Secant Moduli of a Glass Bead-Reinforced Silicone Rubber Specimen

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(Received September 4, 1994)
(Revised June 1, 1995)

ABSTRACT: The influence of volume-fraction of inclusions on the overall stress-strain of a rubber-matrix composite is investigated at the level of dilute concentration. The method developed is based on the approximate mean-field theory developed by Weng and coworkers for plasticity of two-phase composites (Tandon and Weng, 1988; Zhao and Weng, 1989; Qiu and Weng, 1992), using Berveiller-Zaoui's (1979) approximation approach. The constraint due to the matrix phase is characterized by the secant moduli of the matrix, while the interaction of the inclusion is accounted for by the Mori-Tanaka mean-field theory. It is shown that this simple, but approximate theory is capable of predicting the volume fraction dependence of the nonlinear stress-strain relation. An asymptotic solution for this composite system is obtained. By introducing a nonlinear stretch parameter Γ_m and a stress parameter τ_m the stress-stretch master curve of the hardbead-reinforced silicone rubber composite is obtained. The theoretical predictions are in good agreement with the experimental results.

1. INTRODUCTION

THIS PAPER IS concerned with the quantitative determination of the nonlinear stress-strain relation of solid-filled rubber 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 with no voids or nucleation growth. Since the stress-strain relation of the rubber matrix is strongly nonlinear, the solid-filled rubber composite system is nonlinear. With the incom-

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0021-9983/96/1 69-15 $10.00/0

pressible characteristics of the rubber matrix, we only need shear modulus to describe its elastic behavior. However, the solid-filled rubber composite doesn't display the incompressible property as the pure rubber matrix. Most people use the Kerner's experience equation (Kerner, 1956) or modified Kerner equation (Lewis and Nielsen, 1970) to determine the effective shear modulus.

As the composite material is under loading, although the matrix already has the nonlinear deformation, the inclusion still can behave as a linearly elastic material. Due to the plastic deformation of the matrix, its constraint of the inclusion's deformation will also be weakened. This weakened phenomena was discovered by Hill (1965), as he studied the plastic deformation of the polycrystals. He proposed the self-consistent scheme and the relation of tangent moduli for each incremental step to predict the plastic deformation of the polycrystals. Due to the plastic deformation, the tangential moduli keep decreasing and therefore, the constraint of the inclusion's deformation is also weakened as the loading is increasing. The tangent moduli and the Eshelby's transformation tensor are all tangent tensor; therefore, they are anisotropic. In computation, they become cumbersome. Berveiller and Zaoui (1979) 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 (1982) adopted the approach of secant modulus of Berveiller and Zaoui to evaluate the plastic deformation of the polycrystals. His result was very close to Hill's model.

When such a composite is subjected to monotonically increasing, external stress, the initial response is elastic, and its overall elastic strain can be determined from its effective moduli. In Tandon and Weng (1988)–Zhao et al. (1989), an approach based on Mori-Tanaka's method in conjunction with Eshelby's (1957) solution of an ellipsoidal inclusion was adopted with some success.

In this paper, we have adopted the mean stress idea of Mori-Tanaka (1973) and put it together with the eigen-strain idea of Eshelby (1957) 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 reasonably uniform, and therefore the mean-field approach will serve as a good approximation.

The stress-strain relation of the solid-filled rubber composite is highly nonlinear. In order 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 (1957) equivalent-inclusion principle and Mori-Tanaka's (1973) 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 a nonlinear stretch parameter Γ_m and a stress parameter τ_m, the stress-stretch master curve of the solid-filled rubber composite is obtained. The theoretical results are then checked by experiments.

2. NEO-HOOKEAN RUBBER ELASTICITY AND THE ASSOCIATED SECANT MODULI

Let λ_i be the principal stretches of deformation associated with the Cauchy-Green deformation tensor so that for a Neo-Hookean material characterized by a single constant β the principal Cauchy stress components t_k are determined by Truesdell and Noll (1965)

$$\tau_{kk} = \lambda_k^2 - p, \quad k = 1,2,3 \tag{2.1}$$

where $\tau_{kk} \equiv t_k/\beta$ are dimensionless stress components and p an unknown hydrostatic stress. The above equation is augmented by the constant volume constraint relation

$$\lambda_1\lambda_2\lambda_3 = 1 \tag{2.2}$$

Consider now the following generalized tensile loading

$$\frac{t_3}{\beta} = \tau_{33} = P, \quad \frac{t_1}{\beta} = \frac{t_2}{\beta} = \tau_{22} = \tau_{11} = Q \tag{2.3}$$

where Q vanishes for a simple tensile loading. *When loading is known, Equation (2.1) is the third order polynomial equation. There are three roots, two are complex conjugate and the other is positive. Since λ_3 is defined as the stretch deformation, the positive real root therefore must be chosen.* It follows from Equations (2.1) and (2.2) that the associated deformation is

$$\lambda_3 = \Lambda(R) = M(R) + \frac{R}{3M(R)} \tag{2.4}$$

$$\lambda_1 = \lambda_2 = \Lambda^{-1/2} \tag{2.5}$$

$$M(R) = \left\{\frac{1}{2} + \left[\frac{1}{4} - \left(\frac{R}{3}\right)^3\right]^{1/2}\right\}^{1/3} \tag{2.6}$$

$$R = P - Q \tag{2.7}$$

and

$$R = \Lambda^2 - \frac{1}{\Lambda} \tag{2.8}$$

It is clear from the above that the desired material constant β may be deduced by fitting Equation (2.8) to an experimentally measured curve for the simple tension test

$$R = P = \frac{\tau_{11}}{\beta}, \quad Q = 0 \tag{2.9}$$

Recalling that the three principal engineering strains ϵ_{kk} are related to the principal stretches by $\epsilon_{kk} = \lambda_k - 1$, we have

$$\epsilon_{33} = e(R) = \Lambda(R) - 1 \tag{2.10}$$

$$\epsilon_{11} = \epsilon_{22} = e_T(R) = \Lambda^{-1/2}(R) - 1 \tag{2.11}$$

The generalized tensile loading [Equation (2.3)] and the above calculated strains may be made to satisfy the following linear elasticity relations

$$\epsilon_{11} = \frac{\beta}{E^s}[\tau_{11} - \upsilon^s(\tau_{22} + \tau_{33})]$$

$$\epsilon_{22} = \frac{\beta}{E^s}[\tau_{22} - \upsilon^s(\tau_{22} + \tau_{33})] \tag{2.12}$$

$$\epsilon_{33} = \frac{\beta}{E^s}[\tau_{33} - \upsilon^s(\tau_{11} + \tau_{22})]$$

Provided both E^s and υ^s are the so-called secant moduli associated with P and Q, i.e.,

$$\frac{E^s}{\beta} = \frac{R(R + 3Q)}{R[\Lambda - 1] + 2Q[\Lambda - \Lambda^{-1/2}]} \tag{2.13}$$

$$\upsilon^s = \frac{R[1 - \Lambda^{-1/2}] + Q[\Lambda - \Lambda^{-1/2}]}{R[\Lambda - 1] + 2Q[\Lambda - \Lambda^{-1/2}]} \tag{2.14}$$

which are respectively, the secant Young's modulus and secant Poisson's ratio. Secant shear modulus μ^s and secant bulk modulus $\varkappa^s$ follow from the usual definitions. We have

$$\frac{\varkappa^s}{\beta} = \frac{1}{3}\frac{R + 3Q}{\Lambda + 2\Lambda^{-1/2} - 3} \tag{2.15}$$

$$\frac{\mu^s}{\beta} = \frac{1}{2}\frac{R}{\Lambda - \Lambda^{-1/2}} \tag{2.16}$$

For convenience, both indicial and symbolic notations will be used in the text. In the latter case second order tensors will be denoted by boldface, lower case Greek letters and fourth-order tensors by capital letters. Typically, a fourth-order elasticity tensor **C** is used to relate the dimensionless stress τ to the strain ϵ through the representation

$$\beta\tau = \mathbf{C}^s\epsilon, \quad \mathbf{C}^s = (3\kappa^s, 2\mu^s) \tag{2.17}$$

where the last expression signifies the fact

$$C^s_{ijkl} = \mu^s(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{3\kappa^s - 2\mu^s}{3}\delta_{ij}\delta_{kl} \tag{2.18}$$

in which δ_{ij} is the Kronecker delta. In case τ and ϵ are associated with a generalized tensile state in the sense of Equations (2.3), (2.10) and (2.11), the fourth order tensor $\mathbf{C}^s$ is called the secant elasticity tensor. In fact, only generalized tensile states will be considered in this paper. The following notation will be used to characterize Equation (2.3):

$$\tau = (P, Q) \tag{2.19}$$

3. SECANT MODULI OF GLASS BEAD-REINFORCED SILICONE RUBBER

Our object in this section is to establish the basic relation for the average stress of the constituents and the overall strain $\bar{\epsilon}$ and the overall secant moduli C^s of the composite.

The silicone rubber is referred to by the matrix, and the volume fraction of the glass bead is denoted by c_1. Following the exposition of Tandon and Weng (1988) and Weng (1990), we introduce an identically shaped comparison sample made of pure silicone rubber. Let the composite and the pure silicone rubber comparison sample be both subjected to the boundary traction which would give rise to a uniform generalized tensile state

$$\bar{\tau} = (\bar{P}, \bar{Q}), \quad \bar{R} = \bar{P} - \bar{Q} \tag{3.1}$$

The strain ϵ^0 in the comparison sample is related to the above by

$$\beta\bar{\tau} = C^s_0\epsilon^0, \quad C^s_0 = (3\kappa^s_0, 2\mu^s_0) \tag{3.2}$$

where κ^s_0 and μ^s_0 are determined by Equations (2.15) and (2.16) with the substitutions

$$\kappa^s_0 = \kappa^s_0(\bar{R}, \bar{Q}), \quad \mu^s_0 = \mu^s_0(\bar{R}) \tag{3.3}$$

The introduction of the Eshelby's equivalent transformation strain ϵ^*, combined with Mori-Tanaka's mean-field stress concept, the average stress $\tau^{(1)}$ and strain $\epsilon^{(1)}$ of the glass bead are different from the one in the matrix of the composite and can be shown as

$$\epsilon^{(1)} = \epsilon^0 + \tilde{\epsilon} + \epsilon^p \tag{3.4}$$

$$\tau^{(1)} = \tau^0 + \tilde{\tau} + \tau^p \tag{3.4}$$

The above are related by the ordinary elasticity tensor of the beads via

$$\beta\tau^{(1)} = C_1\epsilon^{(1)}, \quad C_1 = (3\varkappa_1, 2\mu_1) \tag{3.6}$$

i.e.,

$$\begin{aligned} \beta\tau^{(1)} &= C_1(\epsilon^0 + \tilde{\epsilon} + \epsilon^p) \\ &= C_0^s(\epsilon^0 + \tilde{\epsilon} + \epsilon^p - \epsilon^*) \end{aligned} \tag{3.7}$$

where ϵ^p and ϵ^* are related by

$$\epsilon^p = S_0^s\epsilon^* \tag{3.8}$$

in which S_0^s is the Eshelby tensor associated with spherical inclusions and C_0^s. After introducing the decomposition, we conclude that

$$\epsilon_{kk}^* = \frac{-\epsilon_{kk}^0}{c_1 + (1 - c_1)\xi_{(0)} + \dfrac{\varkappa_0^s}{\varkappa_1 - \varkappa_0^s}} \tag{3.9}$$

$$\epsilon_{ij}^{*\prime} = \frac{\epsilon_{ij}^{0\prime}}{c_1 + (1 - c_1)\eta_{(0)} + \dfrac{\mu_0^s}{\mu_1 - \mu_0^s}} \tag{3.10}$$

The average stress of the matrix $\tau^{(0)}$ and the average stress of the glass bead $\tau^{(1)}$ can be shown as

$$\tau_{kk}^{(0)} = a_{(0)}\bar{\tau}_{kk}, \quad \tau_{kk}^{(0)\prime} = b_{(0)}\bar{\tau}_{kk}^{\prime} \tag{3.11}$$

$$\tau_{kk}^{(1)} = a_{(1)}\bar{\tau}_{kk}, \quad \tau_{kk}^{(1)\prime} = b_{(1)}\bar{\tau}_{kk}^{\prime} \tag{3.12}$$

where

$$a_{(0)} = \frac{\xi_{(0)} + \dfrac{\varkappa_0^s}{\varkappa_1 - \varkappa_0^s}}{c_1 + (1 - c_1)\xi_{(0)} + \dfrac{\varkappa_0^s}{\varkappa_1 - \varkappa_0^s}} \tag{3.13}$$

$$b_{(0)} = \frac{\eta_{(0)} + \dfrac{\mu_0^s}{\mu_1 - \mu_0^s}}{c_1 + (1 - c_1)\eta_{(0)} + \dfrac{\mu_0^s}{\mu_1 - \mu_0^s}} \tag{3.14}$$

$$a_{(1)} = \frac{1 + \dfrac{\varkappa_0^s}{\varkappa_1 - \varkappa_0^s}}{c_1 + (1 - c_1)\xi_{(0)} + \dfrac{\varkappa_0^s}{\varkappa_1 - \varkappa_0^s}} \tag{3.15}$$

$$b_{(1)} = \frac{1 + \dfrac{\mu_0^s}{\mu_1 - \mu_0^s}}{c_1 + (1 - c_1)\eta_{(0)} + \dfrac{\mu_0^s}{\mu_1 - \mu_0^s}} \tag{3.16}$$

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

$$\bar{\epsilon} = (1 - c_1)\epsilon^{(0)} + c_1\epsilon^{(1)} = c_1\epsilon^* + \epsilon^0 \tag{3.17}$$

Using the decomposing and substituting Equations (3.9) and (3.10) into it, we finally conclude that

$$\beta\bar{\tau} = \bar{C}^s\bar{\epsilon}, \quad \bar{C}^s = (3\bar{\varkappa}_0^s, 2\bar{\mu}_0^s) \tag{3.18}$$

where

$$\frac{\bar{\varkappa}^s}{\varkappa_0^s} = 1 + \frac{c_1}{(1 - c_1)\xi_{(0)} + \dfrac{\varkappa_0^s}{(\varkappa_1 - \varkappa_0^s)}} \tag{3.19}$$

$$\frac{\bar{\mu}^s}{\mu_0^s} = 1 + \frac{c_1}{(1 - c_1)\eta_{(0)} + \dfrac{\mu_0^s}{(\mu_1 - \mu_0^s)}} \tag{3.20}$$

where

$$\eta_{(0)} = \frac{2(4 - 5\upsilon_0^s)}{15(1 - \upsilon_0^s)}, \quad \xi_{(0)} = \frac{1}{3}\frac{(1 + \upsilon_0^s)}{(1 - \upsilon_0^s)} \tag{3.21}$$

and $\bar{C}^s$ is the secant elasticity tensor for the composite at the stress level [Equation (3.1)], $\bar{\varkappa}^s$ and $\bar{\mu}^s$ are the bulk moduli and shear moduli of the composite, respectively. *It is noted that this theoretical model can also lead to identical expres-*

sions for the effective moduli under a prescribed displacement (Tandon and Weng, 1988).

Consider the loading condition as Equation (3.1), then

$$\bar{\tau} = \begin{bmatrix} \bar{Q} & 0 & 0 \\ 0 & \bar{Q} & 0 \\ 0 & 0 & \bar{P} \end{bmatrix} \tag{3.22}$$

and

$$\tau_{kk} = \bar{P} + 2\bar{Q} = \bar{R} + 3\bar{Q} \tag{3.23}$$

$$\bar{\tau}' = \begin{bmatrix} -\dfrac{\bar{R}}{3} & 0 & 0 \\ 0 & -\dfrac{\bar{R}}{3} & 0 \\ 0 & 0 & \dfrac{2\bar{R}}{3} \end{bmatrix} \tag{3.24}$$

Equation (3.18) leads to

$$\bar{\epsilon}_{kk} = \frac{\beta\bar{\tau}_{kk}}{3\bar{\kappa}^s} \tag{3.25}$$

$$\bar{\epsilon}' = \frac{\beta\bar{\tau}'}{2\bar{\mu}^s} \tag{3.26}$$

The overall stress-strain relation of the composite material then can be derived as

$$\bar{\epsilon}_{33} = \bar{e} = \frac{\bar{\epsilon}_{kk}}{3} + \bar{\epsilon}'_{33} = \frac{\beta(\bar{R} + 3\bar{Q})}{9\bar{\kappa}^s} + \frac{\beta\bar{R}}{3\bar{\mu}^s} \tag{3.27}$$

$$\bar{\epsilon}_{11} = \bar{\epsilon}_{22} = \bar{e}_T = \frac{\bar{\epsilon}_{kk}}{3} + \bar{\epsilon}'_{11} = \frac{\beta(\bar{R} + 3\bar{Q})}{9\bar{\kappa}^s} - \frac{\beta\bar{R}}{6\bar{\mu}^s} \tag{3.28}$$

where $\bar{\kappa}^s$ and $\bar{\mu}^s$ are defined as in Equations (3.19) and (3.20), respectively.

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

$$T = \beta\bar{\tau}_{33}(1 + \bar{\epsilon}_{11})(1 + \bar{\epsilon}_{22}) \tag{3.29}$$

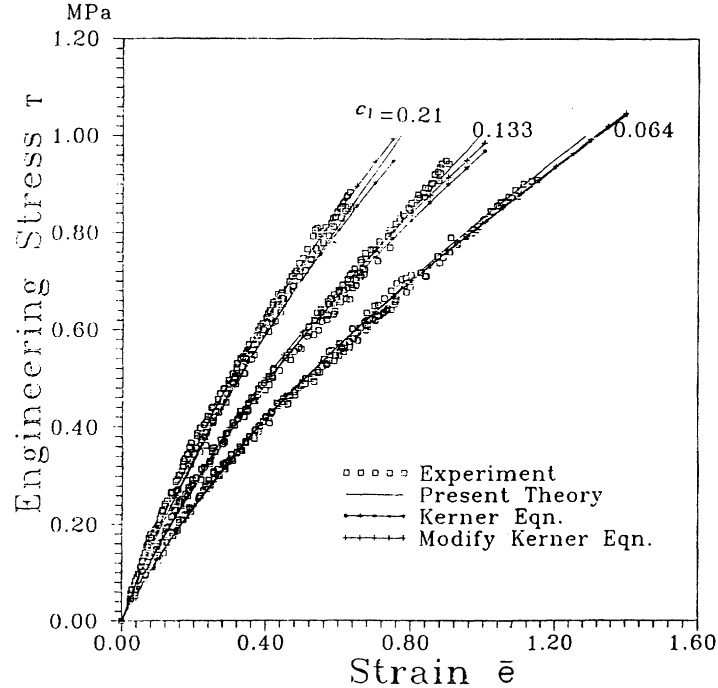

Figure 1. *The engineering stress* T *versus strain* ē *for different volume fractions of glass bead-reinforced silicone rubber composites.*

The final product of this section is the composite stress-strain relation [Equations (3.18) or (3.27) and (3.28)]. The secant elasticity tensor $\overline{C}^s$, 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 Equations (2.13) and (2.14), E_0^s and ν_0^s are derived, therefore, C_0^s is obtained. Using Equations (3.19), (3.20), (3.27) and (3.28), $\overline{C}^s$ and $\bar{\epsilon}$ are derived, respectively. Finally, when we get the stress in the matrix, we go back to Equations (2.13) and (2.14) to check C_0^s. By this iteration procedure we can have the stress-strain solution of this composite system. The typical numerical solutions of T versus $\bar{e}$ curve shown in Figure 1 are in very good agreement with our experimental results and the solution of *Kerner's and Modified Kerner's Equation (1956), (see Appendix).*

4. STRESS-STRAIN MASTER CURVE OF A GLASS BEAD-REINFORCED SILICONE RUBBER

The stiffness of glass bead is usually much harder than the silicone rubber, in the sense that

$$\mu_1 \geq\geq \mu_0^s, \quad \varkappa_1 \geq\geq \varkappa_0^s \tag{4.1}$$

setting μ_0^s/μ_1 and $\varkappa_0^s/\varkappa_1$ to zero in Equations (3.19) and (3.20) we obtain

$$\frac{\bar{\varkappa}^s}{\varkappa_0^s} = 1 + \frac{c_1}{(1 - c_1)\xi_{(0)}} = \frac{(1 - c_1)\xi_{(0)} + c_1}{(1 - c_1)\xi_{(0)}} = \frac{1}{(1 - c_1)d} \tag{4.2}$$

$$\frac{\bar{\mu}^s}{\mu_0^s} = 1 + \frac{c_1}{(1 - c_1)\eta_{(0)}} = \frac{(1 - c_1)\eta_{(0)} + c_1}{(1 - c_1)\eta_{(0)}} = \frac{1}{(1 - c_1)b} \tag{4.3}$$

where

$$d = \frac{\xi}{(1 - c_1)\xi_{(0)} + c_1}, \quad b = \frac{\eta}{(1 - c_1)\eta_{(0)} + c_1} \tag{4.4}$$

Equation (3.11) then can be rewritten as

$$\tau_{kk}^{(0)} = d\bar{\tau}_{kk} \tag{4.5}$$

$$\boldsymbol{\tau}'^{(0)} = b\bar{\boldsymbol{\tau}}' \tag{4.6}$$

Substituting Equations (4.4)–(4.6) into Equations (3.25) and (3.26) leads to

$$\bar{\epsilon}_{kk} = \frac{(1 - c_1)\tau_{kk}^{(0)}}{3\varkappa_0^s} = (1 - c_1)\epsilon_{kk}^{(0)} \tag{4.7}$$

$$\bar{\boldsymbol{\epsilon}}' = \frac{(1 - c_1)\boldsymbol{\tau}'^{(0)}}{2\mu_0^s} = (1 - c_1)\boldsymbol{\epsilon}'^{(0)} \tag{4.8}$$

Equations (4.7) and (4.8) then can be written in tensor form as

$$\bar{\boldsymbol{\epsilon}} = (1 - c_1)\boldsymbol{\epsilon}^{(0)} \tag{4.9}$$

The total strain $\bar{\boldsymbol{\epsilon}}$ of the composite is made of the strain of matrix and glass beads. 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 $\bar{\boldsymbol{\epsilon}}$ is just equal to the volume fraction of the rubber matrix $(1 - c_1)$ times the strain of the rubber matrix $\boldsymbol{\epsilon}^{(0)}$. Equation (4.9) suggests this result.

Using Equations (4.5) and (4.6) the stress distribution of the matrix $\boldsymbol{\tau}^{(0)}$ can be written in its component form as

$$\tau_{33}^{(0)} = \frac{\tau_{kk}^{(0)}}{3} + \tau_{33}'^{(0)} = \frac{d}{3}(\bar{R} + 3\bar{Q}) + \frac{2b}{3}\bar{R} \tag{4.10}$$

$$\tau_{22}^{(0)} = \tau_{11}^{(0)} = \frac{\tau_{kk}^{(0)}}{3} + \tau_{11}'^{(0)} = \frac{d}{3}(\bar{R} + 3\bar{Q}) - \frac{b}{3}\bar{R} \tag{4.11}$$

If we add $[(b - d)/3]\ (\bar{R} + 3\bar{Q})$ on both sides of Equations (4.10) and (4.11) leads to

$$\tau_{33}^{*} = \tau_{33}^{(0)} + \left(\frac{b - d}{3}\right)(\bar{R} + \bar{Q}) = b\bar{P} \tag{4.12}$$

$$\tau_{22}^{*} = \tau_{22}^{(0)} + \left(\frac{b - d}{3}\right)(\bar{R} + \bar{Q}) = b\bar{Q} \tag{4.13}$$

Equations (4.12) and (4.13) can be written in tensor form as

$$\tau^{*} = b\bar{\tau} \tag{4.14}$$

Consider the simple tensile loading, $\bar{Q} = 0$, $\bar{R} = \bar{P}$, then similar to Equation (2.8) we have the constitutive equation of hard bead-reinforced silicone rubber composites

$$\beta\tau_m = \left(\lambda_m^2 - \frac{1}{\lambda_m}\right) \tag{4.15}$$

or

$$\beta\tau_m = \Gamma_m \tag{4.16}$$

where

$$\tau_m = b\bar{P} \tag{4.17}$$

$$\lambda_m = 1 + e_m = 1 + \frac{\bar{\epsilon}}{1 - c_1} \tag{4.18}$$

$$\Gamma_m = \left(\lambda_m^2 - \frac{1}{\lambda_m}\right) \tag{4.19}$$

For pure silicone rubber $c_1 = 0$, and $b = 1$, Equations (4.15) and (4.16) reduce to the constitutive equation of pure silicone rubber. Therefore Equations (4.15) and (4.16) can be represented as the stress-strength master curve of different volume fractions of hard bead-reinforced silicone rubber composites.

5. EXPERIMENTS

The basic elastomers were prepared from silicone rubber cured with the hardener T409103 (from Wacker-Chemie GmbH). The material was cast into sheets of 150 by 150 by 2 mm. The particulate composites were obtained by adding glass spheres 66 to 88 μm in diameter in quantities necessary to obtain volume fractions of 6.4%, 13.3% and 21% in the silicone-mixture. *The Young's modulus of the glass beads is 68500 MPa. The Poisson's ratio of the glass bead is 0.24. The parameter β was found to be 0.4 MPa by the aid of Equation (2.8), and the method of least squares with the data obtained from simple tensile tests.*

Tensile specimens of gage lengths of 20 mm in accordance with ASTM D412 were cut from sheets. All specimens were kept in a desiccator for 15 days for post curing and to stabilize their properties. All the tensile tests were performed in a laboratory environment of 50% relative humidity (RH) and at 25°C.

6. 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 silicone rubber, respectively, at room temperature. The elastic constants of both phases are listed in Section 5. The elastic modulus of glass bead-reinforced silicone rubber can then be found by the aid of Equations (3.27) and (3.28) and the method of least squares with the data obtained from uniaxial tensile tests. Typical results are shown in Figure 2. It showed that the stress-strain

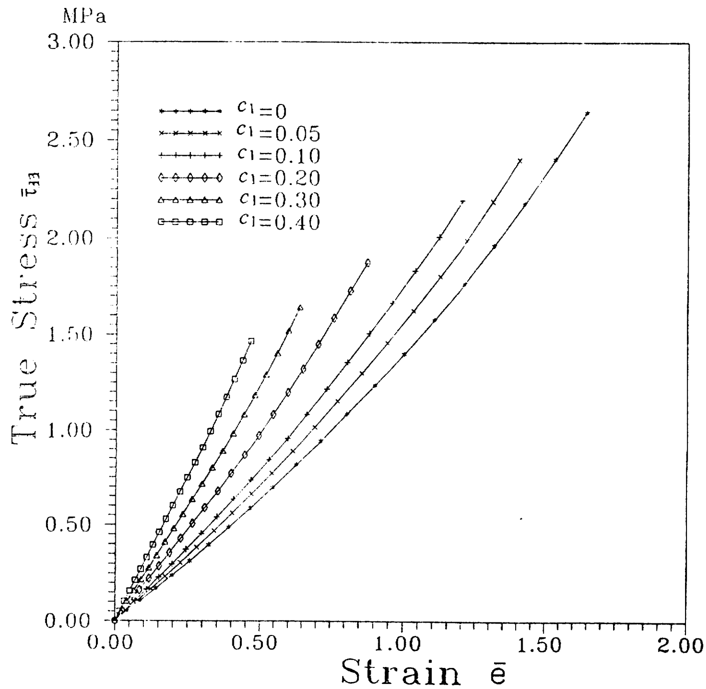

Figure 2. *The Cauchy-stress stress $\bar{\tau}_{33}$ versus strain $\bar{e}$ for different volume fractions of glass bead-reinforced silicone rubber composites.*

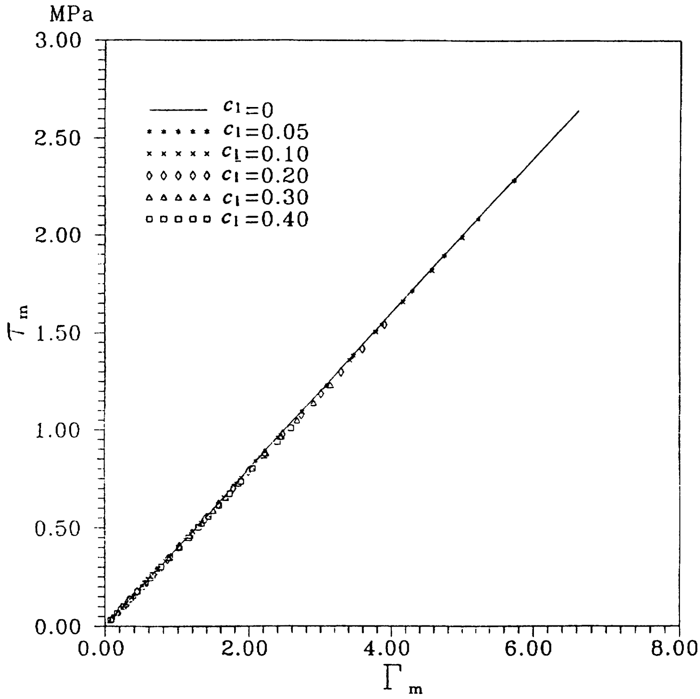

***Figure 3.** The generalized stress-stretch master curve of glass bead-reinforced silicone rubber composites.*

curve of the numerical solution is coinciding with the asymptotic solution [Equation (4.15)].

By introducing a nonlinear stretch parameter Γ_m and a stress parameter τ_m, the stress-stretch relation [Equations (4.15) or (4.16)] of different volume-fraction of inclusions can be regressed to the pure silicone rubber curve. The stress stretch master curve of the glass bead-filled silicone rubber can be found by the aid of Equation (4.16). The results are shown in Figure 3.

7. CONCLUSIONS

The mechanical behavior of solid-filled silicone rubber is found to be nonlinear, depending partly on the content of solid-fillers. To describe such nonlinear characteristics, a nonlinear stretch parameter Γ_m and a stress parameter τ_m, which describe successfully the nonlinear behavior of hard bead-reinforced silicone rubber composites verified by the uniaxial tensile test, have been proposed to the authors' knowledge, for the first time.

APPENDIX

For rubber-filler rigid particles the Kerner equation can be reduced to

$$\frac{\mu_0^s}{\bar{\mu}^s} = 1 + \frac{15(1 - \nu_0^s)}{7 - 5\nu_0^s}\frac{c_1}{c_0} \tag{A1}$$

In this equation, c_0 and c_1 are the volume fraction of the matrix and filler, respectively, and it is assumed that the filler particles are approximately spherical in shape.

Lewis and Nielsen (1970) showed how the Kerner equation can be generalized as

$$\frac{\overline{M}^s}{M_0^s} = \frac{1 + ABc_1}{1 - B\varphi c_1} \tag{A2}$$

where M is any modulus-shear, Young's or bulk. The constant A takes into account such factors of the filler phase and Poisson's ratio of the matrix. The constant B takes into account the relative moduli of the filler and matrix phases. For composites filled with rigid and nearly spherical particles the constants A and B can be assumed as 1.5 and 1.0, respectively. The factor φ depends upon the maximum packing fraction c_m of the filler

$$\varphi = 1 + \left(\frac{1 - c_m}{c_m^2}\right)c_1 \tag{A3}$$

In our case the maximum packing fraction c_m is taken as 0.65.

ACKNOWLEDGEMENTS

The authors wish to express their thanks to Professor Yih-Hsing Pao for many helpful suggestions. This work is supported by the National Science Council of R.O.C., NSC-80-0405-E002-13.

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