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Facilitated Transport through a Liquid Surfactant Membrane with Continuous Phase Resistance: Role of Drop-Size Distribution

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ABSTRACT

An advancing reaction front model with drop-size distribution has been proposed for the case of facilitated transport through a liquid surfactant membrane. The model takes into account the continuous phase and outer liquid membrane phase resistances along with diffusion through a composite emulsion drop. The computed results are found to be in excellent agreement with the experimental data of Ho et al. The outer liquid membrane phase resistance seems to be an important parameter in controlling the overall extraction rate. Analytical solutions to model equations for uniform drop size are also presented in this study. The model is also capable of predicting the effect of various parameters on the overall extraction rate for the limiting cases of inverse Biot modulus tending to zero and infinity.

INTRODUCTION

The use of liquid surfactant membranes in separation science has been an area of active research since its discovery by Li (2). Various types of liquid membranes and the mechanism of mass transfer to and from liquid membrane emulsion drops have been reported by a number of workers. In one variety of liquid membrane system, the solute to be removed from the continuous phase diffuses through the membrane phase to the encapsulated phase. The encapsulated phase contains a reagent which reacts with the solute and forms a product incapable of permeating through the membrane phase. The transport of solute is thereby facilitated because of the maximum concentration difference available across the membrane phase. In general, emulsion drops having diameters of about 0.1–2 mm are large

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compared to those of the encapsulated phase which are usually 1–10 μm in diameter.

During the last few years a number of mathematical models have been developed to describe mass transfer through a liquid surfactant membrane. The existing models can be classified broadly into two distinct categories: 1) membrane film models in which practically all the resistance to mass transfer is assumed to lie in a thin membrane film of constant thickness surrounding the liquid membrane emulsion drop (3–13), and 2) distributed resistance models which consider the diffusional resistance to be distributed throughout the emulsion drop (1, 14–19).

A mechanism of facilitated transport for the case of a liquid membrane system was first suggested by Kopp et al. (20) who used an advancing reaction front model. Ho et al. (1) improved the model significantly by assuming a diffusion-controlled mass transfer mechanism for emulsion drops of uniform size and solving unsteady-state transport in a spherical geometry with changes in the external phase concentration. The continuous phase mass transfer resistance was considered to be negligible in their model. They showed that for all practical applications, the solution to their model with a pseudosteady-state assumption was sufficient to predict the extraction rates. Stroeve and Varanasi (16) extended the model of Ho et al. (1) by including additional resistance in the continuous phase. Kim et al. (15) also developed an unsteady-state model with this approach by including an additional thin outer liquid membrane layer. Continuous phase resistance was neglected in their model, but the solute removal rate was not directly related to the shrinking rate in their model. As expected, their model successfully predicts the experimental results during the initial period of extraction since they use the thickness of the membrane film as an adjustable parameter. Teramoto et al. (21) and Bunge and Noble (22) also developed models for facilitated transport in which they include the assumption of reaction equilibrium, and therefore they did not consider the concept of a reaction front in their models.

However, all the advancing reaction front models developed so far fall short of describing the overall transport mechanism underlying such a system. In the model developed by Ho et al. (1), there is a definite trend in the deviation of their theoretical prediction from experimental values, particularly at longer extraction times. In fact, advancing front models overpredict solute removal rate. The discrepancy between theoretical and experimental results is greater at longer times. The same is expected to be true when the mass ratio of internal reagent to bulk solute is small. This discrepancy has been successfully eliminated in the present model of the role of polydispersity in the time course of extraction by incorporating a drop-size distribution. The inverse Biot modulus, which is a function of drop size, varies with polydispersity and is not a constant in the present model.

Stroeve and Varanasi (16) did not consider the effect of other important parameters, such as diffusivity in the oil phase, D_o, and the distribution coefficient, α, on the extraction rate, particularly when the resistance offered by the continuous phase is negligible and the emulsion drop resistance therefore controls the extraction process.

The present model includes an additional thin outer liquid membrane layer surrounding the emulsion drop. That layer contains no internal reagent. Our model successfully predicts the effect of various parameters on different mass transfer resistances and consequently the extraction rate for the two limiting cases of continuous phase resistance tending to zero and infinity. Membrane leakage is assumed to be negligible.

MATHEMATICAL MODEL

Figure 1 gives a schematic picture of the present model. Unlike Ho et al. (1) and Stroeve and Varanasi (16), the present study describes the permeation mechanism with the help of a combination of a membrane film and the distributed resistance model by assuming an additional thin outer liquid membrane layer that contains no internal droplets. The solute from the continuous phase first diffuses through the outer organic liquid membrane phase of thickness ($R - R_c$) and then through the reacted inner core to the reaction front where it is consumed by the internal reagent in an irreversible and instantaneous reaction. As the reagent is consumed, the reaction front advances and the radius of the unreacted inner core (R_f) shrinks. At the surface of the unreacted inner core, the concentration of the solute is zero.

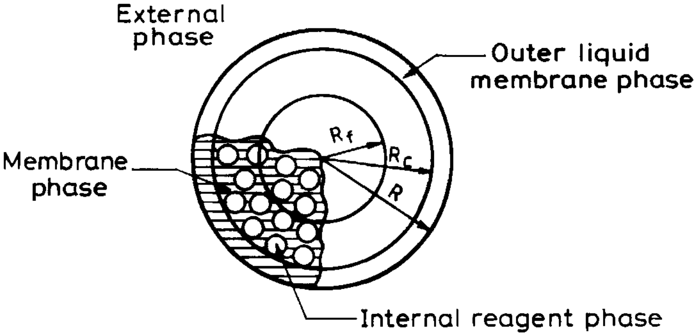

FIG. 1 A schematic diagram of a liquid membrane emulsion drop.

Assuming pseudosteady-state diffusion, Fick's first law of diffusion gives the rate of solute permeation through the outer liquid membrane of organic phase as

$$N = 4\pi r^2 D_o \frac{dc_m}{dr}, \qquad \text{for } R_c < r < R \tag{1}$$

Integration of Eq. (1) between the limits $r = R_c$ and $r = R$ gives

$$C_{ms} - C_{mc} = \frac{N}{4\pi D_o}\left(\frac{1}{R_c} - \frac{1}{R}\right) \tag{2}$$

The equation describing the rate of solute permeation through the reacted inner core is

$$N = 4\pi r^2 D'_{eff} \frac{dc_m}{dr}, \qquad \text{for } R_f < r < R_c \tag{3}$$

where

$$D'_{eff} = D_{eff}\left(\frac{V_i/\alpha + V_m}{V_i + V_m}\right)$$

Here D'_{eff} is the effective diffusivity based on the concentration driving force defined in terms of the membrane phase concentration, C_m, and D_{eff} is the effective diffusivity based on the average concentration in the emulsion mixture.

Integrating Eq. (3) between the limits $r = R_f$ and $r = R_c$, we get

$$C_{mc} = \frac{N}{4\pi D'_{eff}}\left(\frac{1}{R_f} - \frac{1}{R_c}\right) \tag{4}$$

The equation describing the rate of solute extraction in the outer aqueous continuous phase is

$$N = k_e 4\pi R^2 (C_e - C_s) \tag{5}$$

where

$$C_{ms} = \alpha C_s$$

Combination of Eqs. (2), (4), and (5) gives

$$C_e = \frac{N}{4\pi R^2}\left[\frac{1}{k_e} + \frac{R}{R_c}\frac{(R - R_c)}{D_o \alpha} + \frac{R}{D'_{eff}\alpha}\left(\frac{R}{R_f} - \frac{R}{R_c}\right)\right]$$

or

$$C_e = \frac{N}{4\pi R^2}\left[\frac{1}{K} + \frac{R}{D'_{eff}\alpha}\left(\frac{R}{R_f} - \frac{R}{R_c}\right)\right] \tag{6}$$

where

$$\frac{1}{K} = \frac{1}{k_e} + \frac{R}{R_c}\frac{(R - R_c)}{D_o\alpha}$$

is the sum of the continuous phase resistance and the outer liquid membrane phase resistance.

A material balance over the reaction front yields

$$-\frac{d}{dt}\left[\frac{4}{3}\pi R_f^3\left(\frac{V_i}{V_i + V_m}\right)C_{i0}\right] = 4\pi R_f^2 D'_{eff}\frac{dC_m}{dr} \tag{7}$$

Equation (7) gives the rate of change of the advancing front radius as

$$\frac{dR_f}{dt} = -\frac{N}{4\pi R_f^2 C_{i0}\left(\frac{V_i}{V_i + V_m}\right)} \tag{8}$$

The total amount of solute extracted from the aqueous continuous phase is

$$-V_e\frac{dC_e}{dt} = nN \tag{9}$$

If n_j and $f(R_j)$ are, respectively, the number and volume fraction of drops belonging to size range j, then

$$n_j = \frac{(V_i + V_m)f(R_j)}{\frac{4}{3}\pi R_j^3} \tag{10}$$

For a drop of size j, Eq. (8) becomes

$$\frac{dR_{fj}}{dt} = -\frac{N_j}{4\pi R_{fj}^2 C_{i0}\left(\frac{V_i}{V_i + V_m}\right)} \tag{11}$$

and Eq. (9) becomes

$$-V_e\frac{dC_e}{dt} = \sum_j n_j N_j \tag{12}$$

The equations can be nondimensionalized by defining the following dimentionless variables:

$$\chi = R_f/R, \quad h = C_e/C_{e0}, \quad \beta = R/R_c, \quad \tau = \epsilon D'_{eff}t/R_m^2$$
$$\epsilon = \frac{\alpha C_{e0}}{\left(\frac{V_i}{V_i + V_m}\right)C_{i0}}, \quad E = 3\left(\frac{V_i C_{i0}}{V_e C_{e0}}\right), \quad m = \frac{D'_{eff}\alpha}{KR} \tag{13}$$

The dimensionless equations representing the liquid membrane system are

$$\frac{d\chi_j}{d\tau} = -\frac{hR_m^2}{R_j^2\chi_j(m_j\chi_j + 1 - \beta\chi_j)} \tag{14}$$

and

$$\frac{dh}{d\tau} = -hER_m^2\sum_j\frac{f(R_j)\chi_j}{R_j^2(m_j\chi_j + 1 - \beta\chi_j)} \tag{15}$$

with initial conditions:

$$\text{at } \tau = 0, \quad \chi_j = 1/\beta \tag{16}$$

and

$$\text{at } \tau = 0, \quad h = 1 \tag{17}$$

where R_m is the characteristic radius of all drops and is taken as the Sauter mean radius, R_m, in the model. The inverse Biot modulus, m, in this model is the ratio of the sum of the continuous phase resistance and the outer liquid membrane phase resistance to the reacted inner core resistance. The parameter β accounts for the thickness of the outer liquid membrane phase. The equations for the model were numerically solved on a computer by using a fourth-order Runge–Kutta method for the initial value problem.

The above equations can be simplified by using some average drop size. In the present model, the Sauter mean diameter was used to characterize the drop size. Equations (14) and (15) can be rewritten for the average drop size as follows

$$\frac{d\chi}{d\tau} = -\frac{h}{\chi(m\chi + 1 - \beta\chi)} \tag{18}$$

and

$$\frac{dh}{d\tau} = -\frac{hE\chi}{(m\chi + 1 - \beta\chi)} \tag{19}$$

Analytical solutions to the coupled Eqs. (18) and (19) can be obtained for h and τ. They are

$$h = 1 + E/3(\chi^3 - 1/\beta^3) \tag{20}$$

$$= \frac{1}{E}(\beta - m)\ln\frac{\chi^3 + a^3}{a^3 + 1/\beta^3} + \frac{1}{2aE}\left(3\ln\left(\frac{\chi + a}{a + 1/\beta}\right) - \ln\frac{\chi^3 + a^3}{a^3 + 1/\beta^3}\right)$$
$$- \frac{\sqrt{3}}{aE}\left(\tan^{-1}\frac{2\chi - a}{\sqrt{3}a} - \tan^{-1}\frac{2/\beta - a}{\sqrt{3}a}\right) \tag{21}$$

where

$$a = \left(\frac{3 - E/\beta^3}{E}\right)^{1/3}$$

RESULTS AND DISCUSSION

Computer simulations of Eqs. (14) and (15) were performed after selecting a suitable drop-size distribution. The drop-size distribution was selected with diameters in the range of 0.2 to 2 mm so that its average size (Sauter mean diameter) became the same as that found by Ho et al. (1). Table 1 gives the distribution of the sizes of the emulsion drops dispersed in the external continuous aqueous phase at 400 and 600 rpm. The operating conditions for the extraction runs and the estimated values of the various parameters of Ho et al. (1) and those used in the present model are given in Table 2.

TABLE 1
Drop Size Distributions of W/O Emulsion Used for Simulation at 400 and 600 rpm

	Volume fraction	
Drop radius (mm)	For 400 rpm	For 600 rpm
0.1	0.005	0.0006
0.2	0.031	0.1570
0.3	0.086	0.5635
0.4	0.155	0.2404
0.5	0.190	0.0140
0.6	0.196	0.0072
0.7	0.130	0.0057
0.8	0.077	0.0042
0.9	0.055	0.0033
1.0	0.075	0.0041

TABLE 2
Operating Conditions and Parameters for the Extraction of Phenol

	Experimental, Ho et al. (1)	Simulation, present model
Type of separation	Batch	Batch
Agitation speed, N, rpm	400	400
	600	600
Sauter mean radius, R_m, m	5×10^{-4}	5×10^{-4}
	3×10^{-4}	3×10^{-4}
Distribution coefficient, α	0.52	0.1
		0.52
		1.0
Dimensionless parameter, E	3.32	3.32, 2.75, 3, 5, 6
Diffusivity in organic membrane phase, D_0, m²/s	0.65×10^{-10}	0.65×10^{-10}
		2.00×10^{-10}
		5.00×10^{-10}
Diffusivity in internal aqueous phase, D_w, m²/s	9.98×10^{-10}	9.98×10^{-10}
Effective diffusivity, D'_{eff}, m²/s	—	1.64×10^{-10}
Continuous phase mass transfer coefficient, k_e, m/s	—	4.27×10^{-5}
		9.13×10^{-5}
		1.00×10^{-6}

The effective diffusivity, D'_{eff}, was calculated by the method discussed by Ho et al. (1). The value of the continuous phase mass transfer coefficient, k_e, was calculated from the correlation of Skelland and Lee (23) which corresponds to an impeller Reynolds number of 1.71×10^4 (400 rpm) and 2.58×10^4 (600 rpm). The impeller Reynolds number was, in turn, calculated for the standard mixer used by Ho et al. (1) and at stirrer speeds of 400 and 600 rpm.

Effect of Drop Size Distribution

Figure 2 shows the predictions of the present model as well as the theoretical and experimental results of Ho et al. (1) for batch extraction of phenol containing NaOH as the reagent by taking the value of β equal to 1. When the value of β is 1, i.e., a zero outer liquid membrane thickness, the schematic representation of our liquid membrane system reduces to that of Ho et al. (1). The present model is in better agreement with the experimental results of Ho et al. (1) than is their theory. Figure 3 shows the reaction front position for different sized drops at 400 rpm as a function of dimensionless time. It is seen from Fig. 3 that the reaction front penetration is small for larger drops, while for smaller drops the movement of the reaction front is very fast and most of the phenol is consumed in

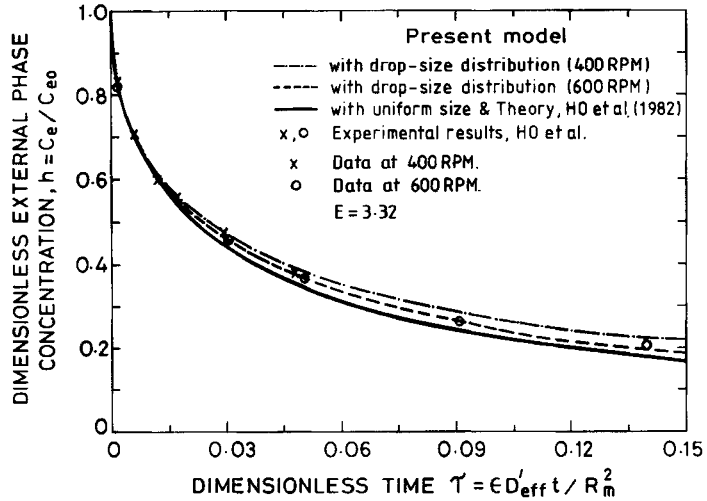

FIG. 2 Comparison of the model predictions with theory and experimental results of Ho et al. (1) for phenol extraction runs.

the early stage. Afterward there is virtually simple physical absorption of phenol, and these smaller drops are no longer effective in extracting the solute, which results in a loss of effective interfacial area for solute transport. It is therefore clear that this discrepancy, which is particularly observed for the advancing front models at longer times, is due to neglect of the polydisperse character of the dispersed emulsion phase. It has also been found that the present model's prediction closely resembles that of the Bunge and Noble (22) model which also eliminated this discrepancy between the theoretical and experimental results. However, Bunge and Noble allowed reaction reversibility in the aqueous phase in their model.

The effect of the drop size distribution or the polydisperse character of the dispersed emulsion phase is not significant, as shown in Fig. 2. This is also in conformity with the predictions of Teramoto et al. (21) and Lorbach and Hatton (24). This is due to the fact that increasing the drop radius, R, decreases the value of m, which leads to an increase in the rate of extraction. However, the interfacial area also decreases with increasing R, thereby balancing the above effect. As seen from Fig. 2, the extraction rate is high until the total interfacial area is available for extracting the solute. With increasing contact time, the smaller drops are depleted from the internal reagent. However, the larger drops are not very effective in extracting the solute because of their smaller specific surface area. Moreover, since larger drops of various sizes offer almost the same resistance

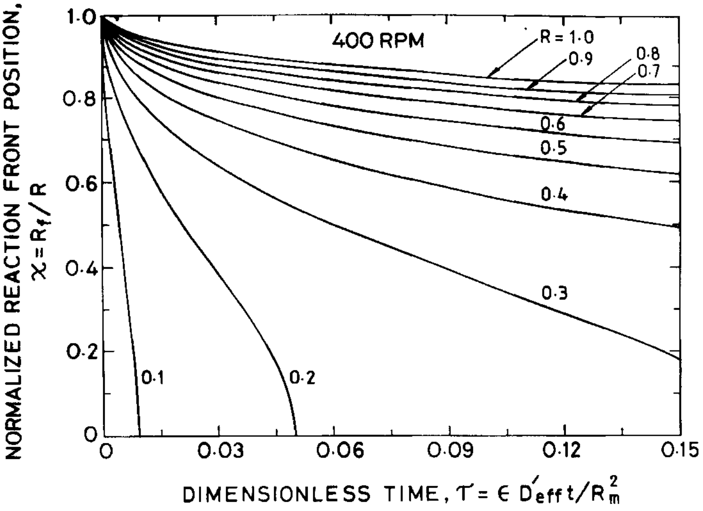

FIG. 3 Effect of R on the progress of the reaction front at a stirrer speed of 400 rpm.

due to short penetration, it is expected that the polydisperse effect will have very little influence on mass transfer behavior.

Figure 2 also shows the present model's prediction for emulsion drops of uniform size. The Sauter mean diameter is taken as the average sizes of the emulsion drops in this model, just as for Ho et al.'s model (1). The comparison of our theory for uniform size with that of Ho et al. (1) is excellent. Our model for uniform size also predicts somewhat higher removal rates than was observed experimentally.

Effect of Outer Liquid Membrane Thickness

The outer liquid membrane phase offers significant resistance to the extraction rate, as observed in Fig. 4. For a small change in the thickness of the outer liquid membrane phase, i.e., in the value of β, the extraction rate changes appreciably. This change is due to the small values of D_o/D_w and α. The outer liquid membrane resistance thus seems to be a very sensitive parameter in controlling the overall extraction rate.

Effect of *E*

Figure 5 shows the change in the extraction rate for various values of E. Increasing E increases the amount of reagent in the emulsion drop so that the reaction front penetrates more slowly. The slower penetration leads to a higher extraction rate because of the shorter diffusion path of the solute needed to reach the reaction front.

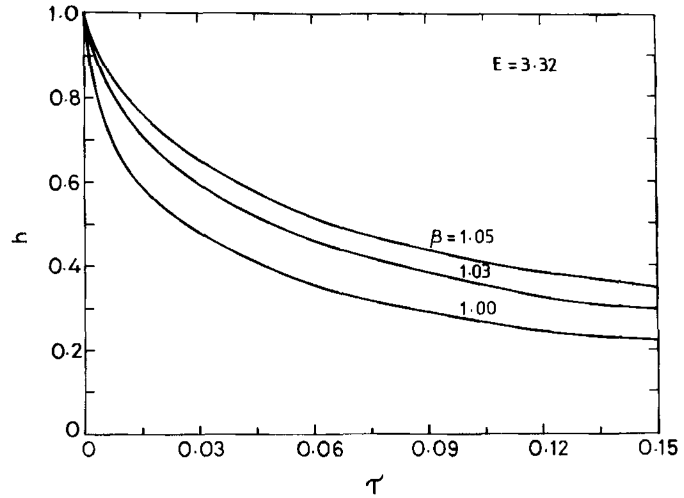

FIG. 4 Variation of dimensionless external phase concentration with τ; effect of β for $E = 3.32$.

For large values of E, i.e., either a high concentration of the internal reagent or a low bulk solute concentration, a significant proportion of the internal reagent remains unreacted in most of the different sized drops, and therefore the effect of drop-size distribution on the extraction rate is small. But when the value of E is small, the internal reagent is likely to be totally consumed by solute, particularly in smaller drops, and the available interfacial area is greatly reduced. The discrepancy between the two models therefore increases with an increase in contact time (Fig. 5). In this case our model, which takes the polydisperse effect into account, correctly predicts the extraction rate.

Role of Different Resistances on the Extraction Rate

The role of different resistances on mass transfer behavior for this system is shown in Fig. 6 for $\beta = 1.003$. When the resistance of the continuous phase is small compared to that of the outer liquid membrane phase, the inverse Biot modulus becomes the ratio of the outer liquid membrane phase resistance to the reacted core resistance. For such a condition, the rate of the solute diffusional rate through the outer liquid membrane phase increases as D_o increases. On the other hand, increasing D_o also increases the values of the effective diffusivity. D'_{eff}, which leads to a faster penetration of the reaction front and therefore a decrease in the rate of extraction. Furthermore, for no change or a negligible change in D_o, a change in the value of α leads to a small change in the value of D'_{eff}. Thus, there

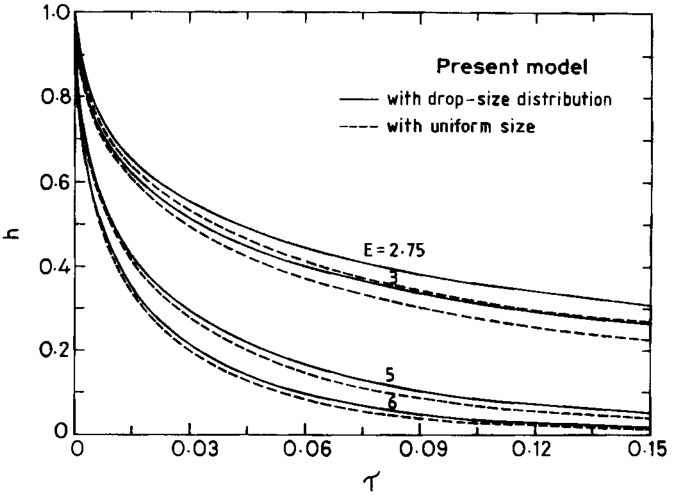

FIG. 5 Variation of dimensionless external phase concentration with τ; effect of E for $\beta = 1$.

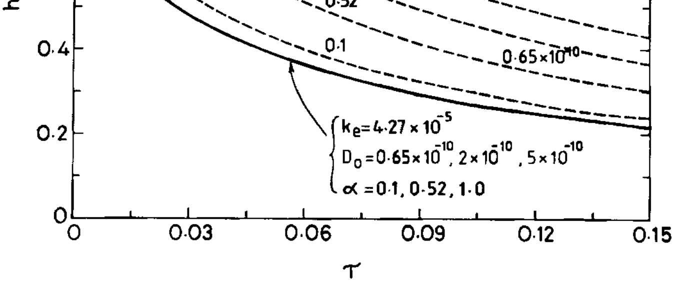

FIG. 6 Variation of dimensionless external phase concentration with τ for $\beta = 1.003$. (——) Effect of D_0 (at $\alpha = 0.52$) and α (at $D_0 = 0.65 \times 10^{-10}$) for $k_e = 4.27 \times 10^{-5}$. (- -) Effect of D_0 (at $\alpha = 0.52$) and α (at $D_0 = 0.65 \times 10^{-10}$) for $k_e = 1 \times 10^{-6}$.

is practically no change in the extraction rate for a change in D_o or in α (Fig. 6). Computed values are also presented in Fig. 6 for different values of D_o and α when the resistance of the continuous phase is appreciable ($k_e = 1 \times 10^{-6}$ m/s) and therefore cannot be neglected. In this case, the inverse Biot modulus becomes the ratio of continuous phase resistance to the reacted core resistance. Here it is observed that for a change in the value of D_o or α, the continuous phase resistance is not affected but the reacted core resistance changes. Consequently, the time for a given extent of extraction changes due to the change in the value of m.

The present model, which uses an adjustable parameter, β, thus provides better understanding of the mechanism of transport and is particularly useful in the design of continuous extraction columns where the effect of polydispersity is important.

NOMENCLATURE

C_m	solute concentration in the membrane phase of an emulsion drop in equivalents per mole
C_{ms}	solute concentration in the membrane phase at the surface of an emulsion drop
C_{mc}	solute concentration in the membrane phase at the inner core of an emulsion drop
C_s	solute concentration in the external continuous phase at the surface of an emulsion drop
C_{e0}	initial solute concentration in the external continuous phase
C_{i0}	initial concentration of reagent in the reagent phase
D_o	diffusivity of solute in the organic phase
D_w	diffusivity of solute in the aqueous phase
D'_{eff}	effective solute diffusivity in the reacted inner core of the emulsion drop
E	dimensionless parameter in the present model
$f(R_j)$	volume fraction of drops of size j
h	normalized solute concentration in the external continuous phase
k_e	continuous phase mass transfer coefficient
m	inverse Biot modulus
n	total number of emulsion drops dispersed in the external continuous phase
N	mass transfer rate of solute
r	radial coordinate
R	emulsion drop radius
R_c	initial radius of the unreacted inner core
R_f	reaction front position

R_m	Sauter mean radius
t	time
V_e	external phase volume
V_i	total internal reagent volume
V_m	total membrane phase volume

Greek Letters

α	distribution coefficient for the solute between the external and membrane phases at equilibrium
β	dimensionless parameter that accounts for the outer liquid membrane thickness
ϵ	dimensionless parameter in the present model
τ	dimensionless time
χ	normalized reaction front

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