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An Examination of the Mori-Tanaka Effective Medium Approximation for Multiphase Composites

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The Mori-Tanaka method is considered in the context of both scalar thermal conductivity and anisotropic elasticity of multiphase composites, and some general properties are deduced. Particular attention is given to its relation to known general bounds, and to the differential scheme. It is shown that the moduli predicted by the method always satisfy the Hashin-Shtrikman and Hill-Hashin bounds for two-phase composites. This property does not generalize to multiphase composites. A specific example illustrates that the method can predict moduli in violation of the Hashin-Shtrikman bounds for a three-phase medium. However, if the particle shapes are all spheres, then the prediction for the multiphase composite is coincident with the Hashin-Shtrikman bounds if the matrix material is either the stiffest or the most compliant phase. It is also shown that the generalized differential effective medium method yields the same moduli as the Mori-Tanaka approximation if certain conditions are satisfied in the differential scheme. Thus, it is required that at each stage in the differential process, and for each phase j ($j = 1, 2, \ldots, n$) *of new material, the average field in the incrementally added phase* j *material must be the same as the average field in the bulk phase* j. *For two phase media,* $n = 1$, *this condition reduces to the less stringent requirement that the ratio of the field in the incrementally added material to the average field in the matrix material is the same as the dilute concentration ratio. The cumulative findings of this paper, particularly those concerning bounds, suggest that the Mori-Tanaka approximation be used with caution in multiphase applications, but is on firmer ground for two-phase composites.*

1 Introduction

Various approximate methods exist for predicting the effective thermal, electrical, and mechanical properties of composites. Among these are the self-consistent scheme, and the differential scheme (Cleary et al., 1980, McGlaughlin, 1977). These effective medium approximations do not require detailed statistical information of the microstructure, but can distinguish between different inclusion shapes. Therefore, such schemes can be useful for statistically homogeneous composites with known inclusion shapes. However, there is always some doubt as to their utility. For example, it is not obvious, a priori, whether the results will automatically satisfy known bounds on the moduli, such as those of Hashin and Shtrikman (1963). At the present time, several methods, including the differential scheme, are known to correspond to realizable media, and hence satisfy the bounds (Avellaneda, 1987).

The Mori-Tanaka approximation is another method that has received attention recently. It is based upon the original work of Mori and Tanaka (1973), and has been used to advantage by, for example, Taya and Mura (1981) and Taya and Chou (1981). Weng (1984) applied the Mori-Tanaka method to the effective medium problem for a two-phase composite with spherical inclusions. Further applications have been given by Benveniste (1986a,b; 1987a,b,c) for the thermal conductivity and mechanical properties of two-phase and multiphase media. Unlike most other approximate methods which require solving implicit equations numerically, the Mori-Tanaka method yields explicit, closed-form answers for the effective properties. As with all other effective medium methods, it hinges upon a mathematical approximation, explained in the following sections. A significant property was discovered by Weng (1984), who showed that the Mori-Tanaka method with spherical inclusions of the softer (harder) phase gives the Hashin-Shtrikman upper (lower) bounds for the bulk and shear moduli. Norris (1985) pointed out that randomly-oriented disk-shaped particles of the softer (harder) phase yields the lower (upper) bounds. Benveniste (1987c) has recently proved, using a clever argument, that the bulk and shear modulus predicted by Mori-Tanaka for a two-phase

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composite with randomly-oriented ellipsoidal particles will lie within the Hashin-Shtrikman bounds. This benign property had previously been noted from the numerical results of Tandon (1986) and Maewal and Dandekar (1987). However, as we show in this paper, the Mori-Tanaka method can give results for multiphase media that are in violation of the Haskin-Shtrikman bounds. It appears, therefore, that two-phase composites are a special case for the method.

The purpose of this paper is to examine the connection between the Mori-Tanaka approximation and known bounds. We also relate the approximation to the generalized differential scheme. One common consequence from both studies is that the two-phase medium is a special case. Both thermal and elastic properties are considered. The analysis is simpler for the scalar thermal conductivity problem, and is presented in Section 2. The theory for the elastic moduli is presented in Section 3, where the major results concerning bounds are derived. The connection with the differential scheme is explored in Section 4.

2 The Effective Thermal Conductivity of a Multiphase Isotropic Composite

2.1 General Equations and Definitions. Consider an $n+1$ phase composite made of isotropic constituents with thermal conductivities k_i, $i=0, 1, 2, \ldots, n$, and occupying total volume fractions c_i, such that $\sum_{i=0}^{n} c_i = 1$. Phase $i=0$ corresponds to the matrix material. The temperature field $\phi(\mathbf{x})$ and the normal component of the heat flux, $\mathbf{q}\cdot\mathbf{n}$, where $\mathbf{q}$ is the flux and $\mathbf{n}$ the unit normal, are both continuous across interfaces between the constituents. The heat flux and temperature field in phase i are related by

$$\mathbf{q}^{(i)} = k_i \mathbf{H}^{(i)}, \tag{1}$$

where

$$\mathbf{H}^{(i)} = -\nabla\phi^{(i)}. \tag{2}$$

Assuming an isotropic distribution of grains, the effective conductivity is isotropic and equal to $k^{(\mathrm{eff})}$, defined by the macroscopic relation

$$\bar{\mathbf{q}} = k^{(\mathrm{eff})}\bar{\mathbf{H}}. \tag{3}$$

An overbar denotes the spatial average of a quantity. Thus, $\bar{\mathbf{H}}$ is the average of $\mathbf{H}$ over the entire composite, and $\bar{\mathbf{H}}^{(i)}$ is the average of $\mathbf{H}^{(i)}$ in phase i. The average $\bar{\mathbf{H}}$ could be imposed, for example, by the boundary condition that $\phi = -\bar{\mathbf{H}}\cdot\mathbf{x}$ on the exterior surface of the composite. Under the assumption of macroscopic isotropy, the vectors $\bar{\mathbf{H}}$, $\bar{\mathbf{H}}^{(i)}$, can be replaced by scalar quantities $\bar{H}$, $\bar{H}^{(i)}$, where $\bar{H}^{(i)}$ is the component of $\bar{\mathbf{H}}^{(i)}$ in the direction of $\bar{\mathbf{H}}$. The effective conductivity follows from equations (1) and (3) as

$$k^{(\mathrm{eff})}\bar{H} = \sum_{i=0}^{n} k_i c_i \bar{H}^{(i)}. \tag{4}$$

Equation (4) can be rewritten

$$k^{(\mathrm{eff})} = k_0 + \frac{\sum_{j=1}^{n} (k_j - k_0) c_j \bar{H}^{(j)}/\bar{H}^{(0)}}{c_0 + \sum_{i=1}^{n} c_i \bar{H}^{(i)}/\bar{H}^{(0)}}. \tag{5}$$

This is an exact equation for the effective conductivity $k^{(\mathrm{eff})}$, but is complicated by the determination of the ratios $\bar{H}^{(j)}/\bar{H}^{(0)}$ for each of the added phases $j=1, 2, \ldots, n$.

2.2 The Mori-Tanaka Scheme and Hashin-Shtrikman Bounds. The Mori-Tanaka effective conductivity k is obtained from equation (5) by assuming that the ratio $\bar{H}^{(j)}/\bar{H}^{(0)}$ is equal to the ratio for a single, isolated grain of phase j embedded in uniform matrix material of infinite extent. Equivalently, the ratio $\bar{H}^{(j)}/\bar{H}^{(0)}$ is taken to be the ratio pertaining in the dilute limit of $c \ll 1$, where c is the total volume fraction of the added phases,

$$c = \sum_{j=1}^{n} c_j = 1 - c_0.$$

The ratio $\bar{H}^{(j)}/\bar{H}^{(0)}$ for randomly-oriented ellipsoidal particles of phase j is described by three depolarization coefficients β_1, β_2, and β_3 for each of the major axes (see Landau and Lifshitz (1960) for explicit formulae). These coefficients are between 0 and 1, and satisfy $\beta_1+\beta_2+\beta_3=1$. They each equal 1/3 for spherical particles; long circular cylinders (needles) have $\beta_1=0$, $\beta_2=\beta_3=1/2$, and thin, circular disks have $\beta_1=\beta_2=0$, $\beta_3=1$. In general,

$$\bar{H}^{(j)}/\bar{H}^{(0)} = \frac{1}{3}\sum_{i=1}^{3}\left(1+\frac{k_j-k_0}{\beta_i^{-1}k_0}\right)^{-1}. \tag{6}$$

Benveniste (1986a) observed that if k_0 is smaller (larger) than all of the other k_j, $j=1, 2, \ldots, n$, then the Mori-Tanaka method with $\bar{H}^{(j)}/\bar{H}^{(0)}$ for spherical particles $j=1, 2, \ldots, n$, gives the Hashin-Shtrikman lower (upper) bound on the effective conductivity.

On the other hand, if all the particles are in the shape of thin, circular disks, the Mori-Tanaka scheme gives, from equations (5) and (6),

$$k = k_0\left[\frac{1+2\bar{k}k_0^{-1}}{2+k_0\overline{k^{-1}}}\right], \tag{7}$$

where

$$\bar{k} = \sum_{j=0}^{n} c_j k_j \text{ and } \overline{k^{-1}} = \sum_{j=0}^{n} c_j k_j^{-1}.$$

For a two-phase composite, $n=1$, in which $k_0<k_1$ ($k_0>k_1$), the prediction of equation (7) corresponds to the Hashin-Shtrikman upper (lower) bound on the effective conductivity. This result does not generalize to the multiphase composite, $n>1$. In fact, equation (7) can violate the Hashin-Shtrikman bounds for $n>1$. For example, k of (7) exceeds the upper bound if $n=2$, $k_1=2k_0$, $k_2=3k_0$ and $c_1=c_2=0.4$. This violation indicates that the Mori-Tanaka scheme is not always realizable for multiphase composites.

It is possible to show for two-phase composites that the Mori-Tanaka k of equations (5) and (6) satisfies the Hashin-Shtrikman bounds. To see this, consider equation (5) with $n=1$ and $\alpha=\bar{H}^{(1)}/\bar{H}^{(0)}$. Then,

$$\frac{\partial k}{\partial\alpha} = \frac{c_0c_1}{(c_0+c_1\alpha)^2} \geq 0. \tag{8}$$

Thus, k is maximum (minimum) for α maximum (minimum). Now consider α of equation (6) as a function of β_1, β_2, and β_3 constrained to the interior and surface of the tetrahedron $\beta_1+\beta_2+\beta_3=1$. It is easily shown that $\alpha(\beta_1, \beta_2, \beta_3)$ attains stationary values at the four vertices, corresponding to plate-like particles, at (1/3, 1/3, 1/3), which is a sphere, and at the points (1/2, 1/2, 0), (1/2, 0, 1/2), and (0, 1/2, 1/2) which are needles. If $k_1>k_0$, then α(sphere) < α(needles) < α(plate). But the sphere and plate values of α correspond to the Hashin-Shtrikman bounds on k, therefore, all other α give intermediate results.

3 The Effective Elastic Moduli of a Multiphase Composite

3.1 The Mori-Tanaka Approximation. Let $\mathbf{L}$ be the effective elastic modulus tensor for a multiphase composite, each phase, $i=0, 1, 2, \ldots, n$ characterized by a fourth-order modulus tensor $\mathbf{L}_i$. The anisotropic Hooke's law for each phase is

$$\sigma_i = \mathbf{L}_i \epsilon_i, \tag{9}$$

where $\epsilon_i = 1/2[\nabla \mathbf{u}_i + (\nabla \mathbf{u}_i)^T]$ is the strain. Let the average strain in phases i and 0 be related by

$$\bar{\epsilon}_i = \bar{\mathbf{T}}_{i0}\bar{\epsilon}_0, \qquad i=1, 2, \ldots, n, \tag{10}$$

then an exact expression for the effective moduli $\mathbf{L}^{(\mathrm{eff})}$ follows in a manner analogous to the derivation of equation (6), as

$$\mathbf{L}^{(\mathrm{eff})} = \mathbf{L}_0 + \left(\sum_{j=1}^{n} c_j(\mathbf{L}_j - \mathbf{L}_0)\bar{\mathbf{T}}_{j0}\right)\left[c_0\mathbf{I} + \sum_{i=1}^{n} c_i \bar{\mathbf{T}}_{i0}\right]^{-1}. \tag{11}$$

Here $\mathbf{I}$ is the fourth-order isotropic indentity tensor, $\mathbf{I}=(1, 1)$ in the concise notation of Hill (1965). In the same notation, an isotropic stiffness tensor is $\mathbf{L}=(3\kappa, 2\mu)$, where κ and μ are bulk and shear moduli, the compliance tensor is $\mathbf{M}=\mathbf{L}^{-1}=(1/3\kappa, 1/2\mu)$, and tensor products are $\mathbf{L}_1\mathbf{L}_2=(9\kappa_1\kappa_2, 4\mu_1\mu_2)$.

In the Mori-Tanaka method $\bar{\mathbf{T}}_{i0}$ is approximated by the analogous quantity for an isolated particle of phase i in an infinite matrix of phase 0. Equivalently, the dilute limit value of $\bar{\mathbf{T}}_{i0}$ is taken. Then equation (1) provides an explicit equation for the Mori-Tanaka effective moduli $\mathbf{L}$. In particular, if the particles of phase i are ellipsoidally shaped and aligned. Eshelby's results provide $\bar{\mathbf{T}}_{i0}$ in the simple form

$$\bar{\mathbf{T}}_{i0} = [\mathbf{I} + \mathbf{S}_{i0}\mathbf{L}_0^{-1}(\mathbf{L}_i - \mathbf{L}_0)]^{-1}, \tag{12}$$

where $\mathbf{S}_{i0}$ depends only upon $\mathbf{L}_0$ and the aspect ratios of the particle of phase i (see, for example, Mura (1982)). The Mori-Tanaka method as defined by equation (11) and the approximation (12) has been called the "direct approach" by Benveniste (1987c). He showed it is identical for two-phase composites to the usual "equivalent inclusion-average stress" formulation of, for example, Weng (1984). The equivalence of the two formulations for multiphase composites is demonstrated in the Appendix.

3.2 Bounds on the Elastic Moduli. If all particle shapes are identical, and the particles are aligned, then the Eshelby tensor $\mathbf{S}_{i0}$ is the same for each phase $i=1, 2, \ldots, n$. Define L_0^0 by

$$\mathbf{L}_0^0 = \mathbf{L}_0\mathbf{S}_{i0}^{-1} - \mathbf{L}_0. \tag{13}$$

Then the Mori-Tanaka effective moduli become

$$\mathbf{L} = \left(\sum_{j=0}^{n} c_j(\mathbf{L}_j + \mathbf{L}_0^0)^{-1}\right)^{-1} - \mathbf{L}_0^0. \tag{14}$$

Walpole (1966) obtained lower (upper) bounds on $\mathbf{L}^{(\mathrm{eff})}$ in the form of equation (14) under the assumption that $(\mathbf{L}_i - \mathbf{L}_0)$ is positive (negative) definite for all $i=1, 2, \ldots, n$. The bounding modulus tensor is $\mathbf{L}^*$, defined by (14), with $\mathbf{L}_0^0 = \mathbf{L}_0^*$, where $\mathbf{L}_0^*$ depends upon the particular type of anisotropy of the composite.

If all the constituents are isotropic, then the well-known Hashin-Shtrikman (1963) bounds for a macroscopically isotropic composite are defined by

$$\mathbf{L}_0^* = (3\kappa_0^*, 2\mu_0^*), \tag{15}$$

where

$$\kappa_j^* = \frac{4}{3}\mu_j, \tag{16a}$$

$$\mu_j^* = \frac{\mu_j}{6}\left[\frac{9\kappa_j + 8\mu_j}{\kappa_j + 2\mu_j}\right]. \tag{16b}$$

The corresponding Eshelby tensor $\mathbf{S}_{i0}$ is for a spherical particle. Thus, we have that the Mori-Tanaka approximation for spherical particles coincides with the lower (upper) Hashin-Shtrikman bounds on κ and μ if $\kappa_j > \kappa_0$, and $\mu_j > \mu_0$, $j=1, 2, \ldots, n$ ($\kappa_j < \kappa_0$, $\mu_j < \mu_0$, $j=1, 2, \ldots, n$). This equivalence has been previously noted by Weng (1984) for a two-phase composite.

While still on the topic of isotropic constituents, we note that if all the particles are randomly-oriented disks, then $\bar{\mathbf{T}}_{i0}$ can still be expressed in the form of equation (12), but $\mathbf{S}_{i0}$ now depends upon $\mathbf{L}_i$,

$$\mathbf{S}_{j0} = (\mathbf{I} + \mathbf{L}_j^*\mathbf{L}_0^{-1})^{-1}. \tag{17}$$

The Mori-Tanaka effective moduli κ and μ reduce in the special case of a two-phase composite to the Hashin-Shtrikman lower (upper) bounds if $\kappa_1 < \kappa_0$ and $\mu_1 < \mu_0$ ($\kappa_1 > \kappa_0$, $\mu_1 > \mu_0$). As for the thermal conductivity, this result does not generalize to multiphase composites.

If all the constituents are aligned, transversely isotropic phases, the moduli tensors can be represented succinctly in the notation of Walpole (1969) as $\mathbf{L}=(2k, l, n, 2m, 2p)$, $\mathbf{M}=\mathbf{L}^{-1}=1/2(n/\gamma, -l/\gamma, 2k/\gamma, 1/m, 1/p)$, where $\gamma = kn - l^2 = kE$, and E is the axial Young's modulus. For example, if x_3 is the symmetry axis, then $k=C_{11}-C_{66}$, $l=C_{13}$, $n=C_{33}$, $m=C_{66}$, and $p=C_{44}$. Positive definiteness requires that k, m, p, and $n-l^2/k$ are each positive. The bounds of Hill (1964) and Hashin (1965) apply to composites made of aligned cylindrical fibers of arbitrary transverse geometry. These bounds have been phrased succinctly by Walpole (1969) for multiphase composites. In this case the tensor $\mathbf{L}_0^*$ is not well-defined, so it is necessary to work with the compliance tensor $\mathbf{M}$. The dual equation to (14) is

$$\mathbf{M} = \left(\sum_{j=0}^{n} c_j(\mathbf{M}_j + \mathbf{M}_0^0)^{-1}\right)^{-1} - \mathbf{M}_0^0, \tag{18}$$

where $\mathbf{M}_0^0 = \mathbf{L}_0^{0^{-1}}$, when the latter is defined. If $(\mathbf{M}_i - \mathbf{M}_0)$ is positive (negative) definite for all $i=1, 2, \ldots, n$, then the lower (upper) bound, $\mathbf{M}^*$, on $\mathbf{M}^{(\mathrm{eff})}$ is given by equation (18), with $\mathbf{M}_0^0 = \mathbf{M}_0^*$, where $\mathbf{M}_0^*$ is

$$\mathbf{M}_0^* = \frac{1}{2}\left(\frac{1}{m_0}, 0, 0, \frac{1}{m_0} + \frac{2}{k_0}, \frac{1}{p_0}\right). \tag{19}$$

Note that Walpole's (1969) $\mathbf{M}_0^*$ contains a typographical error: The correct expression follows from Laws (1974).

The Eshelby tensor $\mathbf{S}_{i0}$ corresponding to equation (19) is that of a circular, cylindrical particle. Therefore, the Hill-Hashin bounds for a multiphase tranversely isotropic fibrous composite of arbitrary transverse geometry corresponds to the Mori-Tanaka approximation with circularly cylindrical particles. This equivalence has been noted by Tandon (1986) for a two-phase composite.

We next develop general results relating the Mori-Tanaka method to the bounds discussed above. The procedure adopted is a generalization of Benveniste's (1987c). Returning to the general assumption that the particles are all identically shaped and aligned, then equation (14) is correct to first-order in c in the dilute limit of $c \ll 1$. Thus,

$$\mathbf{L}^{(\mathrm{eff})} \sim \mathbf{L} = \mathbf{L}_0 + \sum_{j=1}^{n} c_j(\mathbf{L}_j - \mathbf{L}_0)(\mathbf{L}_j + \mathbf{L}_0^0)^{-1}(\mathbf{L}_0 + \mathbf{L}_0^0) + 0(c^2). \tag{20}$$

For a variation $\delta\mathbf{L}_0^0$ of $\mathbf{L}_0^0$ in this equation, the corresponding variation in $\mathbf{L}$ is

$$\delta\mathbf{L} = \sum_{j=1}^{n} c_j(\mathbf{L}_j - \mathbf{L}_0)(\mathbf{L}_j + \mathbf{L}_0^0)^{-1}\delta\mathbf{L}_0^0(\mathbf{L}_j + \mathbf{L}_0^0)^{-1} \times (\mathbf{L}_j - \mathbf{L}_0) + 0(c^2). \tag{21}$$

Thus, a positive (negative) definite change in $\mathbf{L}_0^0$ yields a corresponding positive (negative) definite change in $\mathbf{L}$. By assumption, the dilute limit effective moduli satisfy the bounds for the particular type of microstructure considered. Therefore, if $\mathbf{L}^*$ represents the lower (upper) bounds, then both $\mathbf{L}-\mathbf{L}^*$ and $\mathbf{L}_0^0-\mathbf{L}_0^*$ are positive (negative) definite tensors of the same anisotropy class.

Now consider the finite concentration case. Variation of $\mathbf{L}_0^0$ in equation (14) yields

$$\delta\mathbf{L}=(\mathbf{L}+\mathbf{L}_0^0)\sum_{j=0}^{n}c_j[(\mathbf{L}_j+\mathbf{L}_0^0)^{-1} - (\mathbf{L}+\mathbf{L}_0^0)^{-1}]\delta\mathbf{L}_0^0[(\mathbf{L}_j+\mathbf{L}_0^0)^{-1}-(\mathbf{L}+\mathbf{L}_0^0)^{-1}](\mathbf{L}+\mathbf{L}_0^0), \quad (22)$$

which is positive (negative) definite if $\delta\mathbf{L}_0^0$ is positive (negative) definite. It then follows from the dilute limit result for $\mathbf{L}_0^0-\mathbf{L}_0^*$ that $\mathbf{L}$ of equation (14) satisfies the bounds for finite concentrations. We have thus derived the result that the Mori-Tanaka approximation for multiphase composites, with all particles of the same shape and aligned, satisfies the appropriate Hashin-Shtrikman or Hill-Hashin bounds. This result is not as general as it may seem at first, since the only particle shapes that satisfy these criteria for the Mori-Tanaka approximation are spheres for isotropy, and circular cylinders for fiber-reinforced transverse isotropy. Also, these particular materials correspond to the Hashin-Shtrikman and Hill-Hashin bounds. Although there is no additional information here concerning multiphase composites, these results do have significant application to the particular case of two-phase composites.

The Mori-Tanaka approximation for a two-phase composite ($n=1$) can be written in the form of equation (14) with

$$\mathbf{L}_0^0=(\mathbf{L}_1-\mathbf{L}_0)(\bar{\mathbf{T}}_{10}^{-1}-\mathbf{I})^{-1}-\mathbf{L}_0, \quad (23)$$

where $\bar{\mathbf{T}}_{10}$ is the strain concentration ratio for dilute particulate concentration. Note that $\mathbf{L}_0^0$ of equation (23) is independent of c_1, and holds for any $\bar{\mathbf{T}}_{10}$. Therefore, by the same arguments as before, since the dilute limit moduli must satisfy the lower (upper) bounds on $\mathbf{L}^{(eff)}$, it follows that $\mathbf{L}_0^0-\mathbf{L}_0^*$ is positive (negative) definite. This in turn implies that $\mathbf{L}-\mathbf{L}^*$ is positive (negative) definite at all concentrations, $0<c_1<1$. Hence, the Mori-Tanaka moduli for two-phase composites satisfy the Hashin-Shtrikman or Hill-Hashin bounds, as appropriate. This general result is unique to two-phase composites, since it is not generally possible to write the Mori-Tanaka approximation for multiphase composites in the form of equation (14).

4 The Mori-Tanaka Approximation in Terms of the Differential Scheme

4.1 The Differential Scheme. The generalized differential scheme as understood here is a generalization of Bruggeman's (1935) method to multiphase composites. The present development is similar to that of Norris et al. (1985), where it was applied to a special type of three-phase medium in which one of the added phases was identical to the original matrix material. In order to understand the scheme, consider the scalar conductivity problem for a composite at some finite concentrations of the added phases $j=1, 2, \ldots, n$. Let V be the total volume of the composite, and as before, $\bar{H}$ is the average of the field $H(\mathbf{x})$ in V. Infinitesimal, discrete volumes of the homogeneous composite material are then removed and replaced by homogeneous amounts of phases $j=1, 2, \ldots, n$. Thus, volume dv_j is replaced by phase j, such that the replaced volume is perfectly bonded to the composite, and dv_j contains a representative quantity of the existing composite of conductivity k. If $\tilde{H}^{(j)}$ is the average of H in dv_j, the incremental change in the effective conductivity is

$$dk=\sum_{j=1}^{n}(k_j-k)\frac{\tilde{H}^{(j)}}{\bar{H}}\frac{dv_j}{V}. \quad (24)$$

It is preferable to work with the incremental volume fractions dc_j, rather than with the incremental volumes dv_j. Since the total volume V remains fixed, it is possible to show (Norris et al., 1985) that

$$\frac{dv_j}{V}=dc_j+c_j\frac{dc}{1-c}, \quad (25)$$

and hence,

$$dk=\sum_{j=1}^{n}(k_j-k)\frac{\tilde{H}^{(j)}(t)}{\bar{H}(t)}\left[dc_j+c_j\frac{dc}{1-c}\right]. \quad (26)$$

This becomes an ordinary differential equation by introducing a parameter t to describe the evolution of the composite from homogeneous phase 0 with initial conditions $k(0)=k_0$, $c_j(0)=0, j=1, 2, \ldots, n$. The volume fraction c could be used as the parameter t, for example.

A rigorous justification for the differential equation (26) has been given by Avallaneda (1987). An explicit equation can be obtained, if, for example, the particles are assumed to be in the shape of randomly-oriented ellipsoids. Then $\tilde{H}^{(j)}/\bar{H}$ is given by the right-hand side of equation (6) with k_0 replaced by $k(t)$. Bruggeman's (1935) scheme is contained in (26) as the special case of a two phase composite, $n=1$. The effective medium approximation for multicomponent composites is the limit of (26) as $c\to 1$. Discussions of these and other limiting cases are contained in Norris et al. (1985) and Norris (1985).

4.1 The Connection With the Mori-Tanaka Approximation. The field ratio in equation (26) can be written

$$\frac{\tilde{H}^{(j)}}{\bar{H}}=\frac{\tilde{H}^{(j)}/\bar{H}^{(0)}}{1-c+\sum_{i=1}^{n}c_i\bar{H}^{(i)}/\bar{H}^{(0)}}. \quad (27)$$

Note that the averages $\tilde{H}^{(j)}$ and $\bar{H}^{(j)}$ are generally quite distinct and unrelated. The former is the average field in the incrementally added particles of phase j, while the latter is the average field in the entire volume of phase j in the composite.

Define $\bar{A}$ and $\tilde{A}$ by

$$\bar{A}(t)=\frac{1}{1-c}\sum_{j=1}^{n}c_j\bar{H}^{(j)}/\bar{H}^{(0)}, \quad (28)$$

$$\tilde{A}(t)=\frac{1}{1-c}\sum_{j=1}^{n}c_j\tilde{H}^{(j)}/\bar{H}^{(0)}, \quad (29)$$

An alternative form of the differential scheme follows from equations (27)-(29),

$$(1+\bar{A})dk+kd\bar{A}=d\left[\sum_{j=1}^{n}k_j\frac{c_j}{1-c}\bar{H}^{(j)}/\bar{H}^{(0)}\right] + \sum_{j=1}^{n}(k-k_j)\frac{c_j}{1-c}d[\bar{H}^{(j)}/\bar{H}^{(0)}]. \quad (30)$$

This equation is integrable if it is *assumed* that

$$\bar{H}^{(j)}(t)/\bar{H}^{(0)}(t)=\bar{H}^{(j)}(0)/\bar{H}^{(0)}(0), \quad j=1, 2, \ldots, n, \quad (31)$$

and

$$\tilde{H}^{(j)}=\bar{H}^{(j)}, \qquad j=1, 2, \ldots, n. \quad (32)$$

Then $\bar{A}=\tilde{A}$, and integration of equation (30) subject to the initial conditions $k(0)=k_0$, $c_j(0)=0$, $j\geq 1$, gives precisely equation (5) with $\bar{H}^{(j)}/\bar{H}^{(0)}$ equal to its dilute concentration value. Thus, the differential scheme yields the Mori-Tanaka effective conductivity if equations (31) and (32) hold.

Equations (31) and (32) combined imply that the ratio $\bar{H}^{(j)}(t)/\bar{H}^{(0)}(t)$ remains constant and equal to the dilute value throughout the process. Thus, at each stage of the process, the effective k is given by the Mori-Tanaka method for that concentration. Equation (32) requires, in addition, that the average field in the incrementally added volumes be the same as the bulk average for that phase. It is interesting to note that (32) is not required a priori for two-phase composites, $n=1$, but follows as a consequence of assuming (31). To see this, consider the differential equation (26) for the two-phase composite, which becomes using (27),

$$dk=(k_1-k)\,\frac{\tilde{H}^{(1)}/\tilde{H}^{(0)}}{(1-c_1+c_1\tilde{H}^{(1)}/\tilde{H}^{(0)})}\,\frac{dc_1}{(1-c_1)}. \tag{33}$$

However, it follows from equation (4) with $k^{(\mathrm{eff})}\equiv k$, and $\bar{H}=(1-c_1)\bar{H}^{(0)}+c_1\bar{H}^{(1)}$, that

$$\frac{\bar{H}^{(1)}}{\bar{H}^{(0)}}=\frac{(1-c_1)}{c_1}\left(\frac{k-k_0}{k_1-k}\right). \tag{34}$$

Substituting from equation (34) into equation (33) gives

$$dk=\frac{(k_1-k)^2}{(k_1-k_0)}\,\frac{\tilde{H}^{(1)}}{\bar{H}^{(0)}}\,\frac{dc_1}{(1-c_1)^2}. \tag{35}$$

This equation can be integrated directly if (31) holds for $j=1$. The equality $\tilde{H}^{(1)}=\bar{H}^{(1)}$ then follows by substituting the resulting k into (34).

4.3 Differential Effective Medium Theory for Multiphase Elastic Media. The differential equation for the effective moduli $\mathbf{L}(t)$ can be derived in a manner similar to the derivation of equation (26). Thus,

$$d\mathbf{L}=\sum_{j=1}^{n}(\mathbf{L}_j-\mathbf{L})\tilde{\mathbf{T}}_{j0}\left[(1-c)\mathbf{I}+\sum_{i=1}^{n}c_i\bar{\mathbf{T}}_{i0}\right]^{-1}\left(dc_j+c_j\,\frac{dc}{1-c}\right), \tag{36}$$

where the initial conditions are $\mathbf{L}(0)=\mathbf{L}_0$, $c_j(0)=0$, $j=1, 2, \ldots, n$. The strain concentration tensors $\tilde{\mathbf{T}}_{j0}, j=1, 2, \ldots, n$ in equation (36) compare the strain in the currently added phase j to the strain in phase 0 throughout the composite. The tensors $\bar{\mathbf{T}}_{j0}$ compare the bulk strain in phase j to the bulk strain in phase 0.

In the same way that the differential equation (26) was shown to be integrable if equations (31) and (32) hold, so it can be shown that equation (36) is exactly integrable if both

$$\tilde{\mathbf{T}}_{j0}(t)=\tilde{\mathbf{T}}_{j0}(0), \qquad j=1, 2, \ldots, n, \tag{37}$$

and

$$\tilde{\mathbf{T}}_{j0}=\bar{\mathbf{T}}_{j0}, \qquad j=1, 2, \ldots, n. \tag{38}$$

The case of a two-phase composite is again the exception. The strain concentration tensor $\bar{\mathbf{T}}_{10}$ can then be expressed in terms of the effective moduli as

$$\bar{\mathbf{T}}_{10}=\left(\frac{1-c_1}{c_1}\right)(\mathbf{L}-\mathbf{L}_0)(\mathbf{L}_1-\mathbf{L})^{-1}, \tag{39}$$

which when eliminated from equation (36) gives,

$$d\mathbf{L}=(\mathbf{L}-\mathbf{L}_1)\tilde{\mathbf{T}}_{10}(\mathbf{L}_1-\mathbf{L}_0)^{-1}(\mathbf{L}-\mathbf{L}_1)\,\frac{dc_1}{(1-c_1)^2}. \tag{40}$$

This differential equation depends only upon $\tilde{\mathbf{T}}_{10}$, and can be directly integrated if it is constant. The resultant moduli when substituted into equation (39) give $\bar{\mathbf{T}}_{10}=\tilde{\mathbf{T}}_{10}$, and the moduli themselves are identical to those of the Mori-Tanaka method. It has come to my attention that Laws (1980) also derived the Mori-Tanaka effective medium equation for a two phase composite as a hybrid of the differential scheme and the self-consistent method.

5 Discussion and Conclusions

Our major findings concern the relationship of the Mori-Tanaka approximation to the Hashin-Shtrikman and Hill-Hashin bounds. These bounds will always be satisfied when the approximation is used for two-phase composites. However, this result does not generalize to multiphase media, as demonstrated by a counter example in Section 2.

We have shown that the Mori-Tanaka method can be formulated in terms of the generalized differential scheme provided certain conditions are satisfied by the average fields in the latter. The condition for two-phase media is that the ratio of the field in the incrementally added particles to that in the bulk matrix phase remains constant as the concentration of the added phase goes from zero to its final, finite value. The Mori-Tanaka method, in its "direct approach" formulation, requires that the ratio of the field in the *bulk* added phase to that in the matrix equals its dilute concentration value. It is perhaps surprising that the condition on the incremental field ratio produces the same results as the Mori-Tanaka condition for two-phase media. This is particularly so since the same condition does not suffice to yield the Mori-Tanaka results for multiphase media.

The relationship of the Mori-Tanaka method with the differential scheme offers an alternative way of looking at the former. However, it is not clear whether the relevant conditions, (31)-(32) or (37)-(38), can be realized by specific microgeometries. The answer is, in general, no, since we have shown that the Mori-Tanaka method for multiphase media can give moduli outside the limits of the Hashin-Shtrikman bounds. That it may be possible in two-phase composites is suggested by the fact that the Mori-Tanaka approximation satisfies the bounds, and also because the differential scheme condition (31) or (37) is simpler than that for multiphase media. It remains as an interesting and worthwhile challenge to provide a realization of the method for two phases.

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APPENDIX

Alternative Formulation of the Mori-Tanaka Method

For an average stress in the composite of $\bar{\sigma}$, define the corresponding strain $\hat{\epsilon}_0$ in pure matrix material,

$$\bar{\sigma} = \mathbf{L}_0 \hat{\epsilon}_0. \tag{A1}$$

The perturbed stress and strain in phase 0 in the composite are $\tilde{\sigma}$ and $\tilde{\epsilon}$, where

$$\bar{\sigma} + \tilde{\sigma} = \mathbf{L}_0(\hat{\epsilon}_0 + \tilde{\epsilon}). \tag{A2}$$

The additional perturbed stress and strain in phase i, $i>0$, are σ_i^{pt} and ϵ_i^{pt}, where

$$\begin{aligned} \bar{\sigma} + \tilde{\sigma} + \sigma_i^{pt} &= \mathbf{L}_i(\hat{\epsilon}_0 + \tilde{\epsilon} + \epsilon_i^{pt}), \\ &= \mathbf{L}_0(\hat{\epsilon}_0 + \tilde{\epsilon} + \epsilon_i^{pt} - \epsilon_i^*), \end{aligned} \tag{A3}$$

and ϵ_i^* is the transformation strain in phase i. Taking the average strain throughout the composite, and using ($A1$), gives

$$\tilde{\epsilon} = \sum_{i=1}^{n} c_i(\epsilon_i^* - \epsilon_i^{pt}). \tag{A4}$$

Equation ($A4$) then implies that the average strain is

$$\bar{\epsilon} = \hat{\epsilon}_0 + \sum_{i=1}^{n} c_i \epsilon_i^*. \tag{A5}$$

The effective moduli are defined by $\bar{\sigma} = \mathbf{L}\bar{\epsilon}$, or from ($A1$) and ($A5$),

$$\mathbf{L}\left(\hat{\epsilon}_0 + \sum_{i=1}^{n} c_i \epsilon_i^*\right) = \mathbf{L}_0 \hat{\epsilon}_0. \tag{A6}$$

Some relations between ϵ_i^* and $\hat{\epsilon}_0$ are necessary to solve ($A6$) for $\mathbf{L}$.

For each $i = 1, 2, \ldots, n$ let

$$\epsilon_i^{pt} = \mathbf{S}_{i0} \epsilon_i^*. \tag{A7}$$

The average strain in the matrix phase 0 is, from ($A2$)

$$\bar{\epsilon}_0 = \hat{\epsilon}_0 + \tilde{\epsilon}. \tag{A8}$$

Equations ($A3$), ($A7$), ($A8$), and (12) imply, for each $i = 1, 2, \ldots, n$,

$$\epsilon_i^* = -\mathbf{L}_0^{-1}(\mathbf{L}_i - \mathbf{L}_0)\bar{\mathbf{T}}_{i0}\bar{\epsilon}_0. \tag{A9}$$

Then, equations ($A4$), ($A7$)–($A9$), and (12) give

$$\hat{\epsilon}_0 = \left(c_0 \mathbf{I} + \sum_{i=1}^{n} c_i \mathbf{L}_0^{-1} \mathbf{L}_i \bar{\mathbf{T}}_{i0}\right)\bar{\epsilon}_0. \tag{A10}$$

Equations ($A9$) and ($A10$) can then be substituted into ($A6$) to given an explicit expression for $\mathbf{L}$ that is identical to equation (11). The Mori-Tanaka assumption is that $\mathbf{S}_{i0}$ is equal to the corresponding Eshelby tensor for a single inclusion of phase i in phase 0.