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Micromechanics models of particulate filled elastomer at finite strain deformation

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ABSTRACT

A micromechanics-based model is proposed for the finite strain deformation of filled elastomers based on generalized Eshelby's tensor and Mori–Tanaka's method. The present formulation leads to a clear explanation of the constraint effect of rubber–like matrix on the inclusions. Comparisons with experiments and other micromechanics models are conducted. It is observed that an improvement in predictive capability for the composite with randomly dispersed particles was achieved by the present method. Based on the latest experiment of single molecular chain, a compact network model is fatherly developed to reflect the microstructure effect on the stress–strain relations of rubbery polymer and the resulting composites.

1. Introduction

Rubber-like materials have been widely applied in various industry fields due to their large-deformability. In most of applications, particles or fibers are often introduced into rubber-like materials to make composites in order to improve their mechanical properties. To study the overall performance of hyper-elastic composites, the mechanical behaviors of the composites should be predicted on the basis of their equivalent properties, which are determined by their inherent microstructures [1]. Therefore, a powerful homogenization method is very necessary and impending for such kind of composites. So far, large deformation behaviors of such hyper-elastic materials have been investigated by many approaches, which are grossly classified into micro-mechanics method and network models. Micromechanics studies are first briefly reviewed. Mullins and Tobin [2] systematically measured the stress–strain relations of vulcanized rubbers containing carbon black, and adopted a strain amplification factor to describe the enhanced elastic behavior of filler-reinforced rubbers. They qualitatively interpreted the reinforcement effect of carbon black on the mechanical properties of rubbers. By using Mullins and Tobin's conclusion, Qi and Boyce [3] predicted the effective stress–strain response of hyper-elastic composites. Frankly speaking, although a good agreement between the predictions and experiments was achieved, the interaction between particles and matrix was not embodied fully. Bergstrom and Boyce [4] have investigated the influence of filled particles on the equilibrium stress–strain response of rubber matrix composites by means of testing and FEM. They proposed a new concept based on first strain invariant instead of strain amplification, and found that the new concept could better predict the experimental results. To the best of the authors' knowledge, the large deformation problem of the composites was not really solved until Lopez-Pamies and Ponte Castaneda [5,6] developed the second-order homogenization method. Their method can be regarded as a mechanics-based analytical model to a great extent. However, the specific application of this powerful method cannot but to solve many partial differential equations and code programming. In additional, such method is so complicated and confined to solve some plane strain problems so far. Bouchart et al. [7,8] proposed an accurate algorithm of the second-order method to study highly compressible hyper-elastic composites. Yin et al. [9] developed an effective hyperelastic constitutive model for particle-filled elastomers based on Eshelby's phase transform idea. Although the interaction between particle and matrix is fully considered, some deductions are arguable. For instance, they still adopted Eshelby's tensor of small strain in dealing with large deformation problem. Eshelby's tensor [10] and Mori–Tanaka method [11] are well known as the foundation of micromechanics of heterogeneous materials for their strong physics meaning, and so compact that be easily acceptable. Nemat-Nasser [12,13] firstly extended the classic Eshelby single inclusion theory to finite strain deformation, and obtained the necessary formula of Eshelby's tensor for large deformation. Unfortunately, the validity of their theoretic frame is not checked by the prediction of hyper-elastic composites yet. We do not know whether Eshelby's equivalent inclusion concept fit to large deformation of heterogeneous material or not. In a whole, it is very challenging to establish the deformation compatible conditions between particle and matrix

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during the finite strain deformation, but which is a necessary prerequisite for us in understanding the effective mechanical behaviors of hyper-elastic composites. Many network models are increasely adopted in an attempt to clearly know the inherent morphology evolution. Wang et al. [14] recently measured single polymer chain force-extension behavior by Atomic Force Microscopy (AFM) and confirmed the validity of the worm-like chain (WLC) model. Miehe et al. [15] proposed a micro-sphere model representing a continuous distribution of chain orientations in space, and developed a new micro-mechanically based network model for rubber-like materials. Their models excellently replicated the corresponding experimental data and revealed the inherent microstructure evolution. Böl and Reese [16] presented a finite element method taking account of molecular chain deformation and the interaction between the chains, successfully predicted the large deformation of rubbers. Drozdov and Dorfmann [17] emphasized the mechanical energy instead of entropy theory of polymer chains at the finite strain deformation, and developed a micro-mechanics model for pure polymer and the composites, where the role of particles is equivalent to the cross-links. Brieu and Devries [18] developed a numerical non-incremental algorithm well suited for the homogenization of nonlinearly elastic composites. Abadi [19] presented a homogenization procedure to predict the effective shear response of heterogeneous materials at large deformation. The homogenization procedure is implemented to evaluate shear response of two specific heterogeneous materials, elastomeric composite and reinforced viscoelastic fluid.

The main objective of this contribution is to develop micromechanics-based models in homogenizing the mechanical behaviors of hyper-elastic materials containing inclusions. Main emphasis is played on the interpretation of the synergism mechanism between rubber matrix and inclusions. The accuracy of the homogenization model is checked by the comparison with the experiments and analytic models available in literatures. Some discussions for continuum mechanics method and network model to the heterogeneous systems are expanded subsequently.

2. Eshelby's tensor and Mori–Tanaka's formula

2.1. Constituent materials

The hyper-elastic constituent materials are assumed to be isotropic. The large-deformation behaviors are characterized by the strain energy density functions $W^{(r)}$ (r = P, M), which are often expressed in terms of three invariants, I_1, I_2 and I_3, of the right Cauchy–Green deformation tensor $\mathbf{C} = \mathbf{F}^T \cdot \mathbf{F}$, here the operation $\langle\cdot\rangle$ denotes dot product between second-order tensors. $\mathbf{F} = \partial\mathbf{x}/\partial\mathbf{X}$ is the deformation gradient tensor, where $\mathbf{x}$ is the current position and $\mathbf{X}$ the location at the reference configuration for a arbitrary material point, and its transpose matrix denoted by $\mathbf{F}^T$.

For the matrix phase, Yeoh's constitutive formula is adopted [20], and given by

$$W^{(\mathrm{M})}(\mathbf{F}) = A_{10}(I_1 - 3) + A_{20}(I_1 - 3)^2 + A_{30}(I_1 - 3)^3 \tag{1}$$

where A_{10}, A_{20} and A_{30} are constants determined by fitting with the experimental data.

For the particle phase, the following Neo-Hookean hyper-elastic model is used,

$$W^{(\mathrm{P})}(\mathbf{F}) = B_{10}(I_1 - 3) + (J - 1)^2/B_{20} \tag{2}$$

where $J = \det(\mathbf{F}) = \sqrt{I_3}, B_{10}$ and B_{20} are material constants. Rigid particles are modeled by setting a high value of B_{10} and a small value of B_{20} while the void is obtained with B_{10}=0 and $B_{20} \to \infty$. In the limit case of small strains, Eq. (2) can characterize the linear and isotropic materials with initial bulk modulus $K_P = 2/B_{20}$ and initial shear modulus $\mu_P = 2B_{10}$.

2.2. Eshelby's tensor for finite strain

Let a ellipsoidal region Ω in a material space V undergo a phase transformation, if Ω were free from the constraint imposed by the surrounding material, it would attain a constant transformation deformation gradient $\mathbf{F}^*$. The resulting deformation gradient $\mathbf{F}$ of Ω in the presence of the constraint from the surrounding matrix is spatially uniform when V is homogeneous and unbounded [10]. Therefore, $\mathbf{F}$ is related to the transformation deformation gradient $\mathbf{F}^*$ by

$$\mathbf{F} = \mathcal{L} : \mathbf{F}^* \tag{3}$$

here, the above notation $\langle:\rangle$ is double contraction between fourth-order and second-order tensors. $\mathcal{L}$ is called the generalized Eshelby's tensor, and cannot be solved analytically for the finite strain problem. For an ellipsoidal Ω whose principal axes are parallel to the global coordinate axes x_i with length $2a_i$, ($i = 1 \sim 3$), in the matrix with stiffness C_{ijkl}, the Eshelby's tensor is determined by,

$$\mathcal{L}_{iAjB} = \frac{1}{4\pi a_A a_B}\int_{S(\zeta)} \frac{N_{ij}(\zeta)}{\mathcal{D}(\zeta)}\zeta_A\zeta_B dS(\zeta)\ (A, B \text{ not summed}) \tag{4.a}$$

$$N_{ij}(\zeta) = \frac{1}{2}e_{ikm}e_{ikm}K_{kl}(\zeta)K_{mn}(\zeta) \tag{4.b}$$

$$\mathcal{D}(\zeta) = \det|K_{ij}(\zeta)| \tag{4.c}$$

$$K_{ij}(\zeta) = C^*_{AiBj}\zeta_A\zeta_B\ (A,\ B \text{ summed}) \tag{4.d}$$

$$C^*_{AiBj} = \frac{1}{a_A a_B}C_{AiBj}\ (A,\ B \text{ not summed}) \tag{4.e}$$

where $dS(\zeta)$ is an elementary surface of a unit sphere, $S(\zeta)$, in the ζ-space. In the following section, the particle shape is supposed to be ellipsoidal with $a_3 > a_1 = a_2$, and aspect ratio defined as $\alpha = a_3/a_1$.

2.3. Mori–Tanaka's formula

A two-phase composite system consists of a matrix and particulate phase. Perfect bonding is assumed between the constituents. The particle phase consists of ellipsoidal particles that are of the same shape but can be of different size; they can be aligned or have a random orientation in space.

Suppose a representative volume element (RVE) subjected to linear boundary displacements as follows:

$$\mathbf{x} = \mathbf{F}^0 \cdot \mathbf{X} \tag{5}$$

where $\mathbf{F}^0$ is uniform deformation gradient along the outer boundary. Following Benveniste's formula of Mori–Tanaka mean field concept [21],

$$\mathbf{F}^{\mathrm{M}} = \mathbf{F}^0 + \widetilde{\mathbf{F}}, \quad \mathbf{F}^{\mathrm{P}} = \mathbf{F}^0 + \widetilde{\mathbf{F}} + \mathbf{F}^{pt} \tag{6}$$

where $\widetilde{\mathbf{F}}$ is the average perturbed deformation gradient in the matrix due to the presence of the inclusions, and $\mathbf{F}^{pt}$ is the perturbed part of the same quantity in a typical inclusion with respect to the matrix. Subsequently, the corresponding expression of Mori–Tanaka method is written as

$$\mathbf{C}_{\mathrm{P}} : (\mathbf{F}^0 + \widetilde{\mathbf{F}} + \mathbf{F}^{pt}) = \mathbf{C}_{\mathrm{M}} : (\mathbf{F}^0 + \widetilde{\mathbf{F}} + \mathbf{F}^{pt} - \mathbf{F}^*) \tag{7}$$

2.4. Incremental theory formulation

Based on Eshelby's equivalence principle, together with Mori–Tanaka's mean field concept, the incremental first P–K stress in the particle $d\mathbf{S}^P$ is given by