

Non-linear Constitutive Model for the Oligocarbonate Polyurethane Material*

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Abstract The polyurethane, which was the subject of the constitutive research presented in the paper, was based on oligocarbonate diols Desmophen C2100 produced by Bayer®. The constitutive modelling was performed with a view to applying the material as the inlay of intervertebral disc prostheses. The polyurethane was assumed to be non-linearly visco-hyperelastic, isotropic and incompressible. The constitutive equation was derived from the postulated strain energy function. The elastic and rheological constants were identified on the basis of experimental tests, *i.e.* relaxation tests and monotonic uniaxial tests at two different strain rates, *i.e.* $\dot{\lambda} = 0.1 \text{ min}^{-1}$ and $\dot{\lambda} = 1.0 \text{ min}^{-1}$. The stiffness tensor was derived and introduced to Abaqus® finite element (FE) software in order to numerically validate the constitutive model. The results of the constants identification and numerical implementation show that the derived constitutive equation is fully adequate to model stress-strain behavior of the polyurethane material.

Keywords: Constitutive model; Viscoelastic material; Stress relaxation; Finite elements.

INTRODUCTION

Polymers have been utilised in biomedicine for many years now. They have been applied in such biomedical branches like tissue engineering^[1, 2], orthopaedics^[3, 4] or drug delivery^[5, 6]. Polyurethanes based on oligocarbonate diols are resistant to hydrolysis and oxidation. Moreover, they also exhibit high tensile strength and modulus^[7]. Polyurethane of that kind is one of the most widely tested biomaterial. It is believed to be the first biomedical polyurethane that found application in biomedical devices for its biostability. The fact that the polyurethane comprises carbonate linkages makes this polymer attractive in bio-applications where oxidation is a potential cause of degradation.

Polyurethanes based on oligocarbonate diols can be prepared by various methods:

- in the reaction of diols with phosgene or substances based on phosgene^[8],
- by the copolymerization of oxiranes with carbon dioxide^[9],
- by the polymerization of six- and seven-membered cyclic carbonates^[10],
- by the transesterification of dimethyl- or diethylcarbonate^[11] or alkylene carbonate^[12] with diols,
- by synthesis from five-membered cyclic carbonates and aliphatic diols using azeotropic solvents^[13].

The most popular method is the last but one, however, the last method has been recently proposed and studied (cf.^[13]).

The aim of the paper is to determine the constitutive behavior for the oligocarbonate polyurethane, which

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additionally was modified with hexamethylene diisocyanate and ethylene glycol as chain extenders and with glycerin as a netting agent. The model was then implemented into the finite element (FE) system Abaqus® by means of the user-supplied material code UMAT. The goal of FE simulation was to numerically validate the formulated constitutive law. As a result a ready-to-use in numerical FE analyses visco-elastic model was obtained. The model is unique and can be implemented to simulate the constitutive behavior of various structures made of the oligocarbonate polyurethane.

The research presented in the paper was carried out with a view to applying the polyurethane as one of the components of intervertebral disc prosthesis. Such a prosthesis usually consists of three parts, *i.e.* two metal plates and a plastic core inserted between them. Artificial intervertebral discs are experimentally tested mainly in compressive tests. Some experimental set-ups also include torsional and bending tests. The experiments provide information on the strength of disc prostheses and give the answer whether or not the tested prosthesis can be clinically applied from the mechanical point of view. The answer to that question can be also achieved from numerical simulation of the spine segment with implanted prosthesis. The reader can find some experimental tests performed on various prostheses in the literature, see *e.g.*^[14–16]. In the first paper the authors tested one of the most frequently applied intervertebral disc prostheses, *i.e.* SB Charité. They performed biomechanical static and dynamic compressive tests on two SB Charité prostheses. They reported three strain-stress characteristics in the form of hysteresis loops where the maximal loads corresponded to the stress values 9 MPa, 22 MPa and 40 MPa. The second paper provides the results of similar compressive tests that have been performed on a prosthesis of another type, namely nucleus-sparing prosthesis. The latter cited paper gives a review of clinical application and some biomechanical aspects of various types of intervertebral disc prostheses. The research delivered in the paper contained compressive tests of various types, which were performed in order to determine hyperelastic and viscoelastic constants of the material in interest and formulate a constitutive law for it.

METHOD

Theoretical Consideration

The approach of the constitutive modelling presented in the paper is based on application of a potential function form Ψ_e and is well described in literature^[17, 18]. Potential energy of elasticity per unit of volume is the physical interpretation of the function Ψ_e , which is also referred to as strain energy density or stored energy function. The energy must be invariant of the coordinate system transformation. Thus, potential energy has to depend on invariants of the tensors used to describe material behavior.

A general visco-hyperelastic constitutive law must take into consideration the principle of fading memory developed by Coleman and Noll^[19]. The principle states that the deformation occurred in the recent time history has greater influence on the actual state of stress than the deformation occurred in the more distant time history. Thus, a visco-hyperelastic model must take into account the deformation history. A mathematical description of the principle is formulated as follows:

$$\mathbf{S} = \mathbf{S}^e(\mathbf{C}(t)) + \int_0^t \Phi(\Gamma(t-s), s; \mathbf{C}(t)) ds \quad (1)$$

where $\mathbf{S}$ —second Piola-Kirchhoff stress tensor, $\mathbf{S}^e$ —elastic second Piola-Kirchhoff stress tensor, $\mathbf{C}$ —right Cauchy deformation tensor defined as $\mathbf{C} = \mathbf{F}^T \mathbf{F}$, $\mathbf{F}$ —gradient deformation tensor, Φ —a general tensor-valued function that depends on variables $\Gamma(t-s)$ and t . In equation (1) s represents the historical time variable, whereas t is the current time. The shape memory effects have been recently studied in aliphatic polyether urethane by Ling *et al.*^[20]. The authors investigated the relationships among recovery performance, material parameters and loading conditions. They simulated the shape memory behaviors numerically using a three-dimensional viscoelastic model.

Equation (1) represents a general constitutive law that takes into account hyperelastic and long-term memory contributions. In non-linear visco-hyperelasticity the constitutive model can be formulated by

convolution of strain-dependant function $S^e(\lambda)$ and a time-dependant function $g(t)$:

$$S(\lambda, t) = S^e(\lambda) \times g(t) \quad (2)$$

where S —second Piola-Kirchhoff stress tensor, $g(t) = g_\infty + \sum_{i=1}^n g_i \cdot \exp(-t/\tau_i)$, $g_\infty = 1 - \sum_{i=1}^n g_i$, g_i are characteristic amplitudes and τ_i —relaxation times. After some mathematical operations Eq. (2) can be presented in the form:

$$S(t) = g_\infty S^e(t) + \sum_{i=1}^n \int_0^t g_i \cdot \exp(-(t-s)/\tau_i) \cdot \partial S^e(s) / \partial s \, ds \quad (3)$$

The hyperelastic part of Eq. (3) is calculated using the formula:

$$S_{ij}^e = 2 \cdot \partial \Psi_e / \partial C_{ij} \quad (4)$$

The most known hyperelastic stored energy functions are: Mooney-Rivlin, Ogden, Neo-Hooke, Yeoh, Knowles, Da Silva Soares or Arruda-Boyce. None of those models, however, was adequate to simulate strain-stress behavior of the polyurethane in interest. In Fig. 1 curve fitting for the aforementioned models is shown. It can be clearly noticed that another strain energy function is needed. The presented curves were fitted to the strain-stress curve obtained from the monotonic uniaxial compression test. Strain, in particular nominal strain, is defined here by stretch ratio λ measured in the direction of load. It seems that the mentioned stored energy functions are not able to simulate hyperelastic behavior of the polyurethane material due to their simple mathematical form. It can be seen in Fig. 1 that the experimental stress-strain curve is complex, *i.e.* it has three inflection points. To fit the experimental curve a more sophisticated hyperelastic function was postulated.

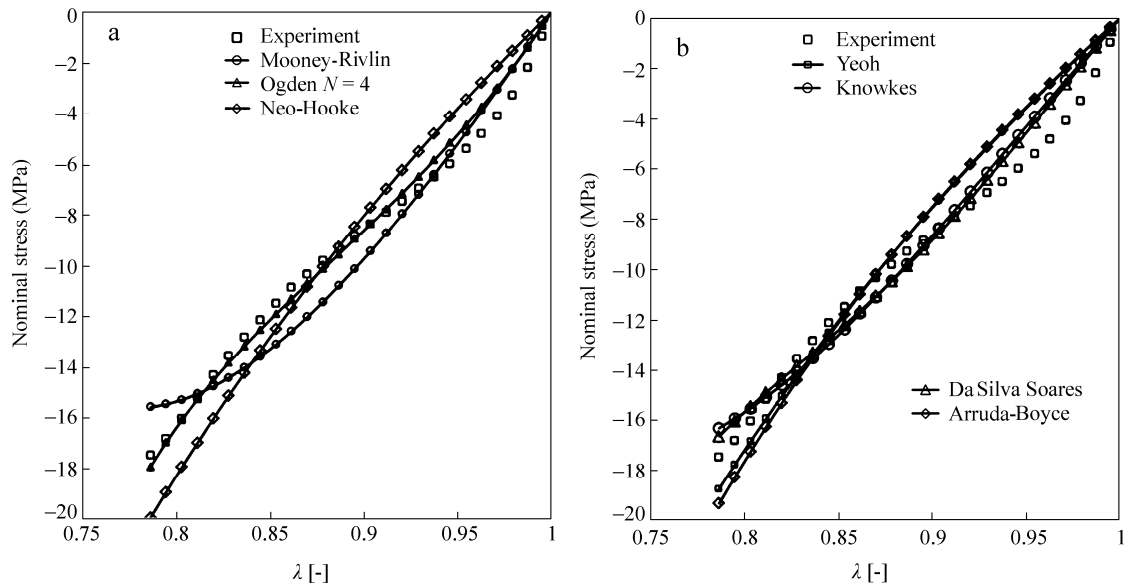


Fig. 1 Curve-fitting of various hyperelastic models to the experimental data

The strain energy function proposed in this paper, which described the polyurethane behavior sufficiently well, was formulated on the basis of hyperelastic function presented in [21]. It is of the following form:

$$\begin{aligned} \Psi_e = & 0.5 \cdot c_1 \cdot (I_1 - 3) + 0.5 \cdot c_2 / c_3 \cdot (I_1^{c_3} - 3^{c_3}) + 0.5 \cdot c_4 / c_5 \cdot (I_1^{c_5} - 3^{c_5}) + \\ & + 0.5 \cdot c_6 \cdot (I_2 - 3) + 0.5 \cdot c_7 / c_8 \cdot (I_2^{c_8} - 3^{c_8}) + 0.5 \cdot c_9 / c_{10} \cdot (I_2^{c_{10}} - 3^{c_{10}}) \end{aligned} \quad (5)$$

The function depends on both the first I_1 and the second I_2 invariants of the tensor $\mathbf{C}$ raised to the power to be determined from experiment. This fact makes the fitting of model (5) to the experimental curve very accurate. In addition, the influence of those invariants is assumed to be equal, although the authors of [21] considered their potential function with weight factors determining the contribution of the invariants separately. Function Ψ_e fulfils the requirements resulting from mathematical theory and the physical nature of solids, *i.e.* convexity, objectivity and material-frame indifference.

The constants c_i , $i = 1, \dots, 10$ are hyperelastic constants of the material. The curve fitting for the model (5) is presented in Fig. 2. It can be clearly seen that the fitting is actually perfect.

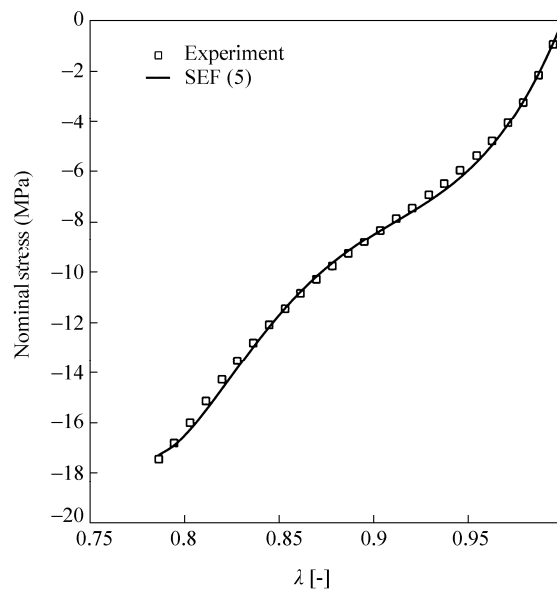


Fig. 2 Curve-fitting of model (5) to the experimental data

The main objective of the paper was to combine two theories to determine the long-term visco-hyperelastic constitutive law for the material using non-linear approach. The first theory was described in [22]. By means of this theory the hereditary integrals in Eq. (3) are numerically solved. The second theory implements the algorithm described in [23] and is applied in order to determine the number of characteristic times τ_i and corresponding characteristic amplitudes g_i . The code based on the algorithm, which is utilised in the studies, “decides” itself how many relaxation times τ_i are needed to describe the rheological response of the material of interest. The combination of the two theories allows one to formulate a constitutive model for the material which describes well its rheological behavior, *i.e.* relaxation phenomenon, dependence on strain rate, hysteresis loop. It has been already utilised in formulation of a constitutive law for another type of polyurethane^[24]. However, no implementation in FE simulation was presented in that paper. In addition, two conventional hyperelastic potential functions proved to be adequate to model the mechanical behavior of the material, *i.e.* Mooney-Rivlin and Ogden models.

Experimental Set-up

The experiments were carried out on a MTS[®] Bionix System. Four cylindrical samples were experimentally studied. The dimensions of the samples were: diameter $d = 11.7$ mm and height $h = 11.85$ mm. The samples were positioned between two metal plates (Fig. 3). In all the experiments the plates were kept lubricated in order

to minimise the friction and avoid barrel-like deformation of the samples. It was assumed that stress and strain are uniformly distributed in the samples. The deformation was measured by means of the MTS[®] strain-gauge extensometer. All the experiments were performed in the average temperature 20 °C. After each test every sample was left for 24 h to let stress relax in it.

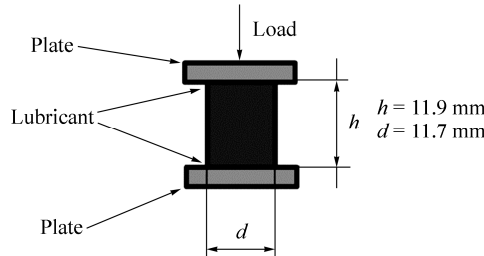


Fig. 3 Experimental set-up

The uniaxial monotonic tests were conducted in order to select the best hyperelastic model for the nanocomposite mechanical behavior. The samples were compressed by 2.5 mm at the constant strain rate $\dot{\lambda} = 10 \text{ min}^{-1}$. The experimental curves were then utilised in the process of curve-fitting and preliminary calibration of the constants. On the basis of those tests the proper form of stored energy function was found.

Next, relaxation tests and compression tests with loading and unloading phases were performed. In the relaxation process the samples were compressed from the initial deformation to the nominal strain 10%, which corresponded to the stress level -7.5 MPa , within approx. 1 sec with constant strain rate. The percentage of stress relaxation in the sample was equal approx. 50%. The relaxation tests performed for higher stress level, *i.e.* -18 MPa , indicated the same material behavior. The sample nominal strain was held fixed for 30 min. On the basis of relaxation tests the number n of relaxation times τ_i and corresponding characteristic amplitudes g_i were determined.

The loading/unloading compression tests were performed at two strain rates, *i.e.* $\dot{\lambda} = 0.1 \text{ min}^{-1}$ and $\dot{\lambda} = 1.0 \text{ min}^{-1}$. The samples were compressed by 2.5 mm, which corresponded to the nominal strain approx. 20%. The constants identification process was conducted for the two experimental curves at the same time. On the basis of the obtained curves the final values of hyperelastic constants c_i , $i = 1, \dots, 10$, and characteristic amplitudes g_i , $i = 1, \dots, n$, were calibrated. The values of the relaxation times were equal to those identified on the basis of relaxation tests. The whole experimental program is presented in Fig. 4.

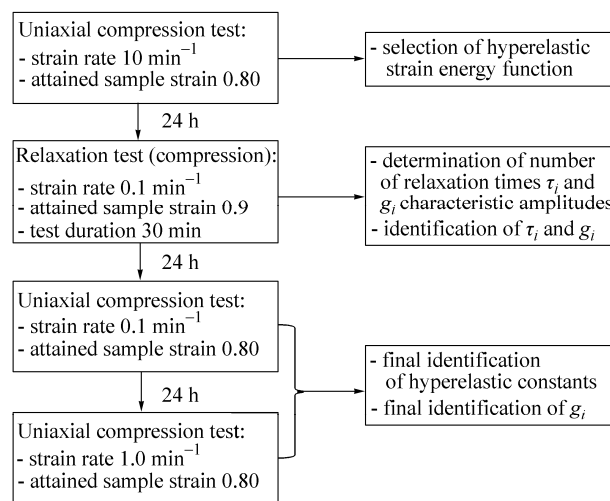


Fig. 4 Program of the experimental tests

Finite Element Implementation

In this research the polyurethane was modelled as a visco-hyperelastic, isotropic and incompressible material. The fact that the constitutive model was to be implemented in FE analyses requires that the potential function Ψ_e be split into volumetric part Θ and isochoric part $\bar{\Psi}$:

$$\Psi_e = \Theta + \bar{\Psi} \quad (6)$$

The volume-changing part was of the following form:

$$\Theta = 1/D \cdot (J - 1)^2 \quad (7)$$

where $1/D$ represents bulk modulus of the infinitesimal theory, J is the determinant of the deformation gradient tensor $\mathbf{F}$. In numerical simulations of incompressible materials the constant D is assumed to be very small making, thus, the bulk modulus tends to infinity. In the presented FE simulation D was 1×10^{-5} . In paper [25] the authors discussed various forms of volumetric potential functions. In the presented research the simplest possible form of the function Θ was assumed.

The volume-preserving part $\bar{\Psi}$ was assumed to be of the form of Eq. (5) with the difference that now it depends on the first $\bar{I}_1$ and second $\bar{I}_2$ invariants of the distortional part of the right Cauchy tensor $\bar{\mathbf{C}}$. Tensor $\bar{\mathbf{C}}$ is calculated using equation (8):

$$\bar{\mathbf{C}} = J^{-2/3} \mathbf{C} \quad (8)$$

The relation between $\bar{I}_1$ and I_1 , as well as that between $\bar{I}_2$ and I_2 , is of the form:

$$\bar{I}_1 = J^{-2/3} I_1, \quad \bar{I}_2 = J^{-2/3} I_2 \quad (9)$$

Now the elastic part of the stress $\mathbf{S}^e$ can be calculated:

$$\mathbf{S}^e = 2 \cdot \partial \Psi_e / \partial \mathbf{C} = J \cdot \partial \Theta / \partial J \cdot \mathbf{C}^{-1} + J^{-2/3} \text{DEV}(\bar{\mathbf{S}}) \quad (10)$$

where:

$$\text{DEV}(\bar{\mathbf{S}}) = \bar{\mathbf{S}} - 1/3 \cdot (\bar{\mathbf{S}} : \bar{\mathbf{C}}) \bar{\mathbf{C}}^{-1} \quad (11)$$

$$\bar{\mathbf{S}} = 2 \cdot \partial \bar{\Psi} / \partial \bar{\mathbf{C}} \quad (12)$$

In Eq. (11) the sign “:” denotes double contraction of two tensors.

The formula for the material stiffness fourth-order tensor $\mathbf{e}$ is as follows:

$$\mathbf{e} = 2 \cdot \partial \mathbf{S}^e / \partial \mathbf{C} = 2 \cdot \partial \left(J \cdot \Theta_J \cdot \mathbf{C}^{-1} + J^{-2/3} \left(\bar{\Psi}_{\bar{\mathbf{C}}} - 1/3 (\bar{\Psi}_{\bar{\mathbf{C}}} : \bar{\mathbf{C}}) \bar{\mathbf{C}}^{-1} \right) \right) / \partial \mathbf{C} \quad (13)$$

where $\Theta_J = \frac{\partial \Theta}{\partial J}$, $\bar{\Psi}_{\bar{\mathbf{C}}} = \partial \bar{\Psi} / \partial \bar{\mathbf{C}}$.

After implementation of the chain rule one obtains the following form of the uncoupled stiffness tensor $\mathbf{e}$:

$$\begin{aligned} \mathbf{e} = & 2 \cdot \Theta_J \cdot \mathbf{C}^{-1} \otimes J_{\mathbf{C}} + 2J \cdot \Theta_J \cdot \mathbf{C}^{-1} + 2J \cdot \Theta_{J,J} \cdot \mathbf{C}^{-1} \otimes J_{\mathbf{C}} + \\ & + 4J^{-2/3} \cdot \left(\bar{\Psi}_{\bar{\mathbf{C}},\bar{\mathbf{C}}} : \bar{\mathbf{C}}_{\mathbf{C}} - 1/3 \cdot \bar{\mathbf{C}}^{-1} \otimes (\bar{\Psi}_{\bar{\mathbf{C}}} : \bar{\mathbf{C}})_{\mathbf{C}} \right) - 4J^{-2/3} \cdot \left(1/3 \cdot (\bar{\Psi}_{\bar{\mathbf{C}}} : \bar{\mathbf{C}}) \mathbf{C}_{\mathbf{C}}^{-1} \right) + \\ & - 4/3 \cdot J^{-2/3} \cdot \left(\bar{\Psi}_{\bar{\mathbf{C}}} - 1/3 \cdot (\bar{\Psi}_{\bar{\mathbf{C}}} : \bar{\mathbf{C}}) \bar{\mathbf{C}}^{-1} \right) \otimes \mathbf{C}^{-1} \end{aligned} \quad (14)$$

where $J_{\mathbf{C}} = \frac{\partial J}{\partial \mathbf{C}}$, $\mathbf{C}_{\mathbf{C}}^{-1} = \frac{\partial \mathbf{C}^{-1}}{\partial \mathbf{C}}$, $\Theta_{J,J} = \frac{\partial^2 \Theta}{\partial J^2}$, $\bar{\Psi}_{\bar{\mathbf{C}},\bar{\mathbf{C}}} = \frac{\partial^2 \bar{\Psi}}{\partial \bar{\mathbf{C}}^2}$, $\bar{\mathbf{C}}_{\mathbf{C}} = \frac{\partial \bar{\mathbf{C}}}{\partial \mathbf{C}}$, $(\bar{\Psi}_{\bar{\mathbf{C}}} : \bar{\mathbf{C}})_{\mathbf{C}} = \frac{\partial (\bar{\Psi}_{\bar{\mathbf{C}}} : \bar{\mathbf{C}})}{\partial \mathbf{C}}$

The sign “ $\otimes$ ” in equation (14) represents the dyadic product of two tensors.

In the contemporary finite element software programs, including Abaqus[®], it is required that the Zaremba-

Jaumann objective stress rate, $\overset{\nabla}{\mathbf{T}}$, of Kirchhoff stress $\mathbf{T}$ is introduced:

$$\overset{\nabla}{\mathbf{T}} = \dot{\mathbf{T}} - \mathbf{W} \cdot \mathbf{T} - \mathbf{T} \cdot \mathbf{W}^T = \mathbf{e}^{T ZJ} : \mathbf{D} \quad (15)$$

where $\mathbf{W}$ and $\mathbf{D}$ are the symmetric and anti-symmetric parts of the spatial velocity gradient $\mathbf{L}$, respectively; $\mathbf{e}^{T ZJ}$ is the tangent modulus tensor for the Zaremba-Jaumann rate of the Kirchhoff stress. The components of the tensor $\mathbf{e}^{T ZJ}$ can be calculated by means of Eq. (16):

$$\mathbf{e}_{ijkl}^{T ZJ} = \mathbf{e}_{ijkl}^{Tc} + \delta_{ik} \mathbf{T}_{jl} + \delta_{jl} \mathbf{T}_{ik} \quad (16)$$

where δ_{ij} is the Kronecker delta. The tensor $\mathbf{e}_{ijkl}^{Tc}$ is associated with the convected rate of the Kirchhoff stress and is obtained by the use of the following transformation rule:

$$\mathbf{e}_{ijkl}^{Tc} = F_{ip} F_{jq} F_{kr} F_{ls} \mathbf{e}_{pqrs} \quad (17)$$

The components of $\mathbf{e}_{ijkl}^{T ZJ}$ were introduced in Abaqus[®] software by means of UMAT user-subroutine code. As the material in interest is modelled as non-linearly visco-hyperelastic, those components were multiplied by the viscous constituent g^v :

$$g^v = g_\infty + \sum_{i=1}^n g_i \cdot (1 - \exp(-\Delta t / \tau_i)) \cdot \tau_i / \Delta t \quad (18)$$

where Δt is a time step of numerical calculations. This results from the definition (13), Eq. (3) and the theory described in [24].

In addition, Cauchy stress was calculated using the formula:

$$\boldsymbol{\sigma} = 1/J \cdot \mathbf{F} \mathbf{S} \mathbf{F}^T \quad (19)$$

To make the set of needed equations complete an expression for Kirchhoff stress has to be introduced:

$$\mathbf{T} = J \boldsymbol{\sigma} \quad (20)$$

In Eq. (19) the second Piola-Kirchhoff stress tensor $\mathbf{S}$ (total stress) was calculated by means of relation (3) and the hyperelastic part of the stress $\mathbf{S}^e$, which has been introduced in Eq. (1), by means of Eq. (10). It has to emphasised, that the hereditary integral in Eq. (3) was determined using the theory presented in [23] and developed in [24]. According to the theory the integral in Eq. (3) can be computed using an algorithm which is based on finite increments of time. Thus, the stress Eq. (3) can be expressed in the form:

$$\begin{aligned} \mathbf{S}(t+1) = & g_\infty \mathbf{S}^e(t+1) + \\ & + \sum_{i=1}^n \left(\exp(-\Delta t / \tau_i) \cdot \mathbf{h}_i(t) + g_i \left(\frac{1 - \exp(-\Delta t / \tau_i)}{\Delta t / \tau_i} \right) (\mathbf{S}^e(t+1) - \mathbf{S}^e(t)) \right) \end{aligned} \quad (21)$$

where Δt is the time increment, $\mathbf{h}_i(t)$ represents the stress at the previous time step. As the initial stress and nominal strain in the material are known the stress at time $t > 0$ can be easily calculated.

RESULTS

Due to the fact that the obtained relaxation curves were practically overlapping it was decided that determination of the number n of relaxation times τ_i was performed on the basis of one relaxation test. The graphical representation of the curve-fitting is shown in Fig. 5. The number of relaxation times was determined by means of the algorithm presented in [24]. The algorithm is based on the iterative calibration of relaxation times and comprises the following steps:

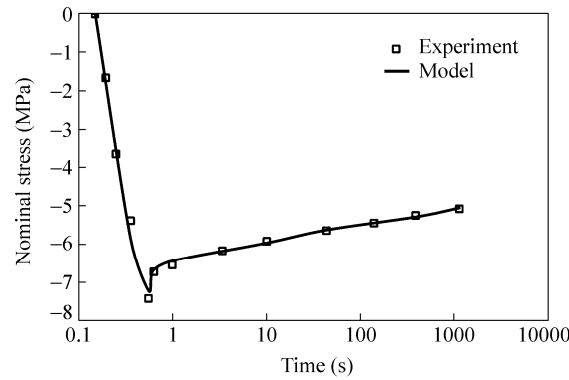


Fig. 5 Curve-fitting for relaxation test

— the range of relaxation times $[\tau_{\min}, \tau_{\max}]$ is determined such that $\tau_{\min}$ equals to the sampling rate and $\tau_{\max}$ is the overall time of the experiment;

— the interval $[\tau_{\min}, \tau_{\max}]$ is divided in n equal parts according to the pattern:

$$\tau_1^{(0)} = \tau_{\min}, \dots, \tau_i^{(0)} = \tau_{\min} 10^{(i-1)\Delta}, \dots, \tau_n^{(0)} = \tau_{\max} \quad (22)$$

$$\text{where } \Delta = \frac{\lg \frac{\tau_{\max}}{\tau_{\min}}}{n-1}$$

— minimisation of the distance between the theoretical curve and experimental one is carried out (curve fitting procedure);

— a new set of relaxation times τ_i^{h+1} for the next time step is determined following the rules: if a characteristic amplitude g_i^h , after curve fitting procedure, is lower than a given threshold the corresponding characteristic time τ_i^h is discarded whereas if g_i^h is higher than the threshold the corresponding relaxation time is left unchanged and in the vicinity new relaxation times are added, according to the formula:

$$\tau_{i-1}^{h+1} = \tau_i^h 10^{-\left(\frac{\Delta}{3}\right)^h}, \quad \tau_i^{h+1} = \tau_i^h, \quad \tau_{i+1}^{h+1} = \tau_i^h 10^{\left(\frac{\Delta}{3}\right)^h} \quad (22)$$

The starting point of the curve fitting problem in step $h+1$ is the solution calculated in the previous step h . The initial values of the constants g_{i-1}^{h+1} and g_{i+1}^{h+1} , corresponding to the new relaxation times, are set to zero. The last two steps are repeated either until all the scalar coefficients g_i exceed the threshold, or until the decrement of the objective function between the consecutive steps is acceptably low.

The values of the identified τ_i and corresponding g_i are presented in Table 1. The mean error was calculated by means of equation:

$$err_{\text{mean}} = \sum_{i=1}^{l_p} err_i / l_p \quad (23)$$

Table 1. Viscous constants calibrated on the basis of relaxation test

g_1 [-]	0.28	τ_1 [s]	0.24
g_2 [-]	0.016	τ_2 [s]	7.95
g_3 [-]	0.049	τ_3 [s]	20.79
g_4 [-]	0.019	τ_4 [s]	92.00
g_5 [-]	0.018	τ_5 [s]	407.09
g_6 [-]	0.075	τ_6 [s]	1801.32

where l_p is the number of measured points, err_i is the error calculated at every sampling point^[26]:

$$err_i = |S_i - \tilde{S}_i| / \max(0.5, |\tilde{S}_i|) \quad (24)$$

In Eq. (24) S_i is the second Piola-Kirchhoff stress component in the direction of load calculated at every sampling point by means of Eq. (3), $\tilde{S}_i$ represents values of the measured second Piola-Kirchhoff stress. The mean error of relaxation curve-fitting was equal to 0.5%.

On the basis of the relaxation curve also the preliminary values of the hyperelastic constants were identified. The values of c_i , $i = 1, \dots, 10$, and characteristic amplitudes g_i , $i = 1, \dots, 6$, determined at this stage of research were the initial values of those quantities in the process of curve-fitting performed in the next step. The final values of c_i and g_i were calibrated on the basis of monotonic uniaxial compression tests performed at two strain rates, i.e. $\dot{\lambda} = 0.1 \text{ min}^{-1}$ and $\dot{\lambda} = 1.0 \text{ min}^{-1}$. It has to be emphasised that the calibration was conducted for the two strain rates at the same time. The values of relaxation times were kept unchanged. The identification of c_i and g_i was performed for each sample. Then the mean values of the constants were calculated and the theoretical strain-stress curves were drawn. In Fig. 6 the final result of curve-fitting process for the mean values is presented. The final values of the visco-hyperelastic constants are shown in Table 2.

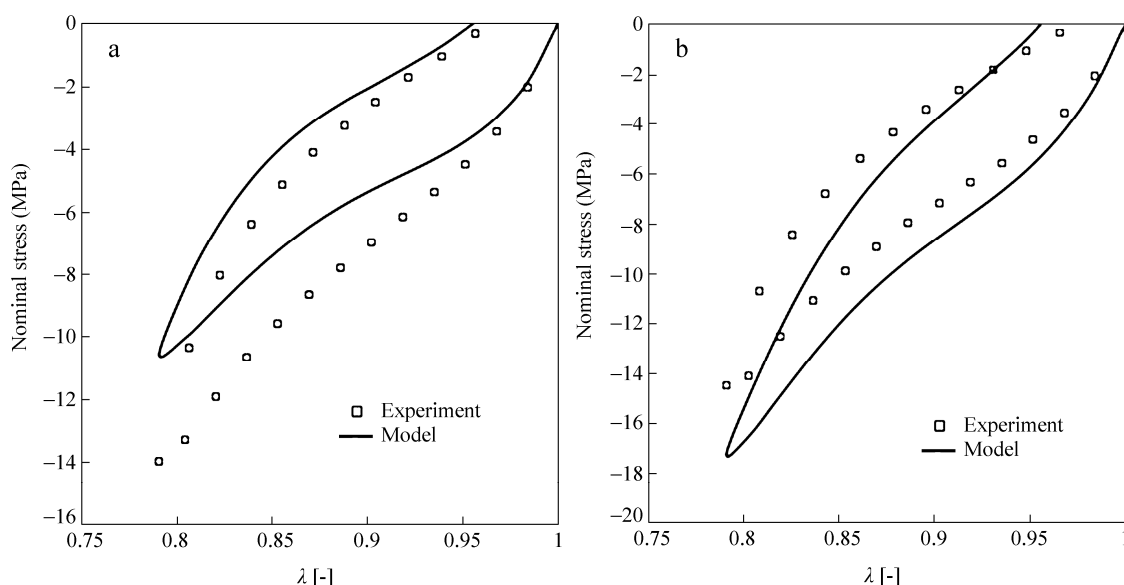
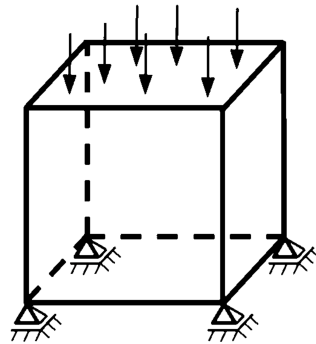
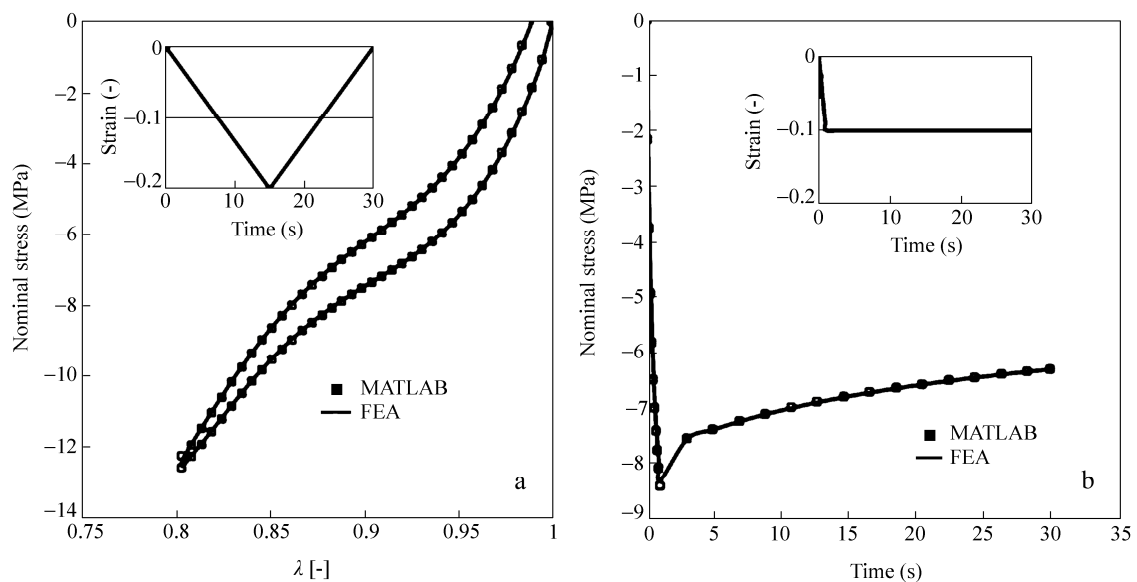


Fig. 6 Curve-fitting for the uniaxial monotonic compression tests: (a) $\dot{\lambda} = 0.1 \text{ min}^{-1}$ and (b) $\dot{\lambda} = 1 \text{ min}^{-1}$

The formulated constitutive model was implemented in FE software Abaqus[®]. FE simulations were performed on a cube $10 \text{ mm} \times 10 \text{ mm} \times 10 \text{ mm}$. The numerical model with the boundary conditions and kinematical load is shown in Fig. 7. In the first analysis the cube was compressed by 2 mm, which corresponded to the nominal strain 20%. In the second one stress relaxation process was simulated at the level of compressive nominal strain 10%. The third FE simulation consisted in compression by 1 mm followed by stretching by 1 mm and unloading phase. The nominal stresses calculated in FE simulations were compared to those calculated in MATLAB[®] by means of Eq. (3). The results of FE simulations are presented in Fig. 8. In Fig. 8(a) the material simulated response to compression of 2 mm is presented with the prediction by model (3) derived from the strain energy function (5). Figure 8(b) shows the simulated and predicted response to static load (stress relaxation). Finally, Fig. 8(c) shows the simulated and predicted behavior to different variants of stretching and compression occurring in a row.

Table 2. Final values of the visco-hyperelastic constants calibrated on the basis of uniaxial monotonic compression tests

	Sample 1	Sample 2	Sample 3	Sample 4	Mean values $\pm$ STD
c_1 [MPa]	-663.27	-663.70	-657.90	-660.88	-661.44 ± 2.66
c_2 [MPa]	11.53	11.53	11.49	11.55	11.52 ± 0.02
c_3 [-]	5.33	5.33	5.34	5.34	5.335 ± 0.005
c_4 [MPa]	2.81	2.82	2.73	2.90	2.81 ± 0.069
c_5 [-]	4.59	4.58	4.63	4.62	4.60 ± 0.023
c_6 [MPa]	-160.54	-160.16	-165.41	-155.87	-160.49 ± 3.90
c_7 [MPa]	-12.73	-12.74	-12.67	-12.66	-12.7 ± 0.04
c_8 [-]	5.30	5.29	5.29	5.30	5.295 ± 0.005
c_9 [MPa]	7.44	7.45	7.40	7.41	7.42 ± 0.023
c_{10} [-]	5.30	5.30	5.29	5.29	5.295 ± 0.005
g_1 [-]	0.299	0.293	0.400	0.302	0.323 ± 0.051
g_2 [-]	0.031	0.026	0.066	0.037	0.04 ± 0.017
g_3 [-]	0.067	0.064	0.074	0.079	0.071 ± 0.006
g_4 [-]	0.045	0.056	0.144	0.094	0.084 ± 0.044
g_5 [-]	0.104	0.119	0.165	0.148	0.134 ± 0.027
g_6 [-]	0.370	0.429	0.376	0.408	0.395 ± 0.027

**Fig. 7** Numerical model adapted to FE simulations in Abaqus®

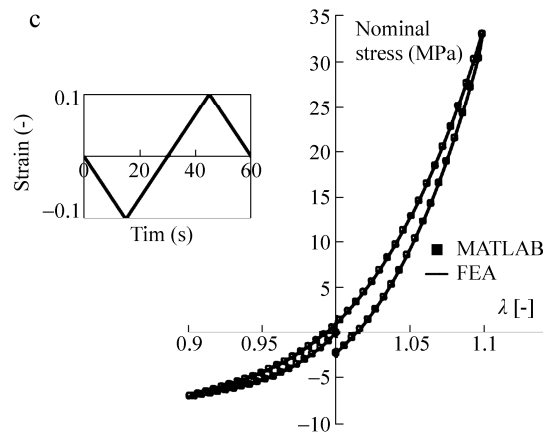


Fig. 8 Comparison of the predicted and simulated material response to (a) compression 2 mm, (b) stress relaxation and (c) compression 1 mm followed by stretch 1 mm and unload phase

CONCLUSIONS

In the course of constitutive law formulation no particular number of relaxation times was assumed. The number was determined on the basis of relaxation tests. The match of theoretical curve to the experimental one was almost perfect. Considering the fact that the constants identification was performed for the two strain rates at the same time and that the range of the strain rates was relatively high, the curve match based on monotonic compression tests was also good. The mean curve fitting error for the strain rate $\dot{\lambda} = 0.1 \text{ min}^{-1}$ was approx. 20%, while the error for the strain rate $\dot{\lambda} = 1 \text{ min}^{-1}$ equalled approx. 25%.

One of the advantages of the approach presented in the paper is that the number of the relaxation times and the characteristic constants in the Prony series is determined from best theoretical relaxation curve fitting. The formulated constitutive law is valid for a wide range of strain rates, takes into account long-term visco-elastic effects and models also the hysteresis loop. The material in the study is considered as incompressible and isotropic. This is a typical assumption attributed to polymer materials.

The results indicate that combination of the theory to calculate numerically the hereditary integral and that incorporating the algorithm of relaxation times determination works fine. The curve match for the two strain rates is good considering the fact that the hysteresis loop was simulated. The combination allows one to formulate a constitutive law that describes the relaxation phenomenon and is valid for quite a wide strain rate range.

Implementation of the formulated constitutive model to FE analyses is crucial, particularly in terms of the material in interest application as one of the components of intervertebral disc prosthesis. Thanks to FE simulations presumptive flaws of the prosthesis can be eliminated^[27–29]. Thus, material behavior simulation is of great importance. The results presented in Fig. 8 proved that the finite element implementation of the derived constitutive law was correct.

The researches performed on prostheses of intervertebral discs (see Introduction) and the constitutive studies presented in the paper suggest that the polyurethane has proper mechanical properties to be applied as the core of those prostheses. The authors of [15] showed that the axial stiffness of the modular intervertebral prosthetic disc (IPD; Dynamic Spine Inc., San Diego, CA, USA) was between 0.38 and 1.02 kN/mm at low and high loads, respectively. On the other hand, the studies described in [14] indicate that the axial stiffness of another intervertebral disc prosthesis, *i.e.* SB Charité, was from 1 to 4 kN/mm. Knowing the dimensions of the samples studied in the paper and the stress-strain behavior of the polyurethane material one can calculate axial stiffness of it. The results show that it is equal to 0.9 kN/mm.

The approach of the constitutive equation formulation presented in the paper is based on application of a potential function form Ψ_e . It is a very convenient approach as it allows one to take into consideration various phenomena, including those occurring in micro scale. In such the cases the use of the higher gradient approach^[30, 31] will be very helpful.

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