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FFD Analysis for the Effect of Fouling on the Permeate Flux in High-Pressure Membranes

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***Abstract*—Porous high-pressure membranes have been widely used for both brackish water and seawater desalination. However, fouling (concentration polarization) extensively reduces permeate flux in high-pressure membranes such that reverse osmosis (RO) and/or nanofiltration (NF). In this study, we have attempted to understand the effect of membrane fouling on the permeate water flux by modeling the salt concentration profile within a membrane of interest. A parabolic (or diffusion) partial differential equation was used to describe the change in salt concentration inside the membrane. Subsequently, the PDE equation was solved numerically, under certain assumptions, by using forward finite difference (FFD) explicit method. It was found that salt accumulation occurs at the membrane feed-side surface and there was a noticeable decrease in water flux as fouling increased. For waters with an initial salt concentration of 10000 ppm (NaCl) and with an average diffusivity of $\mathbf{1.3 \times 10^{-6}\ cm^2/s}$, results showed that both RO/NF would have flux rates of 74.9, 67.4, 22.5, 0, –37.4, –74.9 LMH for the feed-side surface concentrations 0, 1000, 7000, 10000, 15000 and 20000 ppm, respectively, where negative flux indicates a back-flow scenario.**

***Index Terms*—Membrane Separation; Desalination; FFD Fouling Simulation; Reverse Osmosis.**

I. INTRODUCTION

Life qualities of human beings are totally dependent upon the possibility of meeting vast communities demands on drinking water which is prioritized as a top necessity for people [1], [2]. Water desalination through reverse osmosis (RO) and nanofiltration (NF) membranes remains the most common separation process in comparison to other conventional desalination technologies such as multi-effect distillation (MED), multi-stage flash (MSF) and electrodialysis (ED), for the production of fresh water [3]–[5]. However, RO and NF membranes require a high-pressure operation for seawater desalination where fouling issues will inevitably arise on membrane surface due to impurities accumulation and micro-particles/organics precipitation on the membrane permeate-side [4]–[6]. Drioli et al. (2002) [7] suggested that having a pretreatment system (e.g. microfiltration and/or ultrafiltration) prior to RO and NF modules would certainly mitigate fouling issues and extend membrane lifetime for several years. According to Rajamohan et al. (2014) [8], RO membranes are very susceptible to organic compounds; hence, biofouling problems arising from the removal of total organic carbon (TOC) and trihalomethane (THM) from contaminated water are more damaging to RO membranes than the conventional fouling issues from metallic and/or salt ions.

Typically, Membranes separate contaminants from water by passing it through tubular polymer films that will capture impurities on the retentate-side [6], [9]. Fouling takes place when dissolved and particulate matters in the feed water get deposited on the membrane surface-side resulting in increasing membrane resistance to the feed flow [12]. Theoretically, membrane fouling can be categorized into four different types based on the feedwater constitutes as the following: (1) scale (inorganic); (2) particulate; (3) biological and (4) organic compounds [10].

Fouling consequences on membrane systems, particularly on RO and NF modules, are as the following: (1) flux decline and salt passage due to the developed concertation polarization; (2) film degradation and damage from the increased differential/feed pressure; (3) energy loss and poor operation owing to high-pressure and frequent cleaning requirements; (4) treatment cost increase; and (5) permeate quality reduction [11]–[13]. Accordingly, addressing fouling issues in RO/NF membranes through modeling and numerical analysis might be a fruitful way of understanding fouling behavior for better protection.

Here, we investigate the effect of NaCl salts on fouling of RO and/or NF membranes due to the treatment of salt water with different concentrations. Feed waters with various initial concentrations (NaCl) of 10000 ppm (mimicking desalination of groundwater or brackish water) with a chosen fouling concertation range of 0–20000 ppm were studied. Numerical analysis was carried using forward finite difference (FFD) explicit method along with other assumptions to calculate the decline in the flux rates and correlate the observed flux declines to the selected fouling concentrations.

II. ASSUMPTIONS

The effect of membrane fouling was studied in high-pressure membranes which were RO and/or NF with assumptions from previous works [14]–[16]: (1) NaCl rejections of RO and NF are 95% and 50%, respectively; (2) initial salt (NaCl) concentration is 10000 ppm; (3) water has an average membrane diffusivity of $1.3 \times 10^{-6}\ cm^2/s$; and (4) membrane feed-side surface (fouling) concentrations are in the range 0–20000 ppm. Involved study parameters and their selected values which were used in the FFD numerical analysis are shown in Table I.

RO membrane configuration (spiral-wound module) is shown in Fig. 1. Raw water enters the membrane through the module shell-side by applying a high pressure that is enough to push water spirally along the feed spacers

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enveloping a long-rolled thin film composite (TFC) film which is capable of retaining undesired salts and ions. Freshwater is collected from the permeate output (tube-side) owing to the spiral-wound TFC film that would only allow fresh water to pass through diffusion while excluding other contaminants (concentrate output on the shell-side).

TABLE I: FFD MODEL PARAMETERS AND THEIR SELECTED VALUES

Parameter (Symbol)	Value	Unit
Rejection (R); for RO and NF*	95 and 50, respectively	%
Initial salt concentration (C_0)*	10000	ppm
Fouling concentration (C_f)*	0 ~ 20000	ppm
Water diffusivity (D)	1.3×10^{-6}	cm^2 s^{-1}
Membrane thickness ($z = L$)	250	μm
Treatment/passing time (t)	25	s

*for brackish water with sodium chloride salts (NaCl) only.

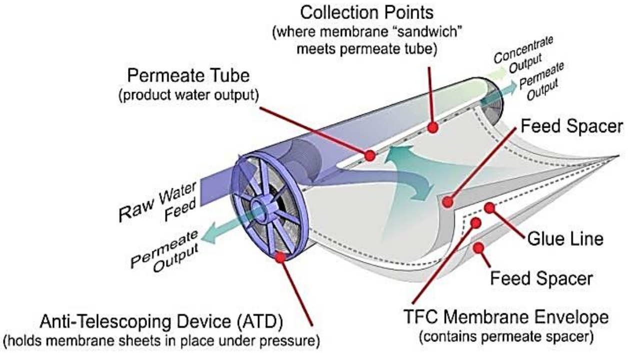


Fig. 1. Spiral-wound RO membrane configuration; Adapted from [17].

III. METHOD AND EQUATIONS

For the FFD numerical analysis, the parabolic (or diffusion) partial differential equation (PDE) from (1) has been used along with the application of the FFD method (explicit) to get (2); where notations referring to space and time are as the following: $\{(i, k) = (z, t)\}$. FFD is a powerful mathematical method used to convert PDEs, such that (1), to equations in algebraic form to be solved numerically. By using the converted and analytically determined equation, as shown in (2), we have been capable of estimating the salt concentration profile within the membrane (equations were solved through a programmed algorithm code in MATLAB) [18]–[20].

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} \tag{1}$$

$$C_i^{k+1} = C_i^k + \frac{D\,\Delta t}{(\Delta z)^2}\left(C_{i+1}^k - 2C_i^k + C_{i-1}^k\right) \tag{2}$$

For further analysis, total water flux across the RO and/or NF membrane was determined from Fick's first law [19] as shown in (3) and at the selected fouling concentrations which were previously reported in Table I. Initial, fouling and total flux rates were calculated from both (4) and (5).

$$J = -D\frac{dC}{dz} = -D\left[\frac{C - C_0}{z - z_0}\right] \tag{3}$$

$$J_0 = -D\left[\frac{C_P - C_0}{L}\right] \;;\; J_f = -D\left[\frac{C_f - C_0}{L}\right] \tag{4}$$

$$J_T = J_0 - \left(J_0 - J_f\right) \tag{5}$$

where J is the flux rate (g cm^{-2} s^{-1}); subscripts: 0, P, f and T refer to initial, permeate, fouling and total respectively, D is water diffusivity into the membrane (cm^2 s^{-1}), C is salt concentration in (g cm^{-3}) and z refers to the spatial position (cm) with respect to membrane thickness, L.

Based on RO and/or NF membrane structure described in previous works [16], [21], we have only considered the polyamide (PA) layer for the flux calculations, and that fouling layer was assumed to only occur in the PA layer. The PA layer was selected instead of the other layers (PSF and PEST from Fig. 2 and Table II) because PA has a very low pore size as compared to the other layers making our selection a valid approximated assumption. The very low pore size in PA makes it responsible for the high membrane rejection and flow resistance in both RO and/or NF membranes. Hence, membrane resistance to water flow from PSF and PEST layers can be neglected due to their larger pores which allow easy water flow with minimal backpressure (despite that PSF and PEST are much thicker than PA, their resistances are still negligible due the larger pores which are three orders of magnitude higher than PA pores).

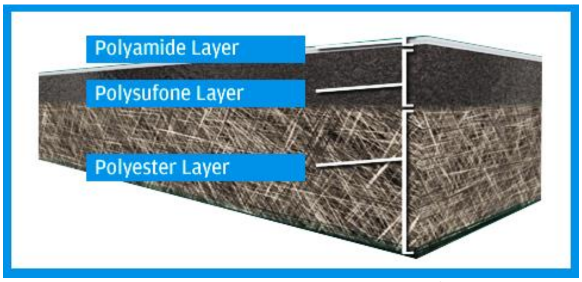


Fig. 2. Membrane layer structure; Adapted from [21]

TABLE II: RO/NF MEMBRANE LAYERS AND THEIR THICKNESS

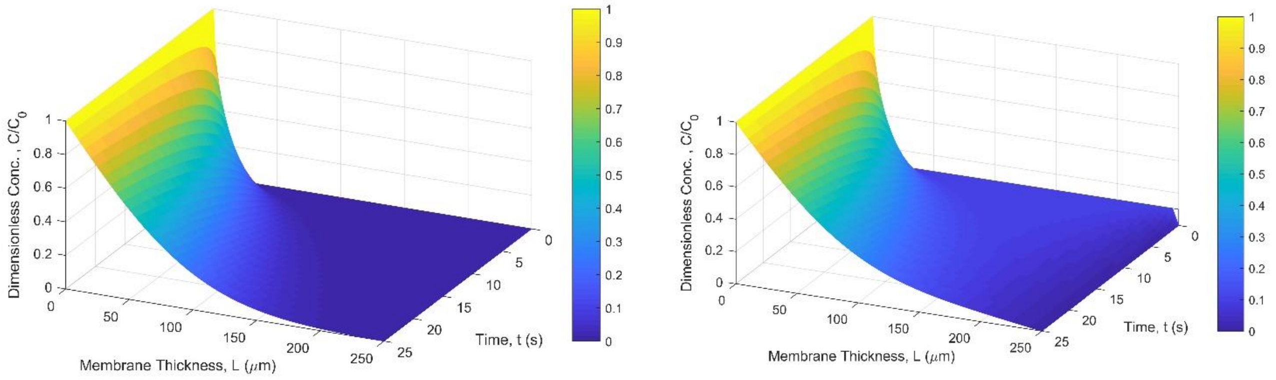
Membrane Layer	Selected	Literature
Polyamide (PA)*	62.5 nm	1 – 200 nm
Polysulfone (PSF)	40 μm	30 – 50 μm
Polyester (PEST)	150 μm	100 – 200 μm

*Flux rates were calculated based on the PA layer only; since the resistance of other layers were neglected due to their larger pores (much larger pore diameters as compared to PA).

IV. RESULTS AND DISCUSSIONS

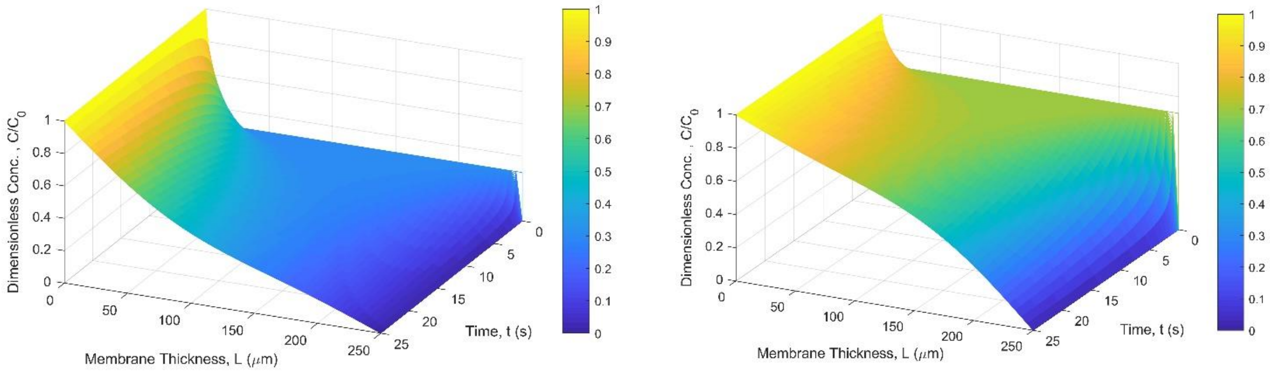
Dimensionless concentration profiles of NaCl salts within the treated water inside the membrane are shown in Fig. 3 with respect to both membrane treatment time (time) and membrane thickness (space) at various selected fouling concentrations. Salt accumulations (NaCl) on the membrane surface were observed to occur much frequently at high fouling concentrations; there should be proportionality between feed salt concentration and membrane fouling rates. High fouling rates resulted in lowering membrane treatment efficiency across the membrane thickness due to the high amounts of NaCl salts accumulated onto the surface which extensively reduced the ideal treatment condition of ~0 ppm salts in the produced water (indicated as blue regions in Fig. 3). As expected, the best instantaneous treatment efficiency was determined when $C_f = 0$; and there was a noticeable decrease in the instantaneous efficiency with the build-up fouling ($C_f > 0\ ppm$) with the worst treatment performance when fouling was furtherly progressed ($C_f \to 2C_0$). In other words, the permeation may get highly contaminated at high fouling rates owing to extensive salt accumulations on the membrane surface and within membrane film (thickness). Typically, membrane cleaning may only help in removal of salts accumulated on the surfaces (as $C_f < 1000\ ppm$); but as fouling progress to higher levels (as $C_f > 7000\ ppm$), membrane cleaning would become more difficult due to arising of inner-membrane fouling besides the surface fouling which might make it more difficult to flush out trapped salt ions to achieve better instantaneous membrane performance.

Impact of membrane fouling and concentration polarization (surface and inner) on the permeate flux rates (feed water across the membrane) is shown in Fig. 4. Since we have selected a fixed initial salt concentration, forward water fluxes (J_0) at initial treatment times were found to be constant across the membrane regardless of C_f value. However, backward fluxes at membrane surface were found to be linearly decreasing with fouling rates (high C_f yield in shifting backward flux values from positive to negative; indicating that fouled membranes progress high surface resistance to water flow which might reverse water direction in severe cases). Thus, reduced water flux defined as ($J_0 - J_f$) have been expected to behave with an almost opposite behavior to J_f in which reduced water flux positively and linearly increases with the increase in fouling concentrations. Total flux rates observed across the membrane have been clearly identified to be linearly decreasing (from 74.9 to 0 LMH) with the increase in the assigned fouling concentrations ($C_f \to C_0$) owing to the high amounts of accumulated slats explained via reduced flux rates; when $C_f > C_0$, membrane would be in the worst case scenario where most of the pores will be clogged from concertation polarization, hence, total water flux rates could be reversed across the membrane where negative flux indicates a back-flow scenario.



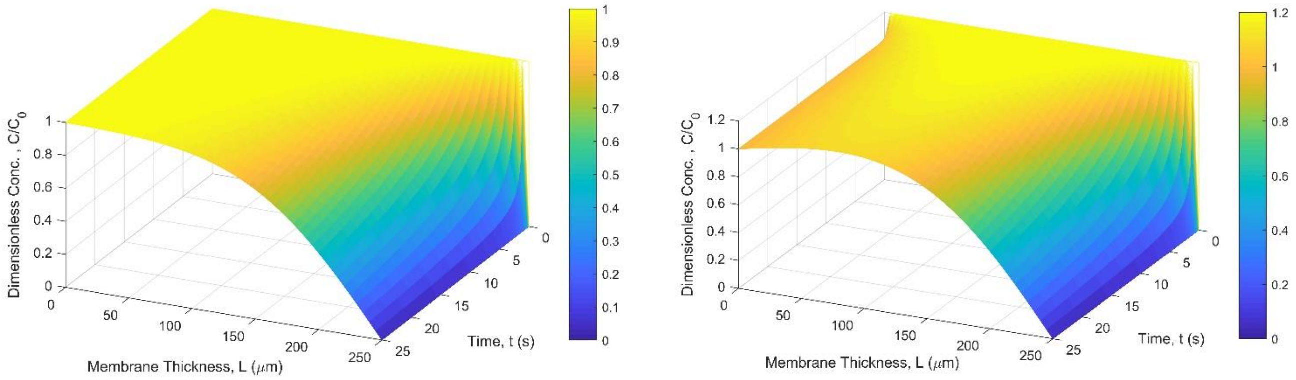
(a) Fouling concentration (C_f) = 0 ppm

(b) Fouling concentration (C_f) = 1000 ppm



(c) Fouling concentration (C_f) = 3000 ppm

(d) Fouling concentration (C_f) = 7000 ppm



(e) Fouling concentration (C_f) = 10000 ppm

(f) Fouling concentration (C_f) = 12000 ppm

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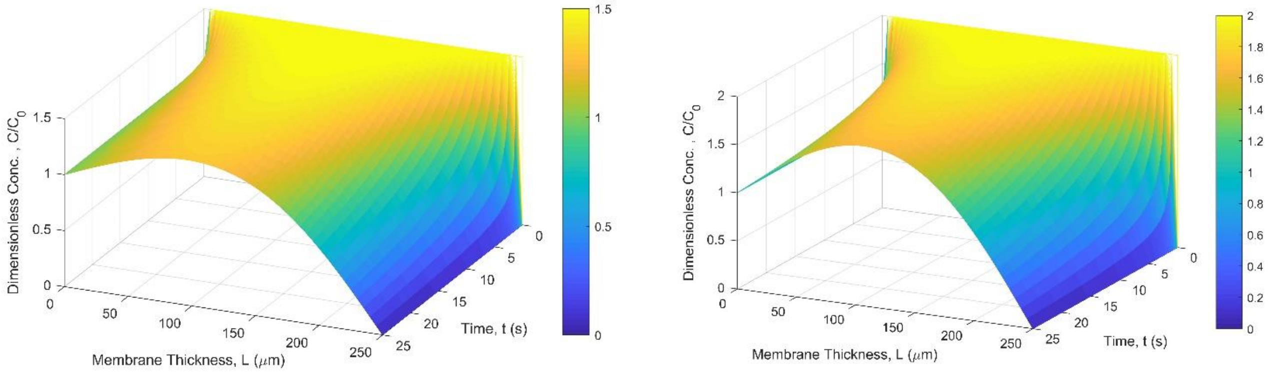
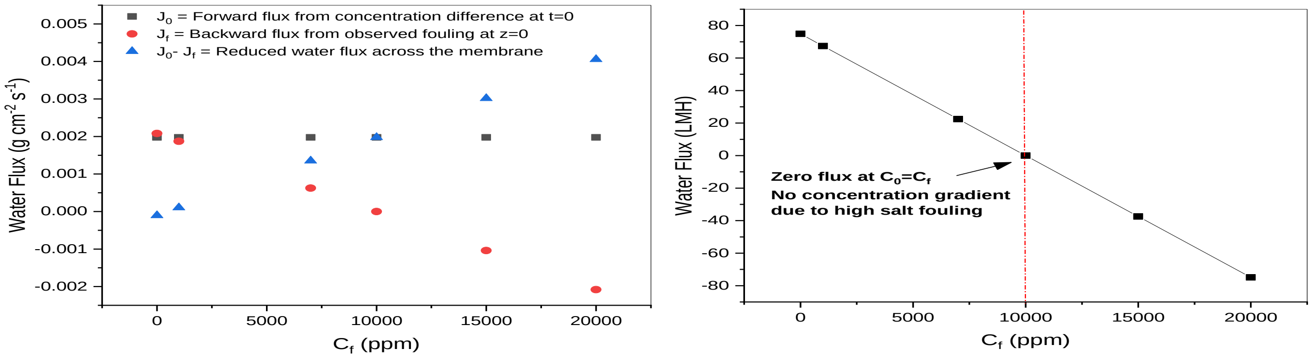


(g) Fouling concentration (C_f) = 15000 ppm (h) Fouling concentration (C_f) = 20000 ppm

Fig. 3. Salt concentration profile as a function of membrane treatment time and thickness at an initial concentration (C_0) of 10000 ppm and different fouling concentrations (C_f) as mentioned below each figure.



(a) Forward, backward and reduced fluxes (b) Total flux: $J_T = J_0 - (J_0 - J_f)$

Fig. 2. Observed water flux across the membrane at an initial concentration (C_0) of 10000 ppm and at different fouling concentrations (C_f).

V. CONCLUSION

Fouling (concentration polarization) associated with brackish water and seawater desalination in high-pressure membranes (RO/NF) was studied numerically in order to determine fouling impact on salt concentration profile and permeate flux rates of treated water. PDE equations were solved analytically through the FFD method and then coded in MATLAB for carrying out the results. Salt accumulations at the membrane feed-side surface were found to increase with fouling concentration which would decrease membrane treatment efficiency. Further, high fouling rates lowered flux rates from salt accumulations ($J_T = 0$ at $C_f = C_0$; and backflow at $C_f > C_0$). Our numerical analysis concluded that fouling has severe impacts on RO/NF membranes where high salt accumulations decrease treatment efficiency and lower permeate flux rates.

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