

Computation of the longitudinal and transverse dispersion coefficient in an adsorbing porous medium using homogenization

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Abstract

In a pioneering paper Bouddour, Auriault and Mhamdi-Alaoui derive upscaled expressions for the dispersion coefficients for reactive flow in a porous medium using the method of homogenization. The method uses a periodic unit cell (PUC), which consists for instance of a spherical grain in a cube, but nothing prohibits defining more complex PUC's. The method leads to a coupled system of equations where the flow is described by Stokes equation and the concentration fluctuation is described by a convection diffusion source term equation. In the PUC we have semi-infinite boundary conditions (BC's). The solution of the equation is not trivial due to the source term and the BC's. We show that commercial finite element software (COMSOL) can be readily used to compute longitudinal and transversal dispersion coefficients in 2-D and 3-D; this makes homogenization accessible to the engineering practice. Details of the complete numerical procedure are discussed in detail. The results are for the first time compared with experimental data of the dispersion coefficient versus Peclet number; there is good agreement. Adsorption enhances the longitudinal dispersion coefficient.

Introduction

A relatively new upscaling method [8], called homogenization, has the advantage that it does not need closure relations [3] [10] [11] [5]. The derivation of the upscaled model equations in homogenization is quite general [8]. Still, for the computation of the transport coefficients, homogenization makes the assumption that the medium is periodic (see, however, [9]). The reason that the method

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has no widespread engineering applications is because it is conceptually difficult and also because the computation of the transport coefficients requires solving non-conventional equations involving periodic boundary conditions. Tardif d’Hamonville et al. [13] developed a 3-D finite element code and solved the equations derived in [3] numerically to find values of all the components of the dispersion tensor as a function of the Peclet number. Their range of interest is confined to relatively low Peclet numbers ($Pe < 5$). We compute the longitudinal dispersion (sum of hydrodynamic dispersion and tortuosity corrected molecular diffusion) both in the absence or presence of retardation (adsorption), using commercial finite element software (COMSOL); this makes homogenization accessible to the engineering practice.

Section 1 presents some background concepts on dispersion. We give an overview of the idea behind homogenization 2, and mention the derived model equations in Section 3. Section 4 deals with the numerical implementation of the ensuing model equations in COMSOL. Section 5 makes a comparison between homogenization data and laboratory experiments reported in the literature. We end with some conclusions.

1 Dispersion

On the microscale, the velocity field is not constant but varying under the influence of the structure of a porous medium. As a result fluid elements containing contaminant particles with more or less the same initial position at $t = 0$ travel along different paths at different velocities. At the pore-scale diffusion mixing between fluid elements arriving at the same time close to the same pore-space occurs. The dispersion coefficient due to velocity heterogeneity and subsequent mixing on the *pore-scale* is called *macroscopic dispersion* or sometimes *microscopic dispersion coefficient*. It spreads the concentration profile. The additional transport term is implemented in the upscaled equation by replacing the molecular diffusion by a dispersion tensor \mathbf{D} . The tensor \mathbf{D} is the sum of the isotropic-tortuosity corrected molecular diffusion tensor $D_m \mathbf{I}$ and the hydrodynamic dispersion tensor \mathbf{D}_d . The tensor can be diagonalized by choosing the flow direction as the main axis in the coordinate system. The component in main flow direction is called the longitudinal dispersion coefficient, whereas the components perpendicular to the main flow direction are called the transverse dispersion coefficient. A common way to present the longitudinal dispersion coefficient is to make a plot of its experimental value versus the Peclet number. The Peclet number is a dimensionless number that expresses the ratio between convective and molecular diffusive transport. A high Peclet number indicates a high fluid velocity and the dispersion tensor components tend to have higher values at high Peclet numbers [1]. At low velocities (low Peclet numbers) molecular diffusion is the most important mixing mechanism. However at higher velocities (high Peclet numbers) mechanical dispersion becomes increasingly important.

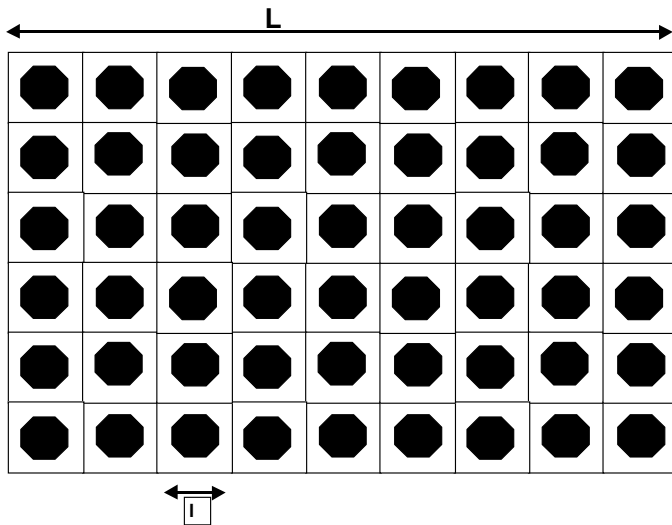


Figure 1: A periodic lattice. In each cell we have a void space and an impermeable grain shown as an octaeder. The length of the entire lattice is L and the length of a single unit cell is l , where $\varepsilon = \frac{l}{L} \ll 1$.

2 Procedure for homogenization

Consider a system of macroscopic (global scale) dimensions with a characteristic length L (see Figure 1). The macroscopic domain consists of a collection of periodic unit cells, on the microscopic (local) scale with characteristic length l . To ensure the separation of scales we need that $\varepsilon = l/L \ll 1$. In our example the periodic unit cell (PUC) (Fig. 2) consists of a (soil) grain in the center with a single phase fluid flowing around it. However, the method can easily be extended using more complex unit cells. The PUC plays a analogous role as the representative elementary volume (REV) [2], but is usually smaller.

Inside the PUC fluid flows according to Stokes law, using periodic boundary conditions and using that the flow along and perpendicular to the grain surface Γ is zero.

The transport equations are non-dimensionalized on inspection [12], where we split all spatial differentiation into a contribution on the local scale and on the global scale. In this procedure L is the characteristic length for global differentiation and l is the characteristic length for local differentiation. We obtain an equation with the usual dimensionless numbers e.g the Peclet number and ε . The magnitude of the dimensionless numbers with respect to ε will be determined. Then we expand the dependent variables into contributions of decreasing significance with respect to ε . After substitution the transport equa-

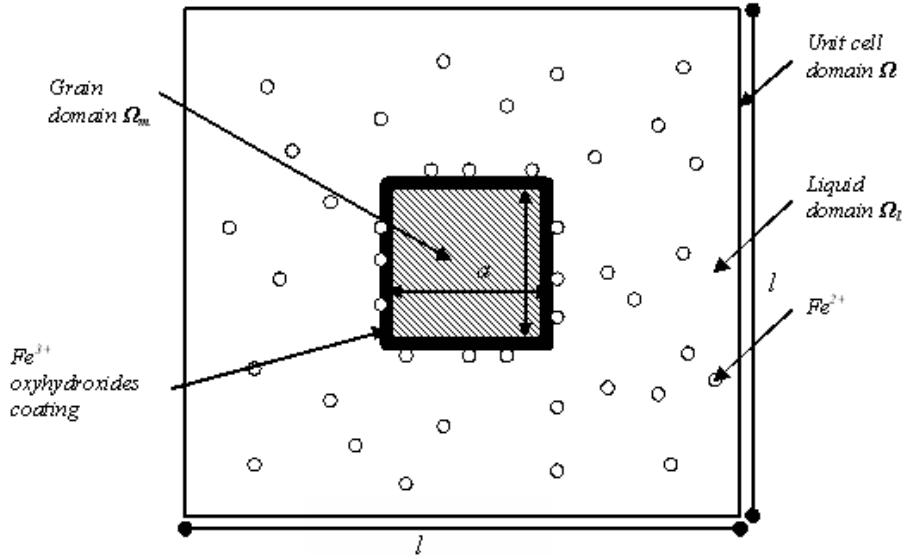


Figure 2: Periodic unit cell and relevant domains. The picture shows the oxyhydroxide coating and the Fe^{2+} ions (not drawn to scale for reasons of illustration).

tions will consist of a sum of terms each with a different order with respect to ε . The upscaling procedure should be valid for a range of ε values of the same order of magnitude. Each of the terms with a specific order in ε constitutes an equation that is separately satisfied. First the equation of the lowest order in ε is solved leading to solutions of the most significant contributions of the dependent variables. Subsequently the equation with one order higher in ε is solved using the previously obtained relations. Finally the next order ε equations are averaged over the PUC leading to the upscaled equation. Combining the results also the expressions for the upscaled parameters, e.g., the dispersion coefficient are obtained.

2.1 Stokes equation

We assume that the flow in the "void" space is governed by Stokes equation. The Stokes equation can be straightforwardly solved numerically introducing the stream function (because we assume incompressible flow $\nabla \cdot \mathbf{v} = \mathbf{0}$) and the vorticity (see [7]). As a result we find in each periodic unit cell the same velocity distribution, because in each PUC the flow is subjected to the same potential gradient. As a result the velocity does not change on the global scale. Only local velocity variations i.e. within the PUC are relevant. It is convenient to separate the divergence ∇ into a global ∇_b and a local ∇_s contribution. We

notice that $\nabla_b \cdot \mathbf{v} = 0$ because of the periodicity assumption and therefore we obtain from $\nabla \cdot \mathbf{v} = \nabla_b \cdot \mathbf{v} + \nabla_s \cdot \mathbf{v} = 0$

$$\nabla_b \cdot \mathbf{v} = 0, \text{ and } \nabla_s \cdot \mathbf{v} = 0 \quad (1)$$

The Stokes flow problem within the periodic unit cell can be solved such that we obtain the velocity field $\mathbf{v} = \mathbf{v}(\mathbf{r}_s)$.

2.2 Solute transport

The convection diffusion equation inside the PUC i.e. in Ω_l can be written as

$$\frac{\partial c}{\partial t} + \mathbf{div}(\mathbf{v}c) = \mathbf{div}(D \mathbf{grad} c) \quad (2)$$

where c is the molar Fe^{2+} concentration, \mathbf{v} the velocity field obtained from the solution of the Stokes equation and D is the molecular diffusion coefficient.

The boundary condition at the grain surface is written as

$$\begin{aligned} (-D \mathbf{grad} c)_n &= \delta \left(\frac{\partial c_a}{\partial t} \right)_\Gamma & \text{at } \Gamma \\ \mathbf{v} &= \mathbf{0} & \text{at } \Gamma, \end{aligned} \quad (3)$$

where c_a is the equilibrium surface concentration at the boundary. Note that $c_s = \delta c_a / (\ell^2 - a^2)$. Periodic boundary conditions are used at the boundaries of the PUC.

3 Upscaled equations

The procedure of homogenization leads to an expression of the dispersion coefficients as follows

$$\mathbf{D}_d = -\frac{D_0 Pe}{\varphi} \frac{1}{|\Omega_l|} \int_{\Omega_l} \mathbf{v} \otimes \chi \, d\mathbf{r}_s \quad (4)$$

and

$$\mathbf{D}_m = \frac{D_0}{\varphi} \left(\varphi \mathbf{I} + \frac{\varphi}{|\Omega_l|} \int_{\Omega_l} \mathbf{grad}_s \otimes \chi \, d\mathbf{r}_s \right) \quad (5)$$

where \mathbf{v} is the dimensionless velocity field and χ is the concentration fluctuation in the unit cell. The velocity field is computed using Stokes equation, a standard option in COMSOL. We use the notation for the dyadic product $\mathbf{v} \otimes \chi$ in which each component of one vector multiplied by another component of the other vector forms a matrix.

For the x-component of χ we can derive the source, convection diffusion equation

$$-\frac{\bar{v}_x}{R} + \mathbf{div}_s (\mathbf{v}(\chi_x + x_s)) = \frac{1}{Pe} \mathbf{div}_s \mathbf{grad}_s (\chi_x + x_s) . \quad (6)$$

with boundary conditions

$$\frac{1}{Pe} (\mathbf{grad}_s (\chi_x + x_s)) \cdot \mathbf{n} = 0 \text{ at } \Gamma , \quad (7)$$

where x_s is the x -coordinate in the unit cell. Here R is the retardation factor.

The concentration fluctuation χ_x is periodic in the unit cell. Equations for the other components of χ can be derived like-wise. The problem of implementing the equation in COMSOL is related to the fact that $(\chi_x + x_s)$ satisfies semi-periodic boundary conditions i.e., a unit concentration at the right must be added to the concentration χ_x at the left.

4 Numerical calculation of the dispersion coefficient

In order to calculate the full dimensional hydrodynamic longitudinal dispersion and effective diffusion coefficients we need to compute the first order concentration correction χ_x (χ_y). We solve the problem both in 3-D using a Finite Element Method software package, COMSOL Multiphysics. COMSOL, formerly FEMLAB is a software package that can solve various coupled engineering and physics problems, e.g., here a combination of Stokes and the convection diffusion equation with a source term. The benefit of using COMSOL with respect to the vorticity approach [4] is that it is capable of solving the problem in three dimensions. COMSOL also allows to compute the volume integrals in Eqs. (4), (5).

For reasons of easy comparison with reference [13] we have defined the geometry in Fig. 3 for the PUC in 3-D. To obtain the unit cells in Fig. 3, we start with a unit cube. In each of the eight corners of the cube, spheres, representing the grains, with radii of 0.583 or 0.510 have been drawn corresponding to porosities of 0.242 and 0.446 respectively. The parts of the sphere that fall outside the unit cube are discarded, whereas the parts inside the cube constitute the grains. The overlapping parts of the spheres inside the cube belong to the porous skeleton.

The steady state incompressible Stokes equation $\mathbf{grad} p = \mu \mathbf{div} \mathbf{grad} \mathbf{v}$ was implemented by setting the appropriate parameters equal to zero, i.e., discarding the inertia term. The internal flow boundaries are set to a no slip condition, i.e., zero velocity at the grain surface. Symmetry conditions at the cube faces parallel to the main flow direction are equivalent to periodic boundary conditions if the sphere is located at the center of the PUC. The semi-periodic boundary conditions for symmetric PUCs can be set by choosing constant pressures at the cube faces perpendicular to the main flow direction. They were chosen such that the average dimensionless longitudinal velocity \bar{v}_x is equal to $1/\varphi$. This choice

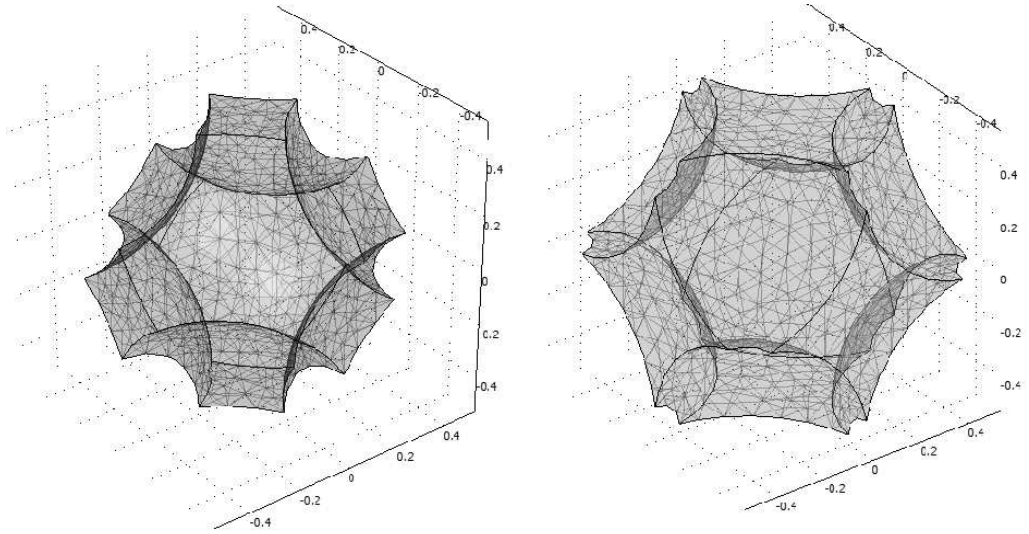


Figure 3: Part of the periodic unit cell (cube) filled with fluids left (a): Coarse Finite Element mesh with radius of spheres at the corner $a = 0.583$ and right (b): finite element mesh for $a = 0.510$ The average x-velocity is equal to $1/\varphi$.

of setting $\bar{v}_x = 1/\varphi$ reduces the number of calculations since we use this velocity as a factor in the source term of Eq. (6). This is accomplished as follows: in the dimensionless calculation in the unit cell with side $\xi = 1$, a pressure difference is applied such that the volume integral of the x-Darcy-velocity in the flowing part is equal to one, and the choice of the Peclet number is made by varying the molecular diffusion coefficient. Here, taking the volume integral equal to one, means that the average Darcy velocity \bar{u} or specific discharge is one and thus \bar{v}_x is equal to $1/\varphi$. However, it is possible to use any value for \bar{u} as long as the Peclet number $Pe = \bar{u}\xi/(\varphi D_0)$. In the literature [2] the Peclet number is usually defined as $\bar{v}d_p/D_0$, where the interstitial velocity $\bar{v} = \bar{u}/\varphi$ and d_p is the grain diameter. For our choice of inscribed radii ($a = 0.51$ or $a = 0.583$) this only leads to a minor difference.

Indeed, the advantage of this procedure is that the solution for the velocity field does not change for computations with various Peclet numbers. We can solve the cell equation Eq. (6) for the longitudinal dispersion coefficient with the stored velocity field as input. We also use boundary condition (7). It turns out to be advantageous to use the diffusion equation in the transient mode: $\partial c/\partial t + \mathbf{v} \cdot \mathbf{grad} c = \mathbf{div}(D \mathbf{grad} c) + S_R$, where we use $c = \chi_x + x_s$ for the longitudinal coefficients. The source term is only non-zero for longitudinal dispersion, i.e., $S_R = \bar{v}_x/R$ while various values of $D = \frac{1}{Pe}$ are used. For long times the solution converges to the solution of the stationary reaction-diffusion-convection equation, i.e., $\mathbf{v} \cdot \mathbf{grad} c = \mathbf{div}(D \mathbf{grad} c) + S_R$.

For the concentration equation semi-periodic boundary conditions are used in the flow direction. To compute the longitudinal dispersion coefficient we use $c(x = 0, y, z, t) = c(x = 1, y, z, t) + 1$ in the flow direction x while we use strictly periodic boundary conditions in the other directions, e.g., $c(x, y = 0, z, t) = c(x, y + 1, z, t)$. For reasons of symmetry the latter can also be replaced by the no flow/symmetry boundary condition. To implement semi-periodic boundary conditions in COMSOL, it appears to be necessary to choose the appropriate Neumann precondition at the inflow and outflow boundary; in this case we implement the Convective Flux condition $(-D \mathbf{grad} c) \cdot \mathbf{n} = 0$ to ensure that the diffusive flux or the concentration gradient is also periodic. This aspect was not incorporated in the COMSOL manual. The remaining external boundaries and the internal boundaries have insulation symmetry boundary conditions, $(-D \mathbf{grad} c + c\mathbf{v}) \cdot \mathbf{n} = 0$. This leads to strictly periodic boundary conditions for χ_x . The transient solution for the convection diffusion equation reaches a steady state solution if the source term $S_R = -\bar{v}_x/R$ counterbalances the difference between the convective flux entering and leaving the PUC. COMSOL can solve the Stokes equation as well as the convection diffusion equation in their conservative form. A Multigrid preconditioner presolves the linear set of equations before COMSOL applies the Generalized Minimal Residual Method (GMRES).

5 Results

Fig. 4 shows the 3-D results. The 3-D simulation with the corner spheres of radii $a = 0.510$ (0.583) was carried out with 5832 (3128) mesh points, with 27420 (14495) tetrahedral Lagrangian quadratic elements. COMSOL uses $\text{shlag}(2, 'c')$ shape functions with integration order 4 and constraint order 2. A simulation with 1731 (955) mesh points and 7746 (4068) elements gave results that deviated at most 0.133% (0.288%). At low Peclet numbers the longitudinal and transversal dispersion coefficient are dominated by the molecular diffusion and from the definition used by engineers they do not depend on the porosity. For low Peclet numbers transverse and longitudinal dispersion are equal. For the configuration in Fig. 3a (3b) D/D_o assumes values of 0.51 (0.69). The measured value is $D \sim 0.7D_o$, is only attained for the configuration shown in Fig 3b. This configuration has a porosity value $\varphi = 0.446$, close to many laboratory tests ($\varphi = 0.35 - 0.45$). The configuration of Fig 3a has a porosity value of 0.242, and here we find $D \sim 0.5D_o$ for low Peclet numbers. At high Peclet numbers the longitudinal D/D_o values increase faster than proportional to the Peclet number. The computed results show the same trends as the experimental results collected for instance in [2]. Deviations between the theoretical results for the configurations of Fig. 3a and 3b are well within the range of experimentally determined values [6].

It should be noted that laboratory experiments to obtain these data are not trivial due to the low values of the dispersion coefficients. Many experimental

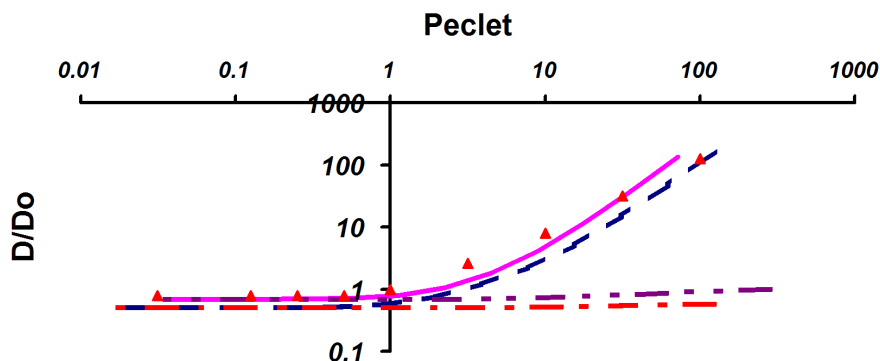


Figure 4: Longitudinal (upper curves) and transverse (lower curves) dispersion divided by molecular diffusion versus Peclet number. The Peclet number is based on the interstitial velocity $v = u/\varphi$. The characteristic dimension is the size of the unit cell. Dashed (dashed-dot-dot) line has unit cell as Fig. 3a (left) and the drawn (dashed-dot) line as Fig. 3b (right). The triangles denote experimental points [2]

data that show large values can be incorrect due to stream line splitting at the entrance and production point, i.e., special fluid distributors at the injection and production point were not used. Small entrapped air bubbles also can cause an apparent increase of the dispersion coefficient. Finally also the sand pack must be homogeneous, which requires special experimental preparation techniques [14]. Suspicious data have been discarded as much as possible in the plot in [2]. High quality data are given here as the triangles in Fig. 4.

Conclusions

- Homogenization is a useful method to obtain upscaled equations and the corresponding transport coefficients in porous media. The method leads to upscaled equations and coefficients for practical implementation that are less dependent on intuition than upscaled equations obtained with REV averaging.
- The derivation of the upscaled equations can be simplified by taking the computation of the Stokes flow field out of the bracket of the homogenization procedure.
- Explicit expressions for the dispersion tensor are obtained, based on comparison to the convective diffusion equation used for contaminant transport. Finite Element Method software, e.g., COMSOL can be used to

solve the unconventional equations, the solution of which is necessary to obtain quantitative results. This makes it possible to use homogenization as an engineering tool.

- The computed longitudinal dispersion coefficients as a function of the Peclet number show good agreement with experimental literature data. This implies that homogenization cannot only be used to obtain upscaled equations but also to obtain estimates of the transport coefficients.
- The new element obtained here from homogenization for modeling transport in porous media is that the longitudinal dispersion coefficient is enhanced if equilibrium adsorption occurs at the grains. The enhancement, even at retardation factors of 10, is small. The transverse dispersion coefficient is not affected by adsorption.

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