

A QUASI-LINEAR VISCOELASTIC RHEOLOGICAL MODEL FOR THERMOPLASTICS AND RESINS

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A new rheological model for polymeric materials has been proposed. The model is based on the concept of utilizing the Knowles stored-energy potential within the framework of quasi-linear viscoelasticity theory. The quasi-linear viscoelastic constitutive equation in its general form has been formulated using the formalism of the internal state variables. The developed constitutive equation allows for capturing the nonlinear-viscoelastic behavior of many polymeric materials such as thermoplastics or resins. The model has been implemented into a FE system. An application of the developed constitutive equation to modeling of the short-term, dissipative response of polyethylene has been presented.

Key words: polymers, rheology, constitutive equation, quasi-linear viscoelasticity, finite element method

Notations

\mathbf{B}	– left Cauchy-Green (C-G) deformation tensor
$\overline{\mathbf{B}}$	– isochoric left C-G deformation tensor
\mathbf{C}	– right C-G deformation tensor
$\overline{\mathbf{C}}$	– isochoric right C-G deformation tensor
\mathbf{c}	– elasticity tensor
\mathbf{c}^{ve}	– viscoelastic material tensor
$\mathbf{c}^{\tau c}$	– material tensor related to convected stress rate
\mathbf{c}^{MZ-J}	– material tensor used by Abaqus
\mathbf{D}	– strain rate tensor
\mathbf{E}	– Green strain tensor
\mathbf{e}_k	– unit vector of a Cartesian base, $k = 1, 2, 3$
\mathbf{F}	– deformation gradient tensor
$\overline{\mathbf{F}}$	– isochoric deformation gradient tensor
\mathbf{H}_k	– viscoelastic stress tensor $k = 1, 2, \dots, N$
\mathbf{I}	– fourth order identity tensor
$\mathbf{I}_{\mathbf{C}-1}$	– fourth order identity tensor in reference configuration
G	– reduced relaxation function
g_∞, g_k	– relaxation coefficients, $k = 1, 2, \dots, N$
I_k	– algebraic invariants of right C-G deformation tensor, $k = 1, 2, 3$
\overline{I}_k	– algebraic invariants of isochoric right C-G deformation tensor, $k = 1, 2, 3$
J	– Jacobian determinant
\mathbf{L}	– velocity gradient tensor
\mathbf{Q}	– orthogonal tensor
\mathbf{S}	– second Piola-Kirchhoff (P-K) total stress tensor
$\overline{\mathbf{S}}$	– auxiliary second P-K stress tensor

\mathbf{S}^e	– second P-K elastic stress tensor
t	– time since loading
U	– volumetric stored elastic energy potential
\mathbf{W}	– spin tensor
W	– stored elastic energy potential
\overline{W}	– isochoric stored elastic energy potential
λ_k	– stretch ratio in k -th direction, $k = 1, 2, 3$
$\boldsymbol{\tau}$	– Kirchhoff stress tensor
$\boldsymbol{\sigma}$	– Cauchy stress tensor
τ	– time variable
τ_k	– k -th relaxation time, $k = 1, 2, \dots, N$
$\mathbf{1}, \delta_{ij}$	– second order identity tensor in absolute and indicial notation
$\text{DEV}[\bullet]$	– operator extracting deviatoric part of tensor in reference configuration
$\text{tr}(\bullet)$	– trace operator
$(\bullet)^T$	– transpose operator
$(\bullet)^{-1}$	– inversion operator
\cdot	– double contraction operator
\otimes	– dyadic product operator
$(\bullet)^\nabla$	– Zaremba-Jaumann (Z-J) objective rate operator

1. Introduction

The polymeric materials find increasingly large number of applications in such fields as aeronautics, automotive industry and biomedical engineering for instance. The experimental results show that polymers exhibit rheological effects such as creep, relaxation, damping and strain rate sensitivity (see Garbarski, 2001).

The theory of linear elasticity has a limited validity for the polymeric materials. In the case of small strains and static loadings, the constitutive relations of linear elasticity can be used in engineering analysis, however, with several restrictions. A certain life span has to be assumed for the construction being designed. Basing on the assumed life span, adequate long-term values of the engineering moduli should be used for calculations. The described approach is admissible in the case of small strains and static loadings, nevertheless it fails for large strains and dynamic loadings when damping and strain rate sensitivity becomes a significant factor.

The most popular theory which allows for taking the rheological effects into account is the linear viscoelasticity based on Boltzmann superposition principle, e.g. Wilczyński (1984). However, it is well known that Boltzmann principle is valid in the case of small strains only. Usually, after reaching the strain level of 0.75%-2% the material behavior becomes nonlinear viscoelastic.

Throughout the years, numerous modifications of Boltzmann superposition principle have been proposed, e.g. Christensen (1971). The developed theories were mostly used for modeling of elastomeric polymers or soft biological tissues. Fung (1981) has introduced the popular theory of quasi-linear viscoelasticity (QLV) destined to model the viscoelastic properties of living tissues such as muscles, ligaments or tendons. A common approach in numerical implementation of QLV is defining stress-like internal state variables along with their evolution equations (see Puso and Weiss, 1998; Goh *et al.*, 2004). The evolution equations usually correspond to the Maxwell rheological model. A similar theory has been used by Kaliske (2000) to model the viscoelastic properties of elastomeric composites and human eye tissue.

Simo (1987) has developed a theory similar to QLV which takes into account the damage effects and neglects the volumetric viscoelasticity. The equations determining the evolution of

the viscoelastic stresses correspond to the generalized Maxwell rheological model. This particular theory, although without the component responsible for damage modeling, enjoys considerable popularity and has been used to describe the viscoelastic response of soft tissues, e.g. Peña *et al.* (2011). Holzapfel (2010) postulated using a slightly different set of viscoelastic state variables and evolution equations. The constitutive equations of that type have been used to model both elastomeric polymers and soft tissues.

An alternative approach involving split of the deformation gradient into viscous and elastic parts were used by Reese and Govindjee (1998) to model the viscoelastic response of vulcanized natural rubber.

Many constitutive models, originally formulated to model large strain nonlinear viscoelasticity of elastomeric polymers or soft biological tissues, are not able to accurately capture the strongly nonlinear viscoelastic response of thermoplastics and thermohardening polymers exhibited in their elastic regions (0-5% stretch). Thus, in this study, the framework of the QLV theory is used to formulate a new rheological model able to capture the nonlinear viscoelastic behavior of thermoplastics and resins. The developed model has been implemented into the FE system Abaqus.

2. Nonlinear elastic model

A hyperelastic material, also called a Green elastic material, is an elastic material which possesses a stored elastic energy potential function $W = W(\mathbf{C})$, e.g. Ogden (1997). In the case of isotropic hyperelastic materials, the stored-energy function must be invariant with respect to a rotation \mathbf{Q} , i.e.

$$W(\mathbf{C}) = W(\mathbf{Q}\mathbf{C}\mathbf{Q}^T) \quad (2.1)$$

This requirement is satisfied by defining the stored-energy potential as a function of three algebraic invariants of the right Cauchy-Green (C-G) tensor \mathbf{C}

$$W(\mathbf{C}) = W(I_1, I_2, I_3) \quad (2.2)$$

where the invariants are specified by the following formulas

$$I_1 = \text{tr } \mathbf{C} \quad I_2 = \frac{1}{2} \left((\text{tr } \mathbf{C})^2 - \text{tr } \mathbf{C}^2 \right) \quad I_3 = J^2 = \det \mathbf{C} \quad (2.3)$$

The second Piola-Kirchhoff (P-K) stress tensor \mathbf{S}^e relative to the reference configuration

$$\mathbf{S}^e = \frac{\partial W}{\partial \mathbf{E}} = 2 \frac{\partial W}{\partial \mathbf{C}} \quad (2.4)$$

and the strain-dependent elasticity tensor

$$\mathbf{C} = 4 \frac{\partial^2 W}{\partial \mathbf{C} \partial \mathbf{C}} \quad (2.5)$$

By inserting Eq. (2.2) into Eq. (2.4) one obtains

$$\mathbf{S}^e = 2 \left(\frac{\partial W}{\partial I_1} + I_1 \frac{\partial W}{\partial I_2} \right) \mathbf{1} - 2 \frac{\partial W}{\partial I_2} \mathbf{C} + 2 I_3 \frac{\partial W}{\partial I_3} \mathbf{C}^{-1} \quad (2.6)$$

which is the general form of the constitutive equation corresponding to the case of material isotropy.

In terms of FEM, it is profitable if the volumetric and isochoric responses are decoupled within the constitutive equation. The decoupling of the material response is facilitated by a multiplicative decomposition of the deformation gradient $\mathbf{F} = (J^{1/3}\mathbf{1})\bar{\mathbf{F}}$, where $J^{1/3}\mathbf{1}$ and $\bar{\mathbf{F}}$ are volumetric and deviatoric components, respectively. Consequently, the isochoric right C-G tensor can be defined as $\bar{\mathbf{C}} = \bar{\mathbf{F}}^T \bar{\mathbf{F}}$ and the associated basis of irreducible invariants takes the form, cf Holzapfel (2010)

$$\bar{I}_1 = \text{tr } \bar{\mathbf{C}} \quad \bar{I}_2 = \frac{1}{2} \left((\text{tr } \bar{\mathbf{C}})^2 - \text{tr } \bar{\mathbf{C}}^2 \right) \quad \bar{I}_3 = \det \bar{\mathbf{C}} = 1 \quad (2.7)$$

The following decoupled stored-energy potential is assumed

$$W(\mathbf{C}) = U(J) + \bar{W}(\bar{\mathbf{C}}) \quad (2.8)$$

where U is the volumetric component whereas \bar{W} states for the isochoric part of the stored-energy potential. By substituting Eq. (2.8) into Eq. (2.4) and using the chain rule, the general form of the decoupled constitutive equation is obtained, i.e.

$$\mathbf{S}^e = J \frac{\partial U}{\partial J} \mathbf{C}^{-1} + J^{-\frac{2}{3}} \text{DEV} [\bar{\mathbf{S}}] \quad (2.9)$$

where

$$\begin{aligned} \text{DEV} [\bar{\mathbf{S}}] &= \bar{\mathbf{S}} - \frac{1}{3} (\bar{\mathbf{S}} \cdot \bar{\mathbf{C}}) \bar{\mathbf{C}}^{-1} \\ \bar{\mathbf{S}} &= 2 \left(\frac{\partial \bar{W}}{\partial \bar{I}_1} + \bar{I}_1 \frac{\partial \bar{W}}{\partial \bar{I}_2} \right) \mathbf{1} - 2 \frac{\partial \bar{W}}{\partial \bar{I}_2} \bar{\mathbf{C}} \end{aligned} \quad (2.10)$$

In addition to the constitutive relation given by Eq. (2.9), an expression for the fourth-order elasticity tensor is required in order to form a finite element stiffness matrix. After inserting Eq. (2.8) into Eq. (2.5), a systematic use of the chain rule leads to the following expression for the elasticity tensor associated with the decoupled stored-energy potential (see Suchocki, 2011)

$$\begin{aligned} \mathbf{c} &= J \frac{\partial U}{\partial J} (\mathbf{C}^{-1} \otimes \mathbf{C}^{-1} - 2\mathbf{I}_{\mathbf{C}^{-1}}) + J^2 \frac{\partial^2 U}{\partial J^2} \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} - \frac{4}{3} J^{-\frac{4}{3}} \left(\frac{\partial \bar{W}}{\partial \bar{\mathbf{C}}} \otimes \bar{\mathbf{C}}^{-1} + \bar{\mathbf{C}}^{-1} \otimes \frac{\partial \bar{W}}{\partial \bar{\mathbf{C}}} \right) \\ &\quad + \frac{4}{3} J^{-\frac{4}{3}} \left(\frac{\partial \bar{W}}{\partial \bar{\mathbf{C}}} \cdot \bar{\mathbf{C}} \right) \left(J^{\frac{4}{3}} \mathbf{I}_{\mathbf{C}^{-1}} + \frac{1}{3} \bar{\mathbf{C}}^{-1} \otimes \bar{\mathbf{C}}^{-1} \right) + J^{-\frac{4}{3}} \bar{\mathbf{c}}_{\bar{W}} \end{aligned} \quad (2.11)$$

where $(\mathbf{I}_{\mathbf{C}^{-1}})_{ijkl} = \frac{1}{2} (C_{ik}^{-1} C_{jl}^{-1} + C_{il}^{-1} C_{jk}^{-1})$ and $\bar{\mathbf{c}}_{\bar{W}}$ is the part of \mathbf{c} which arises directly from the second derivatives of \bar{W} with respect to $\bar{\mathbf{C}}$, i.e.

$$\bar{\mathbf{c}}_{\bar{W}} = 4 \frac{\partial^2 \bar{W}}{\partial \bar{\mathbf{C}} \partial \bar{\mathbf{C}}} - \frac{4}{3} \left[\left(\frac{\partial^2 \bar{W}}{\partial \bar{\mathbf{C}} \partial \bar{\mathbf{C}}} \cdot \bar{\mathbf{C}} \right) \otimes \bar{\mathbf{C}}^{-1} + \bar{\mathbf{C}}^{-1} \otimes \left(\bar{\mathbf{C}} \cdot \frac{\partial^2 \bar{W}}{\partial \bar{\mathbf{C}} \partial \bar{\mathbf{C}}} \right) \right] + \frac{4}{9} \left(\bar{\mathbf{C}} \cdot \frac{\partial^2 \bar{W}}{\partial \bar{\mathbf{C}} \partial \bar{\mathbf{C}}} \cdot \bar{\mathbf{C}} \right) \bar{\mathbf{C}}^{-1} \otimes \bar{\mathbf{C}}^{-1} \quad (2.12)$$

Further specifications can be found after assuming a certain form of the decoupled stored-energy function.

3. New rheological model for polymers

The QLV theory introduced by Fung (1981) is a special case of nonlinear viscoelasticity with a simplifying assumption that the integrand of the convolution integral is split into a time and a strain dependent part, i.e.

$$\mathbf{S}(t) = \int_0^t G(t-\tau) \frac{\partial \mathbf{S}^e[\mathbf{E}(\tau)]}{\partial \tau} d\tau \quad (3.1)$$

where G is a scalar reduced relaxation function and \mathbf{S}^e is a nonlinear elastic term defined by Eq. (2.4). The constitutive equation is called “quasi-linear” since the term \mathbf{S}^e plays the role assumed by the strain in the linear theory of viscoelasticity, cf Christensen (1971).

It is assumed that G is a decreasing function of time and the word “reduced” refers to the condition $G = 1$ for $t = 0$. Alternatively, a fourth order reduced relaxation tensor can be utilized to obtain a model taking into account direction-dependent relaxation phenomena (see Fung, 1981).

A series of exponentials, also known as the Prony series, is a commonly used relaxation function

$$G(t) = g_\infty + \sum_{j=1}^N g_j e^{-\frac{t}{\tau_j}} \tag{3.2}$$

where g_j and τ_j ($j = 1, 2, \dots, N$) are the relaxation coefficients and relaxation times, respectively, whereas g_∞ determines the long-term response.

It can be proved that assuming a Prony series as $G(t)$ makes the QLV constitutive equation equivalent to a generalized Maxwell model formulated by means of the internal state variables (Fig. 1).

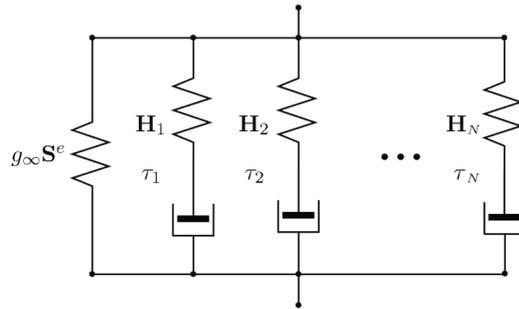


Fig. 1. Mechanical scheme of the rheological model

After introducing N stress-like variables \mathbf{H}_j ($j = 1, 2, \dots, N$), the total stress can be expressed as

$$\mathbf{S}(t) = g_\infty \mathbf{S}^e(t) + \sum_{j=1}^N \mathbf{H}_j(t) \tag{3.3}$$

where the evolution of the j -th viscoelastic stress is governed by the equation

$$\dot{\mathbf{H}}_j + \frac{1}{\tau_j} \mathbf{H}_j = g_j \dot{\mathbf{S}}^e \tag{3.4}$$

which follows from the response of the corresponding Maxwell element. The internal state variables have a physical meaning of non-equilibrium viscoelastic over stresses which slowly decrease under a constant loading.

After time-integrating Eq. (3.4), the following result is obtained

$$\mathbf{H}_j(t) = \int_0^t g_j e^{-\frac{t-\tau}{\tau_j}} \dot{\mathbf{S}}^e d\tau \tag{3.5}$$

By substituting Eq. (3.5) into Eq. (3.3), the integral form of the constitutive equation is recovered. Thus it can be seen that Eqs (3.3) and (3.4) are entirely consistent with Eqs. (3.1) and (3.2).

A computational algorithm developed by Taylor et al. (1970) may be used for discretization of Eqs (3.3) and (3.5). For the time instant $t_{n+1} = t_n + \Delta t$, corresponding to the increment $n + 1$, Eq. (3.3) takes the form

$$\mathbf{S}_{n+1} = g_\infty \mathbf{S}_{n+1}^e + \sum_{j=1}^N \mathbf{H}_{j\ n+1} \quad (3.6)$$

After assuming that for a sufficiently small time step Δt the time derivative $\dot{\mathbf{S}}^e$ in Eq. (3.5) can be approximated by a finite difference, i.e.

$$\dot{\mathbf{S}}_{n+1}^e \approx \frac{\mathbf{S}_{n+1}^e - \mathbf{S}_n^e}{\Delta t} \quad (3.7)$$

a recurrence formula for calculating viscoelastic stresses is obtained

$$\mathbf{H}_{j\ n+1} = e^{-\frac{\Delta t}{\tau_j}} \mathbf{H}_{j\ n} + g_j \frac{1 - e^{-\frac{\Delta t}{\tau_j}}}{\frac{\Delta t}{\tau_j}} (\mathbf{S}_{n+1}^e - \mathbf{S}_n^e) \quad (3.8)$$

which requires the quantities $\mathbf{H}_{j\ n}$ ($j = 1, 2, \dots, N$) and \mathbf{S}_n^e to be stored in memory for computation of these quantities in the next time step.

The viscoelastic material stiffness tensor in the time increment $n + 1$ takes the following form

$$\mathbf{C}_{n+1}^{ve} = 2 \frac{\partial \mathbf{S}_{n+1}}{\partial \mathbf{C}_{n+1}} \quad (3.9)$$

After inserting Eqs (3.6) and (3.8) into Eq. (3.9), one obtains

$$\mathbf{C}_{n+1}^{ve} = \left\{ g_\infty + \sum_{j=1}^N g_j \frac{1 - e^{-\frac{\Delta t}{\tau_j}}}{\frac{\Delta t}{\tau_j}} \right\} \mathbf{C}_{n+1} \quad (3.10)$$

where

$$\mathbf{C}_{n+1} = 2 \frac{\partial \mathbf{S}_{n+1}^e}{\partial \mathbf{C}_{n+1}} \quad (3.11)$$

is the elasticity tensor. It can be seen that the material stiffness tensor corresponding to the QLV model is simply an elasticity tensor multiplied by a proper scalar value. A specific form of the constitutive equation is determined by the choice of the potential function.

Numerous stored-energy potentials have been proposed over the years, however most of them, such as Neo-Hooke, Mooney-Rivlin or Ogden for instance, are destined to model the large-strain elastic response of rubbery materials. Those potentials fail to capture the strongly nonlinear stress-strain relation in the elastic region of thermoplastics and thermohardening polymers (0-5%), cf Suchocki (2011). For that purpose several researchers have proposed various, alternative stored-energy potentials.

Murnagham material model has been used to capture the nonlinear elasticity for small and moderate strains, e.g. Lurie (1990). The Murnagham stored-energy potential uses five material parameters and all three invariants of the right C-G tensor. Bouchart (2008) proposed using Ciarlet-Geymonat stored-energy function in order to model the elastic response of polypropylene. This model uses four material constants. Again, all three invariants of the right C-G deformation tensor were utilized. It appears that Knowles stored-energy function (see Knowles and Sternberg, 1980), used by Soares and Rajagopal (2010) to model polylactide, is particularly interesting due to its simplicity. The model by Knowles uses three material constants but

only one invariant (first) of the right C-G deformation tensor which simplifies the fourth order elasticity tensor and the process of material parameter identification.

The stored-energy function by Knowles was originally formulated in a coupled form, which makes sense for incompressible materials only, cf Knowles and Sternberg (1980). Here, it is presented in a modified, decoupled form which enables modeling of both incompressible and compressible materials, cf Suchocki (2011), and is suitable for FE implementation. It has the following form

$$W = \underbrace{\frac{\mu}{2b} \left\{ \left[1 + \frac{b}{\kappa} (\bar{I}_1 - 3) \right]^\kappa - 1 \right\}}_{\bar{W}(\bar{\mathbf{C}})} + \underbrace{\frac{1}{D_1} (J - 1)^2}_{U(J)} \quad (3.12)$$

where μ is a shear modulus, κ is a “hardening” parameter (the material hardens or softens according as $\kappa > 1$ or $\kappa < 1$), b is an additional parameter which improves curve-fitting and D_1 is the inverse of a bulk modulus.

In order to derive the uncoupled constitutive relation corresponding to the modified Knowles potential, the following derivatives are required

$$\frac{\partial \bar{W}}{\partial \bar{I}_1} = \frac{\mu}{2} \left[1 + \frac{b}{\kappa} (\bar{I}_1 - 3) \right]^{\kappa-1} \quad \frac{\partial \bar{W}}{\partial \bar{I}_2} = 0 \quad \frac{\partial U}{\partial J} = \frac{2}{D_1} (J - 1) \quad (3.13)$$

Inserting Eqs (3.13) into Eqs (2.10) gives

$$\bar{\mathbf{S}} = \mu \left[1 + \frac{b}{\kappa} (\bar{I}_1 - 3) \right]^{\kappa-1} \mathbf{1} \quad (3.14)$$

and

$$\text{DEV}[\bar{\mathbf{S}}] = \mu \left[1 + \frac{b}{\kappa} (\bar{I}_1 - 3) \right]^{\kappa-1} \left(J^{-\frac{2}{3}} \mathbf{1} - \frac{1}{3} \bar{I}_1 \mathbf{C}^{-1} \right) \quad (3.15)$$

where the results $\mathbf{1} \cdot \bar{\mathbf{C}} = \text{tr} \bar{\mathbf{C}} = \bar{I}_1$ and $J^{-2/3} \bar{\mathbf{C}}^{-1} = \mathbf{C}^{-1}$ have been used. Substituting Eqs (3.13)₃ and (3.15) into Eq. (2.9), one obtains the formula for the elastic stress

$$\mathbf{S}^e = \frac{2}{D_1} J (J - 1) \mathbf{C}^{-1} + \mu \left[1 + \frac{b}{\kappa} (\bar{I}_1 - 3) \right]^{\kappa-1} \left(J^{-\frac{2}{3}} \mathbf{1} - \frac{1}{3} \bar{I}_1 \mathbf{C}^{-1} \right) \quad (3.16)$$

The new rheological model for polymers is formulated through the following assumptions:

- The polymer is an isotropic, nonlinear viscoelastic body whose rheological properties are described using QLV theory, i.e. Eq. (3.1).
- The elastic stresses are described by Eq. (3.16), corresponding to the modified Knowles stored-energy potential.
- A Prony series given by Eq. (3.2) is used as the relaxation function.

Summing up, the developed model is described by four elasticity constants and $N + 1$ viscoelasticity constants, of which N are independent.

4. Finite element implementation

In order to implement a constitutive equation into a FE system, the equation must be linearized, which means that a nonlinear constitutive equation must be replaced with a linear relation between finite increments.

This is achieved by taking a material time derivative on Eq. (3.6) and replacing the time derivatives with the finite increments. Thus, for the $n + 1$ time increment

$$\Delta \mathbf{S}_{n+1} = \mathbf{C}_{n+1}^{ve} \cdot \frac{1}{2} \Delta \mathbf{C}_{n+1} \quad (4.1)$$

where the viscoelastic material tensor is given by Eqs (3.10) and (3.11). Subsequently, Eq. (4.1) must be expressed using the Zaremba-Jaumann (Z-J) objective rate of the Kirchhoff stress $\boldsymbol{\tau}$.

The incremental constitutive rate equation takes the form

$$\boldsymbol{\tau}_{n+1}^{\nabla} = J_{n+1} \mathbf{C}_{n+1}^{MZ-J} \cdot \Delta \mathbf{D}_{n+1} \quad (4.2)$$

where the incremental Z-J objective rate of the Kirchhoff stress

$$\boldsymbol{\tau}_{n+1}^{\nabla} = \Delta \boldsymbol{\tau}_{n+1} - \Delta \mathbf{W}_{n+1} \boldsymbol{\tau}_{n+1} - \boldsymbol{\tau}_{n+1} \Delta \mathbf{W}_{n+1}^T \quad (4.3)$$

and

$$\begin{aligned} \Delta \mathbf{W}_{n+1} &= \frac{1}{2} [\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1} - (\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1})^T] \\ \Delta \mathbf{D}_{n+1} &= \frac{1}{2} [\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1} + (\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1})^T] \\ \Delta \mathbf{F}_{n+1} &= \mathbf{F}_{n+1} \mathbf{F}_n^{-1} \end{aligned} \quad (4.4)$$

The material stiffness tensor takes the corresponding form

$$\mathbf{C}_{n+1}^{MZ-J} = \frac{1}{J_{n+1}} (\mathbf{C}_{n+1}^{\tau c} + \mathcal{I}_{n+1}) \quad (4.5)$$

where

$$\begin{aligned} \mathbf{C}_{n+1}^{\tau c} &= (F_{ip} F_{jq} F_{kr} F_{ls} C_{pqrs}^{ve})_{n+1} \mathbf{e}_i \otimes \mathbf{e}_j \otimes \mathbf{e}_k \otimes \mathbf{e}_l \\ \mathcal{I}_{n+1} &= \frac{1}{2} (\delta_{ik} \tau_{jl} + \tau_{ik} \delta_{jl} + \delta_{il} \tau_{jk} + \tau_{il} \delta_{jk})_{n+1} \mathbf{e}_i \otimes \mathbf{e}_j \otimes \mathbf{e}_k \otimes \mathbf{e}_l \end{aligned} \quad (4.6)$$

and \mathbf{e}_k ($k = 1, 2, 3$) being the unit vectors of the Cartesian basis.

For the elasticity tensor corresponding to the modified Knowles model given by Eq. (3.12), see Suchocki (2011).

5. Application to polyethylene

A number of mechanical tests have been carried out in order to verify the developed model's ability to fit the experimental data. The tested material was ultra high molecular weight polyethylene (UHMWPE). UHMWPE is a popular biomaterial with numerous applications as hip joint implants, elbow implants or artificial intervertebral discs for instance. The implant components made of UHMWPE usually play the role of dampers which absorb the mechanical energy, thus making an implant less vulnerable to a failure. UHMWPE's mechanical properties are representative for a class of nonlinear viscoelastic polymers, specifically thermoplastics and thermohardening polymers, for which the magnitude of the elastic strains do not exceed 5%.

The experiments have been carried out in the Laboratory of Strength of Materials, Warsaw University of Technology, Warsaw¹. Three cylindrical specimens made of CHIRULEN 1050 medical UHMWPE were used. Each specimen was subjected to loading-unloading compression test

¹MS Henryk Gołaszewski, from Institute of Mechanics and Printing, Warsaw, had made the specimens and performed the experiments.

Algorithm for the implementation in Abaqus

Input: \mathbf{F}_{n+1} , no. of direct and shear stress components

1. Calculate strain measures from current increment

$$\begin{aligned} J_{n+1} &= \det \mathbf{F}_{n+1} & \mathbf{C}_{n+1} &= \mathbf{F}_{n+1}^T \mathbf{F}_{n+1} \\ \bar{\mathbf{F}}_{n+1} &= J_{n+1}^{-\frac{1}{3}} \mathbf{F}_{n+1} & \bar{\mathbf{B}}_{n+1} &= \bar{\mathbf{F}}_{n+1} \bar{\mathbf{F}}_{n+1}^T \end{aligned}$$

2. Extract stresses from previous increment
3. Calculate elastic stresses from current increment

$$\begin{aligned} \mathbf{S}_{n+1}^{\text{vol}} &= J_{n+1} p_{n+1} \mathbf{C}_{n+1}^{-1} & p_{n+1} &= \partial_{J_{n+1}} U(J_{n+1}) \\ \mathbf{S}_{n+1}^{\text{iso}} &= J_{n+1}^{-\frac{2}{3}} \text{DEV}[\bar{\mathbf{S}}_{n+1}] & \bar{\mathbf{S}}_{n+1} &= 2 \partial_{\bar{\mathbf{C}}_{n+1}} \bar{W}(\bar{\mathbf{C}}_{n+1}) \\ \mathbf{S}_{n+1}^e &= \mathbf{S}_{n+1}^{\text{vol}} + \mathbf{S}_{n+1}^{\text{iso}} \end{aligned}$$

4. Update viscoelastic overstresses ($j = 1, 2, \dots, N$)

$$\mathbf{H}_{j \ n+1} = e^{-\frac{\Delta t}{\tau_j}} \mathbf{H}_{j \ n} + g_j \frac{1 - e^{-\frac{\Delta t}{\tau_j}}}{\frac{\Delta t}{\tau_j}} (\mathbf{S}_{n+1}^e - \mathbf{S}_n^e)$$

5. Calculate total stress from current increment

$$\begin{aligned} \mathbf{S}_{n+1} &= g_\infty \mathbf{S}_{n+1}^e + \sum_{j=1}^N \mathbf{H}_{j \ n+1} \\ \boldsymbol{\sigma}_{n+1} &= \frac{1}{J_{n+1}} \mathbf{F}_{n+1} \mathbf{S}_{n+1} \mathbf{F}_{n+1}^T \end{aligned}$$

6. Calculate viscoelastic stiffness from current increment
7. Store stresses from current increment

with a constant deformation rate $\dot{\lambda}_1$ equal to -0.003 s^{-1} for loading and 0.003 s^{-1} for unloading. The minimum stretch ratio λ_1 equaled 0.954 which corresponded to the maximum Lagrange stress T_{11} of 19.75 MPa. The specimens were tested at the room temperature of 20°C . The Instron 1115 material testing machine was used for the experiments. A sketch of the experimental setup can be seen in Fig. 2. Poisson ratio of 0.46 was measured earlier, thus it was assumed that the material could be treated as incompressible.

The obtained experimental data were further used for the determination of the material parameters. This was achieved by minimizing the error

$$\sum_{k=1}^M \left[(T_{11}(\mathbf{p}))_k - (\tilde{T}_{11})_k \right]^2 = \min \quad (5.1)$$

where $T_{11}(\mathbf{p})$ is the theoretical and \tilde{T}_{11} is the experimental Lagrange stress at the time instant t_k ($k = 1, 2, \dots, M$) and $\mathbf{p} = [\mu, b, \kappa, g_1, g_2, \dots, g_n, g_\infty]^T$ is a vector of the material parameters being optimized. The algorithm used for determining the constants has been programmed in Matlab

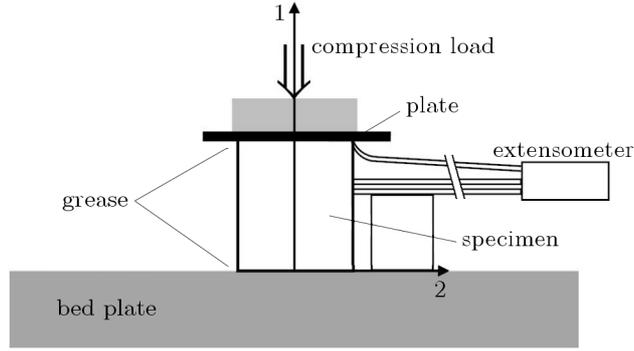


Fig. 2. Experimental set-up

and is described in Suchocki *et al.* (2013). The optimization leads to a relaxation function consisting of $N = 3$ exponential terms. The identified values of the material parameters are collected in Table 1 whereas the results of the curve-fitting can be seen in Fig. 3a and Fig. 3b.

Table 1. Incompressible QLV material parameters of UHMWPE

Elastic constant		Viscoelastic coefficients			
μ	415.13 MPa	g_1	0.13	τ_1	0.82 s
b	266.93	g_2	0.45	τ_2	2.605 s
κ	0.78	g_3	0.17	τ_3	10.84 s

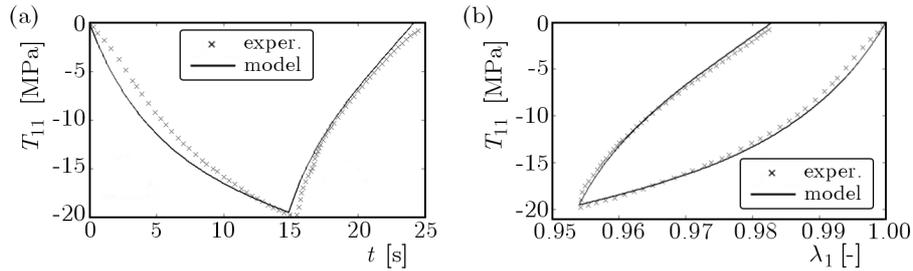


Fig. 3. Fitting of the quasi-linear viscoelastic material model to experimental data

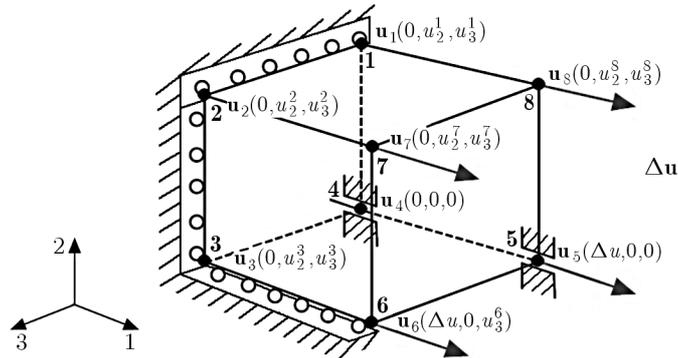


Fig. 4. Boundary conditions used for FE tests

A UMAT (user material) subroutine has been written which allows for using the developed constitutive model within the FE system Abaqus. Exemplary simulations were carried out, involving a $15 \text{ mm} \times 15 \text{ mm} \times 15 \text{ mm}$ UHMWPE block undergoing ramp tension test, ramp compression test and a sinusoidal deformation. The block was meshed with a single finite element. The used set of boundary conditions has been depicted in Fig. 4, where the i -th ($i = 1, 2, 3$)

displacement vector component at j -th ($j = 1, 2, \dots, 8$) node has been denoted as w_i^j . The material incompressibility was modeled by assuming $D_1 = 33 \cdot 10^{-9} \text{MPa}^{-1}$. The simulations were repeated in MATLAB.

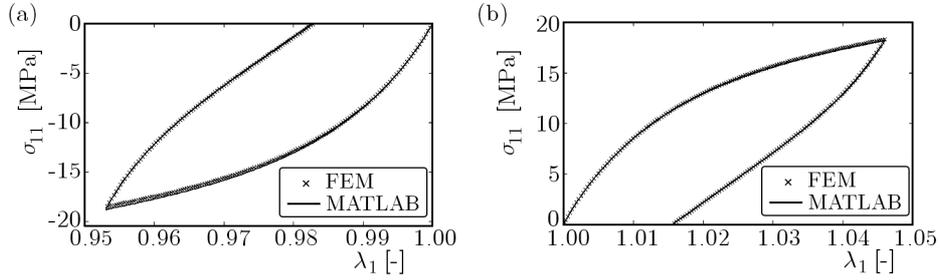


Fig. 5. Polyethylene block undergoing ramp compression and tension tests

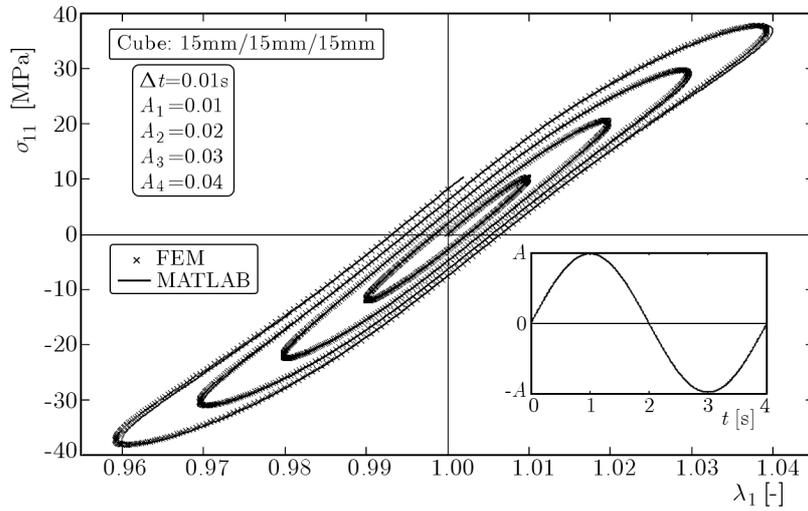


Fig. 6. Polyethylene block undergoing a cyclic strain history – a sinusoidal function with increasing amplitude A : stress versus stretch

The predictions of the FEM simulation utilizing UMAT are in a good agreement with the results produced by MATLAB (see Fig. 5 and Fig. 6). All simulations were repeated for a larger number of finite elements.

6. Conclusions

Many nonlinear viscoelastic constitutive models, originally formulated for elastomeric polymers or soft biological tissues, are not able to accurately describe the mechanical response of thermoplastics and thermohardening polymers. Thus, a new rheological model for polymeric materials has been formulated. The new constitutive equation is obtained by utilizing the Knowles stored-energy function within the framework of the quasi-linear viscoelasticity (QLV) theory, where the relaxation function has been assumed in the form of a series of exponentials, also known as the Prony series.

In order to verify the models' capability of capturing the mechanical behavior of polymers, it was applied to the description of rheological properties of UHMWPE. For the determined set of material parameters, the model described the dissipative response of UHMWPE with a very good accuracy.

The main advantages of the constitutive model for polymers introduced in this study are:

- Very good description of the strongly nonlinear stress-strain relation met in the considered class of polymeric materials.
- Accurate description of rheological effects, especially the hysteresis loop occurring for the cyclic loadings and taking into account its loss of elliptical shape for larger strains.
- Uncoupling volumetric and deviatoric terms within the constitutive relation.

The model has been implemented into the FE system Abaqus by taking advantage of user subroutine UMAT written for that purpose.

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Model reologiczny quasi-liniowej lepkości dla termoplastów i żywic

Streszczenie

Zaproponowano nowy model reologiczny dla materiałów polimerowych. Model bazuje na koncepcji wykorzystania funkcji potencjalnej energii akumulowanej typu Knowlesa w ramach teorii quasi-liniowej lepkości. Równanie konstytutywne quasi-liniowej lepkości w ogólnej postaci sformułowano, wykorzystując formalizm wewnętrznych zmiennych stanu. Opracowane równanie konstytutywne pozwala na opis nieliniowo lepkości wielu materiałów polimerowych, takich jak termoplasty czy żywice. Model wprowadzono do systemu MES. W pracy zaprezentowano zastosowanie opracowanego równania konstytutywnego do modelowania krótkookresowej i dyssypatywnej odpowiedzi polietylenu.

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