NONLINEAR DYNAMICS OF VISCOELASTIC FLOWS IN COMPLEX GEOMETRIES

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Abstract

The nonlinear dynamics of a viscoelastic fluid are investigated experimentally in three model problems: (i) rotational flow between coaxial parallel disks; (ii) entry flow through an abrupt axisymmetric contraction, and (iii) stagnation flow past a circular cylinder confined in a rectangular channel. These test geometries serve as prototypes of the complex flows typically encountered in commercially important processing applications such as spin-coating, fiber-spinning and injection-molding.

Direct visualization of the spatial and temporal structure in each flow is performed with a high resolution digital video camera. Three-color Laser Doppler Velocimetry (LDV) is used to provide spatially-resolved measurements of the individual velocity components throughout each flow cell. The LDV system can measure steady and time-dependent velocities in the range $0.04 \leq \nu \leq 100 \text{ cm/s}$ with a spatial resolution of approximately 50 $\mu$m. The two optical techniques are non-perturbative to the flow and permit both qualitative visual confirmation and quantitative determination of the nonlinear dynamics associated with complex flows of elastic liquids.

The viscoelastic test fluid used in these studies is a highly elastic 'Boger fluid' composed of 0.31 wt% polyisobutylene dissolved in a well-mixed, highly-viscous solvent of 4.86 wt% tetradecane (C14) and 94.83 wt% polybutene (PB). The rheological properties of the fluid have been characterized over a range of temperatures in steady-shear, transient-shear and small-amplitude oscillatory-shear flows. The fluid is highly elastic with a characteristic relaxation time of $\lambda = 0.80$ second, and has an almost constant viscosity across four decades of shear-rate. It is thus possible to study elastic effects in the flow without additional complicating factors such as shear-thinning or fluid inertia.

The relative importance of elastic and viscous effects in each flow are characterized by the Deborah number, which is defined as a ratio of the polymer relaxation time $\lambda$ to a characteristic residence time $\mathcal{I}$ in the flow geometry. This residence time is calculated as $\mathcal{I} = L/\nu$, where $\langle \nu \rangle$ is the average velocity through the test cell, and $L$ is a characteristic length scale in the flow. For the three geometries discussed above, this characteristic length scale is determined to be (i) the disk radius $R$; (ii) the radius of the small downstream tube $R_2$, and (iii) the radius of the cylinder $R_c$, respectively. As the flow rate, and Deborah number, of the fluid in each test geometry is increased, viscoelasticity drives hydrodynamic instabilities that lead to transitions from the steady two-dimensional base flow to more complex three-dimensional or time-dependent fluid motions. The major findings of each set of experiments are summarized below.
Dynamic measurements of the torque and normal force exerted on the plates of standard cone-and-plate and parallel plate test fixtures identify a subcritical Hopf bifurcation from steady, axisymmetric rotational shear flow to a complex three-dimensional, time-dependent motion. Stability measurements for a wide range of shear rates, rotational speeds and disk sizes are used to construct a stability diagram for the rotational flow of Boger fluids which shows that the instability is a sensitive function of the rotation rate $\Omega$ and not of the shear rate $\dot{\gamma}$. Flow visualization shows that the resulting unsteady flow consists of recirculating spiral vortices with a wide range of characteristic wavelengths that propagate radially across the disks.

Laser Doppler velocimetry measurements of the velocity field near the re-entrant lip of the axisymmetric abrupt contraction document a series of nonlinear transitions, commencing with a supercritical Hopf bifurcation to three-dimensional time-dependent flow, followed by subsequent transitions to period-doubling, quasiperiodic and ultimately aperiodic flow regimes. Measurements for six different contraction ratios and two different lip configurations are performed to provide a comprehensive and systematic exploration of the large variations in dynamics associated with small changes in flow geometry. By coupling qualitative video-imaging techniques with the highly accurate LDV measurements the local dynamic behavior of the fluid near the contraction lip is linked with the evolution of the vortex structure that is macroscopically observed in axisymmetric contraction flows.

Direct video-imaging and LDV measurements also document for the first time the onset of an elastic instability in the wake of a circular cylinder confined in a rectangular channel. The transition results in a bifurcation from steady, planar extensional flow in the wake and the formation of a spatially periodic, three-dimensional flow that extends along the symmetry axis of the cylinder. Flow visualization and LDV measurements show that this three-dimensional structure remains steady in time for Deborah numbers close to the critical value before undergoing a second transition to a time-dependent state at higher flow rates. The spatial extent of this cellular structure is limited to the strongly extensional flow near the downstream stagnation point and measurements for different cylinder sizes demonstrate that the onset point and the wavelength of the periodic structure scales more closely with the cylinder radius than with the gap between the cylinder and the channel walls.

The onset of the flow transition in each geometry is found to occur at moderate Deborah numbers of $De \sim 1$ and very small Reynolds numbers of $Re < 0.1$. These transitions are entirely absent in the corresponding flows of Newtonian liquids at equivalent $Re$ and the onset of these instabilities is associated unequivocally with the elastic properties of the polymer solution. The dynamic behavior documented in each of these systems is found to be extremely sensitive to relevant dimensionless geometric parameters such as the aspect ratio of the parallel plates, the contraction ratio, and the cylinder-channel ratio. In each case it is shown that this complex dependence can be represented by the construction of a stability diagram which provides a consistent and rational explanation of the flow phenomena observed in the system. These results represent one of the first applications of concepts from nonlinear hydrodynamic stability theory to experimental studies of non-Newtonian fluid flow. The construction of similar stability diagrams for the flow of other viscoelastic materials — from theoretical considerations and further experimental measurements — is important for defining and evaluating stable operating regimes for viable commercial processes.
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# Table of Contents

List of Figures ..............................................................................9  
List of Photographic Plates ..........................................................19  
List of Tables ...............................................................................20  

## 1. Introduction .........................................................................22  
1.1 Motivation ...........................................................................22  
1.2 Instabilities in the Flow of Viscoelastic Materials .............28  
1.3 Optical Techniques in Non-Newtonian Fluid Mechanics ...31  
1.4 Thesis Aims ..........................................................................37  

## 2. Literature Review .................................................................39  
2.1 Rotational Flows of Viscoelastic Liquids .........................39  
2.2 Viscoelastic Flow Through Axisymmetric Contractions ......49  
  2.2.1 Entry Flows of Newtonian Fluids and Inelastic Liquids ....52  
  2.2.2 Contraction Flows of Shear-Thinning Viscoelastic Solutions ...55  
  2.2.3 Contraction Flows of Boger Fluids ...............................61  
2.3 Viscoelastic Flow Past a Constrained Circular Cylinder ......72  
  2.3.1 Newtonian Flow Past Cylinders and Spheres .............74  
  2.3.2 Viscoelastic Flow Around a Cylinder .........................84  
  2.3.3 Viscoelastic Flow Around Spheres ...............................95  
  2.3.4 Summary .....................................................................105  

## 3. Experimental Apparatus .........................................................107  
3.1 Laser Doppler Velocimetry (LDV) ......................................107  
  3.1.1 Principles of Laser Doppler Velocimetry ..................108  
  3.1.2 The Three-Color LDV System .................................115  
  3.1.3 Special Alignment Considerations for a Six Beam LDV System ..119  
3.2 Flow Visualization ...............................................................125  
3.3 Design of Flow Geometries ...............................................128  
  3.3.1 The Axisymmetric Contraction Geometry ..................128  
  3.3.2 The Constrained Cylinder Geometry .......................131  
3.4 Fluids Handling System .....................................................133
4. The Rheology of Boger Fluids .............................................. 136
  4.1 Rheological Fundamentals .............................................. 136
    4.1.1 Definitions of Viscometric Flows .............................. 136
    4.1.2 The Mechanical Spectrometer .................................. 142
  4.2 Composition and Preparation of Test Fluid ....................... 146
    4.2.1 Boger Fluid Composition ...................................... 147
    4.2.2 Molecular Characteristics of the PIB/PB/C14 Solution ....... 150
  4.3 Rheological Models .................................................. 155
    4.3.1 The Oldroyd-B Model ............................................ 155
    4.3.2 The Bird-DeAguiar Model ...................................... 156
    4.3.3 Multimode Models .............................................. 160

  4.4 Experimental Fluid Characterization ................................ 162
    4.4.1 Linear Viscoelasticity ......................................... 162
    4.4.2 Steady Shear Flow ............................................. 171
    4.4.3 Transient Shear Flows ......................................... 180
    4.4.4 Extensional Flows .............................................. 185
    4.4.5 Summary of the Modeling of Boger Fluids .................... 189

  4.5 Dimensionless Flow Parameters for the PIB Boger Fluid ......... 190

5. The Rotational Flow Instability for Boger Fluids .................. 195
  5.1 Flow Parameters in Rotational Flow ................................ 195
  5.2 Experimental Conditions ............................................ 197
    5.2.1 Data Acquisition and Analysis ................................ 197
    5.2.2 Temperature Effects and their Corrections .................. 198

  5.3 Experimental Results .............................................. 199
    5.3.1 The Cone-and-Plate Geometry ................................ 200
    5.3.2 The Parallel-Plate Geometry ................................ 203

  5.4 Characteristics of the Rotational Flow Instability .............. 210
    5.4.1 Temporal Form of the Instability ........................... 213
    5.4.2 Spatial Form of the Instability .............................. 215
    5.4.3 Flow Stability Diagram for Rotational Flow of Elastic Liquids .... 224
    5.4.4 Interpretation of Literature Results for the 'M1' Test Fluid ...... 228
  5.5 Summary ........................................................................ 231

6. Viscoelastic Flow in Axisymmetric Abrupt Contraction ............. 233
  6.1 Flow Visualization ................................................... 234
  6.2 LDV Measurements of Flow Kinematics Near the Lip ............... 240
    6.2.1 Newtonian Flow .................................................. 240
    6.2.2 The Lip Instability ............................................ 240
    6.2.3 Period-Doubling Transition ................................... 251
    6.2.4 Quasiperiodic Flow Near the Lip .............................. 251
    6.2.5 Aperiodic Flow .................................................. 259

7
6.3 Global Kinematic Modifications to the Flow ......................... 261
  6.3.1 The Diverging Flow Regime...................................... 261
  6.3.2 The Elastic Vortex Instability................................... 265
6.4 Effect of Lip Curvature on the Flow Dynamics ....................... 268
6.5 Discussion of Results ................................................. 277
  6.5.1 Influence of Contraction Ratio on Flow Stability ............... 277
  6.5.2 Application of Phase-Plane Techniques .......................... 284
  6.5.3 Comparison with Numerical Simulations .......................... 288

  7.1 Newtonian Flow .................................................... 297
    7.1.1 Flow in a Rectangular Channel ............................... 297
    7.1.2 Creeping Flow Past a Constrained Cylinder .................. 301
  7.2 Effects of Elasticity on Flow Past the Cylinder ................... 304
    7.2.1 Evolution of Flow Streamlines with $De$ ...................... 304
    7.2.2 The Elastic Wake Instability ................................ 305
    7.2.3 Time-Dependent Flow In the Wake ............................. 317
  7.3 The Effect of the Cylinder-Channel Ratio ............................ 320
  7.4 Discussion of Results .............................................. 332

8. Conclusions ....................................................................... 339

9. References ......................................................................... 344

Appendix A  Listing of Flow Sequences for attached Videotape Entitled
              “Nonlinear Dynamics of Viscoelastic Flow in Complex Geometries”.. 359
List of Figures

Figure 1.1 A typical polymer processing flow and its decomposition into a number of simpler subproblems................................................................. 24

Figure 1.2 A comparison of flow visualization experiments and numerical simulations for viscoelastic flow of a Boger fluid through a 4:1 abrupt, axisymmetric contraction................................................................. 27

Figure 1.3 Onset of flow instabilities in the extrusion of a silicone gum through an orifice die............................................................................................ 30

Figure 1.4 Optical measuring techniques for experimental fluid mechanics........ 33

Figure 2.1 Time-dependent response of the stresses measured in the flow of a Boger fluid during rotational flow between parallel plates at $De = 7$......................................................... 44

Figure 2.2 Multiple solution families for the radial velocity profiles and the torque exerted on the top and bottom plates for rotational flow of an Oldroyd-B fluid model between infinite coaxial disks................................................................. 46

Figure 2.3 Schematic diagram of the axisymmetric contraction geometry. ............. 50

Figure 2.4 Evolution of the streamlines and the vortex reattachment length $\lambda$ with increasing Reynolds number for the flow of Newtonian liquids through axisymmetric contractions................................................................. 54

Figure 2.5 Flow visualization photographs for flow of a shear-thinning PAC/water solution in a 4:1 axisymmetric contraction showing vortex growth and diverging streamlines upstream of the contraction................................................................. 56

Figure 2.6 Evolution of the corner vortex in viscoelastic flow of a PAC/water solution through a 4:1 planar contraction with a smoothly curved lip entrance............. 60

Figure 2.7 Sequence of flow transitions observed in the viscoelastic flow of a PAC/CS Boger fluid through a 7.675:1 axisymmetric contraction................................................................. 65

Figure 2.8 Bifurcation diagram for the entry flow of a PIB/PB/K Boger fluid showing the onset of time-periodic oscillations near the lip of a 4:1 contraction............. 67

Figure 2.9 Qualitative sketches showing the evolution in flow structure in axisymmetric contractions as a function of increasing $De$ and contraction ratio for PIB/PB Boger fluids and PAC/CS Boger fluids................................................................. 69

Figure 2.10 LDV Measurements of the time-dependent centerline axial velocity near the contraction plane for the flow of a PIB/PB/C14 Boger fluid in a 4:1 axisymmetric contraction................................................................. 71
Figure 2.11 Viscoelastic flow past a circular cylinder constrained in a planar slit... 73

Figure 2.12 (a) Schematic diagram for a sphere of radius \(a\) and density \(\rho_s\) settling in a viscous fluid of Newtonian viscosity \(\mu\) and density \(\rho_f\). (b) Comparison of the experimentally measured values of the drag coefficient for a sphere and two different theoretical approximations. ......................................................... 75

Figure 2.13 (a) Unbounded flow of a Newtonian fluid with viscosity \(\mu\) past a fixed circular cylinder of radius \(a\). (b) Streamlines for Newtonian creeping flow past a cylinder in a channel of width \(b = 5a\). ................................................................. 77

Figure 2.14 Curves for calculation of the increase in drag coefficient and pressure drop caused by the presence of walls for creeping flow past a cylinder.............. 79

Figure 2.15 Comparison of the streamlines experimentally observed and numerically calculated for the creeping flow of water past a cylinder at \(Re = 0.16\). ............... 81

Figure 2.16 Creeping flow of a Newtonian fluid past a cylinder. (a) Laminar flow with downstream shifting of the streamlines, \(Re = 1.54\); (b) Laminar flow with ‘standing eddies’, \(Re = 26\). ............................................................... 82

Figure 2.17 Onset of flow instability and the development of a von Kármán ‘vortex street’ in the laminar wake behind a cylinder as the Reynolds number is increased................................................................ 83

Figure 2.18 Viscoelastic flow past a cylinder showing a pronounced \textit{upstream} shift of the streamlines. (a) Calculations of the streamlines for \(Re = 0.1, De = 0.5\). (b) Experimental streamlines past a cylinder of diameter \(2a = 3/8\)”. ......................................................... 88

Figure 2.19 (a) Calculations for flow past a cylinder showing a slight \textit{downstream} shift of the streamlines at \(De = 0.2\). (b) Experimentally determined drag coefficient as a function of \(Re\) for a Newtonian fluid, and an aqueous solution of 2 wt% PAC. (c) Quadratic departure of the viscoelastic drag coefficient from the Newtonian prediction............................................................... 90

Figure 2.20 Contours of constant stress for viscoelastic flow past an unbounded cylinder.................................................................................. 94

Figure 2.21 Flow visualization and centerline velocity measurements demonstrating the formation of a ‘negative wake’ behind a sphere falling through a narrow cylindrical tube of viscoelastic fluid................................. 99

Figure 2.22 (a) Comparison of various authors’ numerically calculated drag reduction for a sphere falling axially in a tube; (b) Comparison of the normalized wall correction factor \(X\) obtained from numerical simulations with the experimental measurements of Mena \textit{et al.} (1987). .............................................................. 101
Figure 2.23  Aperiodic fluctuations in the axial and radial velocity components for viscoelastic flow around a sphere falling along the centerline of a cylindrical tube at $D_{eq} = 30$. ................................................................. 104

Figure 3.1  Geometric arrangement for a typical ‘dual-beam’ laser Doppler velocimeter, and its interpretation in terms of the ‘fringe model’. ................................................................. 109

Figure 3.2  (a) Characteristic Doppler burst generated by a single particle passing through the measuring volume; (b) Frequency spectrum of the Doppler burst calculated by using a fast Fourier transform (FFT). ................................................................. 111

Figure 3.3  (a) The relationship between the measured Doppler frequency and the actual velocity component; (b) The modified velocity-frequency relationship for a ‘dual-beam’ LDV arrangement employing a frequency-shift system. ................................................................. 113

Figure 3.4  Schematic diagram showing the optical and geometric arrangement of the three-color, six-beam laser Doppler velocimetry system. ................................................................. 116

Figure 3.5  Vector diagram showing the orientation of the incident green and violet beam-pairs with respect to the Cartesian coordinate system of the flow cell. ....... 120

Figure 3.6  Analysis of refraction effects on the green and violet beam-pairs at the air-plexiglass interface of the flow cell. ................................................................. 123

Figure 3.7  Schematic diagram of the optical configuration used in video flow visualization experiments. ................................................................. 127

Figure 3.8  (a) Detailed view of the axisymmetric contraction geometry showing the annular inserts employed to vary the contraction ratio $\beta$. (b) Modified lip entrance with smooth radius of curvature $R = 0.5R_2$. ................................................................. 130

Figure 3.9  Experimental design of the flow cell developed for LDV studies of viscoelastic flow past a circular cylinder constrained in a planar channel. ................................................................. 132

Figure 3.10  Flow loop for circulation of the 0.31 wt% PIB Boger fluid. .......... 134

Figure 4.1  The five simple shearing flow experiments used in this thesis to characterize the rheological properties of the Boger fluid. ................................................................. 137

Figure 4.2  The deformation of a unit cell in simple shearrfree flows; (a) uniaxial elongation, (b) biaxial stretching, (c) planar extension. ................................................................. 143

Figure 4.3  Test geometries for rheological characterization of simple shear material functions; (a) the cone-and-plate geometry, (b) the parallel-plate geometry. ....... 145

Figure 4.4  Zero-shear-rate viscometric properties $(\eta_0, \Psi_{1,0})$ of the PIB/C14 solutions as a function of the concentration (in weight percent) of PIB. ................................................................. 148
Figure 4.5 The molecular weight distribution of the high molecular weight polyisobutylene (PIB) polymer and the low molecular weight polybutene (PB) solvent .................................................. 152

Figure 4.6 Steady shear material functions for the nonlinear encapsulated dumbbell model of Bird and DeAguiar (1983) demonstrating the effect of finite extensibility in the dumbbells and the effect of a nonisotropic drag coefficient .................. 159

Figure 4.7 Frequency dependence of the viscous modulus $G''(\omega)$ for the 95.15 wt% PB/C14 solution (Boger fluid solvent) over a range of temperatures .............................................. 163

Figure 4.8 Master curve of the viscous modulus $G''(\omega)$ at $T_0 = 25$ °C for the 95.15 wt% PB/C14 data shown in Figure 4.7 ................................................................. 165

Figure 4.9 Linear viscoelastic properties $\eta'(\omega)$ and $2\eta''(\omega)/\omega$ of the 0.31 wt% PIB/PB/C14 solution over a range of temperatures ...................................................... 166

Figure 4.10 Master curves of the dynamic properties $\eta'(\omega)$ and $2\eta''(\omega)/\omega$ at $T_0 = 25$ °C for the 0.31 wt% PIB/PB/C14 Boger fluid together with the predictions of a single-mode Oldroyd-B model ...................................................... 168

Figure 4.11 Predictions of the linear viscoelastic spectrum for the 0.31 wt% PIB/PB/C14 Boger fluid obtained with a discrete four mode relaxation spectrum ............. 170

Figure 4.12 Temperature dependence of the shift factor $\alpha_T$ for the 95.15 wt% PB/C14 solvent (▲) and the 0.31 wt% PIB/PB/C14 Boger fluid (○) ......................... 172

Figure 4.13 Master curves of the viscosity $\eta$ and first normal stress coefficient $\Psi_1$ measured in steady shear flow for the 0.31 wt% PIB/PB/C14 Boger fluid, together with the single mode predictions of the Oldroyd-B model and the Bird-DeAguiar model ...................................................... 174

Figure 4.14 Master curves of the viscous and elastic material functions for the 0.31 wt% PIB/PB/C14 Boger fluid at 25°C in steady-shear and small-amplitude oscillatory shear flow. Also shown are the final model predictions of the nonlinear 4-mode Bird-DeAguiar model for the dynamic (−−−) and viscometric (-----) data .......... 177

Figure 4.15 Extension of the steady-shear viscosity master curve to high shear-rates using the Instron capillary rheometer ...................................................... 179

Figure 4.16 Comparison of the stress relaxation function $\eta^-(t,\gamma_0)$ at 25 °C for the 0.31 wt% PIB/PB/C14 Boger fluid, plus the predictions of the single mode Bird-DeAguiar model ...................................................... 181

Figure 4.17 Stress relaxation function $\eta^-(t,\gamma_0)$ at 25 °C for the 0.31 wt% PIB/PB/C14 Boger fluid, plus the predictions of the four mode constitutive equations; (-----) linear viscoelastic envelope; (-----) four mode Bird-DeAguiar model ...................................................... 183
Figure 4.18 Time-dependent material function $\Psi^+(t, \dot{\gamma}_0)$ measured in the start-up of steady shearing flow for the 0.31 wt% PIB/PB/C14 Boger fluid ........................................184

Figure 4.19 Relaxation modulus $G(t, \dot{\gamma}_0)$ at 25 °C for the 0.31 wt% PIB/PB/C14 Boger fluid. (----) Linear viscoelastic envelope; (-----) 4-mode Bird-DeAguiar model ........................................................................186

Figure 4.20 Trouton ratios ($\bar{n}_1/\eta_0$) for the Boger fluid in (a) uniaxial elongation ($\kappa = 0$), and (b) planar extension ($\kappa = 1$). (-----) 4-mode Bird-DeAguiar model; (- - -) 4-mode Oldroyd-B model .................................................................188

Figure 4.21 The shear-rate-dependent elastic and inertial flow parameters $De(\dot{\gamma})$, $Re(\dot{\gamma})$ calculated using the 4-mode Bird-DeAguiar model for the 0.31 wt% PIB/PB/C14 Boger fluid ..................................................................................192

Figure 4.22 Comparison of the shear-rate-dependent Deborah number $De(\dot{\gamma})$ that can be achieved for a number of different Boger fluid formulations presented in the literature ..................................................................................................................193

Figure 5.1 Initial transient response of the shear stress $\tau(t)$ and the first normal stress difference $N_1(t)$ in the cone-and-plate geometry upon inception of steady shear flow as the rotation rate and Deborah number are increased ..............................................201

Figure 5.2 Onset of a flow transition and apparent 'anti-thixotropic behavior' in the shear stress and first normal stress difference after a period of steady-shear flow in the cone-and-plate rheometer .................................................................202

Figure 5.3 Experimental measurements of the shear stress over a period of 1500 seconds in the cone-and-plate geometry for Deborah numbers of $De = 4.37$ and $De = 4.41$ .........................................................................................................................204

Figure 5.4 The initial transient response and the long-time behavior of the apparent shear stress $\tau_a(t)$ in the parallel-plate geometry as the rotation rate is increased from $\Omega = 5.65$ to $\Omega = 7.36$ rad/s .......................................................................................................................205

Figure 5.5 The initial transient response and the long-time behavior of the apparent first normal stress difference $N_{1a}(t)$ at a constant Weissenberg number of $We = 58.2 \pm 1.0$ as the Deborah number is increased .......................................................207

Figure 5.6 The initial transient response and the long-time behavior of the apparent shear stress $\tau_a(t)$ at constant Deborah number $De = 4.65 \pm 0.10$ as the Weissenberg number is increased from $We = 29.1$ to $We = 172$ ..................................................................................................................208

Figure 5.7 Hysteresis in the shear stress and the normal stress above the critical Deborah number in the parallel-plate geometry ..........................................................................................209
Figure 5.8 Local form of the flow instability in the parallel-plate geometry for a plate separation of $H = 1.80$ mm. The viscometric base solution (○) becomes unstable at a critical shear rate and the flow becomes time-dependent and three-dimensional (●). ................................................................. 211

Figure 5.9 The apparent first normal stress difference $N_{1a}$ measured in a 'step-shear-rate' experiment with four successively lower shear rates of $\dot{\gamma}a_T = 53.9$, 38.9, 35.6 and 32.7 s$^{-1}$ ................................................................. 212

Figure 5.10 Fast Fourier transform (FFT) of the time-dependent shear stress $\tau_a(t)$ in the parallel-plate geometry at $De = 4.65$. ................................................................. 214

Figure 5.11 Initial exponential growth of the disturbance as measured by the increase in the first normal stress difference above the base response for steady shear flow ................................................................. 216

Figure 5.12 Schematic diagram showing components of apparatus for video-imaging of the rotational flow instability ................................................................. 220

Figure 5.13 Stability diagram for viscometric flow of the Boger fluid between coaxial rotating disks ................................................................. 225

Figure 5.14 Stability diagram for viscometric flow between parallel plates, recalculated by using a shear-rate-dependent relaxation time $\lambda_1(\dot{\gamma})$ that more accurately represents the actual magnitudes of elastic effects in the Boger fluid ................................................................. 227

Figure 5.15 Stability diagram for viscometric flow of the Boger fluid in the cone-and-plate geometry with cone angles of $\theta_0 = 0.04$ and 0.10 radians ................................................................. 229

Figure 6.1 The reattachment length of the vortex observed upstream of the contraction plane as a function of the Deborah number for contraction ratios $2 \leq \beta \leq 8$ ................................................................. 239

Figure 6.2 Centerline axial velocity $u_2(0, \zeta)$ in the large upstream tube for contraction ratios $2 \leq \beta \leq 8$, at flow conditions $De_2 = 0.42, Re_2 = 0.004$ ................................................................. 241

Figure 6.3 (a) Axial velocity profiles $u_2(\xi, -1)$ and (b) radial velocity profiles $u_r(\xi, -1)$ above the contraction plane at a low Deborah number for contraction ratios of $\beta = 2, 3, 4$ and 8 ................................................................. 242

Figure 6.4 Sample time series and frequency spectra showing onset of oscillations of all three velocity components near the lip ($\xi = -1.23, \zeta = -0.32$) of the 4:1 contraction at $De_2 = 1.70$ ................................................................. 244

Figure 6.5 Time series and FFT spectra showing increased amplitude of oscillations in all three velocity components near the lip ($\xi = -1.23, \zeta = -0.32$) of the 4:1 contraction at $De_2 = 2.12$ ................................................................. 245
Figure 6.6  Frequency of oscillations in the axial velocity near the lip of the 4:1 contraction as a function of the Deborah number. ......................................................... 246

Figure 6.7  Square of the amplitude of oscillations in $v_z$ near the lip of the 4:1 contraction as a function of the Deborah number. ......................................................... 247

Figure 6.8  Frequency of oscillations in the radial velocity near the lip as a function of the Deborah number for $\beta = 3$ and 5 ......................................................... 249

Figure 6.9  Square of the amplitude of oscillations in $v_r$ near the lip as a function of the Deborah number for $\beta = 3$ and 5 ......................................................... 250

Figure 6.10  Experimental velocity time series and FFT spectra showing period-doubling of the oscillations in the radial velocity component near the lip of the 4:1 contraction ......................................................... 252

Figure 6.11  Time series and frequency spectrum showing quasiperiodic oscillations in the velocity near the lip of the 5:1 contraction at $De_2 = 3.02$ ..................... 253

Figure 6.12  Dominant frequencies in the time-dependent regimes near the lip of the 3:1 contraction as a function of the Deborah number ......................................................... 254

Figure 6.13  LDV measurements of the steady-state radial and axial velocity profiles near the lip of the 6:1 contraction show the formation of a lip vortex at $De_2 = 2.96$ ..................... 256

Figure 6.14  Stability diagram showing the sequence of nonlinear flow transitions observed in the axisymmetric abrupt contraction as a function of the Deborah number $De_2$ for contraction ratios of $2 \leq \beta \leq 8$ ......................................................... 257

Figure 6.15 (a)  Time series of the axial velocity on the centerline ($\xi = 0.00$, $\zeta = -1.50$) of the 4:1 contraction at $De_2 = 0.4$, 3.1 and 3.7; (b) FFT spectrum of the time-dependent axial velocity shown in (a) at $De_2 = 3.7$ ..................... 258

Figure 6.16  Aperiodic flow along the centerline near the lip ($\xi = 0.00$, $\zeta = -2.25$) of the 4:1 contraction at $De_2 = 4.08$ and $Re_2 = 0.061$ ..................... 260

Figure 6.17  Diverging flow upstream of the contraction plane at high $De_2$ demonstrated by the normalized axial velocity $v_z(0, \zeta)/\langle v_z \rangle$ along the centerline for contraction ratios $\beta = 3$, 4 and 5 ......................................................... 263

Figure 6.18  Transverse axial velocity profiles $v_z(\xi)/\langle v_z \rangle$ at $\zeta = -3.00$ for contraction ratios $\beta = 3$, 4 and 5 at the same $De_2$ as shown in Figure 6.17 ..................... 264

Figure 6.19  Velocity time series and FFT spectra demonstrating two different modes of oscillation for the large elastic vortex: (a) – (b) slow axial pulsing of the vortex at $De_2 = 4.49$ in the 4:1 contraction; (c) – (a) rapid azimuthal rotation of the vortex at $De_2 = 4.33$ in the 5:1 contraction ..................... 266
Figure 6.20 Time-dependent velocity profiles measured far upstream (ζ = -25) of the contraction plane for β = 4 and β = 5. .................................................................269

Figure 6.21 Comparison of the dimensionless vortex reattachment length (χ) for flow through a 4:1 contraction with a sharp lip and with a smoothly curved lip with radius of curvature $R = 0.5R_2$ .................................................................273

Figure 6.22 Time-dependent oscillations in the velocity near the lip of the 4:1 contraction with a curved entrance lip ($\beta = 4\xi$) at $De_2 = 5.02$ and $Re_2 = 0.088$. ..........275

Figure 6.23 Normalized axial velocity $u_2(0, \xi)/\langle u_2 \rangle_2$ along the centerline of the 4:1 contraction with a curved entrance lip ($\beta = 4\xi$) showing diverging flow at $De_2 = 4.60$, $Re_2 = 0.080$. .................................................................276

Figure 6.24 Schematic drawings showing stability diagrams that provide a rational description of the results presented in this thesis and the earlier observations of Boger (1987) for contraction flows of (a) PIB/PB Boger fluids and (b) PAC/CS Boger fluids.................................................................280

Figure 6.25 (a) Autocorrelation function $\mathcal{A}(t)$ of the axial velocity time series at $De_2 = 4.49$ shown in Figure 6.19(a). (b) Phase-plane portrait constructed by using the method of delays to generate the independent coordinates $u(t)$, $u(t + 5.5)$.....286

Figure 6.26 Autocorrelation function $\mathcal{A}(t)$ and phase-plane portrait of the axial velocity time series near the lip of the 4:1 contraction at $(\xi, \zeta) = (1.02, -0.32)$ and $De_2 = 4.49$ .................................................................287

Figure 6.27 (a) FFT of the time-dependent radial velocity component $u_r$ measured at $(\xi, \zeta) = (-1.68, -0.30)$ in the 4:1 contraction for $De_2 = 4.08$. (b) Autocorrelation function $\mathcal{A}(t)$ of the Fourier spectrum shown in (a).........................289

Figure 7.1 Schematic diagram showing viscoelastic flow past a circular cylinder constrained in a planar slit.................................................................296

Figure 7.2 Profiles of the axial velocity $u(x, y)$ in a rectangular channel with aspect ratio of $\Lambda = \Delta x/\Delta y = 10$ at $De = 0.07$, $Re = 0.001$.................................................................298

Figure 7.3 Profiles of the axial velocity component in a rectangular channel with aspect ratio $\Lambda = 6$ at $De = 0.73$, $Re = 0.001$. .................................................................300

Figure 7.4 Comparison of LDV measurements and numerical calculations of the centerline axial velocity in the planar stagnation flow near the cylinder at $De = 0.073$.................................................................302

Figure 7.5 Transverse velocity profiles of the dimensionless axial velocity $u_2/\langle u_2 \rangle$ showing Newtonian creeping flow of the Boger fluid past a circular cylinder at $De = 0.07$, $Re = 0.001$.................................................................303
Figure 7.6  Evolution of the axial velocity profiles \( u_x / \langle u_x \rangle \) along the channel centerline with increasing \( De \); (a) upstream of the cylinder, and (b) in the downstream wake of the cylinder. .................................................................305

Figure 7.7  Profiles of the axial velocity \( u_x(x) \) across the width of the channel at \( (\nu, \xi) = (0, 1.5) \) as a function of \( De \). .................................................................307

Figure 7.8  Magnitude of the axial velocity \( u_x \) measured in the wake of the cylinder as a function of the Deborah number showing the onset of velocity fluctuations at \( De = 1.30 \). .................................................................308

Figure 7.9  Spatial fluctuations in the axial velocity \( u_x(x) \) measured in the downstream wake of the cylinder at \( De = 1.38, Re = 0.017 \). .................................................................310

Figure 7.10 Time-series and frequency spectrum of the axial velocity \( u_x(t) \) in the cylinder wake at the same position and flow conditions as Figure 7.9. .................................................................311

Figure 7.11 (a) Fluctuations in both velocity components \( u_x(x) \) and \( u_y(x) \) across the width of the channel at \( (\nu, \xi) = (0, 1.5) \) and flow conditions of \( De = 1.77, Re = 0.012 \); (b) – (c) Evaluation of the spatial wavelength of the oscillations in each velocity component from FFT spectra. .................................................................314

Figure 7.12  Fluctuations in both velocity components \( u_x(x) \) and \( u_y(x) \) across the width of the channel at \( (\nu, \xi) = (0, 2.0) \) and same flow conditions as Figure 7.11. .... 315

Figure 7.13 Evolution of the spatial structure observed in the centerline axial velocity \( u_x(\xi, 0, \xi) \) within the wake of the cylinder at flow conditions of \( De = 1.83, Re = 0.0158 \). .................................................................316

Figure 7.14 Measurements of the axial velocity profiles along the length of the cylinder at different transverse positions of \( y/R = 0, 0.53, 0.79 \) and 1.10 showing that the three-dimensional cellular structure is confined to narrow region of the cylinder wake. .................................................................318

Figure 7.15 Velocity time-series and FFT spectra showing the onset of time-dependent flow in the cylinder wake for \( De \geq 1.80 \). .................................................................319

Figure 7.16 Centerline axial velocity profiles in the three-dimensional wake show a pronounced shift downstream but increase monotonically to the free stream value with no 'negative wake' or aperiodic fluctuations. .................................................................321

Figure 7.17 Effect of the cylinder-channel ratio \( \beta \) on the centerline axial velocity profiles in the upstream and downstream stagnation flows near the cylinder at the same Deborah number of \( De = 0.073 \). .................................................................323

Figure 7.18 Spatial fluctuations in the axial velocity \( u_x(x) \) measured in the downstream wake of the smaller cylinder (\( \beta = 0.257 \)) at \( De = 2.00 \). .................................................................324
Figure 7.19 Comparison of the axial velocity component $v_x(x)$ at the same
Deborah number of $De = 1.38$ for two different cylinder-channel ratios of
$\beta = 0.257$ and $\beta = 0.503$.................................................................326

Figure 7.20 Comparison of the axial velocity component $v_z(x)$ at the same
upstream flow conditions of $\langle v_x \rangle / H = \text{for the two different cylinder-channel ratios} ....327

Figure 7.21 Evolution of the spatial structure observed in the centerline axial
velocity $v_x(\xi, 0, \zeta)$ within the wake of the smaller cylinder ($\beta = 0.257$) at flow
conditions of $De = 1.83$, $We = 1.07$, $Re = 0.0158$. ........................................328

Figure 7.22 Centerline axial velocity profiles in the wake of the smaller cylinder
($\beta = 0.257$) show a pronounced downstream displacement as the Deborah number
is increased.................................................................330

Figure 7.23 A stability diagram for viscoelastic flow past circular cylinders of
radius $R$ in planar channels of half-width $H$. ...................................................331

Figure 7.24 An approximate representation of the flow-stream lines along the
channel mid-plane showing the spatial evolution of the velocity profiles and the
development of a cellular structure in the cylinder wake.....................................333
List of Photographic Plates

Plate 5.1  Temporal evolution of the free-surface shape in the parallel-plate geometry at a rotation rate of $\Omega = 7.32$ rad/s and a plate separation $H = 1.80$ mm........218

Plate 5.2  Visualization of the spatial structure of the rotational flow instability in the parallel plate geometry at $De = 4.69$.................................222

Plate 6.1  Streak photographs showing evolution of the velocity field with $De$ for flow of the 0.31 wt% PIB Boger fluid through abrupt axisymmetric contractions.................................................................236

Plate 6.2  Viscoelastic flow of the 0.31 wt% PIB Boger fluid through a 4:1 contraction with a smoothly curved lip entrance..............................................271

Plate 6.3  Comparison of the numerically calculated streamlines for a single-mode Modified Upper-Convected Maxwell model (MUCM) and experimental streak photographs for the 0.31 wt% PIB/PB Boger fluid in the 8:1 axisymmetric abrupt contraction.................................................................291

Plate 7.1  Video-imaging of the cellular structure in the wake of the cylinder at flow conditions of $De = 2.48$, $Re = 0.028$.................................................................312
List of Tables

Table 2.1 Previous experimental investigations of viscoelastic entry flows using highly elastic Boger fluids ................................................................. 63

Table 2.2 Previous investigations of viscoelastic flow around a circular cylinder ........................................................................................................... 85

Table 2.3 Previous investigations of viscoelastic flow around a sphere ......................................................................................................................... 96

Table 3.1 Geometric specifications for the measuring volumes associated with each individual beam-pair in the three color LDV configuration ........................................................................................................... 117

Table 3.2 Overall specifications of the three-component LDV system ............................................................................................................................... 118

Table 3.3 Characteristic dimensions of the annular inserts used in the study of viscoelastic flow through axisymmetric abrupt contractions .................................................................................................. 129

Table 4.1 Mark-Houwink Parameters for construction of the GPC universal calibration curve (from Brandup and Immergut, 1978; p. IV-9) ........................................................................................................ 150

Table 4.2 Linear viscoelastic spectrum for the 0.31 wt% PIB/PB/C14 Boger fluid at $T_0 = 25^\circ$C ......................................................................................................................... 169

Table 4.3 Flow activation energy and zero-shear-rate viscosity at $T_0 = 25^\circ$C obtained from experimental measurements of $\eta'(\omega,T)$ ......................................................................................................... 171

Table 4.4 Model parameters used in the 4-mode Bird-DeAguiar formulation constitutive equation ................................................................................................................. 176

Table 5.1 The critical shear stress, rotation rate and Deborah number for onset of the rotational instability in cone-and-plate flow of the ‘M1’ viscoelastic test fluid ........................................................................................................... 230

Table 6.1 Flow parameters at the formation of a time-dependent lip vortex for contraction ratios $\beta = 2, 3, 4$ and $5$. ............................................................................................................... 248

Table 6.2 Flow conditions at the onset of unsteady oscillations in the large upstream vortex for $2 \leq \beta \leq 6$ ........................................................................................................................................ 267

Table 7.1 Critical flow conditions for onset of the three-dimensional cellular instability in the cylinder wake for cylinder-channel ratios of $\beta = 0.257, 0.503$ ........................................................................... 325

Table 8.1 Spatial and temporal structures of viscoelastic flow instabilities in axisymmetric and planar test geometries ...................................................................................................................................... 341
"These instruments have played me so many tricks that I have at last found them out in many of their humours, and have made them confess to me what they would have concealed, if I had not with perseverance and patience courted them"

Sir William Herschel, Astronomer Royal  1781
Chapter 1

Introduction

1.1 Motivation

Annual production of polymeric materials continues to increase, and in 1988 U.S plastic production exceeded 23 million tonnes (Chemical & Engineering News, June 19, 1989). Over 80% of this total consisted of thermoplastics which, following synthesis, may be extruded, injected, drawn or blown into a vast range of final products. The material characteristics and financial value of these products often depends intimately on the operating conditions and the deformation history experienced by the polymer during processing; a fundamental understanding of the fluid mechanics involved can thus be of great commercial importance. Fiber-spinning of nylon filaments for textiles, injection molding of engineering plastic resins for the production of automobile parts and the extrusion of polymer-based propellants are several examples of the integration of complex flows into industrial manufacturing programs.

Newtonian liquids may be characterized by a single fluid property, the constant Newtonian viscosity $\mu$, and at low flow rates the equations of motion are dominated by dissipative viscous forces. Polymeric materials, however, exhibit both viscous and elastic responses during flow and are described as being viscoelastic. This fluid elasticity arises from the ability of the long macromolecules to support non-zero normal stresses in addition to viscous shear stresses as they are being deformed. Polymeric fluids can readily be distinguished from Newtonian liquids by their behavior in simple flows and exhibit a wide range of non-Newtonian phenomena such as rod-climbing and tubeless siphoning (Bird et al., 1987a). In addition, the rheological properties of viscoelastic fluids are found to depend intimately on the local process conditions; for example the material functions of many polymer melts and solutions are found to decrease with increasing shear rate. The
characteristic response of a given viscoelastic material thus varies throughout a complex geometry.

The combination of dissipative viscous effects, coupled with the storage of elastic energy, results in a ‘fading memory’ of past deformations, and a viscoelastic fluid flowing through a complex processing geometry partially ‘remembers’ the previous stress history imposed on it. The importance of fluid elasticity in a particular flow process can be characterized by the Deborah number (Bird et al., 1987a) defined as

$$De = \frac{\lambda}{\tau}$$

where $\lambda$ is a characteristic relaxation time for the viscoelastic material and $\tau$ is a characteristic residence time in the flow of interest. The Deborah number may be interpreted as a ratio of the elastic forces to the viscous forces that are experienced by the flowing polymer. In slow flows with a vanishingly small Deborah number, the fluid behaves as a Newtonian fluid dominated by viscous dissipative forces. However, at large Deborah numbers the fluid exerts significant elastic effects; and, in the limit $De \to \infty$, the fluid acts as a Hookean solid, in which an applied deformation results in perfect elastic energy storage with no viscous dissipation. At Deborah numbers of $O(1)$ however, both elastic effects and viscous effects will be important in the flow, and it is not possible to infer the behavior of the material from the creeping flow of a simple Newtonian fluid. The Deborah number is thus analogous to the familiar Reynolds number in Newtonian fluid mechanics, which scales the relative importance of inertial and viscous forces in the flow.

The Deborah number defined by eq. (1.1) for a particular process is a function of both the material properties of a fluid and the processing rate. As the throughput of a given viscoelastic material is increased, the residence time in the flow geometry decreases, and the Deborah number consequently increases. In many polymer processing operations, the economic viability of the process is directly related to the volumetric throughput of the material. However, increasing the processing rate results in an increase in the value of the Deborah number and the fluid thus acts increasingly like an elastic solid rather than a viscous liquid. This can ultimately lead to the development of unstable flows, and in a number of important industrial processes, such as fiber-spinning and melt extrusion, processing limits are encountered at quite moderate Deborah numbers ($De = 1 - 10$) due to flow transitions that lead to unsteady flow regimes (Pietie and Denn, 1976).

A typical commercial process for extruding PVC piping is shown in Figure 1.1, and illustrates many of the salient features of a complex flow geometry. The solid
Figure 1.1 Schematic diagram of a typical polymer processing flow and its decomposition into a number of simpler subproblems.
thermoplastic resin enters from the left and is heated above its melting point as it is forced along the barrel of the extruder by the rotating helical screw. It is then extruded into an annular shape as it flows past the die mandrill, which is held rigidly in the center of the exit pipe by the 'spider' arms. One of the ultimate aims of viscoelastic fluid mechanics is to be able to predict the morphology and structural properties of the finished product from fundamental knowledge of the material's rheological properties and the geometry of the flow. Optimization of the structure-property relationships can then be achieved by varying the processing conditions and the geometry shape. Unfortunately, the complex flow shown in Figure 1.1 is three-dimensional, nonisothermal, inherently time-dependent and beyond the scope of current numerical simulations. Meanwhile, experimental investigations in such systems are limited to empirical correlations of temperature, pressure drop and the flow stability against flow rate with little fundamental understanding of the relation between the process and product. The problem can, however, be decomposed into a number of simpler, fundamental subproblems as shown in Figure 1.1. These geometries retain the basic fluid mechanics that are associated with characteristics such as time-dependence, extensional flow, the presence of singularities and free surface effects, while still being both numerically tractable and amenable to precise experimental investigations.

Rotational flows occur commonly in processing applications such as extrusion and spin coating, in addition to simpler geometries such as viscometric flow between parallel plates. For the two-dimensional eccentric cylinder geometry shown schematically in Figure 1.1, the flow is primarily a shear flow but the converging and diverging regions of the flow near the narrow part of the 'nip' are highly extensional in character. It is this combination of extensional and shearing components that leads to effective mixing in closely related geometries such as the twin-screw extruder (Pearson, 1981). Entry flows through a suddenly contracting geometry also involve a combination of shearing and extensional deformations and are ubiquitous in the field of polymer processing. The extensional flow near the centerline can result in significant flow-induced molecular orientation and a consequent improvement in the ultimate structural properties. Planar and axisymmetric geometric arrangements have both been studied extensively for a wide range of polymer solutions and melts (White et al., 1987). Stagnation flows past submerged bodies such as cylinders or spheres are also extremely important since the strong extensional flows near the stagnation points result in extension of the macromolecules and the development of large elongational stresses. This stress build-up in the downstream wake of the object can lead to modifications in the velocity field around the object and, more seriously, to the development of microstructural inhomogeneties and defect structures such as 'weld-lines' in the finished product (Tadmor and Gogos, 1979). Finally, free
surface problems are also of importance, since phenomena such as die swell and surface wetting determine the physical characteristics of products produced from polymeric materials in free extrusion or coating flows. Several of these geometries have been specified as benchmark problems for viscoelastic flow (Hassager, 1988) and research in the group at M.I.T. has focused on each of them. With the advent of relatively affordable supercomputers it is now possible to perform large-scale numerical simulations that span physically reasonable process conditions, and that incorporate into each of these subproblems such effects as a spectrum of relaxation times (Rajagopalan et al., 1990a), time-dependence and the propagation of shear-waves during the transient start-up of rotational flow (Burdette, 1989; Northey et al., 1990), and three-dimensional, free surface effects in die-swell (Shiojima and Shimazaki, 1990).

It should therefore be possible to make quantitative comparisons of these computational predictions with precise experimental measurements in each flow geometry. To date though, the agreement between such comparisons has been disappointing. In the last 30 years, significant progress has been made in modelling the dynamic behavior of polymeric fluids, and many constitutive equations have been developed which relate the instantaneous level of stress in the fluid to its past deformation history. These constitutive relations can mimic either qualitatively or quantitatively the viscous and elastic responses of a viscoelastic fluid in a wide range of simple, one-dimensional flows (see for example Bird et al., 1987a, b; Larson, 1988). However, the situation is less satisfactory in the complex, two-dimensional flows, such as those shown in Figure 1.1, which are more representative of those encountered in industrial processes. Qualitative predictions of the flow evolution with increasing $De$ are possible in certain well-studied problems, such as the converging ‘entry flow’ through an orifice; however, numerical simulations and experimental observations of flow characteristics, such as the vortex height and excess pressure drop, often differ significantly (Debbaut et al., 1988) and may even be of opposite sign (Keunings and Crochet, 1984). To demonstrate this discrepancy, the comparisons presented by Crochet (1988) for flow of a highly viscoelastic fluid into a 4:1 axisymmetric contraction are shown in Figure 1.2. The results are arranged in order of increasing flow rate from left to right and, qualitatively at least, the flow visualization experiments and numerical computations predict the same behavior: As the flow rate through the contraction is increased, the increasing importance of elasticity leads to a significant change in the size and shape of the recirculating flow observed in the outer corners of the upstream tube. However, a comparison of the characteristic Deborah number $De$ calculated for the experiments and simulations reveals that the numerical computations under-predict the observed vortex enhancement by an order of magnitude.
Figure 1.2  A comparison of the flow visualization experiments presented by Boger et al. (1986) and the numerical simulations of Debbaud et al. (1988) for viscoelastic flow through a 4:1 abrupt, axisymmetric contraction. (Comparison reproduced from Crochet, 1988).
In most other geometries little is known, even qualitatively, about the evolution of complex viscoelastic flows with increasing $De$. The situation is exacerbated by physically unrealistic responses of the constitutive models or by problems associated with the numerical techniques that develop at moderate values of $De$. These numerical problems are usually associated with the formation of steep stress boundary layers that develop in complex flows, or with a physical behavior of the constitutive equation resulting from the presence of singular points, such as sharp corners, boundary discontinuities and stagnation points, in the computational domain. Typically, as the Deborah number of the simulated flow is increased oscillations develop in the numerical solution fields near the singular point and subsequently grow in magnitude until a limiting Deborah number is reached above which convergent numerical solutions cannot be obtained. This failure results from errors introduced by the numerical technique in approximating the extremely large stresses experienced near singular points and occurs with a number of viscoelastic constitutive equations (Gotsis et al., 1990; Coates et al., 1991). This so-called ‘high Deborah number problem’ is discussed in detail in the recent reviews of Keunings (1987) and Crochet (1989), and its resolution requires cooperative developments in each area of viscoelastic fluid mechanics: theoretical work is necessary to develop constitutive equations which more accurately model the dynamic behavior of polymer molecules in complex flows (Rallison and Hinch, 1988; El-Kareh and Leal, 1989), robust numerical methods are required which allow convergent solutions to be obtained at high Deborah numbers while still preserving the characteristics of the original problem (King et al. 1988; Rajagopal et al. 1990b), and finally, precise experiments are needed which enable evaluation of the components of both the stress and velocity fields throughout the flow domain. A central goal of this thesis is the implementation of optical measuring techniques which are capable of precise kinematic measurements of steady viscoelastic phenomena in addition to permitting the detection and investigation of the structure of flow instabilities which may develop at high Deborah numbers.

1.2 Instabilities in the Flow of Viscoelastic Fluids

As we have mentioned above, the maximum processing rate (or equivalently the maximum Deborah number) that can be achieved in a commercial process is often directly limited by a transition from a steady flow regime to an unsteady time-dependent flow. These flow instabilities can result in severe distortion of the exiting material and lead to worthless products in processes such as coating flows (Baumann et al., 1982), fiber spinning (Kase, 1974) and melt extrusion (den Otter, 1970). To illustrate this, the sequence
of instabilities occurring in the extrusion of a silicone gum through an orifice die are shown in Figure 1.3. At low flow rates, such as that shown in Figure 1.3(a), the moderate 13% die-swell associated with the extrusion of a Newtonian fluid (Bird et al., 1987a) can be observed, the extrudate surface appears smooth, and the fluid is translucent. As the pressure driving force increases, the residence time in the die decreases and the extrudate appears opaque or 'matte' in appearance (Figure 1.3(b)). Closer inspection reveals that the surface consists of regular corrugations, a phenomenon often referred to as 'sharkskin'. The amplitude of these surface oscillations increases rapidly as the Deborah number is increased from $De = 0.30$ to $De = 0.31$, and can be clearly identified in the extrudate shown in Figure 1.3(c). At higher flow rates, these fluctuations become more pronounced and eventually result in complete disruption, or 'melt fracture', of the extruded product, as shown in Figure 1.3(d). To complicate matters further, these extrudate distortions are found to depend intimately on the geometric shape of the orifice (Bagley and Schreiber, 1961), the molecular structure of the polymer being extruded (Petrie and Denn, 1976; White and Kondo , 1977/78) and also the materials used to construct the die (Ramamurthy, 1986; Denn, 1990). Before a computational design tool can be developed which accurately models this level of complexity, careful experimental investigations of flow instabilities are required in which the influence of each of these parameters is understood independently.

In each of the representative test geometries shown in Figure 1.1, flow transitions and instabilities have been experimentally observed with viscoelastic fluids which are entirely absent in corresponding experiments with purely Newtonian liquids. These instabilities develop at very low Reynolds numbers and depend solely on the elastic nature of the material (Pearson, 1976). Experimental observations of elastic instabilities have been made in many flows including Taylor-Couette flow (Giesekus, 1972; Muller et al. 1989), rotational flow in a cone-and-plate rheometer (Magda & Larson 1988), flow around a sphere (Bisgaard 1983), die entry flows (Boger 1982, 1987) and extrudate swell of polymer melts (White, 1973, Piau et al. 1990). To date, most theoretical analyses of viscoelastic flow stability have been limited to relatively simple idealized two-dimensional flows. Akbay et al. (1985) considered the linear stability of plane Couette flow, and Lagnado et al. (1985) considered the stability of Maxwell fluids in more general rectilinear flows containing extensional and shearing components. Phan-Thien (1983, 1985) has studied rotational flow stability between coaxial parallel disks and between a cone and plate for simple quasilinear constitutive equations such as the upper-convected Maxwell and Oldroyd-B models. Recently, Larson (1989) has examined the effects of elasticity and shear-thinning on the form of the inertial instability observed in circular Taylor-Couette flow.
Figure 1.3  Onset of flow instabilities in the extrusion of a silicone gum through an orifice die: (a) ΔP = 0.5 bar, De = 0.14; (b) ΔP = 6 bar, De = 0.30; (c) ΔP = 6.75 bar, De = 0.31; (d) ΔP = 10 bar, De = 0.35. (Reproduced from Piau et al., 1990).
Numerical and theoretical prediction of experimentally documented instabilities has remained beyond the realm of computational ability until recently, when the presence of a purely elastic, time-periodic instability in the Taylor-Couette system was demonstrated both theoretically (Larson et al., 1990) and numerically (Northey et al., 1991) and verified by experiments. However, it is important to note that the experiments were limited to macroscopically-averaged measurements of the unsteady torque exerted by the fluid at the surface of the Couette cell. The measurements were able to determine the approximate onset point of the instability, but were insufficient to verify the more rigorous theoretical predictions concerning the form of the bifurcation, the period of oscillation or the spatial structure of the flow instability. Detailed investigation of such quantities requires the application of more accurate experimental methods which can resolve kinematic information on a local basis. In this thesis two optical techniques are employed to provide both qualitative visual confirmation and quantitative point-wise determination of the nonlinear dynamics associated with complex flows of viscoelastic liquids.

1.3 Optical Techniques in non-Newtonian Fluid Mechanics

In the past, most experimental investigations of the sequence of non-Newtonian transitions observed in complex flows have either involved qualitative techniques such as flow visualization or have attempted to correlate macroscopically observable quantities, *e.g.* excess pressure drop or vortex size, with flow parameters such as the Reynolds number and Deborah number that measure the relative importance of inertia and elasticity in the flow. Although very motivating, few of these experiments have been quantitative enough to provide benchmarks for comparison with theory and computations. Such comparisons are an important next step in the development of the fluid mechanics of viscoelastic liquids. To meet this objective, the experiments must be carried out with a rheologically well characterized fluid, must be accurate enough to describe sufficiently both the spatial and temporal symmetries of the flow, and must provide quantitative measurements that can be directly compared with calculations.

The recent work of Gotsis et al. (1990) compared experimental measurements and numerical predictions for the magnitude of stresses obtained in flow of a polystyrene melt into a slit die and is a significant step in the right direction; however, the macroscopic technique used to obtain the experimental data together with the limited accuracy of the manual analysis highlight the limitations of simple global techniques: relatively little experimental data could be extracted from the photographic images, and these results were
limited to measurements of an overall magnitude of stress. In order to analyze the physical behavior of viscoelastic fluids in complex flows especially near surfaces or sharp corners, and to discriminate effectively between the predictions of different models it is highly desirable to be able to interrogate the flow on a much finer scale and to measure the individual components of the stress tensor.

The application of lasers to experimental fluid mechanics has led to the development of a number of optical techniques which can be used to make kinematic measurements more accurately and on much smaller length scales than was heretofore possible (Goldstein, 1983). These techniques and their inter-relation are shown in Figure 1.4. The methods may be broadly subdivided into two categories; (i) transmitted light techniques which rely on refractive index variations between the fluid undergoing flow and the local laboratory environment, and (ii) scattered light techniques in which the kinematics of the fluid flow are inferred from observing the motion of marker particles. In many applications, this latter category also relies on refractive index variations (e.g. between the index of refraction of the seed particles and the fluid itself); however, they may be collected together and differentiated from (i) by the need to introduce a heterogeneous phase of seed particles. The two scattered light techniques employed in this thesis research are identified in Figure 1.4 by the shaded boxes.

Direct flow visualization is the oldest experimental technique in fluid mechanics, and by suitable choice of media and exposure times can be employed in flow regimes varying from creeping flow to hypersonic flow; see Van Dyke (1982), MerzKirch (1987) and Yang (1989) for examples. The use of a laser light source with a cylindrical lens allows the formation of an intense light sheet which can be used to illuminate planar cross-sections of axisymmetric or three-dimensional flows. Long-time photographic exposures lead to ‘streak photographs’ in which the records of a large number of particle tracks with a given Lagrangian velocity $v(x, t)$ lead to an approximate representation of the streamlines of an Eulerian velocity field $u(x)$. Numerous examples of this method exist in the field of non-Newtonian fluid mechanics, and typical results have already been presented in Figure 1.2 for the creeping flow of a viscoelastic fluid through an axisymmetric contraction. Other examples can be found in the extensive work of Walters and coworkers (Cochrane et al., 1981; Walters and Webster, 1982; Evans and Walters, 1986; Binding et al., 1987). The flow visualization technique, however, yields only qualitative kinematic information about the flow, such as the shape of streamlines, and is limited to steady flows. In unsteady flows the particle tracks vary in time and lead to apparent intersection of fluid streamlines (see for example, Binding et al., 1987). In this thesis, we have used a video-based flow visualization system in which the motion of the particles is recorded in real time and the
Figure 1.4  Optical measuring techniques for experimental fluid mechanics
temporal evolution of the velocity field can easily be followed. The development of charge-coupled devices (CCDs) and the use of conventional macroscopic lens elements allow much higher resolutions to be achieved than has previously been possible with motion picture techniques. More recently the development of digital image processing algorithms coupled with stereophotography and holography have even allowed the direct visualization of three-dimensional flow structures in Newtonian fluids (Hesselink, 1988). These techniques have yet to be applied to the flow of viscoelastic fluids.

Semiquantitative results may be extracted from the simple streak photographs obtained in flow visualization by tedious and inaccurate measurements of the length and direction of the particle tracks (e.g. Cable and Boger, 1978b). The spatial resolution of these measurements is limited, especially in flows with a wide dynamic range of velocities; such as flows involving recirculations or boundary layers. A quantitative improvement in measurements of the velocity field within a fluid can be made by the use of laser Doppler velocimetry (LDV). In this technique, two coherent beams of laser light are focused together at a point to create an interference fringe pattern within the flow of interest. The frequency of light scattered by suspended micron-size particles is measured as they are convected by the fluid flow through the fringe pattern. This light is Doppler shifted in frequency by an amount that is directly proportional to the Lagrangian velocity of the particle, and the velocity can be determined by simple knowledge of the geometric arrangement of the beams and the wavelength of the light. With suitable optical configurations all three components of a velocity vector \( \mathbf{v} \) in the flow can be determined (Durst et al., 1981). The LDV technique is non-invasive, offers a wide dynamic range and can be used to measure steady or time-dependent velocity fields on a point-wise basis. It is limited in resolution only by the minimum diameter of the interrogating laser beams (approximately 50 \( \mu \text{m} \) for a diffraction-limited lens system). The technique has been used successfully in a number of investigations of steady viscoelastic flow, from flow of dilute polymer solutions (Berman and Pasch, 1986; Wunderlich et al., 1988) to flow of polyethylene melts (Kramer and Meissner, 1980; Mackley and Moore, 1986). The recent contributions of Bisgaard (1983), plus those of Muller (1986) and Raiford (1988) within this group, have been the first applications of LDV to studies of flow instabilities and unsteady flows of viscoelastic fluids. In this thesis a fully automated three color LDV system will be used to extend this earlier work and study both steady and transient flows of a viscoelastic fluid in two complex geometries.

Accurate and automated procedures for calculating two-dimensional velocity fields have also been developed recently by the use of pulsed-light techniques, in which the displacement \( \Delta \mathbf{r} \) of markers in the flow is recorded by multiple image exposures that are
separated by an interval $\Delta t$. As indicated in Figure 1.4, these markers in the flow can either be molecular in nature, e.g. drops of molecules which either fluoresce or react with the interrogating sheet of laser light, or they can be particles with a different refractive index to the fluid. Such methods are collected under the general heading of pulsed light velocimetry or PLV, and have recently been reviewed by Heselink (1988) and Adrian (1991). Two distinct modes of operation are possible when using seeding particles; in laser speckle velocimetry (LSV) the concentration of scattering particles in the fluid is so large that the images of these particles overlap in the image plane, and result in random interference patterns known as laser speckle, whereas in particle image velocimetry (PIV) the displacement of individual particles is tracked. In both LSV and PIV, the velocity is obtained directly as $\mathbf{v} = \Delta x / \Delta t$ and the accuracy of the technique depends directly on the resolution of the recorded image and of the interrogation method used to analyze it. Modern optical Fourier transform methods are extremely accurate; however, analysis of the data is computationally intensive: a standard personal computer requires approximately 8 hours to process a single PIV frame. For the slow flow of viscous liquids, high densities of seed particles are typically encountered and to date only LSV has been applied to kinematic studies of viscoelastic solutions. Binnington et al. (1983) have used speckle measurements to determine the velocity of a highly elastic polymer solution through an axisymmetric abrupt contraction, and Van de Griend and Denn (1989) have also applied LSV to the study of co-current contraction flows of Newtonian and viscoelastic liquids.

The chief benefit of PIV is that the entire two-dimensional velocity field is sampled at once. This sampling occurs at the random location of each particle; however given a sufficiently large number of samples it is possible to interpolate between the data points to reconstruct velocity profiles similar to those obtained with LDV. It should be noted that the post-processing required for each individual image frame is extensive and such pulsed light techniques can not provide the real-time, point-wise measurements of time-dependent velocity components that is possible with LDV. Close examination of the work of Binnington et al. (1983) and Van der Griend and Denn (1989) also emphasizes that the technique is not readily applied to low velocities experienced near the walls, or to weak recirculating flows. Hence, LDV and LSV should be regarded as complementary techniques for following the time-dependent evolution of complex flow fields.

Interferometry uses flow-induced variations in the refractive index of a fluid to locally retard the phase of the incident light and produce interference fringes resulting from the positive and destructive interference of initially coherent waves. This technique has been used extensively in Schlieren photography to visualize thermal convection patterns and trans-sonic shocks in Newtonian fluids (Van Dyke, 1982). More recently,
interferometry has been used in non-Newtonian fluids by Pennington et al. (1990) to investigate the free drainage of thin viscoelastic films.

Polarimetry methods involve measurements of the polarized state of the electric vector of the light source. Like LDV, polarimetric methods offer the advantages of being noninvasive and being able to obtain kinematic information over highly localized length scales. In addition the response time of optical methods is much faster compared to mechanical measurements, which is beneficial in transient flows. The application of polarimetry to rheological measurements of material properties is made possible by the fact that most large molecules such as polymer chains are optically anisotropic. In an unperturbed assembly of these molecules, Brownian forces ensure that the particles are randomly distributed and the system behaves as an isotropic medium. However, if external forces are applied to fully or partially orient the system, passage of polarized light through the medium will be affected by the anisotropic polarizability of the molecules. Molecular orientation in liquids can be induced by application of electrical fields (Kerr effect), magnetic fields (Cotton-Mouton effect), acoustic waves (Lucas effect), or by fluid flow, as first observed by Maxwell in 1873 (Janeschitz-Kriegl, 1983). This optical anisotropy is usually quantified in polarimetry experiments by the sample's birefringence ($\Delta n'$) and dichroism ($\Delta n''$). These two quantities are defined respectively as the differences in the values of the real and imaginary parts of the refractive index tensor $n = n' - in''$ along the principal axes. The real part $\Delta n'$ results in a change of phase in the electric vector of light, and the imaginary part $\Delta n''$ causes attenuation of the amplitude. A dichroic material, such as a polarizing filter, preferentially attenuates light of a certain orientation by absorbing or scattering the light. However, a birefringent or 'double-refracting' material such as a polymer induces a differential retardation in the phase of the electric vector, since the real part of the refractive index is different parallel and normal to the chain backbone.

For Newtonian liquids, birefringence measurements may be used to determine directly the shear stress and the streamlines in the flow (Prados and Pebbles, 1959). For viscoelastic flow, it is possible to use flow-induced birefringence (FIB) to calculate both the shear stress and principal normal stress difference in a non-Newtonian fluid by independently measuring two parameters, the light intensity and the angle of rotation of the polarization direction. Two different experimental arrangements have been developed for FIB: two-color flow birefringence (TCFB; Chow and Fuller, 1982) uses two different wavelengths of light to measure the two unknowns; whereas, phase-modulated flow birefringence (PMFB; Frattini and Fuller, 1987) relies on a photoelastic modulator to harmonically separate the two unknown quantities. Further details of the principles of
polarimetry and the development of optical rheometry for inhomogeneous flows, suspensions, and liquid crystals can be found in the recent reviews of Fuller (1988, 1990).

A TCFB system with a resolution of approximately 100 µm is being developed in parallel to this thesis research by Quinzani (1991). It will thus be possible to obtain local measurements of both the velocity and stress components in complex flows of viscoelastic liquids. It is to be hoped that detailed comparison of these independent measurements with accurate numerical simulations will reveal the detailed polymer physics that must be captured to enable the future development of design tools that make possible the quantitative simulation of complex industrial polymer processes.

1.4 Thesis Aims

The discussion above has highlighted a number of the outstanding problems that exist within non-Newtonian fluid mechanics and which will be addressed in this thesis. Research focuses on an experimental investigation of the viscoelastic fluid dynamics exhibited by a highly elastic model polymer solution in three of the test configurations depicted in Figure 1.1. These geometries each embody the importance of a particular aspect of non-Newtonian flow which may be summarized as (a) the rotational flow between coaxial parallel disks, (b) entry flow through an axisymmetric abrupt contraction, and (c) stagnation flow around a cylinder constrained in a slit. Literature reviews of previous investigations relevant to viscoelastic flow in each of these geometries are presented individually in Chapter 2. The optical techniques used in the experimental investigations are detailed in Chapter 3, together with design aspects related to the fluids-handling system and the mechanical construction of each flow geometry.

The test fluid used in this thesis is a viscoelastic solution consisting of polyisobutylene (PIB) dissolved in a solution of polybutene (PB) and tetradecane (C14). The fluid is optically clear with an almost constant viscosity and is highly elastic. Such fluids have come to be referred to ‘Boger fluids’ (Boger, 1977/78) and they have been used extensively in earlier experimental studies of viscoelastic flow. It has been stressed above that it is essential to first understand thoroughly the viscoelastic behavior of polymeric fluids in simple one-dimensional flows, if one hopes to interpret quantitatively the material response observed in more complicated two- and three-dimensional flows. To this end, a detailed rheological characterization of the Boger fluid is presented in Chapter 4, together with a critical analysis of the validity of a number of relatively simple constitutive equations in modeling the fluid rheology.
In Chapter 5, the onset of a hydrodynamic instability is demonstrated in the rotational flow of the Boger fluid. This flow instability is driven by the elasticity of the fluid and leads to a transition from a steady two-dimensional flow to a three-dimensional, time-dependent state. Direct visualization of the flow is used to reveal the spatial structure of the unsteady flow and its temporal evolution. Dynamic measurements of the torque and normal force exerted by the fluid serve to illustrate both the capabilities and limitations of globally-averaged measurements in developing an understanding of viscoelastic flow transitions.

Laser Doppler velocimetry (LDV) is used in Chapter 6 to obtain non-invasive measurements of the viscoelastic flow through abrupt axisymmetric contractions. The LDV results are coupled with video-imaging results to associate specific dynamic behavior with the development and evolution of characteristic structures such as a ‘lip vortex’. A sequence of nonlinear, time-dependent flow transitions are documented in the flow together with the sensitive dependence of the observed dynamics on geometric factors such as the contraction ratio and the shape of the entry region. Similar measurements are presented in Chapter 7 for the evolution of the velocity field observed in viscoelastic flow around a circular cylinder. Finally, general conclusions originating from this research relating to the dynamic behavior of viscoelastic flows in complex geometries are discussed in Chapter 8.
Chapter 2

Literature Review

Three separate flow geometries are considered in this thesis; the rotational flow of a viscoelastic liquid between coaxial disks, viscoelastic flow through an axisymmetric abrupt contraction, and the stagnation flow of a viscoelastic fluid past a cylinder constrained in a flat channel. The motivation for choosing these geometries as paradigms characteristic of actual polymer processing applications has been discussed briefly in Chapter 1, and in particular it is noted that the onset of viscoelastic flow transitions has been observed in each of these systems. In this Chapter the flow geometries are discussed individually in greater detail and previous investigations of viscoelastic flow in each of these systems are reviewed.

For clarity this review is divided into individual Sections covering each flow geometry. This is characteristic of research in viscoelastic fluid mechanics, in which efforts have been concentrated on understanding the evolution of the flow structure as $De$ is increased in a particular flow geometry. However, it will be apparent that the flow transitions and nonlinear dynamics observed in each geometry are closely inter-related and an underlying aim of this thesis is the development of experimental techniques which can investigate the dynamic behavior of viscoelastic fluids in any complex geometry.

2.1 Rotational Flows of Viscoelastic Liquids

Rotational or 'swirling' flows of non-Newtonian liquids are important in many technical applications including spin-coating, design of rotating machinery such as centrifuges and extruders, and especially in rheometry. The simplest such flow is driven by
the rotation of two parallel, infinite coaxial disks, and is approximated by the flow generated in the parallel-plate rheometer.

The rotational flow of Newtonian fluids has been studied intensely since the first investigation by von Kármán in 1921. He considered the problem of the flow induced by an infinite rotating disk placed below a semi-infinite medium of fluid initially at rest, and his analysis introduced a similarity transformation which reduced the full system of Navier-Stokes equations to a pair of nonlinear ordinary differential equations in the axial coordinate $z$. Further studies by a number of researchers have extended this analysis to include suction or injection at the disk surface, rotation of the fluid at infinity, and most importantly here, the flow between two infinite parallel, coaxial disks separated by a gap $H$. A clear account of these developments is given in the recent review of Zandbergen and Dijkstra (1987). In the latter instance of two infinite disks, distinct solutions may be found for the cases of motion of either one or both of the disks and for counter- or co-rotation of the disks. In rheometric applications, it is typical to confine the sample between two parallel plates, and then rotate one of the plates and measure the force exerted by the fluid on the other plate. For this case, the velocity field in a Newtonian fluid is then represented in similarity form by

$$\mathbf{u} = (u_r, u_\theta, u_z) = (r\Omega f'(\zeta), r\Omega g(\zeta), -2\Omega f(\zeta))$$

(2.1)

where $\Omega$ is the rotation rate of the plate, $r$ is the radial coordinate, $\zeta = z/H$ is the dimensionless axial coordinate and $f$, $g$ are functions of $\zeta$ that satisfy the boundary conditions on the plates. The prime $'\prime$ indicates differentiation with respect to the axial coordinate.

The form of the von Kármán solution in eq. (2.1) is of considerable practical interest since at finite Reynolds numbers, the axial velocity $u_z$ is found to be independent of the radial coordinate and uniform across the disk. For this reason, rotational flows are extremely useful in the study and development of mass-transfer processes and surface reactions near a single rotating disk, such as those encountered in the spin-coating and subsequent drying of materials from solution. At higher Reynolds numbers ($200 \leq Re \leq 600$), inertial instabilities which lead to the development of spiral vortices have been observed in the rotational flows of Newtonian fluids. These vortices have been visualized in air by Koyabashi et al. (1980) and in water by Clarkson et al. (1980). Laser Doppler velocimetry measurements have also been performed in water by Szeri et al. (1983).

For creeping flow ($Re = 0$), the velocity functions in eq. (2.1) are given simply as $f(\zeta) = 0$ and $g(\zeta) = \zeta$, and the von Kármán solution reduces to a simple viscometric shear
flow with $v = r \Omega z / H$. For highly viscous materials, such as polymer melts, inertial effects are usually very small ($Re \ll 1$) and the flow in a parallel-plate rheometer is closely approximated by this simple one-dimensional velocity field. Secondary flows should be completely negligible and the onset of inertial flow instabilities is not to be expected.

Phan-Thien (1976, 1983) has shown analytically that for the steady laminar flow of a viscoelastic material between parallel rotating disks, both the Maxwell and Oldroyd-B constitutive models have velocity fields of the same von Kármán form as eq. (2.1). Most importantly, however, his calculations did indicate the presence of instabilities in the solution fields which develop at low Reynolds numbers, but at high values of the Deborah number. These instabilities do not arise from inertial effects in the rotating flow, but rather from the elasticity of the fluid. A more detailed discussion of these instabilities is presented below.

In early experimental investigations of rotational flow transitions in viscoelastic fluids, Griffiths et al. (1969) documented a reversal in the direction of the secondary flow of a dilute polyacrylamide solution near a finite rotating disk. Unlike a Newtonian liquid, in which inertial forces drive a radial secondary flow in the outward direction near the rotating disk, elastic normal forces tend to result in an inwardly directed radial flow. This phenomena may be interpreted physically as arising from the development of azimuthal normal stresses (‘hoop stresses’) which result in an inward driving force. Similar arguments can qualitatively explain the well-known Weissenberg effect (Bird et al. 1987a; Lodge et al. 1988) in which the development of normal stresses in swirling flow of a viscoelastic liquid near a rotating rod result in fluid flow up the rod. Further experimental observations of this reversal in the direction of secondary flow have been made by Hill (1972) and the phenomena has also been predicted numerically by Kramer and Johnson (1972).

Laser Doppler velocimetry has been used by Berman and Pasch (1986) to determine the dimensionless velocity functions $f(\zeta)$ and $g(\zeta)$ at high $Re$ for dilute solutions of polyethylene oxide in water in the boundary layer of a single rotating disk. No flow reversal was documented in this investigation; however, some decrease in the boundary layer thickness was documented at low Deborah numbers. At higher $De$ the boundary layer increased in thickness and fluctuations in the velocity field were also observed. These fluctuations were attributed to an instability in the flow field analogous to that predicted by Phan-Thien for an Oldroyd-B fluid model. However, little rheological characterization of the fluid was provided in this work, and therefore the validity of the Oldroyd-B model in describing the rheological properties of the fluid is unknown. In addition, no visualization of the flow field was presented so that the spatial and temporal structure of this unstable

41
flow is also unknown. The connection of these experimental observations to the analysis of Phan-Thien therefore seems tenuous.

The presence of an elastically-driven instability which results in ‘edge-fracture’ of polymer melts has also been observed in viscometric flow between parallel disks (Hutton, 1969; Tanner, 1983). This instability occurs at low rotation rates and appears to arise from surface tension effects and not from the development of a secondary flow. Typically, polymeric fluids such as melts give rise to large normal stresses in shear flow yet possess relatively low surface tension coefficients (Brandup and Immergut, 1975), and, according to Tanner, this disparity leads to the fracture phenomena in the following manner: The shape of the free surface of the material at the edge of the plates is maintained by surface tension forces which do not depend on the rotational speed of the sample, whereas the normal stress differences which drive secondary flows increase rapidly with the rotation rate. This imbalance of forces acting on infinitesimal perturbations of the meniscus ultimately results in the propagation of cracks through the material analogous to those observed in solid fracture mechanics. Dilute polymer solutions have much lower normal stress coefficients than polymer melts, and this precludes the possibility of observing this type of instability with viscoelastic solutions such as ‘Boger fluids’.

The recent development of highly elastic, model polymer solutions known as Boger fluids (Boger, 1977/78) which possess a large constant viscosity has permitted the observation of elastic modifications to the von Kármán rotational flow in the absence of inertial effects — which tend to eject the sample from between the plates at high $\Omega$. In the course of rheological measurements in a parallel-plate rheometer with a highly elastic Boger fluid consisting of polyacrylamide in corn syrup (PAA/CS), Jackson et al. (1984) documented a transition to a time-dependent, shear-thickening or anti-thixotropic behavior in the fluid. They observed that at high shear rates, measurements of the torque and normal force exerted on the plates by the fluid increased steadily over a period of approximately twenty minutes. This time-dependence resulted in an increase in the apparent viscosity of the fluid and a first normal stress difference $N_1$ that exhibited a greater than quadratic dependence on shear rate. Jackson et al. did not associate this anti-thixotropy with the onset of a flow instability, but attributed it to the shear-induced development of structure within the fluid. Similar behavior for other Boger fluid formulations has been remarked on by Binnington and Boger (1986) and has also been reported recently in experiments with the ‘M1 fluid’ (Laun and Hingmann, 1990; Steiert and Wolff, 1990).

Magda and Larson (1988) were the first to associate anti-thixotropic behavior in rheological measurements with the occurrence of a flow instability, and suggested that above a critical shear rate the simple shear flow expected at low rotation rates became
unstable to a more complicated fluid motion. In their experiments, Magda and Larson used a number of different highly elastic Boger fluids prepared from either polyisobutylene (PIB) or polystyrene (PS) in order to study the effects of fluid rheology on the rotational flow transition. By increasing the molecular weight of the polymeric component, they systematically increased the elasticity and relaxation time of the test fluid. At low shear rates the viscometric behavior of each solution was characteristic of that expected for Boger fluids; both the viscosity and first normal stress coefficient remained constant with shear rate, as described by the simple Oldroyd-B differential constitutive equation (Oldroyd, 1950; 1958). Similar rheological behavior is observed for the PIB Boger fluid used in this thesis, and detailed measurements of the material functions and a discussion of the limitations of the Oldroyd-B model are presented in Chapter 4.

Most importantly, the torque and normal force measurements made by Magda and Larson at low shear rates demonstrated no dependence on the duration of the experiment. However, above a certain critical shear rate a transition to anti-thixotropic behavior was observed in the more elastic fluids; a typical response curve for the measured shear-stress and normal stress difference is shown in Figure 2.1(a). Upon inception of steady shear flow, the shear stress and first normal stress difference grew to a plateau and remained constant for up to forty minutes before rapidly increasing at values significantly greater than the initial plateau. This critical shear rate decreased as the elasticity of the fluid increased and also depended inversely on the gap H between the plates. A similar flow transition was reported in the cone-and-plate geometry and the critical shear rate was found to depend on the reciprocal of the cone-angle θ₀. The time-dependent increase in the normal and shear stresses was observed in experiments with two high molecular weight polyisobutlenes (M₆ = 2.7 × 10⁶ g/mol and 4.0 – 6.0 × 10⁶ g/mol) and an extremely high molecular weight polystyrene (M₆ = 18 × 10⁶ g/mol); however, no flow transition was observed in either the cone-plate or parallel-plate flow of the lowest molecular weight PIB solution (M₆ = 1.3 × 10⁶ g/mol). Little explanation of this trend was provided, although it was suggested that shear-thinning effects could be responsible. From these observations, it is not clear a priori whether a similar flow transition will be observed for the Boger fluid used in this thesis; which is a PIB solution that uses a polymer with an experimentally determined mass-average molecular weight of M₆ = 1.8 × 10⁶ g/mol (see Chapter 4.2).

Although inertial transitions in the flow of a Newtonian fluid between parallel rotating disks have received a great deal of experimental and theoretical attention (see the review of Zandbergen and Dijkstra (1987)), much less is known about rotational flow instabilities in viscoelastic liquids. The only analyses relevant for the almost inertialess swirling flows of Boger fluids are the calculations of Phan-Thien for the stability of
Figure 2.1  (a) Time-dependent response of the shear stress $\tau$ (——) and the first normal stress difference $N_1$ (---) measured during rotational flow of a PIB/PB Boger fluid ($\bar{M}_w = 2.6 \times 10^6$ g/mol.) between parallel plates at $De = 7$. (Reproduced from Magda and Larson, 1988).

(b) Calculated time-dependent response of the radial pressure gradient as a function of $De$, for rotational shear flow of an Oldroyd-B fluid between infinite parallel coaxial disks at $Re = 1$. (Reproduced from Phan-Thien, 1983).
rotational shear flows of an Oldroyd-B model (Phan-Thien, 1983; 1985). The analysis for flow between parallel-plates uses the von Kármán similarity forms for the velocity and stress fields that are valid when the disks are infinite in extent and examines the linear stability of the rotational shear flow to disturbances that can also be represented in the similarity form. Phan-Thien (1983) found that for creeping flow ($Re = 0$) between rotating coaxial disks, these disturbances grow exponentially in time for values of the Deborah number above the critical value of

$$De_c^{(pp)} = \lambda_1 \Omega_c = \frac{\pi}{\sqrt{(1 - \Lambda)(5 - 2\Lambda)}}$$  \hspace{1cm} (2.2)

where $\lambda_1$ is the relaxation time, and $\Lambda = \eta_s / \eta_0$ is the ratio of the solvent viscosity to the total viscosity in the Oldroyd-B model. A similar linear stability analysis for viscometric flow of an Oldroyd-B fluid between an infinitely large cone and plate (Phan-Thien, 1985) found that this flow was unstable to infinitesimal disturbances above a critical rotation rate $\Omega_c$ given by

$$De_c^{(cr)} = \lambda_1 \Omega_c = \pi \sqrt{\frac{2}{5(1 - \Lambda)}}$$  \hspace{1cm} (2.3)

For a Newtonian fluid, $\Lambda = 1$ and the viscometric flow in both geometries is predicted to be stable at all rotation rates. It should be noted that in the original work of Phan-Thien, the dimensionless quantity $(\lambda_1 \Omega_c)$ was identified as a critical Weissenberg number; however, in this thesis it is referred to as a critical Deborah number. The important reasons for making this distinction are discussed in detail in Section 5.2.

In addition to linear stability predictions, Phan-Thien performed direct numerical simulations of the rotational shear flow of an Oldroyd-B fluid between parallel plates at finite $Re$. Typical results for the radial pressure gradient $\partial P / \partial r$ calculated during start-up of rotational shear flow are shown in Figure 2.1(b) for $Re = 1$, $\Lambda = 0.5$. At low Deborah numbers the pressure gradient shows a small initial maximum and then decreases monotonically towards a steady value given by $(3/10) Re - \Lambda De$; however, above the critical value of $De_c^{(pp)}$ given approximately by eq. (2.2), the gradient is observed to approach an initially constant value before rapidly decreasing again.

Very recently, Ji et al. (1990) have presented full numerical solutions for the high Reynolds number rotational flow of an Oldroyd-B fluid over a wide range of $\Lambda$ and $De$. Sample results of these calculations are reproduced in Figure 2.2. The evolution of the torque exerted by the fluid on the plates was calculated for increasing $De$ using arc-length
Figure 2.2  (a) Dimensionless torque exerted on the top and bottom plates as a function of $De$ for rotational flow of an Oldroyd-B fluid model with $\Lambda = 0, 0.5, 1$. (b) Two separate solution families for the dimensionless radial velocity profiles between the two plates. The solid line (-----) represents the antisymmetric solution obtained below the turning point, the dashed lines indicate non-symmetric solutions beyond the turning point; (- - -) $\Lambda = 0$, (- - -) $\Lambda = 0.5$. (Both figures reproduced from Ji et al., 1990).
continuation techniques and is plotted in Figure 2.2(a) for three values of $\Lambda = 0, 0.5,$ and 1. The presence of a turning point which corresponds to the value $De_c^{(PP)}$ obtained by Phan-Thien is clearly observable in each calculation. For Deborah numbers numerically greater than this value it is not possible to find a solution to the problem that can be represented in the von Kármán similarity form, whereas below the turning point, two distinct solution families exist. The two solutions of dimensionless radial velocity function $(v_r/r\Omega) \equiv f'(\zeta)$ at $De = 4$ are shown in Figure 2.2(b) as a function of the axial position $\zeta = z/H$ between the plates for solvent ratios of $\Lambda = 0$ and 0.5. The solid lines indicate the steady-state solution obtained before reaching the turning point. The velocity is antisymmetric about the midpoint $\zeta = 0.5$ and inwardly directed near the top rotating plate. The dashed lines indicate the steady-state radial velocity profiles calculated beyond the turning point. Since the calculations assume a similarity form, the solution is still axisymmetric, however it can be seen that the velocity is no longer antisymmetric about the midpoint (i.e. $f'(\zeta) \neq 0$ at $\zeta = 0.5$). Examination of the other velocity components reveals similar trends, especially in the azimuthal component $v_\theta$ which no longer exhibits a maximum velocity at the rotating plate; i.e. the fluid in the body of the sample is rotating faster than the plate driving the flow. These observations coupled with the principle of exchange of stability (Iooss and Joseph, 1980) indicate that the steady-state profiles obtained beyond the limit point are, in fact, unstable aphysical solutions. An important point to note from the calculations of Ji et al. (1990) is that no solution of the von Kármán similarity form could be found beyond the turning point at $De_c^{(PP)}$. Of course, this does not preclude the existence of other solutions which may be time-dependent or non-axisymmetric, or the possibility that the solution curve in Figure 2.2(a) exhibits another turning point beyond the range of parameter space explored by Ji et al.

Magda and Larson (1988) performed dynamic rheological measurements to determine both the zero-shear-rate viscosity $\eta_0$ of each Bo{ger} fluid and an estimate for the solvent contribution $\eta_s$ to the total viscosity. From these measurements the parameter $\Lambda$ was determined for each fluid and used to calculate the critical Deborah number according to equations (2.2) and (2.3) for onset of unstable flow. These predictions were compared with the shear rates and Deborah numbers actually required to give rise to anti-thixotropic behavior in the cone-and-plate and parallel-plate experiments. With a judicious choice of relaxation time, agreement to within 50% was obtained between the experimental measurements and the theoretical values of $De_c$ predicted by Phan-Thien. The inverse dependence of the critical shear rate $\gamma_c$ on the plate separation $H$ and the cone-angle $\theta_0$ observed by Magda and Larson is also consistent with Phan-Thien’s analysis which predicts an instability determined solely by the rotation rate $\Omega$, not the local shear rate $\gamma$.  

47
The recent work of Laun and Hingmann (1990) and Steiert and Wolff (1990) has corroborated these observations for another Boger fluid consisting of PIB dissolved in polybutene and kerosene. In these later publications, however, the transition has been interpreted as resulting from a critical shear stress \( \tau_c \) in the fluid, rather than in the terms of a kinematic instability developing above a critical Deborah number \( De_c \). This leads to a stability criterion which varies directly with the geometric dimensions of the rheometer used to study the flow transition. In Section 5.4.4 we show how these recent measurements can be re-interpreted in a consistent framework by calculating the critical Deborah number in each flow geometry.

The work of Magda and Larson has been fundamental in identifying the role of this rotational flow instability in causing this so-called anti-thixotropic behavior of Boger fluids. It has also demonstrated that the flow instability does not depend on specific macromolecular interactions, since similar results were obtained with both PIB and PS polymer systems. However, this paper did not provide any details of the spatial and temporal structure associated with the flow that develops consequent to the onset of the instability. The inertialess linear stability analysis of Phan-Thien considers axisymmetric perturbations to the velocity and stress fields which have the functional form of a normal mode analysis in the axial coordinate, and therefore disturbances are represented as sinusoidal perturbations of the form \( \sin(2 \pi k z / H) \). The linearly unstable disturbances that correspond to the critical values given by eqs. (2.2) and (2.3) arise from the \textit{first} mode \( (k = 1) \) of this analysis; and, for example, the spatial form of the unstable radial velocity component is expressed as

\[ v_r = r \Omega [f'(\zeta) + \epsilon \sin(2 \pi \zeta)] \quad (2.4) \]

Disturbances corresponding to higher axial wave-numbers \( (k > 1) \) remain stable at these values of \( De \), and are subsequently found to become unstable to infinitesimal perturbations at higher Deborah numbers corresponding to \( De_c(k) = k De_c(1) \). Therefore, the linear stability analyses predict that the most dangerous disturbance (\textit{i.e.} the one occurring at the lowest \( De \)) leads to a single toroidal recirculation superimposed on the base shear flow, as shown by the additional radial inflow induced near the top rotating disk in Figure 2.2(b). In addition, these disturbances obey the principle of exchange of stability (Iooss and Joseph, 1980), so that as \( De \) is increased to values greater than \( De_c \) the base solution becomes unstable and the flow transition is expected to generate a new steady, axisymmetric flow with a single roll-cell. Such a flow transition should be readily observable in visualization investigations of rotational flow between coaxial disks. Of
course, this prediction is subject to the important caveat that the analysis did not include all possible spatial and temporal disturbances. To do this requires the inclusion of general, three-dimensional perturbations which do not have the similarity form.

The aim of the rotational flow experiments described in Chapter 5 of this thesis is to probe in greater detail the structure of the flow transitions that result in apparent anti-thixotropic behavior in the cone-and-plate and parallel-plate flows of a well-characterized PIB/PB Boger fluid. The polyisobutylene polymer used in the solution is of moderate molecular weight, and the fluid is considerably less elastic than the test fluids employed by Magda and Larson. We show that the onset of these instabilities is governed primarily by the rotation rate $\Omega$, and also occurs reasonably close to the critical Deborah numbers predicted by eqs.\,(2.2) and (2.3). Analysis of the shear stress and normal stress as a function of time demonstrates the initial exponential growth of the unstable mode; however, our measurements indicate the presence of hysteresis in the stress–shear-rate curve, corresponding to a subcritical bifurcation. In addition, the spectral analysis and video-imaging results reported in Chapter 5 show that the instability is nonaxisymmetric and ultimately leads to the development of a three-dimensional, time-dependent flow — in sharp contrast to the stability analysis of Phan-Thien.

2.2 Viscoelastic Flow Through An Abrupt Axisymmetric Contraction

The basic geometry for flow through an axisymmetric abrupt contraction is shown in Figure 2.3, and consists of a large upstream tube of radius $R_1$ which abruptly contracts to a smaller tube with radius $R_2$. The fluid approaching the contraction accelerates from a fully developed upstream profile to assume another fully developed profile at some distance downstream of the contraction plane. A recirculating secondary flow may develop in the corners of the large tube immediately upstream of the contraction plane, depending on the flow rate and fluid rheology. Such entry flows contain both shearing and extensional components and are encountered commonly in many commercially important polymer processing applications, such as extrusion, injection-molding or fiber-spinning. The relative importance of each deformation component is varied by changing the contraction ratio, defined as $\beta \equiv R_1/R_2$. For low contraction ratios, relatively high shear rates are
Figure 2.3  Schematic diagram of the axisymmetric contraction geometry. The contraction ratio is defined as $\beta = R_1 / R_2$ and the dimensionless vortex reattachment length is $\chi = H_V / 2R_1$. 
experienced upstream and the total extensional strain is relatively low, whereas in geometries with higher contraction ratios, the rate of shearing deformation in the large upstream tube is extremely low but the extensional strain and orientation experienced by the polymer molecules as they accelerate into the small downstream tube is increased.

The fluid mechanics of contraction flows is one of the most thoroughly studied experimental systems for complex viscoelastic motion and has been reviewed in detail by Boger (1982, 1987) and by White et al. (1987). A myriad of interesting flow phenomena have been observed with increasing Deborah number: As the flow rate through the contraction is increased, a viscoelastic fluid undergoes transitions from the low flow rate, Newtonian-like behavior to new regimes which may exhibit greatly enhanced vortex size or streamlines that diverge away from the centerline at some distance above the contraction plane. At high flow rates the large vortex observed in many viscoelastic entry flows ultimately becomes unstable, resulting in large fluctuations in the flow field and gross distortion of the viscoelastic material downstream of the contraction, as shown previously in Figure 1.3. These previous investigations have yielded the first glimpses of the rich nonlinear structure of contraction flows and have demonstrated the sensitive dependence of the observed flow transitions on the following parameters: the contraction ratio, the viscoelastic fluid rheology for both polymer melts and solutions, the details of the flow geometry including whether it is planar or axisymmetric, and the precise shape of the contraction lip, i.e. whether it has a sharp entrance or is rounded.

The formation and subsequent development of the secondary flows in this geometry are so pronounced that the flow through an axisymmetric contraction has been made a benchmark problem for numerical simulations of viscoelastic flows (Hassager, 1988). Unfortunately the violent effects on the velocity and stress fields caused by rapid acceleration of the fluid through the contraction, especially near the re-entrant corner, make this problem extremely difficult, as evidenced by the comparison shown previously in Figure 1.2. Reliable numerical results for the entry flows of viscoelastic fluids at moderate Deborah numbers are only just beginning to appear (Marchal & Crochet, 1987; Coates et al., 1991).

As mentioned above, the diversity of viscoelastic phenomena observed in entry flows of non-Newtonian fluids is found to depend intimately on the rheology of the material being studied. For this reason, the ensuing survey of the contraction flow literature is subdivided into sections which briefly review the evolution of the flow structure and stability that is observed in (i) Newtonian and inelastic fluids, (ii) shear-thinning viscoelastic solutions, and finally, (iii) a more detailed discussion of highly elastic liquids such as Boger fluids. Entry flows of polymer melts are not considered directly in this
thesis; however, a comprehensive review of early work is given by White (1973) and a
discourse on the similarities and important differences observed between contraction flows
of polymer melts and polymer solutions can be found in the review of White et al. (1987).

2.2.1 Entry Flows of Newtonian Fluids and Inelastic Liquids

The creeping flow of Newtonian fluids through an abrupt axisymmetric contraction
as shown in Figure 2.3 has been intensely studied both experimentally and numerically,
and can now be considered as essentially a solved problem. A more detailed review of the
historical developments and the approximate boundary layer solutions obtained to the
problem are given by Boger (1982), and here we will concentrate only on the evolution of
the flow structure that is observed as the flow rate through the contraction is increased and
fluid inertia becomes important. Inertial effects in the flow are measured by the Reynolds
number $Re$, which is defined in terms of the fluid properties and downstream flow
conditions as $Re = 2\rho UR_2/\mu$, where $U$ is a characteristic velocity in the small tube and $R_2$
is the tube radius. A number of correlations have been derived from experiments and
verified by numerical computations which accurately predict the evolution of important
flow parameters such as the excess pressure drop, the entry length, and the size of the
vortex in the outer corner of the tube. For example, the entry length $L_e$ is defined as the
distance from the contraction plane into the downstream tube that the fluid travels before the
centerline velocity has attained 99% of its fully-developed value. For contraction ratios of
$\beta \geq 2$, and Reynolds numbers of $Re < 500$, the entry length for Newtonian fluids has been
calculated by Christiansen et al. (1972) and Ventras and Duda (1973) to be given by

$$L_e/R_2 = 0.49 + 0.11 \, Re$$

(2.5)

The effect of fluid inertia is thus to increase the length $L_e$ required for the flow to develop a
fully developed downstream structure.

The most important characteristic observed in the creeping flow ($Re << 1$) of a
Newtonian liquid through an abrupt contraction is the weak recirculation observed in the
outer corner of the upstream tube and shown schematically in Figure 2.3. The size of this
corner vortex is characterized by the vortex reattachment length defined as $\chi = H_V/2R_1$,
where $H_V$ is the vertical height of the vortex and $2R_1$ is the diameter of the upstream tube.
This corner vortex is strictly a result of the Newtonian stresses caused by the kinematical
constraints of the corner, and its presence is predicted by applying the similarity solution of
Moffat (1964) to analysis of the local flow in this region. However, this analysis is restricted to the local flow near the corner and does not predict a characteristic size for the eddy that develops. In experiments for creeping flow of a Newtonian fluid through axisymmetric contractions, the vortex length has been observed (Nguyen and Boger, 1979) and identically predicted (Viriyayuthakorn and Caswell, 1980) to be $\chi = 0.17 \pm 0.01$. At higher Reynolds numbers of $Re \geq 1$, fluid inertia forces the weak recirculation into the outer corner, and the size and strength of the vortex are found to be reduced in size. This effect can be observed in Figure 2.4(a), which shows the excellent agreement between a large number of experimental measurements (Nguyen, 1978; Boger et al. 1986) and the finite-element simulations of Kim-E et al. (1983). At $Re = 100$, these results indicate that the vortex height has decreased to $\chi = 0.05$.

Laser Doppler velocimetry measurements of the kinematics in Newtonian entry flows have been carried out by Burke and Berman (1969) for Reynolds numbers in the range $65 < Re < 400$. This study revealed the evolution of the axial velocity profile at the contraction plane from almost fully developed at low $Re$ to almost flat at high $Re$ together with the development of off-centerline maxima in the velocity. This inertial phenomena has been predicted qualitatively in the early calculations of Christiansen et al. (1972) and more recently a quantitative fit to the LDV data of Burke and Berman has been possible with the finite element solution of Kim-E et al. (1983). Similar detailed LDV measurements of the development of the Newtonian velocity field throughout a planar contraction have also been reported recently by Wunderlich et al. (1988).

In the axisymmetric contraction flow of inelastic or 'power-law' fluids, another parameter becomes important — a fluid viscosity that is a nonlinear function of the local shear-rate in the flow. In reality it is not possible to find an experimental fluid that exhibits a shear-thinning viscosity without also possessing some elasticity, albeit small (see for example; Binding and Walters, 1988; Dhahir and Walters, 1989). Research in this area has thus been concentrated on theoretical predictions which can address in isolation the effects of shear thinning on the kinematics observed in entry flows. More detailed discussions of the modelling of inelastic fluids together with predictions for the flow characteristics such as entry pressure drop and vortex size may be found in the comprehensive study of Kim-E et al. (1983) and the reviews of Boger (1982, 1987). In these works it is shown that the principal consequences of a shear-rate-dependent viscosity are similar to the inertial effects observed at high Reynolds numbers, and that shear-thinning results in an increase in the magnitude of the entrance pressure drop and a decrease in the size of the corner vortex. This effect is clearly illustrated by the finite element simulations of Coates (Raiford et al., 1989) shown in Figure 2.4(b) which indicate a significant reduction in the size and strength
Figure 2.4  (a) Evolution of the vortex reattachment length $\chi$ with increasing Reynolds number for the flow of Newtonian liquids through axisymmetric contractions. (Reproduced from Boger et al., 1986)

(b) Numerically calculated streamlines for flow of a 'Carreau-Yasuda' inelastic fluid model through a 4:1 axisymmetric contraction at $Re = 0$ and $Re = 80$ (Reproduced from Raiford et al., 1989).
of the corner vortex calculated with an inelastic Carreau-Yasuda model (Bird et al., 1987a) at \( Re = 0 \) and \( Re = 80 \).

### 2.2.2 Contraction Flows of Shear-Thinning Viscoelastic Solutions

A large number of previous experimental investigations have considered entry flows of polymer solutions through both planar and axisymmetric contractions in order to understand the modifications to the velocity field introduced by fluid viscoelasticity. Such dilute or semi-dilute polymer solutions typically consist of small quantities (0.05 – 5.0 wt\%) of a high molecular weight polymer such as polyacrylamide (PAC), polyisobutylene (PIB) or polyethylene oxide (PEO) dissolved in an appropriate low viscosity Newtonian solvent. The resulting viscoelastic solutions display both a shear-thinning viscosity and a shear-rate-dependent first normal stress coefficient and can be modelled accurately by multi-mode nonlinear constitutive equations (Quinzani et al., 1990), or by empirical expressions such as the Carreau-Yasuda or Ellis equations (Bird et al., 1987a).

The most comprehensive study to date of the kinematics observed in axisymmetric contraction flows is the work of Cable and Boger (1978a,b; 1979). In these publications, the vortex characteristics, velocity field and stability of the flow of aqueous PAC solutions in 2:1 and 4:1 axisymmetric contractions are presented over a wide range of flow rates. The importance of inertial and elastic effects at a given flow rate are assessed by calculating the Reynolds number and Deborah number of the flow in terms of shear-rate-dependent rheological properties at the characteristic downstream flow conditions. For very low flow rates \( (Re \ll 1, De \ll 1) \) the streamlines visualized by streak photography were identical to those observed in the creeping flow of a Newtonian fluid through an axisymmetric contraction, with a dimensionless vortex reattachment length of \( \chi = 0.17 \), in good agreement with Figure 2.4. As the flow rate was increased, this vortex was observed to rapidly increase in size as shown in the streak photograph reproduced in Figure 2.5(a) at flow conditions of \( Re = 62, De = 0.53 \). The flow is from top to bottom, and the vortex height is calculated as \( \chi = 0.85 \). Similar enhancement of the corner vortex has been observed in other shear-thinning viscoelastic solutions (Rama Murthy, 1974) and in low density polyethylene melts (Ballenger and White, 1971). However, measurements in Newtonian fluids and calculations with inelastic models predict that at \( Re = 62 \) inertial forces and/or shear-thinning effects should have substantially reduced the vortex size to \( \chi \approx 0.12 \), and Cable and Boger thus concluded that this vortex growth regime arises directly from viscoelastic effects in the entry flow region.
Figure 2.5 Flow visualization photographs for flow of a shear-thinning PAC/water solution in a 4:1 axisymmetric contraction showing (a) vortex growth at $Re = 62$, $De = 0.53$ and (b) diverging streamlines at $Re = 165$, $De = 0.653$. (*Reproduced from Cable and Boger, 1978a*).
Observations of this large corner vortex indicated that it exhibited a maximum size of \( \chi = 0.90 \), and at higher flow rates began to decrease in size. Concurrent with this decrease, streak photographs revealed that the streamlines upstream of the contraction plane began to diverge away from the centerline as shown in Figure 2.5(b). Similar effects at high \( Re \) had been previously observed by Rama Murthy and Boger (1972), and this *diverging flow regime* was attributed to inertial effects in the flow. Cable and Boger (1978b) were able to obtain semiquantitative measurements of the axial velocity in this diverging flow by measuring the lengths of particle streaks obtained with strobe photography, and they demonstrated that this diverging flow regime resulted in the development of two symmetric off-center maxima in the velocity profiles. Detailed LDV measurements of this diverging flow effect have recently been obtained with dilute PAA solutions in both axisymmetric and planar contractions (Yoganathan and Yarlagadda, 1984; Wunderlich et al., 1988).

At still higher flow rates, Cable and Boger documented an instability in the diverging flow upstream of the contraction which resulted in the formation of an asymmetric vortex with a vortex length that was a function of the angular coordinate \( \theta \). This vortex was observed to rotate azimuthally around the tube with a regular frequency \( f \sim 0.3 \text{ Hz} \). Similar spiralling oscillations with a frequency \( f \sim 0.5 \text{ Hz} \) have also been documented by Yoganathan and Yarlagadda (1984). From a series of measurements with a number of different, shear-thinning fluid formulations in both 2:1 and 4:1 contractions, Cable and Boger proposed a stability criterion for the onset of unstable flow in terms of a critical stress ratio given by

\[
\frac{N_1}{\tau}_{crit} = 5 + 0.07Re' \tag{2.6}
\]

where \( Re' \) is the generalized Reynolds number defined in terms of the shear-thinning power-law parameters \( K, n \) as \( Re = \rho(2R)^nU^{2-n}/K \).

Inertial effects in contraction flow are thus observed to stabilize the flow and lead to higher critical stresses at the onset of unstable flow. This inertial increase in stability has also been documented in experimental and theoretical analyses of Taylor-Couette flows of dilute polymer solutions (Beavers and Joseph, 1974; Larson, 1989).

In addition to the spiralling flow instability, Rama Murthy (1974) has documented further transitions in axisymmetric contraction flows of dilute PAC solutions. At shear rates of \( \gamma = 2000 \text{ s}^{-1} \) (corresponding to \( Re = 138 \)) the azimuthal swirling vortex no longer shows regular periodic oscillations, but instead exhibits large-amplitude irregular
fluctuations that were described by Rama Murthy as ‘chaotic’. At extremely high flow rates of $\gamma = 5000 \, \text{s}^{-1}$ ($Re = 330$) a further transition resulted in a return to a steady flow regime consisting of an extremely large corner recirculation and a narrow primary inlet stream of almost the same diameter as the downstream tube. Finally, it is noted that recent flow visualization experiments with a shear-thinning PAC solution (Chiba et al., 1990) have documented an instability in flow through a 4:1 planar contraction that results in a transition from simple two-dimensional flow to a complex three-dimensional motion. This transition developed at moderate flow rates ($Re \sim 50$) and the resulting flow was observed to consist of a roughly periodic cellular structure characterized by alternating regions of either an enhanced corner vortex or a divergent flow profile.

More recently, extensive streak photography experiments by Evans and Walters (1986; 1989) have revealed the sensitive dependence of the observed vortex characteristics on the contraction ratio $\beta$ and the precise rheology of the test fluid. For the flow of an 0.5 wt% PAC/water solution through a moderate contraction ratio ($\beta = 4$), the evolution of the flow structure was similar to that documented by Cable and Boger with vortex growth followed by the development of diverging flow. However, by varying the polymer concentration or the contraction ratio a different mechanism for vortex enhancement was observed: as the flow rate through geometries with large contraction ratios ($\beta = 16$ and $\beta = 80$) was increased, the weak Moffatt eddy in the corner expanded inwards, and a second independent vortex also appeared near the re-entrant corner joining the upstream and downstream tubes. This lip vortex subsequently expanded outwards enveloping the corner vortex until it covered the base of the upstream tube. By decreasing the concentration of the PAC to 0.2 wt%, Evans and Walters (1989) varied the fluid rheology so that the elasticity of the fluid was reduced and shear-thinning effects became more important at high shear rates. In this manner a lip vortex could also be produced in the 4:1 contraction and a sequence of photographs with increasing volumetric throughput documented the initial growth of the elastic vortex at moderate flow rates followed by the subsequent inertially-driven collapse of the enhanced corner vortex to form a small intense vortex attached to the lip of the contraction.

A similar coexistence of a corner vortex and a lip vortex was observed visually by Raiford et al. (1989) for flow of a 5.0 wt% PIB/C14 solution through 2:1, 4:1 and 8:1 axisymmetric contractions. This publication also presented LDV measurements that documented the sensitive dependence of the velocity field on $\beta$ and $De$: At high flow rates, the LDV data revealed that instead of developing off-center maxima in the axial velocity profiles near the contraction – as would be observed in inelastic fluids – the interaction of shear-thinning and viscoelasticity result in an extra acceleration of the inner ‘core’ region of
the fluid. Similar results indicating a substantial acceleration in the axial velocity near the centerline have also been obtained recently in numerical simulations with the shear-thinning Phan-Thien–Tanner viscoelastic constitutive model (Debbaut et al., 1988).

The precise conditions that give rise to this lip vortex phenomenon are not known; however, the sensitive dependence on contraction ratio suggests that the interplay between the shear and elongational rheological properties of the test fluid are important. As discussed earlier, the magnitude of the shearing and shear-free components of the flow vary with contraction ratio. For Newtonian and inelastic fluids, experiments and calculations reveal that flow characteristics such as entry length and vortex size may depend on inertial effects and shear-thinning effects but are independent of the contraction ratio for \( \beta \geq 4 \). Similarly, the viscometric properties of these fluids are either constant (for Newtonian fluids) or depend solely on the shear rate (for inelastic ‘power-law’ fluids), and the extensional viscosity \( \bar{\eta} \) of the fluid is given directly by Trouton’s law (Bird et al. 1987a) as \( \bar{\eta} = 3\eta \). However, experimental measurements of the material functions of dilute polymer solutions show that the viscosity displays shear-thinning behavior, whereas the extensional viscosity generally exhibits significant strain-rate thickening in both planar and uniaxial extensional flows (Jones et al., 1987; Fuller et al., 1987). Further details of modelling the extensional properties of viscoelastic liquids can be found in Section 4.4.4 and also in the publication of Quinzani et al. (1990).

In order to investigate the importance of extensional effects by numerical simulation, Debbaut and Crochet (1988; Debbaut, 1990) have empirically formulated modified inelastic and viscoelastic models that predict a tension-thickening in the extensional viscosity. Their calculations have been able to predict the evolution and coexistence of the corner vortex and the elastic lip vortex in a 4:1 axisymmetric contraction, and these models have proved useful in elucidating the interaction of inertial effects and extensional-thickening in the entry region. However, it is important to note that the extensional-thickening behavior formulated in these models occurs only in axisymmetric geometries, and the models cannot predict the development of a lip vortex in a planar geometry, whereas the work of Evans and Walters (1986, 1989) has unequivocally shown that lip vortices form in both planar and axisymmetric contraction flows of dilute polymer solutions.

Evans and Walters also examined the effects of modifying the shape of the re-entrant corner on the structure of the flow observed in planar contractions. Various modifications were considered including ramps, 45° notches and smoothly rounding the corner with a uniform radius of curvature. Typical results are shown in Figure 2.6(a) – (c) at three different flow rates for a planar contraction geometry with a smoothly curved lip
Figure 2.6 Evolution of the corner vortex in flow of an 0.2 wt% PAC/water solution through a 4:1 planar contraction. The left hand side of the entrance has a sharp 90° re-entrant corner, the right-hand side has a smooth radius of curvature $R = 0.8R_2$. (a) Flow rate $Q = 2.50$ cm$^3$/s; (b) $Q = 15.15$ cm$^3$/s; (c) $Q = 22.88$ cm$^3$/s (Reproduced from Evans & Walters, 1989).
entrance of radius \( R = 0.8R_2 \) on one side and a sharp 90° re-entrant corner \( (R = 0) \) on the other. Although Evans and Walters did not calculate Deborah numbers or Reynolds numbers for the flow visualization pictures they presented, both \( De \) and \( Re \) also increase in Figure 2.6 from (a) to (c). As the Deborah number is increased, vortex enhancement can be observed near the sharp lip and the corner recirculation increases in intensity and size as first documented by Cable and Bober. The primary consequence of smoothing the lip entrance is a significant reduction in the size of the corner vortex observed in the outer corner of the upstream flow. Evans and Walters also observed that smoothing the lip corner enhanced the stability of the flow by shifting the onset of unstable flow to higher flow rates. Unfortunately, the authors did not draw any conclusions from these results except to note the sensitive dependence of contraction flow results on the precise shape of the lip geometry. One of the goals of the LDV studies carried out in this thesis has been to address in more detail the role of the re-entrant corner on the nonlinear dynamics and stability of viscoelastic flow through axisymmetric contractions. These results are presented in Chapter 6.4.

2.2.3 Contraction Flows of Bober Fluids

The experimental observations discussed above in Sections 2.2.1 and 2.2.2 have demonstrated that inertia, shear-thinning and viscoelasticity each have pronounced effects on the evolution of the flow structure and the stability of contraction flows. This complex interaction between three competing phenomena makes an unambiguous interpretation of the data extremely difficult. In addition, the range of parameter space accessible in experiments with dilute polymer solutions is generally not typical of the operating regimes characteristic of actual polymer processing applications: the high viscosities of polymer melts results in extremely low Reynolds numbers in the flow, whereas the large molecular weight and high entanglement density of the melt leads to significant elastic effects in the flow and high values of the Deborah number. The relatively small relaxation times and low viscosities of polymer solutions, together with their pronounced shear-thinning characteristics, however, tend to result in flows at high \( Re \) and low \( De \). Even though entry flows of polymer melts are creeping flows \( (Re << 1) \), a wide range of non-Newtonian phenomena (which depend intimately on the rheology and shape of the die entry) have been documented in such contraction flows, and the flow becomes unstable at very low volumetric flow-rates (White, 1973; Petrie and Denn, 1976). The effects of this flow instability have already been demonstrated in Figure 1.3, which shows the development of
oscillations in the extrudate of a PDMS gum from an axisymmetric contraction at a moderate De corresponding to a maximum Reynolds number of Re ~ 10^-2 (Piau et al., 1988). This is approximately three orders of magnitude lower than the values of Re documented by Cable and Boger for unstable flows of shear-thinning polymer solutions.

In order to alleviate this discrepancy, Boger (1977/78; Boger and Nguyêñ, 1978) developed a new class of viscoelastic polymer solutions which more closely represent the rheological behavior of polymer melts, while retaining the experimental flexibility of polymer solutions. These ‘Boger fluids’ consist of a dilute or semi-dilute solution of a high molecular weight polymer dissolved in a small amount of a low viscosity solvent and then added to a highly viscous polymeric solvent. A detailed discussion of the rheology and modelling of Boger fluids is presented in Chapter 4; however, these solutions may be simply described here as highly elastic fluids with a large viscosity that remains almost constant across several decades of shear rate. Boger fluids are thus able to serve as model experimental fluids which more closely approximate polymer melts and allow the investigation of elastic effects in complex flows in the absence of additional complicating phenomena such as shear-thinning or inertial effects. In addition, the relatively simple viscometric properties of Boger fluids allows the direct comparison of experimental observations with both theoretical analyses of flow stability (as described in Section 2.1) and direct numerical simulations of viscoelastic flow in complex geometries (Walters and Webster, 1982; Binding et al. 1987).

In this thesis, a Boger fluid is used to allow a detailed investigation of the effects of fluid elasticity on the stability and flow characteristics observed in axisymmetric contractions, and to examine the sensitive dependence of these properties on the contraction ratio β and the precise shape of the lip entrance. A number of previous researchers have studied contraction flows of Boger fluids; these experimental investigations are summarized in Table 2.1 and discussed in detail below.

The earliest investigations of the flow of Boger fluids through axisymmetric contractions were made by Nguyêñ and Boger (1978, 1979; Boger, 1980) using a polyacrylamide/glucose (PAC/G) fluid formulation in a range of contraction ratios from 4.08 ≤ β ≤ 14.6. These flow visualization experiments revealed the onset of flow instabilities at high De, but low Re, which appeared qualitatively similar to the observations described above in Section 2.2.2 for shear-thinning viscoelastic fluids. The results obtained for flow through a 7.675:1 axisymmetric contraction are reproduced in Figure 2.7, and are described in some detail here because of their relevance to the experiments presented later in Chapter 6. At low Deborah numbers (De << 1) the flow converges radially towards the die entrance and a small recirculation is observed near the contraction plane in Figure 2.7(a).
<table>
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<th>Geometry</th>
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<td>1988</td>
<td>Axisymmetric</td>
<td>$\beta = 2, 4, 8$</td>
<td>PIB/PB/C14</td>
</tr>
<tr>
<td>Boger &amp; Binnington</td>
<td>1990</td>
<td>Axisymmetric</td>
<td>$\beta = 4, 22$</td>
<td>PIB/PB/kerosene</td>
</tr>
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Table 2.1 Previous experimental investigations of viscoelastic entry flows using highly elastic Boger fluids.
Nguyêñ and Boger (1979) concluded that this fluid motion was essentially Newtonian and the size of this Moffat eddy, as measured by the vortex reattachment length $\chi$, was found to agree very well with the size determined from creeping flow experiments in aqueous polymer solutions (Cable and Boger, 1978a) and calculations for a Newtonian fluid at low $Re$ (Kim-E et al. 1983). As the flow rate, and Deborah number, of the flow is increased this vortex dramatically increases in size and extends upstream, as shown in Figure 2.7(b) and (c). At a Deborah number of $De = 2.08$ this large vortex was found to become asymmetric as shown in Figure 2.7(d). Increasing the Deborah number further caused the flow to become time-dependent and the asymmetric vortex was observed to rotate azimuthally around the upstream tube with a constant frequency. At $De = 2.25$ the flow disturbance became even more severe and the large asymmetric vortex pulsated in height periodically, as shown in the sequence of photographs in Figure 2.7(e) – (h). The fluid particles were observed to follow a helical path as they spiralled into the downstream tube. Similar flow patterns have been observed in the entry regions of commercial extrusion dies and result in helical oscillations in the shape of the extrudate (Ballenger and White, 1971). It thus appears that the introduction of Boger fluids has been successful in emulating the creeping flow instabilities of highly elastic polymer melts.

Further experiments reveal, however, that the situation is actually considerably more complicated than was initially thought. The qualitative flow visualization work of Walters and co-workers (Walters and Rawlinson, 1982; Walters and Webster, 1982; Walters, 1985) has been instrumental in demonstrating that although significant vortex growth is observed in the flow of Boger fluids through axisymmetric contractions and geometries with a square cross-section, no vortex enhancement is exhibited in the flow through planar contractions even for very large contraction ratios of 80:1 (Evans and Walters, 1986). In place of vortex enhancement, however, a diverging flow regime – analogous to that originally documented by Cable and Boger (1978b) for shear-thinning fluids – was observed to develop in high $De$ flows of Boger fluids in planar contractions (Binding and Walters, 1988). This results in a pronounced increase in the pressure drop measured through the contraction, and limits the application of entry flows in inferring the extensional flow properties of viscoelastic fluids (Binding, 1988; Binding and Walters, 1988).

This dichotomy between planar and axisymmetric entry flows of Boger fluids remains unexplained. Experiments with polymer melts show that fluids which exhibit high extensional viscosities or unbounded stress growth give rise to substantial vortex growth in both planar, slit dies (White & Baird, 1986) and in tubular, capillary dies (White & Kondo,
Figure 2.7  Sequence of flow transitions observed in the viscoelastic flow of a PAC/CS Boger fluid through a 7.675:1 axisymmetric contraction. (a) $De = 0.14$, $Re = 1.36 \times 10^{-4}$; (b) $De = 0.72$, $Re = 7.8 \times 10^{-4}$; (c) $De = 1.67$, $Re = 1.9 \times 10^{-3}$; (d) $De = 2.08$, $Re = 3.5 \times 10^{-3}$; (e) – (h) Sequence of photographs at $De = 2.25$, $Re = 2.9 \times 10^{-2}$. (Reproduced from Nguyêñ and Boger, 1979).
The picture is similar for semidilute, shear-thinning polymer solutions; the measurements of Jones et al. (1987) show tension-thickening in both the planar extensional viscosity $\eta_p$ and the uniaxial extensional viscosity $\eta_E$, whereas the experiments of Walters & Webster (1982) show that vortex growth also occurs in both planar and axisymmetric geometries. However, despite evidence that Boger fluids exhibit significant strain-rate thickening in both $\eta_p$ and $\eta_E$ (Williams & Williams 1985; Jackson et al. 1987), the flow visualization results of Walters and coworkers have clearly shown that Boger fluids do not show any vortex growth in planar contractions. Detailed LDV and birefringence measurements for the flow of Boger fluids through planar and axisymmetric contractions are required to explain this inconsistency. Little is known about the presence of flow instabilities in the planar geometry; however close inspection of the streak photographs presented by Binding and Walters reveal the development of an asymmetric flow and the formation of unstable vortex at high $De$. This asymmetric flow may arise from the formation of a three-dimensional structure similar to that observed by Chiba et al. (1990) with a shear-thinning polymer solution. Unfortunately Binding and Walters provide no information on the spatial structure of the flow along the neutral direction of the test cell.

The first quantitative measurements of the spatial and temporal structure in the nonlinear flow transitions which occur in a viscoelastic entry flow were presented by Muller (Muller, 1986; Lawler et al. 1986). An automated two-color laser Doppler velocimetry system was used to study the flow of a highly elastic Boger fluid through a 4:1 abrupt axisymmetric contraction. The Boger fluid consisted of a solution of 0.17 wt% polyisobutylene (PIB) in polybutene and kerosene, with a zero-shear-rate relaxation time of 0.047 seconds. The most important result of this work was to show that at a relatively low critical Deborah number, $De_{osc}^{(osc)} = 0.80$, the flow near the contraction lip became three-dimensional, and all three velocity components exhibited time-dependent oscillations. The oscillation frequency was determined from the Fourier spectrum to be approximately 0.2 Hz and was found to increase with Deborah number as shown in Figure 2.8. Hysteresis in the oscillation frequency can also be observed and the measurements indicate another transition between the original time-periodic state and one with approximately twice the period. This behavior is indicative of a subcritical, period-doubling bifurcation. These velocity oscillations were confined to a small region near the contraction lip and were a precursor to the formation of a steady, two-dimensional, elastic vortex near the lip at a second critical Deborah number, $De_{lip} = 1.2$. Subsequent growth of this vortex could not be observed because of constraints to the maximum attainable Deborah number that were imposed by the fluid rheology and construction of the experimental system. One of the goals of this thesis is to employ a similar LDV system to resolve both the spatial and
Figure 2.8  Bifurcation diagram showing the onset of time-periodic oscillations near the lip of a 4:1 contraction at a critical Deborah number $De^{(osc)} = 0.8$ for the flow of PIB/PB/K Boger fluid. The frequency of the oscillations increases with $De$, and the flow eventually reverts back to steady flow at $De^{(lip)} = 1.2$. Hysteresis in the flow is observed in the measurements for decreasing $De$. (Reproduced from Lawler et al. 1986).
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Reproduced from Botter, 1987)

Figure 2.9 Qualitative sketches showing the evolution in flow structure in asymmetric contractions as a function of increasing $D_e$ and contraction ratio for (a) PI/PI and (b) PAC/CS. In each sequence the flow from the top row of sketches and highest in the bottom row. (and $D_e$) is lowest in the top row of sketches and lowest in the bottom row.
The role of time-dependent dynamics near the contraction lip on the formation and growth of the elastic vortex can not be inferred from flow visualization experiments such as those discussed above. In order to investigate these effects, Raiford (1988) extended the earlier LDV work of Muller (1986) to explore the stability of a PIB/PB Boger fluid flowing through axisymmetric contractions with $\beta = 2$, 4, and 8. By modifying the viscometric properties of the fluid Raiford was able to extend the time-dependent measurements of Muller to higher Deborah numbers that corresponded to those attained in the vortex growth and unstable flow regimes by Binnington et al. (1986). Typical LDV measurements of the time-dependent axial velocity on the centerline of the 4:1 contraction as a function of $De$ are shown in Figure 2.10. At a low Deborah number of $De = 0.4$, the flow along the centerline was found to be steady in time and an FFT of the velocity trace revealed no dominant oscillation frequencies. A transition to time-dependent flow near the lip was documented by Raiford at $De^{(osc)} = 1.5$ and the LDV measurements in Figure 2.10 clearly demonstrate that the velocity field remains time-dependent for all $De \geq De^{(osc)}$. As the time-dependent elastic vortex increases in size, the fluctuations in the velocity field increase in severity and ultimately lead to the very large amplitude nonlinear oscillations shown in Figure 2.10. Similar measurements were performed by Raiford for other contraction ratios of $\beta = 2$ and $\beta = 8$; however, little correlation between the results could be made, since the time-dependent behavior of the velocities and the shape of the flow streamlines varied dramatically in each contraction ratio.

The aim of the contraction flow experiments performed in this thesis is to couple highly accurate LDV measurements such as those of Muller (1986) and Raiford (1988) with video flow visualization techniques to associate unambiguously the role of time-dependent dynamics on the evolution in vortex structure observed in axisymmetric contraction flows. The experiments described in Chapter 6 consider contraction ratios of $\beta = 2$, 3, 4, 5, 6 and 8, in order to explore carefully the large variation in dynamics associated with small changes in flow geometry. By systematically varying the contraction ratio it is possible to demonstrate the competing effects of total extensional strain and shear rate in the upstream tube on the flow transitions near the lip entrance. The importance of the severe flow conditions near the re-entrant corner and the effect of the corner shape on the flow dynamics is also investigated by substituting a smooth radius of curvature for the sharp corner at the entrance to the small downstream tube.
Figure 2.10 Measurements of the centerline axial velocity near the contraction plane for the flow of a PIB/PB/C14 Boger fluid in a 4:1 axisymmetric contraction. As $De$ is increased the measurements show the onset of time-dependent oscillations which rapidly increase in amplitude. (Reproduced from Raiford, 1988).
2.3 Viscoelastic Flow Past A Constrained Circular Cylinder

The final flow considered in this thesis consists of viscoelastic flow past a smooth circular cylinder mounted centrally in a long planar channel, as shown in Figure 2.11. The cylinder diameter is $2a$ and the channel width is $2b$. The ratio of the cylinder radius to the channel half-width is defined as the cylinder-channel ratio, $\beta = a/b$. Far upstream and downstream of the cylinder the flow is fully developed plane Poiseuille flow. Along the surface of the cylinder the no-slip boundary condition on the velocity components gives $u_r = 0$, $u_\theta = 0$; and by applying the continuity equation it may be shown that around the cylinder perimeter both the derivatives $\partial u_r/\partial r$ and $\partial u_\theta/\partial \theta$ are also zero. The symmetric points $S_1$, $S_2$ at the front and rear of the cylinder surface are thus stagnation points and polymer molecules will have large residence times in the vicinity of the cylinder, resulting in the development of large molecular extensions and significant elastic stresses. This stress boundary layer will result in modification of the flow field around the cylinder and, as discussed in Chapter 1, this can lead to the formation of weld lines in the wake and considerable degradation in the ultimate material properties of plastics extruded past submerged bodies.

Viscoelastic flow in this geometry has not been afforded the intense attention that has been focused in the past on viscoelastic entry flows. Yet, in many ways, this geometry is ideally suited as a test problem for viscoelastic fluid mechanics for the following reasons: The complex fluid motion around the cylinder is a 'mixed' flow and contains regions where shearing effects are dominant (in the narrow gap between the cylinder and the walls) and other regions where significant extensional effects will be developed (near the upstream and downstream stagnation points). The relative contributions of these effects can be varied by considering different values of the cylinder-channel ratio $\beta$. These characteristics are analogous to those discussed in Section 2.2 for entry flows. However, in contrast to the contraction flow geometry, computational representations of this flow domain do not introduce any geometric singularities, and hence the flow problem is more amenable to numerical simulations. For these reasons the analogous axisymmetric problem of viscoelastic flow around a sphere constrained in a cylindrical tube has recently been adopted as a benchmark problem for numerical computations (Hassager, 1988; Crochet, 1988).

To date, no comprehensive review of previous investigations of viscoelastic flows in these geometries has been published, and therefore the following section has been extended beyond those works directly related to Boger fluids (of which there have been
Figure 2.11  Viscoelastic flow past a circular cylinder constrained in a planar slit. The cylinder of radius $a$ is mounted centrally in the channel which has a width $2b$. A Cartesian coordinate system is defined with its origin at the center of the cylinder.
very few) to encompass a more general survey of viscoelastic flow around cylinders and spheres. Relevant analytic and experimental results for the kinematics and flow stability observed in Newtonian fluids are summarized first in Section 2.3.1 as a background for the more detailed discussions in Sections 2.3.2 and 2.3.3 of the modifications to the kinematics observed in the two closely related cases of non-Newtonian flow past a cylinder and non-Newtonian flow around a sphere.

2.3.1 Newtonian Flow Past Cylinders and Spheres

Solutions to the classical problem of unbounded creeping flow of a Newtonian fluid past a submerged obstacle have been presented by many authors for a variety of body shapes, and are covered in detail in standard texts (see e.g. Batchelor 1967; Happel and Brenner 1973). Here we briefly summarize the results obtained for the two simplest cases of (i) a sphere settling in a viscous fluid and, (ii) creeping flow past an infinitely long cylinder. The coordinate surfaces of these geometries facilitate analytical solutions and ready comparison with direct experimental measurements of settling velocities and drag coefficients.

(i) Creeping Flow Past a Falling Sphere

The problem of a solid sphere settling in an unbounded quiescent fluid of constant Newtonian viscosity $\mu$, is depicted in Figure 2.12(a). The Stokes' equations of motion for creeping flow ($Re = 0$) are solved in spherical coordinates, subject to the boundary conditions of a uniform velocity $U$ on the surface of the sphere and zero velocity in the fluid far away from the sphere. Viscous drag at the no-slip boundary between the sphere and the fluid results in a net drag force $F$ on the sphere. Under steady-state conditions this force is exactly balanced by the gravitational force acting on the sphere such that

$$F = \frac{4}{3} \pi a^3 (\rho_s - \rho_l) g$$  \hspace{1cm} (2.7)

where $\rho_s$ and $\rho_l$ are, respectively, the densities of the sphere and surrounding fluid, and $g$ is the gravitational constant.

Experimental measurements of the drag force $F$ are thus easily realizable by dropping spheres of various densities and radii in the fluid and measuring the steady-state
Figure 2.12  (a) Schematic diagram for a sphere of radius $a$ and density $\rho_s$ settling in a viscous fluid of Newtonian viscosity $\mu$ and density $\rho_f$.

(b) Comparison of measured values of the drag coefficient for a sphere and two theoretical estimates; Stokes law (see eq. (2.9)) and the improved approximation given by equation (2.10). (Reproduced from Batchelor, 1967).
settling velocity. To enable comparison of experimental results on various sphere sizes, it is useful to define a dimensionless drag coefficient \( C_D \) as

\[
C_D = \frac{F}{\frac{1}{2} \rho U^2 \pi a^2}
\]  

(2.8)

Analytical solution of the Stokes' equations results in a total drag force \( F_{Sl} = 6 \pi \mu U \) and substitution in eq. (2.8) results in the following expression for the drag coefficient of Stokes' flow around a sphere:

\[
C_D^0 = \frac{6 \pi \mu U}{\frac{1}{2} \rho U^2 \pi a^2} = \frac{24}{Re}
\]  

(2.9)

where \( Re \) is the Reynolds number and is based on the sphere diameter, \( Re = 2 \rho a U / \mu \).

A more accurate solution to the Navier-Stokes equations at low, but finite, Reynolds number may be obtained by using a singular perturbation technique to match an inner solution near the sphere with a separate outer solution (Batchelor, 1967) and results in a drag coefficient of

\[
C_D = \frac{24}{Re} \left[ 1 + \frac{3}{16} Re + O(Re \ln Re) + \cdots \right]
\]  

(2.10)

The results of comparison between experiments and these two solutions are shown in Figure 2.12(b). Experimental measurements fall between eq. (2.9) and the more accurate eq. (2.10) with good agreement obtained up to \( Re \sim 0.5 \).

(ii) Creeping Flow Past a Fixed Cylinder

The basic geometry for two-dimensional flow past a fixed solid cylinder in an infinite free stream of fluid is shown in Figure 2.13(a). Solution of the inertialess Stokes' equation of motion in a cylindrical polar coordinate system, with origin \( O \) at the symmetry axis of the cylinder, results in a stream function of the form

\[
\psi = U \sin \theta \left( \frac{L}{r} + Mr - \frac{N}{2} r \ln r \right)
\]  

(2.11)

and a drag force per unit length of

\[
F / l = 2 \pi \mu UN
\]  

(2.12)
Figure 2.13  (a) Unbounded flow of a Newtonian fluid with viscosity $\mu$ past a fixed circular cylinder of radius $a$.

(b) Streamlines for Newtonian creeping flow past a cylinder in a channel of width $b = 5a$. (Reproduced from Bairstow et al., 1923)
where \( L, M, N \) are constants to be determined by satisfying the boundary conditions shown in Figure 2.13(a). Although this solution can satisfy the two no-slip boundary conditions on the cylinder surface it results in a divergent velocity as \( r \to \infty \) for any choice of the constant \( N \neq 0 \) and thus fails to satisfy the free stream boundary condition. This inconsistency is known as Stokes' Paradox and by applying dimensional arguments it may be shown to exist for any \textit{unbounded} two-dimensional flow (Happel and Brenner, p47-48).

The situation can be rectified by employing Oseen's improvement to the creeping flow equations of motion, which incorporates an approximation of the importance of inertial effects at large distances from the body. An approximate, but self-consistent, solution for the whole flow field is then obtained in terms of Bessel functions (Lamb, 1911; Bairstow et al., 1923). The drag coefficient for this solution is

\[
C_D^0 = \frac{8\pi}{Re \left[ \frac{1}{2} - \gamma + \ln 8 - \ln Re \right]} \tag{2.13}
\]

where \( Re \) is the dimensionless Reynolds number based on the cylinder diameter, and \( \gamma \) is Euler's constant (\( \gamma \approx 0.577 \)). Expanding the Bessel functions and keeping only first order terms results in the following approximate stream function which is accurate near the cylinder for distances up to \( (r/a) = 1/Re \):

\[
\psi = \frac{Usin\theta}{\frac{1}{2} - \gamma + \ln 8 - \ln Re} \left[ \frac{r}{2} - r\ln\left(\frac{r}{a}\right) - \frac{a^2}{2r} \right] \tag{2.14}
\]

An alternative solution to Stokes' Paradox is to bound the free stream flow by introducing walls at some distance \( b \) from the cylinder origin as shown previously in Figure 2.11. Solutions to the Stokes flow equations may now be sought in terms of the dimensionless geometric ratio of the cylinder radius to channel half-width; defined as \( \beta = (a/b) \). These solutions are difficult to obtain and require use of the method of images (Bairstow et al., 1922, 1923) or series expansions in spherical harmonics (Harrison 1924). The stream function obtained by Bairstow et al. for a cylinder/channel ratio of \( \beta = 0.2 \) is shown in Figure 2.13(b). Near the cylinder surface the streamlines are only slightly perturbed from the Oseen solution, eq. (2.14), however the presence of the walls modifies the more distant streamlines and results in an increase in the drag force on the cylinder. In a detailed series of calculations, Faxén (1946) has obtained series expansions at \( Re = 0 \) for the drag force and pressure drop caused by the cylinder up to terms of order
Figure 2.14 Curves for calculation of the increase in drag coefficient and increased pressure drop caused by the presence of walls for Newtonian creeping flow past a cylinder (from Faxén, 1946). The cylinder-channel ratio is $\beta = (a/b)$. The total drag force is given by $F = \mu U r_2$ and the total pressure drop is $P = \mu U p_2$. The remaining curves $r_1$ and $p_1$ give the drag coefficient and pressure drop for the alternative boundary conditions of a cylinder translating along a channel filled with a viscous fluid at rest.
Taking only the first few terms of this expansion the drag coefficient may be expressed as

\[ C_D(\beta) = \frac{8\pi}{[-0.9157 - (1+0.5\beta^2)\ln\beta + 1.2665\beta^2]} \]  \hspace{1cm} (2.15)

The full expressions are shown in Figure 2.14 for two sets of boundary conditions; the subscript '1' refers to the case of a cylinder translating with velocity \( U \) through a channel filled with fluid at rest, while the '2' indicates the alternative case of a fixed cylinder and plane Poiseuille flow in the channel. As the ratio \( \beta \) increases, the presence of the walls results in a significant increase in the drag on the cylinder. Although the accuracy of the expansion approach is reduced as \( \beta \) increases, for \( \beta = 0.5 \) the drag force on the cylinder is found to increase by a factor of approximately 15 from the Lamb solution given by eq. (2.13).

Since we will be dealing with highly viscous fluids we are primarily concerned with results obtained in creeping flows (\( Re \ll 1 \)); however, extensive experiments and simulations reveal a number of transitions and changes in the flow patterns around a cylinder as the Reynolds number is increased: In creeping flow the drag coefficient decreases with \( Re \), while the distribution of streamlines around the cylinder is perfectly symmetric as shown in Figure 2.15. The top half of the figure shows the streamlines determined experimentally (by streak photography) for the flow of water past a cylinder at \( Re = 0.16 \) (S. Taneda in Van Dyke 1982); the lower half of Figure 2.15 shows the streamlines calculated at \( Re = 0.16 \) by using equation (2.14). As inertial effects begin to become important at \( Re \sim 1 \) the drag coefficient continues to decrease, however the streamlines around the cylinder become asymmetric and are shifted downstream as shown in Figure 2.16(a). At \( Re \sim 9 \) a pair of recirculating vortices with closed streamlines appear in the region immediately aft of the cylinder as shown in Figure 2.16(b). These 'standing eddies' increase in size as the Reynolds number increases and extend downstream as shown in Figure 2.17(a). At \( Re \sim 40 \) the laminar flow in the wake becomes unstable to small perturbations resulting in oscillations in the downstream velocity (see Figure 2.17(b)). At \( Re \sim 60 \) these oscillations disrupt the standing eddies behind the cylinder and the two vortices are shed alternately at each half cycle of oscillation to form a von Kármán 'vortex street' as shown in Figure 2.17(d). The size of these vortices increases with \( Re \) and eventually at \( Re \sim O(1000) \) the flow in the boundary layer and wake becomes turbulent. A great deal of interest currently exists in understanding the local and global temporal characteristics of these wake instabilities and their suppression by 'suction',
Figure 2.15 Creeping flow past a cylinder at $Re = 0.16$. Upper half of the figure shows experimental streamlines in water (S. Taneda in Van Dyke, *An Album of Fluid Motion*, 1982). Lower half shows streamlines determined numerically using Lamb's solution (equation 2.14). *N.B.* In order to show the slow flow near the cylinder the streamlines are not equally spaced.
Figure 2.16  (a) Laminar flow with downstream shifting of the streamlines, $Re = 1.54$
(b) Laminar flow with 'standing eddies', $Re = 26$

*(Reproduced from S. Taneda in Van Dyke, An Album of Fluid Motion, 1982)*
Figure 2.17 Onset of flow instability and the development of a von Kármán 'vortex street' in the laminar wake behind a cylinder as the Reynolds number \( R \) is increased. (Reproduced from Batchelor, 1967)
‘bleed’ and pinning rods; see the recent reviews by Oertel (1990), and Huerre and Monkewitz (1990).

In other laboratory flow systems, such as Taylor-Couette flow, the presence of extremely small amounts of high molecular weight polymers has been found to stabilize the flow and inhibit the onset of inertial flow instabilities (Giesekus, 1972; Beavers and Joseph, 1974; Larson, 1989). This flow stabilization phenomenon is employed commercially in ‘drag reducing’ additives that reduce the pressure drop in turbulent pipe flows (Bird et al., 1987a). A similar flow stabilization is observed in the flow of very dilute polymer solutions past a circular cylinder; the presence of polymer additives is found to delay the onset of vortex formation and to dramatically reduce the frequency of vortex shedding at moderate Reynolds numbers (Usui et al. 1980; Kim and Telionis, 1989). Inertial effects generally remain very small in semi-dilute viscoelastic fluids such as those described in Chapter 4, and we should not expect to see exactly the same sequence of transitions as described above. However, the development of high elastic stresses in the boundary layer near the cylinder will lead to a modification of the local flow field and may lead to the onset of an independent series of elastically-driven instabilities. The importance of these elastic effects relative to viscous effects is characterized using the Deborah number defined earlier in equation (1.1).

2.3.2 Viscoelastic Flow Around a Cylinder

Early interest in viscoelastic flows around cylindrical and spherical objects arose from their central role in standard flow measurement devices such as hot wire anemometers and falling ball viscometers. Elasticity was found to perturb the flow field around the body significantly, and this resulted in alterations in the correlations of heat transfer and drag coefficients used to analyze the experimental measurements (see for example Leslie and Tanner, 1961; Tanner, 1964; Smith et al. 1967). Subsequent research has concentrated on the modifications that viscoelasticity causes to the characteristics of the flow, especially the drag coefficient and the velocity profiles near the cylinder surface. Previous investigations of laminar viscoelastic flow around a cylinder are listed in Table 2.2. Considerable disagreement has been reported in the literature regarding the effects of elasticity on the positions of the flow streamlines and the drag exerted on the cylinder.

In the first detailed study, James & Acosta measured heat transfer coefficients and drag coefficients of extremely small cylinders in dilute polymer solutions over a range of Reynolds number \( Re = 1 - 100 \). The cylinders used were standard hot wire anemometers
<table>
<thead>
<tr>
<th>Authora</th>
<th>Year</th>
<th>Typeb</th>
<th>Ratio $\beta = a/b$</th>
<th>Streamline Shift</th>
<th>Drag Change</th>
<th>$De, Re$ Rangec</th>
<th>Fluid Typed</th>
</tr>
</thead>
<tbody>
<tr>
<td>James &amp; Acosta</td>
<td>1970</td>
<td>E</td>
<td>unbounded</td>
<td>—</td>
<td>increased</td>
<td>$De &lt; 1$</td>
<td>(Re &gt; 1)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>decreased</td>
<td>—</td>
<td>Re &gt; 1</td>
</tr>
<tr>
<td>Ulmann &amp; Denn†</td>
<td>1971</td>
<td>T</td>
<td>0.068</td>
<td>upstream</td>
<td>decreased</td>
<td>$De &lt; 1$</td>
<td>Re &lt;&lt; 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>E</td>
<td></td>
<td>upstream</td>
<td>—</td>
<td>$De = .0032$</td>
<td>Re = $2 \times 10^{-4}$</td>
</tr>
<tr>
<td>Broadbent &amp; Mena†</td>
<td>1974</td>
<td>E</td>
<td>0.334</td>
<td>no change</td>
<td>decreased</td>
<td>$De &lt; 1$</td>
<td>Re &lt; 1</td>
</tr>
<tr>
<td>Mena &amp; Caswell†</td>
<td>1974</td>
<td>T</td>
<td>unbounded</td>
<td>downstream</td>
<td>decreased</td>
<td>$De &lt;&lt; 1$</td>
<td>Re &lt;&lt; 1</td>
</tr>
<tr>
<td>Mizushina &amp; Usui</td>
<td>1975</td>
<td>N</td>
<td>unbounded</td>
<td>upstream</td>
<td>decreased</td>
<td>$De &lt; 1$</td>
<td>Re &gt; 1</td>
</tr>
<tr>
<td>Pilate &amp; Crochet</td>
<td>1977</td>
<td>N</td>
<td>unbounded</td>
<td>—</td>
<td>decreased</td>
<td>—</td>
<td>Re &lt; 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>downstream</td>
<td>increased</td>
<td>—</td>
<td>Re &gt; 1</td>
</tr>
<tr>
<td>Townsend</td>
<td>1980</td>
<td>N</td>
<td>unbounded</td>
<td>downstream</td>
<td>decreased</td>
<td>$De &lt; 1$</td>
<td>Re &lt; 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>downstream</td>
<td>increased</td>
<td>$De &lt; 1$</td>
<td>Re &gt; 1</td>
</tr>
<tr>
<td>Koniuta et al.</td>
<td>1980</td>
<td>E</td>
<td>0.02</td>
<td>upstream</td>
<td>—</td>
<td>—</td>
<td>Re &gt; 1</td>
</tr>
</tbody>
</table>

Table 2.2  Previous Investigations of Viscoelastic Flow Around a Circular Cylinder [continued overleaf]
<table>
<thead>
<tr>
<th>Authors</th>
<th>Year</th>
<th>Type</th>
<th>De, Re</th>
<th>Flow</th>
<th>Boundary Condition</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Christiansen</td>
<td>1980</td>
<td>E</td>
<td>0.094</td>
<td>—</td>
<td>—</td>
<td>De &lt; 1, Re &lt; 1, 1.0% HEC/H₂O</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(see also 1985)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.5% PAC/H₂O</td>
</tr>
<tr>
<td>Manero &amp; Mena</td>
<td>1981</td>
<td>E</td>
<td>0.195</td>
<td>decreased</td>
<td>De &lt; 1, (Re &lt;&lt; 1)</td>
<td>0.5% PAC/H₂O</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>increased</td>
<td>De &gt; 1, (Re &lt;&lt; 1)</td>
<td></td>
</tr>
<tr>
<td>Townsend</td>
<td>1984</td>
<td>N</td>
<td>unbounded</td>
<td>increased</td>
<td>De = 1 – 5, Re &gt; 1</td>
<td>Oldroyd-B</td>
</tr>
<tr>
<td>Chilcott &amp; Rallison†</td>
<td>1988</td>
<td>N</td>
<td>unbounded</td>
<td>increased</td>
<td>De &gt; 1, Re &lt;&lt; 1</td>
<td>FENE dumbell</td>
</tr>
<tr>
<td>Mochimaru</td>
<td>1988</td>
<td>N</td>
<td>unbounded</td>
<td>decreased</td>
<td>(De &lt; 1), Re &gt; 1</td>
<td>Second Order Fluid</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>increased</td>
<td>De &gt; 1, (Re &gt; 1)</td>
<td></td>
</tr>
<tr>
<td>Dhahir &amp; Walters</td>
<td>1989</td>
<td>E</td>
<td>0.60</td>
<td>—</td>
<td>decreased</td>
<td>De &lt; 0.2, Re &lt;&lt; 1, 2.0% PAC/H₂O</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.1% PAC/CS, UCM</td>
</tr>
<tr>
<td>Hu &amp; Joseph</td>
<td>1990</td>
<td>N</td>
<td>0.02</td>
<td>downstream</td>
<td>decreased</td>
<td>De ≤ 1, Re &lt; 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>upstream</td>
<td>increased</td>
<td>De &gt; 1, Re &gt; 1</td>
</tr>
</tbody>
</table>

**Table 2.2** (cont.) Previous Investigations of Viscoelastic Flow Around a Circular Cylinder.

**Notes**

(a) † indicates that viscoelastic flow around a sphere was also considered.

(b) The previous investigations are classified as: E = Experimental; T = Theoretical; N = Numerical.

(c) Previous authors have investigated the effects on the flow field of varying either the Deborah number (De) or Reynolds number (Re).

Quantities in parentheses indicate the parameter is held constant whilst the other parameter is varied.

(d) Abbreviations for the viscoelastic fluid formulations employed in experimental studies are: PAC = polyacrylamide; CMC = carboxymethylcellulose; HEC = hydroxyethylcellulose; PEO = polyethylene oxide; CS = cornsyrup.
of diameters $a \leq 0.006$ in. and the viscoelastic fluids were very dilute solutions of polyethylene oxide (PEO) in water. No rheological characterization of the fluids was performed although the intrinsic viscosity $[\eta]$ was measured and correlated with the molecular weight using a Mark-Houwink expression. The maximum concentration of PEO used was 327 ppm (0.03 wt%) and for such dilute polymer systems the viscosity will increase linearly with polymer concentration, while the relaxation times will be much less than 1 second. At a fixed value of $Re = 100$, increasing the fluid elasticity (by increasing the amount of dissolved PEO) was found to reduce the heat transfer by up to 70% and to increase the drag coefficient by a factor of three from the values obtained for Newtonian fluids. For each PEO solution the drag coefficient was also found to decrease as $Re$ increased; however the decrease in $C_D$ was less for higher concentrations of PEO and appeared to reach a plateau of constant value at high $Re$.

Ultmann and Denn performed the first detailed theoretical analysis of viscoelastic flow past a cylinder using the Upper Convected Maxwell (UCM) model. In order to simplify the equation set, the Oseen approximation was used to linearize the momentum equation and by limiting the flow to small deformations the constitutive equation was reduced to the Maxwell model of linear viscoelasticity (see e.g. Bird et al., 1987a). This latter simplification is a much more severe limitation than the Oseen approximation, since the stagnation points on the cylinder surface can lead to large polymer deformations. The approach has been roundly criticized by Mena and Caswell (1974) and by Zana et al. (1975), since the approximation is not uniformly valid throughout the domain for any nonzero value of $De$ and it results in an overspecified set of boundary conditions. However, the contribution of Ultmann and Denn is significant since it indicated for the first time the possibility of a change of type in the viscoelastic governing equations. The mathematical type of the equations is found to depend on the product of the Reynolds number and Deborah number which can be expressed for a Maxwell model as

$$ReDe = \frac{U^2}{[\eta_0/\rho\lambda_0]} \quad (2.16)$$

The denominator in this expression is the square of the shear wave speed though a linear viscoelastic medium. For $ReDe < 1$ the equation set is elliptic and the velocity solutions are smooth everywhere, however for $ReDe \geq 1$, i.e. for velocities greater than the wave speed, the equation set becomes hyperbolic and discontinuities, or shocks, can propagate along the fluid streamlines. An understanding of the equation type is now known.
Figure 2.18  Viscoelastic flow past a cylinder showing a pronounced *upstream* shift of the streamlines.  
(a) Calculations of the streamlines for $Re = 0.1$: Upper half, Newtonian streamlines; lower half $N_{We} (\equiv De) = 0.5$.  
(b) Experimental streamlines past a cylinder of diameter $2a = 3/8"$: Upper half, corn syrup/water (Newtonian); lower streamlines, 1.7 wt% CMC in water.  
(Reproduced from Ultmann and Denn, 1971).
to be essential to numerical solution of viscoelastic flow problems (see for example Joseph 1985; King et al. 1988; Northey et al. 1990). Although Ultmann and Denn's expression is limited to small values of ReDe, their final expression for the drag coefficient was found to be

\[ C_D = \frac{8\pi[1 - ReDe]}{Re[\frac{1}{2} - \gamma - \ln(Re/8) + 0.5ReDe]} \]  

(2.17)

which reduces to the Stokes-Lamb solution (equation 2.13) when \( De \equiv \lambda_0 U/a = 0 \). The streamlines obtained using this analytic solution, even at low \( De \), indicated a large upstream displacement when compared to the Newtonian streamlines, as shown in Figure 2.18(a). Flow visualization experiments using dye-streaks to follow the streamlines around the cylinder were also presented for flow of a Newtonian fluid and a viscoelastic solution of 1.75 wt\% carboxymethylcellulose (CMC) in water. At a Deborah number of \( De = 3.2 \times 10^{-3} \) the streamlines in the viscoelastic fluid were found to be significantly shifted upstream as shown in Figure 2.18(b). However only one experimental observation was presented and no conclusions can be drawn about the effects of varying the Deborah number on the flow patterns.

Mena and co-workers have carried out several investigations on viscoelastic flow past cylinders and spheres. Mena and Caswell (1974) provided a rigorous matched asymptotic expansion using the Oldroyd-B constitutive equation that is valid for \( Re \ll 1 \) and \( De \ll 1 \). The contribution of elasticity was found to shift the streamlines downstream by a distance of \( O(De) \) as shown in Figure 2.19(a), and to reduce the drag coefficient quadratically from the Stokes-flow solution for flow past a cylinder, \( i.e. \)

\[ C_D(De) = C_D^0 \left[ 1 - O(\lambda U/a)^2 + \cdots \right] \]  

(2.18)

where \( C_D^0 \) is the value obtained by Lamb and given in eq. (2.13). Similar \( O(De^2) \) reductions have also been predicted theoretically for viscoelastic flow around a sphere and are discussed in Section 2.3.3 (Leslie and Tanner 1961; Caswell and Schwarz 1962).

These calculations agreed with the drag force measurements performed by Broadbent and Mena for flow around cylinders and spheres. Their measurements of \( C_D \) in a Newtonian fluid (glycerine) and a shear-thinning aqueous solution of 2.0 wt\% PAC are shown in Figure 2.19(b) together with a comparison of the theoretical approximation for \( C_D^0 \) given by equation (2.13). The departure of the drag coefficient from the Newtonian value was shown to be quadratic by reploting the data in the form \((C_D^0 - C_D)/C_D^0 \) against
Streamlines in flow past a circular cylinder. Newtonian; Elastic.

Figure 2.19  (a) Calculations for flow past a cylinder showing a slight downstream shift of the streamlines at $De = 0.2$ (Reproduced from Mena and Caswell, 1974). (b) Experimentally determined drag coefficient vs. Reynolds number $Re$ for (Θ) a Newtonian fluid, and for (○) an aqueous solution of 2 wt% PAC. (c) Quadratic departure of the viscoelastic drag coefficient from the Newtonian prediction obtained by Lamb. (Both reproduced from Broadbent and Mena, 1974).
(U/a)² as shown in Figure 2.19(c). For $Re < 0.1$ the data lie on a straight line. At higher $Re$ the departure is smaller as higher order terms become important. However, the flow visualization pictures of Broadbent and Mena showed no visually discernable streamline displacement upstream or downstream. In later experiments with cylinders, Manero and Mena (1981) used solutions of PAC in glycerine and water to span a wide range of $De$ at low $Re$. For $De < 1$ a small downstream shift of the streamlines was observed; however, at higher Deborah numbers, $De > 1$, this shift reversed and a large upstream displacement of the streamlines could be seen.

In his thesis, Christiansen (1980) employed two-component laser Doppler velocimetry to investigate in detail the velocity profiles near cylinders. Two experimental fluids were used; a semi-dilute aqueous solution of 1.0 wt% hydroxyethylcellulose (HEC) with moderate elasticity, and an aqueous solution of 0.5 wt% PAC with a long relaxation time which enabled high Deborah numbers to be attained. At low $De < 1$ a greater velocity was measured at a fixed point upstream of the cylinder than at the symmetric point in the downstream wake of the cylinder – indicating a downstream shift in the streamlines. However, at higher $De (∼ 5–10)$ the opposite velocity distribution was observed, suggesting an upstream shift in the streamlines.

Koniuta et al. (1980) also used LDV to measure the $v_z$ velocity component (i.e. the component in the flow direction) near thin wire cylinders in a very dilute ‘drag reducing’ polymer solution of 1000 ppm PEO in water. Flow visualization and LDV measurements downstream of the cylinder at a moderate Reynolds number ($Re ∼ 5$) showed that the wake observed with the polymer solution was broader and slower moving than the corresponding Newtonian flow; indicating a downstream shift of the streamlines. As the Reynolds number was increased the forward stagnation point gradually moved upstream, away from the cylinder surface, while the cylinder became surrounded by a wide boundary layer within which the fluid velocity was very low. This observation is consistent with the measurements of James and Acosta that showed a significant reduction at high $Re$ in the heat transfer from cylinders in dilute polymer solutions.

In the most recent experimental investigation, Dhahir and Walters (1989) measured the drag on a cylinder in a square duct with a high cylinder/channel ratio, $β = 0.6$. In addition to positioning the cylinder centrally in the channel, they also investigated the modification on the drag caused by an asymmetric placement of the cylinder. The effects of fluid rheology were explored by using aqueous solutions of 1.5 – 2.0% PAC, a Boger fluid of 0.1 wt% PAC in CS/H₂O, and a 3.0 wt% solution of Xanthan gum (a rigid rod polymer). For each of these fluids, fluid elasticity was found to result in a reduction of the drag force on the cylinder. Increasing the eccentricity of the cylinder placement was also
found to reduce the drag force while additionally introducing a lift force normal to the flow and directed towards the near wall. Rudimentary numerical simulations on a coarse finite element were also performed by Dhahir and Walters using both a generalized Newtonian fluid (GNF) model, and the UCM constitutive equation. These limited calculations showed the same qualitative dependence of the drag on \( De \) and eccentricity but the agreement was not quantitative, even for the purely Newtonian case. The simulations also failed to show any modifications in the shape of the streamlines as \( De \) was increased.

The first numerical simulations to show the effects of elasticity on the flow characteristics were presented by Pilate and Crochet (1977). Their calculations employed a Second Order Fluid (SOF) model in which elastic effects are introduced as small perturbations from Newtonian behavior, and the results obtained are thus only valid for \( De < 1 \). Pilate and Crochet found that for low \( Re \) and \( De \) the drag coefficient decreased from the Newtonian value, as observed in the experiments of Broadbent and Mena. However, for \( Re > 10 \) the drag on the cylinder increased beyond the Newtonian result in agreement with the results of James and Acosta. At high \( Re \) (but low \( De \)) the streamlines were also found to be shifted slightly downstream, relative to the Newtonian streamlines, and the length of the standing eddies behind the cylinder increased.

Townsend (1980) used a block-iteration finite difference method to calculate solutions for viscoelastic flow past a cylinder using an Oldroyd 4-constant fluid model (Bird et al., 1987a). At \( De = 0.5 \), elasticity resulted in a small shift downstream of the streamlines. The calculations covered a wide range of Reynolds number; however, difficulties associated with the numerical scheme limited the calculations to \( De < 1 \). In this range of parameter space, the calculations of drag coefficient agreed qualitatively with the previous results of Pilate and Crochet: for \( Re < 1 \) elasticity produced a small decrease in \( C_D \), while for higher speed flows (\( Re > 5 \)) a somewhat larger increase in drag was found. In later time-dependent calculations with an Oldroyd-B model, Townsend (1984) extended the range of solutions to \( De \sim 5 \). Once again a slight downstream shift in the streamlines was predicted together with an increase in the cylinder drag. In addition the time-dependent simulation showed the evolution and growth of a zone of stagnant fluid around the cylinder; as previously observed in the flow visualization pictures of James and Acosta and the LDV results of Koniuta et al. (1980).

Very recently quantitative calculations of these experimental phenomena have been made by Delvaux and Crochet (1990) and Hu and Joseph (1990). These numerical simulations considered high Reynolds number flows of the UCM model and showed that the anomalous correlations of transport properties such as drag coefficient and heat transfer rates correspond to a change of type in the governing equations, as first discussed by
Ullmann and Denn (1971). For flows with velocities greater than the shear-wave speed (equivalent to $ReDe \geq 1$) the calculations predicted a large increase in the drag coefficient and a corresponding decrease in the heat transfer coefficient as observed by James and Acosta (1970). Plots of the flow streamlines for $ReDe \geq 1$ also revealed the development of a large region of slowly moving fluid near the cylinder as documented by Koniuta et al. (1980).

Chilcott and Rallison performed time-dependent numerical calculations for viscoelastic creeping flow of a polymer solution past cylinders, spheres and cylindrical bubbles. The fluid was modeled by a FENE dumbbell constitutive equation, in which the macromolecules are considered to be a dilute suspension of noninteracting dumbbells with finite extensibility. The numerical procedure allowed for the development of asymmetric or time-dependent solutions; however, for the range of parameters covered, no instabilities were encountered and steady-state, symmetric solutions were obtained for all Deborah numbers up to $De = 16$. The macromolecular flow structure near the cylinder is shown in Figure 2.20 by plotting contours of constant polymer deformation. High polymeric stresses, corresponding to large molecular extensions are concentrated in three regions: near the forward stagnation point; in the high shear regions on either side of the cylinder, and in the long narrow wake downstream of the rear stagnation point. This wake region consists of highly extended dumbbells which are advected a large distance downstream before relaxing fully. The numerical results again showed a small decrease in $C_D$ for $De < 1$ followed by an increase above the Newtonian value that asymptotically approached a constant value at high $De$. The introduction of a constitutive equation which has a molecular interpretation in these calculations also showed that, in addition to the relaxation time of the fluid, knowledge of the polymer chain extensibility is important in understanding viscoelastic flow near the cylinder. By varying the fully extended length of the dumbbell, and thus the extensional rheology of the fluid, Chilcott and Rallison found they could alter the position of the asymptotic plateau in $C_D$ to a value either above, or below the Newtonian result. For highly extensible polymer molecules the possibility of a 'negative wake' or velocity overshoot (as discussed below for spheres) was also indicated.

The Boger fluid to be used in our experiments has been modeled with a similar FENE dumbbell constitutive equation (see Chapter 4) and the extensibility of the polymer chains is predicted to be extremely high. On the basis of the work by Chilcott and Rallison a negative wake is therefore expected for unbounded flow of a Boger fluid around a cylinder. The authors however were unable to provide a comparison with experimental results since "corresponding measurements of velocity distributions for Boger fluids are unfortunately not available" (Chilcott and Rallison 1988, p413). The LDV measurements

93
Figure 2.20 Contours of constant stress ($\text{tr}[\tau_p]$) for viscoelastic flow past an unbounded cylinder; (a) Overall plot showing extended downstream wake; (b) local structure near the cylinder. (Reproduced from Chilcott and Rallison, 1988)
presented in this thesis will allow detailed experimental investigation of the velocity field near the cylinder and in the downstream wake.

2.3.3 Viscoelastic Flow Around Spheres

A significant volume of literature has also been published on the closely related problem of viscoelastic flow past spherical objects. In addition to a fundamental analysis of the modification of the flow characteristics past a single sphere, this research is motivated by the need to develop an understanding of the behavior of viscoelastic suspensions and slurries that commonly occur in the polymer processing industry. A number of the works discussed above in Section 2.3.2 also considered flow around spheres and are identified with a ∗ in Table 2.2. In general, the qualitative observations for spheres were in agreement with the results discussed for flow around cylinders, and thus are not elaborated on further. In addition to these papers, there have been many other relevant contributions to the problem of viscoelastic flow around a sphere; these publications are summarized in Table 2.3, and have also recently been the subject of a brief review by Walters and Tanner (1991). Since we are primarily concerned with the two-dimensional problem of flow past a cylinder, these papers are not discussed in depth; however, some new phenomena not previously mentioned are highlighted.

Zana et al. (1975) pointed out the errors in the Oseen-like analysis of Uitmann and Denn (1971), and published photographs of viscoelastic flow around a sphere over a wide range of Deborah numbers, including the flow conditions specified by Uitmann and Denn. These flow visualization results showed a small upstream shift in the streamlines which was greatly reduced in extent compared to the results of Uitmann and Denn. Acharya et al. (1976a, b) gave a comprehensive review of early work on this problem, and provided extensive empirical correlations for the drag coefficient of spheres sedimenting in inelastic (‘pow r-law’) and viscoelastic fluids over a wide range of Re.

Sigli and Coutanceau (1977) were the first to investigate experimentally the effect of wall proximity on the drag by varying β; the ratio of sphere to tube diameter. In addition to an upstream shift in the streamlines they also documented a velocity overshoot or ‘negative wake’ downstream of the sphere. Figures 2.21(a) and (b) show photographs of spheres falling in a tube with diameter ratio β = 0.5, for both a Newtonian and a viscoelastic fluid. The fluid is initially at rest far upstream and downstream of the sphere and the camera is held motionless in the laboratory frame of reference. The particle tracks arise from the disturbance in the fluid caused by the sphere as it falls past the camera. For the Newtonian
<table>
<thead>
<tr>
<th>Author</th>
<th>Year</th>
<th>Type</th>
<th>Ratio $\beta = a/b$</th>
<th>Streamline Shift</th>
<th>Drag Change</th>
<th>$De, Re$ Range</th>
<th>Fluid Type</th>
</tr>
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<tr>
<td>Leslie &amp; Tanner</td>
<td>1961</td>
<td>T</td>
<td>unbounded</td>
<td>downstream</td>
<td>decreased</td>
<td>$De &lt;&lt; 1$</td>
<td>$Re &lt;&lt; 1$</td>
</tr>
<tr>
<td>Caswell &amp; Schwarz</td>
<td>1962</td>
<td>T</td>
<td>unbounded</td>
<td>downstream</td>
<td>decreased</td>
<td>$De &lt;&lt; 1$</td>
<td>$Re &lt;&lt; 1$</td>
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<tr>
<td>Zana et al.</td>
<td>1975</td>
<td>E</td>
<td>$0.023 - 0.17$</td>
<td>upstream</td>
<td>—</td>
<td>$De &lt; 20$</td>
<td>$Re &lt; 0.1$</td>
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<tr>
<td>Acharya et al.</td>
<td>1976a,b</td>
<td>E</td>
<td>$0.01 - 0.127$</td>
<td>—</td>
<td>decreased</td>
<td>—</td>
<td>$1 &lt; Re &lt; 100$</td>
</tr>
<tr>
<td>Sigli &amp; Coutanceau</td>
<td>1977</td>
<td>E</td>
<td>$0.25 - 0.75$</td>
<td>upstream</td>
<td>decreased</td>
<td>$De &lt; 0.1$</td>
<td>$Re &lt; 0.1$</td>
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<td>Adachi et al.</td>
<td>1977</td>
<td>N/E</td>
<td>unbounded</td>
<td>upstream</td>
<td>increased</td>
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<td>$0.1 &lt; Re &lt; 60$</td>
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<td>Chhabra et al.</td>
<td>1980</td>
<td>E</td>
<td>$0.01 - 0.07$</td>
<td>—</td>
<td>decreased</td>
<td>$De &lt; 2$</td>
<td>$Re &lt; 0.1$</td>
</tr>
<tr>
<td>Bisgaard</td>
<td>1983</td>
<td>E</td>
<td>$0.3 - 0.5$</td>
<td>downstream</td>
<td>decreased</td>
<td>$De &lt; 50$</td>
<td>$Re &lt; 1$</td>
</tr>
<tr>
<td>Hassager &amp; Bisgaard</td>
<td>1983</td>
<td>N</td>
<td>$0.01 - 0.5$</td>
<td>—</td>
<td>decreased</td>
<td>$De &lt; 1$</td>
<td>$Re &lt;&lt; 1$</td>
</tr>
<tr>
<td>Maalouf &amp; Sigli</td>
<td>1984</td>
<td>N/E</td>
<td>$0.1 - 0.5$</td>
<td>downstream</td>
<td>—</td>
<td>$De &lt; 0.5$</td>
<td>$Re &lt;&lt; 1$</td>
</tr>
</tbody>
</table>

Table 2.3  Previous Investigations of Viscoelastic Flow Around a Sphere  [continued overleaf...]
<table>
<thead>
<tr>
<th>Author</th>
<th>Year</th>
<th>Type</th>
<th>Concentration</th>
<th>Flow Direction</th>
<th>Flow Effect</th>
<th>Deborah Number</th>
<th>Reynolds Number</th>
<th>Fluid Formulations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sugeng &amp; Tanner</td>
<td>1956</td>
<td>N</td>
<td>0.5</td>
<td>Downstream</td>
<td>Decreased</td>
<td>De &lt; 1</td>
<td>Re &lt;&lt; 1</td>
<td>UCM / PTT</td>
</tr>
<tr>
<td>Mena <em>et al.</em></td>
<td>1987</td>
<td>E</td>
<td>0.05 – 0.5</td>
<td></td>
<td>Decreased</td>
<td>De &lt; 3</td>
<td>Re &lt; 1</td>
<td>0.8% PAC/H₂O 0.02% PAC/CS 1.5% CBP/EG</td>
</tr>
<tr>
<td>Lunsmann</td>
<td>1989</td>
<td>N</td>
<td>0.125 – 0.5</td>
<td>Downstream</td>
<td>Decreased</td>
<td>De &lt; 2</td>
<td>Re &lt;&lt; 1</td>
<td>UCM</td>
</tr>
<tr>
<td>Chmielewski <em>et al.</em></td>
<td>1990</td>
<td>E</td>
<td>0.02 – 0.37</td>
<td></td>
<td>Decreased</td>
<td>De &lt; 1</td>
<td>Re &lt;&lt; 1</td>
<td>PAC/CS/H₂O PIB/PB/K</td>
</tr>
<tr>
<td>Tirtaatmadja <em>et al.</em></td>
<td>1990</td>
<td>E</td>
<td>0.03 – 0.21</td>
<td></td>
<td>Increased</td>
<td>De ≤ 2</td>
<td>Re &lt;&lt; 1</td>
<td>PIB/PB/K</td>
</tr>
</tbody>
</table>

Table 2.3 (cont.) Previous Investigations of Viscoelastic Flow Around a Sphere

Notes:  
(a) The previous investigations are classified as: E = Experimental; T = Theoretical; N = Numerical
(b) Previous authors have investigated the effects of the flow field of varying either the Deborah number (De) or Reynolds number (Re).
(c) Abbreviations for the viscoelastic fluid formulations employed in experimental studies are: PAC = polyacrylamide; CMC = carboxymethylcellulose; HEC = hydroxyethylcellulose; PEO = polyethylene oxide; CBP = carbopol; G = glycerine; CS = cornsyrup; EG = ethylene glycol. The constitutive equations used in the numerical investigations are: UCM = upper convected Maxwell; PTT = Phan-Thien – Tanner; T.O.F = third order fluid.
fluid in \((a)\) the flow is symmetric about the equatorial plane of the sphere with a recirculation caused by displacement of fluid in front of the sphere out towards the wall. For the PEO polymer solution \((b)\) the flow patterns are significantly different; the disturbance caused by the sphere extends over greater distances, and the flow is no longer symmetric about the equatorial plane. Downstream on the centerline, near the sphere, the fluid moves in the same direction as the sphere, as in the Newtonian case \((a)\); however farther downstream of the sphere the fluid flows away from the sphere. This phenomenon has been described as a ‘negative wake’ by Hassager (1979). This reverse flow away from the sphere may explain the experimental observations of Riddle et al. (1977) that two spheres sedimenting along their centerline tended to aggregate for small initial separations but separated for initial separations greater than a critical distance. Velocity profiles measured by Sigli and Coutanceau on the centerline upstream and downstream of the sphere are shown in Figure 2.21(c). These velocity profiles are shown in a frame of reference moving with the sphere (at velocity \(-V_0\)) such that the velocity at the sphere surface is zero and the velocity of the fluid far up and downstream is \(V_0\). The negative wake results in a velocity overshoot, such that \(V/V_0 > 1\) at a point downstream of the sphere. The effect of increasing \(De\) can be seen from Figure 2.21(c) to increase the magnitude of this overshoot and move the position of the local maximum closer to the rear stagnation point of the sphere. Sigli and Coutanceau also demonstrated that inertia tended to act in reverse to elasticity; at higher \(Re\) the velocity overshoot decreased in size and was shifted downstream. Experiments in various tube sizes showed that the presence of wall effects at high \(\beta \geq 0.25\) further enhanced the velocity overshoot and moved it closer to sphere. Wall effects were thus found to be equivalent to an increase in the Deborah number of the flow. This enhancement of viscoelastic modifications to the flow caused by the presence of walls has also been documented by Cho et al. (1980) and Chhabra et al. (1981).

Recent work by Sigli and Maalouf (1984) has investigated the effects of body shape and fluid rheology on these velocity profiles. Flow visualization experiments in PEO solutions revealed that a similar ‘negative wake’ was observed with other axisymmetric bodies such as ellipsoids and ovoids. Once again, increasing \(De\) and the presence of wall effects at high \(\beta\) were found to increase the magnitude of the velocity overshoot. Most importantly, however, this study indicated that the ‘negative wake’ phenomena arises from a combination of shear-thinning and elasticity. The negative wake could be clearly identified in experiments with semidilute solutions of PEO in water (a shear-thinning viscoelastic solution); whereas measurements in an inelastic shear-thinning solution of
Figure 2.21 The 'negative wake' effect: Flow visualization of a sphere falling in a narrow tube ($\beta = 0.5$) shows the streamlines in a Newtonian fluid (a) at $Re = 0.2$ are symmetric about the equatorial plane of the sphere. In a viscoelastic fluid (b) at $De = 0.06$ the fluid on the centerline far downstream flows away from the sphere. Measurements of the fluid velocity relative to the sphere (c) show a velocity overshoot downstream (Reproduced from Sigli and Coutanceau, 1977).
carboxymethylcellulose and a constant viscosity PAA/CS Boger fluid revealed modifications in the downstream velocity profiles but no velocity overshoot.

The pronounced effects of wall proximity and viscoelasticity on the flow led to the adoption of the problem of a sphere falling axially in a tube with diameter ratio $\beta = 0.5$ as a benchmark for numerical simulations (Hassager, 1988). The wall correction factor $K$ is defined as the ratio of the drag force experienced by the sphere in a confined geometry, divided by the drag experienced by a sphere falling in an unbounded Newtonian fluid, given by equation (2.9). For a Newtonian fluid, with $\beta = 0.5$ and $De = 0$, this factor is found by using the Faxén formula (Faxén, 1946) to be $K_N = 5.977$; i.e. the drag on the sphere in a narrow tube is increased almost six-fold. Finite element simulations and experimental measurements by Hassager and Bisgaard (1983) showed that the wall correction factor $K$ was a rapidly decreasing function of both the ratio $\beta$ and the Deborah number. Wall effects were also investigated numerically by Sugeng and Tanner (1986) using a boundary element method with a modified Phan-Thien – Tanner (PTT) constitutive equation. The flow streamlines were again found to be shifted downstream and, for $\beta = 0.5$, the presence of the wall was found to reduce the drag from the corresponding Newtonian value. This reduction was found to be 25% for a constant viscosity UCM model and could be enhanced still farther (to 40%) by introducing shear-thinning into the model. In the most recent calculations with the UCM model, Lunsmann (1989) has extended the range of convergent solutions up to $De \sim 1.6$ by using highly refined finite element meshes. Figure 2.22(a) shows a comparison of the numerical predictions for $K$ obtained by each set of authors. The results of Lunsmann show an initially rapid decrease in the drag followed by an almost flat 'plateau' for $De \geq 1.5$ in which the drag reduction does not change significantly.

Boger fluids were used by Chhabra et al. (1980) to investigate the effects of elasticity in the absence of shear-thinning effects. A very slight reduction in drag was observed for low $De (< 0.1)$ in general agreement with the independent theoretical analyses of Leslie and Tanner (1961) and Caswell and Schwarz (1962), which predict an $O(De^2)$ departure from the Newtonian drag (see equation (2.18)). A rapid and much greater reduction in drag was observed for $De \geq 0.1$, which reached an asymptotic value of 26% for $De = 1.0$. Mena et al. (1987) presented a comprehensive series of experiments demonstrating the effects of fluid rheology and the diameter ratio. A number of different sized spheres and tubes allowed the diameter ratio $\beta$ to be varied from 0.05 to 0.5. The effects of shear-thinning and elasticity were isolated by using Newtonian, inelastic, viscoelastic, and highly elastic Boger fluids. For the constant-viscosity PAA/CS Boger fluids, elasticity resulted in an initial quadratic reduction in drag which became asymptotic.
Figure 2.22  (a) Comparison of various authors' numerically calculated drag reduction for a sphere falling axially in a tube (from Lunsmann, 1989). (b) Comparison of the calculated normalized wall correction factor $X$ (from the data of Lunsmann, 1989) with the experiments of Mena et al. (1987).
for $De > 1$, in agreement with Chhabra et al. The viscoelastic drag reduction data for each value of $\beta$ could be reduced by plotting a normalized wall correction factor defined by $X \equiv K/K_N$, where $K_N$ is the Newtonian wall correction factor for the same value of $\beta$. In Figure 2.22(b) we show a comparison between the data from Mena et al. for an 0.02% PAC/CS Boger fluid and the factor $X$ derived from the calculations of Lunsmann et al. The agreement is very encouraging, considering that the calculations are for a UCM model, while Boger fluids are more accurately modeled by an Oldroyd-B constitutive equation (Binnington and Boger 1985). The UCM calculations show both the initial rapid decrease in drag for $De < 1$, and the constant plateau approached for $De \geq 1.5$. For the viscoelastic fluid, Mena et al. found that a shear-thinning viscosity resulted in greater drag reduction and extension of the quadratic region to much higher Deborah numbers ($De \sim 3$) than would be expected from the range of validity of the small perturbation theories. At high $Re$, elastic effects were found to be dominated by shear-thinning effects and the drag reduction for the viscoelastic and inelastic fluids, as measured by the ratio $C_D(De)/C_D^0$, could be correlated simply with knowledge of the shear-thinning viscosity $\eta(\dot{\gamma})/\eta_0$.

Recent experiments with PIB/PB Boger fluids (Chmielewski et al. 1990; Tirtaatmadja et al., 1990) have investigated wall effects and drag reduction for highly elastic fluids in more detail. The measurements of Chmielewski also revealed that the evolution of the drag coefficient $C_D(\beta, De)$ is different for PIB/PB and PAC/CS Boger fluid formulations. Measurements in the PAA/CS fluid for $De \leq 0.7$ showed a monotonic decrease in the drag, in agreement with the earlier measurements of Chhabra et al. and Mena et al. discussed above. However, measurements for a PIB/PB Boger fluid with the same steady-shear viscometric properties $\eta, \Psi_1$, showed a very small initial decrease ($\approx 6\%$) in the drag coefficient ratio $X \equiv C_D(De)/C_D^0$ for $De \leq 0.3$, followed by a larger drag increase (with respect to the Newtonian value) at higher $De$. At the maximum attainable Deborah number of $De = 1.6$ the drag was found to be 20% above the Newtonian value. The dissimilar behavior of the two Boger fluids indicate that the molecular environments of the PIB and PAC are not the same, even though rheological measurements of the viscometric properties in steady shear flow are identical. The stagnation flow in the wake of the sphere is a strongly extensional flow and it thus seems probable that the differences in drag behavior result from different extensional properties of the PIB and PAC molecules. Very recent calculations at MIT by Lunsmann (McKinley et al., 1991c) with the Chilcott–Rallison constitutive model (which incorporates finite molecular extensibility) can reproduce these results, at least qualitatively. For a sphere-tube ratio of $\beta = 0.125$, these calculations show that flows of dumbbells with a low extensibility lead to a monotonic drag reduction, whereas highly extensible dumbbells result in a small drag.
reduction followed by a larger drag increase. Similar results have also been obtained in unbounded flows by Chilcott and Rallison (1988).

Of most direct interest to the thesis research presented in Chapter 7 are the instability measurements of Bisgaard (1983): A single-color LDV system was used to measure the velocity fields around spheres and bubbles in tubes with diameter ratios of $0.1 \leq \beta \leq 0.5$. The test fluid employed was a highly elastic solution of 1.0 wt% polyacrylamide in glycerine with a zero-shear rate-relaxation time of $\lambda_0 = 12$ seconds. In addition to the negative wake effect, rapid fluctuations were consistently measured in the centerline velocity at high Deborah numbers ($De_0 > 30$, based on the zero-shear-rate fluid properties). Measurements of the centerline velocity components ($v_z$, $v_r$) for a sphere of radius $a = 6$ mm falling in a cylindrical tube of radius $b = 50$ mm are presented in Figure 2.23. The axial velocity component shows rapid fluctuations in the downstream wake behind the sphere. In addition, the data in Figure 2.23 indicate that these perturbations appear to be primarily one-dimensional and fluctuations in the radial component, if any, are below the resolution of the LDV system. Subsequent measurements by Bisgaard also showed that these fluctuations in the axial velocity were localized to the wake region near the rear stagnation point, and the flow far upstream and downstream remained steady. Analysis of the Fourier spectrum indicated the oscillations were aperiodic, with no dominant frequency. This instability was determined to be elastic in origin by comparing the time-dependent results to the steady LDV measurements obtained with spheres falling in Newtonian fluids at the same Reynolds number. The experiments also demonstrated that the effects of inertia and elasticity counteract each other: increasing the Deborah number of the flow resulted in more severe fluctuations; however, experiments at higher Reynolds number showed that inertial effects tended to dampen these velocity oscillations.

The similarity between these LDV observations of unstable flow in the uniaxial extensional flow of a viscoelastic fluid in the wake of a sphere and the measurements in this thesis for planar extensional flow in the wake of a cylinder are discussed in Chapter 7.
Figure 2.23 Axial and radial velocity components for viscoelastic flow around a sphere falling along the centerline of a cylindrical tube at $De_0 = 30$. The upstream and downstream stagnation points are marked '1' and '2'. The data shows aperiodic fluctuations in the velocity that indicate an instability in the wake behind the sphere. (Reproduced from Bisgaard, 1983).
2.3.4 Summary of Viscoelastic Flow Around Cylinders and Spheres

Although the picture is still incomplete, the basic aspects of steady, non-Newtonian flow past a sphere or cylinder are now reasonably well understood. A general overview of the flow characteristics may be compiled from the results of the literature survey discussed above, and the evolution of the drag coefficient with $Re$ and $De$ may be summarized as follows:

Initially, at $Re << 0$ and $De = 0$, the drag coefficient $C_D^0$ is described by the solution presented by Lamb using the Oseen equations of motion (equation (2.13)). Careful experiments and accurate perturbation theories both indicate that the introduction of tiny amounts of elasticity to the creeping flow ($De << 1$, $Re << 1$) results in an $O(De^2)$ reduction in drag and an $O(De)$ downstream displacement of the streamlines. For $De > 1$ and $Re << 1$, i.e. highly elastic flows in the absence of inertial effects (such as the conditions experienced in experiments with Boger fluids), the streamlines are shifted downstream and the change in drag coefficient approaches a constant. This asymptotic plateau may either be above or below the Newtonian value, depending on the extensional rheology of the fluid. At very high $De$, flow transitions may occur resulting in a negative wake or the onset of time-dependent flow. For purely Newtonian fluids at high $Re$ the drag coefficient monotonically decreases and inertia results in a shift of the streamlines downstream followed by the development of recirculating eddies. However, the presence of small quantities of polymer in strongly inertial flows ($De \sim 1$, $Re \gg 1$) results in an increase in the drag, relative to the Newtonian value, and the development of a stagnant boundary layer region around the flow. This flow structure is consistent with mathematical representations of a change of type in the governing equation set. In shear-thinning viscoelastic fluids such as semidilute aqueous solutions at high $De$, inertia and shear-thinning effects dominate the effects of elasticity, and the drag reduction can be simply correlated with knowledge of the fluid's shear-thinning viscosity. Meanwhile, the presence of constraining walls near the cylinder or sphere is found to increase the measured drag coefficient significantly and also to enhance the influence of elasticity on the flow.

The only significant disagreement with this summary are the anomalous results of Ultmann and Denn (1971), which suggest both theoretically and experimentally a large upstream shift in the fluid streamlines for flow past a cylinder at low $De$ and low $Re$. The inconsistencies in their theoretical approach have been discussed by Zana et al. (1975) and a correct analysis has been presented by Mena and Caswell (1974). The photographic evidence is more difficult to dispute, however only a single picture at one value of $De$ was presented. Additionally, although it has never been noted in the literature, this upstream
displacement may originate from the unconventional experimental geometry employed by Ulmann and Denn: creeping flow past a cylinder was approximated by mounting a narrow rod (of diameter 3/8") transversely in a circular pipe of diameter 5 1/2" rather than a planar channel. The flow is thus three-dimensional throughout the domain and may not be a true representation of the results expected for uniform two-dimensional flow past a cylinder. This will not be a problem in the experiments discussed in Chapter 7, and the LDV measurements will be used to determine on a point-wise basis the direction and magnitude of the streamline shift arising from elastic effects in the PIB/PB Boger fluid.
Chapter 3

Experimental System

Two separate laser-based optical techniques have been employed in this thesis to provide non-intrusive measurements of the velocity fields in each of the model flow problems. A video-based imaging system permits rapid, qualitative visualization of the global velocity field in each flow, and laser Doppler velocimetry measurements provide a quantitative determination of the spatial and temporal structure of the local flow dynamics. The fundamental principles of each technique are presented in this Chapter, together with a discussion of the design considerations associated with the construction of the test geometries used in the experiments.

3.1 Laser Doppler Velocimetry

Laser Doppler velocimetry (LDV) is a non-invasive optical technique for accurately measuring individual velocity components in a complex flow field. It has a wide dynamic range, is capable of resolving kinematic information on highly localized length scales, and can measure both steady and time-dependent velocities. For these reasons, LDV has been used extensively in many areas of fluid mechanics research, from creeping and laminar flows to high speed turbulent and transonic flows (see for example Goldstein, 1983; Adrian et al., 1989). The LDV technique is of particular utility in viscoelastic fluid mechanics since it is non-perturbative to the local flow field being studied. More traditional measuring techniques, however, are known to be associated with well-documented non-Newtonian phenomena that lead to systematic deviations in kinematic data; common examples include the 'hole-pressure effect' in recess-mounted pressure transducers and the Uepler effect associated with the motion of tracer bubbles. More details can be found in Bird et al. (1987a). A comprehensive listing of previous LDV investigations in viscoelastic flow has been compiled by Raiford (1988), and discussions of LDV measurements with
direct relevance to the flow geometries studied in this thesis have been individually presented in Chapter 2. In this Section the fundamentals of the LDV technique are briefly reviewed together with a more detailed account of the implementation of the three-color system developed in this thesis.

3.1.1 Principles of Laser Doppler Velocimetry

A thorough description of the principles of LDV can be found in the monographs by Drain (1980) and Durst et al. (1981); a brief overview is given below.

The measurement of velocities in a flowing medium using LDV is based on detection and subsequent analysis of the frequency of light scattered from a laser beam that is passed through the sample. Small particles in the fluid pass through the laser beam and scatter light in all directions. The frequency of this scattered light is Doppler-shifted by an amount that is directly proportional to the velocity of the particle. Even for high speed flows this frequency shift (typically about $10^3 - 10^5$ Hz) is small relative to the frequency of the incident radiation ($= 8 \times 10^{14}$ Hz for visible light) and would be impossible to detect unless the scattered light were recombined with the incident light source. This mixed, or heterodyned, light is focused into a photomultiplier and the difference between the two frequencies is determined.

There are a number of different optical configurations that have been presented in the literature for LDV measurements. The LDV apparatus employed in the current work is the most commonly used and consists of a ‘dual-beam’ system, as shown schematically in Figure 3.1. Two coherent laser beams of identical wavelength are focused together by a lens and intersect each other at a half-angle $\alpha$. The intersection point or measuring volume is roughly ellipsoidal in shape, with dimensions of between 100 and 1000 $\mu$m, depending on the initial diameter, $D_e^{-2}$, of the laser beams. The calculation of the particle velocity from this configuration can be simply explained using a ‘fringe model’: When the two incident beams cross, a diffraction pattern is established, which consists of a series of light and dark fringes that result from constructive and destructive interference of the coherent light waves. The spacing of these fringes depends on the wavelength $\lambda$ of the incident radiation, and the half-angle $\alpha$ between the beams. It can easily be shown from geometrical arguments that the fringe spacing is given by

$$d_f = \frac{\lambda}{2\sin \alpha}$$

(3.1)
Figure 3.1 Geometric arrangement for a typical ‘dual-beam’ laser Doppler velocimeter, and its interpretation in terms of the ‘fringe model’.
For the visible laser light and optical configurations commonly used in LDV, this fringe spacing is approximately \(1 < d_f < 5 \ \mu m\). If a seed particle (of characteristic dimension = 1 \(\mu m\)) moving with the fluid passes through the fringe pattern it will scatter light in all directions. The shift in frequency of this scattered light \(f_s\) can be considered to arise from the translation time required for the particle to pass from one bright fringe to the next \((= f_s^{-1})\), and thus the velocity of the particle is determined directly by dividing the fringe spacing by the time required to travel that distance:

\[
v_x = \frac{d_f}{(f_s)^{-1}} = \frac{\lambda f_s}{2\sin\alpha}
\]

(3.2)

Thus, it is from eq. (3.2) that \(v_x\) (the component of velocity normal to the fringe pattern) is directly proportional to the Doppler frequency of the light. In order to calculate the velocity component of the particle one has only to know the wavelength of the light source, the intersection angle between the beams and the Doppler shift of the scattered light.

The LDV technique does not involve any unknown ‘calibration constants’ and the accuracy of the system is limited solely by measurement of the geometric angle \(\alpha\) and accurate determination of the shift frequency \(f_s\). To measure this frequency, the light scattered from the particles passing through the fringe pattern is directed through a color filter into a photomultiplier that is focused directly at the measuring volume. The photomultiplier produces an analog voltage directly proportional to the intensity of laser light being received. A characteristic ‘Doppler burst’ from the passage of a single particle through the measuring volume is shown in Figure 3.2(a). As the particle passes through the measuring volume the intensity of scattered light rapidly rises and falls in synchronization with the pattern of light and dark fringes. These oscillations in the intensity are characterized by the Doppler frequency \(f_s\). In addition, a characteristic Gaussian shape in the overall intensity of the burst can be observed in Figure 3.2(a) since the intensity of the incident laser beams also has a normal or Gaussian distribution. The fluctuating baseline observed on either side of the burst consists of white ‘shot’ noise associated with random thermal fluctuations in the photomultiplier tube.

A number of different techniques have been developed to determine the Doppler frequency \(f_s\) of the oscillations in each burst. The newest and most direct method consists of performing a Fourier analysis of each Doppler burst and determining the dominant components of the resulting frequency spectrum. Such analysis requires the ability to repeatedly compute Fast Fourier Transforms (FFTs) in ‘real time’ and the method has only
Figure 3.2  (a) Characteristic Doppler burst generated by a single particle passing through the measuring volume; (b) Frequency spectrum of the Doppler burst calculated by using a fast Fourier transform (FFT).
become widely available in the last decade. An FFT spectrum of the Doppler burst is shown in Figure 3.2(b). The spectrum was calculated from the 1024 voltage samples in Figure 3.2(a) and the Doppler frequency can clearly be determined as \( f_s = 12.85 \text{ kHz} \). The noise identified in the baseline of Figure 3.2(a) appears in the FFT as random fluctuations which are uniformly distributed across the spectrum (hence they are ‘white’ noise) and approximately two orders of magnitude weaker than the Doppler signal of interest.

In an alternative and simpler detection system, the output from the photomultiplier is sent to a ‘Frequency Tracker’, where a voltage controlled oscillator (VCO) scans a user-specified range of frequencies until it detects the Doppler frequency. A phase-locked loop (PLL) then ‘locks on’ to this frequency and follows its evolution with time. The digital output from the tracker then consists of a single voltage value that is directly proportional to the frequency \( f_s \). This technique offers the benefits of simplicity and the ability to measure unsteady or time-dependent velocities; however it does not provide detailed information such as mean burst frequency and standard deviations about the mean that can be determined from the FFT spectrum. In addition, tracking methods require a high rate of data. The Doppler bursts from the particles in the flow are actually discrete data; however, the PLL produces a continuous voltage proportional to the Doppler frequency of these bursts. If the data rate is too low, the PLL will ‘drop out’, and the VCO will begin scanning for a new frequency. This is a particular problem in very slow flows such as those encountered in recirculations or near walls. The two data acquisition techniques are therefore complimentary to each other, and both frequency trackers and a spectrum analyzer are used in this thesis to measure the velocity components of a flow.

The principle limitation of an optical system such as that shown in Fig. 3.1 is the lack of directionality; the measurement of a Doppler shift frequency \( f_s \) allows determination of the speed of a particle \( |v_x| \) but not the direction, i.e. whether it passed through the measuring volume from top to bottom, or from bottom to top. A plot of the relationship between kinematic velocity and frequency is shown in Figure 3.3(a), and the measurement of a Doppler frequency \( f_s \) corresponds to a calculated velocity of either \(-\lambda f_s / 2 \sin \alpha\) or \(+\lambda f_s / 2 \sin \alpha\). This is unimportant in simple unidirectional flows where the direction of flow is known a priori from simple observation; however, it can be problematic in more complex flows.

A frequency-shift system is employed in order to resolve this ambiguity. One of the two coherent beams shown in Fig. 3.1 is passed through a Bragg cell consisting of a quartz piezoelectric oscillator. This oscillator is driven at a constant frequency of 40 MHz and results in an exit beam which has been shifted in frequency by a variable amount that is in the range \( 2 \text{ kHz} \leq f_0 \leq 10 \text{ MHz} \). In terms of the fringe model described above, this
Figure 3.3  (a) The relationship between the measured Doppler frequency \( f_s \) and the velocity component \( v_x \), calculated from eq. (3.2); (b) The modified velocity-frequency relationship for a 'dual-beam' LDV arrangement employing a frequency-shift system.
frequency differential between the two intersecting beams is considered to result in a fringe pattern that is no longer static but instead moves with a negative velocity that is determined from eq. (3.2) to be \(-\frac{\lambda f_0}{2\sin \alpha}\). This results in the modified velocity–frequency relationship shown in Fig. 3.3(b). A particle at rest in the measuring volume with a velocity \(|v_x| = 0\) thus generates a positive Doppler frequency since it has a positive velocity relative to the moving fringes. Similarly, a particle with a small negative velocity in the range \(-\frac{\lambda f_0}{2\sin \alpha} < v_x < 0\) appears to have a small but positive and unique frequency. The measured shift frequency \(f_s\) and the calculated velocity of the particle are then related by the following mapping

\[
\frac{\lambda f_s}{2\sin \alpha} = |v_x - v_0| = |v_x + \frac{\lambda f_0}{2\sin \alpha}|
\]  
(3.3)

For velocities \(v_x \geq v_0\) this expression can be rearranged to give

\[
v_x = \frac{\lambda(f_s - f_0)}{2\sin \alpha}
\]  
(3.4)

Such a system allows the user to superimpose a global directionality to the flow and measure local flow reversals; for example those associated with the weak recirculating vortices that develop in the axisymmetric contraction. For sufficiently high data rates it is also possible to use a frequency shift with the trackers described above to follow velocities that oscillate in time about zero (Lawler et al., 1986).

With the techniques outlined above it is possible to measure the magnitude and directionality of the velocity \(v_x\) associated a particle moving with the test fluid through the measuring volume. However, in such a dual-beam configuration, the measured component of velocity is always in the plane of the two incident beams and normal to the bisector of the beams. This is sufficient for a simple one-dimensional flow, but to determine the complete velocity vector of a particle in a complex flow it is necessary to measure all three velocity components simultaneously. Such a system is realizable by the addition of more laser beams of different wavelengths and is described in detail below.
3.1.2 Three-Component LDV System

The LDV apparatus employed in this research is a three-color, six-beam system (TSI, Model 9100-12) shown schematically in Figure 3.4. The light from a 4-Watt Argon-ion laser is collimated and separated into three separate wavelengths; green ($\lambda = 514.5 \text{ nm}$), blue ($\lambda = 488 \text{ nm}$) and violet ($\lambda = 458 \text{ nm}$) by using a prism arrangement. The blue and green beams are passed through one set of optical components, while the violet beams travel through a separate, parallel optical train. These optical components are comprised of a series of prisms, mirrors and beam spacers which split each colored beam into two identical coherent beams, resulting in a total of six beams. One beam of each color is also passed through a Bragg cell in order to implement a frequency-shift system as described in Section 3.1.1. The six beams are passed through the beam expanders and then focused to a single point by the two final focusing lenses $L1, L2$. In Figure 3.4, the two blue beams are in the vertical plane and the remaining four beams are in the horizontal plane.

The dimensions of the ellipsoidal measuring volume are determined from knowledge of the focal lengths of the lens ($L1, L2$), the intersection angle of the beams ($\alpha_G, \alpha_B, \alpha_V$), the initial separation of the beam pairs ($d$) and the beam diameter. Since the intensity of a collimated laser light source is distributed in a Gaussian fashion, this beam diameter is, by convention, taken to be the distance $D_e^{-2}$ over which the measured intensity has fallen to $1/e^2$ of its maximum value. After passing through a standard concave lens element (such as the final focusing lens), a Gaussian beam is focused and, for short focal lengths and diffraction limited lens, the narrowest beam diameter occurs very close to the focal point (Klein and Furtag, 1986). The diameter at the beam waist is then given by

$$d_w = 4\lambda F / \pi D_e^{-2}$$

(3.5)

Although the focal length $F$ of both of the final focusing lenses is nominally specified, fluctuations from these specifications are observed due to individual lens variations resulting from manufacturing, polishing and coating operations. A more accurate value for the focal length for each element $L1, L2$ is thus ascertained by accurately measuring the intersection angle between the two focused beams of each color ($2\alpha_i$ where $i = G, B, V$). From simple geometric optics the focal length is then

$$F = d / 2\tan\alpha$$

(3.6)
Figure 3.4  Schematic Diagram showing the optical and geometric arrangement of the three-color, six-beam laser Doppler velocimetry system.
and the minimum diameter of the beam at its waist is therefore

\[ d_w = \frac{2\lambda d_e}{\pi D_e \cdot \tan \alpha} \tag{3.7} \]

The length and diameter of the ellipsoidal measuring volume can be determined from similar geometric arguments, and the spacing of the interference fringes \((d_f)\) formed at the intersection of each beam-pair is determined from eq. (3.2). Although these fringe patterns overlap spatially, the wavelengths \((\lambda)\) of the three colors of laser light are different and therefore the fringe spacing in each direction is different. Thus, in this configuration the system is essentially three independent dual-beam systems, each capable of measuring a single velocity component. The final optical and geometric characteristics for each colored beam-pair are specified in Table 3.1 below.

<table>
<thead>
<tr>
<th>Beam Color</th>
<th>Green</th>
<th>Blue</th>
<th>Violet</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength ((\lambda))</td>
<td>514.5 nm</td>
<td>488 nm</td>
<td>458 nm</td>
</tr>
<tr>
<td>Half Angle ((\alpha))</td>
<td>5.97 ± 0.10°</td>
<td>7.83 ± 0.10°</td>
<td>6.25 ± 0.10°</td>
</tr>
<tr>
<td>Fringe Spacing ((d_f))</td>
<td>2.47 µm</td>
<td>1.79 µm</td>
<td>2.10 µm</td>
</tr>
<tr>
<td>Number of Fringes</td>
<td>21</td>
<td>21</td>
<td>21</td>
</tr>
<tr>
<td>Measuring Vol. Diam. ((d_m))</td>
<td>53.3 µm</td>
<td>37.7 µm</td>
<td>45.3 µm</td>
</tr>
<tr>
<td>Measuring Vol. Length ((l_m))</td>
<td>510 µm</td>
<td>274 µm</td>
<td>414 µm</td>
</tr>
</tbody>
</table>

Table 3.1 Geometric specifications for the measuring volumes associated with each individual beam-pair.

It is seen from equations (3.2) and (3.7) that calculations of both the fringe spacing and the size of the measuring volume are dependent on accurate determination of the intersection angle \(\alpha_i\) for each beam-pair. To measure this angle accurately in the laboratory, an ‘optical protractor’ has been constructed by mounting a laser-compatible front-silvered mirror on a high-precision rotation stage (Newport, Model 472). This stage permits accurate axial rotation of the mirror through 360° with a direct dial readout accuracy of ± 0.02° and an additional vernier scale accurate to ± 0.002°. To determine the intersection angles \(\alpha_i\), the protractor was mounted on the optical bench and aligned such that one of the
laser beams was incident on the mirror and reflected directly back down its own path. The mirror was then rotated on the stage until the incident beam had swept through an angle $2\alpha_i$ and reflected directly down the second beam path. Despite the high resolution of the rotation stage, accuracy was ultimately limited by the ability to visually determine the point at which both beams superposed on each other. Since the beam intensities are Gaussian, such a determination requires use of a laser power meter to determine accurately the center of the beam-spot corresponding to maximum light intensity. Such a power meter was not available and the intersection criterion was measured visually. For beams of diameter $D_e^{-2} = 1.3$ mm and a focal length of $F \approx 298$ mm, the maximum uncertainty in the angle was found to be only $\pm 0.10^\circ$. This results in a maximum error of approximately $\pm 1.7\%$ in the determination of the fringe spacing in eq. (3.2) and in subsequent calculations of the velocity.

The Doppler shifted light that is scattered by particles in the flow is measured in an off-axis, side-scatter mode by three photomultipliers (PM) as shown in Figure 3.4. The violet scattered light is collected through the blue-green focusing lens $L1$, while the blue and green light passes through the violet focusing lens $L2$. The analog voltage output from the PM tubes is directed to three individual frequency trackers (DISA, Model 55N20/21) and a dual channel Spectrum Analyzer (Nicolet, Model 660B) in order to determine the Doppler frequency measured by each beam-pair. The digital output from each of these devices is captured in the computer via standard RS232 and IEE 488 interfaces, respectively. The entire optical train shown in Figure 3.4 is mounted on a computer-controlled, three-dimensional translating table (TSI, Model 9500) which enables point velocity measurements to be made throughout the flow geometry. The final system specifications of the three-color LDV system are given in Table 3.2 below.

<table>
<thead>
<tr>
<th>Optical Axis Separation Angle ($\theta$)</th>
<th>$26.87 \pm 0.10^\circ$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Included Angle ($2\theta + \alpha_r + \alpha_v$)</td>
<td>$65.96^\circ$</td>
</tr>
<tr>
<td>Overall Measuring Volume Dimensions</td>
<td>$38 \times 53 \times 270 \mu$m</td>
</tr>
<tr>
<td>Table Translation Range ($\Delta x, \Delta y, \Delta z$)</td>
<td>$96 \times 48 \times 48$ cm</td>
</tr>
<tr>
<td>Positioning Accuracy</td>
<td>$\pm 4 \mu$m</td>
</tr>
<tr>
<td>Velocity Measuring Range</td>
<td></td>
</tr>
<tr>
<td>Steady State</td>
<td>$0.04 - 400$ cm/s ($\pm 1%$)</td>
</tr>
<tr>
<td>Time-Dependent</td>
<td>$0.1 - 1000$ cm/s ($\pm 2%$)</td>
</tr>
</tbody>
</table>

**Table 3.2** Specifications of the three-component LDV system.
3.1.3 Special Alignment Considerations for a Six-Beam LDV System

In addition to the general characteristics of the LDV system described above there are a number of special considerations that must be carefully addressed in this six-beam geometry.

(i) Non-Orthogonal Velocity Components.

The velocity component measured by each independent dual-beam system is in the plane of the two incident beams and normal to the bisector of the beam-pair. Thus, the blue beam-pair measures a vertical component \( v_B \), while the green and violet beams each measure a horizontal component, \( v_G \) and \( v_V \) respectively. In a laboratory Cartesian coordinate frame, the vertically-aligned blue beam-pair directly determine the vertical component of velocity (into the plane of the page); however, it is seen from Figure 3.4 that the two horizontal components are non-orthogonal and the velocity components of interest, \( v_x \) and \( v_y \) must be resolved from the measured velocities \( v_G \) and \( v_V \). This calculation depends on the relative orientation of the flow cell and the LDV system, as shown in Figure 3.5. For the experimental measurements conducted in this thesis, the flow cell is positioned normal to the bisector of the violet and green beam-pairs, which converge at a half-angle \( \theta \).

A Cartesian coordinate system \( \{x,y,z\} \) is defined as shown with the \( z \)-axis pointing into the plane of the paper. From the vector diagram shown in Figure 3.5 the orthogonal velocity components are given by

\[
\begin{align*}
v_x &= (v_G + v_V) \sin\theta / \sin 2\theta \\
v_y &= (v_G - v_V) \cos\theta / \sin 2\theta \\
v_z &= v_B
\end{align*}
\quad (3.8)
\]

The above equations are valid for any three-dimensional velocity vector \( (v_x, v_y, v_z) \). If the aspect ratio \( (\Delta x: \Delta y) \) of the flow cell is high then the flow field is essentially two-dimensional and the \( x \)-axis corresponds to a neutral direction. An example of such a flow is the planar-slit/constrained-cylinder geometry shown previously in Figure 2.11. In this case, analysis of the vector diagram shows that \( v_G = v_V \) and equation (3.8) can be simplified to give
Figure 3.5  Vector diagram showing the orientation of the incident green and violet beam-pairs with respect to the Cartesian coordinate system of the flow cell.
\[ \begin{align*}
    u_x &= 0 \\
    v_y &= v_G / \sin \theta \\
    v_z &= v_B
\end{align*} \quad (3.9)

In this limit, the magnitude of the green component of velocity \( v_G \) that is physically measured by the LDV system is equal to \( v_y \sin \theta \). For a bisector angle of \( \theta = 26.87^\circ \) (from Table 3.2) this gives \( v_G = 0.45v_y \); i.e. the experimentally measured value is only 45% of the actual velocity. For slow flows, this reduces the detection range available in the horizontal velocity component and also amplifies the measuring error associated with each data point. These limitations must be taken into account in measurements of the velocity components obtained in the cylinder flow geometry.

Such a non-orthogonal, three-color LDV system is thus best suited to strongly three-dimensional flows in which all three velocity components are approximately of equal magnitude. More recent three-dimensional LDV implementations involve the application of fiber-optic techniques which allow relatively straightforward alignment of three mutually orthogonal beam-pairs. These systems are considerably more versatile, especially in weakly three-dimensional flows such as those studied in this thesis.

(ii) Refraction Effects.

As the laser beams pass from the external propagation medium (usually air) into the flow cell they undergo refraction due to the substantially different refractive indices of the two materials. If the incident beam intersects the flow cell at an angle \( \phi \) to the normal vector, then the magnitude of this refraction is determined from Snell’s law

\[ n_r \sin \phi^* = n_a \sin \phi \] \quad (3.10)

where \( n_a = 1.00 \) is the refractive index of air, and \( n_f \) is the refractive index of the fluid. Since \( n_f > n_a \), eq. (3.10) indicates that \( \phi^* < \phi \) and hence the beam is bent towards the normal.

If the air-flow cell interface is curved then calculation of the intersection point of two converging laser beams will depend on their relative positions with respect to the curved surface. Such refraction problems commonly occur for one- or two-color LDV measurements performed in axisymmetric geometries and may be alleviated by placing the curved flow cell inside a ‘viewing box’ of square-cross section that contains fluid of the same refractive index as the flow cell (Walters and Webster, 1982; Raisford, 1988). This
technique has been employed in the measurements described in Chapter 6 for the axisymmetric contraction geometry.

Refraction effects are still more complex in the six-beam system, since all four beams in the horizontal plane shown in Figure 3.5 intersect with the planar flow-cell wall at different angles $\phi_i$ and will thus be refracted by differing amounts. This results in a modified geometric picture of the intersection point, as shown in Figure 3.6. Since each beam is refracted to a different degree, six beams that were precisely aligned in air, will not all intersect each other at a single point in the fluid. To further complicate matters, these intersection points will vary depending on the initial separation of the four beams as they intersect the planar flow cell interface. For these reasons, the final alignment of the non-orthogonal six-beam system must be carried out in the test medium with a depth of fluid characteristic of the problem to be studied.

To perform this alignment, a small plexiglass box of square cross-section was filled with the Boger fluid and an anodized sheet of aluminum was placed in the box at a depth of $y_1 = 2.54$ cm behind the front wall. This depth is characteristic of the distance from the outer plexiglass wall to the centerline of the cylinder for the flow geometry design discussed in Section 3.3.1. Applying a frequency shift $f_i$ to one beam of each color $i (= B, G, V)$ imparts an apparent velocity of $+\lambda f_i / 2 \sin \alpha_i$ to the stationary wall, as described above in Section 3.1.1. The final beam adjustment and focusing of the PM tubes is then performed to optimize the visibility and signal/noise ratio of the Doppler bursts output from each channel.

An analytical analysis of the distortion in the y-direction that is shown in Figure 3.6 has also been performed. By using simple geometric optics it is easy to calculate the refraction of each beam at the air-plexiglass interface and the resultant change in the final intersection point. For an alignment depth of $y_1 = 2.54$ cm, the offset is $y_0 = 1.09$ cm. In addition to this offset, one must also consider the effects of moving the translating table with respect to the flow cell. While such a translation does not affect the measured angles $\theta$, $\alpha_G$, $\alpha_V$ it does affect the initial separation of the four beams as they intersect the plexiglass and the path depth through the fluid. A translation in the x-direction does not change the analysis shown in Figure 3.6; however, for a table translation in the y-direction of magnitude $(0, \Delta y)$, the green measuring volume moves in the fluid by an amount $(\Delta x^*, \Delta y^*)$ given by

$$
\Delta x^* = \Delta y \left[ \frac{\tan \theta \tan (\theta^* - \alpha_G^*) - \tan \theta^* \tan (\theta - \alpha_G)}{\tan \theta^* - \tan (\theta^* - \alpha_G^*)} \right] \quad (3.11a)
$$
Figure 3.6 Analysis of refraction effects on the green and violet beam-pairs at the air-plexiglass interface of the flow cell.
and
\[
\Delta y^* = \Delta y \left[ 1 + \frac{\tan \theta - \tan \theta^* - \tan(\theta - \alpha^*_G) + \tan(\theta^* - \alpha^*_G)}{\tan \theta^* - \tan(\theta^* - \alpha^*_G)} \right]
\] (3.11b)

Similarly the displacement of the violet measuring volume can be calculated from eq. (3.11a, b) by replacing \( \alpha_G, \alpha_G^* \) with \( \alpha_V, \alpha_V^* \). In these expressions the angles \( \theta^*, \alpha_G^*, \alpha_V^* \) are the beam angles following refraction in the fluid and are determined by using Snell’s law (eq.(3.10)), with \( n_a = 1.00 \) and \( n_f = 1.49 \). It should be noted that this analysis is not exact since the difference between the refractive index of the fluid and the plexiglass has been neglected; however, the experimental measurements presented in Chapter 4 indicate these values differ by less than 1% and for small angles of incidence this discrepancy is unimportant.

These equations can be numerically evaluated by using the initial angles specified in Table 3.1 and Table 3.2, and for the six beam configuration used in this thesis a general displacement of the translation table by \( \{\Delta x, \Delta y, \Delta z\} \) results in a movement of the measuring volume of

\[
\begin{align*}
\Delta x^* &= \Delta x - 0.0475\Delta y \\
\Delta y^* &= 1.582\Delta y \\
\Delta z^* &= \Delta z
\end{align*}
\] (3.12)

The results of eqs. (3.11) and (3.12) show that even for a unidirectional movement of the translation table given by \( \{0, \Delta y, 0\} \), the measuring volume moves in both the \( y \)-direction and the \( x \)-direction. For a given displacement of \( \Delta y \) the motion of the measuring volume is amplified by 58% in the \( y \)-direction. This correction is thus extremely important in calculation of accurate velocity profiles. In addition, since the green and violet beam-pairs are in different quadrants of the Cartesian coordinate system, the movement \( \Delta x \) of each beam-pair is of opposite sign. Thus the violet and green measuring volumes gradually diverge from each other as the table is moved in the \( y \)-direction. This results in a progressive worsening of the signal strength measured by the PM tubes and limits the depth of focus obtainable with a non-orthogonal three-color system. Experiments and calculations show that reliable data can be obtained for a table displacement of up to \( \Delta y = \pm 4.0 \text{ cm} \), corresponding to coincident movement of the green and violet measuring volumes by approximately \( \Delta y^* = \pm 6.32 \text{ cm} \) in the \( y \)-direction and a separation by an amount \( \Delta x \equiv 0.19 \text{ cm} \). Any larger \( y \)-displacement and the green measuring volume no
longer overlaps with the intersection point of the violet beams. Since the photomultipliers are aligned in an anti-symmetric off-axis configuration, the focal point of the green and violet PM tubes follow the intersection point associated with the beam-pair of opposite color. When the two measuring volumes separate spatially from each other the PM tubes no longer 'see' the correct measuring volume and this results in a complete loss of the Doppler signal.

(iii) Off-Axis Light Collection

The light scattered by particles moving through the measuring volume is collected in a back-scatter mode through the final focusing lenses $L_1, L_2$ as shown in Figure 3.4. This permits convenient alignment of the PM tubes and easy spatial translation of the complete optical configuration. The previous two-color, four-beam optical arrangement developed by Raiford (1988) employed direct back-scatter data collection in which the light scattered at 180° to the propagation direction was collected by the PM tubes. Since the line-of-sight in this configuration is directly along the propagation direction of the beams, the photomultipliers collect light scattered by particles passing anywhere along the entire length $l_m$ of the measuring volume. If a similar mode of collection were implemented in the three-color system described above, the velocity components determined at any point would actually consist of an average across a measuring volume of maximum length $l_m = 0.5$ mm. Since the characteristic length scale of the flow is only $R_c = 3.18$ mm this technique does not provide acceptable spatial resolution of the velocity components.

To improve this resolution, an off-axis mode of light collection is employed, and the photomultiplier for each color of laser light ‘views’ the appropriate measuring volume from an oblique angle $2\theta$. Mounted at the entrance to each photomultiplier is a pinhole of diameter 50 µm (TSI, Part # 9162). Therefore, with correct alignment, the PM tube only ‘sees’ the central portion of the ellipsoidal measuring volume and light is collected from a cylindrical slice with a diameter of approximately $(50 / \sin 2\theta) = 62$ µm. This configuration improves the spatial resolution of the six-color system by over a factor of eight, and the system is able to resolve velocity gradients accurately on the scale of 100 µm (equivalent to $0.03R_c$).

3.2 Flow Visualization System

In addition to obtaining point-wise velocity measurements with the LDV system, the global dynamics of the flow have been recorded using a video-based flow visualization
system. A schematic diagram of the experimental configuration of this system is shown in Figure 3.7. A single beam from the Ar-ion laser is initially focused through a regular convex lens and then passed through a cylindrical lens to form an expanded plane of laser light that is approximately 100 μm thick. This sheet of light passes through the plexiglass flow cell and illuminates a narrow longitudinal section of the fluid. A CCD video camera is positioned perpendicularly to the propagation direction of the light sheet and records images of particles flowing through the illuminated viewing plane. By replacing the video camera with a regular 35 mm SLR camera (Minolta X700), long time exposure 'streak photographs' have also been produced to record the fluid streamlines in the flow field.

The advantages and limitations of the streak photographs so prevalent in experimental investigations of non-Newtonian fluid mechanics have already been discussed in Chapter 1.3, and in particular it is emphasized that the technique yields only qualitative kinematic information and is limited to steady flows. The video-imaging system described above is thus used to provide an overall picture of the flow field and a qualitative understanding of the unsteady viscoelastic flows studied in this thesis. These observations then serve as guides for highly accurate LDV measurements which can quantify the temporal and spatial evolution of the nonlinear instabilities that occur in each geometry.

The magnification of the flow field possible with the video-based system depends on both the focusing lens element used and the size of the television screen used to display the image. Regular telephoto zoom lens designed for video systems possess undesirably long focal lengths and generally provide a maximum magnification of approximately 12:1. The magnification is improved by using a C-mount adapter (Panasonic, WV-LT23) so that regular 35 mm photographic quality lenses can be mounted on the video camera. Since the imaging area of the CCD is approximately four times smaller than a regular 35 mm photographic negative the linear magnification is increased by 100% but the field of view is correspondingly narrowed. Initial calibration tests with a 50 mm f/3.5 macro lens (Minolta, MD 50 macro) coupled with a 2:1 teleconverter ring and a 19” professional monitor (Sony PLV-1910) showed that a maximum magnification of 49:1 over an 8.2 mm field of view could be achieved with a focal length as low as 5 cm.

The resolution of the video display system is independent of the monitor size and depends solely on the type of lens and the pixel density of the CCD device. In the configuration described above, the image from the macro lens is focused onto a single color-mosaic CCD videochip consisting of 574 (horizontal) × 499 (vertical) pixels. This information density exceeds the bandwidth available with standard NTSC format and the spatial resolution of the system is ultimately limited by the 380 (H) × 505 (V) lines displayed by the video equipment. With this limitation the minimum resolvable feature size
Figure 3.7  Schematic diagram of the optical configuration used in video flow visualization experiments.
is approximately $\Delta x \approx 16 \mu m$. The images are stored on professional quality 1/2" VHS videotape (Maxell BQ T120) at the standard rate of 30 full frames per second. This limits the temporal resolution of the system to events occurring on a time-scale of $\Delta t = 0.033$ seconds; however, the visual 'blurring' of each video frame can be reduced by a 'strobe-shutter' facility of the D5000 camera that enables exposures of 1/1000 second to be recorded still at the rate of 30 image frames per second.

These values are considerably coarser than those obtained with regular photographic film which offers a resolution of 300 lines/mm; a single 35 mm frame thus contains $10,500 \times 7500$ pixels (Adrian, 1990) and can be exposed in 1/2000 second. Faster video cameras (up to 5000 frames per second) and higher pixel density CCDs are available (up to $2000 \times 2000$ pixels), however these devices are prohibitively expensive and also require higher definition non-NTSC storage media. The higher spatial resolution of chemical photographs thus make them suitable for highly accurate particle image velocimetry (PIV) applications, whereas the ability of a standard video-based system to record and display kinematic data in real-time makes it ideally suited to low resolution qualitative studies of unsteady flows and the onset of flow instabilities. The characteristic length scale for each of the flow geometries studied in this thesis is $R \sim 3$ mm and the characteristic time scale of the viscoelastic fluid is approximately $\lambda \sim 1$ s, and the specifications discussed above for the video-based system are thus more than sufficient to resolve steady-state and transient kinematic information in each flow.

### 3.3 Flow Geometry Design

Laser Doppler measurements reported in this thesis have been performed in two flow geometries; (i) the axisymmetric contraction geometry shown schematically in Figure 2.3, and (ii) the constrained cylinder geometry depicted in Figure 2.11. The design considerations associated with each of these flow geometries are individually discussed below.

#### 3.3.1 The Axisymmetric Contraction Geometry

The axisymmetric contraction geometry was originally constructed by Raiford and the design is discussed in more detail in Raiford (1988; Raiford et al. 1989). The flow cell consists of a large polished plexiglass tube of radius $R_0 = 2.54$ cm rigidly connected to a smaller 0.635 cm radius tube. A small polished disk 2.54 cm in radius and 3.81 cm in
height sits at the bottom of the large tube, as shown in Figure 3.8(a). A hole of radius \( R_2 = 0.318 \) cm is carefully machined through the middle of this disk to form the downstream section of the axisymmetric contraction. The contraction ratio \( \beta = R_1/R_2 \) of the geometry is then varied by inserting annular sheaths of internal diameter \( 2R_1 \) and external diameter \( 2R_0 \) into the large upstream section of the flow cell as shown in Figure 3.8(a). The maximum ratio possible with no annular insert is \( R_0/R_2 = 8 \). The length of the upstream flow section is \( 14 \) cm = \( 44R_2 \) in order to ensure that the upstream flow is fully developed as it approaches the contraction plane.

The contraction ratio \( \beta \) is known to affect the flow dynamics observed in viscoelastic entry flow profoundly, and previous experimental investigations with Boger fluids have been reviewed in detail in Chapter 2.2. In order to study systematically the effect of contraction ratio, a large number of annular inserts with differing internal diameters \( 2R_1 \) were prepared. Each insert was machined from a solid cylindrical block of plexiglass to tolerances of \( 1/1000^\circ \) (\( = 0.0025 \) cm, or \( 0.008R_2 \) in dimensionless terms) in order to ensure concentricity with the downstream section of the flow. Following machining the pieces were vapor-polished in an oven with a cross-flow of gaseous acetone in order to remove the surface corrugations resulting from the milling process. The final dimensions of the six contraction ratios studied in this thesis are shown in Table 3.3

<table>
<thead>
<tr>
<th>Inner radius, ( R_1 ) [cm]</th>
<th>0.635</th>
<th>0.953</th>
<th>1.270</th>
<th>1.588</th>
<th>1.905</th>
<th>_ (2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Outer radius, ( R_0 ) [cm](^1)</td>
<td>2.521</td>
<td>2.521</td>
<td>2.521</td>
<td>2.521</td>
<td>2.521</td>
<td>2.54</td>
</tr>
<tr>
<td>Contraction Ratio ( \beta )</td>
<td>2</td>
<td>3</td>
<td>4</td>
<td>5</td>
<td>6</td>
<td>8</td>
</tr>
</tbody>
</table>

**Table 3.3** Characteristic dimensions of the annular inserts used in the study of viscoelastic flow through abrupt axisymmetric contractions.

**Notes**

1. The outside diameter of each cylinder was specified to be \( 0.038 \) cm (\( 15/1000^\circ \)) smaller than the fixed outer tube of the geometry (diameter 5.08 cm) in order to ensure a smooth fit.

2. For the 8:1 contraction no insert is required and the upstream section simply consists of the fixed outer tube.

In all of the contraction flow experiments the same downstream section was used in order to ensure consistency in the flow conditions near the entrance to the downstream tube. To investigate the sensitivity of the observed flow dynamics on the geometry of this
Figure 3.8  (a) Detailed view of the axisymmetric contraction geometry. The contraction ratio $\beta$ is varied by inserting annular sheaths of internal diameter $2R_1$ and external diameter $2R_0$ into the large upstream tube. (b) Modified lip entrance with smooth radius of curvature $R_c = 0.5R_2$. 
re-entrant lip corner, a second downstream section was prepared as shown in Figure 3.8(b). The internal radius \( R_2 \) of the downstream tube is identical to that of the first entrance section; however the re-entrant corner was smoothed by imparting a uniform radius of curvature \( R = 0.159 \text{ cm} = 0.50R_2 \) to the lip entrance. This modified disk can be used with any of the annular inserts discussed above; however, the experimental measurements discussed in Chapter 6.4 were made exclusively in a 4:1 contraction ratio.

### 3.3.2 The Constrained Cylinder Geometry

The second geometry considered in this thesis consists of flow around a right circular cylinder that is mounted transversely in a planar slit, as shown in Figure 2.11. In order to realize this flow experimentally, the flow cell shown in Figure 3.9 (at 50\% of full-scale) was constructed. The main flow channel is formed from two 7.62 cm \( \times \) 43.2 cm (3\" \( \times \) 17\") planar sections of plexiglass that are separated by four triangular spacer blocks 1.27 cm (1/2\") thick. The entire geometry is constructed from a single large sheet of 1/2\" stock plexiglass in order to ensure maximum planarity. The side walls of the flow cell are prepared from 3.81 cm \( \times \) 43.2 cm sections of the same PMMA sheet and are attached by means of 44 countersunk 3/16\" stainless socket-head screws. To ensure a firm pressure-tight seal between the front and side-walls, two rectangular O-rings were assembled from 0.26 cm Viton rubber cord. The flow cell is connected to the braided PVC piping of the main fluid circulation system by 1\" nylon hose adapters at either end of the channel.

The final internal dimensions of the rectangular channel are: depth \( H = 1.27 \text{ cm} \), width \( W = 7.62 \text{ cm} \), and length \( L = 43.2 \text{ cm} \). The aspect ratio of the channel is thus \( W/H = 6:1 \), and is sufficiently high enough to ensure that near the centerline of the geometry edge-effects are negligible and that the flow is essentially one-dimensional plane Poiseuille flow. Experimental verification of this statement is provided by the LDV measurements presented in Chapter 7.1. Upon entering the test section the velocity profile also has to adjust from a fully developed axisymmetric pipe flow to a planar channel flow. The large upstream entry length of 13.97 cm (5.5\") is equivalent to 11\( H \) and is designed so that the flow is a fully developed channel flow well before it reaches the cylinder.

The circular cylinder itself consists of a single 8.89 cm (3.5\") length of plexiglass rod that is held rigidly in place along the centerline of the channel by 0.635 mm deep holes countersunk into the side-walls of the flow cell. It is well-known that viscoelasticity tends to amplify slight asymmetries in flow geometries (Cochrane et al. 1981; Walters, 1985) and great care was taken to ensure that the mounting holes were centered to within 1/1000 of an
Figure 3.9 Side view and front view showing the construction of the flow cell developed for LDV studies of viscoelastic flow past a circular cylinder constrained in a planar channel (Scale = 50% full-size).
inch. Since the viscoelastic test fluid and the plexiglass have almost identical refractive indices, it is possible to pass the focused LDV beams through the front face of the flow cell and directly through the transparent cylinder to obtain velocity measurements on both sides of the cylinder. Following construction of the geometry the large front face of the flow cell was repolished in order to ensure minimal disruption of the incident laser beams. The test geometry was then rigidly mounted on a large aluminum back plate in order to prevent bowing of the planar walls under pressurized flow conditions. This mounting sheet was also anodized in order to reduce the stray reflections of light arising from passage of the laser beams through the geometry.

The majority of the results presented in Chapter 7 consider flow past a cylinder of radius \( R_c = 0.318 \text{ cm} \) \((\equiv 1/8")\), such that the ratio of cylinder diameter to channel depth is \( \beta = 0.50 \). However, the design shown in Figure 3.9 permits easy removal of a channel side-wall and the substitution of another cylinder with a different diameter. It is therefore possible to explore the effect of varying \( \beta \) on the kinematics near the cylinder. Sample results for a smaller cylinder of radius \( R_c = 0.159 \text{ cm} \) \((1/16")\) are presented in Chapter 7.4. By boring additional mounting holes in the side-walls it would also be possible to study viscoelastic flow past periodic arrays of cylinders.

3.4 Fluid Circulation System

The basic design for the fluid handling system was conceived by Muller (1986) and Raiford (1988). It has recently been completely duplicated to enable the independent circulation of two different viscoelastic fluids without the rather protracted and messy operation of stripping and cleaning the pump. The complete circuit for one fluid is shown in Figure 3.10. The test fluid is circulated by a positive displacement pump (Moyno, Model #2L8) connected to a pressurized holding tank. From the tank the fluid passes through the pipes through the test geometry and a valve network before emptying into an atmospheric collection tank. At the test section a T-junction allows the fluid flow to be diverted along two separate paths so that both LDV measurements (presented in this thesis) and flow birefringence measurements (Quinzani, 1991) can be made. The recycle line can be used to control the volume flow rate through the test geometry. The total volume of Boger fluid in the flow loop is approximately 60 gallons, and the volumetric flow rate at the maximum circulation speed is found to be 2 gallons/minute. A fluid element therefore circulates once through the system in approximately 30 minutes. Since the maximum time scale of the fluid
Figure 3.10 Flow loop for circulation of the 0.31 wt% PIB Boger fluid.
is approximately 1–2 seconds (see Chapter 4.3), the macromolecules will thus be fully relaxed upon approaching the test geometry with no residual memory of their previous deformation history.

High density polyethylene (HDPE) containers, PVC drainage piping and braided, flexible PVC tubing are used throughout the system because of their long term resistance to the hydrocarbon solvent (C14) used in the viscoelastic test fluids. The large Moyno pump is designed for low shear-rate operating conditions to reduce degradation of the high molecular weight PIB polymer over a long period of time. The molecular weight distribution was monitored periodically via the gel permeation chromatography (GPC) technique discussed in Section 4.2, and no degradation could be detected over a period of two years.
In order to allow a quantitative interpretation of experimental results obtained in complex, two-dimensional flow geometries it is necessary to understand in detail the rheological behavior of the test fluid in simple one-dimensional flows. In this chapter the preparation and characterization of the PIB test fluid are discussed in detail and the material functions of the Boger fluid are determined in a wide range of simple shear flow experiments. Three different constitutive equations are presented which are capable of quantitatively describing the measured material functions with varying degrees of accuracy. Finally, appropriate dimensionless quantities which measure the relative importance of elastic and inertial effects in the Boger fluid are calculated and compared with values attained in other Boger fluid systems.

4.1 Rheological Fundamentals

4.1.1 Definitions of Test Flows

The material functions of the Boger fluid used in the flow experiments have been fully characterized in a range of steady and transient shear flows using a Rheometrics Mechanical Spectrometer discussed in detail in Section 4.1.2. A comprehensive summary of these standard, one-dimensional flows can be found in Bird et al. (1987a) together with examples of the responses observed in non-Newtonian fluids. The simple shear flow experiments used in this thesis are summarized in Figure 4.1 and briefly described below.

A. Simple Steady Shear Flow

In a simple steady shearing flow, as shown in Figure 4.1(a), a fluid sample is placed between two parallel plates and the top plate is translated at a constant velocity $V$. The velocity of the fluid at any position $y$ is given by $u_x = \gamma y$, where
Figure 4.1  The five simple shearing flow experiments used in this thesis to characterize the rheological properties of the Boger fluid.
the shear rate \( \dot{\gamma} \equiv \gamma_{yx} \) is the constant velocity gradient between the plates. The viscosity of the fluid \( \eta \) is defined in terms of the applied shear rate and the shear stress exerted by the fluid as

\[
\tau_{yx} \equiv - \eta(\dot{\gamma}) \dot{\gamma}
\]  

(4.1)

In a Newtonian fluid, the viscosity has a constant value \( \mu \), however for non-Newtonian materials it is generally a function of the shear rate.

Two additional material functions can be defined in a similar manner to characterize the normal forces exerted by a viscoelastic fluid in steady shear flow. The first and second normal stress coefficients, \( \Psi_1 \) and \( \Psi_2 \) respectively, are defined in terms of the normal stress differences, \( N_1 \) and \( N_2 \), by

\[
N_1 \equiv (\tau_{xx} - \tau_{yy}) = - \Psi_1(\dot{\gamma}) \dot{\gamma}^2
\]  

(4.2)

\[
N_2 \equiv (\tau_{yy} - \tau_{zz}) = - \Psi_2(\dot{\gamma}) \dot{\gamma}^2
\]  

(4.3)

In general, \( \Psi_2(\dot{\gamma}) \) is much smaller and of opposite sign to \( \Psi_1(\dot{\gamma}) \), and is difficult to measure experimentally (Bird et al., 1987a). For Boger fluids in particular, \( \Psi_2(\dot{\gamma}) \) is found to be almost zero (Keentok et al., 1980; Magda et al., 1991), and detailed measurements of the second normal stress coefficient have not been attempted in this thesis.

**B. Small-Amplitude Oscillatory Shear Flow**

In this type of shearing flow the top plate undergoes small-amplitude sinusoidal oscillations in a plane with amplitude \( \gamma^0 \) and frequency \( \omega \), as shown in Figure 4.1(b). Provided that inertial effects are negligible, the velocity profile between the plates is linear and the strain and shear rate in the fluid at time \( t \) are then respectively given by

\[
\gamma_{yx}(0, t) = \gamma^0 \sin \omega t
\]  

(4.4)

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

(4.5)

In general it is found that for viscoelastic materials the measured shear stress is out of phase with both the strain and shear rate. For sufficiently small strain amplitudes \( \gamma^0 \), the measured shear stress may be expressed in terms of the applied shear rate \( \dot{\gamma}^0 \) as
\[ \tau_{yx}(t) = -\eta'(\omega)\dot{\gamma}^0 \cos \omega t - \eta''(\omega)\dot{\gamma}^0 \sin \omega t \] 

(4.6)

or in terms of the shear strain as:

\[ \tau_{yx}(t) = -G'(\omega)\gamma^0 \sin \omega t - G''(\omega)\gamma^0 \cos \omega t \] 

(4.7)

In eq. (4.7), \( G' = \eta'' \omega \) is the storage or elastic modulus, and \( G'' = \eta' \omega \) is the loss or viscous modulus. Since these parameters describe the shear stress that is linear in strain, they are often termed the linear viscoelastic material functions. For reference in future discussion, two important asymptotic limits are noted: For purely elastic materials \( G' \) is equal to the constant shear modulus \( G \), and \( G'' \) is zero; whereas for a Newtonian fluid the dynamic viscosity \( \eta' \) is equal to the Newtonian viscosity \( \mu \), and \( \eta'' \) is zero. Finally it is noted that in the limits of low frequencies or shear rates, simple fluid theory (Bird et al., 1987a) predicts that the steady shear and linear viscoelastic material functions are related by

\[ \lim_{\omega \to 0} \eta'(\omega) = \eta_0 = \lim_{\dot{\gamma} \to 0} \eta(\dot{\gamma}) \] 

(4.8)

\[ \lim_{\omega \to 0} \frac{2\eta''(\omega)}{\omega} = \Psi_{10} = \lim_{\dot{\gamma} \to 0} \Psi_1(\dot{\gamma}) \] 

(4.9)

where \( \eta_0 \) and \( \Psi_{10} \) are defined as the zero-shear-rate viscometric functions.

**C. Start-Up of Steady Shear Flow**

In this experiment (Figure 4.1(c)) the growth of the stresses exerted by the fluid is measured following the inception of a steady shearing flow. The fluid sample is at rest for all times \( t < 0 \) and is assumed to be fully relaxed with all stress components equal to zero. At time \( t = 0 \), a constant velocity gradient \( \dot{\gamma}_0 \) is imposed and the stress response is observed as a function of time. Time-dependent material functions are defined analogously with eqs. (4.1) and (4.2) in terms of the shear stress and first normal stress differences as

\[ \tau_{yx}(t) = -\eta^+(t,\dot{\gamma}_0)\dot{\gamma}_0 \] 

(4.10)

\[ N_1(t) = -\Psi^+(t,\dot{\gamma}_0)\dot{\gamma}_0^2 \] 

(4.11)
A second normal stress coefficient $\Psi_2^+(t, \gamma_0)$ can also be defined, however this quantity has not been measured in these experiments. As indicated in eqs. (4.10) and (4.11), these material functions are usually found to be functions of both time $t$ and applied shear rate $\dot{\gamma}_0$.

D. Stress Relaxation After Cessation of Steady Shear Flow

A stress relaxation experiment following the cessation of steady shear flow is shown in Figure 4.1(d). For all $t < 0$, the system is assumed to be in a fully-developed steady shear flow with stress components given by eqs. (4.1) – (4.3). At time $t = 0$, the shearing flow is instantaneously halted and the fluid stresses decay with time towards their final equilibrium values of zero (assuming the material has no viscoplastic yield stress). Time-dependent material functions for the viscosity and first normal stress coefficient are then defined by

\[ \tau_{yx}(t) = - \eta^{-1}(t, \dot{\gamma}_0) \dot{\gamma}_0 \]  
\[ N_1(t) = - \Psi_1^-(t, \dot{\gamma}_0) \dot{\gamma}_0^2 \]  

(4.12)  
(4.13)

At low shear-rates these material functions are found to be independent of the applied shear rate. However, at high shear rates, shorter-relaxation-time processes dominate the response of the material and it is found that the stresses relax more quickly. Experiments have also shown that, at a given shear rate, the normal stresses relax slower than the shear stress (Bird et al., 1987a).

E. Stress Relaxation After A Sudden Shearing Displacement

The step-strain experiment depicted in Figure 4.1(e) measures the relaxation of the stresses in the fluid following a sudden shearing displacement. A large, constant shear rate $\dot{\gamma}_0$ is applied for a short time interval $\Delta t$ and the total shear strain in the experiment is $\gamma_0 = \dot{\gamma}_0 \Delta t$. The relaxation of the stresses in the material are measured and the relaxation moduli for the shear stress and first normal stress difference are defined respectively by

\[ \tau_{yx}(t) = - G(t, \gamma_0) \gamma_0 \]  
\[ N_1(t) = - G\Psi_1(t, \gamma_0) \gamma_0^2 \]  

(4.14)  
(4.15)

For small shear strains, the shear stress relaxation modulus $G(t, \gamma_0)$ is found to be independent of the applied strain $\gamma_0$, and the stress at time $t$ is linear in the applied strain.
The time-dependent relaxation modulus $G(t)$ thus contains the same linear viscoelastic information about the material as the functions $G'$ and $G''$ defined in eq.(4.7), and interconversion of these material functions is possible (Tschoegl, 1989). At higher strains it is found that the stress is no longer linear in strain, and the relaxation modulus $G(t, \gamma_0)$ decreases with increasing strain. Experiments for a number of polymeric systems have found that the relaxation of the shear stress and first normal stress difference are governed by identical time-dependent functions, the two relaxation moduli are then related by the 

*Lodge-Meissner rule* (Lodge and Meissner, 1972)

\[
\frac{G(t, \gamma_0)}{G_{\psi 1}(t, \gamma_0)} = 1
\]  

(4.16)

Unfortunately it is not possible to verify this relationship for the Boger fluid, since the mechanical spectrometer used in this study is not capable of measuring the decaying response of the normal stress difference $N_1(t)$.

In addition to the five simple one-dimensional shearing flows discussed above, material functions may also be defined to quantify the stress response of viscoelastic fluids in one-dimensional shearrfree flows. In *steady simple shearrfree flow*, the stress tensor is diagonal and only contains three non-zero components. For incompressible fluids there are thus two independent normal stress differences of interest $(\tau_{zz} - \tau_{yy})$ and $(\tau_{yy} - \tau_{xx})$. The velocity field for simple shearrfree flow is given by

\[
\begin{align*}
\nu_x &= -\frac{1}{2} \dot{\varepsilon}(1 + \kappa)x \\
\nu_y &= -\frac{1}{2} \dot{\varepsilon}(1 - \kappa)y \\
\nu_z &= + \dot{\varepsilon}z
\end{align*}
\]  

(4.17)

where $0 \leq \kappa \leq 1$ and $\dot{\varepsilon}$ is the elongation rate $\dot{\varepsilon} \equiv d\nu_2/dz$. Material functions $\overline{\eta}_1$ and $\overline{\eta}_2$ for steady shearrfree flow may then be defined in terms of these parameters as

\[
\begin{align*}
\tau_{zz} - \tau_{xx} &= - \overline{\eta}_1(\dot{\varepsilon}, \kappa)\dot{\varepsilon} \\
\tau_{yy} - \tau_{xx} &= - \overline{\eta}_2(\dot{\varepsilon}, \kappa)\dot{\varepsilon}
\end{align*}
\]  

(4.18) 

(4.19)
In these equations the elongation rate \( \dot{e} \) has been assumed to be constant; however, if it is a function of time, transient shearfree material functions may be defined analogously to eqs. (4.10) – (4.13). Particular values of the parameter \( \kappa \) define special shearfree flows as shown in Figure 4.2. For \( \kappa = 0 \) two situations are possible; uniaxial elongation (Figure 4.2(a)) corresponds to \( \dot{e} > 0 \), whereas biaxial stretching (Figure 4.2(b)) corresponds \( \dot{e} < 0 \). In both of these cases it can be seen from the symmetry of the flow that the \( x \) and \( y \) directions are identical, and thus the second normal stress difference is always zero. The function \( \eta_1 \) is then referred to uniquely as the elongational viscosity, \( \eta \). Planar elongational flow corresponds to \( \kappa = 1 \) and represents either the stretching (\( \dot{e} > 0 \)) or compression (\( \dot{e} < 0 \)) of a viscoelastic sheet with constant width. In this case, both the viscosity functions \( \eta_1 \) and \( \eta_2 \) are nonzero.

Elongational flows are very important in a large number of commercially important polymer processing applications such as fiber-spinning and injection molding, and result in pronounced extension and orientation of the macromolecules. Significant extensional flow components exist in both the contraction flow and cylinder flow geometries discussed in Chapters 6 and 7; knowledge of the elongational viscosity functions \( \eta_1 \) and \( \eta_2 \) is therefore highly desirable. However, the direct measurement of simple shearfree material functions for viscoelastic liquids is extremely difficult since it requires the absence of any solid surfaces which would give rise to shearing components in the stress tensor. A number of different experimental systems have been proposed which give a widely varying range of values for the shear free material functions (see for example the special issues \( \text{J. Non-Newtonian Fluid Mech.}, 11 \) (1981) and 35 (1990)). No experimental measurements of the elongational viscosity have been made in this thesis, however the general trends of the shearfree material functions for other similar Boger fluids are discussed in Section 4.4.4, together with the predictions of the constitutive models discussed in Section 4.3.

### 4.1.2 The Mechanical Spectrometer

The measurement of the material functions defined above have been carried out experimentally using a Rheometrics Mechanical Spectrometer (RMS-800). This rheometer is extremely versatile and allows accurate rheological measurements of the simple shear material functions to be made over a wide spectrum of test conditions: Two separate force-rebalance transducers are available; the FRT-100 ‘fluid transducer’ for low viscosity liquids permits measurements of up to 100 g-cm (0.01 N-m) in the torque (\( \phi \)), and normal forces (\( \phi \)) of up to 100 g (1 N), while the FRT-2000 ‘melt transducer’ has maximum ranges of
Figure 4.2 The deformation of a unit cell in simple shearfree flows; (a) uniaxial elongation, (b) biaxial stretching, (c) planar extension.
2000 g·cm (0.20 N·m) and 2000 g (20 N), respectively. The minimum measurable reading for each transducer was found experimentally to be 0.02% full-scale in torque and 0.10% in normal force, the total dynamic range of the system is thus $2 \times 10^{-6} \leq \mathcal{F} \leq 0.20$ N·m, and $1 \times 10^{-3} \leq \mathcal{F} \leq 20$ N. With the ‘fluids transducer’ installed, the temperature of the fluid sample can be controlled to within ±0.1 °C in the range $-5 \leq T \leq 80$ °C by using a recirculating fluid bath. With the melt transducer installed, an oven/dewar combination permits temperatures in the range $-150 \leq T \leq 500$ °C to be attained; however, the accuracy is much lower (±2 °C) and precise control is difficult near room temperatures. The motor has an operating range of $10^{-6} - 10^2$ rad/s in steady shear and a frequency range of 0.001 $\leq \omega \leq 100$ rad/s in small-amplitude oscillatory shear flow.

Two different test fixtures are available for measuring the material functions in simple shear flows as shown in Figure 4.3. The cone-and-plate device (Figure 4.3(a)) consists of a flat circular plate and a smooth conical section with radius $R$ and a small cone angle $\theta_0$. The lower plate is rotated at a constant rate $\Omega$ and the normal force ($\mathcal{F}$) and torque ($\mathcal{G}$) exerted on the upper fixture are measured. For small cone angles the shear rate across the plate has a constant value $\dot{\gamma} = \Omega/\theta_0$, and it can be shown (Bird et al., 1987a) that the material functions defined in Section 4.1.1 are given in terms of $\mathcal{F}$ and $\mathcal{G}$ as

$$-\tau = \eta(\dot{\gamma}) \dot{\gamma} = \frac{3\mathcal{G}}{2\pi R^3} \quad (4.20a)$$

$$-N_1 = \Psi_1(\dot{\gamma}) \dot{\gamma}^2 = \frac{2\mathcal{F}}{\pi R^2} \quad (4.20b)$$

In transient shear flows, the experimentally measured torque and normal force will be functions of time and equations 4.20(a) and (b) can be used to calculate the transient material functions such as $\eta^+$, $\Psi_1^-$ defined in Section 4.1.1.

In the parallel-plate geometry depicted in figure 4.3(b) two concentric parallel disks of radius $R$ are separated by a narrow gap $H$. The shear rate between the disks in this configuration is now a function of the radial position and experimental results are typically expressed as a function of the rim shear rate $\dot{\gamma}_R = \Omega R/H$ experienced at the outer edge of the disk. The material functions $\eta$ and $\Psi_1$ measured in the parallel-plate geometry are given by similar equations to 4.18(a) and (b); however, since the shearing flow between coaxial parallel disks is nonhomogeneous, the material functions are not given explicitly in terms of the measured forces, but contain additional terms of the form $\ln\mathcal{F}/\ln\dot{\gamma}$ which must be corrected for (Bird et al., 1987a).
Figure 4.3  Test geometries for rheological characterization of simple shear material functions; (a) the cone-and-plate geometry, (b) the parallel-plate geometry.
In this work, two cone-and-plate fixtures were used with radial dimensions of 1.25 cm and 3.00 cm, and cone angles of 0.1 and 0.04 radians, respectively. Parallel-plate measurements were made using two sets of plates with radii of \( R = 1.25 \text{ cm} \) and \( 3.00 \text{ cm} \). The results of these measurements for the Boger fluid are presented below in Section 4.4.

### 4.2 Composition and Preparation of Test Fluid

In 1978, Boger first presented rheological data for fluids which exhibited constant viscosity and high elasticity at room temperature. These so-called ‘Boger fluids’ consist of a high molecular weight polymer that is initially dissolved in a small amount of a low-viscosity intermediate solvent and then added to another highly viscous polymeric solvent. Rheological characterization of these fluids in viscometric flows demonstrated that they exhibit an almost constant viscosity, an initially constant first normal stress coefficient (Boger and Nguyêñ, 1978; Jackson et al., 1984; Binnington and Boger, 1985) and a vanishing second normal stress coefficient (Keentok et al., 1980). The recipe for Boger fluids is not unique and they have been prepared from organic and inorganic systems. The most common formulations consist of polyacrylamide in corn syrup (PAC/CS), polyisobutylene in polybutene (PIB/PB) and, more recently, monodisperse polystyrene in dioctyl phthalate (PS/DOP), (Magda and Larson, 1988).

Recent experimental investigations have examined the flow of Boger fluids in a variety of complex geometries; including flow through axisymmetric sudden contraction and flow around cylinders and spheres as reviewed in Chapter 2, planar contraction flows (Evans and Walters, 1986; Binding and Walters, 1988), and in complex T-shaped geometries (Binding et al., 1987). The essentially constant viscometric properties of these fluids allow them to be modeled by relatively simple constitutive equations. It is then possible to directly compare experimental data with numerical simulations of viscoelastic flow obtained using the appropriate constitutive equation (Binding et al., 1987; Crochet, 1988).

The Boger fluid formulation used in the experiments described in this thesis was originally prepared by Raiford (1988) and has subsequently been fully characterized and modeled in a variety of simple shear flows (see Quinzani et al., 1990). Its high elasticity has enabled high Deborah number \((De \geq 1)\) to be attained in experiments confined to the creeping flow regime \((Re \ll 1)\). In addition, its relatively simple behavior in viscometric
flows will hopefully allow future comparison of experimental results with new finite element simulations now being developed (McKinley et al., 1991c; Coates et al., 1991).

### 4.2.1 Boger Fluid Composition

The original Boger fluid that was developed by Boger (1977/78) consisted of polyacrylamide dissolved in water and corn syrup (Maltose syrup). However, problems have been experienced with sugar crystallization and bacterial growth in these biological systems over extended periods of time, therefore we have chosen to use a purely inert organic fluid formulation. The polymeric component is a high molecular weight polyisobutylene ($M_w = 1.8 \times 10^6$ g/mol) supplied by Exxon Chemicals (Vistanex L-120). This is dissolved in tetradecane, C$_{14}$H$_{30}$ (technical grade 90%, Borden & Remington Co.) and then added to a highly viscous polybutene solvent (Amoco Chemicals, Indopol H-100, $M_w = 1000$ g/mol). After considerable experimentation the final fluid composition was set as:

0.31 wt% Polyisobutylene (PIB)
4.83 wt% Tetradecane (C14)
94.86 wt% Polybutene (PB)

The fluid was prepared by cutting the solid, rubbery polyisobutylene into small pieces, adding the pieces to a known weight of C14 and then rolling the container continuously for two weeks. The C14 serves as a low viscosity solvent which accelerates the gradual swelling and solution of the PIB molecules. Lawler et al. (1986) used kerosene as the intermediary solvent, however tetradecane has been chosen for this study since it is better characterized (kerosene is a mix of short aliphatic chains), has a lower vapor pressure (thus reducing evaporation losses) and is also colorless and odorless. By varying the concentration of PIB in C14 it is possible to vary the viscometric properties of the resulting solution over a very wide range.

It has been shown that for a given non-dilute polymer-solvent system the material functions scale as $\eta_0 \sim (cM_w)^a$ and $\Psi_{10} \sim (cM_w)^{2a}$, where $a = 3.4$ (Bird et al., 1987a). Figure 4.4 shows plots of the zero-shear-rate viscosity and first normal stress coefficient against concentration for stock solutions of 3.0 wt% – 9.0 wt% PIB in C14. Best fit lines suggest the parameter $a = 4.1 \pm 0.3$. It can be seen from Figure 4.4 that by varying the concentration of PIB from 3.0 wt% to 9.0 wt% the viscosity is increased by two orders of magnitude and the first normal stress coefficient is increased over 1000-fold. The 6.0 wt%
Figure 4.4  Zero-shear-rate viscometric properties ($\eta_0$, $\Psi_{1,0}$) of the PIB/C14 solutions as a function of the concentration (in weight percent) of PIB. (○) zero-shear-rate viscosity; (●) zero-shear-rate first normal stress coefficient.
PIB/C14 stock solution was finally selected to achieve high fluid elasticity at a viscosity still within the design range of the Moyno pump and flow apparatus.

The base polybutene solvent is a clear, chemically stable, non-flammable polymeric liquid available in a variety of viscosity grades (Amoco Polybutenes, Technical Bulletin 12-K). Two polybutene grades, H-100 ($M_w = 1000$ g/mol) and H-300 ($M_w = 1300$ g/mol) were considered for this Boger fluid. The viscosities of both fluids were found to be constant over all shear rates, and no normal stresses were observed in these polybutenes; any slight elasticity that may be present in either grade is below the measurable limit of the Rheometrics RMS-800 normal force transducer. The H-100 and H-300 may thus be considered as essentially Newtonian solvents with viscosities $\eta_s = 23$ Pa·s and $\eta_s = 87$ Pa·s respectively. Since the initial design specifications for the fluid recirculation system required a solution viscosity under 30 Pa·s, the polybutene grade H-100 was chosen as the base solvent for our Boger fluid.

The 6.0 wt% PIB/C14 stock solution is added to the polybutene to give a final concentration of 0.31% PIB. To ensure complete mixing of the components the drums are rolled for two weeks and then allowed to stand for a further two weeks for bubbles to rise to the surface. As we have seen from Figure 4.4 the viscometric properties of the solution are highly dependent on the concentration of PIB chains in the fluid, and great care was taken to measure accurately the concentration of all three components during preparation. Although the Boger fluid is strictly a three component system of PIB, PB and C14, it is shown later in this chapter that, from a rheological viewpoint, it can be treated readily as a binary system of high molecular weight polyisobutylene dissolved in a ‘single’ well-mixed solvent of polybutene and tetradeacne. The PIB polymer molecules alone give rise to the elasticity of the Boger fluid and the PB/C14 mixture acts as a ‘single’ highly viscous Newtonian solvent. For the composition listed above, this single solvent consists of PB and C14 in relative concentrations of 95.15 wt% (PB):4.85 wt% (C14) and separate solutions of this composition were also prepared in order to allow a complete rheological characterization of the solvent properties.

The refractive index of the Boger fluid was measured using a Corning refractometer and was found to be $n_f = 1.49$. The refractive index for plexiglass is approximately $n_p = 1.50$ (depending on the residual stress in the material) and the critical angle for total internal reflection is thus $\theta_c = \sin^{-1}(n_f/n_p) = 83^\circ$. The refractive index match is extremely good and we found no need to add additional components (e.g, 2-Bromonapthalene) to increase the fluid refractive index. The fluid density was measured to be $\rho_f = 0.88$ g/cm$^3$. 

149
4.2.2 Molecular Characteristics of the PIB/PB/C14 System

The molecular weight distributions of the PB and PIB components have been determined using the technique of gel permeation chromatography (GPC). In this process dilute polymer solutions are eluted through a series of columns containing a porous gel with a carefully distributed size of pore sizes. The longer polymer molecules are excluded from the small pores due to their greater size and are thus eluted more rapidly. It can be shown that the retention time in the column of a given liquid fraction is proportional to the logarithm of the hydrodynamic volume of the polymers in that fraction (Collins et al., 1973). The hydrodynamic volume of a polymer chain is equal to $[\eta]M$ where $M$ is the molecular weight of the chain, and $[\eta]$ is the intrinsic viscosity (Flory, 1953). The intrinsic viscosity of a given polymer-solvent system can also be empirically related to the molecular weight by a Mark-Houwink expression (Bird et al., 1987a) of the form $[\eta] = KM^a$, and a universal calibration curve can then be constructed for GPC by plotting log($KM^{1+a}$) against the retention time in the column.

A Waters GPC 150C with 4 columns containing a poly(styrene-co-divinyl benzene) ‘styrogel’ of pore sizes $10^2$, $10^3$, $10^4$ and $10^5$ Å, was used to determine the PB and PIB molecular weight distributions. With this distribution of pore sizes the GPC system was able to resolve molecular weights in the range $10^3 \leq M \leq 10^7$ g/mol. Approximately 1.0 wt% Boger fluid was dissolved in toluene to form a dilute solution of PIB, PB and C14 in toluene. Samples of 50 µl were injected into the columns and the concentration of polymer eluted as a function of time was measured using a differential refractometer connected to a strip chart recorder. A universal calibration curve was constructed using almost monodisperse samples of polystyrene (PS) with molecular weights in the range 2350 – 2,700,000 g/mol, and molecular weight distribution of the PIB and PB could then be determined using the Mark-Houwink parameters for PIB and PS given in Table 4.1.

<table>
<thead>
<tr>
<th>Polymer</th>
<th>$T$ [°C]</th>
<th>$K$ [(cm$^3$/g)(mol/g)$^a$]</th>
<th>$a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>PS in Toluene</td>
<td>30</td>
<td>0.00977</td>
<td>0.73</td>
</tr>
<tr>
<td>PIB in Toluene</td>
<td>30</td>
<td>0.020</td>
<td>0.67</td>
</tr>
<tr>
<td>PB in Toluene</td>
<td>30</td>
<td>0.083</td>
<td>0.53</td>
</tr>
</tbody>
</table>

Table 4.1 Mark-Houwink Parameters for construction of the GPC universal calibration curve (from Brandup and Immergut, 1975; pp IV-9)
The final results of the GPC investigation are shown in terms of the weight fraction \( w_i(M_i) \) in Figure 4.5. It should be noted that the weight fraction is normalized such that \( \sum w_i(M_i) = 1 \) for each of the two component, and the relative magnitudes of each curve therefore do not correspond to the concentration of each polymer. The polyisobutylene has an extremely broad molecular weight distribution typical of high molecular weight industrial polymers prepared by free-radical polymerization, and the moments of the distribution were calculated to be \( M_n = 5.6 \times 10^5 \) g/mol and \( M_w = 1.8 \times 10^6 \) g/mol. The polydispersity of the PIB is thus \( p = \frac{M_w}{M_n} = 3.2 \). The polybutene component also has a broad molecular weight distribution; however, it was not possible to fully characterize the fractions of molecular weights \( M_i \leq 900 \) g/mol with this GPC/refractometer/solvent combination. Although the lower molecular weight fractions have a large effect on the number distribution, these components have only a small weighting on the mass-average molecular weight, which was determined for the PB (H-100 grade) to be approximately \( M_w = 1500 \) g/mol; in reasonable agreement with the technical specification of 1100 g/mol (Amoco Polybutenes, Technical Bulletin 12-K).

In order to understand the viscometric properties of the Boger fluid it is instructive to consider the local molecular configuration of the PIB/PB/C14 ternary system. The PIB polymer consists of long unbranched aliphatic molecules with a repeat unit of \([-\text{CH}_2\text{C(CH}_3\text{)}_2\text{]}.\) The repeat unit has a mass \( m_0 = 56 \) g/mol and the polymer has a mass-average molecular weight of \( M_w = 1.8 \times 10^6 \) g/mol. For a freely rotating chain of aliphatic carbon atoms, with a fixed bond angle of \( \theta = 109.5^\circ \) and bond lengths of \( l = 1.54 \) Å, the mean square end-to-end length of the randomly coiled polymer is given by (Flory, 1953)

\[
\langle r^2 \rangle_{ef} = \left( \frac{1 + \cos \theta}{1 - \cos \theta} \right) \left( \frac{2M_w}{m_0} \right) l^2
\]  

(4.21)

For the PIB polymer molecules the r.m.s end-to-end length of the freely-rotating chains is thus \( \sqrt{\langle r^2 \rangle_{ef}} = 5.5 \times 10^{-8} \) m. However, due to the presence of the pendant \(-\text{CH}_3\) groups along the chain backbone plus excluded volume effects, the actual polymer chains will be considerably more expanded than this value. This additional expansion can be calculated from knowledge of the chain configuration at the theta-condition (\textit{i.e.} at the condition where the chain behaves exactly as a random coil). At room temperature of 24°C benzene is a theta solvent for polyisobutylene and from the Mark-Houwink parameters \( (K = 107 \times 10^{-3} \text{ cm}^3/\text{g}, a = 0.50; \text{Brandup and Immergut, 1975}) \) the actual r.m.s end-to-end length is thus calculated to be \( \sqrt{\langle r^2 \rangle_o} = 1.0 \times 10^{-7} \) m. The configuration of polymer
Figure 4.5  The molecular weight distribution of the high molecular weight polyisobutylene polymer and the low molecular weight polybutene solvent.
chains is generally expressed in terms of a characteristic ratio, $C$ which represents the deviation of the mean-square length from that of an ideal three-dimensional random walk,

$$\langle r^2 \rangle_0 = CNl^2$$  (4.22)

where $N = 2M_w/m_0$ is the number of steps in the random walk. Dividing the numerical values of the mean square end-to-end lengths calculated for the PIB solution results in a characteristic ratio of $C = \langle r^2 \rangle_o \langle r^2 \rangle_{of} = 6.70$. Steric hindrance effects thus result in a mean square length for the PIB macromolecules that is almost seven times that calculated from a simple random walk argument.

At conditions away from the theta point, scaling arguments can be used to investigate the conformations of the polymer chains. For the case of a dilute solution of monodisperse long polymer chains in an athermal solvent (i.e. one with no enthalpy of mixing) it is well known that the long chains are expanded relative to an ideal random walk, and the end-to-end length scales as $\sqrt{\langle r_N^2 \rangle} ~ N^{3/5}l$ (Flory, 1953). The effects of this expansion can be observed in the stronger molecular-weight-dependence of the Mark-Houwink parameters ($a > 0.5$) listed in Table 4.1 for dilute polymer solutions away from the theta temperature. Meanwhile, for the case of dilute long polymer chains dissolved in a polymeric melt, consisting of short chains with degree of polymerization $P$, two distinct conformational behaviors are predicted (De Gennes, 1975). If the shorter chains have length $P \geq N^{1/2}$ then both the short and long chains behave ideally with lengths given by equation (4.21). However, if the melt consists of chains with $P \leq N^{1/2}$ then the homogeneous melt remains ideal while the long chains are expanded with an end-to-end length given by $\sqrt{\langle r_N^2 \rangle} ~ N^{3/5}P^{-1/5}l$. Physically, the short $P$ chains behave as a reasonably good solvent for the long chains; however the expansion is weaker by a factor of $P^{-1/5}$ than that observed in a regular non-polymeric solvent.

For a ternary system that consists of polydisperse long polymer chains (PIB) polydisperse shorter chains (PB) and an athermal solvent (C14), the situation will be considerably more complicated still. However, the idealized case of an athermal, bimodal solution consisting of monodisperse long chains ($N$), short chains ($P$) and solvent ($S$) has recently been considered in detail by Joanny et al. (1981). Scaling arguments and renormalization theory lead to six distinct conformational regimes that depend on the volume fractions $\phi_N$, $\phi_P$ and $\phi_S$ of the three components. For a concentrated solution of polymer chains ($\phi_N + \phi_P \gg \phi_S$), Joanny et al. find that the scaling behavior of the long and short chains is similar to that observed in the melt, and the solution exhibits continuous
ideal behavior with end-to-end lengths given respectively by $\sqrt{\langle r_{N}^2 \rangle} \sim N^{1/2}(\phi_N + \phi_p)^{-1/8}$ and $\sqrt{\langle r_p^2 \rangle} \sim N^{1/2}(\phi_N + \phi_p)^{-1/8}$.

For the case of Boger fluid systems, the high volume fraction of polymeric solvent ($\phi_p \sim 0.95$), coupled with the weak $1/8$-power dependence on the volume fractions results in almost perfectly ideal behavior of the long macromolecules. Of course the situation in our PIB/PB/C14 Boger fluid is complicated by the high polydispersities; however, it appears reasonable to use eq. (4.22) to calculate the size of the PIB chains, rather than a typical dilute solution scaling exponent. Finally, it is interesting to note that all Boger fluid formulations prepared to-date consist of small amounts of polymeric components with molecular weights $\sim O(10^6 \text{ g/mol})$ added to viscous polymeric solvents of mol. wt. $O(10^3 \text{ g/mol})$, and these systems are therefore very close to the melt cross-over point at $P = N^{1/2}$, where the scaling of the conformational behavior changes its functional dependence.

In addition to the end-to-end length, it is useful to consider the contour length, $L$ of the macromolecules. This is the maximum length of the PIB chains that could theoretically be attained by completely unravelling the randomly coiled molecules. For an aliphatic carbon chain, such as PIB, the contour length is given by

$$L = \left( \frac{2M_w}{m_0} \right) l \cos^{1/2} (180 - \theta)$$

(4.23)

For the PIB polymer used in this research, the contour length is $L = 8.1 \times 10^{-6}$ m. This is an almost macroscopically observable length scale; however, it would be almost impossible in practice to effect such an extension of the polymer chains, except in an extremely strong and persistent extensional flow.

From the molecular parameters calculated above it is also possible to calculate a critical entanglement concentration $c^*$ for the PIB molecules. Below this critical concentration ($c < c^*$) the solution is dilute and the coiled chains do not overlap each other. The zero-shear-rate viscosity in this regime exhibits a linear dependence on the concentration, similar to the Einstein equation for the concentration dependence of the viscosity for a dilute solution of hard spheres. Above the critical concentration ($c > c^*$) the polymer chains form an interpenetrating network and the zero-shear-rate material functions show a strong power-law dependence on the concentration, as demonstrated earlier in Figure 4.4. For a good solvent, the critical concentration can be calculated from simple molecular arguments to be (De Gennes, 1985):
\[ c^* = \frac{3M_w}{4\pi(r^2)^{3/2}N_A} \]  

(4.24)

where \( N_A \) is the Avogadro number. From the values of the molecular parameters above, the critical concentration of PIB for entanglement is found to be \( c^* = 6.9 \times 10^{-4} \text{ g/cm} \), which is equivalent to an 0.08 wt\% solution of PIB in PB/C14. The 0.31 wt\% solution prepared for this thesis is thus not a true dilute polymer solution, but rather a semi-dilute solution in which the individual chains are moderately entangled and give rise to intermolecular interactions.

### 4.3 Rheological Models

#### 4.3.1 The Oldroyd-B Fluid Model

Boger fluids have been modeled extensively in the literature as dilute solutions of noninteracting, Hookean dumbbells in a viscous, Newtonian solvent (Prilutski et al., 1983; Binnington and Boger 1985; Magda and Larson, 1988). The resulting quasi-linear differential constitutive equation is identical to the Oldroyd-B model derived from continuum mechanics (Oldroyd, 1950) and can be written as

\[ \tau + \lambda_1 \tau_{(1)} = -\eta_0 \left[ \dot{\gamma} + \lambda_2 \dot{\gamma}_{(1)} \right] \]  

(4.25)

where \( \tau \) is the stress tensor, \( \dot{\gamma} \) is the rate of strain tensor = \( (\nabla \mathbf{v} + \nabla \mathbf{v}^* ) \), \( \lambda_1 \) is the relaxation time and \( \lambda_2 \) is a retardation time = \( \lambda_1 \eta_s / \eta_0 \). The subscript \( (1) \) indicates the convected time derivative defined by

\[ \tau_{(1)} = \frac{\partial \tau}{\partial t} + \mathbf{v} \cdot \nabla \tau - \{ \nabla \mathbf{v}^* \cdot \tau + \tau \cdot \nabla \mathbf{v} \} \]  

(4.26)

This model contains three independent parameters: the viscosities \( \eta_s \) and \( \eta_0 \), and the single relaxation time \( \lambda_1 \). In viscometric flow the Oldroyd-B model predicts a constant viscosity, a constant first normal stress coefficient and a zero second normal stress coefficient at all shear rates, and the material functions can be expressed as.
\[ \eta_0 = \eta_s + \eta_p \]
\[ \Psi_1 = 2\eta_0(\lambda_1 - \lambda_2) = 2\lambda_1(\eta_0 - \eta_s) \]
\[ \Psi_2 = 0 \]

(4.27)

where \( \eta_s \) and \( \eta_p \) are, respectively, the solvent and polymer contributions to the total viscosity. Although this model can be fitted to zero-shear viscometric data, it is unable to describe the gradual shear-thinning experimentally observed in the viscoelastic material functions. This deficiency is insignificant in describing the almost constant viscosity of Boger fluids, but is more important in accurately modeling the first normal stress coefficient \( \Psi_1 \). In small-amplitude oscillatory shear flow, the Oldroyd-B constitutive equation predicts frequency dependent linear viscoelastic functions \( \eta' \), and \( 2\eta''/\omega \) which are given by:

\[ \eta'(\omega) = \eta_s + \frac{\eta_0 - \eta_s}{1 + (\lambda_1 \omega)^2} \]

(4.28)

\[ \frac{2\eta''(\omega)}{\omega} = \frac{\eta_0(\lambda_1 - \lambda_2)}{1 + (\lambda_1 \omega)^2} \]

(4.29)

In the limits of low frequencies these functions asymptote to the zero-shear-rate values \( \eta_0 \) and \( \Psi_{10} \) in agreement with eqs. (4.8) and (4.9), before rapidly shear-thinning at high frequencies. This rapid decrease is a consequence of there only being a single relaxation time in the model, and these functions are generally incapable of describing the more gradual frequency dependence of experimental linear viscoelastic data (Ferry, 1980).

### 4.3.2 The Bird-DeAguier Model

The calculations of molecular parameters in Section 4.2.2, revealed that the concentration of PIB chains in the 0.31 wt% PIB Boger fluid exceeds the critical concentration for entanglements \( c^* = 0.08 \) wt\% and the fluid is therefore a semidilute solution. To more accurately model the shear-rate dependence of the material functions, we have used the ‘Encapsulated Dumbbell’ model of Bird and DeAguier (1983) in which the PIB molecules are modeled as a semidilute solution of finitely extensible nonlinear elastic (FENE) dumbbells. In melts and nondilute solutions the polymer chains are entangled and interact with neighboring molecules, they are thus constrained to move (or reptate) in certain preferred directions. This encapsulation is modeled by introducing anisotropy in the Stoke’s hydrodynamic drag acting on the dumbbells and using a non-isotropic velocity
distribution for the Brownian motion of the chains. An approximate closed-form constitutive equation is obtained from the configuration-space distribution function by using the Peterlin Approximation (Bird et al., 1987b) and is written as

\[
\left[ \frac{Z(\sigma \beta - T)}{(Y - T)} - \lambda_H \frac{DlnZ}{Dt} \right] \tau_p + \lambda_1 \tau_{p(1)} = -\lambda_1 Y n k_B T \gamma + \left[ \frac{(\sigma \beta - Y)Z T}{(Y - T)} - \lambda_1 Y \frac{DlnZ}{Dt} \right] n k_B T \beta 
\]

(4.30)

where \( Y = 2 \beta - 1 \) and \( Z = \frac{[1 + (3/b)(1 - T)](Y - T)}{1 - T} \).

In equation (4.27), \( \tau_p \) is the polymeric component of the stress tensor, \( n \) is the number density of dumbbells, \( k_B \) is the Boltzmann constant, and \( T \) is the non-dimensional trace of the polymeric stress:

\[
T = \frac{tr \tau_p}{3 nk_B T} 
\]

(4.31)

The Newtonian solvent's contribution to the stress is \( \tau_s = -\eta_s \gamma \), and the total stress tensor is obtained by combining the solvent and polymer contributions to the stress: \( \tau = \tau_p + \tau_s \).

In addition to the solvent viscosity \( \eta_s \), this nonlinear constitutive equation contains four parameters: \( b \) is a measure of the extensibility of the elastic spring connecting the beads; \( \lambda_1 \) is the single relaxation time-constant for the dumbbells; the parameter \( \sigma \) indicates the degree of anisotropy in the hydrodynamic drag acting on the dumbbell (a value of \( 0 < \sigma < 1 \) reflects a lower viscous drag for motion along the chain backbone); and \( \beta \) describes the deviation from the Maxwellian velocity distribution (if \( 1 < \beta \leq 3/2 \) the Brownian motion perpendicular to the dumbbell axis is decreased relative to Brownian motion along the chain axis). These parameters are inter-related and specifying any two of them, coupled with the zero-shear properties (\( \eta_0 \), \( \Psi_{10} \)) obtained from linear viscoelasticity measurements, completely defines the model (DeAguiar, 1983). We have chosen to fit the parameters \( b \) and \( \sigma \), the remaining parameters are then determined by

\[
\beta = \frac{1}{2} \left[ 1 + \frac{2(\eta_0 - \eta_s)^2}{nk_B T \Psi_{10}} \right] 
\]

(4.32)

\[
\lambda_1 = \frac{\sigma \beta [1 + (3/b)] \Psi_{10}}{2(\eta_0 - \eta_s)} 
\]
In the limit \( \beta = \sigma = 1 \) the model reduces to the dilute FENE dumbbell constitutive equation (Bird et al., 1987b); if additionally \( b \to \infty \) the Hookean dumbbell or Oldroyd-B result eq. (4.25) is recovered. The steady shear and transient material functions for the Bird-DeAguirar model are calculated by solving eq. (4.30) numerically for specific choices of the parameters \( b \) and \( \sigma \). This model contains terms that are nonlinear in stress and is able to represent a wide range of shear-rate-dependent phenomena. The behavior of the steady-shear material functions \( \eta(\dot{\gamma}) \) and \( \Psi_1(\dot{\gamma}) \) is shown in Figure 4.6, as the model parameters \( b \) and \( \sigma \) are varied. In order to highlight the shear-rate-dependence of the polymeric contribution to the viscosity, the constant solvent viscosity \( \eta_s \) has been subtracted from \( \eta(\dot{\gamma}) \). The viscometric properties have also been normalized by the zero-shear-rate values \( (\eta_0 - \eta_s) \) and \( \Psi_{10} \) respectively, and the shear rate has been nondimensionalized with the characteristic time \( (\Psi_{10}/(\eta_0 - \eta_s)) \). The parameter \( \beta \) is also strictly an independent variable in this model; however, for these calculations the value was calculated from the zero-shear-rate material functions determined in Section 4.4.2 with the use of eq. (4.29), and subsequently held constant at \( \beta = 1.13 \). The relaxation time in the model is then

\[
\lambda_1 = 0.794 \sigma \beta [1 + (3/b)]
\]

The effect of finite extensibility in the dumbbells is demonstrated by varying the parameter \( b \) in Figure 4.6(a). At low shear rates, the viscosity and first normal stress difference are constant, before showing shear-thinning behavior at higher deformation rates. The model predicts physically realistic asymptotic slopes at high shear rates of \( \eta \sim \dot{\gamma}^{-2/3} \), and \( \Psi_1 \sim \dot{\gamma}^{-4/3} \). As \( b \) increases and the dumbbells become more extensible, the shear-thinning regime shifts to higher shear rates and for infinitely extensible dumbbells \( (b \to \infty) \) the model would show no shear-thinning behavior. The ‘encapsulation’ effects of an anisotropic velocity distributions \( (\beta \neq 1) \) are also demonstrated for the choice of parameters in Figure 4.6(a), and result in a small increase in \( \eta \) and \( \Psi_1 \) (‘shear-thickening’) at intermediate shear rates, before the finite extensibility of the molecules results in shear-thinning at higher shear rates. The effect of a nonisotropic drag coefficient \( (\sigma \neq 1) \) on the material functions is demonstrated in Figure 4.6(b) for a moderately extensible dumbbell with \( b = 1000 \). Values of \( \sigma < 1 \) (i.e. lower drag along the dumbbell backbone) result in a more rapid decrease in \( \eta \) and \( \Psi_1 \) at low shear rates, followed by an intermediate region which exhibits an inflexion point in the curves before the asymptotic shear-thinning behavior develops at very high shear rates. It is interesting to note that both of the anisotropic effects modeled by \( \beta \) and \( \sigma \) are manifested in the fluid rheology at intermediate shear-rates and can result in either a local shear-thickening or an inflexion point in the material functions. Similar effects have also been computed recently by using more realistic
Figure 4.6  Steady shear material functions for the nonlinear encapsulated dumbbell model of Bird and DeAguiar (1983) demonstrating (a) the effect of finite extensibility in the dumbbells, and (b) the effect of a nonisotropic drag coefficient. (——) normalized viscosity; (-----) normalized first normal stress coefficient.
multibead-spring models with ‘consistently averaged hydrodynamic interactions’ (Magda et al., 1988; Kishbaugh and McHugh, 1990). The approximate treatment of anisotropic effects that is contained in the Bird-DeAguiar model thus appears to result in a relatively simple differential constitutive equation that can mimic the responses predicted in mathematical treatments of more rigorous kinetic theory models. It should be noted that this intermediate shear-thickening behavior cannot be modeled by other well-known nonlinear constitutive models such as the Giesekus model or Phan-Thien – Tanner model.

Despite this improved description of shear-rate-dependent material functions, the presence of only a single relaxation time in the constitutive equation eq.(4.30) limits the ability of the model to describe the linear viscoelastic material functions \( \eta' \) and \( 2\eta''/\omega \). By definition, the linear viscoelastic properties of a material reflect infinitesimal deformations and a linear response of the stress in the material to the applied strain. The nonlinear terms in equation (4.30) are thus negligible in flows such as small-amplitude oscillatory shear flow and the predictions of the Bird-DeAguiar constitutive equation are identical to those of the Oldroyd-B model given in eq. (4.28) and (4.29). To improve the modeling of these properties it is necessary to incorporate a range of relaxation times.

### 4.3.3 Multimode Models

So far, the PIB polymer chains have been modeled as extensible dumbbells in which the molecular weight of each molecule is concentrated in two point-masses connected by a massless, finitely extensible nonlinear spring. In reality, the Boger fluid consists of a semi-dilute solution of polydisperse PIB molecules in an essentially athermal solvent of shorter PB chains. The mass of the molecule is distributed evenly along the length of a flexible backbone, and there exists a wide range of characteristic length scales which can respond to deformation: from the entire contour length of the chain \( L = 8.1 \times 10^{-6} \text{ m} \) down to the individual C—C bond length \( l = 1.54 \times 10^{-10} \text{ m} \). A single relaxation time is obviously insufficient to characterize the many different modes of relaxation available to such long flexible polymer chains. The situation will not be significantly improved by the recent development of esoteric and expensive Boger fluid formulations comprised of highly monodisperse polymer components (e.g. Magda and Larson, 1988); even perfectly monodisperse polymers consist of a large number of ‘beads’ connected in a chain, and kinetic theory for such systems results in constitutive equations such as the Rouse-Zimm, Doi-Edwards and Curtiss-Bird models which predict a spectrum of relaxation times (Bird et al., 1987b).
To account for this range of time-scales we have empirically introduced multiple
time constants by fitting the linear viscoelastic relaxation modulus to the Generalized
Maxwell model

\[ G(t) = \sum_{k=1}^{n} \frac{\eta_k}{\lambda_k} e^{-t/\lambda_k} \quad (4.33) \]

where \( \lambda_k \) and \( \eta_k \) are the relaxation time and viscosity of the \( k \)th mode, and \( G_k = \eta_k/\lambda_k \) is
the shear modulus associated with each mode. A multimode constitutive equation is
constructed by linear superposition of a series of differential models each with a different
time constant and zero-shear-rate viscosity. The total stress tensor is calculated by summing
the individual modal contributions of the polymer plus the additional solvent contribution as

\[ \tau = \tau_s + \sum_{k=1}^{n} \tau_k \quad (4.34) \]

This generalization can be used with the models given by equations (4.25) and
(4.30), or with any other differential constitutive equations, for example those proposed by
Phan-Thien and Tanner, and Aciero et al. (Bird et al., 1987a). Mackay and Boger (1988)
were the first to use this approach in the modeling of Boger fluids, and they attempted to
describe a PIB/PB/K Boger fluid as a dilute solution of Hookean dumbbells dissolved in a
viscoelastic solvent that possessed a small relaxation time and a high viscosity. In the
framework of eq. (4.33) and (4.34), this is equivalent to using a two mode upper
convected Maxwell model with \( \eta_s = 0 \) and \( k = 2 \). This two mode description of the fluid
gave a better fit to the linear viscoelastic material functions over a wider range of
frequencies, however errors of up to 100% in the model predictions still existed. In
addition the superposition of two quasi-linear Maxwell modes with constant relaxation times
and viscosities results in steady-shear material functions which are also independent of
shear rate, and therefore this model cannot describe the shear-rate-dependence of \( \Psi_1 \) that is
observed for Boger fluids. A spectrum of more than two relaxation times is clearly
required, together with constitutive equations that predict shear-thinning in the steady-shear
rheological properties.
4.4 Experimental Fluid Characterization

The rheological properties of the 0.31 wt% PIB/PB/C14 Boger fluid and the 95.15 wt% PB/C14 solvent have been characterized over a wide range of test conditions using the simple shear flow experiments described in Chapter 4.1. These experiments are discussed individually below, and the predictions of the various constitutive models reviewed in Chapter 4.3 are compared to the actual data obtained with the Boger fluid. In this manner the individual strengths and weaknesses of each model are highlighted in a number of different one-dimensional flows.

4.4.1 Linear Viscoelasticity

The linear viscoelastic properties, $G'(\omega)$ and $G''(\omega)$, of the Boger fluid and the PB/C14 solvent were determined over a range of temperatures by using small-amplitude oscillatory shear flow defined by eq. (4.4) – (4.7). The measurements of the viscous modulus $G'' = \eta' \omega$ for the PB/C14 solvent are shown in Figure 4.7. As the temperature is raised from −3.4 °C to 22.6 °C the viscous modulus measured at a given frequency decreases by a factor of 10. This rheological data can be collapsed to form a single master curve at a reference condition $T_0$ through the time-temperature superposition principle (Ferry, 1980). To do this it was assumed that the material is thermo-rheologically simple (Tschoegl, 1989), so that the temperature-dependence of all the relaxation times is given by a single factor $a_T = \lambda_k(T)/\lambda_k(T_0)$, and that the form of the relaxation modulus $G_k(\omega)$ remains constant with changing temperature. In general, it is found that $G_k$ depends on the factor $(T/\rho(T_0))$; however, this dependence has been neglected in this treatment since the effect is negligible over the relatively narrow range of temperatures covered. The shift factor $a_T$ was calculated from the ratio of the zero-frequency dynamic viscosity at temperature $T$ to the value at the reference temperature $T_0$:

$$a_T = \frac{\eta'_0(T)}{\eta'_0(T_0)} = \frac{G''(T)}{G''(T_0)} \quad (4.35)$$

Accordingly, the temperature-dependent rheological parameters are then determined from the following relationships
Figure 4.7  Frequency dependence of the viscous modulus $G''(\omega)$ for the 95.15 wt% PB/C14 solution (Boger fluid solvent) over a range of temperatures.
\[ \omega(T_0) = a_\omega \omega(T), \quad \dot{\gamma}(T_0) = a_\gamma \dot{\gamma}(T) \]
\[ G'(T_0) = G'(T), \quad G''(T_0) = G''(T) \]
\[ \eta'(T_0) = \eta'(T), \quad \eta''(T_0) = \eta''(T) \]
\[ \eta(T_0) = \eta(T)/a_\eta, \quad \Psi_1'(T_0) = \Psi_1'(T)/a_\eta^2 \]  

The master curve for the viscous modulus \( G''(\omega) \) of the PIB/C14 solvent at a reference temperature of \( T_0 = 25 \text{ °C} \) is shown in Figure 4.8. By the use of time-temperature superposition it has been possible to extend the master curve beyond the maximum physically measurable frequency of \( \omega_{\text{max}} = 100 \text{ Hz} \) (a constraint of the RMS-800) by an order of magnitude to \( \omega_{\text{max}} = 1000 \text{ Hz} \). The straight line of slope unity in \( G'' \), together with the absence of any measurable elastic modulus, indicates that the PB/C14 solvent is Newtonian with a constant viscosity which is determined from the zero-frequency-asymptotic slope of Figure 4.8 to be \( \eta_0(T_0) = 8.12 \text{ Pa·s} \). Because of the presence of the polymeric polybutene component, the PB/C14 solvent must have some viscoelastic character, albeit very small. Therefore, the dynamic viscosity of the solvent (and hence the viscous modulus \( G'' = \eta'(\omega) \) should be described by a single relaxation mode of the form given by eq. (4.28), which predicts an initially constant dynamic viscosity, followed by a rapid frequency-thinning at frequencies of \( (\lambda \omega) \sim 1 \). The lines in Figure 4.8 show the results of the model predictions for \( G'' \) with relaxation times of \( \lambda_s = 10^{-3}, 5 \times 10^{-4} \) and \( 10^{-4} \) seconds. A time constant of \( \lambda_s = 10^{-4} \) provides the best fit to the data, and implies that the solvent elasticity is best described by a relaxation time of \( \lambda_s \leq 10^{-4} \) s. Mackay and Boiiger (1988) reported a relaxation time of \( \lambda_s = 2 \times 10^{-4} \) s for a Boiiger fluid solvent consisting of 92.2 wt% poly(1-butene) in kerosene, while Laun and Hingmann (1990) reported a value of \( \lambda_s = 1.3 \times 10^{-4} \) s for a solvent of 93 wt% polybutene in kerosene. Therefore, the value calculated for this 95.15 wt% PB/C14 solution appears to be a reasonable upper bound on the solvent relaxation time.

Similar curves of the linear viscoelastic quantities \( \eta'(\omega) \) and \( 2\eta''(\omega)/\omega \) for the 0.31 wt% PIB Boiiger fluid are shown in Figure 4.9. The measurements of \( \eta'(\omega) \) presented in Figure 4.9(a) demonstrates that the viscosity of the Boiiger fluid is only a weak function of the frequency \( \omega \), but that the zero-shear-rate viscosity \( \eta_0 \) decreases by a factor of 15 as the temperature is raised by 30 °C. These measurements were reduced to give the master curves of the material functions shown in Figure 4.10 by using eq. (4.33) to shift the rheological parameters to a reference temperature of \( T_0 = 25 \text{ °C} \). The master curve of the dynamic viscosity \( \eta'/a_\eta \) shows an initially constant zero-frequency asymptote for frequencies \( \omega \leq 1 \), followed by a small decrease at higher frequencies. The elastic quantity \( 2\eta'/\omega a_\eta^2 \) demonstrates a monotonic frequency-thinning across the entire frequency domain.
Figure 4.8  Master curve of the viscous modulus $G''(\omega)$ at $T_0 = 25^\circ C$ for the 95.15 wt% PB/C14 data shown in Figure 4.7. The lines are the predictions for a single viscoelastic mode given by eq.(4.28) for three different relaxation times: (---) $\lambda_x = 0.001$ s; (---) $\lambda_x = 0.0005$ s; (-----) $\lambda_x = 0.0001$ s.
Figure 4.9  Linear viscoelastic properties $\eta'(\omega)$ and $2\eta''(\omega)/\omega$ of the 0.31 wt% PIB/PB/C14 solution over a range of temperatures.
that could be attained experimentally. The dashed lines in Figure 4.10 show the results of fitting the data to the linear viscoelastic predictions of the single mode Oldroyd-B model given by eqs. (4.28) and (4.29). The solvent viscosity $\eta_s = 8.12 \text{ Pa} \cdot \text{s}$ has already been determined from the measurements of the PB/C14 solvent discussed above, and the remaining two parameters $\eta_0$ and $\lambda_1$ were fit to the data using the Levenberg-Marquardt nonlinear regression method (Press et al., 1985) to minimize the sum of the residuals $R_i$ given by

$$R_i = \left[ \log G'_i - \log G'_{\text{Old}}(\omega_i) \right]^2 + \left[ \log G''_i - \log G''_{\text{Old}}(\omega_i) \right]^2$$  \hspace{1cm} (4.37)

Here $i = 1, \ldots, N$; where $N$ is the number of experimentally determined sets of data points $\{ \omega_i, G'_i, G''_i \}$, and $G'_{\text{Old}}, G''_{\text{Old}}$ are the predictions of the Oldroyd-B model (with $\eta_s = 8.12 \text{ Pa} \cdot \text{s}$) at the frequency $\omega_i$. Each term in eq. (4.34) represents the difference between the measured parameters ($G'_i, G''_i$) and the corresponding values predicted from linear viscoelastic theory. The model parameters were determined to be $\eta_0 = 13.76 \text{ Pa} \cdot \text{s}$ and $\lambda_1 = 0.794 \text{ s}$.

The Oldroyd-B model prediction for the dynamic viscosity $\eta'$ is quite reasonable since the high solvent viscosity $\eta_s$ masks the shear-thinning behavior at high frequencies; however, comparing the data and the prediction for the elastic quantity $2\eta''/\omega$ highlights the inadequacy of the Oldroyd-B model: the predicted curve has the correct zero-frequency asymptote, but over-predicts the elasticity at intermediate shear-rates and finally decays far more rapidly than the experimentally measured dynamic rigidity. A spectrum of relaxation times is required to capture this gradual shear-thinning in the elasticity of the Boger fluid accurately.

To achieve an improved fit to the data, the Oldroyd-B model (eq. (4.25)) was generalized to $k$ modes as described in Section 4.3.3. The relaxation modulus is given by eq. (4.34), and in small-amplitude oscillatory shear flow the linear viscoelastic parameters are given by:

$$\eta'(\omega) = \eta_s + \sum_k \frac{\eta_k}{1 + (\lambda_k \omega)^2}$$  \hspace{1cm} (4.38)

$$\frac{2\eta''(\omega)}{\omega} = \sum_k \frac{2\eta_k \lambda_k}{1 + (\lambda_k \omega)^2}$$  \hspace{1cm} (4.39)
Figure 4.10  Master curves of the dynamic properties $\eta'(\omega)$ and $2\eta''(\omega)/\omega$ at $T_0 = 25^\circ$C for the 0.31 wt% PIB/PB/C14 Boger fluid. The dashed lines (---) are the predictions of the 1-mode Oldroyd-B model with $\eta_0 = 13.76$ Pa·s, $\eta_s = 8.12$ Pa·s, $\lambda_1 = 0.794$ s.
The set of values \( \{ \eta_k, \lambda_k \} \) were determined by employing the Levenberg-Marquardt method again, with a modified set of residual equations in which the terms \( G'_{Old}, G''_{Old} \) are replaced by the predictions of the multimode linear viscoelastic theory. The number of modes \( k \) was chosen to be the minimum number that gave a smooth fit to the data and this was found to be \( k = 4 \) for the range of data shown in Figure 4.10. The introduction of additional modes did not improve the fit and was found to result in two modes \( k = 4, 5 \) with almost identical relaxation times. The final values of the four mode spectra \( \{ \eta_k, \lambda_k \} \) at a reference temperature of \( T_0 = 25 \, ^\circ\text{C} \) are listed in Table 4.2:

<table>
<thead>
<tr>
<th>Mode No.</th>
<th>( \eta_k ) [Pa·s]</th>
<th>( \lambda_k ) [s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.108</td>
<td>2.755</td>
</tr>
<tr>
<td>2</td>
<td>1.677</td>
<td>0.7361</td>
</tr>
<tr>
<td>3</td>
<td>1.657</td>
<td>0.1094</td>
</tr>
<tr>
<td>4</td>
<td>1.211</td>
<td>0.0098</td>
</tr>
<tr>
<td>Solvent</td>
<td>8.118</td>
<td>—</td>
</tr>
</tbody>
</table>

Table 4.2 Linear viscoelastic spectrum for the 0.31 wt% PIB/PB/C14 Boger fluid at \( T_0 = 25 \, ^\circ\text{C} \).

A comparison of the 4-mode model predictions with the experimental data is shown by the solid line in Figure 4.11. The fit is quantitative across the entire range of data and accurately describes the frequency dependence of both \( \eta' \) and \( 2\eta''/\omega \). The zero-frequency asymptotes of the dynamic material functions are given by \( \eta_0 = \eta_s + \Sigma \eta_k = 13.76 \, \text{Pa·s} \), and \( \Psi_{10} = \Sigma 2\eta_k\lambda_k \). The individual spectral contributions of the four modes to the total value of \( 2\eta''/\omega \) given by eq. (4.39) are also shown in Figure 4.11 by the dashed lines. The zero-frequency value of the \( k^{\text{th}} \) mode is \( 2\eta_k\lambda_k \) and the mode begins to frequency-thin at a frequency of \( \omega \sim 1/\lambda_k \). Hence, each discrete spectral mode is dominant \( (i.e.\,\) has the largest numerical value) in different parts of the frequency domain. Similar contributions make up the total dynamic viscosity, however in this case the spectral decomposition is not as clear due to the presence of the large solvent viscosity \( \eta_s \).

The master curve, Figure 4.10, coupled with the linear viscoelastic spectrum given in Table 4.2, allows us to calculate the material functions at any chosen \( \omega \). If, in addition, the shift factor \( \alpha_T \) is known the material functions at any chosen \( T \) can also be calculated.
Figure 4.11  The discrete linear viscoelastic spectrum for the 0.31 wt% PIB/PB/C14 Boger fluid. Hollow symbols show experimentally determined master curves of the linear viscoelastic properties: (○) dynamic viscosity \( \eta' \) and (△) \( 2\eta''/\omega \) [Pa·s²]. (—) Predictions of eq. (4.35) and (4.36) obtained for the discrete four mode relaxation spectrum specified in Table 4.2; (—-—) Individual spectral contributions \( (2\eta''/\omega)_k \) of the four modes.
The shift factors for both the PB/C14 and the PIB/PB/C14 fluid were found to depend on temperature according to an Arrhenius expression

\[ \alpha_T = \exp \left( \frac{\Delta H}{R} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right) \]  

(4.40)

where \( R = 8.314 \) J/mol-K and \( \Delta H \) is the 'activation energy for flow' (Ferry, 1980). The shift factors for each fluid are plotted in Figure 4.12 as functions of the temperature function \( (T^{-1} - T_0^{-1}) \) with a reference condition of \( T_0 = 298 \) K. The slope of this line determines the value \( \Delta H/R \) and the final values for each fluid are given in Table 4.3:

<table>
<thead>
<tr>
<th>Fluid</th>
<th>( \Delta H/R ) [K]</th>
<th>( \eta_0 ) [Pa·s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.31 wt% PIB/PB/C14</td>
<td>7362</td>
<td>13.77</td>
</tr>
<tr>
<td>95.15 wt% PB/C14</td>
<td>7432</td>
<td>8.118</td>
</tr>
</tbody>
</table>

Table 4.3 Flow activation energy and zero-shear-rate viscosity at \( T_0 = 25 \) °C obtained from experimental measurements of \( \eta'(\omega, T) \).

The flow activation energies for both fluids are almost identical as would be expected since both fluids consist predominantly of the PB component. The values of \( \Delta H/R \) are also extremely high and indicate the large temperature sensitivity of the polybutene material; a change of 1 °C in the temperature range \( (8 \leq T \leq 40 \) °C) results in an average change of 8.5% in the viscosity. Measurements of the activation energy for fluids containing only PIB and C14 components show a much lower temperature sensitivity and a flow activation energy of \( \Delta H/R = 2593 \) K (Quinzani et al., 1990). Careful monitoring of the ambient temperature will thus be extremely important in experiments with the PIB/PB/C14 Boger fluid.

4.4.2 Steady Shear Flow

Having accurately determined the linear viscoelastic spectrum of the Boger fluid, attention is now focused on the material functions \( \eta \) and \( \Psi_1 \) observed in steady simple shear flow. Measurements of the viscosity and first normal stress difference were made over a range of temperatures, and master curves at \( T_0 = 25 \) °C were then prepared by using eq. (4.33). The shift factors used in reducing the viscometric functions were calculated
Figure 4.12  Temperature dependence of the shift factor $a_T$ for the 95.15 wt% PB/C14 solvent (△) and the 0.31 wt% PIB/PB/C14 Boger fluid (○).
using eq. (4.40) with the flow activation energy obtained from linear viscoelasticity. Figure 4.13 shows the master curves for \( \eta \) and \( \Psi_1 \) as functions of the reduced shear rate \( \gamma \tau \). The viscosity is approximately constant across four decades of shear rate, however the behavior of the first normal stress coefficient is more complicated; even at very low shear rates of \( \dot{\gamma} = 0.1 \text{ s}^{-1} \) the fluid exhibits some shear-thinning, then at intermediate shear rates \( 3 < \dot{\gamma} < 30 \text{ s}^{-1} \) the first normal stress coefficient exhibits an almost constant plateau before resuming a monotonic decrease at high shear rates. Also shown in Figure 4.13 are the steady shear material functions predicted by the Oldroyd-B model (broken lines) and the one-mode Bird-DeAguiar model (solid lines). For the Oldroyd-B model these predictions are simply flat lines with constant values given by eq. (4.27), where the parameters \( \eta_0 = 13.76 \text{ Pa} \cdot \text{s}, \eta_s = 8.12 \text{ Pa} \cdot \text{s} \) and \( \lambda_1 = 0.794 \text{ s} \) are those determined from linear viscoelasticity. It can be seen from Figure 4.13 that the Oldroyd-B model prediction for the normal stress coefficient \( \Psi_1 \) is extremely poor, even at moderate shear rates. The complex ‘plateau’ region in \( \Psi_1 \) has lead to significant confusion in the literature since it has often been misinterpreted in steady shear measurements as the zero-shear-rate asymptote of the first normal stress coefficient. If \( \Psi_{10} \) is determined experimentally from this region it will be considerably underestimated and the zero-shear-rate asymptotes of the steady shear material function \( \Psi_1 \) and the dynamic quantity \( 2\eta''/\omega \) will not be the same (see for example Binnington and Boger, 1986; Tam et al., 1989). Quinzani et al. (1990) replotted the rheological data of several researchers and demonstrated that this ‘plateau’ in \( \Psi_1 \) also exists for a number of other PIB- and PAC-based Boger fluids. Thus, it is important with such nonlinear fluid rheology to use linear viscoelastic measurements to determine the zero-shear-rate material functions, \( \eta_0 \) and \( \Psi_{10} \), of the fluid.

For the Bird-DeAguiar model, in addition to these zero-shear-rate parameters we require appropriate choices for \( \beta, \sigma \) and \( b \). For a PIB concentration of 0.31 wt% \( (c = 2.73 \times 10^{-4} \text{ g/cm}^3) \) the number density is \( n = 1.37 \times 10^{21} \text{ dumbbells/m}^3 \), and therefore the quantity \( nk_B T = 5.633 \text{ J/m}^3 \) at \( T = 25 \text{ °C} \). The anisotropic velocity coefficient is thus determined from eq. (4.32) to be \( \beta = 1.13 \). Although the parameter \( b \) is regarded as an adjustable fitting parameter, some simple molecular arguments can suggest the order of magnitude that should be expected. It can be shown (Bird et al., 1987b; p76) for an ideal polymer molecule \( (i.e. \text{a Kramers chain} \) consisting of \( N^* \) freely jointed rods of length \( a \), that the chain is equivalent to an extensible dumbbell with a spring constant \( H \equiv 3k_B T/(N^* - 1)a^2 \) and a finite extensibility characterized by \( b = HL^2/k_B T \), where \( L \) is the contour length of the chain. As discussed in Section 4.2.2, an actual macromolecule such as PIB with \( N \) aliphatic carbons of bond length \( l \) is not an ideal bead-rod chain due to fixed bond angles, steric hindrances and excluded volume effects that lead to a
Figure 4.13. Master curves of the viscosity $\eta$ and first normal stress coefficient $\Psi_1$ measured in steady shear flow for the 0.31 wt% PIB/PB/C14 Boger fluid. The zero-shear-rate parameters are predetermined from linear viscoelasticity. The lines show the single mode predictions of the Oldroyd-B model (---), and the Bird-DeAguiar model with $b = 12500$, $\sigma = 0.65$ (-----).
characteristic ratio $C > 1$. However, the parameters $N^*$ and $a$ can still be calculated for such molecules by considering a 'statistically equivalent' ideal chain (Flory, 1953) with the same mean end-to-end length $\langle r^2 \rangle_0$ and the same contour length $L$. With the use of equations (4.21) and (4.22), the parameters $N^*$ and $a$ are defined by

$$C(N - 1)l^2 = \langle r^2 \rangle_0 = (N^* - 1)a^2$$

$$\frac{N - 1}{\cos \frac{1}{2}(180 - \theta)} = L = (N^* - 1)a$$

From the values of $\langle r^2 \rangle_0$ and $L$ determined in Section 4.2.2 for the PIB molecules, these two equations may be solved to give $N^* = N/10 = 4600$, and $a = 8.2l = 1.26 \times 10^{-9}$ m; i.e. the statistically equivalent chain consists of $N/10$ freely jointed rods of length 8.2 times that of a single C–C bond. Combining the two expressions for $H$ and $b$ with eq. (4.38) gives the result $b = 3(N^* - 1)$ and thus for this polyisobutylene (of molecular weight $M_w = 1.8 \times 10^6$ g/mol), $b = 13800$. This is considerably larger than the values of $50 < b < 1000$ typically determined for dilute or semidilute shear-thinning polymer solutions (Christiansen and Bird, 1977/78) and arises from the combination of a high molecular weight polymer (PIB) dissolved in an athermal solvent (PB). It is stressed that this value of $b$ is intended only to be a qualitative estimate of the chain extensibility and that the actual value should be determined, together with the parameter $\sigma$, from fitting the model predictions to the actual fluid rheology.

In Figure 4.13 the final model predictions for the steady shear material functions are shown for values of $b = 12000$, and $\sigma = 0.65$. Although the fit is not quantitative, the Bird-DeAguiar model is able to describe the complicated shear-thinning behavior of the Boger fluid elasticity, including the plateau at intermediate shear rates and the rapid decay in $\Psi_1$ at high shear rates. By varying the parameter $b$ or $\sigma$, it is possible to fit quantitatively either the low shear-rate behavior of $\Psi_1$ or the shear-thinning behavior observed at high shear rates. However, with a single mode model it is not possible to fit the data over the entire range of shear rates. A series of measurements in other PIB/PB/C14 fluids also indicate that this intermediate plateau behavior persists even in extremely dilute Boger fluids (0.01 wt% PIB) well before the critical concentration $c^*$ (Quinzani et al., 1990). In the model this plateau arises from an anisotropic hydrodynamic drag coefficient, and its manifestation in the fluid rheology may be interpreted analogously as a result of hydrodynamic interactions between the PIB polymer chains and the viscous PB solvent.

Despite this improved modeling of the viscometric properties, the single mode Bird-DeAguiar model reduces to the Oldroyd-B constitutive equation in small-amplitude
oscillatory flow and thus cannot improve the single-mode description of the linear viscoelastic properties of Boger fluids previously shown in Figure 4.10. The introduction of a discrete multimode relaxation spectrum was shown to result in a quantitative description of both \( \eta' \) and \( 2\eta'/\omega \) for the Boger fluid and this approach is now extended to the steady shear properties \( \eta \) and \( \Psi_1 \). The multimode constitutive equations are constructed by using the set of \( \{ \eta_k, \lambda_k \} \) that have already been determined from the linear viscoelastic data in Section 4.4.1 and then determining any remaining model parameters by fitting the steady shear material functions. Although a multimode form of the Oldroyd-B model was used with great success in Section 4.4.1, it cannot improve the description of \( \eta \) or \( \Psi_1 \). Since the quasilinear constitutive equation for each independent mode results in a constant viscosity \( \eta_k \) and constant first normal stress coefficient \( 2\eta_k\lambda_k \), the linear combination of modes defined by eq. (4.31) results in constant total viscometric properties of \( \eta_0 = \eta_s + \sum \eta_k \) and \( \Psi_{10} = \sum 2\eta_k\lambda_k \). The multimode predictions will thus be identical to those of the single mode Oldroyd-B model shown in Figure 4.13.

A multimode formulation of the nonlinear Bird-DeAguiar model has also been used to model the steady shear rheology, using the set of parameter values \( \{ \eta_k, \lambda_k, b_k, \sigma_k \} \) shown in Table 4.4:

<table>
<thead>
<tr>
<th>Mode No.</th>
<th>( \eta_k ) [Pa·s]</th>
<th>( \lambda_k ) [s]</th>
<th>( b_k )</th>
<th>( \sigma_k )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.108</td>
<td>2.755</td>
<td>10</td>
<td>0.2</td>
</tr>
<tr>
<td>2</td>
<td>1.677</td>
<td>0.7361</td>
<td>12000</td>
<td>1.0</td>
</tr>
<tr>
<td>3</td>
<td>1.657</td>
<td>0.1094</td>
<td>100</td>
<td>0.8</td>
</tr>
<tr>
<td>4</td>
<td>1.211</td>
<td>0.0098</td>
<td>100</td>
<td>0.9</td>
</tr>
<tr>
<td>Solvent</td>
<td>8.118</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>

**Table 4.4** Model parameters used in the 4-mode Bird-DeAguiar formulation constitutive equation

The results of fitting the fluid rheology with the four mode Bird-DeAguiar model are shown in Figure 4.14. The master curves of the experimental data in steady shear flow (solid symbols) and small-amplitude oscillatory shear flow (hollow symbols) have been superimposed and the 4-mode model predictions in each type of shear flow are shown by solid lines and dashed lines respectively. It can be seen from Figure 4.14 that the complex
Figure 4.14 Viscous and elastic material functions for the 0.31 wt% PIB/PB/C14 Boger fluid at 25°C. Solid symbols are steady shear flow properties: (○) viscosity \( \eta \) [Pa·s] and (▲) first normal stress coefficient \( \Psi_1 \) [Pa·s²]. Hollow symbols denote linear viscoelastic properties: (○) dynamic viscosity \( \eta' \) and (▲) \( 2\eta''/\omega \) [Pa·s²]. Also shown are the model predictions of the nonlinear 4-mode Bird-DeAguiar model for the dynamic (---) and viscometric (——) data.
rheology of the Boger fluid can be described quantitatively by such a multimode nonlinear model. The data in Figure 4.14 demonstrate that in dynamic experiments the limiting value of the elastic quantity $2\eta''/\omega$ at low frequencies is equal to $\Psi_{10}$, in accordance with simple fluid theory (eqs. 4.8 and 4.9), but that $2\eta''/\omega$ decreases far more rapidly than $\Psi_1$ at high frequencies. Similarly, the dynamic viscosity $\eta'$ decreases slightly more rapidly than the steady shear viscosity $\eta$ from the zero-shear-rate value $\eta_0$ towards the ultimate solvent value $\eta_s$. The multimode model also describes accurately the intermediate plateau region observed in $\Psi_1$. The first mode ($\lambda_1$) describes the initial shear-thinning at low shear rates ($\gamma < 1 \text{ s}^{-1}$) and the second mode ($\lambda_2$) describes the intermediate plateau region. Analysis of the individual modal contributions to $\Psi_1$ show that the large value of the parameter $b$ in the second mode coupled with a value of $\beta = 1.13$ results in a mode that shows a small degree of shear-thickening at intermediate shear rates, similar to that demonstrated in Figure 4.6(a). The model is thus capable of describing the local maximum in $\Psi_1$ that can be observed in the plateau region.

Very recently it has been possible to extend the measurement of the Boger fluid viscosity to much higher shear rates by the use of capillary rheometry. The Boger fluid is placed in an upstream reservoir and forced through a narrow orifice of diameter $D = 0.2 \text{ mm}$. By measuring the pressure drop across the orifice and the flow rate it is possible to calculate the fluid rheology by use of the Weissenberg-Rabinowitsch equation (Bird et al., 1987a). The results obtained in an Instron capillary rheometer over a range of shear rates $250 \leq \gamma \leq 20000 \text{ s}^{-1}$ are shown in Figure 4.15 by the solid circles, together with the master curve for the viscosity determined experimentally in the mechanical spectrometer (hollow circles). The agreement between the two data sets is extremely good; the small discrepancy between the data at $\gamma = 200 \text{ s}^{-1}$ is only $\pm 1 \text{ Pa} \cdot \text{s}$ (note the expanded scale of the ordinate axis). This small error is probably a consequence of end effects in the capillary which could not be corrected for due to the unavailability of capillaries with a range of die-lengths (Byars, 1991). The data presented in Figure 4.15 shows that the viscosity of the Boger fluid does not approach a constant Newtonian value $\eta_s$ at high shear rates, but instead shows pronounced shear-thinning at shear rates of $\gamma \sim 10000 \text{ s}^{-1}$. This is the first time that this effect has been demonstrated in a Boger fluid (similar results have recently been presented by Binding et al., 1990). This shear-thinning can readily be accounted for in the multimode formulation by modeling the PB/C14 not as a Newtonian solvent but by an additional polymeric mode. The solid line in Figure 4.15 shows the results calculated for a 5-mode model obtained using the same 4-mode parameter set listed in Table 4.4 and replacing the solvent by a fifth mode with $\eta_5 = \eta_s = 8.118 \text{ Pa} \cdot \text{s}$, $\lambda_5 = \lambda_s = 10^{-4} \text{ s}$ and $b_5 = 10$, $\sigma_5 = 0.2$. Although the fit is not quantitative, it demonstrates that the high shear-
Figure 4.15  Extension of the steady-shear viscosity master curve to high shear-rates; (○) low shear rate data obtained in the RMS-800 mechanical spectrometer; (●) high shear-rate data from the Instron capillary rheometer. The solid line shows the 5-mode model fit obtained by using a finite solvent relaxation time of $10^{-4}$ s.
rate rheology of the PIB/PB/C14 fluid can also be described by a multimode model. A quantitative fit to the data in Figure 4.15 could be achieved by readjusting the model parameters of the 4th and 5th modes; however, considering the experimental uncertainties associated with this preliminary capillary data this was not considered to be justifiable.

Combining these steady shear observations with the dynamic measurements obtained with the PB/C14 solution shows that the solvent is both highly viscous and weakly elastic with a relaxation time of \( \lambda_s \approx 10^{-4} \) s. However, its contribution of \( \Psi_{10(s)} = 2\eta_s\lambda_s \approx 8.1 \times 10^{-4} \text{ Pa}\cdot\text{s}^2 \) to the zero-shear-rate first normal stress coefficient is at least ten times smaller than any of the individual modal contributions in Table 4.4, and four orders of magnitude smaller than the total value \( \Psi_{10} = 8.96 \text{ Pa}\cdot\text{s}^2 \). For the range of shear rates \( 0.1 \leq \dot{\gamma} \leq 200 \text{ s}^{-1} \) that are attained in the flow experiments described in Chapters 6 and 7 it may therefore considered to be a purely Newtonian solvent with negligible elasticity.

4.4.3 Transient Shear Flows

Although the steady shear properties of Boger fluids have been studied intensively by a number of researchers in the past, the transient shear flows have largely been ignored. Larson and coworkers (Magda and Larson, 1988; Larson et al., 1990) have briefly considered stress relaxation following the cessation of steady shear (Figure 4.1(d)) and showed that the mean relaxation time determined from such experiments is larger than that obtained from steady shear flow experiments; however, in previous work few results are presented and the variation with applied shear rate \( \dot{\gamma}_0 \) is not even considered.

This variation of the material functions with applied shear is important as demonstrated by Figure 4.16. The stress relaxation material function \( \eta^-(t;\dot{\gamma}_0) \) is calculated by measuring the decay of the shear stress following the cessation of steady shear flow at time \( t = 0 \) (eq. 4.10) and results are shown for a number of different applied shear rates \( \dot{\gamma}_0 \). As the applied shear rate is increased it is found that the viscosity function \( \eta^-(t;\dot{\gamma}_0) \) decays to zero more rapidly. Also shown in Figure 4.16 are the predictions of the single mode Oldroyd-B (dashed line) and Bird-DeAguiar constitutive models. Stress relaxation for the Oldroyd-B model (eq. 4.22) may be solved analytically and the shear stress relaxation material function is found to be

\[
\eta^-(t;\dot{\gamma}_0) = (\eta_0 - \eta_s) e^{-t/\lambda_t}
\]  
(4.42)
Figure 4.16 Comparison of the stress relaxation function $\eta^-(t,\gamma_0)$ at 25 °C for the 0.31 wt% PIB/PB/C14 Boger fluid, plus the predictions of the single mode Bird-DeAguiar model with $b = 12000$, $\beta = 1.13$, $\sigma = 0.65$. The dashed line (------) also indicates the shear-rate-independent prediction of the Oldroyd-B model.
The model predicts an initial step decrease in the viscosity from $\eta_0$ to $(\eta_0 - \eta_s)$ at $t = 0$, due to the presence of the Newtonian solvent, in reasonable agreement with the rapid initial drop in $\eta^-$ shown in Figure 4.16, but then predicts that the polymeric component decays exponentially. On a semi-log plot equation (4.42) thus predicts a straight line with a slope of $-1/\lambda_1$ that is independent of shear rate; as shown by the dashed line in Figure 4.16. To model the experimentally observed dependence of $\eta^-$ on the applied shear rate, nonlinear terms must be added to the quasi-linear constitutive equation. The material function $\eta^-(t, \gamma_0)$ for the single mode Bird-DeAguiar model was solved numerically by integrating the set of coupled differential constitutive equations for stress over time with a second-order Adams-Bashforth integration routine (Dahlquist and Björck, 1974). The results are shown by the solid lines in Figure 4.16. and it can be seen that, in addition to the step jump at $t = 0$, the model predicts a faster stress relaxation as the applied shear rate is increased. These results illustrate a qualitative agreement with the data; however, once again the presence of only a single relaxation time in the constitutive equation (4.30) results in a simple exponential decay of the shear stress, which gives lines of constant slope on a semi-log plot. Single mode models are thus not able to describe quantitatively the transient shear properties of Boger fluids.

The predictions of the multimode models for the stress relaxation viscosity function are shown in Figure 4.17 for the set of parameters specified in Table 4.4. For the 4-mode Oldroyd-B model, the viscosity $\eta^-$ is given by a sum of exponential modes of the form $\eta_4 \exp(-t/\lambda_4)$ and is shown by the dotted line in Figure 4.17. This 4-mode spectrum describes the stress relaxation observed in the Boger fluid at the lowest shear rates ($\gamma_0 \leq 0.1 \text{ s}^{-1}$), i.e. those that lie within the ‘linear viscoelastic envelope’, but again this quasilinear model fails to predict the dependence on the applied shear rate that is observed for $\gamma \geq 1 \text{ s}^{-1}$. The multimode Bird-DeAguiar model does capture this dependence and describes quantitatively the stress relaxation at low and moderate shear rates. At the highest shear rates of 10 and 40 s$^{-1}$ the experimental data decay more rapidly than the model predictions at small times and more slowly at long times. These results suggest that even a four-mode spectrum fails to capture the full range of time-scales present in the Boger fluid.

Very similar results are obtained for the transient viscous and first normal stress coefficients ($\eta^+, \Psi_1^+$) measured in the start-up of steady shear flow (Figure 4.1(c)). Experimental measurements of $\Psi_1^+$ for the PIB/PB/C14 Boger fluid are shown in Figure 4.18, together with the predictions of the 4-mode Oldroyd-B and Bird-DeAguiar models. For clarity the data have been separated into two parts. The 4-mode Oldroyd-B model again shows no shear-rate-dependence but forms an ‘envelope’ which bounds the data from above. Further evidence for the plateau region and inflection point in the first
Figure 4.17  Stress relaxation function $\eta(t, \gamma_0)$ at $25^\circ$C for the 0.31 wt% PIB/PB/C14 Boger fluid, plus the predictions of the four mode constitutive equations; (-----) linear viscoelastic envelope; (---) four mode Bird-DeAguiar model with parameters given in Table 4.4.
Figure 4.18 Time-dependent material function $\Psi_1^+(t,\dot{\gamma}_0)$ measured in the start-up of steady shearing flow for the 0.31 wt% PIB/PB/C14 Boger fluid. The lines are the predictions of the 4-mode Oldroyd-B model (-----), and the 4-mode Bird-DeAguiar model (----) for the parameter values given in Table 4.2 and 4.4.
normal stress coefficient observed in Figures 4.13 and 4.14 can also be discerned from the results in Figure 4.18. The curves increase monotonically and have the same asymptote at long times in the range of shear rates corresponding to the plateau ($\gamma_0 = 6.0$ and $10.0$ s$^{-1}$). However, at a shear rate of $\dot{\gamma}_0 = 20$ s$^{-1}$ an overshoot in $\Psi_1^+$ is observed and the asymptotic value of $\Psi_1$ obtained as $t \to \infty$ is larger than either of the values observed at $\dot{\gamma}_0 = 10$ s$^{-1}$ or 40 s$^{-1}$. The four-mode Bird-DeAguiar model is able to predict this plateau region at intermediate shear-rates and the appearance of a normal stress overshoot at high shear rates; however, the magnitude of this overshoot is much smaller than that observed in the experimental measurements.

The final transient shear flow that has been considered is the step-strain experiment (Figure 4.1(e)). The measurements of the relaxation modulus $G(t, \gamma_0)$ obtained for the Boger fluid at $T = 25^\circ$C are shown in Figure 4.19. At small strains ($\gamma_0 \leq 1$) the relaxation modulus is independent of the applied strain and is well described by the predictions of the 4-mode linear viscoelastic model. Even at higher strains, the data in Figure 4.19 show that the measured relaxation modulus is only weakly dependent on the applied strain $\gamma_0$, and the same set of relaxation times dominate the flow at all conditions. This is a consequence of the large viscous shear modulus associated with the PB/C14 solvent (see Figure 4.8). The experimental data is well described by the multimode Bird-DeAguiar model, and close inspection of Figure 4.19 shows that the nonlinear model even captures the small strain-dependence of $G(t, \gamma_0)$ at high strains. The prediction of the single mode Oldroyd-B model for the relaxation modulus is also shown in Figure 4.19 by the thick black line. Once again, the presence of a single relaxation time results in a simple exponential decay of $G(t)$ with no dependence on applied strain. The model is thus incapable of describing the shear modulus of the Boger fluid.

4.4.4 Extensional Flows

Although no experimental measurements have been possible for the steady shearfree material functions of the Boger fluid used in this study, it is interesting to examine the predictions of the constitutive models based on the numerical values of the model parameters determined by fitting the linear viscoelastic and steady shear data.

For a single-mode Oldroyd-B model the elongational viscosity in shearfree flows is

$$\bar{\eta}_1 = (3 + \kappa) \eta_k + \frac{(3 + \kappa) (\eta_0 - \eta_k)}{[1 + (1 + \kappa) \lambda \varepsilon] (1 - 2\lambda \varepsilon)}$$ (4.43)
Figure 4.19  Relaxation modulus $G(t,\gamma_0)$ at 25 °C for the 0.31 wt% PIB/PB/C14 Boger fluid. (--•--) Linear viscoelastic envelope; (—•—•) 4-mode Bird-DeAguiar model. The thick line is the modulus $G(t)$ predicted by the single-mode Oldroyd-B model with $\lambda_1 = 0.794$ s and $\eta_s = 8.12$ Pa·s.
At low extension rates \( \dot{\varepsilon} \ll 1 \), the Trouton ratio is \( (\bar{\eta}_l/\eta_0) = 3 \) for uniaxial or biaxial flow \( (\kappa = 0) \), or 4 in planar extensional flow \( (\kappa = 1) \); in agreement with results for a Newtonian fluid (Bird et al., 1987a). However, the infinite extensibility of the Hookean dumbbells results in a physical behavior at higher deformation rates, and the model predicts an infinite extensional viscosity at finite extension rates of \( \dot{\lambda} \dot{\varepsilon} = 1/2 \) and \( \dot{\lambda} \dot{\varepsilon} = -1/(1+\kappa) \).

By incorporating finite extensibility in the dumbbells it is possible to remove this singularity in the extensional viscosity and the single-mode Bird-DeAguiar model predicts bounded extensional viscosities given by

\[
\begin{align*}
\bar{\eta}_l &\rightarrow (3 + \kappa)\eta_0, \quad \dot{\varepsilon} \rightarrow 0 \\
\bar{\eta}_l &\rightarrow 3\eta_s + 2(\eta_0 - \eta_s)\sigma\beta(3 + b)/(2\beta - 1), \quad \kappa = 0, \dot{\varepsilon} \rightarrow +\infty \\
\bar{\eta}_l &\rightarrow 3\eta_s + (\eta_0 - \eta_s)\sigma\beta(3 + b)/(2\beta - 1), \quad \kappa = 0, \dot{\varepsilon} \rightarrow -\infty \\
\bar{\eta}_l &\rightarrow 4\eta_s + 2(\eta_0 - \eta_s)\sigma\beta(3 + b)/(2\beta - 1), \quad \kappa = 1, \dot{\varepsilon} \rightarrow \pm\infty
\end{align*}
\]

(4.44)

The model is capable of displaying either an extensional thickening or thinning behavior, depending on the values of \( \sigma, \beta \) and \( b \). However, the model behavior is monotonic (either increasing or decreasing) and it cannot predict the initial increase in \( \bar{\eta} \) followed by subsequent decline at high \( \dot{\varepsilon} \) that has been observed experimentally in polyethylene melts and that can be modeled by more complex constitutive equations (Wiest, 1989). For the parameter values used in Figure 4.13, the one-mode Bird-DeAguiar model predicts an asymptotic Trouton ratio of \( (\bar{\eta}_l/\eta_0) = 6000 \) as \( \dot{\varepsilon} \rightarrow \infty \).

In multi-mode formulations of the above constitutive equation, the asymptotic values will simply be summations of the model predictions for each individual mode. The model predictions for the the 4-mode Oldroyd-B model and 4-mode Bird-DeAguiar model are shown in Figure 4.20 for (a) uniaxial elongation \( (\kappa = 0) \) and (b) planar extensional flow \( (\kappa = 1) \). At low extension rates, both models predict Newtonian behavior with Trouton ratios given by \( (\bar{\eta}_l/\eta_0) = 3 + \kappa \). At moderate elongation rates, the Oldroyd-B model shows a rapid increase in extensional viscosity and has an infinite value at an extension rate which can be calculated from the longest relaxation time in Table 4.4 to be \( \dot{\varepsilon} = 1/2\dot{\lambda}_1 = 0.181 \) s\(^{-1}\) for both uniaxial and planar extensional flow.

The 4-mode Bird-DeAguiar model predicts a smooth increase in \( \bar{\eta}_l \) and an ultimate Trouton ratio of \( (\bar{\eta}_l/\eta_0) = 3000 \) in both cases. The model thus predicts a finite degree of extensional thickening in the elongational flows of Boger fluids. Although no measurements for shearfree flow of this Boger fluid have been possible, this prediction is
Figure 4.20  Trouton ratios ($\tilde{\eta}/\eta_0$) for the Boger fluid predicted with the parameters from Table 4.4 in (a) uniaxial elongation ($\kappa = 0$), and (b) planar extension ($\kappa = 1$). (——) 4-mode Bird-DeAguiar model; (-----) 4-mode Oldroyd-B model.
consistent with experimental trends observed in other Boger fluid systems. These measurements result in Trouton ratios in the range $70 \leq \bar{\eta}_1/\eta_0 \leq 10^4$ for both planar extensional flow (Williams and Williams, 1985; Jones et al., 1987) and uniaxial extensional flow (Jackson et al., 1984; Walters, 1985). More recent concerted efforts to measure the extensional viscosity of a PIB/PB/K Boger fluid confirm this extensional-thickening trend; however, values of Trouton ratios in the range $10 \leq \bar{\eta}_1/\eta_0 \leq 10^7$ were reported for deformation rates in the limited range $0.7 \leq \dot{\varepsilon} \leq 400$ s$^{-1}$ (see Sridhar, 1990). This extreme scatter in the experimental data preclude a quantitative description of the shearfree material functions, but we may conclude that, in general, Boger fluids exhibit pronounced extensional thickening in elongational flows. One of the outstanding challenges in rheometry remains the development of accurate and consistent methods for determining the elongational properties of viscoelastic solutions.

4.4.5 Summary of the Modeling of Boger Fluids

The large number of experiments discussed above have shown that the simple Oldroyd-B constitutive equation is inadequate for modeling the material functions of Boger fluids in linear viscoelastic, steady-shear and transient shear flows, except in the limit of very low deformation rates. By more realistic modeling of both the polymer chains (incorporating finite extensibility) and the local molecular environment (‘encapsulation’) we have captured more features of the Boger fluid rheology than was possible with the Oldroyd-B model and have also eliminated aphysical behavior; for example, in elongational flow the Bird-DeAguiar model predicts a large, but finite, increase in extensional viscosity. However, the single relaxation time present in this model still prevents anything more than qualitative description of transient flow phenomena, such as stress relaxation.

Although the 4-mode model offers the advantage of a quantitative description of the Boger fluid rheology, it is analytically intractable and hitherto multimode models have proven prohibitively expensive to use in numerical simulations. The one-mode Bird-DeAguiar captures most of the salient features of the fluid rheology and can be used to predict the qualitative flow behavior of the Boger fluid in complex geometries. However, recent finite element formulations developed within the group (Rajagopal, 1990; Northey, 1990) offer the possibility of relatively inexpensive multimode calculations (in which addition of each additional mode increases the number of unknowns in the numerical formulation by approximately 15%), and quantitative comparison between accurate experimental measurements and numerical simulations now appears possible in complex viscoelastic flows.
4.5 Dimensionless Flow Parameters for the PIB Boger Fluid

The Reynolds number and the Deborah number are used as scaling parameters to describe the magnitude of inertial and elastic effects in the flow. From the general definition of the Deborah number given in equation (1.1) it can be seen that expressions are required for the fluid relaxation time and the time scale of the flow. A characteristic shear rate in a complex flow is defined by $\dot{\gamma} = \langle v \rangle / L$, where $\langle v \rangle$ is the average velocity in the flow geometry and $L$ is a characteristic length scale: For the axisymmetric contraction discussed in Chapter 6, we have chosen $L = R_2$, the downstream tube radius, and for the constrained cylinder geometry in Chapter 7 we chose $L = R_c$, the cylinder radius. A representative time for the flow is then to be $\mathcal{I} = L / \langle v \rangle$. For these geometries, the definition of the Deborah number is thus equivalent to a Weissenberg number based on a characteristic strain-rate $\kappa$ in the flow. However, in general, the Weissenberg number and Deborah number are not equal (Bird et al., 1987a) and the two parameters can be varied independently as demonstrated in the rotational flow geometries discussed in Chapter 5.

Having defined an appropriate residence time, it still remains to chose a suitable value for the characteristic fluid relaxation time $\lambda$. The simplest choice is to pick the single Oldroyd-B relaxation time $\lambda_1$ defined in eq. (4.25). This relaxation time has been calculated from the zero-shear-rate material functions determined in linear viscoelasticity to be $\lambda_1 = 0.794$ s; however, as has been demonstrated in Section 4.4.1, a single constant relaxation time is insufficient to describe the shear-rate-dependent behavior observed in Boger fluids. To reflect the gradual shear-thinning observed in the 0.31 wt% PIB/PB/C14 fluid, a shear-rate-dependent mean relaxation time $\lambda(\dot{\gamma})$ is defined from the viscometric properties measured in steady shear as

$$\lambda(\dot{\gamma}) \equiv \frac{\Psi(\dot{\gamma})}{2 \eta(\dot{\gamma})}$$  \hspace{1cm} (4.45)

This relaxation time decreases as the shear-rate increases, and in the limit of zero shear rate is equivalent to the relaxation time obtained from the upper convected Maxwell constitutive model (Bird et al., 1987a); $\lambda_0 \equiv \Psi_{\infty} / 2 \eta_0 = 0.325$ seconds. Shear-rate-dependent Deborah and Reynolds numbers are defined respectively by

$$De(\dot{\gamma}) \equiv \lambda(\dot{\gamma})\dot{\gamma} = \lambda(\dot{\gamma})\langle v \rangle / L$$ \hspace{1cm} (4.46)

$$Re(\dot{\gamma}) = \frac{2 \rho \langle v \rangle L}{\eta(\dot{\gamma})}$$ \hspace{1cm} (4.47)
It is also noted that the definition of a shear-rate-dependent Deborah number given by equation (4.46) is closely related to the stress ratio \( N_f/\tau \) which has often been used in the past to measure the importance of elastic effects in the flow of polymer melts

\[
De(\dot{\gamma}) = \frac{\Psi_1(\dot{\gamma})\dot{\gamma}}{2\eta(\dot{\gamma})} = \frac{1}{2} \left( \frac{N_f}{\tau} \right)
\]  

(4.48)

The quantity \( N_f/2\tau \) is also identified in the literature as the recoverable shear (Petrie and Denn, 1976; Cable and Boger 1979). In the past, it has been common to use definitions of the Deborah number and Reynolds number that are based on the constant zero-shear-rate viscoelastic properties (see for example Bisgaard, 1983; Lawler et al., 1986). This can lead to significant over-estimation of the importance of elasticity in the flow as evidenced by Figure 4.21, which shows a comparison of \( De(\dot{\gamma}) \), \( Re(\dot{\gamma}) \) with the flow parameters \( De_0 \), \( Re_0 \) based on zero-shear-rate properties. The shear-rate-dependent Deborah number for the 0.31 wt\% PIB/PB/C14 fluid is calculated from the viscometric data using eq. (4.43) and is plotted in Figure 4.21 together with the predictions of the 4-mode Bird-DeAguiar model. As the shear rate is increased, the relaxation time \( \lambda(\dot{\gamma}) \) decreases and the shear-rate-dependent Deborah number slowly diverges from the linear function \( De_0 \). At a shear-rate of \( \dot{\gamma} = 100 \text{ s}^{-1} \) this shear thinning results in a value of \( De(\dot{\gamma}) = 0.17 De_0 \). The high viscosity of the solution results in the very low magnitude of the Reynolds number shown in Figure 4.21. In addition, since the viscosity remains almost constant at high shear-rates the shear-rate-dependent quantity \( Re(\dot{\gamma}) \) diverges only slightly from \( Re_0 \). At a value of \( De(\dot{\gamma}) = 1 \), the effective Reynolds number is only \( Re(\dot{\gamma}) = 0.011 \), it is thus possible with the Boger fluid to study the importance of elastic effects in the absence of inertial flow effects.

The shear-rate-dependent Deborah number that can be achieved with this Boger fluid is compared with a number of other Boger fluid formulations in Figure 4.22. The values of \( De(\dot{\gamma}) \) for these other solutions were calculated from the published measurements of \( \Psi_1(\dot{\gamma}) \) and \( \eta(\dot{\gamma}) \) using equation (4.46). It can be seen from Figure 4.22 that while all the different formulations result in qualitatively similar shear-rate-dependent behavior, it is possible to attain a higher \( De(\dot{\gamma}) \) with this PIB/PB/C14 fluid at a given shear-rate than with any of the other Boger fluid formulations. With the same fluids handling system, it should thus be possible to extend our experimental observations to higher \( De \) than was previously possible by Lawler et al. (1986).
Figure 4.21  Predicted elastic and inertial flow parameters as a function of shear-rate for the 0.31 wt% PIB/PB/C14 Boger fluid: (-----) Shear-rate-dependent flow parameters $De(\dot{\gamma})$, $Re(\dot{\gamma})$ calculated using the 4-mode Bird-DeAguiar model; (----) flow parameters $De_0$, $Re_0$ calculated using the zero-shear-rate material functions; (●) experimental data calculated from the master curve (Fig. 4.14).
Figure 4.22  Comparison of the shear-rate-dependent Deborah number $De(\dot{\gamma})$ attainable in a number of Boger fluid formulations; (●) 0.31 wt% PIB/PB/C14 used in this thesis; (○) PIB/Pb/K fluid of Lawler et al. (1986); (□) PIB/PB/K fluid of Binnington et al. (1986); (▲) PAC/CS/H2O fluid of Boger and Nguyen (1979).
In addition to the Deborah number and Reynolds number defined above it is possible to construct an alternative dimensionless group which is known in the literature as the Elasticity number \( E \). This parameter is defined as a ratio of the Deborah number and Reynolds number and thus from eq. (4.46) and (4.47) may be expressed as

\[
E \equiv \frac{De}{Re} = \frac{\lambda \eta}{\rho L^2}
\]  

(4.49)

This parameter has been used by Boger to correlate vortex size in axisymmetric contraction flows of a number of different viscoelastic fluids (Nguyêñ and Boger, 1979) and more recently by Debbaut (1990) in calculations of inertial effects on the formation of lip vortices. The elasticity number \( E \) may be considered to be a ratio of the relative importance of elastic effects to inertial effects in the flow of viscoelastic liquids; high values of \( E \) correspond to almost inertialess creeping flows of highly elastic fluids (e.g. Boger fluids, polymer melts); whereas at low \( E \), inertial effects tend to swamp the elastic contributions to the flow field (e.g. in the flow of dilute, shear-thinning polymer solutions). It can also be seen from eq. (4.49) that the elasticity number has no direct dependence on kinematic quantities such as the fluid velocity \( \langle v \rangle \), or the shear-rate \( \dot{\gamma} \), and is purely a function of the particular fluid and test geometry under consideration. However, if the fluid viscosity \( \eta(\dot{\gamma}) \) and relaxation time \( \lambda(\dot{\gamma}) \) are functions of the shear rate then \( E \) will also indirectly vary with the shear rate, and decreases at high shear rates. The consequences of this decrease in \( E \) have been documented by Raiford et al. (1989) in experiments with a shear-thinning PIB/C14 solution; a small lip vortex was visually observed at \( De = 1.4 \) (\( E = 0.02 \)), however subsequent growth of this vortex was not observed at higher flow rates since the large increase in inertial effects outweighed the small increase in elastic effects.

The elasticity number for the 0.31 wt% Boger fluid can be calculated by using the zero-shear-rate values of the material functions determined in Section 4.4; \( \eta_0 = 13.76 \text{ Pa}\cdot\text{s} \), \( \lambda_0 = 0.325 \text{ s} \), \( \rho = 880 \text{ kg/m}^3 \). Both the axisymmetric contraction geometry and the final version of the cylinder flow geometry have characteristic lengths of \( L = 0.3175 \text{ cm} \) and the elasticity number in the limit of low shear-rates is \( E_0 \equiv 504 \). This value will decrease at high shear rates due to shear-thinning in \( \lambda(\dot{\gamma}) \), however it is readily apparent that elastic effects far outweigh inertial effects in this fluid/geometry system: In contrast the recent calculations of Debbaut (1990) covered the range \( 0.01 < E < 0.05 \).
Chapter 5

The Elastic Instability in Rotational Flows of Boger Fluids

A review of the literature pertaining to rotational or swirling flows of highly elastic Boger fluids has been discussed in detail in Chapter 2.1. A number of researchers have documented the departure of the flow away from the simple viscometric base flow; however, these observations have frequently been interpreted in the framework of an anti-thixotropic flow transition since measurements of the torque and normal force in a rheometer show a time-dependent, apparent shear-thickening of the viscosity and first normal stress coefficient.

In this chapter, rheological experiments using the cone-and-plate and parallel-plate fixtures with the mechanical spectrometer described in Chapter 3 are employed to explore this phenomenon in detail. The flow transition is shown to be an elastically-driven hydrodynamic instability that results in a time-dependent, three-dimensional secondary flow between the plates. Careful measurements of the critical conditions for the onset of the rotational flow transition coupled with video flow visualization are used to characterize both the spatial and temporal form of the instability. These results are compared with the previous measurements of Magda and Larson (1988) for Boger fluids containing higher molecular weight polymers, and with the theoretical predictions of Phan-Thien (1983, 1985) for the stability of rotational flows of the Oldroyd-B fluid model.

5.1 Flow Parameters

It is important to note that there are two independent nondimensional groups which measure the importance of the fluid elasticity in rotational viscometric flow, such as cone-and-plate and parallel-plate flow. The Deborah number is defined as a ratio of the viscoelastic relaxation time \( \lambda \) to the characteristic time scale \( \tau \) of the flow, whereas the Weissenberg number is defined as a product of the fluid relaxation time and a characteristic strain rate \( \dot{\gamma} \) in the flow (Bird et al., 1987a). For viscometric flow between a flat plate and a
cone rotating with an angular velocity $\Omega$ (as shown in Fig. 4.3), an appropriate characteristic time is $\mathcal{T} = 1/\Omega$. For a small cone angle $\theta_0$ the shear rate is uniform throughout the sample and the Deborah number and Weissenberg number are then

$$De^{(cP)} = \lambda \Omega \quad (5.1a)$$

$$We^{(cP)} = \lambda \Omega / \theta_0 \quad (5.1b)$$

For flow between parallel plates of radius $R$ and plate separation $H$, the characteristic time is the same as in the cone-and-plate geometry; however, the shear rate $\dot{\gamma}$ varies radially across the gap. It is conventional to take the characteristic shear rate as the rim shear rate $\dot{\gamma}_R = \Omega R / H$ experienced at the edge of the disks, and the dimensionless groups are then given by

$$De^{(pp)} = \lambda \Omega \quad (5.2a)$$

$$We^{(pp)} = \lambda \Omega R / H \quad (5.2b)$$

Hence, at a fixed rotation rate $\Omega$, it is possible to vary the relative magnitude of $De$ and $We$ by changing either the cone angle $\theta_0$ in the cone-and-plate system or the aspect ratio $R/H$ of the parallel disks.

The calculation of actual numerical values for both $De$ and $We$ depends on appropriate choices for the relaxation time $\lambda$ in equations (5.1) and (5.2). The detailed rheological characterization presented in Chapter 4 clearly showed that a single relaxation time is insufficient to describe quantitatively the nonlinear response of this highly elastic Boger fluid, and that either a spectrum of time constants or a shear-rate-dependent quantity $\lambda(\dot{\gamma})$ is actually required. However, to date, the only theoretical and numerical simulations relevant to rotational flows of Boger fluids have employed the single mode Oldroyd-B fluid model (eq. 4.25). For this reason, we have chosen to use the Oldroyd-B relaxation time which was determined in Chapter 4 from the zero-shear-rate viscometric properties of the Boger fluid to be $\lambda_1 = 0.794$ seconds. The experimental measurements obtained in this chapter can then be compared directly to the stability predictions of Phan-Thien presented in Chapter 2.3.

Inertial effects in the flow are measured by the Reynolds number, which is defined in terms of the finite plate radius, the rotation rate, and the viscosity of the fluid as

$$Re = \frac{\rho \Omega R^2}{\eta} \quad (5.3)$$
In theoretical and numerical studies of inertial rotational flows between infinite parallel disks, there is no characteristic length scale appropriate to describe the radius of the disks and results are commonly presented in terms of the *Ekman* number, defined as \( E = \frac{\eta}{(\rho \Omega H^2)} \). The inverse of this dimensionless group \( E^{-1} \) is directly related to the Reynolds number by the square of the aspect ratio of the plates \((R/H)^2\).

The results presented in this chapter correspond to a maximum rotation rate of \( \Omega = 13.6 \) rad/s and a Deborah number of \( De = 9.32 \). The high viscosity of the Boger fluid used in these experiments minimizes the importance of inertial effects in the flow and the maximum Reynolds number attained was \( Re = 0.159 \), equivalent to Ekman numbers in the range \( 0 < E^{-1} \leq 0.0023 \). These parameter values may be compared to those determined for inertial instabilities in purely Newtonian swirling flows; the recent review of Zandbergen & Dijkstra (1987) indicates that the von Kármán similarity solution is the unique steady-state, two-dimensional solution for values of \( E^{-1} \leq 55 \), while experiments with rotating disks in water find the onset of the primary flow instability to be in the range \( 210 \leq Re \leq 620 \) (Clarkson et al., 1980). Inertial effects in our experimental measurements are thus over three orders of magnitude smaller than those that would give rise to inertial flow instabilities.

5.2 Experimental Conditions

5.2.1 Data Acquisition and Analysis

All the experimental results presented in this chapter were obtained by using the Rheometrics RMS-800 Mechanical Spectrometer described in Chapter 4.2 with both the cone-and-plate and the parallel-plate test fixtures. In order to permit measurements of the large stresses that arise following onset of the flow instability the Spectrometer was fitted with the FRT-2000 'melt transducer' which permits measurement of torques up to 2000 g-cm and normal forces of up to 2000 grams. Measurements of the torque and normal force as functions of time were used to calculate the shear stress \( \tau(t) \) and first normal stress difference \( N_1(t) \) and then output directly to a strip-chart recorder and a personal computer. The data-acquisition software package *(Recap II)* controlling the RMS-800 samples the torque and normal force data at a maximum rate of 256 times per second and permits a maximum record length of 2048 measurements subdivided into 4 user-definable segments. The experiments presented below were performed in the following manner; (i) 512 data points were sampled in the initial 30 seconds, in order to capture in
detail the transient response associated with the start-up of steady shear flow, (ii) another 512 samples equally spaced over 6 minutes captured the onset of the flow transition and the time-dependent response of the torque and normal forces, (iii) finally, in a limited number of experiments, another 512 samples spaced over a period of up to 50 minutes were measured to determine accurately the critical rotation rate for onset of the flow instability.

As discussed in Chapter 4.1.2, shear flow between coaxial parallel disks is nonhomogeneous, and the first normal stress difference \( N_1 \equiv (\tau_{11} - \tau_{22}) \) is not given explicitly in terms of the measured normal force \( \mathcal{F} \). Therefore, measurements of the normal force in the parallel-plate geometry strictly yield an ‘apparent first normal stress difference’, which has to be corrected to account for an implicit term of the form \( d(\ln \mathcal{F})/d(\ln \gamma) \). This correction is straightforward in simple ‘rate-sweeps’ where \( \mathcal{F} \) and \( \mathcal{G} \) are measured as a function of \( \gamma \); however, it is more difficult in time-dependent experiments. Unlike the measurements in the previous chapter, the normal force observations presented below are intended to demonstrate the onset and growth of a flow instability and not to yield precise material functions, therefore this correction has not been performed. Similar arguments apply to measurements of the torque \( \mathcal{G} \) in the parallel-plate geometry; however, this latter correction is negligible for constant viscosity Boger fluids. Thus for the parallel plate geometry results are reported as an ‘apparent shear stress’ \( \tau_a \equiv \eta_a \dot{\gamma}_R = (2\mathcal{G}/\pi R^3) \) and an ‘apparent first normal stress difference’ \( N_{1a} \equiv \Psi_{1a} \dot{\gamma}_R^2 = (4\mathcal{G}/\pi R^2) \). Note also that any contribution from \( \Psi_2 \) is ignored in this latter expression.

\[ \textbf{5.2.2 Temperature Effects and their Correction} \]

The experimental results presented in Chapter 4.4 were obtained using the more sensitive FRT-100 ‘fluids transducer’ which incorporates a refrigerated fluid bath that affords extremely accurate temperature control (± 0.1°C) in the sample. The larger FRT-2000 transducer used in the measurements of the flow instability is designed primarily for work with polymer melts and offers poor temperature control (± 5°C) at typical room temperatures. It has already been shown that viscometric properties such as the viscosity and relaxation time spectrum of the 0.31 wt% PIB Boger fluid are extremely sensitive to temperature (see Chapter 4.4.1), and that these effects may be described by an Arrhenius expression (eq. 4.40) with a flow activation energy of \( \Delta H/\mathcal{R} = 7362 \) K. Thus to interpret measurements of the flow stability of Boger fluids accurately, it is crucial to monitor the ambient laboratory temperature closely and to adjust the material functions accordingly.
Fluid samples were placed between the plates and allowed to stabilize thermally before each run. The temperature was also monitored constantly in each experiment by using a thermocouple mounted on the bottom surface of the lower plate, and the average temperature was then used to calculate the Deborah number attained in the flow as described below. Fresh fluid samples were used for each experiment in order to reduce the possible effects of polymer degradation. From knowledge of the viscometric properties at the reference temperature $T_0 = 25.0$ °C and the time-temperature superposition principle, the true values of the Deborah number and Weissenberg number in an experiment performed with a fluid sample at temperature $T$ are found from the following relations

$$De = \lambda_1(T_0) a_T \Omega \quad (5.4a)$$

$$We = \lambda_1(T_0) a_T \dot{\gamma} \quad (5.4b)$$

Similarly, the Reynolds number may be corrected by accounting for the temperature variation in the fluid viscosity as

$$Re = \frac{\rho \Omega R^2}{\eta(T_0) a_T} \quad (5.5)$$

In this expression the temperature dependence of the fluid density over the narrow range of temperatures in these experiments is neglected.

5.3 Experimental Results

Although qualitatively similar results are obtained for both the cone-and-plate and parallel-plate geometries, for the sake of clarity we present the experimental results and their comparison to the theoretical predictions given by eqs. (2.2) and (2.3) separately in Sections 5.3.1 and 5.3.2. Observations of the temporal and spatial form of the flow instability are then discussed in detail in Section 5.4. The applicable Deborah number and Weissenberg number of the flow in each geometry are calculated from eqs. (5.4a) and (5.4b) by using the Oldroyd-B relaxation time $\lambda_1 = 0.794$ s (evaluated at at $T_0 = 25$°C), the temperature shift factor $a_T$ at temperature $T$ (calculated from eq. 4.30), the rotation rate of the lower plate, and the appropriate geometrical parameters.
5.3.1 The Cone-and-Plate Geometry

The experimentally measured shear stress $\tau(t, \dot{\gamma})$ and first normal stress difference $N_1(t, \dot{\gamma})$ upon the inception of steady shear flow are shown in Figure 5.1. The initial transient response is typical of a Boger fluid; at short times the shear stress shows a rapid initial increase due to the presence of the Newtonian solvent, and the first normal stress difference grows quadratically. A local maximum is observed in each of the measured stresses, and the position of this overshoot shifts to progressively earlier times as the deformation rate is increased. As is well known, these nonlinear effects cannot be described by the quasi-linear Oldroyd-B constitutive model which predicts a simple monotonic increase in $\tau(t)$ and $N_1(t)$, with no dependence on the applied shear rate; however, the behavior can be modeled by the multimode nonlinear constitutive equations discussed in Section 4.4.3.

Extending the experimental measurements to longer times reveals the so-called anti-thixotropic transition, as demonstrated in Figures 5.2(a) and (b). The shear stress and normal stress difference initially remain constant for a period of time before rapidly increasing to a new, time-dependent state. Calculation of the viscosity and first normal stress coefficient from time-averaged values of these force measurements thus indicate an apparent shear-thickening of the fluid sample. It is shown in Section 5.4 that this is not a change in the fluid structure, but the onset of a rotational flow instability. The 'induction time' for the onset of this instability ranges from several minutes to a few seconds and decreases as the angular velocity of the plate is increased. Comparing Figures 5.2(a) and (b) shows that the response of the shear stress and the normal stress is qualitatively very similar, with an identical induction time and comparable time-dependent fluctuations. For brevity, only one of the measured stress components is presented in subsequent figures; however, it is emphasized that in each experiment the onset of a flow transition results in corresponding temporal behavior in both $\tau(t)$ and $N_1(t)$.

The data presented in Figure 5.2 suggest that cone-and-plate flow of the Boger fluid remains stable at $De = 4.41$; however, experiments over longer observation times show that it is in fact unstable. In previous experiments with more elastic Boger fluids, Magda and Larson (1988) have documented extremely long induction times of up to 3000 seconds; therefore, a number of one-hour long experimental runs were conducted over a narrow range of rotation rates to determine accurately the critical Deborah number for the onset of this instability. The shear stress measured at Deborah numbers of $De = 4.37$ and 4.41 is shown in Figure 5.3 for a cone angle of $\theta_0 = 0.10 \text{ rad}$. At $De = 4.37$ the flow remains steady for one hour. Although it is possible that the instability could develop at an even
Figure 5.1  Initial transient response of (a) the shear stress $\tau(t)$ and (b) the first normal stress difference $N_1(t)$ in the cone-and-plate geometry upon inception of steady shear flow as the rotation rate $\omega_0$ and Deborah number are increased.
Figure 5.2  Onset of a flow transition and anti-thixotropic behavior in (a) the shear stress and (b) the first normal stress difference after a period of steady-shear flow in the cone-and-plate rheometer.
later time, experiments of more than 3600 seconds were deemed impractical. The critical Deborah number is thus determined from these measurements to be $De_c = 4.39 \pm 0.01$, which is equivalent to a critical Weissenberg number of $43.9 \pm 0.1$.

The values of the zero-shear-rate material functions determined in Chapter 4.4 are used to determine the viscosity ratio $\beta$ for this Boger fluid as $\eta_s / \eta_0 = 8.12 / 13.76 = 0.59$. Phan-Thien's analysis for cone-and-plate flow of a fluid described by an Oldroyd-B constitutive model with $\beta = 0.59$ is computed from eq. (2.3) to be unstable at a critical Deborah number of $De_c^{(cp)} = 3.10$. The agreement between the theoretical and experimental values is thus reasonably good (to within 40%), and is comparable to the size of the discrepancies measured by Magda and Larson. A detailed discussion of Phan-Thien's linear stability analysis has been presented in Chapter 2.1. In particular it is noted that the analysis is limited to steady, axisymmetric disturbances of the similarity form and to fluids that are described by the quasi-linear Oldroyd-B model. These restrictions are severe, and detailed studies of the spatial and temporal structure of the flow (presented in Section 5.4) show that the flow transition resulting in the stress responses shown in Figure 5.2 is more complex than the form predicted by Phan-Thien.

### 5.3.2 The Parallel-Plate Geometry

Experimental measurements of the flow between parallel plates show a similar flow transition, as evidenced by Figure 5.4. An expanded view of the initial transient response of the shear stress $\tau(t)$ during start-up of steady shear flow is shown in Figure 5.4(a), and the subsequent onset of the flow instability at $De = 4.65$ ($We = 58.2$) and $De = 5.85$ ($We = 73.1$) is displayed in Figure 5.4(b). The flow at $De = 4.49$ remains steady for a period of one hour. The critical flow conditions for onset of the flow instability with a plate separation of $H = 1.00$ mm were determined from a series of experiments of one-hour duration, and were estimated to be $De_c = 4.54 \pm 0.02$, and $We_c = 56.8 \pm 0.3$. The theoretical stability criterion for an Oldroyd-B fluid is determined from eq. (2.2) to be $De_c^{(pp)} = 2.51$, so that the agreement between experimental and analytic predictions of flow stability is less satisfactory than in the cone-and-plate geometry.

In the parallel-plate geometry, it is possible to investigate independently the relative importance of both the Deborah number and the Weissenberg number by varying the aspect ratio $R/H$ of the system. The anti-thixotropic behavior of the apparent first normal stress difference as a function of increasing $De$ is shown in Figure 5.5. The rotation rate $\Omega$ is increased in each experiment and the plate separation $H$ is increased proportionally to

203
Figure 5.3 Shear stress in the cone-and-plate geometry for Deborah numbers of $De = 4.37$ and $De = 4.41$. The flow remains steady at $De = 4.37$, whereas flow at $De = 4.41$ demonstrates onset of the flow instability after approximately 400 seconds.
Figure 5.4  The initial transient response (a) and the long-time behavior (b) of the apparent shear stress $\tau_a(t)$ in the parallel-plate geometry as the rotation rate is increased from $\Omega = 5.65$ to 7.36 rad/s.
maintain constant values of the shear rate $\dot{\gamma}$ and $We$. The initial transients for all values of $De$ superpose, in agreement with the rheological premise that the stresses in the viscometric flow are unique functions of the applied shear rate, not the angular rotation rate of the experiment. However, as can be seen in Figure 5.5(b) the flow instability is a very sensitive function of $De$; the induction time decreases from over a minute to less than 5 seconds and the magnitude of the apparent shear-thickening \( [\tilde{N}_{1d}(t) - \Psi_{1d}(t)^2] \) increases by a factor of two as the Deborah number is increased from 4.65 to 9.32.

Increasing the Weissenberg number at a constant Deborah number has the inverse effect on the flow instability. The results displayed in Figure 5.6 are produced by holding the rotation rate constant at $\Omega = 5.86$ rad/s, corresponding to $De = 4.65$, and decreasing the plate separation to increase the shear-rate and Weissenberg number. Calibration experiments with a silicone fluid standard of known viscosity indicate that accurate measurements can be obtained in the RMS-800 for plate separations in the range $0.25 \leq H \leq 2.00$ mm. It is seen from Figure 5.6(a) that the initial plateau value of the shear stress increases approximately linearly with $We$, as expected for a fluid of constant viscosity. As $We$ increases, the induction time for onset of the instability increases, and the magnitude of the observed shear-thickening \( [\tilde{\tau}_d(t) - \eta_d(\dot{\gamma} t)] \) decreases significantly. A more detailed analysis of these experimental observations is presented in Section 5.4.

Hysteresis and the presence of multiple, steady stress states at a constant shear rate have been demonstrated with controlled stress rheometers by Magda and Larson (1988) and Laun and Hingmann (1990). Since no controlled stress rheometer is available at M.I.T., this hysteresis has been investigated with the RMS-800 by devising a series of 'step-shear-rate' experiments. The results shown in Figures 5.7(a) and (b) are typical of the results obtained in the parallel-plate geometry over a range of shear rates: From an initial set of experiments the critical shear-rate at a plate separation of $H = 1.80$ mm was determined to be $\dot{\gamma}_c = 42.3$ s$^{-1}$, corresponding to a Deborah number of $De_c = 4.83$. The dashed line in Figure 5.7 shows the steady shear stress and normal stress measurements obtained just below the critical value at $\dot{\gamma} = 42.2$ s$^{-1}$, and which remain constant for at least 2400 s. Steady shear flow at $\dot{\gamma} = 40.5$ s$^{-1}$ ($De = 4.63$) is established in a fresh fluid sample for two minutes and is shown in Figure 5.7 by the solid line. The flow is below the critical Deborah number, and measurements of the stresses in other fluid samples at the same shear rate remain steady for at least 2400 seconds (see the dotted lines in Figure 5.7), showing no anti-thixotropic transition. After two minutes a step increase in the shear-rate (to $\dot{\gamma} = 54.0$ s$^{-1}$) is applied to the sample and results in an initial stress overshoot, followed by a short plateau region. The new shear rate of $\dot{\gamma} = 54.0$ s$^{-1}$ ($De = 6.18$) is above the critical conditions for onset of the flow instability, and a rapid increase in both the shear
Figure 5.5  The initial transient response (a) and the long-time behavior (b) of the apparent first normal stress difference $N_{1a}(t)$ at a constant Weissenberg number of $We = 58.2 \pm 1.0$ as the Deborah number is increased.
Figure 5.6  The initial transient response (a) and the long-time behavior (b) of the apparent shear stress $\tau_{a}(t)$ at constant Deborah number $De = 4.65 \pm 0.10$ as the Weissenberg number is increased from $We = 29.1$ to 172.
Figure 5.7 Hysteresis in (a) the shear stress and (b) the normal stress above the critical Deborah number in the parallel-plate geometry. The stresses at $\dot{\gamma} = 40.5 \text{ s}^{-1}$ ($De = 4.63$) are different before and after shearing for 2 minutes at $\dot{\gamma} = 54.0 \text{ s}^{-1}$ to induce onset of the flow instability. For comparison, the steady values of the stresses measured just below the critical shear-rate $\dot{\gamma}_c = 42.2 \text{ s}^{-1}$ ($De = 4.83$) are shown as dashed lines (---).
and normal stress is observed. To prevent excessive degradation of the polymer, this shear-rate is only maintained for two minutes before being reduced back to the previous shear-rate; however, other measurements, such as those shown in Figures 5.4 – 5.6, indicate the flow remains unsteady indefinitely at this $De$. The stress histories presented in Figure 5.7 show that, upon returning to the initial shear-rate, both the shear stress and normal stress remain in a time-dependent unsteady state significantly above the original steady values shown by the dotted line. Thus, multiple stress states are possible at a single shear rate. A series of such experiments are presented in the next section to help evaluate the nature of the bifurcation to unstable flow.

5.4 Characteristics of the Rotational Flow Instability

A series of hysteresis experiments such as the one described above were performed with fresh samples over a range of shear rates to construct a stress – shear-rate bifurcation diagram. The shear-stress data for parallel plates separated by a gap of $H = 1.80$ mm are shown in Figure 5.8 in a local form (Iooss and Joseph, 1980), which is constructed by time-averaging the ultimate value of the stress $\tau_a(t)$ and then subtracting the base solution for steady shear flow, $\tau_a = \eta_0 \dot{\gamma}$. The flow remains steady up to a critical shear rate of $\dot{\gamma}_c = 42.3$ s$^{-1}$; however, above this value the base solution is unstable and a small increase in the shear-rate results in a rapid increase in the stress as the flow bifurcates to a new anti-thixotropic, time-dependent behavior. By decreasing the rotation rate of the plate, it is possible to follow this time-dependent solution family below the critical shear-rate, as shown in Figure 5.8. It is important to note that this curve does not intercept the base solution family; below a shear-rate of $\dot{\gamma} = 33.7$ s$^{-1}$ the time-dependent oscillations of the shear stress decay, and the flow returns to the initial steady state. This return to the stable base solution is demonstrated by the first normal stress measurements shown in Figure 5.9. A step–shear-rate experiment was carried out on a fresh fluid sample with the following deformation history; (a) 120 seconds at a (temperature corrected) shear rate of $\dot{\gamma}a_T = 53.9$ s$^{-1}$, (b) 180 s @ $\dot{\gamma}a_T = 38.9$ s$^{-1}$, (c) 300 s @ $\dot{\gamma}a_T = 35.6$ s$^{-1}$, (d) 600 s @ $\dot{\gamma}a_T = 32.7$ s$^{-1}$. The first shear rate (a) is significantly above the critical shear rate of $\dot{\gamma}_c = 42.3$ s$^{-1}$ and following the initial stress overshoot the flow rapidly becomes unstable; the normal stress difference increases by a factor of four and exhibits rapid temporal oscillations. On subsequently reducing the shear rate in steps (b) and (c) the normal stress difference decreases but remains significantly above the value measured at the subcritical shear rate of $\dot{\gamma} = 42.2$ s$^{-1}$ (shown in Figure 5.9 by the dashed line). However, decreasing
Figure 5.8  Local form of the flow instability in the parallel-plate geometry for a plate separation of $H = 1.80$ mm. The viscometric base solution (○) becomes unstable at $\gamma_c = 42.3$ s$^{-1}$, and the flow becomes time-dependent and three-dimensional (◇).
Figure 5.9  (——) The apparent first normal stress difference $N_{1a}$ measured in a 'step-shear-rate' experiment with four successively lower shear rates of $\gamma a_T = 53.9$, 38.9, 35.6 and 32.7 $s^{-1}$. The dashed line (—) shows the steady value of $N_{1a}$ measured at the subcritical shear-rate of $\gamma = 42.2$ $s^{-1}$.
the shear-rate further in step \((d)\) to \(\dot{\gamma} a_f = 32.3 \text{ s}^{-1}\) results in a slow decay of the oscillations over a period of approximately 120 seconds and the flow returns to the steady base flow given by \(N_{1a} = \Psi_{1a} \dot{\gamma}^2\).

These results, coupled with the constant-stress measurements of Magda and Larson (1988), are indicative of a subcritical Hopf bifurcation in the rotational flow of Boger fluids, as shown schematically by the solid line in Figure 5.8. Near a supercritical Hopf bifurcation, small increases of the control parameter (represented here by the shear rate or the Deborah number) result in small perturbations to the solution field, since a stable, time-periodic flow exists near the unstable fixed point in phase space. Careful experiments are thus able to reveal the sequence of nonlinear flow transitions that lead to aperiodic or chaotic flow regimes (see for example Bergé et al. 1986; Baker and Gollub, 1990). However, small perturbations about a subcritical bifurcation transport the solution to a distant part of the phase-space and lead immediately to large disturbances in the solution field. Such nonlinear instabilities are well-known in the field of fluid dynamics, especially in the initial stages of turbulent transition in Newtonian shear flows (Bayly, Orszag and Herbert, 1988).

### 5.4.1 Temporal Form of the Instability

Dynamic information about the energy distribution and evolution of complex time-varying systems can be obtained by calculating the Fourier spectrum of a time-dependent signal. A Fast-Fourier Transform (FFT) of the time-dependent shear stress \(\tau_a(t)\) measured between parallel plates at \(De = 4.65\) is shown in Figure 5.10. The total record length of the time-dependent signal is \(\Delta T = 560 \text{ seconds}\) and the resolution of the frequency spectrum is thus \(1/\Delta T = 0.0018 \text{ Hz}\). The power spectrum shows the presence of a weak peak \(f = 0.025 \text{ Hz}\) and a number of harmonics which are buried in broad-band noise. The spectral distribution of the FFT presented in Figure 5.10 clearly indicates that this nonlinear flow consists of a number of temporal modes which interact with each other to produce the complex, time-dependent measurements of the shear and normal stresses. Experimental sources of error such as poorly aligned disks or plates which are not mounted parallel might be expected to result in a mechanically-forced oscillatory system. This 'forcing frequency' would be apparent in the Fourier spectrum as a peak at a frequency of \(\Omega/2\pi = 0.93 \text{ Hz}\). This frequency is significantly higher than the peaks present in Figure 5.10 and it is concluded that no such forcing is present and the frequency spectrum corresponds purely to oscillations in the torque arising from the hydrodynamic instability.
Figure 5.10 Fast Fourier transform (FFT) of the time-dependent shear stress $\tau_a(t)$ in the parallel-plate geometry at $De = 4.65$. The power spectrum shows a number of peaks in a background of broad-band noise.
A similar highly nonlinear instability appears to exist in the Couette-Taylor flow of elastic liquids (Larson et al., 1990; Northey et al., 1991) and observations of the torque in a Couette cell show the same nonlinear time-dependent response.

It should also be possible, by carefully controlling the Deborah number, to observe the development and exponential growth of the initial disturbance mode. The local form of the anti-thixotropic increase in the first normal stress difference measured in the cone-and-plate geometry is shown in Figure 5.11 as a function of the Deborah number. This semilogarithmic plot demonstrates that the initial growth of the instability in the cone-and-plate geometry is indeed exponential. Ultimately, however, nonlinear terms inhibit this growth and result in the formation of a new temporally aperiodic state. Similar results are also obtained in the parallel-plate geometry.

These observations are the first investigations of the temporal form of the rotational flow instability in Boger fluids. The earlier observations of Magda and Larson (1988) documented the onset of the flow instability, but were unable to follow its subsequent evolution due to limits in the maximum measuring range of the force transducer. The theoretical and numerical calculations of Phan-Thien also predict an initial exponential growth of this flow instability, in agreement with the experimental data; however, his linear stability analysis provides no information about the interaction with nonlinear terms or the formation of subsequent time-dependent states.

Magda and Larson (1988) did not present any information on the temporal response of the flow instability and were also unable to characterize the spatial structure of the unsteady flow that develops between the plates. The Phan-Thien analysis, however, makes very specific predictions about the spatial form of the most unstable eigenfunction (see Chapter 2.1), in addition to providing a critical value of the Deborah number. If the torque and normal force measurements presented above do correspond to a flow transition of the type considered by Phan-Thien, then it should be possible to observe the toroidal recirculation predicted by the stability analysis.

5.4.2 Spatial Form of the Instability

In order to investigate the spatial characteristics of the secondary flow, the video-imaging system was focused on the free-surface of the fluid sample at the outer edge of the parallel plate geometry. If the experimentally observed disturbance does correspond to the Phan-Thien instability, then the resulting flow should remain axisymmetric and two-dimensional. However, careful frame-by-frame analysis of the videotape reveals that even
Figure 5.11 Initial exponential growth of the disturbance as measured by the first normal stress difference above the base response given by $N_1 = \Psi_1 \dot{\gamma}^2$ for steady shear flow.
the initial exponential growth period of the flow transition results in a nonaxisymmetric
deformation of the fluid surface. In order to demonstrate this, sample photographs of the
free-surface shape in the parallel-plate geometry during an experiment at $De = 5.81$
($= 1.2De_c$) are shown in Plate 5.1, together with an accompanying history of the normal
force measurement. The exposure points of each photograph are located in plate 5.1(a) by
the solid circles. At $t = 20$ seconds the flow shown in Plate 5.1(b) is steady, and the fluid
sample between the plates is approximately cylindrical in shape. After 30 seconds the flow
becomes unstable and the shear-stress and normal stress difference begin to increase
rapidly. The shape of the meniscus in the exponential growth region at $t = 32$ s is shown in
Plate 5.1(c). The surface of the sample is already nonaxisymmetric and severely deformed.
In the fully nonlinear, time-dependent regime that develops at longer times the free surface
shows more rapid fluctuations. The meniscus shape shown in Plate 1(d) at $t = 100$ s has a
complex dependence on both the axial and azimuthal coordinate. This time-dependent
evolution in the free-surface can be seen far more clearly on the accompanying videotape
(see Appendix A).

Although photographs such as those in Plate 5.1 can convey some information
about the nonaxisymmetric disturbance, they reveal little detail of the secondary flow
structure within the bulk of the fluid sample. In an attempt to obtain some qualitative
observations of this spatial form, a simple glass-base rheometer was constructed and is
shown schematically in Figure 5.12. A flat aluminum disk of radius $R = 37.87$ mm was
mounted on the spindle of a d.c. controlled electric motor (ElectroCraft #004) to form the
top plate of a parallel-plate device. The lower plate comprised of a glass laboratory dish
rigidly attached to an optical mount by a large circular clamp. The motor was mounted
vertically on the side of the computer-controlled translating table described in Section 3.1.
By lowering the table until the upper disk touched the lower dish, the two horizontal
surfaces could be adjusted for parallelism and concentricity; subsequent separation of the
plates could then be measured to within $\pm 4$ $\mu$m by moving the translation table vertically.
The video-camera was fitted with a macro lens (Minolta MD Macro 50 mm $f/3.5$), mounted
beneath the glass dish and then focused on the fluid between the parallel plates. The fluid
was illuminated with a standard microscope illuminator (Reichert S653) and a tiny quantity
of seed particles (0.1 wt% rheoscopic concentrate; Kalliroscope Corporation, Groton, MA)
was added to improve the visualization of the secondary flow.

The videotape described in Appendix A contains a clip showing the onset and
development of the flow instability, and representative still frames are reproduced in
Plate 5.2 for a plate separation of $H = 4.81$ mm ($R/H = 7.87$). In order to enhance
resolution of the fine details, an enlarged view of 1/6 of the whole flow field is contained in
Plate 5.1 Evolution of the free-surface shape in the parallel-plate geometry at a rotation rate of $\Omega = 7.32$ rad/s and a plate separation $H = 1.80$ mm; (a) Evolution of the apparent first normal stress difference $N_{1a}$; (b) Symmetric meniscus shape at $t = 20$ s.
Plate 5.1 (cont.)  (c) Asymmetric shape of the free surface during exponential increase in $N_{1a}$ at $t = 31$ seconds; (d) Ultimate surface shape showing the complex dependence on axial and azimuthal position at $t = 100$ s.
Figure 5.12 Schematic diagram showing components of apparatus for video-imaging of the rotational flow instability.
each photograph. The experiment is of the ‘step–shear-rate’ form and the rotation rate is rapidly increased at $t = 1:00$ min from a subcritical value of $\Omega = 3.48$ rad/s ($De = 3.57$) to a higher, supercritical value of $\Omega = 4.57$ rad/s ($De = 4.69$). Plate 5.2(a) shows the fluid streamlines in the flow at $t = 41.9$ s, before the onset of the flow instability. The viscometric flow between the plates results in axisymmetric streamlines which are concentrically aligned about the rotation axis. The edge of the fluid sample is easily identified by the pronounced change in brightness towards the right edge of each image. The small azimuthal streak-lines observed in the otherwise smooth fluid are believed to originate from small foreign objects such as dust particles. At $t = 1:00$ min the rotation rate increases, the flow is now above $De_c$ and the time-dependent response of the stresses is of the form typified by Figure 5.4 with an onset time of approximately 30 seconds. It can be seen from Plate 5.2(b) that the onset of the flow instability is accompanied by a significant change in the spatial structure of the flow. The fluid streamlines are no longer purely axisymmetric but begin to develop a banded radial structure. These ‘roll cells’ are approximately axisymmetric and appear to have a constant radial wavelength which is roughly equal to the plate separation $H$. However, the cells are not steady in time and they propagate both radially outwards from the center of the disk, and radially inwards from the outer rim. This can be seen clearly by examining the pictures in Plate 5.2(b) and (c) which are separated by a time of 10 seconds. When the two sets of roll cells meet each other near the middle of the disk the flow undergoes another significant change in structure as shown in Plate 5.2(d). The flow is no longer axisymmetric and instead consists of a number of irregularly-sized cells which are observed on the videotape to rapidly spiral both inwards and outwards.

Unfortunately, this flow visualization device does not incorporate a force transducer so it is not possible to unequivocally equate these observations with the stress measurements performed in the RM' 800. However, from close observations in both geometries it appears clear that the initial formation and radial growth of the cells results in the initial exponential increase in the shear stress and normal stress difference shown in Figure 5.11, whereas the asymmetric spiralling recirculations documented in Plate 5.2(d) give rise to the irregular temporal oscillations in the stress observed at long times. The fine-scale structure and broad distribution of cellular sizes that can be seen in this photograph also account for the broad-band noise observed in Fourier transforms such as Figure 5.10, since it is clear that there is not one single characteristic wavelength present in the secondary flow. It thus appears that the neutral stability curve for this problem is extremely flat and that a large number of unstable modes are present in the ultimate nonlinear flow that develops.
Plate 5.2 Visualization of the spatial structure of the rotational flow instability in the parallel plate geometry with $H = 4.81$ mm. (a) Viscometric flow at $De = 3.57$; (b) Unstable flow at $De = 4.69$ showing formation of a radial cellular structure at $t = 1:30$ min.
Plate 5.2 [cont.]  (c) Radial propagation of secondary flow across the disk at $t = 1:40$ min; (d) Three-dimensional, time-dependent structure ultimately observed in the flow at $t = 2:10$ min.
Finally it is noted that these observations do not match the linear stability predictions of Phan-Thien discussed earlier. Whereas the experimentally determined onset point of the flow instability in each geometry compares moderately well with the predictions of eqs. (2.2) and (2.3), the observations of the temporal spectrum and spatial form of the secondary flow that ultimately develops are not accurately explained. The normal-mode analysis of Phan-Thien predicts the most unstable mode to have an axial wave-number \( k = 1 \), corresponding to a single, toroidal recirculating cell (for more details, see the discussion in Section 2.1). Higher wave-number instabilities would represent tori that are stacked in the axial dimension between the plates, and the similarity form of Phan-Thien is completely incapable of describing the time-dependent, radial cellular structure shown in Plate 5.2.

5.4.3 A Flow Stability Diagram for Rotational Flow of Elastic Liquids

The large number of experimental results obtained over a range of temperatures, angular rotation rates, plate sizes, and plate separations may be reduced to a master diagram representing the stability of rotational flow in the parallel-plate geometry. Such a stability diagram is shown in Figure 5.13 by calculating the Deborah number and Weissenberg number from equations (5.2a) and (5.2b) for parallel-plate flow of the 0.31 wt% PIB Boger fluid. This parameter space clearly shows the domain of the flow instability, and may be explored in a number of ways: (i) standard ‘rate-sweeps’, with a fluid sample between parallel plates of fixed separation \( H \), traverse lines of constant slope \( We/De \equiv R/H \); (ii) experiments at constant rotation rate \( \Omega \), such as those shown in Figure 5.6, result in vertical translation through Figure 5.13; (iii) conversely, experiments at a constant shear rate (see Figure 5.5) lead to horizontal paths in the parameter space. The linear stability analysis of Phan-Thien (eq. 2.2) predicts that for \( \beta = 0.59 \), unstable flow occurs above a Deborah number of \( De_{c}^{(pp)} = 2.51 \). This analysis is valid for an Oldroyd-B fluid confined between infinite coaxial disks and should thus be approached asymptotically in experiments as the aspect ratio of the plates \( R/H \rightarrow \infty \). It is seen from Figure 5.13 that high rotation rates are required to generate unstable flow for the lowest aspect ratios of \( R/H = 6.25 \). As the plate separation is decreased the critical shear-rate and critical Weissenberg number for unstable flow increase, as first described by Magda and Larson. For intermediate Weissenberg numbers, \( 30 \leq We \leq 160 \), the neutral stability locus also moves towards the theoretical value marked in Figure 5.13 by a broken line. However, as the aspect ratio is increased to its highest value of \( R/H = 50 \), the unstable region moves back towards higher
Figure 5.13 Stability diagram for viscometric flow of the Boger fluid between coaxial rotating disks. Simple shear flow is stable at low Deborah numbers (O) and becomes unstable with respect to a time-dependent, three-dimensional disturbance at high rotation rates (●). The prediction of the linear stability analysis of Phan-Thien (1983) is shown by the dotted line (-----).
values of \( De \). This restabilization appears to be a consequence of the severe shear-thinning in the first normal stress coefficient \( \Psi_1(\dot{\gamma}) \) that is observed in Boger fluids at high shear rates. The increase in stability resulting from shear-thinning behavior is also corroborated by a preliminary stability investigation performed by Phan-Thien (1983).

This shear-thinning in the first normal stress coefficient results in a significant decrease of the effective relaxation time \( \lambda_1(\dot{\gamma}) \) below the Oldroyd-B relaxation time of \( \lambda_1 = 0.794 \) s. It does not appear that this effect can be avoided by using more elastic fluids such as those developed by Magda and Larson; although their larger relaxation times do permit higher \( We \) to be obtained at lower shear rates, they also start shear-thinning at lower shear rates! The effect of shear-thinning on \( De \) and \( We \) in the parallel-disk flow can be taken into account empirically by introducing a shear-rate-dependent relaxation time, defined analogously to that for the Oldroyd-B fluid

\[
\lambda_1(\dot{\gamma}) = \frac{\Psi_1(\dot{\gamma})}{2\eta_p(\dot{\gamma})} = \frac{\Psi_1(\dot{\gamma})}{2[\eta(\dot{\gamma}) - \eta_s]}
\]

(5.6)

Appropriate shear-rate-dependent dimensionless quantities, \( De(\dot{\gamma}) \) and \( We(\dot{\gamma}) \), may then be defined by replacing \( \lambda_1 \) with \( \lambda_1(\dot{\gamma}) \) in equations (5.4a) and (5.4b). The shear-rate-dependent relaxation time defined by eq. (5.6) is evaluated by using the parameters of the 4-mode Bird-DeAguiar model fit listed in Table 4.4 to calculate \( \Psi_1(\dot{\gamma}) \) and \( \eta(\dot{\gamma}) \), as shown in Figure 4.14. Consideration of the shear-thinning in \( \Psi_1(\dot{\gamma}) \) that occurs in Boger fluids at high shear rates leads to significantly lower values of the relaxation time. For example, at a shear-rate of \( \dot{\gamma} = 100 \) s\(^{-1}\), the ratio of relaxation times is \( \lambda_1(\dot{\gamma})/\lambda_1 = 0.28 \). The recalculated stability diagram for the parallel-plate geometry is shown in Figure 5.14. From this diagram it can be seen that even if an instability of the steady similarity form analyzed by Phan-Thien does exist, it will not be observed in experiments with this Boger fluid due to the presence of a more unstable mode that occurs at lower Deborah numbers and leads ultimately to three-dimensional, time-dependent flow. Boger fluids are most accurately described by the Oldroyd-B model at low shear rates and we would thus expect the Phan-Thien analysis to be valid at low values of the shear-rate-dependent quantity \( We(\dot{\gamma}) \), in general agreement with the trend demonstrated in Figure 5.14, however, more detailed conclusions can not strictly be inferred from such empirical calculations.

A stability diagram similar to Figure 5.13 has also been constructed for cone-and-plate flow; however, it is more difficult to explore fully the parameter space since a large number of cones with a distribution of cone angles is required. The results for two cone
Figure 5.14  Stability diagram for viscometric flow between parallel plates, recalculated by using a shear-rate-dependent relaxation time $\lambda_1(\dot{\gamma})$ that more accurately represents the actual magnitudes of elastic effects in the Boger fluid.
angles of $\theta_0 = 0.04$ and 0.10 radians are shown in Figure 5.15 and lie along lines of slope $We/De = 1/\theta_0$. The general trends are qualitatively the same as described above and the agreement with the critical value given by equation (2.4) is closer than that obtained for the parallel-plate geometry. This may arise since for a specified rotation rate, the Weissenberg number and shear-rate in a typical cone-and-plate geometry are lower than those obtained in a parallel-plate geometry. The reason that this elastic flow instability was not observed in the earlier rheological measurements of Quinzani et al. (1990) is clearly explained in Figure 5.15: the fluid characterization described in Chapter 4.3 was performed using only the 0.04 radian cone, which corresponds to the steeper line in Figure 5.13. In this configuration, high shear-rates (and thus high values of $We$) can be achieved at low Deborah numbers which remain below the critical value $De_c^{(CP)}$, and the one-dimensional viscometric flow remains stable up to the maximum measurable limit imposed by the range of the force transducer.

These stability diagrams can be used to guide the choice of experimental conditions for accurate rheological characterizations of highly elastic materials: In order to obtain reliable measurements of the viscometric material functions at high shear rates, it is necessary to conduct experiments in the steady region of the stability diagram. This corresponds to measurements at high $We$, but low $De$, and can be effected by using either very small cone-angles, or large aspect ratios ($R/H$) in the parallel-plate geometry.

5.4.4 Interpretation of Literature Results for the 'M1' Test Fluid

Finally we note that stability diagrams of the form shown in Figures 5.13 and 5.15 can also be used to interpret results obtained recently with the test fluid 'M1'. This fluid is another polyisobutylene-based Boger fluid which has ostensibly been declared a 'standard test fluid' to aid international attempts to measure extensional properties of elastic fluids. The M1 fluid consists of smaller quantities (0.244 wt%) of a higher molecular weight PIB ($\bar{M}_w = 3.8 \times 10^6$ g/mol) dissolved in a 7% kerosene/polybutene solvent, and material properties of the fluid have been published in a special issue of the Journal of Non-Newtonian Fluid Mechanics (vol. 35, 1990). In a series of careful experiments over a range of temperatures, Laun and Hingmann (1990) found that the flow instability in a cone-and-plate geometry with $\theta_0 = 4^\circ$ occurred at a constant critical shear stress of $\tau_c = 135$ Pa. However, their experiments at the same temperature in the same rheometer with a smaller cone angle of $\theta_0 = 2^\circ$ suggest that the instability occurs at the considerably higher critical shear stress of 210 Pa (see Fig. 14 of the Laun and Hingmann manuscript). This apparent
Figure 5.15 Stability diagram for viscometric flow of the Boger fluid in the cone-and-plate geometry with cone angles of $\theta_0 = 0.04$ and 0.10 radians.
contradiction is explained by calculating the angular rotation rate in both experiments: in each case the critical rotation rate is almost unchanged with \( \Omega_c \approx 3.0 \pm 0.2 \text{ rad/s} \). Halving the angle of the cone results in a doubling of the critical shear-rate \( \gamma_c = \Omega_c / \theta_0 \) and thus a doubling of the critical shear stress; therefore, specifying the value of a critical shear stress for onset of the instability does not appear to be a useful criterion.

Similar ideas can explain the observation of Steiert and Wolff (1990) that 'the critical shear rate...does not exactly match the critical value observed for the shear stress (although the instruments and the geometrical dimensions used were different)' Again, calculating the angular velocity in each experiment by using the appropriate geometrical dimensions gives a critical value of \( \Omega_c = 2.63 \text{ rad/s} \) from measurements of a critical stress \( \tau_c = 113 \text{ Pa} \) in a constant stress rheometer, and \( \Omega_c = 2.4 - 3.1 \text{ rad/s} \) from the range of critical shear rates measured in a Weissenberg rheogoniometer. These values of \( \Omega_c \) measured by Steiert and Wolff are self-consistent and also agree well with the numbers calculated above from Laun's data. Identical arguments can be applied to the measurements of Hudson and Ferguson (1990) which give yet another different value of the critical stress \( \tau_c = 180 \text{ Pa} \) in M1 at 20°C. The results of these three investigations are summarized below in Table 5.1:

<table>
<thead>
<tr>
<th>Author</th>
<th>Geometry(1)</th>
<th>Stress ( \tau_c ) [Pa]</th>
<th>( \Omega_c ) [rad/s]</th>
<th>( De_c ) (2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laun &amp; Hingmann</td>
<td>CS ( \theta_0 = 2^* )</td>
<td>210</td>
<td>2.6</td>
<td>1.07</td>
</tr>
<tr>
<td></td>
<td>CS ( \theta_0 = 4^* )</td>
<td>135</td>
<td>3.0</td>
<td>1.23</td>
</tr>
<tr>
<td>Steiert &amp; Wolff</td>
<td>WR ( \theta_0 = 2^* )</td>
<td>210 – 270</td>
<td>2.4 – 3.1</td>
<td>0.98 – 1.27</td>
</tr>
<tr>
<td></td>
<td>CS ( \theta_0 = 4^* )</td>
<td>113</td>
<td>2.63</td>
<td>1.08</td>
</tr>
<tr>
<td>Hudson &amp; Ferguson</td>
<td>WR ( \theta_0 = 1.6^* )</td>
<td>210</td>
<td>2.0</td>
<td>0.82</td>
</tr>
<tr>
<td></td>
<td>CS ( \theta_0 = 2^* )</td>
<td>180</td>
<td>2.1</td>
<td>0.86</td>
</tr>
</tbody>
</table>

Table 5.1 Critical parameters for onset of the rotational instability in cone-and-plate flow of the 'M1' viscoelastic test fluid.

(1) Two geometries were employed; the Constant Stress rheometer (CS) and the Weissenberg Rheogoniometer (WR).
(2) The critical Deborah number \( De_c \) has been calculated using a relaxation time of \( \lambda_1 = 0.41 \text{ s} \) determined from the zero-shear parameters measured by Laun and Hingmann (1990).
Table 5.1 shows clearly that although measurements of a ‘critical stress’ may differ by 100% or more, calculation of a critical Deborah number gives much closer agreement between the sets of data, and the stability criterion $D_e$ will be found to be almost independent of geometrical parameters, shear-rate, and the temperature (assuming the temperature dependence of the relaxation time $\lambda_1(T)$ is known).

The anti-thixotropic behavior of these Boger fluids does not result from a build-up of a ‘transient network formation’ as has been suggested by Sridhar (1990), but arises from a well-defined hydrodynamic transition in which simple viscometric flow becomes unstable with respect to a time-dependent, three-dimensional state consisting of the tangential shear flow and a nonaxisymmetric, secondary flow. If some complex shear-thickening microstructure was created in the fluid, it should be possible to observe it in successive measurements with a particular fluid sample. A series of experiments were performed in which a fluid sample was repeatedly sheared above the critical Deborah number with only 10 seconds between successive measurements. This brief interval should be short enough that any ‘network structure’ remains in the fluid, thus allowing more rapid development of the anti-thixotropic state in subsequent experiments. However, measurements show that in successive runs at the same rotation rate, the initial overshoot and plateau region remain unchanged, whereas the onset of the flow transition takes longer to develop. This increase in the induction time suggests some irreversible molecular degradation occurs and has also been documented by other researchers (Magda and Larson, 1988; Laun and Hingmann, 1990). Gel permeation chromatography (GPC) experiments were performed on fluid samples before and after shearing in the rheometer to determine the molecular weight distribution. However, no statistically significant change in the mass-average molecular weight from the distributions shown in Figure 4.5 could be determined, and any possible degradation in the important high-molecular-weight ‘tail’ was below the small sensitivity and reproducibility errors associated with GPC.

5.5 Summary

The experiments described in this chapter have shown conclusively that an elastically-driven flow transition occurs in the rotational flow of the PIB/PB Boger fluid, and that this instability results in a time-dependent, apparent shear-thickening of the viscosity and first normal stress difference. Measurements of the critical conditions for the onset of the flow transition were made by systematically varying the plate separation in the parallel-plate geometry and demonstrated that this rotational instability is a function of the
angular velocity in the flow and not the shear-rate or shear stress. The disturbance mode initially grows exponentially in time, in agreement with theoretical predictions for an Oldroyd-B model; however, experiments show that the resulting flow is nonaxisymmetric, overstable in time and subcritical in $\gamma$. Spectral analysis also shows that the nonlinear flow which ultimately develops is temporally aperiodic.

The experimentally determined onset point of the flow instability compares moderately well with the predictions of eqs. (2.2) and (2.3); however, the observations of the temporal spectrum and spatial form of the instability are not accurately explained by the normal-mode calculations of Phan-Thien (1983, 1985). His analyses predict a well-defined transition to a new steady-state, axisymmetric motion that corresponds to a single recirculating cell, whereas our observations indicate that the instability leads to a time-dependent, three-dimensional flow consisting of a number of asymmetric cells that propagate radially through the fluid sample. A more accurate stability analysis of this flow requires the incorporation of a number of complex modifications such as disturbances of a more general, three-dimensional form, a more realistic fluid model, the finite nature of the disks, plus the presence of a deformable meniscus. Such an analysis is beyond the capability of current numerical methods.
Chapter 6

Viscoelastic Flow in Axisymmetric Abrupt Constrictions

The flow of viscoelastic liquids through abrupt axisymmetric contractions has been reviewed in detail in Chapter 2.2. Previous experimental studies of the entry flows of highly elastic Boger fluids have demonstrated the onset of unstable flow regimes which appear to be very similar to those documented in commercial extrusion processes. However, these results also indicate that the flow characteristics and stability observed in a particular contraction geometry are a sensitive function of the fluid rheology, the contraction ratio $\beta$, and the shape of the contraction entrance. The aim of the contraction flow experiments described in this Chapter is thus to couple the qualitative video-imaging techniques described in the previous Chapter with highly accurate LDV measurements in order to link the local dynamic behavior of a well-characterized test fluid with the evolution of the vortex structure that is observed macroscopically in axisymmetric contraction flows. The experiments have been performed in six different contraction ratios with two different lip configurations in order to systematically explore the large variations in dynamic behavior associated with small changes in flow geometry.

Simple flow visualization results are presented first in Section 6.1 in order to illustrate the overall structure of the transitions observed throughout the axisymmetric contraction as a function of the Deborah number. Detailed steady-state and time-dependent LDV data are then presented in Section 6.2 to document the sequence of nonlinear transitions that occur in the flow near the contraction lip as the Deborah number is increased. The development of a diverging flow regime and the onset of a second set of dynamics associated with instability of the large elastic vortex are described in Section 6.3. Finally, in Section 6.4 the effects of curving the lip entrance on the dynamics and flow structure observed near the lip of the 4:1 contraction are investigated. A detailed discussion of the experimental results is presented in Section 6.5, together with a few examples illustrating the application of phase-plane techniques to the dynamic behavior of the system.
and the preliminary results of a comparison with a finite-element simulation of viscoelastic entry flow.

The magnitude of elastic and inertial effects in the flow are characterized by the Deborah number and Reynolds number, respectively. General definitions of these dimensionless flow parameters have been provided in eqs (4.46) and (4.47) in terms of the material properties \( \lambda \) and \( \rho \), a characteristic length scale \( L \) and a characteristic velocity \( \langle u \rangle \). For the axisymmetric contraction geometry shown in Figure 2.3, it is conventional to select the downstream tube radius \( R_2 \) and the average velocity \( \langle u_z \rangle_2 \equiv Q / \pi R_2^3 \) in the downstream tube as the characteristic quantities. A characteristic shear rate in the downstream tube is then \( \dot{\gamma}_2 = \langle u_z \rangle_2 / R_2 \). The detailed rheological characterization of the PIB/PB test fluid presented in Chapter 4 has shown that a single constant Maxwellian relaxation time \( \lambda \) is insufficient to accurately describe the viscoelastic behavior of the fluid across a wide range of shear rates. Instead, a shear-thinning relaxation time \( \lambda(\dot{\gamma}) \) is defined in terms of the shear-rate-dependent viscometric properties according to eq. (4.45) and is evaluated at the characteristic shear rate \( \dot{\gamma}_2 \) in the downstream tube. The final definitions of the Deborah number and Reynolds number are thus

\[
De_2 = \frac{\lambda(\dot{\gamma}_2) \langle u_z \rangle_2}{R_2} \tag{6.1}
\]

and

\[
Re_2 = \frac{\rho \langle u_z \rangle_2 R_2}{\eta \dot{\gamma}_2} \tag{6.2}
\]

where the subscript '2' is retained to emphasize that the definitions of \( De_2 \) and \( Re_2 \) are in terms of the downstream tube conditions. These definitions are valid for any viscoelastic fluid in which the shear-rate-dependent viscometric behavior is known; however, equation (6.2) can be simplified further for Boger fluids by replacing \( \eta(\dot{\gamma}_2) \) with \( \eta_0 \), since the viscosity is almost independent of the shear rate.

6.1 Flow Visualization

The macroscopic effects of viscoelasticity on the structure of the velocity field are observed by using the flow visualization procedure described in Chapter 3.2. Images of the velocity field are recorded directly onto videotape and additional long-exposure 'streak photographs' are generated to document the effects of elasticity on the shape of the flow streamlines. Representative photographs are reproduced in this Chapter, and real-time
footage of the dynamic behavior observed in each contraction geometry is contained in the accompanying videotape (Appendix A).

The streamlines observed in a 4:1 contraction at the conditions \( De_2 = 0.90 \) and \( Re_2 = 0.008 \) are shown in Plate 6.1(a). At low Deborah numbers (\( De_2 < 1.0 \)) elastic effects in the flow are negligible compared to viscous effects and the test fluid behaves as a highly viscous Newtonian liquid flowing through an abrupt axisymmetric contraction. The fluid in the upstream tube converges and accelerates directly towards the small tube. A very weak recirculation is observed in the outer corner of the large tube, as predicted by Moffat (1964). The size of this corner vortex is characterized by the dimensionless reattachment length \( \chi = H / 2R_1 \), where \( H \) is the vertical height that the vortex extends upstream and \( R_1 \) is the upstream radius of the tube. Measurements from the time-exposure streak photographs at low \( De_2 \) give a value of \( \chi = 0.17 \) for all contraction ratios, in good agreement with the extensive experiments of Boger et al. (1986) and the numerical simulations of Kim-E et al. (1984) discussed previously in Chapter 2.2.1.

As the flow rate is increased and elastic effects in the flow become important a dramatic change occurs in the shape of the streamlines. The flow field at \( De_2 = 3.40 \) and \( Re_2 = 0.041 \) is shown in Plate 6.1(b). The weak corner vortex observed in Newtonian flow is greatly reduced in size and a separate intense vortex has formed at the re-entrant corner where the upstream tube joins the smaller downstream tube. The formation of this lip vortex is observed to occur in each contraction ratio at a Deborah number \( De_2^{(lip)} \geq 3.0 \). Identical behavior was first documented in another PIB/PB fluid by Boger et al. (1986) and this flow structure is in good agreement with the sketches of Boger (1987) reproduced in Figure 2.9. In addition to the formation of the lip vortex, the flow field shown in Plate 6.1(b) no longer monotonically converges towards the small tube: upstream of the contraction the streamlines near the centerline diverge and fluid flows out towards the stagnant corner of the upstream tube, before rapidly accelerating into the small tube immediately above the contraction plane. This phenomenon has been documented previously in tubular entry flow experiments involving highly shear-thinning viscoelastic fluids at high \( Re_2 \) (as discussed in Chapter 2.2.2), but not in the low Reynolds number flows of Boger fluids.

As the Deborah number is increased the elastic lip vortex increases in size and grows radially outwards towards the wall. Eventually the lip vortex fills the base of the large upstream tube and the flow enters the vortex growth regime in which further increases in \( De \) lead to a rapid increase in the reattachment length \( \chi \) as the elastic vortex expands up the wall of the large tube. The large elastic vortex observed at \( De_2 = 3.92 \) and \( Re_2 = 0.056 \) in the 4:1 contraction is shown in Plate 6.1(c). The reattachment length is
Plate 6.1  Viscoelastic flow through an abrupt 4:1 axisymmetric contraction: (a) low flow rate with Moffat corner vortex, $De_2 = 0.90$, $Re_2 = 0.008$ (exposure time $T = 16$ seconds); (b) development of diverging streamlines and formation of lip vortex; $De_2 = 3.40$, $Re_2 = 0.041$ ($T = 8$ s)
Plate 6.1 (cont.) (c) elastic vortex growth; $De_2 = 3.92$, $Re_2 = 0.056$ ($T = 8$ s).
(d) Coexistence of lip vortex and weak corner eddy in a 6:1 axisymmetric contraction at the same flow conditions as Plate 1(b); $De_2 = 3.40$, $Re_2 = 0.041$ ($T = 12$ s).
determined to be $\chi = 0.21$ and continues to increase with $De_2$ until ultimately the flow becomes visually unstable. At $De_2 = 4.5$ the reattachment length is determined to be $\chi = 0.40$, and the vortex oscillates in size. The vortex shown in Plate 6.1(c) is distinguished from the Newtonian corner vortex shown in Plate 6.1(a) by its size and also by the curvature of the vortex boundary, which is concave for the Moffat eddy at low $De_2$ and becomes convex for the elastic vortex at high Deborah numbers.

The variation of the vortex size $\chi$ (as determined by streak photographs) with Deborah number is shown for each contraction ratio in Figure 6.1. In the vortex growth regime at high $De_2$ (solid symbols) the vortex size is essentially independent of $\beta$; however, at low Deborah numbers the behavior is more complicated. For small contraction ratios ($\beta \leq 5$) the size of the Newtonian corner vortex (hollow symbols) decreases from $\chi = 0.17$ as $De$ increases until it is almost nonexistent when the lip vortex forms at $De_2 = 3.0$. For higher contraction ratios ($\beta = 6$ and $\beta = 8$) the corner vortex is initially the same size with $\chi = 0.17$ for $De_2<<1$; however, it is more effectively spatially isolated from the contraction lip and does not disappear following the formation of the elastic lip vortex. Instead both vortices coexist over a range of Deborah number until the elastic vortex expands outwards from the lip and engulfs the corner vortex. The presence of both vortices is shown in Plate 6.1(d) for the 6:1 contraction at $De_2 = 3.46$. This complex dependence of the vortex size on contraction ratio agrees with the previous investigation of Boger et al. (1986) for PIB/PB Boger fluids. However, it should be noted that similar measurements with PAC/CS Boger fluids have shown that the evolution of the reattachment length $\chi$ with $De_2$ is a sensitive function of the fluid rheology as well as the contraction ratio (Boger, 1987). This variation is summarized in the sketches shown in Figure 2.9. For PAC/CS solutions, the collapse of the Newtonian corner vortex and formation of an independent lip vortex is only observed for $\beta \leq 2$. In higher contraction ratios the coexistence of both a lip and corner vortex, similar to that shown in Plate 6.1(d), is observed. Further discussion of the similarities and differences of PAC/CS and PIB/PB solutions is presented in Section 6.5.

Although macroscopic flow visualization provides a qualitative characterization of the sequence of flow transitions, from a Newtonian corner vortex to an elastic lip vortex and then to vortex growth, a far more detailed picture of the flow transitions is established by relating these global changes to LDV measurements of the local dynamics near the contraction lip and in the bulk of the flow.
Figure 6.1  The vortex reattachment length $\chi (\equiv H_V / 2R_1)$ as a function of Deborah number for each contraction ratio. The hollow symbols correspond to the Newtonian corner vortex and the solid symbols correspond to the elastic vortex which forms near the lip.
6.2 LDV Measurements of Flow Kinematics Near the Lip

6.2.1 Newtonian Flow

The axial and radial velocity components were measured in each contraction ratio at the same volumetric flow rate of \( Q = 0.32 \text{ cm}^3/\text{s} \); corresponding to downstream flow conditions of \( De_2 = 0.42 \) and \( Re_2 = 0.004 \). At this low Deborah number the velocity profiles correspond to a Newtonian flow. The evolution of the axial velocity along the centerline for contraction ratios \( 2 \leq \beta \leq 8 \) is shown in Figure 6.2. The velocity and axial position are nondimensionalized with the downstream average velocity \(<u_2>_2\) and the tube radius \( R_2 \), respectively. Far upstream of the contraction the flow has a fully developed parabolic profile, and the centerline velocity for each contraction ratio is \( u_2 / <u_2>_2 \equiv 2 / \beta^2 \). As the fluid approaches the contraction plane the flow accelerates into the small tube and the data for each contraction ratio superimpose. Downstream of the contraction the flow again assumes a fully developed parabolic profile with centerline velocity \( u_2 / <u_2>_2 \equiv 2 \). Radial profiles of the axial and radial velocity components at \( \zeta = -1.0 \) are shown in Figures 6.3(a) and (b), respectively. The profiles again superimpose when the velocity and position are scaled with the downstream tube conditions, except near the outer walls of the upstream tube. These results indicate that the Newtonian flow near the contraction plane is governed by conditions in the small tube and is relatively independent of the contraction ratio \( \beta \). This scaling has also been documented by Rainford et al. (1989) for axisymmetric contraction flows of a highly shear-thinning solution of 5 wt% polyisobutylene in tetradecane.

6.2.2 The Lip Instability: \( 2 \leq \beta \leq 5 \)

For moderately low Deborah numbers \( (De_2 \leq 1.5) \) the velocity components near the lip remain steady and two-dimensional as the flow rate through the 4:1 contraction is increased. At a critical Deborah number \( De_2^{(osc)} \approx 1.5 \) the flow near the lip undergoes a Hopf bifurcation to a three-dimensional, time-dependent motion and oscillations develop in both the axial and radial velocity components. These velocity fluctuations are small in amplitude and are localized to the lip region upstream of the contraction plane \((-1.5 \leq \xi \leq 1.5, -1.5 \leq \zeta \leq 0)\). LDV measurements further upstream and downstream of the contraction plane remain steady. It is emphasized that the direct flow visualization techniques discussed in Section 6.1 are not able to detect this time-dependent flow; the 'streak' photographs result in a time-averaged picture of the flow field and do not indicate
Figure 6.2  Centerline axial velocity $v_2(0, \zeta)$ in the large upstream tube for contraction ratios $2 \leq \beta \leq 8$, at flow conditions $De_2 = 0.42$, $Re_2 = 0.004$. The velocity profiles are normalized with the average downstream velocity and superimpose near the contraction plane ($-1 < \zeta < 0$).
Figure 6.3  (a) Axial velocity profiles $v_z(x, y, z) - 1$ and (b) radial velocity profiles $v_r(x, y, z)$ above the contraction plane at a low Deborah number for contraction ratios of $\beta = 2, 3, 4$ and 8. The profiles superimpose when scaled with the downstream parameters $R_2$ and $\langle v_z \rangle_2$. 

242
any change to the overall flow pattern, whilst the amplitude of oscillation is too small to be readily detected by straightforward visual examination of the videotape. These time-periodic oscillations are followed by using the three frequency trackers discussed in Chapter 3.1.1, and sample time series for the axial, radial and tangential velocity components near the lip are shown in Figures 6.4 and 6.5. The time-dependent tangential velocity component \( v_\theta \) that develops is observed to oscillate about a zero mean, as first observed by Lawler et al. (1986). The frequency of oscillation in each component is calculated by performing a Fast Fourier Transform (FFT) of the velocity data. The initial frequency of oscillation for each velocity component in the 4:1 contraction is determined from the FFTs in Figures 6.4(b) and (c) to be \( f_1 = 0.0950 \) Hz. As the Deborah number is increased these velocity fluctuations grow in amplitude, the frequency of oscillation increases, and harmonics of the fundamental frequency appear in the FFT spectrum, as shown in Figures 6.5(b) and (c).

The variation of oscillation frequency determined from FFT spectra, such as those in Figures 6.4 and 6.5, is summarized in Figure 6.6 as a function of Deborah number for \( \beta = 4 \). In addition, the square of the oscillation amplitude in \( v_\theta \) near the lip as a function of the Deborah number is shown in Figure 6.7. The critical Deborah number for onset of periodic flow is determined accurately by fitting these data to the results of an asymptotic analysis for a supercritical Hopf bifurcation (Iooss and Joseph, 1980) which predicts that the amplitude of oscillation should be of the form

\[
|v(r, \theta, z, t)| \propto \sqrt{(De - De^{(osc)})} \ e^{i\omega t} \tag{6.3}
\]

Near the onset point the data are linear in agreement with equation (6.3), and extrapolation to \( |v_\theta| = 0 \) determines the critical value of \( De \) to be \( De^{(osc)} = 1.50 \pm 0.02 \). The large nonlinear increase in amplitude at higher Deborah numbers results from the introduction of harmonics of the fundamental oscillation frequency.

A similar Hopf bifurcation in a 4:1 axisymmetric contraction was first observed by Muller (1986, Lawler et al., 1986) for a less elastic PIB/PB/K Boger fluid. In sharp contrast to the results described here, Muller observed the flow to return to a two-dimensional steady state as the Deborah number was increased. With the PIB/PB/C14 fluid used here the flow remains time-dependent for all Deborah numbers greater than \( De^{(osc)} \) and subsequently undergoes a series of nonlinear transitions.

Experiments in other contraction ratios indicate that the local dynamics of the flow transition are highly sensitive to the contraction ratio. The onset of time-periodic flows was
Figure 6.4  (a) Sample time series data showing oscillations of all three velocity components near the lip ($\xi = -1.23$, $\zeta = -0.32$) of the 4:1 contraction at $De_2 = 1.70$. (b) Frequency spectrum of the axial velocity component in (a); the frequency of oscillation is $f_1 = 0.0950$ Hz. (c) Frequency spectrum of the radial velocity component in (a); the frequency of oscillation is $f_1 = 0.0950$ Hz.
Figure 6.5  (a) Sample time series data showing increased amplitude of oscillations in all three velocity components near the lip ($\xi = -1.23, \zeta = -0.32$) of the 4:1 contraction at $De_2 = 2.12$. (b) FFT frequency spectrum of the axial velocity component in (a); the frequency of oscillation is $f_1 = 0.1275 \text{ Hz}$. (c) FFT spectrum of the tangential velocity component in part (a); the frequency of oscillation is $f_1 = 0.1275 \text{ Hz}$. 

\[ 
\begin{align*}
\beta &= 4 \\
De_2 &= 2.12 \\
Re_2 &= 0.016 \\
\end{align*} 
\]
Figure 6.6 Frequency of oscillations in the axial velocity near the lip of the 4:1 contraction ($\xi = 1.15, \zeta = -0.30$) as a function of the Deborah number.
Figure 6.7  Square of the amplitude of oscillations in $v_z$ near the lip of the 4:1 contraction ($\xi = 1.15$, $\zeta = -0.30$) as a function of the Deborah number.
detected for contraction ratios $2 \leq \beta \leq 5$. The flow conditions and critical Deborah number $De_2^{(osc)}$ at onset of oscillations are listed in Table 6.1.

<table>
<thead>
<tr>
<th>$\beta$</th>
<th>$Q$ [cm$^3$/s]</th>
<th>$\gamma_2$ [s$^{-1}$]</th>
<th>$\lambda (\dot{\gamma}_2)$ [s]</th>
<th>$De_2$</th>
<th>$Re_2$</th>
<th>$\tau_w$ [kN/m$^2$]</th>
<th>$N_{1w}$</th>
<th>$N_{1w}/\tau_w$</th>
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<td>0.759</td>
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Table 6.1 Flow parameters at the critical Deborah number $De_2^{(osc)}$ required for onset of time-dependent flow near the lip of the axisymmetric contraction.

Notes
(i) The subscript ‘2’ denotes rheological properties evaluated at the average flow conditions in the downstream tube. The subscript ‘w’ indicates the rheological properties are calculated at the shear rate of the downstream wall where $\dot{\gamma}_w = 4\gamma_2$.

(ii) The stress ratio is defined as $N_{1w}/\tau_w = \Psi_1 (\dot{\gamma}_w)/\eta(\dot{\gamma}_w)$ and is evaluated at the shear-rate of the downstream wall.

The results presented in Table 6.1 demonstrate that the critical value of $De_2^{(osc)}$ displays a minimum at $\beta = 4$ and increases for both larger and smaller contraction ratios. The frequencies of oscillation near the lip for $\beta = 3$ and $\beta = 5$ are shown in Figure 6.8. The critical Deborah numbers of the Hopf bifurcation are determined from the plots of the square of the oscillation amplitude shown in Figure 6.9 to be $De_2^{(osc)} = 1.71$ and 2.20 for $\beta = 3$ and 5, respectively. Again the oscillations developed in all three velocity components and remained localized to the lip region. The flow appeared steady away from the lip with no visual changes to the global structure. Experiments in the 2:1 contraction showed a similar transition to time-periodic flow at $De_2 = 2.04$. However, no Hopf bifurcation was detected in experiments with larger contraction ratios of $\beta = 6$ and $\beta = 8$, up to a critical Deborah number where the large-scale vortex dynamics described in Section 6.3.2 become dominant.
Figure 6.8  Frequency of oscillations in the radial velocity near the lip as a function of the Deborah number for $\beta = 3$ and $5$. 
Figure 6.9  Square of the amplitude of oscillations in $v_r$ near the lip as a function of the Deborah number for $\beta = 3$ and 5.
6.2.3 Period Doubling Transition: $3 \leq \beta \leq 5$

As the Deborah number is increased the amplitude of the oscillations in the velocity components grows, and higher harmonics of the oscillation frequencies $2f_1$, $3f_1$ appear in the frequency spectrum. A subharmonic period-doubling instability is observed in the 4:1 contraction at $De_2 = 2.60$. This transition results in a large increase in the oscillation amplitude and a spectral peak at $f_1/2$ which grows rapidly in strength to dominate the frequency spectrum. These effects are demonstrated by comparing the two sets of time-series and frequency spectra presented in Figure 6.10 for the radial velocity component near the lip ($\xi = 1.68$, $\zeta = -0.30$) of the 4:1 contraction. The frequency of oscillation shown in Figure 6.10(b) before the transition is $f_1 = 0.1275$ Hz, whereas the dominant frequency following the period-doubling transition in Figure 6.10(d) is 0.065 Hz. A similar period-doubling transition is also observed for $\beta = 3$ and $\beta = 5$ at Deborah numbers $De_2 > 2.5$. Further increases in $De_2$ however did not result in subsequent period-doubling transitions and the appearance of $f_1/4$ peaks in the frequency spectrum. Instead, another Hopf bifurcation is observed in the flow at $De_2 \equiv 3.0$ resulting in a quasiperiodic flow state.

6.2.4 Quasiperiodic Flow Near The Lip: $2 \leq \beta \leq 5$

Quasiperiodic flow regimes characterized by two independent frequencies were observed in the contraction ratios $2 \leq \beta \leq 5$ for $De_2 > 3.0$. The radial velocity profile shown in Figure 6.11(a) for flow in a 5:1 contraction contains large nonlinear oscillations as a function of time. Analysis of the frequency spectrum shown in Figure 6.11(b) reveals that the time-dependent flow is comprised of two distinct oscillation frequencies ($f_1$, $f_2$) and a number of other peaks which can be identified as linear combinations of the form $m_1f_1 + m_2f_2$, where $m_1$ and $m_2$ are integers. These two frequencies in the time-dependent flow are described by bifurcation from a limit cycle to a torus (Iooss and Joseph, 1980). The two fundamental frequencies are determined from Figure 6.11(b) to be $f_1 = 0.043$ Hz and $f_2 = 0.063$ Hz within the limits of spectral resolution attainable with the experimental apparatus. This complex series of periodic, period-doubling and quasiperiodic flow transitions can best be represented in the form of an experimentally determined bifurcation diagram, and the evolution of the dominant frequencies in the 3:1 contraction from $De_2 = 1.0 - 3.5$ is summarized in Figure 6.12.
Figure 6.10 Time series data showing period-doubling of the oscillations in the radial velocity component near the lip \((\xi = 1.68, \zeta = -0.30)\) of the 4:1 contraction: The time series \((a)\) at \(De_2 = 2.55\) has small amplitude oscillations, and the fundamental frequency \((b)\) before period-doubling is \(f_1 = 0.1275\) Hz. After the period-doubling transition the time series \((c)\) at \(De_2 = 2.62\) shows larger oscillations and the FFT spectrum \((d)\) contains additional spectral peaks at \(f_1/2\) and \(3f_1/2\).
Figure 6.11 Time series and frequency spectrum showing quasiperiodic oscillations in the velocity near the lip of the 5:1 contraction ($\xi = -1.50, \zeta = -0.30$) at $De_2 = 3.02$: (a) radial velocity profile as a function of time; (b) FFT spectrum.
Figure 6.12 Dominant frequencies in the time-dependent regimes near the lip of the 3:1 contraction as a function of the Deborah number. In the quasiperiodic regime the three strongest peaks in the FFT spectrum are identified as; (●) $f_1$, (○) $f_2$, (Θ) $f_1 + f_2$. 
The development of a quasiperiodic state from a time-periodic regime was approximately independent of contraction ratio for $2 \leq \beta \leq 5$ and occurred in each contraction ratio at a Deborah number $De_2^{(lip)} \approx 3.0$. At this Deborah number the flow visualization results presented in Plate 6.1(b) show a distinct change in the flow field and the formation of an intense lip vortex in the same spatial region where quasiperiodic time-dependence is observed. Analysis of the relevant section of the attached videotape (Appendix A) also shows that this vortex oscillates in size, even though the volumetric flow rate through the contraction is constant. LDV results in the higher contraction ratios $\beta = 6$ and $\beta = 8$ indicate that the flow is not time-dependent at $De_2 = 3.0$; however the formation of a steady lip vortex is still observed in streak photographs such as Plate 6.1(d) and can be measured quantitatively by using the LDV system. Figure 6.13 shows the radial and axial velocity components in the 6:1 contraction near the lip ($\xi = -1.50$, $\zeta = -0.30$) as a function of Deborah number. Initially, the magnitude of both velocity components increase and the radial velocity $v_r$ is negative indicating inward flow towards the throat of the contraction. At a Deborah number of $De_2 = 2.96$ the axial velocity decreases, and the radial velocity reverses in direction as the lip vortex forms and the fluid near the contraction lip flows outwards into the corners of the large tube.

The complex dependence of the steady and time-dependent flow dynamics on contraction ratio is represented graphically by the stability diagram shown in Figure 6.14. The critical Deborah numbers for development of time-periodic flow near the lip, $De_2^{(osc)}$, and for the formation of a lip vortex, $De_2^{(lip)}$, are shown as functions of contraction ratio. These flow transitions are considered to be two separate, competing modes of elastic phenomena. For moderate contraction ratios, $2 \leq \beta \leq 5$, the most unstable mode, i.e. the instability occurring at lowest $De_2$, leads to a time-periodic flow regime near the lip, which is followed by development of a quasiperiodic lip vortex. However, for the larger contraction ratios $\beta = 6$ and $\beta = 8$, the formation of a steady lip vortex (which coexists with the corner vortex) will occur first.

Increasing the Deborah number beyond $De_2^{(lip)}$ resulted in growth of the lip vortex vertically upstream and radially outwards until at $De_2 = 3.8$ the recirculation in the 4:1 contraction covered the base of the large upstream tube. The increasing size of the lip vortex was accompanied by an expansion in the spatial extent of the time-dependent flow and an increase in the amplitude of the velocity oscillations. Sample time series of the axial velocity on the centerline near the lip of the 4:1 contraction are shown in Figure 6.15 as the Deborah number was increased. At low $De_2$ the flow field is Newtonian, and the velocity is steady throughout the upstream and downstream tubes. At $De_2 = 3.10$ a small time-dependent lip vortex has formed near the lip, and velocity data measured within the lip
Figure 6.13 Steady-state radial and axial velocity profiles as functions of Deborah number near the lip of the 6:1 contraction ($\xi = -1.50$, $\zeta = -0.30$). The formation of a lip vortex at $De_2 = 2.96$ is clearly identified by the reversal in flow direction of the radial velocity component.
Figure 6.14 Flow transitions in the axisymmetric abrupt contraction as functions of the contraction ratio for $2 \leq \beta \leq 8$: (●) The critical Deborah number $De_2^{(osc)}$ for Hopf bifurcation to a time-dependent flow regime near the lip; (○) the critical Deborah number $De_2^{(lip)}$ for the formation of a lip vortex. At higher Deborah numbers the large elastic vortex present in the upstream tube becomes unstable to either a pulsing (▲) or rotating (△) flow.
Figure 6.15 (a) Time series of the axial velocity on the centerline ($\xi = 0.00$, $\zeta = -1.50$) of the 4:1 contraction at $De_2 = 0.4, 3.1$ and 3.7; (b) FFT spectrum of the time-dependent axial velocity shown in (a) at $De_2 = 3.7$. 

258
vortex indicate a quasiperiodic flow, as typified by Figure 6.12(a). The axial velocity on the centerline near the lip ($\zeta = -1.50$) now contains small fluctuations about its average value; however, an FFT of the time series at $De_2 = 3.10$ shown in Figure 6.15(a) revealed no dominant frequency of oscillation. At $De_2 = 3.70$ the lip vortex has grown substantially in size ($\chi = 0.12$) and almost extends across the entire width of the upstream tube. Oscillations in the velocity field now extend spatially throughout the throat region and the axial velocity on the centerline shows large amplitude, time-dependent fluctuations. LDV measurements in the lip region at this Deborah number show quasiperiodic behavior; however, farther from the reentrant corner the velocity field is affected predominantly by the large amplitude fluctuations in the size of the lip vortex and velocity measurements on the centerline indicate essentially a time-periodic behavior as shown in Figure 6.15(a). The dominant frequency of oscillation was determined from the Fourier spectrum in Figure 6.15(b) to be $f = 0.1875$ Hz. Farther upstream from the lip the flow remains steady, and LDV measurements at $\zeta = -5.0$ and $\zeta = -10.0$ showed no time-dependent behavior.

6.2.5 Aperiodic Flow: $3 \leq \beta \leq 5$

Upon reaching the corners of the upstream tube, the lip vortex grew rapidly to form the large convex elastic vortex shown in Plate 6.1(c) and the time-dependent nature of the flow underwent a third transition. Experimental time series no longer exhibited clear quasiperiodic oscillations; instead, aperiodic fluctuations in the velocity components were observed with no well resolved spectral peaks. This behavior is demonstrated in Figure 6.16(a) by the axial velocity on the centerline at $\zeta = -2.25$ in the 4:1 contraction. The Deborah number is $De_2 = 4.08$, and the lip vortex has expanded upstream to a height of $\chi = 0.21$. The time series shows both slow and rapid fluctuations in magnitude. The FFT spectrum in Figure 6.16(b) contains broad-band noise across the spectrum with no dominant oscillation frequency. By replotting the data on a logarithmic scale, the level of noise measured in this frequency spectrum is shown to be significantly increased above the instrumental 'white' noise that is observed in the FFT spectra previously presented in the periodic and quasiperiodic flow regimes, particularly in the very low ($< 0.2$ Hz) frequency range. The signal-to-noise resolution of the LDV photomultipliers and the limited time-window of observation possible with this open-flow system prevent the extremely long data-acquisition times necessary to quantify a chaotic flow regime in detail (Benson and Gollub, 1980; Bergé et al., 1986). However, it is clear that upon entering the vortex
Figure 6.16  Aperiodic flow along the centerline near the lip ($\xi = 0.00$, $\zeta = -2.25$) of the 4:1 contraction at $De_2 = 4.08$ and $Re_2 = 0.061$. The axial velocity (a) shows random fluctuations and the FFT (b) has no fundamental frequency of oscillation, but contains broad-band noise across the entire spectrum.
growth regime the flow dynamics become more complex than the quasiperiodic flow observed at lower values of the Deborah number. These velocity fluctuations near the lip persisted as the Deborah number was increased and as the elastic vortex increased in size. Similar FFT spectra were observed for $\beta = 3$ and $\beta = 5$; however, no data could be obtained in the 2:1 contraction since the frequency trackers were unable to follow the rapidly varying LDV signal. In the large contraction ratios ($\beta = 6$ and $\beta = 8$) the flow near the lip remained steady as the Deborah number was increased, until the onset of the global vortex instability described in Section 6.3.2.

Two previous experimental investigations of flow through 4:1 contractions have used similar PIB/PB Boger fluids: Lawler et al. (1986) observed that the time-dependent flow near the lip reverted to steady two-dimensional flow at a second critical Deborah number, and Boger (1987) reported that the time-dependent lip vortex observed qualitatively at low $De$ was followed by a steady two-dimensional vortex growth regime. However, in our experiments the flow in the low contraction ratios ($\beta \leq 5$) remains time-dependent for all $De_2$ above the critical Deborah number, $De_2^{(osc)}$. The reason for this difference is still unclear but it must be related to differences in the rheology of the test fluids. The aperiodic oscillations in the velocity occur over the same range of values of $\beta$ and $De_2$ for which diverging streamlines upstream of the contraction are observed (see Plate 6.1(c)). This diverging flow regime is documented in more detail in Section 6.3.1 and did not occur in the previous investigations with PIB/PB fluids. These velocity fluctuations may thus result from a time-dependent instability connected with the development of a diverging flow structure upstream of the contraction plane.

6.3 Global Kinematic Changes

In addition to the development of localized time-dependent flow near the contraction lip, the flow visualization results described in Section 6.1 have shown that increasing the Deborah number results in modifications of the flow structure throughout the axisymmetric contraction. These phenomena are documented in more detail in this Section.

6.3.1 Diverging Flow Regime: $2 \leq \beta \leq 5$

At high Deborah numbers the flow upstream of the contraction plane becomes divergent and the fluid upstream of the contraction plane moves radially outwards away from the centerline; this phenomenon is demonstrated in Plates 6.1(b) and (c). An increase
in the velocity away from the centerline must result in a reduced axial velocity along
the centerline to conserve mass flow across each plane throughout the upstream tube. Time-
averaged LDV measurements of the axial velocity component along the centerline in the
upstream tube are shown in Figure 6.17 for $\beta = 3, 4$ and 5. Far upstream the velocity
profile remains parabolic and the normalized centerline axial velocity is $\langle u_2 \rangle_2 = 2 / \beta^2$, in
agreement with Figure 6.2. As the fluid approaches the contraction throat it does not
accelerate monotonically but initially decelerates and the velocity reaches a local minimum at
$\zeta = -2.0$. Closer to the contraction plane the velocity increases rapidly. Transverse profiles
of the axial velocity at $\zeta = -1.5$ for contraction ratios $\beta = 3, 4$ and 5 are shown in
Figure 6.18. The profiles are no longer parabolic but show maxima away from the
centerline. Subsequent increases in the Deborah number do not diminish this divergent
effect but result in an increase in the size of the elastic vortex which exists at these flow
rates and an associated shift upstream in the position of the velocity minimum as shown in
Plate 6.1(b) and (c).

It is seen from Figure 6.17 that the magnitude of the deceleration associated with
the diverging flow is strongly dependent on the contraction ratio with the largest decreases
in velocity occurring in the smaller contraction ratios. No velocity minima were observed at
any experimentally attainable Deborah number in the larger contraction ratios ($\beta = 6$ and
$\beta = 8$), and the flow accelerated monotonically towards the throat. Although this
viscoelastic phenomenon first develops at Deborah numbers comparable to those at which
the lip vortex forms it does not appear that the two effects are directly related, since the
diverging flow persists into the vortex growth regime (see Plate 6.1(c)). In addition, lip
vortices have been reported in Boger fluids with no associated diverging flow (Binnington
et al., 1986), whereas a diverging flow regime has been observed in shear-thinning fluids
with no lip vortex formation (Cable and Boger, 1978a; Evans and Walters, 1989). This is
the first time that this phenomenon has been documented in axisymmetric contraction flow
of a Boger fluid, although diverging streamlines have been observed previously by Binding
and Walters (1988) in the flow of a PAC/CS Boger fluid through a 4:1 planar contraction.

The sensitive dependence of the diverging flow on contraction ratio indicates that
flow conditions in the upstream tube play an important role in the sequence of viscoelastic
flow transitions. In steady simple shearing flows a gradual shear-thinning is observed in
viscometric properties such as $\eta$ and $\Psi_1$. However in uniaxial extensional flows, such as
that along the centerline of an axisymmetric contraction, Boger fluids extensionally thicken
and exhibit an elongational viscosity much greater than the viscosity measured in steady
shear flow (see the calculations in Chapter 4.4.4). The data for each contraction ratio
shown in Figure 6.17 show that the maximum extension rate experienced along the
Figure 6.17 Diverging flow upstream of the contraction plane at high $De_2$ demonstrated by the normalized axial velocity $v_z(0, \zeta) / \langle v_z \rangle_2$ along the centerline for contraction ratios $\beta = 3, 4$, and 5.
Figure 6.18 Transverse axial velocity profiles $v_z(\xi) / \langle v_z \rangle_2$ at $\xi = -3.00$ for contraction ratios $\beta = 3, 4$ and 5 at the same $De_2$ as shown in Figure 6.17. The profiles show pronounced off-centerline maxima which vary with contraction ratio.
centerline, \( \dot{\varepsilon} = \partial u_z / \partial z \), is almost the same for \( \beta = 3, 4 \) and 5. Thus the same degree of extensional thickening is expected in each contraction ratio, and the extensional viscosity behavior of Boger fluids alone cannot be used to directly differentiate between the results in each contraction. However, for the same downstream flow conditions \( De_2 \) (and hence the same \( \gamma_2 \)), the average upstream shear rate \( \dot{\gamma}_1 \) is given by \( \dot{\gamma}_1 / \beta^3 \), and therefore differs greatly in each contraction. In addition, the shear rate varies linearly across the upstream tube and reaches a maximum \( \dot{\gamma}_{1w} = 4\dot{\gamma}_1 \) at the wall. The maximum shear rate in the upstream tube and the greatest shear-thinning in the fluid properties is therefore experienced near the wall of the 3:1 contraction. In Section 6.5 we suggest that this variation in the upstream shear rate and the dependence of the total Hencky strain on \( \beta \) may be important in explaining the appearance of the diverging flow field.

6.3.2 Elastic Vortex Instability: \( 2 \leq \beta \leq 6 \)

In addition to the local time-dependent flow dynamics observed near the lip, a separate, macroscopic flow instability is observed at high Deborah numbers. As the flow rate is increased the elastic vortex continues to grow upstream and the vortex size is found to correlate well with the downstream Deborah number as shown in Figure 6.1. At a Deborah number of \( De_2 = 4.4 \) the reattachment length in the 4:1 contraction has reached a maximum steady value of \( \chi = 0.53 \). Any further increase in \( De_2 \) results in the development of an unsteady vortex which remains symmetric but pulsates periodically in size. At \( De_2 = 4.5 \) the reattachment length of this vortex gradually increases with time to a value of \( \chi = 0.73 \) and then rapidly collapses to a much smaller less intense vortex with \( \chi = 0.48 \) before increasing in size again. This periodic pulsating of the vortex is accompanied by highly nonlinear oscillations in the axial velocity, as shown in Figure 6.19(a). The increasing size of the secondary recirculation in the corners of the upstream tube reduces the cross-sectional area for the primary flow through the contraction, and the axial velocity along the centerline therefore increases. When the elastic vortex stops growing in each cycle the axial velocity reaches a maximum and then decreases rapidly as the elastic vortex collapses. The FFT shown in Figure 6.19(b) reveals the relatively strong contributions of the first and second harmonics to the fundamental frequency of oscillation \( f_{\text{pulse}} = 0.0375 \) Hz. These oscillations in vortex size and axial velocity have a much lower frequency than the local dynamics observed near the lip in Section 6.2.1, and the period of oscillation \( T_{\text{pulse}} = 1/f_{\text{pulse}} \) is much longer than the characteristic relaxation times listed in Table 4.2 for this fluid. This instability thus does not appear to be simply
Figure 6.19 Time series and FFT spectra demonstrating different modes of oscillation for the large elastic vortex:

(a) Highly nonlinear large amplitude oscillation in the axial velocity at $D_2 = 4.49$ on the centerline $\xi = 0.0$, $f_{\text{pulse}} = 0.0375$ Hz; (b) FFT spectrum of the time series in (a) gives the frequency of oscillation as $f_{\text{rot}} = 0.1150$ Hz; (c) time series of the axial velocity on the centerline $\xi = -1.75$ of the 5.1 contraction; (d) FFT spectrum of the time series in (c) gives the frequency of oscillation as $f_{\text{rot}} = 0.1150$ Hz.
connected to either the lip kinematics observed at moderate $De_2$ or to the specific relaxation time-scales of the macromolecules in the fluid. LDV measurements in each contraction ratio show that the dynamics of this large vortex instability are highly sensitive to $\beta$; and this flow transition at high $De_2$ appears to be a separate dynamical mode of instability associated with the geometric size of the large upstream tube. The flow conditions at onset of oscillation are summarized below in Table 6.2.

<table>
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<th>$Q$ [cm$^3$/s]</th>
<th>$\gamma_2$ [s$^{-1}$]</th>
<th>$De_2$</th>
<th>$Re_2$</th>
<th>$\tau_w$ [kPa]</th>
<th>$N_{1w}$ [kPa]</th>
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<td>rotating</td>
</tr>
</tbody>
</table>

Table 6.2 Flow conditions at the onset of unsteady oscillations in the large upstream vortex. The oscillation mode observed in each contraction ratio is indicated in the last column. The subscript definitions are as given in Table 6.1.

In the 3:1 contraction a slow pulsing in the vortex size, comparable to that described above, is observed for $De_2 \geq 4.3$; however, the amplitude of the oscillations is smaller and the reattachment length varies between $\chi = 0.51 - 0.60$. The pulsing frequency is $f_{puls} = 0.055$ Hz. A similar weak pulsing instability is also observed for $\beta = 2$. In the larger contraction ratios, $\beta = 5$ and $\beta = 6$, a distinctly different mode of instability is observed. The large vortex becomes asymmetric with a reattachment length that varies with the angular coordinate $\theta$ around the large tube. The vortex does not pulsate and the asymmetric form of the reattachment length remains approximately constant in time; however, the vortex begins to rotate rapidly around the upstream tube. When a longitudinal cross-section is viewed by using the techniques described in Section 3.2, the two sections of the corner vortex on either side of the primary flow through the contraction alternately expand and contract in size as the position of maximum vortex size rotates around the upstream tube. This rotating flow regime can be clearly observed on the videotape (Appendix A) and has also been clearly documented in the photographs of Nguyẻn and
Boger (1979) shown in Figure 2.7. Close inspection of the vortex dynamics on videotape reveals that the actual fluid particles within the vortex have no tangential velocity and that the secondary flow in the recirculating vortex remains two-dimensional. The variation in the vortex height arises from an asymmetry that develops in the streamlines of the converging primary flow and which then precesses azimuthally around the upstream tube. This azimuthal rotation results in periodic oscillations in the axial velocity along the centerline; a typical time series is shown in Figure 6.19(c) for flow in a 5:1 contraction at $De_2 = 4.33$. Comparing Figures 6.19(a) and (c), it is clear that the dynamics in the rotating and pulsing flow regimes are quite different with much faster oscillations observed in the 5:1 contraction. The fundamental frequency for azimuthal rotation was measured as $f_{rot} = 0.1150$ Hz by taking the lowest frequency peak in the FFT spectrum shown in Figure 6.19(d); however, the first harmonic peak ($2f_{rot}$) is of comparable intensity. The oscillation frequencies for both the pulsing and rotating flow regimes increased slowly with increasing Deborah number. With the current flow loop configuration, the maximum flow rate attainable gave $De_2 = 4.7$ and only a few data points could be obtained in each of the time-dependent regimes. No time-dependent vortex dynamics were observed in the 8:1 contraction up to $De_2 = 4.70$; however, we believe that at higher flow rates qualitatively similar behavior would be observed.

The oscillations in the flow associated with variations of the vortex size are not confined to the spatial region near the contraction plane, but also extend far up and downstream. The axial velocity on the centerline at a distance $25R_2$ upstream is shown in Figure 6.20(a) for $\beta = 4$. Large amplitude fluctuations in the velocity are observed and the frequency of oscillation shown in Figure 6.20(b) is the same as the pulsing frequency $f_{pulse}$ observed near the throat of the contraction. The azimuthal rotation of the vortex that occurs for $\beta = 5$ and 6 results in less disturbance to the flow far upstream. Figure 6.20(c) shows the lower amplitude irregular disturbances observed in the centerline velocity $20R_2$ upstream of the contraction plane for $\beta = 5$ at the same flow conditions as Figure 6.20(c). The FFT of this time series is shown in Figure 6.20(d) and identifies some low frequency components, but no clear fundamental oscillation frequency.

6.4 The Effect of Lip Curvature on Flow Dynamics

In previous qualitative investigations of viscoelastic flow through contraction geometries the precise shape of the re-entrant corner at the contraction lip was found to have a pronounced effect on the flow characteristics. Rounding the corner resulted in a
Figure 6.20 Time-dependent velocity profiles far upstream of the contraction plane: (a) Centerline axial velocity at $\zeta = -25$ in the 4:1 contraction; (b) FFT of the time series in part (a) gives $f_{\text{pulse}} = 0.0350$ Hz; (c) the centerline axial velocity far upstream at $\zeta = -20$ in the 5:1 contraction; (d) an FFT of the data in part (c) shows no dominant frequency of oscillation.
reduction in the size of the large corner vortex observed in the axisymmetric contraction flow of Boger fluids (Walters and Webster, 1982) and was reported to increase the stability and reduce the size of the vortex in planar contractions (Evans and Walters, 1986; 1989). This effect is clearly demonstrated by the streak photographs of Evans and Walters presented in Chapter 2.2.2. A smoothly curved lip entrance was machined with a fixed radius of curvature, $R$, to investigate the effect of removing the sharp reentrant corner on the local flow dynamics near the lip. The design of the modified entrance region is shown in Figure 3.8(b). The dimensionless radius of curvature of the curved lip is $R/R_2 = 0.5$, and is the same as that employed by Walters and Webster. However, to prevent the introduction of asymmetry into the flow and the associated distortion of the global flow field described by Evans and Walters, the entire circumference of the re-entrant corner was smoothed instead of only a 180° arc. The results presented here demonstrate that the flow transitions observed in an abrupt contraction are still present for flow through a contraction with a rounded lip, but that the critical values of Deborah number for each transition are increased because of the less severe flow conditions near the reentrant corner of the smooth geometry.

Rounding the lip corner is found to have little effect on the flow characteristics at low $De_2$. A close-up of the flow patterns near the curved lip in a 4:1 contraction at $De_2 = 0.56$ is shown in Plate 6.2(a). The flow converges radially towards the contraction mouth and a weak recirculating eddy, similar to that observed with the sharp lip in Plate 6.1(a), is observed in the stagnant outer corner. The dimensionless reattachment length for the corner vortex is measured as $\chi = 0.17$. Axial and radial LDV scans are not presented here because the data superpose with the results shown in Figures 6.2 and 6.3, except for the region extremely close to the smoothed corner ($-1.5 < \xi < 1.5, -0.3 < \zeta < 0$).

As the Deborah number is increased the consequences of smoothing the lip corner become more pronounced. The variation in vortex size ($\chi$) with $De_2$ is compared in Figure 6.21 for flow through the 4:1 contraction with a sharp lip ($\beta = 4$) and with the curved lip entrance ($\beta = 4c$). At low Deborah numbers the reattachment length of the weak corner vortex for both lip geometries is $\chi = 0.17$. In the sharp lip geometry the corner eddy gradually decreases in size, and a separate lip vortex forms at $De_2^{(lip)} = 3.0$ which grows rapidly in size and engulfs the diminished corner vortex. The vortex then expands into the upstream tube and ultimately begins to pulse in size at $De_2 = 4.4$ as described above in Sections 6.2 - 6.3.

Similar trends are exhibited in the curved lip geometry; however removing the lip singularity shifts these transitions to higher $De_2$. The corner vortex decreases in size very
Plate 6.2  Viscoelastic flow through a 4:1 contraction with a smoothly curved entrance lip: (a) detailed view of the weak Moffat eddy; $De_2 = 0.56$, $Re_2 = 0.005$ (exposure time $T = 128$ s); (b) reduced corner vortex size; $De_2 = 3.24$, $Re_2 = 0.031$ ($T = 64$ s)
Plate 6.2 (cont.)  (c) formation of a highly unsteady, time-dependent vortex near the lip entrance; $De_2 = 4.65$, $Re_2 = 0.091 \ (T = 2 \ s)$; (d) elastic vortex growth regime; $De_2 = 5.32$, $Re_2 = 0.137 \ (T = 4 \ s)$. 
Figure 6.21 Comparison of the dimensionless vortex reattachment length ($\chi$) for flow through a 4:1 contraction with a sharp lip ($\beta = 4$) and with a smoothly curved lip with radius of curvature $R = 0.5R_2$ ($\beta = 4_c$). The hollow symbols ($\square, \bigcirc$) correspond to the shrinking corner vortex, and the solid symbols ($\blacksquare, \bullet$) represent the elastic vortex which develops near the lip edge. The arrows connecting data points at high $De_2$ indicate that the large vortex has become unstable and pulsates in size.
gradually and effectively disappears by $De_2 = 4.5$ when a clearly distinct lip vortex forms. The flow field at $De_2 = 3.24$ is shown in Plate 6.2(b). The weak eddy in the outer corner has greatly decreased in size, no lip vortex can be distinguished, and fluid flows smoothly around the curved lip into the downstream tube. LDV measurements also show that the flow remains steady and axisymmetric. The first flow transition observed in the curved lip geometry was the formation of a small, highly unsteady vortex near the smoothed entrance to the small tube at $De_2 = 4.50$. This time-dependent vortex expanded outwards across the base of the upstream tube and is shown in Plate 6.2(c) at $De_2 = 4.65$. The lip vortex oscillated rapidly in size and was difficult to resolve clearly with the flow visualization procedure. Nonetheless, Plate 6.2(c) is included as the first graphic evidence of the formation of an elastic lip vortex in a contraction geometry with a smoothed lip entrance. Detailed analysis of transient responses on the videotape in Appendix A confirms unequivocally that this vortex originates at the lip entrance and not in the stagnant outer corners of the large tube. The oscillations in the size of the lip vortex are accompanied by rapid periodic fluctuations in the axial and radial velocity components near the lip, as shown in Figure 6.22(a). Both velocity components oscillate with the same frequency which was determined from the FFT data in Figure 6.22(b) to be $f_{\text{curve}} = 0.390$ Hz. This frequency is much higher than the fundamental frequencies measured in any of the flows with sharp reentrant corners.

The time-periodic lip vortex expands outwards with increasing $De_2$ until it reaches the upstream tube wall at $De_2 = 5.20$. At higher Deborah numbers the vortex begins to grow upstream as shown in Plate 6.2(d). It is also temporally stabilized, and the flow returns to a steady two-dimensional state. At $De_2 = 5.5$ the vortex height reaches a maximum size $\chi = 0.45$ and begins to oscillate in a way similar to that described in Section 6.3.2. The frequency of vortex oscillation is once again found to be far more rapid ($f_c \approx 0.400$ Hz) than that observed for a sharp-lipped entrance.

At high flow rates the streamlines in the curved lip geometry were also observed to diverge outwards from the centerline, as previously described in Section 6.3.1 for abrupt contractions. Smoothing the reentrant corner was found to delay this transition and, as may be seen by examining Plates 6.2(c) and 6.2(d), the development of diverging flow does not occur until $De_2 \geq 4.5$. Sample LDV measurements of the axial velocity profile along the centerline are shown in Figure 6.23. At low $De_2$ the velocity monotonically increases as the fluid approaches the contraction plane and reaches a fully developed parabolic velocity profile within one small tube radius ($\zeta = +1$) downstream. At $De_2 = 4.60$ the local velocity exhibits a minimum above the contraction entrance at $\zeta = -2.0$ and a small velocity overshoot along the centerline just downstream of the contraction at $\zeta = +0.70$. 274
Figure 6.22 Time-dependent oscillations in the velocity near the lip of the 4:1 contraction with a curved entrance lip ($\beta = 4e$): (a) The radial and axial velocity components near the lip entrance ($\xi = 1.50$, $\zeta = -0.90$) at $De_2 = 5.02$ and $Re_2 = 0.088$; (b) FFT of the axial velocity component gives the frequency of oscillation as $f_{\text{curve}} = 0.390$ Hz.
Figure 6.23 Normalized axial velocity \( u_2(0, \zeta) / \langle u_2 \rangle_2 \) along the centerline of the 4:1 contraction with a curved entrance lip (\( \beta = 4_c \)): (a) At \( De_2 = 0.42, Re_2 = 0.004 \) the flow converges smoothly towards the small tube and the velocity increases monotonically; (b) at \( De_2 = 4.60, Re_2 = 0.08 \) the flow upstream of the contraction diverges from the centerline and the axial velocity shows a local minimum at \( \zeta = -2.0 \).
6.5 Discussion of Results

The experiments described here demonstrate a rich variety of nonlinear fluid mechanics in the contraction flow of this particular viscoelastic fluid. Flow transitions involving both steady-state, two-dimensional and time-dependent, three-dimensional motions have been documented as a function of the Deborah number of the flow and of the contraction ratio. These results are summarized on the transition diagram, Figure 6.14, and give a rational description of the evolution of the dynamics with $De_2$ and $\beta$.

6.5.1 Influence of the Contraction Ratio on the Stability of Viscoelastic Entry Flows

Any theory for describing flow transitions in the contraction geometry must produce stability curves of the form shown in Figure 6.14, with at least the same qualitative dependence on $De$ and $\beta$ for fluids with the same rheology. Moreover, the flow transitions for other viscoelastic fluids must be described by qualitatively similar transition curves that may be shifted by differences in the material properties of the fluid. One of the most important challenges is to understand the impact of fluid rheology on the details of the flow transitions, i.e. on the shapes and locations of the transition curves in Figure 6.14. Both the shear and extensional characteristics of the fluid are no doubt important, and the variation of the critical Deborah number $De_2^{(asc)}$ with contraction ratio suggests that the upstream flow conditions play an important role in the dynamics observed in the lip region.

The effect of shearing in the upstream tube is characterized by the wall shear rate $\dot{\gamma}_w = 4\gamma_2 / \beta^3$. It is less clear which extensional parameter should be employed to characterize the behavior observed in each contraction ratio. The LDV data presented in Figures 6.2 and 6.7 show that, near the contraction plane, the fluid experiences approximately the same extensional strain rate $\dot{\epsilon}$ in each contraction ratio at the same value of $De_2$. This suggests that the total extensional strain $\epsilon$ may be a more appropriate measure for the flow, as originally suggested by Boger (1987). The total Hencky strain experienced by fluid elements moving along the centerline is

$$
\epsilon = \int_0^{+\infty} \dot{\epsilon} \, dt = \int_{u(-\infty)}^{u(+\infty)} \frac{du}{u} \ln(\beta^2) \tag{6.4}
$$

277
Hence, the upstream shear rate and total extensional behavior are two competing influences which vary quite differently with contraction ratio. For a given $De_2$, low contraction ratios will produce more highly distorted molecules in the upstream flow due to shear, whereas higher contraction ratios will produce greater elongation of the macromolecules because of the larger Hencky strains. The relative importance of shear and extensional characteristics in the fluid can then be used to explain qualitatively the differences in the flow structure caused by changing the contraction ratio $\beta$. In all contraction ratios the flow structure changes from a Newtonian-like pattern as the Deborah number is increased. The shape of the weak corner eddy observed in the outer corner of the upstream tube becomes convex, the intensity of recirculation increases, and the center of rotation shifts towards the center of the tube as shown previously in the sketches of Boger (1987) reproduced in Figure 2.9. The LDV measurements presented in Sections 6.2 and 6.3 allow these qualitative diagrams to be associated with quantitative documentation of hydrodynamic transitions occurring in the flow near the re-entrant corner as follows:

For low contraction ratios ($\beta \leq 5$) the weak recirculation in the corner collapses and a time-dependent lip vortex develops that is clearly isolated from the outer tube walls. As shown in Plates 6.1(a) and 6.2(a), the Moffat vortex is a weak recirculation in the outer corner of the upstream tube with a concave boundary separating it from the primary flow through the contraction. At higher $De_2$ this corner vortex has almost completely disappeared and fluid streamlines near the outer walls extend down to the contraction plane, as shown in Plates 6.1(b) and 6.2(b). The large elastic vortex that develops and grows upstream at higher $De_2$ is shown in Plates 6.1(b) and 6.2(c) to originate in the immediate vicinity of the reentrant lip corner and subsequently grow outward to the outer corner as the Deborah number is increased. This isolated lip vortex gives rise to the complex time-dependent dynamics documented in Section 6.2 and 6.4.

At higher Deborah numbers, the significant shear rates experienced in the upstream tube for small $\beta$ will lead to a shear-thinning in the fluid elasticity. This shear-thinning near the wall, coupled with extensional thickening along the centerline may explain why the flow upstream develops the diverging streamlines documented in Plate 6.1(c) and Figure 6.15. As the contraction ratio is increased the shear rate upstream rapidly decreases leading to less shear-thinning and a decrease in the diverging flow. Unfortunately, to quantify these competing effects requires accurate experimental measurements of the extensional behavior of elastic liquids and of the stress distributions throughout the contraction geometry. Very recently, however, these arguments have been corroborated by the streak photographs of Boger and Binnington (1990) for diverging flow of an 0.244 wt% PIB/PB Boger fluid formulation through a 4:1 axisymmetric contraction: both the measurements in this thesis
for entry flow of an 0.31 wt% PIB/PB Boger fluid, and the new work of Boger and Binnington have documented diverging flows for $\beta = 4$; whereas the work of Boger et al. (1986) for an 0.10 wt% PIB/PB Boger fluid showed no diverging streamlines. Thus, for a fixed contraction ratio of $\beta = 4$ it is found that the diverging flow regime is more pronounced in Boger fluids with higher concentrations of PIB. Rheological characterization shows that the formulations with a higher concentration of the PIB polymer also exhibit a more pronounced shear-thinning in the first normal stress coefficient $\Psi_1$ at higher shear-rates. Hence, shear-thinning appears to be intimately connected with the development of a diverging flow regime.

In the larger contraction ratios ($\beta \geq 6$) the center of rotation of the corner vortex also moves inwards as the Deborah number increases; however, the significantly lower shear-rates in the upstream tube and the higher extensional strain prevent separation of the corner vortex and lip vortex (see Figure 2.9(a)). The recirculation extends across the complete contraction plane, as shown in Plate 6.1(d), and the flow remains steady and two-dimensional. The presence of this recirculation coupled with the much lower shear rates in the upstream tube prevent the development of diverging streamlines, and the flow converges radially into the downstream tube at all $De_2$ experimentally attainable.

The differences in the dynamics of lip vortex formation between PAC/CS and PIB/PB Boger fluids reported by Boger and coworkers (1986, 1987), and shown in Figure 2.9(a) and 2.9(b), must arise from differences in the fluid rheology. The stability diagram for contraction flow of PAC/CS fluids may be similar to the one shown schematically in Figure 6.14; however, the neutral stability curve for onset of the lip instability and subsequent development of an isolated time-dependent lip vortex must be shifted to the left with a minimum near $\beta = 2$. A schematic representation of these ideas is shown in Figure 6.24. Time-dependent entry flows of PAC/CS Boger fluids will thus only be observed in very small contraction ratios and for $\beta \geq 4$ the first flow transition will be development of a lip vortex that extends across the contraction plane and coexists with the corner vortex. Quantitative verification of such arguments requires further LDV experiments in entry flows of PAC/CS fluids and reliable experimental data on the extensional properties of both fluids.

Substantial vortex growth was observed for all contraction ratios $2 \leq \beta \leq 8$. In addition, the flow visualization results have shown that in the low contraction ratios ($\beta \leq 5$) this large elastic vortex does not develop simply from an expansion of the weak Newtonian corner eddy that is observed at low Deborah numbers. The upstream growth of the large elastic vortex ($\chi$) in each contraction is found to correlate well with the downstream Deborah number $De_2$, in agreement with the comprehensive experiments of Boger et al.
Figure 6.24 Schematic drawings showing stability diagrams that provide a rational description of the results presented in this thesis and the earlier observations of Boger (1987) for contraction flows of (a) PIB/PB Boger fluids and (b) PAC/CS Boger fluids.
(1986); however, this relationship does not explain the origin of the large vortices. Experiments with polymer melts (White and Kondo 1977/78; White and Baird 1986; Piau et al., 1988) indicate that vortex growth occurs only when the fluid exhibits a large extensionally-thickening elongational viscosity. This picture is consistent with the limited elongational viscosity data that is available for Boger fluids; recent experimental data for polyisobutylene based Boger fluids suggest that the Trouton ratio varies from 100 to 1000 (Walters, 1989; Sridhar, 1990). The nonlinear multimode constitutive equations which have been accurately fitted to both steady and transient shear flow rheological data for the PIB/PB Boger fluid used in these experiments also predict large Trouton ratios in the range $\overline{\eta}/\eta_0 \sim 1000 - 3000$, as shown in Figure 4.20.

The correlation of vortex growth with extensional properties however fails to explain the anomalous behavior of Boger fluids in planar geometries, previously discussed in Section 2.2.3. Despite evidence that Boger fluids exhibit significant strain-rate thickening in both uniaxial and planar extensional flows (Williams and Williams, 1985; Jackson et al., 1987), the flow visualization results of Walters and coworkers (1982a,b; Binding and Walters, 1988) have clearly shown that Boger fluids do not show any pronounced vortex growth in planar contractions. Little is known about the presence of instabilities in the planar geometry, but once again flow conditions upstream of the contraction plane must be important: For a given contraction ratio $\beta$, the upstream wall shear rate in a planar contraction is $\gamma_{w1} = 4\gamma_{2}/\beta^2$ (a factor of $\beta$ higher than an equivalent axisymmetric geometry) and the Hencky strain will be $\varepsilon = \ln \langle u_2 \rangle_2 / \langle u_2 \rangle_1 = \ln \beta$; i.e. a factor of 2 lower than the value given by eq. (6.4). It is plausible that in planar contraction flows of Boger fluids the higher wall shear rates in the upstream channel lead to shear-thinning in the normal stress coefficient and the development of a diverging flow regime before extensional thickening effects can generate significant vortex enhancement.

At high Deborah numbers the large elastic vortex generated in the upstream tube of the axisymmetric contraction becomes unsteady and undergoes further flow transitions to macroscale time-dependent motions. By combining LDV velocity data and flow visualization it has been possible to quantify two distinct modes of oscillation which are shown in Figure 6.14. In low contraction ratios ($\beta = 2, 3, 4$) the elastic vortex remains axisymmetric but slowly pulses in vertical height with a frequency, $f_{\text{pulse}} = 0.04$ Hz, whereas in the higher contraction ratios ($\beta = 5, 6$) a more rapid azimuthal rotation of the vortex ($f_{\text{rot}} = 0.10$ Hz) is observed. This azimuthal ‘spiralling flow’ has previously been observed in the contraction flow of shear-thinning fluids at high Reynolds number (Rama Murthy, 1974; Cable and Boger, 1979; Yoganathan and Yarlagadda, 1984), and in the flow of a PAC/CS Boger fluid through a 7.675:1 axisymmetric contraction by Nguyêñ and
Boger (1979). In the previous investigations little quantitative information on the vortex dynamics was presented. However, the frequency of oscillation was found to increase with flow rate and to be comparable to the values presented here. Cable and Boger (1979) reported unsteady flow of a shear-thinning polyacrylamide through a 4:1 contraction with a spiralling frequency of 0.3 Hz at a downstream wall shear rate of $\dot{\gamma}_w = 590 \text{ s}^{-1}$ which increased to 0.8 Hz at $\dot{\gamma}_w = 2500 \text{ s}^{-1}$ as the flow rate was increased.

These large vortex oscillations severely disturb the velocity field throughout the upstream and downstream tube and have often been compared to the instabilities encountered in the extrusion of polymer melts through a die: Periodic oscillations develop in the die reservoir upstream of a contraction at stress levels in the die land of approximately $10^2 \text{ kPa}$ and result in helical distortion of the extrudate (see for example den Otter, 1970; Ballenger and White, 1971; White, 1973). The data in Table 6.2 for the 4:1 contraction show that the critical Deborah number for onset of vortex oscillation is $De_2 = 4.40$ ($Re_2 = 0.077$), corresponding to a wall shear rate $\dot{\gamma}_w = 224 \text{ s}^{-1}$ and a wall shear stress of $\tau_w = 2.38 \text{ kPa}$. Due to the much lower viscosity of polymer solutions the magnitude of the wall shear stress calculated at the onset of vortex oscillation is almost two orders of magnitude smaller than that observed in polymer melts and does not appear to be a good criterion for the development of flow instabilities. An alternative criterion that has been proposed is a critical value of the stress ratio $N_1/\tau = \Psi_1 \dot{\gamma}/\eta$ evaluated at the wall shear rate in the downstream tube (Petrie and Denn, 1976; Cable and Boger, 1979). As discussed in Chapter 4.5, the stress ratio is directly proportional to the definition of the Deborah number $De_2$ in eq. (6.1) and the critical value of the stress ratio at the onset of vortex oscillations is listed in Table 6.2 for each contraction ratio. Flow transitions to a spiralling vortex regime have been observed at stress ratios of 5.3 for a monodisperse polystyrene melt (Vlachopoulos and Alam, 1972), between 4.5 and 5.5 for a PAC/CS Boger fluid (Nguyen and Boger, 1979) and at a stress ratio of 5 for shear-thinning polyacrylamide solutions at low Reynolds number (Cable and Boger, 1979). The critical stress ratios $N_{1w}/\tau_w$ given in Table 6.2 for the 0.31 wt% PIB/PB/C14 solution are larger but of a similar magnitude to those presented in the previous investigations. It is also noted that in all of these studies the onset of vortex oscillations is found to occur in the asymptotic shear-thinning region (see Figure 4.22) where the stress ratio is a very weak function of shear rate. In this region, shear thinning results in a rapid decrease in the fluid elasticity (as measured by $\Psi_1$) and inertial effects become increasingly important.

The experiments in Section 6.4 have shown that rounding the sharp re-entrant lip corner does not prohibit the development of elastic phenomena such as a lip vortex or diverging streamlines above the contraction plane. The flow transitions are merely shifted
to higher Deborah numbers. An oscillating lip vortex is still observed in the 4:1 contraction geometry with a smoothly curved lip, although the specific dynamic behavior is significantly altered from that observed with the sharp lip. This suggests that the formation of an elastic lip vortex in axisymmetric contraction flows of Boger fluids reported here and by previous authors (Lawler et al., 1986; Boger et al., 1986; Boger, 1987) is not directly related to the presence of a singularity in the flow, but arises from the accelerating flow near the corner. Smoothing the corner reduces the local extension rate for a given $De_2$ and eliminates the lip vortex, as observed by comparing Plates 6.1(b) and 6.2(b). However, increasing the Deborah number in the curved lip geometry increases the extension rate and stress level near the corner and, at a sufficiently high value of $De_2$, a lip vortex similar to the one documented in the sharp lip geometry results. Experimentally the reduction in stresses near the lip is manifested as a lower pressure drop across the smoothed contraction entrance. Although the entrance pressure drop is not measured explicitly with this apparatus, it was found that for a fixed maximum pressure driving force, $\Delta P = 200$ Pa (30 psi), a higher maximum $De_2$ could be achieved in the curved lip geometry (see Figure 6.21). A further set of experiments is envisaged in which the entrance pressure drop and the critical Deborah number for formation of a time-dependent lip vortex are measured as the radius of curvature of the lip is varied.

Other modifications to the abrupt contraction geometry have been investigated in the literature, including tapering the die entrance to produce a conical converging entrance into the downstream tube. This modification to the local flow near the corner results in similar changes of the viscoelastic flow transitions to the ones we have observed by curving the lip: In polymer melts conical entrances lead to smaller amplitude oscillations of a higher frequency than those observed in a corresponding abrupt geometry (Bagley and Schreiber, 1961; den Otter, 1970), and in polymer solutions a lip vortex near the tapered reentrant corner is still observed (Boger, 1987; Evans and Walters, 1989). The nonlinear transitions leading to elastic vortices may be common to all viscoelastic flows with high extensional strains which result in significant elongation to the polymer chains and the development of large normal stress components in the fluid.

The LDV measurements presented in this Chapter results vividly demonstrate the importance of flow near the lip of the contraction on the quantitative details of the dynamics of viscoelastic entry flows. Although a complete understanding of the relationship between these flow transitions and the details of the fluid rheology must await accurate numerical calculations, these results and the discussion of previous observations clearly show that generic flow transitions do exist in entry flows and that the particular ordering of the transitions is a strong function of the contraction ratio and the fluid rheology.
6.5.2 Application of Phase-Plane Techniques to Axisymmetric Contraction Flows

The experimental approach employed in this thesis for documenting the nonlinear dynamics of contraction flows is similar to that used in other dynamical systems and can be summarized as follows: an external control parameter (in this case, the Deborah number $De_2$) is increased in small steps, and for each value of $De_2$ a long time series of a directly measurable system variable (i.e. the velocity $u_i(t)$ or $v_i(t)$ of the fluid at a fixed point) is collected. The Fourier transform $\mathcal{F}$ of this time series $\tilde{u}(f) = \mathcal{F}\{u(t)\}$ is calculated by an FFT and the frequency spectrum is analyzed to determine if a quantitative change in the system dynamics has occurred. This approach has been particularly successful in documenting the period-doubling and quasiperiodic transitions shown in Figure 6.10 and Figure 6.11. Similar hydrodynamic transitions in Newtonian fluid flows have also been determined from Fourier spectra of LDV time-series by Fenstermacher et al. (1979). The Fourier transform is ideal for quantifying periodically repeating patterns in noisy experimental time-series data such as Figure 6.11(a); however, a limitation of the method is that an FFT calculated from a time-series of finite length cannot accurately represent patterns that are aperiodic or chaotic; i.e. patterns that do not recur periodically in time. Insight into low-dimensional chaotic behavior in other dynamical systems has been achieved by examining ‘phase-portraits’ of the system (Bergé et al., 1986; Baker and Gollub, 1990). These representations of the phase-space of a complex system are typically constructed in simulations of model problems by plotting two independent scalar time series against each other (e.g. for a model fluid system one could choose from the computed velocity $u(t)$, acceleration $\partial u(t)/\partial t$, shear rate $\dot{\gamma}(t)$, or pressure $p(t)$). The resulting trajectory can provide quantitative information on the number and type of fixed point attractors in the system.

Recently, Fraser and Swinney (1986) have also shown that phase portraits of the system can be reconstructed from experimental measurements of a single scalar time series by the ‘method of delay times’. To implement this method, the scalar time series $u(t)$ is expanded into a vector series $\mathbf{v}(t)$ by using a delay time $T$ such that $\mathbf{v}(t) \equiv (u_0(t), v_1(t),...)$, where $u_n(t) = u(t + nT)$. This delay time $T$ must be chosen so that the data $u_0(t)$ and $v_1(t)$ are independent of each other. The simplest method for determining $T$ is from the value of the time $t$ at which the autocorrelation function of the time-series, $\mathcal{R}_u(\tau)$, first passes through zero. This is equivalent to requiring that the two time series $u_0(t)$ and $v_1(t + T)$ are linearly independent of each other. A superior, but computationally more intensive approach derived from information theory has also been developed by Fraser and
Swinney (1986). In this method the value of $T$ is taken as the first minimum of the 'mutual information' of the data; the authors show that is equivalent to minimizing the general dependence (i.e. both linear and nonlinear correlations) in the two time-series $v_0(t)$ and $v_1(t + T)$.

The former, more straightforward approach has been applied to the velocity time series experimentally measured with LDV in the axisymmetric contraction flow. The autocorrelation function $A$ is calculated directly from the the FFTs of the time series $\bar{u}(f)$ as (Press et al., 1985)

$$A[u(t)] = \mathcal{F}^{-1} [\bar{u}(f) \bar{u}^*(f)]$$

(6.5)

where $\mathcal{F}^{-1}$ is the inverse Fourier transform function and $\bar{u}^*(f)$ is the complex conjugate of the complex Fourier transform $\bar{u}(f)$.

A sample autocorrelation of the 1024 data points contained in the nonlinear time series $u_x(0, -25, t)$ shown in Figure 6.20(a) is plotted in Figure 6.25(a). In essence, autocorrelation of the data amplifies the dominant frequency of oscillation in the data and minimizes the small nonlinear contributions of the higher harmonics. At time $t = 0$, the time series is perfectly correlated with itself ($A(0) = 1$), and the correlation is a slowly decaying function of time (since the time series is a record of finite length). The first zero in the autocorrelation function is determined from Figure 6.25(a) to be at $T = 5.5$ seconds. The phase-portrait constructed by plotting $u_x(t + 5.5)$ against $u_x(t)$ is shown in Figure 6.25(b). The data is periodically repeating and reveals the characteristic spatial pattern of a limit cycle about a single fixed-point attractor.

A second example is shown in Figure 6.26 for the time-dependent axial velocity $v_1(t)$ measured near the lip of the 4:1 contraction at $(\xi, \zeta) = (1.02, -0.32)$ and $De_2 = 2.46$. The autocorrelation function shown in Figure 6.26(a) is used to determine the delay time of $T = 1.563$ s. At $De_2 = 2.46$, the flow is in the time-periodic state and the velocity time-series and FFT spectrum are similar to those shown in Figure 6.10(a) and (c), with a dominant oscillation frequency of $f_1 = 0.16$ Hz. The phase-portrait calculated with $T = 1.563$ s is shown in Figure 6.26(b). The limit cycle characteristic of a periodic signal is no longer circular shape and instead shows the beginning of a 'fold' in the upper left corner of the phase-portrait. This is indicative of the appearance of lower frequency components in the spectrum. As these peaks become more pronounced, the fold in the limit cycle becomes larger and the period taken for a complete trajectory of the attractor becomes much larger. This is equivalent to the onset of the period-doubling transition shown in Figures 6.10(b) and (d). Beyond this transition the strongest peak in the spectrum will be
Figure 6.25 (a) Autocorrelation function $A(t)$ of the axial velocity time series at $De_2 = 4.49$ shown previously in Figure 6.19(a). (b) Phase-plane portrait constructed by using the method of delays to generate the independent coordinates $v(t), v(t + 5.5)$. 
Figure 6.26 (a) Autocorrelation function $A(t)$ of the axial velocity time series near the lip of the 4:1 contraction at $(\xi, \zeta) = (1.02, -0.32)$ and $De_2 = 2.46$. (b) Phase-plane portrait constructed by using the method of delays to generate the independent coordinates $u(t), u(t + 1.563)$. 
the \( f_{1/2} \) peak and determination of the first minimum in the autocorrelation function will yield a much larger value of \( T \). This change in the delay time \( T \) effectively results in a rotation of our ‘view’ of the attractor shown in Figure 6.26(b) so that we ‘see’ a new single limit cycle with a period of twice that shown in Figure 6.26.

The significant scatter in the data shown in Figures 6.25(b) and 6.25(b) is important and emphasizes the importance of signal quality restrictions associated with such phase-plane techniques: quantitative determination of more complex systems with chaotic attractors requires very long time series (typically \( 10000 - 30000 \) data points) and high signal-noise ratios of better than \( 1000:1 \) (Broomhead and King, 1986). By contrast, the data acquisition techniques employed in this thesis are limited by low data rates (due to the low velocities) and by ‘shot noise’ in the photomultipliers to signal-noise ratios of \( 100:1 \), at best. Therefore, it is not possible to deduce more than qualitative information about the nonlinear dynamics observed in contraction flows from such techniques. However, the approach is still helpful in emphasizing the dynamic state of the system observed at a given Deborah number. An example from the chaotic flow regime in the 4:1 contraction at \( De_2 = 4.2 \) is shown in Figure 6.27. The FFT of the time series \( v_x(-1.68, -0.30, t) \) shown in Figure 6.27(a) contains broad-band noise across the frequency spectrum similar to that shown previously in Figure 6.16(b). The autocorrelation function of the data is shown in Figure 6.27(b). Again the signal is perfectly correlated with \( A(t) = 1 \) at \( t = 0 \); however, it can be seen from Figure 6.27(b) that the correlation decays very rapidly in time and oscillates about zero (note the expanded scale of the ordinate axis); the velocity at a time \( t + \delta t \) is thus almost linearly independent of the velocity at time \( t \). This is characteristic of a chaotic or aperiodic flow regime (Bergé et al., 1986). Further development of these techniques in viscoelastic flows requires significant improvements in the signal-to-noise ratios of the LDV system or new data-acquisition tools. Phase-portrait methods may, however, be of use in analysis of model nonlinear systems for viscoelastic flows and in time-dependent numerical simulations.

6.5.3 Comparison with Numerical Simulations of Viscoelastic Entry Flow

Although numerical simulations of viscoelastic flow through abrupt contractions with a variety of differential constitutive equations have been presented in the literature, these calculations are plagued by convergence problems associated with the large velocity gradients and stresses near the reentrant corner (see for example Mendelson et al., 1982;
Figure 6.27 (a) FFT of the time-dependent radial velocity component $v_r$ measured at $(\xi, \zeta) = (-1.68, -0.30)$ in the 4:1 contraction for $De_2 = 4.08$. (b) Autocorrelation function $A(t)$ of the Fourier spectrum shown in (a).
Lipscomb et al., 1986; Keunings, 1987). Whether these difficulties are the results of computational difficulties or the inherent lack of integrability of the constitutive equations in the presence of a singularity is an unresolved problem; however, calculations with a model constructed to give an integrable singularity do converge with mesh refinement to high $De$ (Coates et al., 1991). Marchal and Crochet (1987) have presented steady-state, finite-element calculations with an Oldroyd-B fluid model for flow through a 4:1 axisymmetric contraction up to a $De_2$ of over 50 and see the formation of a large vortex in the upstream tube at very high values of $De$. Unfortunately, insufficient detail is given to determine whether the vortex grew out of the Newtonian Moffat eddy or from an elastic vortex originating near the lip. This detail is important in determining the reason for the order-of-magnitude discrepancy in $De$ between numerical simulations and experiments for a given vortex size that can be observed in Figure 1.2.

The steady-state calculations of Coates et al. (1991) have considered entry flows of the Modified Upper-Convected Maxwell, or MUCM, model originally developed by Apelian et al. (1988). Calculations in the 4:1 axisymmetric contraction show the spreading of the Moffat eddy present for the Newtonian flow and a shift in the center of rotation towards the contraction lip as the Deborah number is increased, but they do not show the formation of an independent elastic vortex near the lip. This is not surprising because the experiments documented above have shown that the elastic lip vortex is preceded by the transition to time-periodic flow near the lip. Consideration of the stability diagram shown in Figure 6.14 indicates that two-dimensional, steady-state numerical simulations of entry flows for PIB Boger fluids will be more successful for larger contractions ($\beta \geq 6$) since the flow near the corner singularity remains steady until the inset of large-scale vortex dynamics at high $De_2$. In order to investigate this hypothesis, a detailed comparison between numerical calculations for a single-mode MUCM model and flow visualization studies with the 0.31 wt% PIB/PB/C14 Boger fluid has been performed for flow through an 8:1 abrupt axisymmetric contraction. The results of this comparison are shown in Plate 6.3. Before discussing these results, it is noted that the detailed rheological characterization presented in Chapter 4 has clearly shown that a single-mode nonlinear constitutive equation (for example the Bird-DeAguiar, Giesekus, or MUCM model) can qualitatively describe the behavior of the viscometric properties $\eta$ and $\Psi_1$ measured in simple steady shear flow, but is insufficient to provide a quantitative fit across a wide range of shear-rates. In particular, the MUCM model predicts a monotonically decreasing first normal stress coefficient and cannot fully capture the nonlinear response in $\Psi_1(\gamma)$ shown in Figure 4.14. For this reason the results shown in Plate 6.3 are presented in terms of both a shear-rate-dependent Deborah number $De(\gamma) = De_2$ (calculated by using the shear-thinning
Plate 6.3  Comparison of the numerically calculated streamlines for the Modified Upper-Convected Maxwell model and experimental streak photographs for the 0.31 wt% PIB/PB Boger fluid in the 8:1 axisymmetric abrupt contraction. (a) Low flow rate with concave Newtonian vortex; (b) expansion of recirculation towards lip
Plate 6.3 (cont.) (c) Convex curvature of large elastic vortex; (d) vortex growth into the large, upstream tube.
relaxation time defined in eq. 4.45) and a zero-shear-rate Deborah number defined in terms of the zero-shear-rate relaxation time as $De_0 = \lambda_0 \dot{\gamma}$.

The flow at low $De(\dot{\gamma})$ shown in Plate 6.3(a) reveals the presence of a weak recirculation in the outer corner of the large upstream tube. Both the experimentally visualized particle streaks and the numerically determined streamlines show the concave curvature of the vortex shape and the initial spreading of the vortex towards the re-entrant corner that was originally sketched in 1987 by Boger (see Figure 2.9(a)). The vortex size is identically calculated from either experiment or calculation as $\chi = 0.18$. Increasing the Deborah number results in a change in both the shape and the size of the corner vortex, as shown in the sequence of photographs in Plate 6.3(b) – (d). In plate 6.3(b), the center of rotation of the vortex has shifted towards the sharp re-entrant corner and the curvature of the vortex boundary has become almost flat; however, the vortex size remains almost unchanged at $\chi = 0.19$. Further increases in $De(\dot{\gamma})$ result in the pronounced vortex growth shown in Plate 6.3(c) and 6.3(d). The recirculation extends across the base of the large upstream tube and the vortex boundary shows the characteristic convex curvature associated with a large elastic vortex. The agreement between the experimental measurements and numerical calculations is very encouraging, and is a considerable improvement over the order of magnitude discrepancy calculated by Debbaut et al. (1988) with the Oldroyd-B constitutive equation (see Figure 1.2). However, the comparison is still not quantitative, principally because the single mode MUCM model does not provide a quantitative description of the fluid rheology. In addition, the calculations are limited by convergence difficulties to a maximum shear-rate-dependent Deborah number of $De(\dot{\gamma}) = 0.60$. The maximum calculable vortex size is thus $\chi = 0.31$; whereas, at the highest flow rate shown in Plate 6.3(d) the experimentally observed value is $\chi = 0.43$. It is emphasized that this restriction is not due to instability in the numerical method, but rather is a result of limitations in computational resources. Numerical studies show that the method converges with mesh refinement and in principle higher $De(\dot{\gamma})$ can be achieved by refining the finite element mesh used in the calculations (Coates et al., 1991).

Steady-state, two-dimensional calculations for $\beta = 4$ also show a similar evolution in the vortex shape and size as $De$ is increased (Coates et al., 1991). These simulations can therefore accurately describe the vortex growth observed for flow of PAC/CS Boger fluids through 4:1 axisymmetric contractions (see Figure 2.9(b)), but are insufficient to capture the sequence of time-dependent transitions documented above for a PIB/PB solution in the 4:1 contraction. The qualitative sketches shown in Figure 6.24 indicate that time-dependent, and probably three-dimensional, flow simulations will be needed to unravel fully the details of viscoelastic contraction flows, and their dependence on the contraction

293
ratio β. Such calculations of time-dependent viscoelastic flows are just beginning (Fortin and Esselaoui, 1987; Northey et al., 1990) and are being used to explore flow transitions in other simpler geometries, such as the circular Taylor-Couette problem (Northey et al., 1991).
Chapter 7

Viscoelastic Flow Past a Cylinder Constrained in a Channel

The final geometry considered in this thesis consists of viscoelastic flow past a cylinder of radius $R$ mounted centrally in a long planar channel of half width $H$, as shown in Figure 2.11. The ratio of the cylinder radius to the channel half-width is defined as $\beta = R/H$. Far upstream and downstream of the cylinder the flow is fully developed plane Poiseuille flow. The symmetric points $S_1, S_2$ at the front and rear of the cylinder surface are stagnation points and polymer molecules will have large residence times in the vicinity of the cylinder, resulting in the development of large molecular extensions and significant elastic stresses. This stress boundary layer results in modification of the flow field around the cylinder, and in extrusion processes involving polymer melts (such as that shown in Figure 1.1) can lead to the development of ‘weld lines’ in the wake of the obstacle. Such defect structures lead to considerable degradation in the ultimate material properties of plastics extruded past submerged bodies (Tadmor and Gogos, 1979).

The literature review presented in Chapter 2.3 has shown that considerable discrepancy exists in the literature concerning the effects of elasticity on the shape of the streamlines near a cylinder. In addition the LDV measurements of Bisgaard (1983) in the wake of spheres falling through viscoelastic liquids have alluded to the possibility of unsteadiness in the strongly extensional flow field near a stagnation point. The aim of the LDV measurements presented in this Chapter is to document in detail viscoelastic effects on the steady-state kinematics observed near the cylinder and to investigate the stability of the strong planar extensional flows near the stagnation points $S_1$ and $S_2$.

A Cartesian coordinate system $(x,y,z)$ is defined as shown in Figure 7.1 with the origin $O$ at the center of the cylinder, the $z$-axis aligned along the flow direction, the $y$-axis in the ‘transverse’ direction and the $x$-axis pointing in the ‘neutral’ direction along the cylinder’s axis of symmetry. The characteristic length scale for this geometry is the cylinder radius $R$, and nondimensional coordinates are specified as $\xi = x/R$, $\nu = y/R$, $\zeta = z/R$. Design specifications for the geometry have been discussed in Chapter 3.3, and the
Figure 7.1  Viscoelastic flow past a circular cylinder constrained in a planar slit. The cylinder of radius $R$ is mounted centrally in the channel which has a width $2H$. A Cartesian coordinate system is defined with its origin at the center of the cylinder. Velocity profiles are measured along the channel center-plane (---), in the transverse direction across the channel width (- - -), and in the 'neutral' direction along the length of the cylinder (-- --).
geometric dimensions of the channel are \( H = 6.33 \text{ mm}, \Delta x = 12H = 76 \text{ mm} \). A cylinder-channel ratio of \( \beta = 0.5 \) has recently been proposed as an international benchmark for this geometry and the related axisymmetric problem of a sphere in a cylindrical tube (Hassager, 1988). Careful measurements of the cylinder diameter with a digital micrometer experimentally determined the cylinder radius to be \( R = 3.188 \pm 0.003 \text{ mm} \), yielding a cylinder-channel ratio of \( \beta = 0.5036 \pm 0.0005 \).

The volumetric flow rate through the geometry is used to define the average velocity in the channel as \( \langle u_y \rangle = Q/24H^2 \), and the characteristic residence time of polymer molecules near the cylinder is \( \mathcal{Z} = R/\langle u_y \rangle \). The Reynolds number (\( Re \)) and the Deborah number (\( De \)) for this problem are then defined by equations (4.46) and (4.47), respectively.

### 7.1 Newtonian Flow

#### 7.1.1 Flow in a Rectangular Channel

Far upstream and downstream of the cylinder the Newtonian flow in an infinitely deep channel should be fully-developed, plane Poiseuille flow with a parabolic profile of the form

\[
\frac{v_y}{\langle u_y \rangle} = \frac{3}{2} \left[ 1 - \left( \frac{y}{2R} \right)^2 \right] \tag{7.1}
\]

It is noted that the spatial coordinate in eq. (7.1) has been normalized with the cylinder radius \( R \) (for later convenience), and not with the channel half-width \( H \equiv R/\beta \).

However, unlike the purely two-dimensional, axisymmetric contraction flow discussed in Chapter 6, fluid motion in the channel is three-dimensional and the velocity also varies in the 'neutral' \( x \)-direction, due to the finite extent of the experimental apparatus and the no-slip boundary conditions at the end-walls. In order to ensure that the flow in the channel closely approximates the idealized Poiseuille flow, two different channel geometries were considered with aspect ratios of \( \Lambda \equiv \Delta x / \Delta y = 10 \) and \( \Lambda = 6 \). The flow in the center of the channel should therefore be unaffected by the distant end-walls and almost constant. Profiles of the axial velocity in the rectangular channel with \( \Lambda = 10 \) are shown in Figure 7.2 at \((v, z) = (0, \pm 10)\). In the transverse \( y \)-direction (Figure 7.2(a)) the velocity has a characteristic parabolic profile with a maximum value of \( v_y / \langle u_y \rangle = 1.5 \), as predicted by eq. (7.1). The axial velocity across the width of the channel is shown in Figure 7.2(b)
Figure 7.2  Profiles of the axial velocity $u_z(x, y)$ in a rectangular channel with aspect ratio of $A = \Delta x/\Delta y = 10$. (a) Parabolic dimensionless velocity profile $u_z(y)/\langle u_z \rangle$ in the transverse direction; (b) flat velocity profile $u_z(x)/\langle u_z \rangle$ across the width of channel. The solid lines are calculated with the exact Newtonian solution, eq. (7.4).
and rises very rapidly from zero at either end-wall to assume an approximately constant profile across 80% of the domain from $-16 \leq \xi \leq 16$.

The two-dimensional velocity profile $u_2(x, y)$ for the creeping flow of a Newtonian fluid in a rectangular channel is obtained analytically by solving the Navier-Stokes equations

$$\begin{align*}
v_x &= 0 \\
v_y &= 0 \\
\frac{\partial^2 v_x}{\partial x^2} + \frac{\partial^2 v_x}{\partial y^2} &= -\frac{\partial \phi}{\partial z}
\end{align*}$$

(7.2)

where $\phi \equiv P + \rho gz$ is the modified pressure. The boundary conditions for the flow are

$$\begin{align*}
v_x &= 0, \quad x = \pm 2\lambda R \\
\frac{\partial v_x}{\partial x} &= 0, \quad x = 0 \\
v_z &= 0, \quad y = \pm 2R \\
\frac{\partial v_z}{\partial y} &= 0, \quad y = 0
\end{align*}$$

(7.3)

Equation (7.2) can be solved as an infinite series expansion using a Fourier transform. The pressure gradient $\partial \phi/\partial z$ can be eliminated using the volumetric flow rate $Q = (2\lambda H^2)(u_y)$, and the final velocity profile is expressed in terms of the dimensionless variables $(\xi, \nu)$ as

$$\frac{v_x}{\langle u_x \rangle} = \sum_{p=0}^{\infty} \frac{48(-1)^p}{(2p + 1)^3} \cos\left((2p + 1)\pi \nu/4\right) \left[1 - \frac{\cosh((2p + 1)\pi \xi/4)}{\cosh((2p + 1)\pi \lambda/2)}\right]$$

(7.4)

The series converges rapidly with increasing $p$ due to the size of the denominator and one-dimensional velocity profiles at $\xi = 0$ and $\nu = 0$ have been calculated numerically using the first six terms of the series. These profiles are shown in Figures 7.2(a) and (b) by the solid lines and accurately match the experimental LDV measurements.

Similar measurements of $u_2(x, y)$ for an aspect ratio of $\lambda = 6$ are presented in Figure 7.3 far upstream of the cylinder at $\zeta = -10$. The velocity profiles in (a) and (b) are shown to scale in order to emphasize the difference between the transverse $y$-direction and the neutral $x$-direction. The velocity profiles $u_2(x, y)$ predicted by equation (7.4) for $\lambda = 6$ are also shown in Figure 7.3 by the solid lines. Again the velocity is parabolic in $y$ and is approximately flat across 50% of the channel from $-6 \leq x/R \leq 6$.

The measurements presented in Figures 7.2 and 7.3 show that approximately two-dimensional flow can be realized for aspect ratios of $\lambda = 6$ or $\lambda = 10$. However, for a
Figure 7.3  Profiles of the axial velocity component in a rectangular channel with aspect ratio $A = 6$. (a) Parabolic dimensionless velocity profile $u_z(y)/\langle u_z \rangle$ in transverse direction; (b) flat velocity profile $u_z(x)/\langle u_z \rangle$ across width of channel. The solid lines are calculated with the exact Newtonian solution, eq. (7.4).
fixed pump capacity the maximum velocity (and also maximum $De$) that can be attained varies inversely with the cross-sectional area of the channel. For this reason the smaller, 6:1 channel ratio was selected for the bulk of the LDV measurements presented in this Chapter.

### 7.1.2 Creeping Flow Past a Constrained Cylinder; $\beta = 0.5$  

The evolution of the velocity field around the cylinder at low $De$ is summarized in Figure 7.4. The velocity profile is initially parabolic far upstream ($\zeta = -10$), then begins to flatten as the fluid approaches the cylinder ($\zeta = -3$). Off center maxima develop at $\zeta = -2$, $-1$, and the velocity attains a maximum value at the symmetry plane $\zeta = 0$. At this point the velocity profile on each side of the cylinder is again parabolic; for such an inertialess or ‘creeping’ flow, the fluid momentarily assumes the same velocity profile as would be observed for plane Poiseuille flow between parallel plates separated by a distance $R$. The maximum velocity is $u_r/(u_2) = 3.0$ and occurs at the points $\nu = \pm 3/2$. The velocity profiles are symmetric downstream of the cylinder, and the flow rapidly re-assumes a parabolic profile.

LDV measurements of the axial velocity in the planar stagnation flow near the cylinder are presented in Figure 7.5. Far upstream and downstream of the cylinder the normalized velocity on the channel centerline $(\xi, \nu) = (0, 0)$ has a maximum value of $u_r/(u_2) = 1.5$. As the fluid approaches the cylinder the velocity decreases very rapidly and Figure 7.5 shows that the influence of the cylinder only extends a distance of about $\pm 4R$ from the stagnation points $S_1$ and $S_2$. However, calculations with the Lamb-Oseen solution for the velocity near a cylinder in an unbounded medium (eq. 2.14) however predict reductions in the free stream velocity out to distances of $\zeta \sim 1/Re = 1000$. The rapid deceleration observed experimentally in the confined geometry is due to the constraining effects of the side-walls which deter the fluid from being forced radially outwards by the presence of the cylinder.

Very recently, full two-dimensional numerical solutions for the creeping flow of a Newtonian fluid past a cylinder in a channel with a ratio of $\beta = 0.5$ have been obtained (Lunsmann, 1991). The centerline velocity profiles are shown in Figure 7.5 by the solid lines. The agreement between the numerical computations and the LDV measurements is extremely good in both the upstream and downstream stagnation flow. The maximum difference shown in Figure 7.5 is approximately 7% and occurs downstream of the cylinder at distances of $3.0 \leq \zeta \leq 4.0$. To investigate the reproducibility of this effect,
Figure 7.4 Transverse velocity profiles of the dimensionless axial velocity $\frac{v_x}{(v_x)}$ showing Newtonian creeping flow of the Boger fluid past a circular cylinder at $De = 0.07$, $Re = 0.001$. 
Figure 7.5  Centerline axial velocity in the planar stagnation flow near the cylinder at $De = 0.07$. (●) LDV measurements in flow geometry with $R = 6.37$ mm, $A = 10$; (○) LDV measurements in flow geometry with $R = 3.188$ mm, $A = 6$; (——) Two-dimensional finite element solution of Lunsmann (1991).
LDV measurements were performed at the same $De$ in two separate flow geometries; one with a cylinder radius $R = 3.188$ mm and a 6:1 aspect ratio (solid symbols) and another with a cylinder radius of $R = 6.370$ mm and an aspect ratio of 10:1 (hollow symbols). The two data sets are self-consistent and both show the same, small 7% error. The origin of this discrepancy is unclear, but it seems unlikely to be an inertial effect for the small values of $Re$ attained in the experiments. Theoretical studies suggest that elasticity results in an $O(De)$ downstream shift in the streamlines (Mena and Caswell, 1974), and further calculations with the Oldroyd-B model at $De = 0.2$ also show a very small downstream displacement in the centerline velocity profile. However this shift is insufficient to describe the experimental data shown in Figure 7.5. It thus seems most probable that the discrepancy arises from a nonlinear viscoelastic phenomenon such as the complex shear-thinning observed in $\Psi_1(\dot{\gamma})$ for this Boger fluid, or from three-dimensional effects arising from the finite nature of the test geometries. To distinguish between these two possibilities it will be necessary to perform additional two-dimensional calculations for more realistic constitutive equations and also three-dimensional numerical simulations for a Newtonian fluid.

7.2 Effects of Elasticity on Flow Past the Cylinder

7.2.1 Evolution of Streamlines with $De$

As the Deborah number of the flow past the cylinder is increased, elastic effects result in a progressive modification to the velocity field around the cylinder. This is demonstrated by the centerline profiles shown in Figure 7.6 for the axial velocity in the front and rear stagnation flows near the cylinder. Upstream of the cylinder, in the steady shearing flow, the polymer molecules near the centerline are in an unextended, relaxed configuration, and the velocity profiles near the upstream stagnation point at $\zeta = -1$ superpose when scaled with the average velocity $\langle u_\parallel \rangle$ and the cylinder radius $R$. However, the macromolecules flowing very close to the cylinder experience a strong deformational flow that leads to the development of large molecular extensions and high elongational stresses. This deformation is 'remembered' as the molecules enter the downstream stagnation flow and results in the pronounced shift downstream of the velocity profiles in the wake of the cylinder shown in Figure 7.6.
Figure 7.6 Evolution of the axial velocity profiles $u_z/(v_z)$ along the channel centerline with increasing $De$; (a) upstream of the cylinder, and (b) in downstream wake of the cylinder.
7.2.2 The Elastic Wake Instability

Further increases in the Deborah number result in progressively larger downstream shifts in the velocity profiles until the onset of a flow instability occurs at $De = 1.30$. This instability results in a transition from a steady two-dimensional planar extensional flow in the wake of the cylinder to a steady, but three-dimensional cellular structure. A series of axial velocity profiles in the cylinder wake are presented in Figure 7.7 for increasing $De$. These profiles are measured in the neutral $x$-direction along the channel symmetry plane very close to the cylinder at $(v, \zeta) = (0, 1.5)$, as indicated by the dashed lines in Figure 7.1. At low flow rates the LDV velocity measurements in the wake of the cylinder are flat across the central section of the channel, as expected for a steady, planar stagnation flow. However, as the Deborah number is increased periodic oscillations appear in the velocity profiles, and the measurements presented in Figure 7.7 at $De = 1.38$ and 1.83 reveal large spatial fluctuations in the axial velocity that extend in the neutral $x$-direction along the length of the cylinder.

The computer program controlling the optical translation table has been modified in order to generate these profiles of the velocity in the cylinder wake very rapidly. The measuring volume is positioned at a point $x_1$ and then translated at a fixed velocity $u_x = 1.00$ mm/s to a new point $x_2$. The Doppler frequency measured in the flow is followed in real time by the frequency trackers described in Chapter 3.1 and stored on the computer as a file of 1024 evenly spaced data points. The time-dependent signal response is then converted to a spatial velocity profile by a simple mapping as $v(x) = v(x_1 + u_xt)$. Since the LDV beams intersect the flow cell at an oblique angle (see Figure 3.5) it is not possible to follow the velocity profiles directly to the end walls (located at $x = \pm 38.1$ mm); however the measurements presented in Figure 7.7 and subsequent figures contain velocity measurements across the central 80% of the channel from $x_1 = -30.5$ mm to $x_2 = +30.5$ mm.

The critical Deborah number for the onset of this wake instability was determined by performing a series of velocity scans along the cylinder length as the flow rate was slowly increased. The axial velocity measured at $\zeta = 1.50$ is shown in Figure 7.8 as a function of $De$. At low flow rates the axial velocity is constant across the cylinder and increases approximately linearly with $De$, as shown by the hollow symbols in Figure 7.8. At a critical value of $De_c = 1.30 \pm 0.01$ periodic oscillations appear in the velocity profiles near the cylinder. The maximum and minimum values of the axial velocity are shown in Figure 7.8 by the solid symbols.
Figure 7.7 Profiles of the axial velocity $v_z(x)$ across the width of the channel at $(v, \xi) = (0, 1.5)$ as the Deborah number is increased from $De = 0.70$ to $De = 1.83$. 
Figure 7.8  Magnitude of the axial velocity $v_z$ measured in the wake of the cylinder as a function of the Deborah number; hollow symbols (○) indicate the average value of the flat velocity profile in planar stagnation flow; solid symbols indicate the maximum (▲) and minimum (●) values of the velocity fluctuations measured in the cellular wake structure.
The wavelength of the perturbations in the axial velocity is determined by Fourier analysis of the spatial profile \( u_z(x) \). A sample velocity profile measured in the cylinder wake at \( \zeta = 1.75 \) is shown in Figure 7.9(a). An FFT of this signal results in a series of points that are equally spaced in ‘spatial frequency’ \( f_x \) [mm\(^{-1}\)]. Inversion of these values leads to a power spectrum measuring the spectral contributions of oscillations with a spatial wavelength of \( \lambda_x \) [mm]. The characteristic wavelength of the velocity fluctuations is determined from the FFT spectrum presented in Figure 7.9(b) to be \( \lambda_x = 3.03 \pm 0.15 \) mm. If this value is scaled with the cylinder radius \( (R = 3.188 \) mm), the wavelength of the disturbances is found to be almost equal to the radius of the cylinder; i.e. \( \lambda_x = 0.95R \pm 0.05R \). The LDV system can also been used to investigate the temporal stability of the flow in the cylinder wake. A velocity time series \( u_z(t) \) at \((\xi, \nu, \zeta) = (0, 0, 1.75)\) is presented in Figure 7.10 for the same flow conditions reported in Figure 7.9. The flow in the cylinder wake consists of oscillations that extend spatially across the length of the cylinder; however the velocity measured at a single point remains steady in time and the Fourier spectrum reveals no temporal oscillations. Hence, it is concluded that the elastic instability in the cylinder wake results in the formation of a steady, three-dimensional flow.

Video-imaging of the flow has been used to show unequivocally that these spatial oscillations in the velocity profiles are associated with the development of an evenly-spaced ‘cellular structure’ in the cylinder wake. A narrow longitudinal section of the flow at \( \nu = 0.0 \) is illuminated with a planar sheet of laser light as described in Chapter 3.2 and the video camera is aligned with the \( y \)-axis to visualize the flow along the length of the cylinder. Sequences which show the formation of this cellular structure at varying magnifications are included on the attached videotape (Appendix A) and representative still frames are presented in Plate 7.1. The low magnification image presented in Plate 7.1(a) demonstrates the presence of a periodically alternating banded structure in the cylinder wake. Combined LDV measurements and direct visual observation indicate that the bright areas correspond to the regions of higher axial velocity observed in velocity scans along the length of the cylinder. Plate 7.1(a) also clearly shows that the cellular structure extends across the length of the cylinder and is confined to the downstream wake; the flow upstream of the cylinder appears spatially uniform, and LDV measurements confirm that the velocity profiles here are two-dimensional.

Subsequent flow visualization experiments at higher magnifications of 30:1 show that the velocity field in the cylinder wake is three-dimensional, and particles very close to the cylinder move along the \( x \)-axis in both directions as they flow into the faster-moving, brighter regions of the wake. This effect can be clearly observed on the videotape but is more difficult to capture in still images. Plate 7.1(b) shows the velocity field in the cylinder
Figure 7.9  (a) Spatial fluctuations in the axial velocity $v_x(x)$ measured in the downstream wake of the cylinder at $De = 1.38$, $Re = 0.0117$; (b) FFT spectrum determines the spatial wavelength of the fluctuations to be $\lambda_x = 3.03$ mm.
Figure 7.10 (a) Time-series of the axial velocity $v_z(t)$ in the cylinder wake at the same position and flow conditions as Figure 7.9; (b) Fourier spectrum reveals no dominant frequencies of oscillation.
Plate 7.1  The elastic wake instability at flow conditions of $De = 2.48$, $Re = 0.028$; 
(a) video-imaging of the flow shows the formation of a cellular structure in the 
downstream wake that extends along the length of the cylinder, (b) higher 
magnification image close to the downstream stagnation point shows the velocity in the 
cylinder wake is three-dimensional with a $u_z$ component along the cylinder length.
wake at $De = 2.48$; the particle streak in the upper right section of Plate 7.1(b) clearly shows that there is a $u_x$ component of velocity very close to the cylinder.

This complex three-dimensional flow field in the cylinder wake has been carefully explored using the three-color LDV system. Figure 7.11(a) shows profiles of the $u_x$ and $u_z$ components of velocity measured on the channel centerline very close to the cylinder at $\zeta = 1.5$ and $De = 1.77$, $Re = 0.012$. At these conditions the flow around the cylinder remains symmetric about the channel centerline $v = 0$, and no $y$-component of velocity could be measured with the LDV system. The $u_x$ velocity along the cylinder axis oscillates about zero with a spatial wavelength of $0.95R \pm 0.05R$, as fluid flows in either direction from the slower moving regions into each faster moving cell. LDV measurements also show periodic fluctuations in the axial velocity $u_z$ component very close to the cylinder; however the wavelength of these oscillations is determined from the FFT in Figure 7.11(b) to be $\lambda_x = 0.47R \pm 0.02R$. These measurements show that the axial velocity oscillates with a frequency of twice that observed in the $u_x$ component, and close examination of the velocity profiles shown in Figure 7.11(a) reveal that $u_z$ displays a minimum value at each position where the magnitude $|u_x|$ reaches a maximum.

Similar measurements further from the cylinder at $\zeta = 2.0$ are shown in Figure 7.12. The $u_x$ component of velocity still shows periodic fluctuations about zero; however, the amplitude of oscillations are greatly reduced, and the FFT spectrum shows the presence of only a weak peak at $\lambda_x = 0.95R$. The axial velocity $u_z$ at $\zeta = 2.0$ also contains periodic oscillations; however, the wavelength of these oscillations shown in Figure 7.12(b) has doubled from those measured at $\zeta = 1.5$ (Figure 7.11(b)) to $\lambda_x = 0.95R$. Additional centerline measurements further downstream at $\zeta = 3.0$ and $\zeta = 5.0$ show that the velocity fluctuations are almost one-dimensional with periodic oscillations of wavelength $\lambda_x = 0.95R$ in the axial velocity component $u_z$ and no measurable $u_x$ or $u_y$ components.

The evolution of this complex cellular structure in the downstream wake of the cylinder is shown in Figure 7.13. Near the cylinder, at $\zeta = 1.5$, the complex three-dimensional flow results in the rapid oscillations of wavelength $\lambda_x = 0.5R$ discussed above. Further away from the cylinder at $\zeta = 2.0$, 3.0 there is no measurable $u_x$ component and the axial velocity fluctuates with a period equal to the cylinder radius. The cellular structure slowly decays further from the cylinder as the flow accelerates back to the channel centerline velocity, and 10 cylinder radii downstream the flow has almost recovered the uniform rectangular channel profile documented in Figure 7.2.

Similar LDV measurements coupled with direct flow visualization also show that the three-dimensional wake structure is confined to the narrow region of strongly
Figure 7.11 (a) Fluctuations in both velocity components $v_y(x)$ and $v_z(x)$ across the width of the channel at $(v, \zeta) = (0, 1.5)$ and flow conditions of $De = 1.77$, $Re = 0.012$; (b) the wavelength of oscillations in the axial velocity is determined from the FFT spectrum to be $\lambda_x = 1.50 \pm 0.04$ mm; (c) the wavelength of oscillations in the $x$-component velocity is $\lambda_x = 3.03 \pm 0.15$ mm.
Figure 7.12 (a) Fluctuations in both velocity components $v_x(x)$ and $v_z(x)$ across the width of the channel at $(v, \zeta) = (0, 2.0)$ and the same flow conditions as reported in Figure 7.11. The wavelength of oscillations in both velocity components is determined from the FFT spectra presented in (b) and (c) to be $\lambda_x = 3.03 \pm 0.15$ mm.
Figure 7.13 Evolution of the spatial structure observed in the centerline axial velocity $v_z(\xi, 0, \zeta)$ within the wake of the cylinder at flow conditions of $De = 1.83$, $Re = 0.0158$. 
extensional flow near the center-plane of the channel. Scans of the velocity at a fixed axial position of $\zeta = 1.50$ and different points in the transverse $y$-direction are shown in Figure 7.14. In the center-plane of the channel at $y = 0$ the axial velocity shows the rapid fluctuations of wavelength $\lambda_x = 0.5R$ documented above. The cellular structure can still be observed in flow visualization experiments if the plane of laser light is offset from the centerline and the velocity component $u_z(x)$ at $y = 0.53$ still shows oscillations, with a period equal to the cylinder radius. However, at greater distances from the center-plane the flow between the cylinder and the channel walls is primarily a steady shearing flow with only a weak elongational component, and LDV measurements of the axial velocity remain almost two-dimensional across the channel for all values of $De$.

### 7.2.3 Time-Dependent Flow in the Wake

As the flow rate past the cylinder is increased further a second flow transition is observed, and LDV measurements for $De \geq 1.85$ show that the flow in the wake becomes time-dependent. Representative time-series of the axial velocity in the cylinder wake at $(\xi, y, \zeta) = (0, 0, 1.4)$ are shown in Figure 7.15. The velocity $u_z(t)$ shows a nonlinear time-dependent response which repeats periodically. Video-imaging of the flow reveals that the cellular structures observed in the wake slowly progress outwards from the midpoint of the cylinder ($\xi = 0$) towards the side-walls of the channel. The time-dependent response in the velocity measured at a fixed point in space corresponds to the motion of this regular cellular structure through the measuring volume, and the nonlinear form of the oscillations arises from the complex spatial structure of the axial velocity profile at $\zeta = 1.4$ (see Figure 7.13). The wave speed for this travelling structure is calculated from knowledge of the spatial wavelength $\lambda_x$ of the cells and the oscillation frequency of the time series determined by the FFT spectra shown in Figure 7.15(b) and (d). As the Deborah number is raised from $De = 1.88$ to $De = 3.31$, the frequency of oscillations increases and the wave-speed increases from $U = 0.013$ mm/s to $U = 0.043$ mm/s, respectively. The onset point for this second flow transition was determined from a sequence of velocity time-series measurements obtained over 400 second intervals to be $De_c = 1.83 \pm 0.03$.

Time-dependent velocity oscillations in the wake of a cylinder are encountered in high Reynolds number flows of Newtonian fluids due to the formation of a von Kármán 'vortex street' (see the sequence of photographs in Figure 2.17); however, the maximum Reynolds number attained in the experiments with this Boger fluid is only $Re = 0.04$. The onset of time-dependence in the cylinder wake results solely from translation of the cellular
Figure 7.14 Measurements of the axial velocity profiles along the length of the cylinder at different transverse positions of $v = 0, 0.53, 0.79$ and $1.10$ show that the three-dimensional cellular structure is confined to narrow region of the cylinder wake.
Figure 7.15 Onset of time-dependent flow in the cylinder wake; (a) experimental time series of axial velocity $u_z(t)$ measured at $(\xi, v, \zeta) = (0, 0, 1.4)$, $De = 1.88$; (b) FFT spectrum shows very low frequency of oscillation corresponding to a wave speed of $U \equiv 0.013 \text{ mm/s}$; (c) velocity time series $u_z(t)$ measured at $(\xi, v, \zeta) = (0, 0, 1.5)$, for $De = 3.31$; (d) FFT spectrum shows an increased frequency of oscillation and a wave speed of $U \equiv 0.043 \text{ mm/s}$;
structure along the length of the cylinder and not from some periodic ‘shedd ing’ phenomenon in the stream-wise direction. Profiles of the centerline axial velocity in the stagnation flows upstream and downstream of the cylinder are shown in Figure 7.16. These velocity measurements are made with the frequency trackers by using the algorithm described above and vertically translating the table at a constant velocity \( u_z = 1.50 \text{ mm/s} \). The total data acquisition time is only \( \Delta t = 30\text{s} \) which is much smaller than the period of oscillation documented in Figure 7.15 for translation of the cellular structure along the cylinder axis. The velocity profiles upstream of the cylinder superpose when normalized with the average velocity \( \langle u_x \rangle \) and elasticity does not affect the upstream stagnation flow even at Deborah numbers of \( De > 3 \). However, in the wake downstream of the cylinder elastic effects result in a progressive downstream shift in the position of the streamlines around the cylinder. The LDV measurements for Newtonian flow at \( De = 0.07 \) are also shown in Figure 7.16 to emphasize the magnitude of this downstream displacement; at a fixed point \( \zeta = 5 \) the normalized axial velocity at \( De = 3.08 \) has been reduced by approximately 33% from its value at \( De = 0.07 \), and the flow does not recover a fully developed parabolic profile for distances of over 15R downstream of cylinder.

### 7.3 The Effect of the Cylinder-Channel Ratio, \( \beta \)

The wavelength of the cellular wake structure was determined from the measurements above to scale closely with the radius of the cylinder \( R \). To investigate this scaling further, additional experiments were performed with a smaller cylinder of radius \( R = 1.628 \text{ mm} \) which was centrally mounted in the same channel of half-width, \( H = 6.33 \text{ mm} \). The ratio of the cylinder diameter to channel width is thus reduced to \( \beta = 0.257 \). By analogy with the rotational flow experiments discussed in Chapter 5, two independent dimensionless elastic parameters can be defined for this flow; (i) the Deborah number, which is based on the residence time \( \mathcal{S} = R/\langle u_x \rangle \) for macromolecules near the cylinder, and (ii) a Weissenberg number defined in terms of the characteristic shear rate \( \dot{\gamma} = \langle u_x \rangle / (H - R) \) measured in the gap between the cylinder and the channel wall. The two flow parameters are then defined as

\[
De \equiv \lambda(\dot{\gamma}) \langle u \rangle / R
\]

\[
We \equiv \lambda(\dot{\gamma}) \langle u \rangle / (R - H)
\]
Figure 7.16 Centerline axial velocity profiles in the three-dimensional wake show a pronounced shift downstream with increasing $De$, but increase monotonically to the free stream value with no 'negative wake' or aperiodic fluctuations.
Substitution of the cylinder-channel ratio $\beta = R/H$ in eq. (7.6) shows that the two dimensionless groups are inter-related by $We \equiv \beta De / (1 - \beta)$. Hence, for a cylinder-channel ratio of $\beta = 0.5$, the Weissenberg number is equivalent to the Deborah number; however, for a smaller ratio of $\beta = 0.25$ the larger gap between the cylinder and the channel wall results in lower shear rates and the Weissenberg number is given by $We = \frac{1}{2} De$.

The effect of varying the cylinder-channel ratio is demonstrated by the centerline axial velocity profiles at $De = 0.14$ presented in Figure 7.17. The velocity profiles on the centerline far upstream and downstream of the cylinder superpose when normalized with the average velocity $\langle u_r \rangle$, however the shape of the velocity profiles is very different close to the cylinder, and the velocity decreases more gradually towards the smaller cylinder. This extension in the domain of influence of the cylinder may be explained as follows: the cylinders of varying size $R$ are positioned in the middle of a channel with a constant half-width $H = 12.66$ mm. In a cylindrical coordinate system $(r, \theta)$ centered on the cylinder, the local creeping flow in the channel will be perturbed out to a radial position of $r \sim H$. Along the channel centerline ($\theta = 0$) this equates to a distance of $z \sim H$, or in nondimensional coordinates, $\zeta \sim 1/\beta$. Smaller cylinders therefore perturb the velocity field out to larger dimensionless distances $\zeta$ (but similar physical distances of $z \sim H$) and result in the more gradual change in the velocity shown in Figure 7.17. This change in the shape of the velocity field is important because it indicates that, for a fixed Deborah number, a lower maximum extension rate $\dot{\varepsilon} \equiv \partial u_r / \partial z$ will be experienced near the smaller cylinder. From Figure 7.17 the maximum slope of each velocity profile is calculated to be $\dot{\varepsilon} \equiv 1.1 \langle u_r \rangle / R$ for $\beta = 0.50$, and $\dot{\varepsilon} \equiv 0.63 \langle u_r \rangle / R$ for $\beta = 0.25$; i.e. the reduction is almost a factor of two.

The experiments performed with the smaller cylinder ($\beta = 0.25$) still show the formation of a cellular structure, but the dynamics of the elastic wake instability are dependent on both the Deborah number of the extensional flow near the cylinder and the Weissenberg number in the gap. The centerline axial velocity profile along the length of the cylinder at $\zeta = 3.00$, for flow conditions of $De = 2.00$, $We = 0.727$, $Re = 0.0039$ is shown in Figure 7.18(a). The velocity in the wake shows periodic fluctuations similar to those presented in Section 7.2.2; however, the oscillations have a reduced wavelength of $\lambda_x = 2.30$ mm, equivalent to 1.4 times the new cylinder radius. The FFT spectrum in Figure 7.18(b) also shows that the wavelength selection of the velocity fluctuations is not as clearly defined in this geometry and peaks are observed at $\lambda_x = 2.03$ mm and $\lambda_x = 2.30$ mm.

To determine the critical conditions for onset of the wake instability in this geometry a series of experiments were performed which spanned the same range of flow rates, Deborah numbers and Weissenberg numbers as the experiments in Section 7.2.
Figure 7.17 Effect of the cylinder-channel ratio $\beta$ on the centerline axial velocity profiles in the stagnation flow near the cylinder at the same Deborah number of $De = 0.073$; (○) upstream and (●) downstream profiles for $\beta = 0.257$; (□) upstream and (■) downstream profiles for $\beta = 0.503$. 

$(\xi, \nu, \zeta) = (0, 0, \zeta)$
Figure 7.18 (a) Spatial fluctuations in the axial velocity $v_z(x)$ measured in the downstream wake of the smaller cylinder ($\beta = 0.257$) at $De = 2.00$; (b) The FFT spectrum determines the dominant spatial wavelength of the fluctuations to be $\lambda_x = 2.30 \pm 0.10$ mm.
Measurements of the axial velocity component \( v_z(x) \) at \( \zeta = 3.00 \) for the two different cylinder-channel ratios at the same Deborah number \( De = 1.38 \) are shown in Figure 7.19. The flow past the larger cylinder \( (\beta = 0.5) \) shows the presence of periodic oscillations in the cylinder wake with a cellular wavelength of \( \lambda_c = 0.95R \), whereas the flow near the smaller cylinder \( (\beta = 0.25) \) remains a planar stagnation flow with a flat velocity profile across the channel.

Similar measurements of the axial velocity component \( v_z(x) \) at \( \zeta = 3.00 \) for the two different cylinder-channel ratios at the same average upstream velocity \( \langle u_y \rangle = 3.71 \text{ cm/s} \) are shown in Figure 7.20. The volumetric flow rate \( Q \) and average velocity are the same for each geometry and the channel half-width is constant at \( H = 0.633 \text{ cm} \). Hence the characteristic shear-rate upstream is also a constant \( \langle u_y \rangle / H = 5.86 \text{ s}^{-1} \); however, the 50% smaller radius of the new cylinder results in a Deborah number which is approximately twice that calculated for \( \beta = 0.5 \). Periodic oscillations can now be observed in both velocity profiles and the difference in the wavelength of these oscillations is clearly shown in the FFT spectra presented in Figure 7.20(b) and (d).

Experiments for a number of intermediate flow rates between those presented in Figures 7.19 and 7.20 were performed to determine accurately the critical conditions for the onset of the elastic wake instability. The final results for the two different cylinder-channel ratios are summarized in Table 7.1:

<table>
<thead>
<tr>
<th>( \beta )</th>
<th>( R ) [cm]</th>
<th>( \langle v_z \rangle_c ) [cm/s]</th>
<th>( \dot{\gamma}_c ) [s(^{-1})]</th>
<th>( De_c )</th>
<th>( We_c )</th>
<th>( Re_c )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.257</td>
<td>0.1628</td>
<td>2.35</td>
<td>5.03</td>
<td>1.69 ± 0.05</td>
<td>0.61 ± 0.02</td>
<td>0.0049</td>
</tr>
<tr>
<td>0.503</td>
<td>0.3188</td>
<td>3.49</td>
<td>10.93</td>
<td>1.30 ± 0.01</td>
<td>1.30 ± 0.01</td>
<td>0.0142</td>
</tr>
</tbody>
</table>

**Table 7.1** Critical conditions for onset of the three-dimensional flow wake instability calculated at a reference temperature of \( T_0 = 25 \, ^\circ\text{C} \).

The evolution of the axial velocity profiles in the wake of the cylinder are shown in Figure 7.21. Oscillations in the axial velocity extend approximately ten cylinder radii downstream. The wavelength of these oscillations is determined from Fourier spectra of the velocity profiles to be \( \lambda_x = 2.30 \pm 0.10 \text{ mm} \), equivalent in dimensionless terms to \( \lambda_x = 1.41 \pm 0.06 \, R \). Very close to the cylinder, the axial velocity shows more rapid velocity fluctuations similar to those observed in Figure 7.13. An FFT of the velocity
Figure 7.19 Comparison of the axial velocity component $v_z(x)$ at the same Deborah number $De = 1.38$ for two different cylinder-channel ratios. (a) Velocity fluctuations at $\zeta = 2.0$ for larger cylinder-channel ratio of $\beta = 0.503$; (b) The dominant wavelength of oscillations is determined from an FFT as $\lambda_x = 3.03$ mm. (c) Planar velocity profile at $\zeta = 2.0$ for smaller cylinder-channel ratio of $\beta = 0.257$; (d) FFT reveals no cellular structure in the cylinder wake.
Figure 7.20 Comparison of the axial velocity component $v_2(x)$ at the same upstream flow conditions \( \langle u_x \rangle/H = 5.86 \text{ s}^{-1} \) for two different cylinder-channel ratios. (a) Larger cylinder-channel ratio of $\beta = 0.503$ with velocity fluctuations (b) of dominant wavelength $\lambda_x = 3.03 \text{ mm}$. (c) Velocity profiles at $\zeta = 3.0$ for smaller cylinder-channel ratio of $\beta = 0.257$ show velocity fluctuations (d) with reduced wavelength $\lambda_x = 2.40 \text{ mm}$. 
Figure 7.21  Evolution of the spatial structure observed in the centerline axial velocity $v_z(\xi, 0, \zeta)$ within the wake of the smaller cylinder ($\beta = 0.257$) at flow conditions of $De = 1.83$, $We = 1.07$, $Re = 0.0158$. 
signal at $\zeta = 2.0$ determines the wavelength of these oscillations to be half of the value observed farther from the cylinder, $\lambda_x = 1.08 \pm 0.02$ mm ($0.66 \pm 0.01 R$).

Profiles of the centerline velocity in the cylinder wake are shown in Figure 7.22. The results are similar to those observed in Figure 7.6 and 7.16 for a cylinder-channel ratio of $\beta = 0.50$. Once again, the velocity profile upstream of the cylinder is almost unchanged as the Deborah number is increased from $De = 0.14$ to $De = 3.51$, whereas viscoelastic effects in the downstream wake of the cylinder result in a pronounced shift downstream in the flow streamlines. At $De = 3.51$ the velocity profile is reduced from its free-stream value of $u_x/u_x = 1.5$ for axial distances of more than $20R$ downstream of the cylinder.

The measurements in each geometry have been used to construct an approximate stability diagram for viscoelastic flow past a cylinder in a channel, analogous to the diagrams generated in Chapter 5 for the stability of rotational flows, and in Chapter 6 for contraction flows. The results for the two cylinder-channel ratios $\beta = 0.257$ and $\beta = 0.503$ are shown in Figure 7.23 by the square and circular symbols respectively. The experimental results for a fixed geometric ratio $\beta$ lie along lines of constant slope $We/De = \beta / (1 - \beta)$. The abscissa corresponds to the case of a cylinder in an unbounded viscoelastic fluid ($\beta \to 0$). For low flow rates (shown by the hollow symbols) the velocity profiles in the wake of the cylinder are flat, and the flow is a steady, two-dimensional planar stagnation flow. At the critical conditions given in Table 7.1 this base flow becomes unstable, and the velocity profiles in the cylinder wake develop the three-dimensional cellular structure documented above. It is seen from Figure 7.23 and Table 7.1 that the onset point of the transition varies with the cylinder-channel ratio $\beta$; however, measurements indicate that the critical Deborah number increases by only 30% as the cylinder-channel ratio is decreased from $\beta = 0.50$ to $\beta = 0.26$, whereas the critical Weissenberg number is reduced by a factor of 2. These observations show that the onset point of the transition and the wavelength of the cellular structure scales more closely with the Deborah number and the cylinder radius; although a weaker modulation on the Weissenberg number and the channel gap is also evident. This corroborates the conclusions reached in Section 7.2, that the instability arises primarily from the strong extensional flow in the wake of the cylinder, not from the strong shearing flow between the cylinder and the channel wall.

A full evaluation of the $We/De$ parameter space contained in the stability diagram requires additional experiments for a range of different cylinder-channel ratios $\beta$. 
Figure 7.22  Centerline axial velocity profiles in the wake of the smaller cylinder ($\beta = 0.257$) show a pronounced downstream displacement as the Deborah number is increased. ($\Theta$) $De = 0.14$; (---) $De = 1.74$; (---) $De = 3.51$
Figure 7.23  A stability diagram for viscoelastic flow past circular cylinders of radius $R$ in planar channels of half-width $H$. The flow in the wake of the cylinder is a steady two-dimensional stagnation flow at low Deborah numbers (hollow symbols) but develops a three-dimensional cellular structure at high $De$ (filled symbols).
7.4 Discussion of Results

The measurements presented in this Chapter are the first experimental observations of a three-dimensional elastic instability that occurs within the planar extensional flow in the wake of a circular cylinder. At a critical Deborah number the steady two-dimensional flow undergoes a bifurcation to a steady, three-dimensional motion consisting of a spatially-periodic cellular structure that extends along the length of the cylinder. The LDV measurements and video flow visualization (Appendix A) clearly show that this structure corresponds to cells of fluid that move with a local axial velocity that is higher than the average value. To satisfy continuity constraints these regions are supplied with fluid from neighboring slower moving cells on either side. The flow remains symmetric about the channel centerline (υ = 0) and there is no measurable u_y velocity component in this plane (within the resolution of the three-color LDV system). It is thus possible to define streamlines for this two-dimensional planar region, and Figure 7.24 shows a sketch representing the fluid streamlines along the central plane of the channel. Near the cylinder at ζ = 1.5 the streamlines are curved, and the flow has both u_x and u_y components as fluid alternately converges into the faster moving regions (corresponding to the brighter regions in Plate 7.1(b)) and diverges from the slower moving areas. A profile of the axial velocity component (u_x) at the line ζ = 1.5 shows periodic oscillations with a wavelength λ_x ~ R/2. However, a profile of the u_x velocity component would show periodic oscillations about zero with a wavelength of λ ~ R as fluid diverges from the slower moving regions into the faster-moving cells. Further downstream for ζ > 2.0 the streamlines are almost parallel to each other, and the wake consists of a regularly spaced cellular structure of wavelength λ_x ~ R. At these axial distances the velocity u_x is negligible and profiles of the axial velocity show periodic oscillations of wavelength λ ~ R. Hence, the sketch shown in Figure 7.24 provides a consistent explanation of the complex spatial structure of the velocity field in the cylinder wake that was documented by the LDV velocity profiles presented in Figures 7.11 - 7.13.

Very recently Chiba et al. (1990) have presented qualitative flow visualization results that reveal the onset of a similar transition in the viscoelastic flow of a dilute polyacrylamide solution through a 10:1 planar contraction. Streak photographs of longitudinal cross-sections across the contraction (i.e. along the neutral axis) showed that, at a critical flow rate, the planar extensional flow through the contraction became unstable and developed a three-dimensional structure. This three-dimensional motion consisted of an approximately periodic array of 'bundle-like streams' arising from faster and slower moving regions of fluid that were spaced along the neutral axis of the planar contraction. At
Figure 7.24 An approximate representation of the flow-stream lines along the channel mid-plane ($v = 0$) showing the spatial evolution of the velocity profiles and the development of a cellular structure in the cylinder wake.
higher flow rates these structures were also observed to slowly travel across the width of the contraction. Few quantitative details are provided in this paper; however, the work is the first indication of the development of a planar to cellular transition in viscoelastic flow. No critical Deborah number for the onset of this instability was provided, but the high flow rates result in shear-thinning of the fluid viscosity and Reynolds numbers of $17 \leq Re \leq 53$ in the unstable flow regime. Therefore, inertial effects in this flow must also be important.

A transition that results in anomalous transport properties (such as the heat transfer and drag coefficients) for the flow of dilute polymer solutions past very small cylinders has previously been observed experimentally (James and Acosta, 1970). These experimental results have recently been reproduced numerically by Hu and Joseph (1990) and Delvaux and Crochet (1990). The numerical simulations considered high Reynolds number flow past a cylinder for an Upper-Convected Maxwell (UCM) fluid model and showed that the asymptotic behavior of the transport properties observed by James and Acosta corresponded to a change of type in the governing equation set. The theoretical analysis of Ultmann and Denn (1971) discussed in Section 2.3.2 was the first to consider the possibility of a change of type in viscoelastic flow around a cylinder and showed that it corresponded to a velocity which exceeded the shear-wave speed for the UCM model, $c = \eta_0/\rho \lambda_0$ (see equation 2.16). To interpret their results, Hu and Joseph introduced the concept of a viscoelastic Mach number $M = (v_2)/c$; for $M < 1$ the governing equation set for viscoelastic flow of the UCM model is elliptic, and the velocity solutions are smooth everywhere. However, for $M \geq 1$ (i.e. for velocities greater than the wave speed) the equation set becomes hyperbolic and discontinuities, or shocks, can propagate along the fluid streamlines.

In order to investigate whether the experimental measurements of the elastic wake instability presented above correspond to a change of type in the flow, it is necessary to calculate the shear-wave speed $c$ for the test fluid. The rheological characterization presented in Chapter 4 has shown that the viscoelastic material functions of the 0.31 wt% PIB/PB/C14 Boger fluid are not accurately described by a single-mode, quasi-linear constitutive equation such as the UCM model, and it is not appropriate to use the simple expression above to calculate the wave speed. Recently, Northey (1991) has presented a type analysis for a multimode formulation of the UCM model and has shown that the critical wave speed for propagation of velocity information is

$$c = \sqrt{\frac{1}{\rho} \sum_{k=1}^{N} \frac{\eta_k}{\lambda_k}}$$  (7.7)
where \( \eta_k \) and \( \lambda_k \) are the viscosity and relaxation time of the \( k \)th mode and \( \rho \) is the fluid density.

Equation (7.7) is evaluated for the 0.31 wt% PIB Boger fluid by using the set of \( \{ \eta_k, \lambda_k \} \) determined from the linear viscoelastic measurements presented in Section 4.4.1. However, it can be seen from eq. (7.7) that the presence of a purely Newtonian solvent with \( \lambda_s = 0 \) results in an infinite wave-speed and prohibits a change-of-type in the governing equation set. The high shear-rate experiments in a capillary rheometer presented in Figure 4.15 have shown that, in reality, the highly viscous PB/C14 solvent is not Newtonian, but very weakly elastic with a relaxation time of \( \lambda_s \approx 10^{-4} \) s. By combining this estimate for the PB/C14 solvent relaxation time with the four mode fit given in Table 4.2, the speed of shear-waves through the Boger fluid is calculated to be \( c = 9.61 \) m/s. Joseph et al. (1986) have developed a ‘wave-speed meter’ to determine wave-speeds for a large number of different liquids and the value calculated above agrees extremely well with the range of values \( 8.37 \leq c \leq 22.40 \) m/s measured by Joseph et al. for a Boger fluid consisting of 0.25 wt% PIB in polybutene. The calculated value of \( c = 9.61 \) m/s is far greater than the values of the critical velocity \( \langle u_2 \rangle_c \) given in Table 7.1 and calculation of the ‘viscoelastic Mach number’ at the onset of the elastic wake instability reveals it to be only \( M = 3.63 \times 10^{-3} \) for \( \beta = 0.50 \), \( M = 2.44 \times 10^{-3} \) for \( \beta = 0.25 \). Hence, the transition to three-dimensional flow is not a change-of-type phenomenon and is completely unrelated to the high \( Re \) experiments of James and Acosta (1970) and the related LDV measurements of Koniuta et al. (1980).

The LDV measurements of Bisgaard (1983) for viscoelastic flow past a sphere falling through a cylindrical tube have shown a similar elastic instability at low \( Re \) that results in spatial fluctuations of the velocity in the wake. A representative centerline velocity profile from Bisgaard’s work has been presented in Figure 2.23 and shows that the velocity in the uniaxial extensional flow does not decrease monotonically but develops rapid aperiodic fluctuations. In contrast, the LDV measurements presented above show that profiles of the velocity in the wake vary smoothly and monotonically in the downstream direction (see Figures 7.16 and 7.22), and the instability in the cylinder wake results in a transition to a three-dimensional flow with a well-defined periodic spatial structure. Unfortunately, insufficient information is provided to infer if the instability observed by Bisgaard also results in a three-dimensional flow and an associated loss of axisymmetry in the wake of the sphere. Bisgaard also reported the presence of velocity fluctuations in the strong shearing flow between the sphere and the tube walls, which develop at Deborah numbers lower than those required for the formation of aperiodic fluctuations in the wake.
of the sphere. No similar fluctuations were observed in LDV measurements near the cylinders studied in this thesis and the three-dimensional flow was limited to the region of strongly extensional flow in the wake of the cylinder. The elastic instabilities observed in the wakes of cylinders and spheres therefore appear to be fundamentally different in both their spatial and temporal characteristics.

Of course, this is subject to the important caveat that the experiments of Bisgaard were performed with a shear-thinning polymer solution rather than a constant viscosity Boger fluid and for smaller values of the sphere-tube ratio in the range $0.06 \leq \beta \leq 0.18$. The experimental observations in axisymmetric contractions presented by Boger et al. (1987) and McKinley et al. (1991b) have shown that both the fluid rheology and relevant dimensionless geometric parameters such as $\beta$ are extremely important in governing the precise dynamic behavior associated with the onset of a viscoelastic flow transition. It is, however, clear from the LDV measurements of Bisgaard (1983) and those presented in this thesis that both uniaxial and planar extensional flows of viscoelastic fluids near the downstream stagnation points of submerged bodies can become unstable at high Deborah numbers. The important distinctions between the spatial and temporal structures of the velocity fields in each case are very similar to the differences between planar and axisymmetric contraction flows that have been discussed in Section 2.3 and 6.5, and it appears that the base symmetry of the problem is crucial in studies of viscoelastic flow instabilities. Further discussion of these ideas is presented in the general conclusions of Chapter 8.

The velocity measurements of Sigli and Coutanceau (1977) and Bisgaard (1983) for shear-thinning polymer solutions have both documented the existence of a 'negative wake' in the uniaxial extensional flow behind a sphere. In a laboratory frame of reference – in which the sphere is stationary – this corresponds to a 'velocity overshoot' (see Figure 2.21) that is similar to that observed in the extensional flow along the centerline of axisymmetric contractions. This velocity overshoot develops at low Deborah numbers and is not associated with the onset of time-dependent flow. However, the LDV measurements in the cylinder wake for the 0.31 wt% PiB Boger fluid show that the velocity monotonically increases from zero to the free stream centerline value $v_x = 1.5\langle v_x \rangle$ at all Deborah numbers. Maalouf and Sigli (1984) used streak photography to show that the velocity profiles measured for a Boger fluid in the wake of a sphere were shifted downstream at high $De$ but remained monotonic; however similar measurements of the velocity profiles for aqueous polymer solutions indicated the presence of a velocity overshoot and a negative wake behind the sphere. These observations are consistent with
the LDV measurements presented in this thesis and it is concluded that a negative wake is only observed in viscoelastic fluids which exhibit a shear-thinning viscosity $\eta(\gamma)$.

The recent work of Chmielewski et al. (1990) investigated the elastic dependence of the drag coefficient $C_D(De)$ for spheres falling through PAC/CS and PIB/PB Boger fluids with the same viscometric properties $\eta, \Psi_1$. Measurements showed that the PAC/CS fluid exhibited a monotonic decrease in the drag coefficient below the Newtonian value $C_D^0$ given in eq. (2.9), in agreement with the comprehensive earlier results of Chhabra et al. (1980) and Mena et al. (1987). However, measurements for the PIB/PB Boger fluid showed a very small initial decrease in the ratio of drag coefficients $C_D(De)/C_D^0$ for $De \leq 0.3$, followed by a larger drag increase at higher $De$. At the maximum attainable Deborah number of $De = 1.6$ the drag was found to be increased 20% above the Newtonian value. Similar results for another PIB/PB fluid formulation have also been presented by Tirtaatmadja et al. (1990), and it is plausible that the pronounced increase in the drag coefficient observed for $De > 1$ arises from the onset of a similar elastic wake instability in the axisymmetric stagnation flow behind the sphere.

The dissimilar behavior of the two Boger fluids indicates that the molecular environments of the PIB and PAC macromolecules in solution are not the same, even though rheological measurements of the viscometric properties in steady shear flow are very similar. The stagnation flow in the wake of the sphere is a strong uniaxial extensional flow and it thus seems that the differences in drag behavior must result from different elongational properties of the PIB and PAC chains. The extensive experimental results of Boger and coworkers (1986, 1987) also reveal differences in the sequence of viscoelastic flow transitions observed in the strongly extensional flows of PIB/PB and PAC/CS fluids through axisymmetric abrupt contractions. It therefore appears that the stability of complex flows of Boger fluids is a sensitive function of the elongational properties of the long polymer molecules in solution. The discussion in Chapter 6.5 and the stability diagrams sketched in Figure 6.24 provide a rational interpretation of the differences observed between the dynamic behavior of PAC/CS and PIB/PB Boger fluids in terms of the particular ordering of a sequence of nonlinear hydrodynamic transitions. A similar dependence of the critical onset conditions and precise dynamic behavior is thus to be expected in observations of the elastic wake instability (if it exists) for PAC/CS Boger fluids. To explore this variation further, additional LDV measurements are required for a PAC/CS fluid with the same rheological properties as the 0.31 wt% PIB/PB Boger fluid employed in this research.

The LDV technique has proved to be an extremely powerful tool for documenting the temporal and spatial characteristics of the elastic wake instability. However, the
quantitative results presented in this chapter have only shown one-dimensional ‘slices’ of the complex three-dimensional kinematics that develop in the wake of the cylinder. The direct flow visualization results contained in the attached video-tape (Appendix A) provide a much clearer picture of the velocity field in the wake. However, these images are purely qualitative. The development of experimental techniques which allow the extraction of quantitative kinematic information from such two-dimensional images is an important next step in the documentation of three-dimensional flow instabilities. Optical and digital image processing techniques such as holography and particle image velocimetry (PIV) are two methods that enable rapid evaluation of global velocity fields, and holographic velocimetry has recently been applied to the study of the inertial instability of a Newtonian fluid in the wake of cylinder (Stanislas, 1985). The benefits and drawbacks associated with these approaches have been discussed in Chapter 1, and they complement rather than replace the point-wise measurements provided by LDV. The application of such techniques to the elastic wake instability documented in this Chapter would provide a more detailed understanding of the three-dimensional velocity field in the wake of the cylinder.
Chapter 8

Conclusions

The preparation and characterization of a highly elastic fluid with an almost constant viscosity has permitted the study of elastic effects on the stability of complex flows in the absence of additional complicating factors such as shear-thinning or inertial effects. The experimental results presented in this thesis have documented the spatial and temporal structures of elastic flow instabilities in three separate test geometries which are commonly encountered in commercial polymer processing applications:

• Dynamic measurements of the torque and normal force exerted on the plates of standard cone-and-plate and parallel plate test fixtures have identified a subcritical Hopf bifurcation from steady axisymmetric rotational shear flow to a complex three-dimensional, time-dependent motion. Stability measurements for a wide range of shear rates, rotational speeds and disk sizes have enabled the construction of a stability diagram for rotational flows of Boger fluids which shows that the instability is a sensitive function of the rotation rate \( \Omega \) and not of the shear rate \( \dot{\gamma} \). Flow visualization has shown that the resulting unsteady flow consists of recirculating spiral vortices with a wide range of characteristic wavelengths that propagate radially across disks.

• Laser Doppler velocimetry measurements of the velocity field in the axisymmetric abrupt contraction have documented a series of nonlinear transitions commencing with a supercritical Hopf bifurcation to three-dimensional time-dependent flow, followed by subsequent transitions to period-doubling, quasiperiodic and ultimately aperiodic flow regimes. Measurements for six different contraction ratios and two different lip entrances have permitted a systematic exploration the large variations in dynamics associated with small changes in flow geometry, and the construction of an approximate stability diagram for contraction flows of Boger fluids. By coupling qualitative video-imaging techniques with highly accurate LDV measurements the local dynamic behavior of the fluid near the contraction lip has been linked with the evolution
of the vortex structure that is observed macroscopically in axisymmetric contraction flows.

- Combined flow visualization and LDV measurements have documented for the first time an elastic instability in the wake of a circular cylinder confined in a rectangular channel. The transition leads to a bifurcation from steady, planar extensional flow in the wake and the formation of a spatially periodic three-dimensional flow that extends along the symmetry axis of the cylinder. The spatial extent of this cellular structure is limited to the strongly extensional flow near the downstream stagnation point and the wavelength is found to scale more closely with the cylinder radius than the gap between the cylinder and the channel walls.

In each geometry studied the onset of a flow transition is found to occur at moderate Deborah numbers of $De \sim 1$ and very small Reynolds numbers of $Re < 0.1$. These transitions are entirely absent in the corresponding flows of Newtonian liquids at equivalent $Re$ and the onset of these instabilities is associated unequivocally with the elastic properties of the polymer solution. The first direct kinematic measurements of the nonlinear dynamics in a viscoelastic flow transition were provided by Muller (1986) in the flow of a PIB/PB Boger fluid through a 4:1 axisymmetric contraction. The experimental studies presented in this thesis have shown that instabilities in viscoelastic flow are not restricted to such a specific fluid/geometry combination but in fact appear to be ubiquitous and occur in a number of flows that are of industrial relevance, including rotational flows, entry flows and stagnation flows. The onset of such instabilities in the flow of viscoelastic materials has a direct impact on commercial polymer processing operations: The complex three-dimensional, time-dependent velocity fields will result in the development of local inhomogeneities in the molecular orientation and stress history of the polymeric material. Such microscopic defect structures severely compromise the mechanical integrity and ultimate value of the finished products.

An overview of current knowledge regarding the nonlinear hydrodynamics of viscoelastic flow instabilities can be obtained by uniting the experimental measurements presented in this thesis with the few previous detailed observations of flow transitions in polymeric liquids. Although the list shown below in Table 8.1 is not yet comprehensive enough to span all the model problems that have been discussed in the literature, it is possible to draw some tentative conclusions about the stability of viscoelastic flows in complex geometries. Most importantly, the base symmetry of the problem appears to be crucial in defining the spatial and temporal structure of viscoelastic flow transitions. In
axisymmetric geometries, elastic effects lead to the onset of Hopf bifurcations at \( De \sim 1 \) and the development of flow regimes that are both time-dependent and three-dimensional. As the Deborah number is increased, subsequent transitions lead to period-doubling, quasi-periodic and finally aperiodic states. By contrast, the few experimental investigations of viscoelastic flow instabilities in planar geometries indicate that the first flow transition is a bifurcation from a steady two-dimensional flow to a steady, three-dimensional flow that has a periodic, cellular structure in the 'neutral' direction. At higher \( De \) a second flow transition results in the development of time-dependent flow as this periodic structure begins to translate along the direction of the neutral axis.

<table>
<thead>
<tr>
<th>Axisymmetric Flows</th>
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</tr>
</thead>
<tbody>
<tr>
<td><strong>Geometry</strong></td>
<td><strong>Author</strong></td>
<td><strong>Fluid</strong></td>
<td><strong>Instability Type</strong></td>
</tr>
<tr>
<td>Sphere in a Tube</td>
<td>Bisgaard (1983)</td>
<td>PAC/glycerin</td>
<td>Spatiotemporal fluctuations in wake of sphere</td>
</tr>
<tr>
<td>Axisymmetric Contraction</td>
<td>Nguyễn and Boger (1979)</td>
<td>PAC/glucose</td>
<td>Time-dependent spiralling oscillations of large vortex</td>
</tr>
<tr>
<td></td>
<td>Muller (1986)</td>
<td>PIB/PB/K</td>
<td>3-D, time-dependent flow near contraction lip.</td>
</tr>
<tr>
<td></td>
<td>McKinley et al. (1991b)</td>
<td>PIB/PB/C14</td>
<td>3-D flow with a sequence of transitions to periodic, quasi-periodic, aperiodic regimes.</td>
</tr>
<tr>
<td></td>
<td>McKinley et al. (1991)</td>
<td>PIB/PB/C14</td>
<td>Subcritical Hopf bifurcation to three-dimensional aperiodic flow.</td>
</tr>
<tr>
<td>Taylor-Couette Flow</td>
<td>Larson et al. (1990)</td>
<td>PIB/PB/K</td>
<td>Supercritical Hopf bifurcation to time-dependent cellular structure.</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Rectilinear Flows</th>
<th></th>
<th></th>
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</tr>
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<tbody>
<tr>
<td><strong>Geometry</strong></td>
<td><strong>Author</strong></td>
<td><strong>Fluid</strong></td>
<td><strong>Instability Type</strong></td>
</tr>
<tr>
<td>Planar Contraction</td>
<td>Chiba et al. (1990)</td>
<td>PAC/water</td>
<td>Periodic array of 'bundle-like' streams followed by travelling cellular structure</td>
</tr>
<tr>
<td>Cylinder in Planar Channel</td>
<td>McKinley (1991)</td>
<td>PIB/PB/C14</td>
<td>Steady cellular structure followed by travelling waves</td>
</tr>
</tbody>
</table>

**Table 8.1** Spatial and temporal structures of viscoelastic flow instabilities in axisymmetric and planar test geometries.
The dynamic behavior documented in each of the systems studied in this thesis is found to be extremely sensitive to dimensionless geometric parameters; i.e. the aspect ratio of the plates $R/H$, the contraction ratio $\beta$, and the cylinder-channel ratio $\beta$ respectively. In each case it has been shown that this complex dependence can be represented by the construction of a stability diagram which provides a consistent and rational explanation of the flow phenomena observed in the system. These results represent one of the first applications of concepts from nonlinear hydrodynamic stability theory to experimental studies of non-Newtonian fluid flow. The construction of similar stability diagrams for the flow of other viscoelastic materials — from theoretical considerations and further experimental measurements — is important in defining and evaluating stable operating regimes for viable commercial processes.

The experimental work in this thesis has shown that viscoelastic flow transitions occur in a number of complex geometries; however the results have only been presented for a single highly-elastic fluid consisting of 0.31 wt% PIB in a solution of PB/C14. Further work is required to understand the precise role of the fluid rheology on the stability of the flow. Previous theoretical and experimental work has shown that generally, shear-thinning effects stabilize viscoelastic instabilities (Cable and Boger, 1979; Phan-Thien, 1983; Lagnado et al., 1985), and that the addition of polymer additives inhibits the onset of inertial instabilities (Beavers and Joseph, 1974; Larson, 1989; Kim and Telionis, 1989). However, little is known about the importance of extensional effects on flow stability. Experiments with polyacrylamide- or polyisobutylene-based Boger fluids in identical test geometries have revealed significant differences in the evolution of macroscopic quantities, such as the drag coefficient (Chmielewski et al., 1990) or vortex size (Boger, 1987), with $De$. Since the viscometric properties $\eta$ and $\Psi_1$ of these two fluids are very similar, the deviations must arise from differences in the extensional rheology of the PAC and PIB solutions. However, no quantitative comparisons of the local velocity fields in the PAC and PIB Boger fluids have been presented in either of these previous studies and it is not known how variations in the elongational properties influence flow transitions to time-dependent or three-dimensional regimes. Qualitative ideas have been presented in Chapter 6.5 that propose plausible stability diagrams for contraction flows of PIB/PB and PAC/CS Boger fluids, and a specific ordering of the sequence of flow transitions for each fluid. Additional LDV measurements of the velocity fields and the flow stability of PAC/CS fluids in the axisymmetric contraction geometry and the constrained cylinder geometry are required to verify or refute these arguments.

The complementary optical techniques of flow visualization and three-color laser Doppler velocimetry that have been employed provide both qualitative visual confirmation
and quantitative determination of the nonlinear dynamics associated with complex flows of viscoelastic liquids. These experimental tools are extremely versatile and, in principle, can be adapted to any other flow geometry of interest. The video-imaging system allows rapid visual elucidation of the global flow field; however, to date, these observations have been purely qualitative. It is envisaged that in the future the integration of this system with modern digital image processing techniques such as Particle Image Velocimetry (PIV) will make it possible to obtain rapid quantitative knowledge of the global velocity field. Quantitative experimental measurements of both the steady-state and time-dependent kinematics of complex viscoelastic flows can then be made on a global and point-wise basis with sufficient accuracy to permit critical verification of theoretical constitutive equations and numerical algorithms for the flow of polymeric liquids.
Chapter 9

References


Appendix A

Videotape

A videotape entitled "The Nonlinear Dynamics of Viscoelastic Flows in Complex Geometries" has been submitted with this thesis. The tape is a 30 minute $\frac{1}{2}$" VHS format videocassette recorded in standard NTSC format with no audio track. The tape has a program length of 25 minutes and consists of a sequence of descriptive text titles plus real-time footage recorded using the flow visualization procedure discussed in Chapter 3.

A track listing is given below. (T) indicates a text page describing the following section of video footage (V).

<table>
<thead>
<tr>
<th>Time</th>
<th>Type</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>00:00</td>
<td></td>
<td>Color bar for monitor calibration</td>
</tr>
<tr>
<td>01:30</td>
<td>T</td>
<td>&quot;The Elastic Instability in Parallel-Plate Flow of a Boger Fluid&quot;</td>
</tr>
<tr>
<td>01:38</td>
<td>T</td>
<td>Credits</td>
</tr>
<tr>
<td>01:45</td>
<td>T</td>
<td>Parallel-Plates, Side View, $De = 5.73$, $We = 39.7$</td>
</tr>
<tr>
<td>01:50</td>
<td>T</td>
<td>Graph showing time-dependent response of the first normal stress difference $N_{10}(t)$ [Pa]</td>
</tr>
<tr>
<td>02:00</td>
<td>V</td>
<td>Side view of flow in RMS-800 mechanical spectrometer</td>
</tr>
<tr>
<td>04:00</td>
<td>T</td>
<td>Spatial Structure of Instability</td>
</tr>
<tr>
<td>04:07</td>
<td>T</td>
<td>$De = 3.31 - 4.35$; $We = 26.1 - 34.4$</td>
</tr>
<tr>
<td>04:15</td>
<td>V</td>
<td>Plan view of flow between parallel-plates. Rotation rate and $De$ increases at $t = 01:00$ min. (on screen).</td>
</tr>
<tr>
<td>06:45</td>
<td></td>
<td>End</td>
</tr>
<tr>
<td>07:10</td>
<td>T</td>
<td>&quot;Nonlinear Dynamics of Viscoelastic Flow in Axisymmetric Abrupt Contractions&quot;</td>
</tr>
<tr>
<td>07:22</td>
<td>T</td>
<td>Credits</td>
</tr>
<tr>
<td>07:26</td>
<td>T</td>
<td>Fluid Composition</td>
</tr>
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</table>

359
<table>
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<tr>
<th>Time</th>
<th>Event Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>07:35</td>
<td>T 4:1 Contraction, Newtonian Flow; $De = 0.42; Re = 0.004$</td>
</tr>
<tr>
<td>07:40</td>
<td>V Global View of Velocity Field</td>
</tr>
<tr>
<td>08:05</td>
<td>V Close-Up of Corner Recirculation</td>
</tr>
<tr>
<td>08:19</td>
<td>T 4:1 Contraction, Time-Dependent Flow; $De = 2.11, Re = 0.025$</td>
</tr>
<tr>
<td>08:25</td>
<td>V Close-Up of Corner Vortex</td>
</tr>
<tr>
<td>08:46</td>
<td>T 4:1 Contraction, Lip Vortex; $De = 3.48, Re = 0.043$</td>
</tr>
<tr>
<td>08:52</td>
<td>V Time-Dependent Lip Vortex</td>
</tr>
<tr>
<td>09:26</td>
<td>T 4:1 Contraction, increasing Flow Rate; $De = 0.70 - 4.50$</td>
</tr>
<tr>
<td>09:34</td>
<td>V Step increase in flowrate (and $De$) at $t = 01:00$ min (on screen)</td>
</tr>
<tr>
<td>11:07</td>
<td>V Global view of pulsating elastic vortex</td>
</tr>
<tr>
<td>12:40</td>
<td>T Effect of Contraction Ratio</td>
</tr>
<tr>
<td>12:45</td>
<td>T 5:1 Contraction; $De = 0.74 - 4.66, Re = 0.007 - 0.091$</td>
</tr>
<tr>
<td>12:55</td>
<td>V Step increase in flowrate (and $De$) at $t = 01:00$ min (on screen)</td>
</tr>
<tr>
<td>14:30</td>
<td>T Effect of Lip Curvature, 4:1 Contraction Ratio</td>
</tr>
<tr>
<td>14:35</td>
<td>T No Lip Vortex; $De = 3.48, Re = 0.058</td>
</tr>
<tr>
<td>14:40</td>
<td>V Close-up of curved entry region</td>
</tr>
<tr>
<td>15:05</td>
<td>T Lip Vortex forms; $De = 4.70, Re = 0.108</td>
</tr>
<tr>
<td>15:15</td>
<td>V Small unsteady lip vortex</td>
</tr>
<tr>
<td>15:45</td>
<td>T Steady Vortex Growth; $De = 4.91, Re = 0.124</td>
</tr>
<tr>
<td>15:55</td>
<td>V Large steady elastic vortex</td>
</tr>
<tr>
<td>16:20</td>
<td>T Increasing Flow Rate; $De = 0.47 - 5.05, Re = 0.0</td>
</tr>
<tr>
<td>16:28</td>
<td>V Step increase in flowrate (and $De$) at $t = 01:00$ min (on screen)</td>
</tr>
<tr>
<td>18:10</td>
<td>End</td>
</tr>
</tbody>
</table>

18:30  T "Viscoelastic Flow Around a Constrained Cylinder"

18:40  T Cylinder Radius $R = 3.188$ mm, Channel Depth $H = 12.70$ mm.

18:50  T Side View; $De = 0.78, Re = 0.0055$

19:00  V Side view of flow near upstream stagnation point

19:20  V Side view of flow near downstream stagnation point

19:40  T Front View; $De = 0.78, Re = 0.0055$

19:50  V view along the length of the cylinder

20:10  T Transition to Cellular Structure in Cylinder Wake

$De = 0.43 - 2.48, Re = 0.0027 - 0.021$

20:20  V Step increase in flowrate (and $De$) at $t = 01:00$ min (on screen)
Detail of Three-dimensional Flow in the Wake

\( De = 0.73 - 2.48, \ Re = 0.0055 - 0.021 \)

Step increase in flowrate (and \( De \)) at \( t = 01:00 \) min (on screen)

Structure Extends Along Length of Cylinder; \( De = 24.8, \ Re = 0.021 \)

Overall View of Global Velocity Field

Spatial Extent of Periodic Wake; \( De = 1.50, \ Re = 0.012 \)

Velocity Field at \( y/R = 0.4 \)

sheet of laser light at \( y/R = 0.4 \)

Velocity Field at \( y/R = 1.1 \)

sheet of laser light at \( y/R = 1.1 \)

Return to Centerline, \( y/R = 0.0 \)

move sheet of laser light from \( y/R = 1.0 \rightarrow 0.0 \)

End