A Generalized Self-Consistent Mori-Tanaka Scheme for Prediction of the Effective Elastic Moduli of Hybrid Multiphase Particulate Composites

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A generalized self-consistent Mori-Tanaka method (GSCMTM) is developed in this paper. The method starts with the formulation of the problem of heterogeneous elasticity proposed by Dederichs and Zeller and incorporates the strain equivalence condition of Christensen’s generalized self-consistent method (GSCM) and Hill’s interfacial operator theory into the evaluation of the strain concentration tensors of constituents. For a two-phase composite with spherical inclusions, the effective moduli predicted by GSCMTM coincide with those predicted by the Mori-Tanaka method (MTM). By a two-step homogenization technique, GSCMTM was successfully extended to multiphase composites. The effective moduli predicted by GSCMTM for multiphase hybrid particulate composites agree well with the existing theoretical and experimental results and can be expressed in a simple explicit form.

INTRODUCTION

Particulate filled polymer composites consist of particles of one material in a polymer matrix of another material. The mechanical behaviors of these composites depend upon their constituent properties and any microstructural changes. Under the influence of external load, stress concentration develops at particle interfaces. This changes the initial two-phase composite where all particles are bonded to the matrix into a three-phase composite where some particles become debonded, thereby creating voids or vacuoles. Therefore, the prediction of the effective mechanical properties of such multiphase particulate heterogeneous composites is very important for polymer modification and engineering applications.

In the past decade, several theoretical methods to tackle this class of problems have been developed. Among them are the self-consistent method (1, 2), the differential self-consistent method (3–6), the Mori-Tanaka method (6–8) and the generalized self-consistent method (three-phase sphere model) (9–11). The extensive body of literature on these methods can be found in the papers by Christensen (12), Wang and Weng (13), and Huang et al. (14). However, not all these models admit a physical description at the same level. As noted by Christensen (12), the generalized self-consistent method (GSCM) not only bears attractive capability but also yields the correct asymptotic behavior for rigid inclusions as the volume fraction of the rigid inclusion approaches 1. Accordingly, Christensen demonstrates that the preference should be given the generalized self-consistent method. However, such a classical GSCM can be suitable only for a two-phase composite. Based upon Budiansky’s energy equivalence (2) and Christensen and Lo’s three-phase sphere model (10), Huang et al. (11, 15, 16) developed a generalized self-consistent scheme that can tackle the multiphase composite problem. Nevertheless, the shear modulus estimated by GSCM cannot be expressed in an explicit form and is inconvenient to use. The differential self-consistent method (DSCM), although extended successfully to the case of wide ranges of concentration by Farris et al. (5, 17, 18), bears the same drawback as GSCM—the method cannot present an explicit solution. The Mori-Tanaka method (MTM), based upon the original work of Mori-Tanaka (19), is another method that has received considerable attention recently (6–8). Unlike most other approximate methods, which require solving implicit equations numerically, MTM yields explicit, close-form solution for the effective properties. As pointed out by Weng (6) and Benveniste (8), the knowledge of the strain or stress concentration tensor of constituents is sufficient to determine the effective moduli of composites. However, the determination of such concentration tensors is a tremendously difficult task.

Motivated by the aforementioned observations, a generalized self-consistent Mori-Tanaka method (GSCMTM) is presented by making use of the strain equivalence
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condition of GSCM, Green function techniques, and Hill's interfacial operator theory (20). Based on this scheme, the effective moduli of multiphase particulate composites are obtained and the solutions can be expressed in an explicit form. A comparison with existing experimental data and theoretical results demonstrate the GSCMTM is satisfactory.

THEORETICAL MODEL

Consider an unbounded two-phase composite medium \( V \) with randomly distributed ellipsoidal inclusions. Following Christensen and Lo's inclusion-matrix-composite configuration (10), instead of a composite spheroidal sphere a composite ellipsoidal sphere (ellipsoidal inclusion core + a surrounded matrix shell) is embedded in an infinite homogeneous medium of unknown effective properties (Fig. 1). Each phase is assumed to be homogeneous isotropically elastic and perfect bonding is considered at the interfaces. The tensor of elastic moduli of the inclusion, the matrix and the composite medium are denoted by \( L_i^I \), \( L^M \) and \( L \) respectively. In this paper, the symbol \( \Lambda \) indicates that \( \Lambda \) is a tensor, a vector, or a matrix.

When the homogeneous displacement boundary condition \( u_{j\infty} = E \bar{x} \) is imposed at infinity and a composite medium of as-yet unknown effective properties is taken as the homogeneous reference material, the strain at an arbitrary point in the composite medium can be determined by the following Lippman-Schwinger-Dyson type integral equation (21, 22).

\[
\varepsilon(x) = E - \int_V \Gamma(x-x') \delta L(x') \varepsilon(x') \, dx'
\]

where the symbol : denotes the inner product of tensors. \( E \) is the macroscopic homogeneous strain and \( x \) is the position vector, \( dx' = dx'_1 dx'_2 dx'_3 \), \( \delta L(x) \) is the fluctuating part of elastic moduli of the composite medium and defined by

\[
\delta L(x) = L(x) - \bar{L}
\]

where \( L(x) \) is the local elastic moduli tensor of the composite medium. \( \Gamma(x-x') \) is the strain Green function tensor and defined by

\[
\Gamma_{ijkl}(x-x') = \delta_{ik} \delta_{jl} \epsilon_{ij}
\]

where \( A_i \) denotes a space derivative of \( A \) with respect to \( x \) and the symbol \( \{ij\} \) and \( \{kl\} \) indicate that the

![Fig. 1. Inclusion-matrix-composite medium configuration.](POLYMER COMPOSITES, OCTOBER 1998, Vol. 19, No. 5)
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quantity has been symmetrized with respect to \( (i, j) \) and \( (k, \bar{L}) \). \( \mathcal{G} (x - x') \) is the usual displacement Green function tensor associated with the reference medium.

Hence, the average strain in the composite ellipsoidal sphere can be obtained from (1)

\[
\bar{\varepsilon}^{(2)} = \frac{1}{V_2} \int_{V_2} \int_{V_1} \Gamma (x - x'; \Delta \bar{L}^M; \bar{\varepsilon}(x')) dx' dx
\]

\[
- \frac{1}{V_2} \int_{V_2} \int_{V_1} \Gamma (x - x'; \Delta \bar{L}^M; \bar{\varepsilon}(x')) dx' dx' \tag{4}
\]

where \( V_1, V_M \) and \( V_2 = V_1 + V_M \) are the volumes occupied by the inclusion, the matrix, and the composite ellipsoidal sphere, respectively. \( \Delta \bar{L}^I \) and \( \Delta \bar{L}^M \) are defined by

\[
\Delta \bar{L}^I = \bar{L}^I - \bar{L}^M = \bar{L}^M - \bar{L} \tag{5}
\]

For an ellipsoidal body \( V_2 \), Kroner (23) proved that the integral \( \int_{V_1} \Gamma (x - x'; \Delta \bar{L}^M; \bar{\varepsilon}(x')) dx' \) is uniform. So, we define \( \bar{T} (\bar{L}) \) by

\[
\bar{T} (\bar{L}) = \int_{V_1} \Gamma (x - x'; \Delta \bar{L}^M; \bar{\varepsilon}(x')) dx' \tag{6}
\]

Then Equation 4 can be written as

\[
\bar{\varepsilon}^{(2)} = \bar{\varepsilon}^I + \frac{1}{V_2} \int_{V_2} \bar{T} (\bar{L}) : \Delta \bar{L}^M ; \bar{\varepsilon}^M \tag{7}
\]

where \( \bar{\varepsilon}^I \) and \( \bar{\varepsilon}^M \) are the average strains of the inclusion and the matrix respectively, which are defined by

\[
\bar{\varepsilon}^I = \frac{1}{V_1} \int_{V_1} \bar{\varepsilon}(x; \Delta \bar{L}^I) dx \quad \bar{\varepsilon}^M = \frac{1}{V_M} \int_{V_M} \bar{\varepsilon}(x; \Delta \bar{L}^M) dx \tag{8}
\]

It may be noticed from the Christensen and Lo’s (10) energy condition for the three-phase model that the strain energy stored in the composite sphere being the same as that in the composite medium under the same homogeneous displacement boundary condition is still valid for the present case. After some mathematical manipulations, Christensen and Lo’s energy condition can be turned into the following strain-equivalence condition (24):

\[
\bar{\varepsilon}^{(2)} = \bar{\varepsilon}^I \tag{9}
\]

This means that Christensen’s energy condition for the three-phase model is equivalent to the requirement that the average strain of the composite ellipsoidal sphere be the same as the macroscopic homogeneous strain imposed on the composite medium at infinity.

Applying this strain equivalence condition to Equation 7, we obtain the following general equation:

\[
\frac{V_1}{V_2} \bar{T} (\bar{L}) : \Delta \bar{L}^I \bar{\varepsilon}^I + \frac{V_M}{V_2} \bar{T} (\bar{L}) : \Delta \bar{L}^M \bar{\varepsilon}^M = 0 \tag{10}
\]

Apparently, the effective elastic moduli of the composite material \( \bar{L} \) can be determined from this general equation only if \( \bar{\varepsilon}^I \) and \( \bar{\varepsilon}^M \) or the relation between \( \bar{\varepsilon}^I \) and \( \bar{\varepsilon}^M \) are known. It should be emphasized that Equation 10 is an exact representation of strain equivalence or energy equivalence and is valid for arbitrary ellipsoidal type inclusion including crack inclusion (a limit case of the ellipsoidal inclusion). Therefore, this scheme is an extension of Christensen and Lo’s three-phase model to a certain extent.

We shall assume that the strains are uniform in inclusion. The uniformity of the strains in an inclusion embedded in an infinite space has been demonstrated by Eshelby (25). For a high concentration of inclusions, the hypothesis of strain uniformity certainly fails, but since we are mostly interested by the evaluation of the average strains in the different phases, it remains an interesting working assumption. That hypothesis will be testified in the end of this section to be equivalent to Mori-Tanaka average field approximation. Now according to Hill’s interfacial operator theory (20), there is a strain jump relation across the interface:

\[
\bar{\varepsilon}^M (x') = \bar{\varepsilon}^I + (\mathcal{T} (\bar{L}) - \mathcal{Q} (\bar{L}, \bar{x}')) : \Delta \bar{L}^M ; \bar{\varepsilon}^I \tag{11}
\]

where \( \Delta \bar{L}^M = \bar{L}^I - \bar{L}^M \), \( x' \) is located at the side of the matrix. \( \mathcal{Q} (\bar{L}, \bar{x}') \) is defined by

\[
\mathcal{Q} (\bar{L}, \bar{x}') = \int_{V_2} \Gamma (x^* - x'; \Delta \bar{L}^M; \bar{\varepsilon}(x')) dx' \tag{12}
\]

where \( \Gamma (x^* - x') \) is the strain Green function tensor associated with the matrix material. Hence, according to Equation 11, the average strain of the matrix is written as

\[
\bar{\varepsilon}^M = \bar{\varepsilon}^I + \frac{1}{V_M} \int_{V_M} (\mathcal{T} (\bar{L}) - \mathcal{Q} (\bar{L}, \bar{x}')) : \Delta \bar{L}^M ; \bar{\varepsilon}^I dx' \tag{13}
\]

If \( V_1, V_2 \) are two self-similar ellipsoidal bodies, it is easily proved

\[
\int_{V_2} \mathcal{Q} (\bar{L}, \bar{x}^*) dx^* = \int_{V_2} \int_{V_1} \Gamma (x^* - x') dx' dx^* = 0 \tag{14}
\]

Therefore, Equation 13 can be written as

\[
\bar{\varepsilon}^M = \bar{\varepsilon}^I \tag{15}
\]

where \( \bar{H} \) is defined by

\[
\bar{H} = I + \bar{T} (\bar{L}) : \Delta \bar{L}^M \tag{16}
\]

So, the effective elastic moduli \( \bar{L} \) of composites can be obtained by combining Equations 10 and 15. For an isotropic spherical inclusion problem, \( \bar{L} = (3k, 2\mu) \) and \( \bar{T} (\bar{L}) \) is defined by

\[
T (\bar{L}) = \frac{1}{(3k + 4\mu)} \delta_{ij} \delta_{kl} + \frac{3(k + 2\mu)}{10\mu(3k + 4\mu)} \left( \delta_{ik} \delta_{jl} + \delta_{ij} \delta_{kl} - \frac{2}{3} \delta_{ij} \delta_{kl} \right) \tag{17}
\]

and can be simply expressed as

\[
T (\bar{L}) = \left( \frac{1}{3k^*} - \frac{1}{2\mu^*} \right) \tag{18}
\]

where

\[
k^* = \frac{1}{3} (3k + 4\mu) \quad \mu^* = \frac{5}{6} \frac{\mu(3k + 4\mu)}{k + 2\mu} \tag{19}
\]
where $k$, $\mu$ are the bulk and shear moduli of material respectively.

Now, introduce McLaughlin's tensor operation rule (3). For two arbitrary tensors $\bar{A} = (3a_1, 2a_2)$ and $\bar{B} = (3b_1, 2b_2)$ reads

\[
\begin{align*}
A^{-1} & = (1/3a_1, 1/2a_2) \\
\bar{A} + \bar{B} & = (3(a_1 + b_1), 2(a_2 + b_2)) \\
\bar{A} : \bar{B} & = (9a_1b_1, 4a_2b_2)
\end{align*}
\]

Accordingly, Equation 16 can be written as

\[
\tilde{H} = (h_k, h_\mu)
\]

where

\[
\begin{align*}
h_k & = 1 + \frac{3(k - k_M)}{3k_M + 4\mu_M} \\
h_\mu & = 1 + \frac{6(\mu - \mu_M)(k_M + 2\mu_M)}{5\mu_M(3k_M + 4\mu_M)}
\end{align*}
\]

Combining Equation 10 with 15 as well as 21 and making use of the aforementioned McLaughlin's tensor operation rule, we obtain the effective bulk and shear moduli of a two-phase spherical inclusion based composite

\[
\frac{\bar{k}}{k_M} = 1 + \frac{f_i(k_i/k_M - 1)}{f_i + (1-f_i)h_k}
\]

\[
\frac{\bar{\mu}}{\mu_M} = 1 + \frac{f_i(\mu_i/\mu_M - 1)}{f_i + (1-f_i)h_\mu}
\]

It is interesting to find this result is exactly the same as that of MTM (6), and $H^{-1}$ is actually equal to the strain concentration tensor $\bar{G}$ adopted in the reformulation of MTM derived by Benveniste (8). This means that the assumption that the strain is uniform in the inclusion is equivalent to Mori-Tanaka average field approximation. However, it is emphasized that present scheme is different from both MTM, which is based upon the Eshelby's equivalent inclusion idea (25), and GSCM (10), which needs to solve complicated strain fields of constituents. Furthermore, the present scheme can be extended easily to a multiphase hybrid or coated inclusion problem. Since there exist some connections with GSCM and MTM, we call the present method the generalized self-consistent Mori-Tanaka method (GSCMTM).

**APPLICATIONS TO MULTIPHASE COMPOSITES**

In this section, we evaluate the effective elastic moduli of a particle-void hybrid multiphase composite by GSCMTM. Following the idea of the GSCM (10), such a multiphase composite system (Fig. 2) can be modeled by a three-phase configuration: particle-equivalent matrix (voids + matrix) - homogeneous

**Fig. 2. Particle-void hybrid multiphase composite.**
composite medium (Fig. 3). Here, each phase is assumed to be isotropic and perfect bonding at interfaces is considered. \( L_p, L_v, L_M, L_{EM} \) and \( L \) are the elastic moduli tensor of the particle, the void, the matrix, the equivalent matrix and the composite medium respectively. \( V_p, V_v, V_M \) and \( V_{EM} \) are the volume occupied, respectively, by the particle, the void, the matrix, and the equivalent matrix.

In order to predict the effective moduli of the composite, a two-step homogenization technique is adopted. First, the voids and the matrix are homogenized as a homogeneous equivalent matrix, the effective properties of which are determined by GSCMTM based upon a three-phase configuration: voids – matrix – equivalent matrix. Next, the particles and the equivalent matrix are homogenized as a homogeneous composite medium, the effective properties of which are also determined by GSCMTM based upon a new three-phase configuration: particle – equivalent matrix – composite medium.

According to the scheme derived in the last section, the similar general equation is obtained

\[
f_P T(L_p) \cdot \Delta L_p \cdot \varepsilon_p + f_{EM} T(L_{EM}) \cdot \Delta L_{EM} \cdot \varepsilon_{EM} = 0
\]  

(25)

where \( f_p = V_p/V_0, f_{EM} = V_{EM}/V_3 \) are the volume fraction of the particle and the equivalent matrix respectively, \( V_{EM} = V_v + V_M \) and \( V_3 = V_p + V_v + V_M \). \( \Delta L_p = L_p - L, \Delta L_{EM} = L_{EM} - L \), the relation between the average strain of the equivalent matrix \( \varepsilon_{EM} \) and that of the particle \( \varepsilon_p \) is similar to Equation 15 and can be expressed as follows

\[
\varepsilon_{EM} = P \cdot \varepsilon_p
\]  

(26)

where

\[
\varepsilon_{EM} = \frac{f_v}{f_v + f_M} \varepsilon_v + \frac{f_M}{f_v + f_M} \varepsilon_M
\]  

(27)

\[
P = I + T(L_{EM}) \cdot \Delta L_{EM} = (p_L, p_v)
\]  

(28)

where \( f_v = V_v/V_3, f_M = V_M/V_3 \) are the volume fraction of the void and the matrix respectively. \( \Delta L_{EM} = L_p - L_{EM} \). The effective moduli of the equivalent matrix \( L_{EM} \equiv (3k_{EM}, 2\mu_{EM}) \) can be determined by the first homog-
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Fig. 4. Effective shear modulus for a three-phase composite containing spherical voids and rigid spheres.

\[ k_{EM} = 1 + \frac{f_v(k_v/k_M - 1)}{f_v + f_d h_v} \]  \hspace{1cm} (29)

\[ \mu_{EM} = 1 + \frac{f_v(\mu_v/\mu_M - 1)}{f_v + f_d h_v} \]  \hspace{1cm} (30)

where \( h_v, h_p \) are given by Equation 22 with \( k_v, \mu_v \) replaced by \( k_p, \mu_p \), respectively.

Similarly, introduce McLaughlin's tensor operation rule defined in Equation 20, from Equation 28 we obtain

\[ p_k = 1 + \frac{3(k_p - k_M)}{3k_M + 4\mu_M} \]  \( p_p = 1 + \frac{6(\mu_p - \mu_M)(k_M + 2\mu_M)}{5\mu_M(3k_M + 4\mu_M)} \]  \hspace{1cm} (31)

Accordingly, the effective bulk and shear moduli of multiphase hybrid composite can be determined by the second homogenization step based upon a new three-phase configuration: particle – equivalent matrix – composite medium and can be written in the following simple explicit forms

\[ \frac{k}{k_{EM}} = 1 + \frac{f_p(k_p/k_{EM} - 1)}{f_p + (1 - f_p)p_k} \]  \hspace{1cm} (32)

\[ \frac{\mu}{\mu_{EM}} = 1 + \frac{f_p(\mu_p/\mu_{EM} - 1)}{f_p + (1 - f_p)p_p} \]  \hspace{1cm} (33)

It is easily found that Equations 32 and 33 reduce, respectively, to the corresponding Equations 23 and 24 for a two-phase composite if \( f_v = 0 \).

In order to test the accuracy of GSCMTM, the effective moduli for particle and void hybrid multiphase composites predicted by Equations 32 and 33 are compared with those obtained by the existing methods and experiment. As a first example, we consider a three-phase composite with rigid particles and voids. In a recent work, Molinari and Mouden [26] estimated the effective shear moduli of such a composite by their cluster method. If the cluster contains a single inclusion, the results are identical to those predicted by MTM. For a large cluster size, the results are significantly improved by their proposed method, which better accounts for the particle interactions than

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Fig. 5. Effective Young's modulus for a particle-void hybrid composite.

MTM. The effective shear moduli predicted by present GSCMTM and Molinari and Mouden's cluster method are shown in Fig. 4. It is seen from Fig. 4 that the results from these two schemes are very close.

Next, we compare our predictions with those obtained using MTM for multiphase composites by Weng (6) and using GSCM by Huang et al. (11). Experimental data for a three-phase composite, made of quartz-sand fillers and voids within an epoxy matrix, were obtained by Cohen and Ishai (27). The material properties are: matrix (epoxy) with \( E_M = 2.03 \text{ GPa} \) and \( v_M = 0.4 \); First phase (quartz-sand) with \( E_p = 73.6 \text{ GPa} \) and \( v_p = 0.25 \); and second phase (void) with \( E_v = 0 \). The volume fraction of quartz-sand \( f_p \) is related to the void volume fraction \( f_v \) such that \( f_p = 0.173 (1 - f_v) \). The Young's moduli of the composite are calculated with a constant filler-to-matrix weight ratio of 0.5. Figure 5 shows that the predictions given by making use of the present GSCMTM, MTM (6), and GSCM (11) agree well with the experimental data (27). The solutions from the present approach are very close to those from GSCM and still present very good agreement with the experimental data even for higher void volume fractions, while MTM's predictions have a slight deviation from the experimental data. This example demonstrates that the current scheme makes an accurate prediction of the effective moduli of multiphase hybrid composites.

**SUMMARY AND CONCLUSIONS**

Based upon the strain equivalence condition of Christensen's generalized self-consistent method (GSCM) and Hill's interfacial operator theory, a generalized self-consistent Mori-Tanaka method (GSCMTM) was presented. For a two-phase composite with spherical inclusions, the solutions of GSCMTM coincide with those of MTM. By making use of a two-step homogenization technique, the effective moduli of multiphase hybrid particulate composites are obtained and can be expressed in a simple explicit form. A comparison with the existing methods and experimental results demonstrate that GSCMTM is satisfactory.

The difficulty in extending the current scheme to materials with imperfect interfaces, including interface sliding and partly debonding, lies in the evaluation of the strain concentration tensors of constituents. Study of this problem will constitute a part of our future endeavors.
ACKNOWLEDGMENT

The authors gratefully acknowledge the financial support of this research by the National Natural Science Foundation of China projects by 19572008 & 19632030 and China Postdoctoral Science Foundation.

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