



Damage and healing effects in rubber-like balloons

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ABSTRACT

Inflation experiments on thin rubber-like balloons show a complex, history-dependent hysteretic behavior, important for many technological applications. Typically, this is ascribed to the occurrence of damage processes at the micro-scale level. The experimental pressure–strain and stress–strain responses [Johnson, M.A., Beatty, F.M., 1995. The Mullins effect in equibiaxial extension and its influence on the inflation of a balloon. *Int. J. Eng. Sci.* 33(2), 223–245], suggest that for successive cyclic experiments also the occurrence of healing for previously damaged material may play a crucial role (see [Diani, J., Fayolle, B., Gilormini, P., 2009. A review on the Mullins effect, *Eur. Polym. J.* 45, 601–612] and references therein). In this work we apply a recently proposed, micro-structure-based model for damage and healing effects in rubber-like materials to the inflation problem of a thin spherical balloon. The model, while keeping a computational efficiency, is shown to be in a significant qualitative agreement with the available experimental results.

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1. Introduction

Theoretical and experimental analyses of the inflation of thin balloons constitute a traditional subject of mechanics of materials. In particular, inflation experiments represent a widely used tool for the constitutive characterization of soft materials. Moreover, this field of research is of interest for several technological applications, ranging from aerostatic balloons (Pagitz, 2007) and inflatable structures (Tsunoda and Senbokuya, 2002), to pneumatic micro-actuators and sensors (Yoda and Konishi, 2002). The theoretical difficulties derive both from the typical shape-dependent instability, pointed out by sudden shape and/or volume changes during inflation (e.g. Alexander, 1971; Haughton, 1980; Pamplona et al., 2004; Verron and Marckmann, 2003), and from the important role played by damage and healing effects. A detailed description of some of the main interesting effects observed during these experiments can be found in (Beatty, 1987; Johnson and Beatty, 1995; Müller, 2004).

In this paper, we apply a recent constitutive model (De Tommasi et al., 2006; De Tommasi and Puglisi, 2007) for damage and re-cross-linking processes in rubber-like materials to the study of the inflation of thin spherical balloons. The model is based on a prototypical scheme for the polymeric network which assumes rigid particles (representing filler, such as carbon black or silica particles) connected by two kinds of links: elastic and breakable. In order to model the material disorder, with polymeric chains

characterized by variable lengths, the breakable fraction is assigned by a distribution of links with different activation and breaking thresholds. The remaining fraction is elastic and guarantees a residual strength of the material also at damage saturation. In (De Tommasi et al., 2008) it is shown that the material distribution properties play a crucial role on the evolution of damage, delivering a criterion to describe the observed transition between homogeneous and localized damage structures as well as between ductile and fragile macroscopic responses. Here we consider the three-dimensional extension of this prototypical scheme (see De Tommasi et al., 2006; De Tommasi and Puglisi, 2007) that is based on the assumption that at each material point of a continuum body there exists a distribution of materials with variable activation and breaking thresholds.

Another effect observed in rubber materials, important also in the inflation problem, is represented by the so called healing effect due to the re-cross-linking process of previously broken chains. In the inflation experiments it is shed in evidence by the different material behavior during the unloading–reloading cycles. In this perspective, an extension of the constitutive model described above was proposed in (D'Ambrosio et al., 2008), where the authors assumed that during unloading a given fraction of broken chains may reform. Here we propose an extension of this model which accounts for more general breaking and re-cross-linking behavior. We deduce a simple and compact analytic expression for the resulting constitutive laws and we show that the model keeps the numerical efficiency of the previous version.

The constitutive model is then applied to the case of a thin polymeric spherical balloon under cyclic loading processes of inflation with controlled volume. In order to simplify the analytic computa-

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tions, we restrict our attention to the class of homogeneous equibiaxial deformations, corresponding to spherically deformed configurations. It is well known that, even in this special case, geometric instabilities may occur (Ericksen, 1991; Müller, 2004) and we describe the possible coupling with damage effects. Worth noting, differently from previously proposed models, here we also describe the recross-linking effect, which permits the description of the internal hysteresis phenomenon. Finally, the mechanical behavior of the deduced model is compared with the experimental results reported in (Johnson and Beatty, 1995), obtaining a satisfactory qualitative agreement.

The paper is organized as follows. After a general discussion of the proposed constitutive model (Section 2) we study the case of equibiaxial deformations in Section 3, where the recross-linking effect is also discussed. In Section 4 we apply the model to the balloon problem; finally, connections of our analytical and numerical results with the experimental works of (Johnson and Beatty, 1995) are established and discussed.

2. Damage and healing in polymeric materials

In this section we briefly recall our constitutive assumptions. We refer the reader to (De Tommasi et al., 2006; De Tommasi et al., 2008) for a more detailed discussion of the model with an analysis of its predictability properties.

Let $\mathbf{f}: \mathcal{B}_0 \ni \mathbf{X} \mapsto \mathbf{x} \in \mathcal{B}$ be the deformation of a body \mathcal{B}_0 , $\mathbf{F} := \nabla \mathbf{f}$ the corresponding deformation gradient and $\mathbf{B} := \mathbf{F}\mathbf{F}^T$ the left Cauchy-Green tensor. Assume that at each point $\mathbf{X} \in \mathcal{B}_0$ a fraction $\alpha \in (0, 1)$ (elastic matrix) of the amorphous material is endowed with a hyperelastic, isotropic and incompressible behavior, assigned in terms of an elastic energy density

$$\varphi_e = \varphi_e(I(\mathbf{B}), II(\mathbf{B})), \quad (2.1)$$

with $I(\mathbf{B}) := \text{tr} \mathbf{B}$ and $II(\mathbf{B}) := \text{tr} \mathbf{B}^{-1}$ the first and second invariants of \mathbf{B} . The expression of the Cauchy stress in the elastic matrix is given by

$$\mathbf{T}_e = -\pi \mathbf{I} + 2\varphi_{e,1} \mathbf{B} - 2\varphi_{e,2} \mathbf{B}^{-1}, \quad (2.2)$$

where $\varphi_{e,1}$ and $\varphi_{e,2}$ denote the derivatives with respect to the first and second invariant, respectively, and π is the reactive stress maintaining the incompressibility constraint.

The remaining fraction $(1 - \alpha)$ takes care of the activation, breaking and recross-linking effects at the micro-scale level. Specifically, here we adopt an activation and a breaking criterion based on an isotropic scalar function s of the two invariants of \mathbf{B} , so that the damageable material is respectively activated and broken when

$$s(I, II) = s_a, \quad s(I, II) = s_b, \quad (2.3)$$

with $s_{,I} \geq 0$ and $s_{,II} \geq 0$ and s_a and s_b representing the activation and breaking thresholds, respectively.

Moreover, to take care of micro-structure disorder, we assume that the different activation and breaking events of the material particles at a given point of the body are regulated by a probability density function $f = \hat{f}(s_a, s_b)$, which satisfies $\hat{f}(s_a, s_b) \geq 0$, $\hat{f}(s_a, s_b) = 0$ for $s_a < s_0$ or $s_b \leq s_a$, where s_0 is the value of s at the undeformed state. The function \hat{f} satisfies the normalization condition

$$\int_{s_0}^{\infty} \left(\int_{s_0}^{s_b} \hat{f}(s_a, s_b) ds_a \right) ds_b = 1.$$

To describe the behavior of the material body \mathcal{B}_0 , given a deformation history at $\mathbf{X} \in \mathcal{B}_0$, we denote by \mathbf{F}_a and \mathbf{F}_b the deformation gradients corresponding to the activation and breaking states, respectively. Thus, if $\mathbf{B}_a = \mathbf{F}_a \mathbf{F}_a^T$ and $\mathbf{B}_b = \mathbf{F}_b \mathbf{F}_b^T$ denote the corresponding Cauchy-Green tensors, we have $s(I(\mathbf{B}_a), II(\mathbf{B}_a)) = s_a$ and

$s(I(\mathbf{B}_b), II(\mathbf{B}_b)) = s_b$. We then assume that the response of the damageable fraction of the material depends on the deformation gradient measured from the activation state \mathbf{F}_a ; on setting for the deformation measures from the activation state

$$\hat{\mathbf{F}} := \mathbf{F}\mathbf{F}_a^{-1}, \quad \hat{\mathbf{B}} := \hat{\mathbf{F}}\hat{\mathbf{F}}^T, \quad \hat{I} = I(\hat{\mathbf{B}}), \quad \hat{II} = II(\hat{\mathbf{B}}), \quad (2.4)$$

the response of the damageable part of the material may be given in terms of an energy density φ_d defined as follows:

$$\varphi_d := \begin{cases} 0 & \text{for } s(I, II) < s_a \\ \varphi_d(\hat{I}, \hat{II}) & \text{for } s_a \leq s(I, II) \leq s_b, \\ \varphi_d(\hat{I}_b, \hat{II}_b) & \text{for } s(I, II) > s_b \end{cases}, \quad (2.5)$$

where \hat{I}_b and \hat{II}_b are the invariants of the relative Cauchy-Green strain $\hat{\mathbf{B}}_b$ at the rupture. Hence, the arising expression of the Cauchy stress in the damageable material is given by

$$\mathbf{T}_d := \begin{cases} \mathbf{0} & \text{for } s(I, II) < s_a \\ 2\varphi_{d,1} \hat{\mathbf{B}} - 2\varphi_{d,2} \hat{\mathbf{B}}^{-1} & \text{for } s_a \leq s(I, II) \leq s_b, \\ \mathbf{0} & \text{for } s(I, II) > s_b \end{cases},$$

where $\varphi_{d,1} = \frac{\partial \varphi_d}{\partial \hat{I}}$, $\varphi_{d,2} = \frac{\partial \varphi_d}{\partial \hat{II}}$.

In (De Tommasi et al., 2006) the values of s_a and s_b are considered to be locally regulated by a general probability density $f = \hat{f}(s_a, s_b)$. There the authors show also the predictability of the model by describing simple cyclic uniaxial loading histories that allow to determine the probability function \hat{f} . Moreover, they propose the simplified assumption that the elastic range $\Delta = s_b - s_a$ of breakable material is constant, which leads to a one parameter distribution. Here we take into consideration a slight generalization of this assumption by assuming a more general dependence

$$s_b = \hat{s}_b(s_a) \quad (2.6)$$

with $\frac{ds_b(s_a)}{ds_a} > 0$. By this invertibility assumption, we may introduce the new function

$$s_\ell = \hat{s}_\ell(s) = \hat{s}_b^{-1}(s). \quad (2.7)$$

By this function we may obtain the value of the activation threshold ($s_a = s_\ell(s)$) of the material that breaks at $s = s_b$. This position allows us to reduce to a one parameter distribution function

$$f(s_a) := \hat{f}(s_a, \hat{s}_b(s_a)).$$

In what follows we evaluate the material response of our body under a generic deformation history. We take into consideration two cases: *irreversible damage*, when no healing effect is considered, and *reversible damage*, with the healing effect considered.

2.1. Irreversible damage

First, we analyze a system characterized by irreversible damage, without healing effect. Consider a strain history $\mathbf{F} = \mathbf{F}(t)$ and the associated history of the parameter s , defined as $s(t) = s(\mathbf{F}(t))$. Correspondingly we may define the history of the maximum past value of s : $s_M(t) = \max_{\tau < t} s(\mathbf{F}(\tau))$. Firstly, we notice that given $s = s(t)$, according with Eq. (2.3)₁ the activated material verifies $s_a \leq s(t)$. Secondly, we observe that in view of Eq. (2.3)₂ all the damageable material with $s_b < s_M(t)$ is broken. Granted the invertibility of \hat{s}_b in Eq. (2.6), this breaking condition can be recast in terms of s_a , so that the material verifying

$$s_a < s_f(t) := \hat{s}_b^{-1}(s_M(t)) \quad (2.8)$$

must be broken. Here we have introduced a new state function $s_f = s_f(t)$, whose physical meaning is the value of the activation threshold s_a for the last broken material. This material breaks at an instant $\bar{t} \leq t$ such that $s(\bar{t}) = s_M(\bar{t})$. Moreover, we may assume

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