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Giorgio Carta[a]; Rebecca K. Lewus[a]
[a] DEPARTMENT OF CHEMICAL ENGINEERING, UNIVERSITY OF VIRGINIA, CHARLOTTESVILLE, VIRGINIA, USA

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Film Model Approximation for Particle-Diffusion-Controlled Multicomponent Ion Exchange

GIORGIO CARTA* and REBECCA K. LEWUS
DEPARTMENT OF CHEMICAL ENGINEERING
UNIVERSITY OF VIRGINIA
CHARLOTTESVILLE, VIRGINIA 22903-2442, USA

ABSTRACT

A film-model approximation is developed to predict multicomponent ion exchange in spherical particles. Mass transfer is represented by pseudosteady-state diffusion through a stagnant film of thickness equal to one-fifth of the particle radius. An exact analytic solution is found for the case of homovalent exchange and an approximate solution for the case of heterovalent exchange. Results obtained from the exact solution of the Nernst–Planck model are compared to those obtained using the film-model approximation for the case of ternary exchange in infinite-batch and fixed-bed systems. The predictions based on the film-model approximation are in good agreement with the exact solution for both homovalent and heterovalent ion exchange, indicating that the film-model approximation can be used effectively in numerical simulations of complex ion-exchange systems.

Key Words. Ion exchange; Nernst–Planck model; Linear driving force approximation

INTRODUCTION

It is well known that in ion-exchange processes the effective intraparticle diffusivity is a strong function of composition (1). Generally, for ion exchange on a fully ionized resin, intraparticle diffusion rates are described by the Nernst–Planck equations. Accordingly, the flux of an ion is the result of two additive contributions: one proportional to the ion concentration gradient and

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* To whom correspondence should be addressed.

the other proportional to the electrical potential gradient that develops when the diffusivities of the exchanging counterions are different. The prediction of ion-exchange rates using these equations has been addressed by several authors for both binary (2, 3) and ternary systems (4). The corresponding flux equations, however, are complicated and, in general, have to be solved numerically together with the particle conservation equations in spherical coordinates. This sometimes results in an unmanageable numerical complexity which hinders the implementation of effective codes for the simulation of complex systems (5).

Simplified approaches for the prediction of ion-exchange kinetics have been developed by several authors. For example, for the case of binary exchange, Rendueles de la Vega et al. (6) suggested the use of a constant "corrected Fick diffusivity" whose value is determined empirically by matching a numerical solution of the constant-diffusivity model to the numerical solution of the exact model. For the case of multicomponent exchange, Melis et al. (5) developed an approximation based on the assumption that the intraparticle concentration profiles are parabolic. This approximation was shown to be adequate for moderate values of the diffusivity ratios of the exchanging counterions. However, this approach fails when the diffusivities are very different since in this case the concentration profiles are far from parabolic (7). More recently, Carta et al. (7) developed a film model approximation for binary exchange which was shown to correctly predict the effects of the electric field on the exchange kinetics for both high and low values of the diffusivity ratio. Numerical results obtained for binary exchange in batch and column systems were shown to be in good agreement with those obtained from the numerical solution of the exact model.

In this paper we provide an extension of the film-model approximation to multicomponent systems. An exact analytic solution of the film-model equations is obtained for the case of homovalent exchange, yielding an explicit expression for the ion flux as a function of the average resin loadings and the surface concentrations. An approximate solution is also obtained for heterovalent exchange. In both cases, test numerical results obtained for ternary exchange in batch and column systems are found to be in good agreement with predictions based on the exact model, indicating that the film-model approximation can be used effectively in numerical simulations of complex ion-exchange systems.

THEORETICAL DEVELOPMENT

Multicomponent Model Development

We consider a completely ionized ion-exchange resin with M exchangeable counterions. The flux of ion i is described by

$$J_i = -\overline{D}_i \left(\frac{\partial q_i}{\partial r} + z_i q_i \frac{\mathfrak{F}}{\mathfrak{R}T} \frac{\partial \phi}{\partial r} \right) \tag{1}$$

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where ϕ is the electrical potential gradient. In dimensionless form, this equation can be written as

$$J_i^* = -\frac{1}{\alpha_i}\left(\frac{\partial y_i}{\partial \rho} + z_i y_i \frac{\partial \varphi}{\partial \rho}\right) \tag{2}$$

where $J_i^* = z_i J_i r_\mathrm{p}/\overline{D}_\mathrm{A} q_0$ is a dimensionless flux, y_i is the equivalent fraction of ion i, $\rho = r/r_\mathrm{p}$ is a dimensionless radial coordinate, and $\varphi = \Im\phi/\Re T$ is the dimensionless electrical potential. The parameter $\alpha_i = \overline{D}_\mathrm{A}/\overline{D}_i$ is the ratio of ion diffusivities for a reference counterion A and ion i. Electroneutrality and the condition of no net current transport yield the following equations:

$$\sum_{i=1}^{M} y_i = 1 \tag{3}$$

$$\sum_{i=1}^{M} J_i^* = 0 \tag{4}$$

Combining Eqs. (2)–(4) gives the following expressions for the flux:

$$J_i^* = -\sum_{j=1}^{M-1} \frac{1}{\alpha_{i,j}} \frac{\partial y_j}{\partial \rho} \tag{5}$$

$$\frac{1}{\alpha_{i,i}} = \frac{1}{\alpha_i} - \frac{\dfrac{1}{\alpha_i}\left(\dfrac{1}{\alpha_i} - \dfrac{1}{\alpha_M}\right) z_i y_i}{\displaystyle\sum_{k=1}^{M} \frac{1}{\alpha_k} z_k y_k} \tag{5a}$$

$$\frac{1}{\alpha_{i,j}} = -\frac{\dfrac{1}{\alpha_i}\left(\dfrac{1}{\alpha_j} - \dfrac{1}{\alpha_M}\right) z_i y_i}{\displaystyle\sum_{k=1}^{M} \frac{1}{\alpha_k} z_k y_k} \tag{5b}$$

These flux equations are used in conjunction with the relevant conservation equations and boundary conditions which may be written as follows.

For the particles:

$$\frac{\partial y_i}{\partial \tau} = -\frac{1}{\rho^2} \frac{\partial}{\partial \rho} (\rho^2 J_i^*) \tag{6}$$

$$\rho = 0: \qquad \frac{\partial y_i}{\partial \rho} = 0 \tag{6a}$$

$$\rho = 1: \qquad y_i = y_i^* \tag{6b}$$

$$\tau = 0: \qquad y_i = y_i^0 \tag{6c}$$

$$\bar{y}_i = 3\int_0^1 y_i \rho^2 d\rho \tag{6d}$$

where $\tau = t\overline{D}_\mathrm{A}/r_\mathrm{p}^2$ is a dimensionless time and y_i^* and y_i^0 are the interfacial and initial ionic fractions of ion *i*, respectively.

For a column:

$$\frac{\partial \bar{y}_i}{\partial \tau'} + \frac{\partial x_i}{\partial \xi} = 0 \tag{7}$$

$$\frac{\partial \bar{y}_i}{\partial \tau'} = -3J_i^*\,|_{\rho=1} \tag{7a}$$

$$\xi = 0: \qquad x_i = x_i^\mathrm{F} \tag{7b}$$

$$\tau' = 0: \qquad \bar{y}_i = y_i^0 \tag{7c}$$

where $\tau' = \tau - \xi/\Lambda$, $\xi = \Lambda\overline{D}_\mathrm{A} z/v r_\mathrm{p}^2$, and $\Lambda = (1-\varepsilon)q_0/\varepsilon C_0$. External mass transfer resistances and axial dispersion are neglected in these equations. For an ideal system, y_i^* is related to the fluid-phase equivalent fraction by the mass action law:

$$K_{i,\mathrm{A}}\left(\frac{q_0}{C_0}\right)^{z_i - z_A} = \frac{(y_i^*)^{z_\mathrm{A}}\,(x_\mathrm{A})^{z_i}}{(y_A^*)^{z_i}\,(x_i)^{z_\mathrm{A}}} \tag{8}$$

Dimensional forms of these equations can be found in Ref. 1. A numerical solution is generally required, and in this work it was obtained by finite differences as discussed by Carta et al. (7).

Film-Model Approximation

The film-model approximation is based on the recognition of the fact that for adsorption and ion-exchange calculations, the intraparticle mass transfer resistance can be represented by the resistance offered by a flat solid film of thickness equal to $\frac{1}{5}$ of the particle radius through which pseudosteady-state diffusion occurs (7, 8). For constant diffusivity cases, this representation provides a result mathematically equivalent to the well-known linear driving force approximation (LDF) (9). Accordingly, the rate of mass transfer is proportional to the difference between the solute concentration at the adsorbent particle surface and the average concentration in the particle. However, as shown by Carta and Cincotti and Carta et al., even when the diffusivity varies with composition, integration of the pseudosteady-state conservation equations across the film provides useful quantitatively predictive rate equations. In this case a correction factor dependent on both surface and average concentrations, is multiplied times the linear driving force term to obtain the appropriate rate equation.

The film-model approximation for an ion-exchange system with M counterions can be derived from Eq. (2) following a treatment similar to that of Wildhagen et al. (10). Assuming pseudosteady-state diffusion across the film, we have

$$\frac{dJ_i^*}{d\rho} = 0 \tag{9}$$

Combining Eqs. (2) and (9) gives the differential equation

$$\frac{1}{z_i}\frac{d^2y_i}{d\rho^2} + \frac{dy_i}{d\rho}\frac{d\varphi}{d\rho} + y_i\frac{d^2\varphi}{d\rho^2} = 0 \tag{10}$$

with boundary conditions

$$\rho = 1: \qquad y_i = y_i^* \tag{10a}$$

$$\rho = 1 - \frac{1}{\delta}: \qquad y_i = \bar{y}_i \tag{10b}$$

Summing Eq. (10) over all counterions gives the result

$$\frac{d^2\varphi}{d\rho^2} = -\sum_{j=1}^{M}\frac{1}{z_j}\frac{d^2y_j}{d\rho^2} \tag{11}$$

Exact Solution for Homovalent Exchange

For the case of homovalent exchange, Eq. (11) reduces to

$$\frac{d^2\varphi}{d\rho^2} = 0 \tag{12}$$

or

$$z_i\varphi = m\rho + b \tag{13}$$

Combining Eqs. (2), (9), and (13) yields

$$\frac{dy_i}{d\rho} + my_i = \gamma_i \tag{14}$$

where γ_i is an integration constant. This equation can be integrated across the film with boundary conditions (10a) and (10b) giving

$$\gamma_i = \frac{m(\bar{y}_i - y_i^* e^{m/\delta})}{1 - e^{m/\delta}} \tag{15}$$

The constant *m* is then obtained by imposing that the sum of the fluxes for all counterions is zero. After some algebraic manipulations we obtain

$$m = 5 \times \ln\left(\frac{\sum_{j=1}^{M} \bar{y}_j/\alpha_j}{\sum_{j=1}^{M} y_j^*/\alpha_j}\right) \tag{16}$$

$$J_i^* = -\frac{5}{\alpha_i}\ln\left(\frac{\sum_{j=1}^{M} y_j^*/\alpha_j}{\sum_{j=1}^{M} \bar{y}_j/\alpha_j}\right)\frac{y_i^*\left(\sum_{j=1}^{M}\bar{y}_j/\alpha_j\right) - \bar{y}_i\left(\sum_{j=1}^{M} y_j^*/\alpha_j\right)}{\left(\sum_{j=1}^{M} y_j^*/\alpha_j\right) - \left(\sum_{j=1}^{M}\bar{y}_j/\alpha_j\right)} \tag{17}$$

Finally, the film-model rate equation is obtained by combining Eq. (17) with an overall balance for the particle ($\partial\bar{y}_i/\partial\tau = -3J_i^*$), yielding

$$\frac{\partial\bar{y}_i}{\partial\tau} = \frac{15}{\alpha_i}\ln\left(\frac{\sum_{j=1}^{M}\bar{y}_j/\alpha_j}{\sum_{j=1}^{M} y_j^*/\alpha_j}\right)\frac{\bar{y}_i\left(\sum_{j=1}^{M} y_j^*/\alpha_j\right) - y_i^*\left(\sum_{j=1}^{M}\bar{y}_j/\alpha_j\right)}{\left(\sum_{j=1}^{M} y_j^*/\alpha_j\right) - \left(\sum_{j=1}^{M}\bar{y}_j/\alpha_j\right)} \tag{18}$$

When the diffusivities are all equal, Eq. (18) reduces to the classical LDF approximation $\partial\bar{y}_i/\partial\tau = 15(y_i^* - \bar{y}_i)$. However, when the diffusivities are different, Eq. (18) describes the effect of varying resin-phase composition on the coupled transport process. For batch ion-exchange with a constant surface composition, Eq. (18) can be integrated directly. For other situations, Eq. (18) is coupled to the appropriate conservation equations, for example, Eq. (7) for a fixed bed, replacing the partial differential equations for the particles (Eq. 6) with ordinary differential equations.

Approximate Solution for Heterovalent Exchange

An exact solution of the film-model equations for heterovalent exchange is apparently not possible. However, an approximate solution can be obtained by taking the electrical potential gradient to be equal to the average electrical potential difference across the film divided by the film thickness. Integrating Eq. (11) across the film gives the following result:

$$\frac{\Delta\varphi}{1/5} = m - 5\left(\sum_{j=1}^{M}\frac{1}{z_j}y_j^* - \sum_{j=1}^{M}\frac{1}{z_j}\bar{y}_j\right) = m' \tag{19}$$

Taking $d\varphi/d\rho \sim 5\Delta\varphi$ gives the following result for the flux of ion *i*:

$$J_i^* = -\frac{z_i}{\alpha_i}\frac{m'(\bar{y}_i - y_i^* e^{m'z_i/5})}{1 - e^{m'z_i/5}} \tag{20}$$

The constant m' is obtained by imposing that Eq. (4) is satisfied, which yields

$$\sum_{j=1}^{M}\frac{z_j}{\alpha_j}\frac{(\bar{y}_j - y_j^* e^{m'z_j/5})}{1 - e^{m'z_i/5}} = 0 \tag{21}$$

A trial-and-error procedure is needed to find the value of m' that satisfies this equation.

RESULTS AND DISCUSSION

In order to ascertain the usefulness of the film-model approximation for multicomponent ion exchange, numerical calculations were performed for a ternary system (A, B, C) for batch exchange in a infinite batch and for a fixed bed with constant initial and feed concentrations.

Figure 1(a) shows the average resin loading for batch exchange for the case of diffusivity ratios equal to 1. The resin is assumed to be initially in C-form. In this case the film-model approximation coincides with the LDF approximation and, as expected, the predicted profiles are in reasonable agreement with those obtained from the numerical solution of the exact model, Eqs. (2)–(6). Figure 1(b) shows a comparison of the film-model prediction and the numerical solution of the exact model for diffusivity ratios $\bar{D}_A/\bar{D}_B = 0.1$ and $\bar{D}_A/\bar{D}_C = 0.01$. These ratios imply that A is the slowest counterion and C is the fastest. Since the resin is assumed to be initially in C-form, exchange of the faster counterions C and B occurs rapidly for short times. This causes an overshoot of the B average concentration in the particle above the final equilibrium value (0.5 in this case). For longer times, essentially all of the C is removed from the resin and counterdiffusion of A and B occurs slowly, being largely controlled by the low diffusivity of A. The results of the numerical solution of the exact model and those of the film model approximation are again in reasonable agreement, indicating that this approximation can be used as a simplified rate law for multicomponent ion-exchange calculations. Results for binary exchange were obtained previously by Carta et al. (7), and similar conclusions can be expected for systems with more than three counterions.

Figure 2 shows average resin loadings for the case of heterovalent exchange with $z_A = z_C = 1$, $z_B = 2$, $\bar{D}_A/\bar{D}_B = 11.4$, and $\bar{D}_A/\bar{D}_C = 0.0667$. These values correspond to an experimental system considered by Yoshida and Kataoka (4) involving the exchange of Na^+ (A), Zn^{2+} (B), and H^+ (C) on a strong-acid

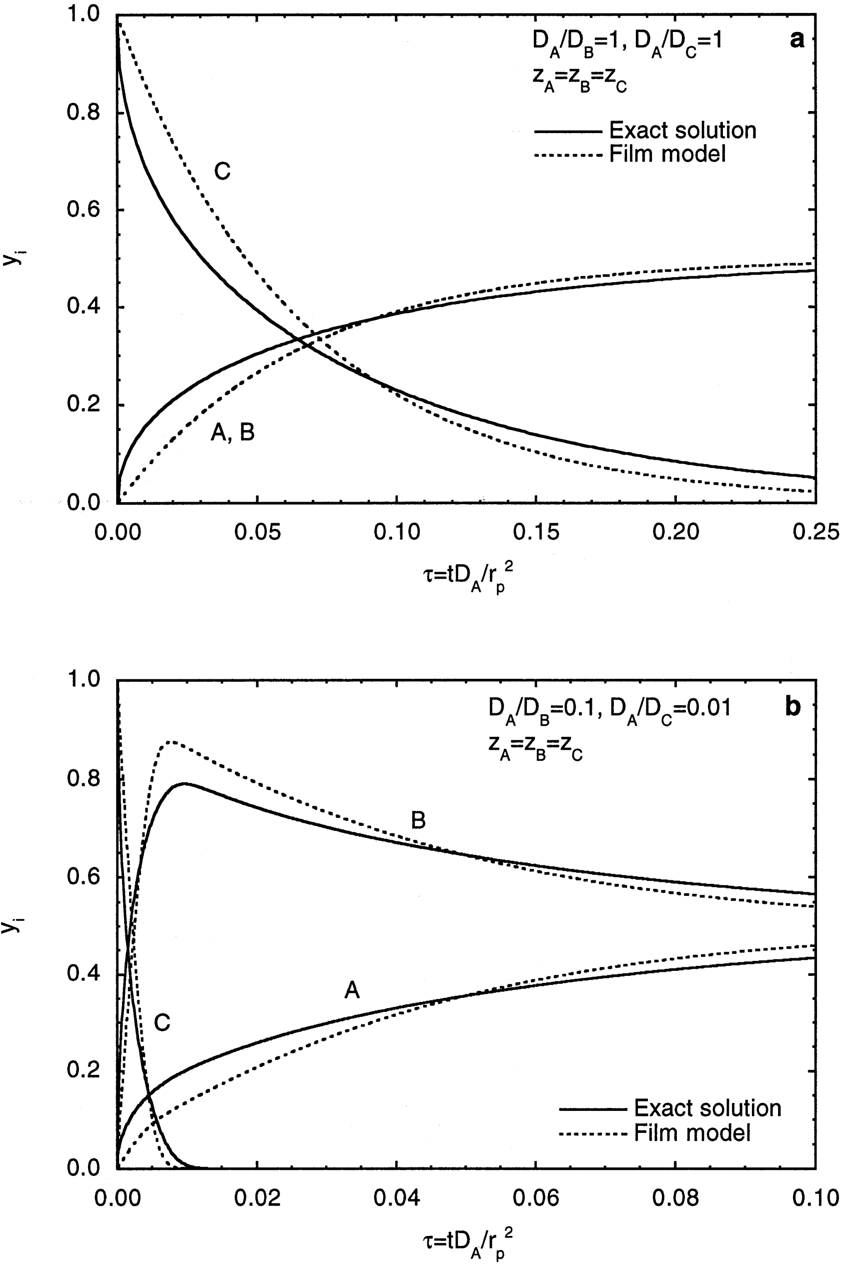

FIG. 1 Comparison of exact solution of Nernst–Planck model and film model approximation for ternary batch exchange in an infinite bath with $y_A^0 = y_B^0 = 0$ and $y_A^* = y_B^* = 0.5$. (a) Equal diffusivities, $\alpha_B = \alpha_C = 1$; (b) unequal diffusivities, $\alpha_B = 0.1$, $\alpha_C = 0.01$.

cation exchanger. Figure 2(a) shows the results for a mixture of A and B displacing C, while Fig. 2(b) shows the results for a mixture of B and C displacing A. In both cases the solution of the film-model approximation and the numerical solution of the exact model are in satisfactory agreement. The latter, of course, is much simpler, requiring only the solution of a system of three or-

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dinary differential equations. The numerical solution of the exact model is in good agreement with the results of Yoshida and Kataoka.

In the batch exchange the film model tends to underpredict the rate for short times and to overpredict it for long times. This occurs because for short times

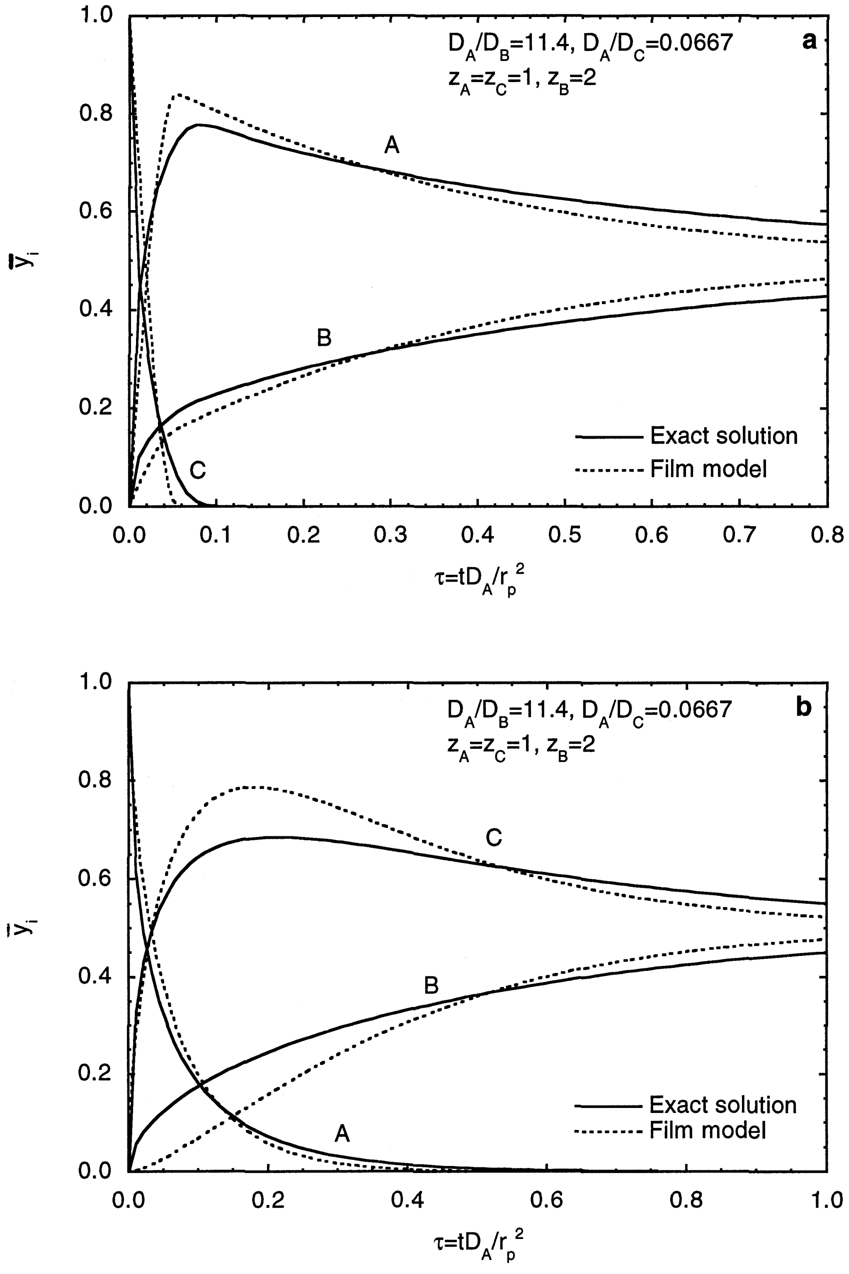

FIG. 2 Comparison of exact solution of Nernst–Planck model and film model approximation for ternary batch exchange in an infinite bath with $z_A = z_C = 1$, $z_B = 2$, $\alpha_B = 11.4$, and $\alpha_C = 0.0667$. Conditions correspond to the experimental system of Yoshida and Kataoka (4). (a) Resin initially in C-form: $y_A^0 = y_B^0 = 0$, $y_A^* = y_B^* = 0.5$; (b) resin initially in A-form: $y_B^0 = y_C^0 = 0$, $y_B^* = y_C^* = 0.5$.

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the depth of penetration predicted by the exact model is less than the value $r_p/5$ assumed by the film model. As time progresses, the depth of penetration predicted by the exact solution becomes greater than $r_p/5$. On the average, however, the film model is in reasonable agreement with the exact solution.

Figure 3(a) shows the breakthrough behavior in a fixed bed for the same system as in Fig. 1(a) assuming equal diffusivities, predicted using the film-

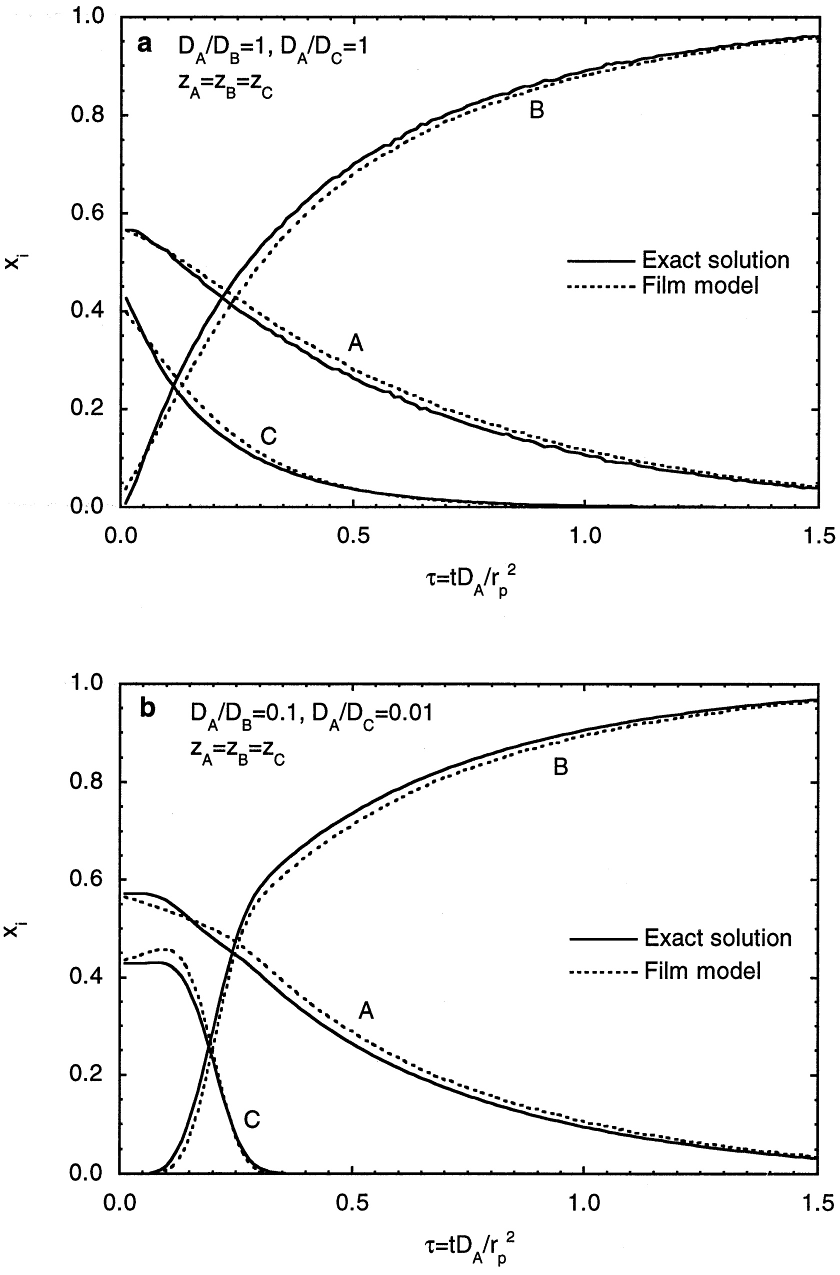

FIG. 3 Comparison of exact solution of Nernst–Planck model and film model approximation for ternary exchange in a fixed bed with $x_A^0 = 0.57$, $x_B^0 = 0$, $x_B^F = 1$, $\Lambda = 75$, and $\xi = 0.667$. (a) Equal diffusivities, $\alpha_B = \alpha_C = 1$; (b) different diffusivities, $\alpha_B = 0.1$, $\alpha_C = 0.01$.

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model approximation and the exact model. Selectivity coefficients were assumed to be $K_{B,A} = K_{C,A} = 0.333$. The resin was assumed to be initially loaded with a mixture of A and C and is displaced by ion B. As expected from the LDF approximation, the film-model approximation is in good agreement with the numerical solution of the exact model. Figure 3(b) shows the same result for the case of diffusivity ratios $\overline{D}_A/\overline{D}_B = 0.1$ and $\overline{D}_A/\overline{D}_C = 0.01$ as in Fig. 1(b). In this case, as a result of the higher diffusivities of B and C, a sharper wave corresponding to the exchange of B and C ions is observed at short times. For longer times, when C is essentially completely eluted from the bed, the exchange of A and B continues to occur at a slow rate, giving rise to shallow effluent profiles. As seen in Figs. 3(a) and 3(b), the agreement between the film model and the exact solution for the fixed bed is, in fact, better than in the batch. This is because, during the development of the mass transfer zone in the resin bed, the overprediction of the exchange rates by the film model at the upstream end of the column (comparable to long times in the batch system) compensates for the underprediction of the exchange rates at the downstream end of the column (comparable to short times in the batch system). Thus, the effluent concentration given by the film model is in substantial agreement with the exact solution at all times and appears to capture quantitatively the effect of electrical coupling of the diffusion fluxes while providing a computationally simple rate law.

CONCLUSIONS

A new approximate rate equation has been obtained for multicomponent ion exchange. The approximation is based on an equivalent film-resistance model. When used to predict batch uptake kinetics and fixed-bed ion-exchange behavior, this model is shown to provide results of accuracy comparable to that obtained when using the classical LDF approximation for systems with constant diffusivity. While the agreement of the film-model approximation and the exact numerical solution is not perfect, in general the results are deemed to be of sufficient accuracy for practical fixed-bed calculations, especially in view of the significant simplification of the computational effort which is afforded by using the new approximate rate equation.

NOMENCLATURE

C_i	concentration of species i (mol/L)
C_0	equivalent concentration (equiv/L)
$\overline{D}_i$	ion diffusivity of species i in the exchanger (cm^2/s)
$\mathfrak{I}$	Faraday constant
J_i	equivalent flux of species i (equiv/cm^2·s)
J_i^*	dimensionless equivalent flux of species i ($= r_p z_i J_i/\overline{D}_A q_0$)
$K_{i,A}$	selectivity coefficient for exchange reaction

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q_i	concentration of species *i* in exchanger phase (mol/L)
q_0	exchanger capacity (equiv/L)
r	particle radial coordinate (cm)
r_p	particle radius (cm)
$\Re$	ideal gas constant
t	time (s)
T	temperature (K)
v	fluid velocity (cm/s)
z	bed length or axial coordinate (cm)
z_i	charge of species *i*
x_i	equivalent fraction of species *i* in solution ($= z_i C_i / C_0$)
x_i^F	equivalent fraction of species *i* in feed solution
y_i	equivalent fraction of species *i* in exchanger phase ($= z_i q_i / q_0$)
$\bar{y}_i$	average equivalent fraction of species *i* in exchanger phase
y_i^*	equilibrium equivalent fraction of species *i* in exchanger phase at particle surface

Greek Letters

α_i	diffusivity ratio ($= \overline{D}_\text{A}/\overline{D}_i$)
$\alpha_{i,j}$	defined by Eqs. (5a) and (5b)
ε	bed void fraction
ϕ	electric potential (V)
φ	dimensionless electrical potential ($= \Im\phi/\Re T$)
Λ	bed capacity factor [$= (1 - \varepsilon) q_0 / \varepsilon C_0$]
ρ	dimensionless radial coordinate ($= r/r_\text{p}$)
τ	dimensionless time ($= \overline{D}_\text{A} t / r_\text{p}^2$)
τ'	dimensionless time ($= \tau - \xi/\Lambda$)
ξ	dimensionless axial coordinate ($= \overline{D}_\text{A} (1 - \varepsilon) q_0 z / r_\text{p}^2 \varepsilon C_0 v$)

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