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Nonisothermal Water Transport through Hydrophobic Membranes in a Stirred Cell

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ABSTRACT

This paper studies the transport of pure water through microporous hydrophobic membranes in a stirred cell when bathed by two phases at different temperatures. The dependence of the phenomena on the stirring rate and on the average temperature has been investigated. The influence of these operating conditions on the mass transfer rate is discussed while keeping in mind the theories of mass and heat transfer within the membrane and adjoining liquids. The concept of temperature polarization is introduced in the transport equations, and it is shown to be important in the interpretation of our experimental results.

INTRODUCTION

Membrane distillation (MD) is a process in which two liquids or solutions at different temperatures are separated by a porous membrane. The liquids or solutions must not wet the membrane or else the pores will be filled immediately as a result of capillary forces. This implies that nonwettable porous hydrophobic membranes must be used for aqueous solutions (1, 2).

When the liquid phases contain pure water and there is no temperature difference, the system is in equilibrium and no transport occurs. If the temperature of one of the two liquid phases is higher than that of the other, a temperature difference exists across the membrane, resulting in a vapor pressure difference. Thus, water will evaporate on the hot side;

the vapor flows from the hot side to the cold side where it condenses. In this way, water transport takes place across the membrane from the hot side to the cold side.

The advantages of membrane distillation are that the distillation process takes place at moderate temperature and that a relatively low temperature difference between the two liquids contacting the microporous hydrophobic membrane presents relatively high fluxes.

The need to supply heat to the evaporation surface of the membrane means that the temperature gradients must be in the liquid phase adjacent to the membrane. The same situation occurs in the condensation surface side. When the bulk phases on both sides of the membrane are stirred, the effective temperature difference between the two sides of the membrane is not the same as the temperature difference between the bulk solutions (Fig. 1). This loss of driving force, brought about by thermal gradients in the fluids bounding the membrane, is known as temperature polarization and was applied to studies of thermoosmosis (3–5) before being used in membrane distillation (6–8).

In the present work we measure water distillation through three different membranes under different operation conditions. Theories of mass and heat transfer within the membrane and adjoining fluids are developed in order to discuss the results obtained and to characterize the temperature polarization.

EXPERIMENTAL

Apparatus, Materials, and Method

Three commercial PTFE (polytetrafluoroethylene) membranes were used. These hydrophobic membranes are marketed by Teknokroma as Lida 200, Lida 500 and Lida 1000, with nominal pore sizes of 0.2, 0.50, and 1.0 μm and porosities of 0.80. These PTFE membranes have limited mechanical strength, and in practice they must be supported by nets generally made of polymer fibers. Therefore, they are composite membranes formed by a porous PTFE layer and a polypropylene support.

Pure water (twice distilled and deionized) was used in the experiments. The water was filtered through a Millipore filter of 0.45 μm nominal pore size before being introduced into the measuring apparatus.

All measurements were made with an experimental device similar to that employed in earlier thermoosmotic studies (3). It is a cell formed by two symmetric cylindrical semicells of 0.1 m length, separated by the membrane which is fixed in a methacrylate holder between two stainless-steel grids. The membrane surface area exposed to the flow was $q = 32.2$

× 10^{-4} m^2. The water in the chamber was stirred by a chain-drive magnetic-cell stirrer assembly. Both semicells are surrounded by concentric thermostatic chambers through which thermostatic fluids at different temperatures flow. Two thermocouples are sealed in the membrane holder so that the temperature of each semicell can be recorded and controlled. The cold chamber was connected to a glass tube placed horizontally and inserted in the top of the half-cell.

In the present work the experiments were performed with distilled water and three microporous hydrophobic membranes. A temperature difference was maintained between the two sides of the membrane, and the volume flux was measured by collecting and weighing the water flow through a glass tube placed horizontally in the cell. The experiments were carried out for different average temperatures in the system and for different stirring rates of the bulk phases. The purpose of this paper is to study the experimental situation and to evaluate some characteristic parameters of the membrane system by flux measurements.

THEORY

Simultaneous heat and mass transport characterize the membrane distillation process. The mass transport through MD membranes is driven by a vapor pressure difference resulting from the imposed temperature difference. This mass transport may be explained by the following mechanisms (6).

(a) When noncondensable gases are contained in the pores of the membrane (e.g., air) as a stagnant film, the molecular diffusion model applies (9):

$$J_D = (1/Y_{ln})(D\epsilon/\chi\delta)(M/RT)(P_1 - P_2) \qquad (1)$$

where D is the water diffusion coefficient, M is the water molecular weight, ϵ is the membrane porosity, χ is the tortuosity factor, δ is the membrane thickness, Y_{ln} is the mole fraction of air (log mean), T is the temperature, P_1 and P_2 are the water vapor pressures corresponding to the temperatures T_{m1} and T_{m2}, respectively, and R is the gas constant.

(b) In most cases, when the pore sizes and the mean free molecular paths in the membrane distillation process are of the same order of magnitude, the Knudsen diffusion model applies (10):

$$J_K = (2/3)(r\epsilon/\chi\delta)(8M/\pi RT)^{1/2}(P_1 - P_2) \qquad (2)$$

where r is the membrane pore radius.

The two models mentioned suggest the following equation by which the transfer may be described:

$$J = C(P_1 - P_2) \tag{3}$$

where C can be considered a combined mass transfer coefficient through the membrane, accounting for the mass transfer resistances of both molecular and Knudsen diffusion.

As vapor pressures within the membrane are not directly measurable, it is convenient to express Eq. (3) in terms of temperatures:

$$J = C|dP/dT|_{T_m}(T_{m1} - T_{m2}) \tag{4}$$

This equation is a good approximation for values of $T_{m1} - T_{m2}$ of less than 10°C. In Eq. (4), dP/dT can be evaluated from the Clausius–Clapeyron equation at the average membrane temperature T_m. Since, as opposed to the temperatures T_{b1} and T_{b2}, the temperatures T_{m1} and T_{m2} are difficult to measure, $T_{b1} - T_{b2}$ is, as a rule, inserted in the above equation. In order to do this, we must introduce the heat transfer coefficients (h_1, h_2) in the liquid films near the membrane, the latent heat transfer (λ) accompanying vapor flux, and the heat transfer by conduction (k_m) across the membrane. In this way, for the stationary thermal flux across the membrane system in Fig. 1 we can write:

$$h_1(T_{b1} - T_{m1}) = (k_m/\delta)(T_{m1} - T_{m2}) + J\lambda = h_2(T_{m2} - T_{b2}) \tag{5}$$

From Eqs. (4) and (5):

$$(T_{b1} - T_{b2})/J\lambda = [C\lambda(dP/dT)]^{-1}[1 + (k_m/\delta h)] + (1/h) \tag{6}$$

where $h = 1/(1/h_1 + 1/h_2)$ is the overall film heat transfer coefficient.

On the other hand, from Eqs. (4) and (5):

$$T_{m1} - T_{m2} = [1 + (H/h_1) + (H/h_2)]^{-1}(T_{b1} - T_{b2}) = \tau(T_{b1} - T_{b2}) \tag{7}$$

where

$$H = C\lambda(dP/dT) + k_m/\delta \tag{8}$$

and

$$\tau = [1 + (H/h_1) + (H/h_2)]^{-1} = [1 + (H/h)]^{-1} \tag{9}$$

is the temperature polarization coefficient.

Equation (6) may be used for the analysis of experimental results for which T_{b1}, T_{b2}, and J are reported because dP/dT is a function of T_m =

$(T_{b1} + T_{b2})/2$, assuming temperature polarization is similar on both sides of the membrane. Specifically, a fit to a linear function of the values of $(T_{b1} - T_{b2})/J\lambda$ versus those of $1/(dP/dT)\lambda$ should yield an intercept of $1/h$ and a slope of $(1/C)[1 + (k_m/\delta h)]$, from which C may be obtained.

RESULTS AND DISCUSSION

Experiments were carried out for a fixed temperature difference $T_{b1} - T_{b2}$ = 10°C between the bulk phases 1 and 2 (Fig. 1). The stirring rate used varied from 150 to 350 rpm in steps of 50 rpm, and the average temperature from 25 to 65°C in steps of 5°C.

The flux results so obtained are shown in Table 1. The distillate flux increases monotonically with increasing stirring rate, corresponding (11, 12) to a decrease of the heat resistance in the boundary layers of the membrane. On the other hand, the distillate flux increases when the absolute temperature level in the membrane is increased, corresponding to an increase of dP/dT as the temperature become larger. In fact, Eq. (6) of the transport model shows how J increases when h and dP/dT are increased.

Plots of Eq. (6) for the experimental water flux corresponding to the same stirring rate and different average temperatures have been used in order to evaluate h and C. These plots have a correlation coefficient higher than 0.98. Three representative plots are shown in Fig. 2.

The h values obtained from the intercepts of plots of Eq. (6) are shown in Table 2 for different stirring rates and membranes, the h error being lower than 10%. The h values for the same stirring rate and different

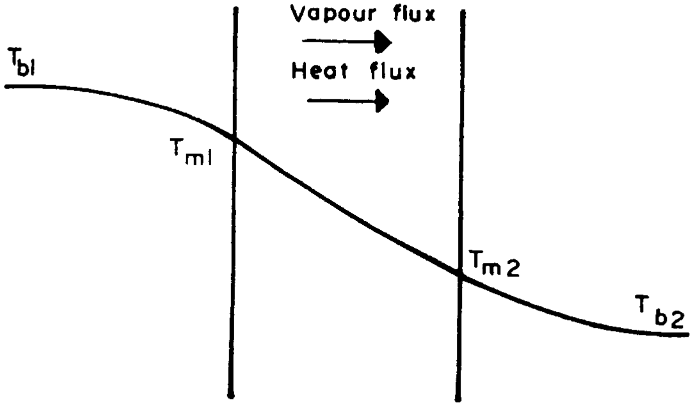

FIG. 1 Schematic representation of membrane distillation.

TABLE 1
Mass Fluxes per Unit Area ($\times 10^3$ kg·m^{-2}·s^{-1}) for the Three Membranes at Various Stirring Rates and Average Temperatures, T_m

Membrane type	rpm	T_m (°C)								
		25	30	35	40	45	50	55	60	65
Lida 200	150	0.84	1.10	1.35	1.68	1.65	1.88	2.11	2.38	2.41
	200	0.93	1.22	1.50	1.81	1.87	2.13	2.35	2.63	2.69
	250	1.00	1.30	1.61	1.89	2.03	2.31	2.52	2.82	2.89
	300	1.05	1.37	1.70	1.95	2.15	2.45	2.65	2.96	3.04
	350	1.10	1.42	1.76	2.00	2.25	2.56	2.75	3.06	3.16
Lida 500	150	1.10	1.24	1.63	1.84	1.98	2.08	2.43	2.74	2.76
	200	1.22	1.39	1.81	1.99	2.19	2.35	2.71	3.05	3.10
	250	1.31	1.49	1.94	2.09	2.34	2.55	2.91	3.28	3.35
	300	1.37	1.57	2.03	2.17	2.45	2.71	3.06	3.46	3.55
	350	1.42	1.63	2.10	2.23	2.54	2.83	3.18	3.59	3.70
Lida 1000	150	1.41	1.71	1.90	2.03	2.14	2.50	2.61	2.88	2.92
	200	1.56	1.89	2.08	2.22	2.34	2.75	2.97	3.25	3.31
	250	1.68	2.03	2.20	2.35	2.48	2.93	3.23	3.52	1.84
	300	1.76	2.13	2.30	2.44	2.58	3.06	3.44	3.73	3.83
	350	1.83	2.20	2.37	2.51	2.65	3.17	3.60	3.89	4.00

membranes are very similar, in agreement with the heat transfer theory for stirred cells (11, 12). In fact, the use of the same experimental device maintains the same hydrodynamic conditions in the layers next to the membrane when the stirring rate is the same.

On the other hand, Schofield et al. (6) showed that most commercial microfiltration membranes have a typical value of k_m/δ of 500 W·m^{-2}·K^{-1}. Taking this value for the membranes studied in this work, the C coefficients have been evaluated from the slopes of plots of Eq. (6). In this way, the following C values are obtained:

$$C(\text{Lida 200}) = (12 \pm 1) \times 10^{-7}\ \text{kg·m}^{-2}\text{·s}^{-1}\text{·Pa}^{-1}$$

$$C(\text{Lida 500}) = (13 \pm 1) \times 10^{-7}\ \text{kg·m}^{-2}\text{·s}^{-1}\text{·Pa}^{-1}$$

$$C(\text{Lida 1000}) = (24 \pm 2) \times 10^{-7}\ \text{kg·m}^{-2}\text{·s}^{-1}\text{·Pa}^{-1}$$

These values are independent of the stirring rate in the system.

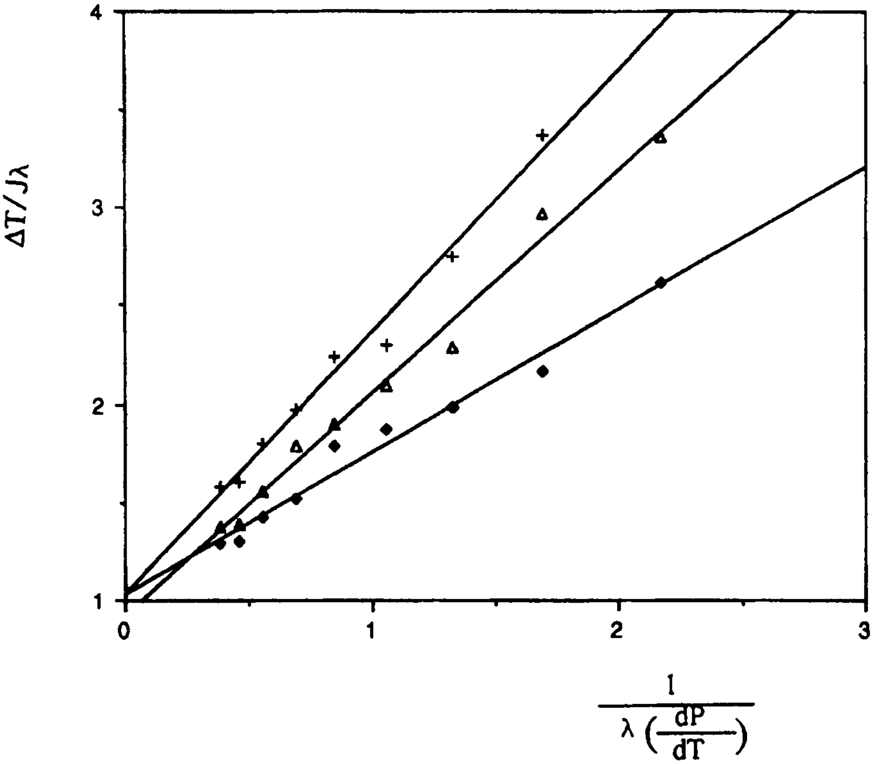

FIG. 2 Plots of $\Delta T/J\lambda$ ($\times$ 10^3 $m^2 \cdot K/W$) vs $1/\lambda(dP/dT)$ ($\times$ 10^9 $kg \cdot K/J \cdot Pa$) corresponding to the results obtained for the Lida 200 (+), Lida 500 (△), and Lida 1000 (◆) membranes when the stirring rate was 250 rpm.

TABLE 2
Overall Film Heat Transfer Coefficient ($W \cdot m^{-2} \cdot K^{-1}$) for Different Membranes and Different Stirring Rates

rpm	Lida 200	Lida 500	Lida 1000
150	857	972	830
200	970	1085	916
250	1053	1172	977
300	1117	1237	1022
350	1168	1289	1057

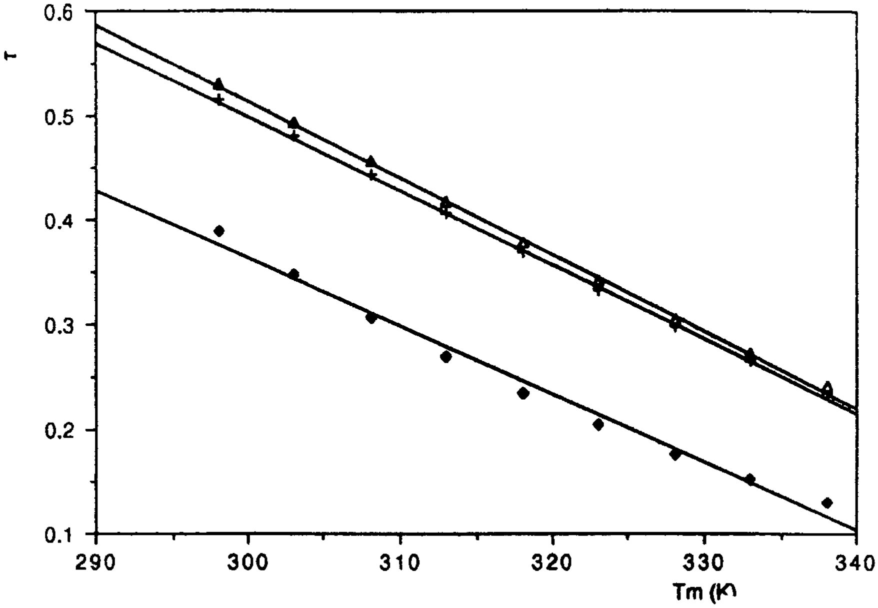

FIG. 3 Temperature polarization coefficient for the different membranes vs average temperature in the system. Stirring rate, 300 rpm. (+) Lida 200, (△) Lida 500, (◆) Lida 1000.

The experimental *C* and *h* values allow us to quantify the τ temperature polarization coefficient. Figure 3 shows the obtained results for the three membranes (and a representative stirring rate) as a function of the average temperature in the system, calculated according to Eqs. (8) and (9). This comportment is different of that obtained in thermoosmosis, where the τ polarization coefficient is independent of the temperature (1). This is because in membrane distillation the *H* coefficient for the membrane, as shown by Eq. (8), accounts for the energy fluxes due to the convective transport of the evaporating vapor occurring simultaneously with heat conduction across the membrane. This increase with temperature is the first contribution which is not present in thermoosmosis processes.

Finally, Fig. 4 shows the τ values obtained for the three membranes as a function of the stirring rate, also calculated according to Eqs. (8) and (9).

All these results highlight the fact that any attempt to compare a membrane distillation transport model with measured data must take into ac-

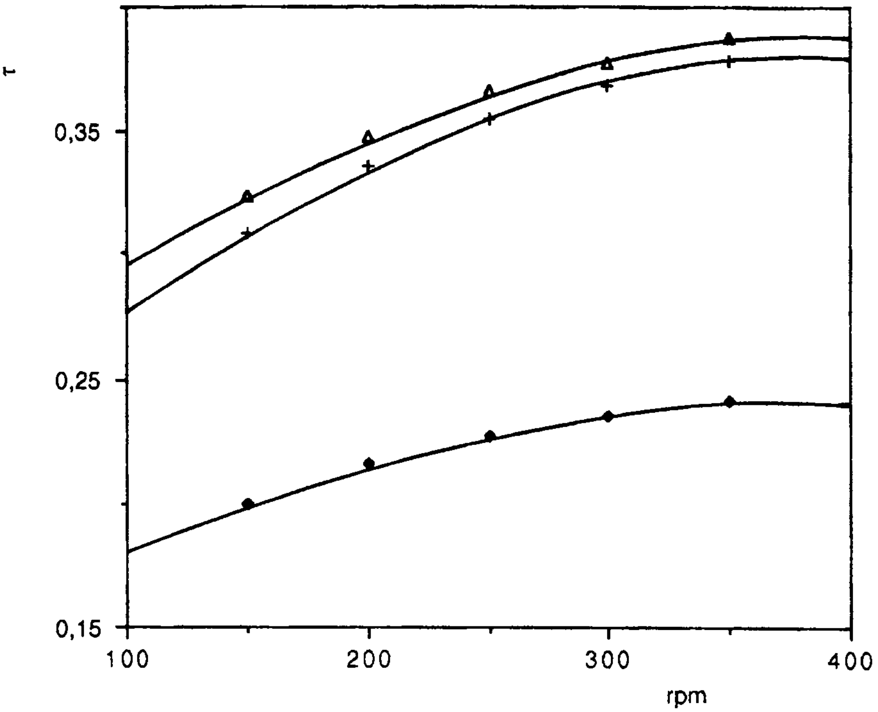

FIG. 4 Temperature polarization coefficient for the different membranes vs stirring rate. Average temperature, 318 K. (+) Lida 200, (Δ) Lida 500, (◆) Lida 1000.

count the significant influence of temperature polarization, which depends of the heat transfer coefficient on the membrane and on the liquid layers.

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