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On the overall elastic moduli of composites with spherical coated fillers

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Abstract

A micromechanical approach is presented to estimate the overall linear elastic moduli of three phase composites consisting of two phase coated spherical particles randomly dispersed in a homogeneous isotropic matrix. The theoretical method is based on Eshelby's equivalent inclusion method and its recent extension by Shodja and Sarvestani [J. Appl. Mech. 68 (2001) 3] to evaluate the local field variables in case of double (multi) inhomogeneities. Using Tanaka–Mori theorem [J. Elasticity 2 (1972) 199] and a decomposition of Green's function integral equation, the pairwise average phase values of stress and strain in two interacting coated particles are estimated. Following Ju and Chen [Acta Mech. 103 (1994) 103; Acta Mech. 103 (1994) 123] the ensemble phase volume average of stress and strain fields can be evaluated within a representative volume element containing a finite number of coated particles. Comparisons with classical bounds are presented to illustrate the accuracy of the proposed method.

Keywords: Composites; Interphase; Coating; Filler; Overall mechanical properties

1. Introduction

The present paper proposes a micromechanical method for the estimation of the global mechanical properties of linear elastic composites with coated spherical microstructures. It is recognized in the literature that the mechanical performance of composite materials is significantly influenced by the presence of interphase layers between constituents (filler and matrix). Hence, this effect was extensively studied over the last decade and a number of micromechanical techniques have been developed. Many of these studies have been devoted to the study of the variation of local field quantities within the constituents. Examples from this category include the methods proposed by Walpole (1978), Mikata and Taya (1985a,b), Benveniste et al. (1989) and Cherkaoui et al. (1994) and recently by Shodja and Sarvestani (2001). Likewise, various techniques have been adopted to predict the effective properties of linear elastic composites in

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presence of interphase layers, e.g., Qiu and Weng (1991), Cherkaoui et al. (1995), Dunn and Ledbetter (1995), El Mouden et al. (1998), Buryachenko and Rammerstorfer (2000) and Ozmusul and Picu (2002a).

The effective mechanical properties of composite materials are defined as the relationship between ensemble averages of the local stress and strain fields over a representative volume element (RVE) of the heterogeneous body (homogenization). A number of micromechanical approaches were proposed in literature to describe how the mechanical properties and microgeometry of constituents affect the overall behavior of composite materials. For comprehensive reviews the reader is referred to the monographs by Beran (1967), Willis (1982), Mura (1987), Nemat-Nasser and Hori (1993) and Torquato (2001). Most of the analytical homogenization procedures are based on Green's function techniques and the Eshelby's equivalent inclusion method (1957, 1959, 1961). Adopting the terminology used by Mura (1987), in the present study the inclusion and inhomogeneity are differentiated in the way that an inclusion is a finite domain with a distribution of eigenstrain, its elastic moduli being the same with those of the matrix; whereas an inhomogeneity is a subdomain with elastic moduli different than the matrix. According to Eshelby's theory, the eigenstrain and consequently strain and stress fields inside a single ellipsoidal inhomogeneity embedded in an unbounded homogeneous isotropic matrix are uniform if the far field applied stress (strain) is uniform.

In the presence of an interlayer between the filler and the matrix, in general, strain and stress fields within the inhomogeneity are not uniform. A number of researchers attempted to bypass the difficulty associated with the evaluation of these non-uniform fields introducing the assumption of thin coating layer (Cherkaoui et al., 1995; El Mouden et al., 1998; Buryachenko and Rammerstorfer, 2000). In this framework, the interphase zone is assumed to have vanishing thickness and its stress and strain fields are obtained using Hill's interfacial operators (1972). This restrictive assumption is not always appropriate and is partly relaxed in the present work. For instance, recent researches performed about the structure of polymer based nanocomposites show the thickness of the affected polymeric zone around the dispersed rigid nanofillers can be comparable to the filler size. This can greatly enhance the volume fraction of interphase zone in the material, which is believed to be the main reason for different macroscopic behavior of this kind of material compared to the polymers reinforced with the micron-size particles (Ozmusul and Picu, 2002a,b; Picu et al., 2003).

An alternative analytical approach to the estimation of macroscopic properties of linear elastic composites with coated microcomponents is a generalization of the composite sphere or cylinder assemblage models proposed by Hashin (1962), Hashin and Rosen (1964) and Christensen and Lo (1979) for self-consistent or generalized self-consistent method in multiphase composites (Qiu and Weng, 1991; Jasiuk and Kouider, 1993; Chu and Rokhlin, 1995). Benveniste et al. (1989) and Chen et al. (1990) presented another solution to this problem, but based on the Mori and Tanaka (1973) scheme. The multi-inclusion model of Hori and Nemat-Nasser (1993, 1994) for multiphase composites was also utilized as an adequate model for functionally graded composites, e.g. Hori and Nemat-Nasser (1993), Dunn and Ledbetter (1995) and Li (2000).

In this paper this problem is approached using a new method applicable to finite concentration of thickly coated particulate systems. The interaction between the particle and interphase is evaluated by means of Eshelby's equivalent inclusion method (EIM) and Tanaka and Mori theorem (1972), following Shodja and Sarvestani (2001). It is assumed that the randomly dispersed spherical fillers of similar size are covered by homogeneous interphase layers with certain thickness and there are well bond interfaces between the various constituents. It is also assumed that all statistical properties corresponding to any arbitrary representative mesodomain of the composite body are statistically homogeneous. An ergodic random field is called statistically homogeneous if multipoint statistical moments of any order are shift invariant functions of spatial variables and, hence, the ensemble averaging could by replaced be volume averaging (Kröner, 1972).

2. Statistical preliminaries

Consider N spherical fillers randomly dispersed in a statistically homogeneous mesodomain V. Let $\mathbf{x}_i$ be the position vector drawn to the center of each particle (O_i) in a Cartesian system with arbitrary origin. A function $\varphi(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N)$ of space coordinates $\mathbf{x}$, and N position vectors $\mathbf{x}_i$ may represent any statistical quantity corresponding to these interacting inhomogeneities. For this random structure, it is possible to define the probability density of the set of variables $(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N)$ denoted by $p(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N)$. This function has the following properties (Mal and Bose, 1974):

$$\begin{aligned} p(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N) &= p(\mathbf{x}_j)p(\mathbf{x}_1, \mathbf{x}_2, \ldots, ', \ldots, \mathbf{x}_N|\mathbf{x}_j) \\ &= p(\mathbf{x}_j)p(\mathbf{x}_i|\mathbf{x}_j)p(\mathbf{x}_1, \mathbf{x}_2, \ldots, ', \ldots, ', \ldots, \mathbf{x}_N|\mathbf{x}_i, \mathbf{x}_j), \end{aligned} \tag{1a}$$

$$p(\mathbf{x}_i) = p(\mathbf{x}_1), \quad p(\mathbf{x}_i|\mathbf{x}_j) = p(\mathbf{x}_2|\mathbf{x}_1) \quad (i \neq j), \tag{1b}$$

where the vertical lines in the arguments denote the usual conditional probabilities. A prime in the first part of (1a) means $\mathbf{x}_j$ is not in the list, while two primes in the second part of (1a) means both $\mathbf{x}_i$ and $\mathbf{x}_j$ are absent.

Since the composite is statistically homogeneous, the positions of a single sphere are equally probable within V, and hence its distribution is uniform with density,

$$\begin{aligned} p(\mathbf{x}_i) &= \frac{1}{V}, \quad \mathbf{x}_i \in V, \\ &= 0, \quad \mathbf{x}_i \notin V. \end{aligned} \tag{2}$$

The distribution of spherical inhomogeneities relative to a given particle at $\mathbf{x}_j$ is spherically symmetric and hence $p(\mathbf{x}_i|\mathbf{x}_j)$ is a function of $|\mathbf{x}_i - \mathbf{x}_j|$ alone,

$$\begin{aligned} p(\mathbf{x}_i|\mathbf{x}_j) &= \frac{1}{V} g(\mathbf{x}_i - \mathbf{x}_j), \quad \mathbf{x}_i \in V, \\ &= 0, \quad \mathbf{x}_i \notin V, \end{aligned} \tag{3}$$

where $g(\mathbf{x}_i - \mathbf{x}_j)$ is a decreasing function of $|\mathbf{x}_i - \mathbf{x}_j|$, called radial pair distribution function. Since the particles cannot interpenetrate and are independent when they are infinitely apart,

$$g(\mathbf{x}_i - \mathbf{x}_j) = 0, \quad \text{for } |\mathbf{x}_i - \mathbf{x}_j| < 2a, \tag{4a}$$

$$g(\mathbf{x}_i - \mathbf{x}_j) = 1, \quad \text{for } |\mathbf{x}_i - \mathbf{x}_j| \to \infty, \tag{4b}$$

where a is the radius of each spherical inhomogeneity. Different forms of the pair distribution function satisfying conditions (4a) and (4b) have been suggested in the literature. In this paper, the following function is used for $g(\mathbf{x}_i - \mathbf{x}_j)$:

$$g(\mathbf{x}_i - \mathbf{x}_j) = 1 + \frac{4}{\pi}\left[\pi - 2\sin^{-1}\left(\frac{\hat{r}}{2}\right) - \hat{r}\left(1 - \frac{\hat{r}^2}{4}\right)^{1/2}\right]\phi, \tag{5}$$

where $\hat{r} = |\mathbf{x}_i - \mathbf{x}_j|/2a$ and ϕ is the volume fraction (Hansen and McDonald, 1986).

We denote the conditional expectations of function φ, when either O_i or O_i and O_j together are held fixed as

$$\langle\varphi\rangle_i = \int \ldots \int \varphi p(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N|\mathbf{x}_i)\mathrm{d}\mathbf{x}_1 \ldots \mathrm{d}\mathbf{x}_N,$$
$$\langle\varphi\rangle_{ij} = \int \ldots \int \varphi p(\mathbf{x}_1, \mathbf{x}_2, \ldots, \mathbf{x}_N|\mathbf{x}_i, \mathbf{x}_j)\mathrm{d}\mathbf{x}_1 \ldots \mathrm{d}\mathbf{x}_N. \quad (6)$$

3. Average fields within a pair of interacting coated spherical fillers

Consider a pair of identically coated spherical fillers located in an infinite isotropic matrix as depicted in Fig. 1. Each coated particle is composed of a spherical filler surrounded by a homogeneous coating. The filler and the coating have different elastic moduli and the ensemble is embedded in an infinite isotropic matrix with yet another elasticity. Filler, coating and matrix are specified as Ω_i, Ψ_i, and Φ domains ($i = 1, 2$) with their elastic moduli being $\mathbf{C}^1$, $\mathbf{C}^2$, and $\mathbf{C}$ respectively. Either of the two coated particles shown in Fig. 1 is referred as a double-inhomogeneity system. Note that Ω_i and Ψ_i may be anisotropic in general. The infinite elastic medium is subjected to uniform far field strain, ε_{ij}^0 (stress, σ_{ij}^0).

According to the EIM, the double-inhomogeneities can be replaced with the equivalent double-inclusions, as shown in Fig. 2. This equivalency holds for proper selection of homogenizing eigenstrain fields $\varepsilon_{ij}^{*(1)}(\mathbf{x})$ and $\varepsilon_{ij}^{*(2)}(\mathbf{x})$ defined in the various domains as

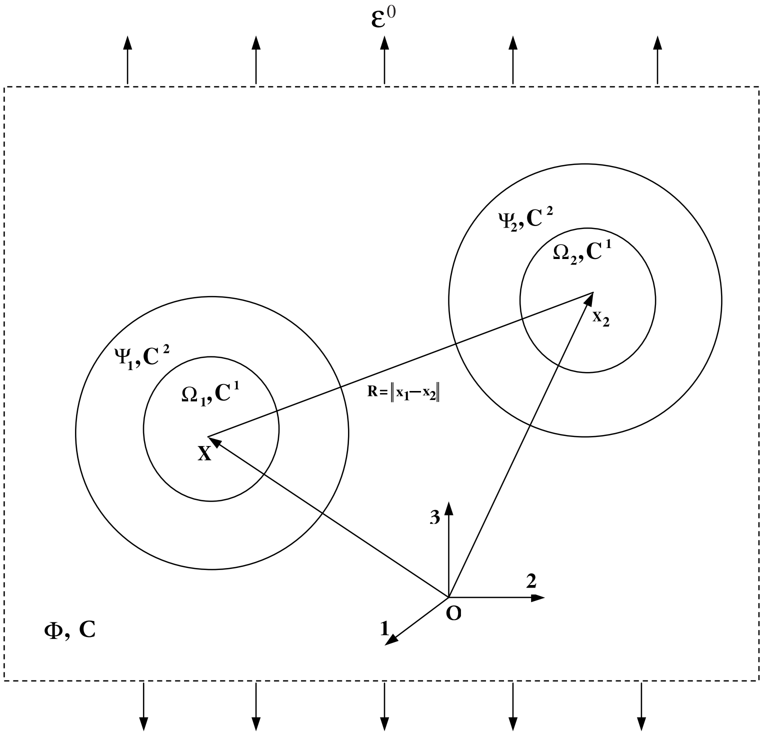

Fig. 1. Two spherical coated fillers embedded in an infinite isotropic matrix.

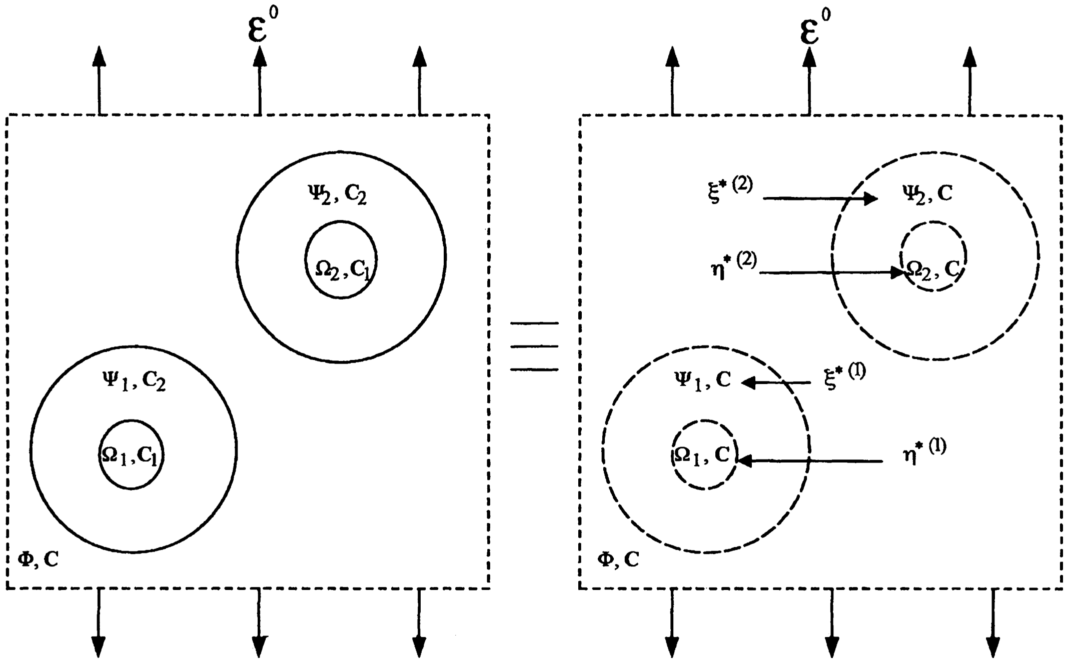

Fig. 2. The pair of interacting double-inhomogeneities can be replaced with the equivalent double-inclusions with proper homogenizing eigenstrains.

$$\varepsilon_{ij}^{*(m)}(\mathbf{x}) = \begin{cases} \eta_{ij}^{*(m)}(\mathbf{x}) & \mathbf{x} \in \Omega_m, \\ \xi_{ij}^{*(m)}(\mathbf{x}) & \mathbf{x} \in \Psi_m \,, \\ 0 & \mathbf{x} \in D, \end{cases} \quad (m = 1, 2). \tag{7}$$

Employing the EIM, the following consistency conditions are obtained ($m = 1$, 2):

$$\begin{aligned} C_{ijkl}^{m}\left(\varepsilon_{kl}^{0} + \varepsilon_{kl}^{\mathrm{d}}(\mathbf{x})\right) &= C_{ijkl}\left(\varepsilon_{kl}^{0} + \varepsilon_{kl}^{\mathrm{d}}(\mathbf{x}) - \eta_{kl}^{*(m)}(\mathbf{x})\right), \quad \mathbf{x} \in \Omega_m, \\ C_{ijkl}^{m}\left(\varepsilon_{kl}^{0} + \varepsilon_{kl}^{\mathrm{d}}(\mathbf{x})\right) &= C_{ijkl}\left(\varepsilon_{kl}^{0} + \varepsilon_{kl}^{\mathrm{d}}(\mathbf{x}) - \xi_{kl}^{*(m)}(\mathbf{x})\right), \quad \mathbf{x} \in \Psi_m, \end{aligned} \tag{8}$$

where $\varepsilon_{ij}^{\mathrm{d}}(\mathbf{x})$ denotes the disturbance in strain field.

The total disturbance in strain due to the presence of the eigenstrain field $\varepsilon_{ij}^{*(m)}(\mathbf{x})$ in the double-inclusion $\Sigma_m = \Psi_m \cup \Omega_m$ can be expressed as (Mura, 1987)

$$\varepsilon_{ij}^{\mathrm{d}}(\mathbf{x}) = \int_{\Sigma_m} \Gamma_{ijkl}(\mathbf{x} - \mathbf{x}')\varepsilon_{kl}^{*(m)}(\mathbf{x}')\mathrm{d}\mathbf{x}', \quad (m = 1, 2), \tag{9}$$

where $\Gamma_{ijkl}(\mathbf{x} - \mathbf{x}')$ is a fourth order tensor defined by

$$\Gamma_{ijkl}(\mathbf{x} - \mathbf{x}') = -\tfrac{1}{2}C_{rskl}[G_{ir,sj}(\mathbf{x} - \mathbf{x}') + G_{jr,si}(\mathbf{x} - \mathbf{x}')]. \tag{10}$$

$G_{ij}(\mathbf{x} - \mathbf{x}')$ is the elastic Green's function for the infinite medium, given for the isotropic case in the form of (Mura, 1987)

$$G_{ij}(\mathbf{x} - \mathbf{x}') = \frac{1}{4\pi\mu}\frac{\delta_{ij}}{|\mathbf{x} - \mathbf{x}'|} - \frac{1}{16\pi\mu(1 - \nu)}\frac{\partial^2}{\partial x_i \partial x_j}|\mathbf{x} - \mathbf{x}'|^2, \tag{11}$$

where μ and v are the shear modulus and Poison's ratio and δ_{ij} is the Kronecker delta. Using the Eqs. (7) and (9), the disturbance in strain reads

$$\boldsymbol{\varepsilon}^{\mathrm{d}}(\mathbf{x}) = \int_{\Omega 1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}' + \int_{\Psi_1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}' + \int_{\Omega_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}' + \int_{\Psi_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'. \tag{12}$$

It is possible to decompose the second and fourth integrals in (12) as

$$\int_{\Psi m} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(m)}(\mathbf{x}')\mathrm{d}\mathbf{x}' = \int_{\Sigma_m} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(m)}(\mathbf{x}')\mathrm{d}\mathbf{x}' - \int_{\Omega_m} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(m)}(\mathbf{x}')\mathrm{d}\mathbf{x}' \quad (m=1,2). \tag{13}$$

This makes possible expressing all terms in (12) as integrals over simply connected spherical domains. Shodja and Sarvestani (2001) used this decomposition for the evaluation of local stress and strain fields in a general double-inhomogeneity system. Here the objective is to estimate the overall macroscopic behavior and hence we look for volume averages of field quantities. Taking the volume average of (12) over Σ_1 and Ω_1, gives

$$\begin{aligned}
\bar{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Sigma_1} &= f\bar{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega_1} + (1-f)\bar{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Psi_1} \\
&= \frac{1}{\Sigma_1}\int_{\Sigma_1}\int_{\Omega 1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} + \frac{1}{\Sigma_1}\int_{\Sigma_1}\int_{\Sigma_1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} \\
&\quad - \frac{1}{\Sigma_1}\int_{\Sigma_1}\int_{\Omega_1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} + \frac{1}{\Sigma_1}\int_{\Sigma_1}\int_{\Omega_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} \\
&\quad + \frac{1}{\Sigma_1}\int_{\Sigma_1}\int_{\Psi_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x},
\end{aligned} \tag{14a}$$

$$\begin{aligned}
\bar{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega_1}(\mathbf{x}) &= \frac{1}{\Omega_1}\int_{\Omega_1}\int_{\Omega_1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} + \frac{1}{\Omega_1}\int_{\Omega_1}\int_{\Sigma_1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} \\
&\quad - \frac{1}{\Omega_1}\int_{\Omega_1}\int_{\Omega_1} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(1)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} + \frac{1}{\Omega_1}\int_{\Omega_1}\int_{\Omega_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} \\
&\quad + \frac{1}{\Omega_1}\int_{\Omega_1}\int_{\Psi_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x},
\end{aligned} \tag{14b}$$

where $f = \Omega/\Sigma$. The bar above any quantity represents the volume average over the region shown in the subscript. For example, the volume average of quantity $\boldsymbol{\chi}(\mathbf{x})$ over region α is $\bar{\boldsymbol{\chi}}_\alpha = 1/\alpha\int_\alpha \boldsymbol{\chi}(\mathbf{x})\mathrm{d}\mathbf{x}$.

Using Tanaka and Mori theorem (1972) and the fact that $\boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}') = \boldsymbol{\Gamma}(\mathbf{x}'-\mathbf{x})$, Eq. (14a) can be written as

$$f\bar{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega_1} + (1-f)\bar{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Psi_1} = \mathbf{S}^{\Sigma_1}\left(f\bar{\boldsymbol{\eta}}^{*(1)}_{\Omega_1} + (1-f)\bar{\boldsymbol{\xi}}^{*(1)}_{\Psi_1}\right) + \frac{1}{\Sigma_1}\int_{\Sigma_1}\int_{\Omega_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} + \frac{1}{\Sigma_1}\int_{\Sigma_1}\int_{\Psi_2} \boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x}, \tag{15a}$$

where $\mathbf{S}^\alpha$ is the well-known Eshelby tensor for interior points of region α. Also application of Hori and Nemat-Nasser's estimation (1993) of the average perturbed strain in a general double-inhomogeneity to (14b) yields

$$\overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega_1} = \mathbf{S}^{\Omega_1}\overline{\boldsymbol{\eta}}^{*(1)}_{\Omega_1} + \left(\mathbf{S}^{\Sigma_1} - \mathbf{S}^{\Omega_1}\right)\overline{\boldsymbol{\xi}}^{*(1)}_{\Psi_1} + \frac{1}{\Omega_1}\int_{\Omega_1}\int_{\Omega_2}\boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\eta}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x}$$
$$+\frac{1}{\Omega_1}\int_{\Omega_1}\int_{\Psi_2}\boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\boldsymbol{\xi}^{*(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x}. \tag{15b}$$

Since Ω_1 and Σ_1 are both spherical inclusions, $\mathbf{S}^{\Sigma_1} = \mathbf{S}^{\Omega_1} = \mathbf{S}$. As an approximation, the eigenstrain fields $\overline{\boldsymbol{\eta}}^{*(2)}$ and $\boldsymbol{\xi}^{*(2)}$ in the integrands in (15a) and (15b) are replaced by their mean values over the corresponding regions Ω_2 and Ψ_2. This assumption is equivalent to keeping the first term and neglecting the higher order moments resulted from a Taylor expansion of $\boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')$ about $\mathbf{x}$ (See Ju and Chen, 1994b). Obviously the validity of this assumption decreases as the inclusions concentration and the thickness of coating layers increase. Nevertheless it will be shown in Section 6 that the results based on this hypothesis for the overall shear moduli even for thickly coated particle systems lie between the relatively narrow bounds proposed by Qiu and Weng (1991) and for overall bulk moduli coincide on their exact evaluation. For the uniform far field stimuli the volume averages of corresponding strains and eigenstrains are equivalent in both double inclusions. Therefore, by means of the decomposition (13), Eqs. (15a) and (15b) can be written as

$$f\overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega} + (1-f)\overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Psi} = \mathbf{S}(f\overline{\boldsymbol{\eta}}^{*}_{\Omega} + (1-f)\overline{\boldsymbol{\xi}}^{*}_{\Psi}) + \mathbf{T}(\Sigma_1,\Omega_2)(\overline{\boldsymbol{\eta}}^{*}_{\Omega} - \overline{\boldsymbol{\xi}}^{*}_{\Psi}) + \mathbf{T}(\Sigma_1,\Sigma_2)\overline{\boldsymbol{\eta}}^{*}_{\Omega}, \tag{16a}$$

$$\overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega}(\mathbf{x}) = \mathbf{S}\overline{\boldsymbol{\eta}}^{*}_{\Omega} + \mathbf{T}(\Omega_1,\Omega_2)\overline{\boldsymbol{\eta}}^{*}_{\Omega} + (\mathbf{T}(\Omega_1,\Sigma_2) - \mathbf{T}(\Omega_1,\Omega_2))\overline{\boldsymbol{\xi}}^{*}_{\Psi}, \tag{16b}$$

where $\mathbf{T}(\alpha,\beta) = \frac{1}{\alpha}\int_{\alpha}\int_{\beta}\boldsymbol{\Gamma}(\mathbf{x}-\mathbf{x}')\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x}$ is a fourth order tensor accounting for the interaction of inclusion α on β belonging to different double-inclusions. The explicit forms of these interaction tensors for the case of two spherical inclusions embedded in an isotropic infinite medium are obtained by Berveiller et al. (1987). For completeness, their results are summarized in Appendix A.

Eqs. (16a) and (16b) provide two sets of linear relations between the unknown average strain fields $\overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega}$ and $\overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Psi}$, and the unknown average eigenstrain fields $\overline{\boldsymbol{\eta}}^{*}_{\Omega}$ and $\overline{\boldsymbol{\xi}}^{*}_{\Psi}$. Taking volume average of the equivalency conditions (8) over regions Ω and Ψ gives

$$\begin{aligned}\mathbf{C}^1(\boldsymbol{\varepsilon}^0 + \overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega}) &= \mathbf{C}(\boldsymbol{\varepsilon}^0 + \overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Omega} - \overline{\boldsymbol{\eta}}^{*}_{\Omega}),\\ \mathbf{C}^2(\boldsymbol{\varepsilon}^0 + \overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Psi}) &= \mathbf{C}(\boldsymbol{\varepsilon}^0 + \overline{\boldsymbol{\varepsilon}}^{\mathrm{d}}_{\Psi} - \overline{\boldsymbol{\xi}}^{*}_{\Psi}).\end{aligned} \tag{17}$$

Average consistency conditions (17) along with relations (16a) and (16b) form a set of algebraic equations, which provide the unknown mean eigenstrain fields $\overline{\boldsymbol{\eta}}^{*}_{\Omega}$ and $\overline{\boldsymbol{\xi}}^{*}_{\Psi}$.

4. Average fields for a random distribution of coated fillers

In order to evaluate the ensemble volume average of quantity $\varphi(\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_N)$ within a composite containing a statistically homogeneous dispersion of identical particles, it is sufficient to compute the average of that quantity over a representative particle in the system. Let us consider the ith particle as the representative one. The ensemble volume average of quantity φ results as (Eq. (6))

$$\langle\varphi\rangle_i = \int\overset{N}{\cdots}\int \varphi p(\mathbf{x}_1,\mathbf{x}_2,\overset{N}{\cdots},\mathbf{x}_N|\mathbf{x}_i)\mathrm{d}\tau_1\overset{N}{\cdots}\mathrm{d}\tau_N. \tag{18}$$

If, as an approximation, the higher order conditional probability densities appearing in (1a) are ignored, then we obtain

$$\langle\varphi\rangle_i = \iiint_{V-E}\varphi(\mathbf{x}_i,\mathbf{x})p(\mathbf{x}|\mathbf{x}_i)\mathrm{d}\mathbf{x}, \tag{19}$$

where E is an excluded volume around the representative particle in the mesodomain V, by condition (3). According to Eq. (19), in order to obtain the ensemble average of φ within the context of pair-wise inclusion interaction, one has to integrate $\varphi(\mathbf{x}_i, \mathbf{x})$ over all possible locations $\mathbf{x}$ for given $\mathbf{x}_i$. This binary inclusion approximation was first used by Ju and coauthors (Ju and Chen, 1994a,b; Ju and Zhang, 1998) to obtain the overall mechanical behavior of a composite system.

Let us assume a statistically homogeneous composite medium that contains randomly dispersed coated spherical particles as described in the previous section. For a RVE V, we defined

$$\boldsymbol{\eta}^{*d}_{\Omega} = \overline{\boldsymbol{\eta}}^{*}_{\Omega} - \overline{\boldsymbol{\eta}}^{*0}_{\Omega}, \quad \boldsymbol{\xi}^{*d}_{\Psi} = \overline{\boldsymbol{\xi}}^{*}_{\Psi} - \overline{\boldsymbol{\xi}}^{*0}_{\Psi}, \tag{20}$$

where $\overline{\boldsymbol{\eta}}^{*0}_{\Omega}$ and $\overline{\boldsymbol{\xi}}^{*0}_{\Psi}$ are the mean eigenstrain introduced in the core and coating layer of a single double-inclusion system located in an infinite isotropic matrix.

For any arbitrary point $\mathbf{x}$ in V,

$$\begin{aligned} \boldsymbol{\eta}^{*d}_{\Omega}(\mathbf{x}_i, \mathbf{x}) &= \sum_{j=1}^{N} \{1 - \vartheta(\mathbf{x}, \mathbf{x}_j)\} \boldsymbol{\eta}^{*d}_{\Omega}(\mathbf{x}_i, \mathbf{x}_j), \\ \boldsymbol{\xi}^{*d}_{\Psi}(\mathbf{x}_i, \mathbf{x}) &= \sum_{j=1}^{N} \{1 - \vartheta(\mathbf{x}, \mathbf{x}_j)\} \boldsymbol{\xi}^{*d}_{\Psi}(\mathbf{x}_i, \mathbf{x}_j), \end{aligned} \tag{21}$$

where $\vartheta(\mathbf{x}, \mathbf{x}_j)$ is the window function defined by

$$\begin{aligned} \vartheta(\mathbf{x}, \mathbf{x}_j) &= 0, \quad \text{if } \mathbf{x} \text{ lies within the } j\text{th coated sphere}, \\ &= 1, \quad \text{if } \mathbf{x} \text{ lies outside the } j\text{th coated sphere}. \end{aligned}$$

Substitution of (3) and (21) into (19) leads to the ensemble volume average of quantities $\boldsymbol{\eta}^{*d}_{\Omega}$ and $\boldsymbol{\xi}^{*d}_{\Psi}$ as

$$\begin{aligned} \langle \boldsymbol{\eta}^{*d}_{\Omega} \rangle &= \frac{N}{V} \iiint_{V-E} \boldsymbol{\eta}^{*d}_{\Omega}(\mathbf{x}_i, \mathbf{x}) g(\mathbf{x}_i - \mathbf{x}) \mathrm{d}\mathbf{x}, \\ \langle \boldsymbol{\xi}^{*d}_{\Psi} \rangle &= \frac{N}{V} \iiint_{V-E} \boldsymbol{\xi}^{*d}_{\Psi}(\mathbf{x}_i, \mathbf{x}) g(\mathbf{x}_i - \mathbf{x}) \mathrm{d}\mathbf{x}. \end{aligned} \tag{22}$$

E here is the excluded volume around each coated particle within a sphere of radius $2R_{\Sigma}$, where R_{Σ} is the radius of the spherical domain $\Sigma = \Omega \cup \Psi$. Finally owing to relation (20), the ensemble average of eigenstrain fields becomes

$$\langle \overline{\boldsymbol{\eta}}^{*}_{\Omega} \rangle = \overline{\boldsymbol{\eta}}^{*0}_{\Omega} + \langle \boldsymbol{\eta}^{*d}_{\Omega} \rangle, \quad \left\langle \overline{\boldsymbol{\xi}}^{*}_{\Psi} \right\rangle = \overline{\boldsymbol{\xi}}^{*0}_{\Psi} + \langle \boldsymbol{\xi}^{*d}_{\Psi} \rangle. \tag{23}$$

5. Overall elastic moduli

The effective stiffness tensor $\overline{\mathbf{C}}$ of a composite material, is a constant relating the ensemble volume averages of strain and stress over the representative mesodomain of the body,

$$\langle \overline{\boldsymbol{\sigma}} \rangle = \overline{\mathbf{C}} \langle \overline{\boldsymbol{\varepsilon}} \rangle. \tag{24}$$

In accordance with Ju and Chen (1994a,b) and dropping the angular brackets denoting the ensemble average operators,

$$\begin{aligned} \overline{\boldsymbol{\sigma}} &= \mathbf{C}(\overline{\boldsymbol{\varepsilon}} - \phi \overline{\boldsymbol{\varepsilon}}^{*}) = \overline{\mathbf{C}} \overline{\boldsymbol{\varepsilon}}, \\ \overline{\boldsymbol{\varepsilon}} &= \boldsymbol{\varepsilon}^{0} + \phi \mathbf{s} \overline{\boldsymbol{\varepsilon}}^{*}, \end{aligned} \tag{25}$$

where $\overline{\boldsymbol{\varepsilon}}^* = f\overline{\boldsymbol{\eta}}^*_\Omega + (1-f)\overline{\boldsymbol{\xi}}^*_\Psi$ and ϕ is the volume fraction of double-inclusions (particles with coating) in the medium. **s** in (25) denotes the "depolarization factor" tensor at the two-point level (Sen and Torquato, 1989). Here, since all coated inclusions are spherical, **s** is equivalent to Eshelby tensor for internal points of a spherical inclusion ($\mathbf{s} = \mathbf{S}$).

The composites with statistically homogeneous distribution of random structure, exhibit isotropic behavior in the macroscopic sense. Here the bulk and shear modulus are selected as the independent material properties of the medium. To find the overall bulk modulus by (25), a far field hydrostatic loading $\varepsilon^0_{11} = \varepsilon^0_{22} = \varepsilon^0_{33}$ is applied on the system. Substitution of the ensemble averages of eigenstrian fields from (23) into (25) leads to the following relations for the macroscopic bulk modulus $\overline{\kappa}$ of the system

$$\overline{\kappa} = \frac{\kappa\left(\overline{\varepsilon}_{11} - \frac{c}{f}\overline{\varepsilon}^*_{11}\right)}{\overline{\varepsilon}_{11}}, \tag{26}$$

where

$$\overline{\varepsilon}_{11} = \varepsilon^0_{11} + c\left(\overline{\eta}^*_{\Omega_{11}} + \frac{1-f}{f}\overline{\xi}^*_{\Psi_{11}}\right)(S_{1111} + S_{1122} + S_{1133}). \tag{27}$$

c shows the volume fraction of the core spherical particle (i.e. $c = f\phi$). Similarly, for determination of the shear modulus $\overline{\mu}$, applying a simple shear strain, ε^0_{12}, gives

$$\overline{\mu} = \frac{\mu\left(\overline{\varepsilon}_{12} - \frac{c}{f}\overline{\varepsilon}^*_{12}\right)}{\overline{\varepsilon}_{12}}, \tag{28}$$

where

$$\overline{\varepsilon}_{12} = \varepsilon^0_{12} + 2c\left(\overline{\eta}^*_{\Omega_{12}} + \frac{1-f}{f}\overline{\xi}^*_{\Psi_{12}}\right)S_{1212}. \tag{29}$$

6. Numerical study

The accuracy of the method proposed here is examined by comparing its predictions of the overall elastic moduli with those predicted by other methods proposed in the literature. The work of Qiu and Weng (1991) on thickly coated particulate composites, and that of Hori and Nemat-Nasser (1993, 1994) on the multi-inclusion model are taken as reference. Qiu and Weng (1991) derived an exact solution for the effective bulk modulus of the thickly coated concentric sphere assemblage. They obtained relatively tight bounds, compared to the Hashin and Shtrikman (1963) and Walpole (1969) bounds, for the overall shear modulus of the system. The multi-inclusion model of Hori and Nemat-Nasser (1993) may be reduced to either the Mori–Tanaka or self-consistent method. In the present work, the Mori–Tanaka scheme is used which assumes that the elasticity of the infinite medium is set equal to that of matrix material.

In the first example two case studies are considered. In the first one, the shear modulus of fiber, coating and matrix are assumed to be 25, 5 and 1 GPa (Fig. 3), while in the second they are 1, 5 and 25 GPa, respectively (Fig. 4). In both cases the filler volume is taken to be half that of the coating volume and the Poisson's ratio is set to 0.3 in all phases. The estimation of the macroscopic bulk modulus vs. the volume fraction of core spherical particle (c) is shown in Figs. 3(a) and 4(a). The agreement between the results of the present study and that of the multi-inclusion method with the exact results of Qiu and Weng (1991) is good. Note that when c is equal to 0.3 the system is almost packed. The comparison of the predicted overall shear modulus with Qiu and Weng's bounds is shown in Figs. 3(b) and 4(b). The results of the present study are within the bounds whereas the prediction of the multi-inclusion method falls beyond. For generating

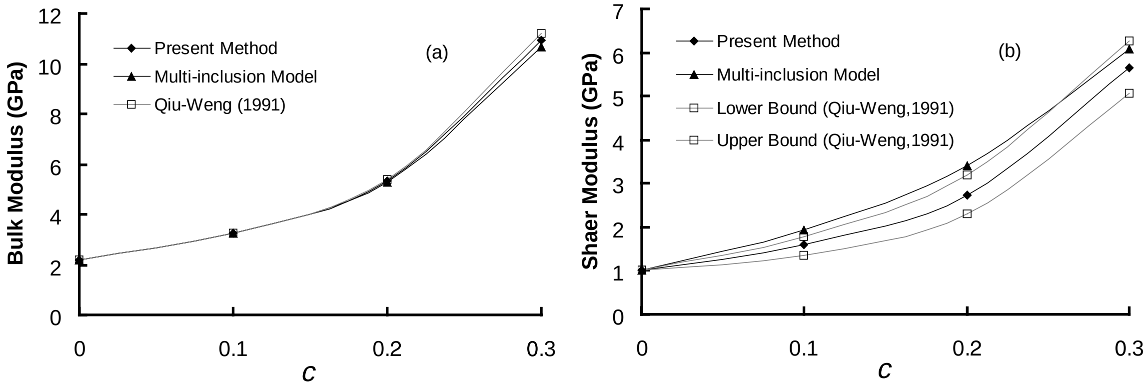

Fig. 3. Comparison between the results of the present method and the prediction of the multi-inclusion model (Hori and Nemat-Nasser, 1993; Qiu and Weng, 1991) for (a) overall bulk modulus and (b) overall shear modulus. The shear modulus of filler, coating and matrix is 25, 5 and 1 GPa, respectively. c is the volume fraction of core spherical particles.

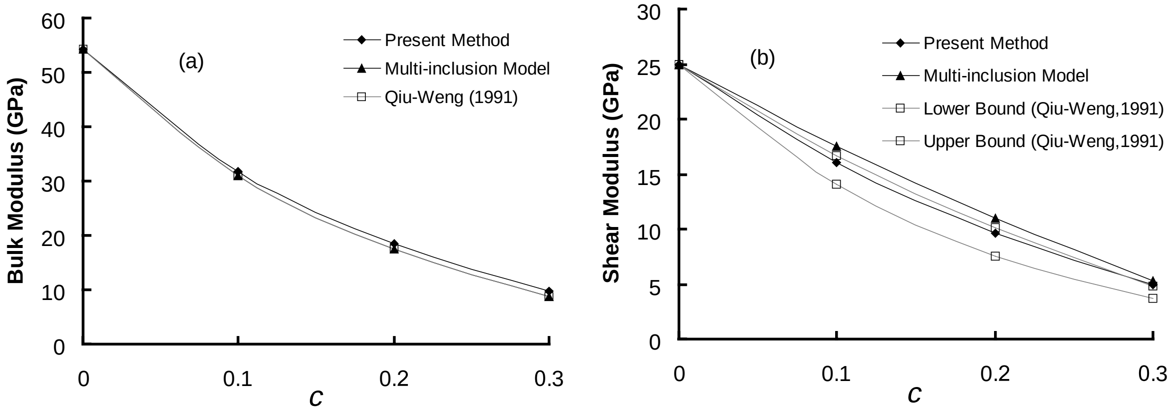

Fig. 4. Comparison between the results of the present method and the prediction of the multi-inclusion model (Hori and Nemat-Nasser, 1993; Qiu and Weng, 1991) for (a) overall bulk modulus and (b) overall shear modulus. The shear modulus of filler, coating and matrix is 1, 5 and 25 GPa, respectively. c is the volume fraction of core spherical particles.

the numerical results of the method presented in this paper in all of discussed examples in this section, the pair distribution function of Eq. (5) has been used.

In another example, the effect of the coating stiffness on the overall mechanical properties of thin coated composites is studied. Fig. 5(a) and (b) show this effect on the overall bulk modulus and shear modulus vs. filler volume fraction for different coating layer stiffness. The shear modulus of filler and matrix is assumed to be 20 and 1 GPa, respectively. In the figures μ_c/μ_m represents the shear modulus ratio of the coating to the matrix. The calculation is performed for three cases $\mu_c/\mu_m = 1$, 5, and 10, where the first case corresponds to no coating layer. Here the normalized thickness of the coating is $t_c/r_f = 0.1$, where t_c and r_f represent the thickness of the coating and radius of the fillers, respectively. It is seen that that the effective properties are sensitive to the stiffness of the coating, especially at high volume fractions.

The last example addresses the effect of the coating thickness (Fig. 6(a, b)). The shear modulus of filler, coating and matrix is chosen to be 20, 10 and 1 GPa, respectively. Numerical results are shown for three

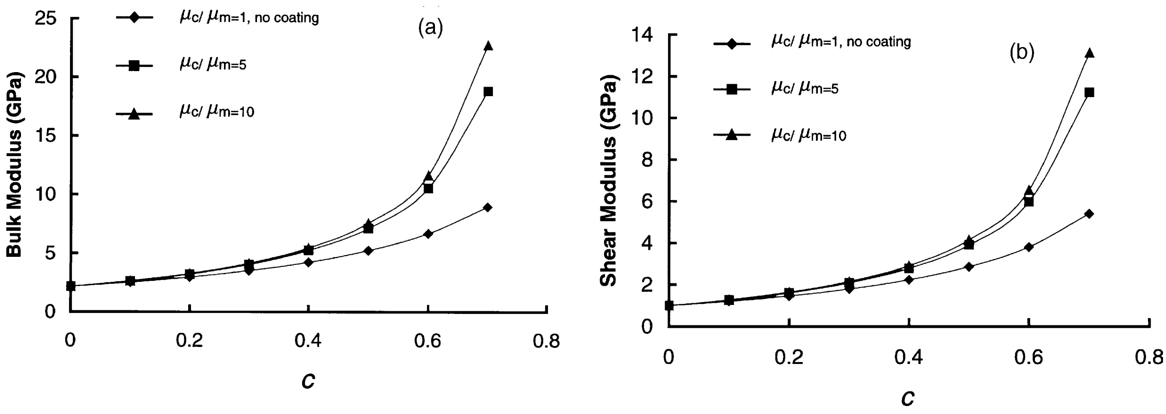

Fig. 5. Effect of the coating stiffness on (a) overall bulk modulus and (b) overall shear modulus of the composite material. c is the volume fraction of core spherical particles. The normalized thickness of the coating is $t_c/r_f = 0.1$.

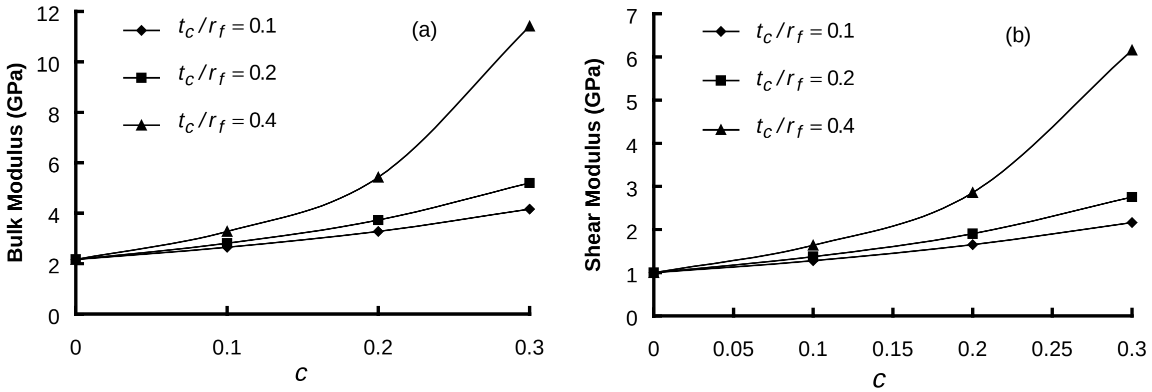

Fig. 6. Effect of the coating thickness on (a) overall bulk modulus and (b) overall shear modulus of the composite material. c is the volume fraction of core spherical particles. $\mu_f/\mu_c/\mu_m = 20/10/1$ GPa.

normalized coating thicknesses, $t_c/r_f = 0.1$, 0.2, and 0.4. It appears that the coating thickness has also a strong effect on the macroscopic properties of the composite material.

7. Conclusion

In this paper a method for the evaluation of the effective elastic moduli of composites with random spherical coated fillers is presented. It is shown that Eshelby's equivalent inclusion method is still applicable in this case for the prediction of the macroscopic elastic properties. The numerical study illustrated that even at high volume fraction of thickly coated composites, the results of the present method almost coincide with Qiu and Weng's (1991) closed form solution for the overall bulk modulus and lies between their proposed bounds for the overall shear modulus. Some other numerical studies are performed to show the effect of coating stiffness and thickness.

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Appendix A

The explicit form of the interaction tensor $\mathbf{T}(\alpha,\beta)$ is presented here. Let α and β be the volume of two spheres, v, the Poisson ratio, and μ, the shear modulus of the isotropic matrix. The radius of spherical fillers α and β are denoted by a and b, and R shows the distance between their centers. The non-zero components of $T_{ijkl}(\alpha,\beta)$ are

$$T_{ijkl}(\alpha,\beta) = Q_{ijmn}(\alpha,\beta)C_{mnkl},$$

where $\mathbf{C}$ is the stiffness of the matrix and

$$Q_{1111}(\alpha,\beta) = Q_{2222}(\alpha,\beta) = \frac{\beta}{16\pi R^2}\frac{1}{\mu(1-v)}\left(1-4v+\frac{5}{9}\rho^2\right),$$

$$Q_{1122}(\alpha,\beta) = Q_{2211}(\alpha,\beta) = \frac{\beta}{16\pi R^2}\frac{1}{\mu(1-v)}\left(-1+\frac{3}{5}\rho^2\right),$$

$$Q_{1133}(\alpha,\beta) = Q_{2233}(\alpha,\beta) = Q_{3311}(\alpha,\beta) = Q_{3322}(\alpha,\beta) = \frac{\beta}{16\pi R^2}\frac{1}{\mu(1-v)}\left(2-\frac{12}{5}\rho^2\right),$$

$$Q_{1212}(\alpha,\beta) = Q_{1221}(\alpha,\beta) = Q_{2121}(\alpha,\beta) = Q_{2112}(\alpha,\beta) = \frac{\beta}{16\pi R^2}\frac{1}{\mu(1-v)}\left(1-2v+\frac{3}{5}\rho^2\right),$$

$$Q_{1313}(\alpha,\beta) = Q_{1331}(\alpha,\beta) = Q_{3113}(\alpha,\beta) = Q_{3131}(\alpha,\beta) = \frac{\beta}{16\pi R^2}\frac{1}{\mu(1-v)}\left(1+4v-\frac{12}{5}\rho^2\right),$$

$$Q_{2323}(\alpha,\beta) = Q_{2332}(\alpha,\beta) = Q_{3223}(\alpha,\beta) = Q_{3232}(\alpha,\beta) = \frac{\beta}{16\pi R^2}\frac{1}{\mu(1-v)}\left(1+v-\frac{12}{5}\rho^2\right),$$

$$Q_{3333}(\alpha,\beta) = \frac{\beta}{16\pi R^2}\frac{1}{\mu(1-v)}\left(-8+8v+\frac{24}{5}\rho^2\right),$$

and $\rho^2 = a^2 + b^2/R^2$. Note that $T_{ijkl}(\alpha,\beta) = 0$ when three indices are different, or when three indices are equal, but different from the fourth.

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