BÁNKI KÖZLEMÉNYEK
1. ÉVFOLYAM 1. SZÁM

Characterisation of large strain viscoelastic properties of polymers

Bálint Fazekas*,Tibor Goda*
*Department of Machine and Product Design, Budapest University of Technology and Economics, H-1111, Budapest, Műegyetem rkp. 3, Hungary
fazekas.balint@gt3.bme.hu
goda.tibor@gt3.bme.hu

Abstract

The most widely used approach to model the large strain elastic and the time-dependent response of polymers in a finite element simulation is the application of the so-called hyper-viscoelastic material model, which is composed of a nonlinear elastic (hyperelastic) and a linear viscoelastic part. In order to determine the constitutive parameters, a simple numerical algorithm has been used here. This method ensures a general strategy to fit the material parameters directly and accurately to the measurements. In this study, the material model and the numerical algorithm have been tested for silicone rubber and polypropylene. Finally, the numerical solutions have been compared with stress relaxation tests.

Keywords: hyperelasticity, finite strain viscoelasticity, Prony-series, parameter identification, polymers

1 INTRODUCTION

Polymers are widely applied materials, among others, in the automotive, the aerospace and the computer industry. They exhibit nonlinear stress-strain relation (Fig. 1 (a)) and time/rate dependent behaviour (Fig. 1 (b)) [1].

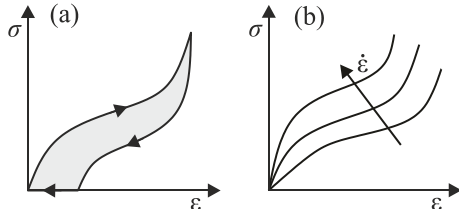

Fig. 1: Material behaviour of viscoelastic solids: (a) loading and unloading, (b) strain-rate dependency

Fig. 1 (a) shows that energy dissipation (hysteresis) is appeared during loading-unloading and the strain-rate influences the overall mechanical response (Fig. 1 (b)). These properties are essential characteristics of viscoelastic materials.

To model this complex mechanical behaviour, a so-called large strain viscoelastic material model has to be used, which consists of a nonlinear elastic and a linear viscoelastic part. The former takes the time-independent, while the latter takes the time– and rate dependent behaviour into consideration [2]. At the same time, it is assumed that the material is linear in rheological sense. In this particular case, the constitutive equation can be split into a strain-dependent and a time-dependent part. Although this type of material model is frequently used, the determination of the model parameters is not a trivial task. This is proved, among others, by the fact that various approaches are available for the determination of the constitutive parameters in the literature. Here, the numerical algorithm proposed by Goh et al. [3] is used, where the convolution integral of the constitutive equation is integrated numerically based on the finite time increments and the material parameters are optimised in one step using measurement results. The method is applied for silicone rubber and polypropylene using tensile stress relaxation tests [4, 5]. Finally, the simulated and the measured stress relaxation behaviours are compared.

2 MATERIAL MODEL

2.1 Large strain elastic model

The large strain time-independent behaviour of polymers can be modelled with hyperelastic material model, where the constitutive equations can be derived from the strain energy density function W. It is assumed that the material is isotropic, therefore W can be expressed with the principal invariants (I_1, I_2, I_3) of the right Cauchy–Green deformation tensor $\mathbf{C}$. In many cases, the material can be considered to be incompressible ($i_3 = 1$). In this study, two different hyperelastic models are used. The strain energy density functions of the models are as follows

$$W^{\mathrm{NH}} = C_{10}(I_1 - 3), \tag{1}$$

$$W^{\mathrm{Y}} = C_{10}(I_1 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3, \tag{2}$$

where W^{NH} is the neo-Hookean [6], while W^{Y} is the Yeoh [7] hyperelastic potential, respectively. C_{10}, C_{20}, C_{30} are material parameters, furthermore

$$I_1 = \mathrm{tr}(\mathbf{C}) = \lambda_1^2 + \lambda_2^2 + \lambda_3^2, \tag{3}$$

where λ_k ($k = 1,2,3$) denotes the principal stretches.

The first Piola–Kirchhoff principal stresses (also known as engineering principal stresses) can be obtained as [8]

$$P_k = \frac{\partial W}{\partial \lambda_k}, k = 1,2,3\,. \tag{4}$$

In case of uniaxial extension, the deformation gradient can be written as

$$\mathbf{F} = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & 1/\sqrt{\lambda} & 0 \\ 0 & 0 & 1/\sqrt{\lambda} \end{bmatrix}, \tag{5}$$

where λ is the stretch ratio in the direction of load. It should be noted that the incompressible condition ($J = \det \mathbf{F} = 1$) was taken into account during the derivation of the deformation gradient **F**. As the strain energy density functions of the chosen hyperelastic models are given by Eqs. (1-2), the uniaxial stress responses can be derived using Eqs. (4-5). The engineering stress response of the neo-Hookean model becomes

$$P_1^{\mathrm{NH}} = 2C_{10}\left(\lambda - \lambda^{-2}\right), \tag{6}$$

while the engineering stress-strain relation of the Yeoh model can be expressed as

$$P_1^{\mathrm{Y}} = 2\left(\lambda^3 - 1\right)\left[C_{10}\lambda^2 + \left(\lambda^3 - 3\lambda + 2\right)\left(2C_{20}\lambda + 3C_{30}\left(\lambda^3 - 3\lambda + 2\right)\right)\right] / \lambda^4 . \tag{7}$$

It should be noted that, in case of time-dependent material behaviour, the strain is expressed as a function of time, thus the following notation is introduced

$$P_0(t) := P_1(\lambda(t))\,. \tag{8}$$

2.2 Large strain viscoelastic model

Since the deformation of the polymers is characterised by large strains and displacements, the hyper-viscoelastic material model has to be used, which takes into consideration both the large strain elastic (hyperelastic) and the viscoelastic behaviour of the material. It should be noted that the model used is linear in the sense that the dimensionless relaxation modulus is not a function of the strain, i.e. in rheological sense. Here the generalised Maxwell-model connected parallel to a nonlinear spring is used (see Fig. 2).

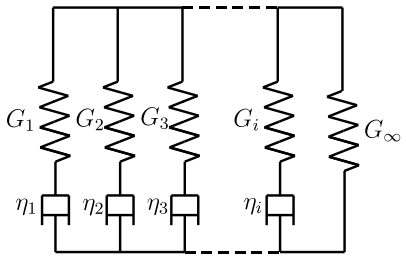

Fig. 2: The material model used: generalised Maxwell-model connected parallel to a nonlinear spring

The constitutive model consists of Maxwell-elements (linear spring and dashpot connected in series) connected in parallel which describes the time-dependent behaviour. Furthermore, it consists of a nonlinear spring, which gives the nonlinear long-term (relaxed) elastic modulus of the material. G_i denotes the i-th shear modulus, η_i is the i-th dynamic viscosity (i=1..N) while G_∞ is the long-term (relaxed) shear modulus.

The constitutive model consists of separable strain-dependent function ($\sigma_0(\varepsilon)$) and time-dependent function ($g(t)$) and can be written as

$$\sigma(\varepsilon, t) = \sigma_0(\varepsilon) g(t)\,, \tag{9}$$

where $\sigma_0(\varepsilon)$ denotes the instantaneous Cauchy stress, which gives the instantaneous elastic (time-independent) response of the material. It can be derived from the hyperelastic potential (strain energy density function). The $g(t)$ normalised function is strain-independent and represents the viscoelastic behaviour of the material. It can be expressed with the Prony-series as

$$g(t) = g_\infty + \sum_{i=1}^{N} g_i e^{-t/\tau_i}\,, \tag{10}$$

where g_i is the i-th relative, while g_∞ is the long-term relative modulus, respectively. Furthermore, N denotes the order of the viscoelastic model and τ_i is the i-th relaxation time. These quantities are defined as follows

$$g_i = \frac{G_i}{G_0}, g_\infty = \frac{G_\infty}{G_0},\ \tau_i = \frac{\eta_i}{G_i}, \tag{11}$$

where G_0 is the instantaneous (glassy) shear modulus. For the long-term and the relative relaxation moduli the following equation can be written

$$g_\infty + \sum_{i=1}^{N} g_i = 1. \tag{12}$$

In general case, the viscoelastic stress response can be expressed in the form of convolution integral as

$$\sigma(\varepsilon, t) = \int_0^t g(t-s) \frac{\mathrm{d}\sigma_0(\varepsilon)}{\mathrm{d}s} \mathrm{d}s\,. \tag{13}$$

It can be split into long-term elastic and viscoelastic contribution as follows

$$\sigma(t) = g_\infty \sigma_0(t) + \sum_{i=1}^{N} \int_0^t g_i e^{-(t-s)/\tau_i} \frac{\mathrm{d}\sigma_0(s)}{\mathrm{d}s} \mathrm{d}s = g_\infty \sigma_0(t) + \sum_{i=1}^{N} h_i(t)\,. \tag{14}$$

The closed-form solution of Eq. (14) cannot be derived for arbitrary hyperelastic potential and strain-history. However, it is possible to compute the stress response numerically based on finite time increments. The detailed derivation of this method is provided by Goh et al. [3].

As the material parameters are fitted to the experimental data and the engineering stress can be obtained from the measurements directly, the incremental stress solution is derived using engineering stresses. At time instant t_{n+1} the engineering stress response may be written as

$$P(t_{n+1}) = g_\infty P_0(t_{n+1}) + \sum_{i=1}^{N} e^{-\Delta t/\tau_i} h_i(t_n) \frac{\lambda(t_n)}{\lambda(t_{n+1})} + \\ + g_i \frac{1-e^{-\Delta t/\tau_i}}{\Delta t/\tau_i} \left[P_0(t_{n+1}) - P_0(t_n) \frac{\lambda(t_n)}{\lambda(t_{n+1})} \right], \quad (15)$$

where $P_0(t_n)$, $P_0(t_{n+1})$ values can be computed with the engineering stress solutions of the neo-Hookean and the Yeoh hyperelastic models (Eqs. (6-7)) and $\Delta t = t_{n+1} - t_n$ denotes the time increment. Since the initial stress-strain state is normally known ($P = \varepsilon = 0$ at time $t = 0$), the incremental stress response can be computed at time $t > 0$. It should be noted that Eq. (15) is only valid for uniaxial extension.

Using Eq. (15), the hyperelastic and the viscoelastic constitutive parameters can be fitted simultaneously, directly to the measurement. The error is defined as

$$\zeta = \sum_{j=1}^{M} \left| P_j^{\exp} - P_j^{\text{comp}} \right|, \quad (16)$$

where $P_j^{\exp}$ and P_j^{comp} are the experimental and the computed engineering stress values, respectively. The objective is to find the optimal material parameters through the minimisation of the error value given by Eq. (16). This minimisation problem is solved by using the 'Solver' routine of Microsoft Excel.

3 Results and discussion

In order to prove the applicability of the hyper-viscoelastic material model and the accuracy of the parameter fitting method, already published uniaxial stress relaxation tests for silicone rubber and polypropylene are used. The details of these experiments are available in [4, 5]. The model parameters for both materials are determined using Eq. (15).

The first step of the fitting method is to investigate that whether the material behaviour is linear in rheological sense or not. As it is mentioned in the former section, the most important character of a linear viscoelastic material is that the strain and time-dependent material behaviour are independent, i.e. $\sigma_0(\varepsilon)$ and $g(t)$ are separable (see Eq. (9)). This behaviour of the material can be studied through the normalised relaxation curves. If these curves are coincident for all strains, the material can be modelled with the presented large strain viscoelastic material model [3, 9]. The normalised relaxation curves of the materials studied are presented in Fig. 3.

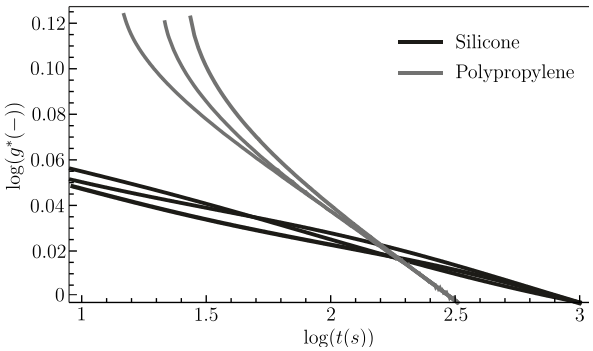

Fig. 3: The normalised stress relaxation curves

Fig. 3 shows that the normalised relaxation curves of the silicone are close to each other at different strain levels, therefore the material behaviour can be considered to be linear in rheological sense. However, in case of polypropylene, the nonlinear viscoelastic behaviour is appeared.

The time-independent behaviour of the silicone and the polypropylene is modelled using the neo-Hookean and the Yeoh hyperelastic models, respectively. The time-dependency is described with third-order viscoelastic model ($N = 3$) for both materials. The parameter fitting method is performed based on the stress relaxation curves using Eq. (15). The obtained material parameters are given in Table 1.

Tabel 1: Material model parameters for silicone and polypropylene

Material parameter	Silicone rubber	Polypropylene
C_{10} [MPa]	0.153	544.26
C_{20} [MPa]	-0.0111	-
C_{30} [MPa]	0.00324	-
g_1 [-]	0.0714	0.125
τ_1 [s]	7.64	5.85
g_2 [-]	0.0723	0.315
τ_2 [s]	240.8	5.84
g_3 [-]	0.01	0.098
τ_3 [s]	200	162.5

It should be noted that the hyperelastic parameters (C_{10}, C_{20}, C_{30}) are required to satisfy the Drucker stability condition. Furthermore, for the viscoelastic parameters (g_i, τ_i), the following conditions have to be taken into account

$$\sum_{i=1}^{N} g_i < 1; \; g_i > 0, \; \tau_i > 0 \text{ for all } i. \quad (16)$$

Using the material model parameters given in Table 1, the finite element (FE) simulation of uniaxial stress relaxation tests has been performed using the commercial finite element software Abaqus [10]. The simulated engineering stress responses and their comparison with the experiments are shown in Figs. 4 and 5.

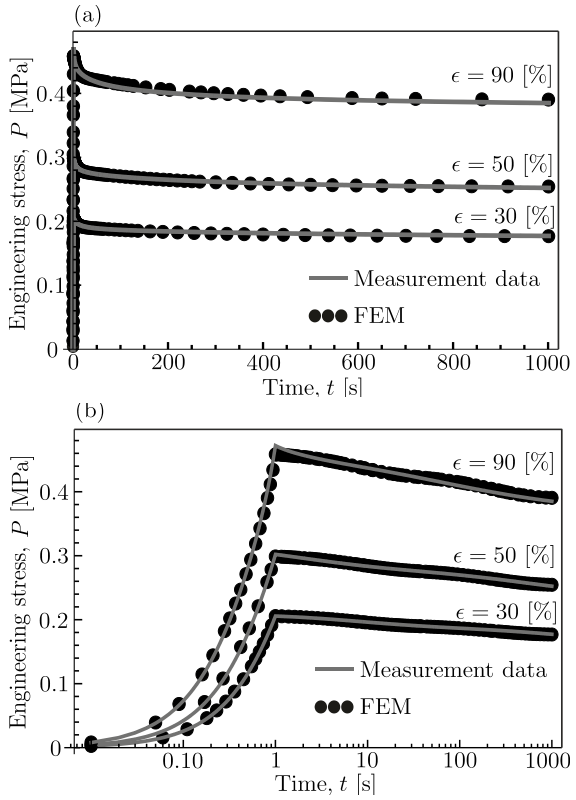

Fig. 4: The simulated and measured engineering stress-time curves for silicone rubber: (a) linear scale, (b) logarithmic scale

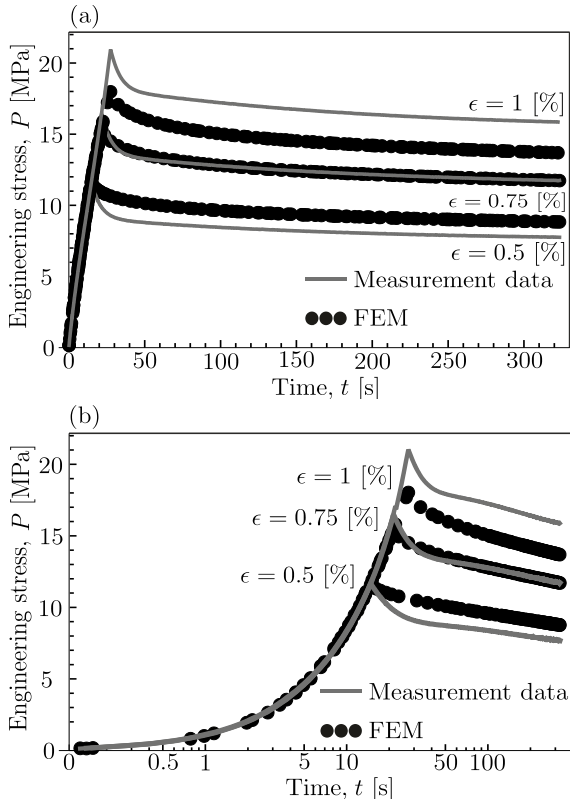

Fig. 5: The simulated and measured engineering stress-time curves for polypropylene: (a) linear scale, (b) logarithmic scale

As seen in Fig. 4, there is a good agreement between the simulated and the measured stress relaxation curves in case of silicone rubber. Therefore, the hyper-viscoelastic material model presented here is able to model the linear viscoelastic material behaviour accurately. In case of polypropylene (Fig. 5), however, the simulated and the measured curves differ considerably especially at 0.5 [%] and 1 [%] strain levels due to the fact that the polypropylene exhibits nonlinear viscoelastic behaviour (see Fig. 3). Consequently, the hyper-viscoelastic material model used here cannot describe the material behaviour with proper accuracy.

4 Conclusions

A simple numerical algorithm has been adopted in order to determine the constitutive parameters of silicone rubber and polypropylene from uniaxial relaxation tests. The results show that the large strain viscoelastic model used is able to model the material behaviour of silicone rubber with proper accuracy. However, in case of polypropylene, the simulated material behaviour differs from the measured one significantly. As it is pointed out the polypropylene exhibits nonlinear viscoelastic behaviour which is not taken into consideration in the material model used.

References

[1] Brinson, H.F., Brinson, L.C., (2015). *Polymer Engineering Science and Viscoelasticity*, Springer.

[2] Bergström, J., (2015). *Mechanics of Solid Polymers: Theory and Computational Modeling*, Elsevier.

[3] Goh, S.M., Charalambides, M.N., Williams, J.G., (2004). Determination of the constitutive constants of non-linear viscoelastic materials, *Mech. Time-Dependent Mater.* 8, 255–268.

[4] Nandi, B., Dalrymple, T., Lapczyk, I. (2014). Importance of Capturing Non-linear Viscoelastic Material Behavior in Tire Rolling Simulations, *Meeting of the Tire Society*.

[5] Dalrymple, T. (2014). Calibration of Polypropylene.

[6] Rivlin, R.S. (1948). Large elastic deformations of isotropic materials I. Fundamental, *Philosophical Trans. of the Roy. Soc. of London, Series A, Mathematical and Physical Sciences*, 240 (822), 459–490.

[7] Yeoh, O. H. (1993). Some Forms of the Strain Energy Function for Rubber, *Rubber Chemistry and Technology*, 66 (5), 754–771.

[8] G.A. Holzapfel, G.A. (2000). *Nonlinear Solid Mechanics*. Wiley.

[9] Gamonpilas, C., McCuiston, R. (2012). A non-linear viscoelastic material constitutive model for polyurea, *Polymer*, 53, 3655–3658.

[10] Abaqus 6.14 Documentation: Dassault Systèmes, USA