



Theory and identification of a constitutive model of induced anisotropy by the Mullins effect



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ABSTRACT

Rubber-like materials present a stress softening phenomenon after a first loading known as the Mullins effect. Some recent experimental data on filled silicone rubber are presented in the literature, using uniaxial and biaxial tests to precondition samples thus induce some primary stress softening. A generic modeling based on the polymer network decomposition into an isotropic hyperelastic one, and a stress-softening evolutive one, is proposed taking into account the contribution of many spatial directions. A new stress softening criterion is built by means of a tensor that measures the repartition of energy in space. A general form of the stress softening function associated to a spatial direction is written by the way of two variables: one, the maximal eigenvalue of the energy tensor; the other, the energy in the considered direction. Finally, a particular form of constitutive equation is proposed. The model is fitted and compared to experimental data. The capacities of such modeling are finally discussed.

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1. Introduction

Rubber-like materials present a stress softening after a first loading cycle, known as the Mullins (1947) effect. Different definitions have been given to the Mullins effect. In this paper the Mullins effect is considered as the difference between the first and second loadings. Moreover, different studies have highlighted that this phenomenon induces anisotropy, since the stress softening is strongly dependent on the second load direction.

In a first approach, many isotropic models were proposed in the literature to describe stress softening. First, physical models taking into account the evolution of the chain network were proposed. Govindjee and Simo (1991) proposed a model based on the macromolecular network evolution by decomposition into a hyperelastic network and an evolving network. Marckmann et al. (2002) considered that the macromolecular network can be represented by the eight chains model (Arruda and Boyce, 1993). The model contains chain lengths and chain densities evolving with the maximal principal stretch. In another way, double network theory was developed (Green and Tobolsky, 1946) considering that the rubber-like material can be decomposed into a hard and a soft phase; the hard phase is transformed into soft phase with the stress softening. Different equations were proposed (Beatty and Krishnaswamy, 2000; Zúñiga and Beatty, 2002). At the same time, the damage theory was often used to describe the stress softening (Simo, 1987; Miehe, 1995; Chagnon et al., 2004). In another way, Li et al. (2008) associated the Mullins effect to the growth of cavities in the material and a compressible model was proposed. In a last point of view, Ogden and Roxburgh (1999) and Dorfmann and Ogden (2003) proposed models based on pseudo-elasticity. All these models fit experimental data more or less accurately in one loading direction, i.e.,

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without changing loading direction between the first and second loadings. For a more exhaustive review about these isotropic models, the reader can refer to [Diani et al. \(2009\)](#).

To improve the modeling and to fit anisotropic stress softening, new approaches were developed taking into account the difference of stress softening in each strain direction. At first, [Göktepe and Miehe \(2005\)](#) generalized the approach proposed by [Govindjee and Simo \(1991\)](#) taking into account a spatial repartition of the chains. In the same way, [Diani et al. \(2006a\)](#) proposed a generalization of the [Marckmann et al. \(2002\)](#) model by means of chains oriented into 42 or more directions in space. Using a phenomenological damage function, this model can describe different stress softening in different directions, with permanent deformation after unloading. [Dargazany and Itskov \(2009\)](#) proposed a similar approach by taking into account the existence of different chains with different lengths in each direction. They integrate the density of probability in each direction, by taking into account the network evolution at each step. [Shariff \(2006\)](#) proposed an anisotropic damage model that describes transverse anisotropy of Mullins effect, taking into account different damages in the three principal strain directions using a second-order damage tensor. In the same way, [Itskov et al. \(2010\)](#) proposed three damage evolution functions for the three principal strain directions. These functions are formulated in terms of material parameters that partly depend on the maximal principal stretch. Recently, [Dorfmann and Pancheri \(2012\)](#) proposed a phenomenological model, based on the theory of pseudo-elasticity, which includes scalar variables in the strain energy function to account for stress softening and changes in material symmetry.

Most of the anisotropic models mentioned above are proposed by analyzing successive tensile tests performed along different directions. In spite of that, [Machado et al. \(2012a\)](#) have recently performed other original tests based on preconditioning with uniaxial tension and biaxial tension tests. Based on [Machado et al. \(2012a\)](#)'s experimental results using silicone rubber, this paper proposes a new approach for modeling the induced anisotropy by the Mullins effect. In [Section 2](#), the global framework of the Mullins effect modeling is presented. In [Section 3](#), a new approach is proposed to write constitutive equations by introducing a tensor that describes the strain energy repartition in the space directions. The conditions to be verified by the equations are detailed. In [Section 4](#), a first constitutive equation is proposed. It is fitted and compared to experimental data. Finally, [Section 5](#) contains some concluding remarks and outlines some future perspectives.

2. Macromolecular approach to model Mullins effect

2.1. Filled silicone behavior

In the last few years, different tests highlighting the stress softening anisotropy have been presented in the literature for different rubber-like materials, see for example [Muhr et al. \(1999\)](#), [Besdo et al. \(2003\)](#), [Hanson et al. \(2005\)](#), [Diani et al. \(2006b\)](#), and [Dorfmann and Pancheri \(2012\)](#). In this paper, attention is focused on the largest and most diverse database concerning Mullins effect anisotropy of a rubber-like material to the best of our knowledge. These data concern the results for the RTV3428 filled silicone rubber ([Machado et al., 2010, 2012a](#)).

First classical experimental tests, i.e., cyclic experiments with an increasing deformation after each cycle, were realized during tensile, pure shear and equibiaxial tensile tests. The data are reported in [Machado et al. \(2010\)](#). Second, stress softening anisotropy is presented in [Machado et al. \(2012a\)](#) induced by two distinguished preconditioning methods. The first one (noted as TT in the following) consists in a first loading in tension and a second loading also in tension, in four different directions. The second preconditioning method (noted as BT) consists in a first biaxial extension loading with constant principal strain directions. It is followed by a second loading in tension along the two principal strain directions of the first biaxial loading. The originality of these data is that loading states are very different between the first and second loads.

These new experimental results question the existing anisotropic constitutive equations and the main reasons are detailed here. The first reason is, [Diani et al. \(2006a\)](#) and [Dargazany and Itskov \(2009\)](#) models present an important permanent deformation that is related to the stress softening. But here, the material exhibits an important stress softening without permanent deformation. The other reason is that, for a second tensile loading orthogonal to the first loading, the models of [Shariff \(2006\)](#) and [Itskov et al. \(2010\)](#) present a stiffer behavior than the virgin material, which is not the case of the filled silicone rubber. Last, all these models are based on a set of material directions and Mullins effect is controlled in each direction only by the maximum stretch reached during the deformation history along the considered direction. Recently, [Merckel et al. \(2011\)](#) analyzed the damage spatial repartition and proposed a softening criterion ([Merckel et al., 2012](#)) that is still the maximum stretch in each direction. Therefore, as pointed out in [Machado et al. \(2012a\)](#), a maximal deformation criterion that depends only on the considered direction is not enough to describe the stress softening for an arbitrary second load direction. This means that, if the maximal principal direction remains the same during the first and second load cycles, the strain energy can be a measure to quantify the Mullins effect in this direction. In the other directions, a coupling effect exists between different directions and it influences the stress softening. Under these circumstances, a new way to handle Mullins effect should be proposed at the sight of [Machado et al. \(2012a\)](#) experimental data.

2.2. Two networks theory

The results presented using silicone rubber-like materials highlight that unfilled silicone rubbers do not present stress softening ([Rey et al., 2013](#)) whereas filled silicone rubbers ([Machado et al., 2010](#)) present an important one. For this silicone

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