



A Mullins softening criterion for general loading conditions

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ABSTRACT

Samples of carbon-black filled styrene butadiene rubbers (SBRs) were submitted to successive nonproportional loadings in order to define a general criterion for the Mullins softening. For this purpose, each sample was initially submitted to uniaxial or biaxial preloadings followed by a cyclic uniaxial tension test. An original experimental analysis aimed at defining the activation threshold for the Mullins softening during cyclic uniaxial loadings. The experimental data provide substantial evidences establishing the surface of the maximum directional stretch undergone by the material as a relevant Mullins softening criterion. The latter was used to successfully predict the Mullins softening surfaces for additional loading cases.

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1. Introduction

Carbon-black filled rubbers exhibit substantial softening when stretched for the first time. This phenomenon is generally referred to as the Mullins softening for the amount of work dedicated to the topic by Mullins (1947, 1949, 1950, 1969). Despite numerous contributions on the Mullins softening over the past decades, no general agreement has been found yet on the activation criterion of this phenomenon for general loading conditions. The objective of this contribution is to provide such a criterion essential for filled rubber constitutive modeling.

The Mullins softening may be illustrated by the stress–stretch response of a filled rubber submitted to a cyclic uniaxial tensile test with increasing maximum stretch. Fig. 1a highlights the stress-softening undergone by a filled rubber when first loaded. One may notice that once the Mullins softening is evacuated, the material behavior evolves very slowly. To the contrary, when stretching the material beyond the maximum intensity previously applied, it softens substantially. One may notice also that while the material softens some residual stretch appears and increases with the maximum stretch applied. Mullins (1947, 1949) conducted extensive experimental studies showing the dependence of filled rubbers softening and residual stretch with the maximum stretch for uniaxial tension tests. The same author revealed the anisotropy of both the Mullins softening and the residual stretch by applying successive nonproportional loadings (i.e. successive loadings, changing the directions of stretching). Subsequent experimental studies focused mainly on proportional loadings, and only a few authors (Laraba-Abbes et al., 2003; Hanson et al., 2005; Diani et al., 2006a,b; Itskov et al., 2006; Dargazany and Itskov, 2009; Machado, 2011) conducted nonproportional loadings showing the anisotropy induced by a preloading when applying a series of loadings according to various directions on the same sample.

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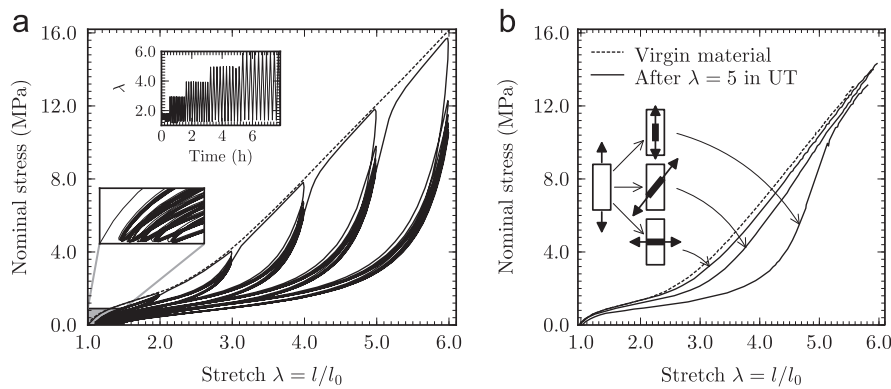


Fig. 1. Mullins softening: (a) Stress–stretch response of a filled rubber under cyclic uniaxial conditions. (b) Evidence of uniaxial tension (UT) preloading induced anisotropy.

At room temperature, the Mullins softening is generally considered as an irreversible damage and therefore is modeled by damage parameters and their evolutions (see [Diani et al., 2009](#) for a review). Most models use an isotropic damage definition for which the damage criterion may be defined in terms of the highest eigenvalue of the deformation gradient tensor ([Govindjee and Simo, 1991, 1992](#)), or of the maximum of any deformation tensor invariant ([Lion, 1996](#); [Krishnaswamy and Beatty, 2000](#); [Chagnon et al., 2004](#); [Elías-Zúñiga, 2005](#)), or of the maximum free energy ([Simo, 1987](#); [Ogden and Roxburgh, 1999](#)), or of other scalar quantities. This idealized isotropic representation of the Mullins softening does not account for the anisotropy induced by the Mullins softening. Actually, very few models deal with an anisotropic criterion. One may cite [Göktepe and Miehe \(2005\)](#) accounting for directional damage parameters depending on the maximum directional free energy, [Diani et al. \(2006a,b\)](#) and [Dargazany and Itskov \(2009\)](#) considering directional maximum stretches as directional criterion of damage activation, and [Itskov et al. \(2010\)](#) using the maximum stretches in the principal directions of the current deformation gradient tensor only. In what follows, an original analysis of nonproportional loading tests provides experimental evidences that the maximum directional stretch surface defines a relevant three-dimensional criterion for the Mullins softening activation.

This paper is organized as follows. The next section presents the experimental setup and describes the original method used to detect the Mullins softening activation. This section also provides the experimental results obtained under nonproportional loading conditions. In [Section 3](#), the Mullins softening criterion is defined and validated for additional loading cases. Finally, concluding remarks close the paper.

2. Experimental determination of the Mullins softening activation

2.1. Experimental setup

For this study, we used a 40 phr carbon-black filled styrene butadiene rubber (SBR) processed by Michelin into final plates of 2.5 mm thickness. The material in-plane isotropy was verified by punching dumbbell samples in various directions in the plates and testing them in uniaxial tension. In order to submit the material to uniaxial and biaxial loading conditions, two testing machines were used. The uniaxial tension tests were conducted on an Instron 5882 uniaxial testing machine at a constant crosshead speed which was chosen in order to reach an average strain rate close to 10^{-2} s^{-1} . Biaxial tests were carried out on a planar biaxial testing device built with four perpendicular electromechanical actuators controlled independently. Any biaxial test is characterized by the biaxial ratio R

$$R = \frac{F_{22} - 1}{F_{11} - 1} \quad (1)$$

with F_{11} and F_{22} being the longitudinal and transverse stretchings, respectively. Biaxial tension conditions were set such that $F_{11} \geq F_{22}$ and R ranging from 0 (pure shear) to 1 (equi-biaxial tension). All biaxial tests were run at constant crosshead speed corresponding to an average strain rate close to 10^{-2} s^{-1} in the direction 1. For proportional uniaxial tension tests, dumbbell samples of dimension 30 mm long and 4 mm wide were used. For biaxial preloading tests, cross-shaped samples were used. For uniaxial preloadings, large dumbbell specimens, 25 mm wide and 60 mm long, were used. Finally, for cyclic uniaxial tension tests 4 mm wide and 10 mm long dumbbell samples were punched in biaxially or uniaxially preloaded specimens. Strains are measured locally by video extensometry for both machines. Uniaxial tensile test resulting stress is defined by $\sigma = F\lambda/S_0$ with F being the force, $\lambda = l/l_0$ being the stretching, and S_0 being the initial sample cross-section. Let us note that σ denotes the Cauchy stress when incompressibility is assumed, which is common for filled rubbers. Biaxial tests were used for preloadings only, and forces were not recorded for these tests. During subsequent cyclic loadings, the tensile stretch is measured using two paint marks and is defined as $\lambda = l/l_0$, with l_0 being the initial distance between the paint marks for the material virgin of any loading.

At this point, let us clarify some notations that will be used along the study, biaxial preloadings are characterized by the ratio R and the maximum value of stretching is denoted as F_{11} , uniaxial preloadings are characterized by the applied

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