Rheol Acta (2000) 39: 444–451

ORIGINAL CONTRIBUTION

M. Kompani
David C. Venerus

Equibiaxial extensional flow of polymer melts via lubricated squeezing flow. I. Experimental analysis

Received: 22 March 2000
Accepted: 31 May 2000

M. Kompani · D. C. Venerus (✉)
Department of Chemical and Environmental Engineering and Center of Excellence in Polymer Science and Engineering
Illinois Institute of Technology
Chicago, Illinois 60616, USA
e-mail: cherenerus@iit.edu

Abstract The technique of lubricated squeezing flow is evaluated using experiments in both constant strain rate and constant stress flows of two low-density polyethylene melts. Experimental parameters that include the lubricant viscosity and sample aspect ratio are systematically varied to examine their effect on the viability of the technique in the linear viscoelastic regime by comparing measured quantities to those predicted by finite linear viscoelastic theory. An evaluation is also made by comparing viscosities measured using the lubricated squeezing flow technique and Meissner's rotating clamp (MAD) rheometer in the nonlinear regime. In both cases, deviations between the expected and measured viscosities were observed, indicating that the technique is not applicable to large strains for constant strain rate and constant stress flows. It is suggested that this limit is the result of lubricant thinning.

Introduction

Equibiaxial extensional flow is a deformation in which a material element is stretched by equal amounts in two directions and compressed in the third direction. This type of deformation occurs frequently in polymer fabrication and forming processes such as blow molding, compression molding, and the production of polymer foams (bubble growth). Despite its significance, our understanding of the rheological behavior of polymeric materials in equibiaxial extensional flow is limited. This is especially true in comparison to our knowledge of rheological behavior in simple shear and uniaxial extensional flows. The reason for this disparity is that experimental methods for generating rheologically-controlled equibiaxial extensional flows are not well developed.

One of the apparently more promising techniques for conducting controlled equibiaxial extensional flow experiments is known as lubricated squeezing flow. This method has many advantages over other existing techniques, but also has some limitations. In this series of papers, a critical analysis of the lubricated squeezing flow technique is conducted using a combination of experimental and modeling approaches. It is hoped that from this work the technique and its limitations can be better understood, and that improved versions of the technique can be developed.

The need to study the rheological behavior of polymer melts in extensional, or shear-free, flow fields was recognized by rheologists many years ago (Bird et al. 1987; Macosko 1994). This led to extensive experimental and theoretical studies on the rheology of polymer melts in uniaxial extension. Conducting controlled uniaxial extensional flows on polymer melts, where the material is stretched in one direction and is compressed equally in the other two directions, represented a significant challenge to experimentalists. Much of the progress in this area is due to Meissner and coworkers (Meissner 1969, 1971, 1972) who developed extensional rheometers based on the 'fixed rotating clamp' (Meissner et al. 1981). As a result, a commercial version of this rheometer that uses rotating belt clamps (Meissner and Hostettler 1994) is now available. The study of uniaxial extensional flows of polymer liquids remains an active area of research.

Excluding lubricated squeezing flow, which will be discussed in the following section, several methods have been developed to study the behavior of polymer liquids in equibiaxial extensional flows. These methods, which include bubble inflation, axisymmetric stagnation flow, and sheet stretching, have been reviewed by Dealy (1978) and by Meissner (1987). In the bubble inflation method, a circular sample is clamped around its perimeter and inflated by imposing a pressure difference across the two sides of the membrane using a low viscosity fluid such as a gas or an oil causing an equibiaxial extension to occur in the region near the pole of the bubble. Because the deformation is not uniform throughout the sheet, several assumptions are usually made in order to determine the stress and strain. Axisymmetric stagnation flow involves impinging two polymer melt streams through lubricated hyperboloid walls to generate the desired flow. Steady state, constant strain rate properties can, in principle, be determined with this method from lubricant pressure or melt birefringence measurements. However, this method also relies on the use of several assumptions to extract deformation and stress.

The sheet stretching method for generating equibiaxial extensional flows of polymer melts is based on the fixed rotating clamp device mentioned earlier (Meissner et al. 1981). This device, which is known as the MAD (Multiaxiale Dehnung) rheometer, consists of eight pairs of rotating belt clamps in a circular arrangement (Meissner 1996; Hachman 1997). Each clamp is mounted on a force transducer and between each pair of clamps is a pair of scissors used to cut the sample periodically as it passes through the clamps. The strain rate is determined by a particle tracking analysis made from a video image of the deforming sample. From this description, one can imagine the complexity of such a device; in fact, only one MAD rheometer exists. This device has been used to obtain constant strain rate stress growth data on several polymer melts at strain rates of up to 0.1 $\sec^{-1}$ to Hencky strains of approximately three (Hachman 1997). This data set, like the device used to obtain it, is quite unique. The main drawbacks of the MAD rheometer are its complexity and its limited availability.

In this first paper, an experimental evaluation of the lubricated squeezing flow technique is conducted. In the following section of this paper, the technique of lubricated squeezing flow is briefly reviewed. In the third section, experimental considerations are discussed followed by a section in which the results of this work are presented. A summary of the study is given in the fifth and final section of this first paper.

Lubricated squeezing flow

The technique of lubricated squeezing flow was developed by Chatraei et al. (1981), and is shown schematically in Fig. 1. A disk-shaped sample of a polymer melt having radius R and thickness $2h$ is squeezed between parallel plates. Each plate is coated with a low viscosity lubricant film of thickness δ, and the total separation between the plates is $2H$. In an ideal experiment, the polymer melt undergoes equibiaxial extension, and shear flow takes place only in the lubricant films. As the deformation proceeds, however, *lubricant thinning* occurs, which can diminish the effectiveness of the lubricant films.

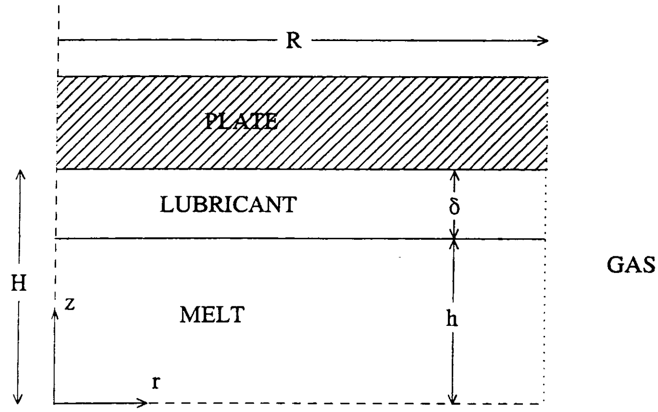

Fig. 1 Schematic diagram of lubricated squeezing flow

The axial symmetry of equibiaxial extensional flow leads to $S_{rr} = S_{\theta\theta}$ where **S** is the extra stress tensor; for constant volume flows, there exists a single, independent stress difference σ_b given by

$$\sigma_b = S_{rr} - S_{zz} \tag{1}$$

Deformation is usually expressed in terms of the Hencky equibiaxial strain, ε_b, given by

$$\varepsilon_b = \ln\left(\frac{R}{R_0}\right) = -\frac{1}{2}\ln\left(\frac{h}{h_0}\right) \tag{2}$$

where R_0 and $2h_0$ are the initial sample radius and thickness, respectively. The second equality in Eq. (2) reflects the constant volume assumption: $R/R_0 = (h_0/h)^{1/2}$.

The attractiveness of LSF as a rheological technique is based on its simplicity which, in turn, depends on the validity of two approximations. The first approximation is that the stress difference is given by (Chatraei et al. 1981)

$$\sigma_b = \frac{F}{\pi R^2} \tag{3}$$

where F is the force exerted on the solid plates. If the sample radius equals or exceeds the plate radius, then R in Eq. (3) indicates the plate radius. The rather simple expression in Eq. (3) appears to have a straightforward interpretation, but it is important to note that implicit in this result are several assumptions that have not been carefully examined. The second approximation is that the Hencky strain can be determined by (Chatraei et al. 1981)

$$\varepsilon_b = -\frac{1}{2}\ln\left(\frac{H}{H_0}\right) \tag{4}$$

which follows from Eq. (2) if the lubricant film thickness is much smaller than the melt thickness: $\delta < h$, since $H = h + \delta \simeq h$. While justified at the initial stage of an experiment, the validity of this approximation is dubious at later times when both h and δ are small and their relative sizes are not known.

The technique of lubricated squeezing flow (LSF) was first used (Chatraei et al. 1981) to conduct constant stress experiments on polydimethyl siloxane and polyisobutylene melts at room temperature. Soskey and Winter (1985) used LSF to carry out step-strain and constant strain rate experiments on polystyrene and low-density polyethylene melts. It was concluded in these studies that the optimum ratio of the melt to lubricant viscosity was from 500 to 1000 and that the maximum achievable strain was approximately 1. Based on these results, a number of studies followed including Khan et al. (1987) who used LSF to carry out step strain tests on several polymer melts, Hsu and Harrison (1991) who carried out constant stress and constant strain rate tests on a polystyrene melt, Takahashi et al. (1993) who carried out constant strain rate tests on polystyrene and polypropylene melts, and Ebrahimi et al. (1995) who carried out constant strain rate tests on a series of monodisperse polystyrene melts. In one study (1991), data was reported for Hencky strains greater than two and there was no mention of lubricant thinning problems. Also, viscosity ratios significantly greater than 1000 were used in several studies (Hsu and Harrison 1991; Takahashi et al. 1993) in contradiction to the results of Soskey and Winter (1985).

Given the absence of data sets against which data from LSF experiments might be checked, it is not surprising that in most of the studies mentioned above attempts to validate the LSF have been somewhat limited. In some cases, comparisons have been made using data obtained at small strains where one would expect lubricant thinning problems to be minimal. One exception is a comparison made using constant stress data from LSF and constant strain-rated data from a sheet stretching device (Chatraei et al. 1981) where reasonably good agreement was found.

From the discussion above, it is evident that the LSF technique, because of its simplicity, versatility and, availability, has a number of advantages compared with other methods for generating equibiaxial extensional flows. However, it appears that the operational limits of LSF are not well established, making the results obtained thus far using this technique somewhat suspect. In other words, the validity of the approximations used in Eqs. (3) and (4) have not been thoroughly examined.

Experimental considerations

LSF experiments were conducted on two different low density polyethylene melts. One is a widely studied material known as IUPACX, which was tested at 125 °C, and the other is a material studied by Meissner (Meissner 1996; Hachman 1997) known as Lupolen 1810H, which was tested at 150 °C. Linear viscoelastic characterization of these polymers was made using small strain amplitude oscillatory shear experiments conducted on a Rheometrics Mechanical Spectrometer (RMS-800). Table 1 shows the discrete spectrum of relaxation times τ_i with weights g_i where: $\tau_1 < \tau_2 \cdots < \tau_N$. From these data, the zero-shear viscosities of the two melts η_0^M were determined and found to be 1.5×10^5 Pa s for IUPACX at 125 °C and 6.5×10^4 Pa s for LUPOLEN at 150 °C.

A series of silicone lubricants (General Electric) with nominal viscosities at 25 °C ranging from approximately 1 Pa s to 500 Pa s were used as lubricants. Shear viscosities of the Newtonian lubricants η_0^L were measured at temperatures from 25 °C to 200 °C in cone and plate flow on the RMS-800. The ratio of the lubricant to melt viscosities at the test temperature is $\alpha = \eta_0^L/\eta_0^M$ and could be varied from 10^{-3} to 10^{-6}.

Two types of deformation histories were considered in this study – stress growth at constant strain rate and creep at constant stress. For stress growth experiments, a constant strain rate $\dot{\varepsilon}_{b0}$ was imposed by controlling the plate separation $H = H_0 \exp(-2\dot{\varepsilon}_{b0}t)$ according to Eq. (4), and measuring the average stress on the plates, $F/\pi R^2$. Figure 2 shows the good agreement between the measured and ideal Hencky strain history in a test where $\dot{\varepsilon}_{b0} = 0.001\,\mathrm{s}^{-1}$. In creep tests, a constant tensile stress difference σ_{b0} is imposed by controlling the plate separation to maintain a constant force $F = \sigma_{b0}\pi R^2$ according to Eq. (3), and measuring the resulting plate separation, $-1/2\ln(H/H_0)$. Figure 3 shows the actual stress difference for a test with $\sigma_{b0} = 800$ Pa.

Predictions from finite linear viscoelastic (FLV) constitutive equations (Dealy 1990) often provide a useful reference for examining rheological data. Once the relaxation spectrum (τ_i, g_i) has been determined for a fluid, the response can be predicted for different deformation histories and flow fields. For example, the equibiaxial viscosity $\eta_b^+ = \sigma_b/\dot{\varepsilon}_{b0}$ in the startup of constant strain rate flow is given by

$$\eta_b^+ = 2\sum_{i=1}^{N} g_i\tau_i\left[\frac{1 - e^{-(1-2\dot{\varepsilon}_{b0}\tau_i)t/\tau_i}}{1 - 2\dot{\varepsilon}_{b0}\tau_i} + 2\frac{1 - e^{-(1+4\dot{\varepsilon}_{b0}\tau_i)t/\tau_i}}{1 + 4\dot{\varepsilon}_{b0}\tau_i}\right] \tag{5}$$

Table 1 Relaxation spectra for LDPEs

i	1810H @ 150 °C		IUPACX @ 125 °C	
	τ_i [s]	g_i [Pa]	τ_i [s]	g_i [Pa]
1	0.0020	110470	0.0059	92985
2	0.0062	16304	0.0193	25010
3	0.0192	27180	0.0632	24200
4	0.0593	15050	0.207	13880
5	0.1840	10613	0.678	8968
6	0.5684	6397	2.21	4960
7	1.759	3625	7.27	2642
8	5.446	1899	23.8	1261
9	16.86	8258	77.9	598.4
10	52.18	300.4	256	144.5
11	161.5	67.6		
12	500.0	10.6		

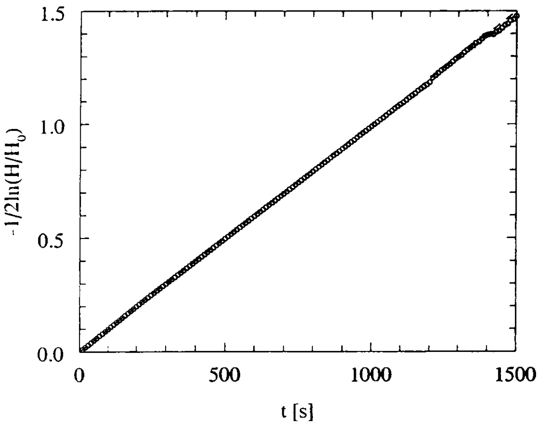

Fig. 2 Measured (○) strain $-1/2\ \ln(H/H_0)$ vs time for constant strain rate $\dot{\varepsilon}_{b0} = 10^{-3}\ s^{-1}$ test. *Dashed line* represents ideal experiment

For small strain rates $\dot{\varepsilon}_{b0}\tau_N \ll 1$, it is clear that

$$\eta_b^+ = 6\sum_{i=1}^{N} g_i\tau_i(1 - e^{-t/\tau_i}) = 6\eta^+ \tag{6}$$

where η^+ is the shear viscosity growth function. Figure 4 shows the good agreement between experimental and calculated shear viscosity growth functions for IUPACX at 125 °C and LUPOLEN at 150 °C. Similarly, FLV theory gives the following expression for flows at constant stress σ_{b0}:

$$\begin{aligned}\sigma_{b0} = \lambda(t)^{-1}\sum_{i=1}^{N}\left[g_i e^{-t/\tau_i} + \frac{g_i}{\tau_i}\int_0^t e^{-(t-\bar{t})/\tau_i}\lambda(\bar{t})\mathrm{d}\bar{t}\right] \\ - \lambda(t)^2\sum_{i=1}^{N}\left[g_i e^{-t/\tau_i} + \frac{g_i}{\tau_i}\int_0^t e^{-(t-\bar{t})/\tau_i}\lambda(\bar{t})^{-2}\mathrm{d}\bar{t}\right]\end{aligned} \tag{7}$$

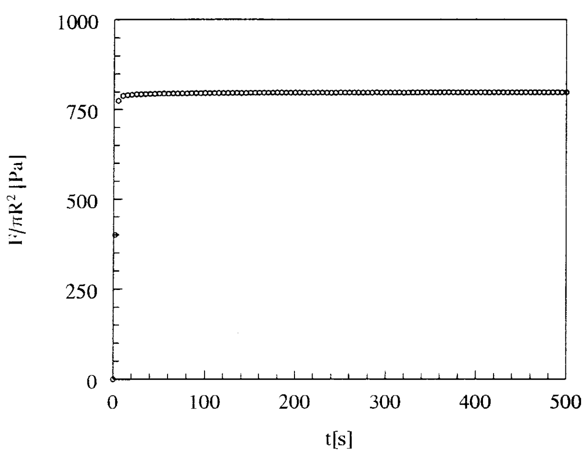

Fig. 3 Measured (○) stress difference $F/\pi R^2$ vs time for constant stress $\sigma_{b0} = 800$ Pa test. *Dashed line* represents ideal experiment

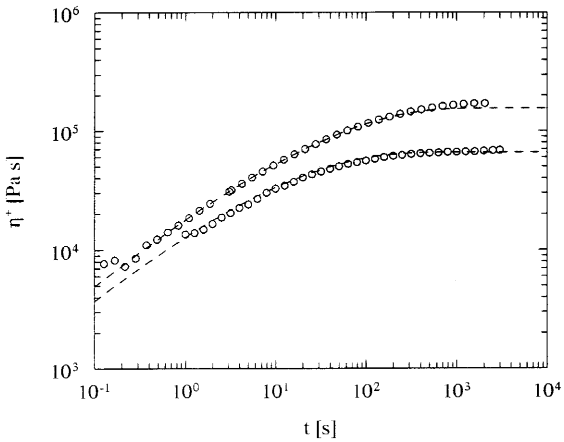

Fig. 4 Comparison of shear viscosities measured (○) at a strain rate of $10^{-3}\ s^{-1}$ and predicted by Eq. (6) (*dashed line*) for IUPACX at 125 °C (*top*) and Lupolen 1810H at 150 °C (*bottom*)

Equation (7) can be solved numerically for $\lambda(t) = h(t)/h_0$, the compression ratio, which is related to the Hencky strain: $\varepsilon_b = -1/2\ln[\lambda]$.

LSF experiments were conducted on a Rheometrics Solids Analyzer (RSA-II). This device has a pair of opposed force rebalance transducers to which circular plate fixtures were mounted. The lower transducer provides linear motion of one plate over the range ± 0.5 mm with a resolution of ± 0.001 mm. The second transducer measures the force required to hold the upper plate stationary with an upper limit of 1 kg and resolution of approximately 10^{-3} kg. The sample fixtures were contained in an environmental chamber wherein the temperature was controlled within ± 0.3 °C. The sample temperature was monitored with an additional thermocouple mounted to the lower plate.

The following procedure was used to produce the lubricated sample configuration shown in Fig. 1. After allowing the system to equilibrate at the test temperature, the gap between the plates was zeroed. Lubricant was loaded on the bottom plate with a syringe and then squeezed with the top plate to a thickness twice that of the desired initial film thickness. The plates were separated leaving approximately equal volumes of the lubricant on each plate. Samples, obtained from compression molded sheets having nominal thicknesses of 1 mm, were placed on the lower plate. Immediately (before the sample could melt) the upper plate was lowered, causing the lubricant on each plate to be squeezed into a film with uniform thickness, δ_0. The initial sample thickness, $2h_0$, and radius, R_0, were determined from room-temperature dimensions taking thermal expansion of the polymer into account. Two or three repeat runs were made and found to be within 15% of the reported averages.

The effects of initial lubricant film thickness and sample radius on the performance of the LSF technique were examined in this study. For convenience in making comparisons with model predictions presented in the second paper, these quantities were made dimensionless using the initial half-thickness of the gap between the plates H_0. The dimensionless initial sample thickness is given by $\chi = h_0/H_0$; the initial sample aspect ratio is given by $\kappa = R_0/H_0$. All constant strain rate experiments were conducted with samples having a radius (that changed with time) smaller than the plate radius, while all constant stress experiments were conducted with samples having an initial radius equal to the plate radius so that the effective sample radius was constant ($\kappa = R/H_0$)

Results and discussion

In this section we present results showing the effects of sample aspect ratio (κ), lubricant viscosity (α), and lubricant thickness ($1-\chi$) on the performance of LSF experiments conducted in both constant strain rate and constant stress modes. The evaluations are made by comparing the measured equibiaxial viscosity, $F/(\pi R^2 \dot{\varepsilon}_{b0})$, or Hencky strain, $-1/2\ln(H/H_0)$, with the expected value, the latter being determined from FLV theory, Eqs. (5) and (7), respectively. In one case, we make comparisons outside the linear regime using constant rate equibiaxial extensional flow data from the rotating-clamp instrument (Meissner 1996; Hachman 1997).

Figure 5 shows the measured biaxial viscosity from LSF experiments at a constant strain rate $\dot{\varepsilon}_{b0} = 10^{-3}$ s^{-1} for the IUPACX melt where the viscosity of the lubricant was varied over several orders of magnitude. A reasonable estimate of the average relaxation time (see relaxation spectrum shown in Table 1) is 100 s. Hence, the dimensionless strain rate for this flow is on the order of 0.1; one would expect the fluid response to be linear viscoelastic. From this figure it appears for lubricant viscosities in the range $10^{-5} \lesssim \alpha \lesssim 10^{-4}$ that there is good agreement between the measured and expected equibiaxial extensional viscosities, but only at relatively small strains: $\varepsilon_b \lesssim 0.2$. At larger strains, the measured viscosity is significantly higher than the expected one. Soskey and Winter (1985) ($\alpha \simeq 2 \times 10^{-3}$) and Ebrahimi et al. (1995) ($\alpha \simeq 10^{-6}$) found similar results using lubricants having different relative viscosities. Takahashi et al. (1993) also examined the effect of lubricant viscosity, but did not observe the systematic dependence on α nor the dramatic increase in viscosity above the expected value as is shown in Fig. 5.

The effect of lubricant thickness on LSF experiments is shown in Fig. 6 for the IUPACX melt with $\dot{\varepsilon}_{b0} = 10^{-3}\,\mathrm{s}^{-1}$. These data show for initial lubricant thicknesses that are 2–6% of the initial sample thickness ($\chi = 0.96 - 0.90$), good agreement between the measured and expected equibiaxial extensional viscosities at small strains is obtained, and that increasing lubricant thickness leads to a slight increase in the strain at which the LSF technique fails. However, increasing the initial lubricant thickness further ($\chi = 0.86$) causes the measured viscosity to fall below the expected value even at small strains. The acceptable range of initial lubricant thickness for LSF established by the data in Fig. 6 is consistent with values used in previous studies.

Figure 7 shows the sensitivity of the LSF technique to sample aspect ratio, again for the IUPACX melt with $\dot{\varepsilon}_{b0} = 10^{-3}\,\mathrm{s}^{-1}$. Surprisingly, the strain at which deviations between the measured and expected equibiaxial extensional viscosities occur appears to increase with decreasing initial sample aspect ratio $\kappa = R_0/H_0$. One would expect edge effects, which would be rather complicated and difficult to quantify, to be more pronounced for small sample aspect ratios. These results are, however, consistent with results from previous studies (Chatraei et al. 1981; Hsu and Harrison 1991) where even smaller aspect ratios ($\kappa \simeq 2$) were used.

Typical results from an LSF experiment conducted in the constant stress mode are shown in Fig. 8 for the IUPACX melt where the viscosity of the lubricant was varied over several orders of magnitude. The applied stress for these data is $\sigma_{b0} = 800$ Pa, which, from linear viscoelasticity, should generate a strain rate of approx-

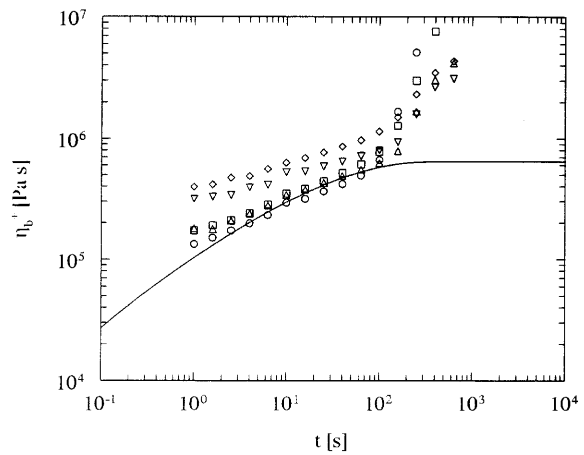

Fig. 5 Comparison of equibiaxial extensional viscosities measured using LSF, $F/\pi R^2\dot{\varepsilon}_{b0}$, (*symbols*) and predicted by Eq. (5) (*solid line*). IUPACX at 125 °C for constant strain rate $\dot{\varepsilon}_{b0} = 10^{-3}\,\mathrm{s}^{-1}$ with $\kappa = 7.7$; $\chi = 0.90$; $\alpha = 1.4 \times 10^{-5}$ (○); 3.8×10^{-5} (△), 1.2×10^{-4} (□), 4.2×10^{-4} (▽), 1.0×10^{-3} (◇)

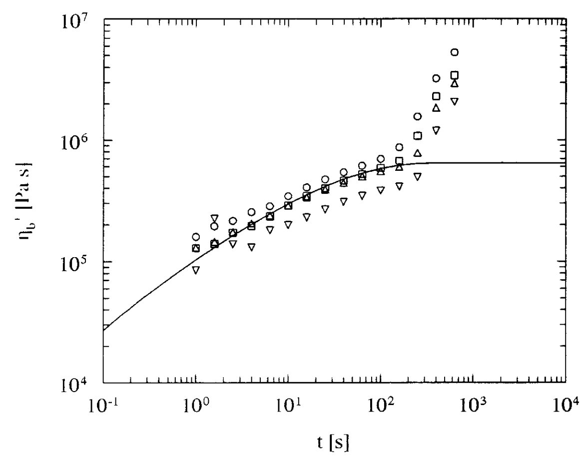

Fig. 6 Comparison of equibiaxial extensional viscosities measured using LSF, $F/\pi R^2\dot{\varepsilon}_{b0}$, (*symbols*) and predicted by Eq. (5) (*solid line*). IUPACX at 125 °C for constant strain rate $\dot{\varepsilon}_{b0} = 10^{-3}\,\mathrm{s}^{-1}$ with $\alpha = 1.2 \times 10^{-4}$; $\kappa = 7.7$; χ = 0.86(▽), 0.90 (□), 0.93 (△), 0.96 (○)

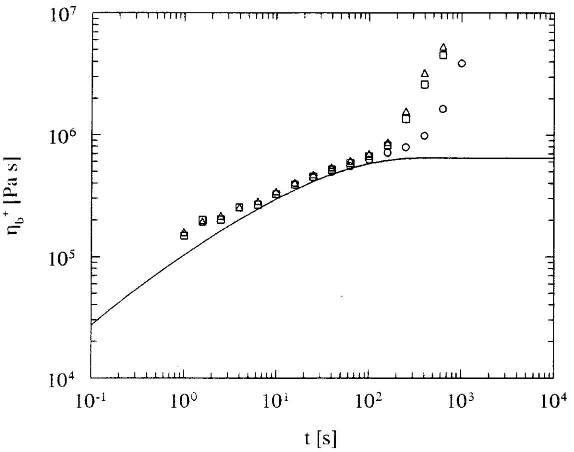

Fig. 7 Comparison of equibiaxial extensional viscosities measured using LSF, $F/\pi R^2\dot{\varepsilon}_{b0}$, (*symbols*) and predicted by Eq. (5) (*solid line*). IUPACX at 125 °C for constant strain rate $\dot{\varepsilon}_{b0} = 10^{-3}\,s^{-1}$ with $\alpha = 1.2 \times 10^{-4}$; $\chi = 0.86$; $\kappa = 4.7$ (○), 7.7 (□), 9.2 (△)

imately $10^{-3}\,s^{-1}$. Initially, the strain rate is approximately independent of lubricant viscosity and then increases with increasing α. Chatraei et al. (1981) found a similar dependence on lubricant viscosity (α). However, the apparent strain at which deviations between the measured and expected strain occur is somewhat lower in this study compared to several previous studies (Chatraei et al. 1981; Hsu and Harrison 1991). The reason for this difference may in part be due to the smaller aspect ratios used in these studies. It is important to note for constant stress LSF experiments where the constant area technique is used that a large amount of the polymer accumulates at the edge of the disks; its effect is difficult to estimate.

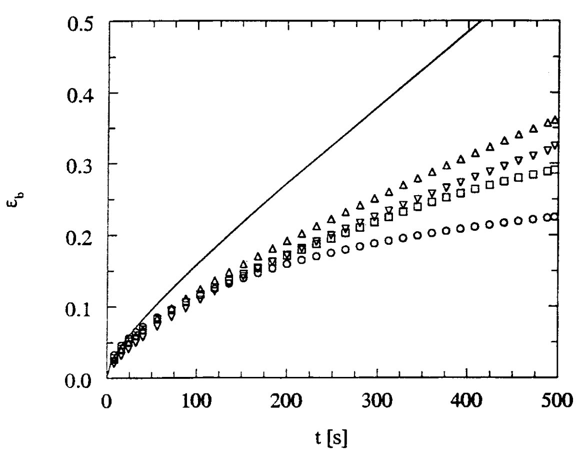

Fig. 8 Comparison of equibiaxial extensional strains measured using LSF, $-1/2\ln(H/H_0)$, (*symbols*) and predicted by Eq. (7) (*solid line*). IUPACX at 125 °C for constant stress $\sigma_{bo} = 800$ Pa with $\kappa = 15.0$; $\chi = 0.90$; $\alpha = 1.1 \times 10^{-6}$ (○), 1.4×10^{-5} (□), 1.2×10^{-4} (▽), 1.0×10^{-3} (△)

The deviations between the experimentally measured and linear-viscoelastic predicted responses shown in Figs. 5–8 appear to represent failures of the LSF technique. However, one should be aware of other possible explanations. For example, one might argue that an increase in the measured equibiaxial extensional viscosity above the linear viscoelastic prediction, Eq. (6), is simply a 'strain hardening' response (Dealy 1990) of the fluid. We also mention that the Hencky strain at which these deviations occur ($\varepsilon_b \lesssim 0.2$–$0.4$) found in this study is similar to that found by Soskey and Winter (1985), but smaller than that reported in several studies (Hsu and Harrison 1991; Takahashi et al. 1993; Ebrahimi et al 1995). A possible explanation for this discrepancy is that a different LSF configuration, involving a centering rod through the sample and an oil bath surrounding the sample, similar to the original setup (Chatraei et al. 1981), was used in these studies.

The availability of equibiaxial extensional viscosity data obtained using the rotating-clamp device (Meissner 1996; Hachman 1997) provides a unique opportunity to evaluate the LSF method outside the linear viscoelastic limit. Figure 9 shows a comparison of equibiaxial extensional viscosities measured using the rotating-clamp rheometer and the LSF technique for the Lupolen 1810H melt at strain rates $\dot{\varepsilon}_{b0}$ equal to 0.01 s^{-1} and 0.1 s^{-1} that roughly correspond to dimensionless strain rates of 1 and 10, respectively. Under these conditions, one would expect the response of the fluid to be nonlinear. It should be mentioned that the larger stresses developed at larger strain rates led to errors in the prescribed strain history due to the compliance of the force transducer in the RSA II. The LSF data shown in Fig. 9 are, however, free of significant compliance

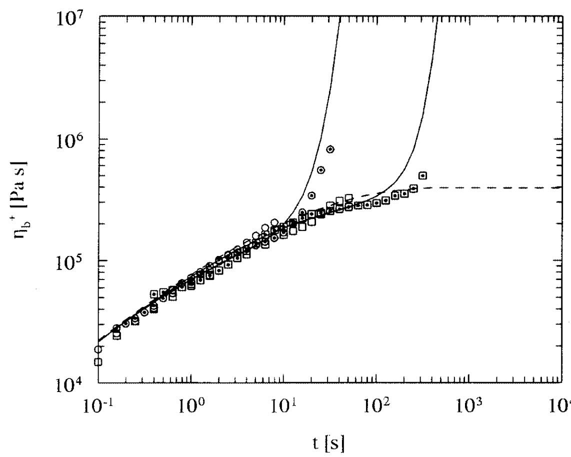

Fig. 9 Comparison of equibiaxial extensional viscosities for Lupolen 1810H at 150 °C. *Solid lines* are predictions of Eq. (5) and *dashed line* is prediction of Eq. (6). Rotating clamp MAD rheometer data (Meissner 1996; Hachman 1997): $\dot{\varepsilon}_{b0} = 10^{-2}\,s^{-1}$ (⊡), 10^{-1} s^{-1} (⊙); LSF data ($F/\pi R^2\dot{\varepsilon}_{b0}$) with $\alpha = 1.2 \times 10^{-4}$ and $\chi = 0.92$: $\dot{\varepsilon}_{b0} = 10^{-2}\,s^{-1}$ (□), 10^{-1} s^{-1} (○)

effects. The rotating-clamp rheometer data exhibit a pronounced increase above the linear viscoelastic prediction (dashed line), or strain hardening, at Hencky strains in the range 1.5–2.0. Also, as expected (Dealy 1990), the rotating-clamp rheometer data are always below the FLV prediction. The data obtained using the LSF technique show deviations from the rotating-clamp data at Hencky strains of approximately 0.2–0.4, a strain level consistent with the data collected at lower strain rates for the IUPACX melt. Also, the LSF data increase above the finite linear viscoelastic prediction, a behavior that would not be expected of typical viscoelastic fluids (Dealy1990). From these data it is reasonable to conclude that effects not associated with the fluid behavior are manifested in LSF experiments at relatively small strains.

Conclusion

The technique of lubricated squeezing flow has been evaluated through a series of experiments in both constant strain rate and constant stress flows using two low-density polyethylene melts. The lubricant viscosity, lubricant thickness, and sample aspect ratio were systematically varied to examine their effect on the technique by comparing the viscosity or strain measured in LSF experiments to those predicted by linear viscoelasticity. Since these experiments were conducted within the linear regime, any difference between measured and predicted quantities could be attributed to a failure of the LSF technique. An evaluation was also made by comparing viscosities measured using the LSF technique and Meissner's rotating clamp (MAD) rheometer at larger strain rates; deviations between the measured viscosities were again observed at small strains. These results suggest that the LSF technique is limited to relatively small Hencky strains for constant strain rate and constant stress equibiaxial extensional flows.

The upper limit on strain found in this investigation is somewhat lower than those reported in previous studies. A possible explanation for this discrepancy is that, unlike the present study, the LSF setup used in several previous studies involved a centering rod passing through the sample and an oil bath surrounding the sample. How such modifications would enhance the LSF technique is not known by the authors. In addition, smaller sample aspect ratios were used in these studies reporting data at larger strains. Again, how a small aspect ratio would enhance the technique is unknown; indeed, one would expect edge effects to be more pronounced in such cases.

The most probable explanation for the maximum strain limit in LSF experiments is lubricant thinning. Clearly, drag flow induced by flow of the melt would cause lubricant to flow from the film, thereby diminishing its effectiveness. The question of how lubricant thinning affects the validity of the approximations in Eqs. (3) and (4), and thus the viability of the technique, is addressed in the second paper of this series using a flow model of LSF. In any event, it appears a modification of the technique that resolves the lubricant thinning problem is necessary if lubricated squeezing flow is to be used to collect meaningful equibiaxial extensional flow data at Hencky strains comparable to those that can be attained using Meissner's rotating clamp (MAD) rheometer.

Acknowledgments The financial support provided by the Amoco Foundation for this study is gratefully acknowledged. The authors are also grateful to Professors J. Meissner and H.C. Öttinger of the ETH, Zürich for making the equibiaxial extensional flow data collected on the MAD rheometer and the test material (Lupolen 1810H) available to us.

References

Bird RB, Armstrong RC, Hassager O (1987) Dynamics of polymeric liquids, vol 1: fluid mechanics. Wiley-Interscience, New York

Chatraei SH, Macosko CW, Winter HH (1981) Lubricated squeezing flow: a new biaxial extensional rheometer. J Rheol 25(4):433–443

Dealy JM (1978) Extensional rheometers for molten polymers: a review. J Non-Newtonian Fluid Mech 4:9–21

Dealy JM (1990) Do polymeric liquids exhibit strain hardening? J Rheol 34(7):1133–1147

Ebrahimi NG, Takahashi M, Araki O, Masuda T (1995) Biaxial extensional flow behavior of mondisperse polystyrene melts. Acta Poly 46:267–271

Hachmann P (1997) Multiaxiale Dehnung von Polymerschmelzen, Ph.D. Thesis, ETH Zürich

Hsu TC, Harrison IR (1991) Measurement of the equibiaxial elongation viscosity of polystyrene using lubricated squeezing. Poly Eng Sci 31:223–230

Khan SA, Prud'homme K, Larson RG (1987) Comparison of the rheology of polymer melts in shear, and biaxial and uniaxial extensions. Rheol Acta 26: 144–151

Macosko CW (1994) Rheology: principles, measurements and applications, VCH, New York

Meissner J (1969) Rheometer zur Untersuchung der deformation-mechanischen Eigenschaften von Kunstoff-Schmelzen unter definierter Zugbeanspruchung. Rheol Acta 8:78–88

Meissner J (1971) Dehnungsverhalten von polyäthylen-schmelzen. Rheol Acta 10: 230–242

Meissner J (1972) Development of a universal extensional rheometer for the uniaxial extension of polymer melts. Trans Soc Rheol 16(3):405–420

Meissner J, Raible T, Stephenson SE (1981) Rotary clamp in uniaxial and biaxial extensional rheometry of polymer melts. J Rheol 25(1):1–28

Meissner J (1987) Polymer melt elongation-methods, results, and recent developments. Polym Eng Sci 27:537–546
Meissner J, Hostettler J (1994) A new elongational rheometer for polymer melts and other highly viscous liquids. Rheol Acta 33:1–21
Meissner J (1996) In: Kadi AA, Dealy JM, James DF, Williams MC (eds) Proceedings of the XIIth International Congress on Rheology, Quebec City, Quebec 7–10
Soskey PR, Winter HH (1985) Equibiaxial extension of two polymer melts: polystyrene and low density polyethylene. J Rheol 29(5):493–517
Takahashi M, Isaki T, Takigawa T, Masuda T (1993) Measurement of biaxial and uniaxial extensional flow behavior of polymer melts at constant strain rates. J Rheol 37(5):827–846