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A CONSTITUTIVE EQUATION FOR NONLINEAR SOLIDS WHICH UNDERGO DEFORMATION INDUCED MICROSTRUCTURAL CHANGES

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Abstract—Classical theories of elasticity assume that the Cauchy stress in the material depends on the deformation gradient of particles in the current configuration. Such as assumption can usually be motivated by the presence of a single micromechanism. Here we consider the possibility that as the material is deformed an additional micromechanism might come into play and have a role in determining the Cauchy stress. We show that "inelastic" behavior of some materials can be explained within the context of such a theory. To illustrate our ideas, we use the ideas of scission and reforming of networks within the context of polymeric materials. The theory is of course much more general and can be used to describe the mechanics of materials in which microstructural changes are induced due to deformations.

I. INTRODUCTION

The general form of the constitutive equation for nonlinear elastic **solids** is based on assumptions which imply that stress arises due to a single material micromechanism, and that this micromechanism does not change as the material deforms. In this constitutive equation, the current value of the Cauchy stress depends only on the gradient of the current configuration with respect to the reference configuration, and this dependence is expressed in terms of a Helmholtz free energy density function. Such a constitutive assumption can be motivated by a single molecular mechanism. For example, if one considers the kinetic theory of rubber elasticity, the Helmholtz free energy is derived from the molecular theories of rubber networks, which consider the influence of such factors as configurational changes in macromolecules, cross-links, and entanglements.

In this paper, we consider materials which undergo microstructural changes when deformed. That is, a new micromechanism arises which affects the mechanical response of these materials and leads to new physical phenomena such as permanent set upon release of stress. Such phenomena are usually associated with materials that are classified as **plastic**. Ideas in plasticity theory owe their genesis to the behavior of metallic solids. However, theories of **plastic response** need to be restudied due to the advent of modern polymeric solids. Many of the phenomena generally classified under the umbrella of **plastic behavior** in polymeric solids can be interpreted, as we shall see, within the context of a different theoretical framework. It is not our aim to press claim to our interpretation as being the unique correct explanation. However, it seems under certain circumstances, for a class of materials, such a theoretical framework is both natural and appealing.

Our research develops the seminal ideas of TOBOLSKY and ANDREWS [1945] and TOBOLSKY, ANDREWS, and HANSON [1946] who proposed the two-network theory for

polymeric materials. In this theory, a certain number of cross-links are present in the initial stress free state, and additional cross-links are introduced in a later state. The initial system of cross-links produces one network or micromechanism. The appearance of the new cross-links leads to a new micromechanism and a second network. TOBOLSKY, PRETTYMAN, and DILLON [1944] postulated another situation in which molecular cross-links are broken and then reformed to produce a new network with a new reference state. LODGE [1964] used a two-network theory to discuss permanent set in rubbers caused by uniaxial and biaxial extension deformation. FONG and ZAPAS [1976] used both the two-network theory and the molecular network model of TOBOLSKY, PRETTYMAN, and DILLON to discuss chemical stress relaxation and permanent set in rubber. A third example of material response in which a second micromechanism occurs could be that of strain induced crystallization. PETERLIN (1976) suggested that this occurs when a number of macromolecular chain segments associate to form a bundle like cluster with fairly good orientation. The remaining chain segments may be in an amorphous region. Thus, a new constituent or micromechanism is formed which contributes to the mechanical response. It is easy to see that all the above-mentioned phenomena can lead to "**plastic behavior**".

WINEMAN and RAJAGOPAL [1990] discussed a constitutive theory based on such material response. They assumed that a microstructural change occurs at some stage of deformation which results in the conversion of a portion of the original material to a new network with a new reference configuration. They then used this two-network model to solve a number of problems including permanent set due to shearing and combined extension and torsion of circular cylinders. In this work, we extend the constitutive equation in order to allow for continuous conversion of the original material as deformation proceeds. The general model is outlined in the next section. A specific example is presented in Section III. Finally, we point out that the ideas presented here are related to those in an earlier work (cf. RAJAGOPAL & WINEMAN [1980]), in which we discussed a method for modeling changes from one manner of response to another based on the ideas of bifurcation and the selection of an appropriate branch.

II. CONSTITUTIVE MODEL

Let s denote a parameter associated with the deformed state. Let $\kappa(0)$ denote the configuration of the material in its initial unstrained, unstressed state, and let $\kappa(s)$ denote its configuration corresponding to the deformed state s. The coordinates of a material configuration $\kappa(s)$ are denoted by $\mathbf{x}(s)$. The deformation gradient of particles in the configuration $\kappa(s)$ with respect to particles in the configuration $\kappa(\hat{s})$ is denoted by $\mathbf{F}(s,\hat{s})$.

We shall present the underlying ideas for a nonlinear elastic material which is incompressible and isotropic in its initial configuration. It is assumed that there is a regime of deformations from the initial configuration in which the mechanical response is governed by a single micromechanism. Within this regime of deformations the constitutive equation has the form

$$\sigma = -p\mathbf{I} + \phi_1\mathbf{B}(s,0) + \phi_{-1}[\mathbf{B}(s,0)]^{-1} \tag{1}$$

where $\mathbf{B}(s,0) = \mathbf{F}(s,0)\mathbf{F}(s,0)^T$, $\phi_1 = \partial W/\partial I_1$, $\phi_{-1} = \partial W/\partial I_2$, I_1 and I_2 denote the principal invariants of $\mathbf{B}$, and $W(I_1, I_2)$ is the Helmholtz free energy density function.

At a certain stage of deformation, a new micromechanism is activated which leads to a change in the microstructure of the material. We shall characterize this event by introducing an activation function (activation criterion) $A(\mathbf{F}(s,0))$ which depends on the deformation gradient of the current configuration with respect to the initial configuration. In our present work we assume that the activation function is objective and scalar valued. However, this need not be so, and it is possible that other theories could be developed where the activation function could be tensor valued. Our activation criterion is akin to the yield criterion in classical plasticity theory. We illustrate some examples using a specific activation criterion. While our criterion is able to model some of the phenomena observed in polymeric solids, it is by no means the only such criterion which can give the desired results. It is our aim to inject a new thought process, and thus we are not very concerned with the precise structure of the activation criterion. As more experimental and theoretical results are obtained a more rational process for choosing the activation function might emerge.

The new micromechanism is activated when

$$A(\mathbf{F}(s,0)) = 0. \tag{2}$$

Material frame indifference, isotropy, and incompressibility imply that

$$A = A(I_1, I_2). \tag{3}$$

The role played by the activation criterion theory is best understood within the context of the specific examples presented later. For example, the activation criterion discussed below determines the value of the state parameter at which a new micromechanism is brought into play.

For convenience of presentation, we focus on a specific micromechanism, that of scission or breaking of network junctions, and their subsequent reforming into new networks. The formation of a second network by means of this micromechanism was introduced by TOBOLSKY and ANDREWS [1945]. FONG and ZAPAS [1976] then allowed for a continuous process of breaking and reforming of network junctions and discussed a possible constitutive equation for the resulting material. The constitutive model introduced here is a modification and generalization of that of FONG and ZAPAS [1976].

It is assumed that during each increment in the deformation, a certain fraction of network junctions of the original material are broken. This fraction depends on the extent of deformation of the original material. The newly broken network junctions then immediately reform to produce a new undistorted network. During further deformation, this newly formed network deforms and contributes to the total stress. It is assumed, for the sake of simplicity, that there is no scission of newly formed networks.

The total stress at each stage of deformation is defined to be the superposition of contributions from the remaining portion of the original material and from each network for formed during the deformation process. Thus, if $\boldsymbol{\sigma}(s)$ is the total stress at the deformation state corresponding to state parameter s,

$$\boldsymbol{\sigma}(s) = \boldsymbol{\sigma}^R(\mathbf{s}) + \int_{s_a}^{s} a(\hat{s})\boldsymbol{\sigma}^N(s,\hat{s})\, d\hat{s} \tag{4}$$

where $\boldsymbol{\sigma}^R(s)$ is the stress in the remaining original material, s_a is the value of the state parameter when activation begins, $a(\hat{s})d\hat{s}$ is the volume fraction of network formed during the interval of deformation when the state parameter increases from $\hat{s}$ to $\hat{s} + d\hat{s}$ and $\boldsymbol{\sigma}^N(s, \hat{s})$ is the stress per unit volume in the newly formed material. The latter depends on the deformation gradient $\mathbf{F}(s, \hat{s})$, which compares the configuration of the network at the current state to the configuration when it was formed at state $\hat{s}$.

Let $b(s)$ denote the volume fraction of the remaining portion of the original material at state s. Then $\boldsymbol{\sigma}^R(s)$ is assumed to have the form

$$\boldsymbol{\sigma}^R(s) = -p\mathbf{I} + b(s)\,[\phi_1\mathbf{B}(s,0) + \phi_{-1}\mathbf{B}(s,0)^{-1}]. \tag{5}$$

It is assumed that the rate of breaking of network junctions equals the rate at which new junctions are formed. This assumption is not essential to the analysis that follows and can be relaxed. This implies that

$$b(s) = 1 - \int_{s_a}^{s} a(\hat{s})\,d\hat{s}. \tag{6}$$

Each new network is assumed to respond like an incompressible nonlinear isotropic elastic material. Moreover, again for the sake of simplicity, the mechanical response is the same for all of the new networks. Thus, we shall assume

$$\boldsymbol{\sigma}^N(s, \hat{s}) = -p\mathbf{I} + \bar{\phi}_1\mathbf{B}(s, \hat{s}) + \bar{\phi}_{-1}\mathbf{B}(s, \hat{s})^{-1} \tag{7}$$

where $\mathbf{B}(s, \hat{s}) = \mathbf{F}(s, \hat{s})\mathbf{F}(s, \hat{s})^T$, and $\bar{\phi}_1, \bar{\phi}_{-1}$ are material property functions which depend on the invariants of $\mathbf{B}(s, \hat{s})$. On combining (4), (5), and (7), the constitutive model during activation takes the representation

$$\begin{aligned}\boldsymbol{\sigma} = &-p\mathbf{I} + b(s)\,[\phi_1\mathbf{B}(s,0) + \phi_{-1}\mathbf{B}(s,0)^{-1}] \\ &+ \int_{s_a}^{s} a(\hat{s})\,[\bar{\phi}_1\mathbf{B}(s, \hat{s}) + \bar{\phi}_{-1}\mathbf{B}(s, \hat{s})^{-1}]\,d\hat{s}.\end{aligned} \tag{8}$$[1]

It is further assumed that at each state of deformation there is a regime for which no further scission occurs and within this regime and $a(\hat{s}) = 0$, and the upper limit of the integral in (8) is fixed at say $s = s^*$. Moreover, (6) shows that $b(s)$ is fixed at the value $b(s^*)$. Equation (8) then becomes

$$\begin{aligned}\boldsymbol{\sigma} = &-p\mathbf{I} + b(s^*)\,[\phi_1\mathbf{B}(s,0) + \phi_{-1}\mathbf{B}(s,0)^{-1}] \\ &+ \int_{s_a}^{s^*} a(\hat{s})\,[\bar{\phi}_1\mathbf{B}(s, \hat{s}) + \bar{\phi}_{-1}\mathbf{B}(s, \hat{s})^{-1}]\,d\hat{s}.\end{aligned} \tag{9}$$

This constitutive expression can be expressed in terms of $\mathbf{F}(s,0)$ and thus can be interpreted as representing local elastic response. In order to show this, note that

$$\mathbf{F}(s, \hat{s}) = \mathbf{F}(s,0)\mathbf{F}(\hat{s},0)^{-1} \tag{10}$$

[1] It should be noted that p in the two eqs (7) and (8) represents different functions. Though this might appear confusing, we choose to use such a notation for the sake of brevity.

from which it follows that

$$\begin{aligned} \mathbf{B}(s,\hat{s}) &= \mathbf{F}(s,0)\mathbf{C}(\hat{s})^{-1}\mathbf{F}(s,0)^T, \\ \mathbf{C}(\hat{s}) &= \mathbf{F}(\hat{s},0)^T\mathbf{F}(\hat{s},0). \end{aligned} \tag{11}$$

When these are substituted into (9), the latter can be written in the form

$$\begin{aligned} \boldsymbol{\sigma} = -p\mathbf{I} &+ b(s^*)\left[\phi_1\mathbf{B}(s,0) + \phi_{-1}\mathbf{B}(s,0)^{-1}\right] \\ &+ \mathbf{F}(s,0)\mathbf{A}_1\mathbf{F}(s,0)^T + \mathbf{F}(s,0)^{-T}\mathbf{A}_2\mathbf{F}(s,0)^{-1} \end{aligned} \tag{12}$$

where

$$\mathbf{A}_1 = \int_{s_a}^{s^*} a(\hat{s})\bar{\phi}_1\mathbf{C}^{-1}(\hat{s})\,d\hat{s} \tag{13}$$

and

$$\mathbf{A}_2 = \int_{s_a}^{s^*} a(\hat{s})\bar{\phi}_{-1}\mathbf{C}(\hat{s})\,d\hat{s}. \tag{14}$$

Each of the several assumptions incorporated into (8) can be generalized. This would lead to an exceedingly complicated model and will be introduced in a future presentation.

III. EXAMPLE

We now provide a specific example of the general constitutive model presented in Section II. Let the material response before activation be neo-Hookean. Then (1) has the form

$$\boldsymbol{\sigma}(s) = -p\mathbf{I} + \mu\mathbf{B}(s,0) \tag{15}$$

where μ is a constant. The deformation state parameter is defined to be the radius in $I_1 - I_2$ space through

$$s = [(I_1 - 3)^2 + (I_2 - 3)^2]^{1/2}. \tag{16}$$

Thus, when the material is in its initial state, $I_1 = I_2 = 3$ and $s = 0$. The activation condition is defined to be

$$A(I_1, I_2) = [(I_1 - 3)^2 + (I_2 - 3)^2]^{1/2} - s_a = 0. \tag{17}$$

When activation begins, each newly formed network is assumed to act as a neo-Hookean elastic material. Then (8) reduces to

$$\begin{aligned} \boldsymbol{\sigma}(s) = -p\mathbf{I} &+ b(s)\mu\mathbf{B}(s,0) \\ &+ \int_{s_a}^{s} \mu a(\hat{s})\mathbf{B}(s,\hat{s})\,d\hat{s}. \end{aligned} \tag{18}$$

For the purposes of this example, $a(\hat{s})$ is chosen to be quadratic on a finite domain,

$$\begin{aligned} a(s) &= \alpha(s - s_a)(s - s_c), \quad s \in (s_a, s_c) \\ &= 0 \quad s \in (s_c, \infty) \end{aligned} \tag{19}$$

where s_c denotes the value of the state parameter at the completion of conversion. It should be noted that, for the specific form of $a(s)$ chosen s_c is bounded. However for other choices it could be unbounded. Let the total volume fraction of converted material be denoted by C, where $C \leq 1$. Then, recalling (6)

$$C = \int_{s_a}^{s_c} a(\hat{s})\, d\hat{s}. \tag{20}$$

It then follows from (19) and (20) that

$$\alpha = \frac{6C}{s_a^2(s_a - 3s_c) + s_c^2(3s_a - s_c)}. \tag{21}$$

According to (17), there are many different deformation states at which activation can begin. Suppose activation has been initiated and the material has been deformed to a state at which $s = s^*$. According to (17) and (18), the material can be deformed to many other states for which $s = s^*$ without further scission of network junctions. If the material is deformed further so that $s < s^*$ then the deformation of the original material can be regarded as having decreased. Then, if the material is deformed so that $s \leq s^*$, there is no further conversion of material and the stress is given by

$$\begin{aligned} \boldsymbol{\sigma} = &-p\mathbf{I} + b(s^*)\mu\mathbf{B}(s,0) \\ &+ \int_{s_a}^{s^*} \mu a(\hat{s})\mathbf{B}(s,\hat{s})\, d\hat{s}. \end{aligned} \tag{22}$$

We now illustrate the predictions of the model for two homogeneous deformations, uniaxial and unequal biaxial extension.

III.1. *Uniaxial extension*

Let the material be subjected to uniaxial extension along the x_1-axis of a cartesian coordinate system. The deformation gradient is $\mathbf{F} = \mathrm{DIAG}(\lambda, 1/\sqrt{\lambda}, 1/\sqrt{\lambda})$ where λ is the stretch ratio. The invariants of the tensor $\mathbf{B}$ are

$$I_1 = \lambda^2 + \frac{2}{\lambda}, \qquad I_2 = 2\lambda + \frac{1}{\lambda^2} \tag{23}$$

and the deformation state parameter, by (16), is

$$s(\lambda) = \left[\left(\lambda^2 + \frac{2}{\lambda} - 3\right)^2 + \left(2\lambda + \frac{1}{\lambda^2} - 3\right)^2\right]^{1/2} \tag{24}$$

which can be shown to be a monotonically increasing function of λ. This can be inverted to give $\lambda = \lambda(s)$. Before activation, when $s < s_a$

$$\sigma_{11} = \mu\left(\lambda^2 - \frac{1}{\lambda}\right), \qquad \sigma_{22} = \sigma_{33} = 0. \tag{25}$$

During network conversion, $s > s_a$, and the stress is given by (18)–(21).

Note that the deformation gradient at state s for the network formed at state $\hat{s}$ is given by

$$\begin{aligned} \mathbf{F}(s,\hat{s}) &= \mathbf{F}(s,0)\mathbf{F}(\hat{s},0)^{-1} \\ &= \mathrm{DIAG}\left[\frac{\lambda(s)}{\lambda(\hat{s})}, \left(\frac{\lambda(\hat{s})}{\lambda(s)}\right)^{1/2}, \left(\frac{\lambda(\hat{s})}{\lambda(s)}\right)^{1/2}\right]. \end{aligned} \tag{26}$$

Thus, when $\lambda = \lambda(s)$ is increasing, $\sigma_{22} = \sigma_{33} = 0$ and

$$\sigma_{11} = b(s)\left(\lambda^2 - \frac{1}{\lambda}\right) + \int_{s_a}^{s} a(\hat{s})\left[\frac{\lambda^2}{\lambda(\hat{s})^2} - \frac{\lambda(\hat{s})}{\lambda}\right] d\hat{s}. \tag{27}$$

A plot of σ_{11} versus λ is shown in Fig. 1. The solid line shows the response for a neo-Hookean material. The other lines show the relation for various amounts of conversion. Conversion is assumed to begin when $s_a = 10$, corresponding to $\lambda = 3.4$, and terminate when $s_c = 30$, corresponding to $\lambda \approx 5.5$. These points are indicated by heavy dots. Just after initial conversion, the $\sigma_{11} - \lambda$ graph becomes less steep than in the neo-Hookean case ($C = 0$), which is usually described as softening of the material. There is then an increase in slope, or a stiffening of the material, just before the end of the conversion process. For conversions of 30% and 40%, the graphs are monotonic. At 50%, the graph has a local maximum followed by a local minimum. These features correspond to a loss of stiffness of the original material due to network scission, followed by an increase in stiffness as the newly formed networks become deformed. For the graphs corresponding to 30% and 40% conversion, we conclude that the rate of loss of stress carrying capacity of the original material is small compared to the rate of increase of stress carrying capacity as new networks are being formed and stretched. At 50% conversion, the loss of the stress carrying capacity occurs faster than the new networks are being formed and stressed. At $s = s_c$ no further conversion occurs, and the deformation in all networks increases, leading to further increase in the stress. It should be pointed out that if $a(s_a) \neq 0$ (or $a(s_c) \neq 0$), there would be a discontinuity in the slope of the σ_{11} versus λ graph at $s = s_a$ (or $s = s_c$). This is avoided in the present example by the particular choice of $a(\hat{s})$ in (19).

Suppose that λ increases to a maximum value at $s = s^*$, and then λ varies so that $s \leq s^*$. According to (22), the relation between σ_{11} and λ is given by

$$\begin{aligned} \sigma_{11} &= b(s^*)\left(\lambda^2 - \frac{1}{\lambda}\right) + \int_{s_a}^{s^*} a(\hat{s})\left[\frac{\lambda^2}{\lambda(\hat{s})^2} - \frac{\lambda(\hat{s})}{\lambda}\right] d\hat{s} \\ &= b(s^*)\left(\lambda^2 - \frac{1}{\lambda}\right) + \lambda^2 \int_{s_a}^{s^*} \frac{a(\hat{s})}{\lambda(\hat{s})^2}\, d\hat{s} - \frac{1}{\lambda}\int_{s_a}^{s^*} a(\hat{s})\lambda(\hat{s})\, d\hat{s}. \end{aligned} \tag{28}$$

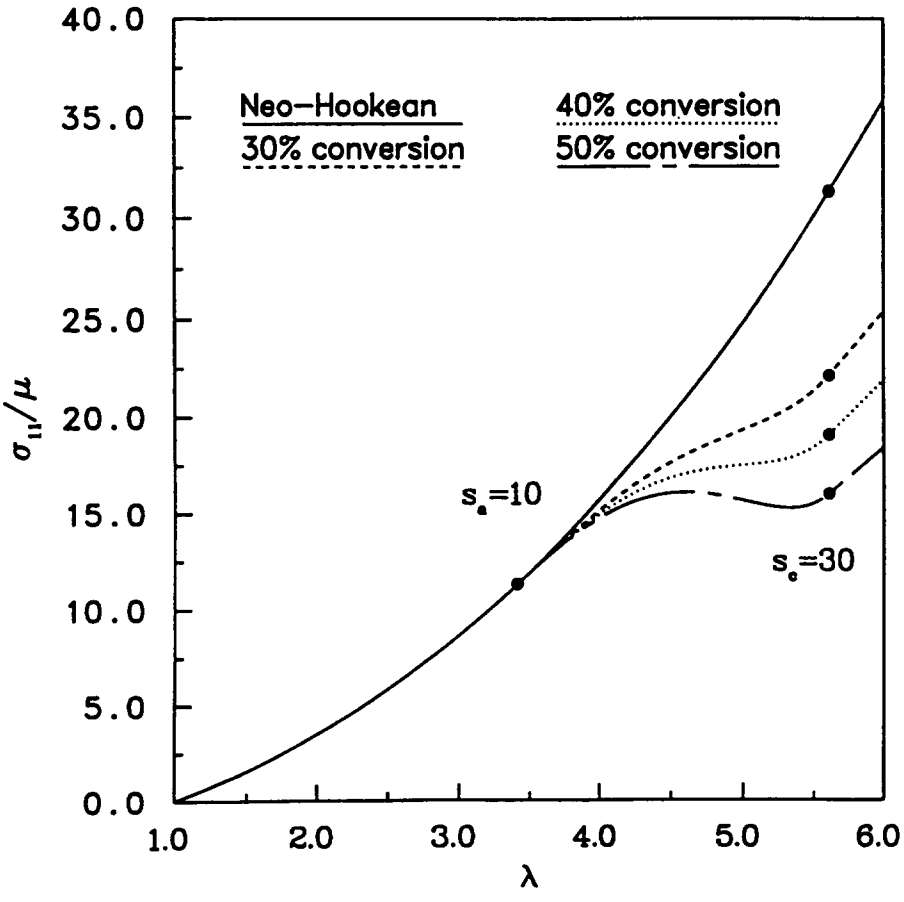

Fig. 1. Uniaxial stress-stretch graphs during elongation, for various amounts of conversion.

Figure 2 shows the σ_{11} versus λ graph corresponding to this equation as λ increases to 5, and then λ decreases (unloads). When $\sigma_{11} = 0$ there is permanent set. In this state, the total stress equals zero, but the original network is in a state of tension and the newly formed networks are in states of compression.

III.2. *Unequal bi-axial extension*

Consider a homogeneous deformation in which the material is subjected to unequal stretches along the x_1 and x_2 axes of a cartesian coordinate system and $\sigma_{33} = 0$. The deformation gradient is $\mathbf{F} = \mathrm{DIAG}(\lambda_1, \lambda_2, (\lambda_1\lambda_2)^{-1})$, and the strain invariants are

$$I_1 = \lambda_1^2 + \lambda_2^2 + (\lambda_1\lambda_2)^{-2}$$
$$I_2 = \frac{1}{\lambda_1^2} + \frac{1}{\lambda_2^2} + (\lambda_1\lambda_2)^{-2}. \tag{29}$$

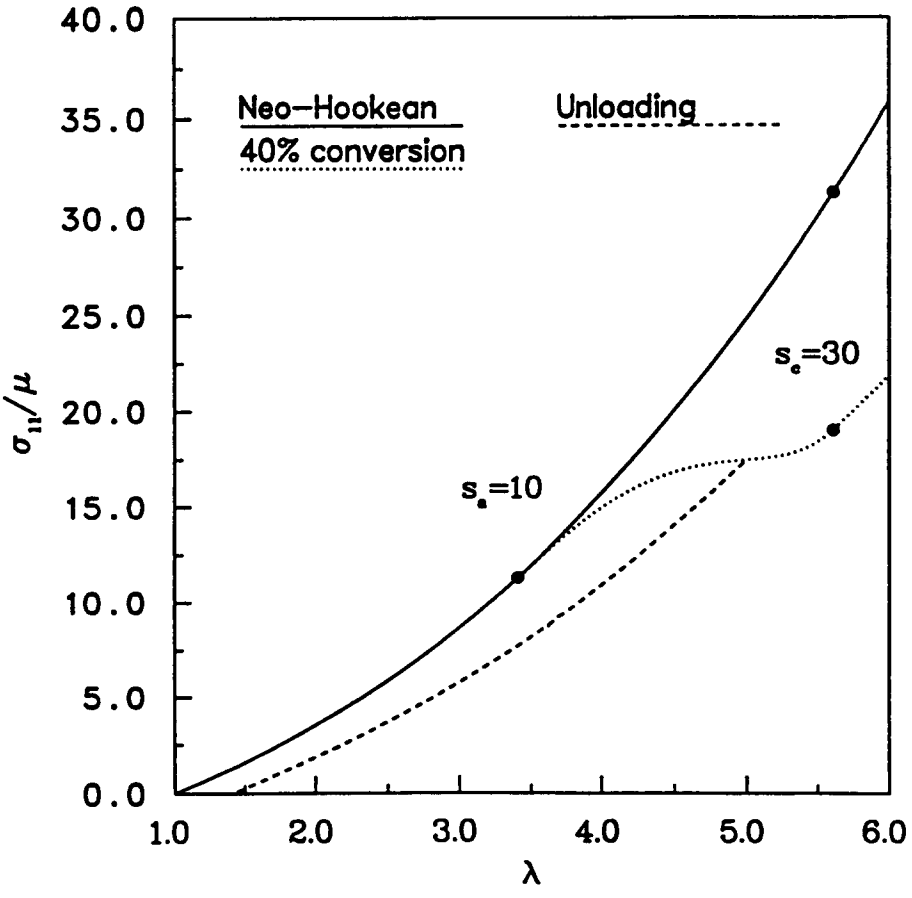

Fig. 2. Uniaxial stress-stretch graphs showing the effect of reversing the stretching when $\lambda = 5$. Results are shown for 0% and 40% conversion.

It is seen from (16) that $s = s(\lambda_1, \lambda_2)$. When $s < s_a$

$$\sigma_{\alpha\alpha} = \mu(\lambda_\alpha^2 - \lambda_3^2), \qquad \alpha = 1,2 \tag{30}$$

where $\lambda_3 = (\lambda_1\lambda_2)^{-1}$.

Now consider an unequal bi-axial stretch history for which

$$\lambda_2 - 1 = \frac{2}{5}(\lambda_1 - 1). \tag{31}$$

Then $s(\lambda_1, \lambda_2) = \bar{s}(\lambda_1)$. This relation can be shown to increase monotonically with λ_1, so that there is an inverse relation $\lambda_1 = \lambda_1(s)$. When $s > s_a$, the constitutive relation is given by (18). The deformation gradient $\mathbf{F}(s, \hat{s})$ is

$$\mathbf{F}(s, \hat{s}) = \mathrm{DIAG}\left(\frac{\lambda_1(s)}{\lambda_1(\hat{s})}, \frac{\lambda_2(s)}{\lambda_2(\hat{s})}, \frac{\lambda_3(s)}{\lambda_3(\hat{s})}\right) \tag{32}$$

and the stress is given by

$$\sigma_{\alpha\alpha} = b(s)\mu(\lambda_\alpha^2 - \lambda_3^2) + \int_{s_a}^{s} a(\hat{s})\mu\left(\frac{\lambda_\alpha^2}{\lambda_\alpha^2(\hat{s})} - \frac{\lambda_3^2}{\lambda_3^2(\hat{s})}\right) d\hat{s}. \tag{33}$$

The predictions of this case are shown in Fig. 3, in which $s_a = 10$, $s_c = 80$, and $C = 0.5$. The upper pair of lines corresponds to σ_{11} and the lower pair corresponds to σ_{22}. The upper line of each pair shows neo-Hookean response in which there is no conversion and the lower line shows the effect of conversion. Notice that both σ_{11} and σ_{22} decrease due to conversion, and both appear to have local maxima.

IV. CONCLUSION

The intent of this work was to show that inelastic behavior in materials can be explained within the context of deformation induced microstructural changes in an initially

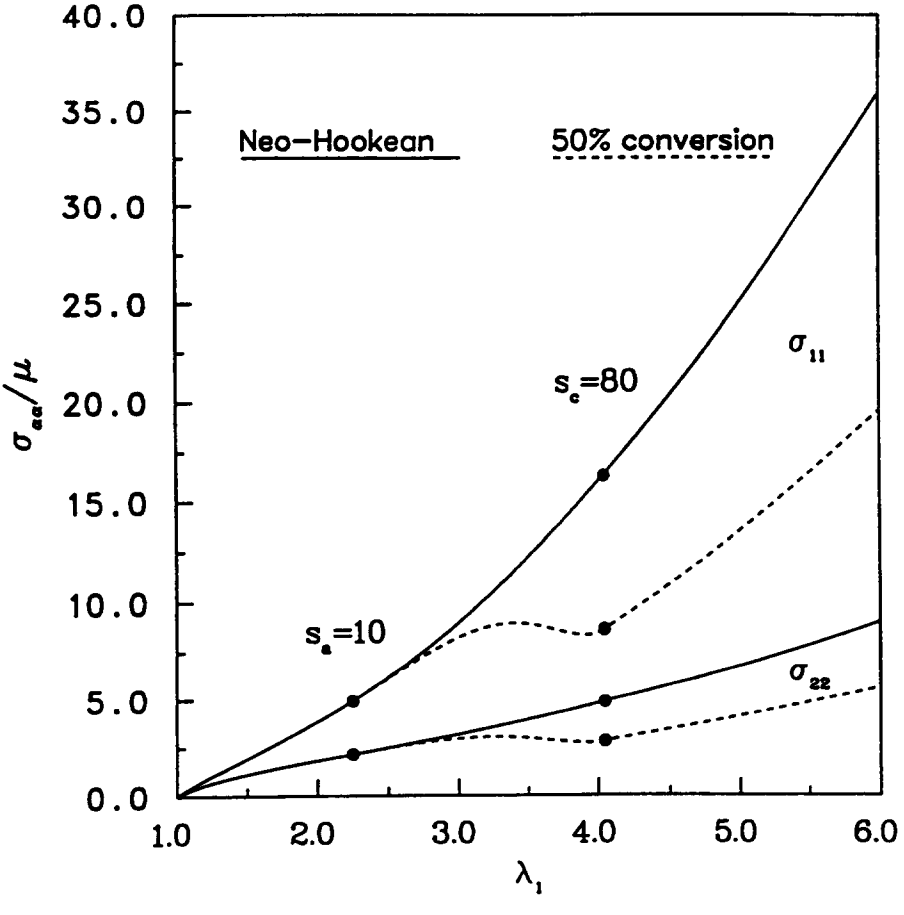

Fig. 3. Stress-stretch graphs during unequal bi-axial stretching according to eq. (31). Results are shown for 0% and 50% conversion.

elastic solid wherein the newly induced micromechanism also leads to an elastic response. In any further deformation, both the micromechanisms come into play thereby determining the response of the solid body. We have shown that phenomena like permanent set on the release of stresses, behavior that is usually associated with inelastic materials, are possible within this theoretical framework. The creation of the second micromechanism is given by an activation criterion similar to a yield condition in plasticity, but is different in the sense that it is fixed once and forever and there is no evolution of a yield surface. The conversion from one micromechanism to another could be either discretized or continuous. Here, we have studied the problems of uni-axial and bi-axial extension of an initially isotropic elastic solid, within the context of a specific activation criterion, allowing for continuous conversion. The results clearly indicate inelastic phenomena of permanent set and hysterisis.

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