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Optimising thermal efficiency of direct contact membrane distillation by brine recycling for small-scale seawater desalination

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Abstract

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Optimising thermal efficiency of direct contact membrane distillation by brine recycling for small-scale seawater desalination

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Desalination

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Abstract: A technique to optimise thermal efficiency using brine recycling during direct contact membrane distillation (DCMD) of seawater was investigated. By returning the hot brine to the feed tank, the system water recovery could be increased and the sensible heat of the hot brine was recovered to improve thermal efficiency. The results show that in the optimal water recovery range of 20 to 60% facilitated by brine recycling, the specific thermal energy consumption of the process could be reduced by more than half. It is also noteworthy that within this optimal water recovery range, the risk of membrane scaling is negligible – DCMD of seawater at a constant water recovery of 70% was achieved for over 24 hours without any scale formation on the membrane surface. In contrast, severe membrane scaling was observed when water recovery reached 80%. In addition to water recovery, other operating conditions such as feed temperature and water circulation rates could influence the process thermal efficiency. Increasing the feed temperature and reducing the circulation flow rates increased thermal efficiency. Increasing the feed temperature could also mitigate the negative effect of elevated feed concentration on the distillate flux, particularly at a high water recovery.

Keywords: direct contact membrane distillation (DCMD); seawater desalination; thermal efficiency; brine recycling; membrane scaling; water recovery.

1. Introduction

Desalination is a practical approach to augmenting fresh water supply in coastal areas [1]. Large-scale seawater desalination can be readily implemented using reverse osmosis (RO) and conventional thermal distillation [2]; however, the provision of small-scale seawater desalination for small and remote coastal communities remains a significant challenge. Indeed, RO requires intensive pre-treatment, high-pressure pumps, and duplex stainless steel piping, all of which are expensive and not practical for small-scale seawater desalination [3, 4]. In the context of small-scale seawater desalination, membrane distillation (MD) can be a favourable alternative particularly because of the potential to directly use solar thermal and low-grade heat as the primary source of energy [5, 6]. Unlike conventional thermal distillation processes, which require a large physical footprint, MD can retain most positive attributes of a typical membrane process, including modulation, compactness, and process efficiency [7, 8]. The optimal thermal energy consumption of MD can be lower than that of conventional thermal distillation [9].

MD is a hybrid separation process that involves phase-change thermal distillation and microporous hydrophobic membrane separation [7, 8, 10]. In MD desalination, the hydrophobic nature of the membrane allows for the transport of water vapour while preventing the permeation of liquid water. As a result, dissolved solutes (i.e. inorganic salts that cannot be evaporated) and suspended particles can be completely rejected by MD. In addition, unlike in RO, the driving force for mass transport in MD is the partial water vapour pressure difference across the membrane, which is mainly induced by a transmembrane temperature difference. Thus, water flux in MD is negligibly affected by the feed water salinity. In other words, MD can be used for desalinating hypersaline feed streams or to achieve high water recovery desalination [11-16]. Given the discontinuity of the liquid phase across the membrane and a small hydraulic pressure on the membrane surface, MD is less susceptible to membrane fouling compared to RO, and hence does not require extensive pre-treatment [7]. More importantly, due to the absence of high hydraulic pressure, which is required for RO, non-corrosive and inexpensive plastic materials can be used for MD's infrastructure (i.e. membrane modules, vessels, and piping), thus significantly reducing its capital costs. Furthermore, by using a microporous membrane to facilitate the transport of water vapour, MD is more compact and thus has a significantly smaller footprint compared to conventional thermal distillation. Finally, MD is often operated at feed temperature ranging from 40 to 80 °C, which coincides with the optimal range of most thermal solar collectors [17]. Given these attributes, MD is arguably the most promising candidate for portable, stand-alone, and solar driven seawater desalination applications [17-19].

In practice, the use of MD for seawater desalination is still largely restricted to pilot-scale demonstrations [7]. Technical challenges, namely intensive energy consumption and membrane pore wetting, must be overcome before seawater desalination by MD can be commercially

realised. As a phase-change separation process, MD consumes significant heating and cooling energy to perform the phase conversion. Consequently, all MD processes reported in the literature demonstrate an energy consumption of several orders of magnitude higher than that of RO [18, 20, 21]. In addition, to sustain its separation functionality, MD requires the membrane pores to be dry. In seawater applications, organic matter and scale formed on the membrane surface can alter the membrane hydrophobicity, which may lead to liquid intrusion into the pores, and, subsequently, water flux reduction and deteriorated distillate quality [22-24].

Depending on the methods applied to generate its driving force, MD can be divided into four basic configurations, including vacuum, air gap, sweeping gas, and direct contact membrane distillation. Among these configurations, direct contact membrane distillation (DCMD) has the simplest arrangement [7], and is deemed best suited for small-scale desalination applications [7, 8]. DCMD has also been the most studied configuration in the MD literature [7]. However, heat loss due to conduction through the membrane in DCMD can be significant because of its simple arrangement (i.e. the hot feed and the cold distillate are both in contact with the membrane). Thus, DCMD may have a lower thermal efficiency (i.e. higher thermal energy consumption per unit volume of distillate) compared to other MD configurations.

Several attempts have been made to reduce energy consumption and thus enhance thermal efficiency of DCMD desalination processes. As a notable example, Lin et al. [25] investigated the coupling of DCMD with an external heat exchanger to recover the latent heat accumulated in the distillate stream, thus enhancing process thermal efficiency. The authors demonstrated that if infinite membrane and heat-exchanging surface was available, a minimum specific heat consumption of DCMD (i.e. with a heat exchanger) of 0.03 MJ/L could be achieved by optimising the ratio between the feed and distillate flow rates. However, it is impractical to have infinite membrane and heat-exchanging surface; thus, in practice, brine recycling can be used to improve water recovery and thermal efficiency [25]. Brine recycling for water recovery and thermal efficiency enhancement has also been suggested by Saffarini et al. [26]. Brine recycling enhances the utilisation of the available membrane surface area. In other words, brine recycling can be used to optimise the thermal efficiency without the need of increasing membrane surface area (or module size). The cost of membrane is significant [27] and this attribute is particularly important for small-scale desalination applications. It is noteworthy that no previous studies have experimentally evaluated brine recycling in DCMD of seawater.

A major challenge for brine recycling during DCMD of seawater is to manage the negative effects of increased feed salinity associated with high water recovery on water flux, distillate quality, and membrane scaling. This study aims to elucidate the relationship between thermal efficiency, water recovery, and membrane scaling in DCMD of seawater with brine recycling. The effects of operating conditions, including water recovery, feed temperature, and water

circulation rates, on thermal efficiency of the process were systematically examined. The risk of membrane scaling at a high water recovery from actual seawater was also investigated.

2. Materials and methods

2.1. DCMD test unit

A flow diagram of the DCMD unit used in this study is shown in Fig. 1. The membrane cell, provided by Aquastill (Sittard, The Netherlands), composed of two polypropylene (PP) semi-cells. Each semi-cell had a flow channel with depth, width, and length of 0.2, 10, and 50 cm, respectively, forming an active membrane area of 500 cm². A flat-sheet, low-density polyethylene (LDPE) membrane (also provided by AquaStill) having nominal pore size of 0.3 μm, thickness of 76 μm, and porosity of 85% was installed between the two semi-cells to form the feed and distillate channels. PP spacers were used in both channels for improved flow turbulence. Two variable-speed gear pumps (Model 120/IEC71-B14, Micropump Inc., Vancouver, Washington, USA) were used to circulate the feed and distillate through the membrane cell. Two rotameters, positioned before the inlet of each channel, were used to monitor the circulation flow rates of the feed and distillate.

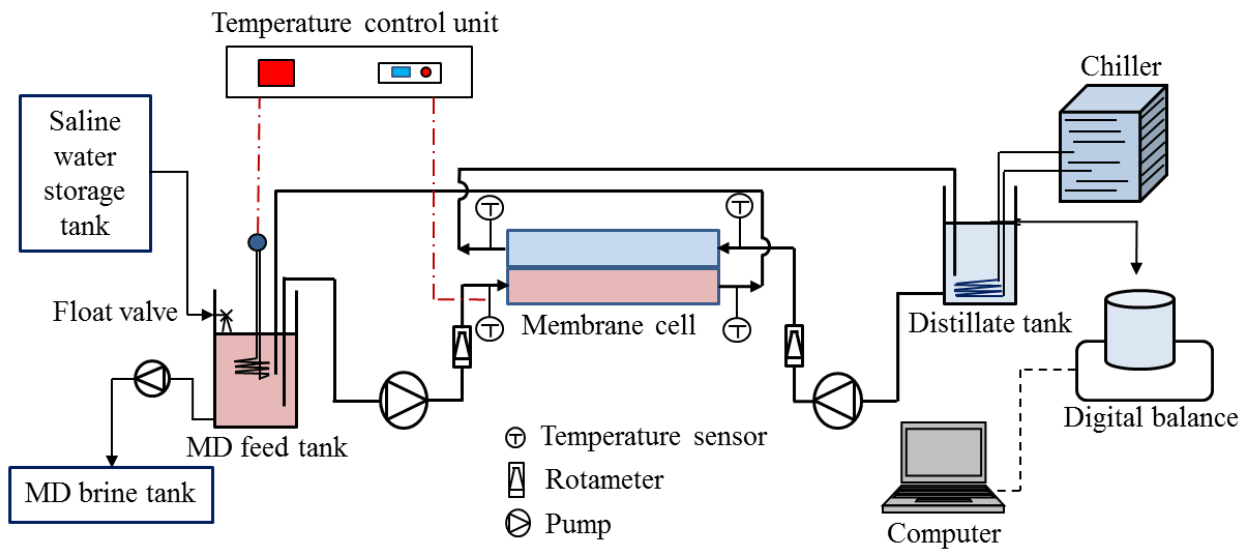


Fig. 1. Flow diagram of the DCMD system used in the study.

Feed water from a storage tank flowed into the MD feed tank by gravity via a float valve. The MD feed tank was heated using a submerged heating element connected to a temperature control unit. A temperature sensor positioned immediately before the inlet of the feed channel was used to regulate the feed water temperature. Another temperature sensor was installed at the outlet of the feed channel to monitor the feed temperature drop along the channel. A peristaltic pump (Masterflex, John Morris Scientific Pty Ltd., Australia) was used to bleed the concentrated brine

from the MD feed tank when necessary (see section 2.3). 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 tank. The temperatures of the distillate entering and leaving the cell were monitored by other two temperature sensors. 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.

2.2. Analytical methods

Electrical conductivity of the feed and distillate was measured using Orion 4-Star Plus meters (Thermo Scientific, Waltham, Massachusetts, USA). Contact angle of membrane surface before and after experiments was measured by the sessile drop technique using a Rame-Hart Goniometer (Model 250, Rame-Hart, Netcong, New Jersey, USA). Milli-Q water was used as the reference liquid for the contact angle measurements. Morphology and composition of membrane surface were examined using a low vacuum scanning electron microscope (SEM) coupled with an energy dispersive spectrometer (EDS) (JOEL JSM-6490LV, Japan). The membrane samples were air-dried and then directly used (i.e. without coating) for SEM-EDS analysis.

2.3. Experimental protocols

2.3.1. Feed solutions

Milli-Q water, synthetic 35,000 mg/L NaCl solution, and pre-filtered seawater were used as feed solutions. Seawater was collected from Wollongong beach (New South Wales, Australia) and was pre-filtered by 0.5 μm filter paper prior to all experiments. The conductivity, pH, and total dissolved solids of this pre-filtered seawater were of 52.5 ± 1.0 mS/cm, 8.35 ± 0.05 , and $37,000 \pm 2000$ mg/L, respectively. The total organic carbon (TOC) concentration of this pre-filtered seawater was less than 2 mg/L. The synthetic NaCl solution was prepared from analytical grade chemical and Milli-Q water.

2.3.2. DCMD of Milli-Q water and saline solutions

DCMD of Milli-Q water was conducted to determine the system's baseline mass transfer coefficient prior to the experiments with the saline solutions. The process was operated at constant water circulation rates (1.25 L/min) and distillate temperature (25 °C), but varied feed temperature (i.e. 35, 40, 45, 50 °C). Water flux of DCMD was measured at each feed temperature at stable conditions for three hours.

Concentrating and constant recovery operating modes were used in the DCMD experiments with saline solutions. In the concentrating mode, 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 (*Rec*) of the system in this mode was the ratio between the accumulated distillate volume and the

initial feed volume. In the constant recovery mode, concentrated brine was bled out and saline solution was allowed to flow into the MD feed tank via the float valve (Fig. 1). The bled-out flow rate is calculated as:

$$F_{brineout} = F_d \left(\frac{1}{Rec} - 1 \right) \quad (1)$$

where $F_{brineout}$ and F_d are the volumetric flow rates (m^3/s) of bled-out brine and produced distillate, respectively, and Rec is the predetermined system water recovery. In the constant recovery mode, Rec is defined as:

$$Rec = \frac{F_d}{F_{sys}} \quad (2)$$

where F_{sys} is the volumetric flow rate of saline water fed into the MD feed tank. In both concentrating and constant recovery modes, a concentration factor (CF) can be determined:

$$CF = \frac{1}{1 - Rec} \quad (3)$$

Prior to constant recovery operation, the feed was first concentrated to reach a predetermined water recovery. Then, the constant recovery mode was initiated and maintained for at least 24 hours before being terminated or switched to another water recovery set point. 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. [To ensure experimental reproducibility, a new membrane sample and 2 L of Milli-Q water as the initial distillate were used for each experiment.](#)

2.4. Mass transfer of water in DCMD

The mass transfer of water vapour across the membrane in DCMD depends on the membrane properties and operating conditions, and can be expressed as:

$$J = K_m \Delta P \quad (4)$$

where J is the water flux of the system ($L/m^2.h$); K_m is the mass transfer coefficient ($L/Pa.m^2.h$); ΔP is the water vapour pressure difference between the vapour-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, pressures, and water circulation rates. K_m can be determined using empirical correlations [7, 28] or experimentally measured [22].

The vapour pressure of pure water at the membrane surface can be calculated using the Antoine equation:

$$P^0 = \exp\left(23.1964 - \frac{3816.44}{T - 46.13}\right) \quad (5)$$

where P^0 is in Pa and T is the temperature in K. For a saline solution, the presence of salts in the solution reduces water activity and, hence, water vapour pressure. Thus, the partial vapour pressure of water at the membrane surfaces in DCMD of saline solutions (P) is calculated as [29]:

$$P = x_{water} \left(1 - 0.5x_{salt} - 10x_{salt}^2\right) P^0 \quad (6)$$

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

For DCMD of a diluted solution, x_{salt} is negligible and thus the concentration polarisation effect can be ignored. On the other hand, due to temperature polarisation, the real transmembrane temperature difference is smaller than that between the bulk feed and distillate (which can be readily measured), thus reducing the driving force for mass transfer. However, the effect of temperature polarisation can be incorporated into the mass transfer coefficient, K_m , and ΔP can be calculated using the average temperatures of the bulk feed and distillate (i.e. $\{T_{f.in} + T_{f.out}\}/2$ and $\{T_{d.in} + T_{d.out}\}/2$, respectively).

2.5. Energy consumption and thermal efficiency in DCMD

In MD, thermal energy is required to heat the saline feed solution and to cool the distillate. In this study, a chiller was used as a heat sink. However, in practice, seawater at ambient temperature can be circulated through a heat-exchanging coil for cooling. Thus, cooling energy was excluded when calculating the process thermal efficiency.

The efficiency of a thermal desalination process can be determined by the specific thermal energy consumption (*STEC*), which is the thermal energy consumed per volume unit of produced distillate [18, 30]. Based on a heat and mass balance (Fig. 2), *STEC* (in MJ/L) of DCMD without brine recycling can be calculated as:

$$STEC = \frac{\rho_{f.in} F_{f.in} C_P (T_{f.in} - T_{sys})}{F_d \times 10^6} \quad (7)$$

where $F_{f.in}$, $T_{f.in}$, and $\rho_{f.in}$ are the inlet volumetric flow rate (m^3/s), inlet temperature ($^{\circ}C$), and inlet density (kg/m^3) of the feed, respectively, T_{sys} is the temperature of the saline water in the storage tank (i.e. assumed to be constant at $25^{\circ}C$), and C_P is the specific heat capacity of solutions ($kJ/kg.^{\circ}C$). When the brine is returned to the feed tank and the system water recovery remains constant at Rec , the heat input into the system is:

$$Q_{input} = \frac{F_d \rho_{sys} C_P (T_{f.in} - T_{sys})}{Rec} + C_p (F_{f.in} \rho_{f.in} - F_d \rho_d) (T_{f.in} - T_{f.out}) \quad (8)$$

where Q_{input} is in kJ/s, ρ_{sys} and ρ_d are densities of saline water in the storage tank and the distillate (kg/m^3), and $T_{f.out}$ is the outlet temperature of the feed. Thus, the heat input of the system could be calculated by measuring the volumetric flow rates of feed and distillate, along with feed inlet and outlet temperatures. Then, *STEC* of the process in constant recovery mode can be calculated as:

$$STEC = \frac{Q_{input}}{F_d \times 10^6} \quad (9)$$

Another useful parameter, particularly when involving heat recovery, is the gained output ratio (*GOR*). *GOR* is the ratio between the heat associated with water vapour transfer and the total heat input, and is calculated as:

$$GOR = \frac{F_d \rho_d \Delta H}{Q_{input}} \quad (10)$$

where ΔH is the latent heat of evaporation of water (kJ/kg). The density, specific heat capacity, and the latent heat of evaporation of saline solutions are dependent on temperature and salinity, and their calculations are given elsewhere [31]. Unless otherwise stated, for comparison purpose, the *STEC* and *GOR* of DCMD reported in this study were calculated for the constant recovery mode.

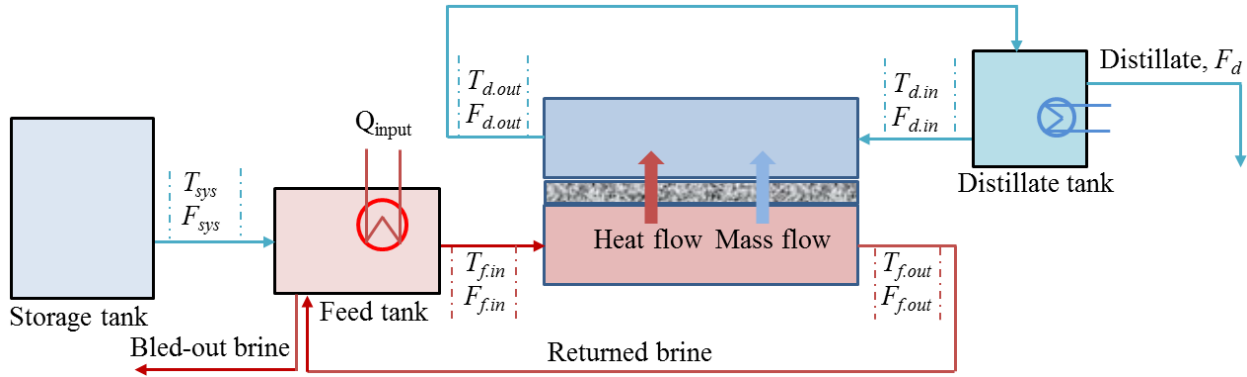


Fig. 2. Heat and mass flow in DCMD with brine recycling.

3. Results and discussions

3.1. Water flux and mass transfer coefficient in DCMD of diluted solution

As expected, water flux increased as the transmembrane temperature difference increased (Fig. 3A). When Milli-Q water was used as the feed, concentration polarisation can be ignored and the mass transfer coefficient of the system, K_m , could be obtained (i.e. $1.01 \times 10^{-3} \text{ L/Pa.m}^2.\text{h}$) based on Eqs. 4 through 6 and a linear regression between the water flux and the calculated ΔP . The linear regression coefficient (R^2 value) was 0.999.

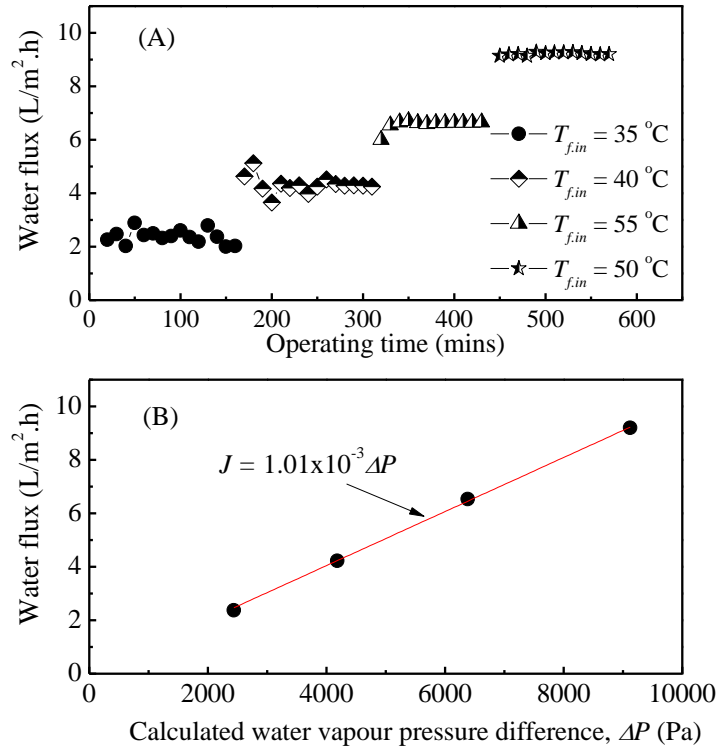


Fig. 3. (A) Experimentally measured water flux at different feed temperatures and (B) water flux as a function of the calculated water vapour pressure difference between the two sides of the membrane in DCMD with milli-Q water feed. Operating parameters: $T_{d.in} = 25 \text{ }^\circ\text{C}$, $F_{f.in} = F_{d.in} = 1.25 \text{ L/min}$.

3.2. Water flux and thermal efficiency in DCMD of saline solution with brine recycling

The obtained K_m value (section 3.1) is valid for DCMD of a diluted feed with negligible concentration polarisation. It is also useful to evaluate the impact of water recovery on water flux in DCMD of saline solutions with brine recycling. For the NaCl feed solution of 35,000 mg/L, as water recovery increases, the remaining feed becomes more concentrated because MD can offer complete salt rejection. However, the increase in water recovery up to about 40% (i.e. in DCMD at feed temperature of 40 °C) did not exert any discernible impact on water flux. The measured water flux coincided with the calculated values using the obtained K_m (Fig. 4A). As water recovery further increased, the increase in salt concentration for each unit increase in water recovery becomes more significant. As a result, the impact of recovery increase on water flux was noticeable at water recoveries above 40% (Fig. 4A). As discussed in section 3.1, the obtained K_m did not take into account concentration polarisation, which was signified at high salt concentration in the feed. Thus, the measured water flux deviated considerably from the calculated values, especially at high water recoveries. In addition, because the polarisation effects are magnified by elevating feed temperature, and hence increasing water flux [15, 32], the

deviation of the measured flux from the calculated values occurred at lower water recovery in the experiments conducted at higher feed temperature (Fig. 4A).

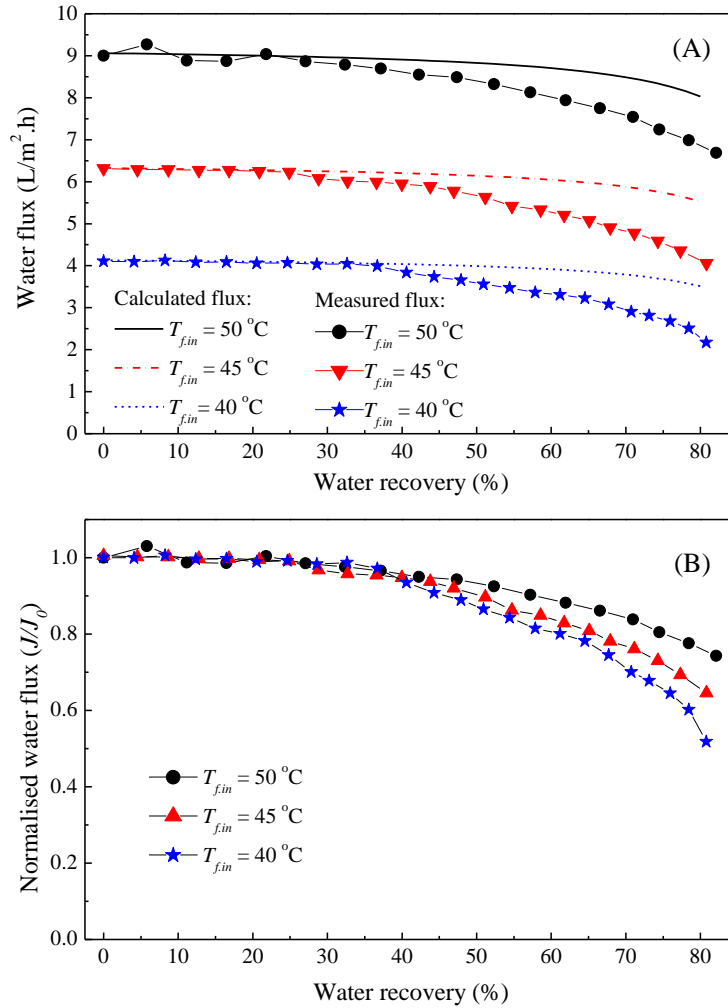


Fig. 4. Influence of feed salinity on water flux as water recovery increased in DCMD of the NaCl 35,000 mg/L solution with brine recycling at different feed temperatures: (A) calculated and experimentally measured water flux as a function of water recovery, and (B) normalised measured water flux as a function of water recovery. Operating conditions: $T_{d.in} = 25\text{ }^{\circ}\text{C}$, $F_{f.in} = F_{d.in} = 1.25\text{ L/min}$.

Operating DCMD at elevated feed temperature helped alleviate the negative effects of feed salinity on water flux at high water recoveries. At water recoveries below 40%, the normalised water fluxes at the three feed temperatures were almost identical and approximated to unity (Fig. 4B), demonstrating the independence of MD flux on feed salinity at low feed concentrations. As water recovery reached 80%, water flux declined by 20% at feed temperature of 50 °C, but it decreased by 50% when operating at feed temperature of 40 °C. This could be attributed to the increasing ratio between the driving force, which exponentially depends on feed temperature, and the feed salinity as feed temperature rises. The reported result is consistent with previous studies

by Duong et al. [30] and Winter et al. [1], emphasising the significant influence of feed salinity on the performance characteristics of the MD processes with a small driving force.

Brine recycling in DCMD could be optimised to increase thermal efficiency (Fig. 5). Increasing water recovery by returning the hot brine to the feed tank resulted in a slight decline in water flux, but a noticeable improvement in *GOR*. A significant reduction in *STEC* was also obtained by brine recycling. Without brine recycling, the *STEC* value (i.e. calculated using Eq. 7) was 16.2 MJ/L. When operating the lab-scale process at the constant recovery mode with brine recycling to obtain the optimal water recovery of 30%, a substantial reduction in *STEC*, to 6.5 MJ/L, could be achieved. It is noteworthy that this calculated *STEC* was from a simple lab-scale DCMD process without any energy recovery. A lower *STEC* value (i.e. 1.6–2.2 MJ/L) can be obtained from pilot-scale DCMD processes with energy recovery and better insulation [33, 34]. However, the inclusion of an external energy recovery system is not suitable for a small-scale system and is beyond the scope of our study.

The optimal water recovery for maximised thermal efficiency is approximately 30%; however, it is noteworthy that changes in both *GOR* and *STEC* values within the water recovery range of 20 to 60% are negligible (Fig. 5). Thus, 20 to 60% can be taken as the optimal water recovery range in DCMD desalination with brine recycling. Exceeding this optimum range, an increase in water recovery led to a small rise in *STEC* and a slight decrease in *GOR*. The decrease in thermal efficiency at water recoveries above 60% can be attributed to the increased effect of feed salinity on water flux at high water recoveries. As water recovery increased, while the required heat input did not change greatly, water flux of the system gradually decreased. Thus, thermal efficiency of the system declined as water recovery exceeded the optimum range.

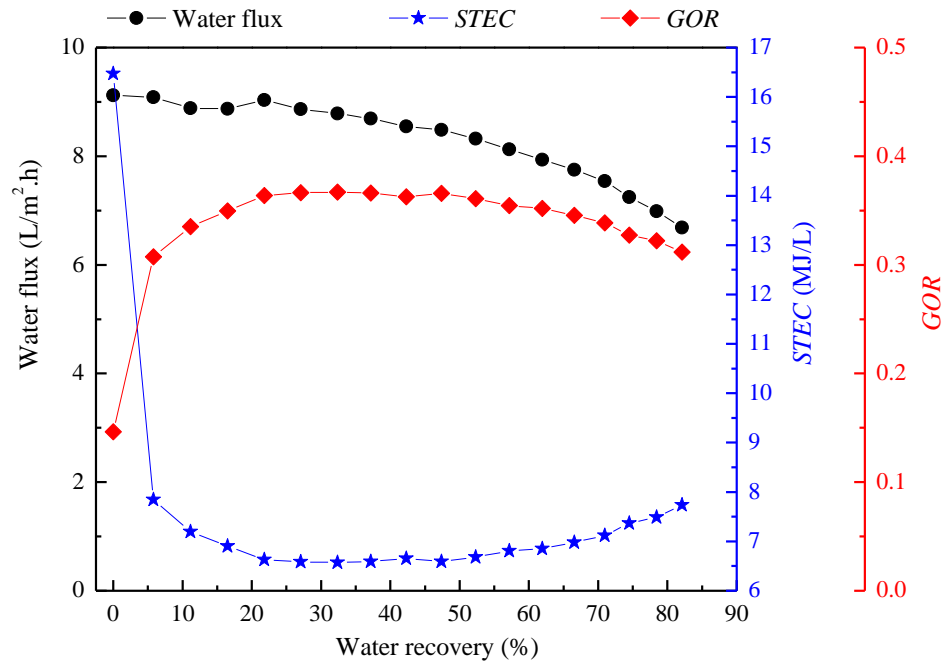


Fig. 5. Water flux, *STEC*, and *GOR* as a function of water recovery in DCMD of the 35,000 mg/L NaCl solution with brine recycling. Operating parameters: $T_{f.in} = 50\text{ }^{\circ}\text{C}$, $T_{d.in} = 25\text{ }^{\circ}\text{C}$, $F_{f.in} = F_{d.in} = 1.25\text{ L/min}$.

In addition to water recovery, the feed temperature and water circulation rates had considerable influence on thermal efficiency of the DCMD process. Increasing feed temperature was beneficial for improving *GOR* and reducing *STEC* of the system (Fig. 6A). At low water recoveries, the influence of feed temperature on *GOR* and *STEC* was unnoticeable. However, at high water recoveries, elevating feed temperature from 45 to 50 °C resulted in 30% increase in *GOR* and a reduction at the same magnitude in *STEC*. The observed increase in thermal efficiency at high water recoveries could be attributed to the mitigated effects of feed salinity at elevated feed temperature as discussed above. Reducing water circulation rates also helped increase thermal efficiency of the system. Operating the system at decreased circulation rates favoured the reduction in *STEC* and improvement in *GOR* (Fig. 6B). This observation is consistent with previous results by Summers et al. [35] and Guan et al. [36] who simulated thermal efficiency of DCMD using a heat-exchanger. It is worth noting that elevating feed temperature and reducing water circulation rates promote concentration polarisation [15, 30, 37], and thus might increase the risk of membrane scaling in DCMD of actual seawater.

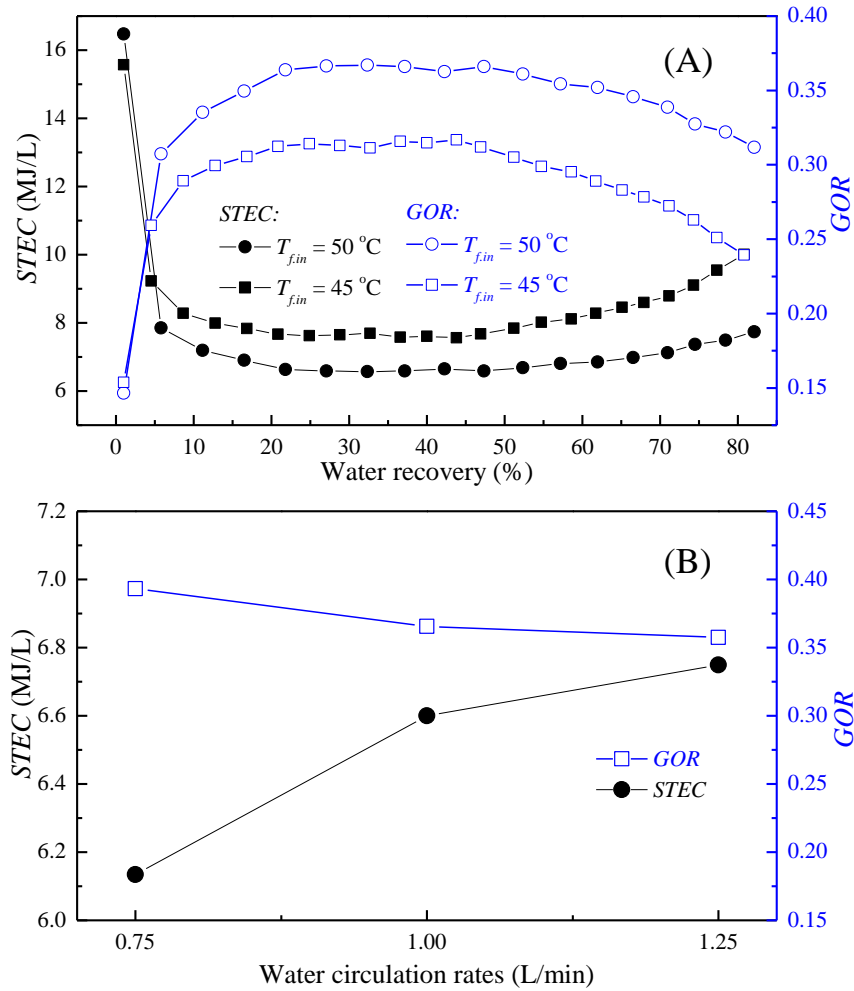


Fig. 6. Influence of operating conditions on thermal efficiency of DCMD with brine recycling. (A) *STEC* and *GOR* as a function of water recovery at feed temperature, $T_{f.in}$, of 45 and 50 °C; other operating conditions: $T_{d.in}= 25\text{ }^{\circ}\text{C}$, $F_{f.in} = F_{d.in}= 1.25\text{ L/min}$. (B) *STEC* and *GOR* at water circulation rates of 0.75, 1.00, and 1.25 L/min, and water recovery of 60%; other operating conditions: $T_{f.in}= 50\text{ }^{\circ}\text{C}$, $T_{d.in}= 25\text{ }^{\circ}\text{C}$.

3.3. Membrane scaling in DCMD with seawater

The performance of DCMD with the pre-filtered seawater operated in the concentrating mode was similar to that observed in the experiment with the NaCl feed solution (Fig. 7). At water recoveries below 70% (i.e. concentration factor, CF , below 3.3), membrane scaling caused due to the presence of sparingly soluble salts did not occur, indicated by the steady decrease in the distillate conductivity, even with increased feed concentration. Thus, there were only effects of temperature and concentration polarisations, and increased feed salinity on water flux, which were also encountered in DCMD with the NaCl solution. As a result, water flux gradually decreased when the system water recovery increased to 70%. As the system water recovery

approached higher values, water flux further decreased. However, no significant reduction in water flux was observed even when the distillate conductivity started increasing, which is an indication of scale formation and membrane pore wetting.

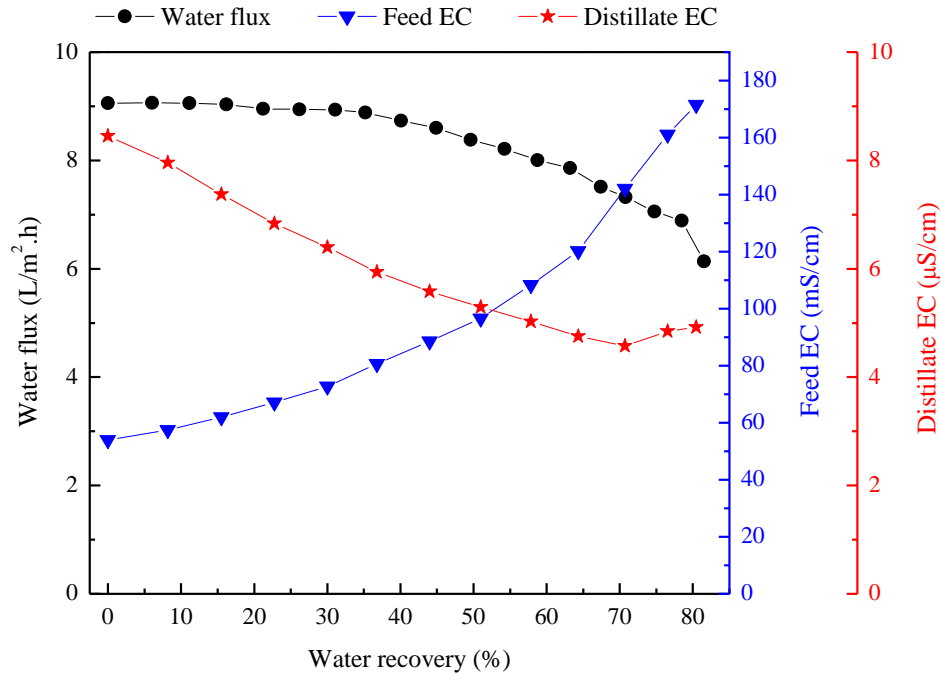


Fig. 7. Water flux, feed and distillate electrical conductivity (EC) as a function of water recovery in DCMD of pre-filtered seawater. Operating conditions: $T_{f.in} = 50$ °C, $T_{d.in} = 25$ °C, $F_{f.in} = F_{d.in} = 1.25$ L/min.

Surface analysis of the membrane after the experiment with pre-filtered seawater confirmed the deposition of scale on the membrane surface. Indeed, contact angle of the scaled membrane decreased to 20° while that of the virgin membrane was 116°. SEM imaging (Fig. 8) reveals a thick and porous layer of salt crystals on the membrane surface at the completion of the experiment. It is possible that the salt crystals did not completely clog the membrane pores, and thus did not result in a significant decrease in water flux. Nevertheless, they drastically altered the hydrophobicity of the membrane surface layer. Qualitative elemental analysis of the scale layer using EDS shows that its main compositions were carbonate and sulphate salts of calcium and magnesium. Morphology of these crystals (Fig. 8) is consistent with calcium sulphate precipitate [38].

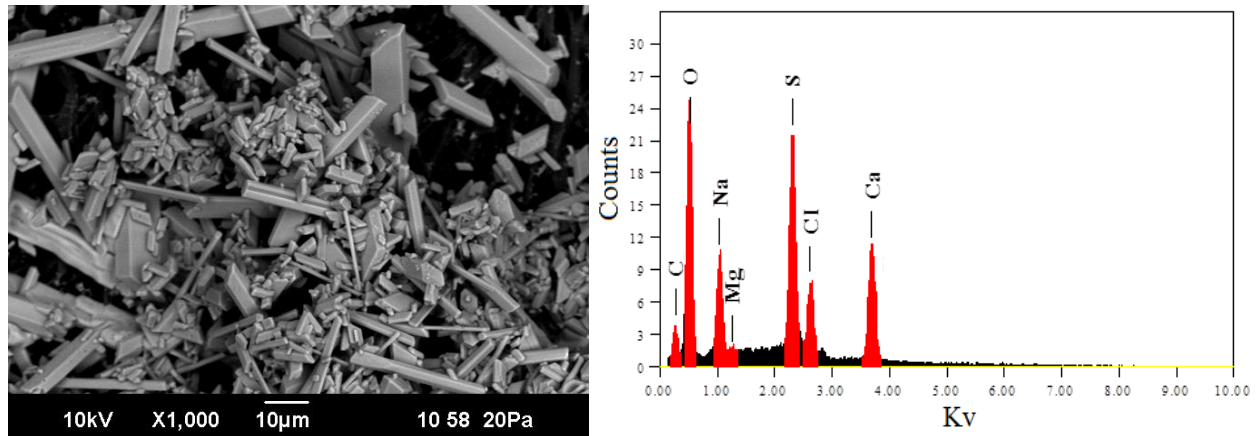


Fig. 8. SEM images and EDS spectra of the membrane after the concentrating DCMD of pre-filtered seawater up to 80% water recovery.

3.4. DCMD of seawater with brine recycling at high water recoveries

DCMD of pre-filtered seawater with brine recycling at constant water recoveries of 60 and 70% (*CF* of 2.5 and 3.3, respectively) could be achieved without any observable membrane scaling. In the concentrating mode, the increase in water recovery resulted in a gradual rise in feed salinity (represented by the feed conductivity) and hence, a steady decrease in water flux (Fig. 9). When the process was operated in the constant recovery mode at 60 and 70% for over 24 hours at each water recovery, both stable feed salinity and a constant water flux were achieved. The conductivity of the distillate steadily decreased during the first 25 hours of the operation before stabilising around 2 $\mu\text{S}/\text{cm}$ for the remainder of the test. [The observed decrease in the distillate conductivity was because of the dilution of the Milli-Q water \(conductivity of 8.5 \$\mu\text{S}/\text{cm}\$ \) which was initially used as the condensing liquid.](#) The obtained constant water flux, along with superior distillate quality, indicates that membrane scaling did not occur throughout the operation. The analysis of membrane surface at the completion of the operation also supports the absence of membrane scaling. Contact angle of the membrane decreased slightly to 100°. No evidence of scaling could be seen from the SEM image of the membrane after DCMD experiment at 70% water recovery (Fig. 10). Traces of sodium, chloride and silica could be observed from the EDS spectra; however, the intensity of these peaks is negligible (Fig. 10A) compared to the EDS spectra of the virgin membrane (Fig. 10B).

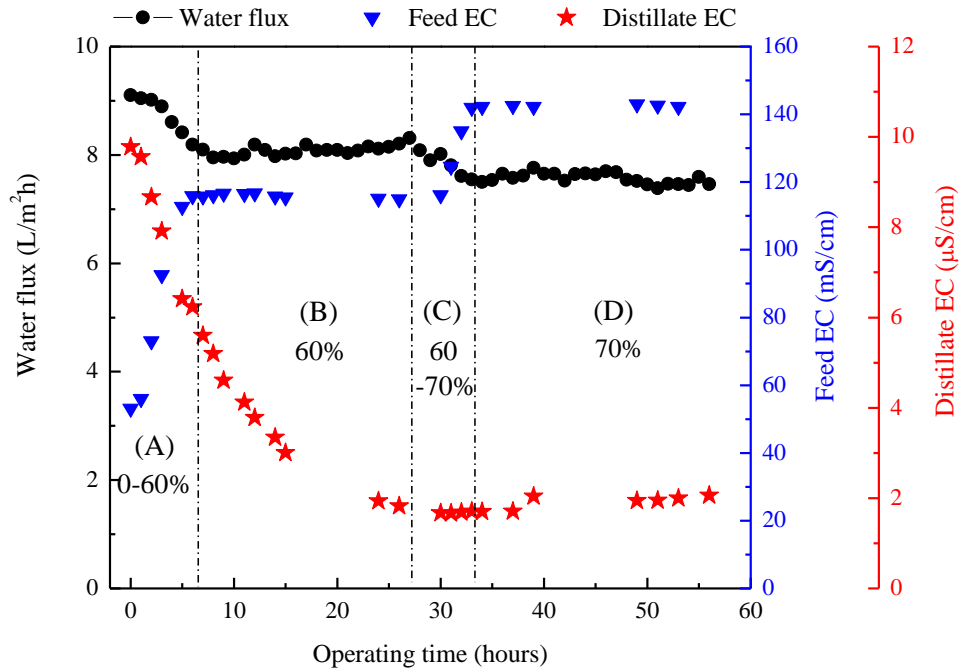


Fig. 9. Water flux, feed and distillate electrical conductivity as a function of operating time in DCMD of pre-filtered seawater with brine recycling at different operation modes: (A) concentrating operation with increased water recovery from 0 to 60%, (B) operation at constant water recovery of 60%, (C) concentrating operation with increased water recovery from 60 to 70%, and (D) operation at constant water recovery of 70%. Operating conditions: $T_{f.in} = 50\text{ }^{\circ}\text{C}$, $T_{d.in} = 25\text{ }^{\circ}\text{C}$, $F_{f.in} = F_{d.in} = 1.25\text{ L/min}$.

There appears to be a trade-off between increasing water recovery and improving performance of the system in DCMD of seawater at high water recoveries. By increasing water recovery from 60 to 70%, the volume of the discharged MD brine can be reduced by 25%. However, this increase in water recovery also resulted in a small reduction in water flux (i.e. from 8 to 7 L/m².h), and a slight decrease in the *GOR* value from 0.35 to 0.33 (corresponding to an increase in the *STEC* value from 6.8 to 7.1 MJ/L). Moreover, excessive water recovery can lead to the formation of scale on the membrane surface as discussed above.

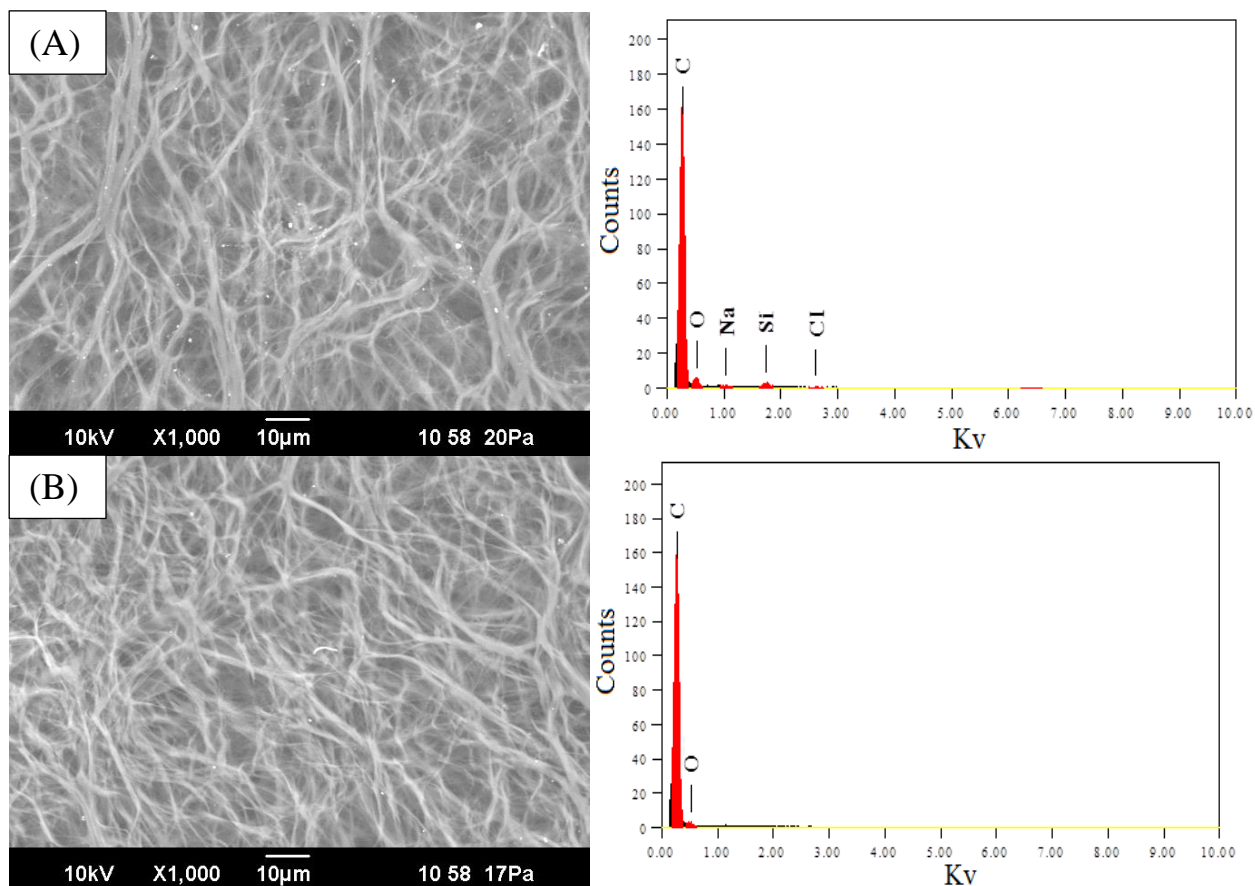


Fig. 10. SEM images and EDS spectra of (A) the membrane after DCMD of pre-filtered seawater at 70% water recovery, and (B) the virgin membrane.

Thermal efficiency of seawater DCMD with brine recycling when operating at the constant water recovery of 60% was more than doubled compared to that of a single-pass process (i.e. *STEC* decreased from 16.2 to 6.8 MJ/L, and *GOR* increased from 0.15 to 0.35). Nevertheless, the achieved thermal efficiency of the DCMD test unit is still lower compared to those reported in the MD literature [18, 30]. It is important to note that the recovery of latent heat from the distillate stream back to the feed stream by using a heat exchanger was not implemented in this study. The thermal efficiency of the DCMD process coupled with the heat exchanger can be improved significantly [25, 33, 36], but will also increase the system complexity, rendering it unsuitable for small-scale operation.

4. Conclusions

Over two-fold reduction in the specific thermal energy consumption of DCMD of seawater could be achieved by brine recycling. The experimental results reveal an optimal water recovery range of 20 to 60% with respect to thermal efficiency. A high water recovery beyond this optimal range led to an increase in the risk of membrane scaling. Indeed, severe membrane scaling was

observed at 80% water recovery. On the other hand, DCMD of seawater at water recovery of up to 70% was operated for over 24 hours without any observable membrane scaling. Results reported here suggest that the risk of membrane scaling within the water recovery range for an optimal thermal efficiency is negligible. In addition to water recovery, feed temperature and water circulation rates had influence on the process thermal efficiency. Elevating feed temperature and reducing circulation rates increased thermal efficiency. Increasing feed temperature also helped reduce the negative effect of increased feed salinity on water flux at a high water recovery.

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References

- [1] D. Winter, J. Koschikowski, and M. Wieghaus, Desalination using membrane distillation: Experimental studies on full scale spiral wound modules, *J. Membr. Sci.* 375 (2011) 104-112.
- [2] M. Elimelech and W.A. Phillip, The Future of Seawater Desalination: Energy, Technology, and the Environment, *Science* 333 (2011) 712-717.
- [3] C. Fritzmann, J. Löwenberg, T. Wintgens, and T. Melin, State-of-the-art of reverse osmosis desalination, *Desalination* 216 (2007) 1-76.
- [4] C. Charcosset, A review of membrane processes and renewable energies for desalination, *Desalination* 245 (2009) 214-231.
- [5] R. Schwantes, A. Cipollina, F. Gross, J. Koschikowski, D. Pfeifle, M. Rolletschek, and V. Subiela, Membrane distillation: Solar and waste heat driven demonstration plants for desalination, *Desalination* 323 (2013) 93-106.
- [6] M. Khayet, Solar desalination by membrane distillation: Dispersion in energy consumption analysis and water production costs (a review), *Desalination* 308 (2013) 89-101.
- [7] E. Drioli, A. Ali, and F. Macedonio, Membrane distillation: Recent developments and perspectives, *Desalination* 356 (2015) 56-84.
- [8] A. Alkhudhiri, N. Darwish, and N. Hilal, Membrane distillation: A comprehensive review, *Desalination* 287 (2012) 2-18.
- [9] G.W. Meindersma, C.M. Guijt, and A.B. de Haan, Desalination and water recycling by air gap membrane distillation, *Desalination* 187 (2006) 291-301.
- [10] L.M. Camacho, L. Dumée, Z. Jianhua, L. Jun-de, M. Duke, J. Gomez, and S. Gray, Advances in membrane distillation for water desalination and purification applications, *Water* (20734441) 5 (2013) 94-196.

- [11] A. Figoli, S. Simone, A. Criscuoli, S.A. Al-Jilil, F.S. Al Shabouna, H.S. Al-Romaih, E. Di Nicolò, O.A. Al-Harbi, and E. Drioli, Hollow fibers for seawater desalination from blends of PVDF with different molecular weights: Morphology, properties and VMD performance, *Polymer* 55 (2014) 1296-1306.
- [12] J J.-P. Mericq, S. Laborie, and C. Cabassud, Vacuum membrane distillation of seawater reverse osmosis brines, *Water Res.* 44 (2010) 5260-5273.
- [13] C. Yang, X.-M. Li, J. Gilron, D.-f. Kong, Y. Yin, Y. Oren, C. Linder, and T. He, CF₄ plasma-modified superhydrophobic PVDF membranes for direct contact membrane distillation, *J. Membr. Sci.* 456 (2014) 155-161.
- [14] G. Chen, Y. Lu, W.B. Krantz, R. Wang, and A.G. Fane, Optimization of operating conditions for a continuous membrane distillation crystallization process with zero salty water discharge, *J. Membr. Sci.* 450 (2014) 1-11.
- [15] K.L. Hickenbottom and T.Y. Cath, Sustainable operation of membrane distillation for enhancement of mineral recovery from hypersaline solutions, *J. Membr. Sci.* 454 (2014) 426-435.
- [16] U.K. Kesieme, N. Milne, H. Aral, C.Y. Cheng, and M. Duke, Economic analysis of desalination technologies in the context of carbon pricing, and opportunities for membrane distillation, *Desalination* 323 (2013) 66-74.
- [17] J. Koschikowski, M. Wiegand, and M. Rommel, Solar thermal-driven desalination plants based on membrane distillation, *Desalination* 156 (2003) 295-304.
- [18] G. Zaragoza, A. Ruiz-Aguirre, and E. Guillén-Burrieza, Efficiency in the use of solar thermal energy of small membrane desalination systems for decentralized water production, *Appl. Energy* 130 (2014) 491-499.
- [19] A. Chafidz, S. Al-Zahrani, M.N. Al-Otaibi, C.F. Hoong, T.F. Lai, and M. Prabu, Portable and integrated solar-driven desalination system using membrane distillation for arid remote areas in Saudi Arabia, *Desalination* 345 (2014) 36-49.
- [20] A. Al-Karaghoul and L.L. Kazmerski, Energy consumption and water production cost of conventional and renewable-energy-powered desalination processes, *Renew. Sust. Energ. Rev.* 24 (2013) 343-356.
- [21] W.G. Shim, K. He, S. Gray, and I.S. Moon, Solar energy assisted direct contact membrane distillation (DCMD) process for seawater desalination, *Sep. Purif. Technol.* 143 (2015) 94-104.
- [22] L.D. Nghiem, F. Hildinger, F.I. Hai, and T. Cath, Treatment of saline aqueous solutions using direct contact membrane distillation, *Desalin. Water Treat.* 32 (2011) 234-241.
- [23] J. Ge, Y. Peng, Z. Li, P. Chen, and S. Wang, Membrane fouling and wetting in a DCMD process for RO brine concentration, *Desalination* 344 (2014) 97-107.
- [24] D.M. Warsinger, J. Swaminathan, E. Guillen-Burrieza, H.A. Arafat, and J.H. Lienhard V, Scaling and fouling in membrane distillation for desalination applications: A review, *Desalination* 356 (2014) 294-313.
- [25] S. Lin, N.Y. Yip, and M. Elimelech, Direct contact membrane distillation with heat recovery: Thermodynamic insights from module scale modeling, *J. Membr. Sci.* 453 (2014) 498-515.

- [26] R.B. Saffarini, E.K. Summers, H.A. Arafat, and J.H. Lienhard V, Technical evaluation of stand-alone solar powered membrane distillation systems, *Desalination* 286 (2012) 332-341.
- [27] S. Al-Obaidani, E. Curcio, F. Macedonio, G. Di Profio, H. Al-Hinai, and E. Drioli, Potential of membrane distillation in seawater desalination: Thermal efficiency, sensitivity study and cost estimation, *J. Membr. Sci.* 323 (2008) 85-98.
- [28] J. Phattaranawik, R. Jiratananon, and A.G. Fane, Heat transport and membrane distillation coefficients in direct contact membrane distillation, *J. Membr. Sci.* 212 (2003) 177-193.
- [29] K.W. Lawson and D.R. Lloyd, Membrane distillation, *J. Membr. Sci.* 124 (1997) 1-25.
- [30] H.C. Duong, A.R. Chivas, B. Nelemans, M. Duke, S. Gray, T.Y. Cath, and L.D. Nghiem, Treatment of RO brine from CSG produced water by spiral-wound air gap membrane distillation – A pilot study, *Desalination* 366 (2015) 121-129.
- [31] A.S. Alsaadi, N. Ghaffour, J.D. Li, S. Gray, L. Francis, H. Maab, and G. L. Amy, Modeling of air-gap membrane distillation process: A theoretical and experimental study, *J. Membr. Sci.* 445 (2013) 53-65.
- [32] H.C. Duong, M. Duke, S. Gray, T.Y. Cath, and L.D. Nghiem, Scaling control during membrane distillation of coal seam gas reverse osmosis brine, *J. Membr. Sci.* (Submitted, 2015).
- [33] Y.-D. Kim, K. Thu, N. Ghaffour, and K. Choon Ng, Performance investigation of a solar-assisted direct contact membrane distillation system, *J. Membr. Sci.* 427 (2013) 345-364.
- [34] G. Zakrzewska-Trznadel, M. Harasimowicz, and A.G. Chmielewski, Concentration of radioactive components in liquid low-level radioactive waste by membrane distillation, *J. Membr. Sci.* 163 (1999) 257-264.
- [35] E.K. Summers, H.A. Arafat, and J.H. Lienhard V, Energy efficiency comparison of single-stage membrane distillation (MD) desalination cycles in different configurations, *Desalination* 290 (2012) 54-66.
- [36] G. Guan, X. Yang, R. Wang, and A.G. Fane, Evaluation of heat utilization in membrane distillation desalination system integrated with heat recovery, *Desalination* 366 (2015) 80-93.
- [37] M. Gryta, Alkaline scaling in the membrane distillation process, *Desalination* 228 (2008) 128-134.
- [38] L.D. Nghiem and T. Cath, A scaling mitigation approach during direct contact membrane distillation, *Sep. Purif. Technol.* 80 (2011) 315-322.