Membrane distillation to regenerate different liquid desiccant solutions for air conditioning

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Desalination

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Abstract: The capacity of membrane distillation (MD) to regenerate three commonly used liquid desiccant solutions (i.e. $CaCl_2$, LiCl, and a mixture of $CaCl_2$/LiCl) for liquid desiccant air-conditioners (LDAC) was investigated. The results demonstrate considerable impact of the concentration polarisation effect on the process water flux during MD regeneration of these three desiccant solutions. For each of these liquid desiccant solutions, the experimentally measured water flux of the MD process was about half of the calculated value using the process mass transfer coefficient (K_m) obtained during the process characterisation in which the concentration polarisation effect was neglected. The observed deviation between the experimentally measured and calculated process water flux indicates the need to include the concentration polarisation effect in the model for calculating water flux. Although Ca^{2+} concentration in the $CaCl_2$ and $CaCl_2$/LiCl liquid desiccant solutions exceeded the solubility limit for $CaCO_3$, membrane scaling was not observed. Nevertheless, there was evidence that membrane fouling might occur during extended MD regeneration of liquid desiccant solutions containing $CaCl_2$.

Keywords: membrane distillation (MD); liquid desiccant air-conditioner (LDAC); liquid desiccant regeneration; mixed liquid desiccant solutions.

1. Introduction

Satisfying the desire for both thermal comfort and energy saving in buildings is a great challenge to the air-conditioning industry. Most air-conditioning systems are currently based on conventional mechanical vapour compression, in which the air is first cooled to below its dew point for dehumidification and then reheated to obtain the desired temperature [1-4]. In these systems, energy is wasted due to over cooling and the subsequent reheating of the air, particularly in areas with high humidity. As a result, there is a growing interest in alternative air-conditioning technologies to meet the thermal comfort requirement but with lower energy footprint [1, 3].

Liquid desiccant air-conditioning (LDAC) is a promising alternative to conventional mechanical vapour compression air-conditioners [5-8]. LDAC systems control air humidity by absorbing moisture from the air into a liquid desiccant stream. The sensible heat load can then be achieved using a complementary evaporative cooler. This technique eliminates the risk of over cooling and the subsequent reheating, and hence is more energy efficient. Indeed, energy consumption by LDAC is about one fourth of that by a vapour compression system. In addition, LDAC can utilise solar thermal energy rather than electricity [2, 7].

Aqueous solutions of halide salts (e.g. LiCl and $CaCl_2$) have been used as liquid desiccants in LDAC. At the same weight concentration, LiCl solution has a high dehumidification capacity but is expensive and toxic if released into the environment. Compared to LiCl solution, $CaCl_2$ solution offers a lower dehumidification capability; however, it is more affordable and less toxic (i.e. indeed $CaCl_2$ is widely used as a domestic moisture absorbent). As a result, mixtures of $CaCl_2$ and LiCl have also been proposed for LDAC applications [2-4].

A core component of LDAC is liquid desiccant regeneration, in which excess water is removed to maintain the dehumidification capacity. Liquid desiccant regeneration accounts for over three-quarters of the total energy consumption of LDAC systems [9]. Most current LDAC systems use a thermal evaporator for liquid desiccant generation. During this process, the weak (i.e. diluted) liquid desiccant solution is heated up to about 90 °C and then sprayed over a packed-bed contact media. Hot air is blown counter-currently along the contact media to remove water vapour from the packed-bed, therefore reconcentrating the liquid desiccant solution. The reconcentrated liquid desiccant is then cooled down prior to re-entering the dehumidifier. Desiccant carry-over is a major issue associated with the current regeneration method due to the direct contact between liquid desiccant solution and the hot air stream. Desiccant carry-over inevitably results in desiccant loss (thus increasing chemical cost), corrosion risk to building equipment, and health concerns due to potential exposure to toxic desiccant chemicals [8, 10, 11].

Several processes have been explored to replace the thermal evaporator for liquid desiccant regeneration in LDAC [12-14]. One of them is membrane distillation (MD), a thermally-driven membrane separation process [6, 15, 16]. MD uses a microporous hydrophobic membrane to separate the liquid desiccant and the permeate streams. The hydrophobic membrane allows only water vapour to permeate through it while retaining all dissolved salts in the liquid desiccant stream [17, 18], thus eliminating desiccant loss due to carry-over [6, 16]. Moreover, MD utilises a transmembrane water vapour pressure gradient as the driving force for salt-water separation, and the liquid phase is discontinued across the membrane [17, 19]. The MD process is not affected by the osmotic pressure of the feed solution. Thus, MD applications have been successfully demonstrated for a range of saline solutions including seawater [20, 21], industrial wastewater [22, 23], and even brines from seawater desalination [24-26] and natural gas exploration [27-30] as well as draw solutions for forward osmosis operation [31, 32]. Finally, because the MD process can be efficiently operated at mild feed temperature [33, 34], low-grade waste heat and solar thermal energy can be utilised to reduce the energy cost of liquid desiccant regeneration in LDAC systems.

To date, there have been only three attempts to explore the feasibility of MD for regeneration of liquid desiccant solution for LDAC, and only LiCl solutions were investigated in all these studies. Choo et al. [4] examined the thermal performance, water flux, and regeneration capacity of a pilot MD process for regeneration of LiCl solutions under various operating conditions, particularly the LiCl concentration and feed temperature. Duong et al. [6] systematically demonstrated the technical viability of MD regeneration of LiCl solutions. Rattner et al. [15] optimised the operating conditions of an MD regeneration process of LiCl solutions for LDAC applications using computer simulation. MD regeneration of $CaCl_2$ solutions and mixed $CaCl_2$/LiCl solutions used for LDAC systems remains a research gap in the literature. Given the difference in scaling propensity and kinetic properties between $CaCl_2$ and LiCl solutions, MD regeneration of $CaCl_2$ solutions and mixed $CaCl_2$/LiCl solutions might deviate from that of LiCl solutions.

This study aimed to demonstrate the viability of the MD process for regeneration of $CaCl_2$ solution and mixed $CaCl_2$/LiCl solution in comparison with LiCl solution. The MD process was first characterised with Milli-Q water to determine the baseline process water flux and mass transfer coefficient. Then, the influence of different liquid desiccants on the process water flux was then manifested in MD experiments with three liquid desiccant solutions (e.g. $CaCl_2$ and LiCl solutions and a mixed $CaCl_2$/LiCl solution). Finally, polarisation effects and membrane scaling behaviour of the MD process during a continuous concentration of the three liquid desiccant solutions were examined.

2. Materials and methods

2.1. Experimental system

A bench scale direct contact MD (DCMD) system was utilised (Fig. 1). The system comprised a plate-and-frame membrane module and hot- and cold-water cycles. The membrane module consisted of two acrylic semi-cells with engraved flow channels with depth, width, and length of 0.3, 9.5, and 14.5 cm, respectively. In the hot-water cycle, feed solution (1.8 L) from the feed tank was heated in a hot water bath using submerged stainless-steel coils prior to entering the feed channel. As the hot feed solution travelled along the feed channel, water evaporated at the membrane surface and permeated through the membrane in vapour form to the distillate channel; thus, the feed solution was concentrated. The concentrated feed solution was then returned to the feed tank. On the other side of the membrane, Milli-Q water (1.8 L) was used as the initial distillate, and was circulated through the distillate channel to condense the permeated water vapour. The temperatures of the feed solution and the distillate were regulated using a temperature control unit and a chiller, respectively. Temperature sensors were placed immediately before the inlets of the feed and distillate channels. Two variable-speed gear pumps and rotameters were used to regulate the feed and distillate circulation rates.

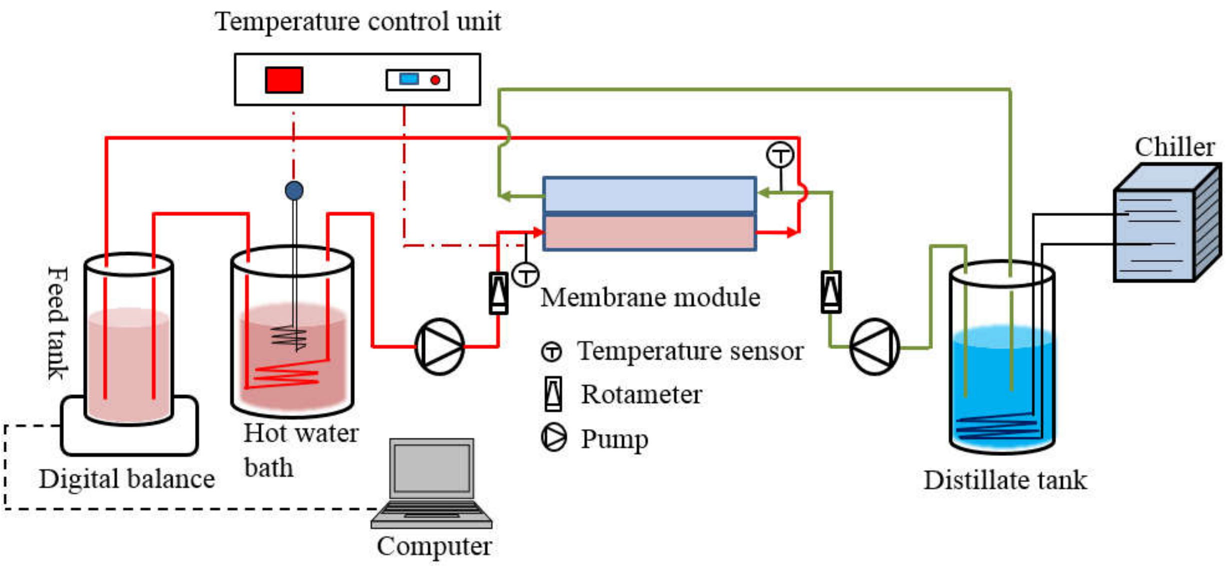

Fig. 1. The schematic diagram of the DCMD system.

2.2. Materials

A flat-sheet hydrophobic polytetrafluoroethylene (PTFE) membrane from Porous Membrane Technology (Ningbo, China) was selectively used in this study. The membrane had thickness, nominal pore size, and porosity of 60 μm, 0.2 μm, and 80%, respectively. The effects of different MD membrane materials including PTFE, polyvinylidene fluoride (PVDF), and polypropylene (PP) on regeneration of liquid desiccant solutions will be investigated in a future study.

Milli-Q water and three liquid desiccant solutions were used as the MD feed in this study. The liquid desiccant solutions contained a single $CaCl_2$ or LiCl salt, or a mixed $CaCl_2$/LiCl salts (1:1 in weight) with a concentration of 20 wt.%. Milli-Q water and laboratory grade anhydrous $CaCl_2$ and LiCl were used to prepare the liquid desiccant solutions.

2.3. Analytical methods

The electrical conductivity of the distillate was measured using an Orion 4-Star Plus pH/conductivity meter (Thermo Scientific, Waltham, Massachusetts, USA). The salt concentration of the feed solution was calculated based on the initial concentration (i.e. 20 wt.%) and the recorded weight of the feed solution with the assumption that the MD process provided a complete salt rejection. The MD process water flux was measured by continuously weighing the feed tank using a digital balance connected to a computer.

The surface morphology of the virgin and fouled membranes was examined using a low vacuum scanning electron microscope (SEM) (JOEL JSM-6490LV, Japan). Membrane samples were air-dried and subsequently coated with a thin layer of gold before the SEM analysis.

2.4. Experimental protocols

2.4.1. Process characterisation

The MD process was characterised with Milli-Q water as the feed. The MD process was operated at constant distillate temperature (20 °C) and feed and distillate circulation rates (0.045 m/s) while the feed temperature was varied from 45 to 65 °C. The MD system was operated for at least one hour or until stable water flux had been achieved. Operation under steady state condition was extended further for one hour for recording the water flux every 5 minutes.

2.4.2. MD regeneration of liquid desiccant solutions

MD experiments with the liquid desiccant solutions were conducted at feed temperature of 55, 60, and 65 °C with the same operating conditions as described in section 2.4.1. During these experiments, the distillate obtained from the process was returned to the feed tank to maintain a constant feed salt concentration. Water flux and distillate conductivity were measured every five minutes for one hour after the process had reached its stable operation.

Extended MD experiments were operated at feed temperature of 65 °C until the process water flux decreased to about 2 L/m^2·h. During these experiments, the obtained distillate was not returned to the feed tank; therefore, the liquid desiccant concentration increased throughout the MD process. At the completion of the MD process, the membrane was either dismounted from the MD system and air dried for SEM analysis. To assess membrane reusability, the extended MD process was repeated, but when the water flux had decreased to 2 L/m^2·h the process was terminated to rinse the system with Milli-Q water for 5 minutes before restarting it with fresh liquid desiccant solutions (i.e. 20 wt.%).

2.5. Water flux and mass transfer coefficient of the MD process

Water flux in DCMD can be calculated theoretically using the membrane mass transfer coefficient and the water vapour pressure difference between the two sides of the membrane as [19, 35]:

$$J = C_m \times (P_{m.feed} - P_{m.distillate}) \tag{1}$$

where J is water flux (L/m^2·h), C_m is the membrane mass transfer coefficient (L/m^2·h·Pa), and $P_{m.feed}$ and $P_{m.distillate}$ are the water vapour pressure (Pa) at the feed and distillate membrane surfaces, respectively. While C_m can be calculated with given membrane properties and operating conditions, $P_{m.feed}$ and $P_{m.distillate}$ are calculated from the temperature immediately at the membrane surface. These values cannot be measured but can only be indirectly calculated from the feed and distillate temperature in the bulk solutions by considering the temperature and concentration polarisation phenomena. Therefore, water flux calculation using the Eq. (1) is impractical. A practical way to circumvent this issue is to use the process mass transfer coefficient and water vapour pressure of the feed and distillate streams to calculate the process water flux as [36-38]:

$$J = K_m \times (P_{feed} - P_{distillate}) \tag{2}$$

where K_m is the process mass transfer coefficient (L/m^2·h·Pa), and P_{feed} and $P_{distillate}$ are the water vapour pressure (Pa) of the feed and distillate streams, respectively. Water vapour pressure of the Milli-Q water feed and the distillate is a function of temperature, and can be calculated using the Antoine equation [19, 35]:

$$P_{distillate} = \exp\left(23.1964 - \frac{3816.44}{T_{distillate} - 46.13}\right) \tag{3}$$

In addition to temperature, water vapour pressure of a saline solution depends on the salt concentration. In this study, the water vapour pressure of the feed solutions containing LiCl or $CaCl_2$ at a given temperature was calculated based on the procedures described by Conde [39].

3. Results and discussions

3.1. MD process characterisation

The MD process was characterised to determine the process water flux and mass transfer coefficient (K_m) at various feed operating temperatures. In good agreement with the MD literature, the process water flux with the Milli-Q water feed increased with the feed operating temperature (Fig. 2) [36, 37, 40]. This is attributed to the exponential relationship between water vapour pressure and temperature as demonstrated in the Antoine equation (Eq. 3). Increasing feed temperature when the distillate temperature remained constant led to an increase in the water vapour pressure difference between two sides of the membrane, which is the actual driving force for water transfer across the membrane, thus increasing the process water flux.

Unlike the water flux, K_m of the MD process decreased almost linearly as the feed temperature increased. Indeed, when the feed temperature was increased from 45 to 65 °C, K_m decreased by around 16% from 1.55×10^{-3} to 1.31×10^{-3} $L/m^2\cdot h\cdot Pa$ (Fig. 2). The decrease in K_m with increasing feed temperature was due to the temperature polarisation phenomenon. During the MD process with Milli-Q water, temperature polarisation rendered the water temperatures at the membrane surfaces different from those in the bulk feed and distillate streams. It is noteworthy that the temperature polarisation effect was embedded in K_m as its determination involved the feed and distillate temperatures in the bulk streams but not at the membrane surfaces (section 2.5). Increasing feed temperature (when other operating conditions were constant) increased the process water flux but also aggravated the temperature polarisation effect. As a result, K_m of the MD process decreased with the increased feed temperature.

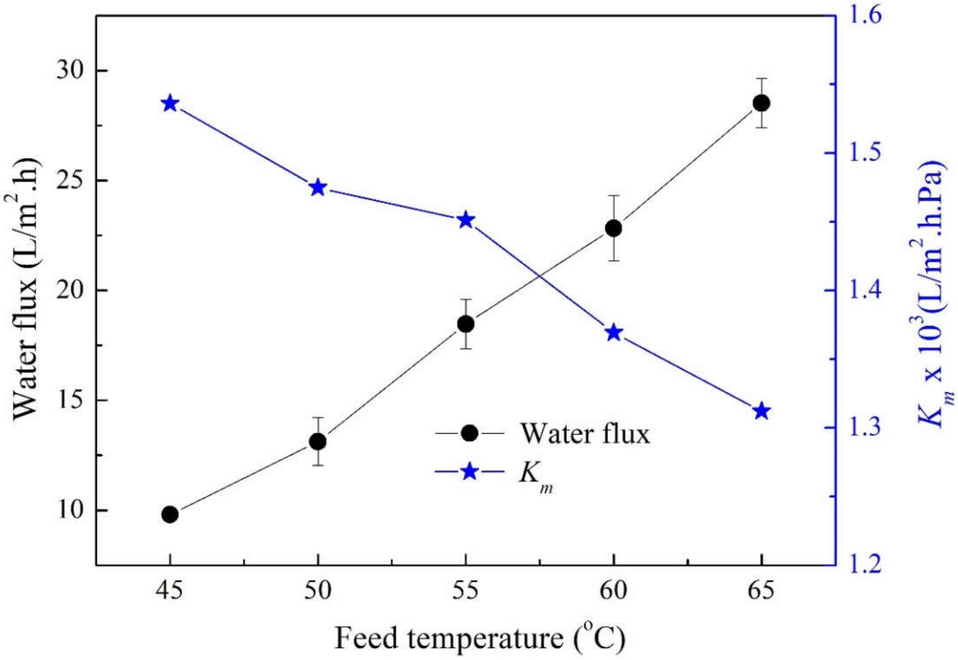

Fig. 2. Water flux and the mass transfer coefficient (K_m) of the MD process with Milli-Q water feed at various operating feed temperatures. Other operating conditions: distillate temperature $T_{distillate}$ = 20 °C, feed and distillate cross flow velocity V_{feed} = $V_{distillate}$ = 0.045 m/s. Error bars represent the standard deviations of 5 measurements.

The observed decrease in K_m during the process characterisation confirms its dependence upon the process operating conditions. This dependence is envisaged being heavier for the MD process with liquid desiccant solutions with hyper salinity. During the process characterisation, Milli-Q water was used as the feed; therefore, only temperature polarisation effect was experienced. With liquid desiccant solutions, the MD process encounters both temperature and concentration polarisation effects. The concentration polarisation effect causes the desiccant concentration at the feed membrane surface higher than that in the bulk feed stream, and hence reduces the water vapour pressure at the feed membrane surface.

3.2. Influences of liquid desiccant on water flux of the MD process

The presence of salts at high concentration in the liquid desiccant solutions markedly reduced the MD process water flux as compared with the Milli-Q water feed (Fig. 3). There were several factors behind the significant reduction in water flux of the MD process with the liquid desiccant solutions. Firstly, given their high affinity to absorb water, LiCl and $CaCl_2$ in the liquid desiccant solutions strongly reduced the water vapour pressure at the feed membrane surface and hence the water vapour pressure gradient across the membrane [6, 39, 41]. Secondly, unlike the MD process with Milli-Q water, the process with the liquid desiccant solutions suffered from the concentration polarisation effect in addition to the temperature polarisation effect. Due to concentration polarisation, salt concentration at the membrane surface was higher than that in the bulk feed stream, further reducing the water vapour pressure at the feed membrane surface and hence the process water flux. This negative influence of concentration polarisation on the process water flux will be discussed more detailed in the section 3.3. Finally, the presence of LiCl and $CaCl_2$ at high concentration increased the dynamic viscosity of the feed stream, and therefore reduced the heat transfer coefficient from the bulk feed stream to the feed membrane surface [39, 42, 43]. Indeed, our calculation using empirical equations from [39] showed that at 60 °C the viscosity of the LiCl 20 wt.% and $CaCl_2$ 20 wt.% solutions was 2.5 and 2.1 times, respectively, higher than that of pure water.

Amongst the three liquid desiccant solutions, the lowest water flux was obtained from the LiCl 20 wt.% solution, followed by the $CaCl_2$/LiCl mixture and $CaCl_2$ 20 wt.% solution (Fig. 3). This order is consistent with the respective water vapour pressure and dynamic viscosity of these liquid desiccant solutions (at the same weight concentration) [39, 44, 45]. At the same weight concentration, LiCl solution has the lowest water vapour pressure and thus the highest dehumidification efficiency amongst these three liquid desiccant solutions. Nevertheless, as can be seen in Fig. 3, high dehumidification efficiency is also associated with the challenge of subsequently removing water to regenerate the liquid desiccant solution.

The water flux when regenerating liquid desiccant solutions also increased with the operating temperature, but at a lower rate compared to that of the process with Milli-Q water feed. Results in Fig. 3 show the impact of concentration polarisation on the water flux of the MD process. Only temperature polarisation could occur during the process characterisation with Milli-Q water as the feed. On the other hand, when a liquid desiccant solution was used as the feed, the MD process was influenced by both temperature and concentration polarisation. Operating the process at a higher feed temperature increased the process water flux, but also aggravated the polarisation effects and therefore negatively impacted the process water flux.

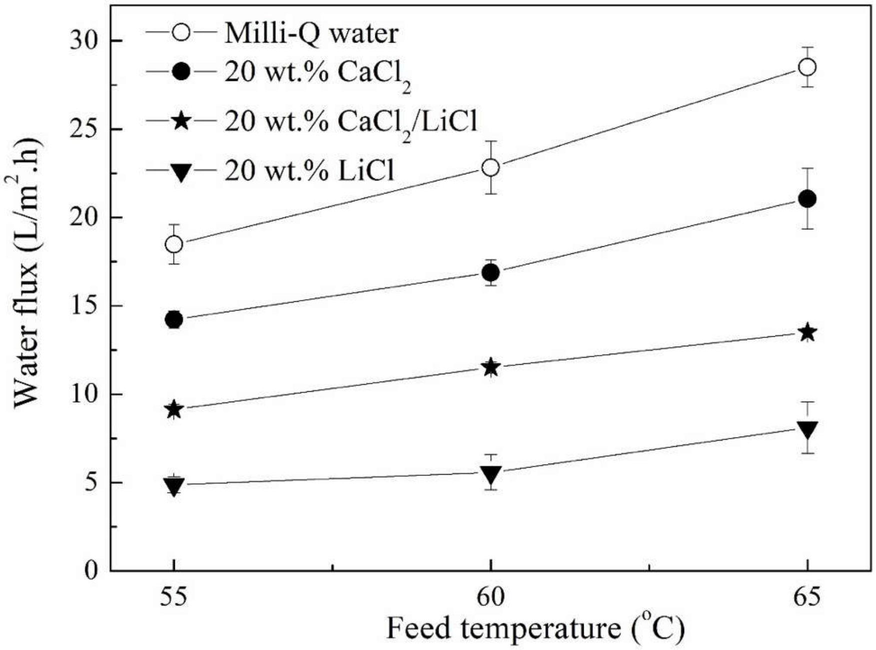

Fig. 3. Water flux of the MD process with various desiccant solutions at different operating feed temperatures. Other operating conditions: distillate temperature $T_{distillate}$ = 20 °C, feed and distillate cross flow velocity V_{feed} = $V_{distillate}$ = 0.045 m/s. Error bars represent the standard deviations of 5 measurements.

3.3. Concentration of liquid desiccant solutions by the MD process

Increasing feed salinity during the concentration of the liquid desiccant solutions inevitably led to reduction in the MD process water flux. As demonstrated in Fig. 4, water flux of the MD process with the three liquid desiccant solutions sharply decreased when they were concentrated. This observed water flux decrease was attributed to the decreased water vapour pressure together with the increased dynamic viscosity of the liquid desiccant solutions as their concentration increased [39, 41, 46]. While the former directly reduced the vapour pressure gradient across the membrane, the latter negatively impacted the heat transfer coefficient from bulk feed solution to the feed membrane surface [19, 35]. It is noteworthy that the heat transfer across the thermal boundary layers is the rate-controlling factor in the DCMD process [19, 35, 47]. The reduced heat transfer coefficient resulted in more severe temperature polarisation, and consequently led to water flux decline.

The deviation between the experimentally measured and the calculated water flux of the MD process with the liquid desiccant solutions demonstrated the significant influence of concentration polarisation on water flux. The experimentally measured fluxes of the MD process with the LiCl and $CaCl_2$ solutions were much lower than the calculated ones using the K_m value obtained during the process characterisation and the Eq. (2). It is worth noting that K_m was determined using Milli-Q water feed when the concentration polarisation effect was negligible. In addition, the feed water vapour pressure in the Eq. (2) was calculated using the salt concentration in the bulk feed stream. For the MD process with the liquid desiccant solutions of hyper salinity, the concentration polarisation effect became particularly noticeable. The concentration polarisation effect increased the salt concentration and hence rendered a smaller water vapour pressure at the feed membrane surface as compared to that in the bulk feed stream, and probably reduced the K_m value (i.e. like the temperature polarisation effect). The results reported here suggest the need to include concentration polarisation effect when simulating the MD process with liquid desiccant solutions, unlike in the MD application for seawater desalination where the concentration polarisation effect is indiscernible and hence often neglected [24, 36, 40].

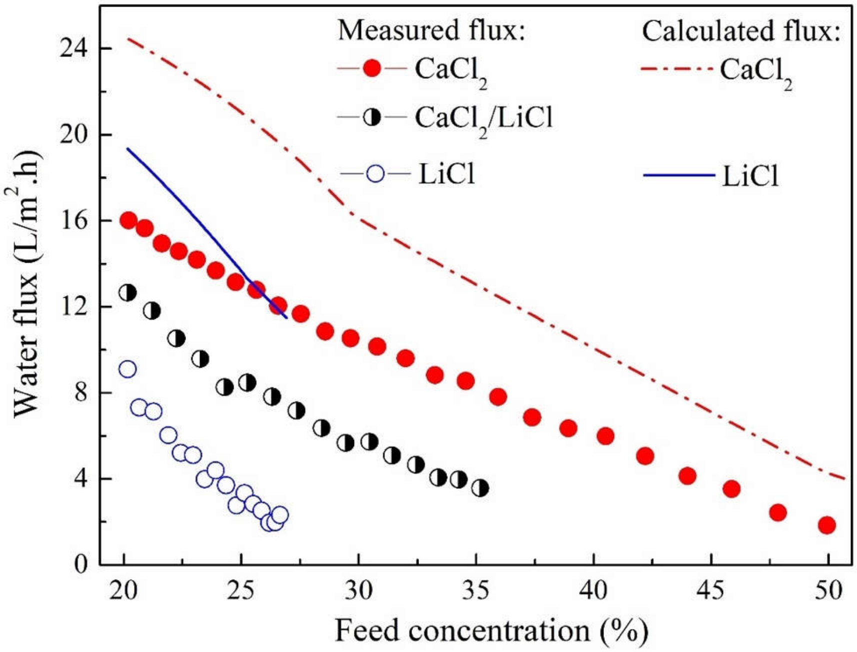

Fig. 4. Experimentally measured and calculated water flux of the MD process during the concentration of the liquid desiccant solutions. Operating conditions: T_{feed} = 65 °C, $T_{distillate}$ = 20 °C, $V_{feed} = V_{distillate}$ = 0.045 m/s.

There was no clear evidence of membrane scaling during the MD regeneration of LiCl and particularly $CaCl_2$ solutions. The electrical conductivity of the obtained distillate remained below 14 µS/cm (i.e. representing a mostly complete salt rejection) throughout the MD process with the three liquid desiccant solutions. Moreover, the water flux of the process with the fresh 20 wt.%

solutions was completely restored after rinsing the used membrane with Milli-Q water. This result is noticeable since Ca^{2+} concentration of 0.18 mol/L (in the 20 g/L $CaCl_2$ desiccant solution) and ambient concentration of CO_2 in the air exceed the solubility limit with respect to $CaCO_3$. Several previous studies have also showed that $CaCO_3$ did not cause scaling during MD operation [48, 49]. However, it is worthwhile to note the difference in the membrane surface after regenerating the LiCl and $CaCl_2$ or $CaCl_2$/LiCl solution (Fig. 5). The membrane surface after regenerating the LiCl solution was clean and similar to that in the virgin condition (Fig. 5A&B). On the other hand, a thin layer of amorphous material could be observed on the membrane surface after regenerating either the $CaCl_2$ or $CaCl_2$/LiCl solution (Fig. 5C&D). These results indicate the need to consider membrane fouling in long-term experiments that simulate the regeneration of liquid desiccant solution over several years.

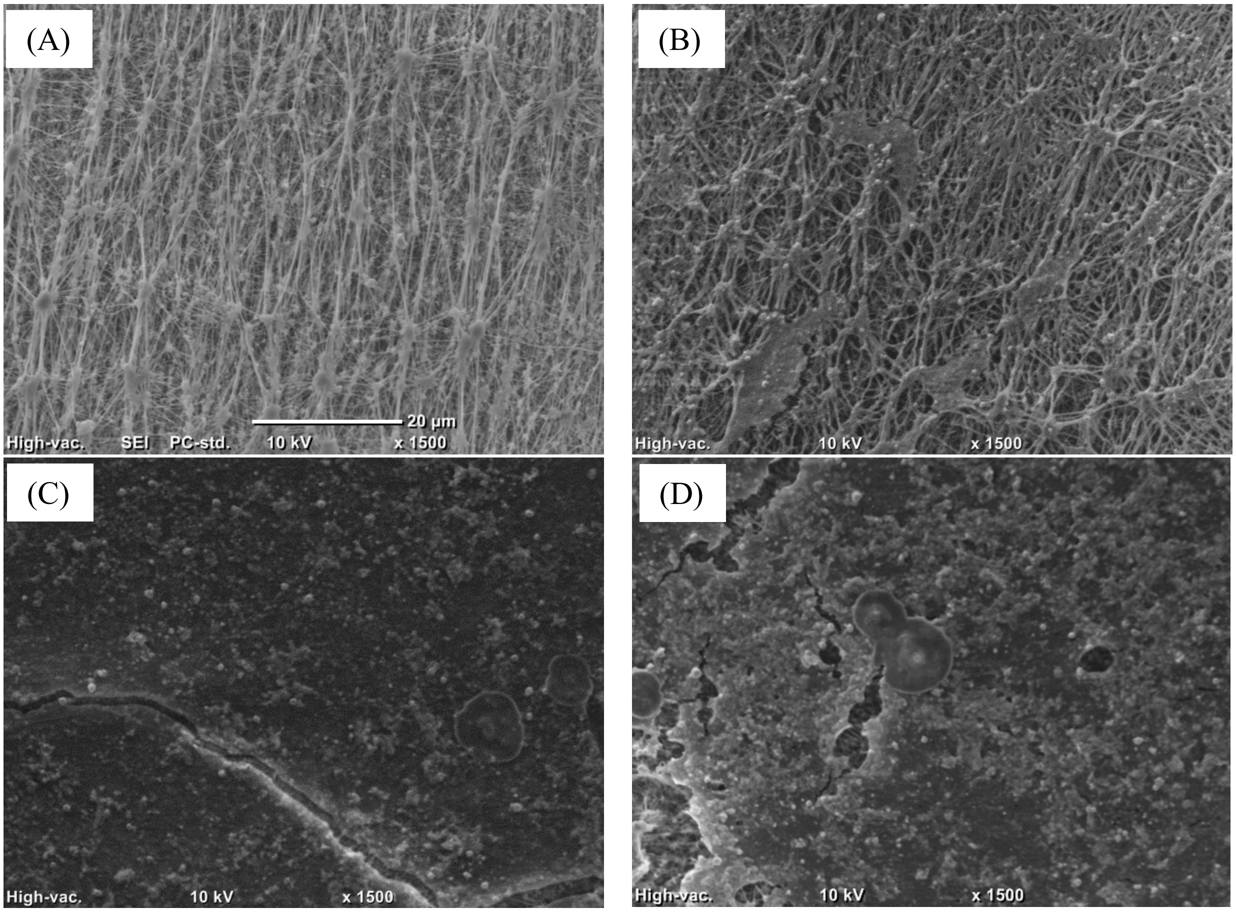

Fig. 5. SEM images of the (A) virgin membrane and the membrane after the 8-hour MD operation with (B) the LiCl solution, (C) the $CaCl_2$ solution, and (D) the mixed $CaCl_2$/LiCl solution.

4. Conclusions

MD regeneration of three liquid desiccant solutions (e.g. $CaCl_2$, LiCl, and mixed $CaCl_2$/LiCl 20 wt.% solutions) was investigated in this study. The results demonstrate considerable influence of the polarisation effects on the process water flux during MD regeneration of these desiccant solutions. The water flux of the MD process with the liquid desiccant solutions was significantly lower than that obtained during the process characterisation with the Milli-Q water feed. This water flux reduction was attributed to the impacts of $CaCl_2$ and LiCl at very high concentrations on the solutions thermodynamic properties (e.g. equilibrium water vapour pressure and viscosity) and particularly the concentration polarisation effect. The discernible concentration polarisation effect also caused the experimentally measured process water flux much lower than the calculated value using the model that neglected the concentration polarisation effect. The experimental results also indicate the need for further research to address the issue of membrane fouling for MD regeneration of liquid desiccant solutions containing $CaCl_2$ during extended operation over several months or years.

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