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ON THE ESTIMATES FOR THE ELASTIC MODULI OF RANDOM VORONOI TRICLINIC POLYCRYSTALS

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Abstract. Our major new results and the previous ones on the bounds for elastic ramdom polycrystals, and most advanced 3D finite elemeni results for random 3D Voronoi polycrystals are resumed and analysed (together for the first time). Recently obtained numerical Dirichlet and Neumann simulation results for the effective elastic moduli of a large 10000-grain-size random Voronoi polycrystal representative volume element (RVE) for a number of triclinic and monoclinic base crystals (Mursheda and Ranganathan, 2017) are compared critically with the bounds on the moduli. Though major parts within the simulation results fall within the bounds of Pham (2011), some Dirichlet upper estimates still lie outside the bounds. Many more RVEs are needed to represent the Voronoi polycrystal on the same RVE-size-level, and larger RVEs are needed for checking the convergence and comparisons with the bounds.

Keywords: effective elastic moduli, random Voronoi polycrystal, triclinic crystal, scatter measures of the estimates.

1. INTRODUCTION

Usual polycrystalline materials on the microscopic scale are composed from crystals (grains) of irregular shapes and random crystalline and shape orientations (without preferable relative directions) that they appear macroscopically isotropic and have such definite macroscopic (effective) properties such that can be tabulated for engineering applications (but often with only very few significant digits). Still, because of the microstructural irregularity, the exact macroscopic elastic moduli of the aggregates can hardly be found, and may not be uniquely determinable. Hence the evaluation of the possible scatter ranges for the macroscopic moduli of the random polycrystals should have the methodological and practical values. From the minimum energy or specialized variational principles, various upper and lower bounds on the effective moduli of random polycrystals have been constructed and calculated for particular crystalline aggregates [1–14].

Voronoi random polycrystal model is widely recognized as the best model representing practical random polycrystalline materials, and methods have been developed

to generate a Voronoi random polycrystal representative volume element (RVE), and to solve the elastic homogenization problem on it [15–18]. Strong mathematical methods and computer resources are required to generate and solve the problem on sufficiently large Voronoi random polycrystal RVEs to get the results of sufficient accuracy. Recently Mursheda and Ranganathan [18] have solved the problem on a large 10000-grain size Voronoi random polycrystal RVE, and their solutions of the respective Dirichlet and Neumann problems for a number of monoclinic and triclinic polycrystals have reached a bench mark result to fall entirely within Hashin-Shtrikman bounds with significant margins. In this paper their numerical results are compared with tighter bounds, with critical discussions and recommendations for further studies.

2. ESTIMATES

The general Voigt-Reuss-Hill bounds on the fourth-rank macroscopic (effective) elastic tensor $\mathbf{C}^{eff}$ of a polycrystalline aggregate derived from the minimum energy principles can be given in the form [10]

$$\langle \mathbf{C}^{-1} \rangle^{-1} \leq \mathbf{C}^{eff} \leq \langle \mathbf{C} \rangle, \tag{1}$$

where $\langle \mathbf{C} \rangle$ designates the volume average of the crystal fourth-rank elastic tensor $\mathbf{C}$ over all space crystalline orientations of the grains within a representative volume element (RVE). As the crystalline orientations of the constituent grains are distributed uniformly over all directions in a random polycrystalline aggregate, the effective elastic tensor of the random aggregate should be isotropic $\mathbf{C}^{eff} = \mathbf{T}(K^{eff}, \mu^{eff})$, where $\mathbf{T}(K, \mu)$ is the isotropic fourth-rank tensor function with the components

$$T_{ijkl}(K, \mu) = K\delta_{ij}\delta_{kl} + \mu(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}), \tag{2}$$

while

$$\begin{aligned} &\langle \mathbf{C} \rangle = \mathbf{T}(K_V, \mu_V), \quad K_V = \frac{1}{9}C_{iijj}\,, \\ &\mu_V = \frac{1}{10}C_{ijij} - \frac{1}{30}C_{iijj}\,, \\ &\langle \mathbf{C}^{-1} \rangle^{-1} = \mathbf{T}(K_R, \mu_R), \quad K_R = [(\mathbf{C}^{-1})_{iijj}]^{-1}, \\ &\mu_R = [\frac{2}{5}(\mathbf{C}^{-1})_{ijij} - \frac{2}{15}(\mathbf{C}^{-1})_{iijj}]^{-1}. \end{aligned} \tag{3}$$

For random polycrystals, where the shape and crystalline orientations of the grains are uncorrelated, tighter bounds have been constructed. The Hashin–Shtrikman bounds can be presented in the form

$$\mathbf{P}_C(\mathbf{C}^-) \leq \mathbf{C}^{eff} \leq \mathbf{P}_C(\mathbf{C}^+), \tag{4}$$

where

$$\begin{aligned} &\mathbf{P}_C(\mathbf{C}^0) = \langle (\mathbf{C}+\mathbf{C}^*)^{-1}\rangle^{-1} - \mathbf{C}^* \,, \\ &\mathbf{C}^0 = \mathbf{T}(K_0,\mu_0)\,, \quad \mathbf{C}^* = \mathbf{T}(K_*,\mu_*)\,, \\ &K_* = \frac{4}{3}\mu_0\,, \quad \mu_* = \mu_0\frac{9K_0+8\mu_0}{6K_0+12\mu_0}\,, \end{aligned} \tag{5}$$

and $\mathbf{C}^0 = \mathbf{C}^+$ is chosen to minimize the upper bound in (4) under the restriction $\mathbf{C} - \mathbf{C}^0 \le \mathbf{0}$ [i.e. $\boldsymbol{\varepsilon} : (\mathbf{C}-\mathbf{C}^0) : \boldsymbol{\varepsilon} \le 0$, for all second order symmetric strain tensor $\boldsymbol{\varepsilon}$, and for all orientations of the elastic tensor $\mathbf{C}$)]; while $\mathbf{C}^0 = \mathbf{C}^-$ is chosen to maximize the lower bound in (4) under the restriction $\mathbf{C}^{-1} - (\mathbf{C}^0)^{-1} \le \mathbf{0}$. Numerical calculations of Hashin–Shtrikman bounds are rather involved, especially for the low symmetry crystal aggregates [12].

The bounds of Pham [10] also have the formal form (4)–(5), where $\mathbf{C}^0 = \mathbf{C}^+$ is chosen to minimize the upper bound in (4) under certain restrictions, including $\mathbf{C}^0 \ge \langle \mathbf{C}\rangle$; while $\mathbf{C}^0 = \mathbf{C}^-$ is chosen to maximize the lower bound in (4) under certain restrictions, including $\mathbf{C}^0 \le \langle \mathbf{C}^{-1}\rangle^{-1}$. Numerical calculations of the bounds are also rather involved. However the bounds and the slightly tighter ones [11, 14] can be well approximated by the (tighter) simple bounds for the specific and idealistic spherical cell polycrystals

$$\mathbf{P}_C(\langle \mathbf{C}^{-1}\rangle^{-1}) \le \mathbf{C}^{eff} \le \mathbf{P}_C(\langle \mathbf{C}\rangle). \tag{6}$$

Voigt–Reuss–Hill bounds can also formally obtained from (4)–(5) when $\mathbf{C}^+$ approaches ∞ (K_0, μ_0, K_*, μ_* approach ∞), and $\mathbf{C}^-$ approaches $\mathbf{0}$ (K_0, μ_0, K_*, μ_* approach 0).

The self-consistent approximation for the effective elastic moduli of the random polycrystal is the solution $\mathbf{C}^0 = \mathbf{C}^{SC}$ of the self-consistent equation

$$\mathbf{C}^0 = \mathbf{P}_C(\mathbf{C}^0). \tag{7}$$

There are 21 independent elastic constants for a (triclinic) crystal of general anisotropy, which in the convenient Voigt two-index notations are given as $C_{11}, C_{12}, C_{13}, C_{14}, C_{15}, C_{16}, C_{22}, C_{23}, C_{24}, C_{25}, C_{26}, C_{33}, C_{34}, C_{35}, C_{36}, C_{44}, C_{45}, C_{46}, C_{55}, C_{56}, C_{66}$.

In the case of monoclinic symmetry, the number of independent elastic constants reduces to 13, with $C_{14} = C_{15} = C_{24} = C_{25} = C_{34} = C_{35} = C_{46} = C_{56} = 0$, and $\{x_1, x_2\}$ being the plane of symmetry.

From the stiffness 6×6-matrix $\{C_{\alpha\beta}\}_1^6$ we construct the scaled inverse compliant matrix $\{S_{\gamma\delta}\}_1^6$ through the operation $F_{\gamma\delta}$ as followed

$$\begin{aligned} &S_{\gamma\delta} = \mathcal{F}_{\gamma\delta}(\{C_{\alpha\beta}\}_1^6) = h\bar{\mathcal{F}}_{\gamma\delta}(\{C_{\alpha\beta}\}_1^6)\,, \quad \gamma,\delta = 1,\dots,6\,, \\ &h = \begin{cases} \dfrac{1}{2} & \text{if } \gamma \text{ or } \delta = 4,5,6 \\ \dfrac{1}{4} & \text{if } \gamma \text{ and } \delta = 4,5,6 \\ 1 & \text{otherwise} \end{cases} \end{aligned} \tag{8}$$

where $\{\bar{\mathcal{F}}_{\gamma\delta}\}_1^6$ is the inverse matrix of $\{C_{\alpha\beta}\}_1^6$.

Then, the Voigt-Reuss-Hill bounds (1)–(3) have particular expressions, in Voigt notations, for the triclinic crystal aggregate

$$K_R \le K^{eff} \le K_V \,, \quad \mu_R \le \mu^{eff} \le \mu_V \,, \tag{9}$$

where

$$\begin{aligned} K_V &= \frac{1}{9}(C_{11}+C_{22}+C_{33}+2C_{12}+2C_{13}+2C_{23}) \,, \\ \mu_V &= \frac{1}{15}(C_{11}+C_{22}+C_{33}-C_{12}-C_{13}-C_{23}+3C_{44}+3C_{55}+3C_{66}) \,, \end{aligned} \tag{10}$$

$$K_R = \mathcal{K}(\{C_{\alpha\beta}\}_1^6), \quad \mu_R = \mathcal{M}(\{C_{\alpha\beta}\}_1^6) \,, \tag{11}$$

$$\begin{aligned} &\mathcal{K}(\{C_{\alpha\beta}\}_1^6) = (S_{11}+S_{22}+S_{33}+2S_{12}+2S_{13}+2S_{23})^{-1}, \\ &\mathcal{M}(\{C_{\alpha\beta}\}_1^6) = \frac{15}{4}(S_{11}+S_{22}+S_{33}-S_{12}-S_{13}-S_{23}+3S_{44}+3S_{55}+3S_{66})^{-1}, \\ &S_{\gamma\delta} = \mathcal{F}_{\gamma\delta}(\{C_{\alpha\beta}\}_1^6), \;\; \gamma,\delta = 1,\dots,6 \,, \end{aligned} \tag{12}$$

and $\mathcal{F}_{\gamma\delta}$ has been defined in (8).

The tensor function $\mathbf{P}_C(\mathbf{C}^0)$ from (5) would have the expanded expression

$$\begin{aligned} \mathbf{P}_C(\mathbf{C}^0) &= \mathbf{T}\Big(P_K(\mathbf{C},K_0,\mu_0), P_\mu(\mathbf{C},K_0,\mu_0)\Big), \\ P_K(\mathbf{C},K_0,\mu_0) &= [(\mathbf{C}+\mathbf{C}^*)^{-1}_{iijj}]^{-1} - K_* \\ &= \mathcal{K}(\{C^{+*}_{\alpha\beta}\}_1^6) - K_* \,, \\ P_\mu(\mathbf{C},K_0,\mu_0) &= [\frac{2}{5}(\mathbf{C}+\mathbf{C}^*)^{-1}_{ijij} - \frac{2}{15}(\mathbf{C}+\mathbf{C}^*)^{-1}_{iijj}]^{-1} - \mu_* \\ &= \mathcal{M}(\{C^{+*}_{\alpha\beta}\}_1^6) - \mu_* \,, \end{aligned} \tag{13}$$

where

$$\begin{aligned} &C^{+*}_{\alpha\alpha} = C_{\alpha\alpha} + K_* + \frac{4}{3}\mu_* \,, \;\; \alpha = 1,2,3; \\ &C^{+*}_{\alpha\beta} = C_{\alpha\beta} + K_* - \frac{2}{3}\mu_* \,, \;\; \alpha,\beta = 1,2,3, \;\; \alpha \ne \beta; \\ &C^{+*}_{\alpha\alpha} = C_{\alpha\alpha} + \mu_* \,, \;\; \alpha = 4,5,6; \\ &\text{other } C^{+*}_{\alpha\beta} = C_{\alpha\beta}, \end{aligned} \tag{14}$$

and functions $\mathcal{K}, \mathcal{M}$ are defined in (12).

The bounds (6) for the subclass of spherical cell polycrystals, which approximate practical equiaxial particulate aggregates, are especially simple

$$K_s^l \le K^{eff} \le K_s^u \,, \quad \mu_s^l \le \mu^{eff} \le \mu_s^u, \tag{15}$$

where

$$\begin{aligned} K_s^l &= P_K(\mathbf{C}, K_R, \mu_R), \\ K_s^u &= P_K(\mathbf{C}, K_V, \mu_V), \\ \mu_s^l &= P_\mu(\mathbf{C}, K_R, \mu_R), \\ \mu_s^u &= P_\mu(\mathbf{C}, K_V, \mu_V). \end{aligned} \tag{16}$$

The self-consistent approximation for the effective moduli of the random polycrystals are the solution $K_0 = K_{SC}, \mu_0 = \mu_{SC}$ of the systems of self-consistent equations

$$\begin{aligned} K_0 &= P_K(\mathbf{C}, K_0, \mu_0), \\ \mu_0 &= P_\mu(\mathbf{C}, K_0, \mu_0). \end{aligned} \tag{17}$$

3. COMPARISONS

A large random Voronoi polycrystalline aggregate (RVE) with up to 10000 grains has been generated in [18]. Having solved numerically the elasticity problem on the RVE and used the Dirichlet and Neumann boundary conditions, the authors obtained, respectively, the upper and lower estimates for the elastic moduli of that RVE, in comparisons with Voigt–Reuss–Hill and Hashin–Shtrikman bounds, and the self-consistent approximations taken from [12, 13] for certain triclinic and monoclinic polycrystals from [13, 19].

Those numerical upper Dirichlet and lower Neumann estimates (K_D, μ_D and K_N, μ_N) of [18], Hashin–Shtrikman bounds (K_{HS}^u, μ_{HS}^u and K_{HS}^l, μ_{HS}^l) of [12,13], the bounds (K^u, μ^u and K^l, μ^l) according to [10], the bounds for spherical cell polycrystals (K_s^u, μ_s^u and K_s^l, μ_s^l) from (15)–(16), Voigt–Reuss–Hill bounds (K_V, μ_V and K_R, μ_R) from (8)–(12), and the self-consistent approximations (K_{SC}, μ_{SC}) from (17) are compared in Tabs. 1, 2. Though formally can be represented through (4), (5), HS bounds are rather complicated, especially for monoclinic and triclinic polycrystals and need special techniques for calculations [12, 13].

Table 1. Dirichlet and Neumann estimates (μ_D and μ_N) of [18], Hashin-Shtrikman bounds (μ_{HS}^u, μ_{HS}^l) of [12, 13], the bounds (μ^u, μ^l) according to [10], the bounds (μ_s^u, μ_s^l) from (15)–(16), Voigt–Ress–Hill bounds (μ_V, μ_R) from (8)–(12), the self-consistent approximations (μ_{SC}) from (17) for the effective shear moduli of triclinic (t) and monoclinic (m) polycrystals (in GPa)

Crystal	μ_R	μ_{HS}^l	μ^l	μ_s^l	μ_N	μ_{SC}	μ_D	μ_s^u	μ^u	μ_{HS}^u	μ_V
An_{96}(t)	35.70	38.2	38.70	38.73	38.95	38.85	39.33	38.97	39.03	39.7	42.45
T...(m)	10.34	11.6	11.89	12.01	12.12	12.13	12.39	12.22	12.22	12.4	13.79
E...(m)	6.21	7.0	7.01	7.01	7.13	7.09	7.31	7.23	7.45	7.7	9.08
O...(m)	4.18	4.9	5.19	5.25	5.44	5.39	5.59	5.53	5.60	5.8	6.91
L...(t)	11.53	14.0	14.48	14.51	15.17	15.03	15.73	15.60	15.86	16.9	20.13

T...(m): Tin difluoride (m)
E...(m): Ethylene diamine tartrate (m)
O...(m): Oxalic acid dihydrate (m)
L...(t): Lithium hydrogen oxalate monohydrate (t)

Table 2. Dirichlet and Neumann estimates (K_D and K_N) of [18], Hashin-Shtrikman bounds (K^u_{HS}, K^l_{HS}) of [12,13], the bounds (K^u, K^l) according to [10], the bounds (K^u_s, K^l_s) from (15)–(16), Voigt–Ress–Hill bounds (K_V, K_R) from (8)–(12), the self-consistent approximations (K_{SC}) from (17) for the effective bulk moduli of some triclinic (t) and monoclinic (m) polycrystals (in GPa)

Crystal	K_R	K^l_{HS}	K^l	K^l_s	K_N	K_{SC}	K_D	K^u_s	K^u	K^u_{HS}	K_V
An_{96}(t)	84.10	86.2	86.59	86.60	86.71	86.68	87.01	86.77	86.79	87.2	88.74
T...(m)	16.47	17.1	17.27	17.30	17.36	17.35	17.44	17.38	17.38	17.5	17.88
E...(m)	15.95	19.2	19.23	19.26	19.63	19.52	20.27	19.98	20.69	21.3	24.46
O...(m)	10.76	11.9	12.12	12.24	12.46	12.42	12.70	12.60	12.60	13.0	14.33
L...(t)	22.14	27.5	27.83	28.59	29.83	29.66	31.11	30.84	30.84	33.2	39.32

Note that the numerical self-consistent approximations (K_{SC}, μ_{SC}) reported here have the accuracy up to 4 significant digits and are more appropriate for comparison with the respective up-to-4-significant-digit numerical upper Dirichlet and lower Neumann estimates (K_D, μ_D and K_N, μ_N) of [18] than the up-to-3-significant-digit accuracy self-consistent values of [13] taken for comparisons in [18].

The self-consistent approximations (K_{SC}, μ_{SC}) fall in the middle of all bounds. Though most of the self-consistent values lie slightly outside (smaller) the numerical lower estimate K_N, μ_N for the particular random Voronoi RVE of [18], except the case $\mu_{SC} > \mu_N$ for Tin difluoride, it does not mean that it should be always the case with other RVEs of the same 10000-grain-size.

All the lower estimates K_N, μ_N and about a half of the upper estimates K_D, μ_D fall within the bounds K^u, μ^u and K^l, μ^l of [10], but a half of the upper estimates K_D, μ_D are slightly larger than the upper bounds K^u, μ^u. Clearly larger random Voronoi RVEs are needed for comparisons with the bounds.

The scatter range measures (in %) for the bounds $S^K_{VR} = \dfrac{K_V - K_R}{K_V + K_R}$, $S^\mu_{VR} = \dfrac{\mu_V - \mu_R}{\mu_V + \mu_R}$, $S^K_{HS} = \dfrac{K^u_{HS} - K^l_{HS}}{K^u_{HS} + K^l_{HS}}$, $S^K = \dfrac{K^u - K^l}{K^u + K^l}$, $S^K_s = \dfrac{K^u_s - K^l_s}{K^u_s + K^l_s}$, ..., and for the estimates $S^K_{DN} = \dfrac{K_D - K_N}{K_D + K_N}$, $S^\mu_{DN} = \dfrac{\mu_D - \mu_N}{\mu_D + \mu_N}$, and the crystal anisotropy index A^U of [20] are compared in Tab. 3. We can see that S^K, S^K_s, S^K_{DN} are comparable, while S^K_{DN} appear predominantly smaller; S^K, S^K_s, S^K_{DN} are considerably smaller than S^K_{HS}, and much smaller than S^K_{VR} (the same observations for the measures on μ).

Though the generation of a large random Voronoi elastic polycrystal RVE with 10000 grains, and the subsequent numerical solution of the elasticity problems on the RVE with Dirichlet and Neumann boundary conditions in [18] are good steps, we need a number of different generations of large random Voronoi elastic polycrystal RVEs with 10000 grains to get more reliable information. The upper envelopes K^u_D, μ^u_D over the upper estimates K_D, μ_D, and the lower envelopes K^l_N, μ^l_N over the lower estimates K_N, μ_N calculated with

those RVEs would better represent the possible scatter ranges for the effective moduli of the random Voronoi elastic polycrystals at the 10000-grain-size RVE level.

Table 3. The scatter range measures $S^{\mu}_{VR} = \dfrac{\mu_V - \mu_R}{\mu_V + \mu_R}$, ... (in %) for the bounds in Tabs. 1, 2, and the crystal anisotropy index A^U of [20]

Crystal	A^U	S^{μ}_{VR}	S^{μ}_{HS}	S^{μ}	S^{μ}_{s}	S^{μ}_{DN}	S^{K}_{VR}	S^{K}_{HS}	S^{K}	S^{K}_{s}	S^{K}_{DN}
An_{96} (t)	1.0	8.6	1.9	0.42	0.31	0.49	2.7	0.58	0.12	0.10	0.23
T...(m)	1.8	14	3.3	1.4	0.84	1.1	4.1	1.2	0.32	0.22	0.23
E...(m)	2.8	19	4.8	3.0	1.5	1.2	21	5.2	3.6	1.8	1.6
O...(m)	3.6	25	8.4	3.9	2.7	1.4	14	4.4	2.0	1.5	0.95
L...(t)	4.5	27	9.4	4.6	3.6	1.8	28	9.4	5.1	3.8	2.1

4. CONCLUSIONS

Available bounds on the effective moduli of random polycrystals are resumed and compared against the numerical Dirichlet and Neumann estimates on a large 10000-grain-size random Voronoi polycrystal RVE for a number of triclinic and monoclinic base crystals reported in [18]. Though the major parts within the simulation results fall within the bounds of [10], about a half of the numerical Dirichlet upper estimate results still lie outside the bounds. Larger-size RVEs and possibly stronger numerical methods and computer resources are needed to test against the bounds.

Even at the same 10000-grain-size level, more random Voronoi polycrystal RVE realizations are needed to make the envelopes of the numerical Dirichlet and Neumann estimate results to really represent the size level and for comparisons with the bounds, starting with the Hashin-Shtrikman to the tighter ones.

Many more RVEs for the Voronoi polycrystal on the same RVE-size-level and larger RVEs are needed for comparisons with the bounds, to see how small the scatter ranges for the effective moduli of random Voronoi polycrystal really are, and could the bounds still be improved significantly.

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