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Osmotic membrane distillation regeneration of liquid desiccant solution used for air-conditioning: A proof of concept

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Abstract. Liquid desiccant air-conditioning (LDAC) has emerged as a promising technology to meet the increasing demand for indoor thermal comfort. In LDAC systems, regeneration of liquid desiccant solution is a critical step. Direct contact membrane distillation (DCMD) has been explored for regeneration of liquid desiccant solutions used in LDAC systems, but faces technical challenges caused by the hyper salinity of the solutions. Osmotic membrane distillation (OMD) is a variant of DCMD that can treat hyper saline waters. In this study, a proof of concept of OMD regeneration of liquid desiccant solution was experimentally demonstrated. Compared to DCMD, the OMD process exhibited significantly higher water flux due to reduced water vapor pressure on the permeate side of the membrane. Indeed, the OMD process of the liquid desiccant 20 % LiCl solution feed achieved water flux two-fold higher than that of the DCMD process could regenerate the LiCl solution of much higher concentration: the OMD process with the initial 20 % CaCl₂ solution osmotic agent concentrated LiCl solution from 20 % to 38.4 % after a continuous 8-hour operation.

Keywords: osmotic membrane distillation, membrane distillation, concentration polarization effects, liquid desiccant air conditioning, liquid desiccant regeneration.

Classification numbers: 3.7.1, 3.8.1.

1. INTRODUCTION

In recent years, liquid desiccant air-conditioning (LDAC) has emerged as a promising technology to meet the growing demand for indoor thermal comfort together with environmental concerns [1, 2]. Compared to traditional vapor compression air-conditioning systems, LDAC can

achieve improved humidity control with significant energy saving [3]. The LDAC process uses a liquid desiccant solution to directly absorb moisture from the air, thus dehumidifying the air without the need for deep cooling to dew point temperature as required in the traditional vapor compression air-conditioning systems. With this working principle, the energy consumption of the LDAC process is significantly reduced compared to that of the traditional vapor compression air-conditioners. The energy consumption of the LDAC process is primarily attributed to the regeneration of liquid desiccant solution. As a result, regeneration of liquid desiccant solution is critical for the LDAC process [4, 5].

Various methods have been proposed for the regeneration of liquid desiccant solutions. However, most of the proposed methods face technical challenges associated with the carry-over of desiccant and the ultra-high salinity of liquid desiccant solutions. For example, in the thermal evaporation method, a hot liquid desiccant solution is sprayed counter-currently with a dry, hot air stream inside a packed bed medium to desorb moisture from the solution and hence regenerate it [5, 6]. Thermal evaporation is workable with liquid desiccant solutions at high concentrations, but in this process the direct contact between liquid desiccant and air streams inevitably leads to the carry-over of tiny desiccant droplets within the air stream [5, 6]. Membrane processes including reverse osmosis (RO) and electrodialysis (ED) have been tested for regenerating liquid desiccant solutions and proved to be effective in eradicating the desiccant solutions hinders the transfer of water through the RO or ED membrane, hence constraining the regeneration capacity of these two processes [7 - 9].

Recently, membrane distillation (MD) has been explored for the regeneration of liquid desiccant solutions [3, 10, 11]. The MD process is a combination of thermal evaporation and membrane separation methods; therefore, it can regenerate liquid desiccant solutions without any problems with desiccant carry-over [3, 11]. Moreover, as the MD process can be powered by solar energy, using MD to regenerate liquid desiccant solutions can reduce the energy cost of LDAC systems. However, like RO and ED, the hyper salinity of liquid desiccant solutions impedes the regeneration capacity of the MD process. A previous study demonstrated that a direct contact membrane distillation (DCMD) process at a feed temperature of 65 °C could concentrate the LiCl solution only to 29 % [3]. To regenerate liquid desiccant LiCl solutions at higher concentrations, the DCMD process requires to be operated at a much higher feed temperature or in a different configuration.

Osmotic membrane distillation (OMD), which is a variant of DCMD, can be an ideal process for liquid desiccant solution regeneration application. In OMD, an osmotic agent (i.e. a concentrated saline solution) is circulated on the permeate side of the membrane instead of the fresh distillate stream in DCMD [12, 13]. Compared to fresh distillate, the osmotic agent offers a noticeably lower water vapor pressure on the permeate side of the membrane, hence increasing the transmembrane water vapor pressure gradient, which is the actual driving force for the transfer of water from the feed through the membrane. Given the low water vapor pressure of the osmotic agent, OMD can treat highly saline feed waters without the need for operating at high feed temperatures. Indeed, OMD has been demonstrated to handle a variety of challenging feed waters including fruit juices and saline brine [12, 14 - 16], but it has not yet been tested to regenerate liquid desiccant solutions used in LDAC systems.

This study aims to demonstrate a proof-of-concept of OMD regeneration of liquid desiccant solutions used in LDAC systems. The performance of the OMD and DCMD processes with the same feed solutions (e.g. fresh water or LiCl solution) under the same conditions was evaluated to highlight the advantages of OMD over DCMD for the regeneration of liquid desiccant solutions. The impacts of operating conditions including LiCl solution feed temperature, osmotic agent concentration, and the circulation rates of LiCl solution feed and osmotic agent on water flux of the OMD process were also examined. Finally, a continuous OMD process with the LiCl solution feed at a mild operating temperature was conducted to elucidate the regeneration capacity of the OMD process.

2. MATERIALS AND METHODS

2.1. Materials

A lab-scale MD setup was used in this study. The setup could be operated in either DCMD or OMD configuration by switching the stream on the permeate side of the membrane from distillate to osmotic agent and vice versa. The setup consisted of three main components: a hot feed cycle, a membrane module, and a cold permeate (e.g. distillate or osmotic agent) cycle. The hot feed cycle was composed of a feed tank, a hot-water bath, and a pump for feed solution circulation. The cold permeate cycle had a permeate tank (containing either fresh distillate for DCMD or osmotic agent for OMD configuration), a cold-water bath, and a pump. The membrane module was consisted of two acrylic semi-cells which were engraved to form the flow channels for the feed and the permeate streams. These flow channels had a width, length, and height of 9.5 cm, 14.5 cm, and 0.3 cm, respectively. A hydrophobic microporous polyvinyl difluoride (PVDF) membrane coupon was inserted between the two semi-cells to separate the feed and permeate channels. The membrane was supplied by Porous Membrane Technology (Ningbo, China) and had pore size, porosity, and thickness of 0.2 µm, 80 %, and 120 µm, respectively. The active membrane surface area for water transfer was 138 cm². Plastic spacers were used in the feed and permeate channels to promote the flow turbulence near the feed and permeate membrane surface.



Figure 1. The schematic diagram of the lab-scale setup used in this study. The setup could be used in either DCMD or OMD configuration only by switching the permeate from distillate (deionized water) to osmotic agent (CaCl₂ solution).

During the DCMD or OMD process, the feed water from the feed tank was circulated through heat-exchanging coils submerged in the hot water bath for heating and then to the feed channel of the membrane module for evaporation. Inside the membrane module, the hot feed water evaporated to form water vapor at the membrane feed surface, and water vapor then diffused through the membrane pores to the permeate surface where it condensed to distillate in the DCMD process or absorbed by the osmotic agent in the OMD process. As water vapor transferred from the feed to the permeate, the feed water was concentrated and cooled. The concentrated water leaving the feed channel was returned to the feed tank. Similarly, on the permeate side of the membrane, cool distillate or osmotic agent from the permeate tank was pumped through submerged coils in the cold-water bath before circulating through the permeate channel of the membrane module. The cool stream condensed or absorbed water vapor that transferred through the membrane pores from the feed water. After leaving the permeate channel, the distillate or osmotic agent was returned to the permeate tank for the next operation cycle like the concentrated feed. The water temperatures at the inlet of the feed and permeate channels were measured using Pt100 temperature sensors, while the circulation rates of the feed and permeate streams were regulated using two flow meters (Figure 1). The temperatures of the feed water and permeate were controlled by a heater and a chiller, respectively.

Lab grade chemicals including lithium chloride (LiCl) and calcium chloride (CaCl₂) and deionized (DI) water were used for experiments in this study. The feed water was either DI water or 20 % LiCl solution. DI water was used as the initial distillate for the DCMD process while CaCl₂ solution was used as the osmotic agent in the OMD process. Indeed, other salt solutions such as NaCl, KCl, K₂CO₃, and MgCl₂ can be used as osmotic agents in the OMD application due to its reasonable cost and low water vapor pressure [12, 14]. The electrical conductivity of the obtained DI water was 12 μ S/cm. In all DCMD or OMD experiments, equal water circulation rates were applied for the feed and permeate streams to avoid membrane pore wetting due to excessive transmembrane hydraulic pressure difference. The initial volumes of the feed and permeate in all DCMD and OMD experiments were 1.5 L.

2.2. Analytical methods and experimental protocols

To assess the performance of the DCMD and OMD processes, water flux was recorded by measuring the change in the volume of the water in the feed tank (i.e. a 2.0 L graduated cylinder). As water transferred from the feed to the permeate, the volume of the feed water was reduced with the operating time. Water flux was then calculated as below:

$$J_{water} = \frac{\Delta V}{S \times \Delta t} \tag{1}$$

where water flux was in L/(m².h); Δt was the incremental time between two consecutive measurements (h); ΔV was the reduced volume of feed water in the incremental time, and S was the active membrane surface area of the membrane module (m²).

The DCMD and OMD experiments with DI water or the 20 % LiCl solution feed were conducted in this study. The experiments with DI water feed were to assess the water flux of the DCMD and OMD processes at different feed temperatures while permeate temperature (25 °C) and feed and permeate circulation rates (0.75 L/min, equivalent to cross flow velocities of 0.045 m/s) were remained unchanged. In the DCMD and OMD experiments with the 20 % LiCl solution feed, the feed temperature and feed/permeate circulation rates were changed to examine their effects on water flux of the DCMD and OMD processes while the permeate temperature was kept at 25 °C. The constant permeate temperature was applied in this study because it had a little effect on water flux of DCMD and OMD processes.

The salt leakage/diffusion from the osmotic agent to the feed during the OMD process was assessed in the experiment with the DI water feed. During this experiment, the electrical conductivity of the DI water feed was regularly monitored using a conductivity-pH meter (Hatch, USA). In the OMD process, salt leakage might occur when one or some membrane pores were wetted followed by the diffusion of $CaCl_2$ from the osmotic agent to the DI water feed, leading to an increase in the feed electrical conductivity. As a thin membrane was used in the OMD process, salt leakage would occur soon after starting the OMD experiment if the membrane was wetted.

2.3. Polarization effects during the DCMD and OMD processes

Polarization effects are common phenomena in all membrane filtration processes, including DCMD and OMD. During these processes, as water vapor and heat transfer from the feed to the permeate, thermal and concentration boundary layers are formed adjacent to the membrane surfaces on both sides of the membrane. The formations of these layers are defined as temperature and concentration polarization effects. A schematic diagram of heat and mass transfer and the resulting temperature and concentration polarization effects during the DCMD and OMD process is illustrated in Figure 2. As illustrated, temperature and concentration polarization effects during the DCMD polarization effects render the temperature difference and water vapor pressure difference between the membrane surfaces lower than those between the feed and permeate streams, hence negatively affecting the process water flux. Of a particular note, compared to DCMD, the OMD process suffers from additional concentration polarization effect on the permeate size of the membrane because the osmotic agent adjacent to the membrane surface is diluted by water transferred from the feed.



Figure 2. Schematic illustration of heat and mass transfer and polarization effects during the DCMD and OMD processes: T_{bf}, T_{mf}, C_{bf}, and C_{mf} are the temperature (T) and concentration (C) of feed water in the bulk and at the membrane surface, T_{mp}, T_{bp}, C_{mp}, and C_{bp} are the temperature and concentration of permeate (distillate in DCMD or osmotic agent in OMD) in the bulk and at the membrane surface.

3. RESULTS AND DISCUSSIONS

3.1. Baseline evaluation of the DCMD and OMD process with DI water feed

The results from the baseline evaluation of the OMD and DCMD processes with deionized (DI) water feed demonstrated the advantage of the former over the latter with respect to water flux under the same operating conditions (Figure 3). Under the same feed and permeate (i.e. distillate or osmotic agent) temperatures and circulation rates, the OMD process achieved discernibly higher water flux than the DCMD process. For example, at the feed and permeate temperatures of 40 °C and 25 °C and feed and permeate circulation rates of 0.75 L/min, the OMD process exhibited a water flux of 18.5 L/(m².h), which was 17 % higher than the water flux of the DCMD process (i.e. 15.8 $L/(m^2.h)$). When the DI water feed temperature was elevated to 70 °C, the water flux of the OMD process was 9 % higher than that of the DCMD process, increasing to 37.0 L/(m².h) compared to 34.1 L/(m².h) for the DCMD process. The higher water flux achieved by the OMD process was attributed to the reduced water vapor pressure on the permeate membrane surface induced by the high concentration of the $CaCl_2$ osmotic agent. It is necessary to emphasize that the driving force for the transfer of water vapor through the membrane in the OMD and DCMD process was the water vapor pressure difference between the two surfaces of the membrane. Under the same feed and permeate temperature, this water vapor pressure difference in the OMD process was higher than that in the DCMD process, hence resulting in improved water flux of OMD compared to the DCMD process.



Figure 3. Water flux of the OMD and DCMD processes with the DI water feed at different feed temperatures. The OMD and DCMD processes were operated at permeate temperature $T_{p.in} = 25$ °C, feed and permeate circulation rates $F_{f.in} = F_{p.in} = 0.75$ L/min. The 20 % CaCl₂ solution was used as the osmotic agent for the OMD process while DI water was used as the initial distillate for the DCMD process. The process was operated at each feed temperature for 1 hour after reaching the stable operation. Error bars represent the standard deviations of triple measurements.

The measurements of the electrical conductivity of the DI water feed confirmed that there was no observable salt leakage from the osmotic agent to the feed water during the OMD process. The electrical conductivity of the feed only increased slightly with the operating time of

the OMD process (Figure 4). This slight increase in the feed electrical conductivity was because of the concentrating effect of the feed as water vapor transferred from the feed to the osmotic agent. Unlike other membrane processes such as reverse osmosis (RO) and forward osmosis (FO), there was no liquid bridge between the feed and the osmotic agent in the OMD process provided that the membrane pores were dry. Thus, dissolved salt could not diffuse from the osmotic agent to the DI water feed when the membrane pores were not wet. The indiscernible increase in the feed electrical conductivity manifested that membrane pore wetting did not occur during the OMD process with the DI water feed and the 20 % CaCl₂ osmotic agent solution.



Figure 4. Electrical conductivity of the DI feed water during the OMD process with the operating time. The OMD process was operated at feed temperature $T_{f.in} = 70$ °C, permeate temperature $T_{p.in} = 25$ °C, feed and permeate circulation rates $F_{f.in} = F_{p.in} = 0.75$ L/min. The 20 % CaCl₂ solution was used as osmotic agent. Error bars represent the standard deviations of triple measurements.

3.2. The performance of the OMD and DCMD processes with the LiCl solution feed under various operating conditions

Water flux is a key performance index of the OMD and DCMD processes during the regeneration of liquid desiccant LiCl solution. Like what was observed with the DI water feed, water flux of the OMD process was noticeably higher than that of the DCMD process under the same operating conditions (e.g. LiCl feed temperature and concentration, permeate temperature, and feed and permeate circulation rates) (Figure 5). Moreover, water flux of both processes increased with the LiCl feed temperature due to the exponential relationship between water vapor pressure and temperature [17, 18]. Of a particular note, water flux of the OMD and DCMD processes with the LiCl solution feed was much lower compared to that observed during the baseline evaluation when DI water was used as the feed. This demonstrated the profound impact of the salinity on the rate of water transfer through the membrane in the OMD and DCMD regeneration of the LiCl solution feed.



Figure 5. Influences of the operating feed temperature on the water flux of the OMD and DCMD processes with the 20 % LiCl solution feed. Other operating conditions: permeate temperature $T_{p,in} = 25$ °C, feed and permeate circulation rates $F_{f,in} = F_{p,in} = 0.75$ L/min. The 20 % CaCl₂ solution was used as the osmotic agent for the OMD process while DI water was used as the initial distillate for the DCMD process. Error bars represent the standard deviations of triple measurements.

The advantage of the OMD over the DCMD process with respect to water flux for the regeneration of the liquid desiccant LiCl solution is magnified when these processes are operated at high water circulation rates. As demonstrated in Fig. 6, elevating water circulation rates resulted in an increase in water flux of the OMD and DCMD processes to different extents. For example, when feed and permeate circulation rates were elevated from 0.5 to 1.0 L/min, water flux of the DCMD process increased by 36.5 % from 11.2 L/(m^2.h) to 15.3 L/(m^2.h) , while the OMD process exhibited a water flux increase by 61.9 % from 13.4 L/(m².h) to 21.7 L/(m².h). The difference in water flux increase resulting from elevating water circulation rates between the OMD and DCMD processes could be attributed to the different magnitude of the concentration polarization effects that occurred during these processes. As illustrated in Figure 2, while concentration polarization occurred only on the feed side of the DCMD process, the OMD process suffered from the concentration polarization effect on both the feed and osmotic agent sides. Elevating feed and permeate circulation rates helped promote the flow turbulence near the feed and permeate membrane surfaces and hence mitigated the negative impact of concentration polarization on water flux. As a result, water flux increased to a higher extent for the OMD process as compared to the DCMD process when raising the feed and permeate circulation rates.

Water flux of the OMD process during the regeneration of liquid desiccant LiCl solution could also be increased by using more concentrated osmotic agent. Indeed, water flux of the OMD process with the 20 % LiCl solution feed linearly increased with the concentration of the CaCl₂ osmotic agent (Figure 7). Of a particular note, water flux of the OMD process increased by 48 % from 17.8 L/(m².h) to 26.4 L/(m².h) when the osmotic agent concentration was elevated from 20 % to 35 %. Moreover, compared with the DCMD process at the same operating temperature and circulation rates, the OMD process with the 35 % CaCl₂ osmotic agent exhibited a nearly double water flux (i.e. 26.4 L/(m².h) compared to 13.9 L/(m².h)). High water flux achieved by using more concentrated osmotic agent would enable the OMD process to

regenerate LiCl solutions of higher concentration without the need for operating the process at excessively high LiCl solution feed temperature. This advantage of the OMD process is vital for the regeneration of liquid desiccant solutions used in LDAC systems as solar thermal energy can be coupled with the OMD regeneration of liquid desiccant solution to reduce the energy cost of the LDAC systems.



Figure 6. The influences of feed and permeate circulation rates on the water flux of the OMD and DCMD processes. Operating conditions: feed temperature $T_{f.in} = 70$ °C, permeate temperature $T_{p.in} = 25$ °C. The 20 % CaCl₂ solution was used as the osmotic agent for the OMD process while DI water was used as the initial distillate for the DCMD process. Error bars represent the standard deviations of triple measurements.



Figure 7. Water flux of the OMD process with the 20 % LiCl solution feed when using the CaCl₂ solution as the osmotic agent at different concentrations. Operating conditions: feed temperature $T_{f.in}$ = 70 °C, permeate temperature $T_{p.in}$ = 25 °C, feed and permeate circulation rates $F_{f.in} = F_{p.in} = 0.75$ L/min.

3.3. Continuous OMD regeneration of the liquid desiccant LiCl solution

The OMD process was capable of concentrating the LiCl solution up to 38.4 % during a continuous 8-hour operation (Fig. 8). During the continuous OMD regeneration of the LiCl feed solution, the process water flux gradually decreased from the initial value of 17.8 L/(m^2 .h) to nearly zero while the LiCl feed solution increased from 20 % to 38.4 %. The decreased water flux with the operating time observed during this continuous OMD process was attributed to a dual effect: the concentration of the LiCl solution feed and the dilution of the CaCl₂ osmotic agent. This dual effect narrowed the water vapor pressure difference between the two sides of the membrane, thus resulting in the decreased water flux with the operating time. When the LiCl feed concentration reached 38.4 % (and the CaCl₂ osmotic concentration reduced to 13.5 %), the transmembrane water vapor pressure difference was neutralized; therefore, water flux approached zero (Figure 8).



Figure 8. Water flux and the LiCl feed concentration during the continuous OMD regeneration of the LiCl solution feed. Operating conditions: initial LiCl feed concentration = 20 %, LiCl feed temperature = 70 °C, initial CaCl₂ osmotic agent concentration = 20 %, permeate temperature $T_{p.in}$ = 25 °C, feed and permeate circulation rates $F_{f.in}$ = $F_{p.in}$ = 0.75 L/min.

The results reported here demonstrated the great advantage of the OMD over the DCMD process for the regeneration of liquid desiccant solutions including LiCl solutions. As previously reported by Duong *et al.* [3], a DCMD process using fresh water as the distillate could increase the LiCl concentration only to 29 % when operated at the feed temperature of 65 °C. The OMD process at the LiCl feed temperature of 70 °C investigated in this study raised the LiCl concentration to 38.4 %. Liquid desiccant LiCl solutions of even higher concentrations could be regenerated by the OMD process when using the osmotic agent of higher CaCl₂ concentrations. However, further studies are required to elucidate the issues associated with the concentration polarization effects and membrane fouling/scaling.

4. CONCLUSIONS

In this study, a proof of concept of OMD regeneration of liquid desiccant solutions used for LDAC systems was demonstrated. The OMD process utilized a concentrated CaCl₂ solution as an osmotic agent on the permeate side of the membrane to boost the water vapor pressure difference across the membrane, thus increasing water flux. Compared to the DCMD process whereby distillate was circulated on the permeate side of the membrane, the OMD process exhibited a water flux two times higher than that of the DCMD process under the same operating conditions (e.g. LiCl solution feed temperature and concentration, and feed and permeate circulation rates). Moreover, the water flux of the OMD process with the LiCl solution feed was increased by elevating the feed temperature, feed and permeate water circulation rates, and particularly the concentration of osmotic agent. Given its increased water flux, the OMD process with the initial 20 % CaCl₂ solution osmotic agent concentrated the LiCl solution up to 38.4 %, manifesting its enhanced capacity for the regeneration of liquid desiccant solutions when comparing with the DCMD process.

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Declaration of competing interest. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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