A REVIEW OF ANALYTICAL METHODS TO DETERMINE EFFECTIVE MECHANICAL PROPERTIES OF COMPOSITES WITH SPHERICAL INCLUSIONS

Anil C. Wijeyewickrema
Associate Professor, Department of Civil Engineering
Tokyo Institute of Technology, O-okayama, Tokyo

Somsak Leungvichcharoen
Graduate Student, Department of Civil Engineering
Tokyo Institute of Technology, O-okayama, Tokyo

Abstract: In this paper different analytical methods to obtain the effective elastic properties of composite materials with randomly distributed spherical inclusions, where all phases are isotropic elastic, are reviewed. The validity of each method depends on whether the inclusions are homogeneous or inhomogeneous, the inclusion gradation and the volume fraction of the phases. Voigt and Reuss approximations, dilute suspension model, self-consistent model are applicable for multi-phase composites, while the composite spheres and three-phase composite models are specified for two-phase composite materials. The more complicated models such as $(n+1)$-phase model, two-step method for four-phase composite, differential effective medium approximation and combined methods are also discussed.

Key Words: Effective properties, elastic moduli, spherical inclusions

1. INTRODUCTION

The scope of this review is restricted to composites with randomly distributed spherical inclusions where all phases are isotropic elastic. Various methods to calculate effective mechanical properties are discussed in many monographs e.g., Christensen (1979), Mura (1987), Aboudi (1991), Nemat-Nasser and Hori (1999), Torquato (2001), and Milton (2002). In the rest of the paper C, M, A, NH will refer to Christensen (1979), Mura (1987), Aboudi (1991) and Nemat-Nasser and Hori (1999), respectively. Useful reviews of some methods used to determine effective properties can be found in the papers by Berryman (1980a,b), Cleary et. al. (1980), Hashin (1983) and Norris (1985). In this section the concepts of representative volume element (RVE) and the averaging of stress and strain, which are essential for calculating effective properties are discussed.

1.1 Representative Volume Element

When the composite has randomly distributed microstructure, the representative volume element (RVE) concept (NH, pg. 11-12), has to be used to obtain the effective properties (see Fig. 1). The size of the RVE should be large compared to the length scale of microstructure to contain a sufficient number of microheterogeneities but still very small compared to the characteristic length of structure, so that continuum mechanics can be applied to analyze the whole structure.
In this paper the constituents of the composite are isotropic, hence, for convenience the stress tensor $\mathbf{\sigma}^{(i)}$ and strain tensor $\mathbf{\varepsilon}^{(i)}$ of phase $i$ are expressed as (A, pg. 1-2),

$$\mathbf{\sigma}^{(i)} = \frac{1}{3} \text{tr}(\mathbf{\sigma}^{(i)}) \mathbf{I} + \mathbf{s}^{(i)}, \quad \mathbf{\varepsilon}^{(i)} = \frac{1}{3} \text{tr}(\mathbf{\varepsilon}^{(i)}) \mathbf{I} + \mathbf{e}^{(i)},$$  \hspace{1cm} (1)

where $\text{tr}$ denotes trace of second order tensor, $\mathbf{I}$ the second order identity tensor, $\mathbf{s}^{(i)}$ and $\mathbf{e}^{(i)}$ the deviatoric part of $\mathbf{\sigma}^{(i)}$ and $\mathbf{\varepsilon}^{(i)}$, respectively. Hooke’s law for each phase can be written as

$$\text{tr}(\mathbf{\sigma}^{(i)}) = 3K^{(i)} \text{tr}(\mathbf{\varepsilon}^{(i)}), \quad \mathbf{s}^{(i)} = 2\mu^{(i)} \mathbf{e}^{(i)},$$ \hspace{1cm} (2)

where $K^{(i)}$ and $\mu^{(i)}$ are bulk and shear moduli of phase $i$.

**1.2 The Stress and Strain averages**

Applying uniform traction $\mathbf{t} = \mathbf{\sigma}^{(i)} \mathbf{n}$ (or linear displacement $\mathbf{u} = \mathbf{\varepsilon}^{(i)} \mathbf{x}$) along the boundary of the RVE, yields a homogeneous state of stress $\mathbf{\sigma}^0$ (or homogeneous state of strain $\mathbf{\varepsilon}^0$) at the boundary, which with the aid of Gauss theorem is equal to the average stress $\bar{\mathbf{\sigma}}$ (or average strain $\bar{\mathbf{\varepsilon}}$) obtained from the volume integral as (A, pg. 4-8),

$$\mathbf{\sigma}^0 = \overline{\mathbf{\sigma}} = \frac{1}{V} \int_V \mathbf{\sigma}(\mathbf{x}) \, dV = \sum_{i=1}^{N} c_i \overline{\mathbf{\sigma}}^{(i)}, \quad \mathbf{\varepsilon}^0 = \overline{\mathbf{\varepsilon}} = \frac{1}{V} \int_V \mathbf{\varepsilon}(\mathbf{x}) \, dV = \sum_{i=1}^{N} c_i \overline{\mathbf{\varepsilon}}^{(i)},$$ \hspace{1cm} (3)

where $V$ is total volume of RVE, $c_i, \overline{\mathbf{\sigma}}^{(i)}, \overline{\mathbf{\varepsilon}}^{(i)}$ are the volume fraction, average stress and strain tensors of phase $i$, respectively. For random distribution of microstructure the composite is considered as isotropic hence

$$\text{tr}(\overline{\mathbf{\sigma}}) = 3\overline{K} \text{tr}(\overline{\mathbf{\varepsilon}}), \quad \overline{\mathbf{s}} = 2\overline{\mu} \overline{\mathbf{e}},$$ \hspace{1cm} (4)

where $\overline{K}$ and $\overline{\mu}$ are effective bulk and shear moduli of the composite. The decoupled equations for bulk and shear moduli in Eqs. (2) and (4) yield a convenient way to determine the effective moduli of the composite. By applying the homogeneous volumetric stress (or homogeneous volumetric strain) at the boundary of RVE, analyzing the micro-mechanics elasticity problem, using Eshelby’s formula (Eshelby, 1956; C, pg. 29) and considering the theorem of minimum complementary energy (or minimum potential energy), finally the lower or upper bound of the effective bulk modulus can be obtained. In a similar manner, by applying the homogeneous simple shear stress (or homogeneous simple shear strain) at the boundary of RVE, the upper or lower bound of effective shear modulus can be obtained. If the upper and lower bounds coincide, the bounds yield the effective moduli of the composite.
2. METHODS TO OBTAIN EFFECTIVE ELASTIC MODULI

In this section the following methods to compute average properties of composites with spherical inclusions are discussed: (a) Voigt and Reuss approximations, (b) dilute suspension model, (c) self-consistent model, (d) composite spheres model, (e) three-phase sphere model, (f) \((n+1)\)-phase sphere model, (g) two-step method for four-phase composites, (h) differential effective medium approximation, (i) combined methods.

2.1 Voigt and Reuss Approximations

Voigt (1889) assumed that the average strain of each phase is equal to the applied strain, which yields (M, pg. 421-424; NH, pg. 213)

\[
\bar{K} = \sum_{i=1}^{n} c_i K^{(i)}, \quad \bar{\mu} = \sum_{i=1}^{n} c_i \mu^{(i)}. \tag{5}
\]

Later Reuss (1929) assumed that the average stress of each phase is equal to the applied stress, which yields (M, pg. 424-425; NH, pg. 212)

\[
\frac{1}{\bar{K}} = \sum_{i=1}^{n} \left( c_i / K^{(i)} \right), \quad \frac{1}{\bar{\mu}} = \sum_{i=1}^{n} \left( c_i / \mu^{(i)} \right). \tag{6}
\]

These approximations are independent of the size and shape of inclusions. It has been proved by Hill (1952), that Voigt and Reuss approximations give the upper and lower bounds for effective stiffness, respectively. Better rigorous bounds are provided by Hashin and Shtrikman (1963). Many bounds have been proposed to improve the Hashin-Shtrikman bounds, see Torquato (1991), Chapters 21-22 of Toquato (2001) and Chapters 21-27 of Milton (2002) for more details.

2.2 Dilute Suspension Model

This model is valid when the volume fractions of inclusions are very small \((c_i \ll 1, i = 1,..,n-1)\), hence the interaction between particles is neglected. For a two-phase composite, the effective moduli are obtained from the solution of a single spherical inclusion in an infinite matrix medium, subjected to homogeneous boundary conditions at infinity. Expression for \(\bar{K}\) and \(\bar{\mu}\) were presented by Bruggemann (1937), but only the expression for \(\bar{K}\) was correct. As noted by Hashin (1964) the correct \(\bar{\mu}\) was derived by Dewey (1947). Eshelby (1957) and Hashin (1959) unaware of the work of Dewey (1947) obtained correct expressions, for both \(\bar{K}\) and \(\bar{\mu}\).

In the case of multi-phase composites, when phases 1 to \(n\)-1 are spherical, it is approximated that the average stress (or strain) in each phase are the average stress (or strain) of a single spherical inclusion of that phase embedded in an infinite matrix medium (NH, pg. 246-248). By applying homogeneous stress boundary conditions at infinity yields

\[
\frac{\bar{K}}{K^{(n)}} = \left[ 1 + \sum_{i=1}^{n-1} c_i \left( \frac{K^{(n)}}{K^{(n)} - K^{(i)}} - s_1 \right)^{-1} \right]^{-1}, \quad \frac{\bar{\mu}}{\mu^{(n)}} = \left[ 1 + \sum_{i=1}^{n-1} c_i \left( \frac{\mu^{(n)}}{\mu^{(n)} - \mu^{(i)}} - s_2 \right)^{-1} \right]^{-1}, \tag{7}
\]

where \(s_1 = \frac{1}{2}(1 + \nu^{(n)})/(1 - \nu^{(n)})\), \(s_2 = \frac{2}{13}(4 - 5\nu^{(n)})/(1 - \nu^{(n)})\), and \(\nu^{(n)}\) is the Poisson’s ratio of phase \(n\) obtained from \(\nu^{(n)} = \frac{1}{2}(3K^{(n)} - 2\mu^{(n)})/(3K^{(n)} + \mu^{(n)})\). Applying a homogeneous
strain boundary condition yields

\[
\frac{\bar{K}}{K^{(n)}} = 1 - \sum_{i=1}^{n-1} C_i \left( \frac{K^{(i)} - s_i}{\bar{K} - s_i} \right)^{-1}, \quad \frac{\bar{\mu}}{\mu^{(n)}} = 1 - \sum_{i=1}^{n-1} C_i \left( \frac{\mu^{(i)} - s_i}{\bar{\mu} - s_i} \right)^{-1}. \tag{8}
\]

It is noted that the series expansions of \( \bar{K} \) and \( \bar{\mu} \) obtained from Eq. (7) agree with \( K \) and \( \mu \) from Eq. (8) to the first order in the volume fraction of inclusion phases.

### 2.3 Self-Consistent Model (SCM)

In this model, the average stain or stress field in the inclusions, is approximated by analyzing the problem of a single inclusion embedded in an equivalent medium subjected to the homogeneous boundary conditions at infinity as shown in Fig. 2a. As noted in Norris (1985), there are two classes of SCM. First is the symmetric SCM presented by Kroner (1958) and it was extended by Budiansky (1965), where all phases are considered as inclusions. Hill (1965) and Berryman (1980a) proved that the results are always within Hashin-Shtrikman bounds for two-phase and multi-phase composites, respectively. In addition, the effective moduli obtained by applying the homogeneous stress or homogeneous strain boundary conditions are equivalent (NH, pg. 228-231). The second class of SCM is asymmetric SCM (Wu, 1966; Walpole, 1969; Boucher, 1974) in which one phase is considered as a matrix phase and all other phases are considered as inclusions. However, the results for spherical inclusions are the same for both classes, hence the coupled equations to calculate effective bulk and shear moduli for an \( n \)-phase composite are given below (NH, pg. 248-250),

\[
\sum_{i=1}^{n} C_i \left( 1 + s_i \frac{K^{(i)} - \bar{K}}{\bar{K}} \right)^{-1} = 1, \quad \sum_{i=1}^{n} C_i \left( 1 + s_i \frac{\mu^{(i)} - \bar{\mu}}{\bar{\mu}} \right)^{-1} = 1, \tag{9}
\]

where \( s_1 = \frac{1}{3} (1 + \bar{\nu}) / (1 - \bar{\nu}) \), \( s_2 = \frac{2}{15} (4 - 5\bar{\nu}) / (1 - \bar{\nu}) \), and \( \bar{\nu} \) is the effective Poisson’s ratio. This model is not limited to small volume fractions.

Budiansky (1965) showed the unusual results of SCM, in which for a two-phase composite material if one phase consists of holes \( (K^{(1)} = \mu^{(1)} = 0) \) and another phase is an incompressible material \( (K^{(2)} = \infty) \), it yields \( \bar{K} = \bar{\mu} = 0 \) for \( C_i > 0.5 \); and if \( K^{(1)} = \mu^{(1)} = K^{(2)} = \infty \) it yields \( \bar{K} = \infty \) for all \( C_i \). Hence SCM is criticized by some authors (C, pg. 59-60), although Milton (1984) proved that symmetric SCM corresponds exactly with the composite, which has a hierarchical microstructure as show in Fig. 2b (see also Torquato, 2001, pg. 465).
2.4 Composite Spheres Model

This model is presented by Hashin (1962) appropriate for a very fine gradation of spherical inclusions embedded in a continuous matrix phase (see Fig. 3a). The schematic figure of this model (Fig. 3b) shows each inclusion surrounded by a spherical shell of matrix material. The ratio of radii is constant and is obtained from the volume fractions. Filling all gaps between composite spheres the distribution requires sizes down to infinitesimal. By analyzing a single representative composite sphere subjected to the homogeneous volumetric stress (or homogeneous volumetric strain) on the surface of the sphere (Fig. 3c), finally the coincident upper and lower bounds of bulk modulus is obtained as (A, pg. 21-22; C, pg. 47-52)

\[
\overline{K} = K^{(2)} + \frac{c_1(K^{(1)} - K^{(2)})}{1 + (1-c_1)(K^{(1)} - K^{(2)})/(K^{(2)} + \frac{4}{3}\mu^{(2)})},
\]

(10)

when the inclusion material is considered as phase 1. However, the shear modulus can be obtained only as bounds because the homogeneous simple shear strain prescribed on the surface of the composite sphere, results in boundary stresses that do not correspond to a state of simple shear stress; and when homogeneous simple shear stresses are prescribed on the surface of the composite sphere, results in boundary strains that do not correspond to a state of simple shear strain. The bounds of shear modulus are recorded in Hashin (1962).

2.5 Three-Phase Sphere Model

Christensen and Lo (1979) succeeded in obtaining the exact shear stiffness for the composite shown in Fig. 3a, by considering a single composite sphere embedded in an infinitely extended equivalent homogeneous material. The three-phase sphere model is shown in Fig. 4. When the homogeneous volumetric stress or volumetric strain boundary condition is prescribed at infinity to calculate the effective bulk modulus, the result is the same as that obtained from the composite spheres model (Eq. (10)). The prescribed simple shear deformation or simple shear stress at infinity yields the same quadratic equation

\[
A(\overline{\mu}/\mu^{(2)})^2 + 2B(\overline{\mu}/\mu^{(2)}) + C = 0,
\]

(11)

for obtaining the shear modulus (A, pg. 24-26; C, pg. 53-58), where the constants \(A, B\) and \(C\) are calculated from Poisson’s ratio and shear modulus of both phases (Christensen and Lo, 1979, 1986). This model is applicable for all ranges of the volume fraction for a two-phase composite with very fine gradation of the inclusions (see Fig. 3a). This method is referred as the generalized self-consistent method (see Hashin, 1983; Christensen, 1990).
2.6 (n+1)-Phase Sphere Model

This model presented by Herve and Zaoui (1993), is a generalized version of the three-phase model, for a composite which contains layered spherical inclusions. By analyzing this model (Fig. 5) with homogeneous boundary conditions prescribed at infinity the exact bulk and shear moduli are obtained. It is noted that the effective bulk modulus $\bar{K} = \bar{K}^{(n)}$ appears in recursive form and is successively obtained from the innermost sphere to the outermost sphere as

$$\bar{K}^{(i)} = K^{(i)} + \frac{(R_{i+1}^3 / R_i^3)(\bar{K}^{(i-1)} - K^{(i)})}{1 + (1 - R_{i+1}^3 / R_i^3)[(\bar{K}^{(i-1)} - K^{(i)})/(K^{(i)} + \frac{4}{3} \mu^{(i)})]}$$

(12)

where $R_i, R_{i+1}$ are the outer and inner radii of phase $i$, respectively, $\bar{K}^{(i)}$ is the effective bulk modulus of the layered spherical inclusion which contains phase 1 to phase $i$. The effective shear modulus can be calculated from the quadratic equation

$$A(\bar{\mu} / \mu^{(n)})^2 + 2B(\bar{\mu} / \mu^{(n)}) + C = 0,$$

(13)

where the constants $A$, $B$ and $C$ are not easily obtained and are given in their paper. Similar to the three-phase model, this model is appropriate for very fine gradation of the inclusions. The application of this model can be found in Herve and Pellegrini (1995), and Garboczi and Bentz (1997).

When $n = 3$, the (n+1)-phase sphere model can be used to analyze the three-phase composite (Fig. 6a), with the four-phase sphere model. This model has been used to study the mechanical properties of concrete (Ramesh, et. al., 1996) and cement-mortar (Hashin and Monteiro, 2002).

2.7 Two-Step Method for Three-phase Composite

In general to obtain the effective properties of a three-phase composite, a four-phase sphere model is required. An alternative method is to use the less complicated three-phase sphere model twice in succession, i.e., a two-step method. Step 1: the three-phase model is used to obtain the effective properties of the layered spherical inclusion (Fig. 6b). Step 2: the three-phase sphere model is used again to obtain the effective properties of a spherical inclusion with effective properties obtained from Step 1 embedded in matrix material (Fig. 6c). Iwase et. al. (2002) showed that $\bar{K}$ obtained by this method agrees with the $\bar{K}$ of the four-phase sphere model, and when applied to cement-based material, the methods give slightly different results for $\bar{\mu}$. The validity of this method is discussed by
Wijeyewickrema and Leungvichcharoen (2003), while a similar method was applied to composites with coated fibers by Sutcu (1992).

Fig. 6. Three-phase composite and schematic diagram of two-step method.

### 2.8 Differential Effective Medium Approximation (DEM)

This scheme was proposed by Roscoe (1952) to study the viscosity of suspensions of rigid spheres and was used by Roscoe (1973) and Boucher (1974) to examine the effective elastic moduli. Mclaughlin (1977) clearly explained this method with a concise formulation for a two-phase composite. The ordinary process of DEM is iteratively replacing (or adding up) the background material by small volume of inclusion and then re-computing the new effective background material (by dilute suspension model) until the final volume fraction is reached. For spherical inclusions (here referred as phase 1), two coupled differential equations are obtained (A, pg. 26-28)

\[
\frac{dK}{dc_2} = \frac{(\bar{K} + \frac{c_1}{3} \bar{\mu})(\bar{K} - K^{(1)})}{c_2(K^{(1)} + \frac{4}{3} \bar{\mu})}, \quad \frac{d\bar{\mu}}{dc_2} = \frac{5\bar{\mu}(\bar{K} + \frac{c_1}{3} \bar{\mu})(\bar{\mu} - \mu^{(1)})}{c_2[3\bar{\mu}(\bar{K} + \frac{4}{3} \bar{\mu}) + 2\mu^{(1)}(\bar{K} + 2\bar{\mu})]}.
\]  

(14)

Cleary et. al. (1980) reviewed the realization of this method compared to SCM. Norris (1985) showed that the effective properties of a composite obtained from this method depend not only on volume fractions but also on the construction path of the composite i.e., the process of inclusion addition. However, Norris et. al., (1985) mentioned that the equivalent equations are obtained from both processes i.e., “replacing” (fixed volume process; see Fig. 7) and “adding up” (variable volume process). The different results can be obtained from alternative process e.g. by iteratively replacing background material with both phases (i.e., materials of the inclusion and the matrix). The method is applicable for multi-phase composites.

Fig. 7. Schematic diagram of DEM.
2.9 Combined Methods

There are some limitations in the analytical models presented, which depend on the assumptions used in the models. To predict the overall properties of some composite materials, a combination of methods may be employed.

For example, Garboczi and Berryman (2001) combined the three-phase sphere model and DEM to obtain the effective moduli of a three-phase composite and applied it to cement-mortar considered as a three-phase composite composed of spherical aggregates, transition zone (ITZ) and cement-paste matrix. The three-phase sphere model is applied to calculate the equivalent moduli of layered spherical inclusions (aggregate and ITZ), and these equivalent properties are used in DEM to obtain the effective elastic moduli of mortar. The results agree very well with finite element results.

Sheng (1990) modeled the sedimentary rock as a three-phase composite composed of fluid, solid and cement material. The mixture of fluid and cement material is considered as matrix phase while solid phase is considered as ellipsoidal inclusions. The elastic moduli of the matrix phase are calculated by SCM, and the overall properties of sedimentary rock are determined by using DEM. The results agree well with the experimental data.

3. DISCUSSION AND CONCLUSIONS

Various models to obtain the effective properties of composite materials are reviewed. No geometric model can cover all cases, each model is appropriate for a composite with a particular microstructure e.g., dilute suspension model is suitable for very small volume fractions, SCM is for a composite with hierarchical microstructure. Composite spheres model, three-phase sphere model, (n+1)-phase sphere model and two-step method and DEM are for composites with very fine grain size distributions of inclusions. The effective moduli for a wider class of materials can be obtained by using combined methods. Due to space constraints not all analytical models in the literature are reviewed in this paper, for instance, the methods based on Mori-Tanaka theorem (Mori and Tanaka, 1973; Weng, 1984; Benveniste, 1987), double-inclusion model (Hori and Nemat-Nasser, 1993; NH pg. 378), contact cement theory (Dvorkin, et. al., 1994, 1999) and Maxwell approximation (Torquato, 2001, pg. 470-473). The summary of all models discussed in this paper is presented in Table 1. To obtain good approximations for the elastic moduli for any composite, it is necessary to use the appropriate analytical model.

Table 1. Summary of the models to predict effective moduli of composites

<table>
<thead>
<tr>
<th>Models</th>
<th>Inclusion Type</th>
<th>Composite Material</th>
<th>Inclusion Gradation</th>
<th>Volume Fraction</th>
<th>Effective Bulk Modulus</th>
<th>Effective Shear Modulus</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Voigt and Rues</td>
<td>any</td>
<td>multi-phase</td>
<td>any</td>
<td>any</td>
<td>bounds</td>
<td>bounds</td>
</tr>
<tr>
<td>2. Dilute suspension</td>
<td>homogeneous</td>
<td>multi-phase</td>
<td>any</td>
<td>small</td>
<td>bounds(^1)</td>
<td>bounds(^1)</td>
</tr>
<tr>
<td>3. Self consistent (SCM)</td>
<td>homogeneous</td>
<td>multi-phase</td>
<td>hierarchical</td>
<td>any</td>
<td>exact</td>
<td>exact</td>
</tr>
<tr>
<td>4. Composite spheres</td>
<td>homogeneous</td>
<td>two-phase</td>
<td>very fine</td>
<td>any</td>
<td>exact</td>
<td>bounds</td>
</tr>
<tr>
<td>5. Three-phase sphere</td>
<td>homogeneous</td>
<td>two-phase</td>
<td>very fine</td>
<td>any</td>
<td>exact</td>
<td>exact</td>
</tr>
<tr>
<td>6. (n+1)-phase sphere</td>
<td>layered</td>
<td>n-phase</td>
<td>very fine</td>
<td>any</td>
<td>exact</td>
<td>exact</td>
</tr>
<tr>
<td>7. Two-step method</td>
<td>layered</td>
<td>three-phase</td>
<td>very fine</td>
<td>any</td>
<td>exact</td>
<td>approx.</td>
</tr>
<tr>
<td>8. DEM</td>
<td>homogeneous</td>
<td>multi-phase</td>
<td>very fine</td>
<td>any</td>
<td>approx.</td>
<td>approx.</td>
</tr>
<tr>
<td>9. Three-phase model + DEM</td>
<td>layered</td>
<td>multi-phase</td>
<td>very fine</td>
<td>any</td>
<td>approx.</td>
<td>approx.</td>
</tr>
<tr>
<td>10. SCM + DEM</td>
<td>homogeneous</td>
<td>three-phase</td>
<td>very fine</td>
<td>any</td>
<td>approx.</td>
<td>approx.</td>
</tr>
</tbody>
</table>

\(^1\) to first order expressions for \( K \) and \( \mu \) coincide.
REFERENCES


