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A non-monotonous damage function to characterize stress-softening effects with permanent set during inflation and deflation of rubber balloons

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ABSTRACT

A non-monotonous stress-softening phenomenological model is applied to study the Mullins effect with residual strains to characterize the inflation and deflation of rubber balloons. It is shown that analytical predictions based on our proposed non-monotonous softening function and the modified stress-softening non-Gaussian average-stretch full-network constitutive equation that accounts for residual strains are consistent with experimental data. Also, we use the constitutive equation for equibiaxial extension to predict stress-softening behavior in a kinematically equivalent simple compression deformation state.

1. Introduction

The Mullins effect in equibiaxial extension has been studied both experimentally and analytically [1,2] in relation to balloon inflation and uniaxial compression. The study of this effect in equibiaxial deformation is greatly important in balloon inflation procedures encountered in biological tissue and biomedical applications, several of which are described recently by Guo [3]. Johnson and Beatty in [1] and Beatty and Krishnaswamy in [2] have investigated the stress-softening effect in the inflation of spherical membranes by assuming that at each material particle there coexists in the virgin material a fraction of material called soft phase and a fraction called hard phase. The material molecular structure is thus regarded as an amorphous mixture of soft phase and hard phase. Due to microstructural damage evolution, the hard phase is continuously transformed to soft phase; and this gives rise to the stress-softening phenomenon. Beatty and Krishnaswamy [4] applied their stress-softening model to describe the Mullins effect observed in the balloon inflation experiments [1]; but they provided no comparison of their analytical predictions with experimental data.

Clément et al. [5] have attributed the Mullins effect to the detachment of slippage of chains having reach their limited extensibility on the filler surface. Just before detaching or slipping, those chains, even if there are a few ones, can withstand large stresses, which gives a high contribution to the elastic modulus but a low contribution to the orientation. Subsequent *in situ* synchrotron wide-angle X-ray diffraction (WAXD) studies by Toki et al. [6] describe that during stretching, the majority of the amorphous chains are isotropic without preferred orientation and that the initial stress in the virgin material is mainly determined by vulcanized chains in the cross-linking network, whereas the final stress in the virgin material is determined by the network of strain-induced crystallization. For the stress-softened material, the effective network structure becomes different from the structure of the virgin path because some crystallities do not melt immediately.

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A network alteration theory for the Mullins effect was introduced by Marckmann et al. in [7]. In this approach, the material softening effect is the result of micro-damages accumulation that occur as a consequence of the breakage of links inside the material. The total or partial recovery of the material stiffness is mainly due to the interactions between polymer chains of the rubber matrix. Horgan et al. in [8] investigated the alteration idea proposed by Marckmann et al. [7] on the basis of the phenomenological model proposed by Gent [9] for rubber-like elasticity since this model facilitates discussion of connections with pseudo-elasticity and the theory of multiple reference configurations [10]. Horgan and co-workers found contradictions to what is reported by Marckmann et al. in [7] and concluded that there is no general agreement on the explanation of the microscopic and mesoscopic origins of the Mullins effect [8].

Ogden and Roxburgh [11] introduced a simple energy-based approach to model the Mullins effect in general biaxial deformations of filled rubber vulcanizates. In this approach, the virgin material response is described by a strain energy function for a perfectly elastic, incompressible and isotropic material in which the damage response of the virgin material is continuously updated. While the softening effect is described by an elastic unloading response through a pseudo-elastic energy function, which includes a certain damage variable that is activated only during unloading, or in subsequent reloading that does not exceed the previous maximum virgin material strain. De Tommasi and Puglisi [12] proposed a stress-softening phenomenological model based on a micromechanical scheme of a polymeric network reinforced with fine filler particles connected by elastic and breakable chains.

On the other hand, Ogden and Roxburgh in [13] and Holzapfel et al. in [14] used the pseudo-elastic theory to account for residual strains during material unloading. Elías-Zúñiga and Beatty [15] proposed a monotonous phenomenological three-dimensional continuum damage model for stress-softening of isotropic, incompressible hyperelastic rubber-like materials that describes the evolution of microstructural damage that begins immediately upon deformation from the natural, undeformed state of the virgin material. They studied the response of rubberlike materials subjected to uniaxial extension based on non-Gaussian molecular network models and found good agreement with uniaxial tension data [15]. However, in a recent publication Kazakevičiūtė-Makovska [16] observed that the experimental data when plotted as the normalized Cauchy stress components τ_{ij}/T_{ij} versus the stretch ratio $\lambda/\lambda_{\max}$ exhibited non-monotonous behavior with the characteristics S-shaped form and because of this, different values of the softening parameters are needed to fit experimental data for a particular choice of the softening function. Furthermore, De Tommasi et al. concluded in [17] that amorphous materials may be characterized by unstable strain domain which gives the possibility of having homogeneous or localized damage with a non-monotone primary loading curve behavior as the distribution properties are varied due to microscopic inhomogeneities. Based on these observations and the conclusions drawn by Kazakevičiūtė-Makovska [16], we propose here a non-monotonous softening damage function of the strain intensity that captures the characteristic S-shaped form in strain controlled experimental data of rubber-like materials subjected to loading and unloading cycles. Afterwards, we use this damage function and the theory of pseudo-elasticity to account for residual strains in a kinematically equivalent compression deformation state to investigate the Mullins effect in the inflation and deflation of rubber balloons by using a non-Gaussian molecular network model [18]. Viscoelastic and anisotropic effects are not included. We find good agreement of the theoretical model in comparison with balloon inflation data by Johnson and Beatty [1] and simple compression data due to Amin et al. [19].

2. Fundamentals of kinematics

We recall briefly some essential relations for finite equibiaxial 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 $\mathbf{X}$ 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 $\varphi = \{O; \mathbf{e}_k\}$ with origin O and orthonormal basis $\mathbf{e}_k$. An isochoric equibiaxial deformation is described by

$$x_1 = \lambda X_1, \quad x_2 = \lambda X_2, \quad x_3 = \lambda^{-2} X_3, \tag{1}$$

in which λ denotes the constant equibiaxial principal stretches in the plane normal to $\mathbf{e}_3$ in φ. The Cauchy–Green deformation tensor $\mathbf{B} \equiv \mathbf{F}\mathbf{F}^T$ has the form

$$\mathbf{B} = \lambda^2\mathbf{e}_{11} + \lambda^2\mathbf{e}_{22} + \lambda^{-4}\mathbf{e}_{33}, \tag{2}$$

where $\mathbf{e}_{jk} \equiv \mathbf{e}_j \otimes \mathbf{e}_k$ and $\mathbf{F}$ is the usual deformation gradient. In the undistorted state $\mathbf{F} = 1$, the identity tensor and the incompressibility $\det\mathbf{F} = 1$ is satisfied by (1). The magnitude of the strain at a material point $\mathbf{X}$, also called the *strain intensity* and denoted by m, is defined by $m \equiv \sqrt{\mathbf{B}\cdot\mathbf{B}} = \sqrt{\operatorname{tr}\mathbf{B}^2}$, where tr denotes the trace operation. Hence, by (2),

$$m = \sqrt{2\lambda^4 + \lambda^{-8}}. \tag{3}$$

We note that $m \geqslant \sqrt{3}$ for all λ, equality holding when and only when $\lambda = 1$, the undeformed state.

A stress-softening material [4], called a *Mullins material*, is characterized as an inelastic material having a selective memory of only the maximum previous strain experienced during its deformation history. The maximum previous strain at $\mathbf{X}$ is defined by $m_{\max} = M \equiv \max_{0\leqslant s\leqslant t} m(s)$, where the material is subjected to a deformation history up to the current time t and s