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A phenomenological energy-based model to characterize stress-softening effect in elastomers

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Abstract

A phenomenological energy-based model for stress-softening of isotropic, incompressible hyperelastic rubberlike materials is derived here. In this model, the microstructural damage is characterized by an exponential softening function that depends on the current magnitude of the strain–energy function and its maximum previous value in a deformation of the virgin material. Theoretical models are presented for uniaxial, equibiaxial and pure shear deformations by using Gaussian and non-Gaussian material molecular network models. The accuracy of the resulting constitutive equations is demonstrated on uniaxial, equibiaxial and pure shear experimental data provided in the literature. Comparisons between the energy-based model and the strain intensity based phenomenological model described in [Elías-Zúñiga A, Beatty MF. ZAMP 2002;53:794–814. [1]] show that the model developed here is slightly superior in following experimental data.

Keywords: Stress-softening effect; Non-Gaussian molecular network models; Strain energy density

1. Introduction

Rubberlike materials are known to exhibit a highly nonlinear elastic behavior under static load, a viscoelastic behavior including hysteresis under cyclic loading, and a stress-softening phenomenon known as the Mullins effect. Since, the modelling of the physical behavior of these materials is complex, we only considered here static behavior and focus on the characterization of Mullins effect of rubberlike materials under several deformation states. An excellent overview of both experimental and theoretical aspects of Mullins effect is provided in Refs. [2–6]. There are also many studies concerning time-independent constitutive equations to characterize aspects of phenomenological descriptions of Mullins phenomenon based on strain–energy functions. Some representative works are papers written by De Souza Neto, Perić and Owen [7], Ogden and Roxburgh [6], Holzapfel, Stadler and Ogden [8], Dorfmann and Ogden [9], and Horgan et al. [10]. These publications focus on phenomenological strain–energy functions that depend on a damage parameter associated with the maximum energy on the primary loading path rather than the specific deformation state that governs the unloading response.

The aim of this paper is to introduce a new phenomenological network model for the Mullins effect based on strain–energy functions of isotropic, incompressible hyperelastic, time independent rubberlike materials. This theoretical damage model is developed in order to provide a description of an idealized form of the Mullins effect for various deformation states. It is important to point out that this phenomenological model closely parallels the strain-based model developed by Elías-Zúñiga and Beatty [1] in which the determination of only two material constants for a neo-Hookean material model: the shear modulus μ_0, and the softening rate parameter b, and three constants for non-Gaussian molecular network models: μ_0, b, and the model specific molecular chain number of links N are required.

The paper is organized as follows. We begin in Section 2 with a brief review of the relations for finite deformations of an incompressible elastic material. In Section 3, we present a brief description of the Ogden and Roxburgh energy-based phenomenological approach to model Mullins effect [6]. In

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Section 4, we present a new constitutive model for stress-softening for which the damage function depends on the magnitude of the energy at a material point; and we introduce in Section 5 a specific damage function that is used in a great variety of deformation states. We then apply the Gaussian, neo-Hookean and non-Gaussian, James–Guth [12] and Arruda–Boyce [13] molecular network models as well as the phenomenological full-network composite model to derive in Section 6 corresponding virgin material and stress-softened material constitutive equations. Results demonstrating the effects of stress-softening for these molecular network models in uniaxial extension, pure shear and equibiaxial deformation are compared in Section 7 with experimental data by Mullins and Tobin [3], Muhr [15], Cheng [16], Johnson and Beatty [17] and by Chagnon [18]. It is shown for these data that our phenomenological energy-based stress-softening material model predicts with great accuracy the Mullins effect in rubberlike materials. In Section 8, we briefly review the strain-based phenomenological model developed by Elías-Zúñiga and Beatty [1] for stress-softening in elastomers. Finally, we compare in Section 9 our proposed energy-based model with the strain-based phenomenological model by applying the corresponding virgin material and stress-softened material constitutive equations with data by Cheng [16], Johnson and Beatty [17] and by Chagnon [18]. It is shown for these data, that our energy-based model is slightly superior to the strain-based model in following experimental data.

2. Preliminaries

We recall briefly some essential relations for finite deformations of an incompressible elastic material. Let us consider a material particle at the place $\mathbf{X}=X_k\mathbf{e}_k$ in an initially undeformed reference configuration of a body. When subjected to a prescribed deformation, the particle at X moves to the place $\mathbf{x}=\mathbf{x}_k\mathbf{e}_k$ in the current configuration of the body in a common rectangular Cartesian frame $\phi=\{O;\mathbf{e}_k\}$ with origin O and orthonormal basis $\mathbf{e}_k$. An isochoric deformation is described by

$$x_1=\lambda_1 X_1,\quad x_2=\lambda_2 X_2,\quad x_3=\lambda_3 X_3 \tag{1}$$

in which λ_i denote the principal stretches in ϕ. The Cauchy–Green deformation tensor $\mathbf{B}\equiv\mathbf{F}\mathbf{F}^T$ has the form

$$\mathbf{B}=\lambda_1^2\mathbf{e}_{11}+\lambda_2^2\mathbf{e}_{22}+\lambda_3^2\mathbf{e}_{33} \tag{2}$$

where $\mathbf{e}_{jk}\equiv\mathbf{e}_j\otimes\mathbf{e}_k$, $\mathbf{e}_i$ are the associated orthonormal principal directions, and $\mathbf{F}$ is the usual deformation gradient. In the undistorted state $\mathbf{F}=1$, and $m=\sqrt{3}$; otherwise, $m>\sqrt{3}$ for all isochoric deformations [5]. The magnitude of the strain at a material point $\mathbf{X}$, also called the strain intensity and denoted by m, is defined by $m=\sqrt{\mathbf{B}\cdot\mathbf{B}}=\sqrt{\mathrm{tr}\mathbf{B}^2}$, where tr denotes the trace operation. The principal invariants I_k of $\mathbf{B}$ are defined by

$$I_1=\mathrm{tr}\,\mathbf{B},\quad I_2=\frac{1}{2}\left[I_1^2-\mathrm{tr}(\mathbf{B}^2)\right],\quad I_3=\det\mathbf{B} \tag{3}$$

so the magnitude m of $\mathbf{B}$ as a function of the invariants is given by

$$m=\sqrt{I_1^2-2I_2} \tag{4}$$

We note that $m\geq\sqrt{3}$ for all λ, equality holding when and only when $\lambda=1$, the undeformed state. Also, for all deformations of an incompressible material we have $I_3\equiv 1$.

3. Review of Ogden–Roxburgh pseudo-elastic model

Ogden and Roxburgh [6] proposed a phenomenological pseudo-elastic model to characterize Mullins effect in filled rubber. They assumed that on the first loading path its stress–strain behavior may be described as a function of the principal stretches by

$$T_i=\lambda_i\frac{\partial\bar{W}}{\partial\lambda_i}(\lambda_1,\lambda_2,\lambda_3,\eta)-p,\quad i=1,2,3,\ \text{no sum} \tag{5}$$

in which the strain–energy function W is related to the pseudo-energy function $\bar{W}$ via

$$\bar{W}(\lambda_1,\lambda_2,\eta)=\eta W(\lambda_1,\lambda_2)+\phi(\eta) \tag{6}$$

where η is an additional scalar continuous damage parameter that takes the value unity on the virgin path, $\phi(\eta)$ is a smooth damage function with $\phi(1)=0$, and $W(\lambda_1,\lambda_2)$ is the strain–energy function that characterizes any primary loading path. On subsequent loading cycles, η satisfies $0<\eta\leq 1$ and attains its unit value only if $W(\lambda_1,\lambda_2)\geq W_{\max}(\lambda_{1m},\lambda_{2m})$, where $W_{\max}$, λ_{1m} and λ_{2m} correspond to the previous maximum energy state. Ogden and Roxburgh have shown that the damage function satisfies the relation

$$-\phi'(\eta)=W(\lambda_1,\lambda_2) \tag{7}$$

which defines η in terms of the deformation state as well as on the specific forms of $W(\lambda_1,\lambda_2)$ and $\phi(\eta)$ used. Also, Ogden and Roxburgh [6] in their pseudo-elasticity model defined the dissipation function $\phi(\eta)$ to have the form:

$$-\phi'(\eta)=m_1\mathrm{erf}^{-1}(r(\eta-1))+W_{\max} \tag{8}$$

where m_1 and r are positive material constant parameters and $\mathrm{erf}^{-1}(\,)$ is the inverse of the error function. They applied it to characterize Mullins effect in simple uniaxial extension data obtained from Mullins and Tobin (1957) with great accuracy. Recently, Dorfmann and Ogden [9] selected a different damage function given by:

$$-\phi'(\eta)=m_1\tanh^{-1}(r(\eta-1))+W_{\max} \tag{9}$$

and they used it to characterize loading, partial unloading and reloading of a particle-reinforced rubber with hysteretic response [9]. We now turn to the development of a new

phenomenological energy-based model to describe stress-softening effect in rubberlike materials.

4. A new phenomenological energy-based model for stress-softening

To characterize the virgin material response of an incompressible and isotropic elastic material, we consider a time independent constitutive equation of the form

$$\mathbf{T}_0 = -p1 + \aleph_1(I_1, I_2)\mathbf{B} + \aleph_{-1}(I_1, I_2)\mathbf{B}^{-1} \tag{10}$$

where $\mathbf{T}_0$ is the Cauchy stress, p is an undetermined pressure, and $\aleph_\Gamma = \aleph_\Gamma(I_1, I_2)$, $\Gamma = 1, -1$, denote the virgin material response functions that can be determined by a strain–energy function $W = \hat{W}(I_1, I_2)$, per unit reference volume, in accordance with

$$\aleph_1 = 2W_1, \quad \aleph_{-1} = -2W_2 \tag{11}$$

wherein $W_\alpha \equiv \partial\hat{W}/\partial I_\alpha$ [5]. During loading of the virgin material, the maximum previous strain–energy is its current value, $W_{\max} = W$. In addition and for many standard forms of strain–energy functions, W is a monotonic increasing function of the amount of stretch from the undeformed state.

The phenomenological model proposed here is based on the assumption that microstructural material damage is characterized by a certain isotropic and monotonic increasing function $\Psi(W;W_{\max})$ that depends on the material strain–energy density W and satisfies the conditions

$$0 < \Psi(W; W_{\max}) < 1, \quad \Psi(W_{\max}; W_{\max}) = 1 \tag{12}$$

where $W_{\max}$ represents the maximum previous strain energy density at the point at which the material is unloaded from the virgin path. The softening function $\Psi(W;W_{\max})$ is determined by a constitutive equation that describes the evolution of microstructural change that begins immediately upon deformation from the natural, undistorted state of the virgin material. We assume that $\Psi(W;W_{\max})$ is a positive monotonic increasing function of the strain intensity on the interval $m \in [\sqrt{3}, M]$ and that it depends on the different forms of strain–energy functions. Thus, our proposed model can be considered as a pseudo-elastic model [6].

If we let $m_{\max} = M$ be the amount of stretch at the point at which the material is unloaded and fix the maximum previous strain energy at $W = W_{\max}$ then, the stress-softened material response for subsequent unloading and loading again from an undeformed state, or from any other elastic point for which $W < W_{\max}$ is defined by the time-independent constitutive equation

$$\boldsymbol{\tau} = \Psi(W; W_{\max})\mathbf{T}_0 \tag{13}$$

where $\boldsymbol{\tau}$ denotes the Cauchy stress in the stress-softened material. It is evident by substituting (6) in (5) that our softening function $\Psi(W;W_{\max})$ is identical to Ogden–Roxburgh damage parameter η defined in Section 3, i.e.

$$\Psi(W; W_{\max}) = \eta \tag{14}$$

and hence, we may use Ogden–Roxburgh pseudo-elastic theory to determine this function. We shall return to this later.

Notice from Eq. (13) that $\boldsymbol{\tau} = \mathbf{T}_0 = 0$, when and only when $m = \sqrt{3}$. Also, the virgin and stress-softened material response values coincide at each softening point for which $W = W_{\max}$ and $m_{\max} = M$. The material behavior described by (13) for $W \le W_{\max}$ is ideally elastic for both loading and unloading as long as the value of W does not exceed its maximum previous value $W_{\max}$. Thereafter, the material recalls its virgin material response described by (10).

In accordance with (12) and (13), the ratios of the non-trivial physical stress components T_{0ij} in the virgin material to the corresponding non-trivial physical components τ_{ij} in the stress-softened material, for a given deformation state, are determined by the inverse of the softening function alone

$$\frac{T_{0ij}}{\tau_{ij}} = \frac{1}{\Psi(W; W_{\max})} \ge 1, \quad i,j = 1,2,3, \text{ no sum} \tag{15}$$

the equality holding when and only when $W = W_{\max}$. In sum, for a given isochoric deformation $\mathbf{B}$, the magnitude of the stress component in the stress-softened material is smaller than the magnitude of the non-trivial corresponding stress component in the virgin material.

We shall see in Section 5 how to obtain a damage function $\phi(\eta)$ that depends on only one positive parameter called the material softening parameter.

5. A stress-softening material model

In order to determine the softening function $\Psi(W;W_{\max})$, we use the Ogden–Roxburgh [6] pseudo-elastic model and select a damage function $\phi(\eta)$ of the form:

$$-\phi'(\eta) = -1\left(\frac{1}{b}\ln\eta\right)^{1/n} + W_{\max} \tag{16}$$

where b is a positive material constant, named the softening parameter and n is a specified positive constant chosen to best fit data for a given rubberlike material. Henceforward, we consider the value of $n = 1/2$. On substitution of Eq. (7) into Eq. (16) and after a little algebraic manipulation, we obtain the expression for the scalar continuous damage parameter η:

$$\eta \equiv \Psi(W; W_{\max}) = \mathrm{e}^{-b\sqrt{(W_{\max} - W)}} \tag{17}$$

Since, $W(\lambda_1,\lambda_2) \le W_{\max}$, it follows that $\eta \le 1$ with equality holding when and only when $W(\lambda_1,\lambda_2) = W_{\max}$.

Substitution of Eq. (11) into Eq. (10) using the general class (13) and recalling Eq. (14), we thus obtain the following simplified Mullins material model:

$$\boldsymbol{\tau} = \mathbf{T}_0 \mathrm{e}^{-b\sqrt{(\bar{W}_{\max} - \bar{W})}} \tag{18}$$

in which each member of the family of elastic stress-softened materials (18), characterized by the previous maximum strain energy density $W_{\max}$, is generated from a specified virgin material model in the class (10). Note that during primary loading of the virgin material, the maximum previous strain energy density is its current value and hence Eq. (18) yields $\boldsymbol{\tau} = \mathbf{T}_0$, the virgin material response. When the virgin material is unloaded from a state of maximum previous strain for which $W(\lambda_1,\lambda_2)$ is either fixed at its maximum previous value, $W(\lambda_1,\lambda_2) = W_{\max}$, or it is decreased the stress-softened material response follows (18). In fact, as long as $W(\lambda_1,\lambda_2) < W_{\max}$ the material response is ideally elastic for decreasing as well as for increasing strain. When the material is further loaded on subsequent second deformation of the stress-softening material from its maximum previous strain at the softening point, the response is again described by the virgin constitutive Eq. (10).

Integration of Eq. (16) with respect to η yields the explicit expression for the damage function $\phi(\eta)$:

$$\begin{aligned}\phi(\eta) &= \int_1^{\eta} \left(\left(\frac{\ln \eta}{b} \right)^2 - W_{\max} \right) \mathrm{d}\eta \\ &= \frac{2}{b^2} \left[\eta \left(1 - \ln \eta + \frac{1}{2} \{\ln \eta\}^2 \right) - 1 \right] \\ &\quad + (1 - \eta) W_{\max} \end{aligned} \tag{19}$$

On substitution of Eq. (17) into Eq. (19), we obtain the dependence of ϕ on $W_{\max}$:

$$\begin{aligned}\phi = \frac{\mathrm{e}^{-b\sqrt{(W_{\max} - W)}}}{b^2} \Big(& 2 - b^2 W + 2b\sqrt{(W_{\max} - W)} \\ & + \mathrm{e}^{b\sqrt{(W_{\max} - W)}} (b^2 W_{\max} - 2) \Big) \end{aligned} \tag{20}$$

We may conclude from Eq. (20) that the damage function ϕ depends on the point, where the virgin material is unloaded from a state of maximum previous strain $W_{\max}$.

6. Material models

To illustrate the application of the stress-softening material model given by Eq. (18), we consider the neo-Hookean Gaussian virgin material and the classical James–Guth 3-chain, Arruda–Boyce 8-chain, and the full network composite non-Gaussian virgin material network models[1]. We shall begin with the neo-Hookean virgin material model.

6.1. A neo-Hookean material

A neo-Hookean material is described by a Gaussian strain–energy function of the form

$$W = \frac{\mu_0}{2}(I_1 - 3) \tag{21}$$

in which μ_0 denotes the shear modulus in the undeformed, natural state of the virgin material. The corresponding maximum strain energy at the greatest previous stretch prior to unloading is given by

$$W_{\max} = \frac{\mu_0}{2}(I_{1\max} - 3) \tag{22}$$

where

$$I_{1\max} = \lambda_{1\max}^2 + \lambda_{2\max}^2 + \lambda_{3\max}^2 \tag{23}$$

and $(\lambda_{1\max},\lambda_{2\max},\lambda_{3\max})$ are the values of $(\lambda_1,\lambda_2,\lambda_3)$ at a point at which unloading begins.

Using Eqs. (10) and (11), the virgin material constitutive equation becomes

$$\mathbf{T}_0 = -p\mathbf{1} + \mu_0 \mathbf{B} \tag{24}$$

In view of (18) and (24), the stress–stretch relations for a stress-softening neo-Hookean material are given by:

$$\boldsymbol{\tau} = (-p\mathbf{1} + \mu_0 \mathbf{B}) \mathrm{e}^{-b\sqrt{W_{\max} - W}} \tag{25}$$

Now consider a simple uniaxial extension for which the Cauchy stress in the virgin material is given by $T_{011} = T$, all other components $T_{0jk} = 0$. The equilibrium equations are satisfied for constant p. The corresponding uniaxial stretch is denoted by $\lambda_1 = \lambda$, and the incompressibility condition $\lambda_1\lambda_2\lambda_3 = 1$ requires that $\lambda_2 = \lambda_3 = 1/\lambda$. Then our constitutive Eqs. (24) and (25) yield the following uniaxial stress–stretch relation for a stress-softened neo-Hookean material:

$$\tau = \mu_0 \left(\lambda^2 - \frac{1}{\lambda} \right) \mathrm{e}^{-b\sqrt{W_{\max} - W}} \tag{26}$$

The corresponding uniaxial engineering stress is denoted $\sigma_s = \tau/\lambda$. Hence, by (26),

$$\sigma_s = \mu_0 \left(\lambda - \frac{1}{\lambda^2} \right) \mathrm{e}^{-b\sqrt{W_{\max} - W}} \tag{27}$$

A similar relation may be obtained for an equibiaxial homogeneous deformation for which $\lambda_1 = \lambda_2 = \lambda$, and $\lambda_3 = \lambda^{-2}$. In this case, the equibiaxial engineering stress-softened neo-Hookean material is described by

$$\sigma_s = \mu_0 \left(\lambda - \frac{1}{\lambda^5} \right) \mathrm{e}^{-b\sqrt{W_{\max} - W}} \tag{28}$$

A pure shear or a plane strain compression is a homogeneous deformation for which $\lambda_1 = \lambda$, $\lambda_2 = 1$, and $\lambda_3 = \lambda^{-1}$. Here, the pure shear engineering stress-softened

[1] Note that in all non-Gaussian material network models reviewed here, the inverse Langevin function can be approximated by $\beta = 3\lambda_r/(1-\lambda_r^3)$, an empirical estimate that exhibits very good comparison with the exact numerical values of $\beta = \mathcal{L}^{-1}(\lambda_r)$ [11].

neo-Hookean material model is described by

$$\sigma_s = \mu_0\left(\lambda - \frac{1}{\lambda^3}\right)\mathrm{e}^{-b\sqrt{W_{\max}-W}} \tag{29}$$

6.2. The James–Guth virgin material model

The strain energy $W = \hat{W}(\lambda_1, \lambda_2, \lambda_3)$ per unit volume for the three-chain, James–Guth virgin material model [12] may be written as

$$W_{3\mathrm{ch}} = \frac{nk\Theta}{3}N_3\sum_{j=1}^{3}\left(\beta_j\lambda_{jr} + \ln\left(\frac{\beta_j}{\sinh\beta_j}\right)\right) - c_3 \tag{30}$$

where k is the Boltzmann constant, n denotes the chain density per unit volume, Θ is the absolute temperature, N_3 is the number of rigid links each of length l in a molecular chain between cross-links of a molecular network consisting of three identical, orthogonal chains that have the same initial root-mean square (rms) chain vector length $r_0 = \sqrt{N_3}l$. The constant c_3 is chosen so that the strain energy vanishes in the natural, undeformed state; and $\beta_j \equiv \mathcal{L}^{-1}(\lambda_{jr})$ is the inverse of the Langevin function $\mathcal{L}(\beta_j) \equiv \coth\beta_j - (1/\beta_j)$. The current chain vector length in an affine deformation is defined by $r_{j\mathrm{chain}} \equiv \lambda_j r_0$, where λ_j represents the macroscopic principal stretch of the continuum along the jth principal axis. The corresponding jth relative chain stretch λ_{jr} is defined by

$$\lambda_{jr} \equiv \frac{\lambda_{j\mathrm{chain}}}{\lambda_L} = \frac{\lambda_j}{\sqrt{N_3}}, \quad j = 1, 2, 3, (\text{no sum}) \tag{31}$$

in which $\lambda_{j\mathrm{chain}} \equiv r_{j\mathrm{chain}}/r_0 = \lambda_j$ defines the jth current chain stretch in the affine deformation, and $\lambda_L \equiv r_L/r_0$, with $r_L \equiv N_3 l$, is the locking chain stretch.

If $(\lambda_{1\max},\lambda_{2\max},\lambda_{3\max})$ are the values of $(\lambda_1,\lambda_2,\lambda_3)$ at which unloading begins, then the maximum strain energy at the greatest previous stretch is given by:

$$W_{3\mathrm{chmax}} = \frac{nk\Theta}{3}N_3\sum_{j=1}^{3}\left(\beta_{j\max}\lambda_{jr\max} + \ln\left(\frac{\beta_{j\max}}{\sinh\beta_{j\max}}\right)\right) - c_3 \tag{32}$$

where

$$\lambda_{jr\max} = \frac{\lambda_{j\max}}{\sqrt{N_3}}, \quad j = 1, 2, 3, (\text{no sum}) \tag{33}$$

and $\beta_{j\max} \equiv \mathcal{L}^{-1}(\lambda_{jr\max})$.

Substitution of (30) into (10) and by using (11), we obtain the constitutive equation for the non-Gaussian, James–Guth virgin material model in the principal reference system:

$$T_{0j} = -p + \frac{\mu_0}{3}N_3\lambda_{jr}\mathcal{L}^{-1}(\lambda_{rj}),$$
$$j = 1, 2, 3, (\text{no sum}) \tag{34}$$

wherein μ_0 represents the shear modulus in the undeformed state and it is given by $\mu_0 \equiv nk\Theta$.

The James–Guth stress-softened material response in the same principal reference system, for exponential softening, is then provided by use of (34) in (18):

$$\tau_j = \left(-p + \frac{\mu_0}{3}N_3\lambda_{jr}\mathcal{L}^{-1}(\lambda_{jr})\right)\mathrm{e}^{-b\sqrt{W_{3\mathrm{chmax}}-W_{3\mathrm{ch}}}}, \quad j = 1, 2, 3, (\text{no sum}) \tag{35}$$

Application of (34) and (35) in the manner described earlier for equilibrium of the neo-Hookean material, yields the following simple uniaxial engineering stress-softened relation for a James–Guth material:

$$(\sigma_s)_{3-\mathrm{ch}} = \frac{\mu_0\sqrt{N_3}}{3}\left(\mathcal{L}^{-1}\left(\frac{\lambda}{\sqrt{N_3}}\right) - \frac{1}{\lambda^{3/2}}\mathcal{L}^{-1}\left(\frac{1}{\sqrt{\lambda N_3}}\right)\right)\mathrm{e}^{-b\sqrt{W_{3\mathrm{chmax}}-W_{3\mathrm{ch}}}} \tag{36}$$

for which $\lambda_{1r} = \lambda/\sqrt{N_3}$, $\lambda_{2r} = \lambda_{3r} = 1/\sqrt{\lambda N_3}$.

In the case of equibiaxial extension for which $\lambda_{1r} = \lambda_{2r} = \lambda/\sqrt{N_3}$, and $\lambda_{3r} = \lambda^{-2}/\sqrt{N_3}$ the equibiaxial engineering stress-softened relation is

$$(\sigma_s)_{3-\mathrm{ch}} = \frac{\mu_0\sqrt{N_3}}{3}\left(\mathcal{L}^{-1}\left(\frac{\lambda}{\sqrt{N_3}}\right) - \frac{1}{\lambda^3}\mathcal{L}^{-1}\left(\frac{1}{\lambda^2\sqrt{N_3}}\right)\right)\mathrm{e}^{-b\sqrt{W_{3\mathrm{chmax}}-W_{3\mathrm{ch}}}} \tag{37}$$

For a pure shear or plane strain compression deformation state, the corresponding compressive engineering stress is $\sigma_1 = T_{01}/\lambda$, the restraining stress is $\sigma_2 = T_{02}$, and the free surface stress is $\sigma_3 = 0$ and hence, the engineering stress-softened equation is given by

$$(\sigma_s)_{3-\mathrm{ch}} = \frac{\mu_0\sqrt{N_3}}{3}\left(\mathcal{L}^{-1}\left(\frac{\lambda}{\sqrt{N_3}}\right) - \frac{1}{\lambda^2}\mathcal{L}^{-1}\left(\frac{1}{\lambda\sqrt{N_3}}\right)\right)\mathrm{e}^{-b\sqrt{W_{3\mathrm{chmax}}-W_{3\mathrm{ch}}}} \tag{38}$$

6.3. The Arruda–Boyce non-Gaussian network model

For the Arruda–Boyce constitutive equation for an average-stretch, full-network of arbitrarily oriented molecular chains, the total strain energy per unit volume is given by

$$W_{8\mathrm{ch}} = \mu_0 N_8\left(\beta\lambda_r + \ln\left(\frac{\beta}{\sinh\beta}\right)\right) - c_8 \tag{39}$$

where λ_r is the relative chain stretch defined by

$$\lambda_r = \frac{\lambda_{\mathrm{chain}}}{\lambda_L} \tag{40}$$

where $\lambda_L = \sqrt{N_8}$, represents the fully extended chain stretch,

N_8 is the chain number of rigid links, each of length l, λ_{chain} is the chain deformation that in the affine deformation is given by

$$\lambda_{\text{chain}} \equiv \sqrt{\frac{I_1}{3}} \tag{41}$$

c_8 is a convenient constant chosen so that the strain energy vanishes in the undeformed state, and β defined by $\beta \equiv \mathcal{L}^{-1}(\lambda_r)$ is the inverse of the Langevin function $\mathcal{L}(\beta)$, thus:

$$\lambda_r = \mathcal{L}(\beta) \equiv \coth\beta - \left(\frac{1}{\beta}\right) \tag{42}$$

Note that the strain energy for the Arruda–Boyce model depends only on the principal invariant I_1 [13,14].

For this molecular network model, the maximum strain energy per unit volume at the greatest previous stretch is:

$$W_{8\text{chmax}} = \mu_0 N_8 \left(\beta_{\max}\lambda_{r\max} + \ln\left(\frac{\beta_{\max}}{\sinh\beta_{\max}}\right)\right) - c_8 \tag{43}$$

where

$$\lambda_{r\max} = \sqrt{\frac{I_{1\max}}{3N_8}} \tag{44}$$

and $\beta_{\max} \equiv \mathcal{L}^{-1}(\lambda_{r\max})$

Substitution of Eq. (39) into Eq. (10) and with the aid of (11), the Cauchy stress–stretch constitutive equation for the Arruda–Boyce 8-chain network model becomes:

$$\mathbf{T} = -p\mathbf{1} + \aleph(I_1)\mathbf{B} \tag{45}$$

where the material response function is defined by

$$\aleph(I_1) \equiv \frac{\mu_0\beta}{3\lambda_r} \tag{46}$$

and $\mathbf{B} = \text{diag}\{\lambda_1^2, \lambda_2^2, \lambda_3^2\}$ in the principal reference configuration. Using Eqs. (7), (17) and (45), the stress–stretch constitutive relation for a stress-softened Arruda–Boyce material is given by:

$$\boldsymbol{\tau} = (-p\mathbf{1} + \aleph(I_1)\mathbf{B})\mathrm{e}^{-b\sqrt{W_{8\text{chmax}} - W_{8\text{ch}}}} \tag{47}$$

Thus, the uniaxial engineering stress–stretch relation for an Arruda–Boyce stress-softened material is obtained by using Eqs. (46) and (47). This yields:

$$(\sigma_s)_{8-\text{ch}} = \frac{\mu_0}{3}\mathcal{L}^{-1}(\lambda_r)\left(\frac{\lambda - \lambda^{-2}}{\lambda_r}\right)\mathrm{e}^{-b\sqrt{W_{8\text{chmax}} - W_{8\text{ch}}}} \tag{48}$$

in which the relative chain stretch can be obtained from Eqs. (40) and (41):

$$\lambda_r = \frac{1}{\sqrt{3N_8}}\left(\lambda^2 + \frac{2}{\lambda}\right)^{1/2} \tag{49}$$

In equibiaxial extension, the corresponding Arruda–Boyce engineering stress-softened relation is:

$$(\sigma_s)_{8-\text{ch}} = \frac{\mu_0}{3}\mathcal{L}^{-1}(\lambda_r)\left(\frac{\lambda - \lambda^{-5}}{\lambda_r}\right)\mathrm{e}^{-b\sqrt{W_{8\text{chmax}} - W_{8\text{ch}}}} \tag{50}$$

where the relative chain stretch is determined by

$$\lambda_r = \frac{1}{\sqrt{3N_8}}\left(2\lambda^2 + \frac{1}{\lambda^4}\right)^{1/2} \tag{51}$$

In the case of a pure shear or plane strain compression deformation state, the compressive engineering stress-softened is described by

$$(\sigma_s)_{8-\text{ch}} = \frac{\mu_0}{3}\mathcal{L}^{-1}(\lambda_r)\left(\frac{\lambda - \lambda^{-3}}{\lambda_r}\right)\mathrm{e}^{-b\sqrt{W_{8\text{chmax}} - W_{8\text{ch}}}} \tag{52}$$

where the relative stretch (44) may be determined from the following equation:

$$\lambda_r = \sqrt{\frac{1}{3N_8}(\lambda^2 + \lambda^{-2} + 1)} \tag{53}$$

6.4. The full-network composite strain energy model

Wu and van der Giessen [19,20] proposed an approximation of their full strain energy network model by considering a linear combination of the James–Guth 3-chain and Arruda–Boyce 8-chain models in accordance with the following equation:

$$W_{\text{full}} = (1-\rho)W_{3\text{ch}} + \rho W_{8\text{ch}} \tag{54}$$

where the parameter ρ is given by the empirical relation

$$\rho = \frac{0.85\lambda_{\max}}{\sqrt{N}} \tag{55}$$

in which $\lambda_{\max} = \max(\lambda_1,\lambda_2,\lambda_3)$ and the factor 0.85 was chosen to provide the best correlation of (54) with numerical integration of the full network equation, $W_{3\text{ch}}$ is given by (30), $W_{8\text{ch}}$ by (39) together with (41) or (42), but only for $N_3 = N_8 = N$. Hence, the 8-chain contribution in (54) becomes increasingly important when $\lambda_{\max}$ approaches the chain locking stretch $\lambda_L = N$ of a single chain. Elías-Zúñiga and Beatty [21] proposed a phenomenological composite model based on relations modelled after (54) and (55), but having distinct coefficients that are tied to the ultimate extensibility of the network:

$$W_c = (1-\rho_{3-\text{ch}})W_{3\text{ch}} + \rho_{8-\text{ch}}W_{8\text{ch}} \tag{56}$$

In this composite full network model, Elías-Zúñiga and Beatty relate the coefficient for the 3-chain model contribution to an average measure of the ultimate chain stretch that accounts for both chain numbers N_3 and N_8. Hence, they defined a somewhat similar constant coefficient for the 3-chain constituent as

$$\rho_{3-\text{ch}} = \frac{a\Lambda_L}{\sqrt{N_3}} \tag{57}$$

where $\Lambda_L \geq 1$ is an average locking stretch defined by

$$\Lambda_L = \sqrt{\frac{N_3 + N_8}{2}} \tag{58}$$

and a is a positive scaling constant chosen to best fit experimental data for a given rubberlike material. It is seen that the coefficient (57) is thus determined by the fraction N_8/N_3.

In the case of the 8-chain network model, Elías-Zúñiga and Beatty defined the 8-chain constituent coefficient as

$$\rho_{8-\mathrm{ch}} = \frac{b_1 \Lambda_{\mathrm{ch}}(\Lambda_L)}{\sqrt{N_8}} \tag{59}$$

where b_1 is another positive, experimental scaling constant and $\Lambda_{\mathrm{ch}}(\Lambda_L)$ is the chain stretch (41) evaluated for $\lambda_{\max} = \Lambda_L$ in (58), that is,

$$\Lambda_{\mathrm{ch}}(\Lambda_L) = \frac{1}{\sqrt{3}} \left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2\right)^{1/2} \Big|_{\lambda_1 = \Lambda_L} \tag{60}$$

in which the principal stretches λ_k are ordered so that $\lambda_1 \geq \lambda_2 \geq \lambda_3$ and we set the greater principal stretch $\lambda_1 = \Lambda_L > 1$ given by (58).

Thus, with the introduction of (34), the stress components of (45) and the use of the constitutive Eq. (18), the engineering stress–stretch relations for the stress-softened composite full network material are given by:

$$\tau_i = \left(\left(1 - \frac{a\Lambda_L}{\sqrt{N_3}}\right)(T_i)_{3-\mathrm{ch}} + \frac{b_1 \Lambda_{\mathrm{ch}}(\Lambda_L)}{\sqrt{N_8}} (T_i)_{8-\mathrm{ch}} \right) \times \mathrm{e}^{-b\sqrt{W_{c\max} - W_c}}, \quad i = 1, 2, 3 \tag{61}$$

7. Comparison with experimental data

We now compare our strain energy-based Gaussian and non-Gaussian constitutive equations for simple uniaxial extension, equibiaxial extension, and pure shear deformations with experimental data obtained by Mullins and Tobin [3], Muhr [15], Cheng [16], Johnson and Beatty [17], and by Chagnon [18].

We begin with Mullins–Tobin data and use the James–Guth model to determine the material constants μ_0, N_3, and b in a plot of the engineering stress versus stretch. We require only three material constants. The shear modulus is first obtained as $\mu_0 = 0.853$ MPa, then the number of chain links $N_3 = 26.91$ is determined so that (34) provides a best fit to the full range of virgin material data shown in Fig. 1. With these values of the virgin material constants, we determine the softening parameter value of $b = 0.295$ (N m)$^{-1/2}$ to best fit the stress-softened material relation (36) to the corresponding Mullins–Tobin stress-softened material data shown in Fig. 1. The value of N_8 [21] may be obtained for simple extension and pure shear deformation states by the equation

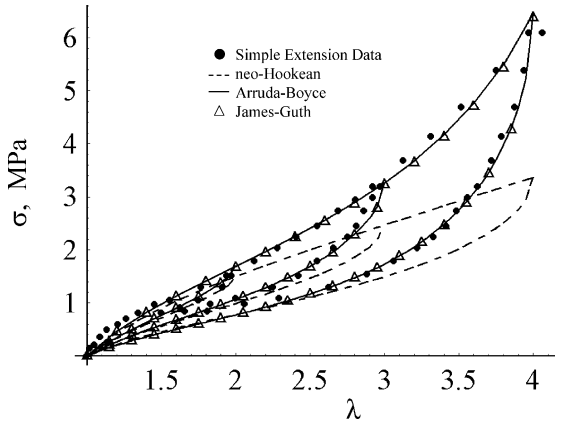

Fig. 1. Comparison of theoretical predictions of three molecular network models with Mullins–Tobin uniaxial extension data for which $\mu_0 = 0.853$ MPa, $N_3 = 26.91$, $N_8 = 9.12$ and $b = 0.295$ (N m)$^{-1/2}$.

$$N_8 = \frac{1}{3}\left(N_3 + \frac{2}{\sqrt{N_3}}\right) \tag{62}$$

and for simple compression and equibiaxial deformation states by

$$N_8 = \frac{1}{3}\left(2N_3 + \frac{1}{N_3^2}\right) \tag{63}$$

whereas the shear modulus and the stress-softening parameter for the Arruda–Boyce model retain the same values. The results predicted by the neo-Hooken Gaussian model, the James–Guth 3-chain and the Arruda–Boyce 8-chain relations are shown in Fig. 1. It is seen from Fig. 1 that both of the non-Gaussian network models compare favorably with Mullins–Tobin data. The results for the neo-Hookean, Gaussian network model vary significantly, particularly at the higher stretch values, where stiffening is apparent. This condition is also evident in Fig. 2, where Cheng experimental data for simple uniaxial extension for a ethylene–propylene–diene terpolymer (EPDM) material is shown [16]. The material constants for the network models

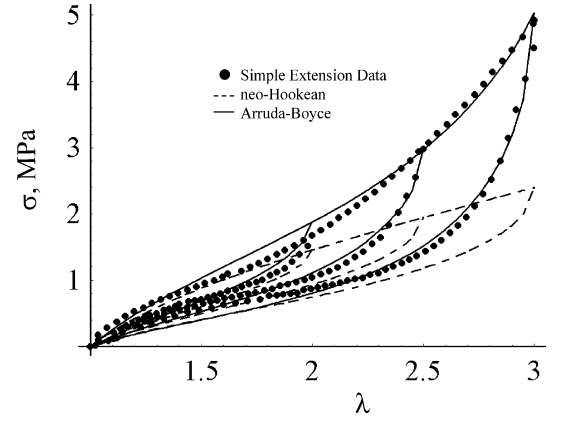

Fig. 2. Comparison of theoretical predictions of Arruda–Boyce molecular network model with Cheng uniaxial extension data for which $\mu_0 = 0.83$ MPa, $N_3 = 14.45$, $N_8 = 5.01$ and $b = 0.48$ (N m)$^{-1/2}$.

are provided by a best-fit analysis of simple uniaxial extension data of the virgin materials. These are listed in the figure captions.

Fig. 3 illustrates analytical results of Eqs. (27), (36) and (48) compared to experimental data in simple uniaxial extension obtain by Muhr in Ref. [15]. Note that the theoretical engineering stresses of the virgin neo-Hookean material model described by Eq. (27) show the highest deviation from experimental data. Our proposed Arruda–Boyce stress-softening constitutive relation for equibiaxial deformation (50) is compared further with balloon inflation experiments BALL 9 and BALL 10 obtained by Johnson and Beatty in Ref. [17]. We can see from Figs. 4 and 5 that the theoretical predictions of Arruda–Boyce 8-chain network model provides an excellent fit over the entire data range.

To further test our phenomenological stress-softening model, the engineering stress–stretch analytical prediction of the composite model given by Eq. (61) is compare in Fig. 6 with pure shear dimensionless data by Chagnon [18]. We may conclude from Fig. 6 that analytical predictions obtained from Eq. (61) capture the qualitative behavior but these do not provide a good fit to experimental data over the entire range. This can be explained by the fact that we are neglecting the viscoelastic material effects in our proposed phenomenological stress-softening model.

We emphasize that in all the above deformation states only three material constants are used in mapping experimental results.

8. Strain intensity phenomenological model

In this section we compare our proposed energy-based phenomenological model with the strain-based phenomenological model introduced by Elías-Zúñiga and Beatty in Ref. [1] by using simple extension and balloon inflation

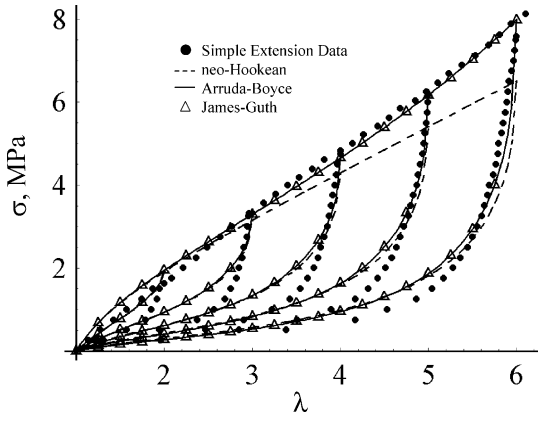

Fig. 3. Comparison of theoretical predictions of three molecular network models with Muhr uniaxial extension data for which $\mu_0=1.09$ MPa, $N_3=130.78$, $N_8=43.65$ and $b=0.45$ (N m)$^{-1/2}$.

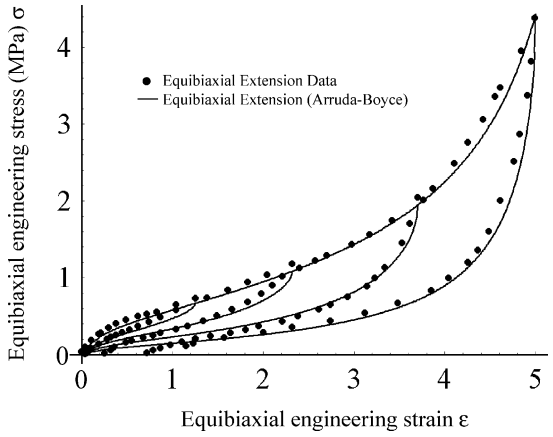

Fig. 4. Comparison of theoretical predictions of Arruda–Boyce molecular network model with equibiaxial inflation data (BALL 9) for which $\mu_0=63$ MPa, $N_8=33.11$, and $b=0.0245$ (N m)$^{-1/2}$.

experimental data. We begin with a briefly review of the strain intensity based phenomenological model.

8.1. Theoretical considerations

To compare the strain-based phenomenological model with our proposed energy based phenomenological model, we briefly recall here some of its main characteristics. In the strain intensity phenomenological model, the virgin material response during loading for which the maximum previous strain is its current value, $m_{\max}=m(t)$, is characterized by the time independent constitutive Eq. (10). The stress-softened material response for subsequent unloading and loading again from an undeformed state, or from any other elastic point for which $m<M$, is defined by the constitutive equation

$$\boldsymbol{\tau} = F(m;M)\mathbf{T}_0 \tag{64}$$

in which $\boldsymbol{\tau}$ denotes the Cauchy stress in the elastic stress-softened material and M represents the maximum previous

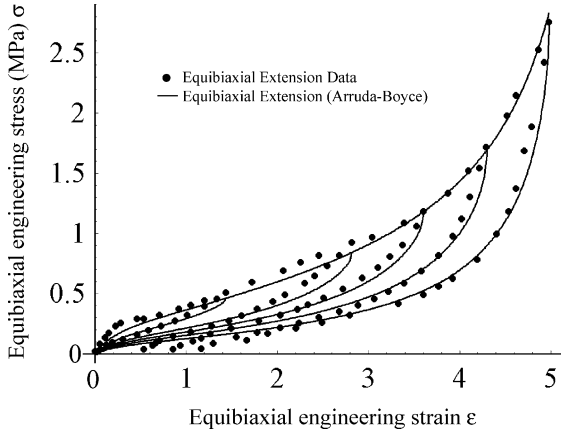

Fig. 5. Comparison of theoretical predictions of Arruda–Boyce molecular network model with equibiaxial inflation data (BALL 10) for which $\mu_0=39$ MPa, $N_8=33.11$, and $b=0.0245$ (N m)$^{-1/2}$.

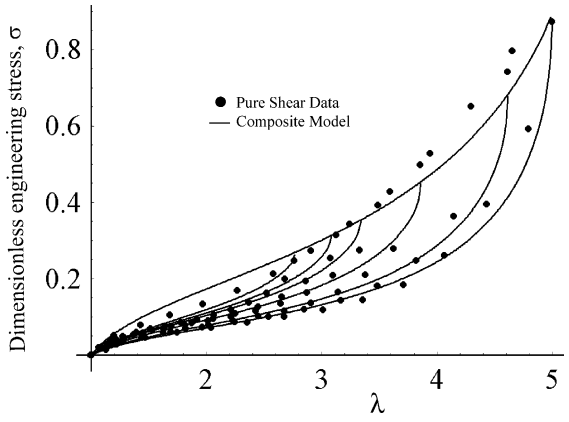

Fig. 6. Comparison of theoretical predictions of the phenomenological composite network model with pure shear dimensionless data for which we use the parameter values of $\mu_0=0.085$, $N_3=40.03$, $N_8=13.45$, $a=b_1=1$, and $b=1.1$.

strain at the point at which the material is unloaded from the virgin path. The isotropic, scalar-valued damage function $F(m;M)$, called the softening function at the damage level $m_{\max}=M$ on the interval $m\in\left[\sqrt{3},M\right]$ satisfies the conditions.

$$0<F(m;M)<1,\quad F(M;M)=1 \tag{65}$$

The softening function $F(m;M)$ is determined by a constitutive equation that describes the evolution of microstructural damage that begins immediately upon deformation from the natural, undistorted state of the virgin material. The constitutive equation that describes the softening function of the phenomenological model proposed by Elías-Zúñiga and Beatty in Ref. [1] is given by

$$F(m;M)=\mathrm{e}^{-b_s\sqrt{(M-m)}} \tag{66}$$

where b_s is a dimensionless positive material constant called the material softening parameter. Its value $F\left(\sqrt{3};M\right)$ characterizes the extent of the damage at M, initiated at $m=\sqrt{3}$.

In the course of loading from the natural state, the maximum value of the strain intensity is its current value, $m(t)=M$; and hence (64) reduces to the virgin material response given by Eq. (10). The material behavior described by (64) for $m\leq M$ is elastic for both loading and unloading until the value of m exceeds its maximum previous value M. During subsequent loading, the material recalls its inelastic virgin material response described by (10). By substituting Eq. (66) into Eq. (64), Elías-Zúñiga and Beatty obtain the following stress-softened material model:

$$\boldsymbol{\tau}=\mathbf{T}_0\mathrm{e}^{-b_s\sqrt{(M-m)}} \tag{67}$$

in which each member of the family of elastic stress-softened materials in (67) is generated from a specified virgin material model in the class (10).

8.2. Numerical results

To assess the accuracy between both phenomenological models, we shall compare their predicted analytical results with experimental data by Cheng [16], Chagnon [18], and Johnson and Beatty [17]. We study the stress-softened material response predicted by Eq. (18) and Eq. (67) by using the Arruda–Boyce 8-chain network model. We shall begin with experimental data by Cheng [16] for simple extension and plot the corresponding engineering stress-softened relations described above. The material parameter values used to simulate predicted results are shown in figure captions. Fig. 7 shows analytical results for both phenomenological models compared to experimental data.

Note that there is a slight difference between both models and that the strain based phenomenological model tends to underestimate experimental data for moderate stretches to about $\lambda=2$; but both models provide a good fit over the entire data range. We next show in Figs. 8 and 9 the comparison of analytical results with balloon inflation data by Johnson and Beatty. Once more, there is a slight difference between the phenomenological models but both predict experimental data fairly well. Finally, we compare in Fig. 10 analytical predictions with dimensionless uniaxial extension experimental data by Chagnon [18]. We can see in Fig. 10, that both phenomenological model results are in good agreement with these experimental data.

From the above figures, we can conclude that the energy based phenomenological model tends to fit, in general, experimental data better that the strain intensity phenomenological model and that the difference between their analytical predictions are more evident for small and moderate stretch values.

9. Concluding remarks

The success of our proposed energy based

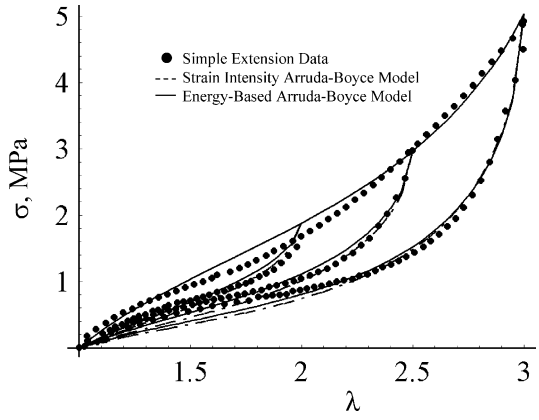

Fig. 7. Comparison of theoretical predictions of Arruda–Boyce molecular stress-softened phenomenological network models with Cheng uniaxial extension data for which $\mu_0=0.83$ MPa, $N_3=14.45$, $N_8=5.01$, $b_s=0.42$ and $b=0.48$ (N m)$^{-1/2}$.

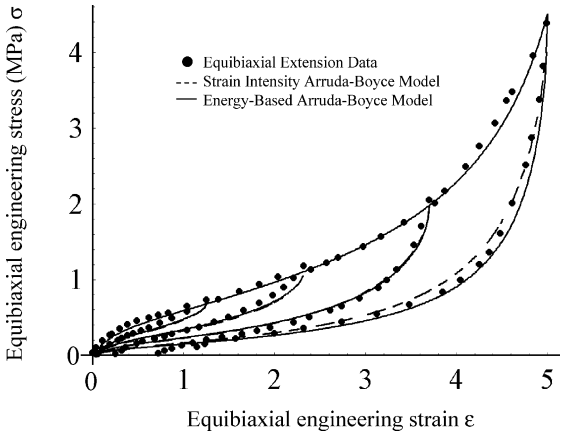

Fig. 8. Comparison of theoretical predictions of Arruda–Boyce molecular stress-softened phenomenological network models with equibiaxial inflation data (BALL 9) for which $\mu_0=63$ MPa, $N_8=33.11$, $b_s=0.19$ and $b=0.0245$ (N m)$^{-1/2}$.

phenomenological stress-softening material model (18) in characterizing the Mullins effect is shown to be very good in modeling the uniaxial extension data by Mullins and Tobin [3], Muhr [15], Cheng [16], and the balloon inflation data by Johnson and Beatty [17]. In the case of pure shear experimental data [18], our model fails in predicting with precision the material stress-softened behavior but this can be explained by the fact that our stress-softened phenomenological model neglects the viscoelastic material effects. However, our model requires determination of only three material constants: the shear modulus μ_0, the model specific molecular chain number of links N, and the softening rate parameter b. Moreover, comparison of our model developed in this paper with the strain based phenomenological model described by Eq. (67) shows that the energy based phenomenological model is slightly superior in following experimental data.

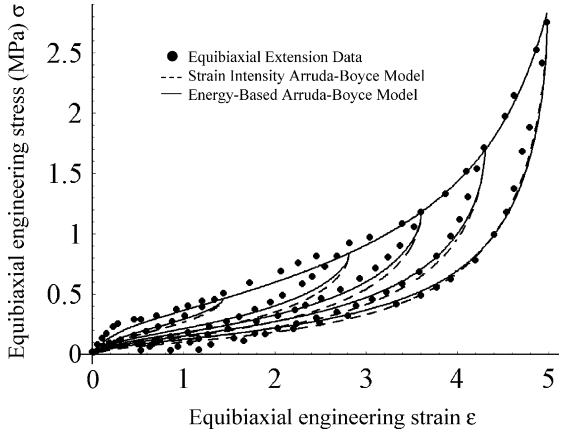

Fig. 9. Comparison of theoretical predictions of Arruda–Boyce molecular stress-softened phenomenological network models with equibiaxial inflation data (BALL 10) for which $\mu_0=39$ MPa, $N_8=33.11$, $b_s=0.19$ and $b=0.0245$ (N m)$^{-1/2}$.

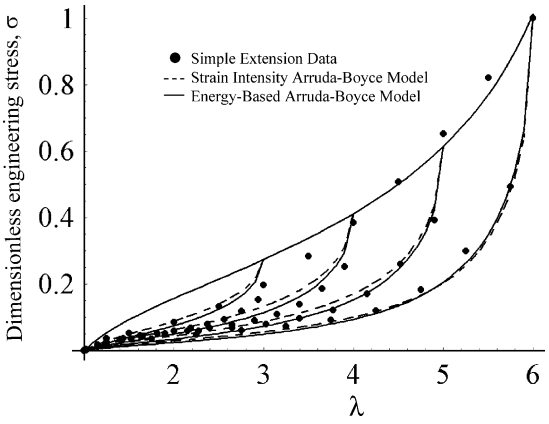

Fig. 10. Comparison of theoretical predictions of the phenomenological network models with uniaxial extension dimensionless data for which the material parameter values are: $\mu_0=0.085$, $N_8=19.5$, $b_s=1.25$ and $b=0.33$.

Finally, it is evident that our energy-based phenomenological model may be applied to three-dimensional deformation states since, the damage function is controlled by the strain energy associated with the primary deformation and not just by the specific deformation from which it is calculated. Besides, the extend of the damage sustained by our energy based phenomenological material model depends on the point, where the virgin material is unloaded from a state of maximum previous strain $W_{\max}$.

Acknowledgements

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