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Preliminary studies of seawater desalination using forward osmosis

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ABSTRACT

Forward osmosis is an emerging technology that has gained increasing attention in the field of seawater desalination. A recent innovation using ammonia-carbon dioxide solution as a draw fluid to remove water from saline feed through membranes has been widely discussed. This process has immense potential for seawater desalination due to its promising low energy consumption. This project attempts to study the forward osmosis performance with respect to different ammonia-carbon dioxide draw solutions and membranes. A simple forward osmosis test rig using a flat-sheet membrane was built. The results suggest that internal concentration polarization in the membrane lowered the anticipated flux. This phenomenon was found to be more prominent in higher concentration feeds. Increasing the temperature improved flux but the effects were less significant for higher concentration feeds. The cellulose triacetate membrane tested would be more compatible with sucrose as draw fluid in terms of flux compared with that of the ammonia carbon dioxide draw solution which is of a high molarity. If the ammonia carbon dioxide should be used as draw solution, the ammonium carbonate would be preferred.

Keywords: Forward osmosis; Cellulose triacetate membrane; Internal concentration polarization; Ammonia-carbon dioxide draw fluid; Osmotic de-swelling

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1. Introduction

Although large-scale thermal desalination plants are still being constructed in the Arabian Gulf states, the great majority of the world's new and planned desalination capacity is established using synthetic membranes [1], mainly in seawater reverse osmosis (SWRO). In fact, energy and membrane replacement are the major cost components of SWRO desalination. The escalation in the energy price, in general, drove researchers to explore more efficient means of low-energy and low-cost desalination processes. Proposed in 1976 [2], forward osmosis (FO) is conceptually a low-energy desalination alternative. FO works by means of osmotic pressure from a highly concentrated draw solution to establish an osmotic pressure gradient across a semi-permeable membrane, thus inducing water flux flow from the feed solution to the draw solution. With a suitable choice of solute as a draw fluid, the osmotic pressure gradient generated across the forward osmosis membrane can be comparable to that of the trans-membrane pressures (TMP) applied to the SWRO process.

A potential draw solute for FO is ammonia-carbon dioxide salt. It is identified due to its high osmotic efficiency and ease of separation from potable water [3]. Water can be easily separated from the diluted draw

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solution by heating near 60°C to yield fresh water, ammonia and carbon dioxide. Both the ammonia and carbon dioxide can then be reused as solutes for the draw fluid.

2. Experimental

2.1. Configuration

The FO experiments were conducted on a laboratory-scale unit, as shown in Fig. 1. The semi-permeable membrane was positioned vertically between the two compartments, one containing the draw solution and the other containing the feed solution. The membrane was orientated such that its active layer faced the draw solution compartment to reduce internal concentration polarization (CP) and thus obtain a higher flux flow [4].

Initially, the fluids in both compartments were filled till excess solution entered in their respective measuring columns A and B of different diameters. During the FO experiment, the permeate passes through the membrane and the volume change is measured from the change in the measuring column solution levels. The volumetric flow rate of permeate is then calculated from the permeate volume and the time interval taken. The isometric drawing of the test rig with dimensions and materials used is shown in Fig. 2.

Also, the FO experiment was conducted with temperature of the feed and draw solutions as a variable parameter by means of heating coil as shown in Fig. 1. A temperature controller was used to maintain the draw and feed solution temperature within ±1°C error.

2.2. Draw and feed solutions

The ammonium carbonate salt used was laboratory grade purchased from Sigma-Aldrich. The ammonium bicarbonate salt and the sea salt (NaCl) were purchased as food-grade chemicals from Poon Huat. All the chemicals were used as received. Solutions of different molarity were mixed by combining the solutes with de-ionized water at pre-determined proportions.

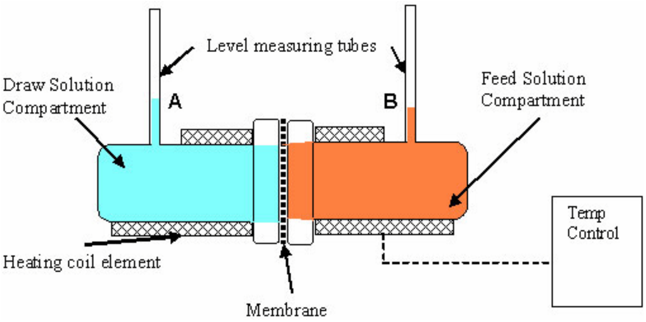


Fig. 1. Schematic diagram of the laboratory-scale forward osmosis set-up.

2.3. Membranes

Three different types of membranes were tested. One of them was a commercially available FO membrane from the Seapack™, supplied by Hydration Technologies. This was the same membrane used by in the other research study [3,4]. To the best knowledge of the author, no other membrane is commercially available for the FO seawater desalination application. The active layer is made up of cellulose triacetate while the support is made of non-woven polyester fibers individually coated with polyethylene. The SEM photograph of the membrane cross-section, as shown in Fig. 3, clearly shows the active layer and fiber support of the FO membrane. A porous section support revealed in the centre portion of the membrane cross-section. It increases the thickness of the FO membrane and hinders the permeate flux due to internal concentration polarization.

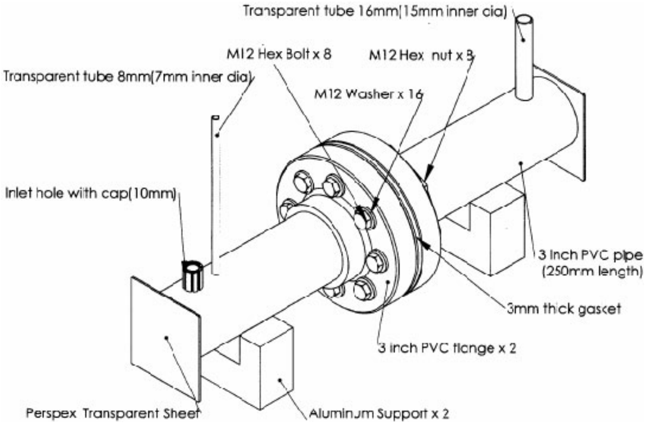


Fig. 2. Engineering drawing of the laboratory-scale forward osmosis set-up.

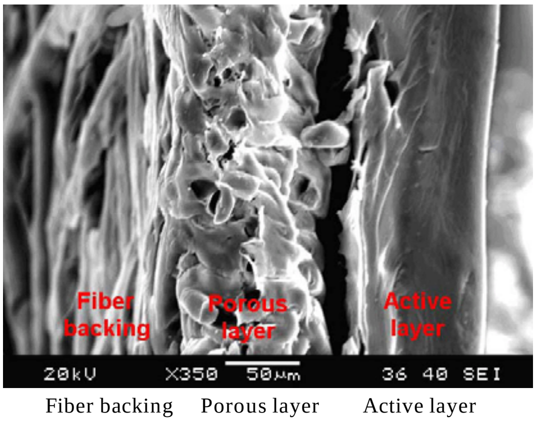
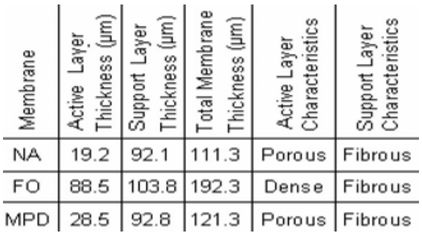


Fig. 3. SEM picture of the FO membrane cross section.



Membrane	Active Layer Thickness (μm)	Support Layer Thickness (μm)	Total Membrane Thickness (μm)	Active Layer Characteristics	Support Layer Characteristics
NA	19.2	92.1	111.3	Porous	Fibrous
FO	88.5	103.8	192.3	Dense	Fibrous
MPD	28.5	92.8	121.3	Porous	Fibrous

Fig. 4. Characteristics of the membranes measured from SEM photographs.

The other two membranes tested are nanofiltration membranes designed for pressure-driven processes. One of them is commercially manufactured while the other was self-cast using M-phenylenediamine (MPD) as the aromatic diamine and trimesoyl chloride (TMC) dissolved in n-hexane as the amine-reactive reactant casted on a ready-made polyacrylonitrile (PAN) membrane. This paper will denote the forward osmosis membrane as FO, the self cast nanofiltration membrane as MPD, and the other nanofiltration membrane as NA. Some useful descriptions of the membranes are seen in Fig. 4. The nanofiltration membranes were used for two reasons. First, they were immediately available in the laboratory. Second, it was intended to allow complete salt passage as a baseline for flux under the selected draw and feed solutions.

3. Results and discussion

3.1. Forward osmosis flux profile

For a fixed concentration (5 M) of the ammonium bicarbonate draw fluid, the accumulated permeate volumes collected vs time for different feed concentrations are presented in Fig. 5. It was observed that rate of water permeate decreased with respect to time. This can be attributed to a phenomenon known as internal concentration polarization (CP) that progressively reduces the flux rate, and has also been experienced in other studies related to FO [5,6].

A reduction of osmotic pressure difference is expected when increasing the feed molarity. This explains the observed lower permeate volume for case of higher feed molarity in Fig. 5.

3.2. Effects of draw and feed solution molarity

This part of the results demonstrates the effect of draw solution (ammonium bicarbonate) and feed (sodium chloride) solution molarity on the membrane water flux.

The water flux is governed by the following equation:

$$J_w = A\,(\sigma\Delta\pi - \Delta P) \qquad (1)$$

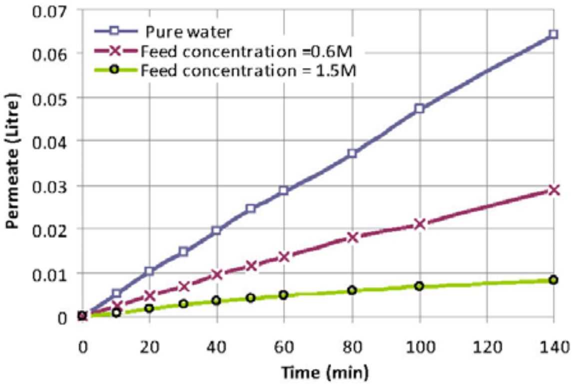


Fig. 5. Accumulated permeate volume for feeds with different molar concentration while maintaining the ammonium bicarbonate draw fluid at constant molar concentration of 5 M.

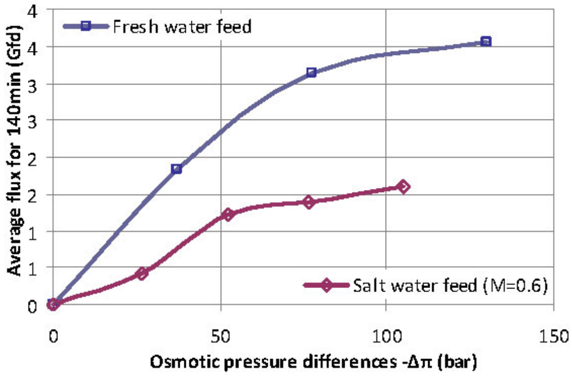


Fig. 6. Water flux over a range of osmotic pressure differences at 50 °C.

where J_w is the water flux, A is the permeability constant of the membrane, σ is the reflection coefficient, $\Delta\pi$ is the osmotic pressure difference, and ΔP is the applied pressure ($\Delta P = 0$ for FO).

The osmotic pressure difference ($\Delta\pi$) depends on the molarity of both the feed and draw solutions. Based on Eq. (1), with a given reflection coefficient (σ) and a water permeability coefficient of the membrane (A), an increase in the feed molarity will cause a reduction in water flux (J_w) as observed in Fig. 6, due to the reduction in $\Delta\pi$.

As described in Section 2.1, the membrane in the test rig was orientated to have the active layer facing the draw solution. In other words, the porous support side of the membrane faced the feed solution. Under this membrane orientation, a higher level of concentrative internal CP would be expected for having feed of higher molarity. This explains why the flux for the concentrated feed was much lower than that of the fresh water feed. For a given osmotic pressure difference of 100 bars, the water flux was lowered by more than 50% for the feed solution of higher molarity. All these suggest that feed solution molarity is an important parameter to the water flux. This also implies that higher water recovery from seawater using

FO would result in a significant reduction in the membrane flux.

In using fresh water (molarity = 0) as feed, the concentrative internal CP effect can be completely eliminated. The water flux of the forward osmosis process with fresh water feed would be the best.

In order to maintain an osmotic pressure difference, the increase in the feed molarity requires the draw fluid molarity to be increased accordingly. A resultant higher draw fluid molarity would lower the reflection coefficient σ because some of the draw fluid solute might cross the membrane towards the feed solution side (known as reflection). The reflection effect due to the increase in the draw fluid molarity reduces the water permeability of the membrane.

3.3. *Effects of temperature*

By varying the temperatures of the FO process, the effect of CP on different feed molarity can be investigated. The rise in fluid temperature reduced the fluid viscosity, increased the diffusion rate of water through the membrane and improved water permeation, as demonstrated in Figs. 7 and 8 for feed molarity below 0.5 M (high osmotic pressure difference). The flux was increased by 51% when process temperature raised from 27 to 50°C for fresh water feed.

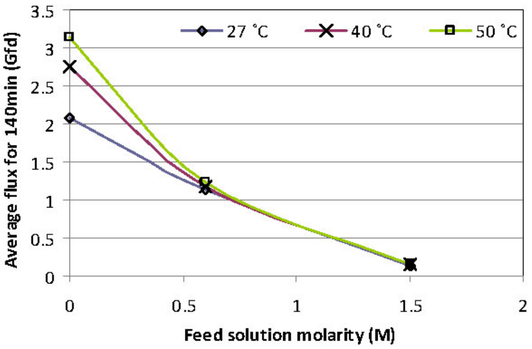
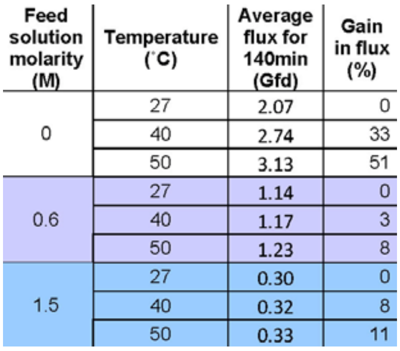


Fig. 7. Water flux over various molarities of sodium chloride feed and temperatures using fixed ammonium bicarbonate draw solution at 2.5 M and FO membrane.



Feed solution molarity (M)	Temperature (˚C)	Average flux for 140min (Gfd)	Gain in flux (%)
0	27	2.07	0
	40	2.74	33
	50	3.13	51
0.6	27	1.14	0
	40	1.17	3
	50	1.23	8
1.5	27	0.30	0
	40	0.32	8
	50	0.33	11

Fig. 8. Water flux for feed of different molarity and temperature using a fixed ammonium bicarbonate draw solution of 2.5 M and FO membrane.

However, the change in water flux due to temperature was almost negligible for cases of high feed molarity (low osmotic pressure difference). The poor flux improvement due to temperature rise for higher feed molarity could be due to three effects that reduces water flux. Firstly, higher reflection rate of ammonia gas could be attributed to the rise in temperature. Secondly, the increase in the feed molarity caused the osmotic de-swelling effect. Lastly, higher feed molarity escalated the internal CP.

3.4. *Performance of different membranes*

Fig. 9 shows the water flux of different membranes used in the experiment. Obviously, the FO membrane has the highest water flux. Since the experimental conditions were identical, differences in water flux demonstrated the differences in the water permeation coefficient (A) value of these three membranes. The water permeation coefficient value is affected by the membrane thickness, partition coefficient of water into the membrane and the diffusivity of water within the polymer membrane based on the studies by Wijmans [7]. The active layer of the FO membrane was the thickest yet it had the highest water flux rate. This means that the difference in the material making up the active surface layer is the main cause for the different water flux achieved.

Another notable point was the change of water flux due to the increase in feed molarity from 0M to 0.6M. The FO membrane had the lowest decline in flux (about 40%) while the NA membrane had the highest decline in flux (>80%). This shows that the FO membrane has a better capability to resist internal CP effect because of its non-woven membrane backing (made of polyester fibers individually coated with polyethylene though thicker). The results in Fig. 10 indicate that the FO membrane has the best salt rejection.

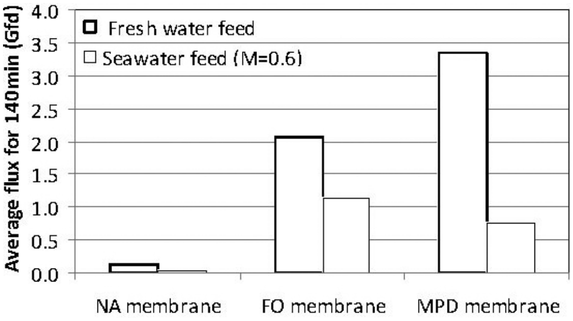


Fig. 9. Water flux comparison using different membranes. Experimental conditions: ammonium bicarbonate draw solution at 2.5 M and 0 M/0.6 M sodium chloride feed at 27°C.

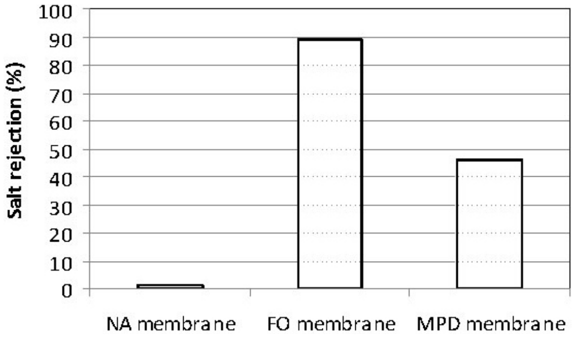


Fig. 10. Salt rejection of membranes using sodium chloride (400 ppm) feed at 27°C.

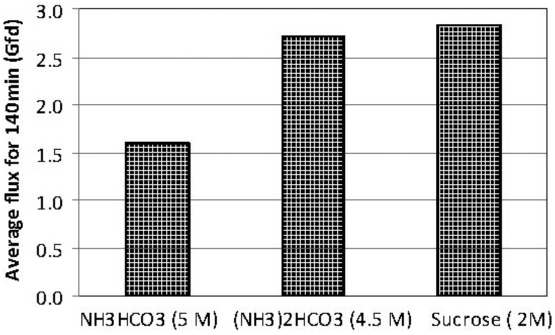


Fig. 11. Water flux obtained from different draw solutions at their near maximum molarity at 50°C. Experimental conditions: sodium chloride feed solution at 0.6M using the FO membrane.

3.5. Effect of draw solution on FO membrane

The theoretical osmotic pressure π (in atm) of a particular solution can be given by:

$$\pi = j\,MRT \qquad (2)$$

where *j* is the Van't Hoff factor, *M* is molarity (M, or moles per liter), *R* is the gas constant (0.0836 L atm/mol K), and *T* is temperature in Kelvin.

Based on Eq. (2), the theoretical osmotic pressure for ammonium carbonate solution should be the highest followed by ammonium bicarbonate and sucrose. Moreover, studies by Gordon et al.[3] showed that solutes with heavier molecular weight tend to produce less flux in the presence of internal CP compared to solutes with lighter molecular weight at the same osmotic pressure difference. This means that sucrose with the highest molecular weight and the lowest osmotic pressure difference should give the lowest flux. However, the results in Fig. 11 offer a different result: sucrose as draw fluid offered the highest flux (instead of the lowest predicted). The flux using sucrose as draw fluid was slightly higher than that of the ammonium carbonate as draw fluid. However, the fluxes of both the sucrose and ammonium carbonate were about 40% higher than that of the ammonium bicarbonate as draw fluid. If the ammonia carbon dioxide should be used a draw fluid, the ammonium carbonate was preferred due to its high flux performance.

These findings suggest the compatibility of the cellulose triacetate membrane with the draw fluid–CA membrane was more compatible to the sucrose as draw fluid compared with the ammonia carbon dioxide draw fluid. The reasons are:

1. Ammonia-carbon dioxide solutions are known to release ammonia gases into the feed side, causing reflection and thus impeding permeate flow.
2. There was a membrane phenomenon known as osmotic de-swelling [3] that caused the reduction of flux due to high fluid concentration. In the experiment, the high molar concentration of the ammonium carbonate and bicarbonate solutions might lower the membrane permeability through osmotic de-swelling.

4. Conclusions

The internal CP reduced the flux of FO progressively. For the same osmotic pressure difference, the effect of concentrative internal CP was greater for higher feed concentration. It reduced flux by 50% when the sodium chloride feed molarity increased by 0.6 M.

The permeate flux was improved when system temperature was raised but the effect was insignificant for concentrated feed. The flux was increased by 50% when the feed temperature was raised from 27 to 50ºC for fresh water as feed.

When comparing the performances of the different membranes, the FO membrane was found to operate more effectively due to the polymer material of the thin active film though it was the thickest membrane.

The FO membrane also showed a better quality in terms of concentrative internal CP. When the feed molarity was raised by 0.6M, the FO membrane only suffered a 40% drop in flux compared with the more than 80% drop in flux for the other two membranes.

Comparing ammonia carbon dioxide and sucrose as draw solutions, the flux of sucrose turned out to be the highest though it had the lowest osmotic pressure difference and heaviest molecular weight. This suggests that the cellulose triacetate membrane was more compatible with sucrose as the draw fluid than the ammonia carbon dioxide solution. If the ammonia carbon dioxide should be used as draw fluid, ammonium carbonate is preferred. Its flux was 40% higher than that of the ammonium.

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