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Statistical spherical cell model for the elastic properties of particulate-filled composite materials

Y. M. POON*,§, W. L. LUK*, F. G. SHIN‡*
*Department of Applied Physics, Materials Research Centre and ‡Centre for Smart Materials. The Hong Kong Polytechnic University, Hong Kong, People's Republic of China
E-mail: apaympoo@polyu.edu.hk

The single spherical cell model of F. J. Guild *et al.* (*J. Mater. Sci. Lett.* **13** (1994) 10) is extended to take into account the statistical spatial distribution of the inclusions in particulate-filled composite materials. Using this model, the elastic properties of a glass-bead filled epoxy composite material were calculated. When compared with the single spherical cell model, we found that the statistical spherical cell model gave predictions consistently closer to the experimental values for both the Young's modulus and the Poisson's ratio.

1. Introduction

Advance in technology has lead to the development of sophisticated particulate-filled composite materials. A variety of them has already found wide range of usage in our daily life. However, their applications can only be properly exploited when their physical behaviors, in particular, the mechanical behaviors, are well understood. For this purpose, predictive models can provide a way to understand the mechanisms behind the physical behaviors. There have been numerous models for the description of the overall properties of these composites. However, not many of these models have taken into account the spatial distribution of the particles in the composite.

In 1988, Davy and Guild [1] applied finite element analysis upon an axisymmetric cylindrical cell model containing a single spherical inclusion. The fact that there are many inclusions were simulated by adding suitable constraints along the boundary of the cylindrical cell, and incorporating hard-core Gibbs point process for the distribution of the centers of the spherical inclusions. The bulk linear elastic behaviors of glass-bead filled polymer of filler volume fraction up to approximately 50% were calculated under the condition of equal stress or equal strain, which should give the upper or the lower bounds for the true values respectively. Good agreement in Young's modulus was found between the experimental and the calculated values. While reasonable agreement in Poisson's ratio was obtained at low filler volume fractions, the agreement was less satisfactory at higher volume fractions. The reason for the increased discrepancies at higher volume fractions, as the authors believed, was that the cylindrical model could not correctly reflect the isotropy of the mechanical properties of the overall composite material.

Guild *et al.* [2] applied finite element method to a single spherical cell model to predict the mechanical properties of rubber-toughened epoxy polymer. The filler volume fraction was varied from 1.07% to 40.5% by changing the ratio of the radius of the spherical cell to that of the spherical inclusion at its center. Internal stresses and nodal displacements were calculated with the spherical cell under unidirectional loading and the multiple-inclusions effect was simulated by constraining the deformed outer surface of the spherical cell and the interface between the matrix and the inclusion to be perfect ellipsoids. Elastic constants such as the bulk modulus, Young's modulus and Poisson's ratio were calculated and compared with the predicted values from the previously mentioned cylindrical cell model.

Then in [3], Guild *et al.* applied the same single spherical cell model to investigate the stress distributions around and within a rubber particle or a void embedded in a matrix of epoxy polymer. The bulk modulus values for various volume fractions of the inclusion phase and the manifestations of the stress distribution on the toughening mechanism were considered. Good agreement was revealed between the predicted and experimental values of a range of mechanical properties of the rubber material.

In this work, Guild's single spherical cell model is extended to a statistical spherical cell model. Using finite element analysis, quantitative mechanical properties were predicted and compared with experimental data.

2. Calculation of elastic properties

For easier reference, we review briefly the theory of Davy and Guild [1] in the following. A particulate-filled

§Author to whom all correspondence should be addressed.

composite material can be divided into various Voronoi cells, each containing one inclusion. A Voronoi cell is defined as the region containing one inclusion such that any point in this region is closer to this inclusion than to any other inclusions. Suppose P_j ($j = 1$ to N) are the centers of the spherical inclusions and $X_j(\omega)$ be the distance from P_j to the boundary of its own Voronoi cell in the direction ω, then the random variable $X(\omega)$ corresponding to a "typical center" is defined to have the following probability function:

$$P(X(\omega) \leq x) = \frac{\xi(\Sigma_{p_j \in V} I_j(x))}{N} \quad (1)$$

where $\xi(\,)$ denotes the expectation value of the enclosed quantity, V is the volume of the composite and $I_j(x)$ is unity when $X_j(\omega)$ is not greater than x, and is zero otherwise. When $X(\omega)$ is averaged over ω, the resulting value is denoted as X, which can be considered as the half-interparticle distance.

For a random distribution of non-overlapping spheres of radius r, X satisfies the Gibb's hard-core point distribution:

$$P(X > x) \approx \exp(-k(x^3/r^3 + 3r/x - 4)) \quad (2)$$

where k is a function of the volume fraction p of the inclusions given by:

$$\int_1^{\infty} y^2 \exp\left(-k\left(y^3 + \frac{3}{y} - 4\right)\right) dy = \frac{1-p}{3p} \quad (3)$$

In our statistical spherical cell model, X is taken to be the radius of the spherical cell and hence each cell has a radius R equals to X and has a spherical inclusion of radius r at its center.

The squared coefficient of variance of the ratio R^3/r^3, denoted by $CV(R^3/r^3)$, can be calculated by:

$$CV(R^3/r^3) = p^2 \xi(R^6/r^6) - 1 \quad (4)$$

where $\xi(X^6/r^6)$ is given by:

$$\xi(X^6/r^6) = 1 + 2\int_0^1 y^{-3} \exp[-k(y^{-1} + 3y^{1/3} - 4)] dy \quad (5)$$

The values of $CV(R^3/r^3)$, together with the k values, for volume fraction p varying from 0.05 up to 0.70 are shown in Table I. Here $R/r = (1/p)^{1/3}$. The values for volume fractions above 0.5 are not shown in Davy and Guilds [1] and the value of k at $p = 0.5$ is misprinted in their paper.

For a given volume fraction p, most spherical cells in the statistical model will have their volume fractions of inclusion close to p. Therefore, a spherical cell, whose volume fraction of inclusion equals to that of the composite material, with the radius ratio denoted by R_p/r, can be selected as the representative sphere. The distribution function of the inter-particle distances between it and the other spheres satisfies the statistical distribution function (2). Employing this statistical distribution, the average stress and strain of the composite is found by a

TABLE I k values and $CV(R^3/r^3)$ values for the spherical cell model

Volume fraction (p)	k	R/r	$CV(R^3/r^3)$
0.05	0.0578	2.7144	0.7755
0.10	0.1319	2.1544	0.6272
0.15	0.2258	1.8821	0.5111
0.20	0.3446	1.7100	0.4140
0.25	0.4958	1.5874	0.3405
0.30	0.6894	1.4938	0.2767
0.35	0.9401	1.4190	0.2235
0.40	1.2694	1.3572	0.1788
0.45	1.7110	1.3050	0.1404
0.50	2.3088	1.2600	0.1104
0.55	3.1483	1.2205	0.0846
0.60	4.3615	1.1856	0.0634
0.65	6.1896	1.1544	0.0461
0.70	9.0949	1.1262	0.0322

volumetric averaging. Using finite element analysis, the internal stresses and displacements can be determined, and hence the Young's modulus and the Poisson's ratio of this representative spherical cell can be found. Suppose f is a property function of the radius ratio R/r, according to [1], the statistically corrected expectation value of f, $\xi\,(f)$ can be found by the following approximation:

$$\xi(f) \approx f_p + d(f) \quad (6)$$

where f_p is the value of f at radius ratio R_p/r, and

$$d(f) = \frac{1}{2}\left(R_p^3/r^3\right)(R^3 f/r^3)'' CV(R^3/r^3) \quad (7)$$

The second derivative in Equation 7 can be approximated by the finite difference:

$$(R^3 f/r^3)'' = \left(f_1 R_1^3/r^3 + f_2 R_2^3/r^3 - 2 f_p R_p^3/r^3\right)/\Delta^2 \quad (8)$$

where Δ is a small value, f_1 is the value of the property function when

$$(R_1/r)^3 = (R_p/r)^3 - \Delta \quad (9)$$

and f_2 the value of the property function when

$$(R_2/r)^3 = (R_p/r)^3 + \Delta \quad (10)$$

Using Equation 6, we can find the statistically corrected, expectation value of f. The choice of the value for Δ is somewhat arbitrary, as long as it is small enough when compared with the value of $(R_p/r)^3$. We have used $\Delta = 0.1$ in our calculations.

In this study, we take f to be the Young's modulus E or the ratio ν/E, where ν is the Poisson ratio. As indicated in reference [1], the effective Young's modulus is then a weighted harmonic mean of the Young moduli of the component spheres and the effective value of the ratio ν/E is a weighted mean of the ratios of the individual spheres. The representative spherical cell is shown in Fig. 1. Due to symmetry, axisymmetric elements can be used and we need only consider one-quarter of the cross-section, labeled by OAB in Fig. 1.

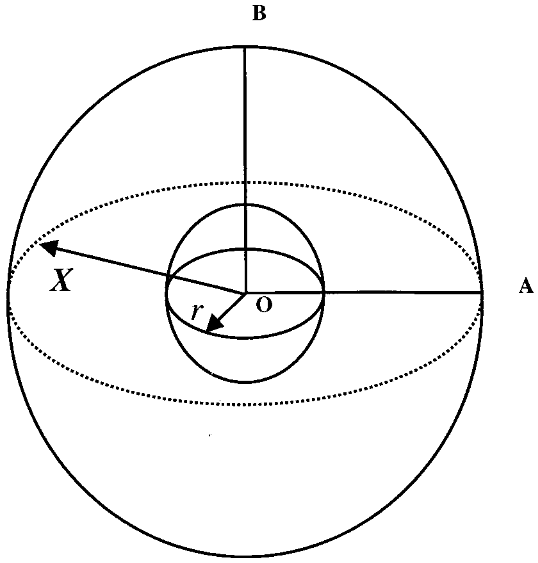

Figure 1 A typical spherical cell.

The mesh for finite element analysis is shown in Fig. 2. The elements used are all 8-node quadrilateral axisymmetric elements except 6-node triangular axisymmetric elements were used around the left lower corner O. The total number of elements is 1024 and the total number of nodes is 3137.

Fig. 3 shows the loading and boundary conditions. The loading condition is such that the cell is under equal-strain deformation along the arc AB due to the load in the y direction. The shape OAB is deformed into OA′B′, which must be part of a perfect ellipse since the overall composite material is isotropic. This shape would not be attained from application of the load alone and constraints must be applied to force this shape. The application of these constraints models the interactions between neighboring spheres.

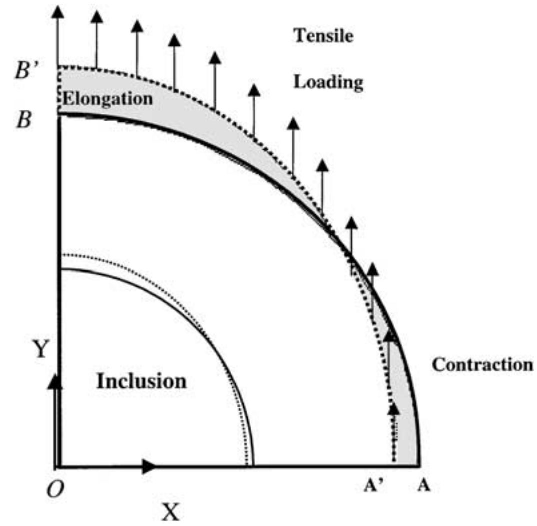

Figure 3 Loading and boundary conditions for the spherical cell model.

When the spherical cell is prescribed with tensile strain ε in the y direction, in order to ensure that the deformed shape must be a perfect ellipsoid, we require that the displacements Δx and Δy in the x and y directions respectively for points on the surface of the spherical cell satisfy:

$$\Delta y = \varepsilon y \tag{17}$$

$$\Delta x = -\nu\varepsilon x \tag{18}$$

where ν is the Poisson's ratio of the composite spherical cell. However, as the value of ν is unknown, we have used the following algorithm to find it.

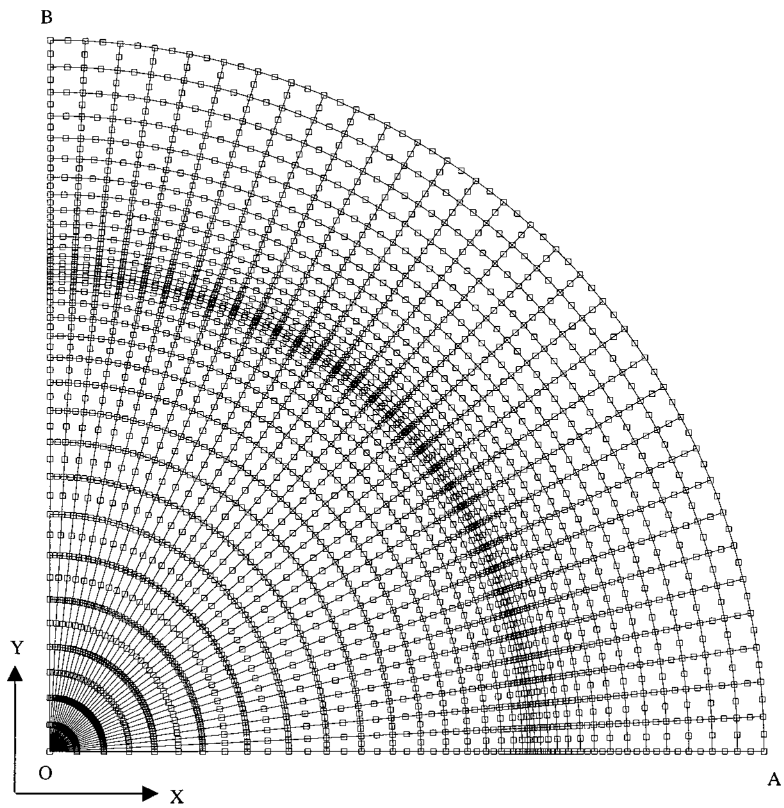

Figure 2 Mesh for finite element analysis.

1. Prescribe to nodes displacements $\Delta y = 0$ and $\Delta x = 0$.

2. Find the bulk modulus of the spherical cell. This can be done by the application of a small constant hydrostatic pressure on the outer surface of the cell. Using finite element analysis, we can find the bulk modulus K using

$$\mathrm{K} = \frac{\text{Hydrostatic pressure}}{\Delta V/V}$$

where $\quad \Delta V/V = 1 - \left(1 - \frac{\Delta R}{R}\right)^3 \approx 3\frac{\Delta R}{R}$

3. Prescribe a small strain value ε, say 0.001, to every outer surface node in Fig. 2 and calculate the corresponding y-direction displacement by Equation 17.

4. (a) (Only to be done for the first time of prescribing displacements of boundary nodes.) Establish a set of "multiple-node connectivity" equations along the outer surface nodes in the x-direction. Or (b) (Should be done when boundary displacements are not newly prescribed.) Prescribe the x-displacements in every outer surface nodes by Equation 18, using the ν_{i+1} value obtained from Step 6.

5. After the finite element analysis, we can find the Young's modulus E_i ($i = 1,2,\ldots$) by averaging the reaction along the plane OA$'$.

6. Using $\nu_{i+1} = \frac{1}{2}(1 - \frac{E_i}{3K})$, we get a new ν value.

7. Repeat Step 4(b) to Step 6 until the successive values of ν are close enough. The "multiple-node connectivity" equations to be used in Step 4(a) can be found by the following.

The original spherical shape of the cell can be described, in parametric form

$$\begin{cases} x = R\cos(\theta) \\ y = R\sin(\theta) \end{cases} \tag{19}$$

where R is the radius of the sphere and θ is the polar angle. Suppose (η, ξ) is a point on the deformed cell, which must have the shape of a perfect ellipse, we have

$$\begin{cases} \eta = aR\cos(\theta) \\ \xi = bR\sin(\theta) \end{cases} \tag{20}$$

where a, b are constants.

By subtracting Equation 19 from 20, we get:

$$\begin{cases} \Delta x = R(a-1)\cos(\theta) \\ \Delta y = R(b-1)\sin(\theta) \end{cases} \tag{21}$$

Because the displacements for every point in the sphere satisfy Equation 21, by arbitrarily choosing two points on the ellipse, say point 1 and point 2, it is easy to show that:

$$\begin{cases} \Delta x_1\cos(\theta_2) = \Delta x_2\cos(\theta_1) \\ \Delta y_1\sin(\theta_2) = \Delta y_2\sin(\theta_1) \end{cases} \tag{22}$$

Suppose there are a series of nodes with node numbers 1 to N along the outer boundary of the ellipse, we get:

$$\begin{cases} \Delta x_i\cos(\theta_{i+1}) = \Delta x_{i+1}\cos(\theta_i) \\ \Delta y_i\sin(\theta_{i+1}) = \Delta y_{i+1}\sin(\theta_i) \end{cases} \tag{23}$$

where $i = 1$ to $N - 1$.

The set of Equations 23 define the "multi-point connectivity" constraints among the nodes on the outer boundary and are used in finite element analysis.

3. Results

The experimental data we studied was for a glass bead filled epoxy resin polymer composite [5]. The Young's modulus for the glass bead and the epoxy resin are 76 GPa and 3.01 GPa respectively. The Poisson's ratios are 0.23 and 0.394, respectively. Using these values,

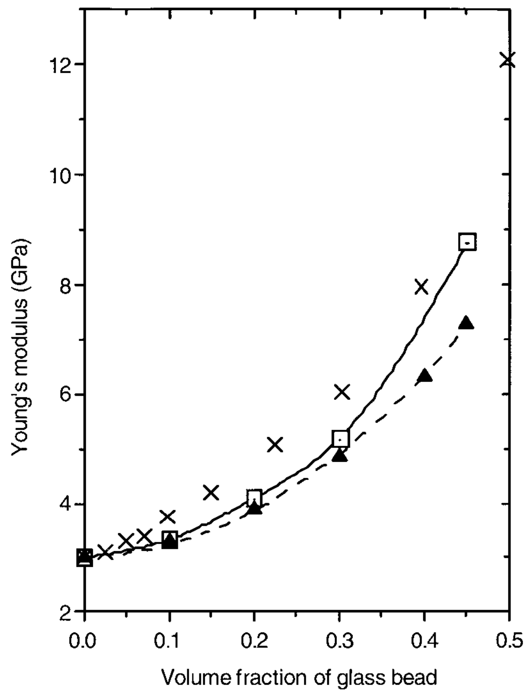

Figure 4 Young's modulus predicted by the statistical spherical cell model (cross: experimental data with error bars [5]; triangle: single spherical model; square: statistical spherical cell model). The error bars for the experimental data are too small to be shown.

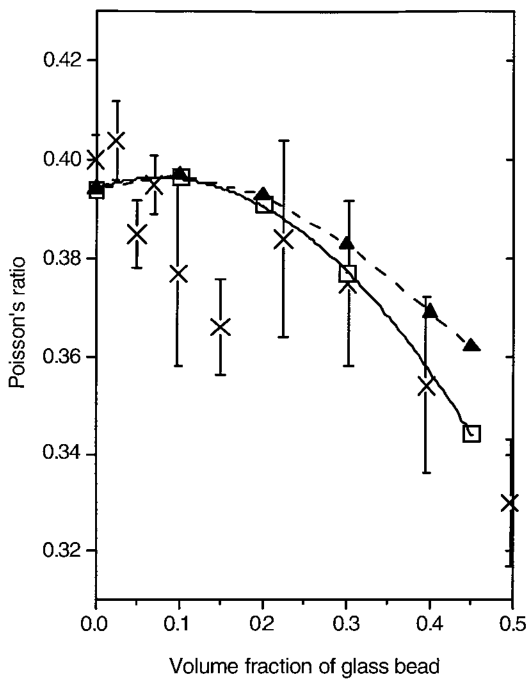

Figure 5 Poisson's ratio predicted by the statistical spherical cell model (cross: experimental data [5] with error bars; triangle: single spherical model; square: statistical spherical cell model).

the Young's modulus predicted by the single spherical cell model is shown in Fig. 4. The statistical spherical cell model and the experimental data are shown for inclusion volume fractions up to 0.45. The experimental data are all higher than the values predicted by the single spherical cell model, with or without statistical correction. Nevertheless, both models fit reasonably well with the experimental data. However, from the figure, we can see that the statistical corrected model shows better agreement with experimental data when the filler volume fraction becomes higher. In Fig. 5, the Poisson's ratio predicted by the single spherical cell model, the values from the statistical spherical cell model and the experimental data are shown. As before, both models are satisfactory. Again, at high filler volume fractions, the values given by the model with statistical correction are closer to the experimental data than those without statistical correction.

4. Conclusion

As pointed out by Guild and Kinloch [3], in the single spherical cell model, the interaction between the inclusions has been taken into account partially and indirectly by the incorporation of the boundary conditions. Our statistical spherical cell model, in addition, also takes into account the inter-particle distance distribution and hence has shown significant improvement in its prediction ability, especially in the calculation of the Poisson's ratio values. Although this work has only demonstrated its advantage in elastic properties calculation, it is quite clear that the method may also be used in dealing with other interesting or important properties.

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