Direct contact membrane distillation: Capability to desalt raw water

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Abstract In this work, the potentialities of membrane distillation to desalt raw waters were investigated. The experiments were performed on a direct contact membrane distillation (DCMD) unit using a flat sheet polypropylene (PP) membrane with a low pore size of 0.064 μm. The effect of relevant operating parameters such as transmembrane temperature difference, hydrodynamic conditions and ionic strength on permeate flux and conductivity was studied. The results indicated that a permeate flux increases with increasing transmembrane temperature difference and Reynolds number, and slightly decreases with increasing ionic strength. The permeate flux reached 4.24 L/m²h at a temperature difference of 60 °C and Reynolds number of 3740 and ionic strength of 8.56 × 10⁻⁴ M. DCMD process using PP with low pore size membrane present a very low salt passage through the membrane which was not affected by feed concentration. DCMD process has been applied during a long period to desalt raw water without any pretreatment. For brackish water, the variation of permeate flux and conductivity were slightly changed as function of operating time. For seawater, the permeate flux decreased slightly and the permeate conductivity increased sharply in which a simple pretreatment step is recommended to ameliorate the performance of DCMD process.

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1. Introduction

Reverse osmosis (RO) and electrodialysis (ED) are the most commercially available membrane technologies used for desalination of brackish water and seawater because of many reasons such as lower specific power consumption and, consequently, lower specific water production cost (Hassan and Fath, 2013). The drawbacks of these membrane processes are related to low water production rate, environmental pollution due to discharge of concentrated water and fouling phenomena.

Membrane distillation (MD) is an emerging and attractive method which can be considered as an alternative solution for conventional desalination processes. MD is a thermally driven process, in which only vapour molecules, not liquid water, are transported through porous hydrophobic membranes (El-Bourawi et al., 2006), where the driving force is the vapour pressure difference created by the temperature
difference across the membrane. MD has many advantages, such as low operating temperatures in comparison to thermal processes, and lower hydrostatic pressure compared to pressure-driven membrane processes (Alkhudhiri et al., 2012). In addition, MD is expected to be a much lower membrane fouling as compared to other membrane processes due to its relatively large pore size of hydrophobic membrane, and the feed water does not require extensive pretreatment step. The pore size of the membranes frequently used in MD process ranged between 0.001 and 1 μm (Khayet and Matsuura, 2011). More suitable membranes in MD process became available, such as hydrophobic membranes made by polytetrafluoroethylene (PTFE), polypropylene (PP), polyethylene (PE), and polyvinylidene fluoride (PVDF).

Based on the method used to activate the vapour pressure gradient across the membrane, MD process may be classified in four configurations (Drioli et al., 2006): (i) Direct contact membrane distillation (DCMD), where the permeate side consists of condensing a cold liquid in direct contact with the membrane; (ii) air gap membrane distillation (AGMD), where the distillate can be recovered on a condensing surface separated from the membrane by an air gap; (iii) sweeping gas membrane distillation (SGMD), where an inert gas is used to sweep the vapour at the permeate side of the membrane; (iv) Vacuum membrane distillation (VMD), where vacuum is applied in the permeate side by using vacuum pumps.

DCMD is one of the famous varieties of the MD process, in which the hydrophobic membrane is in direct contact only with liquid phases, feed water on one side and distilled water on the other (Alklaibi and Lior, 2004). DCMD is considered as the simplest design and appears to be the best for application to the desalinization of various feed saline water such as seawater (Al-Otaibi et al., 2008; Curcio et al., 2010; He et al., 2011; Hsu et al., 2002; Shirazi et al., 2012), brackish water (Hou et al., 2010), brine (Godino et al., 1996; Ji et al., 2010; Martinez-Diez and Florido-Diaz, 2001; Qu et al., 2009) and synthetic salt water (Martinez-Diez and Vazquez-Gonzalez, 1999). The simple configuration of DCMD has proven to be highly efficient in terms of the permeate flux.

In this work, DCMD experiments using polypropylene hydrophobic membrane with low pore size were carried out for the desalinization of different sources of raw water. The effects of relevant operating parameters on the permeate flux and permeate conductivity were studied including transmembrane temperature difference, hydrodynamic conditions and feed ionic strength. Thereafter, the application of DCMD process was investigated to desalt two types of raw waters without any pretreatment steps, such as brackish water and seawater.

2. Materials and methods

2.1. Membrane and DCMD setup

Experiments were carried out using a flat sheet microporous membrane made of polypropylene (PP) polymer with trade name Celgard® 2500 provided by Hoechst-Celanese. The principal membrane characteristics are presented in Table 1.

The flat sheet DCMD laboratory setup is shown in Fig. 1. The setup consisted of two thermostatic cycles (feed and permeate) that were connected to a membrane module which was made by Plexiglas. The feed compartment, connected to a heating resistant, is maintained at a hot temperature and the permeate compartment, connected to a cooling system, is maintained at a cold temperature. The hydrophobic membrane was placed between the two compartments. The effective membrane area is 0.0042 m². The feed and permeate temperatures were measured inside each compartment by a digital

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Characteristics of PP membrane.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Material</td>
<td>Polypropylene</td>
</tr>
<tr>
<td>Nominal pore size</td>
<td>0.064 μm</td>
</tr>
<tr>
<td>Thickness</td>
<td>25 μm</td>
</tr>
<tr>
<td>Porosity</td>
<td>55%</td>
</tr>
<tr>
<td>Manufacturer</td>
<td>Hoechst-Celanese, USA</td>
</tr>
<tr>
<td>Contact angle</td>
<td>120°</td>
</tr>
<tr>
<td>Liquid entry pressure</td>
<td>200 kPa</td>
</tr>
<tr>
<td>Effective membrane area</td>
<td>4.2 × 10⁻³ m²</td>
</tr>
</tbody>
</table>

Figure 1  Schematic diagram of DCMD setup: (1) Permeate tank; (2) Cooling system (3) peristaltic pumps; (4) thermocouples; (5) conductometer; (6) flat sheet module; (7) feed tank; (8) heating resistant.
thermometer with an accuracy of ± 0.1 °C. The feed and the permeate flowed with the help of two peristaltic pumps with a variable flow rate. A conductivity meter was used to monitor changes in the conductivity of permeate reservoir.

The permeate flux (L/m²h) was calculated by the following equation:

\[ J_p = \frac{\Delta V}{\Delta t} \]

where \( \Delta V \) is the volume of permeate (L), \( A \) is the effective membrane area (m²) and \( \Delta t \) is the sampling time (h).

2.2. Analytical methods and feed solutions

Different feed water and permeate solutions were collected at definite interval of all experiments, and were analysed. Electric conductivity and pH solutions were measured using a conductivity/pH meter (Consort CS61). Chloride ion was measured by argentometric titration with Mohr indication (Nollet, 2007). Calcium and magnesium were determined by the EDTA titration method. TDS and sulphate ion were determined by the flame photometric method. Fluoride concentration was determined using ion selective electrode. Nitrate ion was analysed using UV–visible spectrophotometer. The bicarbonate concentration was calculated according to the concentration of inorganic carbon in the solution, which was determined using a TOC analyzer.

The synthetic solutions were prepared by dissolving a reagent grade NaCl salt (supplied by Fluka) in distilled water to obtain different ionic strengths.

The brackish water is collected from the aquifer nape in the south of Tunisia, located in the town of Degache.

The seawater was obtained from the Mediterranean coast of Borj Cedria, northern of Tunisia.

The characteristics of both raw waters are summarized in Table 2.

3. Results and discussion

3.1. DCMD treatment of synthetic NaCl solution

3.1.1. Permeate flux and conductivity as a function of running time

The DCMD experiments were carried out to determine the steady state, or equilibrium, and permeate flux during running time. The behaviors of the permeate flux and permeate conductivity as a function of operating time using PP membrane are presented in Fig. 2. It can be seen that the permeate flux is almost constant. The average values of measured permeate flux ranged between 2.82 and 3.02 L/m²h. Thus, the application of DCMD seems to be more advantageous in desalination of salt solution compared with the pressure driven membrane techniques, where a significant decline of the permeate flux was observed after a short operating period (Kumar et al., 2006).

In terms of water quality, the permeate conductivity was slightly increased from 0.9 to 2 μS/cm, and the permeate quality was stable over time. The permeate flux and insensitive increases of the permeate conductivity indicate that DCMD process presents a high stable performance over operating time, which indicate that NaCl solution did not cause any problem such as membrane wettability or fouling phenomenon, which is in agreement with previous studies (He et al., 2011). For this reason, a selected running time of 2 h seemed to be sufficient for the rest of experiments.

3.1.2. Transmembrane temperature difference

MD is a non isothermal membrane separation process, in which the transmembrane temperature difference between feed and cold sides (\( \Delta T \)) was considered as dominant operating parameters that affect the permeate flux. The effect of temperature difference on DCMD permeate flux was studied in the range 20–60 °C at a constant permeate temperature of 20 °C.
Fig. 3 shows the relationship between temperature difference and DCMD permeate. It can be seen that the permeate flux through PP membrane increased significantly with increasing temperature difference. An increase of $\Delta T$ from 20 to 60 $^\circ$C, resulted in an exponential increase in the permeate flux from 0.75 to 4.24 L/m²h; it is about 4.65-fold. This result could be predicted by the Antoine equation (Eq. (2)) (Khayet, 2011), which expresses the relationship between the vapour pressure, as a driving force for DCMD process, and feed temperature.

$$\ln(P_{\text{sat}}) = A + \frac{B}{T_f + C}$$  

(2)

where $A$, $B$ and $C$ are the regression constants for the specific compounds, $P_{\text{sat}}$ in Pascal and $T_f$ in Kelvin. For water $A = 23.238$; $B = 3841$; $C = 45$.

From Fig. 3, it was observed also that the vapour pressure difference and the permeate flux as a function of transmembrane temperature difference at an ionic strength of $8.56 \times 10^{-2}$ M are almost in the same order of magnitude, which confirms the relationship between $P_{\text{sat}}$ and $\Delta T$.

Moreover, when the temperature is increased, the temperature polarization and the viscosity of the feed saline water declined, which were favourable to enhance the permeate flux (Phattaranawik et al., 2003). This result is in accordance with previous studies using DCMD process (Peng et al., 2005; Yun et al., 2006; He et al., 2011).

3.1.3. Feed hydrodynamic conditions

Reynolds number can be used to characterize the fluid flow state, which is represented by $Re = \frac{vD}{\mu}$, where $\rho$ is density (kg/m³); $v$ velocity (m/s); $D$ hydraulic diameter (m); and $\mu$ is viscosity (kg/ms) of the fluid. Flow liquid is in laminar flow regime when $Re < 2000$ and in turbulent flow regime when $Re > 4000$.

The selected Reynolds number coverts a large spectrum from the laminar to turbulent regime flow; e.g. the Re is between 992 and 16,865. The effect of hydrodynamic conditions on the permeate flux is shown in Fig. 4. It can be observed that the permeate flux increased with increasing Reynolds number. At Reynolds number < 4000, the permeate flux value increases significantly with increasing Reynolds number. At Reynolds number > 4000, the permeate flux increased slightly and can reach an asymptotic value of permeate flux of 3.36 L/m²h.

The increasing Reynolds number improves the mass transfer coefficient in the interface membrane-solution. In addition, the increasing Reynolds number reduces the resistances of transfers; thus reducing the temperature and the concentration polarization effect (Yun et al., 2006; He et al., 2011; Pal and Manna, 2010). This means that the temperature at the membrane surface is approximately more close to that of the bulk streams, and thus the transmembrane temperature difference is larger. This increases the driving force and consequently enhances the flux.

3.1.4. Ionic strength

The effect of ionic strength on the performance of DCMD process was studied, the selected domain for the ionic strength is between 1.71 and $42.78 \times 10^{-2}$ M. The permeate flux and the water vapour pressure as a function of increasing ionic strength were shown in Fig. 5. It can be seen that the permeate flux decreased slightly from about 21% when the ionic strength was increased from $1.71 \times 10^{-2}$ to $42.78 \times 10^{-2}$ M. The ionic strength has no significant effect on the permeate flux as that observed in the case of transmembrane temperature difference or Reynolds number. This result can be explained by the Raoult’s law (Eq. (3)) (Qtaishat et al., 2008). The decrease in the saturation vapour pressure induces the increasing driving force and the performance of DCMD declined.

$$P_{\text{sat}} = (1 - x_i)P^\rho$$  

(3)

where $P_{\text{sat}}$ is the vapour pressure of feed water (Pa); $P^\rho$ is the vapour pressure of pure water ($P^\rho$); $x_i$ is the mole fraction of the solute in the feed water.

By applying Raoult’s law, the vapour pressure of feed water is calculated at a feed temperature of 70 $^\circ$C and its evolution as a function of ionic strength is presented in Fig. 5. It can be observed that the behaviour of both, vapour pressure and permeate flux, as a function of ionic strength are in the same order of magnitude. Which confirm that the DCMD permeate flux through the microporous membrane is proportional to the vapour pressure difference across the membrane.

Also, while the ionic strength increased, the dynamic fluid will change as a result of increasing viscosity and concentration polarization should be added to the temperature polarization, which reduces the imposed DCMD driving force and then the permeate flux (Schofield et al., 1990).

![Figure 4](image1.png) **Figure 4** Permeate flux vs. Reynolds number (ionic strength = $8.56 \times 10^{-2}$ M, $\Delta T = 50$ $^\circ$C, $Re_p = 4196$).

![Figure 5](image2.png) **Figure 5** Permeate flux and vapour pressure vs. ionic strength ($\Delta T = 50$ $^\circ$C, Ref = 3657, Rep = 4196).
The permeate conductivity as a function of increasing feed ionic strength is shown in Fig. 6. An increasing ionic strength in the feed solution from 1.71 to $42.78 \cdot 10^{-2}$ M can lead to a slightly increasing permeate conductivity from 0.6 to 7.5 lS/cm. DCMD process can produce a high quality fresh water recovered after desalting solution with high salinity in the range of brackish and seawater (Hsu et al., 2002).

3.2. DCMD comparison between synthetic and real water at equivalent ionic strength

In this section, DCMD treatment approach between synthetic water and natural raw water at equivalent ionic strength as NaCl was examined.

3.2.1. Synthetic and brackish water

A comparison of the effect of transmembrane temperature difference on the permeate flux between synthetic water and brackish water at equivalent ionic strength as NaCl salt under the same conditions is present in Fig. 7. It can be seen that the permeate fluxes for two types of water increased exponentially with temperature difference as it has been reported previously, but the natural brackish water shows a slightly higher permeate flux than synthetic water at the same ionic strength. This can be attributed to that the equivalent ionic strength synthetic water is higher than the brackish water, in which the divalent ions are abundant in the natural water and cannot be involved in the calculation of the equivalent ionic strength.

3.2.2. Synthetic and seawater

To compare the behaviour of the permeate flux obtained by DCMD desalination of synthetic water and seawater under the same conditions; it must be suitable to work at equivalent ionic strength. Fig. 8 shows the DCMD permeate flux as a function of temperature difference, for synthetic and seawater. The experimental results showed that temperature difference had a significant influence on the permeate flux as it has been reported previously. For both types of water, curves of permeate fluxes as a function of temperature difference are almost superimposed. This trend can be explained by the fact that the ionic strength for synthetic water is almost equal to the equivalent ionic strength for seawater, in which the NaCl salt is the abundant element.

3.2.3. DCMD applications of natural water resources

Two natural saline waters were desalted by DCMD process using polypropylene membrane, including brackish water and seawater. The chemical compositions of two resources of water are presented in Table 1. The DCMD operating conditions were transmembrane temperature difference of 50 °C, permeate Reynolds number of 4196 and feed Reynolds number of 9694, corresponding to turbulent flow. The experimental results of the permeate flux and conductivity for raw seawater and brackish water during 62 h of running time are presented in Fig. 9. It can be observed for both natural saline waters that the permeate flux decreases slightly as a function of time. For brackish raw water, the permeate flux ranged between 3.16 and 2.79 L/m²h, the fitted flux curve shows a decline of 11.7% during 62 h. For raw seawater desalination, without any pretreatment, the permeate flux ranged between 2.46 and 2.28 L/m²h, and from fitted curve, the 62 running hours of experiment shows only 7.3% decrease of the permeate flux. The obtained results confirm that the concentration has a slightly effect on the permeate flux, which was reported previously in the effect of ionic strength.

The permeate conductivity increased for both raw waters as a function of time as shown in Fig. 9. In the case of brackish water with initial feed salinity of 3.58 g/L, the permeate conductivity had been known to increase from 1.1 to 107 lS/cm after 62 h of DCMD operation. In fact, the electrical conductivity
conductivity of permeate is quite below the recommended values for drinking water. The obtained results of desalination of raw brackish water, without pretreatment, confirm that the performance of the DCMD process is maintained quasi constant for a long running period. Indeed, the desalination of raw brackish water has almost apparently no effect on hydrophobic character of the polypropylene membrane during 62 h.

For the DCMD desalination of raw seawater, Fig 9 shows that the permeate electrical conductivity as a function of time had a sensitive decline during the first 53 h from 14.2 to $175 \mu$S/cm, in which a high quality of water was produced. Thereafter, a sharp increase of permeate conductivity was observed to reach $949 \mu$S/cm after 62 h of DCMD operation. This can be attributed to the presence of high amount of total dissolved solids and organic matter in raw seawater, which may affect the membrane wettability and diminished the hydrophobicity of PP membrane. As a result, the transport of partial amount of solutes through the wetted pores from the feed to the distillate compartment can led to an increase in the permeate conductivity. In this case, it is recommended to integrate a simple pretreatment step, such as a 5 micron cartridge filter, to ameliorate the performance of DCMD process.

4. Conclusions

Direct contact membrane distillation using polypropylene membrane with a low pore size is able to desalt aqueous salt solution and natural raw water: brackish and seawater. The effect of relevant operating parameters including feed temperature, feed Reynolds number and initial ionic strength was studied. The following conclusions may be drawn from the obtained results:

- The hydrophobic PP membrane can produce high quality of water with low electrical conductivity for all experimental runs.
- The permeate flux increased exponentially with increasing transmembrane temperature difference through membrane side.
- The DCMD permeate flux was also affected by hydrodynamic conditions. Hence, increase in the feed Reynolds number is accompanied by a sensitive increase in the permeate flux.
- The feed ionic strength had a slight effect on the permeate flux as compared with the effect of transmembrane temperature difference.
- The comparison between synthetic water and real water at equivalent ionic strength shows a great agreement as a function of temperature difference.
- The results of long period DCMD application of raw brackish water and seawater without any pretreatment showed that the PP membrane had slightly decreased permeate flux for both sources during 62 h. The permeate conductivity had slightly increased, reached $171 \mu$S/cm after 62 h for desalting raw brackish water, and a sharp increase was obtained to reach $949 \mu$S/cm for desalting raw seawater due to partial membrane pores wettability.
- The desalination of raw saline water by DCMD process using PP membrane can produce a high water quality with low electrical conductivity below the recommended values for drinking water.

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References


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Yun, Y., Ma, R., Zhang, W., Fane, A.G., Li, J., 2006. Direct contact membrane distillation mechanism for high concentration NaCl solutions. Desalination 188, 251–262.