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Water flux prediction of UV-photografted nanofiltration membrane for forward osmosis application

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Abstract. In the application of forward osmosis, commercial nanofiltration (NF) membrane was modified via ultraviolet(UV)-photografting technique at different grafting parameters; monomer concentration (acrylic acid) and grafting time. However, the performance of the modified membrane measured at the osmotic pressure of 1M NaCl as draw solution limits their applications. In this research, the mathematical modelling was applied to predict the water flux at different osmotic pressure. The mathematical modelling indicated that the generated water flux follows the osmotic pressure but strictly affected by the grafting parameters. The result shows that at grafting time of 3min, the monomer concentration of 15 g/L generates the highest water flux followed by the membrane modified at the monomer concentration of 30 g/L. High water flux was attributed to the presence of strongly hydrophilic groups at new carboxyl layers leading to improved membrane properties. The approach of theoretical modelling on the membrane that is reported in this work allows estimating the water flux at different grafting parameter.

1. Introduction

Forward osmosis (FO) has become an emerging technology for water treatment. FO technology has gained recognition as a suitable complement to the reverse osmosis (RO) technology in a niche application. This is particularly notable in applications where the use of reverse osmosis alone is unfeasible or impossible [1]. According to McCutcheon et al. [2], FO technology has developed as a possible alternative technology for desalination due to its lower energy requirement as compared to both thermal and reverse osmosis desalination processes. Besides low energy consumption, the membrane fouling was reduced as it does not use any external pressure. In this process, a semi-permeable membrane is used for the separation of water but rejects solute molecules or ions by using the osmotic pressure as its driving force.

Among surface modifications, ultraviolet (UV)-photografting has attracted various researchers as it provides advantages in terms of simplicity, usefulness, versatility and low cost [3]. As such, this grafting technology has been widely used in various industries. In the coating industry, the UV-light provides low energy consumption, low emission, low capital investment, low space consumption and marginal substrate heating [4]. In addition, the fabrication via UV photografting produces an integral selective layer due to a strong chemical bond to the substrate. This helps to provide sufficient mechanical stability under relatively high operating pressure and also helps to guide against delamination or leaching of the grafted chains [5].

Recently, our research group discovered that surface modification via UV-photografting enhanced the FO membrane properties. The membrane that was modified at different grafting time (1,3 and 5min) and monomer concentration (5,15,30 and 50g/L) shows significant increases in the water flux [6]. It works by the addition of the carboxylic group (-COOH) on both of the active layer and support layer of the NF membrane. However, the generated water flux was measured at fixed osmotic pressure limits their applications. The experimental work was conducted using deionized (DI) water and 1M of sodium chloride (NaCl) as feed solution (FS) and draw solution (DS), respectively. Thus, the objective of this research is to study the generated water flux at a different osmotic pressure (e.g. variation of NaCl concentration) by using the mathematical model.

2. Mathematical modelling

In the FO technology, both external concentration polarization (ECP) and internal concentration polarization (ICP), as well as the membrane permeability, have to be considered and their models have to be developed separately before combining them to allow for accurate flux prediction in an FO process [7]. McCutcheon and Elimelech [8] have incorporated the dilutive ICP and concentrative ECP in mathematical models using the experimental data and the impacts of both dilutive ICP and concentrative ECP can be investigated concomitantly. The FO configuration was accurately described for water flux (J_v) shown in equation (1). This equation could work as a standalone predictor of flux under a variety of experimental conditions. This combine models can be used to predict the generate water flux and reverse solute flux under a different range of DS concentration and osmotic pressure. In this study, the water flux data was generated by using Excel Microsoft software.

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

Where $\pi_{D,b}$ is the bulk osmotic pressure of the draw solution at the concentration, $C_{D,b}$, $\pi_{F,b}$ is the bulk osmotic pressure of the feed solution at the concentration $C_{F,b}$ and P is the hydraulic pressure applied, which is considered zero in this case. Both terms $\left(\frac{J_v S}{D}\right)$ and $\left(\frac{J_v}{k}\right)$ depict the ICP and ECP respectively, D is the diffusion coefficient value and k is the mass transfer coefficient [9]. The negative and positive of the exponents indicate the concentrative and dilutive at that particular time respectively. The values of mass transfer coefficient (k), water permeability (A) and solute permeability (B) for this membrane were taken from our previous study [6].

Table 1. Parameters for forward osmosis mathematical modelling

Parameter	Symbol	Units	Value/Range
Solute diffusivity	D	m^2s^{-1}	$1.68x10^{-9}$
Bulk draw osmotic pressure	π_D	bar	0-70
Bulk feed osmotic pressure	π_F	bar	0 (DI water)
Water permeability	A	$L.m^{-2}hr^{-1}.bar^{-1}$	6.84-10.67 [6]
Solute permeability	B	$L.m^{-2}hr^{-1}$	15-117 [6]

The value of K can also be calculated as shown in equation (2);

$$K=\frac{S}{D} \tag{2}$$

where S is the structural parameter of the support layer and D is the diffusion coefficient of the solute (NaCl). The solute resistivity coefficient K is related to the structural parameter of the support layer, and the draw solute diffusion affects ICP in the support layer, indicating that is must be a significant parameter in the FO model [10].

3. Results and discussions

3.1 Unmodified NF membrane

In this research, the NF membrane with the commercial name of NF2 was purchased from Amfor Inc. Using equation 1 and data tabulated in Table 1, the model can predict the water flux of the NF2 membrane for FO process up to 60 bars of osmotic pressure. Results of the predicted water flux for the unmodified NF2 membrane are plotted in Figure 1. Using equation (1), the maximum osmotic pressure (60 bars) generate the water flux of 0.68 L.m^{-2}.hr^{-1}. It shows a non-linear behavior as the increases in the osmotic pressure which is attributed to the dilutive ICP [11]. It is named dilutive ICP because the draw solution is normally diluted by the permeate water within the porous layer. The prediction of flux was performed exactly according to what has been described in previous FO modelling study [2].

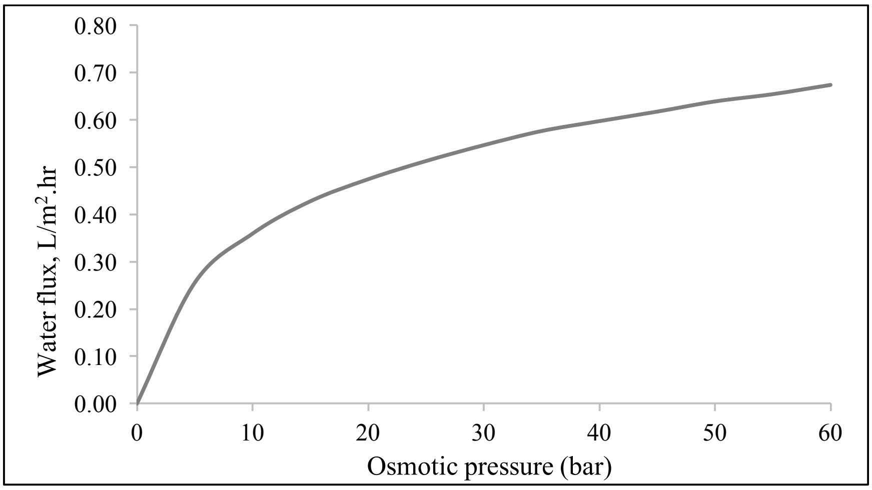

Figure 1 Prediction of water flux for unmodified NFPES membrane at different osmotic pressure

3.2 Modified NF membrane at different monomer concentrations

In general, modified membrane exhibit enhanced water flux property. The predicted water flux at different osmotic pressure generates more than two-fold compared to the unmodified membrane. Figure 2 (a)-(d) shows the effect of osmotic pressure on the permeation flux of water for membrane modified at different monomer concentration. The water flux increases with increasing osmotic pressure were recorded in the modified membrane. It can be explained that higher osmotic pressure facilitates the rapid penetration of water molecules across the FO membrane [12]. Moreover, the increases of water flux was attributed to the addition of carboxyl (-COOH) group on the membrane surface, resulting in higher hydrophilicity [13].

In Figure 2a, the membrane was modified using the monomer concentration of 5 g/L. Maximum water flux recorded at 1.8 L.m^{-2}.hr^{-1} (1min) while the lowest is at 1.6 L.m^{-2}.hr^{-1} (3min). At this monomer concentration, the gap range of maximum water flux at different grafting time was small. Research conducted by Abu Seman et al. [14] reported that at lower monomer concentration, the monomer can penetrate deep into the pores. Thus, the effective grafting on the membrane occurs at the various site due to the mobility of the monomer within the membrane area.

However, for membrane modified at a monomer concentration of 15g/L (Figure 2b) and 30g/L (Figure 2c), the grafting time of 1min and 3min shows higher values of water flux compared to 5min. From the trends, the increases of the grafted layer on the membrane surface lead to the reduction of water flux at higher grafting time. Effective grafting mechanism on membrane surface results in the thickening of the grafted layer thus increases flux resistance [15]. Interestingly, the gap range of water flux was big on both monomer concentrations. This is correlated with the effect of thickness from the graph polymerization reduces the water flux. On the other hand, using the highest monomer concentration (50g/L), the model has predicted the lowest water flux compared to the lower monomer concentrations. The maximum water flux was only at 1.33 $L.m^{-2}.hr^{-1}$ (3min) even though the gap range between the water flux is small. This result is contradicting with the lowest monomer concentration (5g/L). It can be said that due to the high concentration of the monomer, it cannot penetrate the membrane surface. Hence, the graph polymerization was limited to the active layer of the membrane only.

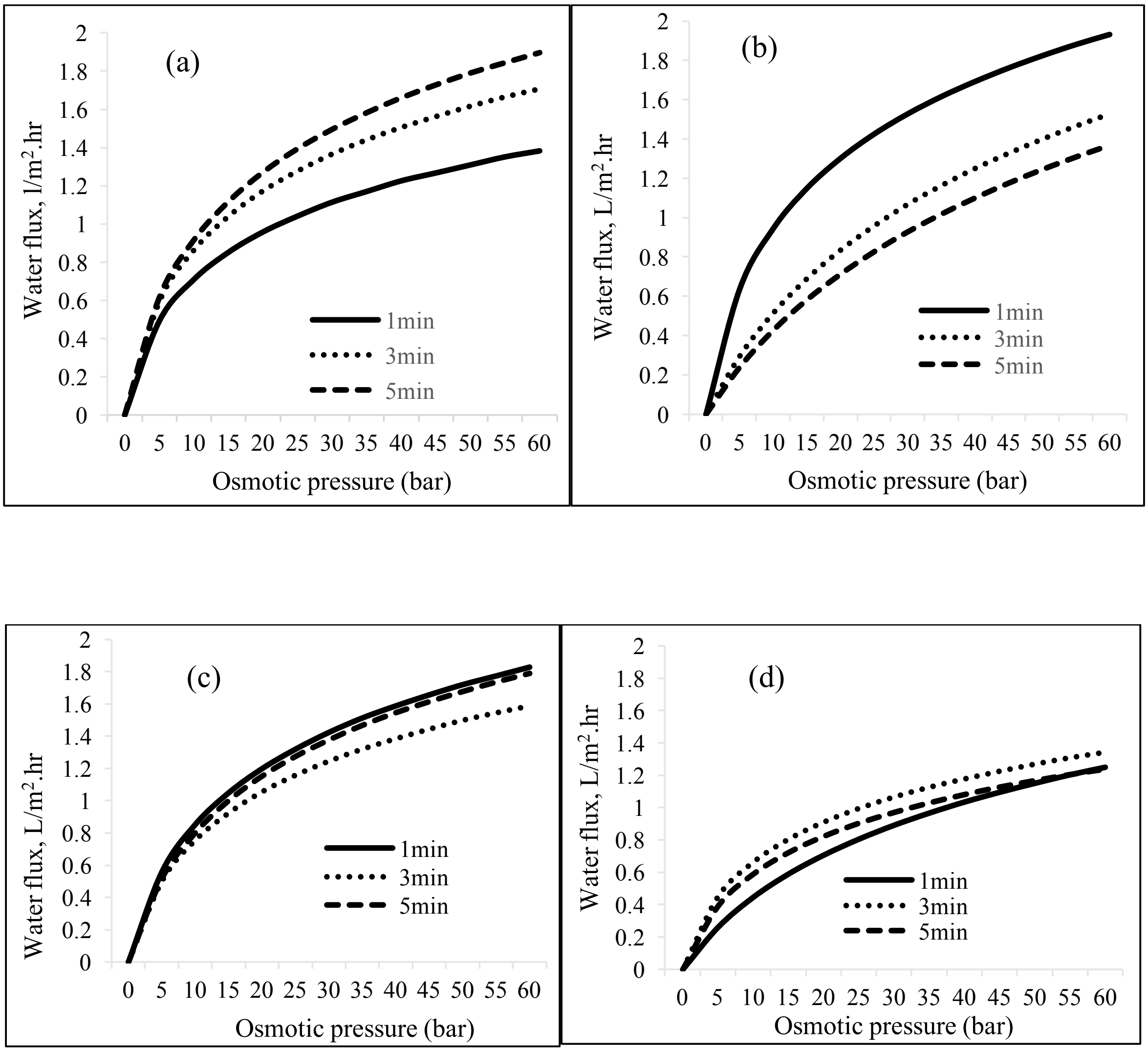

Figure 2. Prediction of water fluxes at monomer concentrations of (a) 5g/L, (b) 15g/L, (c) 30 g/L and (d) 50 g/L.

3.3 Modified NF membrane at different grafting time

Figure 3a-c shows the predicted water flux for modified NF membrane using the osmotic pressure up to 60bar. At different grafting time, the model predicts the highest water flux using the monomer concentrations of 15g/L and 30g/L. It shows that these two monomer concentrations will have a higher impact on the water flux at different osmotic pressure. Interestingly, the lowest (5g/L) and highest (50g/L) monomer concentration have recorded the lowest water flux. It can be said that in the surface modification of the NF membrane via UV-photografting technique, there is a specific range of monomer concentration to increase the water flux. Similar trend was recorded by Taniguchi et al. [16] in their research paper. They have identified that although all the grafted and polymerized monomers increased the membrane properties, there is a specific condition to obtain optimal performance.

On a different angle, there was big gap range for water flux in Figure 3a (1min) and Figure 3c (5min). At grafting time of 1min and 5min, both monomer concentrations of 15g/L and 30g/L recorded a high value compared to the monomer concentration of 5g/L and 50g/L. This finding concludes that the grafting time of 3min will produce a small range gap in the water flux.

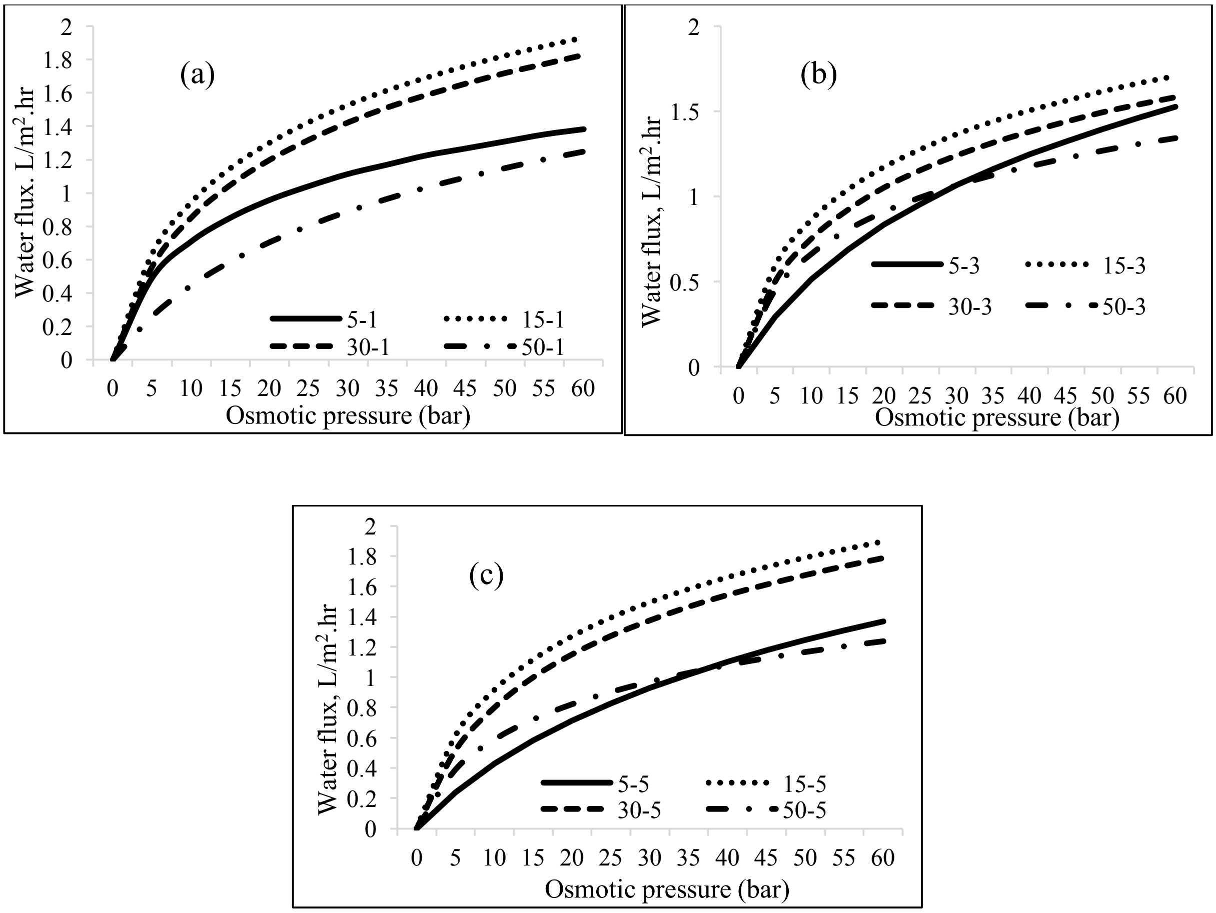

Figure 3. Prediction of water fluxes at grafting times of (a) 1 min, (b) 3 min and (c) 5 min

3.4 Solute resistivity for diffusion

The solute resistivity for diffusion (K) shown in equation (2) is defined as the structural parameter (S) of the support layer over the solute diffusion (D) in the support layer. Thus, as the increases of osmotic pressure (π), the lower the value of K, the higher water flux will be generated. The solute resistivity for diffusion discussed on the unmodified membrane and modified membrane at grafting time of 1 minute. From Figure 4, the unmodified membrane ($K=1.61x10^7$) generates the lowest water flux while sample 15-1min generates the highest water flux ($K=7.21x10^6$). A similar trend was reported by McCutcheon and Elimelech [17] in the flux modelled in the FO mode (dilutive ICP in the absence of concentrative ECP) for membrane with variable solute resistivity.

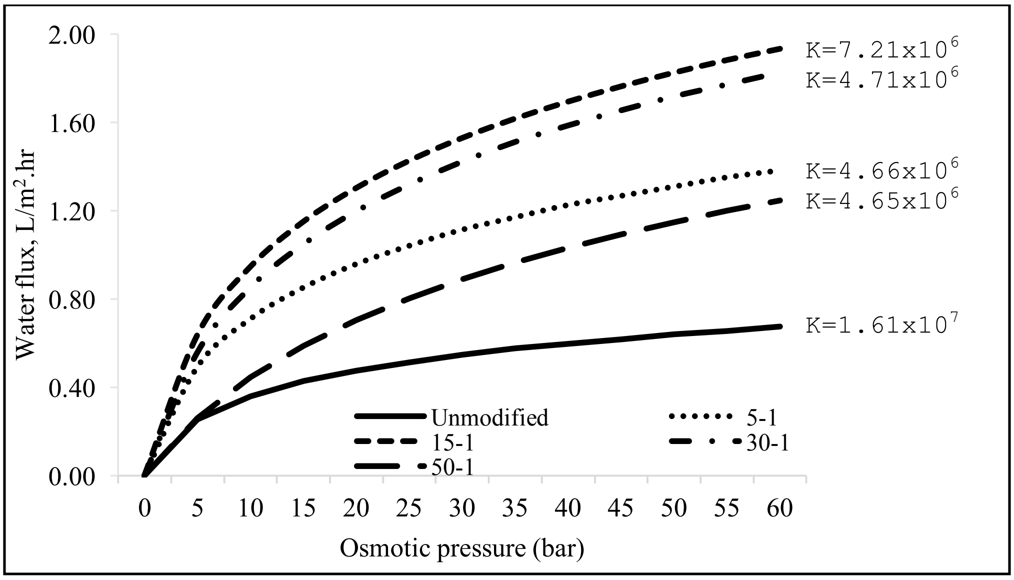

Figure 4. Solute resistivity of the modified membrane at 1min

4. Conclusion

Theoretical modelling was conducted to predict the water flux of the FO membrane modified at different grafting time and monomer concentrations. The model predicts the water flux using the osmotic pressure up to 60 bars. Herein, we have reported that the grafting parameter namely monomer concentration and grafting time plays a significant role in the modified membrane. In addition, the smaller value of the solute resistivity resulting in a higher water flux. Thus, the theoretical modelling is an effective way to predict the performance of the modified membrane.

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