Characteristics and origins of viscoelastic turbulence in a 3-stream contraction-expansion micro-channel

Tai, Jiayan

2016

Tai, J. (2016). Characteristics and origins of viscoelastic turbulence in a 3-stream contraction-expansion micro-channel. Doctoral thesis, Nanyang Technological University, Singapore.

<https://hdl.handle.net/10356/66041>

<https://doi.org/10.32657/10356/66041>

**NANYANG
TECHNOLOGICAL
UNIVERSITY**

**CHARACTERISTICS AND ORIGINS OF
VISCOELASTIC TURBULENCE IN A 3-STREAM
CONTRACTION-EXPANSION MICRO-CHANNEL**

TAI JIAYAN

SCHOOL OF MECHANICAL AND AEROSPACE ENGINEERING

2016

CHARACTERISTICS AND ORIGINS OF
VISCOELASTIC TURBULENCE IN A 3-STREAM
CONTRACTION-EXPANSION MICRO-CHANNEL

TAI JIAYAN

2016

**CHARACTERISTICS AND ORIGINS OF
VISCOELASTIC TURBULENCE IN A 3-STREAM
CONTRACTION-EXPANSION MICRO-CHANNEL**

TAI JIAYAN

TAI JIAYAN

School of Mechanical and Aerospace Engineering

A thesis submitted to the Nanyang Technological University
in partial fulfilment of the requirement for the degree of
Doctor of Philosophy

2016

Abstract

At low Reynolds number ($Re < 1$), the flow of viscous liquids e.g. water, is laminar. An aqueous solution e.g. water becomes viscoelastic when a small amount of polymer additives (< 1 wt%) is added to it; its flow behavior can become drastically different and turbulent i.e. viscoelastic turbulence. This phenomenon has gained increasing attention because it violates the conventional school of thought i.e. high- Re criteria, for creating chaos and disorder in a fluid dynamics system. As the polymer molecules are invisible, the indirect deduction of molecular behavior via motion analyses of tracer additives has been adopted as the main investigative approach in viscoelastic turbulent flows. Based on this approach, reported works attribute viscoelastic turbulence to the release of elastic energy by the polymer molecules, which had been extended due to strong velocity gradients in the flow field. The release of energy occurs over a range of time scales which is dependent on the characteristic time scales of the molecules and bulk viscoelastic liquid. Although the reported characteristic time scales vary significantly, their effects on the structure of the viscoelastic turbulent flow field have not been investigated.

Despite the significant number of investigations based on the conventional approach, the underlying mechanisms in viscoelastic turbulence remains elusive; several outstanding questions on viscoelastic flows remain unresolved e.g. the high Weissenberg number problem, and the drag reduction theory debate. “How do the polymer molecules change the flow field so drastically when they are only present in minute amounts?”. This fundamental question has yet to be clearly answered. Although fluorescent-tagged DNA molecules are commercially available, because

they are costly and impractical for large-scale dynamic flow field investigations, their usage has been mostly limited to tracer additives or stress probes.

Here, a viscoelastic turbulent flow field generated by a 3-stream flow through a contraction-expansion micro-channel was investigated. Based on the conventional approach i.e. tracer additives, the results show that the generated turbulence exhibits characteristics of 2-D turbulent flows, from which information on the molecular and bulk properties of the viscoelastic liquid could be extracted. In addition, a high-speed molecular imaging technique was developed to observe the polymer molecular conformation changes (i.e. extension and relaxation) in a turbulent flow. The technique is capable of image grabbing speeds of more than 1000 frames per second. The developed molecular fluorescein tagging technique has a much lower cost as compared to fluorescein-labeling of DNA, hence making it more feasible for large scale investigations. With the developed technique, the dynamic molecular conformation changes could be correlated with the statistical properties of the flow field obtained based on the conventional approach.

Acknowledgements

I would like to express my heart-felt gratitude to my supervisor, Professor Lam Yee Cheong. His advice and guidance has been invaluable throughout my Ph.D. study.

In addition, I would like to thank NTU for the funding and support, which made this research work possible. The excellent research environment has greatly facilitated the experimental work required in this study.

I would also like to thank all staffs of Materials Laboratory 1, Thermal and Fluids Laboratory and Precision Engineering Laboratory for their support and help in the management and usage of facilities.

Most importantly, I wish to thank my parents and friends for their unwavering support.

Blank Page

Table of Contents

Table of Contents	I
List of Figures	VI
List of Tables	XIII
List of Abbreviations	XIV
List of Symbols	XVI
Chapter 1: Introduction	1
1.1 Background	1
1.2 Objectives	4
1.3 Outline	5
Chapter 2: Viscoelastic Turbulence	9
2.1 Introduction	9
2.1.1 Overview	9
2.1.2 Background	10
2.1.3 Outline	12
2.2 Dimensionless numbers	13
2.3 Viscoelastic contraction flows	15
2.3.1 Overview	15
2.3.2 Macro-channel Contraction flows	16
2.3.3 Micro-channel Contraction flows	18
2.4 Viscoelastic turbulent flows	19
2.4.1 Overview	19
2.4.2 Rotating flows	20
2.4.3 Cross-slot flows	23
2.4.4 Curvilinear flows	24
2.4.5 Contraction and/or expansion flows	28
2.5 Review of viscoelastic turbulent flows	32
2.5.1 Summary	32
2.5.2 Discussion	34
2.6 Thesis planning	36
Chapter 3: High-Re Turbulence	44
3.1 Introduction	44
3.1.1 Background	44
3.1.2 Outline	45
3.2 Governing equations	46

3.2.1	Navier-Stokes equation	47
3.2.2	Viscoelastic models	49
3.3	Viscoelastic turbulence in micro-channels	52
3.4	Characteristics of high-Re turbulent flows	54
3.4.1	3-D turbulence	54
3.4.2	2-D Turbulence	55
3.4.3	Characteristics and measurement techniques	57
3.5	Parameters in turbulence quantification	61
3.6	Review of recent works	67
3.6.1	Introduction	67
3.6.2	Brief review on 3-D turbulent flows	67
3.6.3	Brief review on 2-D turbulent flows	75
3.7	Summary	79
Chapter 4:	Rheology	81
4.1	Introduction	81
4.2	Viscoelasticity	82
4.2.1	Background	82
4.2.2	Stress response to step strain	84
4.2.3	Stress response to shear rate	84
4.2.4	Viscoelastic examples	85
4.3	Maxwell model	86
4.3.1	Introduction	86
4.3.2	Response to constant strain	87
4.3.3	Response to constant strain rate	89
4.3.4	Other viscoelastic liquid models	90
4.4	Viscoelastic fluid properties	91
4.4.1	Intrinsic viscosity	91
4.4.2	Critical overlap concentration	92
4.5	Rheology of test liquids	92
4.5.1	Polyethylene oxide (PEO)	92
4.5.2	Test liquids preparation	93
4.5.3	Steady shear rate tests	94
4.5.3.1	1-D shear flow	94
4.5.3.2	Shear viscosity measurements	96
4.5.4	Extensional tests	97

4.5.4.1	Extensional flow	97
4.5.4.2	Extensional relaxation time measurements	98
4.6	Summary	102
Chapter 5: Experimental Setup		104
5.1	Introduction	104
5.2	Micro-channel chip design	105
5.2.1	Schematic diagram	105
5.2.2	Calculation of dimensionless constants	106
5.3	Particle Image Velocimetry (PIV)	107
5.3.1	Introduction	107
5.3.2	Imaging setup	108
5.3.3	Recording modes	109
5.3.4	PIV algorithm	111
5.4	Image enhancement techniques	112
5.4.1	Introduction	112
5.4.2	Image enhancement algorithm	113
5.5	Summary	114
Chapter 6: Results - Flow Field Analyses		116
6.1	Introduction	116
6.1.1	Scope	116
6.1.2	Outline	116
6.2	Flow profile in a straight planar micro-channel	117
6.3	Experimental details	119
6.4	Mean velocity profiles	120
6.4.1	Region of interest	120
6.4.2	Cross-section profile at $\delta = 0.3$ mm	121
6.4.3	Cross-section axial velocity profile at $\delta = 1.0$ mm	123
6.4.4	Reynolds stresses	125
6.5	Single -point temporal velocity	127
6.5.1	Region of interest	127
6.5.2	Low frequency temporal axial velocity data	127
6.5.3	High frequency temporal axial velocity data	130
6.6	Correlation data	132
6.6.1	Single-point autocorrelation	132

6.6.2	Two-point (spatial) correlation	133
6.7	Spectral analysis	135
6.8	Flow field visualization	138
6.8.1	Vorticity and Galilean Decomposition	138
6.8.2	Proper Orthogonal decomposition (POD)	140
6.9	Polymer concentration and molecular weight	143
6.10	Summary	147
Chapter 7:	Results – Molecular Visualization	150
7.1	Introduction	150
7.1.1	Review	150
7.1.2	Outline	152
7.2	Fluorescein labeling	153
7.2.1	PEO labeling	153
7.2.2	An alternative labeling target - phPAA	154
7.2.3	Raman Spectroscopy	157
7.3	Static Molecular imaging	159
7.3.1	Imaging setup	159
7.3.2	Molecular visualization	159
7.3.3	Molecular size measurement	161
7.4	Experimental details	164
7.5	Dynamic molecular visualization	166
7.5.1	Analyses approach	166
7.5.2	Results	166
7.5.2.1	Aspect ratio	166
7.5.2.2	Molecular relaxation	169
7.5.2.3	Molecular size	169
7.5.3	Discussion	170
7.6	Flow field measurements (PIV)	172
7.6.1	Mean statistics	172
7.6.2	Power spectrum	174
7.7	Summary	178
7.7.1	Overview	178
7.7.2	Final remarks	181

Chapter 8: Conclusions 184

 8.1 Overview 184

 8.2 Future Works 190

References 193

Appendix A 207

Appendix B 210

Appendix C 212

List of Figures

Figure number	Page
Figure 2-1. Presence of corner vortices in flows of aqueous 0.2% PAA solution into a 4:1 contraction (Evans and Walters 1989)	16
Figure 2-2. Summary of the Wi-Re space diagram for flow transitions of different fluids through a 16:1:16 contraction-expansion (Rodd et al. 2005a)	19
Figure 2-3. a) Schematic setup of Groisman and Steinberg (2000) $R=38\text{mm}$, $R2=43.6\text{ mm}$, $d=10\text{ mm}$ b) Power spectra of velocity fluctuations using laser doppler anemometry for shear rates ① 1.25 s^{-1} , ② 1.85 s^{-1} , ③ 2.7 s^{-1} , ④ 4 s^{-1} , ⑤ 5.9 s^{-1}	21
Figure 2-4. a) Probability Density Function of fractional extension (x/L) of DNA molecules at the highest Wi for 5 concentrations. b) square of the peak fractional extension (solid squares) for 5 concentrations, average of the square of the fractional extension (open squares). c) log-log coordinates of data in b). (Liu and Steinberg 2010)	22
Figure 2-5. Flow induced birefringence in a cross-slot flow. Optical retardation at a) $Q = 1.0\text{ }\mu\text{L min}^{-1}$, $Wi = 1.0$, and b) $Q = 2.0\text{ }\mu\text{L min}^{-1}$, $Wi = 2.0$, $Re < 10^{-5}$. The colour scale represents optical retardation in the range a) $0 - 10\text{ nm}$ b) $0 - 30\text{ nm}$ (Haward et al. 2012).	24
Figure 2-6. a) Schematic diagram of channel. Note $R2=6\text{ mm}$, $R1=3\text{ mm}$ (Groisman and Steinberg 2001a) b) Power spectra of fluctuations at $Wi = 6.7$ for 1= longitudinal velocity, 2= transverse velocity.	25
Figure 2-7. a) Large fluctuations in longitudinal velocity b) Autocorrelation function for a) based on 6000 velocity measurements (Burghelea et al. 2004)	26
Figure 2-8. Plot of $(\partial V_{\theta}/\partial r)^{\text{rms}}$ against r/d at various Wi. The boundary layer corresponds to the region from the wall ($r/d = 0$) till the region where $(\partial V_{\theta}/\partial r)^{\text{rms}}$ reaches the maximum value (Jun and Steinberg 2011).	27
Figure 2-9. a) Schematic diagram of micro-channel with two inlets, with one stream containing fluorescent dye. b) negligible mixing near the inlet c) almost complete mixing (up to 80%) near the outlet. (Tatsumi et al.	

2011)	28
Figure 2-10. a) PIV image upstream of contraction, with average velocity plot at section A-A.. b) PIV image downstream of contraction, with average velocity plot at section B-B. The standard deviation is large in both plots. (Lam et al. 2009)	29
Figure 2-11. $De_{ratio} - De_{main}$ space diagram for various flow regimes. Black dot: $El_{main} \sim 7271$, Red hexagon: $El_{main} \sim 2011$ and Grey diamond: $El_{main} \sim 37.1$. (Gan and Lam 2012)	30
Figure 2-12. a) Power spectra of the velocity fluctuations at $x = 3.5$ cm at various flow rates. b) Polymer elongations for $Wi = 1$ and 4: Image 1 is near the outlet, and images 2–5 are further downstream at various distances from the channel wall. (Bonn et al. 2011)	31
Figure 3-1. Power spectrum of a typical 3-D turbulent flow and its energy ranges (Pope 2003).	55
Figure 3-2. Power spectrum of a typical 2-D turbulent flow showing the two inertial ranges (Boffetta 2007).	57
Figure 3-3. Visualization of an isolated turbulent eddy by Galilean decomposition by subtracting 3 different convection velocities (Adrian et al. 2000) a) $U = 0.65 U_{CL}$ b) $U = 0.78 U_{CL}$ c) $U = 0.91 U_{CL}$	60
Figure 3-4. Example of a single point temporal velocity measurement	62
Figure 3-5. a) Normalized pressure drop against Re showing departure at $Re \approx 2000$. b) Magnified portion of insert (Sharp and Adrian 2004)	68
Figure 3-6. Longitudinal velocity correlation function in a micro-channel having a hydraulic diameter of $640 \mu m$ (Li and Olsen 2006a)	68
Figure 3-7. Presence of turbulent structures in a turbulent flow for a square channel with a hydraulic diameter of $640 \mu m$ (Li and Olsen 2006b)	69
Figure 3-8. Deviation of friction factor-Reynolds number product at onset of turbulence (Rands et al. 2006)	70
Figure 3-9. Profiles of a) Cross-section flow profile b) Reynolds stress as a function of wall position from transition of laminar to turbulent flow (Natrajan and Christensen 2007)	71

Figure 3-10. Galilean decomposition at $Re = 2800$, with constant advection velocity $0.79 U_{cl}$ removed. U_{cl} = centreline velocity (Natrajan and Christensen 2007)	71
Figure 3-11. Perspective view of one extracted hairpin-like vortex 112.5 mm downstream of constriction at $Re = 460$ (Vetel et al. 2008)	72
Figure 3-12. a) Flow snapshot b) Distribution of kinetic energy across mode numbers c) POD mode 1 showing 4 pairs of counter rotating vortices. Meyer et al. (2008)	74
Figure 3-13. Comparison of statistics between PIV and LDV at 63% drag reduction. a) stream-wise turbulent intensity b) Reynolds shear stresses (Tamano et al. 2009)	75
Figure 3-14. Power spectra for soap films with different PEO concentrations showing power law scaling at different distances from the grid of cylinders a) Longitudinal and transverse velocity power spectra at a flux of 0.4 ml/s, 7 cm from grid. b) Thickness power spectra at a flux of 0.43ml/s, 7cm and 15 cm from grid (Amarouchene and Kellay 2002)	76
Figure 3-15. Energy spectrum from numerical simulations depicting the two inertial ranges for different spatial resolutions ($A= 2048$, $B= 4096$, $C= 8192$, $D= 16384$) (Boffetta 2007)	78
Figure 3-16. Velocity power spectra at $Wi= 26$ (squares) and $Wi= 45$ (circles) following a power law decay with an exponent of -3.8 (Berti et al. 2008)	79
Figure 4-1. An illustration of viscoelastic behavior of ‘silly putty’	82
Figure 4-2. Response of various materials to applied strain a) Applied constant strain b) Ideal viscous fluid response c) Ideal elastic solid response d) Viscoelastic material response	84
Figure 4-3. Stress response of various liquids as a function of shear rate ..	85
Figure 4-4. a) Weissenberg effect shown by a solution of polyisobutylene in polybutene. b) Die swell effect. (Barnes et al. 1989)	86
Figure 4-5. Schematic diagram of the Maxwell Model	87

Figure 4-6. Response of the Maxwell fluid to step strain. a) step strain b) stress response	87
Figure 4-7. Response of the Maxwell fluid to constant strain rate a) step strain rate b) stress response consisting of elastic and viscous phase	89
Figure 4-8. Schematic diagram of the a) Burgers model b) Jeffrey model.	90
Figure 4-9. a) schematic of 1-D shear flow. b) infinitesimal fluid element representation	95
Figure 4-10. Viscosity plot of test liquids under steady shear rate.	97
Figure 4-11. Schematic diagram of element undergoing simple extensional flow	98
Figure 4-12. Schematic of the sample during extensional tests in CaBER a) at rest b) during extension	99
Figure 4-13. Filament thinning of 0.3% PEO sample during extension. $t_{event} = 100$ ms (Rodd et al. 2005b)	99
Figure 4-14. Plot of filament diameter over time as measured by CaBER for the test liquids.	101
Figure 5-1. Schematic 2-D diagram of the micro-channel. Arrows depict the direction of flow, blue (side-stream), red (main-stream)	105
Figure 5-2. Schematic diagram of the experimental setup.	109
Figure 5-3. Schematic diagram of the two recording modes. a) mode 1. b) mode 2.	110
Figure 5-4. Conventional PIV image pair at two instances, with fluorescent particles being illuminated by a double pulsed laser.	112
Figure 5-5a) Original image b) Background image c) Processed image d) inverted image of c)	113
Figure 6-1. Axial velocity of water across the planar micro-channel	118
Figure 6-2. Schematic diagram of the contraction-expansion micro-channel, showing the plot of a mean axial velocity profile at a distance δ downstream of the contraction. Data in the shaded region has been	

removed due to high noise level.	120
Figure 6-3. a) Normalized axial velocity and b) Turbulent intensities at $\delta = 0.3$ mm	121
Figure 6-4. a) Normalized axial velocity and b) Turbulent intensities at $\delta = 1.0$ mm	123
Figure 6-5. a) Normalized axial velocity and b) Turbulent intensities at $\delta = 2.0$ mm	125
Figure 6-6. Reynolds stresses across the channel at $\delta = 0.3$ mm, 0.5 mm, 1.0 mm and 2.0 mm respectively.	126
Figure 6-7. Locations of interrogation windows A, B and C for single-point velocity measurements	127
Figure 6-8. Low frequency axial velocity measurements at a) interrogation window A ($\delta = 0.3$ mm) b) interrogation window B ($\delta = 0.5$ mm) and c) interrogation window C ($\delta = 1.0$ mm) obtained using recording mode 1. ..	129
Figure 6-9. High frequency axial velocity measurements at a) interrogation window A ($\delta = 0.3$ mm) b) interrogation window B ($\delta = 0.5$ mm) and c) interrogation window C ($\delta = 1.0$ mm) obtained using recording mode 2. ..	131
Figure 6-10. Temporal autocorrelation function obtained using setup 2. Inset shows the magnified view from 0 to 1 s.	132
Figure 6-11. Comparison of spatial correlation function based on calculations and Taylor's hypothesis.	133
Figure 6-12. Approximation of spatial correlation function based on Taylor's hypothesis.	134
Figure 6-13. Power spectrum of the axial velocity fluctuations at for various flow rates at a) interrogation window A ($\delta = 0.3$ mm), b) interrogation window B ($\delta = 0.5$ mm), and c) interrogation window C ($\delta = 1.0$ mm) respectively.	136
Figure 6-14. An instantaneous plot of velocity and vorticity (colour plot) downstream of the contraction from $\delta = 0.1$ mm to $\delta = 1.4$ mm. Insets a-c show structures at various time instances located in different regions, obtained using the Galilean decomposition method with a fraction of the	

convective velocity removed. a) $U - 0.6 \bar{U}$ b) $U - 0.6 \bar{U}$ d) $U - \bar{U}$	139
Figure 6-15. Various POD modes at $Q = 20$ ml/h from $\delta = 0.1$ mm to $\delta = 1.4$ mm a) POD mode 1. A large vortex the scale of the micro-channel width can be observed. b) POD mode 3. Upwards ‘sweeping’ motion. c) POD mode 4. Downwards ‘sweeping’ motion.	141
Figure 6-16. Relative percentage turbulent kinetic energy of the first 13 modes	143
Figure 6-17. Log of the power spectrum of velocity fluctuations for 20 ml/h, at interrogation window A.	144
Figure 6-18. Log-log plot depicting the relationship between the 2nd drop-off frequency and bulk liquid behaviour.	147
Figure 7-1. a) Raman spectrum of PAA, phPAA and phPAA-FITC. Significant bandwidth differences are labeled. The insert (b) shows an enlarged view of the C=S band.	158
Figure 7-2. Snapshots of 5 phPAA-FITC molecule pairs (a – e) showing the original 8-bit image and binary converted image	160
Figure 7-3. Images a) to c) show the snapshots of phPAA-FITC molecules when visualized under the excitation wavelength (left images), in comparison to snapshots obtained under white light illumination (right images).	161
Figure 7-4. Enlarged snapshots of the binary converted image in Figure 7-2a). The MATLAB algorithm fits an ellipse (green) around the phPAA-FITC molecule and measures the major axis (β) and minor axis (α) respectively.	162
Figure 7-5. Histogram plot of the size ranges for 1000 phPAA-FITC molecules measured under static conditions. Insets (a-d) show the snapshots of four molecules, and their corresponding sizes.	163
Figure 7-6. a) Polymer relaxation $(\beta/\alpha)_{\text{normalized}}$ over axial distance ($\delta = 0.1 - 1.5$ mm). Open and closed symbols represent polymers captured at 1 ml/h and 10 ml/h respectively. Significant relaxation occurs at 10 ml/h only. Insets show the relaxation of polymer molecule 9 at b) $\delta = 0.1$ mm and c) 1.0 mm respectively.	167

Figure 7-7. Normalized axial velocity (closed symbols) and turbulent intensities (open symbols) at a) $\delta = 0.3$ mm, and b) $\delta = 1.0$ mm.	172
Figure 7-8. Log-log power spectrum plot of velocity fluctuations for 10 ml/h, at $\delta = 1.0$ mm. The inset shows the power spectrum of the downstream flow field ($\delta = 1.0$ mm, black line) in comparison with the upstream vortex (blue line) from $\text{Log}_{10}(\text{frequency}) = 0.1 - 1.1$	176
Figure 7-9. . Log-log plot depicting the relationship between the 2nd drop-off frequency and bulk liquid behaviour. The data obtained in this section is circled in blue, showing a good agreement with the trend observed for the PEO molecules in Section 6.9.	177
Figure 7-10. Visualization of a single phPAA-FITC molecule flowing past the contraction. Note that the images belong to the same phPAA-FITC molecule. The green lines mark the boundaries of the micro-channel.	182
Figure 7-11. Analyses of the phPAA-FITC molecule length in Figure 7-10. The molecule had been stretched upstream before entering the contraction, and attained its maximum extended length at the singular point of the exit..	183

List of Tables

Table number	Page
Table 2-1. Conditions for onset of unstable flows in macro-contractions. ..	38
Table 2-2. Conditions for onset of unstable flows in micro-contractions. ...	39
Table 2-3. Conditions for onset of instability and quantification techniques used.	40
Table 4-1. Sample fluids composition	94
Table 4-2. Differences between Newtonian and Viscoelastic liquids	102
Table 4-3. Rheological properties of test liquids	102
Table 6-1. Experimental flow parameters and corresponding properties of 1.0%-2M in the contraction.	119
Table 6-2. Viscoelastic properties at various concentrations and molecular weights, with the corresponding cut-off frequencies	146
Table 7-1. Properties of the main-stream and side-stream liquid and the corresponding Re and De.	165

List of Abbreviations

0.1%-2M	0.1 wt% PEO (2×10^6 g/mol)
0.1%-8M	0.1 wt% PEO (8×10^6 g/mol)
0.3%-2M	0.3 wt% PEO (2×10^6 g/mol)
0.3%-8M	0.3 wt% PEO (8×10^6 g/mol)
0.5%-8M	0.5 wt% PEO (8×10^6 g/mol)
1%-2M	1 wt% PEO (2×10^6 g/mol)
1.1%-2M	1.1 wt% PEO (2×10^6 g/mol)
2%-0.6M	2 wt% PEO (0.6×10^6 g/mol)
CaBER	Capillary Breakup Extensional Rheometer
CFD	Computational Fluid Dynamics
CPyCl	Cetyl Pyridinium Chloride
CTAC	Cetyl Trimethylammonium Chloride
DNS	Direct Numerical Simulations
DRIE	Deep Reactive Ion Etching
DRP	Drag Reducing Polymer
DSM	Differential Stress Model
EVM	Eddy-Viscosity Model
FENE	Finitely Extensible Nonlinear Elastic
FFT	Fast Fourier Transform
FITC	Fluorescein Isothiocyanate
FITC-PEO	FITC-labeled PEO
FPS	Frames Per Second
fRe	friction factor - Reynolds number product

HWA	Hot Wire Anemometry
HWNP	High Weissenberg Number Problem
LDA	Laser Doppler Anemometry
LES	Large-Eddy Simulations
NLEVM	Non-Linear Eddy-Viscosity Model
PAA	Polyacrylamide
PDF	Probability Density Function
PEG	Polyethylene Glycol
PEO	Polyethylene Oxide
phPAA	partially hydrolyzed PAA
phPAA-FITC	FITC-labeled phPAA
PIV	Particle Image Velocimetry
PMMA	Polymethyl Methacrylate
PNG	Portable Network Graphics
POD	Proper Orthogonal Decomposition
PTT	Phan-Thein Tanner
RANS	Reynolds-Averaged Navier Stokes
RMS	Root Mean Square
UCM	Upper Convected Maxwell

List of Symbols

Roman

a	mobility factor
A_{initial}	average molecule size at $\delta = 0.1 - 0.2 \text{ mm}$ (μm^2)
A_{final}	average molecule size at $\delta = 1.4 - 1.5 \text{ mm}$ (μm^2)
b	mean rate of enstrophy transfer per unit mass (m^2/s^2)
c	solute concentration (g/ml)
c^*	critical overlap concentration (g/ml)
Cr	contraction ratio
d	depth of micro-channel (μm)
D	rate of deformation tensor (s^{-1})
D	diffusion coefficient (m^2/s)
De	Deborah number, $\frac{2\lambda_e Q}{w_c d^2}$
D_h	hydraulic diameter (mm), $\frac{2w_c d}{w_c + d}$
D_{mid}	mid-diameter of filament (mm)
D_o	initial diameter of filament (mm)
El	Elasticity number, $\frac{De}{Re}$
$E(k)$	isotropic energy spectrum (m^3/s^2)
f	friction factor
fRe	friction factor – Reynolds number product
f_s	sampling frequency (Hz)
F	body force (N)

G	elastic modulus (Pa)
L	contour length (μm)
L_{char}	characteristic length (μm)
L_v	vortex length (mm)
k	wave number (m^{-1})
k_B	Boltzmann constant
K	Kurtosis factor
M_w	molecular weight (g/mol)
N_A	Avogadro's number
P	pressure field (Pa)
Pe	Peclet number, $\frac{\bar{U}L_{\text{char}}}{D}$
Q	total flow rate (ml/h)
Re	Reynolds number, $\frac{2\rho Q}{\eta(w_c+d)}$
$R(s)$	autocovariance function
s	time lag (s)
S	Skewness factor
$S(f)$	temporal power spectrum ($\text{mm}^2/\text{s}^2/\text{Hz}$)
$S(k)$	spatial power spectrum (mm^2/s^2)
t	time (s)
Δt	PIV time interval (s)
t_{flow}	characteristic time scale of system (s)
T	time period which N measurement samples are collected (s)
T_{critical}	critical parameter at which turbulence sets in (s)

Tu	turbulence intensity, $u_{i,rms}/\bar{U}_i$
u	fluctuating axial velocity (mm/s)
u_{rms}	root mean square axial velocity (mm/s)
$u_{i,rms}$	u_{rms} at interrogation window (mm/s)
$\mathbf{u}$	fluctuating velocity component (mm/s)
$\overline{u_i u_j}$	Reynolds shear stress (mm^2/s^2)
$\overline{u_i v_i}/\bar{U}_i^2$	normalized Reynolds shear stresses at interrogation window
U_{CL}	convective velocity (mm/s)
$\mathbf{U}$	velocity vector (mm/s)
$\bar{U}_{bulk}$	mean fluid velocity (mm/s)
$\bar{U}_i$	mean fluid velocity at interrogation window (mm/s)
$\bar{U}_i/\bar{U}_{bulk}$	normalized mean velocity at interrogation window
v	fluctuating transverse velocity (mm/s)
w_u	upstream channel width (μm)
w_c	contraction width (μm)
Wi	Weissenberg number, $\lambda\dot{\kappa}$
x	Cartesian position (μm)
x	molecular extension (μm)
<u>Greek</u>	
α	ellipse minor-axis (μm)
β	ellipse major-axis (μm)
(β/α)	aspect ratio
$(\beta/\alpha)_{\text{normalized}}$	normalized aspect ratio
γ	strain (μm)

$\dot{\gamma}$	shear rate (1/s)
$\dot{\gamma}_c$	characteristic shear rate in De computation (1/s)
δ	downstream contraction displacement (mm)
ϵ	mean rate of kinetic energy transfer per unit mass (m^2/s^2)
$\dot{\epsilon}$	extensional strain rate (1/s)
η	solution viscosity ($\text{mPa}\cdot\text{s}$)
η_e	extensional viscosity ($\text{mPa}\cdot\text{s}$)
η_o	zero-shear rate viscosity ($\text{mPa}\cdot\text{s}$)
η_∞	infinite-shear rate viscosity ($\text{mPa}\cdot\text{s}$)
η_p	polymer contribution to zero-shear viscosity ($\text{mPa}\cdot\text{s}$)
η_s	Newtonian contribution to zero shear viscosity ($\text{mPa}\cdot\text{s}$)
$[\eta]$	intrinsic viscosity (cm^3/g)
$\dot{\kappa}$	average shear rate in Wi computation (1/s)
λ	material relaxation time (ms)
λ_e	extensional relaxation time (ms)
$\lambda_{\text{emission/excitation}}$	emission/excitation wavelengths (nm)
λ_{source}	wavelength of light source (nm)
λ_m	Maxwell relaxation time (ms)
λ_p	single mode polymer relaxation time (ms)
μ	dynamic viscosity ($\text{mPa}\cdot\text{s}$)
ρ	density of fluid (kg/m^3)
$\rho(s)$	normalized autocorrelation function
σ	normal stress tensor (Pa)
σ_s	surface tension (N/m)

τ	characteristic time of fluid (ms)
$\tau_{90\%}$	time interval for 90% polymer relaxation (ms)
$\boldsymbol{\tau}$	shear stress tensor (Pa)
$\tilde{\tau}$	upper-convected time
$\boldsymbol{\tau}_p$	elastic stress tensor (Pa)
$\boldsymbol{\tau}_s$	viscous stress tensor (Pa)
$\boldsymbol{\tau}_{ij}^R$	Reynolds shear stresses (mm^2/s^2)
$\bar{\boldsymbol{\tau}}_{ij,p}$	time-averaged polymer stress tensor (Pa)
$\chi,$	dimensionless vortex reattachment length
ω	radian frequency (rad/s)

Blank Page

1. Introduction

1.1 Background

Turbulence is ubiquitous; it occurs in many forms e.g. capillary wave turbulence ([Newell and Rumpf 2011](#)), financial market turbulence ([Mandelbrot et al. 2005](#)), magnetohydrodynamic turbulence ([Mininni 2011](#)), electrical turbulence ([Winfree 1994](#)), and fluid turbulence ([Pope 2003](#)). The term 'turbulence' is a connotation often associated with the meaning of highly disordered, random and unpredictable behaviour. In fluid dynamics, turbulence is a phenomenon associated with high Reynolds number (Re) i.e. inertial-driven, and has been extensively studied in pipe flows, wind tunnels, the earth's atmosphere and micro-channel flows. High-Re turbulent flows can be classified into two main types, i.e. 2-D and 3-D turbulence. The most commonly encountered turbulent flows are 3-D in nature, where the Richardson's direct energy cascade and Kolmogorov's hypothesis applies ([Pope 2003](#)). However when the aspect ratio is very large, the turbulence may be classified as 2-D, e.g. atmospheric turbulence, soap films, in which an inverse energy cascade exists ([Kraichnan 1967](#), [Leith 1968](#), [Batchelor 1969](#)). The characteristics of high-Re turbulence include good mixing and heat transfer, large velocity fluctuations, increase in resistance to flow, a Kolmogorov's scaling in the inertia-subrange, and the presence of turbulent eddies etc. Although turbulence has been intensively researched, this ubiquitous phenomenon remains the most perplexing fundamental problem of classical physics.

In recent years, another phenomenon i.e. viscoelastic turbulence, has gained increasing attention because it violates the conventional school of thought i.e. high-

Re criteria, for creating chaos and disorder in a fluid dynamics system. Apparent from its name, the driving force of the phenomenon is elasticity; this property is acquired by a viscous liquid when small amounts of polymer additives (< 1 wt%) are dissolved in it, and the liquid becomes viscoelastic. Viscoelastic turbulence occurs when elastic forces are much more dominant as compared to inertial forces in the flow field i.e. elastic-driven. Hence, its manifestation typically occurs in small-scale setups e.g. micro-channels, where elastic effects are amplified under large extensional or shear rates, and inertial forces are negligible ($Re < 1$). Under such conditions, the Deborah number (De) or Weissenberg number (Wi), which describes the relative importance of elastic forces to viscous forces, replaces Re to become the critical parameter that governs the stability of the flow field.

Viscoelastic turbulence originated from the 1960s ([Vinogradov and Ivanova 1968](#)), when experimentalists discovered polymer melt fracture. However this phenomenon did not receive much attention until the late 1970s, when intensive investigations of viscoelastic contraction flows were carried out ([White and Kondo 1977](#), [Nguyen and Boger 1979](#), [Lawler et al. 1986](#), [Wunderlich et al. 1988](#), [Evans and Walters 1989](#), [Chiba et al. 1990](#), [Baloch et al. 1996](#), [Yesilata et al. 1999](#)). In these studies, flow instabilities typically absent in Newtonian liquids were reported. The discovery of such contrasting flow behaviour intrigued many researchers, and this led to an important finding by [Groisman and Steinberg \(2000\)](#), who reported the existence of a power law decay in the velocity spectrum; the first significant statistical measure to bridge the gap between viscoelastic turbulence and the (Kolmogorov's scaling in) conventional high-Re turbulence. Thereafter, viscoelastic turbulence re-emerged and the phenomenon was quickly exploited for

potential polymer-induced micro-mixing applications (i.e. typically difficult at low Re) in the twentieth century ([Groisman and Steinberg 2001a](#), [Gan et al. 2007](#), [Li et al. 2010](#), [Tatsumi et al. 2011](#), [Grilli et al. 2013](#)).

Since then, research in viscoelastic turbulence has progressed. Many reported works focused on obtaining the critical thresholds (De/Wi) for the onset of viscoelastic turbulence in various configurations, using viscoelastic liquids of varying compositions. However, there exists a large discrepancy in the critical thresholds (De/Wi) reported (refer to Tables 2-1, 2-2 and 2-3 on pages 38-43), although the phenomenon was unanimously attributed to the random relaxation of polymer molecules in the viscoelastic liquids, which had been highly extended via large shear gradients, stagnation points, and streamline curvature in the flow field. As such, the manifestations of viscoelastic turbulence can be found in rotating and curvilinear flows, cross-slot flows, and contraction and/or expansion flows etc. ([Liu and Steinberg 2010](#), [Jun and Steinberg 2011](#), [Tatsumi et al. 2011](#), [Haward et al. 2012](#), [Bonn et al. 2011](#), [Gan and Lam 2012](#)), and no experimental findings on its occurrence in straight channel flows have been reported.

Hitherto, investigations on viscoelastic turbulence are largely limited to experimental works, because numerical predictions based on existing models are hindered by the high Weissenberg number problem (HWNP) ([Keunings 1986](#), [Abedijaberi and Khomami 2011](#), [Renardy 2014](#)). Since the polymer molecules are typically invisible, most experimental studies employ indirect measurement techniques e.g. particle image velocimetry (PIV), laser Doppler anemometry (LDA), pressure transducers, optical birefringence etc., to quantify the flow field i.e. velocity, flow resistance and stress. By analysing the motion of tracer additives in

PIV and LDA, the bulk viscoelastic liquid and molecular behaviour are deduced, with the implicit assumption that the liquid is homogenous. Based on the aforementioned quantification techniques, the reported characteristics of viscoelastic turbulence include an increase in flow resistance, increase in velocity fluctuations and turbulent intensities, a power law decay in the velocity power spectra, enhanced mixing efficiencies etc.

Although the existing indirect investigation techniques have enable researchers to gain a deeper understanding of the phenomenon till the present day, the accuracy of these techniques in describing the polymer dynamics has yet to be verified. Hence, it would be a breakthrough to observe the polymer molecules directly. Despite the abundant amount of literature, certain outstanding questions remain unexplained. What are the characteristics of viscoelastic turbulence and its similarities to high-Re turbulence? Hitherto, a comprehensive quantitative investigation and comparison has yet to be reported. Is the flow field characteristics of a viscoelastic turbulent flow related to the type and concentration of the polymer molecules in the viscoelastic liquid? How do such small quantities of polymer molecules perturb the flow so drastically to cause turbulence? Do the existing deductive techniques give a true account of the polymer molecular behaviour in a turbulent flow? These fundamental questions are yet to be answered.

1.2 Objectives

To answer these questions, it is important to review existing quantification parameters employed in high-Re turbulence. These methods can be applied to quantify the viscoelastic turbulent flow field, so that a comprehensive

characterization and comparison can be made. This is an objective of the study, and the quantification techniques will be reviewed in Chapter 3.

The characteristic time scales of a viscoelastic liquid i.e. bulk liquid and molecular relaxation time, varies with the concentration and type of polymer molecules used. Since viscoelastic turbulence is due to the random relaxation of polymer molecules, these characteristic time scales would directly influence the time scales and ‘structures’ of a viscoelastic turbulent flow. Thus, the second objective would be to design an experiment to extract these ‘molecular injected’ time scales from the flow field.

Lastly, it is fundamental to understand the underlying mechanisms of how polymer molecules perturb the flow field, and the relationship between the true molecular behaviour and existing (bulk) deduction techniques. Therefore, a new investigative technique has to be developed to directly visualize the polymer molecular behaviours in situ. This would be the last objective of the study.

These objectives will be further refined and reinforced in Chapter 2.

1.3 Outline

To gain an outlook of the research progress in viscoelastic (elastic-driven) turbulence, and identify the important quantification parameters in high-Re (inertial-driven) turbulence, literature in these two fields will be reviewed in Chapters 2 and 3. In addition, it is important to distinguish the differences between Newtonian and viscoelastic liquids, and to properly characterize the viscoelastic liquids; these are included in Chapter 4. The detailed experimental setup will be elaborated in Chapter 5. The objectives of the study will be addressed via proper

experimental design and the results will be included in Chapters 6 and 7. In the following, a brief outline of each chapter is presented.

Viscoelastic turbulence

Chapter 2 reviews the progress in viscoelastic turbulence research, including earlier contraction flow studies which were mainly based on streak-line visualizations. A review of some recent viscoelastic turbulent works which employ quantification techniques to examine the flow field characteristics is also included. This includes some relevant micro-mixing works which employ viscoelastic turbulence for mixing enhancement.

High-Re Turbulence

In Chapter 3, the two main types of turbulent flows i.e. 2-D and 3-D turbulence, and their distinct characteristics will be elaborated. In addition, the flow field quantification parameters for characterising high-Re wall bounded turbulence will be presented. Selected works on bounded channel flows, which employ the various parameters for quantifying turbulence, will be reviewed. Eddy visualization techniques typically employed to observe the fluctuating flow field e.g. Reynolds decomposition, Galilean decomposition, and Proper Orthogonal Decomposition (POD) etc., will also be briefly discussed.

Rheology

The rich dynamics of viscoelastic flows at diminishing Re shows that viscoelastic liquids are intrinsically different from Newtonian liquids. In Chapter 4, the characteristics of each class of liquid will be presented, and their differences

highlighted. The Maxwell model is one of the simplest viscoelastic models and will be elaborated. To measure its bulk properties, characterization of the test liquids using steady shear rate tests and extensional tests will be included. The effects of polymer concentration and molecular weight on their rheological properties will also be discussed.

Experimental Setup

Particle Image Velocimetry (PIV) is an established technique employed in literature to quantify the flow field, to enable further statistical analyses and eddy visualization. In Chapter 5, the PIV experimental setup for the investigation is presented, together with a brief description of some concepts and steps involved in the PIV analyses, and the micro-channel design used in the flow experiments. In addition, the importance of image enhancement techniques in this study, to increase computation accuracy is highlighted.

Results part 1 – Flow field analyses

In Chapter 6, quantification parameters reviewed in Chapter 3 will be computed based on the velocimetry data obtained from PIV, to correlate the viscoelastic turbulent flow field to that of high-Re turbulence. The statistics that will be presented include mean velocity profiles, high and low frequency velocity data, temporal and spatial correlation functions, and spectral analyses. In addition, flow field visualization based on Galilean decomposition and POD will be included. Based on spectral analyses, the times scales of the flow field will be extracted, and the relationship between the extracted time scales and that of the viscoelastic liquid will be examined.

Results part 2 – Molecular visualization

It is fundamental to directly observe the behaviour of the polymer molecules in a viscoelastic turbulent flow field for a good understanding of the underlying mechanisms involved. In Chapter 7, a high-speed molecular imaging technique developed in this study is presented. Based on this technique, the behaviour i.e. conformation changes of a polymer molecule in a viscoelastic flow can be observed, and correlated to the flow field statistics obtained from PIV. In addition, the relaxation characteristics of the polymer molecules at two flow rates will be compared i.e. laminar vs. viscoelastic turbulence.

Conclusion

A summary of the results will be presented in the final chapter, in alignment with the objectives of the study. In addition, future works will be discussed.

2. Viscoelastic Turbulence

2.1 Introduction

2.1.1 Overview

At low Reynolds number ($Re < 1$), the flow of viscous liquids e.g. water, is laminar. An aqueous solution, e.g. water, becomes viscoelastic when a small amount of polymer additives (< 1 wt%) are added to it. As compared to a viscous liquid, its flow behaviour can become drastically different and turbulent i.e. viscoelastic turbulence. Unlike conventional inertial turbulent flows at high Reynolds number (Re) driven by large inertial forces ($Re \sim 10^3 - 10^4$), e.g. in pipe flows ($Re > 2300$), viscoelastic turbulence occurs due to the manifestations of large elastic forces (i.e. elastic-driven) under low inertial conditions (i.e. $Re < 1$). In these (low Re) flows, the Deborah number (De) or Weissenberg number (Wi), either of which describes the relative importance of elastic forces to viscous forces, becomes the critical parameter that governs the flow transitions and stability of the flow field (the distinction between De and Wi will be discussed in Section 2.2). The elastic forces originate from the extension and relaxation of polymer chains in the polymeric/viscoelastic mixtures, and are only amplified under high extensional and shear rates in smaller conduits (i.e. hydraulic diameters < 1 mm). As the polymer molecules are typically invisible, hitherto the bulk behavior of a viscoelastic solution is usually deduced indirectly by observing the motion of tracer additives, and employing stress quantification techniques, with the implicit assumption that the solution is homogenous.

However, despite the abundant literature on viscoelastic flows, the theory of viscoelastic turbulence remains elusive. Two microscopic theories, i.e. the linear elastic theory (feedback theory) (Fouxon and Lebedev 2003) and non-linear elastic theory (passive theory) (Chertkov 2000), have been put forth to explain the coil-stretch transition of polymer molecules in a viscoelastic turbulent flow; however these theories are only valid for very dilute polymeric solutions ($\ll$ the overlap concentration) and for flows with low elastic stress contribution to the total stress tensor (i.e. little stretching of polymer molecules). Without an adequate understanding of the underlying mechanisms of viscoelastic turbulence, accurate numerical models cannot be formed to describe the dynamics of polymer molecule interacting with the flow field. Hence, the investigation of viscoelastic turbulence is still largely experimental in nature, because numerical computations of viscoelastic flows have been reported to breakdown at low Weissenberg numbers ($Wi \approx 1$), i.e. when polymer molecules are highly stretched; this is termed as “The High Weissenberg Number Problem” (HWNP) (Keunings 1986, Abedijaberi and Khomami 2011, Renardy 2014).

2.1.2 Background

Studies of viscoelastic turbulence originate from the experimental investigations on contraction flows, which date back to the 1970s. In these flows, viscoelastic liquids have been reported to behave very differently from their Newtonian counterparts. Most of the research has been based on a macroscopic approach, investigating the bulk behaviour of the liquid either by experimentation or simulation based on constitutive models. These works focused on the effects of fluid elasticity on

upstream vortex enhancement and flow transitions, i.e. Newtonian like flow, diverging flow, periodic flow, non-periodic flow, onset of viscoelastic instabilities etc., which were reported mainly based on flow visualizations without statistical analyses.. Due to the importance of these pioneering works in propelling the investigations of subsequent viscoelastic turbulent flows in other configurations, a brief review of contraction flows has been included in Section 2.3.

Since the 21st century, investigations of viscoelastic flows have adopted a more quantitative approach. The onset of instability in contraction flows piqued the interest of many researchers; viscoelastic instabilities in other flow configurations were subsequently reported, e.g. rotating flows, cross-slot flows, curvilinear flows, and contraction-expansion flows etc. One of the most significant works was that of [Groisman and Steinberg \(2000\)](#), who were the first to conduct statistical analyses on their velocimetry data. Upon the development of flow instabilities in their flow configuration, they reported the existence of a power law decay in the velocity power spectrum, which is a characteristic of high-Re turbulence. The term ‘elastic turbulence’ ([Vinogradov and Ivanova 1968](#)), initially used to describe polymer melt fracture, resurfaced and was used to describe this phenomenon. Here, the term ‘viscoelastic turbulence’ would be more appropriate because viscoelastic liquids consist of very low polymer concentrations (< 1 wt%) and have much lower viscosities (< 100 mPa·s under high shear rates) as compared to polymer melts.

In agreement with the findings of [Groisman and Steinberg \(2000\)](#), many others have also reported the presence of the power law decay in the velocity power spectrum ([Groisman and Steinberg 2001a](#), [Arratia et al. 2006](#), [Bonn et al. 2011](#), [Tatsumi et al. 2011](#), [Grilli et al. 2013](#), [Zhang et al. 2013](#)). The possibility to

generate turbulence in a micro-channel also led to an increase in micro-channel mixing research (Gan et al. 2007, Groisman and Steinberg 2001a, Li et al. 2010, Tatsumi et al. 2011, Grilli et al. 2013), because flows in miniaturized dimensions, i.e. < 1 mm, are usually laminar (low Re) and inhibit efficient and rapid mixing of fluids, e.g. in chemical reactions. These studies have reported significant improvements in mixing efficiencies beyond the onset of flow instabilities.

In recent years, statistical quantification methods e.g. turbulent intensities, correlation times, velocity power spectrum etc. have also been employed to compare the similarities of viscoelastic turbulence to high-Re turbulence. In addition to velocity measurements (PIV and LDA), other flow field measurement techniques e.g. birefringence stress measurements, molecular stress probes (DNA) etc., have also been employed to supplement the analyses of the viscoelastic turbulent flow field. In many reported works, these results are typically used to deduce the behaviour of the polymer molecules in the flow. However the indirect deduction of molecular behaviour has shown to be inadequate to elucidate the underlying mechanism of how the polymer molecules interact with the flow field in viscoelastic turbulence.

2.1.3 Outline

In Section 2.2, the relevant dimensionless parameters will be presented. The pioneering works on contraction flows were important in propelling the investigations of viscoelastic turbulent flows. Hence, a brief summary of these studies has been included in Section 2.3. A selection of recent viscoelastic turbulent works which have included various quantification techniques has been

included in Section 2.4. Some of these works have also employed the generated turbulence for mixing efficiency investigations. Lastly, a summary and discussion of the works are presented in Section 2.5, together with the thrust and objectives/scope of this study.

2.2 Dimensionless numbers

Before reviewing previous investigations, it is necessary to define the terminology of some common terms relevant to micro-channel flows. The Reynolds number is a dimensionless number named after Osborne Reynolds in 1883 ([Reynolds 1883](#)) that describes the importance of inertial forces to viscous forces for a particular flow field. Typically in pipe flow, laminar flow is characterized by $Re < 2300$ and turbulent flow by $Re > 4000$. For $2300 < Re < 4000$, this is termed as the transition zone ([Munson et al. 2002](#)). In laminar flows, the flow field is smooth and constant with time. However, turbulent flows are characterized by highly fluctuating velocity fields, accompanied by vortices. The Reynolds number, Re , is defined as

$$Re = \frac{\rho \bar{U} L_{char}}{\mu} \quad (2.1)$$

where ρ = density of fluid, $\bar{U}$ = mean fluid velocity, L_{char} = characteristic length and μ = dynamic viscosity of the fluid.

The Deborah number describes the relative importance of elastic forces to viscous forces in a flow field and is typically computed when there is an abrupt change in channel geometry that results in a change in deformation rate e.g. contraction-expansion channels. It is suitable for characterising materials that flow over long time scales, e.g. the Maxwell model, but not for materials which are viscous only on

short time scales but solid for long times, e.g. Kelvin-Voigt model. The Deborah number, De , can be defined as

$$De = \frac{\lambda}{t_{\text{flow}}} = \lambda \dot{\gamma}_c \quad (2.2)$$

where λ is the material relaxation time, t_{flow} is the characteristic time scale of the system, and $\dot{\gamma}_c$ the characteristic shear rate due to the abrupt geometrical change. For example, in contraction-expansion flows, $\dot{\gamma}_c$ corresponds to the shear rate in the contraction. On the other hand, another dimensionless number known as the Weissenberg number, Wi , can be defined as

$$Wi = \lambda \dot{\kappa} \quad (2.3)$$

where $\dot{\kappa}$ is the average shear rate in the flow. Wi is typically computed in place of De , when there are no geometrical changes in the channel dimensions, and the deformation rate is constant throughout the flow e.g. straight channel flows. However, a comprehensive literature review shows that reported works have been using these two numbers interchangeably.

It is common to compute the importance of elastic forces to inertial forces using the elasticity number, El , defined as

$$El = \frac{De}{Re} \quad (2.4)$$

The Peclet number, Pe , describes the relative importance of convection in a particular flow field to diffusion. A large Peclet number indicates the dominance of convection, while a small Peclet number means that Brownian motion effects are important and not negligible. It is defined by

$$Pe = \frac{\bar{U}L_{char}}{D} \quad (2.5)$$

where D is the diffusion coefficient.

2.3 Viscoelastic Contraction flows

2.3.1 Overview

Much research works on viscoelastic contraction flows originated from the 1970s, and results were based on flow visualizations. These studies form the basis of elastic turbulent flows and a brief summary of these works have been included in this section. These investigations can generally be classified under macro-contraction channels (smallest characteristic dimension $\geq 1\text{mm}$) or micro-contraction channels (smallest characteristic dimension $< 1\text{ mm}$) in this study. Note that this ‘macro’ and ‘micro’ terminology is employed in this study for ease of discussions and should not be cross-referenced to other reported works.

In the literature on contraction viscoelastic flows, various contraction ratios ($Cr = w_u/w_c$, refer to Figure 2-1) have been reported. Corner vortices usually form upstream, depending on the flow conditions, i.e. Reynolds and Deborah numbers. These vortices grow in size with increasing El and are quantified based on the dimensionless vortex reattachment length ($\chi = L_v/w_u$). The presence of the vortices has been attributed to be a possible type of stress relief mechanism (White and Baird 1986). Figure 2-1 shows the flow of an aqueous polyacrylamide (PAA) solution through a contraction, accompanied by the presence of salient corner vortices (Evans and Walters 1989).

Figure 2-1. Presence of corner vortices in flows of aqueous 0.2% PAA solution into a 4:1 contraction (Evans and Walters 1989)

2.3.2 Macro-channel Contraction flows

Flows of viscoelastic liquid through macro-channel (≥ 1 mm) contractions date back to the 1970s. Here, a brief summary of these works will be described. Experimental investigations first began with axisymmetric channels due to the ease of connecting circular pipes or capillary tubes together. This was also due to the limitations of micro-fabrication techniques and these studies were targeted at die entry flows. Early works were based on flow field visualizations of Boger fluids i.e. image snapshots, which mainly focused on the effect of elasticity on vortex enhancements (corner and lip vortices, see Figure 2-1) and flow transitions i.e. Newtonian like flow, diverging flow, periodic flow, non-periodic flow, unstable flow. (White and Kondo 1977, Nguyen and Boger 1979, Lawler et al. 1986, Yesilata et al. 1999). In addition, pressure drop measurements were also employed as a means of detecting flow instabilities (Yesilata et al. 1999).

From the 1980s, flows of Boger fluids and shear-thinning liquids, e.g. dilute PAA solutions, in planar channels were the next focus of many research works (Wunderlich et al. 1988, Evans and Walters 1989, Baloch et al. 1996, Chiba et al. 1990, Nigen and Walters 2002, Alves et al. 2005, Poole et al. 2005, Alves et al. 2008, Keegan et al. 2008). Similarly, these works investigated on vortex enhancements, flow transitions, and pressure drop measurements with varying elasticity. Viscoelastic liquids were found to have extra resistance to the flow and higher pressure was required (Nigen and Walters 2002). Experimental and numerical works also began to quantify the flow field via velocimetry measurements, which were realized experimentally using PIV and LDA in most studies. These measurements were used to deduce the behaviour of the polymer molecules in various regions of the flow configuration, e.g. molecular extension near contraction. Elasticity was reported to lead to the formation of ‘cats ears’ in the velocity profile, and result in an earlier acceleration of the center-line velocity, typically accompanied by diverging streamlines (Wunderlich et al. 1988, Alves and Poole 2007). An interesting phenomenon termed as ‘flow inversion’, was also reported in the 3-D square-square contraction flow of Alves et al. (2005, 2008).

However, the critical parameter (De or Wi) for the onset of flow instability began to show significant variations and conflicting results amongst the research works (refer to Table 2-1, page 38). This is due to the different methods in quantifying the relaxation time of the viscoelastic liquids, the different geometries used, and the varying properties of the test liquids.

2.3.3 *Micro-channel Contraction flows*

With the advancement of miniaturization techniques and increasing usage of micro-fabrication technology in the industry from the 1980s, flows in planar micro-channels were the next focus of many studies ([Rodd et al. 2005a, 2007](#), [Oliveira et al. 2007](#), [Gan et al. 2007](#), [Lam et al. 2009](#), [Afonso et al. 2011](#), [Gan and Lam 2012](#)). This is because it is generally harder to create axisymmetric channels using embossing or lithography techniques. Most experimental studies carried out similar investigations as the macro-channel counterpart aforementioned. These investigations typically involve shear-thinning fluids, because large forces are required to pump Boger fluids into such miniaturized dimensions, which could ultimately damage the channels. Due to the large De and Wi that could be attained at such small dimensions (diminishing Re), elastic instabilities could be easily achieved in these studies, and their focuses were on the critical parameters for the onset of flow instability (refer to Table 2-2, page 39).

With a saturation of research on contraction flows, [Rodd et al. \(2005a, 2007\)](#) was one of the pioneers to investigate the flows of viscoelastic liquids past a contraction-expansion micro-channel (16:1:16 contraction-expansion ratio, 400 μm : 25 μm : 400 μm , height = 50 μm). They investigated the vortex growth, pressure drop, and flow transitions that led to elastic instabilities upstream of the contraction. They also noted that elasticity resulted in the growth of vortices upstream but suppressed downstream vortices which could be observed with the Newtonian liquids. One of their most significant contributions was the Wi - Re operating space diagram (see Figure 2-2), which predicts the onset of flow transitions in their flow configuration.

Figure 2-2. Summary of the Wi-Re space diagram for flow transitions of different fluids through a 16:1:16 contraction-expansion (Rodd et al. 2005a)

2.4 Viscoelastic turbulent flows

2.4.1 Overview

The literature on contraction flows (In Section 2.3) has shown that with an increase in elasticity, i.e. high De and low Re , the flow field can become unstable. Most of these research works dictate the onset of viscoelastic instability mainly based on flow visualizations. In recent years, viscoelastic instabilities/turbulence has also been reported in many other geometrical configurations, e.g. rotating flows, cross-slot flows, curvilinear flows, and contraction-expansion flows etc. The amplification of the elastic effects (e.g. due to high shear rates) of the polymer molecules in the absence of inertia forces, together with streamline curvature, have been reported to form the key ingredients for the manifestation of viscoelastic turbulence. In these studies, further quantification via flow field measurements (PIV, LDA) and other methods (birefringence measurements, stress probes etc.) have

been used to uncover the dynamics of the viscoelastic turbulent flow fields, which share similarities to high-Re turbulent flows. In addition, some of these works have also exploited the generation of elastic turbulence in small scale channels, and included investigations on the mixing capabilities of their configuration.

Hitherto, there is no standard definition of a viscoelastic turbulent flow, and the terms ‘turbulence’ and ‘instabilities’ have been used interchangeably. Here, we review the findings of some significant experimental works on both viscoelastic ‘instabilities’ and ‘turbulence’ which have included various quantification techniques, and hence the precise definition of terminology is unimportant in the current context. Generally, the reported characteristics of viscoelastic instabilities/turbulence include an increase in flow resistance, increase in velocity fluctuations and turbulent intensities, a power law decay in the velocity power spectra, enhanced mixing efficiencies, increase in molecular extension of the stress probes, and an increase in optical retardation in birefringence measurements etc. A summary of these studies, and other works that are not elaborated here, have also been included in Table 2-3 (page 40).

2.4.2 Rotating flows

[Groisman and Steinberg \(2000\)](#) were one of the first to report statistical analysis for viscoelastic turbulence at low Reynolds number. For rotating flows of dilute shear thinning PAA solutions (80 ppm in 65% saccharose-water) in a tank (see Figure 2-3 a), they observed transition of laminar flow to instability at low Reynolds numbers ($Re = 0.3$, $Wi = 3.5$) and a non-linear increase in resistance which was much higher than those compared with Newtonian flows. In these situations, the characteristic

Figure 2-3. a) Schematic setup of [Groisman and Steinberg \(2000\)](#) $R = 38\text{ mm}$, $R_2 = 43.6\text{ mm}$, $d = 10\text{ mm}$ b) Power spectra of velocity fluctuations using laser doppler anemometry for shear rates ① 1.25 s^{-1} , ② 1.85 s^{-1} , ③ 2.7 s^{-1} , ④ 4 s^{-1} , ⑤ 5.9 s^{-1}

time spectra of velocity fluctuations at different shear rates followed a power law decay (see Figure 2-3 b), a characteristic of high-Re turbulence. They showed in a separate experiment ([Groisman and Steinberg 2001b](#)) that when viscoelastic turbulence has developed, the relaxing torque due to the polymer solution (25 ppm PAA) was many times greater than that due to the solvent. In addition, this relaxing torque (which was dependent on the relaxation time of the polymer solution) did not immediately decay to zero when the applied rotating torque was stopped.

With a similar setup ($R = 2.25\text{ mm}$, $R_2 = 6\text{ mm}$, $d = 675\text{ }\mu\text{m}$), [Liu and Steinberg \(2010\)](#) reported a method to measure elastic stresses in viscoelastic turbulence ($Wi = 100\text{--}1000$) by visualization of fluorescent T4 DNA probes (10^{-3} ppm , T4GT7 DNA, 165.6 kbp / $1.08 \times 10^8\text{ g/mol}$) stretching in the midst of unstained DNA molecules at different concentrations ($6.29 - 100.64\text{ }\mu\text{g/mL}$ with $35 - 71.5\%$ sucrose). The fluorescent T4 molecules have increased contour lengths (i.e. molecular chain length) of $\approx 28\%$, and relaxation times of $\approx 51\%$ as compared to the unstained molecules. The critical Weissenberg number (Wi_c) for the occurrence

Figure 2-4. a) Probability Density Function of fractional extension (x/L) of DNA molecules at the highest Wi for 5 concentrations. b) square of the peak fractional extension (solid squares) for 5 concentrations, average of the square of the fractional extension (open squares). c) log-log coordinates of data in b). (Liu and Steinberg 2010)

of viscoelastic instability decreased with increasing polymer concentrations, but the elastic stresses (σ_p) in the flow asymptote to a saturated value (σ_p^s) at $Wi/Wi_c \approx 10$ for all concentrations. For each Wi and concentration, approximately 1000 molecular images were measured in an area of $267 \times 267 \mu\text{m}^2$. Based on their measurements, the T4 could be stretched up to 30% above their contour length (refer to Figure 2-4a). They also used the worm-like chain model to approximate the elastic stresses generated in the flow based on the time-averaged dimensions measured. Their calculations indicated that the elastic stresses (2 – 5 Pa) could exceed the viscous stresses by up to 60 times. However, time averaging for each molecule was required due to low frame grabbing rate (≤ 25 Hz) which resulted in significant blurring. This limitation together with the small observation window also inhibited them from investigating the relaxation properties of the DNA molecules over time.

2.4.3 Cross-slot flows

Using Boger fluids i.e., 200 ppm PAA in 97% glycerol aqueous solution, viscoelastic instabilities have also been observed by [Arratia et al. \(2006\)](#) in cross micro-channel flows (650 μm width, 500 μm depth). They observed the transition of a stable Newtonian-like flow to an oscillating time dependent flow at the stagnation point, whereby fluctuations grew larger with increasing shear rate. They reported the onset of time dependent flow to occur at $\text{Re} < 0.01$, $\text{De} = 12.5$. In addition, the power spectra of velocity measurements obtained by PIV also resembled an exponential decay (slope not measured).

[Haward et al. \(2012\)](#) investigated the flow of micellar surfactant liquids in a cross-slot micro-channel (0.2 mm x 1 mm, $\text{Re} = 10^{-8} - 0.16$, $\lambda_M = 2.5$ s, $\text{Wi} = 0.1 - 500$, $\text{El} = 2600 - 10^8$). PIV measurements were carried out for flow visualization and the ensemble average was obtained by averaging 5 successive image pairs. Based on measurements of birefringence (measure of principle stress difference, $\Delta\sigma = \sigma_{xx} - \sigma_{yy}$), they also investigated the microstructural orientation of micelles, assuming the flow was homogenous in the direction of light propagation with negligible effects from the glass channels. At $\text{Wi} < 1.6$, the flow was laminar and Newtonian-like, and a localized region of intense birefringence, i.e. increase in optical retardation, was observed at the stagnation point along the exit direction (see Figure 2-5a). At $\text{Wi} = 1.6$ ($\text{Re} \approx 2 \times 10^{-5}$), the flow became asymmetric, with complete asymmetry being attained at $\text{Wi} = 2$. The birefringence region rotated with increasing asymmetry and a narrow weak birefringent strand was observed in the fully asymmetric state (see Figure 2-5b). At $\text{Wi} = 350$, the flow became unsteady and pressure drop measurements fluctuate erratically. However, quantitative

birefringent measurements were difficult to obtain due to the need for time-averaging.

Figure 2-5. Flow induced birefringence in a cross-slot flow. Optical retardation at a) $Q = 1.0 \mu\text{L min}^{-1}$, $Wi = 1.0$, and b) $Q = 2.0 \mu\text{L min}^{-1}$, $Wi = 2.0$, $Re < 10^{-5}$. The colour scale represents optical retardation in the range a) 0 – 10 nm b) 0 – 30 nm (Haward et al. 2012).

2.4.4 Curvilinear flows

Groisman and Steinberg (2001a) investigated the effect of viscoelasticity on mixing in planar curved channels (see Figure 2-6 a) with a width and depth of 3mm ($D_h = 3\text{mm}$), between two streams of dilute shear thinning PAA solutions (80ppm), one which contained fluorescence dye. As the flow rate was increased, transition to viscoelastic instability occurred ($Re = 0.06$, $Wi = 3.2$). Using the probability density functions and value of moments (down to 0.03), they concluded that good mixing could be achieved in the micro-channel with the optimum at $Wi = 6.7$. At $Wi = 6.7$, they noted that the power spectra of velocity fluctuations (measured using LDA) had broad regions of power law decay which is typical in high- Re turbulent flows (see Figure 2-6 b). However, their mean velocity was recorded to be 6.6mm s^{-1} , with velocity fluctuations having a root mean square (RMS) of 0.594mm s^{-1} . The exponential decay of the higher order moments implied similarity to the chaotic Batchelor regime of mixing (i.e. mixing at scales smaller than the Kolmogorov

Figure 2-6. a) Schematic diagram of channel. Note $R_2 = 6$ mm, $R_1 = 3$ mm (Groisman and Steinberg 2001a)
b) Power spectra of fluctuations at $Wi = 6.7$ for 1= longitudinal velocity, 2= transverse velocity.

microscales). They further verified that this instability in the channel could not be replicated when only Newtonian fluids were used. They achieved a mixing efficiency of more than 70% over a flow path of approximately 340 mm.

Burghlea et al. (2004) repeated the work of Groisman and Steinberg (2001a) with the same channel in a miniaturized form ($100\mu\text{m}$ width and depth, $D_h = 100\mu\text{m}$). Two sample fluids were used, i.e. Solution 1 with 80 ppm of PAA in 35% sucrose in water ($\eta_o = 5.6$ mPa·s), and shear thinning Solution 2 being 80 ppm of PAA in 64.4% sucrose in water ($\eta_o = 138$ mPa·s). They observed a transition from linear to nonlinear increase in resistance, for flows of Solution 1 to be at $Re = 0.017$, $Wi = 1.4$, while that of Solution 2 to be at $Re = 0.00008$, $Wi = 3.5$. They attributed this to be a nonlinear elastic flow transition. Beyond this transition point, velocity strongly fluctuated with time and the velocity autocorrelation function decayed uniformly

Figure 2-7. a) Large fluctuations in longitudinal velocity b) Autocorrelation function for a) based on 6000 velocity measurements (Burghellea et al. 2004)

(refer to Figure 2-7), indicating chaotic-like flow which was also observed previously (Groisman and Steinberg 2001a). These behaviours were not observed when Newtonian fluids were used, whereby only slight smearing occurred at the interface. Mixing experiments conducted with the polymer mixtures revealed an exponentially decaying high order moments, which coincide with the chaotic Batchelor regime of mixing. This provides second evidence that the flow field was viscoelastically turbulent, which was absent when Newtonian fluids were used. An improved mixing efficiency of more than 80% was achieved over a flow path of approximately 20 mm.

Jun and Steinberg (2011) investigated the flow of dilute PAA solution (80 ppm in 65% sucrose-water solvent, 18 Mda, Polysciences, $\lambda = 11.5$ s) through a curvilinear macro-channel (cross-section = 1 mm x 1 mm, $Wi = 0 - 950$, $Re < 0.54$) using PIV (5 Hz). With an increase in Wi , the temporal velocity profiles displayed large fluctuations. They plotted the velocity fluctuations against Wi , and stated the onset of elastic instability and elastic turbulence to occur at $Wi = 200$ and $Wi = 360$ respectively, based on the sudden change in slope. Their investigations showed that

Figure 2-8. Plot of $(\partial V_{\theta}/\partial r)^{\text{rms}}$ against r/d at various Wi . The boundary layer corresponds to the region from the wall ($r/d = 0$) till the region where $(\partial V_{\theta}/\partial r)^{\text{rms}}$ reaches the maximum value (Jun and Steinberg 2011).

the rms values of the velocity gradient, $(\partial V_{\theta}/\partial r)^{\text{rms}}$, were large close to the wall, and identified this region to be a boundary layer (see Figure 2-8), with a characteristic width that was independent of Weissenberg number and proportional to channel width. They stated that the non-uniform distribution of $(\partial V_{\theta}/\partial r)^{\text{rms}}$ indicated the non-uniform elastic stress distribution in the bulk flow, which consists of spatial scales (i.e. spatial correlation function) on the order of the boundary layer width. They hypothesized that this was due to excessive elastic stresses in the boundary layer being ejected into the bulk flow. Their results showed that the localized Wi did not saturate in the bulk flow of elastic turbulence, hence contradicting the prediction by the elastic turbulence theory.

Tatsumi et al. (2011) investigated the flow of polyacrylamide (100 ppm, 18×10^6 g/mol) solution in a curvilinear micro-channel ($84 \mu\text{m} \times 60 \mu\text{m}$, $D_h = 70 \mu\text{m}$) at $Re < 3.6 \times 10^{-3}$. The Deborah or Weissenberg numbers were not computed. Above Re

Figure 2-9. a) Schematic diagram of micro-channel with two inlets, with one stream containing fluorescent dye. b) negligible mixing near the inlet c) almost complete mixing (up to 80%) near the outlet. (Tatsumi et al. 2011)

$= 6.7 \times 10^{-4}$ the flow became unsteady, and their measurements showed a mixing efficiency of up to approximately 80% based on standard deviation analyses of the fluorescent intensity (refer to Figure 2-9). With an image sampling rate of 100 Hz, their PIV measurements showed low frequency and large amplitude velocity fluctuations, with turbulent intensities of 0.3. The power spectrum showed a power law decay in between 0.4 Hz to 5 Hz. They suggested that the instability observed was due to polymers or solution being stretched by the shear stresses in the flow.

2.4.5 Contraction and/or expansion flows

Gan et al. (2007) was the first to investigate the elastic instability produced by a 3-stream flow of dilute polyethylene oxide (PEO) solutions through a planar micro-channel with a sudden contraction-expansion ratio of 8:1:8 (1 mm: 125 μ m: 1 mm). The centre stream contained a more concentrated (1 wt%) PEO solution while the streams at the side were of lower concentration (0.1 wt%). They studied the interaction between the two fluids upstream and noted significant elastically induced instability downstream of the contraction ($Re = 0.025$, $De = 203.5$). The occurrence of viscoelastic instability was observed through the manifestation of ‘viscoelastic whipping’ upstream, and ‘swinging’ flow (up-down sweeping) of the

Figure 2-10. a) PIV image upstream of contraction, with average velocity plot at section A-A. b) PIV image downstream of contraction, with average velocity plot at section B-B. The standard deviation is large in both plots. (Lam et al. 2009)

highly elastic main-stream liquid downstream of the contraction. In addition, they also verified that without elasticity, this instability could not be triggered. In a separate publication (Lam et al. 2009), they reported particle image velocimetry (PIV) results across the upstream and downstream channels which showed large fluctuations especially near the centre-line, downstream of the contraction. This is believed to be the onset of viscoelastic turbulent flow, due to a sudden recovery of the elastic energy in the polymer chains upon exit of the contraction. Figure 2-10 illustrates the PIV results of Lam et al. (2009) across the upstream and downstream channels.

With this instability, significant mixing of up to 68% could be produced downstream of the contraction ($D_h = 261 \mu\text{m}$), over a channel length of approximately only 6 mm. However, criticisms on this setup resembling flow

focusing, and complications caused by shear thinning and other effects have been suggested by [Bryce and Freeman \(2010\)](#).

In a separate publication, [Gan and Lam \(2012\)](#) presented a De_{ratio} – De_{main} operating space (see Figure 2-11) which categorized the flow transitions and mixing efficiencies of the 3-stream flow. They highlighted that the discontinuity in viscoelastic energy i.e. De_{ratio} , between the two fluids was the key explanation for the flow transitions towards viscoelastic instability.

Figure 2-11. De_{ratio} – De_{main} space diagram for various flow regimes. Black dot: $El_{main} \sim 7271$, Red hexagon: $El_{main} \sim 2011$ and Grey diamond: $El_{main} \sim 37.1$. ([Gan and Lam 2012](#))

[Bonn et al. \(2011\)](#) showed that elastic turbulence could occur in straight channels, provided there was a large initial perturbation of the flow. They investigated the flow of polyacrylamide solutions (100 – 1000 ppm, 18×10^6 amu, $\lambda_{Carreau} \approx 1$ s) exiting from a smaller cylindrical tube ($2R = 1$, 3.5 mm) into a larger cylindrical tube ($2R = 5$, 8 mm). Based on Laser Doppler anemometry (LDA), the PAA

Figure 2-12. a) Power spectra of the velocity fluctuations at $x = 3.5$ cm at various flow rates. b) Polymer elongations for $Wi = 1$ and 4: Image 1 is near the outlet, and images 2–5 are further downstream at various distances from the channel wall. (Bonn et al. 2011)

solution exhibited large velocity fluctuations, and the maximum velocity exceeded the mean value by up to 30% (Note: not turbulent intensity). At $Wi = 4$, a power law decay with a slope of -1.5 was observed in the power spectrum (see Figure 2-12a), which persisted till 19.5 cm away from the exit. Subsequently, they seeded a 1000 ppm PAA solution with fluorescent T4 DNA molecules ($\lambda = 80$ s), flowing from a tube ($2R = 100 \mu\text{m}$) exiting into a rectangular channel ($200 \mu\text{m} \times 2 \text{ mm} \times 2 \text{ cm}$ length). To observe the elongation of DNA molecules at various heights and positions in the channel at $Wi = 1$ and $Wi = 4$, instantaneous snapshots of different

DNA molecules were captured. However, the small observation window inhibited them from investigating the relaxation properties of the DNA molecules over time; this could be due to the low capturing speeds (imaging details were not provided). Hence, direct comparisons between the molecular conformation changes and behaviour at each Wi could not be carried out. This is apparent in their figures (Figure 2-12b) whereby the DNA molecule at $Wi = 1$ was observed to be longer than that at $Wi = 4$. This is unexpected because DNA is mono-dispersed and should stretch more at higher Wi .

2.5 Review of viscoelastic turbulent flows

2.5.1 Summary

Studies of viscoelastic turbulence originate from experimental investigations on contraction flows which date back to the 1970s. These studies have been focused on the investigation of the critical De or Wi for the onset of various flow transitions which transit in the following order: Newtonian like flow, diverging flow, periodic flow, non-periodic flow, unstable flow. The investigations were conducted mainly via the streak-line visualizations of the salient and lip vortices, measurement of upstream velocity profiles via PIV or LDA, and pressure measurements. However with the increasing amount of literature, the critical flow parameters (De and Wi) for the onset of flow instability began to show significant differences between research works. This is due to the different methods in quantifying the relaxation time of the viscoelastic liquids, the different geometries used, and varying properties of the test liquids. Tables 2-1 to 2-2 show the summary of the conditions for instability onset in macro- and micro-contraction flows respectively.

The onset of instability in contraction flows piqued the interest of many researchers, and viscoelastic turbulence in other flow configurations was subsequently reported. In addition, these investigations began to adopt a more quantitative approach. Tables 2-3 show the summary of the conditions for instability onset in some of these flow configurations, together with the quantification technique employed. One significant finding by [Groisman and Steinberg \(2000\)](#) was the existence of a power law decay in the velocity power spectrum. This was also supported by the findings of other viscoelastic turbulent works ([Groisman and Steinberg 2001a](#), [Arratia et al. 2006](#), [Bonn et al. 2011](#), [Tatsumi et al. 2011](#), [Grilli et al. 2013](#), [Zhang et al. 2013](#)), and is a signature normally associated with high-Re turbulent flows. Many others also exploited the phenomenon to improve mixing in micro-channels ([Gan et al. 2007](#), [Groisman and Steinberg 2001a](#), [Li et al. 2010](#), [Tatsumi et al. 2011](#), [Grilli et al. 2013](#)) which is usually difficult due to low Re (normally associated with laminar flows). Since then, statistical quantification methods e.g. turbulent intensities, correlation times, velocity power spectrum etc. have also been employed to compare the similarities of viscoelastic turbulence to high-Re turbulence. In addition to velocity measurements (PIV and LDA), other flow field measurement techniques e.g. birefringence stress measurements ([Haward et al. 2012](#)), molecular stress probes (DNA) ([Liu and Steinberg 2010](#), [Bonn et al. 2011](#)) etc., have also been employed to supplement the analyses of the viscoelastic turbulent flow field. However, the application of birefringence measurements and the imaging of DNA probes as stress quantification tools have shown to be inadequate for viscoelastic turbulence investigations because of the need for space and time averaging. Moreover, the usage of fluorescent DNA probes for stress quantification is costly

and is only suitable for small, confined flow enclosures; it also does not reveal the underlying mechanisms in the turbulent flow, which is due to the invisible molecules. Generally, the reported characteristics of elastic instabilities/turbulence include an increase in flow resistance, increase in velocity fluctuations and turbulent intensities, a power law decay in the velocity power spectra, enhanced mixing efficiencies, increase in molecular extension of the stress probes, and an increase in optical retardation in birefringence measurements. In addition, a boundary layer exists for single stream turbulent flows ([Jun and Steinberg 2011](#)).

2.5.2 Discussion

The viscoelastic test liquids amongst the reviewed works were different due to the varying molecules (i.e. PEO, PAA, DNA), molecular weights, and concentration used. If the elastic forces (and hence polymer molecules) are primary in driving the turbulent flow, surely the different characteristics would be revealed in the obtained results? However, despite the differences in molecular and bulk properties of the test liquids, statistical analyses of the reviewed works could only distinguish significant differences when compared to the Newtonian counterpart, but not amongst the various test liquids used. Regardless of the varying molecular and bulk properties, the characteristics of elastic turbulence i.e. large velocity fluctuations, power law decay etc. were generally similar, except for the differences in critical parameters (De or Wi) for instability onset. One plausible explanation could be due to the lack of a comprehensive investigation in a single flow configuration, regarding the effects of varying molecular and bulk properties on the characteristics of viscoelastic turbulence. In addition, it is possible that characteristic time scales of the bulk liquids and molecules within the flow configuration were short, and could

not be captured by the low data acquisition rates used (typically $\leq 10\text{Hz}$). Furthermore, the reviewed elastic turbulent works could be 3-D in nature, and the driving frequencies for the energy transfer in a 3-D turbulent flow could not be observed distinctly in the velocity power spectra. This is further complicated by the polydispersity of the polymer molecules which result in a wide range of characteristic times of the driving elastic forces.

Viscoelastic liquids have typically been studied as a homogenous bulk liquid, and microscopic details e.g. polydispersity effects are usually ignored. The assumption of homogeneity, and the use of bulk properties (i.e. relaxation time obtained under static conditions) for the quantification of critical parameters, are the main reasons for the discrepancy on the critical parameters (De or Wi) for various flow transitions between reported works. Despite the large amount of literature, the mechanism of viscoelastic turbulence remains to be elusive. The indirect quantitative approach found in majority of the literature is clearly inadequate and the research on viscoelastic turbulence has reached a bottle neck. It is fundamental to understand how the small quantities of polymer molecules would result in such significant changes to the flow behavior. In addition, it is necessary to consider the relaxation of polymer molecules, to understand how they interact with the flow field, resists deformation, and cause turbulence. Instead of relying on space and time averaged snapshots, which are unable to capture the molecular relaxation dynamics, a practical and low cost technique that enables real-time observations of the molecular conformation changes (i.e. extension and relaxation) with high clarity is required. This would shed light onto the mechanisms of viscoelastic turbulence,

could result in the development of more exact numerical models, and alleviate the high Weissenberg number problem.

2.6 Thesis planning

Hitherto, a complete description of viscoelastic turbulent flows in micro-channels ($< 1\text{mm}$) is absent. Flow field visualizations tools e.g. Gallilean decomposition, Reynolds decomposition, and Proper Orthogonal Decomposition (POD), typically employed in high-Re turbulent flows to uncover the presence of large scale structures and turbulent eddies, has not been reported. Of particular interest is the phenomenon discovered by [Gan et al. \(2007\)](#) and [Lam et al. \(2009\)](#) in a 3-stream contraction-expansion flow. At the onset of instability ($De = 203.5$, $Re = 0.025$), the main-stream liquid exhibited an up-down sweeping motion across the channel downstream of the contraction, which resulted in high mixing efficiencies. Although micro-PIV was carried out, no further statistical analysis was conducted.

The manifestation of this turbulent ‘sweeping’ motion of the main-stream liquid appears to be 2-D in nature. Hence, this flow configuration is excellent for investigating the effects of varying molecular and bulk properties on the flow field, because the energy injection point (i.e. forcing frequency) in the 2-D energy cascade is distinct ([Kraichnan 1967](#), [Leith 1968](#), [Batchelor 1969](#)). This energy injection corresponds to the elastic energy released by the polymer molecules and transferred to the flow field, and the main dissipation occurs at the largest scales ([Fouxon and Lebedev 2003](#)). It will be highly desirable and advantageous to develop a technique to observe these polymer molecules, and relate the molecular conformation changes (i.e. energy changes) to the properties of the viscoelastic

turbulent flow; this has the potential to reveal the underlying mechanisms of viscoelastic flows.

Hence, the objectives of this study are:-

- 1) To examine the flow field of the 3-stream 8:1:8 contraction-expansion flow, and apply statistical analyses and flow visualization tools on the flow field. Focus will be placed on the downstream flow field to investigate the 2-D nature of the turbulent flow.
- 2) To investigate the effects of varying molecular and bulk liquid properties on the resultant flow field, and to explore the possibility of extracting information from the flow dynamics to deduce the molecular and bulk liquid properties.
- 3) To develop a technique to examine real-time observations of the molecular conformation changes (i.e. extension and relaxation) in a viscoelastic turbulent flow, and correlate the observations to the dynamics of the flow field.

Table 2-1. Conditions for onset of unstable flows in macro-contractions.

Macro- contraction flows						
Investigator	Contraction ratio	Fluids used	Onset of time-dependent unstable flow			Method of obtaining relaxation time (λ)
			Re	De/Wi	El	
Nguyen and Boger 1979	22.2 mm : 2.9 mm	Boger fluid (< 0.1% PAA)	0.0045	Not stated	N.A.	Oscillatory measurements (Maxwell)
Lawler et al. 1986	50.8 mm: 12.7 mm	Boger fluid (0.17% polyisobutylene)	0.044	0.852	19.36	Oscillatory measurements (Maxwell)
Chiba et al. 1990	40mm: 4mm	Shear thinning (0.5 wt% PAA)	51.5	Not evaluated	N.A.	Not evaluated
Yesilata et al. 1999	25.4 mm: 6.35 mm	Boger fluids (0.31% polyisobutylene)	< 0.011	12	>1090.9	Oscillatory measurements (Maxwell)
Alves et al. 2005	24 mm: 6 mm	Boger fluid (PAA 100)	0.9	52	57.8	Oldroyd-B model
		Boger fluid (PAA 300)	0.49	46.7	95.3	
Alves et al. 2008	24 mm: 6 mm	Shear thinning (PAA 500)	0.183	109	595.6	4-mode PTT model

Table 2-2. Conditions for onset of unstable flows in micro-contractions.

Micro-contraction flows						
Investigator	Contraction ratio	Fluids used	Onset of time-dependent unstable flow			Method of obtaining relaxation time (λ)
			Re	De/Wi	El	
Gan et al. 2007	1 mm:125 μ m:1 mm	Shear thinning (1wt% PEO,0.1wt% PEO)	0.025	203.5	8140	CaBER measurements
Rodd et al. 2005a	400 μ m: 25 μ m: 400 μ m	Mild shear thinning (0.05wt%,0.1wt%,0.3wt% PEO)	$Re_{0.05wt\%} \approx 32$, $Re_{0.1wt\%} \approx 14$, $Re_{0.3wt\%} \approx 1.3$.	120	> 92.3	CaBER measurements
Rodd et al. 2007	400 μ m: 25 μ m: 400 μ m	Constant viscosity elastic (0.075wt% PEO)	0.76	179.7	236.4	CaBER measurements

Table 2-3. Conditions for onset of instability and quantification techniques used.

Viscoelastic turbulent flows							
Investigator	Dimensions	Fluids used	Onset of time-dependent unstable flow			Method of obtaining relaxation time (λ)	Quantification methods used.
			Re	De/Wi	El		
Groisman and Steinberg 2000	R1 =38 mm (rotating plate), R2=43.6 mm (cylinder), depth =10 mm	Dilute shear thinning PAA solutions (80ppm)	0.3	3.5	11.7	Phase shift between stress and shear rate in oscillatory experiments.	Torque measurements, LDA velocity measurements, velocity power spectra, light intensity measurements
Groisman and Steinberg 2001a	3mm by 3mm	Dilute shear thinning PAA solutions (80ppm)	0.06	3.2	53.3	Phase shift between stress and shear rate in oscillatory experiments.	LDA velocity measurements, mixing efficiency, probability density functions, velocity power spectra.
Pathak et al. 2004	Width = 85 μ m, depth = 60 μ m	Constant viscosity dilute PAA solution (0.0066 wt%)	not stated	not stated	N.A.	Cross over point for G' and G'' curves	Flow/streakline visualizations, mixing efficiency.
Burghilea et al. 2004	100 μ m width and depth	Constant viscosity solution 1 (80 ppm PAA)	0.017	1.4	82.4	Phase shift between stress and shear rate in oscillatory experiments.	Pressure measurements, mixing efficiency, PIV velocity measurements, root mean square of velocity fluctuations, autocorrelation function.
		Shear thinning solution 2 (80 ppm PAA)	0.00008	3.5	43750		

Arratia et al. 2006	650 μm width, 500 μm depth	Boger fluids (200 ppm PAA)	< 0.01	12.5	> 1250	Oldroyd-B model	PIV velocity measurements, velocity power spectra, mixing efficiency.
Gan et al. 2007, Lam et al. 2009, Gan and Lam 2012	1 mm:125 μm :1 mm	Shear thinning (1wt% PEO,0.1wt% PEO)	0.025	203.5	8140	CaBER measurements	Mixing efficiency, PIV velocity measurements.
Soulages et al. 2009	50 μm by 50 μm – t-shape	Weakly shear thinning (0.075wt% PEO)	0.0041	3.2	780.5	CaBER measurements	Flow/streakline visualizations
	t-shape with cavity		0.0068	5.87	863.2		
XI and Graham 2009	Cross slot = 1 x 1	FENE-P	0.1	66	660	FENE-P	Birefringence calculation, velocity calculations, trace of conformation tensor
Li et al. 2010	60 μm and a width of 300 μm	Shear thinning (1000 ppm CTAC)	0.05	58.1	1162	Giesekus model	Flow/streakline visualizations
	a disc cavity (ϕ = 1mm, depth = 60 μm)		1.4	144.8	103.4		
	width = 200 μm , depth = 50 μm	Shear thinning (200 ppm CTAC)	0.1	7.5	75		
Liu and Steinberg 2010	R = 2.25 mm, R2 = 6mm, d = 675 μm	Shear thinning (6.29 – 100.64 $\mu\text{g/mL}$ T4GT7 DNA)	$\text{Re} \ll 1$	68.9 – 3.1 (6.29 – 100.64 $\mu\text{g/mL}$)	$\gg 31$	Measured by extension of DNA by $> 30\%$ in a shear flow, after flow was stopped	DNA length measurement, Stress approximation from worm-like chain model, probability density function of length and approximated stress.

Bonn et al. 2011	Expansion ratio 1 mm/3.5 mm : 5 mm/8 mm	Shear thinning (100 – 1000 ppm PAA)	$\approx 0.4 - 0.8$	Not stated	-	Carreau model	LDA velocity measurements, velocity power spectra, probability density function of velocity measurements
Jun and Steinberg 2011	1mm by 1mm	Dilute shear thinning PAA solutions (80ppm)	< 0.54	200	> 370	Stress relaxation method	Pressure drop measurements, PIV velocity measurements, root mean square of velocity and velocity gradient, correlation lengths, probability density function.
Tatsumi et al. 2011	84 μm x 60 μm	Shear thinning (100 ppm, 18×10^6 g/mol)	6.7×10^{-4}	Not computed	-	-	Mixing efficiency, PIV velocity measurements, turbulent intensity, velocity power spectrum.
Haward et al. 2012	0.2 mm x 1 mm	Shear thinning (100 mM CPyCl)	$< 10^{-5}$	1.6	$> 160\,000$	Oscillatory measurements (Maxwell)	PIV velocity measurements, pressure drop measurements, birefringent measurements.
Grilli et al. 2013	(3-D simulation) Straight channel 10 x 2 x 5 64 x 64 x 64 grid	Oldroyd-B	0.024	1	41.7	Oldroyd-B	Power spectrum, mixing efficiency, trace of conformation tensor.

Zhang et al. 2013	(2-D simulation) $R_{\text{cylinder}} = 1$ $H_{\text{channel}} = 2$	Giesekus	1	5	5	Giesekus	Turbulent intensity, power spectra, mixing efficiency, vorticity, trace of conformation tensor, probability density function.
Cruz et al. 2014	(2-D simulation) cross-slot = 1 x 1 Number of cells: 12,801 – 201,201	UCM, Oldroyd-B, sPTT	0	UCM: 0.335 Oldroyd-B: 0.425 sPTT: 0.925	-	UCM, Oldroyd-B, sPTT	First normal stress difference, pressure drop calculations, asymmetry parameter

3. High-Re Turbulence

3.1 Introduction

3.1.1 Background

What is turbulence? Perhaps the easiest way to visualize this phenomenon is to imagine a flow with significant ‘furry’ fluctuations in its velocity profile. This fluctuation is random about the mean velocity, exhibiting no sign of periodicity. Turbulence has the ability to transport and mix fluids much better than laminar flows as demonstrated by Osborne Reynolds ([Reynolds 1883](#)), and is also useful in heat transfer. However, the large increase in resistance to flow as a result of excessive energy dissipation is not always desirable, because much more energy is required to maintain the flow. Thus, for such cases where too much energy dissipated in turbulence is undesirable, small concentrations of long chain polymers i.e. drag reducing polymers (DRPs) can be added to reduce friction drag in wall bounded turbulence ([L’vov et al. 2004](#), [Gillissen 2008](#), [Xiong et al. 2010](#), [Karami and Mowla 2012](#), [Khadom and Abdul-Hadi 2014](#)), and reduce turbulent intensities at high Reynolds numbers ($Re > 2000$) ([Oliveira 2001](#), [Pipe and Monkewtiz 2006](#), [Tamano et al. 2009](#), [Japper-Jaafar et al. 2010](#)). This is known as the Tom’s effect, or polymer drag reduction ([Toms 1948](#)).

The classical definition of turbulence states that it occurs when the Reynolds number is high, e.g. above 2300 in pipe flow, and much experimental research on turbulence flows have been conducted in the order of this magnitude. Even though some works have stated the early onset of turbulence due to surface roughness of channel at

Reynolds number of 300-700 (Peng et al. 1994), this is still a fairly large Reynolds number to be achieved in micro-channels. Hitherto there is no unique set of criteria which defines the term ‘turbulence’ although some common experimental observations have been accepted to indicate the presence of turbulence.

In recent years, many experimental works have been conducted which demonstrate the possibility of generating turbulence in the absence of inertia ($Re \ll 1$), by exploiting the elastic effects of viscoelastic fluids which contain only small amounts of polymer additives. This was termed ‘elastic turbulence’ (Vinogradov and Ivanova 1968) and the related literature has been reviewed in Chapter 2. The observed ‘elastic turbulence’ exhibits similar features as that of inertia-driven turbulence such as significant non-periodic velocity fluctuations, increase resistance to flow, power law decay spectra and significant improvement in mixing even in micro-channels (see Chapter 2). However, a comprehensive quantification of the elastic-driven turbulent flow field is absent.

3.1.2 Outline

To gain some understanding on the manifestation of high-Re turbulence from a mathematical standpoint, the governing equations in fluid dynamics and viscoelastic models will be introduced in Section 3.2. The role of the Deborah number in viscoelastic turbulence, and the limitations of existing studies (see Chapter 2) will be recapped in Section 3.3. Next, the characteristics of high-Re turbulence and some existing measurement techniques will be discussed in Section 3.4. Based on these measurement techniques, the parameters typically employed for the quantification of

high-Re turbulence will be reviewed in Section 3.5. These techniques and parameters will be subsequently employed to quantify the viscoelastic turbulent flow field, which is an objective of this study. Lastly, a review of experimental works on high-Re wall bounded turbulence will be included in Section 3.6.

3.2 Governing equations

Before examining the details of turbulence, the governing equations in fluid dynamics are introduced in this section. Note that it is not the focus of this study to solve these equations numerically but more for a basic appreciation of parameters governing a flow field.

For an isotropic and incompressible flow, the governing equations for conservation of mass and momentum is

$$\nabla \cdot \mathbf{U} = 0 \quad (3.1)$$

$$\rho \left[\frac{\partial \mathbf{U}}{\partial t} + \mathbf{U} \cdot \nabla \mathbf{U} \right] = -\nabla P + \nabla \cdot \boldsymbol{\tau} + \mathbf{F} \quad (3.2)$$

where ρ is the density of the fluid, $\mathbf{U}$ the velocity vector, P the pressure field, $\boldsymbol{\tau}$ the total extra stress tensor, and $\mathbf{F}$ the body force which is typically only due to gravity. Equation 3.2 is also known as the Cauchy momentum equation. The first term on the left, $\rho \frac{\partial \mathbf{U}}{\partial t}$, is the inertial acceleration term and the second term on the right, $\nabla \cdot \boldsymbol{\tau}$, is the dissipative viscous term related to the fluid in use. The non-linear convective term, $\rho \mathbf{U} \cdot \nabla \mathbf{U}$, is also inertial in nature. This non-linear term can be caused by a change in velocity with respect to position, or due to a change in geometry. It can be immediately

observed that this non-linear term vanishes, in a fully developed flow because the velocity profile is not a function of the flow direction. This term is related to the turbulent Reynolds stresses and becomes significant in turbulent flows.

3.2.1 Navier-Stokes equation

To describe the flow field, a constitutive relationship for the fluid must be defined. For a Newtonian fluid, the viscous stress tensor is simply

$$\boldsymbol{\tau} = \eta[\nabla \mathbf{U} + (\nabla \mathbf{U})^T] \quad (3.3)$$

Where η is the solvent viscosity.

Note that by substituting equation 3.3 into equation 3.2 and neglecting gravitational forces, one obtains the Navier-Stokes equation (equation 3.4).

$$\rho \left[\frac{\partial \mathbf{U}}{\partial t} + \mathbf{U} \cdot \nabla \mathbf{U} \right] = -\nabla P + \eta \nabla^2 \mathbf{U} \quad (3.4)$$

One typically solves this equation to obtain the velocity flow field which can then be used for computing other parameters such as the flow rate, drag forces or stresses. Some solutions of the Navier-Stokes equation include Poiseuille flow, Couette flow etc., where the non-linear term was set to zero.

In microfluidic flow applications, the inertial terms are usually negligible (i.e. $\text{Re} \rightarrow 0$) due to the constraints in dimensions and velocity, and generally termed as ‘creeping’ flows. Ignoring gravitational effects, the Cauchy momentum equation then becomes the Stokes’ equation given by

$$0 = -\nabla P + \nabla \cdot \boldsymbol{\tau} \quad (3.5)$$

However, at high Reynolds number leading to turbulent flow conditions, it is impossible to solve equation 3.4. Thus, this has led to the practice of averaging these equations and solving for the resulting equations for mean quantities, using the Reynolds-averaged Navier-Stokes (RANS) equation defined as

$$\rho \left[\frac{\partial \bar{U}_i}{\partial t} + \bar{U}_j \frac{\partial \bar{U}_i}{\partial x_j} \right] = -\frac{\partial \bar{P}}{\partial x_i} + \eta \nabla^2 \bar{U}_i - \frac{\partial (\rho \overline{u_i u_j})}{\partial x_j} \quad (3.6)$$

where uppercase letters $\bar{U}_i$, $\bar{U}_j$ and $\bar{P}$ denote the mean components, and lowercase letters u_i and u_j denote the fluctuating components. Note that equation 3.6 is similar to equation 3.4 apart from the Reynolds stress term, $\overline{u_i u_j}$. This non-linear stress term requires additional modelling to obtain closure to the RANS equation for solving, since there is no direct means for relating $u_i u_j$ to $\bar{U}_i$ and $\bar{P}$. Thus, many RANS turbulence models have been created, i.e. eddy-viscosity models (EVM), non-linear eddy-viscosity models (NLEVM) and differential stress models (DSM). However, they are not the focus of this study.

Other methods for direct computing of turbulent flows include Large-Eddy Simulations (LES) where modeling is only applied for small scale eddies, or Direct Numerical Simulations (DNS) whereby no modeling is applied at all. In DNS, turbulent phenomena at all length and time scales can be resolved by simply solving the Navier-Stokes and continuity equations numerically. However, large computational costs, in terms of memory space and time, are involved.

3.2.2 Viscoelastic models

There are many constitutive viscoelastic models for representing the extra stress tensor $\boldsymbol{\tau}$, including the Upper Convected Maxwell (UCM) model, Giesekus model, Phan-Thein Tanner (PTT) model etc. The two main types of models used for modelling polymer solutions or melts are the dumb bell model (e.g. UCM, Giesekus etc) and the network theory model (e.g. PTT). A detailed discussion of these models will not be provided here as it is not the focus of this study; instead interested readers are referred to references by [Bird et al. \(1987\)](#), [Barnes et al. \(1989\)](#) and [Oswald \(2009\)](#). However depending on the flow to be modelled, each of these models has their own advantages and drawbacks. For example, the UCM model has only one relaxation time and makes qualitative correct predictions in the linear regime only. It predicts the appearance of the first normal stress difference but not the second normal stress difference. In addition, it wrongly predicts the divergence of extensional viscosity with elongation rate and does not account for shear thinning behaviour. Nevertheless, this model is very useful for modelling rheological measurements which are usually carried out in the linear regime.

The constitutive UCM equation ([Oswald 2009](#)) for defining stress tensor $\boldsymbol{\tau}$ is

$$\boldsymbol{\tau} + \lambda \left[\frac{\partial \boldsymbol{\tau}}{\partial t} + \nabla \cdot \mathbf{U} \boldsymbol{\tau} \right] = \lambda (\boldsymbol{\tau} \cdot \nabla \mathbf{U} + \nabla \mathbf{U}^T \cdot \boldsymbol{\tau}) + \eta [\nabla \mathbf{U} + (\nabla \mathbf{U})^T] \quad (3.7)$$

Or

$$\lambda \tilde{\boldsymbol{\tau}} + \boldsymbol{\tau} = 2\eta \mathbf{D} \quad (3.8)$$

where $\mathbf{D} = \frac{1}{2}[\nabla \mathbf{U} + (\nabla \mathbf{U})^T]$ is the rate-of-deformation tensor, λ is the relaxation time and $\tilde{\boldsymbol{\tau}}$ is the upper-convected time derivative defined as

$$\tilde{\boldsymbol{\tau}} = \frac{\partial \boldsymbol{\tau}}{\partial t} + \nabla \cdot (\mathbf{U} \boldsymbol{\tau}) - (\nabla \mathbf{U})^T \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \nabla \mathbf{U} \quad (3.9)$$

For better data fitting, superposition of non-linear models such as the Giesekus and PTT models with increased number of modes usually results in an improvement in providing a reasonably good fit. However, the drawback of using non-linear multi-mode models is the large amount of computational time required in numerical simulations. For completeness, the constitutive equation of the Giesekus model for the extra stress tensor is presented here. The Giesekus model originates from the Jeffrey model. In a dilute polymeric solution, the stress tensor can be expressed as a sum of its polymer (elastic) component and its solvent (Newtonian) component as

$$\boldsymbol{\tau} = \boldsymbol{\tau}_p + \boldsymbol{\tau}_s \quad (3.10)$$

where $\boldsymbol{\tau}_p$ is the elastic stress tensor due to the polymeric component, and $\boldsymbol{\tau}_s$ is the viscous stress tensor due to the Newtonian solvent, which is given by

$$\boldsymbol{\tau}_s = \eta_s(\nabla \mathbf{U} + \nabla \mathbf{U}^T) = 2\eta_s \mathbf{D} \quad (3.11)$$

where η_s is the Newtonian solvent viscosity.

The Giesekus constitutive equation (Bird et al. 1987) is defined as

$$\boldsymbol{\tau}_p + \lambda_p \tilde{\boldsymbol{\tau}}_p + a \frac{\lambda_p}{\eta_p} (\boldsymbol{\tau}_p \cdot \boldsymbol{\tau}_p) = 2\eta_p \mathbf{D} \quad (3.12)$$

where λ_p is the single mode polymer relaxation time, a is the mobility factor associated with Brownian motion and anisotropic hydrodynamic drag on the polymer molecules, η_p the polymer contribution to the zero-shear rate viscosity (η_o) given by

$$\eta_o = \eta_s + \eta_p \quad (3.13)$$

Note that if $a = 0$, the UCM model (equation 3.8) is recovered.

In general, multi modes are required to improve the fitting of the constitutive model to physical experimental data. Thus, it is required to introduce a spectrum of viscoelastic modes whereby the total polymeric contribution to the extra stress tensor is now given by

$$\boldsymbol{\tau}_p = \sum_i \boldsymbol{\tau}_{p_i} \quad (3.14)$$

where each mode $\boldsymbol{\tau}_{p_i}$ obeys the equation of similar form to equation 3.12, and is given by

$$\boldsymbol{\tau}_{p_i} + \lambda_{p_i} \tilde{\boldsymbol{\tau}}_{p_i} + a \frac{\lambda_{p_i}}{\eta_{p_i}} (\boldsymbol{\tau}_{p_i} \cdot \boldsymbol{\tau}_{p_i}) = 2\eta_{p_i} \mathbf{D} \quad (3.15)$$

In general if a is chosen appropriately ($0 < a < 1/2$), this equation is capable of predicting non-linear shear-thinning behaviours at high shear rates and the saturation of elongational viscosity at high elongation rates. It can be proven that the Weissenberg number (Wi) is the coefficients of non-linear stress terms of equation 3.15 in dimensionless forms, just like the Reynolds number (Re) being the coefficient of the non-linear velocity terms in the dimensionless Navier-Stokes equation.

To solve numerically for the velocity flow fields and other flow parameters, equations 3.10, 3.11, 3.13, 3.14 and 3.15 must all be solved (assuming a multi-mode Giesekus model is used) and substituted into the Cauchy momentum equation with the respective boundary conditions; this rigorous process may be performed using high speed computers. However, the possibility of employing viscoelastic models to model non-linear behaviour does not mean that turbulence can be modelled accurately with computational fluid dynamics (CFD) software. Hitherto, modelling elastic-driven turbulence in the absence of inertia (i.e. $Re < 1$) due to viscoelastic flows in micro-channels remains challenging. This is also known as the high Weissenberg number problem (HWNP) (Keunings 1986, Abedijaberi and Khomami 2011, Renardy 2014).

For completeness, the time-averaged equation for an incompressible viscoelastic fluid is of the form

$$\rho \left[\frac{\partial \bar{U}_i}{\partial t} + \bar{U}_j \frac{\partial \bar{U}_i}{\partial x_j} \right] = - \frac{\partial \bar{P}}{\partial x_i} + \eta_s \nabla^2 \bar{U}_i + - \frac{\partial (\rho \bar{u}_i \bar{u}_j)}{\partial x_j} + \frac{\partial \bar{\tau}_{ij,p}}{\partial x_j} \quad (3.16)$$

where $\bar{\tau}_{ij,p}$ is the time-averaged polymer stress tensor, which can be defined by the model chosen. Note that the stress is made up of the sum of Newtonian solvent and polymeric contribution.

3.3 Viscoelastic turbulence in micro-channels

It is known that large Re dictates the onset of inertial-driven turbulence due to strong non-linear inertial effects, while a small Re equates to laminar flow. However, is it possible that a large De or Wi will also result in turbulent flow at diminishing $Re (< 1)$,

since it is also related to non-linear stress terms based on the extra stress tensor equation? Indeed, it has been shown in many studies that the possibility of elastic-driven instability/turbulence truly exists even in the absence of inertia, and is dictated by Deborah and Weissenberg numbers (refer to Chapter 2). However, a review of works on viscoelastic turbulence has shown that most of these studies do not provide a complete quantitative analysis regarding the phenomenon they have observed in their setup. One of the few who has made such an attempt was by [Jun and Steinberg \(2011\)](#), who presented data on the velocity fluctuations and velocity gradient in a curvilinear channel, and identified a boundary layer close to the channel wall. Based on their spatial correlation function, they stated that the bulk flow consisted of spatial scales on the order of the boundary layer thickness. However, no data on the velocity power spectra, or evidence of the calculated spatial scales via eddy visualization techniques were presented.

As aforementioned, there have been few attempts in correlating the similarity between viscoelastic (elastic-driven) turbulence to the more commonly observed high-Re (inertia-driven) turbulence. In addition, such detailed statistical analyses have yet to be reported for micro-channel (contraction-expansion) studies. To bridge the gap between elastic-driven turbulence and inertial-driven turbulence, it is necessary to understand the various methods of describing and quantifying turbulent flows at high Reynolds number. These will be described in Section 3.5.

3.4 Characteristics of high-Re turbulent flows

3.4.1 3-D turbulence

The two most famous concepts in turbulence are Richardson's energy cascade and the Kolmogorov's hypothesis (Pope 2003). According to Richardson's view of the energy cascade, turbulence occurs at high Reynolds number and consists of eddies of various sizes, large and small. In 3-D turbulence, this turbulent energy is transferred from the largest to the smallest eddies, whereby the kinetic energy is eventually dissipated because Reynolds number is small and viscous effects are important. Similar to Richardson, Kolmogorov's hypotheses are also based on the assumption of large Reynolds number flows. He stated that the lengthscales of these eddies fall into three ranges, the energy-containing range, the inertial subrange and the dissipation range. The largest eddy lengthscale is found in the energy-containing range, with the smallest being found in the dissipation range. Viscous effects are negligible in the inertial subrange, but significant in the dissipation range. The dissipation range is assumed to be responsible for almost all of the energy dissipation. At sufficiently high Reynolds number, statistics of the small scale turbulent motions are isotropic and is uniquely determined by the viscosity and the dissipation rate. In 3-D turbulence, energy is conserved but not enstrophy (mean squared vorticity). In addition, vortex stretching is present.

In the inertial subrange, the energy flux is such that energy that is injected into the system to create this turbulent flow is balanced by energy dissipation. According to Kolmogorov's theory, the energy and power spectrum in the inertial subrange of 3-D

turbulence follows a $-5/3$ power law scaling (see Figure 3-1). The isotropic energy spectrum, $E(k)$, can be expressed as

$$E(k) = C \epsilon^{\frac{2}{3}} k^{-\frac{5}{3}} \quad (3.17)$$

where C is a constant, ϵ the mean rate of kinetic energy transfer per unit mass, and k is the wave number.

Figure 3-1. Power spectrum of a typical 3-D turbulent flow and its energy ranges (Pope 2003).

3.4.2 2-D Turbulence

However in 2-D turbulent flows (typically aspect ratio $\gg 1$), two cascades exist. Energy is transferred from smaller to larger scales by vortex merger (i.e. inverse energy cascade) and is subsequently dissipated by external friction, while enstrophy (mean squared vorticity) is cascaded to smaller scales and dissipated by viscosity. This is a

significant difference as compared to 3-D turbulence, where all energy is assumed to be dissipated at the small scales by viscosity. Energy and enstrophy are both conserved in 2-D turbulence, and there is no vortex stretching. The energy spectrum of 2-D turbulence has been predicted to have two scalings, a power law decay of $-5/3$ for larger scales and -3 for smaller scales (Kraichnan 1967, Leith 1968, Batchelor 1969). This is depicted in Figure 3-2 (Boffetta 2007). The sizes of the largest vortices are usually constrained due to external friction as a result of a balance between the energy flux from the inverse cascade and the fluid drag forces. Otherwise, the vortices could grow up to the system size.

For 2-D turbulence, the inertial range consists of an energy transfer range which is similar to that of 3-D turbulence, whereby the isotropic energy spectrum, can be expressed as

$$E(k) = C\epsilon^{\frac{2}{3}}k^{-\frac{5}{3}} \quad (3.18)$$

and an enstrophy transfer range where it is described by

$$E(k) = C'b^{\frac{2}{3}}k^{-3} \quad (3.19)$$

Where C' is a constant, b the mean rate of enstrophy transfer per unit mass, and other terms being similar to that in equation 3.17. Note that there is zero enstrophy transfer in the energy transfer range, and there is zero kinetic energy transfer in the enstrophy transfer range.

Figure 3-2. Power spectrum of a typical 2-D turbulent flow showing the two inertial ranges (Boffetta 2007).

3.4.3 Characteristics and measurement techniques

Within the review of published research works regarding elastic-driven turbulence in Chapter 2, a detailed analysis and verification of 2-D or 3-D turbulence has not been reported in micro-channels for small Reynolds number ($Re < 1$), whereby Kolmogorov's hypothesis may not be valid. Indeed, experiments or Direct Numerical Simulation (DNS) that have indicated turbulence in micro-channels mostly occur at high Reynolds number (up to > 3000), with the working fluid being air or water. These are reviewed in Section 3.6.

Much of the reported works have claimed that turbulence is a result of flows deviating from laminar and steady flow-states, whereby velocity profiles are predictable. Methods for identifying transition from laminar to turbulence flow include the observations of the deviations of the friction factor-Reynolds number product with

increasing flow rates (i.e. $fRe = 64$ in laminar flow), fluctuating pressure drop measurements and deviation from linearity with increasing Re , increase in root-mean-square (RMS) velocity fluctuation etc. Others investigated the rise in fluid temperature by measuring the outlet temperatures ([Rands et al. 2006](#)), but this might not be an accurate way to claim the onset of turbulence. Some have reported the plots of autocorrelation functions with space and time to illustrate the randomness of the velocity field as an indication of turbulence. Another observed phenomenon of turbulence is the power law decay in the energy or power spectrum with frequency and wavenumber ([Hädrićh 2001](#), [Pope 2003](#)), which is different for 2-D and 3-D turbulence. Examples of these characteristics of inertial-driven turbulent flows have also been included in Section 3.6.

Measurements of velocity fluctuations are typically performed with stationary hot-wires, Laser Doppler Anemometry (LDA) or Particle Image Velocimetry (PIV) systems. For micro-channels, PIV is a more appropriate method for the measurement of velocity fluctuations, since hot-wires are intrusive and LDA spot sizes might be too large. These measurements can be performed at a single point over time or more points if spatial correlations are to be carried out. For a measurement at a single point, it is statistical stationary if the properties, i.e. mean and variance, do not change with time. Much of the theories on turbulence are made based on the assumption that the turbulence is homogenous, i.e. statistically stationary in space. The joint PDF of velocity statistics of n points should remain unchanged if they are shifted by the same

constant displacement. However, it is idealistic to assume that turbulent flows are homogenous in reality.

As mentioned in Section 3.4.1, Richardson and Kolmogorov's hypotheses of turbulence indicated the presence of eddies of varying sizes. Hence, some reported works have indicated the presence of eddies in their flow visualizations to justify the transition to, and presence of turbulence (Adrian et al. 2000). As these eddies are usually not stationary in space, they must be viewed in the same frame which moves at the same velocity as the vortex core. Reynolds decomposition of velocity field data, whereby velocity is separated into its mean and fluctuating component, is used for revealing small scale vortices and is more appropriate for statistical analysis. However, it removes large-scale features. Thus in order to better visualize the turbulent velocity fields, most reported works employ the usage of Galilean decomposition and CFD that employs Large-Eddy Simulation (LES) decomposition. The LES method involves subtracting a filtered velocity field from the total velocity field and is a better eddy visualization in comparison with the Reynolds and Galilean decomposition. However, the Galilean decomposition is normally employed as it is simple, easily applicable on experimental data, and provides a relatively good result. It involves the subtraction of a convective velocity (U) from the total velocity field. Figure 3-3 shows an example of Galilean decomposition by subtraction of various percentage of the convective velocity (U_{CL}) to reveal an eddy. Note that these two decomposition techniques are purely used for visualization purposes and Reynolds decomposition is still the primary method for turbulent data analysis.

Figure 3-3. Visualization of an isolated turbulent eddy by Galilean decomposition by subtracting 3 different convection velocities (Adrian et al. 2000) a) $U = 0.65 U_{CL}$ b) $U = 0.78 U_{CL}$ c) $U = 0.91 U_{CL}$

In addition to visualizing instantaneous flow field structures e.g. turbulent eddies, it is often desirable to simplify a complex flow field. Hence, proper orthogonal decomposition (POD) can be employed to analyse dynamical flow structures in turbulent flows (Lumley 1967). The technique decomposes (i.e. breaks down) complex flows into simple spatial modes to identify the coherent structures within the turbulent flows. One of the most commonly used POD methods is the snapshot POD technique by Sirovich (1987), which requires velocimetry data obtained from instantaneous PIV measurements i.e. snapshots. Coherent structures containing the highest turbulent kinetic energy can then be retrieved, which may be viewed as ‘dominant’ structures/modes in the complex flow field. Thereafter, snapshots of the actual flow can also be reconstructed with great accuracy using only the most dominant modes. This technique will be illustrated in one of the reviewed works in Section 3.6.

3.5 Parameters in turbulence quantification

In the preceding section, some characteristics of turbulent flows have already been discussed. Statistical methods are usually employed to determine the transition and occurrence of turbulence from laminar flow. Before a review is performed on selected works regarding turbulent flows in Section 3.6, some terms which are used for describing turbulent flows have to be defined for better understanding of this study and the review of works. The definition of these terms will be presented in this section.

Consider N measurements of a single point velocity over a period of time (T), the sampling frequency (f_s) is defined as,

$$f_s = \frac{N}{T} \quad (3.20)$$

where T is in seconds and f_s is in Hz. Measurements using LDA are usually capable of sampling rates up to 1 kHz while that of hot-wires may reach up to 1 MHz. Although technology advancement has resulted in optical PIV systems to attain a sampling frequency of up to 1 MHz with the developments of high-speed CMOS cameras, these systems are usually much more expensive than the LDA and hot-wire setups.

Figure 3-4 shows a single point temporal velocity measurement and point A shows the measured velocity (U_i) at a particular instant. By computing the average of all the measured velocity data, the mean velocity ($\bar{U}$) can be evaluated as

$$\bar{U} = \frac{1}{N} \sum_{i=1}^N U_i \quad (3.21)$$

which is the average of all the data values at the *single point* where it is measured (see Figure 3-4). Here, U_i corresponds to a one point velocity datum measured at one time instant and it can be defined to be composed of the mean velocity and the fluctuation velocity component, u_i (see Figure 3-4).

$$U_i = \bar{U} + u_i \quad (3.22)$$

With the measured velocities, the amplitude of fluctuations with reference to the mean velocity can be obtained. This measurement is termed as the root mean square velocity (u_{rms}) which is defined as

$$u_{rms} = \left(\frac{1}{N-1} \sum_{i=1}^N (U_i - \bar{U})^2 \right)^{1/2} \quad (3.23)$$

It provides information regarding the spread of the collected data points. This is also known as the standard deviation of the distribution and is used to calculate the

turbulence intensity (Tu), which is the normalized data spread based on the mean velocity ($\bar{U}$) at the specific point of measurement.

$$Tu = \frac{u_{rms}}{\bar{U}} \quad (3.24)$$

The turbulent fluctuations give rise to additional stresses i.e. Reynolds stresses (τ_{ij}^R). They represent the average flux of momentum due to the turbulent fluctuations (or random motion of eddies). The Reynolds stresses can also be interpreted to be the momentum exchange between the mean flow and the turbulence. In reported works, the Reynolds stresses is defined without the density coefficient as

$$\tau_{ij}^R = -\overline{u_i u_j} \quad (3.25)$$

Two other quantities for describing the velocity data are the Skewness (S) and Kurtosis factor (K). The Skewness measures if the data points are more favoured towards one side of the mean i.e. lack of statistical symmetry, while the Kurtosis is a measure of the flatness of the probability density function (PDF) of the velocity data. Higher Kurtosis means a higher peak at the mean and extreme deviations from the mean are infrequent.

These two factors are defined as

$$S = \frac{1}{Nu_{rms}^3} \sum_{i=1}^N (U_i - \bar{U})^3 \quad (3.26)$$

$$K = \frac{1}{Nu_{rms}^4} \sum_{i=1}^N (U_i - \bar{U})^4 \quad (3.27)$$

In turbulence, the single point velocity measurement is random in space and time. To study the correlation between the measurements obtained at a particular time (t) with another instant ($t + s$), i.e. points A and B in Figure 3-4, the autocorrelation function is used. This function can be interpreted as how ‘similar’ the velocity fluctuation at one point in time is to that measured previously. The normalized autocorrelation function, $\rho(s)$, is defined as

$$\rho(s) = \frac{R(s)}{u_{rms}^2} = \frac{\overline{u(t)u(t+s)}}{u_{rms}^2} \quad (3.28)$$

where s denotes the time lag and $R(s)$ is the autocovariance. In turbulent flows, $\rho(s)$ drops rapidly and fluctuates about zero with increasing time lag.

Fourier spectra in time and space are frequently used for characterizing the distribution of velocity fluctuations over different length and time scales. According to the Wiener-Khinchin theorem (Pope 2003), the temporal power spectrum, $S(f)$ for a stationary random process, can be obtained through a Fourier transformation of the autocovariance, $R(s)$, given by

$$S(f) = \frac{1}{\pi} \int_{-\infty}^{\infty} R(s) e^{-i2\pi fs} ds \quad (3.29)$$

where f denotes the frequency. The temporal power spectrum describes how the power of the fluctuations is distributed with frequency.

In turbulence, eddies evolve in time and space even though they are advected with the main flow. However, it is not practical to have hot-wires positioned everywhere to

capture the size of these eddies. Neither is it easy nor economical to capture a large field of view with PIV. One solution to these difficulties is based on Taylor's Hypothesis (Pope 2003), which states that turbulence can be considered 'frozen' as it passes by a point or sensor. This hypothesis is valid if the turbulent eddies evolve at a time scale much longer than the advection flow rate past that point, and this criteria (Willis and Deardorff 1976) can be written as

$$u_{rms} < 0.5\bar{U} \quad (3.30)$$

Thus, a size distribution of the eddies may be presented using a spatial power spectrum, $S(k)$, which indicates the energy to wave number distribution, and follows a power law decay with an exponent of -5/3 in the inertial subrange. This spectrum may be directly obtained from the temporal power spectrum with the proper substitution of

$$f = \frac{\bar{U}k}{2\pi} \quad (3.31)$$

where k is the wave number.

Spatial correlation of the velocity components in different regions reveals if the statistical properties in these regions are correlated or 'similar' to each other. In turbulent flows, they may be used to calculate the length scales, i.e. size of turbulent eddies, of the flow. This can be done if there are measurements made at two different positions at the same time, e.g. 2 stationary hotwires, large area PIV. However, if Taylor's hypothesis is valid, the spatial correlation would be equivalent to that obtained temporally and can be measured at a point. Usually, spatial correlation is evaluated

separately in one direction at a time, e.g. in the axial and transverse direction. The autocorrelation functions can be described by

Axial correlation function, $f(x)$:-

$$f(x) = \frac{R_{xx}(x)}{\langle u_{rms}^2 \rangle} = \frac{\langle u(x_o, y_o) u(x_o + x, y_o) \rangle}{\sqrt{\langle u^2(x_o, y_o) \rangle} \sqrt{\langle u^2(x_o + x, y_o) \rangle}} \quad (3.32)$$

Transverse correlation function, $g(y)$:-

$$g(y) = \frac{R_{yy}(y)}{\langle u_{rms}^2 \rangle} = \frac{\langle u(x_o, y_o) u(x_o, y_o + y) \rangle}{\sqrt{\langle u^2(x_o, y_o) \rangle} \sqrt{\langle u^2(x_o, y_o + y) \rangle}} \quad (3.33)$$

where $\langle \rangle$ represents the ensemble average, which is estimated using the time average in the current analysis. Hence the 1-D axial spectra, $E_{xx}(k)$, and transverse spectra, $E_{yy}(k)$, can be written as

$$E_{xx}(k) = \frac{1}{\pi} \int_{-\infty}^{\infty} R_{xx}(x) e^{-ikx} dx \quad (3.34)$$

$$E_{yy}(k) = \frac{1}{\pi} \int_{-\infty}^{\infty} R_{yy}(y) e^{-iky} dy \quad (3.35)$$

Note that this is similar to equation 3.29 if Taylor's frozen-turbulence hypothesis is valid.

3.6 Review of recent works

3.6.1 Introduction

Much of the research and turbulence measurements have been conducted in wind tunnels and large pipes. This is because in bounded flows, it is generally harder to achieve high Reynolds number with miniaturization. The focus of this review is not on turbulence modelling, but to review the measurements and analyses typically involved in defining this phenomenon. A large extent of research has already been carried out, but only a few of the recent investigations will be listed, with a focus on bounded channel flows which are more relevant to the present investigations. In addition, viscoelastic turbulent flows in channels have not been included in this section as most of them have already been reviewed in Chapter 2.

3.6.2. Brief review on 3-D turbulent flows

Majority of the earlier experiments on turbulence were based merely on pressure drop, and descriptions and observations of velocity fluctuations. Velocity correlations, power density spectra and eddy visualizations were incorporated in more recent works. [Zeighami et al. \(2000\)](#) took the repeatability of the velocity data as the criterion to distinguish the transition from laminar to turbulent flow for water flows in planar micro-channels with a height of 150 μm and width of 100 μm . [Sharp and Adrian \(2004\)](#) stated the onset of transition to turbulence occurred at Reynolds numbers in between 1800 and 2000, for flows of fluids (water, 1-propanol etc.) in 50 to 247 μm diameter round glass channels. This was based on the deviation of pressure drops from laminar flows, unstable fluctuations of centreline velocity and the sudden abrupt increase in

root-mean-square value of the centreline velocity as indicators. Figure 3-5 shows the normalized pressure drop and deviation from laminar flow.

Figure 3-5. a) Normalized pressure drop against Re showing departure at $Re \approx 2000$. b) Magnified portion of insert (Sharp and Adrian 2004)

Li and Olsen (2006a) conducted experiments in square micro-channels with hydraulic diameters of $200 \mu\text{m}$ to $640 \mu\text{m}$ and aspect ratios close to 1, and stated that the fully turbulent flow of deionized water occurred at Reynolds number ranging from 2600 to 2900. Deviation from a parabolic laminar profile and an increase in velocity

Figure 3-6. Longitudinal velocity correlation function in a micro-channel having a hydraulic diameter of $640 \mu\text{m}$ (Li and Olsen 2006a)

fluctuations were taken as an indication of transition to turbulence. Furthermore, they interpreted fully developed turbulence at the point whereby fluctuations no longer increased with an increase in Reynolds number. In addition, they noticed a decrease in the normalized spatial correlations of the velocity fluctuations as a result of a fully turbulent flow (see Figure 3-6).

Shortly, [Li and Olsen \(2006b\)](#) conducted the same experiments in square and planar micro-channels with hydraulic diameters of 311 μm to 640 μm at Reynolds numbers based on their previous study which resulted in fully turbulent flows ($2362 < \text{Re} < 3489$). Results of the linear spatial correlations indicated decay to close to zero values for these turbulent flows, which were more drastic near the wall locations, indicating large-scale turbulent structures were smaller near the side walls than the centre of the micro-channels. They also noted the presence of eddies when Galilean decomposition was performed on the instantaneous velocity fields (see Figure 3-7). Based on their

Figure 3-7. Presence of turbulent structures in a turbulent flow for a square channel with a hydraulic diameter of 640 μm ([Li and Olsen 2006b](#))

analysis, they stated that the turbulent length scales calculated in their micro-channels are similar to previous reported works on macro-channel duct flows.

Rands et al. (2006) conducted flow measurements using circular tubes with diameter of 16.6 to 32.2 μm , noting the transition to turbulence at Reynolds number range of 2100 – 2500, and it is higher for smaller diameters. This was based on the deviation of the friction factor-Reynolds number product from the constant of 64 for laminar flow, and the deviation of the viscous heating parameter with increased Reynolds number from a constant of 32. Figure 3-8 shows the friction factor-Reynolds number product with increasing Re.

Figure 3-8. Deviation of friction factor-Reynolds number product at onset of turbulence (Rands et al. 2006)

Natrajan and Christensen (2007) conducted PIV experiments in a 536 μm capillary tube using water as the working fluid. They interpreted transition to turbulence by the deviation of the parabolic laminar flow profile at $\text{Re}=1900$, as shown in Figure 3-9a. This was also characterized by a steady increase in pressure difference from laminar flow (linear trend) and $f\text{Re}$ (constant = 64 for laminar flow) with a significant departure at $\text{Re} = 2200$. In addition, the Reynolds shear stress increased and the maxima shifted

Figure 3-9. Profiles of a) Cross-section flow profile b) Reynolds stress as a function of wall position from transition of laminar to turbulent flow (Natrajan and Christensen 2007)

towards the wall with increasing Re, being connected by a linear region to the centreline (see Figure 3-9b). This is a trait of macro-scale turbulent pipe flows. Fully turbulent state occurred at Reynolds number of 3400, accompanied with a decrease in the growth rate of pressure drop with Re, and more intense single point velocity fluctuations. Galilean decomposition of instantaneous velocity field through the

Figure 3-10. Galilean decomposition at Re = 2800, with constant advection velocity $0.79 U_{cl}$ removed. U_{cl} = centreline velocity (Natrajan and Christensen 2007)

removal of the constant advection velocity also revealed the presence of hairpin-like vortices, a characteristic of wall bounded turbulence in macro-scale. This is depicted in Figure 3-10.

Vetel et al. (2008) conducted PIV in a smooth 2:1:2 axisymmetric contraction-expansion with a constriction diameter of 10.25 mm. They noted high-frequency spatial fluctuations in the velocity field at a Reynolds number of 440 which may indicate the transition to turbulence. However, the flow was periodic and non-stationary. At 112.75 mm downstream of the contraction, hairpin-like vortices (see Figure 3-11) were present, with the top part being similar to horseshoe vortices, which are commonly found in laminar and turbulent boundary layer flows. The transition to turbulence was also characterized by high frequency fluctuations and an increase in spatial perturbation. They stated the occurrence of turbulence at Reynolds number 1044, with high frequency fluctuations but reduced large amplitude fluctuations (i.e. higher kurtosis), yet with stationary velocity measurements, in agreement with others (Ahmed &

Figure 3-11. Perspective view of one extracted hairpin-like vortex 112.5 mm downstream of constriction at $Re = 460$ (Vetel et al. 2008)

Giddens 1983). The instantaneous flow profile was also erratic and the vortex shedding phenomenon was observed.

To identify coherent structures within a cylindrical cavity with a rotating lid (radius = 144 mm), Meyer et al. (2008) applied the snapshot POD technique. The snapshots were obtained using PIV to measure the flow in the range of $Re = 2800 - 6300$, with a glycerine-water mixture as the working fluid. The method involves subtracting the mean velocity field from each of the snapshots to obtain the fluctuating velocity components, i.e. Reynolds decomposition, and subsequently assembling them into a matrix. By solving for the eigenvalues of the matrix, the contribution of each POD mode (i.e. eigenvector) to the total turbulent kinetic energy (i.e. fluctuations) could be evaluated (Fukunaga 1990). Instabilities in the flow developed at $Re > 2800$. At $Re = 3075$, the first two dominant modes were found to contain most ($\approx 2/3$) of the turbulent kinetic energy in the flow. Figures 3-12 a), b) and c) show the snapshots of the flow, the distribution of the turbulent kinetic energy based on the eigenvalues associated with each POD mode, and POD mode 1 respectively (page 74).

Tamano et al. (2009) investigated the statistics of the turbulent boundary layer with drag-reducing aqueous surfactant solutions (65, 100, 150 ppm CTAC) flowing past a test plate in a water tunnel ($0.1 \times 0.3 \text{ m}^2 \times 1.5 \text{ m}$, $Re \approx 30,000$). They employed LDA and PIV to evaluate the velocity profiles in the momentum layer. Figures 3-13a) and b) show the stream-wise turbulent intensities and Reynolds shear stresses at the boundary layer respectively (page 75), for both water and CTAC solution. Note that Re_θ refers to the momentum thickness Reynolds number. For the CTAC solution, there was a slight

a)

b)

c)

Figure 3-12. a) Flow snapshot b) Distribution of kinetic energy across mode numbers c) POD mode 1 showing 4 pairs of counter rotating vortices. [Meyer et al. \(2008\)](#)

Figure 3-13. Comparison of statistics between PIV and LDA at 63% drag reduction. a) stream-wise turbulent intensity b) Reynolds shear stresses (Tamano et al. 2009)

reduction in turbulent intensity with the formation of two peaks at the boundary layer, while the Reynolds shear stresses were greatly diminished. They hypothesised the two visible peaks to be a bi-layered structure, which inhibited the formation of hairpin vortices.

3.6.3 Brief review on 2-D turbulent flows

Hitherto, there are limited studies on 2-D turbulence in bounded channel flows. Most reported works and turbulent flows have been 3-D in nature, but interest in 2-D turbulent flows is gaining attention rapidly in recent years with the two inertial ranges discovered in 1967 (Kraichnan 1967). Hence, three reported works have been included in this section for completeness.

Amarouchene and Kellay (2002) studied the effect of long chain polymer molecules (PEO, 4×10^6 and 8×10^6 g/mol) on the effect of 2-D turbulence in soap films (1% commercial detergent), as fast flowing soap films in a channel (length = 2m, width = 6cm) passed a grid array of cylinders ($\phi = 1 - 3$ mm). The power spectrum obtained

Figure 3-14. Power spectra for soap films with different PEO concentrations showing power law scaling at different distances from the grid of cylinders a) Longitudinal and transverse velocity power spectra at a flux of 0.4 ml/s, 7 cm from grid. b) Thickness power spectra at a flux of 0.43ml/s, 7cm and 15 cm from grid (Amarouchene and Kellay 2002)

using LDA did not show a $-5/3$ scaling, but a -3.3 power law decay was present. They observed that the small additions of polymers resulted in a reduction in turbulent intensity, as well as a reduction in fluctuation amplitudes at low frequencies (see Figure 3-14a). This reduction increased with increasing polymer concentration. However, the

high frequency portion is hardly affected. They reasoned that the addition of polymers may have suppressed or reduced vortex mergers which are responsible for the transfer of energy upscale. In addition, the presence of polymer also suppressed the third moment, a measure of the energy flux. Power spectrum of the thickness fluctuations (passive scalar) of the soap film indicated a $-5/3$ scaling in the absence of polymers, and approximately -1 with the addition of polymers (see Figure 3-14b), which is consistent with theoretical expectations for passive scalars in the viscous – convective subrange of an enstrophy cascade ([Lesieur 2008](#)).

[Boffetta \(2007\)](#) conducted numerical simulations for the 2-D Navier-Stokes equation at spatial resolutions of up to $16,384^2$ (grid size further increased to 32768^2 in [Boffetta and Musacchio 2010](#)), and verified the existence of the two extended inertial ranges in agreement with the double cascade predicted ([Kraichnan 1967](#)). With increased spatial resolution, most of the energy injected was transferred to large scales which were removed by friction, and most of the enstrophy followed the direct cascade to small scales to be dissipated by viscosity. Varying the resolution of the simulation resulted in a change in energy or enstrophy transferred. It does not affect the range of the inverse cascade but strongly affects that of the enstrophy cascade. Figure 3-15 shows the observed energy spectrum which follows a power law decay of $-5/3$ in the inverse cascade, and approximately -3.6 in the direct cascade. The snapshots of energy and enstrophy fluxes showed that they appeared in different physical regions without any correlation. This suggests that it is possible to generate only one cascade in the absence

of the other, evident in other experimental and numerical works ([Amarouchene and Kellay 2002](#), [Berti et al. 2008](#)).

Figure 3-15. Energy spectrum from numerical simulations depicting the two inertial ranges for different spatial resolutions (A = 2048, B = 4096, C = 8192, D = 16384) ([Boffetta 2007](#))

[Berti et al. \(2008\)](#) carried out numerical simulations based on the Oldroyd-B model, using a periodic Kolmogorov flow to study the elastic turbulent regime in two dimensions. In the elastic turbulent regime, the power injection increased with a power law scaling as it transits to the turbulent like regime at $Wi = 15$. Although the maximum Re was only 0.7, there was increased resistance in their flow, similar to high- Re turbulence. In addition, the Reynolds stresses were smaller than 10^{-2} , indicating that the elastic stresses were causing the turbulent flow i.e. elastic-driven, and the inertial terms were irrelevant. Figure 3-16 shows the power spectrum of velocity fluctuations which follows a power-law decay with an exponent of -3.8. In addition, the Lagrangian Lyapunov exponent increases with Wi at low Re , which is different from that observed in high Re flows where the injection of polymers reduces chaoticity by lowering the

Lyapunov exponent (Boffetta et al. 2003). They also stated that Wi plays a similar role in elastic turbulence as non-linearity (Re) in ordinary hydrodynamic turbulence, where the distribution of Lyapunov fluctuations become more asymmetric with increasing Re (Bec et al. 2006).

Figure 3-16. Velocity power spectra at $Wi = 26$ (squares) and $Wi = 45$ (circles) following a power law decay with an exponent of -3.8 (Berti et al. 2008)

3.7 Summary

In this chapter, a brief review has been conducted on the quantification methodologies of inertial-driven turbulent flows, e.g. turbulent intensity, Reynolds stresses, velocity power spectrum, eddy visualization techniques etc., which has been extensively studied. However, most of these quantification parameters have not been included in the reported experimental works of viscoelastic turbulence which have been discussed in Chapter 2. To correlate and obtain better understanding of the differences and

similarities between elastic-driven and inertial-driven turbulence, it is necessary to compute the quantification parameters presented here for comparison.

It has been shown that viscoelastic turbulent flows exhibit similar power law scaling in the velocity power spectra as that of high-Re (2-D/3-D) turbulence in the inertia subrange. One similarity between viscoelastic (i.e. elastic-driven) turbulence and 2-D high-Re (i.e. inertial-driven) turbulence is that the forcing function (which in the former refers to the release of elastic stresses by the strongly extended molecules), cascades energy to large scales (Fouxon and Lebedev 2003). The presence of the $-5/3$ and -3 slopes in the velocity power spectra have both been reported, but none has reported the occurrence of the two slopes simultaneously. It could be possible that like 2-D inertial-driven turbulence, it is possible to generate one cascade in the absence of the other in viscoelastic turbulent flows. Of course, one could argue that the evolution of 2-D or 3-D elastic-driven turbulence in micro-channels could also depend on the aspect ratios of the channels, but evidence has yet to be provided.

In summary, this chapter has presented the common terminologies for characterising inertial-driven turbulent flows. These will be employed in the present study for the characterisation of viscoelastic (elastic-driven) turbulence.

4. Rheology

4.1 Introduction

The manifestation of viscoelastic turbulence (which defies the conventional high-Re criteria) clearly shows that viscoelastic liquids behave very differently from Newtonian liquids. Hence, it is important to understand the characteristics of viscoelastic liquids, and their responses when subjected to strains and stresses. These will be reviewed in Section 4.2, together with some classic viscoelastic examples. The strain/stress responses of viscoelastic liquids show that they exhibit both solid- and liquid-like behaviors, depending on the time scale of the flow. One of the simplest models that describe this behavior is the Maxwell model, which will be presented in Section 4.3. Some other relevant parameters that define the properties of a viscoelastic liquid will be presented in Section 4.4.

The Deborah number (De) is the critical parameter that governs the stability of a viscoelastic micro-channel flow at diminishing Re . Upon a critical De , instabilities ensue and the flow field becomes turbulent, i.e. viscoelastic turbulence. To quantify De and Re for each experimental flow rate (Chapters 6 and 7), the bulk properties of the viscoelastic liquids have to be measured. Hence, shear and extensional tests have been conducted on the test liquids; the measured results, i.e. viscosity and extensional relaxation time, have been included in Section 4.5. In addition, the obtained extensional relaxation time is the relevant characteristic time (of the bulk viscoelastic liquid) in this

study, and could be important in subsequent statistical analyses of the viscoelastic turbulent flow field.

4.2 Viscoelasticity

4.2.1 Background

Viscoelastic liquids consist of macro-molecules, i.e. polymers, typically suspended in a solvent. Unlike Newtonian liquids, the behavior of viscoelastic liquids is non-Newtonian in nature and their properties (e.g. viscosity) vary with the characteristic time scale of the flow field, e.g. shear rate. Due to elasticity imparted by the polymer molecules, a viscoelastic liquid can exhibit both solid-like behavior at short time scales and liquid-like behavior at long time scales. In this study, the relevant time scales are on the order of milliseconds.

Figure 4-1. An illustration of viscoelastic behavior of 'silly putty'

An example of a viscoelastic liquid is 'Silly Putty' ([Oswald 2009](#)) (see Figure 4-1). If a ball of 'Silly Putty' is dropped on a hard surface, it bounces like an elastic solid. However after a few hours, it ends up spreading very slowly (i.e. irreversible

deformation) like a liquid under the force of its own weight. The behavior of ‘Silly Putty’ is a combination of both an elastic solid and a viscous liquid. For a perfectly elastic solid, the mechanical properties can be described by Hooke's law (Oswald 2009). This is a constitutive equation which is not a fundamental law of nature, but a reasonable approximation that is applicable for some materials while it fails in others. The Hooke's law states that,

$$\sigma = G\gamma, \quad (4.1)$$

where σ is the applied stress, G the elastic modulus, and γ represents the strain. This law assumes that for a perfectly elastic solid, the material *responds instantaneously* to any applied stress and it has *infinite memory*, i.e. releasing the applied stress results in the material recovering all deformity.

Similarly for liquids, Newton proposed (Malkin and Isayev 2006) that in parallel and uniform flow, the shear stress is proportional to the velocity gradient. This can be described by the constitutive equation,

$$\tau = \mu\dot{\gamma}, \quad (4.2)$$

where τ is the applied shear stress, μ the dynamic shear viscosity, and $\dot{\gamma}$ the shear rate. For Newtonian liquids, μ is a constant for any given temperature and is independent of shear stress or shear rate. Similarly to a perfect elastic solid, a Newtonian fluid *responds instantaneously* to shear. However, it has *zero memory*, i.e. releasing the applied stress results in the material retaining all deformity. Liquids which do not obey Newton's law of viscosity are termed as non-Newtonian liquids. For these liquids, μ is

not a constant, but is dependent on the shear rate and shear stress applied. Typically, this law fails for liquids such as polymer melts and liquids which contain high molecular weight macro-molecules (polymers) exceeding the critical concentration where Newton's law is valid.

4.2.2 Stress response to step strain

Figure 4-2 shows the stress response of different materials to a sudden step strain at $t = 0$. For the ideal viscous fluid (Newtonian fluid), it experiences a sudden stress impulse which drops to zero immediately. On the other hand, the ideal elastic solid experiences a constant stress instantaneously. However, for the viscoelastic liquid, the stress decays according to a rate that is depending on the relaxation time of the material.

Figure 4-2. Response of various materials to applied strain a) Applied constant strain b) Ideal viscous fluid response c) Ideal elastic solid response d) Viscoelastic liquid response

4.2.3 Stress response to shear rate

Figure 4-3 shows the stress response of the different materials to varying shear rates. As shown, a Newtonian fluid (i.e. an ideal viscous liquid) exhibits a linear stress relationship with shear rate. However, this is not so for viscoelastic liquids,

demonstrated by the non-linear relationship with shear rate. The two most common types of non-Newtonian liquids are either shear thickening or shear thinning fluids. As depicted in Figure 4-3, the rate of increase in stress decreases with increasing shear rate for shear thinning liquids, i.e. a drop in viscosity. The reverse is true for shear thickening liquids, i.e. increasing viscosity with increasing shear rate. Most polymeric solutions are shear thinning in nature, and this is also the characteristic of the test liquids used in the current investigations.

Figure 4-3. Stress response of various liquids as a function of shear rate

4.2.4 Viscoelastic examples

A classical demonstration of viscoelasticity is the Weissenberg or rod-climbing effect ([Barnes et al. 1989](#)) of a viscoelastic liquid, shown in Figure 4-4a. The figure depicts a spinning rod in a beaker of polymeric solution. Polymeric chains are stretched and aligned in the flow direction, resulting in a normal force towards the rod and hence causing the polymer solution to ‘climb up’ the rod. However, for a Newtonian liquid, inertia pushes the liquid towards the wall, resulting in the liquid ‘climbing up’ the beaker walls instead. Another viscoelastic phenomenon is the die swell effect (see

Figure 4-4b), whereby a polymeric fluid swells upon exiting a tube (Barnes et al. 1989). This is due to the relaxation of elastic stresses that build up within the fluid under pressure. An increase in flow rate would amplify the swelling effect.

Figure 4-4. a) Weissenberg effect shown by a solution of polyisobutylene in polybutene. b) Die swell effect. (Barnes et al. 1989)

4.3 Maxwell model

4.3.1 Introduction

The Maxwell model is the simplest viscoelastic liquid model (Oswald 2009), which can be represented by a series connection consisting of a spring, i.e. the elastic component, and a dashpot, i.e. the viscous component (see Figure 4-5). When a stress is applied, the spring reacts almost instantaneously, while the dashpot extends at a rate which is dependent on its characteristic time, before reaching the final strain.

Figure 4-5. Schematic diagram of the Maxwell Model

4.3.2 Response to constant strain

Figure 4-6. Response of the Maxwell fluid to step strain. a) step strain b) stress response

When a sudden constant strain is applied to the Maxwell fluid (see Figure 4-6), there is a step increase in stress experienced by the fluid, due to the elastic component ([Barnes et al. 1989](#)). In the linear region, this stress is given by

$$\sigma_E = G_E \gamma_E, \quad (4.3)$$

where G_E is the shear modulus of the spring. However, the viscous component deforms and increases in strain over time, resulting in a decrease in stress on the elastic component. This phenomenon is also known as creep.

$$\sigma_D = \mu_D \dot{\gamma}_D, \quad (4.4)$$

However, the total stress

$$\sigma = \sigma_E = \sigma_D, \quad (4.5)$$

and the total strain

$$\gamma = \gamma_E + \gamma_D, \quad (4.6)$$

Thus, the stress decreases at an exponential rate given by

$$\sigma = G_E \gamma \exp\left(-\frac{t}{\tau}\right), \quad (4.7)$$

where τ is the characteristic time of the liquid, whereby the stress reduces by 63.2% of its initial value i.e. at $t = 0$. In the linear region, this characteristic time τ can be taken as the time within which a viscoelastic material preserves its shape memory. Beyond this time, the material loses this memory and takes on a new shape.

4.3.3 Response to constant strain rate

For a constant applied shear rate, the Maxwell fluid responds elastically for small strains and in short times (Oswald 2009). However for long times, viscous behavior sets in (see Figure 4-7).

$$\text{For short times,} \quad \sigma = G\dot{\gamma}t \quad (\text{material responds elastically}) \quad (4.8)$$

$$\text{For long times,} \quad \sigma = \mu\dot{\gamma} \quad (\text{viscous behavior sets in}) \quad (4.9)$$

Note: the stress plot is valid only in the linear regime

Figure 4-7. Response of the Maxwell fluid to constant strain rate a) step strain rate b) stress response consisting of elastic and viscous phase

Therefore, the characteristic time of viscoelastic relaxation is given by (see Figure 4-7b)

$$\tau = \mu/G \quad (4.10)$$

Similarly from equations 4.8 and 4.9 we obtain,

$$\sigma(t) = \mu\dot{\gamma} \left[1 - \exp\left(-\frac{t}{\tau}\right) \right], \quad (4.11)$$

when the system is sheared at a constant shear rate at $t=0$.

4.3.4 Other viscoelastic liquid models

Figure 4-8. Schematic diagram of the a) Burgers model b) Jeffrey model

Other similar macroscopic models include the Burger's model and the Jeffrey's model, which are merely variations of the Maxwell model (see Figure 4-8). Extensions of the Jeffrey model and Maxwell model include the convected Jeffrey (Oldroyd) and convected Maxwell models respectively ([Oswald 2009](#)). Although the equations are non-linear, these models are only valid in the linear regime, i.e. shear rates less than $1/\tau$. Beyond the linear regime, these quasi-linear models are unable to predict shear thinning behavior and predicted viscosity results diverge towards infinity for high elongation rates.

Non-linear models for the prediction of behavior in the non-linear regime include the Oldroyd 8-constant model, Giesekus, FENE (Finitely Extensible Non-linear Elastic spring) and the Phan-Thien and Tanner (PTT) model ([Oswald 2009](#)). These models are basically extensions of the Maxwell model. All the aforementioned models assume

incompressible flow, where volume is conserved. These models have their limitations and it is beyond the scope of this study to examine them in detail.

4.4 Viscoelastic fluid properties

4.4.1 Intrinsic viscosity

The intrinsic viscosity of a polymer solution, $[\eta]$, is a measure of the capacity of a polymer molecule to enhance viscosity, depending on the shape and molecular weight of the molecule. It is defined to be the limiting viscosity number as

$$[\eta] = \lim_{c \rightarrow 0} \left(\frac{\eta - \eta_s}{\eta_s c} \right) \quad (4.12)$$

where η_s is the solvent viscosity, η is the solution viscosity, and c is the solute concentration (g/ml) (Oswald 2009). For linear chain polymer molecules, the intrinsic viscosity is related to the molecular weight, M_w (g/mol), by the Mark-Houwink-Sakurada equation, given by

$$[\eta] = K M_w^a \quad (4.13)$$

where constants K and a may be determined from the Polymer Handbook by Brandrup and Immergut (Brandrup et al. 1975). Tirtaatmadja et al. (2006) subsequently carried out linear regression analysis on the intrinsic viscosities of PEO solutions to obtain the composite Mark-Houwink-Sarkurada equation, given by

$$[\eta] = 0.072 M_w^{0.65} \quad (4.14)$$

where $[\eta]$ is the intrinsic viscosity (cm^3/g), and M_w is the PEO molecular weight (g/mol).

4.4.2 Critical overlap concentration

The critical overlap concentration c^* is the concentration at which polymer coils touch and entangle with each other. For solutions consisting of flexible polymer chains (Flory 1953),

$$c^* = 1/[\eta]. \quad (4.15)$$

Beyond the critical limit ($c > c^*$), the solution is a semi-dilute solution and the molecular interactions are strong due to multiple entanglements. Below this limit ($c < c^*$), the molecules behave as independent coils with no entanglements. The polymer solution is thus considered dilute (for $c < c^*$), and its viscoelastic properties are governed by the behaviour of a single polymer molecule.

4.5 Rheology of test liquids

4.5.1 Polyethylene oxide (PEO)

Polyethylene oxide (PEO) is a water soluble, aerobically biodegradable synthetic polyether produced by the polymerization of ethylene oxides. It has a long and highly flexible chain backbone, and can be represented by the linear formulae $(-\text{CH}_2\text{CH}_2\text{O}-)_n$. PEO is available commercially in a wide range of molecular weights ranging from 100,000 g/mol to more than 8,000,000 g/mol . Those with molecular weights less than 100,000 g/mol are usually termed as Polyethylene glycol (PEG). PEO is usually in a

polydispersed form, i.e. comprising of a distribution of molecular weights, and its size distribution can be computed statistically via mass spectrometry or size exclusion chromatography.

In this study, PEO was used to study the highly elastic behaviour of semi-dilute polymeric solutions in a micro-channel. It has been shown that aqueous PEO solutions are affected by pH, and susceptible to thermal-oxidative attack ([Morlat and Gardette 2001](#), [Hassouna et al. 2007](#)), due to the formation of radicals which result in chain scissions mechanism. This results in a reduction in molecular weight, accompanied by a decrease in both viscosity and elastic modulus, which is accentuated with increasing molecular weight ([Dontula et al. 1998](#)). Thus in this study, the sample fluids were stored away from direct heat and light, and disposed of after 5 days.

4.5.2 Test liquids preparation

The water soluble PEO molecules used in this study have an average molecular weight range of $0.6 - 8 \times 10^6$ g/mol (POLYOX, The Dow Chemical Company). The composition of the test liquids was kept similar to that of [Gan et al. \(2007\)](#), comprising of 55.55g of glycerol (Sigma Aldrich) and 46.45g of DI water, while the polymer concentrations and molecular weight was varied. Table 4-1 shows the notation of the various liquids used in this study and the corresponding concentration and molecular weight. In addition, the critical overlap concentration c^* (computed based on equations 4.14 and 4.15) is also presented. The polymer was first dissolved in DI water (46.45g) and stirred at 250 rpm for 2.5 hours to ensure complete solubility. Next, glycerol

(55.55g) was added and the mixture was stirred at 400 rpm for 2.5 hours to ensure homogeneity. To carry out PIV, 0.03g of red fluorescent particles (3 μm , Duke Scientific Co.) were subsequently added to the mixture, and gently stirred at 100 rpm for 30 minutes to obtain the final viscoelastic test liquids.

Table 4-1. Sample fluids composition

Viscoelastic liquids	PEO (g)	Molecular weight (g/mol)	Critical overlap concentration c^* (g)
2%-0.6M	2	600,000	0.24
0.1%-2M	0.1	2,000,000	0.11
0.3%-2M	0.3		
1.0%-2M	1		
1.1%-2M	1.1		
0.1%-8M	0.1	8,000,000	0.05
0.3%-8M	0.3		
0.5%-8M	0.5		

4.5.3 Steady shear rate tests

4.5.3.1 1-D shear flow

To measure the shear properties e.g. viscosity of the test liquids, steady shear rate tests were conducted using the Gemini Hr Nano (plate-plate rheometer). The tests were all carried out at 25°C, which was controlled by a water bath. All tests were conducted with a stainless steel parallel plate (PP 55), and the gap was set to be 180 μm . The rheometer was operated in strain-controlled mode. To illustrate how the viscosity was measured, here we consider the simplified 1-D shear flow of a liquid between a moving plane and a stationary plane (see Figure 4-9a) ([Barnes et al. 1989](#)).

Figure 4-9. a) schematic of 1-D shear flow. b) infinitesimal fluid element representation

Figure 4-9b) shows a schematic representation of an infinitesimal element in the flow, where velocities in the y and z directions are zero, represented as

$$v_x = \dot{\gamma}y, \quad (4.16)$$

$$v_y = v_z = 0 \quad (4.17)$$

For a Newtonian fluid, the stress distribution for this flow can be written as,

$$\tau_{yx} = \mu\dot{\gamma}, \quad (4.18)$$

$$\tau_{xz} = \tau_{yz} = 0, \quad (4.19)$$

$$\sigma_{xx} - \sigma_{yy} = 0, \quad \sigma_{yy} - \sigma_{zz} = 0 \quad (4.20, 4.21)$$

For non-Newtonian liquids, the stress distribution is represented as

$$\tau_{yx} = \eta(\dot{\gamma})\dot{\gamma}, \quad (4.22)$$

$$\tau_{xz} = \tau_{yz} = 0, \quad (4.23)$$

$$N_1(\dot{\gamma}) = \sigma_{xx} - \sigma_{yy}, \quad N_2(\dot{\gamma}) = \sigma_{yy} - \sigma_{zz} \quad (4.24, 4.25)$$

where $\eta(\dot{\gamma})$ is the shear rate dependent dynamic viscosity, $N_1(\dot{\gamma})$ and $N_2(\dot{\gamma})$ are the first and second normal stress differences respectively. $N_1(\dot{\gamma})$ and $N_2(\dot{\gamma})$ are typically non-zero, and are the main causes of the Weissenberg and die swell effects (see Figure 4-4).

Hence, by controlling the strain rate applied on the test liquids i.e. strain-controlled tests, the stress required to maintain the speed of the plate is measured, and hence the viscosity of the test liquids could be calculated using equation 4.22. However, the Gemini Hr Nano is not equipped with a sensitive normal force sensor, and is hence unable to measure N_1 accurately (for dilute viscoelastic liquids).

4.5.3.2 *Shear viscosity measurements*

The steady shear rate ($\dot{\gamma}$) test was carried out in the range $1 \leq \dot{\gamma} \leq 10,000 \text{ s}^{-1}$. As the sample used in this study are shear thinning in nature, the viscosity of the samples decrease with increasing shear rate. Figure 4-10 shows the viscosity-shear rate profile (log-log plot) for various test samples obtained with the Gemini Hr Nano. From the figure, it can be seen that 0.1%-2M behaves like a Newtonian liquid, as its viscosity does not change significantly with increasing shear rate ($\approx 0.011 \text{ Pa}\cdot\text{s}$). Note that the amount of PEO added to 0.1%-2M was below the critical overlap concentration. For the other test liquids where the critical overlap concentration was exceeded, shear thinning behavior is observed to be significant. For example, the measured viscosity of 0.5%-8M dropped by a factor of about 40 times, from $0.87 \text{ Pa}\cdot\text{s}$ ($\dot{\gamma} \approx 1 \text{ s}^{-1}$) to $0.02 \text{ Pa}\cdot\text{s}$

($\dot{\gamma} \approx 10,000 \text{ s}^{-1}$). The shear viscosity range of the test liquids have been included in Table 4-3 (Section 4.6).

Figure 4-10. Viscosity plot of test liquids under steady shear rate.

4.5.4 Extensional tests

4.5.4.1 Extensional flow

Extensional flows are common in many industrial processes, e.g. fiber spinning, inkjet printing etc., but the extensional properties of the liquids are often poorly characterized. Extensional effects become important when there is stream-line acceleration or squeezing mechanism in the flow. In this study, the Capillary Breakup Extensional Rheometer (CaBER) was used to study the extensional properties of the test liquids. Figure 4-11 shows an element in an extensional flow in the y-direction.

Figure 4-11. Schematic diagram of element undergoing simple extensional flow

Obeying the law of mass conservation, the element must contract in the x- and z-directions at a rate of $\dot{\epsilon}/2$ (Barnes et al. 1989). The velocity field of the element can be written as

$$v_y = \dot{\epsilon}y, \quad v_x = -\frac{\dot{\epsilon}}{2}x, \quad v_z = -\frac{\dot{\epsilon}}{2}z \quad (4.26, 4.27, 4.28)$$

where $\dot{\epsilon}$ is the extensional strain rate. This results in extension of the liquid element in the y direction, but compression in the x and z directions, i.e. thinning.

4.5.4.2 Extensional relaxation time measurements

Figure 4-12 shows the schematic setup of the CaBER at rest and during extension. The test liquids were first dispensed into the gap between two parallel plates ($D_o = 6mm$), which was set at 2 mm (see Figure 4-12a). Next, the test liquids were subject to extensional flow by elevating the top plate abruptly within a short time interval of between 30 – 180 ms (i.e. stream-line acceleration) to the preset height of $h = 10.43$ mm (see Figure 4-12b). This results in thinning of the liquid filament (as explained in

Section 4.5.4.1) and its diameter, $D_{mid}(t)$, was measured using a laser micrometer (resolution = 10 μm).

Figure 4-12. Schematic of the sample during extensional tests in CaBER a) at rest b) during extension

Figure 4-13 shows high speed images of the extension and breakup of a viscoelastic fluid filament (Rodd et al. 2005b). After the top plate has been elevated abruptly, the breakup of the viscoelastic liquid initiates with a viscous dominated regime in the first phase. This is followed by a second phase which is governed by elasticity and surface tension, for which fluid viscosity (i.e. viscous effects) does not play a role (Bousfield et al. 1986, Bazilevskii et al. 1990, Renardy 1994, Entov and Hinch, 1997).

Figure 4-13. Filament thinning of 0.3% PEO sample during extension. $t_{event} = 100$ ms (Rodd et al. 2005b)

In the second phase, the extended filament can be described by an exponentially decreasing function, based on the Upper-Convected Maxwell model (Entov and Hinch 1997, Clasen et al. 2004)

$$D_{mid}(t) = D_o \left(\frac{GD_o}{\sigma_s} \right)^{\frac{1}{3}} e^{-\frac{t}{3\lambda_e}} \quad (4.29)$$

where D_{mid} and D_o are the mid-diameter and initial diameter of the filament respectively, G is the elastic modulus of the filament, σ_s is the surface tension, λ_e is the extensional relaxation time of the solution, and t is the elapsed time. Thus, as can be seen from equation 4.29, the diameter decreases exponentially with a decay rate of $(3\lambda_e)^{-1}$.

Figure 4-14 shows the filament diameter of the different sample fluids used in this study measured by the CaBER. Note that these measurements were taken by the laser micrometer after the top plate had reached the preset height (second phase), and exponential fitting of the filament diameter was carried out to obtain the extensional relaxation time (λ_e) of the test liquids. As shown in the figure, the filament of 0.1%-2M decays the fastest while 0.5%-8M decays the slowest. This means that 0.1%-2M had the shortest extensional relaxation time, while that of 0.5%-8M being the longest. All results obtained have been included in Table 4-3 in Section 4.6.

Figure 4-14. Plot of filament diameter over time as measured by CaBER for the test liquids.

4.6 Summary

To understand how viscoelastic liquids result in the rich flow field dynamics as reviewed in Chapter 2, it is important to distinguish between a Newtonian liquid and a viscoelastic liquid. Table 4-2 highlights some differences that have been discussed in this Chapter.

Table 4-2. Differences between Newtonian and Viscoelastic liquids

Newtonian liquid	Viscoelastic liquid
$\gamma = \gamma_0, \tau(t > 0) = 0$	$\gamma = \gamma_0, \tau(t > 0) \neq 0$
$\tau = \mu \dot{\gamma}$	$\tau = \eta(\dot{\gamma}) \dot{\gamma},$
$N_1(\dot{\gamma}), N_2(\dot{\gamma}) = 0$	$N_1(\dot{\gamma}), N_2(\dot{\gamma}) \neq 0$
Low λ_e	High λ_e

In addition, it is necessary to characterize the properties of the test liquids to obtain the relevant dimensionless parameters e.g. Re, De etc. in a viscoelastic flow. The measured properties of the PEO test liquids are summarized in Table 4-3.

Table 4-3. Rheological properties of test liquids

Viscoelastic liquids	η (mPa·s)		λ_e (ms)
	Low shear rate (1 s ⁻¹)	High shear rate (10,000 s ⁻¹)	
2%-0.6M	474.2	68.0	45
0.1%-2M	13.1	9.5	26
0.3%-2M	65.6	16.4	47
1.0%-2M	553.3	36.8	81
1.1%-2M	643.9	35.7	105
0.1%-8M	22.1	9.9	102
0.3%-8M	238.0	15.7	157
0.5%-8M	865.2	21.0	239

Generally, an increase in polymer concentration and molecular weight augments the extent of shear thinning, and results in an increased extensional relaxation time. Although the viscosity varies significantly with shear rate, the extensional relaxation time (λ_e) was found to be independent of the extension rate (i.e. time for abrupt elevation). This is because CaBER measures the longest relaxation time of the largest molecule in the viscoelastic liquid during filament thinning, which occurs after the abrupt elevation was completed. Note that dynamic oscillation tests, which can be used to evaluate the Maxwell (i.e. shear) relaxation time, has been excluded in this section because it has been shown that the stability of the flow field in a contraction-expansion micro-channel is dominated by the extensional properties of the viscoelastic liquid (Tai et al. 2011). In this case, the flow of a viscoelastic liquid past the contraction is analogous to it being stretched abruptly by CaBER.

5. Experimental Setup

5.1 Introduction

In this study, a micro-channel design with similar dimensions to that of [Gan et al. \(2007\)](#) was used to examine the characteristics of viscoelastic turbulence in a 3-stream contraction-expansion flow. Due to the micro-scale dimensions, it was necessary to employ cleanroom micro-fabrication techniques to fabricate the micro-channel, with silicon as the substrate. A summary of the fabrication procedures and schematic diagram of the micro-channel are included in Section 5.2, together with the definition of dimensionless parameters (based on micro-channel dimensions) relevant to this study.

One of the objectives of this investigation was to apply statistical analyses and visualization tools on the viscoelastic turbulent flow field. For this purpose, the velocity was quantified using Particle Image Velocimetry (PIV). In Section 5.3, the PIV experimental setup for the investigation is presented, together with a brief description of some concepts and steps involved in the PIV analyses. Due to the short molecular time scales relevant to the investigations, a high speed camera was employed to obtain high frequency velocity measurements. By acquiring data at the appropriate time scales, it may thus be possible to extract information from the statistical analyses pertaining to the molecular and bulk liquid properties, an objective of this study. As image grabbing was performed under continuous white light illumination, this resulted in higher noise levels and poorer image contrasts, as compared to short pulsed illumination in

conventional PIV setups. Hence, it was crucial to apply image enhancement techniques to improve image quality, to increase the location accuracy of the correlation peak obtained during PIV evaluation. Section 5.4 provides an introduction of some simple image enhancement techniques employed, which greatly improved image clarity.

5.2 Micro-channel chip design

5.2.1 Schematic diagram

Figure 5-1. Schematic 2-D diagram of the micro-channel. Arrows depict the direction of flow, blue (side-stream), red (main-stream)

Figure 5-1 shows a schematic 2-D diagram of the micro-channel that was used in the experiments of the current investigation. This micro-channel consisted of two layers, a bottom silicon layer and a top Pyrex glass layer. The micro-channel was first fabricated by etching a bottom silicon layer using Deep Reactive Ion Etching (DRIE). To etch the micro-channel, a thick layer of photoresist ($\approx 10 \mu\text{m}$, photoresist AZ 9260) was used to

mask other regions. Subsequently, the closed channel was completed by bonding it to a top Pyrex glass layer via anodic bonding. The transparency of the glass layer facilitated observations of the flow field in the micro-channel.

The micro-channel consists of 3 inlets and 1 outlet (see Figure 5-1). The test liquids were pumped into the micro-channel by high force precision pumps (KDS-410) via glass syringes (Hamilton Gastight). The main-stream liquid (red) employed was highly viscoelastic, as compared to the side-stream liquid (blue) which has lower elasticity. The main-stream flow and each of the two side-stream flows contributed to 50% and 25% of the total flow rate (Q) respectively. As the liquids flowed towards the outlet, they were forced through a sudden contraction, which is 1 mm in length and has a width of 125 μm (8:1:8). This resulted in stream-line acceleration and imposed a squeezing mechanism on the liquids concurrently, causing the flow field to be extensional in nature (see Section 4.5.4.1). The micro-channel has a depth of 180 μm , and PIV measurements were taken at the mid-plane i.e. 90 μm .

5.2.2 Calculation of dimensionless constants

Based on the measured properties of the test liquids in Chapter 4, the relevant dimensionless numbers in the micro-channel can be computed. The dimensionless numbers were defined similarly here to [Gan et. al \(2007\)](#). The Reynolds number (Re) was defined as

$$\text{Re} = \frac{\rho \bar{U} D_h}{\mu} = \frac{2\rho Q}{\eta(w_c + d)} \quad (5.1)$$

where D_h is the hydraulic diameter ($D_h = \frac{2w_c d}{w_c + d}$), ρ is the liquid density (kg/m^3), Q is the flow rate (m^3/s), η is the viscosity in the contraction ($\text{Pa}\cdot\text{s}$), w_c is the average contraction width occupied (m), and d is the channel depth (m). Due to the abrupt change in geometry, the Deborah number, De , was defined as

$$De = \lambda \dot{\gamma}_c = \frac{2\lambda_e Q}{w_c d^2} \quad (5.2)$$

where λ_e is the extensional relaxation time (ms) measured with CaBER (Chapter 4), with other variables having the same definition as equation 5.1. Note that $\dot{\gamma}_c$, Re and De were calculated in the contraction region based on the main-stream liquid only. To approximate the space occupied by the main-stream liquid in the contraction, numerical simulations in a straight channel flow (neglecting elasticity) were conducted using COMSOL Multiphysics (see Appendix A), which showed that the main-stream liquid occupied approximately 35 - 42% of the cross-sectional area in the contraction within the operating range investigated.

5.3 Particle Image Velocimetry (PIV)

5.3.1 Introduction

To examine and quantify the flow field dynamics, Particle Image Velocimetry (PIV) was employed. PIV is non-intrusive and is capable of achieving higher resolutions with the appropriate objective lenses, as compared to Laser Doppler Anemometry (LDA) and Hot Wire Anemometry (HWA). To carry out PIV, it was necessary to first seed the

viscoelastic liquid with polystyrene particles (0.1% w/w, 3 μm , Thermo Scientific). This seeding concentration ensured each interrogation window consisted of approximately 10 or more particles at any instant, for obtaining accurate results with a high signal to noise ratio. Subsequently, images of the flow field were captured at the appropriate frame rates. To compute the velocity, the captured images were processed with a cross-correlation PIV MATLAB algorithm. After obtaining the velocity measurements, other relevant quantities such as Reynolds stresses, turbulence intensities etc. can then be computed (see Chapters 6 and 7). The snapshot Proper Orthogonal Decomposition (POD) technique ([Sirovich 1987](#)) can also be applied to visualize the presence of any intricate 2-D structures e.g. turbulent eddies, in the flow field.

5.3.2 Imaging setup

To visualize and quantify the flow field of a viscoelastic turbulent flow, it is necessary to acquire instantaneous snapshots at high speeds. Hence, a monochrome high speed camera (Photron Fastcam SA5, 32 Gb, sensor pixel size = $20 \times 20 \mu\text{m}^2$) was used to capture 8-bit sequential images of the flow. The SA5 is capable of high recording speeds of up to 1,000,000 frames per second (FPS) and high shutter speeds of down to 1 μs . A fibre optic illuminator was used to provide white light illumination. The exposure time for each captured image was reduced by increasing the camera shutter speed. Generally, large exposure times resulted in blurring of the particles, while short exposure times resulted in images that were too dim. In the experiments, a shutter time of 40 μs was found to be appropriate up to the highest flow rate investigated, i.e.

approximately ten times the maximum capturing speed. In addition, the surface of the silicon micro-channel acts as a mirror, which greatly enhances the amount of reflected light received by the camera. Subsequently, the images were recorded and stored in Portable Network Graphics (PNG) format, which is a bitmapped image format employing lossless data compression. Figure 5-2 shows a schematic diagram of the optical setup. The micro-channel was magnified with a 10X telescopic lens (Optem Zoom 70 XL, 1X TV tube, 2X auxiliary lens configuration) which has a depth of field of 30 μm .

Figure 5-2. Schematic diagram of the experimental setup.

5.3.3 Recording modes

For each flow rate investigated, two recording modes were used to capture the images, namely mode 1 (low frequency) and mode 2 (high frequency). In mode 1, the high speed camera was triggered by a function generator (Agilent) and images were captured in 50 Hz bursts over a period of 200 seconds (see Figure 5-3a). Mode 1 was

used to obtain the average velocity profiles over long time durations. For each burst, the high speed camera (SA5) captured 1 image pair, i.e. 2 images separated by a time interval (Δt) between them, which varied for each flow rate. In mode 2, the images were captured in a continuous recording mode, with each image separated from the next by the same time interval as in mode 1 i.e. Δt (see Figure 5-3b). Mode 2 enabled the analyses of velocimetry data over shorter time scales, and progressive changes in velocity can be observed, as compared to the appearance of ‘spikes’ in low frequency velocity measurements. However, due to a limitation in memory storage of the camera (32 Gb), higher flow rates required smaller time intervals (Δt), which resulted in shorter recording durations.

Figure 5-3. Schematic diagram of the two recording modes. a) mode 1. b) mode 2.

5.3.4 PIV algorithm

For velocity computations, two types of correlation algorithms can be applied to each interrogation window, i.e. auto-correlation and cross-correlation. Auto-correlation was used in the past for the processing of PIV images when cameras could not capture at high frame rates. The images captured by these images contain two exposures on a single frame. As such, although the particles may be tracked, it is not discernible which particles belong to the first and second frames. This results in the loss of the directional data, which could be particularly important for flows having complex flow structures. However, since the SA5 is capable of high speed operation, each captured frame only comprise of one exposure; thus, cross-correlation was applied instead. Directional information was hence preserved with this technique.

The captured images from modes 1 and 2 were processed with a MATLAB algorithm which was adapted from [Taylor et al. \(2010\)](#). The algorithm divided each image (1.4 mm by 1 mm) into multiple interrogation windows of sizes 64 x 32 pixels, with a 50% overlap between windows (i.e. the separation between interrogation windows is 32 pixels axially and 16 pixels transversely). Based on the Nyquist theorem, the average particle displacement should be within half the size of an interrogation window to prevent aliasing, which would cause erroneous results. In the current investigations, the displacement limit was set at $\frac{1}{4}$ of each interrogation area, also known as the one-quarter rule. Hence, with the appropriate recording time interval between images frames, cross-correlation was applied on each interrogation window by employing the Fast Fourier Transform (FFT), which is based on the Wiener-Kinchin theorem ([Raffel](#)

et al. 2007). Once the correlation matrix was obtained, the peak was located by employing a three-point Gaussian peak fit, due to the Gaussian nature of the particle image intensities (Raffel et al. 2007). The peak location, which represents the pixel displacement of the interrogation window, could be obtained with sub-pixel accuracy based on this method. The average velocity for each interrogation window could then be computed via dividing the displacement by the recording time interval between the images.

5.4 Image enhancement techniques

5.4.1 Introduction

In the current experimental setup, particle images were captured under white light illumination. This is unlike conventional PIV systems which use high intensity laser pulses to excite fluorescent particles that emit light, and background noise is eliminated (see Figure 5-4).

Figure 5-4. Conventional PIV image pair at two instances, with fluorescent particles being illuminated by a double pulsed laser.

As such, background noises e.g. flickering light, fabrication imperfections, and even stains and dust etc. become significant and are clearly visible in the experimental

recordings. These background noises are impossible to eliminate totally but can be reduced significantly via image pre-processing to improve signal to noise ratio for accurate velocity computations.

5.4.2 Image enhancement algorithm

Figure 5-5a shows the original PIV image captured before image pre-processing, for water flowing through a straight micro-channel at a flow rate of $Q = 20$ ml/h. The micro-channel has a width and depth of $1000\text{ }\mu\text{m}$ and $180\text{ }\mu\text{m}$ respectively.

Figure 5-5 a) Original image b) Background image c) Processed image d) inverted image of c)

It can be observed that the contrast between the background and particles is low, and this can be enhanced by background subtraction from the image. The average background image was obtained by averaging an ensemble of images captured during the experiments i.e. > 100 images, via an in-house written MATLAB algorithm. Figure 5-5b shows the background image of Figure 5-5a, after processing 300 images of the flow using the algorithm. Note that although the images used for background calculation consisted of particles, they became unobservable as a result of the averaging procedure.

After the background image was obtained, it was subtracted from all captured images. However, this did not remove noise that varied with time e.g. light flickers. To remove low-frequency background variations in the subtracted images, an adaptive filter in MATLAB (`wiener2.m`) was applied ([Gonzalez et al. 2004](#)). This filter performs smoothing in regions of small variances, but preserves high-frequency variations i.e. particle edges. In addition, an image sharpening filter in MATLAB (`deconvblind.m`) was applied to restore the sharpness of particle edges that could deteriorate due to motion. Finally, the contrast of the resulting images was uniformly enhanced using an adaptive algorithm in MATLAB (`adapthisteq.m`). Figure 5-5c shows the resulting processed image after the series of steps listed above. For ease of visualization, Figure 5-5d shows the intensity inverted image of Figure 5-5c.

5.5 Summary

In this chapter, a schematic diagram of the micro-channel and a summary of the fabrication procedure has been presented, together with the definitions of Re and De.

Micro-fabrication of silicon chips typically results in superior surface finishes and better dimension tolerances. In this investigation, the smooth mirror-like surface of the micro-channel greatly enhanced the amount of light received by the camera sensor.

A brief introduction to the PIV technique and imaging setup were presented. As white light illumination was used, the appropriate exposure time was employed to reduce blurring. In addition, image enhancement was necessary to improve the signal strength of the cross-correlation results. Here, image pre-processing was carried out to remove random background noise for improving signal-to-noise ratio. These techniques had successfully improved the clarity and contrasts of images (see Figure 5-5), which increases the computational accuracy of the correlation peak location, to resolutions of down to sub-pixel levels ([Raffel et al. 2007](#)). Although not elaborated, one important point to note is that white light illumination limits the technique to the usage of fluids which have the same refractive indices. Thus, the main-stream and side-stream liquid have to be of the same solvent, and other phases e.g. oil cannot be used.

6. Results - Flow Field Analyses

6.1 Introduction

6.1.1 Scope

One of the main objectives of this investigation was to rigorously investigate the downstream turbulent flow field of the contraction-expansion micro-channel (see Chapter 2). It will be of interest to establish linkages between the turbulence behaviours of a viscoelastic fluid to the minute amount of dissolved polymer molecules. Although [Gan et al. \(2007\)](#) and [Lam et al. \(2009\)](#) had reported some velocimetry results, little statistical methods were used for analysis. Thus, this chapter attempts to uncover the underlying flow fields by statistical evaluation and flow visualization downstream of the contraction, in particular the effect of the relaxation of polymer molecules that had been stretched as they flow past the contraction. Hence, the focus here has been placed on the downstream flow field; the analysis of upstream flow field has not been included due to the difficulty in capturing images of the extremely fast moving main-stream liquid upstream, which was approximately ten times faster than the mean flow. In addition, the effects of varying molecular and bulk properties on the flow field will be investigated here.

6.1.2 Outline

The accuracy of the PIV algorithm will be validated in Section 6.2, while the dimensionless parameters and experimental details will be presented in Section 6.3. A high-speed camera (Photron Fastcam SA5) was used for image capturing and these

images were subsequently used for PIV analysis. In the second part of the chapter, an investigation of single and double point statistics will be included. Average cross-sectional profiles in two regions of the downstream flows will also be presented in Section 6.4. Temporal velocity fluctuations with large standard deviation are a signature of turbulent flows and fluctuations at 3 different points downstream of the contraction have been included in Section 6.5. The temporal autocorrelation function and spatial correlation function will be presented in Section 6.6. In addition, the power spectrum density of the velocity fluctuations will be evaluated in Section 6.7. Since the presence of eddies are a characteristic of many turbulent flows, Galilean decomposition and proper orthogonal decomposition (POD) will be employed in Section 6.8, to uncover coherent structures that may have been hidden within the large convection velocities captured by the imaging system. The effect of molecular time scales on the energy cascade will also be discussed in Section 6.9, via the variation of test liquids with different concentrations and molecular weights.

6.2 Flow profile in a straight planar micro-channel

To validate the PIV algorithm, the velocity of water flow at $Q = 20$ ml/h ($Re = 9.66$) in a straight planar micro-channel (see Fig. 5-5a) was computed. The micro-channel has a depth of $180\text{ }\mu\text{m}$ and a width of 1 mm . The PIV algorithm was set to process 2000 image pairs captured at 10 Hz over a recording period of 200s i.e. using mode 1, whereby the time interval between the images within each pair was 0.33 ms . Figure 6-1 shows the fully developed axial velocity in the micro-channel mid-plane i.e. $90\text{ }\mu\text{m}$,

which was taken 5 mm away from the inlet. The interrogation window size used was 64 x 32 pixels, with a 50% overlap between consecutive windows (refer to Section 5.3.4).

Figure 6-1. Axial velocity of water across the planar micro-channel

For comparison, an analytical solution for a fully developed Newtonian flow in a rectangular channel (White 2006) is used. The axial velocity profile is described in equation 6.1:

$$V_z(x, y) = \frac{12Q}{ab\pi^3} \frac{\sum_{n=1,3,5,\dots}^{\infty} \frac{(-1)^{(n-1)/2}}{n^3} \cos(n\pi x/2a) \left(1 - \frac{\cosh(n\pi y/2a)}{\cosh(n\pi b/2a)}\right)}{1 - \frac{192a}{\pi^5 b} \sum_{n=1,3,5,\dots}^{\infty} \frac{\tanh(n\pi b/2a)}{n^5}} \quad (6.1)$$

where $V_z(x, y)$ is the axial velocity, a is half channel width and b is half channel depth. The analytical solution was found to be in good quantitative agreement with the experimental data (see Figure 6-1), with a difference in mean velocity of less than 2%. In addition, the uncertainty of the PIV technique was determined to be approximately 4%.

6.3 Experimental details

In Sections 6.4 to 6.8, results of the 3-stream flow presented will be based on 1.0%-2M as the main-stream liquid, and 0.1%-2M as the side-stream liquid. For each experimental flow rate, the main-stream flow and each of the two side-stream flows contributed to 50% and 25% of the total flow rate respectively. The compositions and rheological properties of the liquids have been presented in Chapter 4. The corresponding average bulk velocity, shear rate, viscosity, Re and De of the main-stream liquid in the contraction have been tabulated in Table 6-1. Two recording modes were used (see Chapter 5), and the time interval (Δt) between images have also been included in Table 6-1. In the following sections, the Re and De numbers at the various flow rates will no longer be included and readers are to refer to Table 6-1 for reference when necessary.

Table 6-1. Experimental flow parameters and corresponding properties of 1.0%-2M in the contraction.

Flow rate, Q (ml/h)	Average bulk velocity (mm/s)	Average shear rate (1/s)	Corresponding Viscosity (Pa·s)	Re	De	Δt (s)
2	24.7	364	0.140	0.019	27	0.004
6	74.1	1164	0.095	0.086	82	0.0013
14	172.8	2883	0.071	0.270	193	0.0004
20	246.9	4262	0.052	0.528	278	0.00025

6.4 Mean velocity profiles

6.4.1 Region of interest

Due to the large amount of data, two cross-sections have been selected for presenting the average axial velocity profile in this section. These two cross-sections are located at $\delta = 0.3$ mm and $\delta = 1$ mm downstream of the contraction respectively (see Figure 6-2 for definition of δ). 10,000 pairs of images were captured based on mode 1 over a period of 200 s, and the statistics i.e. mean velocity, standard deviation, were computed. The chosen interrogation window size of 64×32 pixels did not result in significant data loss and smooth cross-sectional velocity profiles could be obtained without interpolation. However, low lighting conditions and the significantly higher velocity near the walls resulted in a high noise level at the near wall region. Hence, velocimetry data within $100 \mu\text{m}$ from the channel walls will not be presented (see Figure 6-2). In addition, velocimetry data from $\delta = 0 - 0.1$ mm has also been omitted in subsequent sections, because of blurring effects as a result of high velocities at the contraction exit.

Figure 6-2. Schematic diagram of the contraction-expansion micro-channel, showing the plot of a mean axial velocity profile at a distance δ downstream of the contraction. Data in the shaded region has been removed due to high noise level.

6.4.2 Cross-section profile at $\delta = 0.3$ mm

Figure 6-3a shows the normalized mean velocity profiles for different flow rates at $\delta = 0.3$ mm. The profiles were obtained by normalizing the velocity at each interrogation window with the mean channel velocity for that particular flow rate, i.e. $\bar{U}_i / \bar{U}_{bulk}$.

Figure 6-3. a) Normalized axial velocity and b) Turbulent intensities at $\delta = 0.3$ mm

From Figure 6-3a, it can be observed that the velocity profiles resemble inverted Gaussian profiles for flow rates of $Q = 2$ ml/h to $Q = 14$ ml/h. The main-stream fluid exhibits die-swell behaviour upon exiting the contraction. Thus, it pushes the side-stream fluids towards the channel wall to occupy more space for itself; the main-stream fluid velocity is thus relatively lower. With a constrained space of flow the side-stream fluid has to travel at a higher velocity to obey the law of mass conservation.

As the flow rate was increased, the central portion of the velocity profile increased and a distinct peak can be observed at $Q = 20$ ml/h (see Figure 6-3a), which is absent at lower flow rates. This is because the extensional stresses, which have been stored in the

polymer molecules of the main-stream fluid as it passes through the contraction, have not been totally released. Chapter 2 has stated that a polymer exhibits more ‘solid-like’ behaviour at short times (i.e. higher shear rates), but ‘liquid-like’ behaviour at long times (i.e. lower shear rates). Hence at $Q = 20$ ml/h, this phenomenon manifests itself as a central peak. As a result, the relative differences in velocity at the centre and the sides are much lesser than that at lower flow rates. Note that the velocity profile at $Q = 20$ ml/h is not symmetrical, and the central peak is slightly off-centered because of small misalignment errors (not perfectly 90°) between the micro-channel and the camera.

Figure 6-3b shows the turbulent intensity across the flow field at $\delta = 0.3$ mm. For each flow rate, the data was obtained by normalizing the standard deviation of the velocity measurements at each interrogation window with the mean axial velocity at the same interrogation window, i.e. $u_{i,rms}/\bar{U}_i$. The normalized profiles can be observed to approach a symmetrical profile about the centreline with a large ensemble size. Note that for all De, turbulent intensity is largest near the channel walls at the interface between the main-stream and side-stream fluids. This is because the velocity is largest near the channel walls, and the main-stream fluid has the most room to move freely at the interface, to release extensional stresses in the polymer molecules. In addition, turbulent fluctuation at the highest De (i.e. $Q = 20$ ml/h) remains low at the centre near the contraction exit. This is because the main-stream liquid is still solid-like upon exiting the contraction, and hence there is negligible relaxation of the polymer molecules, resulting in low turbulent intensity.

6.4.3 Cross-section axial velocity profile at $\delta = 1.0$ mm

Figure 6-4a shows the normalized mean velocity profile for different flow rates at $\delta = 1.0$ mm. Normalization similar to the velocity profile at $\delta = 0.3$ mm was performed. From Figure 6-4a, it can be seen that the velocity profiles at all flow rates approach inverted Gaussian profiles without a peak in the centre-line. This means that the main-stream fluid had sufficiently relaxed here as compared to that at $\delta = 0.3$ mm. This stress relief mechanism of the main-stream fluid was reported to be the basis of chaotic mixing downstream (Gan et al. 2007, Lam et al. 2009). In addition, the relative difference between the centreline velocity and near wall velocity reduces with increasing flow rate. Besides the die swell effect, this could also be due to penetration of the side-stream fluids into the main-stream with increasing flow rate, which resulted in the chaotic mixing observed by Gan et al. (2007) and Lam et al. (2009).

Figure 6-4. a) Normalized axial velocity and b) Turbulent intensities at $\delta = 1.0$ mm

As compared to Figure 6-3a (i.e. $\delta = 0.3$ mm), the relative normalized difference between the centreline and side-stream is larger at $\delta = 1.0$ mm for all flow rates. This could be because the main-stream fluid experiences a ‘recoil’ mechanism as a result of stress relaxation further downstream at $\delta = 1.0$ mm. Hence, a retardation in axial velocity at the centreline, and a higher velocity near the channel walls is evident. This ‘recoil’ mechanism is due to the sudden release of stresses in the stretched polymer molecules to return to their low energy state (relaxed state) and has been attributed to be the cause of the random ‘sweeping’ action, leading to chaotic mixing, at the contraction exit. This random ‘sweeping’, together with the penetration of side-stream fluids into the main-stream fluids, is not homogenous.

Figure 6-4b shows the turbulent intensity across the flow field at $\delta = 1.0$ mm. The normalization was done in a similar manner to that at $\delta = 0.3$ mm, using equation 3.24. For flow rates up to $Q = 14$ ml/h, the turbulent intensity at this location is quantitatively similar to that at $\delta = 0.3$ mm, and these fluctuations have not been reduced by viscous dissipation. This suggests that unstable interactions caused by the main-stream fluid due to the stress relief mechanism are not yet fully dissipated by viscosity. At $Q = 20$ ml/h, the turbulent intensity at the centre has increased as compared to that at $\delta = 0.3$ mm. This is due to the transformation of the main-stream liquid from a ‘solid-like’ state into a ‘liquid-like’ state, resulting in the release of stresses in the polymer molecules as it transforms, and hence causing an increase in localized fluctuations in this region.

Further downstream at $\delta = 2.0$ mm, the main-stream fluid appears to have almost completely relaxed into a liquid-like state, because the normalized velocity profile for all flow rates have almost converged into a single curve (see Figure 6-5a); the corresponding turbulent intensity curves are smooth across the channel width (Figure 6-5b). The turbulent intensities at $Q = 2$ ml/h and $Q = 6$ ml/h are low and velocity fluctuations have been attenuated significantly by viscosity (compare with Figure 6-4b). However, the velocity fluctuations at $Q = 14$ ml/h and $Q = 20$ ml/h have not been dampened out, and the turbulent intensities remain high (see Figure 6-5b). This shows that the effects of viscoelastic turbulence at high De can be felt even further downstream of the contraction.

Figure 6-5. a) Normalized axial velocity and b) Turbulent intensities at $\delta = 2.0$ mm

6.4.4 Reynolds stresses

Besides turbulent intensities, computation of Reynolds stresses ($\overline{u_i v_i} / \bar{U}_i^2$) enable the study of the turbulent energy transport and allows for comparison with high Re

turbulent flows. The Reynolds stresses can also be interpreted to be the momentum exchange between the mean flow and the turbulence. Figure 6-6 shows the normalized Reynolds stresses for the most turbulent state investigated, i.e. $Q = 20$ ml/h, at various distances (δ) from the contraction exit across the micro-channel width. The momentum flux can be observed to be most significant close to the contraction, and diminishes with axial distance (δ). Such high Reynolds stresses are typically observed in high Re flows in larger channels (Tamano et al. 2009, Japper-Jaafar et al. 2010). There is a change in sign convention for the Reynolds stresses across the microchannel width at each δ , because the turbulence generated at the centre was directed towards each wall in opposite directions.

Figure 6-6. Reynolds stresses at 20 ml/h across the channel at $\delta = 0.3$ mm, 0.5 mm, 1.0 mm and 2.0 mm respectively.

6.5 Single -point temporal velocity

6.5.1 Region of interest

Single-point temporal axial velocity fluctuations downstream of the contraction will be presented here. As there is a significant amount of data downstream, only the velocity measurements at 3 locations along the channel centre-line i.e. $\delta = 0.3$ mm, 0.5 mm, 1.0 mm, have been selected. Figure 6-7 shows the position of the three interrogation windows A, B and C (64×32 pixels, $128 \times 64 \mu\text{m}^2$) respectively, with the PIV measurements done at the channel mid-plane i.e. $90 \mu\text{m}$ depth. The temporal data was evaluated by processing the images using the MATLAB PIV algorithm.

Figure 6-7. Locations of interrogation windows A, B and C for single-point velocity measurements

6.5.2 Low frequency temporal axial velocity data

Figures 6-8a to 6-8c show the temporal velocity profiles at interrogation windows A, B and C over a period of 20 s. The temporal data was evaluated by processing 10,000 image pairs using the MATLAB algorithm. The images were captured in bursts at 50 Hz over a period of 200 s i.e. recording mode 1. For each burst, the high speed camera

(SA5) captures 1 image pair, i.e. 2 images, and the time interval (Δt) between the two images have been listed in Table 6-1, for the different flow rates. It is clearly observed in these figures that fluctuations in temporal axial velocity increases much more significantly at higher flow rates, i.e. higher Deborah number. These were already discussed in Section 6.4, where the turbulent intensities have been presented.

At flow rates of $Q = 2$ ml/h and $Q = 6$ ml/h, the temporal velocity at point A appeared laminar and steady, characterized by small-scale, low amplitude fluctuations. As flow rate was increased up to $Q = 14$ ml/h and $Q = 20$ ml/h, the amplitude of these fluctuations increase with increasing Deborah number, appearing as chaotic and random in time. The peak and trough ‘spikes’ observed could correspond to the large ‘sweeping’ mechanism, which is a result of the release of extensional stresses in the polymer molecules. In addition, a sudden jump in velocity profile, appearing as a step up in mean velocity by approximately 3-4 times in magnitude, can be observed when the flow rate was increased from $Q = 6$ ml/h to $Q = 14$ ml/h (≈ 2 times magnitude). This is an indication that at $Q = 14$ ml/h, the mainstream liquid having transformed from a liquid-like state (i.e. viscous state) to a solid-like state (i.e. elastic state) in the contraction, had not completely relaxed back to its viscous state. Furthermore, the velocity is observed to decrease for all flow rates as the mainstream liquid travels downstream from $\delta = 0.3$ mm to $\delta = 1.0$ mm along the channel mid-line (see Figure 6-8 a-c). This is due to the die-swell effect and the elastic spring-back mechanism, resulting in velocity retardation which is especially pronounced at $Q = 20$ ml/h, whereby the velocity has reduced to a quantitative value similar to that at $Q = 14$ ml/h.

Figure 6-8. Low frequency axial velocity measurements at a) interrogation window A ($\delta = 0.3$ mm) b) interrogation window B ($\delta = 0.5$ mm) and c) interrogation window C ($\delta = 1.0$ mm) obtained using recording mode 1.

6.5.3 High frequency temporal axial velocity data

Figures 6-9a to 6-9c show the temporal velocity profiles for all flow rates at the interrogation windows (A, B and C) over a period of 1 s. These images were captured in continuous bursts, whereby each image was separated from the next by a time interval (Δt) as listed in Table 6-1. This corresponded to an image capturing speed of 250 FPS, 750 FPS, 2500 FPS, and 4000 FPS for the corresponding flow rates of 2ml/h, 6ml/h, 14 ml/h, 20 ml/h respectively. Due to the limited storage memory (32 Gb) of the high speed camera, a set of 50,000 images were captured for each flow rate. Similar to Figures 6-8a to 6-8c, it can be observed that fluctuations in temporal axial velocity generally increase with increasing flow rate, i.e. higher Deborah number. However, instead of the ‘spikes’ that have been observed in the low frequency data in Figures 6-8a to 6-8c, i.e. conventional pulsed laser PIV setups, details on the velocity changes with time can now be more clearly discerned in Figures 6-9a to 6-9c. This is expected because high frequency data acquisition should uncover the transition between the ‘spikes’. The presence of high frequency noise can also be observed at $Q = 14$ ml/h and $Q = 20$ ml/h.

Using the velocity profile at $Q = 20$ ml/h as a benchmark for comparison, it can be observed that the velocity profiles at $\delta = 0.3$ mm and $\delta = 0.5$ mm are similar, i.e. the Taylor’s hypothesis seems to be valid over this short distance. However further downstream at $\delta = 1.0$ mm, the velocity profile appears to deviate slightly. This could be due to further release of stresses into the flow field by the mainstream liquid at $\delta = 1.0$ mm, which was earlier noted to be in a solid-like state at $\delta = 0.3$ mm in Fig. 6-3a.

Figure 6-9. High frequency axial velocity measurements at a) interrogation window A ($\delta = 0.3$ mm) b) interrogation window B ($\delta = 0.5$ mm) and c) interrogation window C ($\delta = 1.0$ mm) obtained using recording mode 2.

6.6 Correlation data

6.6.1 Single-point autocorrelation

In this section, the 1-point temporal autocorrelation function obtained at interrogation window A (as shown in Fig. 6-7) will be presented. This function is based on the most turbulent flow state presented in Section 6.5 i.e. $Q = 20$ ml/h, and describes how the velocity data at that particular location is correlated over time. Figure 6-10 shows the autocorrelation function obtained from the high frequency velocity data in Section 6.5.3 (see Figure 6-9a, $Q = 20$ ml/h). The data selected had a mean and standard deviation which are similar to that obtained from the low frequency recording in Section 6.5.2 (i.e. $< 3\%$ discrepancy). As shown in Figure 6-10, the autocorrelation function rapidly drops from unity and fluctuates about 0. Within a lag of less than 0.05 seconds, the correlation value had dropped to 0 (see Insert), indicating the turbulent nature of the flow.

Figure 6-10. Temporal autocorrelation function obtained using setup 2. Inset shows the magnified view from 0 to 1 s.

6.6.2 Two-point (spatial) correlation

The spatial correlation function describes how far the velocity is correlated with increasing spatial lag i.e. whether the points belong to the same turbulent eddy. The spatial correlation function was evaluated at interrogation window A in the axial direction downstream using equation 3.32 in Section 3.5 (page 66). Due to limited resolution of the recording system, only spatial lags of up to 1.0 mm in the axial direction could be calculated. Based on Taylor's 'frozen turbulence' hypothesis, the single-point temporal autocorrelation function evaluated in Figure 6-10 should be equivalent to the spatial correlation function if the hypothesis is valid. To make a direct comparison, the time lag in Figure 6-10 has to be replaced by the equivalent spatial lag (mm), by multiplying the time lag (s) with the mean velocity (mm/s).

Figure 6-11. Comparison of spatial correlation function based on calculations and Taylor's hypothesis.

Figure 6-11 shows the calculated spatial correlation function evaluated from the PIV flow field, and the approximated spatial correlation function based on Taylor's

hypothesis (single point estimation). There appears to be a reasonably good agreement between Taylor's hypothesis and the calculated spatial correlation function. The slight discrepancy is possibly due to inhomogeneity of the turbulent field downstream as a result of the random polymer stress relaxation and viscous dissipation that may not be negligible. Nevertheless, correlation up to the last spatial lag in this channel showed good agreement and the Taylor's hypothesis can be assumed to be valid.

It is not possible to evaluate the length scales in the turbulent flow based on the calculated spatial correlation function because it did not decrease to zero within the allowable spatial limit. Using Taylor's hypothesis as an estimate, Figure 6-12 shows that flow was correlated up to a spatial lag of 1.44 mm where the function intersects the axis. This could mean the presence of correlated structures or 'eddies' which are of considerable size as compared to the micro-channel dimensions.

Figure 6-12. Approximation of spatial correlation function based on Taylor's hypothesis.

6.7 Spectral analysis

In Chapter 3, it was mentioned that 3-D turbulence at high Reynolds number generally obeys Richardson's energy cascade and Kolmogorov's hypothesis. The energy and power spectrum of its velocity fluctuations follow a power law decay with a slope of $-5/3$. However, this is not the case for 2-D turbulence where two power law decay regions are expected, i.e. $-5/3$ at larger scales and -3 at smaller scales. In this section, the power spectra of axial velocity fluctuations at different flow rates are investigated. To study the effect of the stress relaxation in the main-stream fluid on the downstream flow field, the power spectra was computed based on single-point statistics at interrogation windows A, B and C respectively.

Figures 6-13a to 6-13c show the power spectra of the axial velocity fluctuations, computed using FFT for various flow rates at interrogation windows A ($\delta = 0.3$ mm), B ($\delta = 0.5$ mm) and C ($\delta = 1.0$ mm) respectively. The power spectra were obtained without windowing and overlapping the segments. Data windowing using the Bartlett and Welch windows (Harris 1978) was not performed because the resulting variances were large. Based on the Nyquist criteria, the maximum frequency for each flow rate (i.e. 2 ml/h, 6 ml/h, 14 ml/h, 20 ml/h), is half of the image capturing frequency (i.e. corresponding to 125 Hz, 375 Hz, 1250 Hz, 2000 Hz respectively). As the recording duration was short, only data above 1 Hz has been included. However, it is possible to investigate the power spectrum at higher frequency ranges, which has not been reported in the literature.

Figure 6-13. Power spectrum of the axial velocity fluctuations at for various flow rates at a) interrogation window A ($\delta = 0.3$ mm), b) interrogation window B ($\delta = 0.5$ mm), and c) interrogation window C ($\delta = 1.0$ mm) respectively.

From Figure 6-13a to 6-13c, it can be observed that at $Q = 2$ ml/h, fluctuations are small and the power spectra exhibits a power law drop of less than one decade across the whole spectrum. As the flow rate was increased, relaxation of stresses in the main-stream liquid resulted in instability in the downstream flow, with an increase in power

of the velocity fluctuations at lower frequencies. In addition, the power spectra can be observed to drop with increasing axial distance (δ) from the contraction exit, for flow rates of $Q = 2$ ml/h to $Q = 14$ ml/h. However, this did not occur at $Q = 20$ ml/h and the fluctuations do not appear to decrease with δ .

At $Q = 20$ ml/h (see Figure 6-13a), three distinct features can be observed in the power spectrum i.e. a horizontal plateau followed by two power law decays. The horizontal plateau was observed in the power spectrum from 1 Hz to approximately 20-30 Hz before it decreases. This drop-off frequency is far higher than the reported works of [Groisman and Steinberg \(2001a\)](#) (≈ 0.1 Hz) and [Bonn et al. \(2011\)](#) (< 1 Hz) on elastic turbulence flows, and is in fact closer to the lowest frequency at which the main-stream fluid relaxes i.e. ≈ 11 Hz, which was computed as the inverse of the extensional relaxation time of the liquid. Note that this relaxation time measured is the longest relaxation time of the polymer molecules in the mainstream liquid. Subsequently, the power spectrum follows a power law decay with a slope which is close to the $-5/3$ value as predicted by Kolmogorov in high-Re turbulence. Based on Kolmogorov's hypothesis, this region is termed as the inertial sub-range, where energy is transferred from large to small eddies. However, it should be noted that throughout this experiment, inertia forces are negligible. A second drop-off frequency can be observed clearly i.e. at ≈ 200 Hz, whereby the spectrum decays with a second, steeper power law slope which is close to the characteristic -3 slope typically observed in 2-D turbulence. The second slope appears prominent closer to the contraction exit ($\delta = 0.3$ mm), and is subsequently attenuated further downstream. A second plateau can be observed for $Q =$

6 ml/h, 14 ml/h and 20 ml/h in all interrogation windows i.e. $\approx 400 - 500$ Hz for $Q = 20$ ml/h, corresponding to the small amount of noise present in PIV data in the measurements. A small peak could be observed for $Q = 2$ ml/h at ≈ 80 Hz in all interrogation windows. This peak (≈ 0.0002) has been accentuated by the log scale, and could be due to noise. Another postulation is that the peak could correspond to the high frequency response of the lower molecular weight polymer molecules, which are present because of polydispersity.

6.8 Flow field visualization

6.8.1 Vorticity and Galilean Decomposition

Figure 6-14 shows an instantaneous snapshot of the micro-channel downstream flow field ($\delta = 0.1 - 1.4$ mm) at $Q = 20$ ml/h, with the velocity vectors and vorticity included in the plot (anti-clockwise being positive). The vorticity was computed based on the least squares approach ([Raffel et al. 2007](#)), and the flow is from the right to left. Regions of strong vorticity are localized along the channel walls because the side-stream liquid flowed at a higher velocity compared to the main-stream liquid (refer to mean velocity profiles in Section 6.4), and due to interactions at the interface between the two test liquids. Note that the high vorticity does not imply the presence of a vortex, as evident from the velocity vectors in Figure 6-14. To visualize the presence of interesting flow phenomena that are a result of the elastic stress relaxation downstream, Galilean decomposition was applied ([Adrian et al. 2000](#)). This enables the visualization of non-stationary structures which have been convected away by the flow. Insets 6-14a

Figure 6-14. An instantaneous plot of velocity and vorticity (colour plot) downstream of the contraction, from $\delta = 0.1\text{mm}$ to $\delta = 1.4\text{mm}$. Insets a-c show structures at various time instances located in different regions, obtained using the Galilean decomposition method with a fraction of the convective velocity removed. a) $U - 0.6 \bar{U}$ b) $U - 0.6 \bar{U}$ d) $U - \bar{U}$

to 6-14c show the magnified views of the structures that were observed at different time instants, in various regions of the micro-channel when a fraction of the average convective velocity ($\bar{U}$) was removed. Note that the figures are not drawn to scale.

Insets 6-14a and 6-14b show the extracted views of two instantaneous snapshots located in the same region (enclosed within grey dotted lines, see Fig. 6-14) at $\delta \approx 0.2 - 0.9$ mm, when 60% of the average convective velocity was subtracted. Two distinct phenomena can be observed, namely the upward ‘sweeping’ motion (Inset a) and downward ‘sweeping’ motion (Inset b) of the main-stream liquid, as observed previously by [Gan et al. \(2007\)](#). At another time instant, a large eddy (≈ 1.3 mm x 0.6 mm, see Inset 6-14c) which spans across almost the entire micro-channel (enclosed within red dotted lines, see Fig. 6-14) could be observed when the average convective velocity was totally subtracted. This is in good agreement with the correlated length scale (i.e. 1.44 mm) in Section 6.6.2. However, the eddy could not be visualized or detected clearly with vorticity plots and plots of swirling strength ([Zhou et al. 1999](#)) unlike Galilean decomposition, because it is relatively weaker in comparison to the fast flowing convective flow field. Galilean decomposition also revealed smaller vortices at other time instants in various parts of the micro-channel. For conciseness, these have not been included in Figure 6-14.

6.8.2 Proper Orthogonal decomposition (POD)

Proper orthogonal decomposition (POD) is a commonly used tool to analyse dynamical flow structures in turbulent flows. It was first introduced by Lumley ([Lumley 1967](#)) to

Figure 6-15. Various POD modes at $Q = 20$ ml/h from $\delta = 0.1$ mm to $\delta = 1.4$ mm a) POD mode 1. A large vortex the scale of the micro-channel width can be observed. b) POD mode 3. Upwards 'sweeping' motion. c) POD mode 4. Downwards 'sweeping' motion.

break down complex flows into simple spatial modes, and is often used to identify coherent structures within turbulent flows. Here, the snapshot POD technique by Sirovich ([Sirovich 1987](#)) was applied on the velocimetry data obtained from instantaneous PIV measurements i.e. snapshots, from $\delta = 0.1\text{mm}$ to $\delta = 1.4\text{mm}$ (see Fig. 6-15). The advantage of this technique is the ability to retrieve the coherent structures containing the highest energy, which may be viewed as ‘dominant’ structures/modes in the flow field. Thereafter, snapshots of the actual flow can be reconstructed with great accuracy using only the most dominant modes.

In the current experiments, the POD technique was applied to uncover any coherent structures that may be present in the most turbulent flow state recorded i.e. $Q = 20\text{ ml/h}$. 1000 sets of snapshots captured at 50 Hz were used for the analyses. Figure 6-15 a-c shows the plots of modes 1, 3 and 4. Mode 2 is similar to mode 1 and is not shown. Mode 1 shows the presence of a large structure that spans across the whole micro-channel. Although it could not be visualized completely due to the limited spatial resolution of the camera, it is clear that this structure is a large eddy. This is in good agreement with the estimated correlation length (i.e. 1.44 mm) obtained in Section 6.6.2. In addition, modes 3 and 4 show the upwards and downwards ‘sweeping’ motion of the main-stream liquid respectively upon exit from the contraction, which has been observed previously in the Galilean decomposed flow field in Section 6.8.1 and by [Gan et al. \(2007\)](#).

Figure 6-16. Relative percentage turbulent kinetic energy of the first 13 modes

The first 13 modes were found to account for more than 50% of the turbulent energy. Figure 6-16 shows the distribution of turbulent kinetic energy among the first 13 modes. Mode 1 has the highest energy of approximately 20 %, followed by a significant energy drop for mode 2 i.e. approximately 8 %. It should be noted that the mode energy distribution appears to plateau after the first few modes. Hence, it appears that the viscoelastic turbulent flow observed at $Q = 20$ ml/h is highly complex, since many modes are required to describe the flow. In fact, analyses show that 64 modes are required to capture 75 % of the turbulent kinetic energy.

6.9 Polymer concentration and molecular weight

Viscoelastic turbulence has been generally accepted in the literature to be due to the release of stresses in the polymer molecules, which are undamped by viscosity in the absence of inertia forces. The release of energy by the polymer molecules into the flow

occurs over a range of time scales, which is dependent on the characteristic timescale of each individual polymer molecule, and that of the bulk liquid i.e. interactions with the solvent and other polymer molecules. Unlike inertial-driven turbulence where high energy eddies transfer energy to smaller ones, Figure 6-13 clearly shows that polymer molecules inversely cascade their energy to the larger eddies. Hence, it is clear that the molecular weight of the polymer molecule, and its concentration in the solvent are two factors that would have a direct impact on the ‘structure’ of the turbulent flow. The power spectrum, being a measure of turbulent fluctuations over timescales, would thus be a suitable tool to investigate the effects of polymer concentration and molecular weight on the inverse energy cascades.

The results presented in Sections 6.4 to 6.8 have been solely based on the investigations of a single viscoelastic main-stream liquid i.e. 1.0%-2M (1 wt% PEO, 2×10^6 g/mol). Figure 6-17 shows the log of the power spectrum of the axial velocity fluctuations for $Q = 20$ ml/h at interrogation window A ($\delta = 0.3$ mm). As mentioned in Section 6.7,

Figure 6-17. Log of the power spectrum of velocity fluctuations for 20 ml/h, at interrogation window A.

three distinct regions can be observed i.e. a horizontal plateau (region 1), followed by the first power law decay (region 2) from a drop-off frequency of $\approx 20 - 30$ Hz, and second power law decay (region 3) from a drop-off frequency of ≈ 200 Hz. Using a least squares fitting method, a line was fitted to each of the three regions, and the cut-off frequencies were obtained at the intersections between the fitted lines. This corresponded to drop-off frequency values of 32.58 Hz and 187.41 Hz respectively.

Taking note that these 3 distinct regions observed were most prominent at $Q = 20$ ml/h i.e. $De = 278$, at interrogation window A ($\delta = 0.3$ mm), the power spectra of several main-stream liquids, corresponding to various concentration levels and molecular weights, were subsequently examined. Table 6-2 shows the various main-stream liquids employed, their corresponding polymer concentrations, extensional relaxation times and the 2 drop-off frequencies. The power spectra of all test liquids have been included in Appendix B. All the 3-stream experiments were conducted in the same micro-channel at $De \approx 250$, and the side-stream liquid remained unchanged, i.e. 0.1%-2M.

As observed from Table 6-2, the 1st drop-off frequency is closely related to the molecular weight of the polymer used. This shows that the molecular weight of the polymer molecules determines the frequency and hence size (wave number = frequency/velocity) of the largest coherent structures receiving energy i.e. energy containing eddies, and the range of the inverse energy cascade i.e. an increase in the molecular weight of the molecule results in a larger range of the inverse energy cascade,

and causes the size of the ‘large eddies’ to increase as well. This is logical because polymers with high molecular weights typically have a larger molecular size distribution, resulting in wider molecular timescale ranges that could inject energy into the flow.

Table 6-2. Viscoelastic properties at various concentrations and molecular weights, with the corresponding cut-off frequencies

Molecular weight used (g/mol)	Polymer concentration (wt %)	Extensional relaxation time (ms)	1 st drop-off frequency (Hz)	2 nd drop-off frequency (Hz)	De
600,000	2	45.00	81.00	263.03	231
2,000,000	0.3	46.68	37.30	307.90	261
	1.0	81.13	32.58	187.41	278
	1.1	105.00	31.73	112.56	286
8,000,000	0.1	101.62	12.26	126.33	276
	0.3	156.92	16.66	73.00	283
	0.5	238.71	17.80	54.24	248

The 2nd drop-off frequency appears to be uncorrelated to the molecular weight. On close examination, it is found that this frequency corresponds more closely to the extensional relaxation time of the bulk liquid (refer to Figure 6-18). The 2nd drop-off frequency is typically the forcing frequency in 2-D turbulence, and could possibly be deduced as the effective energy injection frequency of the bulk liquid. A reasonable power law trend can be observed in Figure 6-18. Hence, the results in the current section verifies our hypothesis that the time scales of the viscoelastic liquid have significant effects on the ‘structure’ size and energy cascade in the power spectrum.

Figure 6-18. Log-log plot depicting the relationship between the 2nd drop-off frequency and bulk liquid behaviour.

6.10 Summary

In this chapter, PIV is an important tool for flow field analyses. Hence, the accuracy of the PIV algorithm was first validated using water ($\approx 4\%$ error) by computing the flow velocity in a straight channel, and compared to the analytical solution of [White \(2006\)](#). Subsequently, PIV was employed to analyze the 3-stream viscoelastic flow field downstream of the contraction-expansion micro-channel. The investigation was carried out with 1.0%-2M as the main-stream liquid, and 0.1%-2M as the side-stream liquids, for $2 \text{ ml/h} < Q < 20 \text{ ml/h}$ ($0.019 \leq \text{Re} \leq 0.528$ and $27 \leq \text{De} \leq 278$). Firstly, the normalized mean velocity and turbulent intensity profiles at $\delta = 0.3 \text{ mm}$ and $\delta = 1.0 \text{ mm}$ were computed respectively. At $\delta = 0.3 \text{ mm}$, a ‘solid-like’ behavior was observed at $Q = 20 \text{ ml/h}$, which was manifested as a central peak in the velocity profile. This peak had diminished at $\delta = 1.0 \text{ mm}$, and a ‘recoil’ mechanism was observed. This was due to the relaxation of stresses in the main-stream liquid, which resulted in increased

localized fluctuations, i.e. turbulent intensity. Generally, the turbulent intensities were found to increase with flow rates (i.e. De) at $\delta = 0.3$ mm and $\delta = 1.0$ mm. Further downstream at $\delta = 2.0$ mm, the turbulent intensities at 14 ml/h and 20 ml/h remained high. At $Q = 20$ ml/h, the Reynolds stresses at $\delta = 0.3$ mm were large, with values comparable to high-Re turbulent flows ([Tamano et al. 2009](#), [Japper-Jaafar et al. 2010](#)).

Single point velocity statistics at 3 points ($\delta = 0.3, 0.5, 1.0$ mm) were subsequently presented. Both low frequency (mode 1) and high frequency (mode 2) recordings show that velocity fluctuations increased drastically with increasing De . At $Q = 20$ ml/h, the single point autocorrelation function at $\delta = 0.3$ mm decayed rapidly to zero, indicating the turbulent nature of the flow. The Taylor's hypothesis was assumed valid, and the estimated spatial correlation function was found to decay to zero at a spatial lag of 1.44 mm. To visualize the flow field, Galilean decomposition and POD was employed. At $Q = 20$ ml/h, Galilean decomposition ([Adrian et al. 2000](#)) revealed the presence of a large eddy (≈ 1.4 mm $\times$ 0.7 mm), and the upward and downward 'sweeping' motion of the main-stream liquid previously observed by [Gan et al. \(2007\)](#). These 'structures' of the flow field were shown to be dominant modes when the snapshot POD technique ([Sirovich 1987](#)) was applied. In addition, the POD analyses showed that the flow field at $Q = 20$ ml/h was highly complex, requiring many modes to describe the turbulent flow.

The power spectra of the velocity fluctuations were then evaluated, for $1 \text{ Hz} < \text{frequency} < 1000 \text{ Hz}$. Generally, the flow field loses its stability with increasing De ,

and the power of velocity fluctuations increased. At $Q = 20$ ml/h, two power law decays were observed in the power spectra, with corresponding slopes of $-5/3$ and -3 respectively. The presence of the two slopes is a characteristic of 2-D high-Re turbulent flows (Kraichnan 1967, Leith 1968, Batchelor 1969), whereby the 2nd drop-off frequency corresponds to the forcing frequency (i.e. energy injection point). This phenomenon could be due to the upward and downward ‘sweeping’ motion of the main-stream liquid, which was observed in the Galilean decomposition and POD analyses. Subsequently, the power spectra of six other viscoelastic main-stream liquids consisting of PEO of varying molecular weights and concentrations were compared at $De \approx 250$. The results showed that the energy injection point (i.e. 2nd drop-off frequency) correlated closely to the bulk extensional relaxation properties of the main-stream liquid, which could be the effective energy injection frequency of the liquid. In addition, the molecular weight of the polymer molecule used was found to correlate to the 1st drop-off frequency; the molecular weight of the polymer determines the size of the largest coherent structures and range of the energy cascade. Therefore, these results verify the hypothesis that the time scales of the viscoelastic liquid have significant effects on the ‘structure’ size and energy cascade in the power spectrum.

7. Results – Molecular Visualization

7.1 Introduction

7.1.1 Review

In Chapter 6, the characteristics of viscoelastic turbulence have been examined by analyzing the flow field via PIV. Despite the low Reynolds number (i.e. $Re < 1$), the addition of a small amount of polymer molecules to a viscous liquid i.e. glycerol-water mixture, resulted in a drastically different and turbulent flow behavior i.e. large turbulent intensities, power law decay in power spectrum etc. To explain the phenomenon and flow field characteristics, the stretching and relaxation of polymer molecules have been identified as the root cause of the observed turbulence, with inference from the PIV results. Although the molecular weight of the polymer molecules and the bulk fluid properties of the viscoelastic liquid clearly had a distinct effect on the flow field characteristics i.e. as seen in the power spectrum (refer to Section 6.9), the results do not reveal the underlying mechanics of viscoelastic turbulence.

Hitherto, the deduction of molecular behavior via motion analyses of tracer additives has been adopted as one of the main approach in the literature, because the polymer molecules are invisible. Although fluorescent-tagged DNA molecules are commercially available e.g. T4-DNA labeled with POPO-3, their usage has been limited to tracer additives/probes ([Ingremeau and Kellay 2013](#)) because they are costly and impractical for dynamic flow field investigations. Flow field measurement techniques e.g. Particle

Image Velocimetry (PIV), Laser Doppler Anemometry (LDA), Hot Wire Anemometry (HWA) etc., are often used to deduce the behavior of polymer molecules and bulk solution properties via shear stress calculations, velocity profile measurements, eddy visualization techniques etc. Despite the large amount of work done based on this indirect approach, understanding of the underlying mechanisms in viscoelastic turbulence is limited, and several outstanding questions on viscoelastic flows remain unresolved e.g. the high Weissenberg number problem ([Keunings 1986](#)), the ongoing debate between the viscous theory ([Lumley 1969](#), [Procaccia et al. 2008](#), [Benzi 2010](#), [Thais et al. 2013](#)) and elastic theory ([Tabor and de Gennes 1986](#), [Min et al. 2003](#), [Dallas et al. 2010](#)) on drag reduction ([Toms 1948](#)) etc.

“How do the polymer molecules change the flow field so drastically when they are only present in minute amounts?” This fundamental question has yet to be answered. Direct observation of this minute amount of polymer molecules in viscoelastic flows is clearly the key for a breakthrough. It will be fundamental, yet interesting, to directly observe how the extension, relaxation and interactions between polymer molecules could produce such rich dynamics to result in the phenomena as aforementioned. This knowledge can be extended to elucidate the underlying mechanisms of viscoelastic fluid flow phenomena in biology e.g. plaque formation in arteriosclerosis ([Greene et al. 1980](#), [Hell et al. 1989](#), [Sawchuk et al. 1999](#)), thrombosis mitigation ([Kameneva et al. 2004](#), [Marhefka et al. 2009](#), [Zhao et al. 2010](#)), and in energy conservation applications based on polymer drag reduction e.g. oil pipelines ([Burger et al. 1982](#)) and air-conditioning systems ([Takeuchi 2012](#)) etc.

7.1.2 *Outline*

To address the issues aforementioned, a high-speed molecular imaging technique has been developed to visualize the underlying mechanisms of how polymer molecules behave in a viscoelastic flow. The technique involves fluorescein labeling of polymer molecules so that they appear visible under epi-fluorescence illumination tailored to the excitation wavelength of the dye. The cost is lower compared to fluorescein-labeling of DNA, making it more feasible for larger scale investigations. For this technique, a high speed camera was employed to capture images at up to 1,000 frames per second (FPS), so that polymer molecular conformation changes i.e. extension and relaxation, at high flow velocities could be observed directly. The recorded molecular conformation was correlated to the state of the flow field, i.e. laminar or turbulent flow, which was quantified using the conventional seeding method via Particle Image Velocimetry (PIV). Although not within the scope of the current research, the developed technique can also be extended to other investigations as well, e.g. drag reduction.

The background and details of the fluorescein labeling procedure will be presented in Section 7.2, and the reaction mechanisms for obtaining the fluorescein-labeled molecules i.e. phPAA-FITC are described. For validation of the labeling procedure and to ensure the visibility of the phPAA-FITC molecules, Raman spectroscopy tests and static imaging tests were conducted and their results will be presented in Sections 7.2.3 and 7.3 respectively. With an appropriate algorithm, the sizes of the fluorescein labeled molecules were measured, and the size distribution of the population was obtained (see Section 7.3.3). Next, the experimental details of the viscoelastic flow will be discussed

in Section 7.4. Experimental runs at two representative flow rates ($Q = 1$ ml/h and $Q = 10$ ml/h) have been conducted, and the analyzed results of the conformation changes in the phPAA-FITC molecules at each flow rate, i.e. the aspect ratio and molecular size, will also be presented in Section 7.5.2. Based on the changes in aspect ratio at the most turbulent flow state investigated, i.e. $Q = 10$ ml/h, a molecular relaxation parameter will be defined in Section 7.5.2 to gain an insight of the molecular time scales in the viscoelastic flow. Finally, the conventional PIV measurement method will be presented in Section 7.6 to relate the observed conformation changes to the flow field measurements, via analyses of mean velocity profiles and turbulent intensities. In addition, to obtain the drop-off frequencies previously described in Chapter 6, the power spectrum of the most turbulent flow state, i.e. $Q = 10$ ml/h, will also be examined in Section 7.6.2. In Section 7.6.2, the obtained frequencies will be correlated with previously presented experimental data (Chapter 6), and with the results of the molecular conformation analyses (Section 7.5.2), and the implications will be discussed.

7.2 Fluorescein labeling

7.2.1 PEO labeling

To observe molecular configurations, fluorescence labeling is often employed. This enables the quantification of the labeled species, and allows tracking of its dynamic interactions in a particular phase e.g. water. Fluorescein labeling of polyethylene oxide (FITC-PEO) with fluorescein isothiocyanate (FITC, Sigma Aldrich) has been reported (Kelly and Santore 1995, Rebar and Santore 1996). The reaction mechanism involves

the FITC chemistry attacking the hydroxyl groups on PEO, and the fluorescein molecule was attached to the PEO molecule chain via a thiourethane link. The reaction mechanics can be written as

whereby $\textcircled{\text{F}}$ refers to the fluorophore. However, because there is only one hydroxyl group on each PEO molecule chain (at its end), only one fluorophore could be labeled on each chain. Thus, observing the conformation of the FITC-PEO molecule would be impossible. In addition, the low fluorophore labeling density would result in low emission of fluorescence intensity. This would be a challenge in this study for high speed visualization of dynamical flows in the range of 1000 frames per second. Indeed, no FITC-PEO molecules could be observed in the experimental trials.

7.2.2 An alternative labeling target - phPAA

To overcome this difficulty, partially hydrolyzed polyacrylamide (phPAA) was chosen as a suitable polymer to be labeled; it has multiple nucleophilic bonding sites after hydrolyzation for FITC labeling along its molecular backbone. phPAA was obtained via alkaline hydrolysis of polyacrylamide (PAA, 5×10^6 - 6×10^6 g/mol, Sigma Aldrich), which is commercially available. Firstly, PAA was dissolved in DI water (5 wt% PAA) and gently stirred for 5 hours. Next, 7 parts of sodium hydroxide stock solution (4 wt% NaOH in H_2O) was added to 10 parts of the PAA mixture (100 ml). To

begin hydrolysis, the mixture was heated to 60 °C, while gently stirred for 48 hours to ensure a maximum degree of hydrolysis. It has been reported that the degree of hydrolysis is limited at approximately 67%, because the accumulation of negative charges on the polymeric chains during hydrolysis results in electrostatic repulsion towards hydroxyl ions (Zeynali and Rabbii 2002). The hydrolysis reaction is as follows;

The reaction results in a sodium carboxylate salt (phPAA), and the release of ammonia gas as a reaction byproduct which could be detected via its pungent smell and with a pH indicator. The resulting phPAA molecules contain polar amide groups and ionizable carboxyl groups, hence imparting water solubility to the molecule. The phPAA molecules were subsequently precipitated (1 part phPAA mixture to 10 parts of anhydrous ethanol) and dried at 50°C. The extracted polymer was then stored at 30% relative humidity in a desiccator for 2 days.

Subsequently phPAA was dissolved in DI water (1 wt% phPAA) and gently stirred for 5 hours. When dissolved in an aqueous solution, the carboxyl groups dissociate into negatively charged macro-molecule (i.e. carboxylate) ions and positively charged (i.e. sodium) ions. The carboxylate ions may also combine with the hydronium ions to form carboxylic acid, while the sodium ions have little reaction with the hydroxide ions. These give rise to the polyelectrolyte properties of the solution, resulting in a higher solution viscosity as compared to PAA. The mixture was cooled to 4 °C, and 2 parts of

fluorescein isothiocyanate stock solution (1 wt% FITC in anhydrous ethanol) was added to 10 parts of the phPAA mixture. Assuming an average molecular weight of 5.5×10^6 g/mol for the parent PAA molecule, the ratio of the number of FITC molecules to the number of binding sites on each phPAA molecule would be approximately 1:15. For the subsequent 8 hours, the mixture was incubated in the dark, while gently stirred. This allows the nucleophilic carboxylate ions on the phPAA molecules to react with the electrophilic isothiocyanate groups on the FITC molecules, to form thiourethane links with the fluorophore (Kelly and Santore 1995). The reaction kinetics is as follows;

This completes the fluorescein labeling procedure for the phPAA molecules, i.e. phPAA-FITC. The number of thiourethane links formed, i.e. attached fluorophores, are expected to be limited by the degree of hydrolysis and steric hindrance. However, provided there are sufficient fluorophores to allow high speed visualization, the labeling density is not of interest for this investigation. Based on the approximated binding ratio (1:15), the resultant phPAA-FITC molecules should have a molecular weight in the range of $6.2 - 7.4 \times 10^6$ g/mol. A second precipitation step was carried out (1 part phPAA-FITC mixture to 10 parts of anhydrous ethanol) to extract the phPAA-FITC molecules. The precipitant was subsequently dialyzed with anhydrous ethanol to remove excess free FITC molecules, and stored in a dark desiccator at 30% relative humidity for 7 days.

7.2.3 Raman Spectroscopy

To validate the fluorescein labeling procedure, Raman spectroscopy tests were carried out for PAA, phPAA, and phPAA-FITC for comparison. The polymers were dissolved in DI water and cast on a flat aluminum film, which was fixed on a glass slide. To remove excess water, the cast films were stored in a desiccator at 30% relative humidity for 7 days. A Raman microscope (Renishaw inVia) with a grating spacing of 1800 lines/mm was used to obtain the Raman spectrum, illuminated by a HeNe laser source ($P = 50 \text{ W max}$, $\lambda_{\text{source}} = 633 \text{ nm}$). The glass slides were placed on the microscope and the laser beam was focused onto the polymer films via a 50X objective lens (Leica Plan Epi, $NA = 0.75$). The resulting spectrum was subsequently obtained over 5 accumulations, using a laser power of 25 W.

To remove background noise, a multipoint baseline routine was used, and the spectrum was normalized to the CH_2 Raman band ($2900\text{-}2940 \text{ cm}^{-1}$). Figure 7-1 shows the normalized spectra for PAA, phPAA, and phPAA-FITC. To validate the first part of the labeling procedure, i.e. hydrolysis, the spectrum should indicate a reduction in amide groups in the phPAA sample (due to the release of ammonia gas). Comparing the phPAA spectrum to the PAA spectrum, a drop in the amide I band (C=O stretch, $1550 - 1770 \text{ cm}^{-1}$) can be observed, followed by the attenuation of the peaks at the amide A band (N-H stretch, $\approx 3190 \text{ cm}^{-1}$) and amide B band (N-H stretch, $\approx 3340 \text{ cm}^{-1}$). These observations validated the hydrolysis procedure. The second part of the labeling procedure involves tagging the fluorophore to the phPAA molecule. Since it has been reported in literature that successful attachment of the fluorophore occurs via a

thiourethane link between the molecules, the phPAA-FITC spectrum should not indicate the presence of isothiocyanate groups. Indeed, a comparison of the phPAA-FITC spectrum against the phPAA spectrum revealed a small peak in the C=S band (C=S stretch, $\approx 1180\text{ cm}^{-1}$), and a drop in the peak of the O-H band (O-H stretch, $\approx 3420\text{ cm}^{-1}$). In addition, no peaks could be observed in the isothiocyanate band (stretch, $2020 - 2100\text{ cm}^{-1}$) of the phPAA-FITC spectrum. This indicates the presence of the FITC molecules, which were successfully attached to the phPAA molecules via the thiourethane link. The phPAA-FITC molecule, with multiple FITC groups attached along its molecular backbone, can now be observed at the appropriate fluorophore excitation wavelength, i.e. at $\lambda_{\text{excitation}} = 492\text{ nm}$.

Figure 7-1. a) Raman spectrum of PAA, phPAA and phPAA-FITC. Significant bandwidth differences are labeled. The insert (b) shows an enlarged view of the C=S band.

7.3 Static Molecular imaging

7.3.1 Imaging setup

Although the reaction kinetics has been verified via Raman spectroscopy (Section 7.2.3), it is important to verify the visibility of the phPAA-FITC molecules during image recording. For this purpose, the precipitated phPAA-FITC were dissolved in DI water (1 wt%) and gently stirred for 8 hours. The mixture was then placed in an enclosed Polymethyl methacrylate (PMMA) channel, which was mounted on an inverted microscope (Nikon Eclipse Ti), and illuminated by a mercury white light source. However, the phPAA-FITC molecules could not be detected under white light because of the weak excitation and emission intensities, as compared to the background illumination. To excite the fluorescein labeled phPAA-FITC molecules and remove background noise, the Nikon B-2A filter configuration ($\lambda_{\text{excitation}} = 450\text{-}490\text{ nm}$, $\lambda_{\text{emission}} = 500\text{ nm}$) which corresponds closely to the FITC excitation and emission wavelengths, was used. The high speed camera (Photron Fastcam SA5, 32 Gb, sensor size = 1024×1024 pixels, pixel size = $20 \times 20\text{ }\mu\text{m}^2$) was used to capture static images of the observed fluorescent phPAA-FITC molecules, via a 10X Nikon microscope objective lens (Plan Fluor, numerical aperture = 0.3, depth of focus $\approx 9\text{ }\mu\text{m}$) for magnification, such that one image pixel corresponds to $2 \times 2\text{ }\mu\text{m}^2$.

7.3.2 Molecular visualization

The sizes of the phPAA molecules appear larger than the parent PAA molecules when dissolved in a solvent (Zeynali and Rabbii 2002). This is due to the repulsive forces between the ionized carboxyl groups, which cause the polymer chain to extend and

uncoil. Hence, it is fundamental to investigate the size of the polymer molecules after fluorescein labeling, subsequent to their dissolution in an aqueous medium i.e. DI water. For this purpose, snapshots of the static phPAA-FITC molecules under the excitation wavelength were taken with an exposure time of 20 ms. A MATLAB algorithm was used to convert the 8-bit images into black-and-white 2-bit format: all digital images are stored in binary format for ease of further processing. Figure 7-2 a – e shows 5 image pairs of the original snapshots taken with the high speed camera, paired with the converted binary image. The images are not displayed to scale, and discussion of the actual size of the molecules will be addressed in the next section. A reasonable degree of accuracy can be observed. This also validated the visibility of the phPAA-FITC molecules under the excitation wavelengths.

Figure 7-2. Snapshots of 5 phPAA-FITC molecule pairs (a – e) showing the original 8-bit image and binary converted image

To ensure the observed phPAA-FITC molecules were not specks of dust or undissolved fluorescein clumps, snapshots of the static molecules were captured under the excitation wavelength, and under white light illumination for comparison purposes. Figure 7-3 a) – c) shows three pairs of snapshots taken, with only the illumination

changed. Note that the location of the snapshots was the same for each pair. From the figures, it is evident that the polymer molecules could only be seen under the excitation wavelength, but appeared to be invisible under white light.

Figure 7-3. Images a) to c) show the snapshots of phPAA-FITC molecules when visualized under the excitation wavelength (left images), in comparison to snapshots obtained under white light illumination (right images).

7.3.3 Molecular size measurement

For analyses of phPAA-FITC molecular sizes, a MATLAB algorithm was applied on the converted binary images, to obtain the 2-D area and dimensions of the phPAA-FITC molecule. As observed in Figure 7-2, the static phPAA-FITC molecules have different shapes and sizes. Hence, to obtain the measurements of the molecules, a standard algorithm was applied across all the binary converted snapshots. The area was obtained by calculating the white pixel count of the molecule and scaling it with the equivalent pixel size i.e. $2 \times 2 \mu\text{m}^2$. An ellipse was then fitted along the perimeter of each molecule to obtain the minor axis (α) and major axis (β), which were subsequently used to calculate the aspect ratio (β/α) to quantify the phPAA-FITC

molecular conformation i.e. extension and relaxation. Figure 7-4 shows the enlarged snapshot of the binary converted image obtained from Figure 7-2a after the ellipse fitting algorithm has been applied.

Figure 7-4. Enlarged snapshots of the binary converted image in Figure 7-2a). The MATLAB algorithm fits an ellipse (green) around the phPAA-FITC molecule and measures the major axis (β) and minor axis (α) respectively.

Figure 7-5 shows a histogram plot on the size distribution, subcategorized into $100 \mu\text{m}^2$ incremental size ranges, and obtained by measuring the area of 1,000 phPAA-FITC molecules. Details of the size distribution have been included in Table C-1 in Appendix C. The average dimension of the unstressed phPAA-FITC molecules is 32 by $22 \mu\text{m}^2$, with an aspect ratio (β/α) of 1.5 which does not vary significantly across the various size ranges. The snapshot images of four molecules of varying sizes have also been included in Insets a) – d). It can be observed that the size distribution of the molecules fall into a large range from $100 \mu\text{m}^2$ to over $1500 \mu\text{m}^2$. This is because the parent PAA used for the hydrolysis had a molecular weight ranging from 5×10^6 g/mol to 6×10^6 g/mol. In addition, mechanical stirring, centrifugation during precipitation, the

hydrolysis reaction and fluorescein-labeling procedure could have caused additional degradation and modifications to the polymer chains.

Figure 7-5. Histogram plot of the size ranges for 1000 phPAA-FITC molecules measured under static conditions. Insets (a-d) show the snapshots of four molecules, and their corresponding sizes.

7.4 Experimental details

To visualize the phPAA-FITC molecules in a dynamical flow, the 3-stream contraction-expansion silicon micro-channel (see Figure 5.1, Section 5.2.1) was employed for the investigation. Here, the main-stream and side-stream liquids used consisted of 1.5 wt% and 0.1 wt% phPAA-FITC in DI water respectively. Similar to the static measurements (Section 7.3), the images were recorded with the high speed camera (Photron SA5) via the 10X Nikon microscope lens (Plan Fluor, numerical aperture = 0.3, depth of focus $\approx 9 \mu\text{m}$), with the Nikon B-2A filter configuration ($\lambda_{\text{excitation}} = 450\text{-}490 \text{ nm}$, $\lambda_{\text{emission}} = 500 \text{ nm}$).

To compare the changes in polymer conformation between two extremities, experimental runs at two representative flow rates will be presented i.e. $Q = 1 \text{ ml/h}$ ($\text{Re} = 0.003$, $\text{De} = 41.46$) and $Q = 10 \text{ ml/h}$ ($\text{Re} = 0.217$, $\text{De} = 414.6$). The minimum flow rate, i.e. $Q = 1 \text{ ml/h}$, was limited by the step size of the syringe pump which could result in significant noise in the velocimetry data at lower flow rates. The maximum flow rate, i.e. $Q = 10 \text{ ml/h}$, was limited by the fluorescent intensity (emitted by the phPAA-FITC molecules) that could be detected by the camera. The recording speeds used at $Q = 1 \text{ ml/h}$ and $Q = 10 \text{ ml/h}$ were 125 FPS and 1000 FPS respectively and exposure times were set to 0.2 ms. Only polymer molecules that remained in focus were examined, while out of focus data were discarded. For analyses of molecular conformation, i.e. extension and relaxation, the MATLAB algorithm used for the static size measurements, i.e. to calculate the area and aspect ratio (refer to Section 7.3), was also applied on the recorded images of the phPAA-FITC molecules in the dynamical

micro-channel flow. Here, the investigation focuses on the flow field downstream of the contraction where turbulence occurred. The properties of the test liquids and corresponding Re and De have been included in Table 7-1.

Table 7-1. Properties of the main-stream and side-stream liquid and the corresponding Re and De.

Properties Fluid	Extensional relaxation time (ms)	Flow rate, Q (ml/h)	Contraction shear rate (s ⁻¹)	Contraction viscosity (Pa·s)	Contraction mean velocity (m/s)	Re	De
Main-stream Liquid	266 ± 18	1	154	0.374	0.014	0.003	41.46
		10	1718	0.055	0.146	0.217	414.6
Side-stream Liquid	19 ± 3	1	623	0.008	0.011	0.086	2.327
		10	5710	0.003	0.103	2.241	23.27

Conventional PIV measurements were also carried out to relate the conformation changes of the phPAA-FITC molecules to the flow field. To facilitate this, the test liquids were seeded with 0.1 wt% polystyrene particles (5 µm, Thermo Scientific). This seeding concentration ensured each interrogation window consists of approximately 10 or more particles at any instant, for obtaining accurate results with a high signal to noise ratio. The Nikon B-2A filter was removed and white light illumination was used. For velocity evaluation, the recording rates were such that the one-quarter rule was observed for each flow rate. Each interrogation window consisted of 64 x 32 pixels, and there was a 50% overlap between consecutive windows. Short exposure times i.e. 2 µs, were used to preserve image sharpness. By processing the images using the same MATLAB algorithm discussed in Chapter 5, the resulting velocimetry data was computed.

7.5 Dynamic molecular visualization

7.5.1 Analyses approach

To reduce blurring effects due to the high velocity of the polymer molecules (i.e. phPAA-FITC) upon exiting the contraction, analyses were only carried out from some distance δ ($= 0.1 - 1.5$ mm) away from the contraction. For definition of δ , refer to Figure 6-2 (Section 6.4.1). To compare the degree of relaxation of the phPAA-FITC molecules between the two flow rates, $(\beta/\alpha)_{\text{normalized}}$ was calculated by normalizing the aspect ratio at each δ , with that at $\delta = 0.1$ mm. Note that β/α is the largest at $\delta = 0.1$ mm for each flow rate, because the phPAA-FITC molecule is at its highest extended state (within the δ range investigated). Therefore, a larger change in $(\beta/\alpha)_{\text{normalized}}$ corresponds to a larger release of stored elastic energy. Separately, conventional PIV was also employed to quantify the flow field, and to facilitate the correlation of the polymer conformation changes to the state of the flow field.

7.5.2 Results

7.5.2.1 Aspect ratio

Figure 7-6a shows the normalized aspect ratio i.e. $(\beta/\alpha)_{\text{normalized}}$, of phPAA-FITC molecules with respect to δ , at flow rates of $Q = 1$ ml/h and $Q = 10$ ml/h respectively. For each flow rate, 20 phPAA-FITC molecules were analyzed. However for clarity, only 5 phPAA-FITC molecules are shown for each flow rate. At $Q = 1$ ml/h, it can be observed that there are no significant changes in molecule aspect ratio and $(\beta/\alpha)_{\text{normalized}}$ fluctuates about unity ($\approx 0.9 - 1$). This means that there is little stretching of the

molecules in the contraction, and hence negligible relaxation of the polymer molecules upon exiting the contraction, which resulted in the insignificant release of stored elastic energy by the pHAA-FITC molecules subsequently.

Figure 7-6. a) Polymer relaxation $(\beta/\alpha)_{\text{normalized}}$ over axial distance ($\delta = 0.1 - 1.5$ mm). Open and closed symbols represent polymers captured at 1 ml/h and 10 ml/h respectively. Significant relaxation occurs at 10 ml/h only. Insets show the relaxation of polymer molecule 9 at b) $\delta = 0.1$ mm and c) $\delta = 1.0$ mm respectively.

However, as the flow rate was increased to $Q = 10$ ml/h, a drastic change could be observed. Significant reductions in molecule aspect ratio can be observed at this flow rate (see Figure 7-6a), whereby $(\beta/\alpha)_{\text{normalized}}$ reduces to a range of $\approx 0.5 - 0.8$ at the end of the recording window. This indicates that the molecules had been significantly

stretched in the contraction, and relaxed significantly upon exiting the contraction, which was accompanied by a corresponding release of stored elastic energy in the phPAA-FITC molecules. From Figure 7-6a, it can also be observed that the molecules relaxed randomly and sporadically. In addition, the extent and rate of relaxation varied among the molecules at this flow rate. These could be due to the non-homogeneity in the molecular weight of the polymer molecules, which were also not uniformly distributed in the solution. The captured images of a phPAA-FITC molecule (see Figure 7-6a, molecule 9) at $Q = 10$ ml/h, have also been included in Figure 7-6b ($\delta = 0.1$ mm) and Figure 7-6c ($\delta = 1.0$ mm) respectively. For this molecule, the aspect ratio decreased drastically by over 40%.

At $Q = 10$ ml/h, most of the molecule relaxation occurs before $\delta = 0.4$ mm (see Figure 7-6a). In addition, it can be observed that $(\beta/\alpha)_{\text{normalized}}$ approached a stabilized value and does not change significantly beyond $\delta = 0.6$ mm. Hence, this indicates that the extensional stresses in the molecules would have almost been completely released further downstream at $\delta = 1.4 - 1.5$ mm. Based on the measurements of 20 polymer molecules for each flow rate at $\delta = 1.4 - 1.5$ mm, the average aspect ratio (β/α) was 1.61 and 1.86 at $Q = 1$ ml/h and $Q = 10$ ml/h respectively. Note that this is larger than the aspect ratio measured under static conditions (i.e. 1.5, Section 7.3). The residual stresses observed here could be a result of shear stresses within the flow, which greatly inhibit the stresses within the molecules to be completely released back to the original non-stressed state. Indeed, the aspect ratio upstream (i.e. 4 mm before contraction) was measured to be 1.56 and 1.76 at $Q = 1$ ml/h and $Q = 10$ ml/h respectively.

7.5.2.2 *Molecular relaxation*

To gain an insight of the molecular dynamics at $Q = 10$ ml/h, it is important to study the relaxation of the polymer molecule. Here we define a parameter, $\tau_{90\%}$, which is the duration required for the aspect ratio of each molecule to decrease to within 10% of its final value (i.e. $\delta = 1.4 - 1.5$ mm), after exiting the contraction. $\tau_{90\%}$ can also be perceived as the duration required for the polymer molecule to relax almost completely, and could be easily computed since each image frame corresponded to 1 ms. Analyzing the conformation changes of over 10 phPAA-FITC molecules at $Q = 10$ ml/h, the average $\tau_{90\%}$ was calculated to be ≈ 28.5 ms. Note the huge disparity (≈ 9 times difference) in comparison to the relaxation time measured using CaBER i.e. 266 ms, which measures only the bulk liquid property and not the molecular relaxation time.

7.5.2.3 *Molecular size*

It can be observed from Figure 7-6b and Figure 7-6c that as the aspect ratio varied, the area occupied by the phPAA-FITC molecule also changed. [Pritchard et al. \(2013\)](#) reported that the volume of PAA is reduced, when it is subjected to uniaxial strain in water (Poisson ratio, $\nu = 0.26$). This phenomenon was also observed in the experiments where the polymer molecules were observed to be smaller in size at the contraction exit, and larger as they relaxed further downstream. To quantify the area change in the molecules, $(A_{\text{final}} - A_{\text{initial}}) / (A_{\text{final}})$ was computed, whereby A_{final} was the average area (obtained from the MATLAB algorithm) computed from $\delta = 1.4 - 1.5$ mm, and A_{initial} was the average area computed near the contraction from $\delta = 0.1 - 0.2$ mm. From Figure 7-6b and Figure 7-6c, it can be calculated that the area of polymer molecule 9

decreased by 33.6% near the contraction exit ($\delta = 0.1$ mm, Figure 7-6b) due to the extensional stresses in the contraction, as compared to the average area at $\delta = 1.4 - 1.5$ mm (Figure 7-6c). Further analyses of 20 molecules for each flow rate showed that the area of each molecule decreased by an average of only 4.4 % at $Q = 1$ ml/h, while a 15.6 % area reduction occurred at $Q = 10$ ml/h. The larger area reduction at 10 ml/h was caused by the larger extensional stresses when the Deborah number is higher.

7.5.3 Discussion

The changes in molecular area and $(\beta/\alpha)_{\text{normalized}}$ observed in Section 7.5.2 had been directly attributed to the release of stored elastic energy in the stretched polymer molecules. The much larger change in area and aspect ratio at $Q = 10$ ml/h was discussed to be due to the larger extensional stresses in the contraction at the higher Deborah number. This resulted in larger amount of stored elastic energy in the polymer molecules, which was released subsequently as the polymer molecule moved further downstream, as compared to that at $Q = 1$ ml/h.

It is clear that this elastic energy gained by the molecules were transferred by the flow field (which can be traced back to the power provided by the syringe pump) to the polymer molecules. This raises an important question: If the polymer molecules relax and release their elastic energy, where does the energy go to? Based on the law of energy conservation, the energy has to be transferred to the surrounding medium, i.e. the bulk liquid.

The bulk liquid should gain a momentary increase in energy when a polymer molecule releases its elastic energy. In addition, based on the discussions in Section 7.5.2 and observations in Figure 7-6 a), this increase in energy would have to be sporadic and random as well. To uncover the characteristics of the energy transfer, flow field measurements provide an approach to investigate this energy flux. For this purpose, the conventional PIV technique would be applied in the subsequent section. The experimental details for PIV have been included in Section 7.4.

7.6 Flow field measurements (PIV)

7.6.1 Mean statistics

To obtain average velocity profiles over a longer time span, 10,000 image pairs were captured at 50 Hz i.e. mode 1 recording (refer to Section 5.3.3). The time interval (Δt) between the two images in each pair was 4 ms and 0.5 ms, at $Q = 1$ ml/h and $Q = 10$ ml/h respectively. These allowed the computation of the normalized mean axial velocity profiles ($\bar{U}_i / \bar{U}_{bulk}$) and turbulent intensities ($u_{i,rms} / \bar{U}_i$) across the channel width. Figure 7-7 shows the normalized mean axial velocity profiles and turbulent intensities for the two flow rates investigated i.e. $Q = 1$ ml/h and $Q = 10$ ml/h, at $\delta = 0.3$ mm and $\delta = 1.0$ mm respectively.

Figure 7-7. Normalized axial velocity (closed symbols) and turbulent intensities (open symbols) at a) $\delta = 0.3$ mm, and b) $\delta = 1.0$ mm.

The 3-stream flow of the phPAA-FITC test liquids followed ‘top hat’ profiles, with a slight increment near both ends. At the lower flow rate ($Q = 1$ ml/h), the velocity profiles at $\delta = 0.3$ mm and $\delta = 1.0$ mm appeared unchanged, due to negligible

relaxation of the viscoelastic liquid. This was supported by the low turbulent intensities at both regions, indicating a laminar flow state (see Figure 7-7a and Figure 7-7b). This showed that there was negligible energy flux transferred from the polymer molecules to the flow field, which is also evident from Figure 7-6, whereby no significant changes in molecule aspect ratio can be observed as the molecules traversed downstream at $Q = 1$ ml/h.

However, as the flow rate was increased to $Q = 10$ ml/h, there was a drastic increase in turbulent intensities at both axial distances. At $\delta = 0.3$ mm, a localized diminished peak can be observed in the normalized mean velocity profile as the flow rate was increased to $Q = 10$ ml/h (see Figure 7-7a). In Section 6.4.2, this peak was attributed to the solid-like behavior of the main-stream liquid, whereby the elastic stresses had not been completely released due to the higher Deborah number. This hypothesis is also supported by the relatively attenuated turbulent intensity (see Figure 7-7a) at the channel midpoint (i.e. 0.5 mm), as compared to the surrounding flow field. Physically, this means that the polymer molecules had not completely relaxed at this axial position. Indeed, on closer examination of Figure 7-6a (see Section 7.5.2), it can be observed that most of the molecule relaxation was only completed at approximately $\delta = 0.6$ mm when $Q = 10$ ml/h. Hence, the hypothesis of the ‘solid-like’ behavior and incomplete release of stresses in the main-stream liquid (at $\delta = 0.3$ mm) is supported, and this is in good agreement with the localized diminished peak observed.

At $\delta = 1.0$ mm, the flow at $Q = 10$ ml/h was turbulent, characterized by large turbulent intensities across the whole micro-channel (see Figure 7-7b). The peak in the

normalized mean velocity profile is no longer present, although it did not relax completely to converge with the profile of the lower flow rate ($Q = 1$ ml/h). This indicates the main-stream liquid had transformed from the ‘solid-like’ to a ‘liquid-like’ state, and the stresses in the polymer molecules (i.e. stored elastic energy) had been released. From Figure 7-6a, this is indeed true because it can be observed that $(\beta/\alpha)_{\text{normalized}}$ reached a stabilized value and does not change significantly beyond $\delta = 0.6$ mm, indicating the elastic energy in the molecules has been almost completely transferred to the flow field when the molecules passes $\delta = 1.4 - 1.5$ mm.

The turbulent characteristics of the flow field observed here at $Q = 10$ ml/h are in good agreement with the non-uniform and sporadic release of stored elastic energy by the polymer molecules, as observed in Figure 7-6a (see Section 7.5.2). This erratic transfer of the elastic energy to the flow field resulted in localized increase in kinetic energy (i.e. velocity fluctuations) which was random in space and time, causing the flow to become turbulent.

7.6.2 Power spectrum

In Section 6.9, it has been discussed and observed how the molecular weight and bulk properties of the liquid affect the power spectrum of the velocity fluctuations at high Deborah numbers. Two drop-off frequencies were observed. The first frequency was found to relate to the molecular weight of the polymer used, while the second frequency corresponded more closely to the bulk fluid property, i.e. the CaBER extensional relaxation time. Here, we investigate the power spectrum of the velocity

fluctuations at $Q = 10$ ml/h. The velocity measurements were obtained by analyzing the images captured using recording mode 2 (refer to Section 5.3.3), with a time interval (Δt) of 0.5 ms between images, over a duration of 38 seconds. In the current study, only one power law slope (i.e. region 2) was observed in the power spectrum at $\delta = 0.3$ mm, while two were observed at $\delta = 1.0$ mm. This delayed development of the turbulent flow could be due to the much larger Deborah numbers here as compared to that in Section 6.9, resulting in a delayed relaxation of the polymer molecules and hence a retardation in the energy transfer to the flow field.

Figure 7-8 shows the log-log power spectrum plot of the axial velocity fluctuations at $\delta = 1.0$ mm. Similarly to Section 6.9 (Fig. 6-17), three distinct regions can be observed. However, no linear fitting was done for Region 1 due to the appearance of large scale peaks at low frequencies which hindered the development of a plateau. These were not observed in the PEO experiments in Chapter 6. On further examination, the large scale peaks (see Inset, black line) were found to be augmentations of smaller peaks present in the power spectrum of the upstream vortices (see Inset, blue line). Hence, the characteristics of upstream flow dynamics were transported downstream, and amplified as the polymer stresses were released. Demarcating region 2 to be the linear region between 1.1 – 1.5 on the $\log_{10}$ frequency axis, it can be observed that the power spectra cease to be linear on values smaller than 1.1 (i.e. 12.6 Hz). This corresponds to the 1st drop-off frequency i.e. 12-17 Hz, of the PEO molecules with molecular weights of 8×10^6 g/mol in Section 6.9. However, the molecular weight of the pHAA-FITC molecules here was estimated to range between $6.2 - 7.4 \times 10^6$ g/mol (refer to Section

7.2.2). The small discrepancy observed here could be exacerbated by the lack of a linear region 1 due to the much larger Deborah number, and the exact phPAA-FITC binding ratio being unknown. Further investigation is required to validate this.

Figure 7-8. Log-log power spectrum plot of velocity fluctuations for 10 ml/h, at $\delta = 1.0$ mm. The inset shows the power spectrum of the downstream flow field ($\delta = 1.0$ mm, black line) in comparison with the upstream vortex (blue line) from $\text{Log}_{10}(\text{frequency}) = 0.1 - 1.1$.

In contrast to Region 1, good linearity trends were observed in Regions 2 and 3 (see Figure 7-8) and the same fitting method in Chapter 6 was applied. The regions were found to follow power law slopes of -1.9 (region 2) and -3.3 (region 3) respectively. Based on the linear fitting, the 2nd drop-off frequency between regions 2 and 3 was calculated to be 38.4 Hz. This distinct frequency was not observed in the power spectrum of the upstream vortices, and thus reinforces the findings in Section 6.9 that it is related to the relaxation properties of the viscoelastic liquid downstream. In fact, the 2nd drop-off frequency obtained is in good agreement with the findings in Section 6.9. To visualize how well the data obtained here fits into the previous data, Figure 7-9

shows the data point obtained here (circled in blue) together with all other points presented for the PEO molecules (from Figure 6-18, Section 6.9).

Figure 7-9. Log-log plot depicting the relationship between the 2nd drop-off frequency and bulk liquid behaviour. The data obtained in this section is circled in blue, showing a good agreement with the trend observed for the PEO molecules in Section 6.9.

On further analysis, the 2nd drop-off frequency obtained here was found to correspond closely to the molecular relaxation frequency (i.e. $f = 1/\tau_{90\%} = 35.04$ Hz) calculated in Section 7.5.2.2. This shows that in addition to the relaxation properties of the bulk liquid (i.e. $f = 1/\lambda_{\text{bulk}} = 3.76$ Hz), the 2nd drop-off frequency correlates more closely with the molecular relaxation behavior in the flow field. Based on the law of energy conservation, this further reinforces the 2nd drop-off frequency (i.e. molecular relaxation frequency) as the effective energy injection (i.e. forcing) frequency, whereby the polymer molecules transfer their stored elastic energy into the turbulent flow.

7.7 Summary

7.7.1 Overview

A novel high-speed molecular imaging technique has been presented in this chapter. The technique directly visualizes the conformation changes of polymer molecules in a viscoelastic flow and bypasses the conventional indirect approach i.e. motion analyses of tracer additives. The development of the technique begins with the fluorescein labeling of polymer molecules so that they appear visible when illuminated at the excitation wavelength. Polyacrylamide (PAA) was modified via hydrolysis to obtain partially hydrolyzed PAA (phPAA) which had multiple binding sites for the fluorescein molecules (FITC). The labeling ends with an incubation process involving the electrophilic attack of the carboxyl groups on the phPAA molecules by the isothiocyanate groups on the FITC molecules, to obtain the fluorescein labeled molecule (phPAA-FITC). The whole fluorescein labeling procedure was then validated via Raman spectroscopy tests. Static molecular imaging was subsequently conducted to ensure the visibility of the molecules under the excitation wavelength. An ellipse fitting algorithm was applied to measure the major-axis (β) and minor-axis (α) of 1,000 static and unstressed phPAA-FITC molecules to calculate their aspect ratio (β/α). The algorithm was also used to measure the size of the molecules, which ranged from $100 \mu\text{m}^2$ to over $1500 \mu\text{m}^2$. Despite the large distribution, the aspect ratio of the molecules (≈ 1.5) did not differ significantly across the size ranges.

Experimental runs at two representative flow rates i.e. $Q = 1 \text{ ml/h}$ and $Q = 10 \text{ ml/h}$, were then investigated from $\delta = 0.1 - 1.5 \text{ mm}$ to eliminate blurring at the contraction

exit. The main-stream and side-stream liquids consisted of 1.5 wt% and 0.1 wt% phPAA-FITC in DI water respectively. To compare the degree of relaxation of the phPAA-FITC molecules between the two flow rates, $(\beta/\alpha)_{\text{normalized}}$ was calculated by normalizing the aspect ratio at each δ , with that at $\delta = 0.1$ mm. At $Q = 1$ ml/h, no significant changes in $(\beta/\alpha)_{\text{normalized}}$ and size (≈ 4.4 % reduction) of the phPAA-FITC molecules were observed, indicating negligible stretching and relaxation of the molecules. In contrast, drastic reductions in $(\beta/\alpha)_{\text{normalized}}$ (> 40 %) and size (≈ 15.6 % reduction) were observed at $Q = 10$ ml/h. This indicated significant relaxation due to the polymer molecules being highly stretched in the contraction. Most of the molecule relaxation was observed to have occurred before $\delta = 0.4$ mm, with no significant changes past $\delta = 0.6$ mm. The extent and rate of relaxation were random and sporadic, which could be due to the non-homogenous molecular weight of the phPAA-FITC molecules and their non-uniform distribution in the viscoelastic liquid. The average aspect ratio (β/α) was 1.61 and 1.86 at $Q = 1$ ml/h and $Q = 10$ ml/h respectively, when measured at the end of the recording window ($\delta = 1.4 - 1.5$ mm). The deviation in aspect ratio as compared to measurements under static conditions (i.e. $\beta/\alpha = 1.5$), was due to shear stresses in the flow. This was validated by the aspect ratio measurements upstream of the contraction, which was 1.56 and 1.76 at $Q = 1$ ml/h and $Q = 10$ ml/h respectively. To gain an insight into the molecular dynamics at $Q = 10$ ml/h, the relaxation of the molecules was studied and $\tau_{90\%}$ was measured to be 28.5 ms; this is much smaller than the relaxation time of the bulk liquid i.e. 266 ms.

PIV was subsequently carried out to relate the flow field statistics to the conformation changes of the phPAA-FITC molecules. At $Q = 1$ ml/h, the flow was laminar with low turbulent intensities. This corresponds well with the molecular visualizations whereby no significant changes in molecule aspect ratio were observed. In contrast, as the flow rate was increased to $Q = 10$ ml/h, there were drastic increases in turbulent intensities at both $\delta = 0.3$ mm and $\delta = 1.0$ mm. At $\delta = 0.3$ mm, the mean velocity profile showed a localized diminished peak, indicating an incomplete release of stresses in the main-stream liquid i.e. solid-like behavior. This is in good agreement with the attenuated turbulent intensities at the channel midpoint, and earlier molecular observations which showed that most of the relaxation occurred before $\delta = 0.4$ mm. At $\delta = 1.0$ mm, the peak in the mean velocity profile had disappeared and the turbulent intensities were large across the whole channel width. This indicated that the main-stream liquid had transformed from the ‘solid-like’ to a ‘liquid-like’ state, and most of the stored elastic energy in the phPAA-FITC molecules had been released. This corresponds well with the molecular observations that $(\beta/\alpha)_{\text{normalized}}$ reached a stabilized value and does not change significantly beyond $\delta = 0.6$ mm. The turbulent characteristics observed at $Q = 10$ ml/h are in good agreement with the non-uniform and sporadic release of stored elastic energy by the polymer molecules, as observed in Section 7.5.2.

The 3 regions in the power spectrum were observed only at $\delta = 1.0$ mm, due to the delayed development of the turbulent flow at the high Deborah number investigated. However, the 1st drop-off frequency could not be obtained accurately because region 1 consisted of large scale peaks. Nevertheless, the 1st drop-off frequency was estimated to

be ≈ 12.6 Hz, which falls into the range of the PEO molecules with molecular weights of 8×10^6 g/mol in Section 6.9. Further investigation is required because the phPAA-FITC molecules were estimated to have a molecular weight range of $\approx 6.2 - 7.4 \times 10^6$ g/mol (refer to Section 7.2.2). In contrast, good linearity trends were observed in Regions 2 and 3 with the fitting method in Chapter 6, indicating a 2nd drop-off frequency of 38.4 Hz which corresponded well with previous findings in Section 6.9. More importantly, the 2nd drop-off frequency corresponded closely to the molecular relaxation frequency calculated in Section 7.5.2.2. This validates the 2nd drop-off frequency as the forcing frequency (i.e. energy injection point) whereby the polymer molecules transfer their stored elastic energy to the flow field, causing it to be turbulent.

7.7.2 *Final remarks*

The analyses of the phPAA-FITC molecular conformations were conducted from $\delta = 0.1 - 1.5$ mm. The extent of molecular stretching is expected to be larger nearer to the contraction exit, and molecular extension could have begun upstream before the molecule entered the contraction, due to flow focusing of the main-stream liquid. To gain a broader perspective of how a phPAA-FITC molecule would behave as it flows past the contraction, Figure 7-10 shows the snapshots of a single phPAA-FITC molecule taken at various time intervals at $Q = 10$ ml/h, with a capturing speed of 500 FPS and an exposure time of 50 μ s. The image was obtained with a 4X Nikon microscope objective (Plan Apo, NA = 0.2, DOF ≈ 27 μ m) to obtain a larger overview of the micro-channel. However due to this lower magnification, the images obtained have not been used for analyses in this chapter because the low pixel count for the

minor-axes ($\approx 2 - 3$ pixels) of the molecules would result in large errors. Nevertheless, the major-axes had a more significant pixel count ($\approx 8 - 15$ pixels) and an investigation provided some interesting insights of the polymer behavior.

Figure 7-10. Visualization of a single phPAA-FITC molecule flowing past the contraction. Note that the images belong to the same phPAA-FITC molecule. The green lines mark the boundaries of the micro-channel.

Figure 7-11 shows the major-axes (i.e. length) of the phPAA-molecule in the vicinity of the contraction, obtained with the same algorithm in Section 7.3. The average displacement of the polymer molecule in the contraction (i.e. $x = 1 - 2$ mm) was calculated to be ≈ 44 pixels per frame ($1/500$ s). This translates to the molecule requiring $45 \mu\text{s}$ to travel across 1 pixel. Hence, blurring is minimal since the shutter exposure of $50 \mu\text{s}$ was only slightly longer. From the figure, extension of the molecule can be observed to have occurred before the molecule entered the contraction. The molecule stretched to its maximum extended length at the corner of the contraction exit, before abruptly relaxing to a length which was less than that measured upstream. Hence, the present results show that the molecule extended upstream when subjected to an accelerating flow field. In addition, the singular point (i.e. corner) causes the molecule to be stretched to a greater extent, exceeding the length measured in the contraction.

Figure 7-11. Analyses of the phPAA-FITC molecule length in Figure 7-10. The molecule had been stretched upstream before entering the contraction, and attained its maximum extended length at the singular point of the exit.

8. Conclusions

8.1 Overview

The objectives of this study were 1) to examine the characteristics of a viscoelastic turbulent flow field and compare with that of high-Re turbulence, by applying statistical analyses and flow visualization tools, 2) to investigate the effects of varying molecular and bulk liquid properties on the resultant flow field properties, and explore the possibility of extracting information from the flow dynamics to deduce the molecular and bulk liquid properties, and 3) to develop a technique to examine real-time observations of the molecular conformation changes (i.e. extension and relaxation) in a viscoelastic turbulent flow, and relate the observations to the dynamics of the flow field. To meet these objectives, a 3-stream micro-channel design similar to [Gan et al. \(2007\)](#) was chosen. The focus of this study was placed on the downstream flow, which was reported to be chaotic and unstable ([Gan et al. 2007](#), [Lam et al. 2009](#), [Gan and Lam 2012](#)).

Objective 1

To characterize the viscoelastic turbulent flow downstream and understand its similarities to high-Re turbulence, PIV was employed to quantify the flow field to enable statistical analyses and flow field visualization. The investigation was carried out with 1.0%-2M as the main-stream liquid, and 0.1%-2M as the side-stream liquids, for $2 \text{ ml/h} < Q < 20 \text{ ml/h}$ ($0.019 \leq \text{Re} \leq 0.528$ and $27 \leq \text{De} \leq 278$). For the range of flow rates investigated, the mean velocity profiles at two axial locations ($\delta = 0.3 \text{ mm}$

and $\delta = 1.0$ mm) have been presented. At $Q = 20$ ml/h, the mainstream fluid could not sufficiently relax at $\delta = 0.3$ mm as compared to the lower flow rates. In these cases, the mainstream fluid traverses as an integral solid-like body, resulting in a central peak in velocity profile. However, this peak was absent at $\delta = 1.0$ mm, which may indicate that much of the stored elastic stresses in the main-stream liquid have been relaxed; this was accompanied by a larger turbulent intensity near the channel center-line at $\delta = 1.0$ mm, as compared to that at $\delta = 0.3$ mm. Generally, turbulent intensities increased with increasing flow rate (i.e. De) at each δ , and were not attenuated further downstream (i.e. $\delta = 2.0$ mm) for flow rates of $Q = 14$ ml/h and $Q = 20$ ml/h respectively. At the most turbulent flow state investigated i.e. $Q = 20$ ml/h, large Reynolds stresses were present at $\delta = 0.3$ mm, which decreased with increasing distance from the contraction. Such high turbulent intensities and Reynolds stresses have only been reported in high-Re turbulent flows ([Tamano et al. 2009](#), [Japper-Jaafar et al. 2010](#)).

The single point velocity statistics at 3 points ($\delta = 0.3, 0.5, 1.0$ mm) were recorded using modes 1 (low frequency) and 2 (high frequency). Both low frequency and high frequency measurements showed that velocity fluctuations increased drastically with increasing De . At the most turbulent state investigated ($Q = 20$ ml/h), the single point autocorrelation of the velocity fluctuations at $\delta = 0.3$ mm decayed rapidly to zero. A comparison between the single point autocorrelation function and the spatial correlation function revealed a good quantitative agreement within the maximum calculated spatial-correlation. The Taylor's hypothesis was assumed to be valid, and the correlation function decreased to zero at a spatial lag of 1.44 mm, which is of

considerable size as compared to the micro-channel dimensions. Eddy visualization techniques for high-Re turbulent flows were also employed in this study. Galilean decomposition of the most turbulent flow state (i.e. $Q = 20$ ml/h) revealed the presence of a large eddy (≈ 1.4 mm x 0.7 mm), and the upward and downward ‘sweeping’ motion of the main-stream liquid previously observed by [Gan et al. \(2007\)](#). This ‘sweeping’ motion was also uncovered as two dominant modes in the snapshot POD analyses of the turbulent flow field, which showed that the viscoelastic turbulent flow was highly complex.

The power spectra of the velocity fluctuations were then evaluated, for $1 \text{ Hz} < \text{frequency} < 1000 \text{ Hz}$. Such high frequency ranges have not been reported in the literature. As flow rates (i.e. De) were increased, the flow field loses its stability, and the power of fluctuations at lower frequencies increased. For the most turbulent flow state investigated ($Q = 20$ ml/h), two power law decays could be observed, with corresponding slopes of $\approx -5/3$ and -3 respectively. This is a characteristic of 2-D high-Re turbulent flows ([Kraichnan 1967](#), [Leith 1968](#), [Batchelor 1969](#)), which has not been reported in wall bounded viscoelastic turbulent flows in the literature. The 2-D characteristics of the turbulent flow field could be due to the two dominant modes i.e. upward and downward ‘sweeping’ motion of the main-stream liquid, and was observed in the Galilean decomposition and POD analyses.

Objective 2

Although power law decays similar to that in high-Re turbulent flows was observed in the analyses, it should be noted that the mechanism of viscoelastic instability is

inherently different. Based on Kolmogorov's hypothesis, the $-5/3$ power law decay occurs in the inertial sub-range, where energy is transferred from large eddies (energy containing range) to small ones (dissipation range). In viscoelastic turbulence, the opposite is true. Energy is transferred from the smaller scales to the larger scales via the random relaxation of the polymer molecules, i.e. energy injection frequencies. This observation has not been discussed in details in the literature.

The viscoelastic turbulent flow was found to be 2-D in nature. The energy injection point, i.e. forcing frequency, could be easily located in the power spectra, at the intersection between the two power law decays. Since viscoelastic turbulence is due to the random relaxation of polymer molecules, the location of the energy injection point could possibly be correlated to the relaxation properties of the main-stream liquid. To test this hypothesis, the power spectra of seven viscoelastic liquids consisting of PEO of varying molecular weights and concentrations were compared at $De \approx 250$. Indeed, the energy injection point was found to correlate with the bulk relaxation properties of the main-stream liquid, which is possibly the effective energy injection frequency of the liquid. In addition, another interesting discovery was the correlation between the molecular weight and the 1st drop-off frequency, which showed that the size of the largest coherent structures receiving the energy, and the range of the inverse energy cascade, were dependent on the polymer molecular weight. Hence, the results verify the hypothesis that the time scales of the viscoelastic liquid have significant effects on the 'structure' size and energy cascade in the power spectrum. The transfer of elastic energy from the polymer molecules to the flow field, via the correlation of test liquid

properties to the viscoelastic turbulent flow field statistics presented here, has yet to be reported in the literature.

Objective 3

In Chapter 7, a high-speed molecular imaging technique i.e. up to 1,000 frames per second, was developed. The technique is less costly and thus more suitable for large throughput channel flows, as compared to stained DNA molecules. The procedure involves fluorescein labeling of phPAA molecules, which was obtained after hydrolyzation of PAA. The resultant fluorescein tagged molecules (i.e. phPAA-FITC) were subsequently validated with Raman spectroscopy. In addition, the static size distribution of the phPAA-FITC molecules were measured under epi-fluorescence illumination, based on an ellipse fitting algorithm in MATLAB. The algorithm was also used to investigate the conformation changes (i.e deformation) of the phPAA-FITC molecules at two experimental flow rates in the 3-stream micro-channel (main-stream = 1.5 wt% phPAA-FITC, side-stream = 0.1 wt% phPAA-FITC). To relate the molecular behavior to the flow dynamics, the flow field was quantified using the conventional seeding method (via PIV) concurrently.

When the flow was laminar ($Q = 1$ ml/h, $De = 41.46$), there was negligible stretching and relaxation of the polymer molecules and the flow field was characterized by low turbulent intensities. However when the flow field was turbulent ($Q = 10$ ml/h, $De = 414.6$), the molecules were observed to be highly stretched (i.e. deformation via velocity gradients) with a significant reduction in size at the contraction exit. As the molecules travelled downstream, they relaxed (i.e. via molecular elasticity) and

regained almost their original size, causing the flow field to be turbulent and characterized by large turbulent intensities. The extent and rate of relaxation were random and sporadic, indicating the non-homogeneity of the viscoelastic liquid. Most of the molecular relaxation was observed to have occurred before $\delta = 0.4$ mm, and the bulk behavior of the main-stream liquid was solid-like before that (i.e. at $\delta = 0.3$ mm), as evident from the mean velocity profiles.

The same 2-D turbulent power spectrum was observed only at $\delta = 1.0$ mm, due to the delayed relaxation of the polymer molecules at the much higher Deborah number investigated. However, the 1st drop off frequency was not distinct due to large scale peaks at low frequencies. Nevertheless, the 2nd drop-off frequency (i.e. energy injection point) corresponded closely to the bulk liquid properties, and more importantly to $\tau_{90\%}$ (average time taken for pHAA-FITC molecules to relax by 90%), which validates the energy injection point as the effective transfer of stored elastic energy to the flow field, resulting in viscoelastic turbulence.

In conclusion, the 3 objectives of this study have been successfully accomplished. The viscoelastic turbulent flow field has been characterized, the molecular and bulk liquid properties have been shown to affect the energy cascades in the power spectrum, and a technique has been developed to visualize the polymer molecules in a viscoelastic turbulent flow, and was related with the flow dynamics.

8.2 Future Works

Molecular mixture

In the present study, the main-stream liquid consists of only 1 type of polymer molecule for each set of the experiments conducted, and the energy injection point in the power spectra is distinct. However, when the liquid composition consists of 2 or more types of molecules e.g. of different molecular weight etc., it would be interesting to investigate the mixture effect on the power spectra, to study if two distinct energy injection points can be observed. The same could be done to investigate the largest ‘structure’ size of the mixture, which can be obtained using the 1st drop-off frequency as an appropriate measure. However, based on the results of this study, a limitation exists in which the two drop-off frequencies, if observed, would only be clearly discernible when the molecular weights and relaxation times of the different molecules are very different e.g. ≈ 4 times difference.

Molecular weight measurement

Although the fluorescein-labelling procedure has been validated, its molecular weight has not been measured. This is important because it enables the quantification of the FITC-tagging efficiency, so that the fluorescein labeling procedure can be optimized. The molecular weight can be measured via gel permeation chromatography (GPC) or nuclear magnetic resonance (NMR) spectroscopy etc.

Laser illumination

In the experiments, a mercury lamp was used to excite the fluorescein labeled phPAA-FITC to emit fluorescence. However, with a high intensity laser source, the fluorescent intensity emitted by the molecules would be increased. This would enable higher image capturing speeds to be attained.

Molecular-particle interactions

The change in molecular size could result in localized stresses in the flow field. One way to investigate this is to seed the liquid with a large amount of small sized fluorescent particles e.g. $\approx 0.5 \mu\text{m}$, to increase the number of particles surrounding each visible phPAA-FITC molecule, so that the interactions between the polymer molecules and surrounding particles can be observed when the molecular size changes.

Stress probes

The phPAA-FITC molecules could also be used as stress probes to quantify localized stresses. However, it would be necessary to carry out stress tests via single molecule micromanipulation e.g. using atomic force cantilevers ([Florin et al. 1994](#)) or optical tweezers ([Simmons et al. 1996](#)) etc., to measure the intra-molecular forces (i.e. polymer elasticity). This would allow the conformation length of the molecules to be related to the localized stress it experiences in a hydrodynamic flow field via a calibration curve or equation obtained from the measurements. In addition, this technique can also be used to measure the relaxation time of a single molecule.

Blank Page

References

- Abedijaberi, A., & Khomami, B. (2011). Continuum and multi-scale simulation of mixed kinematics polymeric flows with stagnation points: Closure approximation and the high Weissenberg number problem. *Journal of Non-Newtonian Fluid Mechanics*, 166(11), 533-545.
- Adrian, R. J., Christensen, K. T., & Liu, Z. C. (2000). Analysis and interpretation of instantaneous turbulent velocity fields. *Experiments in fluids*, 29(3), 275-290.
- Afonso, A. M., Oliveira, P. J., Pinho, F. T., & Alves, M. A. (2011). Dynamics of high-Deborah-number entry flows: a numerical study. *Journal of Fluid Mechanics*, 677, 272-304.
- Ahmed, S. A., & Giddens, D. P. (1983). Flow disturbance measurements through a constricted tube at moderate Reynolds numbers. *Journal of Biomechanics*, 16(12), 955-963.
- Alves, M. A., & Poole, R. J. (2007). Divergent flow in contractions. *Journal of Non-Newtonian Fluid Mechanics*, 144(2), 140-148.
- Alves, M. A., Pinho, F. T., & Oliveira, P. J. (2005). Visualizations of Boger fluid flows in a 4: 1 square-square contraction. *AIChE Journal*, 51(11), 2908-2922.
- Alves, M. A., Pinho, F. T., & Oliveira, P. J. (2008). Viscoelastic flow in a 3D square/square contraction: visualizations and simulations. *Journal of Rheology (1978-present)*, 52(6), 1347-1368.
- Amarouchene, Y., & Kellay, H. (2002). Polymers in 2D turbulence: Suppression of large scale fluctuations. *Physical Review Letters*, 89(10), 104502.
- Arratia, P. E., Thomas, C. C., Diorio, J., & Gollub, J. P. (2006). Elastic instabilities of polymer solutions in cross-channel flow. *Physical review letters*, 96(14), 144502.
- Baloch, A., Townsend, P., & Webster, M. F. (1996). On vortex development in viscoelastic expansion and contraction flows. *Journal of Non-Newtonian Fluid Mechanics*, 65(2), 133-149.

- Barnes, H. A., Hutton, J. F., & Walters, K. (1989). *An Introduction to Rheology* (Vol. 3). Netherlands. AE: Elsevier.
- Batchelor, G. K. (1969). Computation of the Energy Spectrum in Homogeneous Two-Dimensional Turbulence. *Physics of Fluids (1958-1988)*, 12(12), II-233.
- Bazilevsky, A. V., Entov, V. M., & Rozhkov, A. N. (1990, January). Liquid filament microrheometer and some of its applications. In *Third European Rheology Conference and Golden Jubilee Meeting of the British Society of Rheology* (pp. 41-43). Springer Netherlands.
- Bec, J., Biferale, L., Cencini, M., Lanotte, A. S., & Toschi, F. (2006). Effects of vortex filaments on the velocity of tracers and heavy particles in turbulence. *Physics of Fluids (1994-present)*, 18(8), 081702.
- Benzi, R. (2010). A short review on drag reduction by polymers in wall bounded turbulence. *Physica D: Nonlinear Phenomena*, 239(14), 1338-1345.
- Berti, S., Bistagnino, A., Boffetta, G., Celani, A., & Musacchio, S. (2008). Two-dimensional elastic turbulence. *Physical Review E*, 77(5), 055306.
- Bird, R. B., Armstrong, R. C., & Hassager, O. (1987). Dynamics of polymeric liquids. Volume 1: fluid mechanics. *A Wiley-Interscience Publication, John Wiley & Sons*.
- Boffetta, G. (2007). Energy and enstrophy fluxes in the double cascade of two-dimensional turbulence. *Journal of Fluid Mechanics*, 589, 253-260.
- Boffetta, G., & Musacchio, S. (2010). Evidence for the double cascade scenario in two-dimensional turbulence. *Physical Review E*, 82(1), 016307.
- Boffetta, G., Celani, A., & Musacchio, S. (2003). Two-dimensional turbulence of dilute polymer solutions. *Physical Review letters*, 91(3), 034501.
- Bonn, D., Ingremeau, F., Amarouchene, Y., & Kellay, H. (2011). Large velocity fluctuations in small-Reynolds-number pipe flow of polymer solutions. *Physical Review E*, 84(4), 045301.

- Bousfield, D. W., Keunings, R., Marrucci, G., & Denn, M. M. (1986). Nonlinear analysis of the surface tension driven breakup of viscoelastic filaments. *Journal of Non-Newtonian Fluid Mechanics*, 21(1), 79-97.
- Brandrup, J., Immergut, E. H., & Grulke, E. A. (1975). *Polymer Handbook* Wiley. New York, 483-497.
- Bryce, R. M., & Freeman, M. R. (2010). Abatement of mixing in shear-free elongationally unstable viscoelastic microflows. *Lab on a Chip*, 10(11), 1436-1441.
- Burger, E. D., Munk, W. R., & Wahl, H. A. (1982). Flow increase in the Trans Alaska Pipeline through use of a polymeric drag-reducing additive. *Journal of petroleum Technology*, 34(02), 377-386.
- Burghlea, T., Segre, E., Bar-Joseph, I., Groisman, A., & Steinberg, V. (2004). Chaotic flow and efficient mixing in a microchannel with a polymer solution. *Physical Review E*, 69(6), 066305.
- Chertkov, M. (2000). Polymer stretching by turbulence. *Physical review letters*, 84(20), 4761-4764.
- Chiba, K., Sakatani, T., & Nakamura, K. (1990). Anomalous flow patterns in viscoelastic entry flow through a planar contraction. *Journal of Non-Newtonian Fluid Mechanics*, 36, 193-203.
- Clasen, C., Verani, M., Plog, J. P., McKinley, G. H., & Kulicke, W. M. (2004). Effects of polymer concentration and molecular weight on the dynamics of visco-elasto-capillary breakup. *Proc. XIVth Int. Cong. on Rheology. Seoul, Korea, publ. Korean Society of Rheology*.
- Cruz, F. A., Poole, R. J., Afonso, A. M., Pinho, F. T., Oliveira, P. J., & Alves, M. A. (2014). A new viscoelastic benchmark flow: Stationary bifurcation in a cross-slot. *Journal of Non-Newtonian Fluid Mechanics*, 214, 57-68.
- Dallas, V., Vassilicos, J. C., & Hewitt, G. F. (2010). Strong polymer-turbulence interactions in viscoelastic turbulent channel flow. *Physical Review E*, 82(6), 066303.

- Dontula, P., Macosko, C. W., & Scriven, L. E. (1998). Model elastic liquids with water-soluble polymers. *AIChE journal*, 44(6), 1247-1255.
- Entov, V. M., & Hinch, E. J. (1997). Effect of a spectrum of relaxation times on the capillary thinning of a filament of elastic liquid. *Journal of Non-Newtonian Fluid Mechanics*, 72(1), 31-53.
- Evans, R. E., & Walters, K. (1989). Further remarks on the lip-vortex mechanism of vortex enhancement in planar-contraction flows. *Journal of Non-Newtonian Fluid Mechanics*, 32(1), 95-105.
- Florin, E. L., Moy, V. T., & Gaub, H. E. (1994). Adhesion forces between individual ligand-receptor pairs. *Science*, 264(5157), 415-417.
- Flory, P. J. (1953). *Principles of polymer chemistry*. New York, NY: Cornell University Press.
- Fouxon, A., & Lebedev, V. (2003). Spectra of turbulence in dilute polymer solutions. *Physics of Fluids (1994-present)*, 15(7), 2060-2072.
- Fukunaga, R. (1990). *Statistical pattern recognition*. Academic Press.
- Gan, H. Y., & Lam, Y. C. (2012). Experimental observations of flow instabilities and rapid mixing of two dissimilar viscoelastic liquids. *AIP Advances*, 2(4), 042146.
- Gan, H. Y., Lam, Y. C., Nguyen, N. T., Tam, K. C., & Yang, C. (2007). Efficient mixing of viscoelastic fluids in a microchannel at low Reynolds number. *Microfluidics and Nanofluidics*, 3(1), 101-108.
- Gillissen, J. J. J. (2008). Polymer flexibility and turbulent drag reduction. *Physical Review E*, 78(4), 046311.
- Gonzalez, R. C., Woods, R. E., & Eddins, S. L. (2004). *Digital image processing using MATLAB*. New Jersey, NJ: Pearson Prentice Hall.
- Greene, H. L., Mostardi, R. F., & Nokes, R. F. (1980). Effects of drag reducing polymers on initiation of atherosclerosis. *Polymer Engineering & Science*, 20(7), 499-504.

- Grilli, M., Vázquez-Quesada, A., & Ellero, M. (2013). Transition to turbulence and mixing in a viscoelastic fluid flowing inside a channel with a periodic array of cylindrical obstacles. *Physical review letters*, 110(17), 174501.
- Groisman, A., & Steinberg, V. (2000). Elastic turbulence in a polymer solution flow. *Nature*, 405(6782), 53-55.
- Groisman, A., & Steinberg, V. (2001a). Efficient mixing at low Reynolds numbers using polymer additives. *Nature*, 410(6831), 905-908.
- Groisman, A., & Steinberg, V. (2001b). Stretching of polymers in a random three-dimensional flow. *Physical review letters*, 86(5), 934.
- Hädrich, T. (2001). Measurement of turbulent spectra of particle-laden flows in a horizontal channel. *Experiments in Fluids*, 31(4), 401-408.
- Harris, F. J. (1978). On the use of windows for harmonic analysis with the discrete Fourier transform. *Proceedings of the IEEE*, 66(1), 51-83.
- Hassouna, F., Morlat-Therias, S., Mailhot, G., & Gardette, J. L. (2007). Influence of water on the photodegradation of poly (ethylene oxide). *Polymer Degradation and Stability*, 92(11), 2042-2050.
- Haward, S. J., Ober, T. J., Oliveira, M. S., Alves, M. A., & McKinley, G. H. (2012). Extensional rheology and elastic instabilities of a wormlike micellar solution in a microfluidic cross-slot device. *Soft Matter*, 8(2), 536-555.
- Hell, K. M. D., Balzereit, A., Diebold, U., & Bruhn, H. D. (1989). Importance of blood viscoelasticity in arteriosclerosis. *Angiology*, 40(6), 539-546.
- Ingremau, F., & Kellay, H. (2013). Stretching Polymers in Droplet-Pinch-Off Experiments. *Physical Review X*, 3(4), 041002.
- Japper-Jaafar, A., Escudier, M. P., & Poole, R. J. (2010). Laminar, transitional and turbulent annular flow of drag-reducing polymer solutions. *Journal of Non-Newtonian Fluid Mechanics*, 165(19), 1357-1372.
- Jun, Y., & Steinberg, V. (2011). Elastic turbulence in a curvilinear channel flow. *Physical Review E*, 84(5), 056325.
- Kameneva, M. V., Wu, Z. J., Uraysh, A., Repko, B., Litwak, K. N., Billiar, T. R., ... & Borovetz, H. S. (2004). Blood soluble drag-reducing polymers prevent

- lethality from hemorrhagic shock in acute animal experiments. *Biorheology*, 41(1), 53.
- Karami, H. R., & Mowla, D. (2012). Investigation of the effects of various parameters on pressure drop reduction in crude oil pipelines by drag reducing agents. *Journal of Non-Newtonian Fluid Mechanics*, 177, 37-45.
- Keegan, F. L., Escudier, M. P., Alves, M. A., & Poole, R. J. (2008, July). Viscoelastic Fluid Flow Through Gradual Contractions: Experiments And Simulations. In *THE XV INTERNATIONAL CONGRESS ON RHEOLOGY: The Society of Rheology 80th Annual Meeting* (Vol. 1027, No. 1, pp. 228-230). AIP Publishing.
- Kelly, M. S., & Santore, M. M. (1995). The role of a single end group in poly (ethylene oxide) adsorption on colloidal and film polystyrene: complimentary sedimentation and total internal reflectance fluorescence studies. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 96(1), 199-215.
- Keunings, R. (1986). On the high Weissenberg number problem. *Journal of Non-Newtonian Fluid Mechanics*, 20, 209-226.
- Khadom, A. A., & Abdul-Hadi, A. A. (2014). Performance of polyacrylamide as drag reduction polymer of crude petroleum flow. *Ain Shams Engineering Journal*, 5(3), 861-865.
- Kraichnan, R. H. (1967). Inertial ranges in two-dimensional turbulence. *Physics of Fluids*, 10(7), 1417.
- L'vov, V. S., Pomyalov, A., Procaccia, I., & Tiberkevich, V. (2004). Drag reduction by polymers in wall bounded turbulence. *Physical Review Letters*, 92(24), 244503.
- Lam, Y. C., Gan, H. Y., Nguyen, N. T., & Lie, H. (2009). Micromixer based on viscoelastic flow instability at low Reynolds number. *Biomicrofluidics*, 3(1), 014106.

- Lawler, J. V., Muller, S. J., Brown, R. A., & Armstrong, R. C. (1986). Laser Doppler velocimetry measurements of velocity fields and transitions in viscoelastic fluids. *Journal of Non-Newtonian Fluid Mechanics*, 20, 51-92.
- Leith, C. E. (1968). Diffusion approximation for two-dimensional turbulence. *Physics of Fluids*, 11(3), 671-672.
- Lesieur, M. (2008). Turbulence in fluids, Fluid Mechanics and its Applications (4th ed.). Dordrecht, NL: Springer Netherlands.
- Li, F. C., Kinoshita, H., Li, X. B., Oishi, M., Fujii, T., & Oshima, M. (2010). Creation of very-low-Reynolds-number chaotic fluid motions in microchannels using viscoelastic surfactant solution. *Experimental Thermal and Fluid Science*, 34(1), 20-27.
- Li, H., & Olsen, M. G. (2006a). MicroPIV measurements of turbulent flow in square microchannels with hydraulic diameters from 200 μ m to 640 μ m. *International Journal of Heat and Fluid Flow*, 27(1), 123-134.
- Li, H., & Olsen, M. G. (2006b). Examination of large-scale structures in turbulent microchannel flow. *Experiments in fluids*, 40(5), 733-743.
- Liu, Y., & Steinberg, V. (2010). Molecular sensor of elastic stress in a random flow. *EPL (Europhysics Letters)*, 90(4), 44002.
- Lumley, J. L. (1967). The structure of inhomogeneous turbulent flows. *Atmospheric Turbulence and Radio Wave Propagation* (eds. A. M. Yaglom and V. I. Tararsky), Nauka, Moscow, 166-178.
- Lumley, J. L. (1969). Drag reduction by additives. *Annual Review of Fluid Mechanics*, 1(1), 367-384.
- Malkin, A. Y., & Isayev, A. I. (2006). *Rheology. Concepts, Methods and Applications*. Toronto, CA: ChemTec Publishing.
- Mandelbrot, B., Hudson, R. L., & Grunwald, E. (2005). The (mis) behaviour of markets. *The Mathematical Intelligencer*, 27(3), 77-79.
- Marhefka, J. N., Zhao, R., Wu, Z., Velankar, S. S., Antaki, J. F., & Kameneva, M. V. (2009). Drag reducing polymers improve tissue perfusion via modification of the RBC traffic in microvessels. *Biorheology*, 46(4), 281.

- Meyer, K. E., Sørensen, J. N., Mikkelsen, R. F., & Watz, B. B. (2008). Frequency and flow structure detection in a cylindrical cavity using POD. *14th Int Symp on Applications of Laser Techniques to Fluid Mechanics*.
- Min, T., Yul Yoo, J., Choi, H., & Joseph, D. D. (2003). Drag reduction by polymer additives in a turbulent channel flow. *Journal of Fluid Mechanics*, 486, 213-238.
- Mininni, P. D. (2011). Scale Interactions in Magnetohydrodynamic Turbulence. *Annual Review of Fluid Mechanics*, 43, 377-397.
- Morlat, S., & Gardette, J. L. (2001). Phototransformation of water-soluble polymers. I: photo-and thermooxidation of poly (ethylene oxide) in solid state. *Polymer*, 42(14), 6071-6079.
- Munson, B. R., Young, D. F., & Okiishi, T. H. *Fundamentals of Fluid Mechanics* (4th ed.). 2002. New York, NY: John Wiley & Sons Inc
- Natrajan, V. K., & Christensen, K. T. (2007). Microscopic particle image velocimetry measurements of transition to turbulence in microscale capillaries. *Experiments in fluids*, 43(1), 1-16.
- Newell, A. C. & Rumpf, B. Wave Turbulence. (2011). *Annual Review of Fluid Mechanics*, 43, 59-78.
- Nguyen, H., & Boger, D. V. (1979). The kinematics and stability of die entry flows. *Journal of Non-Newtonian Fluid Mechanics*, 5, 353-368.
- Nigen, S., & Walters, K. (2002). Viscoelastic contraction flows: comparison of axisymmetric and planar configurations. *Journal of Non-Newtonian Fluid Mechanics*, 102(2), 343-359.
- Oliveira, M. S. N., Alves, M. A., Pinho, F. T., & McKinley, G. H. (2007). Viscous flow through microfabricated hyperbolic contractions. *Experiments in Fluids*, 43(2-3), 437-451.
- Oliveira, P. J. (2001). Method for time-dependent simulations of viscoelastic flows: vortex shedding behind cylinder. *Journal of Non-Newtonian Fluid Mechanics*, 101(1), 113-137.

- Oswald, P. (2009). *Rheophysics: the deformation and flow of matter*. Cambridge, UK: Cambridge University Press.
- Pathak, J. A., Ross, D., & Migler, K. B. (2004). Elastic flow instability, curved streamlines, and mixing in microfluidic flows. *Physics of Fluids (1994-present)*, 16(11), 4028-4034.
- Peng, X. F., Peterson, G. P., & Wang, B. X. (1994). Frictional flow characteristics of water flowing through rectangular microchannels. *Experimental Heat Transfer An International Journal*, 7(4), 249-264.
- Pipe, C. J., & Monkewitz, P. A. (2006). Vortex shedding in flows of dilute polymer solutions. *Journal of Non-Newtonian Fluid Mechanics*, 139(1), 54-67.
- Poole, R. J., Escudier, M. P., & Oliveira, P. J. (2005, December). Laminar flow of a viscoelastic shear-thinning liquid through a plane sudden expansion preceded by a gradual contraction. In *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences* (Vol. 461, No. 2064, pp. 3827-3845). The Royal Society.
- Pope, S. B. (2003). *Turbulent flows*. Cambridge, UK: Cambridge University Press.
- Pritchard, R. H., Lava, P., Debruyne, D., & Terentjev, E. M. (2013). Precise determination of the Poisson ratio in soft materials with 2D digital image correlation. *Soft Matter*, 9(26), 6037-6045.
- Procaccia, I., L'vov, V. S., & Benzi, R. (2008). Colloquium: Theory of drag reduction by polymers in wall-bounded turbulence. *Reviews of Modern Physics*, 80(1), 225.
- Raffel, M., Willert, C., Wereley, S., & Kompenhans, J. (2007). *Particle Image Velocimetry- A Practical Guide*. Springer Berlin Heidelberg (2nd ed.). Berlin, DE: Springer-Verlag Berlin Heidelberg.
- Rands, C., Webb, B. W., & Maynes, D. (2006). Characterization of transition to turbulence in microchannels. *International Journal of Heat and Mass Transfer*, 49(17), 2924-2930.
- Rebar, V. A., & Santore, M. M. (1996). A total internal reflectance fluorescence nanoscale probe of interfacial potential and ion screening in polyethylene

- oxide layers adsorbed onto silica. *Journal of Colloid and Interface Science*, 178(1), 29-41.
- Renardy, M. (1994). Some comments on the surface-tension driven break-up (or the lack of it) of viscoelastic jets. *Journal of Non-Newtonian Fluid Mechanics*, 51(1), 97-107.
- Renardy, M. (2014). The initial value problem for creeping flow of the upper convected Maxwell fluid at high Weissenberg number. *Mathematical Methods in the Applied Sciences*, 38(5), 959-965.
- Reynolds, O. (1883). An experimental investigation of the circumstances which determine whether the motion of water shall be direct or sinuous, and of the law of resistance in parallel channels. *Proceedings of the Royal Society of London*, 35(224-226), 84-99.
- Rodd, L. E., Cooper-White, J. J., Boger, D. V., & McKinley, G. H. (2007). Role of the elasticity number in the entry flow of dilute polymer solutions in micro-fabricated contraction geometries. *Journal of Non-Newtonian Fluid Mechanics*, 143(2), 170-191.
- Rodd, L. E., Scott, T. P., Boger, D. V., Cooper-White, J. J., & McKinley, G. H. (2005a). The inertio-elastic planar entry flow of low-viscosity elastic fluids in micro-fabricated geometries. *Journal of Non-Newtonian Fluid Mechanics*, 129(1), 1-22.
- Rodd, L. E., Scott, T. P., Cooper-White, J. J., and McKinley, G. H. (2005b). Capillary Break-up Rheometry of Low-Viscosity Elastic Fluids. *Applied Rheology*, 15(1), 12-27.
- Sawchuk, A. P., Unthank, J. L., & Dalsing, M. C. (1999). Drag reducing polymers may decrease atherosclerosis by increasing shear in areas normally exposed to low shear stress. *Journal of Vascular Surgery*, 30(4), 761-764.
- Sharp, K. V., & Adrian, R. J. (2004). Transition from laminar to turbulent flow in liquid filled microtubes. *Experiments in Fluids*, 36(5), 741-747.

- Simmons, R. M., Finer, J. T., Chu, S., & Spudich, J. A. (1996). Quantitative measurements of force and displacement using an optical trap. *Biophysical Journal*, 70(4), 1813.
- Sirovich, L. (1987). Turbulence and the dynamics of coherent structures. Part I: Coherent structures. *Quarterly of Applied Mathematics*, 45(3), 561-571.
- Soulages, J., Oliveira, M. S. N., Sousa, P. C., Alves, M. A., & McKinley, G. H. (2009). Investigating the stability of viscoelastic stagnation flows in T-shaped microchannels. *Journal of Non-Newtonian Fluid Mechanics*, 163(1), 9-24.
- Tabor, M., & De Gennes, P. G. (1986). A cascade theory of drag reduction. *EPL (Europhysics Letters)*, 2(7), 519.
- Tai, J., Gan, H. Y. and Lam, Y. C. (2011). Viscoelastic Turbulence for a 3-Stream Flow Through a Sudden Contraction-Expansion Microchannel. *The 2nd Conference on Advances in Microfluidics and Nanofluidics, and Asian-Pacific International Symposium on Lab on Chip*.
- Takeuchi, H. (2012). Demonstration test of energy conservation of central air conditioning system at the Sapporo city office building– Reduction of pump power by flow drag reduction using surfactant. *Synthesiology– English edition*, 4, 136-143.
- Tamano, S., Itoh, M., Inoue, T., Kato, K., & Yokota, K. (2009). Turbulence statistics and structures of drag-reducing turbulent boundary layer in homogeneous aqueous surfactant solutions. *Physics of Fluids (1994-present)*, 21(4), 045101.
- Tatsumi, K., Takeda, Y., Suga, K., & Nakabe, K. (2011). Turbulence characteristics and mixing performances of viscoelastic fluid flow in a serpentine microchannel. In *Journal of Physics: Conference Series* (Vol. 318, No. 9, p. 092020). IOP Publishing.
- Taylor, G. I. (1936). Correlation measurements in a turbulent flow through a pipe. *Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences*, 157(892), 537-546.

- Taylor, Z. J., Gurka, R., Kopp, G., & Liberzon, A. (2010). Long-duration time-resolved PIV to study unsteady aerodynamics. *Instrumentation and Measurement, IEEE Transactions on*, 59(12), 3262-3269.
- Thais, L., Gatski, T. B., & Mompean, G. (2013). Analysis of polymer drag reduction mechanisms from energy budgets. *International Journal of Heat and Fluid Flow*, 43, 52-61.
- Tirtaatmadja, V., McKinley, G. H., & Cooper-White, J. J. (2006). Drop formation and breakup of low viscosity elastic fluids: Effects of molecular weight and concentration. *Physics of Fluids (1994-present)*, 18(4), 043101.
- Toms, B. A. (1948, September). Some observations on the flow of linear polymer solutions through straight tubes at large Reynolds numbers. In *Proceedings of the 1st International Congress on Rheology* (Vol. 2, pp. 135-141). Scheveningen, Holland.
- Vétel, J., Garon, A., Pelletier, D., & Farinas, M. I. (2008). Asymmetry and transition to turbulence in a smooth axisymmetric constriction. *Journal of Fluid Mechanics*, 607, 351-386.
- Vinogradov, G. V., & Ivanova, L. I. (1968). Wall slippage and elastic turbulence of polymers in the rubbery state. *Rheologica Acta*, 7(3), 243-254.
- White, F. (2006). *Viscous fluid flow* (3rd ed.). New York, NY: McGraw-Hill Education.
- White, J. L., & Kondo, A. (1977). Flow patterns in polyethylene and polystyrene melts during extrusion through a die entry region: measurement and interpretation. *Journal of Non-Newtonian Fluid Mechanics*, 3(1), 41-64.
- White, S. A., & Baird, D. G. (1986). The importance of extensional flow properties on planar entry flow patterns of polymer melts. *Journal of Non-Newtonian Fluid Mechanics*, 20, 93-101.
- Willis, G. E., & Deardorff, J. W. (1976). On the use of Taylor's translation hypothesis for diffusion in the mixed layer. *Quarterly Journal of the Royal Meteorological Society*, 102(434), 817-822.

- Winfree, A. T. Electrical turbulence in three-dimensional heart muscle. (1994). *Science*, 266, 1003-1006.
- Wunderlich, A. M., Brunn, P. O., & Durst, F. (1988). Flow of dilute polyacrylamide solutions through a sudden planar contraction. *Journal of Non-Newtonian Fluid Mechanics*, 28(3), 267-285.
- Xi, L., & Graham, M. D. (2009). A mechanism for oscillatory instability in viscoelastic cross-slot flow. *Journal of Fluid Mechanics*, 622, 145-165.
- Xiong, Y. L., Bruneau, C. H., & Kellay, H. (2010). Drag enhancement and drag reduction in viscoelastic fluid flow around a cylinder. *EPL (Europhysics Letters)*, 91(6), 64001.
- Yesilata, B., Öztekin, A., & Neti, S. (1999). Instabilities in viscoelastic flow through an axisymmetric sudden contraction. *Journal of Non-Newtonian Fluid Mechanics*, 85(1), 35-62.
- Zeighami, R., Laser, D., Zhou, P., Asheghi, M., Devasenathipathy, S., Kenny, T., & Goodson, K. (2000). Experimental investigation of flow transition in microchannels using micron-resolution particle image velocimetry. *Thermal and Thermomechanical Phenomena in Electronic Systems, 2000. ITherm 2000. The Seventh Intersociety Conference on* (Vol. 2, pp. 148-153). IEEE.
- Zeynali, M. E., & Rabbii, A. (2002). Alkaline hydrolysis of polyacrylamide and study on poly (acrylamide-co-sodium acrylate) properties. *Iranian Polymer Journal*, 11, 269-275.
- Zhang, H. N., Li, F. C., Cao, Y., Kunugi, T., & Yu, B. (2013). Direct numerical simulation of elastic turbulence and its mixing-enhancement effect in a straight channel flow. *Chinese Physics B*, 22(2), 024703.
- Zhao, R., Marhefka, J. N., Antaki, J. F., & Kamenewa, M. V. (2010). Drag-reducing polymers diminish near-wall concentration of platelets in microchannel blood flow. *Biorheology*, 47(3-4), 193.
- Zhou, J., Adrian, R. J., Balachandar, S., & Kendall, T. M. (1999). Mechanisms for generating coherent packets of hairpin vortices in channel flow. *Journal of Fluid Mechanics*, 387, 353-396.

Blank Page

Appendix A

A.1 Effects of viscosity ratio in a 3-stream flow (A numerical approximation)

In this study, a 3-stream flow was employed, with the main-stream liquid being enveloped by two side-stream liquids. Details of the channel design are presented in Chapter 5. For each experimental run, the main-stream liquid constitutes 50% of the total flow rate (Q), while the two side-stream liquids constitute 25% each. Due to the shear thinning nature of the main-stream liquid, its viscosity varied with flow rate. To approximate the amount of space occupied by the main-stream liquid in the contraction for the calculation of the dimensionless parameters, numerical simulations in a straight channel (see Figure A1) was carried out using COMSOL Multiphysics, neglecting diffusion and elastic effects.

Figure A1. Numerical simulations of the straight channel flow. The inset shows the cross-section of the 3-stream flow located 0.5 mm downstream from the junction. (Red = main-stream, Blue = side-stream)

A straight channel (see Figure A1) with similar cross-sectional dimensions as the contraction was employed, neglecting both upstream and downstream flows. Inlet 1

shows the inflow of the main-stream liquid (red = higher viscosity), while inlets 2 and 3 show the inflows of the side-stream liquids (blue = lower viscosity). Rather fine meshing was employed with element sizes ranging between 3 μm to 9 μm . To approximate the initial viscosity, the average shear rate was calculated based on the assumption that the main-stream liquid occupies 50% of the space in the contraction. The corresponding viscosity was then obtained from the measured data (refer to Figure 4-10.) and input into the simulation. The 3-stream flow was modeled based on the Navier-Stokes equation (equation A1) and the continuity equation (equation A2).

$$\rho \left[\frac{\partial \mathbf{U}}{\partial t} + \mathbf{U} \cdot \nabla \mathbf{U} \right] = -\nabla P + \eta \nabla^2 \mathbf{U} \quad (\text{A1})$$

$$\nabla \cdot \mathbf{U} = 0 \quad (\text{A2})$$

Viscosity was modeled as a species which could be transported and diffused, based on the convection-diffusion equation (equation A3), for ease of computation.

$$D \nabla^2 \eta = \mathbf{U} \cdot \nabla \eta \quad (\text{A3})$$

where D is the diffusion coefficient.

Apart from the inlets and outlets, non-slip boundary conditions (Navier-Stokes equation) and insulation (Convection-Diffusion equation) were applied on all walls. The liquids were modeled as immiscible liquids, and the diffusion coefficient (D) was set at a small value of $10^{-12} \text{ m}^2/\text{s}$. Note that the liquids were in fact miscible, but this assumption was used to preserve the sharp interfaces between the 3 streams (see inset in Figure A1), for ease of approximating the area occupied.

The flow rates and test liquids were varied in this study, and the resulting viscosity ratios ($\mu_{\text{main-stream}} / \mu_{\text{side-stream}}$) ranged from 1.1 to 12.7. An increase in viscosity ratio was found to result in an increase in occupied space by the main-stream liquid. Figure A2 shows the relationship between viscosity ratio and cross-sectional area occupied by the main-stream liquid, based on the flow rates investigated in this study (see Chapter 6). Although the viscosity of the main-stream liquid was up to 12 times that of the side-stream liquid, the area occupied did not vary significantly i.e. 35 – 42%.

Figure A2. Relationship between viscosity ratio and occupied space (main-stream liquid)

Appendix B

B.1 Effects of timescales on the turbulent power spectra

To investigate the effect of the characteristic timescales (i.e. of each individual polymer molecule and that of the bulk liquid) on the turbulent flow field, the molecular weight and concentration of the main-stream liquid was varied. Three molecular weights at various concentrations were used, i.e. 0.6×10^6 g/mol, 2×10^6 g/mol and 8×10^6 g/mol respectively. The properties of the resulting test liquids have been included in Table 6-2 (see Section 6.9).

The experimental runs were completed by replacing the main-stream liquid, i.e. 1.0%-2M with the test liquids in Table 6-2, while the side-stream liquid i.e. 0.1%-2M remained unchanged. The main-stream liquid and side-stream liquid constituted to 50% and 25% of the total flow rate respectively. The experiments were conducted at $De \approx 250$, and the velocimetry data obtained from PIV at interrogation window A ($\delta = 0.3$ mm) was used to compute the power spectra.

Figure B-1 shows the power spectra obtained from the experiments, when the main-stream liquid was varied. Similar to the observations in Section 6.7, two power law decays which were observed. Based on the least squares method, linear trends were fitted to the 3 regions, and the respective drop-off frequencies have been included in Table 6-2 (refer to Section 6.9).

Figure B-1. Double energy cascade of various PEO main-stream liquids. a) 2%-0.6M b) 0.3%-2M
c) 1.1%-2M d) 0.1%-8M e) 0.3%-8M f) 0.5%-8M

Appendix C

C.1 Static measurements of phPAA-FITC molecules

To measure the size of the phPAA-FITC molecules under static conditions, an ellipse was fitted (see Section 7.3) along the perimeter of each molecule to obtain the major axis (β) and minor axis (α), which were subsequently used to calculate the aspect ratio (β/α). Table C-1 shows the size distribution of 1000 phPAA-FITC molecules, subcategorized into $100 \mu\text{m}^2$ incremental size ranges. The average dimension of the unstressed phPAA-FITC molecules is $32 \text{ by } 22 \mu\text{m}^2$, with an aspect ratio (β/α) of 1.5 which does not vary significantly across the various size ranges.

Table C-1 | Size distribution of 1000 phPAA-FITC molecules measured under static conditions. The number of counts for the distribution has been plotted in Figure 7-4. The average aspect ratio is ≈ 1.5 , and does not vary significantly across the size ranges.

Size range (μm^2)	Size (μm^2)		Counts	Major axes, β (μm)		Minor axes, α (μm)		Mean aspect ratio, β/α
	Average	Standard deviation		Average	Standard deviation	Average	Standard deviation	
101-200	167.70	21.42	27	18.91	3.01	15.10	1.62	1.54
201-300	256.65	29.35	130	23.65	3.85	15.03	2.11	1.63
301-400	349.75	29.91	171	26.84	4.05	17.82	2.33	1.56
401-500	449.49	29.33	169	29.72	3.46	20.38	1.99	1.49
501-600	550.89	28.73	173	32.20	3.66	22.77	2.27	1.44
601-700	645.30	25.88	129	35.14	4.09	24.54	2.42	1.46
701-800	746.45	25.83	85	37.34	5.27	26.72	2.52	1.43
801-900	850.86	33.45	35	40.09	4.84	28.45	2.83	1.44
901-1000	940.68	25.83	47	43.45	7.66	29.59	3.43	1.52
1001-1100	1047.29	31.44	17	46.98	5.32	29.83	3.09	1.61
1101-1200	1137.68	29.25	19	48.40	6.84	31.91	3.44	1.55
1201-1300	1240.00	25.42	11	49.75	12.07	35.29	5.63	1.52
1301-1400	1350.67	37.17	3	48.54	3.44	37.50	4.02	1.31
1401-1500	1436.50	20.45	8	57.40	10.43	35.59	4.89	1.67
> 1501	1802.50	363.68	8	59.96	8.33	41.92	5.57	1.44

Blank Page