JOURNAL OF APPLIED POLYMER SCIENCE VOL. 18, PP. 1611–1617 (1974)

Measurements on the Planar Extensional Viscosity of Bulk Polymers: The Inflation of a Thin, Rectangular Polymer Sheet

C. D. DENSON and D. L. CRADY, *Major Appliance Laboratories, General Electric Company, Louisville, Kentucky 40225*

Synopsis

An experimental study of the inflation of a thin rectangular polymer sheet has been conducted to determine whether this technique can be used to measure the planar extensional viscosity of bulk polymers. Viscosities were determined at various extensional strain rates using an undiluted polyisobutylene of high molecular weight. Measurements were also made on the dynamic shear viscosity using orthogonal geometry and compared with "normalized" values of the planar extensional viscosity assuming equivalency of $\sqrt{II_e}$, the second invariant of the extensional strain rate tensor, and the frequency of rotation ω. Industrial fabrication operations where the planar extensional viscosity is expected to play a role include vacuum forming of large rectangular sheets whose length-to-width ratio is large and in injection molds having a center-gated cavity.

INTRODUCTION

In earlier publications,[1,2] we dealt with the problem of measuring the (uniform) biaxial extensional viscosity of bulk polymers and experimentally demonstrated that this particular rheological property could be measured by inflating a thin, circular polymer sheet using an inert gas. The use of a circular sheet in an inflation experiment results in a spherical bubble which grows with time. Since two of the principal stresses are equal at the pole of the bubble, the experiment is similar in nature to a biaxial creep experiment, and the viscosity can be determined by measuring the stress, which is a function of the bubble's internal pressure, and the strain rate, which is a function of the bubble's growth rate.

The presently reported research is an extension of our earlier work and deals with the inflation of a thin, rectangular polymer sheet. As we shall show this geometry provides data on the pure shear or planar extensional viscosity. Measurements on this type of viscosity have apparently not been previously made or reported in the published literature. Thus, we believe this work to be a contribution to the current state of the art.

In planar extensional flows, the velocity field has the form

$$v_1 = f(x_1),\ v_2 = g(x_2),\ v_3 = 0 \tag{1a}$$

© 1974 by John Wiley & Sons. Inc.

where the velocities v_1 and v_2 are related through the continuity equation by

$$\frac{\partial v_1}{\partial x_1} = -\frac{\partial v_2}{\partial x_2}. \tag{1b}$$

Under conditions when the principal strain rate e_{11} is constant and equal to $\dot{\epsilon}$, then

$$\frac{\partial v_1}{\partial x_1} = \dot{\epsilon}$$

and the strain rate matrix is given by

$$\mathbf{e} = \begin{vmatrix} \dot{\epsilon} & 0 & 0 \\ 0 & -\dot{\epsilon} & 0 \\ 0 & 0 & 0 \end{vmatrix} \tag{1c}$$

If one defines a planar extensional viscosity $\bar{\eta}_e$ in the following manner,

$$\bar{\eta}_e = \frac{T_{11} - T_{22}}{e_{11}} \tag{2}$$

then, in view of eqs. (1a) to (1c), it is relatively simple to show (see Appendix) that for a Newtonian fluid

$$\bar{\eta}_e = 4\eta_0. \tag{3}$$

EXPERIMENTAL

Except for the geometry of the hole in the upper plate and the use of a transducer, as opposed to a mercury manometer to measure the bubble's internal pressure, the experimental apparatus and procedure used in this work were identical to those used previously.

The pressure transducer used in this work was a Dynisco Model #PT25-20. It has a range of 0–20 psi, a full scale output of 4 mV/V, and a (combined) error of 0.75% full scale.

Concerning the geometry of the upper plate, we employed one here that had a rectangular hole whose dimensions were 1 in. × 4 in. Thus, when the polymer sheet was inflated, the resulting bubble's shape was that of a right circular cylinder whose cross section (in a plane perpendicular to the cylinder's axis) resembled the segment of a circle. Using this geometry, we were able to closely approximate planar deformation. In fact, measured values of the stretch in the axial direction were found to be less than 1% of those in the polar direction (Fig. 1).

All measurements were made at 23°C using an undiluted polyisobutylene whose viscosity-average molecular weight was 9.9×10^5. This material is manufactured by the Enjay Chemical Company and is designated as Vistanex L-80. In view of the extremely high molecular weight of this material, it was not possible to measure rheological properties under conditions of steady shear. It was possible, however, to obtain measured

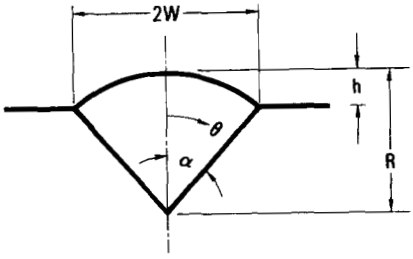

Fig. 1. Sketch of a flat sheet after inflation.

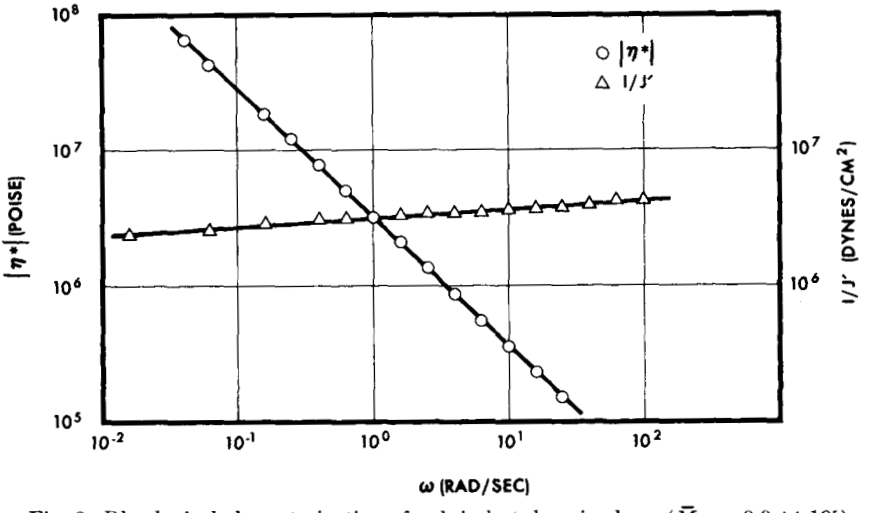

Fig. 2. Rheological characterization of polyisobutylene in shear ($\bar{M}_w = 9.9 \times 10^5$).

values for the complex shear viscosity and modulus using the Rheometrics Mechanical Spectrometer and operating the instrument in the orthogonal mode.[3] All measurements were made using parallel plates having a diameter of 2.5 cm, a distance of separation of 3.8×10^{-1} cm, and offset by 10^{-2} cm. Values for the magnitude of the complex viscosity and compliance are plotted in Figure 2.

DATA REDUCTION

Values for the planar extensional viscosity were calculated using eq. (2). In this apparatus, though,

$$T_{22} = 0$$
$$e_{11} = \dot{\epsilon} \tag{4}$$
$$T_{11} = \frac{PR}{\delta}$$

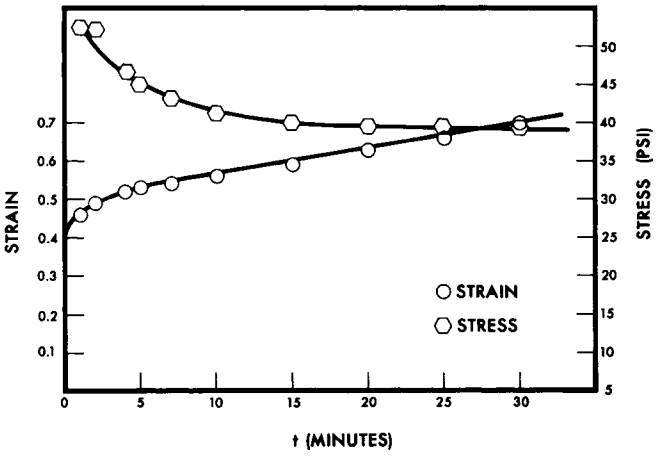

Fig. 3. Stress and strain plotted as a function of time. Although not shown here, the initial stress is zero.

where the stress T_{11} is identically equal to the hoop stress for a thin-walled cylindrical shell. The radius R and the thickness δ were calculated from the following expressions:

$$R = \frac{h}{2} + \frac{W^2}{2h} \tag{5}$$

$$\delta = \delta^* W/\alpha R \tag{6}$$

where

$$\alpha = \sin^{-1}\frac{W}{R}. \tag{7}$$

The strain rate was obtained by measuring the slope of the strain–time curve. Here, the strain is given by

$$\epsilon = \ln\frac{l}{l_0} = \ln\alpha\frac{R}{W}. \tag{8}$$

In using eq. (8), we have implicitly assumed that the strain–hence the thickness—and the stress do not vary with the polar angle. This assumption is probably quite good, for a force balance on the cylindrical bubble shows no dependence of stress (and consequently strain) on polar angle:

$$0 = \frac{\partial P}{\partial r} = \frac{\partial \tau_{rr}}{\partial r} + \frac{1}{r}[\tau_{rr} - \tau_{\theta\theta}] \tag{9a}$$

$$0 = \frac{\partial P}{\partial \theta} + \frac{\partial \tau_{\theta\theta}}{\partial \theta}. \tag{9b}$$

This is in direct contrast to the case of a spherical bubble where a force balance indicates that the stresses are dependent on polar angle. Conse-

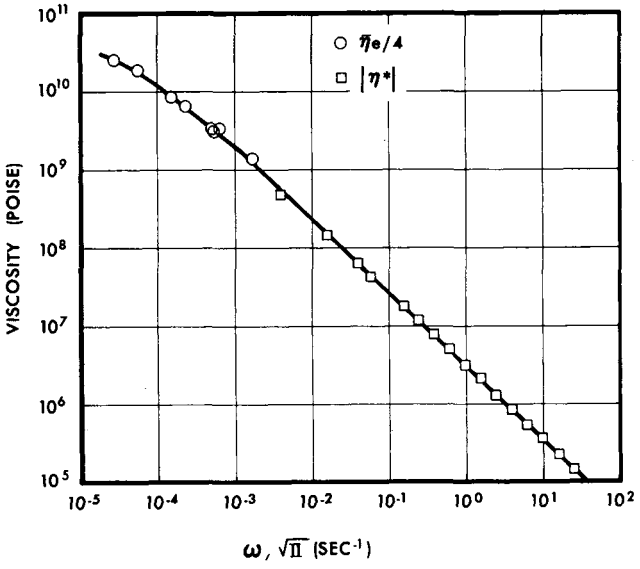

Fig. 4. Combined plot of "normalized" planar extensional and shear viscosities as a function of the second invariant for strain rate and angular velocity, respectively.

quently, calculated values for the uniform biaxial extensional viscosity are in error unless measurements are confined to the polar region.[2,4]

In Figure 3, we show a plot of strain and stress as a function of time. These data are representative of the kind obtained throughout the course of this study. The strain data are typical of those obtained in a creep experiment when a constant stress of known value is instantaneously imposed on a polymeric material. Note that the strain curve consists of three distinct regions. In the first region, there is an instantaneous increase in the strain. This is referred to as the initial elastic deformation, and its magnitude is determined by the modulus of elasticity of the polymer and by the value of the imposed stress. In the second region of the curve, retarded elastic deformation occurs and the strain varies nonlinearly with time. In the third region, flow occurs and, under the conditions of this experiment, the variation of strain with time is linear. It is in this region that we calculate the strain rate. As shown in the upper portion of the graph, the stress is essentially constant throughout the course of the experiment after the initial transients have decayed, which in this case is about 15 min. These transients result because a finite time is required to inject the gas and the stress is not imposed instantaneously.

Values for the planar extensional viscosity at various strain rates are listed in Table I; and in Figure 4, we have plotted values for a "normalized" ($\bar{\eta}_e/4$) planar extensional viscosity as a function of $|\mathrm{II}_e|^{1/2}$. Here, the II_e is given by

$$\mathrm{II}_e = \frac{1}{2}\left[(tr\ \mathbf{e})^2 - \mathbf{e}:\mathbf{e}\right] = 4\dot{\epsilon}^2. \tag{10}$$

TABLE I
Planar Extensional Viscosity at Various Strain Rates

Viscosity $\bar{\eta}_e \times 10^{-10}$, poise	Strain rate $\dot{\epsilon} \times 10^5$, sec^{-1}
10.2	1.4
7.7	2.8
3.4	7.5
2.6	11.0
1.4	25.0
1.3	27.0
1.4	32.0
0.5	86.0

On this same graph, we have plotted $|\eta^*|$ versus ω for the shearing case. The solid line drawn through the data points has no significance and is merely shown to highlight the proximity of the two sets of data. Unfortunately, data were not obtained in a range where values for $\sqrt{II_e}$ coincided with those for ω, and thus no conclusions can be drawn regarding the relationship between the extensional viscosity and shear viscosity. Dealy[5] has pointed out, though, that a direct relationship is not to be expected except at low strain rates where, according to eq. (3), $\eta_e = 4\eta_0$.

Vacuum forming is one polymer fabrication operation where the planar extensional viscosity plays a significant role. This is especially true if large rectangular sheets are being formed and clamping and drawing forces act in one direction as opposed to acting in two directions (biaxial). Thus, one would expect that any analysis of the vacuum-forming process should involve considerations of the planar extensional viscosity, and any correlations between properties of the polymer in the melt and vacuum formability should also be based on the planar extensional viscosity.

SUMMARY

We have demonstrated that the planar extensional viscosity of bulk polymers can be measured by inflating a thin, rectangular polymer sheet using an inert gas. In studying the behavior of polyisobutylene, we found that both the planar extensional viscosity and the shear viscosity decreased with increasing strain rate. No conclusions could be reached, however, regarding the relationship between the two viscosities since data were not obtained at equivalent values of $\sqrt{II_e}$ and ω.

Appendix

For a Newtonian fluid, the total stress tensor and the deviatoric stress tensor are related by

$$T_{ij} = -P\delta_{ij} + 2\eta_0 e_{ij}.$$

The normal stresses are thus given by

$$T_{11} = -P + 2\eta_0 e_{11}$$

$$T_{22} = -P + 2\eta_0 e_{22}$$

$$T_{33} = -P + 2\eta_0 e_{33}.$$

In planar extension, one of the principal strains is zero (e.g., $e_{33} = 0$), and, from the continuity equation,

$$e_{11} = -e_{22}.$$

Therefore, when extension is induced in the 11 direction,

$$T_{11} - T_{22} = 2\eta_0(e_{11} - e_{22}) = 4\eta_0 e_{11}$$

and

$$\bar{\eta}_e = \frac{T_{11} - T_{22}}{e_{11}} = 4\eta_0.$$

Nomenclature

e	strain rate tensor, sec^{-1}
h	height of bubble above surface of plate, cm
J'	compliance, cm^2/dyne
l	length of an extended line segment, cm
l_0	unstretched length of an extended line segment, cm
P	pressure, dynes/cm^2
r	radial coordinate, cm
R	radius of curvature, cm
T_{11}, T_{22}, T_{33}	total stress, dynes/cm^2
W	half-width of rectangular hole, cm
α	polar angle, rad
δ	thickness of distended film, cm
δ^*	thickness of unstretched film, cm
$\dot{\epsilon}$	strain rate, sec^{-1}
ϵ	strain
$\bar{\eta}_e$	planar extensional viscosity, poise
η_0	Newtonian shear viscosity, poise
τ_{rr}, $\tau_{\theta\theta}$	deviatoric stress in radial, polar directions, dynes/cm^2
θ	polar coordinate, rad
ω	angular frequency, rad/sec

References

1. C. D. Denson, and R. J. Gallo, *Polym. Eng. Sci.*, **11**, 174 (1971).
2. D. D. Joye, G. W. Poehlein, and C. D. Denson, *Trans. Soc. Rheol.*, **16**, 421 (1972).
3. J. M. Starita, private Communication, Major Appliance Laboratories Operation, General Electric Co., 1971.
4. D. D. Joye, G. W. Poehlein, and C. D. Denson, *Trans. Soc. Rheol.*, **17**, 287 (1973).
5. J. M. Dealy, *Polym. Eng. Sci.*, **11**, 322 (1971).

Received December 11, 1972
Revised August 14, 1973