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Application of a FO/MD-combined system for the desalination of saline solution

Young-Nam Kwon[a], Min-Jung Kim[b], Yong Taek Lee[b,*]

[a]*School of Urban and Environmental Engineering, Ulsan National Institute of Science and Technology (UNIST), Ulsan 689 798, Republic of Korea, Tel. +82 52 217 2810; Fax: +82 52 217 2809; email: kwonyn@unist.ac.kr*
[b]*Department of Chemical Engineering, Kyung Hee Universiy, Gyeonggi-do 449 701, Republic of Korea, Tel. +82 31 201 2577; Fax: +82 31 202 1946; emails: kmj3203@khu.ac.kr (M.-J. Kim), yongtlee@khu.ac.kr (Y.T. Lee)*

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ABSTRACT

The objective of this study was to systematically investigate the effects of draw solution (DS) chemistry on the performance of a forward osmosis (FO)/membrane distillation (MD)-combined system, and to apply the integrated system to the desalination of a saline solution. Extraction of pure water from saline water was conducted by the FO process, and subsequent production of fresh water and recovery of draw solutes from the DS diluted by the FO process were achieved by MD. Experiments at various temperatures, concentrations, and types of DS showed that the diffusion coefficient of the draw solutes and interaction of the salt ions with water molecules caused severe effects on the performance of the integral system, along with the temperature and concentration of the DS. This study suggests that optimum operating conditions and selection of proper draw solutes with higher diffusion coefficients and lower hydrophilicities can make the FO/MD-combined process a promising candidate for the desalination of saline water.

Keywords: Forward osmosis; Membrane distillation; Draw solution; Desalination

1. Introduction

The worldwide water shortage is emerging as an important issue, and the problem is becoming more serious in many places [1–3]. Furthermore, the demand for fresh water is steadily increasing due to the population growth and rapid industrialization. Nowadays, much research has been actively conducted to solve the freshwater shortage crisis by means of desalination of seawater [4–6]. Desalination is a process to produce fresh water from salty water containing high levels of dissolved inorganic ions, and the current desalination procedures are primarily composed of thermal distillation or membrane separation processes. The thermal technology involves multistage flash distillation (MSF), multiple effect distillation (MED), or the vapor compression method (VC), and the membrane separation uses reverse osmosis (RO), nanofiltration (NF), electrodialysis (ED), forward osmosis (FO) membranes, etc. Compared to the thermal technology, the membrane separation technology requires less capital and operational costs [7]. One of the most widely used technologies in the field of seawater desalination is the RO process, but the driving force for this process is still energy-intensive hydraulic pressure. Furthermore, severe contamination

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*Corresponding author.

and fouling assisted by the hydraulic pressure during the operation requires additional operation and maintenance, such as pretreatment and chemical cleaning [3].

The FO process, however, uses the natural process of osmosis, whereby water molecules diffuse through a membrane from a concentrated solution to a dilute solution. Unlike pressure-driven membrane processes, no high external pressure is required, thus no severe fouling is caused by the hydraulic compaction of the fouling layer on the surface of the FO membrane. The minimal external pressure and lower fouling propensity of the FO process can overcome the intrinsic limitations of the pressure-driven RO process, and thus is expected to provide a cost-effective alternative for the treatment of water, wastewater, and seawater [8]. Maintaining a draw solution (DS) with higher concentration than the feed solution (FS) can generate the osmotic pressure difference across the membrane. Pure water from the feed solution passes through the membrane to keep the osmotic pressure equilibrated, and preferential movement of water from the feed to the DS dilutes the DS. The application of the FO process is mainly limited by the occurrence of internal concentration polarization (ICP), absence of an appropriate draw solute, and lack of an efficient recovery method for the draw solutes [9]. ICP occurs when the pure water transported through the active layer of the membrane from the feed is trapped and builds up inside the support layer, subsequently decreasing the effective concentration gradient across the active layer of the membrane and lowering the osmotic pressure difference [10]. Several researchers have tried to decrease ICP problems by adopting a thin-film composite structure, or by depositing hydrophilic materials on the FO membrane [11–13]. Various molecules have been evaluated as draw solutes for the FO process, including the following: (i) edible hydrocarbons such as glucose, lactose, and sucrose; (ii) inorganic salts such as sodium chloride and magnesium sulfate; and (iii) transformable salts which can change phase when using heat treatment, such as ammonium bicarbonate [8]. Recently, the application of ammonium bicarbonate as a draw solute has received attention since it can dissociate into the ammonium cation and bicarbonate anion in water to produce osmotic pressure, and can later be recovered as ammonia and carbon dioxide gases by a simple heating process [14,15]. However, it seems that heating is not enough to completely remove the ammonium bicarbonate from the solution to the levels suitable for drinking water, since the bicarbonate supplied continuously from the dissolution of atmospheric carbon dioxide gas prevents the evaporation of ammonia to meet the charge balance with the bicarbonate in the solution. For desalination using the FO process, pure water should be extracted from the diluted DS, and the diluted DS should be concentrated again to allow reuse of the DS. Due to the lack of an efficient recovery method, the FO process has been applied without a recovery step for the desalination of saline groundwater for irrigation, and for the offshore membrane enclosures for growing algae [9,16]. The absence of an efficient recovery method has served as a bottleneck for the application of the FO process.

The intrinsic drawbacks of the FO process, such as dilution of the DS during the operation and recovery of the draw solute for reuse, can be overcome by application of the membrane distillation (MD) process. The MD process utilizes the temperature difference between the feed and the cold permeate as a driving force [17,18]. The vapor produced from the feed solution of higher temperatures passes through a non-wettable hydrophobic membrane, and condenses into a cooling solution. MD membranes can have the capability of high permeability and selectivity, even using more concentrated solutions compared to other types of membranes, and the feed water does not have to be boiled past the boiling point. Thus, the MD process has been researched as a low-energy desalination method. In this study, the viability of a combined FO/MD system was investigated as a saline water desalination process, whereby FO was utilized to transport pure water from saline water to the DS, and the MD was applied to extract pure water and concentrate the diluted DS for continuous operation. The effect of concentration, temperature, and types of DS on the permeability of both the FO and MD processes was systematically investigated.

2. Materials and methods

2.1. Materials

The Hydration Technology Innovation (HTI) membrane was investigated as a representative FO membrane. It is a cellulose triacetate membrane. The active surface area exposed to the feed solution and DS was 30.24 cm^2, being 10.8 cm in length and 2.8 cm in width. As a recovery process for the draw solutes in the FO process, an MD membrane manufactured by Econity (Yongin, Korea) was used. The MD membrane used was a hollow fiber-type membrane made of hydrophobic polyvinylidene fluoride (PVDF). The MD module contained 50 fibers with a total surface area of 0.034 cm^2. The characteristics of the FO and MD membranes are shown in Table 1. Sodium chloride, potassium chloride, and magnesium chloride purchased

Table 1
Characteristics of the FO and MD membranes

	FO	MD
Material	Cellulose triacetate	Polyvinylidene fluoride (PVDF)
Type	Flat sheet	Hollow fiber
Membrane size	108 × 28 mm	170 mm length / fiber 1.2 μm / 0.7 μm / 0.5 μm (Outer Dia./Internal Dia./Thick.)
Surface area	0.003024 m^2	0.034 m^2

from Dae Jung (Daejoen, Korea) were dissolved in DI water, and were used as DSs.

2.2. Determination of FO performance

The feed solution for the FO process was prepared by the addition of NaCl to DI water to a concentration of 0.6 M (ionic strength similar to that of normal seawater), and the temperature of the feed was maintained at 25˚C using a water bath circulator. The DSs used in the FO process were 0.6, 2, and 4 M of NaCl at 25, 45, and 60˚C. To examine the effect of the type of DS on the performance of FO, NaCl, KCl, and $MgCl_2$, DS was applied at 4 M and 60˚C. The FO membrane was first presoaked in DI water overnight, and then conditioned in a test cell with a 1 LPM flow rate of FS and DS. After conditioning the FO membranes for 1 h, the performance was evaluated in terms of the permeate flux during operation for the following 3 h. The mass of FS and DS was measured at every hour during 3 h operation using a digital balance, and was used for the calculation of permeate flux. Three measurements of permeate flux were averaged for each representative data point.

The type and characteristics of the DS were closely related to the efficiency of the FO process. Theoretical water flux in FO in the absence of concentration polarization can be estimated using the following equation:

$$J_w = A(\pi_d - \pi_f) \tag{1}$$

where J_w is the water flux, π_d and π_f are the osmotic pressure of the draw and feed solutions, and A is the water permeability coefficient. The membrane water permeability coefficient of the FO membrane was evaluated in a laboratory-scale reverse osmosis test system, revealing a permeability of 2.77×10^{-12} m/s/Pa, which is in the range of those obtained in other reports (1.70×10^{-12} m/s/Pa [19] to 3.75×10^{-12} m/s/Pa [20]).

2.3. Determination of MD performance

The MD module had a diameter of 3 cm and a length of 30 cm. The cylindrical acrylic module was filled with MD hollow fibers using four meshes to keep a constant distance among the membrane fibers, and the vessel was potted using urethane (Haeun, Korea) at one end. The temperature of the module was maintained at 60˚C, and each module was checked for leaking to confirm integrity before each experiment. MD was used for the recovery process of the draw solutes used in FO. Diluted DS from the FO system was used as the feed for MD, and DI water maintained at 10˚C was used as the coolant during the experiments. The MD process was operated in the direct contact membrane distillation (DCMD) mode. Coolants passed along the lumen of the fibers, and pure water evaporated from the outside feed solution (diluted DS) was transported from outside to inside the hollow fiber membrane, condensing on the internal side of the membrane [21,22]. The MD process was independently investigated to evaluate the feasibility of recovery of the draw solute. The feed solution for MD was prepared with the same conditions as the DS of the FO process: 0.6, 2, and 4 M NaCl at temperatures of 25, 45, and 60˚C. The schematic diagram of the FO/MD integrated system is shown in Fig. 1. The presoaking and conditioning process of the MD membranes were the same as those of the FO membranes. After conditioning the MD membranes for 1 h, the performance was evaluated in terms of permeate flux during operation for the following 3 h. The mass of the FS and cooling solution was measured at every hour during 3 h of operation using a digital balance, and was used for the calculation of average permeate flux. Salt rejection was determined by the measurement of the conductivities of the feed and cooling solution every 30 min. Three measurements of permeate flux and six measurements of salt rejection were averaged for each representative data point for performance.

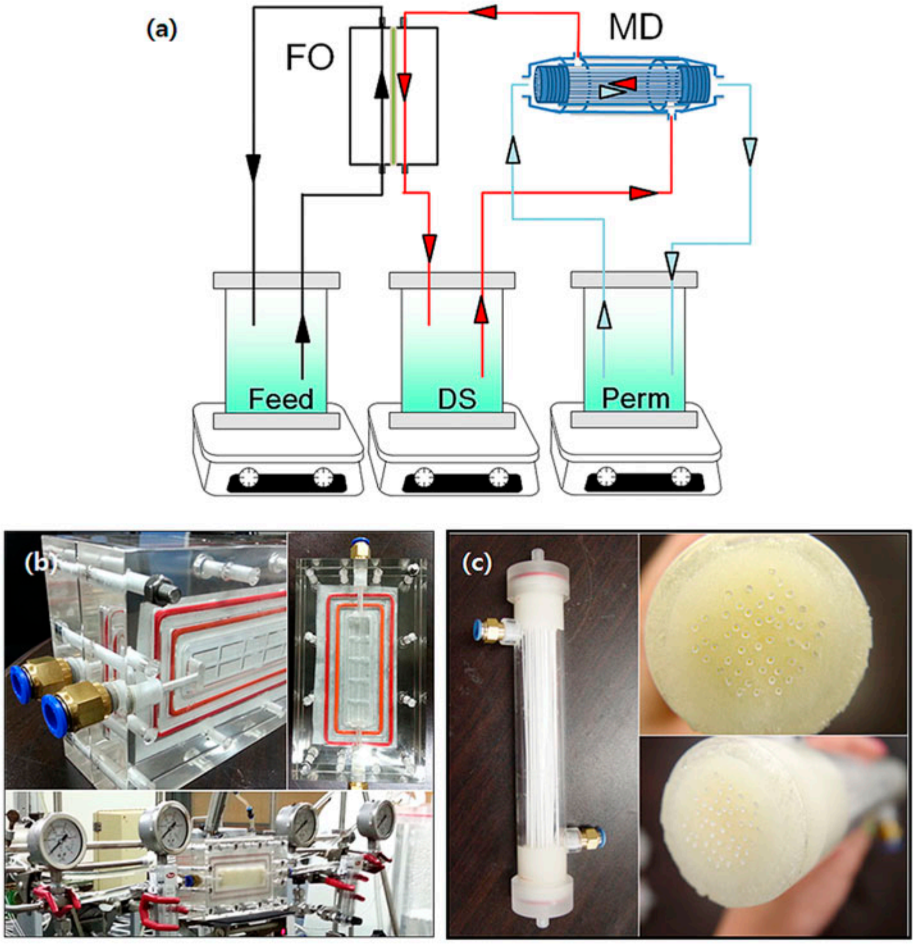

Fig. 1. Schematic flow diagram of FO/MD-combined system (a), and membrane modules of FO process (b), and MD process (c).

3. Results and discussion

An FO/MD-combined system was applied for the desalination of saline feed solution, and viability of the system was evaluated. The FO process was used to preferentially transport pure water from the feed solution to the DS, and the MD process was then applied to extract pure water from the diluted DS and concentrate the diluted DS for reuse. In the combined system, the DS of the FO process is the feed of the MD process. First of all, the optimum operating conditions of the DS in both processes was investigated independently, and the integral system was then run under the optimum conditions.

3.1. Effect of concentrations and temperature of the DS in the FO process

Water flux obtained during FO operation without the subsequent MD process is depicted in Fig. 2, showing the effect of both concentration and temperature of the DS in the process. A 0.6 M NaCl solution at 25˚C was used as the feed solution with an initial volume of 3 L, and 3L of 0.6, 2, and 4 M NaCl solutions were prepared as DSs. The performance of the FO process was investigated at the DS temperatures of 25, 45, and 60˚C.

When the concentration of the FS and DS was equal, 0.6 M at 25˚C, no discernible flux was observed across the FO membrane. The 2 M NaCl DS at 25˚C drove the water in the feed solution to transport across the membrane, causing 1.48×10^{-6} m/s flux. Further increase in the DS concentration up to 4 M produced 2.51×10^{-6} m/s flux. Results showed that the FO flux increased with increasing concentration of the DS. As shown in Eq. (1), the FO water flux depends on the water permeability of the membrane and the osmotic pressure difference across the membrane. Since water permeability is an intrinsic property of membranes, no difference in this value would

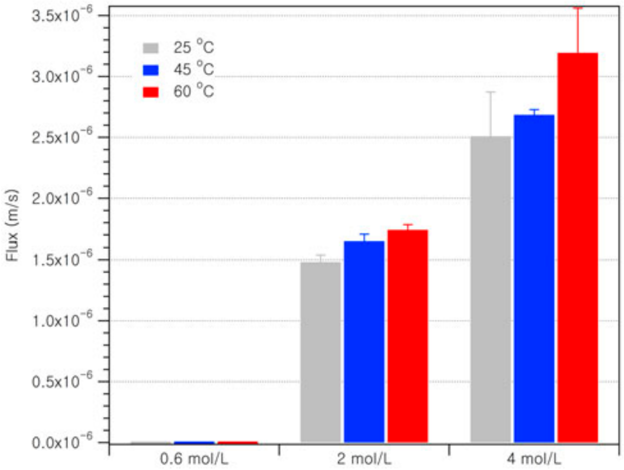

Fig. 2. Water flux in the FO process operated at various concentrations and temperature of DS.

be observed when using the same types of membrane coupon. Thus, the larger osmotic pressure difference caused by the concentration difference increased the water flux.

However, the rate of change in the membrane flux due to concentration ($\Delta J/\Delta c$) at each DS temperature decreased when a higher initial concentration of DS was applied. The rates 1.06, 1,18, and 1.24 μm/s/M at an initial concentration of 0.6 M decreased to 0.51, 0.52, and 0.73 μm/s/M at 2 M initial concentrations under the temperature conditions of 25, 45, and 60°C, respectively. This is due to the more severe internal concentration polarization (ICP) at high DS concentration, which occurs when the diluted DS resulting from the preferential transport of pure water from the feed is trapped inside the support layer of the membrane, decreasing the effective concentration of the DS on the membrane surface. This was further confirmed by comparison of the experimental water flux obtained by filtration test with the theoretical flux derived from equation 1 with an assumption of no ICP (Table 2). The experimental water flux at 2 M DS was 8.5% of its theoretical value, and decreased even more to 5.6% at 4 M DS. Therefore, the larger ICP caused the larger discrepancy between the experimental and theoretical flux at higher concentrations of DS.

When the temperature of the DS increased from 25 to 45°C and to 60°C at a FS temperature of 25°C and a DS concentration of 2 M, the water flux increased about 11 and 18%, respectively. The higher water flux at higher temperatures of DS was also observed in the case of a 4 M DS. Dependence of osmotic pressure on the type of salts, molar concentration of solutes, and solution temperature is well expressed in the van't Hoff equation ($\pi = cRT$, where c is concentration, R is the gas constant, and T is the temperature). The higher temperature difference across the membrane increased the osmotic pressure difference, thus increasing the flux. Operation with 4 M NaCl as the DS at 60°C showed the highest FO flux. These results showed that use of a DS at higher concentrations and temperatures can be beneficiary to allow more extraction of pure water from the feed solution.

Table 2
Experimental water flux and theoretical water flux of the FO process for various concentrations of DS

	Draw solution concentration (M)		
Flux (10^{-6} m/s)	0.6	2	4
Theoretical	–	20.5	57.2
Test	–	1.74	3.19

3.2. *Effect of concentration and temperature of the DS in the MD process*

During the FO process, the driving force of water transport decreases with the operating time due to dilution of the DS. Therefore, recovery of the draw solute from the diluted DS is a critical issue for the FO process. In this experiment, the MD process was evaluated for the recovery and reuse of the draw solutes. The operating parameters of the feed solution for MD were maintained to be the same as those of the DS in FO, since the DS from FO will be used as the feed for MD in the combined FO/MD system. DS diluted after the FO process passes along the outside of the hollow fiber membrane, and cooling water circulating with an initial volume of 3 L at 10°C passes through the inside of the hollow fiber membrane. The water flux was calculated based on the increased amount of cooling water, and the rejection was calculated by measuring the conductivities of both the feed and the permeate.

The effects of temperature and concentration of the DS used in the MD process are shown in Fig. 3. The water flux increased with increasing temperature and decreasing concentration of DS. The increased water flux due to higher temperatures was due to the increased vapor pressure difference across the membrane. The permeate flux in MD depends on the trans-membrane water vapor pressure difference and the membrane characteristic coefficient [6]. The enhanced vapor pressure difference induced with the increasing temperature difference across the membrane could

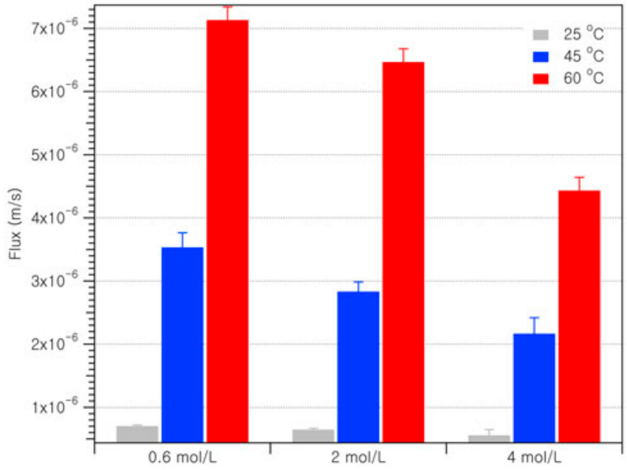

Fig. 3. Water flux in the MD process operated at various temperature and concentrations of DS.

cause faster evaporation of the water molecules at the interface of the feed solution and membrane, and quicker diffusion and condensation of the vapor through the membrane in the membrane/coolant interface.

With increasing concentrations of the DS for MD, the water flux decreased. This resulted from the reduced vapor pressure at high salt concentrations, increasing the surface tension of the solution and decreasing the amount of water vapor produced on the DS side. The rejection of the MD process at various concentrations and temperatures is shown in Table 3. The rejection was maintained above 99.7%, even at high temperature and concentration. This value is equivalent to or higher than the rejection of commercially available RO membranes. This study showed that application of the MD process can be promising when the drawback of low permeate flux in the MD process compared to the RO process is overcome.

3.3. FO/MD-combined system

Finally, the DS used in the FO process was applied as the feed for the MD process to extract pure water and condense the diluted DS. The temperature and concentration of the DS the FO/MD-combined system were fixed at 65˚C and 4 M, respectively, based on the results from the individual FO and MD experiments. In this experiment, the effect of the DS type (NaCl, KCl, and $MgCl_2$) on the FO/MD-combined system was evaluated first, and the draw solute demonstrating the maximum performance was chosen as the representative DS in the integral system. Fig. 4 shows the water flux of the FO process using draw solutes of NaCl, KCl, and $MgCl_2$, and the osmotic pressures and diffusion coefficients of the draw solutes. Osmotic pressure data were obtained from an OLI Stream Analyzer [23], and the diffusion coefficient values were extracted from Refs. [24–26].

Table 3
Salt rejection of the MD process

DS (NaCl) concentration	Conductivity rejection (%) of permeate		
	25˚C	45˚C	60˚C
0.6 M	99.8	99.9	99.9
2 M	99.8	99.8	99.9
4 M	99.7	99.9	99.9

As a draw solute, KCl showed the largest flux followed by NaCl, while $MgCl_2$ showed the lowest flux. Compared to the water flux when using NaCl DS, the water flux of KCl was 66.4% higher. On the other hand, $MgCl_2$ DS showed a 48% lower flux than NaCl DS. This experimental result was unexpected, since the osmotic pressure, a driving force of the FO process, is known to depend on the number of ions and molecules dissociated in the solution. NaCl and KCl are dissociated into water to produce one cation and one anion. However, $MgCl_2$ is dissociated into three parts in water; one divalent cation (Mg^{2+}) and two monovalent anions ($2Cl^-$). According to van't Hoff equation, the theoretical flux is expected to be the highest in the case of $MgCl_2$, while NaCl and KCl should be lower. The osmotic pressure of each of the draw salts obtained from the OLI stream analyzer

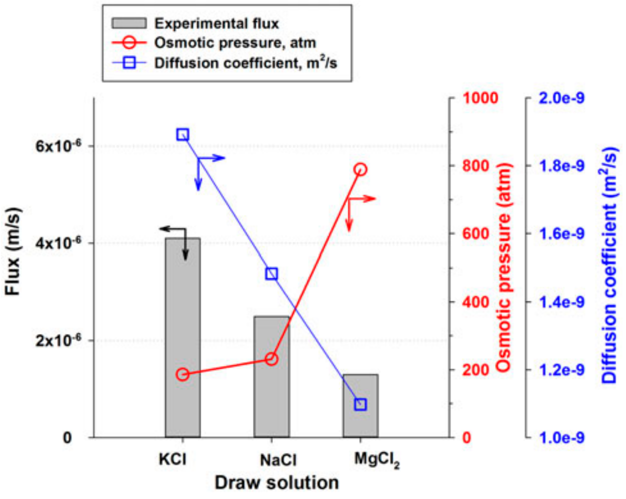

Fig. 4. Water flux of FO membranes in the FO/MD system when using draw solutes of NaCl, KCl, and $MgCl_2$, along with the osmotic pressure [21] and diffusion coefficients of the draw solutes [22–24].

showed the osmotic pressure in the order of $MgCl_2$, NaCl, and KCl at equal molar concentrations [23]. However, experimental results were totally opposite. This discrepancy likely resulted from the different extent of ICP and external concentration polarization (ECP) phenomena, depending on the type of solute. Since magnesium chloride has a lower diffusion coefficient in aqueous electrolyte solutions compared to sodium or potassium chloride, longer residence time of the magnesium chloride in the support layer of the FO membrane would be caused, thus causing a higher decrease in the osmotic pressure difference across the membrane. Furthermore, the higher viscosity of $MaCl_2$ solution as a DS can interfere with the transport of water molecules from the feed to the DS. This behavior was also reported by other researchers [27]. It is likely that the water flux, depending on the type of DS, was primarily determined by the diffusion coefficient of the draw solute.

Maximum flux of the FO/MD-combined system was observed when KCl was used as the DS, as shown in Fig. 5. The water flux of the FO/MD-combined system showed the same pattern as that of the FO-only process. This is likely due to the interaction parameter difference between the water molecules and solutes, along with the diffusion rate of the draw salts. The interaction parameter and the types of draw salts showed the same tendency, as shown in Table 4. With increasing interaction parameters, the interaction between the water molecules and ions becomes stronger, preventing the evaporation of water molecules and finally decreasing the water flux. This study showed that the dilute DS from the FO system is a good feed solution for the MD process, and thus the combined system between FO and MD might be a good combination for the desalination of saline solution and seawater.

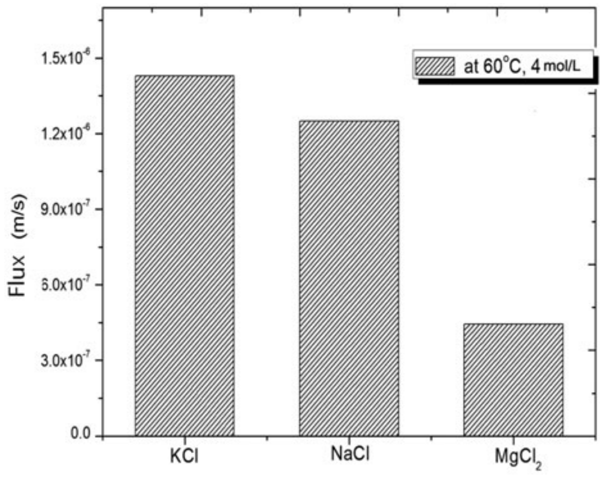

Fig. 5. Water flux of the FO/MD hybrid system.

Table 4
Interaction parameter of DS (KCl, NaCl, $MgCl_2$)

DS (4 M, 60˚C)	δ_j	$(\delta_i - \delta_j)^2$	Interaction parameter
KCl	45	8.41	7.08
NaCl	68	404	9.96
$MgCl_2$	103	3,036	36.11

4. Conclusion

In this study, an FO/MD-integrated system was evaluated for the desalination of saline water, and the effects of concentration, temperature, and type of DS on the water flux of the FO/MD system were investigated. Independent experimental results from the FO and MD processes showed the water flux increased with increasing temperature of the DS. The higher temperature increased the osmotic pressure difference across the FO membrane, and the transmembrane vapor pressure difference across the MD membrane. Both the osmotic pressure and vapor pressure difference are driving forces determining the water flux. With increasing concentrations of the DS, the flux of the FO process increased. However, the flux of the MD process decreased with increasing concentrations of DS. This is due to the increase in osmotic pressure difference in FO, and increase in surface tension and subsequent depression of the water vapor pressure difference through the membrane in MD. The experiments on the effect of DS types in FO showed the water flux to be in the order of KCl > NaCl > $MgCl_2$. This is the opposite order of the osmotic pressure of salts ($MgCl_2$ > NaCl > KCl) obtained from the OLI stream anlyzer (OLI systems Inc., Morris Plains, NJ). The diffusion coefficient difference of the draw salts and viscosity change of the DS when equivalent concentrations of draw solutes are used for the preparation of the DS might have caused this discrepancy. MD was used as a recovery process after FO. The flux of the FO/MD system showed the highest values when using 4 M KCl at 60˚C. Even when the temperatures of the DSs were the same, the water flux among the salts were different. This is due to the difference in interaction parameter value between water molecules and the ions of the salts. Higher interaction forces prevent water molecules from escaping from the DS water body, thus retarding the evaporation and decreasing water flux. As a result, the characteristics of the DS were found to determine the performance of the FO/MD-combined process. In the case of FO, even if the driving force is osmotic pressure, the diffusion coefficient of the DS critically affects the flux. In the case of MD, even if the temperature difference is the driving force, the interaction parameter of water and ions in the feed have a more severe effect on the performance.

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