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Temperature and concentration polarization in membrane distillation of aqueous salt solutions

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Abstract

In this paper we have studied water transport in membrane distillation using a flat PTFE membrane. Experiments have been carried out with water and aqueous solutions of NaCl as feed. The effects of temperature and concentration polarization on the reduction of vapour pressure differences across the membrane with regard to the vapour pressure differences corresponding to the bulk phases which are separated by the membrane, are evaluated. A coefficient which measures this reduction has been introduced. This coefficient and the temperature polarization coefficient coincide when water is used as feed, but they are more and more different when the salt concentration of feed increases.

The measured flux results and the calculated polarization results are discussed for different temperatures, recirculation rates and solution concentrations. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Membrane distillation; Temperature and concentration polarization; Hydrophobic membrane; Vapour permeation

1. Introduction

Membrane distillation (MD) is an evaporative process using hydrophobic microporous membranes. In the MD process studied here, we have a salt solution at an elevated temperature on one side of the membrane and pure water on the other side. The temperature and concentration differences through the membrane result in a vapour pressure difference, so that at the feed/membrane interface water will evaporate. The vapour flows from the warm to the cold side where it condenses. The driving force for the mass transport is a difference of vapour pressure which results from the

temperature and composition of solutions in the layers adjoining the membrane.

The phenomenon of the temperature polarization [1–3] causes the temperatures at the membrane surfaces to differ from the bulk temperatures measured in the feed and in the distillate. This phenomenon is present even when the feed is water and causes an important loss in the driving force for transport with regard to the imposed force. When feed concentration rises, we have to add concentration polarization to temperature polarization which also reduces the imposed driving force and so the mass flux [4–6].

In the MD literature the temperature polarization is measured with the τ coefficient:

$$\tau = \frac{T_{m1} - T_{m2}}{T_{b1} - T_{b2}}, \quad (1)$$

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where T_{m1} and T_{m2} are the temperatures at the hot and cold membrane surfaces, respectively, and T_{b1} and T_{b2} are the temperatures in the feed and permeate bulk solutions, respectively. To quantify the concentration polarization the ζ coefficient is used

$$\zeta = \frac{c_{m1}}{c_{b1}}, \quad (2)$$

where c_{m1} is the salt concentration at the hot membrane surface and c_{b1} is the salt concentration in the feed bulk solution.

In this work and in order to measure the reduction in the effective driving force for the mass transport with regard to the driving force imposed we introduce the vapour pressure polarization coefficient,

$$f = \frac{p_{m1} - p_{m2}}{p_{b1} - p_{b2}}, \quad (3)$$

where p is the vapour pressure and the subscripts have the same meaning as in Eq. (1).

When MD of water or diluted solutions is studied, and values of $(T_{b1} - T_{b2})$ less than 10°C are employed, the following equation is often used in the literature of MD:

$$\frac{(p_{m1} - p_{m2})}{(T_{m1} - T_{m2})} = \frac{(p_{b1} - p_{b2})}{(T_{b1} - T_{b2})}. \quad (4)$$

In this case, τ is considered as an estimation of the reduction of the driving force imposed. Moreover the commonly accepted relation for the mass flux

$$J = C(p_{m1} - p_{m2}), \quad (5)$$

that in accordance with definition (3) can be expressed as

$$J = fC(p_{b1} - p_{b2}) \quad (6)$$

can also be written as

$$J = \tau C(p_{b1} - p_{b2}), \quad (7)$$

where C is the membrane distillation coefficient.

The use of the linearization of the exponential Antoine relation describing the partial vapour pressure as function of the temperature as given in Eqs. (4) and (7) is physically and mathematical not correct. We want to show that in water distillation and at limited temperature differences this linearization is a good approximation. As a consequence f and τ are approximately similar. In this case τ can be considered an

approximated measure of the reduction of driving force imposed.

As distillation of more and more concentrated solutions is considered, the mentioned approximations are more and more incorrect. In the present work we show as, in fact, τ and f are more and more different and it is necessary to evaluate f if a measure of the driving force imposed is looked for. The τ coefficient does not measure the reduction of driving force although concentration polarization is not important.

In this work we have studied the temperature and concentration polarization and have explained the corresponding reduction in the driving force. This analysis is carried out for different values of the recirculation rates, temperatures and concentrations of the solutions studied. We have shown that the difference between τ and f is higher when concentration increases. This difference has been explained as a consequence of temperature and concentration dependence of vapour pressure.

2. Experimental

Experimental tests were performed using a flat PTFE membrane manufactured by Gelman Instrument as TF200 (80% void fraction, 60 μm thickness, 0.2 μm nominal pore size).

The experimental set-up used has been described elsewhere [7]. The membrane module was a flat sheet designed with nine feed channels and nine permeate channels each of 55.0 mm long, 7.0 mm wide and 0.45 mm high. The effective membrane area for the transport was $33.7 \times 10^{-4} \text{ m}^2$. In all experimental runs the membrane was maintained in a horizontal position. The feed solution was pretreated in a thermostatic bath and then pumped onto the membrane low surface. Water was likewise preheated (at a lower temperature than the feed solution) in another thermostatic bath and then pumped onto the upper membrane surface. The recirculation of the fluids on both sides of the membrane was in countercurrent directions.

The temperatures of the bulk liquid phases are measured at the hot entrance (T_{b1-in}), the cold entrance (T_{b2-in}), the hot exit (T_{b1-out}) and the cold exit (T_{b2-out}), of the membrane cell. In the following study the

average values

$$T_{b1} = \frac{T_{b1-in} + T_{b1-out}}{2},$$

$$T_{b2} = \frac{T_{b2-in} + T_{b2-out}}{2}$$

will be considered as bulk temperatures of distilland feed and distillate permeate, respectively.

These temperatures will be different from the temperatures at the hot and cold membrane sides, T_{m1} and T_{m2} , respectively.

In this experimental set-up the permeate continuously flows out of the distillate reservoir, and the corresponding distillate flux was measured by collecting the liquid flowing out of the cold chamber. In all experiments the final composition of the distillate liquid was measured, and a total salt rejection was always observed.

In the present work experiments were conducted with feed sodium chloride solutions of concentrations 0, 0.55, 1.15 and 1.67 molar. In all cases, from the beginning of the run, pure water was recirculated in the cold semicell. Likewise in all cases, the recirculation flow rates on both membrane surfaces were the same.

Different experiments were carried out applying the recirculation rates of 7, 11 and 15 cm³/s (that is a linear velocity down the channel of about 25, 39 and 53 cm/s, respectively). The transmembrane pressure gradient was minimum for all the experiments while the absolute pressure in the membrane varied from 1.05×10^5 Pa for the lower recirculation rate up to 1.3×10^5 Pa for the higher recirculation rate.

Different experiments were carried out for fixed temperatures in the membrane module. The feed temperature T_{b1} varied from 20°C to 50°C at increments of about 7°C, and the cooling water temperature T_{b2} varied from 10°C to 40°C, also at increments of about 7°C. For all these experiments, values of $(T_{b1-in} - T_{b1-out})$ and $(T_{b2-out} - T_{b2-in})$ lower than 1.5°C were obtained. The temperature difference $(T_{b1} - T_{b2})$ was 10°C in all experiments. A water flux from the hot to the cold phase was always observed.

3. Theory

The mass flux through the membrane is driven by a vapour pressure difference resulting from the imposed

temperature and salt concentration differences. This flux may be explained in the framework of different transport models [1,2] (the Knudsen model, the Poiseuille model, and the diffusive model). All the models suggest the relationship (5) between the mass flux J and the driving force $(p_{m1} - p_{m2})$.

Simultaneously to mass transport, heat transport occurs in MD [1]. This heat Q consists of the heat conducted through the membrane and the latent heat accompanying vapour flux

$$Q = (k_m/\delta)(T_{m1} - T_{m2}) + J\lambda, \quad (8)$$

where δ is the membrane thickness and λ is the latent heat of vaporization. In Eq. (8) k_m is the thermal conductivity of the porous membrane that can be calculated as

$$k_m = \epsilon k_g + (1 - \epsilon)k_s$$

with k_g and k_s being the thermal conductivities of the gas and solid phases [7], respectively, and ϵ is the membrane porosity.

Due to temperature polarization, the temperatures T_{m1} and T_{m2} may vary markedly from the measurable bulk temperatures T_{b1} and T_{b2} . In order to calculate the temperature polarization coefficient τ we consider the heat transfer to and from the membrane surface, that for steady conditions are equal to the amount of heat transferred inside the membrane

$$Q = h_1(T_{b1} - T_{m1}) = h_2(T_{m2} - T_{b2}), \quad (9)$$

where h_1 and h_2 are the film heat transfer coefficients.

From Eqs. (8) and (9) we can calculate

$$T_{m1} = T_{b1} - (T_{b1} - T_{b2}) \frac{1/h_1}{1/(h_c + h_v) + 1/h_1 + 1/h_2}, \quad (10)$$

and

$$T_{m2} = T_{b2} + (T_{b1} - T_{b2}) \frac{1/h_2}{1/(h_c + h_v) + 1/h_1 + 1/h_2}, \quad (11)$$

where h_c is the condition coefficient,

$$h_c = \frac{k_m}{\delta}, \quad (12)$$

h_v is the vapour heat transfer coefficient,

$$h_v = \frac{J\lambda}{T_{m1} - T_{m2}}, \quad (13)$$

and the feed and permeate film heat transfer coefficients can be estimated by means of the dimensionless Nusselt number, Nu :

$$h_i = \frac{Nu_i k_i}{d_h}, \quad i = 1, 2, \quad (14)$$

where 1 and 2 refer to feed and permeate, respectively. In Eq. (14) k is the fluid thermal conductivity and d_h is the hydraulic diameter.

In MD the Nusselt number is almost always estimated from empirical correlations. In our experiments we have worked with a flat membrane module and laminar flow (always $Re < 800$). In a previous work [7] we determined, in experiments with water, the h coefficients following an experimental method given by Schofield et al. [1] which has also been used by other authors [8,9]. The more satisfactory agreement between those experimental results and different empirical correlations tested [8,10] was obtained when the Graetz-Lévêque equation:

$$Nu = 1.86 \left(Re Pr \frac{d_h}{L} \right)^{0.33} \quad (15)$$

was used. We have to point out that the disagreement with all the other tested relationships found in bibliography was very important. So this equation has been used for predicting the h_1 and h_2 values in the experiments with different concentrations. In Eq. (15) L is the channel length, and Re and Pr are the Reynolds and Prandtl numbers

$$Re = \frac{d_h v \rho}{\mu}, \quad Pr = \frac{c_p \mu}{k}, \quad (16)$$

where v , ρ , μ and k are the average velocity, the density, the viscosity and the thermal conductivity of the liquid, respectively.

The evaluation of the coefficients h_c , h_v , h_1 and h_2 from the last equations allows us to calculate T_{m1} and T_{m2} from Eqs. (10) and (11). But, as $(T_{m1} - T_{m2})$ is in Eq. (13) and iterative solution is required. Initial values of T_{m1} and T_{m2} were assumed. Then, using the measured J , the values of h coefficients are calculated from (Eqs. (12)–(15)) at the average temperatures $(T_{b1} + T_{m1})/2$ and $(T_{b2} + T_{m2})/2$ in the thermal boundary layers. Then by using Eqs. (10) and (11) the temperatures T_{m1} and T_{m2} are obtained and compared with the initially assumed. The calculation was

repeated until the assumed values for T_{m1} and T_{m2} agreed with the calculated ones to the degree of relative error of 0.1%. In this way we know T_{m1} and T_{m2} for each experiment.

When pure water is used as a feed liquid, the permeation flux is affected simply by the thermal boundary layers as a function of the stirring speed and temperatures. A reduction of the permeate flux occurred with the increase of salt concentration in the feed solution. This results from the decrease of the vapour pressure being in equilibrium with the feed. But the effect of solute in the feed is also to alter the fluid dynamics and to influence the heat transfer (and therefore T_{m1} , T_{m2} and so J) through thermal conductivity and heat capacity. Finally, concentration polarization is formed by the water (and no solute) permeation through the membrane, and so [4] the solute concentration c_{m1} at the feed solution/membrane interface becomes higher than that in the bulk solution, c_{b1} :

$$c_{m1} = c_{b1} \exp(J/\rho K), \quad (17)$$

where K is the solute mass transfer coefficient. In our experiments we will evaluate the K coefficient using the mass transfer analogy of the Graetz-Lévêque equation:

$$Sh = 1.86 \left(Re Sc \frac{d_h}{L} \right)^{0.33}, \quad (18)$$

where Sh and Sc are the Sherwood number and the Schmidt number

$$Sh = \frac{K d_h}{D}, \quad Sc = \frac{\mu}{\rho D} \quad (19)$$

with D being the diffusion coefficient of the solute.

These last relations allow us to evaluate c_{m1} . So we know T_{m1} , T_{m2} and c_{m1} in each experiment and we can evaluate τ and f .

4. Results and discussion

In Figs. 1–3 the distillate fluxes for the three recirculation rates studied are displayed as a function of the imposed temperatures when different salt concentrations are used. These flux results are the average value of at least three experiments obtained when temperatures T_{b1-in} , T_{b1-out} , T_{b2-in} and T_{b2-out} were maintained

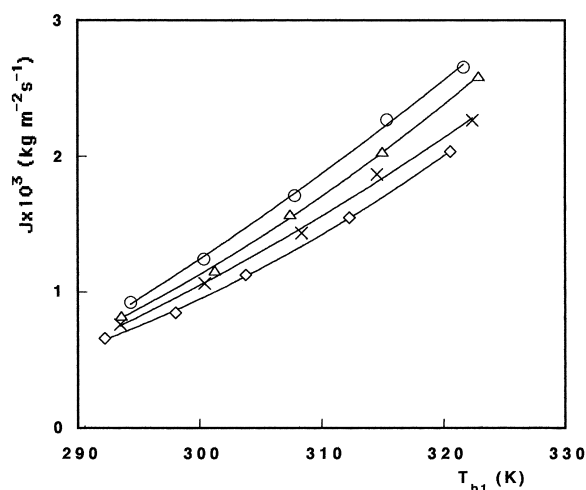


Fig. 1. Water flux vs. feed temperature for a recirculation rate of $7 \times 10^{-6} \text{ m}^3/\text{s}$. The different symbols correspond to different hot feeds: (○) water; (△) sol. 0.55 mol; (×) sol. 1.15 mol; (◇) sol. 1.67 mol. In the experiment $\Delta T_b = 10^\circ\text{C}$.

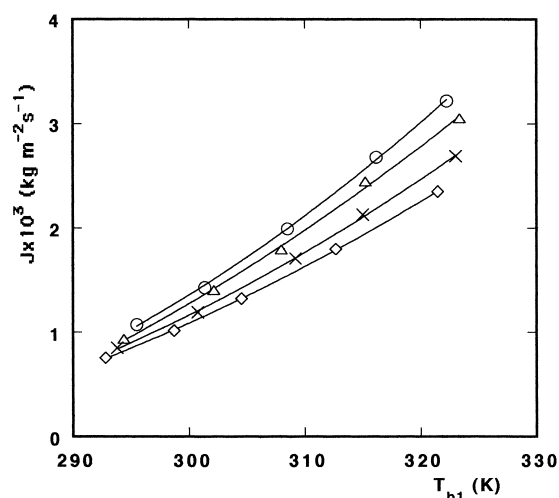


Fig. 3. Water flux vs. feed temperature for a recirculation rate of $15 \times 10^{-6} \text{ m}^3/\text{s}$. In the experiment $\Delta T_b = 10^\circ\text{C}$. Symbols as in Fig. 1.

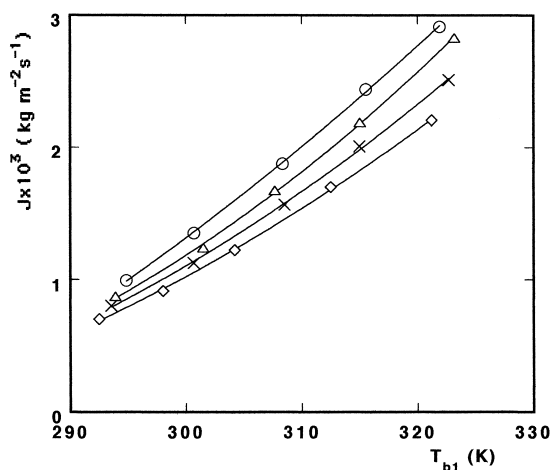


Fig. 2. Water flux vs. feed temperature for a recirculation rate of $11 \times 10^{-6} \text{ m}^3/\text{s}$. In the experiment $\Delta T_b = 10^\circ\text{C}$. Symbols as in Fig. 1.

constant within $\pm 0.1^\circ\text{C}$, and the recirculation rates and concentrations were also constant with oscillations lower than 1%. So, the maximum error in the measurements of fluxes was $\pm 0.00003 \text{ kg/m}^2\text{s}$.

In the model described above C is an adjustable parameter. Thus, in Figs. 1–3 the filled lines correspond to the flux values predicted by the model for $C = 14.5 \times 10^{-7} \text{ kg/m}^2\text{s}\cdot\text{Pa}$. This C value coincides with that estimated in an earlier work [11] following

the mentioned experimental method given by Schofield et al. [1].

We have calculated the corresponding τ coefficients as indicated above after evaluating T_{m1} and T_{m2} from Eqs. (10) and (11) and using the iterative method described. Representative results are shown in Figs. 4–7, and we can conclude from them

1. The polarization temperature is an important factor affecting the mass flux as τ values range between 0.4 and 0.6.
2. In the temperature interval studied (T_{b1} varied from 20°C to 50°C) τ varied significantly, decreasing as the temperature increases. This is due to the exponential rise of the vapour pressure curve which makes the permeate flux increase substantially as the temperature rises. These larger mass fluxes involve more important heat fluxes through the liquid phases, increasing the temperature gradient in the boundary liquid layers and so the temperature polarization. For this reason J does not increase with the temperature as fast as the vapour pressure curve.
3. In the recirculation rate range studied (this varied from 7 to $15 \text{ cm}^3/\text{s}$) the change of τ with the recirculation rate is important. The reason is that when the values of polarization temperature are important an increase of the recirculation rate enhances the heat transport and so τ and J increase.

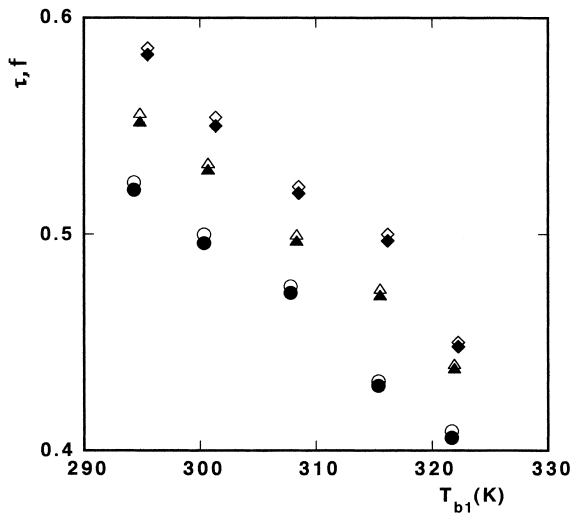


Fig. 4. Temperature polarization and polarization coefficients vs. temperature when the hot feed is water. The different symbols correspond to different recirculation rates. (\circ : τ , \bullet : f) $7 \times 10^{-6} \text{ m}^3/\text{s}$; (Δ : τ , \blacktriangle : f) $11 \times 10^{-6} \text{ m}^3/\text{s}$; (\diamond : τ , \blacklozenge : f) $15 \times 10^{-6} \text{ m}^3/\text{s}$.

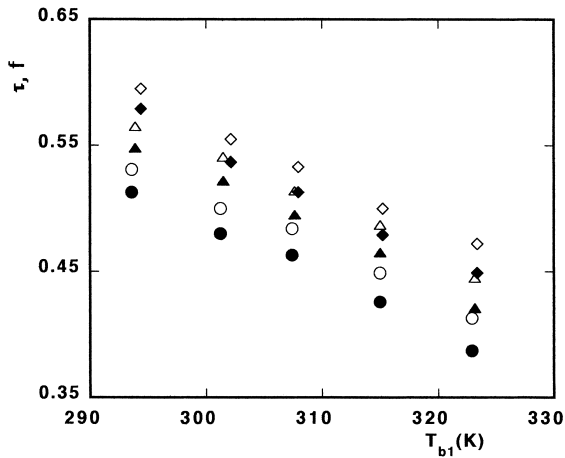


Fig. 5. Temperature polarization and polarization coefficients vs. temperature when the hot feed is a solution 0.55 mol. Symbols as in Fig. 4.

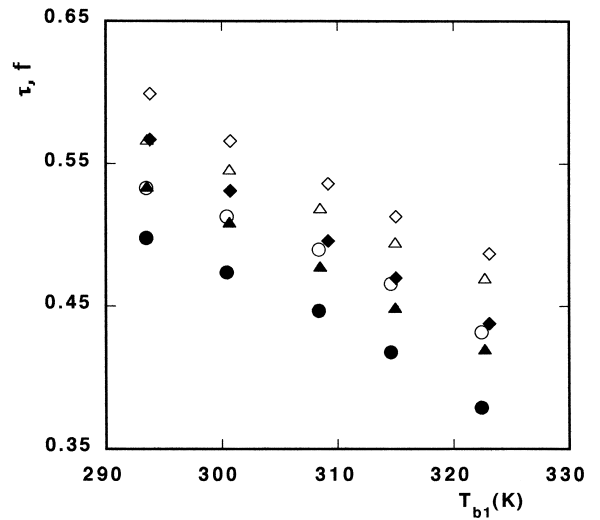


Fig. 6. Temperature polarization and polarization coefficients vs. temperature when the hot feed is a solution 1.67 mol. Symbols as in Fig. 4.

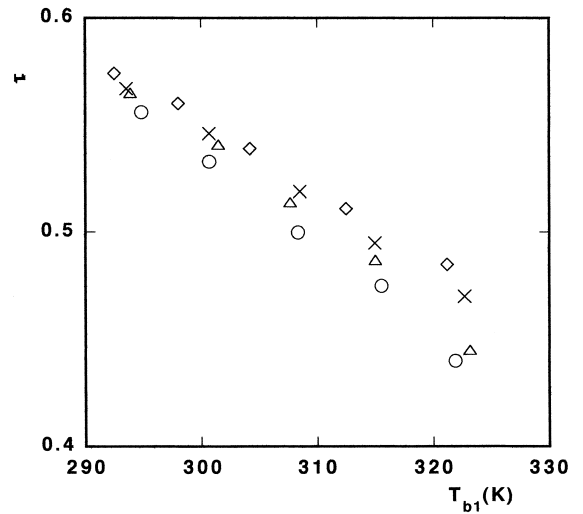


Fig. 7. Temperature polarization vs. temperature for a recirculation rate of $11 \times 10^{-6} \text{ m}^3/\text{s}$. The different symbols correspond to different hot feeds: (\circ) water; (Δ) sol. 0.55 mol; (\times) sol. 1.15 mol; (\diamond) sol. 1.67 mol.

4. In the concentration range studied (c_{b1} varied from 0 to 1.67 mol) we observed an important decrease of J as c_{b1} increases as could be expected due to the fact that the vapour pressure decreases as the concentration increases. At the same time, τ

increases, as we indicate in the representative plot of Fig. 7. This increase of τ with c is the result of two opposed contributions: firstly, h_1 decreases slightly with the concentration (Eq. (15)), and secondly the indicated lower mass fluxes involve

lower heat fluxes through the liquid phases decreasing the temperature gradient in the boundary liquid layers. This last effect is higher than the first and also explains that the increase of τ with concentration is higher when the temperature increases.

As indicated previously after evaluating τ we have evaluated the concentration polarization c_{m1}/c_{b1} from the Eqs. (17) and (18). It was calculated that the membrane wall concentration was as far as 4% higher than the bulk concentration ($c_{m1}/c_{b1}=1.04$). According to Robinson and Stokes [12] this concentration increase means a decrease of the vapour pressure to about 0.2%. As a consequence of the concentration polarization leads to a negligible reduction in the permeate flux in the studied cases. This is an important result as concentration polarization is a major cause of flux reduction in other membrane processes such as ultrafiltration. For the mass transfer coefficients used, higher fluxes will be necessary in order to have an important concentration polarization in MD.

After evaluating τ and ζ we calculated f using the bibliography data of vapour pressure in order to evaluate p_{m1} , p_{m2} , p_{b1} and p_{b2} as a function of temperature and concentration. The obtained results are shown in Figs. 4–6. From these results we can conclude:

1. When water is used as feed, the values of τ and f are similar. In fact, they differ less than 0.6% of the τ values. In this case the value of τ can be considered a good measure of the reduction of driving force imposed by effect of temperature polarization.
2. When $c_{b1} \neq 0$, τ and f differ. This difference is higher when c_{b1} increases, even when concentration polarization effects in pressure vapour reduction are negligible. Now Eq. (4) is not true and the vapour pressure dependence with concentration must be taken into account in order to explain the f values and the important reduction in the flux with regard to the one obtained with water even when τ increases a little with the concentration.

As a general conclusion, the drop of the flux with rising concentrations has several causes: the temperature polarization, the decrease of the vapour pressure and in a much lower scale the concentration polarization. The relative importance of them is shown in the

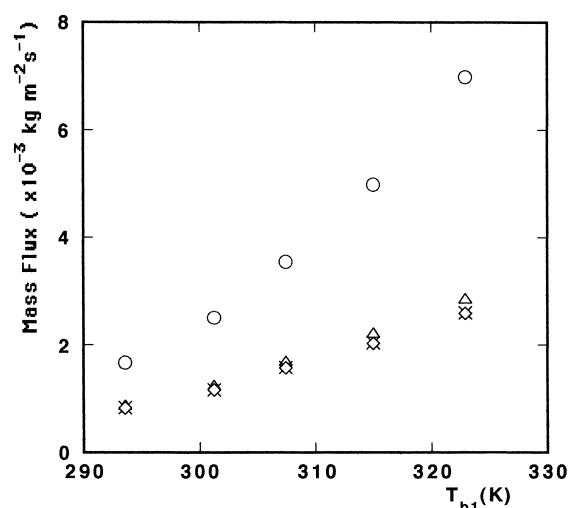


Fig. 8. Mass flux through the membrane vs. feed temperature for a recirculation rate of $7 \times 10^{-6} \text{ m}^3/\text{s}$ and a hot feed of 0.55 mol. J^{id} , J^t and J^v are defined in (Eqs. (21)–(23)): (\circ) J^{id} ; (\triangle) J^t ; (\times) J^v ; (\diamond) J .

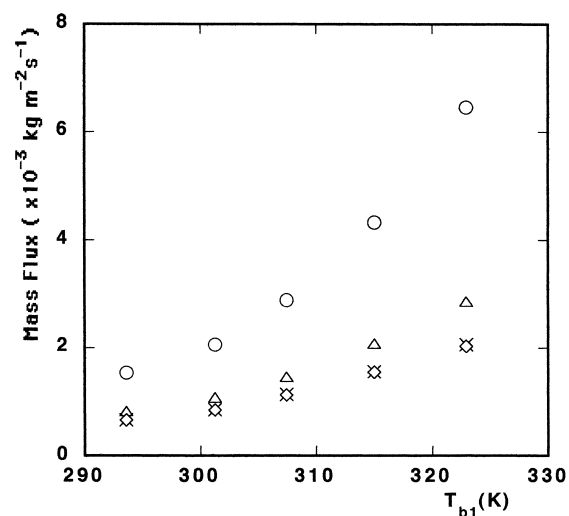


Fig. 9. Mass flux through the membrane vs. feed temperature for a recirculation rate of $7 \times 10^{-6} \text{ m}^3/\text{s}$ and a hot feed of 1.67 molar. Symbols as in Fig. 8.

representative Figs. 8 and 9. Taking into account the dependence of the vapour pressure with temperature and concentration.

$$p = p(T, c) \quad (20)$$

we have defined

$$J^{\text{id}} = C(p_{\text{b1}}^* - p_{\text{b2}}^*) = C[p(T_{\text{b1}}, 0) - p(T_{\text{b2}}, 0)], \quad (21)$$

$$J^{\text{t}} = C(p_{\text{m1}}^* - p_{\text{m2}}^*) = C[p(T_{\text{m1}}, 0) - p(T_{\text{m2}}, 0)], \quad (22)$$

$$J^{\text{v}} = C[p(T_{\text{m1}}, c_{\text{b1}}) - p(T_{\text{m2}}, 0)], \quad (23)$$

where p_{b1}^* , p_{b2}^* , p_{m1}^* and p_{m2}^* are the vapour pressure of pure water at the temperatures T_{b1} , T_{b2} , T_{m1} and T_{m2} , respectively. The results obtained for J^{id} , J^{t} and J^{v} are shown in Figs. 8 and 9 for some representative experimental conditions. The J^{id} values represent the ideal situation which temperature and concentration polarization does not exist and where the reduction of the vapour pressure that occurs when $c \neq 0$ has not been taken into account. The differences between the J^{id} and J^{t} values represent the effect of temperature polarization. The differences between the J^{t} and J^{v} values represent the flux reduction due to the decrease of the vapour pressure of the solution with respect to the pure water. Finally the differences between J^{v} and J values represent the effect of concentration polarization.

These results show the meaning of τ for concentrated solutions not having the meaning of reduction of driving force imposed, which is reserved to f . From these last results we conclude that the temperature boundary layers exert the greatest influence in the mass transport. The permeate flux might be substantially higher if we had no temperature polarization. As the concentration increases the effect of vapour pressure reduction becomes more important. In all cases studied the effect of concentration polarization is negligible.

5. Conclusions

Polarization layers formed on either side of PTFE membrane reduce water permeation in membrane distillation. In the studied experiments only temperature polarization becomes important while concentration polarization has an insignificant influence.

The introduced f coefficient allows us to evaluate effective vapour pressure difference for the transport with regard to the imposed vapour pressure difference. Temperature polarization and the change of vapour

pressure with concentration allows us to explain the f values which mean an important reduction, between 40% and 65%, in the imposed force, depending on the experiment. This reduction is higher when temperature increases, when recirculation rate decreases and when concentration increases.

6. Nomenclature

c	molar concentration (mol/l)
c_p	specific heat (J/kg K)
C	membrane distillation coefficient (kg/m ² s Pa)
d_h	hydraulic diameter of channel (m)
D	diffusion coefficient of solute (m ² /s)
f	polarization coefficient (dimensionless)
h	heat transfer coefficient (W/m ² K)
J	mass flux through the membrane (kg/m ² s).
J^{id}	as defined in Eq. (21) (kg/m ² s)
J^{t}	as defined in Eq. (22) (kg m ² s)
J^{v}	as defined in Eq. (23) (kg m ² s)
k	thermal conductivity of the liquid (W/m K)
k_m	thermal conductivity of the membrane (W/m K)
k_g	thermal conductivity of the air (W/m K)
k_s	thermal conductivity of the solid phase of the membrane (W/m K)
K	solute mass transfer coefficient (m/s)
L	channel length (m)
Nu	Nusselt number (dimensionless)
p	pressure of water vapour (Pa)
p^*	vapour pressure of pure water at the same temperature as the solution (Pa)
Pr	Prandtl number (dimensionless)
Q	heat flux (W/m ²)
Re	Reynolds number (dimensionless)
Sc	Schmidt number (dimensionless)
Sh	Sherwood number (dimensionless)
T	temperature (K)
v	linear velocity (m/s)

Greek symbols

δ	membrane thickness (m)
ϵ	porosity (dimensionless)
η	liquid viscosity (Pa/s)
λ	latent heat of vaporization (J/kg)
ρ	liquid density (kg/m ³)

τ	temperature polarization coefficient (dimensionless)
ζ	concentration polarization coefficient (dimensionless)

Subscripts

b	at the bulk phase
c	conduction
m	at the membrane surface
v	vapour
1	hot solution
2	cold water

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