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ScienceDirect
International Journal of Engineering Science 46 (2008) 758–774
International Journal of Engineering Science
www.elsevier.com/locate/ijengsci

Influence of thermally induced scission and crosslinking on the post-scission inflation of circular elastomeric membranes

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Received 29 November 2007; received in revised form 30 January 2008; accepted 31 January 2008
Available online 18 April 2008

Abstract

When an elastomeric material is deformed and subjected to temperatures above some characteristic value T_{cr} (near 100 °C for natural rubber), macromolecular chains and crosslinks undergo scission, recoiling and re-crosslinking. The process depends on time and temperature and continues until the temperature decreases below T_{cr}. Compared to the virgin material, the new material system has modified properties (reduced stiffness) and permanent set on removal of the applied load.

A constitutive theory that accounts for these changes in macromolecular structure is used to simulate an experiment involving the inflation of an initially flat circular membrane. The membrane is initially at a temperature $T < T_{cr}$. A prescribed volume of fluid inflates the membrane to a surface of revolution. The temperature of the fluid and membrane is increased above T_{cr} for some period of time. Attention is restricted to the case when scission is independent of the deformation of the membrane. It is shown that the time dependence of the pressure in the inflating fluid must satisfy a specific relation involving a time dependent property measured during scission under uniaxial extension. This result provides a useful experimental means for determining when scission is independent of deformation in biaxial deformations.

The temperature is returned to its original value and the pressure is gradually removed. It is shown that compressive stresses may develop as the pressure is reduced. If the membrane is removed from its support, it will have developed a permanently curved shape. Results of a numerical simulation are presented that compares the initial inflated shape to the permanently curved shape.
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Keywords: Chemomechanics; Scission; Crosslinking; Elastomer; Membrane inflation

1. Introduction

The temperature in an elastomeric structural component, such as a bushing, seal or tire, can increase during service due to its operating environment or due to internal dissipation. At a sufficiently high temperature, the

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0020-7225/$ - see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ijengsci.2008.01.020

microstructure of the elastomer can undergo changes consisting of the scission of macromolecular networks and their subsequent crosslinking to form new networks with different stress free configurations. This process is time and temperature dependent and can result in significant changes in the performance of the component.

A constitutive theory for materials undergoing microstructural changes was presented by Wineman and Rajagopal [1] and Rajagopal and Wineman [2]. This theory considered microstructural changes that result from deformation-induced scission. Microstructural changes that result from thermally induced scission has been investigated by the present authors and their co-workers. They used the constitutive structure in [1,2] to formulate a constitutive theory for the interaction of these microstructural changes during large deformations of elastomers [3–6], investigated it experimentally [4–6] and explored its implications for structural response in a variety of applications [7–12]. Much of the experimental work described in [4–6] has been concerned with uniaxial extension in the presence of time dependent temperature fields. One of the interesting and unexpected results of the experimental investigation in [4,5] was that chemical reactions associated with scission and subsequent crosslinking appeared to be independent of stretch during uniaxial extension up to stretch ratios of 3–4. This result, which is in agreement with earlier work by Tobolsky [13] and Scanlan and Watson [14], was used to simplify the formulation in several of the applications [7–12].

In separate ongoing work, multiaxial experiments are being performed in which an initially flat membrane is clamped along a circular boundary and then inflated by pressure over one surface at high temperatures. A facility for carrying out such experiments has been described in [6].

The inflation of a circular membrane by lateral pressure has been important in the development of constitutive equations in nonlinear elasticity. It is an experiment that normally produces a readily measurable axisymmetric non-homogenous deformation. The deformation can be regarded as an assemblage of unequal isochoric biaxial extension states ranging from equal biaxial extension at the membrane crown to pure shear at the supported boundary. Treloar [15] carried out such membrane inflation experiments and measured the deformed profiles and stretch ratio distributions in a vulcanized latex rubber membrane at different levels of inflation. Adkins and Rivlin [16] used the theory of nonlinear elastic membranes to calculate the inflated profiles for several forms of the strain energy function and compared the results with the data provided by Treloar. Studies with other forms of the strain energy function were carried out by Klingbeil and Shield [17] and Hart-Smith and Crisp [18]. Wineman, Wilson and Melvin [19] showed how the measured profiles and stretch ratio distributions can be used as part of a material identification method to determine the form of the strain energy function. Recently, the inflation of a circular membrane was used to determine properties of elastomers by Przybylo and Arruda [20] and of several polymeric materials by Li et al. [21].

Motivated by the importance of the membrane inflation experiment in isothermal nonlinear elasticity, it is natural to consider its use when the membrane material undergoes microstructural changes at elevated temperatures. Thus, the purpose here is to develop a membrane inflation model for use in conjunction with the experiment described in [6]. The constitutive equation for an elastomer undergoing scission and crosslinking at elevated temperatures is outlined in Section 2. The boundary value problem for the inflation of a circular membrane is formulated in Section 3. Section 4 contains a study of an inflated elastic membrane before scission. The thermal and inflation histories are specified in Section 5. Using the assumption that scission is only temperature dependent, not deformation dependent, two tests of experimental results are derived that can be used to determine when this assumption is valid. Section 6 addresses the response of the membrane after the temperature returns to its initial value, at which time scission has ceased and the membrane is deflated. It is shown that there is a deflated state when compressive stresses arise as a result of the constraint at the clamped support, which implies that the membrane will wrinkle on further deflation. It is then shown that if the constraint at the support is removed and the membrane is completely unloaded, the stresses reduce to zero and wrinkling disappears. The initially planar membrane then has a permanently curved axisymmetric shape. Conclusions are presented in Section 7.

2. Constitutive equation

Tobolsky [13] discussed experiments on rubber strips at elevated temperatures that led to the conclusion that there had been chemical changes in the macromolecular structure of the rubber. In these experiments a natural rubber strip at a relatively low temperature, say 20 °C, was subjected to a fixed uniaxial stretch

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