Effect of Several Parameters on Membrane Fouling by Using Mathematical Models of Reverse Osmosis Membrane System

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Abstract

In this paper, models were applied to investigate the parameters that affect membrane fouling. Osmotic pressure across the membrane, salt concentration at the surface of the membrane, solute mass transfer coefficient, effective coefficient diffusion of water, and concentration polarization factor were the main parameters that calculated in this simulation. Sodium chloride was assumed the only salt existed in the feed flux. In addition, changing the applied pressure versus increasing the salt concentration in the feed flux and their effect on the water permeation coefficient was investigated. The results confirmed that concentration polarization gives a good indication about the formation of the fouling layer at the membrane surface and consequently permeate decline.

Keywords: Reverse osmosis; Concentration polarization; Salt permeation; Mathematical model.

1. Introduction

Reverse osmosis (RO) membrane processes are among the most important water treatment technologies. This technology is widely used around the world particularly in the regains where the surface waters are scarcely available such as the islands and the peninsulas. RO can help meet drinking water demands through desalination of seawater and brackish water. In addition, RO membranes are used for many purposes such as water recycling and resource recovery [1]. The main idea of the RO process is to separate the product water (permeate) from the salts in the source water by using high pressure driven through the membrane to overcome the natural osmotic pressure. This process leads to transporting the permeate (desalinated water) through the membrane leaving the minerals in the feed side of the membrane [2]. The major problem that faces the efficient operation of RO system is fouling which can cause a deterioration of the quality and quantity of the desalinated water and result in high cost of water treatment [3]. Fouling of the membrane is the accumulation of the materials on its surface or in its pores. These materials cannot be removed by using backwashing or backflushing processes [4]. Fouling may happen due to microbial growth (biofilms), particulate materials, dissolved organic matters, inorganic materials such as calcium sulfate and calcium carbonate, suspended particles such as aluminum and iron silicates, or colloidal particles such as clays and flocs [5-7]. RO membrane maybe classified as a non-porous membrane; therefore, the major fouling mechanism that can be considered in RO models is the surface fouling of the membrane [8, 9].

Many models and simulation methods for the performance of the RO processes have been developed and proposed. These models, which in general have the same basic equations, consider several elements in the RO membrane system such as the applied pressures, cross-flow velocity, solute-diffusion, concentration polarization, flux, rejection, recovery, fouling, and silt density index (SDI) [10,11]. Hessami et al [12], for example, developed a mathematical model for the RO process based on an assumption of the solutiondiffusion equation and validate it by using a computer software produced by RO membrane manufacturer. Their results revealed that the performance of the membrane was about the same for the output of the model and the computer software. Furthermore, Jamal et al [11] proposed a simulation model to reduce fouling by controlling the concentration polarization of the membrane. They reported that the concentration of the feed and the product, rejection, and flux predicted by the model was almost consistent with experimental data.

In this study, Microsoft Excel was used to develop a simulation of a single stage seawater reverse osmosis unit with spiral-wound modules. The main focus of this analysis addressed the phenomenon of concentration which impacts all RO membrane fouling processes. The effect of using different values of applied pressure in the system was investigated. In addition, increasing the concentration of the salt, sodium chloride in this simulation, and its effect on membrane fouling and consequently on the water permeation was addressed. The assumptions stated in the simulation based on mathematical models developed by Avlonitis et al [13], Geraldes et al [10] and Hoek et al [1].

2. Modeling of RO System

Avlonitis et al [13], Geraldes et al [10] and Hoek et al [1] developed and proposed

mathematical models for the RO systems based on the assumption that the spiral wound modules have flat feed channels because the radius of the module is much bigger than the thickness of the feed channels. The width, length, and height of the channel are W, L, and h, respectively as shown in Figure (1). In addition, the pressure loss in the permeate channels was negligible [10].

Osmotic pressure occurs due to the difference in the solute concentration between the feed and the permeate channels. In the RO process, a pressure larger than the osmotic pressure to first stop and then reverse the flow of water is applied. The difference between the applied pressure and the osmotic pressure is the net driving force [14, 15].

Net Driving Force =
$$(\Delta P - \Delta \pi^{\circ})$$
 (1)

Where ΔP = difference in the applied pressure; and $\Delta \pi^{o}$ = difference in osmotic pressure.

According the assumptions to incompressible and ideal solution behavior, the equation of the osmotic pressure, π° in dilute solutions can be derived by using the concentration and temperature of the solution. In

addition, for the high concentrated solution, osmotic pressure coefficient, \emptyset_c , which is related to the nature and the concentration of the substance (solute) is considered in Eq. (2) [16,

$$\pi^{\circ} = \emptyset_c \frac{{}^n}{V_m} RT \tag{2}$$

Where, \emptyset_c = osmotic pressure coefficient (for seawater, it ranges from 0.85 to 0.95) [17]; n = number of moles of solute; V_m = molar volume of water; R= universal gas constant (0.083145 bar/mole.K); and T= absolute temperature (T°C+273).

Using Eq. (3) which was developed by Geraldes et al [10], water flux rate or solvent permeate flux, F_w^o , can be calculated as follows:

$$F_w^0 = W_n [P - \pi_m + \pi_n] \tag{3}$$

 $F_w^o = W_p [P - \pi_m + \pi_p]$ (3) Where W_p = water permeability coefficient, considers temperature and the characteristics of the membrane and the solute; P = applied pressure; π_m = osmotic pressure in the feed channel; and π_p = osmotic pressure in the permeate channel.

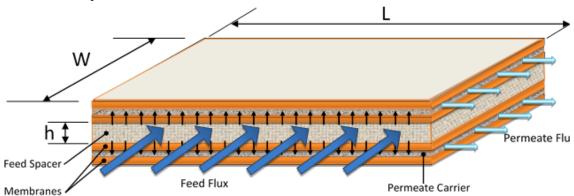


Figure 1: Diagram of the flat channels of spiral wound module.

However, Eq. (4) [16] was used to find the solvent permeate flux through the membrane, F_w^o in this study.

$$F_{w}^{o} = \frac{D_{l,w}^{o}C_{w}V_{m}}{RT\delta_{m}}(\Delta P - \Delta \pi^{\circ})$$
 (4)
Where $D_{l,w}^{o}$ = effective diffusion coefficient

of water through the membrane; C_w = water concentration in the membrane which is provided by the manufacturer; and δ_m = membrane thickness. Eq. (4) indicates that the solvent permeate flux, F_w^o and the thickness of the membrane, δ_m are clearly correlated. In other words, solvent permeate flux decreases by increasing membrane thickness [16].

From Eqs. (3) and (4), water permeability coefficient, W_p , can be found according to the following equation: $W_p = \frac{D^o_{l,w} C_W V_m}{RT \delta_m}$

$$W_p = \frac{D_{l,w}^o C_w V_m}{RT \delta_m} \tag{5}$$

Eq. (6) estimates solute flux through the membrane, F_i^o , which is occurred due to the high solute concentration difference across the membrane [1, 10, 13, 18].

$$F_i^o = K_n (C_m - C_n) \tag{6}$$

 $F_i^0 = K_p(C_m - C_p)$ (6) Where C_m = solute concentration in the feed side; C_p = solute concentration in the permeate side; K_p = solute permeability of the membrane

$$K_p = \frac{D_{l,i}^0 K_d}{\delta_m} \tag{7}$$

which can be calculated from Eq. (7) [18]. $K_{p} = \frac{D_{l,i}^{0}K_{d}}{\delta_{m}}$ In Eq. (7), $D_{l,i}^{0} = \text{effective diffusion}$ coefficient of solute through the membrane; and K_d = solute distribution coefficient which is usually constant for each membrane by the manufacturer. Thus, Eq. (6) can be written in terms of concentration of the solutions, C_i on either side of the membrane [16].

$$F_i^o = \frac{D_{l,i}^o K_d \Delta C_i}{\delta_m} = K_p \Delta C_i \tag{8}$$

In addition, solvent permeate flux and solute flux through the membrane can be found by the formula;

$$F_w^o = \frac{Q_p \rho_w}{A_m}$$
 or $F_i^o = \frac{Q_p c_p}{A_m}$ (9)
Where Q_p = volumetric flow rate of the

permeate; $A_m = cross$ section area of the membrane; and ρ_w = water density.

Solute rejection, R_i° , defined in Eq. (10), is the difference of the solute concentrations between the feed channel and permeate channel

$$R_i^{\circ} = \left[1 + \frac{\kappa_p c_{wp}}{W_p (\Delta P - \Delta \pi^{\circ})}\right]^{-1}$$
 (10)
Where C_{wp} = concentration of water in the

permeate.

The accumulation of the rejected solute or salt on the membrane surface in the feed side results in higher concentration of the solute than the one in the feed channel (solution bulk). This leads to the concentration polarization phenomena which is an indicator for the membrane fouling. Concentration polarization factor, CP, can be calculated from Eq. (11) [1, 16].

$$CP = \frac{c_{im}}{c_{ic,M}} = 1.333 exp \left[\frac{\binom{F_W^0}{\rho_l}}{0.75 V_F} \binom{2}{f} Sc^{2/3} \right]$$
 (11)

Where C_{im} = solute concentration at the membrane surface; $C_{ic,M}$ = solute concentration in the feed channel; V_F = average velocity in the feed channel; ρ_l = solution density in the feed channel; and f = Fanning friction factor which is equal to $(16/R_e)$ in the round tube and $(14.227/R_e)$ in the square channel. R_e = Reynolds number which can be found by Eq. (12). Sc = Schmidt number which can be calculated by Eq. (13) [10].

$$R_e = \frac{\rho_l V_F d_H}{\nu} \tag{12}$$

$$R_e = \frac{\rho_l V_F d_H}{\mu}$$

$$Sc = \frac{\mu}{\rho_l D_{l,i}^0}$$
(12)

In the last two equations, $\mu = dynamic$ viscosity of the solution in the feed channel and d_H = the feed spacer equivalent diameter.

3. Results and Discussions

The previous equations were used to simulate a model of spiral-wound reverse osmosis membrane system by using Microsoft Excel. The parameters used in this simulation are listed in Table (1). Some of the parameters in Table (1) were assumed according to the specifications of the membrane module which are determined by the manufacturers. For example, feed spacer feed spacer equivalent diameter, membrane length and membrane width were determined according to the properties of the Module FilmTec SW30HR-380^a [10]. One leaf of RO membrane was assumed in this simulation; therefore, both volumetric flow rate in the feed and the permeate channels were assumed to be low. Other parameters were determined according to the typical values (osmotic pressure coefficient, \emptyset_c , for example was assumed to be 0.92) [17] or by using the equations which were mentioned in section 2. Since RO membrane is better in rejecting divalent ions than monovalent ions and sodium chloride is a common measurement standard [12], the feed solution was assumed to have sodium chloride only. This assumption seems to be reasonable because the concentration of the sodium chloride in the sea water is very high compared to the other elements [2].

Table 1: Parameter values for model simulation.

Parameter	Value	Parameter	Value
Q _F (cm ³ /sec)	5.1222	F _w (g/cm ² .sec)	0.14330435
$C_i (mg/L)$	10000	A (cm ²)	0.138
Q _p (cm ³ /sec)	0.0192	δm (cm)	0.002
$C_p (mg/L)$	20	Δw (cm)	69
Φ_{c}	0.92	H (cm)	0.0737
n	2	W_{spacer} (cm)	88
$V_{m}(L)$	1	L_{spacer} (cm)	69
R (L.bar/mole.K)	0.083145	d _H (cm)	0.0935
T (°K)	293	V _F (cm/sec)	1.0073
MW (mg/mole)	58500	μ (mg/cm.sec)	1065000
$\rho_1 \left(g/cm^3 \right)$	1.03	υ (cm²/sec)	0.0117
ΔP (bar)	50		

In this simulation, first, the effect of increasing sodium chloride concentration (1000 -30000 mg/L) on the osmotic pressure across the membrane was investigated according to Eq. (2). The results revealed, as shown in Figure (2), that the difference in osmotic pressure increased from 0.766 to 22.987 bar. The total increase of the osmotic pressure was about 97%. Owing to the increase of osmotic pressure across the

membrane, the net driving force for permeate through the membrane is decreased. Thus, more driving pressure needs to be applied to balance the deficit.

In addition, as a result of increased solute, the salt concentration at the surface of the membrane, according to Eq. (6), increased from 0.98 to 29.98 mg/cm³ as illustrated in Figure (3) and hence increasing membrane fouling which can decline the product by reducing the available filtration area.

In terms of solute mass transfer coefficient, $D_{l,i}^{o}$, and effective coefficient diffusion of water through the membrane, $D_{l,w}^{o}$, both coefficients are increased by changing the concentration of the sodium chloride to high values. Figure (4) displays that range of the variation of solute mass transfer coefficient is larger than the range of the effective coefficient diffusion of water.

The results showed that the solute mass transfer coefficient increased from 3.63×10^{-4} to 6.62×10^{-4} m/sec, while the effective coefficient diffusion of water increased from 1.42×10^{-4} to 2.58×10^{-4} m/sec. The results indicated that both solvent and solute coefficients, which are mostly provided by the manufacturer, are not constant when the concentration of the feed water is dramatically increased. As previously mentioned, by increasing the osmotic pressure, more netting driving pressure needs to be applied which results in pushing more solute and solvent through the membrane. The increase of the solute inside the membrane may result in changing its properties or will lead to a deterioration of the permeate quality [19].

Concentration polarization has an important role in enhancing membrane osmotic pressure and surface fouling [20]. Concentration polarization factor (CP) was calculated by using Eq. (11) and, then, plotted versus the variation of salt permeation coefficient as illustrated in Figure (5). According to Figure (5), concentration polarization factor increased by increasing salt permeation coefficient (K_p) as a consequence of changing sodium chloride concentration. The

increase in salt permeation coefficient from 9.28 \times 10⁻⁵ to 5.59 \times 10⁻⁵ m/sec resulted in a dramatic increase in concentration polarization factor from 1.594 to 1.722 which accounts to about 88%. The rapid growth in the CP reflected the increase in the concentration of sodium chloride from 5000 30000 mg/L. This indicated that the concentration polarization at low solute concentrations is not significantly affected. In other words, concentration polarization may not be the only predominant role of membrane fouling in the RO system when low salt concentration water is used as a feed water such as brackish water. However, at high solute concentration, concentration polarization is still as a major character of membrane surface fouling. Finally, changing the applied pressure versus increasing the salt concentration in the feed flux and their effect on the water permeation coefficient was investigated in this simulation by using Eq. (5). Figure (6) displays that water permeation coefficient significantly varies with applied pressure from 30 to 50 bars in the high concentration of sodium chloride. The range of this coefficient is limited in the range of 0.004 to 0.01 when the applied pressure is more than 60 bars. The high water permeation coefficient (larger than 0.01) for the low applied pressure, which falls in the range of 30 to 50 bars, attributed to the increase of the product. At higher applied pressure (more than 50 bars), water permeation coefficient was kept lower than 0.01. High pressure enhances the built up of the salt on the membrane surface and consequently decreases salt rejection and permeate flux as reported by [19].

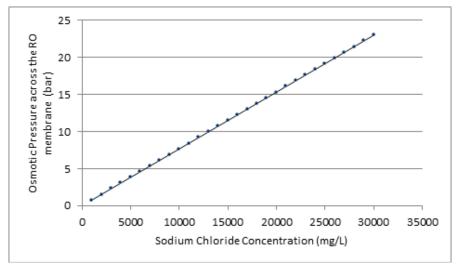


Figure 2: Osmotic pressure across the membrane versus sodium chloride concentration.

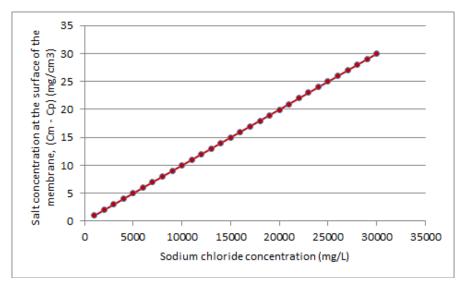


Figure 3: Salt concentration at the surface of the membrane versus sodium chloride concentration.

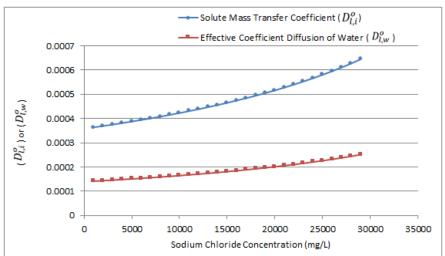


Figure 4: Variation of solute mass transfer coefficient and effective coefficient diffusion of water versus sodium chloride concentration.

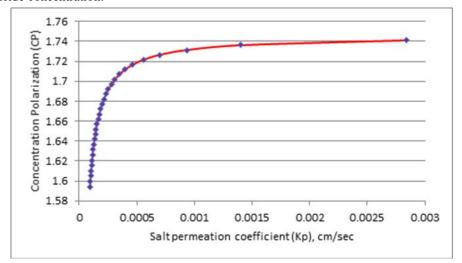


Figure 5: Variation of concentration polarization factor versus salt permeation coefficient.

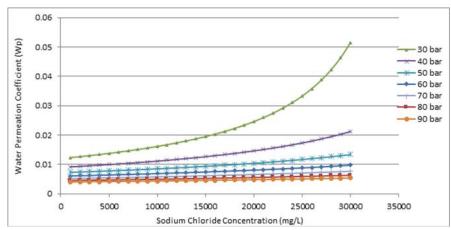


Figure 6: Variation of water permeation versus changing both of applied pressure and concentration of sodium chloride.

4. Conclusion

The aim of this study was to investigate the parameters that affect membrane fouling by using mathematical models. The results showed that the osmotic pressure across the membrane is significantly influenced by increasing the concentration of the solute in the feed flux. In addition, the results confirmed that concentration polarization provides a good indication about the formation of the fouling layer at the membrane surface and consequently permeate decline. Moreover, both solute and water diffusion coefficients through membrane were affected by changing the solute concentration in the feed channel. This indicates that these coefficients. which are mostly provided by the manufacturer, may be varied when the concentration of the feed water is dramatically increased. Finally, for high pressure, water permeation coefficient is slightly increased by increasing the solute concentration. However, at lower applied pressure, the change in the water permeation coefficient was obvious. It is recommended to experimentally study the effect of varying both the applied pressure and the solute concentration on the permeate flux and the salt rejection in the same ranges done in this study.

Nomenclature

0	Volumetric flow rate in the feed channel
Q_{F}	(cm^3/sec)

- Solute concentration in the feed channel C_i (mg/L)
- Volumetric flow rate in the product channel Q_p (cm³/sec)
- Solute concentration in the feed channel
- C_p $\Phi_{\rm c}$ Osmotic pressure coefficient (Dimensionless)
- Number of mole of the solute n
- $V_{\rm m}$ Molar volume of water (L)
- Universal gas constant (L.bar/mole.K) R
- Absolute temperature $(T \circ C + 273) = (^{\circ}K)$ Т
- Molecular weight of the solute (Sodium MW Chloride) (mg/mole)

- Density of liquid (seawater density, 35% $\rho_{l} \\$
- salinity) (g/cm³)
- Difference in the driving pressure cross the ΔΡ
 - membrane (bar)
- Solvent permeate flux through membrane
- $F_{\rm w}$ (g/cm².sec)
- A Section area of the membrane (cm²) Thickness of membrane (cm) δm
- δw Membrane width (cm)
- Feed channel height (cm)
- W_{spacer} Feed spacer width (cm)
- L_{spacer} Effective feed spacer length (cm) $d_{H} \\$ Feed spacer equivalent diameter (cm)
- Average velocity in the feed channel (cm/sec) V_F
- Dynamic viscosity of the solution in the feed
- μ channel (mg/cm.sec)
- Viscosity of the solution in the feed channel
 - (cm²/sec)

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تأثير بعض المتغيرات على ترسيب الأملاح على الأغشية باستخدام المعادلات الرياضية لنماذج تقنية التناضح العكسي باستخدام الاغشية

داود عيسى ساجت قسم هندسة البيئة الجامعة المستنصرية

الخلاصة

تم تطوير العديد من المعادلات الرياضية التي تمثل تقنية التناضح العكسي باستخدام الأغشية من قبل العديد من الباحثين. في هذه الدراسة ، تم استخدام بعض النماذج الرياضية لدراسة تأثير تغيير العناصر التي تؤثر على كفاءة عمل هذه الأغشية. من هذه العوامل الضغط التناضحي عبر الغشاء، معامل كفاءة عبور الماء خلال الغشاء ومعامل التناضحي عبر الغشاء، معامل كفاءة عبور الماء خلال الغشاء ومعامل استقطاب الأملاح على طرفي الغشاء. تم فرض كلوريد الصوديوم هو الملح الوحيد الموجود في الماء الداخل الى منظومة الأغشية كذلك تم دراسة تأثير تغيير الضغط المسلط على الاغشية مقابل زيادة تركيز الأملاح على إنتاجية الغشاء من الماء المار. النتائج أظهرت تأثير تركيز الاستقطاب عالى على انسداد الاغشية وبالتالي انخفاض الماء المار من خلال الغشاء .