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J. Plagge, M. Klüppel

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# A Physically Based Model of Stress Softening and Hysteresis of Filled Rubber Including Rate- and Temperature Dependency

J. Plagge, M. Klüppel

*Deutsches Institut für Kautschuktechnologie e.V., Eupener Str. 33, 30519 Hannover, Germany*

*jan.plagge@DIKautschuk.de*

**Abstract**

A novel physically based material model is presented that describes the complex stress-strain behavior of filled rubbers under arbitrary deformation histories in a constitutive manner. The polymer response is considered by the extended non-affine tube model. Stress softening is taken into account via the breakdown of highly stressed polymer-filler domains under load and homogenization of the medium. Set stress and hysteresis are introduced via a continuous reformation mechanism, characterized by a single critical stress parameter. The latter is predicted to be dependent on temperature and deformation rate by means of Kramers escape rate. This is confirmed for a wide range of temperatures and speeds by fitting to multihysteresis measurements carried out in a heat chamber. Fitting parameters reveal that the mechanism responsible for hysteresis and set stress takes place on the nanometer scale with energies of roughly 100 kJ/mol. The behavior of the fitting parameters is analyzed for varying filler loadings and crosslinker concentrations in EPDM. Simulations of the stress-strain response for several deformation modes are in good agreement with experiments and its mathematical simplicity makes it very promising for applications with Finite Element Methods (FEM).

*Keywords:* A. Microstructures, B. Finite strain, A. Rate-dependent material, B. Viscoelastic material, B. Constitutive behavior

## 1. Introduction

Reinforced elastomers consisting of a polymer and a nano-structured filler like carbon black or silica are present almost everywhere in our lives. The most prominent example is automotive tires, where its incredible wear- and fatigue properties are still unmatched. An unfilled elastomer is almost ideally hyperelastic, e.g. exhibiting small hysteresis and weak rate dependency. The incorporation of fillers greatly toughens and stiffens the polymer, but also creates memory-like material properties as increased hysteresis and stress softening. For small dynamic deformations the latter effect is known as Payne-Effect (Payne, 1962) and is attributed to the irreversible breakdown of a stiff, percolated filler network (Rendek and Lion, 2010). This effect was extensively described by Payne and investigated by Kraus (1984) and Medalia (1978). A similar phenomenon can be observed for larger deformations, which was investigated by Mullins and Tobin (1965): The greater the deformation the material was subject to, the softer the materials response. The change in the response is not immediate, but requires several cycles to reach a steady state. The explanation for this effect is the complex interplay and structural rearrangement of filler particles and the polymer network, as reviewed in Vilgis et al. (2009). In particular, a breakdown or slippage of adsorbed polymer chains at the filler surface (Dannenberg, 1975; Bueche, 1960) and disentanglement of chains adsorbed at the filler surface (Hamed and Hatfield, 1989) or a rearrangement of network junctions in filled systems (Payne and Kraus, 1965) has been proposed. A constitutive model of stress-induced desorption of chains from the filler surface has been derived by Govindjee and Simo based on a statistical mechanics approach (Govindjee and Simo, 1991, 1992). This idea was recently extended to include deformation of filler aggregates as well, allowing the calculation of hysteresis (Dargazany and Itskov, 2013). Reese (2003) built a material model on the basis of the Neo-Hooke material model, taking into account breaking and reforming of chains by using several assumptions about chain distribution to arrive at a continuum level description.