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# On the nonlinear viscoelastic behavior of rubber-like materials: Constitutive description and identification



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#### ABSTRACT

The main concern of this paper is the development of a three dimensional viscoelastic model at finite strain to describe nonfactorizable behavior of rubber-like materials. The model is developed within the framework of rational thermodynamics and internal state variable approach such that the second law of thermodynamics in the form of Clausius–Duhem inequality is satisfied. The nonfactorizable aspect of the behavior is introduced via a strain dependent relaxation times. The model is applied to describe the response of the isotropic Pipkin multi-integral viscoelastic model and the Bromobutyl (BIIR) material, several parameters involved are then identified using quasi-static and dynamic experiments thanks to a least-square minimization procedure. The proposed model is able to reproduce quasi-static response and show a good ability to predict the dynamic response of nonfactorizable rubber-like materials (BIIR) and the multi-integral model of Pipkin in a wide range of strain.

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#### 1. Introduction

It is well known that rubber-like materials exhibit nonlinear viscoelastic behavior over a wide range of strain and strain rates confronted in several engineering applications such as civil engineering, automotive and aerospace industries. This is due to their capacity to undergo high strain and strain rates without exceeding the elastic range of behavior. Further, the time dependent properties of these materials, such as shear relaxation modulus and creep compliance, are, in general, functions of the history of the strain or the stress [1]. Therefore, in a wide range of strain, a linear viscoelasticity theory is no longer applicable for such material and new constitutive equations are required to fully depict the behavior of rubber-like materials for quasi-static and dynamic configurations of huge interest in engineering applications.

The study of viscoelastic behavior of solid materials has a long history and several models have been developed from purely mathematical approaches to applied studies where ease of application is for huge interest. Two main approaches were followed in the development of nonlinear viscoelastic models, which are: the multi-integral approach which was firstly introduced by Volterra (see [2] and references therein) and the internal variable approach. For a general understanding of different approaches in viscoelasticity the reader is directed to the review arti-

cle by Wineman [3]. A significant class of models have been developed following the internal variable approach which consists on a generalization to a three dimensional model of the one dimensional Maxwell model which was firstly suggested by Schapery [4] and followed by the authors in [5–7] and [8] among others. The advantage of these models is their simplicity to be implemented into Finite element industrial software and applied to engineering application such as the work by Ansari and Hassanzadeh-Aghdam [9]. Other contributions to this approach used the fractional derivatives from the Maxwell model to obtain a fractional representation of the constitutive equations, see [10] and [11] among others.

Furthermore, from a phenomenological point of view several models have been developed to describe the nonfactorizable behavior of rubber-like materials, namely the Solid-Liquid viscoelastic model in the series of papers by the authors in [12,13] and [14] for which a generalized measure of deformation has replaced the strain tensor in the linear Boltzmann convolution integral model and the nonlinear viscoelastic model by Schapery [4] in which the creep compliance and the shear relaxation functions were considered stress-dependent and strain-dependent functions, respectively, and the model of Valanis [15] in which a total thermodynamic formulation led to a constitutive equation depending on the deformation via a deformation shift function in analogy with the so-called thermorheoligically simple materials. In the other hand, other

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models based on the microstructure of the polymeric chain have been proposed such as the model by Knauss and Emri [16] in which, following polymer science, time dependent functions were dependent upon the volumetric strain via a strain shift function and the model by Caruthers et al. [17] in which the strain shift function was expressed in terms of the *configurational energy* of the molecular structure.

In this work we shall develop a nonlinear viscoelastic model at finite strain within the framework of rational thermodynamics and the approach of internal state variables, the model is derived through a modification to approaches in [7], [8] and [6] taking into account the dependence of the time dependent functions upon the state of the strain. The model's parameters are then identified using data generated from the multi-integral viscoelastic model of Pipkin [18] and experimental data for bromobutyl (BIIR) from [19] in simple extension and validated using monotonic tests of pure shear.

This paper is organized as follow: in Section 2, a one dimensional nonlinear viscoelastic model is developed using a modified Maxwell rheological model. In the Section 3, this model is extended to the fully nonlinear formulation using a nonlinear set of evolution equation of the internal state variables within the rational thermodynamic framework. The shear relaxation modulus is set to be a function upon the invariants of the right Cauchy-Green strain tensor via a strain shift function analogous to the temperature shift function for the thermorheologically simple materials, this choice is motivated experimentally following the experimental characterization of BIIR from [19]. The constitutive equation for the stress is then obtained by resolving the set of nonlinear evolution equations. In Section 4, a systematic identification procedure of several parameters involved in the model is highlighted. The optimization problems arising from this identification procedure are solved by a modified least square minimization algorithm with Matlab software. Sections 5 and 6 are devoted to the results of this identification procedure using a theoretical data using the Pipkin model [18] and an experimental characterization of the Bromobutyl BIIR from [19], respectively. The capacity of the model to describe the behavior of the material is then outlined.

#### 2. Experimental and rheological motivations

In this section, we develop the rheological and experimental arguments leading to the proposed finite strain viscoelastic model. To motivate the three dimensional model developed below, we first highlight some experimental results leading to this model and then we consider a suitable modification to the generalized Maxwell rheological model to build the one dimensional nonlinear viscoelastic model.

#### 2.1. Experimental motivation

A significant class of rubbers shows nonfactorizable behavior at low and average range of strain. This phenomenon consists on the dependence of the shear relaxation modulus upon strain level. Several works were dedicated to deal with this class of behavior especially the series of papers by Sullivan [14] and O'connell and McKenna [20]. In a recent work [19], an experimental characterization was carried out with three rubber-like materials: the natural rubber (NR), the Bromobutyl (BIIR) and a mixture of these materials (NR-BIIR). Samples of the three materials were subjected to monotonic experiments of simple extension and pure shear with a relaxation of 10 minutes every 50% of strain in order to depict the equilibrium behavior of the materials. Moreover, a dynamic characterization was carried out in simple shear for a wide window of frequency at several temperatures and predeformations in order to construct the master curve of the material. This material showed a dependence of the shear relaxation modulus upon strain. In Fig. 1 it is plotted the logarithm of the shear relaxation modulus G(t) versus the logarithm of time for two different level of strain 10% and 50% for BIIR material. The shear relaxation modulus shows a dependence upon the strain level which leads according to Tschoegl et al. [21] to a shift in the

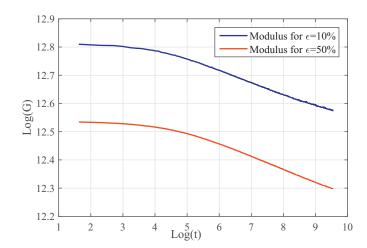


Fig. 1. Dependence of the shear relaxation modulus upon strain for BIIR rubber.

time with a strain dependent function since the shear relaxation modulus at any level could be obtained through a combination of a vertical and horizontal translation from the reference curve at a strain level of 10%. Therefore, a one dimensional viscoelastic model, taking in consideration these results, is developed in the next section through a generalization of the Maxwell rheological model.

#### 2.2. Rheological motivation

Before we develop the three-dimensional viscoelastic model, we shall investigate the following formulation for a standard linear solid. In this model,  $\sigma$  denotes the total stress,  $\varepsilon$  denotes the total strain,  $G_i$  and  $\tau_i$  are the parameters of the Maxwell model. Unlike the rheological model used in [22], the relaxation times  $\tau_i$  are, due to the experimental result outlined above, functions of the total strain  $\varepsilon$ . Furthermore, the stress in the spring of each Maxwell branch is denoted by  $Q_i$  and it is governed by the following evolution equation.

$$\dot{Q}_i + \frac{1}{\tau_i(\varepsilon)} Q_i = \frac{1}{\tau_i(\varepsilon)} G_i \varepsilon, \quad Q_i \big|_{t=0} = 0.$$
 (1)

The total stress  $\sigma$  derive directly from the rheological model as the difference between the elastic equilibrium stress and the non-equilibrium stresses  $Q_i$ .

$$\sigma = G_{\infty} \varepsilon - \sum_{i} Q_{i}. \tag{2}$$

The time parameters of the Maxwell model are set to be a strain dependent function; this idea follows from the description of thermorheologically simple materials behavior see [21] and [23], for which all parameters are temperature dependent via a single variable function called *temperature shift-function*. [5,24] and [20] among others generalized this notion to describe thermorheologically complex materials behavior where the shift function depend upon temperature and stress or strain. Other contributions modeled this phenomena by a strain-rate dependent relaxation times, see [25] and references therein. In our work, since the study was carried out using relaxation data, the time parameters take the following form.

$$\tau_i(\varepsilon) = a(\varepsilon)\tau_i,\tag{3}$$

 $a(\epsilon)$  is a positive strain function, following the dissipation inequality, called strain shift function. Therefore, the law of evolution of Eq. (1) became a linear differential equation over the reduced time  $\xi$ , after considering the form of the time parameters of Eq. (3).

$$\frac{dQ_i}{d\xi} + \frac{1}{\tau_i}Q_i = \frac{1}{\tau_i}G_i \ \varepsilon \quad \text{with} \quad \xi(t) = \int_0^t \frac{dt'}{a(\varepsilon)}, \tag{4}$$

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