Lubricated Squeezing Flow: A New Biaxial Extensional Rheometer

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Synopsis

Squeezing flow between two disks with lubricated surfaces was found to generate a homogeneous compression or equal biaxial extension in a high viscosity polydimethylsiloxane sample. The apparatus is extremely simple: two steel disks with a central rod to provide alignment and prevent sample slip, an LVDT to measure displacement, and a silicone oil bath. The mass and area of the upper disk provide for a constant force boundary condition. The biaxial viscosity was found to be approximately six times the shear viscosity over biaxial extension rates $\dot{\epsilon}$, from 0.003 to 1.0 s^{-1}. Lubrication could be achieved up to Hencky strains of about 2.5. Some data were also taken on the same polyisobutylene sample used by Stephenson and Meissner in their biaxial stretching study. Agreement was very good.

INTRODUCTION

The research reported in this paper is concerned with an experimental method for measuring the biaxial extensional viscosity of viscous polymers. Biaxial flows are important in such polymer forming operations as blow molding, vacuum forming, film blowing, and foaming processes. There is also interest in the area of molecular theory, since it is expected that the behavior of materials under compression (or equal biaxial extension) can be different than under extension (uniaxial extension).[1]

Petrie[2] has reviewed biaxial extension work through 1979 and pointed out that relatively little has been done in this area. Of the

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work to date essentially two methods have been used; bubble inflation and sheet stretching.

In bubble inflation, the most popular method, a thin polymer sheet is inflated using an inert gas or liquid. The experiment is normally carried out at constant stress. The results in this area have been mostly obtained at low extension rates $10^{-4} < \dot{\epsilon} < 10^{-2}\ s^{-1}$,[3,4] except Maerker and Showalter[5] who achieved up to $\dot{\epsilon} = 1.0\ s^{-1}$. The range of total strain, ϵ, has also been limited. Problems have been reported in: (1) obtaining and controlling constant stress, i.e., the bubble bursts before achieving steady state; (2) nonuniformity of the bubble thickness; and (3) unequal deformation except near the pole. All the results by this method show a decreasing extensional viscosity with increasing extension rate.

The sheet stretching method has been recently presented by Stephenson and Meissner.[6] They stretch a sheet of polymer with eight rotating clamps placed in an octagonal pattern. A sophisticated servo control system is required to coordinate the motion of these clamps. Stephenson and Meissner have been able to record the time dependent viscosity function at constant extension rate for $\dot{\epsilon} \leq 10^{-2}\ s^{-1}$.

The main idea in the present work is to compress a viscous material between two disks whose surfaces are lubricated. This can be viewed as the opposite of uniaxial extension experiments, such as the extension of a molten polymer rod.[7] Without lubrication we would have the well known squeezing of Stefan flow which is a combination of shear and extension.[8,9] Lubricated squeezing flow is closely related to a commercial process for forming shapes from lubricated sheets.[10]

The stress in equal biaxial extensional flow has a principal plane normal to the axis of symmetry. Principal planes are defined as planes of vanishing shear stress. In lubricated squeezing flow, the top and the bottom of the samples (note Figs. 1 and 2) are such principal planes. The purpose of the lubricant between sample and wall is achievement of a vanishing shear stress. This orientation of the lubricated interface is a major advantage of lubricated squeezing flow as compared to lubricated die flow,[11,12] and to lubricated stagnation flow.[12–14] In those experiments, the interface of lubrication is curved and it is always in an angle with the principal planes. Therefore, for stagnation flow a finite shear stress is required at the lubricated interface to achieve purely extensional flow in the sample.

THEORY

To calculate extensional viscosity we assume a homogeneous deformation, which implies perfect slip of the sample at the wall. With these assumptions for an incompressible material, the Hencky strain in vertical direction becomes:

$$\epsilon = \ln(h/h_0) \tag{1}$$

where h is the sample thickness; the strain rate is:

$$\dot{\epsilon} = (d\epsilon/dt) = \dot{h}/h \tag{2}$$

and the velocity field is:

$$\begin{aligned} v_z &= \dot{\epsilon}h, \\ v_r &= -\dot{\epsilon}r/2, \\ v_\theta &= 0. \end{aligned} \tag{3}$$

In this experiment we measure thickness as a function of time, then the extension or h/h_0 and Hencky strain in vertical direction are calculated, and $\dot{\epsilon}$ is obtained as the slope of the Hencky strain curve versus time.

The experiment is carried out at constant force. Since the area of the disks is constant and if we can neglect edge effects, then the normal stress difference becomes:

$$T_{zz} - T_{rr} = F/\pi R^2 \tag{4}$$

The radial extension rate:

$$\dot{\epsilon}_r = -\tfrac{1}{2}\dot{\epsilon} = -\tfrac{1}{2}\dot{h}/h \tag{5}$$

is used to define an equal biaxial extensional viscosity:

$$\eta_b = (T_{rr} - T_{zz})/\dot{\epsilon}_r. \tag{6}$$

For a Newtonian fluid, η_b is equal to $6\eta_0$.

EXPERIMENTAL METHOD

The experimental apparatus (see Fig. 1) consists of two coaxial circular disks with a thin rod going through holes in the middle of the disks. The disk diameters were 57.2 and 127 mm. The rod provides alignment and prevents the lubricated sample from sliding from between the disks during the experiment. As shown in Figure 1, the

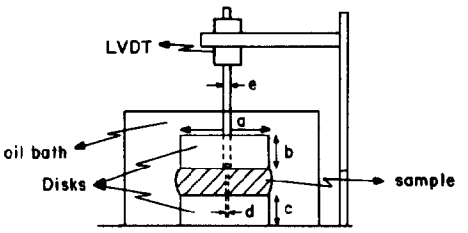

Fig. 1. Schematic of squeezing flow apparatus: (a) either 25.4, 50.8, 57.2 or 127 mm; (b) variable; (c) 19 mm; (d) 1.6 mm; (e) 4.8 mm.

upper disk is attached to the coil of a LVDT (linear variable differential transformer, model 245-004 series 240, Trans Tek, Elington, Connecticut). A strip chart recorder (0–10 V) or a minicomputer with an A/D converter (Data General, Nova II) were used to record h vs. time.

The whole experiment is carried out in an oil bath, the oil acting as lubricant, neutral density medium, and heat transfer agent. Two different oils were used as lubricants:

(1) hydrocarbon (UPS snow white petrolatum) $\rho = 840$ kg/m^3 and $\eta_0 = 0.060$ Pa·s;

(2) silicone (Dow Corning 200 fluid) $\rho = 910$ kg/m^3 and $\eta_0 = 0.453$ Pa·s ($\eta_0 = 4.5$ mPa·s was used for a few experiments).

The test material was an unvulcanized polydimethylsiloxane gum (PDMS) obtained from the Dow Corning Corp., with a weight average molecular weight of 687,000 and density of 980 kg/m^3. Its shear viscosity was found to be approximately constant, $\eta_0 = 2.70 \times 10^4$ Pa·s (at $T = 29$°C), up to a shear rate of 0.02 s^{-1}. PDMS was chosen since it is a fluid at room temperature and since it has a viscosity comparable to the viscosity of molten polymers. Leaving about 30 g of PDMS in the hydrocarbon oil for five days had no effect on η_0; however, the same treatment in the silicone oil reduced η_0 by 10%. One day in silicone oil had no measureable effects on η_0. Typical residence time in the lubricated squeezing tests was less than two hours.

Samples were molded in a lubricated tube of the disk diameter. To completely eliminate air bubbles from the sample required about ten days at ambient condition, but twenty-four hours was sufficient to eliminate all large bubbles. The remaining small bubbles did not appear to effect the stress strain response.

Force is determined from the weight of the upper disk and the

LVDT core. Errors in the force due to changing buoyancy of the LVDT core entering the bath are less than 0.4%. There is also a small amount of frictional force between the core and the LVDT. By careful alignment this fraction can be made less then 0.1 N. In later experiments an air bearing (0.25 in. diam; Dover, Boston, MA) was used on rod e. It made alignment easier and travel smoother but increased friction to about 0.4 N.

RESULTS

In order to check the velocity profile, a vertical ink line was placed in the undeformed sample with a syringe. Figure 2 shows photographs of one of these tracer experiments. Within the accuracy of

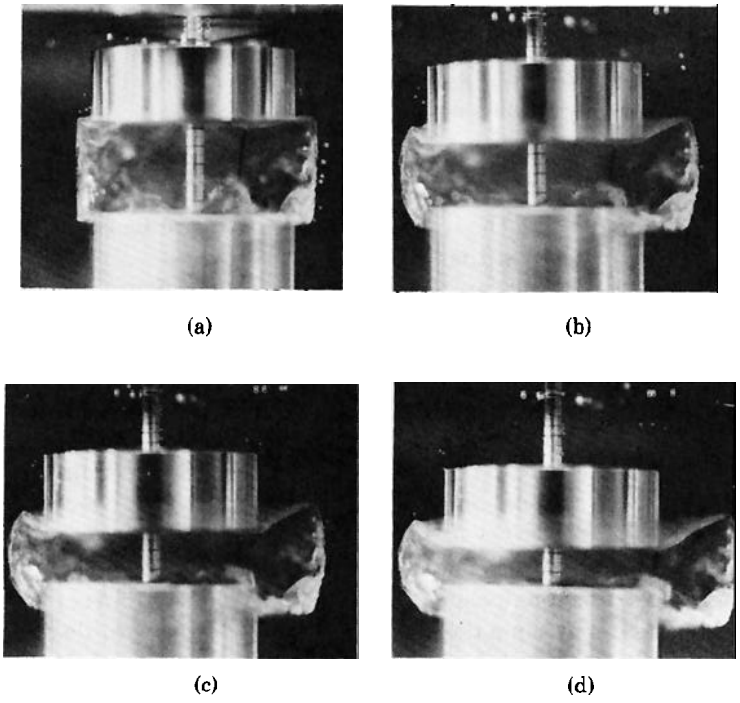

Fig. 2. Photographs of a lubricated squeezing flow experiment with a tracer line; $h_0 = 30$ mm, $R = 28.6$ mm. The central rod here is larger ($d = 4.7$ mm) than that used for the experiments with the LVDT. (a) $h = 25$ mm, (b) 18 mm, (c) 13 mm, (d) 9 mm.

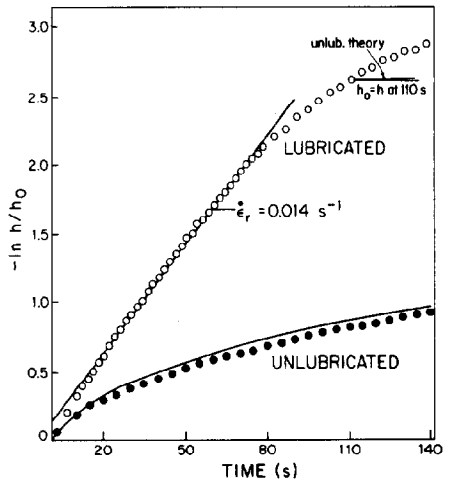

Fig. 3. Typical creep, $\epsilon = \ln(h/h_0)$ vs. time, (O) lubricated experiment F/A = 2.2 kPa, h_0 = 13.13 mm; (Φ) unlubricated experiment F/A = 2.0 kPa, h_0 = 17.01 mm; (—) Newtonian theory for unlubricated squeezing flow.

the photographs and the tracer lines, the velocity profile is flat, and we have perfect slip at the top wall.

Typical creep results are shown in Figure 3. In this figure, note that for approximately the same force the lubricated disks squeeze much more rapidly than the unlubricated ones. The simple Newtonian solution for the unlubricated case overpredicts the deformation but converges toward it at long time. Both of these features are expected for viscoelastic fluids.[8]

The lubricated creep curves shown in Figures 3 and 4 appear to consist of three regions: an initial transient response, then a constant slope (see lines drawn) or steady state region, followed by the gradual loss of lubricant. The steady state region is used below, when determining extensional viscosity. As was mentioned above, perfect slip of the sample along the surface and a flat velocity profile are assumed. All data were obtained at 29°C. A comparison of the third region with the theory for no lubrication (Fig. 3) shows that there must be still some slip.

We tried three oils with different viscosities as lubricant, and the comparison is shown in Figure 4. We can see that lubrication can be

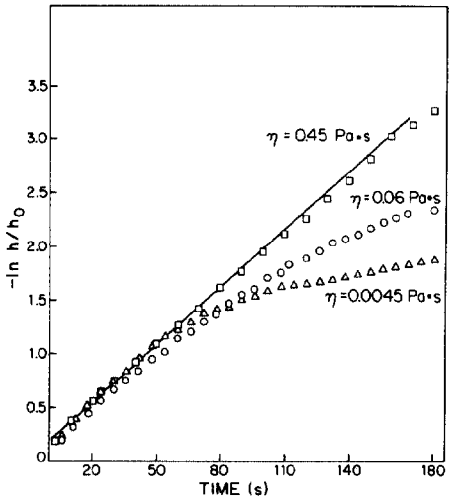

Fig. 4. $\epsilon = \ln(h/h_0)$ versus time, for lubrication fluids of different viscosity.

maintained up to a higher total strain with the lubricant of high viscosity. The cause of this may be simply that more of the viscous lubricant is trapped during the loading process.

We have made a crude attempt to estimate the average lubricant film thickness by quickly lifting up the sample from the lower disk and mopping up the oil film from both surfaces with a tissue. For $R = 25.4$ mm and $F/A = 13.76 \times 10^2$ N/m^2 from the weight gain of the tissue we estimated the lubricant film to be initially 0.5 mm thick. It decreased to 0.2 mm after 100 s, $\epsilon = 2.4$–2.5. These tests were done outside the oil bath; silicone oil was smeared on both disks before loading the sample.

To better illustrate the steady creep region we have plotted $\dot{\epsilon}$ for two of the experiments in Figure 5. "Instantaneous" extension rate was determined by taking differences between thickness data 0.2 to 5 s apart on the original LVDT traces. For each run a reasonable large region of steady extension was found. Except at the lowest force levels, all our results indicated steady extension up to strains of roughly 2.5.

Figure 6 compares the biaxial extensional viscosity values determined from the steady region of the lubricated squeezing experiment

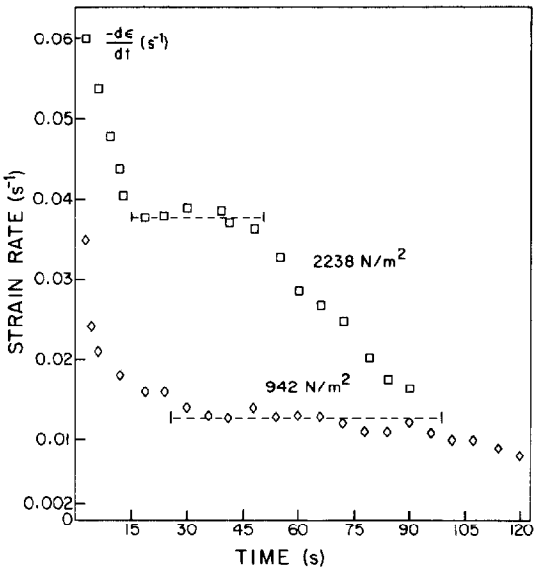

Fig. 5. Extension rate ($\dot{\epsilon} = d\epsilon/dt$) vs time. (□) F/A = 22.38 Pa, h_0 = 23.34 mm, for $12 < t < 50$ s, $\dot{\epsilon} = 0.038\ s^{-1}$; (◇) F/A = 9.42 Pa, h_0 = 29.33 mm, for $18 < t < 102$ s, $\dot{\epsilon} = 0.0128\ s^{-1}$.

with shear viscosity data. The steady shear viscosity was measured in the cone and plate mode using a Rheometrics Mechanical Spectrometer while the magnitude of the complex viscosity was determined in the oscillatory parallel plate mode of a Rheometrics Dynamic Spectrometer. It can be concluded that the extensional viscosity of this PDMS sample is basically Newtonian with thinning above 0.025 s^{-1}. The extensional data is in reasonable agreement in Newtonian region with $6\eta_0$ within the experimental accuracy; $\eta_b = (1.49 \pm 0.14) \times 10^5$ Pa·s while $6\eta_0 = (1.62 \pm 0.13) \times 10^5$ Pa·s. We can also express our results as viscosity versus square root of two times the second invariant of the rate of deformation tensor, or viscosity versus stress, and as one can imagine in both cases the whole shear curve would be shifted to the left (about 0.014 s^{-1}), therefore, extensional data would be higher than $6\eta_0$ in the non-Newtonian region. We also found good agreement between experiments using the four disk diameters. This result also indicates that edge effects and the influence of residual

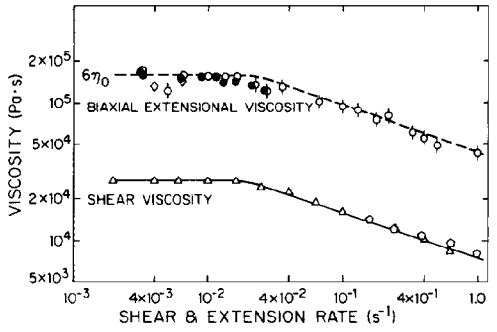

Fig. 6. Viscosities versus shear or extension rates. (△) Shear viscosities (by cone and plate), $T = 25°$ C; (⌂) dynamic viscosities (η^*), $T = 29°$ C; (◊) biaxial extensional viscosity, $R = 12.7$ mm; (○) biaxial extensional viscosity, $R = 25.4$ mm; (●) biaxial extensional viscosity, $R = 28.6$ mm; (◊) biaxial extensional viscosity, $R = 63.5$ mm; (---) $6\eta_0$.

stresses in the extruded material (note Fig. 2) are small. At the lowest stress level friction may be causing larger errors.

Results of Baily[16] and Joye et al.[4] show very similar behavior for polyisobutylene. We therefore tested our apparatus with the sample of Meissner and Stephenson. The steady stretching region is very long, see Figure 7. The high viscosity of the sample seems to be fa-

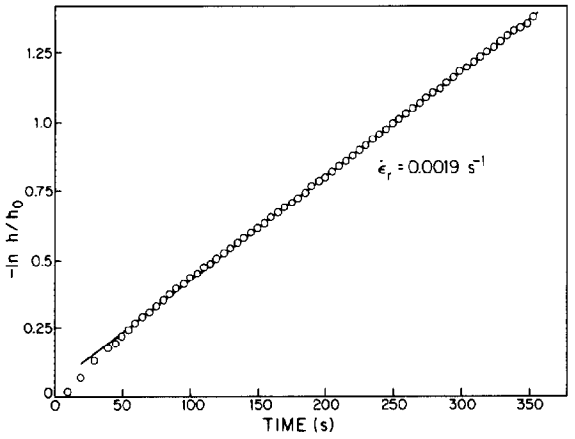

Fig. 7. Lubricated squeezing of polyisobutylene sample with silicone oil as lubricant. The sample is identical with the one used in Ref. 6. $\Delta\tau = 11.3$ kPa, $h_0 = 13.6$ mm.

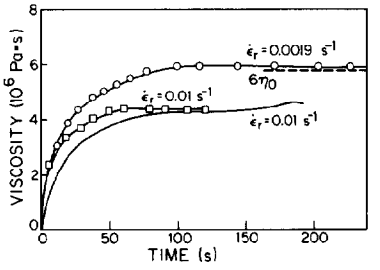

Fig. 8. Lubricated squeezing data are compared to sheet stretching data of Stephenson and Meissner.[6] (—) Biaxial extensional viscosity by sheet stretching[6] at $\dot{\epsilon}_r = 0.01\ s^{-1}$; (□) biaxial extensional viscosity by lubricated squeezing at $\dot{\epsilon}_r = 0.01\ s^{-1}$; (○) biaxial extensional viscosity by lubricated squeezing at $\dot{\epsilon}_r = 0.0019\ s^{-1}$; (- - -) $6\eta_0$ reported in Ref. 6 at $\dot{\gamma} = 0.001\ s^{-1}$.

vorable for the lubricated squeezing technique. Figure 8 compares time dependent viscosities as measured in the start up of lubricated squeezing with the viscosity measured in sheet stretching.[6] At an extension rate $\dot{\epsilon}_r = 0.01\ s^{-1}$, the agreement is remarkable: the extension rate is already outside the linear viscoelastic region. At a lower extension rate, $\dot{\epsilon}_r = 0.0019\ s^{-1}$, the squeezing data are close to $6\eta_0$ of shear flow.

CONCLUSION

It has been experimentally demonstrated that the biaxial extensional viscosity of a high viscosity polymeric liquid can be measured by the lubricated squeezing experiment with total strains as large as $\epsilon = 2.5$ and radial extension rates in the range $0.003 < \dot{\epsilon}_r < 1.0$. In this range our results for a polydimethysilxane sample indicate that the biaxial extensional viscosity is essentially six times the shear viscosity. Our results for polyisobutylene are in a good agreement with Stephenson and Meissner's results.

Presently we are redesigning the apparatus to give higher temperatures, wider stress range and better alignment. We also plan to study pressure distribution, lubriant thickness, and the influence of lubriant viscosity.

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