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A micromechanics-based incremental damage model for carbon black filled rubbers

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ABSTRACT

This paper is to develop a simple micromechanics-based model taking account of progressive damaging for carbon black (CB) filled rubbers. The present model constitutes of the instantaneous Young's modulus and Poisson's ratio characterizing rubber-like material, a double-inclusion (DI) configuration considering the absorption of rubber chains onto CB particles, and the incremental Mori-Tanaka formula to compute the effective stress–strain relations. The progressive damage in filled rubbers is described by the DI cracking, which is represented by the remaining load–carrying capacity. The present predictions are capable of embodying the well-known *S*-shaped response of filled rubbers, and also verified by the comparison with the experimental and analytical results. Moreover, strain localization effect is clearly demonstrated by finite element method (FEM) simulations, and reaches a decisive interpretation to the complicated synergic micro-mechanisms between hard fillers and soft phase in such flexible composites.

1. Introduction

Filled rubbers have been widely applied in various industrial fields due to their outstanding large-deformability. Costa et al. [1] studied the multi-walled carbon nanotubes (MWNT) filled copolymer, and found that the composites possess the electro-mechanical properties with high sensibility at larger strain, and could be used for the electromechanical sensors for large strains applications. Anuar and Zuraida synthesized the thermoplastic elastomer composite reinforced with kenaf fiber [2]. Jovanovic investigates the effect of carbon black (CB) filler on the cure kinetics, mechanical properties, morphology and thermal stability of rubber blends [3]. The resulting macroscopic properties of filled rubbers mainly rely on the inherent microstructure characteristics, and the interrelationship should be established.

Many analytical works have been conducted in explaining such highly nonlinear mechanical behaviors including the damage effect. Firstly, micromechanics approaches are briefly reviewed. Mullins [4], Qi et al. [5] systematically measured the stress–strain relations of vulcanized rubbers containing CB powders, and introduced a strain amplification factor $\chi = E/E_M = 1 + 2.5f_P + 14.1f_P^2$ (E_M–matrix modulus, f_P–particle volume fraction) to describe the enhanced elastic property. Although a good agreement was achieved between the predictions and experiments, the third coefficient depends on the author's appetite and could not be rigorously deduced. Bergstrom et al. [6] investigated the influence of hard particles on the mechanical response of filled rubbers by experiment and FEM. A new concept was proposed based on the first strain invariant instead of strain amplification, and could predict the experiments very well. To the best of authors' knowledge, the finite deformation of composites was not really solved until Ponte Castaneda [7,8] developed a second-order homogenization method, and adopted the optimization computation to solve the energy minimum problem to determine the appropriate deformation compatible conditions between two phases. Unfortunately, solving so many partial differential equations in their model is a challenging work, and only confined to the plane strain case up to now. Recently, the predictions based on the second order theory still could not successfully reproduce FEM simulations [9]; Jiang et al. have systematically studied filled rubbers by molecular chain network model and Eshelby's equivalent inclusion theory, respectively [10–12]. Bouchart et al. [13,14] proposed another alternative formula to study compressible hyper-elastic composites. Omnès et al. [15] developed a generalized self-consistent scheme, containing the occluded rubber, the bound rubber and a percolating network, to predict the elasticity of filled rubber. Yin [16] developed a constitutive model for particle reinforced elastomers based on Eshelby's tensor [17]. Although the interaction between particles and matrix is fully considered, some deductions are arguable. Nemat-Nasser [18,19] innovatively extended the Eshelby's tensor and obtained the corresponding formula for large deformation. Huang et al.

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[20] presented a micromechanical analysis for predicting the stress/strain behavior of the composite made of weft-knit polyester fiber interlock fabric and a polyurethane elastomer matrix. Li et al. [21] studied CB filled rubber experimentally and numerically analyzed the local strain filed, concluded that the local strain distribution in a rubber matrix approximately obeys the statistical Gaussian distribution. Molecular chain network models are commonly adopted for filled rubbers. Based on the additive network configurations, Govindjee [23] developed a micromechanics model and analyzed the Mullins' effect and debonding damage in CB filled rubbers. Furthermore, Dargazany [24] proposed a network evolution model to study the deformation-induced anisotropy and damage. The network models based on the statistics thermodynamics could not stand for the real microstructures, and therefore could not to fatherly consider the damage evolution. Quantifying the effective properties is conventionally done by FEM-based numerical simulation. Unfortunately, only two-dimensional (2D) multi-inclusion composite model could be Drozdov [22] emphasized the mechanical energy instead of entropy theory of polymer chains at finite strain deformation, and developed a micromechanics model for polymer and filled polymer as well. simulated by FEM due to the weak convergence in the finite strain mechanism [9,25]. Moore et al. proposed an efficient framework for predicting filled elastomer damping properties based on imaged microstructures [26]. Their proposed multiscale framework shows a significant improvement in computational speed over direct numerical simulations using the FEM. Morozov et al. developed a realistic model of spatial arrangement of fillers in the rubber matrix [27], and fully considered the following structural parameters, such as the distribution of filler sizes, the fractal characteristics of clusters and the presence of large dense particles. Cantournet et al. investigated the effects of carbon nanotubes on the mechanical behavior of elastomeric materials [28], and presented a systematic approach for reducing the experimental data to isolate the MWNT contribution to the strain energy of the composite. A constitutive model for the large strain deformation behavior of MWNT elastomer composites is then developed. In summary, strain localization effect, especially strain locking-up, play a crucial role during the overall deformation of filled rubbers [29]. However, many existing researches often neglect this deformation stage in order to avoid the resulting difficult induced by locking-up deformation [30].

In this paper, a physics-based constitutive model with considering the damage effect will be developed for filled rubbers. For sake of the convenience to practical application, the model is comparatively simple to be readily implemented in a general purpose FEM code for analyzing the macroscopic composite structures. Additionally, the progressive damage effect is involved by adopting DI configuration to represent the microstructure evolution, and FEM simulation is also conducted to analyze the inherent microdeformation mechanisms.

2. Micromechanics-based damage model

2.1. The tangent stiffness of rubber matrix

Rubber matrix is supposed to be isotropic, hyper-elastic, nearly incompressible and described by the following Ogden's strain energy function [31] due to its high accuracy,

$$W = \sum_{i=1}^{n} \frac{2\alpha_i}{\beta_i}\left(\lambda_1^{\beta_i} + \lambda_2^{\beta_i} + \lambda_3^{\beta_i} - 3\right) \tag{1}$$

here, α_i and β_i are unknown coefficients that are determined by least squares fitting to the uniaxial stress–strain curves. λ_i are the material principal stretches which related to the principal strains via $\lambda_i = 1 + \varepsilon_{ii}$. The equivalent strain is given by

$$\varepsilon_e = \sqrt{\frac{2}{3}\varepsilon : \varepsilon} = \sqrt{\frac{2}{3}\left[\varepsilon_{xx}^2 + \varepsilon_{yy}^2 + \varepsilon_{zz}^2 + 2\left(\varepsilon_{xy}^2 + \varepsilon_{yz}^2 + \varepsilon_{zx}^2\right)\right]} \tag{2}$$

Due to the incompressibility condition $\lambda_1\lambda_2\lambda_3 = 1$. The strain components should satisfy the following relation,

$$\varepsilon_{yy} = \varepsilon_{zz} = (1 + \varepsilon_{xx})^{-1/2} - 1 \tag{3}$$

In order to linearly handle the nonlinear behavior, the whole deformation process is firstly divided into n loading step, i.e., the applied stretch is imposed on the specimen step by step up to the final deformation. The tangent modulus at each step is different, and the instantaneous modulus and Poisson's ratio at the n-th step are then defined by,

$$E^{(n)} = \frac{\sigma_{xx}^{(n)} - \sigma_{xx}^{(n-1)}}{\varepsilon_{xx}^{(n)} - \varepsilon_{xx}^{(n-1)}} \tag{4}$$

$$\nu^{(n)} = \frac{\left(1 + \varepsilon_{xx}^{(n-1)}\right)^{-1/2} - \left(1 + \varepsilon_{xx}^{(n)}\right)^{-1/2}}{\varepsilon_{xx}^{(n)} - \varepsilon_{xx}^{(n-1)}} \tag{5}$$

where σ_{xx} and ε_{xx} denotes the nominal stress and strain in the loading direction, respectively. In this paper, all the terms for the reinforcement and matrix are represented by symbols with subscripts ‘P’ and ‘M’, respectively, and those of the composite are denoted by symbols without any script. All the tensors and vectors are written in boldface letters.

2.2. The double-inclusion (DI) model

As for the actual composites, CB nano-powders easily agglomerate to form a network structure in the rubber matrix. These networks surround a part of polymer chains whose deformation is greatly impeded during the applied stretching. Both experimental TEM observation [29] and numerical simulations [21] confirmed that part of rubber matrix among CB powders stretches more seriously than others, and formed a web-like shaped microstructure during the applied deformation. Motivated by these observations, a novel homogenization procedure shown in Fig. 1 is proposed for studying CB filled rubbers, and the intact and cracked double inclusions (DI) are diagramed. CB phase deforms linear elastically due to its high stiffness in relation to the rubber phase. Therefore, CB particles and the surrounded rubber form a new kind of inclusions dispersed in the matrix. What follows is the homogenization procedure. CB particles are equivalent to a homogeneous layer, and its volume fraction just equals to the given CB concentration as a known parameter. The surrounded rubber is assumed to deform linear elastically, and its material properties are given as the already known parameters. The volume fraction of DI is defined by

$$f_{\mathrm{DI}} = f_{\mathrm{P}}\left(\frac{1 + 0.02139DBP_{abs}}{1.46}\right) \tag{6}$$

where DBP_{abs} denote DBP (dibuty1 phthalate) absorption and already measured for different rubbers [32]. A DI configuration is subsequently constructed as shown in Fig. 1, and its equivalent stiffness can be computed by our previous research [33]. In order to realize this purpose, a new configuration including a particle and its surrounding layer is firstly constructed. During the DI construction, the boundaries of particle and its surrounding layer are supposed to be parallel for simplicity.

In the DI model, a centered particle is surrounded by an inhomogeneous layer, in turn embedded in an infinite matrix $\mathbf{C}_{\mathrm{M}}$, and the layer and particle constructs a DI. The DI includes a layer of