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ORIGINAL CONTRIBUTION

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Evaluation by means of stress relaxation (after a step strain) experiments of the viscoelastic behavior of polymer melts in uniaxial extension

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Abstract As is widely acknowledged, morphology in most materials is far more sensitive to extensional than to shear deformations but, unfortunately, due to the experimental difficulties involved, there are no non-destructive, morphology probing techniques in such flows, i.e., the equivalent of stress relaxation and oscillatory experiments in shear flows. This paper tries to overcome some of those drawbacks by proposing an experimental technique that allows stress relaxation experiments after a step strain in uniaxial extension to be performed. The benefits of this technique are twofold: (a) while the deformation is small enough for the response to be in the linear viscoelastic regime it constitutes a probe of the microstructure of the material and (b) it allows the departure to the non-linear regime to be studied, useful, for example, for the definition of the damping function in uniaxial extensional flow or for the study of the response of materials to fast transient flows with a strong extensional component, such as contraction flows.

In this work the proposed technique, which requires a correction to the apparent (theoretical) strain rate in order to allow the calculation of the true Hencky strains attained during the strain step, is tested and validated for two polyisobutylene melts.

Key words Uniaxial extension · Stress relaxation after step deformation · Modified rotational rheometer

Introduction

Extensional deformation regimes are among the most influential modes of deformation found in common processing situations. Thermoforming, fiber-spinning, and extrusion blow molding are only a few examples of transformation processes where such deformation modes are found to be predominant. It is known that extensional flows are responsible for a higher degree of orientation of the polymeric chains than shear flows, which leads to an improvement of the mechanical properties. Therefore, knowledge of the response of polymeric systems to extensional deformation regimes is very important in order to understand the physical mechanisms behind the technological processes used in the polymer processing industry.

The response to shearing flows is reasonably well understood, not least due to the large number of devices that became available for research purposes. Pure extensional flows, however, aren't easy to achieve, the attainment and maintenance of a pure extensional flow until high enough values of strain are achieved having been found more difficult to obtain than originally anticipated. Among the many difficulties found, obtaining a true extensional flow, fixing and supporting the sample in order to overcome gravitational and end effects are some of the most influential ones (see, for example, Rides et al. 1996a, Schweizer 2000, and Barroso et al. 2001).

A large number of different devices, with several distinct operating principles, were constructed and are described in the literature. Pioneering devices were designed by Cogswell (1968) and Meissner (1969), while

other researchers like Macosko and Lorntson (1973), Vinogradov et al. (1970a, 1970b, 1972), Everage and Ballman (1976), Munstedt (1975, 1979), Laun and Münstedt (1976, 1978), and Maia et al. (1999) have performed modifications to the original devices with the aim of obtaining relatively simple and cheap equipment by making use of the driving and/or the measuring systems of the rheometers. These pieces of equipment (in their various evolutions over the years) have allowed "approximate" uniaxial extension experiments to be performed in several deformation regimes: constant stress, constant strain rate, constant force, and even constant speed. Regarding polymer melts, rotating clamp – as well as translating clamp – rheometers, both working in controlled strain rate and/or controlled stress, have been the preferred means of measuring the response of the materials to uniaxial solicitations. The main goal of these experiments has been the study of the stress and strain growth with the imposition of, respectively, a constant rate of deformation or a constant stress regime, researchers striving to achieve the highest possible rates of deformation and strains whilst assuring that the deformation will be as homogeneous as possible.

Despite this recent appearance of a number of devices that are capable of yielding reasonably accurate and reliable data, some important experimental capabilities are still missing today, namely ones that may allow the study of one or both of the following: (a) the linear viscoelastic response of the materials, i.e., a non-destructive, extensional test of microstructure, since it is well known that the material response is more sensitive to extension than to shear and (b) the conditions that leads to the departure from the non-linear regime, important, for example, for the study of the response of a particular material to fast transient flows with a strong extensional component, such as contraction flows.

Regarding the study of the viscoelastic behavior, there have been a number of reports that deal with the subject. For example, Meissner (1972) reports that his universal extensional rheometer is able to impose any set strain or stress history to the sample. It is thus capable of performing stress relaxation experiments after cessation of steady extensional flow. However, it is not clear from the paper if this is also valid for step deformations and very recently Schweizer (2000) states that even the most updated versions of this instrument (in its commercial form) cannot perform such experiments.

Ide and White (1978) have also performed stress relaxation experiments using a similar idea to the fiber-windup technique and tested a number of polymers. The data concerning these experiments isn't shown, but is used to evaluate theoretical parameters of viscoelastic constitutive equations. However, the authors do point out the fact that in these experiments finite strains must be used. Munstedt (1979) reports that his universal extensional rheometer is able to perform stress relaxation experiments but, like Meissner (1972), no experimental data is presented. Munstedt's apparatus, working under the closed loop control principle, is also able to perform creep and recoverable strain measurements. More recently, Rides et al. (1996b) have performed a modification to a rotational rheometer in order to construct a uniaxial extensional rheometer capable of measuring the relaxation stress of polymer melts. Their technique is similar to the fiber-windup technique of Macosko and co-workers (Macosko and Lorntson (1973; Padmanabhan et al. 1996), one of the plates of the rotational rheometer being replaced by a drum, which acts as a winding device. Stress relaxation measurements are described for two high-density polyethylenes and an extension thickening behavior is found to occur, with an increase of approximately 60% in the relaxation modulus with an almost fourfold increase in the initial strain. Again, however, the minimum duration of the step is around 0.8 s and, therefore, cannot be considered a true step deformation, especially for small strains.

Regarding polymer solutions, there has also been a strong (and very recent) interest in the viscoelastic behavior of these materials, as is testified by the large number of reports such as those of Orr and Sridhar (1996), Spiegelberg and McKinley (1996), van Nieuwkoop and Czernicki (1996), and Remmelgas et al. (1998), to name but some of the pioneering works in this particular subject. However, since all these experiments are carried out for stress relaxation of polymer solutions after finite strains and resorting to translating clamp devices, usually based on the design of Tirtaatmadja and Sridhar (1993), they fall outside the scope of the present work and will not be further addressed.

In addition to the study of the fluid behavior in fast transient extensional or extension-dominated flows, the study of the transition from the linear to non-linear viscoelastic behavior is also important from a fundamental point of view, namely in what regards some of the assumptions taken when integral-type models, such as the simplified K-BKZ-type models (e.g., the Wagner model) are developed. In fact, the general form K-BKZ model is not tractable from the experimental viewpoint, since it involves the derivatives of a potential function that depends on both time and strain (which means that the amount of experimental data required to fully characterize a true K-BKZ fluid is so high that it can safely be considered impossible to obtain).

The most common and widely accepted way of simplifying the general model is through the assumption of the so-called time-strain separability principle, whereby the time- and strain-dependent responses of the stress are assumed to be independent of each other, thus allowing the definition of a memory function that contains the information regarding the linear viscoelas-

tic behavior, i.e., the time-dependency, and a damping function that contains information about the non-linear strain dependency. However, due to the complexity of the model, the simplified version is derived for the particular case of shear flows, assuming time-strain separability to be valid and is later re-extended, via the appropriate generalized strain measures, to other flows.

The above, coupled with the experimental difficulties in performing reliable extensional rheometry already referred above, has meant that all the (vast) work reported in the literature has been carried out on shear flows and, in accordance with the hypothesis of the simplified model, the validity of time-strain separability implicitly has been assumed for other flows, e.g., extensional flow. However, no direct evidence on the validity of the principle from stress relaxation experiments *after a step strain* in extension has ever been reported, thus constituting an open problem.

This paper tries to overcome some of the drawbacks mentioned above by proposing an experimental technique that may allow stress relaxation experiments – after a step strain in uniaxial extension – to be performed.

Experimental

Experimental set-up and conditions

Following on the ideas to modify commercial rheometers, a uniaxial extensional device was built for the measurement of the tensile behavior of polymer melts. The equipment is an add-on fixture to a TA Instruments Weissenberg Rheogoniometer that, besides the measurement of stress growth upon the attainment of a constant strain rate, also allows the stress relaxation after a step strain to be measured. The apparatus, of which a schematic diagram is represented in Fig. 1, has been described elsewhere (Maia et al. 1999) and its concept has been already validated (Maia et al. 1999; Barroso et al. 2001); it will be named MRR (Modified Rotational Rheometer).

Stress relaxation tests in shear and in extension were performed at 25 °C. The materials used in these experiments were two grades of polyisobutylene, PIB (of 0.93 g.cm^{-3} density), Oppanol B12 and B15, with average molecular weights of 60,000 and 85,000 and polydispersity indexes of 1.9 and 2.1, respectively, supplied by BASF Portugal. The stress relaxation tests in shear were performed on the Weissenberg Rheogoniometer using a cone-and-plate geometry, with a diameter of 4 cm and a cone angle of 2°. Small discs of PIB (4 cm diameter) were prepared, time being allowed for them to adjust to the desired gap between the cone and the plate (due to the high viscosity of PIB).

The stress relaxation experiments in extension were also performed using the stress relaxation package of the Weissenberg, which allows setting different initial step strains. The MRR allows a maximum theoretical step Hencky strain of approximately 0.3 units, i.e., a linear deformation of approximately 35%, to be obtained, over a period of less than ten hundredths of a second. The angular displacement of the rollers is measured for each step strain, in order to determine the apparent Hencky strains for any given experiment (that, as will be shown later, are not the real Hencky strains). The samples used in the extensional experiments were made by slowly extruding ($Q \sim 3.5 \times 10^{-2}$ mm^{3}s) the material through a 2 mm diameter orifice by means of a piston, which is then driven by a weight. This allows one to obtain cylindrically shaped samples with a constant diameter (between 3 and 3.5 mm per sample) and good surface finish. Samples showing entrapped air bubbles and surface defects were rejected. The sagging of the samples was removed by means of a slight stretching, at a very low rotation speed. Only after a period long enough for the measured torque to return back to zero was the experiment begun. Image acquisition was performed with a digital video camera, and was analyzed using a MiroVideo acquisition board and the image analysis software package Image Tool.

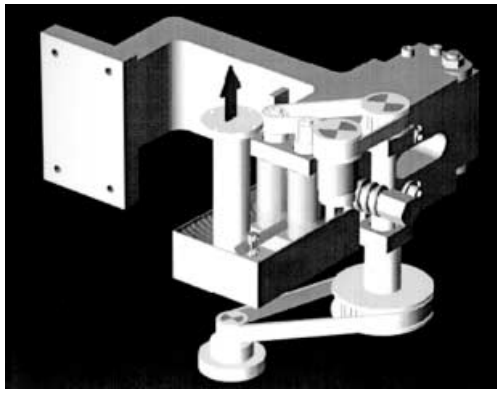

Fig. 1 The MRR

Procedure

When performing stress relaxation *after a step deformation* experiments in uniaxial extension, there are two very important possible sources of error, in addition to the usual ones commonly found in extensional rheometry of polymer melts in general, that need to be taken into account: motion transmission between the rollers and the sample and transient equipment effects.

The first of these is related to whether the angular displacement of the rollers is fully transferred to the samples or not. In fact, previous experiments performed at constant strain rate by several authors (of which Schweizer 2000 and Barroso et al. 2001 are only the most recent) showed that a correction to the apparent strain rate needs to be introduced in order to obtain the true strain rate. In the present case, this correction was performed by determining the true strain rates attained during the experiments via the radius evolution of the samples. This has enabled a power-law type dependence of the true strain rate with the apparent one, with an index of 0.93 for the case of PIB, to be established, as can be seen in Fig. 2. Therefore, although during the stress relaxation experiments in extension the angular displacement of the rollers is measured, it is to be expected that the corresponding linear displacement of the surface of the rollers is not fully converted into linear displacement of the sample. As such, a calibration procedure was established in order to estimate the true displacement of the sample and the corresponding extensional deformation that is represented schematically in Fig. 3.

The first step in the calibrating procedure consists in performing a first series of tests in which the time during which the roller on rotation was measured by means of a high-speed Super VHS video camera, working at 500 frames per second, for different set strain steps. This enabled a relation between the value for the set strain step (which is introduced in the software) and the actual total time that the rollers take to perform a given strain step to be established. The average (apparent) strain rates applied during the strain step are then obtained by dividing the apparent Hencky strains

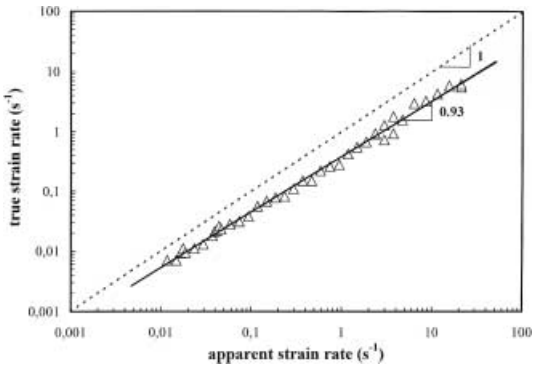

Fig. 2 Dependence of the true strain rate on the apparent strain rate

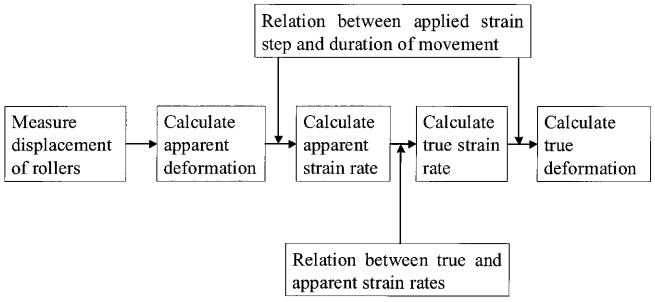

Fig. 3 Algorithm for the determination of true strain

determined from the displacement of the rollers by the estimated time for the duration of the strain step. True strain rates can then be determined via the relationship shown in Fig. 2. The effective Hencky strains were then determined by multiplying the corrected strain rates by the time estimated for the duration of the strain step. The effective Hencky strains are plotted in Fig. 4 and it is clearly seen that a large deviation occurs between these quantities.

The second and most crucial point that needs addressing is related to the shape and magnitude of the measured signal during the experiments. Since the transition to the strained state and back to rest is very fast (of the order of a few hundredths of a second) and because of the particular form of the torque measuring head of the rheogoniometer (in which an elastic torsion bar of known elastic constant is attached to the fixed clamp via a frictionless air bearing) it is expected that transients will appear in the output signal. In fact, after an initial overshoot, damped oscillations are observed in the torque measuring head. Figure 5 shows two examples of this behavior for both materials. The reason for this damping comes from the superposition of the signal originating from the self-damping of the elastic torsion bar of the measuring head to that of the true signal. However, good reproducibility was still achieved as is shown in Fig. 6 where data obtained for two repeats for each set strain step can be observed. Although the rheogoniometer has a built-in feature that allows the dampened oscillation of the bar to be smoothed out, its consequences to the true relaxation data are very serious, as is shown in the Appendix, and it was not used.

Bearing in mind the above results, the strategy adopted was to construct upper and lower boundaries to the measured output signal (other alternatives proved to be very difficult since the data acquisition rate varied along the experiment – a built-in feature of the software of the rheogoniometer that could not be overcome). The functions of choice for the boundaries vary somewhat, since the only criterion is for the evolution of the boundary points to be preserved, one of the most common choices being of the type

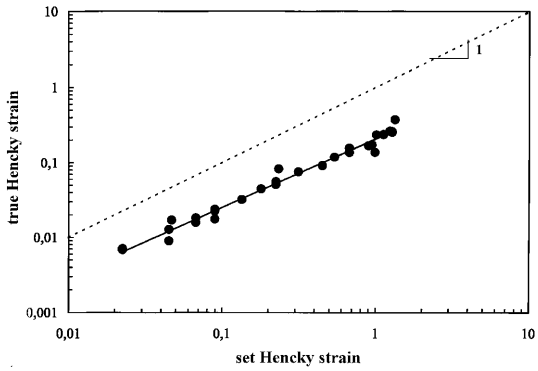

Fig. 4 Correction for the Hencky strain attained during the strain step in extension

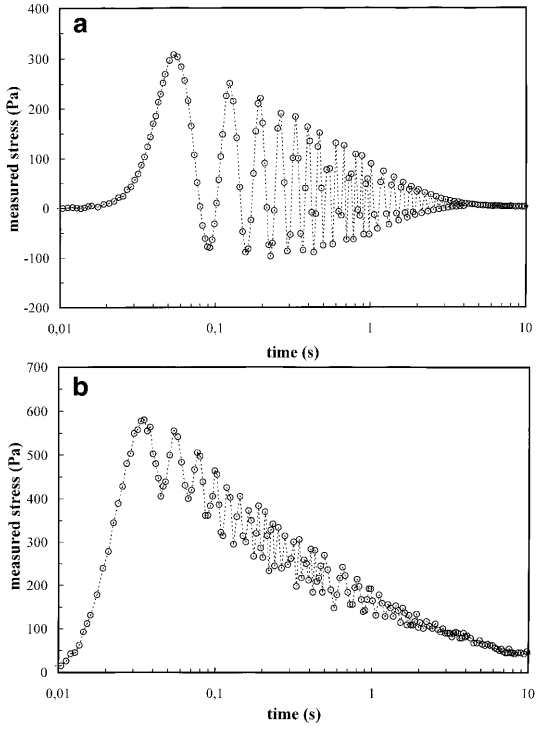

Fig. 5a, b Tensile stress relaxation experiments performed with different torsion bars, without any filtering: **a** run for PIB B12 using torsion bar no. 7; **b** run for PIB B15 using torsion bar no. 8

$$y = \frac{(a + cx + ex^2 + gx^3 + ix^4 + kx^5)}{(1 + bx + dx^2 + fx^3 + hx^4 + jx^5)}$$

The true relaxation signal is then assumed to be the average of two boundaries. Figure 7 shows the result of the application of this procedure to the experiments of Fig. 5.

Results

The behavior observed for the relaxation modulus in shear is the conventional one, time-strain separability

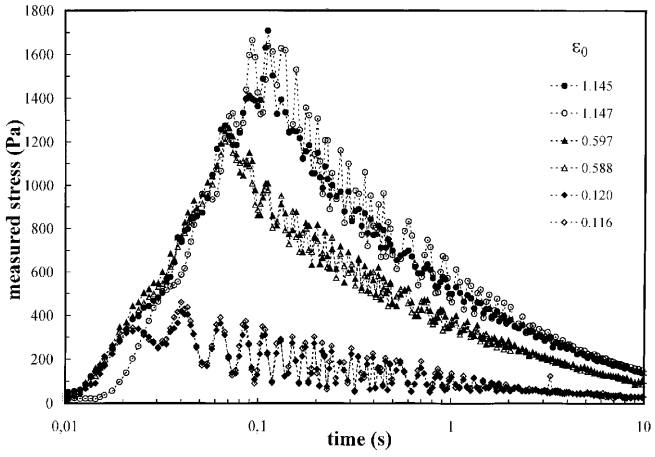

Fig. 6 Reproducibility of stress relaxation experiments

being verified and departure from linear viscoelastic regime occurring at a strain of around 0.4 for PIB B12 and 0.85 for B15 (see Fig. 8, for the example of B15), the maximum deformation being 1.0 and 1.8, respectively. The damping function was then calculated and the results can be seen in Fig. 9 for both B12 and B15, where the characteristic "softening" for increasing strains can be observed. The results indicate that PIB B12 "softens" more rapidly (with increasing strains) than B15 as would be expected on the basis of its similar polydispersity and lower molecular weight.

Figure 10 shows the results obtained for the relaxation modulus in extension for both materials, for various extensional deformation steps. In this, the deformation is the generalized strain measure, $\varepsilon = e^{2\varepsilon_H} - e^{-\varepsilon_H}$, where ε_H is the effective Hencky strain, the generalized deformation ranging from 0.1 to 1.1 (the latter value corresponds to a Hencky strain of 0.3). The most obvious feature is that there is an overlap of the data, which seems to indicate that the range of tensile strains imposed were still within the linear regime for either of these materials. This can be confirmed when the damping function is plotted for both melts (Fig. 11), it being obvious that, within experimental error, no softening is yet occurring. These results are in line with those from steady extension (see Fig. 12 for the example of PIB B15), which indicates a departure from the linear viscoelastic regime at Hencky strains of approximately 0.4 or, equivalently, 50% in linear deformation. Since the maximum generalized strain of 1.1 corresponds to a linear extension of just over 30%, it would then be expected that, for the particular case of these melts, all the experiments were performed within the limits of linear viscoelasticity. Unfortunately, it was not possible to perform experiments at higher strains in order to capture the onset of non-linear behavior, since the maximum angular displacement of the instrument had been achieved.

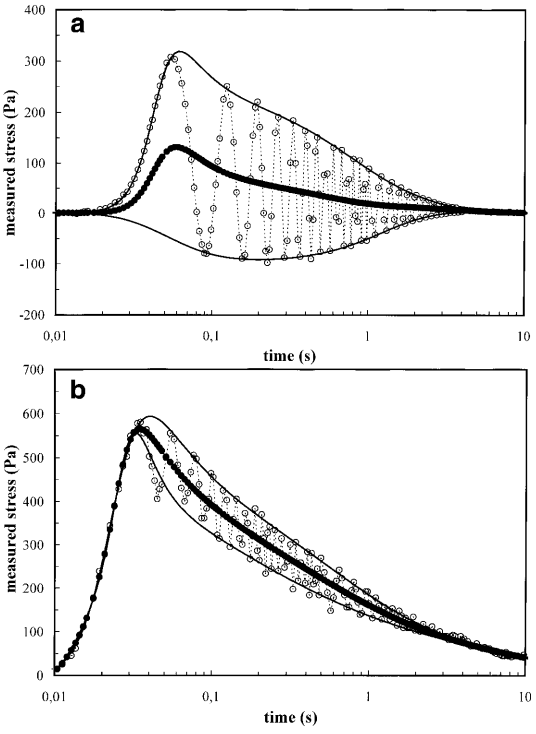

Fig. 7a, b Upper and lower boundary analysis of the relaxation signal for the curves of Fig. 5

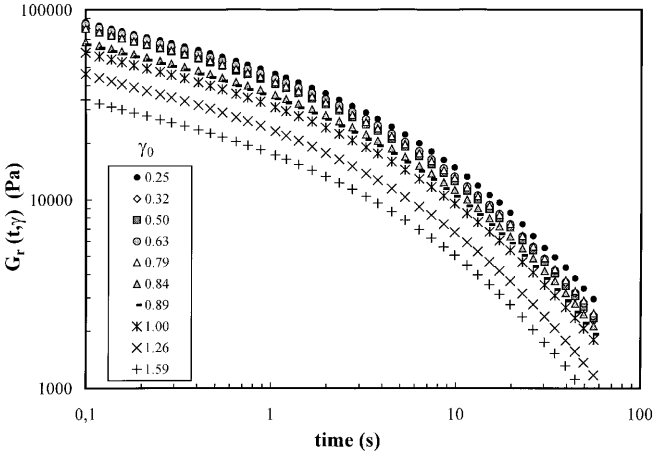

Fig. 8 Shear relaxation modulus for PIB B15

One other feature from Fig. 10 is the different rates at which the two materials relax that are in accordance with the results in shear, the material with lower molecular weight (B12) relaxing at a higher rate. This is a very important result since it clearly shows that the technique is sensitive to changes in the molecular structure and, thus, can be used (as long as the imposed strains are within the linear viscoelastic limit) as a non-destructive extensional probe of the microstructure of polymer melts (and, presumably, also other highly

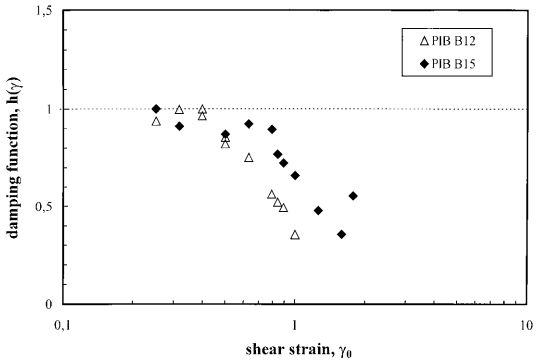

Fig. 9 Damping functions in shear for PIB B12 and B15

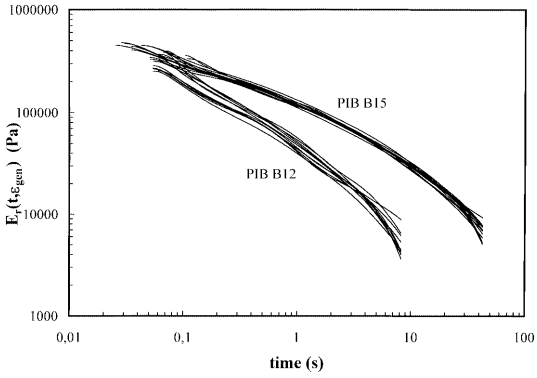

Fig. 10 Tensile relaxation modulus for PIB B12 and B15

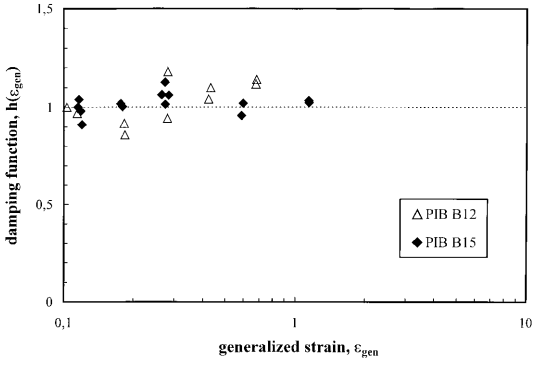

Fig. 11 Damping functions in uniaxial extension for PIB B12 and B15

viscous materials, such as some foodstuff), which is a capability that, as mentioned before, has been non-existent until now.

From fluid mechanics principles, stress relaxation experiments in extension should yield a value of 3 for the ratio between the plateau modulus, i.e., the short time values of the relaxation modulus, in extension and in shear. In the present case, the ratios are approximately 3.7 ± 0.1 for both materials, the slight variation arising

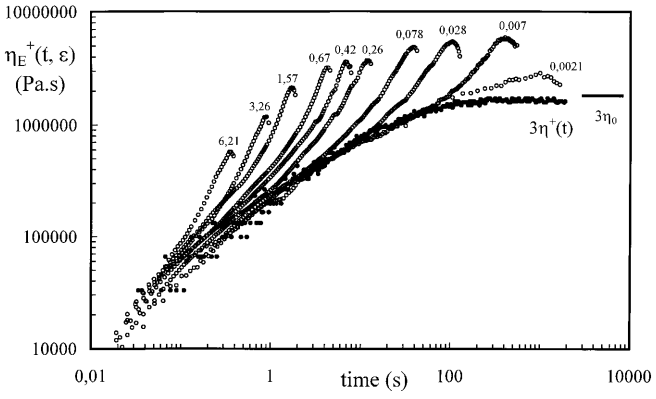

Fig. 12 Transient extensional viscosity at constant strain rate for PIB B15

from the standard deviation of the experiments. Given the high reproducibility of the experiments, i.e., the low standard deviation, the discrepancy between the observed and the expected ratios of plateau modulii cannot be attributed to the experimental technique itself but to errors in the data analysis procedure, especially at very short times where the upper and lower boundary fits tend to overestimate the true signal and, therefore, are probably not the best means of retrieving the relaxation data: work is currently underway to improve this shortcoming.

However, it is the view of the authors that, given the experimental difficulties involved in this type of experiments and the approximations used, the above result can be considered to be in good agreement with the theory and constitute a validation of the technique.

Conclusions

This paper aimed at establishing an operational procedure for performing stress relaxation measurements, after step deformations, in uniaxial extension, for polymer melts. It was shown that, given the inherent difficulties in performing extensional rheometry that add to those specific of the transient response of the equipment to such rapid deformations, it was possible to define such a methodology. For example, the observed relaxation behavior of the two test polyisobutylenes used was that expected from their different molecular weights, thus highlighting the technique's capability to act as a good microstructural probe and, also, the ratio of the plateau modulii in extension and in shear was approximately 3.7, which compares well with the expected value of 3.

However, there is still scope for improvement, namely in two respects:

1. The attainment of higher strains, because, given the mechanical limitations of the instrument, the present

maximum achievable strains may not be high enough to reach the non-linear viscoelastic regime, especially for highly elastic materials.
2. The data analysis technique, at very short times, needs to be improved in order to close the gap between the observed and expected ratios of plateau modulii.

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Appendix
Digitally filtered vs raw data

The hardware of the Weissenberg rheogoniometer allows the use of a digital filter which suppresses the components of the measured electrical signal (obtained from the LVDT torsion transducer) with frequencies higher than the cut-off frequency specified for a given torsion bar and thus acts as a data smoothing technique. Usually, as stated in the Weissenberg's users manual, the value of the cut-off frequency should be 40% lower than the resonant frequency of the torsion bar in use. However, the use of this digital filter in experiments that are extremely quick, such as stress relaxation, can corrupt the experimental results to a point where wrong behaviors will be observed, as will be shown below.

Figure A1 shows some experiments performed for the same extensional step strain, for PIB B12. The results refer to the same test as Fig. 5a, that is performed without digital filter and using torsion bar no. 7 (and where the oscillations are clearly seen) and two other tests performed with digital filtering and torsion bars no. 7 (less rigid) and no. 8 (more rigid), i.e., using different cut-off frequencies. As referred to in the users' manual, different cut-off frequencies lead to a slight increase in the time for updating the measured signal coming from the main controller to the computer, which explains the time lag between the maximums of both tests. In addition, the magnitudes of the maxima are themselves different. When these effects are added, the result is a very fast (artificial) initial decay of the stress.

Figure A2 shows a comparison between the two experiments of Fig. A1 for which torsion bar no. 7 was used. In Fig. A2, the experiment with digital filtering was shifted along the time axis (in order for the maximum to coincide with that of the averaged unfiltered experiment) and it is apparent that the measured stresses coincide after enough time has elapsed for the oscillation of the torsion bar to be digitally filtered, as would be expected.

The net effect of the fast initial relaxation behavior, when the digital filtering is on, however, is a corruption of the data up to a point where the relaxation modulus may actually be "found" to increase (!!) with deformation, as is illustrated in Fig. A3 for the case of PIB B15.

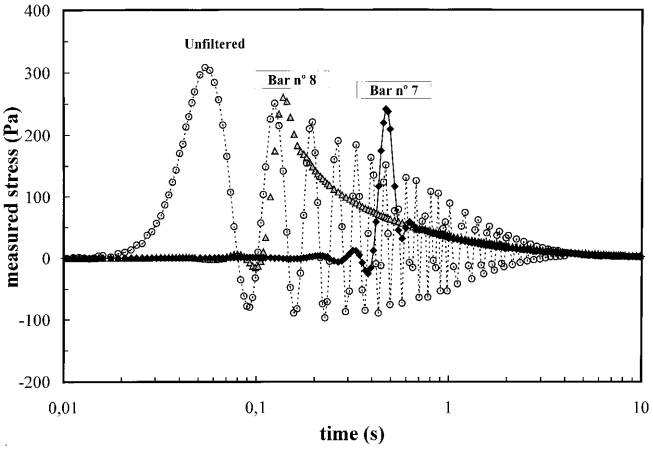

Fig. A1 Comparison between experiments performed with PIB B12 for a constant extensional step strain, using different torsion bars and different values for the digital filtering

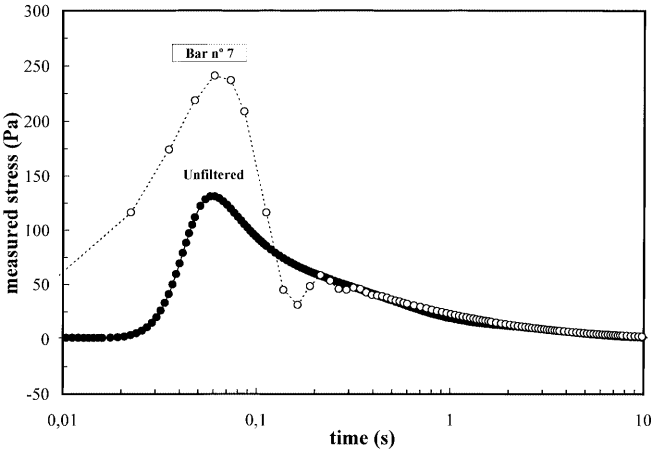

Fig. A2 Comparison between non-filtered (after averaging) and filtered results for PIB B12 (the curves and symbols correspond to those of Fig. A1), with torsion bar no. 7

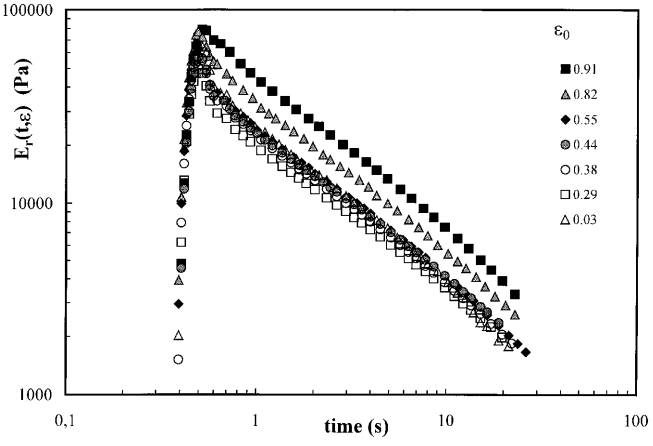

Fig. A3 Tensile relaxation modulus for PIB B15, determined using the filtered data

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