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Influence of Gradual Interphase on Overall Elastic and Viscoelastic Properties of Particulate Composites

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ABSTRACT: The overall elastic and viscoelastic properties of particulate composites with graded interphase have been analyzed analytically. The localization relation for a coated particle with a graded interphase in the radial direction has been firstly derived, and the effective elastic moduli of such composites are then estimated by the mean field theory and generalized self-consistent method respectively. The effective storage and loss moduli of the composite are also determined through the dynamic correspondence principle. The results show that the nature of the interphase (soft and hard) can have important influence on the prediction for effective elastic and viscoelastic properties of the composite, especially for a soft graded interphase, the predictions based on a uniform interphase model overestimates largely the effective elastic and viscoelastic moduli of the composite.

KEY WORDS: graded interphase, particulate composite, overall elasticity, viscoelasticity, micromechanics.

INTRODUCTION

An INTERPHASE LAYER is usually present in a composite system between the matrix and reinforcement due to sizing [1,2], chemical reaction [3] and diffusion process [4]. This interphase layer modifies the stress transfer mechanism between the matrix and fiber, and could be tailored to an optimal compromise between the strength and toughness of the composite.

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The stress distribution in an interphase layer is closely related to the morphology and property of the interphase, these issues appeal recently an intensive experimental study [2,3,5]. Atom Force Microscope (AFM) and nanoindentation are typically used for such purpose, these techniques allow one to measure the hardness variation in a region near the reinforcement, and the modulus variation in the interphase could then be estimated. Recently Gao and Mader [2] examined the local mechanical property variation in the interphase of sized E-glass fibers reinforced by an epoxy resin and a polypropylene (PPm) matrix respectively; the E-glass fiber is sized by γ-APS and PU. They found an exponential variation of local Young's modulus in the interphase for γ-APS and PU sized fiber reinforced epoxy composites, and near constant of the local modulus for γ-APS and PU sized fiber reinforced PPm composites. A local modulus variation is also reported by Torralba et al. [3] due to the composition change in the interphase region. The measured thickness of the interphase is ranged from hundred to several hundred nanometers, depending on composite systems and measuring technique [6].

The presence of an interphase layer between reinforcement and matrix in a practical composite system recalls a more rigorous mechanical modeling. In the early theoretical study, the interphase is usually considered to be homogeneous in mechanical property, and the coated inclusion model are proposed to evaluate the overall properties, such as elasticity [7], plasticity [8] and viscoelasticity [9]. As indicated in the above discussion, an interphase with a gradual mechanical property is present for practical composite systems, to analyze the influence of this kind of interphase, a more rigorous coated-inclusion model, including the modulus variation in the interphase region, must be considered. Extensive works have been conducted for fiber reinforced composites, for which the bulk and shear moduli of the interphase are allowed to vary in the radial direction [10,11]. However, for a particle reinforced composite with a gradual interphase, there is much less work for a general loading condition. Ding and Weng [12] examine the effective bulk modulus for such a composite. Recently Voros and Pukanszky [13] proposed a method to examine the stress distribution for a rigid inclusion with a soft gradual interphase, the modulus of the interphase region is supposed to follow a power-law type variation in the radial direction.

In this paper, we will follow essentially the method proposed by Voros and Pukanszky [13] to solve localization problem, our main concern is focused on the overall elasticity and viscoelasticity of the composite. The influence of the interphase property on the overall elastic and viscoelastic behavior will be analyzed in detail by different micromechanical methods. The paper will be arranged as follows: in the following section, the "Localization Problem for a Spherical Particle with a Graded Interphase" embedded in a homogeneous matrix is solved for a general remote loading condition; in the next section, the "Effective Modulus of the Composite" with such coated particles is determined by both the Mori–Tanaka method and the generalized self-consistent method, comparison with the homogeneous layer model is also provided. The numerical applications for various interphase variations and particle's property are also given; an extension of the method to viscoelasticity for both the matrix and interphase layer will be presented in "Viscoelastic Properties of the Composite".

LOCALIZATION PROBLEM FOR SPHERICAL PARTICLE WITH A GRADED INTERPHASE

In this section, we will determine the local stress and average stress in different phases for a particle with a graded interphase (coated particle), to make the formulation suitable for the following generalized self-consistent method, this coated particle with another layer of matrix (the outer radius of the matrix layer is denoted by c) is embedded into an homogeneous reference material under a general remote loading condition. The particle, matrix and the reference material are assumed to be isotropic, their bulk and shear moduli are denoted by $\kappa_1, \mu_1, \kappa_3 = \kappa_0, \mu_3 = \mu_0$ and κ_4, μ_4 respectively. The bulk and shear moduli of the interphase are assumed to follow the relation such as:

$$\kappa_2(r) = f(r)\kappa_0, \quad \mu_2(r) = f(r)\mu_0 \tag{1}$$

where r is the distance from the particle surface, $f(r)$ is a function characterizing the spatial variation of the interphase property. The relation (1) implies that the Poisson's ratio is constant in the interphase region and equals to that of the matrix. The function $f(r)$ in this paper is taken to be power-law as proposed by Voros and Pukanszky [13], $f(r) = P(r/b)^{\lambda}$. The parameters a and b represent the radii of the particle and interphase region respectively, the parameters P and λ characterize the spatial property variation in the interphase. The choice of this form for the function $f(r)$ guarantees the analytical solutions for the stress and displacement, this greatly simplifies the subsequent analysis, and this type of function can also reflect approximately the modulus variation observed experimentally. In the following, if the modulus of the interphase is greater than that of the matrix, it is called a hard interphase, otherwise it is called a soft interphase, as shown in Figure 1, these depend on the choice of parameters P and λ.

For the problem to be analyzed, the remote loading is split into hydrostatic and pure shear loading, and these two situations will be examined separately.

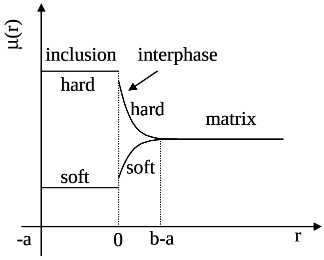

Figure 1. *Illustration of some definitions.*

Solution for a Hydrostatic Stress

In this case, the remote applied macroscopic stresses are: $\Sigma_{xx} = \Sigma_{yy} = \Sigma_{zz} = \Sigma^0$, the other stress components are zero. The expressions for the displacement and the stress in the particle, matrix and the reference medium are of the following form ($i = 1, 3, 4$):

$$u_{r_i} = F_i r + G_i r^{-2} \tag{2}$$

and

$$\begin{aligned} \sigma_{ri} &= 2F_i\kappa_i - 2G_i\mu_i r^{-2} \\ \sigma_{\theta i} &= 2F_i\kappa_i + 2G_i\mu_i r^{-2} \end{aligned} \tag{3}$$

For the interphase layer:

$$u_{r_2} = F_2 r^{s_1} + G_2 r^{s_2} \tag{4}$$

where

$$s_1, s_2 = -\frac{\lambda}{2} \mp \frac{1}{2}\sqrt{\frac{3(\lambda-3)^2\kappa_0 + 4(\lambda+3)^2\mu_0}{3\kappa_0 + 4\mu_0}}$$

With help of the constitutive relation for the interphase layer, the expressions for stresses in this layer are

$$\begin{aligned} \sigma_{r_2} &= \frac{1}{3}P\left(\frac{r}{b}\right)^{\lambda}\{r^{s_1-1}[3(2+s_1)\kappa_0 + 4(s_1-1)\mu_0]F_2 \\ &\quad + r^{s_2-1}[3(2+s_2)\kappa_0 + 4(s_2-1)\mu_0]G_2\} \\ \sigma_{\varphi_2} = \sigma_{\theta_2} &= \frac{1}{3}P\left(\frac{r}{b}\right)^{\lambda}\{r^{s_1-1}[3(2+s_1)\kappa_0 - 2(s_1-1)\mu_0]F_2 \\ &\quad + r^{s_2-1}[3(2+s_2)\kappa_0 - 2(s_2-1)\mu_0]G_2\} \end{aligned} \tag{5}$$

The coefficients $F_i, G_i (i = 1, 2, 3, 4)$ are determined through the continuity conditions at the interfaces between the particle and the interphase, between the interphase and the matrix, and between the matrix and the reference medium. Averaging the stresses over the particle, the interphase layer and also the matrix layer, we get the corresponding average stresses and strains:

$$\overline{\sigma_{xx1}} = \overline{\sigma_{yy1}} = \overline{\sigma_{zz1}} = 3\ F_1\kappa_1 = c_{h1}\Sigma^0,$$
$$\overline{\varepsilon_{xx1}} = \overline{\varepsilon_{yy1}} = \overline{\varepsilon_{zz1}} = F_1 = \frac{c_{h1}}{3\kappa_1}\Sigma^0 = \frac{e_{h1}}{3\kappa_4}\Sigma^0$$
$$\overline{\sigma_{xx2}} = \frac{3P\kappa_0}{a^3 - b^3}\left\{\frac{[a^{2+s_1}(a/b)^{\lambda} - b^{2+s_1}](2+s_1)}{2+s_1+\lambda}F_2 + \frac{[a^{2+s_2}(a/b)^{\lambda} - b^{2+s_2}](2+s_2)}{2+s_2+\lambda}G_2\right\} = c_{h2}\Sigma^0$$
$$\overline{\varepsilon_{xx2}} = \overline{\varepsilon_{yy2}} = \overline{\varepsilon_{zz2}} = \frac{1}{a^3 - b^3}\left[(a^{2+s_1} - b^{2+s_1})F_2 + (a^{2+s_2} - b^{2+s_2})G_2\right] = \frac{e_{h2}}{3\kappa_4}\Sigma^0$$
$$\overline{\sigma_{xx3}} = \overline{\sigma_{yy3}} = \overline{\sigma_{zz3}} = 3F_3\kappa_3 = c_{h3}\Sigma^0\overline{\varepsilon_{xx3}} = \overline{\varepsilon_{yy3}} = \overline{\varepsilon_{zz3}} = F_3 = \frac{c_{h3}}{3\kappa_3}\Sigma^0 = \frac{e_{h3}}{3\kappa_4}\Sigma^0 \tag{6}$$

and $\overline{\sigma_{xx2}} = \overline{\sigma_{yy2}} = \overline{\sigma_{zz2}}$, the other stress components are zero.

Solution for Shear Stress

In this case, the remote applied forces are: $T_1 = \tau n_1, T_2 = -\tau n_2, T_3 = 0$. For the particle, matrix and reference material $(i = 1, 3, 4)$, the displacements and the stresses are [14]

$$\begin{aligned} u_{r_i} &= U_{r_i}(r)\sin^2\theta\cos 2\varphi, \\ u_{\theta_i} &= U_{\theta_i}(r)\sin\theta\cos\theta\cos 2\varphi \\ u_{\varphi_i} &= U_{\varphi_i}(r)\sin\theta\sin 2\varphi \end{aligned} \tag{7}$$

where

$$\begin{aligned} U_{r_i} &= A_1^i r + A_2^i r^3 + A_3^i r^{-2} + A_4^i r^{-4} \\ U_{\theta_i} &= -U_{\varphi_i} = A_1^i r + \frac{7 - 4\nu_i}{6\nu_i}A_2^i r^3 + \frac{2(1+2\nu_i)}{-5+4\nu_i}A_3^i r^{-2} - \frac{2}{3}A_4^i r^{-4} \end{aligned} \tag{8}$$

The corresponding stresses are

$$
\begin{aligned}
\sigma_{r_i} &= \cos 2\varphi \sin^2\theta \mu_i\left(2A_1^i - A_2^i r^2 - A_3^i r^{-3} - A_4^i r^{-5}\right) \\
\sigma_{\theta_i} &= \cos 2\varphi \mu_i\left[\left(A_1^i - \frac{5}{2}A_2^i r^2 + \frac{2\nu_i - 1}{4\nu_i - 5}A_3^i r^{-3} + A_4^i r^{-5}\right)\right. \\
&\qquad \left. + \cos 2\theta\left(2A_1^i + \frac{7(\nu_i + 2)}{6\nu_i}A_2^i r^2 - \frac{2\nu_i - 1}{4\nu_i - 5}A_3^i r^{-3} - \frac{7}{3}A_4^i r^{-5}\right)\right] \\
\sigma_{\varphi_i} &= \cos 2\varphi \mu_i\left[\left(-2A_1^i - \frac{\nu_i + 7}{2\nu_i}A_2^i r^2 - \frac{2\nu_i - 1}{4\nu_i - 5}A_3^i r^{-3} + 3A_4^i r^{-5}\right)\right. \\
&\qquad \left. + \cos 2\theta\left(\frac{11\nu_i + 7}{6\nu_i}A_2^i r^2 - \frac{2\nu_i - 1}{4\nu_i - 5}A_3^i r^{-3} - \frac{5}{3}A_4^i r^{-5}\right)\right] \\
\sigma_{r\theta_i} &= \cos\theta\cos 2\varphi \sin\theta \mu_i\left[2A_1^i + \frac{2\nu_i + 7}{3\nu_i}A_2^i r^2 - \frac{4(\nu_i + 1)}{4\nu_i - 5}A_3^i r^{-3} + \frac{16}{3}A_4^i r^{-5}\right] \\
\sigma_{r\varphi_i} &= \sin\theta \sin 2\varphi \mu_i\left[-2A_1^i - \frac{2\nu_i + 7}{3\nu_i}A_2^i r^2 + \frac{4(\nu_i + 1)}{4\nu_i - 5}A_3^i r^{-3} - \frac{16}{3}A_4^i r^{-5}\right] \\
\sigma_{\theta\varphi_i} &= \cos\theta \sin 2\varphi \mu_i\left[-2A_1^i + \frac{4\nu_i - 7}{3\nu_i}A_2^i r^2 - \frac{4(2\nu_i - 1)}{4\nu_i - 5}A_3^i r^{-3} + \frac{4}{3}A_4^i r^{-5}\right]
\end{aligned}
\tag{9}
$$

where ν_i is the Poisson's ratio for the ith material. From the remote boundary condition and the finite stress in the particle, we have: $A_3^1 = 0,\ A_4^1 = 0,\ A_2^4 = 0,\ A_1^4 = \tau/2\mu_4$.

For the interphase layer, the displacements are still noted by

$$
\begin{aligned}
u_{r2} &= U_{r2}(r)\sin^2\theta\cos 2\varphi \\
u_{\theta 2} &= U_{\theta 2}(r)\sin\theta\cos\theta\cos 2\varphi \\
u_{\varphi 2} &= U_{\varphi_2}(r)\sin\theta\sin 2\varphi
\end{aligned}
\tag{10}
$$

where $U_{r2} = \sum_{i=1}^4 A_i^2 r^{s_i}$; $U_{\theta 2} = \sum_{i=1}^4 A_i^2 \rho_i r^{s_i}$; $U_{\varphi 2} = -U_{\theta 2}$; the parameters ρ_i, s_i are determined by inserting Equation (10) into the equilibrium condition. From Equation (10), the corresponding stresses in the interphase layer are

$$
\begin{aligned}
\sigma_{r_2} &= \frac{2P\mu_0}{2\nu_0 - 1}\left(\frac{r}{b}\right)^{\lambda}\cos 2\varphi\sin^2\theta\sum_{i=1}^{4} r^{s_i - 1}[-s_i + (-2 + s_i + 3\rho_i)\nu_0]A_i^2 \\
\sigma_{\theta_2} &= \frac{2P\mu_0}{2\nu_0 - 1}\left(\frac{r}{b}\right)^{\lambda}\cos 2\varphi\left\{\frac{1}{2}\sum_{i=1}^{4} r^{s_i - 1}[1 + (s_i - 3\rho_i)\nu_0]A_i^2\right. \\
&\qquad \left. + \cos 2\theta\sum_{i=1}^{4} r^{s_i - 1}[-1 + 2\rho_i - (s_i + \rho_i)\nu_0]A_i^4\right\}
\end{aligned}
$$

$$\sigma_{\varphi_2} = -\frac{P\mu_0}{2\nu_0 - 1}\left(\frac{r}{b}\right)^{\lambda}\cos 2\varphi\left\{\sum_{i=1}^{4} r^{s_i-1}[1 - 3\rho_i 1 + (s_i + 3\rho_i)\nu_0]A_i^2 + \cos 2\theta\sum_{i=1}^{4} r^{s_i-1}[-1 + \rho_i - (s_i - \rho_i)\nu_0]A_i^2\right\}$$

$$\sigma_{r\theta 2} = P\mu_0\left(\frac{r}{b}\right)^{\lambda}\cos 2\varphi\sin\theta\cos\theta\sum_{i=1}^{4} r^{s_i-1}[2 + (-1 + s_i)\rho_i]A_i^2$$

$$\sigma_{r\varphi 2} = -P\mu_0\left(\frac{r}{b}\right)^{\lambda}\sin 2\varphi\sin\theta\sum_{i=1}^{4} r^{s_i-1}[2 + (-1 + s_i)\rho_i]A_i^2$$

$$\sigma_{\theta\varphi 2} = -2P\mu_0\left(\frac{r}{b}\right)^{\lambda}\sin 2\varphi\cos\theta\sum_{i=1}^{4} r^{s_i-1}s_i A_i^2 \tag{11}$$

The unknown constants $A_i^j (i = 1, 2, 3, 4; j = 1, 2, 3, 4)$ are determined by the continuity condition.

As for the case of hydrostatic stress, the average stresses in the particle, the interphase layer and the matrix are related to the applied remote stress by:

$$\overline{\sigma_{xx1}} = -\overline{\sigma_{yy1}} = 2\left(A_1^1 + \frac{7a^2}{10\nu_1}A_2^1\right)\mu_1 = c_{s1}\tau$$

$$\overline{\varepsilon_{xx1}} = -\overline{\varepsilon_{yy1}} = A_1^1 + \frac{7a^2}{10\nu_1}A_2^1 = \frac{c_{s1}}{2\mu_1}\tau = \frac{e_{s1}}{2\mu_4}\tau$$

$$\overline{\sigma_{xx2}} = -\overline{\sigma_{yy2}} = 2\mu_0\frac{P}{5(a^3 - b^3)}\sum_{i=1}^{4}\frac{(a^{2+s_i}(a/b)^{\lambda} - b^{2+s_i})(2 + s_i)(2 + 3\rho_i)}{2 + s_i + \lambda}A_i^2 = c_{s2}\tau$$

$$\overline{\varepsilon_{xx2}} = -\overline{\varepsilon_{yy2}} = \frac{1}{5(a^3 - b^3)}\sum_{i=1}^{4}(a^{2+s_i} - b^{2+s_i})(2 + 3\rho_i)A_i^2 = \frac{e_{s2}}{2\mu_4}\tau$$

$$\overline{\sigma_{xx3}} = -\overline{\sigma_{yy3}} = 2\left(A_1^3 + \frac{7(c^5 - b^5)}{5(c^3 - b^3)\nu_3}A_2^3\right)\mu_3 = c_{s3}\tau$$

$$\overline{\varepsilon_{xx3}} = -\overline{\varepsilon_{yy3}} = A_1^3 + \frac{7(c^5 - b^5)}{5(c^3 - b^3)\nu_3}A_2^3 = \frac{c_{s3}}{2\mu_3}\tau = \frac{e_{s3}}{2\mu_4}\tau \tag{12}$$

It is seen that for a pure shear macroscopic stress, the average stresses in the particle, interphase and the matrix layer are also of pure shear.

So until now, we have completely determined the local stresses and their averages over different phases for any applied remote stress. These relations will be used for calculating the effective moduli of the composite.

EFFECTIVE ELASTIC MODULI OF THE COMPOSITE

Mori–Tanaka and Generalized Self-consistent Estimates

With the previous solution, we are ready to construct the effective moduli of the composite made of particles with a graded interphase. In order to take into account the interaction with the other particles, we will follow the methodological idea introduced by Stolz and Zaoui [15]. Now choose the following pattern: a particle with a gradual interphase enclosed by a matrix layer, the volume of the particle to the total volume of the pattern is set to the volume fraction of the particle. This pattern is placed into a reference material under a remote reference stress to determine the stresses in the particle, interphase and the matrix, the relation between the average stress and strain over the pattern gives the effective properties of the composite [16]. By choosing different reference materials, different estimate can be recovered, for example, the reference material is taken to be the matrix material, this gives Mori–Tanaka's estimate [17] (illustrated in Figure 2(a)); and if the reference material is taken to be the yet unknown composite material, this leads to generalized self-consistent estimation [14,16] (illustrated in Figure 2(b)). So with the help of results presented in "Localization Problem for Spherical Particle with a Graded Interphase", we finally have the effective shear and bulk modulus of the composite as (here we assume $\kappa_4 = \kappa_3 = \kappa_0, \mu_4 = \mu_3 = \mu_0$) for Mori–Tanaka's estimate:

$$\kappa_c = \frac{f_1 c_{h1} + f_2 c_{h_2} + f_3}{f_1 e_{h1} + f_2 e_{h2} + f_3}\kappa_0, \quad \mu_c = \frac{f_1 c_{s1} + f_2 c_{s_2} + f_3}{f_1 e_{s1} + f_2 e_{s2} + f_3}\mu_0 \tag{13}$$

where f_1, f_2, f_3 are respectively the volume fraction for the particle, the interphase and the matrix, in which $f_2 = (b^3/a^3 - 1)f_1, f_3 = 1 - f_1 - f_2$.

For the generalized self-consistent estimate, we set $\kappa_4 = \kappa_c, \mu_4 = \mu_c$, and finally get the following two interactive equations that allow one to determine the composite bulk and shear moduli κ_c, μ_c.

$$\frac{f_1 c_{h1} + f_2 c_{h_2} + f_3 c_{h_3}}{f_1 e_{h1} + f_2 e_{h2} + f_3 e_{h3}} = 1, \quad \frac{f_1 c_{s1} + f_2 c_{s_2} + f_3 c_{s_3}}{f_1 e_{s1} + f_2 e_{s2} + f_3 e_{s_3}} = 1 \tag{14}$$

According to Bornert et al. [18], for the hard inclusion with hard interphase, where the matrix is the softest phase, Equation (13) in fact provides a lower bound for the effective shear and bulk modulus for the gradually coated inclusion morphological pattern, and when the reference material is chosen to be the hardest phase (we assume it to be the inclusion phase), the estimate gives the upper bounds.

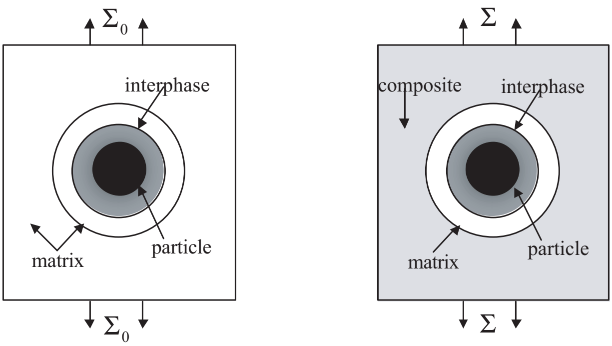

Figure 2. *(a) Concept of Mori–Tanaka method and (b) generalized self-consistent method.*

Numerical Applications

In order to illustrate the proposed method, this section will analyze the predictive capacity through some numerical examples. In the following calculation, the Young's modulus and Poisson's ratio of the matrix are $E_0 = 3.4\,\text{GPa}, \nu_0 = 0.38$, and Young's modulus and Poisson's ratio of the particle is $E_1 = 34.0\,\text{GPa}, \nu_1 = 0.2$ for a harder particle system and $E_1 = 0.34\,\text{GPa}, \nu_1 = 0.2$ for a soft one. Finally for the interphase property, we set $P = 1, \lambda = -20$ for a hard interphase and $P = 1, \lambda = 20$ for a soft interphase, we here choose $P = 1$ to assure the continuity of the material property at interface between the interphase and matrix. The thickness of the interphase, $t = 0.1a$ (a is the radius of the particle) is chosen in all the computation, this means the interphase occupies a volume fraction of one tenth of that for the particle. For comparison, the results with a uniform interphase are also included, which is defined by $f(r) = \int_a^b P(r/b)^{\lambda} dr/(b - a)$.

With the method proposed in "Localization Problem for a Spherical Particle with a Graded Interphase" the shear and the bulk moduli of the composite are estimated by Mori–Tanaka's method and generalized self-consistent method for different combinations of the particle and interphase property. Mori–Tanaka's method is adopted for the uniform interphase. Figure 3 shows the predicted results for a soft interphase, it is seen that for a soft graded interphase and soft particles, the predicted results by different methods and uniform interphase have minor difference. However for hard particles and soft interphase, the uniform interphase model largely overestimates both effective shear and bulk moduli of the composite. And for the effective bulk modulus, both Mori–Tanaka and generalized self-consistent methods coincide, and corresponds to the exact effective bulk modulus for generalized Hashin's sphere assemblage with the pattern considered.

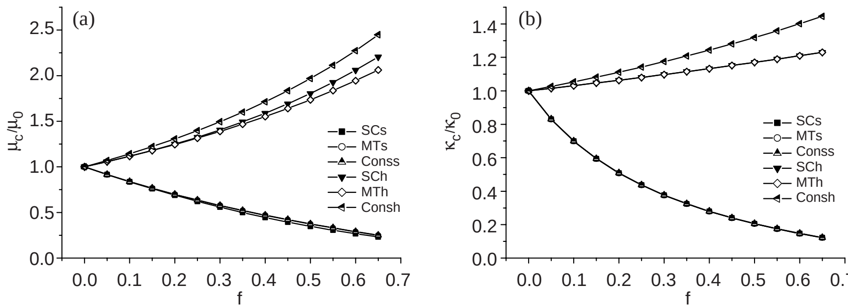

Figure 3. *Comparison of effective moduli predicted by different methods with a soft interphase. (SCs, SCh mean the estimates by generalized self-consistent method for soft and hard particle respectively; MTs, MTh mean the estimates by Mori–Tanaka's method for soft and hard particle respectively; Cons, Consh mean the estimates by Mori–Tanaka's method for a uniform interphase for soft and hard particle respectively): (a) Shear modulus; (b) Bulk modulus.*

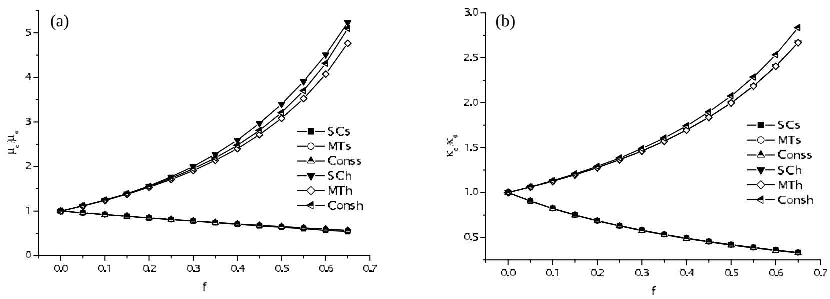

Figure 4. *Comparison of effective moduli predicted by different methods with a hard interphase: (a) Shear modulus; (b) Bulk modulus.*

The predicted effective shear and bulk moduli of the composite with a hard interphase is shown in Figure 4, as for the case of the soft interphase, there is little difference for the predicted results by different methods for a composite with soft particles; and for a composite with hard particles and hard interphase, the uniform interphase model, unlike for the soft interphase can in this case provide a reasonable estimate. Again for the effective bulk modulus, the predictions by Mori–Tanaka and generalized self-consistent methods coincide, the reason for this is explained previously.

Figure 5 illustrates the comparison of the same hard particle and matrix system predicted by generalized self-consistent method for a soft and a hard interphase respectively. Although the particle and the matrix properties remain the same, the nature of the interphase can have a significant influence on the overall behavior of the composite. For a soft interphase, the reinforcing effect of the particle is largely reduced due to the presence of a

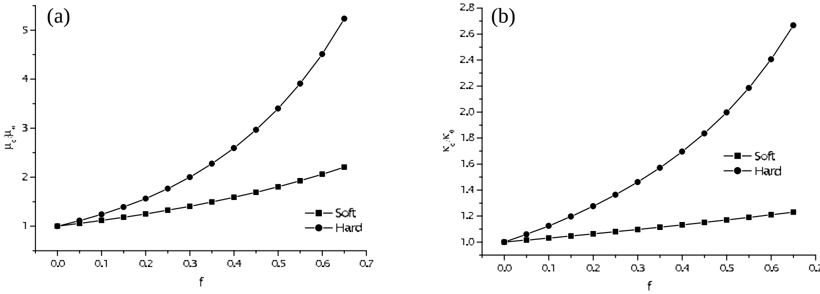

Figure 5. *Influence of the property of the interphase (hard particle and matrix, GSC method): (a) Shear modulus; (b) Bulk modulus.*

soft layer around it, the particle can even loose completely the reinforcing effect and behave like a void if the interphase is soft enough [13]. For this case, the uniform interphase model with the averaging property tends to overestimate the strengthening effect of the particle, and in turn overestimate the effective shear and bulk moduli of the composite as shown in Figure 3.

VISCOELASTIC PROPERTIES OF THE COMPOSITE

Viscoelastic Model and Dynamic Correspondence Principle

In this section, we will explore the influence of viscoelasticity of the matrix and the graded interphase on the overall properties of the composite. Here the matrix and the interphase are assumed to follow a linear isotropic viscoelastic behavior of the following integral form:

$$\sigma_{ij}(t) = \int_{-\infty}^{t} C_{ijkl}(t-\xi)\frac{d\varepsilon_{kl}(\xi)}{d\xi}d\xi \tag{15}$$

where $\sigma_{ij}(t)$, $\varepsilon_{ij}(t)$ are the components of stress and strain, $C_{ijkl}(t)$ is the relaxation function of the material. For an isotropic viscoelastic material, the above constitutive equation can be transformed to the frequency domain by using complex shear and bulk moduli

$$\tilde{s}_{ij} = 2\mu^*(\omega)\tilde{\varepsilon}_{ij}, \quad \tilde{\sigma}_{kk} = 3\kappa^*(\omega)\tilde{\varepsilon}_{kk} \tag{16}$$

$\tilde{A}$ means the corresponding quantity in the frequency domain. $\mu^*(\omega), \kappa^*(\omega)$ are the complex shear and bulk moduli, defined as

$$\mu^*(\omega) = i\omega\int_0^\infty \mu(t)e^{-i\omega t}dt, \quad \kappa^*(\omega) = i\omega\int_0^\infty \kappa(t)e^{-i\omega t}dt \tag{17}$$

In the frequency domain, there is exact correspondence between the elasticity in the real space and the linear viscoelasticity in the frequency domain, just by replacing the moduli by the complex moduli, this is usually called dynamic correspondence principle [19]. These complex moduli are typically written in terms of storage moduli (real part) and loss moduli (imagery part)

$$\kappa^*(\omega) = \kappa'(\omega) + i\kappa''(\omega)$$
$$\mu^*(\omega) = \mu'(\omega) + i\mu''(\omega) \tag{18}$$

The storage modulus characterizes the elastic response of the material, and the loss modulus measures the damping property of the material.

For the graded interphase, its relaxation bulk and shear moduli are assumed to be of the following form:

$$k_2(r,t) = f(r)k_0(t), \quad \mu_2(r,t) = f(r)\mu_0(t) \tag{19}$$

So in the frequency domain, they provide the same relations as their elastic counterparts (complex moduli replace the relaxation moduli). Here we assume the particle is elastic, so by transforming the above viscoelastic problem into the frequency domain, we get an analogous problem which has been determined previously in "Localization Problem for Spherical Particle with a Graded Interphase" and "Effective Elastic Moduli of the Composite".

Numerical Results

Figures 6 and 7 show the predicted storage and loss shear and bulk moduli by the generalized self-consistent method as a function of frequency for 20 and 40% volume fractions of hard particles, the soft and hard

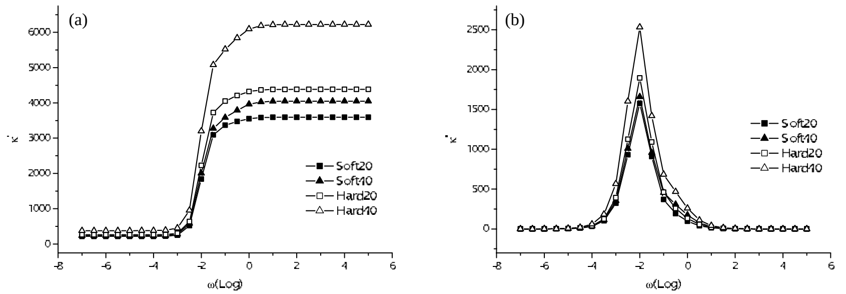

Figure 6. *Influence of interphase on viscoelastic bulk modulus of the composite with 20 and 40% hard particles (GSC estimate: soft20, soft40 mean a soft interphase with 20 and 40% particles respectively; hard20, hard40 mean a hard interphase with 20 and 40% particles respectively): (a) Storage bulk modulus; (b) Loss bulk modulus.*

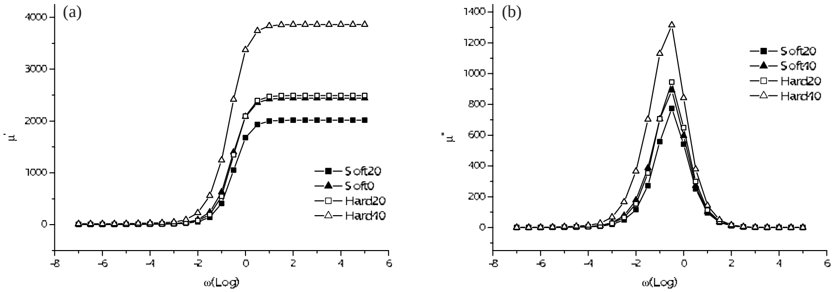

Figure 7. *Influence of interphase on viscoelastic shear modulus of the composite with 20 and 40% hard particles (GSC estimate): (a) Storage shear modulus; (b) Loss shear modulus.*

interphases are examined respectively. As for the elastic case, the type of interphase plays a significant role on the viscoelastic overall behavior for the composite, for example, for the case of composite with 40% hard particles, the predicted storage bulk and shear modulus with the soft interphase may be only one half of those with the hard interphase (the interphase thickness is still one tenth of the particle radius). It is seen from the calculation that the nature of the interphase can have a significant impact both on the elastic and viscoelastic overall properties for composite materials.

CONCLUSIONS

A micromechanical model is proposed for evaluating the effective elastic and viscoelastic properties of a particulate composite with a graded interphase in the radial direction. The localization relations for a coated particle with such an interphase in an infinite matrix under hydrostatic and pure shear loads have been derived, and the effective elastic moduli of the corresponding composite have been evaluated with the Mori–Tanaka mean field theory and the generalized self-consistent method. The dynamic moduli of such composites are also obtained by directly using the corresponding principle between linear viscoelasticity and elasticity.

The computed results show that the presence of an interphase, especially the nature of the interphase (hard or soft) has an important influence on the effective elastic and viscoelastic properties of the composite. For a composite system with hard particles and a soft graded interphase, the interphase can in this case lower significantly the stress transferred to the hard particles, then the overall moduli of the composite, and the uniform interphase model overestimates largely the moduli of the composite. Finally a careful experimental investigation for the composite made of hard particles with multiplayer coating (by varying the property of each layer) is recommended to examine the accuracy of the theoretical prediction.

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