Predicting the performance of large-scale forward osmosis module using spatial variation model: Effect of operating parameters including temperature

JungGil Lee^{a,b} and Noredine Ghaffour^{a,*}

^aKing Abdullah University of Science and Technology (KAUST), Water Desalination and Reuse Center (WDRC), Biological and Environmental Science & Engineering Division (BESE), Thuwal 23955-6900, Saudi Arabia, Email: noredine.ghaffour@kaust.edu.sa

^b Thermal & Fluid System R&D Group, Korea Institute of Industrial Technology, 89 Yangdaegiro-gil, Ipjang-myeon, Seobuk-gu, Cheonan, 331-822, Republic of Korea

Abstract

Forward osmosis (FO) is considered as an energy-efficient process for numerous applications. Although its performance is determined by the spatially varied operation factors and the length of the channel, most of the reported simulation studies rely on length-averaged lumped models. Here, we introduce a one-D model based on heat and mass transfer and transport behavior for both bulk draw and feed channel flows. We find prediction results to be in good agreement with two different experimental results at inlet feed temperatures below 25 °C. However, the difference of water flux (J_w) and reverse salt flux (RSF) between measured and predicted data increases when both feed and draw temperatures also increase. Our theoretical simulation study first reveals that the feed temperature near the membrane active layer surface is the main factor for improving water and salt permeabilities. We find that, with a channel width of 0.3 m and a channel length of 2.5 m, J_w and RSF calculated using the length-averaged based lumped model are overestimated by 13.01% and 13.12%, respectively, compared to those obtained using our new spatial variation model. Our study demonstrates that the length-averaged based lumped model is not an appropriate simulation model to predict the performance of large-scale FO modules at lower inlet velocities.

Keywords: FO simulation, Spatial variation, Length-averaged lumped model, Influence of temperatures, water and salt permeabilities.

1. Introduction

Global demand for freshwater is set to exceed supply, due to the rapid urbanization, industrialization, and climate change resulting from global warming [1-3]. The amount of surface water available for use is also insufficient to meet the global demand [3]. Therefore, to mitigate the water scarcity, freshwater is produced by using traditional desalination and wastewater reuse technologies [1,4]. These two areas are growing very rapidly, [1], with a need to develop novel energy efficient technologies to reduce the water production cost and enhance the water quality. Extensive research is being undertaken to improve the overall productivity, quality, and energy efficiency [4]. Among the most recent technologies being developed, those using membranes such as forward osmosis (FO) have received interest due to their feasibility, high productivity and low energy consumption [4]. Reverse osmosis (RO) is recognized as an efficient water treatment process; it has been rapidly and successfully commercialized at a very large scale [5, 6]. However, the RO is still facing various operational challenges, such as the need for an extensive pretreatment requiring the use of many chemicals for mitigating the scaling and fouling problems, a high electrical energy consumption [1, 6]. FO is a natural mass transfer process with much lower energy consumption and reduced use of chemicals [7]. FO uses a dense, hydrophilic, semi-permeable membrane separating the freshwater spontaneously. The process is driven by the difference in osmotic pressure generated from the difference of feed (low) and draw (high) concentrations. This leads to a reduction in energy consumption, compared to traditional pressure-driven membrane separation techniques [8].

Although the FO process cannot be used to produce fresh water in a stand-alone system, it can be used for various applications with dilution and concentration purposes, in water treatment fields including desalination, landfill leachate, acid mine drainage, and oily wastewater plants [9-13]. For example, FO has found applications in the agricultural field, as a mean to dilute fertilizers for [7,14-17]. To further improve the performance of the FO process, several researchers have worked on the development of novel FO membranes [18-24], the optimization of the draw solution and concentration [10, 25, 26], the design of FO modules [27], and hybrid FO systems [27-30].

Concentration polarization (CP) phenomenon is well-known as the primary challenge leading to an inefficient operation affecting mainly the FO flux. The FO process can have three different configurations: the symmetric mode (symmetric dense membrane), the FO and PRO mode (i.e., FS facing the membrane's active layer and DS facing the porous support layer and vice-versa,

respectively). Among these configurations, in the FO mode, the structure parameter of the membrane support layer determines the internal CP (ICP) facing the draw solution, whereas the feed flow condition determines the external CP (ECP) facing the feed solution.

Water flux (J_w) and reverse salt flux (RSF), as primary indicators of the FO process performance, are closely related to the properties of the membrane, ICP, ECP and the difference of transmembrane osmotic pressure. Due to these complex parameters, it is necessary to develop theoretical prediction models that predict the FO process performance and help to reduce the experimental runs and errors, especially at large module scale. Several theoretical studies of the FO process have been conducted to find the optimal operating conditions or module design, in order to improve the J_w and lower RSF [31-34]. Examples of such prediction models include those from Lee et al., [35] and Loeb et al., [36] for the FO water flux and the theoretical method from Tiraferri et al., [33] for the characterization of the water and salt permeabilities and the structural parameters of the FO membrane. You et al., [37] also studied the influence of temperature on J_w , both experimentally and theoretically. Chowdhury and McCutcheon [38] studied the influence of the inlet feed temperature on J_w and J_s and heat flux across the membrane. They presented the influence of the temperature gradient across the membrane on the FO performance by using general heat flux equation. However, they did not consider the temperature gradient which is caused by the heat transfer for axial direction in the channels. In FO system, the solution temperature can affect the FO performance and it changes along the spatial variation in flow channels due to the heat energy transport across the membrane. Therefore, in order to obtain high accuracy FO simulation data, temperature gradient with the spatial variation needs to be considered.

All the studied theoretical simulations are based on the FO membrane performance, using small lab-scale modules, and they are conducted using a length-averaged lumped model that doesn't take into account spatial variations, in both the draw and feed channels. Although the spatial variation of operating conditions is negligibly small at the lab-scale module, it cannot be neglected for large modules or in multi-stage pilot-scale concepts. This is because the diffusivity of the solutes in the solution and the osmotic pressure vary, depending on the salinity and temperature along the module channel, due to heat and mass transfer. The osmotic pressure is a function of the ion number, salinity, and temperature. The number of theoretical studies that consider spatial variations such as heat and mass transfer, and transport behavior, is low when

applied to FO, especially coupled with a temperature gradient along the module channel. Our motivation for taking into account the temperature gradient, in our study, is the wide variety of FO niche applications, using FO feed and draw solutions with different temperatures [6-38]. The temperature of the exhausted waste water or impaired quality water from industry or the municipality that could be used as FO feed or draw solution may be higher than the atmospheric temperature, thus impacting on the performance of FO [38-41].

This work aims to develop an accurate, integrated FO simulation model which takes into account the mass, momentum and energy balances on both draw and feed flows coupled with heat and mass transfers on the J_w and the RSF. Experimental and theoretical investigations are simultaneously performed to develop an accurate simulation model. Predicted results from two different models (the length-averaged lumped model and the spatial variation model presented here) are compared to better understand the differences between results obtained by the two models at various channel widths and lengths. The influence of temperature of the draw and feed sides on the J_w and RSF, in connection with the different thermal properties, are also investigated. In addition, water and salt permeabilities, at different feed and draw temperatures, are re-estimated. Re-estimated data are mapped to derive the water and salt permeabilities correlations, according to the applied temperatures for accurate prediction. This study allows us to understand, in a comprehensive manner, the influence of the heat and mass transfers and their transport behavior on the FO J_w and RSF.

2. Materials and methods

2.1. Experimental set-up

Fig. 1 showed schematic of the FO experimental set-up. Temperatures of the feed and draw solutions are kept constant during the experiments by using temperature controllers (chiller and boiler). Peristaltic pumps supply the draw and feed solutions to both inlet channels, flowing in a counter-current fashion. The initial volume of the draw and feed solutions is 2 L each. Two conductivity meters measure the conductivities of both the feed and the draw solutions, in real time. Data acquisition is performed via a connection between the computer and the balance and conductivity meters, in real time. The channel dimensions of the FO module are 2 cm x 7 cm x 0.2 cm (width x length x height).

The thicknesses of the TFC-PA membrane's active and support layers (0.86 μm (averaged value) and 99.14 μm , respectively) are measured using SEM image analysis (Carl Zeiss: Merlin^{TM} Field Emission Scanning Electron Microscope), as shown in **Fig. 2**. The membrane thickness is found to be approximately 100 ± 1 μm , measured by a digital micrometer (Digital Micrometer 0.001, 12.7 mm Electronic Thickness Gauge Depth LCD Measurement). The thermal conductivity of PA is 0.27 W/mK [42]. The porosity of the support layer measured using the gravimetric method is $63\% \pm 5$. The weight of isopropyl alcohol, which has a density of 786 kg/m^3 as an organic solvent in the pores, is employed to measure the porosity of the support layer [43].

The latter is defined as the volume of the pores divided by the total volume of the membrane, and can be expressed as follows:

$$\varepsilon_m = \frac{(w_1 - w_2) / D_i}{(w_1 - w_2) / D_i + w_2 / D_p} \quad (1)$$

where w_1 and w_2 are the weights of the wet and dried membranes, respectively and D_i and D_p are the densities of isopropyl alcohol and polymer, respectively.

Figure 1. Schematic of the experimental set-up.

Figure 2. Cross-section SEM image of the TFC-PA membrane's active and support layers provided by Toray Co.

The thermal conductivity of the TFA-PA FO membrane is calculated using Eq. (2).

$$k_{overall} = \left(\frac{\delta_{active}}{k_m} + \frac{\delta_{support}}{\varepsilon_{support} k_w + (1 - \varepsilon_{support}) k_m} \right)^{-1} \quad (2)$$

where δ_{active} and $\delta_{support}$ are the active and support layer thicknesses of the membrane, respectively, $k_{overall}$, k_m , and k_w are the thermal conductivities of the overall, the membrane, the water, and the support layer, respectively, and $\varepsilon_{support}$ is the porosity of the support layer.

3. Theoretical approach

The main driving force of the FO process is the osmotic pressure difference, which is affected by both the concentration and the temperature between the feed and draw solutions (**Fig. 3**). The temperature and concentration of the draw and feed sides can vary, due to the heat and mass transfers across the semi-permeable membrane. Furthermore, these variations can influence the overall performance of the FO system. Therefore, in our study, we use a rigorous one-dimensional simulation model integrating heat and mass transfers coupled with a transport behavior developed by considering spatial variations in both draw and feed flows. The main assumptions for the simulation study are: (i) conduction and convection heat loss to the ambient is negligible; (ii) internal flows are incompressible in steady state; and (iii) roughness of the membrane surface and module at the draw and feed sides is not considered.

Figure 3. Schematic diagram of the FO process.

The basic equation of an ideal J_w in FO can be expressed as:

$$J_w = A(\pi_{D,b} - \pi_{F,b}) \quad (3)$$

where A is the water permeability coefficient of the membrane, $\pi_{D,b}$ and $\pi_{F,b}$ are the osmotic pressures of the bulk draw and feed sides, respectively.

Eq. (3) expresses the ideal J_w without any negative CP effect on the draw or feed sides. However, in a real operation, CP and RSF do affect the performance of the FO process. Therefore, the basic equation of J_w is modified, when considering the ICP (facing draw solution) and ECP (facing feed solution) effects as well as the RSF on the FO performance. It then becomes the following [44]:

$$J_w = A \left(\frac{\pi_{D,b} \exp\left(-\frac{J_w S}{D_{NaCl}}\right) - \pi_{F,b} \exp\left(\frac{J_w}{k}\right)}{1 + \frac{B}{J_w} \left[\exp\left(\frac{J_w}{k}\right) - \exp\left(-\frac{J_w S}{D_{NaCl}}\right) \right]} \right) \quad (4)$$

where $S = \varepsilon_{\text{support}} / \tau \delta_{\text{support}}$ (τ is the tortuosity) is the structural parameter of the membrane support layer, D_{NaCl} is the bulk solute (NaCl) diffusion coefficient, $k = Sh D_{NaCl} / d_h$ (Sh is the Sherwood number) is the mass transfer coefficient, B is the solute (NaCl) permeability coefficient of FO membrane, and $\pi_{D,m}$ and $\pi_{F,m}$ are the osmotic pressure of draw and feed adjacent membrane sides, respectively.

In a real operation, the transmembrane concentrations are determined from the negative CP effects. When the FO mode is used, the ICP appears adjacent to the surface of the transmembrane facing the draw solution, and the ECP occurs adjacent to the surface of the transmembrane facing the feed solution. The transmembrane concentration of draw and feed sides can be calculated using the following equations (Eqs. 5 and 6):

$$\frac{\pi_{D,m}}{\pi_{D,b}} = \exp\left(-\frac{J_w S}{D_{NaCl}}\right) \quad (5)$$

$$\frac{\pi_{F,m}}{\pi_{F,b}} = \exp\left(\frac{J_w}{k}\right) \quad (6)$$

Similarly, the RSF (J_{SR}) can be expressed as [44]:

$$J_{SR} = B \left(\frac{c_{D,b} \exp\left(-\frac{J_w S}{D_{NaCl}}\right) - c_{F,b} \exp\left(\frac{J_w}{k}\right)}{1 + \frac{B}{J_w} \left[\exp\left(\frac{J_w}{k}\right) - \exp\left(-\frac{J_w S}{D_{NaCl}}\right) \right]} \right) \quad (7)$$

where $c_{D,b}$ and $c_{F,b}$ are the bulk solute concentrations of draw and feed sides, respectively.

The diffusion coefficient of NaCl (D_{NaCl}) does not vary more than 3%, within the 0.05-2.0 M range [44]. The correlation of diffusion coefficient for NaCl, according to the temperature, is then derived by using the provided data in [45] and the linear interpolation, and expressed as:

$$D_{NaCl} = 0.0419((T_{D,b} + T_{F,b}) / 2) + 0.4797 (10^{-9} \text{ m}^2\text{s}) \quad (8)$$

where $T_{D,b}$ and $T_{F,b}$ are the bulk temperatures of the draw and feed sides, respectively.

Figure 4. Schematic diagram of mass transfer in the FO process.

A schematic diagram of the mass balance is presented in **Fig. 4**; the respective equations of mass balance at draw and feed sides can be written as follows:

$$x_{D,out} \dot{m}_{D,out} - x_{D,in} \dot{m}_{D,in} = x_{D,in} J_{SR} - x_W J_W \quad (9)$$

$$x_{F,out} \dot{m}_{F,out} - x_{F,in} \dot{m}_{F,in} = x_{D,in} J_{SR} - x_W J_W \quad (10)$$

where $x_W = 0$, due to the assumption that the FO process allows the transport of pure water only across the membrane, $x_{D,in}$, $x_{F,in}$, $x_{D,out}$ and $x_{F,out}$ are the salinities of inlet draw, inlet feed, outlet draw, and outlet feed, respectively, $\dot{m}_{D,in}$, $\dot{m}_{F,in}$, $\dot{m}_{D,out}$ and $\dot{m}_{F,out}$ are the flow rates of inlet draw, inlet feed, outlet draw, and outlet feed, respectively.

3.2 Heat transfer

Figure 5. A schematic diagram of heat transfer in the FO process.

Fig. 5 shows a schematic diagram of heat transfer phenomena in the FO process. The heat transfers from the hot to the cold side through the membrane. Temperature of the feed and draw solutions can affect the difference in osmotic pressure (driving force); it may also affect the water and salt permeabilities of the active membrane layer [46]. If the draw and feed temperatures are different, the heat transfer phenomena should be considered in the FO process. The heat transfers from the hot side to the cold side; this heat transfer is the result of the combination of both the mass transfer (J_w and RSF), and the conduction heat transfer with the sensible heat transfer. Thus, the heat transfer equations in the FO process can be expressed by using the following (i, ii, and iii).

(i) Heat transfer from bulk feed to adjacent feed boundary layer

$$Q_F = h_F (T_{F,b} - T_{F,m}) \quad (11)$$

where $T_{F,b}$ and $T_{F,m}$ are the bulk and transmembrane temperatures at feed side, respectively, and h_F is the convection heat transfer coefficient calculated by a combination of Reynolds number, Re_i and Nusselt number, Nu . It can be expressed by the following equation:

$$h_i = \frac{Nu_i \cdot L}{d_h} \quad i = \text{draw and feed sides} \quad (12)$$

$$Nu_i = 1.85 \left(Re_i Pr_i \frac{d_h}{L} \right)^{0.33} \quad (\text{laminar flow, } Re < 2300)$$

$$Nu_i = 0.023 Re_i^{0.75} Pr_i^{0.33} \quad (\text{turbulent flow, } Re > 2300)$$

where L is module length, and Pr_i is the Prandtl number.

(ii) Heat transfer across the membrane

$$Q_m = (J_w c_p + k_{overall}) (T_{F,m} - T_{D,m}) \quad (13)$$

where c_p is the heat capacity, and $T_{D,m}$ is the transmembrane temperature at draw side.

(iii) Heat transfer from the adjacent draw boundary layer to feed bulk

$$Q_D = h_D (T_{D,m} - T_{D,b}) \quad (14)$$

where h_D and $T_{D,b}$ are the convection heat transfer coefficient and the bulk temperature at draw side, respectively.

At steady state, the overall heat transfer through the whole FO process is given by:

$$Q_F = Q_m = Q_D \quad (15)$$

3.3 Transport behavior

At both draw and feed sides, the momentum, mass, species and energy balances can be expressed by integrated differential equations for the pressure, velocity, salinity and temperature, as follows:

$$\frac{dP_i}{dz} = - \frac{3\mu_i}{h_{c,i}^2} v_i \quad (16)$$

$$\frac{d(\rho_i v_i)}{dz} = - \frac{(J_w - J_{SR})}{h_{c,i}} \quad (17)$$

$$\frac{d(\rho_i v_i x_i)}{dz} = - \frac{(x_w J_w - x_D J_{SR})}{h_c} \quad (18)$$

$$\frac{d(\rho_i v_i c_{p,i} T_i)}{dz} = - \frac{Q_i}{h_{c,i}} \quad (19)$$

where the subscription i is the draw (D) or feed (F), μ is the viscosity, ρ is the density, v is the

velocity, h_c is the channel height, and dz is the grid size.

Boundary conditions for the pressure, velocity, salinity, and temperature, at both draw and feed sides, respectively, are as follows:

$$P_D(L) = P_0, v_D(0) = v_{D,in}, x_D(0) = x_{D,in}, T_D(0) = T_{D,in} \quad (20)$$

$$P_F(0) = P_0, v_F(L) = v_{F,in}, x_F(L) = x_{F,in}, T_F(L) = T_{F,in} \quad (21)$$

where P_0 is the atmospheric pressure.

4. Modeling procedure

Fig. 6 presents a schematic diagram of the solution procedure. The set of integrated ordinary differential equations, for both draw and feed sides, are discretized by using the finite volume method (FVM) and calculated using both the boundary conditions at the draw and feed sides and the characterization of the FO membrane for mass transfer. Broyden's method is used to solve the system of ordinary differential equations.

5. Results and discussion

5.1 Model validation

In order to validate our spatial variation model, predicted results are compared with measured data reported in the literature [44]. **Fig. 7** presents both experimental and simulated results of the flux and the RSF, at various draw concentrations. The water and salt permeabilities and structural parameter of the FO membrane that were used in our study are $1.23 \text{ kgm}^{-2}\text{h}^{-1}\text{bar}^{-1}$, $0.196 \text{ kgm}^{-2}\text{h}^{-1}$, and $328 \text{ }\mu\text{m}$, respectively [44]. We find the predicted data are well matched with the experimental data, at constant inlet draw and feed temperatures of 25°C and velocity of 17.1 cm/s .

Figure 6. Schematic diagram of the solution procedure.

Figure 7. Effect of the inlet draw and feed concentrations on J_w and RSF (experimental [44] and simulated), at constant inlet draw and feed temperatures and velocities of 25°C and 17.1 cm/s, respectively.

Table 1 presents a comparison of real data with predicted data, with a length-averaged lumped model and with our model. Details of the operating conditions (case 1 to case 4) can be seen in Fig. 7. As shown in the table 1, predicted data for J_w and the RSF, using a length-averaged lumped model, have average errors of 2.1% and 2.4%, respectively. On the other hand, average errors of 1.5% and 1.4%, are obtained when using our new model of spatial variation for both draw and feed channels. We find that the difference obtained by the two different models is insignificant, due to the negligible concentration gradient along the feed and draw channels, especially on short channel length. This is because the J_w and the RSF are much smaller than the inlet draw and feed flow rates.

Table 1. Influence of inlet draw and feed salinities on the flux and RSF, for both measured and predicted values using either the length-averaged lumped model or spatial variation model).

Stage	Real (St.)	Length-averaged lumped model	Current developed spatial variation model	Relative Difference		Real (St.)	Length-averaged lumped model	Current developed spatial variation model	Relative Difference	
	J_w (kg·m ⁻² h ⁻¹)			(%)	(%)	J_{SR} (g·m ⁻² h ⁻¹)			(%)	(%)
Case1	8.92	9.04	9.04	1.35	1.35	1.66	1.63	1.68	1.81	1.20
Case2	12.01	11.64	12.18	3.08	1.42	2.24	2.20	2.27	1.79	1.34
Case3	14.9	15.03	15.14	0.87	1.61	2.78	2.69	2.82	3.24	1.44
Case4	17.35	16.81	17.65	3.11	1.73	3.24	3.33	3.29	2.78	1.54

Additionally, in order to validate our own experimental results with prediction data, using our spatial variation model, a validation of the model is performed at various inlet draw salinities. The TFC-PA FO membrane manufactured by Toray Korea is used in all experiments and simulations. Experiments are conducted four times under similar operational conditions to assess the reproducibility of the system performance. The uncertainty of the measured water fluxes is lower than ±10%.

Fig. 8 presents the experimental and predicted results of flux and RSF for our study that were obtained for an inlet draw and feed temperatures of 20°C, inlet draw and feed flow rates of 0.4

L/min, under various inlet draw concentrations ranging from 17.5 g/L to 87.5 g/L NaCl, using deionized water (DI) as feed solution. A, B and S values are $5.964\text{E-}7$ m/s/bar, $1.805\text{E-}7$ m/s and $3.76\text{E-}4$ m, respectively; the predicted results which were calculated by the spatial variation model are in good agreement with the experimental data. As shown in **Fig. 8**, at with a high draw solution, the increase in the J_w and RSF are lower than those obtained for a lower range of draw solution due to the CP effect.

Figure 8. Influence of the inlet draw NaCl salinity on flux and RSF, at constant inlet draw solution and feed temperatures of 20°C and constant inlet draw solution and feed flow rates of 0.4 L/min.

5.2 Influence of draw and feed temperatures

In this section, experimental and theoretical studies are performed with the goal of better understanding the influence of the inlet draw and feed temperatures (20 °C – 40 °C) on the J_w and the RSF, at constant inlet draw and feed flow rates of 0.4 L/min (A, B, and S values of $5.964\text{E-}7$ m/s/bar, $1.805\text{E-}7$ m/s and $3.76\text{E-}4$ m, respectively, and obtained experimentally using an inlet draw and a feed temperature of 20 °C (Fig. 9). Our spatial variation model is used to estimate the predicted results. The experimental results were reproducible within an experimental error of 10% (experiments conducted four times). When the inlet draw temperature increases from 20°C to 40°C, at a constant inlet feed temperature of 20°C, the measured J_w increases from 16.62

324 kg/m²h to 21.15 kg/m²h, and the predicted values also increase from 16.68 kg/m²h to 20.06
 325 kg/m²h. The maximum relative error percentage is 6.26%, at the highest draw solution
 326 temperature of 40°C. At the same time, the measured RSF increases from 6.23 g/m²h to 6.82
 327 kg/m²h and the predicted values also increase from 6.06 g/m²h to 6.84 g/m²h. The maximum
 328 relative error for RSF (2.82%) is observed at the lowest inlet draw and feed temperatures
 329 (20 °C).

Figure 9. Influence of the inlet draw and feed temperatures ranging from (a) 20°C (b) 30°C and (c) 40 °C on the J_{WP} and the RSF (measured and predicted), at inlet draw and feed flow rates of 0.4 L/min (A, B and S values obtained by our experiments were 5.964E-7 m/s/bar, 1.805E-7 m/s and 3.76E-4 m, respectively).

Although the average driving force (osmotic pressure difference) only increases by approximately 3% (from 29.36 bar to 31.27 bar), when increasing the inlet draw temperature from 20 °C to 40 °C at constant inlet feed temperature of 20 °C, the $J_{WP,Exp}$ increases by 21.42%, due to the increase of NaCl's diffusivity as the inlet draw temperature increases. A similar trend was reported by previous research studies [44] that showed that an increase of the draw and feed temperatures led to higher $J_{WP,Exp}$ and reverse salt flux values (more than anticipated), due to an increase in both feed and draw osmotic pressures and a higher diffusivity of NaCl. According to previous published study [45], the increase of the $J_{WP,exp}$ and the RSF, when increasing the temperature by a higher than the increase of the driving force, can be explained by a decrease of viscosity, increase of the diffusion coefficient and increase of the number of water molecules, due to an increase of temperature. However, when a constant inlet draw temperature of 40°C is used, the $J_{WP,Exp}$ increases from 23.57 $\text{kg/m}^2\text{h}$ to 27.93 $\text{kg/m}^2\text{h}$ (18%), as the inlet feed temperature increases from 20°C to 40°C. Meanwhile, the RSF increases from 7.86 $\text{g/m}^2\text{h}$ to 8.84 $\text{g/m}^2\text{h}$, under similar conditions. The increase of the flux and the RSF (measured) is found

unexpectedly higher than that obtained with predicted results when considering an increase of the NaCl's diffusivity as the inlet feed temperature increases. With our current estimation, the increased NaCl diffusivity is considered as an estimated correlation (Eq.8), according to the increase of the draw and feed temperatures, together with the module channel. The maximum difference between measured and predicted results, for the J_{WP} and that RSF are 19.19% and 13.25%, for inlet draw and feed temperatures of 40°C. These results indicate that the feed temperature facing the active layer may affect the pore size of the active layer, as well as the difference of osmotic pressure and NaCl diffusivity. This hypothesis was previously presented by Goosen et al. [46] whose work revealed a change in the membrane's pore size when the feed solution temperature increases from 30 °C to 40 °C. Other studies also found an optimum J_w and RSF under different draw solution temperatures [47]. In our study, we also find that the temperature near the membrane surface affects more the J_w and RSF than the bulk temperatures (Fig. 10).

Figure 10. Influence of the feed and draw temperatures predicted on the deviation of measured water flux.

^aD30F20 means draw temperature of 30 °C and feed temperature of 20 °C, and vice versa, ^bD40F20 means draw temperature of 40°C and feed temperature of 20 °C, and vice versa, and ^cD40F30 means draw temperature of 40 °C and feed temperature of 30 °C, and vice versa.

We find the measured J_w and RSF values to be very close (less than 7% difference) at 30°C and 20°C of the bulk draw and feed temperatures, respectively, in comparison to those obtained at 20°C and 30°, respectively. The transmembrane temperature of the draw and feed sides are estimated to be 25.7°C and 24.4°C for the two cases, respectively. Moreover, when tested at 40°C (bulk draw) and 30°C (bulk feed), we obtain J_w and RSF values that are also very similar (less than 4% difference) to those at 30°C (bulk draw) and 40°C (bulk feed). We estimate the temperatures adjacent to the membrane's draw and feed sides to be 35.7°C and 34.4°C, respectively. Similarly, at 40°C (bulk draw) and 20°C (bulk feed), the J_w and RSF values were also close (less than 11% difference) as compared to those at 20°C (bulk draw) and 40°C (bulk feed). In this case, the estimated transmembrane temperatures of feed and draw sides were 31.5°C and 28.8°C, respectively. Furthermore, at a higher temperature near the feed side of the membrane surface for all cases, the flux and RSF values were always higher compared to those at the draw side. We also found that the increase in feed temperature facing the active layer resulted in higher J_w and RSF as compared to that of draw temperature.

To determine the correlations of the water and salt permeabilities as a function of the draw and feed temperatures, these parameters were re-estimated by the iteration method. In this study, we assumed the change of the properties of structural parameter of the membrane support layer with temperature to be small, because the support layer has much larger pore size, porosity and thickness compared to the active layer. The two parameters A and B in Eqs. (4) and (7), respectively, were estimated from water and salt fluxes experimental data by using the iteration method for solving minimization problems subject to linear equality constraints within the current developed spatial variation model.

Table 2. Re-estimated water (A) and salt (B) permeabilities according to the inlet draw and feed temperatures, at a fixed structural parameter (S). $S = 3.76E-04$ m in all cases.

	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6	Case 7	Case 8	Case 9
	TD:20°C TF:20°C	TD:30°C TF:20°C	TD:40°C TF:20°C	TD:20°C TF:30°C	TD:30°C TF:30°C	TD:40°C TF:30°C	TD:20°C TF:40°C	TD:30°C TF:40°C	TD:40°C TF:40°C
A (m/s/ bar)	5.96E-7	6.05E-7	7.14E-7	7.01E-7	7.75E-7	8.43E-7	8.71E-7	9.06E-7	9.62E-7
B (m/s)	1.81E-7	1.82E-7	1.93E-7	2.19E-7	2.30E-7	2.27E-7	2.58E-7	2.62E-7	2.71E-7

JW,exp (kg/ m ² h)	16.62	18.27	21.15	19.71	22.18	24.58	23.57	25.64	27.93
JW,sim (g/m ² h)	16.67	18.27	21.15	19.71	22.18	24.58	23.57	25.64	27.93
JS,exp (kg/ m ² h)	6.23	6.58	6.82	7.16	7.64	7.68	7.86	8.34	8.84
JS,sim (g/ m ² h)	6.07	6.58	6.82	7.16	7.64	7.68	7.86	8.34	8.84
Error JW/JS (%)	0.3/2.64	0/0	0/0	0/0	0/0	0/0	0/0	0/0	0/0

Table 2 presents the re-estimated water and salt permeabilities, using the iteration method in accordance with the nine cases at fixed structural parameter for coefficient mapping. Each A or B value is estimated individually. Thereby, the error value of J_w/J_s is almost 0% except for case 1 by using iteration method. A, B and S in case 1 were estimated by using the method reported in [33].

As shown in **Table 2**, water and salt permeabilities increase by 161.41% and 149.72%, respectively, as the inlet draw and feed temperatures increase from 20 °C to 40 °C (2 cases). A and B values are defined as a correlation equation, according to the inlet feed and draw temperatures.

To apply the mapped water and salt permeabilities data to our spatial variation model, the correlation of water and salt permeabilities (for the nine cases) are derived as two dimensional (2D) polynomial equations, using the commercial software MatlabR2017b. **Fig. 11** shows the surface fitting of water and salt permeabilities versus the inlet draw and feed temperatures ranging from 20°C to 40°C, respectively. The following correlations for A and B are obtained:

$$A(T_D, T_F) = 3.08472 + 0.14158T_F - 0.03625T_D + 0.000267T_F^2 + 0.00192T_F T_D - 0.00675T_D^2 \quad (10^{-7} \text{ m/s/bar}) \quad (R^2 : 0.971) \quad (22)$$

$$B(T_D, T_F) = 0.8385 + 0.04517T_F + 0.00427T_D - 0.000115T_F^2 + 0.000005T_F T_D + 0.000035T_D^2 \quad (10^{-7} \text{ m/s}) \quad (R^2 : 0.98441) \quad (23)$$

Figure 11. Surface fitting of (a) water and (b) salt permeabilities for inlet draw solution and feed temperatures varying from 20 °C to 40 °C.

To validate the application of the derived correlations of water and salt permeabilities in the spatial variation model, the J_w and the RSF are calculated by using the derived correlations in simulation and then the prediction result was compared with measured data. We find the measured and predicted data to be in good agreement, as shown in **Fig. 12**. The maximum error for the J_w is 3.19% (case 2), and the maximum error for the RSF is 3.5% (case 7). These errors are probably mostly due to a limitation of the 2D polynomial curve fitting to the experimental

425 data.

426

427

428 **Figure 12.** Influence of inlet feed and draw temperatures on (a) J_w and (b)

429 RSF (measured and predicted results with mapped A and B).

430

431 *5.3 Comparison between length-averaged lumped model and spatial variation model*

432 To compare the length-averaged lumped model with the spatial variation model presented

433 here, we perform simulation studies using the two different simulation models, at different

434 channel widths ranging from 0.1 m to 0.3 m, and for channel lengths ranging from 0.5 m to 2.5

m. Simulations are performed at inlet draw and feed flow rates of 0.4 L/min, inlet draw and feed temperatures of 40 °C, and at an inlet draw salinity of 35 g/L. Re-estimated correlations of water and salt permeabilities (see Section 4.3) are used with a fixed structural parameter ($3.76\text{E-}4$). The simulation results estimated by the length-averaged lumped model which was validated with experimental data was utilized for the comparison study with the spatial variation model from the Ref. [44]. At a channel length of 0.07 m for the short module (small effective area), the distribution of the concentration between the inlet and outlet of the feed and draw sides is negligible; this is because the total water and salt fluxes are much smaller than the inlet draw and feed flow rates. Therefore, we cannot observe a significant difference between the results obtained by the two different models, due to the very low (insignificant) dilution and concentration of the draw and feed sides. In other words, a higher J_w , as well as a higher RSF, can also produce a higher distribution of the concentration between the inlet and outlet. Additionally, this difference in concentration at the draw and feed sides can be intensified by the decrease of the inlet draw flow rate due to the mass or the salinity balance in the draw and feed sides.

Our next step is to investigate, using the two different simulation models, the effects of channel width on the J_w and the RSF, at fixed inlet draw and feed volume flow rates of 0.4 L/min, for channel widths ranging from 0.1 m to 0.3 m, a module channel length of 0.5 m, inlet feed and draw temperatures of 40 °C, and inlet draw and feed salinities of 35 g/L and 0 g/L, respectively. Both inlet draw and feed velocities can be adjusted by varying the channel width when the fixed volume flow rate is used. The inlet draw and feed velocities can be decreased from 0.333 m/s to 0.111 m/s, with an increase of the channel width from 0.1 m to 0.3 m.

Fig. 13 presents the effect of channel width (from 0.1m to 0.3 m) on the predicted J_w and RSF, calculated using the two different models at inlet draw and feed temperatures of 40 °C, inlet draw and feed flow rates of 0.4 L/min, channel length of 0.5 m, and inlet draw and feed salinities of the 35 g/L and 0 g/L, respectively. The flux and RSF calculated with the length-averaged lumped model are higher than those obtained by our spatial variation model. In addition, the J_w and RSF calculated by the length-averaged lumped model decrease, as the channel width increases. We find that the difference increases from 1.16% to 3.34% for the J_w , and from 1.22% to 3.38% for the RSF, with an increase in channel width of 0.2 m (from 0.1 m to 0.3 m). This is because, in the FO process, the dilution and concentration of the feed and draw solutions between the inlet and outlet increase, as the inlet draw and feed velocities decrease

(higher residence time). This is a parameter (variation of the velocity distribution along the module) that is not accurately predictable using the length-averaged lumped model. Both simulation models also show, from the above results, that the J_w and RSF are not significantly changed when a high inlet volume flow rate and short channel length are used because the water and salt flux are still much lower than the inlet draw and feed flow rates.

Figure 13. Influence of the channel width on (a) J_w and (b) RSF estimated using the length-averaged based lumped model and our spatial variation model, at various channel widths, inlet draw and feed temperatures of 40 °C, inlet draw and feed flow rates of 0.4 L/min and inlet draw and feed salinities of 35 g/L and 0 g/L, respectively.

Figs. 14 (a) and (b) show the influence of the channel length on the flux and RSF, calculated using the two different models. The maximum difference of flux and RSF obtained using the two different models are 13.01% (J_w) and 13.12% (RSF), respectively, at a channel length of 2.5 m and a width of 0.3 m. This can be explained by the fact that the length-averaged based lumped model does not consider the variations of J_w and RSF along the module as a result of the distribution of concentration. We find that the flux and RSF decrease with an increase in the total channel length due to the dilution and concentration effects along the draw and feed sides. Although the two models do not show any significant variation of the results obtained for the

difference of J_w and RSF, when using a small module (short channel length), we find this difference to be more pronounced as the channel length increases (e.g., large module such as multi-stage membrane distillation (MD) concept), mainly due to the decrease of the transmembrane osmotic pressure (small $\Delta\Pi$) and the temperatures at the membrane's feed and draw interfaces.

Figure 14. Influence of the channel length on (a) the J_w and (b) the RSF, estimated using both the length-averaged based lumped model and our spatial variation model, at a channel width of 0.3

m, inlet draw and feed temperatures of 40 °C, inlet draw and feed flow rates of 0.4 L/min, and inlet draw and feed salinities of 35 g/L and 0 g/L, respectively.

Figure 15. (a) Local draw and feed salinities and (b) local J_w and RSF distributions along the module at a channel width of 0.3 m and length of 2.5 m, inlet draw and feed temperatures of 40 °C, inlet draw and feed flow rates of 0.4 L/min, and inlet draw and feed salinities of 35 g/L and 0 g/L, respectively.

The local draw, feed salinities, local J_w and RSF distributions are shown in **Figs. 15 (a)** and **(b)**. The local draw salinity decreases from 35 g/L to 20.25 g/L along the module channel of 2.5 m, due to the water permeation from the feed side, and at the same time the local feed salinity increases from 0 g/L to 0.014 g/L, due to the RSF effect (Fig. 5a). As shown in **Fig. 15 (b)**, the local J_w decreases from 27.26 kg/m²h to 21.41 kg/m²h and the local RSF increases from 6.67 g/m²h to 8.49 g/m²h, due to a decrease of local driving force resulting from the dilution and concentration effects along the module channel in the draw and feed sides. The distribution of the local draw and feed concentrations are shown in **Figs. 14** and **15**. This study also reveals that the length-averaged lumped model is not appropriate to predict the performance of lower inlet velocities and long channels (large module such as multi-stage concept), hence it cannot give accurate simulation results to scale-up the FO process.

6. Conclusions

A new theoretical model has been developed and validated to describe the influence of spatial variations on the FO flux and RSF. The integrated model involves the thermophysical properties of the feed and the draw solutions and membrane, heat and mass transfer mechanism across the membrane, and the mass, momentum, salinity and energy balances for the draw and feed flows. Theoretical predictions are validated and compared with experimental data from the literature [44] as well as our own experimental data. All predictions are found to be in a good agreement with the experimental results. However, the relative error of the J_w and RSF increases, as the inlet feed temperature increases. Both draw and feed transmembrane temperatures are calculated using our new spatial variation simulation model. This model reveals the effects of the temperatures of the adjacent bulk and feed membrane on the J_w and RSF, in comparison with the effects from the bulk feed and draw temperatures. We also find that at a higher feed temperature near the membrane surface the flux and the RSF are always higher. An assessment of the influence of the feed and draw temperatures on the water and salt permeabilities show a 161.41% increase of the water permeability and 149.72% increase of the salt permeability as both the inlet draw and feed temperatures increase from 20 °C to 40 °C.

We show that the length-averaged based lumped model overestimates the J_w and RSF respectively by up to 13.01% (a decrease from 27.5 kg/m²h to 23.9 kg/m²h) and 13.12%,

respectively, at channel length and width of 2.5 m and 0.3 m, respectively, in comparison with our model that considers the spatial variations along the module channel.

Our work clearly demonstrates that the length-averaged based lumped model is not an appropriate simulation model to predict the performance of the FO process, at lower inlet feed velocities and large modules (long module channel or multi-stage concept) that are key parameters for the FO process scale-up. Moreover, the influence of temperature of draw and feed solutions on the FO water and salt permeability must be considered to accurately estimate the prediction results.

7. Acknowledgments

The research reported in this paper was supported by funding from King Abdullah University of Science and Technology (KAUST), Saudi Arabia. The authors acknowledge help, assistance and support from the Water Desalination and Reuse Center (WDRC) staff, and Mrs. Elisabeth M. Lutanie, Science Editor, Writer, Publication Services and Researcher Support at KAUST for editing the paper.

8. Nomenclature

A	Water permeability coefficient of membrane [m/s·pa]
B	Solute (NaCl) permeability coefficient of membrane [m/s]
C_p	Heat capacity [J/kg·K]
$c_{D,b}$	Bulk solute concentration of draw side [mol/L]
$c_{F,b}$	Bulk solute concentration of feed side [mol/L]
D	Water diffusion coefficient [m ² /s]
d_h	Hydraulic diameter [m]
D_i	Density of isopropyl alcohol [kg/m ³]
D_{NaCl}	Bulk solute (NaCl) diffusion coefficient [m ² /s]
D_p	Density of the polymer [kg/m ³]
dz	Grid length [m]
h_F	Convection heat transfer coefficient at feed side [W/mK]
h_D	Convection heat transfer coefficient at draw side [W/mK]
h_c	Channel height [m]

564	J_W	Water flux [kg/m ² h]
565	J_{SR}	Reverse salt flux [g/m ² h]
566	k	Mass transfer coefficient [m/s]
567	$k_{overall}$	Overall thermal conductivity through the membrane [W/m ² K]
568	k_w	Thermal conductivity of water [W/m ² K]
569	k_m	Thermal conductivity of the polymer [W/m ² K]
570	L	Module length [m]
571	$\dot{m}$	Flow rate [L/min]
572	Nu	Nusselt number [-]
573	P	Pressure [Pa]
574	Pr	Prandtl number [-]
575	Re	Reynolds number [-]
576	S	Structural parameter of the membrane support layer [m]
577	Sh	Sherwood number [-]
578	$T_{F,b}$	Bulk temperature at feed side [K]
579	$T_{F,m}$	Transmembrane temperature at feed side [K]
580	$T_{D,b}$	Bulk temperature at draw side [K]
581	$T_{D,m}$	Transmembrane temperature at draw side [K]
582	v	Velocity [m/s]
583	w_1	Weight of wet membrane [g]
584	w_2	Weight of dried membrane [g]
585	x	Salinity [g/L]
586	μ	Viscosity [Pa·s]
587	δ_{active}	Active layer thicknesses of the membrane [m]
588	$\delta_{support}$	Support layer thicknesses of the membrane [m]
589	$\epsilon_{support}$	Support layer porosity [-]
590	ρ	Density [kg/m ³]
591	$\pi_{D,b}$	Osmotic pressure at bulk draw side [bar]
592	$\pi_{F,b}$	Osmotic pressure at bulk feed side [bar]
593	$\pi_{D,m}$	Osmotic pressure of draw adjacent membrane side [bar]

$\pi_{F,m}$ Osmotic pressure of feed adjacent membrane side [bar]

τ Tortuosity [-]

9. References

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