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C. R. Mecanique 333 (2005) 359–364



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Phenomenological nonlinear modelling of glassy polymers

Fahmi Zaïri ^{a,*}, Krzysztof Woznica ^b, Moussa Naït-Abdelaziz ^a

^a *Université des sciences et technologies de Lille, École polytechnique universitaire de Lille, laboratoire de mécanique de Lille (UMR CNRS 8107), avenue P. Langevin, 59655 Villeneuve d'Ascq cedex, France*

^b *Laboratoire énergétique explosions structures, École nationale supérieure d'ingénieurs de Bourges, 10, boulevard Lahitolle, 18020 Bourges cedex, France*

Received 31 July 2004; accepted 26 January 2005

Presented by Évariste Sanchez-Palencia

Abstract

Compared with the numerous works into the constitutive equations for the mechanical behaviour of metals, very little attention has been devoted to those of polymers. However, a model is required to describe both the complex shape of the stress–strain curves and strain rate sensitivity of glassy polymers. In this Note, a unified viscoelastic-viscoplastic model is presented in which the nonlinear pre-yield behaviour, the strain softening and the strain hardening are described by internal variables, in analogy with the models developed for metals. In order to check the predictive capability of the model, the numerical results are compared with the experimental data (monotone, creep and relaxation tests) of a typical amorphous glassy polymer. **To cite this article:** *F. Zaïri et al., C. R. Mecanique 333 (2005).*

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Résumé

Modélisation phénoménologique non-linéaire pour polymères vitreux. En comparaison avec les nombreux travaux dévolus aux équations constitutives pour le comportement mécanique des métaux, très peu ont été consacré aux polymères. Cependant, un modèle est nécessaire pour décrire la forme complexe des courbes contrainte–déformation et la sensibilité à la vitesse de déformation des polymères vitreux. Dans cet article, un modèle viscoélastique-viscoplastique unifié est présenté dans lequel la non-linéarité avant le pic, l'adoucissement et le durcissement sont décrits par des variables internes en analogie avec les modèles développés pour les métaux. Afin de vérifier les possibilités prédictives du modèle, les résultats numériques sont comparées aux données expérimentales (essais monotones, fluage et relaxation) d'un polymère amorphe vitreux typique. **Pour citer cet article :** *F. Zaïri et al., C. R. Mecanique 333 (2005).*

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Keywords: Computational solid mechanics; Viscoplasticity; Viscoelasticity; Nonlinear behaviour; Glassy polymers

Mots-clés: Mécanique des solides numérique ; Viscoplasticité ; Viscoélasticité ; Comportement non-linéaire ; Polymères vitreux

* Corresponding author.

E-mail address: fahmi.zairi@polytech-lille.fr (F. Zaïri).

Version française abrégée

Dans ce travail, nous proposons une approche phénoménologique de la représentation du comportement fortement non-linéaire et de la dépendance en temps des polymères vitreux.

La vitesse de déformation est décomposée de manière additive en trois parties : $\mathbf{D} = \mathbf{D}^e + \mathbf{D}^{ve} + \mathbf{D}^{vp}$. La partie élastique \mathbf{D}^e suit une loi de Hooke. Les parties viscoélastique \mathbf{D}^{ve} et viscoplastique \mathbf{D}^{vp} sont modélisées à partir d'un modèle unifié [7] utilisant des variables internes simulant la résistance à l'écoulement moléculaire. Dans ce travail, une version modifiée [3,4] du modèle de Bodner et Partom [7], originalement développé pour l'étude de la viscoplasticité des métaux, est utilisée pour représenter le comportement fortement non-linéaire après le pic de contrainte (Éq. (1)). Une variable interne Z_2 , représentant le durcissement et initialement introduit dans [3], a été superposée, dans un précédent travail [4], à une autre variable Z_1 représentant l'adoucissement. La non-linéarité avant le pic de contrainte est modélisée dans ce travail en utilisant le modèle de Bodner et Partom [7] dans sa version originale (Éq. (3)), qui introduit une variable interne supplémentaire Z_3 . Le modèle viscoélastique-viscoplastique résultant est capable de prédire la dépendance à la vitesse de déformation et le comportement typique des polymères vitreux : non-linéarité avant le seuil (Fig. 1), adoucissement et durcissement (Fig. 2).

Une méthode d'identification analytique, que nous avons développée dans un précédent article [4] pour le modèle viscoplastique modifié et que nous avons adapté dans ce travail pour le modèle couplé, permet de remonter à la valeur des paramètres constitutifs. Les équations du modèle viscoélastique-viscoplastique sont intégrées numériquement à l'aide d'un algorithme itératif explicite.

Des essais expérimentaux en traction sur un polycarbonate (PC) ont été réalisés à différentes vitesses de déformation vraies et à température ambiante (Fig. 2) en utilisant un système d'acquisition et de régulation VideoTraction[®]. En complément, des essais de fluage (Fig. 3) et de relaxation (Fig. 4) ont été réalisés avec le même dispositif.

En utilisant la méthode d'identification élaborée, les paramètres constitutifs du PC ont pu être déterminés (Tableau 1). La Fig. 1 montre l'aptitude du modèle à pouvoir appréhender le comportement non-linéaire avant le pic de contrainte. La comparaison entre les prédictions du modèle et les données expérimentales pour trois vitesses de déformation et à température ambiante est réalisée à la Fig. 2. Les prédictions en fluage et en relaxation sont également comparées aux données expérimentales du PC aux Fig. 3 et 4 respectivement. Le modèle viscoplastique modifié associé à la version original du modèle de Bodner et Partom apparaît comme étant assez flexible pour la représentation de la réponse (monotone, fluage et relaxation) de ce polymère vitreux typique.

1. Introduction

The cohesive deformation of ductile amorphous glassy polymers is known to occur by nucleation and growth of shear bands, and to exhibit time dependent behaviour and highly nonlinear response. Macroscopically, the deformation is dominated by an intrinsic post-yield nonlinear behaviour characterised by an intrinsic strain softening immediately after yield and a subsequent re-hardening. In contrast to extensive studies on constitutive modelling of metals, glassy polymers have received little attention. However, we can cite Haward and Thackray [1] who have proposed the first nonlinear model for glassy polymers. Their one-dimensional model connects a model of the rubber-elasticity theory describing the strain hardening with a yielding model. Several refinements of this model have been proposed since in order for example to incorporate the strain softening or to propose a three-dimensional formulation [2]. Although these physical models are in agreement with some polymers, it is potentially difficult to obtain material parameters. Therefore, another approach based on the phenomenological level can be considered by using constitutive equations developed for metals. Indeed, on a macroscopic point of view, although the deformation mechanisms of polymers and metals are different, viscoplastic constitutive equations originally developed for metals can be used to model the strain rate dependent and nonlinear behaviour of glassy polymers [3,4]. Furthermore, glassy polymers are known to have a nonlinear behaviour before the peak and this one can be also modelled with the same approach [5,6].

This Note aims to present a study of the nonlinear viscoelastic and viscoplastic behaviour of glassy polymers on a phenomenological point of view. To predict their stress–strain response under different strain rates, creep and relaxation behaviour, we present a modified viscoplastic model combined with a viscoelastic model, developed initially for metals. Tests were carried out for one representative amorphous glassy polymer, a polycarbonate (PC), on an electro-mechanical Instron device. The implementation of the model shows that the predictions of this one correctly describe experimental data of PC tested in tension at different strain rates, in creep and in relaxation.

2. A viscoelastic-viscoplastic model

The theory of nonlinear behaviour of polymers is still an active research area. This section aims to present nonlinear unified constitutive equations containing time dependent relations.

The strain rate is assumed to be the sum of rate-independent (elastic \mathbf{D}^e) and rate-dependent parts (viscoelastic \mathbf{D}^{ve} and viscoplastic \mathbf{D}^{vp}): $\mathbf{D} = \mathbf{D}^e + \mathbf{D}^{ve} + \mathbf{D}^{vp}$. The rate-independent part \mathbf{D}^e follows Hooke's law and the nonlinearity is included both in viscoplastic and viscoelastic terms (boldface letters refer to second order tensors).

2.1. Viscoplastic part

An unified constitutive model using internal variables, simulating the resistance to molecular flow, is used for the viscoplastic part of the deformation. The Bodner and Partom model [7] constitutes a state variable approach with no yield surface in which the viscoplasticity contribution exists at all non zero stress levels, and it is found to be the more adequate for the viscoplastic part. This model is modified in order to include strain softening immediately after yield and subsequent re-hardening:

$$\mathbf{D}^{vp} = \sqrt{3}D_0 \left(\frac{\sigma_e}{Z_1 + Z_2} \right)^{2n} \frac{\boldsymbol{\sigma}'}{\sigma_e} \quad (1)$$

where $\sigma_e = (3/2\boldsymbol{\sigma}' : \boldsymbol{\sigma}')^{1/2}$ is the von Mises equivalent stress, $\boldsymbol{\sigma}' = \boldsymbol{\sigma} - 1/3\text{tr}(\boldsymbol{\sigma})\mathbf{I}$ is the deviatoric part of the Caychy stress $\boldsymbol{\sigma}$.

In Eq. (1), the internal state variable Z_1 was initially introduced by Frank and Brockman [3] to account for the horizontal plateau upon yield and the subsequent strain hardening exhibited by a glassy polymer. In a previous paper [4], Z_1 was simplified and we have introduced an internal variable Z_2 to account for the effect of strain softening.

First-order differential equations of the two internal variables Z_1 and Z_2 are given by:

$$\dot{Z}_1 = m \left(\frac{Z_1 - (1 - \alpha)Z_{10}}{Z_{10}} \right) \dot{W}^p \quad \text{and} \quad \dot{Z}_2 = h(Z_{2s} - Z_2) \dot{W}^p - AZ_{2s} \left(\frac{Z_2 - Z_{2c}}{Z_{2s}} \right)^r \quad (2)$$

In Eq. (2), $\dot{W}^p = \boldsymbol{\sigma} : (\mathbf{D} - \mathbf{D}^e)$ is the inelastic work rate and the associated rise of temperature is neglected, D_0 is the limiting shear strain rate, n is the strain rate sensitivity parameter controlling the viscosity of flow, Z_{10} is the initial value of Z_1 taken to represent the onset of the plasticity, Z_{2s} is the saturation value of Z_2 , m and h are the hardening and softening rate parameters, respectively, α is a parameter controlling the onset of the re-hardening, A , Z_{2c} and r are three parameters introduced here for the static recovery.

2.2. Viscoelastic part

The nonlinearity is also included in the viscoelastic part. Models originally developed for metals can be also used to describe the nonlinear pre-yield behaviour of polymers [5,6]. The unmodified Bodner and Partom model [7], used to describe the nonlinear pre-peak viscoelastic behaviour (Fig. 1), can be expressed as:

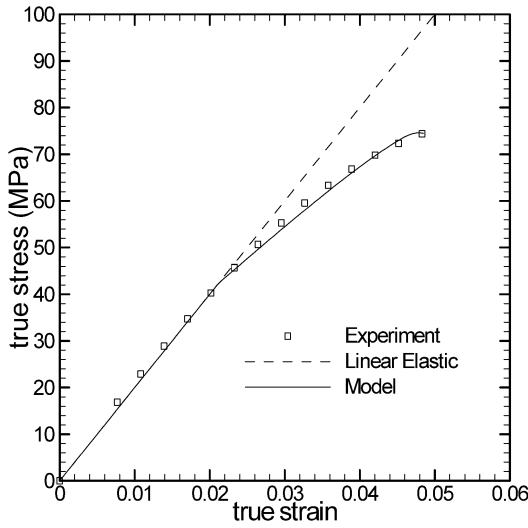


Fig. 1. Nonlinear pre-yield behaviour for PC at 10^{-2} s^{-1} and at room temperature.

Fig. 1. Comportement non-linéaire avant le pic de contrainte du PC à 10^{-2} s^{-1} et à température ambiante.

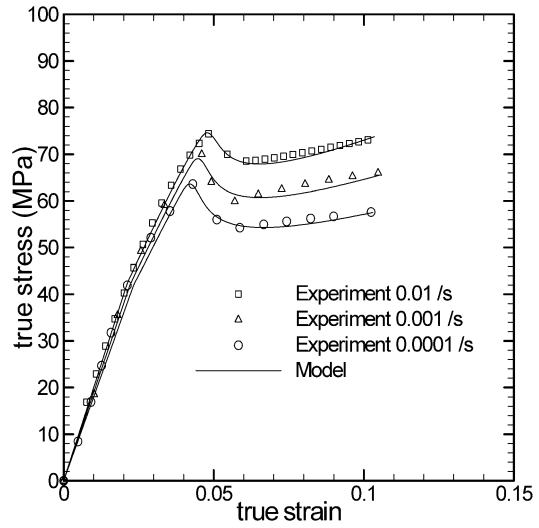


Fig. 2. Tensile stress–strain curves for PC at different strain rates and at room temperature.

Fig. 2. Courbes contrainte–déformation du PC à différentes vitesses de déformation et à température ambiante.

$$\mathbf{D}^{ve} = \sqrt{3} D_0 \exp\left(-\frac{1}{2} \left(\frac{Z_3}{\sigma_e}\right)^{2n}\right) \frac{\sigma'}{\sigma_e} \tag{3}$$

where Z_3 is an added internal state variable and the remaining terms are as defined earlier. Z_3 is governed by the following differential equation:

$$\dot{Z}_3 = q(Z_{3s} - Z_3)\dot{W}^p \tag{4}$$

In Eq. (4) q is the pre-yield hardening rate parameter, Z_{3s} is the saturation value of Z_3 and the initial value of Z_3 is defined by the parameter Z_{30} .

The resulting set of viscoelastic-viscoplastic constitutive equations is able to predict the strain rate dependent behaviour, nonlinear pre-yield behaviour, intrinsic strain softening and subsequent strain hardening of glassy polymers.

2.3. Determination of material parameters

According to the materials and the thermal conditions, the material constants that need to be determined include : E , D_0 , n , Z_{10} , m , h , Z_{2s} , α , q , Z_{3s} , Z_{30} , A , Z_{2c} and r . For identifying these parameters and validating the modified model by experiments, a one-dimensional analytical method was developed. A description of the systematic method for the viscoplastic model parameters determination can be found in a previous paper [4]. However, a summary is given here. The Young’s modulus E is determined from the initial linear region of the stress–strain curves (noticed, for the material studied in this work, a weak dependence of E on the strain rate (Table 1)). The limit D_0 , which can be physically related to Orowan’s equation [7], is fixed arbitrarily [9]. The constants n , Z_{10} and Z_{3s} are determined from a least squares regression analysis of the peak stress versus the strain rate. Z_{30} is determined with the same method but with the stress, corresponding to the beginning of the nonlinear pre-yield behaviour, plotted versus the strain rate. q is calibrated with experimental data before the peak. The parameters h , m , Z_{2s} and α are determined from the work hardening rate defined as the derivative of the stress with the plastic work. The hardening parameters h and m are determined from the negative and positive slope, respectively,

Table 1
Parameter values for PC tested at room temperature

Tableau 1
Valeur des paramètres du PC testé à température ambiante

E MPa			D_0 s^{-1}	n –	q –	Z_{30} MPa	Z_{3s} MPa	Z_{10} MPa	m –	h –	Z_{2s} MPa	α –	A –	Z_{2c} MPa	r –
2000 ($10^{-2} s^{-1}$)	1900 ($10^{-3} s^{-1}$)	1790 ($10^{-4} s^{-1}$)	10^4	10.3	2.5	50	100	176.5	6.5	–1.25	–60	0.9	0.0035	20	2

of the work hardening rate plotted with the true stress. Z_{2s} and α are determined from a work hardening rate value [4]. The parameters A , Z_{2c} and r are calibrated with the stress-time and strain-time curves. If necessary certain hardening parameters are later readjusted to obtain a better fit to the stress–strain curves.

2.4. Numerical integration

It is necessary to integrate in time the rate form of the viscoelastic-viscoplastic constitutive law to obtain strain. To facilitate a possible finite element code implementation, an explicit numerical integration scheme is developed using a Fortran program. Because of its simplicity for updating unknowns over a loading increment, the first order trapezoidal method was found the most suitable. Using the Young's modulus, the current value of both the total strains and the viscoelastic-viscoplastic strains, the stresses are obtained.

3. Comparison between predicted and measured mechanical response

3.1. Experimental procedure and results

A typical glassy polymer, the polycarbonate (PC), was selected for investigating its general response. This polymer was chosen because its plastic behaviour is known to be exclusively controlled by isochoric shear banding mechanisms. Uniaxial tension tests were conducted on an electro-mechanical Instron machine for various true strain rates to obtain stress–strain curves (Fig. 2). In order to localize the deformation in the center, the geometry of the uniaxial tension specimens contains a radius of curvature and in this region two black round markers are made on the front surface and are aligned along the tensile axis. During the test, these markers are analyzed by a VideoTraction[®] system controller and by regulating the cross-head speed, local true strain rate was performed to remain constant.

Furthermore, the strain-time and stress-time response measurements during creep and relaxation, respectively, have been also performed (Fig. 3 and 4) using the electro-mechanical Instron machine. The specimens and the experimental protocol are the same as those used in static tension loading.

It is important to note that tests realized with four markers (two are aligned along the tensile axis and two others along the transversal axis) or seven markers [8] (five along the tensile axis and two others along the transversal axis) reveal that shear bands nucleation really starts, for this material, at the beginning of the nonlinear behaviour before the peak. Therefore, for a description more realistic, the yield must be considered at this instant and the complete model presented in this work takes into account this aspect of pre-plasticity.

3.2. Model validation

Using the procedure we have developed, the values of the parameters obtained are given in the Table 1. Fig. 1 permits us to verify the ability of the model to predict the nonlinearity before the peak. In Fig. 2, the numerical predictions of the model are generated and compared with experimental data for three strain rates and at room temperature. These comparisons show that the model accurately predicts the behaviour of this material and the

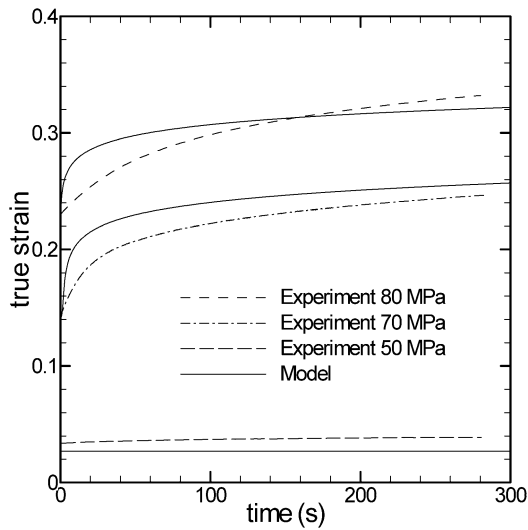


Fig. 3. Creep behaviour for PC with different stress levels.

Fig. 3. Comportement en fluage du PC avec différents niveaux de contraintes.

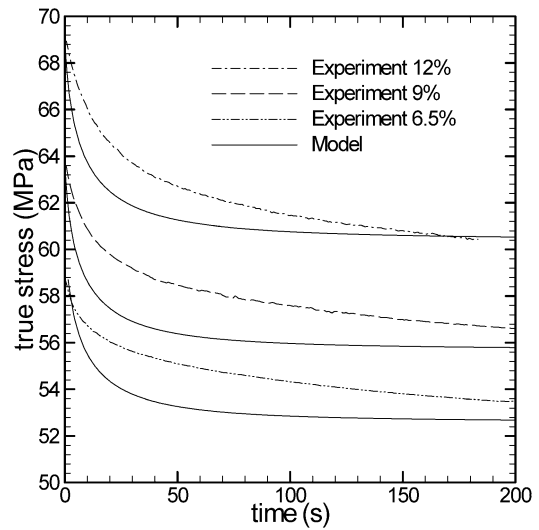


Fig. 4. Loading at a true strain rate of 10^{-3} s^{-1} followed by stress relaxation with different strain levels.

Fig. 4. Chargement à une vitesse de déformation vraie de 10^{-3} s^{-1} suivi de la relaxation de la contrainte avec différents niveaux de déformation.

pertinence of the coupling. The comparison of the predicted behaviour given by the model with the strain-time and stress-time response is made in Fig. 3 and 4, respectively and the agreement is reasonably good.

4. Conclusion

Macroscopic constitutive model originally based on the dislocation theory in metals is used to capture the global behaviour of a polymeric material. For representing the nonlinear behaviour and the effect of rate, a single rate independent set parameters is determined from an original procedure. The modified Bodner and Partom viscoplastic model associated with the original version was shown sufficient flexibility to permit modelling of the representative amorphous glassy polymer response, such as strain rate sensitivity, nonlinear behaviour, creep and relaxation.

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