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Case study

Bounds and self-consistent estimates of the elastic constants of polycrystals

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ABSTRACT

The Hashin–Shtrikman bounds on the elastic constants have been previously calculated for polycrystalline materials with crystallites having general elastic symmetry (triclinic crystallite symmetry). However, the calculation of tighter bounds and the self-consistent estimates of these elastic constants has remained unsolved. In this paper, a general theoretical expression for the self-consistent elastic constants is formulated. An iterative method is used to solve the expression for the self-consistent estimates. Each iteration of the solution gives the next tighter set of bounds including the well-known Voigt–Reuss and Hashin–Shtrikman bounds. Thus, all of the bounds on the elastic constants and the self-consistent estimates for any crystallite symmetry are obtained in a single, computationally efficient procedure. The bounds and self-consistent elastic constants are reported for several geophysical materials having crystallites of monoclinic and triclinic symmetries.

1. Introduction

Information related to the elastic properties of subsurface materials is obtained by relating seismic signatures to well-controlled laboratory experiments (Jones et al., 2009). Often, the laboratory measurements are conducted on single crystals of material, which have well known properties. Relating the single-crystal measurements to processes involving a larger system, e.g., a polycrystal, containing similar crystallites requires modeling the physical interaction of the crystallites and the resulting influence on the larger system. The most commonly employed models for polycrystals are based on statistically homogenizing the crystallites.

Homogenization of the elastic properties extends back to Voigt (1887) who considered the polycrystal's elastic constants to be equal to the average of the crystallite's elastic constants over all possible crystallite orientations. Inherent to Voigt's approach is the assumption of uniform strain throughout the polycrystal. The assumption of strain uniformity satisfies the kinematic compatibility at grain boundary interfaces, while losing continuity of surface traction or static compatibility. Reuss (1929) considered a uniform stress assumption, which satisfies static compatibility while sacrificing kinematic compatibility. Hill (1952) proved, based on the extreme assumptions of Voigt and Reuss, that the Voigt and Reuss estimates of the polycrystal's elastic constants bound the true elastic constants; i.e., the polycrystal's elastic constants that would result if both kinematic and static compatibility at the grain boundaries are satisfied. Hashin and Shtrikman (1962a, 1962b, 1963) developed new variational principles that allowed the next set of bounds to be determined by seeking values of the bulk and shear modulus that are near to regions of positive and negative definiteness of the first-order deviation between the crystallite's and polycrystal's elastic tensors. Watt et al. (1976) argued that the Hashin–Shtrikman bounds are the tightest bounds achievable without precise knowledge of the shapes, sizes, and correlations of the crystallites. Thus, the Hashin–Shtrikman bounds have been applied extensively in geophysical applications where it is difficult to ascertain such microstructural features (Jones et al., 2009; Xu and White, 1996; Berge et al., 1995; Vanorio et al., 2003). However, bounds tighter than those of Hashin–Shtrikman are possible if the microstructure is well characterized.

Watt and Peselnick (1980) and Watt (1986, 1979, 1980) considered the case of statistically isotropic and homogeneous polycrystals composed of uncorrelated, spherically shaped crystallites. Later, Watt (1987) published a computational procedure for the calculation of the Hashin–Shtrikman bounds for each of these cases. Berryman (2005, 2011) provided a convenient procedure to

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calculate the Hashin–Shtrikman bounds for several cases of crystallite symmetry. More recently, Brown (2015) has provided a calculation scheme that includes the most general crystallite symmetry (triclinic).

Following Brown (2015), a computational (MATLAB) procedure is offered that calculates the bounds on the elastic constants of statistically isotropic polycrystals composed of spherically-shaped crystallites belonging to any of the crystallographic symmetry classes. However, in this paper, all orders of bounds (including those of Voigt–Reuss and Hashin–Shtrikman) are considered. The calculation procedure stems from a theoretical expression of the effective elastic constants that one would find when the kinematic and static compatibility is satisfied throughout the polycrystal. An iterative solution is sought where each iteration gives the next tighter set of bounds. Convergence of the bounds produces estimates of the well-known self-consistent elastic constants. Crystals having additional symmetry can be treated as a limiting case of triclinic symmetry by employing the symmetry relations on the elastic constants (Brugger, 1965). The limiting case of cubic symmetry reproduces the formulas derived by Gairola and Kröner (1981).

2. Theory

This section describes the analytical steps to define and reduce the tensors needed in Eq. (7), which is the effective elastic moduli tensor that is used in the iterative computational procedure. Throughout the theory, the components of the fourth-rank tensors use the Voigt index convention where the pairs of indices obtain the following values: 11 → 1, 22 → 2, 33 → 3, 12 → 6, 13 → 5, and 23 → 4. For example, the (i=3, j=3, k=1, l=2) component of the elastic moduli tensor C_{ijkl} is c_{36}. Additionally, the standard summation convention over repeated indices from 1 to 3 is assumed.

Hooke's law for a linearly elastic polycrystalline medium is

$$\sigma_{ij}^{*} = C_{ijkl}^{*}\epsilon_{kl}^{*}, \tag{1}$$

where ϵ^{*} is the infinitesimal strain tensor and C_{ijkl}^{*} is the effective fourth-rank elastic modulus tensor for the medium. Similarly, Hooke's law for an individual spherical crystallite contained in the polycrystal is

$$\sigma_{ij} = C_{ijkl}\epsilon_{kl}, \tag{2}$$

where C_{ijkl} is the elastic modulus of the crystallite. In general, $\epsilon_{ij}^{*} \neq \epsilon_{ij}$ unless $C_{ijkl}^{*} = C_{ijkl}$. The connection between the strain field of the crystallite and polycrystal is

$$\epsilon_{ij} = H_{ijkl}\epsilon_{kl}^{*}. \tag{3}$$

H_{ijkl} is known as the concentration tensor given as (Lubarda, 2002; Qu and Cherkaoui, 2006)

$$H_{ijkl} = \left(C_{ijmn} + R_{ijmn}\right)^{-1}\left(C_{mnkl}^{*} + R_{mnkl}\right), \tag{4}$$

where

$$R_{ijkl} = C_{ijmn}^{*}\left[E_{mnkl}^{-1} - I_{mnkl}\right], \tag{5}$$

with E_{ijkl} being the Eshelby tensor (Eshelby, 1957) and I_{ijkl} being the identity tensor. The Eshelby tensor for a spherical crystallite embedded in the polycrystal is

$$E_{ijkl} = \frac{3c_{12}^{*} - 2c_{44}^{*}}{15\left(c_{12}^{*} + 2c_{44}^{*}\right)}\delta_{ij}\delta_{kl} + \frac{2\left(3c_{12}^{*} + 8c_{44}^{*}\right)}{15\left(c_{12}^{*} + 2c_{44}^{*}\right)}I_{ijkl}, \tag{6}$$

where c_{12}^{*} and c_{44}^{*} are the effective Lamé and shear constants of the polycrystal, respectively. The employment of this Eshelby tensor is not overly restrictive because only the average grain shape of the medium needs to be spherical. Alternate, more complicated forms of the Eshelby tensor may be consulted for the case of a polycrystal with (on the average) ellipsoidally shaped grains. ϵ_{ij} is dependent on the crystallographic orientation of the crystallite with respect to the principal directions of ϵ_{ij}^{*}. However, the volume average of ϵ_{ij} is equal to ϵ_{ij}^{*}. Substituting into Eq. (3), while letting $\langle\rangle$ denote the volume average ($\langle\epsilon_{ij}\rangle = \epsilon_{ij}^{*}$), yields $\langle H_{ijkl}\rangle = I_{ijkl}$. Applying the orientation average to Eq. (4) and rearranging then leads to expressions for the effective elastic moduli tensor,

$$C_{ijkl}^{*} = \langle\left(C_{ijkl} + R_{ijkl}\right)^{-1}\rangle^{-1} - R_{ijkl}, \tag{7}$$

Evaluation of Eq. (7) is completed using the following steps. Find R_{ijkl} by observing that the tensor E_{ijkl} has isotropic symmetry, which permits a straightforward evaluation of E_{ijkl}^{-1}. E_{ijkl}^{-1} also has isotropic symmetry and can be written in terms of the isotropic basis functions $\delta_{ij}\delta_{kl}$ and I_{ijkl}. Thus, the inner products over the repeated indices m and n reduce to simple inner products between the isotropic basis functions. Then, R_{ijkl} can be written in the isotropic form, $R_{ijkl} = r_{12}\delta_{ij}\delta_{kl} + 2r_{44}I_{ijkl}$, where r_{12} and r_{44} are the only independent components of R_{ijkl}. C_{ijkl} and R_{ijkl} can be written with respect to an alternative coordinate system through the use of orthogonal transformation operators, $C'_{ijkl} = a_{i\alpha}a_{j\beta}a_{k\gamma}a_{l\delta}C_{\alpha\beta\gamma\delta}$ and $R'_{ijkl} = a_{i\alpha}a_{j\beta}a_{k\gamma}a_{l\delta}R_{\alpha\beta\gamma\delta}$. Expanding over the repeated indices, and applying the isotropic symmetry relations to R_{ijkl} allows the sum $C_{ijkl} + R_{ijkl}$ to be constructed in the following manner:

$$\begin{aligned} C_{ijkl} + R_{ijkl} = a_{i\alpha}a_{j\beta}a_{k\gamma}a_{l\delta}\left(C_{\alpha\beta\gamma\delta} + R_{\alpha\beta\gamma\delta}\right) = a_{i1}a_{j1}a_{k1} \\ a_{l1}\left(c_{11} + r_{12} + 2r_{44}\right) \\ + a_{i1}a_{j1}a_{k1}a_{l2}c_{16} + a_{i1}a_{j1}a_{k1}a_{l3}c_{15} + a_{i1}a_{j1}a_{k2}a_{l1}c_{16} \\ + a_{i1}a_{j1}a_{k2}a_{l2}\left(c_{12} + r_{12}\right) + \cdots \end{aligned} \tag{8}$$

$C_{ijkl} + R_{ijkl}$ in Eq. (8) has triclinic symmetry like C_{ijkl}. Hence, the inverse of $C_{ijkl} + R_{ijkl}$ follows the same procedure as constructing the inverse of C_{ijkl}. The inverse $L_{ijkl} = \left(C_{ijkl} + R_{ijkl}\right)^{-1}$ is determined by solving the overdetermined system of 81 equations for the 21 unknown components of L_{ijkl} generated by

$$\left(C_{ijmn} + R_{ijmn}\right)L_{mnkl} = I_{ijkl}. \tag{9}$$

For example, the first equation follows from considering $i = j = k = l = 1$ and summing over the repeated indices m and n. Repeating this process for all combinations of i, j, k, l leads to the 81 equations. The average $\langle L_{ijkl}\rangle = \langle\left(C_{ijmn} + R_{ijmn}\right)^{-1}\rangle$ is obtained by equating the invariants of L_{ijkl} and $\langle L_{ijkl}\rangle$ and solving for the two independent components of $\langle L_{ijkl}\rangle$, which are

$$15\ell_{12}^{*} = \ell_{11} + \ell_{22} + \ell_{33} + 4\left(\ell_{12} + \ell_{13} + \ell_{23}\right) - 2\left(\ell_{44} + \ell_{55} + \ell_{66}\right), \tag{10a}$$

$$15\ell_{44}^{*} = \ell_{11} + \ell_{22} + \ell_{33} - \left(\ell_{12} + \ell_{13} + \ell_{23}\right) + 3\left(\ell_{44} + \ell_{55} + \ell_{66}\right). \tag{10b}$$

The evaluation of $\langle L_{ijkl}\rangle^{-1} = \langle\left(C_{ijmn} + R_{ijmn}\right)^{-1}\rangle^{-1}$ is straightforward because $\langle L_{ijkl}\rangle$ has isotropic symmetry. At this point, all of the necessary tensors and operations contained in Eq. (7) are defined. Solving Eq. (7) for C_{ijkl}^{*} in closed form is not possible, except for the special case of cubic crystallographic symmetry, because it is transcendental with C_{ijkl}^{*} appearing in the definitions of R_{ijkl} and E_{ijkl} on the righthand side of Eq. (7). For polycrystals having crystallites of cubic symmetry, the bulk modulus of the polycrystal is equal to the bulk modulus of the crystallites, which leads to the expression $c_{12}^{*} = \left(c_{11} + 2c_{12} - 2c_{44}^{*}\right)/3$ and allows c_{12}^{*} and c_{44}^{*} to be

determined exactly. For this special case, Gairola and Kröner (1981) cast the shear modulus c^*_{44} in two different forms. The first form is a cubic equation in terms of the variable c^*_{44}, which can be solved exactly. The second form is a scalar form of Eq. (7) obtained by applying the cubic symmetry conditions to C_{ijkl} and observing the equivalent bulk moduli. Gairola and Kröner (1981) observed that an iterative solution can be constructed where each updated solution is the next order of bounds and the self-consistent estimate of c^*_{44} is found from the converged solution.

Here, we follow a similar approach where an iterative solution to Eq. (7) is sought to determine C^*_{ijkl}. Apart from Gairola and Kröner (1981), the iterative solution satisfies the general tensorial form of Eq. (7) and is valid for all crystallite symmetries. To begin this procedure, the righthand side receives appropriate zeroth-order estimates of c^*_{12} and c^*_{44}. For example, letting $c^*_{12}=0$ and $c^*_{44}=\infty$ on the righthand side of Eq. (7) produces the first-order Voigt estimates on the lefthand side. Repeating this process by inputting the Voigt estimates on the righthand side of Eq. (7) delivers the third-order estimates on the lefthand side. Subsequent iterations of this process result in all odd-order bounds to the elastic constants. The increasing order of the bounds become tighter after multiple iterations, which eventually converges to the self-consistent estimates of c^*_{12} and c^*_{44}.

The case when $c^*_{12}=0$ and $c^*_{44}=\infty$ produces the odd-order upper bounds to the bulk modulus κ^* and shear modulus c^*_{44}. The odd-order lower bounds are obtained when the procedure starts with $c^*_{12}=\infty$ and $c^*_{44}=0$. Even-order upper bounds are obtained by starting the procedure with $c^*_{12}=\min\left[a_{1i}a_{1j}a_{2k}a_{2l}C_{ijkl}\right]$ and $c^*_{44}=\max\left[a_{2i}a_{3j}a_{2k}a_{3l}C_{ijkl}\right]$, while the even-order lower bounds are obtained when the procedure starts with $c^*_{12}=\max\left[a_{1i}a_{1j}a_{2k}a_{2l}C_{ijkl}\right]$ and $c^*_{44}=\min\left[a_{2i}a_{3j}a_{2k}a_{3l}C_{ijkl}\right]$. The second-order bounds are the well-known Hashin and Shtrikman bounds (Hashin and Shtrikman, 1962a, 1962b). For each of the four initial solutions, the same self-consistent estimates emerge after a suitable number of iterations.

3. Implementation

The MATLAB *m*-file, *bounds.m*, operates as a function to be executed from the MATLAB command window. Contained within the *m-file* is the primary function to be called from the command window

```
function ret=bounds(sel,N,c11,c12,c13,...,c55,
  c56,c66)
```

and the additional function

```
function [s11,s12,s13,...,s55,s56,s66]
  =invtens(c11,c12,c13,...,c55,c56,c66).
```

21 elastic constants are required as inputs into `bounds` along with the inputs of `sel` and `N`. `sel` receives a value of 1, 2, 3, or 4 corresponding to the desired even-order upper, even-order lower, odd-order upper, or odd-order lower bounds, respectively. The designation of upper/lower are the bounds on the bulk and shear modulus. The input `N` is an integer greater than 0 that specifies the number of iterations to be used to solve Eq. (7). The function `invtens` obtains the components of the inverse of a tensor with the 21 components `c11,c12,c13,...,c55,c56,c66`. This function can be used to obtain the closed-form elastic stiffness/compliance relations for general triclinic symmetry.

Inside the function `bounds` is the iterative solution to Eq. (7). The following provides an overview of the case when the odd-order lower bounds is selected (`sel=4`). The iteration procedure starts by letting $c^*_{12}\approx\infty$ and $c^*_{44}=0$,

```
C12(1)=10E6; C44(1)=0;.
```

These values are placed into the righthand side of Eq. (7). Eq. (7) is evaluated iteratively by continually updating the righthand side with the output on the lefthand side. Programmatically this procedure is contained within the for-loop

```
for j=1:N.
```

The various tensor components of E_{ijkl}, E^{-1}_{ijkl}, R_{ijkl}, $C_{ijkl}+R_{ijkl}$, $L_{ijkl}=\left(C_{ijkl}+R_{ijkl}\right)^{-1}$, $\langle L_{ijkl}\rangle$, and $\langle L_{ijkl}\rangle^{-1}$ are calculated within the for-loop as follows.

E_{ijkl}:

```
e12=(3*C12(j)-2*C44(j))/(15*(C12(j)+2*C44
  (j)));
e44=(3*C12(j)+8*C44(j))/(15*(C12(j)+2*C44
  (j)));
```

E^{-1}_{ijkl}:

```
eI12=-e12/(2*e44*(3*e12+2*e44));
eI44=1/(4*e44);
```

R_{ijkl}:

```
r12=C12(j)*(3*eI12+2*eI44-1)+2*C44(j)*eI12;
r44=C44(j)*(2*eI44-1);
```

$L_{ijkl}=\left(C_{ijkl}+R_{ijkl}\right)^{-1}$:

```
[l11,l12,l13,...,l55,l56,l66]
  =invtens(c11+r12+2*r44,c12+r12,...,c56,
   c66+r44);
```

$\langle L_{ijkl}\rangle=\langle\left(C_{ijkl}+R_{ijkl}\right)^{-1}\rangle$:

```
L12=(l11+l22+l33+4*(l12+l13+l23)-2*
  (l44+l55+l66))/15;
L44=(l11+l22+l33-(l12+l13+l23)+3*
  (l44+l55+l66))/15;
```

$\langle L_{ijkl}\rangle^{-1}=\langle\left(C_{ijkl}+R_{ijkl}\right)^{-1}\rangle^{-1}$:

```
LI12=-L12/(2*L44*(3*L12+2*L44));
LI44=1/(4*L44);
```

The components of C^*_{ijkl} on the lefthand side of Eq. (7) are updated:

```
C12(j+1)=LI12-r12; C44(j+1)=LI44-r44;
```

After N iterations, the values of the shear and bulk modulus are returned:

```
ret=[C44; C12+2*C44/3];.
```

4. Results

The *m*-file, *bounds.m*, was created using analytical and closed-form simplifications of several of the needed tensors. The analytical nature of the function permits the symbolic evaluation for the special case of cubic crystallographic symmetry. Evaluating the function symbolically with the 21 elastic constants for cubic symmetry (c_{11}, c_{12}, $c_{13}=c_{12}$, $c_{14}=0$, $c_{15}=0$, $c_{16}=0$, $c_{22}=c_{11}$, $c_{23}=c_{12}$, $c_{24}=0$, $c_{25}=0$, $c_{26}=0$, $c_{33}=c_{11}$, $c_{34}=0$, $c_{35}=0$, $c_{36}=0$, c_{44}, $c_{45}=0$, $c_{46}=0$, $c_{55}=c_{44}$, $c_{56}=0$, $c_{66}=c_{44}$) reproduces exactly the closed-form expressions of the self-consistent shear modulus found in Gairola and Kröner (1981).

Self-consistent estimates of lower symmetry were checked against the results of Berryman (2005, 2011). These estimates

Table 1
Voigt–Reuss, Hashin–Shtrikman bounds and self consistent estimates of the shear modulus μ and bulk modulus κ (in units of GPa). Materials with crystallites of monoclinic symmetry are listed above the first dashed line while materials with crystallites of triclinic symmetry are below the first dashed line. Hexagonal graphite is included below the second dashed line as a high anisotropy example. The input elastic constants were taken from (a) the Landolt–Börnstein tables (Every and McCurdy, 1992), (b) Brown (2015), and (c) Berryman (2005).

Material	μ^R	μ^{-HS}	μ^{SC}	μ^{+HS}	μ^V	κ^R	κ^{-HS}	κ^{SC}	κ^{+HS}	κ^V	A^U
[a]Dibenzyl	1.9	2.1	2.2	2.3	2.5	5.0	5.4	5.4	5.5	5.8	1.7
[a]Ethylene diamine tartrate	6.2	7.0	7.1	7.7	9.1	15.9	19.2	19.5	21.3	24.5	2.8
[a]Oxalic acid dihydrate	4.2	4.9	5.4	5.8	6.9	10.8	11.9	12.4	13.0	14.3	3.6
[a]Potassium cobalt cyanide	8.8	9.5	9.7	9.9	10.7	11.1	11.8	11.9	12.0	12.6	1.2
[a]Tin difluoride	10.3	11.6	12.1	12.4	13.8	16.5	17.1	17.3	17.5	17.9	1.8
[a]Ammonium tetroxalate dihydrate	5.4	6.5	6.7	7.2	8.4	12.1	15.7	16.2	17.4	20.2	3.5
[a]Cesium trihydrogen selenite	12.6	13.2	13.2	13.3	13.8	29.9	31.8	31.9	32.2	33.6	0.6
[a]Copper sulfate pentahydrate	10.5	11.2	11.4	11.5	12.1	36.1	37	37.2	37.4	38.1	0.8
[a]Lithium hydrogen oxalate monohydrate	11.5	14.0	15.0	16.9	20.1	22.1	27.5	29.7	33.2	39.3	4.5
[a]Potassium tetroxalate dihydrate	5.8	6.9	7.2	7.8	9.0	13.3	16.4	17.0	18.0	19.9	3.3
[a]Sodium hydrogen oxalate hydrate	9.6	11.1	11.9	13	15.1	22.3	25.1	26.6	28.4	31.8	3.3
[b]An_0	29.8	33.6	34.5	36.7	41.4	54.1	57.8	58.6	60.3	63.1	2.1
[b]An_{25}	31.1	33.9	34.8	36.1	39.5	64.3	66.2	66.7	67.5	69.2	1.4
[b]An_{37}	33.6	36.5	37.3	38.7	42.3	68.8	70.5	70.9	71.6	73.0	1.4
[b]An_{48}	33.9	36.8	37.7	39.1	42.9	74.1	75.4	75.8	76.4	77.6	1.4
[b]An_{60}	33.9	36.6	37.3	38.3	41.2	73.9	75.3	75.6	76.0	77.0	1.1
[b]An_{78}	34.3	36.7	37.3	38.2	41.1	78.3	80.2	80.5	81.0	82.3	1.0
[b]An_{96}	35.7	38.2	38.9	39.7	42.5	84.1	86.2	86.7	87.2	88.7	1.0
[c]Graphite	9.2	15.4	52.6	120.2	219.4	35.8	42.6	88.0	168.9	286.3	121.1

found agreement to within 1 GPa for the materials considered. Additionally, the implementation was checked against the results of Brown (2015) for plagioclase feldspars having crystallites of triclinic symmetry. The bottom of Table 1 gives these results. The different feldspars are categorized as a function of varying anorthite content denoted by An_x with x being the percentage of anorthite. The Voigt and Reuss estimates agree exactly with Brown (2015) while the Hashin–Shtrikman estimates deviate at most by 0.7 GPa for both the shear and bulk modulus.

The self-consistent estimates of the bulk and shear modulus for polycrystals having monoclinic and triclinic symmetry are seen in Table 1. These estimates were obtained from *bounds.m* using 10 iterations. As expected, the self-consistent estimates are always contained within the Hashin–Shtrikman bounds. Convergence to the self-consistent estimates was obtained in less than 0.1 s for each material. Results for graphite are included in Table 1 to demonstrate the effect of exceptionally high crystallite anisotropy. The anisotropy is quantified using the universal anisotropy index A^U of Ranganathan and Ostoja-Starzewski (2008) given as

$$A^U = \frac{\kappa^V}{\kappa^R} + 5\frac{\mu^V}{\mu^R} - 6. \tag{11}$$

The universal anisotropy index is 0 when the crystallite is isotropic, i.e., the Voigt and Reuss bounds coincide. The vast majority of inorganic crystals have anisotropy in the range $0 \ll A^U \ll 5$ (Ranganathan and Ostoja-Starzewski, 2008). This includes all of the materials in Table 1 except for graphite, which has $A^U = 121.1$. Convergence to the self-consistent estimates for graphite require additional iterations. Effectively, the iterative procedure needs to overcome the crystallite anisotropy to reach a homogenized representation of the polycrystal. Conversely, the polycrystal is *a priori* homogenized for the case when the crystallites are isotropic. Polycrystals with crystallites having strong elastic anisotropy represents situations where the self-consistent estimate differs considerably from the Hill estimate, which is the arithmetic average of the Voigt–Reuss bounds. For the case of graphite, the Hill estimated shear and bulk modulus differs from the self-consistent values by 117.4% and 83%, respectively. The self-consistent values for graphite agree with those reported by Berryman (2005). However, the Hashin–Shtrikman bounds for graphite calculated using the present model differ from Berryman (2005) significantly.

Differences between the bounds reported here and those from Berryman (2005, 2011) and Brown (2015) are caused by different model assumptions and are amplified for crystallites with extremely large anisotropy. However, the converged self-consistent values agree with Berryman (2005) even for highly anisotropic crystallites. The present model is a special case of the bound theorems of Kröner (1986). The bounds generated using the iterative solution represent the bounds on the elastic properties of

a polycrystal with statistical properties denoted as *perfectly disordered* (Kröner, 1986). The self-consistent solution is the rigorous solution for the perfectly disordered polycrystal that achieves global and local continuity of the stress and strain fields. The primary requirement of perfect disorder is the vanishing of statistical correlations amongst crystallites. Thus, the bounds obtained using the present model are ascribed to this special statistical classification of the microstructure. Details of statistical disorder and a discussion of the application of such models to real polycrystals can be located in Kröner (1986).

The supplementary files *bounds.m* and *analysis.m* are included. *analysis.m* can be used to reproduce the entries in Table 1.

5. Conclusions

This article provides an iterative procedure to obtain the elastic constants of statistically isotropic polycrystalline materials. Each iteration corresponds to a set of tighter bounds including the well-known Voigt–Reuss (Voigt, 1887; Reuss, 1929) and Hashin–Shtrikman (Hashin and Shtrikman, 1962a, 1962b, 1963) bounds. Convergence of the iterative procedure produces the self-consistent estimates. The derivation to arrive at the iterative expression was based on a number of analytical simplifications that did not limit the procedure to specific symmetry classes. Thus, the bounds and self-consistent estimates can be calculated for any crystallite material provided the single-crystal elastic constants are known. Output results agree closely with the previously published Hashin–Shtrikman bounds and self-consistent values (Berryman, 2005, 2011; Brown, 2015). Convergence to the self-consistent values for polycrystals with crystallites of triclinic crystallite symmetry was achieved in under 0.1 s.

Appendix A. Supplementary data

Supplementary data associated with this paper can be found in the online version at http://dx.doi.org/10.1016/j.cageo.2016.07.008.

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