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Rheological Properties of Polyamide: Experimental Studies and Constitutive Modeling

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Abstract This work is focused on simulating the rheological effects in polyamide. An experimental study is carried out in order to assess such features of polyamide as: the hysteretic behavior, the strain rate dependence, and the stress relaxation. The material response in tension is investigated. Digital images correlation method (DIC) is employed in order to measure the material compressibility. A newly developed constitutive model, which was previously used to simulate the mechanical response of polyethylene subjected to moderate strains and compressive loadings, is applied to capture the large strain, inelastic behavior of polyamide in tension. The gathered experimental data are utilized to determine the values of constitutive constants of viscoelasticity and plasticity, which describe the rheological properties of polyamide. The determined material parameters are included in the text. Different strategies for evaluating the material parameters are discussed. The proposed constitutive equation is implemented into the finite element (FE) system, ABAQUS, by taking advantage of the user subroutine UMAT, which allows to define custom material laws. Some exemplary FE simulations that were used to investigate the performance of the developed subroutine are described.

Keywords Polyamide; Viscoplasticity; Material parameters; Finite element method; UMAT

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INTRODUCTION

The polymeric materials play an increasingly important role in different branches of industry, such as automotive or aerospace engineering. The classical material models of linear elasticity and viscoelasticity allow for a very limited description of material inelastic response which the polymers exhibit. Thus, for decades, new constitutive models are developed which are aimed at a more accurate simulation of the mechanical properties of polymeric materials.

The constitutive theories which account for both material and geometrical nonlinearity can be roughly divided into three subgroups: the nonlinear elastic models (with the special case of hyperelastic models, *i.e.* the nonlinear elastic models for which a stored energy function exists), the nonlinear viscoelastic models, and the viscoplastic models. The viscoplastic model formulations which result from a straightforward generalization of the nonlinear viscoelasticity are often called the viscoelastic-plastic models. The constitutive equations of nonlinear viscoelasticity and viscoplasticity are usually formulated by utilizing the framework of scalar or tensorial internal state variables. Two basic groups can be distinguished among the utilized internal state variables, *i.e.* the so-called stress-like variables^[1,2] and the strain-like vari-

ables.^[3] The model formulation using the strain-like internal state variables usually utilizes the multiplicative split of the deformation gradient tensor into its elastic and viscous components. The constitutive models which use the stress-like internal state variables are often based on the Maxwell mechanistic model.

A popular strategy for model building, which is usually employed in the case of using stress-like internal state variables, assumes that the material response follows the superposition of elastic, plastic, and viscoelastic behaviors. According to this assumption, the purely elastoplastic response is exhibited for an infinitely slow loading (the so-called material equilibrium response), *e.g.* Käßner *et al.*^[4] A model describing the equilibrium material response is further extended by adding terms which account for such effects as the strain rate dependency and the stress relaxation.

The rate and time effects can be described by defining an explicit strain rate dependence (*e.g.* Pioletti *et al.*^[5]) or by incorporating the internal state variables framework. The change of such variables with time must be determined by proper evolution equations. Different model formulations are possible when the internal state variable method is used. The evolution of tensorial state variables with the physical meaning of non-equilibrium stress can, in the simplest case, follow a linear differential equation derived from Maxwell's rheological model.^[1,2,6] A different approach involves the usage of a nonlinear differential equation, where the relaxation

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time can be defined as a function of the strain rate or the non-equilibrium stress.^[4,7] Furthermore, fractional differential equations can be used to define the evolution of the state variables.^[4,8] In the most popular variant, a fractional evolution equation is employed which corresponds to the so-called fractional exponential relaxation function (a modified Mittag-Leffler function), when the model is written in the integral form. Some more sophisticated model formulations exist on the basis of fractional evolution equations. For instance, a relaxation function can be formulated in a manner which reflects the connection between the spectrum of relaxation time and the polymer's molecular mass distribution (*cf.* Garbarsi^[9]). The major benefit of using the fractional evolution equations is the reduction in the number of material parameters necessary to describe the rheological properties of material. However, such model formulations result in a significant increase of the computational cost of numerical simulations, *e.g.* Ciambella *et al.*^[10] Furthermore, the fractional models allow for a good approximation of the experimental data, but in some cases, better results can be obtained when a classical relaxation function is used, such as a series of exponential terms (the so-called Prony series).

Numerous constitutive model formulations have been developed over many years in order to capture the rheological behavior of thermoplastic polymers. A number of such models were applied to simulate the mechanical response of different kinds of polyamide. For instance, Ghorbel^[11] proposed a viscoplastic constitutive model based on the flow rule which was used for simulating the rheological properties of PA12 in the small strain domain. Similar concepts were followed by Krairi and Doghri^[12] who formulated a viscoplastic-damage constitutive equation for polyamide (Nylon 101), which takes into account the material degradation. The aforementioned model is defined for the range of small strains. An elastoplastic component which utilizes the framework of flow plasticity is used to describe the material equilibrium response. A viscoelastic term containing a three-term Prony series is utilized to simulate the time effects. A similar approach has been proposed by Praud *et al.*^[13] recently to describe the mechanical properties of PA66 by using the infinitesimal strain measures. The methods of flow plasticity combined with the small strain formulation were also used by da Costa Mattos *et al.*^[14] to capture the mechanical response of Nylon 12. Zeng *et al.*^[15] developed a hyperelastic-plastic constitutive model which was used for simulating the large strain behavior of PA6 at elevated temperatures. The experimental data from both uniaxial and biaxial tension experiments were utilized for calibrating the model. Another model formulation which allows to capture the large strain material response was proposed by Maurel-Pantel *et al.*^[16] This model formulation employed the multiplicative split of the deformation gradient into its elastic and viscous components. The framework of large strain viscoplasticity utilizing a flow rule was applied.

In this work, a recently developed viscoelastic-plastic model for polymers is employed to describe the rheological effects in polyamide. This constitutive law was previously utilized to capture the mechanical behavior of polyethylene

subjected to moderate strains and compressive loadings (*cf.* Suchocki^[17]). Herein, the new viscoelastic-plastic constitutive equation is applied to simulate the large strain, inelastic response of polyamide in tension. A large number of experimental tests were performed which enabled to characterize such effects as the stress-strain hysteresis, the strain rate dependence, and the stress relaxation. Digital images correlation method (DIC) was utilized to measure the material compressibility. The gathered experimental data were subsequently used to determine the material parameter values which correspond to the mechanical properties of polyamide. A number of different strategies for evaluation of the material constants were tested and discussed in this study. The assumed framework allowed to achieve good approximation results. The obtained constant values are attached in the text. The proposed constitutive model was implemented into the finite element (FE) system, ABAQUS, by taking advantage of the user subroutine UMAT, which facilitates the definition of custom material laws. The FE simulations which were used to verify the performance of developed subroutine UMAT are discussed.

EXPERIMENTAL SETUP AND TESTS

In order to determine the constitutive constants of polyamide, a series of experiments were conducted by using the static mechanical test system Zwick/Roell Z005 with the maximum load capacity of 5 kN. The specimens were fixed in the testing machine's pressure-jaws with the constant pressure of about 4 MPa (Fig. 1). In order to measure the exact elongation of polyamide specimens (PA66; trade name: Zytel 101), the long-range extensometer with the total measurement range of about 1.2 m was employed.

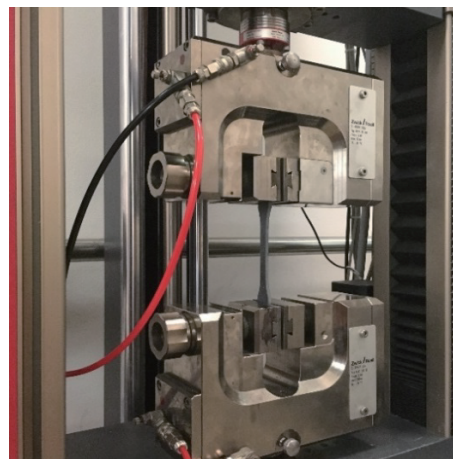


Fig. 1 A specimen fixed in the pressure jaws of Zwick/Roell Z005

For the purpose of estimating the material compressibility, a non-contact, optical method based on digital images correlation (DIC) was utilized. The DIC method allows one to obtain the maps of displacements and macroscopic deformations measured on a surface of the tested specimen. The DIC experimental setup consists of a black and white digital camera with a suitable lens, and a PC with installed software (Fig. 2a).

The presented research used the commercial program Vic2D. Before the test, random spots with high contrast must be applied to the surface (Fig. 2b). After selecting the field for analysis, the specimen's surface is divided into smaller subareas characterized by a unique distribution of gray shades. A computer algorithm compares the digital photographs of a non-deformed sample with the photos taken after the deformation and finds the new positions of analyzed subareas by utilizing the method of best fit. As a result, a displacement map or macroscopic deformations are obtained for the entire analyzed area. The accuracy of displacement measurements depends on the camera resolution and reaches 0.01 pixels.

The standard dog-bone specimens were used with the dimensions of cross-section 4 mm × 10 mm and 150 mm of total length. The specimens were manufactured according to the guidelines of the Polish standard, *i.e.* PN-EN ISO 527-2:2012 “ISO 527-2:2012; Plastics—Determination of tensile properties—Part 2: Test conditions for molding and extrusion plastics”. Three kinds of experiments were performed: the monotonic tension tests, the loading/unloading tests, and the stress relaxation tests. Some exemplary results are presented in Fig. 3.

CONSTITUTIVE MODELING FRAMEWORK

The formulation of the utilized viscoelastic-plastic constitutive equation is presented below. The basic notions of continuum mechanics such as the kinematics of finite deformations and some aspects of finite elasticity are discussed first.

Basic Notions

For the sake of simplicity, it is assumed that the material inelasticity is dependent on the isochoric component of the deformation gradient tensor $\mathbf{F}$. The material response due to a volume change is treated as purely elastic. The decoupling of volumetric and isochoric behaviors can be facilitated by utilizing the multiplicative decomposition of the deformation gradient tensor,^[2] *i.e.*

$$\mathbf{F} = \mathbf{F}_{\text{vol}} \bar{\mathbf{F}} \quad (1)$$

where $\mathbf{F}_{\text{vol}}$ and $\bar{\mathbf{F}}$ are the volumetric and the isochoric components of the deformation gradient tensor, respectively, which are determined by the following formulae:

$$\bar{\mathbf{F}} = J^{-1/3} \mathbf{F}; \quad \mathbf{F}_{\text{vol}} = J^{1/3} \mathbf{I}; \quad J = \det \mathbf{F} \quad (2)$$

Consequently, two different formulations of the right Cauchy-Green (C-G) deformation tensor can be introduced:

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

where $\mathbf{C}$ describes both the isochoric and the volumetric deformations, whereas $\bar{\mathbf{C}}$ is a measure of a purely isochoric deformation.

The method of constitutive modeling proposed in this study is an extension of the finite elasticity, *i.e.* hyperelasticity. Thus, the fundamentals of the decoupled hyperelastic modeling are summarized below. An isotropic material is assumed to be hyperelastic if it possesses a stored elastic energy function which is invariant with respect to a rotation $\mathbf{Q}$:^[2]

$$W(\mathbf{C}) = W(\mathbf{Q}\mathbf{C}\mathbf{Q}^T) \quad (4)$$

The stored energy function plays a role of potential with

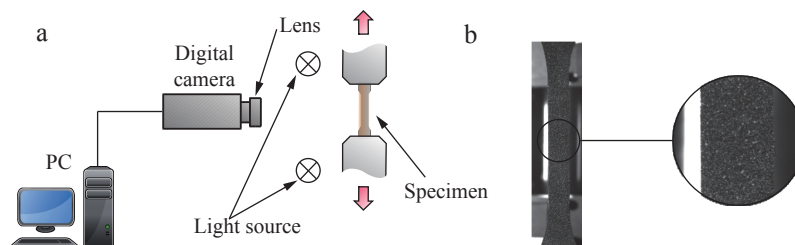


Fig. 2 The experimental setup: (a) A scheme of DIC experimental arrangement; (b) A specimen with the random and high contrast pattern on its surface

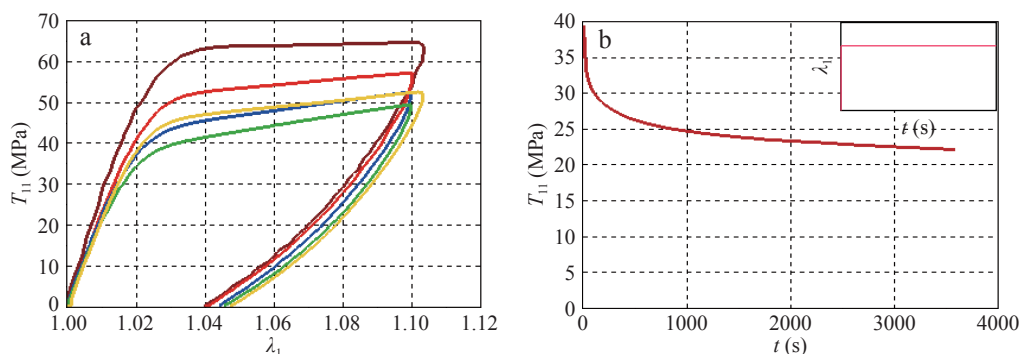


Fig. 3 Exemplary experimental data: (a) The engineering stress versus the stretch ratio plots obtained from the loading/unloading tests performed at different stretch rates; (b) The engineering stress versus time plot obtained from the stress relaxation experiment

respect to the second Piola-Kirchhoff (P-K) stress tensor $\mathbf{S}_0$, *i.e.*

$$\mathbf{S}_0 = \frac{\partial W}{\partial \mathbf{C}} \quad (5)$$

In the case of decoupling the volumetric and the isochoric responses, the stored energy function is assumed to be the sum of its volumetric component $U(J)$ and the isochoric component $\bar{W}(\bar{\mathbf{C}})$:^[2]

$$W(\mathbf{C}) = U(J) + \bar{W}(\bar{\mathbf{C}}) \quad (6)$$

By substituting Eq. (6) in Eq. (5), the decoupled form of the constitutive equation describing a hyperelastic material is found, *i.e.*

$$\mathbf{S}_0 = \mathbf{S}_0^{\text{vol}} + \mathbf{S}_0^{\text{iso}}; \quad \mathbf{S}_0^{\text{vol}} = Jp\mathbf{C}^{-1}; \quad \mathbf{S}_0^{\text{iso}} = J^{-2/3}\text{DEV}[\bar{\mathbf{S}}] \quad (7)$$

with

$$p = \frac{\partial U}{\partial J}; \quad \bar{\mathbf{S}} = 2 \frac{\partial \bar{W}}{\partial \bar{\mathbf{C}}} \quad (8)$$

where $\text{DEV}[\bullet] = [\bullet] - 1/3([\bullet] \cdot \bar{\mathbf{C}})\bar{\mathbf{C}}^{-1}$ is an operator which extracts the deviatoric component of a tensor in the reference configuration.

Viscoelastic-Plastic Constitutive Model Formulation

The constitutive equation of hyperelasticity is extended by incorporating the stress-like tensorial state variables, *i.e.* $\bar{\mathbf{H}}_k$ ($k = 1, 2, \dots, M$) and $\mathbf{H}_j$ ($j = 1, 2, \dots, N$). The total second P-K stress is assumed to be the sum of the elastic component determined by Eq. (7) and the inelastic over stresses

$$\mathbf{S} = \mathbf{S}_0 + \sum_{k=1}^M \bar{\mathbf{H}}_k + \sum_{j=1}^N \mathbf{H}_j \quad (9)$$

where $\mathbf{S}_0$ and $\bar{\mathbf{H}}_k$ ($k = 1, 2, \dots, M$) are responsible for the so-called equilibrium, rate-independent elastoplastic response, whereas the over stresses $\mathbf{H}_j$ ($j = 1, 2, \dots, N$) account for the rate dependency and the stress relaxation.^[17]

The evolution of $\mathbf{H}_j$ ($j = 1, 2, \dots, N$) variables follows a linear differential equation inspired by the Maxwell's mechanistic model,^[2,6] *i.e.*

$$\dot{\mathbf{H}}_j + \frac{1}{\tau_j} \mathbf{H}_j = \Gamma_j \dot{\mathbf{S}}_0^{\text{iso}} \quad j = 1, 2, \dots, N \quad (10)$$

which, after some manipulations, can be rewritten in an integral form:

$$\mathbf{H}_j(t) = \int_0^t \Gamma_j e^{-\frac{t-\tau}{\tau_j}} \frac{\partial \mathbf{S}_0^{\text{iso}}(\tau)}{\partial \tau} d\tau \quad j = 1, 2, \dots, N \quad (11)$$

where Γ_j is the viscoelastic coefficient and τ_j is the relaxation time ($j = 1, 2, \dots, N$).

The evolution of the rate-independent internal state variable $\bar{\mathbf{H}}_k$ ($k = 1, 2, \dots, M$) is governed by the following equation:^[17]

$$\dot{\bar{\mathbf{H}}}_k + \frac{1}{\bar{D}_k M \left(\left| \dot{\bar{\mathbf{C}}} \right| \right)} \bar{\mathbf{H}}_k = \gamma_k \dot{\mathbf{S}}_0^{\text{iso}} \quad k = 1, 2, \dots, M \quad (12)$$

with

$$M \left(\left| \dot{\bar{\mathbf{C}}} \right| \right) = \bar{J}_2^{-1/2}; \quad \bar{J}_2 = \text{tr} \dot{\bar{\mathbf{C}}}^2 = \dot{\bar{\mathbf{C}}} \cdot \dot{\bar{\mathbf{C}}} \quad (13)$$

The integration of Eq. (12) leads to the relation given below:

$$\bar{\mathbf{H}}_k(t) = \int_0^{z(t)} \gamma_k e^{-\frac{z-\zeta}{\bar{D}_k}} \frac{\partial \mathbf{S}_0^{\text{iso}}(\zeta)}{\partial \zeta} d\zeta \quad k = 1, 2, \dots, M \quad (14)$$

where γ_k and $\bar{D}_k$ are the material parameters ($k = 1, 2, \dots, M$) and ζ is a fictitious time variable with the physical meaning of the arc length in nine-dimensional space of components of isochoric right C-G tensor $\bar{\mathbf{C}}$, *i.e.*

$$\zeta = \frac{1}{M \left(\left| \dot{\bar{\mathbf{C}}} \right| \right)}; \quad \zeta(\tau) = \int_0^\tau \sqrt{d\bar{\mathbf{C}}(\tau') \cdot d\bar{\mathbf{C}}(\tau')}; \quad \zeta(t) = z \quad (15)$$

Below the framework of integrating the constitutive equation is presented.

Constitutive Model Discretization

For the time increment $n+1$ the total P-K stress of the second kind is given as:

$$\mathbf{S}_{n+1} = \mathbf{S}_{0\,n+1} + \sum_{k=1}^M \bar{\mathbf{H}}_{k\,n+1} + \sum_{j=1}^N \mathbf{H}_{j\,n+1} \quad (16)$$

The components of the viscoelastic over stresses $\mathbf{H}_{j\,n+1}$ ($j = 1, 2, \dots, N$) are calculated by utilizing the recurrence-update formula developed by Taylor *et al.*,^[18] *i.e.*

$$\mathbf{H}_{j\,n+1} = e^{-\Delta t/\tau_j} \mathbf{H}_{j\,n} + \Gamma_j \frac{1 - e^{-\Delta t/\tau_j}}{\Delta t/\tau_j} (\mathbf{S}_{0\,n+1}^{\text{iso}} - \mathbf{S}_{0\,n}^{\text{iso}}) \quad (17)$$

A recurrence-update formula for the evolution of rate-independent over stresses $\bar{\mathbf{H}}_k$ ($k = 1, 2, \dots, M$) is obtained by applying the central difference method to Eq. (12) which results in:

$$\bar{\mathbf{H}}_{k\,n+1} = \frac{\left(1 - \frac{1}{\bar{D}_k} \frac{\Delta z_{n+1/2}}{2}\right) \bar{\mathbf{H}}_{k\,n} + \gamma_k (\mathbf{S}_{0\,n+1}^{\text{iso}} - \mathbf{S}_{0\,n}^{\text{iso}})}{1 + \frac{1}{\bar{D}_k} \frac{\Delta z_{n+1/2}}{2}} \quad (18)$$

with

$$\Delta z_{n+1/2} = \sqrt{\Delta \bar{\mathbf{C}}_{n+1/2} \cdot \Delta \bar{\mathbf{C}}_{n+1/2}}; \quad \Delta \bar{\mathbf{C}}_{n+1/2} = \bar{\mathbf{C}}_{n+1} - \bar{\mathbf{C}}_n \quad (19)$$

A more specific form of the viscoelastic-plastic constitutive model is obtained by selecting the proper stored energy functions and the numbers of the inelastic over stresses M and N .

ELASTIC-PLASTIC MODELING

In the first approach, the rate-independent, elastoplastic version of the model is applied to fit the experimental data. Thus, in the considered case $N = 0$, whereas $M = 1$ in Eq. (9). The material parameter values determined at this stage are further utilized to evaluate the constitutive constants of the viscoelastic-plastic model.

For the purpose of describing the inelastic response of polyamide, the isochoric stored energy function $\bar{W}$ in the form proposed by Knowles^[19] is utilized, *i.e.*

$$\bar{W}(\bar{I}_1) = \frac{\mu}{2b} \left\{ \left[1 + \frac{b}{\kappa} (\bar{I}_1 - 3) \right]^\kappa - 1 \right\} \quad (20)$$

with $\bar{I}_1 = \text{tr } \bar{\mathbf{C}}$, whereas μ , b , and κ are the material parameters. The volumetric component $U(J)$ of the stored energy function is assumed in the standard form:

$$U(J) = \frac{1}{D_1}(J-1)^2 \quad (21)$$

where D_1 is the inverse of the bulk modulus. The approximation results obtained independently for different uniaxial loading/unloading experimental data are shown in Fig. 4. An excellent agreement between the model predictions and the experimental measurements was found.

The material parameters were determined by the minimization of total quadratic error:

$$\mathcal{F}(\mathbf{p}) = \sum_{r=1}^P [(T_{11}(\mathbf{p}))_r - (\tilde{T}_{11})_r]^2 \quad (22)$$

where $T_{11}(\mathbf{p})$ is the theoretical and $\tilde{T}_{11}$ is the experimental

engineering stress in the axial direction of specimen for the collocation point r ($r = 1, 2, \dots, P$), whereas $\mathbf{p} = [\mu \ b \ \kappa \ \gamma_1 \ \bar{D}_1]^T$ is the column matrix of the optimized constant values. The parameters were determined with the assumption of material incompressibility. Subsequently, the true value of the volumetric modulus, *i.e.* $D_1 = 6.74 \times 10^{-4} \text{ MPa}^{-1}$ was calculated based on the stretch ratio measurements obtained from the DIC system. The discretized version of the model which is given by Eqs. (16)–(19) was utilized for calculation of the theoretical engineering stress. The parameter values determined for different stretch rates $\dot{\lambda}$ are summarized in Table 1. It should be noted that only one material constant changes significantly with the deformation rate, *i.e.* b . The values of the remaining four material parameters are almost independent of the stretch rate. The information about the residual sum of squares (RSS) has been included in Fig. 4.

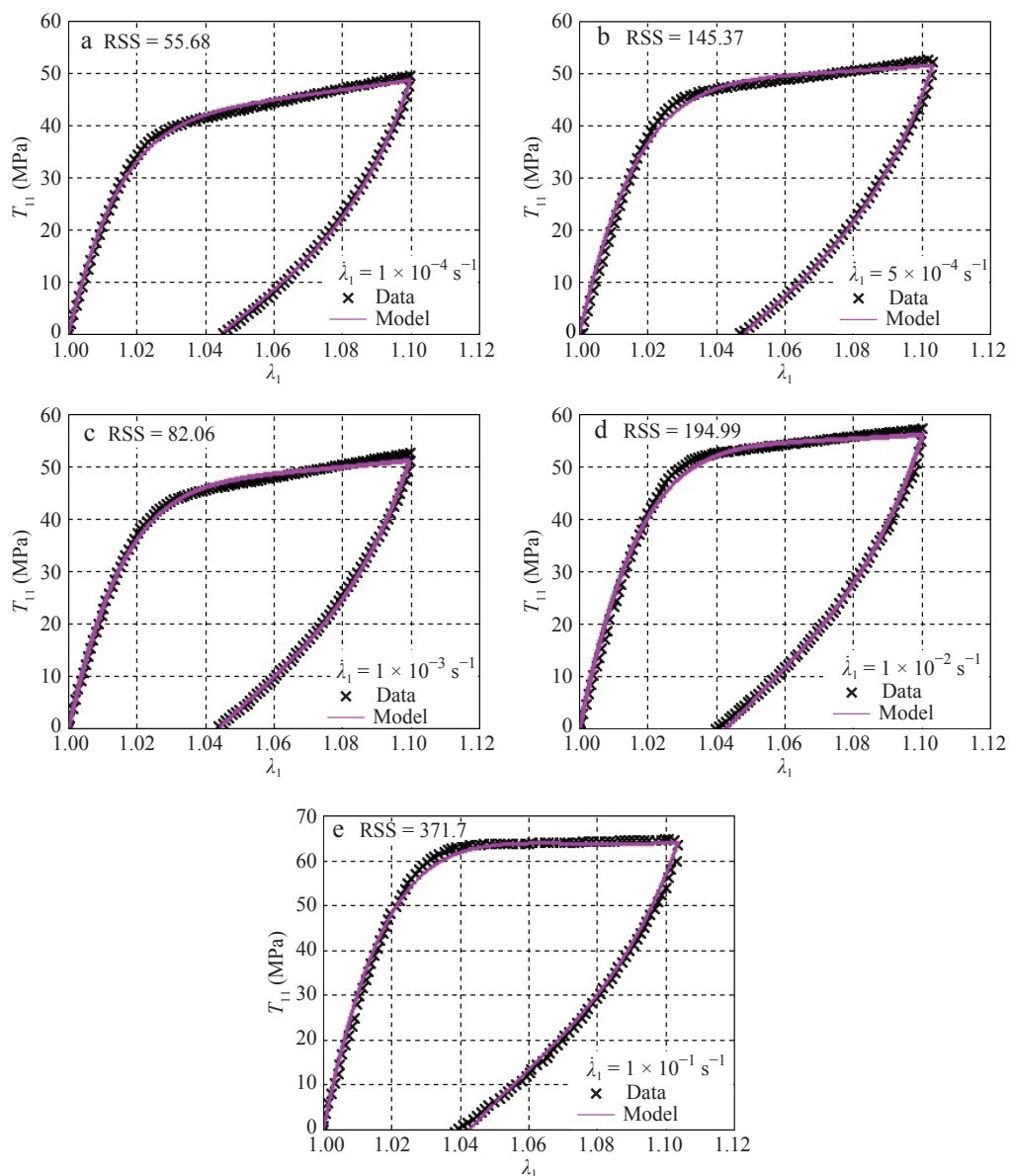


Fig. 4 The comparison of elastic-plastic model predictions and the hysteresis data obtained for polyamide with the deformation rate of (a) $1 \times 10^{-4} \text{ s}^{-1}$; (b) $5 \times 10^{-4} \text{ s}^{-1}$; (c) $1 \times 10^{-3} \text{ s}^{-1}$; (d) $1 \times 10^{-2} \text{ s}^{-1}$; (e) $1 \times 10^{-1} \text{ s}^{-1}$

Table 1 The material parameters of elastic-plastic model determined for polyamide

Material parameter	$\dot{\lambda} = 1 \times 10^{-1} \text{ s}^{-1}$	$\dot{\lambda} = 1 \times 10^{-2} \text{ s}^{-1}$	$\dot{\lambda} = 1 \times 10^{-3} \text{ s}^{-1}$	$\dot{\lambda} = 5 \times 10^{-4} \text{ s}^{-1}$	$\dot{\lambda} = 1 \times 10^{-4} \text{ s}^{-1}$
μ (MPa)	248.92	210.95	191.82	181.09	180.46
b	232.16	282.91	460.63	428.62	695.09
κ	0.78	0.81	0.83	0.83	0.86
γ_1	4.01	3.95	3.99	4.29	4.02
$\tilde{D}_1$	0.05	0.05	0.05	0.05	0.05

VISCOELASTIC-PLASTIC MODELING

For the purpose of formulating a viscoelastic-plastic model for polyamide, the isochoric stored energy function defined by Eq. (20) was utilized. The assumed volumetric energy function is given by Eq. (21). In this section, a variant of the viscoelastic-plastic model is analyzed which utilizes one elastoplastic term ($M = 1$) and two viscoelastic terms ($N = 2$) in Eq. (9). In this case, the material response is determined by nine material parameters which have been gathered in a column matrix, *i.e.* $\mathbf{p} = [\mu \ b \ \kappa \ \gamma_1 \ \tilde{D}_1 \ \Gamma_1 \ \Gamma_2 \ \tau_1 \ \tau_2]^T$. Again, these parameters are found by minimizing the error function defined by Eq. (22). Three selected strategies of evaluating the material parameter values are discussed below. It should be remarked here, that a model formulation using three viscoelastic terms ($N = 3$) was also tested. It was found that the viscoelastic coefficient Γ_3 tended to be zero during the course of the constant value optimization process. Thus, it seems that in the case of simulating rheological properties of the given polyamide 66, there is no point in utilizing more than two viscoelastic terms.

The first method for determining the constitutive constants

involves the usage of experimental data from two rheological tests, *i.e.* a loading/unloading experiment performed at the deformation rate $\dot{\lambda} = 1 \times 10^{-2} \text{ s}^{-1}$, and a relaxation test. The preloading which took place prior to the stress relaxation was performed at the same deformation rate as that used during the loading/unloading experiment. Some material parameter values for the elastoplastic model evaluated previously were utilized during the optimization process as the initial constant values (Table 1, column 3). The material parameter determination was performed in two steps. First, both the hysteresis and the relaxation data were approximated simultaneously. Second, the stress relaxation data were used to recalibrate the previously obtained material parameter set during this step.

The approximation results can be seen in Figs. 5(a) and 5(b). In order to verify the model's performance for the case of large stretches, the theoretical curve obtained for a monotonic uniaxial tension process was compared to the one measured experimentally for the deformation rate $\dot{\lambda} = 1 \times 10^{-2} \text{ s}^{-1}$. The comparison result is shown in Fig. 5(c). The determined material parameter values have been summarized in Table 2 (set 1).

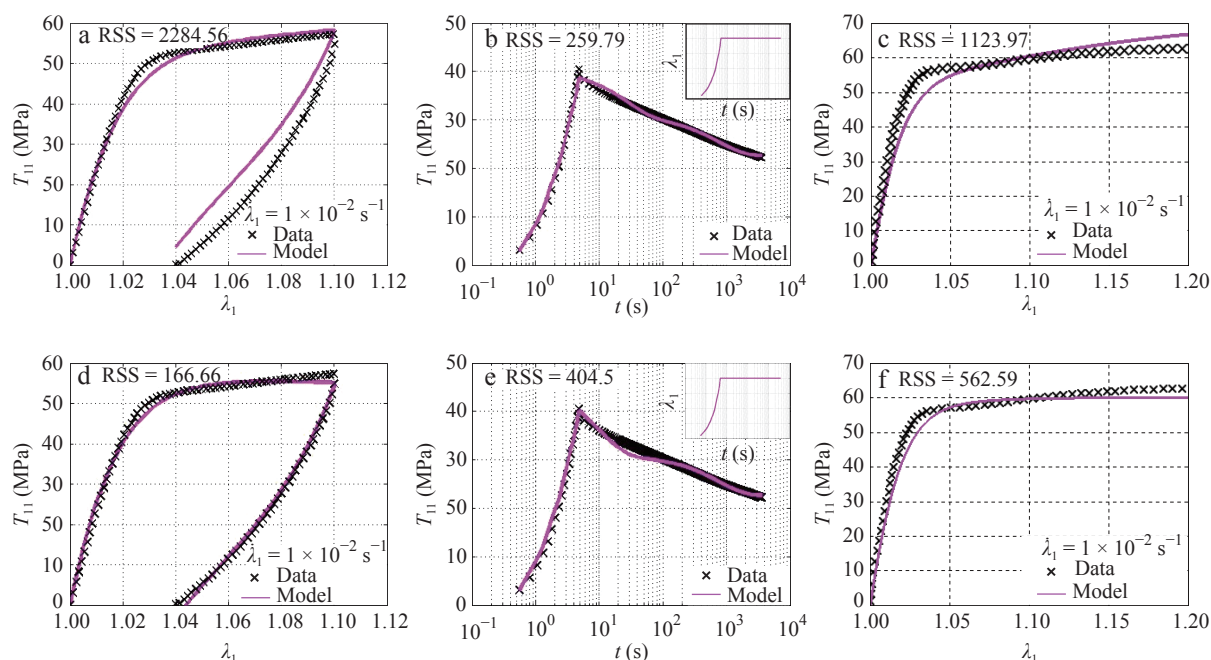


Fig. 5 The comparison of viscoelastic-plastic model predictions (constant sets 1 and 2) and the experimental data for polyamide: (a) The hysteresis data obtained for deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$ (set 1); (b) The relaxation data with preloading at deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$ (set 1); (c) The monotonic loading data obtained for deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$ (set 1); (d) The hysteresis data obtained for deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$ (set 2); (e) The relaxation data with preloading at deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$ (set 2); (f) The monotonic loading data obtained for deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$ (set 2)

Table 2 The material parameters of viscoelastic-plastic model determined for polyamide

Material parameter	Set 1	Set 2	Set 3
μ (MPa)	32.59	32.59	30.36
b	304.26	296.26	249.36
κ	0.78	0.78	0.81
γ_1	19.04	19.04	18.28
$\bar{D}_1$	0.05	0.05	0.06
Γ_1	4.99	6.74	14.30
Γ_2	4.62	4.62	5.90
τ_1 (s)	25.67	9.67	1.25
τ_2 (s)	731.88	731.88	593.01

The second discussed material parameter set is the one obtained by a simultaneous approximation of the previously used hysteresis and stress relaxation data. The results of the fitted curve are exhibited in Figs. 5(d) and 5(e). In Fig. 5(f), the comparison of model predictions and monotonic test data can be observed ($\dot{\lambda} = 1 \times 10^{-2} \text{ s}^{-1}$). The determined constant values have been listed in Table 2 (set 2).

In the third approach, the set of experimental measurements used for curve fitting was extended by including the hysteresis data which were obtained for deformation rate magnitude $\dot{\lambda} = 1 \times 10^{-3} \text{ s}^{-1}$. The previously determined constant values (Table 2, set 2) were utilized as the so-called initial guess during the new optimization process. The results of the curve-fitting can be seen in Fig. 6. A good correlation between the experimental and the theoretical results was found (cf. Figs. 6a–6c). In Fig. 6(d) the comparison of theoretical predictions and monotonic test data ($\dot{\lambda} = 1 \times 10^{-2} \text{ s}^{-1}$)

is presented. The evaluated parameter values have been gathered in Table 2 (set 3).

All the material parameters were determined with the assumption of ideal incompressibility. As mentioned before, the experimental measurements collected by the DIC system were later used to assess the material compressibility and determine the true value of the volumetric modulus.

It can be seen from Figs. 5 and 6 that slightly different results are obtained depending on which experimental data sets were utilized for the curve fitting. For instance, a simultaneous data fitting of a single hysteresis loop along with the relaxation data leads to a satisfactory approximation which is depicted in Figs. 5(d) and 5(e). Subsequent utilization of the relaxation data to recalibrate the obtained parameter values can improve the approximation (Fig. 5b), but this is achieved at the price of worsening the fitting to the hysteresis data (Fig. 5a). The material constants evaluated according to the framework described above (Table 2, constant sets 1 and 2) allow to describe the rate dependence of material's response accurately for the stretch rates from range 0.005–0.05 s^{-1} . The material's sensitivity to the deformation rate can be captured more accurately if two experimental hysteresis loops, obtained for different stretch rates, are utilized during the curve fitting (Figs. 6a and 6b). In this case, a less accurate approximation of the short term relaxation is achieved (Fig. 6c). However, the evaluated constitutive constants (Table 2, set 3) enable to accurately capture the material's rate sensitivity for a broader range of stretch rates, i.e. 0.0005–0.01 s^{-1} .

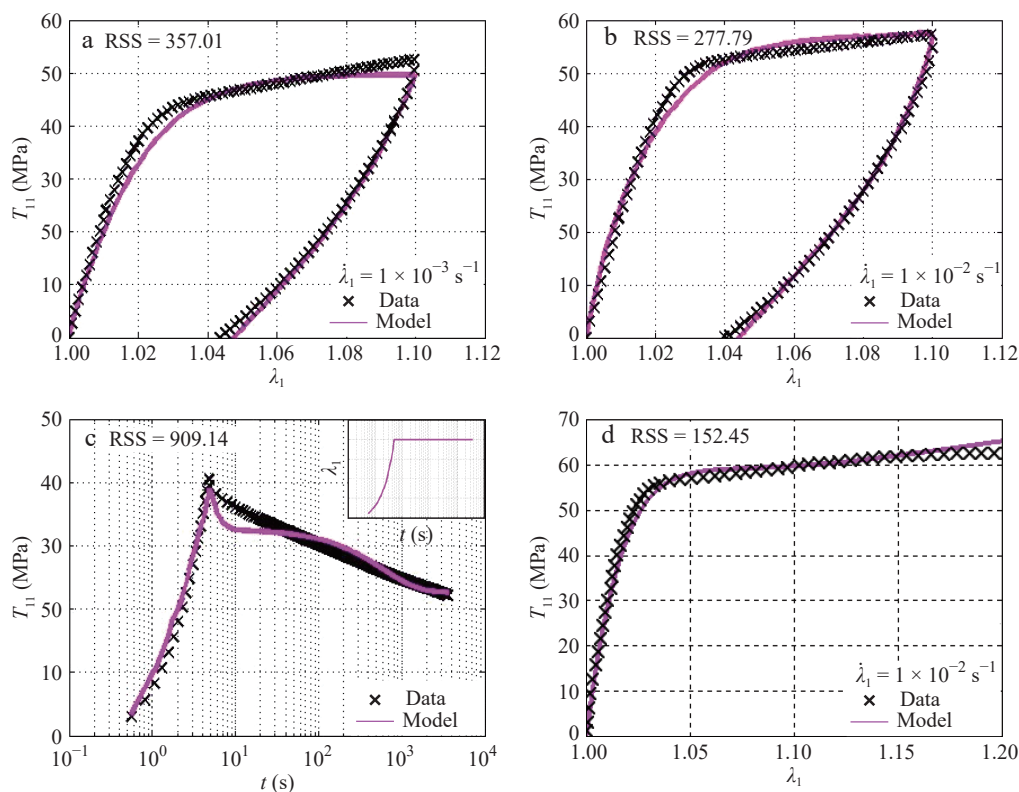


Fig. 6 The comparison of viscoelastic-plastic model predictions (constant set 3) and the experimental data for polyamide: (a) The hysteresis data obtained for deformation rate of $1 \times 10^{-3} \text{ s}^{-1}$; (b) The hysteresis data obtained for deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$; (c) The relaxation data with preloading (deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$); (d) The monotonic loading data obtained for deformation rate of $1 \times 10^{-2} \text{ s}^{-1}$

FINITE ELEMENT IMPLEMENTATION

The presented viscoelastic-plastic constitutive equation was implemented into the FE system ABAQUS by utilizing a user subroutine UMAT,^[20] which was specifically developed for that purpose. The FE implementation requires linearization of the constitutive model's discrete form defined by Eqs. (16)–(19). The derivation of the constitutive model's incremental form is briefly discussed below.

Incremental Form of Constitutive Model

Calculating a directional derivative of the second P-K stress at increment $n+1$, as given by Eq. (16), leads to the following incremental relation in the reference configuration:^[2]

$$\Delta \mathbf{S}_{n+1} = \mathbb{C}_{n+1}^{ve-p} \cdot \frac{1}{2} \Delta \mathbf{C}_{n+1}; \quad \mathbb{C}_{n+1}^{ve-p} = 2 \frac{\partial \mathbf{S}_{n+1}}{\partial \mathbf{C}_{n+1}} \quad (23)$$

with the viscoelastic-plastic tangent operator given as:^[17]

$$\mathbb{C}_{n+1}^{ve-p} = \mathbb{C}_{0n+1}^{vol} + \left[1 + \sum_{k=1}^M \gamma_k \left(1 + \frac{\Delta z_{n+1/2}}{2\tilde{D}_k} \right)^{-1} + \sum_{j=1}^N F_j \frac{1 - e^{-\Delta t/\tau_j}}{\Delta t/\tau_j} \right] \mathbb{C}_{0n+1}^{iso} \quad (24)$$

where the arc length $\Delta z_{n+1/2}$ is defined by Eq. (19)₁. After pushing Eq. (23)₁ forward to the current configuration, the following incremental rate equation is obtained:

$$\boldsymbol{\tau}_{n+1}^\nabla = J_{n+1} \mathbb{C}_{n+1}^{MJ} \cdot \Delta \mathbf{D}_{n+1} \quad (25)$$

where $\boldsymbol{\tau}_{n+1}^\nabla$ is the incremental Zaremba-Jaumann rate of the Kirchhoff stress $\boldsymbol{\tau}_{n+1} = \mathbf{F}_{n+1} \mathbf{S}_{n+1} \mathbf{F}_{n+1}^T$, *i.e.*

$$\boldsymbol{\tau}_{n+1}^\nabla = \Delta \boldsymbol{\tau}_{n+1} - \Delta \mathbf{W}_{n+1} \boldsymbol{\tau}_{n+1} - \boldsymbol{\tau}_{n+1} \Delta \mathbf{W}_{n+1}^T \quad (26)$$

whereas the increments of the strain rate tensor $\Delta \mathbf{D}_{n+1}$ and the spin tensor $\Delta \mathbf{W}_{n+1}$ are given as

$$\Delta \mathbf{D}_{n+1} = \frac{1}{2} \left[\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1} + \left(\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1} \right)^T \right] \quad (27)$$

and

$$\Delta \mathbf{W}_{n+1} = \frac{1}{2} \left[\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1} - \left(\Delta \mathbf{F}_{n+1} \mathbf{F}_{n+1}^{-1} \right)^T \right] \quad (28)$$

The increment of the deformation gradient tensor is defined as follows:

$$\Delta \mathbf{F}_{n+1} = \mathbf{F}_{n+1} \mathbf{F}_n^{-1} \quad (29)$$

The FE system, ABAQUS, requires defining $\mathbb{C}_{n+1}^{MJ}$, *i.e.* the so-called Material Jacobian (MJ) along with the Cauchy stress $\boldsymbol{\sigma}_{n+1} = J_{n+1}^{-1} \boldsymbol{\tau}_{n+1}$ within the UMAT code. During a FE analysis the user subroutine is called by the solver to calculate the MJ and the Cauchy stress. In the considered case:

$$\mathbb{C}_{n+1}^{MJ} = \frac{1}{J_{n+1}} \left(\mathbb{C}_{n+1}^{tc} + \mathbb{H}_{n+1} \right) \quad (30)$$

where

$$\mathbb{C}_{n+1}^{tc} = \left(F_{iP} F_{jQ} F_{kR} F_{lS} \mathbb{C}_{PQRS}^{ve-p} \right)_{n+1} \mathbf{e}_i \otimes \mathbf{e}_j \otimes \mathbf{e}_k \otimes \mathbf{e}_l \quad (31)$$

and

$$\mathbb{H}_{n+1} = \frac{1}{2} \left(\delta_{ik} \tau_{jl} + \tau_{ik} \delta_{jl} + \delta_{il} \tau_{jk} + \tau_{il} \delta_{jk} \right)_{n+1} \mathbf{e}_i \otimes \mathbf{e}_j \otimes \mathbf{e}_k \otimes \mathbf{e}_l \quad (32)$$

with $\mathbf{e}_i$ ($i = 1, 2, 3$) being the unit vectors of a Cartesian base. Some exemplary FE simulations are described below to verify the performance of the developed UMAT code.

Simulations of Homogeneous Deformations

The developed subroutine UMAT was utilized to perform a numerical simulation of uniaxial tension process, for which purpose a cube with the dimensions of $1 \text{ mm} \times 1 \text{ mm} \times 1 \text{ mm}$ was meshed using a single C3D8 element. The following boundary conditions were applied to the cube's geometry (Fig. 7a): $u_1 = 0$ (the displacement component in direction “1”) on the face $ADA'D'$, $u_2 = 0$ on the face $ABCD$, $u_3 = 0$ on the face $CDC'D'$, and $u_1 = 0.1$ on the face $BCB'C'$.

The distribution of the displacement component u_1 at the maximum stretch is shown in Fig. 7(b). The displacement of the frontal face was applied linearly with time. After reaching the maximum value, unloading was performed. The simulation was performed for two different deformation rates, *i.e.* $\dot{\lambda} = 1 \times 10^{-3} \text{ s}^{-1}$ and $\dot{\lambda} = 1 \times 10^{-2} \text{ s}^{-1}$. In order to verify the results generated by ABAQUS, additional simulations of the same deformation processes were conducted using Scilab software. The simulations performed in Scilab were based on utilizing Eqs. (16)–(19) to calculate the stress for a defined time history of the stretch ratio. An excellent agreement was found between the results generated independently by ABAQUS and Scilab (see Fig. 8). The constant set 2 was used for the simulations (Table 2).

Other simulations of homogeneous deformations were performed using some more complex time histories of the stretch ratio. Fig. 9 depicts the comparison of results ob-

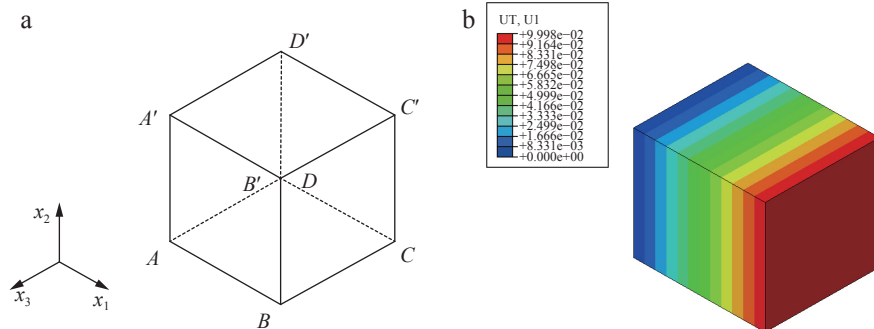


Fig. 7 The homogeneous deformation of single finite element: (a) The coordinate system and the characteristic points; (b) The distribution of displacement component in tension/compression direction

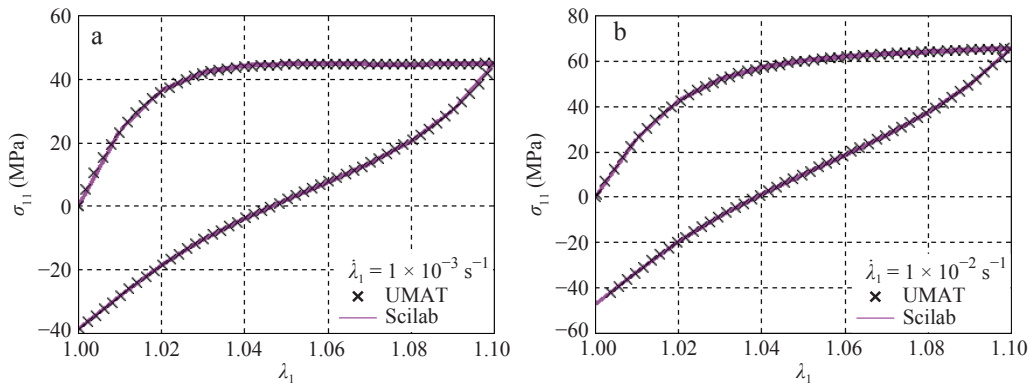


Fig. 8 The polyamide cube undergoing uniaxial loading/unloading with deformation rate of (a) $1 \times 10^{-3} \text{ s}^{-1}$ (b) $1 \times 10^{-2} \text{ s}^{-1}$

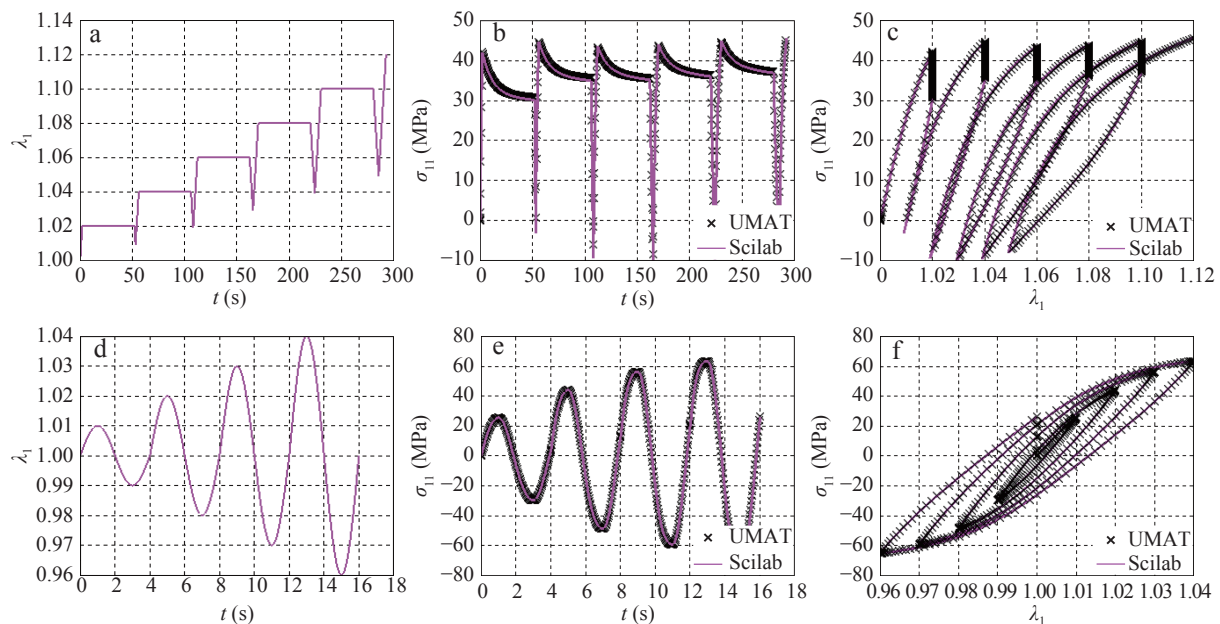


Fig. 9 The polyamide cube undergoing uniaxial tension/compression with the complex loading histories: (a) The axial stretch ratio time history for the multistep relaxation test; (b) The axial stress versus time plot for the multistep relaxation test; (c) The axial stress versus stretch plot for the multistep relaxation test; (d) The axial stretch ratio time history for the cyclic test; (e) The axial stress versus time plot for the cyclic test; (f) The axial stress versus stretch plot for the cyclic test

tained in ABAQUS and Scilab, which are in a perfect agreement. The material parameter set 2 (Table 2) was utilized again.

The FE simulations of the block's deformation were repeated for two different values of the volumetric modulus, *i.e.* $D_1 = 6.74 \times 10^{-4} \text{ MPa}^{-1}$ (as calculated based on the experimental measurements) and $D_1 = 3.30 \times 10^{-7} \text{ MPa}^{-1}$ (which reflects the material near incompressibility). It was found that in the considered case of uniaxial stress states, the differences between results obtained for the two values of volumetric modulus were negligible. This fact supports the proposed method of material parameter identification, which initially makes use of the material incompressibility assumption (with the true value of the volumetric constant being calculated in the next step).

Simulation of Flat Bar Deformation

A simulation of a flat bar in tension is described below as an example of nonhomogeneous deformation. Due to the symmetries in this case, only one-eighth of the bar's geometry

was considered (Fig. 10a). The bar's dimensions are 70 mm $\times$ 70 mm $\times$ 5 mm. There is a centrally placed hole with the radius of 10 mm. The following boundary conditions were defined: $u_1 = 0$ on the face $AA'EE'$, $u_2 = 0$ on the face $BB'CC'$, $u_3 = 0$ on the face $A'B'C'D'E'$. The displacement component u_2 defined on the face $DD'EE'$ was used in the form of a kinematic excitation. The displacement increases linearly until the maximum value of 3.5 mm is reached. Subsequently, it changes with time according to a triangular wave function. The bar's geometry was meshed using C3D8 elements (Fig. 10b). In Fig. 11(a), the contour plot of Huber-von Mises-Hencky (HMH) equivalent stress at the time instant $t = 60 \text{ s}$ is depicted. The distribution of displacement's magnitude at the same time instant can be observed in Fig. 11(b). The time histories of the excitation and the maximum HMH stress are shown in Fig. 12. The simulation was performed using the constant values gathered in set 2 (Table 2) and the volumetric modulus $D_1 = 6.74 \times 10^{-4} \text{ MPa}^{-1}$.

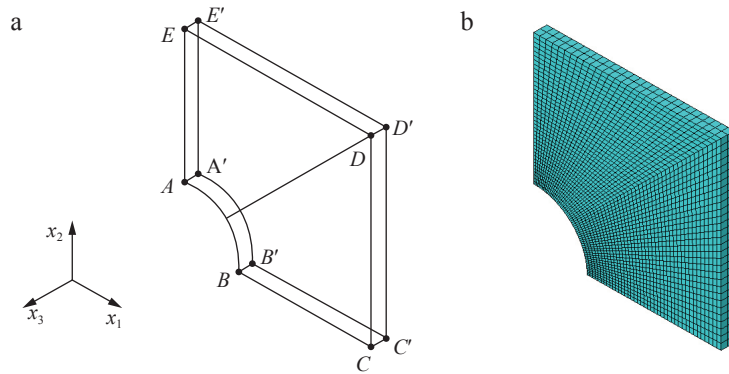


Fig. 10 The flat bar with a hole in tension FE simulation: (a) The coordinate system and characteristic points; (b) The FE mesh (C3D8 elements)

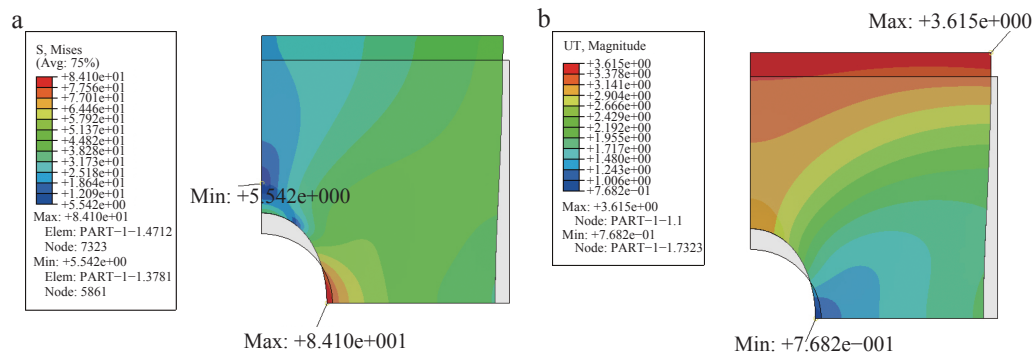


Fig. 11 The flat bar with a hole in tension FE simulation: (a) The contour plot of HMH equivalent stress; (b) The contour plot of displacement vector magnitude

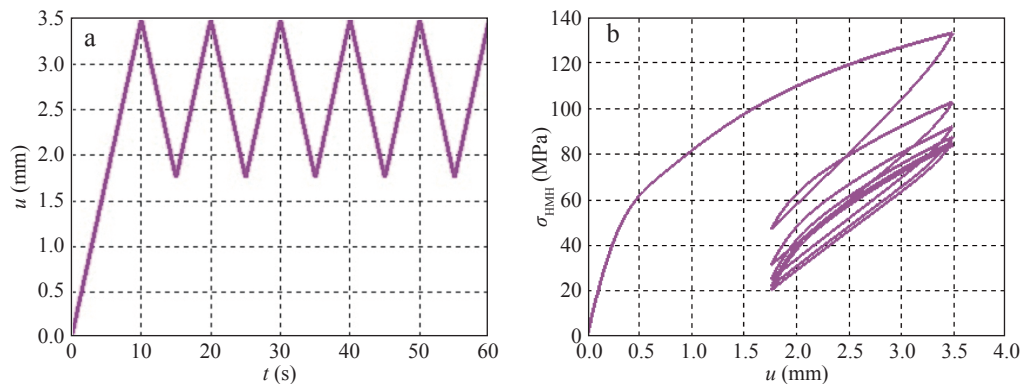


Fig. 12 The flat bar with hole in tension FE simulation: (a) The applied frontal face displacement history; (b) The maximum HMH equivalent stress versus the frontal face displacement

CONCLUSIONS

In this work the inelastic mechanical properties of polyamide 66 were investigated. A thorough experimental study was conducted in order to assess such effects as the hysteretic behavior, the rate-dependence, and the stress relaxation. A constitutive modeling framework was proposed which included a simplified, rate-independent, elastoplastic model and a more sophisticated viscoelastic-plastic constitutive equation.

The material parameter values were determined for the different variants of model and a very good agreement was found between the experimental results and the theoretical

predictions. Upgrading the elastoplastic constitutive law to a viscoelastic-plastic model enables simulating the strain rate dependence and the stress relaxation. This model allows for a good approximation of both the hysteresis and the relaxation experimental data.

The proposed viscoelastic-plastic model was implemented into the FE system ABAQUS by taking advantage of the user subroutine UMAT. The conducted verification tests allow to conclude that the FE implementation of the model was performed correctly. The developed model can be used for the stress and displacement analysis of structures made of polyamide 66 or other thermoplastic polymers.

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