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and the Resulting Droplet Size Distributions
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NOMENCLATURE

Symbol:	Description
C	Concentration of macro-molecules
D	Injector exit diameter, diameter of liquid jet
d_j	Jet swell diameter
I_{nt}	Integral evaluated for contraction flow analysis
k	Consistency index, shear viscosity
K_o, K_u	Phase rejection parameters
L	Length of capillary tube
L/D	Length to diameter ratio
l	Consistency index (extensional viscosity)
L_v	Vortex attachment length
L_s	Length of spindle
n	power law index, shear viscosity
N_1	First normal stress difference
N_2	Second normal stress difference
n'	Flow behavior index
P	Isotropic pressure
P_e	Entry pressure
ΔP	Differential pressure drop
Q_v	Volumetric flow Rate
r	Radial distance
R	Capillary tube radius
R_0	Injector exit radius
Re	Reynolds number
R_i	Radius of spindle, inner radius
R_o	Radius of beaker, outer radius
t	Flow behavior Index (extensional viscosity)
T	Torque at inner or outer cylinder

Greek Letters

α	Growth rate of a disturbance
α_c	Inverse of contraction ratio
γ	Shear rate

γ_p	Reference shear rate 1 Sec-1
γ_R	Wall shear rate
δ_{ij}	Kronecker delta
ϵ	Stretch rate
η_e	Effective viscosity
η_p	Apparent viscosity at reference shear rate
η_s	Shear viscosity non-Newtonian liquids
η_a	Apparent viscosity
η_E	Extensional viscosity non-Newtonian liquids
η_T	Trouton viscosity
η_o	Shear viscosity of suspending agent
η_0	Shear viscosity of non-Newtonian liquids zero shear rate limit
η_{inf}	Shear viscosity of non-Newtonian liquids large shear rate limit
Θ	Contraction flow convergence angle
μ	Newtonian viscosity
$\sigma_{i,j}$	Total stress tensor
τ	Shear Stress
$\tau_{11}, \tau_{22}, \tau_{33}$	Anisotropic normal stress
τ_{ij}	Anisotropic shear stress
τ_n	Shear rate at which fluid becomes non-Newtonian
φ	R_o/R_i
Ω	Angular velocity of inner cylinder or spindle

Subscripts

0	Refers to zero shear rate
i	Inner
inf	Refers to large shear rate
j	Jet property
o	Outer

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Chapter 3

POLYMER RHEOLOGY

Rheology is the science which studies the deformation and flow of materials. Visco-elastic materials respond to an applied force or displacement by exhibiting a combination of elastic and viscous behavior. Most polymeric materials and some concentrated slurries are visco-elastic. Their mechanical properties show a marked time and temperature dependency. Slurries are suspensions where solids are dispersed in liquids. They show in general non-Newtonian behavior. Non-Newtonian liquids exhibit a non linear relationship between the shear stress and the shear rate. As shown in Fig. 3-1, non-Newtonian liquids can be pseudoplastic (shear thinning) or dilatant (shear thickening) and additionally may include a yield stress (Bingham plastic). The viscosity of a pseudoplastic fluid decreases when shear rate increases, and the viscosity of a dilatant increases when the shear rate increases. Many suspensions show time dependent rheological properties. This is mostly encountered in aggregate-forming suspensions: the aggregates are broken down during the application of shear; but this breakdown requires time, during which the rheological character changes. A typical time-dependent rheological phenomenon is thixotropy. Thixotropic fluids show a limited decrease in the shear viscosity of the fluid with time under a suddenly applied constant stress. Thixotropic fluids also show a hysteresis loop and a decaying stress on the shear stress-shear rate plot.

The most successful attempts at describing the steady state shear stress shear rate behavior of non-Newtonian fluids have been largely empirical. It would be much more satisfying if one could derive these functions from theories based upon molecular structure, but most materials of great interest are extremely complex and presently available molecular theories are largely oversimplified and generally inadequate for describing real fluid behavior. Since in this thesis we are mainly dealing with the power law model, a short discussion of the power law model is presented. For more information on the most common empirical models which have been utilized to represent the various classes of observed non-Newtonian behavior consult the book by Ronald Darby [1].

Power Law Model (Ostwald-deWaele)

$$\tau = k |\dot{\gamma}|^{n-1} \dot{\gamma}$$

This is a two parameter model where n is the power law index and k is the consistency index. For $n=1$, the fluid is Newtonian ($k = \mu$); for $n < 1$, the fluid is pseudoplastic (shear thinning) and for $n > 1$, the fluid is dilatant (shear thickening). Because the dimensions of k will depend upon the value of n , it is more correct to introduce a standard or reference shear rate ($\dot{\gamma}_p$, usually taken to be 1 sec^{-1}) so that the power law model can be written as

$$\tau = \eta_p \frac{|\dot{\gamma}|^{n-1}}{|\dot{\gamma}_p|} \dot{\gamma}$$

Here η_p has the dimensions of viscosity (e.g poise), and is in fact the value of the apparent viscosity at the reference shear rate. The apparent viscosity function for this model is

$$\eta_s = \eta_p \frac{|\dot{\gamma}|^{n-1}}{|\dot{\gamma}_p|}$$

The power law model is the most widely used of all models, since it is relatively easy to incorporate in analytical solutions to flow problems, and it can be made to fit the data over a limited range of shear rates. However it has two serious drawbacks. For constant values of n and η_p , it predicts unlimited increasing or decreasing of the apparent viscosity with shear rate; and it predicts either zero or infinite values of the limiting zero shear viscosity for n greater than or less than 1, respectively.

This chapter is an overview of some of the experimental methods which may be used to determine the various material functions. Specifically, consideration is given to the various methods for determining the viscous (apparent viscosity) and normal stress function functions in steady flows. There is no guarantee that a knowledge of all material functions discussed herein provides a complete description of a given viscoelastic fluid. Indeed, other material functions may be defined, such as the oscillatory response of normal stresses and transient stress or strain response. It is impossible to determine exactly how many material functions are sufficient to completely define the rheological behavior of a given material, since this depends largely on the complexity of the material. A typical approach is to postulate a general constitutive model relating stress to deformation and to evaluate experimentally how successful the model is in duplicating experimental data.

Throughout this thesis, the power law model is utilized because of its relative simplicity and its capability of fitting experimental data, at least over a limited range of shear rates, generally three decades or more. The review is not by any means exhaustive as this might require chapters if not books in order to successfully describe some of the observed behavior of polymeric solutions. For more in depth knowledge on any of the subjects discussed consult the references herein cited. Most of the time I present only the formulas that were used leaving most of the derivation out.

The chapter starts with a comparison between shear and extensional flow behavior of real fluids particularly polymer solutions. A definition is given for the normal or "extra" stresses that are encountered in viscoelastic liquids under shear. The various experimental methods for the determination of the extensional properties of fluids are also discussed. This should provide the reason why the contraction flow technique was chosen for the measurement of the extensional viscosity. The die swell technique is also utilized for the determination of the normal stress differences. For the measurement of the shear dependent viscosity, the two methods that were employed to obtain the apparent viscosity at an extensive range of shear rates are also discussed.

3.1 Extensional and Shear Behavior of Polymeric Solutions

3.1.1 Shear Behavior of Polymeric Solutions

Before proceeding with the description of the experimental flow configurations devised to measure the shear and extensional viscosity, some insight as to how a polymer solution can have a low viscosity in shear and a high viscosity in elongational flow is provided by considering the differences between these two types of flows. The familiar shear flow that is commonly produced in a long capillary tube is characterized by a velocity gradient that is normal to the direction of the flow. As a dilute polymer solution flows through a capillary, the polymer molecules tend to be stretched along the principal axis of stress. However, shear flows are rotational. Because of this, very little polymer deformation occurs with a low viscosity solvent even at high rates of shear, Bueche [2], 1960. The net effect is that in dilute solutions, the polymer molecules act more like rigid spheres than flexible coils. Therefore, the primary resistance to shear flow is derived from the viscosity of the solvent. In steady simple shearing the velocity field has the form:

$$U = c y; \quad V = W = 0$$

The particle trajectories are :

$$x(t) = x(0) + c y(0) t; \quad y(t) = y(0) \quad z(t) = z(0)$$

If this is the trajectory of one end of a molecule, the other end being fixed at the origin, then the molecule would become infinitely stretched (if $y(0) \neq 0$) and oriented parallel to the x,z -plane (the slip surface) in the limit of large times. Retractive forces in the polymer chain prevent infinite stretching, and Brownian motion sometimes moves the end of the polymer molecule into the region of negative velocity so that the actual motion of a chain is a tumbling motion with alternate stretching and contraction (Pipkin & Tanner [3], 1977).

3.1.2 Extensional Behavior of Polymeric Solutions

For some time now it has been evident that the shear flow properties of polymer melts and solutions is inadequate in describing many industrial flow situations. Flow moulding of film, fiber spinning from the melt or solution, vacuum forming of sheets, flow through porous media and atomization are all processes in which the extensional behavior is important. There are also theoretical reasons for studying the extension rather than shear flow of polymers, notably because the deformation made in the former is considerably simpler than in the latter. Unfortunately the same is not true experimentally, the difficulties in making extensional measurements with polymer melts and especially with polymer solutions being considerable.

Whereas the direct measurement of extensional viscosity of dilute polymer solutions is difficult, these solutions exhibit a very high flow resistance in porous media and in flows through contractions. This high resistance appears to be related to the uncoiling of the polymer molecules. There have been many attempts to estimate the extensional viscosity of mobile liquids. These attempts have been hindered by the difficulty in generating a purely extensional flow in mobile liquids. The aim is to generate flows with a high extensional component and interpret the data in a way which captures the extensional component in a very convenient and consistent way through a suitably defined extensional viscosity and strain rate, Binding [4].

Elongational or extensional flows occur in many practical situations such as the entrance region to a capillary tube, in flow through an orifice, and in porous media. In all these examples the velocity gradient is in the direction of the flow. Since this type of flow is irrotational except near solid boundaries, a polymer molecule is expected to be oriented in the stress field longer in elongational flow than in shear flow, therefore, larger polymer deformations should occur. Thus elongational flow resistance of a dilute polymer solution

is strongly dependent on the mechanical properties of the polymer, Mannheimer [5], 1983. Steady extensional flow has the following velocity field:

$$U = \dot{\epsilon} x; \quad V = -\dot{\epsilon} y/2; \quad W = -\dot{\epsilon} z/2$$

where $\dot{\epsilon}$ is the constant rate of extension. The particle trajectories are:

$$x(t) = x(0) \exp(\dot{\epsilon} t); \quad y(t) = y(0) \exp(-\dot{\epsilon} t/2); \quad z(t) = z(0) \exp(-\dot{\epsilon} t/2)$$

Molecules are powerfully stretched and oriented by such a flow. Brownian motion prevents perfect orientation, but it cannot lead to tumbling motion as shearing flow [3]. In the steady extensional flow of an isotropic liquid, all shearing stress components are zero and $\tau_{22} = \tau_{33}$, by symmetry. For an incompressible liquid the stress response is completely defined by the dependence of $\tau_{11} - \tau_{22}$ on the rate of extension and the time elapsed since the stretching began:

$$\tau_{11} - \tau_{22} = \dot{\epsilon} \eta_E(\dot{\epsilon}, t)$$

If the rate of extension is large, this initial stage may end in fracture of the specimen. When rupture does not occur, the stretching viscosity $\eta_E(\dot{\epsilon}, \infty)$ must eventually approach a limiting value $\eta_E(\dot{\epsilon}, \infty) = \eta_T(\dot{\epsilon})$. This limiting value is called the steady extensional viscosity, or the Trouton viscosity. Trouton, [6] (1906), found that the extensional viscosity of mixtures of pitch and tar is independent of $\dot{\epsilon}$ and about equal to $3\eta_0$, the value for an incompressible Newtonian fluid with shear viscosity η_0 .

The experimental results of all extensional flow techniques are often expressed in terms of the ratio of the apparent extensional viscosity η_E to the steady state shearing viscosity η_s at a shear rate equal to the rate of extension. It is generally agreed that the observed increases in extensional viscosity are caused by the alignment of the long-chain polymer molecules along the direction of stretching. Both extension and orientation grow larger as the rate of extension increases, and both effects increase the extensional viscosity of the solution. To understand the effect of orientation alone, as modified by Brownian motion, one may consider the results of rigid dumbbells obtained by Bird et al [7], (1970). They found that the excess in the Trouton viscosity over that of the solvent increases by a factor of two as the extension rate increases from zero to infinity.

There is a much greater increase in the extensional viscosity because the distance from one end of the molecule to the other grows larger as the rate of extension increases. This

effect alone can be understood by considering the viscosity of a suspension of rigid, needle-shaped particles, fully aligned along the direction of stretching. Bachelors [8,9] (1970, 1971), has obtained theoretical expressions for the extensional viscosity of dilute and concentrated suspensions of such particles in Newtonian liquids. When the distance between the molecules is small in comparison to their length, the excess of the Trouton or extensional viscosity over that of the solvent, $(3\eta_0)$, is given approximately by:

$$(\eta_T - 3\eta_0)/3\eta_0 = (4/9) (L/D)^2 C / \ln(\pi/C)$$

where L/D is the length to diameter ratio for the particles and C is their volume concentrations. The length to diameter ratio of a randomly coiled polymer chain is effectively unity, when fully extended by an extensional flow field, it can be enormous. Mewis and Metzner, [10] (1974), tested several suspensions with different volume concentrations and particle aspect ratios, and found good agreement with the theory. The highest value of $\eta_T/3\eta_0$ observed was about 260 (with η_0 the shear viscosity of the suspending agent) for particle to diameter ratio of 586 at a volume concentration of about 1%.

3.2 Normal Stress Effects

There are a number of unusual, and rather remarkable, effects exhibited by viscoelastic liquids in steady flow which cannot be attributable to viscous (shear stress) effects. Some of these are illustrated in Fig. 3-2 [1]. In Fig. 3-2a, the shape of the free surface of a liquid in a vessel is depicted when it is subjected to an angular deformation by a rotating rod or cylinder. For fluids without elasticity, the centrifugal force creates a concave vortex in the liquid. However elastic liquids develop stresses which oppose that of the centrifugal force and cause the fluid to climb up a rotating shaft. This is known as the Weissenberg effect. Figure 3-2b illustrates the shape of a free jet issuing from the end of a tube. The jet of an inelastic liquid contracts slightly from the diameter of the tube, while that of an elastic liquid may swell to a diameter that is considerably larger than the tube diameter. In Fig. 3-2c, two parallel discs are immersed in a fluid. One of them has a hole in the center, and the other disc is rotating. For an inelastic liquid, the centrifugal force causes the fluid to flow through the hole and out at the edge, while the elastic liquid is in at the periphery and out through the hole in the center.

These phenomena can be attributed to the "extra" stresses which are developed in elastic fluids under shear, in direction normal to the plane of shear. These normal stresses, are not

isotropic, and are function of the rate of shear and thus disappear when the fluid is at rest. These stresses are illustrated in Fig. 3-3 for the case of simple shear, and are attributed to elasticity inasmuch as they are absent in purely viscous (inelastic) liquids in this type of deformation. The three anisotropic normal stress components are designated here by τ_{11} , τ_{22} , and τ_{33} .

The exact molecular origin of these normal stresses is not entirely understood. However, a qualitative, but considerably oversimplified, explanation is possible in terms of elastic properties. If a fluid (e.g., a polymer solution) is envisioned to be comprised of elastic segments of "rubber bands" which are stretched or extended in the direction of shear, the resulting tensile force can explain many of the observed phenomena. For example, if the fluid is sheared in a rotational flow between concentric cylinders, the contraction force due to the stretched element acts inwards towards the axis of rotation, which forces the fluid in towards the axis. This results in the fluid climbing up the inner cylinder, the Weissenburgh effect. Likewise, the relaxation of the elastic forces when the fluid exits from the open end of a tube in a free jet causes the elastic elements to contract, or "pile up". This would cause the jet to swell when the stresses relax. Thus, one effect of these normal stresses is an additional tensile force in the direction of shearing. More on this in this next paragraph which includes an overview of the normal stress material functions.

The anisotropic normal stress components, illustrated in Fig. 3-3 cannot be measured directly in a fluid system. Measurements can be made, however, of the total stress components. In general for an incompressible liquid the total stress (σ_{ij}) includes contributions from both (isotropic) pressure (P) as well as the fluid shear (anisotropic) stress (τ_{ij}). The total stress tensor can be expressed as follows:

$$\sigma_{ij} = -P \delta_{ij} + \tau_{ij} = \begin{bmatrix} -P + \tau_{11} & \tau_{12} & \tau_{13} \\ \tau_{21} & -P + \tau_{22} & \tau_{23} \\ \tau_{31} & \tau_{32} & -P + \tau_{33} \end{bmatrix}$$

where δ_{ij} is the unit tensor or Kronecker delta, with components

$$\delta_{ij} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix} = \begin{cases} 1 & \text{if } i = j \\ 0 & \text{if } i \neq j \end{cases}$$

In addition we know that for an incompressible fluid the static pressure and the mean normal stress are identical

$$P = -\frac{1}{3} \sum_i \sigma_{ii} \quad (3.1)$$

Therefore, in order to eliminate the contribution of the static pressure to the total normal stress components, resulting in stresses which are characteristic only of the deformation properties of the fluid, only the differences in normal stresses are considered.

$$\sigma_{11} - \sigma_{22} = (-P + \tau_{11}) - (-P + \tau_{22}) = \tau_{11} - \tau_{22} \quad (3.2)$$

Thus, the normal stress character may be completely defined by knowledge of two normal stress differences as a function of deformation (e.g, shear rate). The third equation needed to define the normal stress character is a consequence of the definition of the isotropic pressure.

$$\text{First Normal Stress Difference} = \tau_{11} - \tau_{22} = N_1 \quad (3.3)$$

$$\text{Second Normal Stress Difference} = \tau_{11} - \tau_{33} = N_2 \quad (3.4)$$

$$\text{Pressure Definition: } \tau_{11} + \tau_{22} + \tau_{33} = 0 \quad (3.5)$$

Note that Eq. (3.5) is a consequence of Eq. (3.1), namely

$$P = -\frac{1}{3} \sum_i \sigma_{ii} = (\sigma_{11} + \sigma_{22} + \sigma_{33}) = -\frac{1}{3} (-P + \tau_{11} - P + \tau_{22} - P + \tau_{33})$$

$$P = P + \frac{1}{3} (\tau_{11} + \tau_{22} + \tau_{33})$$

3.3 Experimental Methods for Determination of Extensional Viscosity

Since Trouton [6] carried out his pioneering work on extensional flow, establishing that for Newtonian fluids the extensional viscosity is 3 times the shear viscosity, a variety of techniques have been employed to measure the extensional viscosity of non-Newtonian fluids. In order to measure the steady state extensional viscosity $\eta_T(\dot{\epsilon})$, the fluid must be subjected to a constant rate of extension for so long that the stress reaches a constant value, or a constant stress for so long that the rate of extension becomes constant. Most reported cases in which it is certain that a steady state was reached involve fluids of very high viscosity and rates of extension not greater than 0.1 sec^{-1} . In this limited range the Trouton

viscosity has invariably been found to be either nearly constant or at the highest rates of extension an increasing function of the stretch rate $\dot{\epsilon}$.

Although these rates of extension are extremely low, the steady state shearing viscosity $\eta_s(\dot{\gamma})$ of the same material are usually significantly lower at a shear rate of $\dot{\gamma} = 0.1 \text{ sec}^{-1}$ than they are in the limit of zero rate. Consequently, the ratio of the extensional viscosity to the shearing viscosity is an increasing function of the strain rate even when the Trouton viscosity is still constant. It is relatively easy to obtain much higher rates of extension, but the experiments in which this done always involves flows with a non-constant rates of extension. Spinning experiments usually involve extension rates of the order $\dot{\gamma} = 0.1$ to 10 sec^{-1} . The convergent die-entry flows studied by Cogswell [11] (1969), involve extension rates of order 1 to 100 sec^{-1} . Jet thrust experiments involve extension rates of order 10 to 10^3 sec^{-1} . Rates in the range 100 to 800 sec^{-1} are obtained in the triple jet apparatus proposed by Oliver and Bragg [12], (1974).

The various experimental techniques for measuring the extensional viscosity fall naturally into headings of controllable experiments (where one or more parameters are fixed and the others measured) and non-controllable experiments (where all parameters need to be measured). The parameters involved are usually applied force, velocity, stress, strain and rate of strain.

By their very nature, controllable experiments can only be conducted with polymer melts. Meissner [13] (1983) has reviewed experimental aspects of polymer melt extensional rheometry. The techniques available all involve the preparation of homogeneous isotropic sample in a fully relaxed state. The sample is glued to small metal clamps which are then pulled apart at constant force, constant velocity, constant strain, constant stress, or constant strain rate.

Non-Controllable experiments fall broadly into two groups:

1. For an accelerating jet of fluid, measurements are taken of applied force and jet profile to determine the kinematic parameters.
2. For a fluid passing through a contraction such as a hole or a slit at the end of a reservoir, or through a bed of porous medium, measurements are taken of pressure difference and angle of convergence to determine the kinematic parameters.

These methods have been used for polymer melts, solutions and suspensions. Group 1 consists of three basic geometries. Firstly, a filament of polymer solution or polymer melt is extruded from a die, where it falls under gravity to a take-up device. This is usually a drum or a roller which can also apply an elongating force. Measurements are taken of the force either at the die or take-up roller. As there is no control of rate of elongation along the threadline, the filament diameter profile must be measured. A changing rate of elongation means that the extensional stresses generated are transient.

The second type of experiment involves a fluid filament being drawn upwards against gravity into a capillary from a reservoir of fluid, Chao [14] (1983), and often called tubeless syphon. Measurements of pressure difference and filament diameter are taken to calculate the transient values of the stress and strain rate. The third geometry involves a jet of fluid being extruded horizontally from a nozzle, and either being extended by two other jets impinging on it (the triple jet technique [12]), or by oscillating the nozzle longitudinally, causing instability in the jet, thereby forming spherical droplets between which the fluid is elongated, Schummer (1982) [15].

A typical geometry of group 2 type of experiments is shown in Fig. 3-4. It is observed (Cogswell, 1972 [16] and Nguyen et al. 1979 [17]) that for viscoelastic liquid the flow is usually divided into two regions. The first, which is usually conical has a high extensional flow component, whilst the second is a closed region of recirculating flow. The kinematics of the flow are determined from the cone angle, the flow rate, and the jet thrust from the capillary or pressure drop across the contraction. A good review of these techniques has been given by Cogswell 1978, [18].

For this thesis the contraction flow technique is utilized to investigate the extensional properties of our polymer solutions. Thus a complete description of this method is included in the next section. For more information consult the paper by Binding [4]. The die swell technique proposed by Metzner and co-workers [19] is also used in order to calculate the normal stresses differences. Next section also summarizes the various experimental methods used for determination of the shear viscosity.

3.4 Shear and Extensional Viscosity Measurement.

3.4.1 Shear Viscosity Measurements

For the measurement of shear dependent viscosity, two methods were employed to obtain apparent viscosity data at an extensive range of shear rates. At low shear rates ($0.067 < \dot{\gamma} < 13.4 \text{ s}^{-1}$), the shear dependent viscosity was measured by a concentric cylinder viscometer, the Brookfields viscometer. The mode of operation is to rotate the inner cylinder (spindle) with angular velocity Ω and measure the torque T applied on the spindle. For measurements of the shear dependent viscosity at higher strain rates ($14 < \dot{\gamma} < 20000 \text{ s}^{-1}$), pressure was applied to two capillary tube viscometers operating in a differential mode and the volumetric flow rate Q_v was measured along with the differential pressure ΔP . Expressions were derived to describe the flow conditions present in the viscometers.

3.4.1.a Concentric Cylinder Viscometer:

This flow system involves an angular (rotational) flow in the annulus between concentric cylinders. The general configuration is shown in Fig. 3-5. Figure 3-5 involves a stationary outer cylinder and a moving inner cylinder or spindle. The torque T may be measured at either cylinders. The flow field is described in details in Ref.[1]. The exact expression for the shear rate distribution as a function of radial position for a power law fluid is given by

$$\dot{\gamma}(r) = \frac{2 \Omega \phi^{2/n}}{n (\phi^{2/n} - 1)} \left[\frac{R_i}{r} \right]^{2/n} \quad (3.6)$$

where

$\dot{\gamma}(r)$ = shear rate at any radius r

$\phi = R_o/R_i$

Ω = angular velocity of inner cylinder

n = power law index

R_o = radius of outer cylinder

R_i = radius of inner cylinder

Thus, the shear rate at the inner cylinder is given by

$$\dot{\gamma}(R_i) = \frac{2 \Omega \varphi^{2/n}}{n (\varphi^{2/n} - 1)} \quad (3.7)$$

The shear stress at any surface in the liquid is given by

$$\tau_{r\theta} = \tau(r) = \frac{T}{2 \pi L r^2} \quad (3.8)$$

Thus, the shear stress at the inner cylinder is given by

$$\tau(R_i) = \frac{T}{2 \pi L R_i^2} \quad (3.9)$$

where T is the torque required to overcome the viscous resistance of the fluid and L is the length of the spindle. For a true power law fluid, the power law index, n, can be determined from the slope of Log(T) versus Log(Ω) curve, which is a straight line. For general non-Newtonian fluids, for which this plot is not linear, the point slope n', will still provide a good estimate of the appropriate value of n, but of course it will vary with T (or Ω) [1].

The apparent viscosity for the concentric cylinder viscometer can be defined as the ratio of the shear stress encountered by the inner cylinder (spindle) to the shear rate evaluated at the spindle surface. Hence, a direct measurement of the torque T and the angular velocity Ω of the spindle can be used to express the apparent viscosity. The apparent viscosity for a power law fluid is given by

$$\eta_a = \frac{\tau(R_i)}{\dot{\gamma}(R_i)} = \frac{T n (\varphi^{2/n} - 1)}{4 \pi L R_i^2 \Omega \varphi^{2/n}} \quad (3.10)$$

Equation (3.10) reduces to the corresponding Newtonian viscosity by setting n = 1 and $\eta_a = \mu$, where μ is the fluid's kinematic viscosity. For this case we get

$$\mu = \frac{T (\varphi^2 - 1)}{4 \pi L \Omega R_o^2} \quad (3.11)$$

Equation (3.11) is only valid for Newtonian fluids. If the material is non-Newtonian, the correction factor which converts Eq. (3.11) to Eq. (3.10) is

$$\frac{\mu}{\eta_a} = \frac{(\alpha^2 - 1) \alpha^{2(1-n)/n}}{n(\alpha^{2/n} - 1)} \quad (3.12)$$

where, again, n is the power law index and can be taken as the point slope of $\log(T)$ versus $\log(\Omega)$ curve at the value of T corresponding to $\tau(R_i)$.

3.4.1.b) Capillary Viscometer:

The steady laminar flow in a tube of material whose viscosity depends on the shear rate (i.e., thixotropic time effects are negligible) must be a unique function of the consistency parameters $R\Delta P/2L$ and $4V/R$, where R is the inside radius of the tube, L is the length of the tube, ΔP is the magnitude of the pressure drop and $V = Q_v/A$ is the average velocity (Frederickson [20], 1964). The quantity $R\Delta P/2L$ is the magnitude of the shear stress at wall of the tube for any material. However, $4V/R$ or equivalently $8V/D$ (where $D = 2R$) is the shear rate at the wall ($\dot{\gamma}_R$) only for a special class of fluids (i.e. Newtonian). In all other instances, the shear rate at the wall is given by:

$$\dot{\gamma}_R = \frac{3+1/n'}{4} \frac{8V}{D} \quad (3.13)$$

where

$$n' = \frac{d \log R \Delta P/2L}{d \log 8V/D} \quad (3.14)$$

is the flow index. It is important to note that n' describes the shear dependency of a material. For example, $0 < n' < 1$ indicates that the viscosity is a decreasing function of the shear rate (i.e., shear thinning), while $n' > 1$ indicates that the viscosity is an increasing function of the shear rate (i.e., shear thickening) and $n' = 1$ indicates that the viscosity does not change with shear rate.

The apparent viscosity (η_a) of a material is defined by the ratio of the wall shear stress to the wall shear rate:

$$\eta_a = \frac{R \Delta P/2L}{\dot{\gamma}_R} = \frac{R \Delta P/2L}{\left[\frac{3n'+1}{4n'} \right] \frac{8V}{D}} \quad (3.15)$$

Similarly, an effective viscosity (η_e) can be defined simply by

$$\eta_e = \frac{R \Delta P/2L}{4 V/R} \quad (3.16)$$

which is the viscosity the fluid would have if it were Newtonian. Plots of the shear stress versus apparent (Newtonian) shear rate are in general curved and the slope n' at each point has to be computed. For a true power law fluid n' is expected to be constant; n' in this case is identical to n (the power law index) [1].

3.4.2 Extensional Viscosity Measurement

3.4.2.a Contraction Flow Technique

Cogswell [11, 16, 18] was perhaps the first to recognize the importance of extensional viscosity in contraction flows. He provided a simple analytical analysis, which involved splitting entry flow into separate shear and extensional components, and he was able to estimate the extensional viscosity of polymer melts from entry pressure data. Cogswell's analysis was, strictly speaking, incorrect since he used a shear rate based on a Newtonian fluid in conjunction with a velocity field determined for a power law fluid.

Binding[4] drew heavily on Cogswell's ideas in the belief that "the underlying philosophy behind the analysis is sound". The analysis aimed at investigating the influence of the extensional viscosity on such flows, especially with regard to the occurrence and enhancement of vortex motion in the entry corners. Binding concluded that once significant vortex motion is present, enhancement occurs whenever the Trouton ratio is an increasing function of shear rate or stretch rate. Indeed, the analysis showed that the occurrence of vortices serves as a stress relief mechanism.

Binding[4] presented an approximate analysis for the flow of fluids through planar and axisymmetric contractions. Energy principles were employed to relate the pressure drop to the flow rate and fundamental rheometric properties. Binding's treatment idealizes the flow characteristics to that shown in Fig. 3-4 for the axisymmetric case. The fluid is assumed to flow through the "funnel" defined by the surface, $r=R(z)$ which is such that the convergence angle Θ is small and slowly varying. The axial velocity field is taken to be that corresponding to fully developed flow in a tube of radius R whereas the radial velocity is assumed to be negligible, this being consistent with the restriction on $R(z)$.

In general the components of the stress field for this flow may be expressed as follows:

$$\sigma_{ij} = \begin{bmatrix} -P + \tau_{rr} & 0 & \tau_{rz} \\ 0 & -P + \tau_{\theta\theta} & 0 \\ \tau_{rz} & 0 & -P + \tau_{zz} \end{bmatrix}$$

The first and second normal stress differences are defined by

$$N_1 = \tau_{zz} - \tau_{rr} \quad (3.17)$$

$$N_2 = \tau_{\theta\theta} - \tau_{rr} \quad (3.18)$$

In this case the second normal stress difference N_2 is considerably less important than N_1 because of the quasi-unidirectional character of the flow and therefore it was neglected in the analysis [4]. The components of the stress field are then interpreted in terms of the shear viscosity $\eta_s(\dot{\gamma})$ and an extensional viscosity function $\eta_E(\dot{\epsilon})$ both of which are then expressed as power laws of their respective arguments:

$$\tau_{rz} = \eta_s(\dot{\gamma}) \dot{\gamma} \quad (3.19)$$

$$N_1 = \tau_{zz} - \tau_{rr} = \eta_E(\dot{\epsilon}) \dot{\epsilon} \quad (3.20)$$

$$\text{Extensional Viscosity: } \eta_E(\dot{\epsilon}) = l \dot{\epsilon}^{t-1} \quad (3.21)$$

$$\text{Shear Viscosity: } \eta_s(\dot{\gamma}) = k \dot{\gamma}^{n-1} \quad (3.22)$$

where η_s is the shear viscosity corresponding to a shear rate $\dot{\gamma}$ and η_E is the extensional viscosity corresponding to a stretch rate $\dot{\epsilon}$. Flow behavior is represented by the indices n and t ; flow consistency is represented by the constants k and l .

Basic variational principles are used in order to determine the optimum profile, that corresponding to the least power consumption in the converging section. After evaluating and minimizing the viscous dissipation in the converging region, an energy balance yields the following expression for the "entry pressure". The entry pressure is the pressure in excess of that required to produce fully developed conditions over the upstream and downstream geometries.

$$P_e = \frac{2k(1+t)^2}{3t^2(1+n)^2} \left(\frac{lt(3n+1)n^t I_{nt}}{k} \right)^{1/1+t} \dot{\gamma}_{R0}^{t(n+1)/1+t} \{1 - \alpha_c^{3t(n+1)/1+t}\} + \frac{3\rho Q_v 2(3n+1)2(1-\alpha_c^4)}{2(2n+1)(5n+3)\pi^2 R_0^4} \quad (3.23)$$

where

$$\dot{\gamma}_{R_0} = (3n+1)Q_v / n\pi R_0^3 \quad (3.24)$$

is the wall shear-rate in a capillary of radius R_0 , and

$$\alpha_c = \frac{R_0}{R_1}$$

is the inverse of the contraction ratio. Q_v is the volumetric flow-rate and I_{nt} is determined by

$$I_{nt} = \int_0^1 \left[\text{abs} \left\{ 2 - \left(\frac{3n+1}{n} \right) \phi^{1+1/n} \right\} \right]^{t+1} \phi d\phi$$

The first and second terms on the right hand side of Eq. (3.23) are contributions from both shear and extension, and inertia respectively. Note that Eq. (3.24) is identical to Eq.(3.13).

Procedure for obtaining the extensional viscosity power law parameters.

From the entry pressure equation a plot of $\text{Log}(P_e - \text{inertia term})$ against $\text{Log}(Q_v)$ is expected to have a constant slope of : $t(n+1)/(1+t)$. Knowing n from the shear viscosity data, t may thus be determined. A simple numerical algorithm is then employed to evaluate the integral I_{nt} . Finally, l is obtained from the straight forward application of the entry pressure equation. As an auxiliary test to the theory, vortex lengths are measured from the photographs of the flow field. The vortex attachment length L_v predicted by the analysis is given by the following equation:

$$\frac{L_v}{2R_1} = \frac{(1+t)}{2(n+1)(2t-3n-1)} \left\{ \frac{l(3n+1)n^t I_{nt}}{k} \right\}^{1/(1+t)} \cdot \frac{(t-n)(1+t)}{\dot{\gamma}_{R_0}} \left\{ 1 - \alpha_c^{(2t-3n-1)(1+t)} \right\} \alpha_c \quad (3.25)$$

3.4.2.b Die Swell Technique

In order to calculate the normal stresses from die-swell, the following relationship proposed by Metzner and co-workers [19] is used. The arrangement is similar to that shown in Fig. 3-2b. Please refer to Fig. 3-2b for the definition of the various variables.

$$N_1 = \tau_{11} - \tau_{22} = \frac{\rho D^2 (8V/D)^2}{64 n'} \left\{ (n'+1) \left(\frac{3n+1}{2n+1} \right) - \left(\frac{D}{d_j} \right)^2 \left[n'+1 + \frac{d \log D/d_j}{d \log 8V/D} \right] \right\} \quad (3.26)$$

where n' is the flow index, n is the power law index. Measurements of the die-swell ratio (D/d_j) along with the volumetric flow rate are used to evaluate the normal stress differences. The applicability of Eq. (3.26) to calculate the normal stress difference has been questioned by Boger and Denn [21]. Of particular concern is the need for information on the rearrangement of the velocity profile at the exit of the tube. The use of Eq. (3.26) to calculate the normal stresses was generally limited to conditions for which the flow index was constant so that $n = n'$ and high Reynolds numbers ($100 < Re = \rho VD/\eta_e < 500$).

Fig. 3-1 Shear Stress vs Shear Rate for Time Independent Fluids

Viscous

Viscoelastic

Fig. 3-2a

Viscous

Viscoelastic

Fig. 3-2b

Viscous

Viscoelastic

Fig. 3-2c

Fig.3-2 Behavior of Viscoelastic Liquids in Simple Shear

Fig.3-3 Anisotropic Normal Stress in Simple Shear

Fig.3-4 Schematic of Contraction Flow

Fig.3-5 Concentric Cylinder Viscometer

Chapter 4

EXPERIMENTAL APPARATUS

The principal elements of the experimental apparatus are the following:

- **Liquid delivery system:**
 - Weighed mixing tank
 - Mixer
 - Mono cavity feed pump
 - Micro-motion mass flow meter
 - Pressure Transducers
 - Pressure regulators
 - Solenoid Valve
 - Co-axial atomizer (Extensional viscometer)
 - Spray chamber
 - Collecting tank
 - Recirculating pump
- **Air delivery system:**
 - House or compressor air
 - Solenoid valve
 - Filters, regulators and pressure transducers
 - Thermal mass flow meter
 - Rotameters and pressure gauges
 - Exhaust system
- **Viscometers:**
 - Concentric cylinder viscometer (Brookfield)

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Side-stream capillary viscometer (House made)
Extensional viscometer (House made)
Die-swell setup (House made)

- **Optical system:**

Phase Doppler particle analyzer
Greenfield digital spray analyzer
Microflash
35 mm camera
Optical diffusers and windows

- **Electronics:**

Charge amplifier
Microflash controller
Oscilloscope
Power Supply
Ammeters
Voltsmeters

4.1 System Design and Operation

The air supply system is shown in Fig. 4-1. Sources of the atomizing air are either the house air which supplies air at pressures up to 90 psig or a compressor which delivers air up to 180 psig. The atomizing air passes through a 2.5 cm diameter line containing a control valve, filter, regulator and a pressure transducer. The thermal mass flow meter is used for accurate mass flow rate measurements. The air line then splits into two identical streams, each containing a pressure regulator and a pressure gauge. Each of the streams is further subdivided into two streams, thus forming four identical air lines to the atomizer. Each of the lines contains a rotameter and a pressure gauge for flow monitoring. The final connection to the co-axial atomizer (which is shown in Fig. 4-3) is provided by flexible quick-connect hoses having internal diameters of 1.25 cm. The four air jets flow radially into a converging channel through four diametrically opposite inlet ports.

A schematic of the fuel delivery system is shown in Fig. 4-2. The fuel delivery system was designed to supply a pulsation free flow. It can operate in both the continuous and intermittent modes. A second fuel line was installed between the supply tank and the slurry flow meter. This was done in order to by-pass the fuel delivery pump. The co-axial

atomizer (extensional viscometer) is rated at 30 psig for the chamber pressure. In order to protect the atomizer from pressure buildup in case of blockage in the injection orifice, a bypass around the pump is required. The second fuel line is driven by compressed house air, regulated at 30 psig, maximum pressure. A Micro-Motion model D-6 slurry flow meter is used for accurate mass flow rate measurements.

4.2 The Co-Axial Atomizer and Extensional Viscometer

The co-axial atomizer and extensional viscometer is shown in Fig. 4-3. This atomizer was specifically designed to allow measurement of the entry pressure at different locations in the cylindrical chamber. This chamber has an internal diameter of 2.75 in and a total length of 10 in. Two circular plates, with large diameter bores (i.e., 3.5 in), are glued at each end of the cylindrical chamber, thus providing a leak proof, rigid assembly. Both the chamber and the plates are made of plexi-glass. All other parts are made of aluminum. Extruding from the chamber is a 3 mm inside diameter tube. The tube is epoxied in place to a large disc, and is centered by a profiled cone. The air flows radially into a converging channel through four diametrically opposite inlet ports. Extreme care was taken in order to make a smooth contour and minimize flow disturbances. Pressure is measured by 4 U-tube manometers at four locations. The air channel ends with a circular orifice having an ID of 0.5 in. The total contraction ratio is 70:1. The air channel is topped with a profiled cap. Both the cone and the air channel cap have the same profile. All parts are centered by centering pins. Leaks are sealed by cork gaskets. Each end plate is mounted to the chamber body by 6 bolts. Extra support to the total assembly is provided by four rigid poles. The poles are fixed to a mounting plate, which in turn is mounted to a bracketed assembly. The mounting support is made of rigid I-beams. The total unit (i.e. chamber, nozzle, mounting support) is fixed to a rugged three dimensional traversing mechanism.

A solenoid valve, and a pressure transducer were installed upstream of the atomizer for intermittent fuel injection. This was necessary in order to accommodate the piezoelectric transducer's operation mode requirements. The transducers operate in dynamic or quasi-static modes, only. Most of the operations are conducted in the quasi-static mode.

The transducers setup configuration is shown in Fig. 4-4. Four holes, of diameters 0.5 in, were tapped in the positions indicated to accommodate "flushmounted" transducers and adapters. The transducers are of the piezoelectric type and have diameters of 6 mm. The purpose of the charge amplifiers is to convert the electric charge generated by the transducers into a voltage. Short-term static and dynamic pressure acting on the diaphragm

becomes a proportionate compressive force in the quartz sensing element, producing a high impedance electrostatic charge signal. Measurements in the low frequency range, termed quasi-static measurements, are made with these transducers. The lower cutoff frequency mainly depends on the properties of the non-ideal charge amplifier, and the insulation resistance of the quartz. Since all charge amplifiers have a finite insulation resistance, and therefore a finite cutoff frequency, purely static measurements with no lower cutoff frequency limit, are inherently impossible. By adjusting the amplifier's time constant, τ , to long range, the voltage drift problems can be controlled, thus allowing quasi-static measurements to be made. For quasi-static measurements using quartz transducers, the measuring time from the onset of loading should not exceed 0.05τ , for which there is an error of 5%. If the admissible discharge error is 1%, the measuring time must not exceed 0.01τ .

4.3 The Concentric Cylinder Viscometer

The concentric cylinder viscometer operates by measuring the torque applied to a rotating spindle submerged in a beaker containing the liquid to be tested. The spindle is attached directly to a pre-calibrated torsion spring. The whole assembly rotates as unit. By rotating the spindle at different angular velocities, variable shear rates can be generated. Once an equilibrium state has been reached at which the torque applied balances the viscous resistance, the equilibrium torque can be measured at a given angular spindle velocity. To obtain the viscosity of the fluid, one uses a calibration chart, either obtained from the instrument manufacturer or experimentally determined, to multiply the dial torque by the calibration factor.

For the Brookfield viscometer, the outer cylinder was a stationary beaker while the inner cylinder was a rotating spindle. Figure 3-5 is diagram of the Brookfield concentric cylinder viscometer. The spindle used was the model LV1. This viscometer had speed settings which could generate eight shear rates ranging from 0.067 to 13.4 s^{-1} . The torque required to rotate the spindle was measured by a dial reading on the viscometer. The instrument had a viscosity measurement range 1 to 20,000 cP.

Remarks Concerning Rotational Viscometers:

The following are some general remarks on the use of rotational viscometers and some factors that affect their ideal behavior. These factors include the gap between the rotating surfaces, the onset of turbulence in the test section of the viscometer, and end effects. The

formulas presented in Section 3.4.1a presuppose that the torque is a single valued function of the angular velocity. Thus this technique cannot be used when properties vary with time of shearing or with the previous shear history. It is also desirable to use the narrowest possible gap both to minimize the uncertainties in the strain rate and minimize the difference in shear history between material near the inner and material near the outer cylinder during the test. Since materials in different paths of the annulus will have experienced different shear stresses, only average properties at the time of taking a reading can be determined. Care must also be exercised that turbulent motion does not occur. It is clear that the definition of viscosity implies the existence of a laminar flow. In turbulent flow a larger energy input is required to maintain the motion. This increased energy input is reflected by a greater apparent viscosity. To minimize the influence of end effects, the torque at any given rate of rotation must be measured with the inner cylinder (spindle) immersed at varying heights of the fluid. The graph of torque vs. immersed length should be linear, and the slope gives the torque per unit length. The intercept on the torque axis gives the torque on the lower end of the inner cylinder.

4.4 Capillary Viscometers

Side stream capillary tubes are utilized for the determination of high shear rate region of the viscosity curve (Fig. 4-5). In this type of viscometer, shear rates in excess of 10^4 s^{-1} can be accomplished. In order to eliminate the errors associated with the velocity adjustment region in the entrance and exit to the capillaries (especially in the case of visco-elastic liquids, where swelling occurs) a differential pressure measurement system is utilized. The side stream capillaries are installed up stream of the contraction flow chamber. The capillary tube viscometer was supplied with liquid by way of a side stream originating near the atomizer (Fig. 4-3). After accumulating a quantity of liquid in a smaller tank, liquid was driven through the capillary tubes by compressed air. A highly accurate differential pressure transmitter was used to obtain the pressure drop across two points on each capillary tube. The volumetric flow rate was obtained by measuring the volume of liquid issued from a tube in a given amount of time. Capillary tube diameters were selected to obtain a small overlapping region of shear rates. The highest liquid flow rates through the largest diameter tube corresponded to the wall shear rates experienced by the smaller tube at relatively lower flow rates. Hence, a continuous range of shear rates was obtained from 10 to $21,000 \text{ s}^{-1}$. The large and Small tube diameters were 4.39 mm and 1.80 mm, with a length to diameter ratio of 80 and 189, respectively

To ensure that fully developed laminar flow was present in the capillary tubes, sufficient tube extensions were provided for entry length upstream of the high pressure taps. Tube extension was also used downstream of the low pressure taps to prevent air from rushing into the capillary tube exit during operation. The tubes were situated horizontally with their exits exposed to the atmosphere.

4.5 The Die-Swell Setup

The apparatus shown schematically in Fig. 4-6 was used to measure the die-swell at high shear rates. A digital image analyzer with a close-up lens was positioned near the outlet of the capillary tube to measure the jet diameter. The measurement were performed by printing a hard copy of the computer screen; the diameter of the jet was subsequently measured on the printed image. The jet diameter and volumetric flow rate were measured at different flow conditions.

4.6 The Phase Doppler Particle Analyzer

The Phase Doppler (PDPA) light scattering technique developed by Bachalo and Houser [22] for spray analysis was used to make measurements of joint droplet size-velocity distribution functions in the sprays (Fig. 4-7). These measurements are made in situ and non-intrusively which is important when measuring easily deformable particles or probing turbulent flow fields. The PDPA instrument is an extension of the laser Doppler velocimeter. The PDPA provides many useful parameters for spray characterization including temporal and spatial mean drop size, local drop size distribution, drop velocity, volume flux, and number density.

4.7 The Greenfield Digital Spray Analyzer

A digital image analyzer (Greenfield) was used to make droplet size measurements. The instrument setup and optical configuration is shown in Fig. 4-6. The video imaging system includes: a CCD camera, microscope lens, fast strobed light, image processing hardware and software. The light source is on the opposite side of the jet from the CCD camera, so that the camera records back-lit images. The strobe light is synchronized with the camera and emits a flash of about 200 nanosecond duration, just before the camera begins a frame scan. The camera scans two 240-line fields and interlaces them to produce a 480-line frame. The time duration for a full frame is 33 milliseconds. Drops appear as dark objects on a white background. A digitizer on the image processing board turns each

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picture into an array of 640 by 480 pixels, each of which has a gray scale level of 0 (black) to 255 (white). The frame scan algorithm sums up the dark pixels in each drop representation, and computes a pixel diameter based on that area, assuming circularity. The software transforms the pixel diameter into a diameter in microns on the basis of a prior calibration using objects of known size. For non spherical droplets, the software reports the diameter of a circle of an equivalent area. Often the droplets, acting as spherical lenses, contain a white center or hole due to light refraction effects. The scanning algorithm effectively fills in these holes during the computation of area. At any one particular optical setting, the dynamic range of the instrument can extend to 150:1, with an overall range covering 1 to 18000 μm .

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Fig. 4-1 Air Supply System

Fig.4-2 Liquid Supply System

Fig.4-3 The Co-Axial Atomizer Extensional Viscometer

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Fig.4-4 Transducer Setup Configuration

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Fig.4-5 Side Stream Capillary Viscometer

Fig.4-6 Die-Swell Setup

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Fig.4-7

The Phase Doppler Particle Analyzer

Chapter 5

RHEOLOGICAL MEASUREMENTS

Non-Newtonian viscosities were measured over five decades of shear rates $\dot{\gamma}$ for 12 solutions of polymeric materials. By using the Brookfield viscometer and the capillary tube viscometers, the shear viscosity was determined as a function of shear rate. Both the upper and lower limiting shear viscosities were determined for all solutions. By using the die-swell technique, the first normal stress difference was determined for all solutions. The contraction flow technique was also used for the determination of the extensional viscosity. For all relevant equations and a short description of the techniques consult Chapter 3. For a description of the experimental setup consult Chapter 4.

The Non-Newtonian liquids selected for the experiment were aqueous solutions of Xanthan Gum and Polyacrilamide E10 polymers. Tests were performed on 0.05, 0.10, 0.15, 0.20, 0.30% concentrations by mass for the Xanthan Gum polymer, and 0.0125, 0.025, 0.05, 0.1, 0.15, 0.30, and 0.50% concentrations by mass for the Polyacrilamide E10. Xanthan Gum and Polyacrilamide E10 were supplied by Aldrich Chemical Company and Allied Colloid Company, respectively. The solutions were mixed in a large tank using the mixer described in Chapter 4. The tank was filled to a capacity of 200 liters. The desired polymer was then added in in a series of small amounts by distributing it evenly across the mixture's free surface while the mixer was running. When the polymer had been distributed throughout the fluid, another small amount was added and mixed. This procedure was repeated until all the polymer had been evenly distributed throughout the liquid. This procedure consumed as much as 3 hours and was longest for the most viscous mixtures.

5.1 Shear Viscosity Measurements

5.1.1 Experimental Details

Viscosity measurements at low shear rates ($\dot{\gamma} < 13.4 \text{ s}^{-1}$) were performed using the Brookfield viscometer. To obtain viscosity data at higher shear rates ($\dot{\gamma} > 13.4 \text{ s}^{-1}$), the capillary tube viscometers were used. Viscosity measurements were first made with the Brookfield viscometer before the solutions were run through the capillary tube viscometers. By increasing the volumetric flow rate and recording the corresponding pressure drop, the functional relationship between shear stress and shear rate can be determined. The large and small capillary tubes were operated in the range of shear rates, $10 < 8V/D < 2000 \text{ s}^{-1}$ and $1800 < 8V/D < 20000 \text{ s}^{-1}$, respectively. To maintain laminar flow at shear rates in excess of 2000 s^{-1} , the smaller diameter tube was used after the larger diameter one. By using the smaller diameter tube, sufficiently high shear rates could be generated at which the liquid flow behavior approached a Newtonian limit of viscosity. To observe any degradation of the polymer solutions which may have been caused by the high shear rates, the solutions were measured again in the Brookfield viscometer after exiting from the capillary tubes. The viscosity of all solutions at the exit of the capillary tubes was generally 95 to 100% of the original values measured immediately after mixing a fresh batch. Although no precise control over temperature was exercised during the rheological measurements, the ambient room temperature was maintained at 22 degrees Celsius.

Silicon oil, with a viscosity of 48 cP, was used as a reference standard for calibration. Since the viscosities of silicon oils are not very sensitive to temperature, they are widely used as calibration liquids. After first calibrating the Brookfield viscometer with silicon oil, the instrument measured the viscosity of an aqueous solution of glycerine as being 21 cP. The aqueous solution of glycerine was then used to calibrate the capillary tube viscometers.

In taking data with the viscometers, there are two different types of errors. The first kind involves inadequate temperature control which cause errors of the order of 3-4%. Errors in temperature control were minimized by taking data at the same room temperature. The second type of errors are due to the inaccuracies of the instruments. The Brookfield viscometer was accurate within 1% of a full scale reading. The instrument made repeatable measurements within 0.2% of a full scale reading, over a range of shear rates, when measuring the viscosity of the glycerin water mixture. For the capillary tube measurements, the major errors at low pressure were due to the accuracy of the measurement of the differential pressure. At high pressure, the error in measuring the volumetric flow rate were most significant. For Newtonian liquids, this leads to an error in the mean of about 2%.

For non-Newtonian liquids there was a less extensive region where $\eta(\dot{\gamma}) = \eta_{\infty}$, the upper limiting viscosity, and the errors in η_{∞} were 3%. Absolute errors in $\eta(\dot{\gamma})$ are somewhat difficult to estimate. However, in the region where there was an overlap between the Brookfield data, the large and the small capillary data, the curves were essentially undistinguishable. Hence these two different methods provide a check on the absolute level of the measurements.

5.1.2 Results

Figures 5-1 and 5-2 show a plot of the shear stress as a function of shear rate for both the Xanthan gum polymer and the Polyacrilamide E10 solutions, respectively. The shear dependency is illustrated by the slope of the tangent line to the flow curve (i.e., the flow index n') in Figures 5-1 and 5-2. Plots of the shear stress vs. shear rate were generally curved and the slope was determined at each point. The slope was less than 1 for all values of shear rate and for all solutions, indicating shear thinning behavior.

The apparent viscosity data for the five different Xanthan gum concentrations is shown in Fig. 5-3. For each concentration, three distinct regions can be observed on the plot of apparent viscosity vs. shear rate. At the lower shear rates, $\dot{\gamma} < 10 \text{ s}^{-1}$, the apparent viscosity tends to deviate from values in the range ($10 < \dot{\gamma} < 3500 \text{ s}^{-1}$). This is mostly emphasized with the 0.05 and the 0.10% concentrations. Almost all theories predict that the $\eta(\dot{\gamma})$ should be an even function of the shear rate. Hence at low shear rate the shear viscosity can be represented by a parabola,

$$\eta(\dot{\gamma}) = \eta_0 (1 - \tau_n^2 \dot{\gamma}^2)$$

where higher order terms have been neglected. τ_n is a measure of the the strain rate at which non-Newtonian viscosity become important, Bruce and Schwartz [23]. In the mid-range of shear rates ($10 < \dot{\gamma} < 3500 \text{ s}^{-1}$), the apparent viscosity conforms to a power-law function. In the highest range of shear rates, $\dot{\gamma} > 3500 \text{ s}^{-1}$, the apparent viscosity becomes nearly constant and the solutions tend towards Newtonian behavior, with viscosities relatively close to that of water. Viscous behavior of this kind can be classified as "structural viscosity" since the change in flow behavior from region to region is a function of the structure of the macro-molecules and how they interact at different shear rates [1].

Figure 5-4 shows the apparent viscosity data for the Polyacrilamide E10 solutions. The same general tendencies are observed, although in this case it is seen that the parabolic region is more extensive. Consequently, the determination of the zero shear viscosity was

more accurate. It is also seen that η_{∞} is substantially higher for the Polyacrilamide E10 solutions than for the Xanthan gum solutions. Polyacrilamide solutions are relatively more stable under shear and they maintain their structural integrity more effectively than the Xanthan gum solutions.

The degree of shear thinning exhibited by each solution was characterized by the apparent viscosity power law index in the mid-range of shear rates. All concentrations were shear thinning with the power law index $n < 1$. For each concentration, the power law parameters and apparent viscosity limiting values are summarized in Tables 5-1 and 5-2 for the Xanthan gum and Polyacrilamide E10 solutions, respectively. η_0 is the viscosity value that is reached in the limit as the shear rate tends towards zero. For the Xanthan gum solutions, the data was not extensive in the parabolic region and thus, the uncertainties in estimating η_0 were generally high. A least squares approximation of the value η_0 was not possible because of insufficient data points in the low shear rate range. The values listed for η_0 were obtained graphically and only provide a rough estimate of the true value of η_0 . The value η_{∞} is the apparent viscosity value which is reached as the fluid becomes Newtonian at high shear rates. It is observed from Table 5-1 that n is a strong function of the solution concentration with a decreasing value of n for increasing values of concentration. The values of n represent a highly shear thinning behavior of the Xanthan gum aqueous solutions. The values of η_0 and η_{∞} increase with increasing concentration. However, η_{∞} does not increase as drastically as η_0 with increasing concentration. Newtonian behavior tends to begin for all solutions at a shear rate of approximately 4,000. For the Xanthan gum solutions, the Newtonian limit of viscosity is close to that of water.

Table 5-2 shows the power law parameters and the limiting viscosities for the Polyacrilamide E10 solutions. A least square analysis of the data in the parabolic region showed a consistently lower values for η_0 , when contrasted to the Xanthan gum values at the same concentration. For the same concentrations, η_{∞} was however higher for the Polyacrilamide solutions. The Polyacrilamide solutions also show a lower degree of shear thinning.

Table 5-1: Power-Law Parameters and Limiting Viscosities for Xanthan Gum Solutions

<u>Concentration (%)</u>	<u>k (cP-Sⁿ⁻¹)</u>	<u>n</u>	<u>η_0 (cP)</u>	<u>η_{∞} (cP)</u>
0.05	28.6	0.604	30	1.58
0.10	110.2	0.487	115	1.69

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0.15	321.0	0.383	822	1.84
0.20	664.4	0.333	2416	2.15
0.30	1804.8	0.248	14761	2.82

Table 5-2: Power-law Parameters and Limiting Viscosities for Polyacrilamide E10 Solutions

<u>Concentration (%)</u>	<u>k (cP-Sⁿ⁻¹)</u>	<u>n</u>	<u>η_0 (cP)</u>	<u>η_∞ (cP)</u>
0.0125	1.90	0.956	2.10	1.210
0.025	3.20	0.923	3.40	1.330
0.05	7.30	0.855	7.912	1.530
0.10	18.70	0.757	20.21	2.164
0.15	84.45	0.700	93.00	3.900
0.30	517.7	0.595	1243	6.300
0.50	1644.4	0.444	9342	12.10

5.2 Die-Swell Measurements

5.2.1 Experimental Details

The apparatus shown schematically in Fig.4-6 was used to measure the die-swell at high shear rates. A digital image analyzer with a close-up lens was positioned near the outlet of the capillary tube to measure the jet diameter. The measurement were performed by printing a hard copy of the computer screen; the diameter of the jet was subsequently measured on the printed image. The jet diameter and volumetric flow rate were measured at different flow conditions.

The major causes of errors in the first normal stress measurements are accurate determination of the die-swell ratio, which is limited by the Greenfield digital spray analyzer. The error is relatively small ($< 1\%$) due to the large jet magnification. The errors due to the measurement of the volumetric flow rate and thus the shear rate are of the order of 2%. In order to calculate the normal stress difference from die-swell measurements, a reasonably accurate estimate of the shear dependency as a function of $8V/D$ is needed. Over the range of $8V/D$ for which die-swell measurements were made ($300 \text{ sec}^{-1} < 8V/D < 6000 \text{ sec}^{-1}$), the shear dependency of the polymeric solutions is represented by the values of n' shown in Tables 1 and 2. The measurement of the die-swell ratio was limited to the region

where the flow index n' is nearly constant and is equal to the power law index n , and thus the errors in determining the power law index (of order of 3%) contributed the most to the die-swell test errors.

5.2.2 Results

The phenomenon of die-swell is attributed to the relaxation of the normal stresses (see Section 3-2). Figure 5-5 shows a typical image of the die-swell phenomenon. Die-swell data for the Xanthan gum and Polyacrilamide solutions are summarized in Tables 5-3 and 5-4 respectively. It should be mentioned that the Reynolds numbers, based on the apparent viscosity, was greater than 100 for all data of Tables 3 and 4. These data, together with the measured shear dependencies, were used to calculate the normal stresses from an equation proposed by Metzner and co-workers [19], (Eq.3-26). The applicability of the Eq.3-26 in calculating the normal stress difference has been questioned by Boger and Denn [21]. Of particular concern is the need for information on the rearrangement of the velocity profile at the exit of the tube. Keeping in mind the potential limitation of Eq.3-26, for relating normal stress to die-swell, the calculated values of the first normal stress function N_1 as a function of shear rate for the Xanthan gum and Polyacrilamide E10 solutions are given in Figs. 5-6.

The single most important difference between the Xanthan gum solutions and the Polyacrilamide E10 solutions is the much higher levels of normal stresses developed in the E10 solutions when compared to the Xanthan gum solutions. Despite the fact that all polymeric solutions exhibit a more conventional shear dependent viscosity (i.e., shear thinning), die-swell measurements indicate that Polyacrilamide develop large normal stresses at high shear rates. Although, the quantitative relation between die-swell and normal stresses is quite complex, the differences in normal stress behavior between the Xanthan gum solutions and the Polyacrilamide solutions are quite significant and no error of approximation can justify such a marked difference. The die-swell that has been observed for the Polyacrilamide solutions in the capillary tube experiments suggests that the extensional viscosity levels for the E10 solutions are large and that the extensional viscosity is probably the most significant rheological mechanism that inhibits breakup.

In spite of the close resemblance of the shear viscosity behavior of the Xanthan gum and the E10 solutions, it is seen that E10 solutions develop normal stresses that order of magnitude larger than those of the Xanthan gum solutions. These normal stresses are much higher than their associated shear stresses. Consequently, the development of the large

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normal stresses appears to be the most important rheological mechanism that inhibits atomization of viscoelastic liquids.

Table 5-3 Die-swell Measurements of Aqueous Solutions of Xanthan Gum

0.20%		0.30%	
$8V/D [s^{-1}]$	d_j/D	$8V/D [s^{-1}]$	d_j/D
981	0.892	359	0.937
1081	0.892	610	0.937
1372	0.892	1004	0.937
1921	0.892	1526	0.915
2029	0.892	1996	0.937
2395	0.937	2908	0.937
2719	0.915	3130	0.959
3093	0.915	3358	0.937
3318	0.915	3792	0.937
3692	0.915	3877	0.915
4206	0.937	4105	0.937
4690	0.915	4191	0.937
5288	0.915	4553	0.937
6053	0.915	5018	0.915
8248	0.915	5693	0.937

Table 5-4 Die-swell Measurements of Aqueous Solutions of Polyacrilamide E10

0.0125%		0.025%		0.05%		0.30%		0.50%	
$8V/D$	d_j/D	$8V/D$	d_j/D	$8V/D$	d_j/D	$8V/D$	d_j/D	$8V/D$	d_j/D
536	1.004	979	1.047	282	1.071	359	1.200	330	1.240
599	1.026	1436	1.047	384	1.071	610	1.200	526	1.250
723	1.004	1568	1.046	1497	1.071	1004	1.250	599	1.340
773	1.004	2680	1.052	2544	1.062	1526	1.320	773	1.350
1297	1.004	3022	1.047	2744	1.070	1996	1.360	1297	1.450
1696	1.004	3102	1.047	2844	1.071	2908	1.430	1696	1.470
2195	1.004	3459	1.047	2944	1.071	3358	1.450	2195	1.520
2844	0.995	3528	1.047	3017	1.069	3877	1.460	2844	1.550
3875	0.999	4424	1.047	3877	1.071	4191	1.430	3720	1.610
4590	1.004	4798	1.047	3991	1.071	4553	1.430	4423	1.640
6835	1.004	5060	1.047	7184	1.075	5693	1.460	6835	1.660

5.3 Extensional Viscosity Measurements

5.3.1 Experimental Details

Pressure measurements in the contraction flow chamber were performed using the piezoelectric transducers described in detail in Section 4.2. The measurements were conducted in a quasi-static mode. Transducers 1 and 2 were not operational during the measurements. Contamination of the transducer leads with the humidity complicated the measurements and rendered the measurements inaccurate. Transducer lead contamination resulted in an almost instantaneous discharge of the piezoelectric crystal even if the charge amplifiers time constant was set to long. In the present investigation, it was found that, to within experimental error, the remaining 2 pressure transducers (3 and 4) indicated the same pressure. This common pressure could be quite high and therefore there was every indication that the drop in pressure between that recorded on the transducers and atmospheric pressure occurred over a relatively small region close to the exit. In the interpretation of the data on the basis of the Binding theory, the transducer reading was therefore taken to be the entry pressure P_e .

5.3.2 Results

Figures 5-7, 5-8, and 5-9 show typical photographs of the flow field in the contraction flow chamber corresponding to the 0.15%, 0.3%, and 0.5% Polyacrilamide E10 solutions respectively. The liquid volumetric flow rate was maintained constant for at $Q_v = 1.06 \times 10^{-6} \text{ m}^3/\text{s}$. As can be seen in these photographs, the highest concentrations yield the most enhancement. The vortex in Fig. 5-9, corresponding to the 0.5% concentration of E10, extends to approximately 0.8 times the total length of the contraction flow chamber; its length is 7.66 in. Increasing the liquid flow rate beyond that shown in Fig. 5-9, renders the flow unstable. Entry pressures in excess of 1.5 times the original pressure before flow destabilization are recorded.

Figure 5-10 depicts the most common flow configurations seen when vortex enhancement is arrested by the upper wall of the chamber. This invariably resulted in flow destabilization. Perhaps the most provocative result of flow destabilization is the significant increase of entry pressure after the flow is destabilized. Figure 5-10 highlights several important features of the flow after destabilization. Nearly periodic oscillations between the five configurations shown are seen. The increase in pressure drop after flow destabilization corroborates the basic approach taken by Binding [4] in minimizing the energy consumption in the entry corner in his contraction flow analysis (see Section 3.4.2a). Vortex enhancement occurs in order to reduce the energy losses in the entry corner. When the vortex length is equal to the chamber length, no additional enhancement is possible and

the flow is destabilized. The large vortex configuration is destroyed leading to the flow configurations shown in Fig. 5-10. Schematic 1 of Fig. 5-10 shows the flow configuration when vortex length reaches the total length of the contraction flow chamber. At this time vortex enhancement (enlargement) is arrested. In Schematic 2, the vortex is then forced to retract due to the increased inertia of the incoming fluid into the chamber. In configuration 3, an imbalance develops; the vortex becomes asymmetric. Relatively long periodic oscillations between configuration 3 and 4 develop. The process can take as much as 2 minutes. In configuration 3, it is seen that enhancement occurs in the lower right portion of the chamber while retraction occurs in the upper left portion. In configuration 4 the opposite is true. The directions of the arrows in configurations 3 and 4 indicate either retraction or enhancement. Ultimately configuration 5 sets in. Throughout the process (configurations 1 to 5) there is a steady increase in the entry pressure. It should be mentioned however that there are relatively mild oscillations in pressure between configuration 3 and 4. It can be concluded that vortex enhancement occurs in order to reduce the viscous dissipation in the entry corner. When vortex enhancement is arrested by the upper wall of the chamber, the ultimate result is always destabilization of the flow and relatively large increases in the entry pressure result. The final result of interest is that for flows of viscoelastic materials through contractions sufficient chamber length should be provided to allow for vortex enhancement.

In Fig. 5-11, entry pressure vs flow rate data are given for the 0.15, 0.3, and 0.5% aqueous solutions of Polyacrilamide E10. It is clear that the data plotted logarithmically are well represented by a straight line over the available flow range. This was found to be the case for the all E10 solutions. From Fig. 5-11, the extensional power law parameters could be obtained, as described in Section 3.4.2a. The extensional viscosities thus determined are shown in Fig. 5-12. Only the 0.3% and 0.5% solutions of E10 are shown in Fig. 5-12. The accuracy of the measurements for the 0.15% solution was not ascertained, and thus the 0.15% solution data is not included; little or no vortex enhancement occurs for the 0.15% solution of E10 (see next paragraph). Table 5-5 shows the power law parameters in extension for the 0.3% and 0.5% solutions of E10. Also Shown in Table 5-5 are the power law parameters in extension obtained from the graphical data of reference [24] for 1% and 2% solutions of E10. As can be seen in Fig. 5-12 and Table 5-5, all solutions are extension thickening.

Table 5-2: Power-law Parameters in Shear and Extension Polyacrilamide E10 Solutions

<u>Concentration (%)</u>	<u>k (cP-Sⁿ⁻¹)</u>	<u>n</u>	<u>l (cP-Sⁿ⁻¹)</u>	<u>t</u>
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0.30	517	0.595	0.294	4.93
0.50	1644	0.444	5.8	4.50
1.0	3256	0.318	524.3	3.90
2.0	21770	0.226	53820	2.30

A useful indication of the applicability of the contraction flow analysis is provided by the comparison of the vortex attachment length predicted by the theory and the value determined from laser sheet flow visualization. Comparisons are provided in Figs. 5-13, 5-14, and 5-15 for the 0.15%, 0.3%, and 0.5% solutions respectively. The agreement is regarded particularly good for the 0.3% and the 0.5% solutions. It is seen however that there is marked discrepancy in the vortex length predicted by the theory for the 0.15% solution and the experimental data. We have attempted to interpret the experimental data for the 0.15% concentration on the basis of the Binding analysis, however it should be mentioned that the observed flow characteristic violated some of the assumptions of the analysis. In particular little or no vortex enhancement occurred for the 0.15% concentration. This was also true for Polyacrilamide concentrations below 0.15% and all Xanthan gum concentrations. Thus it can be concluded that the contraction flow technique is of little value for the rheological control of dilute polymer solutions. The success of the extensional viscosity measurements for the semi-dilute solutions (0.3% and 0.5% solutions) is weakened however since at these high concentration the liquid did not atomize; Chapter 9 discusses the atomization of these liquids. Thus the contraction flow technique is of little value for the rheological control of extensional viscosity particularly when applied to the atomization problem.

The Trouton ratio, which is the ratio of the extensional viscosity to the shear viscosity, is represented graphically in Fig. 5-16. As can be seen in Fig. 5-16, the extensional viscosity could be quite high when compared to the shear viscosity, reaching about 10^3 times the shear viscosity in the limit of the highest shear rates considered in the experiment. It is therefore of importance to consider the extensional viscosity levels of non-Newtonian liquids when describing breakup behavior of these liquids. Unfortunately, the contraction flow technique was shown to be inadequate for the rheological control of the extensional behavior of dilute polymeric materials. The success of extensional viscosity measurements for semi-dilute solutions is eclipsed by their unusual capability to resist breakup. It was not possible to assess the atomization quality of the more concentrated solutions. Chapter 9 will discuss these measurements thoroughly.

Fig. 5-1 Shear Stress vs Shear Rate for Xanthan Gum Solutions

Fig. 5-2 Shear Stress vs Shear Rate for Polyacrilamide E10 Solutions

Fig.5-3 Shear Viscosity Data Xanthan Gum Solutions

Fig.5-4 Shear Viscosity Data Polyacrilamide E10 Solutions

a)

b)

Fig.5-5 Die-Swell a) 0.3% Xanthan Gum, b) 0.5% Polyacrilamide E10 at $8V/D = 5246 \text{ S}^{-1}$.

Fig.5-6 First Normal Stress Difference Xanthan Gum and Polyacrilamide E10 Solutions

Fig. 5-7 Flow Field 0.15% aqueous Solution of Polyacrilamide E10, $Q_v = 1.0 \times 10^{-6} \text{ m}^3/\text{s}$

Fig. 5-8 Flow Field 0.30% aqueous Solution of Polyacrilamide E10, $Q_v = 1.06 \times 10^{-6} \text{ m}^3/\text{s}$

Fig. 5-9 Flow Field 0.50% aqueous Solution of Polyacrilamide E10, $Q_v = 1.06 \times 10^{-6} \text{ m}^3/\text{s}$

Figure 5-10 A Typical Sequence of Flow Configurations after Flow Destabilization

Figure 5-11 Entry Pressure vs Flow Rate Data for the 0.15, 0.3, and 0.5% aqueous Solutions of Polyacrilamide E10.

Figure 5-12 Extensional Viscosity Data for the 0.3, and 0.5% aqueous Solutions of Polyacrilamide E10.

Fig. 5-13 Vortex Attachment Length vs Flow Rate for the 0.15% aqueous Solutions of Polyacrilamide E10.

Fig. 5-14 Vortex Attachment Length vs Flow Rate for the 0.3% aqueous Solutions of Polyacrilamide E10.

Fig. 5-15 Vortex Attachment Length vs Flow Rate for the 0.5% aqueous Solutions of Polyacrilamide E10.

Fig. 5-16 Trouton Ratio for the 0.3, and 0.5% aqueous Solutions of Polyacrilamide E10.

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