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Membrane scaling during seawater desalination by direct contact membrane distillation

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Abstract

Seawater desalination by membrane distillation (MD) has great potential for fresh water provision in small and remote areas. Amongst four basic MD configurations, direct contact membrane distillation (DCMD) has a simple arrangement; thus, it is most suited for small-scale seawater desalination application. In this study, membrane scaling during a seawater DCMD desalination process was systematically investigated. Mass transfer coefficient of the DCMD system was first determined with Milli-Q water. The obtained mass transfer coefficient was used to simulate the influence of feed salinity increase and membrane scaling on water flux. The simulation results were then validated by experimental data. Results reported here demonstrate a notable influence of feed salinity increase and membrane scaling on water flux, particularly at a high water recovery. The rapid increased feed salinity during the concentration of seawater at water recoveries above 50 % magnified both temperature and concentration polarization effects, thus reducing the experimentally measured water flux compared to the calculated one. In addition, membrane scaling caused by the precipitation of $CaSO_4$ and $MgSO_4$ at high water recoveries further reduced the measured water flux. Moreover, feed operating temperature had a profound effect on both water flux and membrane scaling. Increasing feed temperature favored higher water flux but also escalated membrane scaling. Finally, a DCMD process of seawater at a water recovery of 70 % without any observable membrane scaling was obtained either by operating the process at a reduced feed temperature or by anti-scalant addition. The results reported in this study demonstrate the viability of DCMD for small-scale seawater desalination in Vietnam given its long coastline together with a large number of islands and great solar energy availability.

Keywords. Membrane distillation (MD), direct contact membrane distillation (DCMD), seawater desalination, membrane scaling, scaling mitigation techniques.

1. INTRODUCTION

Sufficient fresh water provision for small communities in remote areas remains a considerable challenge. Large-scale seawater desalination using reverse osmosis (RO) and conventional thermal distillation (e.g. multi-stage flash, and multi-effect distillation) has been implemented to effectively supply fresh water for centralized communities [1]. Indeed, RO desalination, which is a pressure driven filtration process, requires high-pressure pumps and hence duplex stainless steel piping, intensive physical and chemical pre-treatment, and skilled operators. Similarly, conventional distillation processes require large physical footprint and are considered energy-intensive [2]. As a result, both RO and conventional thermal distillation might not be an ideal technology platform for fresh water supply in small and remote areas. Freshwater provision for these areas requires a small-scale, robust, and economically feasible desalination process.

Membrane distillation (MD), which is a combination of conventional thermal distillation and a membrane separation process, can be a promising candidate for small-scale seawater desalination application in remote areas. In MD, a hydrophobic microporous membrane is used as a physical barrier to prevent the permeation of liquid water while allowing the transfer of water vapor through the membrane pores [3, 4]. As a result, in seawater MD desalination all dissolved salts and nonvolatile compounds are retained by the membrane, and ultra-pure water can be obtained as the distillate [3, 4]. In

addition, unlike RO, MD utilizes a water vapor pressure difference induced by a temperature gradient across the membrane as its driving force. Thus, water flux in MD is negligibly affected by the osmotic pressure of the feed, allowing MD operation at higher water recoveries than RO [4]. More importantly, given the absence of high hydraulic pressure, components for a MD system can be made from inexpensive plastic materials, thus resulting in considerable cost savings. Finally, energy supply to MD processes can be sourced from low-grade waste heat or solar thermal energy given its operating temperature in the range from 40 to 80 °C [5-7].

MD has been practiced in four basic configurations, including direct contact membrane distillation (DCMD), air gap membrane distillation (AGMD), vacuum membrane distillation (VMD), and sweeping gas membrane distillation (VMD). Amongst these configurations, DCMD has the simplest process arrangement with both feed and distillate streams in direct contact with the membrane [3, 4]. As a result, DCMD has been the most widely used configuration in the MD literature, and it is deemed the best suited for small-scale seawater desalination application [4, 8]. Also because of its simple arrangement, DCMD exhibits lower process thermal efficiency compared to other configurations [8]. However, the thermal efficiency limitation of DCMD can be tolerated given the availability of waste heat or solar thermal energy on site.

A major technical challenge to seawater DCMD desalination 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). At high process recoveries, sparingly soluble salts present in seawater can exceed their saturation limits and precipitate to form scale layers on the membrane surface. The formation of scales on the membrane results in reduction in water flux and the quality of fresh water product, membrane damage, increased energy consumption, and thus increasing operation costs [9-12].

Given the detrimental effects of membrane scaling, this study aimed to investigate membrane scaling during a DCMD process of actual seawater. First, the mass transfer coefficient of the DCMD system with Milli-Q water at various operating conditions was experimentally determined. Given the mass transfer coefficient, the influence of increased feed salinity and particularly membrane scaling on water flux during DCMD concentration of seawater was examined. Finally, membrane scaling mitigation techniques, including optimizing the feed temperature and anti-scalant addition, were demonstrated for a seawater DCMD desalination process at high water recoveries for an extended period.

2. MATERIALS AND METHODS

2.1. Materials

2.1.1. The lab-scale DCMD system

A schematic diagram of the lab-scale DCMD system used in this study is shown in Fig. 1. The system employed a plate-and-frame membrane module composed of two acrylic semi-cells and a hydrophobic flat-sheet PTFE membrane. Two semi-cells were engraved to form flow channels with depth, width, and length of 0.3, 9.5, and 14.5 cm, respectively, generating an active membrane area of 138 cm^2 for water transfer. The flat-sheet PTFE membrane, provided by Porous Membrane Technology (Ningbo, China), had thickness, nominal pore size, and porosity of 60 μm, 0.2 μm. and 75%, respectively.

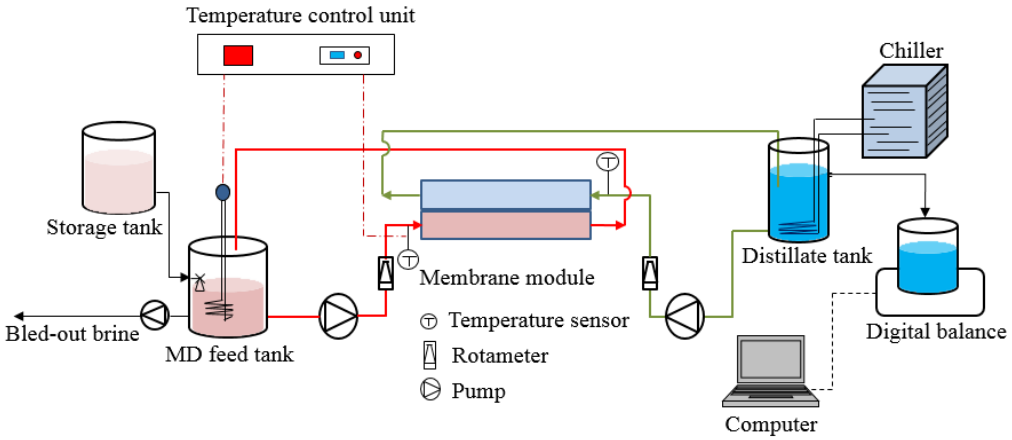

Figure 1: The schematic diagram of the lab-scale DCMD system

Pre-filtered seawater from the storage tank flowed into the MD feed tank via a float valve by gravity (Fig. 1). The seawater was heated in the feed tank using a heating element connected to a temperature control unit. A temperature sensor placed immediately before the inlet of the feed channel was connected to the temperature control unit to regulate the feed temperature. A chiller (SC200-PC, Aqua Cooler, Sydney, New South Wales, Australia) was used to control the distillate temperature through a stainless steel heat-exchanging coil submerged directly into the distillate reservoir. Two variable-speed gear pumps (Model 120/IEC71-B14, Micropump Inc., Vancouver, Washington, USA) were used to circulate the feed and distillate through the feed and distillate channel, respectively. Two rotameters, positioned before the inlet of each channel, were used to monitor the circulation rates of the feed and distillate. A digital balance (PB32002-S, Mettler Toledo, Inc., Hightstown, New Jersey, USA) connected to a computer was used to weigh the excess distillate flow for determining the water flux.

2.1.2. Feed solutions and anti-scalant

Milli-Q water and pre-filtered seawater were used as feed solutions. Milli-Q water having electrical conductivity of 10±2 μS/cm was produced by a Milli-Q® Integral Water Purification System (Merck Millipore, Australia). Seawater was collected from Wollongong beach (New South Wales, Australia) and was pre-filtered by 0.45 μm filter papers prior to all experiments. The pre-filtered seawater had conductivity, pH, and total dissolved solids (TDS) of 52.5±0.5 mS/cm, 8.35±0.05, and 37,000±2000 mg/L, respectively. The total organic carbon (TOC) concentration of this pre-filtered seawater was less than 2 mg/L.

A commercial anti-scalant, Osmotreat OSM35 (Osmoflo Pty Ltd, Adelaide, Australia), was used in the DCMD experiment with seawater at 70 % water recovery. According to the manufacture, Osmotreat OSM35 can inhibit a broad spectrum of scalants, including the sparingly soluble salts of calcium and magnesium.

2.2. Analytical methods

A Rame-Hart Goniometer (Model 250, Rame-Hart, Netcong, New Jersey, USA) was used to measure the contact angle of the membrane surface following the standard sessile drop method. Milli-Q water was used as the reference liquid. At least 5 droplets (i.e. each with volume of 12 μL) were tested for each membrane sample.

A low vacuum scanning electron microscope (SEM) coupled with an energy dispersive spectrometer (EDS) (JOEL JSM-6490LV, Japan) was used to examine the morphology and composition of membrane surfaces. Membrane samples were air-dried and subsequently sputtered with a thin layer of gold prior to SEM-EDS analysis.

Orion 4-Star Plus meters (Thermo Scientific, Waltham, Massachusetts, USA) were used to monitor the electrical conductivity (EC) of the feed and distillate during DCMD experiments with the pre-filtered seawater.

2.3. Experimental protocols

DCMD of Milli-Q water was conducted to characterize the system and to determine its mass transfer coefficient. Milli-Q water at temperature of 40, 50, and 60 °C was introduced to the feed channel at flow rate of 0.5, 0.75, and 1.0 L/min (i.e. equivalent to cross flow velocity of 0.03, 0.045, 0.06 m/s, respectively). The distillate at a constant temperature of 25 °C was circulated though the distillate channel at the same flow rate to the feed. Water flux of the process at each operating conditions was measured for 1 hour after the attainment of stable operation. The water flux of the process was calculated as:

$$J = \frac{\Delta V_{distillate}}{S \times \Delta t} \quad (1)$$

where J was the water flux (L/m^2.h), $\Delta V_{distillate}$ was the volume of distillate (L) obtained in a time interval Δt (h), and S was the active membrane surface for water evaporation (m^2).

DCMD of pre-filtered seawater was operated under the same conditions as described above. Two operation modes, namely concentrating and constant recovery, were employed. The concentrating mode was operated in the experiments to examine the influence of increased feed salinity and membrane scaling on the process performance. During the concentrating operation, the volume of feed solution in the feed tank was allowed to decrease, thus resulting in an increase in feed salinity over time. The water recovery of the system in this mode was the ratio between the accumulated distillate volume and the initial feed volume. The constant recovery mode was operated in the DCMD experiment at 70 % water recovery using membrane scaling mitigation techniques. The pre-filtered seawater was first concentrated by the DCMD process. When the process had reached 70 % water recovery, the

constant recovery mode operation was initiated by bleeding out the concentrated brine while allowing the pre-filtered seawater to flow into the MD feed tank (Fig. 1). The brine bled-out flow rate was calculated as:

$$F_{brineout} = \frac{3}{7} F_{distillate} \quad (2)$$

where $F_{brineout}$ and $F_{distillate}$ were the volumetric flow rates (m^3/s) of bled-out brine and produced distillate, respectively. Given this ratio between the bled-out and distillate flow rate, a constant feed concentration and thus a constant process water recovery of 70 % could be obtained. The DCMD process at 70 % water recovery was maintained for at least 24 hours. At the end of the experiments with the pre-filtered seawater, the membrane sample was removed for subsequent contact angle measurement and SEM-EDS analysis.

2.4. Mass transfer of water in DCMD

The mass transfer of water across the membrane in DCMD could be expressed as:

$$J = K_m \Delta P \quad (3)$$

where K_m was the mass transfer coefficient ($L/Pa.m^2.h$); ΔP was the water vapor pressure difference between the vapor-liquid interfaces formed at two sides of the membrane (Pa). The mass transfer coefficient is a function of membrane properties and operating conditions, including feed and distillate temperatures and water circulation rates. K_m can be determined using empirical correlations [13, 14] or experimentally measured [8].

The vapor pressure of pure water at the membrane surface was calculated using the Antoine equation:

$$P^0 = exp\left(23.1964 - \frac{3816.44}{T - 46.13}\right) \quad (4)$$

where P^0 was in Pa and T was the temperature in K. For seawater feed, the water vapor pressure at the membrane surfaces (P) was calculated as [3]:

$$P = x_{water}\ 1 - 0.5x_{salt} - 10x_{salt}^2\ P^0 \quad (5)$$

where x_{water} and x_{salt} were the molar fraction of water and salts, respectively.

Temperature and concentration polarization effects are intrinsic problems for MD, particularly DCMD, processes with saline solution feeds (Fig. 2). For the DCMD process of Milli-Q water, x_{salt} was negligible and thus the concentration polarization effect could be ignored. On the other hand, due to temperature polarization, the actual transmembrane temperature difference ($T_{f,m}$-$T_{p,m}$) was smaller than that between the bulk feed and distillate stream (T_f-T_p), thus reducing the driving force for mass transfer. However, the effect of temperature polarization could be incorporated into the mass transfer coefficient, K_m, and ΔP could be calculated using the temperature of the feed and distillate stream, which were measured using temperature the sensors (Fig. 1).

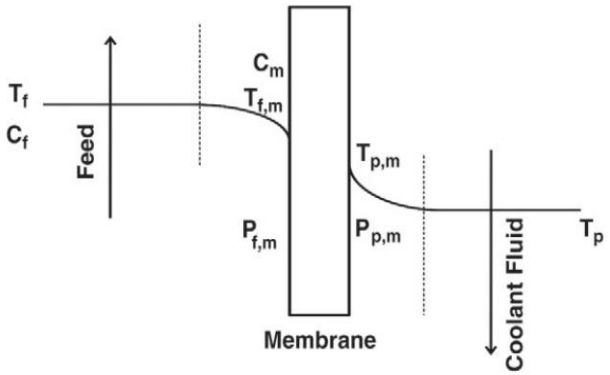

Figure 2: Temperature and concentration polarization effects in DCMD (adapted from [4])

3. RESULTS AND DISCUSSION

3.1. Characterization of the DCMD system with Milli-Q water

Feed temperature and water circulation rates exerted strong influence on the water flux of the DCMD process with Milli-Q water. As expressed in Eq. (4), increasing feed temperature resulted in an exponential increase in the water vapor pressure difference between the feed and distillate stream, thus favoring a higher water flux. Indeed, the water flux of the DCMD process increased by 40%, 45%, and 50 % when elevating the feed temperature from 40 to 60 °C at water circulation rates of 0.5, 0.75, and 1.0 L/min, respectively (Fig. 3A). Operating the DCMD process with Milli-Q water at higher water circulation rates also elevated water flux. Increasing water circulation rates promoted turbulence of the feed and distillate stream, and thus mitigated temperature polarization effect, hence leading to an increase in water flux. It is noteworthy that temperature polarization effect of DCMD escalates with increased feed temperature. As a result, water circulation rates exerted a greater influence on water flux in the DCMD process at higher feed temperature (Fig. 3A).

Compared to water flux, the process mass transfer coefficient (K_m) was influenced by feed temperature and water circulation rates in different manners (Fig. 3B). It should be noted that temperature polarization effect was incorporated into the experimentally measured K_m of the DCMD

process. Temperature polarization effect rendered the temperature at the feed membrane surface lower than in the bulk feed and at the distillate membrane surface higher than in the bulk distillate (Fig. 2), thus reducing the actual driving force of the DCMD. As a result, temperature polarization effect negatively affected the mass transfer coefficient of the process. Increasing feed temperature escalated temperature polarization effect, thus resulting in decreased K_m. In contrast increasing water circulation rates helped mitigating temperature polarization effect. As a result, K_m increased with water circulation rates (Fig. 3B).

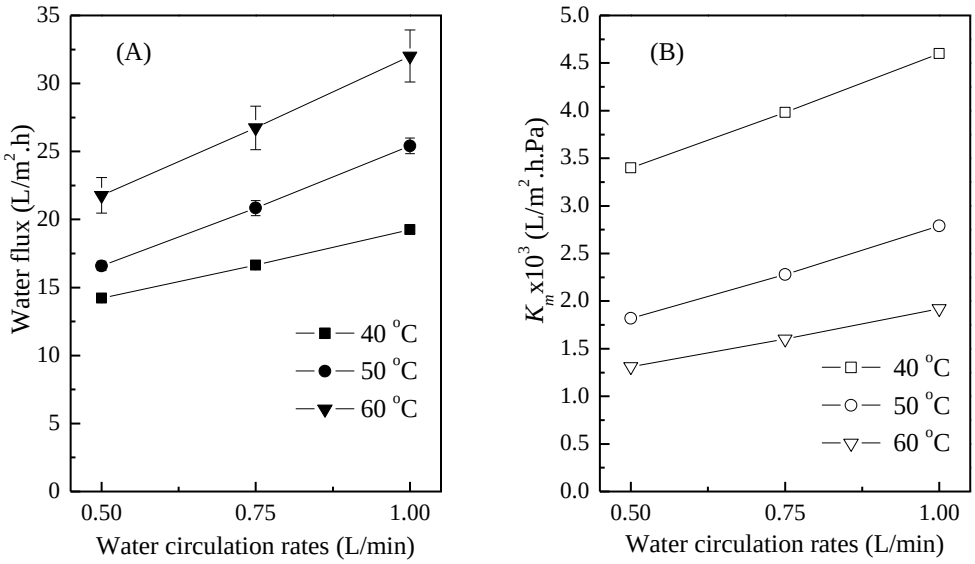

Figure 3: The influence of feed temperature and water circulation rates on (A) water flux and (B) the mass transfer coefficient of the DCMD system with Milli-Q water at a constant distillate temperature, $T_{distillate}$, of 25 °C

3.2. DCMD with pre-filtered seawater

The above K_m values were obtained during a DCMD process with Milli-Q water, in which the concentration polarization effect was negligible. For the DCMD of the pre-filtered seawater, the concentration polarization effect existed, thus affecting water flux of the process. However, the determined K_m values were useful for the preliminary evaluation of increased feed salinity during the concentration of seawater on water flux of DCMD.

Increase in feed salinity associated with increased process water recovery during DCMD concentration of seawater resulted in a decrease in water flux (Fig. 4). Increasing feed salinity reduced both water molar fraction and water activity (i.e. as expressed in Eq. 5), thus leading to a reduction in the water vapor pressure of the seawater feed. When the distillate temperature was maintained constant at 25 °C, the water vapor pressure of the distillate stream was constant. The reduction in the water vapor pressure of the feed reduced the water vapor pressure difference across the membrane, which was the driving force of the DCMD process. As a result, water flux decreased with increased water recovery (Fig. 4). It is noteworthy that the negative influence of increased feed salinity on water flux at process water recoveries below 50 % was unnoticeable. This again confirms the advantage of MD over RO for seawater desalination.

At high process water recoveries (i.e. > 50 %), both temperature and concentration polarization effects were magnified due to the rapid increase in feed salinity and hence the feed viscosity with increased water recovery. For DCMD, the temperature polarization effect was significant. In addition, the concentration polarization effect rendered the salt concentration at the membrane surface higher than in the bulk feed, hence further reducing water flux. As a result, the experimentally measured water flux at high process water recoveries deviated from the calculated flux (Fig. 4). The deviation was stronger for the process having higher water flux because increasing water flux exacerbated both temperature and concentration polarization effects [15, 16].

In addition to magnified polarization effects, scale formation on the membrane surface further reduced water flux of the DCMD process at high water recoveries. The experimentally measured water flux was significantly lower than the

calculated values when the process reached 80 % water recovery, particularly at feed temperature of 60 °C (Fig. 4). The scale layers aggravated temperature and concentration polarization effects and reduced water vapor pressure at the membrane surface [17]. They also reduced the active membrane surface area for water evaporation. As a result, water flux decreased rapidly following the occurrence of membrane scaling at high water recoveries.

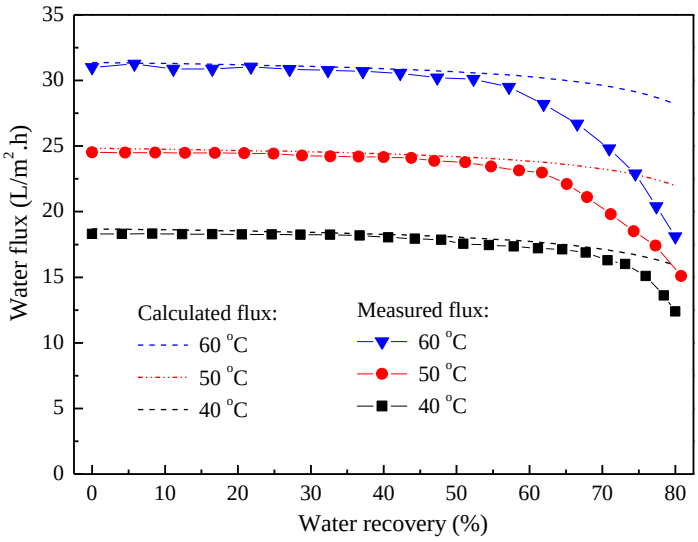

Figure 4: Calculated and experimentally measured water flux as functions of water recovery during DCMD of pre-filtered seawater. Operating conditions: distillate temperature, $T_{distillate}$, of 25 °C, water circulation rates F_{feed} = $F_{distillate}$ = 1.0 L/min

The analyses of the membrane surfaces at the end of DCMD experiments with pre-filtered seawater confirm membrane scaling occurrence. Membrane surface was covered by layers of salt crystals (Fig. 5). Nevertheless, the scale layers did not totally prevent the transfer of water vapor through the membrane given their porous nature. Indeed, at the end of the experiment (i.e. water recovery of 80 %), water flux of the process was 12, 15, and 18 L/m^2.h at feed temperature of 40, 50, and 60 °C, respectively. The EDS analyses of the the virgin and scaled membranes reveal that the scale layers mainly composed of calcium and magnesium salts of sulfate. The formed scale layers also altered the hydrophobicity of the membrane surface and rendered it so hydrophilic that its contact angle could not be measured by the sessile drop method. It is worth mentioning that the contact angle of a virgin PTFE membrane was 130°.

Operating feed temperature exerted a notable influence on not only water flux but also membrane scaling during DCMD of seawater. Increasing feed temperature from 40 to 60 °C nearly doubled the initial water flux of the process. However, increasing feed temperature and the resultant increase in water flux also magnified polarization effects and promoted membrane scaling. Given the temperature-inversed solubility of $CaSO_4$ (i.e. at temperature above 40 °C), which mainly composed the scale layers, increasing feed temperature depressed the solubility of $CaSO_4$. Concentration polarization raised the concentration of $CaSO_4$ at the membrane surface. As a result, operating the process at higher feed temperature increased the supersaturation of $CaSO_4$ at the membrane surface, leading to more severe membrane scaling. The SEM analyses of scales membranes (Fig. 5) also confirmed the influence of feed temperature on the severity of membrane scaling. Larger and more orthorhombic scale crystals were formed at higher feed temperature.

3.3. Membrane scaling mitigation during DCMD

Two membrane scaling mitigation techniques, including reducing feed temperature and adding anti-scalant to the feed, were deployed for the DCMD process of seawater at constant water recovery of 70%. A stable DCMD operation with pre-filtered seawater feed without anti-scalant addition at feed temperature of 40 °C and 70 % water recovery was obtained for 24 hours. Both water flux and distillate EC of the process remained stable throughout the operation (Fig. 6). This could

be attributed to reduced supersaturation levels of scalants at the membrane surface achieved by lowering feed temperature and thus water flux and polarization effects. A similar stable operation was obtained for the DCMD process at 60 °C and 70 % water recovery with the pre-filtered seawater feed dosed with 0.5 mg/L of anti-scalant. The added anti-scalant increased the induction time and thus delayed to crystallization of salts. As a result, the scale formation on the membrane surface was effectively prevented. It is worth noting that the DCMD process with scale mitigation techniques could produce distillate of superior quality compared to seawater RO – the MD distillate with EC as low as 3 μS/cm was obtained from seawater even at a process water recovery of 70 % (Fig. 6).

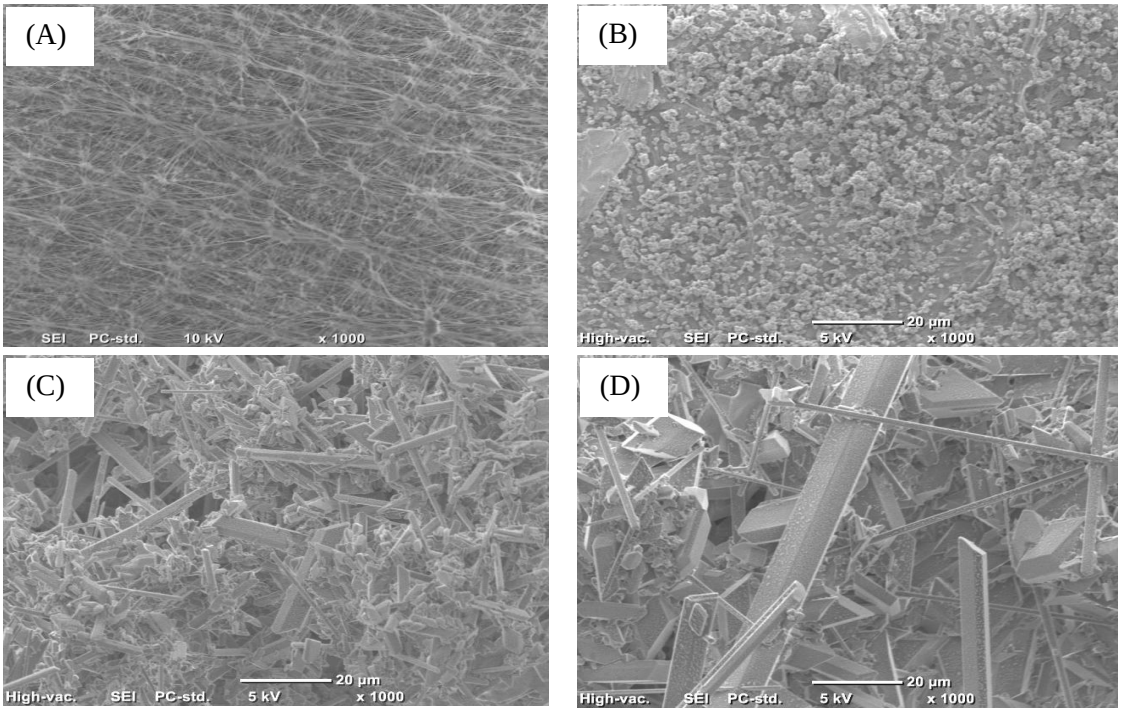

Figure 5: SEM images of (A) a virgin membrane and scaled membrane at the end of the DCMD process with pre-filtered seawater at feed temperature of (B) 40 °C, (C) 50 °C, and (D) 60 °C

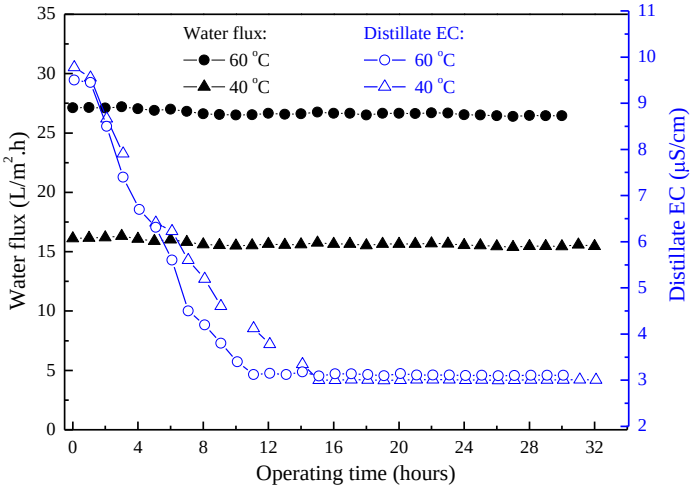

Figure 6: Water flux and distillate electrical conductivity (EC) during DCMD of seawater at a constant water recovery of 70 % with scaling mitigation techniques

The results reported here demonstrate the great viability of MD for small-scale and decentralized seawater desalination application in Vietnam. With little feed water pre-treatment (i.e. simple pre-filtration and a small dose of anti-scalant), seawater MD desalination process can produce stable water

flux of super quality. Given the water flux of 27 $L/m^2.h$ at feed temperature of 60 °C, a small DCMD system with 10 m^2 of membrane surface can produce 2,160 L of fresh water for 8 hours. More importantly, the main energy source for MD is thermal energy which can be sourced from low-grade waste heat or solar thermal energy. Vietnam has long coastline, a large number of islands, and widespread availability of solar thermal energy. Therefore, seawater MD desalination can be a technology platform for fresh water provision in remote coastal areas in Vietnam.

4. CONCLUSION

Results from this study demonstrate notable influence of increased feed salinity and membrane scaling on water flux at high water recoveries during the DCMD process of seawater. At water recoveries above 50 %, significant impacts of temperature and concentration polarization effects on water flux were observed, resulting in noticeable deviation between the experimentally measured and the calculated water flux. The formation of scale layers on the membrane surface at high water recoveries further reduced the measured water flux. Feed operating temperature exerted strong effects on water flux and scaling behavior of the process. Reducing feed temperature led to a decrease in water flux but also reduced the severity of membrane scaling. Finally, a stable DCMD process of seawater (i.e. with respect to water flux and distillate EC) at a constant water recovery of 70 % was obtained for over 24 hours by either anti-scalant addition or operating the process at low feed temperature (i.e. 40 °C). The experimental results obtained in this study demonstrate the viability of MD for seawater desalination in Vietnam.

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