VISCOELASTIC PROPERTIES OF FILLED RUBBER. EXPERIMENTAL OBSERVATIONS AND MATERIAL MODELLING

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The paper presents an application of a phenomenological material model for the viscoelastic stress response at large strains. The model is used for the simulation of carbon-black filled rubber in monotonic and cyclic deformation processes under isothermal conditions. The material stress response is decomposed into two constitutive parts which act in parallel: an elastic equilibrium stress response and a rate-dependent viscoelastic overstress response. The response of a particular filled rubber in the cyclic and relaxation tests was measured experimentally. The parameters of the constitutive functions are determined from the experimental data by an identification process employing nonlinear optimization methods. The paper concludes with a simulation by FEM of the cyclic loading of a simple rubber specimen.

Key words: *rubber behaviour, viscoelasticity, large strains, stress relaxation, cyclic loading*

1. Introduction

Rubber materials are applied in various branches of mechanical engineering because of their damping properties. One of such applications is the cushion of tram-wheels by rubber segments manufactured by Bonatrans Bohumin. During the operation the segments are under the temporally constant compressive preload due to the shrinkage between the corpus of wheel and the hoop and under the dynamic compressive and shear loads due to the transfer of the vehicle weight during the wheel rotation and the transfer of torque. The static preload leads to compressive permanent set of segment and the periodic deformation leads to hysteresis behaviour and heat generation which considerably affects properties of rubber. The modelling and FEM calculation of the structural response requires a constitutive model which captures the complex material behaviour. The present paper focuses on the viscoelastic behaviour of the filled rubber used for the cushion segments in cyclic and relaxation experiments.

2. Material behaviour and modelling

The experimentally observed mechanical response of a filled rubber may be subdivided into four fundamentally different effects [1] which together characterize the typical overall response:

– a dominating elastic ground-stress response characterized by large elastic strains,

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– a finite viscoelastic over-stress which governs rate-dependent effects such as relaxation and creep phenomena,
– finite plastoelastic over-stress behaviour responsible for rate- independent hysteresis phenomena associated with relaxed equilibrium states,
– a damage response within the first loading cycles which induces a considerable stress softening.

The ground-stress response is usually modelled in the phenomenological framework of finite elasticity by Mooney-Rivlin or Ogden models [2–5], or by Arruda and Boyce model [6] in terms of the micromechanically based kinetic theory of polymer chain deformations.

Finite viscoelastic overstress response is apparent in creep and relaxation tests. Cyclic loading tests show a typical frequency-dependent hysteresis. The width of the hysteresis increases with increasing stretch rates. The constitutive theory of finite linear viscoelasticity is a major foundation for modeling rate-dependent material behaviour based on the phenomenological approach. This general theory is formulated using functionals with fading memory properties. The stress is decomposed into an equilibrium stress that corresponds to the stress response at an infinite slow rate of deformation and a viscosity-induced overstress. The overstress is expressed as an integral over the deformation history and a relaxation function is specified as a measure for the material memory [7–10]. The thermodynamic consistency requires the relaxation function to be positive with negative slope and to possess a positive curvature [11]. Within this restriction certain number of decreasing exponentials can be superimposed, referred as a so-called Prony series. Such model requires a large number of material parameters that are difficult to estimate. Another innovative approach [11, 12] uses compact relaxation function based on power law, the Mittag-Leffler function [13], in describing Payne effect, and involves only a very few number of material parameters.

There exists another possibility of constructing finite strain models of viscoelasticity by considering the multiplicative decomposition of the deformation gradient into elastic and inelastic parts [14–18, 26]. In this approach, a suitable hyperelasticity model is employed to reproduce the elastic responses represented by the springs, while the dashpot represents the inelastic or the so-called internal strain. Its temporal behavior is determined by an evolution equation that is consistent with the second law of thermodynamics.

The third phenomenological effect observed in filled rubberlike materials is the partial rate-independent response. This phenomenon can be identified as a hysteresis of the relaxed equilibrium response within cyclic deformation processes and is usually denoted as a plastic effect [1]. If this effect is significant enough, the theory of viscoplasticity applies. If the equilibrium hysteresis appears to be negligible, a viscoelasticity theory is sufficient [19].

The important phenomenon in filled rubbers is the typical stress softening during the first loading cycles. This so-called Mullins effect can be interpreted as a damage effect where the evolution of the damage depends critically on the maximum stretch reached in the deformation history. A phenomenological model of Mullins effect was proposed by Ogden and Roxburgh [20] and recently by Marckmann [21].

3. Model for finite viscoelasticity

The material model of finite strain viscoelasticity used in our work follows from the concept of Simo [7] and Govindjee & Simo [22]. The finite element formulation of the model was elaborated by Holzapfel [23] and used by Holzapfel & Gasser [24] to calculate the viscoelastic deformation of fiber reinforced composite material undergoing finite strains. The model was incorporated into the new version of ANSYS 10.

The model is based on the theory of compressible hyperelasticity with the decoupled representation of the Helmholtz free energy function with the internal variables (Holzapfel [4], p. 283):

$$\Psi(\boldsymbol{C},\boldsymbol{\Gamma}_1,\dots,\boldsymbol{\Gamma}_m) = \Psi^{\infty}_{\mathrm{VOL}}(J) + \Psi^{\infty}_{\mathrm{ISO}}(\bar{\boldsymbol{C}}) + \sum_{\alpha=1}^{m} \Upsilon_\alpha(\bar{\boldsymbol{C}},\boldsymbol{\Gamma}_\alpha) \ , \qquad \bar{\boldsymbol{C}} = J^{-2/3}\,\boldsymbol{C} \ . \tag{1}$$

The first two terms in (1) characterize the equilibrium state and describe the volumetric elastic response and the isochoric elastic response as $t \to \infty$, respectively. The third term is the dissipative potential responsible for the viscoelastic contribution, C is right Cauchy-Green strain tensor and J is the determinant of the deformation gradient. The derivation of the 2$^{\mathrm{nd}}$ Piola-Kirchhoff stress tensor S with volumetric and isochoric parts:

$$\boldsymbol{S} = 2\,\frac{\partial\Psi(\boldsymbol{C},\boldsymbol{\Gamma}_1,\dots,\boldsymbol{\Gamma}_m)}{\partial\boldsymbol{C}} = \boldsymbol{S}^{\infty}_{\mathrm{VOL}} + \boldsymbol{S}^{\infty}_{\mathrm{ISO}} + \sum_{\alpha=1}^{m} \boldsymbol{Q}_\alpha \tag{2}$$

where $\boldsymbol{S}^{\infty}_{\mathrm{VOL}}$ and $\boldsymbol{S}^{\infty}_{\mathrm{ISO}}$ is the volumetric and the isochoric stress response respectively and the overstress $\boldsymbol{Q}_\alpha$ is stress of 2$^{\mathrm{nd}}$ Piola-Kirchhoff type.

$$\boldsymbol{S}^{\infty}_{\mathrm{VOL}} = J\,\frac{\mathrm{d}\Psi^{\infty}_{\mathrm{VOL}}(J)}{\mathrm{d}J}\,\boldsymbol{C}^{-1} \ , \qquad \boldsymbol{S}^{\infty}_{\mathrm{ISO}} = J^{-2/3}\ \mathrm{Dev}\left[2\,\frac{\partial\Psi^{\infty}_{\mathrm{ISO}}(\bar{\boldsymbol{C}})}{\partial\bar{C}}\right] \ , \tag{3}$$

$$\boldsymbol{Q}_\alpha = J^{-2/3}\ \mathrm{Dev}\left[2\,\frac{\partial\Upsilon_\alpha(\bar{\boldsymbol{C}},\boldsymbol{\Gamma}_\alpha)}{\partial\bar{\boldsymbol{C}}}\right] \ , \tag{4}$$

$$\mathrm{Dev}(\cdot) = (\cdot) - \frac{1}{3}[(\cdot):\boldsymbol{C}]\,\boldsymbol{C}^{-1} \tag{5}$$

where Dev$(\cdot)$ is the deviatoric operator in the Lagrangian description. Motivated by the generalized Maxwell rheological model (Fig. 1), the evolution equation for the internal variable Q_α takes on the form (6).

$$\dot{\boldsymbol{Q}}_\alpha + \frac{\boldsymbol{Q}_\alpha}{\tau_\alpha} = \dot{\boldsymbol{S}}_{\mathrm{ISO}\alpha} \ , \tag{6}$$

$$\boldsymbol{S}_{\mathrm{ISO}\alpha} = J^{-2/3}\ \mathrm{Dev}\left[2\,\frac{\partial\Psi^{\infty}_{\mathrm{ISO}\alpha}(\bar{\boldsymbol{C}})}{\partial\bar{\boldsymbol{C}}}\right] \ , \tag{7}$$

$$\Psi_{\mathrm{ISO}\alpha}(\bar{\boldsymbol{C}}) = \beta^{\infty}_\alpha\ \Psi^{\infty}_{\mathrm{ISO}}(\bar{\boldsymbol{C}}) \ , \tag{8}$$

$$\boldsymbol{S}_{\mathrm{ISO}\alpha} = \beta^{\infty}_\alpha\ \boldsymbol{S}^{\infty}_{\mathrm{ISO}}(\bar{\boldsymbol{C}}) \ . \tag{9}$$

$\beta^{\infty}_\alpha \in (0,\infty)$ in the expressions (8) and (9) is the nondimensional strain energy factor [7, 22] and is the relaxation time. The closed form solution of the linear evolution equation is given by the convolution integral and the recurence updated formula [4] for the internal stress

$$\begin{gathered}\boldsymbol{Q}_\alpha = \exp\left(-\frac{T}{\tau_\alpha}\right)\boldsymbol{Q}_{\alpha 0} + \int_0^T \exp\left(-\frac{T-t}{\tau_\alpha}\right)\beta^{\infty}_\alpha\,\dot{\boldsymbol{S}}^{\infty}_{\mathrm{ISO}}(\bar{\boldsymbol{C}})\,\mathrm{d}t \ , \\ (\boldsymbol{Q}_\alpha)_{n+1} = \exp(2\,\xi_\alpha)\,(\boldsymbol{Q}_\alpha)_n + \exp(\xi_\alpha)\,\beta^{\infty}_\alpha[(\boldsymbol{S}^{\infty}_{\mathrm{ISO}})_{n+1} - (\boldsymbol{S}^{\infty}_{\mathrm{ISO}})_n] \ , \qquad \xi_\alpha = -\frac{\Delta t}{2\,\tau_\alpha} \ .\end{gathered} \tag{10}$$

The material is assumed to be slightly compressible, the volumetric and isochoric (Mooney-Rivlin) parts of Helmholtz free energy function were chosen in the form

$$\Psi^{\infty}_{\mathrm{VOL}}(J) = \frac{1}{d}\,(J-1)^2\;, \qquad \Psi^{\infty}_{\mathrm{ISO}}(\bar{\boldsymbol{C}}) = c_1\,(\bar{I}_1 - 3) + c_2\,(\bar{I}_2 - 3)\;, \tag{11}$$

where $\bar{I}_1$ and $\bar{I}_2$ are first and second modified invariants of $\bar{\boldsymbol{C}}$ and the parameters c_1, c_2 and d are to be determined from experiments. The viscoelastic behavior is modeled by use of $\alpha = 2$ relaxation processes with the corresponding relaxation times τ_α and free energy factors β^{∞}_{α}. The stretch ans the second Piola-Kirchhoff stress in the loading direction of test specimens were determined from experimental measurements. The seven material parameters were calculated by nonlinear optimization methods in Matlab.

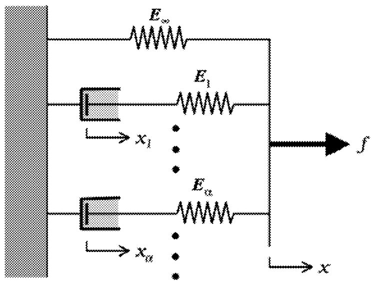

Fig.1: Maxwell rheological model

4. Experiment

The experiments were conducted in the laboratories of IT AV CR in Plzen and of TUL in Liberec. The specimens of filled rubber BAE 8534 used for compression tests were rectangular in shape, 47×23×25 mm. The rubber has the shear modulus of about 1 MPa and was manufactured by Rubena Hradec Kralove. In order to study the fundamental viscoelastic behavior of filled rubber, an experimental scheme was applied to each specimen. The scheme comprises of cyclic compression tests, multi-step relaxation tests and simple relaxation tests. All tests were performed at constant temperature under strain control and the experimental data were recorded by a personal computer. Prior to an actual test, each virgin specimen was subjected to a pre-loading process to remove the Mullins' softening effect. Mullins' effect is apparent on Fig. 2, where the hysteresis and the permanent set are also visible. This softening effect is partially recovered after a period of time depending on material in question (approximately one week in our case).

4.1. Cyclic tests at different strain rates

To study the material rate-dependence, specimens were subjected to cyclic compressive loading with constant strain rates. Fig. 3 presents the stress-strain responses as obtained from cyclic compression tests. The tests of compression was conducted with six different strain rates. A comparison of the stress responses indicates a strongly pronounced rate-dependent behaviour during loading, whereas a much weaker rate-dependence is observed during unloading. In addition, the presence of hysteresis and the permanent set is visible. All responses during loading suggest a diminishing trend in the increase of the stress with

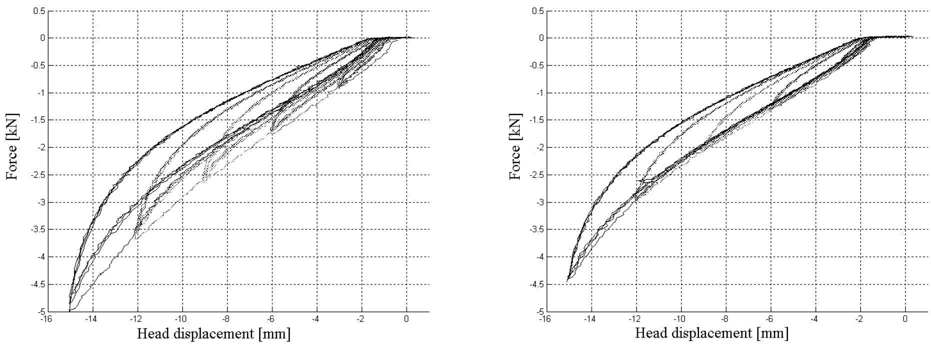

Fig.2: Mullins' effect; step cyclic loading; left – specimen without pre-loading, right – specimen after pre-loading to the maximum strain

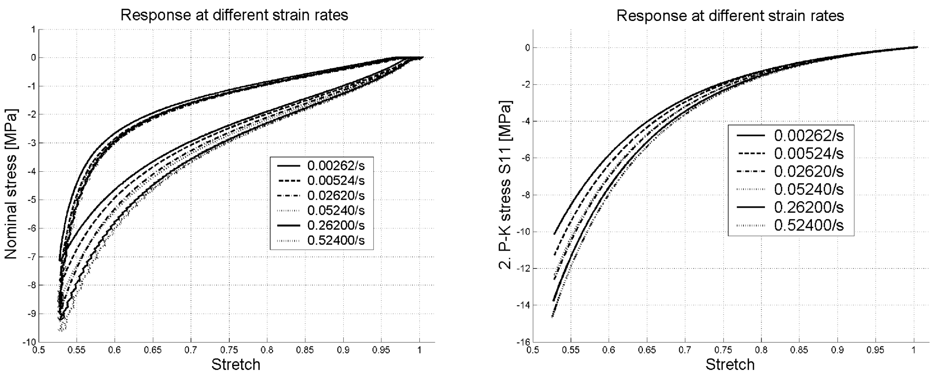

Fig.3: Cyclic tests at different strain rates; left – experiment, right – fitting of Mooney-Rivlin hyperelastic model to experimental data

increasing strain rate. Such a behaviour can be related to the approach of the material towards the so-called instantaneous stress response.

4.2. Relaxation tests

The relaxation behaviour at different strain levels is examined in detail through single-step and multi-step relaxation tests at Fig. 4 and 5. In the compression tests, a strain rate of 0.05 mm/s was applied during the loading path. The stress relaxation was being recorded for 1200 s. Fig. 4 shows the time histories of force at different strain levels in compression regime. All the curves reveal the existence of a very fast stress relaxation during the first 10 seconds after loading followed by a very slow rate of relaxation that continues in an asymptotic sense. This conforms with observations reported by Haupt and Sedlan [19]. Comparing the results obtained at different strain levels, it can be seen that relaxation tests conducted at higher strain levels possess larger overstresses and subsequently show a faster stress relaxation than those at lower strain levels with lower overstresses as reported also by Amin [25]. In the classical approach, equilibrium states are reached if the duration of the relaxation periods is infinitely long. Thus, the stresses measured at the termination points of the relaxation periods are approximate values of the equilibrium stress. The difference between the current stress and the equilibrium stress is the so-called overstress.

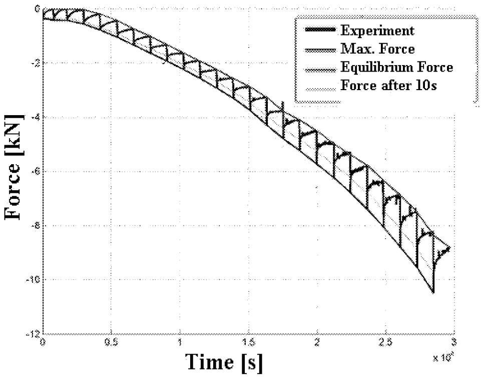

Fig.4: Step relaxation time history

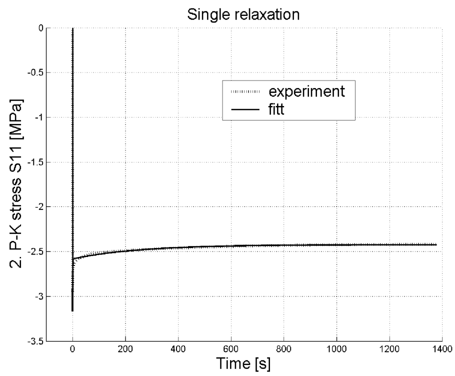

Fig.5: Single relaxation time history

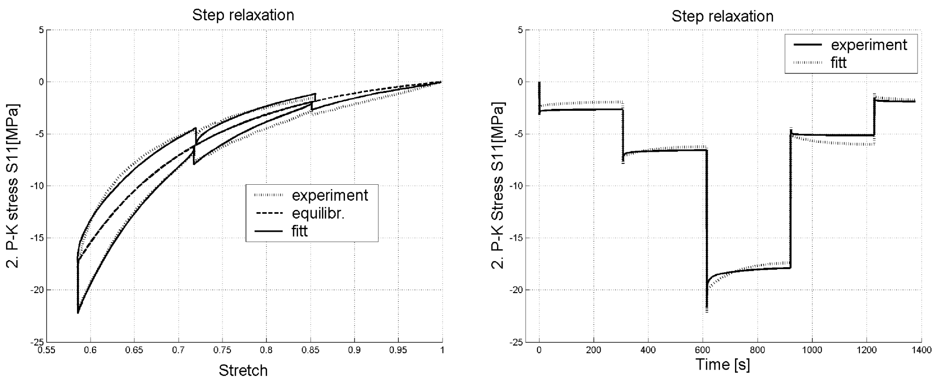

Fig.6: Fitting of the viscoelastic material model to experimental data

The constitutive model presented in the Section 3 was compared with the experimental data. The prediction of the step relaxation test is shown in Figure 6. These two graphs indicate that the model predicts the equilibrium stress and the uploading and unloading experimental data very well. However, for higher strain values, the behaviour during relaxation is not predicted well. The reason for this is that the model predicts the same time-dependence for different strain levels, but as was discussed above, the filled rubber exhibits faster relaxation during higher strains [26].

5. Finite element simulation

The cyclic loading test of the viscoelastic material at finite strains was simulated by FEM in Comsol Multiphysics 3.2 and in ANSYS 10. The material model described above was implemented into Comsol Multiphysics. The Structural Mechanics and PDE modules were used for the calculation of time dependent stresses in a rubber block loaded by time dependent displacement. The time dependent loading and the resulting stresses are shown at Fig. 7.

The ANSYS implementation based on Holzapfel viscoelastic model [9] uses the combination HYPER and PRONY options [29]. The parameters of the model were determined from the results of the cyclic experiments using a nonlinear least squares method. The capability of the material model to simulate the rate-dependent response of rubber is pre-

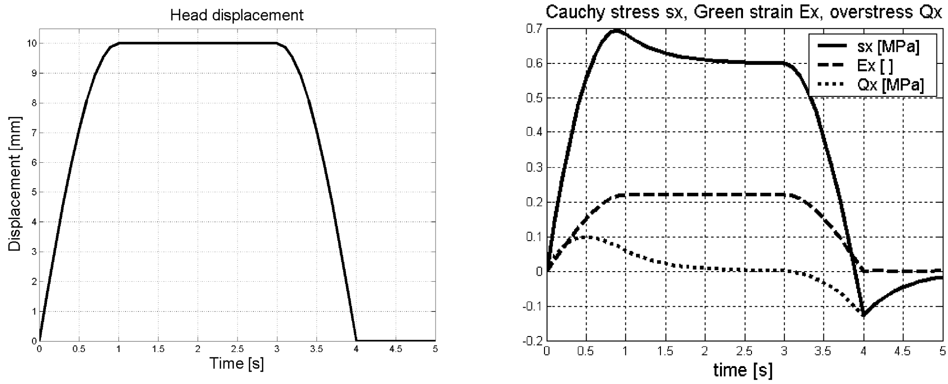

Fig.7: Simulation of cyclic test in Comsol Multiphysics

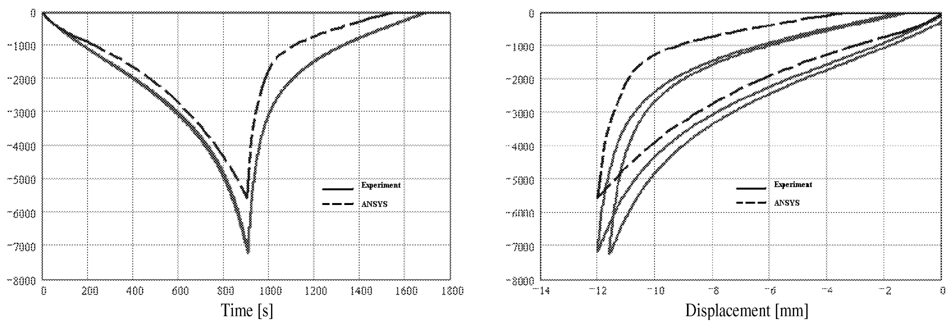

Fig.8: Experimental measurement compared with the simulation of the cyclic test in ANSYS

sented in Fig. 8 by comparing the stress calculated in ANSYS with experimental data. The results show good correlation between simulation and experiment for slow strain rates in compression. In general, the accuracy in predicting the experimental response was found to be better for slow strain rates and in the unloading stage of compression.

6. Conclusion

The experimental investigation demonstrated the time dependent behaviour of the filled rubber at large strains. The constitutive model used gives good quantitative agreement for different strain rates cyclic and relaxational behaviour. The model is already incorporated into finite element simulation. The incorporation of the viscoelastic effect into filled rubber behavior is the first step toward the coupled thermo-viscoelastic model and toward the possibility to calculate by FEM the dissipative heating of segments used for the rubber-sprung wheels. The improvement of the material model is achievable by considering nonlinear viscosity phenomena in the constitutive model [25] and to introduce the history dependence of viscosities [27] which leads to nonlinearly coupled equations. The effective relaxation times depending on amplitude and temperature can be also applied [28].

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