Journal of the Mechanics and Physics of Solids 81 (2015) 58–74

Contents lists available at ScienceDirect

Journal of the Mechanics and Physics of Solids

journal homepage: www.elsevier.com/locate/jmps

Micro-sphere model for strain-induced crystallisation and three-dimensional applications

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ARTICLE INFO

Article history:
Received 30 September 2014
Received in revised form
1 February 2015
Accepted 13 May 2015
Available online 15 May 2015

Keywords:
Strain-induced crystallisation
Micro-sphere model
Anisotropy
Pure shear test
Constitutive model

ABSTRACT

Strain-crystallising rubber exhibits interesting properties: for instance, fatigue lifetime is known to be modified by this microstructural evolution which dissipates energy and creates a strong anisotropic reinforcement. We develop herein a micro-sphere 3D constitutive model for such strain-crystallising rubber. It is based on a simplified 1D micro-mechanical model that we extend with a micro-sphere approach to a full thermodynamically consistent evolutive anisotropic model. A specific numerical strategy is then proposed. The model is assessed on several significative configurations and reproduces the main experimental features while predicting the evolution of anisotropy as a function of the loading history. We finally show that it can also predict the crystallised zone in front of a mode I crack.

1. Introduction

Natural Rubber (NR) has an ability to form small crystals under strain (Strain Induced crystallisation, SIC). This remarkable property is recognised to be at the origin of its excellent mechanical properties, namely resistance to crack growth and fatigue behaviour. Experimental studies related to this phenomenon are abundant; we are particularly interested by simultaneous mechanical and X-Ray diffraction experiments as they may correspond to our modelling efforts. These experiments have been well-gathered in Huneau (2011). SIC is still a current topic of interest both from an experimental and a modelling perspective. In particular, since 2011, several Wide Angle X-Ray Diffraction (WAXD) studies have been published that deal with crystallisation kinetics and morphology at the mesoscale (Hernandez et al., 2013; Albouy et al., 2013; Bruening et al., 2013; Candau et al., 2012; Tosaka et al., 2012; Toki et al., 2013; Che et al., 2013).

Of particular interest are cyclic traction experiments (Fig. 1) in which crystallinity, elongation and stresses are simultaneously measured. Such experiments lead to several key observations (Fig. 1):

- From 0 to A: on loading, crystallisation is absent.
- From A to B: crystallisation starts at $\lambda = 4.5$, it is very slow at the beginning, then accelerates. The curve in green circles is measured for dynamic loading at 80 °C, a temperature where no crystallisation is present, and scaled assuming stress varies linearly with temperature. This allows us to ascribe the A to B low modulus to crystallisation. It is believed (Marchal, 2006) that two effects are competing: crystallites act as reticulation nodes, therefore enhancing the modulus, but Flory relaxation of the amorphous chains decreases the stress.

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http://dx.doi.org/10.1016/j.jmps.2015.05.004
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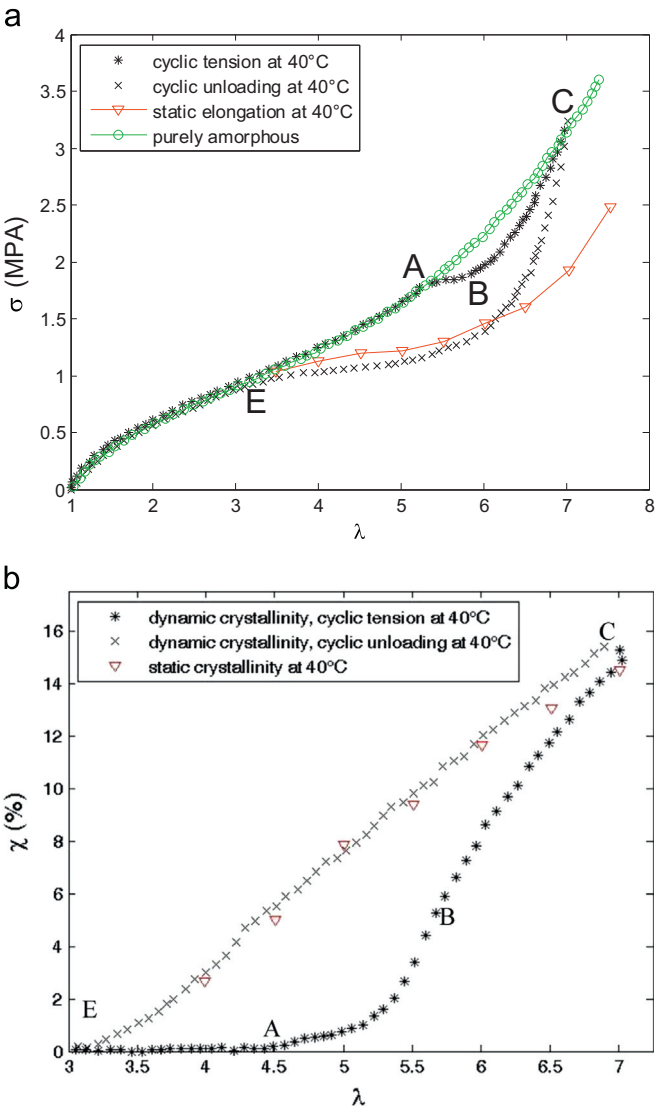

Fig. 1. Response curve of a specimen submitted to cyclic uniaxial tension: cycles of stress and crystallinity as a function of elongation (dynamic measurements). The green curve (green circles) is a loading curve, at a temperature where there is no crystallisation, scaled with temperature (see text); hysteresis is absent with these conditions (not shown). The red triangles correspond to static measurements (see text). Taken from Marchal (2006). (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

- From B to C: the material hardens at high elongation: the slope of the stress curve is noticeably higher than what is observed for a totally amorphous phase (green circles). It is assumed that crystallites are the more efficient as reticulation nodes the greater the elongation.
- From C to E: during unloading chains return progressively to an amorphous state (kinetics experiments show that on unloading equilibrium is reached within fraction of milliseconds, Rublon et al., 2012). The reference is given by experiments of static loading: a stress is imposed at 40 °C. Subsequently the sample is heated at 80 °C, at fixed elongation, then the sample is cooled and its stress and crystallinity are measured at 40 °C (red triangles). This allows thermodynamic equilibrium to be reached. From C to E, one can see that there is no delay of crystallites de-crystallisation by comparing the static crystallinity measurement to the cyclic unloading.

There is a long tradition of SIC modelling dating back to Flory (1947). These models have been discussed and modified later by Gaylord (1990). They assume that the material is at any time at equilibrium. One challenging difficulty is then to introduce an hysteretic response. Different models have been recently developed for this purpose by Kroon (2010), Mistry and Govindjee (2014), Guilie et al. (2013) or Guilie (2014). They share three basic ingredients: a micromechanical description of the individual chain, a constitutive law describing the evolution of crystallinity at local level, and a micro-to-macro extension paradigm.