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Computational evaluation of strain-rate-dependent deformation behavior of rubber and carbon-black-filled rubber under monotonic and cyclic straining

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Abstract

The molecular chain network model for elastic deformation behavior and the reptation theory for viscoelastic deformation behavior are used to derive a constitutive equation for rubber. The new eight-chain-like model contains eight standard models consisting of Langevin springs and dashpot to account for the interaction of chains with their surroundings. Monotonic and cyclic deformation behavior of rubber with relaxation under different strain rates have been examined. The results reveal the roles of the individual springs and dashpot, and the strain rate dependence of materials in the monotonic and cyclic deformation behaviors, particularly softening and hysteresis loss, that is, the Mullins effect, occurring in stress–stretch curves under cyclic deformation processes. The validity of the results is checked through comparison with experimental results. The deformation behaviors of a plane strain rubber unit cell containing carbon-black (CB) under monotonic and cyclic straining are investigated by computational simulation using the proposed constitutive equation and homogenization method. The results reveal the substantial enhancement of the resistance of CB-filled rubber to macroscopic deformation, which is caused by the marked orientation hardening due to the highly localized deformation of rubber. The role of strain rate sensitivity on such characteristic deformation behaviors as increases in the resistance to deformation, hysteresis loss, and the effects of the distribution morphology and the volume fraction of CB on the deformation behavior is clarified. The increases in the volume fraction and in the aggregation of the distribution of CB substantially raise the resistance to deformation and hysteresis loss.

Keywords: Strain-rate-dependent behavior; Carbon-black-filled rubber; Cyclic deformation behavior; Hysteresis loss; Homogenization method; Molecular chain network theory

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1. Introduction

It is well known that rubber exhibits a complex deformation behavior under monotonic and cyclic straining. Hysteresis loss, that is, the Mullins effect [1] during cyclic loading processes, depends on the strain rate applied, which is strongly related to the ultimate properties of carbon-black (CB)-filled rubber [2]. Blending of CB induces a marked change in such mechanical properties as the resistance to deformation and hysteresis loss. The manifestations of hysteresis and the viscoelastic response of unfilled rubber were explained by the change in the entanglement situation of the molecular structures [3], slipping of the molecular chain [4] and the interaction of springs with the surroundings [5–7]. For the CB-filled rubber, phenomenological constitutive equations [4,6,8,9] have been proposed, however, the details of the mechanism of the manifestation of the characteristic response caused by the microscopic deformation behavior depending on the volume fraction and distribution morphology of CB have not yet been clarified. Therefore, it is indispensable to establish a suitable constitutive equation that can

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reproduce the strain-rate-dependent nature of the rubber and a computational model for evaluating the effect of the CB-filling on the deformation behavior of CB-filled rubber.

Thus far, we have developed a nonaffine model that accounts for the change in the entanglement situation of the molecular network structure [10,11] and constructed a computational model of CB-filled rubber using the proposed constitutive equation and homogenization method [12]. The strain-rate-independent mechanical characteristics of unfilled and CB-filled rubber have been investigated [3], and the mechanism of the enhancement of the mechanical characteristics of the rubber upon filling CB have been clarified. Here, in order to reproduce the strain rate sensitivity of the response of CB-filled rubber, we develop a viscoelastic constitutive equation based on the reptation theory [5,7] for the viscoelastic nature of the rubber and the molecular chain network theory for the nonlinear elastic response. The validity of the proposed constitutive equation will be examined against the experimental data. A unit cell model is constructed to evaluate the effect of the volume fraction and distribution morphology of CB on the deformation behavior of CB-filled rubber under different strain rate conditions. The elaborated 3D model will enable us to clarify the effect of the distribution morphology on the deformation behavior of CB-filled rubber.

2. Constitutive equation

To duplicate the experimentally observed characteristic features of the rubber, the microstructure of rubber is assumed to consist of long molecular chains randomly distributed in space. A single chain, which consists of several segments containing monomers, is defined by two linkages, which are assumed to be chemically or physically entangled points of molecular chains. The physical links are, in general, not permanent and may change depending on deformation. On the other hand, chemical links are permanent and preserve the entanglement situation. The decrease in the number of entangled points upon deformation causes an increase in the average number of segments N in a single chain, enhanced extensibility, and a reduction in the stiffness of the material, i.e., softening, which all play very important roles in the manifestation of the hysteresis of the cyclic deformation behavior of strain-rate-independent rubber [3]. To account for the change in the number of entangled points, a nonaffine molecular chain network theory was developed [11], in which the number of entangled points is expressed as a suitable function of temperature and an appropriately defined measure of deformation. However, in this investigation, we focus our attention on the evaluation of the effect of the viscoelastic nature of the rubber on the characteristic deformation behavior of CB-filled rubber and we restrict our attention to the affine model.

The principal deviatoric stress s_i and principal stretch λ_i relations for the eight-chain model become [13]

$$s_i = \frac{1}{3} C^R \sqrt{N} \frac{\lambda_i^2 - \lambda_c^2}{\lambda_c} L^{-1}\left(\frac{\lambda_c}{\sqrt{N}}\right), \quad L(x) = \coth x - \frac{1}{x},$$
$$\lambda_c^2 = \frac{1}{3}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2), \tag{1}$$

where $C^R = nk_BT$ is a constant, n is the number of chains per unit volume, k_B is Boltzmann's constant, and L is the Langevin function.

The long molecular chains are in contact with other chains at many points, which is the potential source of the viscoelastic response. Therefore, a chain easily moves along the chain direction; however, its movement with respect to its normal direction is restricted. A tube can represent this type of restriction; upon the application of deformation, the molecular chain relaxes with respect to the chain and normal directions in a short time, and subsequently, it moves along the chain direction with long-term relaxation. This is the reptation theory that can easily account for the interaction of chains and their surroundings [5,7]. The viscoelastic response of rubber under the application of stress σ_γ can be represented by

$$d^\gamma = \dot{\gamma}^\gamma \frac{1}{\sqrt{2}\tau^*} \sigma'_\gamma, \quad \tau^* = \left(\frac{1}{2}\sigma'_\gamma \cdot \sigma'_\gamma\right)^{1/2}, \tag{2}$$

where d^γ is the viscoelastic strain rate, σ'_γ is the deviatoric part of σ_γ, and the corresponding shear strain rate $\dot{\gamma}^\gamma$ is a function of stretch λ_γ and stress τ^* [7]:

$$\dot{\gamma}^\gamma = C_1(\lambda_\gamma - 1)^{C_2} \tau^{*m}, \quad \lambda_\gamma = \int_0^t \dot{\lambda}_\gamma \, dt = \int_0^t \lambda_\gamma d^\gamma \, dt. \tag{3}$$

The material parameters C_1 and C_2 are functions of strain rate, because of the characteristic nature of chain relaxation [7].

The most typical characteristic of the present eight-chain model is that it contains eight standard models, as depicted in Fig. 1. This is because the viscoelastic response of rubber is caused by the interaction of chains with their surroundings; therefore, the component chains of the eight-chain model should exhibit a viscoelastic response. The two springs α and β represent single Langevin chains, and dashpot γ represents the viscoelastic nature of rubber, which is modeled using the reptation theory [5,7]. With C_α^R and C_β^R, and N_α and N_β for springs α and β, respectively, as

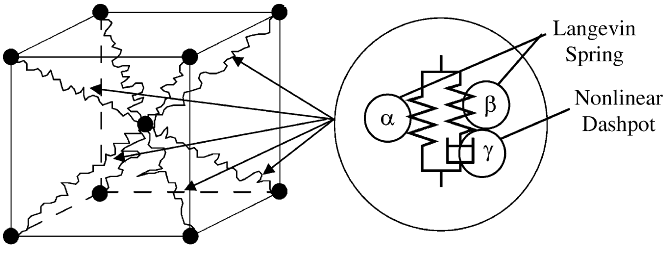

Fig. 1. Eight-chain model containing standard elements. α and β are Langevin springs and γ is dashpot.