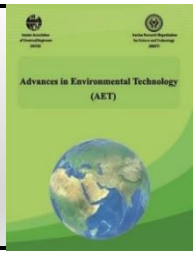
Advances in Environmental Technology 3 (2019) 141-148

Advances in Environmental Technology

journal homepage: http://aet.irost.ir



Mass transfer study in saline water treatment by forward osmosis process

Razieh Ahmadizadeh, Soheila Shokrollahzadeh, Seyed Mahdi Latifi*

Department of Chemical Technologies, Iranian Research Organization for Science and Technology (IROST), Tehran, Iran

ARTICLE INFO

Article history:
Received 3 December 2019
Received in revised form
14 Jun 2020
Accepted 22 Jun 2020

Keywords:
Forward osmosis (FO)
Brine wastewater
Concentration polarization
Mass transfer

ABSTRACT

Forward osmosis (FO) is an energy-saving separation process that can be used in desalination applications. This work investigated the effect of mass transfer phenomenon on the FO desalination process. For this purpose, the water flux was studied through a bench scale system using a flat sheet FO membrane and feeds with various salinity. Then, the mass transfer resistances, which appear in the form of concentration polarization (CP) for the FO process, were evaluated qualitatively and quantitatively, using the collected experimental data and by employing a mathematical model. The results indicated that the increase in feed salinity led to a decrease in water flux due to the counteracted part of the draw solution osmotic pressure, thus leading to a lower effective osmotic pressure and driving force. Also, according to the results, there was a significant difference between the theoretical and experimental fluxes, indicating the influence of the mass transfer effects on the osmotic pressure drop. The modeling results showed that the internal concentration polarization (ICP) still held more contribution to the osmotic pressure loss. Furthermore, it was observed that as the feed solution concentration increased, both the ICP and dilutive external concentration polarization (DECP) decreased, whereas the concentrative ECP (CECP) intensified. Therefore, increasing the CECP led to a significant reduction in the effective osmotic pressure. In addition, increasing the draw solution concentration was accompanied by a much more severe ICP that limited the enhancement of effective flux.

1. Introduction

Forward osmosis (FO) is known as a novel membrane technology for water and wastewater treatment that can be used in such processes as desalination [1], wastewater treatment [2], power supply [3], food industries [4] and RO brine reducing or reuse [5]. In this process, the osmosis phenomenon acts as a driving force for transporting water from a saline feed solution to a greater concentrated solution, as draw solution across a semi-permeable membrane without demanding any external force [6,7]. Recently, several review papers have given an extensive account of the advantages and disadvantages of the forward osmosis process [8-10]. The comprehensive application of FO, especially in comparison to the RO process, is due to its two significant advantages: lower energy consumption and lower fouling propensity [11]. In fact, the absence of hydraulic pressure in the FO process has resulted in minimal irreversible fouling and lower operating costs [12,13]. However, mass transfer effects, which appear in the form of concentration polarization (CP), challenge the water recovery in the FO process and causes some non-linearity in operation [14]. The mass transfer effects mostly include the external concentration polarization (ECP) and the internal concentration polarization phenomena. By permeating water from the feed solution across the membrane, concentrative and dilutive ECP occurs in the feed and draw solution sides, respectively. In the ICP phenomenon, the draw solution is diluted by the infused water through the porous support layer, which causes a reduction in the difference of the effective osmotic pressure and has a more severe impact on decreasing water flux than ECP. Literature surveys reveal several modeling studies on FO [15-18] in which mass transfer equations have been

*Corresponding author. Tel: +98 2156276624
E-mail address: latifi@irost.ir
DOI: 10.22104/AET.2020.3946.1195

proposed based on film and convective-diffusion theories. Literature surveys reveal that research has been conducted on concentrative polarization modeling to predict the resulted water flux. Loeb et al. [19] modeled ICP by introducing the solute resistivity for diffusion through the porous layer. Tan and Ng [15] studied the CP impact on flux behavior with the extending the diffusion coefficient of the solute represented as a non-linear power series. Qin et al. [20] predicted water flux using CP modeling and introduced a simple approach to choose a suitable draw solution. Since the ICP has been more severe in reducing the water flux in many mathematical models, the ECP effects have been ignored. Indeed, some investigations developed mathematical models that are able to simulate the ECP on both sides of the membrane. Suh et al. [21] and Wang et al. [22] investigated the effect of the feed and draw solution concentrations (while keeping the theoretical osmotic pressure constant) and found that with an increase in the solution concentrations, the effective osmotic pressure across the active layer decreased as a result of the ICP and ECP effects. As mentioned above, the effects of mass transfer play an important role in FO performance, particularly in higher amounts of salinity; so, modeling these phenomena can help us investigate influencing parameters and predict the process performance in various conditions. Literature surveys imply that in most of the research carried out in this area, the concentration of the draw solution has been investigated as a variable, and the used feed solution mostly included deionized water. In this work, the aim was to investigate the effect of solute concentration variation in the feed solution on the mass transfer resistances. For this, a set of bench scale experiments were conducted to measure the water flux through the FO process with various concentrations of feed and draw solutions. Then, using the collected experimental data and employing a mathematical model, the CP phenomena (concentrative ECP, ICP, and dilutive ECP) were evaluated qualitatively and quantitatively.

2. Materials and methods

The FO membrane used in the study was supplied by Hydration Technology (HTI, Albany, Oregon, US) and was made of cellulose triacetate embedded in a polyester screen mesh. The NaCl of analytical grade (99.9% Fisher) was used to prepare the feed solution and used as a draw solution.

2.1. Forward osmosis cross flow set-up

The laboratory system employed was carried out under the FO mode (i.e., active layer facing the feed solution) with an effective membrane area of 95.76 cm^2. A schematic of the experimental setup is shown in Figure 1. The feed and draw solutions (placed in 3L tanks and kept at 25°C) were recirculated with a flow rate of 3 L/min by using two low-pressure magnetic pumps (MP-045 B, CSE Co., Korea). The feed solution tank was placed on a load cell (Model 640 single point, Revere Transducers Europe BV, Denmark), and weight changes were recorded for calculating the permeate water flux. The conductivity of the draw solution was continuously measured via a conductivity probe (LF 96, WTW Co., Germany) to regulate the draw solution concentration by using a reservoir containing 4 M NaCl stock solution.

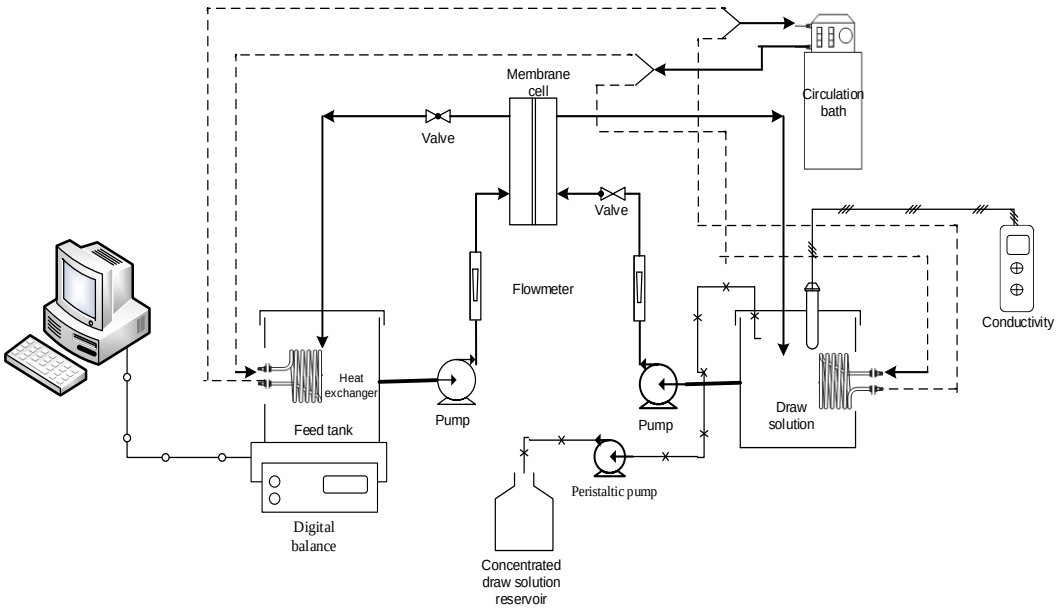


Fig. 1. Schematic diagram of the laboratory-scale forward osmosis system.

3. Experiments

The experiments were conducted to investigate the effect of different brine on the FO process with dissolved NaCl concentrations (0-30 g/l) in the feed solution. The draw solutions were 1 M and 2 M NaCl. Table 1 presents the experimental conditions and corresponding osmotic pressures of the solutions calculated by using an OLI Stream Analyser 3.2 (OLI Systems Inc., Morris Plains, NJ).

Table 1. Values of NaCl concentrations in feed and draw solutions (and corresponding osmotic pressure).

No.	Concentration of feed solution, NaCl (g/L)	Concentration of draw solution NaCl (M)	Bulk osmotic pressure, of feed solution π_f, (atm)	Bulk osmotic pressure of draw solution , π_D(atm)
1	0	1	0.00	46.77
2	5	1	3.91	46.77
3	10	1	7.76	46.77
4	15	1	11.62	46.77
5	20	1	15.51	46.77
6	25	1	19.43	46.77
7	30	1	23.39	46.77
8	15	2	11.62	100.39
9	20	2	15.51	100.39
10	25	2	19.43	100.39
11	30	2	23.39	100.39

3.1. Water Flux (J_w) in forward osmosis process

The theoretical water flux through a semi-permeable membrane by osmotic pressure gradient (Δπ) is calculated as [23]:

$$J_w = A\Delta\pi = A(\pi_D - \pi_F) \quad (1)$$

where J_w is the water flux, A is the water permeability coefficient of the membrane, and π_D and π_F are the bulk osmotic pressures of the draw and feed solutions, respectively. For this type of CTA membrane, A was reported to be about 0.857 L/m^2.hr.bar [24]

The experimental water flux is determined by measuring the weight difference of the feed solution at regular time intervals using a computer and a data logging system (LabVIEW) as follows:

$$J_W = \frac{\Delta W}{\rho \times t \times S} \quad (2)$$

where ΔW, ρ, t and S are the decrease in weight of the feed solution(g), density of water, membrane effective area (m²), and time interval (h), respectively.

3.2. Determination of the performance ratio (PR) in the FO process

It has been recounted in previous studies that theoretical flux corresponding to the osmotic pressure differential (π_D-π_F) is higher than the experimental water flux, resulting from the effects of internal concentrative polarization and external concentrative polarization. PR quantifies these effects by considering the percentage of the bulk effective osmotic pressure used for producing water flux in the FO process and is defined as [6].

$$PR = \frac{J_{Experimental}}{J_{Theoritical}} \times 100 \quad (3)$$

3.3. Effect of concentration polarization on drop of osmotic pressure

In this work, both the ECP and ICP of the FO process were computed using an earlier developed model. Figure 2 depicts the profile of the solute concentration in the FO membrane adjacent. In Figure 2, $C_{f,m}$ is the solute concentration on the membrane surface of the active layer, C_i represents the solute concentration at the interface between the active layer and support layer, and $C_{D,m}$ stands for the solute concentration on the membrane surface of the support layer. Under steady-state conditions, C_{Fm}, C_i, and C_{Dm} can be expressed by [22].

$$C_{F,m} = \left(C_{F,b} + \frac{J_S}{J_W}\right)\exp\left(\frac{J_W}{k_f}\right) - \frac{J_S}{J_W} \quad (4)$$

$$C_i = \left(C_{D,m} + \frac{J_S}{J_W}\right)\exp(-J_W K) - \frac{J_S}{J_W} \quad (5)$$

$$C_{D,m} = \left(C_{D,b} + \frac{J_S}{J_W}\right)\exp\left(-\frac{J_W}{k_d}\right) - \frac{J_S}{J_W} \quad (6)$$

where J_S(mol/m^2.h) is reverse salt flux, J_W(L/m^2.h) stands for water flux, K is solute resistivity for diffusion through the porous layer, $C_{F,b}$ is feed solution concentration, and $C_{D,b}$ stands for draw solution concentration. k_f and k_d are mass transfer coefficients for the feed and draw solutions, respectively.

According to Figure 2, ICP, CECP, and DECP can be defined as follows:

$$ICP = C_{D,m} - C_i \quad (7)$$

$$DECP = C_{D,b} - C_{D,m} \quad (8)$$

$$CECP = C_{F,m} - C_{F,b} \quad (9)$$

$$Effective = C_i - C_{F,m} \quad (10)$$

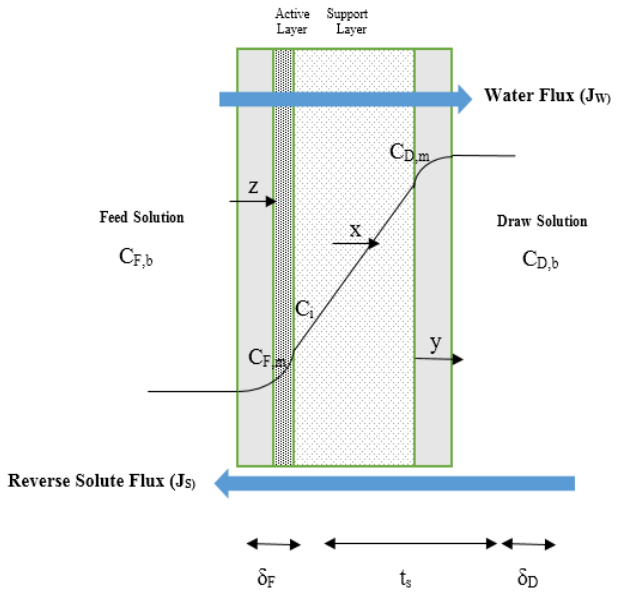


Fig. 2. Schematic of the feed and draw solution concentration profile across FO membrane.

4. Results and discussion

The feed solution concentration is an important parameter that influences the resulting water flux in the FO process. Figures 3 and 4 illustrate the water flux profiles at different feed salinities using 1M and 2M draw solutions, respectively. Figures 3 and 4 show that the water flux is slightly reduced as time passes, but after a while, no decrease is observed. Since the draw solution concentration is kept almost constant during the process, this decrease can be attributed to increasing the concentration of feed solution that leads to decreasing the effective driving force. Also, it is obvious in Figures 3 and 4 that increasing the feed salinity causes a reduction in the water flux as a result of decreasing the osmotic driving force. A comparison between the results shown in Figures 3 and 4 reveals that the draw solution with a higher concentration results in an enhanced water flux due to increasing osmotic difference as the driving force.

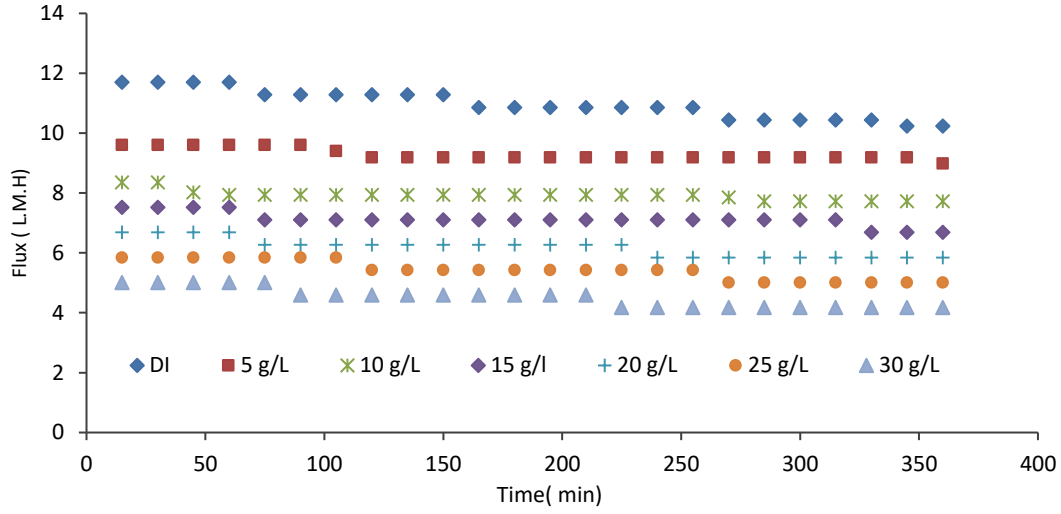


Fig. 3. Effect of NaCl concentration (0 to 30 g/l) on Water flux (Draw solution 1M NaCl and temperature 25 °C).

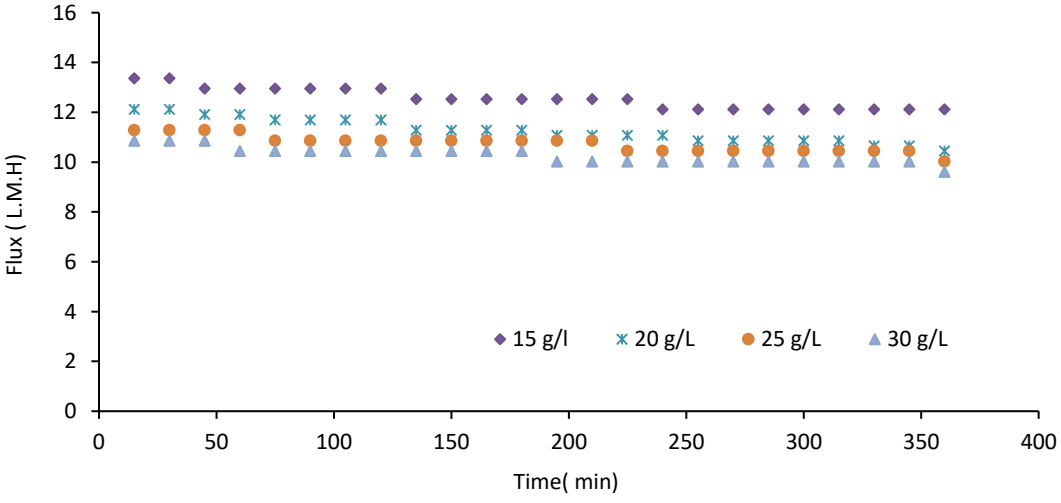


Fig. 4. Effect of NaCl concentration (15 to 30 g/l) on water flux (Draw solution 2M NaCl and temperature 25 °C).

4.1. Modeling without considering concentrative polarizations (CPs)

Figure 5 depicts the difference between the experimental water flux measurements with the theoretical one calculation in various concentrations of feed and draw solutions. This difference stems from the fact that in the theoretical calculation, the flux is only the function of the osmotic pressure difference (Eq. 1), whereas such mass transfer effects as ECP and ICP play a significant role in real obtained flux. It can be seen in Figure 5 that by increasing the feed solution concentration, the difference between the theoretical and experimental water flux goes down and can be ascribed to the ICP reduction as a result of a decrease in water flux.

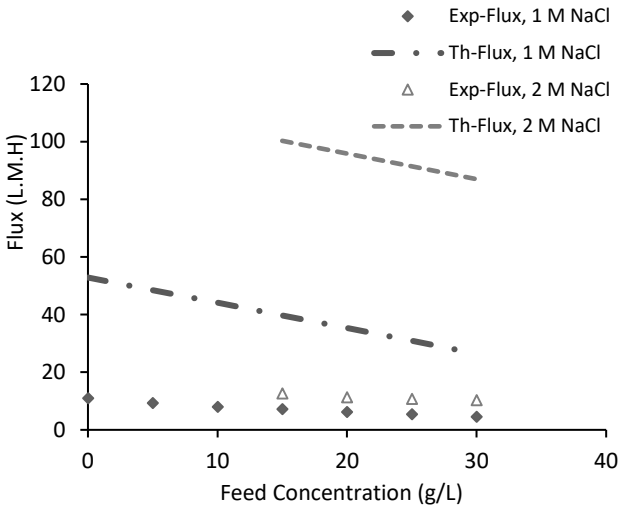


Fig. 5. Experimental and the theoretical water flux under various concentrations of feed (from 0 to 30 g/l) and draw solutions (1-2 M NaCl) in temperature 25 °C. (EXP-Flux: experimental flux, Th: theoretical).

As mentioned before, the discrepancy between the theoretical and experimental results can be shown with the PR (%) for various feed and draw solution concentrations (Table 2). The significant difference observed in Table 2 between the experimental and theoretical results stems from the fact that the theoretical flux is only corresponded to the osmotic pressure differential (without considering CPs), while in fact CPs have a great influence on the obtained flux. Also, it can be seen in Table 2 that by increasing the draw solution concentration from 1M to 2M, the PR% decreases significantly while this is not the case for increasing the feed solution concentration. This finding implies that the ICP that results from the draw solution concentration is more effective than the ECP that is caused by the feed solution concentration, which is in agreement with previous researches [6,25].

Table 2. Performance ratio determined using theoretical and experimental flux for various NaCl solutions as feed. Membrane permeability coefficient (A) = 1.13 $Lm^{-2}h^{-1}atm^{-1}$

Feed Conc. (g/L)	Draw Conc. (M)	FLUX (L.M.H) (Theoretical)	FLUX (L.M.H) (Experimental)	PR (%)
0	1	52.85	10.97	20.7
5	1	48.43	9.29	19.2
10	1	44.08	7.92	18.0
15	1	39.72	7.12	17.9
20	1	35.32	6.18	17.5
25	1	30.89	5.43	17.6
30	1	26.42	4.51	17.1
15	2	100.28	12.55	12.5
20	2	95.88	11.26	11.7
25	2	91.45	10.2	11.15
30	2	86.98	9.41	10.8

4.2. Modeling by considering CPs

In this section, the effects of concentrative polarizations in the feed and draw solutions sides were introduced to the water flux prediction model by using Eqs. 4 to 10; the results are presented in Figures 6 to 8.

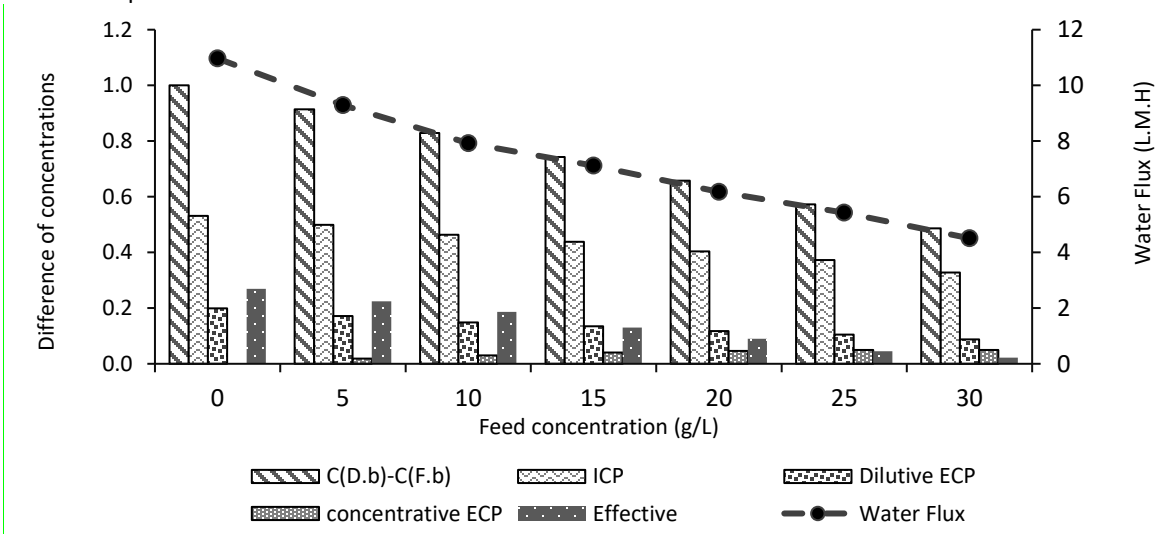


Fig. 6. Evaluation of osmotic pressure drop by CPs simulation in various concentrations of feed (from 15 to 30 g/l) and draw solution 1M NaCl.

As Figures 6 and 8 show, in the lower feed salinity, that was connected with the higher water flux, the severity of the concentrative ECP on the driving force was reduced, and ICP played a prevailing role in the performance of the FO process. In this condition, the solute concentration of the draw solution bulk was greater than that of the support layer, which led to the larger difference between $C_{D,m}$ and C_i (Figure 7). On the other hand, the higher feed salinity led to the accumulation of solute on the active layer membrane, which resulted in decreasing the ICP and dilutive ECP influences (shown in Figures 6 and 8). A comparison between Figures 6 and 8 revealed that as the draw solution concentration increased from 1 M to 2 M, the ICP and dilutive ECP was enhanced more than 2.5 and 3 times, respectively. Under this condition, it could be predicted that any further increase in the draw solution concentration was counteracted by a much more severe ICP, resulting in a less effective flux enhancement. Evaluating the abovementioned modeling results reveals that in all the studied conditions, the mass resistance of the ICP is greater than that of the ECP and can be attributed to the significant difference between mass diffusion coefficients through porous and liquid media. This finding is verified by other works [26] that investigated the contributions of mass transfer resistances in driving force loss.

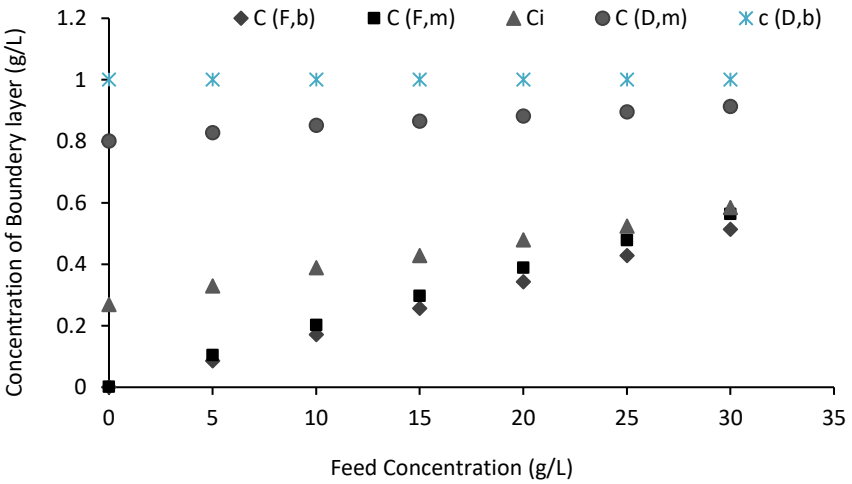


Fig. 7. The solute concentration in the FO membrane adjacent.

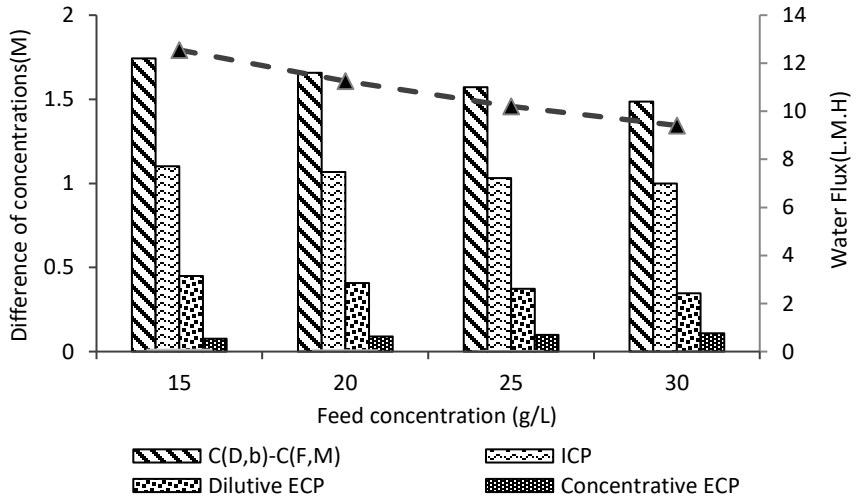


Fig. 8. Evaluation of osmotic pressure drop by CPs simulation in various concentrations of feed (from 15 to 30 g/l) and draw solution 2M NaCl.

5. Conclusions

This work focused on the effects of mass transfer on the FO process in various concentrations of feed solution. According to the obtained results, with increasing feed solution salinity, the water flux decreased, and the ICP effects became limited in the FO process. In addition, the difference between the theoretical flux, calculated from the solution thermodynamics, and that resulting from the

experiments revealed that increasing the draw solution concentration led to a reduction in the percentage utilization of the effective osmotic pressure (PR %) due to an increase in the dilutive ICP phenomena.

Nomenclature

A water permeability coefficient of the membrane (L/m^2.hr.bar)
$C_{D,b}$ draw solution concentration (M)
$C_{D,m}$ solute concentration on the membrane surface of support layer (M)
$C_{F,b}$ feed solution concentration (M)
$C_{f,m}$ solute concentration on the membrane surface of active layer (M)
C_i solute concentration at the interface between the active layer and support layer (M)
J_S reverse salt flux (L/m^2.h)
J_w water flux (L/m^2.h)
K solute resistivity for diffusion within the porous support layer (s/m)
k_d mass transfer coefficient of draw side (m/s)
k_f mass transfer coefficient of feed side (m/s)
S membrane effective area (m²)
t time interval (h)
ΔW decrease in weight of the feed solution (g)
π_D bulk osmotic pressure of the draw solution (bar)
π_F bulk osmotic pressure of the feed solution (bar)
ρ density of water (kg/m^3**)**

Acknowledgement

The authors are grateful to the South Pars Gas Company (SPGC) for its support.

References

[1] Qin, J. J., Lay, W. C. L., Kekre, K. A. (2012). Recent developments and future challenges of forward osmosis for desalination: a review. *Desalination and water treatment*, *39*(1-3), 123-136.

[2] Hey, T., Bajraktari, N., Davidsson, Å, Vogel, J., Madsen, H. T., Hélix-Nielsen, C., Jönsson, K. (2018). Evaluation of direct membrane filtration and direct forward osmosis as concepts for compact and energy-positive municipal wastewater treatment. *Environmental technology*, *39*(3), 264-276.

[3] Kim, J., Jeong, K., Park, M. J., Shon, H. K., Kim, J. H. (2015). Recent advances in osmotic energy generation via pressure-retarded osmosis (PRO): a review. *Energies*, *8*(10), 11821-11845.

[4] Hasanoğlu, A., Gül, K. (2016). Concentration of skim milk and dairy products by forward osmosis. *Journal of the Turkish Chemical Society Section B: Chemical engineering*, *1*(1), 149-160.

[5] Johnson, D. J., Suwaileh, W. A., Mohammed, A. W., Hilal, N. (2018). Osmotic's potential: An overview of draw solutes for forward osmosis. *Desalination*, *434*, 100-120. pressure retarded osmosis (PRO). *Separation and purification technology*, *156*, 856-860.

[6] J McCutcheon, J. R., McGinnis, R. L., Elimelech, M. (2006). Desalination by ammonia–carbon dioxide forward osmosis: influence of draw and feed solution concentrations on process performance. *Journal of membrane science*, *278*(1-2), 114-123.

[7] Cath, T. Y., Childress, A. E., Elimelech, M. (2006). Forward osmosis: principles, applications, and recent developments. *Journal of membrane science*, *281*(1-2), 70-87.

[8] Qasim, M., Darwish, N. A., Sarp, S., Hilal, N. (2015). Water desalination by forward (direct) osmosis phenomenon: A comprehensive review. *Desalination*, *374*, 47-69.

[9] Johnson, D. J., Suwaileh, W. A., Mohammed, A. W., Hilal, N. (2018). Osmotic's potential: An overview of draw solutes for forward osmosis. *Desalination*, *434*, 100-120

[10] Akther, N., Sodiq, A., Giwa, A., Daer, S., Arafat, H. A., Hasan, S. W. (2015). Recent advancements in forward osmosis desalination: a review. *Chemical engineering journal*, *281*, 502-522.

[11] Tow, E. W., Warsinger, D. M., Trueworthy, A. M., Swaminathan, J., Thiel, G. P., Zubair, S. M., Myerson, A. S. (2018). Comparison of fouling propensity between reverse osmosis, forward osmosis, and membrane distillation. *Journal of membrane science*, *556*, 352-364.

[12] Li, L., Liu, X. P., Li, H. Q. (2017). A review of forward osmosis membrane fouling: Types, research methods and future prospects. *Environmental technology reviews*, *6*(1), 26-46.

[13] Linares, R. V., Li, Z., Yangali-Quintanilla, V., Ghaffour, N., Amy, G., Leiknes, T., Vrouwenvelder, J. S. (2016). Life cycle cost of a hybrid forward osmosis–low pressure reverse osmosis system for seawater desalination and wastewater recovery. *Water research*, *88*, 225-234.

[14] Phuntsho, S., Hong, S., Elimelech, M., Shon, H. K. (2014). Osmotic equilibrium in the forward osmosis process: Modelling, experiments and implications for process performance. *Journal of membrane science*, *453*, 240-252.

[15] Tan, C. H., Ng, H. Y. (2008). Modified models to predict flux behavior in forward osmosis in consideration of external and internal concentration polarizations. *Journal of membrane science*, *324*(1-2), 209-219.

[16] McCutcheon, J. R., Elimelech, M. (2007). Modeling water flux in forward osmosis: implications for improved membrane design. *AIChE journal*, *53*(7), 1736-1744.

[17] Phillip, W. A., Yong, J. S., & Elimelech, M. (2010). Reverse draw solute permeation in forward osmosis: modeling and experiments. *Environmental science and technology*, *44*(13), 5170-5176.

[18] Bae, C., Park, K., Heo, H., Yang, D. R. (2017). Quantitative estimation of internal concentration polarization in a spiral wound forward osmosis membrane module compared to a flat sheet membrane module. *Korean journal of chemical engineering*, *34*(3), 844-853.

[19] Loeb, S., Titelman, L., Korngold, E., Freiman, J. (1997). Effect of porous support fabric on osmosis through a Loeb-Sourirajan type asymmetric membrane. *Journal of membrane science*, *129*(2), 243-249.

[20] Qin, J. J., Chen, S., Oo, M. H., Kekre, K. A., Cornelissen, E. R., Ruiken, C. J. (2010). Experimental studies and modeling on concentration polarization in forward osmosis. *Water science and technology*, *61*(11), 2897-2904.

[21] Suh, C., Lee, S. (2013). Modeling reverse draw solute flux in forward osmosis with external concentration polarization in both sides of the draw and feed solution. *Journal of membrane science*, *427*, 365-374.

[22] Wang, Y., Zhang, M., Liu, Y., Xiao, Q., Xu, S. (2016). Quantitative evaluation of concentration polarization under different operating conditions for forward osmosis process. *Desalination*, *398*, 106-113.

[23] Helfer, F., Lemckert, C., Anissimov, Y. G. (2014). Osmotic power with pressure retarded osmosis: theory, performance and trends–a review. *Journal of membrane science*, *453*, 337-358.

[24] Ortega-Bravo, J. C., Ruiz-Filippi, G., Donoso-Bravo, A., Reyes-Caniupán, I. E., Jeison, D. (2016). Forward osmosis: Evaluation thin-film-composite membrane for municipal sewage concentration. *Chemical engineering journal*, *306*, 531-537.

[25] Phuntsho, S., Shon, H. K., Hong, S., Lee, S., Vigneswaran, S. (2011). A novel low energy fertilizer driven forward osmosis desalination for direct fertigation: evaluating the performance of fertilizer draw solutions. *Journal of membrane science*, *375*(1-2), 172-181.

[26] Bui, N. N., Arena, J. T., McCutcheon, J. R. (2015). Proper accounting of mass transfer resistances in forward osmosis: Improving the accuracy of model predictions of structural parameter. *Journal of membrane science*, *492*, 289-302.