Non-dilute multiple coated fiber composites with BCC microstructures: Effect of shape and fiber orientation

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Summary

A micromechanical approach for an accurate estimation of the effective moduli of composites reinforced with non-dilute distribution of coated particles is developed. In this treatment the short and long range interactions of the reinforcing particles are appropriately incorporated through the homogenizing eigenstrain field. For demonstration, the problems of composites with body centered cubic (BCC) distribution of coated spherical particles as well as coated ellipsoidal particles are solved. The effect of concentration, shape, and the orientation of the coated ellipsoidal reinforcements on overall behaviour of composites are thoroughly examined.

Introduction

Particle reinforced composites have been increasingly paid attention in engineering due to their prominent thermo-mechanical properties. A particle ensemble in a particulate composite, in general consists of an innermost phase (core) enclosed by several phases of distinct elastic constants, the outermost phase being its surrounding matrix. Functionally graded (FG) transition zones may be created inadvertently by chemical reactions between particles and matrix or may be created intentionally to improve thermo-mechanical properties.

The existing treatments to the estimation of overall mechanical behaviour of particulate composites are essentially divided into two categories, namely average field theory and homogenization theory, Hori and Nemat-Nasser [1]. In the average field theory, the overall mechanical behaviour is estimated using various averaging schemes such as the dilute distribution assumption, the self-consistent method or the Mori-Tanaka method, see Nemat-Nasser and Hori [2], and Hori and Nemat-Nasser [3] for the averaging schemes. On the other hand in the homogenization theory, the equivalent inclusion method is used as the fundamental base, which entails lengthy intricate mathematical manipulations. Because of this mathematical complexity, homogenization theory is mostly used for periodic microstructures while it is preferred to use simpler estimations of average filed theory in the case of random microstructure, however the averaging schemes are questionable when dealing with non-dilute distributions of particles.

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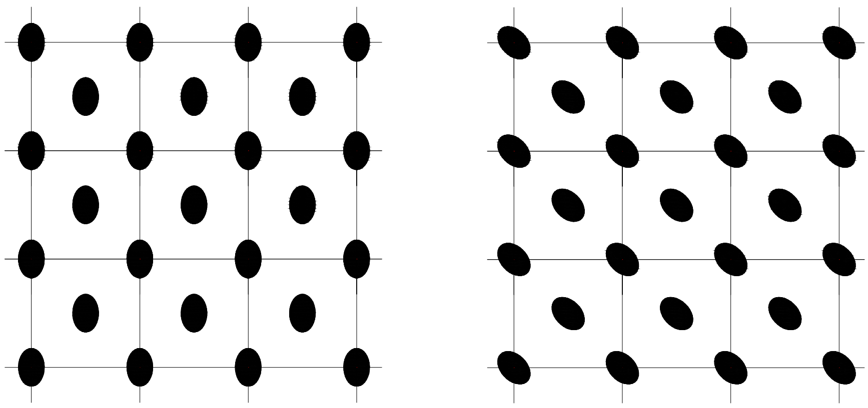
Nemat-Nasser and co-workers [4-6] used homogenization theory to estimate overall moduli of composites with non-dilute distribution of non-coated ellipsoidal particles with periodic microstructures. El Mouden et al. [7] used a different variation of the homogenization theory together with the aid of interfacial operators to derive effective properties of composites with aligned thinly coated particles. Shodja and Roumi [8], based on the equivalent inclusion method (EIM) for multi-inhomogeneities proposed by Shodja and Sarvestani [9], presented a theory for estimation of effective properties of composites with periodic distribution of multi-phase reinforcement particles. Their work is devoted to simple cubic (SC) arrangements of particles, and restricted to vertically aligned coated ellipsoidal reinforcements.

In this paper, based on homogenization a robust technique for the evaluation of the overall elastic properties of composites containing strongly interacting multiple coated ellipsoidal particles with BCC microstructure is presented. The present theory accommodates thickly and/or functionally graded coated particles, and rigorously accounts for both the intra-particle and inter-particle interactions. In the discussion section, as a simple demonstration, the experiment of thinly coated spherical particles of Amdouni et al. [8] will be modeled via BCC microstructure. The more interesting examples pertaining to high concentration of thickly / FG coated ellipsoidal particles will be considered. The object of the present work is to examine the effect of concentration , orientation and shape of such reinforcement particles on the overall mechanical properties of composites.



(a) (b)

Fig. 1: A typical BCC distribution of inhomogeneities,

a) Vertically aligned, b)Slanted Particles

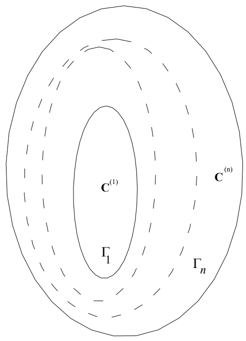


Fig. 2 : A typical multi-inhomogeneity

Average Elastic Fields: General Formulae for Coated Ellipsoidal Particles

Consider a BCC distribution of coated particles throughout the space as shown in Fig. 1, possible microstructures of a particle and its surrounding coatings are shown in Fig. 2. The homogenizing eigenstrains, $\boldsymbol{\varepsilon}^*(\mathbf{x})$ and the associated elastic strain field $\boldsymbol{\varepsilon}^d(\mathbf{x})$ may be expressed by Fourier series:

$$\boldsymbol{\varepsilon}^*(\mathbf{x}) = \sum_{\xi} \widehat{\boldsymbol{\varepsilon}}^*(\xi)\, e^{i\,\xi.\mathbf{x}}, \quad i = \sqrt{-1}, \ \xi_j = \frac{2\pi}{\Lambda_j} n_j, \qquad j=1,2,3 \quad \text{no sum on j} \tag{1}$$

$$\widehat{\boldsymbol{\varepsilon}}^*(\xi) = \frac{1}{V}\int_D \boldsymbol{\varepsilon}^*(\mathbf{x})\, e^{-i\,\xi.\mathbf{x}} d\mathbf{x} \tag{2}$$

$$\boldsymbol{\varepsilon}^d(\mathbf{x}) = \sum_{\xi} \widehat{\boldsymbol{\varepsilon}}(\xi)\, e^{i\,\xi.\mathbf{x}} \tag{3}$$

$$\widehat{\boldsymbol{\varepsilon}}(\xi) = \frac{1}{V}\int_D \boldsymbol{\varepsilon}^d(\mathbf{x})\, e^{-i\,\xi.\mathbf{x}} d\mathbf{x} \tag{4}$$

where $\sum_{\xi}$ is in general a triple summation corresponding to ξ_i, $i = 1,2,3$, which are in turn associated with the periodicity of the distribution of the eigenstrain field in the directions x_i, $i = 1,2,3$, respectively. $V = \Lambda_1 \Lambda_2 \Lambda_3$ is the volume of a typical cell D, Λ_i, $i = 1,2,3$ are the dimensions of the cell (parallel-piped).

In the homogenization scheme, each inhomogeneity is replaced with an equivalent inclusion containing appropriate eigenstrains such that the stress field in the original and homogenized problem is equivalent. Thus, for the α *th*-phase one may write

$$\mathbf{C}^{(\alpha)}:\left(\boldsymbol{\varepsilon}^{0}(\mathbf{x})+\boldsymbol{\varepsilon}^{d}(\mathbf{x})\right)=\mathbf{C}^{m}:\left(\boldsymbol{\varepsilon}^{0}(\mathbf{x})+\boldsymbol{\varepsilon}^{d}(\mathbf{x})-\boldsymbol{\varepsilon}^{*}(\mathbf{x})\right) \tag{5}$$

where $\mathbf{C}^{m}$ and $\mathbf{C}^{(\alpha)}$ are the elastic moduli of matrix and α *th*-phase, respectively. $\boldsymbol{\varepsilon}^{0}(\mathbf{x})$ is the far-field strain, $\boldsymbol{\varepsilon}^{d}(\mathbf{x})$ is the disturbance strain due to the presence of inhomogeneities. The equilibrium equation reads:

$$\nabla.[\mathbf{C}^{m}:(\boldsymbol{\varepsilon}^{0}+\boldsymbol{\varepsilon}^{d}-\boldsymbol{\varepsilon}^{*})]=0 \tag{6}$$

In the context of the present paper the far-field strain is assumed to be constant, substitution of Eqs. (1) and (3) into (6) yields

$$\boldsymbol{\varepsilon}^{d}(\mathbf{x})=\frac{1+e^{-i\,\xi.\,\mathbf{c}}}{V}\sum_{\xi}\mathbf{M}(\xi):\int_{\Omega}\boldsymbol{\varepsilon}^{*}(\mathbf{x}')\,e^{\,i\,\xi.(\mathbf{x}-\mathbf{x}')}d\mathbf{x}' \tag{7}$$

where $\mathbf{c}$ is a vector whose components are half of the dimensions of a typical cell and accounts for BCC distribution, Ω is the space occupied by an ellipsoidal particle and its surrounding coatings . For an isotropic matrix

$$\mathrm{M}_{ijkl}=\frac{1}{2\xi^{2}}\left\{\xi_{j}(\delta_{il}\xi_{k}+\delta_{ik}\xi_{l})+\xi_{i}(\delta_{jl}\xi_{k}+\delta_{jk}\xi_{l})\right\}-\frac{1}{1-\nu}\frac{\xi_{i}\xi_{j}\xi_{k}\xi_{l}}{\xi^{4}}+\frac{\nu}{1-\nu}\frac{\xi_{i}\xi_{j}}{\xi^{2}}\delta_{lk} \tag{8}$$

where

$$\xi^{2}=\xi.\xi \tag{9}$$

and ν is the Poisson's ratio of the matrix. The volume average of $\boldsymbol{\varepsilon}^{d}(\mathbf{x})$ over α *th*-phase, D_{α} with volume V_{α} is

$$\begin{aligned}<\boldsymbol{\varepsilon}^{d}(\mathbf{x})>_{\mathrm{D}_{\alpha}}&=\frac{1}{V_{\alpha}}\int_{\mathrm{D}_{\alpha}}\boldsymbol{\varepsilon}^{d}(\mathbf{x})\,d\mathbf{x}\\&=\frac{1+e^{-i\,\xi.\,\mathbf{c}}}{V\,V_{\alpha}}\Big(\sum_{\xi}\mathbf{M}(\xi):\mathrm{H}_{\mathrm{D}_{\alpha}}(\xi)\int_{\Omega}\boldsymbol{\varepsilon}^{*}(\mathbf{x}')\,e^{-i\,\xi.\,\mathbf{x}'}d\mathbf{x}'\Big)\end{aligned} \tag{10}$$

Eqn. (10) gives the average strain field in a particulate composite due to the presence of the eigenstrain field. In Eq. (10)

$$\mathrm{H}_{\mathrm{D}_\alpha}(\xi) = \int_{\mathrm{D}_\alpha} \mathrm{e}^{\,\mathrm{i}\xi.\mathbf{x}}\mathrm{d}\mathbf{x} \tag{11}$$

For a slanted ellipsoidal particle, Ω whose semi-principal axes a_i, $i=1,2,3$ do not coincide with directions of the global Cartesian coordinates x_i, $i=1,2,3$

$$\mathrm{H}(\xi) = \frac{3\,(\sin\eta - \eta\cos\eta)}{\eta^3}\mathrm{V}_\Omega \tag{12}$$

$$\eta = \sqrt{(\mathrm{a}_1\bar{\xi}_1)^2 + (\mathrm{a}_2\bar{\xi}_2)^2 + (\mathrm{a}_3\bar{\xi}_3)^2} \tag{13}$$

$$\bar{\xi} = \mathbf{Q}\,.\xi \tag{14}$$

Where the ξ_i, $i=1,2,3$ are along the corresponding directions of x_i, $i=1,2,3$, where as the directions of $\bar{\xi}_i$, $i=1,2,3$ coincide with the local coordinates along the principal axes $\bar{x}_i$, $i=1,2,3$ of ellipsoid. V_Ω is the volume of Ω, and $\mathbf{Q}$ is the rotation matrix.

Elastic Moduli of a Particulate Composite

The overall elastic moduli of a statistically homogeneous composite is defined by $\mathbf{C}^*$, which relates the average stress and strain fields as

$$\langle\boldsymbol{\sigma}\rangle_{\mathrm{D}} = \mathbf{C}^* : \langle\boldsymbol{\varepsilon}\rangle_{\mathrm{D}} \tag{15}$$

where

$$\langle\boldsymbol{\sigma}\rangle_{\mathrm{D}} = \frac{1}{V}\int_D \boldsymbol{\sigma}(\mathbf{x})\,\mathrm{d}\mathbf{x} \tag{16}$$

$$\langle\boldsymbol{\varepsilon}\rangle_{\mathrm{D}} = \frac{1}{V}\int_D \boldsymbol{\varepsilon}(\mathbf{x})\,\mathrm{d}\mathbf{x} \tag{17}$$

are average elastic stress and strain fields, respectively. On the other hand, by implementing homogenization theory, the elastic fields can be written as:

$$\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{C}^{\mathrm{m}} : \left(\boldsymbol{\varepsilon}^{0} + \boldsymbol{\varepsilon}^{\mathrm{d}}(\mathbf{x}) - \boldsymbol{\varepsilon}^{*}(\mathbf{x})\right) \tag{18}$$

$$\boldsymbol{\varepsilon}(\mathbf{x}) = \boldsymbol{\varepsilon}^{0} + \boldsymbol{\varepsilon}^{\mathrm{d}}(\mathbf{x}) \tag{19}$$

Combining Eqs. (15) through (19) and using $\left\langle \boldsymbol{\varepsilon}^{\mathrm{d}} \right\rangle_{\mathrm{D}} = \mathbf{0}$, one obtains

$$\mathbf{C}^{*} : \boldsymbol{\varepsilon}^{0} = \mathbf{C}^{\mathrm{m}} : \boldsymbol{\varepsilon}^{0} - \mathbf{C}^{\mathrm{m}} : \left\langle \boldsymbol{\varepsilon}^{*} \right\rangle_{\mathrm{D}} \tag{20}$$

Thus, the overall elastic moduli of a particulate composite can be estimated provided that the average eigenstrain field is known. Accurate estimations of averages require special attention addressed in the following section.

A Treatment for Thick/FG Coatings

It should be emphasized that, at high concentrations the variation of eigenstrain field within the phases of the particles may be highly nonlinear, and so the size of the region over which the average of eigenstrain field is computed is the key factor responsible for possible deterioration of the accuracy of the results. To ensure that the average of eigenstrain field is tolerably accurate, each domain will be subdivided into small enough sub-domains, followed by the treatment given by Shodja and Sarvestani [9] for multi-inhomogeneity systems. Suppose that, after subdividing the desired phases of a particle into a rational number of sub-domains, a total of *NS* sub-domains are encountered. The average of the elastic strain over a sub-domain Γ_{α} is now obtained by modifying Eq. (10) as follows

$$<\boldsymbol{\varepsilon}^{\mathrm{d}}>_{\Gamma_{\alpha}} = \frac{1+\mathrm{e}^{-\mathrm{i}\,\xi.\,\mathbf{c}}}{V V_{\alpha}} \big(\sum_{\xi} \mathbf{M}(\xi) : \mathrm{H}_{\Gamma_{\alpha}}(\xi) \sum_{\gamma=1}^{\mathrm{NS}} \int_{\Gamma_{\gamma}} \boldsymbol{\varepsilon}^{*(\gamma)}(\mathbf{x}')\, \mathrm{e}^{-\mathrm{i}\,\xi.\,\mathbf{x}'} \mathrm{d}\mathbf{x}'\big) \tag{21}$$

Using the superposition proposed by Shodja and Sarvestani [9]

$$<\boldsymbol{\varepsilon}^{\mathrm{d}}>_{\Gamma_{\alpha}} = \frac{1+\mathrm{e}^{-\mathrm{i}\,\xi\,.\,\mathbf{c}}}{V V_{\alpha}} \sum_{\xi} \mathbf{M}(\xi) : \mathrm{H}_{\Gamma_{\alpha}}(\xi) \{ \int_{\Sigma_{\mathrm{NS}}} \boldsymbol{\varepsilon}^{*(\mathrm{NS})}\, \mathrm{e}^{-\mathrm{i}\,\xi.\,\mathbf{x}'} \mathrm{d}\mathbf{x}'$$

$$+ \sum_{\gamma=1}^{\mathrm{NS-1}} \int_{\Sigma_{\gamma}} \left(\boldsymbol{\varepsilon}^{*(\gamma)} - \boldsymbol{\varepsilon}^{*(\gamma+1)}\right) \mathrm{e}^{-\mathrm{i}\,\xi.\,\mathbf{x}'} \mathrm{d}\mathbf{x}' \; \} \tag{22}$$

For thin enough phases we will get

$$< \boldsymbol{\varepsilon}^{d} >_{\Gamma_\alpha} = \frac{1+e^{-i\,\xi\,.\,c}}{V\,V_\alpha} \sum_{\xi} \mathbf{M}(\xi) : \mathrm{H}_{\Gamma_\alpha}(\xi) \{ \left\langle \boldsymbol{\varepsilon}^{*(NS)} \right\rangle_{D_{NS}} \mathrm{H}_{\Sigma_{NS}}(-\xi)$$

$$+ \sum_{\gamma=1}^{NS} (\left\langle \boldsymbol{\varepsilon}^{*(\gamma)} \right\rangle_{\Gamma_\gamma} - \left\langle \boldsymbol{\varepsilon}^{*(\gamma+1)} \right\rangle_{\Gamma_{\gamma+1}}) \, \mathrm{H}_{\Sigma_\gamma}(-\xi) \ \} \tag{23}$$

where $\Sigma^{1} = \Gamma^{1}$ and $\Sigma^{\mathbf{j}} = \Gamma^{\mathbf{j}} \bigcup \Sigma^{\mathbf{j-1}}$.

The average consistency equation associated with the $\alpha\,th$-phase is readily available from Eq. (5)

$$\mathbf{C}^{(\alpha)} : \{ \boldsymbol{\varepsilon}^{0} + \left\langle \boldsymbol{\varepsilon}^{d}(\mathbf{x}) \right\rangle_{\Gamma_\alpha} \} = \mathbf{C}^{m} : \{ \boldsymbol{\varepsilon}^{0} + \left\langle \boldsymbol{\varepsilon}^{d}(\mathbf{x}) \right\rangle_{\Gamma_\alpha} - \left\langle \boldsymbol{\varepsilon}^{*}(\mathbf{x}) \right\rangle_{\Gamma_\alpha} \} \tag{24}$$

Upon substitution of (23) into (24) the average eigenstrain field over each sub-domain is calculated, subsequently with the aid of Eq. (20) the overall elastic moduli will be estimated.

Numerical Results

As mentioned earlier the present theory is developed for periodic distribution of the thickly coated reinforcement particles having BCC microstructures. The formulation for random distribution departs greatly from the periodic treatment and will be presented in another work. In subsequent sections, it is intended to examine several examples based on BCC type periodicity.

Experiment of Amdouni et al.

Amdouni et al. [8] performed an experimental study on epoxy matrix composites reinforced with thinly coated glass beads. The introduction of the elastomer as a thin layer at the filler / matrix interface improves the impact and fracture properties of the composite without reducing the elastic modulus. The elastic modulus, E and Poisson's ratio ν of the constituent materials are $\mathrm{E}^{m} = 2.9\,\mathrm{GPa}$, $\nu^{m} = 0.4$, $\mathrm{E}^{f} = 73\,\mathrm{GPa}$, $\nu^{f} = 0.2$, $\mathrm{E}^{c} = 10\,\mathrm{GPa}$, and $\nu^{c} = 0.49$, where the superscripts m, f, and c denote matrix, filler, and coating, respectively. Verification of this experiment is the simplest case considered by the present theory. The ratio of the thickness of the coating to the radius of

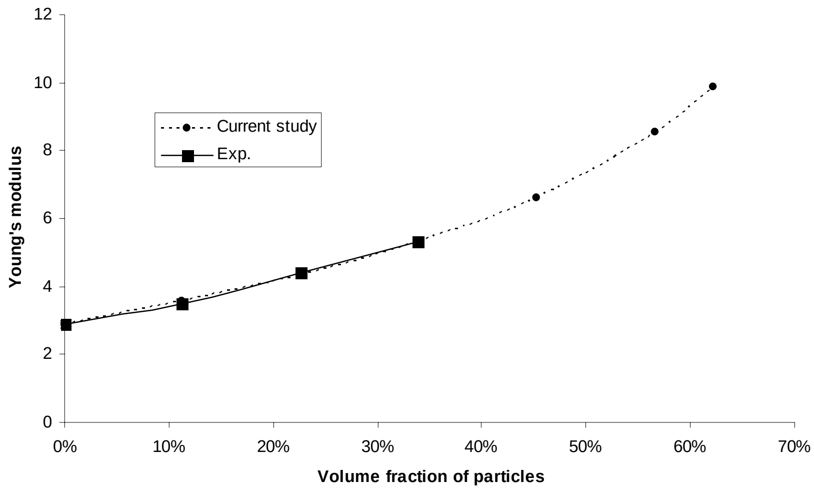


Fig. 3 : Comparison of theoretical and experimental values of Young's modulus

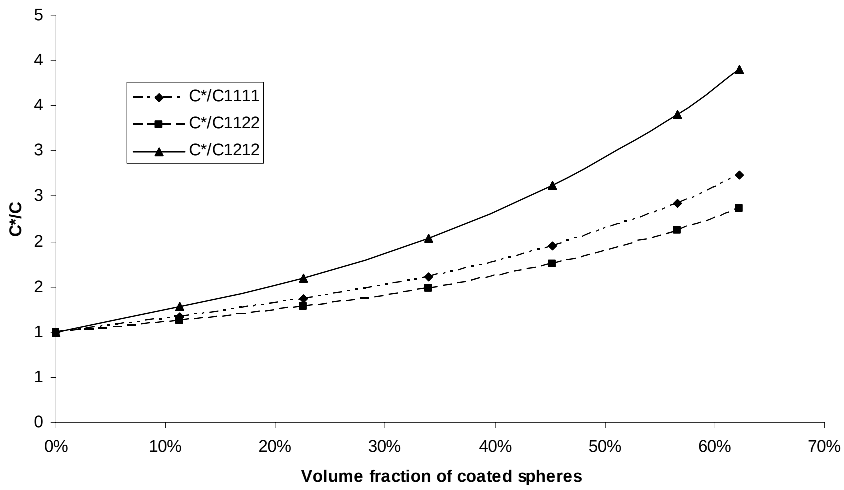


Fig. 4 : The computed values of elastic moduli

the filler is taken to be 4.2%. From Fig. 3, for the Young's modulus versus volume fraction of particles, it is observed that there is a good correspondence between the experimental data of Amdouni et al. [8] and the results obtained by the present theory.

The experimental data are available for volume fractions of up to 33%, where as the predictions by the theory extend to volume fractions of up to 60%. The values of C^*/C_{1111}, C^*/C_{1122}, C^*/C_{1212} versus volume fraction have also been computed, and are depicted in Fig. 4. The results are in good agreement with those given by El Mouden et al. [7].

Composites Containing Thickly Coated Ellipsoidal Particles

In this section the ratios of the shear moduli are taken to be $\mu^{\text{fiber}}/\mu^{\text{matrix}} = 10$, $\mu^{\text{coating}}/\mu^{\text{matrix}} = 5.5$, and the Poisson's ratio $\nu = 0.3$ for all phases. As the second demonstration, a composite containing thickly coated ellipsoidal particles, whose ratios of the principal axes are $a_2/a_1 = a_3/a_1 = 0.7$ is examined. The long axis of each coated particle is vertically oriented, and the ratio of the thickness of the coating (along the principal directions of the ellipsoid) to the respective radius of the ellipsoid is 0.25. The ratios of the dimensions of the cell D are taken to be $\Lambda_2/\Lambda_1 = \Lambda_3/\Lambda_1 = 0.7$. The values of C^*/C_{1212}, C^*/C_{2323}, and the bulk modulus k^*/k are plotted against the total volume fraction of the core particle and its coating, the results are displayed in Fig. 5. To increase the accuracy; every coating is subdivided into four layers. The volume fraction of 52% corresponds to a rather high concentration of particles.

The effect of shape on the components of the overall elastic moduli $\mathbf{C}^*$, Young's moduli E^*, and the bulk modulus k^* is explored through variation of the dimensions of the principal axes of ellipsoid. The results of such studies are clearly demonstrated in Table 1, where $P1 = a_2/a_1 = \Lambda_2/\Lambda_1$, and $P2 = a_3/a_1 = \Lambda_3/\Lambda_1$. Note that, the volume fractions $f^{\text{fiber}} = 30\%$, and $f^{\text{coating}} = 22\%$ are kept fixed in all the cases.

Table 1 : Study of the shape effect

P1	*P2*	C^*/C_{1212}	C^*/C_{2323}	C^*/C_{3131}	k^*/k	E^*/E_1	E^*/E_2	E^*/E_3
1.00	1.00	2.439	2.439	2.439	2.160	2.263	2.263	2.263
0.70	0.70	2.391	2.436	2.391	2.218	3.084	2.106	2.106
0.50	0.50	2.189	2.313	2.189	2.228	4.215	1.928	1.928
0.30	0.30	1.825	1.863	1.825	2.007	4.506	1.671	1.671
0.10	0.10	1.402	1.377	1.402	1.746	4.688	1.375	1.375
0.10	0.50	1.387	1.389	4.345	2.497	4.651	1.673	4.136

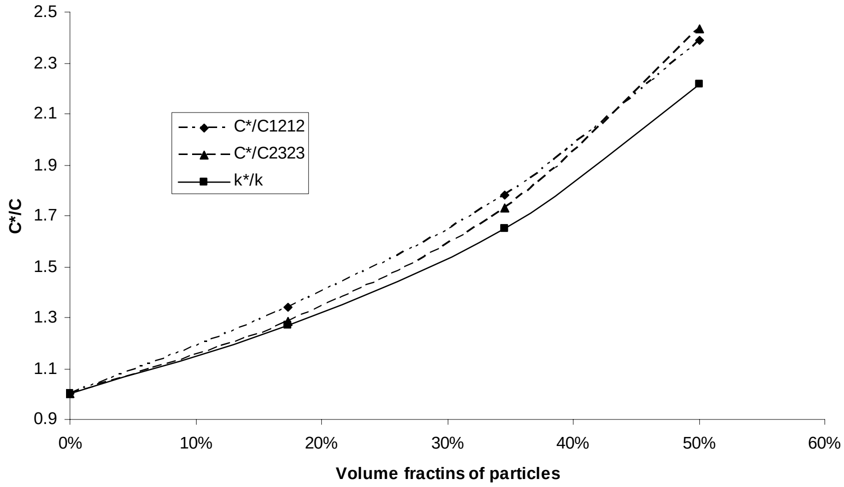


Fig. 5 : Prediction of bulk and shear moduli

Particulate Composites with Thick Interfacial Zones

This example concerns with composite systems reinforced with ellipsoidal reinforcements having BCC microstructure. It is assumed that there is a transition zone between each particle and the matrix, where the shear moduli, of the interphase particle varies according to

$$\mu^{coating}(r) = \mu^{fiber} + (\mu^{matrix} - \mu^{fiber})\left(\frac{r - r^{fiber}}{\Delta}\right)^2 \quad , \qquad r^f \leqslant r \leqslant r^f + \Delta\,, \tag{25}$$

where r is the distance away from the fiber-interphase boundary, Δ is the thickness of the transition zone, and r^f is the radius of the fiber.

The results of eight different configurations are tabulated in Table 2. The ratios of the principal axes of each ellipsoid are, 1; 0.5:0.75 in all cases and the dimensions of the cell D are equal, $\Lambda_1 = \Lambda_2 = \Lambda_3$. The first column of Table 2 refers to the particles' orientation, determined by specifying the orientations of the long and short axes denoted by the subscripts L and S, respectively. The ratio of the shear moduli of fiber to matrix is

$\mu^{fiber}/\mu^{matrix} = 10$. The volume fraction of fibers, $f^{fiber} = 12\%$, and that of the fibers and their transition region, $f^{particle} = 20\%$ are kept fixed in all cases.

Table 2 : Study of the particle orientation

Particle Orientation	C^*/C_{1212}	C^*/C_{2323}	C^*/C_{3131}	k*/k	E^*/E_{11}	E^*/E_{22}	E^*/E_{33}
$(1,0,0)_L,(0,1,0)_S$	1.303	1.307	1.377	1.277	1.422	1.254	1.325
$(1,0,0)_L,(0,0,1)_S$	1.377	1.307	1.303	1.277	1.422	1.325	1.254
$(1,0,0)_L,(0,.866,-.5)_S$	1.319	1.320	1.355	1.276	1.423	1.263	1.296
$(1,0,0)_L,(0,.707,-.707)_S$	1.336	1.324	1.336	1.275	1.422	1.276	1.276
$(0,0,1)_L,(0,1,0)_S$	1.307	1.303	1.377	1.277	1.325	1.254	1.422
$(.75,-.650,-.125)_L,(.433,.625,-.650)_S$	1.367	1.319	1.319	1.272	1.312	1.365	1.265
$(.577,.577,.577)_L,(-.707,.707,0)_S$	1.352	1.364	1.321	1.272	1.264	1.312	1.360
$(.5,-.854,.146)_L,(.5,.146,-.854)_S$	1.340	1.343	1.341	1.269	1.277	1.317	1.306

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