Performance evaluation of ePTFE and PVDF flat-sheet module direct contact membrane distillation

Ching-Jung Chuang, Kuo-Lun Tung, Yang-Hsiang Fan, Chi-Dong Ho and James Huang

ABSTRACT

This paper reports experiments using a flat-sheet module with 0.18 – 0.45 μm ePTFE (expanded polytetrafluoroethylene) and PVDF (polyvinylidene fluoride) membranes to show the effects of membrane properties, salt concentration and fluid hydrodynamics on the permeate flux and salt rejection of DCMD (direct contact membrane distillation). A theoretical prediction of the permeate flux was carried out, and was in close agreement with the experimental results. In addition, the energy integration of the process was also analyzed in order to evaluate module design to increase energy efficiency. According to the simulated results of the energy integration design, a combination of simultaneous cooling of the permeate stream and an additional heat exchanger to lower the temperature of the permeate stream not only enhances the MD flux, but also reduces energy consumption.

Key words | DCMD, desalination, energy integration design, ePTFE, membrane distillation, PVDF

INTRODUCTION

Membrane distillation (MD) is a process based on the transport of vapour through the pores of hydrophobic membranes followed by condensation on the downstream side of the membrane. The vapour driving force is provided by a temperature difference across the membrane, and the hydrophobic nature of the membrane prevents penetration of the aqueous liquid solution into the pores. This results in the formation of a vapour-liquid interface at the pore entrance. If both the heated feed and cold permeate streams are liquids and kept in direct contact with the membrane, the configuration is known as direct contact membrane distillation (DCMD), the most common and versatile configuration for membrane distillation. Because DCMD does not need an external condenser, it is suitable for applications in which the volatile component is water (Lawson & Lloyd 1996).

MD has been known for over 40 years as an alternative approach for producing highly pure water, used mainly for desalination. Because it has the potential to work at low temperatures using low-grade or waste energy, it offers many advantages over traditional distillation processes. Despite this great potential, however, the process is still not a commercial water desalination process. This is partially due to its relatively lower flux compared to the reverse osmosis (RO) process, which is the case because of the unavailability of membranes with the most suitable characteristics for the process (Alklaibi & Lior 2004; El-Bourawi et al. 2006).

The MD process involves a coupled mass and heat transfer transport stepped from the feed side to the membrane and the permeate side, as shown in Figure 1. The flux depends on membrane characteristics such as porosity, pore size and thickness, as well as on the temperature difference between the two sides and the flow geometry in the membrane module. Many researchers have studied the effects of the operating parameters on the flux through the membrane and/or the heat efficiency, which is defined as the ratio between the heats contributed to
evaporation and the total heat exchanged through the membranes. Few researchers have considered the heat efficiency of MD when heat recovery or external heat exchangers are applied to cool the permeate streams. This work analyzes a flat-sheet DCMD module with ePTFE and PVDF membranes theoretically and experimentally in order to study the effects of membrane pore size, membrane thickness and flow channel spacer on the permeate flux. Then, a new DCMD configuration is presented based on the concept of cooling the permeate side stream to enhance flux and energy efficiency, and is also analyzed by simulation.

**THEORY**

**Heat transfer**

Membrane distillation is a physical process involving both heat and mass transfer. The heat transfer can be described in three steps (Figure 1): 1) convective transfer of heat from the bulk feed to the membrane surface \( (Q = h_f(T_f - T_{m1})) \), 2) heat transfer through the membrane in the form of heat conduction and vapour latent heat \( (Q = h_m(T_{m1} - T_{m2})/\delta + J_v\Delta H_v) \), where only the latent heat of evaporation is the heat used effectively and the heat transfer by conduction is considered as heat lost, and 3) convective transfer from the membrane to the bulk permeate stream \( (Q = h_p(T_{m2} - T_p)) \).

Based on these heat transfer resistances in series, the steady state temperatures at the membrane-liquid interface can be derived as Termplyakul et al. (2005) and Srisurichan et al. (2006):

\[
T_{m1} = T_f - \frac{J_v\Delta H_v + h_m(T_{m1} - T_{m2})/\delta}{h_f}
\]

\[
T_{m2} = T_p + \frac{J_v\Delta H_v + h_m(T_{m1} - T_{m2})/\delta}{h_p}
\]

where \( h \) is the heat transfer coefficient (generally obtained by empirical relationships in terms of the Reynolds number \( R_e \), Prandtl number \( P_r \) and flow channel geometry) (Phattaranawik et al. 2003a), \( \delta \) is the membrane thickness, \( h_m \) is the thermal conductivity of the membrane, and \( J_v \) and \( \Delta H_v \) denote the vapour mass flux through the membrane and the latent heat of vapour, respectively.

**Mass transfer**

The membranes used for mass transfer of vapour in MD generally have a pore size ranging from 0.1 to 0.5 \( \mu \)m, with temperature between 20 and 80°C. Therefore, both molecular diffusion and Knudsen diffusion play important roles in transporting vapours through the feed membranes

Figure 1 | Mass and heat transfer resistance in DCMD.
membranes. The following equation has been derived for mass transfer flux through a porous membrane (Phattaranawik et al. 2003b):

\[
J_v = \frac{D_{ei} M P_{m1} - P_{m2}}{\chi \delta \frac{RT}{(P - P_m)_{ln}}} \tag{3}
\]

where \(D_e\) is the effective mass diffusion coefficient, calculated by the following relationship of the Knudsen diffusion coefficient, \(D_k\), and the molecular diffusion coefficient, \(D\), as:

\[
\frac{1}{D_e} = \frac{1}{D_k} + \frac{1}{D} \tag{4}
\]

In Equation (3), \(\chi\) is the tortuosity factor of the membrane, \(\varepsilon\) is the porosity of the membrane, \(M\) the molecular weight of vapour, \(R\) the gas constant, \(T\) the average temperature inside the pore, \(P_m\) the vapour pressure at the membrane-liquid interface, \(P\) the total pressure in the pore and \((P - P_m)_{ln}\) denotes the log mean vapour pressure difference of \((P - P_{m1})\) and \((P - P_{m2})\).

Making use of the transport models mentioned above and the iterative proceeding shown in the flow chart of Figure 2, the permeate flux can be predicted for given experimental conditions if the membrane characteristic parameters, mass diffusion coefficients in membrane and heat transfer coefficients are known. In addition, the method also allows the evaluation of the heat efficiency defined as the ratio between the latent heat flow accompanying vapor flux and the total heat across the membrane (i.e. the total heat exchanged by the feed).

**EXPERIMENTAL**

A flow chart describing the DCMD experiment in this work is schematically presented in Figure 3. A plate-and-frame membrane module 146 mm long and 90 mm wide was used to perform DCMD with a permeation section of 0.0091 m². The flow channels on both the feed and permeate sides had the same clearance of 3 mm, and in some of the experiments, inserts were placed in the channel to reduce the temperature polarization at the membrane-liquid interface. The permeate flux was obtained from the weight of water overflow from the constant-level cooling tank.

Two different flat sheet membranes, ePTFE and PVDF, which are frequently used in membrane distillation applications, were used in this study and their specifications are shown in Table 1. One of the ePTFE membranes was provided by our Membrane Technology Centre (CMT) and other membranes used were supplied by the Millipore Inc.

Experiments were performed using aqueous NaCl solutions (0, 30,000 and 60,000 mg/L), and the hot feed and permeate liquid were pumped tangentially to the membrane from thermostatic reservoirs at circulation rates of 6 and 8 L/min, respectively. The temperatures of hot feed were controlled in a range of 30 to 70°C, and

**Table 1** | Membrane properties

<table>
<thead>
<tr>
<th>Material</th>
<th>Pore size (µm)</th>
<th>Porosity, (\varepsilon)</th>
<th>Thickness, (\delta) (µm)</th>
<th>Thermal conductivity, (k_m) (Wm⁻¹K⁻¹) (Phattaranawik et al. 2003a)</th>
<th>Supplier</th>
</tr>
</thead>
<tbody>
<tr>
<td>ePTFE</td>
<td>0.20</td>
<td>0.91</td>
<td>60</td>
<td>0.0507</td>
<td>Millipore</td>
</tr>
<tr>
<td>ePTFE</td>
<td>0.45</td>
<td>0.90</td>
<td>60</td>
<td>0.0310</td>
<td>Millipore</td>
</tr>
<tr>
<td>ePTFE</td>
<td>0.18</td>
<td>0.90</td>
<td>55</td>
<td>0.0310</td>
<td>Millipore</td>
</tr>
<tr>
<td>PVDF</td>
<td>0.20</td>
<td>0.85</td>
<td>200</td>
<td>0.0393</td>
<td>Millipore</td>
</tr>
<tr>
<td>PVDF</td>
<td>0.45</td>
<td>0.75</td>
<td>140</td>
<td>0.0412</td>
<td>Millipore</td>
</tr>
</tbody>
</table>
the permeate side was maintained at 20°C to provide the temperature difference between hot feed and permeate liquid ranging from 10 to 50°C in the experiments.

RESULTS AND DISCUSSION

Comparison of DCMD permeability between ePTFE and PVDF membranes

The influence of the temperature difference between the water feed side and the 20°C permeate side on the vapour flux through 0.2 μm ePTFE and PVDF membranes is shown in Figure 4. EPTFE membranes clearly provide a much higher MD flux than the PVDF membranes. This is mainly due to the different active-layer thickness and porosity of the two membranes. A thinner active layer and higher porosity leads to lower mass transfer resistance. Figure 5 shows a comparison between the MD flux of commercial ePTFE and that of the membrane prepared by our membrane centre. Both membranes have very similar pore size and porosity characteristics (Table 1). The higher flux obtained using the ePTFE membrane from CMT results from its thinner active layer, which reduces the mass transfer resistance.

A comparison of the experimental and predicted fluxes of pure water and 30,000 ppm NaCl solution with 0.45 μm ePTFE and PVDF membranes is shown in Figure 6, where the curves represent the predicted flux, and the symbols represent the experimental data at the corresponding temperatures. The results indicate that the 30,000 ppm aqueous NaCl solution has a flux only about 10% less than that of pure water. The salt rejection measured for both membranes was greater than 99.8%. In accordance with theoretical predictions, a membrane tortuosity factor of 2.0 was used, which has been widely supported by many researchers (Schofield et al. 1990). Clearly, the predicted values are in good agreement with the experimental data.

DCMD configuration with a cooling stream on the permeate side

The driving force for vapour flux in DCMD is the temperature difference across the membrane. The traditional configuration of DCMD generally uses an external heat...
exchanger to cool the permeate stream. This study proposed a different configuration to increase the temperature difference across the membrane by decreasing the temperature of the permeate stream. As shown in Figure 7, the permeate stream flowing through the membrane cell was simultaneously cooled by a cooling water stream, and the permeate stream leaving the membrane cell was further cooled by an external heat exchanger to reach the temperature given for the permeate stream entering the MD cell. Based on the theoretical models and iterative method mentioned above, the proposed process was simulated by taking into account the rate of heat transfer from permeate side to the cooling side. The parameters used for the process simulation are shown in Table 2. With a feed temperature of 80°C and a permeate stream temperature of 30°C, the calculated outlet temperatures of the feed and permeate streams are also shown in Figure 7. The calculated flux was 75.99 kg/m²hr, which is larger than the value of 71.63 kg/m²hr calculated for the process without simultaneous cooling of the permeate stream. Because of the driving force for vapour transport through membranes decreases along with the permeate side flow path length due to the rise of temperature in permeate stream, such a proposed system for preventing a significant rise in permeate stream temperature has the potential for application in enhancing flux of large-scale DCMD modules.

Table 3 shows the calculated flux, the area required for the external heat exchanger and the energy consumption per unit volume of water permeated. With a temperature difference $\Delta T = 50^\circ$C, the required area of the external heat exchanger is about three times the membrane cell area, and the required heat exchanger area decreases with a decreasing temperature difference, since the lower flux reduces the temperature rise in the permeate stream flowing across the MD cell. One of the advantages of applying MD is that it can utilize low-grade or waste heat energy. If the energy available to heat the feed is completely contributed by waste heat and only fluid pumping work is considered, the calculated energy consumption per unit volume of permeated water in the study is in the range of 3.13–5.36 MJ/m³, which is smaller than the 9–16 MJ/m³ reported in large-scale RO processes (Meindersma et al. 2006).

**CONCLUSION**

The water flux in DCMD was compared using ePTFE and PVDF membranes. The results showed that the ePTFE membranes permeate more distillate water than the PVDF
membranes because the former have a higher porosity and a thinner active-layer. An iterative process based on transport models was developed to predict DCMD flux. The experimental water fluxes obtained were in good agreement with those predicted by the transport model. According to the simulated results of the MD process using a combination of simultaneously cooling the permeate stream and adding an external heat exchanger to reduce the permeate stream temperature, such a process could increase flux and lead to an energy consumption smaller than that of RO processes when waste energy is available to heat the feed.

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