

A DIFFERENTIAL SCHEME FOR THE EFFECTIVE MODULI OF COMPOSITES

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A differential scheme to compute the effective moduli of composites is presented. The method is based on the idea of realizability, i.e. the composite is constructed explicitly from an initial material through a series of incremental additions. The construction process is uniquely specified by parametrizing the volume fractions of the included phases. The properties of the final composite depend upon the construction path taken and not just on the final volume fractions. Assuming the grain shapes are ellipsoidal, a system of ordinary differential equations for the moduli is obtained which is integrated along the path. The present method includes as special cases of paths or endpoints the differential scheme of Roscoe-Boucher and the self-consistent scheme of Kroner-Hill, respectively. The method includes a realization of the Hashin-Shtrikman bounds for a two-phase composite with $(\kappa_1 - \kappa_2)(\mu_1 - \mu_2) \geq 0$. For example, the upper bounds are achieved by imbedding disks of the stiffer material in a matrix of the more compliant material.

1. Introduction

The subject of this paper is the mechanical properties of composites. The composites are assumed to consist of discrete homogeneous 'grains' which are perfectly bonded together. The grain structure can be such that one phase is suspended in a matrix of another phase, or the two phases can be distributed in a symmetric fashion. The grains can be aligned, leading to anisotropy, or randomly oriented, to give an isotropic composite. In general, any number of distinct phases can be considered. A similar method for estimating the electrostatic properties of composites has been discussed by Norris, Callegari and Sheng (1984).

Various methods have been proposed over the years for computing the macroscopic effective moduli of multi-phase elastic composites. Rigorous procedures exist for composites with periodic microstructure (Nemat-Nasser and Taya, 1981; Nemat-Nasser et al., 1982; Tao and Sheng, 1984; Nunan and Keller, 1984). However, exact solu-

tions are of little use when the underlying structure is random. In this case it is common to use one of several 'effective medium theories'. Foremost among these methods has been the symmetric self-consistent scheme of Kroner (1958). This method is analogous to the effective medium approximation (EMA) used in electrostatics and due originally to Bruggeman (1935). Hill (1965b) examined the Kroner scheme for spherical grains and found it to be consistent with the Hashin-Shtrikman bounds for two-phase isotropic composites. However, another class of 'self-consistent' theories proposed by Wu (1966), Walpole (1969) and Boucher (1974) differ from Hill's formulation in that the two phases are not treated on a symmetric basis in the final equations. Both of these schemes are identical for spherical grains, a fact which has led to some confusion in distinguishing between them. The distinctions between these theories and others are discussed in detail by Koringa et al. (1979) and by Berryman (1980). Good reviews of the relevant literature can be found in Watt, Davies and O'Connell (1976), Cleary, Chen and Lee (1980), Christensen (1979) and Hashin (1983); see also Budiansky (1970). In this paper we consider only the self-consistent scheme of Kroner

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and Hill, which we refer to as EMA to avoid ambiguity. The EMA has also been derived in the context of scattering theory by Korringa (1973), Gubernatis and Krumhansl (1975), and by Berryman (1980). The key in all the scattering theory approaches is to imbed typical grains of either phase in a homogeneous matrix with unknown elastic moduli. The moduli are then obtained by requiring the averaged forward scattered field for the ensemble of grains to equal zero. In the low frequency or quasistatic limit this is equivalent to the EMA.

The second class of effective medium theories considered here is due originally to Roscoe (1954) and Boucher (1975). This is the differential effective medium theory (DEM), which unlike the EMA, does not treat the phases on an equal footing. In a two-phase composite one phase is taken as the matrix and the other is added incrementally in such a way that the newly added material is always in dilute concentration with respect to the current effective medium. The microgeometry corresponding to DEM has been described by Sen, Scala and Cohen (1981) and also by Sheng and Callegari (1984). McLaughlin (1977) has shown that the DEM theory for spherical grains gives moduli that satisfy the Hashin-Shtrikman bounds. A good review of the elastic DEM theory is contained in the article by Cleary, Chen and Lee (1980). DEM has received attention recently because it predicts the empirical Archie's law for the effective conductivity of rocks (Sen, Scala and Cohen, 1981; Sheng and Callegari, 1984). Other differential schemes based on the idea of iterating the dilute approximation have been proposed by Henyey and Pomphrey (1982) for cracked solids and by Zimmerman (1984a and b) with spherical pores. The method of Zimmerman predicts moduli that are inconsistent with the Hashin-Shtrikman bounds and so cannot correspond to a physically realizable solid.

The purpose of this paper is to develop a theory which contains both DEM and EMA as special cases. The key to our approach is the idea of realization. An effective medium theory is called realizable if we can describe a construction process to make a composite material with effective moduli predicted by the theory. Thus, the Hashin

-Shtrikman bounds for the bulk modulus of a two-phase randomly disordered solid can be realized through the well known packed-spheres geometry (e.g. (Hashin, 1983)). There were very few other realizable geometries known until recently. A major breakthrough came when Milton (1984a and b) showed that EMA corresponds to a class of aggregates with well defined microgeometries. For a two phase composite these aggregates are constructed briefly as follows: one starts with an arbitrary homogeneous matrix material with moduli $L^{(0)}$. Then grains of the two phases are imbedded in the matrix such that: (1) the grains are in a dilute suspension, and (2) the relative volume fractions of the two phases are the same as in the final material. This results in an effective medium with moduli $L^{(1)}$. The process is repeated with material $L^{(1)}$ as the matrix and with grains which are an order of magnitude larger in size than the previous grains. This results in an effective medium with moduli $L^{(2)}$. The iteration is continued such that at each stage J the imbedded grains are in dilute concentration and the required volume fractions are satisfied. The imbedded grains at stage J are much larger in size than those of stage $J-1$ and the imbedded grains are sufficiently separated so that grain-grain interactions are negligible. Milton (1984b) showed that the EMA result is obtained by letting $J \rightarrow \infty$ and taking certain other limits. We refer to the original paper for more details.

A similar type of hierarchy of aggregates can be defined for DEM. For a two-phase composite of materials 1 and 2 we start with a matrix of phase 2, for example, and imbed grains of phase 1 in dilute concentration. The next stage consists of imbedding grains of phase 1 that are an order of magnitude larger than the previous ones, and so on. The DEM hierarchy of composites continues until phase 1 occupies its correct volume fraction. In this sense it is different from EMA, where the original matrix material is completely replaced. The DEM composite is unsymmetric in the two phases by definition. If material 2 has no rigidity ($\mu_2 = 0$) then the DEM material has no rigidity because the added phase, which has rigidity, is not connected. However, the EMA result for the two phase composite has rigidity when the volume

fraction of the solid phase exceeds a certain value, the rigidity threshold, which depends upon the grain shapes considered (Hill, 1965b).

2. The construction process

In this section we describe a procedure whereby the generalized theory can be realized. The process considered here is just one of many that could be envisaged. It is characterized by keeping the volume of the composite fixed at V_0 during the entire process. An alternative process in which the volume increases is described briefly in Section 3. However, it turns out that the differential equations we obtain in Section 3 are independent of the construction process.

Begin with volume V_0 of material 0, an anisotropic, homogeneous, linearly elastic solid with elastic moduli tensor L_0 , which we refer to as the 'backbone' material. Grains of materials 1 and 2, are imbedded in material 0 in such a way that the volume remains fixed at V_0 . This involves removing and replacing some of the original material. Materials 1 and 2 are also anisotropic, homogeneous, linearly elastic solids with moduli L_1 and L_2 respectively. The composite now has a new modulus tensor L , different from L_0 .

The construction process continues by removing the current material and replacing it with grains of materials 1 and 2. Let ϕ_0 , ϕ_1 and ϕ_2 be the current volume fractions of the materials, such that

$$\phi_0 + \phi_1 + \phi_2 = 1. \quad (1)$$

At each replacement, the removed material must have the same volume fractions of materials 0, 1 and 2 as the total volume. Thus, at each stage the material is assumed to be homogeneous. This can be realized if the replacement grains are always an order of magnitude greater in size than those at the previous removal-replacement. In addition, the grains must be dispersed at random and occupy an infinitesimal volume fraction. A schematic of the iterative process is shown in Fig. 1.

The construction process is uniquely defined by a path in the (ϕ_1, ϕ_2) plane, see Fig. 2. Let the path be parametrized by an arc length parameter t , which is analogous to time, so that on the path we have $\phi_1 = \phi_1(t)$, $\phi_2 = \phi_2(t)$. The origin of time is conveniently chosen so that $\phi_1(0) = \phi_2(0) = 0$. It then follows that for each $t \geq 0$ the material is homogeneous, i.e. any given volume v of the composite contains $\phi_2 v$ of material 2.

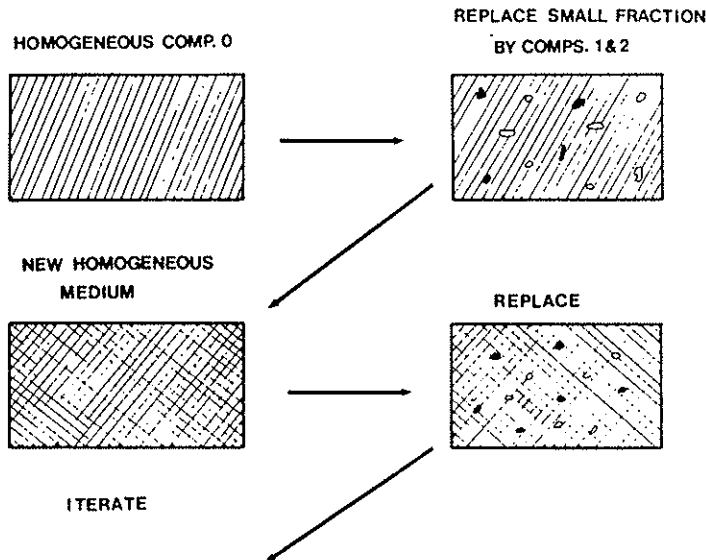


Fig. 1. The iterative construction process of the general theory.

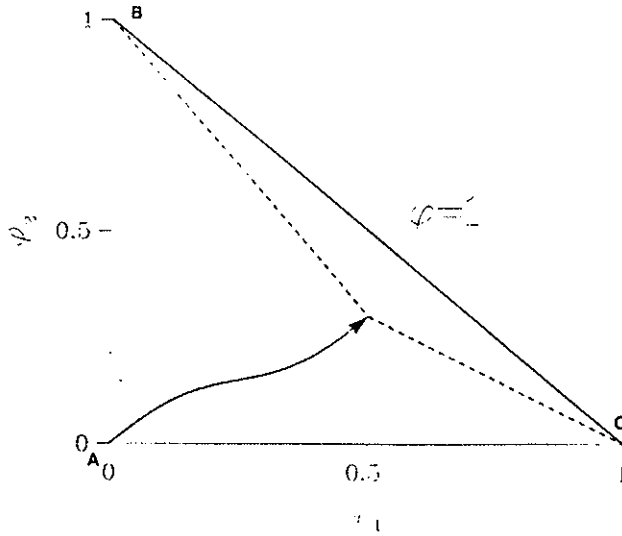


Fig. 2. A homogenization path in the (ϕ_1, ϕ_2) plane. The range of further possible points is the small triangle.

We now examine the incremental removal-replacement at time t . Let the volume of material removed by Δv . The same volume is replaced with volume Δv_1 of material 1 and volume Δv_2 of material 2 such that $\Delta v_1 + \Delta v_2 = \Delta v$. The volume of material 1 in the solid before the replacement was $\phi_1 V_0$. After replacement, the volume of material 1 is

$$V_0(\phi_1 + \Delta\phi_1) = V_0\phi_1\left(1 - \frac{\Delta v}{V_0}\right) + \Delta v_1. \quad (2)$$

This relates the increment in ϕ_1 to the corresponding increments in v_1 and v_2 . A similar relation follows for $\Delta\phi_2$. Note that v_1 and v_2 measure the total volumes of materials 1 and 2, respectively, that have been added during the construction process up to the current value of t . Therefore v_1 and v_2 describe a path in the (v_1, v_2) plane as t varies. We can parametrize this path by t such that $v_1 = v_1(t)$ and $v_2 = v_2(t)$ along the path. Define the derivative $\dot{\phi}_1$ as

$$\dot{\phi}_1(t) = d\phi_1/dt. \quad (3)$$

Similarly, we define the derivatives $\dot{\phi}_2$, $\dot{v}_1$ and $\dot{v}_2$ so that (2) becomes

$$V_0\dot{\phi}_1 = (1 - \phi_1)\dot{v}_1 - \phi_1\dot{v}_2.$$

This relation and the corresponding one for $\dot{\phi}_2(t)$

can be inverted to give $\dot{v}_1$ and $\dot{v}_2$ as

$$\begin{aligned} \frac{\dot{v}_1}{V_0} &= \frac{(1 - \phi_2)\dot{\phi}_1 + \phi_1\dot{\phi}_2}{1 - \phi}, \\ \frac{\dot{v}_2}{V_0} &= \frac{(1 - \phi_1)\dot{\phi}_2 + \phi_2\dot{\phi}_1}{1 - \phi} \end{aligned} \quad (4)$$

where $\phi \equiv \phi_1 + \phi_2$. Thus, the construction process can be described by a path in the (ϕ_1, ϕ_2) plane or its equivalent path in the (v_1, v_2) plane.

Paths in either plane begin at the origin. The range of v_1 and v_2 is the quarter space $v_1 \geq 0$, $v_2 \geq 0$. It may seem paradoxical that v_1 and v_2 can become as large as we wish while the volume of composite remains fixed at V_0 . However, v_j , $j = 1$ or 2 does not represent the current volume of material j in the composite, but the volume of material j that has been used up in the removal-replacement process.

The paths in the (ϕ_1, ϕ_2) plane are restricted to the triangle $\phi_1 \geq 0$, $\phi_2 \geq 0$ and $\phi \leq 1$. A further restriction follows from the physical requirement that $\dot{v}_1$ and $\dot{v}_2$ are both positive, meaning materials 1 and 2 are always added, not taken out. This implies, using (4) and the identity

$$\frac{(1 - \phi_2)\dot{\phi}_1 + \phi_1\dot{\phi}_2}{(1 - \phi)^2} = \frac{d}{dt} \frac{\phi_1}{1 - \phi},$$

that the two quantities $\phi_1/(1-\phi)$ and $\phi_2/(1-\phi)$ must both be monotonically increasing functions of t . Geometrically, this means that the direction vector (ϕ_1, ϕ_2) can only point from the current point (ϕ_1, ϕ_2) into the triangle with vertices at the points $(1, 0)$, $(0, 1)$ and (ϕ_1, ϕ_2) , see Fig. 2. Thus, the triangle which defines the permissible range in the (ϕ_1, ϕ_2) plane decreases in size as t increases. Additional relations between the variables (ϕ_1, ϕ_2) and (v_1, v_2) are discussed by Norris, Callegari and Sheng (1984). It is shown there, and it will become clear later, that (ϕ_1, ϕ_2) are the preferred set of variables because they are independent of the particular construction process.

3. Differential equations for the moduli

The current elastic modulus L is a function of t through ϕ_1 and ϕ_2 and the path which these variables follow from $(0, 0)$ to the current point. Consider the removal of volume $\Delta v = \Delta v_1 + \Delta v_2$ from the current homogeneous composite and its replacement by grains of materials 1 and 2. The elastic modulus after the removal-replacement is $L + \Delta L$, defined by

$$\bar{\sigma} = (L + \Delta L) \bar{\epsilon} \quad (5)$$

where $\bar{\sigma}$ and $\bar{\epsilon}$ are the stress and strain averaged over the composite after the replacement. Thus,

$$\begin{aligned} V_0 \bar{\epsilon} &= \int_{V_0 - \Delta v} \epsilon \, dv + \int_{\Delta v} \epsilon \, dv, \\ V_0 \bar{\sigma} &= L \int_{V_0 - \Delta v} \epsilon \, dv + \int_{\Delta v} \hat{L} \epsilon \, dv \end{aligned} \quad (6)$$

where $\hat{L} = L_1$ in Δv_1 and L_2 in Δv_2 . Equations (5) and (6) imply the exact result

$$\Delta L \bar{\epsilon} = \frac{1}{V_0} \int_{\Delta v} (\hat{L} - L) \epsilon \, dv \quad (7)$$

Now consider an isolated grain of material 1 or 2 in the otherwise homogeneous composite. Assume the grain size is small enough compared with the sample size that we can define a homogeneous strain ϵ_0 at 'infinity'. We make the further assumption that the grain is ellipsoidal in shape.

This allows us to consider limiting cases of disk or needle shaped grains later. The main reason for specifying the inclusion to be an ellipsoid is the remarkable result of Eshelby, that the resultant strain in the inclusion is homogeneous. Specifically, in the inclusion,

$$\epsilon = T \epsilon_0 \quad (8)$$

where T is Wu's fourth order tensor (Wu, 1966; Korrington et al., 1979; Berryman, 1980) which depends upon the matrix moduli L , the inclusion moduli $\hat{L}$ and the aspect ratios of the ellipsoid. The dependence of T upon $\hat{L}$ can be made explicit by introducing Eshelby's fourth order tensor S (Eshelby, 1957; Mura, 1982)

$$T = [I + SL^{-1}(\hat{L} - L)]^{-1} \quad (9)$$

where I is the symmetric unit fourth order tensor,

$$I_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}). \quad (10)$$

The tensor S depends only upon L and the aspect ratios of the ellipsoid. When L is isotropic it turns out that S depends only on the Poisson's ratio (Mura, 1982).

The strain in a typical grain of material j , $j = 1, 2$ after the replacement is

$$\epsilon = T_j \epsilon_0 \quad \text{in } \Delta v_j \quad (11)$$

where T_j is Wu's strain concentration tensor for a material j inclusion in matrix L . Equation (11) assumes the grains are sufficiently separated that grain-grain interactions are negligible. This is true in the limit $\Delta v \rightarrow 0$, which we will consider shortly. Averaging (11) over all grains of material j , gives

$$\frac{1}{\Delta v_j} \int_{\Delta v_j} \epsilon \, dv = \bar{T}_j \epsilon_0, \quad j = 1, 2 \quad (12)$$

where the overbar denotes the average over aspect ratios and orientations. Note that the grain sizes do not play a role in this theory. A random orientation of grains results in an isotropic average tensor. We adopt the concise notation $L = (3\kappa, 2\mu)$ due to Hill (1965a) for the isotropic tensor L ,

$$L_{ijkl} = \kappa \delta_{ij} \delta_{kl} + \mu (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl}) \quad (13)$$

so that $L^{-1} = (\frac{1}{3}\kappa, \frac{1}{2}\mu)$ and $L_1 L_2 = (9\kappa_1 \kappa_2, 4\mu_1 \mu_2)$.

The isotropic strain concentration tensor is then (Berryman, 1980)

$$\bar{T}_j = (P_j, Q_j), \quad j = 1, 2 \quad (14)$$

where

$$P = \frac{1}{3} T_{iikk}, \quad Q = \frac{1}{3} (T_{ikik} - P). \quad (15)$$

The scalars P_j and Q_j , $j = 1, 2$ depend upon the inclusion material L_j , the matrix material L and the ellipsoid aspect ratios. In general, we can consider a distribution of aspect ratios which can vary with t . Explicit formulas for P and Q are given by Berryman (1980) for grains which are spheroids of arbitrary aspect ratio and when both matrix and inclusion are isotropic.

The incremental change in the moduli, given by (7) is now

$$\Delta L \bar{\epsilon} = (L_1 - L) \bar{T}_1 \epsilon_0 \frac{\Delta v_1}{V_0} + (L_2 - L) \bar{T}_2 \epsilon_0 \frac{\Delta v_2}{V_0}. \quad (16)$$

In the limit as $\Delta v = \Delta v_1 + \Delta v_2 \rightarrow 0$, we have

$$\bar{\epsilon} = \epsilon_0 \left[1 + O\left(\frac{\Delta v}{V_0}\right) \right].$$

The strain ϵ_0 is arbitrary, so that (16) implies

$$\Delta L = (L_1 - L) \bar{T}_1 \frac{\Delta v_1}{V_0} + (L_2 - L) \bar{T}_2 \frac{\Delta v_2}{V_0}$$

or, in the limit,

$$\dot{L}(t) = (L_1 - L) \bar{T}_1 \frac{\dot{v}_1}{V_0} + (L_2 - L) \bar{T}_2 \frac{\dot{v}_2}{V_0}. \quad (17)$$

Eliminating $\dot{v}_1$ and $\dot{v}_2$ using (4) gives us the path-dependent set of coupled differential equations

$$\begin{aligned} \dot{L}(t) = & (L_1 - L) \bar{T}_1 \dot{\phi}_1 + (L_2 - L) \bar{T}_2 \dot{\phi}_2 \\ & + [(L_1 - L) \bar{T}_1 \phi_1 + (L_2 - L) \bar{T}_2 \phi_2] \frac{\dot{\phi}}{1 - \phi} \end{aligned} \quad (18)$$

where $\phi(t) = \phi_1(t) + \phi_2(t)$. The initial conditions are $L(0) = L_0$.

Although (18) appears more cumbersome than (17), it is the preferred or canonical form. Equation (17) is phrased in terms of variables (v_1, v_2)

which are particular to the construction process considered. To see this more clearly, consider an alternative construction process in which the volume of composite material is allowed to increase. Begin at $t = 0$ with an initial volume v_0 . At each step in the homogenization incremental volumes Δv_1 and Δv_2 of materials 1 and 2 are added to the current total volume $V(t) \equiv v_0 + v_1 + v_2$. The volume fractions ϕ_1 and ϕ_2 of materials 1 and 2 are

$$\phi_1 = v_1/V, \quad \phi_2 = v_2/V. \quad (19)$$

Note that (19) defines a one-to-one relation between points in the (v_1, v_2) plane and points in the (ϕ_1, ϕ_2) plane. This contrasts with the previous construction process where there did not exist such a functional relationship, but just a differential relationship (4). The difference is trivially apparent; we are now retaining all the added material within the mixture. There is thus no indeterminacy in how the new material is distributed in terms of volume fractions. We refer to Norris, Callegari and Sheng (1984) for further discussion of either process. The relevant homogenization equation for the process under consideration may be derived in a similar manner to the analysis preceeding (17). We omit the details and just cite the result analogous to (17), viz.

$$\dot{L}(t) = (L_1 - L) \bar{T}_1 \frac{\dot{v}_1}{V} + (L_2 - L) \bar{T}_2 \frac{\dot{v}_2}{V}. \quad (20)$$

The difference between the two systems of equations is that the constant V_0 in (17) is replaced in (20) by V , which is a function of t . However, using (19), or its inverse

$$\frac{v_1}{v_0} = \frac{\phi_1}{1 - \phi}, \quad \frac{v_2}{v_0} = \frac{\phi_2}{1 - \phi},$$

we can eliminate $\dot{v}_1$ and $\dot{v}_2$ from (20) and once again obtain the system (18).

4. General results

4.1. Ordinary differential scheme (DEM)

Various differential schemes have been proposed over the years for the effective properties of two-phase composites. The first such scheme was

due to Roscoe (1952) who considered spherical grains. The same concept was also proposed by Boucher (1976) who considered arbitrary ellipsoidal grains. A concise formulation of the scheme is given by McLaughlin (1977). A good review of the differential effective medium or DEM approach is given by Cleary, Chen and Lee (1980). This paper also has an exhaustive list of relevant pre-1980 literature. Since then the method has been discussed by Sen, Scala and Cohen (1980), Norris, Sheng and Callegari (1984) in the context of the dielectric problem, and by Sheng and Callegari (1984) for the elastic problem.

Briefly, the idea behind DEM is to begin with one phase, say material 2, as the starting matrix or 'backbone'. Then material 1 grains are added such that their concentration increases incrementally from zero to the final value. The additional inclusions are imagined at each increment to be imbedded in a homogeneous composite material, made up of the matrix phase and the previously added inclusions. Thus, the ordinary DEM corresponds to a homogenization path along the ϕ_1 -axis, see Fig. 2. Put $\phi_2 = \dot{\phi}_2 = 0$ in (18) to obtain

$$\frac{dL}{d\phi_1} = \frac{1}{1-\phi_1} (L_1 - L) \bar{T}_1 \quad (21)$$

with initial condition $L = L_2$ at $\phi_1 = 0$. This is precisely the system of ordinary differential equations derived by McLaughlin (1977, equation (4)). Ordinary DEM, or any differential scheme developed previously, corresponds to paths along the ϕ_1 or ϕ_2 -axis. This result is to be expected, since previous schemes only considered imbedding one phase within the other. The present generalized scheme allows both phases to be imbedded simultaneously. There is thus a great deal more freedom in the generalized scheme. The freedom is reflected in the infinitely wide choice of paths in the (ϕ_1, ϕ_2) plane from which we choose. This novel path dependent aspect is discussed in more detail later. Finally, we note that the 'new self-consistent method' proposed by Zimmerman (1984a and b) is given by (21) with the volume normalization term $1/(1-\phi_1)$ omitted. The results of this method are known to violate the Hashin-Shtrikman bounds (Zimmerman, 1984a) and so we do not consider it further.

4.2. Effective medium approximation (EMA)

We now show here the EMA results from the generalized differential scheme. The EMA equations for a two-phase composite are

$$c_1(L_1 - L)\bar{T}_1 + c_2(L_2 - L)\bar{T}_2 = 0. \quad (22)$$

Here c_1 and c_2 are the concentrations of materials 1 and 2, such that $c_1 + c_2 = 1$. Equation (22) is a set of equations in the unknown moduli L . The tensors $\bar{T}_j$, $j = 1, 2$ are the averaged strain concentration tensors for inclusions of material j in the homogeneous composite with moduli L . The present formulation of EMA is consistent with the self consistent imbedding method (SCI) of Koringa (1973) and Koringa et al. (1979), or the self consistent method of Berryman (1980). Both Koringa (1973) and Berryman (1980) arrive at similar equations by setting averaged forward scattering amplitudes to zero in the low frequency limit of wave propagation. The EMA of (22) is also equivalent to the self consistent scheme of Hill (1965). However, later self-consistent methods due to Wu (1966), Walpole (1969) and Boucher (1974) among others, are not the same as (22). The latter theories do not treat the two phases in a symmetric fashion, whereas (22) gives no preference to one phase or the other. The difference between the two classes of self-consistent methods is discussed in detail by Berryman (1980).

In order to see how the EMA results from the generalized scheme, consider an arbitrary homogenization path which ends at $\phi_1 = c_1$, $\phi_2 = c_2$. As $(\phi_1, \phi_2) \rightarrow (c_1, c_2)$, we have $1 - \phi \rightarrow 0$. In this limit, it is clear from (18) that the only way L can remain finite is if L satisfies (22) at $(\phi_1, \phi_2) = (c_1, c_2)$. Thus, any path which ends on $\phi = 1$ results in a composite with moduli determined by the EMA.

Recently, Milton (1984a and b) has shown the EMA moduli to be realized through a hierarchical process analogous to the construction process of Section 2. Milton's procedure amounts to taking any permissible homogenization path in the (ϕ_1, ϕ_2) plane from $(0, 0)$ to (c_1, c_2) . Note that the EMA moduli are independent of the backbone L_0 . This is because all the original material has been replaced at $\phi = 1$, or $\phi_0 = 0$.

4.3. Isotropic constituents with random orientation

In this case the tensors $\bar{T}_1$ and $\bar{T}_2$ are isotropic, given by (14) and (15). Let the current moduli $L(t)$ be $L = (3\kappa, 2\mu)$. The system (18) becomes two coupled equations for κ and μ

$$\dot{\kappa}(t) = (\kappa_1 - \kappa)P_1 \left(\dot{\phi}_1 + \frac{\phi_1 \dot{\phi}}{1 - \phi} \right) + (\kappa_2 - \kappa)P_2 \left(\dot{\phi}_2 + \frac{\phi_2 \dot{\phi}}{1 - \phi} \right), \quad (23a)$$

$$\dot{\mu}(t) = (\mu_1 - \mu)Q_1 \left(\dot{\phi}_1 + \frac{\phi_1 \dot{\phi}}{1 - \phi} \right) + (\mu_2 - \mu)Q_2 \left(\dot{\phi}_2 + \frac{\phi_2 \dot{\phi}}{1 - \phi} \right) \quad (23b)$$

with initial conditions $\kappa(0) = \kappa_0$, $\mu(0) = \mu_0$. The scalars P_j and Q_j , $j = 1, 2$ depend upon κ , μ , κ_j and μ_j , $j = 1, 2$ and on the aspect ratios of the inclusions. We cite the following results from Berryman (1980) for later use: for spheres,

$$P_j = \frac{\kappa + \kappa_j^*}{\kappa_j + \kappa_j^*}, \quad Q_j = \frac{\mu + \mu_j^*}{\mu_j + \mu_j^*}, \quad (24)$$

and for disks (or plates)

$$P_j = \frac{\kappa + \kappa_j^*}{\kappa_j + \kappa_j^*}, \quad Q_j = \frac{\mu + \mu_j^*}{\mu_j + \mu_j^*}, \quad (25)$$

where

$$\begin{aligned} \kappa^* &= \frac{4}{3}\mu, & \mu^* &= \frac{1}{6}\mu \left(\frac{9\kappa + 8\mu}{\kappa + 2\mu} \right), \\ \kappa_j^* &= \frac{4}{3}\mu_j, & \mu_j^* &= \frac{1}{6}\mu_j \left(\frac{9\kappa_j + 8\mu_j}{\kappa_j + 2\mu_j} \right). \end{aligned} \quad (26)$$

We note that the two ordinary differential equations (23) are coupled when the inclusions are spheres. However, the equations decouple for disk-shaped inclusions. This fact is used to advantage in Section 5.

4.4. Exact solution for equal phase rigidities

When the constituents have equal shear moduli, $\mu_1 = \mu_2 = \mu_0 \equiv \mu$, but different bulk moduli, then the exact solution is known for any isotropic

geometry. This result, due to Hill (1963), states that for an isotropic composite of materials 1, 2 and 0 (the backbone) the effective bulk modulus κ is given by

$$\frac{1}{\kappa + \frac{4}{3}\mu} = \frac{\phi_0}{\kappa_0 + \frac{4}{3}\mu} + \frac{\phi_1}{\kappa_1 + \frac{4}{3}\mu} + \frac{\phi_2}{\kappa_2 + \frac{4}{3}\mu} \quad (27)$$

We now show that (27) follows from (23).

First, we have identically that $\dot{\mu} = 0$, so that (23) is just a single ordinary differential equation for $\kappa(t)$. The key to our proof is the result of Hill (1963) that P_1 and P_2 in (23), are independent of the inclusion shapes for equal phase rigidities. This result is easily checked for spheroids by inspection of the formulas in Berryman (1980). Substituting for P_1 and P_2 from (24) into (23) and rearranging, gives

$$\begin{aligned} \frac{d}{dt} [(\kappa + \kappa^*)(1 - \phi)]^{-1} \\ = \frac{d}{dt} \left\{ \phi_1 [(\kappa_1 + \kappa^*)(1 - \phi)]^{-1} \right. \\ \left. + \phi_2 [(\kappa_2 + \kappa^*)(1 - \phi)]^{-1} \right\}. \end{aligned}$$

Integrating, and using $\phi_0 + \phi_1 + \phi_2 = 1$ and the initial condition $\kappa(0) = \kappa_0$, we obtain (27). Finally, we note the trivial result that (23) gives the correct bulk modulus when $\kappa_0 = \kappa_1 = \kappa_2$ and the shear moduli differ. However, in this case there is no universal formula for μ .

5. Realization of the Hashin-Shtrikman bounds

5.1. Proof of result

The Hashin-Shtrikman bounds on κ and μ for an isotropic two-phase composite (Walpole, 1966) with $(\kappa_1 - \kappa_2)(\mu_1 - \mu_2) \geq 0$, are

$$\begin{aligned} \kappa_L &= \frac{\kappa_1 \kappa_2 + \kappa_L^* (c_1 \kappa_1 + c_2 \kappa_2)}{\kappa_L^* + c_1 \kappa_2 + c_2 \kappa_1}, \\ \kappa_G &= \frac{\kappa_1 \kappa_2 + \kappa_G^* (c_1 \kappa_1 + c_2 \kappa_2)}{\kappa_G^* + c_1 \kappa_2 + c_2 \kappa_1}, \\ \mu_L &= \frac{\mu_1 \mu_2 + \mu_L^* (c_1 \mu_1 + c_2 \mu_2)}{\mu_L^* + c_1 \mu_2 + c_2 \mu_1}, \\ \mu_G &= \frac{\mu_1 \mu_2 + \mu_G^* (c_1 \mu_1 + c_2 \mu_2)}{\mu_G^* + c_1 \mu_2 + c_2 \mu_1}, \end{aligned} \quad (28)$$

where suffices L and G indicate lower and upper bounds and κ_l^* , κ_g^* are the lesser and greater of κ_1^* , κ_2^* , and μ_l^* , μ_g^* are the lesser and greater of μ_1^* , μ_2^* . We consider the case $(\kappa_1 - \kappa_2)(\mu_1 - \mu_2) \geq 0$, and show that the lower bounds are achieved by DEM with phase g as matrix and phase l imbedded as disk-shaped inclusions. Similarly, the upper bounds are achieved by DEM with phase l as matrix and phase g imbedded as disks. A similar result has previously been derived by Boucher (1976).

It has been known for a long time that the Hashin-Shtrikman bounds on the bulk modulus, but not the shear modulus, can be realized by a packed-sphere microgeometry (Hashin, 1962; Milton, 1981). The present DEM-disk geometry achieves the bounds on κ and μ simultaneously. The upper bounds in particular are very interesting because they correspond to the stiffest composite obtainable with given concentrations of materials 1 and 2. Another realization of the bounds has been given by Lurie and Cherkav (1984). They use the fact that the packed-sphere microgeometry realizes the bounds on the shear modulus asymptotically, i.e. at dilute concentrations of one phase or another. A composite is then formed by iterating the dilute packed-sphere microgeometry infinitely many times. The procedure is very analogous to the construction process of the present paper. In fact, Lurie and Cherkav end up with a differential equation which they integrate up to get the Hashin-Shtrikman bound for μ .

Milton (1984c) has independently deduced that the Hashin-Shtrikman bounds are realizable. Expanding on an idea due to Schulgasser (1977), Milton defines a rank N laminate through a hierarchical procedure on N length scales. As $N \rightarrow \infty$, the scheme is analogous to the construction process of Section 2 with laminar or disc-shaped inclusions. Milton's result is that the Hashin-Shtrikman bounds are attained by an infinite rank laminate. Also independently, Murat and Tartar (1984) have shown that the bounds in the conductivity and on the bulk modulus are attained by rank N laminates, where N is the dimensionality of the space. They have also found laminate geometries of finite rank that attain the shear modulus bounds.

We now prove our assertion. Consider the lower bound κ_L on the bulk modulus. Differentiating (28) with respect to either c_1 or c_2 and using $c_1 + c_2 = 1$, we find after a bit of rearrangement that

$$\frac{d\kappa_L}{dc_j} = \left(\frac{\kappa_j - \kappa_L}{1 - c_j} \right) \left(\frac{\kappa_L + \kappa_j^*}{\kappa_j + \kappa_j^*} \right), \quad j = 1 \text{ or } 2. \quad (29)$$

Now choose $j = l$ in (29), then it follows that κ_L is given by $\kappa(c_l)$ where $\kappa(\phi_l)$ satisfies the initial value problem

$$\frac{d\kappa(\phi_l)}{d\phi_l} = \left(\frac{\kappa_l - \kappa}{1 - \phi_l} \right) \left(\frac{\kappa + \kappa_l^*}{\kappa_l + \kappa_l^*} \right), \quad \kappa(0) = \kappa_g. \quad (30)$$

Referring to (23) and (25), we note by inspection that (30) corresponds to (23) with $\phi_g = \phi_g = 0$ and the phase l inclusions as disks. Also, we require the backbone to be phase g . Thus, (30) is equivalent to the ordinary DEM equation (21) with phase g as matrix and phase l as disk inclusions, as we claimed above. The proof for κ_G , μ_L and μ_G is now apparent from the similarity between the P_j and Q_j in (25) and between κ_L , κ_G and μ_L , μ_G in (28).

5.2. Discussion

The above results must be used with caution when the bulk modulus or rigidity of either phase tends to zero. For instance, suppose one phase is a vacuum. Then, the Hashin-Shtrikman lower bounds on κ and μ are both zero. The upper bounds are non-zero, but according to our results they correspond to disks of the solid phase imbedded in the vacuum, which is apparently nonsense. Similarly, according to our results the lower bounds are realized by voids in the shape of disks imbedded in the solid phase. Looking at P_j and Q_j of (25) for disks, we see there is a singularity when $\kappa_j = \mu_j = 0$ unless $\kappa = \mu \equiv 0$, and hence the lower bounds follow. However, in arriving at (25), we have used the results for a spheroid of vanishingly small aspect ratio with non-vacuous included material ($\kappa_j \neq 0$, $\mu_j \neq 0$). The additional limits of $\kappa_j \rightarrow 0$, $\mu_j \rightarrow 0$ must be handled with care. If we first let the moduli vanish and subsequently take the limit of zero aspect ratio we obtain singular P_j and Q_j , but the integrals of P_j and Q_j over the

vanishingly small void-volume remain finite. In this case we obtain a theory for a cracked solid. There are several conflicting available theories on the moduli of cracked solids, some based upon differential schemes (Bruner, 1976; Henyey and Pomphrey, 1982), others on self-consistent schemes (Budiansky and O'Connell, 1976; Horii and Nemat-Nasser, 1983) and others on low-frequency scattering (Piau, 1980). It is not the goal of this paper to discuss the mechanical properties of cracked solids. We will show in another paper how a differential scheme for cracked solids follows from the general equations (18).

We note that the Hashin-Shtrikman bounds are predicted in certain limiting cases of other effective medium theories. Boucher (1974) was apparently the first to notice this. Boucher considered an unsymmetric self-consistent scheme (SCS) identical to the scheme of Wu (1966) and Walpole (1969), but unlike the symmetric EMA of Koringa et al. (1979), Berryman (1980) and the present paper. A major conclusion of Boucher's paper is that when $(\kappa_1 - \kappa_2)(\mu_1 - \mu_2) \geq 0$, the Hashin-Shtrikman bounds are equivalent to the estimated moduli of the SCS with disk-shaped inclusions. Specifically, the upper (lower) bound is given by the SCS moduli for phase g (phase l) inclusions imbedded in a matrix of phase l (phase g). The shape effect of the inclusions on the overall module has been examined by Wu (1966) and Walpole (1969). Both authors surmise that disk-shape inclusions are best for strengthening (weakening) the matrix material when the included phase is stiffer (more compliant) than the matrix phase. Wu (1966) based his conclusion on numerical comparisons for different shapes and Walpole (1969) based his on an analytic comparison of results for needle shapes and disk shapes. However, both authors had closed-form results for disk shaped inclusions (Wu, 1966, equations (23) and (24); Walpole, 1969, equation (61)) but failed to observe that these were the same as the Hashin-Shtrikman bounds. The Hashin-Shtrikman bounds are also predicted by the theory of Kuster and Toksoz (1974), as noted by Cheng (1978). In this case the required inclusion shape is spherical and $(\kappa_1 - \kappa_2)(\mu_1 - \mu_2) \geq 0$ again assumed. The upper (lower) bounds correspond to inclusions of phase l

(phase g) imbedded in a matrix of phase g (phase l). However, unlike EMA, DEM and the generalized theory of this paper, it is not known whether the above theories actually correspond to realizable microgeometries. Berryman (1980) has shown that the Kuster-Toksoz estimates for disc inclusions violate the Hashin-Shtrikman bounds. Therefore, the Kuster-Toksoz theory is not always realizable. Whether it and the theory of Wu, Walpole and Boucher are ever realizable remains an open question.

Another theory, due to Weng (1984), also gives the Hashin-Shtrikman bounds. The equation for the effective moduli is given by (B6) (see Appendix B) with $\hat{S} = c_1 S$, where S is the Eshelby tensor for the inclusions. When $(\kappa_1 - \kappa_2)(\mu_1 - \mu_2) > 0$ and S is taken as that for an isolated spherical inclusion, the upper (lower) bounds as κ and μ are achieved simultaneously for $\mu_1 > \mu_2$ ($\mu_1 < \mu_2$). It can also be checked that the upper (lower) bounds are attained for $\mu_1 < \mu_2$ ($\mu_1 > \mu_2$) if S for an arbitrarily oriented disk-like inclusion is used.

Finally, on the theme of the Hashin-Shtrikman bounds, we note that the bounds are also predicted for periodic composites when a certain approximation is made. The exact formulation for the effective moduli of periodic composites has been presented by Nemat-Nasser and Taya (1981) and Nemat-Nasser et al. (1982). The simplest approach to approximating the solution of the relevant equations is to assume the transformation strain in each inclusion to be constant. Using this approximation, Nemat-Nasser et al. (1982) considered periodically distributed spherical voids and found the estimated bulk modulus for the composite to be the Hashin-Shtrikman upper bound. Their result relied upon a certain relation which they did not prove. This relation is proved in Appendix B, where we also generalize their result to the case of solid inclusions. A similar result for the shear modulus is also derived in Appendix B.

6. Path dependence of the homogenization process

6.1. Example of different paths to the same endpoint

The coupled system of ordinary differential equations (18) depends critically upon the path we

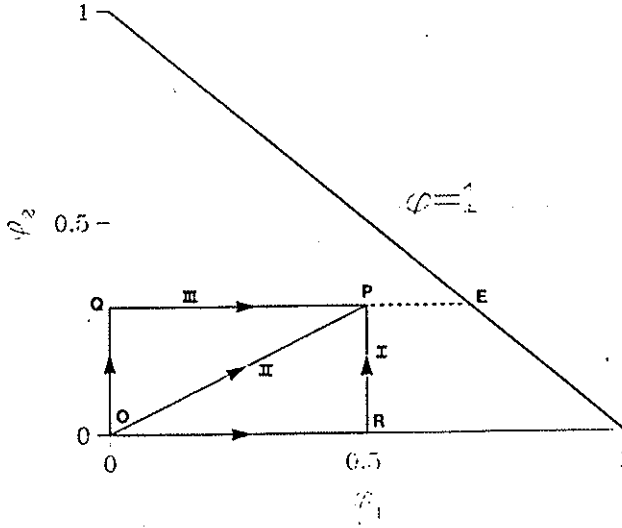


Fig. 3. Three paths to the same endpoint $P(\phi_1, \phi_2)$.

choose to consider in the (ϕ_1, ϕ_2) plane. In fact, two different paths from $(0, 0)$ to the same endpoint will, in general, not give the same result at the endpoint.

For example, consider the three paths in Fig. 3 with the backbone material 0 the same as material 1. Thus, the 'composite' is pure material 1 at 0. As we move along path I to the point R , the imbedded inclusions are also pure material 1. No matter what shapes the grains are, there is no change in material properties along OR . The process between R and P is defined by $\phi_1 = \text{constant}$. However, ϕ_0 , the volume fraction of material 1 associated with the starting material decreases between R and P as ϕ_2 increases. The removal-replacement process does not distinguish between the ϕ_0 and ϕ_1 domains of material 1, with the result that some ' ϕ_1 material' is always removed. Hence material 1 must be put back into the composite in the replacement process in addition to material 2. The relative injection rates of phases 1 and 2 along RP follows from (4) with $\dot{\phi}_1 = 0$ as

$$dv_1/dv_2 = \phi_1/(1 - \phi_1).$$

The relative injection rate is constant on the whole of path II, and equals

$$dv_1/dv_2 = \phi_1/\phi_2.$$

Thus, the material properties are always changing on path II. Similarly, the properties change all along path III. Between O and Q , only grains of material II are added. This particular process ($O \rightarrow Q$) corresponds to the previously studied differential scheme, or ordinary DEM of Boucher (1976), McLaughlin (1977) and Cleary et al. (1980). The solution to the ordinary DEM equations is unambiguous because there is no arbitrariness in the choice of path.

The three paths in Fig. 3 are composed of piecewise straight segments. Consider the system of ordinary differential equations (18) along a straight segment between the points (ϕ_{1A}, ϕ_{2A}) and (ϕ_{1B}, ϕ_{2B}) in the (ϕ_1, ϕ_2) plane. Define the line segment by $\phi_j = \phi_{jA} + t\Delta\phi_j$, $j = 1, 2$ where $0 \leq t \leq 1$ and

$$\Delta\phi_j = \phi_{jB} - \phi_{jA}, \quad j = 1, 2.$$

Substituting into (18) and eliminating the artificial variable t in favor of ϕ , we deduce the following autonomous system of ordinary differential equations for the straight line segment,

$$\frac{d}{d \ln(1 - \phi)} L = p_1(L - L_1)\bar{T}_1 + p_2(L - L_2)\bar{T}_2 \quad (31)$$

where

$$\begin{aligned} p_1 &= \frac{(1 - \phi_{2A})\Delta\phi_1 + \phi_{1A}\Delta\phi_2}{\Delta\phi_1 + \Delta\phi_2}, \\ p_2 &= \frac{(1 - \phi_{1A})\Delta\phi_2 + \phi_{2A}\Delta\phi_1}{\Delta\phi_1 + \Delta\phi_2}. \end{aligned} \quad (32)$$

Thus, $p_1 + p_2 = 1$ and we note that $\Delta\phi_1$ and $\Delta\phi_2$ are not infinitesimal quantities. The system of equations (31) reduce to two coupled equations for κ and μ when all the phases are isotropic. These two equations decouple in the particular case that the inclusions of phases 1 and 2 are disks. When the inclusions are spheres, the equations are

$$\frac{d\kappa}{d \ln(1 - \phi)} = \left[\frac{p_1}{\kappa_1 + \kappa^*} + \frac{p_2}{\kappa_2 + \kappa^*} - \frac{1}{\kappa + \kappa^*} \right] (\kappa + \kappa^*)^2 \quad (33a)$$

$$\frac{d\mu}{d \ln(1 - \phi)} = \left[\frac{p_1}{\mu_1 + \mu^*} + \frac{p_2}{\mu_2 + \mu^*} - \frac{1}{\mu + \mu^*} \right] (\mu + \mu^*)^2 \quad (33b)$$

where κ^* and μ^* are defined in (26). The EMA equations for κ and μ when both phases are spherical grains follows from (33) by setting the two right hand sides to zero with $p_1 = c_1$ and $p_2 = c_2$. These equations have been discussed in detail by Hill (1965b).

6.2. Numerical examples for voids

In the following numerical examples we have taken $\kappa_2 = \mu_2 = 0$. The ratios κ/κ_1 and μ/μ_1 depend on the material properties of phase 1 through the Poisson's ratio ν_1 , or alternatively through the ratio $R_1 \equiv \kappa_1^*/\kappa_1 = 4\mu_1/3\kappa_1 = 2(1 - 2\nu_1)/(1 + \nu_1)$. Results are shown in Figs. 4 and 5 for spherical grains and $R_1 = 0.5$ ($\nu_1 = \frac{1}{3}$). The DEM and EMA estimates are also plotted for comparison. The DEM results were found by integrating as far as Q in Fig. 3. Explicit and implicit solutions to the EMA and DEM equations, respectively, are given in Appendix A. The curves for paths I, II and III in Figs. 4 and 5 were calculated for $\phi_1 = 0.2$. The

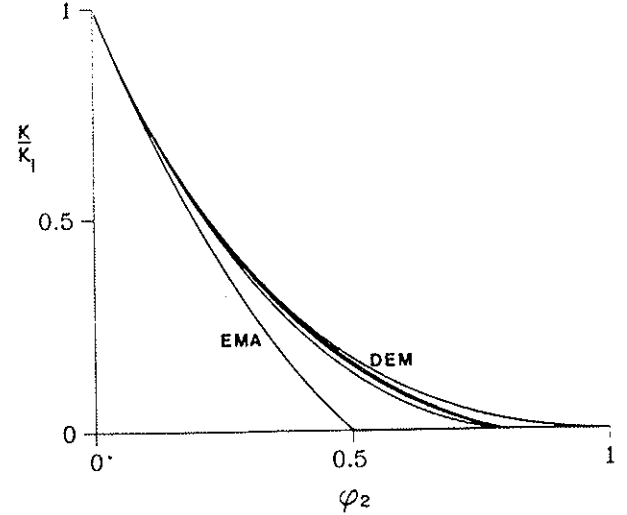


Fig. 4. Plots of κ/κ_1 for a solid-void composite with spherical grains and $\nu_1 = \frac{1}{3}$. The top curve is the usual DEM. The next 3 curves are for paths I, II and III with $\phi_1 = 0.2$. The bottom curve is the EMA result.

range of ϕ_2 is thus $0 \leq \phi_2 \leq 0.8$. We note the different results for each path. At the upper limit, $\phi_2 = 0.8$, all three paths reduce to the EMA result as expected from Section 4.2 since $\phi \rightarrow 1$ as $\phi_2 \rightarrow 0.8$. We note from Fig. 5 that $R \equiv \kappa^*/\kappa \rightarrow 1$ at the rigidity threshold for all curves. In Appendix A we show that $R \rightarrow 1$ independently of ν_1 . Recent numerical calculations by Tao and Sheng (1984)

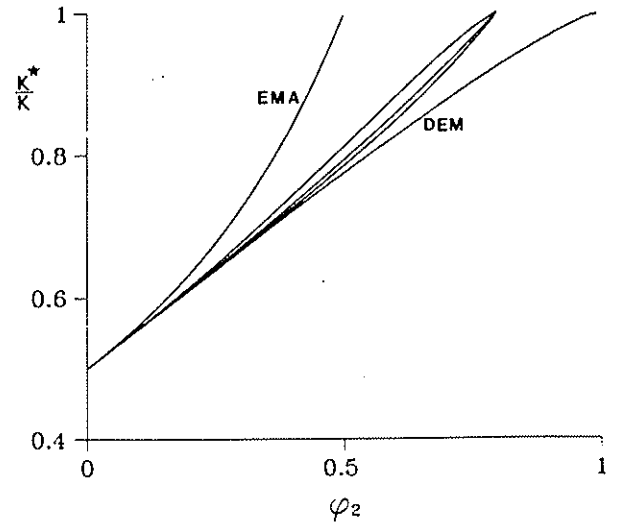


Fig. 5. Same as Fig. 4, for $R = 4\mu/3\kappa$. The ordering of the curves is, from top to bottom, EMA, paths III, II and I, DEM.

for periodic composites of fused solid spheres in a porous frame show that $R \approx \frac{1}{2}$ at the threshold and is material dependent. The discrepancy between the two results highlights the critical effect of microgeometry on the macroscopic moduli.

Finally, we illustrate how the results of the general method approach those of EMA as the backbone disappears. For a given value of $\phi_0 = 1 - \phi$, we have integrated the equations along path II of Fig. 3 from $(0, 0)$ to (ϕ_1, ϕ_2) , where $\phi_1 = 1 - \phi_0 - \phi_2$. The range of ϕ_2 is thus 0 to $1 - \phi_0$. The ratio κ/κ_1 is shown in Fig. 6 for several values of ϕ_0 . We note the significant deviation from the EMA result ($\phi_0 = 0$) even when $\phi_0 = 10^{-5}$. A similar situation exists for the electrical conductivity of a conductor-insulator composite. The EMA gives a conductivity threshold at $\frac{1}{3}$ for spherical grains. The approach to EMA as $\phi_0 \rightarrow 0$ has been discussed in detail by Norris, Callegari and Sheng (1984). They show that the conductivity right at threshold is $O(|\log \phi_0|^{-1})$ as $\phi_0 \rightarrow 0$. A comparison of Fig. 6 with the results of Norris, Callegari and Sheng (1984) suggests a similar behavior for κ at the rigidity threshold.

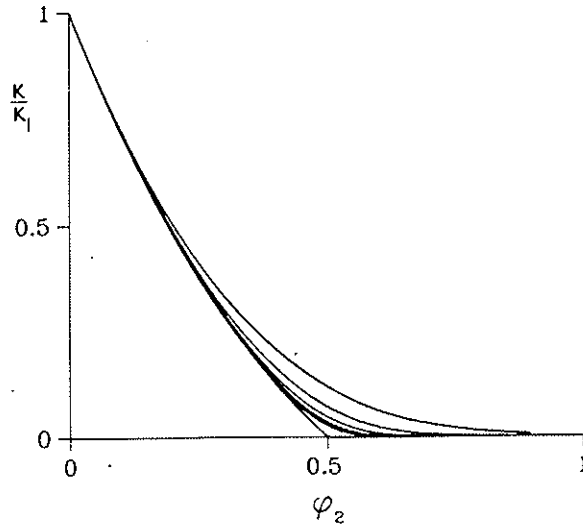


Fig. 6. Curves illustrating the approach to the EMA result for κ/κ_1 as the volume fraction of the backbone vanishes. The curves are, from the top down, $\phi_0 = 10^{-1}, 10^{-2}, 10^{-3}, 10^{-4}, 10^{-5}$ and the bottom curve is EMA, or $\phi_0 = 0$.

Appendix A. Results for voids

Specific solutions to the EMA and DEM equations are presented for spherical grains when one phase is vacuum (voids). The EMA equations, which follow from (33), are the same as those considered by Hill (1965b) and Budiansky (1965). The DEM equations also follow from (33) and one considered by McLaughlin (1977). The EMA solution has been given by Nemat-Nasser et al. (1982), but the DEM solution, to our knowledge, has not appeared previously.

Material 1 is defined by the moduli κ_1 and $\kappa_1^* = \frac{4}{3}\mu_1$. Define the ratio $R_1 = \kappa_1^*/\kappa_1$. The EMA solution is

$$\kappa^* = \begin{cases} \kappa_1^* \gamma, & c_2 < \frac{1}{2}, \\ 0, & c_2 \geq \frac{1}{2}, \end{cases} \quad (A.1)$$

$$\kappa = \kappa^* \left\{ \left[c_2 + \frac{1 - c_2}{1 + (R_1 \gamma)^{-1}} \right]^{-1} - 1 \right\}$$

where

$$4\gamma = 2 - 5c_2 - (3 - c_2)/R_1$$

$$+ \left\{ [2 - 5c_2 - (3 - c_2)/R_1]^2 \right.$$

$$\left. + 24(1 - 2c_2)/R_1 \right\}^{1/2}.$$

The DEM solution is defined implicitly by

$$\kappa^* = \kappa_1^* \left(\frac{R - 1}{R_1 - 1} \right)^{5/3}, \quad \kappa = \kappa^*/R \quad (A.2)$$

where R satisfies

$$\left(\frac{R - 1}{R_1 - 1} \right)^5 \left(\frac{R_1 + 1}{R + 1} \right) = (1 - c_2)^6.$$

Thus, as $c_2 \rightarrow 1$, we have $\kappa^*/\kappa \rightarrow 1$ and $\kappa = O((1 - c_2)^2)$. This should be compared with the analogous result for the effective conductivity σ of a conductor-insulator composite. As $c_2 \rightarrow 1$, the conducting phase disappears and DEM with spherical grains predicts $\sigma = O((1 - c_2)^{3/2})$ (Sen, Scala and Cohen, 1981).

Appendix B. Composites with periodic microstructure

Following Nemat-Nasser et al. (1982) we consider a matrix of material 1 containing periodically distributed inclusions of material 2. The unit cell is a parallelepiped with dimensions Λ_1 , Λ_2 and Λ_3 along the coordinate axes and volume $V = \Lambda_1 \Lambda_2 \Lambda_3$. The inclusion Ω occupies volume V_Ω in the unit cell and the volume fraction of phase 2 is $c_2 = V_\Omega/V = 1 - c_1$.

Let an overall strain ϵ^0 be prescribed in the composite. The transformation strain $\epsilon^*(x)$ in Ω is the solution to

$$\epsilon_{ij}^0 = A_{ijkl} \bar{\epsilon}_{kl}^* - \frac{1}{V} \sum_{n_p=0}^{\pm\infty} g_{ijkl}(\xi) \frac{1}{V_\Omega} \int_\Omega e^{i\xi \cdot x} d\xi \times \int_\Omega \epsilon_{kl}^*(x') e^{-i\xi \cdot x'} dx' \quad (B1)$$

where

$$A = (L_1 - L_2)^{-1} L_1, \quad (B2)$$

$$\bar{\epsilon}^* = \frac{1}{V_\Omega} \int_\Omega \epsilon^*(x) dx, \quad (B3)$$

$$g_{ijkl}(\xi) = \frac{1}{2} \left[\bar{\xi}_j (\delta_{il} \bar{\xi}_k + \delta_{ik} \bar{\xi}_l) + \bar{\xi}_i (\delta_{jl} \bar{\xi}_k + \delta_{jk} \bar{\xi}_l) \right] - \bar{\xi}_i \bar{\xi}_j \bar{\xi}_k \bar{\xi}_l / (1 - \nu_1) + \delta_{kl} \bar{\xi}_i \bar{\xi}_j \nu_1 / (1 - \nu_1), \quad (B4)$$

$$\bar{\xi}_i = \xi_i / \sqrt{\xi_k \xi_k},$$

$$\xi_i = 2\pi n_i / \Lambda_i \quad (\text{no sum}),$$

and where a prime on Σ indicates the $n_p n_p = 0$ is excluded in the summation. Also, ν_1 is the Poisson's ratio for material 1, assumed to be isotropic. Define the pseudo-Eshelby tensor $\hat{S}$ by

$$\epsilon^0 = (A - \hat{S}) \bar{\epsilon}^*, \quad (B5)$$

then the effective moduli of the composite are L were

$$L = L_1 [I - c_2 (A - \hat{S})^{-1}]. \quad (B6)$$

This equation for L follows directly from (7), (8) and (9), correct to first order in c_2 , with $\hat{S} = S$. A more precise analysis by Weng (1984) also gives (B6) but with $\hat{S} = c_1 S$.

The difficult step in the above procedure is solving the system of integral equations (B1). The simplest approximation is to replace $\epsilon^*(x)$ in (B1) with its average $\bar{\epsilon}^*$, defined in (B3). This is the approach adopted by Iwakuma and Nemat-Nasser (1983), see also Nemat-Nasser and Taya (1981) and Nemat-Nasser et al. (1982). In the following we assume that phase 2 is also isotropic. Consider first Case 1 of Nemat-Nasser et al. (1982), where the prescribed strain is $\epsilon_{ij} = \frac{1}{3} \epsilon^0 \delta_{ij}$ and the transformation strain is assumed to be $\epsilon_{ij}^*(x) = \bar{\epsilon}_{ij}^* = \frac{1}{3} \bar{\epsilon}^* \delta_{ij}$. The effective bulk modulus κ is from (B6),

$$\kappa = \kappa_1 - 3c_2 \kappa_1 / (A_{ijjj} - \hat{S}_{ijjj}) \quad (B7)$$

where, from (B2)

$$A_{ijjj} = 3\kappa_1 / (\kappa_1 - \kappa_2). \quad (B8)$$

Following Nemat-Nasser et al. (1982), we have

$$\hat{S}_{ijjj} = \left(\frac{1 + \nu_1}{1 - \nu_1} \right) \sum_{n_p=0}^{\pm\infty} P(\xi) \quad (B9)$$

where

$$P(\xi) = c_2 \left| \frac{1}{V_\Omega} \int_\Omega e^{i\xi \cdot x} dx \right|^2. \quad (B10)$$

Now,

$$\begin{aligned} \sum_{n_p=0}^{\pm\infty} P(\xi) &= \sum_{n_p=0}^{\pm\infty} P(\xi) - c_2 \\ &= \frac{c_2}{V_\Omega^2} \int_\Omega dx \int_\Omega dx' \sum_{n_p=0}^{\pm\infty} e^{i\xi \cdot (x-x')} - c_2 \\ &= \frac{c_2}{V_\Omega^2} \int_\Omega dx' \int_\Omega dx V \delta(x - x') - c_2 \\ &= 1 - c_2. \end{aligned} \quad (B11)$$

This result was assumed but not proven by Nemat-Nasser et al. (1982), see equation (7.7) of their paper. The result is independent of the inclusion shape. Combining (B7), (B8), (B9) and (B11) we obtain

$$\kappa = \frac{\kappa_1 \kappa_2 + \kappa_1^* (c_1 \kappa_1 + c_2 \kappa_2)}{\kappa_1^* + c_1 \kappa_2 + c_2 \kappa_1}. \quad (B12)$$

From (28), we see that (B12) is the upper (lower) bound on the bulk modulus if $\mu_1 > \mu_2$ ($\mu_1 < \mu_2$). Nemat-Nasser et al. (1982) obtained (B12) for the special case of voids, $\kappa_2 = \mu_2 = 0$.

Next, we consider Case 2 of Nemat-Nasser et al. (1982) where $\epsilon_{12}^0 = \epsilon_{21}^0 \neq 0$ and the rest of ϵ_{ij}^0 are zero. We again take the simplest approximation, assuming $\epsilon_{12}^*(x) = \epsilon_{21}^*(x) = \bar{\epsilon}_{12}^* = \bar{\epsilon}_{21}^* \neq 0$, with the rest = 0. The additional assumption that L and $\hat{S}$ are isotropic, implies from (B6) that the effective shear modulus is

$$\mu = \mu_1 - \frac{1}{2}c_2\mu_1/(A_{1212} - \hat{S}_{1212}) \quad (B13)$$

where

$$A_{1212} = \frac{1}{2}\mu_1(\mu_1 - \mu_2)^{-1}. \quad (B14)$$

$\hat{S}_{1212}$ follows from equations (3.8) and (3.9) and Appendix A of Nemat-Nasser et al. (1982), as

$$\hat{S}_{1212} = \sum_{n_p=0}^{\pm\infty} P(\xi) \left[\frac{1}{2}(\bar{\xi}_1^2 + \bar{\xi}_2^2) - \bar{\xi}_1^2 \bar{\xi}_2^2 / (1 - \nu_1) \right]. \quad (B15)$$

This expression cannot be simplified in general. Computed numerical values of the various series in (B15) are given by Nemat-Nasser and Taya (1981), Nemat-Nasser et al. (1982) and Iwakuma and Nemat-Nasser (1983) for different inclusion geometries. However, an interesting result is obtained if we replace the three terms, $\bar{\xi}_1^2$, $\bar{\xi}_2^2$ and $\bar{\xi}_1^2 \bar{\xi}_2^2$ in the square brackets in (B15) by their respective averages over the surface of the unit sphere. Thus,

$$\bar{\xi}_1^2 \rightarrow \langle \bar{\xi}_1^2 \rangle = \frac{1}{3},$$

$$\bar{\xi}_2^2 \rightarrow \langle \bar{\xi}_2^2 \rangle = \frac{1}{3},$$

$$\bar{\xi}_1^2 \bar{\xi}_2^2 \rightarrow \langle \bar{\xi}_1^2 \bar{\xi}_2^2 \rangle = \frac{1}{15}$$

where

$$\langle g \rangle \equiv \frac{1}{4\pi} \int_0^{2\pi} d\phi \int_0^\pi d\theta g(\theta, \phi) \sin \theta,$$

and $\bar{\xi} = (\cos \theta, \sin \theta \cos \phi, \sin \theta \sin \phi)$. Then (B15) becomes using (B11),

$$\begin{aligned} \hat{S}_{1212} &= c_1 \left[\frac{1}{3} - \frac{1}{15(1 - \nu_1)} \right] \\ &= \frac{1}{2} \frac{c_1 \mu_1}{\mu_1 + \mu_1^*} \end{aligned} \quad (B16)$$

where μ_1^* is defined in (26). Combining (B13), (B14) and (B16),

$$\mu = \frac{\mu_1 \mu_2 + \mu_1^* (c_1 \mu_1 + c_2 \mu_2)}{\mu_1^* + c_1 \mu_2 + c_2 \mu_1}. \quad (B17)$$

Comparison with (28) shows that (B17) is the upper (lower) bound on the shear modulus if $\mu_1 > \mu_2$ ($\mu_1 < \mu_2$), assuming $(\kappa_1 - \kappa_2)(\mu_1 - \mu_2) > 0$.

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