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Parameters That May Influence the Correction Factor in Membranes Subjected to Thermal Gradients

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Abstract

Various treatments are generalized through which the phenomenon of polarization of temperatures can be approached, and an expression for its correction factor which gives an account of the parameters that may influence its value is achieved. An equation is found which enables the determination of some experimental parameters which are difficult to measure through direct methods. The equation mentioned above has been analyzed for both porous and nonporous membranes. In the case of porous membranes, this equation is found useful as long as the thermoosmotic coefficient employed is the one obtained after having taken into account the mechanism which produces the mass transfer that occurred through the membranes. It has not been possible to verify the usefulness of the equation mentioned in the case of nonporous membranes. As to porous membranes, a correction factor (of temperature) depending on the temperature is achieved from the thermoosmotic coefficient.

INTRODUCTION

When a membrane separates two aqueous solutions, each at a different temperature, a mass flow is generated through it. The efficiency of this process is characterized in a wide variety of ways and it depends on various parameters, such as characteristics of membranes, experimental device, values of temperatures, concentration factor, correction factor of temperatures, etc. From these, among the most important is the correction factor of temperatures, which regards the phenomenon of polarization of temperatures. This phenomenon is due to the fact that the temperature difference between the two phases adjacent to the membrane, ΔT_b, is different from the temperature difference between the two faces of the membrane, ΔT_m, as a result of the existence of layers adhering to the membrane.

The estimation of the effects of temperature polarization has been studied by different authors (*1–7*). Belluci (*1*), as to nonporous membranes, considers the heat transfer within the membrane to be due only to a conduction phenomenon. On the other hand, Schofield et al. (*2*), as to porous membranes, presume that a part of the heat which penetrates the membrane is consumed in conduction and the other part in latent heat transfer accompanying mass flux from one phase to the other, without considering the case where mass flux in the membrane conveys heat generation.

In this study those various treatments through which the phenomenon of polarization of temperatures can be approached are generalized. We study the influence that the terms appearing in the expression of the correction factor of temperatures may have, the values that this factor may take, and the way to determine several experimental parameters, like h and k, difficult to measure through direct methods.

THEORETICAL FUNDAMENTAL

In the case where both conduction and the exchange of heat associated with mass transfer take place inside the membrane, the equation of continuity of heat flow in a steady state in the system Phase 1–membrane–Phase 2 (Fig. 1) can be expressed as

$$h_1(T_{b1} - T_{m1}) = (k/\delta)(T_{m1} - T_{m2}) + (M/q)J_RQ = h_2(T_{m2} - T_{b2}) \qquad (1)$$

where k is the overall thermal conductivity of the membrane of area exposed to transport q and thickness δ, h_1 and h_2 are the coefficients of film heat transfer on both sides of the membrane, M is the molecular mass of the permeant, Q is the term for the heat exchange inside the membrane, J_R is the mass flow at a stirring rate R, and T_{mi} and T_{bi} are the temperatures of each one of both faces of the membrane and of their adjacent phases, respectively.

On the assumption that the difference of temperatures between the adjacent phases to the membrane is not very big and that $h_1 = h_2 = h_i$, Eq. (1) can be expressed as

$$\Delta T_b = \Delta T_m[1 + (2k/\delta h_i)] + (2J_RMQ/qh_i) \qquad (2)$$

Equation (2) is a general expression which allows the difference of temperatures between the faces of the membrane, ΔT_m, to be known through certain parameters that can be determined from various experimental methods.

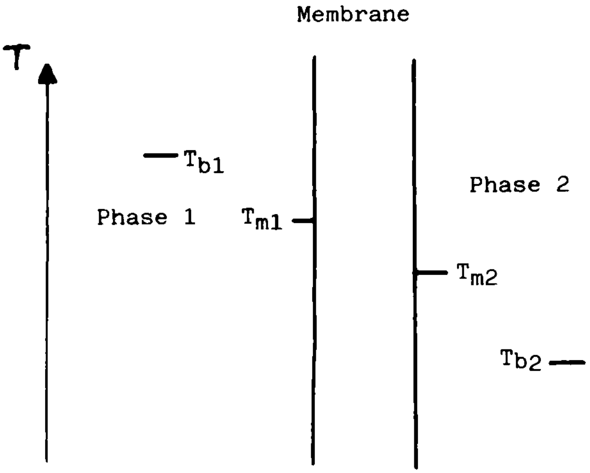

FIG. 1. Temperature profile in the system Phase 1–membrane–Phase 2.

If we bear in mind that $J_R = (q/\delta)B^{\mathrm{exp}}\Delta T_b = (q/\delta)B^{\mathrm{cor}}\Delta T_m$, where B^{exp} is the thermoosmotic coefficient obtained directly from the experimental results, and B^{cor} is the thermoosmotic corrected coefficient (that is to say, the one which could be obtained after having eliminated the effect of temperature polarization), then we reach an expression for the correction factor of temperatures as

$$f = \frac{\Delta T_b}{\Delta T_m} = \frac{B^{\mathrm{cor}}}{B^{\mathrm{exp}}} = \frac{1 + (2k/\delta h_i)}{1 - (2B^{\mathrm{exp}}MQ/\delta h_i)} \tag{3}$$

which allows its determination from the known values of the different parameters appearing in the expression.

Equation (3) can be transformed into

$$\frac{1}{B^{\mathrm{exp}}Q} = \frac{1 + (2k/\delta h_i)}{B^{\mathrm{cor}}Q} + \frac{2M}{\delta h_i} \tag{4}$$

which can be used to obtain parameters hard to determine from others known by experiment. Specifically, a fit to a linear function of the values of $1/B^{\mathrm{exp}}Q$ opposite to those of $1/B^{\mathrm{cor}}Q$, will allow determination of the value of h_i from the ordinate at the origin, and the value of k from the slope of the fit.

ANALYSIS OF RESULTS

The determination of the correction factor following Eq. (3) can be achieved either from the ratio B^{cor}/B^{exp}, which requires knowing the value of B^{cor}, or from the various parameters of the last member of Eq. (3).

If f is worked out as the ratio B^{cor}/B^{exp}, its value will depend on how B^{cor} is determined. Among the references, we can find different ways to get to know B^{cor} (*1*, *3*, *5–7*) which are based mainly on the fact that the layers on the faces of the membrane will be eliminated as soon as the stirring rate of the phases adjacent to the membrane is infinite. In this way, the effect of temperature polarization is removed and the thermo-osmotic coefficient corresponding to this situation will be B^{cor}.

The usual procedure consists of fitting the values of $1/B^{exp}$ to a straight line as a function of the inverse of the stirring rate of the adjacent phases to the membrane, obtaining the value of B^{cor} from the ordinate at the origin of the fit, which corresponds to an infinite stirring rate. Nevertheless, the way of using the results varies from one author to another.

In this work we have proceeded to a systematic analysis of the results in Refs. *3* and *7*, fitting the values of $1/B^{exp}$ or $1/(\Delta P/\Delta T)$ to a linear function of the inverse of the stirring rate in the phases adjacent to the membrane, as follows:

(1) Considering the values of B^{exp} or $\Delta P/\Delta T$ at a stirring rate of zero.
(2) Not considering the values of B^{exp} or $\Delta P/\Delta T$ at a stirring rate of zero.
(3) Without considering the values of B^{exp} or $\Delta P/\Delta T$ at zero and 50 rpm.

Table 1, 2, and 3 present the values of B^{cor} and its correspondent f for a determined stirring rate obtained from some of the instances studied. The statistical parameters that characterize the fits are also presented.

An analysis of the results shows that the fits are statistically better when the null stirring rate is not taken into account, and therefore the values of B^{cor} and f obtained in this way must be considered the most correct ones.

TABLE 1
Results of the Fits of Data in Ref. *3* to Determine $(\Delta P/\Delta T)^{cor}$

	Correlation coefficient	Zero comparison, Student T test	$(\Delta P/\Delta T)^{cor}$ ($N/m^{-2}\cdot K$)
All stirring rate	0.9973	$t_1 = 15$ $t_2 = 23$	4001.12
0 rpm is eliminated	—	—	—
0 and 50 rpm are elminated	0.9985	$t_2 = 70$ $t_2 = 31$	3483.1

TABLE 2
Results of the Fits of Data for Membrane 1 in Ref. 4 to Determine B^{cor} (T = 40°C, ΔT = 10°C) and f (T = 40°C, ΔT = 10°C, stirring rate = 200 rpm)

	Correlation coefficient	Zero comparison, Student T test	B^{cor} (mol/m·s·K)	f
All stirring rate	0.9951	$t_1 = 4$ $t_2 = 18$	3.52×10^{-6}	2.50
0 rpm is eliminated	0.9995	$t_1 = 55$ $t_2 = 53$	2.07×10^{-6}	1.47
0 and 50 rpm are eliminated	0.9959	$t_1 = 25$ $t_2 = 15$	2.44×10^{-6}	1.45

The correction factor, f, can also be determined through the last member of Eq. (3). This way has been employed to consider the experimental conditions required to obtain all the possible values that the correction factor may take. That is to say, as a rule there is nothing which could prevent f from either taking positive or negative values, or being higher or lower than unity. Therefore, with both ΔT_b and $1 + (2k/\delta h_i)$ always positive, the sign f may take will be affected by the sign both B^{exp} (or J_R) and Q have, since these can be either positive or negative. In short, the following cases may arise:

(A) $f > 1$:
 Positive: $J_RQ < 0$ or $0 < J_RQ < (\delta h_i/2M)$
 Negative $J_RQ > (\delta h_i/2M)$ together with $J_RQ > 0$
(B) $f < 1$:
 Positive: $k\Delta T_b < -J_RQM$ together with $J_RQ < 0$
 Negative: Not possible

TABLE 3
Results of the Fits of Data for Membrane 2 in Ref. 4 to Determine B^{cor} (T = 35°C, ΔT = 10°C) and f (T = 35°C, ΔT = 10°C, stirring rate = 200 rpm)

	Correlation coefficient	Zero comparison, Student T test	B^{cor} (mol/m·s·K)	f
All stirring rate	0.9947	$t_1 = 2.9$ $t_2 = 17$	1.95×10^{-6}	2.78
0 rpm is eliminated	0.9999	$t_1 = 194$ $t_2 = 203$	1.04×10^{-6}	1.48
0 and 50 rpm are eliminated	0.9998	$t_1 = 122$ $t_2 = 82$	1.05×10^{-6}	1.50

The values of B^{exp} in mol/m·s·K as found in the references vary between 3×10^{-10} for nonporous membranes (with a thickness of the order of 10^{-6} m and a thermal conductivity $k = 0.45$ W/m·K) and a range of 10^{-6} to 10^{-7} for porous membranes (with a thickness of the order of 10^{-4} m and $k = 0.05$ W/m·K). The values of Q range from -170 kJ/kg up to 2357 kJ/kg, with typical values of h_i between 100 to 10,000 W/m^2·K.

With these data, an analysis of the possible values of f leads to the following results.

Negative f: Both J_R and Q must be either positive or negative at the same time. In every case studied, the values of J_R (or B^{exp}) are positive, which requires the values of Q to also be positive. Under these conditions, no instance of $J_RQ > (\delta h_i/2M)$, which would allow a negative correction factor to appear, has been found in the references.

Positive f: Considering that it has always been found that $J_R > 0$, a value of $f < 1$ implies $Q < 0$; this condition is observed for nonporous membranes but they do not observe the other condition needed: $k\Delta T_b < -J_RQM$. Finally, $f > 1$ requires either $J_RQ < 0$ be fulfilled, which happens in the cases of nonporous membranes, or $0 < J_RQ < (\delta h_i/2M)$, which is observed for porous membranes.

The study of Eq. (4) has been carried out considering the use of both nonporous and porous membranes. In the first case, measurements of the thermoosomosis of water have been carried out with cellophane membrane 600P with a thickness of $(56.4 \pm 1.8) \times 10^{-6}$ m subjected to the same treatment and with the experimental device described in a previous work (*6*). The measurements have been carried out at various average temperatures and under various temperature differences, and the values of B^{exp} were obtained at the different stirring rates presented in Table 4. The values of B^{cor} appearing in the last line of Table 4 were obtained through using the method described above.

The various values of B^{exp} at a given stirring rate, together with those corresponding to B^{cor} under the same conditions of T and ΔT from Table 4, have been used to make a linear fit of $1/B^{exp}$ as a function of $1/B^{cor}$ according to Eq. (4). Although the parameters of the fit have statistically acceptable values, considering the value of $Q = 13$ J/g given in Ref. *9*, the values of h_i and k, obtained from the ordinate at the origin and from the slope of the fit, respectively, do not have a physical meaning, which can be due to either the insufficient amount of data or the fact that the value used for Q may not be applicable in this case.

For porous membranes it is known (*2*) that the term of nonconductive heat appearing in Eq. (1) is caused by a transfer of latent heat of evaporation accompanying the mass flow, and the driving force for mass transfer is related to the difference in water vapor pressure between the two sides

TABLE 4
Values of B^{exp} and B^{cor} ($\times$ 10^{10} mol/m·s·K) for Various Experimental Situations (T_1 = 37.5°C, ΔT_1 = 5°C), (T_2 = 40°C, ΔT_2 = 10°C), (T_3 = 42.5°C, ΔT_3 = 15°C). Cellophane 600P Membrane

rpm	$B_1^{\mathrm{exp}}(T_1,\Delta T_1)$	$B_2^{\mathrm{exp}}(T_2,\Delta T_2)$	$B_3^{\mathrm{exp}}(T_3,\Delta T_3)$
150	2.58	1.93	1.50
180	3.07	2.28	1.76
210	3.39	2.52	1.92
250	3.77	2.79	2.11
290	4.11	3.03	2.27
340	4.44	3.27	2.42
∞	9.14	6.54	4.31

of the membrane. Since the vapor pressure inside the membrane cannot be directly measured, the flow should be expressed as a function of the temperatures in such a way that if the temperature difference is not too big, it can be expressed as

$$J^* = C(dP/dT)_T \Delta T_m \tag{5}$$

where C is a coefficient of mass transfer through the membrane, related to a true thermoosmotic coefficient, $(B^{\mathrm{cor}})^*$, with the same meaning used before, by

$$C(dP/dT)_T = M(B^{\mathrm{cor}})^*/\delta \tag{6}$$

and $(dP/dT)_T$ can be determined by using the Clausius–Clapeyron equation.

By means of some algebraic changes and by using Eqs. (5) and (6), Eq. (4) can be expressed as

$$\frac{1}{B^{\mathrm{exp}}Q} = \frac{M[1 + (2k/\delta h_i)]}{C(dP/dT)_T Q\delta} + \frac{2M}{\delta h_i} \tag{7}$$

By fitting the values of $1/B^{\mathrm{exp}}Q$ as a linear function of $1/(dP/dT)_T Q$ according to Eq. (7), the value of h_i can be obtained from the ordinate at the origin of the fit. Also, by using Eq. (6) and knowing the value of k, the value of $(B^{\mathrm{cor}})^*$ can be obtained from the slope of the fit.

The data of porous membranes from Refs. *7* and *8* have been analyzed through Eqs. (4) and (7). By an analysis through Eq. (4), the values of h_i

TABLE 5
Values of $(B^{cor})^*$ (mol/m·s·K) and of f^* at Various Temperatures in °C and ΔT = 10°C for Membrane 1 in Ref. 7. Stirring Rate = 200 rpm

Temperature	$(B^{cor})^* \times 10^7$	f^*
35	17.72	1.61
40	22.41	1.59
45	28.10	1.77
50	34.91	1.79
55	43.02	2.11

and k can be determined on the assumption that B^{cor} and Q are known. By Eq. (7), h_i and $(B^{cor})^*$ can be determined, supposing k and Q are known.

When Eq. (4) is applied to the data from Ref. 7, using the B^{cor} obtained through a fit as described before, we find that, in the case of either Membrane 1 or Membrane 2, when the temperature difference and the rate of stirring have been fixed and heat Q is considered to be the latent heat of evaporation of water at a temperature T, the values of h_i are not physically acceptable although the statistical parameters of the fits are good.

The data of Ref. 7 have also been applied to Eq. (7). Using the same value of Q as before, and an average value of k from those found in the references for this type of membrane (k = 0.05 W/m·K), a physically significant value of h_i together with good statistical fits is obtained. With these fits we obtain the true thermoosmotic coefficients $(B^{cor})^*$ which allows us to find correction factors of temperature f^*. These results are presented in Table 5. The same procedure has been employed with the data of Ref. 8 with similar results, which are presented in Table 6.

TABLE 6
Values of $(B^{cor})^*$ (mol/m·s·K) and f^* at Various Temperatures in °C for the Membrane in Ref. 8

Temperature	$(B^{cor})^* \times 10^7$	f^*
20	2.05	1.42
30	3.45	1.47
40	5.57	1.63
50	8.65	1.65
60	13.02	1.83
70	18.99	1.81

An examination of the values of the correction factor of temperature in Tables 5 and 6 shows that these values are connected with temperature. An analysis of correlation between the pairs of values (f^*, T) has allowed us to establish the existence of a linear correlation between them.

CONCLUSIONS

No experimental situation has been found in which a correction factor of temperature results either in a negative or in a lower than unity value.

The B^{cor} determined through a linear fit of $1/B^{\mathrm{exp}}$ as a function of the inverse of the rate of stirring in the cell is not adequate to be used in Eq. (4) for porous and nonporous membranes.

$(B^{\mathrm{cor}})^*$ is dependent on temperature, therefore so is f^*, possibly due to the equation by which the former is determined.

The use of Eq. [7] allows values such as h_i and k to be determined.

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