

On the Modeling of Miscible Flow in Multi-Component Porous Media

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Abstract. An analytical model of miscible flow in multi-component porous media is presented to demonstrate the influence of pore capacitance in extending diffusive tailing. Solute attenuation is represented naturally by accommodating diffusive and convective flux components in macropores and micropores as elicited by the local solute concentration and velocity fields. A set of twin, coupled differential equations result from the Laplace transform and are solved simultaneously using a differential operator for one-dimensional flow geometry. The solutions in real space are achieved using numeric inversion. In addition, to represent more faithfully the dominant physical processes, this approach enables efficient and stable semi-analytical solution procedure of the coupled system that is significantly more complex than current capacitance type models. Parametric studies are completed to illustrate the ability of the model to represent sharp breakthrough and lengthy tailing, as well as investigating the form of the nested heterogeneity as a result of solute exchange between macropores and micropores. Data from a laboratory column experiment is examined using the present model and satisfactory agreement results.

Key words: Miscible flow, micropore convection, micropore diffusion, heterogeneity, breakthrough curve.

0. Nomenclature

Roman Letters

- a* rate coefficient of internal flow
- b* velocity ratio (v_1/v_2)
- h* dispersion ratio (D_2/D_1)
- c_1 macropore concentration
- c_2 micropore concentration
- \bar{c}_1 macropore concentration in Laplace space
- \bar{c}_2 micropore concentration in Laplace space
- c_1^0 macropore concentration at source location
- c_2^0 micropore concentration at source location
- D_1 macropore dispersion coefficient
- D_2 micropore dispersion coefficient
- f* fraction of pore space occupied by fluid in primary channel
- L* length of laboratory sample column
- K* mass exchange rate
- t* time from initial stage
- v_1 primary flow channel velocity

v_2 micropore interstitial velocity
 x distance from source
 y dimensionless distance

Greek Letters

γ equivalent Péclet number
 τ dimensionless time, or injected pore volume

1. Introduction

For a homogeneous porous medium, miscible flow is usually defined in terms of a single fluid with the independent variable taken as the concentration. For multi-component media, where characteristic lengths are distinctly different between the flow-interacting units, two fluids remain separate and occupy their own volume, but fluids are interchangeable during the transport process due to their miscibility. It is generally recognized that heterogeneous porous media present capacitance effects during miscible displacement due to noninstantaneous equilibrium of concentration between mobile fluid in macropores and immobile fluid in micropores. The latter results from the stagnant fluid frequently isolated in dead-end pores. The geometric relationship between micropores and macropores is illustrated in Figure 1. Large open channels present in macropores comprise the primary flow pathways, with interstitial micropores acting as capacitors in storing or recharging the flow to the macropores, depending on the relative concentration gradient between the two component spaces.

The impact of heterogeneities at various scales precludes the use of the classical convection-dispersion approach because the transport process can not be represented by superposition of a simple diffusive process. Capacitance models have been applied to replicate the nonhomogeneous behavior observed in porous media exhibiting storage perturbation. Observation of abrupt solute breakthrough accompanied by extensive tailing is often attributed to the storage interchange between flowing liquid in macropores and stagnant fluid in micropores. (Deans, 1963; Coats and Smith, 1964; Passioura, 1971; Passioura and Rose, 1971; Piquemal, 1992, 1993). In addition to its utility in modeling tracer dispersion in naturally heterogeneous porous media, capacitance models has been used in the simulation of particle transport in partially clogged porous media where the heterogeneity may evolve with the progress of clogging. Consequently, local rectification of flow pathways corresponds to morphological and structural changes occurring in the clogged porous media (Bouhroum, 1993). Dual-porosity models provide a natural analog to capacitance models where fractures and matrix blocks are considered as interacting porous components. The rate of mass transfer depends on either the relative magnitude of concentration gradient, or the degree of permeability contrast between the two interacting media (Tang *et al.*, 1981; Bibby, 1981; Huyakorn *et al.*,

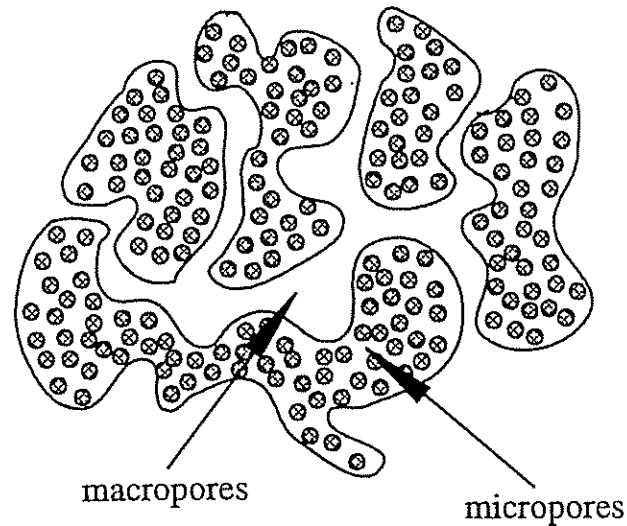


Fig. 1. Micropore-macropore media in capacitance model.

1983; Nilson and Lie, 1990; Rowe and Booker, 1990; Sudicky and McLaren, 1992; Harrison *et al.*, 1992; Leo and Booker, 1993).

Despite the wide utilization of the capacitance concept, these models provide poor representation of the extended tailing phenomenon observed in moderately and highly heterogeneous porous media (Joy and Kouwen, 1991; Imdakm and Sahimi, 1991; Koenders and Williams, 1992; Joy *et al.*, 1993). The lack of a physical basis in supporting the capacitance model was noted by Bouhroum (1993) in reference to experimental results. Bouhroum (1993) showed that the early solute penetration and long tailing of breakthrough curves were the result of velocity fluctuations induced by layers of different permeability and particle clogging, respectively. The existence of low permeability zones delays the rate of solute transport while simultaneously increasing the skewness of the breakthrough curves. Although these experimental results were adequately approximated by capacitance models through manipulation of input parameters, the fidelity of the match was more a result of the curve-fitting procedure rather than an adequate physical replication of actual transport processes. One undesirable feature of capacitance approaches rests on its physically spurious assumption that there is no flow within the micropore space. In reality, flow in the micropores should be either diffusive at low flow velocities or convective at high local velocities. Under more significant concentration gradients and at higher local flow velocities, both diffusion and convection need be included in representing transport in micropores. As pointed out by Udey and Spanos (1993) in the construction of a mathematical model for miscible flow in nonhomogeneous media, the two sets of equations (one representing macropores and another representing micropores) should be identical. In other words, the effects of diffusion (dispersion) and convection should be maintained for both macropore and micro-

pore spaces. The incorporation of micro-convection and micro-diffusion in the formulation of an alternative capacitance model constitutes the primary objective of the following work.

2. Analytical Formulation and Solutions

The capacitance model developed by Coats and Smith (1964) can be equivalently expressed as

$$D_1 \frac{\partial^2 c_1}{\partial x^2} - v_1 \frac{\partial c_1}{\partial x} = f \frac{\partial c_1}{\partial t} + K(c_1 - c_2), \quad (1)$$

$$0 = (1 - f) \frac{\partial c_2}{\partial t} - K(c_1 - c_2), \quad (2)$$

where c_1 and c_2 are the solute concentrations for macropores and micropores, respectively; D_1 is the macropore dispersion coefficient, v_1 is the average velocity in primary flow channel, f is the fraction of pore space occupied by mobile fluid, K is the rate of mass exchange between macropores and micropores, x is the distance from source, and t is the time. The zero on the left hand side of Equation (2) implies that liquid in the micropores is stagnant.

Considering the dispersive as well as convective flow processes within the micropores, two terms should be used to substitute the zero on the left-hand side of Equation (2). Therefore

$$D_2 \frac{\partial^2 c_2}{\partial x^2} - v_2 \frac{\partial c_2}{\partial x} = (1 - f) \frac{\partial c_2}{\partial t} - K(c_1 - c_2), \quad (3)$$

where D_2 is the micropore dispersion coefficient, and v_2 is the micropore interstitial velocity.

Equations (1) and (3) represent the governing equations of solute transport in porous media incorporating micropore dispersion and convection. It is more convenient to utilize dimensionless variables in the formulation by replacing the dimensional variables:

$$\begin{aligned} b &= \frac{v_1}{v_2}, & h &= \frac{D_2}{D_1}, \\ y &= \frac{x}{L}, & \tau &= \frac{v_1 t}{L}, \\ \gamma &= \frac{v_1 L}{D_2}, & a &= \frac{K L}{v_1}, \end{aligned} \quad (4)$$

where b is the coefficient representing the proportionality between flow velocities in macropores and micropores, h is the ratio of dispersion coefficients between micropores and macropores, L is an arbitrary length representing the longest possible

solute travel distance, y is the dimensionless distance, τ is the dimensionless time or injected pore volume, γ is the equivalent Peclet number considering the ratio between macropore velocity and micropore dispersion, and a is the rate coefficient of internal flow.

In dimensionless form, Equation (1) and (3) may be written as

$$\frac{1}{h\gamma} \frac{\partial^2 c_1}{\partial y^2} - \frac{\partial c_1}{\partial y} = f \frac{\partial c_1}{\partial \tau} + a(c_1 - c_2), \quad (5)$$

$$\frac{1}{\gamma} \frac{\partial^2 c_2}{\partial y^2} - \frac{1}{b} \frac{\partial c_2}{\partial y} = (1 - f) \frac{\partial c_2}{\partial \tau} - a(c_1 - c_2). \quad (6)$$

For application of a step change in solute concentration at one end and no variation in solute flux at the other end, the boundary and initial conditions may be defined as

$$\begin{aligned} c_1 &= c_1^0, c_2 = c_2^0 \quad (y = 0), \\ \frac{\partial c_1}{\partial y} &= \frac{\partial c_2}{\partial y} = 0 \quad (y = 1), \\ c_1 &= c_2 = 0 \quad (t = 0), \end{aligned} \quad (7)$$

where c_1^0 and c_2^0 are the macropore and micropore concentration at source location, respectively.

Using the Laplace transform, Equations (5) and (6) can be modified into ordinary differential equations and the time dependence removed as

$$\frac{1}{h\gamma} \frac{d^2 \bar{c}_1}{dy^2} - \frac{d\bar{c}_1}{dy} = fs\bar{c}_1 + a(\bar{c}_1 - \bar{c}_2), \quad (8)$$

$$\frac{1}{\gamma} \frac{d^2 \bar{c}_2}{dy^2} - \frac{1}{b} \frac{d\bar{c}_2}{dy} = (1 - f)s\bar{c}_2 - a(\bar{c}_1 - \bar{c}_2). \quad (9)$$

The boundary conditions in the Laplace domain are given as

$$\begin{aligned} \bar{c}_1 &= \frac{\bar{c}_1^0}{s}, \quad \bar{c}_2 = \frac{\bar{c}_2^0}{s} \quad (y = 0), \\ \frac{d\bar{c}_1}{dy} &= \frac{d\bar{c}_2}{dy} = 0 \quad (y = 1). \end{aligned} \quad (10)$$

The method of differential operators (*Mathematical Handbook*, 1979) is applied in this work to accommodate the coupled behavior of Equations (8) and (9). The differential operators, D^n , is applied as

$$D^n f(x_i) = \frac{d^n f(x_i)}{dx_i^n}, \quad (11)$$

where i indexes an arbitrary variable, and n is the order of the differential equations.

Applying the differential operators to Equations (8) and (9), gives

$$\left(\frac{D^2}{h\gamma} - D - \beta_2\right) \bar{c}_1 + a\bar{c}_2 = 0, \quad (12)$$

$$\left[\frac{D^2}{\gamma} - \frac{D}{b} + \beta_1\right] \bar{c}_2 + a\bar{c}_1 = 0, \quad (13)$$

where

$$\beta_1 = -[a + (1 - f)s], \quad \beta_2 = a + fs. \quad (14)$$

Solving Equations (12) and (13) simultaneously, yields

$$\bar{c}_1 = \frac{-1}{a} \left[\frac{D^2}{\gamma} - \frac{D}{b} + \beta_1\right] \bar{c}_2. \quad (15)$$

Substituting Equation (15) into (12) results in

$$(D^4 + B_1D^3 + B_2D^2 + B_3D + B_4)\bar{c}_2 = 0, \quad (16)$$

where

$$\begin{aligned} B_1 &= -\gamma \left(h + \frac{1}{b}\right), & B_2 &= h\gamma \left(\frac{\beta_1}{h} + \frac{\gamma}{b} - \beta_2\right), \\ B_3 &= h\gamma^2 \left(\frac{\beta_2}{b} - \beta_1\right), & B_4 &= -h\gamma^2(\beta_1\beta_2 + a^2). \end{aligned} \quad (17)$$

Equation (16) has four roots from the following equations (*Mathematical Handbook*, 1979):

$$D^2 + \xi_1 D + \phi_1 = 0, \quad (18)$$

$$D^2 + \xi_2 D + \phi_2 = 0, \quad (19)$$

where

$$\begin{aligned} \Delta_1 &= 8z + B_1^2 - 4B_2, \\ \xi_1 &= 0.5(B_1 + \sqrt{\Delta_1}), & \xi_2 &= 0.5(B_1 - \sqrt{\Delta_1}), \\ \phi_1 &= z + \frac{B_1 z - B_3}{\sqrt{\Delta_1}}, & \phi_2 &= z - \frac{B_1 z - B_3}{\sqrt{\Delta_1}}, \end{aligned} \quad (20)$$

where z is any real root of the following equation:

$$8z^3 + 4E_1z^2 + E_2z + E_3 = 0, \quad (21)$$

and where

$$\begin{aligned} E_1 &= -B_2, \\ E_2 &= 2B_1B_3 - 8B_4, \\ E_3 &= B_4(4B_2 - B_1^2) - B_3^2. \end{aligned} \quad (22)$$

Further, assume

$$z = u - \frac{E_1}{6}, \quad (23)$$

where the parameter u can be determined from the third-order equation

$$u^3 + pu + q = 0, \quad (24)$$

and where

$$\begin{aligned} p &= 0.125 \left(E_2 - \frac{2E_1^2}{3} \right), \\ q &= 0.125 \left(\frac{2E_1^3}{27} - \frac{E_1E_2}{6} + E_3 \right). \end{aligned} \quad (25)$$

The three roots of u may be described as

$$\begin{aligned} u_1 &= F_1 + F_2, \\ u_2 &= \omega_1 F_1 + \omega_2 F_2, \\ u_3 &= \omega_2 F_1 + \omega_1 F_2, \end{aligned} \quad (26)$$

where

$$\begin{aligned} \Delta^* &= \left(\frac{q}{2} \right)^2 + \left(\frac{p}{3} \right)^3, \\ F_1 &= \left(-\frac{q}{2} + \sqrt{\Delta^*} \right)^{\frac{1}{3}}, \quad F_2 = \left(-\frac{q}{2} - \sqrt{\Delta^*} \right)^{\frac{1}{3}}, \\ \omega_1 &= -0.5 + \frac{\sqrt{3}}{2}i, \quad \omega_2 = -0.5 - \frac{\sqrt{3}}{2}i. \end{aligned} \quad (27)$$

Three possible solutions of the real root, z , exist depending upon the signs of Δ^* in Equation (27), as indicated in the following:

When $\Delta^* > 0$, the real root is

$$z_1 = F_1 + F_2 - \frac{E_1}{6}. \quad (28)$$

If $\Delta^* = 0$, then the real root becomes

$$z_1 = -\sqrt[3]{4q} - \frac{E_1}{6}. \quad (29)$$

However, if $\Delta^* < 0$, the real root is recovered in trigonometric form as

$$z_1 = 2\sqrt[3]{r} \cos\left(\frac{\alpha}{3}\right) - \frac{E_1}{6} \quad (30)$$

where

$$r = \sqrt{-\left(\frac{p}{3}\right)^3},$$

$$\alpha = \arccos\left(-\frac{q}{2r}\right). \quad (31)$$

Once the real root, z , is determined for Equation (21), the four roots of Equations (18) and (19) can then be expressed as

$$\Delta_2 = \frac{\xi_1^2}{4} - \phi_1, \quad \Delta_3 = \frac{\xi_2^2}{4} - \phi_2,$$

$$\psi_1 = -\frac{\xi_1}{2} + \sqrt{\Delta_2}, \quad \psi_2 = -\frac{\xi_1}{2} - \sqrt{\Delta_2},$$

$$\psi_3 = -\frac{\xi_2}{2} + \sqrt{\Delta_3}, \quad \psi_4 = -\frac{\xi_2}{2} - \sqrt{\Delta_3}. \quad (32)$$

The solutions are further complicated as a result of uncertainty in the signs of Δ_1 , Δ_2 , and Δ_3 . For a choice of representative physical parameters, Δ_1 is predominantly positive. The solutions due merely to the changing signs of Δ_2 and Δ_3 are reported in the following. For completeness, the solution for the negative Δ_1 is described in the Appendix.

2.1. SOLUTIONS WHEN $\Delta_1 \geq 0$, $\Delta_2 \geq 0$ AND $\Delta_3 \geq 0$

Concentration in the micropores may be determined from Equation (16) as

$$\bar{c}_2 = g_1 e^{\psi_1 y} + g_2 e^{\psi_2 y} + g_3 e^{\psi_3 y} + g_4 e^{\psi_4 y}, \quad (33)$$

where constants g_1, g_2, g_3 and g_4 are determined by satisfying the boundary conditions. The concentration in the macropores can be derived from Equation (15) as

$$\bar{c}_1 = \frac{1}{a\gamma}(\eta_1 g_1 e^{\psi_1 y} + \eta_2 g_2 e^{\psi_2 y} + \eta_3 g_3 e^{\psi_3 y} + \eta_4 g_4 e^{\psi_4 y}), \quad (34)$$

where

$$\begin{aligned} \eta_1 &= -\psi_1^2 + \frac{\gamma\psi_1}{b} - \gamma\beta_1, & \eta_2 &= -\psi_2^2 + \frac{\gamma\psi_2}{b} - \gamma\beta_1, \\ \eta_3 &= -\psi_3^2 + \frac{\gamma\psi_3}{b} - \gamma\beta_1, & \eta_4 &= -\psi_4^2 + \frac{\gamma\psi_4}{b} - \gamma\beta_1. \end{aligned} \quad (35)$$

A system of equations can be constructed through satisfying the boundary conditions given in Equation (10). After solving the system of equations through the method of elimination, the constants g_1, g_2, g_3 and g_4 are expressed as

$$g_1 = \frac{H_1}{M}, \quad g_2 = \frac{H_2}{M}, \quad g_3 = \frac{H_3}{M}, \quad g_4 = \frac{H_4^\phi}{M}, \quad (36)$$

and assume:

$$\begin{aligned} \eta_5 &= \psi_1 e^{\psi_1}, & \eta_6 &= \psi_2 e^{\psi_2}, & \eta_7 &= \psi_3 e^{\psi_3}, & \eta_8 &= \psi_4 e^{\psi_4}, \\ \eta_9 &= \eta_1 \eta_5, & \eta_{10} &= \eta_2 \eta_6, & \eta_{11} &= \eta_3 \eta_7, & \eta_{12} &= \eta_4 \eta_8 \\ \zeta_1 &= \frac{\bar{c}_1^0 a \gamma}{s}, & \zeta_2 &= \frac{\bar{c}_2^0}{s}, \end{aligned} \quad (37)$$

where

$$\begin{aligned} M &= (\eta_2 - \eta_1)(\eta_7 \eta_{12} - \eta_8 \eta_{11}) + (\eta_1 - \eta_3)(\eta_6 \eta_{12} - \eta_8 \eta_{10}) \\ &\quad + (\eta_4 - \eta_1)(\eta_6 \eta_{11} - \eta_7 \eta_{10}) + (\eta_3 - \eta_2)(\eta_5 \eta_{12} - \eta_8 \eta_9) \\ &\quad + (\eta_2 - \eta_4)(\eta_5 \eta_{11} - \eta_7 \eta_9) + (\eta_4 - \eta_3)(\eta_5 \eta_{10} - \eta_6 \eta_9). \end{aligned} \quad (38)$$

$$\begin{aligned} H_1 &= (\zeta_2 \eta_2 - \zeta_1)(\eta_7 \eta_{12} - \eta_8 \eta_{11}) + (\zeta_1 - \zeta_2 \eta_3)(\eta_6 \eta_{12} - \eta_8 \eta_{10}) \\ &\quad + (\zeta_2 \eta_4 - \zeta_1)(\eta_6 \eta_{11} - \eta_7 \eta_{10}) \end{aligned} \quad (39)$$

$$\begin{aligned} H_2 &= (\zeta_1 - \zeta_2 \eta_1)(\eta_7 \eta_{12} - \eta_8 \eta_{11}) + (\zeta_2 \eta_3 - \zeta_1)(\eta_5 \eta_{12} - \eta_8 \eta_9) \\ &\quad + (\zeta_1 - \zeta_2 \eta_4)(\eta_5 \eta_{11} - \eta_7 \eta_9). \end{aligned} \quad (40)$$

$$H_3 = (\zeta_2 \eta_1 - \zeta_1)(\eta_6 \eta_{12} - \eta_8 \eta_{10}) + (\zeta_1 - \zeta_2 \eta_3)(\eta_5 \eta_{12} - \eta_8 \eta_9)$$

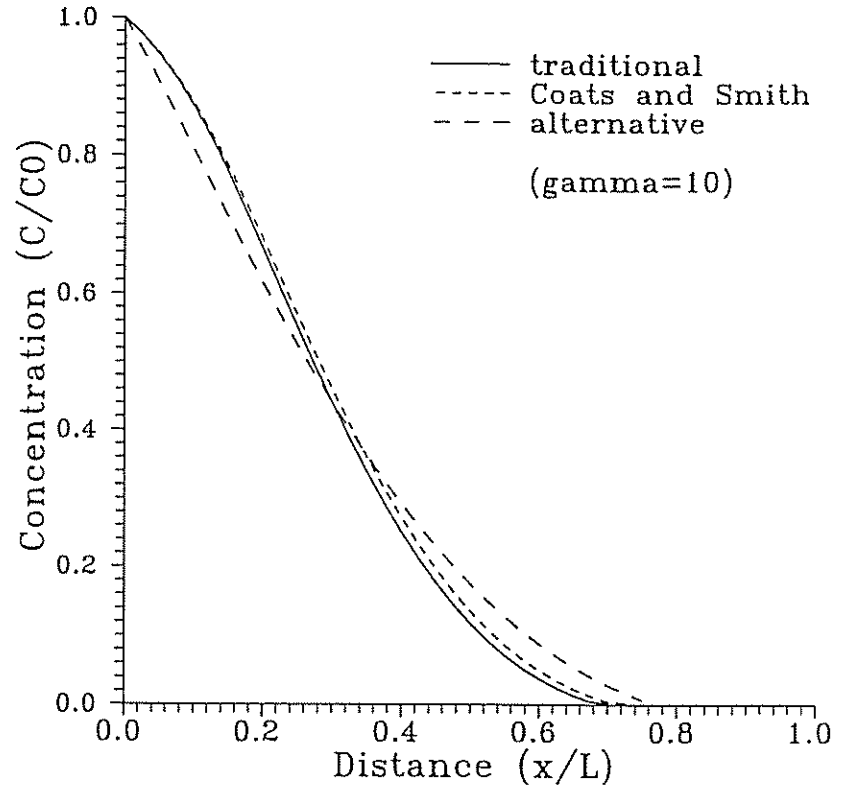


Fig. 2. Comparison of spatial concentration when $\gamma = 10$.

$$+ (\zeta_2 \eta_4 - \zeta_1)(\eta_5 \eta_{10} - \eta_6 \eta_9) \quad (41)$$

$$H_4 = (\zeta_1 - \zeta_2 \eta_1)(\eta_6 \eta_{11} - \eta_7 \eta_{10}) + (\zeta_2 \eta_2 - \zeta_1)(\eta_5 \eta_{11} - \eta_7 \eta_9)$$

$$+ (\zeta_1 - \zeta_2 \eta_3)(\eta_5 \eta_{10} - \eta_6 \eta_9). \quad (42)$$

2.2. SOLUTIONS WHEN $\Delta_1 \geq 0$, $\Delta_2 < 0$ AND $\Delta_3 \geq 0$

For this condition, two of the four roots from Equations (18) and (19) become complex variables, and may be rewritten as

$$\psi_1 = -\frac{\xi_1}{2} + i\sqrt{-\Delta_2}, \quad \psi_2 = -\frac{\xi_1}{2} - i\sqrt{-\Delta_2}. \quad (43)$$

ψ_3 and ψ_4 are unchanged from Equation (32). The concentration in micropores can be alternatively expressed as

$$\bar{c}_2 = g_1 e^{w_1 y} \cos(w_2 y) + g_2 e^{w_1 y} \sin(w_2 y) + g_3 e^{\psi_3 y} + g_4 e^{\psi_4 y}, \quad (44)$$

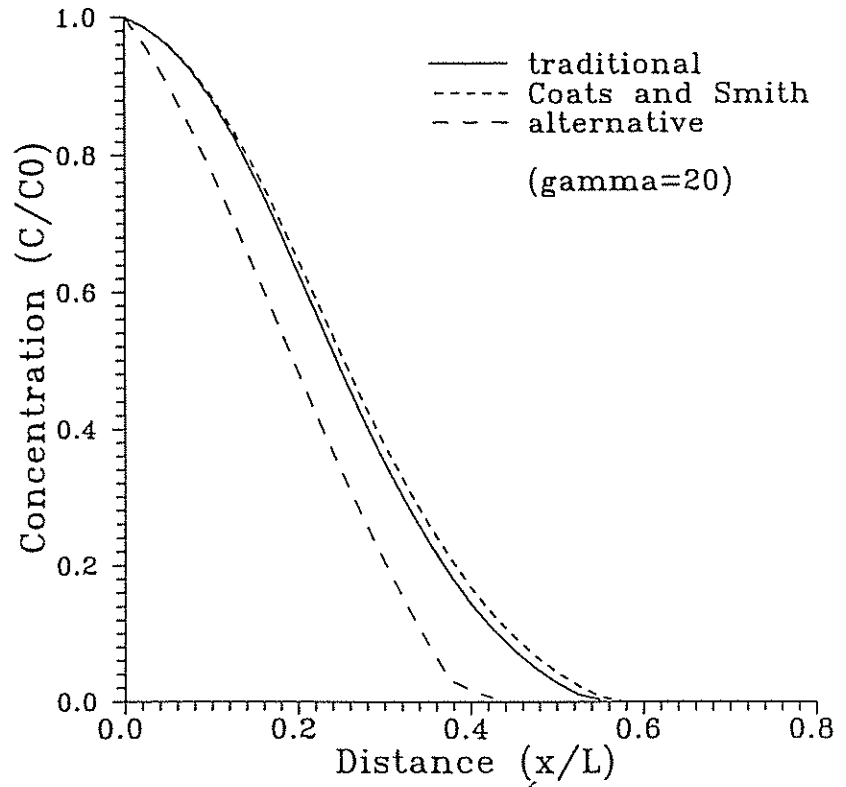


Fig. 3. Comparison of spatial concentration when $\gamma = 20$.

where

$$w_1 = -\frac{\xi_1}{2}, \quad w_2 = \sqrt{-\Delta_2}. \quad (45)$$

Similarly, the concentration within the macropores is described as

$$\begin{aligned} \bar{c}_1 = & \frac{1}{a\gamma} \{g_1 e^{w_1 y} [\lambda_1 \cos(w_2 y) - \lambda_2 \sin(w_2 y)] \\ & + g_2 e^{w_1 y} [\lambda_1 \sin(w_2 y) + \lambda_2 \cos(w_2 y)] \\ & + \lambda_3 g_3 e^{\psi_3 y} + \lambda_4 g_4 e^{\psi_4 y}\}, \end{aligned} \quad (46)$$

where

$$\begin{aligned} \lambda_1 = & -w_1^2 + w_2^2 + w_1 \frac{\gamma}{b} - \gamma \beta_1, & \lambda_2 = & -w_2 \left(2w_1 - \frac{\gamma}{b}\right), \\ \lambda_3 = & -\psi_3^2 + \frac{\gamma \psi_3}{b} - \gamma \beta_1, & \lambda_4 = & -\psi_4^2 + \frac{\gamma \psi_4}{b} - \gamma \beta_1. \end{aligned} \quad (47)$$

For convenience, assume

$$\begin{aligned}
\alpha_1 &= e^{w_1}[w_1 \cos(w_2) - w_2 \sin(w_2)], \\
\alpha_2 &= e^{w_1}[w_1 \sin(w_2) + w_2 \cos(w_2)], \\
\alpha_3 &= \psi_3 e^{\psi_3}, \quad \alpha_4 = \psi_4 e^{\psi_4}, \\
\alpha_5 &= e^{w_1}[(\lambda_1 w_1 - \lambda_2 w_2) \cos(w_2) - (\lambda_2 w_1 + \lambda_1 w_2) \sin(w_2)], \\
\alpha_6 &= e^{w_1}[(\lambda_1 w_2 + \lambda_2 w_1) \cos(w_2) + (\lambda_2 w_1 - \lambda_1 w_2) \sin(w_2)], \\
\alpha_7 &= \lambda_3 \alpha_3, \quad \alpha_8 = \lambda_4 \alpha_4.
\end{aligned} \tag{48}$$

Following a similar procedure as previously, the constants g_1, g_2, g_3 and g_4 in Equations (44) and (46) are derived as in Equation (36) but with the substitution of M and $H_i (i = 1, 2, 3, 4)$ as

$$\begin{aligned}
M &= (\alpha_2 \alpha_7 - \alpha_3 \alpha_6)(\lambda_1 + \lambda_4) + (\alpha_2 \alpha_8 - \alpha_4 \alpha_6) \\
&\quad (\lambda_1 - \lambda_3) + (\alpha_1 \alpha_6 - \alpha_2 \alpha_5)(\lambda_4 - \lambda_3) \\
&\quad + \lambda_2[\alpha_8(\alpha_3 - \alpha_1) + \alpha_4(\alpha_5 - \alpha_7) + \alpha_1 \alpha_7 - \alpha_3 \alpha_5].
\end{aligned} \tag{49}$$

$$\begin{aligned}
H_1 &= \zeta_2[\lambda_2(\alpha_3 \alpha_8 - \alpha_4 \alpha_7) - \lambda_3(\alpha_2 \alpha_8 - \alpha_4 \alpha_6) + \lambda_4(\alpha_2 \alpha_7 - \alpha_3 \alpha_6)] \\
&\quad - \zeta_1(\alpha_4 \alpha_6 - \alpha_2 \alpha_8 + \alpha_2 \alpha_7 - \alpha_3 \alpha_6).
\end{aligned} \tag{50}$$

$$\begin{aligned}
H_2 &= -\zeta_2[\lambda_1(\alpha_3 \alpha_8 - \alpha_4 \alpha_7) - \lambda_3(\alpha_1 \alpha_8 - \alpha_4 \alpha_5) + \lambda_4(\alpha_1 \alpha_7 - \alpha_3 \alpha_5)] \\
&\quad + \zeta_1(\alpha_3 \alpha_8 - \alpha_4 \alpha_7 - \alpha_1 \alpha_8 + \alpha_4 \alpha_5 + \alpha_1 \alpha_7 - \alpha_3 \alpha_5).
\end{aligned} \tag{51}$$

$$\begin{aligned}
H_3 &= \zeta_2[\lambda_1(\alpha_2 \alpha_8 - \alpha_4 \alpha_6) - \lambda_2(\alpha_1 \alpha_8 - \alpha_4 \alpha_5) + \lambda_4(\alpha_1 \alpha_6 - \alpha_2 \alpha_5)] \\
&\quad - \zeta_1(\alpha_2 \alpha_8 - \alpha_4 \alpha_6 + \alpha_1 \alpha_6 - \alpha_2 \alpha_5).
\end{aligned} \tag{52}$$

$$\begin{aligned}
H_4 &= -\zeta_2[\lambda_1(\alpha_2 \alpha_7 - \alpha_3 \alpha_6) - \lambda_2(\alpha_1 \alpha_7 - \alpha_3 \alpha_5) + \lambda_3(\alpha_1 \alpha_6 - \alpha_2 \alpha_5)] \\
&\quad + \zeta_1(\alpha_2 \alpha_7 - \alpha_3 \alpha_6 + \alpha_1 \alpha_6 - \alpha_2 \alpha_5).
\end{aligned} \tag{53}$$

2.3. SOLUTIONS WHEN $\Delta_1 \geq 0, \Delta_2 < 0$ AND $\Delta_3 < 0$

Under this condition, all four roots from Equations (18) and (19) become complex variables. ψ_1 and ψ_2 have identical forms to equation (43). ψ_3 and ψ_4 are

$$\psi_3 = -\frac{\xi_2}{2} + i\sqrt{-\Delta_3}, \quad \psi_4 = -\frac{\xi_2}{2} - i\sqrt{-\Delta_3}. \tag{54}$$

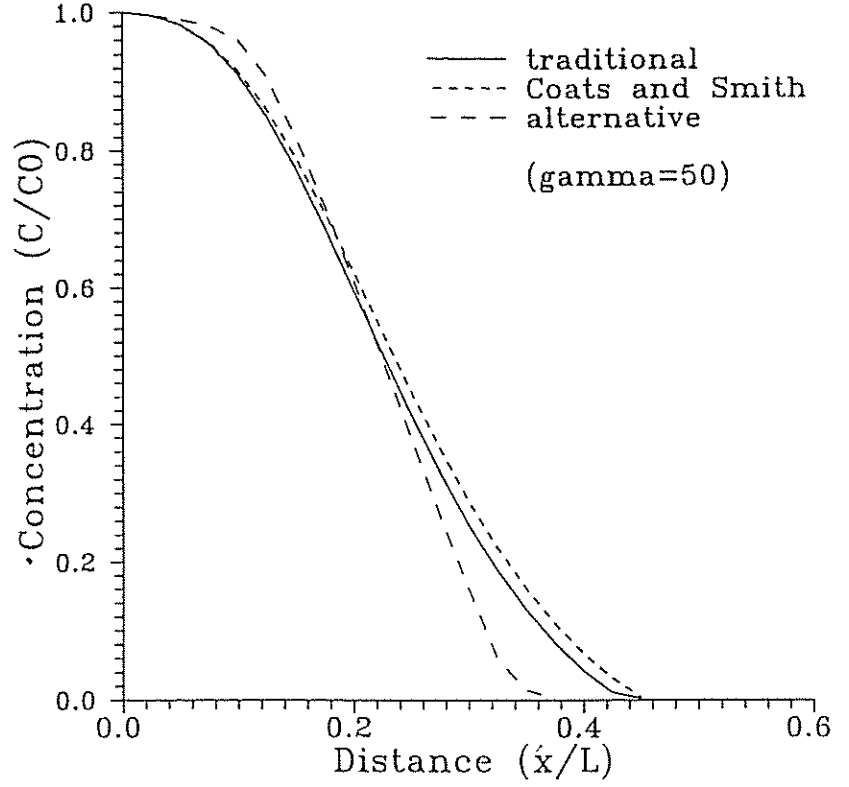


Fig. 4. Comparison of spatial concentration when $\gamma = 50$.

The concentration in micropores can be written as

$$\begin{aligned} \bar{c}_2 = & g_1 e^{w_1^* y} \cos(w_2^* y) + g_2 e^{w_1^* y} \sin(w_2^* y) \\ & + g_3 e^{w_3^* y} \cos(w_4^* y) + g_4 e^{w_3^* y} \sin(w_4^* y), \end{aligned} \quad (55)$$

where w_1^* and w_2^* are identical to w_1 and w_2 given in Equation (45), with w_3^* and w_4^* defined as

$$w_3^* = -\frac{\xi_2}{2}, \quad w_4^* = \sqrt{-\Delta_3}. \quad (56)$$

The concentration in macropores can be derived as

$$\begin{aligned} \bar{c}_1 = & \frac{1}{a\gamma} \{ g_1 e^{w_1^* y} [\lambda_1 \cos(w_2^* y) - \lambda_3 \sin(w_2^* y)] \\ & + g_2 e^{w_1^* y} [\lambda_1 \sin(w_2^* y) + \lambda_3 \cos(w_2^* y)] \} \end{aligned}$$

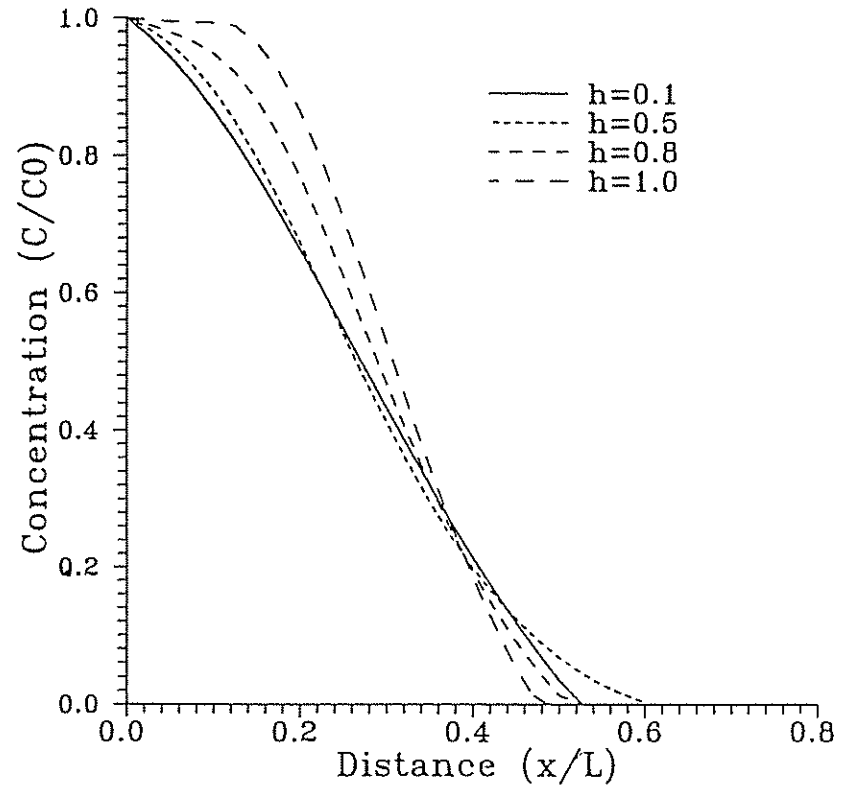


Fig. 5. Spatial concentration for various 'h'.

$$\begin{aligned}
 & + g_3 e^{w_3^* y} [\lambda_2 \cos(w_4^* y) - \lambda_4 \sin(w_4^* y)] \\
 & + g_4 e^{w_3^* y} [\lambda_2 \sin(w_4^* y) + \lambda_4 \cos(w_4^* y)] \}, \quad (57)
 \end{aligned}$$

where

$$\begin{aligned}
 \lambda_1 & = -(w_1^*)^2 + (w_2^*)^2 + w_1^* \frac{\gamma}{b} - \gamma \beta_1, \\
 \lambda_2 & = -(w_3^*)^2 + (w_4^*)^2 + w_3^* \frac{\gamma}{b} - \gamma \beta_1, \\
 \lambda_3 & = -w_2^* \left(2w_1^* - \frac{\gamma}{b} \right), \quad \lambda_4 = -w_4^* \left(2w_3^* - \frac{\gamma}{b} \right). \quad (58)
 \end{aligned}$$

After satisfying the boundary conditions, the system of equations for obtaining the constants $g_1, g_2, g_3,$ and g_4 can be constructed as follows:

$$\begin{bmatrix} 1 & 0 & 1 & 0 \\ \lambda_1 & \lambda_3 & \lambda_2 & \lambda_4 \\ \alpha_1 & \alpha_2 & \alpha_3 & \alpha_4 \\ \alpha_5 & \alpha_6 & \alpha_7 & \alpha_8 \end{bmatrix} \begin{bmatrix} g_1 \\ g_2 \\ g_3 \\ g_4 \end{bmatrix} = \begin{bmatrix} \zeta_2 \\ \zeta_1 \\ 0 \\ 0 \end{bmatrix} \quad (59)$$

where

$$\begin{aligned} \alpha_1 &= e^{w_1^*} [w_1^* \cos(w_2^*) - w_2^* \sin(w_2^*)], \\ \alpha_2 &= e^{w_1^*} [w_1^* \sin(w_2^*) + w_2^* \cos(w_2^*)], \\ \alpha_3 &= e^{w_3^*} [w_3^* \cos(w_4^*) - w_4^* \sin(w_4^*)], \\ \alpha_4 &= e^{w_3^*} [w_3^* \sin(w_4^*) + w_4^* \cos(w_4^*)], \\ \alpha_5 &= e^{w_1^*} [(\lambda_1 w_1^* - \lambda_3 w_2^*) \cos(w_2^*) - (\lambda_3 w_1^* + \lambda_1 w_2^*) \sin(w_2^*)], \\ \alpha_6 &= e^{w_1^*} [(\lambda_1 w_2^* + \lambda_3 w_1^*) \cos(w_2^*) + (\lambda_1 w_1^* - \lambda_3 w_2^*) \sin(w_2^*)], \\ \alpha_7 &= e^{w_3^*} [(\lambda_2 w_3^* - \lambda_4 w_4^*) \cos(w_4^*) - (\lambda_4 w_3^* + \lambda_2 w_4^*) \sin(w_4^*)], \\ \alpha_8 &= e^{w_3^*} [(\lambda_4 w_3^* + \lambda_2 w_4^*) \cos(w_4^*) + (\lambda_2 w_3^* + \lambda_4 w_4^*) \sin(w_4^*)]. \end{aligned} \quad (60)$$

Because the condition of $\Delta_3 < 0$ is unlikely encountered for parameters selected in the present study, detailed discussion of the solution of Equation (59) is omitted.

Solute concentrations c_1 and c_2 may be recovered in time by invoking a numerical inversion technique. In this work the Stehfest algorithm (Stehfest, 1970) is utilized.

3. Modeling of Miscible Flow

The model incorporating micropore diffusion and convection developed in this paper is compared to the analytical results obtained from a conventional dispersion-convection model (Bear, 1972), and to the model developed by Coats and Smith (1964). The sensitivity of the model is tested through parametric investigation. The model is also verified against experimental data. The relative concentration is represented for the macropore space only. Table I summarizes the selected modeling parameters for the designated figures.

The comparison of spatial concentration between the conventional single-porosity model (SP), the Coats and Smith's model (CS) and the present model (DP) for various equivalent Péclet numbers, γ , is shown in Figures 2–4. For the

and

$$\begin{aligned} \bar{c}_1 = & \frac{1}{a\gamma} \{g_1 e^{W_1 y} [\lambda_1 \cos(W_2 y) - \lambda_2 \sin(W_2 y)] \\ & + g_2 e^{W_3 y} [\lambda_3 \cos(W_4 y) + \lambda_4 \sin(W_4 y)] \\ & + g_3 e^{W_1 y} [\lambda_2 \cos(W_2 y) + \lambda_1 \sin(W_2 y)] \\ & + g_4 e^{W_3 y} [\lambda_4 \cos(W_4 y) - \lambda_3 \sin(W_4 y)]\} \end{aligned} \quad (69)$$

where

$$\begin{aligned} \lambda_1 = & -W_1^2 + W_1^2 + W_1 \frac{\gamma}{b} - \gamma\beta_1, & \lambda_2 = & -W_2 \left(2W_1 - \frac{\gamma}{b}\right), \\ \lambda_3 = & -W_4 \left(2W_3 - \frac{\gamma}{b}\right), & \lambda_4 = & -W_3^2 + W_4^2 + W_3 \frac{\gamma}{b} - \gamma\beta_1. \end{aligned} \quad (70)$$

Assuming

$$\begin{aligned} \alpha_1 = & e^{W_1} [W_1 \cos(W_2) - W_2 \sin(W_2)], \\ \alpha_2 = & e^{W_3} [W_3 \sin(W_4) + W_4 \cos(W_4)], \\ \alpha_3 = & e^{W_1} [W_1 \sin(W_2) + W_2 \cos(W_2)], \\ \alpha_4 = & e^{W_3} [W_3 \cos(W_4) - W_4 \sin(W_4)], \\ \alpha_5 = & e^{W_1} [(\lambda_1 W_1 - \lambda_2 W_2) \cos(W_2) - (\lambda_2 W_1 + \lambda_1 W_2) \sin(W_2)], \\ \alpha_6 = & e^{W_3} [(\lambda_3 W_3 + \lambda_4 W_4) \cos(W_4) + (\lambda_4 W_3 - \lambda_3 W_4) \sin(W_4)], \\ \alpha_7 = & e^{W_1} [(\lambda_2 W_1 + \lambda_1 W_2) \cos(W_2) + (\lambda_1 W_1 - \lambda_2 W_2) \sin(W_2)], \\ \alpha_8 = & e^{W_3} [(\lambda_4 W_3 - \lambda_3 W_4) \cos(W_4) - (\lambda_3 W_3 + \lambda_4 W_4) \sin(W_4)]. \end{aligned} \quad (71)$$

After satisfying the boundary conditions, the constants g_1, g_2, g_3 and g_4 in Equations (68) and (69) are derived as follows:

$$g_1 = \frac{H_1^*}{M^*}, \quad g_2 = \frac{H_2^*}{M^*}, \quad g_3 = \frac{H_3^*}{M^*}, \quad g_4 = \frac{H_4^*}{M^*}, \quad (72)$$

where

$$\begin{aligned} M^* = & \lambda_2 [\alpha_3 (\alpha_8 - \alpha_5) + \alpha_7 (\alpha_1 - \alpha_4)] - \lambda_3 [\alpha_2 (\alpha_8 - \alpha_5) + \alpha_6 (\alpha_1 - \alpha_4)] \\ & + (\lambda_4 - \lambda_1) (\alpha_2 \alpha_7 - \alpha_3 \alpha_6). \end{aligned} \quad (73)$$

$$H_1^* = \zeta_2 [\lambda_3 (\alpha_3 \alpha_8 - \alpha_4 \alpha_7) - \lambda_2 (\alpha_2 \alpha_8 - \alpha_4 \alpha_6)]$$

$$+ (\zeta_2 \lambda_4 - \zeta_1)(\alpha_2 \alpha_7 - \alpha_3 \alpha_6). \quad (74)$$

$$H_2^* = -\zeta_2[\lambda_1(\alpha_3 \alpha_8 - \alpha_4 \alpha_7) - \lambda_2(\alpha_1 \alpha_8 - \alpha_4 \alpha_5) + \lambda_4(\alpha_1 \alpha_7 - \alpha_3 \alpha_5)] \\ + \zeta_1(\alpha_3 \alpha_8 - \alpha_4 \alpha_7 + \alpha_1 \alpha_7 - \alpha_3 \alpha_5). \quad (75)$$

$$H_3^* = \zeta_2[\lambda_1(\alpha_2 \alpha_8 - \alpha_4 \alpha_6) - \lambda_3(\alpha_1 \alpha_8 - \alpha_4 \alpha_5) + \lambda_4(\alpha_1 \alpha_6 - \alpha_2 \alpha_5)] \\ - \zeta_1(\alpha_2 \alpha_8 - \alpha_4 \alpha_6 + \alpha_1 \alpha_6 - \alpha_2 \alpha_5). \quad (76)$$

$$H_4^* = -\zeta_2[\lambda_1(\alpha_2 \alpha_7 - \alpha_3 \alpha_6) - \lambda_3(\alpha_1 \alpha_7 - \alpha_3 \alpha_5) \\ + \lambda_2(\alpha_1 \alpha_6 - \alpha_2 \alpha_5)] + \zeta_1(\alpha_2 \alpha_7 - \alpha_3 \alpha_6). \quad (77)$$

Solute concentrations c_1 and c_2 in real time may be obtained by numerical inversion. For brevity, solutions when $W_2 < 0$ and/or $W_4 < 0$ are omitted.

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