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Manuscript received November 6, 1973; revision received February 15 and accepted February 22, 1974.

An Experimental Investigation of Viscous Heating in Some Simple Shear Flows

Theoretical investigations of viscous heating in the flow of fluids with an exponential dependence of viscosity on temperature have shown that, for a given shear stress, two shear rates are possible. Above a critical value, the stress decreases as the shear rate increases.

The present work is an experimental study of this phenomenon in plane and circular Couette flows and in cylindrical Poiseuille flow. Arochlor^R 1260, a high viscosity Newtonian fluid with an extremely sensitive viscosity-temperature dependence is used as the test fluid. The results clearly show that two shear rates for Couette flow exist for one measured wall shear stress. Because of the viscosity-pressure dependence of the fluid, the Poiseuille flow results are inconclusive.

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SCOPE

Rheologists have long been aware of the impossibility of maintaining viscous flow without some dissipation of mechanical energy into heat. For the past 50 years, investigators in many fields have attempted to estimate and measure the magnitude of the temperature rise due to frictional heating and the effect this increased temperature has on measured flow properties.

One of the most significant results of previous investigations is the existence of a maximum shear stress, below which, for a given stress, the shear rate assumes two values. At low fluid speeds, the stress increases because of the increased shear rate. As the speed increases, the vis-

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cosity decrease due to frictional heating becomes significant. Eventually, a speed is reached at which the stress increase due to the increased shear rate is more than compensated for by the stress decrease due to decreased viscosity. The shear stress then starts to decrease.

The object of this study is to present experimental evidence of the existence of a maximal shear stress in simple viscometric flow of a Newtonian fluid. The existence of this phenomenon has significance in virtually all polymer processing, not only in rheological measurements. The experiments were carried in three configurations, plane Couette flow, circular Couette flow, and circular Poiseuille flow using a Newtonian fluid exhibiting a very strong dependence of viscosity upon temperature, Arochlor 1260.

CONCLUSIONS AND SIGNIFICANCE

Although the phenomenon of maximal stress in flow with frictional heating has been proposed for several years, this is the first known study in which it is measured experimentally in viscometric flows. Plane Couette flow of the Arochlor 1260, a polychlorinated biphenyl, was conducted in the Porter-Manrique High Shear Viscometer. The flow did exhibit the maximal shear stress, but the data fell below the predicted curve. This discrepancy could be readily accounted for by only a slight eccentricity of the concentric cylinders. An equivalent experiment was conducted in a Epprecht viscometer. The results indicate excellent agreement with the predicted performance. Finally, the Arochlor 1260 was processed in an Instron Capillary Rheometer. Manifestation of the frictional heating phenomena was clearly apparent despite significant compressibility effects.

The principal significance of this effort lies in the verification of the maximal shear stress, but equally significant were the observations which reinforce the importance of accurate determination of the thermal behavior of viscometer boundaries.

Viscous heating in simple shear flow of a Newtonian liquid has been the subject of a large number of theoretical investigations. These studies have established beyond question the nature of the velocity and temperature profiles for these flows of a fluid with an exponential dependence of viscosity upon temperature.

However, the large majority of papers mentioned in the subsequent section contain no experimental data. Several investigators noted the existence of a maximum shear stress below which, for a given stress, the shear rate assumes two values. Although this behavior, sketched qualitatively in Figure 1, have been predicted, they have never been reported as having been measured in the laboratory.

In this work, the literature germane to viscous heating phenomena is reviewed and experimental evidence for the shear stress maximum is presented.

HISTORICAL ACCOUNT OF PREVIOUS WORK

The earliest investigators of this problem dealt with heat generation in lubricating films. Much of the earliest work is quite significant. Kingsbury (1933) suggested a graphical procedure for determining the temperature rise and shear stress in plane Couette flow with an arbitrary viscosity-temperature dependence. Duncan (1934) referenced an earlier work by Bratt (1927) in which the equations for the velocity and temperature profiles for a Newtonian fluid with an exponential dependence of viscosity on temperature were actually solved. This is a very surprising result, since many investigators attempted to solve this problem, but no complete solution was reported until the 1960's. Duncan himself outlined several methods of solution for the governing equation. However, he makes no mention of the double-valued shear rate found by later investigators. It is interesting to speculate whether or not Bratt realized this feature of the solution and why the publications committee to which he sent his work was unconvinced and rejected the paper. Unfortunately, Bratt's and Duncan's works seem to have been completely overlooked. Solutions to those equations, attempts at solutions, and solutions to much simpler problems have appeared many times since.

Although the viscosity of most fluids is extremely sensitive to temperature, many workers believed that any realistic model for this dependence would lead to insoluble equations. Hersey (1936) and Hersey and Zimmer (1937) developed approximate formulae for estimating heat effects in capillary flow. The latter work contained some experi-

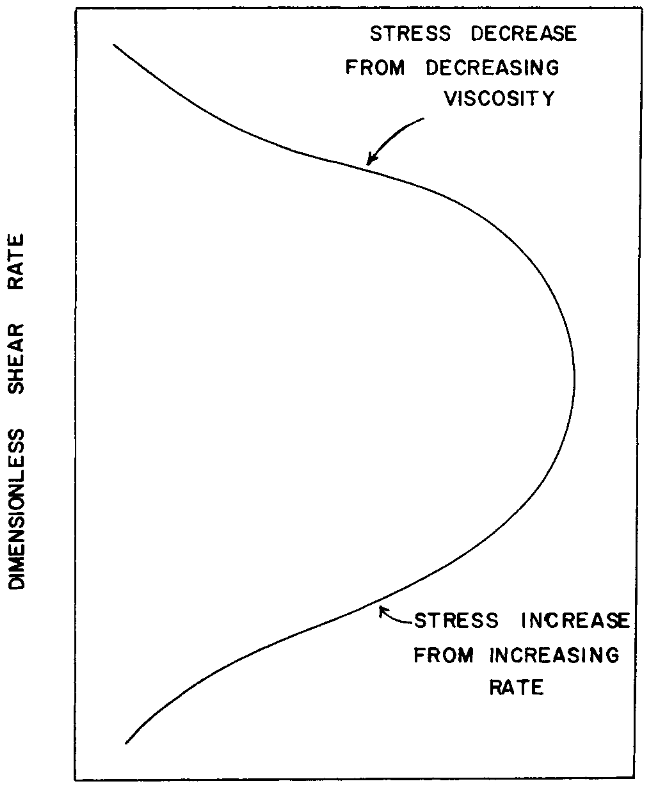

Fig. 1. The double-valued shear stress in flows with viscous heating.

mental data which agreed reasonably well with the theory.

Nahme (1940), Philippoff (1942), Hagg (1944), and Hausenblas (1950) attempted to determine the velocity and temperature profiles of a Newtonian fluid with temperature-dependent viscosity in Couette and Poiseuille flows. Hausenblas solved the equations for a hyperbolic dependence on temperature. Because of this assumption, he did predict a limiting value of the shear stress but could not observe its double-valued character. Nahme, Philippoff, and Hagg assumed an exponential temperature dependence. Nahme's solution is incomplete, for he failed to realize that two velocity and temperature profiles are possible for a given shear stress. Philippoff, believing the governing equations for Poiseuille flow to be insoluble, presented approximate solutions. Only Hagg, whose work, like Bratt's dealt with heat effects in lubricating films, predicted that a maximum shear stress exists and that below this value, the stress is double-valued in the shear rate. He presents a plot of a dimensionless shear rate, now called the Brinkman number, *vs.* a dimensionless shear stress, which clearly shows the double-valued character of the solution. Hagg performed some experiments on two low viscosity lubricating oils, but the shear rates were too low to observe the turnaround in stress. Hagg's work has been completely overlooked in the American literature, but unlike Bratt and Duncan, many Russian investigators were aware of his contribution.

An attempt to estimate the temperature rise in Couette flow with an arbitrary viscosity-temperature relationship was made by Weltmann and Kuhns (1952). Their work, which contains many errors, involved a graphical solution using the method of successive approximations.

One of the most significant advances came with the work of Brinkman (1951). He presented rigorous solutions for the temperature profile in capillary flow taking into account heat conduction, convection, and generation by dissipation. Bird (1955) extended Brinkman's solution to the case of non-Newtonian fluids. Toor (1956, 1957) investigated compressible fluids in capillary flow. These works are all based on a temperature-independent viscosity. The presence of the convection terms in the energy equation makes similar calculations with a temperature-dependent viscosity intractable when done by hand. Gee and Lyon (1957) attempted one of the first computer solutions for the velocity and temperature profiles of a non-Newtonian compressible fluid with temperature-dependent physical properties in tube flow.

Gorazdovskii and Regirer (1956) investigated the Couette flow problem with a hyperbolic dependence for viscosity. They concluded that a maximum shear stress exists, as did Regirer (1958) in his work on plane Couette flow with an arbitrary viscosity-temperature relationship. They did not, however, realize that for a given wall shear stress, two shear rates are possible.

This double-valued shear rate-shear stress behavior was predicted by Bolen and Colwell (1958) and McKelvey (1963, pp. 326-328) in their analyses of dispersive mixing in fluids with an exponential viscosity-temperature dependence. Such a result is important in this area of polymer processing since the magnitude of the shear stress developed is viewed as determining the quality of the resulting mixture. These investigations provided theoretical support for earlier experimental work by Jones and Snyder (1951). These workers had performed mixing experiments on a Banbury mixer and found that as they increased the rotor speed at constant temperature the quality of mixing increased, passed through a maximum, and then decreased.

Ball and Colwell (1960) arrived at a similar conclusion in their computer study of the plane Poiseuille flow of a polymer melt. These are probably the first authors, with the exception of Hagg, and possibly Bratt, to predict that the stress decreased with increasing shear rate above a critical value.

Since that time, the literature on viscous heating has grown enormously, although not every investigator seemed to realize that two velocity and temperature profiles may exist at a given shear stress. Kearsley (1962) solved the fully-developed tube flow problem with an exponential dependence of viscosity upon temperature and recognized this distinguishing feature. Kaganov (1963), however, investigated the same problem for an arbitrary viscosity-temperature relationship and concluded a limiting pressure gradient exists, making no mention of the double-valued solutions. Aslanov (1963) examined Couette flow, but failed to present a complete solution. Later, Kaganov (1965) recognized there should be two solutions in Couette flow, but he did not determine them.

In a series of papers, Gruntfest (1963, 1965a, 1965b), Gruntfest, Young and Johnson (1964) and Gruntfest and Becker (1965) contended that thermal feed-back contributed in large measure to the shear-thinning properties of fluids. They solved the equations for time-dependent Couette and Poiseuille flows of a Newtonian fluid with an exponential dependence of viscosity on temperature on an analog computer. These solutions do show the double-valued behavior. Gruntfest (1965a) attributes the solution of the equation for the steady plane flow problem to Copple, Hartree, Porter, and Tyson (1939), who solved it in their study of the electrical strength of dielectrics. Brodnyan (1966) refuted many of Gruntfest's contentions, especially his implication that non-Newtonian flow behavior was largely an experimental artifact. Brodnyan did agree that viscous heating could cause serious errors in viscometry.

Bird and Turian (1962) were the first to investigate viscous heating in a cone and plate viscometer. Assuming constant viscosity and a small cone angle, they obtained an approximate solution to the resulting partial differential equation using variational techniques. Turian and Bird (1963) solved the plane Couette flow problem with both a series approximation for the temperature dependence and an exponential viscosity-temperature dependence. They did not, however, seem to realize that in the latter case two solutions exist for one value of the wall shear stress. They present some data obtained on a cone and plate instrument which agree well with their theory at low Brinkman numbers. The shear rates were too low to observe the stress maximum experimentally. Turian (1965, 1969) extended their analysis to power-law and Ellis fluids in plane Couette flow. In the first paper, he used the series approximation for temperature dependent thermal conductivity and viscosity. An exponential dependence on temperature for the zero-shear viscosity was employed in the second paper. Turian obtained an analytical solution to the equations and found that two solutions are possible for a given shear stress. However, he believed that only one solution would be observed physically.

An approximate method of accounting for viscous heating in capillary flow with an adiabatic wall, based on neglecting the radial velocity components, was developed by Gerrard and Philippoff (1965). Their data, taken for a 12 poise oil in a pressure-driven rheometer, agreed quite well with the theory. These authors were perhaps the first to point out that the adiabatic boundary condition is probably much better than the isothermal condition for most capillary rheometers.

Joseph (1964, 1965) solved the equations of conservation of momentum and energy for a variety of viscosity-temperature relationships in Couette and Poiseuille flows. He reviewed much of the previous work and clarified many incorrect conclusions regarding the stability of the flow. He realized that a maximum stress exists but apparently believed that solutions corresponding to shear rates beyond the stress maximum were somehow not readily realizable physically.

The Couette flows of a Newtonian fluid with an exponential viscosity-temperature relationship were treated by Bostandzhyan, Merzhanov, and Khudyaev (1965). A Bingham plastic fluid in the same flow system was studied by Ibragimov (1965). Both investigations indicated that two solutions are possible for a given applied shear stress.

The capillary flow problem was once again studied by Gerrard (1965, 1966), this time in conjunction with Steidler and Appeldorn. They present computer solutions along with experimental data for both the adiabatic and isothermal wall problems. The fluid they used also showed a viscosity-pressure dependence, which they accounted for in their numerical solution.

Martin (1967) investigated the plane and circular Couette flow and the Poiseuille flow of a power-law fluid with an exponential dependence of viscosity on temperature. Martin presents complete solutions for all three flow situations and for both adiabatic and isothermal boundary conditions in the Couette flows. His plots of a parameter which is the square root of the Brinkman number vs. dimensionless shear stress clearly show that two different shear rates can result in the same stress.

Powell and Middleman (1968) examined the transient thermal response of the bob in Couette flow. The bob is assumed to be hollow and of finite mass and thermal capacity. They include the effects of viscous heating but assume that the viscosity is constant. Their results show when the shear rate-shear stress data should be taken so that it can be temperature corrected using either the isothermal or the adiabatic boundary condition on the temperature field.

The work of Gavis and Laurence (1968a, b) on the Couette flows of both Newtonian and power-law fluids also show the double-valued shear stress. Gavis and Laurence present Brinkman number-shear stress curves, as well as the velocity and temperature profiles for sample values of the shear stress. These authors investigated both the isothermal and adiabatic boundary conditions. Nihoul (1970) showed how the procedure used by Gavis and Laurence to obtain their solutions could be extended to include a temperature dependent thermal conductivity as well as viscosity. He also stated that the velocity and temperature profiles were unique in the Brinkman number. Fixing this parameter, one could then determine a single velocity and temperature profile as well as shear stress.

Sukanek (1971) investigated the Poiseuille flow problem for power-law fluids and showed how the velocity and temperature profiles could be expressed uniquely in terms of the Brinkman number.

Nihoul (1971) examined the same problem and concluded, as Joseph did, that the portion of the shear stress curve above the maximum has no physical meaning. This contention is refuted by Sukanek and Laurence (1972) who argue that such a conclusion is only possible for pressure-driven rheometers.

The effects of viscous heating in lubrication have been recently examined by Crook (1961), Dyson (1970), Kannel and Walowit (1970), and Fein (1971). Crook reported the results of experiments which showed that one frictional traction (shear stress) is obtained at two different values of the sliding speed (shear rate). Crook attempted to explain these results on the basis of frictional heating. Dyson reported similar experimental results found by several previous workers. An analysis of the effect of boundaries of finite thermal resistance was presented by Fein. Kannel and Walowit proposed a simplified method of accounting for temperature- and pressure-dependent viscosity. Their theoretical results are very close to the experimental curves of Crook.

Gould (1971) has examined the plane Poiseuille flow problem with pressure- and temperature-dependent viscosity. His analysis of the problem is good, although his discussion of instability is misleading since his definition of stability is unclear.

One of the most recent papers on the subject is that of Lebon and Mathieu (1972), which again treats the plane Couette flow problem for a non-Newtonian fluid and exponential viscosity behavior. This work is more an exercise in irreversible thermodynamics and variational calculus than an explication of viscous heating phenomena, but its methods may be useful in the analysis of more complex flow situations than those discussed in this review.

EXPERIMENTS

Test Fluid

One of the major drawbacks of the fluids used in the few experiments on viscous heating that have been reported is that their viscosities were too low to obtain sufficiently large Brinkman numbers. The Brinkman number, as defined in this study, is

$$Br = \frac{\beta \mu_0 V^2}{k T_0} \qquad (1)$$

the parameter,

$$\mu = \mu_0 \exp \{-\beta(T - T_0)/T_0\} \qquad (2)$$

Since in all the reported experiments the fluids had a viscosity of less than 20 poise, it would take a velocity of at least 300 cm/s to produce a Brinkman number of approximately 10. This is based on a temperature of about 300°K, a β value of 50 (very temperature sensitive) and a thermal conductivity of 4×10^4 g cm/s^3 (°K), a reasonable value for many liquids.

One relatively easy way of increasing the Brinkman number, without using exceedingly high velocities, is to choose a fluid with a high viscosity. The Arochlor^R series of chlorinated biphenyls manufactured by the Monsanto Company, have all of the desired properties and offer a wide range of viscosities. One in particular, Arochlor^R 1260 (hereafter called simply 1260), is an excellent fluid to use in the study of viscous heating phenomena. It has a very high viscosity, is extremely temperature sensitive, and behaves as a Newtonian fluid over a very broad shear rate range.

Two samples of 1260, obtained at different times from the Monsanto Company, were used in this study. The viscosities were measured on an Epprecht Couette viscometer over a temperature range of 20° to 45°C. A Haake constant temperature bath was used to set and maintain a constant wall temperature. Figure 2 is a plot of the viscosity of 1260, in poise, vs. reciprocal absolute temperature. It shows the extreme temperature sensitivity of the viscosity. At 20°C, the viscosity is greater than 10^5 poise and falls to about 400 poise a 40°C. For every 2°C temperature rise, the viscosity drops by a factor of about 2. Because of the extreme temperature dependence, Equation (2) is valid for the 1260 over only about a 10°C temperature range. The density, heat capacity, and thermal conductivity of the 1260 are:

$C_p = 0.235$ cal/g °C (Monsanto Company)

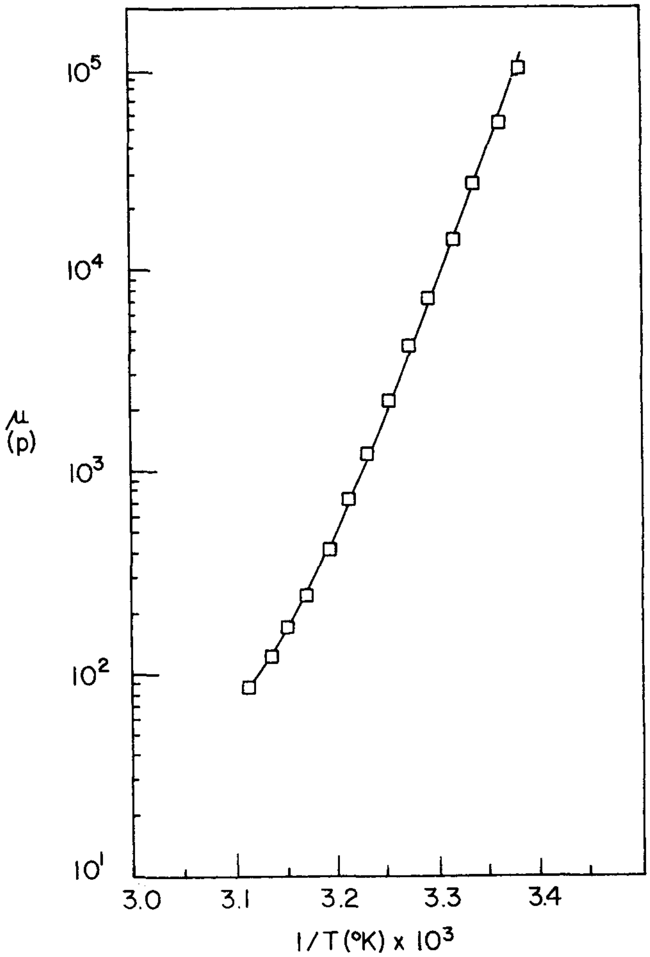

Fig. 2. Viscosity of Arochlor 1260^R vs. reciprocal absolute temperature.

ρ = 1.6 g/cm^3 (Monsanto Company)
k = 9.4 × 10^3 g cm/s^3 (°K) (Kruse, 1972)

These values vary only slightly with temperature.

Plane Couette Flow

The first flow situation in which the maximum shear stress has been measured is plane Couette flow. The High Shear Couette Viscometer described by Manrique and Porter (1972) was used in this study. Several inner cylinders are available giving gaps between the two cylinders from 5 × 10^{-5} cm to 1.27 × 10^{-2} cm. Shear rates above 10^6 s^{-1} can be obtained. Because of the very small gaps, the flow closely approximates plane Couette flow.

The system is similar in design and layout to that described by Barber, Muenger, and Villforth (1955). The details of the experimental device may be found in Manrique (1972). The outer cylinder and retaining cup assembly is mounted on ball bearings. The shear force is measured by the torque required to restrain this assembly from rotating. The test material is supplied through a groove around the circumference of the other cylinder. Thermocouple wells in the outer cylinder permit the temperature profile in the cylinder to be measured. The fluid temperature at the wall of the outer cylinder can be found by extrapolation. A 1 hp. electric motor with a variable speed controller drives the inner cylinder through a long flexible drive rod. The outer cylinder is surrounded by a constant temperature bath and so may be assumed isothermal. There is no accurate way to thermostat the inner cylinder. However, because it is hollow, it might be assumed to act as an adiabatic boundary.

Gavis and Laurence (1968a) present their solutions in terms of a double-valued constant. However, for plane Couette flow, the solution can be given explicitly in terms of the Brinkman number. The relation between the dimensionless shear stress λ and the Brinkman number is, for an adiabatic inner wall and isothermal outer wall

$$\lambda = \frac{4}{2 + Br}\left[\sinh^{-1}\frac{Br^{1/2}}{2}\right] \tag{3}$$

The definition of λ is

$$\lambda = \frac{\beta h^2 \tau_\omega^2}{\mu_0 k T_0} \tag{4}$$

The widest gap, 1.27 × 10^{-2} cm, was used in the present study.

From Equation (3), the maximum value of λ is approximately 0.88 and occurs at Br = 4.55. The Br vs. λ curve from Equation (3) is shown by the solid line of Figure 3.

Samples of the 1260 were run on the high shear viscometer in an attempt to measure this double-valued shear stress. The characteristic velocity used in the definition of the Brinkman number is, in this case:

$$V = \Omega_i R_i \tag{5}$$

The 1260 was heated with a hot air gun and poured into a syringe which was connected to the groove in the interior of the outer cylinder. With the inner cylinder positioned, the 1260 was forced into the gap by applying pressure to the syringe plunger. The temperature was maintained with a silicon oil constant temperature bath. When the outer wall came to thermal equilibrium as indicated by constant temperature readings from the thermocouples, the inner cylinder was placed in motion at a constant rotational speed. The torque reading started at a high value, and as the temperature profile in the fluid developed, it decreased. The steady torque value was recorded and a new rotational speed selected.

A complete listing of the data can be found in Sukanek (1972). The experimental data are plotted in Figure 3.

The measured data do not agree with the theory. In all cases, the experimentally determined shear stress falls well below the theoretical value for a Newtonian fluid in plane Couette flow with an isothermal outer wall and an adiabatic inner wall. However, the data clearly show the shear stress to be double-valued in the Brinkman number. The experimental Brinkman number at the maximum shear stress is approximately 3.7, very close to the predicted value.

Although the experimentally determined values of λ are much lower than those predicted theoretically, it should be remembered that λ is defined in terms of the square of the shear stress. A small error in τ_ω will produce a large error in λ.

Several explanations can be offered for the discrepancy between theory and experiment. Although the connecting tubing between the syringe and the outer cylinder and interior of the apparatus was heated with a hot air gun during the gap-filling operation, it is possible that some regions of the interior were not heated sufficiently. If this were true the 1260, because of its extremely high viscosity at low temperatures, would not completely fill the gap between the two cylinders. A partially filled gap would result in shear stresses lower than anticipated.

Another explanation for the low shear stresses is based on the self-centering nature of the inner cylinder. At the high viscosity and shear stresses used in these experiments, the inner cylinder might not be properly centered. Pinkus and Sternlicht (1961, pp. 41-48) have performed an approximate analysis of flow in a journal bearing, which is the same as Couette flow between eccentric cylinders. The ratio of the torque on the outer cylinder for eccentric cylinders T to that for concentric cylinders T_o is

$$\frac{T}{T_o} = \frac{2(1-\epsilon^2)^{1/2}}{2+\epsilon^2} \tag{6}$$

Equation (6) shows that the torque measured at the outer cylinder is always less for eccentric cylinder than for concentric ones. An eccentric inner cylinder would not only lower the torque but would also affect the heat generation and produce temperature variations around the cylinder.

It was noted above that, because of the extreme temperature sensitivity of the viscosity of 1260, Equation (2) is strictly valid over only a 10°C temperature range. Figure 4 is a plot of the theoretical maximum temperature rise vs. Brinkman number in plane Couette flow when the inner wall is adiabatic. As seen in Figure 3, the highest Brinkman number attained in this study was less than 20. This gives a temperature rise of about 9°C. No serious errors were introduced into the analysis by assuming β to be constant for each experimental run.

Circular Couette Flow

The viscous heating experiments in circular Couette flow were performed on an Epprecht Couette viscometer. A series of cups and bobs are provided with the instrument. The particular measuring system that is chosen depends on the approximate viscosity of the fluid. Measuring system MS-E was used in the experiments reported here.

The test fluid is placed in the cup, and the bob, a small cylinder with conically tapered ends, is inserted and attached to the body of the viscometer. An electric motor rotates the bob at one of 15 possible rotational speeds. The torque acting on the bob is measured by the deflection of a lever-arm. The torque reading is converted to a shear stress by an appropriate conversion factor depending on the geometry of the system and the mechanics of the torque measuring system.

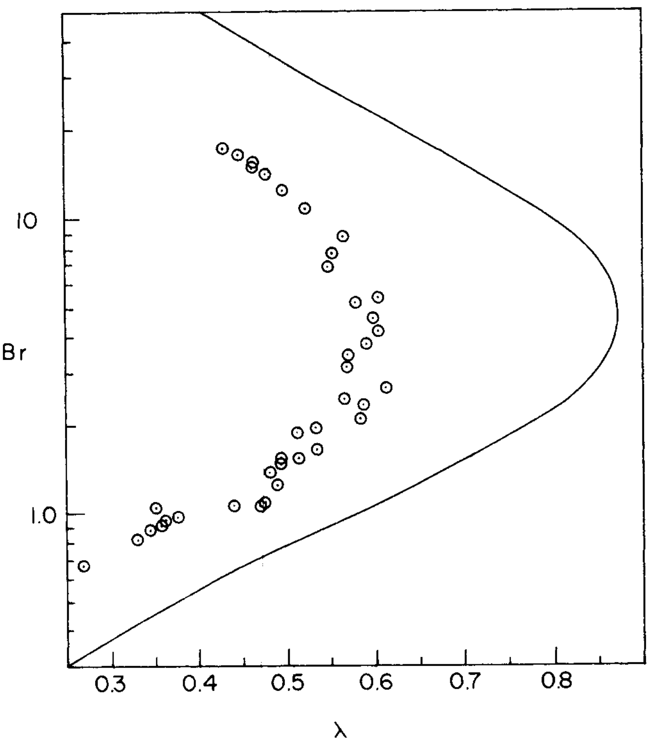

Fig. 3. *Br* vs. λ in plane Couette flow. Theoretical curve and experimental values.

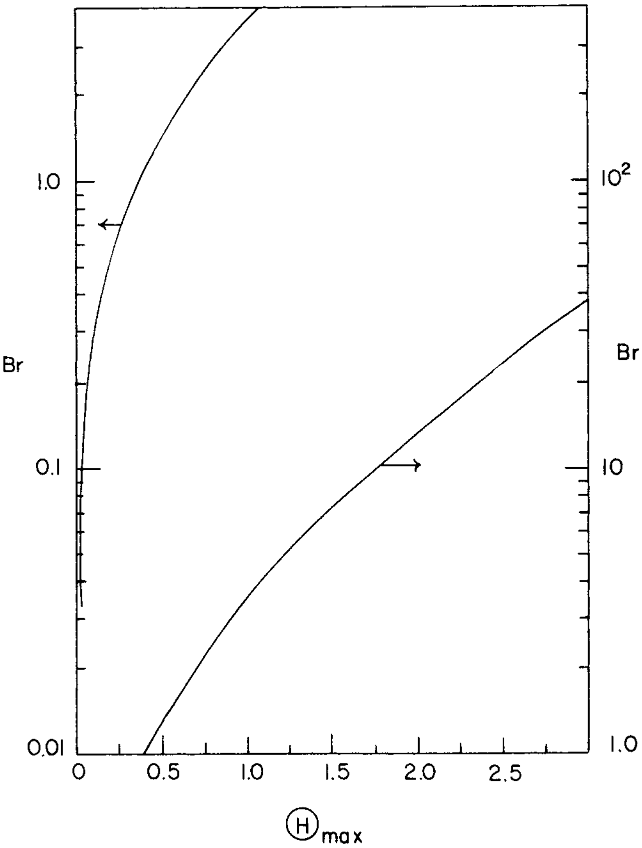

Fig. 4. Theoretical maximum temperature rise in plane Couette flow. Inner wall abiabatic. $\theta = \beta\,(T - T_o)/T_o$.

The outer cylinder could be held at a constant temperature using the Haake constant temperature bath. The bob was first assumed to act as an adiabatic boundary. For circular flow with these boundary conditions, a relationship similar to Equation (3) can be developed from the work of Gavis and Laurence (1968a). Because of the circular geometry, it is not as simple as Equation (3)

$$\begin{aligned} Br &= \frac{2}{\alpha^2\kappa^2}(\alpha^2-1)\sinh^2(\alpha \ln \kappa) \\ \alpha^2 &= q\lambda/2 \\ q &= \frac{[\alpha\cosh(\alpha\ln\kappa)+\sinh(\alpha\ln\kappa)]^2}{(\alpha^2-1)\kappa^2} \end{aligned} \tag{7}$$

The Brinkman number is defined as before, Equation (1). The shear stress parameter λ is given by

$$\lambda = \frac{\beta R^2{}_i \tau_\omega{}^2}{\mu k T_0} \tag{8}$$

The quantities α and q may be viewed simply as parameters, and κ is the ratio of the radii of the outer to inner cylinders. In the present experiments, the value of κ was 3.755.

Figure 5 shows the viscosity-shear rate behavior of 1260 on the Epprecht viscometer. The dashed lines, for three different temperatures, show the fluid to be Newtonian. These data were taken before the temperature profile could develop, and so represent the true shear dependence.

The solid lines of Figure 5 are the apparent viscosity-shear rate data taken after the temperature profiles were fully developed. When this thermal equilibrium was achieved, the shear stress was constant in time. The apparent highly non-Newtonian behavior indicated by these curves is due solely to viscous dissipation.

The data are plotted in the form Br vs. λ in Figure 6. These clearly show that above a certain critical Brinkman number, the shear stress decreases with increasing shear rate. As in the plane flow case, the observed data do not fall on the theoretical curve for an adiabatic inner wall. This behavior, however, can be readily explained.

The bob of the Epprecht viscometer is solid and small in size. There is no insulation of any kind on the rod connecting the bob to the motor. Therefore, it is likely that all the heat generated in the fluid cannot accumulate in the bob; it must be conducted away, up the connecting shaft. The inner boundary is neither adiabatic, nor isothermal. This contention is supported by the location of the experimental data. They fall between the theoretical curves for the adiabatic and isothermal boundary conditions.

A theoretical investigation of this problem was presented by Sukanek (1972). The appropriate boundary condition on the temperature field at the bob is, in dimensionless form:

$$\frac{d\theta}{dn} + Nu\,\theta = Nu\,\theta^* \tag{9}$$

The notation $d\theta/dn$ denotes the component of the temperature gradient normal to the surface. Two adjustable parameters, Nu and θ^*, are introduced. Nu is a Nusselt number, a measure of how much of the heat transferred to the bob is conducted away. When $Nu = 0$, no heat leaves the bob; it is adiabatic. When $Nu \to \infty$, all the heat is conducted away, and the bob stays at a constant temperature. In this case, the temperature θ^* is the dimensionless bob temperature.

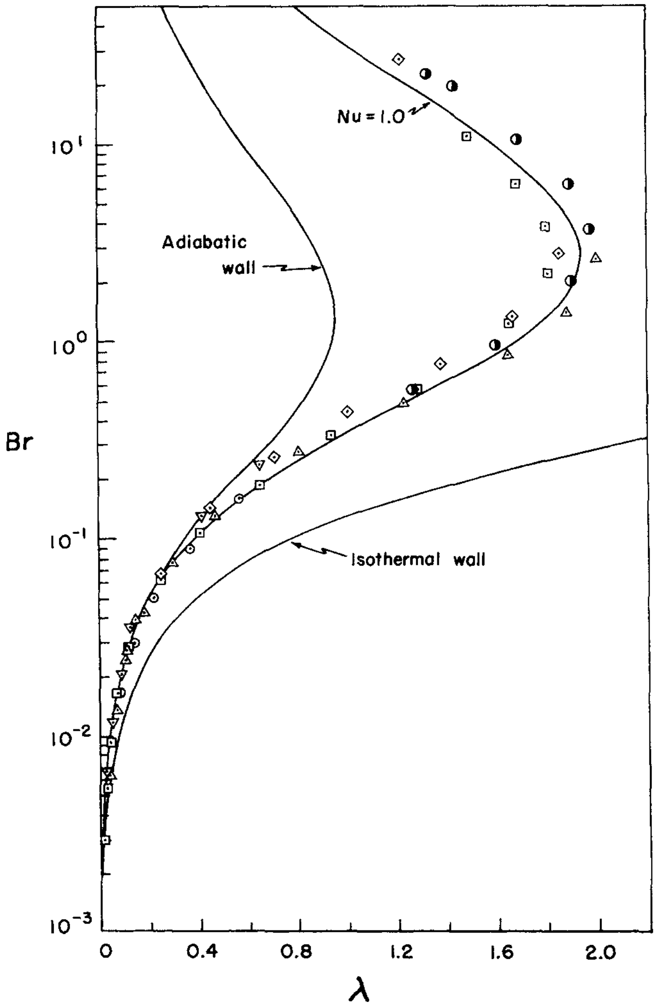

Fig. 6. Br vs. λ in circular Couette flow. Theoretical curves and experimental values. $K = 3.755$. The temperatures are: ○, 298.2°K; ◇, 300.8°K; ◑, 300.95°K; ◑, 301.35°K; ◐, 301.75°K; □, 303.15°K; △, 308.0°K.

The analysis of Sukanek (1972) predicts the curve shown in Figure 6 for a Nusselt number of unity and a θ^* of 0. This curve fits the data exceptionally well. The small deviations of the experimental points from this curve might be due to small variations in θ^*.

Figure 7 is a plot of the theoretical temperature rise for the adiabatic wall vs. the Brinkman number. The completely adiabatic inner wall will give the largest temperature rise and so will present the strongest test for the applicability of Equation (2). The largest temperature rise in these experiments predicted in this manner is less than 12°C. Therefore, the assumption that β is a constant for each of the test temperatures is valid.

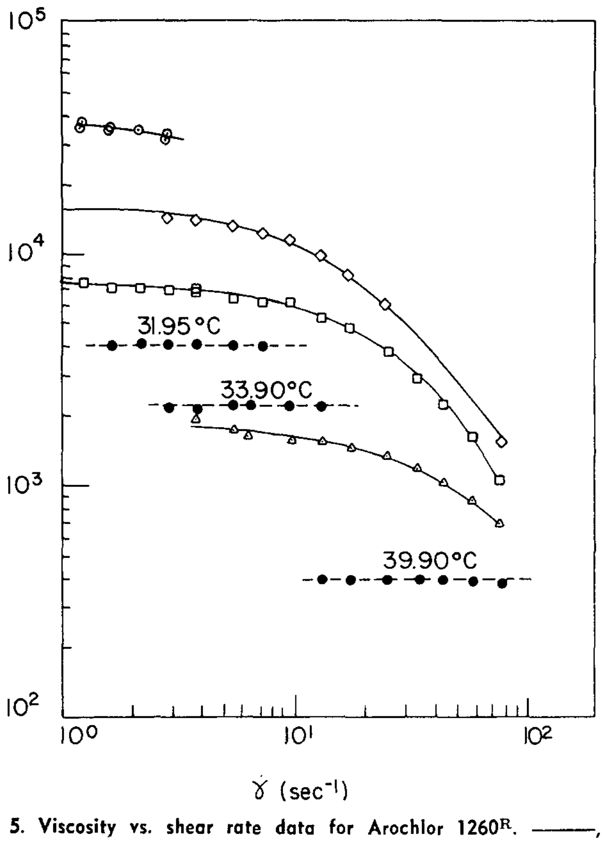

Fig. 5. Viscosity vs. shear rate data for Arochlor 1260R. ———, apparent viscosity; - - - -, actual viscosity. The temperatures are: ⊙, 25.05°C; ◇, 27.65°C; □, 30.05°C; △, 35.05°C; ● actual viscosity data.

Cylindrical Poiseuille Flow

It is generally accepted that in Couette flows for a given shear stress two flows are possible. This has been shown experimentally in the previous two sections. However, there is some controversy over the Poiseuille flow problem. Whereas Nihoul (1971) contends that the upper branch of the shear stress curve is physically meaningless, Sukanek and Laurence (1972) maintain that the results should depend upon the experimental system. A pressure-driven capillary, for example, would produce only the lower half of the curve. An instrument in which the average velocity can be fixed should produce the entire curve.

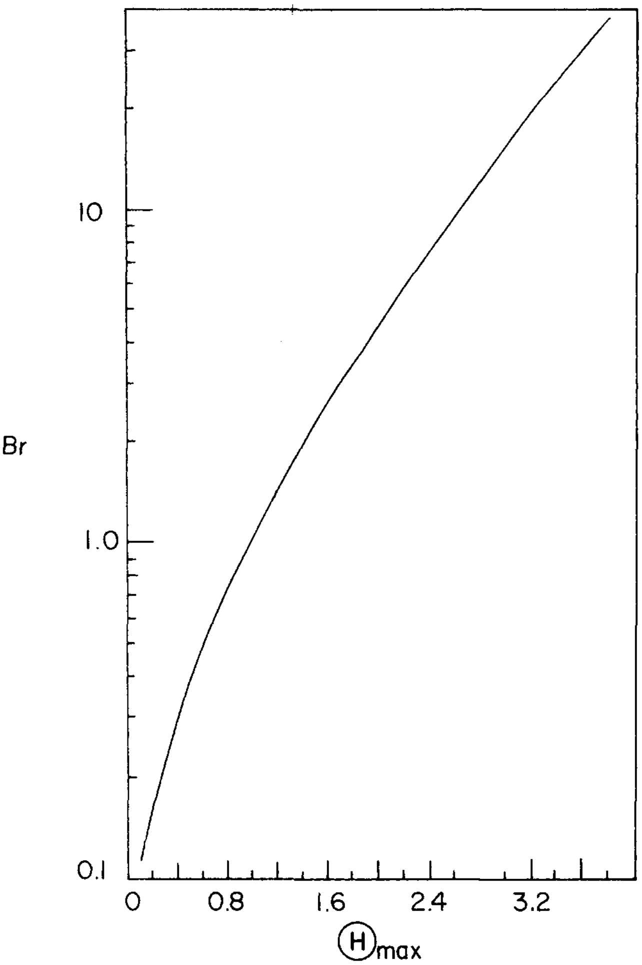

Fig. 7. Theoretical maximum temperature rise in circular Couette flow. Inner wall abiabatic. $K = 3.755$. $\theta = \beta\,(T - T_o)/T_o$.

In this section, experiments performed with the 1260 on a Instron capillary rheometer will be discussed. Unlike the pressure-driven instruments of Gerrard and Philippoff and Gerrard, Steidler, and Appeldorn, the Instron forces the fluid through the capillary at a constant, pre-set, average velocity.

Two capillaries were used, both with an L/D of approximately 100. Capillary number 1 had a diameter of 0.07665 cm; capillary number 2, 0.0508 cm.

The results of the Instron experiments are shown in Figures 8 and 9, where the apparent viscosity is plotted against the wall shear rate. It can be seen that another factor, not taken into account by the viscous heating analysis of Sukanek and Laurence, is important. The viscosity of 1260 is pressure- as well as temperature-dependent. No data, however, are currently available to show the quantitative dependence of the viscosity of Arochlor 1260 on pressure.

This pressure-dependence of viscosity has been examined by Penwell and Porter (1969) and Penwell, Porter, and Middleman (1971) for polystyrene. The results of these investigations also showed that as the shear rate increased so did the apparent viscosity. Penwell and Porter ascribe this phenomenon to a decrease in the free volume which causes a shift in the glass transition temperature. The shift in T_g can then account for the increase in apparent viscosity.

Figures 8 and 9 show that the capillary diameter has a strong effect on the apparent viscosity, especially at the lower test temperatures. At low shear rates, where the pressure-dependence is important, the smaller capillary showed a much lower apparent viscosity.

As the shear rate increase, viscous dissipation becomes more and more important. At low wall temperatures, the fluid is so pressure-sensitive that even moderately high shear rates cannot be reached. As the wall temperature increases, the pressure dependence softens. At these wall temperatures a shear rate can be reached where the heat

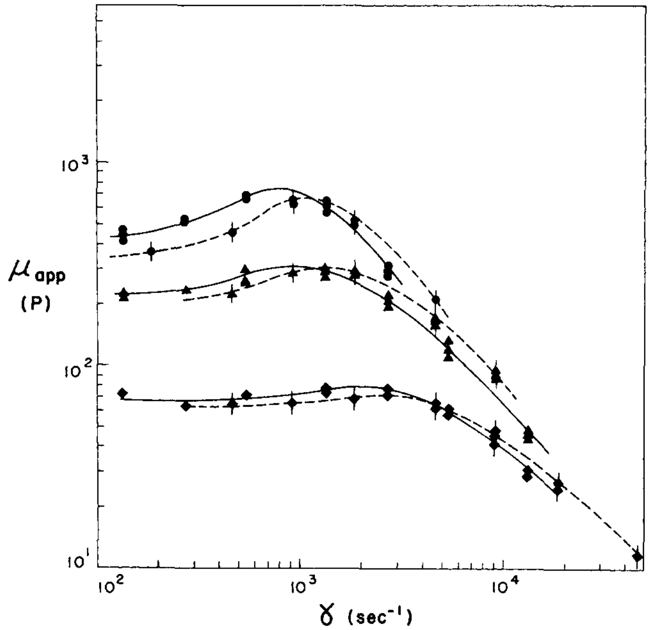

Fig. 8. Apparent viscosity vs. shear rate data for Arochlor 1260R in capillary flow. Low temperatures. ———, capillary no. 1; - - - -, capillary no. 2. The temperatures are: ○, 33.5°C; □, 36.5°C; △, 38.5°C; ▽, 39.5°C. Symbols with | refer to capillary no. 2.

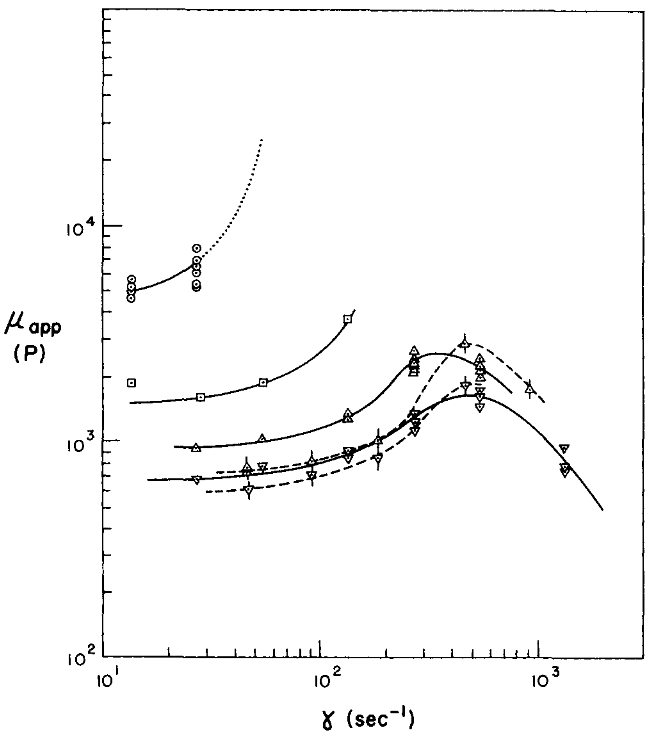

Fig. 9. Apparent viscosity vs. shear rate data for Arochlor 1260R in capillary flow. High temperatures. ———, capillary no. 1; - - - -, capillary no. 2. The temperatures are: ●, 41.5°C; ▲, 43.5°C; ◆, 48.5°C. Symbols with | refer to capillary no. 2.

generated in the fluid more than compensates for the viscosity rise due to pressure. The apparent viscosity then begins to decrease.

At these higher shear rates, where viscous heating is predominant, the smaller capillary gives a somewhat higher apparent viscosity. It is relatively easy to show that this type of diameter dependence is predicted by the viscous heating theory of Sukanek and Laurence (1972)*.

The Brinkman number-shear stress data for the capillary experiments are shown in Figure 10. The Brinkman number is defined in terms of the average velocity through the capillary. The shear stress parameter λ is defined as

$$\lambda = \frac{\beta R^2 \tau_\omega^2}{\mu_0 k T_0} \tag{10}$$

where R is the capillary radius.

The data, despite their variability, do show a definite trend. There is definitely a limiting shear stress, $\lambda = 70$. However, the data do not show the predicted double-valued behavior clearly. At only two temperatures, 41.5°C and 43.5°C, did the highest Brinkman number point fall at a λ below the previous one.

The scatter in the data can be accounted for in at least two ways. First, in making a plot such as Figure 10, no account is taken of the pressure dependence. Introducing another temperature-dependent parameter characteristic of the pressure variation of viscosity would probably bring the points for the different temperatures closer together.

A second reason for variations in the data of Figure 10 for the different temperatures is that it is quite possible that the thermal boundary conditions are different for each test temperature. The isothermal wall boundary condition is probably not a good thermal approximation of the Instron wall. As Gerrard and Philippoff (1965) and Gerrard, Steidler, and Appledorn (1966) found in their investigations of capillary flow, there is no doubt a temperature profile in the rheometer wall. The Instron wall is kept at constant temperature by supplying a heat flux to the wall at a specified rate and time as prescribed by a temperature controller. For each test temperature, this rate and time would be different. A boundary condition similar to Equation (9) is necessary to describe the system, but with a different value of Nu for each test temperature. This would give a different theoretical curve for each temperature.

Brinkman numbers higher than those shown in Figure 10 could not be attained for several reasons. At the low temperatures, the pressure effect was too great to reach high shear rates. At the highest temperatures, the viscosity was too low. At intermediate temperatures, when the flow rate through the capillary was increased beyond the highest value reported here, rapid oscillations in the shear stress resulted. Three examples of the phenomenon are shown in Figure 11 for temperatures of 36.5°, 38.5°, and 41.5°C. These curves represent the force variations with time. The abscissa of these curves is the force measurement. The ordinate, actually a chart distance, has been converted to time. Each run was terminated automatically when the plunger reached a certain length into the rheometer barrel. While the recorder pen behaved in this manner, the output of the capillary appeared as pulses rather than in the smooth stream characteristic of the lower throughputs. As the force increased, a small drop of 1260 appeared to accumulate at the exit. Then, as the force decreased, the drop fell and a new one began to grow. At no time, however, did the flow cease entirely. While these drops grew, the flow rate did appear to slow down considerably.

The cause of this behavior is undetermined. However, it seems likely that it is due to instability of the flow. A physical explanation is based on the large temperature variations in the fluid at high Brinkman numbers. If the temperature at the capillary center were extremely high, a small fluctuation in temperature, pressure, or velocity might cause the fluid at the center to appear to slip past the cooler, more viscous fluid near the wall. This would result in a spurt of fluid at the capillary exit, with a corresponding stress decrease. After this spurt left the capillary, the stress would begin to rise to its previous value, and the whole process would be repeated.

Table 1 lists the conditions where this apparent instability was observed.

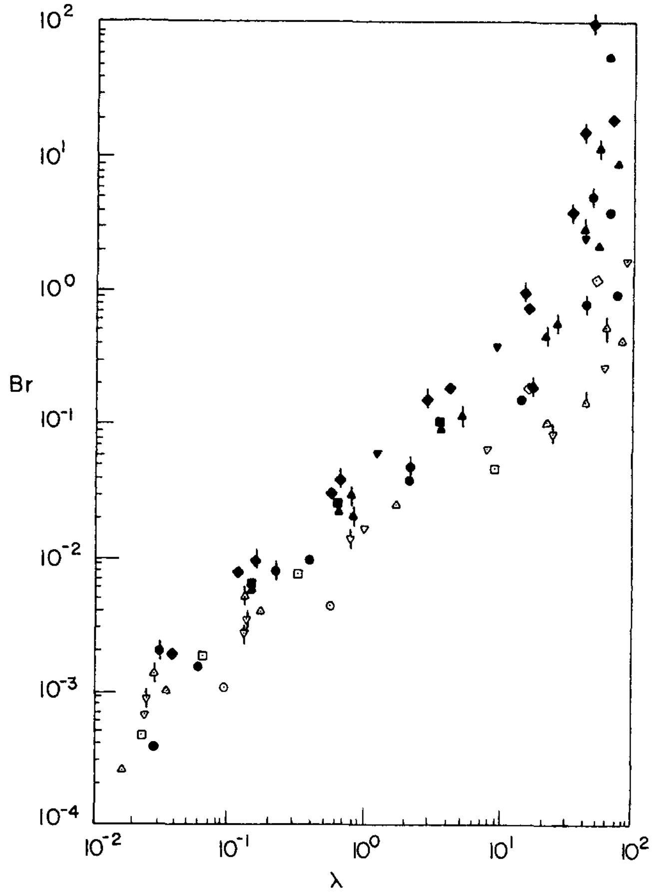

Fig. 10. Br vs. λ data for capillary flow. The temperatures are: ⊙, 33.5°C; □, 36.5°C; △, 38.5°C; ▽, 39.5°C; ◇, 40.5°C; ●, 41.5°C; ■, 43.0°C; ▲, 43.5°C; ▲, 45.5°C; ◆, 48.5°C. Symbols with | refer to capillary no. 2.

* The dimensionless pressure drop P of Sukanek and Laurence (1972) is defined as

$$P \equiv \frac{\tau_\omega D}{\mu_0 V} = \frac{8\,\tau_\omega}{\dot{\gamma}\,\mu_0} \tag{A1}$$

In terms of the Brinkman number, this is found to be

$$P = \frac{16}{2 + Br} \tag{A2}$$

Combining (A1) and (A2) and rearranging

$$\frac{\tau_\omega/\dot{\gamma}}{\mu_0} = \frac{\mu_{app}}{\mu_0} = \frac{2}{2 + Br} \tag{A3}$$

The Brinkman number is independent of the diameter, while $\dot{\gamma}$ depends on D^{-1}. Therefore, for a given $\dot{\gamma}$, the average velocity, and hence Br, increases with increasing D. Since the apparent viscosity depends on the reciprocal of the Brinkman number, increasing D at a given $\dot{\gamma}$ decreases μ_{app}/μ_0.

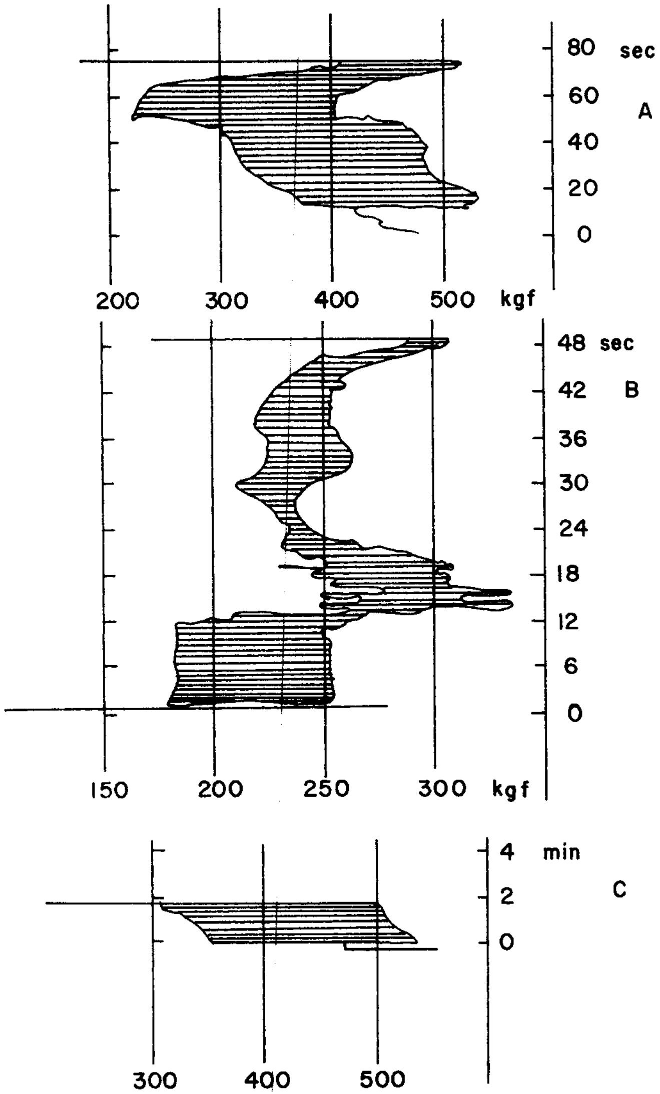

Fig. 11. Force vs. time behavior from Instron recorder during unstable flow. Capillary no. 1. (Note: Only the outline of the trace is shown here): A:$T_o = 36.5°C$, $V_x = 10$ cm/min; B:$T_o = 41.5°C$, $V_x = 20$ cm/min; C:$T_o = 38.5°C$, $V_x = 2$ cm/min.

TABLE 1. CONDITIONS FOR OBSERVED UNSTABLE FLOW

Cap. diam., in.	T_0, °C	V_x*, cm/min	ΔF**, kg_f
0.03018	38.5	5	230-710
	41.5	20	220-260
	36.5	1	500-900
	39.5	10	220-530
0.020	38.5	2	300-540
	39.5	1	390-710
	41.5	10	262-350
	43.5	20	220-255

* V_x is the speed of the Instron crosshead.
** ΔF is the approximate range of the force variations.

Conclusions Concerning Experimental Rheology

The effect of viscous heating on the stress measurements of fluids in simple shear has been studied theoretically for about 45 years. However, little experimental work has been done on this problem. No one has ever attempted to verify in the laboratory the predicted double-valued shear stress until now.

In the Couette instruments, which operate under atmospheric pressure, the measurement of the stress decrease at high Brinkman numbers has been accomplished. The results show that the amount of dissipated heat which remains in the fluid, and, hence, the Brinkman number-shear stress behavior, is a strong function of the boundary conditions.

This has important consequences in viscometry. Rheologists have long known that viscous heating effects must be accounted for in order to accurately determine the viscosity of high molecular weight liquids. The present work clearly shows this. Taken at face value, the apparent viscosity-shear rate curves of Figure 5 show the 1260 to be extremely shear thinning. However, in reality, it is Newtonian.

Results of this kind are common in investigations of viscous dissipation. More significantly, the present study show the importance of accurately determining the thermal behavior of the viscometer boundaries. Before any viscous heating correction can be applied to experimental data, the extent to which the walls of the instrument depart from truly adiabatic or isothermal behavior must be found. A fluid such as Arochlor[R] 1260, whose viscosity is extremely temperature sensitive, would be of great value in such a determination.

NOTATION

Br = Brinkman number
c = mean gap thickness between eccentric cylinders
C_p = heat capacity
D = diameter
e = distance between centers of eccentric cylinders
h = gap thickness
k = thermal conductivity
Nu = Nusselt number
P = dimensionless pressure drop
q = shear parameter defined by Equation (2.7)
R = radius
Ri = radius of inner cylinder
T = temperature
T_0 = reference temperature
T_g = glass transition temperature
V = characteristic velocity

Greek Letters

α = stress parameter defined by Equation (2.7)
β = viscosity-temperature coefficient
$\dot{\gamma}$ = apparent shear rate
ϵ = eccentricity, e/c
κ = ratio of outer to inner diameter
λ = dimensionless shear stress
μ = viscosity
μ_0 = viscosity at temperature T_0
μ_{app} = apparent viscosity
ρ = density
τ_ω = wall shear stress
T = torque on outer eccentric cylinder
T_o = torque on outer concentric cylinder
θ = dimensionless temperature
θ^* = dimensionless surface temperature
Ω_i = angular velocity

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Manuscript received August 27, 1973; revision received and accepted February 6, 1974.