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Heat transfer intensification and scaling mitigation in bubbling-enhanced membrane distillation for brine concentration

Guizi Chen^{1,2}, Xing Yang³, Yinghong Lu^{1,2}, Rong Wang^{*, 1,2}, Anthony G. Fane^{1,2}

1. School of Civil and Environmental Engineering, Nanyang Technological University,
Singapore 639798

2. Singapore Membrane Technology Centre, Nanyang Environment and Water Research
Institute, Nanyang Technological University, 1 Cleantech Loop, Singapore 637141

3. Institute for Sustainability and Innovation, College of Engineering and Science,
Victoria University, PO Box 14428, Melbourne, Victoria 8001, Australia

*Corresponding author at: School of Civil and Environmental Engineering,

Nanyang Technological University, Singapore 639798

Singapore. Tel.: +65 6790 5327; fax: +65 6791 0676.

E-mail address: rwang@ntu.edu.sg (R. Wang).

Abstract

Membrane distillation (MD) technology is being extensively studied to address operational challenges such as undesired thermal efficiency and scaling phenomenon in recovering valuable solutes and minimizing brine disposal. This study has explored the working mechanisms of utilizing gas-liquid two-phase flow to enhance heat transfer and mitigate scaling formation in MD concentration process, based on the quantification of heat-transfer coefficients and local scaling-resistance associated with bubble size properties.

With the aid of direct observation and statistical analysis on the bubble characteristics in a specially-designed direct contact MD (DCMD) module, it was found that the bubbles with small mean bubble size and narrow size distribution were preferred for creating even flow distribution, intensifying mixing and enhancing surface shear rate. Compared to non-bubbling DCMD, the heat-transfer coefficient and temperature polarization coefficient (*TPC*) reached up to 2.30- and 2.13-fold, respectively, at an optimal gas flowrate of 0.2 L min⁻¹. With the theoretical expressions for local scaling resistance derived based on the resistance-in-series model, the relative permeation flux ($J_{w/o|t=tI} / J_{w/o|t=0}$) in non-bubbling MD was quantified and found to rapidly decline by 65% as the concentration process progressed, consistent with the increasing trend of the ratio of local scaling resistance to the overall resistance (r_{fl} / r_{ov}). Fortunately, the introduction of gas bubbles has shown benefits for

supersaturation brine concentrating MD process - remarkably decreased the local-scaling resistance due to bubble-intensified shear stress and enhanced hydrodynamics. Also, the total water removal for the brine concentration process was significantly improved by 131% and the discharged brine volume was reduced accordingly at appropriately selected gas flow rates. Nevertheless, at inappropriately high gas flowrates, high energy consumption and potential fiber breakage should be avoided.

Keywords: membrane distillation; gas bubbling; heat-transfer resistance; scaling resistance; desalination

1. Introduction

Membrane distillation (MD) is well recognized as a potential alternative technology for desalination due to the benefits of being operated at atmospheric pressure and moderate temperature conditions, high salt rejection rate and ability to incorporate free low-grade heat [1, 2]. As a coupled mass and heat transfer process, MD is driven by vapor pressure difference (caused by temperature gradient) across the membrane [3, 4]. Hence, compared to the pressurized desalination processes (*e.g.*, reverse osmosis (RO)), the low sensitivity to feed salinity makes MD an attractive technology for maximizing brine recovery and even zero discharge [2, 3, 5, 6].

However, technical challenges that impede the industrialization of MD brine processing remain as: membrane development and polarization effects as well as fouling/scaling issues. In recent years, there is an emerging interest on novel MD membrane material development [7-10] and heat/mass-transfer analysis [11-14] associated with flow studies in membrane modules [15-18] for general desalination applications. However, only a handful of papers are available in the open literature for high concentration MD systems, which are subject to surface scaling deposition and major flux decline [2, 5, 6, 12, 19, 20]. For instance, Chan et.al [2] observed a dramatic flux decline in the MD crystallization (MDC) brine concentration process due to crystal deposition and scaling formation on the surface of

flat sheet membranes, when the feed concentration reached a critical level of supersaturation. Later on, other researchers [21] found that the flux declined rapidly at higher feed temperatures (*e.g.*, 70 °C), which was attributed to membrane scaling and wetting accelerated by salt saturation within the boundary layer in hollow fiber-based MDC module. A recent MD study [22] proposed that an early prediction of the scaling layer formation is possible through the crystallization kinetics on the membrane surface in hollow fiber modules for high concentration MD. Nevertheless, thus far only few attempts have been made to explore the intensification methods for improving thermal efficiency and mitigating scaling formation in MD brine processing [23, 24], not to mention a lack of development on scaling control strategies associated with quantitative resistance analysis of the scaling layer.

As one of the most effective and simple anti-scaling techniques, the concept of gas-liquid two-phase flow was first introduced to the field of membrane separation in the 1990s [25] and has been widely used in membrane bioreactors (MBRs), in which the air sparging serves double purposes of providing aeration and causing two-phase flow to enhance flux and control fouling [26-32]. The main working principles are owed to the bubble-induced liquid/fiber movement (secondary flows) intensified surface shear, and hence displacement of concentration polarization layers. Prior studies on MBRs and low pressure membrane systems showed that it is essential to characterize the performance of the bubbling system and

distinguish the contribution from bubbles of different sizes [33, 34]. Yet, gas bubbling has only been recently applied in the MD process for improving heat transfer and alleviating fouling. A recent MD study has examined the effectiveness of intermittent gas bubbling on the concentration process of Chinese tea extract in terms of foulant removal rate and fouling resistance analysis [23]; Other researchers [24] reported that a bubbling-assisted MD system achieved up to 72% flux enhancement and effectively postponed the occurrence of critical point of flux decline caused by the supersaturation of salt solution. However, a comprehensive evaluation is still to be carried out for characterizing the performance of two-phase flow MD system and distinguishing the intensification effect from bubbles of different properties (size & size distribution, etc).

As an extension of the previous bubbling studies, this paper aims to explore the working mechanisms of gas bubbling on heat transfer intensification and scaling alleviation for MD brine processing. With the aid of direct observation on the flowing bubbles in the MD module, the bubble characteristics (*i.e.*, bubble size and bubble size distribution) are quantified through the mathematical function **GaussAmp** (*i.e.*, Amplitude version of Gaussian peak function). An in-depth theoretical analysis is conducted to further demonstrate the benefits of utilizing gas bubbling for MD heat-transfer intensification, via the calculation of heat-transfer and temperature-polarization coefficients. Also, the effectiveness of scaling

alleviation in two-phase flow MD with supersaturated feed solution is evaluated through the local scaling resistance, which is quantified based on the resistance-in-series theory. The practicability to operate the bubbling-assisted MD process beyond supersaturation is also investigated.

2. Theory

2.1 Assessment of heat transfer process intensification by bubbling

2.1.1 Heat transfer mechanisms in DCMD

Theoretically, the permeation flux of DCMD J could be calculated in term of the transmembrane temperature difference, as:

$$J = C \frac{dP}{dT} \big|_{T_m} (T_{fm} - T_{pm}) \quad (1)$$

where C is the overall mass transfer coefficient [14], $\text{kg m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$. T_m is the membrane temperature, K. T_{fm} and T_{pm} are the membrane surface temperature on the feed and the permeate sides, K, respectively. By assuming that the temperature polarization effect is similar on both sides of the membrane, T_m can be estimated by $(T_f + T_p)/2$. The above equation is accurate with $(T_{fm} - T_{pm})$ less than 10 K and pure water used as feed. In this case, the Clausius-Clapeyron equation is applicable to determine the vapor pressure gradient dP/dT across the membrane [15],

$$\frac{dP}{dT} \big|_{T_m} = \frac{P\gamma M}{RT^2} \big|_{T_m} \quad (2)$$

where γ is the latent heat-of-vaporization, J kg^{-1} , M is the molecular weight of water, g mol^{-1} , R is the gas constant (8.314 J K^{-1}) and P is obtained from the Antoine equation [35].

The overall heat-transfer flux in MD, q , consists of the conductive heat flux q_c across the membrane and the latent heat transfer q_v accompanying the vapor flux J [19]:

$$q = q_c + q_v = \left(\frac{k_m}{\delta_m}\right)(T_{fm} - T_{pm}) + \gamma C \frac{dP}{dT} \Big|_{T_m} (T_{fm} - T_{pm}) = H(T_{fm} - T_{pm}) \quad (3)$$

where H is the overall heat-transfer coefficient based on the transmembrane temperature difference, δ_m is the wall thickness of the membrane, and k_m is the overall thermal conductivity of the membrane, $\text{W m}^{-1} \text{ K}^{-1}$. The k_m / δ_m value of the PVDF fiber used in this study is determined as $274 \text{ W m}^{-2} \text{ K}^{-1}$ [36]. The overall heat-transfer flux can be re-expressed as:

$$q = h_f (T_f - T_{fm}) = h_p (T_{pm} - T_p) \quad (4)$$

where h_f and h_p are the local heat-transfer coefficients for the hot feed and the cold permeate sides, respectively, J K^{-1} . Defined as the fraction of effective driving force $(T_{fm} - T_{pm})$ over the total thermal input $(T_f - T_p)$, the temperature polarization can be quantified via the combination of Eqs. (3) and (4), as:

$$(T_{fm} - T_{pm}) = \frac{T_f - T_p}{1 + H / h_f + H / h_p} = TPC \times (T_f - T_p) \quad (5)$$

Therefore, TPC can be determined through the calculation of various heat transfer coefficients. Combing Eqs. (1) with (5) for dilute solutions

$$J = C \frac{dP}{dT} (T_f - T_p) / (1 + H / h) \quad (6)$$

where h is termed as the overall film heat transfer coefficient $1/(1/h_f + 1/h_p)$. Substituting the definition of H ($H = C\gamma dP/dT + k_m/\delta_m$ from Eq. (3)) into Eq. (6) and rearranging gives:

$$\frac{\Delta T}{J\gamma} = \frac{1}{dP/dT} \frac{1}{C\gamma} \left(1 + \frac{k_m/\delta_m}{h}\right) + \frac{1}{h} \quad (7)$$

Therefore, with the measurable quantities k_m/δ_m , $\Delta T = T_f - T_p$ and J as well as the predetermined points of dP/dT from Eq. (2), the unknown parameters h and C can be calculated from the intercept and the slope by plotting $\Delta T/J\gamma$ against $1/(dP/dT)$ [14].

Hence, the temperature polarization coefficient (TPC) and ratio of conductive heat loss q_c/q can be quantified.

According to the above mechanism of heat transfer analysis in DCMD, the comparison of the overall film heat transfer coefficient h , TPC and conductive heat loss q_c/q with and without gas bubbling can be determined to indicate the intensification effect on heat and mass transfer in two-phase flow MD. Also, the MD performance improvement can be evaluated by the enhancement factor Φ of permeation flux:

$$\Phi = J_w / J_{w/o} \quad (8)$$

where the subscripts, w and w/o indicate a DCMD system with and without gas bubbling, respectively.

2.2 Analysis of scaling resistance in DCMD with and without bubbling

2.2.1 Resistance-in-series heat transfer model for general DCMD with scaling

In DCMD, by introducing the equivalent overall mass transfer resistance, r_{ov} , the transmembrane flux J can be re-expressed as [37]:

$$J = \frac{P_f - P_p}{r_{ov}} \quad (9)$$

where P represents the vapor pressure of the bulk fluid, Pa; f and p denote feed and permeate, respectively. The vapor pressure difference ($P_f - P_p$) is the equivalent overall driving force for mass transfer in DCMD, corresponding to the bulk temperature difference ($T_f - T_p$).

In the high concentration brine treatment process using batch-mode DCMD, scalant components such as NaCl tend to nucleate from the liquid phase due to super-saturation of the feed solution. As a result, the crystal particles would deposit on the membrane surface and form a scaling layer because of the membrane-solute interaction, which may greatly reduce the local driving force (transmembrane vapor pressure difference) and impose an additional heat-transfer resistance in MD [2]. Nevertheless, compared to pressurized membrane processes [4], the scaling layer in MD composed by NaCl crystals is much thinner and hence negligible to account for the hydrodynamic resistance of the bulks. Based on the temperature profiles across the entire domain in DCMD with scaling formation, as illustrated in Fig. 1, this overall driving force can be divided into four parts [4]:

$$P_f - P_p = (P_f - P_{sl}) + (P_{sl} - P_{fm}) + (P_{fm} - P_{pm}) + (P_{pm} - P_p) \quad (10)$$

Also, the overall heat-transfer resistance r_{ov} consists of four steps in series: (1) resistance due to polarization effect on the feed side, r_f ; (2) resistance of the scaling layer, r_{sl} ; (3) resistance of the membrane, r_m ; (4) resistance due to polarization effect on the permeate side, r_p . Hence, the r_{ov} can be expressed as:

$$r_{ov} = r_f + r_{sl} + r_m + r_p \quad (11)$$

in which r_f , r_{sl} , r_m and r_p represent the respective local vapor pressure drop per unit flux generated ($\text{Pa m}^2 \text{ h kg}^{-1}$) associated with the decrease of temperature due to the heat transferred across step 1 to 4.

2.2.2 Quantification of scaling-layer resistance with and without bubbling

In high concentration DCMD with continuous scalant nucleation and deposition adjacent to the membrane surface, the scaling layer thickens along the operation time. At the beginning of NaCl scaling formation, *i.e.*, time $t = 0$ when $r_{sl} = 0$, based on Eq. (9) the initial trans-membrane flux can be expressed as:

$$J_{w/o|t=0} = \frac{P_f - P_p}{r_{fwo} + r_m + r_p} \quad (12)$$

Hence, the overall resistance of non-bubbling DCMD is written as:

$$r_{fwo} + r_m + r_p = \frac{P_f - P_p}{J_{w/o|t=0}} \quad (13)$$

With an increasing number of crystals depositing on the membrane surface at $t = t_I$, when $r_{sl} \neq 0$, the permeation flux can then be calculated as:

$$J_{w/o|t=t_I} = \frac{P_f - P_p}{r_{f_{w/o}} + r_m + r_p + r_{sl_{w/o}}} \quad (14)$$

By combining Eqs. (12), (13) and (14) and rearranging, the scaling-layer resistance of the non-bubbling DCMD system $r_{sl_{w/o}}$ at $t = t_I$ can be quantified as:

$$r_{sl_{w/o}} = \left(\frac{J_{w/o|t=0}}{J_{w/o|t=t_I}} - 1 \right) \times \frac{(P_f - P_p)}{J_{w/o|t=0}} \quad (15)$$

With the introduction of gas bubbles in DCMD, only the hydrodynamics (boundary layer) at the feed side would be affected while the rest of the local resistances remain unchanged. Thus, the trans-membrane flux of a bubbling-assisted DCMD system $J_{w|t=0}$ and $J_{w|t=t_I}$, at $t=0$ and $t=t_I$, respectively, are expressed as:

$$J_{w|t=0} = \frac{P_f - P_p}{r_{f_w} + r_m + r_p} \quad (16)$$

$$J_{w|t=t_I} = \frac{P_f - P_p}{r_{f_w} + r_m + r_p + r_{sl_{w}}} \quad (17)$$

Similar to the derivation of Eq. (15), the overall resistance of a bubbling-assisted DCMD is written as:

$$r_{sl_{w}} = \left(\frac{J_{w|t=0}}{J_{w|t=t_I}} - 1 \right) \times \frac{(P_f - P_p)}{J_{w|t=0}} \quad (18)$$

In a word, the scaling-layer resistance in DCMD with and without bubbling can be quantified as a function of time t through Eqs. (15) and (18), respectively, where the permeation fluxes $J_{w|t=0}$, $J_{w|t=t_1}$, $J_{w/o|t=0}$ and $J_{w/o|t=t_1}$ can be obtained experimentally and the corresponding vapor pressure P_f & P_p can be calculated based on known temperature conditions (T_f & T_p) using the Antoine equation [35].

2.3 Bubble characteristics in two-phase flow

With the gas stream continuously introduced into the membrane module, bubbles of various shapes and sizes are formed depending on the gas flow rate, air sparger dimension and fluid viscosity. The number of bubbles in a sufficiently small size domain, namely frequency, could quantify bubble characteristics under certain bubbling conditions. A built-in GaussAmp mathematical function was applied to the changing curves of frequency with the bubble size (bubble size distribution) in Origin software [38]:

$$y = y_0 + y_1 e^{\left(-\frac{(x-x_c)^2}{2x_w^2}\right)} \quad (19)$$

where y is the bubbling frequency for a specific size range, x is the bubble size, mm, y_0 , y_1 , x_c and x_w are constants. Based on Eq. (19), the bubble size distribution curve of a two-phase flow could be obtained accordingly, including key parameters such as the mean bubble size, x_c , and variance of size distribution, x_w^2 . Thus, the module performance can be associated with the bubble characteristics under various bubbling conditions.

3. Experimental

3.1 *Material properties, module fabrication and air sparging*

In the current study a polyvinylidene fluoride (PVDF) hollow fiber membrane specifically developed for MD applications was used to fabricate lab-scale membrane modules. Relevant membrane properties and module specifications are listed in Table 1. To introduce gas bubbles into the MD system, specially-designed modules with air sparger, as illustrated in Fig. 2a, were fabricated by potting the PVDF hollow fiber membranes into an 85 mm × 8 mm × 256 mm rectangular Teflon housing. Six 1-mm diameter pinholes as air inlets were created and evenly distributed at the bottom of the module; they were then connected to a nitrogen cylinder to introduce even bubble distribution inside the membrane module, as presented in Fig. 2b. It is noted that the modules were installed in a vertical orientation to maintain regular bubble shape and movement. Also, for direct observation and image collections of the bubbles, a transparent module housing/shell was adopted and a scale was attached to the observation region to measure bubble size with varying flow rate, as shown in Fig. 2c. Hence, bubble images can be clearly captured and analyzed under various gas flow rates, which were measured by an air flow meter (Dwyer, 0.1 ~ 1.0 L min⁻¹). The bubble characteristics, *i.e.*, mean bubble size and variance of bubble size distribution can be obtained according to Eq. (19).

3.2 DCMD experiments

In this study, three sets of MD experiments were carried out: 1) Set A, to study the effect of gas flow rate on the bubble characteristics and MD performance, with 3.5 wt% NaCl solution as feed; 2) Set B, to analyze the local heat-transfer resistance under different bubble flow conditions, via pure water tests; 3) Set C, to investigate the growth of NaCl scaling resistance on the membrane surface and anti-scaling efficiency by gas bubbles, with 7.0 kg 23.5 wt% NaCl solution as feed. The Module #1 (6 hollow fibers packed) was used in Set A experiments for conveniently capturing and analyzing bubble characteristics; while Module #2 (35 fibers packed) was employed for Sets B and C experiments to minimize the experimental errors from permeation flux fluctuation.

All the experiments were conducted in a batch-mode DCMD setup, which was described in our previous work [24]. In brief, both the feed and permeate streams were circulated countercurrently through the shell and lumen sides of the hollow fiber modules by peristaltic pumps (Masterflex®, Cole Palmer), respectively. The feed flow rates were kept constant as 0.41 L min^{-1} (the superficial velocity on feed side was 0.01 m s^{-1}), and the permeate flow rates were kept as 0.15 L min^{-1} (in Sets A and B experiments) and 0.28 L min^{-1} (in Set C experiments). The effect of gas flow rate on module performance was investigated

at 0.2 L min^{-1} , 0.5 L min^{-1} and 0.8 L min^{-1} , respectively. The permeation flux was recorded and measured in every 10 minutes. In addition, the operating temperature ranges were deliberately controlled for different sets of experiments. For instance, in Set B pure water tests, the feed-side temperature T_f was kept in a range of $300 \text{ K} \sim 340 \text{ K}$; while the permeate-side temperature T_p was varied between $292 \text{ K} \sim 328 \text{ K}$, respectively, to ensure a transmembrane temperature difference lower than 10 K [39]. For Set C tests with highly concentrated feed, the T_f and T_p were kept at 334 K and 294 K , respectively. The starting point ($t = 0$) for scaling formation (saturation concentration of NaCl solution) was identified and marked when a dramatic flux decline was observed during the batch-mode operation [2].

It is noted that the sodium chloride used to prepare the synthetic brine was purchased from Merck with an analytical purity of 99.5%. Also, all the experiments were repeated three times and showed good reproducibility with water fluxes within $\pm 5\%$. The conductivity meter had an accuracy of $\pm 0.1 \text{ ms cm}^{-1}$ (feed side) and $\pm 0.1 \text{ } \mu\text{s cm}^{-1}$ (permeate side), respectively. The temperature and flow rate variations were strictly controlled within $\pm 0.4 \text{ }^\circ\text{C}$ and $\pm 0.01 \text{ L min}^{-1}$, respectively.

4. Results and Discussion

4.1 Heat transfer process intensification by bubbling

4.1.1 Permeation flux vs. gas flow rates

Figure 3 shows the ratio of flux enhancement Φ (Eq. (8)) as a function of gas flow rate based on the results of Set A DCMD experiments with low salinity feed. Clearly, the Φ values for all gas flow rates are above unity, which is mainly due to the two-phase flow intensification effect on mass and heat transfers. With the improved flow mixing and intensified surface shear rate induced by bubbling, the thickness of the feed-side thermal boundary layer adjacent to the membrane surface is greatly reduced, leading to an increase of driving force (*i.e.*, trans-membrane temperature gradient, vapor pressure difference) and subsequently the permeation rate. Interestingly, compared with the non-bubbling system as benchmark (the permeation flux $J = 7.95 \text{ kg m}^{-2} \text{ h}^{-1}$), it is observed that the Φ initially increases till reaching a maximum value of 1.27 at $Q_g = 0.2 \text{ L min}^{-1}$ and then slightly decreases with increasing gas flow rate. The slight decrease of Φ with a further increase on the gas flow rate may be caused by the formation of large bubbles or slugs resulting in local channeling/by-passing and uneven flow distribution, which slightly offsets the flux enhancement ratio Φ due to the negligible increment of the surface shear intensity at a higher gas flow (Section 2.3). Hence, finer bubbles tend to be more effective in terms of performance enhancement.

4.1.2 Characterization of bubble properties vs. MD performance

To study the working mechanisms induced by bubbling in DCMD, bubble images were captured during experiments to analyze the bubble frequency in every sufficiently small size domain and size distribution curves in Origin (Eq. (19)). Figs. 4 a, b & c show the captured bubble images at various gas flowrates based on Set A experiments, with the corresponding histograms of bubble size distribution displayed in Figs. 4 d, e & f, respectively. In general, the images in Fig. 4 a, b & c qualitatively illustrate that the larger bubbles with more uneven diameters are formed at a higher gas flowrate. Accordingly, the distribution curves obtained from bubble size analysis in Figs. 4 d, e & f show that the bubble size distribution tends to be more spread out with increasing gas flowrate, *i.e.*, the size deviation from the mean value increases and the percentage of number of large bubbles increases as the curve flattens along the x direction (bubble size). According to the relationship between the bubbling frequency and bubble size given in Eq. (19), the mean bubble size and variance of size distribution curves at different gas flowrates were derived and presented in Fig. 5. The results show that the mean bubble size increases steadily from 2.80 mm to 3.81 mm and the corresponding variance increases dramatically from 0.28 to 1.80 with increasing gas flowrate from 0.2 L min⁻¹ to 0.8 L min⁻¹, which have quantitatively confirmed the information obtained from the bubble images illustrated in Fig. 4. Combined with the MD performance results shown in Fig. 3, it is clear that a narrow bubble size distribution with a smaller mean size at a lower gas

flow rate has helped to achieve a higher permeation flux, due to more effective local mixing and surface renewal of the liquid boundary layers. Thus, a careful choice of gas flowrate has to be made to avoid large bubbles or slugs that compromise module performance, not to mention the unnecessary energy consumption of aeration and potential fiber breakage and/or membrane wetting problems during long-term operation.

4.1.3 Heat-transfer assessment with and without gas bubbling in MD (pure water tests)

The Set B experiments were conducted to investigate the intensification mechanisms of heat transfer in bubbling-assisted DCMD. Based on Eq. (7), experimental data was analyzed and plotted following the linear relationship of $\Delta T / J\gamma$ and $1/(dp/dT)$, as shown in Fig. 6, which shows the comparison between the original (Fig. 6a) and bubbling-assisted DCMD systems (Figs. 6b-6d, with gas flow rates of 0.2, 0.5 and 0.8 L min⁻¹, respectively). The linearity of the data with a correlation coefficient $R^2 > 0.99$ further confirms the accuracy of applying Eq. (7) for the selected temperature range ($T_f - T_p < 10$ K). Therefore, the film heat transfer coefficient h and overall mass transfer coefficient C were calculated based on the intercept and slope of the linear regression equation. Accordingly, for non-bubbling system the film heat transfer coefficient was obtained as 177.10 J K⁻¹, while the heat transfer coefficient were derived as 407.46 J K⁻¹, 358.92 J K⁻¹ and 270.41 J K⁻¹ for gas flow rates of 0.2, 0.5 and 0.8 L min⁻¹, respectively. Obviously, the enhancement ratio (130% for gas flow

rates of 0.2 L min^{-1} , 102% for gas flow rates of 0.5 L min^{-1} and 53% for gas flow rates of 0.8 L min^{-1}) decreases with increasing gas flow rate, which can be well-associated with the MD flux results and bubble size distribution curves discussed in Sections 4.1.1 and 4.1.2.

Also, based on the heat transfer coefficients obtained from Fig. 6, the corresponding temperature polarization coefficient (*TPC*) and the ratio of conductive heat loss to overall heat flux (q_c/q) were calculated at varied gas flowrates. The respective results are given as the functions of membrane temperature T_m in Figs. 7 & 8. In Fig. 7 it is observed that the *TPC* values decrease generally with increasing membrane temperature T_m (296 ~ 333 K) for a DCMD system with or without bubbling. This is because with the bulk temperature conditions kept constant (T_f, T_p), a higher evaporation rate is caused by a higher membrane wall temperature at the feed side, T_{fm} , and subsequently leads to a higher condensation rate and a more rapid increase of the wall temperature at the permeate, T_{pm} . As a result, a lower trans-membrane temperature difference ($T_{fm} - T_{pm}$) and hence a smaller *TPC* occur at a higher membrane temperature T_m (Eq. (5)). Nevertheless, compared to the non-bubbling DCMD system, the introduction of gas bubbling obviously showed a significant improvement on the temperature polarization coefficient *TPC* and DCMD driving force ($T_{fm} - T_{pm}$) at a given membrane temperature T_m . Consistent with the heat transfer coefficients obtained from Fig. 6, the *TPC* curves in Fig. 7 decrease with increasing gas flowrate when gas bubbles are

introduced – the highest *TPC* enhancement ratio of 113% (over the non-bubbling DCMD system) is achieved at a lower gas rate of 0.2 L min⁻¹ and membrane temperature of 333 K. It has again confirmed that a low gas stream with fine bubbles and narrow bubble size distribution can greatly intensify the heat-transfer process and facilitate the mitigation of temperature polarization effect.

Similarly, in Fig. 8 the ratio of conductive heat loss to total heat flux, q_c/q , also decreases with increasing membrane temperature at a constant q , regardless of the gas rate. The main reason is that the q_c increases linearly with the trans-membrane membrane temperature difference ($T_{fm} - T_{pm}$); while the q_v has an exponentially relationship with ($T_{fm} - T_{pm}$), as indicated in (Eq. (3)). Clearly, the non-bubbling DCMD presents a lower ratio of q_c/q (from 24.36% at $T_m = 297$ K to 5.55% at $T_m = 332$ K) than any bubbling-assisted systems. This can be explained by the lower *TPC* and hence a smaller trans-membrane temperature difference ($T_{fm} - T_{pm}$) (*i.e.*, driving force) caused by the poorer mixing and flow dynamics on the feed side under non-bubbling system. On the contrary, with enhanced hydrodynamics and reduced liquid boundary layers under bubbling conditions, a dramatic increase of the driving force ($T_{fm} - T_{pm}$) and higher permeation flux is the penalty for a subsequent rise of the conductive heat q_c . Therefore, a compromise has to be made for MD system design based on this trade-off relationship between permeation flux and conductive heat loss.

4.2 Quantification of local scaling resistance in DCMD with and without bubbling

With the encouraging results of flux enhancement achieved (Section 4.1) in two-phase flow MD systems, further investigations are conducted to evaluate its effectiveness in scaling control in treating highly concentrated feeds.

4.2.1 Flux decline vs. scaling growth in non-bubbling MD

To explore the relationship of permeation flux decline and growth of scaling layer on the membrane surface, Set C experiments were carried out for high concentration batch-mode DCMD without incorporating bubbling. The results are shown in Fig. 9 in terms of the relative permeation flux ($J_{w/o|t=t1} / J_{w/o|t=0}$) (*i.e.*, the ratio of instantaneous trans-membrane flux to the initial flux value at $t=0$) and the ratio of local scaling resistance to the overall resistance (r_{sl} / r_{ov}) along the operating time. Clearly, a drastic flux decline of 65% is observed after 3.5-hour operation under supersaturation conditions of the concentration process. This is mainly due to the continuous NaCl crystal deposition on the membrane surface and hence rapid growth of scaling layer with time, which is consistent with the previous research findings [2, 7, 24, 40].

To ascertain the relationship of flux decline and scaling formation, the ratio of local scaling resistance to overall resistance (r_{sl} / r_{ov}) (Eqs. (13) and (15)) was calculated and also plotted against operation time t in Fig. 9. Obviously, the curve of (r_{sl} / r_{ov}) shows an increasing trend: at $t = 0$, the scaling resistance r_{sl} is close to zero because of the insignificant initial deposition of fine crystals on the membrane surface; as t increases and more pure water is extracted from the feed, the ratio (r_{sl} / r_{ov}) increases rapidly due to the constant rise of supersaturation level (driving force for crystallization) and subsequent accumulation of more and larger crystals on the membrane surface, which inevitably leads to severe membrane pore blockage and declined permeation flux (Fig. 9). Eventually, the resistance of scaling r_{sl} / r_{ov} reaches 65% at $t=3.5$ h and has clearly become the dominant factor for major flux drop and deterioration of heat and mass transfers in MD.

To associate the results presented in Fig. 9 with the tendency of crystal deposition and/or scaling formation on the membrane surface, post-experiment membrane inspection was conducted at different operation times. The SEM imaging results are shown in Fig. 10. At $t = 0$ h, no crystal deposition is observed on the membrane surface, as shown in Fig. 10(a); while at $t = 1.5$ h a small amount of fine crystals appear on the surface (Fig. 10(b)). Later on, a substantial crystal coverage is found at $t = 3.5$ h (Fig. 10(c)). Therefore, consistent with the tendency of flux decline along the operation time (Fig. 9), the physical observation of gradual

crystal deposition on the membrane surface has confirmed the increasing proportion of scaling resistance (*i.e.*, r_{sl}/r_{ov}) in high concentration DCMD.

4.2.2 Flux enhancement vs. scaling mitigation in bubbling-assisted MD

To reveal the fundamentals of process enhancement and scaling mitigation by incorporating gas bubbling into MD, Set C experiments including four contrast tests for high concentration batch-mode DCMD were carried out at varied gas flowrates with other operating conditions held constant. With the non-bubbling system as benchmark (*i.e.*, gas flowrate=0), the scaling resistance in high concentration DCMD with bubbling was quantified based on Eq (18). The scaling-resistance curve as a function of operating time for each gas flowrate is plotted in Fig. 11. Similarly to the discussions for Fig. 9, a general increasing trend of the scaling resistance curve is observed at a given gas flowrate. Overall, compared with the non-bubbling MD system, the scaling resistance curves at gas flowrates of 0.2 & 0.5 L min⁻¹ are significantly lower with the introduction of two-phase flow. However, a further increase of gas flowrate to 0.8 L min⁻¹ has an unexpected influence on the scaling resistance compared to the benchmark – an initial reduction and a subsequent dramatic increase as the solution saturation level elevates. The main reason is that in the initial stage of crystal formation, the introduction of bubble flow can effectively enhance the surface shear intensity and prevent crystal deposition on the membrane surface; while at high gas flowrate

(e.g., at 0.8 L min^{-1} with mean bubble size of 3.81 mm and variance of 1.80, Fig. 5), the introduction of large and uneven bubbles or slugs would instead lead to severe flow channeling and rapid decrease on the volume holdup in the membrane module, which has further accelerated the nucleation of new crystals and growth of existing particles adjacent to membrane surface. Subsequently, more severe scaling phenomenon was observed. As previously discussed in Section 4.1, a high gas flowrate is not necessarily cost-effective and could even cause an adverse effect on the module performance and scaling issue in MD.

To further analyze the effectiveness of fine and uniform bubbles under deliberately-selected gas flowrate of $0.2 \text{ \& } 0.5 \text{ L min}^{-1}$, a magnified local snapshot is given in Fig. 11 for a clear display of their scaling resistance curves benchmarked against that of the non-bubbling MD system with saturated feed solution. As stated previously, it is observed that scaling resistance has been significantly reduced with the injection of gas bubbles, which may induce secondary flows and physically displace the concentration layer adjacent to membrane surface. In general, compared to steadily increasing trend of the non-bubbling curve, the resistance curves with bubbling initially show an insignificant increase (*i.e.*, 0 ~ 2 hour, Fig. 11) and then a relatively rapid rise after certain operation time (*i.e.*, > 2 hour, Fig. 11). The possible reason is: as initial nucleation occurs with the increasing solution saturation level, the enhanced shear stress on the membrane surface induced by the two-phase flow is

expected to partially remove small crystals from the nascent scaling layer. This observation is consistent with our previous research findings [24], which have confirmed that the critical point of major permeation flux decline associated with scaling formation could be significantly delayed with the introduction of two-phase flow into high concentration MD. Yet, a continuously increasing supersaturation level and more rapid nucleation rate in batch-mode MD would inevitably lead to a deterioration of crystal aggregation and deposition on the membrane surface.

A further comparison is made to evaluate the operability of gas-bubbling DCMD system beyond supersaturation in terms of its capacity to remove water and reduce discharge in a given operation time, defined as water product, W , $\text{g}\cdot\text{m}^{-2}$. The values of W can be obtained from dividing the weight of water remove in the range of given time by the membrane area used in the batch mode DCMD. The results are displayed via a bar chart in Fig. 12 for various gas flowrates. Compared to the non-bubbling MD system, the introduction of bubbling exhibits great potential in maximizing the water removal from supersaturated brines – the highest enhancement ratio of 2.3-fold at gas flowrate of 0.5 L min^{-1} , followed by 2- and 1.83-fold at 0.2 and 0.8 L min^{-1} , respectively. Clearly, appropriately chosen bubbling conditions can help achieve maximum anti-scaling effect and postpone the water flux decline in MD brine concentration.

5 Conclusions

This study has explored the intensification mechanisms of gas bubbling for heat transfer enhancement and scaling alleviation in MD brine processing. Relatively small bubble size and narrow size distribution **at a low gas flow rate** are preferred to attain even flow distribution and enhanced surface shear intensity. The highest heat-transfer coefficients were obtained with fine bubbles at optimized bubbling conditions. Correspondingly, the temperature polarization effect was greatly mitigated with the penalty of an unavoidable higher conductive heat loss to the membrane matrix. The trade-off relationship suggested that a compromise has to be made to accomplish a desired MD thermal efficiency.

Besides heat transfer resistance, fouling/scaling is another significant roadblock to the **wider adoption** of membrane distillation technology, especially for high concentration operation. In the MD brine concentration experiment without bubbling, the rapid decline of the measured relative flux ($J_{w/o|t=t1} / J_{w/o|t=0}$) accorded with the increasing contribution of the local scaling resistance to the overall mass transfer resistance (r_{sl} / r_{ov}), implying that the resistance of scaling has become the controlling factor for permeation flux drop. In comparison, the introduction of gas bubbling into MD could significantly reduce the scaling resistance, due to the high shear stress induced by gas bubbles and the removal of tiny

deposited crystals from the membrane surface. However, an unreasonably high gas flow rate has the adverse effect due to the formation of large and uneven bubbles or slugs resulting in local by-passing and increased frequency of crystal collision.

In a word, to effectively control scaling formation for a more operable MD brine concentration process (especially beyond supersaturation level), optimized aeration conditions with a smaller mean bubble size and narrower bubble size distribution are preferred. Nevertheless, the incorporation of supplementary membrane cleaning techniques is still necessary for effectively restoring the membrane performance as the scaling layer matures.

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Nomenclature

C	overall mass transfer coefficient ($\text{kg m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$)
d_{max}	maximum diameter of membrane pore size (μm)
d_{mean}	mean diameter of membrane pore size (μm)
d_o	outside diameter of hollow fiber (mm)
d_s	housing diameter of membrane module (mm)
E_t	tensile module (MPa)
H	effective heat transfer coefficient (J K^{-1})
h	the local heat-transfer coefficient (J K^{-1})
J	transmembrane flux ($\text{kg m}^{-2} \text{s}^{-1}$)
k_m	overall thermal conductivity of the membrane ($\text{W m}^{-1} \text{K}^{-1}$)
L	effective fiber length (mm)
M	molecular weight of water (g mol^{-1})
P	vapor pressure (Pa)
q	over heat-transfer flux in MD (J)
q_c	conductive heat flux across the membrane (J)
q_v	latent heat transfer accompanying the vapor flux (J)
Q	volume flow rate (L min^{-1})
R	gas constant (8.314 J K^{-1})
R^2	dimensionless of linear fitting correlation coefficient
r	mass transfer resistance ($\text{Pa m}^2 \text{h kg}^{-1}$)
t	time (s)
T	temperature (K)

TPC	temperature polarization coefficient (dimensionless)
ΔT	temperature difference (K)
W	water product output per unit membrane area during the recorded DCMD running time (g m^{-2})
x_w	constant in GaussAmp function
x	bubble size (mm)
x_c	constant in GaussAmp function
y	bubbling frequency in a specific size range
y_0	constant in GaussAmp function
y_l	constant in GaussAmp function

Greek letters

γ	latent heat-of-vaporization (J kg^{-1})
Φ	dimensionless ratio of flux enhancement
$\tilde{\alpha}$	porosity (%)
δ_m	thickness of hollow fiber (μm)
$\ddot{\alpha}_b$	strain at break (%)

Subscripts

g	gas
m	membrane
f	feed
sl	scaling

<i>fm</i>	membrane surface on the feed side
<i>ov</i>	overall
<i>p</i>	permeate
<i>pm</i>	membrane surface on the permeate side
<i>w</i>	with bubbling
<i>w/o</i>	without bubbling

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