PREDICTION OF INTERPHASE PROPERTIES OF A THREE-PHASE COMPOSITE USING THREE-PHASE AND FOUR-PHASE COMPOSITE MODELS

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ABSTRACT

Here the composite material is assumed to consist of three homogeneous, isotropic elastic phases, namely spherical inclusions, interphase and matrix. In this study, methods to predict the elastic properties of the interphase are presented. The effective properties of this composite can be calculated exactly using the four-phase composite sphere model, and also by applying the less complicated three-phase composite sphere model twice, i.e., the two-step method. Both methods are used in this study, where contour plots of the effective Young’s modulus and effective Poisson’s ratio of the composite is obtained. From this contour plot the elastic properties of the interphase can be predicted when the properties of the inclusion, matrix and composite are known. Numerical results are presented and it is found that for concrete, the two-step-method yields results which are sufficiently close to the results obtained from the four-phase composite sphere model. The validity of the two-step method is discussed.

Keywords: Composite, Effective properties, Elastic moduli, Four-phase model, Three-phase model.

INTRODUCTION

For a composite material if the properties of all constituents i.e. Young’s modulus, Poisson’s ratio, shape of inclusions, arrangement and volume fractions are known, then the effective properties of the composite may be predicted. In practice, the effective properties of composites can be measured from experiments, while for some composite materials the properties of constituents are difficult to measure e.g. cement-based material.

In the case of elastic composite material composed of spherical inclusions, the three-phase composite sphere model for obtaining the effective bulk modulus of a two-phase composite material was first presented by Hashin (1962). The quadratic equation to calculate the effective elastic shear modulus of the same model was obtained by Christensen and Lo (1979). The generalization of the three-phase composite sphere model to the multi-layer spherical inclusion was presented by Herve and Zaoui (1993). The four-phase composite sphere model was used to...
study cement-based material by Ramesh et al. (1996). The inverse method related to four-phase composite sphere model was presented by Hashin and Monteiro (2002) in obtaining the properties of the interphase between aggregate and cement paste.

In this paper two schemes to determine the elastic constants of interphase are presented, the first method is by using the four-phase composite sphere model and the second method is by using the less complicated three-phase composite sphere model twice, in succession (two-step method). The numerical results from the two methods are presented and compared. In the first numerical example by using the available experimental data for concrete, aggregate and cement mortar, the elastic properties of the interphase (i.e., interfacial transition zone; ITZ) are obtained. In the second example a composite with very stiff inclusions is considered. It is shown in both examples that the bulk modulus is the same for both methods while the other elastic moduli are different, which agrees with the analytical results. The validity of the two-step method is discussed. The contour plots presented in this paper are useful in the optimization problem.

THREE-PHASE COMPOSITE SPHERE MODEL

In calculating the effective properties of a two-phase composite material, which consists of spherical inclusions embedded in a homogeneous matrix material (Fig. 1), the three-phase composite sphere model is used. The model consists of a two-phase composite sphere surrounded by an equivalent homogeneous infinite medium as shown in Fig. 2. Each phase is homogeneous and isotropic elastic, and perfect bonding is assumed at each interface. The derivation of the effective bulk modulus was presented by Hashin (1962), while Christensen and Lo (1979) obtained the quadratic equation to calculate the effective shear modulus.

Basic equations

The equilibrium equation when there are no body forces is

\[ \nabla \cdot \sigma = 0, \]

(1)

where \( \sigma \) is the symmetric stress tensor and \( \nabla \cdot \sigma \) is the divergence of \( \sigma \).

The infinitesimal strain tensor \( \varepsilon \) is related to the displacement vector \( u \) by
\[ \varepsilon = \frac{1}{2} [\nabla \otimes \mathbf{u} + (\nabla \otimes \mathbf{u})^T], \tag{2} \]

where \( \nabla \otimes \mathbf{u} \) is the gradient of \( \mathbf{u} \) and superscript \( ^T \) denotes the transpose of a tensor.

The stress and strain tensors are split into isotropic and deviatoric parts as

\[ \sigma = \frac{1}{3} \text{tr}(\sigma) \mathbf{I} + \mathbf{s}, \quad \varepsilon = \frac{1}{3} \text{tr}(\varepsilon) \mathbf{I} + \mathbf{e}, \tag{3} \]

where “tr” is the trace of a tensor, \( \mathbf{I} \) is the second order identity tensor, \( \mathbf{s} \) and \( \mathbf{e} \) are the deviatoric stress and strain tensors, respectively. For phase \( (i) \) Hooke’s law can be expressed as

\[ \text{tr}(\sigma^{(i)}) = 3K_i \text{tr}(\varepsilon^{(i)}), \quad \mathbf{s}^{(i)} = 2\mu_i \mathbf{e}^{(i)}, \tag{4} \]

where \( K_i \) is bulk modulus and \( \mu_i \) is shear modulus. The spherical coordinate system \((R, \psi, \theta)\) is used in the following analysis.

**Effective bulk modulus**

The displacement field for phase \( (i) \) under uniform dilatation or uniform pressure at infinity can be written as (Love, 1944)

\[ u_R^{(i)}(R) = F_i R + G_i R^{-2}, \quad u_\psi^{(i)}(R) = 0, \tag{5} \]

where \( F_i \) and \( G_i \) are arbitrary constants. For the three-phase sphere model \( i = 1, 2, 3 \). From Eqs. (2)-(5) the expressions for stresses are

\[ \sigma_{RR}^{(i)} = 3K_i F_i - 4\mu_i G_i R^{-3}, \quad \sigma_{\psi\psi}^{(i)} = \sigma_{\theta\theta}^{(i)} = 3K_i F_i + 2\mu_i G_i R^{-3}, \quad \sigma_{R\psi}^{(i)} = \sigma_{R\theta}^{(i)} = \sigma_{\psi\theta}^{(i)} = 0. \tag{6} \]

The non-singular stress condition at the origin and the continuity conditions at the interfaces are

\[ \sigma^{(i)}(0) \quad \text{should be finite,} \quad u^{(j)}(R_j) = u^{(i+1)}(R_j), \quad \mathbf{e}_R^{(j)}(R_j) = \mathbf{e}_R^{(i+1)}(R_j), \tag{7} \]

where \( \mathbf{e}_R \) is the base vector in the radius direction \( R \). Substituting Eqs. (5)-(7) (for \( i = 1, 2, 3 ; j = 1, 2 \)) into Eq. (A.3) the effective bulk modulus \( K_e \) is obtained as

\[ K_e = K_1 = K_2 + \frac{f_1(K_1 - K_2)}{1 + (1 - f_1)[(K_1 - K_2)/(K_2 + 4\mu_2/3)]}, \tag{8} \]

where \( f_1 = R_1^3 / R_2^3 \) is the volume fraction of the inclusion.

**Effective shear modulus**

The displacement field for phase \( (i) \) under simple shear deformation can be written as (Love,
where \( v_i \) is Poisson’s ratio and \( A_i, B_i, C_i \) and \( D_i \) are arbitrary constants \((i = 1, 2, 3)\). From Eqs. (2)-(4) and (9) the relevant stresses are expressed as

\[
\begin{align*}
\sigma_{rr}^{(i)} &= 2(A_i + \frac{3v_i}{1-2v_i}B_i R^2 - 12C_i R^{-5} - 2\frac{5-v_i}{1-2v_i}D_i R^{-3}) \mu_i \sin^2 \psi \cos 2\theta, \\
\sigma_{r\theta}^{(i)} &= (A_i - \frac{7+2v_i}{1-2v_i}B_i R^2 + 8C_i R^{-5} + 2\frac{1+v_i}{1-2v_i}D_i R^{-3}) \mu_i \sin 2\psi \cos 2\theta, \\
\sigma_{r\phi}^{(i)} &= -2(A_i - \frac{7+2v_i}{1-2v_i}B_i R^2 + 8C_i R^{-5} + 2\frac{1+v_i}{1-2v_i}D_i R^{-3}) \mu_i \sin \psi \sin 2\theta.
\end{align*}
\]

Substituting Eqs. (7), (9)-(10) (for \( i = 1, 2, 3; j = 1, 2 \)) into Eq. (A.3) and after some manipulation the effective shear modulus \( \mu_e = \mu_3 \) can be obtained from

\[
a(\mu_e / \mu_2)^2 + 2b(\mu_e / \mu_2) + c = 0,
\]

where

\[
a = 8(\bar{\mu}_i - 1)(4 - 5v_i) \eta_i f_i^{10/3} - 2 [63(\bar{\mu}_i - 1) \eta_i + 2\eta_i \eta_2] f_i^{7/3} + 252(\bar{\mu}_i - 1) \eta_i f_i^{5/3} - 50(\bar{\mu}_i - 1)(7 - 12v_i + 8v_i^2) \eta_i f_i + 4(7 - 10v_i) \eta_i, \\
b = -2(\bar{\mu}_i - 1)(1 - 5v_i) \eta_i f_i^{10/3} + 2 [63(\bar{\mu}_i - 1) \eta_i + 2\eta_i \eta_1] f_i^{7/3} - 252(\bar{\mu}_i - 1) \eta_i f_i^{5/3} + 75(\bar{\mu}_i - 1)(3 - v_i) \eta_i f_i + 3 \eta_i f_i^{3/3}, \\
c = 4(\bar{\mu}_i - 1)(5v_i - 7) \eta_i f_i^{10/3} - 2 [63(\bar{\mu}_i - 1) \eta_i + 2\eta_i \eta_1] f_i^{7/3} + 252(\bar{\mu}_i - 1) \eta_i f_i^{5/3} + 25(\bar{\mu}_i - 1)(v_i^2 - 7) \eta_i f_i^{3/3} + (7 + 5v_i) \eta_i f_i, \\
\eta_1 = (49 - 50v_i v_2)(\bar{\mu}_i - 1) + 35(\bar{\mu}_i - 1)(v_i - 2v_2) + 35(2v_i - v_2), \\
\eta_2 = 5v_i(\bar{\mu}_i - 8) + 7(\bar{\mu}_i + 4), \quad \eta_3 = \bar{\mu}_i (8 - 10v_i) + (7 - 5v_i), \quad \bar{\mu}_i = \mu_i / \mu_2.
\]

**FOUR-PHASE COMPOSITE SPHERE MODEL**

The multi-phase composite sphere model presented by Herve and Zaoui (1993) is a generalization of the three-phase composite sphere model. The four-phase composite sphere model (Ramesh et al., 1996) is a special case of the multi-phase composite sphere model. The
composite consists of three phases namely, spherical inclusions, interphase and the matrix material as shown in Fig. 3. Each phase is homogeneous and isotropic elastic and perfect bonding is assumed at each interface. The model consists of a three-phase composite sphere surrounded by an equivalent homogeneous infinite medium as shown in Fig. 4. The basic equations to analyze this model are the same as given in Section 2.

Effective bulk modulus

Substituting Eqs. (5)-(7) (for \( i = 1,..4; j = 1,2,3 \)) into Eq. (A.3) the effective bulk modulus \( K_{eff} \) is obtained in recursive form as

\[
K_{eff} = K_4 = K_3 + \frac{(f_1 + f_2)}{1/(K_e - K_3) + (1 - f_1 - f_2)/(K_3 + 4\mu_3 / 3)},
\]

(13)

where

\[
K_e = K_2 + \frac{\overline{f_1}(K_1 - K_2)}{1+(1-\overline{f_1})[(K_1 - K_2)/(K_2 + 4\mu_2 / 3)]},
\]

(14)

\( \overline{f_1} = f_1/(f_1 + f_2) = R_1^3 / R_2^3 \) is a relative volume fraction, \( f_1 \) and \( f_2 \) are the volume fractions of inclusion and interphase phases, respectively.

Effective shear modulus

Substituting Eqs. (7), (9)-(10) (for \( i = 1,..4; j = 1,2,3 \)) into Eq. (A.3) and after some manipulation the effective shear modulus \( \mu_{eff} = \mu_4 \) can be obtained from

\[
4d_{12}^{(3)} (\mu_{eff} / \mu_3)^2 + (d_{23}^{(3)} - 2d_{13}^{(3)} + 3d_{24}^{(3)}) (\mu_{eff} / \mu_3) - d_{34}^{(3)} = 0,
\]

(15)

where
\[ d_{ij}^{(3)} = p_{1j}^{(3)} p_{2i}^{(3)} - p_{ij}^{(3)} p_{2i}^{(3)}, \]  

\[ \mathbf{p}^{(n)} = \mathbf{p}^{(n-1)} \mathbf{s}^{(n)}, \quad \mathbf{p}^{(1)} = \begin{bmatrix} 1 - 2\nu_1 & 1 - 2\nu_1 & 1 - 2\nu_1 \\ -6\nu_1 & -7 + 4\nu_1 & 3\nu_1 \\ -7 - 2\nu_1 \end{bmatrix}, \]

\[ \mathbf{s}^{(n)} = \begin{bmatrix} -2(5 - \nu_n) & 24 f_{n-1}^{-2/3} & -2\nu_n & -14 f_{n-1}^{-1} \\ 3(1 + \nu_n) & -24 f_{n-1}^{-2/3} & 7 + 2\nu_n & -21 f_{n-1}^{-1} \\ -5 - 4\nu_n & 6\nu_{n-1} f_{n-1}^{-2/3} & -4\nu_n f_{n-1}^{-1} & 14 f_{n-1}^{-1} f_{n-1} \\ -3 - 6\nu_n & 6\nu_{n-1} f_{n-1}^{-2/3} & -7 - 4\nu_n f_{n-1}^{-1} & 21 f_{n-1}^{-1} f_{n-1} \end{bmatrix}, \]

and \[ f_{n-1} = \frac{\sum_{i=1}^{n-1} f_i}{\sum_{i=1}^{n} f_i}, \quad \bar{\mu}_{n-1} = \mu_{n-1}/\mu_n. \]

**TWO-STEP METHOD**

It is possible to use a two-step procedure to calculate the effective bulk modulus \( \hat{K}_{\text{eff}} \) and the effective shear modulus \( \hat{\mu}_{\text{eff}} \) of a three-phase composite.

Step 1: The three-phase composite model is used for medium “1” and “2” and the effective properties \( K_e \) and \( \mu_e \) are determined.

Step 2: The three-phase composite model is used for the equivalent homogeneous medium of step 1 and phase “3” and the effective properties \( \hat{K}_{\text{eff}} \) and \( \hat{\mu}_{\text{eff}} \) are calculated. It is found that \( \hat{K}_{\text{eff}} = K_{\text{eff}} \) while \( \hat{\mu}_{\text{eff}} \neq \mu_{\text{eff}} \).

A similar method was applied for composites with coated fibers by Sutcu (1992).

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**Fig. 5.** The two-step method to calculate effective properties.
NUMERICAL RESULTS

The effective properties of the three-phase composite is determined by using the four-phase
composite sphere model, and the three-phase composite sphere model with the two-step method.
Since each phase is assumed to be homogeneous and isotropic elastic, the bulk modulus and
shear modulus are determined from Young’s modulus and Poisson’s ratio from

\[ K_i = \frac{E_i}{3(1-2\nu_i)}, \quad \mu_i = \frac{E_i}{2(1+\nu_i)}, \quad (i = 1, 2, 3). \]  (19)

Example 1

The three-phase composite considered is concrete, which consists of gravel as coarse
aggregate, interfacial transition zone (ITZ) and mortar. The elastic constants of gravel and
mortar are given in Table 1, when \( E_i, \nu_i \) \( (i = 1, 3) \) are taken from Ramesh et al. (1996) who
assumed values for \( \nu_i \) and obtained \( E_i \) from Hirsch (1962). Shear and bulk modulus are
calculated from Eq. (19).

<table>
<thead>
<tr>
<th>Inclusion (gravel)</th>
<th>( E_1 = 59.6 ) GPa</th>
<th>( \nu_1 = 0.15 )</th>
<th>( K_1 = 28.38 ) GPa</th>
<th>( \mu_1 = 25.91 ) GPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>Interphase (ITZ)</td>
<td>( E_2 = 15.0 ) GPa</td>
<td>( \nu_2 = 0.20 )</td>
<td>( K_2 = 8.33 ) GPa</td>
<td>( \mu_2 = 6.25 ) GPa</td>
</tr>
<tr>
<td>Matrix (mortar)</td>
<td>( E_3 = 19.17 ) GPa</td>
<td>( \nu_3 = 0.26 )</td>
<td>( K_3 = 13.31 ) GPa</td>
<td>( \mu_3 = 7.61 ) GPa</td>
</tr>
</tbody>
</table>

* these values are assumed only for the results presented in Fig. 6

In Fig. 6, assuming the properties of ITZ as \( E_2 = 15.0 \) (GPa) and \( \nu_2 = 0.20 \) for all possible
ranges of volume fractions i.e., \( 0 \leq f_1 + f_2 \leq 1 \) and \( 0 \leq f_2/(f_1 + f_2) \leq 1 \), the results from
the four-phase sphere model and the difference between the results using both methods are plotted.
It is seen that for concrete the difference between the results of these two methods are less than
0.5% of the moduli obtained using the four-phase model. In addition, the effective bulk
modulus of both methods \( K_{eff} \) and \( \tilde{K}_{eff} \) are the same.

Next, the contour plots of effective Young’s modulus and Poisson’s ratio obtained from both
methods for some combinations of volume fraction of constituents \( (f_1, f_2) = (0.4, 0.05),
(0.4, 0.1), \) \( (0.5, 0.05) \) and \( (0.5, 0.1) \) with the properties of ITZ as \( 5 \leq E_2 \leq 50 \) (GPa) and
\( 0 \leq \nu_2 \leq 0.4 \) are presented in Fig. 7. From the contour plots it is found that the effective
Young’s modulus \( E_{eff} \) is not affected too much by \( \nu_2 \). The measured Young’s modulus of this
concrete reported by Hirsch (1962) for volume fraction of coarse aggregate \( f_1 = 0.4 \) is
\( E_{con} = 29.7 \) (GPa) and for \( f_1 = 0.5 \) is \( E_{con} = 32.5 \) (GPa). From which a narrow range for \( E_2 \)
of ITZ may be determined as shown in Table 2. If Poisson’s ratio for concrete was measured
and available, rather than a narrow range for \( E_2 \), both \( E_2 \) and \( \nu_2 \) can be determined.
Fig. 6. The effective properties for Example 1; $\Delta E_{\text{eff}} = \hat{E}_{\text{eff}} - E_{\text{eff}}$; $\Delta v_{\text{eff}} = \hat{v}_{\text{eff}} - v_{\text{eff}}$; $\Delta \mu_{\text{eff}} = \hat{\mu}_{\text{eff}} - \mu_{\text{eff}}$; $\Delta K_{\text{eff}} = \hat{K}_{\text{eff}} - K_{\text{eff}} = 0$. 
**Fig. 7.** The contour plot of effective Young’s modulus and Poisson’s ratio of Example 1.

**Table 2 Estimated Young’s modulus of ITZ for Example 1 when 0<ν<0.4**

<table>
<thead>
<tr>
<th>$f_1 = 0.4, E_{\text{conc}} = 29.7$ GPa</th>
<th>$f_1 = 0.5, E_{\text{conc}} = 32.5$ GPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_2 = 0.05$</td>
<td>$f_2 = 0.1$</td>
</tr>
<tr>
<td>21.70 ≤ $E_2$ ≤ 23.38</td>
<td>19.72 ≤ $E_2$ ≤ 21.46</td>
</tr>
<tr>
<td>21.78 ≤ $\hat{E}_2$ ≤ 23.46</td>
<td>19.52 ≤ $\hat{E}_2$ ≤ 21.35</td>
</tr>
</tbody>
</table>

Note: $E_2$ and $\hat{E}_2$ are obtained by using four-phase model and two-step method respectively.
Fig. 8. The effective properties for Example 2; \( \Delta E_{\text{eff}} = \hat{E}_{\text{eff}} - E_{\text{eff}} \); 
\( \Delta \nu_{\text{eff}} = \hat{\nu}_{\text{eff}} - \nu_{\text{eff}} \); \( \Delta \mu_{\text{eff}} = \hat{\mu}_{\text{eff}} - \mu_{\text{eff}} \); \( \Delta K_{\text{eff}} = \hat{K}_{\text{eff}} - K_{\text{eff}} = 0 \).
Example 2

To investigate under what conditions the 4-phase composite model and the two-step method give widely different answers a composite with a very stiff inclusion was considered. The extreme combination of constituent materials assumed is shown in Table 3. Figure 8 shows the contour plot of the effective moduli and the difference between the results obtained from these two methods for $0 \leq f_1 + f_2 \leq 1$ and $0 \leq f_2 / (f_1 + f_2) \leq 1$. It can be seen that when $f_1 + f_2 = 0.6, f_2 / (f_1 + f_2) = 0.12$ the difference in Young’s modulus of these two methods is greater than 50%, hence, the two-step method may not be valid for the whole range of three-phase composite materials. The bulk modulus of both methods gives the same value, as for Example 1. The contour plots in Fig. 8 are useful for the optimization problem such that the composite with the required properties may be created with the optimum cost.

<table>
<thead>
<tr>
<th>Young’s modulus $E_i / E_3$</th>
<th>Poisson’s ratio $\nu_i$</th>
<th>Bulk modulus $K_i / E_3$</th>
<th>Shear modulus $\mu_i / E_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inclusion</td>
<td>$E_1 / E_3 = 100,000$</td>
<td>$\nu_1 = 0.40$</td>
<td>$K_1 / E_3 = 166,666.7$</td>
</tr>
<tr>
<td>Interphase</td>
<td>$E_2 / E_3 = 0.1$</td>
<td>$\nu_2 = 0.49$</td>
<td>$K_2 / E_3 = 1.66667$</td>
</tr>
<tr>
<td>Matrix</td>
<td>$E_3 / E_3 = 1.0$</td>
<td>$\nu_3 = 0.45$</td>
<td>$K_3 / E_3 = 3.33333$</td>
</tr>
</tbody>
</table>

CONCLUSIONS

The effective elastic moduli of a three-phase composite are calculated by using the four-phase sphere model and by using a two-step method. The bulk modulus calculated using both methods is the same while other moduli are not the same. The analysis of four-phase sphere model is exact while the two-step method is an approximate scheme. These models are applied to concrete and by comparing with the experimental results the elastic properties of the interfacial transition zone are conjectured or if the parameters of the transition zone are known the effective properties of concrete can be predicted. The results show that the two-step method gives sufficiently correct answers for concrete. However, for composite materials where the stiffness of the phases widely differ, the two-step method may not give results close to the four-phase model. The contour plot of the effective moduli presented in this paper is useful for optimization problem.

REFERENCES

Hashin Z. and Monteiro, P. J. M. (2002), “An inverse method to determine the elastic properties of the interphase between the aggregate and the cement paste,” Cement and Concrete Research, 32,


**APPENDIX: ESHELBY’S FORMULA**

Eshelby (1956) gave an expression for the strain energy $U$ stored in a composite medium subjected to a uniform field at infinity as

$$U = U_0 \pm \frac{1}{2} \int_S (t_i u_i^0 - t_i^0 u_i) dS,$$  \hspace{1cm} (A.1)

where $t_i$ and $u_i$ are the traction and displacement components of the composite medium on the surface $S$ of the inclusion, $t_i^0$, $u_i^0$ and $U_0$ are traction and displacement components on the surface $S$ and the stored strain energy of the homogeneous medium (with no inclusion) subjected to the same uniform field. The $+$ or $-$ sign in Eq. (A.1) is used when the uniform strain field or stress field is prescribed, respectively.

The strain energy of the composite model shown in Fig. 2 is $U$ and when the inclusion and matrix are replaced by the equivalent medium the strain energy is $U_0$, therefore the energy condition to be satisfied is

$$U = U_0,$$  \hspace{1cm} (A.2)

By analyzing the boundary value problem under uniform conditions at infinity combined with the energy conditions in Eqs. (A.1) and (A.2) the effective properties may be obtained from the condition,

$$\int_{\theta=0}^{2\pi} \int_{\psi=0}^{\pi} \left[ \sigma_{R\theta}^0 u_R + \sigma_{R\psi}^0 u_\psi + \sigma_{\theta\theta}^0 u_\theta - u_R^0 \sigma_{RR} - u_\psi^0 \sigma_{R\psi} - u_\theta^0 \sigma_{\theta\theta} \right] R_n^2 \sin \psi \, d\psi \, d\theta = 0,$$  \hspace{1cm} (A.3)

where $n = 2$ or 3 for the three-phase or the four-phase sphere model, respectively.