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Bounds and estimates for elastic constants of random polycrystals of laminates

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Abstract

In order to obtain formulas providing estimates for elastic constants of random polycrystals of laminates, some known rigorous bounds of Peselnick, Meister, and Watt are first simplified. Then, some new self-consistent estimates are formulated based on the resulting analytical structure of these bounds. A numerical study is made, assuming first that the internal structure (i.e., the laminated grain structure) is not known, and then that it is known. The purpose of this aspect of the study is to attempt to quantify the differences in the predictions of properties of the same system being modelled when such internal structure of the composite medium and spatial correlation information is and is not available.

1. Introduction

In the history of studies of random heterogeneous media, the earliest work on electrical, elastic, and viscous media (Maxwell, 1873; Rayleigh, 1892; Einstein, 1905; Voigt, 1928; Reuss, 1929; Bruggeman, 1935) involved ad hoc procedures intended to provide sensible estimates of the physical constants of interest in such systems. Much later the early work on bounding methods first showed that some of the known estimates were in fact rigorous bounds (Hill, 1952) and subsequently produced quite accurate and useful bounds (Hashin and Shtrikman, 1962) that were then proven to be optimal in the sense that for certain special classes of microstructures the bounding values could be attained. Later still it was established that certain choices of the ad hoc estimates or effective medium theories had special relationships to the bounds. In particular some of these estimates were shown always to lie between the rigorous upper and lower bounds on the material constants (Berryman, 1980a,b, 1982; Milton, 1985; Norris, 1985; Avellaneda, 1987).

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Bounding methods obviously have the great advantage of rigor, but the disadvantage that, for real material constants and each fixed choice of volume fraction, there are two numbers and, for complex material constants, a closed curve in the complex plane describing the bounds (Gibiansky and Milton, 1993; Milton and Berryman, 1997; Gibiansky et al., 1999). But, for practical applications, users often want estimates instead of such bounds—especially in applications to porous media where the bounds may commonly be far apart, and the lower bound often vanishes. Clearly estimates together with additional measures of the probable range of errors in those estimates would be highly desirable for many applications. And surely if bounds are available, then estimates can often be found. Hill (1952) resolved this dilemma famously by averaging (i.e., using either the mean or the geometric mean) the well-known Voigt and Reuss bounds for elastic constants, thereby producing the very well-known Voigt–Reuss–Hill (VRH) estimates. In other cases, known estimates have already been shown to lie between the bounds, but in fact if the analytical form of the bounds had been known first, then often these common estimates could very easily have been deduced directly from the bounds (Berryman, 1982).

The author has recently shown (Berryman, 2004b) how the Peselnick and Meister bounds (Peselnick and Meister, 1965) for random polycrystals of laminates can be used to provide both bounds and self-consistent estimates of the shear modulus in the special case of heterogeneous elastic media having constant bulk modulus. The present paper will expand on this idea by showing that in general the algorithmic form of the Peselnick–Meister–Watt bounds (Peselnick and Meister, 1965; Watt and Peselnick, 1980) for hexagonal crystals can be simplified into explicit formulas, and subsequently rewritten so it is straightforward to obtain self-consistent estimates for both bulk and shear modulus for this same type of random polycrystal of laminates. But no restrictions (except the usual physical ones, such as positivity) need to be imposed here on the range of values present for bulk or shear moduli in the composite.

The main focus of the paper is to present a new formulation of the rigorous bounds on bulk and shear modulus for any isotropic random polycrystal whose component crystals have hexagonal symmetry. The presentation is limited here to results for hexagonal symmetry because this is the one relevant to the model material considered, i.e., the random polycrystal of laminates. This model is especially useful for showing directly how much improvement (tightening of the bounds) is achieved when something is known about the microstructure, as it is in this model. The more commonly studied Hashin–Shtrikman bounds (Hashin and Shtrikman, 1961, 1963) use only the volume fraction information, whereas in this model we have both volume fraction information and also some very precise knowledge of the local spatial arrangement of the constituents in the layers.

The second section presents the model of random polycrystals of laminates (hereafter called "the model") and provides some details of the basic analysis. Then the third section shows how to simplify the Hashin–Shtrikman-type bounds of Peselnick, Meister, and Watt to analytical forms for both bulk and shear moduli. The fourth section shows how to construct self-consistent estimates from these analytical bounds. Examples of the use of the formulas are presented in the fifth section. The results are discussed and the conclusions are summarized in the final section. Appendix A summarizes some recent results needed in the main text concerning certain product formulas in hexagonal anisotropic media. Appendix B displays the original version of the Peselnick–Meister–Watt bounds and also shows two examples of the details of the algorithmic calculations, together with the final results for the self-consistent estimates.

2. Structure of the model

2.1. Elasticity of layered materials

We assume that a typical building block of the random system is a small grain of laminate material whose elastic response for such a transversely isotropic (hexagonal) system can be described by:

$$\begin{pmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33} \\ \sigma_{23} \\ \sigma_{31} \\ \sigma_{12} \end{pmatrix} = \begin{pmatrix} c_{11} & c_{12} & c_{13} & & & \\ c_{12} & c_{11} & c_{13} & & & \\ c_{13} & c_{13} & c_{33} & & & \\ & & & 2c_{44} & & \\ & & & & 2c_{44} & \\ & & & & & 2c_{66} \end{pmatrix} \begin{pmatrix} e_{11} \\ e_{22} \\ e_{33} \\ e_{23} \\ e_{31} \\ e_{12} \end{pmatrix}, \tag{1}$$

where σ_{ij} are the usual stress components for $i, j = 1$–3 in Cartesian coordinates, with 3 (or z) being the axis of symmetry (the lamination direction for such a layered material). Displacement u_i is then related to strain component e_{ij} by $e_{ij} = (\partial u_i/\partial x_j + \partial u_j/\partial x_i)/2$. This choice of definition introduces some convenient factors of two into the 44, 55, 66 components of the stiffness matrix shown in (1).

Although some of the results presented here are more general, we will assume for definiteness that this stiffness matrix in (1) arises from the lamination of N isotropic constituents having bulk and shear moduli K_n, μ_n, in the $N > 1$ layers present in each building block. It is important that the thicknesses d_n always be in the same proportion in each of these laminated blocks, so that $f_n = d_n/D$, where $D = \sum_{n'} d_{n'}$ is the sum over all layer thicknesses in one grain. But it is not important what order the layers were added to the blocks, as Backus's formulas (Backus, 1962) for the constants show. For the overall behavior for the quasistatic (long wavelength) behavior of the system we are studying, Backus's results [also see Postma (1955) and Milton (2002)] state that

$$\begin{aligned} c_{33} &= \left\langle \frac{1}{K + 4\mu/3} \right\rangle^{-1}, & c_{13} &= c_{33} \left\langle \frac{K - 2\mu/3}{K + 4\mu/3} \right\rangle, \\ c_{44} &= \left\langle \frac{1}{\mu} \right\rangle^{-1}, & c_{66} &= \langle \mu \rangle, \\ c_{11} &= \frac{c_{13}^2}{c_{33}} + 4c_{66} - 4\left\langle \frac{\mu^2}{K + 4\mu/3} \right\rangle, & c_{12} &= c_{11} - 2c_{66}. \end{aligned} \tag{2}$$

This bracket notation can be correctly viewed: (a) as a volume average, (b) as a line integral along the symmetry axis x_3, or (c) as a weighted summation $\langle Q \rangle = \sum_n f_n Q_n$ over any relevant physical quantity Q taking a constant value Q_n in the n-th layer.

2.2. Random polycrystals of laminates

For our general modeling problem having arbitrary (but non-negative) values of the μ_n's and K_n's, suppose we construct a random polycrystal by packing small bits of this laminate material into a large container (see Fig. 1) in a way so that the axis of symmetry appears randomly over all possible orientations and also such that no misfit of surfaces (and therefore no porosity) is left in the resulting composite. [*Note*. It is known that small amounts of misfit porosity can make a significant difference to the results (Berryman, 1994), but we will not study this issue here.] If the ratio of grain size to overall composite is small enough so the usual implicit assumption of scale separation applies to the composite—but not so small that we are violating the continuum hypothesis—then we have an example of the type of material we want to study.

For each individual grain in this polycrystal, Eqs. (2) are valid locally (i.e., for locally defined coordinates), and the grain bulk modulus K_R is given by (20) for all the grains. The factors $3K_R$ and $2G_{eff}^{v}$ are not necessarily eigenvalues of elastic stiffness for individual grains. The Voigt average for shear is given in Appendix A by (22), which is an upper bound on the isotropic shear modulus of the random polycrystal (Hill, 1952).

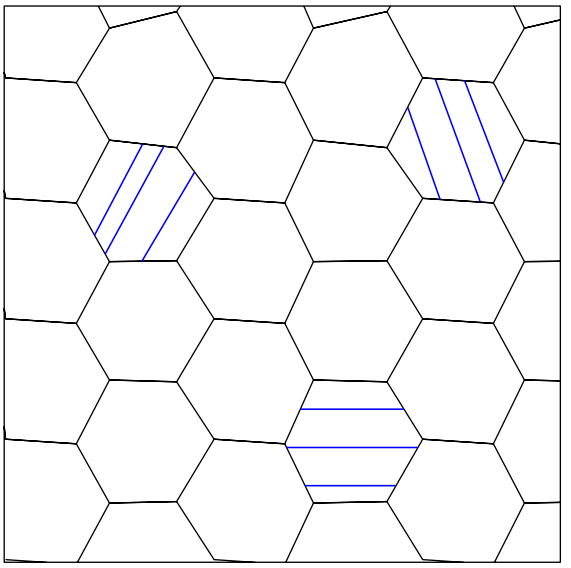

Fig. 1. Schematic illustrating the random polycrystals of laminates model. Grains are assumed to fit tightly so there is no porosity. But the shapes of the grains are not necessarily the same, and the symmetry axes of the grains (three examples are shown here) are randomly oriented so the overall polycrystal is equiaxed (statistically isotropic).

We distinguish between "correlated" and "uncorrelated" bounds. For example, the most familiar bounds—after the uncorrelated Voigt and Reuss bounds (i.e., the volume averaged mean and harmonic mean respectively of the constituents' constants)—are the uncorrelated Hashin–Shtrikman bounds:

$$K_{\rm HS}^{\pm} = \left[\sum_{n=1}^{N} \frac{f_n}{K_n + 4\mu_{\pm}/3}\right]^{-1} - 4\mu_{\pm}/3 \tag{3}$$

and

$$\mu_{\rm HS}^{\pm} = \left[\sum_{n=1}^{N} \frac{f_n}{\mu_n + \zeta_{\pm}}\right]^{-1} - \zeta_{\pm}, \tag{4}$$

where

$$\zeta_{\pm} = \frac{\mu_{\pm}}{6}\left(\frac{9K_{\pm} + 8\mu_{\pm}}{K_{\pm} + 2\mu_{\pm}}\right), \tag{5}$$

with K_+ and K_- being the highest and lowest values of K_n in the system, and similarly $\mu_+(\mu_-)$ being the highest (lowest) value of the shear moduli μ_n. By greatly simplifying several earlier presentations, Milton (1981), developed examples of correlated bounds where the correlations were introduced explicitly through spatial correlation functions. But here we introduce correlations instead through the grain laminations. The Voigt and Reuss bounds (20)–(23) are then considered correlated because of the assumed internal grain-like structure. Some limited numerical comparisons of these bounds, and a few others, to the Peselnick–Meister–Watt bounds (Peselnick and Meister, 1965; Watt and Peselnick, 1980) were given previously by the author (Berryman, 2004a). It was found that the best and also most relevant bounds were clearly the Peselnick–Meister–Watt bounds (Peselnick and Meister, 1965; Watt and Peselnick, 1980), which are presented again in Appendix B.

3. Elastic constant bounds for the model

3.1. Simplified bounds on bulk modulus

The formulas for the Hashin–Shtrikman-type bounds on polycrystals of grains having hexagonal symmetry are summarized in Appendix B. These bounds were derived by Peselnick and Meister (1965) with a correction added later by Watt and Peselnick (1980). The bounds are expressed algorithmically—not as formulas. In order to gain the insight needed to deduce (later in this paper) effective medium approximations based on such bounds, it is most helpful to have explicit formulas. So it will be our goal to find appropriate expressions for bulk modulus and, in the next subsection, also for shear modulus.

The main observation that helps us to find such formulas in this case is based on the easily verified fact that

$$1 + 2\beta_\pm G_\pm = -2\beta_\pm \zeta_\pm, \tag{6}$$

where $\zeta_\pm = \frac{G_\pm}{6}\left(\frac{9K_\pm + 8G_\pm}{K_\pm + 2G_\pm}\right)$ is defined exactly as in (5), but the meanings of the arguments $G_\pm$ and $K_\pm$ differ. In (6), the values $G_\pm$ and $K_\pm$ are those given in Appendix B, having the significance of the shear and bulk moduli of the isotropic comparison material. In (5), the bulk and shear moduli are the lowest and highest values present among all the values found in the system. In contrast, for the values in (6) and Appendix B, we typically have $G_- = c_{44}$ and $G_+ = c_{66}$, and then the values of $K_\pm$ are computed from (29)–(31). In (5), the values of $K_\pm$ are just the highest and lowest bulk modulus values in the system—thus resulting in the Hashin–Shtrikman–Walpole bounds (Walpole, 1966)—and not necessarily very closely correlated with the layer bounding values $\mu_\pm$.

To obtain the desired result for bulk modulus, first rearrange (24) into the form

$$K_{\mathrm{PM}}^{\pm} = \frac{K_{\mathrm{V}} + K_\pm 2\beta_\pm (G_\pm - G_{\mathrm{eff}}^{\mathrm{v}})}{1 + 2\beta_\pm (G_\pm - G_{\mathrm{eff}}^{\mathrm{v}})}. \tag{7}$$

Then, making use of (29), we have

$$K_{\mathrm{PM}}^{\pm} = \frac{K_{\mathrm{V}}[1 + 2\beta_\pm (G_\pm - G_{\mathrm{eff}}^{\mathrm{r}})]}{1 + 2\beta_\pm (G_\pm - G_{\mathrm{eff}}^{\mathrm{v}})}. \tag{8}$$

And, finally, substituting (6) into (8), we obtain the desired result

$$K_{\mathrm{PM}}^{\pm} = \frac{K_{\mathrm{V}}(G_{\mathrm{eff}}^{\mathrm{r}} + \zeta_\pm)}{(G_{\mathrm{eff}}^{\mathrm{v}} + \zeta_\pm)}. \tag{9}$$

This is the main result of this sub-section.

Note that $\zeta_\pm$ is a monotonic function of both arguments $G_\pm$ and $K_\pm$. As $K_\pm$ ranges from 0 to ∞ for fixed $G_\pm$ (which can happen as the model parameters vary), $\zeta_\pm$ lies in the bounded range $\frac{2}{3}G_\pm \leqslant \zeta_\pm \leqslant \frac{3}{2}G_\pm$. As $G_\pm$ varies from 0 to ∞, $\zeta_\pm$ also ranges from 0 to ∞. In particular, when $\zeta_- = 0$, we have

$$K_{\mathrm{PM}}^{-} = K_{\mathrm{R}} \tag{10}$$

and, when $\zeta_+ = \infty$, we have

$$K_{\mathrm{PM}}^{+} = K_{\mathrm{V}}, \tag{11}$$

which are obviously the lower and upper bounds on K given by Reuss and Voigt, respectively. Thus, this analytical formula parameterizes the bounds in terms of the parameters $\zeta_\pm$, which are still determined by the formulas given for $G_\pm$ and $K_\pm$ in Appendix B. But, as we will soon see, the formula (9) has the advantage that it is easy to use as the basis for an effective medium approximation.

An interesting limit that is readily checked is the case of constant layer shear modulus, $\mu = \text{const.}$ Then, $G^{\text{v}}_{\text{eff}} = G^{\text{r}}_{\text{eff}}$, and from the product formula [see Appendix A and/or Berryman (2004a)], $K_{\text{V}} = K_{\text{R}}$. In this special case, the grains are actually isotropic, and the grain bulk modulus K^* satisfies Hill's equation

$$K^* = \left[\sum_{n=1}^{N} \frac{f_n}{K_n + 4\mu/3}\right]^{-1} - 4\mu/3. \tag{12}$$

Furthermore, since $K_{\text{V}} = K_{\text{R}}$, this is the correct result also for the overall bulk modulus $K^* = K_{\text{V}}$ of the system we are modeling. It is easy to check that Backus's formulas give exactly (12) for this limiting case.

3.2. Simplified bounds on the shear modulus

To find the simplified version of (25) for the overall shear modulus, we first shift $G_\pm$ to the left-hand side, then multiply by $-2\beta_\pm$, and add unity to both sides of the result. We find that

$$[1 + 2\beta_\pm(G_\pm - \mu^\pm_{\text{PM}})] = \frac{1}{1 + 2\beta_\pm B_2^\pm}. \tag{13}$$

Using (6) to simplify the left-hand side, we then have

$$\mu^\pm_{\text{PM}} + \zeta_\pm = -\frac{1}{2\beta_\pm(1 + 2\beta_\pm B_2^\pm)}. \tag{14}$$

The right-hand side of (14) can be greatly simplified. When this (rather tedious algebra) has been accomplished, the formula (14) can be inverted to give

$$\frac{1}{\mu^\pm_{\text{PM}} + \zeta_\pm} = \frac{1}{5}\left[\frac{1 - \alpha_\pm(K_{\text{V}} - K_\pm)}{G^{\text{v}}_{\text{eff}} + \zeta_\pm + \frac{\alpha_\pm}{2\beta_\pm}(K_{\text{V}} - K_\pm)} + \frac{2}{c_{44} + \zeta_\pm} + \frac{2}{c_{66} + \zeta_\pm}\right], \tag{15}$$

which is the desired form of the Peselnick–Meister–Watt bounds on overall shear modulus of a polycrystal of grains having hexagonal symmetry. Parameters $\alpha_\pm$, $\beta_\pm$ are defined in (26). This is the main result of this subsection.

To check one known limit of this formula, consider the case when $K_n = K = \text{const.}$ considered previously in Berryman (2004b). Then, $K_{\text{V}} = K = K_\pm$, and the formula (15) reduces correctly to

$$\mu^\pm_{\text{PM}} = \left[\frac{1}{5}\left(\frac{1}{G^{\text{v}}_{\text{eff}} + \zeta_\pm} + \frac{2}{c_{44} + \zeta_\pm} + \frac{2}{c_{66} + \zeta_\pm}\right)\right]^{-1} - \zeta_\pm. \tag{16}$$

If we choose $\zeta_\pm = 0$ or ∞, then (16) reduces to the formulas (23) and (22) for the correlated Reuss and Voigt bounds on the polycrystal's overall shear modulus. For this special case, the $\zeta_- \to 0$ limit is correct because $G^{\text{v}}_{\text{eff}} = G^{\text{r}}_{\text{eff}}$ when $K_n = \text{const.}$

Next, without placing any special restrictions on the layer constants, we can also check whether (15) reduces correctly to (22) as $\zeta_+ \to \infty$ and (23) as $\zeta_- \to 0$. When $\zeta_+ \to \infty$, $K_+ \to K_{\text{V}}$, so (15) reduces to (16) in this limit, and therefore performs as it should. As $\zeta_- \to 0$, $K \to K_{\text{R}}$, $\alpha_- \to -1/K_{\text{R}}$, and $\beta_- \to \infty$. The expected result (23) is then obtained because $K_{\text{V}}/G^{\text{v}}_{\text{eff}}K_{\text{R}} = 1/G^{\text{r}}_{\text{eff}}$ follows from the product formulas.

Uniaxial shear energy $G^{\text{v}}_{\text{eff}}$ plays a dominant role in both formulas (15) and (16)—along with c_{44} and c_{66}—even though $2G^{\text{v}}_{\text{eff}}$ is only rarely an eigenvalue of this system, while $2c_{44}$ and $2c_{66}$ are both always eigenvalues (twice over).

4. Elastic constant estimates for the model

We are now in position to create some useful effective medium approximations based on the formulas for the rigorous bounds (9) and (15) derived in the previous section. In each case the choices to be made seem quite apparent based both on the form of the bounds, and on prior experiences with other bounds and self-consistent estimates. The resulting formulas obtained this way will be called the "self-consistent" or SC estimate based on these correlated bounds.

We take the self-consistent estimate for bulk modulus to be

$$K^* = \frac{K_V(G^r_{eff} + \zeta^*)}{(G^v_{eff} + \zeta^*)} = \frac{G^v_{eff}K_R + \zeta^* K_V}{G^v_{eff} + \zeta^*}, \tag{17}$$

where

$$\zeta^* = \frac{\mu^*}{6}\left(\frac{9K^* + 8\mu^*}{K^* + 2\mu^*}\right). \tag{18}$$

In (18), K^* is determined by (17) and μ^* is determined by the self-consistent expression for the shear modulus to follow. In fact, μ^* is obtained similarly from (15) and we have

$$\frac{1}{\mu^* + \zeta^*} = \frac{1}{5}\left[\frac{1 - \alpha^*(K_V - K^*)}{G^v_{eff} + \zeta^* + \frac{\alpha^*}{2\beta^*}(K_V - K^*)} + \frac{2}{c_{44} + \zeta^*} + \frac{2}{c_{66} + \zeta^*}\right]. \tag{19}$$

The parameters α^* and β^* are defined as in (26) taking $K_\pm \to K^*$ and $G_\pm \to \mu^*$. In all cases, these formulas are obtained by replacing the terms in the bounds everywhere so that $K_\pm \to K^*$ and $G_\pm \to \mu^*$. The result is a set of fairly complicated coupled equations that are most conveniently solved by numerical iteration.

This iteration process is expected to converge rapidly to definite unique answers for both K^* and μ^*, and especially so if it can be shown that the individual formulas are monotonic functionals of their arguments. It is well-known that ζ^* is a monotonic functional of both arguments (Berryman, 1982). It is also quite easy to check that K^* is a monotonic functional of ζ^*. Since $K^* \leqslant K_V$ will always be satisfied, μ^* is easily shown to

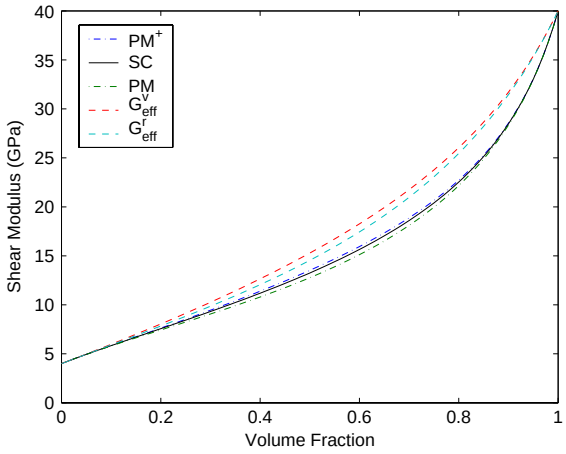

Fig. 2. Comparison of the Peselnick–Meister shear modulus bounds and the self-consistent estimate over all choices of volume fraction of the stiffer component, for the same case considered in Figs. 3 and 4. Here the uniaxial shear energies G^v_{eff} (strain) and G^r_{eff} (stress) are both plotted as well to show that the condition $G^v_{eff} \geqslant \mu^*_{SC}$, which is the condition found sufficient to guarantee monotonicity of the self-consistent shear functional, is easily satisfied in all cases.

be a monotonic functional of ζ^*. The only remaining issue to check is whether μ^* is also a monotonic functional of K^*. A rather tedious analysis (which will therefore not be shown here) indicates that μ^* is indeed a monotonic functional of K^* as long as $\mu^* \leqslant G^{\rm v}_{\rm eff}$. However, since $G^{\rm v}_{\rm eff}$ is not the overall Voigt average of the shear modulus (but rather the energy per unit volume of the uniaxial shear component), it is not immediately clear that this inequality will necessarily be obeyed. We point out however that Fig. 5 of Berryman (2004b) showed explicitly, for the special case of $K_n = \text{const.}$, that $\mu^-_{\rm PM} \leqslant \mu^*_{\rm SC} \leqslant \mu^+_{\rm PM} \leqslant G^{\rm v}_{\rm eff}$. So it is quite possible that $\mu^*_{\rm SC} \leqslant G^{\rm v}_{\rm eff}$ is generally true—at least for the random polycrystal of laminates model. In any case, the iteration scheme itself tells us quickly enough whether the functional is behaving well or not; if not, then the convergence will either be slow or non-existent, i.e., jumping from one unstable point in the (μ^*, K^*) space to another. Such unpleasant behavior has not been observed.

Fig. 2 illustrates the behavior found in practice for the examples that follow. Both the bounds and the self-consistent estimates always lie below the two values $G^{\rm v}_{\rm eff}$ and $G^{\rm r}_{\rm eff}$. Convergence is generally found in 2 or 3 iterations, as might be expected from the narrow range of values permitted by the bounds.

5. Examples

Figs. 3 and 4 illustrate the results obtained so far in the text. Fig. 3 presents the bulk modulus results, and Fig. 4 presents the shear modulus results. Three types of bounds (both upper and lower) are shown along with two effective medium estimates. The Hashin–Shtrikman bounds ($\rm HS^{\pm}$) for an isotropic composite having isotropic constituents, when no information about spatial correlation functions or other types of local ordering in the medium are known. These bounds are the outer most bounds shown here. (There are also uncorrelated Voigt and Reuss bounds available, but these fall outside of the all the ones presented here. In fact, the Voigt bound is just a straight line between the two end points in each case.)

The next best bounds are the Voigt and Reuss bounds based on knowledge of the crystalline nature of the aggregate (VX and RX). Although these bounds are somewhat crude, they are nevertheless better/tighter bounds than the ''uncorrelated'' Hashin–Shtrikman bounds.

The best bounds are the Hashin–Shtrikman-type bounds of Peselnick, Meister, and Watts ($\rm PM^{\pm}$). These bounds take the local correlations of the crystalline (laminated) components into account, and are very

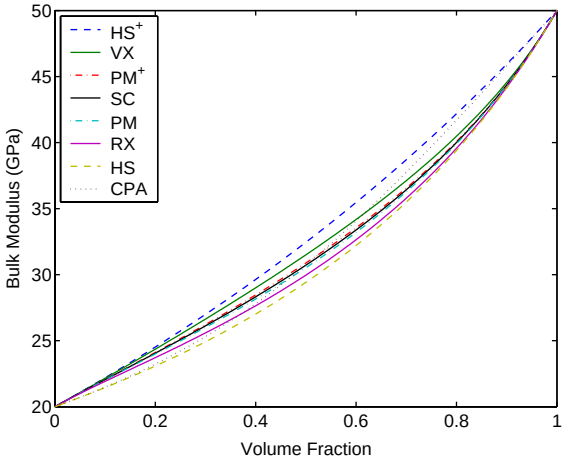

Fig. 3. Comparison of various bulk modulus bounds and estimates from the text. The abscissa is the volume fraction of the stiffer component. This stiff component has $K = 50$ GPa and $\mu = 40$ GPa, while the compliant component has $K = 20$ GPa and $\mu = 4$ GPa.

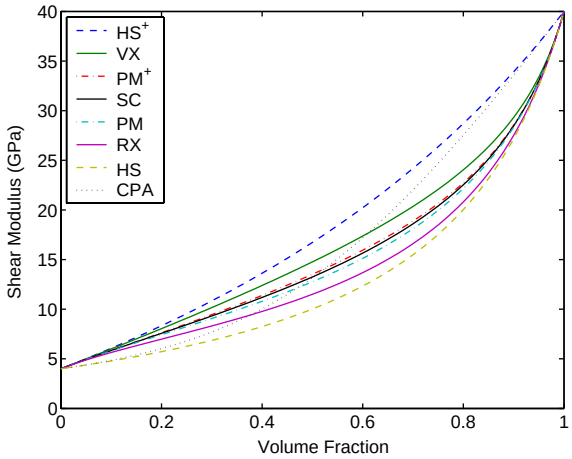

Fig. 4. Comparison of various shear modulus bounds and estimates from the text. Component elastic constant values and significance of the volume fractions are the same as in Fig. 3.

accurate bounds in the sense that they are very close to each other for the present, fairly high contrast, example. Then, the self-consistent estimate SC (based as it is on the analytical form of the PM bounds) is seen to fall within these tightest bounds, as expected.

In contrast, the same type of self-consistent bound—for which we use the technical term the CPA (for "coherent potential approximation") and which is based instead on the original "uncorrelated" Hashin–Shtrikman bounds [by taking $K_{\pm} \to K^{*}$ and $\mu_{\pm} \to \mu^{*}$ in Eqs. (3) and (4)]—does not do very well here. At least it is not accurately reflecting the true microstructure when the components are implicitly assumed to be spherical in shape (as is true in this way of constructing the CPA from $HS^{\pm}$). In this illustration, for both the bulk and shear estimates, it starts out hugging the lower bound HS^{-} and then cuts across the diagram and ends up hugging the upper bound HS^{+}. It is clear that this behavior is wrong for this particular type of composite because the layering in the grains tends to emphasize the weaker component at high concentrations of the strong component, and the stronger component at low concentrations of the stronger component. This type of behavior could have been obtained from the CPA, but it would have required input of additional information about the microstructure [and also a different type of derivation—see Berryman (1980b)]. We expect (and can easily confirm numerically) that the CPA will give quite similar results to those observed here if instead the components were assumed to be penny-shaped or disk-shaped objects, which would have been more consistent with the actual microstructure implicit in the "random polycrystal of laminates" model.

6. Conclusions and discussion

Formulas (9) and (15) are general rigorous bounds for the elastic constants of any random polycrystal having crystalline grains with hexagonal symmetry. Similarly, Eqs. (17) and (19) are the self-consistent estimates for the elastic constants of any random polycrystal having crystalline grains with hexagonal symmetry derived directly from these bounds.

Traditional effective medium theories have typically been formulated using physical arguments to arrive at thought experiments leading to definite predictions about the behavior of complex systems. A small subset of these formulations (Milton, 1985; Norris, 1985; Avellaneda, 1987) has been shown to correspond to realizable (at least in principle) microstructures and, therefore, to the conclusion that these

approximations should always satisfy any rigorous bounds known for the physical constants. But, such realizability conditions are *not* always easy to establish and *are always* subject to the criticism (Christensen, 1990) that, even though the implicit microstructure is realizable, it is nevertheless not the pertinent microstructure for the system we need to study in the laboratory or in the field—being instead a typically hierarchical microstructure (Milton, 1985) requiring many levels for validity of the required separation of scales.

But there are many bounds on physical constants available now (Milton, 2002). So the question arises: Can we make use of these bounds in developing new effective medium estimates rather than in justifying them after the fact? The present work has shown that this is possible. To achieve this goal in elasticity, it is most useful to have elasticity bounds expressed as a pair of formulas, and preferably formulas that have the same type of functional form. The original Peselnick–Meister–Watt bounds, for example, were expressed in terms of an algorithm – not as explicit formulas. Although it is obviously possible to arrive at estimates in either case (numerical curves can be averaged), it seems most useful to the author to have the formulas—for then formulas for the estimates are most easily and intuitively obtained. Whenever this is possible, the estimates presumably describe the physical behavior of the system for a typical microstructure from the ensemble of microstructures assumed in the original derivation of the bounds themselves.

Success here in simplifying the Peselnick–Meister bounds for random polycrystals having hexagonal symmetry also suggests that other bounds of this algorithmic type might also be simplified. Determining the answer to this question will be one direction of work to follow. Another direction will involve additional complications that are introduced when the model layers are porous and the pores are saturated or partially saturated by fluids. Then, the model may serve as a semi-analytical model for the geomechanics of earth reservoirs.

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Appendix A. Voigt and Reuss bounds and a product formula in elasticity

The bulk modulus for each laminated building block (or crystalline grain if you like) is that given by the compressional Reuss average K_R of the corresponding compliance matrix s_{ij} [the inverse of the usual stiffness matrix c_{ij}, whose non-zero matrix elements are shown in (1)]. The well-known result is $e = e_{11} + e_{22} + e_{33} = \sigma/K_{\text{eff}}$, where $1/K_{\text{eff}} = 1/K_R = 2s_{11} + 2s_{12} + 4s_{13} + s_{33}$. This quantity can be expressed in terms of the stiffness elements as

$$\frac{1}{K_R - c_{13}} = \frac{1}{c_{11} - c_{66} - c_{13}} + \frac{1}{c_{33} - c_{13}}. \tag{20}$$

The Voigt average for bulk modulus is well-known to be

$$K_V = [2(c_{11} + c_{12}) + 4c_{13} + c_{33}]/9. \tag{21}$$

Even though $K_{\rm eff}$ is the same for every grain, since the grains themselves are not isotropic, the overall bulk modulus K^* of the random polycrystal is not necessarily the same as the value $K_{\rm eff}$ for the individual grains (Hill, 1952). Hashin–Shtrikman bounds on K^* for random polycrystals whose grains have hexagonal symmetry (Peselnick and Meister, 1965; Watt and Peselnick, 1980). show in fact that the value $K_{\rm R}$ lies outside (actually below) the bounds in many situations (Berryman, 2004b).

In general an upper bound on the overall shear modulus of an isotropic polycrystal (Hill, 1952) is given by the Voigt average over shear of the stiffness matrix, which may be written as

$$\mu_{\rm V} = \frac{1}{5}(G^{\rm v}_{\rm eff} + 2c_{44} + 2c_{66}). \tag{22}$$

This expression can be taken as the definition of $G^{\rm v}_{\rm eff}$. Eq. (22) implies that $G^{\rm v}_{\rm eff} = (c_{11} + c_{33} - 2c_{13} - c_{66})/3$. In fact $G^{\rm v}_{\rm eff}$ is the energy per unit volume in a grain when a pure uniaxial shear strain of unit magnitude is applied to the grain along its axis of symmetry (Berryman, 2004a,b).

Then, the Reuss average for shear is

$$\mu_{\rm R} = \left[\frac{1}{5}\left(\frac{1}{G^{\rm r}_{\rm eff}} + \frac{2}{c_{44}} + \frac{2}{c_{66}}\right)\right]^{-1}, \tag{23}$$

which is also a rigorous lower bound on the overall shear modulus of the polycrystal (Hill, 1952).

For each grain having hexagonal symmetry, the product formulas $3K_{\rm R}G^{\rm v}_{\rm eff} = 3K_{\rm V}G^{\rm r}_{\rm eff} = \omega_+\omega_-/2 = c_{33}(c_{11} - c_{66}) - c_{13}^2$ are valid (Berryman, 2004a). The symbols $\omega_\pm$ stand for the quasi-compressional and quasi-uniaxial shear eigenvalues for all the grains. Whenever the bulk modulus in this model is uniform, the product formulas show immediately that $G^{\rm r}_{\rm eff} = G^{\rm v}_{\rm eff}K_{\rm R}/K_{\rm V} = G^{\rm v}_{\rm eff}$, since $K_{\rm R} = K_{\rm V} = K$. Thus, for this special case, pure compression or tension ($e_{11} = e_{22} = e_{33}$) is an eigenvector corresponding to stiffness eigenvalue $3K$. Uniaxial shear strain ($e_{33} = -2e_{11} = -2e_{22}$) is then also an eigenvector and $2G^{\rm v}_{\rm eff} = 2G^{\rm r}_{\rm eff}$ is the corresponding eigenvalue.

Appendix B. Peselnick–Meister–Watt bounds for hexagonal symmetry

Hashin–Shtrikman-style bounds (Hashin and Shtrikman, 1961, 1962, 1963) on the bulk and shear moduli of isotropic random polycrystals composed of hexagonal grains have been derived by Peselnick and Meister (1965), with later corrections by Watt and Peselnick (1980). Derivations will be found in the references. The structure of the algorithm for computing these bounds is illustrated in Figs. 5 and 6.

Parameters used to optimize the Hashin–Shtrikman bounds are $K_\pm$ and $G_\pm$, which have the significance of being the bulk and shear moduli of two isotropic comparison materials. G_+ and K_+ are the values used in the formulas for the upper bounds, and $G_-$and K_- for the lower bounds. Formulas for the bounds are

$$K^\pm_{\rm PM} = K_\pm + \frac{K_{\rm V} - K_\pm}{1 + 2\beta_\pm(G_\pm - G^{\rm v}_{\rm eff})} \tag{24}$$

and

$$\mu^\pm_{\rm PM} = G_\pm + \frac{B_2^\pm}{1 + 2\beta_\pm B_2^\pm}, \tag{25}$$

where

$$\alpha_\pm = \frac{-1}{K_\pm + 4G_\pm/3}, \quad \beta_\pm = \frac{2\alpha_\pm}{15} - \frac{1}{5G_\pm}, \quad \gamma_\pm = \frac{1}{9}(\alpha_\pm - 3\beta_\pm) \tag{26}$$

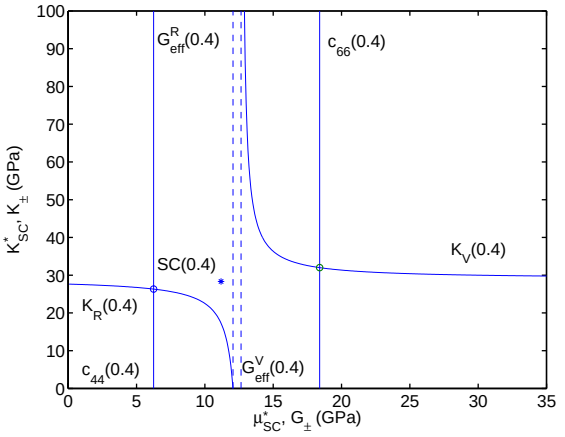

Fig. 5. Illustrating the graphical construction leading to the optimum parameters for the comparison material of the lower and upper Peselnick–Meister–Watt bounds (G_-, K_-), (G_+, K_+), shown as circles, and the self-consistent estimate (μ^*, K^*) obtained from the analytical form, shown as an asterisk. The case shown is for the volume fraction of the stiffer component from Figs. 3 and 4 given by $f_1 = 0.4$. Values of the constants entering the expressions (see Appendix A) are: $K_V = 29.0055$, $c_{44} = 6.2500$, $c_{66} = 18.4000$, $G^r_{eff} = 12.0571$, and $G^v_{eff} = 12.6506$, all in units of GPa. The two parts of the solid curve are determined by (29).

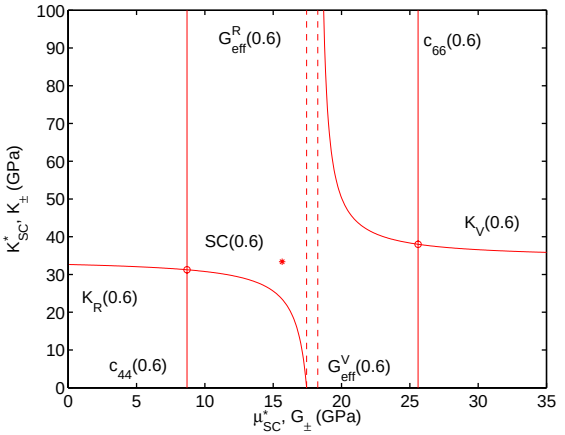

Fig. 6. Same as Fig. 5 for the volume fraction of the stiffer component from Figs. 3 and 4 given by $f_1 = 0.6$. Values of the constants entering the expressions are: $K_V = 34.1792$, $c_{44} = 8.6957$, $c_{66} = 25.6000$, $G^r_{eff} = 17.4441$ and $G^v_{eff} = 18.2641$, all in units of GPa.

and

$$B_2^{\pm} = \frac{1}{5}\left[\frac{G^v_{eff} - G_\pm}{\mathscr{D}_\pm} + \frac{2(c_{44} - G_\pm)}{1 - 2\beta_\pm(c_{44} - G_\pm)} + \frac{2(c_{66} - G_\pm)}{1 - 2\beta_\pm(c_{66} - G_\pm)}\right], \tag{27}$$

with

$$\mathscr{D}_\pm = 1 - \beta_\pm(c_{11} + c_{12} + c_{33} - 3K_\pm - 2G_\pm) - 9\gamma_\pm(K_V - K_\pm). \tag{28}$$

Optimum values of the moduli for the comparison materials have been shown to be (in our notation)

$$K_\pm = \frac{K_V(G^r_{eff} - G_\pm)}{(G^v_{eff} - G_\pm)}, \tag{29}$$

where, for K_-,

$$0 \leqslant G_- \leqslant \min(c_{44}, G^{\mathrm{r}}_{\mathrm{eff}}, c_{66}) \tag{30}$$

and where, for K_+

$$\max(c_{44}, G^{\mathrm{v}}_{\mathrm{eff}}, c_{66}) \leqslant G_+ \leqslant \infty. \tag{31}$$

Note that, when $G_- = 0$, $K_- = K_{\mathrm{R}}$, because $K_{\mathrm{R}} = K_{\mathrm{V}} G^{\mathrm{r}}_{\mathrm{eff}}/G^{\mathrm{v}}_{\mathrm{eff}}$ from the product formulas (Berryman, 2004a). When $G_+ \to \infty$, $K_+ \to K_{\mathrm{V}}$.

For the laminated materials considered here, the minimum condition in (30) will never be satisfied by c_{66} except in the trivial case of constant shear modulus. Each of the other two arguments can possibly become the minimum under certain non-trivial circumstances. For the materials considered, the maximum condition in (31) will always be uniquely satisfied by c_{66}, except again for the trivial case of constant shear modulus.

Peselnick and Meister (1965) had originally obtained all these results except for the additional condition in (30) that permits c_{44} to be replaced in some circumstances by $G^{\mathrm{r}}_{\mathrm{eff}}$. This new condition was added later by Watt and Peselnick (1980).

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