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Time domain finite element estimates of dynamic stiffness of viscoelastic composites with stiff spherical inclusions

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ABSTRACT

A weighted residual time domain unstructured mesh finite element approach was used to obtain numerical estimates for the dynamic stiffness of medium and high stiffness contrast composites consisting of viscoelastic epoxy matrix filled with stiff spherical inclusions. Both random and regular microstructure composites were studied. It was shown that over the broad temperature and inclusion fraction ranges studied, the generalized self-consistent model (Christensen and Lo, 1979) provided accurate predictions for the effective dynamic stiffness of random composites obeying classical Percus–Yevick hard sphere statistics. Assuming sphere flocculation, we have studied the role of microstructural effects and found that upon increasing stiffness contrast, they become progressively more important for both the effective storage and loss moduli. As a consequence, for the reliable predictions of effective stiffness of high contrast composites with rubber like matrices, one should necessarily use homogenization models accounting not only for the inclusion concentration but also for the finer microstructural details. However, it was also found that for such high stiffness contrast viscoelastic composites, the ratio of their effective storage and loss shear moduli (the damping factor) remains essentially unchanged as compared to that of pure unfilled matrix so it is the invariability of the damping factor that can be viewed as a special signature of underlying micromechanical mechanism of reinforcement.

1. Introduction

Viscoelastic materials exhibit time and rate dependent mechanical responses in which they store the energy as an elastic solid and dissipate it as a viscous fluid. The theory of viscoelastic materials is well developed and it has been thoroughly treated by Gross (1953), Christensen (1971), Pipkin (1972) and Ferry (1980), among others. For linear viscoelastic materials under steady state harmonic conditions, the stress and the strain are related by complex viscoelastic moduli $\mathbf{C} = \mathbf{C}' + i\mathbf{C}''$, where $\mathbf{C}'$ and $\mathbf{C}''$ are termed as the storage and loss moduli, respectively. The loss tangent of a particular component C is defined by $\tan\delta = C''/C'$, where δ denotes the phase angle by which the strain lags behind the stress in steady state harmonic oscillations. The elastic-viscoelastic correspondence principle allows one to convert static elastic solutions to steady state harmonic viscoelastic solutions simply by replacing the static elastic moduli by the corresponding complex viscoelastic moduli and treating the underlying field variables as complex harmonic variables. The correspondence principle is equally applicable to both homogeneous and heterogeneous materials (Hashin, 1965 and 1970) and it has been broadly used for obtaining viscoelastic solutions.

Viscoelastic composites consisting of a polymer matrix filled with stiff fillers are widely used in vibration damping and noise reduction applications (Jones, 2001; Ward and Sweeney, 2013). Commonly, such composites have a stiffness contrast of the order of 10^2 – 10^6 and it is an interesting problem to assess whether one can use traditional micromechanics based models for reliable predictions of the dynamic stiffness of such high stiffness contrast composites. In the last decades, different research groups have addressed this issue. In particular, Shaterzadeh et al. (1998) studied the temperature dependence of dynamic shear modulus $G = G' + iG''$ of epoxy matrix composites reinforced by spherical glass spheres and compared their experimental results with predictions of the Generalized Self-Consistent (GSC) model introduced by Christensen and Lo (1979). For obtaining model predictions, measured dynamic matrix shear modulus was used but it was assumed without justification that the required dynamic Poisson's ratio of the matrix be real valued and have the same temperature dependence as the matrix storage modulus G'. Composites with surface treated and untreated spherical glass microbeads were studied. It was found that for medium contrast composites with solid-like epoxy matrix, the surface treatment had practically no

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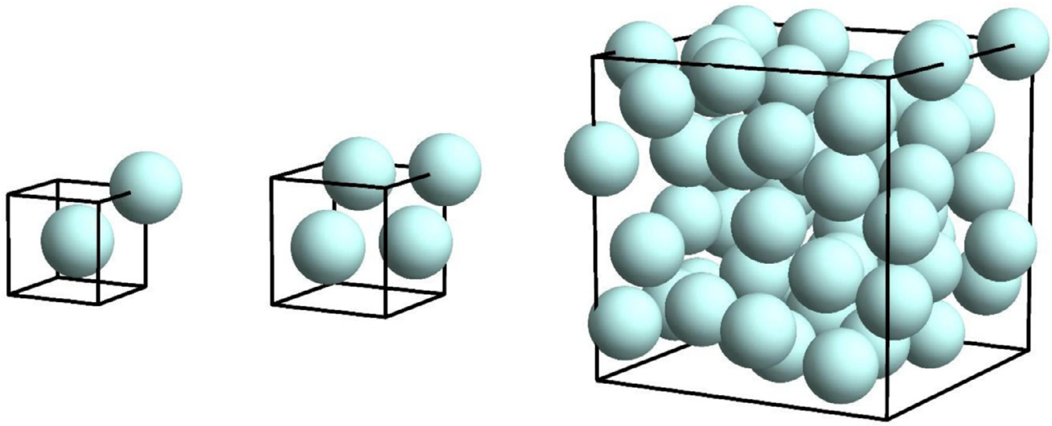

Fig. 1. Periodic computer models with non-overlapping identical spheres dispersed at a volume fraction of $c = 0.3$. From left to right: a body-centered cubic (bcc), a face-centered cubic (fcc) and a random Monte Carlo model with 64 spheres.

effect on the effective shear modulus while for high contrast composites with rubber-like epoxy matrix, the effect of surface treatment was significant. It was observed that the surface treatment caused changes in the composite microstructure, with the treated beads being more uniformly distributed than untreated ones. This experimental study illustrates some general problems encountered with experimental validation of homogenization models, namely, incomplete set of constituent properties and the important role of interfacial phenomena at matrix/filler interfaces that affect effective stiffness both through incomplete bonding and also through alternations in the composite microstructure. For viscoelastic composites with nanosized inclusions, the inclusion surface area per unit volume increases greatly and the interfacial effects become prominent so for reliable predictions, one should necessarily explicitly account for the presence of the interfacial layers (see, e.g., Shashidhar and Shenoy, 2001; Berriot et al., 2002, 2003; Heinrich and Klüppel, 2002; Gauthier, 2004; Ozmusul et al., 2005; Goertzen and Kessler, 2008; Fritzsche and Klüppel, 2011; Litvinov et al., 2011; Stockelhuber et al., 2011; Pourhossaini and Razzagni-Kashani, 2014; Underwood and Kim, 2014).

Various homogenization methods have been proposed for predicting the effective stiffness of composite materials from that of the constituents and materials microstructure, as reviewed by Christensen (2005), Milton (2002) and Torquato (2002). For linear elastic composites, a number of finite element studies were performed using random microstructure computer models and the results were used to assess the predictive potential of the models (Gusev 1997; Tucker and Liang, 1999; Gusev et al., 2000, Böhm and Han, 2001; Gusev, 2001; Böhm et al., 2002; Segurado and Llorca, 2002 and 2006; Lusti, et al., 2002; Kanit et al., 2003; Lusti and Gusev, 2004; Kari et al., 2007; Gusev, 2016). Compared to laboratory experiments, computer simulations allows one to control both the constituent properties, the interfacial bonding and the materials microstructure. Recently, a comprehensive validation program has been carried out by Ghossein and Lévesque (2012, 2014 and 2015). A grid based finite element method with a fast Fourier transform procedure of Moulinec and Suquet (1998) accelerated with the algorithm of Eyre and Milton (1999) was employed to obtain numerical predictions for a great variety of composites with spherical and ellipsoidal inclusions and the results were compared with predictions of various commonly used homogenization models.

Grid based homogenization methods can be remarkably fast and it has been demonstrated that using sufficiently large grids with up to 256^3 pixels, one can obtain reliable predictions for random microstructure composites with low and medium stiffness contrasts (Garboczi and Day, 1995; Gusev, 2007; Ghossein and Lévesque, 2012, 2014 and 2015; Brenner and Suquet, 2013; Garboczi and Kushch, 2015). However for random composites with a stiffness contrast of the order of 10^2 and higher, progressively larger grids are required for reliable estimates. It has been shown that for such composites, unstructured mesh finite element approach becomes advantageous (Gusev, 2016).

In this work we employ a time domain unstructured mesh finite element method to study the effective dynamic stiffness of a viscoelastic composite consisting of a model epoxy resin matrix reinforced by both random and regular dispersions of non-overlapping identical silica spheres. We use periodic models with up to a hundred spheres and study composites with a stiffness contrast from 30–3000 corresponding to the whole spectrum of silica/epoxy stiffness contrasts commonly observed in the temperature scans over the glass transition region of a typical epoxy resin matrix. Our main goals are to assess the predictive potential of the GSC model for high stiffness contrast viscoelastic composites and to understand the role of microstructural effects in the effective dynamic stiffness of such high contrasts composites.

2. Time domain finite element homogenization procedure

2.1. Periodic models

We have studied periodic computer models with non-overlapping identical spheres. Perfect bonding was assumed at the matrix/inclusion interfaces. Both regular and random microstructure models were studied, see Fig. 1. Unbiased Monte Carlo runs were employed to generate random models obeying the Perckus–Yevick hard sphere statistics. Delaunay tessellation was used to create unstructured morphology-adaptive periodic meshes with tetrahedral elements. Further details on the Monte Carlo and mesh generation procedures are given elsewhere (Gusev, 2001, 2016).

2.2. Spatial discretization

We consider viscoelastic materials with linear constitutive relations of the form

$$\boldsymbol{\sigma} = \mathbf{D}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}_0) + \mathbf{Q}(\dot{\boldsymbol{\varepsilon}} - \dot{\boldsymbol{\varepsilon}}_0) \tag{1}$$

where $\mathbf{D}$ and $\mathbf{Q}$ are, respectively, position-dependent elasticity and friction matrices with some appropriate time independent materials coefficients, ε and $\boldsymbol{\sigma}$ instantaneous strain and stress, respectively, and ε_0 uniform harmonic initial strain defined by

$$\boldsymbol{\varepsilon}_0(t) = \mathbf{E}\sin(\omega t) \tag{2}$$