Brackish water treatment for reuse using vacuum membrane distillation process
Muttucumaru Sivakumar, Mohammad Ramezanianpour and Glen O’Halloran

ABSTRACT
The availability of fresh water is vital for all human activities and in particular for improving living conditions, health and overall well-being. Pressure on scarce fresh water resources can be reduced by treating and reusing brackish water by advanced membrane treatment technologies. In this study, brackish water originating from effluent discharge of a local coal mine, seawater, groundwater and salt water swimming pool is treated by a laboratory-scale vacuum membrane distillation (VMD) system. VMD is an emerging technology that has the potential to become as important as conventional distillation system and aims to remove particles and dissolved impurities by evaporation and condensation techniques that mimic what occurs in nature. This study validates the mathematical modeling of the transport mechanisms used in the VMD process using data collected for different experimental situations. The response of flux rate to various process operating parameters, including pressure, temperature, flow rate and salinity concentration, is also demonstrated. This thermally driven separation process enables to remove 99.9% of total dissolved solids (TDS) from brackish water. The quality of the permeate water from all four water sources studied is of acceptable standards for potable use; however, it requires mineralization efforts before direct consumption.

Key words | brackish water, heat and mass transfer, hydrophobic membrane, vacuum membrane distillation, water reuse

INTRODUCTION
Rapid urbanization, population growth and associated industrial and agricultural activities together with climate change are known major factors in increasing fresh water demand across the world. A sustainable and integrated urban water management strategy must involve alternative water sources to protect fresh drinking water supplies. Brackish water originating from mine water effluent (Sivakumar et al. 2015), swimming pool salt water (Ramezanianpour & Sivakumar 2015), sea water and groundwater are such sources that when treated with advanced membrane technologies can be reused for a variety of purposes.

Membrane distillation (MD) is a thermally driven separation process which uses hydrophobic porous membranes to separate a gaseous phase from the entry liquid (Qtaishat et al. 2008). A negative pressure or coolant flow on the permeate side creates physical separation in all four configurations of MD, namely direct contact membrane distillation, sweeping gas membrane distillation, vacuum membrane distillation (VMD) and air gap membrane distillation (Khayet 2011). VMD is increasingly becoming recognized and accepted as a cost competitive and energy efficient alternative to membrane separation processes such as reverse osmosis (RO) and conventional MD (Mericq et al. 2010; Khayet & Matsuura 2011). VMD can compete with RO in terms of specific energy consumption; however, it will require higher membrane area than RO
(Cabassud & Wirth 2003). VMD process produces ultra-pure water as it rejects all non-volatile constituents such as ions, dissolved non-volatile organics, colloids and pathogenic microorganisms. The VMD technique has a number of advantages over conventional MD techniques. Perhaps the most notable is the fact that distilled water can be produced at lower operating temperatures, resulting in lower operating costs (Safavi & Mohammadi 2009). During VMD, a liquid–vapor interface is created at the membrane surface and prevents contact between the liquid feed and membrane. This interface minimizes heat transfer losses within the system and reduces the potential for membrane fouling. Therefore, lower energy consumption is required to achieve similar flux rates compared with pressure-driven desalination processes (Cabassud & Wirth 2003). The only concern is the hydrophobicity of the membrane, as it acts as a barrier between liquid solution and gas phase.

There is much literature available on the theory and physics of mass transfer behind VMD (Lawson & Lloyd 1996; Alklaibi & Lior 2005; Al-Obaidani et al. 2008; Khayet 2011), and small scale experimental systems have been designed, constructed and installed (Xu et al. 2006; Criscuoli et al. 2008; Sivakumar et al. 2013). Studies into the energy and cost-effectiveness of large scale VMD systems have shown it to be, theoretically, very competitive with other distillation methods (Chen et al. 2011; Ben Abdallah et al. 2013). In a recent study, it has been shown that a solar-based VMD process is one of the most sustainable distillation systems among the three desalination processes studied due to its weights obtained in comparative judgment of systems and total weight of indicators in an optimized analytical hierarchy process (Sivakumar & Ramezanianpour 2013). The multi-criteria decision analysis was performed for a solar-based VMD, solar-based electro-coagulation followed by ultrafiltration and solar-based membrane biological reactor system. Indicators in three major environmental, economic and social criteria are incorporated to avoid subjectivity in decision-making and raise awareness of the actual effect of each parameter. To assist in the design and implementation of effective desalination methods, more research is warranted to determine the effectiveness of VMD to treat specific water sources and the effects of their properties on flux and fouling of membranes that can reduce efficiency and increase operational costs.

THEORETICAL BACKGROUND

Membrane characteristics play an important role in the flux rate. Higher flux rate can be achieved by higher porosity, larger pore size and suitable coating of the membranes. It has been shown that membrane coating with chitosan resulted in higher rate of permeate flux (Chanachai et al. 2010). However, larger pore size reduces the critical entry pressure required for feed-water to infiltrate the membrane pores. Thereby, determination of an ideal pore size is extremely influenced by the membrane material and the characteristic of feed solution. The mass transfer mechanisms in a VMD system can be described by the kinetic theory of gases. The ratio of mean free path, λ, of the transported vapor molecules to the diameter of the membrane pores, d, namely Knudsen number (Kn = λ/d), provides a guideline to determine the accurate mechanism of mass transfer (Lawson & Lloyd 1997). A model of the Knudsen flow, the viscous flow and Knudsen-viscous type of diffusion can explain mass transfer in a VMD process for Kn > 1, Kn < 0.01 and 0.01 < Kn < 1, respectively.

For a membrane with small pores (Kn > 1), molecule-pore wall collisions are dominant. Thereby, the Knudsen diffusion equation, which can be adapted for most membrane distillation configurations, is implemented to predict the performance of VMD process. The Knudsen diffusion model expressed in Equation (1) assumes Knudsen flow regime for vapor passing through small holes in a thin wall (Lawson & Lloyd 1997; Khayet et al. 2004). The number of molecules passing through a pore is directly proportional to the driving pressure of the gas and inversely to its molecular weight

\[
N_w = \frac{K \Delta P}{R \delta T_{fm}} \left( \frac{8RT}{\pi M} \right)^\frac{1}{2}
\]

where \(N_w\) is the molar flux (mol/m² s), \(\Delta P\) (Pa) is the pressure difference between the partial pressure of the solution and the absolute vacuum pressure, \(R\) is the gas constant (8.31 J/mol K), \(\delta\) is the membrane thickness (m), \(T_{fm}\) indicates temperature at feed side of the membrane surface (K), \(M\) is the molecular weight of water (18.01528 g/mol) and \(K\) is the Knudsen diffusion constant (m). Water
temperature at the feed side of the membrane can be calculated as

\[
T_{mf} = T_{m1} - \frac{\dot{m}v H_{lv}}{H_{l}}
\]

(2)

where \(T_{m1}\) is feed-solution temperature (K), \(\dot{m}v\) is vapor mass flow rate (kg/s), \(H_{lv}\) is specific enthalpy of vaporization (J/kg), \(h_{f}\) is heat transfer coefficient (W/m² K) and \(A_{m}\) is the total membrane surface area (m²).

For membrane with large pores (\(K_n < 0.01\)) viscous flow arises so the molecule–molecule collision is dominant. In this case, Equation (3) was proposed to determine the total flux (Lawson & Lloyd 1997; Khayet & Matsuura 2004)

\[
N_w = \frac{\pi r^4 P_{ave}}{8 \mu RT_{mf} A_m \tau \delta} \Delta P
\]

(3)

where \(r\) is the average of the pores’ radius (m), \(P_{ave}\) is average partial pressure (Pa), \(A_{m}\) is the total membrane surface area (m²), \(\tau\) is the membrane tortuosity and \(\mu\) is dynamic viscosity of feed solution (Pa s). In transition region (0.01 < \(K_n < 1\)) both molecule–molecule and molecule–pore wall collisions have to be considered. A Knudsen-viscous model was developed in the transition region to describe VMD performance as Equation (4) (Khayet 2011)

\[
N_w = \frac{\Delta P}{RT_{mf} \delta} \left[ K \left( \frac{8RT}{\pi M} \right)^{1/2} + B \frac{P_{ave}}{\mu} \right]
\]

(4)

where \(B\) is a constant and depends on membrane pore radius, tortuosity and porosity. The above models predict flux response to changes in temperature, pressure, flow rate and feed solution density and salinity for a specific membrane. For the chosen membrane and experimental conditions used in this research, the Knudsen number \(K_n\) was found to be >1.

**MATERIALS AND METHODS**

The experimental arrangement of a typical laboratory-scale VMD process using a hydrophobic membrane is presented schematically in Figure 1. The VMD experiments were undertaken by monitoring the effects of various operating parameters on flux for treating brackish water. Feed-water was warmed up in a reservoir by a temperature controlled...
Water bath circulation system. Warmed up feed-water was circulated through the membrane module by a digitally controlled Masterflex peristaltic pump. The effects of temperature and flow rate were monitored using a temperature probe and flow meter incorporated at the membrane inlet. The Emfion polyethylene terephthalate (PFR) membrane filter cartridge (Pall, Australia) used had a surface area and pore diameter of 0.8 m² and 0.2 μm, respectively, and contained a double layer polytetrafluoroethylene (PTFE) hydrophobic membrane in pleated surface form. Vacuum pressure on permeate side of the membrane was applied when the temperature reached a desired value. To ensure accurate control of vacuum pressure, a ball valve and a pressure sensor were fitted into the Javac vacuum pump. A condensation column placed across the vacuum pump and the permeate side of the membrane condensed vapor into a liquid phase. Initially a liquid nitrogen trap was used to condense water vapor. This had significant problems including difficulty of getting constant supply of liquid nitrogen, evaporation at an extremely fast rate and accumulation of ice crystals on the outside surface from the surrounding air during the transporting of the trap to the scales for weighing. Hence, a conventional glassware condensation column is substituted, as shown in Figure 1.

To investigate the desalination capabilities of VMD process for commercial and industrial applications, feed solutions such as Milli-Q water, saline solution made by NaCl and various brackish water sources were collected for analysis. Groundwater samples were collected from Wagon Wheels farm, located in Stanwell Tops, NSW. Seawater was collected from Wollongong harbor. Saline mine water was collected from the BHP Billiton owned Westcliff mine site located in Appin NSW. Swimming pool salt water samples from the University of Wollongong’s swimming pool were also collected. Water quality parameters such as total dissolved solids (TDS), pH, total coliforms and the concentration of Na, Ca, Mg and Fe were analyzed for each sample before and after the treatment process.

The pH and TDS of raw and treated water samples were measured by Eutech Instrument PCD650. The procedures recommended in Standard Method 9221B (APHA 2012) were followed in determining the total coliform count calculated in colony forming units per 100 mL of sample (cfu/100 mL). The measurements of Ca, Mg, Fe and Al were undertaken using an Atomic Absorption Spectrophotometer Model AA-6500 (Shimadzu) based on Standard Method 3111B (APHA 2012).

RESULTS AND DISCUSSION

The variation of permeate flux with vacuum pressure, temperature, flow rate and salinity of feed-water for a given set of experimental conditions was studied in this research. Sodium chloride was mixed with distilled water to obtain different concentrations of saline solutions. The energy required to operate the VMD process incorporated the power of vacuum pump (110–115 W), circulation pump (15.5–87.6 W) and water bath circulator (5–500 W) and the operating time which was measured using a power meter for each test. The calculation of energy consumption to produce 1 kg of permeate water is based on the variation of the three significant parameters: pressure, temperature and flow rate. The experiments were also carried out for long periods in order to observe the rate of flux decline. This is attributed to the scaling on the membrane surface. The irreversible permeate flux decline was determined by following VMD test using Milli-Q water.

The effects of vacuum pressure were investigated by decreasing permeate side absolute pressure from 20 to 5 kPa and running the system at various vacuum intervals within this range while all other parameters remained constant. The variation of the flux extracted from Milli-Q water distillation with changing vacuum pressure is shown in Figure 2. The Knudsen diffusion model predicted the measured flux accurately. As illustrated in Figure 2, flux tended to reach zero for pressure above 20 kPa as the pressure above this value is not suitable for vaporization of feed solution at 65 °C. Vacuum pressure was found to be the variable having the most significant effect on flux among the variables studied (Sivakumar et al. 2013). It was observed that an increase in pressure on the permeate side of the membrane resulted in a severe decrease in permeate flux rate due to a significant reduction in the driving force for trans-membrane flux. The specific energy consumption is defined as the energy required to obtain a unit mass of permeate water (Safavi & Mohammadi 2009) and is also shown in Figure 2. The specific energy consumption is
found to be the lowest at high vacuum pressure that causes high flux rate.

For the experiments, feed temperatures from 55 to 75 °C were selected in steps of 5 °C, as temperatures above the maximum value are not only considered detrimental to the membranes but will also require more energy for heating. Feed-water temperature had a considerable effect on vapor permeation flux. Figure 3 illustrates the relationship between feed-water temperature and the flux permeation rate extracted from Milli-Q water distillation for a given set of experimental conditions. The permeate flux significantly increases as feed-water temperature increases. This can be attributed to the fact that an increase in temperature increases the water vapor pressure exponentially. The Knudsen model was also seen to be acceptable in the estimation of the flux for given experimental conditions. However, at high temperatures the experimental data appear to be higher than the predicted value, presumably due to the changes in membrane characteristics. Figure 3 also illustrates the variation of energy consumption per unit mass of permeate water with temperature. As expected, energy consumption per unit mass of permeate increases with temperature. Safavi & Mohammadi (2009) reported an order of magnitude higher energy consumption rate of 3.5 kWh/kg for their VMD process when operated at a permeate pressure of 4 kPa with feed-water set at 55 °C. However, their membrane had an area of 4 cm² which is significantly smaller than the membrane employed in this study. It is important to note that the membrane area and type deployed in a VMD process can critically affect specific energy consumption (Cabassud & Wirth 2003).

The effect of feed-water flow rate on flux was investigated by increasing the flow from 0.5 to 2 L/min while all other variable parameters were kept constant. Figure 4 illustrates the resulting fluxes extracted from Milli-Q water distillation for variation of flow rate. It also shows that doubling the flow raises flux only marginally by about 6%. The slow increase in flux is due to increase in flow Reynolds number which influences the Nusselt number, heat transfer coefficient, vapor temperature and finally the saturation pressure of feed solution. The energy consumption per unit mass of permeate water remained relatively constant with variation of flow rate. The energy consumption
increased from 134.1 to 156.3 Wh, which is insignificant compared with the results achieved for pressure and temperature. It is illustrated that higher flow rate assists in extracting more distilled water with small changes in energy consumption. However, the feed side pressure has to be controlled to prevent pore wetting problem.

The effects of initial salinity concentration were also investigated by increasing the initial feed-water concentration from 0 to 21 g/L while all other parameters remained constant. Figure 5 shows that increasing feed-water salinity concentration has negligible effect on flux and this is one of the significant advantages of using VMD for desalination purposes. A reduction of 5.8% was observed for a short-term experiment using 21 g/L salinity solution. The flux decline at high concentration of 100–300 g/L was reported as less than 35% by Safavi & Mohammadi (2009), whereas in other studies the total flux decline of 13–28% was derived at feed concentrations of 50–120 g/L NaCl (Martínez-Díez & Florido-Díaz 2001; Wirth & Cabassud 2002). A comprehensive study of the scaling phenomenon during MD process followed by developing a mathematical model for the decline rate of the permeate flux has been performed (Ramezanianpour & Sivakumar 2014). However, in this research the removal efficiency of the system in terms of salt rejection was investigated. The modeled flux was marginally above the experimental data due to the effect of scaling phenomena. This is attributed to the fact that the model flux did not take into consideration the effect of concentration polarization with salinity. Salt removal efficiency of over 99.9% was achieved in the VMD process irrespective of feed salt concentrations.

The treatment of groundwater, seawater, mine water effluent and salt pool water was investigated by running the system and measuring the flux at various intervals. The VMD process was carried out at 65 °C temperature, 5 kPa pressure and 1 L/min flow rate while the feed solution was recirculated through the membrane. It was found that the initial flux was below the modeled clean water flux and a decrease of 6.8, 6.1, 5.3 and 5.8% occurred over the duration of the experiment for treatment of groundwater, seawater, mine effluent water and salt water, respectively. The flux decline is due to the deposition of suspended and dissolved solids on membrane surface. The TDS removal efficiency remained constant at 99.99 ± 0.05% for all cases. The membrane cleaning with a citric acid (C6H8O7) solution was carried out after the experimental run with brackish water sources. The flux rate value was restored to between 97 and 99% of the original value.

A number of water quality tests were performed on raw feed-water and the corresponding treated product water in order to investigate removal efficiency and hence the ability of VMD to produce potable water quality. These values have then been compared with the guideline values in Table 1. It was generally found that the metals and TDS removal efficiencies were in the high 99% removal. Standard total coliform tests using the membrane filter procedure were carried out on both feed-water and permeate samples to determine microbial removal efficiency. However, for some samples, low values for total coliforms within feed-water samples were found. In all cases the pH values dropped between the feed-water and permeate. This is most likely caused by the removal of alkalinity and adsorption of atmospheric carbon dioxide in the permeate water. All permeate water quality values were significantly below the Australian Drinking Water Guideline (ADWG) and WHO guideline values.

**CONCLUSION**

A laboratory study has been undertaken to show the importance of VMD for desalinating different types of
brackish water. The influence of various operating parameters on flux rate and specific energy consumption was measured. Among the operating variables studied, it was found that vacuum pressure was the most influential parameter, followed by feed-water temperature, feed-water flow rate and finally salinity. The experimental flux values were also compared with the theoretically determined values for the same operating conditions. In general the theoretical predictions using the Knudsen type of diffusion model agree well with experimental observations. Moreover, higher vacuum pressure was found to be advantageous for increasing flux in an energy efficient manner. The specific energy consumption was unaffected by the increase in feed flow rate. However, increase in temperature raised the specific energy consumption mainly for heating the feed-water. If VMD process is to be sustainable, then it is recommended that renewable energy sources like solar energy are used for this purpose.

The results showed that VMD process produces desalinated and distilled water from a variety of saline, brackish and contaminated water sources. A flux decline of \( 0.6\% \) was found for each of the water sources tested as a result of scale formation or particulates being deposited on the membrane surface. Contaminant removal efficiency remained considerably high for measured contaminants and water sources. The limited water quality parameters measured in the permeate fell within the potable water guideline values, which indicates that the permeate water is suitable for drinking. However, the values indicate that mineralization may be necessary for direct potable consumption.

**REFERENCES**


Ben Abdallah, S., Frikha, N. & Gabsi, S. 2013 Design of an autonomous solar desalination plant using vacuum membrane distillation, the MEDINA project. Chemical Engineering Research and Design 91 (12), 2782–2788.


**Table 1 | Water quality analysis**

<table>
<thead>
<tr>
<th>Parameter (mg/L)</th>
<th>ADWG</th>
<th>WHO</th>
<th>Feed-water</th>
<th>Permeate</th>
<th>Feed-water</th>
<th>Permeate</th>
<th>Feed-water</th>
<th>Permeate</th>
<th>Feed-water</th>
<th>Permeate</th>
<th>Feed-water</th>
<th>Permeate</th>
</tr>
</thead>
<tbody>
<tr>
<td>TDS</td>
<td>500</td>
<td>1,000</td>
<td>417</td>
<td>2.41</td>
<td>3,685</td>
<td>2.12</td>
<td>2,332</td>
<td>2.66</td>
<td>4,495</td>
<td>2.25</td>
<td></td>
<td></td>
</tr>
<tr>
<td>pH</td>
<td>6.5–8.5</td>
<td>6.5–9.2</td>
<td>7.31</td>
<td>6.11</td>
<td>7.98</td>
<td>6.15</td>
<td>7.68</td>
<td>6.32</td>
<td>7.28</td>
<td>6.38</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Calcium</td>
<td>60–200</td>
<td>100–200</td>
<td>3.00</td>
<td>0.08</td>
<td>410</td>
<td>0.73</td>
<td>14.4</td>
<td>0.74</td>
<td>71.39</td>
<td>0.20</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Magnesium</td>
<td>N/A</td>
<td>N/A</td>
<td>8.69</td>
<td>0.09</td>
<td>338</td>
<td>0.48</td>
<td>2.72</td>
<td>0.03</td>
<td>2.88</td>
<td>0.01</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Iron</td>
<td>0.3</td>
<td>0.2</td>
<td>12.6</td>
<td>0.07</td>
<td>0.32</td>
<td>0.00</td>
<td>1.92</td>
<td>0.04</td>
<td>N/A</td>
<td>N/A</td>
<td></td>
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</tr>
<tr>
<td>Aluminum</td>
<td>0.2</td>
<td>0.2</td>
<td>0.67</td>
<td>0.00</td>
<td>0.18</td>
<td>0.00</td>
<td>3.38</td>
<td>0.00</td>
<td>N/A</td>
<td>N/A</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total coliforms</td>
<td>1</td>
<td>0</td>
<td>10</td>
<td>0.00</td>
<td>1,230</td>
<td>10</td>
<td>0.00</td>
<td>0.00</td>
<td>N/A</td>
<td>N/A</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

All values in mg/L, except total coliforms, which are measured in cfu/100 mL, and pH.


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