CHAPTER 1

INTRODUCTION

1.1 MOTIVATION

The following thesis is an experimental study examining the confined swirling flow of viscoelastic fluids. The aim is to provide experimental observations of the flow kinematics of well characterised elastic fluids in a simple geometry involving a complex flow which consists of both primary and secondary flow fields. A torsionally driven cavity is used where the fluid is confined in an enclosed cylinder with a stationary top lid and a rotating bottom lid as depicted in figure 1.1. The rotation of the bottom lid produces a primary flow in the azimuthal direction which causes both an inertial (centrifugal) force directed radially outwards and, for viscoelastic fluids, a normal force directed radially inwards. For purely Newtonian fluids, the general secondary flow consists of a vortex which forces the fluid outwards along the rotating lid, up the stationary walls, inwards along the stationary lid and down the central axis. For highly elastic fluids, the general secondary flow is found to be in the opposite direction with the vortex directing the fluid inwards along the rotating lid and up the central axis. The flow fields for both a Newtonian fluid and a highly elastic fluid are shown as part of figure 1.1. A variety of complicated flow patterns are created as the levels of inertia and elasticity are varied.

Examples of non-Newtonian flow phenomena, and in particular the effects of fluid elasticity, are well documented in the literature and illustrated in a book by Boger & Walters (1993). Figure 1.2 illustrates just two examples of the dramatic differences between the flow behaviour of a Newtonian and a particular non-Newtonian fluid. Non-Newtonian fluids are also commercially very important and are found throughout a range of industries with several examples listed in table 1.1. Theoretical understanding of the flow of non-Newtonian fluids is essential to aid design of efficient fluid processes and to ensure adequate product quality.
**FIGURE 1.1 -** Torsionally driven cavity flow cell.
FIGURE 1.2 - Two examples of flow phenomena caused as a result of fluid elasticity for non-Newtonian fluids with comparison to the flow observed for Newtonian fluids. The top example is the 'rod-climbing' or Weissenberg effect, and the bottom example is extrudent die swell.
Visualisation studies are performed to understand the kinematics and to define or evaluate the governing parameters in the flow field of interest. Flow visualisation and quantitative measurements are also necessary for the validation of theoretical predictions using the equations of motion and continuity. In Newtonian fluid dynamics, the constitutive equation defining the deformation behaviour of a fluid is Newton’s law of viscosity. The combination of Newton’s law with the equations of motion gives the Navier-Stokes equations. However, even for Newtonian fluids, analytical solution of the Navier-Stokes equations and equation of continuity are only available for a limited number of flow problems and numerical methods are usually employed. Turbulent flows are very complicated and require enormous computational time to solve. Empirical models are then used to simplify the equations of motion such that the flow behaviour can be predicted in a realistic time for turbulent situations. In non-Newtonian fluid dynamics, only a limited knowledge about constitutive equations exists. There are many theories available which attempt to describe non-Newtonian fluids with many of these only applying to particular classes of fluids and/or for certain types of flows. Flow visualisation is therefore essential to ensure a particular constitutive equation is capable of predicting the essential features in a flow field rather than relying only on theoretical results. The validation of constitutive equations is also performed through rheological tests on the non-Newtonian fluids of interest and then comparing the measurements to those predicted theoretically. Both flow visualisation and rheological measurements provide the means to understand and describe the general flow kinematics of non-Newtonian fluids as well as to validate proposed constitutive equations.

The major characteristic of many non-Newtonian fluids is that the viscosity increases or decreases with shear rate. Another feature of many non-Newtonian fluids is the retention of a fading ‘memory’ of their flow history which is typically termed ‘elasticity’. The memory effect is characterised by a relaxation time, or a spectrum of relaxation times, depending on the constitutive model. High molecular weight polymer solutions and melts are commonly found to be both shear thinning and highly elastic. Therefore, it is difficult to discern between the effects of shear
thinning and elasticity for the flow phenomena observed using such fluids. Boger (1977/78) introduced the constant-viscosity elastic liquid to experimentally isolate elasticity from shear thinning. The so-called Boger fluids traditionally consist of dilute or semi-dilute concentrations of a high molecular weight polymer dissolved in a highly viscous solvent. The introduction of Boger fluids also made it possible to use the simple non-Newtonian constitutive models which predict or assume a constant viscosity. Previously, these models could only be used to qualitatively predict the effects of elasticity in flow because the predictions were compared to experiments which used shear thinning elastic fluids.

There are relatively few examples where three-dimensional and swirling flow fields of non-Newtonian fluids have been examined both experimentally and theoretically. Common benchmark problems in non-Newtonian fluid dynamics are essentially two-dimensional, such as in planar or tubular contractions and for flow around a sphere. However, these two geometries have areas where the fluid experiences high extension, which subsequently dominate the entire flow behaviour. The most simple constitutive models, and therefore the most desirable models for use in computation, predict an infinite extensional viscosity at high extension rates and subsequently cannot accurately predict the flow of polymer solutions in the contraction geometry and around a sphere. However, the flow behaviour in the torsionally driven cavity is dominated by either normal stress effects and/or inertia. Therefore, it is envisaged that the use of simple constitutive models, such as the Oldroyd-B model, in the equations of motion may be capable of quantitatively predicting the flow of Boger fluids in the torsionally driven cavity.

The torsionally driven cavity is used in this thesis as a proposed benchmark problem in which to theoretically predict the flow behaviour of viscoelastic fluids. The flow field is examined experimentally by using a variety of Boger fluids of different viscosity and elasticity. The secondary flow is examined for situations ranging from where inertial forces dominate to when elastic and viscous forces dominate. The Boger fluids consist of either dilute concentrations of a high molecular weight
flexible polyacrylamide polymer or semi-dilute concentrations of a high molecular weight semi-rigid xanthan gum polymer in aqueous solutions. Both of these polymers are abundant throughout a range of industries with several applications listed in tables 1.2 and 1.3 respectively. The rheological properties of the solutions used in this thesis are thoroughly characterised and compared to several constitutive models. Laser-induced florescence is used to qualitatively examine sectional streamline patterns, while particle image velocimetry offers a quantitative measurement of the radial and axial velocity field in the secondary flow plane. The combination of a well defined flow field with both qualitative and quantitative flow measurements in the torsionally driven cavity and using well characterised fluids, makes this thesis an excellent source of information for comparison to theoretical predictions.

1.2 THESIS OUTLINE

The terminology, with reference to the swirling flow apparatus, used throughout this thesis and the literature may vary slightly with the following terms used interchangeably: confined swirling flow; torsionally driven cavity; disk-and-cylinder system; and an enclosed cylinder with a rotating lid, disk or end-wall.

Chapter 2 reviews the scientific literature on the swirling flow of fluids of both a Newtonian and non-Newtonian nature from both an experimental and theoretical perspective. General discussion is initially pursued on the phenomena of vortex breakdown which is observed in swirling flows of Newtonian fluids. The behaviour of both Newtonian and non-Newtonian fluids in the torsionally driven cavity is then examined. Lastly, the chapter focuses on several rotating flows of viscoelastic fluids and the observation of flow instabilities in such flows.

Chapter 3 describes the experimental methods and materials used in this work. The chapter is split into two parts. The first part describes the methods used to characterise the non-Newtonian fluids using rheological constitutive models. The
second part of the chapter details the torsionally driven cavity and flow visualisation techniques.

Chapter 4 presents the rheology and constitutive model parameters of the test fluids. This chapter is also split into two parts where the first part examines the properties of the low-viscosity fluids used in chapter 5, while the second part examines the properties of the high-viscosity solutions which are used in chapter 6.

Chapter 5 investigates the confined swirling flow of low-viscosity Boger fluids for situations where inertia dominates the flow field. The effects of slight fluid elasticity on vortex breakdown is examined.

Chapter 6 investigates the confined swirling flow of high-viscosity Boger fluids. The inertial forces are systematically decreased by using fluids of increasing viscosity. When the viscosity is high such that the Reynolds number is small, the inertia is negligible and the flow kinematics are dominated only by elasticity and viscosity.

Chapter 7 consists of conclusions and recommendations for future work.

<table>
<thead>
<tr>
<th>Industry</th>
<th>Non-Newtonian fluids</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mineral</td>
<td>slurries/suspensions, polymer addition</td>
</tr>
<tr>
<td>Paint</td>
<td>suspensions and polymeric liquids</td>
</tr>
<tr>
<td>Plastics</td>
<td>molten polymer</td>
</tr>
<tr>
<td>Food</td>
<td>suspensions and polymeric liquids</td>
</tr>
<tr>
<td>Chemical</td>
<td>emulsions</td>
</tr>
<tr>
<td>Agriculture</td>
<td>polymer addition</td>
</tr>
<tr>
<td>Printing</td>
<td>polymer addition</td>
</tr>
<tr>
<td>Oil</td>
<td>waxy crude oil, polymer liquids</td>
</tr>
</tbody>
</table>
### TABLE 1.2 - Commercial uses of polyacrylamide.

**Compounding and Formulating:**
- Pulp and paper - filler retention aid, dry-strength improvement. Tape-joint cement.

**Processing aids:**
- Sugar clarification
- Borax manufacture
- Inorganic fibre slurries: asbestos-cement products, insulation board
- Phosphoric acid purification
- Electrorefining processes
- Magnesia production
- Coal washeries
- Aluminium sulphate manufacture
- Cement manufacture
- Lime sulphur filtration

**Industrial reclamation:**
- Flue-dust recovery
- Petroleum refinery oil reclamation

**Water treatment:**
- Primary flocculents
- Raw water treatment
- Filler retention and drain improvement
- White-liquor clarification
- Hot and cold lime softening
- Potable water treatment
- Flotation-type save-alls
- Sludge treatment

**Pulp and paper manufacture:**
- Green-liquor clarification
- White-water clarification

**Mining and ore processing:**
- Underground mine water
- Thickening flotation concentrates
- Leaching - copper, gold, uranium
- Zinc calcines
- Hydraulic backfill
- Settling of slimes
- Tailings disposal

**Oil industry:**
- Secondary oil recovery
- Drilling-water clarification

**Miscellaneous applications:**
- Friction reduction
- Film forming
- Used in the production of: ammonia, barium brines clay, drugs, fine chemicals, inorganic salts, lithium, soda ash, and pigments.
- Viscosity agent
- Adhesives
**Table 1.3 - Commercial uses of xanthan gum (Zirnsak 1995).**

**Food Industry:**

<table>
<thead>
<tr>
<th>Category</th>
<th>Product</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pie fillings</td>
<td>Salad dressings</td>
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<tr>
<td>Cheeses</td>
<td>Milk and cream products</td>
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<tr>
<td>Ice confectionery</td>
<td>Table syrups</td>
</tr>
<tr>
<td>Sauces</td>
<td>Gravies</td>
</tr>
<tr>
<td>Beverages</td>
<td>Batters</td>
</tr>
<tr>
<td>Dough</td>
<td>Cake mixes</td>
</tr>
<tr>
<td>Canned products</td>
<td>Powdered drink mixes</td>
</tr>
<tr>
<td>Toppings</td>
<td>Retort pouches</td>
</tr>
<tr>
<td>Puddings</td>
<td>Dressings</td>
</tr>
<tr>
<td>Powdered soup mixes</td>
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**Oil Industry:**

<table>
<thead>
<tr>
<th>Category</th>
<th>Product</th>
</tr>
</thead>
<tbody>
<tr>
<td>Drill hole cleaning</td>
<td>Drilling fluids</td>
</tr>
<tr>
<td>Workover fluids and completion fluids in oil wells</td>
<td>Hydraulic fracturing fluids</td>
</tr>
<tr>
<td>Enhanced oil recovery fluids</td>
<td></td>
</tr>
</tbody>
</table>

**Agricultural Industry:**

<table>
<thead>
<tr>
<th>Category</th>
<th>Product</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flowable pesticides</td>
<td>Flowable fungicides</td>
</tr>
<tr>
<td>Flowable herbicides</td>
<td>Flowable insecticides</td>
</tr>
<tr>
<td>Liquid feed supplements of farm animals</td>
<td></td>
</tr>
</tbody>
</table>

**Miscellaneous applications/uses:**

<table>
<thead>
<tr>
<th>Category</th>
<th>Product</th>
</tr>
</thead>
<tbody>
<tr>
<td>Removal of rust, welding rods, weld slag, and other debris from gas pipelines.</td>
<td>Paint</td>
</tr>
<tr>
<td>Textile printing pastes</td>
<td>Acid and alkaline cleaners</td>
</tr>
<tr>
<td>Ceramic glazes</td>
<td>Slurry explosives</td>
</tr>
<tr>
<td>Abrasive cleaners</td>
<td></td>
</tr>
<tr>
<td>Ceramic processes based on extrusion</td>
<td>Paper</td>
</tr>
<tr>
<td>Ink</td>
<td>Pigment suspensions</td>
</tr>
<tr>
<td>Space dyeing solutions</td>
<td>Industrial emulsions</td>
</tr>
<tr>
<td>Foundry coatings</td>
<td>Toothpaste</td>
</tr>
<tr>
<td>Polishes</td>
<td>Shampoos</td>
</tr>
<tr>
<td>Wallpaper adhesives</td>
<td>Coal in water slurries</td>
</tr>
<tr>
<td>Beauty creams</td>
<td>Make-up</td>
</tr>
<tr>
<td>Lotions</td>
<td></td>
</tr>
</tbody>
</table>
CHAPTER 2

LITERATURE REVIEW OF SWIRLING FLOW

2.1 INTRODUCTION

The following chapter is a literature review on the swirling flow of Newtonian and non-Newtonian fluids. Swirling flows are abundant throughout industrial processes and the properties of swirl are well utilised. The swirl of Newtonian fluids is essentially well understood and can be modelled using computational fluid dynamics with the Navier-Stokes equations. However, elastic fluids are not uniquely described in all flows using an equivalent set of Navier-Stokes type equations and therefore the behaviour of elastic fluids due to swirl is difficult to predict and therefore difficult to understand. Yet swirl is present throughout the processing of non-Newtonian fluids and knowledge of the behaviour of such fluids due to swirl is desirable.

The central swirling flow of interest in this thesis is that produced in a torsionally driven cavity. The torsionally driven cavity is one of the simplest geometries which involves swirl and is ideal for attempting to predict experimental observations as a test of numerical codes and of non-Newtonian constitutive models. The prediction of the flow behaviour in such a geometry may act as a precursor to solving more difficult problems associated with swirl, for example in complex mixing processes.

The first part of this chapter examines the confined swirling flow of Newtonian fluids from both an experimental and theoretical perspective. An introduction to the phenomena of vortex breakdown, which is commonly observed in numerous geometries involving strong swirling flows, is included. The second part of the chapter analyses the confined swirling flow of non-Newtonian fluids also from an experimental and theoretical perspective. The last part of the chapter discusses other rotating flows of viscoelastic fluids with particular reference to elastic flow instabilities.
2.2 VORTEX BREAKDOWN AND THE CONFINED SWIRLING FLOW OF NEWTONIAN FLUIDS

In the confined cylindrical swirling flow of Newtonian liquids, in which the fluid is situated in an enclosed cylinder with rotating bottom lid, the rotation of the lid produces a non-uniform centrifugal force along the base and a secondary flow generated in the cylinder normal to the primary flow. An Ekman layer is present on the rotating lid with a thickness of the order $(\nu \Omega R^3)^{0.5}$ where $\nu$ is the kinematic viscosity, $\Omega$ is the rotation rate and $R$ is the lid radius (Lopez, 1990). The Ekman layer acts as a centrifugal pump by driving the fluid outwards along the rotating base, up the side walls and down the central axis in a spiral motion where it is then sucked back into the boundary layer as depicted in figure 2.1(a). As the rotation rate of the disk is increased, a widening of the vortex core near the disk is observed, followed by the production of a stagnation point on the central axis and a weak recirculation zone which is characteristic of an axisymmetric vortex breakdown bubble and shown pictorially in figure 2.1(c).

A majority of the research on the confined swirling flow of Newtonian fluids focuses on the phenomena of vortex breakdown because of its application to a broad range of flow geometries including those found in aeronautics. Therefore, before further discussion on the confined swirling flow of Newtonian fluids is undertaken, the general phenomena of vortex breakdown is briefly reviewed.

2.2.1 Introduction to Vortex Breakdown

Vortex breakdown refers to the situation where a sudden transition of a vortex flow structure occurs with an abrupt change in character. The breakdown is usually associated with the development of a flow stagnation point and often with regions of reversed axial flow. Vortex breakdown was reportedly first observed experimentally as the ‘bursting’ of trailing edge vortices from aircraft travelling at high angles of attack by Peckham & Atkinson (1957), Elle (1960), Werlé (1960) and Lambourne & Bryer (1961). An example of two forms of breakdown occurring upon a delta wing is
FIGURE 2.1 - Secondary flow patterns for a Newtonian fluid at conditions showing (a) inertia driven vortex, (b) pre-incipient breakdown, and (c) vortex breakdown.
shown in figure 2.2. A great deal of aeronautical research investigating vortex breakdown arose primarily due to its detrimental consequences on limiting the angle of attack for jet fighter aircraft. The extensive literature has shown that the lift, drag and pitching moment of a delta wing all undergo an abrupt deterioration as the location of vortex breakdown, with increasing angle of attack, moves upstream over the trailing edge of the wing. Vortex breakdown can also cause buffeting, unsteadiness and poor control of aircraft.

Experimental investigations on the breakdown of vortices over delta wings is extremely difficult due to the complicated nature of the leading edge vortex, unsteadiness and a lack of axial symmetry. A simple vortex tube was introduced by Harvey (1962) who observed vortex breakdown as air travelled axially along a circular tube where the degree of swirl imparted on the air was controlled by a set of adjustable vanes. The introduction of a diverging tube by Sarpakaya (1971) allowed the characterisation of several types of breakdown forms and in particular he observed the transformation between the two main types of breakdown - asymmetric "spiral-type" and axisymmetric "bubble-type" - by increasing the degree of swirl. The two forms of breakdown are shown for the case over a delta wing and in swirling pipe flow in figures 2.2 and 2.3 respectively. An enclosed cylinder with rotating end-wall was used by Escudier (1984) to investigate vortex breakdown in an even more refined experiment due to the axis symmetric geometry and well defined boundary conditions producing a well posed problem which is ideal for numerical solution of the Navier-Stokes equations (Lopez 1990).

Vortex breakdown has been researched heavily over the last 40 years with the first theories proposed by Squire (1960), Jones (1960), Ludweig (1961), and Benjamin (1962). Detailed discussion on the mechanisms and theories governing breakdown may be found in review articles by Hall (1972), Leibovich (1978, 1984), Escudier (1988), and Delery (1994). More recent discussions on the phenomena have been made by Berger and Erlebacher (1995), Keller (1995), Rusak (1996), and Wang & Rusak (1997).
Figure 2.2 - Vortex breakdown over a delta wing (reprinted from Lambourne & Bryer 1961).
FIGURE 2.3 - Vortex breakdown in a diverging tube showing (a) spiral type, and (b) nearly axisymmetric ‘bubble’ type breakdown (reprinted from Sarpakaya 1971).
In general, breakdown flow fields may be broken up into three spatial regimes: the approach flow, the breakdown region and the downstream flow (Leibovich 1978). The approach flow consists of a concentrated vortex core with the flow being laminar or with relatively low turbulent intensities and axial flow variations being gradual. The axial velocity is often observed to be jet-like, with speeds on the axis exceeding those outside the core by an amount comparable to the maximum azimuthal speed. The breakdown region is often characterised by rapid changes in axial flow direction and comprises of three subintervals as follows:

- The approach flow is decelerated and a stagnation point is formed on the vortex axis,
- flow reversal then occurs near the vortex axis for both the spiral-type breakdown and the bubble-type breakdown,
- the original direction of axial flow is restored in the third subinterval, which is marked by a large increase in turbulent intensity or, for laminar-approach flows, by the first unmistakable signs of transition to turbulence.

Downstream of the breakdown zone, a new vortex structure is established with an expanded core. Axial velocity profiles closely resemble a conventional wake behind a solid obstacle, with centreline speeds less than those outside the vortex core, and the flow is invariably turbulent. The various vortex breakdown forms are generally observed to oscillate in position during tube swirling flow experiments, with the entire breakdown form also observed to be unsteady in position and move back and forth in an unpredictable way along the tube, as described by Leibovich (1978).

Hall (1972) considered that the primary conditions necessary for vortex breakdown to be a high a degree of swirl, a positive or adverse pressure gradient (where the pressure increases in the direction of flow) and a divergence of the stream tubes in the vortex core immediately upstream of the breakdown, and that it is a balance of these factors which determine whether vortex breakdown occurs. Two definitions which have generally been applied to examine the ratio of swirl and axial velocity are
the angle of swirl ($\phi_s$) and the swirl parameter ($S$), which are given by Hall (1972) and Delerey (1996) respectively as follows:

$$\phi_s = \tan^{-1}\left(\frac{v_\phi}{v_z}\right)$$  \hspace{1cm} (2.1)

$$S = \frac{\Gamma_0}{r V_z}$$  \hspace{1cm} (2.2)

$v_\phi$ and $v_z$ are the swirl (or azimuthal) and axial velocity components respectively, $\Gamma_0$ is the circulation or swirl, $r_c$ is the radius of the vortex core and $V_z$ is the axial velocity just outside the vortex core. The maximum value of $\phi_s$ upstream of breakdown is invariably greater than 40 - 50° while $S$ is found to be between 1.28 and 1.41 (Hall 1972, Delerey 1996). Therefore, this criteria essentially infers that a steady vortical flow, where the axial velocity component is positive, can only exist if its swirl velocity is not too large compared to the axial velocity. If the steady vortical flow, which is referred to as being supercritical, enters an adverse pressure gradient, its axial motion will be decelerated and an increase in $\phi_s$ and $S$ will follow and a change in the vortical flow field will take place with a jump to a subcritical state.

There are several theories or explanations of breakdown which have been well discussed in the literature and in particular by the reviews previously mentioned. Four of the theoretical methods which have been used to try and understand breakdown are: the quasi-cylindrical approximation and analogy to boundary layer separation, hydrodynamic instability, the solution of the axisymmetric Navier-Stokes equations, and the concept of critical state. All the above theories have limitations but are very useful in understanding breakdown.

It is variations on the critical state or ‘wave’ theory, first proposed by Squire (1960), Benjamin (1962, 1967) and Bossel (1967, 1969), which gives the most consistent explanation of the breakdown phenomena. The theory assumes that if a long standing wave can exist on a vortex, small disturbances coming from downstream will propagate in the upstream direction and ultimately cause breakdown. A
supercritical flow is one in which waves propagating against the flow are carried downstream and hence cannot support a standing wave. A subcritical flow is then one in which the waves may propagate upstream and a standing wave may exist in the flow. Hence, in a supercritical flow, increasing the swirl or decreasing the axial component will drive the flow towards a subcritical flow state. An extension of the critical state theory has been tested both in swirling pipe flow (Darmofal & Murman 1994) and in a torsionally driven cavity (Brydon & Thompson 1998), where the trapping of small perturbations is considered responsible for the growth of vortex breakdown bubbles (Leibovich 1970; Bilanin & Widnall 1973). Wang & Rusak (1997) use stability analysis together with steady-state solutions of the Navier-Stokes equations to provide a thorough explanation of the mechanism leading to axisymmetric vortex breakdown in high Reynolds number swirling flows in a pipe. They found that the mechanism is governed by the propagation of small- and large-amplitude disturbances in the pipe and its interaction with the inlet conditions. As a result of the loss of stability of an initial columnar swirling flow, when the swirl ratio is at or above a critical level, the base columnar state evolves into another relatively stable equilibrium state which represents a flow around a stagnation zone.

Although there is still no unique consensus on the underlying mechanism for the occurrence of vortex breakdown, the combination of advanced numerical techniques and increased computer power to solve the unsteady Navier-Stokes equations, such that the theories can be fully tested, are beginning to elucidate the mechanism. A complete understanding of the mechanisms for vortex breakdown is very realisable in the near future.
2.2.2 Confined Swirling Flow of Newtonian Fluids - Experimental

Vortex breakdown in an enclosed cylinder with a rotating lid was first observed experimentally using flow visualisation techniques by Vogel (1968, 1975), Hill (1972), and Ronnenberg (1977) for a limited range of parameter space and with only one breakdown bubble observed. Escudier (1984) observed the formation of up to three breakdown bubbles and produced a detailed diagram, represented in figure 2.4, showing the existence domain of vortex breakdown with respect to two governing dimensionless groups, the aspect ratio ($H/R$) and a rotational Reynolds number ($Re$). The Reynolds number may be defined as follows:

$$Re = \frac{\rho (2\pi \Omega) R^3}{\eta}$$  \hspace{1cm} (2.3)

$\rho$ is the density (kg/m$^3$), $\Omega$ is the disk rotation rate (s$^{-1}$), $R$ is the disk radius (m), $H$ is the cylinder height (m) and $\eta$ is the viscosity (Pa s). Escudier (1984) observed that the breakdown regions were highly axisymmetric, which supported his view that vortex breakdown is inherently axisymmetric and departures from axisymmetry are the result of instabilities not directly associated with the breakdown process. The recirculation zone inside the breakdown region was observed to contain low interior velocities. A steady oscillatory flow, where the stagnation point bubbles moved up and down, was also observed at high Reynolds numbers ($Re > 2600$ depending on $H/R$) with vortex breakdown still highly axisymmetric. The flow was ultimately observed to became unsteady and then turbulent with a further increase in the Reynolds number. Fujimura et al. (1997) examined the location of the stagnation points during spin-up and spin-down of the rotating lid. Fujimura et al. (1997) found that equilibrium after spin-up from rest was reached after more than 25 seconds ($1970 < Re < 2450$, $H/R = 2.5$).

There are only a few reports which have measured velocity distributions in the disk and cylinder system for Newtonian fluids and these have mainly investigated the
Figure 2.4 - Existence domain of vortex breakdown for Newtonian fluids (extracted from Escudier 1984). Circular symbols represent experimental points produced for a Newtonian fluid in the current thesis.
techniques in determining the velocity rather than examining the measurements themselves. In the absence of breakdown, tangential velocity measurements have been made by Bien & Penner (1970) and radial and tangential velocity measurements were made by Hill (1972). Prasad & Adrian (1993) have also demonstrated the use of stereoscopic particle image velocimetry to obtain accurate measurement of the tangential, radial and axial velocity profiles for a Newtonian fluid at low Reynolds number. Only a limited set of velocity measurements were made in the presence of breakdown by Ronnenberg (1977) and Buchave et al. (1991).

2.2.3 Confined Swirling Flow of Newtonian Fluids - Theoretical

The cylinder with rotating lid provides the simplest geometry in which vortex breakdown is observed. This flow field is therefore ideal for numerical studies into the vortex breakdown phenomena using the time dependent Navier-Stokes equations. Investigation into vortex breakdown at steady state conditions using the numerical solution of the axisymmetric Navier-Stokes equations has been primarily performed by Lutg & Abboud (1987), Neitzel (1988), Sørensen & Daube (1989), Lopez (1990), Brown & Lopez (1990), Tsiitverbilt (1993), Sørensen & Christensen (1995), Gelfgat et al. (1996), and Brydon & Thompson (1998). The numerical works were validated by comparing the predicted streamlines with the streaklines observed from the flow visualisation images of Escudier (1984) with excellent agreement, as shown in figure 2.5 for the numerical model of Lopez (1990). Brydon & Thompson (1998) provide the complete existence domain for breakdown which compares very closely to the experimental results of Escudier (1984).

The numerical investigations indicate that an Ekman boundary layer with thickness of the order \((\nu \Omega R)^{0.5}\) acts as a centrifugal pump on the rotating lid (Greenspan 1968; Schlichting 1979; Lopez 1990). The boundary layer on a rotating disk is the layer of fluid which is carried by the disk owing to friction and is thrown outwards due to centrifugal forces. The boundary layer may simply be estimated by balancing the centrifugal force on a fluid element adjacent to the disk with the radial component of the disk shearing stress. The fluid then travels up the stationary side walls and
Figure 2.5 - Comparison between experimentally observed dye-lines (Escudier 1984) and numerically predicted sectional streamline patterns of vortex breakdown for a Newtonian fluid at $Re = 2494$ and $H/R = 2.5$ (reprinted from Lopez 1990).
inwards along the stationary lid to form a secondary flow which is cellular in appearance with the sectional streamlines depicted diagrammatically in figure 2.1(a) for a Reynolds number of \( Re \approx 1000 \) with \( H/R \approx 2.5 \). Angular momentum \( (I = rv) \) and total head \( (H = P/\rho + \frac{1}{2}(v_x^2 + v_y^2 + v_z^2)) \) are acquired in the Ekman boundary layer but are lost in the viscous boundary layers on the stationary side walls and stationary lid. The angular momentum and total head were found by Lopez (1990) to be relatively constant along stream surfaces in the meridional plane.

Lopez (1990) describes the breakdown process as the result of the advection of angular momentum towards the central axis as the fluid flows radially inwards along the stationary lid from the corner of the side wall at Reynolds numbers a little below those required for breakdown \( (Re \approx 1600-1800 \) for \( H/R = 2.5 \)). Preservation of the angular momentum causes the angular velocity to increase, and consequently an increase in centrifugal acceleration to a local maxima results as the fluid flows axially down the centre of the cylinder towards the rotating lid. The stream surfaces then deform and take on a concave shape resulting in a stationary centrifugal (or inertial) wave, as pictorially shown in the streamlines of figure 2.1(b). The amplitude of the inertial waves increases and their wavelength decreases with further increases towards the Reynolds number required for breakdown. The associated axial deceleration is then large enough to cause the flow to stagnate under the crest of the wave and cause an adverse pressure gradient, resulting in vortex breakdown (figure 2.1c).

The numerical analysis of Lucht & Abboud (1987) show that Hall’s (1972) criteria for the occurrence of breakdown using the angle of swirl, defined by (2.1), is met with a value of the swirl angle just below the stationary lid increasing to \( \phi_s \approx 40^\circ \) once breakdown takes place for \( H/R = 2 \). Lucht & Abboud (1987) also note that as the Reynolds number is increased further, the axial velocity component increases slightly relative to the azimuthal component which causes the breakdown bubbles to disappear.
Lopez (1990) and Brown & Lopez (1990) introduce the azimuthal component of vorticity \( \omega_\theta = \frac{dv_r}{dz} - \frac{dv_z}{dr} \) as a useful parameter for examining the criteria for the occurrence of breakdown. Lopez (1990) observed a region around the central axis where a change in sign of \( \omega_\theta \) (to positive) corresponded to the occurrence of a ‘waviness’ in the sectional streamlines. This region of positive \( \omega_\theta \) is initially small and situated just above the Ekman layer at low Reynolds number \( (Re < 1000, H/R = 2.5) \). As the Reynolds number is increased \( (Re \approx 1600-1800, H/R = 2.5) \), the region of positive \( \omega_\theta \) increases and includes the area where the stream surfaces become wavy and where vortex breakdown is observed with further increases in Reynolds number. Lopez & Perry (1992), Lopez (1990) and Brown & Lopez (1990) conclude that an azimuthal component of vorticity induces the reverse flow on the axis and that a positive \( \eta_\theta \) is a necessary condition for the occurrence of vortex breakdown. The azimuthal vorticity is produced by the tilting and stretching of the vorticity vector which consists mainly of an axial component (due to the swirling motion), and an azimuthal component (due to the meridional motion). They also applied their theory to the swirling pipe flow and established an alternative criteria for the occurrence of breakdown based on the relationship between the angle of the velocity vector and the vorticity vector on stream surfaces upstream of breakdown as follows:

\[
\phi_\phi > \phi_\omega \tag{2.4}
\]

\( \phi_\omega = \tan^{-1}(\omega_\theta / \omega_\phi) \) where \( \omega_\phi \) and \( \omega_\theta \) are the azimuthal and radial components of vorticity respectively. Brown & Lopez (1990) show that vortex breakdown occurs when the ratio of the helix angle of the velocity vector to that of the vorticity vector is greater than one.

Gelfgat et al. (1996), similarly to Lopez (1990), conclude that a necessary condition for vortex breakdown is a concave form of the stream surfaces, which may be considered as the cause of the change in sign of the azimuthal component of vorticity. However, Gelfgat et al. (1996) shows that vortex breakdown does not necessarily occur when the \( \omega_\phi \) is negative or when the stream surfaces are concave in shape by
observing these conditions at low cylinder aspect ratios where vortex breakdown is not observed to occur at any Reynolds number.

The oscillatory instability and unsteady flow behaviour which Escudier (1984) observed at high Reynolds number, as shown in figure 2.4, has been primarily investigated by Sørensen & Daube (1989), Lopez (1990), Lopez & Perry (1992), Liao & Young (1995), Sørensen & Christensen (1995), and Gelfgat et al. (1996). With the advancement of numerical codes and computer technology, the ability to solve unsteady flow problems is becoming possible, such that greater insights may be gained into the changing kinematics of various flow structures observed at high Reynolds number. The unsteady flow field for Newtonian fluids is not of primary interest in this thesis and therefore will not be discussed further.

2.2.4 Related Rotating Flows of Newtonian Fluids

Related investigations to the confined swirling flow which involve the cylinder geometry include the co-rotation or counter-rotation of two lids, and the use of an open cylinder with a rotating bottom lid and a free surface. Also related is the infinite or finite disk flow, where the disk may be situated in an infinite or finite sea of fluid. Other swirling flows are those found between rotating coaxial plates and a cone-and-plate.

Roesner (1990) investigated experimentally vortex breakdown in a cylinder with two rotating lids and found that, when at incipient breakdown, co-rotation of the lids resulted in breakdown while counter rotation resulted in the disappearance of the breakdown bubble. Numerical investigations into the two rotating lid system have been conducted by Valentine & Jahnke (1994), Lopez (1995), Gelfgat et al. (1996) and Watson & Neitzel (1996). Watson & Neitzel (1996) found that the criteria of Brown & Lopez (1990) was met at the location of the breakdown bubble in their flow domain. However, the criteria was not met upstream of breakdown, nor at the incipient state of breakdown, which questions the use of the criteria of Brown and Lopez (1990) as a predictive tool.
Spohn et al. (1993) examined experimentally the secondary flow in an open cylinder with one rotating lid, and found that the conditions for vortex breakdown changed noticeably from those observed for a closed cylinder. The differences to the closed cylinder case included: breakdown occurred at lower Reynolds numbers; a breakdown bubble was present even at the maximum Reynolds number tested \((Re = 3500)\); breakdown was observed at aspect ratios as low as \(H/R = 0.5\); and breakdown bubbles were generally much larger in size. A breakdown bubble attached to the free surface was also observed, which cannot be explained by classical vortex breakdown theories (e.g. Benjamin 1962, Ludweig 1961) which assume a cylindrical vortex core upstream of breakdown.

Rotating disk flows include those generated by an infinite or finite disk in semi-infinite fluid and those where the fluid is bounded by two infinite or finite disks. Rotating disk flows have been widely investigated in Newtonian fluid mechanics and were first investigated analytically by von Karman (1921) for an infinite rotating disk in a semi-infinite fluid. The problem was made solvable by von Karman (1921) due to the discovery of a similarity transformation which produced a set of ordinary differential equations. It should be noted that in the torsionally driven cavity flow, the bounding side walls eliminates the similarity condition and so the method cannot be used in the cavity flow. Many variants of the von Karman (1921) problem have been published with difficulties arising due to the multiplicity of solutions, as highlighted in communications of Batchelor (1951) and Stewartson (1953) and the more recent articles on coaxial disk flows by Szeri et al. (1983ab). The subject is vast and the review of Zandbergen & Dijkstra (1987) is highly recommended for discussion on the characteristics of a variety of solutions for the one-disk and two-disk problem, and on the stability of stationary solutions. Other reviews for disk flows of Newtonian fluids include those by McLeod (1975), Parter (1982), van Wijngaarden (1985), and Brady & Durlofsky (1986).
2.2.5 Summary

In the confined swirling flow of Newtonian fluids, centrifugal forces drive a secondary flow outwards along a rotating lid, along perpendicular stationary walls, along the stationary lid and down the central axis. The flow is dominated by inertia at high Reynolds number and axisymmetric vortex breakdown results due to the divergence of the streamlines near the rotating lid, leading to an adverse pressure gradient down the central axis such that once the swirl level reaches some critical limit, the vortex ‘bursts’. In the subcritical state prior to breakdown, the axial sectional streamlines are stationary inertial waves which propagate upstream when the critical swirl level is reached, causing a supercritical state and vortex breakdown.

2.3 THE CONFINED SWIRLING FLOW OF NON-NEWTONIAN FLUIDS

The swirling flow of non-Newtonian fluids is much more complex than that for Newtonian fluids. Several more parameters are required to describe non-Newtonian fluids including those associated with a shear thinning viscosity and fluid elasticity. In a rotating flow, elasticity is demonstrated through the primary normal stress difference, which is zero for a Newtonian fluid. The effect of the primary normal stress difference is demonstrated by analysing a fluid element in the swirling flow which takes on the appearance of that shown in figure 2.6. The normal stresses acting in the direction of the primary flow cause a tension along the streamlines, with the resulting ‘elastic’ force acting radially inwards in the opposite direction to the centrifugal force. The effects of inertia and elasticity may be regarded as opposing each other in a swirling flow.

The introduction of non-Newtonian fluids into the confined swirling flow experiment creates two more independent sets of parameters for the problem: elasticity and shear thinning. Shear thinning may be represented by either a single or group of shear thinning parameters, which may be found in numerous viscosity models (eg. Carreau
**FIGURE 2.6** - Fluid element in a swirling flow. Indicated are the opposing centrifugal and 'elastic' forces where the elastic component arises from a tension along the streamline due to the normal stress $\sigma_{\theta\theta}$. 
models and ‘power’ law models, see Bird et al. 1987a). If the fluid is shear thinning, the Reynolds number is based on the zero-shear rate viscosity and given the symbol \( Re_p \). Elasticity is typically represented by a Weissenberg number (\( We \)), which is measured by evaluating of the ratio of the characteristic time of the fluid (eg. Maxwell relaxation time: \( \lambda_m \)) and characteristic time of the process (eg. \( 1/2\pi\Omega \)) as follows:

\[
We = \lambda_m 2\pi\Omega
\] (2.5)

Another dimensionless number which will be used is the elasticity number (\( El \)), which measures the ratio of the elastic forces to inertial forces, and is represented as follows:

\[
El = \frac{We}{Re} = \frac{\lambda_m \rho_0}{\rho R^2}
\] (2.6)

A feature of the Elasticity number is that it is independent of rotation rate of the lid, provided the relaxation time is constant and not shear rate dependent. The flow of non-Newtonian fluids in a torsionally driven cavity will now be discussed in detail.

### 2.3.1 Experimental Observations

The confined swirling flow of non-Newtonian fluids was realised by Hill, Huppler & Bird (1966) as a possible flow geometry to act as a test case for the ability of constitutive equations to describe non-Newtonian flow behaviour. It was demonstrated that for a highly elastic and shear thinning fluid, the rotation of the disk generated a secondary flow which was in the opposite direction to that for a Newtonian fluid as a result of induced normal stresses in the fluid. Subsequent publications by Hill (1969,1972), Böhme, Ruburt & Stenger (1992), Day et al. (1996), Escudier & Cullen (1996) and Wusch & Böhme (1996) have all used fluids which are highly shear thinning and contain varying amounts of elasticity. A description of the experimental observations follows with a summary presented in table 2.1.
Böhme et al. (1992) performed experiments using solutions with 0.1% carboxymethylcellulose in glycerol-water solvents to produce two highly shear thinning solutions in order to investigate the effect of shear thinning on vortex breakdown. A shear thinning parameter ($\beta$) was defined by Böhme et al. (1992) which was independent of the rotation rate of the disk and defined by:

$$\beta = \frac{\eta_s (\eta_k - \eta_s)}{\rho \tau_c d^2} \quad (2.7)$$

$\eta_s$ is the solvent viscosity, $c$ is the polymer concentration, $d$ is the diameter of the disk, and $\tau_c$ is a constant reference stress which is determined by fitting the viscosity curve to a master curve. The fluids investigated by Böhme et al. (1992) had shear thinning parameters of $\beta = 0.13$ and $\beta = 1.0$ with $\beta = 0$ indicating a Newtonian fluid. Experiments were performed at high Reynolds number, where inertia dominated and the elasticity was considered negligible, although no measurement of any elastic material properties were presented. ‘Newtonian-like’ flow was observed, however, the existence domain for vortex breakdown decreased in size as the shear thinning parameter was increased. The breakdown domain also shifted to higher values of aspect ratio, as shown in figure 2.7 where the domain for the two shear thinning fluids is compared to that for a Newtonian fluid.

Escudier & Cullum (1996) observed the confined swirling flow of highly shear thinning carboxymethylcellulose solutions for concentrations of 0.75 - 1.5% and with Reynolds numbers of $Re < 174$. The primary normal stress difference was measured but the fluid was considered relatively inelastic due to low values in elasticity number ($El < 0.002$). At all rotation rates examined, a vortex was observed on the disk which was dominated by inertia such that the secondary flow was in the ‘Newtonian’ direction and driven outwards along the rotating disk. However, a counter rotating vortex was observed in the upper portion of the flow cell which was driven in the ‘reverse’ direction with a very slow secondary velocity and was near stagnant. An upward flowing jet of fluid containing a wavy structure was also present in several observations of Escudier & Cullum (1996) along the axis of symmetry.
FIGURE 2.7- Comparison of the existence domain of vortex breakdown by Bohme et al (1992) between a Newtonian fluid and two shear thinning fluids.
A range of highly elastic shear thinning polyacrylamide solutions (0.016 - 0.52 \%) were used by Hill (1969, 1972), in solvents of glycerol and water, to examine the effect of elasticity in swirling flow. ‘Reverse’ flow occurred for highly elastic liquids at low Reynolds number where the secondary flow was inwards along the rotating lid against centrifugal forces, upwards along the central axis away from the rotating disk and then along the outside stationary walls. At higher levels of Reynolds number and lower values of elasticity, complex flow patterns were observed where an inertially driven ‘ring’ vortex formed at the edge of the rotating disk, counter rotating with the main ‘reverse’ flow vortex structure, as shown pictorially in figure 2.8. Further increases in disk speed caused the growth of the outer edge vortex and then a highly unsteady pattern where the flow changed direction in a ‘confused’ manner. For low concentrations of polymer (0.03\%), Hill (1972) observed the flow to transform from ‘reverse’ flow at low Reynolds number to ‘Newtonian’ flow at high Reynolds number. Hill (1972) only observed ‘Newtonian’ flow without any apparent elastic behaviour for concentrations of polymer of 0.015 \%. Hill (1972) also measured the tangential and radial velocities for one highly elastic liquid in the ‘reverse’ flow state.

Day et al. (1996) used a highly elastic shear thinning polyacrylamide solution (2.5\% in water) and observed ‘reverse’ secondary flow at low Reynolds number. On increasing the Reynolds number, Day et al. (1996) observed the formation of a ring vortex on the centre of the disk and an instability where the core of the main vortex was observed to spiral with the primary motion of the fluid, as depicted in figure 2.9.
**Figure 2.8** - Depiction of the secondary flow patterns observed by Hill (1972) for shear thinning viscoelastic fluids showing (a) Newtonian-like flow, (b) 'reverse' or elasticity driven flow, (c) counter-rotating vortices with an inertia driven ring vortex located on the edge of the rotating disk. (a) and (b) were also predicted by Kramer & Johnson's (1972) analysis using a second order fluid with $El = 0$ and $El = 0.5$ respectively.
FIGURE 2.9 - Depiction of the three-dimensional spiral instability observed for a shear thinning elastic polyacrylamide solution by Day et al. (1996).
<table>
<thead>
<tr>
<th>Observation</th>
<th>$Re_0$</th>
<th>$We_0$</th>
<th>$El_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Hill et al (1972)</strong> $H/R = 1$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.016-0.52 % PAA in 10-54% glycerol-water</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>'Reverse' flow</td>
<td>&lt; 1</td>
<td>&lt; 0.5</td>
<td>0.1 - 60</td>
</tr>
<tr>
<td>Unsteady flow/instability</td>
<td>0.1 - 0.4</td>
<td>0.3 - 0.5</td>
<td>0.7 - 4</td>
</tr>
<tr>
<td>Counter-rotating vortices</td>
<td>0.2 - 20</td>
<td>0.3 - 0.5</td>
<td>0.02 - 2</td>
</tr>
<tr>
<td>'Newtonian' flow</td>
<td>0.4 - 90</td>
<td>0.02 - 0.4</td>
<td>0.004-0.04</td>
</tr>
<tr>
<td><strong>Day et al. (1996) $H/R = 1.8$</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.5 % PAA in water</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>'Reverse' flow</td>
<td>0.88</td>
<td>0.88</td>
<td>1</td>
</tr>
<tr>
<td>Instability</td>
<td>1.57</td>
<td>0.94</td>
<td>&lt; 0.6</td>
</tr>
<tr>
<td><strong>Escudier and Cullen (1996) $1.5 &lt; H/R &lt; 2.84$</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.75 - 1.5 % CMC in water or glucose-water</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Counter rotating vortices</td>
<td>7 - 174</td>
<td>&lt; 0.014</td>
<td>&lt; 0.002</td>
</tr>
<tr>
<td><strong>Böhme et al. (1992) $1 &lt; H/R &lt; 3$</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.1 % CMC in 60-80 % glycerol-water</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vortex breakdown</td>
<td>&gt;1000</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
2.3.2 Theoretical Analyses

Numerical methods have been used in an attempt to predict the flow patterns observed for non-Newtonian fluids in confined swirling flow for both elastic and inelastic fluids. The constitutive equations used by Böhme et al (1992) and Escudier & Cullum (1996) described inelastic fluids while those used by Kramer (1969) and Kramer & Johnson (1972), Nirschl & Stewart (1984), and more recently by Chiao & Chang (1990), described elastic fluids.

Böhme et al. (1992) performed a finite-element simulation and used a generalised Newtonian model in order to model only the shear thinning viscosity of the fluids he used in his experiments at high Reynolds number. Vortex breakdown was predicted numerically with reasonable accuracy but with some departure from the size and location of the first breakdown bubble observed experimentally. Böhme et al (1992) associated the deviations between the experiments and numerical prediction as possibly due to the elasticity of the fluid, which was not considered in the constitutive equation used.

Escudier & Cullen (1996) used the commercial computational fluid dynamics package ‘Polyflow’ at moderate Reynolds numbers, with the shear thinning viscosity behaviour for their experimental fluids represented by the Cross model, however fluid elasticity was not taken into account. The numerical model predicted ‘Newtonian’ like flow to govern the whole flow cell and it was not able to predict the counter rotating vortices observed in the experiments. Therefore, the constitutive model used was inadequate to describe the flow kinematics of their slightly elastic shear thinning fluids.

Kramer (1969) and Kramer & Johnson (1972) were the first to try and predict the effect of elasticity in confined swirling flow and hence reproduce the experimental observations made by Hill (1972). Kramer & Johnson (1972) used a perturbation theory for a weak secondary flow superimposed on an arbitrary primary flow using
both a second order fluid model, which assumes a constant viscosity and a constant primary normal stress coefficient, and the WJFLMB constitutive model of Spriggs et al. (1966), which assumes a power law form of material functions. Figures 2.8 and 2.10 show the qualitative observations made by Kramer & Johnson (1972) when they used the second order fluid model for constant Reynolds number and varied the normal stress coefficient, which equated to a variation in the elasticity number of $0 \leq El \leq 0.5$. An elastic vortex will be defined here as a secondary flow vortex which is flowing radially inwards along the rotating disk and up the central axis (i.e. ‘reverse’ flow) while an inertially driven vortex is defined as one which flows in typical Newtonian fashion and radially outwards along the rotating disk due to centrifugal forces. As the elasticity number was increased from $El = 0$ to $El = 0.0125$, a small elastically driven vortex formed on the outer edge of the rotating disk in an otherwise Newtonian flow field. The elastic vortex then governed a majority of the flow field with a further increase in elasticity number to $El = 0.025$, and only a small inertial vortex remained on the centre of the rotating disk. ‘Reverse’ flow was then predicted for an increase in elasticity number to $El = 0.5$ with elastic effects fully dominating. The predictions using the second order fluid model were not found to qualitatively predict the observations of Hill (1972), except for the case when the flow was dominated by elasticity and only ‘reverse’ flow was observed. The inadequacy of the second order model to describe the full flow kinematics is not surprising considering the model does not account for shear thinning or fluid memory, and hence it is only capable of predicting the rheology of a fluid to a very limited degree. The second order model was also found to be highly inaccurate when comparing the analytical velocity measurements with those made by Hill (1972). However, the WJFMB model, which considers the variation in shear dependence of relaxation time and viscosity, was found to predict the tangential and radial velocity profiles with reasonable accuracy for the case when ‘reverse’ flow was observed.

A global spectral method was used by Chiao & Chang (1990) while an orthogonal collocation method was used by Nirschl & Stewart (1984) with both methods using
FIGURE 2.10 - Secondary flow patterns predicted by Kramer & Johnson (1972) for a constant-viscosity second order fluid with (a) $El = 0.0125$, and (b) $El = 0.025$. 


the Criminale-Ericksen-Filbey (CEF) constitutive equation in attempt to predict the observations of Hill (1972). Shear thinning and elasticity were taken into consideration in the constitutive model by using the Carreau A model (Bird et al. 1987a) to describe the material functions for the rheological data of Hill (1972). However, Chiao & Chang (1990) state that the CEF fluid has some physical limitations and was a mathematical obstacle due to third order terms in the equation. However, the order of magnitude of the third order terms was considered negligible relative to the second order terms, provided the Reynolds number was high and Weissenberg number low or vice versa. This was considered to be the case for most of Hill’s (1972) experiments.

Both Nirschl & Stewart (1984) and Chiao & Chang (1990) were able to predict ‘reverse’ flow in many of the cases for when Hill (1972) observed it experimentally. Also, the predicted tangential and radial velocity profiles compared very well to Hill’s (1972) measurements for a ‘reverse’ flow situation. However, Nirschl & Stewart (1984) were unable to predict the counter rotating inertially driven vortex on the outer edge of the disk, which was observed by Hill (1972). Instead, an inertial vortex was predicted to form on the centre of the disk, which was similarly predicted by Kramer & Johnson (1972) at moderate values of elasticity number. Chiao & Chang (1990) were able to predict a counter rotating vortex on the outer edge of the disk for several of the cases observed experimentally by Hill (1972). Chiao & Chang (1990) also predicted a region of temporal instabilities and chaotic flow, which were believed to be consistent with some observations made by Hill (1972) at high Reynolds number.

Wünsch & Böhme (1996) used a single-integral constitutive equation (Wagner model) to predict their own experimental results for a shear thinning elastic polyacrylamide solution. Although limited details were presented, the flow patterns were found to be similar to those observed by Hill (1972). Wünsch & Böhme (1996) state that the experimentally observed flow behaviour was qualitatively predicted when the Weissenberg number was altered for a set Reynolds number.
2.3.3 Related Cylindrical Swirling Flows

The open cylinder with rotating bottom lid was used experimentally for a shear thinning elastic liquid (2.5% polyacrylamide in water) and a constant viscosity elastic liquid (silicon oil) by Böhme, Voß & Warnecke (1985). 'Reverse' flow was observed at low Reynolds number ($Re < 0.013$ for 2.5% polyacrylamide) and a bulge in the free surface was produced which depended on the primary normal stress difference. The effect was termed the Quelleffekt because the fluid flowed upwards along the axis of symmetry as a source or Quell. Böhme et al. (1985) used second order theory assuming a sufficiently slow flow and solved the flow problem using a numerical finite element method. The numerical results for the surface bulge agreed well with experiment. Böhme et al. (1985) found that the zero-shear rate normal stress coefficients could be determined by measuring the displacement of the free surface, and found that surface tension of the fluid had insignificant influence on the result. It was also found that the axial bulge deformation was quadratic in the angular velocity of the rotating disk for low angular velocities. Debbaut & Hocq (1992) used the Oldroyd-B and Johnson-Segalman constitutive models to predict the bulge shape observed by Böhme et al (1985). The relative importance of the two normal stress differences was examined by using the Johnson-Segalman equation because the second normal stress difference is quantified while in the Oldroyd B model it vanishes. The surface bulge was found to be larger using the Oldroyd B model indicating that the second normal stress difference acts against the first normal stress difference as far as the bulge shape was concerned. Both models assume a constant viscosity for the test fluid, but the predictions were compared to the behaviour of a fluid which was highly shear thinning. However, very good quantitative agreement on surface displacement was found between the predictions of Debbaut & Hocq (1992) and the experiments by Böhme et al. (1985). Siginer (1991) found that surface tension was important when measuring surface deformation to yield normal stress coefficients. Siginer (1991) also predicted sectional streamline patterns for the secondary flow and found that various sets of counter rotating vortices were observed which were dependent on the elasticity of the fluid and cylinder aspect ratio.
2.3.4 Summary

All previous experimental investigations on the confined swirling flow of non-Newtonian fluids has been performed using shear thinning elastic liquids, although in some cases the elasticity was considered relatively negligible. When the relaxation time has been determined for the various fluids, it has also been invariably shear rate dependent. Both the Reynolds number and the Weissenberg number are consequently evaluated using the zero-shear rate value of viscosity and with a shear rate dependent relaxation time respectively. This is despite the fact the lid may be rotating at substantial rates and that the material functions will vary throughout the flow cell. Comparison between different experiments and fluids has been difficult due to the inability of the Reynolds number, Weissenberg number (or elasticity number) and a shear thinning parameter to fully characterise the flow field. If the fluid is shear thinning, it is difficult to distinguish between the effects of shear thinning and those of elasticity, especially when the Reynolds number is high. It is then difficult to ascertain the role of elasticity in all of the above mentioned observations. Also, prediction of the flow field for non-Newtonian fluids has not been able to produce all the observations made by Hill (1972), due to limitations in the constitutive equations used or in the data presented by Hill (1972). However, some good comparisons between experiments and numerical results were found for the case of 'reverse' flow of Hill's (1972) most elastic fluid. Numerical prediction in confined swirling flow is possible, however, there is a need for experimental results using well characterised fluids, which can be described by more sophisticated constitutive models than those that have been previously used.

2.4 ROTATING FLOWS OF VISCOELASTIC FLUIDS AND ELASTIC FLOW INSTABILITIES

The following discussion is an outline on swirling flows of viscoelastic fluids for geometries other than the torsionally driven cavity. The swirling flows to be discussed include rotating disk flows and Taylor-Couette flow. These flows are
related to the torsionally driven cavity flow and hence the general behaviour of viscoelastic fluids in such flows is applicable. Review articles which include discussion on the subject are those of Petrie & Denn (1976), Larson (1992), and Shaqfeh (1996) while various elastic flow phenomena are detailed in a variety of rheological texts (eg. Tanner 1985; Bird et al. 1987a; Barnes, Hutton & Walters 1989) and collectively illustrated in Boger & Walters (1993). Detailed discussion will not be pursued, with the following section offered only as an introduction to the types of phenomena observed in the swirling flow of viscoelastic fluids.

2.4.1 Rotating Disk Flows

Rotating disk flows include those generated by an infinite or finite disk in semi-infinite fluid and those where the fluid is bounded by two infinite or finite disks. Rotating disk flows have been widely investigated in Newtonian fluid mechanics and were first investigated by von Karman (1921) for an infinite rotating disk in a semi-infinite fluid and was discussed previously in section 2.3.4.

In the rotating disk flow of viscoelastic fluids, experimental investigations using submerged finite disks include those by Hansford & Litt (1968) and Griffiths, Jones & Walters (1969) using fluids which also had a shear thinning viscosity. Hansford & Litt (1968) observed three different flow regimes during a study on mass transfer in viscoelastic fluids, with a qualitative depiction shown in figure 2.11. At low disk rotation speeds, the secondary motion of the fluid was inwards along the disk in the opposite direction to that for Newtonian fluids. At intermediate disk speeds a counter rotating vortex was produced on the centre of the disk, along with a ‘spiral’ moving away from the disk, while ‘Newtonian-like’ flow was observed at high disk speeds. Griffiths et al. (1969) provide a theoretical analysis of the secondary flow of a viscoelastic fluid using an Oldroyd-type equation of state. When an infinite disk was used, the secondary flow was observed to be radially inwards along the rotating disk in the same direction as that for a Newtonian fluid. This was contrary to the secondary flow when a finite disk was used in a cylindrical vessel where the flow was inwards along the disk in both a theoretical and experimental analysis. Therefore,
FIGURE 2.11 - Depiction of three flow regimes observed by Hansford & Litt (1968) in the rotating disk flow of a viscoelastic fluid indicating (a) centrifugal flow, (b) toroidal flow (counter-rotating vortices), and (c) elastic 'reverse' flow.
inwards motion for a viscoelastic fluid arises from the finiteness of the disk, which subsequently causes the whole flow field to be in the opposite direction to that for an infinite disk or for a Newtonian fluid. At medium levels of elasticity, a counter rotating vortex was observed to form on the centre of the disk.

Numerous analytical studies have been performed for the case of a rotating disk in a fluid using non-Newtonian equations of state and most generally taking the von Karman (1921) type approach (Jain 1961; Rathna 1962; Mitschka & Ulbrecht 1966; Griffiths et al. 1969; Kelkar et al. 1972; Elliot 1971; Williams 1976; Williams 1980; Ulbrecht & Gasparetto 1980; Phan-Thien 1983). For the case of an infinite rotating disk, different predictions on whether ‘reverse’ flow should occur for viscoelastic fluids were made, while ‘reverse’ flow was predicted and observed for a finite disk. Examples of the prediction of essentially ‘Newtonian-like’ flow for viscoelastic fluids includes the work of Griffiths et al. (1969), Elliot (1971), Williams (1976) and Phan-Thien (1983) while ‘reverse’ flow was predicted by Sharma (1959) and Rathna (1962). The predictions of Sharma (1959) and Rathna (1962) used an approximate momentum-integral method which was used for Newtonian fluids by von Karman (1921). However, Cochran (1934) showed that the method of von Karman was not sufficiently precise in determining velocity profiles and that the profiles can be somewhat arbitrarily chosen to satisfy certain conditions. Therefore, Williams (1976) concluded that the momentum-integral approach was not appropriate for an investigation of secondary flow phenomenon for non-Newtonian fluids. Instead, Williams (1976) used the method of Cochran (1934) and Benton (1966) for slow steady flows of Newtonian fluids, but with the Oldroyd-B constitutive equation, and did not predict ‘reverse’ flow. A more recent analytical study on the infinite rotating disk problem using viscoelastic constitutive models is that of Buggish & Anksel (1990). Buggish & Anksel (1990) also highlight that the Newtonian and non-Newtonian problems are essentially identical and that elastic effects arise only when suction to the plate is applied.
Berman & Pasch (1986) examine the effect of viscoelasticity on the boundary layer on a finite rotating disk using polyethyleneoxide solutions and compare the observations with the analytical study of Phan-Thien (1983a). In both cases the boundary layer was observed to initially show a slight decrease in size and then a much larger increase as the Weissenberg number was raised. Phan-Thein (1983a) used the Maxwell fluid model and also predicted that the secondary circulation (radial velocity) would decrease with increasing Weissenberg number ($We = 1 - 2$). In the experiments of Berman & Pasch (1986), their dilute polyethyleneoxide solutions showed a substantial decrease in radial velocity with increasing Weissenberg number.

The submerged rotating disk flow has also been used in the study of drag reduction (Kale, Mashelkar & Ulbrecht 1975; Ulbrecht & Gasparetto 1980; Berman & Pasch 1986; and Choi & Jhon 1996). Elastic fluids were observed to experience a lower torque, and hence drag coefficient, when compared to Newtonian fluids. Under laminar conditions, a reduction in torque was associated with a reversal of the secondary flow (Ulbrecht & Gasparetto 1980). In turbulent flows, there is still no universally accepted model for drag reduction. Proposed mechanisms include that a reduction in turbulence is the result of polymer extension, which suppresses the formation of eddies, or that it is the result of boundary layer alterations associated with the addition of polymer (Morgan & McCormick 1990).

Similar experimental observations to rotating submerged disk flows have been made for the case of a rotating sphere submerged in viscoelastic fluids (Giesekus 1963, 1964, 1965, 1969; Walters & Waters 1963, Walters & Savins 1965). A secondary flow was also observed in the ‘reverse’ direction for viscoelastic fluids when compared to Newtonian fluids, while decreasing elasticity or increasing inertia caused the production of counter rotating vortices. The flow patterns for a rotating sphere have also been predicted (Giesekus 1963; Thomas & Walters 1964; and Williams 1980). Qualitative predictions for the rotation of a sphere compared reasonably well with the experimental observations.
Other rotating disk flows include those for two parallel disks and for the cone-and-plate geometries. These flows are important due to their direct application in rheometry, where they are used to determine viscoelastic material functions. Early experimental investigations (Cox 1962; Hoppmann & Miller 1963; Hoppmann & Baronet 1964; Giesekus 1965, 1967) and analytical studies (Rao 1962; Bhatnager & Rathna 1963; Giesekus 1967) on such flows identified the operating constraints for use in rheometry such as the need for small gaps between the plates (disk or cone), in order to minimise secondary flows associated with either inertia or elasticity.

Flow transitions in parallel disk flow and cone-and-plate flow were first observed by Jackson, Walters & Williams (1984), where an increase over time was observed for the torque and normal force in anti-thixotropic fashion for a polyacrylamide Boger fluid, using a parallel plate rheometer. Binnington & Boger (1986) and Steiert & Wolf (1990) subsequently observed similar flow transitions for other Boger fluids. The flow transitions were characterised as a hydrodynamic instability, through the experimental investigations of Magda & Larson (1988), Laun & Hingmann (1990) and McKinley et al. (1991), which depended on the angular rotation rate while the critical shear rate was inversely dependent on the gap size. The characterisation of the flow field has been assisted by numerous analytical studies (Phan-Thien 1983ab, 1985; Walsh 1987; Ji, Rahagopal & Szeri 1990; Crewther, Huilgol & Jozsa 1991, Avagliano & Phan-Thien 1996, Renardy & Renardy 1998). Multiplicity of solutions have been found with bifurcation of the flow and generation of instabilities.

The experimental and analytical analysis of Byars et al. (1994) has provided the most comprehensive understanding of the instability observed between parallel plates by using two polyisobutylene Boger fluids. Spiral vortices were observed to emanate at some critical radius \( R_1 \), and travel radially outwards to disappear beyond a second critical radius \( R_2 \), as depicted in figure 2.12. The spirals have the form of Archimedean spirals, with the secondary flow being three-dimensional and time-dependent, while the spatial characteristics scale well with the rotation rate and the axial separation of the disks. A linear stability analysis using the Chilcott-Rallison
FIGURE 2.12 - Parallel plate flow with an upper rotating disk showing (a) geometry, and (b) depiction of an elastic instability in the form of a spiral vortex.
model was found to describe the wave speed, wavelength, and azimuthal structure of the flow instability reasonably well, while the critical radii were under predicted. A mechanism of the instability, proposed by Byars et al. (1994), was based on a disturbance-energy analysis used previously to analyse purely elastic instabilities in Taylor-Couette and Taylor-Dean flows (Joo & Sheqfeh 1991, 1992, 1994). The analysis involves determining the energy transfer between the mean flow and the disturbance using a mechanical energy balance. The mechanism is associated, through curvilinear streamlines, with the creation of a perturbation in the azimuthal normal stress ("hoop stress", $\sigma_{\omega}$) as a result of the non-linear coupling between the base state velocity field and the disturbance polymeric stress. Similar results were also found for the cone-and-plate geometry, except the spiral instability covers the entire flow domain because there is a constant shear rate throughout the flow cell (Byars, 1995).

2.4.2 Taylor-Couette Flow

Another swirling flow of relevance is the Taylor-Couette flow which has been most recently reviewed by Larson (1992) and Shaqfeh (1996). These reviews will be largely used in the following discussion. In the circular Couette flow, fluid is confined in the gap between two cylinders, with the flow driven by the rotation of one or both cylinders. An inertial instability was first observed for Newtonian fluids by Rayleigh (1880), and followed by Taylor (1923) and Chandreskar (1961), with a depiction of the secondary flow field shown in figure 2.13. The stationary inertial instability was found to have the form of toroidal flow cells, which were approximately equal in size with the gap width, and were spread axially along the cylinder as the centrifugal force flung fluid to the outside. Wavy vortices were generated as the Reynolds number was increased further, with turbulence resulting at very high Reynolds numbers.

The effects of slight elasticity on the inertial instability were investigated by Ginn & Denn (1969) and Giesekus (1966), where weak elastic effects were described using a second order fluid model. The analysis predicted that elasticity would stabilise the
Figure 2.13 - Taylor-Couette geometry consisting of coaxial cylinders. Depiction of the secondary flow cells once a critical Reynolds number is exceeded for an inertial instability.
inertial instability, provided the second normal stress difference was not close to zero. The stabilising effect has been found to occur for many cases experimentally but not in all situations (Denn & Roisman 1969; Sun & Denn 1972; Friebe 1976). As the elasticity was raised, the effect on the inertial instability and the general flow behaviour depended on the competing values of primary normal stress difference and second normal stress difference, as well as the level of shear thinning. The primary normal stress difference and shear thinning have a destabilising effect while the second normal stress difference has a stabilising effect on the inertial instability. Viscoelastic fluids have also been observed to show wavy cells which are time-periodic and similar to those observed for Newtonian fluids, except that the viscoelastic wavy cells occur at a lower Reynolds number than the stationary instability. A ‘spiral type’ instability can also occur at still lower Reynolds numbers (Friebe 1976).

At very low Reynolds numbers ($Re << 1$), Geisekus (1966) observed the onset of a cellular instability in Taylor-Couette flow for a viscoelastic fluid. However, it was only recently that Muller, Larson & Sheqfah (1989) observed the dramatic increase in torque on the rotating cylinder while performing viscosity measurements in the system for a Boger fluid. The Reynolds number was much less than one and the increase occurred after five to ten minutes, after which the measurement oscillated in time. The flow field after the transition was a well defined banded vortex structure which evolved over time into a complicated multi-wavelength structure. Further experiments on Boger fluids have elucidated the general characteristics of what is regarded as a purely elastic instability and the critical conditions for its occurrence (Larson et al. 1990; Sheqfah et al. 1992; Muller et al. 1993; Braumert & Muller 1995).

Numerous analytical studies have been performed which mainly apply perturbations to the flow of model fluids (eg Oldroyd-B fluid) in both a linear and non-linear analysis with a discussion of the results found in the review by Shaqfah (1996). A mechanism by which polymers create a purely elastic instability was proposed by
Larson et al. (1990) for the axisymmetric mode of the instability. Their analysis considered that the azimuthal normal stress, or ‘hoop’ stress, in a base-state can be acted upon by a fluctuation in the velocity gradient, which creates an even larger hoop stress and thus it moves away from its original state. The instability mechanism for non-axisymmetric mode was examined by Joo & Sheqfeh (1994) using an energy analysis, however, it is essentially the same as for the axisymmetric case, except that the perturbation is caused by a non-axisymmetric velocity variation. These mechanisms are similar to that described previously for parallel plate flow.

2.5 SUMMARY

The effects of fluid elasticity in various swirling flows have been examined. In confined geometries, elasticity is apparent in a swirling flow as a large azimuthal normal stress acting along curved primary streamlines. The normal stress causes a tension along the streamline, in the form of a ‘hoop stress’, resulting in a force directed radially inwards in the opposite direction to centrifugal forces. The secondary flows produced when a viscoelastic fluid with high elasticity is subject to swirl is found to be in the ‘reverse’ direction to what would be observed for a Newtonian fluid. Flow geometries where ‘reverse’ flow has been observed include: the torsionally driven cavity; submerged rotating disk, sphere, or rod flow; and parallel-plate and cone-and-plate (large gaps) flows. Complicated flows may also be generated comprising of a mixture of inertial and elastic driven vortices depending on the governing parameters for the flow such as the Reynolds and Weissenberg numbers and the level of shear thinning. Unstable flow fields may also be generated as a result of the competing elastic and inertial forces. Purely elastic instabilities are typically observed for highly elastic non-shear thinning solutions in swirling flows for Reynolds numbers of the order of, or less than, one. The most well documented mechanism for the elastic instability is that caused by slight fluctuations in the velocity field which reinforces the ‘hoop stress’, altering the original flow state.
Viscoelastic fluids are not described by a unique equation of state in the same way as Newtonian fluids. Constitutive models which are proposed to describe a viscoelastic fluids must be tested by comparing both their ability to describe viscoelastic material properties (such as the viscosity, primary normal stress, storage modulus and extensional viscosity), and their ability to predict the flow kinematics accurately when substituted into the equations of motion. Therefore, many experiments have been performed throughout the literature to provide information for which to test non-Newtonian constitutive models, with the most well documented model flows being the contraction flow and flow around a sphere.

This thesis sets out to provide an experimental study on the confined swirling flow of viscoelastic fluids using a torsionally driven cavity. The torsionally driven cavity has been chosen due to its well defined, yet three-dimensional, geometry. All previous experimental work has involved shear thinning fluids, and hence the resulting flow kinematics cannot be attributed solely to the effects associated with fluid elasticity. In this thesis, elasticity is isolated by examining the confined swirling flow of a collection of constant viscosity elastic liquids (Boger fluids) which are considered as ‘ideal’ fluids. It is envisaged that the results presented will be ideal for comparison to numerical predictions in confined swirling flow. Constitutive models and numerical techniques for steady and unsteady flow of viscoelastic fluids can therefore be tested in a complex flow using well characterised fluids.
CHAPTER 3

EXPERIMENTAL METHODS AND MATERIALS

3.1 INTRODUCTION

The following chapter discusses the experimental techniques in essentially two parts:

- fluid preparation and characterisation
- confined swirling flow apparatus and flow visualisation techniques.

The majority of the fluids used in this study are constant-viscosity elastic liquids which are commonly referred to as Boger fluids (Boger 1977/78). The solutions are typically made up of dilute concentrations of polymer, which have molecular weights of the order of one million, in viscous solvents. The important characteristic of Boger fluids is their constant viscosity because it enables the separation of elastic and shear thinning effects in the flow kinematics of viscoelastic fluids. The method of preparation for all the fluids used in this study is outlined along with descriptions of the polymers and solvents. The techniques used to characterise the fluids are discussed and involves a description of several rheological instruments and their accuracy. The test fluids are essentially characterised such that they can be modelled using non-Newtonian constitutive equations and hence some of the key models are explored with particular reference to their material functions.

The confined swirling flow apparatus is a torsionally driven cavity which is described in detail. The flow visualisation techniques used are presented and in particular the technique of Particle Image Velocimetry (PIV), used to obtain radial and axial velocity measurements in the secondary flow plane, is examined in detail.
3.2 POLYMERS, SOLUTIONS AND PREPARATION

The following section initially describes the solvents used and their properties. The polymer materials are then examined with a brief review of their properties followed by the method used to make the viscoelastic test fluids. A listing is also provided of all the fluids used in the confined swirling flow apparatus with each solution given a reference code.

3.2.1 Solvents

Various aqueous solvents were constructed using either de-ionised water or a viscous fluid and de-ionised water. The de-ionised water was obtained from a Millipore Milli-RO\textsuperscript{®}4 water purification system which utilised 10 and 3 µm pre-filters, a 10µm carbon filter and a reverse osmosis unit. The water typically had a conductivity of about 2 - 8 µS/cm and pH ranging from 5.5 - 7. The viscous fluid additives used were either wheat syrup, fructose (powder), or glycerol. The wheat syrup was 43°Be provided by Weston Bioproducts Ltd., Melbourne, Australia, with a refractive index of 1.5. It was yellow in colour but optically clear. The syrup comprised of a combination of several sugar groups including dextrose (18%), maltose (13%) maltotriose (12%) and higher sugars according to the manufacturer. The glycerol was technical grade (98 %) purchased from Ajax Chemicals Ltd. Australia. The fructose was technical grade obtained in powder form from Hoechst Australia Ltd..

The viscosity of the solvents in de-ionised water is presented as a function of concentration in figure 3.1 using rheological techniques discussed in section 3.4. Several of the results for wheat syrup and glycerol have previously been published by Zirnsak (1995), Weisser (1996) and Mun (1996). The viscosity of each solvent rises dramatically with increasing concentration.
FIGURE 3.1 - Viscosity of various Newtonian solvents in de-ionised water as a function of concentration for temperatures of about 20-22 °C.
3.2.2 Polymers

The following polymers were used in this study with abbreviations given in parentheses:

- polyacrylamide, Separan AP30 - (PAA or PAA AP30)
- polyacrylamide, MG500 - (PAA MG500)
- xanthan gum, Keltrol - (XG)
- polyethyleneoxide, poly-dispersed - (PEO)
- polyethyleneoxide, mono-dispersed - (PEOm)

The polymers were either provided or purchased from the following companies: the two polyacrylamides were provided by DOW Chemical Ltd., USA; the xanthan gum was provided by Kelco, Division of Merck & Co. Inc., USA; the poly-dispersed polyethyleneoxide (PEO) was manufactured by E.I. Dupont de Nemours & Company and purchased from Aldrich Chemicals Ltd., Melbourne, Australia; and the mono-dispersed polyethylene oxide was purchased from Polymer Laboratories Inc, Amherst, USA. Each polymer will now be considered in turn.

**POLYACRYLAMIDE**

Solutions of polyacrylamide are used to study the swirling flow of high molecular weight flexible polymer. The molecular weight of the PAA Seperan AP30 has been reported in the literature to span the range of $2-4 \times 10^6$ by Zirnsak (1995), Hur (1987), Eisenbrand & Goddard (1982), Tam & Tiu (1989ab) and Lawler *et al.* (1986). Separan AP30 is a polyelectrolyte and contains both amide and carboxylic groups, with a 25-30% degree of ionisation, and is approximately represented by the chemical formula presented in figure 3.2. PAA MG500 is considered by the manufacturer to be similar to Separan AP30.

Polyacrylamide Separan AP30 can be classified as an anionic polyelectrolyte in neutral and alkaline solutions and the rheology of polyacrylamide solutions is strongly affected by pH. Under acidic conditions the ionisation is repressed and the

56
FIGURE 3.2 - The chemical structure of polyacrylamide Separan AP30.
polymer assumes a non-ionic character, which results in a low viscosity and low elasticity of the solutions. Under basic conditions (pH ≈ 8), the viscosity and degree of elasticity reaches a maximum due to a large degree of ionisation of the polymer chain (Volk & Friedrich 1980).

The solvent properties can be controlled in order to alter a flexible polymer’s conformation in solution (Flory 1953). A good solvent is one in which the polymer and solvent have strong attractive energy such that the interaction between polymer segments is repulsive, which causes an excluded volume effect. The excluded volume effect refers to the volume from which a given polymer molecule effectively excludes all others. In a θ solvent, the polymer experiences a slight repulsive interaction towards the solvent which cancels the excluded volume effect. θ conditions are typically only found at a single temperature and the fluids in this thesis are generally regarded as good solvents. Salt may be used to cause a shielding of the negative charges on the polyacrylamide polymer so that different parts of the polymer can approach more closely. This causes a coiling up of the molecule and a reduction of the hydrodynamic size in solution (Ait-Kadi et al. 1987; Ferguson et al. 1990; Muller et al. 1979; Tam & Tiu 1989ab).

**XANTHAN GUM**

Solutions of xanthan are used to study the swirling flow of a high molecular weight semi-rigid polymer. The molecular weight of Keltrol xanthan gum is reported in the literature to be $2.4 \times 10^6$ by Zirnsak (1995), $1.95 \times 10^6$ by Paradossi & Brant (1982), $3 \times 10^6$ by Norton et al. (1984), $3.5 \times 10^6$ by Sato et al. (1984) and $7.4 \times 10^6$ by Holzwarth (1978). Zirnsak (1995) determined the hydrodynamic length of the Keltrol xanthan gum sample used in this work for solutions in the presence of 0.02% sodium azide as $1250\pm 50$ nm with an aspect ratio of 625 from light scattering experiments.

Xanthan gum is an extracellular polysaccharide produced by the micro-organism *xanthomanos campestris*. Xanthan typically exists in an ordered conformation in solution having a semi-rigid rod-like structure stabilised by intramolecular non-
covalent bonding (Norton et al. 1984). However, the conformation and degree of rigidity of the polymer changes due to the influence of temperature and ionic strength (salinity). Self association of the polymer can also occur even in dilute concentrations to form aggregates in solution (Holzworth 1981; Southwick et al. 1983).

**POLYETHYLENEOXIDE**

Polyethyleneoxide (PEO) is a non-ionic synthetic polymer having the general formula $\text{H}($OCH$_2$CH$_2$)$_n\text{OH}$. PEO consists of a flexible linear chain and is readily soluble in water and solvents such as acetonitrile and anisole, chlorinated solvents such as dichloroethylene and chloroform, and aromatic hydrocarbons such as benzene and toluene at elevated temperatures. PEO shows an unusual property in water as it displays an inverse solubility-temperature relationship, which is possibly due to a hydrophilic-hydrophobic balance in the polymer system as discussed by Bailey & Koleske (1987).

Polyethers such as PEO are subject to oxidative attack and must be stabilised with an antioxidant to avoid degradation of the polymer. The degradation of PEO in aqueous and organic solutions is accelerated by strong acids, certain oxidising agents, UV light, and certain heavy metal ions. The effectiveness of the latter two components is related to the solution stability of various polymer samples upon long-term, natural ageing. These facts are interpreted by assuming that PEO degrades primarily by autooxidation, giving hydroperoxides which decompose by polymer chain cleavage. Certain oxidising materials and compounds which form stable free radicals are effective in retarding the oxidative degradation (McGary 1960).

It has been observed that PEO tends to aggregate in solution, whereby the extent of aggregation is dependent on solvent type, temperature, polymer solution concentration and type of salt solution. There is speculation about whether the aggregation is due to the behaviour of the PEO molecule itself or due to impurities associated with the sample (Polverari & van de Ven 1996; Devenand & Selser 1990, 1991; Woodley 1992; Bailey & Koleske 1976, 1987; Porsch & Sunderlöf 1995). The
observed aggregation may be the result of oxidation, which produces free radicals in solution which then provide sources for hydrogen bonding and intramolecular interaction.

Several polydispersed samples of PEO (Aldrich) were used here with the viscosity molecular weight given by the manufacturer as: $1 \times 10^6$, $2 \times 10^6$, $4 \times 10^6$, and $8 \times 10^6$. A majority of the polydispersed samples contained trace amounts of Butylated Hydroxy Toluene (BHT) which acts as an antioxidant. The PEO from Polymer Laboratories was essentially mono-dispersed ($M_w/M_n = 1.10$) and the weight average molecular weight was supplied as $M_w = 0.963 \times 10^6$. The mono-dispersed samples of polyethyleneoxide did not contain BHT.

3.2.3 Polymer Solution Preparation

Each polymer was used as provided by the manufacturer. Sodium azide ($\text{NaN}_3$) was added to most solutions at a concentration of less than 0.02 wt% to act as a biocide. Butylated Hydroxy Toluene (BHT), at a concentration of less than 1 ppm, was also added as an anti-oxidant to the PEO solutions which did not already contain BHT when supplied.

All polymers were prepared by first dissolving the appropriate amount of polymer (typically 0.1 wt%) into millipore water. The polymer was added gradually to the water while continually swirling the container, such that the polymer was dispersed throughout the solution and did not associate into clumps. Sodium azide was added to the stock solution (<0.02%). BHT was added to some of the PEO solutions (<1ppm). The solutions were usually placed on a roller mixer device at very low rotation rates for 12-48 hours. Occasionally the polyacrylamide and xanthan gum solutions were heated gently to about 40 °C to enhance dissolution. Dispersion of PEO in solution could be enhanced by heating the solutions, although dissolution of PEO decreases with increasing temperature (Bailey & Koleske 1976). Once the polymer was fully dissolved, the polymer solutions were added in appropriate
amounts to the viscous solvent and then the make-up water is added. The solutions were then mixed using a four pronged impeller at low rotation rates for 8-12 hours.

3.2.4 The Test Fluids

Three sets of fluids were used in confined swirling flow apparatus which are referred to as follows:

- low-viscosity Boger fluids
- Boger fluids (medium-viscosity and high-viscosity Boger fluids)
- shear thinning elastic fluids

Each set of fluids will be considered in turn.

LOW-VISCOSITY BOGER FLUIDS:

Low-viscosity fluids are classed as those solutions where high molecular weight polymer \( M_w > 1 \text{ million} \) is dissolved in a Newtonian solvent with the final solution having a constant viscosity and no measurable primary normal stress difference. These fluids were used in the confined swirling flow apparatus for conditions where the flow was dominated by inertia and vortex breakdown was observed for Newtonian fluids. The viscosity of the fluids range from 10 to 100 mPa s. Two solvents were used to make the low-viscosity Boger fluids, and the solvent used depended on the type of polymer being dissolved. These solvents are as follows:

- 76% glycerol - used for polyacrylamide and xanthan gum
- 60% glycerol - used for polyethylene oxide

Sodium azide \((\text{NaN}_3)\) in concentrations of less than 0.02% was used as a biocide in all solutions. Butylated hydroxy toluene (BHT) was used at concentrations of less than 1ppm as an antioxidant for those solutions containing PEO. The concentrations in each solution varied for each polymer and are expressed in terms of parts-per-million (ppm) of the final solution weight. The test fluids used in the confined swirling flow include the following concentrations of polymers:
- polyacrylamide: 25, 35, 45, 75 ppm
- xanthan gum: 25, 45, 75 ppm
- polyethyleneoxide: 150, 300, 1000 ppm for $M_w \approx 1,000k$ (PEO - 1M)
  500, 1000 ppm for $M_w \approx 2,000k$ (PEO - 2M)
  50, 200, 400 ppm for $M_w \approx 4,000k$ (PEO - 4M)

**BOGER FLUIDS**

Medium and high-viscosity Boger fluids are classed as those solutions where high molecular weight polymer ($M_w > 1$ million) is dissolved in a Newtonian solvent with the final solution having a constant viscosity and measurable primary normal stress difference. These Boger fluids have moderate to high viscosities ranging from 0.15 to 23 Pa s. They were used in the confined swirling flow apparatus for conditions ranging from where the flow was governed primarily by inertia to when the flow was dominated by elasticity. Only polyacrylamide and xanthan gum were used to make the Boger fluids with various solvent types and concentrations. The fluids and their compositions are listed in table 3.1. The polyacrylamide fluids which are referred to as fluids A to E are listed in order of increasing viscosity and elasticity. Fluid F was not examined in detail, and will only be briefly referred to, but was similar rheologically to Boger fluid B. Fluid X was the only xanthan gum fluid considered.

**SHEAR THINNING ELASTIC FLUIDS:**

Only two fluids were used in the confined swirling flow apparatus which were shear thinning and elastic. Both contained polyacrylamide and their composition is listed in table 3.2. The composition of fluid $F_{ST}$ is almost identical to Boger fluid F except it does not contain salt. The 2.5% polyacrylamide fluid is a concentrated polyacrylamide solution.
TABLE 3.1 - Composition of the 'high-viscosity' Boger fluids used in the confined swirling flow apparatus. AP30 and MG500 are two types of Separan polyacrylamide while XG Keltrol is a xanthan gum.

<table>
<thead>
<tr>
<th>LABEL</th>
<th>POLYMER type</th>
<th>wt%</th>
<th>VISCOSOUS SOLVENT type</th>
<th>wt%</th>
<th>WATER wt%</th>
<th>NaCl wt%</th>
<th>NaN₃ wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>AP30</td>
<td>0.015</td>
<td>glycerol</td>
<td>82</td>
<td>15.24</td>
<td>2.73</td>
<td>0.02</td>
</tr>
<tr>
<td>B</td>
<td>AP30</td>
<td>0.015</td>
<td>glycerol</td>
<td>90</td>
<td>8.305</td>
<td>1.66</td>
<td>0.02</td>
</tr>
<tr>
<td>C</td>
<td>AP30</td>
<td>0.015</td>
<td>wheat syrup</td>
<td>85</td>
<td>14.97</td>
<td>-</td>
<td>0.02</td>
</tr>
<tr>
<td>D</td>
<td>MG500</td>
<td>0.025</td>
<td>wheat syrup</td>
<td>88</td>
<td>11.96</td>
<td>-</td>
<td>0.02</td>
</tr>
<tr>
<td>E</td>
<td>AP30</td>
<td>.0396</td>
<td>wheat syrup</td>
<td>93</td>
<td>6.94</td>
<td>-</td>
<td>0.02</td>
</tr>
<tr>
<td>F</td>
<td>MG500</td>
<td>0.04</td>
<td>fructose</td>
<td>68.8</td>
<td>29</td>
<td>2.1</td>
<td>0.02</td>
</tr>
<tr>
<td>X</td>
<td>XG</td>
<td>0.02</td>
<td>wheat syrup</td>
<td>92</td>
<td>7.96</td>
<td>-</td>
<td>0.02</td>
</tr>
</tbody>
</table>

Keltrol

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TABLE 3.2 - Composition of two shear thinning elastic solutions used in the confined swirling flow apparatus.

<table>
<thead>
<tr>
<th>LABEL</th>
<th>POLYMER type</th>
<th>wt%</th>
<th>VISCOUS SOLVENT type</th>
<th>wt%</th>
<th>WATER wt%</th>
<th>NaCl wt%</th>
<th>NaN₃ wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fₜ겠다</td>
<td>MG500</td>
<td>0.04</td>
<td>Fructose</td>
<td>70</td>
<td>30</td>
<td>-</td>
<td>0.02</td>
</tr>
<tr>
<td>2.5% PAA</td>
<td>MG500</td>
<td>2.5</td>
<td>-</td>
<td>-</td>
<td>97.48</td>
<td>-</td>
<td>0.02</td>
</tr>
</tbody>
</table>
3.3 POLYMER CHARACTERISATION PROCEDURES

The molecular weight of a polymer molecule is extremely important in characterisation of the fundamental properties of a polymer solution. Discussed are two techniques for determining polymer molecule weight: the direct method of Gel Permeation Chromatography (GPC) in conjunction with light scattering, and the empirical method using the intrinsic viscosity. The size and conformation of a polymer molecule also plays an important role in the behaviour of polymer solutions in flow. The method of Photon Correlation Spectroscopy used to determine the hydrodynamic size of the polymers in solution is outlined.

3.3.1 Direct Molecular Weight Determination

The molecular weight of several polymer samples was determined with the assistance of Dr. Greg Allen of the Industrial Plant Biopolymer Co-operative Research Centre at The University of Melbourne using Gel Permeation Chromatography (GPC). GPC, which is also referred to as Size Exclusion Chromatography (SEC), is a liquid column chromatographic technique in which a sample solution is introduced onto a column filled with rigid porous gel, and is carried through the column by the solvent. Size separation is achieved by differential pore permeation, where larger molecules are kept away from the wall of the pore by steric interference, while smaller molecules approach the pore more closely. Larger molecules are then eluted from the column earlier than the smaller molecules and may be detected using techniques such as differential refractometry and spectrophotometry. In conjunction with the solute concentration, continual monitoring of the molecular weight of the effluent may be performed using techniques of osmometry, viscometry, light scattering, and ultracentrifugation. Light scattering, referred to as low angle laser light-scattering photometry, is probably the most common technique, where the intensity of laser light scattered by the polymer solution in the detector cell is proportional to both the molecular weight and concentration. Therefore, the absolute molecular weight may be determined using, for example, a differential refractometer in line with a light
scattering photometer, which is the method used in this work (Styring & Hamielec 1989).

Solutions of 1 g/L of polymer in 0.1 M sodium nitrate (NaNO₃) were first filtered through 0.45 μm Millipore filters (Milllex-HA disposable cellulose filtration unit or disposable membrane discs). The action of the NaNO₃ electrolyte is to minimise any electrostatic interactions that may occur between the solute and the column surface. The solutions were then pumped at a flow rate of 0.8 ml/min into two Waters Ultrahydrogel (hydroxylated polymethacrylate based gel) SEC columns with pore sizes of 250 Å and 2,000 Å respectively, which were used to separate each solution into components of different molecular weight. The aqueous eluent was filtered through a 0.6 μm Millipore type BD disposable membrane disc. The on-line detectors were a differential refractometer (Model: Waters 410) and a multi-angle laser light scattering instrument (Model: DAWN F) from Wyatt Technology Corporation. The light scattering data was analysed with the ASTRA software package from Wyatt Technology Corporation. This software also allowed the determination of the molecular weight by integrating the differential refractive index output and comparing this with the injected polymer mass.

Generally a few tests were required for each polymer tested and only the results with reproducible molecular weight distributions were used. This was achieved by only accepting those molecular weights which were concordant to within 20% and were within the expected range based on available literature for each polymer. An example of the possible accuracy of the process was examined using the mono-dispersed polyethylene oxide (PEOm). The weight average molecular weight was measured as 965 000 which is similar to the value supplied by the manufacturer of 963 000.

3.3.2 Intrinsic Viscosity

The intrinsic viscosity was measured for several polymer solutions to either determine the viscosity average molecular weight or the polymer molecule’s longest relaxation
time in a particular solvent. The following is a description of the method used for determining the intrinsic viscosity.

In even very dilute polymer solutions, the polymer molecules are very large relative to those of the solvent, and hence, it is typically found that the viscosity of such solutions is greater than that of the solvent. For a very dilute polymer solution, there is no interaction between polymer molecules, and the viscosity of the solution is that of the solvent plus the contribution of the individual polymer molecules. It is therefore possible to relate parameters derived from dilute solution viscosity measurements to molecular weight and polymer chain dimensions, and also to the interaction between polymer and solvent. These parameters can also be used in the study of chain stiffness, chain branching, polydispersity and association of polymer in solution.

The intrinsic viscosity, \([\eta]\), is defined as the zero concentration limit of the reduced viscosity \((\eta_{rs} = \eta_p / c)\), where \(\eta_p\) is the specific viscosity. The specific viscosity is defined as the relative polymer contribution to viscosity: \(\eta_p = (\eta_p - \eta_s) / \eta_s\). The intrinsic viscosity is then divided by the viscosity due to the solvent as follows:

\[
[\eta] = \lim_{c \to 0} \frac{\eta_p - \eta_s}{c \eta_s} = \lim_{c \to 0} \eta_{red}
\] (3.1)

\(c\) is the concentration of polymer, \(\eta_p\) is the zero-shear viscosity, \(\eta_s\) is the solvent viscosity. Therefore, the intrinsic viscosity is determined graphically by plotting \(\eta_{red}\) versus \(c\) and extrapolating to zero concentration. It is also found that extrapolation to zero concentration of the inherent viscosity \((\eta_{inh} = \frac{1}{c} \ln(\eta_p + 1))\) can also be used to determine the intrinsic viscosity and the same result for \([\eta]\) should be achieved.

The most common relation between dilution solution viscosity and polymer concentration is that of Huggins (1942):
\[ \frac{\eta'}{c} = [\eta] + k'[\eta]^2 c \] (3.2)

\[ \frac{1}{c} \ln \left( \frac{\eta}{\eta_0} \right) = [\eta] - k''[\eta]^2 c \] (3.3)

\( k' \) is the Huggins slope constant and is representative of the polymer-solvent system. The alternative expression of Kramear (1938) may also be used:

\( k'' \) is Kramear's constant. Huggin's slope constant and Kramear's constant are related by: \( k' + k'' = 1/2 \).

The intrinsic viscosity is typically determined by measuring the viscosity using either Cannon-Fenske or Ubbelohde viscometers due to their ability to precisely detect small differences in viscosity at low polymer concentrations. Incorrect determination of the viscosity and therefore intrinsic viscosity can arise if a solution is shear thinning and measurements should be made at low shear rates, such that the viscosity equates to the zero-shear viscosity. This may be achieved by choosing the appropriate viscometer, which are essentially capillary viscometers, and evaluating the wall shear rate. The viscometers typically used here were Cannon-Fenske viscometers (either '75' or '150' series) or a Ubbelohde viscometer of type '531-10' combined with an automated Schott-Geräte AVS350 intrinsic viscometer.

The intrinsic viscosity may be used to determine the viscosity molecular weight \( (M_n) \) using the Mark-Houwink equation as follows:

\[ [\eta] = K M_n^\alpha \] (3.4)

\( K \) and \( \alpha \) are determined from a double logarithmic plot of intrinsic viscosity and molecular weight. These parameters have been published for many systems in such references as the Polymer Handbook (Bandrup & Immergut 1975). The Mark-Houwink parameters for the polymers used in this study are listed in table 3.3. The parameters for polyacrylamide are reported for the non-ionic state and as such are not
appropriate for polyacrylamide AP30 and MG500, which are anionic. It is necessary to repress the ionic nature of the polyacrylamides before measuring the intrinsic viscosity for molecular weight determination.

Figure 3.3 shows an example of the extrapolation curve used for the determination of intrinsic viscosity for the PEOm polymer in de-ionised water at 25°C. The intrinsic viscosity was determined as 471 mL/g with the Huggin’s constant as $k’ = 0.36$ and Kramear’s constant as $k'' = 0.14$. The two sets of Mark-Houwink parameters in table 3.3 for PEO were used to determine the viscosity molecular weight with values of 964,000 and 1,117,000 calculated respectively. The first value is essentially identical to the measurement for molecular weight using GPC while the second value is within 15%.

<table>
<thead>
<tr>
<th>POLYMER</th>
<th>$K$ (cm$^3$/g)</th>
<th>$\alpha$</th>
<th>Conditions</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polyacrylamide</td>
<td>$5.6 \times 10^{-3}$</td>
<td>0.8</td>
<td>water, 25°C</td>
<td>Molyneux (1984)</td>
</tr>
<tr>
<td>(nonionic) Xanthan Gum</td>
<td>1.70</td>
<td>1.14</td>
<td>0.1M NaCl</td>
<td>Milas et al. (1985)</td>
</tr>
<tr>
<td></td>
<td>1.70</td>
<td>1.2</td>
<td>0.01M NaCl</td>
<td>Liu &amp; Norisue (1988)</td>
</tr>
<tr>
<td></td>
<td>0.063</td>
<td>0.93</td>
<td>0.5% NaCl</td>
<td>Muller et al. (1984)</td>
</tr>
<tr>
<td>Polyethyleneoxide</td>
<td>$1.03 \times 10^{-2}$</td>
<td>0.78</td>
<td>water, 25°C</td>
<td>Bailey &amp; Koleske (1976)</td>
</tr>
<tr>
<td></td>
<td>$1.192 \times 10^{-2}$</td>
<td>0.76</td>
<td>water, 25°C</td>
<td></td>
</tr>
</tbody>
</table>
FIGURE 3.3 - Determination of intrinsic viscosity for mono-dispersed polyethyleneoxide in de-ionised water at 25°C with the addition of 1 ppm BHT. Shown is the reduced and inherent viscosity for two sets of measurements taken one week apart.
3.3.3 Hydrodynamic Size Measurement

The hydrodynamic size of a polymer in some solutions was determined using Photon Correlation Spectroscopy (PCS) with the assistance of Dr. Dave Dunstun of the Industrial Plant Biopolymer Co-operative Research Centre at The University of Melbourne in a manner similar to that used by Ung et al. (1997). Samples of about 50mL were initially centrifuged at 10,000 rpm for 1-2 hours to remove dust and unwanted impurities. A commercial Malvern 4700 system was then utilised, equipped with an argon ion laser operating at 488 nm. Analysis of the correlation function, measured at a scattering angle of $\theta = 90^\circ$, was carried out using the CONTIN algorithm. The diffusion coefficients ($D$) were then converted into an effective hydrodynamic radius ($R_h$) using the Stokes-Einstein equation: $D = kT/\xi$, where $\xi$ is the friction coefficient given by $6\pi\eta_h R_h$.

The xanthan gum polymer molecule is regarded as a semi-rigid rod macromolecule in solution and the hydrodynamic radius may be used to determine the length ($L$) and radius of gyration ($R_g$) of the xanthan gum molecule by using relations given by Broersma (1960) and Young et al. (1978) for rigid rod molecules as follows:

$$L = R_s\left(2\sigma - 0.19 - \frac{8.24}{\sigma} + \frac{12}{\sigma^2}\right)$$  \hspace{1cm} (3.5)

$$R_g = \left(\frac{L^2}{12} + \frac{r^2}{2}\right)^{1/2}$$  \hspace{1cm} (3.6)

$\sigma = \ln(L/r)$ is the aspect ratio of a rod and $r$ is the radius of the 'rigid rod' and assumed to be equal to approximately $r = 2$ nm, which is of the same order as that observed in the literature (Zirnsak 1995).

The intrinsic viscosity can also be related to the molecular structure by the following expression (Flory and Fox, 1951):
\[ [\eta] = \frac{\phi R_x}{M_w} \] (3.7)

\( \phi \) is a universal constant and equals \( 2.1 \times 10^{23} \) mol\(^{-1}\).

### 3.3.4 Polymer Solution Classification

There are a few techniques which are used to classify whether solutions can be regarded as dilute, semi-dilute, or concentrated. Only two methods will be considered here. Flexible polymers tend to occupy a spherical region in solution with this volume effectively determined by measuring the intrinsic viscosity and hydrodynamic size. For a solution to be regarded as dilute, there must be no interaction between individual molecules. Hence, the criteria for ‘diluteness’ usually corresponds to when the spherical region occupied by the polymer is equal to the solution volume. For flexible polymer solutions, a standard method is to evaluate the dimensionless concentration \([\eta]c\). The solution is then regarded as dilute when \([\eta]c < 1\) (Flory 1953). An alternative method has been given by Doi & Edwards (1986) in which a solution is considered dilute when \(c N_v V_r / M_w < 1\), where \(c\) is the weight of polymer per unit volume and \(V_r\) is the specific volume of a polymer molecule defined by \(V_r = \frac{(4/3)\pi R_h^3}{M_w}\). Both methods should correspond to similar estimates of the concentration of polymer required before the solution is no longer dilute.

For rigid rod-like molecules, a similar approach to that for flexible polymers is taken, however, due to the large aspect ratio of rigid molecules, the spherical region required for the molecule to freely rotate without interference from other molecules is quite large. For rigid systems, the criteria is that \(c N_v V_r / M_w < 1\), where \(V_r\) is the spherical region available for the rigid molecule to freely rotate, and is defined by \(V_r = \frac{(4/3)\pi (L/2)^3}{M_w}\) (Note: Doi & Edwards (1986) use \(V_r \approx L^3\)).
3.4 POLYMER SOLUTION CHARACTERISATION AND RHEOLOGY

The following section outlines several rheological techniques which are used in the characterisation of polymer solutions. The rheological properties which are used to characterise non-Newtonian fluids are termed material functions. These properties are also necessary for the evaluation of non-Newtonian equations of state, which are referred to as either constitutive equations or rheological models. Simple flow fields are required to determine the material properties and these are broken into three groups: steady shear, small-amplitude oscillatory, and extensional flow. Each flow field, along with the instrumentation used, will be discussed. Several rheological models are examined with reference to the equations used to predict or utilise material functions measured for non-Newtonian fluids.

The instrumentation utilised for the determination of the material functions is summarised in table 3.4.

3.4.1 Steady Shear Rheology

A simple shear flow is one in which $v_x = \dot{\gamma}(t)y$ and $v_y = v_z = 0$ as indicated in figure 3.4(a). The stress tensor in such a flow is thus defined by the following:

$$
\sigma = \begin{pmatrix}
\sigma_{xx} & \sigma_{xy} & 0 \\
\sigma_{yx} & \sigma_{yy} & 0 \\
0 & 0 & \sigma_z
\end{pmatrix}
\quad (3.8)
$$

Note that $\sigma_{xy} = \sigma_{yx}$. The first (or primary) normal stress difference ($N_1$) and the second normal stress difference ($N_2$) are then defined as follows:

$$
N_1 = \sigma_{xx} - \sigma_{yy} 
\quad (3.9)
$$

$$
N_2 = \sigma_{yy} - \sigma_z 
\quad (3.10)
$$
Two flow geometries which are commonly utilised to simulate steady-state shear flow are the cone-and-plate and cup-and-bob devices, which are displayed in figure 3.4(b) and 3.4(c) respectively. Both geometries must be used with very small gaps between the stationary and movable surfaces, and low cone angles (< 4°) are used for the cone-and-plate geometry. This allows the stress distribution, and hence the material functions, to be constant throughout the sample. The instruments used for steady shear flow are listed as part of table 3.4.

**Table 3.4** - List of rheometers used in this study for measuring material functions of Newtonian and non-Newtonian fluids. Properties indicated are: \( \eta \) - viscosity; \( \mu \) - Newtonian viscosity; \( N_I \) - primary normal stress difference; \( G' \) and \( G'' \) - storage and loss modulus; \( [\eta] \) - Intrinsic viscosity; \( \eta_e \) - extensional viscosity.

<table>
<thead>
<tr>
<th>Geometry</th>
<th>Instrument</th>
<th>Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cone-and-plate</td>
<td>Weissenberg R19, R20, R21 rheogimeter</td>
<td>( \eta, N_I, G', G'' )</td>
</tr>
<tr>
<td></td>
<td>Carri-Med CSL100 and CSL2100 rheometer</td>
<td>( \eta, G', G'' )</td>
</tr>
<tr>
<td>Cup-and-bob</td>
<td>Contraves LS40 rheometer (Nanyang University)</td>
<td>( \eta, G', G'' )</td>
</tr>
<tr>
<td>Capillary/Intrinsic</td>
<td>Schott-Geräte AVS350 with Ubbelohde viscometer of type ‘531-10’</td>
<td>( \mu, [\eta] )</td>
</tr>
<tr>
<td>viscosity</td>
<td>Cannon-Fenske viscometers of type ‘75’ or ‘150’</td>
<td>( \mu, [\eta] )</td>
</tr>
<tr>
<td>Opposed jets</td>
<td>Rheometrics RFX extensional rheometer</td>
<td>( \eta_e^a )</td>
</tr>
<tr>
<td>Filament stretching</td>
<td>Filament stretching device (Monash University)</td>
<td>( \eta_e, \eta_e^+ )</td>
</tr>
</tbody>
</table>
FIGURE 3.4 - Geometries used for measuring rheological properties. Indicated are (a) steady shear flow between parallel plates, (b) cone-and-plate geometry and (c) co-axial cylinder or ‘cup-and-bob’ geometry.
A description of the theory for determining rheological properties using the cone-and-plate geometry may be found in Walters (1975), Barnes et al. (1989) and Bird et al. (1987a). Two types of cone-and-plate instruments were used for steady shear measurements, either a constant rate rheometer (e.g. the Weissenberg R19) or a constant stress rheometer (e.g. the Carri-Med CSL100). In the constant rate device, the plate is rotated at a constant rate and the resulting shear stress may be determined from the measurement of torque \( (C) \) on the cone. The shear rate, shear stress \( (\sigma_{xy}) \) and viscosity \( (\eta) \) are given by:

\[
\dot{\gamma} = \frac{\omega}{\theta} \quad \text{(3.11)} \\
\sigma_{xy} = \frac{3C}{2\pi r^3} \quad \text{(3.12)} \\
\eta(\dot{\gamma}) = \frac{\sigma_{xy}}{\dot{\gamma}} \quad \text{(3.13)}
\]

\( \omega \) is the angular rotation rate of the plate, \( \theta \) is the cone angle and \( r \) is the radius of the cone and plate. In the characterisation of viscoelastic fluids, a force may result from the rotation of the plate which acts to separate the plates. The total thrust \( (F) \) on the bottom plate may then be used to determine the primary normal stress difference and typically placed in terms of a primary normal stress coefficient \( (\Psi_1) \) as follows:

\[
N_1(\dot{\gamma}) = \frac{2F}{\pi R^2} = \Psi_1(\dot{\gamma})\dot{\gamma}^2 \quad \text{(3.14)}
\]

The normal stress coefficient is used to characterise the elasticity of a viscoelastic fluid and is zero for a Newtonian fluid. Two material parameters are thus determined as functions of the shear rate: the viscosity and the primary normal stress coefficient. It should also be noted that the value of the viscosity as the shear rate approaches zero usually plateaus with shear rate and is termed the zero-shear-rate viscosity \( (\eta_0) \) while the primary normal stress coefficient can also plateau to a limiting value \( (\Psi_1,0) \). A constant primary normal stress coefficient is obtained when the primary normal stress varies quadratically with shear rate.
Rheological tests on several Dow Corning silicon oils (200 fluid) were performed using each steady shear rheometer in order to ensure the instrument and the technique for using the instrument was accurate. Figure 3.5 shows the viscosity measured using Carri-Med CSL rheometer for three silicon oils. The viscosity of the silicon oils cover three orders of magnitude and span the range of viscosity for most of the fluids used in this work. The measurements indicate that there were minimum shear rates, which depend on the viscosity, below which the rheometer was inaccurate and above which the results were within 5% of the expected values. Therefore, the approximate shear rate required for accurate measurements using the Carri-Med CSL was above 0.1 s⁻¹, 1 s⁻¹ and 10 s⁻¹ for viscosities of around 1000 mPa·s, 100 mPa·s and 10 mPa·s respectively. The steady shear elastic properties for 12,500 cSt silicon oil are shown in figure 3.6. These measurements were made using the Weissenberg R19 and compared with previous experimental data provided by Hur (1987). Very good agreement was found with Hur’s results with differences of less than 5%, as shown in figure 3.6. Figure 3.6 also shows that it is possible to get normal stress differences as low as \( N_t = 1 \) Pa with a reasonable degree of accuracy.

Steady shear measurements were also obtained using cup-and-bob geometry courtesy of the rheology group at Nanyang Technological University, Singapore, headed by Dr. K.C. Tam, who performed experiments on several of the solutions used in the present work. The instrument used was a Contraves LS40, which is regarded as highly sensitive and very accurate for low viscosity solutions. The measurements were reliable at shear rates which were two orders of magnitude lower than those for the cone-and-plate geometries. Two measurement systems were used with specifications and dimensions as follows:

- DIN406: bob radius = 3.0 mm, cup radius = 3.25 mm, length = 9 mm, gap = 0.25 mm
- MS41S/1S: bob radius = 5.5 mm, cup radius = 6 mm, length = 8 mm, gap = 0.5 mm
FIGURE 3.5 - Viscosity measurements for silicon oils at 25°C using the Carri-Med CSL100 rheometer.
FIGURE 3.6 - Steady shear properties of 12,500 cSt silicon oil and comparison with those of Hur (1987).
3.4.2 Small-Amplitude Oscillatory Shear

Small-amplitude oscillatory measurements provide another means to examine the elasticity of a viscoelastic fluid using the cone and plate or cup-and-bob geometry shown in figure 3.4. Oscillatory tests involve the measurement of the response of the fluid to a small amplitude sinusoidal oscillation. In cone-and-plate flow, the strain applied to the plate and the strain rate are given by:

\[ \gamma(t) = \gamma^0 \sin(\omega t) \]  
\[ \dot{\gamma}(t) = \gamma^0 \omega \cos(\omega t) \]

where \( \gamma^0 \) is the amplitude of the applied strain and \( \omega \) is the frequency. The resulting shear stress may be given in terms of amplitude (\( \sigma^0 \)) and phase shift (\( \delta = (\frac{\pi}{2} - \phi) \)) as follows:

\[ \sigma_\omega(t) = \sigma^0 \sin(\omega t + \delta) \]  
\[ \sigma_\varphi(t) = \sigma^0 \omega \cos(\omega t - \phi) \]

These may be expanded and rewritten in terms of the in-phase and out-of-phase parts of the shear stress and placed in terms of viscoelastic material functions (\( G', G'', \eta', \eta'' \)) as follows:

\[ \sigma_\eta(t) = -\gamma^0 (G' \sin(\omega t) + G'' \cos(\omega t)) \]  
\[ \sigma_\varphi(t) = -\gamma^0 \omega (\eta' \cos(\omega t) + \eta'' \sin(\omega t)) \]

The storage modulus (\( G' \)) is defined as the stress in-phase with the strain in a sinusoidal shear deformation divided by the strain, and is a measure of the stored elastic energy. \( G' \) represents the solid-like response of a material, and for a perfectly elastic solid is equal to the constant shear modulus (\( G \)) for a perfectly elastic solid with the loss modulus equal to zero. The loss modulus (\( G'' \)) is defined as the stress \( 90^\circ \) out-of-phase with the strain divided by the strain, and is a measure of the energy dissipated during flow. \( G'' \) represents the viscous component or liquid-like response

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of a material to the deformation. The dynamic viscosity ($\eta'$) and dynamic rigidity ($\eta''$) are related to $G'$ and $G''$ as follows:

$$\eta' = \frac{G''}{\omega} \quad (3.21)$$

$$\eta'' = \frac{G'}{\omega} \quad (3.22)$$

The material functions, $G'$, $G''$, $\eta'$ and $\eta''$, are referred to as the linear viscoelastic properties, because they are determined from the shear stress which is linear in strain for small deformations. Thus, two more independent material functions are measured by utilising small-amplitude oscillatory shear: the storage modulus and loss modulus. It should also be noted that as the frequency approaches zero, $\eta'$ approaches $\eta_0$ and $2G'/\omega^2$ approaches $\Psi_{1,0}$.

The linear viscoelastic properties for 12,500 cSt silicon oil are presented in figure 3.7 and compared to data provided by Hur (1987). Close agreement to the measurements of Hur (1987) was found, except there were large differences for the storage modulus when $2G' < 0.3$ Pa at frequencies below about 1 rad/s.

The low viscosity solutions used in this study contain only small degrees of elasticity due to low polymer concentrations. Hence, any dynamic properties measured must be examined to ensure the limitations of the rheometer was not breached. The limits for the measurement of $G'$ using the Carri-Med CSL100 was undertaken by M.Y. Lee and S. Sexton in the rheology group at The University of Melbourne by measuring the linear viscoelastic properties of a large range of fluids with different viscosities (Lee & Sexton 1994). The results are shown in figure 3.8, which indicates that the same $G'$ was obtained for air, water and for 1-1000 cSt silicon oils. A ‘base-line’ has been drawn to represent the limiting curve and this is represented by: $G' = 0.0015a^2$. If the Maxwell constitutive model is applied to the base-line and assuming $\lambda_M \omega << 1$, then the Maxwell relaxation time would be given by $\lambda_M = 0.0015/\eta$. This would indicate that the relaxation time for water is 1.5s, which is clearly too large and
FIGURE 3.7 - Dynamic rheological properties for 12500 cSt silicon oil and comparison with those of Hur (1987). Also shown is the primary normal stress difference as a function of shear rate.
FIGURE 3.8 - Storage modulus for a range of fluids using the CarriMed CSL100 rheometer (Lee & Sexton 1994).
therefore incorrect. Therefore, the data near the base-line cannot be real and was produced purely as an artefact of the instrument. Any results in the vicinity of the base-line are therefore considered unreliable.

3.4.3 Elongational and ‘Shear Free’ Flow

The velocity field for uniaxial elongational, which is a shear free flow, is given in terms of the elongation rate ($\dot{\varepsilon}(t)$) as follows:

$$v_x = -\frac{1}{2} \dot{\varepsilon} x, v_y = -\frac{1}{2} \dot{\varepsilon} y, v_z = \dot{\varepsilon} z$$  \hspace{1cm} (3.23)

The stress tensor in elongational flow is defined by:

$$\sigma = \begin{pmatrix} \sigma_{xx} & 0 & 0 \\ 0 & \sigma_{yy} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}$$  \hspace{1cm} (3.24)

In uniaxial elongational flow, $\sigma_{xx} = \sigma_{yy}$ and the extensional viscosity ($\eta_e$) is given by the following normal stress difference:

$$\sigma_{xx} - \sigma_{yy} = -\eta_e(\dot{\varepsilon})\dot{\varepsilon}$$  \hspace{1cm} (3.25)

The extensional viscosity may also be represented as a Trouton ratio, which is defined by:

$$Tr = \frac{\eta_e}{\eta}$$  \hspace{1cm} (3.26)

The Trouton ratio has a value of $Tr = 3$ for a Newtonian solution but may be found to be extremely large for a viscoelastic fluid. The two rheometers which have been used to determine an extensional viscosity, the opposed jet apparatus and the filament stretching device, will now be discussed in turn.
THE OPPOSED JET APPARATUS:

The Rheometrics RFX opposed jet rheometer was used to examine the behaviour of viscoelastic solutions in an apparent elongational flow field, as shown in figure 3.9. The RFX device is used to measure an apparent extensional viscosity ($\eta^*$) as a function of strain rate. The term ‘apparent’ is used because the velocity field is not uniform across the nozzle face, such that the extensional rate is not constant with respect to radial position, and consequently it is unlikely that steady-state polymer conformations can be achieved. There is also insufficient strain and residence time in the flow for the molecule to fully extend. The conformation and residence times in the extensional flow field of the polymer chains will be heterogeneous across the nozzle face and if the chains are slow to orientate or stretch then the apparent extensional viscosity will be underestimated (Chai 1990). In the case of Newtonian fluids and rigid polymer chains (eg. xanthan gum), orientation is rapid such that a reasonable measure of the extensional viscosity is expected. However, large flexible molecules (eg. polyacrylamide) will not have enough time to fully orient and stretch, and therefore the extensional viscosity will be underestimated (Fuller et al. 1987; Tirtaatmadja & Srihda 1993).

The opposed jet instrument was first proposed by Fuller et al. (1987) and a schematic of the device is shown in figure 3.9. Fluid is drawn into opposed jets with the right nozzle arm fixed while the left nozzle arm is free to rotate about a pivot. The fluid exerts a hydrodynamic force onto the nozzles during flow, which is balanced by applying a torque ($T_m$) to the pivot arm to prevent movement of the left arm. The force ($F_R$) used to balance the hydrodynamic force is related to the fluid’s extensional viscosity and can be used to define an apparent extensional stress difference ($\sigma_e = \sigma_a - \sigma_n$) as follows:

$$\sigma_e = \frac{F_R}{A} = \frac{T_m}{AL}$$  \hspace{1cm} (3.27)
Figure 3.9 - Schematic of opposed jet apparatus (Rheometrics RFX) and a depiction of possible streamlines patterns in flow field.
$L$ is the length of the lever arm ($L = 7.62 \text{ cm}$) between the nozzle and transducer, $A$ is the area of the nozzle opening ($A = \pi R^2$) and $R$ is the nozzle radius. Assuming a uniform jet entrance velocity, the apparent extensional rate ($\dot{\varepsilon}$) in the flow field is defined in terms of the volumetric flow rate through a nozzle ($Q$) and the gap between the nozzles ($d_n$) as follows:

$$\dot{\varepsilon} = \frac{Q}{A d_n}$$

(3.28)

The apparent extensional viscosity ($\eta''$) can then be calculated according to:

$$\eta'' = \frac{\sigma}{\dot{\varepsilon}}$$

(3.29)

One problem with the opposed-jet apparatus is that an upturn in the extensional viscosity is measured at high rates of strain for Newtonian fluids of low viscosity ($\eta_e < 75 \text{ mPas}$) which Hermansky & Boger (1995) associated with fluid inertia. Hermansky & Boger (1995) developed a method to correct for the 'inertia' which relied on determining a coefficient, $a'(R)$, which was dependent only on jet size. The relationship between the measured and corrected Trouton ratio was given as follows:

$$\frac{\eta_e}{\eta_s} = \frac{\eta''}{\eta_s} - a'(R) \left[ \frac{1}{4 \pi LR^2} \right] \frac{\rho d_n^2 \dot{\varepsilon}}{\eta_s}$$

(3.30)

$\eta_e$ is the corrected extensional viscosity and $\eta_s$ is the shear viscosity. The parameter $a'(R)$ was found to be constant for a particular jet and was reported to be a factor of 100 too large in the original paper. The value for $a'(R)$ should read $7.08 \times 10^{-9}$, $26.4 \times 10^{-9}$, and $168 \times 10^{-9} \text{ m}^3$ for the 0.5 mm, 1 mm and 2 mm diameter nozzles. Contrary to the postulation of Hermansky & Boger (1995), the analyses of Schunk et al. (1990), Pasquali & Scriven (1996) and Donutala et al. (1998) have shown that inertia will actually act to decrease the extensional viscosity in the opposed-jet apparatus, however, they were unable to quantitatively account for the observed increase in extensional viscosity of Newtonian fluids. Therefore, although the correction proposed by Hermansky & Boger (1995) does not account for 'inertia', it
does provide a good means of distinguishing the difference between an elastic and inelastic material of low viscosity. This is particularly the case when the opposed nozzle apparatus is virtually the only instrument commercially available, at this time, which can determine if a very low viscosity fluid is elastic or inelastic.

The Trouton ratio is shown for two Newtonian fluids in figure 3.10. The measured and corrected Trouton ratio for 60% glycerol, which has a shear viscosity of 11.7 mPas, is presented for three jet sizes. The uncorrected Trouton ratio for 60% glycerol is $Tr \approx 3$ at low strain rates but increases dramatically at moderate to high levels of strain rate and hence requires correction to obtain somewhat meaningful results. The corrected Trouton ratios for the 0.5 mm and 1 mm jets are reasonably constant but when the correction factor for the 2 mm jet is applied, the Trouton ratio is overcorrected and becomes negative. In figure 3.10, the correction factor for the 2 mm jet was halved (ie. $a'(R) = 84 \times 10^{-9}$ m$^3$) such that when applied the corrected Trouton ratio was relatively constant. Although the correction coefficients of Hermansky & Boger (1995) should be applicable for all Newtonian fluids, they were only tested on relatively few samples and should not be applied without caution. Before the correction is applied to an elastic fluid, it is always recommended that it be applied to a Newtonian fluid with a similar shear viscosity to the elastic fluid, to ensure accuracy. The corrections should only be used when the rise in Trouton ratio for a Newtonian fluid is excessive, as for 60% glycerol. The measured Trouton ratio for a slightly more viscous Newtonian fluid, 76% glycerol, is also presented in figure 3.10 and the rise in Trouton ratio is negligible below extreme values of extension rate (10,000 s$^{-1}$). Therefore, corrections are not required for fluids with a shear viscosity greater than about 40 mPas.

**FILAMENT STRETCHING DEVICE:**

The filament stretching device for determining the steady uniaxial extensional viscosity of polymer solutions was first developed by Tirtaatmadja & Sridhar (1993). A depiction of the instrument is shown in figure 3.11, where the fluid sample is held between two disks which move apart at an increasing rate so that the extension
FIGURE 3.10 - Measured and corrected Trouton ratio for a 60% glycerol Newtonian solvent (η= 11.7 mPa s) using the opposed jet apparatus. Also shown is the measured Trouton ratio for 76% glycerol (η=39.5 mPa s) using the 1 mm diameter jet.
Figure 3.11 - Depiction of the filament stretching device used for uniaxial extensional viscosity measurements
rate along the filament midpoint is held constant. The instrument has the advantage over the opposed jet apparatus, as well as many other devices available for measuring the extensional viscosity, by producing a flow field which is a very close approximation to pure uniaxial elongation. Previous experimental measurements for the steady-state and transient extensional viscosity have compared very closely to predicted values using a variety of constitutive models (Tirtaatmadja & Sridhar 1995; Orr & Sridhar 1996; Spiegelberg & McKinley 1996).

Measurements of the extensional viscosity using the filament stretch device were performed on several elastic fluids used in this study. The measurements were performed by Dr. Duc At Nguyen and Prof. Tam Srihdar at Monash University, Victoria, Australia.

3.4.4 Rheological Models and Material Functions

The purpose of measuring the rheological properties of materials is to provide parameters which will enable a description of the fluid’s behaviour when the fluid is flowing. A constitutive equation is required to describe the stress tensor in the equations of motion. The simplest constitutive equation is Newton’s law of viscosity, which is used to produce the Navier-Stokes equations in order to predict the flow of, what are now known as, Newtonian fluids. Fluids which do not behave according to Newton, are simply called ‘non-Newtonian’ fluids, and their rheological behaviour is much more complicated. Numerous constitutive equations have been proposed to describe various classes of non-Newtonian fluids and a few of the simplest, and therefore the more popular models, are described in appendix A. The books by Bird et al (1987) and Larson (1988) are recommended for more in depth discussion on constitutive models. The following section will briefly outline the main material functions utilised in describing the fluids used in this thesis and their relationship within a few simple constitutive models. Notation used in the following section is described in appendix A.
The simplest constitutive equation used to describe a fluid is Newton’s law of viscosity with the stress tensor ($\sigma$) defined by:

$$\sigma = -\mu \dot{\gamma}$$  \hspace{1cm} (3.31)

$\mu$ is the Newtonian viscosity (constant) and $\dot{\gamma}$ is the rate-of-strain tensor. Newton’s law describes any inelastic time independent incompressible fluid where there is a linear relationship between the stress and the shear rate with these fluids, subsequently labelled Newtonian fluids.

When there is not a linear relationship between shear stress and shear rate, the stress tensor may be altered by incorporating the concept of a non-Newtonian viscosity to obtain the *generalised Newtonian model*, which is defined:

$$\sigma = -\eta(\dot{\gamma}) \dot{\gamma}$$  \hspace{1cm} (3.32)

$\eta(\dot{\gamma})$ is the non-Newtonian viscosity as a function of the shear rate. The generalised Newtonian model can only represent the change in viscosity with shear rate for a non-Newtonian fluid, and does not include any reference to elastic properties. It is therefore only of use for solutions which are relatively inelastic. Several models have been introduced to empirically represent the non-Newtonian viscosity function. The simplest generalised Newtonian model is the *power law* model which describes the non-Newtonian viscosity as follows:

$$\eta = K \dot{\gamma}^{n-1}$$  \hspace{1cm} (3.33)

$K$ is referred to as the consistency index and $n$ is the power law exponent. The power law exponent may be used as a measure of the variability of the viscosity with shear rate with the value of $n = 1$ indicating a constant viscosity. When $n < 1$, the fluid is shear thinning, and when $n > 1$, the fluid is shear thickening. A majority of the fluids used in this study are Boger fluids which have a constant viscosity such that the power law index is close to unity. Deviations from unity will be expressed in terms
of the power law exponent and when the exponent is above 0.9, the fluid will be considered to have a constant viscosity.

A continuum mechanics approach has been used to develop two useful constitutive models to account for fluid elasticity. The first is the Maxwell model, which is based on a theory of viscoelasticity where the fluid is considered as both viscous and elastic by utilising both Newton’s law of viscosity and Hooke’s law for an elastic solid. The Maxwell equation is given by:

\[ \sigma + \lambda_M \frac{\partial}{\partial t} \sigma = -\eta_0 \dot{\gamma} \]  

(3.34)

\( \lambda_M \) is the Maxwell relaxation time and \( \eta_0 \) is a constant-viscosity. The equation may be converted into an objective relationship by replacing the time derivative with the convected time derivative to obtain the upper-convected Maxwell model, which is given in appendix A. The Maxwell relaxation time is determined by substituting the upper-convected Maxwell model into the equations for steady shear flow:

\[ \lambda_M = \frac{N_i}{2\eta_0 \dot{\gamma}^2} = \frac{\Psi_i}{2\eta_0} \]  

(3.35)

When the primary normal stress difference is quadratic with shear rate and the viscosity constant, a constant relaxation time results. This relationship will be used extensively in this thesis to calculate the characteristic relaxation time of a polymer solution. The Maxwell relaxation time will be subsequently used in the evaluation of the dimensionless numbers governing the fluid mechanics in confined swirling flow experiments, i.e. the Weissenberg number and elasticity number.

The upper-convected Maxwell model may be transformed to include an additional time derivative of \( \dot{\gamma} \) to obtain the more realistic Oldroyd-B constitutive model. In non-convected form, the Oldroyd-B model is referred to as the Jeffrey’s model and given by:
\[ \sigma + \lambda_1 \frac{\partial}{\partial t} \sigma = -\eta_0 (\dot{\gamma} + \lambda_2 \frac{\partial}{\partial t} \dot{\gamma}) \]  

(3.36)

\( \lambda_1 \) is the relaxation time and \( \lambda_2 \) is the retardation time. The relaxation times are determined in the same manner as for the upper-convected Maxwell model, with the resulting material functions for steady shear flow of an Oldroyd-B fluid being:

\[ \eta = \eta_0, \ \Psi_1 = 2\eta_0 (\lambda_1 - \lambda_2) \text{ and } \Psi_2 = 0 \]  

(3.37)

Therefore, the Oldroyd-B model predicts a constant viscosity and constant first normal stress coefficient, while the second normal stress coefficient is zero. These properties are observed for flexible polymer Boger fluids at low deformation rates. The retardation time is also calculated in terms of the relaxation time and is given by:

\[ \lambda_2 = \frac{\eta_0}{\eta_0} \lambda_1 \]  

(3.38)

The high viscosity dilute flexible polymer Boger fluids produced in this thesis have been designed such that at low shear rates, the normal stress difference is a quadratic function of shear rate. Therefore, the primary normal stress coefficient will reach a plateau at low shear rates, which allows the upper-convected Maxwell and Oldroyd-B models to be obeyed in steady shear and the appropriate constant relaxation times to be determined. The linear viscoelastic properties for both the upper-connected Maxwell and Oldroyd-B models may be determined by substituting the stress tensor into the equations governing small-amplitude oscillatory shear. Both models predict that at low frequencies the storage modulus varies quadratically with frequency while at high frequencies a constant value is obtained. The equations for the uniaxial extensional viscosity may also be calculated in the same fashion except using the equations governing shear-free flow. Both models predict that the steady-state extensional viscosity increases with strain rate and asymptotes to infinity as the strain rate approaches the reciprocal of twice the relaxation time.
The Oldroyd-B model is also derived by idealising a polymer molecule as an elastic dumbbell. The elastic dumbbell model represents the molecule as two beads connected by a non-bendable Hookean spring. The relaxation time for the elastic dumbbell model may be determined using kinetic theory, which incorporates the intrinsic viscosity. Therefore, the Oldroyd-B relaxation time is determined from the intrinsic viscosity and defined by:

\[ \lambda_1 = \frac{[\eta] \eta M}{RT} \]  

(3.39)

This equation will be used to determine model parameters for low viscosity dilute flexible polymer solutions when the primary normal stress is too low to measure. The Maxwell relaxation time may be determined by noting above that \( \lambda_M = \lambda_1 - \lambda_2 \).

The Maxwell and Oldroyd-B models are really only suitable for flexible polymer solutions and apply to polyacrylamide and polyethyleneoxide. The other polymer used in this work is xanthan gum which is regarded as a rigid or semi-rigid polymer. The rigid dumbbell model is the simplest mechanical model for the description of dilute solutions of rod-like macromolecules. It accounts for the orientability of the polymer molecules in the flow field while ignoring molecular stretching and bending motions, which are not considered significant for this class of macromolecules. The macromolecule is represented by two point masses or beads joined by a massless rod, with the solvent presumed to only interact at the beads. The model is described in appendix A with the rigid dumbbell relaxation time given by:

\[ \lambda_D = \frac{m[\eta] \eta_r M}{RT}, \quad m = (m_1 + m_2)^{-1} \]  

(3.40)

For a finite concentration the relaxation time is equal to:

\[ \lambda_D = \frac{m(\eta - \eta_r) M}{cRT} \]  

(3.41)

The values of \( m_1 \) and \( m_2 \) depend on the details of the model, as listed by Ferry (1980). In this thesis, the parameters for the rigid dumbbell model are \( m_1 = 3/5 \) and \( m_2 = 2/5 \),
as given by Bird et al. (1987b). The steady shear and linear viscoelastic properties may be determined for the appropriate rigid dumbbell model in the same fashion as was stated for the Maxwell and Oldroyd-B models. The extensional viscosity approaches a constant at relatively low extension rates such that as \( \dot{e} \to \infty \), the limiting extensional viscosity is:

\[
\eta_e = 3\eta_0 + 6ckT\lambda_0
\]  

(3.42)

The extensional viscosity predicted using (3.42) will be compared to measured values in the next chapter. The behaviour of the semi-rigid polymer to act in a similar fashion to a high aspect ratio rigid rod will also be examined by comparing the extensional viscosity to Batchelor’s theory for a semi-dilute suspension of perfectly aligned rigid rods, as defined by (Batchelor 1970, 1971):

\[
\eta_e = 3\eta_0 \left(1 + \frac{4\phi a^2}{9\ln(\pi / \phi)}\right)
\]  

(3.43)

\( \phi \) is the volume fraction of rods.

More complicated constitutive models were also used to describe the rheology of the non-Newtonian fluids used in this thesis. The material properties for the polyacrylamide Boger fluids were predicted using the following constitutive models: Oldroyd-B (Oldroyd 1950), FENE-P (Warner 1972; Peterlin 1966; Bird, Dotson & Johnson 1980), Giesekus (Giesekus 1982, 1983; Bird & Weist 1985) and KBKZ (Kaye 1962; Berstein, Kearsley & Zapas 1963; Papanastasiou, Sriven & Macosko 1983). Details of these models are found in appendix A and throughout the literature (Bird et al. 1987ab; Byars, Binnington & Boger 1997; Tirtaatmadja & Sridhar 1995; Tirtaatmadja 1993).

Both single and multiple relaxation modes were used to describe the polyacrylamide fluid rheology. In the limit of small deformations, all the models mentioned reduce to the Maxwell model (Bird et al. 1987). For multi-mode models, this is given by:
\[ \sigma_i + \lambda_i \frac{\partial \sigma_i}{\partial t} = -\eta \dot{\gamma} \]  

(3.44)

\( \sigma_i \) is the shear stress corresponding to the \( i \) th relaxation mode, such that the polymer contribution to the overall shear stress for \( k \) relaxation modes is given by: \( \sigma_p = \sum_{i=1}^{k} \sigma_i \).

The linear viscoelastic spectrum \( \{ \eta_b, \lambda_i \} \) are obtained from small amplitude oscillatory measurements.

Apart from the Oldroyd-B model, all the aforementioned models contain adjustable parameters which are determined from the measurements of the primary normal stress difference and/or the extensional viscosity. The FENE-P model, where each polymer macromolecule is treated as a dumbbell connected by a non-linear finitely extensible spring, contains the parameter \( b_n \), as defined in Bird et al. (1987b). This parameter is indicative of the degree of flexibility of the polymer molecules, such that as \( b \rightarrow \infty \), the model approaches the Oldroyd-B model. The Giesekus model was derived from a molecular model of Hookean dumbbells with a non-linear stress term related to anisotropic drag and incorporates an adjustable 'mobility factor', \( \alpha_i \). The mobility factor is restricted to \( 0 \leq \alpha_i \leq 0.5 \) for realistic predictions and reduces to the Oldroyd-B model when \( \alpha_i = 0 \). The Giesekus model predicts a uniaxial extensional viscosity which approaches \( \eta_e = 2\eta_0/\alpha_i \) at high strain rates and a second normal stress coefficient of \( \Psi_{2,0} = -(\alpha_i/2) \Psi_{1,0} \), where \( \Psi_{1,0} \) and \( \Psi_{2,0} \) are the two limiting values of the normal stress coefficients. The KBKZ model is an integral type constitutive equation and contains two sets of adjustable parameters, \( \alpha_i \) and \( \beta_i \), which are determined from the shear and extensional properties respectively. The adjustable parameters for all models were determined for each relaxation mode. The parameters for a single mode were also determined, however it should be noted that a single relaxation mode could not describe the dynamic properties except at low frequencies.

The preceding section has detailed the methods for determining material functions, which are used to describe and characterise the behaviour of non-Newtonian fluids.
The measured material functions may be used in constitutive models to predict the behaviour of a particular fluid in flow, by substituting the appropriate stress tensor into the equations of motion. A full comparison between a few selected models and experimental rheometrical data will be presented in the next chapter, such that the limitations of the models can be established. The wealth of experimental rheological data combined with a comparison of selected models will enable a reasonable choice to be made of the most appropriate constitutive equation, and their appropriate material parameters, for anyone wishing to predict the observation of the behaviour of the Boger fluids used in the torsionally driven cavity described in chapters 5 and 6.

3.5 CONFINED SWIRLING FLOW APPARATUS

The following section details the confined swirling flow apparatus, which is an enclosed torsionally driven cavity. The flow parameters which govern the geometry for Newtonian and viscoelastic fluids will also be discussed.

3.5.1 The Torsionally Driven Cavity

The experimental apparatus, as shown in figure 3.12 and also described by Day et al. (1996), consists of an acrylic cylinder with radius $70 \pm 0.25$ mm, situated in a rectangular acrylic water bath, with dimensions $402 \times 402 \times 592$ mm, to reduce image distortion effects. The bottom lid of the cylinder was a stainless steel disk which was rotated using a 3-phase AC motor via a v-belt and pulley arrangement with a selectable reduction gearbox for the lower disk speed range. The rotation rate of the disk was controlled using a variable frequency unit connected to the AC motor and was measured using a frequency counter with a resolution of approximately $\pm 0.002s^{-1}$. The stationary top lid was movable and lockable and positioned according to the height to radius ratio required to within $\pm 0.5$ mm. Both disk and cylinder were designed and built to ensure axial symmetry with the disk rotating with a lateral tolerance of $\pm 50$ $\mu$m.
FIGURE 3.12 - Torsionally driven cavity apparatus.
Various top lids were used depending on the viscosity of the fluid, such that air bubble entrainment was minimised as the lids were lowered into the fluid and set in place. A lid comprising of one central small capillary hole (0.5 mm diameter), connected to a needle and a 1 mm diameter tube for dye insertion, was used for fluids with a low-viscosity ($\eta < 1.5$ Pa s) and also contained a flush mounted thermocouple. An alternative lid comprising of 5 small holes (1 mm diameter) arranged regularly in a line across the lid surface was used for medium-viscosity fluids ($1.5 < \eta < 3$ Pa s). Also, a third lid comprising of a central small hole (1 mm diameter) and an off centre large diameter hole ($\approx 10$ mm diameter) was used for high-viscosity fluids ($\eta > 3$ Pa s), such that once the lid was lowered into the fluid, a large flat plug screw could be used to block the hole. Subsequent flow measurements showed that no detectable asymmetries in the flow field were present for any of the three different lids.

During a given experiment, the temperature of the working fluid was found to increase due to viscous heating, especially at high rotation rates and for viscous liquids. In order to control heating effects, the bath water was circulated through an Haake F3 environmental controller such that most experiments were conducted between 20 and 21 °C. For the lids without a flush mounted thermocouple, a thermocouple probe was inserted through the capillary holes in the lids, or alternatively, for continual measurement the thermocouple could be placed in the capillary holes just outside the flow cell. The temperature of the test fluid was regularly measured to within ± 0.05 °C, such that the governing dimensionless numbers could be determined accurately by taking into consideration temperature variation of the material functions.

### 3.5.2 Dimensional Analysis

The characteristic parameters used in the dimensional analysis of the torsionally driven cavity are as follows: characteristic time of the process = $1/2\pi\Omega t$, characteristic length = $R$; and the characteristic velocity = $2\pi\Omega R$. The Reynolds number, which is the ratio of inertial and viscous forces, is defined as follows:
\[
Re = \frac{\rho(2\pi\Omega)R^2}{\eta}
\]  
(3.45)

\(\rho\) is the density (kg/m\(^3\)), \(\omega\) is the disk rotation rate (s\(^{-1}\)), \(R\) is the disk radius (m), \(H\) is the cylinder height (m) and \(\eta\) is the viscosity (Pa s).

Elasticity is typically represented by a Weissenberg number (\(We\)), or similarly the Deborah number (\(De\)), which are measured by evaluating of the ratio of the characteristic time of the fluid (eg. Maxwell relaxation time: \(\lambda_m\)) and characteristic time of the process (eg. 1/2\(\pi\omega\)). The Weissenberg number is evaluated as follows:

\[
We = \lambda_m 2\pi\omega
\]  
(3.46)

Another dimensionless number which will be used is the elasticity number (\(El\)) which is the ratio of the Weissenberg number and the Reynolds number and may be represented as follows:

\[
El = \frac{We}{Re} = \frac{\lambda_m \eta_0}{\rho R^2}
\]  
(3.47)

The elasticity number gives the ratio of the elastic forces to inertial forces due to the rotation of the disk. The inertial forces on an element of fluid are given by the centrifugal force as \((\rho v_\theta^2 / R)dV\), while the elastic forces are determined from the first normal stress difference as \((N_1)dA\). Therefore, using the definition for the Maxwell relaxation time in equation (3.35), and assuming \(\dot{\gamma} = \Omega\) and \(v_\theta \approx \Omega R\) near the disk, then the above expression for the elasticity number is obtained. A feature of the Elasticity number is that it is independent of the rotation rate of the lid, provided the relaxation time and viscosity are constant and not shear rate dependent. It can therefore be used as a material parameter and is used to compare the level of elasticity between different fluids.
3.6 VISUALISATION TECHNIQUES

Two techniques which were used to visualise the secondary flow field in the torsionally driven cavity were laser induced fluorescence and particle image velocimetry (PIV). Laser induced fluorescence is used to capture the ‘streak-lines’ in the secondary flow plane in order to obtain a qualitative indication of the flow kinematics. PIV is used to measure the axial and radial velocity distribution in the secondary flow plane. These two techniques are described as follows and the errors associated with PIV are also discussed.

3.6.1 Laser Induced Fluorescence

Illumination of the secondary flow plane was performed using a Coherent Highlight argon-ion laser, operating at 0.5W, piped through an optical fibre to a cylindrical lens. This lens produced a blue-green laser light sheet with a thickness of 1-2 mm. Dye flow visualisation was conducted, in order to observe ‘streaklines’, by dissolving fluorescein powder (≈ 0.2 g/L) into a small quantity of the test fluid. The dye was then added via either a syringe-tube-needle arrangement in the centre of the stationary lid used for low-viscosity fluids, or by a syringe-tube-capillary arrangement in the other lids.

Colour photography of the dye streaklines was typically performed at an exposure of 1/4 - 1/2 s and aperture f2 to f4, using a 35 mm SLR camera with a noct-Nikkor 58 mm lens and 1BUV filter with EPP 100 ISO film. Images were slightly distorted in the radial direction such that the equivalent radial image distance is equal to 108% of the actual distance. Video imaging was performed using a Sony Hi8 video camera (model no. DXC537P) with zoom lens.

3.6.2 Particle Image Velocimetry

Velocity data in the secondary flow plane was obtained using the two-dimensional optical technique of particle image velocimetry (PIV) (Pickering & Halliwell 1985; Adrian 1991). A similar technique, laser speckle velocimetry (LSV), has also been
used previously by Binnington, Troup & Boger (1983) to obtain velocity profiles for
Boger fluids. PIV was used in preference to the alternative measurement technique
Laser Doppler Anemometry (LDA) (Durst, Lehmann & Tropea 1981) due to the
prohibitively long acquisition periods that would be required for LDA when
measuring the higher viscosity flows, where fluid velocities were less than 1 mm/s.

The remainder of this section will describe the PIV system, which was based on
simple multiple exposure photography for imaging and digital autocorrelation
techniques for data processing (Adrian 1991; Meinhart, Prasad & Adrian 1993).

To record the PIV images, a pulsed light source was provided by either a 12 sided
rotating mirror system as described by Gray et al. (1991) and shown in figure 3.13, or
a mechanical shutter with light sheet optics. In both cases, a 0.5W argon ion laser
light source was used with a fibre optic delivery and collimation lens. Due to the
greater illumination power of the rotating mirror system, this was generally used for
the higher flow speeds when $25 < Re < 4000$. These flow ranges required
corresponding mirror speeds of 35 - 500 rpm to provide pulse separations of 10 - 143
ms, and exposure periods of 1/8 - 1 s to ensure a minimum of five exposures on any
given image. Below a mirror speed of 35 rpm, unacceptable errors occurred with the
mirror velocity. Lower flow speeds were then measured with either a mechanical
shutter system or by ‘blacking’ out 6 to 10 sides of the rotating mirror. In this case
exposure times ranged between 0.05 - 60 s to provide the required number of pulses
per image. A Nikon F4 camera with a noct-Nikkor 58mm f1.2 lens was used to
record the images onto 35mm Kodak Tmax400 (TMY) film. The film was
subsequently developed with Ilford Microphen developer at 23°C for 11 minutes to
provide the required resolution. The 35mm transparencies were then digitised into
3500 x 2500 pixel 8 bit grayscale images at a resolution of 2700 dpi by using a
Polaroid SprintScan 35 scanner. The flow was seeded using fluorescent rhodamine
particles with diameter of 10-80 μm (30 μm average) in conjunction with a HOYA O
[G] orange camera filter in a method similar to Northrup, Kulp & Angel (1991). This
technique improved image contrast by reducing flare off the perspex walls of the rig.
FIGURE 3.13 - Torsionally driven cavity experimental set-up showing the rotating mirror used for PIV studies.
The processing of images was carried out using autocorrelation and post-processing software developed by Dr. Nick Lawson at the University of Melbourne. Grid size ranged from a $40 \times 20$ grid used for a $H/R = 1$ to a $40 \times 50$ grid for a $H/R = 2.5$. For a majority of the processed images, an interrogation window resolution of $128 \times 128$ pixels was chosen for optimum accuracy (Prasad et al. 1992). The software was set up to vary the size of the dc peak mask in the correlation plane so that it was proportional to the particle image diameter (Lawson, Coupland & Halliwell 1997). These results were subsequently validated using an absolute measurement range of 5-25% of the interrogation region length as recommended by Keane & Adrian (1991). Vector direction was determined using a-priori knowledge of the flow and any missing data points were interpolated and smoothed using algorithms described by Landreth & Adrian (1988). In this case a smoothing radius of three interrogation regions was chosen to match the spatial scales in the majority of the flow.

Figure 3.14 shows a typical PIV image with corresponding vector map and streamline plot. The two-dimensional PIV vector maps are measurements of velocity in the axial and radial plane. From this data, a predictor-corrector integration algorithm in the commercial software package of ‘Tecplot’ was used to obtain streamline traces. It should be noted, however, that due to the three-dimensional nature of the flow field, the streamlines in the following analysis are representative of the instantaneous flow in the cross plane and are termed ‘sectional streamline patterns’ (Perry & Steiner 1987).

As mentioned previously, the flow field is highly three-dimensional and contains a strong out-of-plane component, termed the azimuthal velocity ($v_\phi$). In the worst case, this component will displace particles out of the light sheet between exposures causing data dropout in the vector map. Therefore, areas near the outer surfaces and in particular near the rotating disk, were found to have the greatest dropout and only the central region near the axis of symmetry contained reliable data, where the out of plane velocity component was lower. This problem was partly overcome by recording several PIV images with different pulse separations and then combining the
FIGURE 3.14 - Secondary flow field for 75 ppm polyacrylamide low-viscosity Boger fluid at $Re = 2100$, $We = 0.7$ and $H/R = 2$ showing (a) PIV image, (b) vector map, and (c) sectional streamline patterns.
sets of validated data. However, in the case where the secondary flow velocity was the same order of magnitude or greater than the azimuthal velocity component, the majority of PIV vectors were obtained. Other errors were also generated around the axis of symmetry when particles were displaced across the centre with the primary flow due to either an excessive laser sheet thickness, high velocities in the primary flow and/or misalignment of the laser sheet position.

The measured radial and axial velocities, given the symbols \( v_r \) and \( v_z \) respectively, will be made dimensionless for comparative purposes by dividing by a characteristic velocity. The characteristic velocity is chosen to be the maximum azimuthal velocity \( (v_\text{max})_{\text{azimuthal}} = 2\pi \Omega R \) in the system, which is located at the edge of the rotating disk. The co-ordinate system is taken as \( (r, \theta, z) \) with \( r = 0 \) and \( z = 0 \) corresponding to the centre of the rotating disk. Therefore, a positive axial velocity corresponds to the fluid flowing upwards vertically away from the rotating disk. The azimuthal component of vorticity \( (\omega_\theta) \) was determined from the measured velocity data using the following definition:

\[
\omega_\theta = \frac{\partial v_r}{\partial z} - \frac{\partial v_z}{\partial r}
\]  

(3.48)

Components of the rate-of-strain tensor which could be determined are \( \dot{\gamma}_r, \dot{\gamma}_\theta, \dot{\gamma}_z, \dot{\gamma}_a, \dot{\gamma}_\rho \). These were determined from the following definition for the rate-of-strain tensor from Bird et al. (1987a):

\[
\dot{\gamma} = \nabla \mathbf{v} + (\nabla \mathbf{v})^T
\]  

(3.49)

3.6.3 PIV Error Analysis

For the PIV system, the velocity vector, \( \mathbf{V} \), at any grid point is calculated from a calibrated magnification, \( M \), a particle image displacement, \( \Delta x \), and a pulse separation, \( \Delta t \), such that:
\[ \mathbf{V} = \frac{\Delta s}{M \Delta t} \]  

(3.50)

Therefore, if the uncertainty in measurement of the quantities \( M, \Delta s \) and \( \Delta t \) is represented by the percentage errors \( \delta(M) \), \( \delta(\Delta s) \) and \( \delta(\Delta t) \) respectively, the total percentage error in velocity measurement, \( \delta(V) \), can be found from:

\[ \delta(V) = \sqrt{[\delta(M)]^2 + [\delta(\Delta s)]^2 + [\delta(\Delta t)]^2} \]  

(3.51)

The magnification \( M \) (pixels/mm) was estimated from known image dimensions such as the height and diameter of the flow cell and the error was typically one pixel in 1000 or \( \delta(M) = 0.1\% \). The maximum deviation in the mirror speed was noted as 3 rpm in 167 rpm, resulting in a maximum pulse separation error of \( \delta(\Delta t) = 1.8\% \). For the error in particle image displacement, \( \delta(\Delta s) \), previous work by Keane & Adrian (1991) will be used to estimate the error from \textit{a-priori} information on the flow.

\textit{A-priori} velocity information is required since the particle displacement error is strongly dependent on spatial velocity gradients (Keane and Adrian, 1991) with the worst error occurring in the region of highest spatial gradient. From previous work (Lugt & Abboud, 1987), the maximum gradient has been predicted at the edge and towards the centreline of the flow, and is of the order of 10 mm/s across a given interrogation region of size 1.3 mm. Therefore, with a mean particle image size of 100 \( \mu \)m and a magnification of \( M = 0.15 \), the error in particle displacement is estimated to be in the range \( 2.5\% < \delta(\Delta s) < 19.5\% \) for a corresponding range of pulse separations \( 15 \text{ms} < \Delta t < 143 \text{ms} \). The total error in measurement from equation (3.51) will then equal \( 3.1\% < \delta(V) < 19.6\% \). This estimate indicates that it is desirable to keep the laser pulse separation to a minimum due to the errors generated by velocity gradients. Unfortunately, at lower pulse separations, the lower range of secondary flow velocities cannot be resolved due to insufficient particle image separations. Hence, a number of PIV vector maps were taken for a given flow with different pulse separations, and the different sets of validated vectors combined. This technique allows the user to restrict the total error in measurement to \( \delta(V) < 10.0\% \) and also
permit tuning of the data to account for the out of plane effects mentioned previously. Any remaining non valid vectors are then interpolated and the complete map smoothed to remove correlation noise.

A comparison is shown in figure 3.15 between PIV experimental measurements of the centreline axial velocity and those predicted using the numerical model of Lught & Abboud (1987) for a Newtonian fluid at Reynolds numbers of $Re \approx 1000$ and $Re \approx 1500$. Each plot shows axial velocities which were determined using a range of pulse separation times of between 15 ms and 143 ms. The experimental results compare very well to the predicted measurements across the entire length of the cylinder. Deviations of around 10% between the experimental measurements and those predicted were found at the minimum peak in axial velocity, where the velocity gradients were highest. This deviation matches the level of accuracy predicted in the previous error analysis and thus gives a sufficient degree of confidence in the technique for following study of centreline flow fields.
FIGURE 3.15 - Comparison of the axial velocity along the centre line (r ~ 0) measured using PIV with that predicted by Lught & Abboud (1987) for a Newtonian fluid at (a) $Re = 1000$, and (b) $Re = 1500$. 
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