Sorption Kinetics for Asymmetric Binary and Ternary Laminate Slabs in Finite and Semi-Infinite Well-Stirred Baths

H. GARTH SPENCER, *Department of Chemistry, Clemson University, Clemson, South Carolina 29631*, and JAMES A. BARRIE, *Department of Chemistry, Imperial College of Science and Technology, SW7 2AY, England*

Synopsis

Equations describing the sorption kinetics of asymmetric membranes composed of AB and ABC laminate slabs in finite and in semi-infinite well-stirred baths are presented, and some properties of these systems are discussed.

INTRODUCTION

Equations have been developed for diffusion from a well-stirred finite bath into a homogeneous slab[1–3] which can be used to evaluate diffusion and solubility coefficients[4–6]; this work has been extended to include symmetric ABA laminate slabs.[7] Equations for diffusion from a well-stirred semi-infinite bath into a homogeneous slab, a symmetric ABA laminate slab, and an asymmetric AB laminate slab are also available.[2,8] The purpose of this paper is to extend the development to describe sorption kinetics for the AB laminate slab in a well-stirred finite bath and for the ABC laminate slab in both a well-stirred finite bath and a well-stirred semi-infinite bath.

DIFFUSION EQUATIONS

AB Laminate in a Finite Bath

The membrane is a slab comprising lamina A of thickness a and lamina B of thickness b in perfect contact at $x = 0$, where x is the distance coordinate normal to the interface. The solute concentration c in the well-stirred bath of volume V is initially c^0. The initial concentrations in the layers A and B are respectively C_A^i and C_B^i and are uniform corresponding to equilibrium with a bath concentration c^i. Equilibrium is maintained at each of the interfaces according to the following relations: $C_A = cK_A$ at $x = -a$; $C_A = C_B K$ at $x = 0$; and $C_B = cK_B$ at $x = b$; where K_A, K, and K_B are constants. The diffusion coefficients for the respective laminae are D_A and D_B.

The differential equations describing the diffusional transport are

$$\frac{\partial C_A}{\partial t} = D_A \frac{\partial^2 C_A}{\partial x^2} \qquad -a < x < 0$$
$$\frac{\partial C_B}{\partial t} = D_B \frac{\partial^2 C_B}{\partial x^2} \qquad 0 < x < b \tag{1}$$

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The initial and boundary conditions for the system are

$$C_A(-a,0) = C_A^0 \qquad C_B(b,0) = C_B^0$$

$$C_A(x,0) = C_A^i, -a < x \leqslant 0 \qquad C_B(x,0) = C_B^i, 0 \leqslant x < b \tag{2}$$

$$D_A \left(\frac{\partial C_A}{\partial x}\right)_{x=0} = D_B \left(\frac{\partial C_B}{\partial x}\right)_{x=0} \qquad t \geqslant 0 \tag{3}$$

$$K = \frac{K_A}{K_B} = \frac{C_A(0,t)}{C_B(0,t)} = \frac{C_A(-a,t)}{C_B(b,t)} \qquad t \geqslant 0 \tag{4}$$

$$\delta^2 \left(\frac{\partial C_A}{\partial x}\right)_{x=-a} - \left(\frac{\partial C_B}{\partial x}\right)_{x=b} = \frac{b}{H_B D_B}\left(\frac{\partial C_B}{\partial t}\right)_{x=b} \tag{5}$$

where $\delta^2 = D_A/D_B$, $H_B = K_B V_B/V$, and V_B is the volume of lamina B.

Application of the Laplace transform method[9] provides the solution to the set of eqs. (1)–(5) which can be expressed as

$$C_I(x,t) = C_I^f + (C_I^f - C_I^i) \sum_{n=1}^{\infty} I_n(x) \exp(-D_B R_n^2 t/b^2) \tag{6}$$

with I = A or B. The concentration of diffusant in the laminae at equilibrium C_I^f is given by

$$C_I^f = \frac{C_I^0 + C_I^i(H_A + H_B)}{1 + H_A + H_B} \tag{7}$$

with $H_A = K_A V_A/V = \lambda K H_B$. Also,

$$A_n(x) = 2\{(\cos\alpha_n - \cos R_n)\sin(\alpha_n x/a) + (\sin\alpha_n + \delta K \sin R_n)\cos(\alpha_n x/a)\}/U_n \tag{8}$$

$$B_n(x) = 2\{\delta K(\cos\alpha_n - \cos R_n)\sin(R_n x/b) + (\sin\alpha_n + \delta K\sin R_n)\cos(R_n x/b)\}/U_n \tag{9}$$

$$\begin{aligned} U_n = {}& \delta K \Theta_n [(\lambda/\delta)\sin\alpha_n - \sin R_n] - \delta K \Theta_n'(\cos\alpha_n - \cos R_n) \\ & + \Phi_n[(\lambda/\delta)\cos\alpha_n + \delta K \cos R_n] \\ & + \Phi_n'(\sin\alpha_n + \delta K \sin R_n) \\ & + \delta K[(H_A + H_B)\sin R_n \cos\alpha_n + (\delta K H_B + H_A/\delta K)\cos R_n \sin\alpha_n] \end{aligned} \tag{10}$$

where

$$\begin{aligned} \alpha_n &= R_n \lambda/\delta \\ \Theta_n &= H_B \cos R_n - R_n \sin R_n \\ \Theta_n' &= -\Phi_n - \sin R_n \\ \Phi_n &= H_B \sin R_n + R_n \cos R_n \\ \Phi_n' &= \Theta_n + \cos R_n \\ \lambda &= a/b = V_A/V_B \end{aligned} \tag{11}$$

The coefficients R_n are the nonzero positive roots of

$$\delta K\Theta (\cos \alpha - \cos R) - \Phi (\sin \alpha + \delta K \sin R) + \delta K H_B (\cos \alpha \cos R - \delta K \sin \alpha \sin R - 1) = 0 \quad (12)$$

The quantity usually measured is the concentration of diffusant in the finite bath; using $cK_A = C_A$ and solving eq. (6) for $x = -a$, one obtains

$$c(t) = c^f + (c^0 - c^i) \sum_{n=1}^{\infty} Z_n \exp(-D_B R_n^2 t/b^2) \quad (13)$$

where

$$Z_n = 2(\delta K \sin R_n \cos \alpha_n + \cos R_n \sin \alpha_n)/U_n \quad (14)$$

The fractional change of the diffusant concentration remaining in the bath is given by

$$M(t) = \frac{c(t) - c^f}{c^0 - c^f} = \sum_{n=1}^{\infty} X_n \exp(-D_B R_n^2 t/b^2) \quad (15)$$

with

$$X_n = Z_n(1 + H_A + H_B)/(H_A + H_B) \quad (16)$$

At large t, the first term in eq. (15) dominates, and the expression reduces to

$$\ln M(t) = \ln X_1 - D_B R_1^2 t/b^2 \quad (17)$$

ABC Laminate in a Finite Bath

The slab of thickness $(a + l)$ comprises three laminae, A, B, and C, of thickness a, b, and $l - b$, respectively. Perfect contact is maintained between laminae A and B at $x = 0$ and between B and C at $x = b$. Initially the slab is in equilibrium with a well-stirred finite bath of volume V with diffusant concentrations of c^i, C_A^i, C_B^i, and C_C^i in the bath and laminae A, B, and C, respectively. To initiate the experiment, the bath concentration is changed from c^i to c^0 at $t = 0$. Equilibrium is maintained at each phase interface throughout, described by $cK_A = C_A$ at $x = -a$; $C_A = C_B K$ at $x = 0$; $C_C = C_B K^*$ at $x = b$; and $cK_C = C_C$ at $x = l$; also $cK_B = C_B$. The diffusion coefficients D_A, D_B, and D_C in the respective laminae are constant.

The differential equations describing transport are given by eq. (1) with the additional equation

$$\frac{\partial C_C}{\partial t} = D_C \frac{\partial^2 C_C}{\partial x^2} \qquad b < x < l \quad (18)$$

The initial and boundary conditions are

$$C_A(-a,0) = C_A^0 \qquad C_C(l,0) = C_C^0$$

$$C_A(x,0) = C_A^i \qquad -a < x \leqslant 0$$

$$C_B(x,0) = C_B^i \qquad 0 \leqslant x \leqslant b$$

$$C_C(x,0) = C_C^i \qquad b \leqslant x < l \quad (19)$$

$$C_A(0,t) = KC_B(0,t) \qquad C_C(b,t) = K^* C_B(b,t)$$

$$C_A(-a,t)/C_C(l,t) = K_A/K_C = K/K^* \qquad t \geqslant 0 \quad (20)$$

$$D_A\left(\frac{\partial C_A}{\partial x}\right)_{x=0} = D_B\left(\frac{\partial C_B}{\partial x}\right)_{x=0}$$

$$D_B\left(\frac{\partial C_B}{\partial x}\right)_{x=b} = D_C\left(\frac{\partial C_C}{\partial x}\right)_{x=b}$$

$$\left(\frac{\delta}{\delta^*}\right)^2\left(\frac{\partial C_A}{\partial x}\right)_{x=-a} - \left(\frac{\partial C_C}{\partial x}\right)_{x=l} = \frac{b}{KD_C H_B}\left(\frac{\partial C_A}{\partial t}\right)_{x=-a} \qquad t \geqslant 0 \tag{21}$$

with $\delta^2 = D_A/D_B$ and $(\delta^*)^2 = D_C/D_B$.

Application of the Laplace transform method provides the solution to the problem in the form of eq. (6), where I now denotes A, B, or C and C_I^f. The final or equilibrium concentrations of diffusant in the laminae are given by

$$C_I^f = [C_I^0 + C_I^i (H_A + H_B + H_C)]/(1 + H_A + H_B + H_C) \tag{22}$$

with $H_I = K_I V_I/V$. Also,

$$A_n(x) = [F_A \sin(\alpha_n x/a) + G_A \cos(\alpha_n x/a)]/W_n \tag{23}$$

$$B_n(x) = [F_B \sin(R_n x/b) + G_B \cos(R_n x/b)]/W_n \tag{24}$$

$$C_n(x) = [F_C \sin(\beta_n^* x/l) + G_C \cos(\beta_n^* x/l)]/W_n \tag{25}$$

with $\lambda = a/b$, $\lambda^* = (l - b)/b$, $\alpha_n = R_n\lambda/\delta$, and $\beta_n^* = R_n(\lambda^* + 1)/\delta^*$. The remaining coefficients are given by

$$F_A = \sin R_n \sin \alpha_n^* - \delta^* K^* \cos R_n \cos \alpha_n^* + \delta^* K^* \cos \alpha_n \tag{26}$$

$$G_A = \delta K \delta^* K^* \sin R_n \cos \alpha_n^* + \delta K \cos R_n \sin \alpha_n^* + \delta^* K^* \sin \alpha_n \tag{27}$$

$$F_B = \delta K F_A \qquad G_B = G_A \tag{28}$$

$$\begin{aligned} F_C = &-\delta K \cos \beta_n^* \\ &+ \delta K [\cos R_n \cos(R_n/\delta^*) + \delta^* K^* \sin R_n \sin(R_n/\delta^*)] \cos \alpha_n \\ &- [\sin R_n \cos(R_n/\delta^*) - \delta^* K^* \cos R_n \sin(R_n/\delta^*)] \sin \alpha_n \end{aligned} \tag{29}$$

$$\begin{aligned} G_C = &\ \delta K \sin \beta_n^* - \delta K [\cos R_n \sin(R_n/\delta^*) - \delta^* K^* \sin R_n \cos(R_n/\delta^*)] \\ &\times \cos \alpha_n + [\sin R_n \sin(R_n/\delta^*) + \delta^* K^* \cos R_n \cos(R_n/\delta^*)] \sin \alpha_n \end{aligned} \tag{30}$$

and

$$\begin{aligned} W_n = \{&-H_B \delta^* K^* (\delta^* K^* + K\lambda + \lambda^*/\delta^*) \sin R_n \sin \alpha_n \\ &+ H_B \delta^* K^* (\delta^* K^* \lambda/\delta + \delta K \delta^* K^* + \delta K \lambda^*/\delta^*) \cos R_n \cos \alpha_n \\ &- [\Delta_n' + \delta K (K^* \lambda^* + 1) \Gamma_n] \sin R_n \\ &+ [(K^* \lambda^* + 1) \Delta_n + \delta K \Gamma_n'] \cos R_n\} \sin \alpha_n^* \\ &+ \{H_B \delta^* K^* [\lambda/\delta + \delta K (K^* \lambda^* + 1)] \sin R_n \cos \alpha_n \\ &+ H_B \delta^* K^* (1 + K\lambda + \lambda^*/\delta^*) \cos R_n \sin \alpha_n \\ &+ [\delta K \delta^* K^* \Gamma_n' + (\delta^* K^* + \lambda^*/\delta^*) \Delta_n] \sin R_n \\ &+ [\delta^* K^* \Delta_n' + \delta K (\delta^* K^* + \lambda^*/\delta^*) \Gamma_n] \cos R_n\} \cos \alpha_n^* \\ &+ \delta^* K^* \{(\Gamma_n' - \Delta_n \lambda/\delta) \sin \alpha_n + (\Gamma_n \lambda/\delta - \Delta_n') \cos \alpha_n\} \end{aligned} \tag{31}$$

where

$$\alpha_n^* = R_n \lambda^*/\delta^*$$

$$\Delta_n = \delta K H_B \cos \alpha_n - R_n \sin \alpha_n$$

$$\Gamma_n = \delta K H_B \sin \alpha_n + R_n \cos \alpha_n \tag{32}$$

$$\Delta_n' = -\Gamma_n - \sin \alpha_n$$
$$\Gamma_n' = \Delta_n + \cos \alpha_n$$

and the R_n are the nonzero positive roots of

$$\begin{aligned}-\delta K^2 H_B \delta^* K^* - K\delta^* K^* (-\Gamma \sin \alpha + \Delta \cos \alpha) \\ -(KH_B \delta^{*2} K^{*2} \sin \alpha \cos R + K \Delta \sin R \\ + \delta K^2 H_B \delta^{*2} K^{*2} \cos \alpha \sin R \\ + \delta K^2 \Gamma \cos R) \sin \alpha^* \\ - (KH_B \delta^* K^* \sin \alpha \sin R - K\delta^* K^* \Delta \cos R \\ - \delta K^2 H_B \delta^* K^* \cos \alpha \cos R + \delta K^2 \delta^* K^* \Gamma \sin R) \cos \alpha^* = 0 \quad (33)\end{aligned}$$

Solving eq. (6) for $x = -a$ and using $cK_A = C_A$, one obtains eq. (13) for the concentration of diffusant in the bath with

$$\begin{aligned}Z_n = 2\{\delta^* K^* \cos (R_n \lambda^*/K^*) [\delta K \cos \alpha_n \sin R_n + \cos R_n \sin \alpha_n] \\ + \sin (R_n \lambda^*/K^*) [\delta K \cos R_n \cos \alpha_n - \sin R_n \sin \alpha_n]\}/W_n \quad (34)\end{aligned}$$

The reduced concentration in the bath is given by eq. (15) with

$$X_n = Z_n(1 + H_A + H_B + H_C)/(H_A + H_B + H_C) \quad (35)$$

As before, for large t eq. (15) reduces to eq. (17) with X_n given by eq. (35).

ABC Laminate in a Semi-Infinite Bath

The system differs from the previous case in that the finite bath is replaced by a semi-infinite bath so that the bath concentration remains constant at c^0, as do the concentrations in the two membrane surfaces in contact with the bath, i.e., $C_A(-a,t) = C_A^0$ and $C_C(l,t) = C_C^0$.

The diffusive transport is described by eqs. (1) and (18) with the following initial and boundary conditions:

$$\begin{aligned}C_A(x,0) &= C_A^i \qquad -a < x \leqslant 0 \\ C_B(x,0) &= C_B^i \qquad 0 \leqslant x \leqslant b \quad (36) \\ C_C(x,0) &= C_C^i \qquad b \leqslant x < l\end{aligned}$$

$$C_A(-a,t) = C_A^0 \qquad C_C(l,t) = C_C^0 \qquad t \geqslant 0 \quad (37)$$

$$C_A(0,t) = KC_B(0,t) \qquad C_C(b,t) = K^* C_B(b,t) \qquad t \geqslant 0 \quad (38)$$

$$D_A \left(\frac{\partial C_A}{\partial x}\right)_{x=0} = D_B \left(\frac{\partial C_B}{\partial x}\right)_{x=0} \qquad D_B \left(\frac{\partial C_B}{\partial x}\right)_{x=b} = D_C \left(\frac{\partial C_C}{\partial x}\right)_{x=b} \qquad t \geqslant 0 \quad (39)$$

The Laplace transform method gives the solutions

$$C_I(x,t) = C_I^0 + (C_I^0 - C_I^i) \sum_{n=1}^{\infty} I_n(x) \exp (-D_B R_n^2 t/b^2) \quad (40)$$

where I = A, B, or C and

$$\begin{aligned}A_n(x) = 2\{(-\Lambda_n \sin \beta_n^* - \Xi_n \cos \beta_n^* + \delta^* K^* \cos \alpha_n) \sin (\alpha_n x/a) \\ + (\delta K \Psi_n \sin \beta_n^* + \delta K \Omega_n \cos \beta_n^* + \delta^* K^* \sin \alpha_n) \cos (\alpha_n x/a)\}/S_n \quad (41)\end{aligned}$$

$$\begin{aligned}B_n(x) = 2 [(-\Lambda_n \sin \beta_n^* - \Xi_n \cos \beta_n^* + \delta^* K^* \cos \alpha_n) \delta K \sin (R_n x/b) \\ + (\delta K \Psi_n \sin \beta_n^* + \delta K \Omega_n \cos \beta_n^* + \delta^* K^* \sin \alpha_n) \cos (R_n x/b)]/S_n \quad (42)\end{aligned}$$

$$C_n(x) = 2\,[(\Lambda_n \sin \alpha_n + \delta K\, \Psi_n \cos \alpha_n - \delta K \cos \beta_n{}^*) \sin (\beta_n{}^* x/l) + (\Xi_n \sin \alpha_n + \delta K\, \Omega_n \cos \alpha_n + \delta K \sin \beta_n{}^*) \cos (\beta_n{}^* x/l)]/S_n \quad (43)$$

$$S_n = R_n\{[(\delta K + \lambda/\delta)\Lambda_n - (K\lambda^*\delta/\delta^*)\Omega_n] \cos \alpha_n \sin \beta_n{}^* + [(\lambda^* - 1)\Lambda_n/\delta^* - (\delta K + 1)\Omega_n] \sin \alpha_n \sin \beta_n{}^* + [(\delta K + \lambda/\delta)\Xi_n + (K\lambda^*\, \delta/\delta^*)\Psi_n] \cos \alpha_n \cos \beta_n{}^* \quad (44)$$

with

$$\Lambda_n = \delta^* K^* \cos R_n \sin (R_n/\delta^*) - \sin R_n \cos (R_n/\delta^*) \quad (45)$$

$$\Omega_n = \delta^* K^* \sin R_n \cos (R_n/\delta^*) - \cos R_n \sin (R_n/\delta^*) \quad (46)$$

$$\Xi_n = \delta^* K^* \cos R_n \cos (R_n/\delta^*) + \sin R_n \sin (R_n/\delta^*) \quad (47)$$

$$\Psi_n = \delta^* K^* \sin R_n \sin (R_n/\delta^*) + \cos R_n \cos (R_n/\delta^*) \quad (48)$$

The R_n are the nonzero positive roots of

$$\sin \alpha\, (\Lambda \sin \beta^* + \Xi \cos \beta^*) + \delta K \cos \alpha\, (\Psi \sin \beta^* + \Omega \cos \beta^*) = 0 \quad (49)$$

The increase in the amount of diffusant in the membrane at time t over the initial amount is given by

$$M(t) = \int_{-a}^{0} [C_A(x,t) - C_A^i]dx + \int_0^b [C_B(x,t) - C_B^i]\, dx + \int_b^l [C_C(x,t) - C_C^i]\, dx \quad (50)$$

and the final or equilibrium value of $M(t)$ is given by

$$M^f = (C_A^0 - C_A^i)a + (C_B^0 - C_B^i)b + (C_C^0 - C_C^i)(l - b) \quad (51)$$

where C_B^0 is the concentration in B in equilibrium with the bath concentration c^0. The reduced fractional change in the diffusant mass in the membrane is

$$1 - (M(t)/M^f) = [-1/(K\lambda + K^*\lambda^* + 1)] \sum_{n=1}^{\infty} (KA_n + B_n + K^*C_n) \times \exp (-D_B R_n^2 t/b^2) \quad (52)$$

where

$$A_n = 2\delta[(\Lambda_n \sin \beta_n{}^* + \Xi_n \cos \beta_n{}^* - \delta^* K^* \cos \alpha_n) (\cos \alpha_n - 1) + (\delta K\, \Psi_n \sin \beta_n{}^* + \delta K\, \Omega_n \cos \beta_n{}^* + \delta^* K^* \sin \alpha_n) \sin \alpha_n]/R_n S_n \quad (53)$$

$$B_n = 2[(\Lambda_n \sin \beta_n{}^* + \Xi_n \cos \beta_n{}^* - \delta^* K^* \cos \alpha_n)\, \delta K(\cos R_n - 1) + (\delta K\, \Psi_n \sin \beta_n{}^* + \delta K\, \Omega_n \cos \beta_n{}^* + \delta^* K^* \sin \alpha_n) \sin R_n]/R_n S_n \quad (54)$$

$$C_n = 2\delta^*\, \{[\Lambda_n \sin \alpha_n + \delta K\, \Psi_n \cos \alpha_n - \delta K \cos \beta_n{}^*]\, [\cos \beta_n{}^* - \cos (R_n/\delta^*)] + [\Xi_n \sin \alpha_n + \delta K\, \Omega_n \cos \alpha_n + \delta^* K^* \sin \beta_n{}^*] \times [\sin \beta_n{}^* - \sin (R_n/\delta^*)]\}/R_n S_n \quad (55)$$

At large t the first term in eq. (52) dominates, and the expression reduces to

$$\ln\, [1 - (M(t)/M^f)] = \ln\, [-(KA_1 + B_1 + K^*C_1)/(K\lambda + K^*\lambda^* + 1)] - D_B R_1^2 t/b^2 \quad (56)$$

DISCUSSION

In principle it should be possible to determine the K_I and D_I of, for example, the AB laminate in a finite bath from the transient and equilibrium sorption behavior of two samples with different values of the thickness ratio $\lambda = a/b$. From eq. (7) it follows that

$$(c^f - c^0)/(c^i - c^f) = H_A + H_B = (1 + \lambda K)H_B \quad (57)$$

so that two equilibrium measurements provide H_A, H_B, K_A, and K_B, provided λ has been determined. From the limiting slope of ln $M(t)$ vs. t, the product $D_B R_1^2$ is obtained according to eq. (17). To proceed further and calculate both D_B and D_A, it is first necessary to determine iteratively values of R_1 and δ which satisfy both eqs. (12) and (16). This procedure is possible with a computer, but the degree of accuracy is likely to be low as summations with many terms involved.

If K_B and D_B have been determined independently, then the corresponding parameters for the other lamina, K_a and D_a, follow more directly. Thus, a single equilibrium sorption suffices for the measurement of K_A and H_A using the known values of K_B and H_B in eq. (57). From the limiting slope of ln $M(t)$ vs. t, one obtains R and δ, and hence D_A can be determined iteratively from eq. (12).

The greater value of the equations lies in their use to predict the transient and equilibrium sorption behavior of a laminate membrane when the D_I and K_I values are known for each of the laminae. Once again using the AB laminate membrane in a finite bath as an example, H_A and H_B are calculated from the K_I, the membrane dimensions, and the bath volume. As δ is also known, the R_n can be determined iteratively from eq. (12) and $M(t)$ evaluated from eq. (15). Restriction to the region of large t requires only R_1 and eq. (17). The concentration profiles, at least in the later stages of sorption where fewer terms are required in the summation, can be calculated from eq. (6). The same procedure can be applied to the ABC laminate, although it becomes necessarily more lengthy.

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