Size-dependent effective properties of a heterogeneous material with interface energy effect: from finite deformation theory to infinitesimal strain analysis

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Summary

In this paper, the change of the elastic fields induced by the interface energies and the interface stresses from the reference configuration to the current configuration is considered. It is emphasized that the governing equations taking into account the interface energy effect should be established within the framework of finite deformation in the first place, and then the approximations of governing equations for a finitely deformed multi-phase elastic medium by an infinitesimal strain analysis can be formulated. Hence it can be seen that the asymmetric interface stress has to be used in the Young-Laplace equation. According to the above mentioned formalism, analytical expressions of the size-dependent effective moduli of a particle-filled composite material with interface energy effect are derived. It is shown that, different from the results obtained by previous researchers, the liquid-like surface/interface tension, as a residual stress-type term, also influences the effective property of the composite

1 Introduction

The concept of surface free energy in solids was first introduced by Gibbs [1]. Since then this concept was further developed by many researchers (e.g. [2,3,4,5,6,7,8,9,10,11,12]). As the characteristic size of a solid approaches the nano-scale, for instance, for nano-size structures and nanocomposites, the surface/interface energy effect on its mechanical and physical properties becomes substantial and thus needs to be taken into account in the deformation analysis. In this regard, two kinds of fundamental equation are necessary in the solution of boundary-value problems for stress fields with surface/interface effect. The first is the surface/interface constitutive relations, and the second is the discontinuity conditions of the stress across the interface, namely, the Young-Laplace equations. The above fundamental equations can be used to predict the effective moduli of a composite material with surface/interface energy effect. However, even if an infinitesimal analysis is employed, these equations should be established within the framework of finite deformation in the first place. The reasons for this are: (1) In the study of the mechanical behavior of a composite material or a structure, what we are concerned with is the mechanical response from the reference configuration to the current

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configuration. During the deformation process, the size and the shape of the interface will change, hence the curvature tensor in the governing equations will change too. This means that the deformation will change the residual elastic field induced by the interface energy, and the effect of the interface energy manifests itself precisely through the change of the residual elastic field due to the change of configuration. Therefore, this is essentially a finite deformation problem. (2) For the interface energy model, there should be a residual elastic field due to the presence of the interface energy (and the interface stress) in the material, even though there is no external loading. Thus, by taking into account the change of the residual elastic field due to the change of configuration, the influence of the liquid-like surface tension on the effective properties of a composite material can also be included. In this paper, we will focus on the discussions of the interface energy model. (3) Recently, Huang and Wang [13] derived the constitutive relations for hyperelastic solids with the surface/interface energy effect at finite deformation. These constitutive relations are expressed in terms of the free energy of the interface per unit area at the current configuration, denoted by γ. In particular, for an isotropic interface, γ can be written as a function of J_1 and J_2, where

$$J_1 = \mathrm{tr}\mathbf{U}_s = \mathrm{tr}\mathbf{V}_s, \;\; J_2 = \det\mathbf{U}_s = \det\mathbf{V}_s \tag{1}$$

and $\mathbf{U}_s$ and $\mathbf{V}_s$ are the right and left stretch tensors of the interface, respectively. In the case of small deformation, the interface strain can be approximately expressed as

$$\mathbf{E}_s = \frac{1}{2}\left(\mathbf{u}\nabla_{0s} + \nabla_{0s}\mathbf{u}\right) = \mathbf{U}_s - \mathbf{i}_0 \tag{2}$$

where ∇_{0s} is the surface gradient operator on the reference configuration κ_0. $\mathbf{u}\nabla_{0s}$ is the displacement gradient of the interface, and $\mathbf{i}_0$ is the second-rank identity tensor in the tangent plane of the interface in the reference configuration. Thus Eq. (1) can be written as

$$J_1 = 2 + \mathrm{tr}\mathbf{E}_s, \;\; J_2 = 1 + \mathrm{tr}\mathbf{E}_s + \det\mathbf{E}_s \tag{3}$$

For an isotropic interface, it can be shown that when the deformation is small, the interface Piola-Kirchhoff stresses of the first and second kinds can be expressed as

$$\mathbf{S}_s = J_2\left(\frac{\partial\gamma}{\partial J_1} + J_2\frac{\partial\gamma}{\partial J_2} + \gamma\right)\mathbf{i}_0 + J_2\frac{\partial\gamma}{\partial J_1}\mathbf{E}_s - J_2\left(\frac{\partial\gamma}{\partial J_1} + J_2\frac{\partial\gamma}{\partial J_2} + \gamma\right)\left(\nabla_{0s}\mathbf{u}\right) \tag{4}$$

$$\mathbf{T}_s = J_2\left(\frac{\partial\gamma}{\partial J_1} + J_2\frac{\partial\gamma}{\partial J_2} + \gamma\right)\mathbf{i}_0 - J_2\left(\frac{\partial\gamma}{\partial J_1} + 2J_2\frac{\partial\gamma}{\partial J_2} + 2\gamma\right)\mathbf{E}_s \tag{5}$$

and the Cauchy stress of the interface can be given by

$$\boldsymbol{\sigma}_s = \left(\frac{\partial\gamma}{\partial J_1} + J_2\frac{\partial\gamma}{\partial J_2} + \gamma\right)\mathbf{i}_0 + \frac{\partial\gamma}{\partial J_1}\mathbf{E}_s \tag{6}$$

Therefore, even if the infinitesimal deformation approximation is used, $\mathbf{S}_s$, $\mathbf{T}_s$ and the Cauchy stress of the interface $\boldsymbol{\sigma}_s$ are not the same. This situation is completely different from that in the three dimensional analysis in classical elasticity, in which there is no residual stress in the reference configuration. This means that in the study of the interface energy effect on the mechanical properties of a heterogeneous material, only starting from a finite deformation theory can we correctly choose an appropriate infinitesimal interface stress to be used in the governing equations.

In the following, we shall derive the approximate expressions of the changes of the interface stress and the Young-Laplace equation due to the change of configuration under infinitesimal deformation. As an application of the present theory, we also give the analytical expressions for the effective moduli of a particle-reinforced composite. It is shown that a liquid-like surface/interface tension also affects the effective moduli, which has not been discussed in the literature.

2 Infinitesimal Deformation Approximation

It is well known that in the infinitesimal analysis in classical elasticity, the governing equations, such as the equilibrium equations, are based on one configuration. However, in order to study the interface energy effect, the residual elastic field induced by the interface energy should be taken into account. As was mentioned before, in the study of the macroscopic response of a composite material or a structure, we are usually not interested in the interface induced residual elastic field itself in the reference configuration or in the current one, but the change of this residual elastic field from the former to the latter. Obviously, the difference of the Cauchy stress (in the bulk material as well as at the interface) based on Eulerian description can not be used to describe this change. This is because after and before deformation, the Cauchy stresses are not in the same configuration, so they are not comparable. Hence the Lagrangian description is preferable. The generalized Young-Laplace equation based on the Lagrangian description was given by Huang and Wang [13], and can be expressed in terms of the interface Piola-Kirchhoff stress of the first kind as follows:

$$\begin{cases} \mathbf{N} \cdot \llbracket \mathbf{S}^0 \rrbracket \cdot \mathbf{N} = -\mathbf{S}_s : \mathbf{b}_0 \\ \mathbf{P}_0 \cdot \llbracket \mathbf{S}^0 \rrbracket \cdot \mathbf{N} = -\mathbf{S}_s \cdot \nabla_{0s} \end{cases} \tag{7}$$

where the symbol $\llbracket \cdot \rrbracket$ denotes the discontinuity of a quantity across the interface, $\mathbf{S}^0$ is the first kind Piola-Kirchhoff stress in the bulk material. $\mathbf{P}_0 = \mathbf{I} - \mathbf{N} \otimes \mathbf{N}$, and $\mathbf{I}$ is the unit tensor in three-dimensional space. $\mathbf{N}$ is the unit normal vector to the interface in the reference configuration κ_0. $\mathbf{b}_0$ is the curvature tensor of the interface in κ_0.

It is seen that the change of the residual elastic field induced by the interface energy can be described by the difference of the above equation, which gives

$$\begin{cases} \mathbf{N} \cdot \llbracket \Delta\mathbf{S}^0 \rrbracket \cdot \mathbf{N} = -\Delta\mathbf{S}_s : \mathbf{b}_0 \\ \mathbf{P}_0 \cdot \llbracket \Delta\mathbf{S}^0 \rrbracket \cdot \mathbf{N} = -\Delta\mathbf{S}_s \cdot \nabla_{0s} \end{cases} \tag{8}$$

where Δ denotes the difference of the quantities between the current and reference configurations. The above discussion indicates that in order to take into account the interface energy effect correctly, the interface Piola-Kirchhoff stress of the first kind $\mathbf{S}_s$ should be utilized in the analysis. This is the key point of the present paper, and it seems to be ignored by previous researchers in the study of the effective properties of a heterogeneous material with interface energy effect.

Now an infinitesimal deformation approximation can be performed as follows. In the case of infinitesimal deformation, $\Delta\mathbf{S}^0$ in Eq.(8) can be approximated by the difference of the bulk Cauchy stress between the current and reference configurations, and $\mathbf{S}_s$ can be expressed in terms of the free energy of the interface γ by Eq. (4). In order to simplify the discussion, we can further linearize the expression of the interface free energy. For this, γ will be expanded as follows:

$$\gamma = \gamma_0 + \gamma_1 (J_1 - 2) + \gamma_2 (J_2 - 1) + \frac{1}{2}\gamma_{11} (J_1 - 2)^2 + \gamma_{12} (J_1 - 2)(J_2 - 1) + \frac{1}{2}\gamma_{22} (J_2 - 1)^2 + \cdots \tag{9}$$

In the above expression, γ_0 is equivalent to the surface tension of a liquid-like material. $J_1 - 2$ and $J_2 - 1$ are first-order small quantities. If we only keep the first-order small quantities in Eq. (4) and neglect higher-order small quantities, then from

$$J_2 \left(\frac{\partial \gamma}{\partial J_1} + J_2 \frac{\partial \gamma}{\partial J_2} + \gamma \right) = \gamma_0^* + \left(\gamma_0^* + \gamma_1^* \right) \mathrm{tr}\, \mathbf{E}_s \tag{10}$$

and

$$J_2 \frac{\partial \gamma}{\partial J_1} = \gamma_1 + \left(\gamma_0 + \gamma_{11} + \gamma_{12} \right) \mathrm{tr} \mathbf{E}_s \tag{11}$$

we have

$$\mathbf{S}_s = \gamma_0^* \mathbf{i}_0 + \left(\gamma_0^* + \gamma_1^* \right) \left(\mathrm{tr} \mathbf{E}_s \right) \mathbf{i}_0 - \gamma_0^* \nabla_{0s} \mathbf{u} + \gamma_1 \mathbf{E}_s \tag{12}$$

$$\boldsymbol{\sigma}_s = \gamma_0^* \mathbf{i}_0 + \gamma_1^* \left(\mathrm{tr} \mathbf{E}_s \right) \mathbf{i}_0 + \gamma_1 \mathbf{E}_s \tag{13}$$

where $\gamma_0^* = \gamma_0 + \gamma_1 + \gamma_2$, $\gamma_1^* = \gamma_1 + 2\gamma_2 + \gamma_{11} + 2\gamma_{12} + \gamma_{22}$. Gurtin and Murdoch [5] have also previously derived a similar expression by assuming that the interface stress is an isotropic linear function of the interface strain. Here, we have derived the interface stress in terms of the interface energy, and will emphasize the change of the interface stress due to the change of configuration in the following discussion. We also apply this theoretical framework to predict the effective moduli of heterogeneous media with the interface energy effect. In the reference configuration κ_0, the "residual" Piola-Kirchhoff stress of the first kind of the interface is given by

$$\mathbf{S}_s \big|_0 = \gamma_0^* \mathbf{i}_0 \tag{14}$$

So, in the case of infinitesimal deformation, the difference of the interface Piola-Kirchhoff stress of the first kind between the current and reference configurations, $\Delta \mathbf{S}_s$, can be expressed as

$$\Delta \mathbf{S}_s = \left(\gamma_0^* + \gamma_1^* \right) \left(\mathrm{tr}\, \mathbf{E}_s \right) \mathbf{i}_0 - \gamma_0^* \nabla_{0s} \mathbf{u} + \gamma_1 \mathbf{E}_s \tag{15}$$

It can be seen that, in general, at least three independent material parameters γ_0^*, γ_1^* and γ_1 are needed in the above equation. Only for some special cases, for example, a spherical inhomogeneity embedded in an infinite matrix material subjected to axisymmetric loading, can $\nabla_{0s} \mathbf{u}$ be expressed as a symmetric second-order tensor in two-dimensional space. Then, Eq. (15) becomes

$$\Delta \mathbf{S}_s = \left(\gamma_0^* + \gamma_1^* \right) \left(\mathrm{tr}\, \mathbf{E}_s \right) \mathbf{i}_0 - \left(\gamma_0^* - \gamma_1 \right) \mathbf{E}_s \tag{16}$$

The right hand side of the above expression can be formally written as $\lambda_s \left(\mathrm{tr}\, \mathbf{E}_s \right) \mathbf{i}_0 + 2\mu_s \mathbf{E}_s$, where λ_s and μ_s are called interface moduli, which are given by

$$\begin{cases} \lambda_s = \gamma_0^* + \gamma_1^* \\ \mu_s = -\frac{1}{2}\left(\gamma_0^* - \gamma_1 \right) = -\frac{1}{2}\left(\gamma_0 + \gamma_2 \right) \end{cases} \tag{17}$$

It is interesting to note that in some cases μ_s may be negative. This is because in general the surface/interface energy γ_0 at κ_0 is positive, since otherwise a liquid or a solid would gain energy upon fragmentation (e.g., cf. [9], page 595), and γ_2 is the change rate of the interface energy due to the change of the interface area, and the negative μ_s has been confirmed by Shenoy [14] in his atomistic calculations. Substituting $\Delta\mathbf{S}_s$ in Eq. (15) or (16) into Eq. (8) yields the discontinuity conditions of the traction across the interface in the reference configuration κ_0. These discontinuity conditions, together with other governing equations, can be used to predict the macroscopic mechanical response of composites with the interface energy effect.

3 Effective Moduli of a Particle-Filled Composite

As an illustrative example of the above theoretical framework, in this section, we study the effective properties of a composite reinforced by spherical particles. There have been many models in classical micromechanics which can be used to predict effective moduli of composites (e.g. [15, 16, 17, 18]). There also have been many works for inhomogeneities with imperfect interface bonding conditions or interface effects (e.g. [19, 20, 21, 22, 23, 24]). Previously, Sharma and Ganti [25] have calculated the effective bulk moduli of spherical particle-filled composites with the interface effect using the composite spheres assemblage model (CSA, [26]), and Duan et al. [23] have calculated the effective bulk and shear moduli of such composites using the composite spheres assemblage model, the Mori-Tanaka method (MTM, [27]) and the generalized self-consistent method (GSCM, [28]). The difference between the present work and those of Sharma and Ganti [25], and Duan et al. [23] is that here, starting from the finite deformation theory proposed by Huang and Wang [13], we have derived the infinitesimal deformation approximations of the interface constitutive relation and the Lagrangian description of the Young-Laplace equation by considering the change of configuration. Hence we can explicitly demonstrate the necessity of using the asymmetric interface stress in the Young-Laplace equation and show the influence of the residual surface/interface tension γ_0^* on the effective elastic moduli. For the present inhomogeneity problem with the interface energy effect, if we take the inhomogeneity together with the interface as an “equivalent inhomogeneity”, then the micromechanical schemes for two-phase composites are readily applicable. In this case, the volume averages of the stress and strain for the “equivalent inhomogeneity” must be calculated on the matrix side due to the discontinuity of the stress across the interface. Therefore, the key point is how to derive the stress discontinuity conditions across the interface correctly. This paper takes into account the change of the interface stress in Eq. (8) and Eq. (15) due to the change of configuration, and the interface moduli are directly related to the parameters of the interface energy, thus providing an in-depth understanding of the interface energy effect.

Now consider a two-phase composite composed of a matrix and randomly distributed spherical inhomogeneities. The radius of the inhomogeneity is assumed to be *a*. The effective stiffness tensor of the composite can be expressed as

$$\bar{\mathbf{L}} = \mathbf{L}_0 + f\left(\mathbf{L}_* - \mathbf{L}_0\right) : \mathbf{A} \tag{18}$$

where $\mathbf{L}_0$ and $\mathbf{L}_*$ are the stiffness tensors of the matrix and the “equivalent inhomogeneity” (i.e. an inhomogeneity together with the interface), f is the volume fraction of the inhomogeneities, and **A** denotes the fourth-order strain concentration tensor of the “equivalent inhomogeneity”. If we use the Mori-Tanaka approximation method (e.g. [29]), then **A** is given as

$$\mathbf{A} = \mathbf{A}^0 : \left[\left(1 - f\right) \mathbf{I}^{(1)} + f\mathbf{A}^0 \right]^{-1} \tag{19}$$

where $\mathbf{I}^{(1)}$ is the fourth-order unit tensor, and $\mathbf{A}^0$ is the strain concentration tensor of the "equivalent inhomogeneity" in an infinite matrix corresponding to dilute distribution of inhomogeneities. If the matrix and particles are all isotropic and the inhomogeneities are randomly distributed, then the composite material is statistically isotropic and the elastic moduli in Eq. (18) can be written as

$$\begin{cases} \bar{\mathbf{L}} = 3\bar{K}\mathbf{I}_m + 2\bar{\mu}\mathbf{I}_s \\ \mathbf{L}_0 = 3K_0\mathbf{I}_m + 2\mu_0\mathbf{I}_s \\ \mathbf{L}_* = 3K_*\mathbf{I}_m + 2\mu_*\mathbf{I}_s \end{cases} \tag{20}$$

where K_0, K_* and $\bar{K}$ are the bulk moduli of the matrix, the "equivalent inhomogeneity" and the composite, respectively. μ_0 , μ_* and $\bar{\mu}$ are shear moduli of the matrix, the "equivalent inhomogeneity" and the composite, respectively. $\mathbf{I}_m = \frac{1}{3}\mathbf{I} \otimes \mathbf{I}$, $\mathbf{I}_s = \mathbf{I}^{(1)} - \mathbf{I}_m$. Eq. (18) can be further decoupled into

$$\begin{cases} \bar{K} = K_0 + f\left(K_* - K_0\right)A_m \\ \bar{\mu} = \mu_0 + f\left(\mu_* - \mu_0\right)A_s \end{cases} \tag{21}$$

where A_m and A_s are the constants in the strain concentration tensors corresponding to the bulk and shear moduli, respectively. They are given by

$$A_m = \frac{K_0}{K_0 + (1-f)(K_* - K_0)\omega_m}, \quad A_s = \frac{\mu_0}{\mu_0 + (1-f)(\mu_* - \mu_0)\omega_s} \tag{22}$$

where

$$\omega_m = \frac{3K_0}{3K_0 + 4\mu_0}, \quad \omega_s = \frac{6(K_0 + 2\mu_0)}{5(3K_0 + 4\mu_0)} \tag{23}$$

It can be seen that the key point is to calculate the elastic moduli K_* and μ_* of the "equivalent inhomogeneity". To this end, Eqs. (8) and (15) are needed. For the spherical inhomogeneity of radius a, $\nabla_{0s}\mathbf{u}$ can be expressed in terms of the physical components (u_r, u_θ, u_φ) in a spherical polar coordinate system

$$\begin{aligned} \nabla_{0s}\mathbf{u} = &\left(\frac{\partial u_\theta}{r\partial\theta} + \frac{u_r}{r}\right)\mathbf{e}_\theta \otimes \mathbf{e}_\theta + \left(\frac{\partial u_\varphi}{r\partial\theta}\right)\mathbf{e}_\theta \otimes \mathbf{e}_\varphi + \left(\frac{1}{r\sin\theta}\frac{\partial u_\theta}{\partial\varphi} - \cot\theta\frac{u_\varphi}{r}\right)\mathbf{e}_\varphi \otimes \mathbf{e}_\theta \\ &+ \left(\frac{1}{r\sin\theta}\frac{\partial u_\varphi}{\partial\varphi} + \cot\theta\frac{u_\theta}{r} + \frac{u_r}{r}\right)\mathbf{e}_\varphi \otimes \mathbf{e}_\varphi \end{aligned} \tag{24}$$

For axisymmetric loading, $u_\varphi = 0$, and u_r and u_θ are not dependent on φ, hence $\nabla_{0s}\mathbf{u}$ is a symmetric tensor

$$\nabla_{0s}\mathbf{u} = \left(\frac{\partial u_\theta}{r\partial\theta} + \frac{u_r}{r}\right)\mathbf{e}_\theta \otimes \mathbf{e}_\theta + \left(\cot\theta\frac{u_\theta}{r} + \frac{u_r}{r}\right)\mathbf{e}_\varphi \otimes \mathbf{e}_\varphi \tag{25}$$

In this case, Eq. (15) can be replaced by Eq.(16). Noting that in the reference configuration, the curvature tensor on the surface of the sphere with radius *a* is

$$\mathbf{b}_0 = -\frac{1}{a}\mathbf{i}_0 \tag{26}$$

Eq. (8) can be written as

$$\begin{cases} [\![\sigma_{rr}]\!]\big|_{r=a} = \dfrac{1}{a^2}\left(\gamma_0^* + 2\gamma_1^* + \gamma_1\right)\left(2u_r + u_\theta \cot\theta + \dfrac{\partial u_\theta}{\partial \theta}\right)\Bigg|_{r=a} \\ [\![\sigma_{r\theta}]\!]\big|_{r=a} = \dfrac{1}{a^2}\Bigg[\left(\gamma_0^* + \gamma_1^*\right)u_\theta + \left(\gamma_1^* + \gamma_1\right)\left(u_\theta \cot^2\theta - \dfrac{\partial^2 u_\theta}{\partial \theta^2} - \dfrac{\partial u_\theta}{\partial \theta}\cot\theta\right) \\ \qquad\qquad -\left(\gamma_0^* + 2\gamma_1^* + \gamma_1\right)\dfrac{\partial u_r}{\partial \theta}\Bigg]\Bigg|_{r=a} \end{cases} \tag{27}$$

In order to calculate the bulk modulus K_* of the "equivalent inhomogeneity", let us consider the inhomogeneity problem where a spherical inhomogeneity is embedded in an infinite medium under a hydrostatic loading with the remote strain

$$\mathbf{E}^\infty = \frac{1}{3}E_m\mathbf{I} \tag{28}$$

In this case, the displacement and stress fields in the inhomogeneity and matrix are given by

$$\begin{cases} u_r^i = F_i r + G_i / r^2 \\ \sigma_{rr}^i = 3K_i F_i - 4\mu_i G_i / r^3 \end{cases} \tag{29}$$

The superscript *i* =1, 0 denotes the quantities of the inhomogeneity and matrix, respectively, F_1, F_0, G_1 and G_0 are constants to be determined. In addition to the displacement continuity condition at the interface *r* = *a*, the elastic solution needs to satisfy the stress discontinuity condition in Eq.(27), that is,

$$\left(\sigma_{rr}^0 - \sigma_{rr}^1\right)\Big|_{r=a} = \frac{2}{a^2}\left(\gamma_0^* + 2\gamma_1^* + \gamma_1\right)\left(u_r\big|_{r=a}\right) \tag{30}$$

From the above conditions, the non-singular condition at the origin and the condition at the infinity, the constants in Eq. (29) can be determined and thus the bulk modulus K_* of the "equivalent inhomogeneity" can be given by

$$K_* = \frac{tr\langle\boldsymbol{\sigma}_*\rangle}{3tr\langle\boldsymbol{\varepsilon}_*\rangle}\Bigg|_{r=a} = K_1 + \frac{2\left(\gamma_0^* + 2\gamma_1^* + \gamma_1\right)}{3a} \tag{31}$$

In the above expression, $\langle\boldsymbol{\sigma}_*\rangle$ and $\langle\boldsymbol{\varepsilon}_*\rangle$ represent the volume averages of the stress and strain of the "equivalent inhomogeneity" that includes the inhomogeneity and the interface.

The shear modulus μ_* of the "equivalent inhomogeneity" can be calculated by imposing a pure deviatoric remote strain at the infinity

$$\mathbf{E}^\infty = E_e\left[\mathbf{e}_3 \otimes \mathbf{e}_3 - \frac{1}{2}\left(\mathbf{e}_1 \otimes \mathbf{e}_1 + \mathbf{e}_2 \otimes \mathbf{e}_2\right)\right] \tag{32}$$

where $\mathbf{e}_1$, $\mathbf{e}_2$ and $\mathbf{e}_3$ are the base vectors in a rectangular Cartesian coordinate system. From the solution of Lur'e [30], the displacement and stress fields in the inhomogeneity and matrix can be expressed by

$$\begin{cases} u_r^1 = \left(12\nu_1 A r^3 + 2Br\right) P_2\left(\cos\theta\right) \\ u_r^0 = \left(E_e r + \dfrac{2\left(5-4\nu_0\right)}{r^2} C - \dfrac{3}{r^4} D\right) P_2\left(\cos\theta\right) \\ u_\theta^1 = \left[\left(7-4\nu_1\right) A r^3 + Br\right] \dfrac{dP_2\left(\cos\theta\right)}{d\theta} \\ u_\theta^0 = \left[\dfrac{1}{2} E_e r + \dfrac{\left(2-4\nu_0\right)}{r^2} C + \dfrac{1}{r^4} D\right] \dfrac{dP_2\left(\cos\theta\right)}{d\theta} \\ \sigma_{rr}^1 = 2\mu_1 \left(-6\nu_1 A r^2 + 2B\right) P_2\left(\cos\theta\right) \\ \sigma_{rr}^0 = 2\mu_0 \left[E_e - \dfrac{4\left(5-\nu_0\right)}{r^3} C + \dfrac{12}{r^5} D\right] P_2\left(\cos\theta\right) \\ \sigma_{r\theta}^1 = 2\mu_1 \left[\left(7+2\nu_1\right) A r^2 + B\right] \dfrac{dP_2\left(\cos\theta\right)}{d\theta} \\ \sigma_{r\theta}^0 = 2\mu_0 \left[\dfrac{1}{2} E_e + \dfrac{2\left(1+\nu_0\right)}{r^3} C - \dfrac{4}{r^5} D\right] \dfrac{dP_2\left(\cos\theta\right)}{d\theta} \end{cases} \tag{33}$$

The superscripts 1 and 0 denote the quantities of the inhomogeneity and matrix, respectively. ν_1 and ν_0 are Poisson ratios of the inhomogeneity and matrix, respectively. $P_2\left(\cos\theta\right)$ is the second-order Legendre polynomial. *A*, *B*, *C* and *D* are constants to be determined. Again, in addition to the displacement continuity condition at the interface $r = a$, the elastic solution needs to satisfy the stress discontinuity condition in Eq. (27). Then, the unknown constants can be determined in a way similar to that for the bulk modulus. The shear modulus μ_* of the "equivalent inhomogeneity" can be given by

$$\begin{cases} \mu_* = \left.\dfrac{\left\langle \boldsymbol{\sigma}_* \right\rangle_e}{3\left\langle \boldsymbol{\varepsilon}_* \right\rangle_e}\right|_{r=a} = \dfrac{\mu_1 L + L_0 + L_1 + L_2}{L + L_3} \\ L = 10\left[4\mu_0\left(7-10\nu_1\right) + \mu_1\left(7+5\nu_1\right)\right] \\ L_0 = 5\left[4\mu_0\left(10\nu_1 - 7\right) + 3\mu_1\left(7-15\nu_1\right)\right]\left(\gamma_0^* - 2\gamma_1 - \gamma_1^*\right)/a \\ L_1 = \left[12\mu_0\left(10\nu_1 - 7\right) + 5\mu_1\left(91-139\nu_1\right)\right]\left(\gamma_1 + \gamma_1^*\right)/a \\ L_2 = 10\left(10\nu_1 - 7\right)\left[\left(\gamma_0^* - 2\gamma_1 - \gamma_1^*\right)^2 + 3\left(\gamma_1 + \gamma_1^*\right)^2 + 4\left(\gamma_0^* - 2\gamma_1 - \gamma_1^*\right)\left(\gamma_1 + \gamma_1^*\right)\right]/a^2 \\ L_3 = -4\left(10\nu_1 - 7\right)\left(5\gamma_0^* + 7\gamma_1 + 12\gamma_1^*\right)/a \end{cases} \tag{34}$$

where $\left\langle \boldsymbol{\sigma}_* \right\rangle_e$ and $\left\langle \boldsymbol{\varepsilon}_* \right\rangle_e$ are the effective average stress and the effective average strain of the "equivalent inhomogeneity" , respectively.

It can be easily seen that K_* and μ_* are not only functions of the elastic moduli of the inhomogeneity, but also the functions of the size of it. Substituting the obtained K_* (Eq.(31)) and μ_* (Eq.(34)) into Eq.(21), we obtain the analytical expressions of the effective moduli $\bar{K}$ and $\bar{\mu}$ of the composite

$$\bar{K} = \frac{1}{3}\left[\frac{12K_0\mu_0(1-f)+3K_1(3K_0+4\mu_0 f)+2(3K_0+4\mu_0 f)(\gamma_0^*+2\gamma_1^*+\gamma_1)/a}{3K_0 f+4\mu_0+3K_1(1-f)+2(1-f)(\gamma_0^*+2\gamma_1^*+\gamma_1)/a}\right] \quad (35)$$

$$\begin{cases} \bar{\mu} = \mu_0 + \dfrac{15\mu_0 f(1-\nu_0)\left[(\mu_1-\mu_0)L-\mu_0 L_3+L_0+L_1+L_2\right]}{(1-f)\left[LL_4+10(L_5+L_6)+2(4-5\nu_0)L_2\right]+15\mu_0 f(1-\nu_0)(L+L_3)} \\ L_4 = 2\mu_1(4-5\nu_0)+\mu_0(7-5\nu_0) \\ L_5 = \left[2\mu_0(7-10\nu_1)(5\nu_0-1)-3\mu_1(15\nu_1-7)(4-5\nu_0)\right](\gamma_0^*-2\gamma_1-\gamma_1^*)/a \\ L_6 = \left[2\mu_0(7-10\nu_1)(19-11\nu_0)+\mu_1(91-139\nu_1)(4-5\nu_0)\right](\gamma_1+\gamma_1^*)/a \end{cases} \quad (36)$$

It is noted that the above effective bulk modulus can also be obtained by using the composite sphere assemblage model (e.g. [31]).

4 Discussions

In the following, we discuss two special cases, namely, the effective moduli of a porous material containing nano-voids and a two-phase composite where the interface is liquid-like. First, for the porous material containing spherical nano-voids, the effective moduli are

$$\bar{K}_{void} = \frac{1}{3}\left[\frac{12K_0\mu_0(1-f)+2(3K_0+4\mu_0 f)n_3}{3K_0 f+4\mu_0+2(1-f)n_3}\right] \quad (37)$$

$$\begin{cases} \bar{\mu}_{void} = \dfrac{\mu_0}{2}\left[\dfrac{4(1-f)m_1\mu_0^2+4(2m_2-fm_1)\mu_0 n_1+42m_4\mu_0 n_2+(m_1 f+2m_3)n_2 n_3}{2(2fm_3+m_1)\mu_0^2+4(fm_3+m_2)\mu_0 n_1+21m_4\mu_0 n_2+(1-f)m_3 n_2 n_3}\right] \\ m_1 = 7-5\nu_0,\ m_2 = 5-4\nu_0,\ m_3 = 4-5\nu_0,\ m_4 = 1-\nu_0 \\ n_1 = (\gamma_0^*+\gamma_1^*)/a,\ n_2 = (\gamma_1-\gamma_0^*)/a,\ n_3 = (\gamma_0^*+2\gamma_1^*+\gamma_1)/a \end{cases} \quad (38)$$

Second, if the interface of a two-phase composite containing spherical inhomogeneities behaves like that of a liquid, namely, $\gamma_0^* = \gamma_0$, $\gamma_1^* = \gamma_1 = 0$, then the effective moduli of the composite are

$$\bar{K}_{(\gamma_0)} = \frac{1}{3}\left[\frac{12K_0\mu_0(1-f)+3K_1(3K_0+4\mu_0 f)+2(3K_0+4\mu_0 f)\gamma_0/a}{3K_0 f+4\mu_0+3K_1(1-f)+2(1-f)\gamma_0/a}\right] \quad (39)$$

$$\begin{cases} \bar{\mu}_{(\gamma_0)} = \mu_0 + \dfrac{15\mu_0 f(1-\nu_0)\left[(\mu_1-\mu_0)L-\mu_0 L_3^*+L_0+L_1+L_2^*\right]}{(1-f)\left[LL_4+10L_5+2(4-5\nu_0)L_2^*\right]+15\mu_0 f(1-\nu_0)(L+L_3^*)} \\ L_2^* = 10(10\nu_1-7)\gamma_0^2/a^2 \\ L_3^* = -20(10\nu_1-7)\gamma_0/a \end{cases} \quad (40)$$

This means that the surface/interface tension γ_0 does affect the effective moduli of the composite.

In order to compare the above results with those obtained by other interface models, let us consider an interface with the following properties: The displacement across the interface is continuous, but the traction is allowed to have a discontinuity across the interface; moreover, we assume that there is no residual elastic field induced by the interface tension when the material is not subjected to any external loading. This kind of interface can also be regarded as an equivalency of a thin and stiff interphase (e.g. [20, 32]). Under infinitesimal deformation, the constitutive relation of the above interface can be written as

$$\boldsymbol{\sigma}_s = \lambda_s^{'}\left(tr\mathbf{E}_s\right)\mathbf{i}_0 + 2\mu_s^{'}\mathbf{E}_s \tag{41}$$

It can be seen that Eq. (41) can be obtained directly by setting $\gamma_0^* = 0$ in Eqs. (12), (13) and (17), with

$$\lambda_s^{'} = \gamma_1^*,\ \mu_s^{'} = \frac{1}{2}\gamma_1 \tag{42}$$

In this case, there is no need to distinguish between the interface Piola-Kirchhoff stress of the first kind $\mathbf{S}_s$ and the Cauchy stress of the interface $\boldsymbol{\sigma}_s$. Hence we can apply the infinitesimal deformation formulation from the outset.

Based on Eq.(41), the effective bulk and shear moduli of the composite filled with spherical particles can easily be obtained by simply setting $\gamma_0^* = 0$ in Eqs. (35) and (36). Obviously, the effective moduli so-obtained are not influenced by the residual interface tension. In particular, it can be seen from Eqs. (39) and (40) that the effective moduli of the composite material with a liquid-like interface will be the same as those of a perfectly-bonded composite material, if Eq. (41) is utilized.

Figures 1 and 2 show the variations of the effective bulk and shear moduli predicted in Eqs. (37) and (38) for polypropylene containing spherical voids. The bulk modulus of the matrix material is K_0 = 2.5 GPa, and the shear modulus is μ_0 = 0.5 GPa. It is assumed that the surface is liquid-like with a surface tension γ_0 = 0.05 J/m^2. The volume fractions of the voids are assumed to be f = 20% and f = 20% respectively. In the figures, $\bar{K}_0$ and $\bar{\mu}_0$ are the effective bulk and shear moduli of the material without the surface energy effect. It is seen from these figures that the surface effect decreases with the increase of the size of the voids, and can be neglected when the radius of the void is larger than 30 nm.

Several points should be mentioned: 1) In general, the displacement gradient of the interface is not symmetric. For instance, for a cylindrical inclusion embedded in an infinite matrix, the displacement gradient of the interface can be expressed in terms of the physical components (u_r, u_θ, u_z) in a cylindrical polar coordinate system (r, θ, z):

$$\nabla_{0s}\mathbf{u} = \left(\frac{1}{r}\frac{\partial u_\theta}{\partial \theta} + \frac{u_r}{r}\right)\mathbf{e}_\theta \otimes \mathbf{e}_\theta + \frac{1}{r}\frac{\partial u_z}{\partial \theta}\mathbf{e}_\theta \otimes \mathbf{e}_z + \frac{\partial u_\theta}{\partial z}\mathbf{e}_z \otimes \mathbf{e}_\theta + \frac{\partial u_z}{\partial z}\mathbf{e}_z \otimes \mathbf{e}_z \tag{43}$$

Obviously, under an anti-plane shear deformation, Eq.(43) is not a symmetric tensor. In this case, an asymmetric Piola-Kirchhoff stress Eq.(15) has to be used in Eq.(8). 2) In this paper, we only used the Mori-Tanaka approximation method to predict the effective moduli of a composite. As was pointed out by Weng [33], for spherical inclusions, the effective moduli derived from the Mori-Tanaka method are identical to the Hashin-Shtrikman bounds [34]. Actually, once K_* and μ_* are obtained, the effective moduli can also be calculated by using other micromechanical schemes such as the generalized self-consistent method [28], the double-inclusion method [16], and the IDD estimate [35]. This will not be further discussed here; 3) Although, only inhomogeneities with the same radius were considered in the above discussion, the method can also be applied to the materials with inhomogeneities of different sizes. As K_* and μ_* are related to the size of the inhomogeneity, inhomogeneities with different radii should be treated as different “equivalent inhomogeneity”; 4) We have assumed that the interface is elastically isotropic. However, in many materials such as single crystals, the interface may be anisotropic. In this case, the interface constitutive relations for anisotropic materials given by Huang and Wang [13] should be used, and the effective moduli of the composite can still be predicted by using the above procedure. The only difference is that there are more material parameters and the expressions of the effective moduli may become complicated.

5 Concluding Remarks

Beginning with the finite deformation analysis of a multi-phase hyperelastic medium, the interface energy effect on the macroscopic mechanical behavior of a composite is studied. Particular emphasis is placed upon the approximate formulation of a finitely deformed multi-phase elastic medium by an infinitesimal deformation analysis. It is noted that due to the existence of the interface energy, even under no external loading, there is still a “residual elastic field” induced by the interface stress. During the deformation process of a composite from the reference configuration to the current configuration, the changes of the size and shape of the interface result in the change of this “residual elastic field”. The novelty of the present paper is that the governing equations depicting the change of the “residual elastic field” due to the change of configuration are derived under infinitesimal deformation approximation, leading to the use of the asymmetric interface stress in the prediction of the effective properties of heterogeneous materials with interface energy effect. Therefore, the influence of the residual surface/interface tension can also be taken into account. The theoretical framework is applied to obtain the analytical expressions of the effective moduli of a composite containing spherical inhomogeneities. It is shown that the mechanical behavior of the composite exhibits size-dependent effect when the interface energy effect is considered. The results in this paper can find application in the studies of nanocomposites.

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Figures Captions:

(1)Figure 1: Variation of normalized effective bulk modulus with the radius of voids (in Gragh1.EPS).

(2)Figure 2: Variation of normalized effective shear modulus with the radius of voids (in Gragh2.EPS).

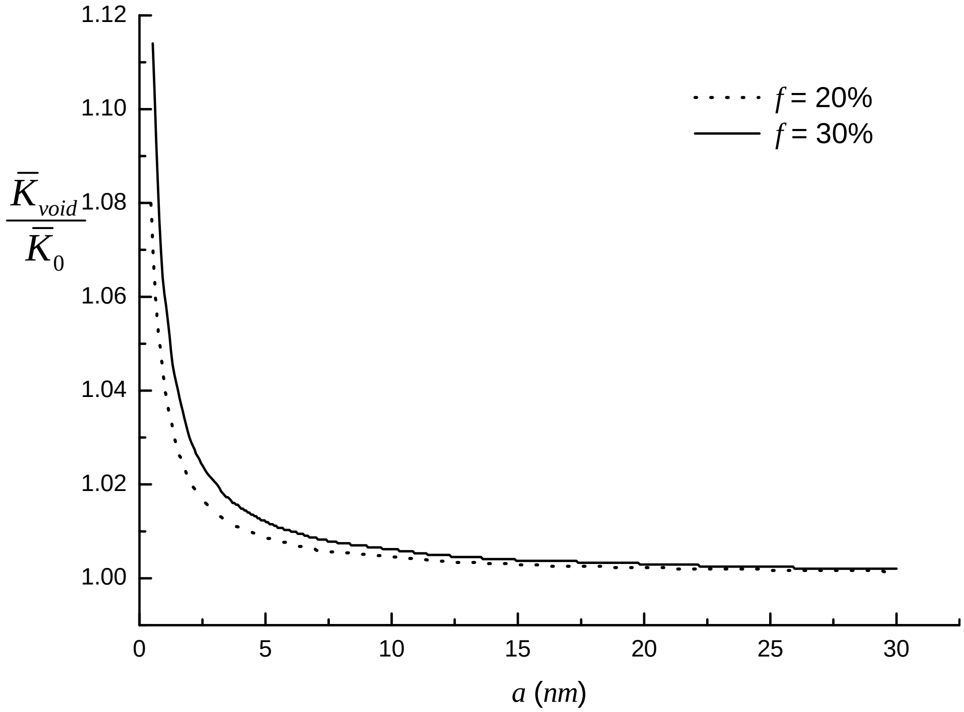

Figure 1 Variation of normalized effective bulk modulus with the radius of voids

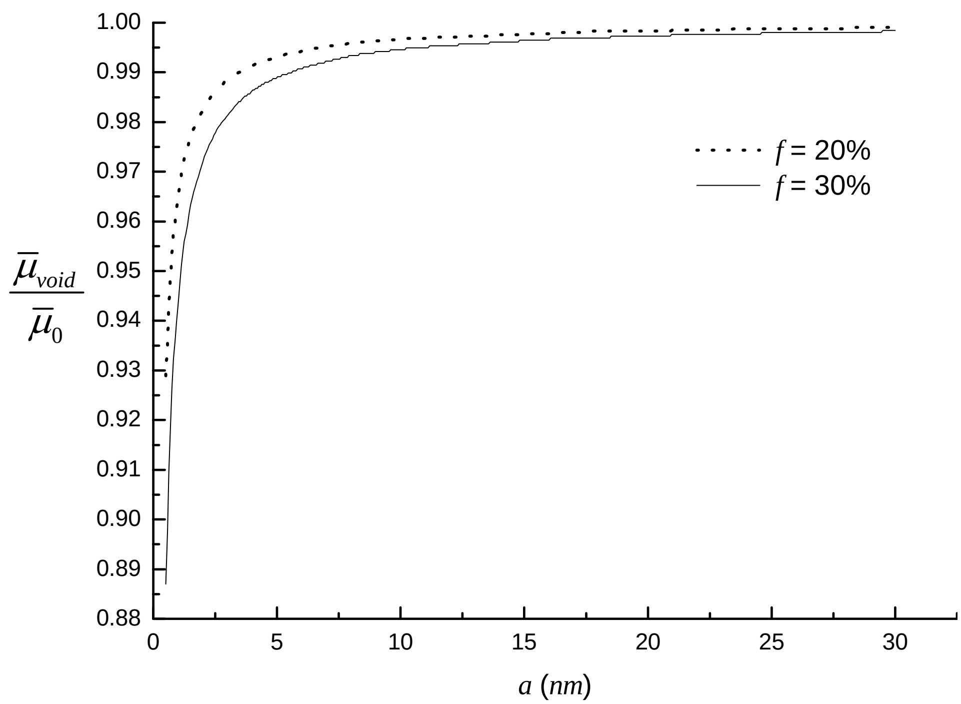

Figure 2 Variation of normalized effective shear modulus with the radius of voids