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Molecular chain orientation in polycarbonate during equal channel angular extrusion: Experiments and simulations

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ABSTRACT

The objective of this contribution is to provide some insights of the molecular chain orientation in polycarbonate (PC) during an equal channel angular extrusion (ECAE) process. Experimentally, wide-angle X-ray scattering was carried out to characterize the microstructure of the ECAE-deformed glassy amorphous polymer. The deformation behavior of the PC specimen in the ECAE process was simulated using a physically-based viscohyperelastic–viscoplastic constitutive model that was implemented into a finite element code. The large inelastic deformation response of the glassy amorphous polymer was modeled by assuming that the deformation mechanisms are split into an intermolecular resistance and a molecular network resistance. The intermolecular resistance captures the initial response and the yielding rate-dependence due to the isotropic resistance to deformation resulting from intermolecular barriers to chain-segment rotation. The molecular network resistance reproduces the anisotropic strain hardening response at large strains due to stretching and chain orientation. The progressive molecular chain orientation during the ECAE process and its distribution were analyzed. The predicted results, favorably compared with the experimental observations, show a high degree of anisotropy and heterogeneity in orientation of polymer molecular chains in the ECAE-deformed PC.

1. Introduction

The equal channel angular extrusion (ECAE) process is an ingenious deformation method that submits a specimen to a large plastic deformation in the solid state without changes in the specimen cross-section. Contrary to traditional solid-state forming techniques (such as forging, ram-extrusion and die-drawing) involving continuous specimen cross-section reduction, the ECAE method preserves the specimen cross-section which allows thus to repeat several times the process and to induce extremely large plastic strains in the specimen. The ECAE method, developed in the beginning of the 1980s by Segal et al. [1], consists in extruding a specimen through a die constituted of two equal cross-section channels. Fig. 1 shows schematically the ECAE process in the *L* configuration. The specimen being forced to pass, in the solid state, from the entrance channel to the exit one, it undergoes a large simple shear strain in the crossing plane of the two channels. In a very useful background paper, Beyerlein and Toth [2] have widely reported the considerable interest that the ECAE method has known in the last few years due to its impressive ability to control the microstructure and properties of crystalline metals. While the crystalline metals have received much attention (about 200 papers per year are published), the polymeric materials have not. The ECAE process was applied to semi-crystalline and glassy amorphous polymers [3–26]. Most of these investigations focused on the microstructure and properties of ECAE-deformed polymers, and particularly on the texture development in semi-crystalline polymers. Less works dealt with the molecular chain orientation in glassy amorphous polymers and its connection with the mechanical behavior, see e.g. [5,6,8] for polycarbonate and [13] for polymethylmethacrylate. Following the process parameters but also the structure and properties of the ECAE-deformed polymer, the stress state can change from the expected homogeneous simple shear to a complex inhomogeneous stress field. That results in an inhomogeneous molecular chain orientation in the extruded polymer. Over the last years, several numerical studies were carried out in order to examine the deformation behavior and the plastic strain heterogeneity in ECAE-deformed polymers [17–22], but they were never related to the microstructure evolution. In the present contribution, finite element simulations, using a physically-based constitutive model, were carried out in order to evaluate the degree and distribution

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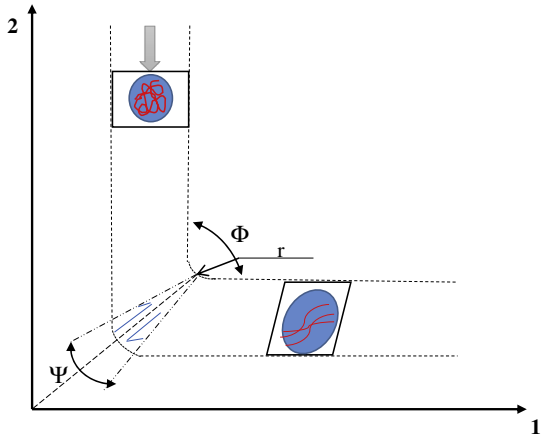

Fig. 1. Schematic illustration of the ECAE process.

of molecular chain anisotropy in polycarbonate when it is extruded by the ECAE process. In order to give an appreciation of the simulation results, experiments, giving an overview of the molecular chain anisotropy in the glassy amorphous polymer after extrusion, were performed.

The present contribution is organized as follows. In Section 2, we present the experiments concerning the mechanics of the ECAE process and the microstructural characterization. Section 3 is devoted to the simulations by describing the constitutive modeling and the computational formulation of the problem. The predicted results are discussed and verified by comparisons with the experimental observations. Finally, some concluding remarks are given in Section 4.

2. Experiments

In this section, experiments were performed to quantify the anisotropy and distribution of polymer molecular chains in ECAE-deformed polycarbonate.

2.1. Material and specimens

The polycarbonate (PC) used in this investigation was purchased from Goodfellow in the form of 10 mm thick compression-molded sheets. The 10 mm thick sheets were cut into 10×75 mm long billets for ECAE processing. Although material isotropy is expected, billets were cut from the as-received PC sheets along the same direction.

The PC intrinsic stress–strain response under finite strains was determined by means of a universal testing machine Instron-5800 connected to a non-contact optical extensometer able to maintain constant the local true axial strain rate during the test. Fig. 2a presents the PC flow behavior at different true strain rates and at room temperature. The highly nonlinear mechanical response of the PC material, which is the signature of most of glassy polymers, can be clearly observed. Under small deformation, the PC material exhibits a relatively stiff linear response, followed by a strain softening at yield. At moderate deformation, the PC material shows progressive stiffening followed by a dramatic strain hardening at large deformation. The strain rate affects the initial flow stress, but the nonlinearity appears to be strain rate independent.

2.2. ECAE experiments

ECAE experiments were conducted at room temperature and under a constant ram speed of 45 mm/min. The ECAE die is made of stainless steel and is characterized by a square channel cross-section of 10×10 mm, an internal angle Φ of 90° between the two channels, an outer corner angle Ψ of 10° and an inner radius r of 2 mm. It was developed to be easily set-up on a universal testing machine Instron-5800. The Instron testing machine has a capacity of 350 kN and an appropriate load-cell of 50 kN. The die was lubricated using silicone grease before extrusion. An example of load-ram displacement curve is presented in Fig. 2b. The extrusion load exhibits a strong increase at the beginning of the process, followed by a softening. At large displacement, a strong hardening phenomenon is observed. That may be correlated with the intrinsic strain hardening of the PC material and therefore to the molecular chain orientation.

2.3. Molecular chain orientation characterization

Wide-angle X-ray scattering (WAXS) experiments were carried out in order to quantify the orientation parameter of the macromolecular chains after ECAE processing. Experiments were carried out using a GeniX micro-source (Xenocs, France) operated at 50 kV and 1 mA. The Cu Kα radiation (λ = 1.54 Å) was selected and focused by means of FOX2D mirrors. Beam size was adjusted with two sets of scaterless slits. The 2D-WAXS patterns were recorded on a Photonic Science VHR CCD camera. Corrections were applied for background scattering, geometry and intensity distortions of the detector. Both incoming and transmitted intensities were measured synchronously with WAXS data acquisition. Thereby, the transmission factor, defined as the ratio between the transmitted

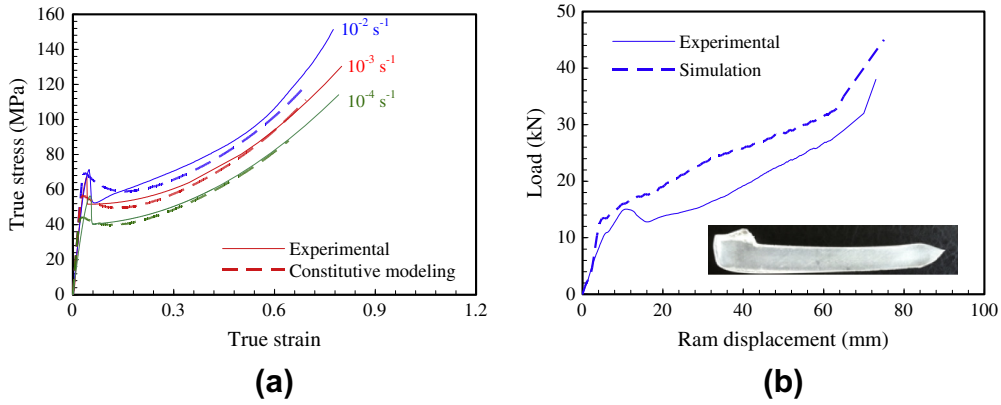

Fig. 2. Experimental and simulated (a) intrinsic stress–strain responses and (b) ECAE load-displacement curves.