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Multi-mechanism modeling of amorphous polymers

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ABSTRACT

The paper is devoted to a multi-mechanism (MM) model for the mechanical behavior of amorphous glassy polymers. A finite strain formulation through updated Lagrangian formalisms is used. In the proposed phenomenological model, three mechanisms are respectively associated to three physical regimes of plastic deformation. The model was successful in describing the stress–strain behavior of glassy polymers for different strain rates and range of temperatures. The description of the three regions observed in the monotonic stress–strain curves is obtained through a coupling matrix between the isotropic hardening variables. A modular strategy based on the determination of the material parameters in three steps is proposed.

1. Introduction

Structural polymers are frequently used in engineering components. The mechanical behavior of glassy polymers has been the subject of considerable research. The literature is abundant in studies devoted to experimental and numerical investigations of mechanical behavior of these polymers. In the following, works related to semi-crystalline polymers are not discussed. Numerous experimental studies have been carried out on polymers to characterize their mechanical behavior in terms of temperature and strain rate sensitivity (Hope et al., 1980; Richeton et al., 2006; Prasad et al., 2009; Nasraoui et al., 2012; Forquin et al., 2012; Cheng and Ghosh, 2013). Various models were developed and tested to simulate these responses. Rheological models extended to three-dimensional case under finite strain assumption have been proposed, for instance, by Alcoutlabi and Martinez-Vega (2003), Anand and Ames (2006), Dupaix and Boyce (2007), Ames et al. (2009), Anand et al. (2009), Srivastava et al. (2010), Shim and Mohr (2011), Fleischhauer et al. (2012), Helbig and Seelig (2012). Some of them were devoted to phenomenological modeling (Zaïri et al., 2005b; Cheng and Ghosh, 2013) or developed within a thermodynamics framework (Drozdov, 1999; Miehe et al., 2009; Bouvard et al., 2013). Studies devoted to constitutive models including damage involve the works of Zaïri et al. (2005a, 2011), Hocine et al. (2011) among others. Some models are focused on predicting the mechanical behavior of amorphous polymers for a wide range of temperatures and strain rates. A non exhautive list of these studies includes the works of Richeton et al. (2005, 2007). All these models have, as a common root, the separation of stress–strain curve into three regimes (Stachurski, 2003): (i) initially, the deformation is nonlinear until a maximum stress, the yield stress, is reached, (ii) the stress decreases due to softening even in absence of necking (iii) molecular orientation provides a mechanism for hardening that predominates at large strains.

The purpose of this article is to offer a new point of view for the description of the glassy polymer mechanical behavior. The stress strain curve is described using the so called multi-mechanism (MM) approach. This class of models refers also as well to *multi-mechanism*, *multi-phase* or *multi-potential*. The MM model considered in this work refers to "*multi-mechanism*" due to the presence of the three regimes discussed above. The model formulation is detailed in Section 2 within a finite strain framework. Indeed, the loading strain ranges between 10% and 150% in the following simulations. To assess the model reliability, the proposed MM is compared in Sections 3 and 4 with experimental data of polycarbonate (PC) and poly(methyl metacrelate) (PMMA) respectively. The PC and PMMA are, with polystyrene (PS), the more commonly used glassy polymers. The selected tests, taken from the literature, were performed at different strain rates and different temperatures. All the experimental results consist mainly of compressive tests and are not concerned with necking phenomenon according to the cited references. A detailed identification procedure of the material parameters is shown in Section 3. In Section 5, a FE (finite element) simulations of plane-strain forging experiments at 25 °C, under isothermal conditions, were performed on PC specimens as in the work of Srivastava et al. (2010). In that section, the material

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parameters are first identified using the stress–strain curves. The prediction of the model is then compared with experiments in terms of final forged shape.

2. Modeling

The proposed approach is based upon the investigation of multi-mechanism and multi-criteria models. The proposed model is an extension to the use of three mechanisms of previous "two mechanism–two criteria" models. This general framework includes the models adopted by authors like Saï et al. (2004) to predict the stress–strain behavior of metals and applied for the case of polymers by Regrain et al. (2009) to model Polyamide 6 creep behavior and account for the contribution of both amorphous and crystalline phases. A detailed review of such approach can be found in the work of Saï (2011).

The use of a finite strain formulation through updated Lagrangian formalisms is needed to model large-strain deformation of the studied amorphous polymers. The material behavior is based on Green–Naghdi transformation of the stress–strain problem into an "equivalent material referential". This kind of formulation can be applied to materials with tensorial internal variables without modifying the local evolution rules (Ladeveze, 1980). The model is described by:

$$\underset{\sim}{\mathbf{L}} = \underset{\sim}{\dot{\mathbf{F}}}\,\underset{\sim}{\mathbf{F}}^{-1} \qquad \underset{\sim}{\mathbf{D}} = \frac{1}{2}(\underset{\sim}{\mathbf{L}} + \underset{\sim}{\mathbf{L}}^{T}) \qquad \underset{\sim}{\boldsymbol{\Omega}} = \frac{1}{2}(\underset{\sim}{\mathbf{L}} - \underset{\sim}{\mathbf{L}}^{T}) \tag{1}$$

where $\underset{\sim}{\mathbf{F}}$ is the deformation gradient, $\underset{\sim}{\mathbf{L}}$ the rate of deformation, $\underset{\sim}{\mathbf{D}}$ the stretch rate and $\underset{\sim}{\boldsymbol{\Omega}}$ the rotation rate. The stretch rate tensor is transported into a local rotated referential:

$$\underset{\sim}{\dot{\mathbf{e}}} = \underset{\sim}{\mathbf{R}}^{T}\underset{\sim}{\mathbf{D}}\,\underset{\sim}{\mathbf{R}} \tag{2}$$

where the rotation tensor $\underset{\sim}{\mathbf{R}}$ is determined by the polar decomposition of the deformation gradient $\underset{\sim}{\mathbf{F}} = \underset{\sim}{\mathbf{R}}\,\underset{\sim}{\mathbf{U}}$. $\underset{\sim}{\mathbf{R}}$ and $\underset{\sim}{\mathbf{U}}$ describes respectively a pure rotation and a pure stretch tensor. The integrated strain tensor is decomposed into both elastic and inelastic parts. Thanks to updated Lagrangian formulation, constitutive relations can be expressed as in small strain hypothesis. Therefore dealing with the elastic strain tensor is equivalent to a hypoelastic formulation in agreement with a Green–Naghdi stress rate. The stress measure is here the Cauchy stress $\underset{\sim}{\sigma}$ obtained by using the conjugate stress $\underset{\sim}{\mathbf{S}}$ which results from the material behavior integration:

$$\underset{\sim}{\sigma} = \det{}^{-1}(\underset{\sim}{\mathbf{F}})\underset{\sim}{\mathbf{R}}\,\underset{\sim}{\mathbf{S}}\,\underset{\sim}{\mathbf{R}}^{T} \tag{3}$$

Under the small deformation assumption, the total strain can be decomposed into an elastic part and an inelastic one: $\underset{\sim}{\varepsilon} = \underset{\sim}{\varepsilon}_{el} + \underset{\sim}{\varepsilon}_{in}$. The elastic strain $\underset{\sim}{\varepsilon}_{el}$ is calculated thanks to the generalized Hooke's law:$\underset{\sim}{\sigma} = \underset{\approx}{\boldsymbol{\Lambda}} : \underset{\sim}{\varepsilon}_{el}$ where $\underset{\sim}{\sigma}$ is the Cauchy stress tensor or macroscopic stress resulting from the material behavior integration and $\underset{\approx}{\boldsymbol{\Lambda}}$ is the fourth-rank tensor of elastic moduli. The inelastic strain $\underset{\sim}{\varepsilon}_{in}$ is the sum of the irreversible deformations of the three observed mechanisms: $\underset{\sim}{\varepsilon}_{in} = \underset{\sim}{\varepsilon}_{v1} + \underset{\sim}{\varepsilon}_{v2} + \underset{\sim}{\varepsilon}_{v3}$.

In the MM models, each mechanism I is associated to a stress tensor $\underset{\sim}{\sigma}_I$ calculated from a stress concentration rule. Several approaches were used to obtain $\underset{\sim}{\sigma}_I$ and strains through different transition rules. It is assumed in the present work that the macroscopic stress $\underset{\sim}{\sigma}$ is equal to the individual stress for the three mechanisms: $\underset{\sim}{\sigma} = \underset{\sim}{\sigma}_1 = \underset{\sim}{\sigma}_2 = \underset{\sim}{\sigma}_3$. The construction of a constitutive model is in general based on the definition of a yield function, a flow potential and a hardening potential. The proposed model belongs to the MM class model in which each mechanism has its own plastic multiplier. Each stress tensor ($\underset{\sim}{\sigma}_1$, $\underset{\sim}{\sigma}_2$ and $\underset{\sim}{\sigma}_3$) is involved in a yield function (f_1, f_2 and f_3) to define the elastic domain related to each mechanism. Since the present study is only dedicated to polymer behavior under monotonic loading, the kinematic hardening variables were not considered.

$$f_1 = J(\underset{\sim}{\sigma}_1) - R_1 - R_{01} \qquad f_2 = J(\underset{\sim}{\sigma}_2) - R_2 - R_{02}$$
$$f_3 = J(\underset{\sim}{\sigma}_3) - R_3 - R_{03} \tag{4}$$

where $J(\underset{\sim}{\sigma}) = \sqrt{1.5\underset{\sim}{s} : \underset{\sim}{s}}$ and $\underset{\sim}{s}$ is the deviatoric part of the tensor $\underset{\sim}{\sigma}$. R_{01}, R_{02} and R_{03} denotes the initial size of the elastic domain for the mechanism 1, 2 and 3 respectively. The isotropic hardening variables (R_1, R_2 and R_3) that characterize the size change in the elastic domain related to each yield surface are described by three internal variables (r_1, r_2 and r_3) respectively. In this formulation, an isotropic-isotropic coupling has been introduced between the hardening variables to account for the interaction between the different mechanisms:

$$\begin{pmatrix} R_1 \\ R_2 \\ R_3 \end{pmatrix} = \begin{pmatrix} Q_{11} & Q_{12} & Q_{13} \\ Q_{21} & Q_{22} & Q_{23} \\ Q_{31} & Q_{32} & Q_{33} \end{pmatrix} \begin{pmatrix} r_1 \\ r_2 \\ r_3 \end{pmatrix} \tag{5}$$

Q_{11}, Q_{22} and Q_{33} are isotropic hardening modulus. (Q_{IJ}, $I \neq J$) are coupling parameters that play an essential role in the polymer behavior. For example, a positive value of Q_{12} allows to the mechanism 2 (if active) to attenuate the mechanism 1. Increasing Q_{12} leads to desactivate the mechanism 1. Inversely, a negative value of Q_{21} allows to a premature activation of the mechanism 2 caused by the mechanism 1. This feature can be helpful, in the case of the studied materials, to activate the third mechanism only after the softening stage. Previous works dealing with MM models have shown that the kinematic coupling allows to describe several mechanical effects (i.e. rate-sensitivity, plasticity–creep interaction, ratcheting, etc.). According to the normality flow rule, the viscoplastic strain rates may be expressed as:

$$\underset{\sim}{\dot{\varepsilon}}_{v1} = \dot{v}_1\underset{\sim}{\mathbf{n}}_1 \qquad \underset{\sim}{\dot{\varepsilon}}_{v2} = \dot{v}_2\underset{\sim}{\mathbf{n}}_2 \qquad \underset{\sim}{\dot{\varepsilon}}_{v3} = \dot{v}_3\underset{\sim}{\mathbf{n}}_3 \tag{6}$$

$\dot{v}_1$, $\dot{v}_2$ and $\dot{v}_3$ are three viscoplastic multipliers and are the driving forces for the evolution laws. They can be written using sinh rules to account for the strong nonlinearity of the creep rate as a function of the stress

$$\dot{v}_I = \dot{v}_{0I}\sinh\left(\langle\frac{f_I}{K_I}\rangle^{n_I}\right) \qquad I = 1, 3 \tag{7}$$

The McCauley-brackets $\langle\rangle$ denotes the positive part: $\langle x\rangle=0$ if $x<0$ and x otherwise. K_1, n_1, K_2, n_2, K_3 and n_3 are viscoplastic parameters related to the viscosity of the different mechanisms. The evolution laws of the isotropic hardening variable are given by:

$$\dot{r}_1 = \dot{v}_1(1 - b_1r_1) \qquad \dot{r}_2 = \dot{v}_2(1 - b_2r_2) \qquad \dot{r}_3 = \dot{v}_3(1 - b_3r_3) \tag{8}$$

It is worth noting that the evolution rules of the isotropic internal variables was intentionally simplified compared to the evolution rules that come from thermodynamical consideration as demonstrated in the work of Saï (2011) in which "flow" coupling is considered in addition to the "state" coupling (Eq. (5)):

$$\dot{r}_1 = \dot{v}_1\left(1 - \frac{b_1R_1}{Q_{11}}\right) \qquad \dot{r}_2 = \dot{v}_2\left(1 - \frac{b_2R_2}{Q_{22}}\right)$$
$$\dot{r}_3 = \dot{v}_3\left(1 - \frac{b_3R_3}{Q_{33}}\right) \tag{9}$$

In this work, using the isotropic evolution rules Eq. (8), coupling between the three mechanisms results then only from Eq. (5).

The MM model is implemented into the material library ZMaT of the FE code ZSeT (Besson et al., 1998), using a θ-method solved