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Material behaviour

Comparison of voiding mechanisms in semi-crystalline polyamide 6 during tensile and creep tests

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ABSTRACT

Behaviour of a semi-crystalline polymer, polyamide 6, described by loading curves, as well as necking and whitening phenomena, is related to its micro-structural evolution in terms of void morphology and distribution during both tensile and creep tests. Notched bars have been subjected to creep tests interrupted at the onset of the tertiary creep stage and at the onset of final rupture. Inspections of these specimens using Synchrotron Radiation Tomography have been coupled with statistical image analysis treatment to obtain spatial distributions of void length and void volume fraction. Cavitation mechanisms observed and quantified during creep and tensile tests were similar: from penny shaped voids (diameter larger than height) perpendicular to the drawing direction to cylindrical voids (diameter equal to the height) arranged in columns during the neck extension. The void volume fraction distributions along radial and axial directions presented an inverted parabolic profile with a maximum located at the centre of the sample.

1. Introduction

The present paper addresses the question of the durability of polymeric engineering structures such as pressure vessels or pipes submitted to sustained internal pressure for tens of years (50 years). Such structures are subjected to creep loading under a multiaxial stress state. Creep tests at low stress levels require several years and cannot be easily programmed in a laboratory. Moreover, the methodology used to predict lifetimes is nowadays based on extrapolation hypotheses that may be questionable. Indeed, some premature failures of these structures are regularly observed [1]. The macroscopic and viscoelastic behaviours of semi-crystalline polymers subjected to creep loading and slow crack growth are widely studied in the literature [2–4]. However, very few studies about damage mechanisms and cavitation phenomenon within these materials deformed under constant load have been published [5]. In this paper, the methodology suggested to predict lifetimes of such structures is based on the evolution of the microstructure during creep loading. The technique used here to assess damage features at micrometre resolution was Synchrotron Radiation Tomography (SRT). This technique presents numerous advantages. First, regarding sample preparation, SRT is a non-destructive technique and SRT inspections do not require any preparation, unlike SEM inspections of semi-crystalline polymers [6]. Indeed, the latter must be preceded by cutting the samples (observations of microtomed or cryo-fractured surfaces), Au–Pd coating of these surfaces and sometimes by chemical etching. Moreover, voids can be visualized in 3D and directly in space with precise spatial location with the SRT technique whereas, other existing techniques such as IPSLT [7,8] or X-ray scattering [9] provide only an average value over a prescribed volume (no gradient). This ability to explore the whole volume of a sample also gives access to information through the thickness of the inspected samples and to spatial gradients of the parameters of interest. Such observations can now even be made with nanometre resolution, as shown for HDPE [10] and for PA6 [11].

To enhance void growth and to reproduce a well-controlled multiaxial stress state similar to the ones encountered in engineering structures, creep tests were carried out on notched round bars [12]. Macroscopic data such as creep curves or sample deformation were recorded. Whitening and necking on smooth or notched tensile specimens, which have been observed during

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several studies on semi-crystalline polymers, have been attributed to the presence of voids in the deformed polymeric material [12–14]. Previous works on the same PA6 material have related the onset of whitening with the yield stress during steady strain rate load and at the end of the secondary creep during steady loading [15,16]. Under these conditions, the neck extends in the drawing direction accompanied by void growth. In order to study the microstructural evolution, creep tests were stopped at characteristic stages during the creep loading, allowing 3D through thickness SRT-inspections. Qualitative results were first gathered and are discussed below: void shape evolution and their spatial distributions were studied. These observations have been related to quantitative results on the radial and axial evolution of void volume fraction and void characteristic lengths. The void shape was seen to change from prolate (flattened disks oriented perpendicularly to the drawing direction) to obloid (arrangement in columns of cylindrical voids the height of which increased, in the loading direction). These deformation micromechanisms have been already observed and studied during steady strain rate loading [16,17], and a comparison between these data and the present creep related data is made below.

2. Experimental

2.1. Material description

The material under study was a semi-crystalline polymer: a thermoplastic polyamide 6 (PA6) properties of which have been published elsewhere [18,19]. Modulated Differential Scanning Calorimetry (MDSC) was used to determine the main physico-chemical properties that are recalled here: the glass transition temperature $T_g = 53$ °C, the melting point $T_f = 219$ °C and the crystallinity index $\chi = 43\%$. The examination of specimens by Scanning Electron Microscopy (SEM) after chemical etching revealed the spherulitic microstructure of the material, characterised by a spherical shape and a mean diameter of about 5 μm for the spherulites. An attempt was made to inspect pre-existing voids on PA6 fracture surfaces obtained by cryofractography [15]. SEM images of these surfaces were analysed, leading to an initial void volume fraction estimated at about 1% and a void mean diameter around 0.1 μm.

2.2. Sample preparation

The notch root radius was R = 4 mm for the three samples studied (denoted samples A, B and C). The specimens had gauge section diameter of 7.2 mm and a gauge length of 65 mm. The net (minimal) section diameter was 3.5 mm for Samples A and C and 4 mm for Sample B (Table 1). This geometry leads to a moderate stress triaxiality ratio of about 0.54 in the centre of the notched region of the specimens. These samples were dried until a level of humidity of 0% in the material and stored afterward in a dessicator. Then, they were tested at room temperature (20 °C) and at a relative humidity of 50% using an electromechanical tensile rig, monitoring both axial load and cross-head displacement. The experimental procedure consisted of carrying out a tensile creep test up to failure of the specimen in order to plot the general trends of the creep strain history and to determine the time to creep failure of the PA6 material [18]. Similar to smooth specimen results, creep strain curves obtained on notched round bars exhibited three stages: the creep strain rate decreased rapidly with time (stage I, primary creep), then it reached a steady-state and minimal value (stage II, secondary creep), followed by a rapid increase and fracture (stage III, tertiary creep). The tests presented here were stopped around the second inflexion of the creep strain curve (end of the secondary creep) and during the tertiary creep. The purpose was then to study the microstructure evolution of the specimens by following the same approach as the one applied to monotonic tensile tests [17]. In this loading case, it can be recalled that necking and whitening appear for the same material and geometry at the yield (maximum) stress.

Table 1 summarizes characteristic data for the specimens investigated here. The engineering net stress (σ_{net}) is defined as F/S_0 where F is the load and S_0 is the net section of the notched round bar. The deformed notch opening displacement (δ) is defined as the distance between both notch shoulders. This quantity was measured on the deformed samples represented on Fig. 1a. The Creep Measured Displacement (CMD: Δu_c) is the difference between the applied displacement at the end of the test (u) and at the end of the loading stage (u_1):

$$u = u_1 + \Delta u_c$$

It has been mentioned above that the initial net section radius (R_0) was not the same for the three samples. They are shown in Table 1. The net section radii of the deformed samples (R) were also measured. The creep responses of the material together with the pictures of the deformed specimens are displayed in Fig. 1a. The height of the whitened and necked zone (h_w) was measured on these pictures. The values of δ and R are useful for the sake of normalization. This normalization procedure permits comparison of the results from the different samples.

Sample A had the minimum applied stress level ($\sigma_{net} \approx 71$ MPa). For Sample B, the applied stress was increased ($\sigma_{net} \approx 75.4$ MPa) to reduce the test duration and to study the influence of the stress level on the cavitation. Finally, Sample C was obtained after a long period of creep at a low stress level similar to that applied to Sample A ($\sigma_{net} \approx 71.5$ MPa). Since the applied stress was different for each sample in Fig. 1a, the plot does not reflect at what point from the end of the secondary creep the tests were stopped. First, the end of the secondary creep was identified by studying the evolution of the creep strain rate. Then, for each sample, CMD (Δu_c) and time were normalized by their value at the end of the secondary creep: Δu_{II} and t_{II}, respectively. In Fig. 1b, curves obtained after this normalization procedure are superimposed. It can be clearly observed that samples A and B have been deformed up to the beginning of the tertiary creep as soon as the necking accompanied by whitening appeared in the net section. The necked and whitened zone in the vicinity of the net section were, nevertheless, more extended in Sample B than in Sample A. Samples A and C were submitted to a similar net stress but Sample C has been deformed until a more advanced state of creep deformation, when the tertiary creep was well established, prior to the final rupture.

Table 1
Characteristic measures on deformed samples.

	σ_{net} (MPa)	Δu_c (mm)	δ (mm)	R (mm)	R_0 (mm)	h_w (mm)	h_w/δ(−)
Sample A	71	1.93	8.7	1.58	1.75	0.7	0.08
Sample B	75.4	2.06	8.1	1.81	2	1.5	0.19
Sample C	71.5	3.23	9.4	1.31	1.75	2.5	0.27