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TRANSPORT PROCESSES IN POROUS MEDIA. 1. CONTINUOUS MODELING

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Abstract: A course grained space-time average of quantities assigned to the molecules of a corpuscular physical system is defined. It is shown that these averages are almost everywhere continuous space-time functions and they satisfy identities similar to the balance equations from continuum mechanics. Further, through averages over the statistical ensemble, everywhere continuous fields, and balance equations are derived. It is shown that a Lagrangian description of the transport by an advection-diffusion equation, can be obtained. In this frame, a macroscopic continuous model of motion in porous media is proposed, the Darcy-Buckingham flux law and the porosity dependent advection-diffusion equation are derived.

1 INTRODUCTION

The adequacy of the diffusion equation as a model of the transport in porous media is a very much commented question [Sposito et all., 1986]. The attempts to derive it in a stochastic framework, based on the equivalence between the Ito stochastic differential equation and deterministic Fokker-Planck equation, did not take into account the porosity. Moreover the validity of the effective diffusion equation at large scales is only an assertion, not a rigorous mathematical result [Suciu et all., 1996]. The methods based on homogenization or renormalization give good mathematical results but assume nonphysical hypotheses on the structure of the porous media or statistics of velocity. For instance, the Darcy law providing the filtration velocity is mathematically founded by homogenization method under periodicity assumptions on the porous media structure. [Sanchez-Palencia, 1980].

Sposito [1978] gives a statistical mechanical derivation of both balance equations and Darcy law. The approach is in the spirit of the statistical mechanics transport theory of Kirkwood [1967]. An additional space average over the pore space introduces continuous fields but the porous media structure is not explicitly taken into account. The utility of this theory is limited by the necessity of the knowledge of the probability density function which describes the microscopic dynamics of the porous media as a physical system.

None of the previously discussed methods give a continuous model of the porous media entirely based on a microscopic description. We try to fill this gap as it follows. We describe the porous media at the microscopic scale as a physical system of an arbitrary and finite number of molecules of the solid matrix and of the components of the fluid filling it. Using the method of Vamoş et all. [1996a,b] we derive continuous fields and balance equations and we propose a continuous model of the porous media.

In the section 2, a coarse-grained space-time average is defined. We prove that if the microscopic physical quantities are described by analytic functions then their coarse-grained averages are almost everywhere continuous and satisfy identities similar to macroscopic balance equations.

In the section 3, we describe the physical system by a stochastic process defined, in the sense of Doob, as random variable valued in the space of the trajectories of the constituent particles. The expectation of coarse-grained averages gives smooth continuous fields. This approach enables us to improve the insight of the classical statistical mechanics definition of the continuous fields [Kirkwood, 1967]: continuous fields are the limits, for small space-time scales, of the expectations of the coarse-grained space-time averages. By averages over the statistical ensemble of the identities derived in the previous section we obtain balance equations. In this way, the macroscopic balance equations can be derived for any microscopic quantities, not only for conservatives ones, as it is usual in statistical mechanics. So, using the balance equation corresponding to the positions of the microscopic particles, we can write the concentration balance equation in the advection-diffusion form. This result suggests developments in data analysis and numerical algorithms.

Applying this continuous modeling to porous media, in the section 4, we find an expression of the porosity similar to that introduced by Hilfer [1991]. We also derive the Darcy-Buckingham law and the porosity-dependent advection and diffusion equations.

2 COARSE GRAINED AVERAGES

We consider a classical mechanics system consisting of N molecules. The microscopic discrete description of this system is given by a set of analytic functions

$\varphi_i : I \longmapsto \mathbb{R}$, $I = [0,T] \subset \mathbb{R}$ $(1 \le i \le N)$. The α components of the corresponding position vectors $\mathbf{r}_i$, $x_{\alpha i} : I \longmapsto \mathbb{R}$ $(\alpha = 1,2,3)$, and of the velocities $\boldsymbol{\xi}_i$, $\xi_{\alpha i} : I \longmapsto \mathbb{R}$ $(\alpha = 1,2,3)$, can be treated as particular cases of functions φ_i.

We define the *coarse-grained average* of the physical quantity φ as a function $\langle\varphi\rangle : \mathbb{R}^3 \times (\tau, T-\tau) \longmapsto \mathbb{R}$,

$$\langle\varphi\rangle(\mathbf{r},t) \;=\; \frac{1}{2\tau\mathcal{V}} \sum_{i=1}^{N} \int\limits_{t-\tau}^{t+\tau} \varphi_i(t')\, H^+(a^2 - (\mathbf{r}_i(t') - \mathbf{r})^2)\, dt' \tag{2.1}$$

where $\tau < T/2$ and a are arbitrary positive real parameters, $\mathcal{V} = 4\pi a^3/3$ is the volume of the sphere $S(\mathbf{r},a)$ and H^+ is the left continuous Heaviside function. Since $H^+(a^2 - (\mathbf{r}_i(t') - \mathbf{r})^2)$ vanishes if the i-th particle is located outside the sphere $S(\mathbf{r},a)$, then a nonvanishing contribution to $\langle\varphi\rangle$ is due only to particles which lie in $S(\mathbf{r},a)$ over the interval $(t-\tau, t+\tau)$. This average characterizes the mean distribution of the physical quantity φ about the point of position $\mathbf{r}$ and about the moment t.

Proposition 1. *As a function of* $\mathbf{r}$ *and* t, $\langle\varphi\rangle$ *possesses partial derivatives a.e. continuous in* $\mathbb{R}^3 \times (\tau, T-\tau)$.

Proof: The function $H^+(a^2 - (\mathbf{r}_i(t') - \mathbf{r})^2)$ in (2.1) takes only the values 0 and 1. The jumps occur when the i-th molecule enters or leaves the open sphere $S(\mathbf{r},a)$. These moments are among the solutions u_i of the equation

$$h_i(\mathbf{r}, u_i) \;\equiv\; (\mathbf{r}_i(u_i) - \mathbf{r})^2 - a^2 \;=\; 0. \tag{2.2}$$

Here $|\, h_i(\mathbf{r},t) \,|^{1/2}$ is the distance, at the moment t, between the i-th molecule and the surface $\partial S(\mathbf{r},a)$ of the sphere $S(\mathbf{r},a)$. Since the functions $x_{\alpha i}$, and hence h_i, are analytic with respect to u_i, and taking into account that I is a closed interval, it follows that either equation (2.2) has a finite number of solutions or h_i vanishes identically [Sveshnikov and Tikhonov, 1978, p.78]. In the last case the molecule moves along the surface $\partial S(\mathbf{r},a)$ and does not enter the sphere, hence $H^+(a^2 - (\mathbf{r}_i(t) - \mathbf{r})^2)$ is identically zero and no jumps occur. Since $\mathbf{r}_i(u_i)$ is a known function, then the isolated zeros of (2.2) are implicit functions $u_i(\mathbf{r})$. The implicit function theorem can be applied only at interior points of the range of h_i, the same with the range of $\langle\varphi\rangle$. Therefore it does not ensure the existence of $u_i(\mathbf{r})$ for $u_i = \tau$ and for $u_i = T - \tau$, i.e. $\mathbf{r} \in \partial S(\mathbf{r}_i(\tau), a)$ and $\mathbf{r} \in \partial S(\mathbf{r}_i(T-\tau), a)$. For $u_i \in (\tau, T-\tau)$, and for every finite $\mathbf{r}$, the function $\partial h_i/\partial x_\alpha = 2(\, x_{\alpha i}(u_i) - x_\alpha)$, where x_α are the components of $\mathbf{r}$, is continuous on every neighborhood of the point $(u_i, \mathbf{r})$. If

$$\frac{\partial h_i}{\partial u_i} \;=\; 2(\mathbf{r}_i(u_i) - \mathbf{r}) \cdot \boldsymbol{\xi}_i(u_i) \;\neq\; 0 \tag{2.3}$$

then the function $u_i(\mathbf{r})$, given by the implicit function theorem, exists in a neighborhood of $\mathbf{r}$ and possesses the derivatives

$$\frac{\partial u_i}{\partial x_\alpha} \;=\; -\frac{\partial h_i}{\partial x_\alpha} \Big/ \frac{\partial h_i}{\partial u_i} \;=\; \frac{x_{\alpha i}(u_i) - x_\alpha}{(\mathbf{r}_i(u_i) - \mathbf{r}) \cdot \boldsymbol{\xi}_i(u_i)}\,, \; \alpha = 1,2,3. \tag{2.4}$$

Let $U_i' = \{u_{i1}', u_{i2}', ..., u_{in'}'\}$ and $U_i'' = \{u_{i1}'', u_{i2}'', ..., u_{in''}''\}$ be the solutions of (2.2) which denote the moments when the i-th molecule enters (leaves) the sphere $S(\mathbf{r}, a)$. Since the sphere $S(\mathbf{r}, a)$ is open, $H^+(a^2 - (\mathbf{r}_i(t) - \mathbf{r})^2)$ is left (right) continuous function of t when the molecule enters (leaves) the sphere. Hence for any $t \in (t - \tau, t + \tau)$, we have

$$\begin{aligned} H^+(a^2 - (\mathbf{r}_i(t) - \mathbf{r})^2) = \; & H^+(a^2 - (\mathbf{r}_i(t - \tau) - \mathbf{r})^2) \\ & + \sum_{k'=1}^{n'} H^+(t - u_{ik'}') - \sum_{k''=1}^{n''} H^-(t - u_{ik''}''), \end{aligned} \tag{2.5}$$

where H^- is the right continuous Heaviside jump function.

Let $U_i = (U_i' \cup U_i'') \cap (t - \tau, t + \tau)$. The integrand of each term in (2.1) is a continuous function of t' on $(t-\tau, t+\tau)\backslash U_i$, with the exception of a finite number of jump discontinuities; hence it is Riemann integrable. Taking the derivative of each term in (2.1) with respect to t we get

$$\begin{aligned} \partial_t \langle\varphi\rangle(\mathbf{r}, t) = \; & \tfrac{1}{2\tau V} \sum_{i=1}^{N} [\varphi_i(t + \tau) H^+(a^2 - (\mathbf{r}_i(t + \tau) - \mathbf{r})^2) \\ & - \varphi_i(t - \tau) H^+(a^2 - (\mathbf{r}_i(t - \tau) - \mathbf{r})^2)]. \end{aligned} \tag{2.6}$$

Since φ_i is a continuous function in $(\tau, T - \tau)$, the time derivative (2.6) is not continuous when the Heaviside function H^+ is discontinuous, i.e. on the set

$$\bigcup_{i=1}^{N} \{(\mathbf{r}, t) \mid t \in (\tau, T - \tau) \text{ and } \mathbf{r} \in \partial S(\mathbf{r}_i(t \mp \tau), a)\},$$

which has null Lebesgue measure in $\mathbf{R}^3 \times (\tau, T - \tau)$, hence, $\partial_t \langle\varphi\rangle$ is a.e. continuous.

In order to study the space differentiability of $\langle\varphi\rangle$ we write the terms from (2.1) using (2.5) as

$$\begin{aligned} & H^+(a^2 - (\mathbf{r}_i(t - \tau) - \mathbf{r})^2) \int_{t-\tau}^{t+\tau} \varphi_i(t')\, dt' + \\ & + \sum_{k'=1}^{n'} \int_{t-\tau}^{t+\tau} \varphi_i(t') H^+(t' - u_{ik'}')\, dt' - \sum_{k''=1}^{n''} \int_{t-\tau}^{t+\tau} \varphi_i(t') H^-(t' - u_{ik''}'') dt'. \end{aligned} \tag{2.7}$$

The terms from (2.7) depend on $\mathbf{r}$ through the function $H^+(a^2-(\mathbf{r}_i(t-\tau)-\mathbf{r})^2)$ (with zero derivatives, excepting the points where it has jump discontinuities) and the functions $u_{ik'}'$, $u_{ik''}''$, implicitely defined by (2.2). When $u_{ik'}'$ and $u_{ik''}''$ are not equal to the integration limits and if the conditions required by the implicit functions theorem (interior poins of the range of $\langle\varphi\rangle$, and (2.3)) are fulfilled, the only nonvanishing derivatives of the terms from (2.7) are

$$- \sum_{k'=1}^{n'} \varphi_i(u'_{ik'}) \frac{\partial u'_{ik'}}{\partial x_\alpha} + \sum_{k''=1}^{n''} \varphi_i(u''_{ik''}) \frac{\partial u''_{ik''}}{\partial x_\alpha}.$$

Summing over i we get

$$\partial_\alpha \langle \varphi \rangle (\mathbf{r}, t) = \frac{1}{2\tau \mathcal{V}} \sum_{i=1}^{N} \sum_{u \in U_i} \varphi_i(u) \frac{x_{\alpha i}(u) - x_\alpha}{\mid (\mathbf{r}_i(u) - \mathbf{r}) \cdot \xi_i(u) \mid}. \tag{2.8}$$

From the previous no-jump conditions for H^+ and H^- and the two conditions required by the implicit function theorem, it follows that the space derivative is not continuous on the set

$$\begin{aligned} \bigcup_{i=1}^{N} \{ \; & \{(\mathbf{r}, t) \mid t \in (\tau, T - \tau) \text{ and } \mathbf{r} \in \partial S(\mathbf{r}_i(t \mp \tau), a)\} \\ & \cup \{(\mathbf{r}, t) \mid t \in (\tau, T - \tau) \text{ and } \mathbf{r} \in \partial S(\mathbf{r}_i(\tau), a)\} \\ & \cup \{(\mathbf{r}, t) \mid t \in (\tau, T - \tau) \text{ and } \mathbf{r} \in \partial S(\mathbf{r}_i(T - \tau), a)\} \\ & \cup \{(\mathbf{r}, t) \mid \text{ there exists some } t',\ t' \in (\tau, T - \tau) \text{ such that} \\ & \mathbf{r} \in \partial S(\mathbf{r}_i(t'), a) \text{ and } (\mathbf{r}_i(t') - \mathbf{r}) \cdot \boldsymbol{\xi}_i(t') = 0\}\}. \end{aligned}$$

This set is of null Lebesgue measure in $\mathbb{R}^3 \times (\tau, T - \tau)$, hence $\partial_\alpha \langle \varphi \rangle$ is a.e. continuous function. □

The a.e. continuity of the partial derivatives ensures the continuity of $\langle \varphi \rangle$ with respect to $(\mathbf{r}, t) \in \mathbb{R}^3 \times (\tau, T - \tau)$ and then $\langle \varphi \rangle$ is an a.e. continuous field.

Proposition 2. *If the partial derivatives exist then $\langle \varphi \rangle$ satisfies the identity*

$$\partial_t \langle \varphi \rangle + \partial_\alpha \langle \varphi \xi_\alpha \rangle = \langle \frac{d}{dt} \varphi \rangle. \tag{2.9}$$

Proof: From (2.1) and (2.5), it follows that the average $\langle \frac{d}{dt} \varphi \rangle$ can be written as

$$\begin{aligned} \langle \tfrac{d}{dt} \varphi(\mathbf{r}, t) \rangle &= \tfrac{1}{2\tau \mathcal{V}} \sum_{i=1}^{N} \{ H^+(a^2 - (\mathbf{r}_i(t - \tau) - \mathbf{r})^2) \varphi_i(t') \mid_{t-\tau}^{t+\tau} + \sum_{k'=1}^{n'} \varphi_i(t') \mid_{u'_{ik'}}^{t+\tau} \\ & - \sum_{k''=1}^{n''} \varphi_i(t') \mid_{u'_{ik''}}^{t+\tau} \} = \tfrac{1}{2\tau \mathcal{V}} \sum_{i=1}^{N} \{ H^+(a^2 - (\mathbf{r}_i(t - \tau) - \mathbf{r})^2) \varphi_i(t') \mid_{t-\tau}^{t+\tau} \\ & + (n' - n'') \varphi_i(t + \tau) + \sum_{i=1}^{N} \sum_{u \in U_i} \varphi_i(u) \tfrac{(x_{\alpha i}(u) - x_\alpha) \xi_{\alpha i}(u)}{|(\mathbf{r}_i(u) - \mathbf{r}) \cdot \boldsymbol{\xi}_i(u)|} \} \end{aligned} \tag{2.10}$$

If the first term in (2.6) is expressed by (2.5), the sum over the first two terms under brackets in (2.10) gives the time derivative $\partial_t \langle \varphi \rangle$. The product of the analytic functions φ and $\xi_{\alpha i}$ is also an analytic function and, from (2.8), the second term in (2.10) is $\partial_\alpha \langle \varphi \xi_\alpha \rangle$. □

The integrand in (2.1) is an analytic function except for a finite number of points where it has finite jumps; thus, it is a bounded-variation function. There exists some theorem stating that all bounded variation functions can be uniquely spited into a sum between a jump function and a continuous one [Kolmogorov and Fomine, 1974]. Due to its analyticity, the continuous part is also absolutely continuous. According to Lebesgue's theorem, the absolutely continuous part is then given by an integral from the derivative of the bounded variation function,

$$\int_{t-\tau}^{t+\tau} \tfrac{d\varphi_i}{dt} H^+(a^2 - (\mathbf{r}_i(t') - \mathbf{r})^2)dt' = \quad [\varphi_i(t+\tau)\, H^+(a^2 - (\mathbf{r}_i(t+\tau) - \mathbf{r})^2)$$

$$-\varphi_i(t-\tau)\, H^+(a^2 - (\mathbf{r}_i(t-\tau) - \mathbf{r})^2)]_{ac}. \tag{2.11}$$

Comparing (2.11) with (2.6) we get that the absolutely continuous part of the time derivative is given by $(\partial_t\langle\varphi\rangle)_{ac} = \langle d\varphi/dt\rangle$. Thus the advection-like term in (2.9) is the time derivative due to jumps, $(\partial_t\langle\varphi\rangle)_{jumps} = -\partial_\alpha\langle\varphi\xi_\alpha\rangle$. Indeed, we have seen in the proof of the proposition 1 that the space derivative is expressed with the aid of the derivative of implicit space functions of the time moments at which the molecules enter in (or leave) the sphere. It accounts for the balance of the molecular quantities φ into a sphere and a time interval on which an imaginary measurement is performed. This is the physical meaning of identities (2.9) as "microscopic" balance equations. Although they are a.e. continuous, the fields defined by (2.1) and balance equations (2.9) give a continuous description of the physical system equivalent to that given by the corresponding N-dimensional dynamical system. In order to formulate a boundary- and initial-value problem for them we also need the whole microscopic information enclosed in the sets on which the fields $\langle\varphi\rangle$ are not defined.

In [Vamoş et al., 1996a,b] the Propositions 1 and 2 are proved in the more general case when the functions φ_i are only piece-wise analytic and when the particles of the physical system may be created or destroyed.

3 CONTINUOUS FIELDS AND BALANCE EQUATIONS

Let us compare the approach based on coarse-grained averages with the statistical mechanics one introduced by Kirkwood [1967] and used by Sposito [1978]. As a function of $\mathbf{r}_i$, H^+ from (2.1) is the characteristic (or indicator) function of the sphere $S(\mathbf{r},a) : H^+(a^2 - (\mathbf{r}_i(t') - \mathbf{r})^2) = 1_{S(\mathbf{r},a)}(\mathbf{r}_i(t'))$. For $\varphi = 1$, the definition (2.1) can be written as

$$\langle 1\rangle(\mathbf{r},t) = \frac{1}{2\tau}\int_{t-\tau}^{t+\tau} \tilde{c}(\mathbf{r},t)dt', \text{ where}$$

$$\tilde{c}(\mathbf{r},t) = \frac{1}{V}\sum_{i=1}^{N}\int_{\mathbf{R}^3} 1_{S(\mathbf{r},a)}(\mathbf{r}')\delta(\mathbf{r}' - \mathbf{r}_i(t'))d\mathbf{r}'. \tag{3.1}$$

The relation (3.1) looks like the Kirkwood's statistical mechanical definition for the concentration field, where, similarly to (3.1), a linear functional defined as a sum of Dirac distributions is considered. The difference is that in statistic mechanics a probability density (considered to be a C_0^∞ function) is used instead of the characteristic function. The probability densities satisfies the differential (Liouville) or integro-differential (Boltzmann) equations. It is by the use of these equations that the macroscopic balance equations are derived. In our approach (1) may be considered as a linear combination of Heaviside distributions. The identity (2.9) is a relation between the time and space derivatives of these distributions valid in almost all points $(\mathbf{r},t)$. An average with a suitable smoothing kernel of the a.e. fields (2.1) and of the identities (2.9) is sufficient to provide both everywhere continuous fields and balance equations. Thus only the existence of a probability density, as a smoothing kernel, is necessary; the knowledge of its evolution equation is not needed. We shall prove this in the following by the use of a general stochastic averaging procedure.

3.1 STOCHASTIC DESCRIPTION

Let $(\Omega, \mathcal{A}, P)$ be a probability space. We consider the stochastic process

$$\boldsymbol{\eta} : \Omega \longmapsto Y^I, \text{ where } Y = \mathbb{R}^{6N} \text{ and } I \subseteq \mathbb{R}. \tag{3.2}$$

The *states space* Y is the usual position-velocity space of statistical mechanics and Y^I is *the phases space* (the space of the samples, or trajectories of the stochastic process which describes the physical system). For a fixed value $\omega \in \Omega$, we note by $\mathbf{y}(\cdot;\omega) = \boldsymbol{\eta}(\omega)$ the sample $t \longmapsto \mathbf{y}(t;\omega)$, where $\mathbf{y}(t;\omega) = (\mathbf{r}(t;\omega), \boldsymbol{\xi}(t;\omega))$, $\mathbf{r}(t;\omega) = (\mathbf{r}_1(t;\omega), ..., \mathbf{r}_N(t;\omega))$, and $\boldsymbol{\xi}(t;\omega) = (\boldsymbol{\xi}_1(t;\omega), ..., \boldsymbol{\xi}_N(t;\omega))$. So, this stochastic process is defined in the sense of Doob, as a random variable into a phase space [Doob, 1953, chap. II, Iosifescu and Tăutu, 1972, p. 164]. The *distribution* of this random variable is defined by $P_{\boldsymbol{\eta}}(B) = P(\{\boldsymbol{\eta} \in B\})$, $\forall\, B \in \mathcal{B}^I$, where $\mathcal{B}^I$ is a $\sigma-$algebra in the phase space Y^I. The measure space $(\, Y^I, \mathcal{B}^I, P_{\boldsymbol{\eta}})$ is also a probability space, isomorphic to the basic probability space $(\Omega, \mathcal{A}, P)$. The *expectation*, $M[f]$, of a physical quantity described by a function $f(\boldsymbol{\eta}(\omega))$ is defined as a Lebesgue integral with respect to the probability measure P and, due to a change of variables theorem, it equals the Lebesgue integral with respect to the distribution $P_{\boldsymbol{\eta}}$ [Malliavin, 1995, p.187]:

$$M_\Omega[f] = \int\limits_{\Omega} f(\boldsymbol{\eta}(\omega)) P(d\omega) = \int\limits_{Y^I} f(\mathbf{y}) P_{\boldsymbol{\eta}}(d\mathbf{y}) \tag{3.3}$$

According to Kolmogorov theorem [Wentzell, 1981, p.81], the distribution is uniquely defined by consistent *finite-dimensional distributions*:

$$P_{t_1 \cdots t_n}(B_1 \times \cdots \times B_n) = P(\{\boldsymbol{\eta}_{t_1}(\omega) \in B_1 \cdots \boldsymbol{\eta}_{t_n}(\omega) \in B_n\}),$$

where $\boldsymbol{\eta}_{t_1} = \mathbf{y}(t_1;\cdot), \cdots, \boldsymbol{\eta}_{t_n} = \mathbf{y}(t_n;\cdot)$ are the projections of the random variable $\boldsymbol{\eta}$ for n fixed time moments, $t_1 \cdots t_n$, $B_n = (B_1 \times \cdots \times B_n) \in \mathcal{B}^n$ and $\mathcal{B}^n$ is the Borelian

$\sigma-$algebra in Y^n. The *densities* of the finite-dimensional distributions, defined by the Radon-Nikodym theorem through $\int_{B_n} p(\mathbf{y}_1, t_1 \cdots \mathbf{y}_n t_n) d\mathbf{y}_1 \cdots d\mathbf{y}_n = P_{t_1 \cdots t_n}(B_n)$, are given by [Suciu et al., 1996]:

$$p(\mathbf{y}_1, t_1 \cdots \mathbf{y}_n t_n) = M_\Omega[\delta(\mathbf{y}_1 - \eta_{t_1}(t_1, \omega)) \cdots \delta(\mathbf{y}_n - \eta_{t_n}(t_n, \omega))] \qquad (3.4)$$

From (3.4), the 1-dimensional density of the process defined by (3.2), $p(\mathbf{y}, t) = p_N(\mathbf{r}, \boldsymbol{\xi}, t)$, $p_N : \mathbb{R}^{6N} \times I \to \mathbb{R}_+$, gets the form

$$p_N(\mathbf{r}_1, ..., \boldsymbol{\xi}_N, t) = \int\limits_\Omega \delta(\mathbf{r}_1 - \mathbf{r}_1(t, \omega)) \cdots \delta(\boldsymbol{\xi}_1 - \boldsymbol{\xi}_N(t, \omega)) P(d\omega). \qquad (3.5)$$

Using (3.5) the "one-particle" density from the statistical mechanics becomes

$$\begin{aligned} p_1(\mathbf{r}, t) = & \sum_{i=1}^{N} \int\limits_{\mathbb{R}^{6N-1}} p_N(\mathbf{r}_1, ..., \mathbf{r}_{i-1}, \mathbf{r}, \mathbf{r}_{i+1}, \cdots, \boldsymbol{\xi}_N, t) d\mathbf{r}_1 \cdots d\mathbf{r}_{i-1} d\mathbf{r}_{i+1} \cdots d\boldsymbol{\xi}_N \\ = & \sum_{i=1}^{N} \int\limits_\Omega \delta(\mathbf{r} - \mathbf{r}_i(t, \omega)) P(d\omega) = M_\Omega[\sum_{i=1}^{N} \delta(\mathbf{r} - \mathbf{r}_i(t, \omega))]. \end{aligned} \qquad (3.6)$$

Statistical mechanics deals with *dynamical descriptions* of the physical system at the microscopic level, given by functions defined in the states space, $f_i(\mathbf{r}_1, ..., \boldsymbol{\xi}_N, t)$, 1 □ i □ N. Kirkwood [1967] defines *continuous fields* $F(\mathbf{r}, t)$, associated to microscopic quantities f_i, by counting the contribution of all particles to the value of the field in the point $\mathbf{r}$ at the moment t:

$$F(\mathbf{r}, t) = \sum_{i=1}^{N} \int\limits_{\mathbb{R}^{6N-1}} (f_i p_N)(\mathbf{r}_1, ..., \mathbf{r}_{i-1}, \mathbf{r}, \mathbf{r}_{i+1}, \cdots, \boldsymbol{\xi}_N, t) d\mathbf{r}_1 \cdots d\mathbf{r}_{i-1} d\mathbf{r}_{i+1} \cdots d\boldsymbol{\xi}_N. \qquad (3.7)$$

Using the definition (3.4) of the 1-dimensional density p_N, we get the equivalent expression

$$F(\mathbf{r}, t) = M_\Omega[\sum_{i=1}^{N} f_i(\mathbf{r}_1(t, \omega), ..., \mathbf{r}_{i-1}(t, \omega), \mathbf{r}, \mathbf{r}_{i+1}(t, \omega), \cdots, \boldsymbol{\xi}_N(t, \omega), t) \delta(\mathbf{r} - \mathbf{r}_i(t, \omega))]. \qquad (3.8)$$

If the density p_N is a smooth function and f_i are Riemann integrable functions then the field defined by (3.7) is a smooth space-time function, everywhere in $\mathbb{R}^3 \times I$. For $f_i \equiv 1$, (3.7) becomes the definition of the concentration field which, from (3.6), equals the one-particle density: $c(\mathbf{r}, t) = p_1(\mathbf{r}, t)$.

The coarse graining approach from the previous section uses a *kinematical description* of the physical system by time functions $\varphi_i(t)$. The connection with the dynamical description of the statistical mechanics can be established if one defines φ_i, using the stochastic process (3.2), by

$$\varphi_i(t) = \varphi_i(\mathbf{r}(t; \omega), \boldsymbol{\xi}(t; \omega), t). \qquad (3.9)$$

the functions defined by (3.9) are analytical or, at least, piece-wise anaalytical time functions, then thecoarse-grained averages (2.1) are a.e. continuous fields, by the Proposition 1, and, by The Proposition 2, these fields verify the identity (2.9). Using the previous stochastic description of the physical system we shall prove the following proposition.

Proposition 3. *If the functions (3.9) are at least piece-wise analytical, then the expectation of the space-time coarse-grained averages (2.1) equals a space-time average of the continuous field F_φ, defined by (3.7), given by*

$$M_\Omega[\langle\varphi\rangle](\mathbf{r},t;a,\tau)=\frac{1}{2\tau V}\int\limits_{t-\tau}^{t+\tau}dt'\int\limits_{S(\mathbf{r},a)}F_\varphi(\mathbf{r}',t')d\mathbf{r}', \tag{3.10}$$

and it verifies the identity

$$\partial_t M_\Omega[\langle\varphi\rangle]+\partial_\alpha M_\Omega[\langle\varphi\xi_\alpha\rangle]=M_\Omega[\Big\langle\frac{d}{dt}\varphi\Big\rangle]. \tag{3.11}$$

Proof: Using (3.9), and the obvious relation $H^+(a^2-(\mathbf{r}_i(t',\omega)-\mathbf{r})^2)=1_{S(\mathbf{r},a)}(\mathbf{r}_i(t',\omega)$, the expectation (3.3) of the coarse grained averages (2.1) becomes:

$$M_\Omega[\langle\varphi\rangle]=\int\limits_\Omega P(d\omega)[\tfrac{1}{2\tau V}\sum_{i=1}^{N}\int\limits_{t-\tau}^{t+\tau}\varphi_i(\mathbf{r}(t';\omega),\xi(t';\omega),t')\,H^+(a^2-(\mathbf{r}_i(t',\omega)-\mathbf{r})^2)\,dt']=$$
$$=\tfrac{1}{2\tau V}\int\limits_{t-\tau}^{t+\tau}dt'M_\Omega[\sum_{i=1}^{N}\int\limits_{\mathbb{R}^3}\varphi_i(\mathbf{r}_1(t',\omega),...,\mathbf{r}',\cdots,\xi_N(t',\omega),t')1_{S(\mathbf{r},a)}(\mathbf{r}')\delta(\mathbf{r}'-\mathbf{r}_i(t',\omega))d\mathbf{r}']$$
$$=\tfrac{1}{2\tau V}\int\limits_{t-\tau}^{t+\tau}dt'\int\limits_{S(\mathbf{r},a)}d\mathbf{r}'M_\Omega[\sum_{i=1}^{N}\varphi_i(\mathbf{r}_1(t',\omega),\cdots,\mathbf{r}',\cdots,\xi_N(t,\omega),t')\delta(\mathbf{r}'-\mathbf{r}_i(t',\omega))],$$

where the integrand in the last equality is the continuous field defined by (3.8), and also equivalent to (3.7). This proves (3.10). Because $\varphi_i(t)$ are at least piece-wise analytical functions the coarse grained averages (2.1) verify the identity (2.9). The expectation is a Lebesgue integral with respect to $P(d\omega)$ and it commutes with the time derivative ∂_t and space derivative ∂_α. Thus the expectation of the terms in (2.9) gives (3.11). □

The Proposition 3 relates the usual approach of statistical mechanics, based on dynamical microscopical descriptions, with the coarse-grained approach, based on kinematical descriptions. If we consider the limits, for $a\longrightarrow 0$ and $\tau\longrightarrow 0$, of the expectations (3.10) of the coarse grained averages, we get

$$\lim[M\langle\varphi\rangle](\mathbf{r},t;a,\tau)=F(\mathbf{r},t). \tag{3.12}$$

Hence, the Kirkwood's continuous fields (3.7) correspond to expectations of some fictitious measurements, on small space-time scales, modeled by coarse grained averages (2.1).

3.2 BALANCE AND DIFFUSION EQUATIONS

For $\varphi_i \equiv 1$, $1 \le i \le N$, (3.10) gives the concentration field

$$c(\mathbf{r},t) = M_\Omega[\langle 1\rangle](\mathbf{r},t), \tag{3.13}$$

We assign to each "particle" of the continuous medium *a mean value of the physical quantity* φ by

$$\bar{\varphi}(\mathbf{r},t) = \frac{1}{c(\mathbf{r},t)} M_\Omega[\langle\varphi\rangle](\mathbf{r},t) \tag{3.14}$$

For $\varphi_i \equiv \xi_{\alpha i}$, (3.14) defines the $\alpha-$ component of the *Eulerian velocity field*, $\mathbf{u}$, of the continuum medium by

$$u_\alpha(\mathbf{r},t) = \overline{\xi_\alpha}(\mathbf{r},t). \tag{3.15}$$

With (3.13-15), the identity (3.11) takes the usual form of a balance equation [Müller, 1985]:

$$\partial_t(c\,\bar{\varphi}) + \partial_\alpha(c\,\bar{\varphi}u_\alpha) + \partial_\alpha(c\overline{\varphi(\xi_\alpha - u_\alpha)}) = c\,\overline{\frac{d}{dt}\varphi}. \tag{3.16}$$

Because in the Proposition 3 only the analyticity requirement and the existence of the stochastic description were sufficient to prove the identity (3.11), such equations can be derived for any physical quantities φ not only for conservatives (or "collisional invariants") ones as it is usual in the methods based on the Liouville equation [Shinbrot, 1973].

For $\varphi_i \equiv 1$ we get the concentration balance equation

$$\partial_t c + \partial_\alpha(cu_\alpha) = 0, \tag{3.17}$$

i. e. a continuity equation. With $\xi_\alpha = dx_\alpha/dt$ in (3.15), we get $u_\alpha = \overline{dx_\alpha/dt}$, and, using (3.11), (3.17) takes the form

$$\partial_t c + \partial_\alpha(cv_\alpha) = \partial_\alpha\partial_\beta(c\,D_{\alpha\beta}). \tag{3.18}$$

The quantity v_α in (4.18) is defined by

$$v_\alpha(\mathbf{r},t) = \partial_t\overline{x_\alpha}(\mathbf{r},t) + u_\beta\partial_\beta\overline{x_\alpha}(\mathbf{r},t) = \frac{d\overline{x_\alpha}}{dt}. \tag{3.19}$$

If we consider the "fluid particle" consisting of the system of particles lying into the sphere $S(\mathbf{r},a)$ during the time interval $[t-\tau, t+\tau]$ we have a Lagrangian description of the mass transport process and $\overline{x_\alpha}$ is the α-component of the center of mass of the fluid particle. So, the material derivative (4.19) defines the α-component of the *Lagrangian velocity field*, $\mathbf{v}$. The meaning of $D_{\alpha\beta}$ is that of a *diffusion tensor* and it looks like

$$D_{\alpha\beta}(\mathbf{r},t) = \overline{x_\alpha}(\mathbf{r},t)u_\beta(\mathbf{r},t) - \overline{x_\alpha\xi_\beta}(\mathbf{r},t). \tag{3.20}$$

Thus, we proved

Proposition 4. *If the diffusion tensor (3.20) is positivelly defined, then the concentration balance equation takes the form of the advection-diffusion equation (3.18).* □

If we define the correlation-like quantity $cor(x_\alpha, \xi_\beta) = \overline{(x_\alpha \xi_\beta)}/(\overline{x_\alpha} u_\beta)$ (the correlation corresponding to our averaging procedure: the ensemble average, (3.3), of a coarse grained average (2.1)) the diffusion tensor (3.20) becomes

$$D_{\alpha\beta}(\mathbf{r}, t) = \overline{x_\alpha}(\mathbf{r}, t) u_\beta(\mathbf{r}, t)[1 - cor(x_\alpha, \xi_\beta)(\mathbf{r}, t)]. \tag{3.21}$$

For a deterministic movement of the constituent particles $cor(x_\alpha, \xi_\beta) \equiv 1$ and, from (3.21), $D_{\alpha\beta} \equiv 0$, i.e. a first test of this formula.

The averaging procedure from this paper also constitutes the ground of a cellular automata numerical method for diffusion processes [Vamoş et al., 1997, Suciu at al. 1997, 1998]. Using this algorithm, it was easy to check that for a diffusion process, simulated by random walkers cellular automata, the relation (3.20) gives the true values of the diffusion coefficients

The diffusion-like equation (3.18) was derived without any approximation and it is equivalent to the continuity equation (3.17). The positivity of the coefficients (3.20) is a criterion to say when there exists a diffusive description of the transport process, strictly equivalent to the advective one given by (3.17). Thus, we expect that the positivity of the diffusion coefficients (3.20) could be an useful tool to check if the measured, or simulated, transport processes may be described, at a given space-time scale, by an advection-diffusion equation.

4 TRANSPORT PROCESSES IN POROUS MEDIA

With the results from sections 2 and 3, we try to derive a continuous model of a fluid flow in the void space of a consolidated porous medium.

We consider a system of N molecules, $N = N^m + N^{c^1} + N^{c^2} + \cdots$, where N^m is the number of molecules from the solid matrix of the porous medium and N^{c^1}, $N^{c^2} \cdots$ are the numbers of molecules of the fluid components. To each molecular species we assign a volume $\mathcal{V}^m$, respectively $\mathcal{V}^{c^1}$, $\mathcal{V}^{c^2}, \cdots$ and a molecular mass $\mathcal{M}^m$, respectively $\mathcal{M}^{c^1}$, $\mathcal{M}^{c^2}, \cdots$.

4.1 CONTINUOUS MODEL OF THE SOLID MATRIX

For the continuous model of the solid matrix we first consider a time scale τ much greater than the period of the molecular vibrations and a space scale a much greater than the molecular dimensions and much smaller than the mean pores diameter, d_p,

$$(\mathcal{V}^m)^{\frac{1}{3}} \ll a \ll d_p. \tag{4.1}$$

Because, at the considered time scale τ, the molecules of the solid matrix have fixed positions, the coarse-grained average of $\mathcal{M}^m$, by (2.1), becomes

$$\langle\mathcal{M}^m\rangle(\mathbf{r}) = \frac{\mathcal{M}^m}{\mathcal{V}}\sum_{i=1}^{N} H^+(a^2-(\mathbf{r}_i-\mathbf{r})^2). \tag{4.2}$$

The value of the function defined by (4.2) is zero if inside the sphere $S(\mathbf{r},t)$ there exists no molecule of the solid matrix species. The expectation of (4.2) is given by (3.10) as

$$M_\Omega[\langle\mathcal{M}^m\rangle](\mathbf{r};a,\tau) = \frac{1}{2\tau\mathcal{V}}\int_{t-\tau}^{t+\tau} dt' \int_{S(\mathbf{r},a)} F_{\mathcal{M}^m}(\mathbf{r}',t')d\mathbf{r}'. \tag{4.3}$$

Following (3.12), for small scales, closed to the lower bound of the range (4.1), we can estimate have $M_\Omega[\langle\mathcal{M}^m\rangle] \sim F_{\mathcal{M}^m}$. Then, the expectation (4.3), for small scales, approximates the continuous mass field $F_{\mathcal{M}^m}$, associated with the molecules of the solid matrix. This is a bulk density. If $\mathbf{r}$ is a position in the interior of the solid matrix, then $F_{\mathcal{M}^m} = \rho^m$, i.e. it is the density of the solid material. Because (4.2) is zero in void spaces so is its expectation (4.3). We define *the characteristic function of the solid matrix* by the ratio between the bulk density $F_{\mathcal{M}^m}$ and the true density ρ^m,

$$1^m(\mathbf{r}) \;=\; \frac{F_{\mathcal{M}^m}}{\rho^m}. \tag{4.4}$$

The function defined by (4.4) is continuous, excepting the points on the surface of the solid matrix, which have zero Lebesgue measure. Thus $1^m(\mathbf{r})$ is Riemann integrable.

At a scale much greater than the mean pores diameter,

$$a \gg d_p, \tag{4.5}$$

we define the local average *porosity* by the ratio between the volumes of void space and solid matrix contained in $S(\mathbf{r},t)$,

$$\begin{aligned}\Theta(\mathbf{r}) =& \ \tfrac{1}{\mathcal{V}}\int_{S(\mathbf{r},a)} (1-1^m(\mathbf{r}'))d\mathbf{r}' = \\ =& \ \tfrac{1}{\mathcal{V}}\int_{\mathbf{R}^3}(1-1^m(\mathbf{r}'))H^+(a^2-(\mathbf{r}'-\mathbf{r})^2)d\mathbf{r}'.\end{aligned} \tag{4.6}$$

Here $(1-1^m)$ is the characteristic function of the void space and $H^+(a^2-(\mathbf{r}'-\mathbf{r})^2)$ is the characteristic function of the sphere. For the modelling consistency, we remark that, for small a, given by (4.1), we have $\Theta \sim (1-1^m)$. The porosity $\Theta(\mathbf{r})$, defined by (4.6), at scales (4.5), much greater than the pores dimensions, is a positive continuous function. The definition (4.6) corresponds to the *local porosity* introduced by Hilfer [1991], where, instead of spheres of radius a, a cubic cells ("Bravais lattice") were considered.

The solid matrix volume ratio, Θ^m, has an analogous definition and we have $\Theta^m + \Theta = 1$. Generally, at scales much greater than the pores dimensions, we have the following relation for the components of the porous media,

$$\Theta^{m}+\Theta^{c^1}+\Theta^{c^2}+\cdots=\frac{F_{M^m}}{\rho^m}+\frac{F_{M^{c^1}}}{\rho^{c^1}}+\frac{F_{M^{c^2}}}{\rho^{c^2}}+\cdots=1. \tag{4.7}$$

In the framework of the theory of mixtures the relation (4.7) "reflects the assumption that the mixture does not contain void spaces" [Bowen, 1984, p. 67]. Thus, under the scale assumptions (4.1) and (4.5), the definitions (4.4) and (4.6) can provide continuous models for the solid matrix and, respectively, the "mixture" formed by the fluid and solid matrix.

4.2 DARCY-BUCKINGHAM FLUX LAW

Now, consider a space scale obeying (4.5) and a time scale corresponding to experimental measurements. The *volume fraction of the component c*, θ^c, can be defined as a continuous field by the expectation of $\langle\mathcal{V}^c\rangle$, according to (3.10), as

$$\theta^c(\mathbf{r},t)=\mathcal{V}^c M_\Omega[\langle 1\rangle](\mathbf{r},t)=\mathcal{V}^c c_a^c(\mathbf{r},t), \tag{4.8}$$

were c_a^c is the apparent, or bulk, concentration of the molecules from the c-species. The identity (3.11) becomes

$$\partial_t\theta^c+\partial_\alpha(u_\alpha^c\theta^c)=0, \tag{4.9}$$

where $c_a^c u_\alpha^c=M_\Omega[\langle\xi_\alpha^c\rangle]$. Hence, (4.9) is a continuity equation for the volume fraction of the c component. The center of mass of the fluid particle corresponding to the c-component of the porous medium has the components $R_\alpha^c=\overline{x_\alpha^c}=M_\Omega[\langle x_\alpha^c\rangle]/c_a^c$ and the corresponding Lagrangian velocity is $v_\alpha^c=dR_\alpha^c/dt$. Then, in the condition of Proposition 4, the continuity equation (4.9) takes the equivalent form

$$\partial_t\theta^c+\partial_\alpha(v_\alpha^c\theta^c)=\partial_\alpha\partial_\beta(K_{\alpha\beta}\,\theta^c), \tag{4.10}$$

where

$$K_{\alpha\beta}(\mathbf{r},t)=R_\alpha^c u_\beta^c\left(1-\frac{\overline{x_\alpha^c\xi_\beta^c}}{R_\alpha^c u_\beta^c}\right). \tag{4.11}$$

From (4.9-10) we get the following definition of the flux of the volume fraction θ_α^c :

$$J_\alpha=\theta^c(u_\alpha^c-v_\alpha^c)=-\partial_\beta(K_{\alpha\beta}\,\theta^c). \tag{4.12}$$

Let us consider the case when θ^c represents the water content of a soil or aquifer. If we define *the filtration velocity* by $\mathbf{u}_f=(\mathbf{u}^c-\mathbf{v}^c)\theta^c$, then (4.12) is the well known *Darcy-Buckingham flux law*, written for the volumetric water flux density [Sposito, 1986]. For instance, when the saturated aquifer case is considered, the water content has the form

$$\theta^c(x,y,z,t)=\begin{cases}0 & \text{for } z>f(x,y,t)\\ \Theta & \text{for } z\ \square\ f(x,y,t)\end{cases},$$

where $x\in\mathbb{R}$, $y\in\mathbb{R}$, $z\in[0,\infty)$, Θ is the porosity given by (4.6) and $f(x,y,t)$ is a function describing the free surface (the water level in porous medium). If we suppose the coefficients (4.11) to be constants and we integrate (4.12) with respect to z we obtain the following expressions for the components of the volumetric flux,

$$\begin{aligned} J_x(x,y,t) = & \int_0^\infty u_{fx}(x,y,z,t)\theta^c(x,y,z,t)dz = & -K_{xx}\Theta\partial_x f(x,y,t) - K_{xy}\Theta\partial_y f(x,y,t) \\ J_y(x,y,t) = & \int_0^\infty u_{fy}(x,y,z,t)\theta^c(x,y,z,t)dz = & -K_{yx}\Theta\partial_x f(x,y,t) - K_{yy}\Theta\partial_y f(x,y,t) \end{aligned}$$

Thus, (4.12) takes the usual form of the Darcy law for saturated aquifers [Bowen, 1984],

$$\mathbf{J} = -\tilde{\mathbf{K}}\nabla f.$$

4.3 POROSITY DEPENDENT ADVECTION-DIFFUSION EQUATION

Now, we consider that the c-species molecules constitute a solute of a fluid contained in a saturated porous medium. Then, the fluid volume fraction equals the porosity Θ. The true concentration, $c(\mathbf{r},t)$, measured on fluid samples, is

$$c(\mathbf{r},t) = \frac{\zeta_a^c(\mathbf{r},t)}{\Theta(\mathbf{r})}. \tag{4.13}$$

Dividing the equation (4.10) by $\mathcal{V}^c$ and using (4.13), we get the porosity dependent advection-diffusion equation

$$\Theta\partial_t c + \partial_\alpha(v_\alpha^c c\Theta) = \partial_\alpha\partial_\beta(K_{\alpha\beta}\ c\Theta). \tag{4.14}$$

The equations (4.9) and (4.14) are usually taken as the starting point in modeling the transport of non-reactive dissolved solutes through soils and aquifers [Sposito et all., 1986, Shvidler, 1993].

5 CONCLUSIONS

The model of transport in porous media we have proposed here is based on a microscopic kinematical description of the physical system. No dynamical properties were assumed. Also, no statistical assumptions and evolution equation for the probability densities were necessary. This is the distinctive feature of our method with respect to both stochastic and statistical mechanical approaches. The stochastic theory of transport does not use kinematic or dynamic descriptions of the system but it starts with the study of some abstract stochastic process. The associated Fokker-Planck equation becomes the diffusion equation and the constitutive laws for the diffusion coefficients are expressed through statistic correlations [Suciu et. all., 1996]. The statistical mechanical approach of Sposito [1978] uses the dynamical microscopic description in the states space of the physical system and the evolution equation of the probability density function.

The macroscopic balance equation for the water content is inferred and the constitutive law is found as a function of the velocity correlation. The Darcy law follows for a simplified model of noninteracting fluid molecules (the so called "darcions"). Thus the utility of statistical mechanical approach is conditioned by the knowledge of some stochastic quantities.

The relationship between the model and the measurement scales represents the highest difficulty in modeling continuous media, mainly heterogeneous ones as mixtures and porous media. If this relation is disregarded "the development of transport equations ... may result in field variables that are nothing more than unwanted noise" [Cushman, 1986]. Or, more categorical, " If the scales of measurements associated with experimental methods are not accounted for in theories of transport in hierarchical porous media, then the theories are metaphysical and of little practical consequence" [Cushman, 1990]. In subsurface (soils and aquifers) transport theories, the controversial questions are the existence of the "macrodispersion" and the "scale effect". Cushman expresses his doubt that such effects really do exist and shows that they can be explained by the inadequacy between the model and measurement scales. In our approach, the definition of continuous fields by the expectation of the coarse grained averages and the identity (2.9), which coarse grained averages verify, leads to macroscopic balance equations. The explicit use of the space-time parameters a and τ into the definition (2.1) of the coarse grained average answers to the criticism of Cushman that field variables and constitutive parameters have unambiguous meaning only for specified space-time scales.

As discussed in sub-section 3.2, we expect that this approach may be developed towards the analysis of the experimental data and, also, it may be useful in the implementation of the cellular automata simulation methods for transport processes in porous media.

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