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Connection between electrical conductivity and diffusion coefficient of a conductive porous material filled with electrolyte

I. Sevostianov[a,b,*], A. Trofimov[b], J. Merodio[c], R. Penta[c,d], R. Rodriguez-Ramos[e]

[a] *Department of Mechanical and Aerospace Engineering, New Mexico State University, Las Cruces, NM 88001, USA*
[b] *Center for Design, Manufacturing, and Materials, Skolkovo Institute of Science and Technology, Skolkovo, Russia*
[c] *Departamento de Mecanica de los Medios Continuos y T. Estructuras, Universidad Politecnica de Madrid, Madrid, 28040, Spain*
[d] *School of Mathematics & Statistics, University of Glasgow, Glasgow G12 8SQ, UK*
[e] *Facultad de Matematica y Computacion, Universidad de La Habana, La Habana, CP 10400, Cuba*

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ABSTRACT

The paper focuses on the cross-property connection between the effective electrical conductivity and the overall mass transfer coefficient of a two phase material. The two properties are expressed in terms of the tortuosity parameter which generalized to the case of a material with two conductive phases. Elimination of this parameter yields the cross-property connection. The theoretical derivation is verified by comparison with computer simulation.

1. Introduction

The practical implementation of homogenization schemes to calculate the overall properties of heterogeneous materials often requires information that is not available. The accurate predictions are based on the knowledge of the properties of constituents, relative volume functions, and parameters characterizing the microstructure. While material properties of the constituents and their volume fractions are generally known, information about morphology of the material may be incomplete or inappropriate. This information, however, may be reconstructed by measuring the set of properties different from ones of interest and using cross-property connections.

Existence of cross-property connections has been recognized first from the observations of qualitative nature. For instance, geophysicists noticed that cracks in rocks increase both the elastic compliance and the fluid permeability; in fracture mechanics, attempts have been made to relate the loss of elastic stiffness of a deteriorating microstructure to lifetime predictions. Quantitative theoretical results on cross-property connections started to appear in 1950s. In works of Klinkenberg (1951), Wyllie and Rose (1950), and Wyllie and Spangler (1952) method of evaluation of hydraulic conductivity of porous rock through electrical conductivity measurements has been developed. Pores were filled with electrolyte and the solid skeleton of the porous material was considered as a perfect electrical insulator impenetrable for the liquid. Bristow (1960) derived explicit connection between elastic constants and electrical conductivity for a material containing multiple randomly crack orientated cracks. Levin (1967) interrelated the effective bulk modulus and the

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[*] Corresponding author at: Department of Mechanical and Aerospace Engineering, New Mexico State University, Las Cruces, NM 88001, USA.
E-mail address: igor@nmsu.edu (I. Sevostianov).

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effective thermal expansion coefficient of a two phase isotropic composite. Prager (1969) constructed Hashin–Shtrikman-based bounds for the effective magnetic permeability (or electrical conductivity) in terms of the effective thermal conductivity of a two-phase isotropic material. Later, many results have been obtained on correlation between linear elastic and conductive (thermal or electrical) properties of heterogeneous materials. Cross-property bounds have been obtained by Milton (1984) and Gibiansky and Torquato (1995, 1996a, b); explicit approximate connections have been derived by Sevostianov and Kachanov (2002) (see also Sevostianov, 2003; and review Sevostianov & Kachanov, 2009). Exact connections between normal compliance and spreading resistance of two contacting surfaces have been developed by Barber (2003), Sevostianov and Kachanov (2004) and Sevostianov (2010). Connections between electrical conductivity and fluid permeability of a porous material have been developed by Torquato (1990) and Avellaneda and Torquato (1991) under assumption that the solid skeleton does not conduct electricity and the physical properties are refer to the porous space filled with electrolyte (such a situation is typical, for example, for geophysical applications). *Generally speaking, cross-property connections can be developed if microstructural parameters controlling two physical properties are either the same or similar* (see review of Sevostianov & Kachanov, 2009).

Another possibility appears when the properties of interest are governed by *supplemental parameters*, like in the case of elastic properties and electrical conductivity of saturated rock, where the latter is determined by microgeometry of the porous space and elastic properties are controlled by the morphology of the solid phase. Berryman and Milton (1988) derived variational cross-property bound for such a case. Sevostianov and Shrestha (2010), using results of Sevostianova, Leinauer, and Sevostianov (2010), derived connections between fluid permeability of a porous material and electrical conductivity through the solid skeleton. They developed variational bounds and explicit closed-form connection between the two physical properties. Their results were numerically verified by Garsia and Sevostianov (2012)

In the present paper, we consider a porous material with electrically conductive skeleton. The pores are assumed to be filled with electrically conductive liquid. Diffusion of a substance of interests is possible in the liquid as well as in the skeleton. The paper focuses on the problem of the evaluation of the overall mass transfer coefficient of such a material from the electrical conductivity measurements. The derivation is based on the elimination of the microstructural parameter – tortuosity of the porous space – that governs both the properties. Analytical derivations are compared with FEM calculations.

2. Tortuosity as a microstructural parameter in the context of mass transfer and electrical conductivity: the concept and the history of terminology

The electrical and mass transport performances of any porous material are strongly dependent on their three-dimensional (3D) microstructures, which include the porosity, pore sizes and shapes and connectivity of the porous space. These microstructural parameters can be collectively described as the “tortuosity of the porous space” (see, for example, Chen, Wang, Giuliani, & Atkinson, 2013).

The term tortuosity, to the best of our knowledge, has been first introduced by Thomson and Tait (1879) in the context of curvature of a non-plane curve (see Sections 7–9 of their book). Noting $\delta\phi$ the angle between the osculating planes at two points at a distance δs from one another along the curve, they defined tortuosity τ of a curve as a derivative

$$\tau_{T-T} = \frac{d\phi}{ds} \tag{2.1}$$

(we use subscript “T-T” to identify definition of Thomson and Tait). In the beginning of XX century term tortuosity was adopted in medicine to describe (qualitatively) spatial curvature of blood vessels (see, for example Edington (1901) or Cairney (1924)). Later, Carman (1939) suggested to use this term to describe curvilinear character of porous space in the context of hydraulic conductivity. Carman defined it as the ratio of the effective length (L_e) of the fluid flow path to the apparent length (L_a) of a specimen:

$$\tau_C = L_e/L_a \tag{2.2}$$

(subscript C stays for definition of Carman).

Due to obvious uncertainty of this definition and difficulties associated with its practical implications, Winsauer, Shearin, Masson, and Williams (1951), Wyllie and Rose (1950), and Cornell and Katz (1953) suggested to use different kind of tortuosity τ related to the resistivity factor F (sometimes this factor is also called electrical formation factor or electrical retardation factor):

$$F \equiv k^0/k^{eff} = \tau/\psi, \tag{2.3}$$

where k^{eff} is the bulk (effective) electrical conductivity of the porous material completely saturated by electrolyte, k^0 is the conductivity of the electrolyte, and ψ is the ratio of the apparent cross-sectional area of the conducting electrolyte to the total cross-sectional area of the specimen. This ratio, however, varies from one cross-section to another and it is unclear, which value is appropriate – maximum, minimum, average or anything else (see Sevostianova et al., 2010).

Perkins, Osoba, and Ribe (1956) suggested an experimental procedure to estimate tortuosity τ and showed that, for completely saturated sandstone, the resistivity factor, tortuosity and porosity p are interrelated by