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# A finite viscoelastic constitutive model for filled rubber-like materials

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#### abstract

Filled rubber-like materials show a significant dependence on predeformation, frequency and amplitude when they are loaded with static predeformations superimposed by harmonic deformations. In order to investigate this predeformation-, frequency- and amplitude-dependent material behavior, a recently developed constitutive model of finite viscoelasticity is proposed based on the models of Lion and Kardelky (2004) and Hofer and Lion (2009). The constitutive equations are geometrically linearized in the neighborhood of predeformation and then formulated for incompressible materials. Finally, the process of parameter identification and some numerical aspects are shown. Additionally, the experimental verification and some numerical simulations will also be supplied. The results show that our model cannot only be used for loading cases with small strains but also with large strains. It may have some meanings for practical engineering applications.

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#### 1. Introduction

Because of superiorly elastic and damping properties, filled rubber-like materials have been widely used in industry and private life ([Dai et al., 2006; Omnès et al., 2008; Misra and Yuan, 2012;](#page--1-0) [Dezfuli and Alam, 2013; Siqueira et al., 2014; Spizzuoco et al.,](#page--1-0) [2014\)](#page--1-0). The filler, such as carbon black and silica, enhances essential properties of polymers like stiffness, strength and abrasion resistance [\(Chiu et al., 2003; Kaleem et al., 2012; Vesenjaka et al.,](#page--1-0) [2012; Wang et al., 2013a,b; Wang and Han, 2013\)](#page--1-0). From numerous engineering applications, we know that there are many fields where constitutive models are required to precisely predict the dynamical mechanical responses of filled polymers under infinitesimal and finite thermo-mechanical deformations ([Williams, 1963; Tormey and Britton, 1963; Reggi and](#page--1-0) [Woytowitz, 1993; Hu and Zhu, 2011; Spizzuoco et al., 2014](#page--1-0)).

As is known to us all, the mechanical behavior of filled rubberlike materials is mainly hyperelastic ([Reese and Wriggers, 1997;](#page--1-0) [Boyce and Arruda, 2000; Miehe et al., 2004; Ogden, 2005; Miehe](#page--1-0) [and Goktepe, 2005; Goktepe and Miehe, 2005; DeBotton et al.,](#page--1-0) [2006; Pamiesa and Castaneda, 2006a,b; Li et al., 2012; Hao et al.,](#page--1-0) [2015a,b\)](#page--1-0) but, more or less significant, a lot of inelastic phenomena were observed [\(Weia et al., 2004; Garc](#page--1-0)ı[aTarrago et al., 2007; Asta](#page--1-0) [and Ragni, 2008; Machado et al., 2010; Yu et al., 2012; Wang and](#page--1-0) [Han, 2013](#page--1-0)). If a virgin specimen made of filler-reinforced rubber be observed ([Mullins, 1948, 1969; Mullins and Tobin, 1957](#page--1-0)). It is a softening phenomenon which is caused by the strain-induced breakage of weak physical bonds. Experimental data and constitutive models expressing the Mullins effect have been published ([Machado et al., 2010, 2012; Rebouah et al., 2013](#page--1-0) as example). The Payne effect, which was found out by [Payne \(1961\)](#page--1-0) and seems very similar to the Mullins effect, has also been observed by many researchers [\(Lion and Kardelky, 2004; Hofer and Lion, 2009;](#page--1-0) [Rendek and Lion, 2010a,b; Blom and Kari, 2011; Netzker et al.,](#page--1-0) [2010; Austrell and Olsson, 2012; Österlöf et al., 2014\)](#page--1-0). The both two phenomena are strain-induced nonlinearities, and the Payne effect can recover in a very short time of several seconds or minutes while the Mullins effect is usually irreversible at room temperature. Since this paper focuses on the Payne effect, any non-recurrent or irreversible stress-softening phenomena are not taken into account. For further insight into modeling investigations of the Payne effect, the reader is referred to [Lin and Schomburg](#page--1-0) [\(2003\), Lion and Kardelky \(2004\), Luoa et al. \(2010\), Drozdov](#page--1-0) [\(2007a,b\), Mendiguren et al. \(2012\), Pascon and Coda \(2013\)](#page--1-0) and [Luo et al. \(2013\)](#page--1-0) and so on.

is cyclically excited with large amplitudes, the Mullins effect can

As we are also aware, the mechanical behavior of filled rubberlike materials under finite deformations is highly dependent on frequency, deformation rate and predeformation ([Liak and](#page--1-0) [Negami, 1966; Enelund and Josefson, 1997; Alvarez et al., 1991;](#page--1-0) [Miiller et al., 1995; Havriliak, 1996; Pan, 1996; Busfield et al.,](#page--1-0) [2000; Dickens, 2000; Metzler and Nonnenmacher, 2003;](#page--1-0) [Ramrakhyani et al., 2004; Shaska, 2005; Setua et al., 2006; Jrad](#page--1-0)





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[et al., 2013; Nimpaiboon et al., 2013; Diani et al., 2013; Arikoglu,](#page--1-0) [2014; Hao et al., 2015a,b](#page--1-0)). In the scope of continuum mechanics, a wide basis of comprehensive and profound models for the description of rubber-like materials have been developed. Frequently, linear or finite viscoelastic formulations are derived and applied. Experimental data and constitutive theories to describe such behavior have been proposed ([Pan, 1996; Yang](#page--1-0) [et al., 2000; Lin and Schomburg, 2003; Reese, 2003; Amin et al.,](#page--1-0) [2006; Gil-Negrete et al., 2006; Garc](#page--1-0)ı[aTarrago et al., 2007; Rendek](#page--1-0) [and Lion \(2010a,b\), Blom and Kari, 2011, 2012, Ooi and Ripin,](#page--1-0) [2011, Austrell and Olsson, 2012, Wollscheid and Lion, 2013;](#page--1-0) [Lindberg et al., 2014; Österlöf et al., 2014\)](#page--1-0).

Since the filled rubber-like materials are nonlinear viscoelastic materials, their dynamic properties are also temperaturedependent [\(Hou and Chen, 2012\)](#page--1-0). In this article, the temperature-dependence is neither researched nor taken into account in the constitutive model.

In the present work, based on [Lion and Kardelky \(2004\)\) and](#page--1-0) [Hofer and Lion \(2009\)](#page--1-0) models, a developed finite viscoelastic constitutive model expressing the predeformation-, frequency- and amplitude-dependence shall be formulated. On the above basis, common modeling ideas will be supplied. Starting with a linear and one-dimensional motivation of the constitutive approach, an appropriate model of finite nonlinear viscoelasticity will be formulated. In contrast with the most common approach which uses general Maxwell elements and bases on a multiplicative decomposition of the deformation gradient into elastic and inelastic parts, our model employs the fractional Maxwell elements and is a functional formulation without any decomposition of the deformation gradient. For the purpose of taking the amplitudeinduced nonlinearities into account, the constitutive model is formulated with intrinsic time scales which are equivalent to variable viscosities. These intrinsic time scales are driven by the internal state variables, which can be considered as phenomenological measures of the current state for the filled polymers. By choosing adequate fractional Maxwell elements (or evolution equations for the internal state variables), the influence of the Payne effect can be incorporated into the formulation.

In order to obtain closed-form solutions for the storage and loss moduli, the proposed model has to be linearized with respect to small dynamic excitation around a static predeformation. On the basis of these expressions, a set of material parameters will be identified to describe the rubber-like material's mechanical behavior. Then, some numerical aspects of the proposed model and some simulation results obtained by the Matlab package will also be given in the present paper. Our paper will finish with some conclusions.

#### 2. The proposed constitutive model

The main focus of this section will be the formulation of a constitutive theory to describe the predeformation-, frequency- and amplitude-dependence of mechanical behaviors for rubber-like materials. Starting with a one-dimensional illustration of the basic method of the model and some fundamentals of nonlinear continuum mechanics, a theory of nonlinear finite viscoelastic behaviors will be represented. In contrast with the common approach of the multiplicative finite viscoelastic property, the present approach is based on a functional representation of the overstress without need of a viscous intermediate configuration. Although the multiplicative viscoelastic behavior is capable of describing the frequency- and amplitude-dependence ([Pascon and Coda, 2013\)](#page--1-0), in consideration of an easy-to-use with fewer model constants and a simple-to-realize with good-natured parameter identifications, the presented approach of a functional theory of nonlinear finite viscoelastic property is preferred.

It is important to mention that our formulation is quite different from the multiplicative approach, where the Helmholtz free energy is formulated based on the elastic part of the deformation gradient. Here, the constitutive equations base on differential equations of the fractional order using nonlinear stress and strain measures. Due to the incorporation of structural-dependent viscosity, these differential equations are linear with respect to intrinsic time scales. But with respect to the real time, these differential equations are nonlinear. In this paper, the model is investigated to describe the predeformation-, frequency- and amplitudedependent phenomena of the rubber-like materials around a certain working point. The object of our work is to develop a time domain formulation of the nonlinear viscoelastic behavior which represents predeformation-, frequency- and amplitude-dependent properties and can be used to simulate the mechanical behavior of rubber-like materials.

#### 2.1. One-dimensional modeling approach

In order to describe the frequency-dependent behavior of rubber-like materials, it is quite common to use a set of Maxwell elements [\(Zhang and Richards, 2007; Renaud et al., 2011; Zhao et al.,](#page--1-0) [2014](#page--1-0)), which can be described by the linear differential equation of first order

$$
\begin{cases}\n\sigma = E_0 \varepsilon + \sum_{k=1}^{M} \sigma_{ovk} \\
\dot{\sigma}_{ovk} = E_k \dot{\varepsilon} - \frac{1}{z_{0k}} \sigma_{ovk}\n\end{cases}
$$
\n(1)

where  $E_k$  and  $z_{0k}$  are the model parameters of the overstress part. In order to describe the frequency-dependent behavior with a smaller number of model parameters, we replace the first order of time derivative in Eq. (1) with the fractional order of time derivative which has been adopted by many authors ([Lewandowski and](#page--1-0) [Chorazyczewski, 2010; Hou and Chen, 2012](#page--1-0)). Therefore, Eq. (1) becomes

$$
\begin{cases}\n\sigma = E_0 \varepsilon + \sum_{k=1}^{M} \sigma_{ovk} \\
D^{\alpha_k} \sigma_{ovk} = E_k D^{\alpha_k} \varepsilon - \frac{1}{2\alpha_k} \sigma_{ovk} \\
\end{cases} , \quad \alpha_k \in (0, 1]
$$
\n(2)

where  $\alpha_k$  is the order of the fractional derivative. The unit of  $z_{0k}$  is still second. The fractional derivative operator  $D^{\alpha_k}$  is defined by [Shahsavari and Ulm \(2009\)](#page--1-0)

$$
D^{\alpha_k}f(t) = \frac{1}{\Gamma(1-\alpha_k)}\frac{d}{dt}\int_0^t \frac{f(\kappa)}{(t-\kappa)^{\alpha_k}}d\kappa\tag{3}
$$

where  $\Gamma(x)$  is the Gamma function defined by

$$
\Gamma(x) = \int_0^\infty t^{x-1} e^{-t} dt, \quad \text{Re}(x) > 0 \tag{4}
$$

In addition, when  $\dot{\varepsilon} \equiv 0$ ,  $\sigma_{ovk} \equiv 0$ . Thus, Eq. (2)<sub>1</sub> becomes

$$
\sigma = E_0 \varepsilon \tag{5}
$$

Eq. (5) implies that when the deformation is static or quasistatic, Eq. (2) describes the linear elastic behavior. This may be correct for small deformation cases, while there is obvious nonlinear elastic behavior (hyperelasticity) for large deformation cases. Therefore, the item of  $E_0 \varepsilon$  in Eq. (2) should be replaced by a hyperelastic model to represent the nonlinear elastic behavior. In this paper, a Helmholtz free energy function is adopted. The Helmholtz free energy function in this paper is denoted as  $\psi_{\text{eq}}$ , which also expresses the equilibrium part. Then Eq.  $(2)$  is rewritten as

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