Effective conductivity by fluid analogy for a porous insulator filled with a conductor

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By combining identities relating effective conductivity to tortuosity and tortuosity to induced mass, a general formula for the effective (electrical or thermal) conductivity of a porous insulator filled with a conductor is obtained. This formula depends on an induced-mass factor which arises by treating the conducting material as an inviscid fluid. This induced-mass factor can be estimated with the use of an effective-medium theory. For random arrays of equal spheres, the estimates of conductivity obtained with the use of this fluid analogy are in good agreement with recent exact values derived for periodic arrays of insulating spheres to closest packing.

Recently[1-3] an important link has been established between the effective electrical conductivity σ^* of a porous insulator saturated with a conducting fluid and a geometrical structure factor α (also known[3,4] as the "electric tortuosity") depending on the topology of the interconnected pore space. Another identity relating this tortuosity to the induced mass[5] of the solid particles oscillating in the presence of the pore fluid had been noted and exploited previously.[6] In this Brief Report, we combine these two identities to produce a comparatively simple formula for the effective (electrical or thermal) conductivity of a porous insulator filled with a conductor. The conductor need not be a fluid but, while developing the fluid analogy, we will often speak of the conductor as if it were an inviscid, incompressible fluid.

The identity relating effective conductivity and tortuosity which was established first theoretically[1,2] and then confirmed experimentally[3] is

$$\sigma^*/\sigma = \phi/\alpha \ , \tag{1}$$

where σ and ϕ are, respectively, the conductivity and volume fraction of the conducting material. We assume that all of the void space of the insulator is interconnected and filled with conductor so ϕ is the porosity of the insulator.

In a porous material saturated with fluid, the macroscopic kinetic energy per unit volume can be written (following Biot[7]) as

$$2T = \rho_{11}\vec{U}\cdot\vec{U} + 2\rho_{12}\vec{U}\cdot\vec{V} + \rho_{22}\vec{V}\cdot\vec{V} \ , \tag{2}$$

where $\vec{U}$ is the (local) average velocity of the solid (isotropic) porous frame, $\vec{V}$ is the average velocity of the pore fluid, and the densities ρ_{11}, ρ_{12}, ρ_{22} are parameters to be determined. The coefficient ρ_{22} is well known[1,2,7] to be related to α by

$$\rho_{22} = \alpha\phi\rho_f \ , \tag{3}$$

where ρ_f is the fluid density. Equation (3) has been used in the derivation[1,2] of (1).

When the frame is stationary $\vec{U}=0$ and the fluid flows through it with local (microscopic) velocity $\vec{v}$, the kinetic energy has the form

$$2T = \frac{\rho_f}{\Omega}\int_{\Omega_f}\vec{v}\cdot\vec{v}\,d^3x = \rho_{22}\vec{V}\cdot\vec{V} \ , \tag{4}$$

where Ω_f is the fluid volume and Ω is the total volume being considered. Defining $\vec{v}=\vec{V}+\Delta\vec{v}$ such that

$$\int_{\Omega_f}\Delta\vec{v}\,d^3x = 0 \ , \tag{5}$$

we find

$$2T = \rho_f\phi V^2 + \frac{\rho_f}{\Omega}\int_{\Omega_f}\Delta\vec{v}\cdot\Delta\vec{v}\,d^3x \ , \tag{6}$$

since $\Omega_f/\Omega=\phi$.

For an isolated particle in an inviscid, incompressible fluid, the integral corresponding to the one in (6) is defined[5] as

$$\rho_f\int_{\Omega_f}\Delta\vec{v}\cdot\Delta\vec{v}\,d^3x = m_{ij}^{(0)}V_iV_j \ , \tag{7}$$

where the summation convention has been assumed. The induced-mass tensor $m_{ij}^{(0)}$ has the general form

$$m_{ij}^{(0)} = r_{ij}^{(0)}\Omega_0\rho_f \ , \tag{8}$$

where $\Omega_0\rho_f$ is the mass of the displaced fluid and r_{ij} is a dimensionless tensor of order unity ($r_{ij}=\frac{1}{2}\delta_{ij}$ for spherical particles). When many particles are present, (7) becomes

$$\frac{1}{\Omega}\int_{\Omega_f}\Delta\vec{v}\cdot\Delta\vec{v}\,d^3x = \frac{1}{\Omega}\sum_n r_{ij}^{(n)}\Omega_n V_iV_j \ . \tag{9}$$

By noting that $\sum_n\Omega_n=(1-\phi)\Omega$ and by assuming that the porous medium is isotropic on the macro-

scopic scale, we can define a scalar induced-mass factor r for such a medium by

$$\frac{1}{\Omega}\sum_n r_{ij}^{(n)}\Omega_n = r(1-\phi)\delta_{ij} \ . \tag{10}$$

Combining (4), (6), (9), and (10), we find directly that

$$\rho_{22} = [\phi + r(1-\phi)]\rho_f \ . \tag{11}$$

The coefficient ρ_{22} is related to α by (3) so we find, in general, that

$$\alpha = 1 + r(\phi^{-1} - 1) \ . \tag{12}$$

Equation (12) was derived previously[6] in a less direct fashion. We present this new derivation to stress the generality of the result. In particular, the validity of Eq. (12) is not dependent on the validity of Biot's macroscopic equations[7] for elastic waves in fluid-saturated porous media. It therefore follows from (1) and (12) that the equation

$$\sigma^*/\sigma = \phi^2/[\phi + r(1-\phi)] \tag{13}$$

is a rigorous formula for the effective conductivity in terms of the induced-mass factor r.

By considering known rigorous bounds on σ^*, we can learn something about the behavior of the induced-mass factor. Two simple rigorous upper bounds on σ^* are the Hashin-Shtrikman upper bound[8]

$$\sigma^*/\sigma \leq 2\phi/(3-\phi) \tag{14}$$

and the Miller upper bound[9] for symmetric-cell materials with spherical cells

$$\sigma^*/\sigma \leq 2\phi^2/(1+\phi) \ . \tag{15}$$

The corresponding lower bounds vanish in both cases for all $\phi < 1$. These inequalities can be translated into bounds for r. We find

$$\tfrac{1}{2}\phi \leq r \leq \infty \ \text{(Hashin–Shtrikman)} \tag{16}$$

and

$$\tfrac{1}{2} \leq r \leq \infty \ \text{(Miller)} \ . \tag{17}$$

It is interesting to note that setting $r = \frac{1}{2}$ (the value for isolated spheres), as was done in Ref. 6, leads to the Miller bound (15) for spherical-cell materials. Real materials will not generally be symmetric-cell materials so we will leave further discussion of the Miller bounds to a future publication.

A simple interpretation can be given to the Hashin-Shtrikman bound (16). Suppose we imagine imbedding one new sphere in a fluid already containing a finite concentration of particles. For random placement of the sphere, the fluid displaced by the new particle will, on the average, have density given approximately by $\phi\rho_f$ since the other particles act like holes in the fluid. Thus the induced mass of the new sphere is one-half the mass of the displaced fluid or

$$r\rho_f\Omega_0 \simeq \tfrac{1}{2}\phi\rho_f\Omega_0 \ , \tag{18}$$

so

$$r \simeq \tfrac{1}{2}\phi \ , \tag{19}$$

which is just the Hashin-Shtrikman lower bound on r.

The preceding argument gives a value of r which is generally too low—actually achieving the lowest allowable value of r. To improve this argument, we must answer the question "What is the correct effective density of the displaced fluid?" However, our previous discussion has already at least partially answered this question. The effective macroscopic density of the fluid is given by ρ_{22} in (11). This density ρ_{22} has two terms: the first term corresponds to the fluid with holes and the second term is the backflow contribution. Thus, if we ignore details like the precise location or distribution of the other particles, an elementary effective-medium argument suggests that, for a spherical inclusion,

$$r\rho_f = \tfrac{1}{2}\rho_{22} \ . \tag{20}$$

For particles which are not spherical in shape, the factor $\frac{1}{2}$ must be replaced by the appropriately averaged factor (e.g., $\frac{2}{3}$ for randomly oriented needles[2]). However, we will restrict our present discussion to spherical particles.

Substituting (11) into (20) and solving for r, we find that

$$r = \phi/(1+\phi) \tag{21}$$

and

$$\sigma^*/\sigma = \tfrac{1}{2}\phi(1+\phi) \ . \tag{22}$$

To check the accuracy of (22), we first consider low-concentration expansions. Define the packing fraction η by $\eta = 1-\phi$ and note that Eq. (22) is

$$\sigma^*/\sigma = 1 - \tfrac{3}{2}\eta + \tfrac{1}{2}\eta^2 \tag{23}$$

in terms of η. For periodic arrays of spheres, it is known[10] that

$$\sigma^*/\sigma = 1 - \tfrac{3}{2}\eta + \tfrac{3}{4}\eta^2 + O(\eta^3) \ . \tag{24}$$

Jeffrey[11] has shown that

$$\sigma^*/\sigma = 1 - \tfrac{3}{2}\eta + 0.588\eta^2 + O(\eta^3) \tag{25}$$

for random arrays of equal spheres at low concentrations if the distribution of spheres satisfies the well-stirred approximation (the probability of finding a second particle center is zero inside and constant outside the first particle's exclusion sphere). McCoy and

Beran[12] have shown that the coefficient of η^2 depends on the choice of distribution, and it is not difficult to show that a distribution with higher probability of finding another particle close to a particle at the origin will lead to a smaller coefficient of η^2 than the one given by Jeffrey. For the hard-sphere radial distribution function,[13] corrections to Jeffrey's coefficient are of $O(\eta)$ for small η and therefore do not alter the result (25). However, the sign of these corrections again tends to decrease the contribution from the second-order term.

Thus, for low particle concentrations, Eq. (23) gives a good approximation to the exact results for random arrays to second order in η. It is not surprising that (23) does not agree with (24) to second order since the effective-medium approach does not assume the particles are "well separated" (no particle center within a radius $R/\eta^{1/3}$ of a given particle when R is the radius of the particles) as is the case for the periodic arrays.

For large particle concentrations, exact results are known[10] for periodic arrays of spheres but not for random arrays. However, it is a common supposition that results for random arrays of spheres should not differ too much from results for periodic arrays at high concentrations. In Table I, comparison is made between recent exact results of Sangani and Acrivos[10] for periodic arrays and the predictions of (23) for random arrays of spheres. Agreement between the two sets of values is quite good.

Although the formula (13) is a rigorous identity for σ^*, its usefulness is limited in at least three ways. First, the formula applies only to porous *insulators*. If the porous frame is also a conductor, the present approach for calculating conductivity does not apply because Eq. (1) is no longer true. However, even if the frame is conducting, the formula (12) for the tortuosity α (important for Biot's theory[7] as has been amply shown elsewhere[3,6]) is still valid. Second, if some of the pore space is isolated from the connected pore space, the relevant value of ϕ (i.e., the volume fraction of the connected pore space) will be more difficult to measure. Except for measuring ϕ, the presence of isolated pores will not affect the present arguments as long as the porous frame is an insulator since isolated pores (whether filled with conductor or not) are completely surrounded by insulator and, hence, cannot contribute to the conductivity.

A third limitation of formula (13) is that the induced-mass factor r may not be especially easy to calculate for arbitrary shapes and arrangements of particles. The factor r may nevertheless prove to be a useful tool for correlating experimental data. For example, the tortuosity data of Johnson *et al.*[3] for sintered packings of glass spheres have been reduced to the equivalent induced-mass factor and plotted in Fig. 1. We see that most of these data fall in the range $r = 0.35 \pm 0.05$. No explanation of this virtually constant value of r can be offered at the present time.

We conclude that the fluid analogy provides both a

TABLE I. Comparison of the values of effective conductivity σ^*/σ for random arrays of spheres estimated using Eq. (23) and for periodic arrays of spheres calculated by Sangani and Acrivos (Ref. 10). The four values of packing fraction η correspond to the three close-packed periodic arrays—simple cubic ($\eta = \pi/6$), body-centered cubic ($\eta = \sqrt{3}\pi/8$), face-centered cubic ($\eta = \sqrt{2}\pi/6$),—and to random close packing for hard spheres (Ref. 13) ($\eta = 0.64 \pm 0.02$). The value of σ^*/σ quoted in parentheses for periodic arrays at $\eta = 0.64$ was obtained by interpolation from the exact results for sc and bcc arrays.

	σ^*/σ	
$\eta = 1-\phi$	Random arrays of spheres	Periodic arrays of spheres
0.5236	0.352	0.344
0.64	0.245	(0.250)
0.6802	0.211	0.217
0.7405	0.163	0.160

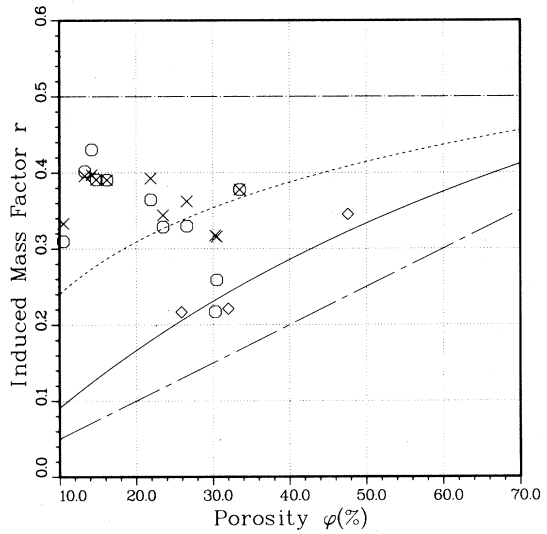

FIG. 1. Induced-mass factor as a function of porosity for various models. Dash–long-dash line is the Hashin-Shtrikman lower bound (16). Dot-dash line is the Miller lower bound (17) for spherical-symmetric–cell materials. Solid line is Eq. (21) for random arrays of spheres. Diamonds are exact results for periodic arrays of spheres from Sangani and Acrivos (Ref. 10) (also see Table I). Data [courtesy of Johnson *et al.* (Ref. 3)] from measured tortuosity of porous structures fabricated by sintering packings of glass beads are shown as crosses for superfluid acoustic measurements and octagons for electrical measurements. The dashed line is for the self-similar estimate (Refs. 2 and 14) for effective conductivity $\sigma^*/\sigma = \phi^{3/2}$.

rigorous new identity satisfied by σ^* for porous insulators and also a surprisingly simple method of estimating the induced-mass factor for random arrays of spheres. More detailed calculations including corrections to (20) and comparisons to other theories are planned to appear in a later publication.

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