

## A novel hybrid forward osmosis – nanofiltration (FO-NF) process for seawater desalination: Draw solution selection and system configuration

C.H. Tan, H.Y. Ng\*

*Division of Environmental Science & Engineering, Faculty of Engineering, National University of Singapore, Block EA #03-12, 9 Engineering Drive 1, Singapore 117576*

*Tel. +65-6516 4777; Fax +65 6774 4202; email: esenghy@nus.edu.sg*

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### ABSTRACT

A hybrid forward osmosis–nanofiltration (FO-NF) process for seawater desalination is proposed in this study. Seven potential draw solutions for the FO-NF process were investigated using laboratory-scale forward osmosis (FO) and nanofiltration (NF) test cells. Results from both FO and NF tests suggested that the hybrid FO-NF process is a feasible process for seawater desalination. Water fluxes of about 10 L/m<sup>2</sup> h, for both FO and NF processes could be achieved. Solute rejection of the FO membrane was maintained at over 99.4% for all seven draw solutes tested. Solute rejection of the NF membrane for the four selected draw solutions could achieve a maximum of 97.9%. A hybrid FO-NF process with two-pass NF regeneration is proposed to achieve good quality product water that meets the recommended drinking water total dissolved solids (TDS) guideline, of 500 mg/L from the World Health Organisation. It is possible to desalinate seawater with this system configuration to obtain product water with TDS of 113.6 mg/L. Finally, MgSO<sub>4</sub> and Na<sub>2</sub>SO<sub>4</sub> are proposed as the most likely draw solution for the hybrid process.

**Keywords:** Forward osmosis; Nanofiltration; Seawater desalination

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### 1. Introduction

Recently, several studies on the forward osmosis (FO) process were aimed at utilizing FO in desalination. However, most of those studies have not matured or proven feasible. The FO process utilizes a draw solution that has a higher osmotic pressure than the feed solution and this osmotic pressure gradient across a highly-selective membrane results in water flux across the membrane. Various draw solution at high concentration can have exceedingly large osmotic pressure, potentially leading to a more energy efficient water flux and recoveries as compared to the more expensive reverse osmosis (RO) process. One of the major challenges in the

development of FO process for seawater desalination is the lack of an ideal draw solution. Previously, McCutcheon et al. [1] proposed an ammonia-carbon dioxide FO process for seawater desalination and a pilot plant was set up by Elimelech [2], which reportedly uses lower pumping energy than RO process. However, the availability of waste heat and the stringent quality of the final product water might restrict the progress of using this draw solute. Nonetheless, these studies opened up opportunities for further research on FO process for various applications. As demonstrated by McGinnis et al., an FO system for seawater desalination can theoretically use 85% less energy as compared to RO system [3]. Hence other potential draw solutions together with a suitable recovery system need to be studied to further develop the FO process for seawater desalination.

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\*Corresponding author

To utilize FO for desalination, it is usually necessary to combine the FO process with a post-treatment process in order to separate the draw solute from the product water, and various post-treatments have already been studied [4]. Thermal decomposition and other membrane processes, such as RO, have been proposed as the regeneration means of the draw solution. However, the energy cost associated with using RO for the regeneration of the draw solution is significantly higher, due to its high operating hydraulic pressure, which in effect defeats the purpose of using the FO process for desalination. Instead, nanofiltration (NF), which is a low pressure membrane process, can be used for effective removal of bivalent ions and other larger molecular weight solutes. Both Wang et al. [5] and Mohammad et al. [6] demonstrated that as high as 99% rejection can be achieved for bivalent salt solution [5,6]. Therefore, the NF process can be proposed as an alternative regeneration method for the draw solution.

In this study, a hybrid forward osmosis-nanofiltration (FO-NF) process, which is expected to be lower in operating cost than current desalination technologies, is proposed for seawater desalination. This FO-NF process would be able to achieve regeneration of the draw solution and production of clean product water. The FO-NF process requires a draw solution that can generate a large osmotic pressure as a driving force for the FO process to extract water from the seawater feed. This draw solution must also be able to be removed via the NF post-treatment to produce clean product water and regenerate the draw solution for reuse in the FO process. Seven potential draw solutes namely NaCl, KCl, MgCl<sub>2</sub>, CaCl<sub>2</sub>, MgSO<sub>4</sub>, Na<sub>2</sub>SO<sub>4</sub> and C<sub>6</sub>H<sub>12</sub>O<sub>6</sub> were tested using laboratory-scale FO and NF test cells to assess their suitability as FO draw solutions.

The first objective of this study is to investigate the feasibility of the seven potential solutes, as mentioned above, as the draw solutes in the FO process. A laboratory-scale FO test setup was used to study the performances of these draw solutes in terms of water fluxes and solute rejection. The second objective is to further select the solutes (from the seven solutes tested) for testing using a laboratory-scale NF setup, to evaluate the performance of these solutes in the post-treatment recovery. Finally, the system configuration of the hybrid FO-NF process will be proposed based on the target quality of the product water.

## 2. Materials and methods

### 2.1. Feed and draw solutions

The feed and draw solutions were prepared using deionized water. The feed solution was 0.6 M NaCl

solution which simulated seawater. The draw solutions used for this study include NaCl, KCl, MgCl<sub>2</sub>, CaCl<sub>2</sub>, MgSO<sub>4</sub>, Na<sub>2</sub>SO<sub>4</sub> and C<sub>6</sub>H<sub>12</sub>O<sub>6</sub>. The maximum concentration prepared for these draw solution was as high as 5 M, depending on their solubility.

### 2.2. Membranes tested and orientation

The non-porous FO membrane (Hydration Technologies Inc., Albany, OR) used in the FO process was the same as the one used in Tan and Ng [7]. The FO membrane is made from a cellulose triacetate (CTA) polymer with a polymesh embedded within this polymer for additional mechanical support. It has excellent salt rejection and can achieve 99% NaCl rejection [1]. It is currently the only FO membrane that is commercially available. The FO membrane is usually tested in two different orientations, namely normal (PRO) or reverse (FO) mode. For the normal mode, the dense selective layer faces the draw solution while the porous support substructure faces the feed solution. For the reverse mode, it is opposite in orientation with that of the normal mode.

The NF membrane used in the NF regeneration process was a thin-film composite NF membrane (HL) from GE Osmonics, with a manufacturer nominal MgSO<sub>4</sub> rejection of 98%. The NF membrane is used as per normal, with the tighter layer facing the feed solution.

### 2.3. Experimental setup

The FO experiments were conducted using a dual-channel cross-flow FO membrane cell. A schematic diagram of the laboratory-scale unit used in this study is provided in Tan and Ng [7]. The dimensions of each channel are 2, 250 and 30 mm for channel height, length and width, respectively. The draw and feed solutions would flow co-currently in each channel on either side of the membrane, both of which were controlled independently by a centrifugal pump (Cole-Parmer, Barrington, IL) and the flow-rates were each monitored with a flow-meter (Blue-white Industries Ltd., USA). The volumetric flow-rate for all experiments was held constant at 2.0 L/min (0.444 m/s). Both the feed and draw solutions were maintained at 25 °C by using heaters/cooler with temperature controllers and were kept homogeneous by two independent mechanical stirrers. A weighing scale (SB16001, Mettler Toledo, Germany) connected to a computer with a data logging software was used to monitor the weight changes of the draw solution as a result of the water flux across the membrane to quantitatively calculate the water flux of the

process. In precedence to each experimental run, air was purged from both the flow channels, and the fluid flowing in both channels were allowed to stabilize for 5 min before measurements were taken. The conductivities of the feed and draw solutions were taken before and after each experimental run to determine the actual solute rejection of the solute by the FO membrane.

The NF experiments were conducted using a laboratory-scale cross-flow NF membrane cell. A schematic diagram of the laboratory-scale unit used in this study is provided in Mo et al. [8]. The feed water (diluted draw solution) was circulated into the membrane cell by a diaphragm pump (Hydra-cell D-03, Wanner Engineering, Inc., Minneapolis, MN). The hydraulic pressure was varied up to a maximum of 630 psi (45 atm) and the desired pressures were achieved by adjusting the bypass needle valve and back-pressure regulator. The applied pressure and retentate flow rate were monitored by a digital pressure gauge (PSI-Tronix, Inc. Tulane, CA) and a variable area flow meter (Blue-White industries, Ltd., Huntington Beach, CA), respectively. The permeate flow rate was measured by a digital flow meter (Optiflow 1000, Agilent Technologies, Palo Alto, CA). Both the feed and retentate were recycled back to the feed tank. The feed water was maintained at  $25 \pm 0.5$  °C by a chiller (Model CWA-12PTS, Wexten Precise Industries Co., Taiwan). The HL NF membrane was compacted for 2 h at 600 psi prior to actual experiments. The feed and permeate conductivities were measured using a conductivity meter (1214000, Thermo Scientific Orion, Beverly, MA) and the solute rejection for the various solutions were then calculated.

### 3. Results and discussion

#### 3.1. Performances of FO draw solutions based on FO test

Water fluxes for the seven different draw solutes used in this study were determined from laboratory-scaled FO setup for varying draw solution concentrations. The FO experiments were conducted in the reverse mode (FO mode) and with 0.6 M NaCl solution as the feed water. These results are as shown in Fig. 1. It is observed that as the concentration (or osmotic pressure) of the draw solution increased, the water flux also increased. However, the increase in water flux for most of the draw solutions was not linear to that of the increase in osmotic pressure, and this was attributed to the effect of concentration polarization (CP) occurring on both sides of the FO membrane [7]. According to Fig. 1, both NaCl and KCl draw solutions produced the highest water flux of more than 25 L/m<sup>2</sup> h (LMH),

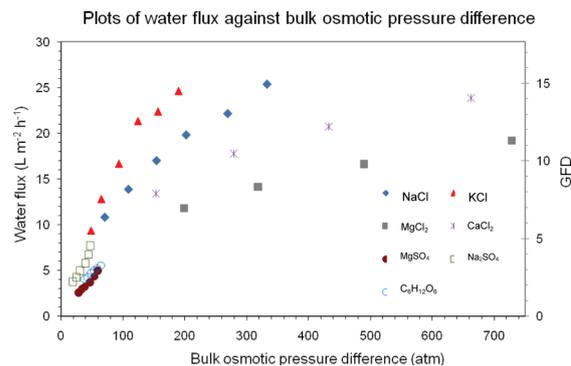


Fig. 1. Graph of water flux against bulk osmotic pressure difference for the seven draw solutes investigated using the laboratory-scale FO test cell. The draw solution concentrations ranged from as high as 5 M–0.6 M. The feed solution was 0.6 M NaCl solution, which simulated seawater. The test is conducted in the reverse (FO) mode and maintained at 25 °C.

while both MgCl<sub>2</sub> and CaCl<sub>2</sub> generated the largest osmotic pressure (as high as 1,400 atm, not shown in Fig. 1) because of their high solubility in water. Even though MgCl<sub>2</sub> and CaCl<sub>2</sub> could generate significantly larger osmotic pressures than all other draw solutes tested, the corresponding water fluxes were only comparable to that of NaCl and KCl. This was probably due to the more severe CP effect for both the MgCl<sub>2</sub> and CaCl<sub>2</sub> draw solutions. The remaining three draw solutes (Na<sub>2</sub>SO<sub>4</sub>, MgSO<sub>4</sub> and C<sub>6</sub>H<sub>12</sub>O<sub>6</sub>) produced much lower water fluxes than the above mentioned four draw solutes. This suggests that the CP effects were much more severe for these three draw solutes. According to Tan and Ng [7], CP effects could reduce water fluxes by as much as 50% of theoretical water fluxes [7].

As the FO membrane is non-porous, it is expected to achieve a very high solute rejection for all the solutes tested. Table 1 shows the average solute rejection for

Table 1  
Average solute rejection of the FO membrane tested for the seven draw solutes investigated

Draw solute	Solute rejection of FO membrane (%)
NaCl	99.47
KCl	99.48
CaCl <sub>2</sub>	99.98
MgCl <sub>2</sub>	99.97
MgSO <sub>4</sub>	99.99
Na <sub>2</sub> SO <sub>4</sub>	99.97
C <sub>6</sub> H <sub>12</sub> O <sub>6</sub>	99.86

each of the seven draw solutes that were obtained from the FO test over the testing duration of 2 h. Results indicated that the solute rejection of this FO membrane was always higher than 99.47% (NaCl) and could achieve a maximum of 99.99% rejection ( $\text{MgSO}_4$ ).

In order to evaluate the feasibility of the hybrid FO-NF process, the seven draw solutes will have to be further selected and their performance in NF process, based on water flux and rejection efficiency, evaluated so as to produce good quality product water. According to Mohammad et al. [6], their experimental NF results showed that for NaCl and KCl at high concentrations, the maximum NaCl rejection achieved is less than 40% and that of KCl is only 65% [6]. Therefore both NaCl and KCl were not selected for further NF test in this study.  $\text{CaCl}_2$  was also not selected for further NF test because  $\text{CaCl}_2$  solution at high concentration when exposed to atmosphere produces precipitates, which will be formed when  $\text{CO}_2$  gas dissolves into the solution to form  $\text{CaCO}_3$  precipitates. Finally, the performances of the remaining four draw solutes,  $\text{MgCl}_2$ ,  $\text{MgSO}_4$ ,  $\text{Na}_2\text{SO}_4$  and  $\text{C}_6\text{H}_{12}\text{O}_6$ , were tested using the laboratory-scale NF test setup.

### 3.2. Performances of FO draw solutes based on NF test

The second part of this study involved the testing of  $\text{MgCl}_2$ ,  $\text{MgSO}_4$ ,  $\text{Na}_2\text{SO}_4$  and  $\text{C}_6\text{H}_{12}\text{O}_6$ , using a laboratory-scale NF test cell to determine the regeneration of the four FO draw solutions and also to evaluate the quality of the product water for the FO-NF process. Previously, no other NF studies have investigated the solute rejection of NF process at very high solute concentration. In Mohammad et al. [6], the maximum solution concentration that was investigated was only 20,000 ppm ( $\sim 0.1$  M). Therefore it is pertinent for this study to study the NF process for solution concentration higher than 0.1 M. Fig. 2 shows the water fluxes at varying solution concentrations for  $\text{MgCl}_2$ ,  $\text{MgSO}_4$ ,  $\text{Na}_2\text{SO}_4$  and  $\text{C}_6\text{H}_{12}\text{O}_6$ , with a hydraulic driving force of 35 atm (490 psi). For all the plots of different solutions, they follow similar trend, whereby the water fluxes decreased when the solution concentration was decreased. This is due to the increase in opposing osmotic pressure, which would reduce the effective hydraulic driving force for the production of permeate across the NF membrane. As the solution concentration was increased, the opposing osmotic pressure would increase and thereby, reducing the effective hydraulic pressure.

Results in Fig. 2 suggest that the draw solution concentration must not be too high as it reduces the water flux to a value this is too low for efficient permeate production for NF. When 0.6 M  $\text{Na}_2\text{SO}_4$  solution was used as the NF feed water, a water flux of only 5.38 LMH

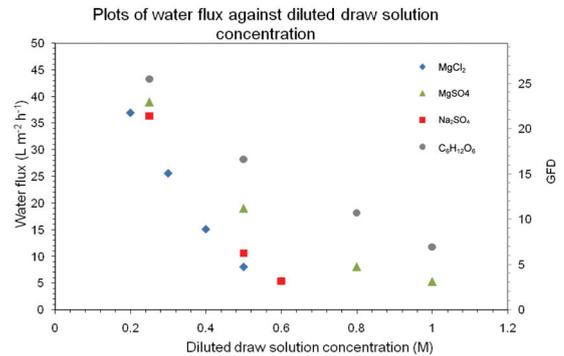


Fig. 2. Graph of water flux against solution concentration for the selected four draw solutes investigated using the laboratory-scale NF test cell. The NF feed solution concentrations ranged from as high as 1 M–0.2 M. The hydraulic pressure was maintained at 490 psi (35 atm). Temperature of the system was maintained at 25 °C.

was produced. However, when the concentration was reduced to 0.5 M, the water flux almost doubled to 10.6 LMH. Although  $\text{C}_6\text{H}_{12}\text{O}_6$  draw solution had slightly higher fluxes than the other three solutes,  $\text{C}_6\text{H}_{12}\text{O}_6$  was not recommended for use as the draw solution for the hybrid process because it is susceptible to degradation by microorganisms, thereby rendering the reusability of the  $\text{C}_6\text{H}_{12}\text{O}_6$  draw solution difficult. Hence, we can also conclude that the selection of the draw solute is also based on its stability and must not degrade (physical, chemical or biological) upon reuse. Therefore organic draw solutes should be avoided for recycled usage.

Solute rejection of the NF process is also another important parameter for determining the draw solution for effective use in the FO-NF system. Fig. 3 shows the results for solute rejection against NF feed solution

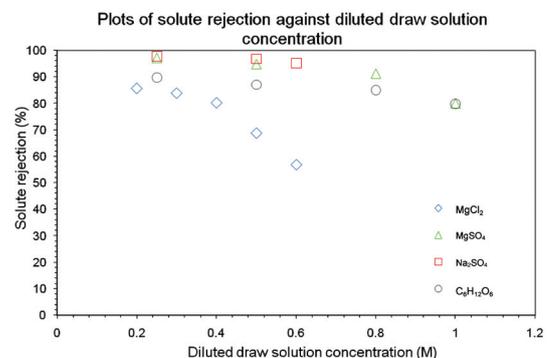


Fig. 3. Graph of solute rejection against diluted draw solution concentration for the selected four draw solutes investigated using the laboratory-scale NF test cell. Process parameters are the same as Fig. 2.

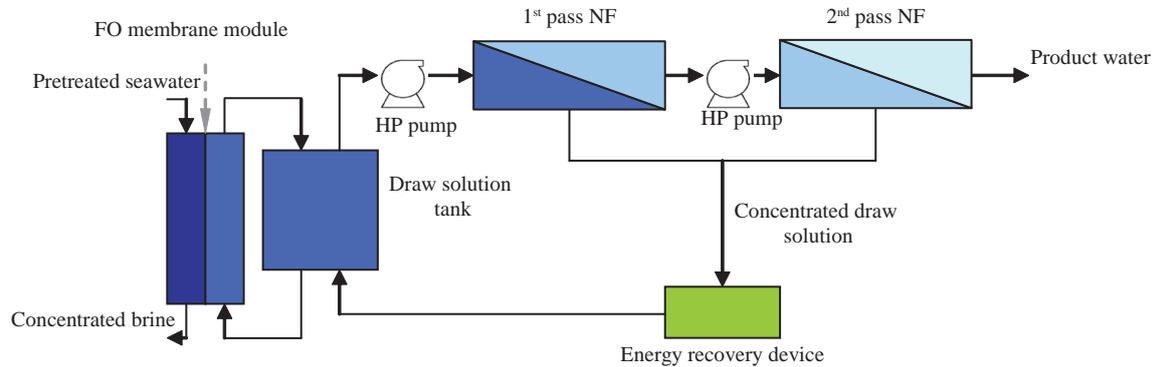


Fig. 4. Proposed configuration of hybrid FO-NF system for seawater desalination.

concentration for the four draw solutes tested. It can be observed that  $\text{Na}_2\text{SO}_4$  rejection by NF membrane could be maintained above 90% for increasing solution concentration. The solute rejection for the other three solutes ( $\text{MgCl}_2$ ,  $\text{MgSO}_4$  and  $\text{C}_6\text{H}_{12}\text{O}_6$ ) decreased to below 90% at higher solute concentration, with  $\text{MgCl}_2$  rejection decreasing to less than 57% when the NF feed solution was at 0.6 M  $\text{MgCl}_2$ . Therefore, a single-pass NF process is not capable of producing good quality product water and that another polishing step, such as a second-pass NF, is necessary to further improve the quality of the final product water.

### 3.3. System configuration of the hybrid FO-NF process to produce drinking water

Results obtained from the FO and NF tests indicate that the most likely draw solutions to be used for the hybrid FO-NF process are  $\text{MgSO}_4$  and  $\text{Na}_2\text{SO}_4$ . Since a single-pass NF post-treatment to recycle the draw solution is not sufficient to produce a sufficient product water quality, the FO-NF process needs to be modified to encompass a two-pass NF regeneration process as shown in Fig. 4. This ensures that the product water is within the recommended drinking water guideline provided by the World Health Organisation (Guidelines for drinking-water quality (GDWQ), WHO 2006). Table 2 shows the permeate water quality for the

hybrid FO-NF process with two-pass NF regeneration. Both permeate qualities when using  $\text{MgSO}_4$  and  $\text{Na}_2\text{SO}_4$  as draw solutions were within the recommended total dissolved solids (TDS) guideline, with the hybrid process using  $\text{Na}_2\text{SO}_4$  draw solution achieving a low TDS value of 113.6 mg/L. This shows that the hybrid FO-NF process with two-pass NF regeneration could potentially desalinate seawater into good quality drinking water. Further optimization of the proposed FO-NF process is required to enhance the water flux and further improve the permeate water quality.

## 4. Conclusions

In conclusion, the results obtained from laboratory-scale FO and NF tests suggest that both  $\text{MgSO}_4$  and  $\text{Na}_2\text{SO}_4$  could be used as potential draw solutes for the hybrid FO-NF process. Depending on the concentration of the draw solutes, acceptable water fluxes for both FO and NF processes could be achieved. Solute rejection of over 99.4% for the FO membrane could be maintained for all seven draw solutes tested. Likewise, a maximum of 97.9% solute rejection of the NF membrane for the four selected draw solutions could be achieved. In order to achieve good quality product water that meets the recommended TDS of the GDWQ from WHO, a hybrid FO-NF process with single-stage two-pass NF regeneration is proposed. It is possible

Table 2

Expected permeate water quality for a hybrid FO-NF process with two-pass NF regeneration

Solute	Diluted draw solution concentration (M)	Expected permeate quality		
		After first pass NF (mg/L)	After second pass NF (mg/L)	WHO TDS Guidelines (mg/L)
$\text{MgCl}_2$	0.6	24,510	3921.6	500
$\text{MgSO}_4$	1	24,000	480	
$\text{Na}_2\text{SO}_4$	1	11,360	113.6	

to desalinate seawater with this process to obtain product water with TDS of 113.6 mg/L. Further work needs to be done to optimize the draw solution concentration for use in this proposed process. The effective water flux of the process should be further enhanced to be comparable with that of current RO operating flux, if not higher. Lastly, energy requirement of the system has to be calculated to evaluate the system's feasibility when compared to RO process for seawater desalination.

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