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Technical Notes

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Micromechanics of Nonlinear Behavior in Solid-Filled Mooney–Rivlin Rubber Specimen

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Nomenclature

C = fourth-order elasticity tensor, secant elasticity tensor
c_1 = volume fraction of the glass bead
E = secant Young's modulus
p = unknown hydrostatic stress
$S_{(0)}$ = Eshelby tensor associated with spherical inclusions and $C_{(0)}$
$\bar{\varepsilon}$ = total strain of the composite is made of the strain of matrix and glass beads
ε_k = three principal engineering strains
ε^p = related by $S_{(0)}\varepsilon^*$
ε^0 = strain in the comparison sample
$\varepsilon^{(0)}$ = average strain in the matrix of the composite
$\varepsilon^{(1)}$ = average strain of the glass beads
ε^* = equivalent transformation strain
κ = secant bulk modulus
λ_i = principal stretches of deformation associated with the Cauchy–Green deformation tensor
λ_m = nonlinear stretch parameter
μ = secant shear modulus
$\bar{\tau}$ = uniform stress of the composite
τ_k = principal Cauchy stress components
τ_m = nonlinear stress parameter
$\tau^{(0)}$ = average stress in the matrix of the composite
$\tau^{(1)}$ = average stress of the glass beads
v = secant Poisson's ratio

I. Introduction

THE stress analysis of solid-propellant grains requires a constitutive of stress–strain relationship for the solid-propellant material. Linear elastic models have been widely considered as inadequate models of propellant behavior. This article is concerned with the quantitative determination of the nonlinear stress–strain relation of solid-filled hydroxyl-terminated polybutadiene (HTPB) composite, where solid particles are homogeneously dispersed in the rubber matrix. Here we assume that the solid particle and the rubber matrix are perfectly bonded without either voids or nucleation growth. Since the stress–strain relation of the rubber matrix is highly nonlinear, the solid-filled rubber composite system is nonlinear.

As the rubber composite material is under loading, although the matrix already has the nonlinear deformation, the inclusion still can behave as an elastic material. Because of the nonlinear deformation of the matrix, its constraint of the inclusion's deformation will also be weakened. Chen and Chang[1] took the undeformed state of the material as the reference point, using the secant modulus of the total stress–strain relation. Via the secant modulus approach, the elastic modulus now becomes isotropic. Therefore, the computation will become easier. Weng[2–4] adopted the approach of secant modulus of Berveiller and Zaoui[5] to evaluate the plastic deformation of the polycrystals. His result was very close to Hill's[6] micromechanics model of elastoplastic polycrystals.

In this article, we have adopted the mean stress idea of Mori and Tanaka[7] and put it together with the eigenstrain idea of Eshelby[8] to predict the effective elastic moduli of this composite system. At dilute concentration of particles, however, the stress field and strain field in the matrix are uniform, and therefore the mean-field approach will serve as a good approximation. When such a composite is subjected to a constant external stress, the initial response is elastic, and its elastic strain can be determined from its effective moduli.

The stress–strain relation of the solid-filled rubber composite is highly nonlinear. To capture the elastic modulus response of every loading status, we have used the secant moduli in our theoretical derivation. The theory makes use of a linear comparison material, whose elastic moduli at every instant is chosen to coincide with the average secant moduli of the rubber matrix to reflect its nonlinear state. By using Eshelby's[8] equivalent–inclusion principle and Mori and Tanaka's[7] mean-field method, the composite is subsequently replaced by the comparison material filled with equivalent transformation strains. This approach allows us to find the average stress of the matrix in terms of macroscopic stress, and then by appealing to the constitutive equation of the rubber matrix, the overall stress–strain relation of the two-phase system can be easily determined. By introducing λ_m and τ_m, the stress–stretch master curve of the solid-filled rubber composite is obtained. These theoretical results are then checked by experiments.

II. Mooney–Rivlin Rubber Elasticity and Associated Secant Moduli

Let λ_i be the principal stretches of deformation associated with the Cauchy–Green deformation tensor, so that for a Mooney–Rivlin material characterized by two constants a_1 and a_2, the principal Cauchy stress components τ_k are determined by[9]

$$\tau_k = 2[\lambda_k^2 \cdot a_1 - (a_2/\lambda_k^2)] + p, \qquad k = 1, 2, 3 \tag{1}$$

The previous equation is augmented by the constant volume constraint relation

$$\lambda_1\lambda_2\lambda_3 = 1 \tag{2}$$

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Consider the following generalized tensile loading:

$$\tau_3 = P, \qquad \tau_2 = \tau_1 = Q \tag{3}$$

where Q vanishes for a simple tensile loading. It follows from Eqs. (1) and (2) that the associated deformation is

$$\lambda_3 = \Lambda = -\frac{a_2}{4a_1} - \frac{M}{4\sqrt{3}a_1\sqrt{A}} + \frac{\sqrt{|N| + B}}{a_1\sqrt{24\cdot A\cdot M}} \tag{4}$$

where

$$M = (2R^2a_1 - 72a_1^2a_2 + 4Ra_1A + 3a_2^2A + 2a_1A^2)^{1/2}$$

$$A = (-R^3 + 108a_1^3 - 162Ra_1a_2 - 108a_2^3 + L)^{1/3}$$

$$L = 2\sqrt{27}[108(a_1^3 + a_2^3)^2 + (2R^3 + 324a_1a_2R)(a_2^3 - a_1^3) + R^2a_1a_2(207a_1a_2 + 4R^2)]^{1/2}$$

$$B = (M/2)(12Ra_1A + 9a_2^2A - M^2)$$

$$N = \sqrt{27}A^{3/2}(2Ra_1a_2 + a_2^3 - Ba_1^3) \tag{5}$$

$$\lambda_1 = \lambda_2 = \Lambda^{-1/2} \tag{6}$$

$$R = P - Q \tag{7}$$

$$R = [\lambda_3^2 - (1/\lambda_3)][2a_1 + (2a_2/\lambda_3)] \tag{8}$$

It is clear that the desired material constants a_1 and a_2 may be deduced by fitting Eq. (8) to an experimentally measured curve for the simple tension test

$$R = P = \tau_3, \qquad Q = 0 \tag{9}$$

Recalling that the three principal engineering strains ε_k are related to the principal stretches by $\varepsilon_k = \lambda_k - 1$. The generalized tensile loading [Eq. (3)] and the previous calculated strains may be made to satisfy the linear elasticity relations. Both μ and κ modules follow from the usual definitions. We have

$$\kappa = \frac{1}{3}\frac{R + 3Q}{\Lambda + 2\Lambda^{-1/2} - 3} \tag{10}$$

$$\mu = \frac{1}{2}\frac{R}{\Lambda - \Lambda^{-1/2}} \tag{11}$$

III. Initial Stress of the Rubber Matrix

Consider a representative volume of a glass bead-reinforced Mooney–Rivlin rubber that is assumed to be statistically homogeneous and macroscopically isotropic. The Mooney–Rivlin rubber is referred to as the matrix, and the volume fraction of the glass bead is denoted by c_1. Following the exposition of Weng,[4] we introduce an identically shaped comparison sample made of pure Mooney–Rivlin rubber. Let the composite and the pure Mooney–Rivlin rubber comparison sample both be subjected to the boundary traction, which will give rise to a uniform generalized tensile state:

$$\bar{\tau} = (\bar{P}, \bar{Q}), \qquad \bar{R} = \bar{P} - \bar{Q} \tag{12}$$

The strain ε^0, in the comparison sample is related to the previous equation by

$$\bar{\tau} = C_0\varepsilon^0, \qquad C_0 = (3\kappa_0, 2\mu_0) \tag{13}$$

The average strain, $\varepsilon^{(0)}$, $\tau^{(0)}$ in the matrix of the composite cannot be the same as ε^0, $\bar{\tau}$ and may be represented by

$$\tau^{(0)} = \bar{\tau} + \tilde{\tau} = C_0\varepsilon^{(0)} = C_0(\varepsilon^0 + \tilde{\varepsilon}) \tag{14}$$

The introduction of ε^*, combined with Mori and Tanaka's concept, leads to the identity

$$\begin{aligned}\tau^{(1)} &= C_1(\varepsilon^0 + \tilde{\varepsilon} + \varepsilon^p)\\ &= C_0(\varepsilon^0 + \tilde{\varepsilon} + \varepsilon^p - \varepsilon^*)\end{aligned} \tag{15}$$

where ε^p and ε^* are related by

$$\varepsilon^p = S_0\varepsilon \tag{16}$$

in which S_0 is the Eshelby tensor (calculated in the elastic comparison material) associated with the spherical inclusions and C_0.

Finally, since $\bar{\tau}$ of Eq. (12) is also the uniform stress of the composite, we have

$$\bar{\tau} = c_1\tau^{(1)} + (1 - c_1)\tau^{(0)} \tag{17}$$

Following Eqs. (14–17) we have

$$\varepsilon^* = [A^{-1} - (1 - c_1)S_0 - c_1I]^{-1}\cdot\varepsilon^0 \tag{18}$$

$$A^{-1} = I - C_0C_1 \tag{19}$$

The total strain of the composite is given by the weighted average of those strains of the matrix and inclusion, i.e.,

$$\bar{\varepsilon} = (1 - c_1)\varepsilon^{(0)} + c_1\varepsilon^{(1)} = c_1\varepsilon^* + \varepsilon^0 \tag{20}$$

Substituting Eq. (18) into the previous equation and using Eqs. (13) and (15), we finally conclude that

$$\bar{\tau} = C\bar{\varepsilon}, \qquad C = (3\bar{\kappa}, 2\bar{\mu}) \tag{21a}$$

$$\tau^{(0)} = C_0(A - S_0)[A^{-1} - (1 - c_1)S_0 - c_1I]C_0\bar{\tau} \tag{21b}$$

where

$$C = C_0\{I + c_1[A^{-1} - (1 - c_1)S_0 - c_1I]^{-1}\}^{-1} \tag{22}$$

and c is the secant elasticity tensor for the composite at the stress level [Eq. (12)].

For convenience of measurement in the simple tension test, we need to use the relation between the engineering stress T and true stress $\bar{\tau}_{33}$ as

$$T = \bar{\tau}_{33}(1 + \bar{\varepsilon}_{11})(1 + \bar{\varepsilon}_{22}) \tag{23}$$

The final product of this section is the composite stress–strain relation (22). The secant elasticity tensor C, however, depends on $\bar{\tau}$ via $\tau^{(0)}$, the stress of the matrix of the composite. Thus, an iteration procedure must be improvised to facilitate the computation. We use the simpler hard-bead approximation to illustrate the procedure described as follows. Using Eqs. (10) and (11), μ_0 and κ_0 are derived; therefore, C_0 is obtained. Using Eqs. (18), (20), and (22), C and $\bar{\varepsilon}$ are derived, respectively. Finally, by way of Eq. (14), we have $\tau^{(0)}$. Then, we go back to Eqs. (10) and (11) to check C_0. By this iteration procedure, we can have the stress–strain solution of this composite system. The typical numerical solutions of T vs $\bar{\varepsilon}$ curve shown in Fig. 1 are in very good agreement with experimental results.

IV. Stress–Strain Master Curve of a Glass Bead-Reinforced Mooney–Rivlin Rubber

Since the stiffness of glass bead is usually much harder than the HTPB rubber, in the sense that

$$\mu_1 >> \mu_0, \qquad \kappa_1 >> \kappa_0 \tag{24}$$

Using the decomposition theory of the stress, strain, and secant

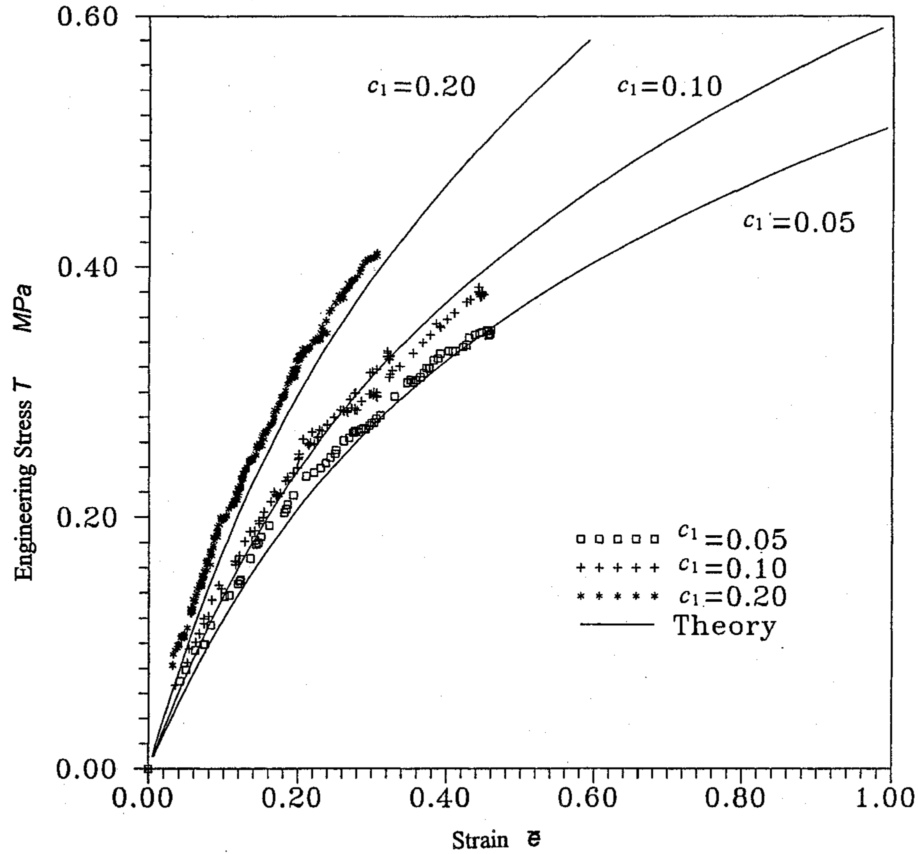

Fig. 1 Engineering stress T vs strain $\bar{\varepsilon}$ for different volume fraction of glass bead-reinforced HTPB rubber composites.

elasticity tensor, they can be decomposed to the deviatoric part and hydrostatic part, individually as follows: by setting μ_0/μ_1 and κ_0/κ_1 to zero in Eqs. (21b) and (22), we obtain

$$\frac{\bar{\kappa}}{\kappa_0} = 1 + \frac{c_1}{(1-c_1)\xi} = \frac{(1-c_1)\xi + c_1}{(1-c_1)\xi} = \frac{1}{(1-c_1)d} \tag{25}$$

$$\frac{\bar{\mu}}{\mu_0} = 1 + \frac{c_1}{(1-c_1)\eta} = \frac{(1-c_1)\eta + c_1}{(1-c_1)\eta} = \frac{1}{(1-c_1)b} \tag{26}$$

$$d = \frac{\xi}{(1-c_1)\xi + c_1}, \qquad b = \frac{\eta}{(1-c_1)\eta + c_1} \tag{27a}$$

$$\eta = \frac{2(4-5v_0)}{15(1-v_0)}, \qquad \zeta = \frac{(1+v_0)}{3(1-v_0)} \tag{27b}$$

$$\tau_{kk}^{(0)} = d\bar{\tau}_{kk} \tag{28}$$

$$\tau'^{(0)} = b\bar{\tau}' \tag{29}$$

Substitution of Eqs. (27a–29) into Eqs. (21a) leads to

$$\bar{\varepsilon}_{kk} = \frac{(1-c_1)\tau_{kk}^{(0)}}{3\kappa_{(0)}} = (1-c_1)\varepsilon_{kk}^{(0)} \tag{30}$$

$$\bar{\varepsilon}' = \frac{(1-c_1)\tau'^{(0)}}{2\mu_{(0)}} = (1-c_1)\varepsilon'^{(0)} \tag{31}$$

Equations (30) and (31) then can be written as tensor form as

$$\bar{\varepsilon} = (1-c_1)\varepsilon^{(0)} \tag{32}$$

If the stiffness of the glass beads is much harder than the matrix then we can treat the glass beads as rigid-body without any deformation, therefore, the total strain $\bar{\varepsilon}$ is just equal to the volume fraction of the rubber matrix $(1-c_1)$ times the strain of the rubber matrix $\varepsilon^{(0)}$. Equation (32) suggests this result. Using Eqs. (28) and (29) the stress distribution of the matrix $\tau^{(0)}$ can be written in its component forms as

$$\tau_{33}^{(0)} = \frac{\tau_{kk}^{(0)}}{3} + \tau_{33}'^{(0)} = \frac{d}{3}(\bar{R} + 3\bar{Q}) + \frac{2b}{3}\bar{R} \tag{33}$$

$$\tau_{22}^{(0)} = \tau_{11}^{(0)} = \frac{\tau_{kk}^{(0)}}{3} + \tau_{11}'^{(0)} = \frac{d}{3}(\bar{R} + 3\bar{Q}) - \frac{b}{3}\bar{R} \tag{34}$$

If we add $(b - d/3)(\bar{R} + 3\bar{Q})$ on both sides of Eqs. (33) and (34), this leads to

$$\tau_{33}^{*} = \tau_{33}^{(0)} + [(b-d)/3](\bar{R} + \bar{Q}) = b\bar{P} \tag{35}$$

$$\tau_{22}^{*} = \tau_{22}^{(0)} + [(b-d)/3](\bar{R} + \bar{Q}) = b\bar{Q} \tag{36}$$

Equations (35) and (36) can be written in tensor form as

$$\tau^{*} = b\bar{\tau} \tag{37}$$

Consider the simple tensile loading $\bar{Q} = 0$, $\bar{R} = \bar{P}$, then similar to Eq. (8) we have the constitutive equation of hardbead-reinforced Mooney–Rivlin rubber composites

$$\tau_m = [\lambda_m^2 - (1/\lambda_m)][2a_1 + (2a_2/\lambda_m)] \tag{38}$$

where

$$\tau_m = b\bar{P} \tag{39}$$

$$\lambda_m = 1 + e_m = 1 + [\bar{\varepsilon}/(1-c_1)] \tag{40}$$

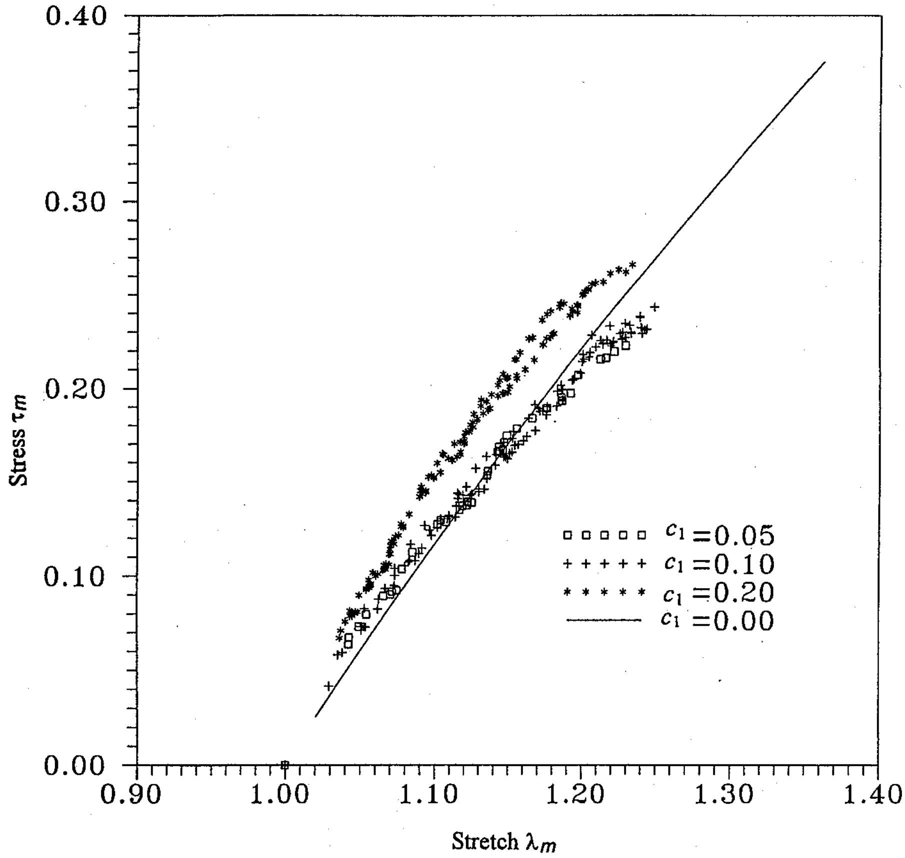

Fig. 2 Generalized stress-stretch master curve of glass bead-reinforced HTPB rubber composites.

For pure Mooney–Rivlin rubber $c_1 = 0$ and $b = 1$, Eq. (38) reduces to the constitutive equation of pure Mooney–Rivlin rubber. Therefore Eq. (38) can be represented as the stress–stretch master curve of different volume fractions of hardbead-reinforced Mooney–Rivlin rubber composites.

V. Experiments

The basic elastomers were prepared from HTPB cured with toluylen-2, 4-diisocyant. The material was cast into sheets of 150 by 150 by 2 mm. The particulate composites were obtained by adding glass spheres 66–88 μm in diameter in quantities necessary to obtain volume fractions of 5, 10, and 20% in the HTPB mixture. Young's modulus of the glass beads is 68,500 MPa. Poisson's ratio of the glass bead is 0.24. The parameters a_1 and a_2 were found to be 0.0518 and 0.157 MPa, respectively, by the aid of Eq. (8), and the method of least squares with the data obtained from simple tensile tests. Tensile specimens of gauge lengths of 20 mm in accordance with ASTM D412 were cut from sheets. All specimens were kept in a desiccator for seven days for postcuring and to stabilize their properties. All of the tensile tests were performed in a laboratory environment of 50% relative humidity and at 25°C.

VI. Results and Discussions

It is now of interest to examine the influence of volume–fraction of inclusions on the stress–strain as predicted by the theory. To this end we assume in our calculations that the inclusions and the matrix take the properties of glass beads and the Mooney–Rivlin rubber, respectively, at room temperature. The elastic constants of both phases are listed in Sec. IV. It showed that the stress–strain curve of the numerical solution is coinciding with the asymptotic solution [Eq. (38)].

By introducing λ_m and τ_m, the stress–stretch relation [Eq. (38) or (39)] of different volume–fraction of inclusions can be regressed to the curve of pure Mooney–Rivlin rubber. The stress–stretch master curve of the glass bead-filled Mooney–Rivlin rubber can be found by the aid of Eq. (39). The results are shown in Fig. 2.

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