

# Challenges in Forward Osmosis of Seawater Using Ammonium Bicarbonate as Osmotic Agent

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**Abstract** This study aimed at exploring whether product quality, membrane fouling and salt reverse flow would be challenges in forward osmosis (FO) of seawater using  $\text{NH}_4\text{HCO}_3$  as an osmotic agent. Experiments were conducted with a lab scale FO system containing effective membrane area of  $95 \text{ cm}^2$ . Synthetic seawater (SSW) with 3.5-7.0 mg/L boron and a real seawater (RSW) were used as feeds and 1.5-2.5 M  $\text{NH}_4\text{HCO}_3$  as draw solutions. The experimental operation could be stabilized within 0.5 h. For the SSW, boron rejection ranged of 47-85% and increased with increasing water flux while boron in the permeate was greater than 0.8mg/L. Water flux with RSW was 3 times lower than that with SSW, indicating that there might be serious membrane fouling with RSW. It was surprisingly observed that non volatile organic in the FO permeate was 8-10 mg/L, which was from the draw solution although  $\text{NH}_4\text{HCO}_3$  used was analytical grade. Additional water cost would be  $\$0.4/\text{m}^3$  because of  $\text{NH}_4\text{HCO}_3$  loss. It was concluded that product quality in terms of high TOC contaminant in  $\text{NH}_4\text{HCO}_3$  and low boron removal, serious fouling with RSW and salt reverse flow could be challenges for the FO process using  $\text{NH}_4\text{HCO}_3$  as osmotic agent for seawater desalination.

**Keywords:** seawater desalination, forward osmosis, ammonium bicarbonate, boron removal, membrane fouling, salt loss

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## 1. Introduction

Forward osmosis (FO) is known as the net movement of water across a semi-permeable membrane driven by a difference in chemical potential across the membrane, i.e., passage of water is from lower concentration side to higher one because the lower the concentration, the higher the water chemical potential. Although the FO phenomenon was observed in 1748, the interests in research on FO and in engineered applications of FO have been increasingly attractive since 2000 [1,2]. However, there are still three major challenges of FO in municipal scale seawater desalination: a) fabrication of FO membranes with high water flux and high draw solute rejection; b) being lack of appropriate draw solutions; and c) cost-effective post-treatment on a total system approach [3].

In desalination application, the energy consumption for seawater desalination using reverse osmosis (RO) is currently 3-3.5 kWh/m<sup>3</sup>. Newly developed FO technology could potentially lower desalination energy consumption since osmotic pressure, not hydraulic pressure as in the case of RO, provides a natural driving force for water transport through the FO membrane. It is reported that the Manipulated Osmosis Technology developed by

University of Surrey could reduce energy consumption to  $<1.6 \text{ kWh/m}^3$  using proprietary osmotic agents as a draw solution. The proprietary process had also been demonstrated using a 1 m<sup>3</sup>/h pilot unit and is now ready for commercialization [4]. Khaydarov [5] developed a solar powered direct osmosis desalination process with a pilot testing of 1 m<sup>3</sup>/h capacity. He reported the specific power consumption of the desalination process could be less than 1 kWh/m<sup>3</sup> excluding the requirement on solar energy.

In recent years, there has been increasing interest in an integrated FO process for seawater desalination developed and intensively studied by Elimelech and his research group at Yale University [6,7,8,9]. In the process, ammonium bicarbonate ( $\text{NH}_4\text{HCO}_3$ ) is utilized as the osmotic agent and a thermal process such as the distillation is proposed as the post-treatment to strip and recover the osmotic agent. Potential advantages of  $\text{NH}_4\text{HCO}_3$  osmotic agent include high osmotic pressure which can result in high water flux and utilisation of waste heat (if available) to recover the osmotic agent at  $<60 \text{ }^\circ\text{C}$ . Other researchers [10-16] have also studied the FO process with  $\text{NH}_4\text{HCO}_3$  as the osmotic agent. However, existing studies on the  $\text{NH}_4\text{HCO}_3$  concept focus mainly on effects such as osmotic agent concentration, temperature, and role of internal concentration polarization on water flux. Study of this process using real seawater as a feed

has not been reported so far and there is also a lack of data on product quality. Practical issues such as loss of osmotic agent in the FO process for implementation in the municipal scale have also not been adequately discussed. Objective of this study is to address the above issues and discuss on challenges in using  $\text{NH}_4\text{HCO}_3$  as osmotic agent in a FO process for seawater desalination.

## 2. Experimental Materials and Methods

The lab scale FO unit used in experiments is shown in Figure 1 [17]. Two FO membranes from Hydration Technologies, Inc. were used in the study. The FO membrane with a diameter of 110 mm (effective membrane area of  $95 \text{ cm}^2$ ) was positioned vertically between the two compartments in a membrane holder without any additional support. The active layer of FO membrane was orientated to face the draw solution in order to reduce internal concentration polarization (CP) and thus obtain high water flux [18].

The experiments were carried out at temperature of  $25 \pm 1^\circ\text{C}$ . During experiments, feed and draw solution were re-circulated using Pump I and Pump II with a flow rate of 9 l/min (MD-6Z, supplied by Iwaki Co. Ltd), respectively. Due to the intensive mixing, external concentration polarization phenomena were expected to be negligible.

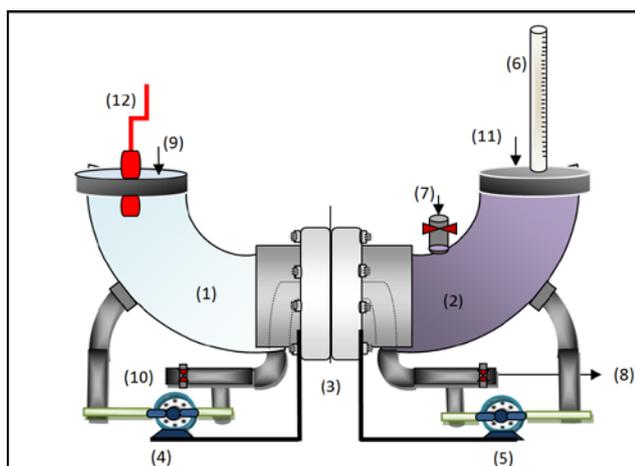


Figure 1. Lab scale FO unit [17]

Where, (1) Feed side (2.6L), (2) Draw solution side (2.6L), (3) FO membrane, (4) Recirculation Pump I, (5) Recirculation Pump II, (6) Measuring tube, (7) Draw solution inlet, (8) Draw side sampling, (9) Feed inlet (open compartment), (10) Feed sampling, (11) Sealed compartment, (12) pH reader.

Two types of feed water were used: 0.5 M NaCl solutions with different boron concentrations (3.5-7.0 mg/L) were used as synthetic seawater (SSW), and real seawater (RSW) which was collected from the Variable Salinity Plant of PUB Singapore. The RSW was pre-treated by ultrafiltration and was solids free. Different concentration of  $\text{NH}_4\text{HCO}_3$  was used as draw solution for the experiments. When the feed water was SSW, the draw solution was 2.5M  $\text{NH}_4\text{HCO}_3$  solution with osmotic pressure around 100 bar. When the feed water was RSW, draw solution was 1.5M or 2.5 M  $\text{NH}_4\text{HCO}_3$  with osmotic pressures around 60 bar and 100 bar, respectively. For overview, the designed experimental conditions are summarized in Table 1. Characteristics of the real seawater (RSW) are given in Table 2.

Table 1. Experimental conditions

S/N	Feed		Draw solution	Remark
	Composition	B concentration (mg/L)	$\text{NH}_4\text{HCO}_3$ concentration (M)	
Test 1	SSW	4.5	2.5	M1
Test 2	SSW	5.0	2.5	M1
Test 3	SSW	7.0	2.5	M1
Test 4	SSW	0	2.5	M2
Test 5	SSW	3.5	2.5	M2
Test 6	SSW	6.0	2.5	M2
Test 7	RSW	3.3	2.5	M2
Test 8	RSW	3.3	1.5	M2

Table 2. Characteristics of RSW

Parameter	Units	Value
pH		7.7
Conductivity	$\mu\text{S/cm}$	$3.6 \times 10^4$
Turbidity	NTU	0.3
Total Organic Carbon (TOC)	mg/l	2.1
Boron (B)	mg/l	3.3
Chloride (Cl)	mg/l	15500
Magnesium (as Mg)	mg/l	961
Sodium (Na)	mg/l	6877

The permeate flow rate, concentrations of solutes in feed and draw solution were measured during experiments. The experimental duration was 1-2 hrs. The parameters pH and conductivity were measured by a benchtop pH/conductivity meter (model WTW). Boron and metal ion analyses were performed using a Perkin Elmer model 2100 Inductively Coupled Plasma Emission Spectrometer. TOC was measured using Shimadzu TOC analyzer model 5000A as per USEPA 415.1 standard method.

Analysis on water flux, solute flux and rejection of the referred solute (Boron) is provided as per Eq.1, Eq.2 and Eq.3, respectively.

$$\text{Water flux } J_w = \Delta V / A_m / \Delta t \quad (1)$$

$$\text{Solute flux } J_s = \Delta W / A_m / \Delta t \quad (2)$$

$$\begin{aligned} \text{Rejection of solute} &= (1 - C_p / C_f) \times 100\% \\ &= (1 - J_s / J_w / C_f) \times 100\% \end{aligned} \quad (3)$$

Where:  $J_w$  is water flux, ( $\text{l/m}^2/\text{h}$  or LMH);  $\Delta V$  is the volume increase measured from the measuring tube, (L);  $A_m$  is effective membrane area, ( $\text{m}^2$ );  $\Delta t$  is the time recorded, (h);  $J_s$  is solute flux, ( $\text{g/m}^2/\text{h}$ );  $\Delta W$  is the mass increase of the solute measured from draw solution side, (g);  $C_f$  and  $C_p$  are the solute concentration in the feed and permeate (NOT the draw solution), respectively.

## 3. Results and Discussion

### 3.1. Characteristics of Feeds and Draw Solutions

Characteristics of feed and draw solutions in experiments are summarized in Table 3. The duration of each experiment was about 120 minutes. It is interesting to note that about one unit increase in pH of the feed was observed from start to end of the experiment in all tests except Test 8. This could be attributed to the reverse flow of  $\text{HCO}_3^-$  ions from the draw solution of 2.5 M  $\text{NH}_4\text{HCO}_3$  to the feed, resulting in a significant increase of  $\text{OH}^-$  concentration ( $\text{HCO}_3^- \rightarrow \text{OH}^- + \text{CO}_2$ ) in the feed. Meanwhile, the loss of  $\text{HCO}_3^-$  ions from the draw solution

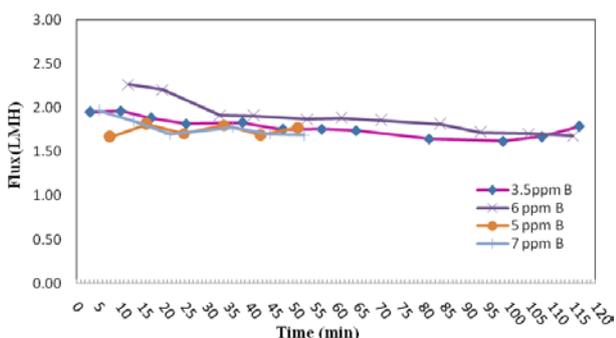
resulted in an increase of H<sup>+</sup> concentration (CO<sub>2</sub> + H<sub>2</sub>O → H<sup>+</sup> + HCO<sub>3</sub><sup>-</sup>), and hence a reduced pH. This observation is not well understood. However, it has implication on ions transport in a FO process and should be studied further. No significant change in conductivity was observed at both feed and draw solution sides during the experiments. Although the experiments were run in batch, the dilution of the draw solution due to permeation of feed water could be considered negligible because max net volume increase over the experimental duration was about 10 mL compared to the draw solution compartment volume at 2.6L (Less than 1% in volume). Quasi-constant concentration in the draw solution may therefore be assumed.

**Table 3. Characteristics of feed and draw solutions in experiments**

S/N	Feed		Draw solution	
	pH	Conductivity (μS/cm) x10 <sup>4</sup>	pH	Conductivity (μS/cm) x10 <sup>4</sup>
Test 1	8.1->9.0	NA ->4.76	8.6->8.3	NA ->12.60
Test 2	7.7->9.0	4.64->5.01	8.7->8.4	12.26->12.30
Test 3	7.8->9.0	4.66->4.97	8.5->8.3	12.20->12.46
Test 4	7.9->9.2	4.63->4.69	8.5->8.3	12.21->12.29
Test 5	7.6->9.1	4.60->4.88	8.6->8.3	12.21->12.52
Test 6	7.9->9.2	4.61->4.96	8.7->8.3	12.24->12.54
Test 7	7.7->8.7	3.60->3.71	8.5->8.4	12.43->12.48
Test 8	7.7->7.4	3.60->3.69	8.2->8.0	9.17-> 9.26

### 3.2. Determination of the Steady State

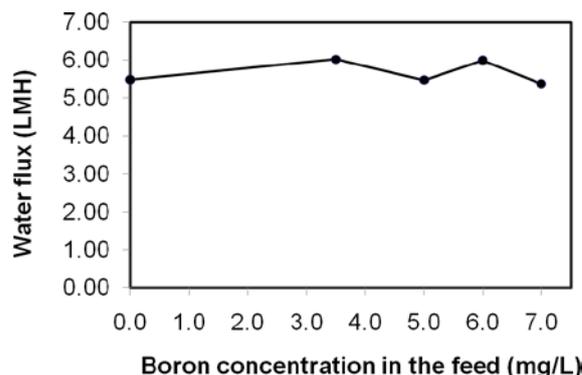
A couple of experiments were conducted prior to the test to ascertain the effect of varying Boron concentration on water flux (Figure 2). The results indicate that the experiment could reach a steady state (in terms of water flux) within 30 minutes. During this time, the water flux decreased with the time before levelled off to a stable value. This observation could be explained as the draw solution was pre-filled in the experimental system while the feed was filled just before the experiment started. Internal concentration polarization (ICP) occurred within the support layer of the membrane which faced the feed side. The development of the ICP resulted in a decrease in driving force and hence the flux within the first 30 minutes. The ICP stabilized thereafter and flux became stabilized correspondingly [19].



**Figure 2. Water flux vs. time**

Another set of experiments that tracked the steady state water flux under different Boron concentration indicates that boron concentration in the feed of SSW

had no significant influence on water flux when 2.5 M NH<sub>4</sub>HCO<sub>3</sub> was used as the draw solution (Figure 3).



**Figure 3. Water Flux vs. Boron Concentration in the Feed**

### 3.3. Challenges on Water Quality and Fouling

Analysis of boron concentration and rejection at different water fluxes in experiments are summarized in Table 4. Water flux in Group 2 (Tests 4-6) was much higher than that in Group 1 (Tests 1-3) due to different types of FO membranes. As a result, boron rejections (76-85%) in Group 2 were higher than that in Group 1 (57-67%). Large difference in water flux between Group 2 (the synthetic seawater) and Group 3 (Tests 7 & 8, real seawater) could be attributed to membrane fouling by compounds found in the real seawater and not in synthetic water. Water flux in Test 8 was lower than that in Test 7 due to the reduced concentration of draw solution used. Again, Boron rejection in Test 8 was lower than that in Test 7.

**Table 4. Analysis of boron concentration and rejection at different water fluxes**

S/N	Group	Water flux (LMH)	Boron concentration (mg/L)		Boron rejection (%)	Remark
			Feed	Permeate		
Test 1	1	2.1	4.55	2.06	57	M1, SSW
Test 2	1	2.0	-	-	-	M1, SSW
Test 3	1	2.2	7.15	2.33	67	M1, SSW
Test 4	2	5.4	0	-	-	M2, SSW
Test 5	2	6.0	3.43	0.820	76	M2, SSW
Test 6	2	6.0	6.06	0.918	85	M2, SSW
Test 7	3	1.7	3.30	1.44	56	M2, RSW
Test 8	3	1.5	3.3	2.07	39	M2, RSW

A few discussions could be made. Consistent with this study, Tang *et al.* [20] also found higher fouling potential with NOMs in the feed when the FO membrane active layer was faced to draw solutions. The water flux has significant impact on the boron rejection observed. With increasing water flux, higher boron rejection was achieved in an FO process due to dilution effect that is similar to RO process. This observation is consistent with Jin *et al.*

[21]. In that study, boron rejection in the range of 29-62% was observed with NaCl as draw solutions. The reduced boron rejection observed when using RSW as compared to SSW could be attributed to the greater fouling encountered under the RSW condition due to fouling-enhanced concentrative ICP effect. [20,21].

Mane *et al.* [22] associated the increase in boron rejection with the dilution effect at higher flux in RO studies. Similar trend was reported by Cengeloglu *et al.* [23], Koseoglu *et al.* [24], Prats *et al.* [25] and Magara *et al.* [26]. Based on the results of Group 3 in this study, boron concentration in the permeate could be dramatically reduced if a new FO membrane with much high water flux can be developed in the future, which could be a challenge for FO desalination [3].

The average water flux achieved with synthetic seawater (0.5 M NaCl) as feed and 2.5 M  $\text{NH}_4\text{HCO}_3$  as draw solution was 5.7 LMH in Group 2. McCutcheon *et al.* [6] obtained water flux of 13.5 LMH using similar feed and draw solution at 50°C and the temperature-corrected flux could be 6.4 LMH [19]. In terms of water flux, this study is consistent with the work of McCutcheon *et al.* [6], and the key difference is the experimental temperature.

For all the tests, boron concentrations in the permeate were greater than the previous WHO guideline for drinking water (0.5mg/L) but would be within the current one (2.4 mg/L) [27]. The results indicate that boron removal may be just adequate for seawater desalination using FO, and under specific circumstances, a further post-treatment for boron removal would be required.

Furthermore, it was also observed that the TOC in the draw solution of Test 7 was 9-10 mg/L, which was greater than the TOC in RSW feed at 2.1 mg/L as shown in Table 5. Analysis indicated that the TOC was non volatile organic and it was due to the  $\text{NH}_4\text{HCO}_3$  draw solution (2.5M). Such high TOC value would not be able to meet drinking water guideline.

**Table 5. Challenge on water quality for high TOC contaminant**

Feed (RSW) (mg/L)	Draw solution with FO permeate (mg/L)	Draw solution* (2.5M $\text{NH}_4\text{HCO}_3$ ) (mg/L)
2.1	9-10	9-10

\* non volatile organic in 2.5M  $\text{NH}_4\text{HCO}_3$

The results demonstrated that the product water quality using  $\text{NH}_4\text{HCO}_3$  as the draw solution could be a serious issue, which so far had not been discussed extensively in the literature. Another issue of concern is the membrane fouling for practical FO application in seawater desalination.

### 3.4. Challenge of Loss of $\text{NH}_4\text{HCO}_3$ Osmotic Agent

Another practical issue for implementation of FO with  $\text{NH}_4\text{HCO}_3$  osmotic agent is the loss of draw solute through the membrane into the feed ( $J_s'/J_w$ ). Table 6 shows the daily amount of  $\text{NH}_4\text{HCO}_3$  draw solute that needs to be replenished for a FO process due to loss through the membrane for different  $J_s'/J_w$  scenarios. The implication is that a municipal scale FO plant with capacity of 100,000  $\text{m}^3/\text{day}$  will lose at least 200,000 kg of  $\text{NH}_4\text{HCO}_3$  on a per day basis, which need replenishment. Additional production cost for topping-up of the loss of  $\text{NH}_4\text{HCO}_3$  will be  $\$0.4/\text{m}^3$  if  $\text{NH}_4\text{HCO}_3$  cost is  $\$0.2/\text{kg}$ . From a

logistics point of view, a FO process that uses  $\text{NH}_4\text{HCO}_3$  draw solute will not be feasible for a municipal scale application at the current membrane development. The limitation must be addressed via great enhancement of FO membrane performance with high water permeability & high solute selectivity such that  $J_s'/J_w \leq 0.01 \text{ g/L}$  [3]. This is a challenge to the FO research community. Future development needs to take into account the inter-dependent relationship between membrane development and draw solution selection, and under the consideration of cost-effectiveness.

**Table 6. Loss of  $\text{NH}_4\text{HCO}_3$  osmotic agent in FO**

$J_s'/J_w$ , ratio of reverse salt flux/water flux (g/L)	Loss of osmotic agent at 100,000 $\text{m}^3/\text{d}$ (ton/day)	Reference
2.9	290	[15]
2.0-2.5	200-250	[14]

Note:  $J_s'$  here refers to the flux the draw solute ( $\text{NH}_4\text{HCO}_3$ ) through the FO membrane into the feed water due to reverse diffusion [14,15].

## 4. Conclusions

From findings in this study, the following conclusions can be drawn:

- 1) When synthetic seawater solution was used as the feed water with boron concentration between 3.5 and 7.0 mg/L, boron rejection by the FO process was in the range of 57-85%. Boron rejection could be increased with increasing water flux.
- 2) When real seawater that was pretreated with UF was used as the feed water, serious membrane fouling was observed ( $J_w$  was 3 times lower compared to that with synthetic seawater) and boron rejection reduced to 39-56% due to fouling-enhanced concentrative ICP effect.
- 3) The impaired product water quality in terms of TOC and huge loss of osmotic agent when  $\text{NH}_4\text{HCO}_3$  is used as the draw solution are challenges for application of FO process for seawater desalination.

It is recommended that future development needs to take into account the inter-dependent relationship between membrane development and draw solution selection, and under the consideration of cost-effectiveness.

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