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Desalination in Cross-flow Vacuum Membrane Distillation under the Negative Membrane Pressure Difference

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Abstract: This study demonstrates desalination using cross-flow Vacuum Membrane Distillation (VMD) under negative membrane pressure differences. The membrane pressure difference is defined as the partial pressure difference of water vapour between the upstream membrane surface and the downstream membrane surface. The negative membrane pressure differences are resulted from the low feed temperatures (40-60°C) and the high downstream pressures (14.5-40 kPa). The experimental results showed that positive fluxes (2-3.6 kg m⁻² h) take place although the system is operated under the negative membrane pressure differences. The humidity on upstream side higher than the humidity on downstream side allows the water vapour transfers from the feed to the permeate sides. Relatively high feed temperatures and relatively low downstream pressures resulted in high enthalpy differences and hence high fluxes. The hydrostatic pressure on the feed circulation loop contributed a minor effect on the total flux when membrane pore wetting takes place. The rejection of salt under these conditions was at least 96%.

Key words: Vacuum membrane distillation, desalination, negative membrane pressure difference, enthalpy difference, hydrostatic pressure

INTRODUCTION

Phase change is the common characteristic of Membrane Distillation (MD) process in which only vapour(s) are thermally driven through a porous hydrophobic membrane. The application of MD has been examined in producing the ultrapure water from salt solutions (Khayet and Cojocar, 2012; Qtaishat and Banat, 2013), concentrating the juices (Kozak *et al.*, 2009; Jensen *et al.*, 2011), organic extractions (Hasanoglu *et al.*, 2012) and ammonia removal (Sarbatly and Chiam, 2012). In MD, an aqueous solution is kept in direct contact with one of the membrane surface and a hydrostatic pressure of the solution must not exceed the 'liquid entry pressure of water (LEP_w)' of the membrane. Because of the membrane material is water repellent and no liquid penetration inside the pores, a liquid-vapour (L-V) interface is formed at the membrane pore entrances. Inside the pores, the hydrophobic characteristic of the pore wall is able to inhibit the vapour from condensation.

It was quoted that direct contact MD (DCMD) is the most studied configuration which occupied more than 60% of the MD studies (Khayet, 2010). In ultrapure water

production, however, one of the major drawbacks in DCMD is that a negative flux results from a negative pressure drop when pure water is brought into direct contact with the downstream side of the membrane (Lagana *et al.*, 2000). On the upstream side, the partial pressure of water decreases with the concentration of solute that dissolved in the feed solution. Up to a level of the solute concentration, an osmotic pressure due to the pure water on the downstream side of the membrane can exceed the osmotic pressure of the feed solution on the upstream side of the membrane. Unlike the reverse osmosis process, additionally, the MD is preferable operates at atmospheric pressure.

The osmotic pressure is eliminated when a stagnant air gap, a cold inert sweeping gas or a vacuum space is applied on the MD downstream side; which these configurations are respectively termed as air gap MD (AGMD), sweeping gas MD (SGMD) and vacuum MD (VMD). Similar with the DCMD, the driving force across the membrane in the AGMD, SGMD and VMD is the partial pressure difference corresponding to the temperature and composition at the both sides of membrane surfaces. Compared to the DCMD, however, a

positive flux may be obtained in the AGMD, SGMD and VMD when the enthalpy of water vapour at the membrane upstream surface is higher than that at the membrane downstream surface even the membrane pressure difference is negative.

In the case of VMD configuration, for a modelling study of a single component transport process such as desalination assuming that the partial pressure of water vapour on the membrane downstream surface equals the absolute pressure of vacuum. The liquid water evaporates at the L-V interface due to the latent heat of vaporization. On the downstream compartment where vacuum is applied, the moist of air is relatively low. The water vapour transports from the L-V interface to the vacuum space through the membrane because of the humidity gradient exists. The enthalpy of water vapour near the L-V interface is higher than the enthalpy of water vapour in the downstream compartment. In a non-isothermal process like VMD, the water vapour condenses back to liquid water onto a surface inside the downstream compartment where the latent enthalpy absorbed during the evaporation is released as sensible heat onto the surface.

In a cross-flow VMD system, the feed solution flows parallel to the membrane surface and then both concentrated solution (rejection) and vapour streams leave the system. The rejection is circulated on the upstream membrane side and the vapour is collected into a condenser. A hydrostatic pressure of the feed solution may be created in the circulation loop on the upstream side during the operation. However, the hydrostatic pressure may have no effect on producing water vapour. For the hydrophobic membrane, water vapour is the only component can transport through the membrane in the desalination process. As a result, 100% of the salts are rejected. On the other hand, when the hydrostatic pressure slightly exceeds the LEP_w of the membrane during the operation, the liquid feed begins to penetrate the largest pores and then permeate through the membrane (Lawson and Lloyd, 1997). Under this condition, not only the water vapour but the liquid may also contribute to the total flux in the system when the membrane pressure difference is negative.

The objective of this research is to study the effect of the low feed temperatures and the high downstream pressures on the cross-flow VMD fluxes. The process conditions are selected in order to observe the influence of membrane pressure difference on the fluxes.

THEORY

In the present study of the desalination process, the membrane pressure difference is defined as the partial

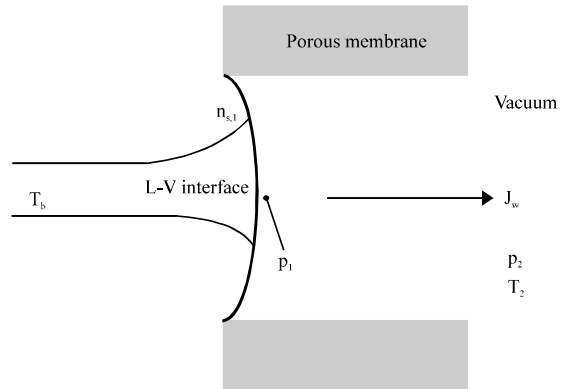


Fig. 1: A schema of typical VMD configuration

pressure difference of water vapour at the membrane upstream surface and the membrane downstream surface. A schema of typical VMD configuration illustrated in Fig. 1, the partial pressure difference of water vapour across the membrane (Δp_m) is calculated as follows:

$$\Delta p_m = p_1 - p_2 \tag{1}$$

where, p_1 and p_2 are the partial pressure of water vapour on the feed (upstream) membrane surface and the partial pressure of water vapour on the permeate (downstream) membrane surface, respectively. In this study, the feed temperatures ranging from 40 to 60°C operate at downstream pressure of 14.5 kPa and the downstream pressures ranging from 14.5 to 40 kPa operate at feed temperature of 60°C, are selected in order to obtain negative values of Δp_m . The VMD operation is operated in laminar flow rates ($700 < Re < 1000$).

The saline water is a non-ideal binary mixture; thus, the partial pressure on the feed side at the membrane surface is described as follows:

$$p_1 = (1 - n_{s,1}) \gamma_w P_w^\circ \tag{2}$$

where, $n_{s,1}$ is the mole fraction of salt solute at the membrane surface, γ_w is the activity coefficient of water and P_w° is the saturation pressure of pure water. The activity coefficient of water in the saline water can be determined as follows (Gryta, 2002):

$$\gamma_w = 1 - 0.5n_{s,1} - 10n_{s,1}^2 \tag{3}$$

The saturation pressure of pure water can be described by the Antoine equation:

$$P_w^\circ = \exp \left[23.1964 - 3816 \frac{44}{T_1 - 46.13} \right] \tag{4}$$

where, T_1 is the feed/membrane interface temperature.

Because of the water vapour at the L-V interface is saturated and dissolved gases content is too low where the relative humidity is almost equals 100%, the partial pressure on the membrane surface of the permeate side is approximated to the absolute pressure of vacuum applied on the downstream side (P_v):

$$p_2 \cong P_v \quad (5)$$

Therefore, the driving force due to the partial pressure difference of water vapour can be calculated as follows:

$$\Delta p_m = (1 - n_{s,i}) \cdot (1 - 0.5n_{s,i} - 10n_{s,i}^2) \cdot \exp \left[23.1964 - 3816 \cdot \frac{44}{T_1 - 46.13} \right] - P_v \quad (6)$$

The evaporation of water at the L-V interface reduces the temperature and increases the salt concentration at the membrane surface; these phenomena are respectively called temperature and concentration polarizations. The temperature and concentration polarization effects are described by heat and mass transfer coefficients, respectively. The heat and mass balance equations in the VMD process are, respectively expressed as follows (Mericq *et al.*, 2010; Alkhudhiri *et al.*, 2012):

$$J_w \Delta H_w = h_f (T_b - T_1) \quad (7)$$

and:

$$J_w M_w = \rho_f k_s \ln \frac{n_{s,i}}{n_{s,b}} \quad (8)$$

where, J_w , ΔH_w , M_w , ρ_f , h_f , k_s , T_b and $n_{s,b}$ are the water vapour flux, heat of vaporization, water molecular weight, density of feed in the bulk phase, heat transfer coefficient, mass transfer coefficient of salt, temperature of the feed in the bulk phase and molar fraction of salt in the bulk phase, respectively.

The temperature and molar fraction of salt at the upstream membrane surface can be calculated from the Nusselt ($Nu = h_f d_f / k$) and Sherwood ($Sh = k_s d_f / D_s$) numbers. These numbers can be given by the Sider-Tate empirical correlation (Khayet, 2010; Alkhudhiri *et al.*, 2012):

$$Nu = 1.86 \cdot (Re \cdot Pr \cdot d_f / L)^{1/3} (\mu_1 / \mu_b)^{0.14} \quad (9)$$

$$Sh = 1.86 \cdot (Re \cdot Sc \cdot d_f / L)^{1/3} (\mu_1 / \mu_b)^{0.14} \quad (10)$$

where, d_b , L , μ_1 and μ_b are, respectively the hydraulic diameter, length of feed flow channel, water viscosity at feed/membrane interface and bulk feed viscosity. The Re , Pr and Sc are Reynolds, Prandtl and Schmidt numbers, respectively. These numbers are evaluated at the bulk temperature. The correction factor in the right side of the Eq. 9 and 10 is employed for there is a significant temperature difference between the bulk and the membrane surface.

In order to estimate the temperature and molar fraction of salt at the upstream membrane surface, the Re and Pr are first estimated at the bulk feed temperature, T_b and the heat transfer coefficient, h_f is evaluated using Eq. 9. The bulk feed temperature is calculated as the mean temperature between the temperatures at the inlet and outlet membrane module. Next, from Eq. 7 the feed/membrane interface temperature, T_1 , is obtained. New values of Re and Nu numbers are calculated at the mean temperature ($(T_b + T_1) / 2$). Then, the heat transfer coefficient is calculated again from Eq. 9 and then the T_1 from Eq. 7. The procedure is repeated until the difference between two successive calculated feed/membrane interface temperatures, T_1 , is less than 0.01%. By this method, the Temperature Polarization Coefficient (TPC) is evaluated according to $TPC = T_1 / T_b$. The μ_1 in Eq. 10 is calculated based on the final value of T_1 . The obtained k_s is substituted in Eq. 8 and the $n_{s,i}$ is then estimated. The Concentration Polarization Coefficient (CPC) can be estimated according to $CPC = n_{s,i} / n_{s,b}$.

EXPERIMENTAL SECTION

Membrane and membrane module: A 0.2 μm pore size hydrophobic porous flat membrane made of polyvinylidene fluoride supplied by Whatman GmbH, Germany (Westran S PVDF) was used in this study. The LEP_w of the membrane was measured to be approximately 142 kPa. An aluminium membrane module was fabricated in our lab and used in this study. The module has a rectangular feed flow channel with a dimension ($H \times L \times W$) of $13 \times 102 \times 70$ mm³ and therefore the effective membrane area was 71.4 cm². The module consists of two detachable compartments, one is the upstream compartment and another is downstream compartment. The membrane together with a rubber gasket was sandwiched between the compartments and the compartments were attached tightly by using bolts and nuts. Inside the downstream compartment, beneath the membrane a perforated aluminium support was embedded to prevent the membrane from breakage.

Vacuum membrane distillation (VMD) experiments: The experimental VMD apparatus used in this study is shown

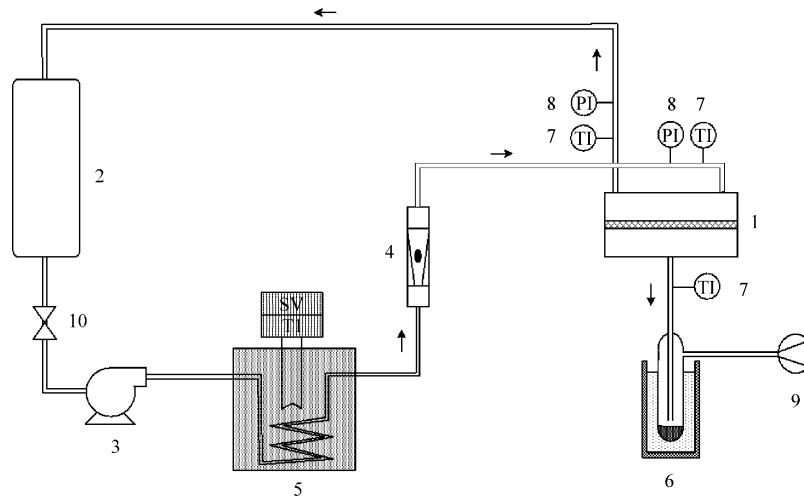


Fig. 2: Schematic diagrams of the experimental VMD apparatus; 1: Module with a flat membrane, 2: Feed reservoir, 3: Feed pump, 4: Rotameter, 5: Temperature control bath with heating coil, 6: Liquid nitrogen trap, 7: Temperature transmitters, 8: Pressure gauges, 9: Vacuum pump and 10: Feed valve

in Fig. 2. The feed solution was pumped using a digital gear pump (Micropump Drives 75211-35 and Pump head 07003-04, Cole-Parmer Instrument Co., Illinois, USA) through a heating circulation bath (Model 9012A12E, Polyscience, Niles, IL USA) and then it entered the membrane module. A vacuum on the downstream side was created by a vacuum pump (Rocker 300, Today’s Instruments Co. Ltd., Taiwan) which was equipped with a vacuum gauge pressure. The feed temperature was kept constant by using a temperature controller equipped in the heating circulation bath. A cold trap (Z225789, Sigma Aldrich Corp., St. Louis, MO USA) immersed in liquid nitrogen and housed in a dewar (Z225770, Sigma Aldrich Corp., St. Louis, MO USA) was used to condense the water vapour permeates.

The feed solution was a mixture of NaCl and distilled water. The concentration of NaCl in all samples was determined using a conductivity meter (CON 1500, Eutech Instrument Pte Ltd., Ayer Rajah Crescent, Singapore). Before starting the experiments, the VMD system only allowed the feed streamline to circulate for about 40 min for preheating purpose. When the feed temperatures on the inlet streamline, the outlet streamline and the feed tank reached a steady state condition, the VMD operation was started. Each of the experiments was operated for 1 h. The experimental flux of water vapour ($J_{w,exp}$) for every experiment was calculated by the following equation:

$$J_{w,exp} = \frac{m}{A.t} \quad (11)$$

where, m is the total mass of water vapour that permeates through the membrane, A is the effective membrane area and t is the operation time.

RESULTS AND DISCUSSION

Effect of feed temperature: The effect of feed temperature on desalination by VMD was studied in this work by conducting different experiments at different feed temperatures (T_f : 40-60°C). The feed flow rate was 1.5 L min⁻¹ and the corresponding Re ranging from 714 to 861, the NaCl concentration was 3.5-wt% and the downstream pressure was constant at 14.5 kPa. The TPC was ranging from 0.88 to 0.91 and the CPC was 1.002.

The experimental water vapour flux ($J_{w,exp}$) through the membrane for different conditions corresponding to the membrane pressure difference is presented in Fig. 3. The membrane pressure difference was calculated based on Eq. 6. The Fig. 3 shows the positive fluxes were obtained even the processes were operated under negative membrane pressure differences. This could be attributed to the partial pressure differences of water vapour did not play an essential role in controlling the fluxes when the bulk feed temperatures were below 60°C and downstream pressure of 14.5 kPa in this system. The conductivity of the permeated water was between 0.90 and 1.20 mS cm⁻¹ which were larger than the conductivity of distilled water (<2 μS cm⁻¹). This could be probably the hydrostatic pressure built in the feed circulation loop wetted the larger

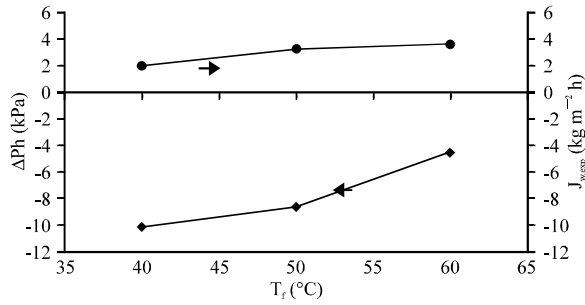


Fig. 3: Effect of feed temperatures on water vapour flux. Re: 714-861; downstream pressure = 14.5 kPa and feed concentration = 3.5-wt% NaCl

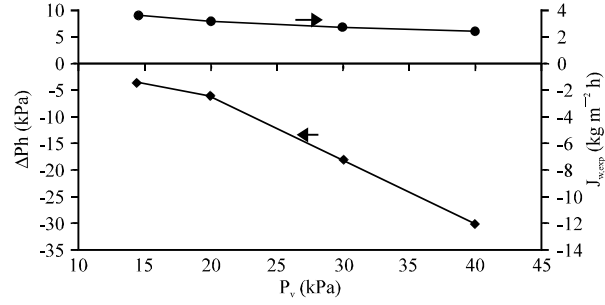


Fig. 4: Effect of downstream pressure on water vapour flux. Re: 842-924; feed temperature = 60°C and feed concentration = 3.5-wt% NaCl

membrane pores during the VMD process and the wetted pores allow the saline liquid transported through the membrane.

Additionally, the water vapour transferred through the porous membrane could also due to the enthalpy difference between the upstream and the downstream sides. The enthalpy on the upstream side was higher than the enthalpy on the downstream side permitted the water vapour transferred from the feed to permeate side. In other words, the upstream side is more humid than the downstream side. Under these conditions, the water vapour flux increased with the feed temperature because the enthalpy difference increased. The hydrostatic pressure at the membrane surface was able to push the water vapour through the porous membrane because of the suction effect by the vacuum pump on the downstream side. By using a calibrated curve (conductivity, mS cm⁻¹ versus NaCl concentration, wt%), the salt rejection in this system was at least 98%.

Effect of downstream pressure: The effect of pressure of the membrane downstream side was investigated by performing different experiments each at a specified vacuum pressure on the downstream compartment while the other process conditions remained to be constant; feed flow rate was 1.5 L min⁻¹ and the corresponding reranging from 842 to 924 (different Re was because of the instable heat loss of the feed bulk temperatures), feed solution concentration was 3.5-wt% and feed temperature of 60°C. The selected downstream pressures were 14.5, 20, 30 and 40 kPa. The TPC was ranging from 0.90 to 0.93 and the CPC was 1.002.

The results are shown in Fig. 4 in terms of the experimental water vapour flux and the calculated partial pressure difference of water vapour at the corresponding downstream pressure. The positive fluxes obtained experimentally implied that the VMD processes were driven by the enthalpy differences of water vapour

because the processes were operated under negative membrane pressure differences. The hydrostatic pressure might also contribute to the total flux because the permeated water quality was ranging 1.15 to 2.21 mS cm⁻¹, however, the effect of this pressure was minor when compared to the enthalpy difference. At a higher downstream pressure, the enthalpy of water vapour on the downstream compartment was higher because the humid air content was high. Consequently, the enthalpy difference decreased with increasing the downstream pressure. However, the rate of humidification on the downstream side or water vapour flux through the membrane increased with decreasing the downstream pressure. The salt rejection under these conditions was at least 96%.

CONCLUSION

In VMD desalination process, a positive flux can be obtained even though the process is worked under a negative membrane pressure difference. In this case, a humidity difference replaces the partial pressure difference to be the driving force of the separation process. The water vapour flux through the membrane increases with the feed temperatures because the humidity on the upstream side increases correspondingly. When the downstream pressures decrease, the fluxes increase as well because the humidity on the downstream side decreases. In addition, when membrane pore wetting occurs in the system where feed liquid is allowed to transport through the membrane, the hydrostatic pressure built in the feed circulation loop will be a considerable factor in contributing the total flux.

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NOMENCLATURE

d = Diameter (m)
D_s = Diffusion transfer coefficient (m² sec⁻¹)
h = Heat transfer coefficient (W m⁻² K)
H = Height (m)
ΔH = Heat of vaporization (J kg⁻¹)
J = Flux (kg m⁻² sec or kg m⁻² h)
k = Thermal conductivity (W m⁻² K)
k_s = Mass transfer coefficient (m sec⁻¹)
L = Length (m)
M = Molecular weight (kg mol⁻¹)
n = Molar fraction (-)
Nu = Nusselt number (-)
p = Partial pressure (Pa)
P = Absolute pressure (Pa)
Δp = Partial pressure difference (Pa)
P^o = Vapour pressure (Pa)
Pr = Prandtl number (-)
Re = Reynolds number (-)
T = Temperature (K)
W = Width (m)
b = Bulk
exp = Experimental
f = Feed
h = Hydraulic
m = Membrane surface
s = Solute
v = Vacuum
w = Water
1 = Upstream membrane side
2 = Downstream membrane side
γ = Activity coefficient (-)
μ = Viscosity (Pa s)
ρ = Density (kg m⁻³)

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