A novel multi-stage direct contact membrane distillation module:

Design, experimental and theoretical approaches

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Abstract

An economic desalination system with a small scale and footprint for remote areas, which have a limited and inadequate water supply, insufficient water treatment and low infrastructure, is strongly demanded in the desalination markets. Here, a direct contact membrane distillation (DCMD) process has the simplest configuration and potentially the highest permeate flux among all of the possible MD processes. This process can also be easily instituted in a multi-stage manner for enhanced compactness, productivity, versatility and cost-effectiveness. In this study, an innovative, multi-stage, DCMD module under countercurrent-flow configuration is first designed and then investigate both theoretically and experimentally to identify its feasibility and operability for desalination application. Model predictions and measured data for mean permeate flux are compared and shown to be in good agreement. The effect of the number of module stages on the mean permeate flux, performance ratio and daily water production of the MDCMD system has been theoretically identified at inlet feed and permeate flow rates of 1.5 l/min and inlet feed and permeate temperatures of 70 °C

and 25 °C, respectively. The daily water production of a three-stage DCMD module with a membrane area of 0.01 m² at each stage is found to be 21.5 kg.

Keywords: multi-stage DCMD; module design; desalination; composite membrane; experiment; modeling

1. Introduction

Global climate change and variability have a negative impact on water supply and quality in remote areas by diminishing both the availability of water and the dilution of contaminants, such as pollutants, sediment and salts. These effects are expected to be exacerbated by a limited and inadequate water supply, low infrastructure and insufficient water treatment. Because it is unfeasible to adopt large-scale and mature seawater desalination technologies such as multi-stage flash (MSF), multi-effect desalination (MED) and reverse osmosis (RO) in such regions, seawater desalination markets will necessarily require an economic seawater desalination system with a small scale and footprint in order to supply sufficient fresh water to isolated areas. The membrane distillation (MD) process may be an alternative for providing potable water in such areas. MD is a thermally-driven separation process using a microporous hydrophobic membrane. The transmembrane partial vapor pressure difference is the driving force of the MD process, and is generated by the temperature gradient formed between the liquid-vapor interfaces across the membrane. This MD process has many attractive features, such as low operating temperature and hydraulic pressure, low sensitivity to salt concentration, a small footprint, a near 99.9% rejection of non-volatile solutes (Alkhudhiri et al., 2012; Curcio and Drioli, 2005), independent performance of high osmotic pressure or concentration polarization (Al-Obaidani et al., 2008; Cheng et al., 2011; El-Bourawi et al., 2006; Francis et al., 2013, 2014a; Kim et al., 2013; Lawson and Lloyd, 1997; Lee and Kim, 2013, 2014; Lee et al., 2015a, 2015b; Maab et al., 2013; Shim et al., 2014), potentially low maintenance requirements (Guillén-Burrieza et al., 2011) and high permeate flux in lab-scale membrane tests (Cath et al., 2004; Francis et al., 2014b; Gilron et al., 2007; Li and Sirkar, 2004). Due to the aforementioned advantages, MD has exhibited high potential as a seawater desalination process; however, the major barriers hindering its commercialization, aside from the MD membrane, include module design, membrane pore wetting, low permeate flux, flux degradation with time, as well as uncertain energy and economic costs. In order to overcome these limitations, various studies on module and process design utilizing a heat recovery concept have been conducted (Blanco Gálvez et al., 2009; Duong et al., 2015; González-Bravo et al., 2015; Guan et al.,

2015; Khayet et al., 2005; Kim et al., 2013, 2015; Lee et al., 2011; Maab et al., 2012; Zhao et al., 2013) and various MD membranes have been developed (Feng et al., 2006; Francis et al., 2014c; Huo et al., 2009; Khayet et al., 2005; Peng et al., 2005; Prince et al., 2013; Winter et al., 2013). A composite membrane, which has high permeability with an appropriate pore size and porosity, low thermal conductivity with a high thickness and porosity, good thermal stability and high mechanical strength, has been developed and may meet the intricate demands of MD (Khayet et al., 2005; Lee et al., 2015a).

A multi-stage MD process with various configurations such as series, parallel and series/parallel arrangements of the MD modules has been studied to efficiently increase water production and system performance (González-Bravo et al., 2015; Lee and Kim, 2014). It has been shown that the multi-stage concept for the air gap membrane distillation (AGMD) and vacuum membrane distillation (VMD) processes could reduce energy consumption by recovering the latent heat of water vapor condensation (Guillén-Burrieza et al., 2011; Zhao et al., 2013). By applying the multi-stage scheme, an improvement in MD permeate flux can be achieved with thermal entrance effects, which affects the enhancement of heat transfer coefficient near the inlet flow region of each stage as the thermal boundary layer begins to develop near the inlet region (Phattaranawik et al., 2003a; Welty et al., 2009). In addition, due to the separated module configuration, the multi-stage scheme may provide supplementary advantages, such as simple maintenance, replacements, as well as the ability to check the leaks in the module (Lee and Kim, 2014). Multi-stage MD systems have been studied by many researchers (Blanco Gálvez et al., 2009; Kim et al., 2015; Lee and Kim, 2014; Lee et al., 2015b; Zhao et al., 2013). Lee and Kim (2014) presented various configurations of multi-stage VMD systems. Among the proposed systems which have various configuration manners, an optimized multi-stage VMD system configuration was determined via cost evaluation. Kim et al. (2015) proposed a solar-assisted multi-stage VMD system with a heat recovery unit, which could increase thermal efficiency and water production. Blanco Gálvez et al. (2009) reported an innovative solar-powered AGMD desalination system. The solar powered multi-stage AGMD system was developed and experimentally determined to improve the energy efficiency and reduce water production costs. The memsys had commercialized a vacuum-multi effect membrane distillation (V-MEMD) module, which achieved highly efficient heat recovery as compared to conventional thermal desalination processes. The solar-driven memsys system showed good operating performance with a flux at approximately 7 l/m²h on a sunny day with a seawater feed (Zhao et al., 2013).

All of the aforementioned studies have attempted to develop a commercialized MD system using a multi-

stage concept due to negligible conductive heat loss through the membranes of both the AGMD and VMD processes. However, the AGMD has several drawbacks such as complex module design and low permeate flux, and the VMD has several disadvantages such as treatment of non-condensable gases, requirement of an additional vacuum pump and membrane pore wetting, which readily occurs when vacuuming the permeate side of the membrane. In contrast, the direct contact membrane distillation (DCMD) process has the simplest configuration and potentially the highest permeate flux among all of the possible MD processes. In addition, the effects of the diffusion of non-condensable gases on the permeate flux is negligible in the DCMD process due to the very small quantity, as compared with a high DCMD permeate flux (Khayet and Matsuura, 2011). Furthermore, DCMD can be configured easily in a multiple stages to achieve improvements in compactness, cost-effectiveness, productivity and versatility. However, previous experimental and theoretical studies for DCMD process with a multi-stage concept were conducted with a rather simple module configuration via external pipelines between the stages (Gilron et al., 2007; Lee et al., 2011; Song et al., 2008). It was evident that a multi-stage concept could have a higher water production due to a higher effective membrane area. The module configuration proposed in previous works, however, can lead to additional heat losses to the ambient through the external pipelines, larger footprint, more complex structure as well as higher capital cost and operating cost due to higher pumping power caused by high pressure drop. In order to ameliorate such shortcomings, completely different module configurations need to be designed and studied both theoretically and experimentally.

The ultimate objective of this work is to develop a high-performance multi-stage direct contact membrane distillation (MDCMD) process that is applicable to a small scale and footprint desalination system. In this study, therefore, an innovative MDCMD module under countercurrent-flow operation has been first designed and both theoretically and experimentally examined to demonstrate the feasibility and operability of the module design for desalination. A rigorous numerical model, which was developed in our previous work (Lee et al., 2015a), has been modified to incorporate a thermal entrance effect near the inlet flow region of each stage for the performance prediction of the MDCMD process. Further investigations have been conducted to identify the effect of the number of module stages on the mean permeate flux, performance ratio and daily water production of the MDCMD system.

2. Experimental set-up and procedure

The requirements of an ideal membrane applied for the MD process are (i) higher permeability with an optimal pore size and porosity, (ii) lower thermal conductivity with higher thickness and porosity, (iii) higher liquid entry pressure with water and smaller maximum pore size, (iv) higher mechanical strength, (v) excellent thermal stability and (vi) chemical resistance (Khayet et al., 2005; Lee et al., 2015a). However, it exhibits a conflict between the requirements of an ideal membrane such as higher mass transfer and lower conductive heat loss. It is well known that the above conflict can be resolved by using a microporous hydrophobic/hydrophilic (or hydrophobic) composite membrane (Khayet et al., 2005; Lee et al., 2015a). As shown in the SEM images (clockwise from top left: 100x, 500x, 1,000x and 10,000x magnifications) in Fig. 1 (Lee et al., 2015a), therefore, a commercial hydrophobic microporous PTFE/PP composite membrane (Sterlitech Corporation) has been used for this study. It appears that the knot-fibril net-structured PTFE active layer (dark gray in top right of Fig. 1) is partially covered by the PP scrim support layer (white gray). Here, the PTFE active layer is not covered by the PP support layer at the permeate side, indicating an effective area for diffusion that can be expressed by the surface porosity, defined as the surface area of the PTFE active layer exposed to the permeate side divided by the total membrane surface area. The surface porosity is found to be 42% using CAD software based on the SEM images. The physical properties of the PTFE/PP composite membrane are given in Table 1. Note that the composite membrane has a significantly thinner and more porous active layer than the scrim support layer, which can result in decreased mass transfer resistance and increased heat transfer resistance.

Fig. 2 represents a schematic (Fig. 2a) and picture (Fig. 2b) of the novel MDCMD module made of polycarbonate material. The module consists of two exterior units and a pair of interior units. The number of interior units depends on the number of stages in more than single stage units. Therefore, the number of interior units equal to the number of stages minus 1, while in the case of a single stage, the interior unit is not required. As shown in this figure, rotation coupling implementation is employed to assemble the adjacent units. Here, the module requires no external pipelines between the stages due to the incorporation of internal flow channels with a first-in-last-out (FILO) configuration, which helps form a uniform flow in the channel width direction, resulting in improved MD performance. This multi-stage module concept can enhance compactness, productivity, versatility and cost-effectiveness in the DCMD process. The specifications of the MDCMD module include a 0.01 m^2 ($0.1 \text{ m} \times 0.1 \text{ m}$) effective membrane area in each stage and a 3 mm channel height in both the feed and permeate sides. As membrane support and turbulence promoters, polypropylene (PP) mesh spacers have been implemented in both the feed and permeate channels. Detailed specifications of the spacer

are shown in Table 2.

A schematic (Fig. 3a) and picture (Fig. 3b) of the experimental set-up are shown in Fig. 3. Deionized (DI) and tap water are used as bulk permeate and feed solutions, respectively. The tap water flows through the active layer side of the membrane, while DI water is circulated through the support layer side of the membrane in a countercurrent-flow manner. The feed and permeate temperatures are kept constant using a heater and chiller (RW-1025G, Lab Companion), respectively. The permeate production rate is measured using an electronic balance (CUX-6200H, CAS). The temperature, pressure and volume flow rate at both the inlet and outlet of the module are simultaneously monitored at both the feed and permeate sides. In order to detect the leaks or pore wetting of the membrane, the conductivity of the feed and permeate solutions at the outlet of the module is measured using a conductivity/resistivity sensor; the corresponding signal is transmitted to a PC via a transmitter (M300, Mettler-Toledo Thornton) and data acquisition device. The performance of the MDCMD system using a composite PTFE/PP membrane has been investigated by varying the inlet feed temperature in the range of 50 °C – 70 °C and the number of stages from 1 to 3 while keeping inlet permeate temperature constant at 25 °C. The inlet feed and permeate volume flow rates in the range of 1.0 l/min to 2.5 l/min are tested for single-stage modules and for two- and three-stage modules: their values are 1.0 l/min and 1.5 l/min, respectively, due to a lack of heating and cooling capacities. For reproducibility, all experiments are conducted more than three times and the maximum deviation in the measured permeate fluxes is found to be less than $\pm 6\%$.

3. Theoretical approach

To demonstrate the feasibility and operability of the MDCMD module designed in the present work, a theoretical investigation has been also performed. As shown in Fig. 4, the heat and mass transfers occur simultaneously across the boundary layers near the membrane surfaces and through the PTFE/PP composite membrane. Also, the presence of turbulence or eddy currents induced by the spacers in both channels will yield better flow characteristics. As a result, the thickness of the thermal boundary layer in the spacer-filled channel is less than that in the empty channel, which brings the membrane surface temperatures closer to the bulk temperatures. Thus, temperature polarization can be reduced, and permeate fluxes can be enhanced due to a larger vapor pressure difference through the PTFE/PP composite membrane. Detailed theoretical models and solution procedure for the heat and mass transfer through the composite membrane and transport phenomena on

the bulk feed and permeate flows have been demonstrated in our previous work (Lee et al., 2015a).

In the present work, the theoretical model developed previously has been modified to incorporate the thermal entrance effect at each stage as a consequence of the adoption of the multi-stage concept. In order to estimate the heat transfer coefficient, h , in both the spacer-filled feed and permeate channels, therefore, the modified Dittus-Boelter's correlation on the flat surface of a fully-developed turbulent flow has been employed, as given in Eq. (1). For the spacer-filled channel, it has been known that the heat transfer coefficient is best described by the turbulent flow correlation at low Reynolds numbers ($Re < 2100$) (Al-Sharif et al., 2013; Da Costa et al., 1994; Fane et al., 1987; Lee et al., 2015a; Lydersen, 1979; Phattaranawik et al., 2003a, 2003b, 2001; Schofield et al., 1987; Welty et al., 2009; Zhang et al., 2012).

$$Nu = \frac{hd_h}{k} = k_{sp} 0.023(1 + 6d_h / L) Re^{0.8} Pr^{0.33}, \quad (1)$$

with $Re = \rho v d_h / \mu$, $Pr = \mu c_p / k$ and $d_h = \frac{4\epsilon_{sp}}{2(w_c + h_c) / w_c h_c + 4(1 - \epsilon_{sp}) / d_f}$ for a spacer-filled channel,

where k_{sp} is the spacer factor, d_h is the channel hydraulic diameter (i.e., 9.8×10^{-4} m), k is the thermal conductivity of the bulk feed or permeate, L is the effective membrane length, ϵ_{sp} is the spacer porosity, w_c is the channel width, h_c is the channel height and d_f is the diameter of the spacer filament.

As can be expected from the spacer factor included in Eq. (1), the spacer can be employed as a membrane support and turbulence promoter, resulting in an enhancement of heat and mass transfers and flow resistance (Da Costa et al., 1994; Lee et al., 2015a; Phattaranawik et al., 2003a). As illustrated in Fig. 5, the main geometric parameters of the spacer factor are (i) hydrodynamic angle (in degree, θ), (ii) mesh size (l_m), (iii) filament diameter (d_f), (iv) spacer thickness (h_s) and (v) spacer porosity (ϵ_{sp}), which can be estimated from the aforementioned four geometric parameters as follows (Da Costa et al., 1994; Lee et al., 2015a):

$$\epsilon_{sp} = 1 - \frac{\pi d_f^2}{2l_m h_s \sin \theta}. \quad (2)$$

In this study, therefore, the spacer factor correlation can be defined as follows:

$$k_{sp} = c_1 \left(\frac{d_f}{h_s} \right)^{c_2} \left[\sin \left(\frac{\theta}{2} \right) \right]^{c_3} \exp \left[c_4 \ln(\epsilon_{sp}) \right]^{c_5}. \quad (3)$$

Five parameters (c_1, c_2, c_3, c_4, c_5) in the spacer factor above are fitted to the experimental permeate flux data using the conjugate gradient method (CGM) (Kim and Kim, 2009). The coefficient of determination, R^2 , which indicates the percentage of variability in the dependent variables (the variance about the mean), is implemented

to analyze the fitting degree of spacer factor with the measured data and is expressed as (Steel and Torrie, 1960):

$$R^2 = 1 - \frac{\sum_{i=1}^N (J_{m,exp,i} - J_{m,sim,i})^2}{\sum_{i=1}^N (J_{m,exp,i} - \bar{J}_{m,exp})^2}, \quad (4)$$

$$\text{with } \bar{J}_{m,exp} = \frac{1}{N} \sum_{i=1}^N J_{m,exp,i},$$

where $J_{m,exp}$ and $J_{m,sim}$ are the mean value of the measured and predicted permeate fluxes, respectively, and N is the number of measured permeate flux data.

For the performance investigation of the MDMCD process using a multi-stage concept, the mean performance ratio (PR) is expressed as (Lee et al., 2015a)

$$PR = \frac{1}{L} \int_0^L \eta_z dz, \quad (5)$$

where η_z is the local performance ratio, determined as the ratio of the vaporization heat associated with the permeate flux to the heat transferred through the membrane, and is given by (Lee et al., 2015a)

$$\eta_z = \frac{\varepsilon J_z \Delta H|_{al} + (1-\varepsilon) J_z \Delta H|_{al-sl}}{Q_m}, \quad (6)$$

where ε is the surface porosity of the composite membrane mentioned in the first paragraph of Section 2, J_z is the local permeate flux, $\Delta H|_{al}$ and $\Delta H|_{al-sl}$ are the enthalpy of evaporation at the mean temperature through the active layer membrane and the active/support layer membrane, respectively, and Q_m is the heat flux through the membrane.

In this study, as stated before, tap water is employed as bulk feed solution (experiment and simulation) and thus its salinity is assumed to be zero for the simulation. This is because it has been shown that the MD flux is not significantly affected by the feed water salinity (Francis et al., 2013, 2014a, 2014b; Lee et al., 2015a; Maab et al., 2013), at least in the range of seawater desaliation application, including treating brines (Xu et al., 2016).

4. Results and discussion

4.1. Performance evaluation and model validation

In order to identify the feasibility and operability of the multi-stage module design with a countercurrent-flow configuration developed in the present work, experimental and theoretical investigations on its performance have been carried out by varying the inlet feed temperature in the range of 50 °C – 70 °C at the

inlet permeate temperature of 25 °C, and the feed and permeate volume flow rates in the range of 1.0 l/min – 2.5 l/min. In this section, a brief comparison and description of the measured and predicted results of the MDCMD modules are described and a more detailed interpretation of the physical characteristics of their results is discussed in the following section.

With an increase in the inlet feed temperature from 50 °C to 70 °C at the stream flow rate of 1.0 l/min for a single-stage module, the measured mean permeate flux increases from 11.9 kg/m²h to 31.2 kg/m²h, whereas the model prediction shows an increase in mean permeate flux from 10.9 kg/m²h to 29.1 kg/m²h (Fig. 6). The mean performance ratio increases asymptotically from 62.2% to 80.7% (black lines in Fig. 6). It is shown that at the stream flow rate of 1.5 l/min, the measured mean permeate flux increases from 14.1 kg/m²h to 37.7 kg/m²h and the model-based permeate flux increases from 13.8 kg/m²h to 37.7 kg/m²h, while the mean performance ratio increases gradually from 62.8% to 81.3% (red lines in Fig. 6). A good agreement between the measured and predicted data is observed, with maximum relative deviations of 8.4% for 1.0 l/min and 3.5% for 1.5 l/min. It is noted that a higher permeate flux is achieved at a higher feed temperature, as the partial vapor pressure increases exponentially with inlet feed temperature. Furthermore, a higher flow rate provides enhanced heat transfer through both the feed and permeate boundary layers and then reduces the temperature polarization, which indicates a higher transmembrane temperature difference and results in a higher permeate flux in the DCMD process (Al-Obaidani et al., 2008; Cath et al., 2004; El-Bourawi et al., 2006; Guillén-Burrieza et al., 2011; Kim et al., 2013; Li and Sirkar, 2004). The increase of feed flow rate leads to shorter residence time maintaining higher temperature difference across the membrane, hence higher flux (Alsaadi et al., 2013).

When increasing the inlet feed temperature at the stream flow rate of 2.0 l/min for the single-stage module, the measured mean permeate flux increases from 16.3 kg/m²h to 45.5 kg/m²h and the simulated mean permeate flux increases from 15.9 kg/m²h to 46.0 kg/m²h. The mean performance ratio increases from 63.0 % to 82.2 % (black lines in Fig. 7). With a stream flow rate of 2.5 l/min, meanwhile, the measured mean permeate flux increases from 16.5 kg/m²h to 48.7 kg/m²h, while the predicted mean permeate flux also increases from 18.3 kg/m²h to 52.3 kg/m²h. The performance ratio increases from the 63.7 % to 82.5 % (red lines in Fig. 7). Here, the maximum relative errors between the experimental and simulated results on the mean permeate flux are found to be 2.5% and 11.0% for 2.0 l/min and 2.5 l/min, respectively. As shown in Figs. 6 and 7, all of the relatively large deviations are observed at lower feed temperatures of 50 °C, which may be attributed to the under- or over-estimation of the MD coefficient, but also can be due to the lower permeate flux obtained under

these conditions, hence a lower accuracy in the measurements.

For the two-stage DCMD module at a stream flow rate of 1.0 l/min (black lines in Fig. 8), the measured mean permeate flux increases from 11.5 kg/m²h to 29.4 kg/m²h, whereas the predicted mean permeate flux increases from 10.4 kg/m²h to 27.6 kg/m²h, with an increase in the inlet feed temperature from 50 °C to 70 °C. The mean performance ratio increases from 62.2 % to 80.5 %. At the inlet feed and permeate flow rates of 1.5 l/min (red lines in Fig. 8), the measured mean permeate flux increases from 13.2 kg/m²h to 36.7 kg/m²h and the model-based permeate flux increases from 13.5 kg/m²h to 36.2 kg/m²h. The performance ratio increases gradually from 63.1 % to 81.6 %. The model predictions of the permeate flux are also in good accordance with the measured results, with maximum deviations of 10.2% and 5.4% for 1.0 l/min and 1.5 l/min, respectively.

For the three-stage module at a stream flow rate of 1.0 l/min (black lines in Fig. 9), the measured mean permeate flux increases from 10.1 kg/m²h to 24.0 kg/m²h and the model prediction shows an increase in mean permeate flux from 9.82 kg/m²h to 25.2 kg/m²h, with an increasing inlet feed temperature. The predicted mean performance ratio increases from 61.8 % to 79.8 %. With a stream flow rate of 1.5 l/min (red lines in Fig. 9), the measured mean permeate flux increases from 11.4 kg/m²h to 29.9 kg/m²h, while the predicted mean permeate flux increases from 12.8 kg/m²h to 34.0 kg/m²h. The mean performance ratio also increases from 62.6 % to 81.2 %. Here, the maximum relative deviations between the measured and predicted results on the mean permeate flux are found to be 5.0% for 1.0 l/min and 13.7% for 1.5 l/min.

Based on the aforementioned experimental results, the convective heat transfer coefficient at the feed and permeate boundary layers is obtained from the procedures described in Section 3. The heat transfer coefficient obtained is correlated with spacer characteristics via the multiple linear regressions. The best fit between the measured and simulated permeate fluxes is achieved with a squared correlation coefficient of 0.944 (Table 3).

At inlet feed and permeate flow rates of 1.5 l/min, the model-based mean permeate flux and daily water production achieved by varying the inlet feed temperature from 50 °C to 70 °C are compared with regard to the number of module stages: single- (black lines), two- (red lines) and three-stage (blue lines) (Fig. 10). It is noted that as the number of module stages increases from single- to two- and three-stage, on average, the mean permeate flux (solid lines) decreases by 4% and 9%, whereas the daily water production (dashed lines) increases by 92% and 172%, or 1.92 times and 2.72 times, with an increase in the effective membrane area.

The relative good agreement between the prediction results and experimental data, therefore, demonstrates that the multi-stage module concept may achieve enhanced compactness, productivity, versatility and cost-

effectiveness in the DCMD process.

4.2. Influence of the multi-stage concept

As the feasibility and operability of the multi-stage module designed in the present work has been previously demonstrated, in this section, the effect of the number of module stages on the mean permeate flux, performance ratio and daily water production of the MDCMD system has been theoretically identified at inlet feed and permeate flow rates of 1.5 l/min and inlet feed and permeate temperatures of 70 °C and 25 °C, respectively.

The local permeate flux, performance ratio (Fig. 11a) and temperature (Fig. 11b) profiles along the normalized axial distance with respect to various numbers of module stages (N_s) from one to twenty are compared. Here, the dimensionless axial distance (horizontal axis) is defined as $[L(i-1) + z] / (N_s L)$, where L is the membrane length at each stage, i is the number of module stages ($0 \leq i \leq N_s$), and z is the local axial coordinate ($0 \leq z \leq L$). It is clearly shown that the local permeate flux decreases linearly along the membrane length, and the permeate flux throughout the membrane decreases greatly as the number of stages is increased. This effect is particularly evident with lower quantities of stages, and then decreases asymptotically with larger quantities of stages (Fig. 11a). With an increase in the number of stages (membrane length) in a countercurrent-flow configuration, the heat transfer rate through the membrane via both the latent heat of vaporization induced by permeate flux and heat conduction increases with stream longer retention times. Hence, as shown in the axial temperature variations (Fig. 11b), or bulk feed (T_f), liquid-vapor interface at the feed side ($T_{f,m}$), liquid-vapor interface at the permeate side ($T_{p,m}$) and bulk permeate (T_p), the temperatures at the feed and permeate sides decrease and increase along the flow directions, respectively, which results in a lower transmembrane temperature difference ($T_{f,m} - T_{p,m}$) as a driving force, yielding a lower permeate flux by means of a proportionality of flux to the driving force. At a given number of stages, the temperature changes at each stage of the bulk feed and permeate streams are almost identical, but its value near the front of the stages is marginally larger than that near the rear of the stages due to a relatively higher permeate flux. The average temperature changes at each stage are 1.87 °C, 1.59 °C, 1.37 °C, 1.19 °C and 1.06 °C, respectively, with an increase in the number of stages from one to twenty, which is attributed to an asymptotic decrease of the transmembrane flux. As the number of module stages increases, a relatively smaller flux decline is observed at the front of the module stages (Fig. 11a), which is because a higher permeate flux can be achieved at higher

temperatures (Fig. 11b), as expected from the exponential relationship between temperature and vapor pressure in the Antoine equation (Felder and Rousseau, 2000):

$$\log_{10}(133.3P_{w,m}) = 8.07131 - \frac{1730.63}{T + 233.426} \quad (7)$$

where $P_{w,m}$ is the water vapor pressure (in Pa) without dissolved species in the water and T is the temperature (in °C).

It is also shown that, with an increasing number of stages, the local performance ratio increases slightly near the front of the module stages (less than the dimensionless axial length of 0.3), while its value near the rear of the stages decreases significantly, mainly due to a lower permeate flux (Fig. 11a). The lower the transmembrane temperature difference, the lower the conductive heat loss across the membrane via Fourier's law of conduction, hence, a higher performance ratio can be obtained at the front of the stages.

In addition, the mean permeate flux (J_m), daily water production (J_{wp}) and mean performance ratio (PR) are compared with regard to the number of stages by keeping the other operating parameters constant as before (Fig. 12). It is shown that with an increase in the number of module stages from one to thirty, the mean permeate flux decreases from 38.5 kg/m²h to 15.3 kg/m²h and the mean performance ratio decreases from 81.6 % to 74.1 %, while the daily water production increases asymptotically from 9.24 kg/day to 110.0 kg/day. It is noted that the multi-stage concept can achieve a higher daily water production with a higher effective membrane area. However, this increase could actually mean an asymptotic increase in daily water production with regard to the number of module stages; as noted the maximum daily water production may be prohibitive under a given operating condition.

5. Conclusions

In this paper, a novel multi-stage direct contact membrane distillation (MDCMD) system is designed and its performance is examined both theoretically and experimentally. The MDCMD module with a countercurrent-flow configuration, applicable to a small scale and footprint desalination system while retaining process performance, is first designed, and then experimental and theoretical studies are successfully performed to demonstrate the feasibility and operability of module design in terms of permeate flux, daily water production and performance ratio. Here, the theoretical model is modified to incorporate a thermal entrance effect at the inlet flow region of each stage, which has a positive influence on the DCMD performance. Model predictions and measured data for mean permeate flux are compared and shown to be in good agreement. It is shown that as

the number of module stages increases from single- to two- and three-stage, the mean permeate flux decreases by 4% and 9%, respectively, while the daily water production increases by 1.92 times and 2.72 times, respectively, with an increase in the effective membrane area, which makes the proposed MDCMD module an attractive and energy-efficient concept for desalination application.

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Nomenclature

d_f	Filament diameter [m]
h_c	Channel height [m]
h_s	Spacer thickness [m]
$\Delta H _{al}$	Enthalpy of evaporation at the mean temperature through the active layer membrane [J/kg]
$\Delta H _{al-sl}$	Enthalpy of evaporation at the mean temperature through the active/support layers membrane [J/kg]
$J_{m,exp}$	Measured mean permeate flux [kg/m ² h]
$J_{m,sim}$	Predicted mean permeate flux [kg/m ² h]
J_z	Local permeate flux [kg/m ² h]
k_{sp}	Spacer factor [-]
l_m	Mesh size [m]
N	Number of measured data [-]
N_s	Number of module stages [-]
Q_m	Heat flux through the membrane [W/m ²]
w_c	Channel width [m]
z	axial coordinate [m]

Greek letters

ε	Surface porosity [%]
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1	ε_m	Membrane porosity [%]
2	ε_{sp}	Spacer porosity [%]
3	η_z	Local performance ratio [%]
4	θ	Hydrodynamic angle [°]

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Table captions

Table 1 Characteristics of the PTFE/PP composite membrane.

Table 2 Specifications of the spacer used in this study.

Table 3 The values of parameters in the spacer factor determined via the CGM.

Figure captions

Fig. 1. SEM images of a commercial PTFE/PP composite membrane. Clockwise from top left: 100x, 500x, 1,000x and 10,000x magnifications (Lee et al., 2015a).

Fig. 2 (a) Schematic and (b) picture of the MDCMD module with four and three stages, respectively.

Fig. 3 (a) Schematic and (b) picture of the experimental set-up.

Fig. 4. Schematic diagram of temperature polarization in both empty and spacer-filled channels.

Fig. 5. Geometric characteristics of a non-woven net spacer and flow pattern in a spacer-filled channel.

Fig. 6. Effect of inlet feed temperature and flow rate (1.0 l/min and 1.5 l/min) on the mean permeate flux (measured and predicted) and performance ratio (predicted) of a single-stage module.

Fig. 7. Effect of inlet feed temperature and flow rate ((2.0 l/min and 2.5 l/min)) on the mean permeate flux (measured and predicted) and performance ratio (predicted) of a single-stage module.

Fig. 8. Effect of inlet feed temperature and flow rate (1.0 l/min and 1.5 l/min) on the mean permeate flux (measured and predicted) and performance ratio (predicted) of a two-stage module.

Fig. 9. Effect of inlet feed temperature and flow rate (1.0 l/min and 1.5 l/min) on the mean permeate flux (measured and predicted) and performance ratio (predicted) of a three-stage module.

Fig. 10. Effect of inlet feed temperature on the predicted mean permeate flux and daily water production of single-, two- and three-stage modules.

Fig. 11. Comparison of predicted (a) local permeate flux (J_z), performance ratio (η_z), and (b) temperature (T_f , $T_{f,m}$, $T_{p,m}$ and T_p) along the normalized axial distance of single-, five-, ten-, fifteen- and twenty-stage modules.

Fig. 12. Comparison of predicted mean permeate flux, daily water production and mean performance ratio of multi-stage modules ranging from single to thirty.

Table 1 Characteristics of the PTFE/PP composite membrane.

Material	PTFE	PP
Thickness ^a , δ_m (μm)	20 ± 2	80 ± 2
Porosity ^a , ε_m (%)	70 ± 5	34 ± 5
Mean pore size ^a , r (μm)	0.5 ± 0.02	0.1 ± 0.02
Liquid entry pressure ^b , LEP_w (kPa)	207	160

^a data measured in this work, ^b data provided by manufacturer

Table 2 Specifications of the spacer used in this study.

Material	PP
Spacer thickness, h_s (mm)	1.35
Filament diameter, d_f (mm)	0.675
Angle between filaments, θ (deg)	90
Mesh size, l_m (mm)	1.26

Table 3 The values of parameters in the spacer factor determined via the CGM.

c_1	c_2	c_3	c_4	c_5	R^2
1.223	0.375	0.957	0.513	0.262	0.944

Top view **Side view**

Highlights

- ▶ An innovative MDCMD module with a countercurrent-flow configuration is proposed
- ▶ Feasibility and operability of module design are demonstrated experimentally and theoretically
- ▶ Model predictions and measured data for mean permeate flux are shown to be in good agreement
- ▶ Effect of the number of module stages on the performance of the MDCMD system is identified