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A micro–macro approach to rubber-like materials. Part III: The micro-sphere model of anisotropic Mullins-type damage

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Abstract

A micromechanically based non-affine network model for finite rubber elasticity and viscoelasticity was discussed in Parts I and II [Miehe, C., Göktepe, S., Lulei, F., 2004. A micro–macro approach to rubber-like materials. Part I: The non-affine micro-sphere model of rubber elasticity. J. Mech. Phys. Solids 52, 2617–2660; Miehe, C., Göktepe, S., 2005. A micro–macro approach to rubber-like materials. Part II: Viscoelasticity model for polymer networks. J. Mech. Phys. Solids, published on-line, doi:10.1016/j.jmps.2005.04.006.] of this work. In this follow-up contribution, we further extend the micro-sphere network model such that it incorporates a deformation-induced softening commonly referred to as the Mullins effect. To this end, a continuum formulation is constructed by a superimposed modeling of a crosslink-to-crosslink (CC) and a particle-to-particle (PP) network. The former is described by the non-affine elastic network model proposed in Part I. The Mullins-type damage phenomenon is embedded into the PP network and micromechanically motivated by a breakdown of bonds between chains and filler particles. Key idea of the constitutive approach is a two-step procedure that includes (i) the set up of micromechanically based constitutive models for a *single chain orientation* and (ii) the definition of the macroscopic stress response by a *directly evaluated homogenization* of state variables defined on a micro-sphere of space orientations. In contrast to previous works on the Mullins effect, our formulation inherently describes a *deformation-induced anisotropy* of the damage as observed in experiments. We show that the experimentally

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observed *permanent set* in stress–strain diagrams is achieved by our model in a natural way as an anisotropy effect. The performance of the model is demonstrated by means of several numerical experiments including the solution of boundary-value problems.

Keywords: Micromechanics, Rubber-like materials; Mullins effect; Network models; Permanent set; Experiments; Simulations

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1. Introduction

In Parts I and II (Miehe et al., 2004; Miehe and Göktepe, 2005) of our work on rubbery polymers we developed network models for non-affine elasticity and viscoelasticity, respectively. In the present report we further exploit the basic features of these micro-sphere models for the description of the deformation-induced stress softening in rubbers with a particular account of induced anisotropy.

1.1. The phenomenon of stress softening in rubbers

Investigations on the effect of stress softening in cyclic tensile stretching of rubber specimens can be traced back to the beginning of the last century starting with the works Bouasse and Carrière (1903) and Holt (1932). The observed deformation-induced softening phenomenon has been referred to as the *Mullins effect* due to his detailed investigations starting from the late 1940s. Mullins (1948, 1969) reported the following basic phenomenological observations: (i) A substantial softening only occurs in rubbers at stretch values greater than a maximum obtained in the previous deformation history. (ii) The degree of softening increases with the initial volume fraction of the filler, characterizing the softening effect as the breakdown of the stiffening microstructure due to fillers. (iii) An anisotropic directional dependence of the softening was observed and supported by swelling experiments. (iv) The healing of softening observed was very slow and complete recovery was never achieved. (v) A softening behavior was also observed in compression tests, as also reported recently by Bergström and Boyce (1998). An *idealized description* of the Mullins effect in a quasi-static cyclic uniaxial tensile test with successively increasing stretches is schematically depicted in Fig. 1b. This idealization neglects slight viscous hystereses of the unloading–reloading curves depicted in Fig. 1a, which are observed also at very slow deformation rates. A common procedure is to interpret either retraction or reloading curves in Fig. 1a as the idealized softened response of Fig. 1b, see for example Mullins and Tobin (1957), James and Green (1975) or Marckmann et al. (2002). Furthermore, it was observed that the retraction curves get zero stress value at non-zero strain levels. This so-called *permanent set phenomenon* is a basic indication of a *characteristic anisotropy* due to the strain-induced softening response. It has often been eliminated in context of isotropic damage models as shown in Fig. 1b by shifting the unloading curves to the left by the amount of the permanent set, see for example Mullins and Tobin (1957). In contrast, an important goal of this