

Concentration dependence modeling of reverse osmosis

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ABSTRACT

Osmosis is the natural flow of a solvent through a semi-permeable membrane from a less concentrated solution to a more concentrated one. This is driven by the osmotic pressure difference. If opposing external pressure is used that exceeds the osmotic pressure, the solvent will flow from the more concentrated solution towards the less concentrated one. Besides the external pressure, the flux of the solvent (water) describing the reverse osmosis is the function of the osmotic pressure, which, in the case of an ideal solution, according to the van't Hoff principle, is proportional to the salt concentration. The current study examines the concentration dependence of the permeate flux, complementing the traditional model with the osmotic pressure difference caused by the concentration polarization stemming from the crossflow. In the calculations for the real solution, the osmotic coefficient was used instead of the empirical forms of osmotic pressure, making the model more correct theoretically as well. The validity of the model was certified and the parameters determined with independently measured results based on literature regarding the reverse osmotic desalination of seawater.

Keywords: Reverse osmosis; Flux; Mass transfer coefficient; Concentration dependence; Ideal solution; Real solution

1. Introduction

Osmosis is the natural flow of a solvent through a semi-permeable membrane from a less concentrated solution to a more concentrated one. The driving force is the osmotic pressure difference, which is the function of the solvent, the type of dissolved material, and the concentration. When the developed hydrostatic pressure is the same as the osmotic pressure, the result is an osmotic equilibrium, when the net mass transfer through the membrane is zero. If we apply over-pressure on the side of the more concentrated solution, the direction of the natural osmosis is reversed. The solvent will flow from the more concentrated solution towards the less concentrated one. So this

process, reverse osmosis, is the reversal of natural osmosis due to hydrostatic pressure. In the case of reverse osmosis, the driving force is the hydrostatic pressure difference, and the mass transfer mechanism is molecular diffusion [1].

The permeate flux is proportional to the difference between the overpressure and the osmotic pressure differences, the mass transfer coefficient (permeability) [2]. Based on the van't Hoff law, the osmotic pressure and, accordingly, the permeate flux are linearly dependent on the concentration [3], while measurements show a different concentration dependence [4]. The van't Hoff law is applicable to small concentrations (ideal solution), while, in the case of real solutions, there is a considerable difference from the linear

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law, for which there are several empirical equations [5,6]. Osmotic pressure calculated this way can modify the linear dependence of permeate flux from the concentration of the solution [7].

In the case of reverse osmosis, as with all cross-flows, there is a concentration difference in the solution, which is perpendicular to the membrane wall [8]. This concentration polarisation further modifies the value of the osmotic pressure and, through this, the concentration dependence of the flux.

A study was performed using five commercial thin-film composite (TFC) membranes and an analog TFC membrane. The solution diffusion model coupled with film theory was used to calculate the permeate (water) and concentrate (salt) to account for the effect of concentration polarisation. The mass transfer coefficient was independent and a linear reduction in (salt) concentrate was observed while the permeate water remained constant below 45 g L^{-1} NaCl, however, when the salt concentration at the membrane surface exceeded 45 g L^{-1} , water permeance either increased, remained constant, or decreased [9].

For the reverse osmosis membrane process, one-dimensional steady-state and dynamic mathematical models have been developed based on the solution–diffusion model coupled with the concentration polarisation mechanism [10]. A detailed simulation using the dynamic model was carried out to gain a deeper insight into the process. The effect of the feed flow rate, pressure, temperature, and concentration of pollutants on the performance of the process measured in terms of salt rejection, recovery ratio and permeate flux has been investigated.

Concentration polarization and osmotic effects have been developed for the cross-flow filtration in a membrane channel [11], focusing on the coupling between laminar hydrodynamics and mass transfer with the steady Navier–Stokes equations under the Prandtl approximation, which offers a simplified framework to enforce the non-linear coupling between filtration and concentration polarisation at the membrane surface.

This study examines the concentration dependence of the permeate flux, complementing the traditional model with the osmotic pressure difference caused by the concentration polarization stemming from the crossflow. In the calculations for the real solution, we used the osmotic coefficient instead of the empirical forms of osmotic pressure, making the model more correct theoretically as well. The validity of the model was certified and the parameters were determined with independently measured results based on literature regarding the reverse osmotic desalination of seawater [4,12].

2. Model

2.1. Permeate flux

In the case of reverse osmosis, permeate flows depend on the diffusion of mass transport, both for salt and water. The permeate flux (J_p), which is essentially the water flux J_w , is created by the $\Delta p = p_f - p_p$ transmembrane pressure difference between the feed side (p_f) and the permeate side (p_p) and the $\Delta\pi$ osmotic pressure difference (Fig. 1), namely:

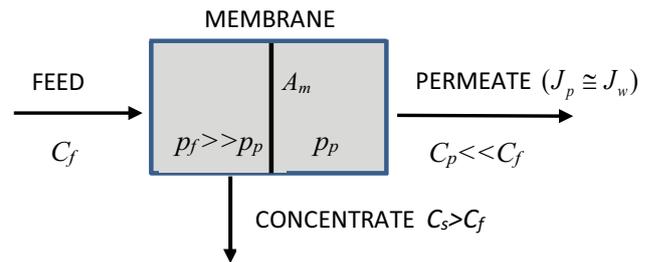


Fig. 1. Schematics of reverse osmotic membrane filtering with flux, concentrate, and pressure values.

$$J_w = K_w (\Delta p - \Delta\pi) \quad (1)$$

where K_w can be defined as the mass transfer coefficient for water (permeability) [2,13].

2.2. Osmotic pressure, thin solution

In Eq. (1) the osmotic pressure difference is dependent on the (feed) concentration:

$$\pi = \alpha CR_g T \quad (2)$$

According to the van't Hoff law, the osmotic pressure is linearly correlated with the concentration, where $\alpha = K_a \nu$, furthermore, K_a is the dissociation constant and ν is the dissociation number (in the case of NaCl, $K_a = 1$, $\nu = 2$, i.e., $\alpha = 2$). The osmotic pressure difference in Eq. (1) is the pressure difference between the two sides of the membrane:

$$\Delta\pi = \alpha \cdot (C_f - C_p) R_g T = \alpha \cdot C_f R_g T \quad (3)$$

where C_f is the feed-side concentration, and C_p is the exit or permeate-side concentration. Since $C_f \gg C_p$ for $\frac{C_f - C_p}{C_f} \geq 0.985$ (see below), therefore C_p close to zero when compared to C_f

$$J_w = K_w (\Delta p - \alpha C_f R T) \quad (4)$$

Accordingly, the permeate flux from Eqs. (1) and (3) can be written and that is linearly dependent on the C_f feed concentration [3,13].

2.3. Polarization

Crossflow takes place inside the membrane, during which the convection flow of the solvent (water) takes the (salt) molecules in the solution towards the membrane [13]. The direct flux is (Fig. 2):

$$J_s^+ = J_w C_f \quad (5)$$

The salt flux through the membrane, similarly to Eq. (5), can also be formulated as follows:

$$J_s = J_w C_p \quad (6)$$

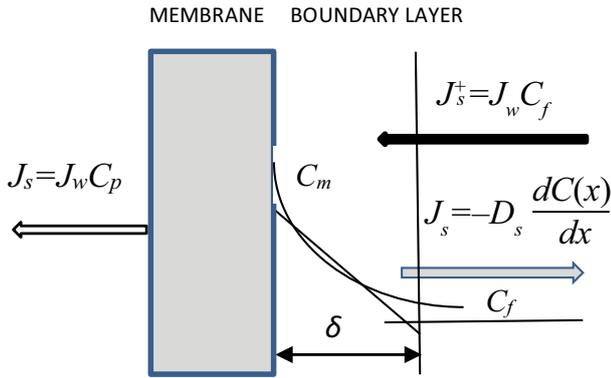


Fig. 2. Forming of polarisation layer in the case of crossflow with the fluxes.

Since $C_p \ll C_f$ only a small part of the salt molecules can cross the membrane, and the larger part gets stuck along the membrane wall, which causes a concentration difference (polarization) between the membrane wall and the flowing liquid (Fig. 2). As a result of the concentration difference, an indirect flux (J_s^-) forms opposite to the direct flux (J_s^+), for which the Fick-I law can be applied [13], that is:

$$J_s^- = -D_s \frac{dC(x)}{dx} \quad (7)$$

where D_s is the diffusion coefficient of the salt in the solvent (water), furthermore, $C(x=0) = C_m$ and $C(x \rightarrow \infty) = C_f$

In the steady state the following mass equation can be formulated for fluxes:

$$J_s^+ = J_s^- + J_w C_p \quad (8)$$

Substituting Eqs. (5)–(7), the next relation can be written for the water flux and salt concentrations:

$$J_w (C_f - C_p) = -D_s \frac{dC(x)}{dx} \quad (9)$$

In Eq. (9) the gradient can be approximated in the usual way (Fig. 2), namely:

$$\frac{dC(x)}{dx} = \frac{C_f - C_m}{\delta} \quad (10)$$

Accordingly, the mass equation ($C_p = 0$) can be simplified as follows:

$$J_w C_f = D_s \frac{C_m - C_f}{\delta} \quad (11)$$

where δ is the thickness of the boundary layer formed by the stuck salt molecules, and furthermore:

$$K_s^- = \frac{D_s}{\delta} \quad (12)$$

is the back mass transfer coefficient for the salt [13].

Regrouping Eq. (11) and using Eq. (12), the salt concentration along the membrane wall (C_m) can be formulated as follows:

$$\frac{C_m}{C_f} = \frac{J_w + K_s^-}{K_s^-} = 1 + \frac{J_w}{K_s^-} = \beta \quad (13)$$

In Eq. (13) β is the so-called concentration polarisation ($\beta > 1$) dimensionless number [14], which, through J_w in accordance with Eq. (4), can itself be dependent on the feed concentration C_f

2.4. Osmotic coefficient, the real solution

Eq. (2) for osmotic pressure is only relevant for ideal (thin) solutions. For a greater concentration range, there is a considerable difference from the linear correlation Eq. (2). According to experimental data, a polynomial of the third degree [5] or a power function [6] provide a better approximation. Using the theoretical equation for real solutions instead of the empirical approach for Eq. (3) [15], the osmotic pressure for the higher concentration now will be:

$$\Delta\pi = \alpha R_g T f_0 C_f \quad (14)$$

where f_0 the osmotic coefficient

$$f_0 = 1 - \frac{3.1 \cdot 10^4}{\epsilon_0 T^{\frac{3}{2}}} (v \cdot C_f)^{0.5} \cong 1 - 0.0105 \cdot C_f^{0.5} \quad (15)$$

is the function of the feed concentration, ϵ_0 is the dielectric constant of the solvent (water), and C_f is the salt concentration (mol m^{-3}). According to this, the permeate flux, instead of Eq. (4), will be the following for real solutions:

$$J_w = K_w (\Delta p - \alpha R_g T C_f^*) \quad (16)$$

and

$$C_f^* = f_0 C_f \quad (17)$$

equivalent concentration means the concentration value that would be the value associated with the identical osmotic pressure in the case of an ideal solution. Due to the Eq. (17) concentration dependence of the osmotic coefficient, the concentration dependence of the permeate flux differs from Eq. (4) linear.

2.5. Corrected osmotic pressure model

Henceforth, in the case of polarization, Eq. (14) osmotic pressure is dependent on the salt concentration by the membrane wall C_m . By substituting the value of this from Eq. (13), into Eqs. (16) and (17) for C_f in the case of polarisation, the following implicit equation for the Eq. (16) permeate (water) flux has been got for real solutions:

$$J_w = K_w \left[\Delta p - \alpha R_g T \left(1 + \frac{J_w}{K_s^-} \right) C_f^* \right] \quad (18)$$

Taking J_w out Eq. (18) and developing that into a series for the first degree of C_f^* , the permeate flux can be approximated well and expressed as the following dual-factor polynomial:

$$J_w \cong K_w \left[(1 - \alpha R_s T q C_f^*) \Delta p - \alpha R T C_f^* \right] \quad (19)$$

or

$$J_w \cong K_w \left[\Delta p - (1 + \alpha R_s T q \Delta p) C_f^* \right] \quad (20)$$

where

$$q = \frac{K_w}{K_s^-} \quad (21)$$

is the ratio of the two mass transfer coefficients. Eqs. (19) and (20) are different from Eq. (16) in that, the flux is also dependent on $\Delta p C_f^*$ (corrected osmotic pressure model). Without considering the polarisation ($K_s^- \rightarrow \infty$, $q \rightarrow 0$ full equalization), it becomes Eq. (16).

Furthermore, as a result of the solution flow, instead of Eq. (12) the back mass transfer coefficient for the salt will be as follows:

$$K_s^- = \text{Sh} \frac{D}{d} \quad (22)$$

introducing Sh, the Sherwood number characteristic of the flow, and d is a characteristic size [16].

The Sherwood number depends on the nature of the flow (laminar, turbulent) and the system geometry: tube, d diameter, l length of the lamella, etc., and on its parameters: v flow rate, η (water) viscosity, ρ solution density. In the case of laminar flow, the Sherwood number [17]:

$$\text{Sh} = 1.62 \left(\text{ReSc} \frac{d}{l} \right)^{\frac{1}{3}} \quad (23)$$

$\text{Re} = \frac{dv\rho}{\eta}$ is the Reynolds number and $\text{Sc} = \frac{\eta}{\rho D_s}$ is the Schmidt number.

2.6. Salt rejection

The salt flux can be written in the next form:

$$J_s = K_s (C_f - C_p) \cong K_s C_f \quad (24)$$

where K_s is the mass transfer coefficient of salt through the membrane, and $C_f \gg C_p$, therefore $C_p = 0$.

The salt rejection factor (expressed as a percentage) as a parameter characteristic of selectivity, can be expressed in the following form [13]:

$$\mathbf{R} = \left[1 - \frac{C_p}{C_f} \right] \cdot 100(\%) \quad (25)$$

Using Eqs. (6) and (24) and rearranging Eq. (25) as follows (conjugated salt rejection factor):

$$100 - \mathbf{R} = K_s \frac{100}{J_w} \quad (26)$$

provides a line as the function of the inverse of the permeate flux, the slope of which is the mass transfer coefficient for salt K_s .

3. Experiments

3.1. Materials

The measurements necessary for proving the model, the permeate flux and salt rejection as the function of concentration (Fig. 3) and pressure (Fig. 4) were taken from the studies of Cadotte et al. [5] and Al-Mutaz, Al Ghunaimi [12] on a 3.5% salt solution detailed in another paper [18]. Considering the measurement units used in practice units of measurement have been recalculated for flux ($\text{Lm}^{-2} \text{h}^{-1} = 2.78 \times 10^{-7} \text{ m}^3 \text{ m}^{-2} \text{ s}^{-1}$), for pressure ($\text{bar} = 10^5 \text{ Pa}$), and for concentration ($\text{m}\% = 171 \text{ mol m}^{-3}$), and finally in Eqs. (4) and (16) in case of 25°C ($T = 298^\circ\text{K}$) $\alpha R_s T = 7.57 \text{ bar m}\%^{-1}$.

3.2. Mathematical method

The characteristics of the transmembrane process with R^2 were calculated from Eqs. (19)–(20) and Eq. (26) by linear regression (EXCEL). The regression line is:

$$y = ax + b \quad (27)$$

where a and b are the regression parameters.

The student test was used, t_{n-2} [19], where n is the number of measurements and $f = n-2$ is the variability. The estimated values of the parameters are \hat{a} and \hat{b} and that of Eq. (27) is \hat{y} (point estimation). The 0-hypothesis is $H_0: \hat{y} = y$, which is accepted at a 95% probability level, if $t_{n-2} > t_{f,0.95}$, where $t_{f,0.95}$ is the value belonging to a 95% (one-side) probability level (student's test table).

The standard errors of parameters, s_a and s_b , were calculated by the known formulas referred to in a parallel work in this volume [20]. Finally a confidence interval (both sides) of 95% is determined for $a[\hat{a} - t_{f,0.975} s_a; \hat{a} + t_{f,0.975} s_a]$ and for b as well and $P = 1-0.95 = 0.05$ is the significance level (interval estimation) [20].

3.3. Pressure dependence

The pressure dependence of the water flux has been calculated from Eq. (19) by Eq. (27) with parameters: $y = J_w$, $x = \Delta p$, $a = K_w(1-7.57qC_f^*)$ and $b = -7.57K_w C_f^*$. The values measured for the parameters of the curve with a given concentration ($C_f = 3.5 \text{ m}\%$) are as follows: $\hat{a} = 1.28 \text{ Lm}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$, $\hat{b} = -28.9 \text{ Lm}^{-2} \text{ h}^{-1}$, furthermore, the fitting parameters are $R^2 = 0.9948$, $n = 6$, $f = 4$, from this, $t_f = 28.2$. The critical value is $t_{4,0.95} = 2.13$ (one-side probability), thus the 0-hypothesis is accepted (Fig. 4). The standard errors of parameters are: $s_a = 0.02 \text{ Lm}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ (1.6%) and $s_b = 0.3 \text{ Lm}^{-2} \text{ h}^{-1}$ (1%).

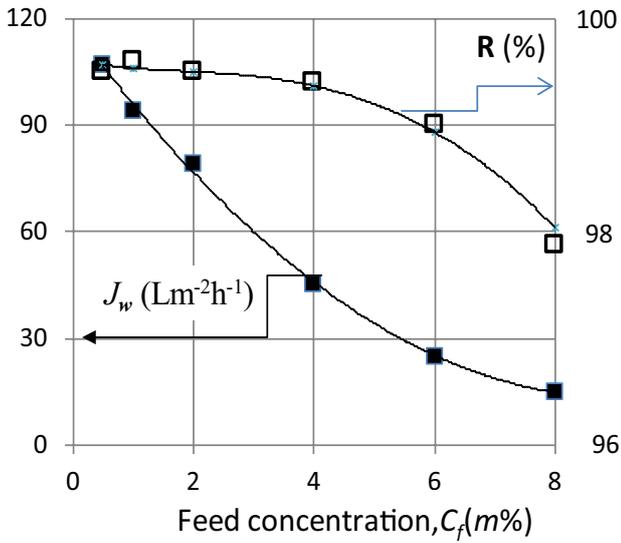


Fig. 3. The permeate (water) flux, J_w , and salt rejection, R , as a function of the feed concentration at $\Delta p = 67$ transmembrane pressure difference on the feed side [5,12].

The osmotic pressure associated with C_f concentration (3.5 m%) in the case of $J_w = 0$, the transmembrane pressure difference from Eq. (27) is:

$$\Delta p(J_w = 0) = \Delta \hat{\pi} = -\frac{b}{a} \quad (28)$$

the estimated value of which is 22.5 bar, the standard error (1.6% + 1% = 2.6%, the relative errors are summarised) is 0.58 bar and the confidence interval of 95% is (20.9; 24.1 bar).

3.4. Concentration dependence

The concentration dependence of the water flux has been determined from Eq. (20) by Eq. (27), with $a = K_w(1 + 7.37q\Delta p)$ and $b = K_w\Delta p$. The estimated values are $\hat{a} = -21.2 \text{ Lm}^{-2} \text{ h}^{-1} \text{ m\%}^{-1}$ and $\hat{b} = 121 \text{ Lm}^{-2} \text{ h}^{-1}$, furthermore, $R^2 = 0.9859$, $n = 6$, $f = 4$; from this $t_f = 16.7 > 2.78$, that is, the 0-hypothesis is accepted (Fig. 5). The standard errors of the fitting parameters will be $s_a = 0.9 \text{ Lm}^{-2} \text{ h}^{-1} \text{ m\%}^{-1}$ (4%) and $s_b = 4 \text{ Lm}^{-2} \text{ h}^{-1}$ (3.5%).

The estimated value of K_w from b ($\Delta p = 67$ bar, [4,12]) is $\hat{K}_w = 1.81 \text{ Lm}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ ($0.00181 \text{ mh}^{-1} \text{ bar}^{-1}$) and its standard error in Eq. (27) is $s_k = 0.06 \text{ Lm}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$, furthermore, the confidence interval is 95% [1.64; 1.98 $\text{Lm}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$]. The estimated value of q is $\hat{q} = 0.0021 \text{ bar}^{-1}$, $s_q = 0.00016 \text{ bar}^{-1}$ and the confidence interval is [0.0018; 0.0026 bar^{-1}]. The estimated back diffusion coefficient from Eq. (21) is $\hat{K}_s = 0.85 \text{ Lm}^{-2} \text{ h}^{-1}$.

3.5. Salt rejection

The salt rejection parameters were calculated from Eq. (26) using the Eq. (27) regression line (Fig. 6) with $y = 100 - R$, $x = 100/J_w$ and $a = K_s$. The estimated values of salt mass transfer coefficient K_s is $\hat{K}_s = 0.45 \text{ Lm}^{-2} \text{ h}^{-1}$ (0.00045 mh^{-1}),

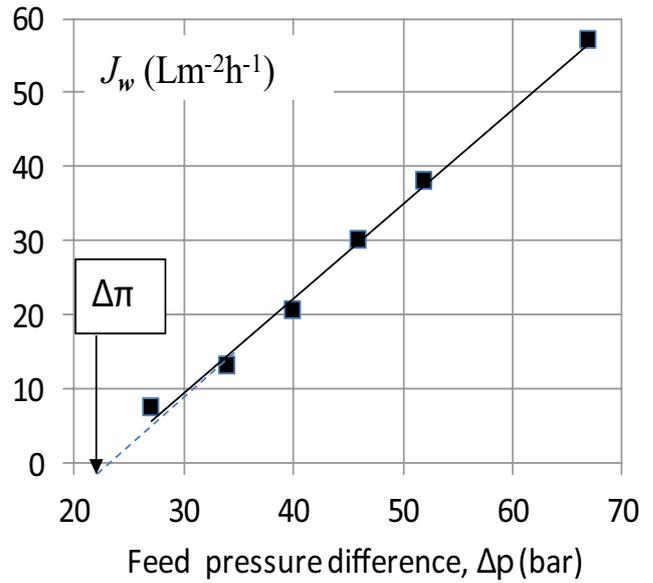


Fig. 4. The permeate (water) flux, J_w , as the function of the feed (transmembrane) pressure difference, at a $C_f = 3.5$ m% feed-side salt concentration [5,12].

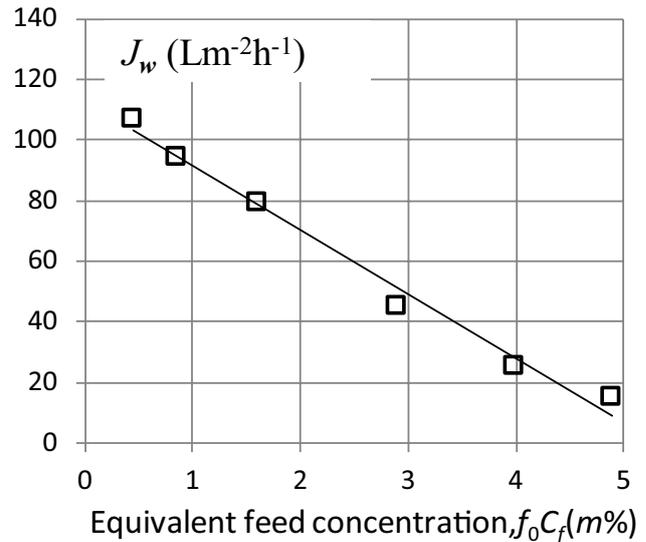


Fig. 5. Permeate (water) flux, J_w , as the function of the equivalent feed concentration ($C_f^* = f_0 C_f$) for $\Delta p = 67$ bar feed-side transmembrane pressure difference [5,12].

$R^2 = 0.9482$, $f = 3$, and from this $t_f = 7.4 > 2.35$ therefore the 0-hypothesis is accepted. The standard error of the estimation is $s_{ks} = 0.036 \text{ Lm}^{-2} \text{ h}^{-1}$ and the confidence interval of 95% is [72; 92 mh^{-1}] as well.

4. Discussion

In the case of reverse osmosis, the permeate flux depends on the Δp transmembrane pressure difference and the C_f feed concentration. The pressure dependence is linear, and the transmembrane pressure associated with $J_w = 0$

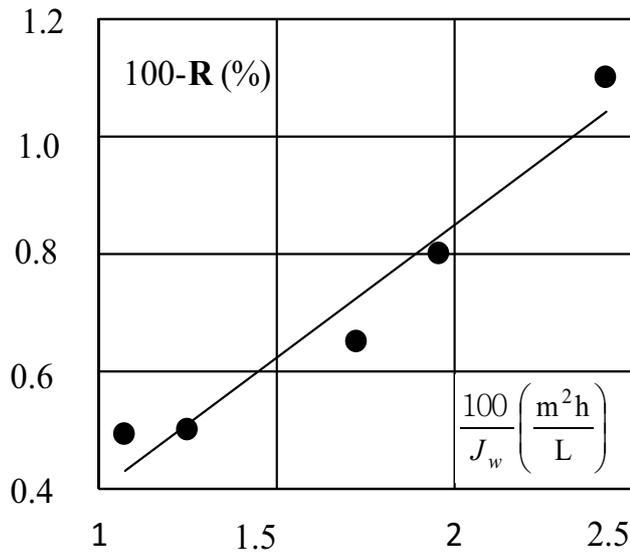


Fig. 6. The function of the salt rejection conjugate as a function of the inverse permeate flux [5,12].

is equal to the osmotic pressure, $\Delta p = \Delta \pi$. If $\Delta p < \Delta \pi$, it is considered osmosis, if $\Delta p > \Delta \pi$, it is reverse osmosis.

In the case of reverse osmosis, the osmotic pressure, and through this the permeate flux, according to the van't Hoff law, is a linear function of the concentration, C_p , while measurement results show a higher degree of correlation. According to our model, this dependence is partly because the osmotic pressure of the real solution differs from the van't Hoff law, and partly because of the polarisation within the membrane.

Instead of the empirical formulas normally used in literature, the deviation from the van't Hoff law was considered by using the osmotic coefficient f_0 . The product of the concentration and the osmotic coefficient, as equivalent concentration, C_f^* can be defined, and it means the concentration value that would produce identical osmotic pressure in the case of an ideal solution.

The permeate flux as the function of equivalent concentration can be approximated by a linear equation. In the case of an empirical model the permeate flux depends separately on transmembrane pressure (Δp) and salt concentration (C_p), while in a developed one it depends on the product of both $\Delta p C_f$ too. This latter factor is the consequence of the polarization, which can be altered somewhat by changing the system characteristics and parameters with the back mass transfer constant through the Sherwood number.

5. Conclusion

During the process, the concentrate becomes thicker along the membrane length l . Increasing the osmotic pressure raises the driving force and thus the permeate flux decreases, therefore a maximum (effective) membrane length $\Delta p = \Delta \pi(l_{\max})$ can be defined, above which $J_w = 0$. Its value depends on the flow rate of the solution, which, in turn, is dependent on the trans-membrane pressure applied (Bernoulli law). If l is increased, the permeate quantity will

grow until this value, and better yields are achieved. In the case of $l < l_{\max}$, the flux determined experimentally is an average flux characteristic of the system, and accordingly, the mass transfer coefficients K_w and K_s will be average values, which depend on the system characteristics.

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Symbols

C	—	Concentration, mol m ⁻³ , m%
D	—	Diffusion coefficient, m ² h ⁻¹
J_w	—	Water flux, Lm ⁻² h ⁻¹ , m h ⁻¹
J_s	—	Salt flux, m% m ⁻² h ⁻¹
K_w	—	Water transfer coefficient, Lm ⁻² h ⁻¹ bar ⁻¹ , m h ⁻¹ bar ⁻¹
K_s	—	Salt transfer coefficient, Lm ⁻² h ⁻¹ , m h ⁻¹
K_a	—	Dissociation constant
R	—	Salt rejection factor, %
Re	—	Reynolds number, —
R_g	—	Gas constant, 8.314 J mol ⁻¹ K ⁻¹
Sc	—	Schmidt number, —
Sh	—	Sherwood number, —
T	—	Temperature, K, °C
P	—	Significance level, %
R^2	—	Correlation coefficient
a	—	Parameter, —
b	—	Parameter, —
d	—	Characteristic size, diameter, m
f	—	Variability
f_0	—	Osmotic coefficient
l	—	Length, m
n	—	Number of measurements
p	—	Pressure, Pa, bar
s	—	Standard error
q	—	Rate, —
t_f	—	Student test
v	—	Flow rate, m s ⁻¹

Greek

ν	—	Dissociation number
π	—	Osmotic pressure, Pa, bar
δ	—	Thickness, m
β	—	Concentration polarization, —
ϵ_0	—	Dielectric constant
η	—	Viscosity, m ² s ⁻¹
ρ	—	Density, kg m ⁻³

Indexes

f	—	Feed
p	—	Permeate

- ^s — Salt
^w — Water
 – — Equivalent value
 – — Back
 ^ — Estimated value

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