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A micromechanical model for interpenetrating multiphase composites

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Abstract

The dependence relation between the macroscopic effective property and the microstructure of interpenetrating multiphase composites is investigated in this paper. The effective elastic moduli of such composites cannot be calculated from conventional micromechanics methods based on Eshelby's tensor because an interpenetrating phase cannot be extracted as dispersed inclusions. Employing the concept of connectivity, a micromechanical cell model is presented for estimating the effective elastic moduli of composites reinforced with either dispersed inclusions or interpenetrating networks. The model includes the main features of stress transfer of interpenetrating microstructures. The Mori–Tanaka method and the iso-stress and iso-strain assumptions are adopted in an appropriate manner of combination, rendering the calculation of effective moduli quite easy and accurate.

Keywords: Interpenetrating phase composite; Microstructure; Effective properties; Micromechanics; Finite element method; Connectivity

1. Introduction

Recently, considerable attention has been directed toward interpenetrating or co-continuous phase composites. In contrast with most conventional composites in which only the matrix phase is spatially continuous, an interpenetrating phase composite has at least two phases that are each interconnected in three dimensions and construct a topologically continuous network throughout the microstructure. This new generation of composites possesses some physical and mechanical properties that are evidently different from and often superior to conventional fiber- or particle-reinforced composites [1–4]. The determination of the overall effective properties of interpenetrating multiphase composites is of great interest in both their engineering applications and theoretical analysis, but little work has been done as yet on this subject.

Micromechanical analysis of heterogeneous materials provides their overall behavior from the known properties of individual constituents. Various estimation schemes (e.g., non-interacting or dilute concentration method, self-consistent method, and Mori–Tanaka method) have been

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proposed to calculate the effective properties (e.g., elastic moduli, thermal conductivity, electrical conductivity, dielectric constants, piezoelectric coefficients, and magnetic permeability) of heterogeneous materials [5,6]. However, most of the previous work has been conducted for those materials comprised of well-defined inclusions (e.g., spheres, whiskers, flakes, and fibers) in a connected matrix, because it is more difficult to treat interpenetrating multiphase composites.

Recently, Schmauder and coworkers [4,7,8] developed a self-consistent matricity model to simulate numerically the mechanical behavior of an isotropic two-phase composite with a coarse interpenetrating microstructure. Levassort et al. [9] presented a unit cell model to estimate the effective electromechanical moduli of an interpenetrating two-phase piezoelectric composite. Wegner and Gibson [2,3] suggested a numerical model to estimate the effective properties of interpenetrating binary composites. This model was directed mainly towards simulating the mechanical properties of isotropic materials reinforced with coarse particles in such a large volume fraction that they are interconnected as a three-dimensional network. These models are difficult to represent the complicated microstructures of interpenetrating multiphase composites, especially when the spatial distribution of individual phases and the macroscopic properties of composites are anisotropic. Using statistical correlation functions, Torquato and coworkers [6,10] derived the multipoint bounds for heterogeneous materials, which are sharper than the Voigt–Reuss (or one-point) bounds and the Hashin–Shtrikman (or two-point) bounds. Until recently, however, applications of high-point bounds are still very limited because of the prohibitive complexity involved in ascertaining the statistical correlation functions for engineering composites.

One of the key issues in estimation of the effective constitutive relation of a composite is the appropriate modeling of the stress transfer relation of the constituent phases with specific microstructures. In interpenetrating composites, the reinforcing phase can transfer stresses effectively in all directions. The present paper aims to examine the relationship between the macroscopic effective property and the microstructure of interpenetrating multiphase composites. The concept of connectivity in [11] is used to characterize the main topological and stress transfer features of the complicated microstructure of composites. A unit cell model is defined in terms of the microstructural parameters including volume fractions, connectivity, and anisotropic spatial distribution of phases. The presented micromechanics method can calculate the effective properties of composites with both interpenetrating phases and dispersed inclusions.

2. Unit cell model

2.1. Characterization of microstructures

Many natural interpenetrating phase materials in biology (e.g., bones in mammals and the trucks and limbs of many plants) have anisotropic, interpenetrating microstructures [1]. Manmade interpenetrating phase composites derived by such approaches as directed metal oxidation and colloidal methods also possess somewhat anisotropic properties. However, there is still a lack of micromechanics methods for simulating the mechanical behaviors of anisotropic interpenetrating phase composites.

Now, consider a macroscopically homogeneous n-phase composite, in which both the elastic properties and spatial arrangements of individual phases may be anisotropic. Assume that in the composite, n_1 phases (say Phases $1, 2, \ldots, n_1$) are each continuously self-connected in three dimensions, and the other n_2 phases (i.e., Phases $n_1 + 1, n_1 + 2, \ldots, n$ with $n_1 + n_2 = n$) appear as well-defined, disconnected inclusions. If a constituent material exists both in the form of continuous networks and dispersed inclusions, then it is treated as two different phases, a self-connected or interpenetrating phase and a dispersed inclusion phase. Connectivity suggested by Newnham et al. [11] is a practical concept for describing the spatial arrangement of each phase in such a composite since it gives the number of dimensions in which each component is self-connected. For example, 10 different connectivities are possible for binary composites, including 0–0, 0–1, 0–2, 0–3, 1–1, 1–2, 1–3, 2–2, 2–3, and 3–3. The connectivity characterizing the microstructural feature of the above defined n-phase composite is designated as 3_1–3_2–$\cdots$–3_{n_1-1}–3_{n_1}–0_{n_1+1}–0_{n_1+2}–$\cdots$–0_n, where the subscripts stand for the corresponding phases. A phase of fiber or flake shape, with connectivity being 1 and 2, respectively, can also be incorporated easily into the present model, but is omitted here for conciseness.

Let us first specify according to the material microstructure an interpenetrating phase, say the n_1th one, as the matrix of the composite, which usually has a relatively high volume fraction. It might be assumed, for simplicity, that all the isolated inclusions of the n_2 phases are embedded in this host matrix. Then the interpenetrating matrix and all the n_2 constituent phases of inclusions are regarded as a single hybrid phase, which is continuously self-connected in three dimensions. Thus, the microstructure of the n-phase composite is simplified as an interpenetrating n_1-phase material in which all the phases are each continuously self-connected. Its connectivity can be simplified by 3_1–3_2–$\cdots$–3_{n_1-1}–3_{n_1} (3_{n_1}–0_{n_1+1}–0_{n_1+2}–$\cdots$–0_n), where the expression in the parentheses indicates the microstructure of the composite matrix phase, as defined above.

2.2. Unit cell

Refer to a Cartesian coordinate system (o–$x_1x_2x_3$). For such a multiphase composite with complex, interpenetrating microstructure, a cubic cell of unit volume, filled by the n_1 interpenetrating phases, can be specified according to the volume fractions, connectivity and spatial arrangement of the phases. Each self-connected phase is present in the unit cell as three mutually orthogonal branches with rectangular cross-sections, as shown in Fig. 1. The dimensions of the three branches can be determined according to the volume fraction and anisotropic spatial distribution of this phase. The anisotropic microstructures of an interpenetrating phase, say the αth one, is described by only three size parameters, a_α , b_α and c_α, Fig. 1. Since the cubic cell is assumed to be of unit volume, the parameters a_α, b_α and c_α are normalized or non-

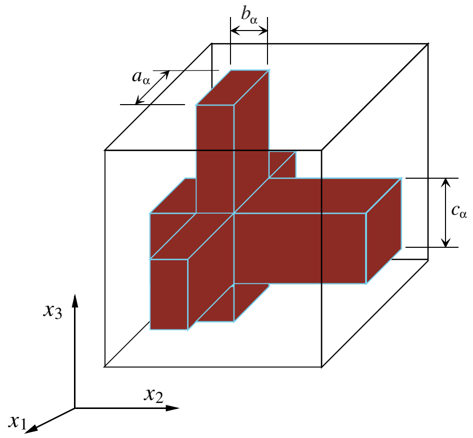

Fig. 1. A single interpenetrating phase in the unit cell.

dimensional. The interpenetrating phases $1, 2, \ldots,$ and $n_1 - 1$ are characterized by $n_1 - 1$ orthogonally branched components embedded in the host matrix. This unit cell model keeps the most significant features of microstructures and stress transfer relations in interpenetrating multiphase composites. More parameters may be introduced further in the cubic cell to characterize the microstructures more exactly, but this will certainly make the numerical computation and parameter determination cumbersome.

Mathematically, the spatial distribution of the phases can be determined from a certain number of cross-sections of different directions, provided that the composite is statistically homogeneous in the macroscopic sense. For a two-phase composite with its principal axes aligned in the x_1-, x_2- and x_3-directions, for example, a cross-section normal to one principal axis, say x_1, is shown in Fig. 2 [12]. The cross-section fractions of the reinforcing (white) phase and the (black) matrix in this cross-section can be determined by image analysis and designated as s_{1,x_1} and $s_{2,x_1} = 1 - s_{1,x_1}$, respectively, where the first subscript denotes the corresponding phase, and the second stands for the direction of the normal of the corresponding cross-section. Similarly, one can get s_{1,x_2}, $s_{2,x_2} = 1 - s_{1,x_2}$, s_{1,x_3} and $s_{2,x_3} = 1 - s_{1,x_3}$. In the special case of an isotropic composite, the cross-section fractions of phases are independent on the directions, i.e., $s_{1,x_1} = s_{1,x_2} = s_{1,x_3}$ and $s_{2,x_1} = s_{2,x_2} = s_{2,x_3}$.

Fig. 2. A cross-section of an interpenetrating two-phase composite [12].

Thus, the size parameters of the αth phase in the unit cell are related to the cross-section fractions by

$$b_\alpha c_\alpha = s_{\alpha,x_1}, \quad a_\alpha c_\alpha = s_{\alpha,x_2}, \quad a_\alpha b_\alpha = s_{\alpha,x_3}, \tag{1}$$

from which one obtains

$$a_\alpha = \left(\frac{s_{\alpha,x_2} s_{\alpha,x_3}}{s_{\alpha,x_1}}\right)^{1/2}, \quad b_\alpha = \left(\frac{s_{\alpha,x_1} s_{\alpha,x_3}}{s_{\alpha,x_2}}\right)^{1/2}, \quad c_\alpha = \left(\frac{s_{\alpha,x_1} s_{\alpha,x_2}}{s_{\alpha,x_3}}\right)^{1/2}. \tag{2}$$

The volume fraction of an interpenetrating phase is related to the three size parameters by

$$f_\alpha = a_\alpha b_\alpha + a_\alpha c_\alpha + b_\alpha c_\alpha - 2a_\alpha b_\alpha c_\alpha. \tag{3}$$

In the case of isotropic distribution, one obtains $a_\alpha = b_\alpha = c_\alpha$, and then $f_\alpha = a_\alpha^2(3 - 2a_\alpha)$.

To illustrate the method for defining a unit cell, two examples are schematized in Fig. 3, corresponding to two five-phase composites. In Fig. 3(a), three phases are self-connected in three dimensions and the other two exist in the form of dispersed inclusions, and in Fig. 3(b), four phases are self-continuous and the other is of inclusion shape. Such unit cells for multiphase composites are of engineering interest in analysis, design and characterization of the actual materials. On one hand, more than two phases are often necessary to yield better comprehensive properties of a composite. On the other hand, if a constituent material of a composite exists in different forms (network or particles), it should be considered as different phases in the theoretical model for easier calculation, as aforementioned.

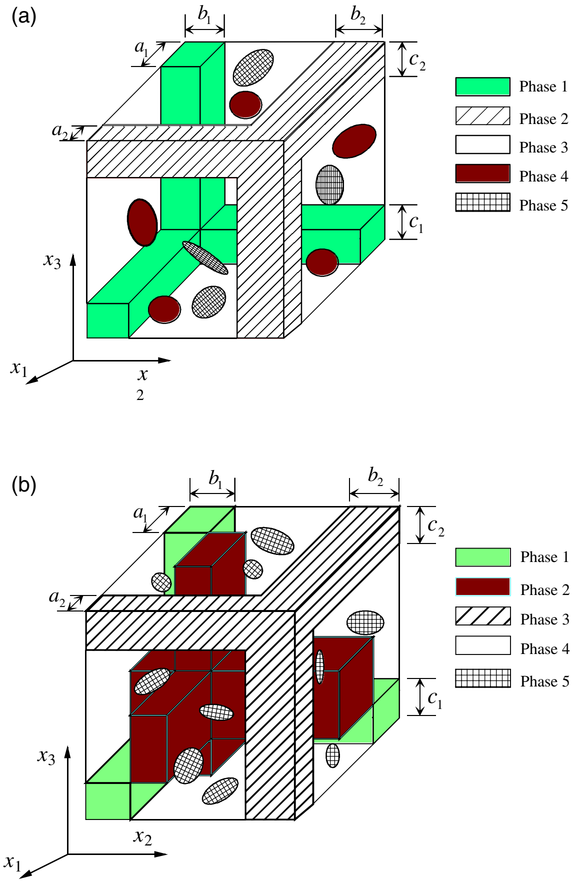

Fig. 3. Two unit cells of interpenetrating multiphase composites.

3. Estimation of effective elastic moduli

As mentioned in Section 1, composites of current interest not only have phases of dispersed inclusion but also continuously self-connected phases. To provide an analytical estimate on the effective elastic moduli of a composite with such complicated microstructures, some assumptions and simplifications are always necessary. The effective properties (for example, elastic moduli, elastoplastic constitutive relation and thermal conductivity) of such composites can be estimated from the above-defined unit cell model. We present here a method for calculating the overall effective elastic moduli via a two-step procedure.

First, the effective moduli of the above-defined hybrid matrix phase, in which a self-connected phase (the n_1th one) containing the n_2 inclusion-dispersed phases, is determined by one of the conventional micromechanics methods (e.g., dilute concentration method, self-consistent method, generalized self-consistent method, Mori–Tanaka method). These methods derive the overall behavior of heterogeneous materials from the known properties of the individual constituents (e.g., matrix and inclusions) [5]. In the present paper, only the Mori–Tanaka method [13] will be employed because it may easily derive the effective moduli of inclusion-dispersed composites with good accuracy even for a high volume fraction of inclusions. However, it is known that the Mori–Tanaka method leads to asymmetrical elastic tensors in some cases. This drawback can generally be overcome by appropriate symmetrization of the elastic stiffness or compliance tensors. Implementation of other estimation methods aforementioned into the present model is also thinkable but omitted here for short.

The Mori–Tanaka method [13] estimates the effective moduli by assuming that each inclusion is placed in an infinite pristine matrix and subjected, in the far field, to the average stress $\boldsymbol{\sigma}_{\mathrm{m}}$ or average strain $\boldsymbol{\varepsilon}_{\mathrm{m}}$ of the matrix. For a composite with an interpenetrating matrix containing n_2 reinforcing phases in the form of differently oriented inclusions, the Mori–Tanaka method gives the effective stiffness tensor $\mathbf{C}$ as [5]

$$\mathbf{C} = \left(f_{\mathrm{m}}\mathbf{C}_{\mathrm{m}} + \sum_{\alpha=1}^{n_2} f_{\mathrm{r}\alpha}\langle \mathbf{C}_{\mathrm{r}\alpha} : \mathbf{A}_\alpha \rangle \right) : \left(f_{\mathrm{m}}\mathbf{I} + \sum_{\alpha=1}^{n_2} f_{\mathrm{r}\alpha}\langle \mathbf{A}_\alpha \rangle \right)^{-1}, \tag{4}$$

where the subscripts m and r represent quantities of the hybrid matrix and the reinforcing phase, respectively, α implies the αth reinforcing phase, f_{m} and $f_{\mathrm{r}\alpha}$ denote the volume fractions, and $\mathbf{C}_{\mathrm{m}}$ and $\mathbf{C}_{\mathrm{r}\alpha}$ the elastic stiffness tensors of the corresponding phases. Throughout this paper, a boldface letter stands for a two- or four-order tensor, and a colon between two tensors denotes contraction (inner product) over two indices. The fourth-order tensor $\mathbf{A}_\alpha$, which is the average strain-concentration tensor, is defined by

$$\boldsymbol{\varepsilon}_{\mathrm{r}\alpha} = \mathbf{A}_\alpha : \boldsymbol{\varepsilon}_{\mathrm{m}}, \tag{5}$$

where $\boldsymbol{\varepsilon}_{\mathrm{r}\alpha}$ denotes the average strain in the αth reinforcing phase (inclusions). The partial concentration factor $\mathbf{A}_\alpha$ was given by Walpole [14] as

$$\mathbf{A}_\alpha = \left[\mathbf{I} + \mathbf{S}_\alpha : (\mathbf{C}_{\mathrm{m}})^{-1} : (\mathbf{C}_{\mathrm{r}\alpha} - \mathbf{C}_{\mathrm{m}}) \right]^{-1}, \tag{6}$$

where $\mathbf{S}_\alpha$ denotes Eshelby's tensor. Eqs. (4)–(6) are adopted here to determine the elastic moduli of the above-defined hybrid matrix in the unit cell.

The second step is to estimate the effective elastic moduli of the cubic cell, either by a finite element method or by an approximate analytical method. For a composite with dispersed inclusions, the iso-stress and iso-strain assumptions lead, respectively, to the lower and upper bounds of elastic moduli. These two methods were first introduced by Voigt and Reuss, and, therefore, are also referred to as the Voigt and Reuss methods, respectively. However, an appropriate combination of the iso-stress and iso-strain assumptions may yield the effective elastic moduli of the cubic cell in a manner much easier than the finite element numerical analysis.

To this end, the unit cell, which consists of n_1 self-connected phases (see Fig. 3), is divided into $n_1 \times n_1$ sub-cells, each consisting of n_1 series blocks. The effective moduli of each sub-cell are determined by adopting the iso-stress assumption. Finally, the elastic moduli of the whole cell can be calculated from the $n_1 \times n_1$ parallel sub-cells by using the iso-strain assumption.

Such a parallel-series decomposition method is schematized in Fig. 4 for an interpenetrating two-phase composite. Evidently, there are three possible directions to divide the cell. If the composite is isotropic, estimates of the effective Young's modulus and shear modulus are independent of the dividing direction. For an anisotropic composite

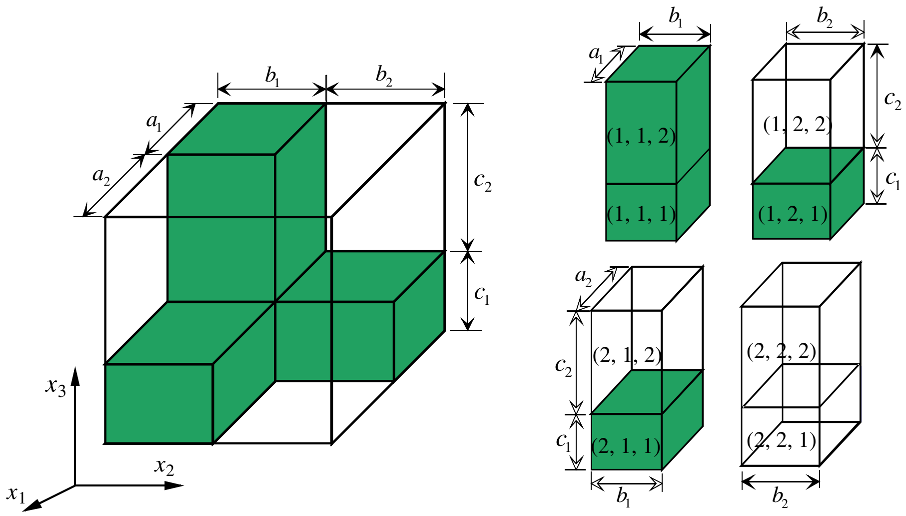

Fig. 4. (a) A unit cell for an interpenetrating two-phase composite, and (b) its decomposition.

with oriented network phases, the effective elastic moduli in different directions should be derived from the corresponding decomposition direction. In the isotropic case, for example, the effective Young's modulus can be determined by

$$E = \sum_{\alpha=1}^{n_1} \sum_{\beta=1}^{n_1} \left\{ \left[\left(\sum_{\gamma=1}^{n_1} \frac{V_{\alpha\beta\gamma}}{E_{\alpha\beta\gamma}} \right)^{-1} \sum_{\gamma=1}^{n_1} V_{\alpha\beta\gamma} \right] \times \sum_{\gamma=1}^{n_1} V_{\alpha\beta\gamma} \right\}, \tag{7}$$

where (α, β, γ) denotes the serial number of a sub-cell in the x_1-, x_2- and x_3-directions (Fig. 4(b)), $E_{\alpha\beta\gamma}$ and $V_{\alpha\beta\gamma}$ denote the Young's modulus and volume of the (α, β, γ) sub-cell.

For composites reinforced by distributed inclusions, only the matrix phase is self-connected in three dimensions. Then the unit cell is reduced to a representative volume element (RVE), which is extensively employed in the micromechanics of composites. In this simple case, therefore, the present model is completely equivalent to the conventional micromechanics models. The effective moduli of the composite can be given directly from such methods as that of Mori–Tanaka [5,13].

4. Illustrations and discussions

4.1. An interpenetrating two-phase composite

For an interpenetrating binary composite, the effective moduli can be estimated easily from the unit cubic cell in Fig. 4(a) by adopting the combination of iso-stress and iso-strain assumptions, as shown in Fig. 4(b). We take a simple case of engineering significance as an example, where both the interpenetrating phases are isotropic, linearly elastic and uniformly distributed in all directions. Here, it is seen that the geometrical parameters $a_1 = b_1 = c_1 = a$ and $a_2 = b_2 = c_2 = 1 - a$, and the volume fraction of the reinforcing phase $f_1 = 3a^2 - 2a^3$. Then, the effective Young's modulus E and shear modulus G of such a 3–3 composite are derived from Eq. (7) in the following explicit form:

$$\begin{aligned} E &= a^2 E_r + (1-a)^2 E_m + 2a(1-a)\left(\frac{a}{E_r} + \frac{1-a}{E_m}\right)^{-1}, \\ G &= a^2 G_r + (1-a)^2 G_m + 2a(1-a)\left(\frac{a}{G_r} + \frac{1-a}{G_m}\right)^{-1} \end{aligned} \tag{8}$$

which are functions merely of the volume fraction of the reinforcing phase via the relation $f_1 = 3a^2 - 2a^3$, though the effects of interpenetrating microstructures have been included.

To examine the accuracy of the above method based on the decomposition of parallel and series sub-cells, commercial FE program ABAQUS-6.2.1 was used to calculate the effective elastic moduli of the unit cell in Fig. 4(a). The periodic displacement boundary conditions are prescribed on the unit cell. A bi-continuous composite made of 420 stainless steel and 150P bronze is taken as an example. The Young's modulus and shear modulus of the stainless are 210 and 81.4 GPa, and those of the bronze are 110 and 41.35 GPa, respectively [2,3]. The analytical solution in Eq. (8), the results of the finite element method and the experimental results of Wegner and Gibson [2] are shown in Fig. 5. Evidently, the approximate analytical method based on the sub-cell decomposition agrees very well with the numerical method and the experimental results.

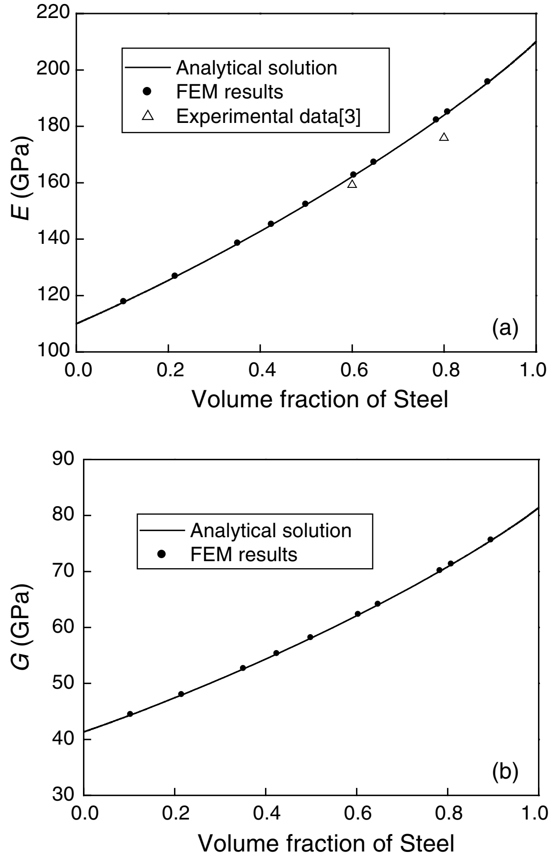

Fig. 5. Effective moduli of the co-continuous stainless steel/ bronze composite: (a) Young's modulus and (b) shear modulus.

4.2. A four-phase composite with two self-connected phases

A four-phase composite synthesized by Torquato et al. [12] is analyzed here, in which the B_4C and Al phases are self-connected in three dimensions and the AlB_2 and Al_4BC phases exist in the form of dispersed inclusions. The elastic properties and volume fractions of all the phases are given in Ref. [12] and, for completeness, are listed in Table 1.

The B_4C phase is chosen as the self-connected host matrix. According to the two-step calculation procedure, the effective moduli of the composite matrix comprising of the B_4C, AlB_2 and Al_4BC phases are first determined from the Mori–Tanaka method. Its Young's modulus and shear modulus are determined, respectively, as $E_m = 414.72$ GPa and $G_m = 176.19$ GPa. Using the relation: $f_1 = 3a^2 - 2a^3$ and the volume fraction of Al, $f_1 = 0.16$, the parameter a equals to 0.2533. Then, the effective bulk modulus and shear modulus are derived from Eq. (8) as $K = 169.9$ GPa and

Table 1
Volume fractions and elastic moduli of phases in a four-phase composite [12]

Phase number, α	Phase material	Volume fraction, f_α	Bulk modulus, K_α (GPa)	Shear modulus, G_α (GPa)
1	Al	0.16	67.6	25.9
2	B_4C	0.66	226.0	192.0
3	AlB_2	0.02	170.0	120.0
4	Al_4BC	0.16	175.0	129.0

$G = 126.8$ GPa, which agree well with the experimental data of Torquato et al. [12], $K = 176$ GPa and $G = 125$ GPa.

5. Conclusions

The mechanical properties, for example, stiffness, strength, and fracture toughness, of composites depend not only on the volume fractions but also, to varying extents, on the spatial distribution of the constituents. The effective elastic moduli of interpenetrating phase composites cannot be obtained from the Eshelby's tensor, which has provided a sound physical basis for the conventional micromechanics of composites, because an interpenetrating phase cannot be extracted as dispersed inclusions. By using the concept of connectivity, a unit cell model is presented in this paper to determine the overall effective properties of composites reinforced with either dispersed inclusions or interpenetrating networks. The Mori–Tanaka method together with the iso-stress and iso-strain assumptions are employed and combined in an appropriate manner, which enables an easy and accurate determination of the effective elastic moduli. Although the attention of this work is focused mainly on the effective elastic moduli, the presented cell model can also be extended easily to evaluate the three-dimensional elastoplastic constitutive relation and other effective properties of composites.

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