Non-Steady-State Transport of Diffusants in Ternary Laminate Slabs

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Synopsis

Equations describing transient diffusion in ternary laminate slabs are presented. The equations developed can be applied to: (I) asymmetric ABC slabs separating two semi-infinite baths, each bath containing either the same or different concentration(s) of permeant; (II) asymmetric ABC slabs separating a semi-infinite bath from an impermeable substrate; (III) asymmetric ABC slabs with impermeable substrates attached to laminas A and C. Further, the equations can be applied to slabs containing initially uniform, but not necessarily equilibrial, concentrations of permeant in each lamina. Some numerical examples of the aforementioned diffusion systems I and II are discussed.

INTRODUCTION

Equations have been obtained for diffusion from a well-stirred semi-infinite bath into a homogeneous slab.[1] Spencer and Barrie have expanded this work to include symmetric ABA and asymmetric AB laminate slabs,[2] and recently an asymmetric ABC laminate slab.[3] However, the initial conditions imposed upon the last-mentioned system were rather restrictive in that the individual laminas were considered to be initially in equilibrium with each other. In addition, the slab separated two semi-infinite baths, each bath containing the same concentration of diffusant. This paper extends the work of Spencer and Barrie to include more general initial and boundary conditions. Furthermore, modification of the distance coordinate results in the derivation of equations which are easier to use.

DIFFUSION EQUATIONS

Definitions, Assumptions, and General Diffusion Equations

The membrane is a slab comprising three laminas, lamina 1 of thickness l_1, lamina 2 of thickness $l_2 - l_1$, and lamina 3 of thickness $l_3 - l_2$. A schematic representation of the ternary laminate slab is shown in Figure 1. The concentrations in each lamina at $t = 0$ are uniform; C_1^i in lamina 1, C_2^i in lamina 2, and C_3^i in lamina 3.

It is assumed that equilibrium is maintained at the phase interfaces when $t > 0$. Further, it is assumed that the solubility follows Henry's law in each lamina, so that when $t > 0$, $C_1 = K_{12}C_2$ at $x = l_1$ and $C_2 = K_{23}C_3$ at $x = l_2$, where K_{12} and K_{23} are constants. Another useful quantity, as will become apparent is $K_{13} = K_{12}K_{23}$. The diffusion coefficients in the laminas, D_1, D_2, and D_3 are constant.

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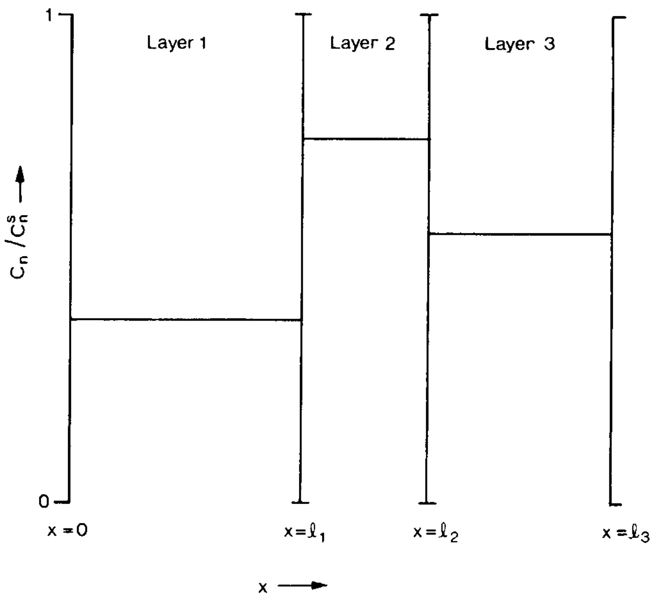

Fig. 1. Schematic representation of the ternary laminate slab at $t = 0$. Quantity C_n^s is the saturation concentration of diffusant in layer $n (n = 1,2,3)$ which is in equilibrium with pure diffusant. In general the ratios C_1^i/C_1^s, C_2^i/C_2^s, and C_3^i/C_3^s are different and may lie anywhere between 0 and 1.

Spencer and Barrie[3] have considered a case of a ternary laminate slab in which $C_1^i = K_{12}C_2^i$, $C_2^i = K_{23}C_3^i$, and $C_1^i = K_{13}C_3^i$. However, it is useful to have solutions for $C_1^i \neq K_{12}C_2^i$, $C_2^i \neq K_{23}C_3^i$, and $C_1^i \neq K_{13}C_3^i$, i.e., nonequilibrial initial concentrations of permeant. In this paper it will be assumed that the laminas, in general, are not initially in equilibrium with each other, but the derived solutions to the general diffusion equations can also cope with equilibrial or mixed equilibrial/nonequilibrial initial conditions.

When the ternary slabs are exposed to the semi-infinite baths (systems I and II to be considered) at $t \geqslant 0$, the exposed surfaces are assumed to be in equilibrium with their respective contact baths.

The differential equations describing transient diffusion are

$$\frac{\partial^2 C_1}{\partial x^2} = \frac{1}{D_1}\frac{\partial C_1}{\partial t}, \qquad 0 < x < l_1$$
$$\frac{\partial^2 C_2}{\partial x^2} = \frac{1}{D_2}\frac{\partial C_2}{\partial t}, \qquad l_1 < x < l_2$$
$$\frac{\partial^2 C_3}{\partial x^2} = \frac{1}{D_3}\frac{\partial C_3}{\partial t}, \qquad l_2 < x < l_3 \qquad (1)$$

The following dimensionless parameters are defined:

$\delta_{12} = (D_1/D_2)^{1/2}$; $\delta_{23} = (D_2/D_3)^{1/2}$; $\delta_{13} = (D_1/D_3)^{1/2}$
$\lambda_{12} = l_1/(l_2 - l_1)$; $\lambda_{23} = (l_2 - l_1)/(l_3 - l_2)$; $\lambda_{13} = l_1/(l_3 - l_2)$
$\xi_1 = x/l_1$; $\xi_2 = (l_2 - x)/(l_2 - l_1)$; $\xi_3 = (l_3 - x)/(l_3 - l_2)$
$\tau = D_1 t/l_1^2$

Equations (1) can be written in a semidimensionless form. Concentration is not made dimensionless for reasons which will become apparent. Thus

$$\frac{\partial^2 C_1}{\partial \xi_1^2} = \frac{\partial C_1}{\partial \tau}, \qquad 0 < \xi_1 < 1$$
$$\frac{\partial^2 C_2}{\partial \xi_2^2} = \frac{\delta_{12}^2}{\lambda_{12}^2}\frac{\partial C_2}{\partial \tau}, \qquad 1 > \xi_2 > 0 \tag{2}$$
$$\frac{\partial^2 C_3}{\partial \xi_3^2} = \frac{\delta_{13}^2}{\lambda_{13}^2}\frac{\partial C_3}{\partial \tau}, \qquad 1 > \xi_3 > 0$$

The Laplace transformation method[4] provides the solution to eqs. (2), subject to the appropriate boundary and initial conditions. The application of the Laplace transform technique to systems I, II, and III is outlined in the Appendix.

System I: Asymmetric ABC Slab Separating Two Semi-Infinite Baths, Each Bath Containing Different Concentrations of Diffusant

The initial and boundary conditions for this sytem are

$$C_3 = C_3^i, \qquad 1 \geqslant \xi_3 > 0, \qquad \tau = 0$$
$$C_2 = C_2^i, \qquad 1 \geqslant \xi_2 \geqslant 0, \qquad \tau = 0 \tag{3}$$
$$C_1 = C_1^i, \qquad 0 < \xi_1 \leqslant 1, \qquad \tau = 0$$

where, in general, $C_1^i \neq K_{12}C_2^i$, $C_2^i \neq K_{23}C_3^i$, $C_1^i \neq K_{13}C_3^i$;

$$C_3 = C_3^0, \qquad \xi_3 = 0, \qquad \tau \geqslant 0$$
$$C_1 = C_1^0, \qquad \xi_1 = 0, \qquad \tau \geqslant 0 \tag{4}$$

where C_1^0 and C_3^0 are the constant concentrations in the two membrane surfaces which are in equilibrium with the two semi-infinite contact baths; in general $C_1^0 \neq K_{13}C_3^0$, i.e., the concentrations of diffusant in the two baths are different;

$$(C_1)_{\xi_1=1} = K_{12}(C_2)_{\xi_2=1}, \qquad (C_2)_{\xi_2=0} = K_{23}(C_3)_{\xi_3=1}, \qquad \tau > 0 \tag{5}$$

$$-\frac{\delta_{12}^2}{\lambda_{12}}\left(\frac{\partial C_1}{\partial \xi_1}\right)_{\xi_1=1} = \left(\frac{\partial C_2}{\partial \xi_2}\right)_{\xi_2=1}, \qquad \frac{\delta_{23}^2}{\lambda_{23}}\left(\frac{\partial C_2}{\partial \xi_2}\right)_{\xi_2=0} = \left(\frac{\partial C_3}{\partial \xi_3}\right)_{\xi_3=1}, \qquad \tau > 0 \tag{6}$$

The Laplace transform method gives the solutions

$$C_3 = X(\xi_3) + \sum_{m=1}^{\infty} \frac{A_m \sin[(\delta_{13}/\lambda_{13})\alpha_m \xi_3] \exp(-\alpha_m^2 \tau)}{R_m} \tag{7}$$

$$C_2 = Y(\xi_2) + \sum_{m=1}^{\infty} \frac{\{B_m \cos[(\delta_{12}/\lambda_{12})\alpha_m \xi_2] + E_m \sin[(\delta_{12}/\lambda_{12})\alpha_m \xi_2]\} \exp(-\alpha_m^2 \tau)}{R_m} \tag{8}$$

$$C_1 = Z(\xi_1) + \sum_{m=1}^{\infty} \frac{F_m \sin(\alpha_m \xi_1) \cdot \exp(-\alpha_m^2 \tau)}{R_m} \tag{9}$$

where the α_m are the positive roots of an auxiliary equation determined by the boundary conditions.

These solutions are presented in Table I, with

$$\Lambda(\alpha_m) = \delta_{12}K_{12}\cos\alpha_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) + \sin\alpha_m \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) \quad (10)$$

$$\Omega(\alpha_m) = \delta_{12}K_{12}\sin\alpha_m \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) + \cos\alpha_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) \quad (11)$$

$$\Xi(\alpha_m) = \delta_{12}K_{12}\cos\alpha_m \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) - \sin\alpha_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) \quad (12)$$

$$\Psi(\alpha_m) = \cos\alpha_m \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) - \delta_{12}K_{12}\sin\alpha_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) \quad (13)$$

In eqs. (7)–(9) $X(\xi_3)$, $Y(\xi_2)$, and $Z(\xi_1)$ are the concentrations, respectively, in layers 3, 2, and 1 as t (or τ) $\rightarrow \infty$.

It is useful to be able to calculate the average concentration of diffusant in any layer at time t (or dimensionless time τ). Defining the average concentration in layer $n (n = 1,2,3)$ as C_n^*, it is easily shown that

$$C_n^*(\tau) = \int_0^1 C_n(\xi_n,\tau)\, d\xi_n \quad (14)$$

from which

$$C_1^*(\tau) = C_1^0 + \frac{(1/2)(K_{13}C_3^0 - C_1^0)}{1 + \delta_{12}^2K_{12}/\lambda_{12} + \delta_{13}^2K_{13}/\lambda_{13}} + \sum_{m=1}^{\infty} \frac{F_m[1 - \cos\alpha_m]\exp(-\alpha_m^2\tau)}{\alpha_m R_m} \quad (15)$$

$$C_2^*(\tau) = \frac{(\delta_{12}^2/2\lambda_{12})(C_1^0 - K_{13}C_3^0) + K_{23}C_3^0(1 + \delta_{12}^2K_{12}/\lambda_{12}) + (K_{23}\delta_{13}^2/\lambda_{13})C_1^0}{1 + \delta_{12}^2K_{12}/\lambda_{12} + \delta_{13}^2K_{13}/\lambda_{13}} + \frac{\lambda_{12}}{\delta_{12}}\sum_{m=1}^{\infty} \frac{\left\{B_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right) + E_m\left[1 - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right)\right]\right\}\exp(-\alpha_m^2\tau)}{\alpha_m R_m} \quad (16)$$

$$C_3^*(\tau) = C_3^0 + \frac{(\delta_{13}^2/2\lambda_{13})(C_1^0 - K_{13}C_3^0)}{1 + \delta_{12}^2K_{12}/\lambda_{12} + \delta_{13}^2K_{13}/\lambda_{13}} + \frac{\lambda_{13}}{\delta_{13}}\sum_{m=1}^{\infty} \frac{A_m\{1 - \cos[(\delta_{13}/\lambda_{13})\alpha_m]\}\exp(-\alpha_m^2\tau)}{\alpha_m R_m} \quad (17)$$

Equations (7)–(9) and (15)–(17) are very complicated. However, with simpler initial conditions, the solutions also become simpler. Consider the case $C_1^i = K_{12}C_2^i$, $C_2^i = K_{23}C_3^i$. Then R_m, $X(\xi_3)$, $Y(\xi_2)$, $Z(\xi_1)$, and the auxiliary equation remain the same, but

$$A_m = \delta_{13}(C_1^0 - C_1^i) + (C_3^0 - C_3^i)\left[\Lambda(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \delta_{23}K_{23}\,\Xi(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]$$

$$B_m = K_{23}(C_3^0 - C_3^i)\Lambda(\alpha_m) + \delta_{13}K_{23}(C_1^0 - C_1^i)\sin[(\delta_{13}/\lambda_{13})\alpha_m]$$

$$E_m = \delta_{12}(C_1^0 - C_1^i)\cos[(\delta_{13}/\lambda_{13})\alpha_m] - K_{23}(C_3^0 - C_3^i)\Xi(\alpha_m)$$

TABLE I
System I: Solutions to Diffusion Equations

Layer No.		Solution
3	$X(\xi_3) = C_3^0 + \dfrac{(\delta_{13}^2\xi_3/\lambda_{13})[C_1^0 - K_{13}C_3^0]}{[1 + \delta_{12}^2K_{12}/\lambda_{12} + \delta_{13}^2K_{13}/\lambda_{13}]}$	$A_m = \delta_{13}(C_1^0 - C_1^i) + \delta_{13}(C_1^i - K_{12}C_2^i)\cos\alpha_m$ $+ \delta_{23}(C_2^i - K_{23}C_3^i)\Xi(\alpha_m)$ $+ (C_3^0 - C_3^i)\left[\Lambda(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \delta_{23}K_{23}\Xi(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]$
2	$Y(\xi_2) = \dfrac{(\delta_{12}^2\xi_2/\lambda_{12})[C_1^0 - K_{13}C_3^0] + K_{23}C_3^0[1 + \delta_{12}^2K_{12}/\lambda_{12}] + (K_{23}\delta_{13}^2/\lambda_{13})C_1^0}{[1 + \delta_{12}^2K_{12}/\lambda_{12} + \delta_{13}^2K_{13}/\lambda_{13}]}$	$B_m = \left[K_{23}(C_3^0 - C_3^i) + (K_{23}C_3^i - C_2^i)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]\Lambda(\alpha_m)$ $+ [\delta_{13}K_{23}(C_1^0 - C_1^i) + \delta_{13}K_{23}(C_1^i - K_{12}C_2^i)\cos\alpha_m]\sin[(\delta_{13}/\lambda_{13})\alpha_m]$ $E_m = [\delta_{12}(C_1^0 - C_1^i) + \delta_{12}(C_1^i - K_{12}C_2^i)\cos\alpha_m]\cos[(\delta_{13}/\lambda_{13})\alpha_m]$ $- \{K_{23}(C_3^0 - C_3^i) + (K_{23}C_3^i - C_2^i)\cos[(\delta_{13}/\lambda_{13})\alpha_m]\}\Xi(\alpha_m)$
1	$Z(\xi_1) = C_1^0 + \dfrac{\xi_1[K_{13}C_3^0 - C_1^0]}{[1 + \delta_{12}^2K_{12}/\lambda_{12} + \delta_{13}^2K_{13}/\lambda_{13}]}$	$F_m = K_{13}(C_3^0 - C_3^i) + (K_{13}C_3^i - K_{12}C_2^i)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)$ $+ (C_1^0 - C_1^i)\left[\delta_{23}K_{23}\Omega(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \Psi(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]$ $+ (C_1^i - K_{12}C_2^i)\left[\delta_{23}K_{23}\sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]$
	$R_m = \frac{\alpha_m}{2}\left\{\left[\left(\frac{\delta_{13}\delta_{23}K_{23}}{\lambda_{13}} + \frac{\delta_{12}}{\lambda_{12}}\right)\Xi(\alpha_m) + \Psi(\alpha_m)\right]\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \left[\left(\frac{\delta_{13}K_{23}}{\lambda_{12}} + \frac{\delta_{13}}{\lambda_{13}}\right)\Lambda(\alpha_m) + \delta_{23}K_{23}\Omega(\alpha_m)\right]\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right\}$	
	Auxiliary equation: $\Lambda(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) + \delta_{23}K_{23}\Xi(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) = 0$	

$$F_m = K_{13}(C_3^0 - C_3^i) + (C_1^0 - C_1^i)\left[\delta_{23}K_{23}\Omega(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \Psi(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]$$

Now consider the case $C_1^i = K_{12}C_2^i$, $C_2^i = K_{23}C_3^i$, and $C_1^0 = K_{13}C_3^0$. The last-mentioned condition is tantamount to specifying similar concentrations of diffusant in the two semi-infinite baths. Thus

$$X(\xi_3) = C_3^0; \qquad A_m = \delta_{13}K_{13}(C_3^0 - C_3^i) + (C_3^0 - C_3^i) \times \left[\Lambda(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \delta_{23}K_{23}\Xi(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]$$

$$Y(\xi_2) = K_{23}C_3^0; \qquad B_m = K_{23}(C_3^0 - C_3^i)\Lambda(\alpha_m) + \delta_{13}K_{13}K_{23}(C_3^0 - C_3^i)\sin[(\delta_{13}/\lambda_{13})\,\alpha_m]$$

$$E_m = \delta_{12}K_{13}(C_3^0 - C_3^i)\cos[(\delta_{13}/\lambda_{13})\alpha_m] - K_{23}(C_3^0 - C_3^i)\Xi(\alpha_m)$$

$$Z(\xi_1) = C_1^0; \qquad F_m = K_{13}(C_3^0 - C_3^i) + K_{13}(C_3^0 - C_3^i) \times \left[\delta_{23}K_{23}\Omega(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \Psi(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right)\right]$$

With the above coefficients A_m, B_m, E_m, and F_m, and values of $X(\xi_3)$, $Y(\xi_2)$, and $Z(\xi_1)$ we have a solution equivalent to that obtained by Spencer and Barrie,[3] except that the new equations are somewhat more tractable. It can be seen that the term $(C_3^0 - C_3^i)$ turns up in A_m, B_m, E_m, and F_m. Thus in this instance the solutions can be made dimensionless by dividing both sides of eqs. (7)–(9) by $(C_3^0 - C_3^i)$. However, in the more general case elaborated in Table I there is no obvious choice of dimensionless concentration, so that the reason for rendering the diffusion equations into the semidimensionless eqs. (2) becomes obvious.

System II: Asymmetric ABC Slabs Separating a Semi-Infinite Bath from an Impermeable Substrate

This system is equivalent to a free symmetrical ABCBA laminate slab separating two semi-infinite baths, each bath containing the same concentration of diffusant. The initial conditions are given by eqs. (3). Boundary conditions (5) and (6) are retained, but boundary conditions (4) are modified to

$$\begin{aligned} C_3 &= C_3^0, & \xi_3 &= 0, & \tau &\geqslant 0 \\ \partial C_1/\partial \xi_1 &= 0, & \xi_1 &= 0, & \tau &\geqslant 0 \end{aligned} \tag{18}$$

The Laplace transform method gives the solutions

$$C_3 = C_3^0 + \sum_{m=1}^{\infty} \frac{G_m \sin[(\delta_{13}/\lambda_{13})\beta_m\xi_3]\cdot\exp(-\beta_m^2\tau)}{S_m} \tag{19}$$

$$C_2 = K_{23}C_3^0 + \sum_{m=1}^{\infty} \frac{\{H_m \cos[(\delta_{12}/\lambda_{12})\beta_m\xi_2] + M_m \sin[(\delta_{12}/\lambda_{12})\beta_m\xi_2]\}\exp(-\beta_m^2\tau)}{S_m} \tag{20}$$

$$C_1 = K_{13}C_3^0 + \sum_{m=1}^{\infty} \frac{N_m \cos(\beta_m \xi_1) \cdot \exp(-\beta_m^2 \tau)}{S_m} \tag{21}$$

where β_m are the positive roots of an auxiliary equation determined by the boundary conditions. The solutions to the diffusion equations are presented in Table II. The quantities $\Lambda(\beta_m)$ etc., are defined by eqs. (10)–(13) with α_m replaced by β_m.

In eqs. (19)–(21) C_3^0, $K_{23}C_3^0$, and $K_{13}C_3^0$ are the concentrations, respectively, in layers 3, 2, and 1 as t(or τ) $\to \infty$.

Using eq. (14) the average concentrations in each layer are found to be

$$C_1^*(\tau) = K_{13}C_3^0 + \sum_{m=1}^{\infty} \frac{N_m \sin\beta_m \cdot \exp(-\beta_m^2 \tau)}{\beta_m S_m} \tag{22}$$

$$C_2^*(\tau) = K_{23}C_3^0 + \frac{\lambda_{12}}{\delta_{12}} \sum_{m=1}^{\infty} \frac{\left\{H_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\right) + M_m\left[1 - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\right)\right]\right\} \exp(-\beta_m^2 \tau)}{\beta_m S_m} \tag{23}$$

$$C_3^*(\tau) = C_3^0 + \frac{\lambda_{13}}{\delta_{13}} \sum_{m=1}^{\infty} \frac{G_m\{1 - \cos[\delta_{13}/\lambda_{13})\beta_m]\} \exp(-\beta_m^2 \tau)}{\beta_m S_m} \tag{24}$$

Consider the case $C_1^i = K_{12}C_2^i$, $C_2^i = K_{23}C_3^i$, which has recently been solved by Spencer and Barrie.[5] Then the coefficients G_m, H_m, M_m, and N_m are simplified to

$$G_m = (C_3^0 - C_3^i)\left[\Psi(\beta_m) \sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right) + \delta_{23}K_{23}\Omega(\beta_m) \cos\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right]$$

$$H_m = K_{23}(C_3^0 - C_3^i)\Psi(\beta_m)$$

$$M_m = K_{23}(C_3^0 - C_3^i)\Omega(\beta_m)$$

$$N_m = K_{13}(C_3^0 - C_3^i)$$

Substituting these values of G_m, etc., into eqs. (19)–(21), and dividing both sides of the resulting equations by $(C_3^0 - C_3^i)$, leads to entirely dimensionless expressions.

System III: Asymmetric ABC Slabs with Impermeable Substrates Attached to Laminas A and C

The initial conditions are given by eqs. (3). Boundary conditions (5) and (6) are retained, but boundary conditions (4) are modified to

$$\begin{aligned} \partial C_3/\partial \xi_3 = 0, &\quad \xi_3 = 0, \quad \tau \geqslant 0 \\ \partial C_1/\partial \xi_1 = 0, &\quad \xi_1 = 0, \quad \tau \geqslant 0 \end{aligned} \tag{25}$$

The Laplace transform method gives the solutions

$$C_3 = \frac{C_3^i + C_2^i\lambda_{23} + C_1^i\lambda_{13}}{1 + \lambda_{23}K_{23} + \lambda_{13}K_{13}} + \sum_{m=1}^{\infty} \frac{P_m \cos[(\delta_{13}/\lambda_{13})\gamma_m \xi_3] \cdot \exp(-\gamma_m^2 \tau)}{T_m} \tag{26}$$

$$C_2 = \frac{K_{23}(C_3^i + C_2^i\lambda_{23} + C_1^i\lambda_{13})}{1 + \lambda_{23}K_{23} + \lambda_{13}K_{13}} + \sum_{m=1}^{\infty} \left\{\left[Q_m \cos\left(\frac{\delta_{12}}{\lambda_{12}}\gamma_m \xi_2\right) + U_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\gamma_m \xi_2\right)\right] \cdot \exp(-\gamma_m^2 \tau)\right\} (T_m)^{-1} \tag{27}$$

TABLE II
System II: Solutions to Diffusion Equations

Layer No.	Solution
3	$G_m = (C_3^0 - C_3^i)\left[\Psi(\beta_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right) + \delta_{23}K_{23}\Omega(\beta_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right] + \delta_{23}(K_{23}C_3^i - C_2^i)\Omega(\beta_m) - \delta_{13}(C_1^i - K_{12}C_2^i)\sin\beta_m$
2	$H_m = \{K_{23}(C_3^0 - C_3^i) + (K_{23}C_3^i - C_2^i)\cos[(\delta_{13}/\lambda_{13})\beta_m]\}\Psi(\beta_m) - \delta_{13}K_{23}(C_1^i - K_{12}C_2^i)\sin\beta_m \sin[(\delta_{13}/\lambda_{13})\beta_m]$
	$M_m = \{K_{23}(C_3^0 - C_3^i) + (K_{23}C_3^i - C_2^i)\cos[(\delta_{13}/\lambda_{13})\beta_m]\}\,\Omega(\beta_m) - \delta_{12}(C_1^i - K_{12}C_2^i)\sin\beta_m \cos[(\delta_{13}/\lambda_{13})\beta_m]$
1	$N_m = K_{13}(C_3^0 - C_3^i) + (K_{13}C_3^i - K_{12}C_2^i)\cos[(\delta_{13}/\lambda_{13})\beta_m] + (C_1^i - K_{12}C_2^i)\left[\delta_{23}K_{23}\sin\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\right)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right) - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\right)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right]$
	$S_m = \frac{-\beta_m}{2}\left\{\left[\left(\frac{\delta_{13}\delta_{23}K_{23}}{\lambda_{13}} + \frac{\delta_{12}}{\lambda_{12}}\right)\Omega(\beta_m) + \Lambda(\beta_m)\right]\cos\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right) + \left[\left(\frac{\delta_{13}K_{23}}{\lambda_{12}} + \frac{\delta_{13}}{\lambda_{13}}\right)\Psi(\beta_m) + \delta_{23}K_{23}\Xi(\beta_m)\right]\sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right\}$
	Auxiliary equation: $\Psi(\beta_m)\cos[(\delta_{13}/\lambda_{13})\beta_m] - \delta_{23}K_{23}\Omega(\beta_m)\sin[(\delta_{13}/\lambda_{13})\beta_m] = 0$

$$C_1 = \frac{K_{13}(C_3^i + C_2^i\lambda_{23} + C_1^i\lambda_{13})}{1 + \lambda_{23}K_{23} + \lambda_{13}K_{13})} + \sum_{m=1}^{\infty} \frac{V_m \cos(\gamma_m \xi_1) \cdot \exp(-\gamma_m^2 \tau)}{T_m} \quad (28)$$

where the γ_m are the positive roots of an auxiliary equation determined by the boundary conditions. The solutions to the diffusion equations are presented in Table III. The $\Lambda(\gamma_m)$, etc., are defined by eqs. (10)–(13) with α_m replaced by γ_m.

In eqs. (26)–(28) the terms on the right hand sides of the equations outside the summation signs represent the concentrations, respectively, in layers 3, 2, and 1 as t(or τ) $\rightarrow \infty$.

Using eq. (14) the average concentrations in each layer are found to be

$$C_1^*(\tau) = \frac{K_{13}(C_3^i + C_2^i\lambda_{23} + C_1^i\lambda_{13})}{1 + \lambda_{23}K_{23} + \lambda_{13}K_{13})} + \sum_{m=1}^{\infty} \frac{V_m \sin\gamma_m \cdot \exp(-\gamma_m^2 \tau)}{\gamma_m T_m} \quad (29)$$

$$C_2^*(\tau) = \frac{K_{23}(C_3^i + C_2^i\lambda_{23} + C_1^i\lambda_{13})}{1 + \lambda_{23}K_{23} + \lambda_{13}K_{13}} + \frac{\lambda_{12}}{\delta_{12}} \sum_{m=1}^{\infty} \left(\left\{ Q_m \sin\left(\frac{\delta_{12}}{\lambda_{12}}\gamma_m\right) + U_m \left[1 - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\gamma_m\right)\right] \right\} \exp(-\gamma_m^2\tau) \right) (\gamma_m T_m)^{-1} \quad (30)$$

$$C_3^*(\tau) = \frac{C_3^i + C_2^i\lambda_{23} + C_1^i\lambda_{13}}{1 + \lambda_{23}K_{23} + \lambda_{13}K_{13}} + \frac{\lambda_{13}}{\delta_{13}} \times \sum_{m=1}^{\infty} \frac{P_m \sin\left[(\delta_{13}/\lambda_{13})\gamma_m\right] \exp(-\gamma_m^2 \tau)}{\gamma_m T_m} \quad (31)$$

NUMERICAL EXAMPLES OF DIFFUSION IN TERNARY LAMINATE SLABS

The main use for the solutions presented in this paper is in predicting concentration profiles in ternary laminate slabs, knowing values of δ and K.

One case which requires knowledge of transient diffusion in multilaminate slabs is that of power cables without metallic sheaths. The diffusant of interest frequently is water, but transfer of plasticizers may also be of concern. Cable geometry is, of course, essentially cylindrical, but there are several reasons for using a plane slab as an approximation.

First, in many high-voltage power cables the thicknesses of the polymeric layers are small in relation to the radius of the conductor. When this is so the series solutions for cylindrical coordinates converge very slowly.

Secondly, solutions for annuli are very complicated. The binary laminate annulus yields solutions[6] which are equivalent in complexity to those for the ternary laminate slab. Solutions for the ternary laminate annulus are so complicated as to be of little practical use.

Thirdly, diffusion data and the assumptions, e.g., the constancy of diffusion coefficients and the applicability of Henry's law, are rarely accurate, so that the plane slab approximation to the annulus is acceptable, if the objective is simply to obtain an order-of-magnitude estimate of concentration at a particular time.

We will now consider three particular instances in which water is the diffusant.

TABLE III

System III: Solutions to Diffusion Equations

Layer No.	Solution
3	$P_m = \delta_{13}(C_1^i - K_{12}C_2^i)\sin\gamma_m - \delta_{23}(K_{23}C_3^i - C_2^i)\Omega(\gamma_m)$
2	$Q_m = (K_{23}C_3^i - C_2^i)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right)\Psi(\gamma_m) + \delta_{13}K_{23}(C_1^i - K_{12}C_2^i)\sin\gamma_m\cos\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right)$
	$U_m = (K_{23}C_3^i - C_2^i)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right)\Omega(\gamma_m) - \delta_{12}(C_1^i - K_{12}C_2^i)\sin\gamma_m\sin\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right)$
1	$V_m = (K_{13}C_3^i - K_{12}C_2^i)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right) - (C_1^i - K_{12}C_2^i)\left[\delta_{23}K_{23}\cos\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right)\sin\left(\frac{\delta_{12}}{\lambda_{12}}\gamma_m\right) + \sin\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right)\cos\left(\frac{\delta_{12}}{\lambda_{12}}\gamma_m\right)\right]$
	$T_m = \frac{\gamma_m}{2}\left\{\left[\left(\frac{\delta_{13}K_{23}}{\lambda_{12}} + \frac{\delta_{13}}{\lambda_{13}}\right)\Psi(\gamma_m) + \delta_{23}K_{23}\Xi(\gamma_m)\right]\cos\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right) - \left[\left(\frac{\delta_{13}\delta_{23}K_{23}}{\lambda_{13}} + \frac{\delta_{12}}{\lambda_{12}}\right)\Omega(\gamma_m) + \Lambda(\gamma_m)\right]\sin\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right)\right\}$
	Auxiliary equation: $\Psi(\gamma_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right) + \delta_{23}K_{23}\Omega(\gamma_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\gamma_m\right) = 0$

TABLE IV
Values of D, C, and l used in Examples (T = 25°C)

Layer	D_n(cm²/s)	C_n^s(g/cm³)	Length(cm)
1	5.2×10^{-9}	9.2×10^{-5}	$l_1 = 0.5$
2	8.4×10^{-9}	2×10^{-3}	$l_2 - l_1 = 0.3$
3	3.5×10^{-8}	2×10^{-3}	$l_3 - l_2 = 0.3$

The composite slab comprises an outer plasticized poly(vinyl chloride) (PVC) sheath (layer 3), an intermediate layer based on unvulcanized butyl rubber (layer 2) and a low-density polyethylene insulation (layer 1). The PVC sheath is contacted with a semi-infinite bath of pure water. The data used for the following examples are presented in Table IV. Diffusion coefficients and saturation concentrations for PVC and unvulcanized butyl rubber were determined experimentally by the author. Literature values for the diffusion coefficient of water in low-density polyethylene vary enormously. A low diffusion coefficient was deliberately selected as representing the most optimistic value, water being considered detrimental to the electrical integrity of the insulation. It should be realised, however, that the main purpose of the exercise is to demonstrate the applicability of the diffusion equation solutions.

Example 1

For the first example we will consider System II with all three laminas initially free of diffusant. In addition $C_1^i = C_2^i = C_3^i = 0$ and $C_3^0 = C_3^s$, where the superscript s denotes saturation with diffusant. This is a specific case of a more general solution obtained by Spencer and Barrie.[5]

With the above-mentioned restrictions, application of eqs. (19)–(21) and Table II gives

$$\frac{C_3 - C_3^s}{C_3^s} = \sum_{m=1}^{\infty} \left\{ \left[\Psi(\beta_m) \sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right) + \delta_{23}K_{23}\Omega(\beta_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right] \sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\xi_3\right) \exp(-\beta_m{}^2\tau)\right\}(S_m)^{-1} \quad (32)$$

$$\frac{C_2 - C_2^s}{C_2^s} = \sum_{m=1}^{\infty} \left\{ \left[\Psi(\beta_m) \cos\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\xi_2\right) + \Omega(\beta_m)\sin\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\xi_2\right)\right] \exp(-\beta_m{}^2\tau)\right\}(S_m)^{-1} \quad (33)$$

$$\frac{C_1 - C_1^s}{C_1^s} = \sum_{m=1}^{\infty} \frac{\cos(\beta_m\xi_1)\cdot\exp(-\beta_m{}^2\tau)}{S_m} \quad (34)$$

with the β_m being the nonzero positive roots of the auxiliary equation presented in Table II. It will be seen that the above equations are entirely dimensionless. Similar solutions in nondimensionless form were obtained by Le Poidevin.[7]

Using the data of Table IV the first four roots of the auxiliary equation were found to be $\beta_1 = 1.528$, $\beta_2 = 2.660$, $\beta_3 = 4.718$ and $\beta_4 = 6.715$. With this information the concentration profile at 3000 h was computed and the result is shown in Figure 2.

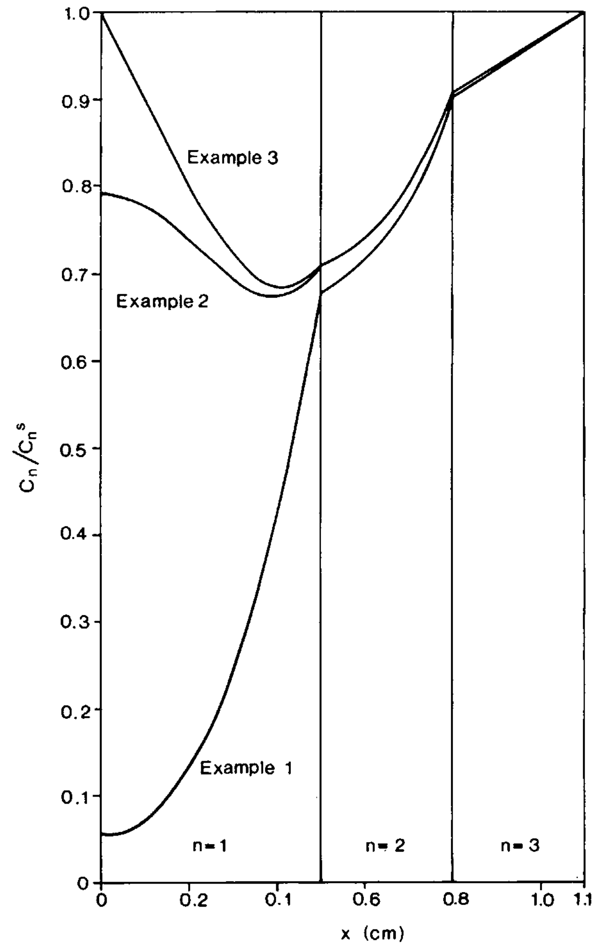

Fig. 2. Concentration profiles at 3000 h for examples 1, 2, and 3.

Example 2

For the second example we will consider System II with laminas 2 and 3 initially free of diffusant, but lamina 1 saturated with diffusant. This situation sometimes arises in power cables when the extruded polyethylene insulation is crosslinked, and is saturated (or supersaturated) with water either from the steam heat transfer medium or as the result of the decomposition of the chemical crosslinking agent. We will assume that the insulation is crosslinked in an inert gas heat transfer medium, and that the resultant extrudate has the same diffusion properties as uncrosslinked low-density polyethylene and is initially saturated with water.

We use eqs. (19)–(21) and Table II, together with the restrictions $C_2^i = C_3^i = 0$, $C_1^i = C_1^s$, and $C_3^0 = C_3^s$, to obtain

$$\frac{C_3 - C_3^s}{C_3^s} = \sum_{m=1}^{\infty} \left\{ \left[\Psi(\beta_m) \sin\left(\frac{\delta_{13}}{\lambda_{13}} \beta_m\right) + \delta_{23} K_{23} \Omega(\beta_m) \cos\left(\frac{\delta_{13}}{\lambda_{13}} \beta_m\right) - \delta_{13} K_{13} \sin\beta_m \right] \sin\left(\frac{\delta_{13}}{\lambda_{13}} \beta_m \xi_3\right) \cdot \exp(-\beta_m^2 \tau) \right\} (S_m)^{-1} \quad (35)$$

$$\frac{C_2 - C_2^s}{C_2^s} = \sum_{m=1}^{\infty} \left\{\left[\Psi(\beta_m) - \delta_{13}K_{13}\sin\beta_m \sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right]\cos\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\xi_2\right)\right.$$
$$\left. + \left[\Omega(\beta_m) - \delta_{12}K_{12}\sin\beta_m \cos\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right]\sin\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\xi_2\right)\right\}\exp(-\beta_m^2\tau)\,(S_m)^{-1} \tag{36}$$

$$\frac{C_1 - C_1^s}{C_1^s} = \sum_{m=1}^{\infty} \left\{\left[1 + \delta_{23}K_{23}\sin\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\right)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right) - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\beta_m\right)\right.\right.$$
$$\left.\left. \times \cos\left(\frac{\delta_{13}}{\lambda_{13}}\beta_m\right)\right]\cos(\beta_m\xi_1)\exp(-\beta_m^2\tau)\right\}(S_m)^{-1} \tag{37}$$

with the β_m again being the nonzero positive roots of the auxiliary equation presented in Table II.

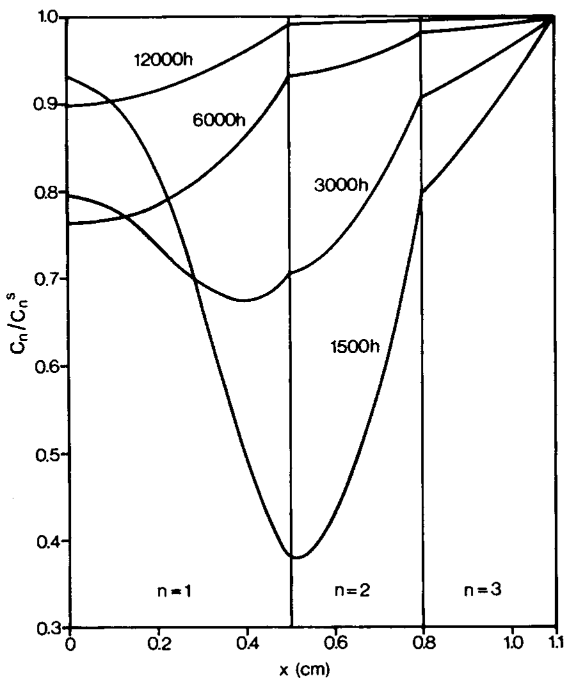

Fig. 3. Concentration profiles at various times for example 2.

The concentration profile at 3000 h is shown in Figure 2 and may be compared with those of examples 1 and 3. Figure 3 shows the concentration profiles for example 2 at various times. As might be expected at low times there is a minimum in the concentration. This minimum moves towards $x = 0$ (the boundary with the impermeable substrate) as time goes by. Between 3000 h and 6000 h the minimum reaches $x = 0$, and remains at $x = 0$ thereafter. This is sensible because, as mentioned earlier, this laminate slab can be considered as one half of a symmetrical ABCBA slab which separates two semi-infinite baths, each bath having in it the same concentration of diffusant, in these examples both baths containing liquid water.

Example 3

We will now consider an example of System I. In the context of water diffusion in a power cable this situation could arise if the conductor is stranded, thus permitting the movement of liquid water along the conductor and consequently diffusion into the laminate from the insulation side as well as from outside the sheath.

Let us consider the restrictions $C_2^i = C_3^i = 0$, $C_1^i = C_1^s$, $C_3^0 = C_3^s$, and $C_1^0 = K_{13}C_3^0$. The last-mentioned restriction applies because the concentration of diffusant in the two "baths" is the same. Thus from eqs. (7)–(10) and Table I it can be shown that

$$\frac{C_3 - C_3^s}{C_3^s} = \sum_{m=1}^{\infty} \left\{ \left[\delta_{13}K_{13}\cos\alpha_m + \Lambda(\alpha_m)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \delta_{23}K_{23}\,\Xi(\alpha_m)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) \right] \sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\xi_3\right) \cdot \exp(-\alpha_m^2\tau) \right\} (R_m)^{-1} \quad (38)$$

$$\frac{C_2 - C_2^s}{C_2^s} = \sum_{m=1}^{\infty} \left(\left\{ \left[\Lambda(\alpha_m) + \delta_{13}K_{13}\cos\alpha_m \sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) \right] \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\xi_2\right) + \left[\delta_{12}K_{12}\cos\alpha_m \cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \Xi(\alpha_m) \right] \sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\xi_2\right) \right\} \exp(-\alpha_m^2\tau) \right) (R_m)^{-1} \quad (39)$$

$$\frac{C_1 - C_1^s}{C_1^s} = \sum_{m=1}^{\infty} \left\{ \left[1 + \delta_{23}K_{23}\sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_m\right)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_m\right) \right] \sin(\alpha_m\xi_1)\exp(-\alpha_m^2\tau) \right\} (R_m)^{-1} \quad (40)$$

with the α_m being the nonzero positive roots of the auxiliary equation presented in Table I.

Using the data of Table IV the first four roots of the auxiliary equation were found to be $\alpha_1 = 2.547$, $\alpha_2 = 3.232$, $\alpha_3 = 6.216$, and $\alpha_4 = 6.798$.

Figure 4 shows the concentration profiles for example 3 at various times. As was the case with example 2 there is a minimum in the concentration at moderate times. This minimum moves in the direction of $x = 0$ as time goes by. The minimum, however, never appears at $x = 0$, because the boundary condition $\partial C_1/\partial x = 0$, at $x = 0$, does not apply in this example. As t (or τ) becomes large the minimum approaches its final position within the ternary laminate slab. In the particular case of the Table IV data the minimum ends up in layer 1. The location of the minimum at large times is found by setting $\partial C_1/\partial\xi_1 = 0$ and considering only the first term of the infinite series of eq. (40), since second and subsequent terms can be neglected.

Thus

$$\frac{\partial C_1}{\partial\xi_1} \simeq \left\{ \alpha_1 C_1^s \left[1 + \delta_{23}K_{23}\sin\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_1\right)\sin\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_1\right) - \cos\left(\frac{\delta_{12}}{\lambda_{12}}\alpha_1\right)\cos\left(\frac{\delta_{13}}{\lambda_{13}}\alpha_1\right) \right] \cos(\alpha_1\xi_1)\exp(-\alpha_1^2\tau) \right\} (R_1)^{-1} = 0$$

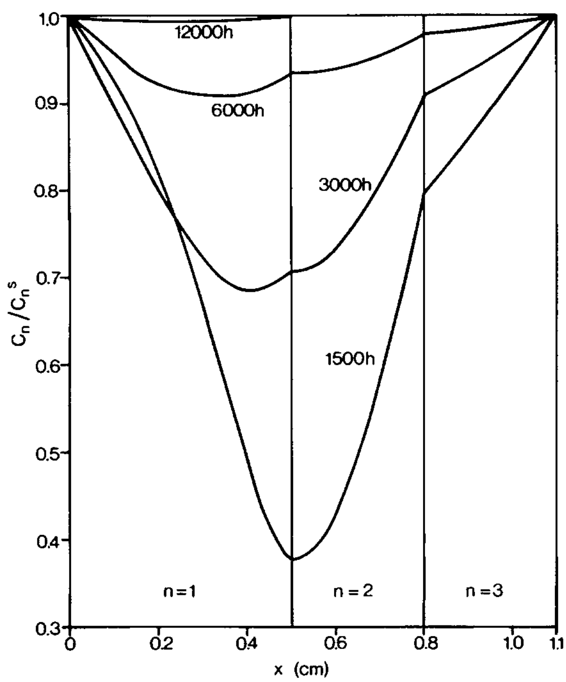

Fig. 4. Concentration profiles at various times for example 3.

The exponential term is small, but nonzero. Thus to find the position of the minimum at large t (or τ) it is necessary to solve the equation

$$\cos(\alpha_1\xi_1) = 0$$

for the first nonzero value of ξ_1. Hence we need to find ξ_1 from

$$\alpha_1\xi_1 = \tfrac{1}{2}\pi \tag{41}$$

From eq. (41) it is found that $\xi_1 = 0.617$ or $x = 0.309$ cm. As Figure 4 shows, the minimum does indeed reach its final resting place at this position. Note that if the first nonzero value of ξ_1 from eq. (41) had been greater than unity, it would have implied that the minimum was in layer 2 or 3. It would have been necessary to apply the same procedure of truncation, differentiation, and setting to zero to C_2 and C_3 until either $0 \leqslant \xi_2 < 1$ or $0 < \xi_3 \leqslant 1$ had been found.

Finally, Figure 2 shows the concentration profile at 3000 h for example 3, along with examples 1 and 2. This figure provides a useful check for the correctness of some of the complicated solutions that have been presented in this paper. For instance, it will be seen that the concentration profiles in layers 2 and 3 are virtually identical for examples 2 and 3, even though the solutions to the diffusion equations are very different, as are the set of roots α_m and β_m, $m = 1, 2, 3, 4$. The reason for the similarity is that the permeability coefficient (defined in general as $P_n = D_nC_n^s$) of the layer 1 is much lower than those of layers 2 and 3. Thus for moderate times at least, the concentration profiles in layers 2 and 3 could have been obtained with a fairly high degree of accuracy by discarding layer 1 and considering an impermeable substrate attached to layer 2 at $x = l_1$. However, although layer 1 has very little influence on the concentration profiles in layers 2 and 3, what happens inside layer 1 *is* important.

APPENDIX: APPLICATION OF THE LAPLACE TRANSFORM METHOD

The Laplace transform method of solving the diffusion (or heat conduction) equations is treated in detail by Carslaw and Jaeger.[4] However, because the coordinate system used in this paper is novel, the derivations of solutions to the diffusion equations are briefly outlined.

The Laplace transforms of the partial differential equation set (2), subject to the initial conditions (3), are

$$\begin{aligned} \frac{d^2\overline{C}_1}{d\xi_1^2} - p\overline{C}_1 &= -C_1^i, && 0 < \xi_1 < 1 \\ \frac{d^2\overline{C}_2}{d\xi_2^2} - \frac{\delta_{12}^2}{\lambda_{12}^2}p\overline{C}_2 &= -\frac{\delta_{12}^2}{\lambda_{12}^2}C_2^i, && 1 > \xi_2 > 0 \\ \frac{d^2\overline{C}_3}{d\xi_3^2} - \frac{\delta_{13}^2}{\lambda_{13}^2}p\overline{C}_3 &= -\frac{\delta_{13}^2}{\lambda_{13}^2}C_3^i, && 1 > \xi_3 > 0 \end{aligned} \tag{A1}$$

where p is the variable of the Laplace transform $\overline{C}_n(p)$ of $C_n(\tau)$.

System I

The boundary conditions when transformed become

$$\overline{C}_3 = C_3^0/p, \quad \xi_3 = 0; \qquad \overline{C}_1 = C_1^0/p, \quad \xi_1 = 0 \tag{A2}$$

$$(\overline{C}_1)_{\xi_1=1} = K_{12}(\overline{C}_2)_{\xi_2=1}; \qquad (\overline{C}_2)_{\xi_2=0} = K_{23}(\overline{C}_3)_{\xi_3=1} \tag{A3}$$

$$-\frac{\delta_{12}^2}{\lambda_{12}}\left(\frac{d\overline{C}^1}{d\xi_1}\right)_{\xi_1=1} = \left(\frac{d\overline{C}_2}{d\xi_2}\right)_{\xi_2=1}; \qquad \frac{\delta_{23}{}^2}{\lambda_{23}}\left(\frac{d\overline{C}_2}{d\xi_2}\right)_{\xi_2=0} = \left(\frac{d\overline{C}_3}{d\xi_3}\right)_{\xi_3=1} \tag{A4}$$

Solutions of eq. (A1) which satisfy eqs. (A2)–(A4) are

$$\overline{C}_3 = \frac{C_3^0 - C_3^i}{p}\cosh\left(\frac{\delta_{13}}{\lambda_{13}}q\xi_3\right) + A^\dagger \sinh\left(\frac{\delta_{13}}{\lambda_{13}}q\xi_3\right) + \frac{C_3^i}{p}$$

$$\overline{C}_2 = B^\dagger \cosh\left(\frac{\delta_{12}}{\lambda_{12}}q\xi_2\right) + E^\dagger \sinh\left(\frac{\delta_{12}}{\lambda_{12}}q\xi_2\right) + \frac{C_2^i}{p}$$

$$\overline{C}_1 = \frac{C_1^0 - C_1^i}{p}\cosh(q\xi_1) + F^\dagger \sinh(q\xi_1) + \frac{C_1^i}{p}$$

where $q = p^{1/2}$.

The factors $A^\dagger$, $B^\dagger$, $E^\dagger$, and $F^\dagger$ are found from eqs. (A3) and (A4).

System II

Boundary conditions (A3) and (A4) are retained, but condition (A2) is replaced by

$$\overline{C}_3 = C_3^0/p, \quad \xi_3 = 0; \qquad d\overline{C}_1/d\xi_1 = 0, \quad \xi_1 = 0 \tag{A5}$$

Solutions of eq. (A1) which satisfy eqs. (A3)–(A5) are

$$\overline{C}_3 = \frac{C_3^0 - C_3^i}{p}\cosh\left(\frac{\delta_{13}}{\lambda_{13}}q\xi_3\right) + G^\dagger \sinh\left(\frac{\delta_{13}}{\lambda_{13}}q\xi_3\right) + \frac{C_3^i}{p}$$

$$\overline{C}_2 = H^\dagger \quad \cosh[(\delta_{12}/\lambda_{12})q\xi_2] + M^\dagger \quad \sinh[(\delta_{12}/\lambda_{12})q\xi_2] + C_2^i/p$$

$$\overline{C}_1 = N^\dagger \quad \cosh(q\xi_1) + C_1^i/p$$

The factors $G^\dagger$, $H^\dagger$, $M^\dagger$, and $N^\dagger$ are found from eqs. (A3) and (A4).

System III

Boundary conditions eqs. (A3) and (A4) are retained, but eq. (A5) is replaced by

$$\frac{d\overline{C}_3}{d\xi_3} = 0, \qquad \xi_3 = 0; \qquad \frac{d\overline{C}_1}{d\xi_1} = 0, \qquad \xi_1 = 0 \tag{A6}$$

Solutions of eqs. (A1) which satisfy eqs. (A3), (A4), and (A6) are

$$\overline{C}_3 = P^\dagger \quad \cosh\,[(\delta_{13}/\lambda_{13})q\xi_3] + C_3^i/p$$

$$\overline{C}_2 = Q^\dagger \quad \cosh\,[(\delta_{12}/\lambda_{12})q\xi_2] + U^\dagger \quad \sinh[(\delta_{12}/\lambda_{12})q\xi_2] + C_2^i/p$$

$$\overline{C}_1 = V^\dagger \quad \cosh(q\xi_1) + C_1^i/p$$

The factors $P^\dagger$, $Q^\dagger$, $U^\dagger$, and $V^\dagger$ are found from eqs. (A3) and (A4)

To evalute $C_1(\tau)$, $C_2(\tau)$, and $C_3(\tau)$ for systems I, II and III, the Inversion theorem is used. All systems have simple poles at $p = 0$. The other poles for systems I, II, and III, lie, respectively, at $p = -\alpha_m^2$, $-\beta_m^2$, and $-\gamma_m^2$.

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