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On the Viscosity Maximum during Uniaxial Extension: Material Property or Measuring Artifact?

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Abstract. An experimental investigation of the viscosity maximum observed during uniaxial extension of a low density polyethylene is presented. For this purpose, traditional integral viscosity measurements on a Münstedt-type extensional rheometer are combined with local measurements based on the in-situ visualization of the sample under extension. For elongational experiments at intermediate constant strain rates local maximum of the extensional viscosity is systematically observed. Images of samples within this regime show that secondary necks develop along the sample at the moment when the maximum of the integral viscosity is observed. The emergence of a maximum of the integral elongational viscosity is, thus, related to the distinct non-uniformity of deformation states and most probably not to the rheological properties of the material. Moreover, the position of the viscosity maximum was found to be dependent on the initial geometry of the specimen. This indicates again that the appearance of the transient viscosity maximum is not a material property but an artifact caused by a geometrical non-uniformity of highly stretched specimens.

Keywords: Polymer melts, Rheology, Elongational flow.

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INTRODUCTION

Uniaxial extension is the dominant flow in many industrial processes and, therefore, accurate measurements of the extensional rheological properties of polymer melts are very important. From a more fundamental point of view, reliable measurements of the rheological properties in extension are crucial for validating existing theoretical models and suggesting new approaches. In spite of the universally recognized need for reliable elongational measurements of polymer melts, the development of extensional rheometric equipment has progressed slowly during the past three decades. Versatile design of an extensional rheometer has met several practical difficulties and one of the toughest is to generate a homogeneous extensional flow. Several techniques to measure the elongational properties of polymer melts have been proposed: the Rheometrics Melt Extensiometer (RME) by Meissner [1], the supporting oil-bath design by Münstedt [2], the Sentmanat extensional rheometer (SER) [3] and the Filament Stretching Rheometer (FSR) [4,5]. It is worth noting that for each of these approaches the homogeneity of deformation states is crucial for

reliably assessing the elongational properties of the material. Though previously recognized by most experimentalists, this issue did not receive the proper attention. Only very recently the negative influence of sample non-uniformity during elongation has been made explicit, by measuring locally both the stresses and the strain and showing that in the case of strongly non-uniform samples the classical extensional measurements become completely unreliable [6]. Moreover, it has been demonstrated that even initially homogeneous (perfectly cylindrical) samples lose their uniformity at high enough Hencky strains and thus, the impact of sample non-uniformity on the viscosity measurements is always an issue to worry about during extensional tests. Also very recently, a full numerical simulation of the extension process in a SER rheometer demonstrated that the loss of sample homogeneity during deformation leads to a strong strain localization along the sample which ultimately translates into unreliable measurements of the transient elongational viscosity [7].

Since the early days of extensional rheology it has been observed that some strain hardening materials under uniaxial extension display a clear maximum in the transient extensional viscosity shortly before the physical rupture of the sample [8-10]. Moreover, Rassmusen et al. [11] observed even an overshoot behavior, i.e. an extended plateau (over several Hencky strain units prior to the physical rupture of the sample) after the viscosity maximum. During the experiments presented in [11] the geometric uniformity of the sample has been locally monitored by a laser which images a small region of the sample around its centre, measures in situ the mid-point diameter of the sample and passes this information to a feedback loop which maintains a constant rate of deformation at the mid-point of the sample. However, no experimental evidence (e.g. video imaging of the entire sample under deformation) for the homogeneity of the sample during the elongation process is presented.

The existence of a true viscosity overshoot is important from a theoretical point of view, too, as reliable elongational data are needed for a verification of molecular theories. The POM-POM model for branched polymer melts [12] and the molecular stress function (MSF) model [13] predict a monotone increase of the transient extensional viscosity to a plateau. Other theoretical works, however, are able to predict an overshoot in viscosity [14,15]. Even more worrying, recent theoretical models seem to be able to fit both a maximum and/or a steady state of the transient extensional viscosity [15].

In this paper the origin of the viscosity maximum in uniaxial extension of low-density polyethylene is investigated by means of in-situ optical measurements which allow one to evaluate the homogeneity of deformation precisely.

EXPERIMENTAL

The material used in this study is a low-density polyethylene from Lyondell Basell with the trade name Lupolen 1840 D. A detailed description of the sample preparation for elongational experiments can be found elsewhere [16].

The experiments at constant strain rate at 140°C have been conducted with a Mündstedt-type extensional rheometer which is illustrated in Figure 1a. The specimen **S** under investigation is clamped between the plates **P1** and **P2** of the rheometer and immersed in a silicone-oil bath **C** to minimize gravity and buoyancy effects. While the

bottom plate **P2** is stationary, the top plate **P1** is moved vertically by an AC-servo motor **M**, controlled by the computer **PC1**. The sample is illuminated from behind by two linear light sources **LS1** and **LS2**, disposed as shown in the schematic top view presented in Figure 1b. A major difficulty in imaging a considerably elongated sample comes from the high aspect ratio of the corresponding field of view, which during extensional experiments at large Hencky strains may be as large as 50:1. Therefore, a high resolution telecentric lens with the entrance pupil located at infinity, (VisionMes 225/11/0.1, Carl Zeiss) was used in this study which images the sample in parallel light and delivers frames with very uniform brightness and free of distortions, perspective errors and edge position uncertainties. Images of the sample under elongation are acquired in real time using a high resolution (3000 by 1400 pixels full frame, which translates into roughly 60 μm spatial resolution) low noise camera (Pixelink from Edmunds Optics) at a speed of 3 frames per second. The video camera is connected to a second computer, **PC2**. The image acquisition is digitally synchronized with the rheometer via a transistor-transistor logic (**TTL**) trigger signal sent by the rheometer drive **D** to the camera.

FIGURE 1. (a) Schematic view of the experimental apparatus: C - oil bath, P1 and P2 - top and bottom plates of the rheometer, S - the sample under investigation, M - AC servo motor, D - the control drive of the rheometer, PC1,2 - personal computers, TL - telecentric lens, CCD - video camera.
(b) Sample illumination and imaging (top view): LS1 and LS2 - linear light sources.

RESULTS AND DISCUSSION

Integral measurements of the transient elongational viscosity μ^+ at a constant rate of deformation $\dot{\epsilon}$ are presented in Figure 2. The integral viscosity is obtained by measuring the transient tensile force, using the assumption that the cross-section of the sample decreases exponentially with time and it is constant along the sample at any state of deformation. Each data set has been acquired until the physical rupture of the sample occurred. Except for the linear range of deformation, $\epsilon_H < 1$, the shape of the transient elongational viscosity curve depends considerably on the constant rate at which the material is deformed. Thus, depending on the rate of deformation, the integral transient viscosity may display either a clear maximum (curves 2–4, Figure 2) or a monotonic increase (curve 5, Figure 2).

FIGURE 2. Transient elongational viscosities at various rates of deformation $\dot{\epsilon}$: (1) 0.002 s^{-1} ; (2) 0.015 s^{-1} ; (3) 0.02 s^{-1} ; (4) 0.025 s^{-1} ; (5) 0.09 s^{-1} . Each data set has been acquired until the physical rupture of the sample occurred. The measurements were performed at $T = 140^\circ\text{C}$.

To get a more complete picture of how the shape of the transient viscosity curve is influenced by the deformation conditions and to identify the deformation regime in which a stress maximum is observed, measurements similar to those presented in Figure 2 were performed in a wide range of strain rates covering nearly three decades. The results of these measurements are summarized in Figure 3 which presents the Hencky strains corresponding to a maximum in the tensile stress and to the physical rupture of the sample as a function of the elongational rate. The Hencky strains presented in Figure 3 have been identified using integral measurements of the transient tensile force stress.

As can be seen from Figure 3, one can distinguish three different deformation regimes. At very low elongational rates ($\dot{\epsilon} \leq 0.01 \text{ s}^{-1}$) the Hencky strains corresponding to a stress maximum and to the physical rupture of the sample are practically independent of the strain rate and they are separated by roughly 0.8 Hencky strain units. As the rate of deformation increases a second deformation regime is observed in the range from 0.01 to 0.1 s^{-1} . The Hencky strains corresponding to a stress maximum and to the physical rupture of the sample depend significantly on the strain rate applied and they get progressively closer to each other as the strain rate increases. This finding suggests that the emergence of a stress maximum and the physical rupture of the sample are interconnected phenomena. Within this deformation regime, the integral elongational viscosity displays a local maximum which becomes sharper as the rate of deformation increases. Ultimately, at high elongational rates, $\dot{\epsilon} > 0.1 \text{ s}^{-1}$, a third deformation regime is attained. Within this regime a local maximum of the

tensile stress is no longer observed (cf. curve 5 in Figure 2) and the Hencky strain corresponding to the physical rupture of the sample becomes practically independent of the strain rate. A more detailed characterization of the deformation regimes can be found elsewhere [16].

FIGURE 3. Dependence of the Hencky strain corresponding to the stress maximum and physical rupture of the sample on the strain rate applied.

The following discussion is focused on the phenomenon of the viscosity maximum observed in the second deformation regime, i.e. at moderate elongational rates. In order to reveal the origin of this effect, in-situ optical investigations were performed and the homogeneity of deformation was evaluated.

In a range of high Hencky strains ($\epsilon_H > 3.5$), prior to the physical rupture of the sample, necks develop in the proximity of the midpoint of the sample as visible in Figure 4. The exact location of these necks is not reproducible in subsequent experiments. With increasing rate of deformation the geometric inhomogeneity of the sample becomes even more pronounced. Just after a local maximum in the viscosity is observed ($\epsilon_H \approx 3.5$), a secondary neck emerges slightly below the center point of the sample (cf. curve 2 in Figure 2 and Figure 4a). It can be seen that as the Hencky strain increases, the secondary neck becomes sharper (its local diameter decreases rapidly) and moves slowly along the sample. Another localized neck is formed at $\epsilon_H \approx 3.59$ and this ultimately leads to the physical rupture of the sample in a finite time.

FIGURE 4. Sequence of specimen images under deformation at the strain rate of 0.015 s^{-1} ($T=140^\circ\text{C}$). The aspect ratio of each image has been modified in order to enhance the clarity. The dotted squares indicate the location of the necks. The corresponding Hencky strains ε_H are as follows: a) 3.55; b) 3.56; c) 3.58; d) 3.59; e) 3.60; f) 3.62; g) 3.63.

These experimental findings suggest that the maximum of the transient extensional viscosity of some polymer melts does not reflect true rheological features of the material and is solely related to a severe inhomogeneity of deformation states due to the emergence of secondary necks along the sample. Another hint supporting this conclusion was obtained from the measurements on samples with different initial geometries. If the viscosity maximum is a material property then it should be independent of geometric aspects. However, the results in Figure 5 clearly show that the shape of the transient elongational viscosity curves depends strongly and systematically on the initial aspect ratio of the sample L_0/D_0 . For the highest initial aspect ratio ($L_0/D_0 = 3.03$) a monotone increase of the elongational viscosity is observed. In this case, the maximum travel distance of the top plate of the rheometer is insufficient to reach high enough Hencky strains at which the viscosity maximum could be observed. As the initial aspect ratio of the sample decreases, a viscosity maximum emerges. With decreasing L_0/D_0 of the sample the location of the viscosity maximum changes systematically: the maximum occurs at smaller total elongations and its value decreases considerably.

FIGURE 5. Measurements of the transient elongational viscosity for LDPE samples with different initial aspect ratios L_0/D_0 ($T=140^\circ\text{C}$, $\dot{\epsilon} = 0.07 \text{ s}^{-1}$).

CONCLUSIONS

A systematic experimental investigation of the role of sample homogeneity during measurements of the transient elongational viscosity on a polymer melt exhibiting strain hardening was performed. Using the in-situ optical method introduced by Burghelea et al. [16] it was shown that the emergence of the viscosity maximum is associated with a strong non-uniformity of sample in the form of secondary necks. Moreover, the local maximum of the transient elongational viscosity depends systematically on the initial aspect ratio of the sample. In view of these findings, it can be concluded that a viscosity maximum emerges as an experimental artifact rather than an intrinsic property of the material under uniaxial extension.

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