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# Aging-dependent strain localization in amorphous glassy polymers: From necking to shear banding

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## ABSTRACT

This study investigates the influence of physical aging on the tensile deformation behaviors of poly(ethylene terephthalate)-glycol. The polymers are subjected to a quenched and an annealed heat treatment, followed by deformation in tension. The digital image correlation (DIC) is used to capture the surface strain. The DIC results show that the deformation localization occurs in the post-yield strain softening region. The quenched and annealed polymers exhibit different localization types, representing as necking for quenched polymers and shear banding for annealed polymers. With the help of the DIC, the nucleation and propagation of the localization zones are fully captured. The DIC results show that the reference location with the maximum strain does not change with the deformation. At the same strain level, the maximum strain in the shear bands is much larger than that in the necks, which explains the failure strain of the annealed polymers is smaller than the quenched polymers. The stretch rate also has a strong effect on the localization behaviors. A larger stretch rate results in a stronger localization due to an increase in the amount of strain softening.

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## 1. Introduction

Deformation localization, such as crazing (Legrand, 1969; Argon, 2011; Jiang et al., 2017), necking (Neale and Tugcu, 1985; Wu and van der Giessen, 1995; Li and Buckley, 2009) and shear banding (Brady and Yeh, 1971; Wu and Turner, 1973; Chau and Li, 1979), can result in an early failure of polymers, which significantly limits the applications of polymers as load-bearing materials. While crazing occurs in the pre-yield region, the necking and shear banding appear in the post-yield region. The post-yield strain softening in polymers is the intrinsic reason for the occurring of necking and shear banding (Govaert et al., 2000; Li and Buckley, 2010), which allows the coexistence of zones with different strain. The glassy polymers exhibit strain hardening at large deformation. Thus, the deformation localization zone propagates after initializing, which is different from the localization behaviors in metals (Wu and van der Giessen, 1994; 1995).

The nature of the deformation localization depends on temperature, loading rate, physical aging and mechanical deformation (Legrand, 1969; Dooling et al., 2002; Stoclet et al., 2014; van Melick et al., 2003; Govaert et al., 2000; Archer and Lesser, 2011; Dobovšek and Polonica, 2015). Jang et al. (1984) demonstrated that

ductile failure changed to crazing-induced brittle failure when decreasing temperatures and increasing strain rates for a variety of polymers. Dooling et al. (2002) showed that a transition from homogeneous deformation to necking in poly (methyl methacrylate) can occur when deformed above a critical strain rate or below a critical temperature. Temperature and strain rate also influence the morphology of shear bands. Coarse bands often appear at low temperatures and high rates, while fine bands can be observed at high temperatures and low rates (Dobovšek and Polonica, 2015). Physical aging can induce a more severe localization behavior, while mechanical pre-deformation can reverse the effects of physical aging. Yang et al. (1996) found that a homogeneous deformation was observed for the quenched poly (phenylene oxide), which changed to necking as aging and a brittle failure for the heavily aged condition. Deformation localization in some polymers can be eliminated by pre-deformation such as cold rolling (van Melick et al., 2003) and drawing (Unwin et al., 2002). Several excellent reviews are also available for localization behaviors in polymers. Tomita et al. (1998) provided a full discussion on the onset of necking, the propagation of necked region, the temperature change and the transition of necking to shear banding with pre-drawing. Meijer and Govaert (2005) reviewed the influence of the nonequilibrium structure on the localization behaviors of amorphous polymers.

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Based on the constitutive theories of glassy polymers (Boyce et al., 1992; Wu and Van der Giessen, 1996; Tomita et al., 1997; Tomita, 2000; Chowdhury et al., 2008; Li and Buckley, 2010; Xu et al., 2017; Kweon and Benzerga, 2013), the intrinsic strain softening plays a fundamental role on the deformation localization behaviors. Physical aging, also named as structural relaxation, determines the amount of strain softening (Xiao and Nguyen, 2015; Xiao et al., 2017). When approaching the glass transition temperature, the polymer structure falls out of the equilibrium. This nonequilibrium state continuously evolves towards an equilibrium state. This aging process can result in a more sluggish polymer structure, accompanied with a decrease in volume, enthalpy and an increase in viscosity, yield strength (Xiao et al., 2013; Liu et al., 2015). The mechanical deformation can induce an opposite effect on the polymer structure compared with physical aging, driving the polymer structure away from the equilibrium state, a process known as mechanical rejuvenation (Kierkels et al., 2008; Semkiv and Hütter, 2016). The amount of stress softening increases with aging and decreases with mechanical pre-deformation, which explains the effects of the physical aging and mechanical pre-deformation on the localization behaviors (Cross and Haward, 1978; Govaert et al., 2000).

The traditional experimental methods, such as strain gauge, can not capture the full field deformation behaviors. Also, those methods can only measure the small strain behaviors. The recent development of the non-contacted optical experimental techniques, such as digital image correlation (DIC), has provided a powerful tool to investigate the inhomogeneous deformation of various materials, such as biomaterials (Murienne et al., 2016), concrete (Lei et al., 2017), metals (Cai et al., 2016; Tung et al., 2010; Gao et al., 2015; Pan et al., 2015) and polymers (Grytten et al., 2009; Jerabek et al., 2010). Yu et al. (2016) applied DIC to study the necking of aluminum alloys. Zhu et al. (2012) used DIC to investigate the shear band nucleation and broadening in the nanocrystalline Ni sheet. Pan et al. (2016) applied high speed stereo-DIC to describe the deformation of an aluminum panel under impact. Compared with metals, the deformation of polymers typically involves with large deformation. Heinz and Wiggins (2010) applied the DIC to characterize the strain distribution during compression and found homogeneous deformation can only be observed in the pre-yield region. Ye et al. (2015) demonstrated that 3D DIC can fully capture the necking behaviors of high-density polyethylene up to a true strain 1.8. Poulain et al. (2013) and Engqvist et al. (2014) compared the performance of the DIC and other techniques to measure the localization behaviors of polymers at large deformation and found the DIC was more reliable. Pan et al. (2009) reviewed the methodologies of the DIC technique and the accuracy of strain estimation using DIC.

In this work, we will utilize the DIC to investigate the influence of physical aging on the strain localization behaviors of poly(ethylene terephthalate)-glycol (PETG). PETG is an amorphous copolymer of poly(ethylene terephthalate) (PET). However, it has not been as widely used as PET in industrial areas mainly due to the fact that PETG does not show stretch-induced crystallization as PET does. Recent works have shown that PETG has its own advantages, such as transparent, easy processing, high toughness and excellent mechanical properties (Kattan et al., 2001; Dupaix and Boyce, 2005). This investigation can further enrich our understanding of PETG and potentially promote the applications of PETG.

The paper is arranged as follows. The experimental procedures are shown in Section 2. Section 3 presents the deformation localization behaviors of the quenched and annealed polymers, followed by a discussion regarding the obtained results. The conclusion part summarizes the main findings and the future directions.

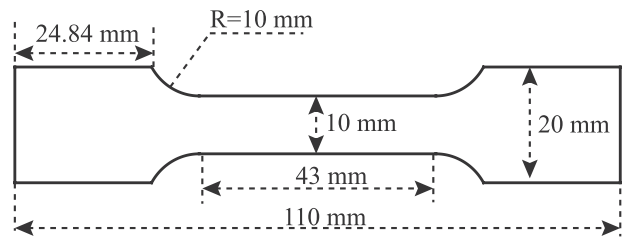


Fig. 1. The geometry of the specimens used in uniaxial tension tests.

## 2. Experimental methods

The PETG film with 1.59 mm (1/16 inch) thickness was obtained from McMaster-Carr Supply Company. The film was used in as-received conditions and was laser cut to the specific experimental geometry before testing.

### 2.1. Differential scanning calorimetry and dynamic temperature sweep tests

The differential scanning calorimetry (DSC) was used to characterize the phase transition behaviors of the obtained PETG, while the dynamic mechanical analysis (DMA) was used to describe the glass transition behaviors. Square specimen weighting 10 mg was cut from the PETG film and placed in a differential scanning calorimeter TA Q20 (TA Instruments, New Castle, DE). The specimen was heated from 25 °C to 110 °C at 5 °C/min.

To measure the dynamic modulus, film specimen with size 15 mm × 5 mm × 1.59 mm was used. The test was run on a dynamic mechanical analyzer TA Q800 (TA Instruments, New Castle, DE). The as-received specimen subject to a 0.2% dynamic strain in the tension mode was heated from 20 °C to 130 °C at 2 °C/min. The frequency of the dynamic test was chosen as 1 Hz.

### 2.2. Uniaxial tension tests

The geometry of tensile specimens is shown in Fig. 1. Before the thermal treatment, the specimens were sprayed with random speckles. To remove the effects of previous thermal history, the specimens were first equilibrated at 110 °C for 30 min in a thermal chamber. The quenched specimens were obtained through transferring the specimens from the thermal chamber to water at room temperature. For the annealed condition, the temperature of the thermal chamber was decreased step-wise by 10 °C each time and held for 30 min until room temperature. The total process took 4 to 5 h. The size of the specimen showed no change after thermal treatment, which indicated no molecular orientation existed in as-received materials. The uniaxial tension tests were performed on the quenched and annealed specimens using Electronic Universal Material Testing Machine Instron 3360 Series Testing Machine at room temperature (20–24 °C) with a 30 kN load cell. Three loading rates were chosen: 0.774 mm/min, 7.74 mm/min and 77.4 mm/min. During the tension tests, a high precision CCD camera with the M0814-MP's megapixel lens of Japan Computer was used to record the surface of the specimens. The resolution of each image was 1624 × 1224 pixels<sup>2</sup>. In our setup, one pixel represents 0.04 mm. The strain resolution of this DIC system is 0.01%.

## 3. Results

The DSC result in Fig. 2-a shows that the PETG exhibits a glass transition behavior. No endothermic peak was observed during the heating process. This indicates that no melting transition has been observed until 110°C. This is consistent with the observations in Kattan et al. (2001). PETG tends to resist crystallization,

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