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First-Order Mass Transfer in a Diffusion-Dominated (Immobile) Zone of an Axisymmetric Pore: Semi-Analytic Solution and Its Limitations

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대칭형 다공성 매질의 확산주도 영역에 관한 1차 물질이동 방정식

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Abstract Comparison of the classical mobile-immobile zone (MIM) model to the derived model led to several conclusions. If the MIM model is to be applied, the initial concentration in the immobile zone has to be down-scaled by a correction factor that is a function of pore geometry. The MIM model was valid only after sufficiently long time has passed, i.e., only after the diffusion front reaches the deepest pore wall in the immobile zone. The MIM mass-transfer coefficient α, was inversely proportional to the square of the pore depth. Also it did not depend on the mobile-zone flow velocity, contrary to the number of laboratory and field observations. The classical MIM model displayed a rapid exponential decay of immobile-zone concentration. Meanwhile at large times, the newly derived model displayed similar exponential decay. This was contrary to the mounting evidence of power-law BTC tails observed in laboratory and field settings

요 약 본 연구에서는 기존 MIM Zone model과 새로이 유도한 모델과 비교를 통해 몇가지 결론을 도출하였다. MIM model 이 적용되면 immobile 영역에서의 초기농도는 기공의 형태에 의해 달라지는 보정계수에 의해 실제농도보다 저평가되었으며 이는 오직 확산이 기공의 가장 깊은 부분까지 시행된 이후에 유효함을 확인하였다. 물질이동계수, α 는 기공의 깊이에 따라 반비례하며, 유동 구역의 유속에는 종속되어지지 않는다. 기존의 MIM model 은 확산주도영역의 농도가 급속하게 감소하는 현상을 보여주는데 새로이 유도된 모델의 경우도 충분한 시간이 경과한후 비슷한 현상을 보였으며 이는 기존의 여러 실험에서 관찰된 power-law BTC 의 상반되는 결과를 보여준다.

Key Words : Mobile-immobile zone model, Numerical Analysis, Diffusion, Mass-transfer

1. Introduction

Understanding mass transfer in porous media is crucial for protecting our groundwater resources and, in particular, for forecasting the fate of contaminants in subsurface and designing soil and aquifer remediation.

Since the existence of advection-dominated (mobile) and diffusion- dominated (immobile) zones in porous media and their roles in contaminant transport were recognized, a vast body of research has been built on

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these concepts. Introduced by Coats and Smith [1964] and van Genuchten and Wierenga [1976], the two-site mobile-immobile zone (MIM) model has found applications in a large number of column experiments and field tracer tests. Some of these refinements dealt with highly skewed breakthrough curves (BTCs) and/or BTCs with long and heavy tails that cannot be represented by the MIM model was soon observed and accounting for more than one mass transfer processes became necessary [Villeramaux, 1981, 1987, 1990; Brusseau *et al*,. 1989; Valocchi, 1990; Sardin *et al.*, 1991; Haggerty and Gorelick, 1995] developed a general multirate model that allows for arbitrary number of rates.

Many of the BTCs collected in laboratory and field experiments display a power-law behavior in the tail, i.e., concentration proportional to a power of time at large times. The power-law behavior in the tail can be captured by models with power-law memory functions or by the fractal mobile-immobile zone (MIM) model for solute transport developed by Schumer *et al*. [2003]. Interestingly, the same power-law BTC can be accurately represented by more than one probability density function (pdf) of the mass-transfer rate coefficient [Cunnigham *et al*., 1997; Haggerty and Gorelick, 1998]. Not surprisingly, different distributions of the mass-transfer coefficients can thus yield similar BTCs.

Haggerty and Gorelick [1995] demonstrated that the mobile/immobile and multirate mass transfer equations available in the literature are closely related to the basic mass transfer equation:

$$\frac{\partial C_{im}}{\partial t} = \alpha(C_m - C_{im}) \tag{1}$$

where C_{im}, C_m, and α are concentration in the immobile zone, concentration in the mobile zone, and a first-order rate coefficient, respectively. Coats and Smith [1964] and Goodknight *et al*. [1960], derived this equation by considering an immobile (stagnant) zone of volume V_{im} connected to the main channel of the mobile zone via a narrow neck of cross-sectional area A and length l. They assumed that i) the diffusion through the neck is in steady state and ii) the concentration in the immobile zone stays uniform. Conservation of mass and Fick's law with molecular diffusion D lead then directly to Eq.(1) with

$$\alpha = \frac{D\,A}{V_{im}\,l} \tag{2}$$

In the MIM model the above expression is modified to account for the relative zone fractions.

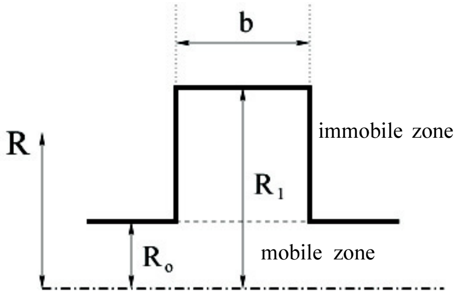
We note that the simplifying assumptions invoked by Coats and Smith [1964] and Goodknight *et al*. [1960] are severe. Indeed, the first one does not hold when a concentration in the mobile zone varies rapidly enough, as it may when a concentration front is passing, while the second one is even more unrealistic, as there is no plausible physical mechanism for mixing the immobile zone.

In order to properly estimate the time needed for aquifer remediation, it is crucial that models of mass transfer in porous media accurately resolve the tail (long-time) concentrations. Although BTCs with power-law tails are often referred to as "anomalous" Haggerty *et al*., 2001; Schumer *et al*., 2003; and others], the question arises whether such BTCs are exceptional or typical. A light could be shed on this question by carefully studying the derivation and limitations of the basic mass-transfer equation, Eq.(1), that lies at the foundation of the discussed models. Kim *et al.*, [2010] have done it for a two-dimensional rectangular pore without invoking the oversimplifying assumptions.

In this paper we derive and study the limitations of the mass-transfer equation for an immobile zone of an axisymmetric pore.

2. Derivation of the Mass-Transfer Equation for an Immobile Zone of an Axisymmetric Pore

Consider an axisymmetric pore of depth $a = R_1 - R_0$ presented in Figure 1. Since we focus here only on the immobile zone, we drop the subscript and from now on denote the concentration in that zone by C. We assume no-flux boundary conditions on the walls and a C_m concentration on the boundary between the zones. With no loss of generality we assume $C_m = 0$. We also assume uniform initial concentration of C_0.



[Fig. 1] Schematic of an axisymmetric pore. b is the pore width and R stands for radius.

Although in reality the immobile zone is not stagnant and a vortex could form there driven by the flow in the mobile zone, we assume that the immoblie zone is perfectly still. This assumption represents an idealization, unless an infinitesimally thin permeable membrane is placed between the zones. Due to symmetry the problem reduces to

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r}\right) \tag{3}$$

subject to

$$C|_{t=0} = C_0 \tag{4}$$

$$\frac{\partial C}{\partial r}|_{r=R_1} = 0 \tag{5}$$

$$C|_{r=R_0} = 0 \tag{6}$$

where r and t are space (radial) and time coordinates.

Let $\tilde{C}(r,p) = \mathcal{L}\{C(r,t), t \to p\}$ be the Laplace transform with respect to time of $C(r,t)$. Applying the transform, $\mathcal{L}\{(\ , \ ), t \to p\}$, to Eqs.(3), (5), and (6), employing Eq.(4) leads to

$$D\left[\frac{d^2\tilde{C}}{dr^2} + \frac{1}{r}\frac{d\tilde{C}}{dr}\right] - p\tilde{C} = -C_0 \tag{7}$$

subject to

$$\frac{\partial \tilde{C}}{\partial r}|_{r=R_1} = 0 \tag{8}$$

$$\tilde{C}|_{r=R_0} = 0 \tag{9}$$

with

$$q = \sqrt{\frac{p}{D}} \tag{10}$$

the general solution of Eq.(7) and its derivative with respect to r are given by

$$\tilde{C}(r,p) = \frac{C_0}{p} + A I_0(qr) + BK_0(qr) \tag{11}$$

and

$$\frac{\partial \tilde{C}}{\partial r} = q\left[A I_1(qr) - BK_1(qr)\right] \tag{12}$$

where I_n and K_n are n-th order modified Bessel functions of the first and second kind, respectively, and A and B are arbitrary constants.

Now, Eqs.(8) and (12) lead to

$$I_1(qR_1)A - K_1(qR_1)B = 0 \tag{13}$$

while Eqs.(9) and (10) yield

$$I_0(qR_0)A + K_0(qR_0)B = -\frac{C_0}{p} \tag{14}$$

Solving the system Eqs.(13), (14) for A and B and plugging them back into Eq.(11) yields the semi-analytic solution of the problem

$$\tilde{C}(r,p) = \frac{C_0}{p}\left[1 - \frac{K_1(qR_1)\,I_0(qr) + I_1(qR_1)\,K_0(qr)}{I_0(qR_0)\,K_1(qR_1) + I_1(qR_1)\,K_0(qR_0)}\right] \tag{15}$$

The average concentration in the pore is

$$\begin{aligned}\bar{C}(t) &= \frac{1}{V_{pore}}\int_{V_{pore}} C\,dV \\ &= \frac{1}{\pi(R_1^2 - R_0^2)b}\int_{R_0}^{R_1} C(r,t)(2\pi r b\,dr)\end{aligned} \tag{16}$$

while its Laplace transform is

$$\tilde{\bar{C}}(p) = \frac{2}{R_1^2 - R_0^2}\int_{R_0}^{R_1} \tilde{C}(r,p)\,r\,dr \tag{17}$$

Now, Eqs.(15) and (17) yield

$$\widetilde{\overline{C}}(p) = \frac{2\,C_0}{(R_1^2 - R_0^2)\,p}\left[\frac{R_1^2 - R_0^2}{2} - \right.$$
$$\left.\left[\int_{R_0}^{R_1} \frac{K_1(q R_1)\, I_0(q r) + I_1(q R_1)\, K_0(q R_1)\, K_0(q r)}{K_1(q R_1)\, I_0(q R_0) + I_1(q R_1)\, K_0(q R_0)} r\, dr\right]\right] \quad (18)$$

The integrals of the Bessel functions in Eq.(18) follow from *Abramowitz and Stegun* [1972, (6.6.28)]} as $\int I_0(z) z\, dz = I_1(z)\, z$ and $\int K_0(z) z\, dz = -K_1(z)\, z$. With them Eq.(18) reduces to

$$\widetilde{\overline{C}}(p) = \frac{C_0}{p}\left[\frac{2\,R_0}{(R_1^2 - R_0^2)\,q}\,\frac{I_1(q R_1) K_1(q R_0) - I_1(q R_0) K_1(q R_1)}{I_0(q R_0) K_1(q R_1) + I_1(q R_1) K_0(q R_0)}\right] \quad (19)$$

or with Eq.(10)

$$\widetilde{\overline{C}}(p) = \frac{C_0}{q^2 D}\left[1 - \frac{2\,R_0}{(R_1^2 - R_0^2)}\,\frac{1}{q}\,\frac{I_1(q R_1) K_1(q R_0) - I_1(q R_0) K_1(q R_1)}{I_0(q R_0) K_1(q R_1) - I_1(q R_0) K_0(q R_1)}\right] \quad (20)$$

Numerical Laplace-transform inversion suggests that for large times $\overline{C}(t)$ decays exponentially. We'll match it therefore with

$$C_{im}(t) = \beta\, e^{\alpha\, t} \Rightarrow C_{im}(p) = \frac{\beta}{p + \alpha} \quad (21)$$

$$\{t \gg 0 \Leftrightarrow |p| \ll 1\} \Rightarrow$$
$$\overline{C_{im}}(p) = \frac{\beta}{\alpha}\,\frac{1}{1 + p/\alpha} = \frac{\beta}{\alpha}\left[1 - \frac{p}{\alpha} + \frac{p^2}{\alpha^2} - \cdots\right] \quad (22)$$

Switching to the q variable via Eq.(10), we have

$$\left|\frac{q^2\, 2D}{\alpha}\right| \ll 1 \Rightarrow \overline{C_{im}}(q) = \frac{\beta}{\alpha}\left[1 - \frac{q^2\, 2D}{\alpha} + \cdots\right] \quad (23)$$

It follows from Eq.(23) that

$$\lim_{q \to 0} \widetilde{\overline{C_{im}}}(q) = \frac{\beta}{\alpha} \Rightarrow \beta = \alpha \lim_{q \to 0} \widetilde{\overline{C_{im}}}(q) \quad (24)$$

and

$$\lim_{q \to 0}\left(\frac{1}{q}\,\frac{d\widetilde{\overline{C_{im}}}}{dq}\right) = \frac{2\,\beta\, D}{\alpha^2} \quad (25)$$

Thus

$$\alpha = -2\,D \lim_{q \to 0} \widetilde{\overline{C_{im}}}(q) / \lim_{q \to 0}\left(\frac{1}{q}\,\frac{d\widetilde{\overline{C}}_{im}}{dq}\right) \quad (26)$$

$$\beta = -2\,D\left(\lim_{q \to 0} \widetilde{\overline{C_{im}}}(q)\right)^2 / \lim_{q \to 0}\left(\frac{1}{q}\,\frac{d\widetilde{\overline{C}}_{im}}{dq}\right) \quad (27)$$

For Eqs.(23) and (20) to match for long times we can calculate α and β from Eqs.(26) and (27) with (20) used for $\widetilde{\overline{C_{im}}}(q)$. The results are

$$\alpha = \frac{12\,D}{R_0^2}\,\frac{y^2\{(1 - y^2)(y^2 - 3) - 4\ln y\}}{17 - 30 y^2 + 15 y^4 - 2 y^6 + 12 \ln y\,(3 - 2 y^2 + 2 \ln y)} \quad (28)$$

$$\beta = C_0\,\frac{12\, y^2\{(1 - y^2)(y^2 - 3) - 4\ln y\}^2}{(1 - y^2)\{17 - 30 y^2 + 15 y^4 - 2 y^6 + 12 \ln y\,(3 - 2 y^2 + 2 \ln y)\}} \quad (29)$$

where

$$y = \frac{R_0}{R_1} \quad (30)$$

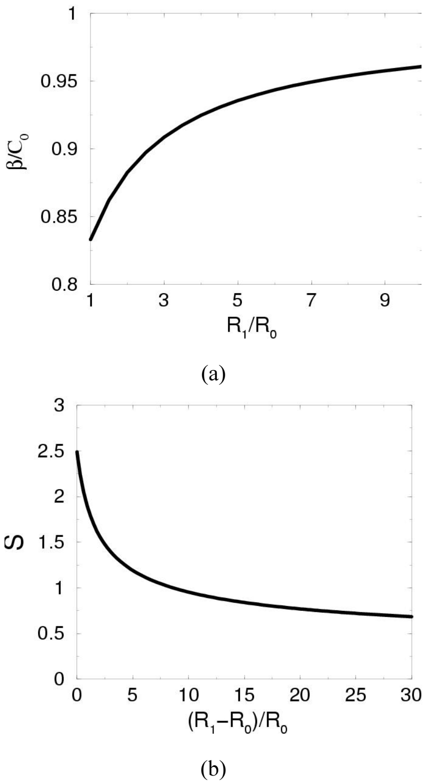
3. Results

We note that β / C_0 represents a correction factor to the initial condition required for the MIM model to match the exact semi-analytic solution in late times. In Figure 2a we plot this correction factor as a function of R_1 / R_0. In a parallel study of a 2-D pore we found the corresponding correction factor to be $\beta / C_0 = 5/6$ [Kim *et al*, 2010]. In that study we also found the mass-transfer coefficient for a 2-D pore to be inversly proportional to the pore depth, $\alpha = 2.5\, D / a^2$. The mass-transfer coefficient for an axisymmetric pore, Eq.(28), can be recast in an analogous form

$$\alpha = S \frac{D}{(R_1 - R_0)^2} \quad (31)$$

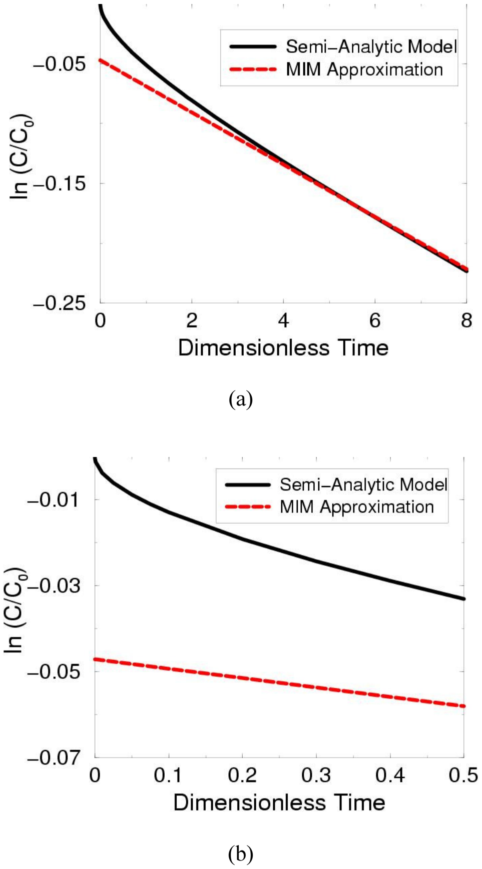
with the shape factor S being a function of the dimensionless pore depth $(R_1 - R_0)/R_0$. We plot this shape factor in Figure 2b.

The semi-analytic solution Eq.(20) is inverted to the time domain via the Stehfest algorithm [Stehfest, 1970], and the results are presented in Figure 3, where we plot the dimensionless concentration, C / C_0 versus dimensionless time, $\tau = \alpha t$ for $R_1 / R_0 = 8$. As in a 2-D pore, the traditional mobile-immobile zone (MIM) model can match the exact semi-analytic solution only if the initial condition is scaled down by the correction factor plotted in Figure 2a, as seen from comparing Figure 3a (new semi-analytic solution) to Figure 3b (the MIM model). Although the new semi-analytic solution differs from the MIM model for sufficiently early times, as seen from Figure 3b, it quickly settles in an exponential decay and matches the MIM model, as seen from Figure 3a.



[Fig. 2] (a) the correction factor β/C_0 as a function of R_1/R_0, (b) the shape factor S as a function of dimensionless pore depth $(R_1-R_0)/R_0$ of an axisymmetric pore.

It is clear from Figure 3 that the diffusion from the immobile zone can be described by the classical MIM model only for times sufficiently long for the diffusion front to reach the deepest wall in the pore. It is quite remarkable that the MIM model, defined by a first-order differential equation with only a time derivative, Eq.(1), matches for most of the times the appropriately averaged exact solution of the governing parabolic differential equation, Eq.(3), involving not only a time derivative but also a second spatial derivative.



[Fig. 3] The mobile-immobile zone (MIM) model versus the newly derived semi-analytic model: (a) long times, (b) early times.

4. Conclusion

A new semi-analytic solution, Eq.(19), has been derived for the diffusion into or from an immobile zone of a axisymmetric pore. Comparison of the classical mobile-immobile zone (MIM) model to the derived model leads to the following:

- If the MIM model is to be applied, the initial concentration in the immobile zone has to be down-scaled by a correction factor that is a function of pore geometry.
- The MIM model is valid only after sufficiently

long time has passed, i.e., only after the diffusion front reaches the deepest pore wall in the immobile zone.

- The MIM model should fail for sufficiently rapid concentration fluctuations at time scales below the time the diffusion front reaches the deepest pore wall.
- The MIM mass-transfer coefficient α, given by Eq.(28), is inversely proportional to the square of the pore depth.
- As long as the mobile-immobile zone mass transfer timescale is appreciably longer than the advection timescale, the mass-transfer coefficient α in the MIM model does not depend on the mobile-zone flow velocity. This is contrary to the number of laboratory and field observations [*Brusseau*, 1992; *Bajracharya and Barry*, 1997; and others]}.
- The classical MIM model displays a rapid exponential decay of immobile-zone concentration. At large times, the newly derived model displays similar exponential decay. This is contrary to the mounting evidence of power-law BTC tails observed in laboratory and field settings [*Farrell and Reinhard*, 1994; *Werth et al.*, 1997; *Meigs and Beauheim*, 2001; and others].
- As has been pointed out in the literature for over half a century [*Gluecakauf*, 1955; *Rao et al.*, 1980ab; *Haggerty et al.*, 2004; and others], the MIM model does not fully represent the physics of mass transfer. And neither does the newly derived model.

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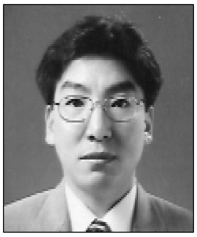

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