On the modelling of amplitude and frequency-dependent properties in rubberlike solids

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ABSTRACT: In these days the development of vehicles is partly based on numerical simulations using computer programs developed to simulate the dynamics of multi-body systems. Constitutive models in combination with finite element implementations are required for the prediction of the mechanical behaviour of, e.g., rubberlike solids within vehicle components, which then provide a basis for simulations in multi-body systems. One aim of the present communication is to deal with the experimental investigation of the amplitude and frequency-dependent mechanical properties in rubberlike solids at constant temperature. Quasi-static uniaxial, equibiaxial and simple shear tests on natural rubber compounds were carried out in order to analyze their mechanical responses including the typical stress softening effect. In addition, cyclic uniaxial tension tests were performed for the determination of dynamical properties. Another aim of this communication is to summarize a constitutive model which captures the typical mechanical phenomena observed during the experiment investigation. A method for parameter identification is briefly described and a set of numerical data presented.

1 INTRODUCTION

Dynamic simulations of vehicles during the design process by use of, for example, multi-body systems, are becoming more and more important. Such simulations require reliable constitutive descriptors of rubber-metal components, which have numerous applications in the vehicle industry. Based on comprehensive material test data for the rubber, the development of constitutive models, when incorporated into the finite element method, provides a powerful tool for predicting the nonlinear mechanical behaviour of the rubber within vehicle components. This avoids the need for extensive experimental testing of individual components. However, in order to obtain reliable constitutive models, appropriate static and dynamic experimental investigations on rubber-metal components are unavoidable for providing a sound basis for the development of so-called “force elements” that are used frequently in multi-body systems.

In this investigation we restrict our attention to isothermal processes, although in a more general situation thermo-mechanical heating effects should be considered, as in, e.g., Miehe (1995) or Holzapfel & Simo (1996a) amongst others. In this paper we provide a brief overview of the testing and modeling of rubberlike solids, then we describe novel experimental data and summarize a model that allows the simulation of amplitude and frequency-dependent material properties in rubberlike solids under different levels of temperature and static prestrain.

2 OVERVIEW – STATE-OF-THE-ART

2.1 Experiments

From the historical point of view the first experimental investigation on rubber seems to have been performed by Joule (1859). Experimental results on the nonlinear elastic behaviour of elastomers were published in the early work of Treloar (1944). Treloar’s data are still widely used in research and development. The dependence of the mechanical properties (e.g., stiffness changes) on the magnitude of the amplitudes, or more precisely on the maximal strain energy, to which the material has been subjected under static loading conditions was analyzed by Mullins (1947), and is known as the Mullins effect. In the case of dynamic loading the storage and loss moduli depend on the amplitude. The greater the amplitude the lower is the storage modulus, while the loss modulus exhibits a maximum at intermediate amplitude levels. It is the so-called Fletcher-Gent or Payne effect (see, e.g., Payne 1967,
and references therein). The difference between the (static) Mullins and the (dynamic) Fletcher-Gent effects is that the latter can be recovered but the former cannot. It is remarkable that investigators have succeeded in at least recovering partially the Mullins effect by means of sufficient heat treatment over a long time period. Sjöberg & Kari (2003) have tested the influence of simultaneously applied sinusoidal or noise excitation signals on a vibration isolator. As a rule of thumb they concluded that the reference stiffness measured at a small (excitation) amplitude and high frequency is reduced, and damping is increased when the initial excitation is superimposed on a noise excitation with a large amplitude and a low frequency. For the evaluation of dynamic material tests at different temperatures the assumption of thermo-rheologically simple material behaviour is commonly used. Hence, the viscoelastic characteristics (such as relaxation function or storage modulus) can be shifted horizontally along a logarithmic time or frequency axis in order to compose a single master curve. The principle of time-temperature superposition was first observed by Leadermann (1943), but it does not hold for all elastomers.

2.2 Constitutive models for rubber

Constitutive models may be phenomenologically motivated, as with, e.g., the Mooney-Rivlin model, which is frequently used for industrial applications, or micromechanically motivated, as in the case of the “flexible” Van der Waals model developed by Kilian (1981), who discusses the analogy between rubber and gases using only four parameters. The Arruda & Boyce (1993) model has a physical basis, in which the individual polymer chains are described by a non-Gaussian statistical theory. Moreover, the Arruda-Boyce model needs only the identification of two parameters. For other free-energy functions we refer to the books by, e.g., Holzapfel (2000) and Saccomandi & Ogden (2004).

The Mullins effect is usually regarded as a damage effect in the material since its recovery time is very long, if indeed there is recovery at all. The first model of this phenomenon goes back to Kachanov (1958), who used two damage variables: a continuous damage variable that describes in effect the Mullins effect, and a so-called continuous damage variable, which is used to simulate fatigue behaviour. A pseudo-elastic approach that captures the Mullins effect was developed by Ogden & Roxburgh (1999) and extended by Dorfmann & Ogden (2004) to account for permanent set.

Equilibrium hysteresis, which is observed during static cyclic testing, can also be modelled by viscoelastic or elastoplastic rheological models (see, e.g., Holzapfel, 1996b, Kaliske & Rothert, 1998, Reese & Govindjee, 1998, Simo & Hughes, 1998). Furthermore, the model of Chaboche et al. (1979), which was originally developed for the simulation of the kinematic hardening of steel, may also be able to reproduce the equilibrium hysteresis of elastomers.

2.3 Models capturing amplitude-dependent properties

2.3.1 Kraus model and pseudo-elastic models

The so-called Kraus equations provide the storage module $E'$ and the loss module $E''$ as a function of the strain amplitude $\Delta \varepsilon$, i.e.

$$E' = E'_\infty + \frac{C_1}{1 + \left(\frac{\Delta \varepsilon}{\Delta \varepsilon_c}\right)^{2m}}, \quad E'' = E''_\infty + \frac{C_2 \left(\frac{\Delta \varepsilon}{\Delta \varepsilon_c}\right)^m}{1 + \left(\frac{\Delta \varepsilon}{\Delta \varepsilon_c}\right)^{2m}}$$

(1)

where $E'_\infty$, $E''_\infty$, $C_1$, $C_2$, $\Delta \varepsilon_c$ and $m$ are material parameters to be identified. These parameters may be physically interpreted. The relations (1) have the drawback that they do not describe frequency and temperature effects, and the model is not applicable to three-dimensional transient simulations.

Pseudo-elastic models are documented by, e.g., Ogden & Roxburgh (1999) and Saccomandi & Ogden (2004). Essentially the so-called secondary curves follow different paths during loading and unloading, i.e. different hyperelastic materials are assigned for loading and unloading. In addition two more functions are defined for the cases of reloading and any subsequent unloading between the two secondary curves. These models do not consider any rate dependence.

2.3.2 Elastoplastic models

The generalized Prandtl model or other elastoplastic models (e.g., that of Chaboche et al. 1979) are able to simulate the amplitude-dependent behaviour of rubberlike materials, although in a strain-rate independent manner. Hence, elasto-viscoplasticity should be used, as has been proposed by Kämmle (1986), who has extended the generalized Maxwell model with Coulomb friction elements (ideal plastic elements).

The so-called Rho-model was suggested by Lambertz (1994), and refined and extended to the three-dimensional regime by Stommel (1999). The essence of the model is the application of non-Coulomb friction elements instead of numerous Coulomb elements to the generalized Maxwell model.

2.3.3 Models based on structural variables

Lion (1998, 1999) has introduced structural variables for the modelling of amplitude-dependent and thixotropic material behaviours of rubberlike solids. In this case the viscosities depend on the temperature and the deformation history. The deformation
history and time dependences (i.e. the thixotropic effects) are taken into account by structural variables, which are internal variables used as measures of the amplitudes. This model is discussed in more detail in Section 4.

2.4 Numerical modelling

Hyperelastic material models have been available for a long time and many have been implemented in commercial finite-element (FE) software. These include the models of Ogden (1972, 1997), Holzapfel (1996b), Bergström & Boyce (1998), Ogden & Roxburgh (1999), etc. For the computation of combined finite elastic-viscoelastic-plastoelastic stress and damage it is worth of mentioning the work by Miehe & Keck (2000). These authors have also tested the Rho-model and the thixotropy model with their own FE software.

3 EXPERIMENTAL TESTS

3.1 Manufacturing of specimens

In our study the material of a rubber-metal vehicle part was analyzed. There are two ways to obtain specimens for material testing: (i) cutting directly from the vehicle part, or (ii) vulcanization from the raw material. The disadvantage of (i) is that the form and size of the specimen are strongly limited by the available vehicle part. In addition, cutting may change the material properties of the specimen (by inducing residual stresses). The main difficulty of method (ii) is, however, in the assessment of the vulcanization parameters, and ensuring the same material properties for the specimen and the vehicle part. For our study we have chosen procedure (ii). We used a carbon-black-filled natural rubber with Shore hardness 60, which is frequently used in railway vehicle bogies. Pressure and temperature data for the vulcanization and the raw compound were provided by the manufacturer. Vulcanization times for the different specimens were determined (~7 minutes) by preliminary testing on a vulcameter machine. Three types of specimens were produced: so-called “dog-bone” specimens for uniaxial tension-compression tests (see Fig. 1), cylindrical specimens for shear-sandwich tests (diameter = 19.71 mm, height = 5.28 mm), and square plates for equibiaxial tests (edge length = 164.1 mm, thickness = 2.67 mm).

3.2 Protocol for material testing

All our tests were carried out under strain (or displacement) controlled excitation. Before the experiments were actually performed all specimens were thermally pre-conditioned at room temperature of 20 ± 2°C for the duration of at least 12 hours. Mechanical pre-conditioning was then performed at room temperature, except for the specimens prepared for studying the Mullins effect. The mechanical pre-conditioning consisted of a cyclic phase with 12 cycles followed by a load-free relaxation phase of at least 10 minutes duration. The excitations during the cyclic part were triangle strain signals and the maximal strain rate was 1/60 s\(^{-1}\). The Mullins effect was tested for three deformation modes (uniaxial compression, uniaxial tension and equibiaxial tension) at room temperature (20 ± 2°C); see Fig. 2.

Table 1. Deformation ranges during quasi-static material tests.

<table>
<thead>
<tr>
<th>Mode of deformation</th>
<th>minimal strain*</th>
<th>maximal strain*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uniaxial compression</td>
<td>-0.3</td>
<td>0</td>
</tr>
<tr>
<td>Uniaxial tension</td>
<td>0</td>
<td>1.0</td>
</tr>
<tr>
<td>Equibiaxial tension</td>
<td>0</td>
<td>0.72</td>
</tr>
<tr>
<td>Simple shear**</td>
<td>-0.7</td>
<td>0.7</td>
</tr>
</tbody>
</table>

* Engineering strains, which refer to the load-free reference configuration.

** Given values are tangents of the shear angle.

Dynamic tests on dog-bone specimens were carried out with all combinations of the parameters listed in Table 2. Experiments were performed with...
harmonic excitation to determine the storage module $E'$ and the loss module $E''$ using the Fourier method.

First, the pre-conditioned specimen was cooled down to the lowest temperature ($-30^\circ$C), then, after an equilibration time of 30 minutes, it was deformed in compression up to $-20\%$ engineering strain. This was done with a speed of $500$ mm/s. Subsequently the specimen relaxed for 10 minutes. For verification the force-displacement curves during the relaxation were recorded. The dynamic excitation started with the smallest (engineering) strain amplitude. The frequency was varied from the smallest to the highest value. After the frequency variation the static pre-strain remained constant, and the amplitude was increased to the next higher value. The frequency sweep was carried out again, and this procedure was repeated until the highest amplitude was reached. The static pre-strain was then set to the next level, and the specimen was again allowed to relax for 10 minutes. The test continued in a similar way until the highest pre-strain level in tension had been reached. Subsequently, the temperature was increased with a heat up rate of $5-20^\circ$C/min to the next higher level, and, as for the first temperature, the whole procedure was repeated.

The model used is based on the three-dimensional finite strain viscoelasticity formulation in Holzapfel (1996b, 2000), and on the thixotropy model of Lion (1999). We use the well-established volumetric-isochoric split of the deformation gradient $F$, i.e.

$$F = \left(J^{1/3}\right)\bar{F}, \quad \det(\bar{F}) = 1, \quad \bar{C} = \left(J^{2/3}\right)\bar{C}, \quad \bar{C} = \bar{F}^T \bar{F},$$ (2)

where $\bar{C} = F^T F$ is the right Cauchy-Green tensor, $\bar{C}$ its modification defined in (2), $I$ the identity tensor, and $J$ the volume ratio. To characterize viscoelastic processes we use the isotropic constitutive model $\Psi(C, \theta, \Gamma_1, ..., \Gamma_m) = \Psi_{vol}^{\alpha}(J, \theta) + \Psi_{iso}^{\alpha}(\bar{C}, \theta) + \sum_{\alpha=1}^{m} \Psi_{iso, \alpha}^{\alpha}(\bar{C}, \theta, \Gamma_\alpha(\delta)),$ (3)

where $\theta$ is the temperature, $\Gamma_\alpha$ are strain-like internal (tensor) variables, $\delta$ is a scalar measure of the strain amplitude, $\Psi_{vol}^{\alpha}$ and $\Psi_{iso}^{\alpha}$ are free energy functions per unit reference volume describing, respectively, the (elastic) volumetric and isochoric responses, and $\Psi_{iso, \alpha}$ are scalar-valued functions characterizing the non-equilibrium state. Next we introduce the assumption

$$\Psi_{iso, \alpha} = \beta_{\alpha} \Psi_{iso}^{\alpha},$$ (4)

where $\beta_{\alpha} \in [0, \infty)$ are given non-dimensional free-energy factors, and $\Psi_{iso}^{\alpha}$ is a function characterizing the non-equilibrium state. Now, the algorithmic update formula for the second Piola-Kirchhoff stress tensor is given by

$$S_{\alpha+1} = \left(S_{vol}^{\alpha} + S_{iso}^{\alpha} + \sum_{\alpha=1}^{m} Q_{\alpha}(\delta)\right)_{\alpha+1},$$ (5)

where $S_{vol}^{\alpha}, S_{iso}^{\alpha}$ are the volumetric and isochoric parts of the (equilibrium) second Piola-Kirchhoff stress, respectively, while $Q_{\alpha}$ are stress-like internal variables characterizing the non-equilibrium stresses. Omitting the derivation we obtain the second-order accurate recurrence update formula

$$Q_{\alpha, n+1} = \frac{\Delta t}{\tau_{\alpha}} a(\delta, \theta) Q_{\alpha, n} +$$

$$\frac{1 - \exp(-\frac{\Delta t}{\tau_{\alpha}} a(\delta, \theta))}{\tau_{\alpha} a(\delta, \theta)} \left(\hat{S}_{iso, \alpha+1}^{\alpha} - \hat{S}_{iso, \alpha}^{\alpha}\right),$$ (6)

where $\Delta t$ is time increment, $\tau_{\alpha}$ is the relaxation time for the Maxwell element $a$, $a(\delta, \theta) = a_d(\delta) a_t(\theta)$, the product of scalar-valued functions for determining the effective relaxation times, more precisely the viscosities: $a_d(\delta)$ is an amplitude-frequency shift function; $a_t(\theta)$ is the well-known temperature-frequency shift function, and $\hat{S}_{iso, \alpha+1}^{\alpha}, \hat{S}_{iso, \alpha}^{\alpha}$ are isochoric second Piola-Kirchhoff stresses derived from the free-energy function. At this point we introduce a surface $\Phi = 0$ in the mechanical strain space according to

$$\Phi = \sqrt{2/3} \| \mathbf{E} \| - \varphi, \quad \| \mathbf{E} \| = \sqrt{\mathbf{E} : \mathbf{E}},$$ (7)

where $\varphi$ is a scalar measure of the maximum strain reached at the recent past, and $\mathbf{E} = (\mathbf{C} - I)/2$ is the Green-Lagrange strain tensor. In order to specify the scalar amplitude measure $\delta$ we define a second surface $\Delta = 0$ according to

$$\Delta = \sqrt{2/3} \| \mathbf{E} \| - \varphi - \delta.$$

(8)

For the evolution equations for $\varphi$ and $\delta$ we refer to Lion (1999).

Table 2. Parameters for the dynamic tests on dog-bone specimens under uniaxial loading.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Levels</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature</td>
<td>°C</td>
<td>-30, 0, 30, 80</td>
</tr>
<tr>
<td>Pre-strain*</td>
<td>%</td>
<td>-20, -10, 0, 10, 20, 30, 50, 70, 90</td>
</tr>
<tr>
<td>Strain amplitude*</td>
<td>%</td>
<td>0.1, 0.5, 1, 3, 5, 10</td>
</tr>
<tr>
<td>Frequency</td>
<td>Hz</td>
<td>0.01, 0.1, 0.3, 1, 3, 10, 15, 20, 30</td>
</tr>
</tbody>
</table>

* Engineering strains, which refer to the load-free reference configuration

4 CONSTITUTIVE MODEL

4.1 Outline of the model

The model used is based on the three-dimensional finite strain viscoelasticity formulation in Holzapfel (1996b, 2000), and on the thixotropy model of Lion (1998). We use the well-established volumetric-isochoric split of the deformation gradient $F$, i.e.
4.2 Method for parameter identification

The parameters of the hyperelastic model may be determined on the basis of the quasi-static tests. The first step in the parameter identification of the dynamic tests was the computation of the horizontal shift of the storage modulus \( E' \) with respect to the temperature. Note that \( E' \) was measured in the frequency range 0.01–30 Hz at four temperatures, six amplitudes and at nine pre-strain levels. The measured \( E' \) values, for different temperatures but for the same strain amplitude and pre-strain level, were shifted along the frequency axis to obtain a more or less continuous curve (i.e. a master curve) so that the \( E' \) values at the reference temperature (here +30°C) remain unshifted; see Fig. 3.

As a first approximation we restrict the proposed method to the temperature range 0 to +80°C, because for this range the master curves have rather similar shapes. Subsequently, analogously to the temperature shift, a horizontal shift was performed with respect to the amplitude to obtain a single master curve for each pre-strain. For the reference strain amplitude we chose 0.5% because for this amplitude we have the most complete experimental data. Power-law type curves may then be fitted to the shifted test data; see Fig. 4.

The generalized Maxwell model can be linearized with respect to the region of the static pre-strain \( \varepsilon_{\text{pre}} \), and hence the storage modulus may be expressed as

\[
E'(\omega, \varepsilon_{\text{pre}}) = E^\infty(\varepsilon_{\text{pre}}) + \frac{\sum_{a=1}^{m} E_a(\varepsilon_{\text{pre}})(\omega \tau_a)^{2}}{1+(\omega \tau_a)^{2}},
\]

where \( E' \) is the storage modulus, \( E^\infty \) is the stiffness at the given level of pre-strain as \( t\to\infty \), \( \omega \) is the angular frequency, and \( E_a \) and \( \tau_a \), \( \alpha = 1, \ldots, m \), are the stiffness and relaxation time of the Maxwell element \( \alpha \). We assumed a fixed set of relaxation times \( \tau_a \) for each pre-strain level and identified the values \( E_a \).

Note that by analogy with (4) we may also write

\[
E_a(\varepsilon_{\text{pre}}) = \beta_d E^\infty(\varepsilon_{\text{pre}}).
\]

At this stage the scaling functions \( a_d(\theta) \) and \( a_f(\theta) \) can be determined by taking into account the shifts needed to obtain the master curves.

4.3 Material parameters

The isochoric part of the equilibrium stress was characterized by the Van der Waals model with the parameters \( \mu = 1.01199 \text{ N/mm}^2 \), \( \lambda_m = 3.7525 \), \( a = 0.72124 \), \( \beta = 0 \) (HKS 2000), which were identified by using uniaxial and equibiaxial tension test data. We used the power-law function \( E' = A_1 f^G + A_2 f^H \), with the frequency denoted by \( f \). For illustrative purposes we chose the pre-strain level of \(-10\%\) and obtained the parameters \( A_1 = 2.0061 \text{ N/mm}^2 \), \( A_2 = 2.0629 \text{ N/mm}^2 \), \( G = 0.009972 \), \( H = 0.066794 \). We assumed that at the other pre-strain levels only the parameters \( A_1, A_2 \) differ by a scalar factor. The amplitude shift function has the form \( a_{\varepsilon} = A_l(1+{B_\theta})^C \), with the parameters \( A = 967.8 \), \( B = 258.1 \), \( C = 4.4 \). As a temperature-frequency shift function \( a_T \) we used the WLF equation (see Stommel 1999) with the parameters \( c_1 = 8.1 \), \( c_2 = 104.2K \) and \( T_0 = 303K \). Figure 5 illustrates experimental data together with the related numerical results. In future work we shall analyze three-dimensional problems on the basis of the finite element method.

Figure 3. Experimental master curves for six different strain amplitudes \( \Delta \varepsilon_i \), \( i = 1, \ldots, 6 \), after the horizontal shift with respect to the temperature, where the reference temperature was chosen to be +30°C. The pre-strain level is \(-10\%\) (engineering strain).

Figure 4. Experimental data (circles) for the reference strain amplitude \( \Delta \varepsilon = 0.5\% \) after the temperature and amplitude shifting procedure at a reference temperature of +30°C, and a pre-strain level of \(-10\%\). The solid line indicates a fitted power-law curve.

Figure 5. Experimental data (circles) at temperature +30°C and at a pre-strain level of \(-10\%\) for six strain amplitudes (compare also with Fig. 3), and numerical results (solid lines).
5 CONCLUSION

The experimental investigations demonstrated that the amplitude and frequency dependence of the dynamic moduli are strongly influenced by the static pre-strain and temperature. The proposed model, formulated to capture isothermal, three-dimensional finite deformations, can predict the amplitude-dependent viscoelastic material behaviour, and enables thixotropic effects to be incorporated.

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7 REFERENCES


