Computational evaluation of effective material properties of composites reinforced by randomly distributed spherical particles

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Abstract

The aim of presenting this paper is to evaluate the effective material properties of spherical particle reinforced composites for different volume fractions up to 60%. A numerical homogenization technique based on the finite element method (FEM) with representative volume element (RVE) was used to evaluate the effective material properties with periodic boundary conditions. The numerical approach is based on the FEM and it allows the extension of the composites with arbitrary geometrical inclusion configurations, providing a powerful tool for fast calculation of their effective material properties. Modified random sequential adsorption algorithm (RSA) was used to generate the three-dimensional RVE models of randomly distributed spherical particles. The effective material properties obtained using the numerical homogenization techniques were compared with different analytical methods and good agreement was achieved. Several investigations had been conducted to estimate the influence of the size of spherical particles and of the RVE on effective material properties of spherical particle reinforced composites.

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1. Introduction

Short fiber and particle reinforced composites do have an advantage of easy manufacturing and good mechanical properties. Since particles are easily mixed with the liquid matrix resin, and the mixture can be injection or compression moulded to produce components with complicated shapes, composites composed of spatially distributed particles have become popular in a wide variety of industrial applications. In addition, using these spatially distributed fibers and particles as reinforcing elements in a controlled manner can provide more balanced properties, which leads to an improved through-the-thickness stiffness/strength and better ability to build complex shapes. A typical problem in solid mechanics is evaluation of the effective elastic properties of a composite material made of a statistically isotropic random distribution of isotropic and elastic spherical particles embedded in a continuous, isotropic and elastic matrix. Although some analytical and semi analytical models have been developed to evaluate the effective material properties of the fiber and particle reinforced composites based on the homogenization techniques, they are often reduced to specific cases. Numerical models seem to be a well suited approach to describe the behavior of these materials, because there is no restriction on the geometry, on the material properties, and on the number of phases in the composite. Therefore the finite element method has been used to determine the effective material properties of short fibers or
particles reinforced composites. In order to obtain realistic predictions of a new material's macroscopic behavior by the computational means, three-dimensional numerical simulations of statistically representative micro-heterogeneous material samples are unavoidable.

A number of classical micro-mechanics theories have been developed and published in the literature. The Voigt approximation [33] is one of the simplest models used to evaluate the effective properties of a composite. It was originally introduced to estimate the average constants of polycrystals. Here it is assumed that the strain throughout the bulk material is uniform. The inverse assumption to Voigt is the Reuss Approximation [26], which assumes that the stress is uniform throughout the phases. But neither Voigt nor Reuss are correct. Under the Voigt model the implied tractions across the boundaries of the phases would in general violate equilibrium, and under the Reuss model the resulting strains would force debonding of the phases. Using variational principles Hashin [14] and Hashin–Shtrikman [15] established bounds on materials that could be considered as “Mechanical mixtures of a number of different isotropic and homogeneous elastic phases” and, in bulk, regarded as statistically isotropic and homogeneous. These two point bounds had been improved by the three point bounds [23,24,28] that incorporate information about the phase arrangement through the statistical correlation parameters. Eshelby [9–11] considered the problem of an ellipsoidal inclusion in an infinite isotropic matrix. What Eshelby realized was that this problem should be equivalent to the problem of a region in a matrix with different material properties. In one case the inhomogeneity of the strain field would be due to the transformation, and in other due to different constitutive properties. Eshelby assumed a well defined matrix. This is not always true, in polycrystalline materials a variety of properties can be exhibited but there is no clearly defined matrix phase. In these cases the interactions between the particles (regions) are more significant. The Mori–Tanaka [25] method was designed to calculate the average internal stress in the matrix containing precipitates with eigenstrains. Benveniste [1] reformulated it so that it could be applied to composite materials. He considered isotropic phases and ellipsoidal phases. Torquato [29,30] derived approximate relations to estimate effective shear and bulk moduli of two and three-dimensional isotropic dispersions by truncating, after third order terms, an exact series expansion for the effective stiffness tensor of d-dimensional two phase composites that perturbs about certain optimal dispersions. Recently, Segurado and Llorca [21] and Böhm et al. [5] have assessed the effective coefficients of randomly distributed spherical particles using the RSA algorithm and compared those with different analytical methods. Also Gusev [12,13,17,22] made experiments of randomly distributed short fiber composites and compared them with numerical results. A good agreement has found. But, because of the limited amount of literature which deals with randomly distributed spherical particle reinforced composites is available and restricted to lower volume fractions of particles, we have been motivated to work in this direction. In our opinion micro–macro mechanical approaches offer new insights in the material behavior of such spatially distributed fiber and particle reinforced composites and may result in new procedures to develop realistic material models for design and optimization purposes.

In this paper we focused on the evaluation of effective material properties of randomly distributed spherical particle reinforced composites using numerical homogenization techniques (finite element based RVE) with periodic boundary conditions. An attempt was made to generate higher volume fraction RVE models with different sizes of spherical particles. Several investigations were conducted in order to determine the influence of the size of the spherical particles and of the RVE on effective material properties of such composites, which is explained in the subsequent sections. Section 2 explains the procedure of generating randomly distributed spherical particle reinforced composites, numerical homogenization techniques and periodic boundary conditions. Section 3 discusses the results on the spherical particle reinforced composites. In Section 4, conclusions of this paper are presented.

2. Numerical homogenization of randomly distributed spherical particle composite

2.1. Generation of the RVE

The homogenized effective elastic constants of the composites were obtained by the finite element analysis of a periodic cubic RVE of volume $L^3$ consisting of randomly distributed non-overlapping hard spherical particles. The RVE was generated using the RSA algorithm [32] modified to provide for a user specified minimum distance between neighboring inclusions and for the periodicity on opposite boundary surfaces. The centre distance between spherical particle $i$ and all other particles, which are previously accepted $j = 1, \ldots, i - 1$ have to exceed a minimum value $(2r + s)$, where $r$ is the radius of the spherical particle and $s$ is the minimum distance between any two spherical particles, imposed by the practical limitations for creating an adequate finite element mesh. If any surface of the spherical particle $i$ cuts any of the cubic RVE surfaces, this condition has to be checked with the spherical volumes on the opposite surfaces because the microstructure of the composite is periodic. Also the surface of the spherical particles should not be very close to the cubic RVE surfaces as well as to the corners of the RVE in order to avoid
the presence of distorted finite elements during meshing. The RSA algorithm with the combination of the above conditions was used to generate the RVE models of a composite up to desired volume fractions. In general with the identical spherical particles, these algorithms can generate up to 30% volume fractions. For higher volume fractions, different sizes of spherical particles were used and these were deposited inside the RVE in descending manner. With this approach the volume fraction achieved was up to 60% with minimum distortion of the finite elements and the adequate mesh. Fig. 1(a) shows the randomly distributed spherical particles RVE model.

2.2. Numerical homogenization technique

The mechanical and physical properties of the constituent material are always regarded as a small-scale/microstructure. One of the most powerful tools to speed up the modelling process, both the composite discretization and the computer simulation of composites in real conditions, is the homogenization method. The main idea of the method is to find a globally homogeneous medium equivalent to the original composite, where the strain energy stored in both systems is approximately the same. In the numerical homogenization technique, the common approach to model the macroscopic properties of fiber or particle composites is to create the RVE that should capture the major features of the underlying microstructure. The finite element RVE models of the spherical particles reinforced composites are shown in Fig. 1(b) and (c).

All finite element calculations were made with the commercial FE package ANSYS. The matrix and the spherical particles were meshed with 10 node tetrahedron elements with full integration. To obtain the homogenized effective material properties, periodic boundary conditions were applied to the RVE by coupling opposite nodes on the opposite boundary surfaces. In order to apply these periodic boundary conditions on the FE model of the RVE, the meshes on the opposite boundary surfaces must be the same. For each pair of displacement components at the two corresponding nodes with identical in-plane co-ordinates on two opposite boundary surfaces, a constraint equation (periodic boundary condition Eq. (4) in the next section) was imposed. For this purpose any three faces of the RVE along the three co-ordinate directions are meshed with the triangular area elements, and these area meshes are copied to the opposite faces of the RVE. Then, meshing the volumes of the spherical particles and of the matrix is performed. Finally the area mesh is deleted. In this way identical meshes were achieved on the opposite boundary surfaces of the RVE as shown in Fig. 1(c), which is required in order to apply periodic boundary conditions. The mesh size used was fine enough to represent accurately the geometry of the spheres and the matrix. But applying these constraint equations on all opposite nodes at opposite boundary surfaces interactively in ANSYS is a very time consuming task due to a great number of coupled nodes. So we automated this process by using the ANSYS Parametric Design Language (APDL) to generate all required constraint equations. Furthermore, we used the APDL for the evaluation of needed average strains and stresses and evaluated the effective material properties in the end. The developed APDL-Scripts in combination with the ANSYS batch processing provide a powerful tool for the fast calculation of homogenized material properties for composites with a great variety of inclusion geometries.

2.3. Periodic boundary conditions to RVE

Composite materials can be represented as a periodic array of the RVEs. Therefore, the periodic boundary conditions must be applied to the RVE models. This implies that each RVE in the composite has the same deformation mode and there is no separation or overlap between the neighboring RVEs after deformation. These
periodic boundary conditions on the RVE surfaces described in Cartesian co-ordinates are given by Suquet [27]:

\[
\mathbf{u}_i = \mathbf{S}_{ij} \mathbf{x}_j + v_i.
\]  

(1)

In the above Eq. (1) \( \mathbf{S}_{ij} \) are the average strains, \( v_i \) is the local fluctuation on the boundary surfaces, which is generally unknown and is dependent on the applied global loads. A more explicit form of the periodic boundary conditions, suitable for the RVE models can be derived from the above general expression. For the RVEs, the displacements on a pair of opposite boundary surfaces (with their normal along the \( x_j \) axis) are

\[
\begin{align*}
\mathbf{u}^{K^+}_i &= \mathbf{S}_{ij} x_j^{K^+} + v_i^{K^+}, \\
\mathbf{u}^{K^-}_i &= \mathbf{S}_{ij} x_j^{K^-} + v_i^{K^-},
\end{align*}
\]

(2) \( (3) \)

where index \( K^+ \) means along the positive \( x_j \) direction and \( K^- \) means along the negative \( x_j \) direction on the corresponding opposite boundary surfaces. The local fluctuations \( v_i^{K^+} \) and \( v_i^{K^-} \) around the average macroscopic value are identical on two opposite faces of the RVE due to the periodic conditions. So, the difference between the above two equations is the applied macroscopic strain condition

\[
\mathbf{u}^{K^+}_i - \mathbf{u}^{K^-}_i = \mathbf{S}_{ij} (x_j^{K^+} - x_j^{K^-}).
\]

(4)

It is assumed that the average mechanical properties of the RVE are equal to the average properties of the particular composite. The average stresses and strains in the RVE are defined by

\[
\begin{align*}
\mathbf{S}_{ij} &= \frac{1}{V} \int_V \mathbf{S}_{ij} \, dV, \\
\mathbf{T}_{ij} &= \frac{1}{V} \int_V \mathbf{T}_{ij} \, dV,
\end{align*}
\]

(5) \( (6) \)

where \( V \) is the volume of the periodic RVE.

The effective material coefficients can be calculated by using the constitutive equations of the material properties as the ratio of corresponding average stresses and average strains by applying appropriate boundary conditions along with these periodic boundary conditions. For further details see our previous articles [2–4, 19].

3. Results and discussions

Three-dimensional spherical particle RVE models were created using the modified RSA algorithm. With identical spherical particles (monodisperse) using this algorithm, it is possible to generate up to 30% volume fractions. It is not possible to generate higher volume fraction RVE models because of the jamming limit. In order to generate higher volume fraction RVE models, a different approach was used i.e., with different sizes of the spherical particles and these should be deposited inside the RVE in a descending manner i.e., first deposit the all possible largest particles in the RVE, then deposit the next possible largest particles inside the RVE by preserving the minimum distance between any two particles and periodicity on the opposite boundary surfaces. This process would be continued up to achieving the desired volume fraction or maximum possible volume fraction for the given size of spherical particles and minimum distance between the particles. Several investigations were made in order to determine the influence of the size of the spherical particles on the effective material properties of these composites. The results showed that the influence of the size of the spherical particles on the effective material properties was not significant in the linear elastic case. Taking this fact into consideration and, using different sizes of the spherical particles, the effective material properties of these composites were obtained for up to 60% volume fractions. With this algorithm, even higher volume fractions can be achieved, but there are some restrictions in the FE packages like the limited maximum number of the elements that can support the analysis and the presence of distorted finite elements if the gap between any two particles is reduced to a certain distance. The material properties used for the analysis are for the matrix material \( E_m = 70 \) GPa, \( v_m = 0.3 \) and for the spherical particles \( E_f = 450 \) GPa, \( v_f = 0.17 \) [5]. The results of the numerical methods were compared with different analytical methods which are Torquato’s third order approximations [31] (TOA), rigorous three point bounds [23, 24, 31] (3PB), Hashin–Strikman two point bounds [15] (HS), Mori–Tanaka estimates [25] (MTM), self-consistent method [16, 20] (SCM) and generalized self-consistent method [6] (GSCM). Also several investigations were made to determine the effect of the size of the RVE on the effective material properties of these composites.

3.1. The effect of the size of the RVE on the effective material properties

The RVE is generally regarded as a volume \( V \) of a heterogeneous material that is sufficiently large to be statistically representative of the composite, i.e., to effectively include a sampling of all micro-structural heterogeneities that occur in the composite [18]. Such a RVE should exhibit the properties of the composite medium and these properties can be shown to be relatively insensitive to the macroscopically uniform boundary conditions. In other words, they fluctuate about a mean with a wave length small compared to the dimensions of the volume. Unlike as in many analytical methods, in the case of numerical homogenization techniques, one must take care of the minimum size of the RVE (with spatially distributed particles or short fibers), which is required to give appropriate effective material
properties of a macroscopic composite structure. If the size of the RVE considered is less than the minimum size required, it may lead to a wrong prediction of effective material properties. Numerical homogenization techniques can help in determining the critical size of the RVE. In order to determine the minimum size of RVE, identical spherical particles were considered and by changing the size of the cubic RVE, the effective material properties were obtained for 30% volume fraction. Fig. 2 explains the variation of effective material properties regarding the change in size of the RVE. Here onwards in all graphs, the legend ‘ERROR-FEM’ represents the fluctuation (error) of effective material properties around the mean value, which were obtained from the ensemble averages of the effective properties of different RVE samples. The effective material properties ($E$—Young’s Modulus, $v$—Poisson’s ratio and $G$—shear modulus) shown in Fig. 2 were obtained from the ensemble average of five different RVE samples at each length of the cubic RVE. The error was around 5% if we consider $L/D \geq 2$ where $L$ is the length of the cubic RVE and $D$ is the diameter of spherical particles which were of constant size ($D = 0.24$ mm) in this case. If $L/D \geq 10/3$, the error was around less than 1%. This is in agreement with Drugan and Willis [7,8] predictions. The differences in effective material properties between $L/D \geq 10/3$ and $L/D \geq 5$ were very small and the error was less than 1%. From these results, it can be observed that it is sufficient to consider the size of the RVE as $L = (10/3)D$.

3.2. The effect of the size of the spherical particles on the effective material properties

Different studies were made to determine the effect of the size of the spherical particles on effective material properties of these composites. Fig. 3 explains the effect of the size of the spherical particles on the effective material properties. Here the size of the RVE remains constant and by varying the size of the particles, effective material properties were obtained at 30% volume fraction. From Fig. 3, it can be observed that there were no significant variations by changing the size of the spherical particles on the effective material properties. Although slight variations can be observed in the effective material properties with change in the size of the particles, these might be due to greater number of particles inside the RVE by reducing its size for the same 30% volume fraction. As explained in the previous section, if $L/D \geq 10/3$ the variations in the effective material properties were very small and less than 1%. This can also be observed in Fig. 3 for all effective material properties.

Fig. 2. Variation of the effective material properties with change in RVE size (a) Young’s modulus, (b) Poisson’s ratio and (c) shear modulus.
Also the numerical homogenization techniques were applied to two different types of the RVE models, one with the same size of the spherical particles and another with random size of spherical particles at 30% volume fraction. The following table explains the differences between these two cases. In both cases ten different RVE samples were considered and the effective material properties were obtained from the ensemble averages of the different samples. From Table 1, it can be observed that the differences in effective properties are negligible between the random size of spherical particles and the same size of spherical particles.

Fig. 4 shows the comparison between the effective material properties of the numerical homogenization techniques and Torquato’s third order approximations for monodisperse and polydisperse spherical particles. From Fig. 4 it can be observed that the differences of the effective material properties were very small between the monodisperse and the polydisperse spherical particle reinforced composites. From these studies it can be concluded that the effective material properties of the spherical particle reinforced composites depend only on the volume fraction. The size of the particles has not a significant influence on the effective material properties in the linear elastic case.

### 3.3. Comparison of calculated effective material properties with other methods from literature

The elastic modulus ($E$), Poisson’s ratio ($v$), shear modulus ($G$) and bulk modulus ($K$) were evaluated for different volume fractions from 10% to 60%. Five different RVE models with randomly distributed spherical particles were considered for each volume fraction, and subjected to uniaxial tensile as well as the shear deformation along the three axes of co-ordinates. The ensemble average of the effective material properties at each volume fraction were considered as effective material properties of the total composite at that particular volume fraction with a certain error. The effective material properties, which were obtained using the numerical homogenization technique, were very close to the TOA approximation of the monodisperse spherical particle composites and were within the three point bounds as well as the

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Volume fraction = 30%</th>
<th>$E$ (GPa)</th>
<th>$v$</th>
<th>$G$ (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Same size</td>
<td>115.11</td>
<td>0.2651</td>
<td>44.00</td>
<td></td>
</tr>
<tr>
<td>Random size</td>
<td>114.31</td>
<td>0.2670</td>
<td>43.79</td>
<td></td>
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Fig. 3. Variation of the effective material properties with change in size of the spherical particles (a) Young’s modulus, (b) Poisson’s ratio and (c) shear modulus.
Fig. 4. Comparison of the effective material properties with monodisperse and polydisperse analytical approximations (a) Young’s modulus, (b) Poisson’s ratio, (c) shear modulus and (d) bulk modulus.

Fig. 5. Comparison of the effective material properties with different analytical methods (a) Young’s modulus, (b) Poisson’s ratio, (c) shear modulus and (d) bulk modulus.
two point Hashin–Shtrikman bounds. The convergence of the effective material properties was rapid and the error of the numerical simulations was very small even at 60% volume fraction due to the periodic boundary conditions applied to the RVE. The error between the TOA and the FEM was very small in case of shear modulus and bulk modulus, which was about less than 1%. In the case of the Young's modulus and Poisson's ratio it was around maximum of less than 3%. When compared with three point bounds, the numerical results were much closer to the upper bound for the case of the Young's modulus and the Poisson's ratio, and closer to the lower bound in the case of shear modulus. The results of analytical methods of MTM and GSCM were same for the spherical particle composites and differences between numerical results and GSCM were first noticeable at 30% volume fraction and at 60% volume fraction, it is about 8%. The SCM method overestimated the effective material properties at higher volume fraction and at 60% volume fraction, the error between numerical method and SCM was about 13%. Evidently all results of numerical method and analytical methods were in between Hashin and Shtrikman bounds and most results of different methods were closer to the lower bound (see Fig. 5).

The isotropy of the RVE models was achieved using the modified RSA algorithm and this was explained in terms of the effective material properties which were obtained using three co-ordinate directions for different volume fraction as shown in Fig. 6. The effective material properties, which were obtained using the three co-ordinate directions, are the same and the error is less than 1%.

4. Conclusions

The numerical homogenization tools have been developed for the evaluation of the effective material properties of short fibers and spherical particles reinforced composite structures. The effective material properties of the spherical particle reinforced composites obtained using these tools were compared with the results of different analytical methods. Our numerical predictions were in between the three point bounds and close to the results of Torquato’s third order approximation. We also studied the influence of the size of the RVE on the effective material properties. From these studies it can be concluded that \( L/D \geq 10/3 \) is sufficient to predict the effective material properties with a small error. Several investigations were made to determine the influence of the size of the spherical particles on the effective material properties. The results showed that the effective material properties depend mainly on the volume fraction. There were no significant variations with respect to the change in size of the particles. This
statement is valid for linear elastic case for the evaluation of the effective material properties only. There may be some influence in non-linear case, debonding and damage predictions. Further investigations have to be carried out to determine the influence of the size of the spherical particles on the behavior of composites at macrolevel. A generalized procedure has been developed to calculate different effective coefficients for all desired volume fractions based on the ANSYS Parametric Design Language. This tool reduces the manual work and time and can be used as a template to evaluate the effective coefficients of randomly distributed spherical particle reinforced composites up to 60% volume fraction. Finally, the tool which we developed can be applied to any number of phases i.e., there are no restriction regarding the number of materials, geometry and material symmetry and this can be used effectively to determine material coefficients of different types of fiber and particle reinforced composites.

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References