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Heat Transfer Effects in Facilitated Transport Liquid Membranes

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ABSTRACT

facilitated transport. two limiting regimes where steady-state analytical solutions are available; diffusion-limited (*reaction equilibrium*) and reaction-limited (*frozen condition*). For intermediate cases, numerical solutions are available. All of these models are valid for isothermal conditions. It is possible in practice that the system may not be isothermal. The gas streams on each side of the membrane may be at different temperatures and/or there can be heat of reaction effects. These effects can cause the total facilitated flux to deviate from the isothermal case.

The results of including temperature effects are incorporated in a dimensionless factor θ. θ is the facilitation factor for the non-isothermal case divided by the facilitation factor for the isothermal case. Results show that the imposition of a temperature gradient can cause a significant increase or decrease in the facilitated flux. In extreme cases, there can even be a reversal in the direction of the facilitated flux. The heat of reaction had no noticeable effect for the conditions studied.

INTRODUCTION

under isothermal, one-dimensional conditions is described elsewhere (2, 10, 11).

The mass transfer in facilitated transport can be affected if a temperature gradient is imposed across the liquid film. This non-isothermal condition is quite likely to occur in practice since it is difficult to precisely control the temperature on both sides of the film due to process conditions. Previous researchers have studied various aspects of the above problem. Most have been concerned with combined heat and mass transfer with a reversible reaction taking place, either in the medium being studied or at a boundary. The primary focus has been on the enhancement of heat transfer. Fay and Riddell (12) studied the case of heat transfer from dissociating air with a finite reaction rate. They used numerical methods to integrate the laminar boundary layer equations. Broadwell (13) studied this same problem using a linearized reaction rate expression. Hirschfelder (14) considered the more general problem of combined heat and mass transfer near reaction equilibrium. He developed an effective thermal conductivity to incorporate these combined effects. Brian and Reid (15), Brian and Bodman (16), and Goddard, Schultz, and Suchdeo (17) gave theoretical solutions for enhanced heat transfer due to chemical reaction. Goddard et al. specifically demonstrated that the analysis of facilitated mass transport could be applied to enhanced heat transport.

The objective of this study is to demonstrate the effect of heat transfer on the mass transport rate. It will be shown that

heat transfer can either enhance or impede the facilitated mass transport.

Problem Formulation

The most common reaction mechanism studied for facilitated transport is (7, 10, 11)

$$A + B \underset{k_2}{\overset{k_1}{\rightleftarrows}} AB \qquad (1)$$

where A = permeate

B = carrier

AB = permeate-carrier complex

The differential mass balances which describe this reaction mechanism at steady-state are

$$0 = D_A \frac{d^2C_A}{dx^2} - k_1C_AC_B + k_2C_{AB} \qquad (2)$$

$$0 = D_B \frac{d^2C_B}{dx^2} - k_1C_AC_B + k_2C_{AB} \qquad (3)$$

$$0 = D_{AB} \frac{d^2C_{AB}}{dx^2} + k_1C_AC_B - k_2C_{AB} \qquad (4)$$

where

D_i = diffusion coefficient of component i

C_i = concentration of component i in the liquid film.

$$k_1 = k_{10} e^{-E_f/RT} \tag{5}$$

$$k_2 = k_{20} e^{-E_r/RT} \tag{6}$$

The above equations assume one-dimensional transport across a planar membrane.

A steady-state energy balance can also be written across the membrane.

$$0 = -k \frac{d^2T}{dx^2} + (\Delta Hr)\, D_{AB} \frac{d^2C_{AB}}{dx^2} \tag{7}$$

where:

k = thermal conductivity of liquid film

ΔHr = heat of reaction

The boundary conditions are

@x = 0	$C_A = C_{Ao}$	$\frac{dC_B}{dx} = \frac{dC_{AB}}{dx} = 0$	$T = T_o$
@x = L	$C_A = 0$	$\frac{dC_B}{dx} = \frac{dC_{AB}}{dx} = 0$	$T = T_L$

This second boundary condition corresponds to the limiting condition of maximizing the concentration gradient across the liquid film. Here L is the membrane thickness.

The following assumptions are made:

1. $D_B = D_{AB}$. This equality is based on assuming that the carrier (B) and the complex (AB) are approximately equal in size.

2. The thermal conductivity and diffusion coefficients are considered to be constant over the temperature range shown in Table 1. This is obviously an approximation but the temperature effect on the kinetic rate constants is much larger than the effect on either the thermal conductivity or the diffusion coefficients.

3. Ionic effects are neglected.

4. Heat of reaction (ΔHr) is negligible. This term was originally used in the simulations and found to be negligible for the conditions studied.

A mass balance on the carrier yields

$$C_T = C_B + C_{AB} \tag{8}$$

where C_T is the initial carrier concentration in the liquid film.

The following dimensionless variables are introduced (11, 18)

$$T^* = \frac{T - T_o}{T_L - T_o} \tag{9}$$

$$X = \frac{x}{L} \tag{10}$$

$$C_A^* = \frac{C_A}{C_{Ao}} \tag{11}$$

$$C_B^* = \frac{C_B}{C_T} \tag{12}$$

$$C_{AB}^* = \frac{C_{AB}}{C_T} \tag{13}$$

$$E_f^* = \frac{E_f(T_L - T_o)}{RT_o^2} \tag{14}$$

$$E_r^* = \frac{E_r(T_L - T_o)}{RT_o^2} \tag{15}$$

$$K = \frac{k_{10}'}{k_{20}'} \tag{16}$$

$$\varepsilon = \frac{D_{AB}}{k_{20}'L^2} \tag{17}$$

$$\alpha = \frac{D_{AB}}{D_A}\frac{C_T}{C_{Ao}} \tag{18}$$

where

$$k_1 = k_{10}'e^{E_f^*T^*} \tag{19}$$

$$k_2 = k_{20}'e^{E_r^*T^*} \tag{20}$$

$$k_{10}' = k_{10}e^{-E_{f/RT_o}} \tag{21}$$

$$k_{20}' = k_{20}e^{-E_{r/RT_o}} \tag{22}$$

Equations (19) and (20) were obtained by using the Laurent expansion for the exponential term in equations (6) and (7) and retaining only the zero and first order terms. This is valid if $RT^*(T_L - T_o) \ll T_o$. This condition is valid for the cases studied here.

The dimensionless equations and boundary conditions become

$$0 = \frac{d^2C_A^*}{dX^2} - \frac{\alpha K}{\varepsilon} e^{E_f^*T^*}\left[C_A^*(1-C_{AB}^*) - \frac{1}{K}e^{(E_r^*-E_f^*)T^*}C_{AB}^*\right] \tag{23}$$

$$0 = \frac{d^2C_{AB}^*}{dX^2} - \frac{K}{\varepsilon} e^{E_f^*T^*} \left[C_A^* (1-C_{AB}^*) - \frac{1}{K} e^{(E_r^*-E_f^*)T^*} C_{AB}^* \right] \quad (24)$$

$$0 = \frac{d^2T^*}{dX^2} \quad (25)$$

$$@X = 0 \qquad C_A^* = 1 \qquad \frac{dC_{AB}^*}{dX} = 0 \qquad T^* = 0$$

$$@X = 1 \qquad C_A^* = 0 \qquad \frac{dC_{AB}^*}{dX} = 0 \qquad T^* = 1$$

The facilitation factor (F) is defined as the total facilitated flux divided by the simple diffusion flux of the permeate. For this system, F becomes

$$F = - \left. \frac{dC_A^*}{dX} \right|_{X = 1} \quad (26)$$

The problem formulation was cast in dimensionless form for two reasons. Dimensionless variables allow one to generalize the solution so it is useful for a range of specific variables. Also, the proper choice of dimensionless variables allows one to give physical interpretations to these variables and the values of these dimensionless variables help to describe the physical situation.

Results

Equations (23), (24), and (25) were solved numerically using a computer package D02GAF (NAG library) to evaluate F. To determine the effect of the temperature gradient on the facilitation factor, θ is defined as

$$\theta = \frac{F(\text{non-isothermal case})}{F \quad (\text{isothermal case})} \tag{27}$$

A value of θ greater than 1 would correspond to an additional enhancement of the facilitated flux, while a value of θ less than 1 would reflect a reduction in the facilitated flux. θ equals 1 corresponds to no effect of temperature on the facilitated flux. The dimensionless variables K, ε, α, E_f^*, and E_r^* were varied to obtain θ for these different physical and operating conditions. The range in physical properties and operating conditions used in this study are shown in Table 1. Results are shown in figures 1a to 1h which demonstrate the effect of each dimensionless variable on θ .

The carrier concentration is directly proportional to α. As α increases, the effect on θ, whether positive or negative, is also increased. This is due to the fact that the reaction rate and, therefore, the facilitation is affected by carrier concentration.

E_f^* and E_r^* are a measure of the change in the kinetic rate constant with temperature. The sign of E_f^* and E_r^* is determined by the direction of the temperature increase. A positive sign indicates $T_L > T_o$ and a negative sign indicates $T_L < T_o$. Note that E_f^* and E_r^* must both be positive or both negative. For E_f^* and E_r^* positive, $\theta > 1$ if $E_f^* \leq E_r^*$ and $\theta < 1$ if $E_f^* \geq E_r^*$. This is consistent from a physical standpoint. If $E_f^* < E_r^*$, the forward reaction rate constant will increase at a slower rate than the reverse reaction rate constant. So, at X = 0, the forward reaction occurs and the reverse reaction is faster relative to the forward reaction at X = 1 than it would be in the isothermal case since the temperature is higher. More permeate is released and the facili-

Table 1

Values of various parameters used in model simulation

$$k_{10}' = 7.15 \times 10^{3}\ \mathrm{cm^3/mol \cdot s}$$

$$k_{20}' = 9.00 \times 10^{-3}\ \mathrm{s^{-1}}$$

$$D_{AB} = 2.0 \times 10^{-6}\ \mathrm{cm^2/s}$$

$$D_{A} = 1.5 \times 10^{-5}\ \mathrm{cm^2/s}$$

$$50\mu\mathrm{m} \leq L \leq 10^{-2}\mathrm{m}$$

$$0.1 \leq C_T/C_{Ao} \leq 500$$

$$1 \times 10^{-6}\ \frac{\mathrm{mol}}{\mathrm{cm^3}} \leq C_T \leq 5 \times 10^{-5}\ \frac{\mathrm{mol}}{\mathrm{cm^3}}$$

$$295\mathrm{K} \leq T_o \leq 325\mathrm{K}$$

$$1\mathrm{K} \leq \left| T_L - T_o \right| \leq 30\mathrm{K}$$

$$1 \times 10^{4}\ \frac{\mathrm{J}}{\mathrm{mol}} \leq E_f \leq 7 \times 10^{4}\ \frac{\mathrm{J}}{\mathrm{mol}}$$

$$1 \times 10^{4}\ \frac{\mathrm{J}}{\mathrm{mol}} \leq E_r \leq 7 \times 10^{4}\ \frac{\mathrm{J}}{\mathrm{mol}}$$

tation is increased. The same reasoning explains the decrease in Θ if $E_f^* > E_r^*$. For E_f^* and E_r^* negative, the above conclusions are reversed since the temperature gradient is in the opposite direction. When E_f^* and E_r^* are equal and small, Θ equals 1. This corresponds to no temperature effect on facilitation.

ε is an inverse Damkohler number and relates diffusion to reaction times. A small value of ε corresponds to rapid reaction

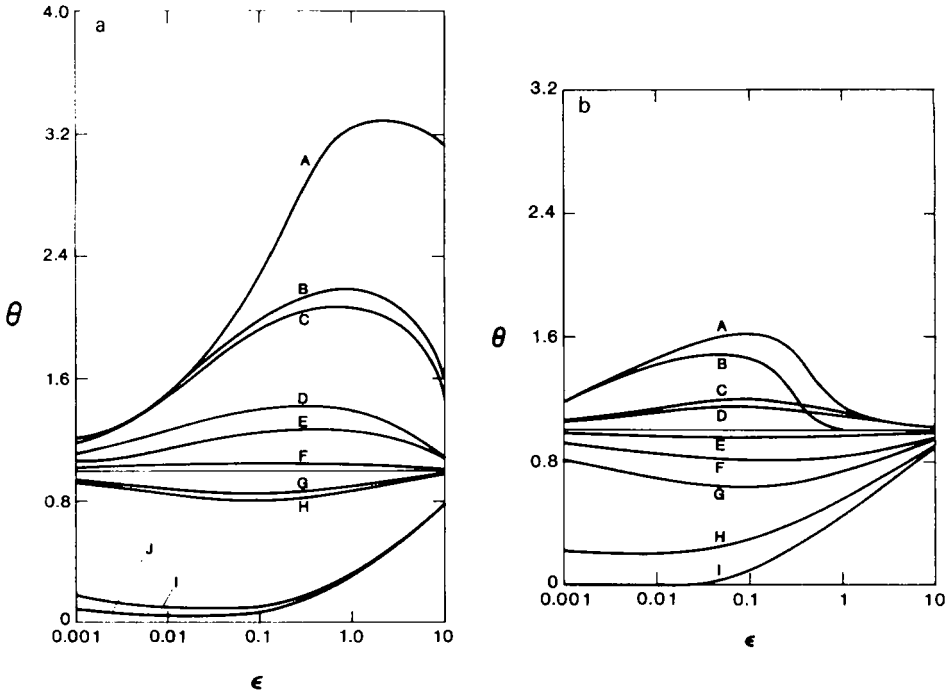

FIGURE 1a. θ vs ε for K = 5, α = 10, with A) $E_f^* = 10$, $E_r^* = 10$; B) $E_f^* = 1$, $E_r^* = 10$; C) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 10$; D) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 1$; E) $E_f^* = 1$, $E_r^* = 1$; F) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 1$; G) $E_f^* = 1$, $E_r^* = 1$; H) $E_f^* = 1$, $E_r^* = 0.001, 0.01$; I) $E_f^* = 10$, $E_r^* = 1$; J) $E_f^* = 10$, $E_r^* = 0.001, 0.01, 0.1$.

FIGURE 1b. θ vs ε for K = 5, α = 10, with A) $E_f^* = -10$, $E_r^* = -0.001, -0.01, -0.1$; B) $E_f^* = -10$, $E_r^* = -1$; C) $E_f^* = -1$, $E_r^* = -0.001, -0.01$; D) $E_f^* = -1$, $E_r^* = -0.1$; E) $E_f^* = -0.001, -0.01, -0.1$; F) $E_f^* = -1$, $E_r^* = -1$; G) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -1$; H) $E_f^* = -10$, $E_r^* = -10$; I) $E_f^* = -0.001, -0.01, -0.1, -1$, $E_r^* = -10$.

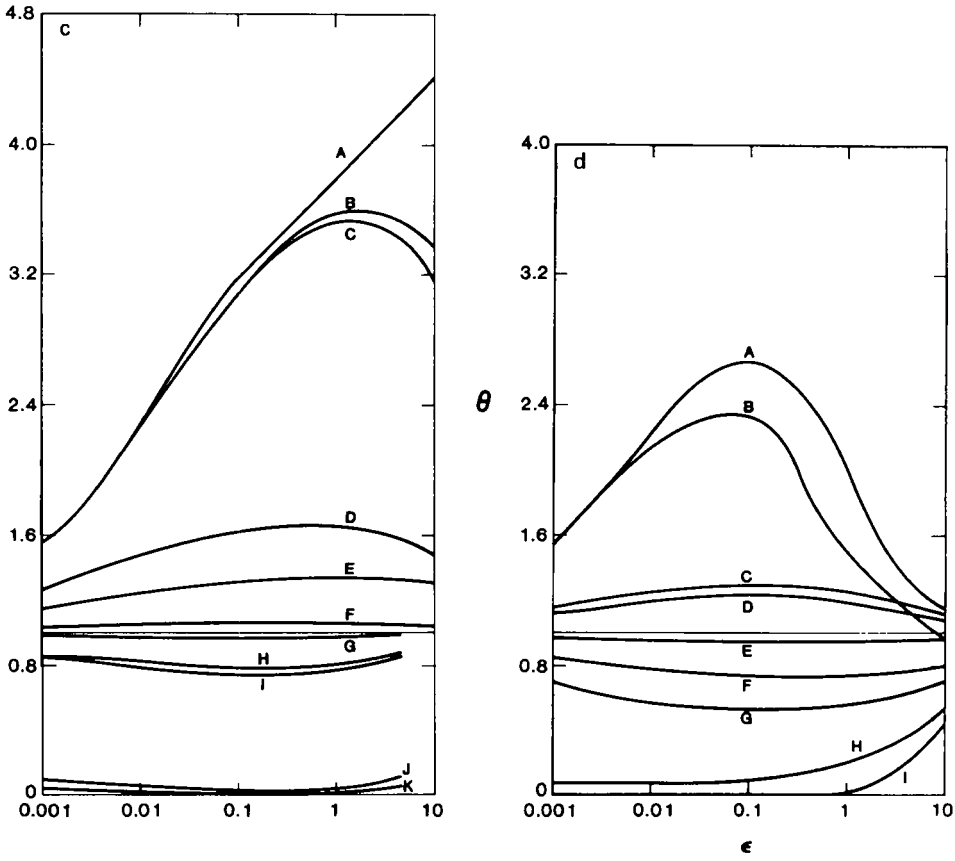

FIGURE 1c. θ vs ε for K = 5, α = 100, with A) $E_f^* = 10$, $E_r^* = 10$; B) $E_f^* = 1$, $E_r^* = 10$; C) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 10$; D) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 1$; E) $E_f^* = 1$, $E_r^* = 1$; F) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 0.1$; G) $E_f^* = 0.1$, $E_r^* = 0.001, 0.01$; H) $E_f^* = 1$, $E_r^* = 0.1$; I) $E_f^* = 1$, $E_r^* = 0.01, 0.001$; J) $E_f^* = 10$, $E_r^* = 1$; K) $E_f^* = 10$, $E_r^* = 0.001, 0.01, 0.1$.

FIGURE 1d. θ vs ε for K = 5, α = 100, with A) $E_f^* = -10$, $E_r^* = -0.001, -0.01, -0.1$; B) $E_f^* = -10$, $E_r^* = -1$; C) $E_f^* = -1$, $E_r^* = -0.001, -0.01$; D) $E_f^* = -1$, $E_r^* = -0.1$; E) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = 0.1$; F) $E_f^* = -1$, $E_r^* = -1$; G) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -1$; H) $E_f^* = -10$, $E_r^* = -10$; I) $E_f^* = -0.001, -0.01, -0.1, -1$, $E_r^* = -10$.

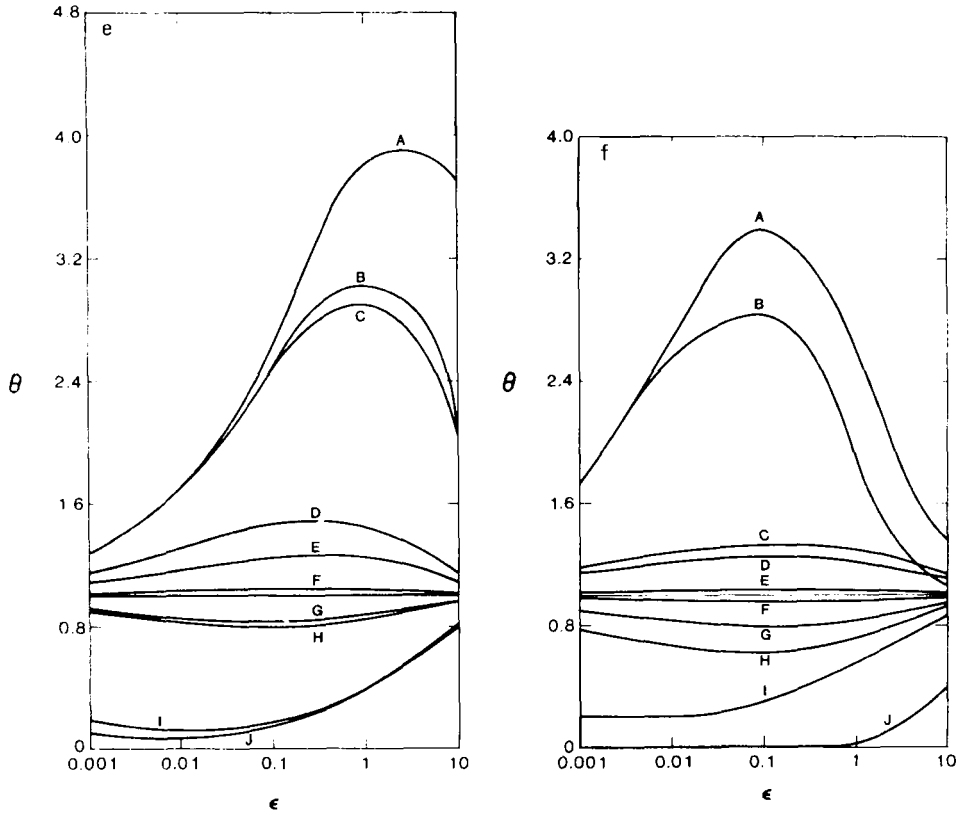

FIGURE 1e. θ vs ε for K = 10, α = 10, with A) $E_f^* = 10$, $E_r^* = 10$; B) $E_f^* = 1$, $E_r^* = 10$; C) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 10$; D) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 1$; E) $E_f^* = 1$, $E_r^* = 1$; F) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 0.1$; G) $E_f^* = 1$, $E_r^* = 0.1$; H) $E_f^* = 1$, $E_r^* = 0.001, 0.01$; I) $E_f^* = 10$, $E_r^* = 1$; J) $E_f^* = 10$, $E_r^* = 0.001, 0.01, 0.1$.

FIGURE 1f. θ vs ε for K = 10, α = 10, with A) $E_f^* = -10$, $E_r^* = -0.001, -0.01, -0.1$; B) $E_f^* = -10$, $E_r^* = -1$; C) $E_f^* = -1$, $E_r^* = -0.001, -0.01, -0.1$; D) $E_f^* = -1$, $E_r^* = -0.1$; E) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -0.1$; F) $E_f^* = -1$, $E_r^* = -1$; G) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -1$; H) $E_f^* = -10$, $E_r^* = -10$; I) $E_f^* = -0.001, -0.01, -0.1, -1$, $E_r^* = -10$.

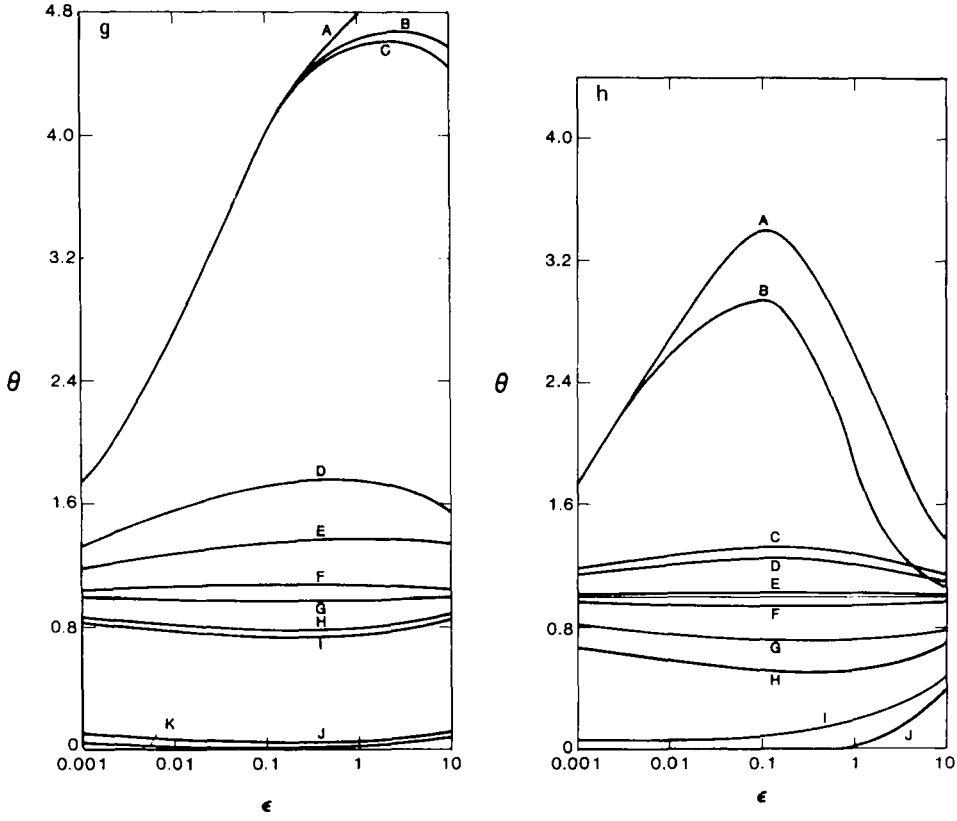

FIGURE 1g. θ vs ε K = 10, α = 100, with A) $E_f^* = 10$, $E_r^* = 10$; B) $E_f^* = 1$, $E_r^* = 10$; C) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 10$; D) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 1$; E) $E_f^* = 1$, $E_r^* = 1$; F) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 0.1$; G) $E_f^* = 0.1$, $E_r^* = 0.001, 0.01$; H) $E_f^* = 1$, $E_r^* = 0.1$; I) $E_f^* = 1$, $E_r^* = 0.001, 0.01$; J) $E_f^* = 10$, $E_r^* = 1$; K) $E_f^* = 10$, $E_r^* = 0.001, 0.1, 0.1$.

FIGURE 1h. θ vs ε for K = 10, α = 100, with A) $E_f^* = -10$, $E_r^* = -0.001, -0.01, -0.1$; B) $E_f^* = -10$, $E_r^* = -1$; C) $E_f^* = -1$, $E_r^* = -0.001, -0.01, -0.1$; D) $E_f^* = -1$, $E_r^* = -0.1$; E) $E_f^* = -0.1$, $E_r^* = -0.001, -0.01$; F) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -0.1$; G) $E_f^* = -1$, $E_r^* = -1$; H) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -1$; I) $E_f^* = -10$, $E_r^* = -10$; J) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -10$.

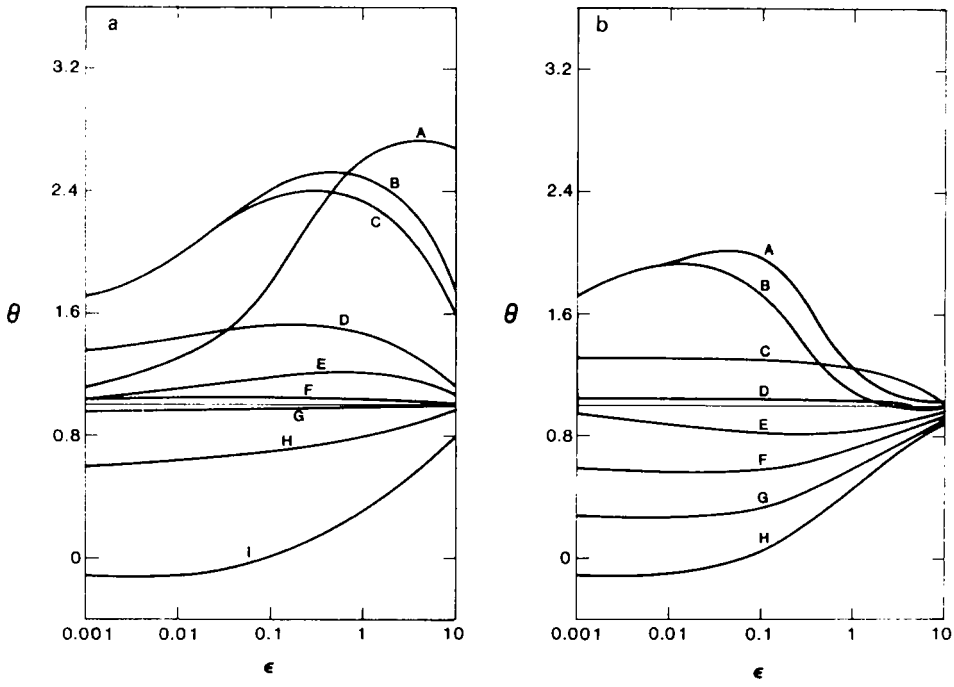

FIGURE 2a. θ vs ε for $C_A^*|_{\chi=1} = 0.1$, $K = 5$, $\alpha = 10$, with A) $E_f^* = 10$, $E_r^* = 10$; B) $E_f^* = 1$, $E_r^* = 10$; C) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 10$; D) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 1$; E) $E_f^* = 1$, $E_r^* = 1$; F) $E_f^* = 0.001, 0.01, 0.1$, $E_r^* = 0.1$; G) $E_f^* = 0.1$, $E_r^* = 0.001, 0.01$; H) $E_f^* = 1$, $E_r^* = 0.001, 0.01, 0.1$; I) $E_f^* = 10$, $E_r^* = 0.001, 0.01, 0.1, 1$.

FIGURE 2b. θ vs ε for $C_A^*|_{\chi=1} = 0.1$, $K = 5$, $\alpha = 10$, with A) $E_f^* = -10$, $E_r^* = -0.001, -0.01, -0.1$; B) $E_f^* = -10$, $E_r^* = -1$; C) $E_f^* = -1$, $E_r^* = -0.001, -0.01, -0.1$; D) $E_f^* = -0.1$, $E_r^* = -0.001, -0.01$; E) $E_f^* = -1$, $E_r^* = -1$; F) $E_f^* = -0.001, -0.01, -0.1$, $E_r^* = -1$; G) $E_f^* = -10$, $E_r^* = -10$; H) $E_f^* = -0.001, -0.01, -0.1, -1$, $E_r^* = -10$.

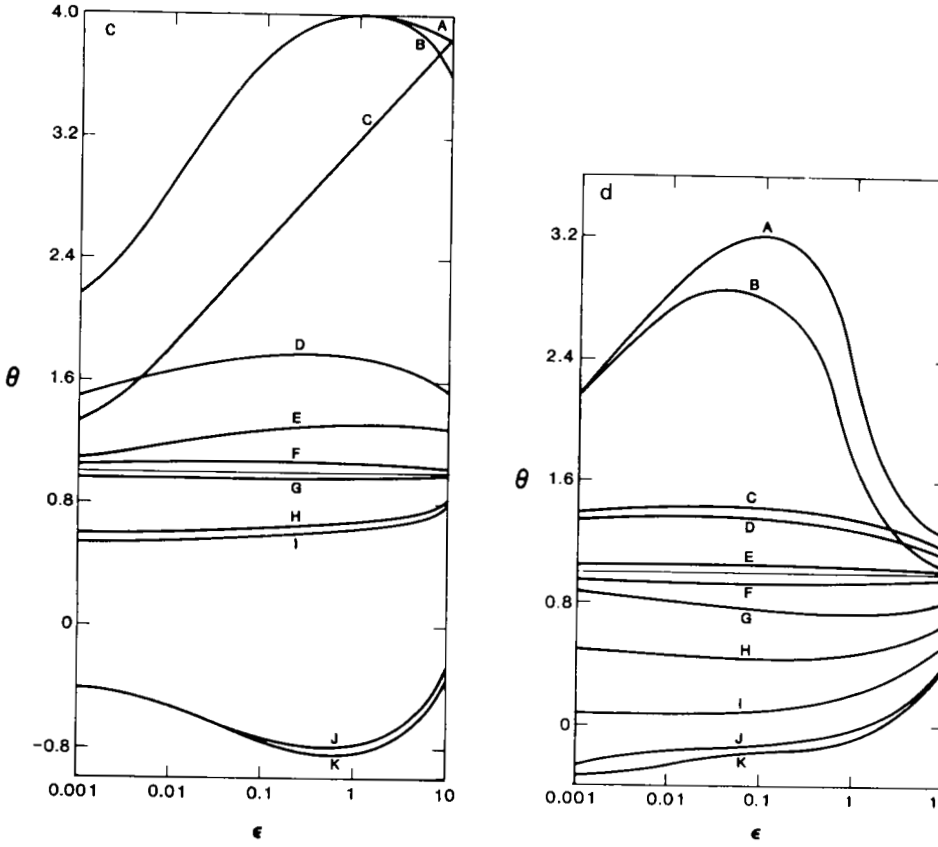

FIGURE 2c. θ vs ε for $C_A^*|_{\chi=1}$ = 0.1, K = 5, α = 100, with A) E_f^* = 1, E_r^* = 10; B) E_f^* = 0.001, 0.01, 0.1, E_r^* = 10; C) E_f^* = 10, E_r^* = 10; D) E_f^* = 0.001, 0.01, 0.1, E_r^* = 1; E) E_f^* = 1, E_r^* = 1; F) E_f^* = 0.001, 0.01, 0.1, E_r^* = 0.1; G) E_f^* = 0.1, E_r^* = 0.001, 0.01; H) E_f^* = 1, E_r^* = 0.1; I) E_f^* = 1, E_r^* = 0.01; J) E_f^* = 10, E_r^* = 0.001, 0.01, 0.1; K) E_f^* = 10, E_r^* = 1.

FIGURE 2d. θ vs ε for $C_A^*|_{\chi=1}$ = 0.1, K = 5, α = 100, with A) E_f^* = -10, E_r^* = -0.001, -0.01, -0.1; B) E_f^* = -10, E_r^* = -1; C) E_f^* = -1, E_r^* = -0.001, -0.01; D) E_f^* = -1, E_r^* = -0.1; E) E_f^* = -0.1, E_r^* = -0.001, -0.01; F) E_f^* = -0.001, -0.01, -0.1, E_r^* = -0.1; G) E_f^* = -1, E_r^* = -1; H) E_f^* = -0.001, -0.01, -0.1, E_r^* = -1; I) E_f^* = -10, E_r^* = -10; J) E_f^* = -1, E_r^* = -10; K) E_f^* = -0.001, -0.01, -0.1, E_r^* = -10.

times and the system approaches reaction equilibrium. A large value of ε corresponds to rapid diffusion times and there is very little reaction taking place. In both limits of very small and very large ε, Θ should approach 1 if both E_f^* and E_r^* are small. For large ε, there is very little reaction and the facilitation is close to 1 and unaffected by changes in the kinetic rate constants. For very small ε, the system is at reaction equilibrium and the facilitation is controlled by diffusion.

K is a dimensionless equilibrium constant. Kemena, Noble, and Kemp (18) determined that the optimal value of K was between 1 and 10 for large variations in α and ε. Over this range, the effect of K on Θ was small.

When the permeate concentration at X = 1 is non-zero, the temperature effect can have some interesting results. Figures 2a through 2d show that Θ can become negative. This situation corresponds to a reversal in the direction of the permeate flux. This reversal is due to the fact that the reverse reaction is very large at X = 0 and the forward reaction is very large at X = 1. This effect is noted by Verhoff et al. (19) who assumed that the chemical reaction is near equilibrium to obtain an analytical solution.

The solution for Θ is presented graphically. This allows one to estimate the effect of the temperature gradient on the total facilitated flux. The isothermal facilitation factor can be estimated analytically (20, 21) or determined graphically (11). At present, there is no data available in the open literature for comparison with the model results.

CONCLUSIONS

The effect of a temperature gradient on the permeate flux through a liquid film in facilitated transport has been studied. A dimensionless number Θ has been defined which is the ratio of the total facilitated flux under non-isothermal conditions to the total facilitated flux under isothermal conditions. Θ provides a measure of the temperature effect on facilitated transport.

The major effect of the temperature gradient is the change in the reaction rate constants with temperature. It has been shown that Θ can be increased or decreased significantly from 1 depending on the physical properties and operating conditions of the system. This result means that temperature effects should not be ignored, even for small temperature differences across the liquid film.

The solution for Θ is presented graphically. This allows one to estimate the effect of the temperature gradient on the facilitated factor.

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