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I am submitting herewith a dissertation written by Raymond Leslie Boles, Jr., entitled "An Experimental Study of Viscoelastic Flow in a Conical Entrance Region." I recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Chemical Engineering.

DeBryne
Major Professor

We have read this dissertation
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DG Thomas

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Herbert S Lee

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James L White

Accepted for the Council:

Hilton A. Smith
Vice President for
Graduate Studies and Research

AN EXPERIMENTAL STUDY OF VISCOELASTIC FLOW
IN A CONICAL ENTRANCE REGION

A Dissertation
Presented to
The Graduate Council of
The University of Tennessee

In Partial Fulfillment
of the Requirements for the Degree
Doctor of Philosophy

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Raymond Leslie Boles, Jr.
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ABSTRACT

An experimental study has been made of the pressure drop in the conical entrance region to a capillary tube. Two fluids, a Newtonian oil and a viscoelastic polymer solution, were used. The entrance half angle was varied from 10.8° to 90° ; the wall shear rate in the capillary was varied from 50 sec^{-1} to 1000 sec^{-1} .

For Newtonian flow, Oka's creeping flow solution was shown to be of the correct form to predict viscous pressure drop effects, although it gave quantitative agreement only at the larger half angles (46.7°). An adaptation of this solution to the power law viscosity model provided a satisfactory prediction of non-Newtonian viscous effects. Weissberg's solution for flow in a sharp-edged entrance ($\alpha=90^\circ$) gave excellent agreement with experimental data.

For viscoelastic flow, the data were correlated semiempirically using an equation that involved a nonlinear viscous term and an elastic term. An attempt to solve the conical flow problem using the theory of Bernstein, Kearsley, and Zapas yielded an intractable result, but it was possible to identify a group with the character of a dimensionless normal deformation rate. This group, which also has the properties of a Deborah number, was found to characterize the elastic pressure drop reasonably well.

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CHAPTER I

INTRODUCTION

The increased production of polymer products in the past two decades has inspired much activity in the field of non-Newtonian fluid mechanics. Efforts are being made to describe, in terms of constitutive equations, the flow of materials which exhibit both elasticity and viscosity. Such equations are used to relate stress and strain to fluid properties. Because the stress distribution in viscoelastic materials depends on the strain history as well as on the instantaneous rate of deformation, most of the constitutive equations which have been proposed are either inadequate for describing the observed phenomena, or too complicated for use in any but simple viscometric flows. If engineering use is to be made of these equations, methods must be developed for applying them to more complex flow problems.

The study of accelerative flows such as flow in a cone, flow between non-parallel flat plates, and flow in tube entrances and exits represents a large effort in that direction. The first step in such problems is to identify the pertinent variables. Dimensionless groups may then be formulated and used to correlate experimental data and to guide the evaluation of constitutive theories. Eventually the most promising constitutive theories will be evaluated

by means of detailed computer calculations.

Bogue and Doughty (10) and Spriggs, Huppler, and Bird (39) have discussed those constitutive equations which seem most promising. In recent work LaNieve (20) used an empirical power law model to correlate end effects for viscoelastic flow at the entrances and exits of capillary tubes. Adams (2) has simplified the constitutive theory of Bernstein, Kearsley, and Zapas (6, 7, 8) and used it to predict the pressure drop for viscoelastic flow between converging and diverging flat plates.

In this investigation the pressure drop due to end effects at the conical entrance of a capillary tube was measured. Two fluids were used, a Newtonian oil and a viscoelastic polymer solution. The length of the capillary, the cone angle, and the shear rate were varied. The Newtonian fluid was used to test the validity and applicability of the existing theory for slow Newtonian flow in a cone. Data for the viscoelastic fluid were correlated with a dimensionless group suggested by the theory of Bernstein, Kearsley, and Zapas. This group has the properties of the Deborah number suggested by Reiner (32) and more recently by Metzner, White, and Denn (26). It consists of a characteristic material time divided by a characteristic process time. If the Deborah number is very large the material may be said to exhibit solid elastic behavior; if the Deborah

number is very small the material may be said to exhibit viscous fluid behavior.

The results of the author's previous work (11) with inelastic fluids provided additional background for this investigation. Pressure drop and flow rate measurements for the flow of inelastic non-Newtonian fluids in cones suggested that limiting solutions for the cases of viscous and inertial flows could be superimposed to give a reasonable prediction of the pressure drop due to the combined effects. It seemed plausible that such an approach might also be applicable to viscoelastic flow, in which case the normal stress effect must also be considered. An equation for calculating the pressure drop due to combined effects would be of the form:

$$\Delta P = \left[\begin{array}{c} \text{inertia} \\ \text{term} \end{array} \right] + \left[\begin{array}{c} \text{shear stress} \\ \text{term} \end{array} \right] + \left[\begin{array}{c} \text{normal stress} \\ \text{term} \end{array} \right]. \quad (\text{I-1})$$

In accelerating viscoelastic flow there is a contribution to the shear stress term due to the strain history of the fluid. Likewise, it has been shown (19) that under certain conditions of accelerating inelastic flow, normal stress can become measurable. Therefore, it may be more useful to consider an equation of the following form for calculating pressure drops due to combined effects:

$$\Delta P = \left[\begin{array}{c} \text{inertia} \\ \text{term} \end{array} \right] + \left[\begin{array}{c} \text{viscous} \\ \text{term} \end{array} \right] + \left[\begin{array}{c} \text{elastic} \\ \text{term} \end{array} \right]. \quad (\text{I-2})$$

This allows one to separate the elastic effect reported here by deducting the viscous and inertial contributions from the total pressure drop. Further evidence that this is a reasonable approach is given by Adams (2). His result for BKZ flow in converging and diverging ducts shows that the viscous terms may be recognized as separate from the elastic terms.

CHAPTER II

FLOW AT A SHARP-EDGED CAPILLARY ENTRANCE

Capillary flow has been used to measure viscosity for more than a century. The basic capillary viscometer consists of two reservoirs connected by a capillary tube. Pressure is applied to one reservoir, forcing the liquid through the capillary into the other reservoir. If the fluid is Newtonian, the viscosity is related to the pressure drop and flow rate by the classical Hagen-Poiseuille equation for developed flow in a tube:

$$\eta = \frac{\pi R^4 \Delta P(\text{CAP})}{8QL}, \quad (\text{II-1})$$

where $\Delta P(\text{CAP})$ is the pressure loss due to developed flow in the capillary.

In practice the measured pressure drop is not that due to developed flow, but also includes dissipation effects due to entrance flow, exit flow, and kinetic energy. If the capillary is used simply to determine the viscosity, these effects may be rendered negligible by using a sufficiently long capillary or by making a suitable correction in terms of an equivalent length of capillary. If, however, the capillary is used in an extrusion or

spinning operation, the character of the entrance and exit flow may seriously affect the quality of the product (13, 23, 46). For this reason entrance and exit flow are two important problems which need to be studied more thoroughly.

A mechanical energy balance around a capillary tube viscometer shows the individual terms which make up the total pressure drop:

$$\frac{\Delta P(\text{TOT})}{\rho} = \Delta K.E. + \underbrace{F_{up} + F_{inside}}_{\text{entrance}} + \underbrace{F_c}_{\substack{\text{developed} \\ \text{flow}}} + \underbrace{F_{down}}_{\text{exit}}, \quad (\text{II-2})$$

where F indicates irreversible energy dissipation. LaNieve (20) discusses in detail each term of Equation II-2. For this viscometer the kinetic energy effects are negligible. Likewise, F_{inside} , the effect of undeveloped flow inside the tube entrance is negligible (4, 29). The present study is concerned only with the effects of developed flow in the tube, F_c , and with the end effects outside the tube, F_{up} and F_{down} . They will be discussed in detail in this chapter.

I. PRESSURE DROP DUE TO DEVELOPED FLOW

The pressure loss due to developed flow in a tube is caused entirely by viscous dissipation. A momentum balance may be used to relate the shear stress at the wall of the

tube to the pressure drop (9):

$$\tau_R = \frac{R\Delta P(CAP)}{2L}. \quad (II-3)$$

Mooney and Rabinowitsch derived an expression for the wall shear rate of a general time-independent fluid. This expression is given by Wilkinson (49) as

$$-\left(\frac{dv}{dr}z\right)_R = \frac{3n'+1}{4n'} \frac{8V}{d}, \quad (II-4)$$

where

$$n' = \frac{d \ln \left(\frac{R\Delta P(CAP)}{2L} \right)}{d \ln \left(\frac{8V}{d} \right)}. \quad (II-5)$$

The shear stress and shear rate may be related empirically by the power law:

$$\tau = kD^n, \quad (II-7)$$

where k and n are material parameters. It can be easily shown that $n = n'$. Combining Equations II-3, II-4, and II-6 gives the pressure drop due to developed flow of a power law fluid in a tube:

$$\Delta P(CAP) = \rho F_C = \frac{2L}{R} k \left(\frac{3n+1}{4n} \frac{4Q}{\pi R^3} \right)^n. \quad (II-7)$$

For a Newtonian fluid $n = 1$, k becomes the viscosity, and Equation II-7 reduces to the Hagen-Poiseuille relationship.

II. PRESSURE DROP DUE TO END EFFECTS OUTSIDE THE TUBE

Evaluation of End Effects

Philippoff and Gaskins (29) found that the end effects outside the tube could be evaluated by using tubes of different L/R values. They performed a macroscopic energy balance about the viscometer. Losses due to kinetic energy and undeveloped flow were neglected. The elastic energy transported out of the capillary was assumed to be the axial normal force P_{11} whose value at the wall is $(P_{11})_R$. The energy balance is

$$\Delta P(\text{TOT}) = \rho F_{\text{up}} + \rho F_{\text{down}} + (P_{11})_R + \Delta P(\text{CAP}). \quad (\text{II-8})$$

It was assumed that

$$(P_{11})_R = \tau_R S_R, \quad (\text{II-9})$$

where S_R is the recoverable shear at the wall. The irreversible viscous dissipation at the ends is treated as an additional effective length of capillary:

$$\rho F_{\text{up}} + \rho F_{\text{down}} = \frac{2n_c}{R} \tau_R. \quad (\text{II-10})$$

From Equation II-3 the energy loss in the capillary is

$$\Delta P(\text{CAP}) = \frac{2L}{R} \tau_R. \quad (\text{II-11})$$

Combining Equations II-8 to II-11 gives

$$\Delta P(\text{TOT}) = \tau_R \left[\frac{2L}{R} + 2n_c + S_R \right]. \quad (\text{II-12})$$

If one plots $\Delta P(\text{TOT})$ versus L/R the result should be a straight line of slope $2\tau_R$ and of intercept $-(n_c + S_R/2)$. For very elastic fluids n_c is negligible and S_R is very large. For inelastic fluids $S_R=0$, and the entire value of the intercept represents viscous dissipation. Bagley (5) derived equations which allow one to separate the elastic and viscous effects provided the fluid obeys Hooke's law. For most viscoelastic fluids, however, it is impossible to distinguish between a large value of S_R and a large value of n_c .

Bagley (4) has shown experimentally that a plot of $\Delta P(\text{TOT})$ versus L/R gives a straight line as predicted by Equation II-12. His experiments employed very short tubes, including a sharp-edged orifice with $L/R=0$. The fact that the straight line relationship was verified even at low values of L/R seems to indicate that the state of the flow at $L=0$ is the same as at very large values of L ; i.e., the flow is fully developed at the tube entrance. Other experi-

ments show, however, that the increase in the diameter of the exit stream from a capillary due to the release of normal stresses is very much dependent on the L/R ratio of the tube (3, 24). These two observations lead to the conclusion that pressure drop experiments are not a very sensitive measure of the degree of developed flow in the capillary.

Prediction of Viscous End Effects

Several analyses have attempted to describe the end effects outside the tube (34, 35, 44, 45, 48). While the exact character of the flow is still not known, the analyses have been successful in predicting the viscous dissipation due to converging flow prior to the entrance and diverging flow at the exit of the tube for slow flows of very viscous Newtonian fluids. For low flow rates the flows in the entrance and exit regions are so similar that the dissipation in each region may be assumed the same.

For creeping flow the equation of motion reduces to

$$\nabla \cdot \bar{\tau} = 0, \quad (\text{II-13})$$

which becomes

$$\nabla P = \eta \nabla^2 \bar{v} \quad (\text{II-14})$$

for an incompressible Newtonian fluid. By introducing the

stream function this can be reduced to Laplace's equation:

$$\nabla^2 \psi = 0. \quad (\text{II-15})$$

This equation was first solved for creeping flow through an orifice by Sampson (35) in 1891. Later it was corrected by Roscoe (34).

More recently Weissberg (48) solved the problem of slow Newtonian converging entrance flow in the oblate spheroidal coordinate system. He found that the streamlines coincided with one of the coordinate axes, allowing the use of only one velocity component. The oblate spheroidal coordinate system is defined in terms of the entrance problem in Figure 1. The equation for the velocity component as a function of position in the entrance region is

$$u(t) = -\frac{3Q}{2\pi R^2} \frac{x^2}{(t^2+1)^{\frac{1}{2}}(t^2+x^2)^{\frac{1}{2}}}. \quad (\text{II-16})$$

The pressure difference between a point far in front of the entrance and a point in the converging flow field is

$$P_{\infty} - P(x) = \frac{3\eta Q}{\pi R^2} \left[\frac{x}{R(x^2+t^2)} \frac{1}{R} \tan^{-1} \left(\frac{1}{x} \right) \right]. \quad (\text{II-17})$$

Evaluating Equation II-17 for the point at the centerline in the plane of the entrance ($t=0$) gives

$$\Delta P(\text{ENT}) = \rho F_{\text{up}} = \frac{3\eta Q}{2R^3}. \quad (\text{II-18})$$

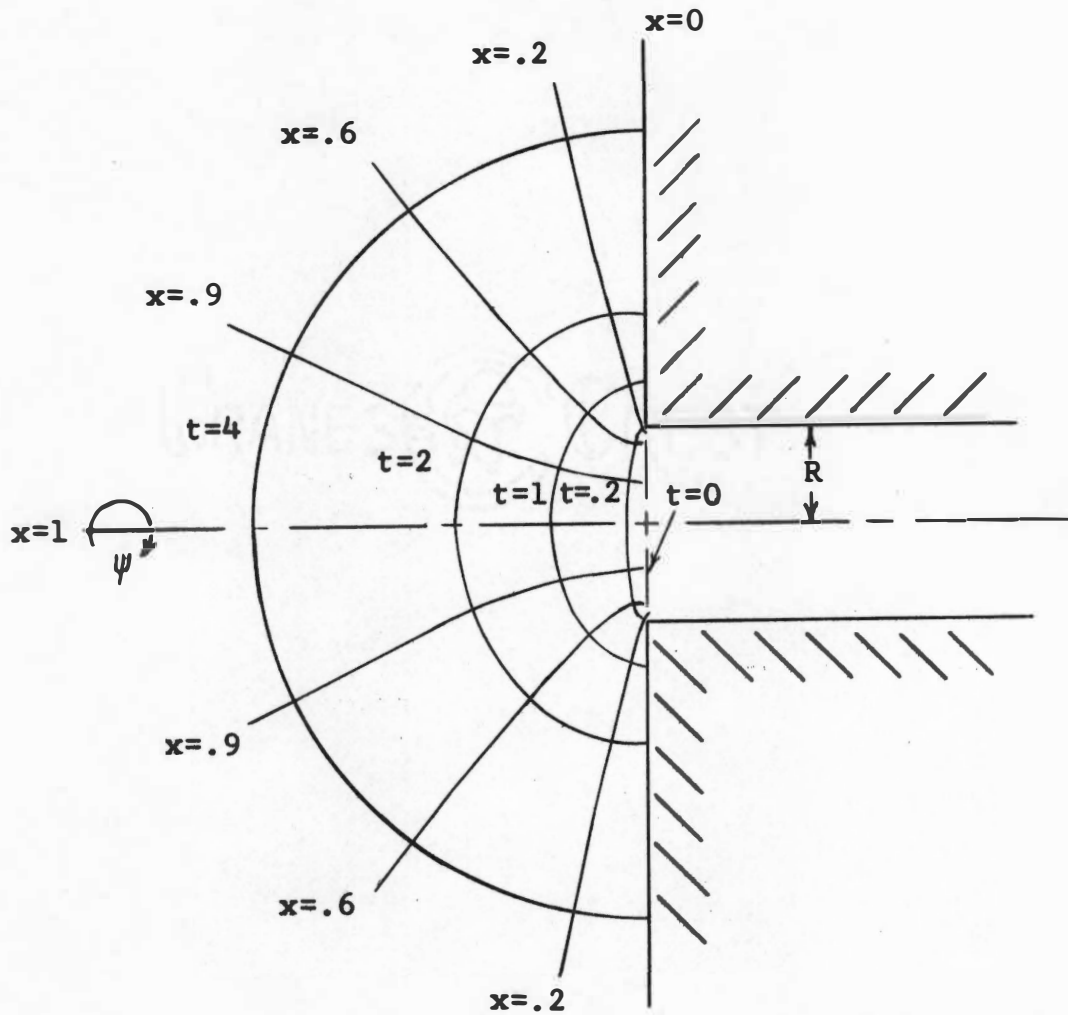


Figure 1. The Capillary Entrance in Oblate Spheroidal Coordinates.

Assuming that the streamlines are the same at the exit as at the entrance for creeping flow:

$$\Delta P(\text{ENDS}) = \rho F_{\text{up}} + \rho F_{\text{down}} = \frac{3\eta Q}{R^3}. \quad (\text{II-19})$$

A solution in spherical coordinates for creeping flow of a Newtonian fluid through a tapered tube is given by Oka (28). He solved the Navier-Stokes equations assuming flow in the radial direction only. If one assumes for the case of the plane entrance (180° included angle) that the radial flow assumption is not reasonable any closer to the entrance than the distance equal to the capillary radius R , the centerline pressure drop is

$$\Delta P(\text{ENT}) = \frac{2}{\pi} \frac{\eta Q}{R^3}. \quad (\text{II-20})$$

Comparison of this with Weissberg's result shows that the spherical coordinate solution predicts a pressure drop which is less by a factor of about 2.3. This seems to indicate that most of the Newtonian entrance loss occurs in a region very close to the entrance where $r < R$.

CHAPTER III

FLOW IN A CONE

The problem of creeping Newtonian flow in a cone was first solved by Harrison (16) in 1919 and Bond (12) in 1925. More recently Oka (28) rediscovered their solution. Newtonian flow with inertia effects has been treated by Ackerman (1), Nishimura and Oka (27), and Slezkin (38). Sutterby (40, 41, 42) and Boles (11) have reported experimental studies of inelastic non-Newtonian flow with inertial effects. Some experimental work on the flow of viscoelastic fluids has been reported (13, 36, 37), but in these studies emphasis was on the effect of cone angle on extrudate irregularities in the flow of polymer melts. Kaloni (17, 18) performed a theoretical investigation of flow of a viscoelastic fluid in a cone using the Oldroyd constitutive equation and a perturbation scheme for solution of the differential equations. His conclusion was that the effect of the elasticity of the fluid was to cause a secondary flow. His results were of a form which was inconvenient for comparison with experimental measurements of pressure drop and flow rate.

The problem of flow in a cone is best treated in the spherical coordinate system (Figure 2) where the radial flow

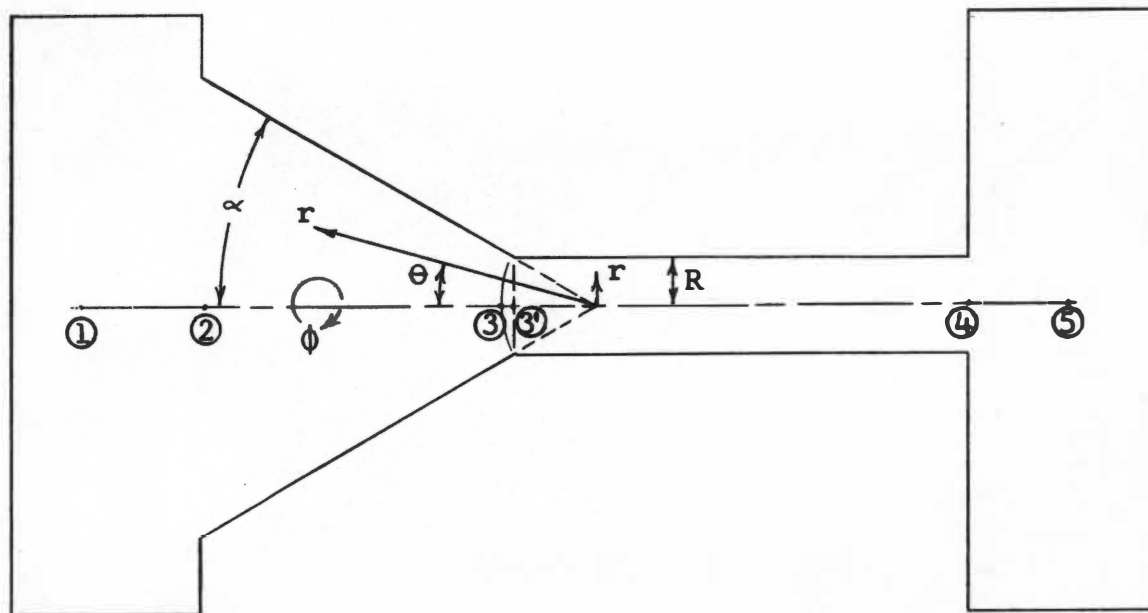


Figure 2. The Spherical Coordinate System:
Flow from a Cone into a Tube.

assumption may be made. The axially-symmetric cylindrical system with rz -coordinates was considered, but discarded because it necessitates the use of two velocity components. The following treatment of the conical flow problem makes use of the spherical coordinate system with the assumptions that $v_\phi=0$, $v_\theta=0$, and $\partial(\)/\partial\phi = 0$. The inertial terms are neglected here but are discussed in Appendix B.

The equations of motion in spherical coordinates may be expressed as follows:

$$-\frac{\partial P_{rr}}{\partial r} = \frac{1}{r \sin \theta} \frac{\partial}{\partial \theta} (\tau_{r\theta} \sin \theta) + \frac{P_{rr} - P_{\theta\theta}}{r} + \frac{P_{rr} - P_{\phi\phi}}{r}, \quad (\text{III-1})$$

and

$$-\frac{\partial P_{\theta\theta}}{\partial \theta} = \frac{1}{r} \frac{\partial}{\partial r} (r^2 \tau_{r\theta}) + \tau_{r\theta} + \cot \theta (P_{\theta\theta} - P_{\phi\phi}). \quad (\text{III-2})$$

These equations have been expressed in terms of the total stress, P_{ij} , where

$$P_{ij} = -Pg_{ij} + \tau_{ij}. \quad (\text{III-3})$$

This is the quantity which is actually measured in most experiments instead of the isotropic pressure, P .

The continuity equation for radial flow in a cone is

$$r^2 v_r = f(\theta). \quad (\text{III-4})$$

The components of the deviator stress may then be written:

$$\begin{bmatrix} \tau_{rr} & \tau_{r\theta} & \tau_{r\phi} \\ \tau_{\theta r} & \tau_{\theta\theta} & \tau_{\theta\phi} \\ \tau_{\phi r} & \tau_{\phi\theta} & \tau_{\phi\phi} \end{bmatrix} = \frac{\eta}{r^3} \begin{bmatrix} -4f & f' & 0 \\ f' & 2f & 0 \\ 0 & 0 & 2f \end{bmatrix}. \quad (\text{III-5})$$

I. NEWTONIAN FLOW

Assume creeping radial converging flow of a Newtonian fluid in a cone of half-angle α . Expressing the equations of motion in terms of the isotropic pressure:

$$\frac{\partial P}{\partial r} = \frac{\eta}{r^4 \sin \theta} \frac{\partial}{\partial \theta} (\sin \theta f'), \quad (\text{III-6})$$

and

$$\frac{\partial P}{\partial \theta} = \frac{2\eta}{r^3} f'. \quad (\text{III-7})$$

Eliminating P between Equations III-6 and III-7 gives

$$\frac{d}{d\theta} \left[\frac{1}{\sin \theta} \frac{d}{d\theta} (f' \sin \theta) \right] + 6f' = 0. \quad (\text{III-8})$$

Integration with respect to θ gives

$$\frac{1}{\sin \theta} \frac{d}{d\theta} (f' \sin \theta) + 6f = C, \quad (\text{III-9})$$

where C is the integration constant. The boundary conditions are

$$(1) \quad f'(0) = 0, \quad (\text{III-10})$$

$$(2) \quad f(\alpha) = 0, \quad (\text{III-11})$$

$$(3) \quad Q = - \int_0^{2\pi} \int_0^\alpha r^2 v_r \sin \theta \, d\theta d\phi. \quad (\text{III-12})$$

Oka (28) recognized Equation III-9 as a form of the Legendre differential equation. His solution is

$$f = r^2 v_r = \frac{-3Q \sin^2 \alpha}{2\pi(1-\cos \alpha)^2(1+2\cos \alpha)} \left[1 - \left(\frac{\sin \theta}{\sin \alpha} \right)^2 \right]. \quad (\text{III-13})$$

The pressure drop between two radial positions r and r_1 is

$$P-P_1 = - \frac{3Q(\cos 2\theta+1/3)\eta}{2\pi(1-\cos \alpha)^2(1+2\cos \alpha)} \left(\frac{1}{r^3} - \frac{1}{r_1^3} \right). \quad (\text{III-14})$$

These results make two rather interesting predictions. From Equation III-13 it can be seen that the velocity becomes zero when $\theta=\alpha$ and when $\theta=\pi-\alpha$. Therefore, for values of $\theta > \pi-\alpha$, where $\alpha > \pi/2$, the flow is reversed in sense. Equation III-14 shows that the pressure is proportional to $(\cos 2\theta+1/3)$. Therefore, when $\cos 2\theta=-1/3$ or $\theta=54^\circ 45'$, there is a reversal of the radial pressure gradient.

Bond (12) photographed tracer bubbles for the flow of glycerine into cones of very large half-angles ($\alpha=90^\circ$, 110° , 141° , 160°). His experiments did not verify the prediction of backflow, but he observed that for $\alpha > 90^\circ$ the

radial flow assumption was not valid. For $\alpha = 90^\circ$ the radial flow assumption was observed to be valid except at very small values of r . Tracer measurements of the velocity for $\alpha = 90^\circ$ differed little from the prediction of Equation III-13. The prediction of reversal of the radial pressure gradient was verified at the wall for $\alpha = 90^\circ$.

If Equation III-14 is to be compared with experimental measurements, one should determine whether a correction is necessary for the deviatoric normal stress. From Equations III-3, III-5, and III-14, it can be shown that

$$P_{\theta\theta} = \frac{3\eta Q(\cos 2\alpha + 1/3)}{2(1 - \cos \alpha)^2(1 + 2\cos \alpha)r^3}. \quad (\text{III-15})$$

Note that $P_{\theta\theta}$ is not a function of θ , the angular position in the cone. Thus, pressure drop measurements at the wall of the cone should give

$$P_{\theta\theta}(r) - P_{\theta\theta}(r_1) = \frac{3\eta Q(\cos 2\alpha + 1/3)}{2(1 - \cos \alpha)^2(1 + 2\cos \alpha)} \left(\frac{1}{r^3} - \frac{1}{r_1^3} \right). \quad (\text{III-16})$$

This is the negative of the quantity which would be obtained using Equation III-14; i.e.,

$$P_{\theta\theta} = -P. \quad (\text{III-17})$$

Therefore no correction is needed for pressure drop measurements at the wall of a cone. This observation could have

been made very easily by noting that $\tau_{\theta\theta} = 2\eta f/r^3 = 0$ at the wall. If one wished to predict pressure variations such as those measured by pitot tubes, a similar derivation could be performed for the total stress in the r -direction, though in this case the deviatoric normal stress would be important.

A more common problem is that of combining pressure equations for simple geometries to predict pressure drops for complicated flow systems. A convenient way to do this is to match the solutions along a straight line such as the centerline. For instance, the conical solution might be matched with the tube flow solution in the manner described below.

For flow in a pipe, the appropriate equation of motion is

$$\frac{\partial P_{zz}}{\partial z} = \frac{\partial P}{\partial z} = \frac{1}{r} \frac{\partial(r\tau_{rz})}{\partial r}. \quad (\text{III-18})$$

When integrated this gives the classical Hagen-Poiseuille equation, which may be written

$$-\Delta P_{zz} = \Delta P = \frac{8Q\eta L}{\pi R^4}. \quad (\text{III-19})$$

Matching total stresses along the centerline gives

$$\begin{aligned}
P_1 - P_5 = & \left[P_1 - (-P_{rr})_2 \right]_{\xi} + \left[(-P_{rr})_2 - (-P_{rr})_3 \right]_{\xi} \\
& + \left[(-P_{rr})_3 - (-P_{zz})_3 \right]_{\xi} + \left[(-P_{zz})_3 - (-P_{zz})_4 \right]_{\xi} \\
& + \left[(-P_{zz})_4 - P_5 \right]_{\xi} .
\end{aligned} \tag{III-20}$$

If, wherever the total stress appears in Equation III-20, one substitutes the sum of the isotropic pressure and the deviatoric stress according to Equation III-3, the result is

$$\begin{aligned}
P_1 - P_5 = & \left[P_1 - P_2 \right]_{\xi} + \left[P_2 - P_3 \right]_{\xi} + \left[P_3 - P_3 \right]_{\xi} \\
& + \left[P_3 - P_4 \right]_{\xi} + \left[P_4 - P_5 \right]_{\xi} .
\end{aligned} \tag{III-21}$$

Note that all deviatoric stresses in Equation III-20 have been eliminated by cancellation of terms.

All terms of Equation III-21 can be calculated directly or estimated by equations presented previously, except the term $\left[P_3 - P_3 \right]_{\xi}$. This involves the small but important region where the flow in the cone merges with the flow in the tube. In this region it is unlikely that the velocity can be described by rays or by the parabolic profile of the tube. If the radial flow assumption were valid one could say that the deviatoric stress reaches its maximum value in this region. Furthermore, if the velocity profile were fully developed at the tube entrance, as indicated by pressure drop measurements (4), one could say that

the deviatoric stress disappears in this region. Thus, it would seem that the fluid suddenly loses its acceleration as it enters the tube. It may be argued that the deviatoric normal stress, τ_{rr} , represents tension in the accelerating flow field. In the region near the exit the tension is relaxed so that the net change in isotropic pressure is small. Parenthetically, it should be noted that if this argument is correct, then one would expect a very viscous Newtonian fluid to expand at the exit of an accelerating flow field, though certainly not to the extent that has been observed with viscoelastic fluids.

II. NON-NEWTONIAN FLOW

An exact solution such as the one just discussed has not been reported for the problem of non-Newtonian flow in a cone. Tomita (45) obtains a solution for radial flow of a power law fluid in a 90° entrance between $r \rightarrow \infty$ and $r=R$ by assuming a form for the stream function ψ . His assumption is viewed with suspicion, however, because it gives a velocity profile which is independent of the power law parameter n . A different approach to this problem is given below.

For a power law fluid the viscosity is given by

$$\eta = k \left| \frac{1}{r} \frac{\partial v_r}{\partial \theta} \right|^{n-1} \quad (\text{III-22})$$

Combining Equations III-1 to III-5 with Equation III-22, the equations of motion may be written:

$$\frac{\partial P}{\partial r} = \frac{k}{r^{3n+1} \sin \theta} \frac{\partial}{\partial \theta} (f'^n \sin \theta). \quad (\text{III-23})$$

and

$$\frac{\partial P}{\partial \theta} = \frac{(5-3n)k}{r^{3n}} f'^n + \frac{2(n-1)k}{r^{3n}} f f'^{n-2} f''. \quad (\text{III-24})$$

An analytical solution for the velocity profile of a power law fluid in a cone was not found. However, by analogy with the Newtonian velocity profile (Equation III-13) and the power law velocity profile for flow in a tube, the following expression was assumed:

$$f = r^2 v_r = \frac{-3Q \sin^2 \alpha}{4\pi(1-\cos \alpha)^2(1+2\cos \alpha)} \left(\frac{3n+1}{n+1} \right) \cdot \left[1 - \left(\frac{\sin \theta}{\sin \alpha} \right)^{\frac{n+1}{n}} \right]. \quad (\text{III-25})$$

This expression reduces properly to the Newtonian solution when $n=1$. Combining Equations III-23 and III-25 and integrating with respect to r gives

$$P_1 - P = \frac{k}{3n \sin \alpha} \left[\frac{3Q \sin \alpha \cos \theta (3n+1)}{4\pi n (1-\cos \alpha)^2 (1+2\cos \alpha)} \right]^n \cdot \left[2\cos \theta - n \frac{\sin^2 \theta}{\cos \theta} \right] \left[\frac{1}{r_1^{3n}} - \frac{1}{r^{3n}} \right]. \quad (\text{III-26})$$

For $n=1$ this reduces to the Newtonian expression of Equation III-14. For small values of α Equation III-26 reduces to the result reported in the author's master's thesis (11).

At the centerline Equation III-26 may be written

$$\begin{aligned} [P_1 - P]_{\frac{1}{2}} = & \frac{2k}{3n \sin \alpha} \left[\frac{3Q \sin \alpha (3n+1)}{4\pi n (1 - \cos \alpha)^2 (1 + 2 \cos \alpha)} \right]^n \\ & \cdot \left[\frac{1}{r_1^{3n}} - \frac{1}{r^{3n}} \right]. \end{aligned} \quad (\text{III-27})$$

Equation III-27 can be combined with Equation II-7 for flow of a power law fluid in a tube in the same manner that the Newtonian result was combined with the Hagen-Poiseuille equation.

III. VISCOELASTIC FLOW

An attempt was made to solve the problem of viscoelastic flow in a cone using the constitutive theory of Bernstein, Kearsley, and Zapas. Analytical results were not obtained because of the complicated form of the integrals involved. Descriptions of the rigorous approach to the problem and of an approximate approach which is valid only at very low flow rates are given in Appendix A.

From the attempts to solve the problem it became evident that the pressure drop would depend on two dimensionless groups, Fs^* and $F's^*$, where $F=f/r^3$. These groups

have the properties of Deborah numbers in that they involve the ratio of a material time to a process time.

Metzner, White, and Denn (26) postulate that the relevant process time is the residence time of a fluid particle in the accelerating flow field. In reality, however, the residence time is not easily obtained for most problems. In practice this problem may be avoided by noting that the above-mentioned groups are related to the residence time for any assumed velocity profile. When an explicit solution to a specific problem is available, one group may be more important. For example, if the pressure drop between two taps at the wall is of interest, $F'_w s^*$ would seem to be the appropriate Deborah number for correlating the elastic contribution because the quantity f , being zero at the wall, does not appear in the solution of the problem. On the other hand, if the overall pressure drop across a larger system, such as a tube in addition to a cone, is being considered it is intuitively reasonable to make a balance along the centerline, as discussed previously. In such a case F' will not appear in the solution (because $F'_0=0$) and $F_0 s^*$ would seem to be the appropriate Deborah number.

The quantity F is really the derivative of V_r with respect to r . It may, therefore, be thought of as a normal deformation rate. This function allows one to circumvent

the problem of defining a residence time. The function f is constant along a ray. One may postulate that the particle begins to accelerate at $r \rightarrow \infty$, and terminates its acceleration in the vicinity of the tube entrance. The lower limit of r is artificially specified as $r = R \csc \alpha$, which is the distance from the virtual origin of the cone to the point at the wall where the cone joins the tube. Although it may be correctly argued that the fluid particle continues to accelerate up to the tube entrance (or, indeed, well into the tube), this choice of r allows one to treat the case of the sharp-edged entrance ($\alpha = 90^\circ$) in the same manner that the conical entrance is treated. It may also be argued that the radial flow assumption is not valid any closer to the entrance than has been specified. Thus, the choice of the lower limit of r must remain arbitrary until a more complete analysis of the composite problem has been made.

Adams (2) was able to solve analytically the problem of viscoelastic flow between converging and diverging flat plates using the BKZ theory. His result for the radial total stress at the centerline, though complex, is of the following form:

$$\begin{aligned} \left[P_{rr}(\infty) - P_{rr}(r) \right]_{\phi} &= \left[4F_0 - F_0'' \right] \frac{\eta_0}{2} \\ &+ \frac{\gamma_0}{s^*} \phi(F_0 s^*, F_0' s^*, a), \quad (\text{III-28}) \end{aligned}$$

where

$$F_o = \left. \frac{f}{r^2} \right|_c,$$

and

$$F_o'' = \left. \frac{f''}{r^2} \right|_c.$$

His result for the total stress $P_{\theta\theta}|_w$ which one measures with taps at the wall is

$$\begin{aligned} [P_{\theta\theta}(\infty) - P_{\theta\theta}(r)]_w &= -\frac{F'_w \eta(F'_w s^*)}{2} \\ &+ \frac{1}{4a} \frac{\gamma_o}{s^{*2}} \ln[1 + a F'^2_w s^{*2}], \quad (\text{III-29}) \end{aligned}$$

where

$$F'_w = \left. \frac{f'}{r^2} \right|_w,$$

$$F'_w = \left. \frac{f''}{r^2} \right|_w,$$

and

$$\eta(F'_w s^*) = \eta_o - \frac{\gamma_o}{2s^*} \ln(1 + a F'^2_w s^{*2}).$$

The first term in both results can be interpreted as a viscous effect, being cast in terms of η , and can therefore be generalized to other viscous models.

Equation III-28 is of the same form as that suggested by LaNieve (20) for end effects in sharpened-edged capillary

flow, although LaNieve's model was much more empirical. From a dimensional analysis argument, he suggested that

$$\Delta P(\text{END}) = \alpha_2 H_{\text{ch}} \lambda_{\text{ch}}^{b_2} D^{b_2} + \alpha_3 H_{\text{ch}} \lambda_{\text{ch}}^{b_3} D^{b_3}, \quad (\text{III-30})$$

where H_{ch} has units of stress and λ_{ch} has units of time. Taken together $H_{\text{ch}} \lambda_{\text{ch}}^n$ is merely the power law parameter k . The quantity $\lambda_{\text{ch}} D$ could be interpreted as a form of the Deborah number. LaNieve used empirical power law relationships for both the shear and the normal stresses. They are

$$\tau_{12} = H_{\text{ch}} \lambda_{\text{ch}}^n D^n, \quad (\text{III-31})$$

and

$$\tau_{11} - \tau_{22} = H_{\text{ch}} \lambda_{\text{ch}}^{2m} D^{2m}. \quad (\text{III-32})$$

He found that for large values of shear rate, $b_3=2m$. On the other hand, he used the Coleman-Noll second order theory, which is valid only for very low shear rates, to predict that $b_3=2$.

From these results of Adams and LaNieve, one would reasonably expect that the pressure drop in the conical entrance region would be given by

$$\Delta P(\text{ENT}) = \Delta P(\text{VISC}) + \Delta P(\text{EL}), \quad (\text{III-33})$$

where $\Delta P(\text{VISC})$ is the viscous effect and $\Delta P(\text{EL}) = \phi_2(F_0 s^*)$ is the elastic effect. The viscous effect may be given by the

power law solution (Equation III-26): The quantity F_0 will likewise be taken from the power law solution (Equation III-25). No functional form will be assumed for $\Delta P(EL)$, but one would expect from the results of LaNieve and Adams that

$$\Delta P(EL) \sim (F_0 s^*)^2 \quad \text{for small } F_0 s^*,$$

and that

$$\Delta P(EL) \sim (F_0 s^*)^{2m} \quad \text{for large } F_0 s^*.$$

CHAPTER IV

EXPERIMENTAL: EQUIPMENT, PROCEDURE, AND TREATMENT OF DATA

I. APPARATUS

The experimental pressure drop measurements were made with a modified capillary viscometer which was designed by H. L. LaNieve and built by the Chemical and Metallurgical Engineering Shop at The University of Tennessee. Details of the equipment design are given by LaNieve (20). A brief description will be given here.

The apparatus is shown schematically in Figure 3. Basically, the viscometer consists of two reservoirs joined by a capillary tube. Regulated nitrogen pressure forces the fluid from the entrance reservoir, through the tube, and into the exit reservoir. The flow rate is determined by measuring the volume of air displaced from the exit reservoir. The modification consists of small cones which can be fastened to the capillary holder forming a conical entrance into the tube.

Reservoir Design

The reservoirs are cylindrical with capacities of about 400 ml. One end of each reservoir is flanged and

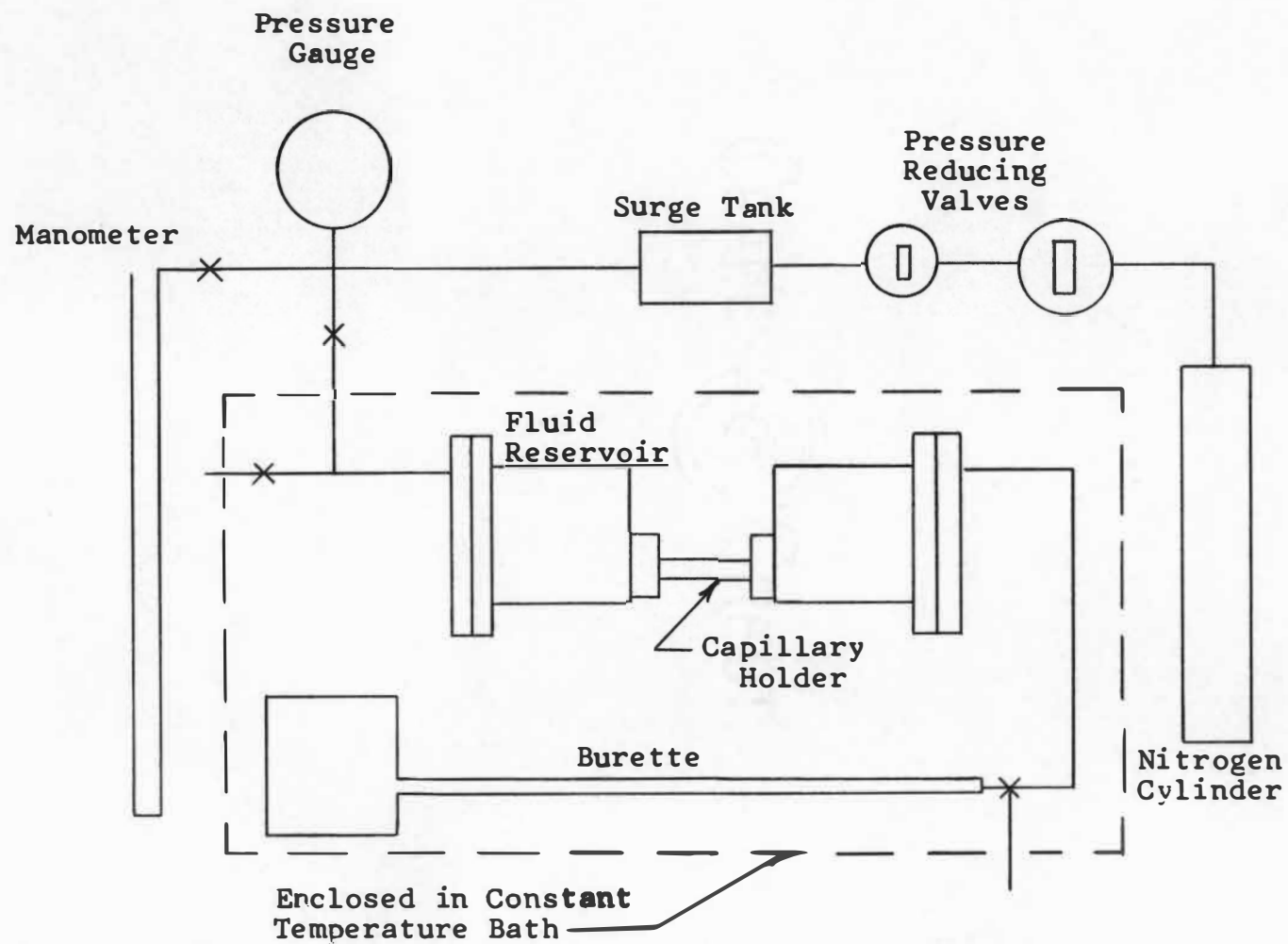


Figure 3. Experimental Apparatus.

sealed with an O-ring. The flange plate contains a pressure fitting near the top for connection with either the pressure source or the flow measuring system. The capillary tubes are encased in holders which enter the reservoirs through pressure fittings in the ends opposite the flanges. These pressure fittings were designed to permit the exchange of capillary tubes while the viscometer is charged with fluid.

Capillary Design and Calibration

In order to obtain the desired shear rates (between 10^2 and 10^3 sec.⁻¹) and not extend the pressure and flow rate beyond the capacity of the instrument, it was necessary to use capillary tubes with an inside diameter of about 1 mm. Glass tubes were used.

The diameters of the longer tubes were determined by measuring the length of a mercury drop in the tube with a cathometer. The mercury was then weighed and the diameter calculated by relating the weight and volume of the mercury drop. This procedure was performed for several sizes of mercury drops at several positions in the tube.

For the shorter capillary tubes (about 1/8 and 1/4-inch long) the above method of calibration was found to be inadequate. The diameters of the short capillaries were determined with a 5X microscope, using a micrometer eyepiece calibrated in tenths of a millimeter. Of course, this

method was inadequate for the longer capillaries because non-uniformity of the tube diameter could not be detected. The dimensions of the capillary tubes and holders are given in Table I.

In order to withstand high pressures and to permit easy handling, the capillaries were encased in brass holders. The tubing was cemented into the holders with hard setting epoxy resin glue. The ends of the holder with the glass protruding were then sanded down with a wet belt sander. Because of the size of the pressure fittings used to secure the capillary holders to the reservoirs, there was a minimum length for the holders of about 3.5 inches. This was greater than the desired length of some of the capillaries, making it necessary to devise another method for holding the capillaries. For this reason the brass holders for the short capillaries were drilled to 0.5-in. inside diameter. The glass tubing was mounted in brass slugs of 0.5-in. outside diameter and of the desired capillary length. The brass slugs were mounted in the holders. The flush end of the holder was used as the entrance, giving a very large entrance ratio. The exit ratio was 1 mm. to 0.5 in. or about 1:10.

Cone Design

Conical entrances with half-angles (α) of 10.8° , 22° ,

TABLE I

DIMENSIONS OF CAPILLARIES AND HOLDERS

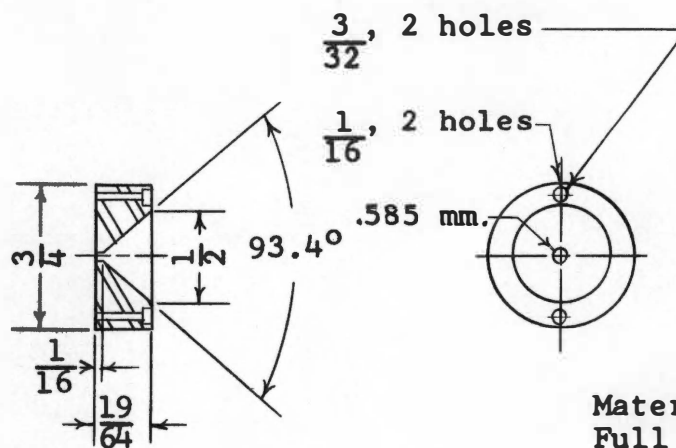
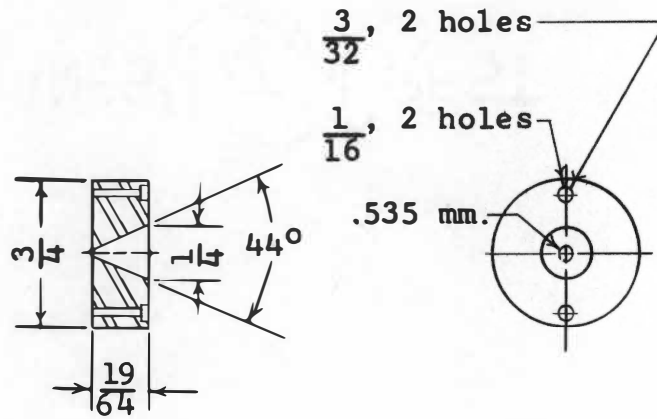
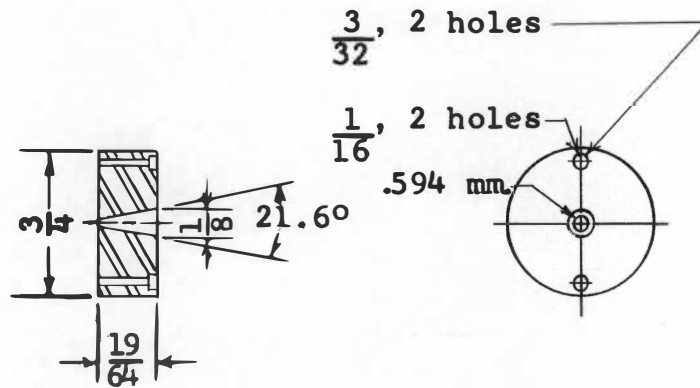
Capillary	Radius R (mm.)	Length L (in.)	Length to Radius Ratio L/R	Barrel Radius R _B (in.)	Barrel Length L _B (in.)
A1	.538	3.531	166.7	-	0
A2	.549	1.125	52.1	.25	2.672
A3	.568	.2185	9.77	.25	3.688
A4	.525	.125	6.05	.25	3.516
A5	-	-	-	.25	3.905

and 46.7° were used as well as a sharp-edged entrance ($\alpha=90^\circ$). The cones were constructed of brass and held to the capillary holders by 00-80 machine screws. Detail drawings of the cones are shown in Figure 4. An assembly drawing of a capillary holder and cone is shown in Figure 5. The ends of the cones were intended to be the same size as the glass capillaries. However, microscopic examination revealed small deviations.

One capillary holder was drilled to 0.5-in. diameter and used without a capillary. In this way the conical entrances alone could be used, giving an L/R ratio of nearly zero. Ultimately, measurements made without the glass capillary tubes were used to determine the end effects. This made the deviation between the capillary diameter and the diameter of the small end of the cone of little consequence.

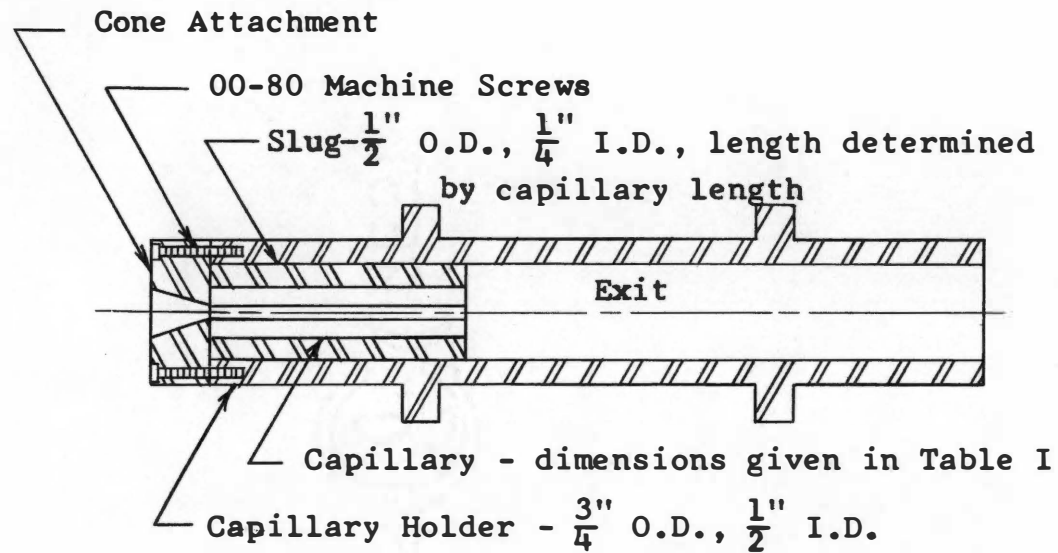
Pressure Control System

Regulated nitrogen pressure from a gas cylinder was used to push the fluid through the capillary. A 30 cu. in. surge tank was used to further smooth pressure variations. The high pressure of the gas cylinder made it necessary to use a large pressure regulator whose response was too slow to give adequate control. For this reason a smaller, faster responding pressure regulator was used in series with the



Material--Brass
Full Scale

Figure 4. Detail Drawing of Conical Attachments.



Material--Brass
Full Scale

Figure 5. Section of Assembled Cone and Capillary Holder.

larger one. For pressures of 1 to 25 p.s.i.g., a Watts type 118 model M regulator was used. For pressures of 25 to 125 p.s.i.g., a regulator made by Arrow Tools, Inc. was used.

At low pressures a 1000 cm. mercury manometer was used to measure the pressure. At higher pressures an Ashcroft laboratory test gauge with a range of 0 to 200 p.s.i.g. marked in one p.s.i.g. graduations was used. It was calibrated with a dead weight gauge and found to be off zero, reading low by 3.1 p.s.i.g. The pressure readings were corrected accordingly.

Temperature Control System

Although the experimental measurements were made at room temperature, the room temperature fluctuation was enough to affect both the viscosity and the flow rate measurement significantly. The temperature was controlled by immersing both the viscometer and the flow measuring system in a constant temperature bath. The bath permitted control of $\pm 0.05^{\circ}\text{C}$. at 25°C . The temperature was measured with a thermometer which was calibrated against a National Bureau of Standards thermometer. It had a range of 19° - 34°C ., with divisions of 0.02°C .

Flow Rate Measuring System

The flow rate of the fluid through the capillary tube was determined by measuring the volume of air displaced from

the exit reservoir in a measured amount of time. The air flowed into a 5 ml. precision bore burette with .01 ml. graduations, displacing water from it. The bore of the burette was small enough to maintain a smooth interface between the air and the water. The travel of the interface between graduations (usually for an interval of 2.5 ml.) was timed with an electric timer calibrated in hundredths of a minute. The level of the water reservoir was maintained sufficiently high to allow gravity to refill the burette, but the back pressure due to the water was negligible. The burette and water reservoir were held just below water level in the constant temperature bath, permitting the temperature to be regulated while the graduations could also be read.

II. TEST FLUIDS

Two fluids were used in these experiments. One was a Newtonian oil obtained from the National Bureau of Standards. It is designated viscometer calibrating oil OB, lot 39. The National Bureau of Standards calibrated viscosity is 218.1 poise at 25° C.

The other fluid was a solution of 12 per cent polyisobutylene in Decalin. It was chosen because of its highly viscoelastic properties. The polyisobutylene was obtained from the Enjay Chemical Company and designated as Vistanex L-100. It has a molecular weight of approximately 81,000 - 99,000. The density was estimated at 0.896 gm./cm.³ by

weighing a measured volume of the solution.

The solution was mixed by placing the proper amounts of polymer and solvent in a jar and rolling the jar on a motorized roller while heat was applied from a heat lamp. Approximately three weeks were required to obtain a homogeneous solution.

III. EXPERIMENTAL PROCEDURE

The experimental procedure for obtaining data will be briefly described. The procedure was the same for the Newtonian oil and the viscoelastic polymer solution.

About 300 ml. of fluid was charged into the entrance reservoir of the assembled viscometer. The viscometer was secured to an aluminum frame and flexible leads to the pressure supply and flow measuring system were connected. The apparatus was submerged in the constant temperature bath and about one hour was allowed for the temperature to become constant at 25°C. and for the fluid to relax.

The desired pressure was applied to the entrance reservoir and maintained constant throughout a run. The pressure, the time required for 2.5 ml. of fluid to flow through the capillary, and the temperature were measured and recorded. The pressure was then changed and this procedure repeated. Approximately 20 to 30 runs were needed to cover the desired shear rate range (10^2 to 10^3 sec^{-1}).

The viscometer was then removed from the bath and the capillary replaced by one of another L/R ratio, attached to a cone of a different angle. This procedure was repeated until all combinations of capillary lengths and cone angles had been used.

Modifications of the above procedure were performed by LaNieve (20) in order to determine the effects of reducing the entrance ration to 10:1 and of letting the fluid jet into the air instead of the stagnant fluid. These changes had no effect on the results.

IV. TREATMENT OF DATA

The experimental data from the capillary viscometer consisted of the measured pressure drop, the volumetric flow rate, the dimensions of the capillaries and the dimensions of the cones. This section will describe the necessary corrections to these data and the procedure for obtaining the end effect. Also, the results of the viscosity measurements are presented in this section.

Corrections

An excellent discussion of the sources of error in capillary viscometers and their correction is given by Van Wazer, Lyons, Kim, and Colwell (47). In addition to the errors which they discuss, there are several sources of

error which are peculiar to this viscometer.

The effects of fluid head in the viscometer and of back pressure due to water in the flow measuring device were calculated and found to be negligible.

The effects of viscous heating in the capillary are discussed by Gerrard and Philippoff (15). As will be shown later, the viscosity of oil OB was observed to decrease slightly at the higher shear rates, indicating that viscous heating could be important in experiments. However, at the relatively low pressures reported here, Gerrard and Philippoff observed that the effect of viscous heating on viscosity was only about 3 per cent. Since oil OB has a temperature gradient of about one-third that of the petroleum oil which they used, one would conclude that the effect was much smaller in this experiment.

Corrections were made for the flow in the barrel of the capillary holder for the very pseudoplastic polymer solution. Barrel corrections for the Newtonian oil were calculated and found to be negligible. Equation II-7 was used for these corrections. For flow in the barrel this equation becomes

$$\Delta P(\text{BAR}) = \frac{2L_B}{R_B} k \left(\frac{4Q}{\pi R_B^3} \frac{3n+1}{4n} \right)^n. \quad (\text{IV-1})$$

In a gas-driven viscometer the pressure drop is set

and the flow rate is measured for each capillary. For this reason, it is often necessary to interpolate between data points to obtain the necessary values of pressure drop for each capillary at a constant value of shear rate. This interpolation was carried out graphically by plotting the total pressure drop versus the effective shear rate for each capillary. Smooth curves through the data points were then used for interpolation. The raw capillary data presented in Appendix C were taken from these plots, since data in this form are more useful for calculations.

Evaluation of End Effects

The end effects were evaluated by the method of Philippoff and Gaskins (29), and Bagley (4). This method was discussed in detail in Chapter II. The total pressure drop, $\Delta P(\text{TOT})$, was plotted versus L/R . The end effect, $\Delta P(\text{ENDS})$, was determined from the intercept at $L/R=0$. Figures 6 through 9 are typical of the graphs used to determine the end effects.

If the discrepancies between the bores of the capillaries and cones caused a serious error, it is not evident from these graphs. The data for $\alpha=90^\circ$, where the discrepancies do not exist, show no more scatter than the data for the smaller angles. For $\alpha=10.8^\circ$ and $\alpha=22^\circ$, the end effect was taken as the total pressure drop at $L/R=0$. For $\alpha=46.7^\circ$, the cone had a very short straight section at the

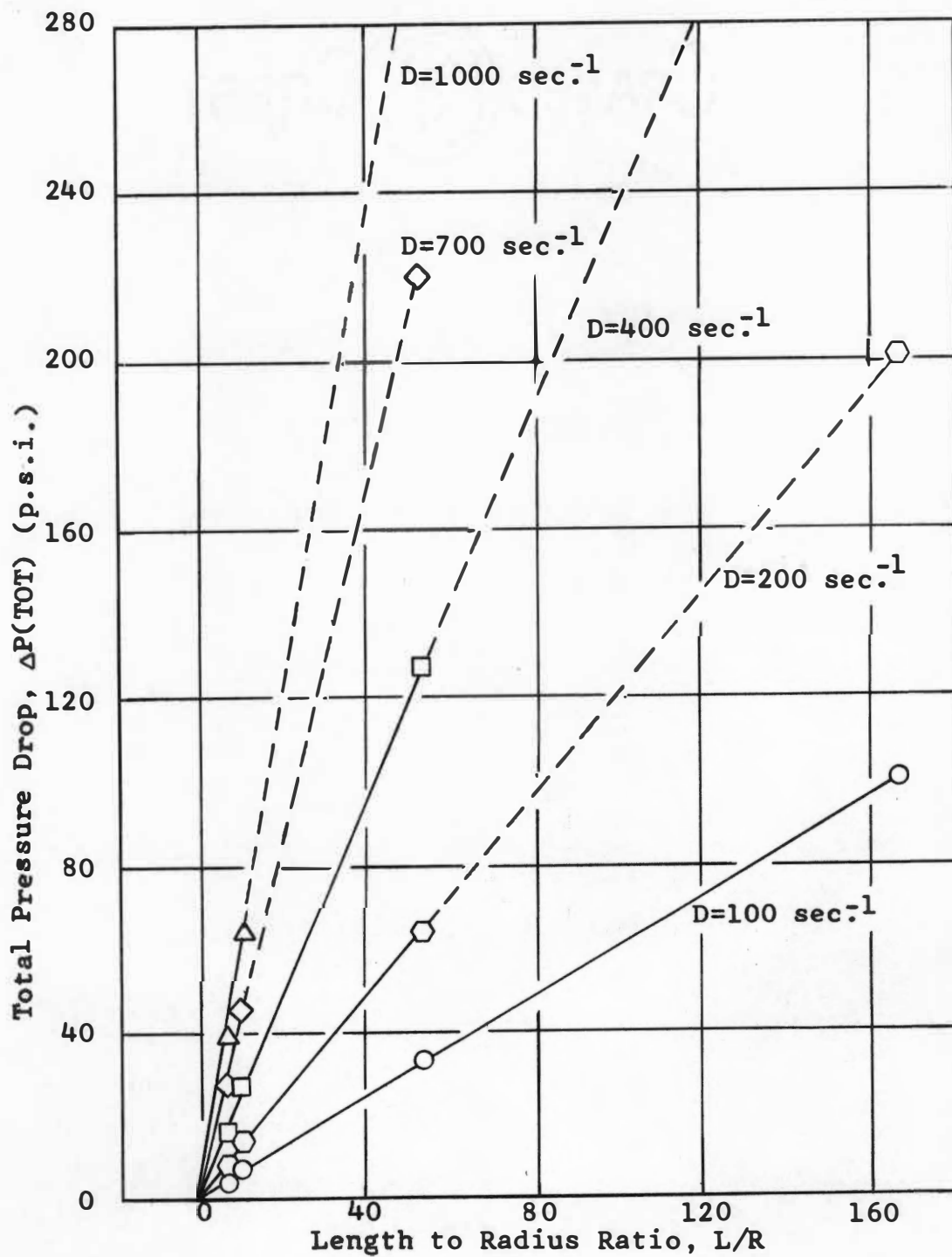


Figure 6. Typical Graph for Evaluating End Effects For Oil OB with $\alpha=90^\circ$.

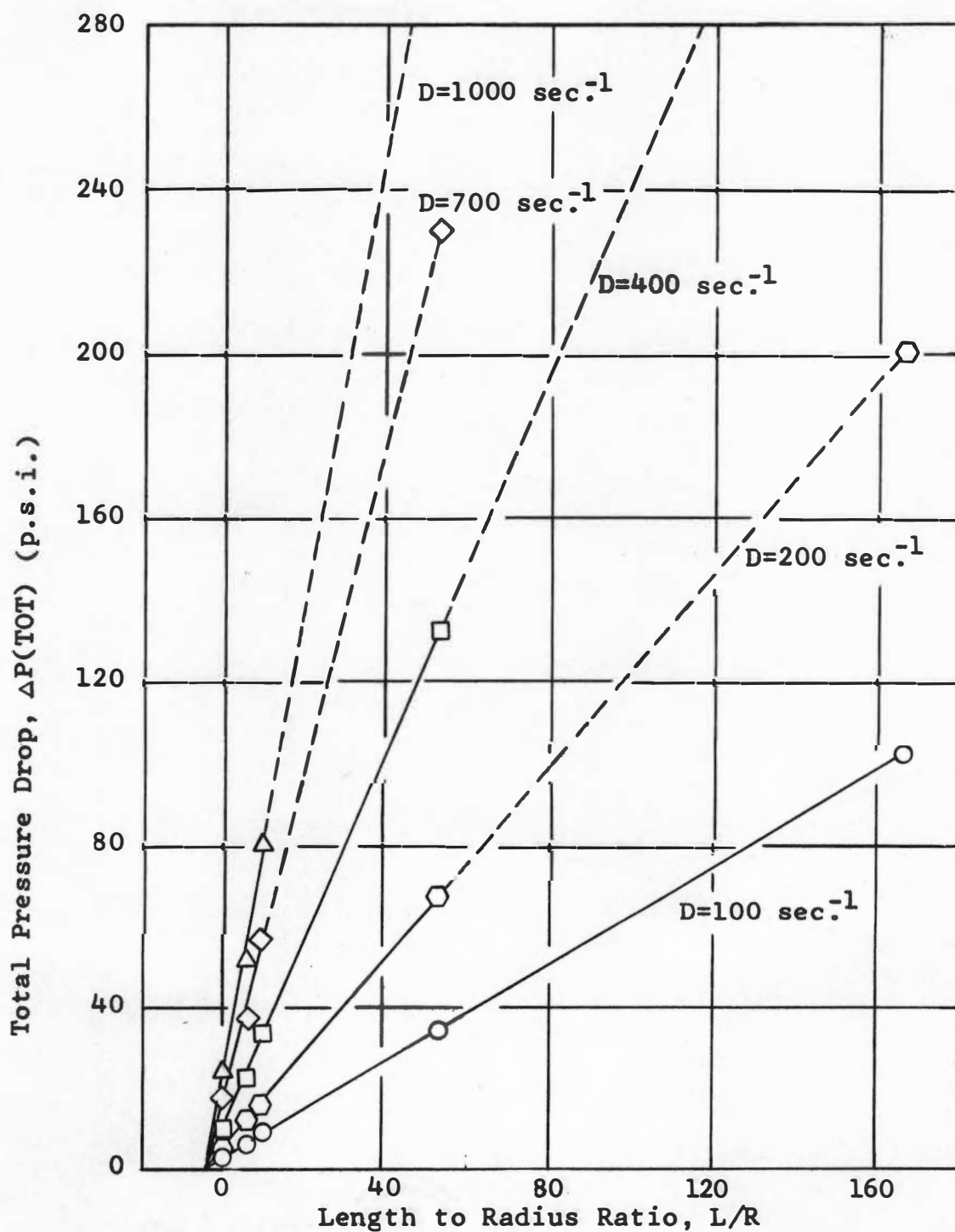


Figure 7. Typical Graph for Evaluating End Effects
For Oil OB with $\alpha=10.8^\circ$.

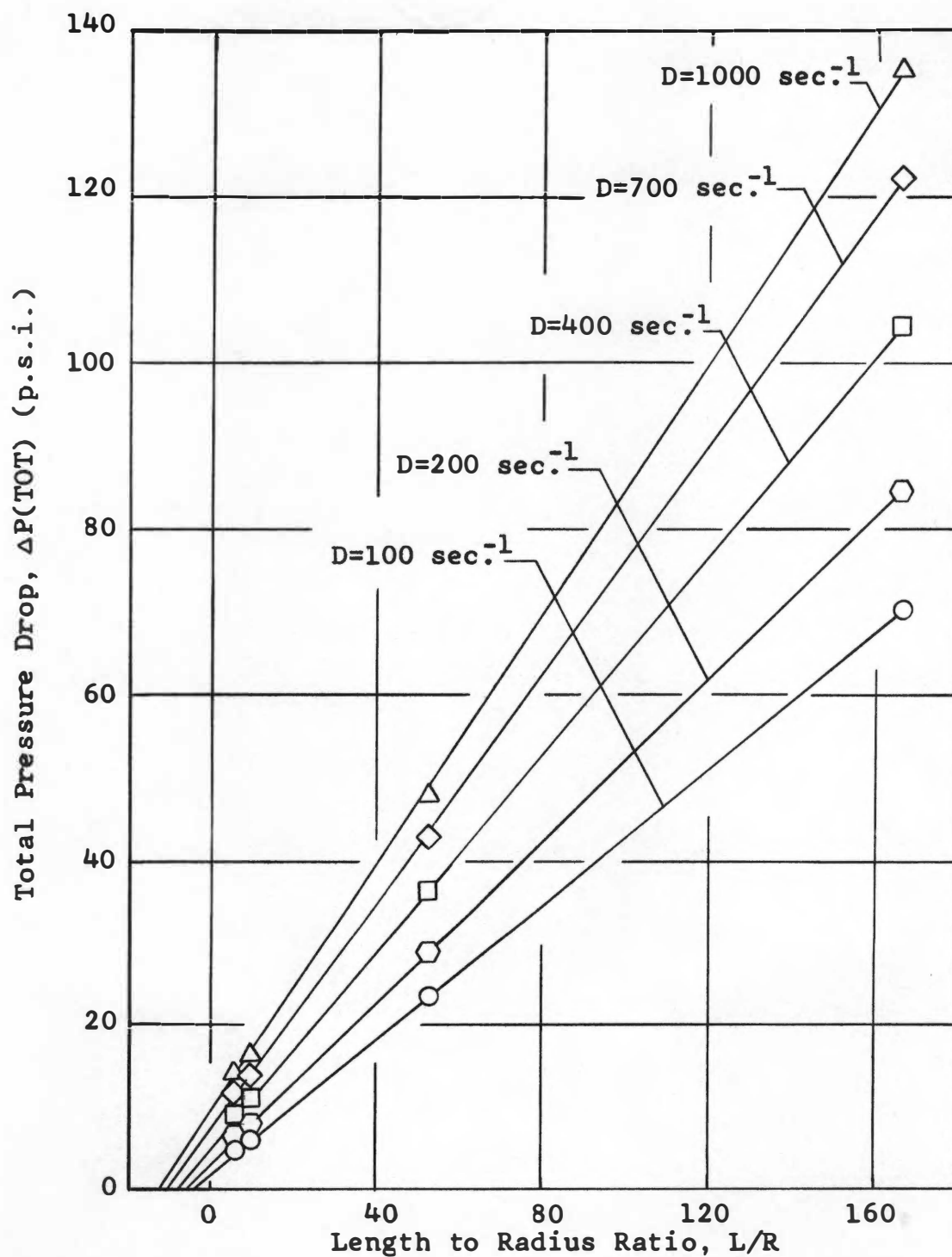


Figure 8. Typical Graph for Evaluating End Effects for 12 Per Cent Polyisobutylene Solution with $\alpha=90^\circ$.

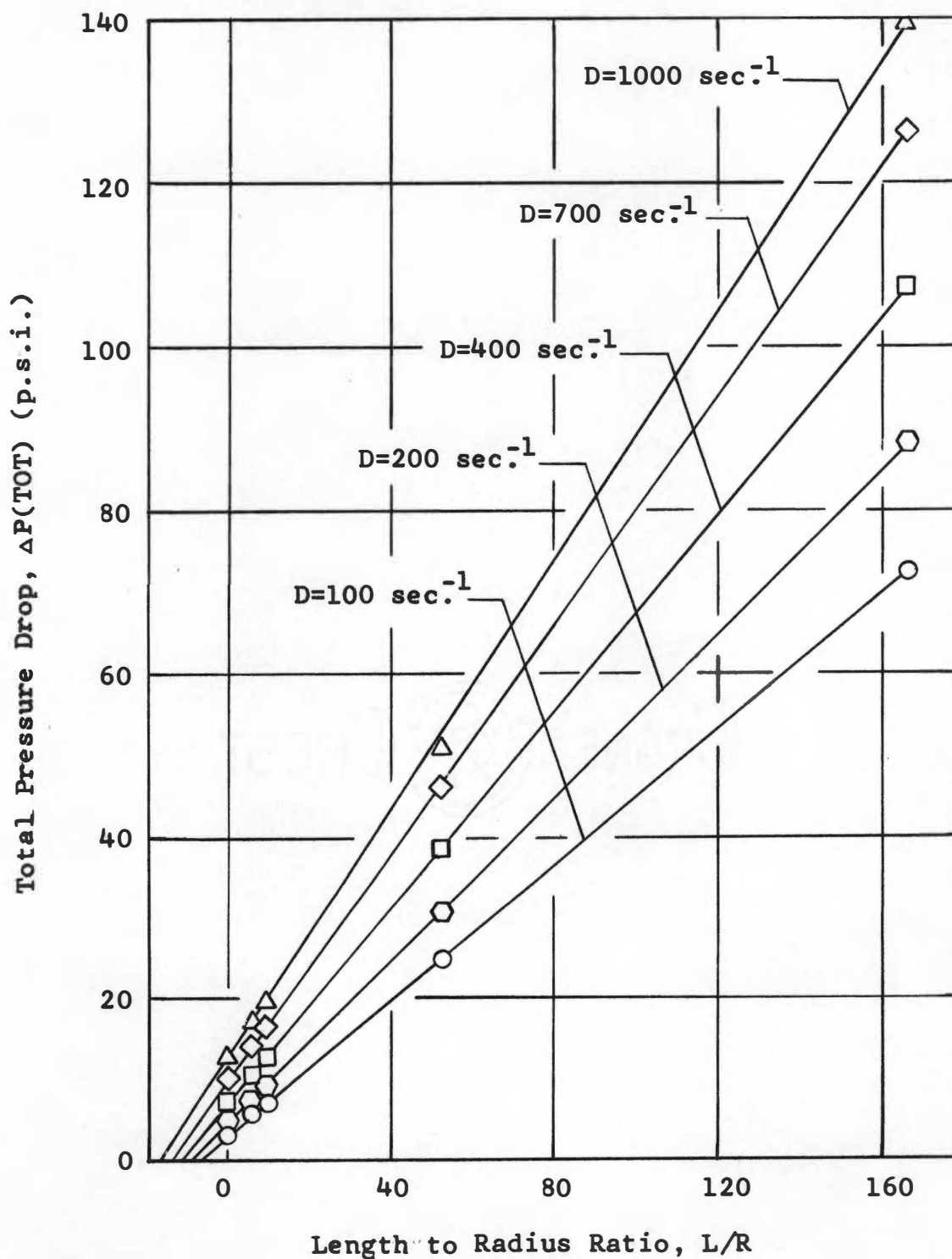


Figure 9. Typical Graph for Evaluating End Effects for 12 Per Cent Polyisobutylene Solution with $\alpha=46.7^\circ$.

small end, necessitating an extrapolation to $L/R = -2.7$.

The linearity of these plots, even at small values of L/R , indicates that the effect of velocity profile development inside the tube is negligible, insofar as pressure drop measurements are concerned. This observation was, of course, predicted by Philippoff and Gaskins (29), and observed by Bagley (4).

Calculation of Viscosity

The data for $\alpha = 90^\circ$ were used to compute the apparent viscosity of the two fluids as a function of shear rate. The apparent viscosity is given by

$$\eta_{app} = \frac{\tau_w}{D}, \quad (IV-2)$$

where τ_w is one-half the slope of the $\Delta P(TOT)$ versus L/R plot, and D is the shear rate at the wall of the tube. A plot of τ_w versus $4Q/\pi R^3$ allows one to determine the power law parameters n and k . Knowing n , D is given by

$$D = \frac{3n+1}{4n} \frac{4Q}{\pi R^3}. \quad (IV-3)$$

The apparent viscosities of the two materials are compared with data from the Weissenberg rheogoniometer in Figure 10. Capillary data for oil OB were lower, while rheogoniometer data were higher than the National Bureau of

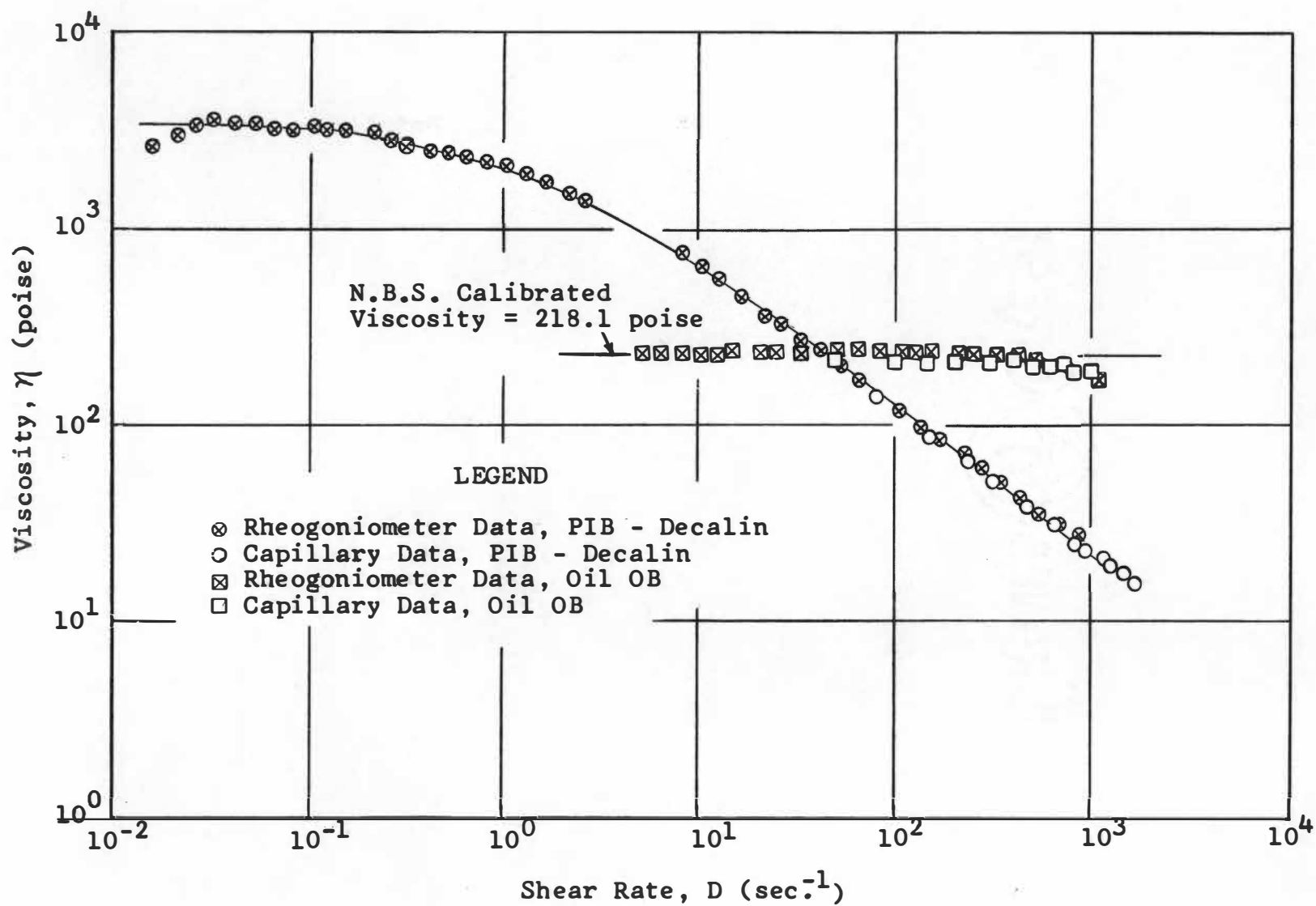


Figure 10. Apparent Viscosities of the Test Fluids.

Standards calibrated value of 218.1 poise at 25°C. The probable reason for these discrepancies is the extreme sensitivity of the oil to temperature. It should be noted that both the capillary data and the rheogoniometer data revealed that oil OB becomes slightly non-Newtonian at high shear rates.

Excellent agreement between the capillary data and the rheogoniometer data was obtained for the polyisobutylene solution. From the data at very low shear rates, the zero-shear viscosity was estimated as:

$$\eta_0 = 3250 \text{ poise.}$$

Viscosity data in the range of shear rates from 10 to 10^3 fit the power law viscosity model where

$$\eta = 3710 D^{-.734}.$$

The power law index, n is 0.266.

The second order fluid parameter, γ , defined by

$$\gamma = \frac{\tau_{11} - \tau_{22}}{D^2} \quad (\text{IV-4})$$

was determined from rheogoniometer measurements for the polyisobutylene solution. It is plotted as a function of shear rate in Figure 11. These data can be fitted to a power law model also, where

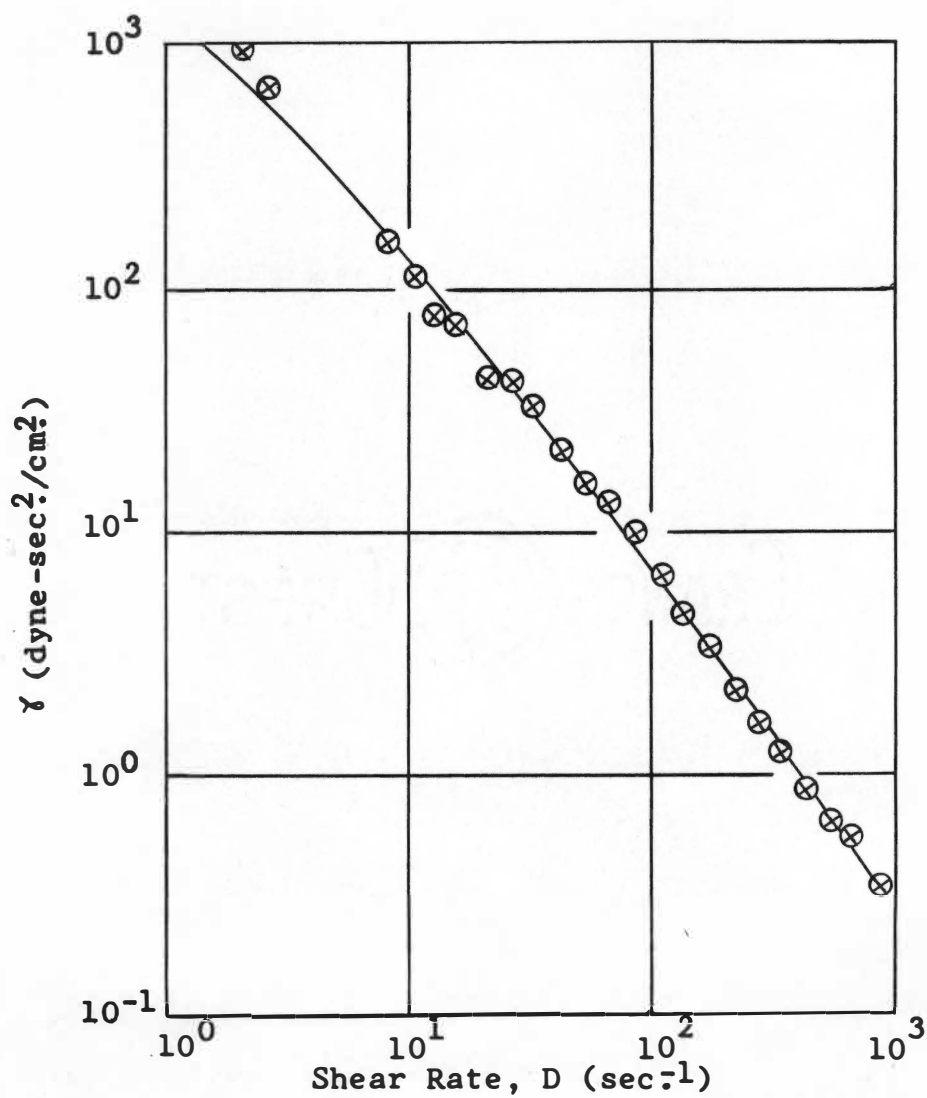


Figure 11. Second Order Parameter, γ , Determined from Rheogoniometer Measurements.

$$\tau_{11} - \tau_{22} = k' D^{2m}.$$

(IV-5)

The parameters k' and $2m$ were evaluated as:

$$k' = 4220 \text{ dyne sec}^{2m}/\text{cm}^2$$

$$2m = 0.61 .$$

The low shear rate value, γ_0 , could not be determined from these data.

CHAPTER V

CORRELATION OF END PRESSURE DROP

In this experiment the only significant contributions to the end pressure drop were the entrance effect upstream and the exit effect downstream of the capillary tube. The entrance pressure drop was of primary interest in this analysis, since the entrance angle, α , was varied, while the exit angle was always 90° . Assuming that the flow is slow enough so that the streamlines are the same at the 90° exit as at the 90° entrance, the entrance pressure drop for any approach angle may be computed very easily:

$$\Delta P(\text{ENT}, \alpha) = \Delta P(\text{ENDS}, \alpha) - \frac{1}{2} \Delta P(\text{ENDS}, 90^\circ). \quad (\text{V-1})$$

The entrance pressure drop was assumed to consist of a viscous effect and an elastic effect, each of which exists as a separate function which one could hope to derive as a limiting solution. The reasonableness of this assumption has been discussed in previous chapters. If this assumption is valid, the elastic contribution to the pressure drop may be computed from the following equation:

$$\Delta P(\text{EL}) = \Delta P(\text{ENT}) - \Delta P(\text{VISC}). \quad (\text{V-2})$$

The viscous contribution may be computed from Equation III-14

for a Newtonian fluid and from Equation III-27 for a non-Newtonian fluid in a conical entrance. For a sharp-edged entrance ($\alpha=90^\circ$) the viscous effect is computed from Weissberg's result, Equation II-18.

I. NEWTONIAN END EFFECTS

The entrance pressure drop is plotted versus shear rate for oil OB in Figure 12. For the 90° entrance the data are compared with Weissberg's prediction. For the conical entrances the data are compared with Oka's result.

The agreement of the data with theory was very good for entrance angles of 90° and 46.7° . For $\alpha=10.8^\circ$ and $\alpha=22^\circ$, however, the theory predicted pressure losses which were significantly lower than were measured.

In using Oka's equation it was assumed that

$$r = R \operatorname{ctn} \alpha. \quad (\text{V-3})$$

This is equivalent to integrating from the tube entrance (point 3' in Figure 2) outward. For Newtonian flow the outer limit, r_1 , may be considered infinite. With these assumptions Equation III-14 becomes

$$\Delta P(\text{ENT}) = \left(\frac{40}{\pi R^3} \right) \frac{\eta}{2 \operatorname{ctn}^3 \alpha (1 - \cos \alpha)^2 (1 + 2 \cos \alpha)}. \quad (\text{V-4})$$

Equation V-4 predicts a minimum pressure drop at the centerline for an angle of about 30.6° . This explains why a

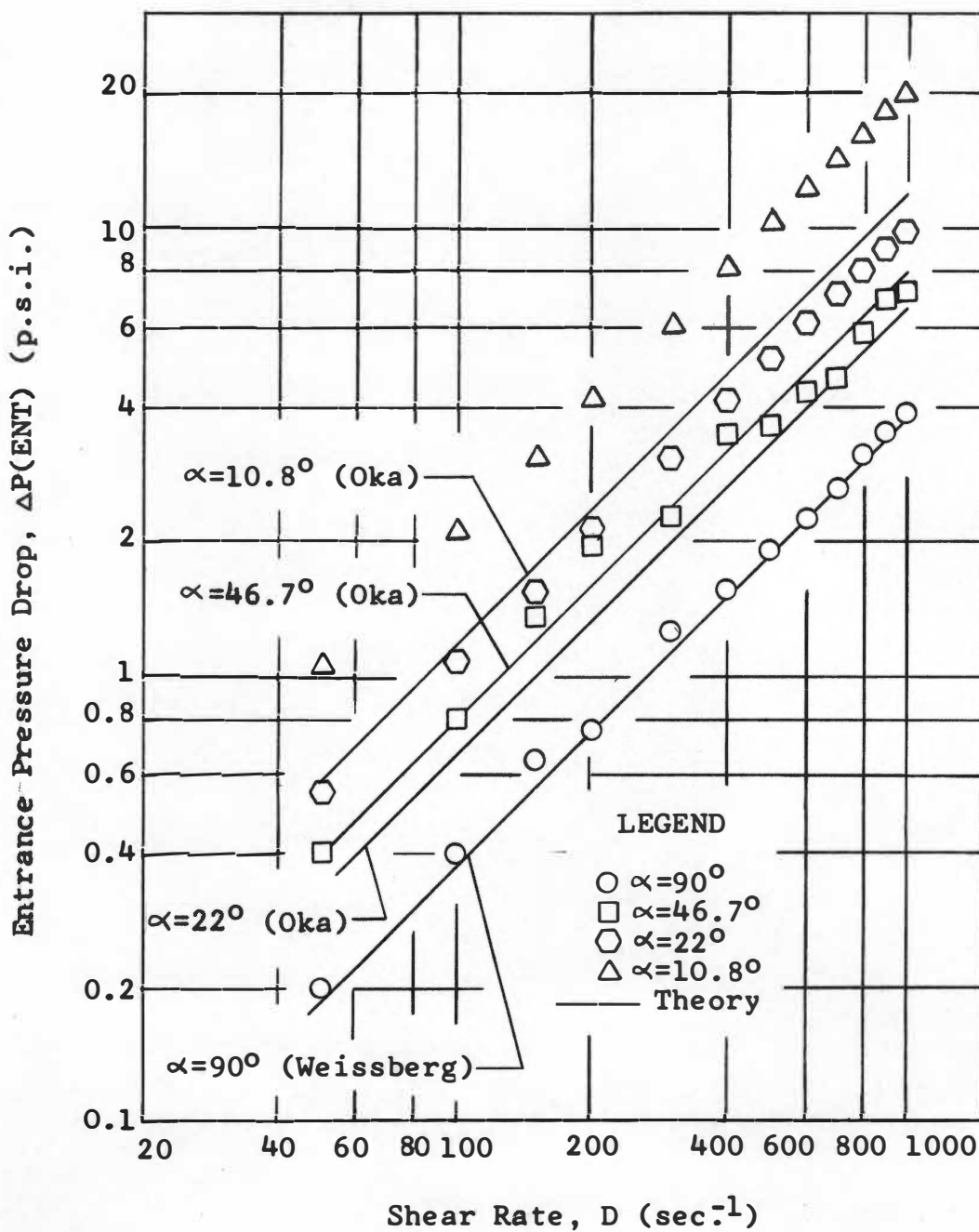


Figure 12. Newtonian Entrance Effects for Oil OB.

lower pressure drop is predicted for $\alpha=22^\circ$ than for $\alpha=46.7^\circ$. It must be explained that this strange behavior is not a result of Oka's solution, but of the way r was chosen to vary with α . The theory could have been forced to agree with the measurements by choosing a smaller value of r , assuming that for small α radial flow is still a good approximation for a short distance after the fluid leaves the cone and enters the tube. However, such action would not have been consistent with the method of determining end effects; i.e., the Philippoff-Gaskins-Bagley analysis evaluates only end effects outside the tube.

Another method of forcing the theory to fit the data is to assume that the fluid channels through the center of the cone, creating an effective angle of approach. This has been observed for flow of certain polymer melts through cones (13, 46). However, the phenomenon seemed to depend on the structural properties of the melt, since it was observed for some materials but not observed for others. If such an approach were taken here, agreement with Equation V-4 could be obtained by assuming effective angles of 6° and 14° for actual angles of 10.8° and 22° , respectively. For an actual angle of 46.7° the effective angle may be either 46° or 21° , Equation V-4 being double-valued in α .

The excess pressure drop observed for the small angle cones could be explained by arguing that the radial flow

assumption becomes invalid near the exit of the cone. The more complex flow in this region would give rise to more energy dissipation, hence a higher pressure drop. However, one would intuitively expect this effect to be more pronounced for large entrance angles instead of the reverse, as one would conclude from this study.

From the data presented here, one can say that Oka's solution gives qualitative agreement with experiment. Probably, quantitative agreement could be obtained if one knew how to define the distance r for the experiment. Clearly, a more complete analysis is needed for the composite problem of flow from a conical entrance into a tube.

II. VISCOELASTIC END EFFECTS

Figure 13 shows the entrance pressure drop as a function of shear rate for the viscoelastic polyisobutylene solution. It can be seen that this plot is not strongly dependent on the entrance angle except for the case of $\alpha=10.8^\circ$ at low shear rates. The reason for this is illustrated in Figures 14 through 17, where the individual terms which make up the end effect are plotted.

The viscous entrance pressure drops in the conical entrances were calculated from Equation III-27. The limiting values of radius were $r = R \cot \alpha$ and $r_1 = R \cot \alpha + 0.754$. This is equivalent to integrating from

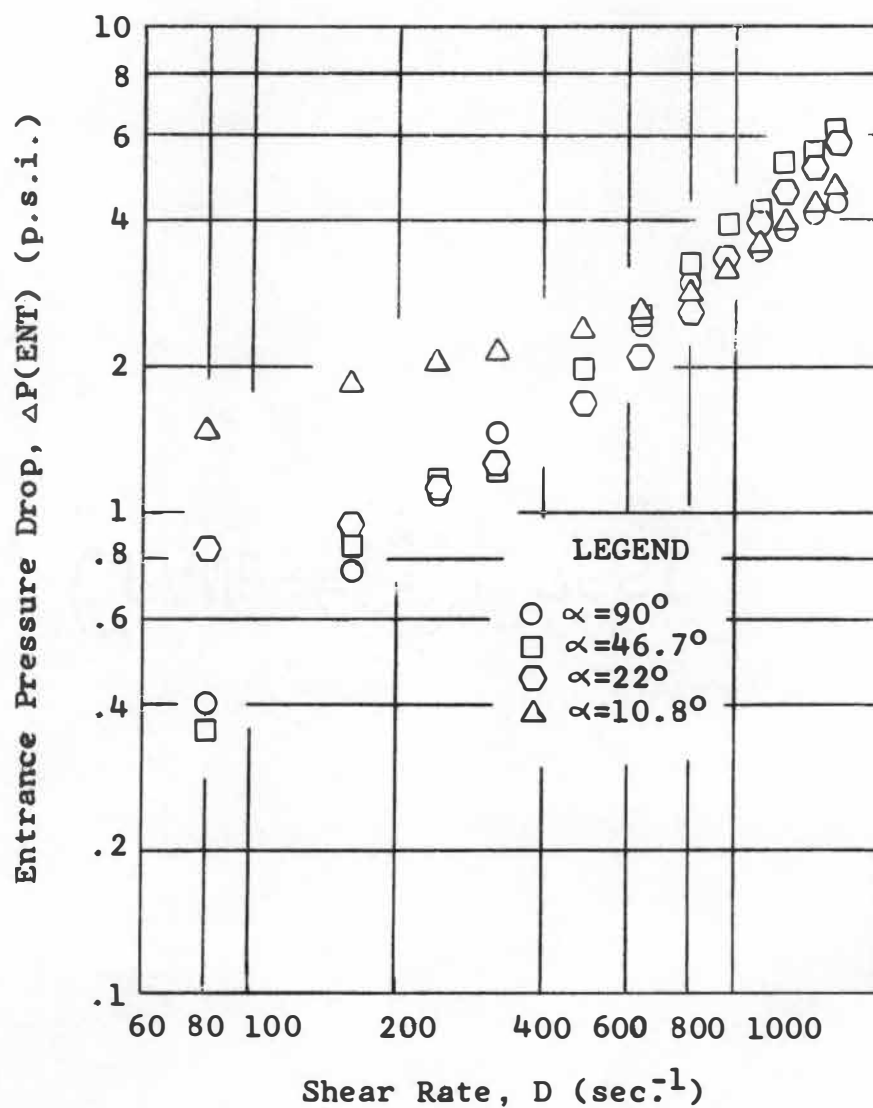


Figure 13. Entrance Pressure Drop as a Function of Shear Rate for PIB Solution.

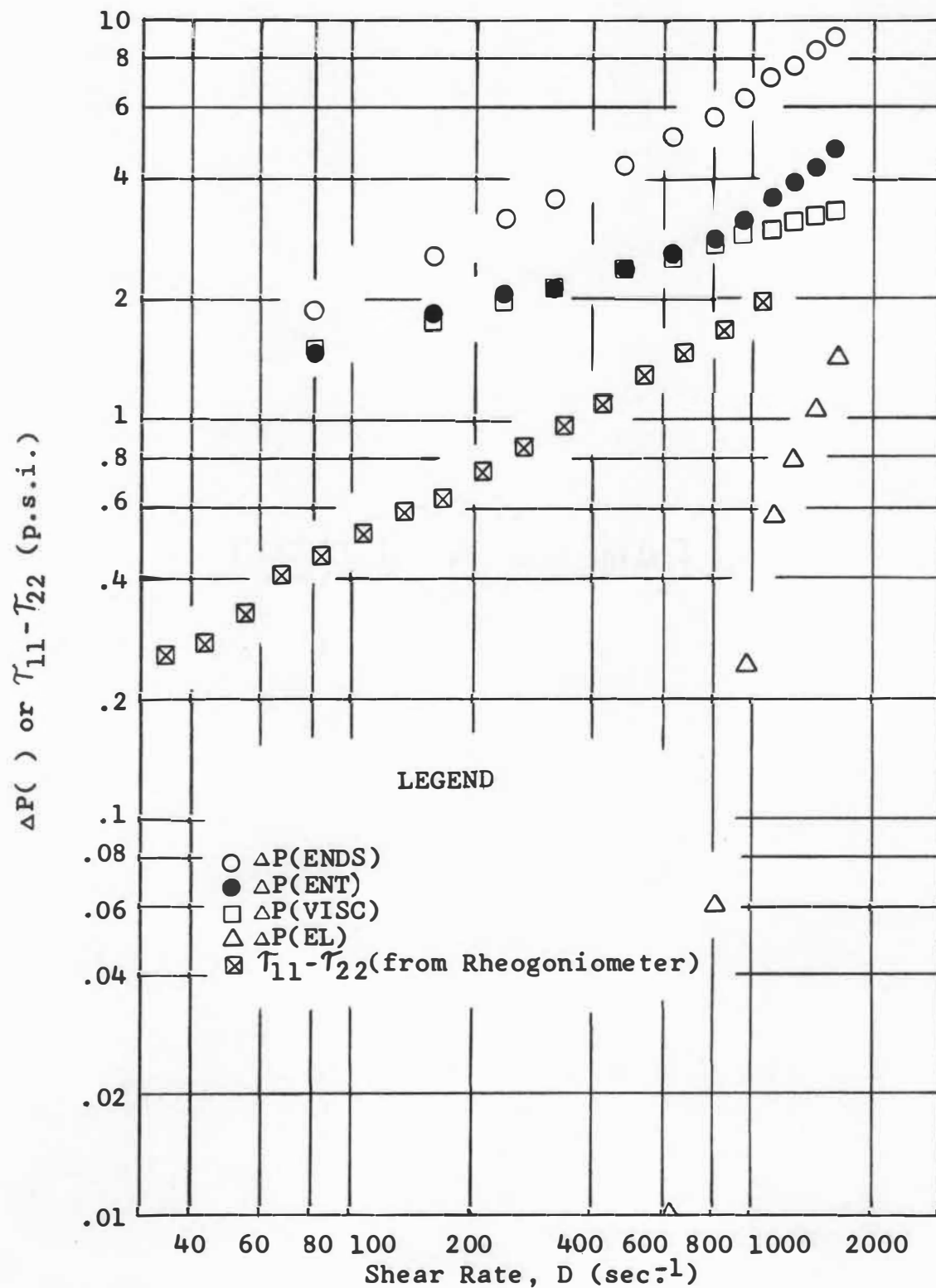


Figure 14. Contributions to the End Pressure Drop for PIB Solution in a Cone with $\alpha=10.8^\circ$.

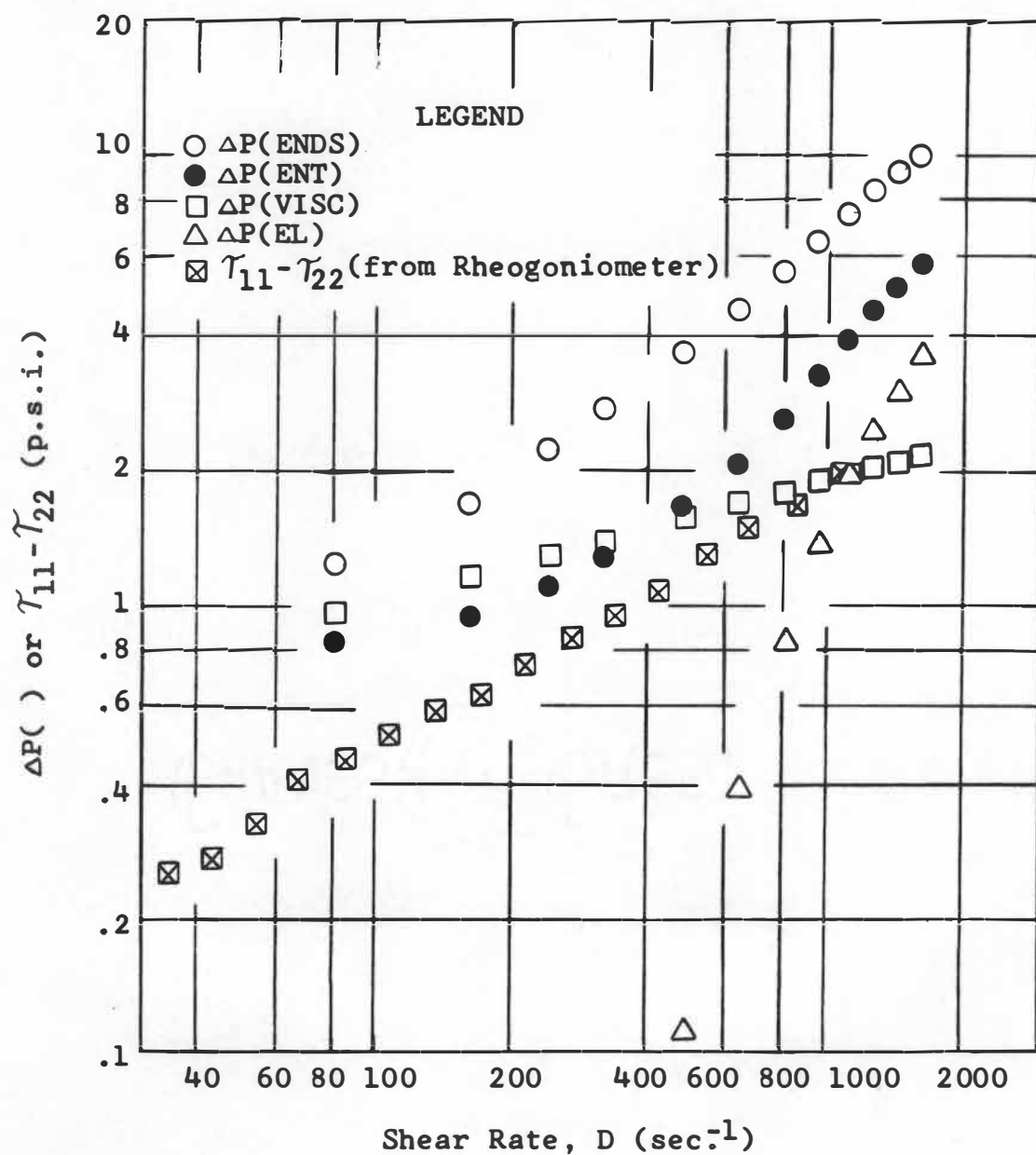


Figure 15. Contributions to End Pressure Drop for PIB Solution in a Cone with $\alpha=22^\circ$.

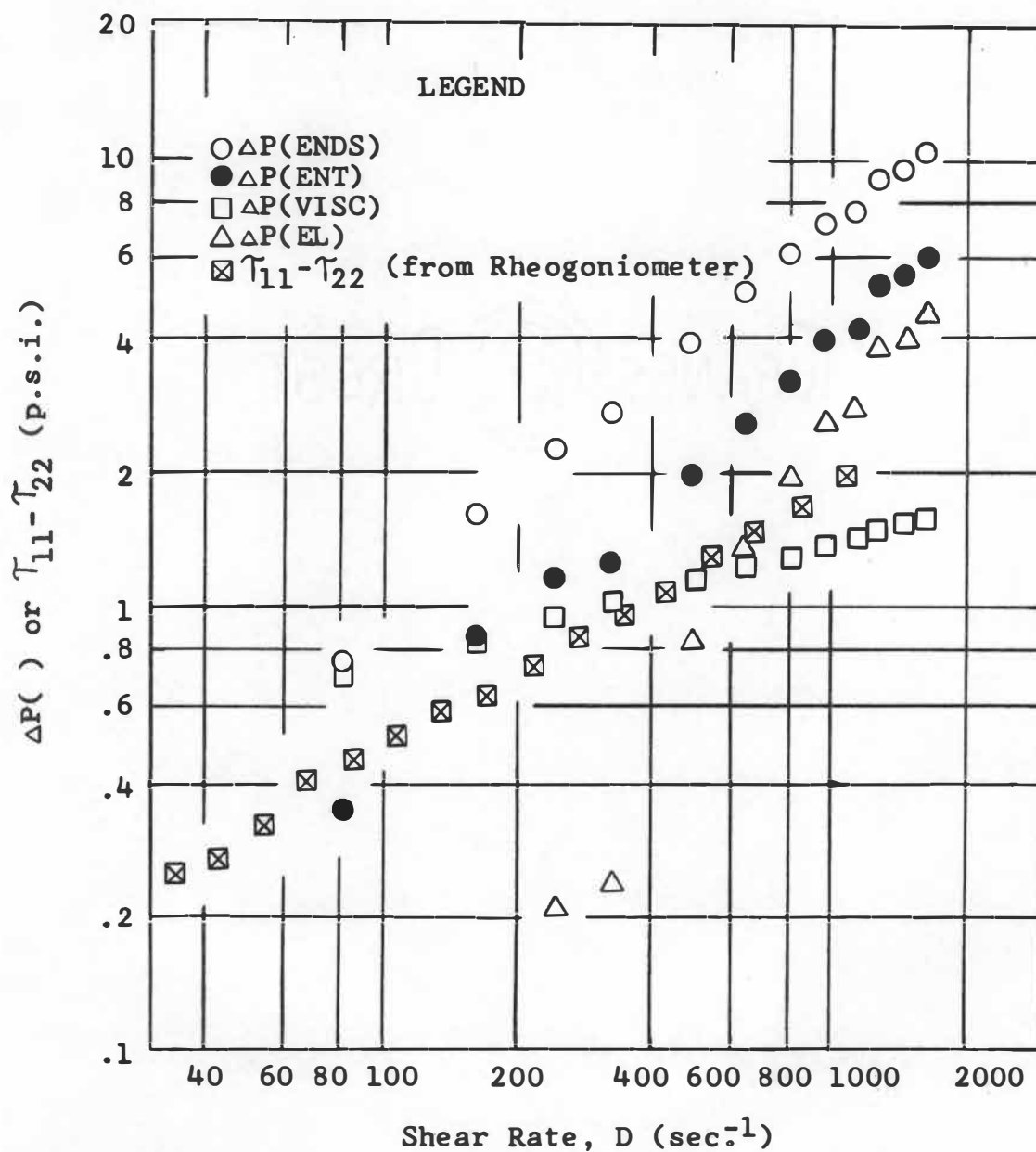


Figure 16. Contributions to End Pressure Drop for PIB Solution in Cone with $\alpha=46.7^\circ$.

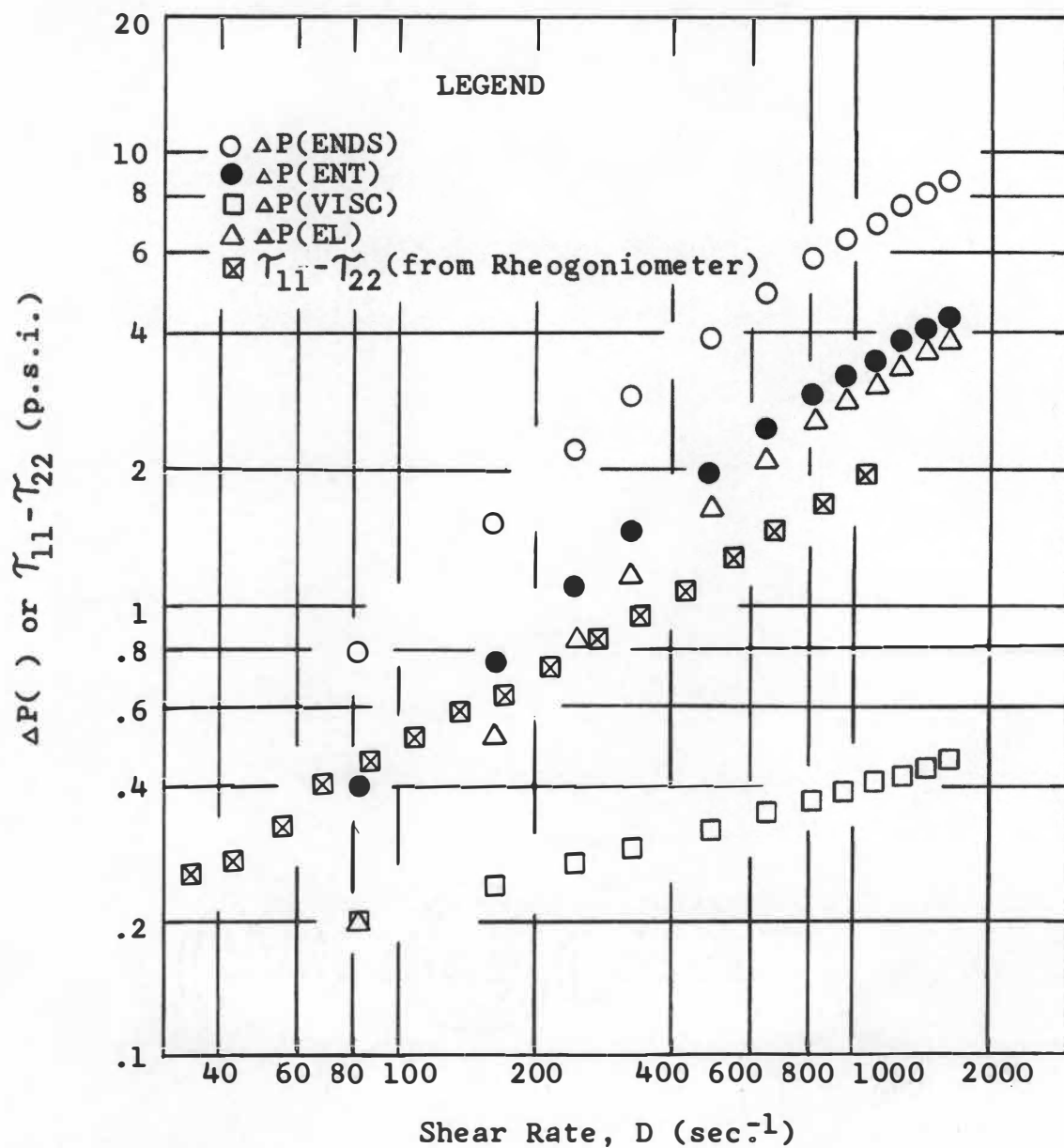


Figure 17. Contributions to End Pressure Drop for PIB Solution in Sharp-Edged (90°) Entrance.

the tube entrance (point 3' in Figure 2, page 15) to the cone entrance (point 2 in Figure 2, page 15). Note that the outer limit, r , cannot be assumed infinite for the case of a very pseudoplastic fluid.

For the 90° entrance, Weissberg's result was used to calculate the viscous pressure drop. It was slightly modified to the following form:

$$\Delta P(\text{VISC}) = \frac{3\pi}{4} \eta_{\text{app}} D, \quad (\text{V-5})$$

where

$$D = \frac{4Q}{\pi R^3} \frac{3n+1}{4n}$$

and η_{app} is the apparent viscosity corresponding to a shear rate, D . This approximation is questionable since one would expect that, for a very pseudoplastic fluid, there would be a significant viscous pressure drop at a greater distance from the tube than would be expected for a Newtonian fluid. However, when this method was used by LaNieve and compared with an analysis of Tomita (44) for a power law fluid, the results did not depend on which analysis was used. The probable reason is that the elastic effect is so predominant for the case of the 90° entrance that the viscous effect could be neglected altogether without serious error.

For the smaller entrance angles ($\alpha=10.8^\circ$, 22°) it is

seen that the viscous contribution is completely dominant at the lower shear rates. However, as the shear rate increases, the elastic effect becomes more significant. At the highest shear rate ($D=1610 \text{ sec}^{-1}$) the elastic effect comprises 30 per cent of the total pressure drop for the 10.8° cone and 63 per cent of the total pressure drop for the 22° cone. As the entrance angle is increased, the viscous effect becomes less important. At $\alpha=90^\circ$ the elastic effect comprises 50 per cent of the total pressure drop at the lowest shear rate ($D=80.6 \text{ sec}^{-1}$) and over 90 per cent of the total pressure drop at the highest shear rate ($D=1610 \text{ sec}^{-1}$). This should be expected, since at the larger angles the fluid accelerates more rapidly, thus increasing the normal stress; while at the smaller angles there is more restriction at the walls, hence more viscous force acting on the fluid.

Comparing the elastic pressure drop with the normal stress difference, as measured with the Weissenberg rheogoniometer, one observes that the two curves have the same slopes at high shear rates for the 90° entrance. This is in agreement with the observations of LaNieve (20). For $\alpha=46.7^\circ$ the slope of the elastic pressure drop curve seemed to be approaching that of the normal stress curve, but for the smaller angles no relationship between the two quantities was observed.

The elastic contribution to entrance pressure drop is

plotted as a function of shear rate, D , in Figure 18. The effect of entrance angle on the elastic contribution is seen to be quite pronounced at low shear rates, the elastic effect being nil for small entrance angles. At the higher shear rates there appears to be little dependence on angle, except for the 10.8° entrance where the elastic contribution is still relatively small at high shear rates.

The correlation between elastic effects is considerably improved if the Deborah number, N_{Deb} , is used as the abscissa instead of the shear rate. Figure 19 is a plot of the elastic pressure drop versus $F_0 = N_{Deb}/s^*$. Since only one fluid is being considered, it is not necessary to specify the characteristic material time in order to call this a Deborah number correlation. Note that as the entrance angle increases, the Deborah number corresponding to a particular shear rate in the tube also increases. This means that the residence time of a fluid particle in the accelerating flow field is inversely proportional to the magnitude of the entrance angle. One would expect, therefore, that for the 90° entrance, the fluid moves very slowly until it enters the region very close to the entrance where it is suddenly accelerated.

The two lines in Figure 19 indicate slopes of 2 and 2m. For the larger angles the data agree with the predictions discussed in Chapter III; i.e., for small Deborah

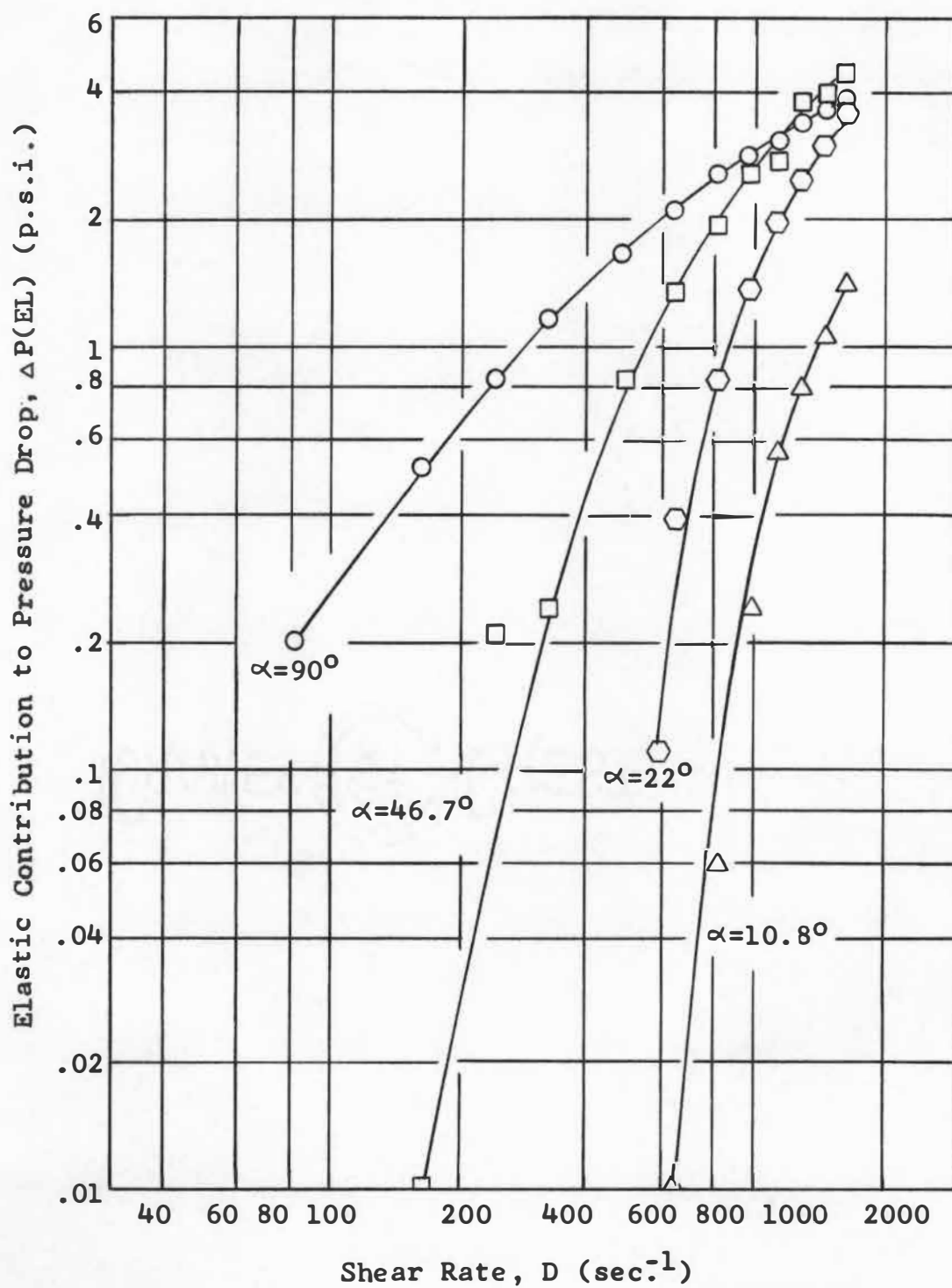


Figure 18. Elastic Contribution to Pressure Drop for PIB Solution in Conical Entrance Region.

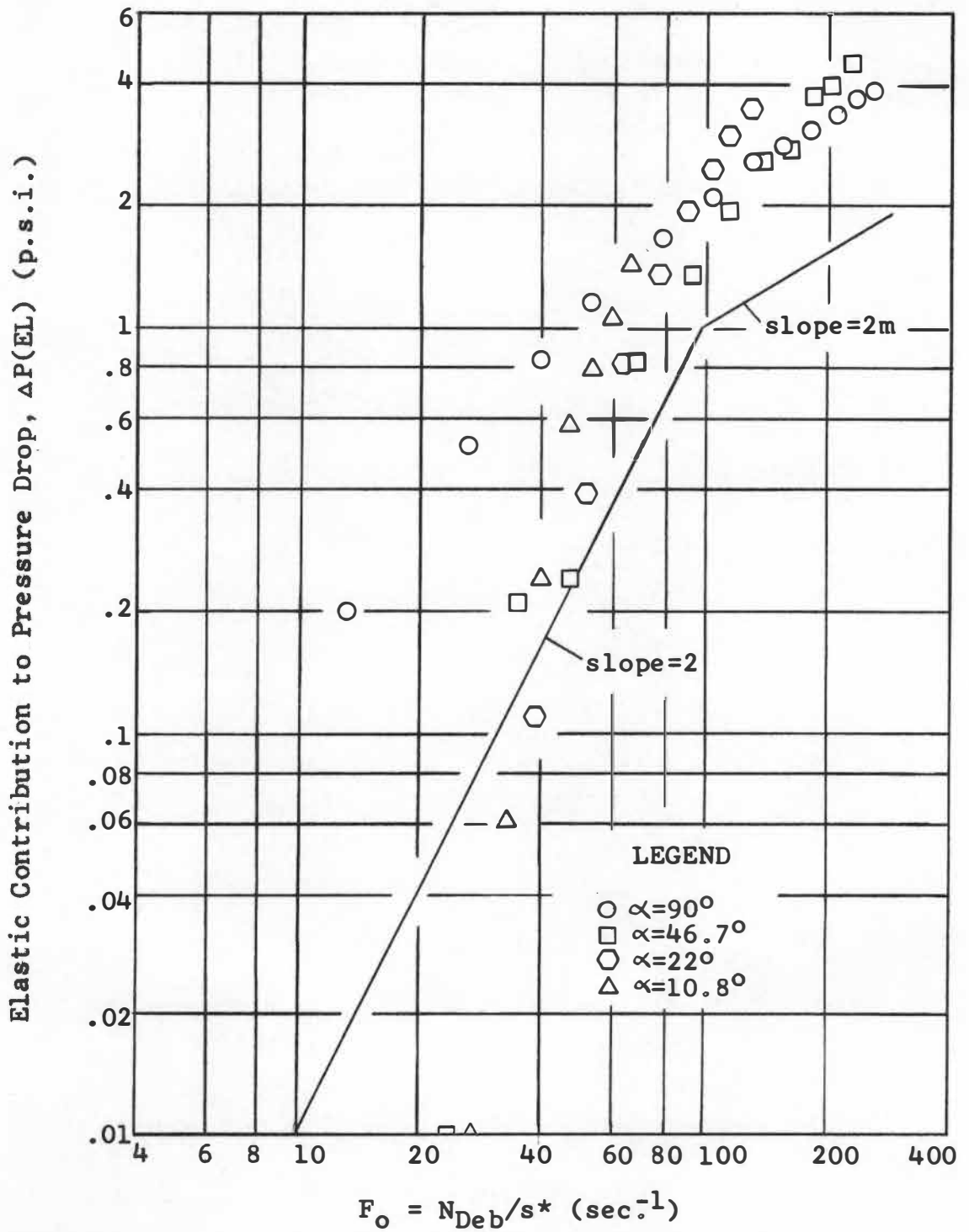


Figure 19. Elastic Contribution to Pressure Drop as Correlated by the Deborah Number.

numbers the elastic contribution is approximately a function of the Deborah number squared, and at high Deborah numbers the elastic contribution is approximately a function of the Deborah number to the $2m$ power. For the smaller angles the slope is somewhat greater than 2. This may be the result of errors introduced by the method of determining end effects; i.e., subtracting small numbers of comparable magnitude.

This correlation is as good as could be reasonably expected for the conical entrances. For the sharp-edged entrance the curve exhibits a slightly different character, indicating that the Deborah number may not be properly defined. Actually this should not be unexpected since the definition of the Deborah number used here assumes a radial flow field. From examination of Weissberg's velocity profile, Equation II-16, and the oblate spheroidal coordinate system, Figure 1, page 12, one can see that the flow field is not radial in the region near the entrance where most of the energy dissipation is presumed to take place. Figure 20 shows the entrance pressure drop as a function of $F_0 = N_{Deb}/s^*$. This correlation is very dependent on the entrance angle, indicating that the viscous contribution is dependent on α as well as $F_0 s^*$. This could have been seen, of course, from an examination of Equations III-25 and III-27. Figures 19 and 20 seem to support the conclusion, drawn from the work of LaNieve (20) and Adams (2), that the entrance pressure drop could be calculated by simply adding

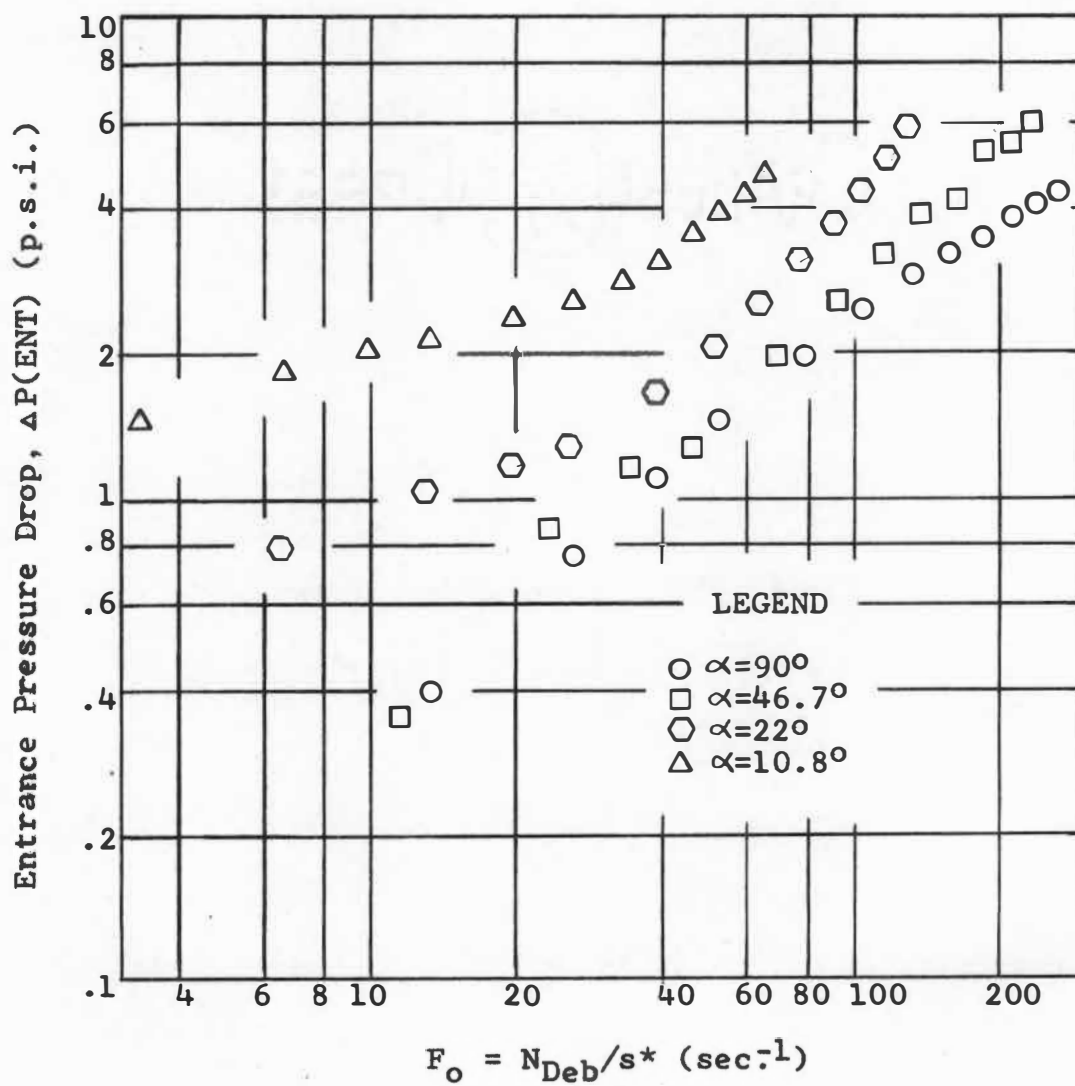


Figure 20. Entrance Pressure Drop as a Function of the Deborah Number.

limiting solutions for viscous effects and elastic effects if these solutions were known. Of course, if the elastic effect could be calculated from a particular constitutive equation, the viscous effect would almost surely be obtainable from the same constitutive equation. The purpose of calculating the viscous effect from a different model in this study was to determine which dimensionless groups the elastic effect depends upon.

III. SUMMARY OF CORRELATIONS

In summary, the data were correlated with an equation of the form:

$$\Delta P(\text{ENT}) = \Delta P(\text{VISC}) + \Delta P(\text{EL}), \quad (\text{V-6})$$

where

$$\Delta P(\text{VISC}) = \frac{2k}{2n \sin \alpha} \left[\frac{3Q \sin (3n+1)}{4\pi n (1-\cos \alpha)^2 (1+2\cos \alpha)} \right]^n \cdot \left[\frac{1}{(R \cot \alpha)^{3n}} - \frac{1}{(R \cot \alpha + .754)^{3n}} \right] \quad (\text{V-7})$$

for $\alpha = 10.8^\circ$, 22° , and 46.7° , and

$$\Delta P(\text{VISC}) = \frac{3\pi}{4} \eta_{\text{app}} D \quad (\text{V-8})$$

for $\alpha = 90^\circ$. Equations V-7 and V-8 reduce properly to Oka's solution and Weissberg's solution respectively for Newtonian

flow ($n=1$).

The elastic term,

$$\Delta P(EL) = \phi_2(F_0 s^*), \quad (V-9)$$

has remained undefined, although it has been shown that

$$\phi_2 \sim (N_{Deb})^b \quad (V-10)$$

where $b \approx 2m$ for high Deborah numbers and $b \approx 2$ for low Deborah numbers. The function F_0 was defined as

$$F_0 = \frac{40}{\pi R^3} \left(\frac{3n+1}{n+1} \right) \frac{3}{16} \frac{\sin^5 \alpha}{(1-\cos \alpha)^2 (1+2\cos \alpha)}. \quad (V-11)$$

CHAPTER VI

CONCLUSIONS AND RECOMMENDATIONS

I. CONCLUSIONS

The following conclusions were made as a result of this investigation:

1. For a Newtonian fluid in a converging cone, Oka's creeping flow solution is of the correct form to predict viscous pressure drop effects. It gives good quantitative agreement for $\alpha=46.7^\circ$ and would probably do so at the smaller angles if the boundaries of the radial flow field were correctly specified. An adaptation of this solution to the power law viscosity model provides a satisfactory prediction of non-Newtonian viscous effects. Weissberg's solution gives excellent agreement with experimental data for Newtonian flow in a 90° entrance.

2. For viscoelastic flow in a converging cone, the data were correlated semi-empirically using an equation that involved a viscous term and an elastic term. Adam's treatment of flow between non-parallel plates showed that the pressure drop consists of a viscous term (involving a zero shear viscosity) plus nonlinear terms, although explicit nonlinear viscosity terms could not be identified. The

present author's treatment of the analogous problem in a cone identified a group which can be thought of as a dimensionless normal deformation rate. It was assumed that this group characterizes the elastic pressure drop, and that a nonlinear viscous term characterizes the viscous pressure drop.

3. The above-mentioned dimensionless group may also be thought of as a Deborah number, since the normal deformation rate has the units of time and is related to the residence time of the fluid in the accelerating flow field. This definition of the Deborah number is reasonable in that it increases as the entrance angle to the capillary increases, and therefore as the acceleration of the fluid increases. It is not claimed to be a unique definition of the Deborah number.

4. The elastic pressure drop in the conical entrance region correlates reasonably well with the Deborah number. It was found that

$$\Delta P(EL) \sim N_{Deb}^b,$$

where $b \approx 2$ for small values of the Deborah number, and $b \approx 2m$ for large values of the Deborah number. The quantity $2m$ is the slope of the $\log(\tau_{11} - \tau_{22})$ versus $\log D$ plot.

5. The elastic effect on pressure drop is insignificant for the case of small entrance angle and low shear rate.

As the shear rate is increased (approaching 10^3 sec^{-1}) the elastic effect becomes of increasing importance. For large entrance angles the viscous effect tends to be unimportant compared to the elastic effect.

6. The constitutive theory of Bernstein, Kearsley, and Zapas is too complex mathematically for direct analytical solution of the conical flow problem. It does, however, predict a useful dimensionless group, the Deborah number discussed above, for correlating elastic pressure drop effects.

II. RECOMMENDATIONS

The following recommendations are made for further study of flow in the entrance region of a capillary:

1. LaNieve's (20) data for viscoelastic flow in a sharp-edged entrance should be correlated with the Deborah number. Such a correlation would involve accurate determination of the material time constant, s^* , from dynamic data (as discussed in Appendix A) or by some more direct method. From Adams' (2) results one would expect that the correlation would take the form of a plot of $\Delta P(\text{EL})/(\gamma_0/s^*)$ versus N_{Deb} .

2. A visual or photographic study of accelerating flow fields should be made in order to determine the flow lines and velocity profiles, and in particular, the region

where radial flow is a good approximation. Such a study should include Newtonian, inelastic non-Newtonian, and viscoelastic fluids.

3. An analysis of the 90° entrance problem should be made using a simple viscosity model such as the power law.

4. Experiments such as those reported here should be performed for a variety of fluids, including some fluids which exhibit negligible elastic effects. In this connection, a more rigorous analysis of non-Newtonian viscous flow in a cone, in which the velocity profile is not pre-assumed, would be quite useful.

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APPENDICES

APPENDIX A

TREATMENT OF CONICAL FLOW PROBLEM USING BKZ THEORY

An attempt was made to solve the problem of the flow of a viscoelastic fluid in a converging cone using the constitutive theory of Bernstein, Kearsley, and Zapas (6, 7, 8). The theory is briefly stated below, and a description is given of the rigorous approach to the problem. A partial solution is given using "second order" approximations. Although these approximations are not appropriate to the present problem, they do enable one to identify a useful dimensionless group which arises naturally from the BKZ theory.

I. STATEMENT OF BKZ THEORY

The theory of Bernstein, Kearsley, and Zapas is modeled after elastic theory of solid mechanics. For incompressible, isothermal flows it can be stated as follows:

$$P_{ij} = -Pg_{ij} - 2 \int_0^\infty \left[\frac{\partial^2 W}{\partial I \partial s} C_{ij}^{-1}(t,s) - \frac{\partial^2 W}{\partial II \partial s} C_{ij}(t,s) \right] ds, \quad (A-1)$$

where

$$C_{ij}^{-1} = g^{ab} g_{ki} g_{lj} \frac{\partial x^k}{\partial X^a} \frac{\partial x^l}{\partial X^b}$$

relative Finger strain tensor ,

$$C_{ij} = g_{ab} \frac{\partial X^a}{\partial x^i} \frac{\partial X^b}{\partial x^j}$$

relative Cauchy strain tensor ,

s = backward running time ($s=0$ now),

x^i = coordinates of points at time t ,

X^i = coordinates of points at earlier time $t-s$,

$W = W(I, II, s)$, an unspecified scalar function,

and

I, II = scalar invariants of C_{ij}^{-1} .

The function W is analogous in a mathematical sense to the strain energy function of solid elastic theory. However, one should not try to associate the physical interpretation of strain energy with the scalar W , because W is a decaying function of time whereas the strain energy is independent of time.

Although W is unspecified in the BKZ theory, Zapas has suggested a heuristic form of the function which can be stated as follows (50):

$$W(I, II, s) = \frac{\alpha(s)}{2} (I-3)^2 + \frac{3}{4} m(s) \ln \frac{I+II-3}{3} + 7 \left[\frac{3}{4} m(s) - C(s) \right] \ln \frac{I+6}{II+6} + C(s) (I-3), \quad (A-2)$$

where $m(s)$, $C(s)$, and $\alpha(s)$ are decaying functions of time.

Depending on the choice of $m(s)$, $C(s)$, and $\alpha(s)$, a variety of fluid behavior can be predicted. The function $C(s)$ allows description of a high shear rate plateau in the viscosity curve. The function $\alpha(s)$ allows prediction of shear-thickening (dilatant) behavior. The function $m(s)$ allows prediction of shear-thinning (pseudoplastic) behavior, which is characteristic of polymers. It must be emphasized that Equation A-2 is not in any sense unique. Several forms for W have been suggested, any of which might have the properties needed to describe viscoelastic flow phenomena.

A special case of the BKZ theory will be considered, where $\alpha(s)=C(s)=0$. This special case is valid for polymer solutions only at shear rates below the upper Newtonian region. Although Zapas does not explicitly choose the form of $m(s)$, a series of exponentials is convenient and reasonable. The function will then reduce to the classical spectrum of Maxwell elements in linear viscoelastic deformations:

$$m(s) = \sum_{i=1}^{\infty} G_i e^{-s/\lambda_i} \equiv \int_{-\infty}^{\infty} H(\lambda) e^{-s/\lambda} d \ln \lambda. \quad (A-3)$$

or

$$-m'(s) = \sum_{i=1}^{\infty} \frac{G_i}{\lambda_i} e^{-s/\lambda_i} \equiv \int_{-\infty}^{\infty} \frac{H(\lambda)}{\lambda} e^{-s/\lambda} d \ln \lambda. \quad (A-4)$$

Use of either the explicit or the continuous forms of Equations A-3 and A-4 in conjunction with Equation A-2 makes the stress equation intractable.

Adams (2) was motivated to use the results of Doughty's dynamic experiments (14) to compute $-m'(s)$ numerically. He used values of $H(\lambda)$ versus λ to integrate Equation A-4 numerically, then fitted the results empirically to obtain the following form for 12 per cent polystyrene in Aroclor:

$$-m'(s) = \frac{A}{(1+bs)^2}, \quad 0 < s < s^*, \quad (\text{A-5})$$

where A has units of stress/time, b has units of reciprocal time, and s^* is a maximum integration time, chosen so that the expression for viscosity in the simple shear case integrates to the correct simple shear value, η_0 . The parameters A , B , and s^* can be related to the parameters η_0 and γ_0 for simple shear flow (see Adams (2)), giving three independent material parameters η_0 , γ_0 , and s^* .

Using Adams' empirical form for $-m'(s)$, W is given by:

$$W(I, II, s) = -\frac{3}{4} \frac{As}{(1+bs)} \left[\ln \frac{I+II-3}{3} + 7 \ln \frac{I+6}{II+6} \right]. \quad (\text{A-6})$$

The derivatives in Equation A-1 are

$$\frac{\partial^2 W}{\partial I \partial s} = -\frac{3}{4} \frac{A}{(1+bs)^2} \left[\frac{1}{I+II-3} + \frac{7}{I+6} \right], \quad (A-7)$$

and

$$\frac{\partial^2 W}{\partial II \partial s} = -\frac{3}{4} \frac{A}{(1+bs)^2} \left[\frac{1}{I+II-3} - \frac{7}{II+6} \right]. \quad (A-8)$$

II. RIGOROUS APPROACH TO CONICAL FLOW PROBLEM

Consider the radial converging flow of a viscoelastic fluid in a cone of half-angle α . The problem will be described in the spherical coordinate system, where it will be assumed that $V_\Theta = 0$, $V_\Phi = 0$, and $\partial(\)/\partial\Phi = 0$. Inertial effects will be assumed negligible. The continuity equation is

$$r^2 v_r = f(\Theta). \quad (A-9)$$

The r -component of the momentum equation is

$$\begin{aligned} -\frac{\partial P(rr)}{\partial r} &= \frac{1}{r \sin \Theta} \frac{\partial}{\partial \Theta} [\tau(r\Theta) \sin \Theta] \\ &+ \frac{P(rr) - P(\Theta\Theta)}{r} + \frac{P(rr) - P(\Phi\Phi)}{r}, \end{aligned} \quad (A-10)$$

where

$$P(rr) = -P + \tau(rr). \quad (A-11)$$

From the definition of time scale

$$s = \int_R^F \frac{dr}{V_2} . \quad (A-12)$$

Using Equation A-9 and integrating gives

$$s = \frac{1}{F} \int_R^F r^2 dr = \frac{1}{3F} (r^3 - R^3) . \quad (A-13)$$

The coordinates of points at two different times are then related by

$$\left. \begin{aligned} r &= (3fs + R^3)^{1/3} \\ \theta &= \Theta \\ \phi &= \Phi . \end{aligned} \right\} \quad (A-14)$$

The components of the relative Finger strain tensor may be written

$$\begin{bmatrix} C^{-1r}_r & C^{-1r}_\theta & C^{-1r}_\phi \\ C^{-1\theta}_r & C^{-1\theta}_\theta & C^{-1\theta}_\phi \\ C^{-1\phi}_r & C^{-1\phi}_\theta & C^{-1\phi}_\phi \end{bmatrix} = \begin{bmatrix} h^2 + h^{-1} F'^2 s^2 & r h^{-1} F' s & 0 \\ \frac{1}{r} h^{-1} F' s & h^{-1} & 0 \\ 0 & 0 & h^{-1} \end{bmatrix} , \quad (A-15)$$

and the components of the relative Cauchy strain tensor may be written

$$\begin{bmatrix} C_r^r & C_\theta^r & C_\phi^r \\ C_r^\theta & C_\theta^\theta & C_\phi^\theta \\ C_r^\phi & C_\theta^\phi & C_\phi^\phi \end{bmatrix} = \begin{bmatrix} h^{-2} & -rh^{-2}F's & 0 \\ -\frac{1}{r}h^{-2}F's & h+h^{-2}F'^2s^2 & 0 \\ 0 & 0 & h \end{bmatrix}, \quad (\text{A-16})$$

where

$$h = \left(1 - \frac{3fs}{r^3}\right)^{2/3} = (1 - 3Fs)^{2/3}$$

and

$$F = \frac{f}{r^3}.$$

The scalar invariants of the Finger strain tensor are

$$\left. \begin{aligned} \text{I} &= h^2 + h^{-1}(F'^2s^2 + 2) \\ \text{II} &= 2h + h^{-2}(F'^2s^2 + 1) \\ \text{III} &= 1 \end{aligned} \right\}. \quad (\text{A-17})$$

Using Equations A-1 to A-8 the shear stress is given by

$$\begin{aligned} \tau(r\theta) = \frac{3}{2} \int_0^{s^*} \frac{A}{(1+bs)^2} & \left[\frac{F'sh^{-1}(1+h^{-1})}{h^2 + 2h - 3 + h^{-1}(F'^2s^2 + 2) + h^{-2}(F'^2s^2 + 1)} \right. \\ & \left. + \frac{7h^{-1}F's}{h^2 + 6 + h^{-1}(F'^2s^2 + 2)} - \frac{7h^{-2}F's}{2h + 6 + h^{-2}(F'^2s^2 + 1)} \right] ds. \quad (\text{A-18}) \end{aligned}$$

The difference in normal stresses is given by

$$P(rr)-P(\theta\theta) = P(rr)-P(\phi\phi)$$

$$= \frac{3}{2} \int_0^{s^*} \frac{A}{(1+bs)^2} \left[\frac{h^2+h+h^{-1}(F'^2 s^2 -1)+h^{-2}(F'^2 s^2 -1)}{h^2+2h-3+h^{-1}(F'^2 s^2 +2)+h^{-2}(F'^2 s^2 +1)} \right. \\ \left. + \frac{7[h^2+h^{-1}(F'^2 s^2 -1)]}{h^2+6+h^{-1}(F'^2 s^2 +2)} + \frac{7[h+h^{-2}(F'^2 s^2 -1)]}{2h+6+h^{-2}(F'^2 s^2 +1)} \right] ds. \quad (A-19)$$

Obviously Equations A-18 and A-19 cannot be handled analytically without making some rather drastic assumptions. The crux of the problem is that the spherical coordinates do not allow the strain tensors to collapse as they do in cartesian and cylindrical coordinates; i.e., the strain tensors take the form of a polynomial raised to a fractional power. This is a very difficult form to treat analytically.

The equations could, of course, be integrated by computer, but it is felt that this would not be a profitable course of action at this time. Such an approach would require decisions on such subtle problems as the validity of the radial flow assumption, the range of validity of the empirical memory function, and the wisdom of choosing the BKZ constitutive equation to the exclusion of other promising theories. The scope of this thesis has been limited, rather, to using the BKZ theory to gain insight into the pertinent variables of this very complex problem and into the interplay of the elastic and viscous effects.

III. "SECOND ORDER" APPROACH TO CONICAL FLOW PROBLEM

Let us circumvent the problem of unwieldy strain tensors by approximating each of the terms in Equations A-15 and A-16 with a Maclaurin's series, truncated after the s^2 term. Thus

$$C^{-1j}_i(s) = C^{-1j}_i(0) + \left. \frac{\partial C^{-1j}_i}{\partial s} \right|_{s=0} s + \left. \frac{\partial^2 C^{-1j}_i}{\partial s^2} \right|_{s=0} \frac{s^2}{2!} + \dots, \quad (\text{A-20})$$

and

$$C^i_j(s) = C^i_j(0) + \left. \frac{\partial C^i_j}{\partial s} \right|_{s=0} s + \left. \frac{\partial^2 C^i_j}{\partial s^2} \right|_{s=0} \frac{s^2}{2!} + \dots \quad (\text{A-21})$$

Such an approach was suggested by the familiar "second order" approach to the Coleman-Noll theory.

The strain tensors then become

$$\begin{bmatrix} C^{-1r}_r & C^{-1r}_\theta & C^{-1r}_\phi \\ C^{-1\theta}_r & C^{-1\theta}_\theta & C^{-1\theta}_\phi \\ C^{-1\phi}_r & C^{-1\phi}_\theta & C^{-1\phi}_\phi \end{bmatrix} = \begin{bmatrix} 1-4Fs+(2F^2+F'^2)s^2 & r[F's+2FF's^2] & 0 \\ \frac{1}{r}[F's+2FF's^2] & 1+2Fs+5F^2s^2 & 0 \\ 0 & 0 & 1+2Fs+5F^2s^2 \end{bmatrix}, \quad (\text{A-22})$$

and

$$\begin{bmatrix} C_r^r & C_\theta^r & C_\phi^r \\ C_r^\theta & C_\theta^\theta & C_\phi^\theta \\ C_r^\phi & C_\theta^\phi & C_\phi^\phi \end{bmatrix} = \begin{bmatrix} 1+4Fs+14F^2s^2 & -r[F's+4FF's^2] & 0 \\ \frac{-1}{r}[F's+4FF's^2] & 1-2Fs+(F'^2-F^2)s^2 & 0 \\ 0 & 0 & 1-2Fs-F^2s^2 \end{bmatrix}. \quad (A-23)$$

The scalar invariants of the Finger strain tensor become

$$\left. \begin{aligned} I &= 3+(12F^2+F'^2)s^2, \\ II &= 3+(12F^2+F'^2)s^2, \\ III &= 1, \end{aligned} \right\} \quad (A-24)$$

where all terms of order higher than s^2 have been neglected.

The shear stress can now be written

$$\tau(r\theta) = \frac{3}{2} \int_0^{s^*} \frac{A}{(1+bs)^2} \cdot \left[\frac{2(F's+3FF's^2)}{3+2(12F^2+F'^2)s^2} - \frac{14FF's^2}{9+(12F^2+F'^2)s^2} \right] ds, \quad (A-25)$$

and the normal stress difference

$$P(rr) - P(\theta\theta) = P(rr) - P(\phi\phi) = \frac{3}{2} \int_0^{s^*} \frac{A}{(1+bs)^2} ds \quad (A-26)$$

$$\cdot \left[\frac{-2(6Fs + 9F^2s^2 - F'^2s^2)}{3 + 2(12F^2 + F'^2)s^2} + \frac{84F^2s^2}{9 + (12F^2 + F'^2)s^2} \right] ds.$$

Equations A-25 and A-26 can be integrated analytically, using the method of partial fractions. The first integrand of Equation A-25, for example, can be written

$$\frac{3A(F's + 3FF's^2)}{(1+bs)^2 [3 + 2(12F^2 + F'^2)s^2]} = \frac{B_1}{(1+bs)^2} \quad (A-27)$$

$$+ \frac{B_2}{(1+bs)} + \frac{B_3s + B_4}{3 + 2(12F^2 + F'^2)s^2}.$$

Equations A-25 and A-26 then become

$$\begin{aligned} r(r\theta) = & B_1 \int_0^{s^*} \frac{ds}{(1+bs)^2} + B_2 \int_0^{s^*} \frac{ds}{(1+bs)} + B_3 \int_0^{s^*} \frac{sds}{3 + 2(12F^2 + F'^2)s^2} \\ & + B_4 \int_0^{s^*} \frac{ds}{3 + 2(12F^2 + F'^2)s^2} - C_1 \int_0^{s^*} \frac{ds}{(1+bs)^2} \\ & - C_2 \int_0^{s^*} \frac{ds}{(1+bs)} - C_3 \int_0^{s^*} \frac{sds}{9 + (12F^2 + F'^2)s^2} \end{aligned}$$

(equation continued)

$$-C_4 \int_0^{s^*} \frac{ds}{9+(12F^2+F'^2)s^2}, \quad (\text{A-28})$$

and

$$\begin{aligned} P(rr)-P(\theta\theta) = P(rr)-P(\phi\phi) = & D_1 \int_0^{s^*} \frac{ds}{(1+bs)^2} + D_2 \int_0^{s^*} \frac{ds}{(1+bs)} \\ & + D_3 \int_0^{s^*} \frac{sds}{3+2(12F^2+F'^2)s^2} + D_4 \int_0^{s^*} \frac{ds}{3+2(12F^2+F'^2)s^2} \\ & + E_1 \int_0^{s^*} \frac{ds}{(1+bs)^2} + E_2 \int_0^{s^*} \frac{ds}{(1+bs)} + E_3 \int_0^{s^*} \frac{sds}{9+(12F^2+F'^2)s^2} \\ & + E_4 \int_0^{s^*} \frac{ds}{9+(12F^2+F'^2)s^2}. \end{aligned} \quad (\text{A-29})$$

Integration of Equations A-28 and A-29 gives

$$\begin{aligned} \gamma(r\theta) = & \frac{(B_1+C_1)s^*}{1+bs^*} + \frac{(B_2+C_2)}{b} \ln(1+bs^*) \\ & + \frac{B_3}{4(12F^2+F'^2)} \ln \left[1 + \frac{2}{3}(12F^2+F'^2)s^{*2} \right] \\ & + \frac{C_3}{2(12F^2+F'^2)} \ln \left[1 + \frac{1}{9}(12F^2+F'^2)s^{*2} \right] \end{aligned}$$

(equation continued)

$$\begin{aligned}
& + \frac{B_4}{\sqrt{6(12F^2+F'^2)}} \tan^{-1} \left[\frac{s^* \sqrt{6(12F^2+F'^2)}}{3} \right] \\
& + \frac{C_4}{\sqrt{9(12F^2+F'^2)}} \tan^{-1} \left[\frac{s^* \sqrt{9(12F^2+F'^2)}}{9} \right], \quad (A-30)
\end{aligned}$$

and

$$\begin{aligned}
P(rr) - P(\theta\theta) = P(rr) - P(\phi\phi) &= \frac{(D_1+E_1)s^*}{(1+bs^*)} + \frac{(D_2+E_2)}{b} \ln(1+bs^*) \\
& + \frac{D_3}{4(12F^2+F'^2)} \ln \left[1 + \frac{2}{3} (12F^2+F'^2)s^{*2} \right] \\
& + \frac{E_3}{2(12F^2+F'^2)} \ln \left[1 + \frac{1}{9} (12F^2+F'^2)s^{*2} \right] \\
& + \frac{D_4}{\sqrt{6(12F^2+F'^2)}} \tan^{-1} \left[\frac{s^* \sqrt{6(12F^2+F'^2)}}{3} \right] \\
& + \frac{E_4}{\sqrt{9(12F^2+F'^2)}} \tan^{-1} \left[\frac{s^* \sqrt{9(12F^2+F'^2)}}{9} \right]. \quad (A-31)
\end{aligned}$$

The constants B_i , C_i , D_i , and E_i are all complicated functions of r . By making further approximations one could obtain expressions for $\gamma(r0)$ and $P(rr) - P(\theta\theta)$ which could be substituted into the momentum equation and integrated with respect to r to obtain an expression for pressure drop. The results of such a procedure would be similar to those reported by Adams for viscoelastic flow between non-parallel flat plates. In this case quantitative results would not be

expected, however, because the approximations made in Equations A-20 and A-21 would be valid only at very low shear rates.

From Equations A-10, A-30, and A-31 it can be seen that the pressure drop will be a function of the dimensionless variables Fs^* and $F's^*$. These variables have the properties of the Deborah number which has been suggested by Reiner (32) and more recently by Metzner, White, and Denn (26). The parameter s^* is a characteristic time for the material, while the quantities $1/F$ and $1/F'$ have units of time and are characteristic of the process. Let $Fs^* = N_{Deb}$ and $F's^* = N'_{Deb}$. If N_{Deb} (or N'_{Deb}) is large for a particular process, the material is behaving like a solid. If N_{Deb} (or N'_{Deb}) is small the material is behaving like a fluid. The quantity F' is the shear rate in the cone, while F may be interpreted as a normal deformation rate which is related to the residence time in the accelerating flow field. Because $F=0$ at the wall and $F'=0$ at the centerline, one would suppose that F'_ws^* is the appropriate group for correlating data at the wall and that F_0s^* is the appropriate group for correlating data at the centerline.

APPENDIX B

CONICAL FLOW WITH INERTIAL EFFECTS

In the author's master's thesis an experimental study of the converging and diverging flow of an Attagel clay slurry in a cone was described (11). A correlation method was suggested which gave a dimensionless pressure gradient as a function of a modified local Reynolds number $R_{r\alpha}^*$, the half angle of the cone α , and the power law index n . Since that time other workers have published papers (to be noted later) which deal with the same problem. In particular, these papers have contributed to a better understanding of the inertial effects in the cone and have provided additional data which agree with the author's correlation scheme.

Nishimura and Oka (27) used a method of successive approximations to obtain a Newtonian solution for flow in a converging cone. Their solution considered two velocity components and did not neglect the inertial effects, as had been done in previous work (12, 16, 28). A similar Newtonian solution has been presented by Slezkin (38). Sutterby (40, 41, 42) and Boles (11) performed experiments with two different non-Newtonian materials. These results will be presented and discussed for the case of converging

flow. The reader may refer to reference (11) for a detailed discussion of the theory, and to references (11) and (41) for detailed discussions of the experimental methods. A brief description of the essential elements of the theory and some of the results are presented below.

I. THEORY

The "scalar viscosity model" or "generalized Newtonian fluid model" was used to relate the stress to the rate of deformation:

$$\tau_{ij} = -g_{ij}P + \eta d_{ij}. \quad (B-1)$$

In general, the scalar viscosity is a function of the three scalar invariants of the rate of deformation tensor I_d , II_d , and III_d . For incompressible fluids, $I_d=0$. Some concern has been expressed over the fact that III_d is not zero in most non-viscometric flows, including conical flow (43). Usually one proceeds with the assumption that the effect of III_d is negligible and, if a good correlation with experimental data is obtained, concludes that the assumption is valid. From the theoretical standpoint, the omission of III_d is justified by relations presented by Reiner (33), who showed that if η_c is zero, and if the energy dissipation function is positive, then η depends only on II_d . These relations are discussed in some detail by Keen (19).

In this investigation the generalized power law model was used to describe the viscosity. It may be written as

$$\eta = k \left| II_d \right|^{\frac{n-1}{2}}. \quad (B-2)$$

Objections have been made to the power law in view of its inability to describe a low shear limiting viscosity and of the unusual units of the parameter k . It does, however, have the advantage of being simple. As Tanner (43) points out, one of the objections to the power law model is partially reconciled in multidimensional flow because of the accelerative term in II_d (see Equation B-4); that is, it does not predict an infinite viscosity at the centerline.

In order to obtain explicit results for correlating pressure drop data, certain "reasonable" assumptions will now be made. These are as follows: (1) the flow is radial ($V_r = \text{finite}$, $V_\theta = 0$) and non-spiralling ($V_\phi = 0$), (2) the angle of the cone is small ($\sin \theta \approx \theta$), and (3) the pressure gradient is a function of the radial position in the cone only.

The first assumption may be stated in spherical coordinates by the equations:

$$r^2 V_r = f(\theta),$$

$$V_\theta = 0, \quad V_\phi = 0. \quad (B-3)$$

With these equations the second invariant of deformation rate becomes

$$II_d = \left(\frac{1}{r} \frac{\partial v_r}{\partial \theta}\right)^2 + 12\left(\frac{v_r}{r}\right)^2. \quad (B-4)$$

Combining Equations B-2, B-3, and B-4 gives

$$\eta = \frac{k}{r^{3(n-1)}} \left[1 + \frac{12f^2}{f'^2} \right]^{\frac{n-1}{2}} |f'|^{n-1}. \quad (B-5)$$

If a parabolic velocity profile is assumed and f and f' are approximated by their averages over θ , $\bar{f}$ and $\bar{f}'$, the bracketed term becomes

$$G \approx \left[1 + \left(\frac{2n+1}{3n+1} \right)^2 3\alpha^2 \right]^{\frac{n-1}{2}}. \quad (B-6)$$

Equation B-5 may then be rewritten as

$$\eta = \frac{KG}{r^{3(n-1)}} |f'|^{n-1}. \quad (B-7)$$

Note that G reduces properly for Newtonian flow ($n=1$) and for non-accelerative flow ($\alpha=0$). For $\alpha=10^0$, G varies from 0.956 at $n=0$ to 1.000 at $n=1$. It is therefore concluded that the effect of the second term in the bracket in Equation B-6 is small in the present experiments (that is, $G \approx 1$).

Under the assumptions stated, the r-component of the equations of motion may be written as

$$V_r \frac{\partial V_r}{\partial r} = -\frac{\partial P}{\partial r} + \frac{1}{r\theta} \frac{\partial}{\partial \theta} \left(\eta \frac{\partial V_r}{\partial \theta} \theta \right) + \frac{1}{r^2} \frac{\partial}{\partial r} \left(2r^2 \eta \frac{\partial V_r}{\partial r} \right) + \frac{4\eta V_r}{r^2}, \quad (\text{B-8})$$

or

$$\frac{\partial P}{\partial r} = \underbrace{\frac{1}{r^4 \theta} \frac{\partial}{\partial \theta} (\eta f' \theta)}_{\text{shear}} + \underbrace{2 \frac{f^2}{r^5}}_{\text{inertial}} - \underbrace{\frac{4f}{r^3} \frac{\partial \eta}{\partial r}}_{\text{normal}} \quad (\text{B-9})$$

where

$$\eta = k \left| \frac{f'}{r^3} \right|^{n-1}. \quad (\text{B-10})$$

The three terms on the right hand side of Equation B-9 will now be approximated separately.

Shear Component

The shear component is treated first by considering Equation B-9 in the absence of other terms. With $\partial P / \partial r = dP/dr$, this differential equation may be solved to give

$$P^* = \frac{16}{R_*^* r \alpha} \quad (\text{converging})$$

$$P^* = -\frac{16}{R_*^* r \alpha} \quad (\text{diverging})$$

(shear component) (B-11)

where

$$P^* = \frac{r\alpha}{\rho \bar{V}^2} \left. \frac{dP}{dr} \right|_w, \quad \text{dimensionless pressure gradient,}$$

$$R_{r\alpha}^* = \frac{(2r\alpha) \bar{V}^{2-n} \rho}{\frac{k}{8} \left(\frac{6n+2}{n} \right)^2}, \quad \text{local Reynolds number,}$$

$$\bar{V}(r) = \frac{Q}{\pi r^2 \alpha^2}, \quad \text{mean velocity at some position } r,$$

Q = volumetric flow rate.

The velocity profile is given by

$$f(\theta) \equiv r^2 v_r = \frac{-Q}{\pi \alpha^2} \left(\frac{3n+1}{n+1} \right) \left[1 - \left(\frac{\theta}{\alpha} \right)^{\frac{n+1}{n}} \right]. \quad (\text{B-12})$$

These results are, of course, just those for power law flow in a straight pipe if r is replaced by the pipe radius, R (49). An interesting feature of this result is that the Reynolds number is r -dependent (it varies from point to point in the cone). This is another way of saying that the equations of motion have rejected the radial flow assumption, although we will continue to use this assumption as a first approximation.

Inertial Component

The inertial component may be approximated (once the velocity profile is known) by using an overall mechanical energy balance:

$$\frac{dP}{dr} = -\frac{\rho}{2} \frac{d}{dr} \left[\frac{\bar{v}_r^3}{\bar{v}_r} \right] = \frac{2\rho}{r^5} \frac{\int_0^\alpha f^3 \theta d\theta}{\int_0^\alpha f \theta d\theta}. \quad (\text{B-13})$$

For low flow rates, Equation B-12 is suitable and one obtains the result:

$$P^* = \left[\frac{6(3n+1)^2}{(2n+1)(5n+3)} \right] \alpha \quad (\text{inertial component}). \quad (\text{B-14})$$

For very high flow rates one intuitively expects the velocity profile to be flat (i.e., stress effects to be absent). For this case one obtains

$$P^* = 2\alpha \quad (\text{inertial component}). \quad (\text{B-15})$$

One would, of course, like to identify the Reynolds number range where Equation B-14 is valid and where Equation B-15 is valid. The inertial term in the Newtonian solution of Nishimura and Oka (27) reduces to $P^*=4\alpha$ (Equation B-14 with $n=1$) in the limit of small angles, as one would expect from a solution constructed about the slow flow asymptote. Their solution cannot, however, be easily extended to high Reynolds numbers. Thus, the best one is able to do is to bound the inertial contribution with Equations B-14 and B-15. Fortunately for most non-Newtonian fluids the bracketed term in

Equation B-14 is lower than 4 and in fact approaches 2 as n becomes small.

Normal Component

From Equation B-9 the normal component is seen to be zero when evaluated at the wall.

The final correlating equation is obtained by adding the shear and inertial contributions:

$$P^* = A\alpha \pm \frac{16}{R_{r\alpha}^*} \quad (B-16)$$

The plus sign is for converging flow and the minus sign is for diverging flow. The parameter A is bounded by

$$2 < A < \frac{6(3n+1)^2}{(2n+1)(5n+3)}. \quad (B-17)$$

With regard to diverging flow, note that there is a Reynolds number at which the shear and inertial components exactly cancel, giving a "null point."

In summary, then, an analytical correlating equation has been formulated by adding together certain limiting results: that for pure shear flow and that for pure inertial (Bernoulli) flow. Viscoelastic normal stresses are not included, these being beyond the scope of the scalar viscosity model. The correlation procedure is quite similar to that of Sutterby (40, 41, 42), the essential

difference being that the power law model is used here in contrast to the modified Eyring model used by Sutterby. The modified Eyring model predicts a zero shear viscosity, but it is believed that several sets of power law parameters, evaluated in several shear rate ranges, serve nearly as well and permit simpler analytical results.

The radial flow assumption can be made throughout. The Nishimura and Oka solution (for Newtonian fluids) eliminates this assumption and produces a higher order approximation for the pressure drop. Their solution is a series expansion about the slow flow asymptote and reduces to Equation B-16 (with A given by Equation B-14 and $n=1$) in the limit of small cone angles. For Newtonian fluids their solution provides a basis for a theoretical extension to "modest" cone angles, but this degree of rigor was not felt justified by the data. Direct evidence in favor of the radial flow assumption is the numerical solution of Keen (19) who treated flow between non-parallel flat plates, using the power law viscosity model; this problem also rigorously rejects the radial flow assumption but can nonetheless be treated approximately with it.

II. RESULTS

The results for straight pipe flow and for diverging cone flow are presented in reference (11). Only the converging flow results of Boles (11) and Sutterby (40, 41, 42)

will be presented here.

The results for converging slurry flow are compared with Equation B-16 in Figure 21. The inertial parameter A is bounded by the results shown in Equation B-17. A representative run from Sutterby's work is shown in Figure 22; the material in this run was 0.7 per cent Natrosol in water flowing in a converging cone with $\alpha \approx 70^\circ$. In calculating the Reynolds numbers, power law parameters corresponding to the wall shear stress in the cone (or slightly below it) were used. While there is some scatter, probably due to rheological difficulties in the case of the slurry, and some differences in agreement with the high Reynolds number asymptotes, there can be said to be reasonable agreement throughout.

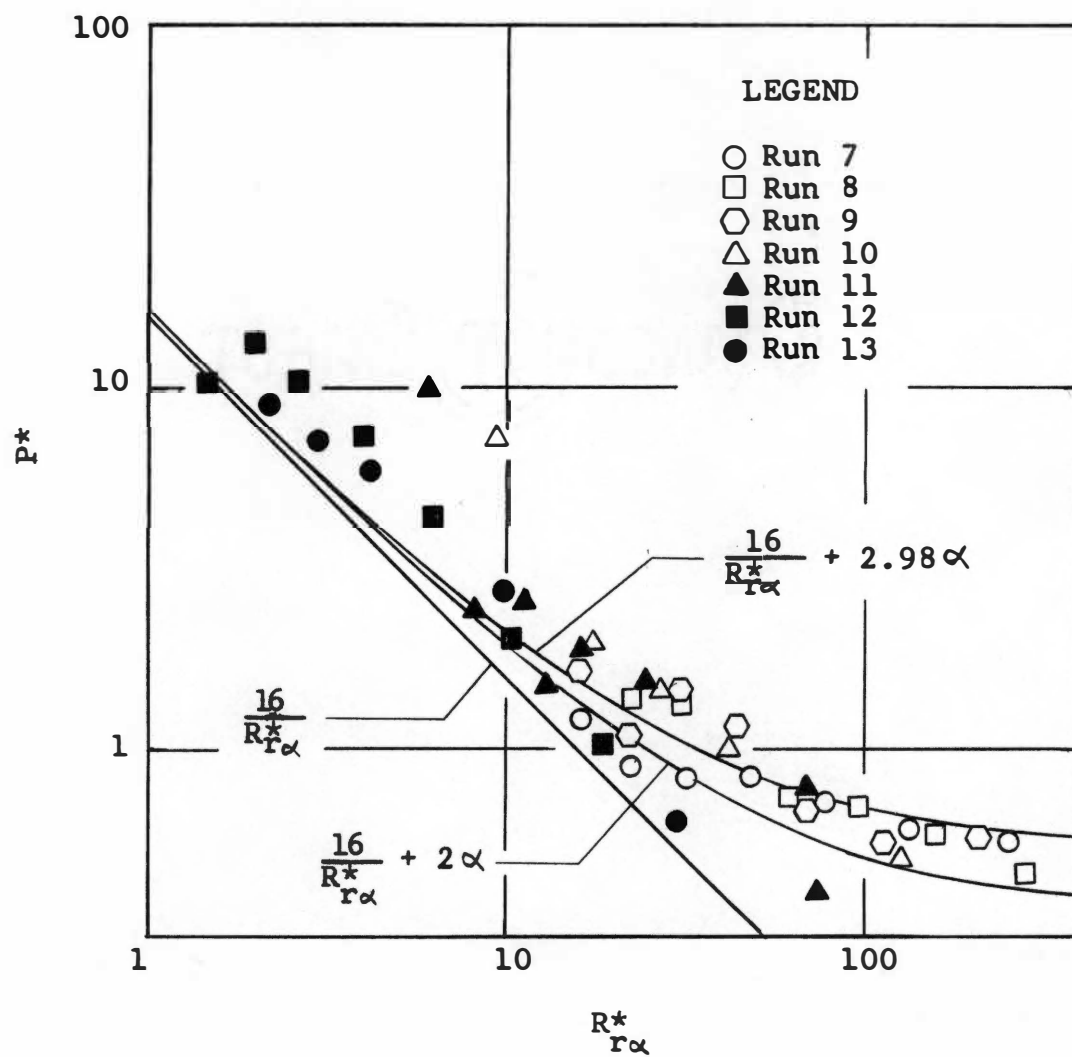


Figure 21. Slurry Data for a Converging Cone.

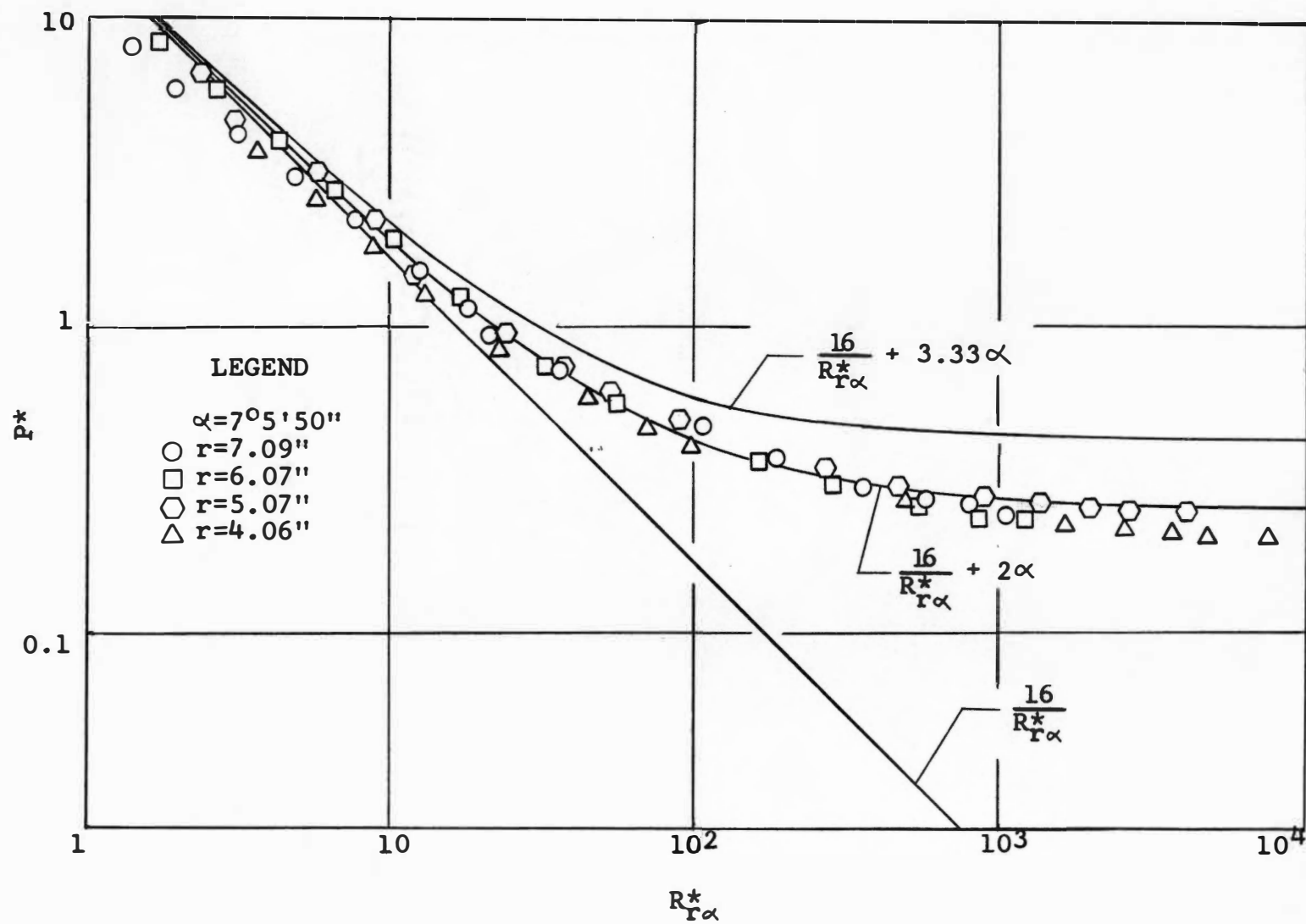


Figure 22. Natrosol Solution Data for a Converging Cone (Sutterby).

APPENDIX C

TABULATION OF DATA

In capillary experiments the pressure drop is imposed and the flow rate is measured. Since it is desirable to have the shear rate as the independent variable, it is often necessary to interpolate between data points to obtain the necessary values of pressure drop at a given rate of shear for each capillary. This interpolation was carried out graphically by plotting the log total pressure drop versus the log effective shear rate ($4Q/\pi R^3$) for each capillary. Smooth curves through the data points were used for interpolation. The raw capillary data presented in Tables II through IX were taken from these plots. In the case of the longer tubes the desired high pressures could not be obtained for oil OB. These values were obtained by extrapolating the plots of $\log \Delta P(\text{TOT})$ versus $\log (4Q/\pi R^3)$. The extrapolation is considered reasonable since the plots gave straight lines. Such values are marked by an asterisk in Tables II through V.

Rheogoniometer data for 12 per cent polyisobutylene in Decalin and for oil OB are presented in Tables X and XI.

TABLE II

CAPILLARY DATA FOR OIL OB WITH $\alpha=90^\circ$

$4Q/\pi R^3$ (sec ⁻¹)	L/R:	$\Delta P(TOT)$ (p.s.i.)		
	166.4	52.7	9.92	6.05
50	51.7	16.6	3.45	1.98
100	101	32.5	6.80	3.90
150	*151	48.1	10.1	5.80
200	*201	64.0	13.3	7.75
300	*299	95.0	20.0	11.5
400	*398	127	26.2	15.4
500		*157	32.5	19.2
600		*188	39.0	23.0
700		*220	45.5	26.9
800		*250	51.5	30.7
900		*280	58.0	34.6
1000		*310	64.0	38.2

*Denotes data obtained by extrapolation.

TABLE III

CAPILLARY DATA FOR OIL OB WITH $\alpha=46.7^\circ$

$4Q/\pi R^3$ (sec. ⁻¹)	$\Delta P(TOT)$ (p.s.i.)					
	L/R:	166.4	52.7	9.92	6.05	0
50		52.7	16.8	4.42	2.85	1.40
100		106	33.6	8.75	5.68	2.75
150		*161	50.0	13.0	8.50	4.10
200		*217	67.0	17.3	11.3	5.40
300		*330	100	25.6	16.9	8.00
400			134	34.0	25.0	10.7
500			*167	42.2	28.0	13.2
600			*200	50.3	33.4	15.8
700			*234	58.4	39.0	18.3
800			*266	67.0	44.8	21.0
900			*300	74.5	50.0	23.5
1000			*332	83.0	55.0	26.1

*Denotes data obtained by extrapolation.

TABLE IV

CAPILLARY DATA FOR OIL OB WITH $\alpha=22^\circ$

$4Q/\pi R^3$ (sec. ⁻¹)	$\Delta P(TOT)$ (p.s.i.)					
	L/R:	166.4	52.7	9.92	6.05	0
50		52.7	17.1	4.20	2.80	0.75
100		107	33.7	8.15	5.47	1.47
150		*163	50.0	12.0	8.0	2.18
200		*219	66.0	15.9	10.7	2.88
300		*330	98.0	23.3	15.8	4.27
400			130	30.8	20.9	5.62
500			*162	38.0	26.0	7.00
600			*194	45.2	31.0	8.35
700			*225	52.5	35.9	9.65
800			*256	59.6	40.8	11.1
900			*286	66.5	45.6	12.4
1000			*319	74.0	50.1	13.6

*Denotes data obtained by extrapolation.

TABLE V

CAPILLARY DATA FOR OIL OB WITH $\alpha=10.8^\circ$

$\frac{4Q}{\pi R^3}$ (sec ⁻¹)	L/R:	$\Delta P(TOT)$ (p.s.i.)				
		166.4	52.7	9.92	6.05	0
50		52.0	17.2	4.30	3.14	1.26
100		102	34.0	8.45	6.00	2.50
150		*151	50.5	12.5	8.79	3.70
200		*201	67.2	15.6	11.5	4.91
300		*300	100	24.6	16.7	7.30
400		*395	132	32.7	22.0	9.65
500			*165	40.9	27.0	12.0
600			*197	48.9	32.0	14.4
700			*230	56.2	37.0	16.7
800			*262	64.1	41.8	19.1
900			*293	72.0	46.6	21.5
1000			*326	80.0	51.0	23.7

*Denotes data obtained by extrapolation.

TABLE VI

CAPILLARY DATA FOR 12 PER CENT POLYISOBUTYLENE
IN DECALIN WITH $\alpha=90^\circ$

$\frac{4Q}{\pi R^3}$ (sec. ⁻¹)	$\Delta P(TOT)$ (p.s.i.)				
	L/R:	166.4	52.7	9.92	6.05
50		57.0	18.6	4.03	3.07
100		70.0	23.1	5.57	4.36
150		78.0	26.2	6.68	5.34
200		84.5	28.7	7.65	6.20
300		95.5	32.9	9.30	7.58
400		104	36.1	10.6	8.83
500		110	38.6	11.8	9.90
600		116	41.0	12.8	10.8
700		122	42.9	13.7	11.6
800		126	44.8	14.6	12.5
900		131	46.5	15.4	13.2
1000		135	47.9	16.1	14.0

TABLE VII

CAPILLARY DATA FOR 12 PER CENT POLYISOBUTYLENE
IN DECALIN WITH $\alpha=46.7^\circ$

$\frac{4Q}{\pi R^3}$ (sec. ⁻¹)	$\Delta P(TOT)$ (p.s.i.)					
	L/R:	166.4	52.7	9.92	6.05	0
50		59.0	20.0	5.04	3.96	1.65
100		72.0	24.4	6.72	5.28	2.73
150		80.7	27.7	7.87	6.23	3.59
200		88.0	30.4	8.90	7.00	4.36
300		98.5	34.5	10.7	8.70	5.70
400		107	38.2	12.3	10.2	6.94
500		114	41.0	13.7	11.6	7.90
600		120	43.5	15.0	12.8	9.00
700		126	45.8	16.0	13.9	9.90
800		131	47.6	17.1	15.1	10.9
900		135	49.4	18.1	16.0	11.5
1000		139	50.9	19.1	16.9	12.2

TABLE VIII

CAPILLARY DATA FOR 12 PER CENT POLYISOBUTYLENE
IN DECALIN WITH $\alpha=22^\circ$

$\frac{4Q}{\pi R^3}$ (sec. ⁻¹)	L/R:	166.4	52.7	$\frac{\Delta P(TOT)}{(p.s.i.)}$ 9.92	6.05	0
50		59.5	19.0	5.15	3.81	1.23
100		72.5	23.3	5.77	4.53	1.69
150		81.0	26.1	6.66	5.18	2.21
200		88.0	28.8	7.53	5.87	2.72
300		99.0	33.1	9.30	7.14	3.62
400		107	36.4	10.8	8.44	4.52
500		115	39.2	12.2	9.70	5.50
600		120	41.9	13.6	10.9	6.42
700		126	43.9	14.9	12.1	7.35
800		131	46.1	16.1	13.3	8.28
900		136	48.0	17.3	14.4	9.10
1000		139	49.8	18.5	15.5	10.0

TABLE IX

CAPILLARY DATA FOR 12 PER CENT POLYISOBUTYLENE
IN DECALIN WITH $\alpha=10.8^\circ$

$4Q/\pi R^3$ (sec^{-1})	L/R:	166.4	52.7	$\Delta P(\text{TOT})$ (p.s.i.) 9.92	6.05	0
50		59.0	19.6	4.82	3.97	1.85
100		72.0	24.3	6.17	5.06	2.56
150		80.2	27.1	7.06	5.83	3.13
200		87.0	29.3	7.83	6.45	3.56
300		97.5	32.8	9.10	7.43	4.32
400		106	36.0	10.2	8.44	5.04
500		112	38.4	11.2	9.40	5.70
600		118	40.8	12.1	10.2	6.32
700		124	42.8	13.0	10.9	7.02
800		128	44.4	13.9	11.6	7.69
900		132	46.1	14.6	12.1	8.30
1000		136	47.8	15.4	12.8	9.00

TABLE X

RHEOGONIOMETER DATA FOR 12 PER CENT POLYISOBUTYLENE
IN DECALIN AT 25°C.

K (sec^{-1})	τ_{12} (dynes/cm^2)	$\tau_{11} - \tau_{22}$ (dynes/cm^2)
1.73×10^{-2}	4.5×10^1	-
2.17	6.3	-
2.75	9.0	-
3.46	1.17×10^2	-
4.34	1.44	-
5.46	1.80	-
6.89	2.16	-
8.64	2.79	-
1.095×10^{-1}	3.42	-
1.375	4.23	-
1.730	5.40	-
2.170	6.57	-
2.75	7.92	-
3.46	9.54	-
4.34	1.15×10^3	-
5.46	1.38	9.0×10^2
6.89	1.62	1.3×10^3
8.64	1.93	1.52

TABLE X (continued)

K (sec. ⁻¹)	τ_{12} (dynes/cm. ²)	$\tau_{11} - \tau_{22}$ (dynes/cm. ²)
1.095x10 ⁰	2.30	1.80
1.375	2.65	2.28
1.730	3.06	3.46
2.170	3.37	4.50
2.75	3.87	4.97
8.58	6.47	1.21x10 ⁴
1.08x10 ¹	6.92	1.44
1.36	7.27	1.47
1.71	7.82	2.07
2.16	8.10	1.94
2.72	9.00	2.97
3.41	9.43	3.66
4.29	1.02x10 ⁴	3.94
5.41	1.08	4.70
6.78	1.15	5.87
8.56	1.21	6.56
1.08x10 ²	1.27	7.46
1.36	1.35	8.45
1.71	1.43	9.26
2.16	1.53	1.06x10 ⁵

TABLE X (continued)

K (sec ⁻¹)	τ_{12} (dynes/cm ²)	$\tau_{11} - \tau_{22}$ (dynes/cm ²)
2.72	1.63	1.23
3.41	1.72	1.38
4.29	1.83	1.59
5.41	1.93	1.87
6.78	2.06	2.14
8.56	2.16	2.42
1.08×10^3	2.29	2.83

TABLE XI

RHEOGONIOMETER DATA FOR OIL OB AT 25°C.

K (sec^{-1})	τ_{12} (dynes/cm^2)	η^a (poise)
5.33×10^0	1.23×10^2	2.31×10^2
6.72	1.555	2.32
8.44	1.963	2.33
1.06×10^1	2.45	2.31
1.34	3.10	2.32
1.68	3.96	2.36
2.12	4.98	2.35
2.67	6.30	2.36
3.36	7.95	2.37
4.22	1.008×10^2	2.39
5.33	1.260	2.36
6.72	1.583	2.36
8.44	2.010	2.38
1.06×10^2	2.47	2.33
1.34	3.11	2.32

TABLE XI (continued)

K (sec^{-1})	τ_{12} (dynes/cm ²)	η^a (poise)
1.68	3.96	2.35
2.12	4.95	2.33
2.67	6.17	2.31
3.36	7.65	2.28
4.22	9.45	2.24
5.33	1.14×10^4	2.15
6.72	1.35	2.01
8.44	1.585	1.88
1.06×10^3	1.800	1.70

^aNational Bureau of Standards calibrated value is 218.1 poise at 25°C.

LIST OF SYMBOLS

LIST OF SYMBOLS

<u>Symbol</u>	<u>Meaning</u>
a	- material constant, dimensionless.
A	- material constant, dimensionless; parameter defined by Equation B-17.
A_i	- partial fractions constants.
b	- time constant, sec^{-1}
b, b_3	- constants, dimensionless.
B_i	- partial fractions constants.
$c(s)$	- decay function, dynes/cm^2
C	- integration constant.
C_i	- partial fractions constants.
C_{ij}^{-1}, C_{ij}	- nonlinear strain tensors.
d	- diameter, cm.
D	- shear rate, sec^{-1}
D_i	- partial fractions constants.
E_i	- partial fractions constants.
f	- radial flow parameter, cm^3/sec .
f'	- derivative of f with respect to θ , cm^3/sec .
f''	- derivative of f' with respect to θ , cm^3/sec .
F	- radial flow parameter, sec^{-1} ; irreversible energy dissipation, dynes/cm^2
F'	- derivative of F with respect to θ , sec^{-1}
F''	- derivative of F' with respect to θ , sec^{-1}

<u>Symbol</u>	<u>Meaning</u>
F_c	- dissipation during developed capillary flow, dynes/cm ²
F_{down}	- dissipation during exit flow from a tube, dynes/cm ²
F_{inside}	- dissipation due to developing velocity profile just inside tube entrance, dynes/cm ²
F_{up}	- dissipation during flow upstream of tube entrance, dynes/cm ²
g_{ij}	- metric tensor.
G	- defined by Equation B-6, dimensionless.
G_i	- material constant, dynes/cm ²
h	- defined by Equations A-16 and A-17, dimensionless.
H_{ch}	- characteristic stress, dynes/cm ²
$H(\lambda)$	- relaxation function, dynes/cm ²
I, II, III	- scalar invariants of Finger strain tensor.
I_d, II_d, III_d	- scalar invariants of d_{ij} .
k	- fluid parameter in power law model (fluid consistency index), dynes-sec ⁿ /cm ²
K	- shear rate, sec ⁻¹
$m(s)$	- memory function, dynes/cm ²
$m'(s)$	- derivative of memory function, dynes/cm ²
n	- fluid parameter in power law model (fluid consistency index), dimensionless.

<u>Symbol</u>	<u>Meaning</u>
n'	- Mooney-Rabinowitsch exponent, dimensionless.
n_c	- Couette coefficient, dimensionless.
N_{Deb}	- Deborah number, dimensionless.
P	- pressure, p.s.i.
P^*	- dimensionless pressure gradient.
$\Delta P(EL)$	- elastic pressure difference, p.s.i.
$\Delta P(ENT)$	- pressure difference at entrance of capillary, p.s.i.
$\Delta P(EXIT)$	- pressure difference at exit of capillary, p.s.i.
$\Delta P(TOT)$	- total pressure difference, p.s.i.
$\Delta P(VISC)$	- viscous pressure difference, p.s.i.
P_{ij}	- total stress tensor, p.s.i.
Q	- volumetric flow rate, cm^3/sec .
r	- radial coordinate, cm.
R	- radius of tube, cm.
$R_{r\alpha}^*$	- effective local Reynolds number in cone, dimensionless.
s	- backward running time index, sec.
s^*	- material time constant, sec.
S_R	- recoverable shear, dimensionless.
t	- time, sec.; coordinate direction in oblate spheroidal coordinates.
u	- velocity in oblate spheroidal coordinates, cm./sec.

<u>Symbol</u>	<u>Meaning</u>
V_r	- radial component of velocity, cm./sec.
V_θ	- component of velocity in θ -direction, cm./sec.
V_ϕ	- component of velocity in ϕ -direction, cm./sec.
$\bar{V}$	- average velocity, cm./sec.
W	- a scalar function, dynes/cm ²
x	- coordinate direction, cm.
x^i	- coordinates at time t .
X^i	- coordinates at some past time.
z	- coordinate direction, cm.

Greek Letters

α	- half-angle of cone, degrees.
$\alpha(s)$	- decay function, dynes/cm ²
α_2	- viscous end effect coefficient, dimensionless.
α_3	- elastic end effect coefficient, dimensionless.
γ	- first normal stress coefficient, dynes-sec ² /cm ²
γ_0	- low shear rate value of γ , dynes-sec ² /cm ²
η	- viscosity, poise.
η_0	- low shear rate value of η , poise.
θ	- spherical coordinate direction, radians.
λ	- relaxation time, sec.
λ_i	- discrete relaxation time, sec.
λ_{ch}	- characteristic relaxation time, sec.
ρ	- density, gm./cm ³
τ	- shear stress, dynes/cm ²

<u>Symbol</u>	<u>Meaning</u>
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τ_R	- shear stress at wall of tube, dynes/cm ²
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τ_{ij}	- stress tensor.
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ϕ	- spherical coordinate direction, radians.
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ϕ, ϕ_2	- general functions.
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ψ	- stream function.
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VITA

Raymond L. Boles, Jr., was born on March 14, 1939, in Covington, Kentucky. His parents, Mr. and Mrs. Raymond L. Boles, Sr., and his two younger brothers, Ronald and Gary, reside in Knoxville. He was married on June 6, 1964, to Carolyn McKamey of Knoxville.

After graduating from Knoxville Fulton High School in June, 1957, the author entered The University of Tennessee. He received B.S. and M.S. degrees in Chemical Engineering in June, 1962, and March, 1965, respectively.

While at The University of Tennessee, the author was a member of the student chapter of the American Institute of Chemical Engineers, Tau Beta Pi national honorary fraternity, Alpha Chi Sigma chemistry fraternity, and the Sigma Xi national research society.

As an undergraduate, the author was employed as a co-op student with the Chemical Technology Division of Oak Ridge National Laboratory. He was also employed by the Reactor Chemistry Division of Oak Ridge National Laboratory during the summer of 1962.

Upon completion of the Ph.D. degree, the author plans to work in industry.