Performance modelling of direct contact membrane distillation using a hydrophobic/hydrophilic dual-layer membrane

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**ABSTRACT**

HFP-co-PVDF/N6 hydrophobic/hydrophilic dual-layer membrane was used to study desalination with direct contact membrane distillation (DCMD). A one-dimensional (1-D) model was proposed to predict the flux and thermal efficiency. Heat and mass transfer equations were solved numerically for the combined hydrophilic and hydrophobic layers. The membrane characteristics of the hydrophobic layer were considered for the calculation of the mass transfer coefficients, while the hydrophilic layer was ignored since it was assumed to be filled with water. However, the hydrophilic layer was taken into account during the calculations of conductive heat transfer. Therefore, the equations are different, compared to single-layer hydrophobic membranes. It was found that with the same hydrophobic membrane characteristics, the single-layer membranes performed with better flux and thermal efficiency than the dual-layer membranes. Furthermore, the improvement of flux and thermal efficiency by an addition of the hydrophilic layer has not been observed experimentally, and it is suggested that the improved performance for dual-layer membranes reported previously is due to improved permeability by using thinner and more porous hydrophobic layers that can be mechanically reinforced by the hydrophilic layer. The validation of the model was conducted by comparing the experimental results for single- and dual-layer membranes with the modelling results. The predicted flux and thermal efficiency by the modelling were within 10% error to the experimental results.

**Key words:**

**HIGHLIGHTS**

- Mathematical models for predicting the flux and energy efficiency of dual-layer membranes.
- Experimental results have been used to validate the models.
- It is found that the thickness of the hydrophilic layer has a large effect on the flux and thermal efficiency.

**1. INTRODUCTION**

Membrane distillation (MD) is a separation process that has been known for over 50 years (Camacho \textit{et al.} 2013). It has been considered for different applications such as desalination, wastewater treatment, and dairy applications (Mostafa \textit{et al.} 2017). Various configurations of MD have also been considered such as direct contact MD (DCMD), air gap MD, vacuum MD, and sweeping gas MD (Li \textit{et al.} 2011). MD is a thermally driven process, in which heat and mass transfer occur simultaneously across the membrane (Camacho \textit{et al.} 2013). In DCMD, evaporation and condensation take place on the feed and permeate side, respectively. Vapour molecules transport through the membrane pores and condense on the permeate side for DCMD. This requires the membrane pores to be kept non-wetted during the process. In comparison with other desalination technologies, high-concentrated solution and pure water product can be achieved with MD (Enrico Drioli & Criscuoli 2009; Susanto 2011; Alkhudhiri \textit{et al.} 2012). MD has some disadvantages beside its advantages. Those are high thermal energy consumption, heat loss by conduction, and membrane wetting and fouling (Pangarkar \textit{et al.} 2011; Alkhudhiri \textit{et al.} 2012; Qtaishat & Banat 2013). The performance of MD depends on the membrane properties, process conditions, and the module design (Susanto 2011; Winter \textit{et al.} 2013). Proper module design should provide high-rate mass transfer, high turbulence for feed and permeate, and efficient evaporation. The suitable membrane in the process required to be resisted to wetting, high temperature, and fouling and scaling (Wang & Chung 2015). Mass transfer resistance from the membrane can be minimised by using membranes with low thickness and tortuosity (Adnan \textit{et al.} 2012). Thermal resistance can be increased using thicker membrane, so...
that heat loss can be prevented. The membranes with higher porosity can increase both MD permeability and thermal resistance for which MD flux and thermal efficiency both increase (Khayet et al. 2004). Modelling and computation can be accounted for as vital tools when optimising those aforementioned parameter effects on the performance of MD (Susanto 2011).

Different types of hydrophobic/hydrophilic composite membranes were prepared over the last decade (Essalhi & Khayet 2014). The concept of dual-layer composite membranes was claimed to improve MD flux for desalination in DCMD because of their low resistance to mass flux by decreasing water vapour transport path length through the thin hydrophobic top layer and their low conductive heat loss attributed by the thick hydrophilic layer (Khayet et al. 2005; Qtaishat et al. 2009a; Essalhi & Khayet 2014). Hou et al. (2012) fabricated polyvinylidene fluoride (PVDF) flat-sheet composite membrane using hydrophilic polyester non-woven fabric for MD. Through DCMD tests, the composite membrane achieved as high as 47.6 kg/m² h permeate flux with feed inlet and permeate inlet temperatures at 80.5 and 20.0 °C, respectively (Hou et al. 2012). In Qtaishat et al. (2009b), two different types of surface modifying macromolecules (SMMs) were blended into the host hydrophilic polymer polyetherimide (PEI) to prepare hydrophobic/hydrophilic porous composite membrane. They found that most of the dual-layer composite membranes performed at 55% higher fluxes than that of commercial polytetrafluoroethylene (PTFE) membrane. Qtaishat et al. (2009c) used hydrophobic/hydrophilic dual-layer membranes, which were blended hydrophilic polysulfone with hydrophobic SMMs. Some of the dual-layer membranes have exhibited higher DCMD fluxes than commercial PTFE membrane. Although M1 (SMMs/PS) dual-layer membrane that was used in Qtaishat et al. (2009c) had the highest flux among the other membranes in the study, M12 (SMMs/PEI) membrane from the study of Khayet et al. (2005) achieved higher flux than that of M1 membrane due to its thinner hydrophobic top layer. The hydrophobic layers prevent the penetration of water into the membrane pores and provide mass transfer resistance to vapour flow, while both hydrophobic and hydrophilic layers contribute to heat transfer (Khayet 2011). On the whole, it has been claimed that the hydrophobic layer of the membrane should be as thin as possible, whereas its pore size and porosity should be as large as possible to achieve high MD permeability (Khayet 2011).

Mathematical modelling of MD can lead to further awareness of process mechanisms (Hitsov et al. 2017). There have been a number of models that simulated various types of MD phenomena, and those models have their strengths along with some limitations (Hitsov et al. 2015). Models can be divided into three types: 0-D models, 1-D models, and 2-D models (Alsaadi et al. 2013). 0-D models do not consider the changes in fluid conditions along the module. Bulk averaged fluid conditions and module properties are used as inputs in this type of model. 1-D models divide the module into small elements along its length in the flow direction, so that in each element, temperature and flow properties can change along the membrane. 2-D models involve complex computational fluid dynamic approaches to describe the heat and mass transfer across the feed and permeate channels and membrane modules, which can be useful when changes in flow and temperature occur in two dimensions. 2-D models are computationally more expensive and require detailed calculations and longer time to solve compared to 1-D models (Alsaadi et al. 2013).

Modelling heat and mass transfer in DCMD is an approach to understand the effects of different design parameters on the performance of DCMD (Deshpande et al. 2017). MD modelling is based on heat and mass transfer equations of the process and incorporates membrane properties such as porosity and pore size (Zhang et al. 2011). Previous work has mainly focused on theoretical models and experimental studies for various operating conditions (Khayet 2011).

Performance modelling of hydrophobic membranes has been undertaken in previous studies for different MD configurations (Zhang et al. 2012, 2013; Lawal et al. 2014). Flux predictions were performed for PTFE membrane in DCMD under various process parameters such as velocity, module length, and feed temperature (Zhang et al. 2009). Ibrahim & Alsalhy (2013) predicted flux by performance modelling for DCMD using hollow fibre membrane and found that the effect of feed or permeate flow rate on permeate flux was less sensitive than the effect of feed temperature. MD performance of compressed PTFE membrane was modelled by Zhang et al. (2012, 2013), and flux and thermal efficiency were predicted at different pressures. Model validations were undertaken by comparing the predicted flux with experimental results as well as the exit temperatures for the feed and permeate, and good agreements were found between experimental results and model predictions. Lee et al. (2015) studied theoretical modelling of DCMD process using commercial hydrophobic microporous PTFE/PP composite membrane. The model predicted the flux successfully in comparison with experimental data, and
the surface porosity was found to be a significant factor among parameters such as thicknesses of active and support layer on the process performance (Lee et al. 2015). Winter et al. (2013) introduced new integrated and backing modelling approach for DCMD using different membranes with and without backing. The model included the parameters based on geometrical specifications of membrane and scrim/non-woven backing structures. Choosing the optimised backing structures providing high porosity and small coverage of the effective membrane surface had improvements on membrane performance. Zhang et al. (2011) developed a model for flat-sheet hydrophobic PTFE membrane in DCMD. The effect of process parameters, such as temperature and module length on flux, was observed. The model predicted the permeate flux at different module lengths and found that permeate flux reduced as the module length increased. One of the advantages of mathematical models is that, once validated, they can be used to scale up MD from laboratory scale to industrial scale.

0-D modelling was applied for hydrophobic/hydrophilic dual-layer membranes. The first model was developed by Qtaishat et al. (2009a), and the model was used to observe the hydrophobic and hydrophilic layer membrane characteristics’ influence on permeate flux. Optimum membrane characteristics were identified for high-efficiency MD. Heat and mass transfer equations were also derived for the model. Other modelling work examined the optimisation of membrane characteristics for hollow fibre hydrophobic/hydrophilic membrane to enhance the permeate flux (Bonyadi & Chung 2007). The effect of thermal conductivity of hydrophilic layer of dual-layer hollow fibre membrane was examined by experimental work and the model (Su et al. 2010). The study showed that increasing the thermal conductivity of the hydrophilic layer increased permeate flux. Nevertheless, the trend for flux changed once the thermal conductivity of the hydrophilic layer reached at certain level because of the limited temperature difference across the hydrophilic layer.

In this study, a 1-D model was developed to predict the permeate flux and energy efficiency for hydrophobic and hydrophobic/hydrophilic membranes in DCMD under various operating conditions. The predictions from the model were compared with the experimental results and show very good agreement. From this investigation, the effect of the hydrophilic layer on the flux was predicted and it is found that for the dual-layer membrane used, the hydrophilic layer resulted in a lowering of flux. The reasons for this observation will be discussed.

2. THEORY

Figure 1 shows the schematic diagrams of single- and dual-layer membranes for the current DCMD study (Li et al. 2011). The temperature and concentration variations across the feed and permeate channels, as well as that across the membrane, are shown.

The difference between the single- and dual-layer membranes is the extra hydrophilic layer in the dual layer. Figure 1 shows that for the dual-layer membrane, the thickness of the hydrophobic layer is in general much less than that in a single-layer membrane. The reasons for using the dual layer include: (1) the distance for vapour transport is reduced, and (2) the hydrophilic layer can be made strong to support the hydrophobic layer. In developing the 1-D model for heat and mass transfer across the membrane, the length of the module in the flow direction is generally discretised into small elements. Mass and heat transfer in both the flow and cross-flow directions are then calculated in each element based on energy and mass conservations. The equations given in the following sections apply to all membrane elements.

2.1. Heat transfer

The heat transfer through the hydrophobic membrane passes through three regions: bulk feed to the membrane surface, membrane, and permeate surface to the bulk permeate. The heat transfer mechanisms for each of these regions can be described as below:

Bulk feed to membrane surface:

$$Q_i = h_i(T_{b,f} - T_{m,i})$$ (1)

Through the membrane:

$$Q_m = h_m(T_{m,i} - T_{m,p}) + J \Delta H_v$$ (2)
Membrane surface to bulk permeate:

\[ Q_p = h_p(T_{m,p} - T_{b,p}) \]  

(3)

where \( Q_f, Q_m, \) and \( Q_p \) are the heat transfers across the feed, the membrane, and the permeate, respectively, \( T_{b,f} \) and \( T_{b,p} \) are the bulk temperatures of the feed and permeate, \( J \) is the permeate flux, \( \Delta H_v \) is the latent heat of vaporisation, and \( T_{m,f} \) and \( T_{m,p} \) are the temperatures at the membrane surfaces on the feed and permeate sides.
respectively. $h_f$, $h_m$, $h_p$ are the heat transfer coefficients at the feed, membrane, and permeate sides, respectively, in Equations (1–3), and the heat transfer is defined as being per unit area.

Assuming no heat loss from the module, the total heat transfer at steady state can be written as follows:

$$Q = Q_f = Q_m = Q_p$$  \hspace{1cm} (4)$$

The overall heat flux for single-layer membranes can be found as follows:

$$Q = \left( \frac{1}{h_f} + \frac{1}{h_m + (J_w \Delta H_v)/(T_{m,f} - T_{m,p}) + \frac{1}{h_p}} \right)^{-1} (T_{b,f} - T_{b,p})$$  \hspace{1cm} (5)$$

Heat transfer through the hydrophobic/hydrophilic dual-layer membrane is different compared to hydrophobic single-layer membranes due to the additional transport section, which is through the hydrophilic membrane layer. Heat flux through the hydrophilic layer is considered to be conductive because of water filling the layer. Heat transport associated with the flow of water through the hydrophilic layer is considered negligible because the flux is low and is ignored.

Through hydrophilic membrane layer:

$$Q_s = h_s(T_{m,p} - T_{s,p})$$  \hspace{1cm} (6)$$

At steady state,

$$Q = Q_f = Q_s = Q_p$$  \hspace{1cm} (7)$$

$$Q = U(T_{b,f} - T_{b,p})$$  \hspace{1cm} (8)$$

$$U = \left( \frac{1}{h_f} + \frac{1}{h_t + (J_w \Delta H_v)/(T_{m,f} - T_{m,p}) + \frac{1}{h_p}} \right)^{-1}$$  \hspace{1cm} (9)$$

The convective heat transfer coefficients can be estimated by using correlations of dimensionless numbers such as Nusselt, Reynolds, and Prandtl numbers.

$$Nu = f(Re, Pr)$$  \hspace{1cm} (10)$$

The Nusselt number is defined as follows:

$$Nu_i = \frac{h_i d_i}{k_i} \hspace{1cm} i = f, p$$  \hspace{1cm} (11)$$

The Prandtl number is used as a correction factor for the Nusselt number (Lawal et al. 2014):

$$Pr = \frac{\mu C_p}{k}$$  \hspace{1cm} (12)$$

And the Reynolds number:

$$Re = \frac{\rho u d}{\mu}$$  \hspace{1cm} (13)$$

where $k$ is the thermal conductivity of water, $\rho$ is the density of the water, $C_p$ is the specific heat of water, $d$ is the hydraulic diameter of the channel, and $u$ is the average velocity of the liquid.

The Nusselt number is chosen particularly in relation to flow regime in the channel (Khayet 2011; Zhang et al. 2012).
Re < 2,100 laminar regime:

\[ \text{Nu} = 1.86 \left( \frac{\text{Re}Pr \left( \frac{d_h}{L} \right)}{L} \right)^{1/3} \] (14)

2,100 < Re < 10,000 transitional regime:

\[ \text{Nu} = 0.116(\text{Re}^{2/3} - 125)\text{Pr}^{4/3} \left[ 1 + \left( \frac{d_h}{L} \right)^{2/3} \right] \] (15)

Re > 10,000 turbulent regime:

\[ \text{Nu} = 0.023\text{Re}^{4/5}\text{Pr}^{1/3} \] (16)

The conductive heat transfer coefficients of the hydrophilic sub-layer and hydrophobic top-layer can be calculated from the following equations:

\[ h_s = \frac{k_s}{\delta_s} \] (17)
\[ h_t = \frac{k_t}{\delta_t} \] (18)

where \( k_s, k_w, k_t, \) and \( k_g \) are the thermal conductivities of the hydrophilic membrane polymer, water in the pores, hydrophobic membrane polymer, and the gas contained in the pores; \( \delta_s, \delta_t, \) and \( \varepsilon_s, \varepsilon_t \) are the thickness and porosity of the hydrophilic and hydrophobic layer of the composite membrane, respectively.

The thermal efficiency (EE) was calculated from the ratio of flux (\( f \)) and latent heat transfer (\( f_{\text{latent}} \)) to mass flux (\( m_f \)), specific heat capacity (\( C_p \)), and temperature difference between bulk temperature (\( T_b \)) inlet and bulk temperature outlet (\( T_{bo} \)) as described by the following equation (Zhang et al. 2010; Swaminathan et al. 2016).

\[ \text{Thermal efficiency} = EE = \frac{Jf_{\text{latent}}A}{m_f C_p (T_b - T_{bo})} \times 100 \] (19)

Thermal efficiency for MD can be also described as the proportion of vaporisation latent heat to the total heat transferred (via latent heat and conductive losses) from the feed to the permeate (Alkhudhiri et al. 2012). Thermal efficiency can be enhanced by adequate membrane thickness, high feed temperature, and flow rates (Al-Obaidani et al. 2008; Duong et al. 2015).

Temperature polarisation is the difference between interface temperatures and the bulk temperatures (Zhang 2011). Temperature polarisation causes the resistance in the boundary layer of the membrane. Temperature polarisation coefficient can be given as in the following equation (Ge et al. 2014).

\[ \text{TPC} = \frac{T_{mf} - T_{mp}}{T_{bf} - T_{bp}} \] (20)

2.2. Mass transfer

Mass transfer through the hydrophobic single-layer membrane occurs in three regions: mass transport through boundary layers at the feed side, mass transfer through membrane pores, and mass transfer from membrane surface to the permeate side. Although dual-layer membranes have an additional hydrophilic layer, the equations remain the same as for hydrophobic single-layer membranes. The hydrophilic layer of the membrane is assumed to be filled with water, and the velocity of water flow through the hydrophilic layer is assumed to be low, so that the hydraulic resistance to mass transfer from the hydrophilic layer can be neglected.

Mass flux across the membrane can be calculated by the following equation:

\[ J = C_m(p_{v_{mf}} - p_{v_{mp}}) \] (21)
where $C_m$ is the mass transfer coefficient, and $p_{vm,f}$, $p_{vm,p}$ are the partial pressures on the feed and permeate sides, respectively. Partial pressures can be calculated by the Antoine equation in the following equation.

$$p_v = \exp \left(23.528 - \frac{3.841}{T - 45}\right)$$  \hspace{1cm} (22)

where $T$ is the mean temperature, $(T_{b,f} + T_{b,p})/2$, at the membrane interface.

Depending upon Knudsen number,

$$K_n = \frac{l}{d}$$  \hspace{1cm} (23)

the mass transfer coefficient can be chosen. If Knudsen number is greater than 1, Knudsen flow dominates and $C_m$ can be described as follows (Khayet et al. 2004):

$$C_m = \frac{2}{3} \frac{\varepsilon \tau \varepsilon_t \delta_t}{\tau \delta_t} \left(\frac{8M}{\pi RT}\right)^{1/2}$$  \hspace{1cm} (24)

If $K_n < 0.01$, molecular diffusion dominates and $C_m$ can be described as follows:

$$C_m = \frac{\varepsilon \tau \varepsilon_t \delta_t}{\tau \delta_t} \frac{P D M}{P_a \pi RT}$$  \hspace{1cm} (25)

If $0.01 < K_n < 1$, Knudsen/molecular diffusion mechanism dominates mass transport (Qtaishat et al. 2008; Soni et al. 2009),

$$C_m = \left[\frac{3}{2} \frac{\varepsilon \tau \varepsilon_t \delta_t}{\tau \delta_t} \left(\frac{\pi RT}{8M}\right)^{1/2} + \frac{\varepsilon \tau \varepsilon_t \delta_t}{\tau \delta_t} \frac{P_a RT}{\pi PD M}\right]^{-1}$$  \hspace{1cm} (26)

where $\delta_t$, $\tau$, $\varepsilon_t$, and $r_{ps}$ are the thickness, tortuosity, porosity, and pore size of the top hydrophobic layer of the membrane. $P_a$ is the air pressure, $P$ is the total pressure inside the pore, $M$ is the molecular weight of the water, $R$ is the gas constant, and $T$ is the absolute temperature.

The mass transfer equations are considered to be the same for the single- and dual-layer membranes, since the hydrophobic layer of the dual-layer membrane was placed on the feed side.

During the mass transfer, salt ions cannot pass through the membrane, which causes the accumulation of salt ions in the feed and the increase of salt concentration near the membrane surface. This phenomenon is termed as concentration polarisation (Hitsov et al. 2015). Concentration polarisation occurs in the MD feed channels and reduces transmembrane flux. However, for low concentrations of feed solution, this phenomenon might be negligible, as the vapour pressure is not greatly affected. Concentration polarisation does not give burden computationally to the model.

### 3. EXPERIMENTAL

The characterisation of membranes and DCMD experiments were undertaken. Characteristics of the membranes were undertaken to determine the porosity, thickness, pore size, and contact angles of the membranes. The membrane used for the tests were commercially available single-layer polyethylene (PE) membrane (Aquastill) and a bespoke dual-layer PVDF-co-HFP-N6 (PH/N6) membrane fabricated via electrospinning and sourced from the University of Technology Sydney. The membranes used, their code names, and the composition are listed in Table 1.
3.1. Membrane characterisation tests

As Equation (27) shows, the mass flux depends on membrane properties such as pore size \( r \), porosity \( \varepsilon \), thickness \( \delta \), and tortuosity \( \tau \) (Zhang et al. 2010), i.e. \( \alpha \) is the exponent of pore size which is in the range of 1–2.

\[
C_m \propto \frac{r^\alpha \varepsilon}{\delta \tau}
\]  
(27)

It shows by correlation above, increasing the porosity and pore size of the membrane increases the permeate flux, whereas increasing tortuosity and thickness reduces the permeate flux. Therefore, the ideal membrane should have higher porosity, adequate pore size (between 0.1 and 0.5 + 0.08 µm; Hou et al. 2012; Prince et al. 2013), and lower thickness and tortuosity. These characteristics are required in the modelling and are measured in this study.

3.1.1. Porosity test

Porosities of the membranes were measured using acetone and water due to their different wetting properties for the hydrophobic and hydrophilic layers (Zhang 2011). The water wetted the hydrophilic layer and the acetone wetted both the hydrophilic and hydrophobic layers. The mass differences of a dry and wet membrane were used to determine the porosity of the hydrophilic and hydrophobic layers. Water was first used to measure the porous volume of hydrophilic membrane support layer, so that the porosity of the hydrophilic layer could be determined using the hydrophilic layer thickness and area. The volume of the membrane \( V_m \) and support layer \( V_{support} \) was found using acetone, as it wetted both the membrane and hydrophilic layer pores. The porous volume of the support layer was calculated by the following equations:

\[
V_{support} = \frac{m_{support}}{\rho_{support}}
\]  
(28)

\[
V_m - V_a = V_{support}
\]  
(29)

\[
b = \frac{V_a}{A}
\]  
(30)

\[
e = 1 - \frac{(m_{total} - m_{support})/\rho}{(V_m - V_{support})}
\]  
(31)

where \( V_a \) is the active layer volume, \( m_{support} \) is the mass of the support layer, \( \rho_{support} \) is the density of the support layer, \( \rho \) is the membrane density, and \( m_{total} \) is the membrane mass with active and support layers (Zhang et al. 2010).

The density of active layer can be calculated from the following equation:

\[
\rho_{active\ layer} = \frac{m_{active\ layer}}{V_{flask} - \left( m_{acetone\ +\ membrane} - m_{active\ layer} \right) / \rho_{acetone}}
\]  
(32)

where \( m_{active\ layer} \) is the active layer mass, \( V_{flask} \) is the volume of the volumetric flask, \( m_{acetone\ +\ membrane} \) is the total mass of the acetone and membrane in the volumetric flask.

3.1.2. Pore size (porometer)

Mean pore size, maximum pore size, minimum pore size, and pore size distributions of the membranes were determined by Porometer Quantachrome 3GZ. Wet and dry run were conducted consecutively. Isopropyl
alcohol was used as wetting liquid due to its low surface tension. Test was conducted with increasing the transmembrane pressure, gas was sent through the holder, and the gas flow was measured as a function of the transmembrane pressure.

3.1.3. Thickness (scanning electron microscope)
Scanning electron microscope (SEM) was used to observe the membrane surface and membrane cross-sections. The thicknesses of the membranes were measured from membrane cross-sections. Membranes were frozen in liquid nitrogen and then cut with a blade to exhibit the clean cross-sections.

3.1.4. Contact angle measurement
Hydrophobicity of the membranes was measured by Contact Angle Analyser Kruss DSA25. Contact angles of the membranes were determined by the sessile drop method. A 4 μl drop was placed on the membrane surface, and the contact angle was determined using a camera and image analysis. The mean contact angle was determined from two replicate measurements. Both sides of the dual-layer membrane were tested.

3.2. MD test
Figure 2 shows a schematic diagram of the experimental set-up for the DCMD tests similar to that used in Zhang (2011). It consisted of a membrane module, heater, a chiller, the feed and permeate pumps, a balance, and a conductivity meter. 1% w/w (10 g/L) salt concentration (saline water) was used as feed solution. The feed solution was held in the feed tank, and the permeate solution was collected in the product reservoir. The product reservoir was weighed throughout the experiment. DCMD tests were run for 4 h for each experimental condition, and inlet and outlet temperatures of the feed and permeate streams and the module inlets and outlets were recorded by temperature logging. The conductivity of the permeate solution was recorded throughout the experiment.

DCMD tests were conducted at a variety of different feed and permeate inlet temperatures and flow rates for different membranes. The various experimental conditions can be found in Table 2. For all the experiments, the mass flow rates for both the feed and permeate channels were kept the same in each experiment.

Experimental conditions were kept constant throughout each experiment. Effects of different experimental conditions on flux and energy efficiency were examined and modelled. Membrane dimensions of M1 and M2 were 135 × 135 mm and 135 × 95 mm (L × W), respectively. As the width of the M2 membrane differed from the M1 membrane, two different module arrangements were used to accommodate the variations in the size of membrane samples between the single- and dual-layer membranes. The module and spacer dimensions can be found in Table 3. These dimensions were used in modelling the DCMD process.
4. RESULTS AND DISCUSSION

4.1. Membrane characterisations

Measured membrane characteristics are given in Table 4, and Figures 3 and 4 show the SEM images of the M1 and M2 membranes.

Characteristics of the membranes were used in the modelling to calculate the mass transfer and conductive heat transfer coefficients for the M1 single-layer and M2 dual-layer membranes.

M2 dual-layer membrane had more porous surface than M1 single-layer membrane, as it can be seen from Figure 3(a)–3(c). High porosity can lower conductive heat flux and increase the water vapour transport coefficient through the membrane (Zhang et al. 2010). Although it is desirable to have high porosity for better MD flux, the other characteristics such as pore size and thickness are also crucial factors.

Contact angles of the membranes indicate their hydrophobicity, and the hydrophobic layers of both M1 and M2 were greater than 100°. Hydrophobic membranes are desirable for DCMD to prevent wetting, so that membrane can be used for a longer time. Although contact angle values were not used in the modelling, it was important to see their hydrophobicity in order to assume the character of wetting tendency when contacting the feed.

SEM surface images of membranes show surface porosities, while images of cross-sections identify the membrane thicknesses. Thickness can be a limitation for mass transfer across the membrane, and lower permeate flux might be the reason for this.

From the images below, it can be seen that the hydrophobic and hydrophilic layers of M2 membrane had more porous surface than M1 membrane.

| Table 2 | Different experimental conditions for DCMD tests
| Membranes | Flow type | Flow rates (ml/min) | Velocity (m/s) | Feed inlet temperature (°C) | Permeate inlet temperature (°C) |
|-----------|-----------|---------------------|----------------|-----------------------------|-------------------------------|
| M1        | Co-current| 530                 | 0.093          | 60                          | 20                            |
|           |           | 600                 | 0.105          | 70                          |
| Counter-current| 530   | 0.093          | 60                          | 20                            |
|           |           | 600                 | 0.105          | 70                          |
| M2        | Co-current| 450                 | 0.079          | 50                          | 10                            |
|           |           | 530                 | 0.093          | 60                          | 20                            |
|           |           | 600                 | 0.105          | 70                          | 30                            |
|           |           | 750                 | 0.131          | 80                          |
| Counter-current| 450   | 0.079          | 50                          |
|           |           | 530                 | 0.093          | 60                          |
|           |           | 600                 | 0.105          | 70                          | 20                            |
|           |           | 750                 | 0.131          | 80                          |

| Table 3 | Spacer and module dimensions
| Flow channel (mm) | Width | Spacer (mm) |
|-------------|-------|-------------|
| Length (L) | (W1) | (W2) | Depth (D) | Filament diameter (d_f) | Thickness (h_sp) | Mesh size (l_m) |
| 135         | 135   | 95   | 0.8      | 0.4                  | 0.8              | 3               |

| Table 4 | Characteristics of the membranes
| Membrane codes | Porosity (%) | Contact angle (°) | Maximum pore size (μm) | Mean pore size (μm) | Thickness (μm) |
|-------------|---------------|------------------|------------------------|---------------------|-----------------|
| M1          | 75            | 101              | 0.39                   | 0.21                | 14.3            |
| M2 Hydrophilic layer | 91 | 50 | 0.44 | 0.25 | 13.6 |
| M2 Hydrophobic layer | 81 | 111 | 0.39 | 0.31 | 93.9 |
The layers of M2 were detached to allow SEM imaging to identify the layer thicknesses more easily, where hydrophobic and hydrophilic layers are represented as red and black arrows, respectively. The hydrophobic layer was thicker than the hydrophilic layer, as shown in Figure 4(b).

The differences in thicknesses of hydrophobic layers of M1 and M2 can be seen in Figure 4(a) and 4(b). The hydrophobic layer of M2 was thicker than the hydrophobic layer of M2. However, the thinness of the membranes was preferable over thicker membranes in DCMD due to membrane performance.

4.2. Model validation

Programmes were written using MATLAB to solve the heat and mass transfer equations given in Section 2. A flow chart for the solution procedure is given in Supplementary Appendix. The prediction of the permeate flux was undertaken at different feed inlet temperatures and at different flow rates for the M1 and M2 membranes. The predicted model permeate fluxes at different flow rates and feed inlet temperatures for M1 and M2 were validated with experimental results. In the following figures, the experimental results are presented with points and the modelling results with lines.

Following assumptions have been made to simplify the coding for predicting the flux and thermal efficiency in the model. These assumptions were:

1. The temperature gradient across the width of the membrane was neglected.
2. Heat loss through the membrane module to the environment was neglected.
3. Sensible heat transferred by the permeate is neglected.
4. Permeate mass passing the membrane is neglected, as the single-pass recovery is approximately 5%.
5. Concentration polarisation effect was neglected due to the low concentration of salt used in the experiments compared to the concentration at which significant vapour pressure depression occurs. Hence, the model is only valid for low salt concentrations.

Temperature profiles of M1 and M2 were observed along the membrane length for co- and counter-current flows and can be seen in Figures 5 and 6. Temperature change between feed and permeate bulk is decreasing parallel to
each other for counter-current flow, whereas co-current temperatures are approaching each other. $T_{bf}$ and $T_{bp}$ are the bulk temperatures at the feed and permeate side, and $T_{mf}$ and $T_{mp}$ are the membrane interface temperatures at the feed and permeate, respectively. Temperatures at the interface of the membranes were calculated by the model.

Temperature profiles for single- and dual-layer membranes were found to be similar; however, temperature profile for M2 has another temperature point between the interfaces of hydrophilic layer and permeate solution, which is $T_{sp}$.

The temperature difference between $T_{sp}$ and $T_{bp}$ was observed to be very small because the thin hydrophilic layer of the membrane was filled with water, in which only conductive heat transfer takes place without latent heat transfer.

### 4.2.1. Predicting the permeate flux at different temperatures and velocities

The permeate flux increased for both membranes when increasing the flow rate due to enhanced turbulence along the channel. Figure 7 shows the flux change at different flow rates with co-current flow at 60 °C feed and 20 °C permeate inlet temperatures.
Experimental reproducibility of the experimental results was within the maximum error range, which was 10%. The permeate flux was observed to be higher with M1 membrane at different flow rates. Figure 8 shows the flux change at different flow rates for counter-current flow. The trend for flux was similar compared to co-current flow with both M1 and M2 membranes. M2 achieved its highest flux at 0.105 m/s with counter-current flow. Figure 9 shows the permeate fluxes at different feed inlet temperatures for M1 and M2.

Increasing the feed temperature while keeping the permeate inlet temperature at 20 °C increased the permeate flux for both membranes M1 and M2. M1 membrane achieved higher permeate flux than M2 membrane. Lower flux for the M2 dual-layer membrane can be correlated to heat and mass transfer coefficients that are based on the membrane characteristics. Heat and mass transfer across the membrane were limited due to M2 membrane’s higher thickness. An additional hydrophilic layer of M2 has increased the thermal resistance and decreased
Figure 7 | Flux versus velocity at 60 °C feed inlet and 20 °C permeate inlet temperatures with co-current flow.

Figure 8 | Flux at different velocities at 60 °C feed inlet temperature and 20 °C permeate inlet temperature with counter-current flow.

Figure 9 | Flux at different feed temperatures with co-current flow for M1 and M2.
the overall heat transfer. The agreement between the model predictions and the experimental results was better than changing the feed inlet velocities as those in Figures 7 and 8.

Figure 10 shows the permeate flux versus feed inlet temperatures with counter-current flow. The increase for the flux in counter-current flow was not as high as the increment with co-current flow for M1 membrane because of the higher temperature difference between feed and permeate bulk solutions.

The model accuracy with the experimental results was better when changing the feed inlet temperature than feed and permeate velocities (see Figures 7–10). This is probably related to more accurate modelling for heat transfer through the membrane compared to mass transfer, particularly given the simplified assumption made for mass transfer through the hydrophilic support layer. Also, the model has more parameters that were influenced by temperature rather than velocity. The same sensitivity was shown by the experiments as the model. The reason was when the flow was fully developed, further increments of fluxes increase thermal differences marginally.

Having higher porosity, larger pore size, and thinner layer was favourable for higher flux. Although M2 membrane had the high porosity, the performance was lower than that of the M1 membrane. The thickness and relatively smaller pore size of the dual-layer membrane M2 could be the factors for the relatively poor mass transfer.

4.2.2. Validation of energy efficiencies at various temperatures and velocities

Energy efficiencies for hydrophobic single-layer and hydrophobic/hydrophilic dual-layer membrane were compared, and validation was made comparing the thermal efficiency obtained from calculations to the experimental data as shown in Figures 11 and 12. It can be seen that MD with co-current flow was more efficient
than counter-current flow. Single- and dual-layer membranes exhibited different energy efficiencies. As it can be seen from Equation (19), energy efficiency is the ratio between the heat transfer from the vapour transfer across the membrane to the heat loss from the feed. Figure 11 demonstrates the energy efficiency variation with the feed inlet temperature for co-current flow regime. M1 membrane was more energy-efficient than M2 membrane.

Lower energy efficiency for M2 membrane can be explained because of its low flux and hydrophilic layer. The temperature difference between interface and bulk temperatures was higher for dual-layer membrane compared to single-layer membrane, which increases the temperature polarisation coefficient according to Equation (20). The addition of the hydrophilic layer can be a limitation for heat transfer, as it increases conductive heat losses to the permeate so that it reduces the thermal efficiency of the membrane. Therefore, dual-layer membrane performance for permeate flux and energy efficiency was less than single-layer membrane.

The temperature difference between \( T_{s,p} \) and \( T_{b,p} \) was observed to be very small because the thin hydrophilic layer of the membrane was filled with water, in which only conductive heat transfer takes place without latent heat transfer. The hydrophobic layer thickness of the M2 membrane caused more heat loss across the membrane compared to the thinner M1 membrane. The greater thickness of the layer reduced the conductive heat transfer across the layer.

Thermal efficiencies with counter-current flow were lower than with co-current flow. The reason was the higher temperature difference between feed inlet and outlet due to the flow directions leading to greater localised temperature polarisation.

Thermal efficiencies of the DCMD system can be increased by increasing the membrane pore size and porosities, which can also lead to higher flux.

5. CONCLUSIONS

A novel 1-D numerical model for dual-layer membranes was developed and verified experimentally with a hydrophobic/hydrophilic Electrospun PH/N6 membrane. The model was aimed to predict the permeate flux and energy efficiency for DCMD.

Mass and heat transfer equations for M1 single-layer membrane and M2 dual-layer membrane were solved numerically. To do so, heat and mass transfer coefficients were calculated after measuring the membrane characteristics. The mass and heat transfer equations for the hydrophobic/hydrophilic dual-layer membrane accounted for conductive heat transfer across the hydrophilic layer and neglected mass transfer effects because the flow rate was assumed negligible.

The validation of the hydrophilic/hydrophobic dual-layer membrane model was achieved by comparing the experimental results with modelling results. The accuracy for the model was in general within 10% error (5% experimental error and 5% modelling error), which makes reliable DCMD performance modelling. The accuracy for flux variation at different velocities was less compared to that at different feed inlet temperatures. This identifies that the hydrodynamic simplifications for the dual-layer membrane lead to greater error in the heat transfer equations used.
The permeate flux increase was higher with the increase in feed inlet temperature compared to the increase in flow rate. Despite M2 membrane having higher porosity than M1 membrane, M1 membrane performed better when it came to mass flux and energy efficiency compared to M2 membrane. This 1-D model can be used to predict flux and thermal efficiency for either single or dual-layer membranes. This successful attempt of the 1-D model for dual-layer membranes can lead to in-depth analysis for heat and mass phenomena for membranes, which can also be extended to the 2-D model.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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