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Modeling of the mechanical behavior of amorphous glassy polymer based on the quasi-point defect theory—Part II: 3D formulation and finite element modeling of polycarbonate

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ABSTRACT

A three-dimensional formulation of the quasi-point defect theory has been developed and the corresponding constitutive equations have been implemented in the finite element package Abaqus, via the writing of an UMAT file. The proposed tool has been completed to support the large strain description and thus can perform a general structure calculation. Referring to a unique set of parameters identified in the part I of this paper for glassy amorphous bisphenol-A polycarbonate (PC-BPA), various commonly used tests were then considered as structural patterns and modeled. The whole mechanical response as well as localization phenomena measured via video technique is well predicted by the calculations. These results also highlight the need to consider the spatial dimension in a sense that the mechanical behavior up to large strain is related to structural modifications, especially for materials such as amorphous polymers that exhibit yield, softening and hardening intrinsic phenomena.

1. Introduction

Until now, the conception procedure of thermoplastic polymer components for industrial applications based on finite elements calculation suffered from the lack of adapted and robust constitutive equations to describe the peculiar mechanical behavior of this kind of material. Indeed, its complex intrinsic behavior is hardly reproduced in a wide set of external conditions. Two different types of models have been developed up to an implementation in a finite element code in order to describe such behavior.

The first type of models is purely phenomenological. Depending on the complexity of the proposed equations and of the number of used parameters, these models can simulate a more or less large number of experimental conditions, and the assumptions embedded are related to the effects focused on. For instance, Lu and Ravi-Chandar [1] consider an elasto-plastic description via a trilinear model to illustrate the localization phenomena. In many efforts, elasto-viscoplastic description of amorphous polymers are developed for FE calculations [2,3]. Finally, authors of Refs. [4–6] implement a viscoelastic–viscoplastic phenomenological description in their FE code. In those theories, we can point out the lack of physical significance and the possible non-uniqueness of the parameters, even for the most complex and therefore the most complete models. Those limitations compromise the validity of the equations as soon as the experimental conditions are enlarged.

The second type of models aims to consider the microstructure of the polymer (even though phenomenological concepts are included to a certain extend). Many theories consider two distinct internal physical contributions to the total resistance to deformation as initially proposed by Haward and Trackay [7]. The first one is a resistance to flow based on a microstructural scenario [8–10] that leads to the prediction of the yield phenomena. In addition to this flow rule, a network resistance is added, leading to the strain hardening and allowing the model to account for the large strain behavior. This was done by making the analogy between the elasticity of a cross-linked network of flexible chains (rubber) and the hardening of an entangled network of chains that are (locally) relaxed by virtue of shear yielding (hardening in an amorphous glass). More or less refinement in the definition of the active chains, i.e. the number of active polymer “crosslinks” in the strained material has then been considered [11–15]. However, it is noteworthy that most of these models do not propose a global vision of the whole viscoelastic–viscoplastic behavior of amorphous polymer, being essentially interested in its viscoplastic aspect. Recently, Anand and Ames [16] proposed a generalization of this approach. In his model, a set of Kelvin–Voigt elements accounts for the inelastic micro-mechanisms and 31 parameters mostly phenomenological need to be determined. This illustrates the complexity for

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describing the numerous peculiarities of the mechanical behavior of amorphous polymers in their glassy state.

Contrary to these approaches, the quasi-point defect (QPD) theory [17–20] describes the mechanical behavior of amorphous polymer in the linear as well as non-linear domains. This model is suited for all the non-crystalline solids at temperatures below and in the vicinity of the glass transition temperature and has shown its efficiency to simulate a large set of experimental conditions. In the present paper, we will briefly recall the main equations of the physical-based model. A complete justification of the following set of equations can be found in the companion paper [21]. These equations are then introduced in a 3D formalism and implemented in the FE code Abaqus [22]. Finally, finite elements simulations of different types of mechanical tests are performed to illustrate the efficiency of this new tool. Because typical glassy behavior exhibiting yield, softening and hardening is known to favor strain localization and/or stabilization (see [2,11,23,24] and reference herein); the validation will be performed with the comparison between experimental data and calculation at both global and local levels.

2. Constitutive equations

The constitutive equations of amorphous polymers are derived from the QPD theory [20] and are presented in details and for the first time in the Part I of this paper. Based on a microstructural scenario, this model provides the kinetic equation for the non-elastic shear strain rate of thermoplastic polymer under a thermo-mechanical stimulus. The non-elastic deformation is described through localized and correlated microstructural movements, as widely acknowledged in the literature [25–28]. Moreover, a reversible (so-called anelastic) and a permanent component (viscoplastic) can be dissociated. The distributed characteristic times, associated to the anelastic part, correspond to those involved in the β and low temperature part of the α relaxations observed in DMA experiments (depicted β and *an*, respectively). The distributed viscoplastic characteristic times (vp) are those responsible for the upper part of the α relaxation. Finally, three populations of characteristic times lead to the general formulation of the non-elastic shear strain rate $\dot{\gamma}_{nel}$

$$\dot{\gamma}_{nel}(t) = \sum_{n} \dot{\gamma}_n(t) = \sum_{n} \sum_{i} \frac{{}^{\infty}\gamma_n^{i(t)-} \gamma_n^i(t)}{\tau_n^i(t)} \tag{1}$$

where the i subscript corresponds to the ith distributed element of the nth contribution and the n subscript corresponds to the process involved, either the β, *an* or *vp* one. The equilibrium shear strain of each process is defined by a limited strain value ${}^{\infty}\gamma_n^{i(t)}$. It is function of the activation shear stress σ_a and the compliance increase ΔJ_n associated to the corresponding relaxation process

$${}^{\infty}\gamma_n^i(t) = W_n^i \Delta J_n \sigma_a(t) \tag{2}$$

The distribution weights W_n^i as well as the associated relaxation times τ_n^i are defined in Appendix A1.

The microstructural state is taken into account through a so-called correlation parameter χ that is introduced in the equation defining the relaxation times. Moreover, this parameter depends on the temperature (Eq. (3)) and evolves with the non-elastic deformation (Eq. (4))

$$\begin{aligned} \chi(T) &= \chi(T_g) & T &< T_g \\ \chi(T) &= \chi(T_g) + A_\chi(T - T_g) & T &> T_g \end{aligned} \tag{3}$$

$$\chi(T, \varepsilon) = \chi(T) + A_{an}(\gamma_{an}) - A_{vp}\gamma_{vp} \tag{4}$$

The different parameters A_χ, A_{an} and A_{vp} are given in Table 1.

Table 1
Parameters of the QPD model.

Parameters	***Values***	***Units***	***Definition***
Linear behavior (DMA tests)			
Secondary relaxation			
$\boldsymbol{G_{\beta\_u}}$	**1296**	MPa	Storage shear modulus before the β relaxation $(=1/\mathbf{J_{el}})$
$\boldsymbol{G_{\beta\_r}}$	**648**	MPa	Relaxed shear storage modulus of the β relaxation $(=\mathbf{G_{\alpha\_u}})$
$\boldsymbol{U_\beta}$	**0.8**	eV	Activation energy of the β relaxation
$\boldsymbol{\tau_{\beta 0}}$	$\mathbf{10^{-21}}$	s	Pre-exponential factor of the β relaxation
$\boldsymbol{B_\beta}$	**0.1**		Distribution parameter
Main relaxation			
$\boldsymbol{\Delta G_{an}}$	**213.3**	MPa	Intensity of the anelastic process $(=1/\boldsymbol{\Delta J_{an}})$
$\boldsymbol{G_{\alpha\_r}}$	**1.8**	MPa	Relaxed shear storage modulus of the β relaxation
$\boldsymbol{t_0}$	$\mathbf{5 \times 10^{-21}}$	s	Time scale parameter related to the shift between the two main relaxations
$\boldsymbol{\chi}$	**0.3**		Disorder parameter
$\boldsymbol{\chi'}$	**0.95**		Distribution parameter of the viscoplasticity
$\boldsymbol{A_\chi}$	**0.0065**		Temperature contribution to the disordered state (up to T_g)
Non-linear behavior (finite strain tests)			
$\boldsymbol{\sigma_0}$	**185**	MPa	Yield stress to overcome the energy barrier for conformational change at 0 K
$\boldsymbol{A_{an}}$	**0.9**		Anelastic contribution to the disordered state (related to the softening)
$\boldsymbol{A_{vp}}$	**0.1**		Viscoplastic contribution to the disordered state (related to the plateau value)
$\boldsymbol{\Omega}$	**0.001**	MPa^{-1}	Pressure dependence
Limited extensibility criterion			
$\boldsymbol{\varepsilon_{eq}}$	**0.55**		Equivalent limited strain
“hardening modulus”	**400**	MPa	Elastic constant for the elastic behavior assumed once the limited strain has been reached

Note that $\boldsymbol{\Delta J_{vp}} = (1/\boldsymbol{G_{\alpha\_r}} - 1/\boldsymbol{G_{\alpha\_u}}) - \boldsymbol{\Delta J_{an}}$ and $\Delta \boldsymbol{J_\beta} = (1/\boldsymbol{G_{\beta\_r}} - 1/\boldsymbol{G_{\beta\_u}})$.

This model makes a strong link between the viscoelastic behavior of the material, characterized by DMA and the mechanical finite deformation behavior generally characterized by classical mechanical tests (tensile, compression and shear tests). Finally, with the set of constitutive equations recalled above and in the Appendix A1, the QPD model gives a global description of an amorphous polymer from the linear to the non-linear range with account for temperature, loading conditions and strain rate or pulsation. Its validation in simple uniaxial stress states (assumed homogeneous) has shown its limitation (see part I) when localization phenomenon becomes important. Its 3D generalization and calculations accounting for the spatial dimensions are then needed.

3. 3D generalization

3.1. Constitutive equations

The general three-dimensional problem involves large transformations including large deformations and large rotations. In such conditions, the kinematics formulation is based on the Kroner–Lee multiplicative decomposition [29,30] of the