

MĚŘENÍ A IDENTIFIKACE VISOELASTICKÝCH PARAMETRŮ PLNĚNÝCH PRYŽÍ

MEASUREMENT AND IDENTIFICATION OF VISCOELASTIC PARAMETERS OF FILLED RUBBER

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Abstrakt

Odezva pryže na mechanické zatížení je závislá na rychlosti zatěžování a pryž při cyklickém zatěžování vykazuje hysterezi. I když toto chování pryže je dobře známo a je důležité pro funkci prýžových elementů konstrukcí, málokdy jsou viskoelastické vlastnosti zahrnuty v modelech prýžových materiálů. V článku jsou popsány základní testy pro určení viskoelastických parametrů pryže a metoda identifikace těchto parametrů z experimentálních měření pomocí nelineární optimalizace.

Klíčová slova: viskoelasticita, relaxace, pryž, materiálové testy, identifikace parametrů.

Abstract

The rate-dependent behavior of filled rubber is investigated in compression regimes. The viscosity-induced rate-dependent effects are described. The parameters of a constitutive model of finite strain viscoelasticity are determined by nonlinear optimization methods.

Keywords: viscoelasticity, relaxation, filled rubber, mechanical testing, identification of material parameters.

INTRODUCTION

Rubber materials are applied in various branches of mechanical engineering because of their damping properties. One of such applications of the orthotropic cord-reinforced rubber composite in engineering practice is the sheet of air-springs used for the vibro-isolation of driver seats. Hyperelastic models of such composite material at finite strains were developed in our laboratory and used successfully for FEM simulation of deformations of a cylindrical air-spring (Tran Huu Nam [20]; Urban, R., [21]). Nowadays we are interested in the dissipative behaviour of cord-reinforced rubber composites - in their viscoelastic properties which determine their damping function. The modelling and FEM calculation of the structural response requires a constitutive model which captures the complex material behaviour. A viscoelastic model for the fully three-dimensional stress and deformation response of cord-reinforced rubber composites at finite strains requires the knowledge of material parameters of their components. The relaxation response of each compound of the composite is modelled separately and the global response is obtained by an assembly of all contributions. Viscoelastic properties of the cords were subject of our previous work. The present paper focuses on the viscoelastic behaviour of the filled rubber in cyclic and relaxation experiments.

The constitutive theory of finite linear viscoelasticity is a major foundation for modelling rate-dependent filled-rubber behaviour based on the phenomenological approach. This general

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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 (Simo, 1987; Holzapfel and Simo [19]; Holzapfel [7]; Kaliske and Rother [11]). The thermodynamic consistency requires the relaxation function to be positive with negative slope and to possess a positive curvature (Haupt and Lion [5]). Within this restriction certain number of decreasing exponentials can be superimposed, referred as a so-called Prony series. 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. This process may invite a large number of material parameters in the model that are difficult to estimate. Another innovative approach (Haupt [4], Haupt and Lion [5]) uses compact relaxation function based on power laws, the Mittag-Leffler function (Lion and Kardelky [15]), in describing Payne effect, and involves only a very few number of material parameters.

There exists another possibility to establish finite strain models of viscoelasticity by considering the multiplicative decomposition of the deformation gradient into elastic and inelastic parts (Lion [13], Reese & Govindjee [17], Bonet [2], Laiarinandrasana [12], Reese [18], Bergstrom & Boyce [22]). The temporal behavior is determined by an evolution equation that is consistent with the second law of thermodynamics.

MODEL FOR FINITE VISCOELASTICITY

The origin of the material model of finite strain viscoelasticity used in our work is the concept of Simo [19] and Govindjee & Simo [3]. The finite element formulation of the model was elaborated by Holzapfel [7] and used by Holzapfel & Gasser [9] to calculate the viscoelastic deformation of fibre 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 [10], p. 283)

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

The first two terms in (1) characterize the equilibrium state and describe the volumetric elastic response and the isochoric elastic response as $t \rightarrow \infty$, respectively. The third term is the dissipative potential responsible for the viscoelastic contribution. The derivation of the 2nd Piola-Kirchhoff stress with volumetric and isochoric terms:

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

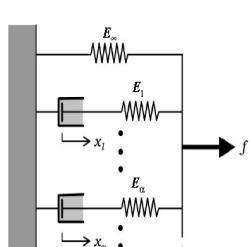
where \mathbf{S}_{VOL}^∞ and \mathbf{S}_{ISO}^∞ is the volumetric and the isochoric stress response respectively and the overstress \mathbf{Q}_α is stress of 2nd Piola-Kirchhoff type.

$$\mathbf{S}_{VOL}^\infty = J \frac{d \psi_{VOL}^\infty(J)}{d J} \mathbf{C}^{-1}, \quad \mathbf{S}_{ISO}^\infty = J^{-2/3} \text{Dev}[2 \frac{\partial \psi_{ISO}^\infty(\bar{\mathbf{C}})}{\partial \bar{\mathbf{C}}}] \quad (3)$$

$$\mathbf{Q}_\alpha = J^{-2/3} \text{Dev}[2 \frac{\partial Y_\alpha(\bar{C}, \Gamma_\alpha)}{\partial C}], \quad (4)$$

$$\text{Dev}(.) = (.) - 1/3[(.) : C]C^{-1}. \quad (5)$$

where $\text{Dev}(.)$ 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).



$$\dot{Q}_\alpha + \frac{Q_\alpha}{\tau_\alpha} = \dot{S}_{ISO\alpha}, \quad (6)$$

$$S_{ISO\alpha} = J^{-2/3} \text{Dev}[2 \frac{\partial \psi_{ISO\alpha}(\bar{C})}{\partial \bar{C}}], \quad (7)$$

$$\Psi_{ISO\alpha}(\bar{C}) = \beta_\alpha^\infty \Psi_{ISO}^\infty(\bar{C}), \quad (8)$$

Fig. 1. Maxwell rheological model

$$S_{ISO\alpha} = \beta_\alpha^\infty S_{ISO}^\infty(\bar{C}). \quad (9)$$

$\beta_\alpha^\infty \in (0, \infty)$ in the expressions (8) and (9) is the nondimensional strain energy factor and τ_α is the relaxation time. The closed form solution of the linear evolution equation is given by the convolution integral and the recurrence updated formula for the internal stress

$$\begin{aligned} Q_\alpha &= \exp(-T/\tau_\alpha) Q_{\alpha 0} + \int_0^T \exp(-(T-t)/\tau_\alpha) \beta_\alpha^\infty \dot{S}_{ISO}^\infty(t) dt, \\ (Q_\alpha)_{n+1} &= \exp(2\xi_\alpha)(Q_\alpha)_n + \exp(\xi_\alpha)\beta_\alpha^\infty[(S_{ISO}^\infty)_{n+1} - (S_{ISO}^\infty)_n], \quad \xi_\alpha = -\frac{\Delta t}{2\tau_\alpha}. \end{aligned} \quad (10)$$

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

$$\psi_{VOL}^\infty(J) = \frac{1}{d}(J-1)^2, \quad \psi_{ISO}^\infty(\bar{C}) = c_1(\bar{I}_1 - 3) + c_2(\bar{I} - 3), \quad (11)$$

where 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 material is assumed to be isotropic. To evaluate the experimental results the finite strain model needs to be degenerated into the corresponding one-dimensional form in accordance with the deformations applied in the experiments, e.g., uniaxial compression. If a specimen is subjected to uniaxial homogeneous compression, the principal stretch λ_1 in the loading direction becomes compressed and those in the two other directions λ_2 and λ_3 are under tension. Thus, the deformation gradient F reads as:

$$F = \begin{bmatrix} \lambda_1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{bmatrix}. \quad (12)$$

For the isotropic material $\lambda_2 = \lambda_3 = \lambda_1^{-v}$ (for the incompressible material $v = 0.5$, we suppose $v = 0.49$ for the slightly compressible material). The modified right Cauchy-Green deformation tensor in (1) and its first invariant are:

$$\bar{\mathbf{C}} = J^{-\frac{2}{3}} \begin{bmatrix} \lambda_1^2 & 0 & 0 \\ 0 & \lambda_2^2 & 0 \\ 0 & 0 & \lambda_3^2 \end{bmatrix}, \quad J = \lambda_1 \lambda_2 \lambda_3, \quad \bar{I} = J^{\frac{2}{3}} (\lambda_1^2 + \lambda_2^2 + \lambda_3^2) \quad (13)$$

The modified equilibrium isochoric part of 2nd Piola-Kirchhoff stress tensor in (3) reads as:

$$\bar{\mathbf{S}}_{ISO}^\infty = 2 \frac{\partial \psi_{ISO}^\infty(\bar{\mathbf{C}})}{\partial \bar{\mathbf{C}}} = 2(c_1 + c_2 \bar{I}_1) \mathbf{I} - 2c_2 \bar{\mathbf{C}} . \quad (14)$$

The components of the 2nd Piola-Kirchhoff stress tensor \mathbf{S}_{VOL}^∞ and \mathbf{S}_{ISO}^∞ in (3) for $\lambda_2 = \lambda_3$ are:

$$\begin{aligned} \bar{\mathbf{S}}_{VOL}^\infty &= J(J-1) \frac{2}{d} \text{diag} \begin{bmatrix} \lambda_1^{-2} \\ \lambda_2^{-2} \\ \lambda_3^{-2} \end{bmatrix}, \\ \mathbf{S}_{ISO}^\infty &= \frac{2}{3} J^{-\frac{2}{3}} \text{diag} \begin{bmatrix} 2c_1 [1 - \left(\frac{\lambda_2}{\lambda_1}\right)^2] J + 2c_2 \lambda_2^2 J^{-\frac{2}{3}} [1 - \left(\frac{\lambda_2}{\lambda_1}\right)^2] J \\ c_1 [1 - \left(\frac{\lambda_1}{\lambda_2}\right)^2] J + c_2 \lambda_2^2 J^{-\frac{2}{3}} [1 - \left(\frac{\lambda_1}{\lambda_2}\right)^2] J \\ dtto \end{bmatrix}. \end{aligned} \quad (15)$$

The total 2nd Piola-Kirchhoff stress tensor is the sum of volumetric, isochoric and overstress parts in (2). Since the lateral boundaries of the experimental specimen are stress-free the 2nd Piola-Kirchhoff stress tensor in experiments is given by

$$\mathbf{S} = \begin{bmatrix} S_{11} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \quad (16)$$

The components S_{22} and S_{33} of the principal stress tensor then must be zero

$$\begin{aligned} S_{22} &= S_{VOL22}^\infty + S_{ISO22}^\infty + \sum_{\alpha=1}^m Q_{\alpha22} = 0, \\ J(J-1) \frac{2}{d} \lambda_2^{-2} + \frac{2}{3} J^{-\frac{2}{3}} \left\{ c_1 [1 - \left(\frac{\lambda_1}{\lambda_2}\right)^2] J + c_2 \lambda_2^2 J^{-\frac{2}{3}} [1 - \left(\frac{\lambda_1}{\lambda_2}\right)^2] J \right\} + \sum_{\alpha=1}^m Q_{\alpha22} &= 0. \end{aligned} \quad (17)$$

We substitute d from eq. (17) into the expression for the principal stress S_{11} .

$$\begin{aligned}
S_{11} &= S_{11}^\infty + \sum_{\alpha=1}^m Q_{\alpha 11}, \\
S_{11}^\infty &= 2J^{-\frac{2}{3}} \left\{ c_1 \left[1 - \left(\frac{\lambda_2}{\lambda_1} \right)^2 \right] + c_2 \lambda_2^2 J^{-\frac{2}{3}} \left[1 - \left(\frac{\lambda_2}{\lambda_1} \right)^2 \right] \right\}, \\
Q_{\alpha 11} &= \int_0^T \exp(-(T-t)/\tau_\alpha) \beta_\alpha^\infty \dot{S}_{11}^\infty(t) dt, \\
Q_{\alpha 11/0} &= 0.
\end{aligned} \tag{18}$$

In order to establish a relation between theory and experiment, we note the equivalence of the experimentally recorded stresses S_{11} with the 2nd Piola-Kirchhoff stress S_{11} in eq. (18). Furthermore, the values of the stress recorded at the termination points of the relaxation periods are regarded as equilibrium stresses S_{11}^∞ . Based on this concept, it is possible to solve nonlinear second eq. (18) alone to calculate the hyperelastic constants c_1 and c_2 by a numerical optimization method. Thus, the experimentally recorded stress relaxation history of S_{11} can be analyzed to obtain the corresponding relaxation times τ_α and free energy factors β_α^∞ . Next the bulk modulus constant d can be obtained from eq. (17).

The stretches λ_1 in the loading direction and the components S_{11} of the 2nd Piola – Kirchhoff stress of test specimens are determined from experimental results:

$$\begin{aligned}
\lambda_1 &= -\Delta / L_0 + 1, \\
S_{11} &= -P / A_0 / \lambda_1,
\end{aligned} \tag{19}$$

where Δ is the compressive displacement of specimen boundary, P is the compressive force, A_0 and L_0 are the initial area and initial length of specimen respectively.

RELAXATION TESTS

The relaxation behaviour at different strain levels is examined in detail through multi-step relaxation test. In the compression tests, a strain rate of 0,05mm/s was applied during the loading path. The stress relaxation was recorded for 1200 s. Fig. 2 shows the time histories of force at different strain levels in compression regime. All curves reveal the existence of a very fast stress relaxation during the first 10 seconds followed by a very slow rate of relaxation that continues in an asymptotic sense. This conforms with observations reported by Haupt and Sedlan [6]. Comparing the results obtained at different strain levels, it can be seen that relaxation tests carried out at higher strain levels possess larger over-stresses and subsequently show a faster stress relaxation than those at lower strain levels with lower over-stresses as reported also by Amin [1]. 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.

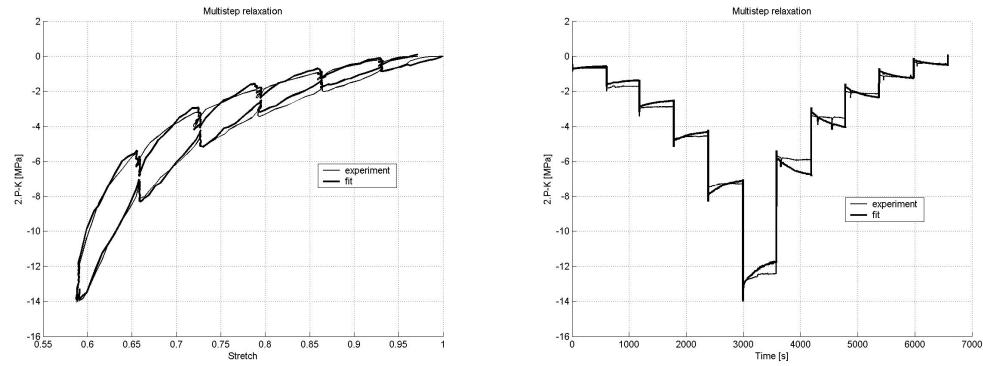


Fig. 2 Multi-step relaxation experiment

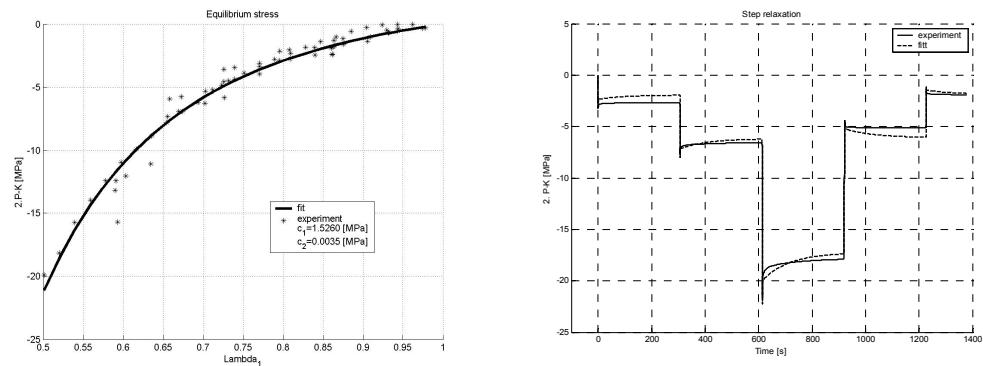


Fig. 3 Equilibrium stress and multi-step relaxation

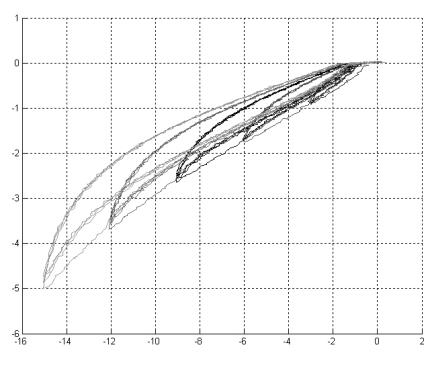


Fig.4 Cyclic tests - Mullins'effect

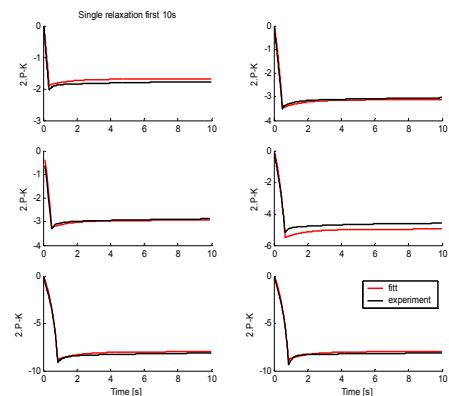


Fig.5 Single relaxation - first 10 s

Fig.2 compares the experimental data and the curves fitted to the proposed material model by nonlinear least squares method. The curve at Fig.3 approximates the equilibrium stress

$S_{VOL}^\infty + S_{ISO}^\infty$. The good performance of the model in capturing the main material features is obvious.

To characterize better the viscosity properties, a series of simple relaxation tests at different stretch levels were carried out. In this course, a stretch rate of 0.05/s followed by a hold time of 20 min was used in the tests. The results are at Fig. 5.

All tests were performed in Hydrodynamic laboratory of TUL in Liberec 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. 4, where the hysteresis and the permanent set are also visible. The second Piola – Kirchhoff stress and the stretch in the loading direction of test specimens were determined from experimental results. The seven material parameters were calculated by nonlinear optimization methods in Matlab.

CONCLUSION

Step-strain relaxation and single relaxation of a filled rubber were modelled with viscoelastic theory. The parameters of the model were determined from relaxation data by employing a nonlinear optimization methods. The proposed model is then compared with experimental data for filled rubber subjected to different loading histories. It is shown that the model gives good quantitative agreement for different relaxational behaviour. An intensive research of the mechanical behaviour of carbon black filled rubbers is currently in progress and will be the topic of a future report.

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REFERENCES

- [1] AMIN, A.F.M.S., LION A., SEKITA, S., OKUI, Y.: *Nonlinear dependence of viscosity in modeling the ratedependent response of natural and high damping rubbers in compression and shear: Experimental identification and numerical verification*. Int. J. of Plasticity, 2005, in press
- [2] BONET, J.: *Large strain viscoelastic constitutive models*. Int. J. Solid Struct. 38, 2001, 2953–68.
- [3] GOVINDJEE, S., SIMO, J.C.: *Mullins' effect and strain amplitude dependence of the storage modulus*. Int. J. Solid Struct. 29, 1992, 1737–1751.
- [4] HAUPT, P.: *Continuum Mechanics and Theory of Materials*. Springer, 2002, ISBN 354043111X
- [5] HAUPT, P., LION, A.: *On finite linear viscoelast. of incompress. isotrop. materials*. Acta Mech. 159, 2002, 87–124.
- [6] HAUPT, P., SEDLAN, K.: *Viscoplasticity of elastomeric materials: experimental facts and constitutive modelling*. Arch. Appl. Mech. 71, 2001, 89–109.
- [7] HOLZAPFEL, G.A.: *On large strain viscoelasticity: continuum formulation and finite element applications to elastomeric structures*. Int. J. Numer. Meth. Eng. 39, 1996, 3903–3926.
- [8] HOLZAPFEL, G.A., SIMO, J.C.: *A new viscoelastic constitutive model for continuous media at finite thermomechanical changes*. Int. J. Solid Struct. 33, 1996, 3019–3034.

- [9] HOLZAPFEL, G. A., GASSER, T.C.: *A viscoelastic model for fiber-reinforced composites at finite strains: Continuum basis, computational aspects and applications.* Computer Methods in Applied Mechanics and Engineering, Vol. 190, 2001, 4379-4430
- [10] HOLZAPFEL, G. A.: *Nonlinear solid mechanics.* Wiley, N.Y., 2000, ISBN 0471823198
- [11] KALISKE, M., ROTHERT, H.: *Formulation and implementation of three-dimensional viscoelasticity at small and finite strains.* Comput. Mech. 19, 1997, 228–239.
- [12] LAIARINANDRASANA, L., PIQUES, R., ROBISSON, A.: *Visco-hyperelastic model with internal state variable coupled with discontinuous damage concept under total Lagrangian formulation.* Int. J. Plasticity 19, 2003, 977–1000.
- [13] LION, A.: *A physically based method to represent the thermo-mechanical behavior of elastomers.* Acta Mech. 123, 1997, 1–25.
- [14] LION, A.: *Thixotropic behavior of rubber under dynamic loading histories: experiments and theory.* J.Mech.Phys. Solids, Vol.46, 1998, no.5, 895-930
- [15] LION, A., KARDELKY, C.: *The Payne effect in finite viscoelasticity constitutive modelling based on fractional derivatives and intrinsic time scales.* Int. J. Plasticity 20, 2004, 1313–1345.
- [16] NEMETH, I., SCHLEINZER, G., OGDEN, R.W., HOLZAPFEL, G.A.: *On the modelling of amplitude and frequencydependent properties in rubberlike solids.* In.: Constitutive Models for Rubber IV, P.-E. Austrell and L. Keri (eds.), A.A. Balkema Publishers: Leiden, 2005, 285-298.
- [17] REESE, S., GOVINDJEE, S.: *A theory of finite viscoelasticity and numerical aspects.* Int. J. Solid Struct. 35, 1998, 3455–3482.
- [18] REESE, S.: *A micromechanically motivated material model for the thermo-viscoelastic material behaviour of rubber-like polymers.* International Journal of Plasticity 19, 2003, 909–940
- [19] SIMO, J.C.: *On a fully three dimensional finite strain viscoelastic damage model: formulation and computational aspects.* Comput. Meth. Appl. Mech. Eng. 60, 1987, 153–173.
- [20] TRAN HUU NAM: *Mechanical properties of the composite material with elastomeric matrix reinforced by textile cords,* 2004, PhD thesis,TUL of Liberec
<http://www.kmp.vslib.cz/lide/index.php?clovek=nam&lang=cs>
- [21] URBAN, R.: *Modelování konstrukčních prvků z pryže vyztužené nitěmi,* Doktorská disert. práce, TUL v Liberci, 2004,
<http://www.kmp.vslib.cz/lide/index.php?clovek=urban&lang=cs>
- [22] BERGSTROM, J. S., BOYCE, M. C.: *Constitutive modelling of the large strain time-dependent behavior of elastomers.* J. Mech. Phys. Solids., Vol. 46, 1998, pp. 931-954