

Numerical Study on a Formation of Concentration Polarization on a Reverse Osmotic Membrane Surface

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Abstract: A global water crisis has led to thriving industry of desalination seawater for past decades. Desalination process of seawater through reverse osmotic (RO) membrane separation is the most widely used in a water treatment process as its effectiveness in separating the dissolved solute from seawater solution and able to produce clean water. However, scaling problems, organic fouling, particulate matter, and biofouling are common types of foulant that can be occurred on the surface membranes layer during the separation process. Thus, a numerical study on the separation process using RO membrane was conducted where 2D spiral wound membrane model was developed by COMSOL[®] Multiphysics 5.5 software which the aim is to determine the factors that influence the formation of scaling on the RO membranes surface layer. In this study, concentration polarization of calcium carbonate on membrane surface was determined at three range of parameters; pressure ($P=15-35$ bar), temperature ($T=30-60^{\circ}\text{C}$) and concentration of ion calcium carbonate ($C_i=5000-15,000$ mg/L). It shows a significant effect on the formation of concentration polarization on the membrane surface where at $T=30, 45$ and 45°C , the permeate flux obtained are 21.9, 24.6 and 26.8 $\text{L}/\text{m}^2\text{hr}$, respectively. While, for the ion concentration factor, the obtained permeate flux for 5000, 10,000 dan 15,000 mg/L, are 22.3, 25.7 dan 27.4 $\text{L}/\text{m}^2\text{hr}$, respectively and for the pressure factor, the permeate flux obtained are 21, 28 and 34 $\text{L}/\text{m}^2\text{hr}$ at $P=15, 25$ and 35 bar, respectively. Through this study, it was found that this developed spiral wound membrane model can estimate the potential scaling of the ion calcium carbonate on the membrane surface layer up to 70% efficiency. Thus, this model can be applied in the desalination of seawater to identify the optimum operating parameters, aid in the dosage selection of anticipants, and estimate fouling mitigation effects in the pressure-driven membrane filtering process.

Keywords: *Reverse Osmosis, Membrane Fouling, Concentration Polarization, Scaling; Spiral Wound*

1. Introduction

Currently, more than 845 million people are living with severe water shortages, and an estimated about 2.8 billion people worldwide will be facing a serious water shortage by

2025, according to a United Nations report [1]. Due to water crisis, several efforts have been made to address this problem including water treatment, rainwater collection and desalination of seawater. It was found about 71% of the earth's surface consists of water, which 97.4% is seawater, while 2.6% is freshwater [2]. Thus, due to the versatility of

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these seawater uses, the desalination of seawater process has become a rising industry over the past few decades to ensure the sustainability of clean water resources. The development of desalination of seawater plants around the world goes hand in hand with the membrane technologies such as forward osmosis, reverse osmosis (RO), electrodialysis, membrane distillation, filtration etc. [3-6]. (2020). Among these said technologies, RO desalination of seawater has been used in one of the large-scale industrial membrane processes as it uses less energy as well it can be distinguished by the membrane polymer processing material, structure, and its configuration.

Based on membrane configuration, RO membranes can be divided into spiral membranes, hollow fibers, and flat sheets (plate and frameworks) [7]. Among these configurations, spiral wound membrane has a high compaction density and allows for easy cleaning. Furthermore, it also has a large per unit volume, strong design that prevents membrane cracking and has relatively low construction capital and minimal operating costs [8]. The spiral wound membrane has various configurations where it can be designed with different spacecraft, membrane types, lengths, and diameter sizes that allow it to be used in a variety application such as ultrafiltration, gas permeation and sulfate removal [8]. Due to that, RO membrane is more practically to use for desalination of groundwater as it can develop formation of calcium-soluble salt crystals; calcium sulfate and calcium carbonate in membrane cells [9].

Yet, one of the main concerns for the mostly membrane designers and operators is to reduce the formation of membrane impurities that occurred during salt precipitation process (scaling) as tend to be when the solubility of salts exceeds their solubility limits in a solution and will cause the precipitation of salts [10]. Mostly, membrane separation can be operated in crossflow and dead-end mode [2]. The crossflow filtering is dominant in the most RO and nano filtration processes as it has a potential of stable production rate with a continuous crossflow to control the polarization of membrane concentration and impurities (Wang & Wang 2018). In addition, these RO membrane processes are usually driven by a pressure to achieve optimal separation with its ability to move an element more easily as compared to the other separation processes. Through the RO process, a feed flow that containing solute, and solvent will permeate through the membrane layer, while the solute will be partially filtered.

However, concentration polarization (CP) tends to occur as the flow concentration of a particular component increases or decreases at the boundary layer near to the membrane surface due to the transportation of the selected components that pass through the membrane. Thus, increased a mass transfer plays an important role, so that the CP formation can be reduced where it can improve the

performance of the RO separation process. Also, the operating pressure in the RO process need to overcome the osmotic pressure due to difference in a concentration (chemical potential difference) that allows water to permeate from higher concentrated solution to the lower concentrated solution. In this case, salt will accumulate around the membrane layer and cause CP on the side of the membrane feed, and it known as an external concentration polarization. Meanwhile, the low permeates concentrations passing through the support layer will cause minimal and negligible polarization [11-12]

For more understanding on this phenomenon, numerous experiment, and modelling studies [13-20] have been reported on the CP formation and flux scaling due to deformation of ion carbonate on the membrane surface layer by various imagining technique such as dynamic model on crystallization theory [21], digital holographic interferometry [22] etc. It shows the salt formation (scaling) will cause desalination module to unable function properly [9]. Hence, preventive measures for the formation of this scaling problem need to be studied so that its effect can be reduced. Many past research based on experimental, theoretical, and empirical models are developed to study this CP formation phenomenon [23-28]. Even so, due to complexity nature of membrane features and with the advance in computing science, it can be solved and simulated using computational fluid dynamic (CFD) within time consuming. As CFD analysis not only saves a time and cost effective, nonetheless also is more environmentally friendly in most of cases. Even though the CFD prediction will not completely replace the experimental investigation for obtaining information for design purposes, but it is believed that CFD methods play a vital role in providing insight into the complexity of the flow phenomenon [29].

Therefore, in this study, 2D spiral wound membrane was developed and simulated with the aim is to determine the factors that influence the formation of scaling on the RO membrane surface layer. The CP formation on the membrane layer was observed and determined at various range of parameters such as at the inlet pressure; $P=15-35$ bar, temperature; $T=25-60^{\circ}\text{C}$ and inlet concentration of calcium carbonate ion; $C_i=5000-15,000$ mg/L and so its effects of the CP formation on the membrane surface were studied.

2. Methodology

2.1 Spiral wound membrane model design

A 2D model of a spiral wound membrane for RO separation was developed using AutoCAD[®] software as shown in Figure 1(a). The height and length of the spiral wound membrane channel is 1 mm and 6 mm, respectively which was chosen based on the common range used in the

most commercial spiral wound membrane RO modules. Then, this model was imported to the COMSOL[®] Multiphysics version 5.5 software to simulate and observe the formation of concentration polarization on the membrane surface layer at various range of parameters. Once the 2D spiral membrane model was developed, the physics equations and boundary conditions were defined and set up, followed by meshing generation, and then setting the models with different data sets and parameters to simulate to observe the flow profile on the membrane surface layer. For meshing generation, a free-triangular mesh and finer mesh size was employed and chosen as shown in Figure 1(b) since it was adequate to guarantee a low Reynolds number with less than 3 for all situations practically in the whole domain.

2.2 Physical properties and parameters

In this study, a constant viscosity of salt solution is $\nu=1.0 \mu\text{m}^2/\text{s}$, and a permeability of sodium chloride solution in the liquid solution is $D=1.5 \times 10^{-9} \text{ m}^2/\text{s}$. A 100% complete salt rejection was selected to obtain an accurate value for scaling formation that occurred due to polarization of calcium carbonate concentration during the separation process and the linear dependency of the osmotic pressure on salt concentration ($\Delta\pi=k_cw$, $k=75 \text{ kPa m}^3/\text{kg}$) are used in this simulation. If the salt rejection is not 100%, the calculation of ion flux across the membrane layer become more complicated. The physical properties of the materials used is shown in Table 1 and the permeability of a solution vary according to the concentration of dissolved substance [30-32].

2.3 Governing equation and physic model

In this simulation, Equations (1) - (4) were applied that show an expression for the solution used to solve diffusion model where m_A is mass fraction of salt, π is osmotic pressure of the solution (Pa), μ is viscosity of the solution (Pa.s), D_{AB} is diffusion coefficient of salt and ρ is fluid density [33]. Based on the solution-diffusion model, the driving force for volumetric flux is a pressure difference and the osmotic pressure of the ionic solution that can be calculated using Van't Hoff equation which varies on the ion concentration and absolute temperature.

$$\pi = 805.1 \times 10^5 m_A \quad (1)$$

$$\mu = 0.89 \times 10^{-3} (1 + 1.63 m_A) \quad (2)$$

$$D_{AB} = \times 10^{-9} (1 - 14 m_A), 1.45 \times 10^{-9} \quad (3)$$

$$\rho = 997.1 + 694 m_A \quad (4)$$

2.4 Define a boundary condition and model physics

By applied a solution-diffusion model, the boundary

conditions on the membrane surfaces are defined where the volumetric flow through the membrane is a function of inlet pressure, temperature, and ion concentration. Nernst-Planck equations was used for transporting the ion solute through the membrane [10] and the diffusion flux flow of this model is relatively smaller than the flow velocity in the membrane channel. The COMSOL[®] Flow Mode model was used to simulate and analysis on a hydrodynamic state of RO membrane separation. Considering sodium and chloride ions are the main ions in solution, the density and viscosity of the solution correspond to the NaCl solution with the same amount of concentration. The ionic velocity calculated by the flow model was used for a flow convection condition across a membrane layer.

During the RO membrane separation, the flow of solution in the channel is flowed to the high concentration solution. Then, the absorption of water through the membrane is driven by pressure differences in the feed flow and diffuse the flow through a membrane layer. A formation of concentration polarization of ion calcium carbonate and salt precipitation were occurred under specific chemical conditions. To observe a formation of a concentration polarization on the membrane surface, various range of the temperature ($T=25-60^\circ\text{C}$), pressure ($P=15-35\text{bar}$) and the concentration of ion calcium carbonate ($C_i=5000-15,000 \text{ mg/L}$) were studied. At the higher temperature, the concentration formation of calcium carbonate scaling is expected to be increased as a CO_2 gas is generally more soluble under the higher pressure. As the pressure drops, the carbon dioxide gas will oxidize from the solution, causing a decrement in the concentration of carbonic acid and the formation of calcium carbonate scaling tend to be occurred [18, 34-35].

3. Results and discussions

3.1 Concentration polarization at various parameters

Figure 2 shows a flow profile of the concentration polarization distribution that formed on a membrane surface layer at three range of the ion concentrations; $C_i=5000, 10,000$ and $15,000 \text{ mg/L}$ where the temperature was maintained at 25°C and the separation process was simulated within 30 seconds. The color bar indicates the range of averaged concentration polarization distribution of calcium carbonate scaling during the separation process where the red color shows the maximum value; 1 mol/m^3 , and the blue color shows the lowest value; 0. The results show the scaling distribution of concentration polarization formation increase with increasing of time and ion concentration where in the case of the dilute ions concentration; $C_i=5000 \text{ mg/L}$, the formation of scaling on the membrane surface is quite lower which is around 0.1 mol/dm^3 . However, as the ions concentration increases and become denser, the formation of scaling on the membrane

surface layer gradually increases and become dominant at $C_i=15,000\text{mg/L}$ that shown in the redder color distribution around the membrane surface layer.

From this flow profile result, the effect of ion concentration on the polarization deformation of CaCO_3 concentration on the membrane surface layer is plotted qualitatively as shown in a Figure 3. The CaCO_3 is formed when the solution is saturated with the calcium and bicarbonate ions. It shows the concentration polarization is gradually increases with increasing of ion concentration and time. It was occurred as the denser ion concentration; it will affect the RO process due to the rate of concentration polarization on the membrane surface layer which prevents the diffusion process from taking place effectively.

Besides, with the dense salt concentrations and higher saturation levels at the membrane surface layer, it will increase the nucleus formation at the RO membrane surface. Once the nucleation occurs, scaling will be formed and cover membrane surface as well as reduce the permeability of the membrane for the diffusion process to occur [31]. Therefore, the denser ion concentrations will result in the higher osmotic pressures, a higher tendency of suspended solids to be formed in the membrane surface layer, and a higher possibility of scaling will be occurred. Also, the concentration of calcium carbonate is highly dependent on the level of dissolved carbon dioxide gas in the liquid solution as it exists in the water ion in the form of bicarbonate ions. As supersaturating condition is occurred, bicarbonate ions begin to decompose, releasing CO_2 gas and formed carbonate ions which can result in precipitation of calcium carbonate in presence of calcium cations [36].

Since the formation of the concentration polarization phenomenon is more likely to occur at higher temperatures, a simulation at the various range of temperature with a constant pressure and inlet velocity was studied. Figure 4 shows the flow distribution of the concentration polarization that occurred on the membrane surface layer at three temperatures; $T=30, 45$ and 60°C in 30s where the ion concentration was fixed at $C_i=10,000\text{mg/L}$. The color bar indicates the range of concentration polarization distribution of the calcium carbonate scaling during the separation process where the red color shows the maximum value, and the blue color shows the lowest value. The results show the scaling distribution of the concentration polarization formation is increased with the higher temperature and separation time. Overall, it shows the effect of the temperature on the formation of scaling on the membrane surface is less significant at lower temperature, $T=30^\circ\text{C}$; $C_o < 0.5 \text{ mol/dm}^3$. However, as the temperature is increases, the formation and thickness of scaling on the membrane surface gradually increases especially at the center area of membrane layer which can be visualized in the mixture of yellow and green color at $T=60^\circ\text{C}$.

From this temperature flow profile result, the effect of the temperature on the concentration polarization of calcium

carbonate on the membrane surface are plotted qualitatively as shown in a Figure 5. It shows the highest concentration polarization formation was observed at $T=60^\circ\text{C}$, and lowest at $T=25^\circ\text{C}$. It has been verified that the concentration polarization occurs at a high rate with increasing of the temperature as the temperature affects a water viscosity, solubility, and mass transfer coefficient of solute. It is because at the higher temperature, the water's viscosity will decrease, and this condition will cause an increment in the diffusion flux. This condition will contribute to the formation of the significant scales on the membrane surface [37]. The temperature also affects a solubility of CaCO_3 where at the higher temperature, the solubility rate of CaCO_3 is lower which causing concentration polarization to easily be formed on the membrane surface. However, in the most cases, the solubility rate of the similar inorganic salt does not change or increase with increasing of temperature.

Figure 6 shows the flow profile distribution of the concentration polarization that occurred on the membrane surface layer at three pressures; $P=15, 25$ and 35 bar in 30s of separation process. The color bar indicates the range of concentration polarization distribution of ion calcium carbonate scaling during separation process where the red color shows the maximum value, and the blue color shows the lowest value. The flow observation shows the formation of the scaling distribution and the thickness of the concentration polarization on the membrane surface layer is increased as the higher pressure and time elapsed. It can be seen at $P=35$ bar, the thickness of concentration polarization formation on the membrane surface is distributed equally and higher as compared to others condition. However, the effect of pressure on the formation of scaling on the membrane surface is relatively small and low at all tested pressures which is less than 0.3 mol/dm^3 as compared to the ion concentration and temperature's effects.

From these observation results and color distribution on the membrane surface, it shows concentration polarization is highest at the $P=35$ bar. It was happened due to the higher pressure induces less formation of CaCO_3 on the membrane surface due to small solubility rate of CaCO_3 , then the concentration polarization will occur [38]. Besides, the pressure effect shows strongly influencing on the formation of calcium carbonate scales [36] as the formation of a calcium carbonate from the carbonate anions is dependent on the quantity of carbon dioxide present in the solution. At the higher carbon dioxide partial pressure, the solubility rate of calcium carbonate is more than double due to air saturation.

As the pressure is applied to the feed flow portion on the membrane surface layer during the separation process, the solute will accumulate on the membrane surface, whereas the solvent will pass through the membrane surface layer. Due to the accumulation of solute that occurs at the membrane surface, the concentration of solute in the diffusion part is quite lower than the concentration in the

feed part. This condition will cause solute concentration to gradually increase at the membrane surface due to the accumulation of solute from the convective flow. At certain times, the convective flow of solutes at the membrane surface layer will be offset by the flux of solutes passing through membrane and the diffusion flow from membrane surface to bulk flow to form the concentration polarization on the membrane surface layer [8, 39].

3.2 Effect of concentration polarization on separation efficiency

The concentration polarization formed at the membrane surface will reduce the efficiency of the separation process as well it might limit the flux rate and permeability of the membrane process. Once the concentration polarization occurs, it will reduce the flow of water flux and solute rejection, causing the precipitation due to high surface concentrations that exceed the solubility limit and change the membrane separation properties. Figure 7 shows the effect of ions solution concentration and temperature on the diffusion permeate flux of separation membrane. In the Figure 7(a), it shows slightly small varies where the lowest value of the diffusion flux is shown at $C_i=15,000$ mg/L and the highest value was observed at $C_i=5000$ ppm due to formation of concentration polarization on the membrane surface. Meanwhile, on the temperature effect as shown in the Figure 7(b) shows the highest value of the diffusion flux was observed at $T=45$ °C and the lowest value at $T=25$ °C. This indicates the diffusion flux increases with increasing of temperature as the ion diffusion rate was increases. Since the flux is influenced by adsorption of soluble metal hydroxide species on the membrane surface, the formation of a cake layer is consisting of the metal hydroxide deposits, concentration polarization, and osmotic pressure [40]. Thus, the organic aggregates and the inorganic particles will lower the membrane permeability flux.

From all these simulation results, it was observed that in the RO separation process, the concentration polarization phenomenon is an important factor and need to be overcome as it might be led to the formation of larger salt concentration differences between the membrane layer as well will increase the osmotic pressure of the solution at the membrane surface layer. Moreover, it will also assistance the formation of membrane dirt scales in the membrane surface layer and eventually result in reducing a low performance of RO separation process. However, the main adverse effect of the concentration polarization is to reduce the diffusion flux by increasing the hydraulic resistance to water flow as well as reducing the pressure driving force across the membrane by increasing the osmotic pressure in the boundary layer.

This phenomenon in turn will lead to an increasing of impurities on the membrane surface as it will increase the concentration of contaminants such as bacteria, organic and

inorganic substances at near to the membrane surface, which exacerbates the formation of membrane impurities. As the denser concentration of solution, the diffusion flux will also decrease. This is because as the concentration of the solution is increases, the osmotic pressure will also increase, thus the driving force will decrease that in turn will cause the diffusion flux to decrease.

3.3 Comparison of results on the current study with the previous researchers

This study concludes that the highest concentration polarization formation was observed at $T=60$ °C and $P=35$ bar. The similar finding resulted by Ning 2015 where the concentration formation of calcium carbonate scaling was increased at the higher temperature as CO_2 gas is generally more soluble under the higher pressure [18]. It has been verified that the concentration polarization occurs at a high rate with increasing of the temperature as the temperature affects a water viscosity, solubility, and mass transfer coefficient of solute. As the higher temperature, the water's viscosity will decrease, and this condition will cause an increment in the diffusion flux. Also, this condition will contribute to formation of significant scales on membrane surface [37]. The higher temperature also resulted in lower solubility rate of $CaCO_3$ which causing concentration polarization to easily be formed on the membrane surface. In case of pressure effect, Muryanto et al. 2019 and Bai et al. 2014 also found that as the pressure drops, the carbon dioxide gas will oxidize from the solution, causing a decrement in the concentration of carbonic acid and the formation of calcium carbonate scaling tend to be occurred [34-35]. A studied by Troup et al. 1978 shows the pressure strongly influencing on the formation of calcium carbonate scales where at the higher carbon dioxide partial pressure, the solubility rate of calcium carbonate is more than double due to air saturation [36].

4. Conclusion

Numerical simulation has been conducted and successfully simulated by using AutoCAD® and COMSOL® Multiphysics software to observe the effect of temperature, pressure, and ions concentration on the scaling formation and concentration polarization on the membrane surface layer. The simulation results show in a diluted ion concentration, with the higher temperature and lower pressure resulted in an increasing of the rate of formation of $CaCO_3$ on membrane surface with the salt is precipitated at $C_o \geq 24$ mg/L. At $T=30, 45$ and 60 °C, the diffusion flux obtained are 21.9, 24.6 and 26.8 L/m^2hr , respectively. At $C_i=5000, 10,000$ and $15,000$ mg/L, the diffusion flux obtained are 22.3, 25.7 and 27.4 L/m^2hr , respectively. Next, at $P=15, 25$ and 35 bar, the diffusion flux obtained are 21, 28 and 34 L/m^2hr , respectively. This finding will help to

estimate the formation of concentration polarization and scaling in the membrane separation process. Future studies may be possible by considering crystal nucleation simulations (scales), which basis of crystal growth studies for various crystals as well as the membrane surfaces covered by these crystals.

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6. References

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Figures

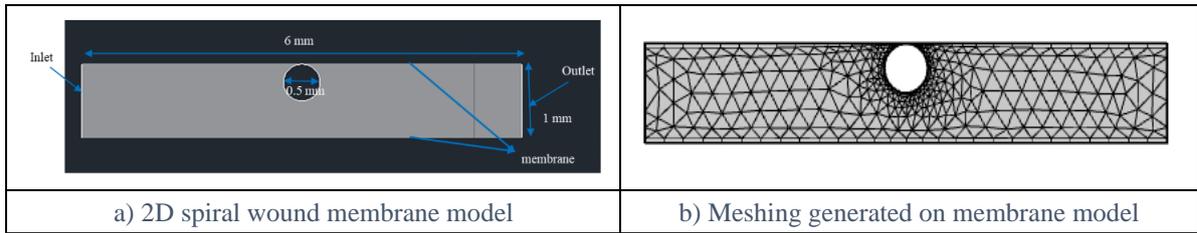


Figure 1. Configuration and dimension of 2D spiral wound membrane model

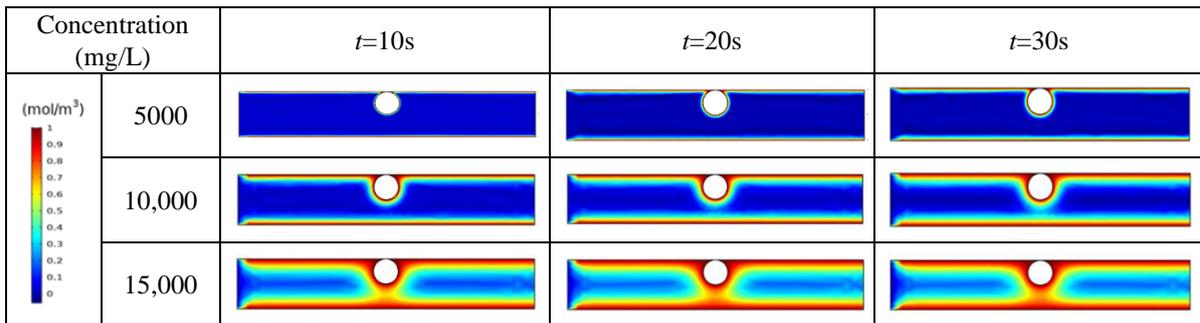


Figure 2. Concentration polarization profile in the membrane model at different inlet concentration in $t=30s$ of separation process

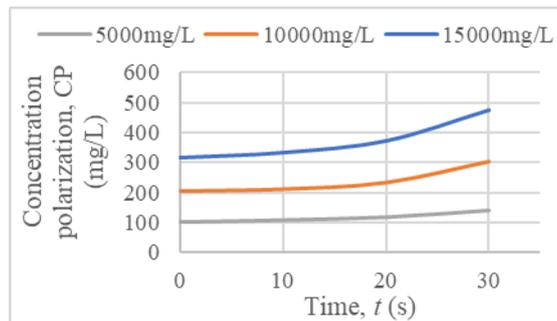


Figure 3. The effect of ion solution concentration on the polarization of $CaCO_3$ concentration

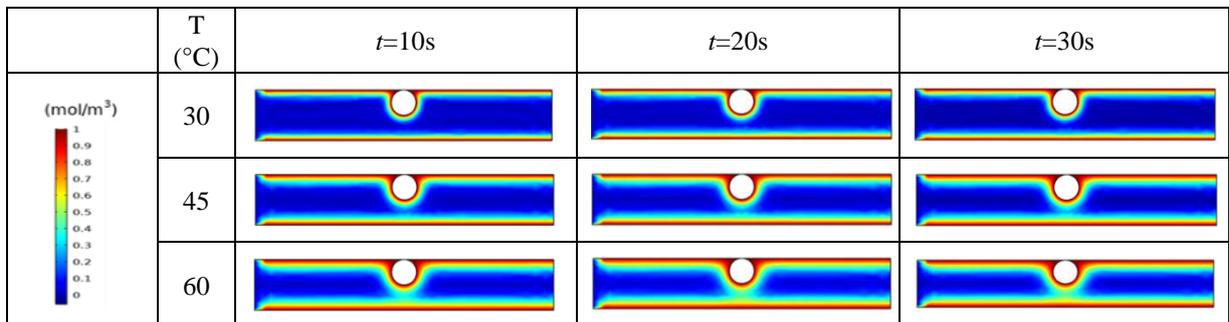


Figure 4. Concentration polarization profile in the membrane model at different inlet temperature in $t=30s$ of separation process

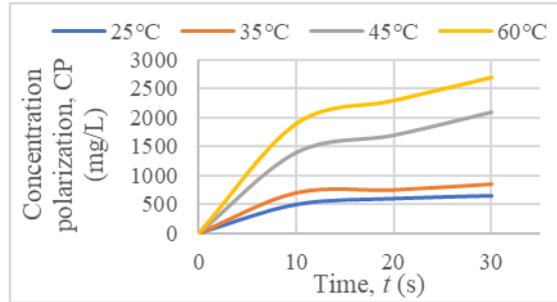


Figure 5. Temperature effect on concentration polarization in 30s

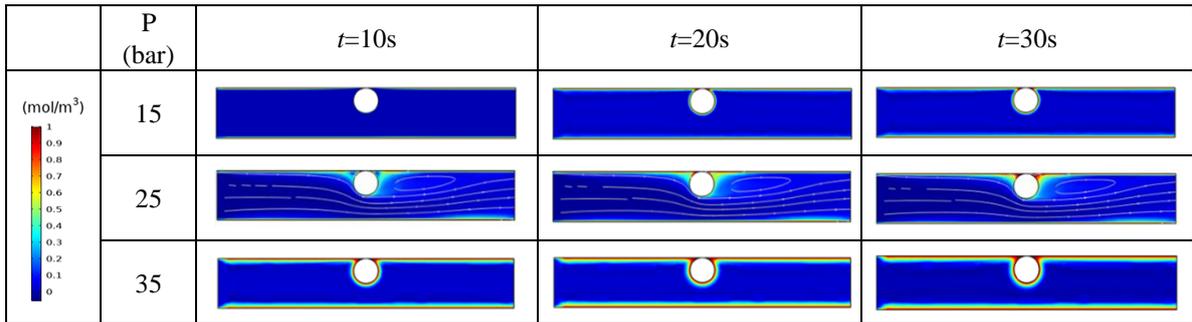


Figure 6. Concentration polarization profile in the membrane model at different inlet pressure in t=30s

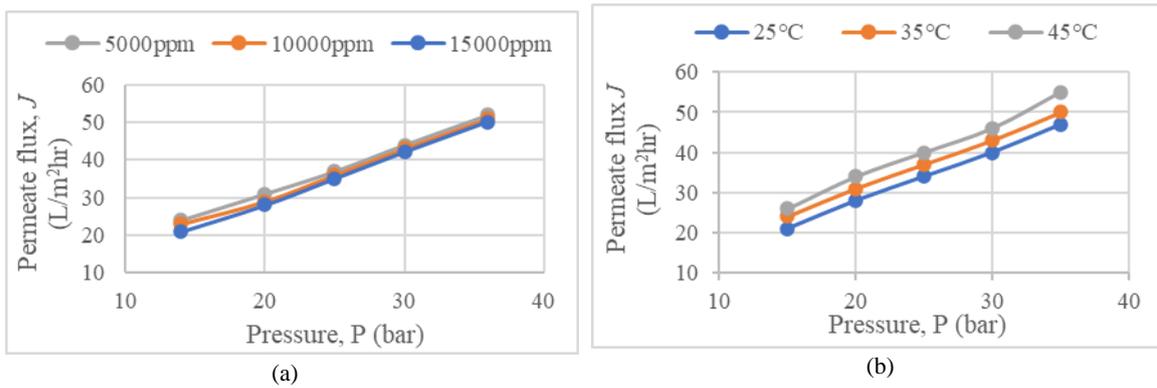


Figure 7. The effect of a) ions concentration polarization and b) temperature on permeate flux

Table

Table 1. Physical properties

Properties	Value
Liquid density, ρ	1000 kg/m ³
Liquid dynamic viscosity, μ_l	10 ⁻³ Pa/s
Diffusion coefficient NaCl, K_D	1.3 × 10 ⁻⁹ m ² /s
Inlet velocity, v	0.25 m/s
Membrane permeability, ϵ_m	9.56 × 10 ⁻¹² m/s·Pa
Viscosity, μ	1.0 × 10 ⁻⁶ m ² /s
Permeability NaCl, ϵ_s	1.5 × 10 ⁻⁹ m ² /s