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APPLIED MICROMECHANICS OF POROUS MATERIALS

EDITED BY

LUC DORMIEUX
ECOLE NATIONALE DES PONTS ET CHAUSSEES,
CHAMPS SUR MARNE, FRANCE

FRANZ-JOSEF ULM
MASSACHUSETTS INSTITUTE OF TECHNOLOGY,
CAMBRIDGE MA, U.S.A.

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PREFACE

All natural composite materials (soils, rocks, woods, hard and soft tissues, etc.) and many engineered composites (concrete, bioengineered tissues, etc.) are multiphase and multiscale material systems. The multiphase composition of such materials is permanently evolving over various scales of time and length, creating in the course of this process the most heterogeneous class of materials in existence, with heterogeneities that manifest themselves from the nanoscale to the macroscale. The most prominent heterogeneity of such natural composite materials is the porosity, ie. the space left in between the different solid phases at various scales, ranging from interlayer spaces in between minerals filled by a few water molecules, to the macropore space in between microstructural units of the material in the micrometer to millimeter range. This porosity is the key to understanding and prediction of macroscopic material behavior, ranging from diffusive or advective transport properties to stiffness, strength and deformation behavior.

The specific nature of the mechanical behavior of multiphase porous materials was early on recognized in the groundbreaking works of M.A. Biot and K. Terzaghi, who developed the macroscopic basis of what is now known as 'Poromechanics'. Ever since, poromechanics has entered a large number of engineering applications ranging from civil and environmental engineering to petroleum engineering and more recently biomechanical engineering. In the 1970's, a breakthrough was achieved with pioneering works that relate macroscopic laws to microstructural properties. Furthermore, as new experimental techniques such as nanoindentation, now provide an unprecedented access to micromechanical properties and morphologies of materials, it becomes possible to trace these features from the nanoscale to the macroscale of day-to-day engineering applications, and predict transport properties, stiffness, strength and deformation behaviors within a consistent framework of 'Micromechanics of Porous Media'. The focus of this course which took place in July 2004, was to review fundamentals and applications of this rapidly emerging discipline of Applied Mechanics and Engineering Science.

This book assembles the lecture notes on 'Applied micromechanics of porous materials'. It is composed of three parts: (I) Transport properties of porous media; (II) Microporomechanics; (III) Materials Applications. Part I and II introduces the two fundamental homogenization theories of micromechanics of porous media, namely asymptotic expansion techniques and linear and nonlinear mean-field theories based on the concept of a representative elementary volume (Part II). The first is illustrated for the upscaling of fluid mass transport phenomena through the pore space that involve both advection and diffusion, and allows for a rigorous derivation of the permeability and tortuosity tensor. Linear and nonlinear mean-field theories are most effective for upscaling of the elastic and inelastic solid response of porous materials, which is illustrated for cracked porous media and for plastic deformation of saturated *porous materials. Finally, the combination of microporomechanics theory with advanced experimental micromechanical techniques (incl. Nanoindentation, Atomic Force Microscopy and Environmental Scanning Electron Microscopy) is illustrated in Part III of these lecture notes, and is applied to the multiscale investigation of the poroelastic properties of cement-based materials, shales and bones.*

From the onset, the course was designed as a well-balanced blend of theory and hands-on application of microporomechanics to a large range of porous materials in the linear and nonlinear regimes. Intended for a first course in 'micromechanics of porous media' for graduate students, researchers and engineers working at the forefront of Engineering Mechanics and Materials Science, we trust that these lecture notes be a source of imagination.

Luc Dormieux
Franz-Josef Ulm

CONTENTS

Part I

Transport properties of porous media

Transport in Porous Media: Upscaling by Multiscale Asymptotic Expansions

Jean-Louis Auriault*

* Laboratoire Sols, Solides, structures (3S), UJF, INPG, CNRS UMR 5521, Grenoble, France

Abstract Transport in porous media is investigated by upscaling the pore scale behaviour. We use the technique of multiscale asymptotic expansions which seems to be the most efficient method to obtaining macroscopic equivalent behaviours. Different transport phenomena are addressed: fluid flow through a saturated porous medium (Darcy's law), diphasic flow (coupled Darcy's laws), solute transport (diffusion, advection, dispersion) and fluid flow through deformable porous media (consolidation).

1 Physical Motivation

Heterogeneous media with a large number of heterogeneities cannot be described by considering each of the heterogeneities, that would yield to intractable boundary value problems. The well known method is to replace, *if possible*, the heterogeneous media by an homogeneous one, the description of which is valid at a very large scale (the macroscopic scale) with respect to the heterogeneity scale. As a continuous equivalent description, the derived macroscopic behaviour should be intrinsic to the medium and to the excitation, i.e. it should be independent of the macroscopic boundary conditions.

There are two main ways of deriving this macroscopic description. The first one is a directly macroscopic approach, which is often associated with experiments and is called the phenomenological approach. Many physical laws have been first derived by this kind of approaches. It is the case of Darcy's law, Darcy (1856) and of Biot's laws Biot (1962). The second kind of continuous approach allows to derive the macroscopic behaviour from the local description. This is an upscaling technique. Upscaling techniques allow the derivation of an equivalent macroscopic continuous description from the description of a Representative Elementary Volume (REV). The equivalent description is called the homogenized description. Different techniques are available which address random as well as periodic heterogeneous media. Among all, the most popular are the statistic modelling (Kröner, 1972), the self-consistent method, see Zaoui (1987), the volume averaging method, see, e.g., Nigmatulin (1981) and the homogenization for periodic structures (Sanchez-Palencia, 1974; Keller, 1977; Bensoussan et al., 1978).

When an upscaling technique is in use, the macroscopic behaviour is derived from the description at the heterogeneity scale that describes the physical process over a representative elementary volume (REV). The existence of a such a volume is required for any continuous macroscopic representation of the physical system, and, as a consequence, is required for applying any upscaling technique. By definition, the REV is i) sufficiently large for representing the heterogeneity scale[1], and ii) small compared to the macroscopic volume. As a consequence, a condition of separation of scales is required. This fundamental condition can be expressed as:

$$\frac{l}{L} = \varepsilon << 1, \tag{1.1}$$

where l and L are the characteristic lengths at the REV scale and at the macroscopic scale, respectively.
This definition intuitively conjures up a geometrical separation of scales, whereas this fundamental condition must also be verified regarding the excitation (i.e., the physical process). For instance, consider the propagation of a wave in an heterogeneous medium. The wavelength actually constitutes a third characteristic length. Intuitively, we see that a continuous approach for describing this physical process will be possible only if the heterogeneity scale is small compared to the wavelength; a wavelength of the order of the heterogeneity length scale would lead to wave-trapping effects, which could not be completely described by an equivalent continuous behaviour at the macroscopic scale. For fluid flow in porous media as the medium is excited by a pressure gradient, the characteristic length of the excitation is thus related to the pressure gradient.
Therefore, the fundamental condition of separation of scales is expressed as $l/L << 1$, where L is the macroscopic characteristic length and is either geometrical or related to the excitation: the existence of the REV and, as a consequence, the condition of separation of scales, are not only constrained to geometrical considerations but also related to the excitation (i.e., the physical process). The analysis should focus on what we call the physical system, that consists of both the medium and the excitation.

In the lecture, we use the *method of homogenization for periodic structures*, which is also called the method of multiscale asymptotic expansions to investigate transport in porous media. This method has been introduced by Sanchez-Palencia (1974, 1980), Keller (1977), and Bensoussan et al. (1978). More recently, a more physical methodology based on dimensionless analysis has been introduced by Auriault (1991). This approach highlights the conditions under which homogenization can be applied. The presentation of this methodology is the purpose of the present paper. In this technique, a systematic use is made of the separation of scale parameter which, although small, is not generally null ($\varepsilon \neq 0$). That procures some advantages to the technique: (1) avoiding prerequisites at the macroscopic scale: the macroscopic equivalent description is obtained from the heterogeneity scale description plus the condition of separation of scales, only; (2) modelling finite size macroscopic samples and phenomena with finite macroscopic characteristic lengths ($\varepsilon \neq 0$); (3) modelling macroscopically nonhomogeneous media or phenomena;

[1]The REV size is typically equal or less than times the heterogeneity size, Cherel et al. (1988); Anguy et al. (1994). Therefore, a macroscopic equivalent behaviour cannot be deduced from an arbitrary boundary value problem concerning a single such REV: the result will be strongly dependent on the boundary conditions in use.

(4) modelling problems with several separations of scales by introducing several separation of scales parameter; (5) modelling several simultaneous phenomena; (6) determining whether the system "medium+phenomena" is homogenizable or not, i.e., whether or not a continuous equivalent macroscopic description exists; and (7) providing the domains of validity of the macroscopic models.
Evidently, real porous media are rarely periodic. However, there exists similarity in the behaviour of periodic and random media, on the condition that a separation of scales is present. Consider the right-angled parallelepipedal REV Ω_{REV} of a random medium. Construct a period, Ω, by introducing three successive plane symmetries with respect to three nonparallel faces of Ω_{REV}. The periodic medium of period Ω, and the random medium of REV Ω_{REV}, possess similar structure of their macroscopic description (although an eventual anisotropy could be modified by the plane symmetries). Therefore, we assume in the following that the heterogeneous medium is periodic, without lost of generality. In many applications, it was shown that this gives reasonable results (Quintard and Whitaker, 1993) for effective parameters.

In part 2, we rapidly present the method of multiscale asymptotic expansions. Then we successively address flow in saturated rigid porous media in part 3, diphasic flow in part 4, solute transport in part 5 and flow in deformable porous media in part 6. All these investigations show two separated scales, only.

2 Multiscale Asymptotic Expansion Technique

A periodic medium of period Ω and a separation of scales, $l/L = \varepsilon \ll 1$ are assumed. Such a period of a porous medium is shown in Fig.1. Let $\mathbf{X}$ be the physical space

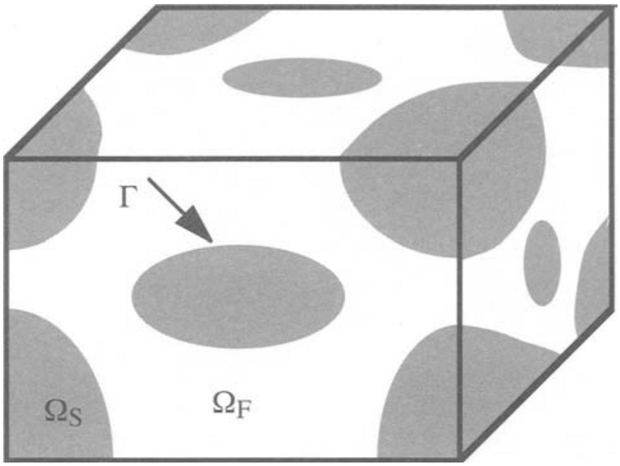

Figure 1. Period of the porous medium

variable of the medium. From the two characteristic lengths l and L, two dimensionless space variables can be defined:

$$\mathbf{y} = \mathbf{X}/l \quad \text{is the dimensionless "microscopic" space variable,}$$
$$\mathbf{x} = \mathbf{X}/L \quad \text{is the dimensionless "macroscopic" space variable.}$$

As a consequence of the separation of scales, $\mathbf{y}$ and $\mathbf{x}$ are two separated variables. Any space dependent quantity φ required to describe the physical process can be described, *a priori*, as a function of these two separated variables:

$$\varphi = \varphi(\mathbf{y}, \mathbf{x}). \tag{2.1}$$

Obviously, these space variables are related by $\mathbf{x} = \varepsilon \mathbf{y}$. The macroscopic equivalent model is obtained from the description at the heterogeneity scale by (Auriault, 1991):
1. assuming the medium to be periodic, without loss of generality since the separation of scales condition is fulfilled. That implies that the different φ's are periodic with respect to space variable $\mathbf{y}$. Note that the eventual dependence of φ with respect to $\mathbf{x}$ shows the eventual macroscopic inhomogeneity of the medium,
2. writing the local description in a dimensionless form, by using l or L as the characteristic length (the choice of l or L is a technical point of no effect on the final result) and by using characteristic values φ_c for the different quantities φ entering the local description: $\varphi = \varphi_c \varphi^*$, where an asterisk shows a dimensionless quantity,
3. deriving dimensionless numbers from the governing equations and evaluating these

dimensionless numbers with respect to the scale ratio ε. A dimensionless number Q is said to be $\mathcal{O}(\varepsilon^p)$ if

$$\varepsilon^{p+1} << Q << \varepsilon^{p-1}. \tag{2.2}$$

For the sake of simplicity, we assume here that all dimensionless numbers can be evaluated by integer powers of ε. Once all dimensionless numbers have been estimated, the governing equations can then be written as :

$$\sum_{q=0}^{\infty} \varepsilon^q \ A^{*(q)} = 0, \tag{2.3}$$

where the q's are integers. The $A^{*(q)}$'s are generally some dimensionless operators. Although the value of ε is fixed in each point of the medium for a given upscaling problem, the corresponding macroscopic equivalent model at stake, if it exists, is valid for all values of ε that respect the above evaluations of the dimensionless numbers. Thus parameter ε can be considered as varying in some range.
4. looking for the dimensionless unknown fields in the form of asymptotic expansions in powers of ε. As a consequence of both the separation of scales and the periodicity, all physical quantities can be looked for in the form of asymptotic expansions in powers of ε (Bensoussan et al., 1978):

$$\varphi^*(\mathbf{y}, \mathbf{x}) = \varphi^{(0)}(\mathbf{y}, \mathbf{x}) + \varepsilon^1 \varphi^{(1)}(\mathbf{y}, \mathbf{x}) + \varepsilon^2 \varphi^{(2)}(\mathbf{y}, \mathbf{x}) + ... \tag{2.4}$$

where the $\varphi^{(i)}$'s are $\mathbf{y}$ periodic and $\varphi^{(0)}$ is $\mathcal{O}(1)$,
5. solving the successive local boundary-value problems that are obtained after introducing these expansions in the local dimensionless description, and equating like powers of ε,
6. obtaining the macroscopic equivalent model from compatibility conditions which are the sufficient and necessary conditions for the existence of solutions to the successive local boundary-value problems. At some given order of ε, a local balance equation will yield an equation of the form

$$\nabla_y \cdot \mathbf{\Phi}^{(i+1)} + \nabla_x \cdot \mathbf{\Phi}^{(i)} = F^*, \tag{2.5}$$

where $\mathbf{\Phi}^{(i+1)}$ and $\mathbf{\Phi}^{(i)}$ are $\mathbf{y}$-periodic tensors and F^* is a source term. For simplicity, we assume here that (2.5) is valid over all Ω^* in the distribution sense. Other situations are possible, see, e.g., balance (3.22) below. This latter equation will lead to the macroscopic description. It actually expresses the balance at the $\mathbf{y}$ scale of the periodic quantity $\mathbf{\Phi}^{(i+1)}$, in presence of the source term $S^* = F^* - \nabla_x . \mathbf{\Phi}^{(i)}$. Integrating it over the period gives what is called a "compatibility condition". This compatibility condition must be checked (Fredholm alternative), otherwise the original equation has no solution and the problem is not homogenizable. It is written as

$$< \nabla_x \cdot \mathbf{\Phi}^{(i)} >_{\Omega^*} - < F^* >_{\Omega^*} = 0, \tag{2.6}$$

where $< . >_{\Omega^*}$ denotes the volume average with respect to space variable $\mathbf{y}$ over the period and is defined by

$$< . >_{\Omega^*} = \frac{1}{\mid \Omega^* \mid} \int_{\Omega_y^*} . \, \mathrm{dV}^*, \tag{2.7}$$

and
7. checking if the aforementioned process yields results that verify the estimations of dimensionless numbers in point 3: if not, e.g. if $A^{*(0)} = 0$, then the system (medium + phenomena) under consideration is not homogenizable: there exists no equivalent macroscopic description. Of course, an averaging process is possible in such a case, but it yields pseudo-effective coefficients that depend on the boundary conditions of the macroscopic boundary value problem and on L, thus leading to scale effects. The macroscopic mathematical model loses its legitimacy. The homogeneised solution is valid in the bulk macroscopic volume. Near the macroscopic boundary, the periodicity is broken and boundary layers are introduced to match the homogeneised solution to the macroscopic boundary conditions. The thickness of these boundary layers is generally of approximately one period size for diffusion or elastic problems, Sanchez-Palencia (1987). Their investigation yields the homogeneised boundary condition to be used, see Ene and Sanchez-Palencia (1975) for an example. Boundary layers are not studied in the present lecture.

3 Darcy's Law

For a sufficiently low pore Reynolds number Re, the steady state flow of incompressible Newtonian fluids through a Galilean porous media with connected pores is described by the celebrated Darcy's law. The velocity $\mathbf{v}$ is linearly related to the gradient of pressure

$$v_i = -\frac{K_{ij}}{\mu}\frac{\partial p}{\partial X_j}, \tag{3.1}$$

where $\mathbf{K}$ is the permeability tensor and μ is the viscosity. Let us recover Darcy's law by upscaling the pore scale behaviour, see Ene and Sanchez-Palencia (1975), Sanchez-Palencia (1980).

We consider a periodic rigid porous matrix of period Ω saturated by an incompressible fluid. The pores are connected; they occupy Ω_F and the pore surface is denoted Γ, see Figure 1. The macroscopic characteristic length is L and the characteristic pore size is l. Characteristic lengths L and l are generally difficult to estimate *a priori*. We just assume for the moment a separation of scales

$$\frac{l}{L} = \varepsilon \ll 1. \tag{3.2}$$

Characteristic lengths L and l will be made precise later.

3.1 Pore Scale Description of the Flow

The flow is slow, laminar, permanent and isothermal: at the pore scale, the Reynolds number and the transient Reynolds number are assumed as small, $Re_l \leq \mathcal{O}(\varepsilon)$, $Rt_l \leq \mathcal{O}(\varepsilon)$ (Subscript l shows that the dimensionless numbers are calculated by using l as the characteristic length). Therefore, the fluid velocity $\boldsymbol{v}$ and the pressure p verify

$$\mu\frac{\partial^2 v_i}{\partial X_j \partial X_j} - \frac{\partial p}{\partial X_i} = 0, \tag{3.3}$$

$$\frac{\partial v_i}{\partial X_i} = 0 \qquad \text{in } \Omega_F, \tag{3.4}$$

$$v_i = 0 \qquad \text{on } \Gamma. \tag{3.5}$$

The above set is put in dimensionless form by introducing the dimensionless space variable $y_i = X_i/l$ and adequate characteristic quantities, v_c, p_c and μ_c : $\mathbf{v} = v_c\mathbf{v}^*$, $p = p_c p^*$, $\mu = \mu_c\mu^*$. The above set introduces the dimensionless number

$$\mathcal{Q}_l = \frac{|\frac{\partial p}{\partial X_i}|}{|\mu\frac{\partial^2 v_i}{\partial X_j \partial X_j}|} = \frac{l p_c}{\mu_c v_c}. \tag{3.6}$$

Number $\mathcal{Q}_l$ can be evaluated in function of ε by noticing that the flow at the pore level is caused by a macroscopic gradient of pressure which equilibrates the viscous force

$$\frac{p_c}{L} = \mathcal{O}(\frac{\mu_c v_c}{l^2}). \tag{3.7}$$

We deduce that $\mathcal{Q}_l = \mathcal{O}(\varepsilon^{-1})$. The dimensionless form of the set (3.3-3.5) is the following, with $\mathcal{Q}_l = \varepsilon^{-1}$

$$\varepsilon\mu^* \frac{\partial^2 v_i^*}{\partial y_j \partial y_j} - \frac{\partial p^*}{\partial y_i} = 0, \tag{3.8}$$

$$\frac{\partial v_i^*}{\partial y_i} = 0 \qquad \text{in } \Omega_F^*, \tag{3.9}$$

$$v_i^* = 0 \qquad \text{on } \Gamma^*. \tag{3.10}$$

3.2 Upscaling

By following the multiple scale expansion technique (Sanchez-Palencia, 1980), the velocity $\mathbf{v}^*$ and the pressure fluctuation p^* are looked for in the form of asymptotic expansions of powers of ε

$$\mathbf{v}^* = \mathbf{v}^{(0)}(\mathbf{x}, \mathbf{y}) + \varepsilon\, \mathbf{v}^{(1)}(\mathbf{x}, \mathbf{y}) + \varepsilon^2\, \mathbf{v}^{(2)}(\mathbf{x}, \mathbf{y}) + \ldots \tag{3.11}$$

$$p^* = p^{(0)}(\mathbf{x}, \mathbf{y}) + \varepsilon\, p^{(1)}(\mathbf{x}, \mathbf{y}) + \varepsilon^2\, p^{(2)}(\mathbf{x}, \mathbf{y}) + \ldots \tag{3.12}$$

Introducing these expansions in (3.8-3.10) gives by identification of the like powers of ε successive boundary value problems to be investigated. The first order approximation of the pressure verifies

$$\frac{\partial p^{(0)}}{\partial y_i} = 0, \qquad p^{(0)} = p^{(0)}(\mathbf{x}). \tag{3.13}$$

The first order approximation of the velocity $\mathbf{v}^{(0)}$ and the second order approximation of the pressure $p^{(1)}$ are determined by the following set

$$\mu^* \frac{\partial^2 v_i^{(0)}}{\partial y_j \partial y_j} - J_i - \frac{\partial p^{(1)}}{\partial y_i} = 0, \tag{3.14}$$

$$\frac{\partial v_i^{(0)}}{\partial y_i} = 0 \qquad \text{in } \Omega_F^*, \tag{3.15}$$

$$v_i^{(0)} = 0 \qquad \text{on } \Gamma^*, \tag{3.16}$$

where $\mathbf{v}^{(0)}$ and $p^{(1)}$ are Ω^*-periodic. Vector $J_i = \partial p^{(0)}/\partial x_i$ is the macroscopic pressure gradient that causes the flow. To investigate this boundary value problem, it is convenient to introduce the space $\mathcal{V}$ of periodic divergence free vectors, defined on Ω^*_F, null on Γ^*, with the scalar product:

$$(\mathbf{u}, \mathbf{v})_{\mathcal{V}} = \int_{\Omega^*_F} \mu^* \frac{\partial u_i}{\partial y_j} \frac{\partial v_i}{\partial y_j} \, \mathrm{dV}^*, \tag{3.17}$$

which satisfies the following symmetry

$$(\mathbf{u}, \mathbf{v})_{\mathcal{V}} = (\mathbf{v}, \mathbf{u})_{\mathcal{V}}. \tag{3.18}$$

Now, multiply the two members of (3.14) by $\mathbf{u} \in \mathcal{V}$ and integrate over Ω^*_F. By using integration by parts, the divergence theorem, the periodicity and the boundary condition on Γ^*, we obtain

$$\forall\ \mathbf{u} \in \mathcal{V}, \qquad (\mathbf{u}, \mathbf{v}^{(0)})_{\mathcal{V}} = -(\mathbf{u}, \mathbf{J})_{\mathrm{L}^2}, \tag{3.19}$$

It gives $\mathbf{v}^{(0)}$ as a linear vectorial function of $\mathbf{J}$

$$v_i^{(0)} = -\lambda^*_{ij} J_j, \tag{3.20}$$

where $\boldsymbol{\lambda}^*(\mathbf{y})$ is the pore scale conductivity tensor field. When the viscosity μ is a constant, this relation can be put in the form

$$v_i^{(0)} = -\frac{k^*_{ij}}{\mu^*} J_j, \tag{3.21}$$

where $\mathbf{k}^*(\mathbf{y})$ is the pore scale permeability tensor field.

Then the volume balance of $\mathbf{v}^{(1)}$

$$\frac{\partial v_i^{(1)}}{\partial y_i} + \frac{\partial v_i^{(0)}}{\partial x_i} = 0, \tag{3.22}$$

gives by averaging a compatibility condition which stands for the macroscopic volume balance and introduces the Darcy's law

$$\frac{\partial \langle v_i^{(0)} \rangle}{\partial x_i} = 0, \quad \langle v_i^{(0)} \rangle = -\Lambda^*_{ij} J_j = -\frac{K^*_{ij}}{\mu^*} J_j, \quad \mathbf{K}^* = \langle \mathbf{k}^* \rangle = \frac{1}{|\Omega^*|} \int_{\Omega^*_F} \mathbf{k}^* \, \mathrm{dV}^*, \tag{3.23}$$

where $\boldsymbol{\Lambda}^*$ and $\mathbf{K}^*$ are the macroscopic dimensionless conductivity and permeability tensors, respectively.

The vector $\mathbf{k}^*_{.j}$ is solution of the following variational form

$$\forall\ \mathbf{u} \in \mathcal{V}, \qquad (\mathbf{u}, \mathbf{k}^*_{.j})_{\mathcal{V}} = \mu^* (\mathbf{u}, \mathbf{I}_{.j})_{\mathrm{L}^2}, \tag{3.24}$$

where $\mathbf{I}$ is the identity matrix.

3.3 Macroscopic Flow Model

By returning to dimensional quantities, relations (3.23) become

$$\frac{\partial\langle v_i\rangle}{\partial X_i} = \mathcal{O}(\varepsilon\frac{\partial\langle v_i\rangle}{\partial X_i}), \qquad \langle v_i\rangle = -\frac{K_{ij}}{\mu}\frac{\partial p}{\partial X_j} + \mathcal{O}(\varepsilon\langle v_i\rangle), \qquad \mathbf{K} = l^2\mathbf{K}^*. \qquad (3.25)$$

Remark that the macroscopic model is defined within a relative error $\mathcal{O}(\varepsilon)$. By using formulation (3.24), it is possible to show that tensor $\mathbf{K}$ is positive and symmetrical, (Sanchez-Palencia, 1980), see below.

Volume averaged velocity is a flux. It is important to check the physical meaning of the macroscopic quantities that are introduced by the upscaling process. Quantity p in (3.25) does not pose problem: it is directly defined as the first approximation of the local pore pressure, which first approximation is a constant over the period. Therefore, quantity p in (3.25) is a pressure. On an other hand $\langle\mathbf{v}\rangle$, which is a volume average can be shown to be a Darcy velocity, i.e. a flux. This is the consequence of the solenoidal character of $\mathbf{v}^{(0)}$. Let start from the identity :

$$\frac{\partial}{\partial y_k}(v_k^{(0)}y_i) = \frac{\partial v_k^{(0)}}{\partial y_k}y_i + v_k^{(0)}I_{ik}.$$

After integrating over Ω_F^*, using the divergence theorem and the adherence condition on Γ^*, we obtain:

$$\langle v_i^{(0)}\rangle = |\Omega^*|^{-1}\int_{\delta\Omega_F^*\cap\delta\Omega^*} v_k^{(0)}y_iN_k \text{ dS}^*,$$

where $\delta\Omega_F^*$ and $\delta\Omega^*$ are the boundaries of Ω_F^* and Ω^*, respectively, and N is the unit outward normal to Ω_F^*. Let l_i^* be the dimensionless length of the period along direction y_i and let Σ_i^* be the cross-section of the period at $y_i = l_i^*$, see Figure 2. Surface $\Sigma_{F_i}^*$ is the fluid part of Σ_i^* . Because of the Ω^*-periodicity of $v_k^{(0)}y_i$ in direction $y_j, j \neq i$, and because it cancels out at $y_i = 0$, we are left with :

$$< v_i^{(0)} >= |\Omega^*|^{-1}\int_{\Sigma_{F_i}^*} v_i^{(0)}l_i^* \text{ dS}^* = |\Sigma_i^*|^{-1}\int_{\Sigma_{F_i}^*} v_i^{(0)}\text{dS}^*,$$

(no summation on i). Therefore, $< \mathbf{v}^{(0)} >$ is a flux and $< \mathbf{v} >$ too at its first order of approximation.

Tensors $\mathbf{\Lambda}$ and $\mathbf{K}$ are symmetrical. Consider formulation (3.19) with on the one hand $\mathbf{v}^{(0)} = \boldsymbol{\lambda}_{.q}^*$, $\mathbf{u} = \boldsymbol{\lambda}_{.p}^*$ and on the other hand $\mathbf{v}^{(0)} = \boldsymbol{\lambda}_{.p}^*$, $\mathbf{u} = \boldsymbol{\lambda}_{.q}^*$. Due to the symmetry of the scalar product, we obtain

$$\int_{\Omega_F^*} \mu^*\frac{\partial\lambda_{ip}}{\partial y_j}\frac{\partial\lambda_{iq}^*}{\partial y_j}\text{ dV}^* = -\int_{\Omega_F^*}\lambda_{pq}^*\text{dV}^* = -\int_{\Omega_F^*}\lambda_{qp}^*\text{dV}^*.$$

Tensor $\mathbf{\Lambda}$ is symmetrical, and tensor $\mathbf{K}$ too.

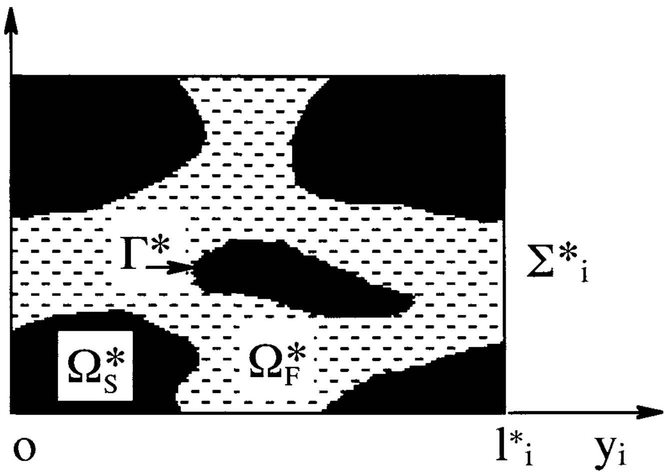

Figure 2. Period Ω^* of the porous medium, two-dimensional case

Tensors $\mathbf{\Lambda}$ and $\mathbf{K}$ are positive. Consider now (3.19) with $\mathbf{u} = \mathbf{v}^{(0)}$

$$|\Omega^*|^{-1} \int_{\Omega_F^*} \mu^* \frac{\partial v_i^{(0)}}{\partial y_j} \frac{\partial v_i^{(0)}}{\partial y_j} \, \mathrm{dV}^* = -|\Omega^*|^{-1} \int_{\Omega_F^*} v_i^{(0)} \frac{\partial p^{(0)}}{\partial x_i} \, \mathrm{dV}^* = \frac{\partial p^{(0)}}{\partial x_i} \Lambda_{ij}^* \frac{\partial p^{(0)}}{\partial x_j}.$$

The left hand member is positive. Then, the right hand member is positive and tensor $\mathbf{\Lambda}$ is positive, as well as tensor $\mathbf{K}$.

Evaluation of l and L. It is possible to *a posteriori* obtain an evaluation of l. It generally depends on the flow direction $\mathbf{n}$. From the third relation in (3.25) we have

$$l_{\mathbf{n}} \approx \sqrt{K_{ij} n_i n_j}, \tag{3.26}$$

which in the isotropic case reduces to

$$l \approx \sqrt{K}. \tag{3.27}$$

The macroscopic characteristic length L is either a characteristic size of the macroscopic boundary value problem either a length related to the flow itself. This later one can be *a posteriori* evaluated from the macroscopic pressure field

$$L \approx \frac{p}{|\nabla p|}. \tag{3.28}$$

A consequence of these estimations is that L generally depends on $\mathbf{X}$ if the porous medium is not macroscopically homogeneous. It results in a dependence of ε on $\mathbf{X}$, $\varepsilon = \varepsilon(\mathbf{X})$.

3.4 Domain of validity of the model

For permanent and quasi-static slow flow. physical considerations showed that $\mathcal{Q}_l = \mathcal{O}(\varepsilon^{-1})$, which is the domain of validity of the model (3.25). Let us have a look to other estimations of $\mathcal{Q}_l$.

Smaller estimations of $\mathcal{Q}_l$. Let $\mathcal{Q}_l$ be decreased to $\mathcal{Q}_l = \varepsilon^q$, $q \geq 0$, and let us explore the case $\mathcal{Q}_l = 1$. The dimensionless form of the set (3.3-3.5) is now in the form

$$\mu^* \frac{\partial^2 v_i^*}{\partial y_j \partial y_j} - \frac{\partial p^*}{\partial y_i} = 0, \tag{3.29}$$

$$\frac{\partial v_i^*}{\partial y_i} = 0 \qquad \text{in } \Omega_F^*, \tag{3.30}$$

$$v_i^* = 0 \qquad \text{on } \Gamma^*. \tag{3.31}$$

The first orders approximation of the velocity $\mathbf{v}^{(0)}$ and of the pressure $p^{(0)}$ are determined by the following set

$$\mu^* \frac{\partial^2 v_i^{(0)}}{\partial y_j \partial y_j} - \frac{\partial p^{(0)}}{\partial y_i} = 0, \tag{3.32}$$

$$\frac{\partial v_i^{(0)}}{\partial y_i} = 0 \qquad \text{in } \Omega_F^*, \tag{3.33}$$

$$v_i^{(0)} = 0 \qquad \text{on } \Gamma^*, \tag{3.34}$$

where $\mathbf{v}^{(0)}$ and $p^{(0)}$ are Ω^*-periodic. This system is similar to the system (3.14-3.16) after replacing $p^{(1)}$ by $p^{(0)}$ and letting $\mathbf{J} = \mathbf{0}$. Therefore, we obtain $\mathbf{v}^{(0)} = 0$, which is in a contradiction with $|\mathbf{v}^{(0)}| = \mathcal{O}(1)$. Estimation $\mathcal{Q}_l = 1$ does not correspond to a homogenizable situation. We also obtain $p^{(0)} = p^{(0)}(\mathbf{x})$. The next order yields a boundary value problem for $\mathbf{v}^{(1)}$ and $p^{(1)}$. It is easy to check that this problem is similar to system (3.14-3.16) after replacing $\mathbf{v}^{(0)}$ by $\mathbf{v}^{(1)}$. We then recover Darcy's flow

$$v_i^{(1)} = -\frac{k_{ij}^*}{\mu^*} J_j.$$

The homogenization process shifts $\mathcal{Q}_l = \mathcal{O}(1)$ to $\mathcal{Q}_l = \mathcal{O}(\varepsilon^{-1})$. Estimation $\mathcal{Q}_l = \varepsilon^q$, $q \geq 0$ yields $\mathbf{v}^{(i)} = 0$, $i \leq q+1$, which is also a non-homogenizable situation. All these estimations are of no physical meaning.

Higher estimations of $\mathcal{Q}_l$. If $\mathcal{Q}_l$ is increased by increasing the gradient of pressure, $\mathcal{Q}_l = \varepsilon^{-q}$, $q \geq 2$, we obtain also a non-homogenizable situation when assuming Stokes equation to be valid. In the case $\mathcal{Q}_l = \varepsilon^{-2}$, we have at the local scale

$$\varepsilon^2 \mu^* \frac{\partial^2 v_i^*}{\partial y_j \partial y_j} - \frac{\partial p^*}{\partial y_i} = 0,$$

$$\frac{\partial v_i^*}{\partial y_i} = 0 \qquad \text{in } \Omega_F^*,$$

$$v_i^* = 0 \qquad \text{on } \Gamma^*.$$

At the lower order we recover

$$\frac{\partial p^{(0)}}{\partial y_i} = 0, \qquad p^{(0)} = p^{(0)}(\mathbf{x}).$$

However, the second order gives

$$\frac{\partial p^{(0)}}{\partial x_i} + \frac{\partial p^{(1)}}{\partial y_i} = 0.$$

The periodicity of $p^{(1)}$ yields $p^{(0)}$ = constant, which is not admissible since $\partial p^{(0)}/\partial x_i$ should be $\mathcal{O}(1)$. We deduce that the estimation $\mathcal{Q}_l = \varepsilon^{-2}$ corresponds to a non-homogenizable situation. However, continuing the homogenization process gives the Darcy's flow at the next order. The homogenization process shifts $\mathcal{Q}_l = \mathcal{O}(\varepsilon^{-2})$ to $\mathcal{Q}_l = \mathcal{O}(\varepsilon^{-1})$. It is easy to show that estimations $\mathcal{Q}_l = \varepsilon^{-q}$, $q \geq 2$ are non-homogenizable.

Concluding remarks. The above results are resumed in Table 1. Obviously, the adopted pore scale behaviour (3.3-3.5) is not convenient to describe other numerous flows in porous media. When the gradient of pressure is increased, inertial non-linearities ap-

Table 1. Stokes' flow through rigid porous media.

$\mathcal{Q}_l = \mathcal{O}(\varepsilon^q)$, $q \geq 0$	$\mathcal{Q}_l = \mathcal{O}(\varepsilon^{-1})$	$\mathcal{Q}_l = \mathcal{O}(\varepsilon^{-q})$, $q \geq 2$
Non-homogenizable	Darcy's law	Non-homogenizable

pear. Deviation to Darcy's law in this case was investigated theoretically by Wodie and Levy (1991) and Mei and Auriault (1991), experimentally in Rasoloarijaona and Auriault (1994), Skjetne and Auriault (1999a), and numerically by Firdaouss et al. (1997). For small Reynolds numbers the obtained flow law is cubic with respect of the seepage velocity. For compressible fluids, the local balance equation (3.4) is modified. Darcy's law remains valid, but the macroscopic volume balance becomes non-linear, Auriault et al. (1990). in the case of low pressure gas flow, wall-slip appears on the pore surface, that yields Klinkenberg's law, Skjetne and Auriault (1999b), Chastanet et al. (2004). Transient flows were also investigated by homogenization in Levy (1979), Auriault (1980) and Burridge and Keller (1981), in which the phenomenological law previously introduced by Biot (1941) is recovered. Let us mention also non-Newtonian fluid flow in Auriault et al. (2002a) and flow in non-Galilean porous matrix, Auriault et al. (2000), Auriault et al. (2002b). All these studies concern single porosity porous media. Double or multiple porosity media introduce more than two separated scales, that generally strongly modifies the macroscopic behaviour, see e.g. Auriault and Boutin (1992, 1993).

4 Immiscible Two-phase Flow in Porous Media

The aim of this part is to investigate the governing equations that describe the flow of two immiscible fluids through a Galilean rigid porous medium (Auriault and Sanchez-Palencia, 1986; Auriault, 1987). The two fluids are shown by subscripts 1 and 2, respectively. To simplify the analysis we assume that

- A separation of scales is present, $l/L = \varepsilon \ll 1$, see the previous part for the definition of the two lengths l and L.

- The porous medium is rigid and Galilean and the two fluids are viscous Newtonian and incompressible.

- The two viscosities are of the same order of magnitude with respect to ε.

- As in the previous part, flows are slow (inertia is negligible), isothermal and time variations are neglected in Navier-Stokes equations.

- Each volume Ω_1 and Ω_2 occupied by each fluid is connected. In such flows, the interface Γ_{12} is generally moving. We assume this movement to be slower than the flow itself: if $\mathbf{n}$ is a unit normal to Γ_{12}, the fluid velocity on this surface verifies $\mathbf{v}.\mathbf{n} = \mathcal{O}(\varepsilon|\mathbf{v}_1|) = \mathcal{O}(\varepsilon|\mathbf{v}_2|)$. This assumption is coherent with equation (4.12) below. Integrating (4.12) over Ω^*_α, applying the divergence theorem and considering the periodicity of $\mathbf{v}^{(0)}_\alpha$ yields

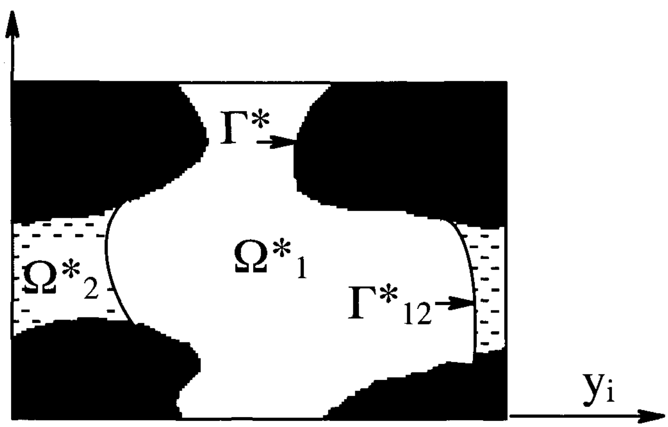

Figure 3. Period Ω^* of the non-saturated porous medium, cross section

$$\int_{\Gamma^*_{12}} \mathbf{v}^{(0)}_\alpha.\mathbf{n}\ \mathrm{dS}^* = 0.$$

- The Weber number $\mathcal{W}$ is $\mathcal{O}(1)$: the capillary pressure p_{ca} is of the same order of magnitude as the pressure in the two fluids. Laplace equation is written

$$p_{ca} = \sigma(\frac{1}{R_1} + \frac{1}{R_2}), \tag{4.1}$$

where σ is the surface tension assumed here as a constant and the two principal radii of curvature R_1 and R_2 are of the order of magnitude of the characteristic length l of the pores. Therefore, we have

$$\mathcal{W} = p_c l \sigma^{-1} = \mathcal{O}(1). \tag{4.2}$$

- There is no mass transfer between the two fluids.

4.1 Pore Scale Description.

By following the same route as for the one fluid flow, it can be shown that

$$\mathcal{Q}_{1_l} = \mathcal{O}(\mathcal{Q}_{2_l}) = \mathcal{O}(\varepsilon^{-1}). \tag{4.3}$$

Therefore, with the above assumptions, the dimensionless local description is in the form

$$\varepsilon\mu_\alpha^* \frac{\partial^2 v_{\alpha i}^*}{\partial y_j \partial y_j} - \frac{\partial p_\alpha^*}{\partial y_i} = 0, \tag{4.4}$$

$$\frac{\partial v_{\alpha i}^*}{\partial y_i} = 0 \qquad \text{in } \Omega_\alpha^*, \tag{4.5}$$

$$v_{\alpha i}^* = 0 \qquad \text{on } \Gamma^*, \tag{4.6}$$

where $\alpha = 1, 2$ and on Γ_{12}^*

$$(\sigma_{2_{ij}}^* - \sigma_{1_{ij}}^*) n_j = p_{ca}^* n_i, \tag{4.7}$$

where the $\sigma_{\alpha_{ij}}^* = -p_\alpha^* I_{ij} + \varepsilon\mu^*(\frac{\partial v_{\alpha_i}^*}{\partial y_j} + \frac{\partial v_{\alpha_j}^*}{\partial y_i})$ are the stress components, and

$$v_{1_i}^* = v_{2_i}^*, \qquad v_{\alpha_i}^* n_i = \mathcal{O}(\varepsilon|\mathbf{v}_\alpha^*|). \tag{4.8}$$

4.2 Upscaling.

After introducing *ad hoc* asymptotic expansions in the above description, and identifying like powers of ε, we obtain from (4.4) at the first order

$$\frac{\partial p_\alpha^{(0)}}{\partial y_i} = 0, \qquad p_\alpha^{(0)} = p_\alpha^{(0)}(\mathbf{x}), \tag{4.9}$$

and from (4.7)

$$p_{ca}^{(0)} = p_2^{(0)} - p_1^{(0)} = p_{ca}^{(0)}(\mathbf{x}). \tag{4.10}$$

The first approximation of the capillary pressure is a constant over the period, as well as the first approximation of the mean curvature of the surface Γ_{12}^*.

At the following order, we obtain a boundary value problem for $\mathbf{v}_\alpha^{(0)}$ and $p_\alpha^{(1)}$, $\alpha = 1, 2$

$$\mu_\alpha^* \frac{\partial^2 v_{\alpha i}^{(0)}}{\partial y_j \partial y_j} - \frac{\partial p_\alpha^{(1)}}{\partial y_i} - \frac{\partial p_\alpha^{(0)}}{\partial x_i} = 0, \tag{4.11}$$

$$\frac{\partial v_{\alpha i}^{(0)}}{\partial y_i} = 0 \qquad \text{in } \Omega_\alpha^*, \tag{4.12}$$

$$v_{\alpha i}^{(0)} = 0 \qquad \text{on } \Gamma^*, \tag{4.13}$$

and on Γ_{12}^*

$$(\sigma_{2_{ij}}^{(1)} - \sigma_{1_{ij}}^{(1)})n_j = p_{ca}^{(1)} n_i, \tag{4.14}$$

$$v_{1_i}^{(0)} = v_{2_i}^{(0)}, \qquad v_{\alpha_i}^{(0)} n_i = 0, \tag{4.15}$$

and where $\mathbf{v}_\alpha^{(0)}$ and $p_\alpha^{(1)}$ are Ω^*-periodic. To investigate the above boundary value problem, let us introduce the space $\mathcal{V}_1$ of Ω^*-periodic vectors, defined over $\Omega_F^* = \Omega_1^* + \Omega_2^*$, with zero divergence, zero valued on Γ^*, continuous on Γ_{12}^*, with zero flux through Γ_{12}^*, and with the scalar product

$$(\mathbf{u}, \mathbf{v})_{\mathcal{V}_1} = \int_{\Omega_F^*} \mu_\alpha^* \frac{\partial u_i}{\partial y_j} \frac{\partial v_i}{\partial y_j} \, \mathrm{dV}^*. \tag{4.16}$$

Multiply the two members of equation (4.11) by $\mathbf{u} \in \mathcal{V}_1$ and integrate over Ω_F^*. Since this equation can be written

$$\frac{\partial \sigma_{\alpha_{ij}}^{(1)}}{\partial y_j} = -\frac{\partial p_\alpha^{(0)}}{\partial x_i},$$

we have in Ω_1^*, after using the periodicity, the adherence condition on Γ^*, and the incompressibility at the first order

$$\int_{\Omega_1^*} u_i \frac{\partial \sigma_{1_{ij}}^{(1)}}{\partial y_j} \, \mathrm{dV}^* = -\int_{\Omega_1^*} u_i \frac{\partial p_1^{(0)}}{\partial x_i} \, \mathrm{dV}^* = \int_{\Omega_1^*} \left(\frac{\partial u_i \sigma_{1_{ij}}^{(1)}}{\partial y_j} - \sigma_{1_{ij}}^{(1)} \frac{\partial u_i}{\partial y_j}\right) \mathrm{dV}^*$$

$$= \int_{\Gamma_{12}^*} u_i \sigma_{1_{ij}}^{(1)} N_{1_j} \, \mathrm{dS}^* - \int_{\Omega_1^*} \mu_1^* \frac{\partial u_i}{\partial y_j} \frac{\partial v_{1_i}^{(0)}}{\partial y_j} \, \mathrm{dV}^*,$$

where $\mathbf{N}_1$ is the outward unit normal to Ω_1^*. In the same way, we obtain in Ω_2^*

$$\int_{\Omega_2^*} u_i \frac{\partial \sigma_{2_{ij}}^{(1)}}{\partial y_j}\,\mathrm{dV}^* = -\int_{\Gamma_{12}^*} u_i \sigma_{2_{ij}}^{(1)} N_{1_j} \mathrm{dS}^* - \int_{\Omega_2^*} \mu_2^* \frac{\partial u_i}{\partial y_j}\frac{\partial v_{2_i}^{(0)}}{\partial y_j}\,\mathrm{dV}^*.$$

Finally, adding member to member the two last equalities, and noticing that, because of $(4.15)_2$

$$\int_{\Gamma_{12}^*} u_i(\sigma_{1_{ij}}^{(1)} - \sigma_{2_{ij}}^{(1)}) N_{1_j} \mathrm{dS}^* = -\int_{\Gamma_{12}^*} u_i p_{ca}^{(1)} N_{1_i} \mathrm{dS}^* = 0,$$

give the equivalent variational form

$$\forall\, \mathbf{u} \in \mathcal{V}_1, \qquad \int_{\Omega_F^*} \mu_\alpha^* \frac{\partial u_i}{\partial y_j}\frac{\partial v_{\alpha i}^{(0)}}{\partial y_j}\,\mathrm{dV}^* = \int_{\Omega_F^*} u_i \frac{\partial p_\alpha^{(0)}}{\partial x_i}\,\mathrm{dV}^*. \tag{4.17}$$

This formulation ensures the uniqueness of $\mathbf{v}^{(0)}$. The problem being linear, let us introduce particular solutions. Let

$$\boldsymbol{\lambda}_1^{*j}(\mathbf{y}) = \begin{cases} \boldsymbol{\lambda}_{11.j}^* & \text{in } \Omega_1^* \\ \boldsymbol{\lambda}_{12.j}^* & \text{in } \Omega_2^* \end{cases}$$

be the solution for

$$\frac{\partial p_1^{(0)}}{\partial x_i} = \delta_{ij}, \qquad \frac{\partial p_2^{(0)}}{\partial x_i} = 0,$$

and

$$\boldsymbol{\lambda}_2^{*j}(\mathbf{y}) = \begin{cases} \boldsymbol{\lambda}_{21.j}^* & \text{in } \Omega_1^* \\ \boldsymbol{\lambda}_{22.j}^* & \text{in } \Omega_2^* \end{cases}$$

be the solution for

$$\frac{\partial p_1^{(0)}}{\partial x_i} = 0, \qquad \frac{\partial p_2^{(0)}}{\partial x_i} = \delta_{ij}.$$

Then the general solution is in the form

$$v_{1_i}^{(0)} = -\lambda_{11_{ij}}^* \frac{\partial p_1^{(0)}}{\partial x_j} - \lambda_{12_{ij}}^* \frac{\partial p_2^{(0)}}{\partial x_j}, \tag{4.18}$$

$$v_{2_i}^{(0)} = -\lambda_{21_{ij}}^* \frac{\partial p_1^{(0)}}{\partial x_j} - \lambda_{22_{ij}}^* \frac{\partial p_2^{(0)}}{\partial x_j}, \tag{4.19}$$

The volume balance gives at the second order

$$\frac{\partial v_{\alpha i}^{(1)}}{\partial y_i} + \frac{\partial v_{\alpha i}^{(0)}}{\partial x_i} = 0. \tag{4.20}$$

Each equation for $\alpha = 1, 2$, yields a compatibility equation which is obtained by volume averaging

$$\frac{\partial \langle v_{1_i}^{(0)} \rangle}{\partial x_i} = -|\Omega^*|^{-1} \int_{\Gamma_{12}^*} v_{1_i}^{(1)} N_{1_i} \mathrm{dS}^* = \phi \frac{\partial S_1}{\partial t^*}, \tag{4.21}$$

$$\frac{\partial \langle v_{2_i}^{(0)} \rangle}{\partial x_i} = -|\Omega^*|^{-1} \int_{\Gamma_{12}^*} v_{2_i}^{(1)} N_{2_i} \mathrm{dS}^* = \phi \frac{\partial S_2}{\partial t^*}, \tag{4.22}$$

where ϕ is the porosity and S_α is the saturation of fluid α, with $S_1 + S_2 = 1$. The macroscopic volume balances (4.21) and (4.22) yield a generalized flow law in the form

$$\langle v_{1_i}^{(0)} \rangle = -\Lambda_{11_{ij}}^* \frac{\partial p_1^{(0)}}{\partial x_j} - \Lambda_{12_{ij}}^* \frac{\partial p_2^{(0)}}{\partial x_j}, \tag{4.23}$$

$$\langle v_{2_i}^{(0)} \rangle = -\Lambda_{21_{ij}}^* \frac{\partial p_1^{(0)}}{\partial x_j} - \Lambda_{22_{ij}}^* \frac{\partial p_2^{(0)}}{\partial x_j}, \tag{4.24}$$

where

$$\mathbf{\Lambda}_{\alpha\beta}^* = \langle \boldsymbol{\lambda}_{\alpha\beta}^* \rangle = \frac{1}{|\Omega^*|} \int_{\Omega_\alpha^*} \boldsymbol{\lambda}_{\alpha\beta}^* \, \mathrm{dV}^*.$$

4.3 Macroscopic Two-phase Flow Model

By returning to dimensional quantities, relations (4.23) and (4.24) become

$$\frac{\partial \langle v_{\alpha i} \rangle}{\partial X_i} = -\phi \frac{\partial S_\alpha}{\partial t} + \mathcal{O}(\varepsilon \frac{\partial \langle v_{\alpha i} \rangle}{\partial X_i}), \quad \langle v_{\alpha i} \rangle = -\frac{\Lambda_{\alpha\beta_{ij}}}{\mu} \frac{\partial p_\beta}{\partial X_j} + \mathcal{O}(\varepsilon \langle v_{\alpha i} \rangle), \tag{4.25}$$

where

$$\mathbf{\Lambda}_{\alpha\beta} = \frac{l^2}{\mu_c} \mathbf{\Lambda}_{\alpha\beta}^*, \qquad \alpha = 1, 2, \tag{4.26}$$

are conductivity tensors. The macroscopic problem is closed by giving the relation between the capillary pressure p_{ca} and one of the saturations, e.g., S_1.
Remark that, on the contrary to one fluid flow, permeabilities cannot be introduced unless in the very restrictive case where $\mu_1 = \mu_2 =$ constant. Volume average velocities $\mathbf{v}_\alpha^{(0)}$ are shown to be fluxes by following the same route as for the one fluid flow. It is also possible to show (Auriault, 1987) that the conductivities verifies the symmetries

$$\Lambda_{\alpha\alpha_{ij}} = \Lambda_{\alpha\alpha_{ji}}, \qquad \Lambda_{\alpha\beta_{ij}} = \Lambda_{\beta\alpha_{ji}}. \tag{4.27}$$

In practical applications, the coupling terms are often neglected: $\Lambda_{\alpha\beta_{ij}} = 0$ when $\alpha \neq \beta$, see Zarcone and Lenormand (1994) for an experimental checking. That can be obtained from the above analysis by letting negligible the interface between the two fluids: $|\Gamma_{12}| \approx 0$.

5 Diffusion/Convection in Porous Media

We consider again a rigid Galilean porous matrix with connected pores. The pores are saturated by a viscous Newtonian incompressible fluid which contains a solute at low concentration. The solute is transported in the pores by diffusion and advection. Temperature is constant. We neglect adsorption on the pore surface Γ and absorption in the solid Ω_S.
It is convenient to introduce (Auriault and Adler, 1995), characteristic times associated with advection and diffusion, respectively: $T_l^{adv} = l/v_c$, $T_l^{diff} = l^2/D_c$ at scale l and $T_L^{adv} = L/v_c$, $T_L^{diff} = L^2/D_c$ at scale L, where D_c and v_c are characteristic values of the molecular diffusion tensor and the fluid velocity, respectively. Their ratio is equal to the Péclet number $\mathcal{P}e$, which is also the ratio of the advection term to the diffusion term in equation (5.1) below

$$\mathcal{P}e_l = \frac{T_l^{diff}}{T_l^{adv}}, \qquad \mathcal{P}e_L = \frac{T_L^{diff}}{T_L^{adv}}.$$

The whole study can be done by comparing $\mathcal{P}e$ to the ratio ε between the microscopic and the macroscopic space scales of the medium. Four regimes can be distinguished. When the local Péclet number $\mathcal{P}e_l = v_c l/D_c = T_l^{diff}/T_l^{adv}$ is of order ε^2 (macroscopic Péclet number $\mathcal{P}e_L = v_c L/D_c = T_L^{diff}/T_L^{adv} = \mathcal{O}(\varepsilon)$), diffusion is predominant at the large and small scales; convection does not appear at the macroscopic scale at the first order of approximation, and one obtains a macroscopic diffusion equation with a macroscopic diffusion tensor. When the local Péclet number $\mathcal{P}e_l$ is of order ε, $\mathcal{P}e_L = \mathcal{O}(1)$, diffusion is still predominant at the small scale, but convection becomes of the same order of magnitude at the large scale. When the local Péclet number $\mathcal{P}e_l$ is equal to 1, $\mathcal{P}e_L = \mathcal{O}(\varepsilon^{-1})$, the situation is more complex; convection and diffusion are equivalent at the small spatial scale, but convection is predominant for short times and large spatial scale. It is only at the second order of approximation that diffusion appears at the large scale in the form of dispersion. Finally, when the local Péclet number $\mathcal{P}e_l$ is assumed to be of the order of ε^{-1}, $\mathcal{P}e_L = \mathcal{O}(\varepsilon^{-2})$, convection is predominant everywhere and the concentration is constant along streamlines at the macroscopic scale. Hence, the concentration distribution depends mostly upon the concentration imposed at the external boundaries of the macroscopic volume. Because of this dependency, the problem cannot be homogenized anymore.

5.1 Formulation of the Local Problem

The physical processes of molecular diffusion and convection of the solute can be described by the following mass balance equation

$$\frac{\partial c}{\partial t} + \frac{\partial}{\partial X_i}(-D_{ij}\frac{\partial c}{\partial X_j} + v_i c) = 0 \qquad \text{in } \Omega_F, \tag{5.1}$$

$$D_{ij}\frac{\partial c}{\partial X_i}N_i = 0 \qquad \text{on } \Gamma, \tag{5.2}$$

where c is the concentration (mass of pollutant per unit volume of fluid), **D** is the molecular diffusion tensor which is generally an isotropic tensor, t is the time variable, **N** is the unit vector normal to Γ and **v** is the fluid velocity. Velocity **v** is investigated in the previous part 3 and we will make use of the analysis herein in the present study.

We aim at describing diffusion and advection in the porous medium at the macroscopic scale. Therefore, we use T_L^{diff} as the characteristic time when diffusion is predominant or equivalent to advection at macroscale to make dimensionless the above set of equations

$$t_c = T_L^{diff} = \frac{L^2}{D_c} \text{ when } \mathcal{P}e_l = \mathcal{O}(\varepsilon^2) \text{ or } \mathcal{P}e_l = \mathcal{O}(\varepsilon), \tag{5.3}$$

and we use $t_c = T_L^{adv}$ when advection is predominant

$$t_c = T_L^{adv} = \frac{L}{v_c} \text{ when } \mathcal{P}e_l = \mathcal{O}(1) \text{ or } \mathcal{P}e_l = \mathcal{O}(\varepsilon^{-1}). \tag{5.4}$$

The dimensionless form of the local description is as follows (l is the characteristic length in use)

$$\mathcal{P}_l\frac{\partial c^*}{\partial t^*} + \frac{\partial}{\partial y_i}(-D^*_{ij}\frac{\partial c^*}{\partial y_j} + \mathcal{P}e_l v^*_i c^*) = 0 \qquad \text{in } \Omega^*_F, \tag{5.5}$$

$$D^*_{ij}\frac{\partial c^*}{\partial y_j}N_i = 0 \qquad \text{on } \Gamma^*. \tag{5.6}$$

It introduces two dimensionless numbers

$$\mathcal{P}_l = \frac{|\frac{\partial c}{\partial t}|}{|\frac{\partial}{\partial X_i}(D_{ij}\frac{\partial c}{\partial X_j})|} = \frac{l^2}{D_c t_c}, \tag{5.7}$$

and the Péclet number is defined by

$$\mathcal{P}e_l = \frac{|\frac{\partial}{X_i}(v_i c)|}{|\frac{\partial}{\partial X_i}(D_{ij}\frac{\partial c}{\partial X_j})|} = \frac{v_c l}{D_c}. \tag{5.8}$$

In the following, we apply the homogenization process to the four above mentioned different cases of interest: predominant diffusion at macroscale, $t_c = T_L^{diff}$, $\mathcal{P}e_l = \varepsilon^2$, $\mathcal{P}_l = \varepsilon^2$; diffusion and advection equivalent at macroscale, $t_c = T_L^{diff} = T_L^{adv}$, $\mathcal{P}e_l = \varepsilon$, $\mathcal{P}_l = \varepsilon^2$; predominant advection at macroscale, $t_c = T_L^{adv}$, $\mathcal{P}e_l = 1$, $\mathcal{P}_l = \varepsilon$; strong advection, $t_c = T_L^{adv}$, $\mathcal{P}e_l = \varepsilon^{-1}$, $\mathcal{P}_l = 1$. We look for c^* in the form of an asymptotic expansion

$$c^* = c^{(0)}(\mathbf{x},\mathbf{y},t^*) + \varepsilon c^{(1)}(\mathbf{x},\mathbf{y},t^*) + \varepsilon^2 c^{(2)}(\mathbf{x},\mathbf{y},t^*) + \cdots, \tag{5.9}$$

where the components $c^{(i)}(\mathbf{x},\mathbf{y},t^*)$ are $\mathbf{y}$-periodic of period Ω^*, and where space variable $\mathbf{x}$ is expressed by $\mathbf{x} = \varepsilon\mathbf{y}$.

5.2 Predominant Diffusion at Macroscale

The observation time is now

$$t_c = T_L^{diff} = \varepsilon T_L^{adv},$$

which yields

$$\mathcal{P}e_l = \varepsilon^2, \qquad \mathcal{P}_l = \varepsilon^2.$$

The normalized equations (5.5) and (5.6) take the following form

$$\varepsilon^2 \frac{\partial c^*}{\partial t^*} + \frac{\partial}{\partial y_i}\left(-D_{ij}^* \frac{\partial c^*}{\partial y_j} + \varepsilon^2 v_i^* c^*\right) = 0 \qquad \text{in } \Omega_F^*, \tag{5.10}$$

$$D_{ij}^* \frac{\partial c^*}{\partial y_j} N_i = 0 \qquad \text{on } \Gamma^*. \tag{5.11}$$

Upscaling. After introducing expansion (5.9), we obtain at the lower order of ε

$$\frac{\partial}{\partial y_i}\left(D_{ij}^* \frac{\partial c^{(0)}}{\partial y_j}\right) = 0 \qquad \text{in } \Omega_F^*, \tag{5.12}$$

$$D_{ij}^* \frac{\partial c^{(0)}}{\partial y_j} N_i = 0 \qquad \text{on } \Gamma^*, \tag{5.13}$$

where $c^{(0)}$ is a periodic function of $\mathbf{y}$. Let us introduce the space $\mathcal{C}$ of periodic functions defined in Ω_F^*, of zero average over Ω_F^* (an extraneous condition just introduced to insure the Hilbertian character of the space), with the scalar product

$$(\alpha,\beta)_{\mathcal{C}} = \int_{\Omega_F^*} D_{ij}^* \frac{\partial \alpha}{\partial y_i}\frac{\partial \beta}{\partial y_j}\, \mathrm{dV}^*.$$

Multiplying the two sides of (5.12) by $\alpha \in \mathcal{C}$, and integrating over Ω_F^* yield after some analysis to the following weak formulation

$$\forall \alpha \in \mathcal{C}, \qquad \int_{\Omega_F^*} D_{ij}^* \frac{\partial \alpha}{\partial y_i}\frac{\partial c^{(0)}}{\partial y_j}\, \mathrm{dV}^* = 0. \tag{5.14}$$

This formulation insures the existence and uniqueness of $c^{(0)} \in \mathcal{C}$. That yields to the trivial solution of (5.16)-(5.17)

$$c^{(0)} = c^{(0)}(\mathbf{x}, t^*). \tag{5.15}$$

The first order concentration is a constant over the period Ω^*. After taking into account this result, the second order term $c^{(1)}$ is the solution of the set

$$\frac{\partial}{\partial y_i}(D^*_{ij}(\frac{\partial c^{(0)}}{\partial x_j} + \frac{\partial c^{(1)}}{\partial y_j})) = 0 \qquad \text{in } \Omega^*_F, \tag{5.16}$$

$$D^*_{ij}(\frac{\partial c^{(0)}}{\partial x_j} + \frac{\partial c^{(1)}}{\partial y_j})N_i = 0 \qquad \text{on } \Gamma^*, \tag{5.17}$$

where $c^{(1)}$ is a periodic function of $\mathbf{y}$. Multiplying the two sides of (5.16) by $\alpha \in \mathcal{C}$, and integrating over Ω^*_F yield to the following weak formulation

$$\forall \alpha \in \mathcal{C}, \qquad \int_{\Omega^*_F} D^*_{ij} \frac{\partial \alpha}{\partial y_i}(\frac{\partial c^{(0)}}{\partial x_j} + \frac{\partial c^{(1)}}{\partial y_j}) \, \mathrm{dV}^* = 0. \tag{5.18}$$

This formulation insures the existence and uniqueness of $c^{(1)} \in \mathcal{C}$, that gives the solution of the linear problem (5.16)-(5.17) in the form

$$c^{(1)} = \chi_i^{*+} \frac{\partial c^{(0)}}{\partial x_i} + \bar{c}^{(1)}(\mathbf{x}, t^*), \tag{5.19}$$

where $\boldsymbol{\chi}^{*+}$ is a periodic vector, of zero average which component χ_k^{*+} verifies

$$\forall \alpha \in \mathcal{C}, \qquad \int_{\Omega^*_F} D^*_{ij} \frac{\partial \alpha}{\partial y_i}(I_{jk} + \frac{\partial \chi_k^{*+}}{\partial y_j}) \, \mathrm{dV}^* = 0. \tag{5.20}$$

Function $\bar{c}^{(1)}$ is an arbitrary function of $\mathbf{x}$ and t^* which is introduced due to the extraneous condition $< \boldsymbol{\chi}^{*+} >_{\Omega^*_\Gamma} = 0$. Finally, the boundary value problem for $c^{(2)}$ reduces to, after introducing the above result for $c^{(0)}$ (the term $\partial/\partial y_i(v^{(0)}c^{(0)})$ cancels out)

$$\frac{\partial c^{(0)}}{\partial t^*} - \frac{\partial}{\partial y_i}(D^*_{ij}(\frac{\partial c^{(1)}}{\partial x_j} + \frac{\partial c^{(2)}}{\partial y_j})) - \frac{\partial}{\partial x_i}(D^*_{ij}(\frac{\partial c^{(0)}}{\partial x_j} + \frac{\partial c^{(1)}}{\partial y_j})) = 0 \qquad \text{in } \Omega^*_F, \tag{5.21}$$

$$D^*_{ij}(\frac{\partial c^{(1)}}{\partial x_j} + \frac{\partial c^{(2)}}{\partial y_j})N_i = 0 \qquad \text{on } \Gamma^*, \tag{5.22}$$

where $c^{(2)}$ is $\mathbf{y}$-periodic. On the contrary to the boundary value problems for $c^{(0)}$ and $c^{(1)}$, the boundary value problem for $c^{(2)}$ yields a compatibility condition which is obtain by integrating (5.21) over Ω^*

$$\phi \frac{\partial c^{(0)}}{\partial t^*} - \frac{\partial}{\partial x_i}(D^{*+}_{ij} \frac{\partial c^{(0)}}{\partial x_j}) = 0, \tag{5.23}$$

where ϕ is the porosity and where the effective dimensionless diffusion tensor $\mathbf{D}^{*+}$ is defined by

$$D_{ij}^{*+} = \frac{1}{|\Omega^*|} \int_{\Omega_F^*} D_{ik}^* (I_{jk} + \frac{\partial \chi_j^{*+}}{\partial y_k}) \, \mathrm{dV}^*. \tag{5.24}$$

Macroscopic diffusion model. By returning to physical quantities, we obtain

$$\phi \frac{\partial c}{\partial t} - \frac{\partial}{\partial X_i}(D_{ij}^+ \frac{\partial c}{\partial X_j}) = \mathcal{O}(\varepsilon \phi \frac{\partial c}{\partial t}), \qquad \mathbf{D}^+ = D_c \, \mathbf{D}^{*+}, \tag{5.25}$$

which relative accuracy is $\mathcal{O}(\varepsilon)$. Tensor $\mathbf{D}^+$ is a purely diffusive effective tensor. By following a similar route as for investigating the properties of the permeability $\mathbf{K}$ in part 3, it can be shown from formulation (5.18) that tensor $\mathbf{D}^+$ is positive and symmetrical.

5.3 Diffusion and Advection Equivalent at Macroscale

The observation time is now

$$t_c = T_L^{diff} = T_L^{adv},$$

which gives the following dimensionless numbers

$$\mathcal{P}e_l = \varepsilon, \qquad \mathcal{P}_l = \varepsilon^2.$$

The normalized equations (5.5) and (5.6) take now the following form

$$\varepsilon^2 \frac{\partial c^*}{\partial t^*} + \frac{\partial}{\partial y_i}(-D_{ij}^* \frac{\partial c^*}{\partial y_j} + \varepsilon v_i^* c^*) = 0 \qquad \text{in } \Omega_F^*, \tag{5.26}$$

$$D_{ij}^* \frac{\partial c^*}{\partial y_j} N_i = 0 \qquad \text{on } \Gamma^*. \tag{5.27}$$

Upscaling. After introducing expansion (5.9) in the above set, it easy to show that the boundary value problems for $c^{(0)}$ and $c^{(1)}$ are unchanged. Therefore results (5.15) and (5.19) are still valid

$$c^{(0)} = c^{(0)}(\mathbf{x}, t^*), \qquad c^{(1)} = \chi_i^{*+} \frac{\partial c^{(0)}}{\partial x_i} + \bar{c}^{(1)}(\mathbf{x}, t^*). \tag{5.28}$$

However, the boundary value problem for $c^{(2)}$ is changed into

$$\frac{\partial c^{(0)}}{\partial t^*} - \frac{\partial}{\partial y_i}(D_{ij}^* (\frac{\partial c^{(1)}}{\partial x_j} + \frac{\partial c^{(2)}}{\partial y_j})) - \frac{\partial}{\partial x_i}(D_{ij}^* (\frac{\partial c^{(0)}}{\partial x_j} + \frac{\partial c^{(1)}}{\partial y_j}))$$

$$+ \frac{\partial}{\partial y_i}(c^{(0)} v_i^{(1)} + c^{(1)} v_i^{(0)}) + \frac{\partial}{\partial x_i}(c^{(0)} v_i^{(0)}) = 0 \quad \text{in } \Omega_F^*, \tag{5.29}$$

$$D^*_{ij}(\frac{\partial c^{(1)}}{\partial x_j} + \frac{\partial c^{(2)}}{\partial y_j})N_i = 0 \qquad \text{on } \Gamma^*, \tag{5.30}$$

where $c^{(2)}$ is $\mathbf{y}$-periodic. The compatibility condition is now in the form

$$\phi \frac{\partial c^{(0)}}{\partial t^*} - \frac{\partial}{\partial x_i}(D^{*+}_{ij} \frac{\partial c^{(0)}}{\partial x_j} - < v^{(0)} >_i c^{(0)}) = 0, \tag{5.31}$$

where $\mathbf{D}^{*+}$ is the effective diffusion tensor already defined in the previous case. The macroscopic velocity $< \mathbf{v}^{(0)} >$ was defined in part 3.

Macroscopic diffusion-advection model. After coming back to dimensional quantities, we have

$$\phi \frac{\partial c}{\partial t} - \frac{\partial}{\partial X_i}(D^+_{ij} \frac{\partial c}{\partial X_j} - < v >_i c) = \mathcal{O}(\varepsilon \phi \frac{\partial c}{\partial t}), \qquad \mathbf{D}^+ = D_c \, \mathbf{D}^{*+}, \tag{5.32}$$

which relative accuracy is $\mathcal{O}(\varepsilon)$. Tensor $\mathbf{D}^+$ is a purely diffusive tensor: it is not modified by the advection.

5.4 Predominant Advection at Macroscale

The observation time is now

$$t_c = T^{adv}_L = \varepsilon T^{diff}_L,$$

and we have

$$\mathcal{P}e_l = 1, \qquad \mathcal{P}_l = \varepsilon.$$

The normalized equations (5.5) and (5.6) take the following form

$$\varepsilon \frac{\partial c^*}{\partial t^*} + \frac{\partial}{\partial y_i}(-D^*_{ij} \frac{\partial c^*}{\partial y_j} + v^*_i c^*) = 0 \qquad \text{in } \Omega^*_F, \tag{5.33}$$

$$D^*_{ij} \frac{\partial c^*}{\partial y_j} N_i = 0 \qquad \text{on } \Gamma^*. \tag{5.34}$$

Upscaling. The boundary value problem for $c^{(0)}$ is now in the form

$$\frac{\partial}{\partial y_i}(D^*_{ij} \frac{\partial c^{(0)}}{\partial y_j} - v^{(0)}_i c^{(0)}) = 0 \qquad \text{in } \Omega^*_F, \tag{5.35}$$

$$D^*_{ij} \frac{\partial c^{(0)}}{\partial y_j} N_i = 0 \qquad \text{on } \Gamma^*, \tag{5.36}$$