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Macroscopic modelling of solute transport in porous media saturated by two immiscible liquids

B.Kh. Khuzhayorov

Complex research institute of regional problems, Uzbekistan Academy of Sciences, Samarkand City, Uzbekistan

J.-L. Auriault & P. Royer

Laboratoire 3S CNRS, INPG, UJF, Domaine Universitaire, Grenoble Cedex 9, France

ABSTRACT: We consider solute transport in a porous medium saturated by two immiscible liquids with different viscosities. Liquid with high viscosity can form isolated stagnant zones (immobile liquid zone – IMLZ), whereas liquid with low viscosity forms a mobile liquid zone – MLZ. Diffusion properties of MLZ and IMLZ can considerably differ. The homogenization method is used to obtain a macroscopic description of the problem. Several cases are defined by means of the order of magnitude of local non-dimensional Dämkohler number, of the Péclet number and of the ratio of diffusion coefficients of liquids. For each case macroscopic solute transport models are obtained and analyzed. It is shown that the influence of the IMLZ appears by means of a source/sink term in the macroscopic equation. This indicates that solute mass-exchange between the MLZ and IMLZ can considerably change the solute concentration distribution in the MLZ.

1 INTRODUCTION

During the displacement of heavy oil by water solutions of chemicals, isolated stagnant oil zones surrounded by water can be formed due to the great differences in viscosities of oil and water. Thus, the water phase forms a mobile liquid zone (MLZ) whereas oil occupies an immobile liquid zone (IMLZ). The solute can diffuse through the MLZ/IMLZ interface, and due to high differences in viscosities of both liquids, the diffusion coefficients in the MLZ and in the IMLZ can be significantly different. The presence of IMLZ in porous media yields to significant changes of hydrodynamic parameters of solute transport, and, in particular, to anomalous breakthrough curves (BTCs). One of these anomalous effects is tail effect.

One of the first models accounting for the presence of IMLZ was introduced by Coats & Smith (1964), where the IMLZ is presented as a dead-end pore one, which yields a capacitance model. The dispersion equations of Coats and Smith (1964) can be written in the form

$$\frac{\partial}{\partial t}(\theta_m c_m) + \frac{\partial}{\partial t}(\theta_{im} c_{im}) = \nabla \cdot (\theta_m \mathbf{D}_m \cdot \nabla c_m) - \nabla \cdot (\theta_m v \nabla c_m), \quad (1)$$

$$\frac{\partial}{\partial t}(\theta_{im} c_{im}) = \alpha_{im}(c_m - c_{im}), \quad (2)$$

where c_m, c_{im} are the mass concentrations of solute, \mathbf{D}_m is the dispersion coefficient, \mathbf{v} is the average velocity in the mobile liquid, θ_m, θ_{im} are the fractions of the pore volume occupied by mobile and immobile liquids, respectively, α_{im} is a mass transfer coefficient, and indexes «m» and «im» correspond to mobile and immobile liquids, respectively. Note that Eqs (1), (2) were written by Coats & Smith (1964) in the one-dimensional case, with $D = \text{const}$, $\theta_m = \text{const}$ and with the coefficient θ_m absent in the RHS of (1) (it was introduced later by subsequent authors).

Van Genuchten & Wierenga (1976) proposed a model generalizing the Coats & Smith (1964) model. They introduced two regions, the dynamic and stagnant zones, with fraction f of the sorption sites in the MLZ, and fraction $1 - f$ in the IMLZ. For a nonlinear instantaneous adsorption law $s = a_1 c^{b_1}$, $a_1, b_1 = \text{const}$, and when $D_m, \theta_m, \theta_{im}, f, v$ are constants, resulting equations in one-dimensional case are in the form

$$\theta_m R_m \frac{\partial c_m}{\partial t} + \theta_{im} R_{im} \frac{\partial c_{im}}{\partial t} = \theta_m D_m \frac{\partial^2 c_m}{\partial x^2} - v \theta_m \frac{\partial c_m}{\partial x}, \quad (3)$$

$$\theta_{im} R_{im} \frac{\partial c_{im}}{\partial t} = \alpha_{im}(c_m - c_{im}), \quad (4)$$

where $R_m = 1 + f \rho a_1 b_1 c_m^{b_1-1} / \theta_m$, $R_{im} = 1 + (1 - f) \times \rho a_1 b_1 c_{im}^{b_1-1} / \theta_{im}$, and ρ is the bulk density of the porous medium.

There are many works where the tail effect of BTCs is well described by Eqs (1), (2) or (3), (4) (Coats & Smith 1964, van Genuchten & Wierenga 1976, Gaudet et al. 1977, de Smedt & Wierenga 1979 etc.).

Piquemal (1992) analyzed the miscible fluid displacement in porous media with a mobile and immobile fraction by means of volume or surface averaging methods. The results show strong limitations: Péclet numbers should be large enough, interfacial area between mobile liquid and solid should be much larger than that between mobile and immobile liquids. On the other hand, during the application of the averaging technique the author used a phenomenological relation.

Adsorption of solute in porous media was investigated by Auriault & Lewandowska (1993a, b, 1995, 1996, 1997), Canon et al. (1999) by using the method of homogenization of periodic structures.

To our knowledge, macroscopic modelling of solute transport in porous media with MLZ and IMLZ in presence of adsorption, has not been still investigated. We address this problem in the present paper. During the following investigations we repeatedly make use of the results and the methodology developed by Auriault & Lewandowska (1993a, b, 1995, 1996, 1997), as well as by Auriault (1983, 1991), Auriault & Adler (1995), Auriault & Royer (1993).

2 STATEMENT OF THE PROBLEM

Consider a porous medium which domain Ω consists of three parts: Ω_1 is the mobile liquid, where convection-diffusion phenomena occur; Ω_2 is the immobile liquid, where only diffusion can occur, and Ω_3 is the solid domain. Domains Ω_β , $\beta = 1, 2, 3$ are assumed to be connected and Ω_3 is undeformable (Fig. 1).

We denote by ϕ_1 the volume fraction of Ω_1 , i.e. $\phi_1 = \frac{|\Omega_1|}{|\Omega|}$, and by analogy $\phi_2 = \frac{|\Omega_2|}{|\Omega|}$. Obviously,



Figure 1. A porous medium with MLZ and IMLZ. pore volume occupied by mobile liquid (Ω_1), volume occupied by immobile liquid (Ω_2), solid volume (Ω_3).

$\phi = \phi_1 + \phi_2$ represents the total porosity of the porous medium.

Common boundaries between domains Ω_β , $\beta = 1, 2, 3$ are denoted $\Gamma_1 = G_1 \cap G_3 \cap G$, $\Gamma_2 = G_1 \cap G_2 \cap G$, $\Gamma_3 = G_2 \cap G_3 \cap G$. The domain Ω is assumed to be periodic, with a period of characteristic length l . Let L be a macroscopic characteristic length. Then we can introduce the small parameter of separation of scales $\varepsilon = l/L$.

The fluids are incompressible and liquid motion in Ω_1 is quasi-static, so we neglect the initial and transient terms in the Navier-Stokes equation. Then at the pore scale the fluid motion in Ω_1 can be described by Stokes' equation and the local equation of continuity

$$\mu \nabla^2 \mathbf{v} - \nabla p = 0, \quad \nabla \cdot \mathbf{v} = 0, \quad (5)$$

with the no-slip condition on the solid walls and on the surface of the IMLZ Ω_2

$$\mathbf{v} = 0 \quad \text{on } \Gamma_1 \cup \Gamma_2. \quad (6)$$

We consider a problem of solute transport in Ω_1 with possible adsorption on Γ_1 and mass-exchange between Ω_1 and Ω_2 domains through Γ_2 . In Ω_2 only diffusion of the solute can occur whereas in Ω_1 solute is transported by both convection and diffusion. So the concentration of the solute c_1 in Ω_1 obeys the convection-diffusion equation

$$\frac{\partial c_1}{\partial t} + \nabla \cdot (\mathbf{v}c_1) = \nabla \cdot (\mathbf{D}_1 \cdot \nabla c_1) \quad (7)$$

and the solute concentration c_2 in Ω_2 is described by the diffusion equation

$$\frac{\partial c_2}{\partial t} = \nabla \cdot (\mathbf{D}_2 \cdot \nabla c_2), \quad (8)$$

where $\mathbf{D}_1, \mathbf{D}_2$ are the molecular diffusion tensors in Ω_1 and Ω_2 , respectively.

According to the above description, the boundary conditions are in the form:

$$-\mathbf{n}_1 \cdot (\mathbf{D}_1 \cdot \nabla c_1) = \alpha \frac{\partial c_1}{\partial t} \quad \text{on } \Gamma_1, \quad (9)$$

$$\mathbf{n}_1 \cdot (\mathbf{D}_1 \cdot \nabla c_1) = -\mathbf{n}_2 \cdot (\mathbf{D}_2 \cdot \nabla c_2) \quad \text{on } \Gamma_2, \quad (10)$$

$$c_1 = c_2 \quad \text{on } \Gamma_2, \quad (11)$$

$$-\mathbf{n}_2 \cdot (\mathbf{D}_2 \cdot \nabla c_2) = 0 \quad \text{on } \Gamma_3, \quad (12)$$

where α is an adsorption parameter, $\mathbf{n}_1, \mathbf{n}_2$ are unit vectors normal to Γ_1 and Γ_2 and oriented from Ω_1 to Ω_2 and Ω_3 and from Ω_2 to Ω_1 and Ω_3 , respectively. Obviously, we have $\mathbf{n}_1 = -\mathbf{n}_2$ on Γ_2 .

3 NORMALIZATION

We use the macroscopic point of view (Auriault 1991). Therefore, the characteristic length in use for normalization is the macroscopic characteristic length L , e.g., the characteristic length of the porous media sample. We introduce the following non-dimensional variables

$$\begin{aligned} \bar{c}_1 &= \frac{c_1}{c_0}, \quad \bar{c}_2 = \frac{c_2}{c_0}, \quad \bar{x} = \frac{x}{L}, \quad \bar{v} = \frac{v}{U}, \\ \bar{\mathbf{D}}_1 &= \frac{\mathbf{D}_1}{D_1}, \quad \bar{\mathbf{D}}_2 = \frac{\mathbf{D}_2}{D_2}, \quad \bar{t} = \frac{t}{t_c}, \end{aligned} \quad (13)$$

where c_0, D_1, D_2, U, t_c are characteristic values of $c_1(c_2), \mathbf{D}_1, \mathbf{D}_2, v, t$, respectively, and the overscript « $\bar{\cdot}$ » shows non-dimensional variables. Then we can write the set (7)–(12) in the following dimensionless form after choosing $t_c = \frac{L^2}{D_1}$, which is the characteristic diffusion time in the MLZ

$$\frac{\partial \bar{c}_1}{\partial t} + \text{Pe} \cdot \nabla \cdot (\bar{\mathbf{v}} \bar{c}_1) = \nabla \cdot (\bar{\mathbf{D}}_1 \cdot \nabla \bar{c}_1), \quad (7')$$

$$\frac{\partial \bar{c}_2}{\partial t} = D \cdot \nabla \cdot (\bar{\mathbf{D}}_2 \cdot \nabla \bar{c}_2), \quad (8')$$

$$-\mathbf{n}_1 \cdot (\bar{\mathbf{D}}_1 \cdot \nabla \bar{c}_1) = \text{Da} \cdot \frac{\partial \bar{c}_1}{\partial t} \quad \text{on } \Gamma_1 \quad (9')$$

$$\mathbf{n}_1 \cdot (\bar{\mathbf{D}}_1 \cdot \nabla \bar{c}_1) = -D \mathbf{n}_2 \cdot (\bar{\mathbf{D}}_2 \cdot \nabla \bar{c}_2) \quad \text{on } \Gamma_2 \quad (10')$$

$$\bar{c}_1 = \bar{c}_2 \quad \text{on } \Gamma_2 \quad (11')$$

$$-\mathbf{n}_2 \cdot (\bar{\mathbf{D}}_2 \cdot \nabla \bar{c}_2) = 0 \quad \text{on } \Gamma_3, \quad (12')$$

where $\text{Pe} = \frac{UL}{D_1}$ is the Péclet number, $\text{Da} = \frac{\sigma}{L}$ is the Dämcöhler number, and $D = \frac{D_2}{D_1}$. For the sake of the simplicity we keep the same notations, but all quantities are now dimensionless quantities.

4 HOMOGENIZATION

Since the macroscopic point of view (Auriault 1991) was adopted, we use the following differential rule

$$\nabla \cdot = \nabla_x \cdot + \frac{1}{\varepsilon} \nabla_y \cdot \quad (14)$$

For large Péclet numbers, we can introduce a second characteristic time $t_c = \frac{L^2}{D_1}$ and the corresponding dimensionless time τ . When using these two time scales, the differentiation rule with respect to time becomes

$$\frac{\partial}{\partial t} \Rightarrow \frac{\partial}{\partial t} + \text{Pe} \cdot \frac{\partial}{\partial \tau} \quad (15)$$

We also introduce the following notations for the volume and surface averages

$$\begin{aligned} \langle \cdot \rangle_{\Omega_1} &= \frac{1}{|\Omega_1|} \int_{\Omega_1} \cdot d\Omega, \quad \langle \cdot \rangle_{\Omega_2} = \frac{1}{|\Omega_2|} \int_{\Omega_2} \cdot d\Omega, \\ \langle \cdot \rangle_1 &= \frac{1}{|\Omega|} \int_{\Omega_1} \cdot d\Omega, \quad \langle \cdot \rangle_2 = \frac{1}{|\Omega|} \int_{\Omega_2} \cdot d\Omega, \quad (16) \\ \langle \cdot \rangle_{\Gamma_1} &= \frac{1}{|\Omega|} \int_{\Gamma_1} \cdot dS, \quad \langle \cdot \rangle_{\Gamma_2} = \frac{1}{|\Omega|} \int_{\Gamma_2} \cdot dS. \end{aligned}$$

Obviously, we have $\langle \cdot \rangle_\beta = \phi_\beta \langle \cdot \rangle_{\Omega_\beta}$, $\beta = 1, 2$. And we introduce the following specific surfaces of the porous medium

$$\begin{aligned} \gamma_1 &= \frac{|\Gamma_1|}{|\Omega|}, \quad \gamma_2 = \frac{|\Gamma_2|}{|\Omega|}, \\ \gamma_{12} &= \gamma_1 + \gamma_2 = \frac{|\Gamma_1| + |\Gamma_2|}{|\Omega|}. \end{aligned} \quad (17)$$

Then, the total specific surface of the porous medium is defined by

$$\gamma_{p.m.} = \frac{|\Gamma_1| + |\Gamma_3|}{|\Omega|}.$$

Note that in general we have $\gamma_{12} \neq \gamma_{p.m.}$

Problem (5)–(6) is independent of problem (7)–(12). It is a well-known problem, see for example Auriault & Adler (1995). The main result is in the form

$$\langle \mathbf{v}_0 \rangle_1 = -\mathbf{K}_f \cdot \nabla_x p^0, \quad (18)$$

where $\mathbf{K}_f = (\mathbf{k}_f)_1$, \mathbf{k}_f is solution to the corresponding cell problem.

We analyze now different cases defined by the orders of magnitude of the non-dimensional numbers Pe , Da , and D . As in Auriault & Adler (1995), Auriault & Lewandowska (1993b, 1996) we consider the values $\text{Pe} = O(\varepsilon), O(1), O(\varepsilon^{-1})$. In the present paper, we restrict ourselves to small $D = O(\varepsilon)$ and small or moderate adsorption, i.e. $\text{Da} = O(\varepsilon), O(1)$. Cases with $\text{Da} = O(\varepsilon^q)$, $q \geq 2$ lead to macroscopic equations where adsorption does not appear.

Case 1. $\text{Pe} = O(\varepsilon)$, $\text{Da} = O(\varepsilon)$, $D = O(\varepsilon)$. The non-dimensional equations and the corresponding boundary conditions are

$$\frac{\partial c_1}{\partial t} + \varepsilon \nabla \cdot (\mathbf{v} c_1) = \nabla \cdot (\mathbf{D}_1 \cdot \nabla c_1) \quad \text{in } \Omega_1, \quad (19)$$

$$\frac{\partial c_2}{\partial t} = \varepsilon \nabla \cdot (\mathbf{D}_2 \cdot \nabla c_2) \quad \text{in } \Omega_2, \quad (20)$$

$$-\mathbf{n}_1 \cdot (\mathbf{D}_1 \cdot \nabla c_1) = \varepsilon \alpha \frac{\partial c_1}{\partial t} \quad \text{on } \Gamma_1, \quad (21)$$

$$\mathbf{n}_1 \cdot (\mathbf{D}_1 \cdot \nabla c_1) = -\varepsilon \mathbf{n}_2 \cdot (\mathbf{D}_2 \cdot \nabla c_2) \quad \text{on } \Gamma_2, \quad (22)$$

$$c_1 = c_2 \quad \text{on } \Gamma_2, \quad (23)$$

$$-\mathbf{n}_2 \cdot (\mathbf{D}_2 \cdot \nabla c_2) = 0 \quad \text{on } \Gamma_3. \quad (24)$$

We look for the unknowns in the form of asymptotic expansions

$$\begin{aligned} c_1 &= c_1^0(x, y, t) + \varepsilon c_1^1(x, y, t) + \varepsilon^2 c_1^2(x, y, t) + \dots, \\ c_2 &= c_2^0(x, y, t) + \varepsilon c_2^1(x, y, t) \\ &\quad + \varepsilon^2 c_2^2(x, y, t) + \dots, \\ \mathbf{v} &= \mathbf{v}^0(x, y, t) + \varepsilon \mathbf{v}^1(x, y, t) + \varepsilon^2 \mathbf{v}^2(x, y, t) + \dots, \end{aligned} \quad (25)$$

Introducing these expansions in Eq. (19) gives at the successive orders of ε

$$\varepsilon^{-2} : \nabla_y \cdot (\mathbf{D}_1 \cdot \nabla_y c_1^0) = 0, \quad (26)$$

$$\begin{aligned} \varepsilon^{-1} : \nabla_y \cdot (\mathbf{D}_1 \cdot (\nabla_x c_1^0 + \nabla_y c_1^1)) \\ + \nabla_x \cdot (\mathbf{D}_1 \cdot \nabla_y c_1^0) = 0, \end{aligned} \quad (27)$$

$$\begin{aligned} \varepsilon^0 : \frac{\partial c_1^0}{\partial t} + \nabla_y \cdot (\mathbf{v}^0 c_1^0) = \nabla_x \cdot (\mathbf{D}_1 \cdot (\nabla_x c_1^0 + \nabla_y c_1^1)) \\ + \nabla_y \cdot (\mathbf{D}_1 \cdot (\nabla_x c_1^1 + \nabla_y c_1^2)). \end{aligned} \quad (28)$$

In the same way, we have with Eq. (20)

$$\varepsilon^{-1} : \nabla_y \cdot (\mathbf{D}_2 \cdot \nabla_y c_2^0) = 0, \quad (29)$$

$$\begin{aligned} \varepsilon^0 : \frac{\partial c_2^0}{\partial t} = \nabla_x \cdot (\mathbf{D}_2 \cdot \nabla_y c_2^0) + \nabla_y \\ \times (\mathbf{D}_2 \cdot (\nabla_x c_2^0 + \nabla_y c_2^1)). \end{aligned} \quad (30)$$

From the boundary conditions (21)–(24) we have at successive orders of ε on Γ_1 :

$$-\mathbf{n}_1 \cdot (\mathbf{D}_1 \cdot \nabla_y c_1^0) = 0, \quad (31)$$

$$-\mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c_1^0 + \nabla_y c_1^1) = 0, \quad (32)$$

$$-\mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c_1^1 + \nabla_y c_1^2) = \alpha \frac{\partial c_1^0}{\partial t}, \quad (33)$$

on Γ_2 :

$$\mathbf{n}_1 \cdot \mathbf{D}_1 \cdot \nabla_y c_1^0 = 0, \quad (34)$$

$$\mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c_1^0 + \nabla_y c_1^1) = -\mathbf{n}_2 \cdot \mathbf{D}_2 \cdot \nabla_y c_2^0, \quad (35)$$

$$\begin{aligned} \mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c_1^1 + \nabla_y c_1^2) = -\mathbf{n}_2 \cdot \mathbf{D}_2 \\ \times (\nabla_x c_2^0 + \nabla_y c_2^1), \end{aligned} \quad (36)$$

$$c_1^0 = c_2^0, \quad c_1^1 = c_2^1, \dots, \quad (37)$$

and on Γ_3 :

$$-\mathbf{n}_2 \cdot (\mathbf{D}_2 \cdot \nabla_y c_2^1) = 0, \quad (38)$$

$$-\mathbf{n}_2 \cdot (\mathbf{D}_2 \cdot (\nabla_x c_2^0 + \nabla_y c_2^1)) = 0, \quad (39)$$

$$-\mathbf{n}_2 \cdot (\mathbf{D}_2 \cdot (\nabla_x c_2^1 + \nabla_y c_2^2)) = 0. \quad (40)$$

Eq. (26) with (31) and (34) gives

$$c_1^0 = c_1^0(x, t). \quad (41)$$

Then Eq. (27) with (41) becomes

$$\nabla_y \cdot (\mathbf{D}_1 \cdot (\nabla_x c_1^0 + \nabla_y c_1^1)) = 0. \quad (42)$$

Eq. (29) with (37), (38) gives

$$c_2^0 = c_2^0(x, t). \quad (43)$$

From (37a), (41) and (43) we have

$$c^0 = c_1^0(x, t) = c_2^0(x, t), \quad (44)$$

and (35) becomes

$$\mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c^0 + \nabla_y c_1^1) = 0 \quad \text{on } \Gamma_2. \quad (45)$$

Consider Eq. (42) together with (32) and (45). The solution of this problem is well-known (Auriault 1983)

$$c_1^1 = \chi_1 \cdot \nabla_x c^0 + \bar{c}_1^1(x, t), \quad (46)$$

where the vector function χ_1 is Ω_1 periodic with $(\chi_1)_1 = 0$ and is the solution of the cell problem

$$\nabla_y \cdot (\mathbf{D}_1 \cdot (\mathbf{I} + \nabla_y \chi_1)) = 0 \quad (47)$$

with

$$-\mathbf{n}_1 \cdot \mathbf{D}_1 \cdot \nabla_y \chi_1 = 0 \quad \text{on } \Gamma_1 \cap \Gamma_2. \quad (48)$$

Integration of (28) over Ω_1 yields

$$\begin{aligned} \phi_1 \frac{\partial c^0}{\partial t} = \nabla_x \cdot ((\mathbf{D}_1 \cdot (\mathbf{I} + \nabla_y \chi_1))_1 \cdot \nabla_x c^0) \\ + \frac{1}{\Omega} \int_{\Omega_1} \nabla_y \cdot (\mathbf{D}_1 \cdot (\nabla_y c_1^1 + \nabla_y c_1^2)). \end{aligned} \quad (49)$$

Let us transform the integral term in (49)

$$\begin{aligned} \frac{1}{\Omega} \int_{\Omega_1} \nabla_y \cdot (\mathbf{D}_1 \cdot (\nabla_x c_1^1 + \nabla_y c_1^2)) d\Omega \\ = \frac{1}{\Omega} \int_{\Gamma_1 \cup \Gamma_2} \mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c_1^1 + \nabla_y c_1^2) dS \\ = \frac{1}{\Omega} \int_{\Gamma_1} \mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c_1^1 + \nabla_y c_1^2) dS \\ + \frac{1}{\Omega} \int_{\Gamma_2} \mathbf{n}_1 \cdot \mathbf{D}_1 \cdot (\nabla_x c_1^1 + \nabla_y c_1^2) dS \\ = -\alpha \gamma_1 \frac{\partial c^0}{\partial t} - \frac{1}{\Omega} \int_{\Gamma_2} \mathbf{n}_2 \cdot \mathbf{D}_2 \cdot (\nabla_x c^0 + \nabla_y c_2^1) dS. \end{aligned} \quad (50)$$

To obtain (50) we have used (33) and (36). To investigate the last integral term in (50), we integrate (30) over Ω_2

$$\phi_2 \frac{\partial c^0}{\partial t} = \frac{1}{\Omega} \int_{\Omega_2} \nabla_y \cdot (\mathbf{D}_2 \cdot (\nabla_x c^0 + \nabla_y c_2^1)) d\Omega.$$

Then, using the divergence theorem gives

$$\begin{aligned} \phi_2 \frac{\partial c^0}{\partial t} &= \frac{1}{\Omega} \int_{\Gamma_2} \mathbf{n}_2 \cdot \mathbf{D}_2 \cdot (\nabla_x c^0 + \nabla_y c_2^1) dS \\ &+ \frac{1}{\Omega} \int_{\Gamma_3} \mathbf{n}_2 \cdot \mathbf{D}_2 \cdot (\nabla_x c^0 + \nabla_y c_2^1) dS. \end{aligned}$$

With (39) we obtain

$$\frac{1}{\Omega} \int_{\Gamma_2} \mathbf{n}_2 \cdot \mathbf{D}_2 \cdot (\nabla_x c_2^0 + \nabla_y c_2^1) dS = \phi_2 \frac{\partial c^0}{\partial t}, \quad (51)$$

and according to (49), (50), (51) we have

$$(\phi_1 R_1 + \phi_2) \frac{\partial c^0}{\partial t} = \nabla_x \cdot (\mathbf{D}_1^* \cdot \nabla_x c^0), \quad (52)$$

where

$$\mathbf{D}_1^* = \frac{1}{\Omega} \int_{\Omega_1} \mathbf{D}_1 \cdot (\mathbf{I} + \nabla_y \chi_1) d\Omega, \quad (53)$$

$$R_1 = 1 + \alpha \gamma_1 / \phi_1. \quad (54)$$

Note that the macroscopic Eq. (52) contains only the local transient derivative $\frac{\partial c^0}{\partial t}$ in the LHS: diffusion does not appear in Ω_2 . Equation (52) can be rewritten in the form

$$\phi R_{app} \frac{\partial c^0}{\partial t} = \nabla_x \cdot (\mathbf{D}_1^* \cdot \nabla_x c^0), \quad (55)$$

where

$$R_{app} = 1 + \alpha \gamma_1 / \phi. \quad (56)$$

If the IMLZ is absent, the retardation factor R_{app} in (55) becomes $R_d = 1 + \alpha \cdot \gamma_{p,m} / \phi$.

By comparing R_{app} with R_d we obtain $R_{app} < R_d$: the presence of a IMLZ leads to a smaller retardation factor. With $\gamma_1, \alpha, \phi = \text{const}$, equation (55) becomes

$$\phi \frac{\partial c^0}{\partial t} = \nabla_x \cdot (\mathbf{D}_{1app}^* \cdot \nabla_x c^0), \quad \mathbf{D}_{1app}^* = \frac{\mathbf{D}_1^*}{R_{app}} \quad (57)$$

We conclude that the presence of the IMLZ yields a larger apparent dispersion coefficient.

Case 2. $Pe = O(1)$, $Da = O(\varepsilon)$, $D = O(\varepsilon)$. Among equations (19)–(24), only (19) is changed

$$\frac{\partial c_1}{\partial t} + \nabla \cdot (\mathbf{v} c_1) = \nabla \cdot (\mathbf{D}_1 \cdot \nabla c_1). \quad (58)$$

In this case we have

$$\begin{aligned} (\phi_1 R_1 + \phi_2) \frac{\partial c^0}{\partial t} + \nabla_x \cdot ((\mathbf{v}^0)_1 c_1^0) \\ = \nabla_x \cdot (\mathbf{D}_1^* \cdot \nabla_x c_1^0), \end{aligned} \quad (59)$$

where \mathbf{D}_1^* and R_1 are determined by (53) and (54), respectively. By repeating the same process as above, we obtain from (59)

$$\phi \frac{\partial c^0}{\partial t} + \nabla_x \cdot ((\mathbf{v}^0)_{1app} c^0) = \nabla_x \cdot (\mathbf{D}_{1app}^* \cdot \nabla_x c_1^0), \quad (60)$$

where

$$(\mathbf{v}^0)_{1app} = \frac{1}{R_{app}} (\mathbf{v}^0)_1, \quad \mathbf{D}_{1app}^* = \frac{1}{R_{app}} \mathbf{D}_1^*.$$

We see that the presence of the IMLZ yields to a weaker reduction of the apparent fluid velocity and of the dispersion tensor in comparison with the case when all porous space is filled by mobile liquid. In the latter situation we have (Bear, 1972)

$$(\mathbf{v}^0)_{1app} = \frac{1}{R_d} (\mathbf{v}^0)_1, \quad \mathbf{D}_{1app}^* = \frac{1}{R_d} \mathbf{D}_1^*.$$

In other words, the presence of the IMLZ leads to a weakening of the influence of the adsorption on the convection–dispersion phenomena.

Case 3. $Pe = O(\varepsilon^{-1})$, $Da = O(\varepsilon)$, $D = O(\varepsilon)$. Since Pe is larger than $O(1)$ we introduce a second time scale τ , and the time differentiation rule becomes

$$\frac{\partial \cdot}{\partial t} \Rightarrow \frac{\partial \cdot}{\partial t} + \frac{1}{\varepsilon} \frac{\partial \cdot}{\partial \tau}. \quad (61)$$

The two time scales appear only in the convection–diffusion equation in Ω_1 , and in boundary conditions on $\Gamma_1 \cup \Gamma_2$. Due to $\mathbf{v} = 0$ in Ω_2 , one time scale t is present in this region.

Eq. (8) is now written in the form

$$\frac{\partial c_1}{\partial t} + \frac{1}{\varepsilon} \nabla \cdot (\mathbf{v} c_1) = \nabla \cdot (\mathbf{D}_1 \cdot \nabla c_1). \quad (62)$$

In the present case we obtain two macroscopic equations

$$\frac{\partial c^0}{\partial t} + \nabla_x \cdot ((\mathbf{v}^0)_{\Omega_1} c^0) = 0. \quad (63)$$

$$\begin{aligned} (\phi_1 R_1 + \phi_2) \frac{\partial c^0}{\partial t} + \frac{\partial (c_1^1)_1}{\partial \tau} + \nabla_x \cdot ((\mathbf{v}^1)_1 c^0 + (c_1^1 \mathbf{v}^1)_1) \\ = \nabla_x \cdot (\mathbf{D}_{1app}^{**} \cdot \nabla_x c^0), \end{aligned} \quad (64)$$

where

$$\mathbf{D}_1^{**} = \frac{1}{\Omega} \int_{\Omega_1} \mathbf{D}_1 \cdot (\mathbf{I} + \nabla_y \chi_2) d\Omega, \quad (65)$$

and χ_2 is a vector function of the same nature as χ_1 .

When compared to (59), macroscopic equation (64) shows more complex transient and convective terms. By using (18), we can rewrite (64) in the following form

$$\begin{aligned} (\phi_1 R_1 + \phi_2) \frac{\partial c^0}{\partial t} + \frac{\partial \langle c_1^1 \rangle_1}{\partial \tau} + \nabla_x \cdot (\langle v^1 \rangle_1 c^0 + c_1^1 \langle v^0 \rangle_1) \\ = \nabla_x \cdot (\mathbf{D}_{app}^{**} \cdot \nabla_x c^0), \end{aligned} \quad (66)$$

where $\mathbf{D}_{app}^{**} = \mathbf{D}_1^{**} + \mathbf{A} \cdot \nabla_x p^0$, $\mathbf{A} = (\mathbf{k}_f \cdot \chi_2)_1$.

Other cases. It can be shown that **case 4** with $Pe = O(\varepsilon)$, $Da = O(1)$, $D = O(\varepsilon)$ and **case 5** with $Pe = O(1)$, $Da = O(1)$, $D = O(\varepsilon)$ lead to situations which are non-homogenizable (Auriault 1991, Auriault & Lewandowska 1993b, 1996). In **case 6** with $Pe = O(\varepsilon^{-1})$, $Da = O(1)$, $D = O(\varepsilon)$ we obtain the following macroscopic eqns

$$\phi_1 R_1 \frac{\partial c^0}{\partial \tau} + \langle v_0 \rangle_1 \cdot \nabla_x c^0 = 0. \quad (67)$$

$$\begin{aligned} \phi \frac{\partial c^0}{\partial t} + \frac{\partial \langle c_1^1 \rangle_1}{\partial \tau} + \alpha \frac{\partial \langle c_1^1 \rangle_{\Gamma_1}}{\partial t} \\ + \nabla_x \cdot (\langle v^1 \rangle_1 c^0 + \langle v^0 c_1^1 \rangle_1) \\ = \nabla_x \cdot (\mathbf{D}_1^{***} \cdot \nabla_x c^0), \end{aligned} \quad (68)$$

where

$$\mathbf{D}_1^{***} = \frac{1}{\Omega} \int_{\Omega_1} \mathbf{D}_1 \cdot (\mathbf{I} + \nabla_y \chi_3) d\Omega, \quad (69)$$

and χ_3 is a vector function which is the solution to the corresponding local cell problem. Similarly to (66), eq. (68) can be written in the form

$$\begin{aligned} \phi \frac{\partial c^0}{\partial t} + \frac{\partial \langle c_1^1 \rangle_1}{\partial \tau} + \alpha \frac{\partial \langle c_1^1 \rangle_{\Gamma_1}}{\partial t} \\ + \nabla_x \cdot (\langle v^1 \rangle_1 c^0 + \langle \bar{c}_1^1 \rangle_1 \langle v^0 \rangle_1) \\ = \nabla_x \cdot (\mathbf{D}_{app}^{***} \cdot \nabla_x c^0), \end{aligned} \quad (70)$$

where $\mathbf{D}_{app}^{***} = \mathbf{D}_1^{***} + \mathbf{A} \cdot \nabla_x p^0$, $\mathbf{A} = (\mathbf{k}_f \cdot \chi_3)_1$.

The above obtained results show that only cases with $Da = O(\varepsilon)$, $D = O(\varepsilon)$ and $Pe = O(\varepsilon), O(1)$ yield qualitatively similar models to phenomenological model (3)–(4). In all other cases analyzed here, macroscopic equations are different.

5 CONCLUSIONS

Let us summarize our results. In cases with $Da = O(\varepsilon)$, $D = O(\varepsilon)$ and $Pe = O(\varepsilon), O(1)$ the presence of the IMLZ leads to the reduction of the retardation factor of the porous media. The macroscopic equations are of diffusion or convection–diffusion type, with a source/sink term. The latter characterizes the presence of the IMLZ. At large Péclet numbers, the macroscopic solute transport process can be divided into two stages: 1) at small times (time scale τ) only solute convection with local transient change of the macroscopic concentration c^0 (Eqs (63), (67)) is observed; 2) at longer times (time scale t) we have a convection–diffusion process with complex transient phenomena (Eqs (64), (68)).

In presence of strong adsorption ($Da = O(1)$) and small or moderate Péclet numbers ($Pe = O(\varepsilon), O(1)$) the situation is non-homogenizable. When $Pe = O(\varepsilon^{-1})$, both transient behaviour and dispersion tensor are dependent on adsorption, among other parameters.

It was shown that phenomenological models by Coats & Smith (1964), van Genuchten & Wierenga (1976) are in good agreement with the macroscopic models obtained here in cases with $Da = O(\varepsilon), D = O(\varepsilon)$ and $Pe = O(\varepsilon), O(1)$, only.

In conclusion, the presence of IMLZ in porous media can considerably alter the solute concentration field in the MLZ and then alter the macroscopic concentration field.

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The work proposed under the classical constraint of length scale separation. The main goal is further investigated from a structural point of view by solving the associated discrete problem over a simple periodic model of regular medium. Equivalent permeability results are validated by comparison with experimental data obtained on the same geometrical configuration over a wide range of excels and by using its approximations resulting from permeability composition. Finally, two-phase flow is investigated from a phenomenological point of view by performing oil-water flow experiments on the same type of medium.

1. INTRODUCTION

1.1. General background

Vugular porous media are a special type of heterogeneous media made of a porous matrix embedding large pores (referred to as vugs) which characteristic length-scale is much larger – by several orders of magnitude – than the characteristic pore size of the matrix. This type of medium is commonly encountered in geological formations such as carbonate or limestone reservoirs and is of particular interest in petroleum engineering for instance. Typical diameter of vugs ranges between one millimetre and one centimetre and their presence often results from local frequent dissolution. Core analysis on vugular samples has often exhibited some unexpected behavior like early water breakthrough during imbibition experiments (de Zabala & Kanuth 1995) and this suggests that a particular attention should be dedicated to the macroscopic model and transfer properties of such media. In this work, attention is focused on molecular transport in vugular porous media for one-phase flow from a theoretical, analytical and experimental point of view while preliminary results of observation during two-phase flow in a model vugular medium are presented.

1.2. Purpose of the study

The situation under consideration is depicted in Figure 1 where we have represented the three different scales under concern: the pore scale with the characteristic length scales L_p and l_p for the solid and fluid phase respectively; the Darcy scale at which matrix and vugs can be clearly identified with their respective



Figure 1. Typical scales for vugular porous media.

characteristic length scales L_v and l_v and the large scale for which the typical size is L . The purpose is first to investigate the possibility of representing the vugular structure over the volume V by an equivalent homogeneous medium for one-phase creeping flow, and evaluate the equivalent permeability of the medium. Secondly, we started observing the behavior of such a system during two-phase oil-water flow.

2. UP-SCALING AND MACROSCOPIC MODEL

2.1. Boundary value problem

In order to derive one-phase flow model on an equivalent homogeneous medium over volume V the starting