SEAWATER DESALINATION USING AIR GAP MEMBRANE DISTILLATION – AN EXPERIMENTAL STUDY ON MEMBRANE SCALING AND CLEANING

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ABSTRACT

The connection between operating temperature and membrane scaling/cleaning during an air gap membrane distillation (AGMD) process of seawater has been systematically elucidated in this study. Experimental and mathematically simulated data demonstrate the profound influences of feed salinity and membrane scaling on water flux at various operating temperatures. Feed salinity exerted significant impacts on water flux at high operating temperatures because of aggravated polarization effects. Membrane scaling and the subsequent membrane cleaning efficiency were also strongly affected by operating temperatures. Indeed, membrane scaling was more severe and occurred at a lower water recovery when operating at 60−50 °C (feed−coolant temperature) compared to that at 35−25 °C. Moreover, membrane cleaning with fresh water and vinegar was less effective for the membrane scaled at 60−50 °C compared to 35−25 °C. Finally, membrane cleaning using vinegar was much more efficient than fresh water. Given the availability of vinegar at household level, vinegar cleaning can potentially be a low cost and readily accessible approach for MD maintenance for small-scale seawater desalination applications in remote coastal communities.

Keywords: membrane distillation, air gap membrane distillation, membrane scaling, membrane cleaning, seawater desalination.

1. INTRODUCTION

Seawater desalination is a practical approach to secure drinking water supply for small and remote coastal communities around the world [1]. Large-scale seawater desalination using reverse osmosis (RO) and conventional thermal distillation such as multi-stage flash (MSF) has been effectively implemented to provide freshwater for large and centralized communities. Indeed, RO desalination requires a pressure of about 60 bar (hence the need for high-pressure pumps and duplex stainless steel materials), intensive pre-treatment, and skilled operators. On the other hand, MSF demands a large physical and energy footprint. As a result, both RO and

MSF desalination are not applicable for small and remote areas. Freshwater provision for these areas requires an alternative desalination process that can negate all the above drawbacks inherent in RO and MSF technologies.

Membrane distillation (MD) is a combination of conventional thermal distillation and a membrane separation process. MD utilizes a hydrophobic, microporous membrane as a physical barrier for separation and a temperature gradient across the membrane as the driving force for mass transfer of water. Given its notable merits, including a complete salt rejection, less susceptibility to feed concentration, process compactness, and particularly ability to use low-grade waste heat and solar energy, MD can possibly be an ideal alternative to RO and MSF for small-scale and stand-alone seawater desalination applications in remote coastal regions [2 - 4].

MD processes can be operated in four basic configurations, including direct contact MD, vacuum MD, sweeping gas MD, and air gap MD. Amongst these configurations, air gap MD (AGMD) exhibits the highest process thermal efficiency with the lowest process simplicity. Therefore, AGMD is the most widely used for small-scale seawater MD desalination [2, 3].

A major technical challenge to seawater MD application in remote areas is membrane scaling associated with the desire for a high process water recovery (i.e. the volumetric ratio between fresh water product and seawater feed). Membrane scaling results in a reduction in water flux and the quality of fresh water product, membrane damage, and energy consumption increase [5, 6].

Given the detrimental effects of membrane scaling, this study aimed to elucidate membrane scaling and cleaning in a seawater AGMD process. The mass transfer coefficient of the AGMD system was experimentally determined. Then, the influence of feed salinity and membrane scaling on water flux was examined. Finally, the efficiency of scaled membrane cleaning with fresh water and vinegar was investigated.

2. MATERIALS AND METHODS

The lab-scale AGMD unit used in this study consisted of a plate-and-frame AGMD membrane module, two variable-speed gear pumps for water circulation, a heating element to heat the seawater feed, and a chiller to cool the coolant. Flat-sheet polytetrafluoroethylene (PTFE) membrane provided by Porous Membrane Technology (Ningbo, China) was used in the AGMD membrane module. The membrane had thickness, nominal pore size, and porosity of 60 μm, 0.2 μm, and 80 %, respectively.

Milli-Q water and seawater were used as the feed. Milli-Q water had electrical conductivity of 45±5 μS/cm. Seawater was collected from Wollongong beach (New South Wales, Australia), and pre-filtered by 0.45 μm filter papers. The pre-filtered seawater had total dissolved solids (TDS), electrical conductivity, and pH of 37,000 ± 2000 mg/L, 52.5 ± 1.0 mS/cm, and 8.35 ± 0.05, respectively. The total organic carbon (TOC) concentration of this pre-filtered seawater was less than 2 mg/L. Fresh water and a vinegar purchased from a local super market were used as cleaning agents in AGMD membrane cleaning experiments. As per instructions, the vinegar had acetic acid content of 8.0 ± 0.5 % and had pH of 2.55 ± 0.05.

AGMD of Milli-Q water was conducted to experimentally determine the process mass transfer coefficient (K_m). The water flux (J) of the process with Milli-Q water was measured at various feed and coolant temperatures. Then, K_m could be calculated as:

$$K_m = \frac{J}{\Delta P} \tag{1}$$

where K_m and J were in L.Pa^{-1}.m^{-2}.h^{-1} and in L.m^{-2}.h^{-1}, respectively; ΔP was the water vapor pressure difference between the feed and coolant streams (Pa). ΔP was calculated as:

$$\Delta P = P^0_{feed} - P^0_{coolant} \tag{2}$$

The water vapor pressure of the feed and coolant stream was calculated using the Antoine equation:

$$P^0 = exp\left(23.1964 - \frac{3816.44}{T - 46.13}\right) \tag{3}$$

where T was the temperature of the stream.

AGMD of seawater was operated in batch mode. Seawater feed (4 L) was continuously concentrated until the process water flux declined to zero or a process water recovery of 80% was achieved. Then, membrane cleaning with fresh water or commercial vinegar was initiated. Membrane cleaning was conducted at the same water circulation rates and at room temperature (i.e. 25 °C). Membrane cleaning efficiency was assessed based on the restoration of membrane surface hydrophobicity using contact angle measurement, and the visual analysis of membrane surface using scanning electron microscope (SEM) images.

During the AGMD process with seawater, the presence of dissolved salts reduced the water activity of the feed solution, thus lowering its water vapor pressure as expressed in Eq. (4):

$$P_{feed} = x_{water} a_{water} P^0 \tag{4}$$

where a_{water} was dependent on feed salinity as:

$$a_{water} = 1 - 0.5 x_{salt} - 10 x_{salt}^2 \tag{5}$$

where x_{salt} and x_{water} were the molar fraction of salt and water in the feed. In addition, concentration polarization effect in AGMD rendered the salt concentration at the membrane surface ($C_{m.f}$) higher than that in the bulk feed solution ($C_{b.f}$). The polarization effect was dependent on the process water flux as expressed in Eq. (6):

$$\frac{C_{m.f}}{C_{b.f}} = \exp\left(\frac{J}{k}\right) \tag{6}$$

where k was the mass transfer coefficient of salt.

3. RESULTS AND DISCUSSIONS

3.1. Mass transfer of AGMD with Milli-Q water

Increasing feed−coolant temperature while maintaining a constant temperature difference (ΔT) between the feed and coolant stream resulted in an increase in water flux but a decrease in mass transfer coefficient (Figure 1). The increase in water flux at higher feed−coolant temperature could be attributed to the exponential relationship between water vapor pressure and temperature as demonstrated in Eq. (3). Indeed, elevating feed−coolant temperature from 35−25

to 60−50 °C increased ΔP from 1.28 to 3,68 kPa, thus increasing water flux from 2.5 to 5 L.m^{-2}.h^{-1}. By contrast, K_m decreased from 1.8×10^{-3} to 1.3×10^{-3} L.Pa^{-1}.m^{-2}.h^{-1} (Figure 1). The decrease in K_m with increased feed−coolant temperature demonstrated the influence of temperature polarization on water flux of AGMD. The values of K_m were determined using the measured temperatures of the feed and the coolant streams instead of temperatures at the feed membrane surface and at the condenser surface. Temperature polarization effect rendered the temperature difference between the feed membrane surface and the condenser surface (i.e. the actual driving force of the process) smaller than that between the feed and coolant stream (ΔT), thus reducing water flux and hence K_m of the process. Increasing feed−coolant temperature elevated water flux and therefore magnified temperature polarization effect.
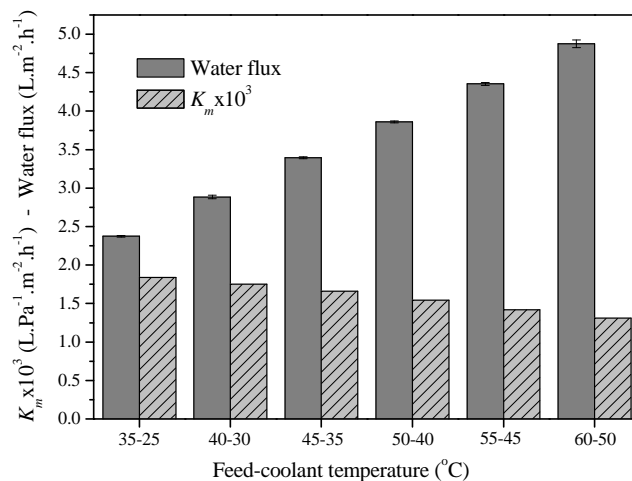

Figure 1. Experimentally measured water flux and mass transfer coefficient (K_m) during AGMD process with Milli-Q water at various feed−coolant temperature, a constant ΔT of 10 °C, and water circulation rate $F_{feed} = F_{coolant} = 0.5$ L/min. Error bars represent the standard deviation of water flux measurements.

3.2. AGMD of seawater

The influence of feed salinity on water flux and distillate conductivity during AGMD of seawater is demonstrated in Figure 2. Increasing feed salinity during the concentration of seawater led to a reduction in water flux of AGMD. This was attributed to the decrease in water activity and thus the reduction in water vapor pressure of the feed stream with increased feed salinity as expressed in Eqs. (4−5) [7].

Compared to simulated water flux, the experimentally measured flux decreased more as the seawater feed was concentrated (Figure 2). The deviation between the experimentally measured and the simulated flux was because of concentration polarization effect and membrane scaling. The simulation of water flux using the K_m values obtained during AGMD of Milli-Q water excluded the influence of concentration polarization effect. Concentration polarization effect caused the salt concentration at the membrane surface higher than that in the bulk feed solution, thus reducing water flux. Operating AGMD at higher feed−coolant temperature and hence higher water flux aggravated concentration polarization as expressed in Eq. (6) [7]. Therefore, the deviation between the measured and simulated water flux was more at feed−coolant temperature of 60−50 °C compared to that at 35−25 °C (Figure 2).
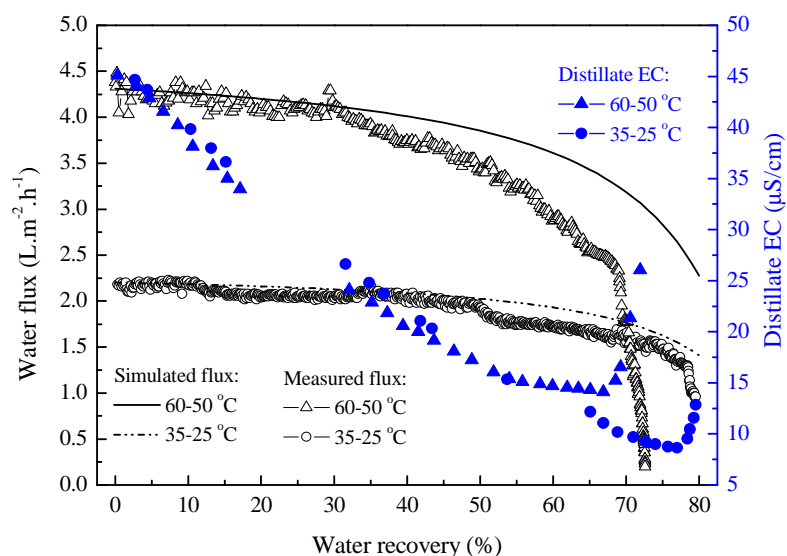
Figure 2. Simulated and experimentally measured water flux and distillate electrical conductivity (EC) as functions of process water recovery during AGMD with seawater. Water circulation rate $F_{feed} = F_{coolant} = 0.5$ L/min.

Membrane scaling caused by the precipitation of sparingly soluble salts (e.g. $CaSO_4$ and $MgSO_4$) in seawater further reduced the measured flux (Figure 2). As the seawater feed was concentrated, the concentration of these salts exceeded their saturation limits. Concentration polarization effect further increased the supersaturation of the salts at the membrane surface, leading to the formation of scale layers on the membrane. The scale layers promoted temperature and concentration polarization effects, and reduced water vapor pressure at the membrane surface and the active membrane area for water evaporation [8, 9]. Therefore, water flux rapidly decreased from 2.5 $L.m^{-2}.h^{-1}$ to almost zero and from 1.5 to 1.0 $L.m^{-2}.h^{-1}$ as the process water recovery exceeded 70 % and 75 % at feed−coolant temperature of 60−50 and 35−25 °C, respectively (Figure 2).

Membrane scaling also resulted in decline in distillate quality (Figure 2). At the beginning of the AGMD process, the electrical conductivity of the distillate gradually decreased. The gradual decrease in distillate conductivity before membrane scaling demonstrated the ability of AGMD for pure water production from seawater. Indeed, distillate with conductivity as low as 10 μS/cm was obtained from seawater. When membrane scaling occurred, the scale layers altered the hydrophobicity of the membrane surface [10, 11], leading to partial intrusion of seawater through the membrane pores. Consequently, distillate conductivity started increasing following the formation of scale layers on the membrane (Figure 2).

Operating feed−coolant temperature influenced not only the water flux but also membrane scaling in AGMD of seawater. Increasing feed−coolant temperature from 35−25 to 60−50 °C doubled water flux, and at the same time escalated membrane scaling. Membrane scaling occurred at a lower water recovery when operating at 60−50 °C compared to that at 35−25 °C (Figure 2). The operating temperature also affected the efficiency of subsequent membrane cleaning as will be discussed in the next section.

3.3. Efficiency of membrane cleaning

The efficiency of membrane cleaning could be evaluated by the restoration of membrane surface hydrophobicity. Compared to fresh water, vinegar demonstrated a superior membrane cleaning efficiency under the same cleaning conditions (i.e. water circulation rates, temperature, and cleaning duration) (Figure 3). Given its hydrophobic nature, the contact angle of the virgin membrane used in this study was 130°. The scale layers formed at the membrane rendered its surface so hydrophilic that its contact angle could not be determined. Cleaning the scaled membrane with vinegar effectively removed scalants from the membrane surface, thus returning it to a hydrophobic condition (i.e. contact angle of the scaled membrane at 60−50 and 35−25 °C increased to 120° and 125°, respectively, after cleaning with vinegar). It is worth noting that the vinegar contained a high content of acetic acid that might have increased the solubility and thus the removal of the sparingly soluble salts from the membrane surface. The slight decrease in contact angle of the vinegar cleaned membrane compared to the virgin membrane was expected because deterioration in membrane hydrophobicity has been reported in DCMD process with only pure water [5]. Cleaning the scaled membrane with fresh water was unable to remove all scale deposits from the membrane surface (i.e. confirmed by SEM images of the scaled membrane surfaces). The scales remained on the membrane surface significantly reduced its hydrophobicity. Thus, the contact angle of the scaled membrane surface following fresh water cleaning was far below 90° (Figure 3).

The operating feed−coolant temperature slightly affected the efficiency of subsequent scaled membrane cleaning. Cleaning with both vinegar and fresh water was more efficient for the membrane scaled at 35−25 °C compared to that at 60−50 °C (Figure 3). As discussed above, membrane scaling at 60−50 °C was more severe than at 35−25 °C, resulting in thicker and possibly more compacted scale layers at 60−50 °C compared to 35−25 °C. The morphology of the scale layers appeared to exert an effect on the efficiency of the subsequent membrane cleaning.
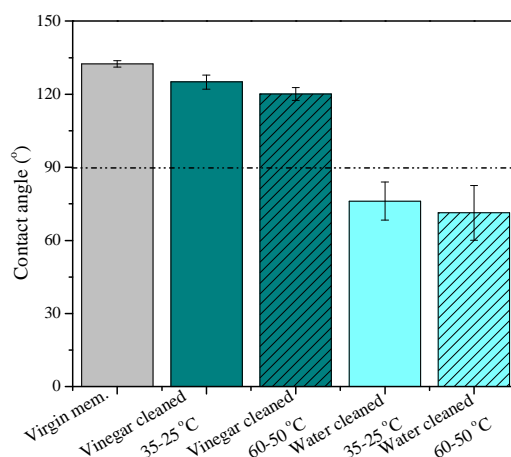

Figure 3. Contact angles of the virgin membrane and the scaled membranes at 35−25 and 60−50 °C after cleaning with vinegar and fresh water. Error bars represent the standard deviation of 5 repeated measurements.

The results reported here have significant implications for pilot or small-scale seawater AGMD application, in which membrane modules with long membrane channels are employed.

Along the membrane channels, the feed temperature significantly decreases from 70 to 35 °C, and the coolant temperature increases from 25 to 60 °C [12, 13]. The change in feed−coolant temperature will result in an uneven distribution of water production along the membrane channels − more distillate is obtained at the higher temperature end of the membrane module compared to the low temperature end. Also because of the uneven distribution of feed−coolant temperature, membrane scaling will occur at the high temperature membrane area before the low temperature one. Finally, when membrane scaling occurs, it will be harder to clean the membrane area scaled at higher temperature compared to that at low temperature. The scale remnants on the membrane after cleaning will act as crystal nuclei, and thus accelerating membrane scaling in the next seawater AGMD cycle [6]. As a result, repetitive membrane scaling and cleaning in AGMD of seawater will inevitably lead to deterioration in process performance. In this context, effective scaling prevention techniques, including but are not limited to anti-scalant addition [14 – 16], utilization of ultrasonic and gas bubbling [17, 18], or process optimization [9, 19, 20], are highly recommended.

4. CONCLUSIONS

AGMD of an actual seawater feed were investigated. The experimental results demonstrate a profound influence of feed salinity and operating temperature on water flux, scaling behavior, and the efficiency of subsequent membrane cleaning. Feed salinity reduced the water flux of the AGMD process with seawater compared to that with fresh water, particularly at higher operating temperatures due to the aggravated polarization effects. Increasing feed−coolant temperature from 35−25 to 60−50 °C doubled water flux but also escalated membrane scaling during seawater desalination with AGMD. At feed−coolant temperature of 60−50 °C, membrane scaling occurred at a lower water recovery compared to that at 35−25 °C. The efficiency of membrane cleaning with fresh water and vinegar was also lower for the membrane scaled at 60−50 °C compared to at 35−25 °C. Vinegar cleaning demonstrated a superior efficiency to fresh water cleaning. Given the accessibility to vinegar at household level, membrane cleaning using vinegar can be a practical scaling control method for small-scale seawater MD desalination applications.

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