



## Performance Evaluation of Cellulose Tri Acetate-Polyamide Forward Osmosis Membrane: A Mathematical Model Approach

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### ABSTRACT

Forward Osmosis (FO) membrane is a recent membrane process used in many applications. One of the challenges in FO technology is membrane material, which suits the FO process. The selection of membrane material for FO membrane application is crucial as it determines the membrane efficiency. When characterizing the FO membrane, besides its physical and chemical properties, the intrinsic parameters; A, B, and S, which represent water permeability, solute permeability, and structural parameters, respectively, are also essential. Experiments evaluate these intrinsic parameters, which may require more work and cost. As an alternative, the mathematical model approach is beneficial in determining these intrinsic parameters. This research work applied mathematical models and was solved using Microsoft Excel to evaluate intrinsic parameters for lab-made Cellulose Tri Acetate-Polyamide (CTA-PA) membrane. Besides that, a mathematical model was also applied to predict the CTA-PA membrane performance in terms of water flux and its suitability for power generation. CTA-PA membrane with lower CTA content in the support layer exhibited higher values of A, B, and S. Lower CTA content also contributed to higher flux and higher power density as predicted by the model. The mathematical model was successfully applied in this work to determine the intrinsic parameters and predict the performance.

## 1. Introduction

Forward osmosis (FO) as a non-pressurized membrane process has gained increasing attention in wastewater treatment [1-4] and desalination [1,5,6]. Even the FO membrane has been produced commercially by a minimal company such as HTI, Oasys, Aquaporin, Porifera, Toray, modern Water, Toyobo, Fluid Technology Solution, and Trevi system [7]. The research still keeps increasing, especially on the membrane material. Polyethersulfone (PES), polysulfone (PSF) and Poly(vinylidene) Fluoride (PVDF) are the common polymer materials used for membrane fabrication [8-10], especially for the pressurized-driven membrane process. Most commercial pressure-driven membranes are

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unsuitable for the FO process due to a thicker porous substrate layer (plus a non-woven fabric layer for mechanical strength to withstand high pressure). The accumulation of draw solutes in this porous substrate layer, namely internal concentration polarization (ICP), creates a concentration barrier, thus reducing the adequate osmotic pressure, leading to a lower water flux [11]. ICP typically occurs in the porous substrate layer. Hence, optimization of the substrate layer is essential. Parameter  $S$  is a standard parameter used to evaluate the effectiveness of the substrate layer in the FO membrane, representing the membrane resistance to solute mass transport (i.e., draw solute). The lower  $S$  value indicates better FO performance, and  $S$  is generally linked to the physical properties of the membrane substrate layer as follows in Eq. (1) [12]:

$$S = \frac{\tau \times l}{\varepsilon} \quad (1)$$

It is clear that, from Eq. (1), the tortuosity factor ( $\tau$ ), thickness ( $l$ ), and porosity ( $\varepsilon$ ) of the substrate layer significantly affect the  $S$ . All of these characteristics (tortuosity, thickness, and porosity) can be controlled during the membrane substrate fabrication process. This  $S$  value can be calculated once  $\tau$ ,  $l$ , and  $\varepsilon$  values are measured by a series of experiments, which require more work and cost. Alternatively, the  $S$  value can be determined using a mathematical model related to mass transfer fundamentals. Other than the  $S$  parameter, the other two essential parameters to characterize FO membrane are  $A$  and  $B$ , representing water permeability and solute permeability, respectively. Previously, the intrinsic parameters were used in the mathematical model to predict the FO membrane performance (water flux) [13] using a commercial cellulose acetate (CA) membrane provided by Hydration Technologies, Inc. (Albany, OR) [14].

However, the work was limited to the FO process parameters such as membrane orientation, feed and draw concentrations and membrane structural properties only. Instead of investigating the effect of FO process parameters on the performance, the current work was focused on the membrane itself. Using a mathematical model approach, this research aims to evaluate the intrinsic parameters ( $A$ ,  $B$ , and  $S$ ) and predict the performance of the thin film composite CTA-PA FO membranes fabricated with different CTA content in the support layer.

## 2. Methodology

### 2.1 Intrinsic Parameters Characterization

The intrinsic parameters of the membrane support and active layers were evaluated using mathematical modelling. Usually, the parameters  $A$  and  $B$  depend on the active layer characteristics, while parameter  $S$  is responsible for the support layer. The ideal FO membrane should possess high  $A$ , low  $B$ , and smaller  $S$ . The evaluation of the intrinsic parameters  $A$ ,  $B$ , and  $S$  is accomplished by the following modelling Eqs. (2) and (3) through a non-linear least square analysis approach using data obtained from our previous work [15] as tabulated in Tables 1 and 2. The intrinsic values were calculated using an Excel spreadsheet provided by Tiraferri *et al.*, [16], simplified in a visual basic (VB) platform.

$$J_w = A \left[ \frac{\pi_{D,b} \exp\left(-\frac{J_w S}{D}\right) - \pi_{F,b} \exp\left(\frac{J_w}{k}\right)}{1 + \frac{B}{J_w} \left[ \exp\left(\frac{J_w}{k}\right) - \exp\left(-\frac{J_w S}{D}\right) \right]} \right] \quad (2)$$

$$J_s = B \left[ \frac{C_{D,b} \exp\left(-\frac{J_w S}{D}\right) - C_{F,b} \exp\left(\frac{J_w}{k}\right)}{1 + \frac{B}{J_w} \left[ \exp\left(\frac{J_w}{k}\right) - \exp\left(-\frac{J_w S}{D}\right) \right]} \right] \quad (3)$$

The S value represents the changes in membrane support layer structural properties (porosity, tortuosity and thickness, as represented in Eq. (1). In this work, the support layer is assumed to have a pore structure with cylindrical shape ( $\tau=1$ ) and well-defined porosity and thickness. The membrane must be fully wetted before the FO test to make sure the "incomplete" wetting does not overestimate the S value due to inaccurate water flux during osmotic process.

**Table 1**  
FO membrane preparation [15]

Membrane code	CTA content	Interfacial Polymerization (IP) condition for PA active layer
M1	8wt%	2wt% m-phenylenediamine (MPD), 0.15wt%TMC
M2	10 wt%	(Trimesoyl Chloride) and 2 minutes reaction time.
M3	12wt%	

**Table 2**  
Data for  $J_w$  and  $J_s$  for the fabricated CTA-PA membrane [15]

Membrane code	Draw solution (DS), concentration (M)	Feed solution (FS)	$J_w$ (LMH)*	$J_s$ (gMH)*
M1	0.5	Pure water	7.3155	1.1995
	0.75		9.119	1.4457
	1.0		11.1786	2.2982
	1.25		12.1643	2.726
M2	0.5	Pure water	4.8762	1.0311
	0.75		3.7976	1.3587
	1.0		7.7857	1.808
	1.25		8.6655	2.2213
M3	0.5	Pure water	2.2976	0.1029
	0.75		3.3333	0.1715
	1.0		4.1429	0.2389
	1.25		4.8333	0.3087

**Note:** \*LMH= L/m<sup>2</sup>.h; gMH= g/m<sup>2</sup>.h

## 2.2 Prediction of CTA-PA Membrane Performance

The water flux data presented in Table 2 were recorded for selected DS (0.5, 0.75, 1.0 and 1.25 M) only hence Eq. (2) now can be used to predict the water flux ( $J_w$ ) at any DS concentration with different feed solution (FS) quality. Using intrinsic parameters (A, B and S) values obtained from the previous section,  $J_w$  was predicted by applying Eq. (2). To achieve this, Eq. (2) must be rearranged as follows:

$$J_w - A \left[ \frac{\pi_{D,i} \exp\left(\frac{J_w S}{D}\right) - \pi_{F,i} \exp\left(\frac{J_w}{k}\right)}{1 + \frac{B}{J_w} \left[ \exp\left(\frac{J_w}{k}\right) - \exp\left(\frac{J_w S}{D}\right) \right]} \right] = 0 \quad (4)$$

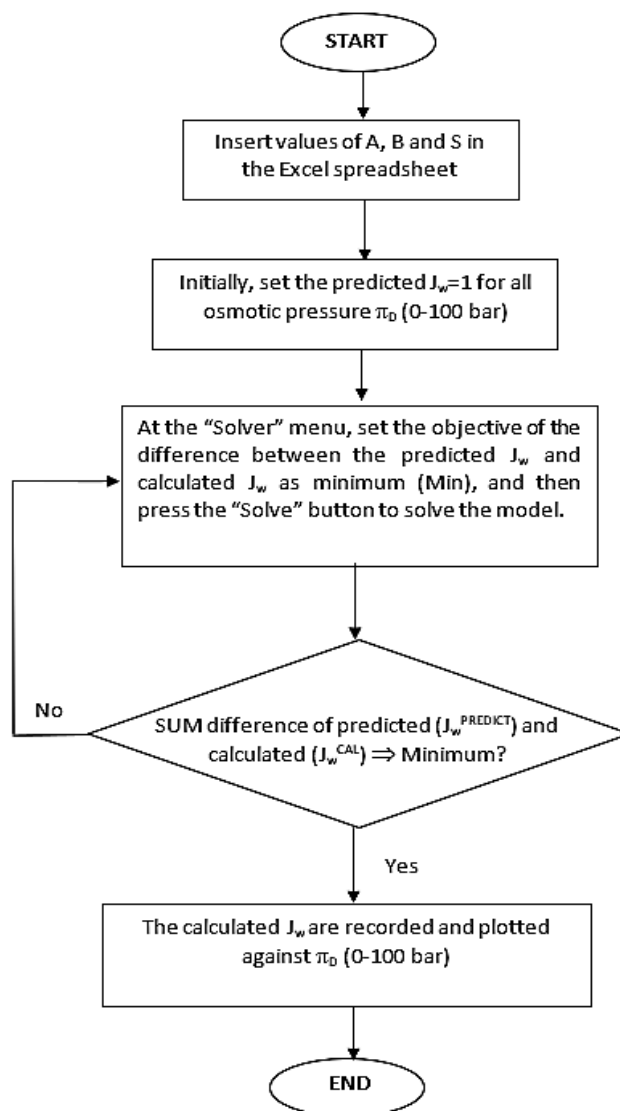
The "goal seek" approach of the numerical method could be applied to solve this complex equation. The Excel spreadsheet has a goal-seeking tool built-in where the equation can be solved by setting "0" or "minimum" for the difference between the 1<sup>st</sup> term and 2<sup>nd</sup> term of Eq. (4). However, the "goal seek" method in Excel spreadsheet has limited to 1 solution at one time. It is impractical when the other parameters (i.e., DS and FS concentration) are adjusted. As the current work would predict the  $J_w$  for a range of DS and FS concentrations, the model in Eq. (4) was solved using an Excel spreadsheet with the other function, namely Solver.  $J_w$  at different DS concentrations was obtained by minimizing the sum of differences between the predicted ( $J_w^{\text{PREDICT}}$ ) and calculated ( $J_w^{\text{CAL}}$ ).

The flow process of the numerical method for “goal seek” approach has been simplified in Figure 1. For the details Excel spreadsheet used in this current work, please refer to the attached supplementary file (S1). On top of that, the fabricated CTA-FO membrane also could be applied for power generation through Pressure Retarded Osmosis (PRO) process. Therefore, by applying the values of A, B, and S for each membrane, the power density  $W$  ( $W/m^2$ ) could be predicted using Eqs. (5) and (6).

$$J_{w,PRO} = A \left[ \frac{\pi_{D,b} \exp\left(-\frac{J_{w,PRO}}{k}\right) - \pi_{F,b} \exp\left(\frac{J_{w,PRO}}{D}\right)}{1 + \frac{B}{J_{w,PRO}} \left[ \exp\left(\frac{J_{w,PRO}}{D}\right) - \exp\left(-\frac{J_{w,PRO}}{k}\right) \right]} - \Delta P \right] \quad (5)$$

$$\text{Power density, } W = J_{w,PRO} \times \Delta P \quad (6)$$

Similar to the water flux prediction for FO, the water flux for PRO mode ( $J_{w,PRO}$ ) was calculated and solved using the Solver function for a set of applied pressure,  $\Delta P$ . The same procedure illustrated in Figure 1 was applied in PRO mode to obtain  $J_w$ . Finally, the obtained  $J_w$  is inserted in Eq. (6) to calculate the power density,  $W$ . The parameters for predicting water flux and power density are summarized in Table 3.



**Fig. 1.** Flowchart of numerical approach for “goal seek” using Excel spreadsheet

**Table 3**

Parameters for FO and PRO mathematical modelling

Parameter	
Bulk draw osmotic pressure, $\pi_D$ (bar)	0-100, 60 (brine solution) 0 (pure water)
Bulk feed osmotic pressure, $\pi_F$ (bar)	29.73 (seawater)
Solute diffusivity for NaCl, $D$ (m <sup>2</sup> /s)	$1.48 \times 10^{-9}$
Volume flow rate $\dot{V}$ (mL/min)	167 and 1000
Mass transfer Coefficient, $k$ (m/s)	$2.35 \times 10^{-6}$ (167 mL/min) $9.00 \times 10^{-6}$ (1000 mL/min)

### 3. Results and Discussion

#### 3.1 Intrinsic Parameters

The intrinsic parameters for the CTA-PA membrane were evaluated using a computational mathematic approach using a Microsoft Excel spreadsheet and presented in Table 4. The amount of CTA polymer content in the membrane formulation affects the intrinsic parameters. The water permeability  $A$  was reduced by almost 50% when the CTA content increased from 8wt% to 10wt%. A significant drop of the  $A$  value was observed at the highest CTA content of 12wt%. This reduction trend was also observed for the other two parameters,  $B$  and  $S$ . This indicates that the support layer could be tailored to the required properties by changing the amount of polymer during the membrane formulation.

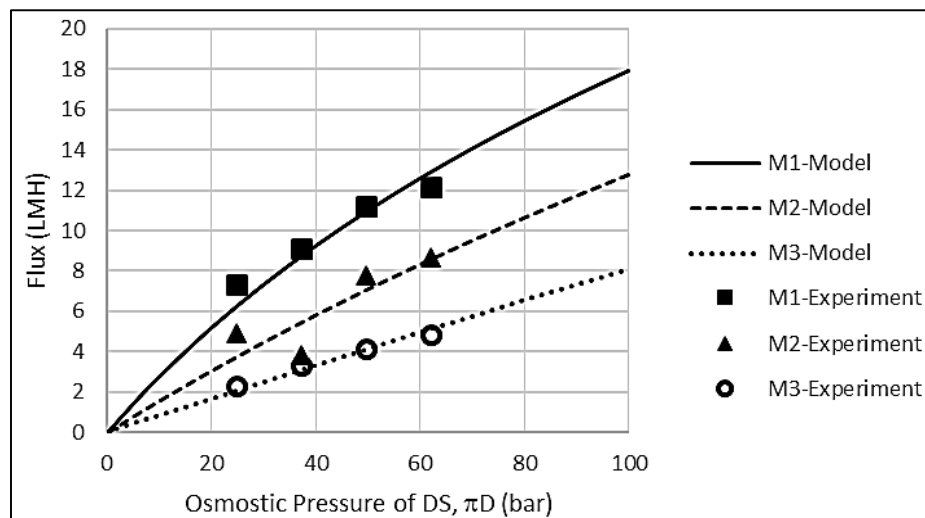
**Table 4**

Intrinsic parameters for CTA-PA FO membrane

Membrane code	$A$ (LMH.bar)	$B$ (LMH)	$S$ ( $\mu\text{m}$ )
M1	0.305	0.0514	152
M2	0.163	0.0355	98
M3	0.085	0.0042	32

#### 3.2 Water Flux Prediction for a Range of DS

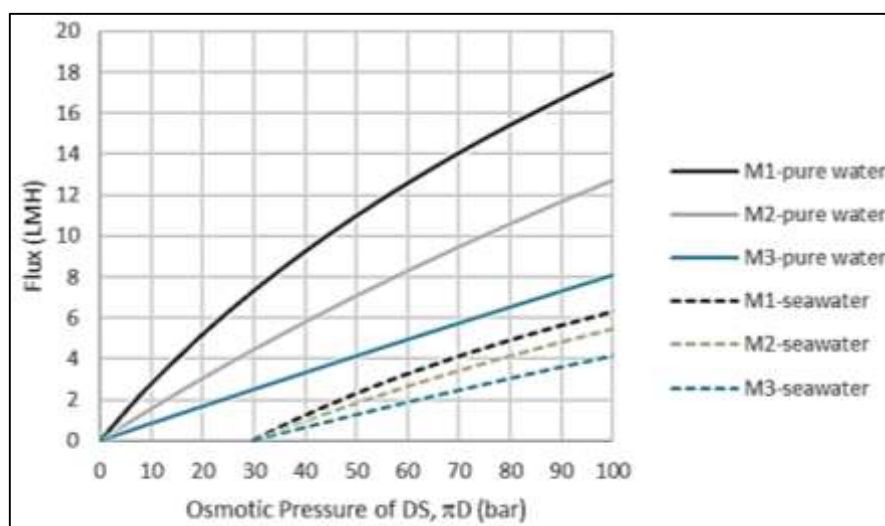
The data of water fluxes in Table 2 were obtained experimentally using four different DS concentrations of 0.5, 0.75, 1.0, and 1.25M representing osmotic pressure ( $\pi$ ) of 24.78, 37.17, 49.55, and 61.94 bar, respectively. This section predicted the water permeate fluxes from low DS concentration (0 M equivalent to  $\pi = 0$  bar) to high concentration, 2M ( $\pi = 100$  bar). Figure 2 shows the water flux for the M1 membrane is the highest while the M3 membrane exhibited the lowest flux compared to others as expected, which is indicated by the lowest  $A$  value reported in Table 4. The predicted fluxes almost fit the experimental data, showing the success of the mathematical model applied in this research work. This would help the user to predict the membrane performance (i.e., water flux) at any DS concentration when selecting an appropriate membrane for a specific purpose.



**Fig. 2.** Prediction of water flux for M1, M2 and M3 membranes at different DS concentrations using pure water as feed solution (FS)

### 3.3 Water Flux Prediction for Seawater as Feed

The FO membrane would also be useful for the desalination process. Therefore, the prediction of water flux to treat seawater is essential. In this part, the FS concentration was set as an average value of 0.6M ( $\pi_F=29.73$  bar) to represent the common salt concentration of seawater [17]. The predicted water flux of the membrane for seawater is presented in Figure 3. The water flux of the seawater was lower than pure water for all three membranes. This is because the net osmotic pressure generated for feed seawater is lower than that generated by feed pure water. From Figure 3, it was observed that the permeate water flux could only be produced when the  $\pi_D$  is greater than 29.73 bar, where this higher osmotic pressure starts to drive the water molecules moving from the feed side (FS-seawater) to the DS side and the water flux become higher at higher  $\pi_D$ . It is important to note that there is no permeate flux at  $\pi_D=29.73$  bar since the net osmotic pressure is 0 ( $\Delta\pi_{net}=\pi_D-\pi_F=0$ ).

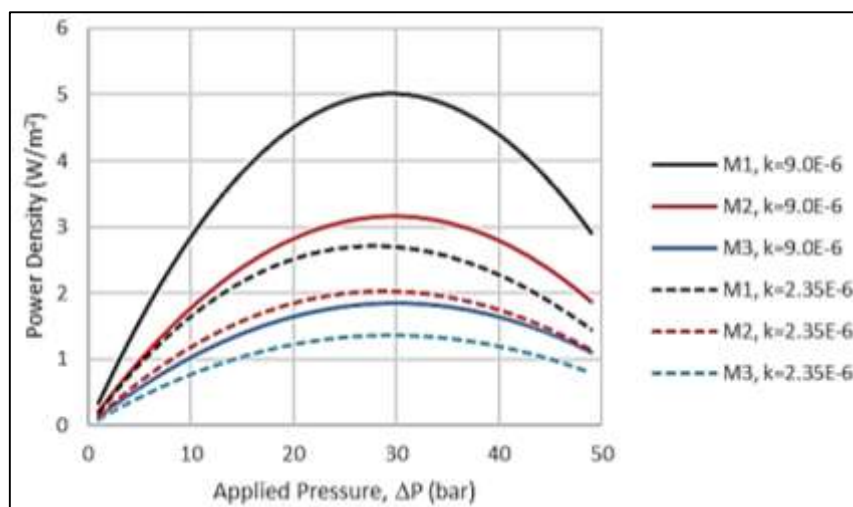


**Fig. 3.** Prediction of water flux for M1, M2 and M3 membranes at different DS concentrations using pure water and seawater as feed solution (FS)

### 3.4 Prediction of Power Density

The suitability of the CTA-FO membrane for PRO application was investigated using a mathematical model. The concentration of the draw solution was set as 1.20M ( $\pi_D=60$  bar), representing brine solution, while pure water was used as feed ( $\pi_F=0$ ). It can be seen in Figure 4 that the power density generated by the M1 membrane is the highest among the fabricated membranes, as expected from the higher flux produced by membrane M1 reported in the previous section. Referring to Eq. (6), the power density is affected by the water flux produced. The higher water flux produced would generate higher power density at a specified pressure. For example, at 30 bar of applied pressure, a maximum power density ( $W_{max}$ ) of 5 W/m<sup>2</sup> could be achieved by the M1 membrane, while M2 could generate roughly 3.16 W/m<sup>2</sup> only followed by M3 with the lowest value of 1.84 W/m<sup>2</sup>.

Besides that, it was found that the mass transfer coefficient significantly influences the power generated. This phenomenon was obvious for the M1 membrane and moderate for the M2 membrane with increments up to 86.6% and 57.2%, respectively. The power density increased with increasing the  $k$  value, indicating the higher volume flow rate (higher flow velocity) would reduce external concentration polarization (ECP) [18-23] and enhance the net effective osmotic pressure leading to higher water flux. However, the M3 membrane shows that the volume flow rate has a less significant effect on the water flux due to the dense membrane which is indicated by the extremely low water permeability ( $A$ ).



**Fig. 4.** Prediction of power density for M1, M2 and M3 membranes at different applied pressure ( $\Delta P$ ). Conditions:  $\pi_D=60$  bar,  $\pi_F=0$ ,  $k=2.35 \times 10^{-6}$  m/s &  $k=9.0 \times 10^{-6}$  m/s)

### 4. Conclusions

The mathematical model has been successfully employed to determine intrinsic parameters ( $A$ ,  $B$ , and  $S$ ) for the CTA-PA FO membrane. The results revealed the model fitted almost all of the experiment data, and it is beneficial for predicting the water flux at different conditions (i.e., different DS and FS concentrations). Using the obtained  $A$ ,  $B$ , and  $S$  values, the mathematical model for PRO mode was applied to predict the CTA-PA membrane suitability for power generation. The membrane formulation and operating conditions significantly affect the water flux and power density. Further



investigation is required to optimize membrane formulation to obtain higher water flux with maximum power density ( $W_{\max}$ ) for PRO application.

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