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Effective medium approximation and exact formulae for electrokinetic phenomena in porous media

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Abstract

Electrokinetic phenomena in porous media are studied by application of the effective medium theory and the theory of duality transformation. We deduce new exact relations and analytical formulae for the effective constants of the macroscopic tensor. We also prove that the effective tensors obtained by these approaches coincide for 2D problems. The obtained results for the electrokinetic processes are closely related to similar results derived for piezoelectric composites because of a common mathematical background.

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1. Introduction

This paper is primarily devoted to the determination of the macroscopic properties of porous media undergoing electrokinetic phenomena by extension of the effective medium theory (EMT). We also develop the theory of duality transformation (TDT) of 2D media and establish a deep relation between EMT and TDT.

First, let us review results devoted to *scalar problems* when pure conductivity or elasticity is discussed. EMT is based upon local consideration of the composite material/porous media, when an inclusion is embedded in a homogeneous medium whose effective conductivity/permeability is the unknown to be determined by averaging the local structure. Application of EMT leads to analytical formulae for the effective conductivity of composite materials. It was originated by Bruggemann [1] and developed by Kirkpatrick [2] who proposed approximating the medium by square or cubic networks. Self-consistent methods, the methods of Mori–Tanaka [3], differential effective medium methods and their relations and extensions were discussed in [4–7, 9, 10].

The scalar theory of duality transformation of 2D media was discovered by Keller [11] for two-phase media and independently by Matheron [12] for general media. It is based on the observation that a divergent-free field produces a curl-free field when rotated locally

by 90° . Application of TDT yields an expression for the conductivity of a two-phase composite when phases are interchanged. In particular, the famous square-root formula for the effective conductivity was deduced in [13, 14]. For multiphase media, other exact formulae were obtained in [15–19, 24].

The following duality transformation between two different conductivity problems was proposed by Milton [20]. Let $\sigma(\mathbf{x})$ be the conductivity tensor in a medium; define the conductivity tensor $\sigma'(\mathbf{x})$ by

$$\sigma'(\mathbf{x}) = [a\sigma(\mathbf{x}) + b\mathbf{R}][c\mathbf{I} + \mathbf{R}\sigma(\mathbf{x})]^{-1} \quad (1)$$

where a, b, c, d are constants; \mathbf{R} is the matrix for 90° rotation. Assuming that the media are macroscopically homogeneous, Milton proved that their effective conductivity tensors are also related via (1).

Shvidler (see [21]) found a relation for the components of the effective tensor assuming three properties: (S1) the principal axes of the local conductivity tensor do not depend on the space variables; (S2) the principal components have isotropic correlations and the same distribution; (S3) the distributions of the fluctuations of the logarithm of the principal components are even functions.

Further discussion of 3D scalar problems for statistical (continuous) distributions of phases can be found in [22, 23].

The literature on EMT and TDT in the scalar case is very rich. Here, we mention the latter papers, where one can find references to many other results (see also the books by Milton [25] and Torquato [26]).

In the present paper we discuss a mathematical model of the electrokinetic phenomena applying EMT and TDT. A theoretical and numerical study of the electrokinetic phenomena was made in [27].

Let \mathbf{u} denote the seepage velocity, \mathbf{i} the current density, p the pressure, ψ the electric potential. Electro-osmotic effects are governed by the following equations [27]

$$\mathbf{u} = -\mathbf{k} \cdot \nabla p - \boldsymbol{\alpha} \cdot \nabla \psi \quad \mathbf{i} = -\boldsymbol{\alpha} \cdot \nabla p - \boldsymbol{\sigma} \cdot \nabla \psi \quad (2a)$$

$$\nabla \cdot \mathbf{u} = 0 \quad \nabla \cdot \mathbf{i} = 0 \quad (2b)$$

where ∇ is the gradient operator, \mathbf{k} the ratio of the permeability tensor and of the field viscosity, $\boldsymbol{\sigma}$ the electrical conductivity tensor, and $\boldsymbol{\alpha}$ is the electroosmotic coupling tensor. These tensors are random matrix functions of the position \mathbf{x} ; these random functions are required to be stationary. Hence, the macroscopic behaviour of the medium can be described by a constant tensor \mathbf{L} . Applying the results of [28], one can deduce a general relation between the components of \mathbf{L} .

Although EMT and TDT for electrokinetic phenomena have not been discussed before, one can use known results for piezoelectric composites where the electric and elastic fields are coupled [29]. Benveniste [28] extended Milton's approach [20] to piezoelectric composites in an anti-plane mode deformation which is coupled to an in-plane electric field. A transformation like (1) which maps the local fields into an equivalent class of heterogeneous media was given. A correspondence relation between the effective constants of these media was also derived. A phase interchange connection for two-phase media was deduced. Milgrom and Shtrikman [30] studied the effective properties of the coupled multifield of a polycrystal made of uniaxial crystals. Schulgasser [31] and Cheng [32] established exact relations for overall moduli of piezoelectric composites consisting of two and many transversely isotropic phases. Grabovsky developed a general algebraic theory to find exact relations for effective moduli of composites (see, for instance, [33]). Olson and Avellaneda [34]

applied EMT and extended Hashin–Shtrikman bounds to isotropic polycrystals. They obtained expressions for the effective moduli in the effective medium approximations wherein each grain behaves like a sphere surrounded by a homogenized medium. Dunn and Taya [35] calculated the effective tensor of the piezoelectric two-phase composites by application of various theories and methods such as the dilute approach, EMT, the Mori–Tanaka method and the differential scheme. The application of each method was based on the exact solution of a 3D vector–matrix problem for an ellipsoidal inclusion in an infinite medium.

When locally isotropic media are considered, (2) becomes

$$\mathbf{u} = -k\nabla p - \alpha\nabla\psi \quad \mathbf{i} = -\alpha\nabla p - \sigma\nabla\psi \quad (3a)$$

$$\nabla \cdot \mathbf{u} = 0 \quad \nabla \cdot \mathbf{i} = 0 \quad (3b)$$

where k , α and σ are now scalars. Following Matheron, we derive new formulae which can be also applied to piezoelectric phenomena. Moreover, we extend EMT to coupled processes described by equations (3). There is a remarkable relation between these two techniques. It was numerically shown in [36] that Matheron’s formula and the EMT provide the same result. In the present paper, this result is established analytically. It should be mentioned that the question of the exact relation of the effective medium approximation by certain two-phase microstructures was discussed in [37, 38].

In contrast with the previous works devoted to duality transformations for *vector–matrix problems*, we impose conditions on the statistical properties of the local quantities that allow us to obtain not only relations between the effective properties, but also exact formulae for them when media are described by continuous local laws.

The present paper is organized as follows. First, we construct effective medium approximations for (3) in 2D (section 2) and 3D (section 3). We deduce the effective medium approximation for n -phase media (see (15) for 2D and (26) for 3D) and for general media in integral form. This relation is written as an integral over the local parameters k , α , σ (see (16) for 2D and (27) for 3D) and as an integral on the porosity ϵ (see (21) for 2D and (28) for 3D). The latter relation is obtained under the assumption that all the local parameters k , α , σ depend on the local porosity. Section 4 is devoted to the simple scalar case, where we deduce the duality identity for general media and analytically prove the identity of Matheron’s formula and EMT in 2D. We present the scalar results in a separate section in order to expose the relation between the continuous and discrete phase distributions. It should be noted that our formula (36) was deduced by Shvidler (see [21]) in another form in a special case (compare the assumptions (S1)–(S3) and our hypothesis (H)). In section 4, we also prove that the effective conductivity obtained by TDT coincides with the effective conductivity obtained by EMT for continuous phase distribution when assumption (H) is fulfilled. This fact was already noted in [38] for two-phase media. In section 5, we deduce a duality identity in the vector–matrix case. In theorem 2, we propose two exact formulae for the effective tensor for media satisfying conditions (H1) and (H2). These conditions are analogous to Mendelson’s condition [14] for two-phase materials and to Matheron’s conditions for general media (see (M1) and (M2) in section 4). Respectively, the exact formulae from theorem 2 can be considered as a vector–matrix counterpart of the square-root formula [11, 13] for two-phase materials and to Matheron’s formulae (33) and (34) for general media. Section 6 is devoted to the relation between EMT and vector-matrix TDT. In general both methods are known to have a limited range of validity. But in 2D under conditions (H1) and (H2), the effective tensor obtained by EMT and TDT for the vector–matrix gives exactly the same result. Numerical results and examples are presented in section 7.

2. EMT in 2D

Following Bruggeman [39] (p 442), we construct the effective medium theory for the coupled equations (3). Let us introduce the local matrix

$$\mathbf{I} = \begin{pmatrix} k & \alpha \\ \alpha & \sigma \end{pmatrix}. \quad (4)$$

A matrix \mathbf{L} which describes the macroscopic behaviour of the medium can be defined as

$$\mathbf{L} = \begin{pmatrix} K & A \\ A & \Sigma \end{pmatrix}. \quad (5)$$

Consider a 2D basic element in the complex plane \mathbf{C} under the form of a disc of radius r_k made of a material with constant coefficients defined by a constant matrix \mathbf{I}_k of the form (4). The disc $D_k^+ = \{z \in \mathbf{C} : |z - a_k| < r_k\}$ is immersed in a material which occupies the domain $D_k^- = \{z \in \mathbf{C} : |z - a_k| > r_k\}$ whose properties are defined by the matrix \mathbf{L} defined by (5). The whole material is submitted to constant macroscopic gradients $\overline{\nabla p}$ and $\overline{\nabla \psi}$.

Let p^+ , ψ^+ and p^- , ψ^- denote the potentials in the domains D_k^+ , D_k^- , respectively. Let, for instance, $p_n^+ := \frac{\partial p^+}{\partial n}$ be the normal derivative of the interior potential p^+ . The following conjugation conditions hold on $L_k = \{z \in \mathbf{C} : |z - a_k| = r_k\}$ for equations (3)

$$\begin{pmatrix} p^+ \\ \psi^+ \end{pmatrix} = \begin{pmatrix} p^- \\ \psi^- \end{pmatrix} \quad (6a)$$

$$\mathbf{I}_k \begin{pmatrix} p_n^+ \\ \psi_n^+ \end{pmatrix} = \mathbf{L} \begin{pmatrix} p_n^- \\ \psi_n^- \end{pmatrix}. \quad (6b)$$

We shall solve the problem (6) by introduction of the complex potentials $\phi_i^\pm(z)$ ($i = 1, 2$) which are analytic in the domains D_k^\pm

$$\phi^\pm(z) = \begin{pmatrix} \phi_1^\pm(z) \\ \phi_2^\pm(z) \end{pmatrix}. \quad (7)$$

We define

$$\begin{pmatrix} p^+ \\ \psi^+ \end{pmatrix} = \text{Re } \phi^+(z) \quad z \in D_k^+ \quad \begin{pmatrix} p^- \\ \psi^- \end{pmatrix} = \text{Re } (\phi^-(z) + \mathbf{e}z) \quad z \in D_k^- \quad (8)$$

where the vector $\mathbf{e} = \begin{pmatrix} e_1 \\ e_2 \end{pmatrix}$ corresponds to the applied field. More precisely, it can be written in the following complex form $e_1 = p_1 - ip_2$, $e_2 = \psi_1 - i\psi_2$, where $\overline{\nabla p} = (p_1, p_2)$ and $\overline{\nabla \psi} = (\psi_1, \psi_2)$. Standard manipulations yield the complex potentials

$$\phi^+(z) = 2z(\mathbf{I} + \mathbf{L}^{-1}\mathbf{I}_k)^{-1} \mathbf{e} + 2(\mathbf{I} + \mathbf{L}^{-1}\mathbf{I}_k)^{-1} \mathbf{c} \quad (9)$$

$$\phi^-(z) = \frac{1}{2}(\mathbf{I} + \mathbf{L}^{-1}\mathbf{I}_k) \left(2 \left(\frac{r_k^2}{z - a_k} + \overline{a_k} \right) (\mathbf{I} + \mathbf{L}^{-1}\mathbf{I}_k)^{-1} \mathbf{e} + 2(\mathbf{I} + \mathbf{L}^{-1}\mathbf{I}_k)^{-1} \overline{\mathbf{c}} \right) + \mathbf{c} \quad (10)$$

where \mathbf{c} is an arbitrary complex vector, and \mathbf{I} is the unit tensor. We are interested in the coefficient \mathbf{c}_k of $\phi^-(z)$ on $\frac{1}{z - a_k}$ which expresses the dipole moment of the complex potential $\phi^-(z)$

$$\mathbf{c}_k = r_k^2 (\mathbf{I} - \mathbf{L}^{-1}\mathbf{I}_k) (\mathbf{I} + \mathbf{L}^{-1}\mathbf{I}_k)^{-1} \mathbf{e}. \quad (11)$$

Then, the total dipole moment corresponding to a disk of property \mathbf{l}_k is proportional to

$$\pi_k(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l}_k)(\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}_k)^{-1} \mathbf{e} \tag{12}$$

where π_k is the volumetric probability of the k th phase. These probabilities verify

$$\sum_{k=1}^n \pi_k = 1. \tag{13}$$

One demands that the total moment created by all phases composing the porous medium vanishes. Hence,

$$\sum_{k=1}^n \pi_k(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l}_k)(\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}_k)^{-1} \mathbf{e} = \mathbf{0}. \tag{14}$$

Since (14) should be valid whatever \mathbf{e} , we obtain

$$\sum_{k=1}^n \pi_k(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l}_k)(\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}_k)^{-1} = \mathbf{0}. \tag{15}$$

The effective tensor \mathbf{L} is the solution of equation (15) which can be also written in the following continuous form

$$\int_{\Omega_l} \pi(\mathbf{l})(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l})(\mathbf{I} + \mathbf{L}^{-1}\mathbf{l})^{-1} d\mathbf{k} d\alpha d\sigma = \mathbf{0} \tag{16}$$

where Ω_l is the set of admissible k, α, σ . We can put $\Omega_l = \mathbf{R}^3$ and $\pi(\mathbf{l}) = 0$ for not admissible variables, for instance for $k < 0, \sigma < 0$.

It is easily checked that (15) and (16) reduce to Kirkpatrick's formula [2] when \mathbf{l} is a diagonal tensor.

Using (4) and (5), (16) can be expanded as four scalar equations

$$\int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} (A^2 - \alpha^2 + (k - K)(\sigma + \Sigma)) d\mathbf{k} d\alpha d\sigma = 0 \tag{17a}$$

$$\int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} (\alpha \Sigma - \sigma A) d\mathbf{k} d\alpha d\sigma = 0 \tag{17b}$$

$$\int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} (A^2 - \alpha^2 + (k + K)(\sigma - \Sigma)) d\mathbf{k} d\alpha d\sigma = 0 \tag{17c}$$

$$\int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} (\alpha K - kA) d\mathbf{k} d\alpha d\sigma = 0 \tag{17d}$$

where $\Delta_0 = \det(\mathbf{I} + \mathbf{L}) = (k + K)(\sigma + \Sigma) - (\alpha + A)^2$.

One of the relations (17a) is dependent on the others because of the expected symmetry of \mathbf{L} . Let us prove it. Subtracting (17c) from (17a), we obtain

$$\int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} (-K\sigma + k\Sigma) d\mathbf{k} d\alpha d\sigma = \int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} (K\sigma - k\Sigma) d\mathbf{k} d\alpha d\sigma. \tag{18}$$

This yields the relation

$$\frac{1}{\Sigma} \int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} \sigma d\mathbf{k} d\alpha d\sigma = \frac{1}{K} \int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} k d\mathbf{k} d\alpha d\sigma. \tag{19}$$

Equation (17b) implies that

$$\frac{1}{\Sigma} \int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} \sigma d\mathbf{k} d\alpha d\sigma = \frac{1}{A} \int_{\Omega_l} \pi(\mathbf{l}) \frac{1}{\Delta_0} \alpha d\mathbf{k} d\alpha d\sigma. \tag{20}$$

Then, (17d) follows from (19) and (20). Therefore, one can consider the three relations (17a)–(17c) as three equations for the macroscopic properties K , A , Σ .

Finally, let us assume that all the local properties only depend on the local porosity ϵ which is assumed to be a random and stationary function of space (cf [27] for detailed examples). Then, (16) yields

$$\int_0^1 f(\epsilon)(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l}(\epsilon))(\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}(\epsilon))^{-1} d\epsilon = 0 \quad (21)$$

where $f(\epsilon)$ is the probability density of the porosity ϵ .

3. EMT in 3D

Following the previous section and [39], we construct an effective medium approximation for equations (3). It can be directly checked that in this case the solution of the problem (6) for a sphere of radius r_k centred at \mathbf{x}_k has the form

$$\begin{pmatrix} P^+ \\ \psi^+ \end{pmatrix} = 3(2\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}_k)^{-1} \mathbf{e}(x^{(1)} - x_k^{(1)}) \quad (22)$$

$$\begin{pmatrix} P^- \\ \psi^- \end{pmatrix} = \mathbf{e}(x^{(1)} - x_k^{(1)}) + (\mathbf{I} - \mathbf{L}^{-1}\mathbf{l}_k)(2\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}_k)^{-1} \mathbf{e} \frac{r_k^3}{r^3} (x^{(1)} - x_k^{(1)}) \quad (23)$$

where $r = |\mathbf{x} - \mathbf{x}_k|$. Here, for simplicity, we take the external field along the $x^{(1)}$ -direction

$$\mathbf{e}(x^{(1)} - x_k^{(1)}) = \begin{pmatrix} \overline{\nabla P} \\ \overline{\nabla \psi} \end{pmatrix} (x^{(1)} - x_k^{(1)}). \quad (24)$$

The dipole moment corresponding to the contribution of the phase described by \mathbf{l}_k is proportional to

$$\pi_k(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l})(2\mathbf{I} + \mathbf{L}^{-1}\mathbf{l})^{-1} \mathbf{e}. \quad (25)$$

Equating the total moment to zero, we obtain in the discrete case

$$\sum_{k=1}^n \pi_k(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l}_k)(2\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}_k)^{-1} = 0 \quad (26)$$

and in the continuous case

$$\int_{\Omega_l} \pi(l)(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l})(2\mathbf{I} + \mathbf{L}^{-1}\mathbf{l})^{-1} dk d\alpha d\sigma = 0. \quad (27)$$

A relation analogous to (21) can be written as

$$\int_0^1 f(\epsilon)(\mathbf{I} - \mathbf{L}^{-1}\mathbf{l}(\epsilon))(2\mathbf{I} + \mathbf{L}^{-1}\mathbf{l}(\epsilon))^{-1} d\epsilon = 0. \quad (28)$$

Hence, (27) and (28) are identical to (16) and (21), except for the factor 2.

4. Scalar problem in two-dimensional EMT and TDT

As mentioned in the introduction, there is a relation between EMT and TDT. First, we discuss this relation in the simple scalar case, when $\alpha = 0$. However, we discuss in the present section

more complicated locally anisotropic media. In this case, (3) is separated into two similar independent systems of equations. One of them is

$$\mathbf{u} = -\mathbf{k}\nabla p \quad \nabla \cdot \mathbf{u} = 0 \tag{29}$$

where the permeability $\mathbf{k} = \mathbf{k}(x, y)$ is expressed as a matrix function (tensor function) defined on the plane \mathbf{R}^2 . We assume that $\mathbf{k}(x, y)$ can be averaged (homogenized), i.e., $\mathbf{k}(x, y)$ is a doubly periodic matrix function or that $\mathbf{k}(x, y)$ can be considered as a stochastic stationary process on \mathbf{R}^2 (for details see [40, 41]). We are looking for the macroscopic permeability tensor \mathbf{K} which is determined by the relation

$$\langle \mathbf{k}\nabla p \rangle = \mathbf{K}\langle \nabla p \rangle \tag{30}$$

where the brackets denote the spatial average

$$\langle \cdot \rangle := \lim_{n \rightarrow \infty} |D_n|^{-1} \int_{D_n} \cdot \, dx \, dy \tag{31}$$

D_n ($n = 1, 2, \dots$) is a sequence of domains exhausting the plane \mathbf{R}^2 .

Let us introduce the resistivity tensor $\mathbf{h} := \mathbf{k}^{-1}$. Matheron [12] considered a medium when the following two hypotheses are fulfilled:

(M1) $\mathbf{k}(x, y)$ is invariant under rotations.

For a random field, (M1) means that the distribution of \mathbf{k} is invariant under rotations. This condition implies that the medium is macroscopically isotropic and that the following tensors are spherical

$$\langle \mathbf{k} \rangle = k_0 \mathbf{I} \quad \langle \mathbf{h} \rangle = h_0 \mathbf{I} \quad \mathbf{K} = K \mathbf{I} \tag{32}$$

where k_0, h_0 and K are scalars.

(M2) $\mathbf{k}(x, y)/k_0$ and $\mathbf{h}(x, y)/h_0$ have the same spatial distribution.

Using (M1) and (M2), Matheron deduced the exact formulae

$$K = \sqrt{k_0/h_0} \tag{33}$$

and

$$\ln K \mathbf{I} = -\langle \ln \mathbf{k} \rangle. \tag{34}$$

In the particular case where $\mathbf{k}(x, y)$ takes only two constant scalar values k_1 and k_2 with the same probability $1/2$, (33) yields the Dykhne–Mendelson formula [13, 14]

$$K = \sqrt{k_1 k_2}. \tag{35}$$

Instead of hypotheses (M1) and (M2), let us impose a weaker condition:

(H) $\mathbf{k}(x, y)\langle \mathbf{k}(x, y) \rangle^{-1}$ and $\mathbf{h}^*(x, y)\langle \mathbf{h}^*(x, y) \rangle^{-1}$ have the same spatial distribution.

Here, \mathbf{h}^* denotes the adjoint of \mathbf{h} . Then, it follows from [42] that

$$|K| = \frac{|\langle \mathbf{k} \rangle|}{|\langle \mathbf{k}/|\mathbf{k}| \rangle|} \tag{36}$$

where $|\cdot|$ denotes the determinant of the matrix. In the particular case where $\mathbf{k}(x, y)$ takes only two constant values $k_1 \mathbf{I}$ and $k_2 \mathbf{I}$ with the same probability $\frac{1}{2}$, we obtain Keller’s identity [11]

$$k_x k_y = k_1 k_2. \tag{37}$$

Here, k_x is the effective permeability of the medium in the x -direction; k_y is the effective permeability in the y -direction of another medium which is deduced from the original one by exchanging the phases with different permeabilities k_1 and k_2 .

Let us come back now to the effective medium approximation in 2D. In the considered scalar case, equation (16) for \mathbf{L} becomes

$$X \equiv \int_0^\infty d\sigma p(\sigma) \frac{\sigma - \sigma_e}{\sigma + \sigma_e} = 0 \quad (38)$$

where $p(\sigma)$ is the probability density of the local conductivities, and σ_e the effective conductivity corresponding to the tensor \mathbf{L} .

Adler and Berkowitz [36] used numerical simulations of flow through random lattice models for the lognormal distribution of local conductivities

$$p(\sigma) = \frac{1}{b\sigma\sqrt{2\pi}} \exp\left(-\frac{(\ln \sigma - a)^2}{2b^2}\right). \quad (39)$$

They compared the simulated effective conductivities σ_s which are equal to e^a for the lognormal distribution (39) to the effective medium approximation σ_e from (38). They found that σ_s is in agreement with σ_e . We now confirm this numerical result analytically.

Theorem 1. Equation (38) has a unique solution

$$\sigma_e = e^a. \quad (40)$$

Proof. The existence and uniqueness for (38) follow from the general EMT, where a unique effective conductivity σ_e exists for macroscopically isotropic media.

We now prove that this unique solution verifies (40). Let us change the variables in the integral (38)

$$\sigma = \frac{e^{2a}}{s}. \quad (41)$$

Then,

$$X = - \int_0^\infty p(\sigma) \frac{s - e^{2a}/\sigma_e}{s + e^{2a}/\sigma_e} ds. \quad (42)$$

Use the relation

$$\frac{e^{2a}}{s^2} p\left(\frac{e^{2a}}{s}\right) = p(s). \quad (43)$$

Equation (42) for σ_e which is equivalent to equation (38), can be written as

$$\int_0^\infty p(s) \frac{s - \frac{e^{2a}}{\sigma_e}}{s + \frac{e^{2a}}{\sigma_e}} ds = 0. \quad (44)$$

Since (38) and (44) have a unique solution, these equations give the same result

$$\sigma_e = \frac{e^{2a}}{\sigma_e} \quad (45)$$

which implies (40) and proves the theorem.

Let us note that formula (40) coincides with Matheron's formula [12]. \square

5. Vector-matrix problem in 2D and TDT

Consider a medium described by the coupled equations (3) in the plane \mathbf{R}^2 . It is convenient to write the first two relations (3) in the matrix form

$$\begin{pmatrix} \mathbf{u} \\ \mathbf{i} \end{pmatrix} = -\mathbf{Y} \begin{pmatrix} \nabla p \\ \nabla \psi \end{pmatrix} \quad (46)$$

where

$$\mathbf{I}' = \begin{pmatrix} k & 0 & \alpha & 0 \\ 0 & k & 0 & \alpha \\ \alpha & 0 & \sigma & 0 \\ 0 & \sigma & 0 & \alpha \end{pmatrix}. \tag{47}$$

Let us introduce ‘the cut matrix’ of dimensions 2×2 which coincides with (4)

$$\mathbf{I} = \begin{pmatrix} k & \alpha \\ \alpha & \sigma \end{pmatrix}. \tag{48}$$

Then, (46) is equivalent to the relations

$$\begin{pmatrix} u_1 \\ i_1 \end{pmatrix} = -\mathbf{I} \begin{pmatrix} \frac{\partial p}{\partial x} \\ \frac{\partial \psi}{\partial x} \end{pmatrix} \quad \begin{pmatrix} u_2 \\ i_2 \end{pmatrix} = -\mathbf{I} \begin{pmatrix} \frac{\partial p}{\partial y} \\ \frac{\partial \psi}{\partial y} \end{pmatrix} \tag{49}$$

whre $\mathbf{u} = (u_1, u_2)$, $\mathbf{i} = (i_1, i_2)$.

Introduce the inverse matrix \mathbf{m}

$$\mathbf{m} := \mathbf{I}^{-1} = \frac{1}{|\mathbf{I}|} \begin{pmatrix} \sigma & -\alpha \\ -\alpha & k \end{pmatrix} \tag{50}$$

and the matrices

$$\mathbf{l}_0 := \begin{pmatrix} \langle k \rangle & \langle \alpha \rangle \\ \langle \alpha \rangle & \langle \sigma \rangle \end{pmatrix} \quad \mathbf{m}_0 := \begin{pmatrix} \langle \frac{\sigma}{|\mathbf{I}|} \rangle & -\langle \frac{\alpha}{|\mathbf{I}|} \rangle \\ -\langle \frac{\alpha}{|\mathbf{I}|} \rangle & \langle \frac{k}{|\mathbf{I}|} \rangle \end{pmatrix}. \tag{51}$$

Let us recall that $|\mathbf{I}| = k\sigma - \alpha^2$ is the determinant of \mathbf{I} . We assume that the following hypotheses are fulfilled:

- (H1) the matrices \mathbf{l}_0^{-1} and $\mathbf{m}\mathbf{m}_0^{-1}$ have the same spatial distribution law,
- (H2) the homogenized medium is isotropic.

According to (H2), the macroscopic tensor \mathbf{L} can be introduced as a matrix of dimensions 2×2

$$\mathbf{L} = \begin{pmatrix} K & A \\ A & \Sigma \end{pmatrix}. \tag{52}$$

There is a correspondence between \mathbf{I} and \mathbf{L} . For instance, K is the macroscopic permeability corresponding to the local permeability $k(x, y)$.

Theorem 2. Consider a medium satisfying hypotheses (H1) and (H2). Then,

$$\mathbf{L}^2 = \mathbf{m}_0^{-1} \mathbf{l}_0 \tag{53}$$

which is equivalent to the relation

$$\ln \mathbf{L} = \langle \ln \mathbf{I} \rangle. \tag{54}$$

The proof is parallel to Matheron’s idea [12]. Let us operate rotations by 90° and introduce the vectors

$$\mathbf{G} = (G_1, G_2) := (-u_2, u_1) \quad \mathbf{\Gamma} = (\Gamma_1, \Gamma_2) := (-i_2, i_1). \tag{55}$$

The introduced vectors \mathbf{G} and $\mathbf{\Gamma}$ are gradients, since for instance,

$$\frac{\partial G_1}{\partial y} - \frac{\partial G_2}{\partial x} = -\frac{\partial u_2}{\partial y} - \frac{\partial u_1}{\partial x} = 0 \tag{56}$$

because of the first relation (3b). Hence, there exist two potentials q and ω such that $\mathbf{G} = \nabla q$ and $\mathbf{\Gamma} = \nabla \omega$. Two vectors \mathbf{F} and $\mathbf{\Phi}$ can be defined as

$$\mathbf{F} = (F_1, F_2) := \left(-\frac{\partial p}{\partial y}, \frac{\partial p}{\partial x} \right) \quad \mathbf{\Phi} = (\Phi_1, \Phi_2) := \left(-\frac{\partial \psi}{\partial y}, \frac{\partial \psi}{\partial x} \right). \quad (57)$$

\mathbf{F} and $\mathbf{\Phi}$ are considered as conservative fluxes since they verify

$$\frac{\partial F_1}{\partial x} + \frac{\partial F_2}{\partial y} = -\frac{\partial^2 p}{\partial x \partial y} + \frac{\partial^2 p}{\partial y \partial x} = 0 \quad \frac{\partial \Phi_1}{\partial x} + \frac{\partial \Phi_2}{\partial y} = 0. \quad (58)$$

In terms of the matrix \mathbf{m} , the relations (49) become

$$\begin{pmatrix} \frac{\partial p}{\partial x} \\ \frac{\partial \psi}{\partial x} \end{pmatrix} = -\mathbf{m} \begin{pmatrix} u_1 \\ i_1 \end{pmatrix} \quad \begin{pmatrix} \frac{\partial p}{\partial y} \\ \frac{\partial \psi}{\partial y} \end{pmatrix} = -\mathbf{m} \begin{pmatrix} u_2 \\ i_2 \end{pmatrix}. \quad (59)$$

Using the vectors \mathbf{G} , $\mathbf{\Gamma}$, \mathbf{F} , $\mathbf{\Phi}$, one can write (59) in the form

$$\begin{pmatrix} F_1 \\ \Phi_1 \end{pmatrix} = -\mathbf{m} \begin{pmatrix} \frac{\partial q}{\partial x} \\ \frac{\partial \psi}{\partial x} \end{pmatrix} \quad \begin{pmatrix} F_2 \\ \Phi_2 \end{pmatrix} = -\mathbf{m} \begin{pmatrix} \frac{\partial q}{\partial y} \\ \frac{\partial \psi}{\partial y} \end{pmatrix}. \quad (60)$$

Thus, (49) and (60) can be reduced to formally identical systems

$$\begin{pmatrix} u_1 \\ i_1 \end{pmatrix} = -\mathbf{\Pi}_0^{-1} \begin{pmatrix} \frac{\partial p'}{\partial x} \\ \frac{\partial \psi'}{\partial x} \end{pmatrix} \quad \begin{pmatrix} u_2 \\ i_2 \end{pmatrix} = -\mathbf{\Pi}_0^{-1} \begin{pmatrix} \frac{\partial p'}{\partial y} \\ \frac{\partial \psi'}{\partial y} \end{pmatrix} \quad (61)$$

and

$$\begin{pmatrix} F_1 \\ \Phi_1 \end{pmatrix} = -\mathbf{m}\mathbf{m}_0^{-1} \begin{pmatrix} \frac{\partial q'}{\partial x} \\ \frac{\partial \psi'}{\partial x} \end{pmatrix} \quad \begin{pmatrix} F_2 \\ \Phi_2 \end{pmatrix} = -\mathbf{m}\mathbf{m}_0^{-1} \begin{pmatrix} \frac{\partial q'}{\partial y} \\ \frac{\partial \psi'}{\partial y} \end{pmatrix}. \quad (62)$$

Here,

$$\begin{pmatrix} p' \\ \psi' \end{pmatrix} = \mathbf{l}_0 \begin{pmatrix} p \\ \psi \end{pmatrix} \quad \begin{pmatrix} q' \\ \omega' \end{pmatrix} = \mathbf{m}_0 \begin{pmatrix} q \\ \omega \end{pmatrix}. \quad (63)$$

The operation of homogenization applied to (61) and (62) yields the macroscopic equations

$$\begin{pmatrix} \overline{u_1} \\ \overline{i_1} \end{pmatrix} = -\mathbf{L}\mathbf{l}_0^{-1} \begin{pmatrix} \overline{p_x} \\ \overline{\psi_x} \end{pmatrix}, \quad \dots, \quad \begin{pmatrix} F_2 \\ \Phi_2 \end{pmatrix} = -\mathbf{M}\mathbf{m}_0^{-1} \begin{pmatrix} \overline{q_y} \\ \overline{\omega_y} \end{pmatrix} \quad (64)$$

where the macroscopic parameters are denoted by capital letters and overbars. \mathbf{L} is the macroscopic tensor (see (52)). By construction, the tensor \mathbf{M} can be related to the original medium in the following way. First, we rotate the medium by 90° . The macroscopic properties do not change because of hypothesis (H2). Next, we invert the local tensor and introduce the macroscopic tensor \mathbf{M} for the medium described by this inverted local tensor. Hence, \mathbf{M} is simply \mathbf{L}^{-1} . Identical media have equal macroscopic properties as expressed by the relation

$$\mathbf{L}\mathbf{l}_0^{-1} = \mathbf{L}^{-1}\mathbf{m}_0^{-1}. \quad (65)$$

This is equivalent to (53).

Recall that if \mathbf{S} is a symmetric positive definite matrix, its logarithm also exists [43]. $\ln \mathbf{S}$ is the matrix with the same eigenvectors as \mathbf{S} and its eigenvalues are the logarithms of the eigenvalues of \mathbf{S} . Because of the relation $\mathbf{l}\mathbf{m}^{-1} = \mathbf{I}$, where \mathbf{I} is the identity matrix of dimension 2×2 , we have

$$\ln \mathbf{l} = -\ln \mathbf{m}. \quad (66)$$

Because of hypothesis (H1)

$$\langle \ln \mathbf{I}_0^{-1} \rangle = \langle \ln \mathbf{m} \mathbf{m}_0^{-1} \rangle \quad (67)$$

and because of (53)

$$\ln \mathbf{L} = \frac{1}{2} \ln \mathbf{I}_0 \mathbf{m}_0^{-1}. \quad (68)$$

Comparison of these two last formulae and the use of (68) yield (54).

The theorem is proved.

6. Vector–matrix case in 2D. Relation between EMT and TDT

In the present section, we extend the results of section 4 to equations (3) in 2D. The components of the matrix \mathbf{I} defined by (4) depend on k , α and σ . Let us introduce new variables k' , α' and σ' and the matrix

$$\mathbf{m} = \begin{pmatrix} k' & \alpha' \\ \alpha' & \sigma' \end{pmatrix}. \quad (69)$$

We solve equation (16) with respect to \mathbf{L} assuming that the differential $\pi(\mathbf{I}) dk d\alpha d\sigma$ is invariant under the transformation \mathbf{I} to \mathbf{m}

$$\mathbf{I} = \mathbf{L} \mathbf{m}^{-1} \mathbf{L}^{-1} \boldsymbol{\kappa}^{-1} \quad (70)$$

where $\boldsymbol{\kappa}$ is a non-degenerate constant matrix. This invariance corresponds to hypothesis (M2) for the scalar case from section 4. Invariance of the differentials under transformation (70) means that

$$\pi(\mathbf{I}) dk d\alpha d\sigma = \pi(\mathbf{m}) dk' d\alpha' d\sigma'. \quad (71)$$

Substitution of (70) into (16) and use of (71) yields

$$\int_{\Omega_{\mathbf{m}}} \pi(\mathbf{m}) (\mathbf{I} - \mathbf{m}^{-1} \mathbf{L}^{-1} \boldsymbol{\kappa}^{-1}) (\mathbf{I} + \mathbf{m}^{-1} \mathbf{L}^{-1} \boldsymbol{\kappa}^{-1}) dk' d\alpha' d\sigma' = 0. \quad (72)$$

The kernel of (72) can be written as follows:

$$(\mathbf{I} - \mathbf{m}^{-1} \mathbf{L}^{-1} \boldsymbol{\kappa}^{-1}) (\mathbf{I} + \mathbf{m}^{-1} \mathbf{L}^{-1} \boldsymbol{\kappa}^{-1}) = -(\mathbf{I} - \boldsymbol{\kappa} \mathbf{L} \mathbf{m}) (\mathbf{I} + \boldsymbol{\kappa} \mathbf{L} \mathbf{m})^{-1}. \quad (73)$$

Then, (72) becomes

$$\int_{\Omega_{\mathbf{m}}} \pi(\mathbf{m}) (\mathbf{I} - \boldsymbol{\kappa} \mathbf{L} \mathbf{m}) (\mathbf{I} + \boldsymbol{\kappa} \mathbf{L} \mathbf{m})^{-1} dk' d\alpha' d\sigma' = 0. \quad (74)$$

One can consider (16) and (74) as equations with the same structure for \mathbf{L}^{-1} and $\boldsymbol{\kappa} \mathbf{L}$. This yields the relation $\mathbf{L}^{-1} = \boldsymbol{\kappa} \mathbf{L}$ or simply

$$\mathbf{L}^2 = \boldsymbol{\kappa}^{-1}. \quad (75)$$

If $\boldsymbol{\kappa}^{-1} = \mathbf{m}_0^{-1} \mathbf{l}_0$, where $\mathbf{m}_0 := \langle \mathbf{m} \rangle$, $\mathbf{l}_0 := \langle \mathbf{l} \rangle$, we obtain a counterpart of Matheron's formula

$$\mathbf{L}^2 = \mathbf{m}_0^{-1} \mathbf{l}_0. \quad (76)$$

Hence, instead of (17a)–(17d), under the assumptions (71) and (70) one can solve the much simpler equation (76).

7. Vector–matrix case in 2D: examples and numerics

Hypothesis (H1) is formulated in a compact matrix form. Let us write it in an expanded form for the elements of the matrix. Then, (H1) implies that the following scalar values have the

same distribution laws by pairs

$$\begin{aligned}
 \frac{\langle \sigma \rangle k - \langle \alpha \rangle \alpha}{\Delta_0} & \quad \text{and} \quad \frac{\langle \frac{k}{\Delta} \rangle \sigma - \langle \frac{\alpha}{\Delta} \rangle \alpha}{\Delta \Delta_1} \\
 \frac{\langle k \rangle \alpha - \langle \alpha \rangle k}{\Delta_0} & \quad \text{and} \quad \frac{\langle \frac{\alpha}{\Delta} \rangle \sigma - \langle \frac{\sigma}{\Delta} \rangle \alpha}{\Delta \Delta_1} \\
 \frac{\langle \sigma \rangle \alpha - \langle \alpha \rangle \sigma}{\Delta_0} & \quad \text{and} \quad \frac{\langle \frac{\alpha}{\Delta} \rangle k - \langle \frac{k}{\Delta} \rangle \alpha}{\Delta \Delta_1} \\
 \frac{\langle k \rangle \sigma - \langle \alpha \rangle \alpha}{\Delta_0} & \quad \text{and} \quad \frac{\langle \frac{\sigma}{\Delta} \rangle k - \langle \frac{\alpha}{\Delta} \rangle \alpha}{\Delta \Delta_1}
 \end{aligned} \tag{77}$$

with the following notation

$$\Delta = k\sigma - \alpha^2 \quad \Delta_0 = \langle k \rangle \langle \sigma \rangle - \langle \alpha \rangle^2 \quad \Delta_1 = \left\langle \frac{k}{\Delta} \right\rangle \left\langle \frac{\sigma}{\Delta} \right\rangle - \left\langle \frac{\alpha}{\Delta} \right\rangle^2. \tag{78}$$

In order to calculate K , A and Σ of the matrix \mathbf{L} , we write the components of (53)

$$\begin{cases} K^2 + A^2 = a \\ A(K + \Sigma) = b \\ A^2 + \Sigma^2 = c \end{cases} \tag{79}$$

where

$$\begin{aligned}
 a & := \frac{1}{\Delta_1} \left(\left\langle \frac{k}{\Delta} \right\rangle \langle k \rangle + \left\langle \frac{\alpha}{\Delta} \right\rangle \langle \alpha \rangle \right) \\
 b & := \frac{1}{\Delta_1} \left(\left\langle \frac{\alpha}{\Delta} \right\rangle \langle k \rangle + \left\langle \frac{\sigma}{\Delta} \right\rangle \langle \alpha \rangle \right) \\
 c & := \frac{1}{\Delta_1} \left(\left\langle \frac{\alpha}{\Delta} \right\rangle \langle \alpha \rangle + \left\langle \frac{\sigma}{\Delta} \right\rangle \langle \sigma \rangle \right).
 \end{aligned} \tag{80}$$

Moreover, the following relation must be fulfilled in order to satisfy (53)

$$\langle \alpha \rangle \left(\left\langle \frac{\sigma}{\Delta} \right\rangle - \left\langle \frac{k}{\Delta} \right\rangle \right) = \left\langle \frac{\alpha}{\Delta} \right\rangle (\langle \sigma \rangle - \langle k \rangle). \tag{81}$$

The relations (79) can be considered as a system of equations with respect to K , A and Σ . After tedious calculations, we obtain that system (79) has a solution if and only if $pq \geq 1$, where $p := \frac{a}{b}$, $q := \frac{c}{b}$. This positive solution has the form

$$\begin{aligned}
 A & = \sqrt{\frac{b}{p+q \pm 2\sqrt{pq-1}}} \\
 K & = A(p \pm \sqrt{pq-1}) \\
 \Sigma & = (q \pm \sqrt{pq-1}).
 \end{aligned} \tag{82}$$

It is possible to check that the conditions $pq \geq 1$ and $|p+q| \geq 2\sqrt{pq-1}$ are always fulfilled.

Let us consider a numerical example of a correlated medium where all local parameters k , α , σ depend on the porosity ϵ [27]

$$k = k_0 \epsilon^m \quad \sigma = \sigma_0 \epsilon^n \quad \alpha = \alpha_0 \epsilon^m. \tag{83}$$

We assume that the porosity ϵ satisfies the cut lognormal distribution determined by the probability density

$$f(\epsilon) = \frac{1}{c_0} \frac{1}{\sqrt{2\pi} \sigma^* \epsilon} \exp\left(-\frac{\ln \epsilon - \ln \epsilon_0}{2\sigma^{*2}}\right) \tag{84}$$

where ϵ_0 is the expected porosity; σ^* is the standard deviation. The constant c_0 is chosen in such a way that

$$\int_0^1 f(\epsilon) d\epsilon = 1. \tag{85}$$

Following [27], let us put (in SI units)

$$m = 3.75 \quad n = \frac{2}{3}m \quad \epsilon_0 = 0.3 \quad k_0 = 10^{-12} \quad \alpha_0 = 3 \times 10^{-9} \quad \sigma_0 = 10^{-2}. \tag{86}$$

Then, (82) yields the effective constants

$$K = 9.17642 \times 10^{-15} \quad A = 1.64201 \times 10^{-18} \quad \Sigma = 0.000461544. \tag{87}$$

Hypothesis (H1) assumes that the matrices $\mathbf{\Pi}_0^{-1}$ and $\mathbf{m}\mathbf{m}_0^{-1}$ have the same spatial distribution law. Instead of (H1), we can consider three other cases, when the following matrices have the same distribution laws by pairs

$$\mathbf{l}_0^{-1}\mathbf{l} \text{ and } \mathbf{m}\mathbf{m}_0^{-1} \quad \mathbf{\Pi}_0^{-1} \text{ and } \mathbf{m}_0^{-1}\mathbf{m} \quad \mathbf{\Pi}_0^{-1}\mathbf{l} \text{ and } \mathbf{m}_0^{-1}\mathbf{m}. \tag{88}$$

For instance, if $\mathbf{\Pi}_0^{-1}$ and $\mathbf{m}_0^{-1}\mathbf{m}$ have the same distribution law, we obtain

$$\mathbf{L}\mathbf{l}_0^{-1}\mathbf{l} = \mathbf{m}_0^{-1}. \tag{89}$$

Let us write (89) by elements

$$\begin{cases} \langle \sigma \rangle K^2 + 2\langle \alpha \rangle AK + \langle k \rangle A^2 = a_1 \\ \langle \sigma \rangle AK + \langle k \rangle A\Sigma - \langle \alpha \rangle (A^2 + K\Sigma) = b_1 \\ \langle \sigma \rangle A^2 - 2\langle \alpha \rangle A\Sigma + \langle k \rangle \Sigma^2 = c_1 \end{cases} \tag{90}$$

where

$$\begin{aligned} a_1 &:= \frac{\Delta_0}{\Delta_1} \left\langle \frac{k}{\Delta} \right\rangle \\ b_1 &:= \frac{\Delta_0}{\Delta_1} \left\langle \frac{\alpha}{\Delta} \right\rangle \\ c_1 &:= \frac{\Delta_0}{\Delta_1} \left\langle \frac{\sigma}{\Delta} \right\rangle. \end{aligned} \tag{91}$$

One can consider (90) as a system of algebraic equations with respect to K, A, Σ . Let us note that there is no additional condition like (81) in this case. We do not know a solution of (90) in explicit form, but it can be easily found by computer.

8. Conclusion

Electro-osmotic phenomena in porous media are governed by the coupled equations (3). Following Bruggeman, we applied the effective medium theory for (3) by embedding a single circular (spherical) inclusion in a medium whose tensor \mathbf{L} is the unknown macroscopic property. Using a self-consistent approach, we deduced the relations (15), (16) and (21) for \mathbf{L} in 2D; and the relations (26), (27) and (28) in 3D.

Following Matheron, we also developed the theory of duality transformations for equations (3) in 2D. We imposed conditions on the statistical properties of the local coefficients of (3) and deduced the exact formulae (53), (54) for the macroscopic tensor \mathbf{L} .

We proved that the effective tensors obtained by both methods (EMT and TDT) in 2D under conditions (H1) and (H2) give the same result.

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