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Analytical effective elastic properties of particulate composites with soft interfaces around anisotropic particles

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ABSTRACT

Understanding the effects of interfacial properties on effective elastic properties is of great importance in materials science and engineering. In this work, we propose a theoretical framework to predict the effective moduli of three-phase heterogeneous particulate composites containing spheroidal particles, soft interfaces, and a homogeneous matrix. We first derive the effective moduli of two-phase representative volume elements (RVEs) with matrix and spheroidal inclusions using the variational principle. Subsequently, an analytical model considering the volume fraction of soft interfaces around spheroidal particles is presented. The effective moduli of such three-phase particulate composites are eventually derived by the generalized self-consistent scheme. These theoretical schemes are compared with experimental studies, numerical simulations, and theoretical approximations reported in the literature to verify their validity. We further investigate the dependence of the effective elastic modulus on the interfacial properties and the geometric characteristics of anisotropic particles based on the proposed theoretical framework. Results show that the interfacial volume fraction and the effective elastic modulus of particulate composites are strongly dependent on the aspect ratio, geometric size factor, volume fraction, and particle size distribution of ellipsoidal particles.

1. Introduction

Interfaces interacted by anisotropic particles are crucial components in a variety of particulate materials like polymer, colloidal, ceramic, and cementitious composites [1–4]. Understanding the effects of interfacial characteristics on effective elastic properties as average features by homogenization that well reflect the macroscopic mechanical responses of particulate composites, is a problem of great interest in materials research & development [3–7]. Specifically, the estimation of effective moduli of particulate composites is of prime importance to better capture the behaviors of composites and to evaluate the success of their design. In the present work, we focus on spheroids as shape-anisotropic particles over a broad range of aspect ratios with widespread applications in specific materials [1,4–6].

It has been experimentally observed by several imaging equipment that interfaces as a weak link have a complex network that adjacent interfaces possess an overlap potential in some particulate composites, such as cementitious, ceramic, and colloidal composites, where the formation of interfaces normally attributes to the packing of discrete grains against aggregate or wall surfaces, namely, the so-called "wall" effect [8–10]. This also gives rise to the physical natures of a relative high porosity and low rigidity for interfaces around aggregates. As such, interfaces are usually viewed as a compliant interphase (i.e., soft interfaces) between inclusions and matrix [2,4,7–9], as well as particulate composites as a three-phase composite structure consist of inclusions, soft interfaces, and matrix.

Over the past decades, the researches for effective elastic properties of three-phase particulate composites have attracted much attention, especially for the three-phase composites containing interfaces. The pioneering work was from Christensen and Lo [11] that applied the generalized self-consistent scheme [12] to study the effective shear modulus of three-phase composites with

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spherical inclusions. Thereafter, many seminal empirical and theoretical formulae have been proposed to predict effective moduli of three-phase composites, such as bounds models [13,14], Mori-Tanaka scheme [15], differential effective medium approximation [16], generalized self-consistent scheme [12,17], and series expansions [18], and other effective medium methods. Fu et al. [19] and Wang and Pan [20] have well summarized the existing studies in this area, interested readers may refer to the two reviews. Also, three kinds of interfacial model are often used to simulate the properties of interfaces in those effective medium methods: the linear-spring model, interface stress model, and interphase model [7]. The first two models assume interfaces occupying a zero volume in composites that is essentially a two-phase composite structure, whereas, the third one is a three-phase model, composed of inclusions, interphase, and matrix, similar to the present case. From view of the abovementioned micromechanics schemes, the estimation of effective moduli of composites requires knowledge of the volume fraction and elastic properties of individual phases [11–20]. Therefore, as an important microstructural characteristic, the volume fraction of interphase should be considered to investigate the effective elastic properties of three-phase composites. As demonstrated by Torquato [5], the more microstructural characteristics of composite media are explored, the more accurate their effective properties can be estimated. It is worth mentioning that Garboczi and Bentz [21] presented a theoretical approximation for the volume fraction of soft interfaces around spherical particles, and the theoretical model was further employed to predict the effective conductivity of cementitious composites. Although such the outstanding contribution may provide guidance for the effective elastic properties of particulate composites, isotropic spheres cannot reflect the anisotropy nature of particles in particulate composites. Also, the volume fraction of soft interfaces around anisotropic particles has received relatively little attention in terms of theoretical modeling until fairly recently [4,22–24]. Moreover, it is quite challenging to evaluate the effect of such the interfacial property on the elastic moduli of particulate composites with anisotropic ellipsoidal particles and soft interfaces.

In the present study, heterogeneous particulate composites consist of a homogeneous matrix, anisotropic spheroidal particles, and soft interfaces. Perfect bonding conditions are assumed to prevail at both the particle/interface and the interface/matrix. We attempt to develop a theoretical framework to predict the effective moduli of particulate composites, in order to provide an efficient tool for their design. We first demonstrate a theoretical scheme for predicting the effective moduli of two-phase composites with spheroidal inclusions. We give an analytical approximation for the volume fraction of soft interfaces around ellipsoidal particles. Subsequently, by the generalized self-consistent scheme, the prediction of the effective moduli of heterogeneous three-phase particulate composites is explicitly presented in details. The rest of this article is organized as follows. Section 2 demonstrates the effective moduli of two-phase composites. Section 3 presents the volume fraction of soft interfaces around ellipsoidal particles. In Section 4, the effective moduli of three-phase composites are proposed. Subsequently, the theoretical results are given and discussed in Section 5. Finally, this article is completed with some concluding remarks in Section 6.

2. Effective moduli of two-phase composites

As mentioned above, several attempts have been made for theoretically investigating the effective moduli of particulate composites. It is worth mentioning that Wills and co-workers [25,26] applied a generalized variational principle to propose estimates of the Hashin-Shtrikman (HS) type for composites with ellipsoidal inclusions and considering their spatial distribution configurations. In that approximation, the RVE of an ergodic M-phase heterogeneous composite consists of M-1 types of ellipsoidal inclusions, distributed in a homogeneous matrix (denoted as phase 1, with modulus tensor $\mathbf{E}_1$). It is assumed that there are m_r inclusions of type r ($r = 2, 3, \ldots, M$), with modulus tensor $\mathbf{E}_r$. The basic derivation of effective moduli is illustrated in Supplementary Information (see S1). In that framework, a suitable choice for the comparison material is the matrix material itself, i.e., $\mathbf{E}_0 = \mathbf{E}_1$, so that the polarization field vanishes exactly in the matrix phase and the microstructural tensor $\langle \mathbf{A}_{rs} \rangle$ for this kind of RVE can be expressed by

$$\langle \mathbf{A}_{rs} \rangle = f_r(\delta_{rs}\mathbf{L}_{ir} - f_s\mathbf{L}_{drs}), (r, s = 2, \ldots, M) \tag{1}$$

where f_r is the volume fraction of inclusions of type r, tensors $\mathbf{L}_{ir}$ and $\mathbf{L}_{drs}$ are associated with the inclusion and distribution shape tensors $\mathbf{Z}_{ir}$ and $\mathbf{Z}_{drs}$, respectively. $\mathbf{L}_{ir}$ is defined as [27].

$$\mathbf{L}_{ir} = \frac{1}{4\pi|\mathbf{Z}_{ir}|}\int\limits_{|\xi|=1} \mathbf{C}_0(\xi)\left|\mathbf{Z}_{ir}^{-1}\cdot\xi\right|^{-3}\mathrm{d}S(\xi) \tag{2}$$

where the integration is operated over the unit sphere $|\xi| = 1$, $C_{0mn} = 1/E_{1mn}\xi_m\xi_n$, and ξ_m is one of components of the unit vector. Similarly, $\mathbf{L}_{drs}$ can also be expressed by an analogous to Eq. (2) with $\mathbf{Z}_{ir}$ replaced by $\mathbf{Z}_{drs}$. The inverses of the eigenvalues of $\mathbf{Z}_{ir}$ and $\mathbf{Z}_{drs}$ are the semi-axes of the ellipsoidal inclusion and the distribution ellipsoid, respectively. If the distribution of inclusions is the same, i.e., $\mathbf{L}_{drs} = \mathbf{L}_d$, $\mathbf{L}_d$ and $\mathbf{L}_{ir}$ can be characterized by Eshelby tensors $\mathbf{H}_V$ and $\mathbf{H}_r$, which represent the geometric factor tensors of the distribution ellipsoid and the ellipsoidal inclusions of type r correspondingly. Herein, we follow the works of Torquato [5] and Duan et al. [27], where the 2 s-order tensors are displayed by the elliptical integrals

$$\mathbf{H}_j = \frac{a_{1j}a_{2j}a_{3j}}{4\pi}\int\limits_{|\xi|=1}\frac{\mathbf{C}_0(\xi)\cdot\mathbf{E}_1}{\Upsilon^3}\mathrm{d}S(\xi), (j = r, V) \tag{3}$$

with

$$\Upsilon^2 = a_{1j}^2\xi_1^2 + a_{2j}^2\xi_2^2 + a_{3j}^2\xi_3^2$$

where a_{1j}, a_{2j}, and a_{3j} are the semi-axes of the j th ellipsoidal inclusion. For a spheroidal inclusion, its symmetry axis is aligned along the Z-axis, namely, $a_{1j} = a_{2j} = a$, and $a_{3j} = b$. $\mathbf{H}_V$ and $\mathbf{H}_r$ have exactly diagonal eigenvalues, that is

$$\mathbf{H}_j = \begin{bmatrix} H_j & 0 & 0 \\ 0 & H_j & 0 \\ 0 & 0 & 1-2H_j \end{bmatrix}, (j = r, V) \tag{4}$$

with

$$H_j = \frac{1}{2}\left\{1 + \frac{1}{\kappa^2-1}\left[1 - \frac{\kappa}{2\sqrt{\kappa^2-1}}\ln\left(\frac{\kappa+\sqrt{\kappa^2-1}}{\kappa-\sqrt{\kappa^2-1}}\right)\right]\right\}, \kappa \geq 1 \tag{5a}$$

$$H_j = \frac{1}{2}\left\{1 + \frac{1}{\kappa^2-1}\left[1 - \frac{\kappa}{\sqrt{1-\kappa^2}}\arctan\left(\frac{\sqrt{1-\kappa^2}}{\kappa}\right)\right]\right\}, \kappa \leq 1 \tag{5b}$$

where κ is the aspect ratio of spheroid defined as $\kappa = b/a$. If $\kappa > 1$, the