Extensional Flow of Bulk Polymers

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A study was made of the behavior of polyisobutylene under motion at a constant stretch history for both strip biaxial extensional flow and simple extensional flow. Steady-state non-Newtonian viscosities were observed at various constant stretch histories. Newtonian viscosities for both strip biaxial and simple extensional flow were found to be in agreement with the classical theory.

The results of this study provide an essential part of the experimental background necessary for the development of a new general stress-strain-time relation for uncrosslinked and lightly crosslinked polymers.

Introduction

An experimental investigation of the behavior of uncrosslinked polymers subjected to a constant stretch history has been conducted for both pure shear extensional flow and simple extensional flow.

A standard Instron testing machine, in which crosshead displacement varies linearly with time was modified so that crosshead displacement varies exponentially with time. This change permits testing at constant Hencky strain rate ($\dot{\epsilon} = \dot{\ell}/\ell$), which provides a constant stretch history for both simple extensional flow and strip biaxial extensional flow. This results in a visualization of the steady flow for both types of deformation and enables one to derive the non-Newtonian viscosity from the steady-state region.

It has been shown (Reference 1) previously that in steady-state flow under constant stretch history, the stress functional of a non-Newtonian fluid reduces to a function of the three Rivlin-Ericksen tensors $\underset{\sim}{A}^{(1)}$, $\underset{\sim}{A}^{(2)}$, $\underset{\sim}{A}^{(3)}$, i.e.,

$$\underset{\sim}{\tau} = \underset{\sim}{f}(\underset{\sim}{A}^{(1)}, \underset{\sim}{A}^{(2)}, \underset{\sim}{A}^{(3)}) \tag{1}$$

where $\underset{\sim}{\tau}$ is an extra stress tensor.

Since f is an isotropic tensor function, one may obtain explicit expression for f from the representation theorem of three tensors. By restricting the discussion to slow motion, one may represent the function by a first, second, third, and higher order approximation. In a later report, we will attempt to represent the function by the fourth-order approximation and to determine the material constants from measurement of flow in both extension and strip biaxial extensional flow.

In the present study, the steady-state non-Newtonian viscosities of uncrosslinked polyisobutylene were determined at various constant stretch histories. It was found that for slow strain rates (i.e., the region of Newtonian flow) the viscosity of strip biaxial extensional flow, η_{sb}, is equal to 4/3 the simple extensional viscosity, η_E; this result is consistent with the classical theory of Newtonian flow.

Experiment

The test setup is shown schematically in Figure 1. The main part of the setup is a Barber-Coleman Model 7401 "Chronotrol" (c), which is a program controller that uses the meter movement of a Model 401P millivolt input chassis (c). Programming is accomplished by changing the position of the millivoltmeter control set point by means of a slowly rotating disc or cam. The cam is rotated by a motor driven gear train, with the cam follower linked to the control set point through a cable drive. The Chronotrol provides a linear or nonlinear program control from any predetermined time-variable cycle. During any given cam cycle, an appropriate dc voltage is provided from the power supply (f) to the variable crosshead speed control accessory (b) of the Instron tester (a) through a servosystem. The servosystem consists of a reversible motor (d), a polarity reversing relay (g) and two ten-turn potentiometers. Using this setup, the crosshead speed is controlled by the shape of the cam.

Consider a rod-shaped sample of initial length ℓ_0 fixed at one end and extended in the direction of the principal extension. The velocity field v_i of the steady extensional flow is given by

$$v_i = \dot{\epsilon}_i x_i \qquad (i \text{ not summed}) \tag{2}$$

The following relationships for the displacement ℓ and the crosshead speed v can be derived:

$$\ell = \ell_0 e^{\dot{\epsilon}t} \text{ and } v = \dot{\epsilon}\ell_0 e^{\dot{\epsilon}t} \tag{3}$$

The correspondence of actual strain regime to that programmed depends on fluctuation of the motor speed, of the variable crosshead drive accessory, and on the accuracy of the programmed cam. Thus, during the run, the actual displacement of the crosshead as a function of time is continually
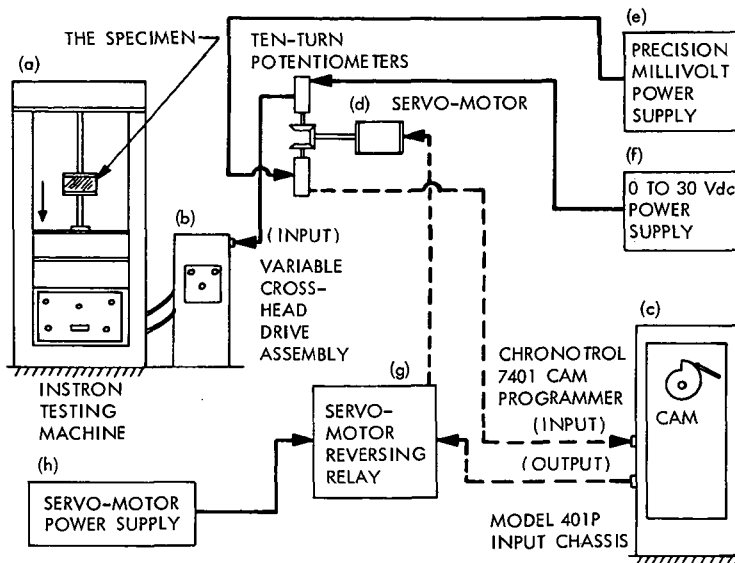
(a) INSTRON FLOOR MODEL TT-D TESTING MACHINE
(b) INSTRON MODEL A-35 VARIABLE SPEED CROSSHEAD DRIVE ACCESSORY
(c) BARBER-COLMAN·MODEL 7401 CHRONOTROL WITH A BARBER-COLMAN
    MODEL 401P MILLIVOLT INPUT CHASSIS
(d) dc SERVO-MOTOR, CONNECTED TO TWO BOURNS TEN-TURN POTENTIO-
    METERS BY A COMMON SHAFT
(e) ELECTRONIC DEVELOPMENT CORP., PRECISION MILLIVOLT POWER SUPPLY
(f) HEWLETT-PACKARD MODEL 721A POWER SUPPLY
(g) ADVANCE MODEL AM2C115 A.C. RELAY
(h) TRANPAC MODEL SR81 POWER SUPPLY

Figure 1. Schematic diagram of apparatus

compared to that programmed. If there exists any discrepancy, the position of the cam is carefully adjusted to bring them into agreement. The estimated maximum difference between the actual and programmed crosshead displacement is about 1%. Figure 2 illustrates the linearity between $\ln\lambda$ and time for several rates.

Results and Discussion

The material studied was uncrosslinked high molecular weight polyisobutylene (Vistanex L-80, Enjay Chemical Co.). All measurements were carried out at room temperature. Figure 3 shows the relationship of the true stress T_{11} to the stretch ratio λ for simple extensional flow at various strain rates while Figure 4 shows the corresponding behavior for strip biaxial flow. Since the stretch ratio at the constant strain rate $\dot{\epsilon}$ depends on time according to the relation $\lambda = e^{\dot{\epsilon}t}$, Figures 3 and 4 represent the relation of true stress vs time, and show the existence of a steady flow region (i.e., asymptotic region) for both simple and strip biaxial extension flow.
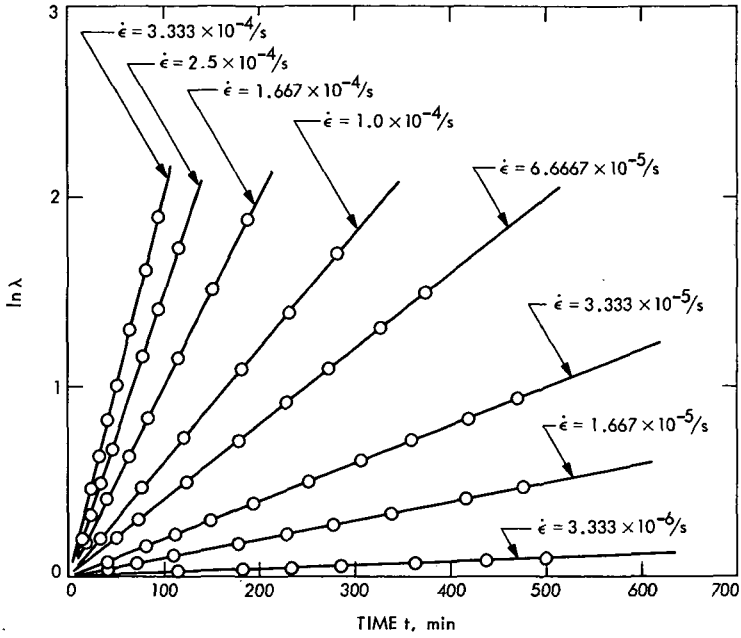
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Figure 2. Dependence of lnλ on time for several strain rates
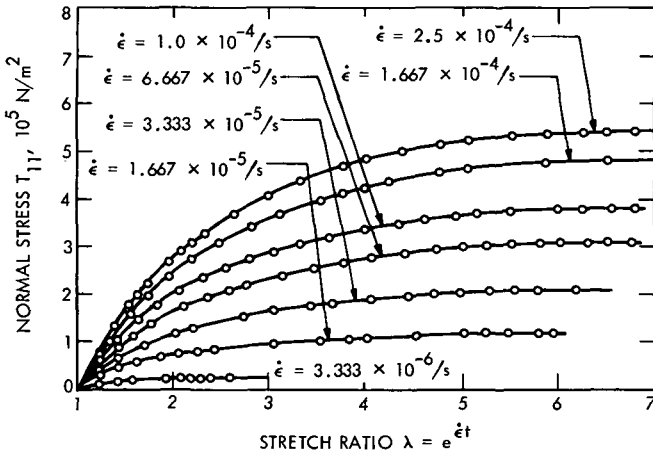

Figure 3. True stress vs total deformation in simple extension for various constant strain rates

The existence of steady flow for simple extension has been reported previously (Reference 2). But this is the first time that steady flow has been experimentally shown for strip biaxial flow.

Figure 5 shows the apparent non-Newtonian viscosity as a function of the normal stress at various strain rates for both simple extensional flow and
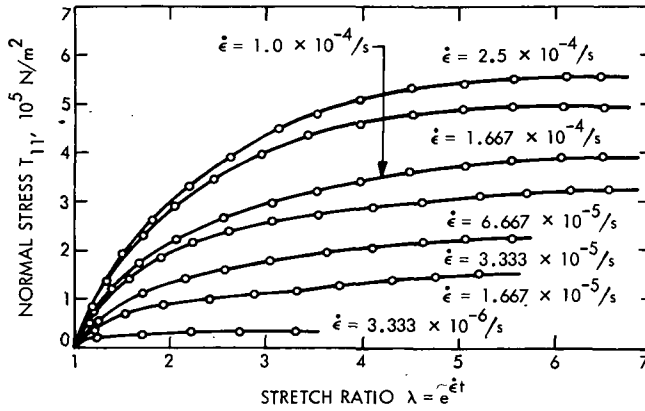
Figure 4. True stress vs total deformation in strip biaxial
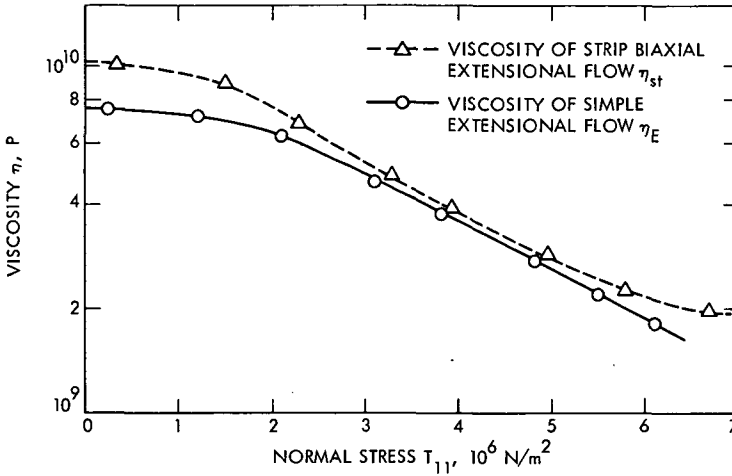extension for various constant strain rates

Figure 5. Dependence of simple extension viscosity η_E and strip biaxial
extension viscosity η_{st} on the normal stress T_{11}

strip biaxial extensional flow. It shows clearly that the viscosity decreases
with increase in the normal stress for both types of test. Figure 6 shows the
dependence of the non-Newtonian viscosity on the constant strain rate. It
may be seen that the viscosity is a monotonically decreasing function of the
strain rate. It is interesting to note that at higher strain rates, the difference
between the viscosity in simple extensional flow and in strip biaxial
extensional flow decreases.

For the transient region (i.e., the initial parts of the curves in Figures 3
and 4), however, it appears that in contrast to steady-state, the differences
between the simple and biaxial case increase at high strain rates. The reason
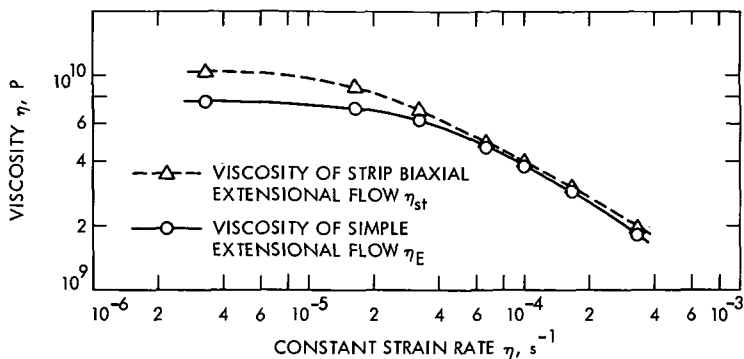for this behavior is not understood. Perhaps at high $\dot{\epsilon}$ elastic effects

Figure 6. Dependence of simple extension viscosity η_E and strip biaxial viscosity η_{st} on constant strain rate $\dot{\epsilon}$

predominate because of polymer network entanglements. The two modes of deformation for a nonlinear elastic regime have distinctly different characters as shown in studies on finite deformation of highly crosslinked rubberlike materials. On the other hand, in the region of steady flow, the material is liquidlike, and, as expected, exhibits the same character of rectilinear flow for both types of deformation.

In a later report a new model for elastic liquids will be developed to attempt to describe the transient state. This in conjunction with the fourth-order approximation to the steady-state region will aid in obtaining a total picture of the stress-strain-time relation for uncrosslinked and lightly crosslinked polymers for motion with constant stretch history.

Acknowledgment

References

1. Wang, C. C., *Arch. Ration. Mech. Anal.*, Vol. 20, No. 23, p. 329, 1965.

2. Ballman, R. L., *Rheologica Acta*, Vol. 4, p. 137, 1965.