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Structural versus microstructural evolution of semi-crystalline polymers during necking under tension: Influence of the skin-core effects, the relative humidity and the strain rate

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ABSTRACT

This study deals with the appearance of heterogeneity of strain and stress in a uniaxial specimen deformed by extension. For this purpose, two semi-crystalline polymers (an isotactic polypropylene and a polyamide 6), were selected: flat geometries showing a marked microstructural skin-core effect for the PP and homogeneous round bars for PA6 which were systematically compared during tensile loading. The specimens underwent necking at various strain rates and two relative humidities. The minimum neck curvature radius and the net section area decreased at high strain rates and low relative humidity (dried samples). The gradual localization of the deformation by the necking process was analysed by inspecting the changes in the microstructure. Tomography allowed the observations of voids appearing within spherulites in the form of two symmetrical fans, one at each end of the spherulites. A full study of these polar fans has been carried out in terms of morphology, general arrangement and characteristic length. The latter allowed axial and transverse deformations to be experimentally measured at the scale of the individual voids, the polar fans and the spherulites. The strain distributions were plotted with respect to the axial and radial directions. Heterogeneous strains were found: i) to be due to the necking for the two materials; ii) and due to the initial skin-core effects in the PP. These heterogeneities imply that the evolution of the volume variation of the specimen by measurement at the surface could not accurately reflect through-volume effects, as the assumptions of isotropy and homogeneity are not satisfied. Experimental data derived from uniaxial tensile tests should be enriched to build relevant constitutive models.

1. Introduction

Weight saving for engineering structures constitutes one of the motivations promoting the use of organic materials for the replacement of metals. This usually requires the redesign of industrial components in order to comply with their safety in use. Amongst the large range of structural polymers, thermoplastics have gained increasing interest thanks to their lower fabrication costs. Indeed, pre-designed shapes can be processed very close to their final complex profiles. Designing such complex shapes needs numerical computation generally based on finite element (FE) codes, relying on constitutive models based on the true stress-strain curve.

Traditionally, uniaxial tensile tests have been the principal means of obtaining the engineering stresses and strains of the materials under load. Since the true stress-strain curve is derived from engineering stress-strain plot using transverse properties (Section 2: **Theory**), the development of VideoTraction [1] and full field measurement combined with digital image correlation (DIC) improved knowledge of the mechanical behaviour of a given material. However, these strain 2D data still lack through-thickness information. To overcome this difficulty, it is generally admitted that:

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- the stress and the strain are homogeneously distributed within the gauge length of the tested specimen;
- by isotropy, the out of plane deformation is similar to that measured transversely.

This work aims at checking experimentally the validity of assumptions of isotropy and homogeneity, using an approach based on the relation between evolution of the microstructure during deformation and the mechanical properties. To this end, Section 3 details the specimens and the spherulitic microstructure of the two semi-crystalline polymers under study: an isotactic polypropylene (PP) and a polyamide 6 (PA6). These specimens were subjected to tensile tests with specific strain rates and preconditioning so as to provoke necking during the test. Experimental data during the necking process were collected, allowing the identification of the stage in the stress-strain curve where the minimum neck curvature radius was reached. This change in the shape of the specimen highlights, at the macroscopic scale, heterogeneity induced by the deformation. The last part of section 3 attempts to reveal the heterogeneity at the microscopic scale. To this end, necked specimens were produced so as to collect comprehensive through-thickness data in 3D from the deformed microstructures and, further, to depict their evolution. Unlike observation techniques such as SAXS, WAXS [2,3] and IPSLT [4] used to investigate such information on semi-crystalline polymers, synchrotron radiation computed tomography (SRCT) [5–7] has been used here. At a resolution of about 0.7 μm, the heights and the diameters of microstructural patterns such as individual voids and cluster of voids arranged into polar fans could be evaluated. From these SRCT data, the deviation from the homogeneity assumption could be analysed.

Section 4 sets three points of discussion: i) the outcome of the through thickness SRCT data in comparison with traditional measurements at the surface (load, full-field displacement); ii) the determination of the local strains that can be split into shear and volumetric parts; iii) the consequence of a better understanding of the link between microstructural evolution and macroscopic mechanical properties on the FE constitutive models.

2. Theory

Engineering stress-strain curves are obtained experimentally but constitutive modelling requires the true stress versus true strain curve to be correctly integrated. Let (r, θ, z) be the cylindrical coordinates where the load applied to the tensile specimen is in the z direction (Fig. 1).

Consider a representative volume element (RVE) where the stress and strain are supposed to be homogeneous. In Appendix A, it is shown that calculation of the true strain is straightforward, by integrating the increment of strain.

$$\varepsilon_z = ln\left[1 + \frac{\Delta l_z}{l_z^0}\right] \tag{1}$$

where l_z is the actual length l in the z direction and l_z^0 is the initial value of l_z.

The true stress is a function of the transverse strains ε_r and ε_θ (Appendix A):

$$\sigma_z = \frac{F}{S} = \frac{F}{S_0} e^{-(\varepsilon_r + \varepsilon_\theta)} \tag{2}$$

where F is the tensile load S_0 and S are the initial and actual section areas, respectively. Assuming that the material is both isotropic and isochoric (incompressible), the classical relationship between the engineering stress F/S_0 and the true stress can be derived from Eq. (2), so that:

$$\sigma_z = \frac{F}{S_0}\left(1 + \frac{\Delta l_z}{l_z^0}\right) \tag{3}$$

In the present work, the polymers under investigation were considered to be isotropic, at least initially, but not isochoric. Indeed, in contrast to metals in the plastic regime, polymeric materials are known to be pressure sensitive [1,9–13] due to volume changes. Therefore, the calculation of the true stress should be performed using Eq. (2). A partial answer can be given by full field strain measurement obtained by Digital Image Correlation (DIC). Indeed, ε_r and ε_θ were also measured *at the surface* during the deformation. However, there was no guarantee that these transverse deformations were homogeneous throughout the thickness, especially on samples with pre-existing skin-core effects.

Moreover, when the specimen underwent necking, the applied load together with the material response induce heterogeneity inside the necked zone. For notched round bars, the Bridgman formulae reported in Appendix A might be more relevant to describe the multiaxial stress state within the net section [8,14]:

$$\begin{aligned} \sigma_z(r) &= \sigma_{eq}\left[1 + ln\left(1 + \frac{a^2 - r^2}{2aR}\right)\right] \\ \sigma_r(r) &= \sigma_\theta(r) = \sigma_{eq}\left[ln\left(1 + \frac{a^2 - r^2}{2aR}\right)\right] \end{aligned} \tag{4}$$

where: a is the net section radius, R is the neck curvature radius and σ_{eq} is the Von Mises equivalent stress.

Note that the principal stresses in Eq. (4) depend on the geometrical parameters a and R. Measuring their current values during the stretching of the sample allowed estimation of the multiaxial true stress state within the net section of the material.

3. Materials and methods

3.1. Materials and specimens

3.1.1. Polypropylene and polyamide 6 semi-crystalline polymers

Two semi-crystalline polymers have been considered in this study: an isotactic polypropylene (PP) and a polyamide 6 (PA6) [6,17,18]. The glass transition temperatures T_g and the degrees of crystallinity of the two materials, evaluated using modulated differential scanning calorimetry are given in Table 1. The pronounced dependence of T_g on the water content was studied and summarized in Table 2.

3.1.2. Specimens

For PP, ISO flat dumbbell specimens were provided after injection moulding. The gauge length l_z^0 was equal to 25 mm (Fig. 1a).

Optical microscope observations on microtomed slices of the rectangular section in Fig. 2a showed increasing sperulite diameter from the surface (skin) towards the core of the sample. Some quasi-parabolic spherulites [15] were observed at the interface between the skin and the core, due to the thermal gradient during crystallization [16]. Fig. 2b illustrates the regular spherulites in the core. Their diameter is fairly homogeneous with an average value of about 60 μm. This change in both morphology and size of spherulites through the thickness is defined here as the skin-core effect.

For PA6, the 10 mm thick plate also exhibited skin-core effects [6]. However, machined round bars extracted from mid-thickness of this plate allowed homogeneous equi-axed spherulites to be obtained. The characteristic lengths of these round bars are given in