

Entry Flow of Viscoelastic Fluids at Macro- and Micro-Scale

by

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Abstract

The main objective of the present thesis is to investigate laminar entry flows of viscoelastic fluids using simple geometries at the macro- and micro-scale. The channels used include in their design contraction and/or expansion regions in order to generate strong extensional flows, mimicking real industrial polymer processing units, in which several remarkably complex flow phenomena emerge due to elastic effects. Newtonian fluid flow is also investigated for comparison purposes and in order to validate the experimental techniques, by comparison with the results obtained from numerical simulations using a finite-volume code.

The experiments at macro-scale used *high*-viscosity polymeric fluids to promote low Reynolds number (Re) flow conditions, while the measurements using microfluidic devices were conducted with dilute polymeric solutions, with viscosities similar to that of water. Due to the small characteristic length scales of the microfluidic devices, low Re and high elasticity flow conditions are observed. In both cases, visualizations of the flow patterns were undertaken using streak line photography, detailed velocity field measurements were performed using particle image velocimetry and pressure drop measurements were also carried out in order to characterize the fluid flow behavior.

The microfluidic channels used in the experiments were produced in PDMS by soft-lithography using a SU-8 mold. In parallel, a new technique based on in-situ photopolymerization of a liquid phase, induced by UV light, was also developed. This technique has the great advantage of having reduced fabrication times using simple protocols that do not require clean room facilities, and all the expensive equipment associated with it. In this thesis, we lay down the foundations of this technique for future applications.

At the macro-scale, the investigation focused on the Newtonian and viscoelastic fluid flow through square-square sudden contractions and expansions. Various contraction and expansion ratios were tested ranging from 2.4 to 12. A Boger fluid and a shear-thinning viscoelastic fluid were used in the experiments, and comparison with the flow of a Newtonian fluid was also performed. For all fluids studied, the flow presents a complex three-dimensional character evidenced by a helical flow in the structure of the recirculations that are formed. Moreover, a rich variety of flow patterns arise for both viscoelastic fluids: Newtonian-like flow at low flow rates, lip vortex activity, vortex growth, diverging flow and unstable flow at high flow rates, due to onset of elastic instabilities. Interestingly, the contraction flow of the Boger fluid presents, for higher contraction ratios, atypical localized diverging streamlines near the re-entrant corner, a

novel type of flow pattern. The experimental results obtained for the 3D laminar flow of the Newtonian and viscoelastic fluids are compared with the numerical results and overall a good agreement is found, particularly for Newtonian fluid flow.

Studies at the micro-scale were also conducted with the ultimate goal of developing an efficient microfluidic rectifier, or microfluidic diode, which is able to operate efficiently even under creeping flow conditions. The viscoelastic behavior of the dilute polymer solutions used is enhanced due to the small length scales, and anisotropic flow resistance is obtained even for creeping flow conditions. The effect of the microchannel shape on the flow resistance anisotropy was investigated, and a microfluidic channel shaped as a succession of hyperbolic contractions/abrupt expansions presented the best performance in terms of flow diodicity, due to the strong extensional flow that is generated along the smooth hyperbolic contractions. In addition, the influence of the channel bounding walls on the efficiency of the microfluidic rectifier was investigated in order to optimize the performance of the proposed microfluidic device. The experimental results showed that increasing the distance between top and bottom planar walls leads to higher rectification effects, in great excess of other shapes reported in the literature in similar studies.

Finally, the extensional flow behavior of two viscoelastic fluids, which have a shear rheology similar to that of human blood, is analyzed. Despite the similar shear rheology, distinct flow behavior was observed for the two blood analogues solutions, demonstrating the key importance of extensional properties in the proper selection of blood analogue fluids.

Keywords: Viscoelasticity; Flow visualization; Particle image velocimetry; 3D contraction flow; 3D expansion flow; Microfluidic rectifier; Numerical simulations; Extensional flow; Elastic instabilities; Blood analogue.

Resumo

O principal objectivo da presente tese é investigar escoamentos de entrada de fluidos viscoelásticos em condições de regime laminar, usando geometrias simples à macro- e micro-escala. Os canais usados incluem regiões com contracções e/ou expansões de forma a gerar escoamentos fortemente extensionais, que se assemelham aos encontrados em unidades processuais reais da indústria de polímeros, nas quais diversos fenómenos, notavelmente complexos, ocorrem devido a efeitos elásticos. O escoamento de fluidos newtonianos é também investigado para referência e com o intuito de validar as técnicas experimentais, por comparação com os resultados obtidos por simulação numérica, usando um código de volumes-finitos.

Nas experiências à macro-escala usaram-se fluidos poliméricos de elevada viscosidade para promover condições de escoamento a baixos números de Reynolds (Re). As medições usando dispositivos microfluídicos foram conduzidas usando soluções poliméricas diluídas que apresentam viscosidades semelhantes à da água. Neste caso, devido às reduzidas dimensões características dos dispositivos microfluídicos, é possível promover escoamentos a baixos Re em que os efeitos elásticos são notórios, mesmo usando fluidos pouco viscosos e pouco elásticos. Em ambos os casos, realizaram-se visualizações dos padrões de escoamento usando uma técnica fotográfica de elevado tempo de exposição, mediu-se em detalhe o campo de velocidades usando velocimetria por imagem de partículas e mediram-se ainda as quedas de pressão de modo a caracterizar o escoamento.

Os microcanais usados nas experiências foram fabricados em PDMS por litografia suave usando um molde de SU-8. Em paralelo, foi também desenvolvida uma nova técnica baseada na fotopolimerização *in-situ* de uma fase líquida, induzida por luz ultravioleta. Esta técnica acarreta grandes vantagens pois tem tempos de fabricação reduzidos, usando protocolos simples que não necessitam da existência de uma sala limpa e todos os equipamentos dispendiosos associados à técnica standard. Nesta tese, estabelecem-se os fundamentos desta nova técnica para futuras aplicações.

A investigação à macro-escala focou-se no escoamento de fluidos newtonianos e viscoelásticos através de contracções e expansões abruptas quadrado-quadrado. Foram testadas várias razões de contracção e expansão desde 2.4 até 12. Nas experiências foi usado um fluido de Boger e um fluido viscoelástico reofluidificante, sendo os respectivos escoamentos comparados com o de um fluido newtoniano. Para todos os fluidos estudados, o escoamento apresenta um carácter tridimensional complexo, evidenciado por um escoamento com

recirculações em que o fluido se move numa trajectória helicoidal. Para além disso, foi observada uma grande variedade de padrões de escoamento para ambos os fluidos viscoelásticos: escoamento semelhante ao newtoniano para baixos caudais, vórtices de entrada, crescimento dos vórtices, escoamento divergente e escoamento não-estacionário a elevados caudais, devido ao aparecimento de instabilidades elásticas. Para elevadas razões de contracção, o escoamento do fluido de Boger apresenta ainda um novo tipo de padrão de escoamento em que as linhas de corrente, localizadas imediatamente a montante da contracção, são fortemente divergentes. Os resultados experimentais são comparados com os resultados numéricos, tendo-se verificado uma boa concordância entre ambos, particularmente para o escoamento do fluido newtoniano.

Os estudos à micro-escala foram conduzidos com o objectivo de desenvolver um rectificador (ou díodo) que opere eficientemente em condições de inércia desprezáveis. O comportamento viscoelástico das soluções poliméricas usadas é realçado devido às reduzidas dimensões características e obtém-se uma resistência anisotrópica do escoamento para ambos os sentidos, até mesmo quando a inércia do escoamento é desprezável. O efeito da forma do microcanal na anisotropia do escoamento foi investigado e um microcanal formado por uma sucessão de contracções hiperbólicas/expansões abruptas foi o que apresentou o melhor desempenho em termos de diodicidade, devido ao forte escoamento extensional gerado ao longo da contracção hiperbólica. A influência das paredes do canal na eficiência do rectificador microfluídico foi também analisada por forma a optimizar o funcionamento do dispositivo proposto. Os resultados experimentais mostram que um aumento da distância entre as paredes planas do topo e da base, leva ao aumento dos efeitos rectificativos, bem acima dos revelados para outras formas relatadas na literatura para estudos similares.

Finalmente, analisou-se o escoamento extensional de dois fluidos viscoelásticos que apresentam uma reologia de corte semelhante à do sangue humano. Apesar da reologia de corte dos dois fluidos análogos ser idêntica, os padrões de escoamento observados com as duas soluções são bem distintos, demonstrando a importância das propriedades extensionais para uma selecção adequada de fluidos análogos do sangue.

Palavras-chave: Viscoelasticidade; visualização do escoamento; velocimetria por imagem de partículas; escoamento 3D numa contracção; escoamento 3D numa expansão; rectificador microfluídico; simulações numéricas; escoamento extensional; instabilidades elásticas; análogos do sangue.

Résumé

Le principal objectif de cette thèse est d'étudier l'écoulement d'entrée de fluides viscoélastiques en régime laminaire, en utilisant des géométries simples à macro- et micro- échelle. Les canaux utilisés incluent des régions avec des contractions et /ou des expansions afin de déclencher un écoulement fortement extensionnel qui ressemble à celui observé dans les unités processuelles réelles de l'industrie de polymères, dans lesquelles divers phénomènes, très complexes, émergent à cause des effets élastiques. L'écoulement de fluides newtoniens est aussi étudié comme référence avec le but de valider les techniques expérimentales, en comparaison avec les résultats obtenus par simulation numérique, en utilisant un code de volumes finis.

Dans les expériences à macro échelle, des fluides polymériques de haute viscosité sont utilisés pour promouvoir des conditions d'écoulement à un nombre *Reynolds* (*Re*) réduit. Mesures avec l'aide de dispositifs micro-fluidiques ont été conduites, en utilisant des solutions polymériques diluées ayant des viscosités qui ressemblent à celle de l'eau. Dans ce cas, à cause des dimensions réduites caractéristiques des dispositifs micro-fluidiques, il est possible de développer un écoulement à bas *Re*, où les effets élastiques sont visibles même quand des fluides peu visqueux et peu élastiques sont utilisés. Dans les deux cas, des visualisations de modèle d'écoulement ont été réalisées, en utilisant une technique photographique avec un long temps d'exposition, puis des mesures détaillées du champ de vitesse ont été effectuées à l'aide de la vélocimétrie par images de particules ainsi que les chutes de pression de façon à caractériser l'écoulement.

Les micro-canaux utilisés dans les expériences ont été fabriqués en *PDMS* par lithographie douce, en utilisant un modèle de SU-8. Parallèlement, une nouvelle technique a été aussi développée, inspirée par la photopolymérisation *in-situ* d'une phase liquide, induite par la lumière ultraviolette. Cette technique a de grands avantages puisque la durée de fabrication est réduite, en utilisant des protocoles simples qui n'exigent pas e'utilisation d'une salle propre ni d'équipements coûteux associés à la technique standard. Dans cette thèse, les principes fondamentaux de ce nouveau procédé sont établis pour de futures applications.

À la macro-échelle, l'investigation s'est focalisée dans l'écoulement de fluides newtoniens et visco-élastiques par contraction et expansion abruptes carré-carré. Plusieurs raisons de contraction et expansion ont été testées du 2.4 jusqu'au 12. Pour cette expérience, un fluide de *Boger* et un fluide viscoélastique pseudoplastique sont utilisés et les respectifs écoulements ont été comparés à celui d'un fluide newtonien. Pour tous les fluides étudiés, l'écoulement présente un caractère complexe tridimensionnel, mis en évidence par un écoulement de re-circulation où

le fluide se déplace dans une structure hélicoidale. En plus, une grande variété de modèles d'écoulement pour les deux fluides visco-élastiques a été observée: écoulement semblable au newtonien pour de bas débits vortex d'entrée, croissance de vortex, écoulement divergent et écoulement non-stationnaire à débits élevés dû à l'apparition d'instabilités élastiques. En cas de fortes raisons de contraction, l'écoulement du fluide de *Boger* présente aussi un nouveau type de modèle d'égouttage où les lignes de courant localisées immédiatement à montant de la contraction, sont fortement divergentes. Les résultats expérimentaux sont comparés aux résultats numériques, en se vérifiant une bonne concordance entre les deux, particulièrement dans l'écoulement du fluide newtonien.

Les études à micro-échelle ont été conduites pour développer un rectificateur ou diode qui opère avec efficacité en condition d'inertie négligeable. Le comportement viscoélastique des solutions polymériques utilisées est mis en évidence grâce aux petites dimensions caractéristiques, et une résistance anisotrope de l'écoulement est obtenue, même quand l'inertie de l'écoulement est négligeable. L'effet de la forme du micro-canal dans l'anisotropie de l'écoulement a été étudié et un micro-canal formé par une succession de contractions hyperboliques/expansions abruptes a été celui qui a présenté la meilleure exécution en ce qui concerne la diodicité, dû à un fort écoulement extensionnel développé dans la contraction hyperbolique. L'influence des parois du canal dans l'efficacité du rectificateur microfluide a été aussi étudié de façon à optimiser le fonctionnement du dispositif proposé. Les résultats expérimentaux ont montré qu'en augmentant la distance entre les parois du plafond et de la base provoque l'accroissement des effets rectificatifs, en excès de ceux qui ont été révélés par d'autres procédés décrits dans des études similaires.

Finalement, l'écoulement extensionnel de deux fluides visco-élastiques qui présentent une réologie de coupe semblable à celle du sang humain a été observé. Bien que la réologie de coupe de deux fluides analogues soit identique, les procédés d'écoulement observés avec les deux solutions sont bien différents, montrant l'importance des propriétés extensionnelles pour une sélection adéquate de fluides analogues.

Mots-clés: Viscoélasticité ; visualisation de l'écoulement ; vélocimétrie par images de particules ; écoulement 3D dans une contraction ; écoulement 3D dans une expansion; rectificateur micro-fluidique ; simulations numériques; écoulement extensionnel ; instabilités élastiques ; analogue au sang.

*To my grandmother
who always hold my heart in her hands*

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Nomenclature

Roman

a	Parameter of the Carreau-Yasuda model
A	Cross section of the filament
AR	Aspect ratio
a_T	Shift factor
a, b	Parameters of the hyperbolic function
c	Concentration
c^*	Critical overlap concentration
C	Couette correction
CR	Contraction ratio
D_0	Diameter of the filament at time $t = 0$
$\mathbf{D}$	Rate of deformation tensor
D	Characteristic dimension
D	Diffusion coefficient
D_c	Length of a smooth contraction
De	Deborah number
Di_Q	Diodicity evaluated at a constant flow rate
$Di_{\Delta P}$	Diodicity evaluated at a constant pressure drop
D_p	Diameter of the plates
d_e	Effective image diameter
d_p	Diameter of the particle
d_s	Diffraction-limited spot size of a point source of light
El	Elasticity number
ER	Expansion ratio
f	Frequency of oscillation
f	Force applied
f	Diaphragm aperture of a digital camera
$f()$	Function
F	Normal force
G	Elastic modulus
G'	Loss modulus
G''	Storage modulus
G^*	Complex modulus
H	Half width of the square duct
h	Depth of the microchannel
h	Gap between plates
h_f	Final height
h_i	Initial height

i	Imaginary constant ($i^2 = -1$)
k	Index
k_B	Boltzmann's constant
K_H	Huggins constant
L	Length
L^*	Characteristic length
L_c	Contraction length
M	Magnification
M_w	Molecular weight
n	Refractive index
n	Power law index
N	Total number of images
N_1	First normal stress difference
N_2	Second normal stress difference
N_A	Avogadro's number
p	Pressure
Q	Volumetric flow rate
Q_m	Mass flow rate
R	Radius
R	Universal gas constant
Re	Reynolds number
s_0	Objective working distance
T	Absolute Temperature
T_0	Reference absolute temperature
t	Time
t_c	Critical time
t_{flow}	Characteristic time of the flow
Tr	Trouton ratio
U	Average axial velocity
u	Instantaneous velocity component in the x direction
u	Velocity vector
U_{down}	Average velocity in the downstream channel
U_{up}	Average velocity in the upstream channel
V	Particle visibility
v	Instantaneous velocity component in the y direction
V_{fr}	Fraction of particles
Wi	Weissenberg number
x, y, z	Cartesian coordinates
x_0	Reference pressure port location
x_1, x_2, x_3	Pressure ports locations
x_R	Vortex length

Greek

β	Solvent viscosity ratio
β	Cut-off level of the edges of the particle image
δ	Phase angle
ΔH	Activation energy
Δp	Pressure drop
Δp_{extra}	Extra pressure drop
Δp_{total}	Total pressure drop
Δt	Time interval
δt	Time step
$\Delta x, \Delta y$	Total particle displacement in the x and y directions
$\Delta x, \Delta y, \Delta z$	Cell dimensions
δz	Depth of field
δz_m	Total measurement depth
ε	Extensibility parameter
$\dot{\varepsilon}$	Strain rate
ε_H	Hencky strain
ε_x	Relative error due to Brownian motion in the x -direction
ε_y	Relative error due to Brownian motion in the y -direction
$\dot{\gamma}$	Shear rate
γ_0	Strain amplitude
Γ	Time parameter
η	Shear viscosity
η_0	Shear viscosity at zero shear-rate
η_E	Extensional viscosity
η_p	Polymer shear viscosity
η_s	Solvent viscosity
η^*	Complex dynamic viscosity
$[\eta]$	Intrinsic viscosity
λ	Relaxation time
λ	Light wavelength
λ_0	Light wavelength in vacuum
λ_{Zimm}	Zimm relaxation time
Λ	Aspect ratio
Λ	Time parameter of the Carreau and Carreau-Yasuda models
Λ_i	Initial aspect ratio
Λ_f	Final aspect ratio
μ	Dynamic viscosity
ν	Solvent quality exponent
Ψ_1	First normal stress coefficient
Ψ_2	Second normal stress coefficient
ρ	Density

ρ_0	Density at the reference temperature
σ	Surface tension
σ_x	Displacement due to Brownian motion in the x-direction
σ_y	Displacement due to Brownian motion in the y-direction
τ_{xx}	Normal stress
τ_{yy}	Normal stress
τ_{xy}	Shear stress
τ_w	Wall shear stress
τ	Extra-stress tensor
τ_p	Polymeric solute contribution to the extra-stress tensor
τ_s	Newtonian solvent contribution to the extra-stress tensor
θ	Angle
$\mathfrak{S}$	Torque
$\mathfrak{S}_{\min}$	Rheometer minimum resolvable torque
Ω	Angular velocity
ω	Spin velocity

Acronyms

2D	Two-dimensional
3D	Three-dimensional
BP	Band pass
CaBER	Capillary break-up extensional rheometer
CAC	Critical aggregation concentration
CAD	Computer Aided Design
CCD	Charge Coupled Device
CUBISTA	Convergent and Universally Bounded Interpolation Scheme for Treatment of Advection
CV	Control volumes
DC	Direct current
DMPA	2,2-dimethoxy-2-phenyl-acetophenone
DOF	Depth of field
dpi	Dots per inch
Em	Emission
Ex	Excitation
FENE	Finite Extensible Nonlinear Elastic model
IBA	Isobornyl acrylate
LDV	Laser Doppler velocimetry
LP	Low pass filter
MHS	Mark-Houwink-Sakurada
NA	Numerical aperture
NC	Number of cells
Nd-YAG	Neodymium-doped yttrium-aluminum-garnet
NMP	No-moving parts
PAA	Polyacrylamide

PDMS	Polydimethylsiloxane
PEO	Polyethylene oxide
PIV	Particle image velocimetry
PTT	Phan-Thien and Tanner
PVC	Polyvinyl chloride
RBC	Red blood cells
SAOS	Small-amplitude oscillatory shear
SDS	Sodium dodecyl sulphate
SEM	Scanning electron microscopy
SU-8	Designation of epoxi-bored photoresist
TeGDMA	Tetraethylene glycol dimethacrylate
UCM	Upper convected Maxwell model
URP	Ultra-rapid prototyping
UV	Ultraviolet
WBC	White blood cells
XG	Xanthan gum
μ PIV	Micro-particle image velocimetry
μ TAS	Micro total analysis systems

Subscripts

1, 2	Upstream and downstream square channel
<i>Backward</i>	Backward direction
<i>c</i>	Centerline
<i>crit</i>	Critical
<i>d</i>	Downstream
<i>f</i>	Final
<i>fd</i>	Fully developed
<i>Forward</i>	Forward direction
<i>i</i>	Index
<i>i</i>	Initial
<i>k</i>	Mode index of the linear viscoelastic spectra for the Boger fluid
<i>max</i>	Maximum
<i>min</i>	Minimum
<i>p</i>	Polymer
<i>s</i>	Solvent
<i>u</i>	Upstream
<i>x, y, z</i>	Cartesian directions

Superscripts

T	Transpose

INTRODUCTION

1.1. Background

1.1.1. Newtonian vs. non-Newtonian fluids

Understanding and analyzing the flow of fluids has attracted much attention due to its importance to human life. Effectively, fluid flow occurs in a multiplicity of natural and technical fields. Water flowing in a river or blood flowing in the body of living beings are basic examples of spontaneous fluid flows in nature. In addition, fluid flows take place in a wide range of technological processes with relevance to industry, which exist in order to provide many products essential to human life.

A fluid is defined as a substance that moves and deforms continuously as long as a shear stress is applied (White, 2008). On the opposite extreme, it is possible to define another class of materials, known as (elastic) solids, which may experience small deformations when exposed to a shear stress, but will not flow, i.e. a solid material will not deform continuously. These two concepts are the basis of Newton's law of viscosity and Hookean elasticity theory, respectively, developed in the 17th century. Later, a hybrid concept, which comprises the elastic and viscous behaviors of a material was introduced, the so-called viscoelastic material.

For a Newtonian fluid, the applied shear stress (τ_{xy}) is proportional to the resulting rate of deformation ($\dot{\gamma}$), in accordance with Newton's law of viscosity:

$$\tau_{xy} = \mu \dot{\gamma}. \quad (1.1)$$

On the other hand, for other fluids, the relation between shear stress and rate of deformation is nonlinear. These fluids are called non-Newtonian, and typical examples include polymeric solutions, melts and colloidal suspensions. Non-Newtonian fluids may be purely viscous or viscoelastic, also called memory fluids. For the former, the stress does not depend on the deformation history of the flow, being a function of the instantaneous deformation rate. For viscoelastic fluids the stress depends not only on the instantaneous deformation rate, but also on the kinematic history of the deformation. A viscoelastic fluid exhibits a combination of properties of a viscous liquid and an elastic solid, i.e. reacts to deformations over a short time scale as an elastic solid and flows as a liquid over long time scales (Denn, 2004).

Purely viscous non-Newtonian fluids, or generalized Newtonian fluids (GNF), can be classified in three categories according to the dependence of the shear stress on the rate of deformation (White, 2008): (i) pseudoplastic fluid (or shear-thinning fluid), in which the shear viscosity decreases as the shear rate increases; (ii) dilatant fluid (or shear thickening fluid) in which the shear viscosity increases with shear-rate; (iii) Bingham plastic, in which the fluid behaves as a solid until a minimum shear stress (known as the yield stress) is applied. The generic rheological behavior of the various types of GNF fluids is illustrated in Figure 1.1, where the behavior of a Newtonian fluid is also sketched. We note that for a GNF fluid, Newton's law of viscosity can be generalized to the following constitutive equation:

$$\tau_{xy} = \eta(\dot{\gamma}) \dot{\gamma}, \quad (1.2)$$

where the shear viscosity η is a function of the shear rate.

For viscoelastic fluids in a tangential (Couette) flow between two parallel plates with one of them being translated along the x -direction at constant velocity, besides the shear stress (τ_{xy}) that needs to be applied to promote the flow, the first normal stress difference, $N_1 = \tau_{xx} - \tau_{yy}$, and the second normal stress difference, $N_2 = \tau_{yy} - \tau_{zz}$, in which τ_{xx} , τ_{yy} and τ_{zz} , are the normal stresses, are of rheological importance. It is often more convenient to use the normal stress coefficients, Ψ_1 and Ψ_2 , defined as $\Psi_1 = N_1 / \dot{\gamma}^2$ and $\Psi_2 = N_2 / \dot{\gamma}^2$, respectively. In contrast, for Newtonian

fluids, the normal stresses and consequently the normal stress differences are null in a Couette flow, showing an important difference between GNF and viscoelastic fluids. A simple example illustrating the different behavior of Newtonian and viscoelastic fluids occurs when a rod rotates in a reservoir containing a liquid. If the liquid is a Newtonian fluid, the free surface near the rotating rod depresses, whereas if the liquid is a viscoelastic fluid it tends to climb up the rod. This latter phenomenon is known as the Weissenberg effect and arises due to the existence of normal stresses in the viscoelastic flow.

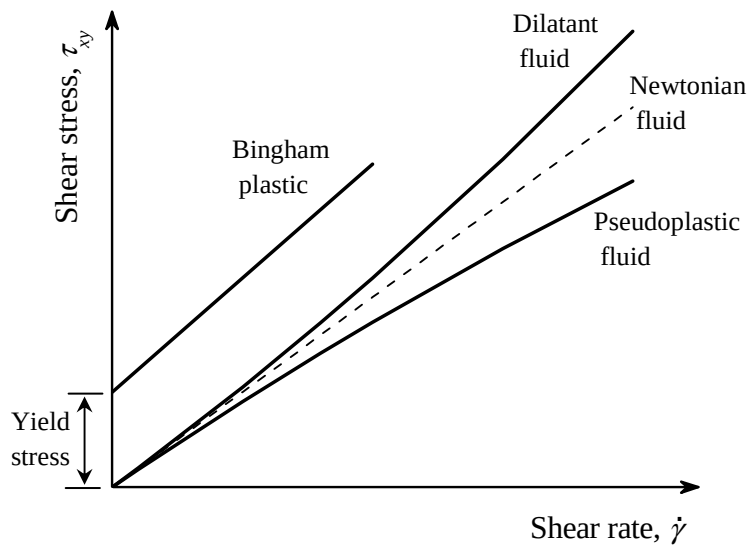


Figure 1.1. Shear stress (τ_{xy}) as a function of the shear-rate ($\dot{\gamma}$) for various purely viscous materials: a Newtonian fluid; an ideal Bingham plastic; a dilatant or shear-thickening fluid; a pseudoplastic or shear-thinning fluid.

Newtonian fluid flow behavior was thoroughly investigated in the past, and the resultant flow phenomena are effectively well understood, at least for laminar flow conditions. On the other hand, due to the complex nature of non-Newtonian fluids, and in particular those exhibiting viscoelasticity, much research efforts are still needed in order to obtain further insights regarding their flow behavior. Indeed, non-Newtonian fluid flow is of great importance, as non-Newtonian fluids are ubiquitous in nature (e.g. saliva, blood) and in common use products (e.g. paints, shampoo). Furthermore, the flow of viscoelastic fluids is particularly important since it is used in a broad range of industrial applications. Typical examples include the production of plastics, rubbers, paints or contact adhesives, applications with biological fluids as well as in the performance of lubricants and processing of foodstuffs. In the process units of such industries, flow through contractions and/or expansion geometries are frequent. Hence, it is essential to

understand the nonlinear viscoelastic fluid flow in such type of geometries, in order to optimize the flow behavior (e.g. minimizing the occurrence of flow instabilities that often occur with viscoelastic fluids) and consequently the product quality, throughput and production profits.

The goal of this thesis is to investigate the non-linear flow of polymeric fluids, with viscoelastic behavior, within complex geometries containing contractions and/or expansions. In this so-called entry-flow problem, a strong extensional flow is generated in the contraction/expansion region, and viscoelastic behavior is enhanced. In contrast, near the channel walls the flow is shear dominated. The simultaneous presence of extensional- and shear-dominated regions of the flow, can lead to a rich and complex variety of flow patterns, difficult to predict due to the complex nature of the fluid rheology (Boger and Walters, 1993). Furthermore, the diverse phenomena resulting from viscoelastic fluid flow are markedly different from those observed for Newtonian fluid flow, due to the nonlinear characteristics of the former, which makes the investigation of the viscoelastic fluid flow both interesting and challenging.

1.1.2. Dimensionless numbers

In order to characterize the flow and establish different flow regimes, the use of dimensionless numbers is very useful. These dimensionless groups assess the importance of different competing effects, expressing their relative importance (and form a sort of parameter space that should be explored for a given flow). In the various chapters of this thesis, several dimensionless numbers will be used in the characterization of the flow, and some of those dimensionless parameters are reviewed in this section, emphasizing the competing effects that are at play for each case.

i) Reynolds number

The Reynolds number is the most cited dimensionless number in flows of Newtonian fluids, either at macro- or micro-scales. The Reynolds number, Re , is usually defined as:

$$Re = \frac{\rho U D}{\eta}, \quad (1.3)$$

where ρ is the density of the fluid, U is a characteristic velocity, η is the shear viscosity of the fluid, and D is a characteristic dimension of the geometry. This dimensionless number was

introduced by Stokes in 1851 and made popular by Osborne Reynolds (1883) with the intention of explaining the experimental flow patterns obtained in the Newtonian fluid flow in a pipe. This dimensionless number expresses the competing effects between inertial forces ($\rho U^2/L$) and viscous forces ($\eta U/D^2$) per unit volume. If the value of Re is very small (say, $Re \ll 1$), inertial effects can be ignored and the viscous fluid flow is called creeping (or inertialess) flow; on the other hand, if Re is high, inertial effects are dominant and the nonlinear (inertial) terms in the momentum equation prevails, eventually leading to turbulent flow. The flow of non-Newtonian fluids typically occurs at low Reynolds numbers, due to their usual high viscosities.

ii) *Deborah number*

This dimensionless group has great importance in rheology and was introduced by Reiner (1964) following the idea that everything flows, depending on the time scale. Reiner was inspired by the song of Deborah presented in the English version of the Old Testament, where the prophetess Deborah sang: “The mountains flowed before the Lord” (Judges 5:1-5). Using an analogy, the Deborah number (De) was defined by Reiner (1964) as the ratio between the relaxation time of the fluid and the time of observation. Implicit in Deborah’s song is the concept that the rheological response of a given material depends on the time scale of observation: mountains eventually flow on geological time scales ($De \ll 1$), but are rigid bodies for human time scales ($De \gg 1$) (Squires and Quake, 2005).

From a practical point of view, De relates the characteristic time of the fluid, λ , with a characteristic time of the flow, t_{flow} (Bird *et al.*, 1987), which can be estimated as $t_{flow} = D/U$, resulting in the following definition used throughout this thesis:

$$De = \frac{\lambda U}{D}. \quad (1.4)$$

As already stated, if a shear stress is applied on a viscoelastic fluid over a period of time, and then is rapidly removed, the fluid returns to the original state (shear free) gradually, in contrast to a Newtonian fluid, which returns immediately to the initial state of null shear stress. The time necessary for the viscoelastic fluid to revert to the initial state is proportional to the relaxation time, λ .

Two limiting cases must be taken into consideration: first, when the De is very small (say, $De \rightarrow 0$), elastic effects are negligible and the viscoelastic fluid has a Newtonian-like behavior;

second, when De is very large ($De \rightarrow \infty$), elastic effects dominate and the fluid behavior approaches that of an Hookean solid. In practical terms, for $De > 1$ elastic effects start to dominate, and the flow behavior has elastic and viscous components.

iii) Weissenberg number

The Weissenberg number is another important dimensionless group used in non-Newtonian fluid mechanics, especially in studies of viscoelastic fluid flow through contractions, in which there is an important elongational component of the flow. In this thesis this dimensionless group is defined as:

$$Wi = \lambda \dot{\epsilon}, \quad (1.5)$$

where $\dot{\epsilon}$ is the strain rate. For contraction flows, the strain rate is often estimated based on the average velocity of the downstream (U_{down}) and upstream (U_{up}) channels, $\dot{\epsilon} = (U_{down} - U_{up}) / D_c$, in which the length D_c is a characteristic dimension of the contraction (e.g. the length of the contraction for a smooth contraction).

iv) Elasticity number

The elasticity number relates the relative importance of elastic to inertial effects, and is usually defined as the ratio between the Deborah and Reynolds numbers:

$$El = \frac{De}{Re} = \frac{\lambda \eta}{\rho D^2}. \quad (1.6)$$

The elasticity number depends on the dimension of the device and the material functions of the fluid. For Boger fluids, it has the advantage of being independent of the flow kinematics (namely flow rate and velocity).

The small length scales typical of microfluidics allow for high-elasticity flows to be investigated more easily, and consequently regions in the Re - De parameter space so far unattainable at the macro-scale are easily attained in microfluidic flows (Rodd *et al.*, 2005; Squires and Quake, 2005).

1.1.3. Macro- vs. micro-scale entry flow

The flow of viscoelastic fluids past a contraction has been extensively investigated with the goal to better understand polymer processing operations. The complex flow generated in such simple geometrical arrangement led to its selection, during the 5th International Workshop on Numerical Methods in Non-Newtonian Flows, that took place in Lake Arrowhead, U.S.A. (Hassager, 1988), as one of the most important benchmark flow problems in computational rheology, even nowadays. The 4:1 planar contraction flow of an upper-convected Maxwell (UCM) fluid has been the primary selection for benchmark purposes, and the majority of studies published so far are for this configuration using either UCM or Oldroyd-B fluids. Therefore, two-dimensional (2D) geometrical arrangements have been much more investigated than the corresponding three-dimensional (3D) counterpart, due to the simplicity of the geometry and also because of the less demanding computational power necessary for accurate predictions of 2D flows. Given the lack of studies involving 3D configurations, it is very important and promising to study those flows, not only to understand the flow behavior in complex 3D flows, but also to provide accurate data to validate 3D numerical codes, providing useful benchmark data. Typically, non-Newtonian fluid flow in polymer processing units occurs in complex 3D geometries, at low Reynolds numbers (e.g. extrusion processes and injection molding). Therefore, in order to better understand and predict accurately the flow in such processes, with industrial relevance, it is essential to study polymeric fluid flow behavior as well as the influence of the rheology of different polymeric solutions on the fluid flow within fluidic devices, in which the Reynolds numbers are small and consequently inertial effects are small or even negligible. It is worth noticing that in such inertialess flow conditions, elastic effects are the driving force for the rich and complex phenomena that arise in viscoelastic fluid flow.

When viscoelastic fluid flow occurs in macro-scale devices, in which the characteristic dimension is higher than (typically) 1 cm, small Reynolds numbers are only achieved when fluids with relatively high shear viscosities are used. Alternatively, if the viscoelastic fluid flow occurs in microfluidic devices, with characteristic dimensions ranging from about 1 μm up to $10^3 \mu\text{m}$, the Reynolds numbers are typically small, even for low viscosity fluids, and creeping flow conditions are easily attained, as illustrated schematically in Figure 1.2. As a result, the nonlinear convection term in the Navier-Stokes equations can be neglected, while the deformations of the polymer molecules, enhanced by the small dimensions of the geometry, bring elastic nonlinearities into play, with the corresponding onset of elastic instabilities and

ultimately leading to elastic turbulence (Groisman and Steinberg, 2000; Burghlea *et al.*, 2007; Fardin *et al.*, 2010).

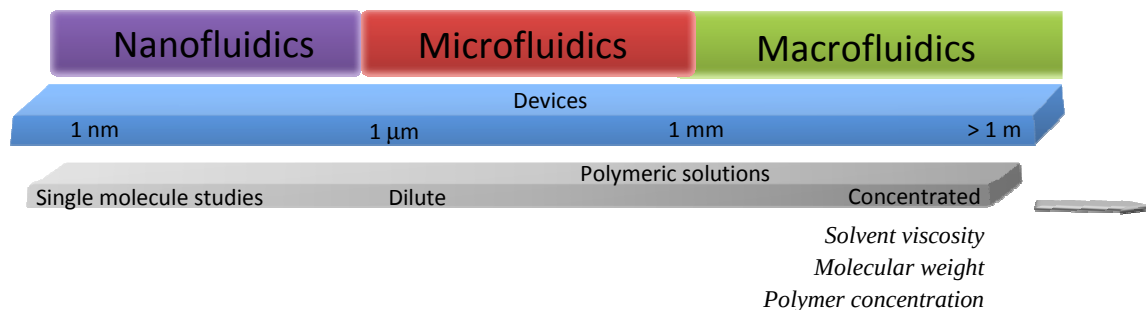


Figure 1.2. Comparison between nano-, micro- and macro-scale flows in terms of typical devices length scales and concentration of the polymeric fluids usually investigated.

The development of microfluidic devices to transport and manipulate small amounts of fluids (at the micro- or nano-liter scale) have stimulated great interest, not only in classical fluid mechanics, but particularly for rheologists in the study of non-Newtonian fluid flows (Pipe and McKinley, 2009), which is the general scope of the present thesis, and also in the development of integrated micro-systems for many other applications namely in biology and chemistry (Whitesides, 2006). Scaling down from macro-systems to micro lab-on-a-chip systems, volume forces become considerably less relevant and surface forces become dominant as the characteristic dimensions are progressively reduced (Bruus, 2008), leading to flow phenomena usually not observed at the macro-scale.

In the area of microfluidics, many advances were accomplished in the last two decades and the concept of *lab-on-a-chip* has expanded to diverse areas of application (Nguyen and Wereley, 2006). As happened with microelectronics in the 1960's, which became extremely successful as the size of integrated circuits (and consequently of electronic equipment) was decreased and became more powerful, in the area of microfluidics a similar goal is envisioned with various functions incorporated into one small device so that an autonomous and portable workstation is produced (Thorsen *et al.*, 2002). Several components for fluid manipulation (e.g. pumps, valves) or separation and detection techniques (e.g. electrophoresis, chromatography, electrochemical detection) have been improved and optimized in order to be employed in those micro-systems. Currently, microfluidic technology offers a variety of promising tools in a broad range of applications, especially in life sciences industries as for example in pharmaceutical industry for

drug delivery (e.g. Hilt and Peppas, 2005; Chen *et al.*, 2009) or in medicine for diagnostic screening devices (e.g. Polla *et al.*, 2000; Ziober *et al.*, 2007) or even in biology for separation of biological molecules (e.g. Tegenfeldt *et al.*, 2004; Dittrich and Manz, 2005; Yi *et al.*, 2006), to mention only a few examples. Furthermore, the amount of liquids used in microfluidic devices is very small, leading to a significant reduction in the quantity of reagents and waste when compared with macro-scale devices and the analysis time is reduced as well. Despite the amazing progress seen in the last two decades, there is still a lack of commercial developments of microfluidic technologies (Whitesides, 2006), and the quest for the “killer application” of microfluidics is still underway (Becker, 2010).

In microfluidic devices, adding small amounts of macromolecules to a Newtonian solvent is sufficient to generate elastic effects. Effectively, dilute polymeric solutions with relatively small relaxation times ($O(1\text{ ms})$) can experience high shear rates (or deformations in small time scales) and therefore high elasticity flow conditions are achieved, i.e. high De , Wi or El , when flowing in microchannels. Furthermore, when high elasticity flow conditions are reached under inertialess flow conditions, it is possible to more clearly identify and analyze the complex fluid flow behavior due to elasticity. In fact, viscoelastic fluid flow presents various interesting characteristics as documented by Boger and Walters (1993). In entry-flows, the transitions experienced by the fluid lead to different flow phenomena and instabilities, which can also occur in polymer process operations, namely the appearance of lip vortices near the re-entrant corner as well as recirculating vortices upstream of the contraction, the onset of diverging streamlines and time-dependent flow instabilities (Hulsén, 1993; Larson *et al.*, 1994; Grillet *et al.*, 2000; Owens and Phillips, 2002; Alves and Poole, 2007; Afonso *et al.*, 2010). The critical flow condition for the onset of elastic instabilities depends on the rheological properties of the fluid and the geometrical shape of the channel. When the elastic instabilities arise under creeping flow conditions, above a critical Deborah number, they are called as purely-elastic instabilities. Such type of instabilities is investigated in the present thesis for entry flows.

Viscoelastic flow field has been characterized in several investigations using different techniques. Visualizations of the flow patterns is perhaps the most employed in Newtonian and non-Newtonian fluid flow (Bagley and Birks, 1960; Van Dyke, 1982; Walters and Jones, 1988; Boger and Walters, 1993; Nigen and Walters, 2002; Rodd *et al.*, 2005). Nevertheless, there are other optical techniques, which allow a quantitative analysis of the flow field. Experiments using laser Doppler velocimetry (LDV) (e.g. Mackley and Moore, 1986; Compton and Eaton,

1996; Wassner *et al.*, 1999) and particle image velocimetry (PIV) (e.g. Adrian, 1991; Santiago *et al.*, 1998; Meinhart and Zhang, 2000) have been used frequently in order to measure the velocity field of the flow. In addition, birefringence measurements have also been used to probe the stress field (e.g. Geffroy and Leal, 1990; Quinzani *et al.*, 1994; Li *et al.*, 2000). Moreover, pressure drop measurements are also useful for the flow behavior analysis (e.g. Eisenbrand and Goddard, 1982; Binding *et al.*, 2006).

1.2. Road map for this dissertation

This dissertation compiles the experimental and numerical research work carried out with the main goal of investigating Newtonian and viscoelastic fluid flow through 3D entry flow geometries, which promote strong extensional flows at low Reynolds numbers, including the limit of creeping flow conditions. Different studies, as schematically illustrated in Figure 1.3, were carried out at the macro- and micro-scales with the same common denominator: rheology and viscoelasticity. In this section, we describe in detail the specific objectives and motivation of this work.

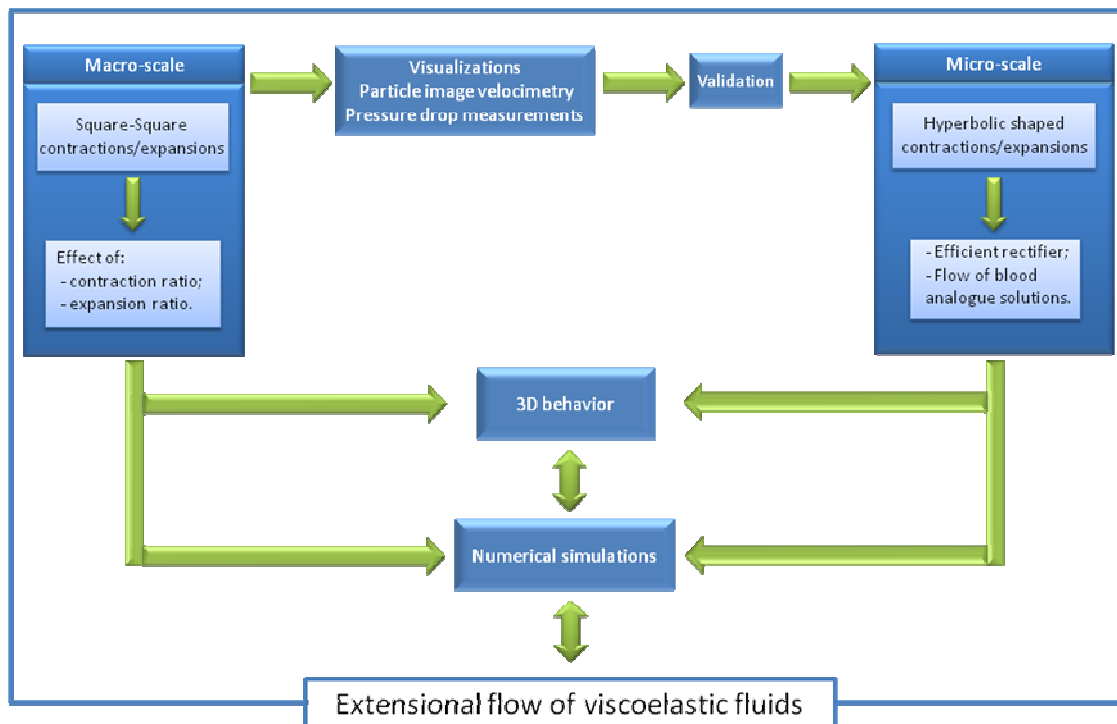


Figure 1.3. Illustration of the objectives of this thesis and the links between different studies.

The miniaturization of flow devices allows for strong viscoelastic effects to be achieved as a consequence of the high deformations rates attained due to the small lengthscales of microfluidic

devices. Experiments in microfluidic flows had never been performed in the host research group laboratory at the beginning of this investigation as the microfluidics laboratory was being set-up. The decision at the initial phase of the work was therefore to put the experimental techniques, and in particular the new PIV system, into operation first at the macro-scale, and then move to the micro-scale. In spite of the obvious differences between the two length-scales, the flow phenomena observed at the macro- and micro-scales share some similarities and can be used as a complement to each other. Therefore, the objectives defined for the macro-scale experiments are linked with those outlined for microfluidics experiments. Furthermore, since the experimental techniques employed at both scales are similar, the macro-scale experiments served also to test the equipments and validate the results prior to performing the micro-scale experiments.

At the macro-scale the investigation focused on the viscoelastic fluid flow through abrupt contractions and expansions in three-dimensional (3D) channels with a square cross-section. Contractions or expansions are excellent platforms to promote extensional flow. In addition, the choice of this geometrical arrangement was motivated by the highly three-dimensional character achieved in flows through this type of geometries, and was further triggered by the lack of experimental and numerical works concerning it, besides a few preliminary studies undertaken in the host research group (Alves *et al.*, 2005; Alves *et al.*, 2008) and elsewhere (Sirakov *et al.*, 2005). Also, since the flow through microfluidic devices are also 3D in nature, due to the typical low aspect ratios of these devices, the use of a 3D arrangement in the macro-scale experiments was natural, allowing for complementary studies at the macro- and micro-scales to be carried out. Furthermore, the investigation of 3D viscoelastic fluid flows is of great relevance in order to provide accurate benchmark data in complex 3D flows, useful for validating numerical codes.

An additional objective established for work at the macro-scale, was to study the effect of the contraction/expansion ratio on the fluid flow behavior. The contraction ratio (CR) is defined as the ratio between the side lengths of the upstream and downstream channels and the expansion ratio (ER) is defined as the ratio between downstream and upstream side lengths of the square channels, so that CR and ER are always greater than one. Furthermore, two viscoelastic fluids with different rheological behavior were employed: a fluid with the shear viscosity nearly independent of the shear-rate, known as a Boger fluid (Owens and Phillips, 2002) and a shear-thinning fluid in which the shear viscosity decreases with the shear-rate. This choice was done in order to probe independently the effects of elasticity and shear-thinning viscosity upon the complex flow behavior.

The flow of dilute polymeric solutions was investigated at the micro-scale with two practical applications in mind: (i) development of a microfluidic rectifier, or fluidic diode, in which the flow resistance varies with the direction of the flow (thus acting like the equivalent of a diode); (ii) test the rheological behavior of different blood analogue solutions under strong extensional flow using hyperbolic contractions.

For application (i), we designed and modified the shape of microfluidic devices in order to achieve large flow resistance anisotropy, even under creeping flow conditions, thus acting as an efficient microfluidic rectifier. This investigation was motivated by the works of Groisman and Quake (2004) and Nguyen *et al.* (2008), which explore the nonlinear properties of viscoelastic fluids under strong extensional flows. In spite of the viscoelastic fluid flow through abrupt contractions being sufficient to promote a strong extensional component of the flow, we found that using a series of hyperbolic contractions allowed to achieve a nearly constant extensional rate at the centerline of the channel (Oliveira *et al.*, 2007), leading to a much higher flow anisotropy as compared with previous studies (Groisman and Quake, 2004; Nguyen *et al.*, 2008). Dilute and semi-dilute solutions were used in the experiments so that a wide range of polymer concentrations was covered.

Application (ii) is related with the investigation of the flow of viscoelastic fluids with shear rheology similar to human blood, known as blood analogue solutions. To probe the elastic behavior of different blood analogues proposed in the literature (e.g. Liepsch *et al.*, 1991; Thurston, 1996; Vlastos *et al.*, 1997), we use a hyperbolic shaped contraction/expansion geometry capable of imposing a strong extensional flow. Blood analogues are often used for in vitro experiments when the use of whole blood is not practical and this work showed that extensional properties should also be taken into account when selecting a blood analogue fluid, in particular when considering conditions equivalent to those found in microcirculation.

In all cases, both at macro- and micro-scales, the experimental results are compared with numerical predictions. The numerical code used in the simulations has been developed in the host research group for more than a decade (Oliveira *et al.*, 1998; Alves *et al.*, 2003; Afonso *et al.*, 2009) and is applicable to Newtonian and non-Newtonian fluid flow. This numerical code is based on the finite-volume method and in this thesis, no work development was undertaken and the calculations were performed with the aim of reproducing the experimental observations. In a

first stage, the numerical simulations were performed with the main objective of validating the experimental results. For that purpose, all the experimental results obtained for the Newtonian fluid flows were compared with the corresponding numerical predictions. In addition, three-dimensional numerical simulations of Newtonian and non-Newtonian fluids were also carried out with the aim of enabling a thorough investigation of the 3D flow characteristics, which might not be easily observable experimentally, and assessing elastic effects on the fluid flow.

1.3. Outline of the thesis

This dissertation reports the investigation of strong extensional flows of viscoelastic fluids under low inertia flow conditions. The central part of this thesis is composed of 6 scientific articles published (3), submitted (2) or to be submitted for publication (1) in international refereed journals during the course of the PhD studies, each of which represents an independent chapter of this thesis (Chapter 3 to 8). Thus, in each chapter, we start by providing a literature review of previous experimental and numerical studies that are strictly related with the key topic discussed in that chapter. The experimental set-up and techniques used as well as the fluids and their rheological characterization are then presented, followed by the numerical method, governing equations and computational meshes used. Subsequently, the results are presented and discussed and the main conclusions are summarized. It is important to note that in the review of each chapter, we summarize and discuss the background investigations previously performed and present the guidelines of our work. Moreover, since the numerical code utilized in the simulations was employed in a user's perspective, as explained in Section 1.2, only the fundamentals that are essential to this research work are described. On the other hand, an entire chapter (Chapter 2) is devoted to clarify and delineate the principles of the experimental techniques used. This was done since experimental techniques and methods were used at macro- and micro-scale investigations, and it became essential to establish the connections and explain the differences of the fundamental principles.

Chapter 2 details the experimental techniques employed. The working principles are described and the differences between macro- and micro-scale experiments are established. Moreover, basic concepts of rheometry are presented.

Chapter 3 presents the results obtained for the flow of Newtonian and Boger fluids through square-square contractions. The Boger fluid is a viscoelastic fluid which exhibits a shear viscosity nearly independent of the shear rate. Visualization of the flow patterns and PIV

measurements of the velocity field, as well as pressure drop measurements, are used to characterize the flow. The experimental results obtained for the Newtonian fluid flow are compared with those obtained from numerical simulations and very good agreement is observed. Moreover, the effect of the contraction ratio on the flow behavior is established. This chapter was published as a journal paper in the *Journal of non-Newtonian Fluid Mechanics* (Sousa *et al.*, 2009).

Chapter 4 presents an investigation of the flow of a shear-thinning viscoelastic fluid through square-square contractions. Visualization of the flow patterns and PIV measurements of the velocity field are employed to analyse the flow. The experimental results obtained for the Newtonian and the shear-thinning fluid flow are compared with the results obtained from numerical simulations, showing a reasonably good agreement. This chapter was published as a journal paper in the *Chemical Engineering Science* journal (Sousa *et al.*, 2011).

Chapter 5 provides a detailed study of Newtonian and viscoelastic fluid flow through square-square expansions. The three-dimensional laminar flow of a Newtonian and two viscoelastic fluids with different rheological behavior (a Boger fluid and a shear-thinning fluid, as used in Chapters 3 and 4 for contraction flows) is considered. In this case, visualizations of the flow patterns, PIV measurements of the velocity field and pressure drop measurements are the experimental techniques used to characterize the flow. Different expansion ratios are investigated and the experimental results are compared with numerical predictions. This chapter was submitted for publication in the *Journal of non-Newtonian Fluid Mechanics* (Sousa *et al.*, 2010b).

Chapter 6 presents a novel microfluidic rectifier able to operate efficiently under creeping flow conditions with viscoelastic fluids. Different geometries are analyzed in order to create an efficient microfluidic rectifier, with enhanced diodicity, and the microfluidic geometry that shows better performance is investigated in more detail. Microchannels with hyperbolic shape are employed, in order to generate strong extensional flow and consequently achieve significant anisotropic flow resistance between the two flow directions for each microchannel. A comparison between the flow of Newtonian and non-Newtonian fluids is shown. This chapter was published in the *Journal of non-Newtonian Fluid Mechanics* (Sousa *et al.*, 2010a).

Chapter 7 presents the analysis of the wall effects on the optimized shape of the microfluidic rectifier developed in Chapter 6. Two microchannels with the same geometrical configuration

are used, but with significantly different depths, demonstrating that the bounding walls have a significant effect on the observed diodicity. The microfluidic rectifiers are characterized in detail experimentally, using flow visualizations, pressure drop measurements and detailed velocity field measurements using micro-PIV. This chapter will be submitted soon for possible publication as a journal paper (Sousa *et al.*, 2010c).

Chapter 8 provides a detailed investigation on the flow of non-Newtonian fluids with a rheological behavior similar to that of human blood, known as blood analogue solutions, through microfluidic hyperbolic or abrupt contractions/expansions. The experimental results obtained from visualizations of the flow patterns, micro-PIV measurements and pressure drop measurements for a range of flow rates are discussed. Furthermore, a comparison between the experimental results and the results obtained numerically for a Newtonian fluid and a generalized Newtonian fluid with a shear rheology similar to human blood is also presented. This chapter was submitted to the journal *Biomicrofluidics* (Sousa *et al.*, 2010d).

This dissertation ends with a discussion of the main conclusions, the milestones achieved and identification of important topics for future research in related areas.

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EXPERIMENTAL TECHNIQUES AND PROCEDURES

2.1. Introduction

In this chapter, a detailed description of the experimental techniques employed in this research work is presented. The working principles and procedures pertaining to each experimental technique are described in a general manner and the fundamental concepts are introduced. In particular, the differences between the experimental techniques used at the macro- and micro-scales are pointed out and explained. More specific considerations for each individual investigation work carried out, are given in the respective chapter.

2.2. Microfluidic device fabrication

2.2.1. PDMS soft-lithography

The microfluidic devices used in this work were fabricated in polydimethylsiloxane (PDMS) using soft-lithography. The choice of this silicone-based polymer is related first, to its optical properties such as transparency, essential to access the flow visually and second, to the straightforward, rapid, very well documented and inexpensive fabrication method, which produces microchannels with well-defined features and sizes. Furthermore, PDMS is chemically inert, has a homogeneous structure and is biocompatible, and is therefore widely employed for replica molding (Sia and Whitesides, 2003).

The micro-fabrication process is comprised of four fundamental steps:

- (i) design of the micro-geometries using a CAD software;
- (ii) manufacture of the mask using the CAD drawings;
- (iii) manufacture of the mold using the mask;
- (iv) fabrication of the PDMS microchannels using the mold.

The geometries of the microchannels were drawn using AutoCAD[®] (Step (i)) and the masks were sent to be *printed* in a specialized workshop which transfers the drawing onto the mask substrate (Step (ii)). Chrome masks are generally used when high-resolution, stable masks are required. This type of mask is obtained by sputtering a chrome layer onto a glass or quartz substrate in which the CAD image is etched. Chrome is a very hard and resistant material which together with a stable substrate with good optical characteristics such as glass produces robust, durable and stable masks. The devices fabricated using this kind of mask typically exhibit well-defined boundaries and sharp edges with a resolution of about 1-5 μm . There are other alternatives such as transparency masks, in which an emulsion layer of gelatin and silver halide is printed in a polyester film. Though cheaper, this type of mask is not rigid and presents lower durability and stability to temperature and humidity variations. Therefore, film masks need to be stored and used in a controlled environment (temperature and humidity) when dimensional accuracy is essential. The devices fabricated using this kind of mask typically exhibit increased wall roughness and a lack of definition of the boundaries and therefore are used mostly when feature sizes greater than 50 μm are required (Zaouk *et al.*, 2006).

Since the spatial resolution of the mask has a major impact on the final quality of the mold and microchannels, chrome masks were used for the manufacture of PDMS microchips required for the present work. This guarantees stability and allows for the fabrication of micro-devices with higher resolution and better-defined features with sizes smaller than 20 μm . An example of a chrome mask used in this work is shown in Figure 2.1.

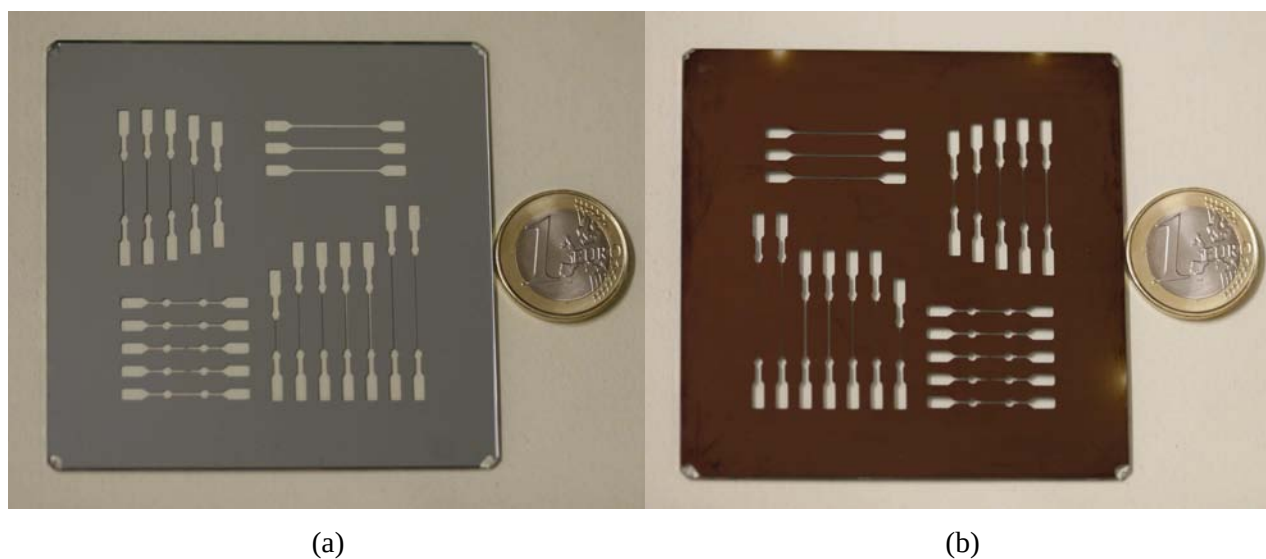


Figure 2.1. Chrome mask used in the fabrication process of the PDMS devices: (a) glass side; (b) chrome side.

The manufacture of the mold (Step (iii)) must be performed in a clean room, where the level of contaminants, such as dust, microorganisms, chemical vapors or other type of particles are strictly controlled, and minimized, in order to avoid interference in the production of the mask and the mold. Furthermore, humidity and temperature are also monitored and kept within certain levels. In the host research group laboratory there are no clean room facilities and consequently, the molds used in this work were ordered from external laboratories. The molds were fabricated by photolithography, a technique which was originally used for manufacturing microelectronic components, like integrated circuits (Xia and Whitesides, 1998). A SU-8 photoresist was used to create a positive-relief on the mold surface. SU-8 is a commonly used negative photoresist developed by IBM (Lorenz *et al.*, 1997; Zhang *et al.*, 2001; Li *et al.*, 2003). The main advantage of this photoresist is its ability to achieve a high degree of crosslinking after exposure, resulting in resistant features with good side-wall resolution (Bubendorfer *et al.*, 2007). The mold containing the inverse structure of the microchannels to be produced is then used to fabricate the microchannels. An example of a mold used in this work is shown in Figure 2.2.

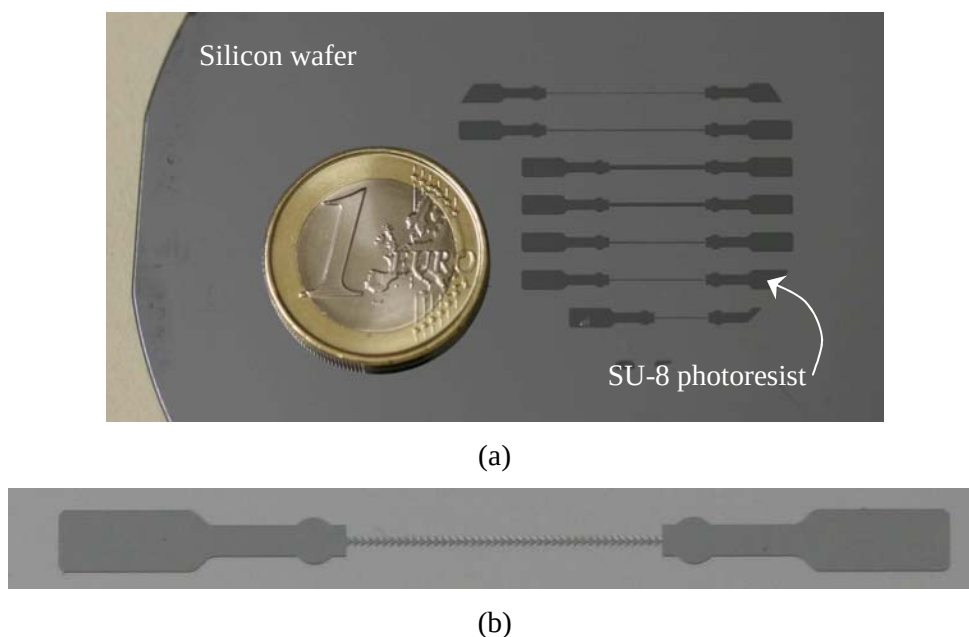


Figure 2.2. Example of a SU-8 mold used in the fabrication process of the PDMS devices: (a) overview of part of the SU-8 mold; (b) zoomed view of the inverted structure of a channel in a SU-8 mold.

The microfluidic devices were fabricated using soft-lithography, by casting PDMS on the mold (Step (iv)). PDMS soft-lithography is more robust and less dependent on environmental conditions than the photo-lithography technique used to manufacture the SU-8 molds. Therefore,

the PDMS microchannels used in the present experiments were fabricated in house in a *regular* microfluidics laboratory (i.e. without strict control of dust, humidity and temperature conditions).

The PDMS used in this work (Sylgard 184, Dow Corning) is commercially available as a pre-polymer kit composed of a PDMS oligomer and a crosslinking agent or curing agent, which are mixed in certain proportions to produce the polymer used to fabricate the micro-devices. This polymer belongs to the category of elastomers and has the following repeating unit: $[(\text{CH}_3)_2\text{SiO}]$. The Young's modulus of PDMS is approximately 1 MPa which makes it a relatively stiff material (Thorsen, 2003). This value, however, depends on the extension of cross-linking between chains, ranging from 0.42 MPa to 1.5 MPa for mixtures in a proportion of 20:1 and 5:1, respectively and at the standard temperature of cure, 80°C (Oliveira and Cerqueira, 2010).

With a mold available, multiple copies of the (inverted) pattern can be produced in a straightforward manner since the same mold can be used repeatedly without a significant reduction in its performance. The fabrication of PDMS microchips by soft-lithography is a well-established process, however fabrication procedures have to be adapted and optimized according to the existing materials and conditions in the host laboratory. An overview of the fabrication procedure adopted in this experimental work is shown schematically in Figure 2.4 and described in detail below.

Step 1. Silanize the SU-8 molds

A few drops of trichlorosilane (tridecofluoro-1,1,2,2-tetrahydrooctyl-1-trichlorosilane, United Chemical Technologies) are placed on a Petri dish containing the mold for 20 minutes. This process should be carried out in an exhaustion chamber and needs to be performed to allow the PDMS substrate to be easily removed from the mold (in step 7) after curing. Note that this step needs to be done once before the fabrication process and then repeated when the PDMS becomes difficult to remove from the mold.

Step 2. Prepare of the PDMS mixtures

The microfluidic chips are composed by the PDMS substrate, which contains the channels structure, and a thin PDMS layer that covers the glass slide used for sealing the micro-devices. Different ratios of oligomer to curing agent were tested for the two PDMS layers in order to obtain a chip with a high-quality seal. The best results were achieved when the components are

mixed in a 5:1 ratio for the substrate and a 20:1 ratio for the glass slide film. A vortex mixer is used to homogenize the solutions.

Step 3. Degass the mixtures

In order to remove the air bubbles that appear during the mixing procedure, the solutions are degassed for several minutes using a desiccator connected to a vacuum pump.

Step 4. Pour 5:1 PDMS mixture over SU-8 mold

The silanized mold is covered with the 5:1 polymer solution and degassed with a vacuum pump. Remaining air bubbles are removed by piercing with a thin syringe needle.

Step 5. Prepare glass slides and spincoat with 20:1 PDMS mixture

Glass slides are cleaned with acetone and ethanol and dried in the oven. The 20:1 PDMS mixture is then spincoated onto the cleaned glass slide using a spin coater (Laurell WS-650S-6NPP). A 30 μm thick layer of PDMS is obtained using spin speeds of $\omega = 5000$ rpm for $t = 50$ s following a 10 s ramping time (Figure 2.3). Different layer thicknesses can be obtained by changing the spincoating velocities (cf. Appendix A).

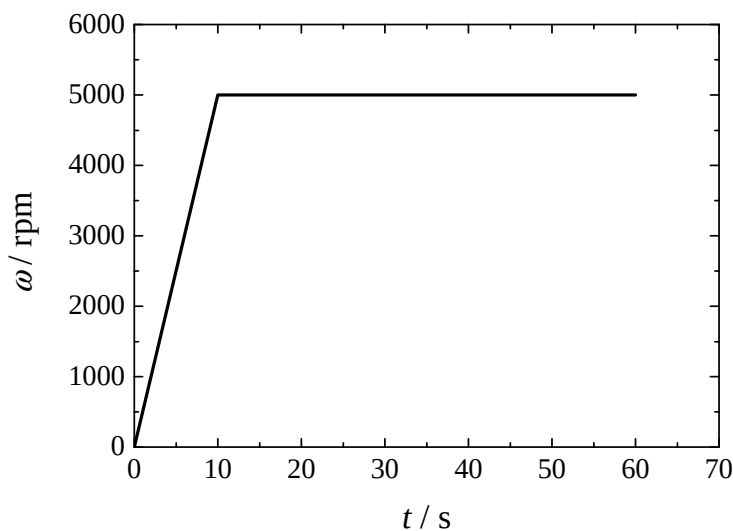


Figure 2.3. Spincoating protocol used to coat the glass slide with a 30 μm thick PDMS layer.

Step 6. Bake the PDMS layers

Place the mold with a layer of PDMS as well the PDMS coated glass slide in the oven to cure the PDMS for 20 minutes at 80°C. It is important that the oven remains at a nearly-constant temperature for the entire duration of the curing process.

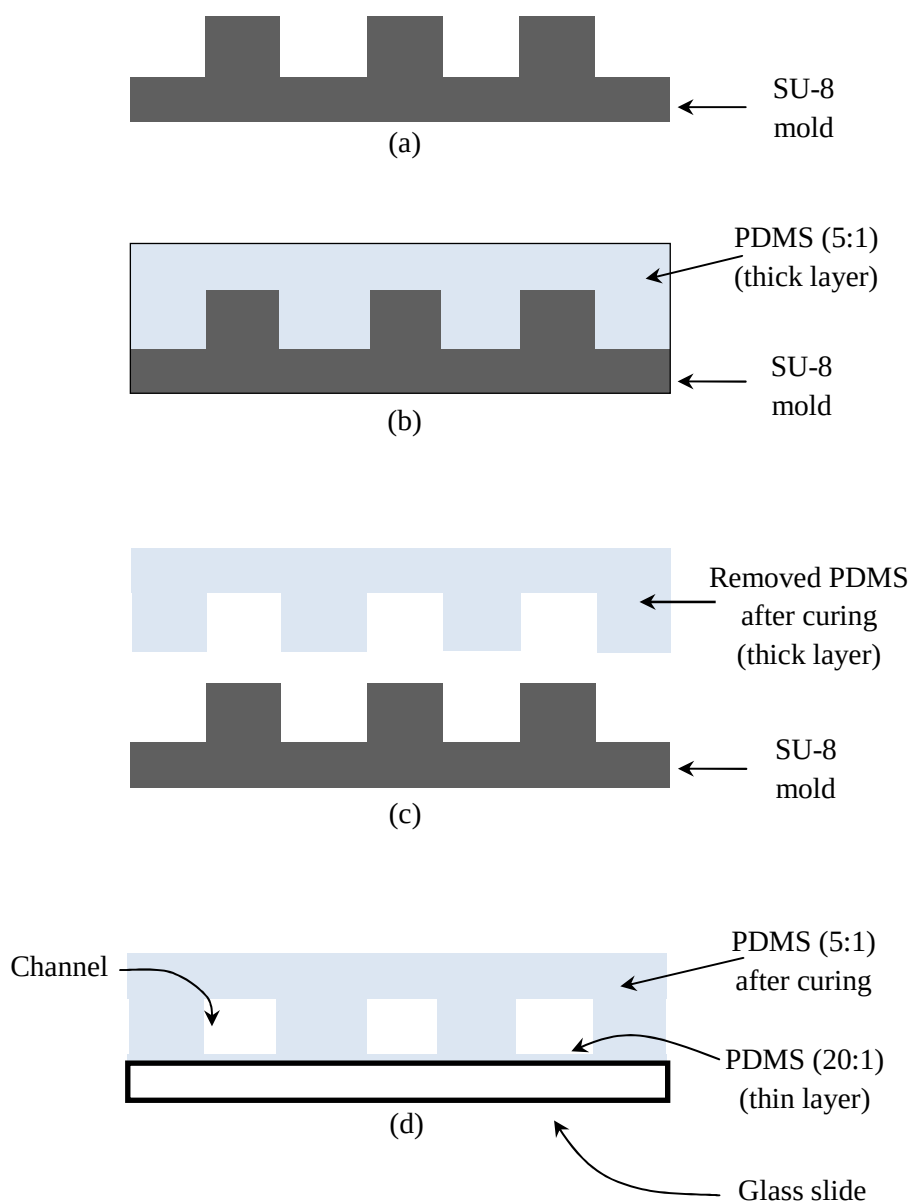


Figure 2.4. Overview of the PDMS device fabrication procedure. (a) Cross-section of the SU-8 mold with a positive relief. (b) The mixture of oligomer and curing agent, prepared in a ratio of 5:1, is poured onto the SU-8 mold (thick layer) and placed in the oven to cure during 20 minutes. (c) After curing, the PDMS thick layer is removed from the SU-8 mold and access ports are created. (d) The PDMS layer containing the channel structure is bonded to the glass slide covered with a thin layer of PDMS and placed in the oven to further seal the channel.

Step 7. Create access ports

After curing, the PDMS layer containing the microchannel structures is cut and peeled off from the mold. Inlet, outlet and pressure ports are created by punching holes at the desired locations in the PDMS substrate using a small syringe tip. The walls of the syringe tip must be smooth and straight for best results. The PDMS residues resultant from punching the holes should be removed carefully in order to maintain the microchannels as clean as possible.

Step 8. Seal the microchannels

Finally, the PDMS microchannels are sealed with the PDMS covered glass slide and placed back into the oven for approximately 12 hours at 80°C to promote a better adhesion and complete the cure of PDMS.

2.2.2. In-situ photopolymerization

The absence of clean room facilities at the host research group laboratory implies that the masks and molds necessary for the standard soft-lithography technique, described in Section 2.2.1, had to be manufactured abroad, which became a time-limiting step in the fabrication procedure. As such, a new alternative technique for microchannel fabrication was implemented in parallel. With this new technique, microsystems are fabricated using in-situ photopolymerization of a liquid-phase photo-curable material and consequently the existence of a clean room is no longer fundamental for the fabrication of fluidic devices.

This technique, called ultra-rapid prototyping (URP), consists of the following steps:

- a) assembly;
- b) filling;
- c) exposing;
- d) cleaning.

An empty chamber with access ports is assembled, filled with a liquid prepolymer and exposed to ultraviolet (UV) light ($\lambda = 365$ nm) through a high-resolution film mask (Kim and Beebe, 2007). After the photopolymerization reaction occurs, the exposed material forms a solid structure that defines the channel and the unreacted liquid, contained within the channel, is removed by washing with a solvent. This technique offers a great versatility, since various designs of microfluidic systems can be fabricated in a few minutes by changing the mask.

Moreover, a bonding step, which is often a yield-limiting step in fabrication, is not necessary (Khoury *et al.*, 2002).

A HybriWell™ hybridization chamber (HBW2160, Grace Bio-Labs, Inc., Bend, OR) is attached to the edges of a standard microscope glass slide (76 mm × 26 mm × 1 mm) to form an empty chamber (Figure 2.5(a)). The thickness of the adhesive gasket, which attaches to the glass slide, defines the depth of the empty chamber and consequently the depth of the microchannel that will be patterned (Figure 2.5(b)). Chambers 250 μm in depth were used, but different dimensions are also available. The top of the HybriWell™ is composed of a flexible polycarbonate sheet with a thickness of 180 μm. The chamber is prepared as shown in Figure 2.5.

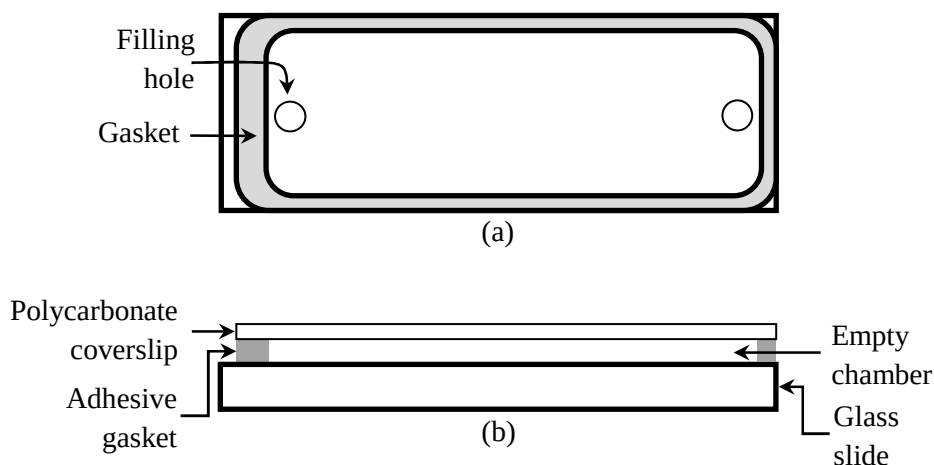


Figure 2.5. Cartridge preparation. (a) A HybriWell™ chamber with pre-drilled holes is attached to a microscope glass slide (top view). (b) The created cartridge is covered by a polycarbonate sheet and the sidewalls are formed by the adhesive gasket (side view).

The prepolymer was prepared by mixing the following components: the monomer, isobornyl acrylate (IBA, Sigma-Aldrich); the crosslinker, tetraethylene glycol dimethacrylate (TeGDMA, Polyscience); the photoinitiator, 2,2-dimethoxy-2-phenyl-acetophenone (DMPA, Sigma-Aldrich). The composition of the components of the prepolymer solution is presented in Table 2.1. The prepolymer was prepared in a dark bottle in order to inhibit polymerization by exposure to ambient light.

Table 2.1. Composition and function of the prepolymer components.

Component	Function	% wt.
Isobornyl acrylate, IBA	monomer	92
Tetraethylene glycol dimethacrylate, TeGDMA	crosslinker	5
2,2-Dimethoxy-2-phenyl-acetophenone, DMPA	photoinitiator	3

The IBA-based prepolymer is a transparent and inert material, with low viscosity, the polymerization reaction is photo-induced at 365 nm and low shrinkage occurs during the photopolymerization (López-García *et al.*, 2006).

The IBA prepolymer is supplied using a syringe without the tip, and the cavity is then completely filled by capillary effects and the existing air is totally removed (Figure 2.6(a)). The presence of oxygen within the cavity leads to a reduction of the polymerization rate since oxygen reacts with other free radicals at a much faster rate than IBA (Kim and Beebe, 2007). The photomask is a transparency film, in which the shape of the microchannel is printed in black with a high-resolution laser printer (typically 20 000 dpi) (Figure 2.6(b)). This photomask is then positioned under the glass slide and both are placed on top of an UV light source chamber as illustrated in Figure 2.6(c). The photopolymerization reaction occurs in a few seconds, but the time necessary to obtain the solid polymer depends on the different parameters, such as the UV light intensity or the age of the material. Another parameter initially thought to be important, the crosslinker concentration, was found to have a negligible impact, as discussed by García-Lopez *et al.* (2006).

Initially, the microchannels were created by exposing the HybriWell™ chamber, containing the prepolymer, and the photomask to a standard UV lamp. However, as a result of the multiple directions of the emission light, the resulting microchannels presented undesirable characteristics, such as non-vertical walls. With the appearance of such difficulties and looking for new alternatives, we found a novel solution, that consisted in using the microscope system for the fabrication of the microchannels.

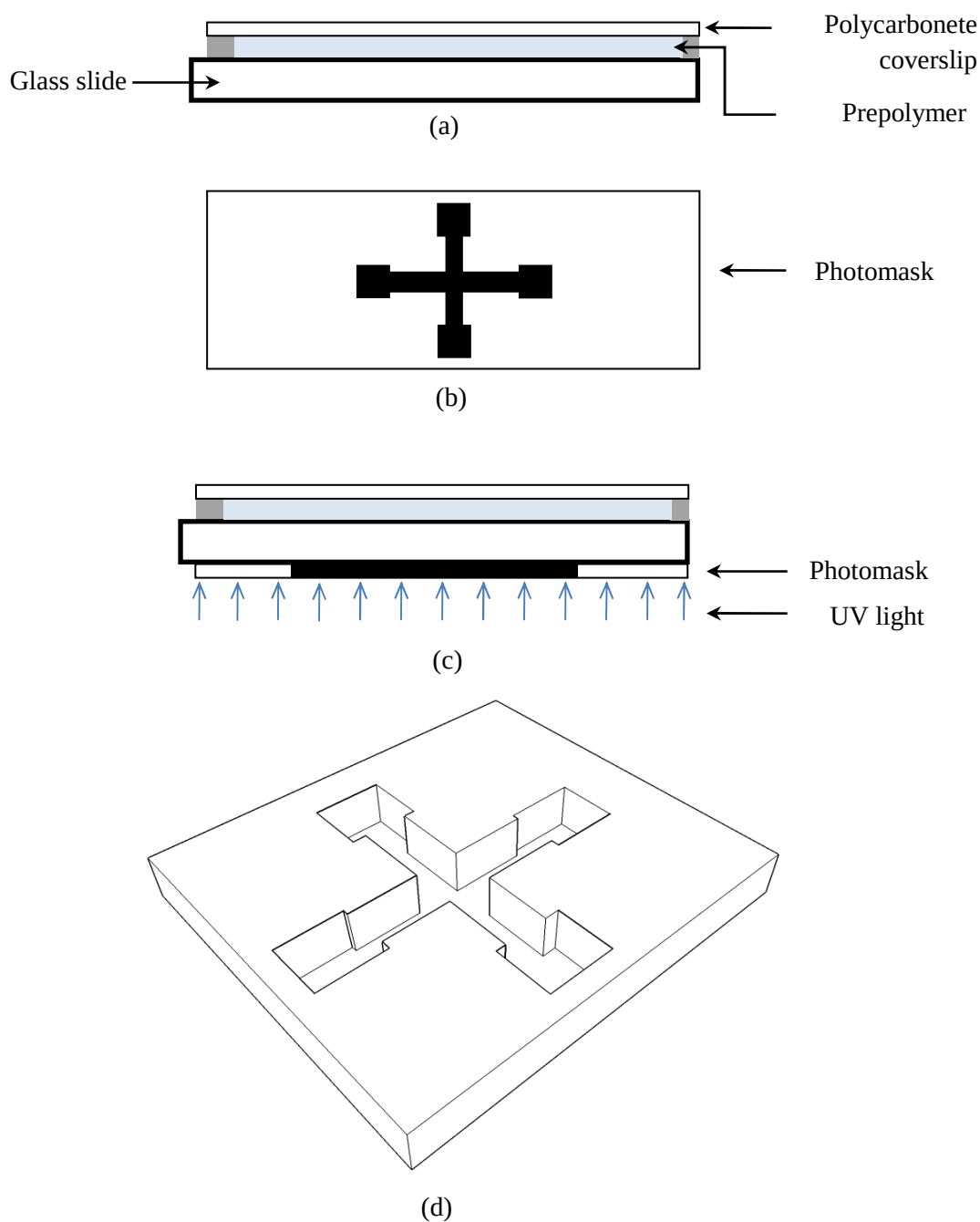


Figure 2.6. Overview of the microfluidic device fabrication using in-situ polymerization. (a) A HybriWell™ chamber is assembled and filled with the prepolymer mixture (side view). (b) The photo-mask used to select which regions will be cured (top view). (c) Arrangement used for the photo-polymerization. (d) 3-D view of the created channel.

Our microscope system includes a programmable xy moving stage, which was used to move the assembled chamber (HybriWell™ chamber filled with prepolymer) together with the photomask, as shown in Figure 2.6(c), during the photopolymerization process. The microscope mercury lamp (100 W) was used as the light source for photopolymerization. Since this lamp has a multispectral light, the microscope was fitted with a UV filter cube (Chroma Technology Corp., excitation 365, dichroic 380, emission LP 400 nm) so that only UV light with a wavelength of $\lambda = 365$ nm reaches the prepolymer. Different objectives with low magnifications and low numerical apertures (NA) were tested in order to guarantee that the incident light on the prepolymer was as vertical as possible. For this work, a 5× (NA = 0.12) and 2.5× (NA = 0.07) objectives were used. The size and shape of the cross-section of the generated light volume that passes through the objective was controlled by adjusting the microscope diaphragm aperture. A large aperture leads to a higher cross-sectional area of light focused on the liquid, whereas a low aperture leads to a small cross-sectional area. If the incident light has a small area and is collected at the center of the spotlight available within the optical system, a nearly vertical pathway of light is attained. The question concerning the direction of the light is crucial in order to obtain microchannels with vertical walls. The exposing time of the prepolymer to the UV light, and consequently the photopolymerization reaction rate, is controlled by the aperture of the microscope diaphragm and the velocity of the xy microscope moving stage. It was found that an overexposure produced channels with dimensions smaller than the photomask while an underexposure led to channel dimensions larger than the photomask. Typically, using our set-up, the velocity of the xy stage was set to 50 $\mu\text{m/s}$ using a 2.5× objective (NA = 0.07).

After the photopolymerization, the unreacted monomer existing in the microchannel region is removed. For this, we impose first a low flow rate of air through the microchannel, and then water followed by ethanol to wash away unpolymerized liquid. The fluidic access ports are made by sticking Tygon tubing via an o-ring glued to the chamber ports located on the polycarbonate coverslip (cf. Figure 2.5(a)).

At the present moment, using in-situ photopolymerization we cannot yet produce microchannels with small-sized features with such high quality as those obtained by PDMS soft-lithography in part due to the lower resolution of the transparency mask as compared to the chrome mask used in the soft-lithography technique. In particular, it is difficult to obtain sharp corners using this simple technique. Nevertheless, the quality of the channels fabricated

(illustrated in Figure 2.7(a)) is sufficient to perform exploratory microfluidic experiments, such as flow visualization (depicted in Figure 2.7(b)), with a much shorter fabrication time.

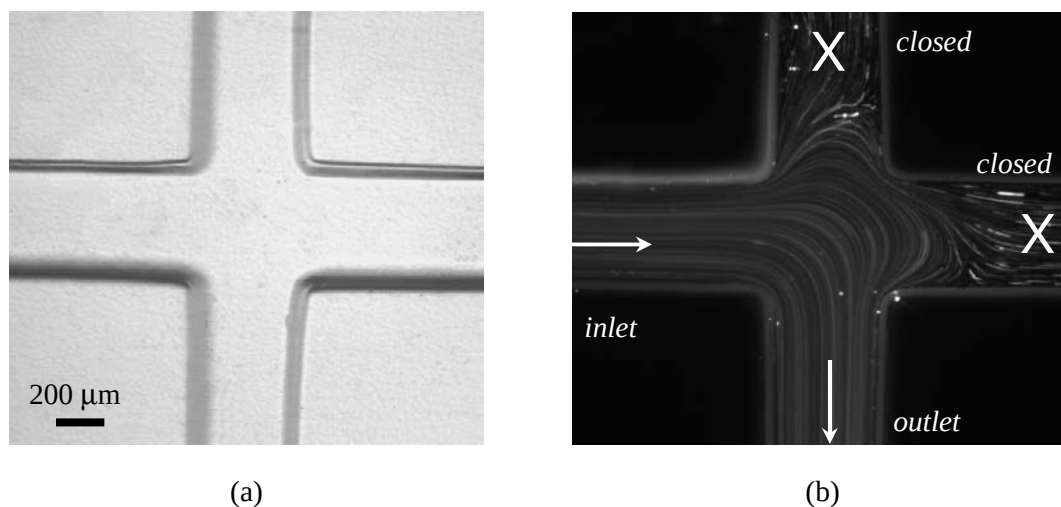


Figure 2.7. Cross-channel fabricated using in-situ photopolymerization: (a) micrograph of a channel after fabrication; (b) fluid flow patterns.

This new technique has many advantages: the fabrication time is reduced, the fabrication process has a low cost and the protocol is simple. Furthermore, it is also possible to use this technique to produce microfluidic structures distinct from microchannels. For instance, a mold can be created using photopolymerization and then used to fabricate various microchannels by soft-lithography. In addition, this technique is of utmost value when structures such as posts need to be incorporated within an already assembled microchannel.

In summary, this technique can be seen as a complement to the standard soft-lithography and can be useful in scientific research where a large number of different geometries need to be tested. In this case, one can take advantage of the fact that photopolymerization does not strictly require the use of a mask (a transparency mask is applied in this work only to improve the resolution of the channels) since the microscope moving stage can be programmed to move according to the design the required structures, and in this case can be used to produce a microfluidic channel in about 2-4 hours, from the initial design to the final sealing and connection to the tygon tubing.

2.3. Flow characterization

2.3.1. Imaging techniques

Imaging techniques are of great importance in order to visualize the fluid flow and understand its behavior when spatial and temporal gradients exist (Sinton, 2004). It allows a qualitative and quantitative analysis by assessing different flow phenomena through flow patterns or measuring the velocity field of the flow. In the present work, two particle-based flow visualization techniques were employed to track the flow: streakline photography (to characterize the flow patterns) and particle image velocimetry (PIV, to quantify the velocity field).

These techniques rely on a tracer being introduced within the bulk fluid in order to follow the fluid motion as accurately as possible. For this, the fluid was seeded with particles that were selected according to the following properties (Durst *et al.*, 1981):

- particle density similar to the fluid density;
- chemically inactive, non-toxic, non-corrosive and non-volatile;
- optimum size to follow accurately the flow and scatter sufficient light in order to be detected.

Apart from the particles that are added to the fluid, the visualization methods are completely non-intrusive.

The two imaging techniques were employed both in the macro- and micro-scale experiments performed in this work. Although the principles of the methods at both scales are similar, the practical implementation presents some differences, particularly with respect to the imaging set-up and the flow illumination concept. In macroscopic experiments a light sheet is used to illuminate the tracer particles within the plane of the flow under study while, in the experiments at the micro-scale, volume illumination is used. These distinctions will be explained in detail in the following sub-sections.

2.3.1.1. Imaging Set-up

Macro-scale

At the macro-scale experiments, a laser coupled with a cylindrical lens is used to generate a single wavelength light sheet, which illuminates the plane of the flow under investigation. When illuminated, the tracer particles reflect the light, which is then captured using a digital camera. A

generic set-up for the macroscopic experiments is illustrated in Figure 2.8. The true depth of field is given by the thickness of the laser light sheet, since only the illuminated particles are captured by the camera and every particle in the depth of field of the recording lens is well-focused (Nguyen and Wereley, 2006) and consequently, the background noise from out of focus particles is minimized. Typically the thickness of the sheet of laser light is of the order of 0.5 - 1 mm.

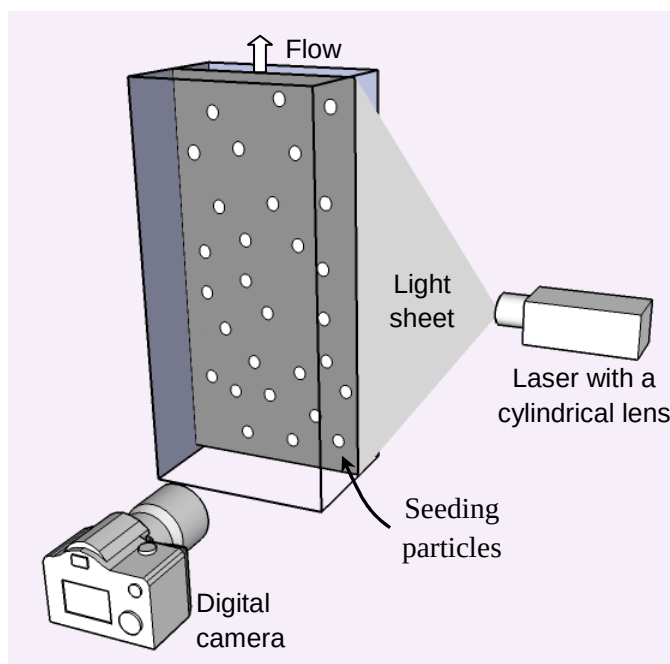


Figure 2.8. Schematic of the basic principles of the optical techniques used at the macro-scale experiments.

Micro-scale

In the experiments at the micro-scale, epi-fluorescence microscopy is the basis for the two optical techniques used. For that, we used an inverted fluorescence microscope (DMI 5000M, Leica Microsystems GmbH) properly equipped with a light source, appropriate filters and a digital camera. All the techniques utilize a similar generic imaging setup, which is illustrated in Figure 2.9.

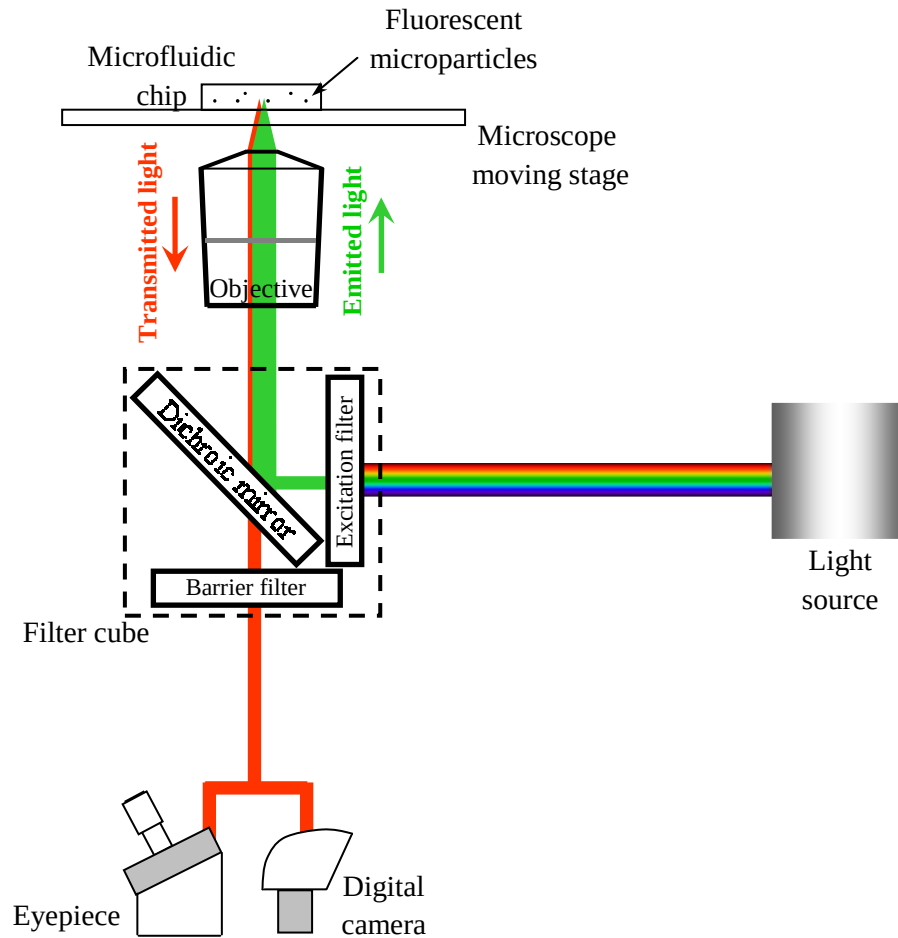


Figure 2.9. Schematic diagram of a generic optical system used in epi-fluorescence.

In fluorescence microscopy, instead of the highly reflective particles used at the macro-scale, fluorescent particles, which exhibit different excitation and emission spectra, are used as tracer particles. Furthermore, it often relies on multispectral light generated by the light source and, as consequence, the optical train required is rather complex as shown in Figure 2.9. The light emitted by the light source reaches an excitation filter which allows the passage of a band of wavelengths (range of transmission: $\lambda = 530\text{-}545\text{ nm}$). This excitation light is then reflected by a dichroic mirror, passes through the objective and finally reaches the transparent microchip containing the channel filled with the seeded fluid. The fluorescent particles employed here have excitation and emission peaks at 520 and 580 nm, respectively. This difference between excitation and emission spectra of a fluorescent specimen is denominated as Stokes' shift. Subsequently, the emitted light passes back through the objective and the dichroic mirror, which is able to separate the reflected and emitted light ($\lambda > 565\text{ nm}$ are transmitted while $\lambda < 565\text{ nm}$

are reflected), and is filtered by a barrier filter (range of transmission $\lambda > 610$ nm) before reaching the eyepiece or the digital camera.

2.3.1.2. Streak line photography

Macro-scale

In the macroscopic experiments performed, a laser diode was used as a light source: either a green 3 mW 532 nm laser diode (Imatronic, model LLM115) or a red 5 mW 635 nm laser diode (Vector, model 5200-20). In some cases, for which very large recirculations were achieved, the length of the light sheet obtained with these laser diodes was not sufficient to illuminate the whole region of interest. In these cases, a pulsed Nd-YAG laser (employed in the PIV measurements) was used at the maximum frequency (15 Hz), since the light sheet obtained using Nd-YAG laser was longer and more intense than that produced by the laser diodes. Moving the laser diodes further away from the region of interest results in a large illumination area, but the light intensity becomes insufficient and less focused. The fluids under investigation were seeded with 10 μm PVC reflective particles at a concentration of approximately 30 ppm. A 3504 $\times$ 2336 pixels Canon EOS 30D digital camera equipped with a macro lens (Canon EF100mm, f/2.8) was utilized to capture the path of the particles during a certain time interval, which is designated as the exposure time. For long enough exposure times (which have to be tuned according to flow conditions), the particles appear as streaks. As a result, we obtain a visual representation of the pathlines of the illuminated particles, which represent the fluid motion.

Micro-scale

The light source used in streak line photography at the micro-scale is a mercury lamp, which is attached to the microscope. Nile red fluorescent micro-particles, 1 μm in diameter, with excitation and emission wavelengths of 520 and 580 nm, respectively, were used to seed the working fluids at a concentration of 0.1%. All streak line images were captured using a 10 $\times$ (NA = 0.25) objective in order to see the largest possible area of the channel (and therefore have an overview of the flow), while being able to capture in detail the flow features of interest. In addition, a CCD camera (DFC350 FX, Leica Microsystems GmbH) was used to acquire the images and movies, which were recorded as a complement to the images for a more complete analysis, in particular for unsteady flow.

2.3.2. Particle image velocimetry

Particle image velocimetry is an imaging technique used for the characterization of the flow kinematics that relies on measuring the local velocity components from the average displacement of tracer particles (Δx) in an interrogation region over a known time (Δt) (Wereley and Meinhart, 2005). This process is carried out using a double-frame imaging technique with a cross-correlation algorithm for extracting local particle displacements (Stone *et al.*, 2002).

Macro-scale

Particle image velocimetry experiments were performed at the macro-scale to measure the velocity field of the flow through square channels with a constriction (cf. Chapters 3-5). A 532 nm doubled Nd:YAG pulsed laser (Solo PIV III, New Wave Research) was used in order to illuminate the flow. The images were acquired using a digital CCD camera (Flow Sense 2M from Dantec Dynamics) coupled with a lens (Nikon AF Micro 60 mm) and a filter, which eliminates background noise and allows that only light with a wavelength near 532 nm reaches the CCD sensor.

The time between pulses (Δt) was adjusted for best results according to specific flow conditions. For time between pulses up to $30 \times 10^3 \mu s$, the PIV system can be operated in double frame mode, while for Δt above that value the system worked in single frame mode. In this latter case, the frequency between pulses was used in order to create double frame images and subsequently process the data and obtain the final vector map. A set of 60 double frame images were recorded for each case.

Since in this investigation the velocity gradient within the square channel is high and different flow phenomena appear depending on the flow conditions (such as recirculations and diverging streamlines) an adaptive correlation method was applied to improve the resolution of the flow. The adaptive correlation is an iterative process in which the interrogation area is successively refined. In this method, the in-plane dropout or loss of particles in the interrogation areas between light pulses is reduced. The thickness of the light sheet is 1 mm and the out-of-plane spatial resolution is given by this value (Wereley and Meinhart, 2005). Each pair of images originates a vector map and an ensemble average of those vector maps provide the final vector map of the flow. A detailed list of procedures used during macro-PIV measurements is presented in Appendix B.

Micro-scale

One of the main differences that distinguishes μ PIV from *regular* macro-PIV is the flow illumination method as briefly pointed out in Section (2.3.1.1). In *regular* PIV, a light sheet is used to illuminate the tracer particles, while in μ PIV volume illumination is used as shown schematically in Figure 2.10. This type of illumination leads to high background noise from out-of-focus particles, since in this case both in-focus and out-of-focus particles appear in the image. Moreover, when particle size is reduced, interactions between particles and particles-fluid can originate Brownian motion.

The light source was a double-pulsed 532 nm Nd:YAG laser (Dual Power 65-15, Dantec Dynamics) and the camera was a 2048×2048 pixels CCD digital camera (Flow Sense 4M, Dantec Dynamics) running on double frame mode. Two different objectives were used: 20× (NA = 0.4) and 40× (NA = 0.55) objectives. The 20× microscope objective was used to cover the whole area of the test section under investigation and the 40× objective was used in order to increase the resolution near the contraction regions and therefore achieve higher accuracy in those specific regions.

With the aim of improving the procedure to measure the velocity field of the Newtonian and non-Newtonian fluid flow in the complex microgeometries used in this investigation, a preliminary validation study was carried out (results in Appendix C). For this, velocity field measurements were performed using a straight microchannel and a Newtonian fluid was employed as the working fluid. The experimental velocity profiles obtained were then compared with the corresponding analytical solution for fully-developed flow conditions. This preliminary study enabled a more informed choice of parameters in subsequent experiments.

For micro-scale experiments at steady flow conditions, 150 pairs of images were acquired and an ensemble average correlation, using all pairs of images, was performed in order to obtain the final vector maps. Wereley and Meinhart (2005) proposed this method to improve the accuracy of μ PIV measurements, in which there is generally low particle image density. The same authors (Wereley and Meinhart, 2005) have also described several other factors that affect spatial resolution and velocity accuracy. These are discussed below in the context of the present experiments.

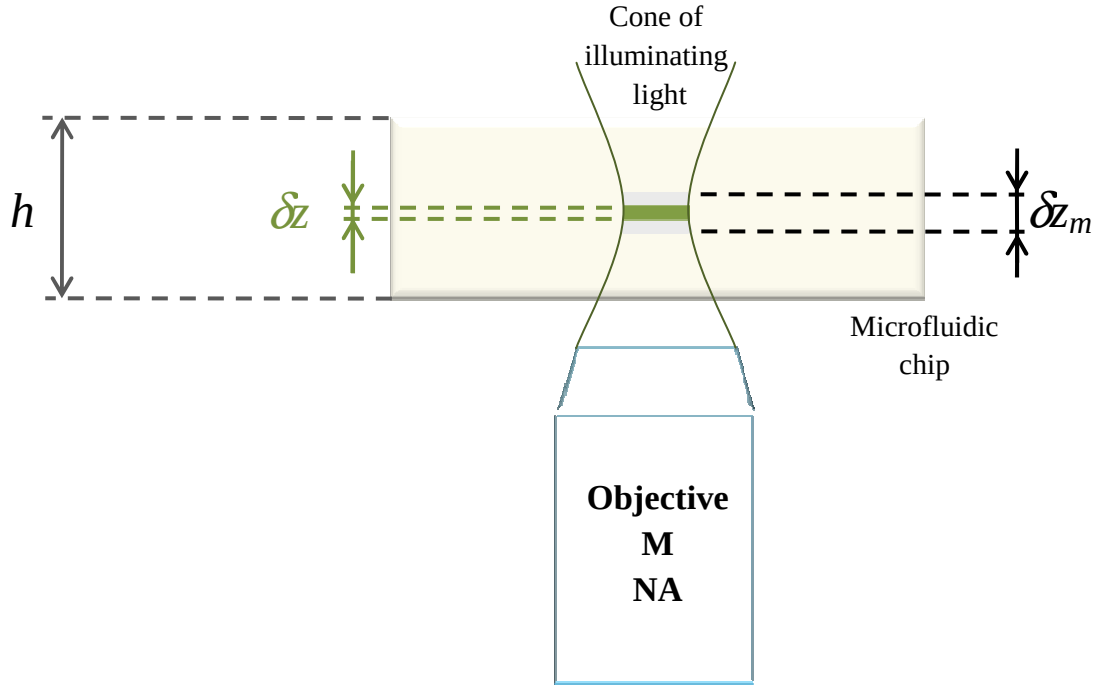


Figure 2.10. A schematic diagram representing the volume illumination concept used in epi-fluorescence. The total measurement depth, δz_m , the depth of field, δz , and depth of the microchannel, h , are also represented.

a) In-plane Spatial Resolution

The diameter of each fluorescent particle should be resolved in 3-4 pixels of the acquired image. The diffraction-limited spot size of a point source of light, d_s , is given by (Wereley and Meinhart, 2005):

$$d_s = 2.44(M + 1) \frac{\lambda}{2NA}, \quad (2.1)$$

in which, M is the magnification of the objective, NA is the numerical aperture of the objective and λ is the wavelength of light. The effective image diameter, projected back into the flow, d_e/M , is defined as (Wereley and Meinhart, 2005):

$$\frac{d_e}{M} = \frac{[d_s^2 + M^2 d_p^2]^{1/2}}{M}, \quad (2.2)$$

where d_p is the real diameter of the particle. For the present experimental set-up, the referred parameters are listed in Table 2.2.

Since the particle diameter should be resolved within 3-4 pixels, the uncertainty of the correlation peak location is equivalent to $d_e/(10M)$ (Prasad *et al.*, 1992). Moreover, the relation between this uncertainty and the particle displacement should be lower than 2%. Therefore, the maximum particle displacements considered in the present experiments should be 9.6 μm and 13 μm for the 20 $\times$ or 40 $\times$ objectives, respectively.

Table 2.2. Numerical aperture, diffraction-limited spot size and effective particle diameter projected back into the flow for the two objectives used in present investigation.

Objective	20 $\times$	40 $\times$
NA	0.4	0.55
$d_s / \mu\text{m}$	37	53
$(d_e/M) / \mu\text{m}$	1.9	2.6

b) Out-of-plane spatial resolution

The out-of-plane thickness for μPIV experiments is sometimes given by the depth of field (cf. Figure 2.10). However, in μPIV volume illumination, out-of-focus particles are captured in the images and contribute to increase the background noise level. Therefore, unfocused particles should be considered in a more realistic model. Indeed, for each optical set-up, the total measurement depth, δz_m , is the depth over which there is a contribution of unfocused particles and is calculated as the sum of three different effects: the effect due to diffraction $[3n\lambda_0/(NA^2)]$, the effect due to geometrical optics $(2.16d_p/\tan\theta)$ and the effect due to the finite size of the particle (d_p) (Meinhart *et al.*, 2000):

$$\delta z_m = \frac{3n\lambda_0}{(NA)^2} + \frac{2.16d_p}{\tan\theta} + d_p, \quad (2.3)$$

where n is the refractive index, λ_0 is the wavelength of the light in vacuum and $\tan\theta \simeq \sin\theta = NA/n$. For μPIV experiments using the 20 $\times$ objective, the total measurements depth is $\delta z_m = 13.9 \mu\text{m}$, whereas for the set-up using the 40 $\times$ objective, $\delta z_m = 7.89 \mu\text{m}$.

c) Particle visibility

Particle visibility is the ratio of the intensity of an in-focus particle image to the background light produced by the out of focus particles (Olsen and Adrian, 2000). Furthermore, at least, an in-focus particle visibility (V) of about 1.5 is necessary for a reasonable high quality particle image. The fraction of particles by volume, V_{fr} , for a specific particle visibility is given by (Wereley and Meinhart, 2005):

$$V_{fr} = \frac{2d_p^3 M^2 \beta^2 \left(s_0 - \frac{h}{2}\right) \left(s_0 - \frac{h}{2} + h\right)}{3Vhs_0 \left[M^2 dp^2 + \frac{1.49\lambda^2}{NA^2} (M+1)^2\right]} \times 100\%, \quad (2.4)$$

where the parameter that defines the cut-off level of the edges of the particle image is $\beta^2 = 3.67$ (Adrian and Yao, 1985), s_0 is the objective working distance and h is the depth of the microchannel. Therefore, for a visibility of 1.5, a maximum particle concentration of 0.1 % (v/v) is admissible for the experimental set-up using the 20× (NA = 0.4) objective and a concentration of 0.2 % (v/v) when the 40× (NA = 0.55) objective is used.

d) Brownian motion

Fluorescent tracer particles should follow the fluid motion accurately without modifying the fluid velocity. However, when particles have small dimensions and in particular, when the fluid velocity is very low, Brownian motion can cause particle trajectories to fluctuate rather than to follow the pathlines of the fluid flow (Wereley and Meinhart, 2005).

The relative error due to Brownian motion in μ PIV is given by:

$$\varepsilon_x = \frac{\sigma_x}{\Delta x} = \frac{1}{u} \sqrt{\frac{2D}{\Delta t}}, \quad (2.5)$$

$$\varepsilon_y = \frac{\sigma_y}{\Delta y} = \frac{1}{v} \sqrt{\frac{2D}{\Delta t}}, \quad (2.6)$$

where σ is the displacement due to Brownian motion in the corresponding x or y direction, Δx and Δy are the total particle displacement in the x and y direction, u and v are the characteristic

velocity components in the x and y direction, Δt is the time interval between images and D is the diffusion coefficient, $D = k_B T / 3\pi\mu d_p$, in which k_B is the Boltzmann's constant ($k_B = 1.3807 \times 10^{-23} \text{ m}^2 \text{ kg s}^{-2} \text{ K}^{-1}$), T is the absolute temperature of the fluid and μ is the dynamic viscosity of the fluid.

Since most of the velocity properties analysed refer to the axial velocity at the centerline of the microchannel, ε_x is the important relative error to take into account. The higher Brownian error is calculated for the experiments using water at the lowest flow rate, $Q = 0.08 \text{ ml h}^{-1}$ for which the time between images used is $\Delta t = 150 \text{ }\mu\text{s}$. The value of the diffusion coefficient is $D = 8.70 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ corresponding to $\varepsilon_x = 0.014$. The resulting Brownian error can be substantially reduced to $\varepsilon / \sqrt{N}$ by averaging over several particles in a single interrogation spot and by ensemble averaging over several images (Wereley and Meinhart, 2005). Using 150 pairs of images and assuming an average of 5 particles in a single interrogation area reduces the relative error due to Brownian motion to 0.1%, which is the maximum error expected in all experiments.

2.3.3. Pressure drops measurements

Pressure drop measurements were performed in the micro- and macro-scale investigations. For this, the two ports of the differential pressure sensors were connected to the channel at the extremes of the test section, so that the pressure drop between a position upstream and downstream of the region of interest was measured.

Four pressure sensors (Honeywell 26PC) covering various differential pressure ranges (up to 7 kPa, 34 kPa, 100 kPa and 200 kPa) were employed according to the pressure drop to be measured, which depends on the geometrical arrangement as well as on the flow rates studied. Before use, the differential pressure sensors were calibrated utilizing two different methods: a static column of water for sensors covering pressure differences up to 34 kPa and a compressed air line with a manometer (Wika Instrument Corporation, model 332.50, accuracy of $\pm 2 \text{ kPa}$) for sensors covering higher differential pressures, up to 200 kPa. The calibration data of one pressure sensor is presented in Appendix D as an example. The pressure sensors were powered using a 12 V power supply (Lascar electronics, PSU 206) and the output data was acquired using a data acquisition card (NI USB-6218, National Instruments), which in turn was connected to the

computer through an USB port. In addition, the output data was recorded and monitored using LabView v7.1 software.

The pressure sensors typically showed a transient response until steady-state was reached, as illustrated in Figure 2.11. For Newtonian fluids, a constant value of the pressure drop, corresponding to the steady-state, was reached more rapidly than for polymeric memory fluids as can also be seen in Figure 2.11. After steady-state is attained, the signal reaches a plateau with superimposed low amplitude oscillations due mostly to electronic noise, and the average pressure drop is calculated by taking the arithmetic mean of the signal with time.

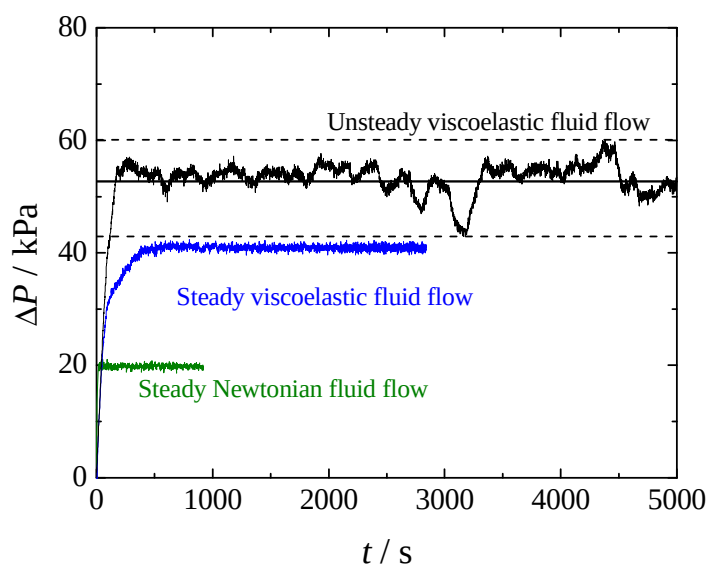


Figure 2.11. Pressure drop measured experimentally as a function of time. The Newtonian fluid is water and the viscoelastic fluid is an aqueous solution of polyethylene oxide (0.1 wt. %). The data shown for both fluids was obtained for the flow through the microfluidic rectifier, as will be presented in Chapters 6 and 7. For the unsteady viscoelastic fluid flow, the calculated average value of the pressure drop is shown as a solid thick line and the upper and lower values are shown as dashed lines.

For high Deborah numbers when the viscoelastic fluid flow becomes unsteady due to the onset of elastic instabilities, the pressure sensor response does not reach a steady value, i.e. the value of the pressure drop varies with time as illustrated in Figure 2.11. Pressure drop measurements for unsteady flow were only performed at the micro-scale and for those cases, in addition to the average value, the maximum and minimum values were also registered.

2.4. Fluid characterization

2.4.1. Solution composition and preparation

In this thesis, Newtonian and non-Newtonian fluids were used. Since the flow of Newtonian fluids have been extensively studied so far, in this thesis we also employed Newtonian fluids in order to improve the experimental procedures prior to the use of complex fluids and also in order to compare with numerical predictions and validate the experimental results. At the macro-scale, the Newtonian fluid used is a solution composed of water and glycerin, with a composition such that the shear viscosity is comparable to that of the viscoelastic fluids used in the macro-scale experiments. At the micro-scale study, water was used as the reference fluid.

Non-Newtonian fluids were prepared by adding a small amount of a polymer to a Newtonian solvent so that viscoelastic properties were achieved. The polymers used were: polyacrylamide, denoted as PAA, polyethylene oxide, denoted as PEO and xanthan gum also abbreviated as XG.

a) Polyacrylamide

Polyacrylamide results from a free-radical polymerization reaction of the monomer acrylamide, whose chemical structure is shown in Figure 2.12:

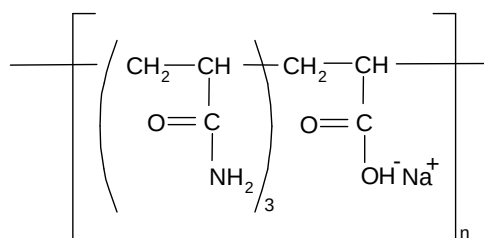


Figure 2.12. Chemical structure of the repeating unit of anionic polyacrylamide.

This polymer is widely used in biology and in industry as a thickening and flocculating agent (Lewis and Richard, 2002). Degradation of this polymer occurs with the scission of the chain which leads to a significant change of the rheological properties of polymer solutions.

b) Polyethylene oxide

Polyethylene oxide is used frequently as a drag reducing agent and has found applications in numerous areas. This polymer is used in the pharmaceutical industry, e.g. for drug delivery; in

detergents; in personal care products; in paints, etc (Braun and Rosen, 2000). The chemical structure of the monomer is represented in Figure 2.13.

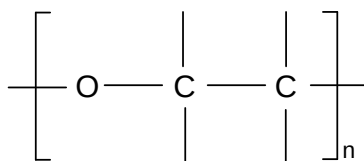


Figure 2.13. Chemical structure of the repeating unit of polyethylene oxide.

c) Xanthan gum

Xanthan gum is a long-chain heteropolysaccharide which is soluble in water and produced by microorganisms, namely *Xanthomonas campestris* (Braun and Rosen, 2000). The chemical structure of this gum is shown in Figure 2.14.

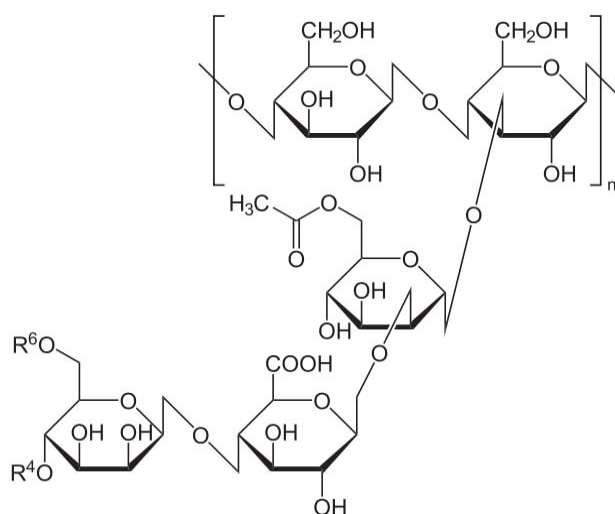


Figure 2.14. Chemical structure of the xanthan gum monomer.

This polymer has a large commercial importance as a thickening agent due to its chemical and rheological properties and because it is non-toxic and can be used to create highly viscous solutions. For these reasons, xanthan gum has been employed in a vast variety of industrial applications, such as in food, pharmaceutical, textile and petroleum applications (García-Ochoa *et al.*, 2000).

All polymers chosen for this thesis have long chains and consequently high molecular weights which confer to the non-Newtonian fluids, a significant elasticity level and viscoelastic flow behavior.

In the investigation at the macro-scale, PAA at different concentrations was used in the preparation of the viscoelastic fluids. The polymer was dissolved in water and glycerin by agitation at very low speeds, in order to avoid rupture of the polymer chains and consequently their degradation. Sodium chloride (NaCl) was also added to some fluids containing PAA to stabilize electrical charges of the polymer and render a Boger fluid rheological behavior (Stokes, 1998). In addition, a biocide agent (Kathon LXE, Rohm and Haas) was also added to the fluids at a concentration of 25 ppm (w/w) in order to prevent biofilm formation and biological degradation of the polymer solution.

For microfluidics measurements, polymers with various molecular weights were utilized in the preparation of the fluids. In this case, the polymers were dissolved in the Newtonian solvent using a magnetic stirrer at very low speeds (typically 60 rpm). The solutions were prepared in plastic bottles and were kept in the refrigerator to minimize biological degradation. Solutions composed of PEO were maintained in dark bottles due to photoinduced degradation of this polymer under certain conditions (Hassouna *et al.*, 2008). The solutions prepared were divided into two parts: one fraction to which fluorescent tracer particles were added, to be used in flow visualization and particle image velocimetry measurements, and another fraction to be used in pressure drop measurements that do not require the use of tracer particles.

For flow visualizations, a surfactant, sodium dodecyl sulphate (SDS), was added to reduce the adhesion of the tracer particles onto the PDMS channel walls. Above a certain concentration, which is known as the critical aggregation concentration (CAC), the surfactant tends to form micelle-like aggregates on backbone of the polymer chain (Hai and Han, 2006). For this reason, a concentration below the CAC was used in this investigation so that interactions between the surfactant and the polymer do not exist and there is no impact on the rheological properties of the fluid. The CAC for a PEO-SDS system is about 4-5 mM (Miller and Cooper-White, 2009). Above this concentration, the SDS molecules form aggregates on the backbone of the PEO chain and electrostatic repulsion between the components leads to an extension of the chain. Hence, a concentration of SDS of 0.1 % (w/w) (~ 3.5 mM) was used in the preparation of the PEO fluids. Moreover, the viscoelastic fluids containing SDS in their composition were characterized

rheologically in order to ensure that rheological behavior is similar to that found for the same viscoelastic fluid without the surfactant agent. For the PAA solution, a concentration of SDS of 0.05 % (w/w) was used. In this case, the CAC is 0.173 g SDS/g PAA (Hai and Han, 2006), which corresponds to 2.16×10^{-3} % (w/w) for the 125 ppm PAA solution.

2.4.2. Shear rheometry

2.4.2.1. Steady shear measurements

Material functions, such as shear viscosity (η), first and second normal stress differences (N_1 and N_2 , respectively) or first and second normal stress coefficients (Ψ_1 and Ψ_2 , respectively) are measured in steady shear rheometry using a rotational rheometer, in which a fluid sample is confined by the system geometry and a rotational motion is promoted in order to achieve a simple shear flow. In general, rotational rheometers work under controlled strain or under controlled stress (Barnes, 2000).

The most common measurement geometries used are the parallel plates, the cone and plate and the concentric cylinders. For the parallel plates and the cone and plate geometries, the two components are separated by a fixed distance, the bottom plate remains static and the other rotates with a certain angular velocity. On the other hand, in the concentric cylinders, the sample is placed in small gap between two cylinders, which are in relative motion. Usually the inner cylinder rotates at constant angular velocity while the outer cylinder is kept static. A detailed description of the different geometries and their advantages/disadvantages can be found in Barnes *et al.* (1989).

In this thesis, all fluids were characterized using a cone and plate geometry except the shear-thinning fluid employed in the macro-scale experiments which was characterized using parallel plates. These two types of arrangements were chosen to characterize the working fluids, even low-viscosity fluids, because of the larger shearing surface that these systems have when compared with the concentric cylinders available in the host research group laboratory.

In this section we focus on theoretical considerations and on the details of the measurement technique. A specific description of the measurements performed for each fluid used is presented in the corresponding chapter.

d) Cone and plate geometry

Figure 2.15 shows schematically the cone and plate geometry. In this arrangement, the cone is truncated at the center in order not to touch the plate. Small cone angles are important for accurate normal stress measurements and this arrangement ensures that the shear-rate remains constant everywhere in the sample. On the other hand, the smaller the cone angle, the greater the shear rate for a determined rotational speed. This geometry provides great precision and accuracy and is the favorite system for rheological measurements (Strivens and Schoff, 2010).

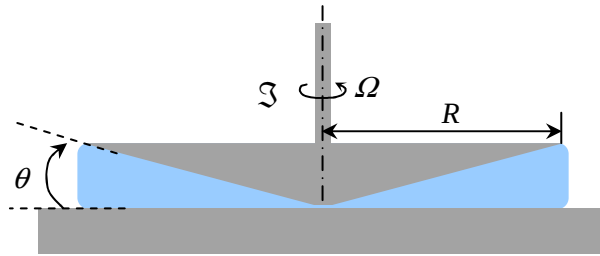


Figure 2.15. Schematic representation of the cone and plate geometry. The cone is rotating on top and the fixed plate is placed on bottom. The fluid sample is placed in the gap between them.

The shear-rate is constant throughout the sample and is given by (Barnes *et al.*, 1989):

$$\dot{\gamma} = \frac{\Omega}{\theta}, \quad (2.7)$$

where Ω is the angular velocity of the rotating cone with angle θ . The shear stress can be calculated from the torque necessary to apply on the cone (S) by:

$$\tau_{xy} = \eta(\dot{\gamma})\dot{\gamma} = \frac{3S}{2\pi R^3}. \quad (2.8)$$

For viscoelastic fluids a normal force, F , also arises and tends to separate the cone from the plate. Using the value of this force, it is possible to calculate the first normal stress difference or the first normal stress coefficient by relating it with the applied shear-rate (Barnes *et al.*, 1989):

$$N_1 = \Psi_1(\dot{\gamma})\dot{\gamma}^2 = \frac{2F}{\pi R^2}. \quad (2.9)$$

For high rotational velocities, negative normal stress effects appear which tend to pull the plates together. When this happens, the normal force measured is smaller than the true value and its reduction is given by (Barnes *et al.*, 1989):

$$\Delta F = \frac{3\pi\rho\Omega^2 R^4}{40}. \quad (2.10)$$

For all flow curves measured, the lower and upper limits that should be considered for accurate measurements of the shear viscosity were calculated. The lower boundary of the shear viscosity was determined using Eq. (2.11) and considering 20 times the minimum resolvable torque specifications of the shear rheometer:

$$\eta = \frac{3(20\mathfrak{S}_{\min})}{2\pi R^3} \frac{1}{\dot{\gamma}}, \quad (2.11)$$

where $\mathfrak{S}_{\min}$ is the rheometer minimum resolvable torque, which is equal to 1×10^{-7} N m for the shear rheometer used in the measurements.

The laminar flow of a Newtonian fluid between a rotating cone and a fixed plate is a purely rotational steady shear flow when the following criterion is observed (Sdougos *et al.*, 1984):

$$\frac{\Omega\theta^2\rho R^2}{12\eta} < 0.5. \quad (2.12)$$

Therefore, the upper limit of the measurable shear viscosity, based on the onset of inertial instabilities (Taylor vortices) is given by:

$$\eta = \frac{\Omega\theta^2\rho R^2}{6}. \quad (2.13)$$

e) Parallel plates geometry

The parallel plates arrangement is schematically shown in Figure 2.16. In the parallel plates geometry, the strain and shear-rate vary across the radius of the disk, from zero at the center to a maximum at the edge. In this case, the shear-rate is given by:

$$\dot{\gamma} = \frac{r \Omega}{h}, \quad (2.14)$$

where h is gap between the plates and r is the radial coordinate. The shear viscosity is given by:

$$\eta(\dot{\gamma}) = \frac{1}{\dot{\gamma}} \frac{3\mathfrak{I}}{2\pi R^3} \left(1 + \frac{1}{3} \frac{d \ln \mathfrak{I}}{d \ln \dot{\gamma}} \right), \quad (2.15)$$

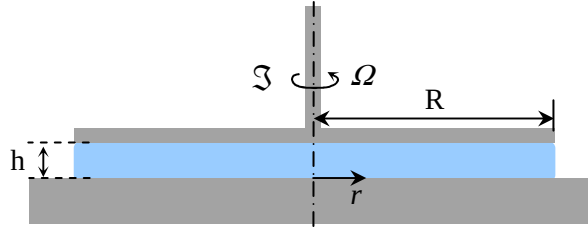


Figure 2.16. Schematic representation of the parallel plate geometry. The rotating plate is on top and the fixed plate on bottom. The fluid sample is placed in their gap.

In general, the second normal stress difference has a small value and therefore, measurements using a cone and plate as well as parallel plate geometries must be performed in order to obtain separated values of N_1 and N_2 . Using the normal force data, it is possible to calculate the normal stress difference (Barnes *et al.*, 1989):

$$N_1 - N_2 = \frac{2F}{\pi R^2} \left(1 + \frac{1}{2} \frac{d \ln F}{d \ln \dot{\gamma}} \right). \quad (2.16)$$

In this case, the laminar flow of a Newtonian fluid between a rotating and a fixed plate is a purely rotational steady shear flow when the following criterion is observed (Savins and Metzner, 1969):

$$\frac{\Omega \rho h^2}{12\eta} < 0.5. \quad (2.17)$$

Therefore, the upper limit of the measurable shear viscosity, based on the onset of inertial instabilities (Taylor vortices) is given by:

$$\eta = \frac{\Omega \rho h^2}{6}. \quad (2.18)$$

2.4.2.2. Dynamic measurements

The viscoelastic fluids used in the macroscopic measurements were also characterized in small-amplitude oscillatory shear (SAOS) flow. In contrast, the viscoelastic fluids utilized in microfluidic experiments were not characterized using these dynamic rheological experiments since the solutions are dilute and semi-dilute and the rheometer is not able to perform accurate measurements for such low viscosity/elasticity solutions. The geometries employed in the steady shear measurements were also employed in the SAOS measurements. Initially, the linear viscoelastic spectrum (range of amplitude values in which the shear and elastic modulus are constant) was determined by applying a fixed frequency. Then, measurements for a range of frequencies were performed so that dynamic properties were determined.

In SAOS experiments, the fluid sample is subject to a small sinusoidal oscillating strain or deformation (γ) as a function of time, according to:

$$\gamma(t) = \gamma_0 \sin(\omega t), \quad (2.19)$$

where γ_0 is the strain amplitude and ω is the angular frequency.

For a viscoelastic fluid, the resultant shear stress, $\tau_{xy}(t)$, is also sinusoidal but presents a phase lag when compared with the strain:

$$\tau_{xy}(t) = \tau_0 \sin(\omega t + \delta), \quad (2.20)$$

where δ is the phase angle, which covers the range of 0 to $\pi/2$ as the viscous component increases. Therefore, the response of the viscoelastic fluid to the applied strain is divided into two components: an elastic component in phase with the strain and a viscous component out of phase by an angle $\pi/2$. The trigonometric function presented in Eq. (2.20) can be expanded and the shear-stress becomes:

$$\tau_{xy}(t) = \tau_0 [\cos(\delta) \sin(\omega t) + \sin(\delta) \cos(\omega t)], \quad (2.21)$$

or,

$$\tau_{xy}(t) = \gamma_0 [G' \sin(\omega t) + G'' \cos(\omega t)], \quad (2.22)$$

where G' is the storage modulus and G'' is the loss modulus, which are defined as:

$$G' = \frac{\tau_0}{\gamma_0} \cos(\delta), \quad (2.23)$$

and

$$G'' = \frac{\tau_0}{\gamma_0} \sin(\delta). \quad (2.24)$$

The storage modulus expresses the magnitude of the energy that is stored in the material (elastic part) and the loss modulus the energy that is lost as viscous dissipation per cycle of deformation (Rao, 1999). For a Newtonian fluid, $G' = 0$ and the stress and strain are out of phase ($\delta = \pi/2$). For a perfectly elastic solid, all energy is stored, i.e. $G'' = 0$ and the stress and strain are in phase ($\delta = 0$).

Using complex variables, it is possible to define a complex modulus (G^*) and a dynamic viscosity (η^*),

$$|G^*| = \sqrt{(G')^2 + (G'')^2}, \quad (2.25)$$

$$\eta^* = \frac{G^*}{\omega} = \frac{G''}{\omega} - \left(\frac{G'}{\omega} \right) i, \quad (2.26)$$

where i is the imaginary constant ($i = \sqrt{-1}$).

2.4.3. Extensional rheometry

Material functions in extensional flow are also very important in order to describe the rheological behavior of the fluid in extensional flow. However, these are usually difficult to measure, especially for low-viscosity fluids for which it is difficult to generate homogeneous extensional flows.

Uniaxial extensional flow may be generated by grabbing a cylindrical sample of fluid on each end and pulling them apart as illustrated in Figure 2.17.

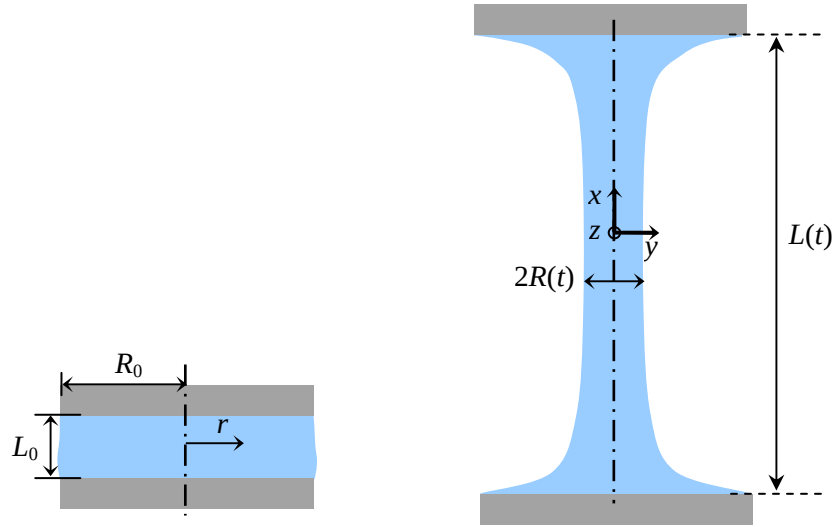


Figure 2.17. Schematic representation of a cylindrical filament being stretched in uniaxial elongational flow.

The velocity components can be expressed as,

$$u_x = \dot{\epsilon}x, u_y = -\frac{1}{2}\dot{\epsilon}y, u_z = -\frac{1}{2}\dot{\epsilon}z, \quad (2.27)$$

where $\dot{\epsilon}$ is the extensional strain rate ($\equiv \partial u / \partial x$), which is ideally a constant value. Since the sample has a cylindrical shape, the radial velocity component can be defined in terms of cylindrical coordinates:

$$u_r = -\frac{1}{2}\dot{\epsilon}r. \quad (2.28)$$

Considering that L is the cylindrical filament length and R is the radius of that filament, the extensional rate can be expressed as,

$$\dot{\epsilon} = \frac{1}{L} \frac{dL}{dt}. \quad (2.29)$$

Integrating from L_0 to L , Eq. (2.29) becomes

$$L = L_0 e^{\dot{\epsilon}t}. \quad (2.30)$$

and consequently, the total deformation experienced by the fluid, designed as Hencky strain is $\varepsilon_H = \dot{\varepsilon}t = \ln(L/L_0)$. Taking into account mass conservation as well as axial symmetry and integrating Eq. (2.29) from R_0 to R , the variation of the filament radius is given by

$$\frac{R}{R_0} = \exp\left(-\frac{\dot{\varepsilon}}{2}t\right). \quad (2.31)$$

If surface tension and gravity are neglected, the stress response to the elongation is defined as the normal stress difference, $\sigma_{xx} - \sigma_{rr}$, which equals to the ratio between the force applied on the sample, f , and the cross section of the filament, A , ($\sigma_{xx} - \sigma_{rr} = f/A$). Hence, the normal stress difference becomes:

$$\sigma_{xx} - \sigma_{rr} = \frac{fe^{\dot{\varepsilon}t}}{\pi R_0^2} = \dot{\varepsilon}\eta_E(\dot{\varepsilon}, t), \quad (2.32)$$

where η_E is the extensional viscosity. Using this material function and the shear viscosity at zero shear-rate, η_0 , it is possible to define the Trouton ratio, Tr , as:

$$Tr = \frac{\eta_E}{\eta_0}. \quad (2.33)$$

In the present investigation, a uniaxial extensional flow was applied to the samples of the working fluids in order to measure their relaxation time under extensional flow. For this purpose, a capillary-breakup extensional rheometer, CaBER (Haake CaBER 1, Thermo Scientific), was used. During CaBER measurements, the liquid is placed between two circular plates, which are separated by a distance h_i , and then an axial step strain is imposed in a short time (typically 50 ms) until a final height, h_f , is reached. Subsequently, the liquid thread is left to relax and breakup under the combined action of capillary and extensional viscoelastic forces. The diameter of the filament $D(t)$ is monitored as a function of time using the equipment's laser micrometer. Circular plates of $D_p = 6$ mm in diameter were used and the initial and final aspect ratios employed were varied according to the fluid used as will be explained in each chapter of results, with the objective of minimizing inertial effects that can be significant for low viscosity solutions and lead to erroneous measurements.

According to the findings of Entov and Hinch (1997) the variation of the filament diameter of a Newtonian fluid can be described as:

$$\frac{D(t)}{D_0} = \frac{\sigma}{6\eta D_0} (t_c - t), \quad (2.34)$$

while for a viscoelastic fluid,

$$\frac{D(t)}{D_0} = \left(\frac{GD_0}{\sigma} \right) e^{(-t/(3\lambda))}, \quad (2.35)$$

where t_c is the critical time for the Newtonian fluid breakup, σ is surface tension and G is elastic modulus of the viscoelastic fluid with relaxation time λ . For a Newtonian fluid, the filament diameter decreases linearly with time whereas for a viscoelastic fluid, this relationship is exponential. Thus, the relaxation time of the viscoelastic fluids used in this investigation were determined by fitting the experimental data in the linear region of $\log[D(t)]$ as a function of t . The relaxation time obtained from a CaBER experiment can be regarded as the characteristic time scale for viscoelastic stress growth in uniaxial elongational flow (Rodd *et al.*, 2006).

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PART I

Entry flow of viscoelastic fluids at macro-scale

THREE-DIMENSIONAL FLOW OF NEWTONIAN AND BOGER FLUIDS IN SQUARE-SQUARE CONTRACTIONS

Abstract

The flow of a Newtonian fluid and a Boger fluid through sudden square-square contractions was investigated experimentally aiming to characterize the flow and provide quantitative data for benchmarking in a complex three-dimensional flow. Visualizations of the flow patterns were undertaken using streak-line photography, detailed velocity field measurements were conducted using particle image velocimetry (PIV) and pressure drop measurements were performed in various geometries with different contraction ratios. For the Newtonian fluid, the experimental results are compared with numerical simulations performed using a finite-volume method, and excellent agreement is found for the range of Reynolds number tested ($Re_2 \leq 23$). For the viscoelastic case, recirculations are still present upstream of the contraction but we also observe other complex flow patterns that are dependent on contraction ratio (CR) and Deborah number (De_2) for the range of conditions studied: $CR = 2.4, 4, 8, 12$ and $De_2 \leq 150$. For low contraction ratios strong divergent flow is observed upstream of the contraction, whereas for high contraction ratios there is no upstream divergent flow, except in the vicinity of the re-entrant corner where a localized atypical divergent flow is observed. For all contraction ratios studied, at sufficiently high Deborah numbers, strong elastic vortex enhancement upstream of the contraction is observed, which leads to the onset of a periodic complex flow at higher flow rates. The vortices observed under steady flow are not closed, and fluid elasticity was found to modify the flow direction within the recirculations as compared to that found for Newtonian fluids. The entry pressure drop, quantified using a Couette correction, was found to increase with the Deborah number for the higher contraction ratios.

3.1. Introduction

The study of entry-flow problems, in which a fluid flowing through a duct of *large* cross-section progresses through a contraction into a smaller one, has been the subject of numerous studies with Newtonian and viscoelastic fluids (e.g. Boger, 1987; White *et al.*, 1987; Boger *et al.*, 1993; Nigen and Walters, 2002; Alves *et al.*, 2004; Oliveira *et al.*, 2007a). Besides its great usefulness for understanding a variety of real-flow phenomena that exists in many engineering applications, such as extrusion and injection molding processes, it is also a classic benchmark flow problem used in computational rheology (Hassager, 1988; Brown and McKinley, 1994). The majority of experimental and numerical works found in the literature on the subject of contraction flows focuses on axisymmetric and/or planar (or quasi-planar) arrangements. Planar flow configurations are particularly amenable for comparison with numerical predictions, since usually three-dimensional (3D) effects are negligible and experimental results can be adequately described using computationally less demanding two-dimensional (2D) simulations. Axisymmetric contractions are ubiquitous in real-flow problems, and under steady flow conditions are also less demanding than full 3D simulations. Numerical research on 3D sudden contractions is less frequent since these require much more powerful computational resources. However, in some cases of interest, e.g. in microfluidic contraction geometries, the flow is markedly three-dimensional due to reduced characteristic aspect ratios (Rodd *et al.*, 2006; Gulati *et al.*, 2008; Oliveira *et al.*, 2008). In such cases, a simple 2D approach is often qualitatively inadequate to describe the flow (Oliveira *et al.*, 2007b). Contractions with significant 3D effects are not only interesting from a fundamental point of view, but are also useful for validating 3D numerical codes. As such, the square-square geometry, in which the contraction occurs in two perpendicular directions, is a candidate for a reference 3D test-case, since it offers a good compromise between geometric simplicity and complex 3D flow structure.

Entry flows through square-square contractions (or analogous configurations) have been documented only in a handful of papers, which have focused mostly on the flow patterns (Walters and Webster, 1982; Xue *et al.*, 1998a, 1998b; Mompean and Deville, 1997, 2002; Sirakov *et al.*, 2005; Alves *et al.*, 2005, 2008). Some similarities between the flow through square-square and circular contractions were reported both in terms of the actual flow and in terms of the variation of the strain-rates (Walters and Rawlinson, 1982; Alves *et al.*, 2005, 2008). Most experimental and numerical studies typically report results concerning a 4:1 contraction

ratio (CR) in both directions (although $CR = 13.3$ has also been considered by Walters and Rawlinson, 1982). Indeed, this is motivated by the fact that the 4:1 contraction ratio was chosen as a benchmark test-case during the 5th International Workshop on Numerical Methods in non-Newtonian flows (Hassager, 1988). As a result, the flow patterns in this particular configuration are well documented for Newtonian and Boger fluids (Xue *et al.*, 1998a, 1998b; Alves *et al.*, 2005). The flow of viscoelastic fluids through this type of contraction presents a rich variety of flow structures spanning a number of regimes, such as lip vortex activity, vortex growth, diverging flow and unstable flow. Despite these advances, a comprehensive description of the flow in a square-square contraction, in which the effects of both geometric and rheological parameters are analyzed, is still lacking.

In this paper, we move toward this goal by presenting detailed experimental results for the flow of Newtonian and Boger fluids through square-square contractions with several different contraction ratios. Besides reporting global characteristics such as flow patterns and vortex length, as in previous studies, we also present particle image velocimetry (PIV) measurements, and pressure-drop measurements across the contraction. In addition we study the flow behavior in different planes of the geometry, in contrast with previous work in which only the center plane was analyzed (Alves *et al.*, 2005, 2008). Furthermore, we go beyond the previously studied 4:1 contraction and examine the effect of contraction ratio, defined as the ratio between the width of the large duct and the width of the smaller duct, on the flow characteristics for a wide range of Reynolds and Deborah numbers. The contraction ratio is known to have a major influence on the viscoelastic flow characteristics in converging configurations, as predicted for axisymmetric (Oliveira *et al.*, 2007a) and planar (Alves *et al.*, 2004) contractions. Additionally, we present numerical results of 3D simulations for Newtonian fluids in the studied geometries.

In the following section the experimental set-up and methods are briefly described. Fluid characterization is discussed in Section 3.3. The numerical method and governing equations used in the numerical calculations are briefly outlined in Section 3.4. In Sections 3.5 and 3.6 we present and discuss in detail the experimental results for the Newtonian fluid and for the Boger fluid, respectively. Furthermore, the Newtonian results are compared with the numerical simulations. The final section of this paper summarizes the main conclusions of this work.

3.2. Experimental set-up and methods

3.2.1. Experimental set-up

Figure 3.1 shows a diagram of the experimental set-up. The key part of the experimental rig is a square column of length $L = 1.75$ m, which is composed of two parts: a fixed square duct, with width $2H_1 = 24.0$ mm, and an interchangeable duct of smaller cross-section which fits tightly inside the larger one. The inner width of the inset square channel can be set to $2H_2 = 10.0$ mm, 6.0 mm, 3.0 mm or 2.0 mm, defining 2.4:1, 4:1, 8:1 and 12:1 contraction ratios, respectively. The upstream and downstream sections are denoted by subscripts 1 and 2, respectively. The dashed lines in Figure 3.1 represent the air circulation lines (vacuum), which are used to promote the flow.

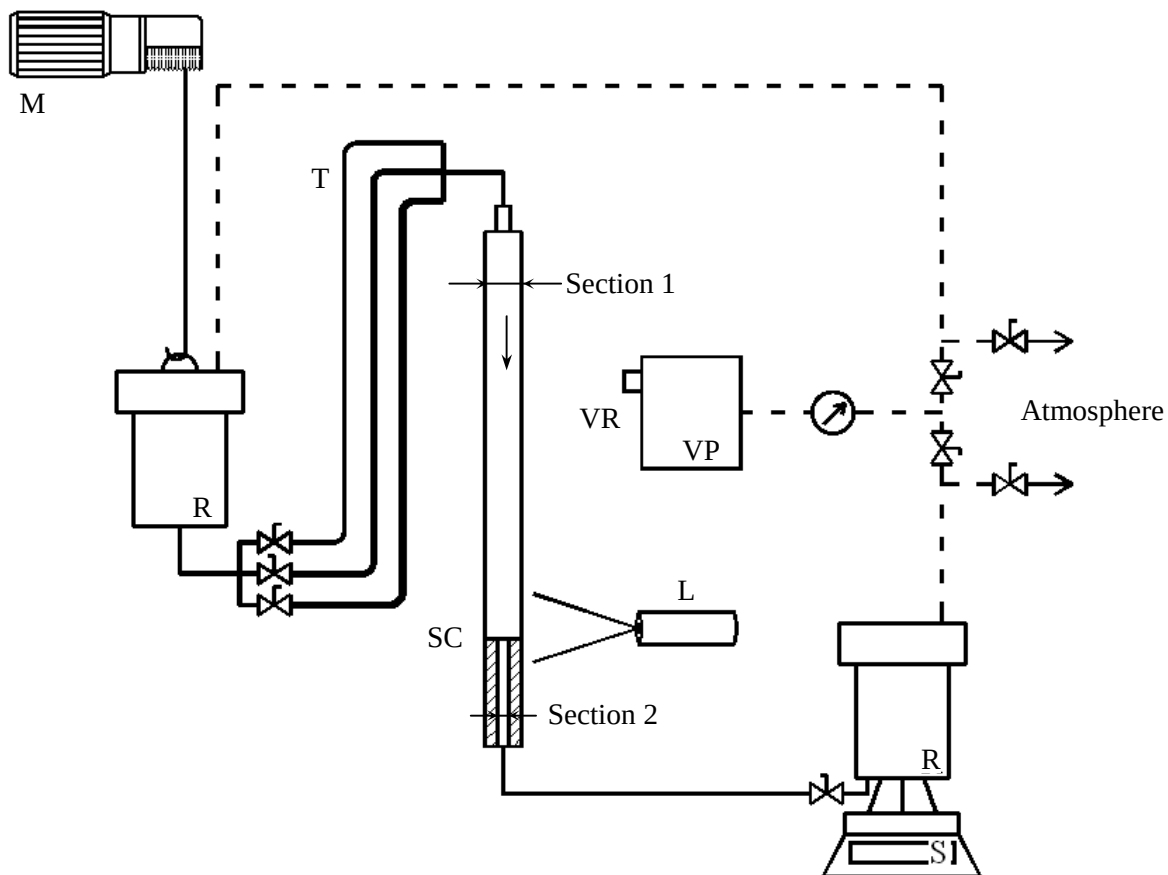


Figure 3.1. Experimental set-up. (L, Laser diode with cylindrical lens; M, Electric motor with winch; R, Reservoirs; S, Weight Scale; SC, Square contraction; T, Tubes; VP, Vacuum pump; VR, Vacuum regulator).

The left hand-side reservoir (Figure 3.1) is suspended in a steel cable connected to an electric motor, which allows the reservoir to be moved vertically by approximately 4 m. This reservoir is connected to the upstream duct by three parallel tubes of different diameters, while the second reservoir is located on top of a weighing scale (KERN DS 16k0.2; with readout of 0.2 g and maximum range of 16 kg) and is connected to the downstream duct by a 13 mm internal diameter tube.

The mass flow rate was set by adjusting the following parameters: the difference between the liquid levels in the reservoirs (attained by moving the vertical position of the left hand-side reservoir); the vacuum applied to the right hand-side reservoir using a vacuum pump (KNF Loboport N811-KT.18); and the diameter of the pipe that connects the reservoir to the duct (this diameter could be set to 4, 6 or 15 mm). The value of flow rate was monitored using LabView 7.1 that records the time and the mass of fluid in the right hand-side reservoir during the experiment. We note that we do not use any regulating valve to adjust the flow rate in order to avoid degradation of the fluid by passing through narrow orifices.

The experimental set-up was designed to provide a versatile mechanism that allows varying the contraction ratio, modifying the flow rate and adjusting the optical laser/camera arrangement in a simple and straight-forward way. For the purpose of flow visualization, the experimental set-up was made of transparent acrylic and the optical measurement system was adjusted to comply with the requirements of streak-line photography and PIV as described in sections 3.2.2.1 and 3.2.2.2, respectively. To allow for pressure drop measurements (detailed in section 3.2.2.3), four ports were positioned axially along the contraction geometry. One of the ports (the reference port, P_0) was located upstream of the contraction plane ($x = 0$), and the remaining ports (P_1 , P_2 and P_3) were located downstream of the contraction at positions that depend on the size of the smaller duct. Table 3.1 displays the precise port locations for each contraction ratio considered.

Table 3.1. Axial positions of the pressure ports (P_0 , P_1 , P_2 and P_3). The contraction plane is located at $x = 0$

	$CR = 2.4$	$CR = 4$	$CR = 8$	$CR = 12$
x_0 /mm	-152	-152	-153	-154
x_1 /mm	+99	+49	+30	+13
x_2 /mm	+149	+99	+48	+29
x_3 /mm	+249	+149	+98	+47

3.2.2. Experimental techniques

3.2.2.1. Streak-line photography

A detailed study of the flow patterns was carried out using long time exposure streak-line photography. The fluid was seeded with highly reflective PVC tracer particles $10\text{ }\mu\text{m}$ in diameter, at a concentration of approximately 30 ppm. The flow was continuously illuminated using a 635 nm 5 mW laser diode (Vector, model 5200-20) with a cylindrical lens incorporated in order to transform the spot laser beam into a light sheet that illuminates the plane under study. The streak-line images were captured using a digital camera (Canon EOS 30D) equipped with a macro lens (Canon EF100mm, f/2.8) placed perpendicularly to the laser light sheet. The exposure time was varied depending on the flow conditions, ranging from $O(1\text{ s})$ up to $O(10^3\text{ s})$.

Most streak-line images were taken at the center plane, i.e. mid-way between the front and the back wall of the duct section. In some cases, streak-line images were also obtained at different parallel planes, which required simultaneous adjustment of the light source and the camera. This was accomplished using a manual 2D traverse and a dial comparator with a relative positioning precision of $\pm 0.01\text{ mm}$.

3.2.2.2. Particle image velocimetry (PIV)

PIV was used to measure the velocity field in the vicinity of the contraction. The position of the optical components, such as the laser and the camera, is similar to that used for flow visualizations. However, PIV imaging requires very short exposure times compared with streak-line photography which typically uses exposure times on the order of seconds.

A doubled pulsed Nd:YAG laser, with a maximum energy of 50 mJ (Solo PIV III from New Wave Research), combined with appropriate optical components, produces a light sheet that illuminates the measurement plane. A digital CCD camera (Flow Sense 2M from Dantec Dynamics coupled with a Nikon AF Micro Nikkor 60 mm f/2.8D lens), running on double frame mode and positioned perpendicularly to the light sheet, was used to acquire the images. The time interval between each frame was adjusted according to the flow rate, the interrogation area, and the section of the flow being analyzed.

The velocity field was determined by processing the images with FlowManager v4.60 software, using a cross-correlation algorithm to generate a two-dimensional velocity vector map for each image pair. Subsequently, the velocity field is submitted to a validation process using a moving average criterion. This process is carried out for a minimum of 60 double images for each steady-state experiment and an averaged velocity field is determined. In order to increase the accuracy of PIV measurements when the variation of velocity throughout the region of interest is high, the flow region was divided in smaller sub-regions and PIV measurements were performed independently for each sub-region using an appropriate time between frames according to the local velocities. The full velocity field could then be reconstructed by compiling the data obtained for each sub-region. A thorough discussion of the PIV technique can be found in Keane and Adrian (1992) and Wereley and Meinhart (2001).

3.2.2.3. Pressure drop measurement

The pressure drop across the contraction region was measured using Honeywell 26PC differential pressure sensors (26PCA FA6D) which can cover a differential pressure range of ± 6.9 kPa. Each sensor is connected to the reference port P_0 , located upstream of the contraction plane, and one of the following ports located downstream of the contraction: P_1 , P_2 or P_3 (cf. Table 3.1). For the steady-state experiments, the transient output of the sensors was recorded until the steady value of pressure drop was reached. The data was acquired through a data acquisition card (NI USB-6218, National Instruments) using LabView v7.1. Prior to use, the sensors were calibrated using a static column of water for the whole differential pressure range.

3.3. Fluid characterization

In the experiments we used either a Newtonian fluid or a Boger fluid. A summary of the composition of the fluids is given in Table 3.2. The fluids were characterized using an MCR301 shear rheometer (Anton Paar), and the density (ρ) was measured using an hydrometer (readability of 0.001 kg/m^3 ; range $1200\text{-}1300 \text{ kg/m}^3$). Details of the rheological measurements are given in Sections 3.3.1 and 3.3.2 for the Newtonian and the Boger fluid, respectively.

Table 3.2. Composition of fluids in mass concentrations and density.

Fluid	PAA [ppm]	Glycerin [%]	Water [%]	Kathon [ppm]	NaCl [%]	ρ^* [kg·m ⁻³]
Newtonian	-	84.84	15.16	25	-	1221
Boger	100	90.96	7.52	28	1.50	1249

*The density was measured at 293.2 K.

3.3.1. Newtonian Fluid

The Newtonian fluid used in this work was a solution of glycerol (84.8%) and water (15.2%). A biocide (Kathon LXE, Rohm and Haas) was added to the solution at a concentration of 25 ppm in order to minimize bacteriological growth, and thus prevent fluid degradation.

The temperature effect on the steady shear viscosity was measured using the shear rheometer with a cone-plate fixture (75 mm in diameter and 1° angle), for temperatures ranging from 288.2 K to 303.2 K. The results are shown in Figure 3.2 for the Newtonian fluid and comparison is made against a 85% aqueous solution of glycerol as reported by Sheely (1932).

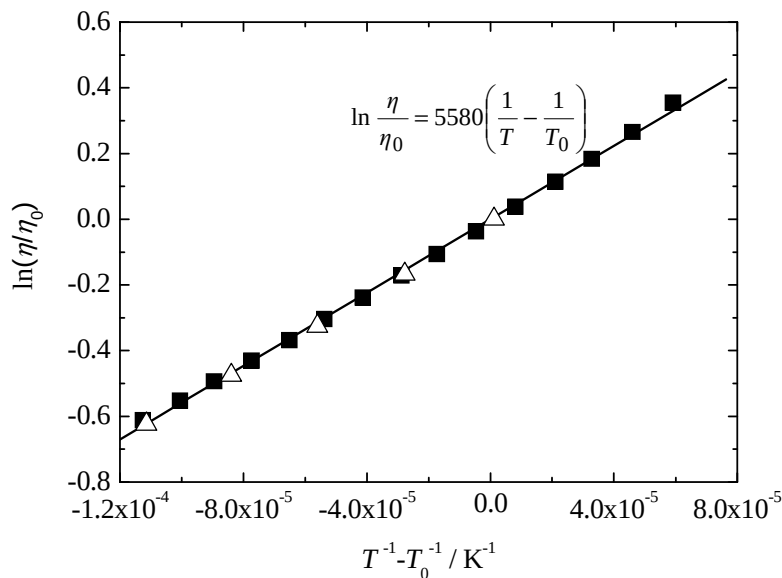


Figure 3.2. Temperature effect on the shear viscosity for the Newtonian fluid studied (■) and comparison with a 85 wt. % glycerol solution (△), reported in (Sheely, 1932) ($T_0 = 293.2$ K).

The dependency of the shear viscosity on the temperature can be properly described by an Arrhenius equation in the form:

$$\ln(a_T) = \frac{\Delta H}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right), \quad (3.1)$$

where ΔH represents the activation energy for flow, T_0 the reference absolute temperature (293.2 K), R the universal gas constant and a_T the shift factor, usually defined as (Bird *et al.*, 1987):

$$a_T = \frac{\eta(T)}{\eta(T_0)} \frac{T_0}{T} \frac{\rho_0}{\rho}, \quad (3.2)$$

in which $\eta(T_0)$ is the shear viscosity at the reference temperature T_0 , $\eta(T)$ is the shear viscosity at a given temperature T , while ρ_0 and ρ are the fluid densities at the reference temperature and at temperature T , respectively. For the range of measurements, the temperature variation is limited and small enough to consider that the fluid density and the ratio T_0/T do not change significantly. Thus, the shift factor becomes (Bird *et al.*, 1987):

$$a_T = \frac{\eta(T)}{\eta(T_0)}. \quad (3.3)$$

A fit of the experimental data to the Arrhenius equation (also shown in Figure 3.2) demonstrates quantitatively the dependency of the shear viscosity with the temperature for the Newtonian fluid. The shear viscosity for the Newtonian fluid at the reference temperature is $\eta(T_0) = 0.0982$ Pa s and $\Delta H/R = 5580$ K.

3.3.2. Boger Fluid

The Boger fluid was prepared by dissolving a small amount of PAA (100 ppm) into a Newtonian fluid of moderate viscosity. The shear viscosity (η) and the first normal stress difference coefficient (Ψ_1) were measured in steady shear flow, and the storage and loss moduli (G' , G'') in dynamic shear flow. A cone-plate geometry with 50 mm diameter and 2° angle was used.

The steady shear tests were carried out for temperatures in the range between 283.2 K and 303.2 K. The shift factors a_T were determined and $\Delta H/R$ was calculated to be 6780 K using 293.2 K as the reference temperature. Figure 3.3 displays the shear viscosity, η , as a function of the shear rate for the reference temperature, $T_0 = 293.2$ K. The shear viscosity of the Boger fluid

is approximately constant for the whole range of shear rates tested ($0.7 \text{ s}^{-1} \leq \dot{\gamma} \leq 100 \text{ s}^{-1}$). Also shown in this figure is the first normal-stress difference coefficient (Ψ_1). For a critical shear rate between 20 and 50 s^{-1} an elastic instability sets in which originates a strong increase in the shear and normal stresses (McKinley, 1991). Above this critical shear-rate the rheological measurements are not meaningful. We also show in Figure 3.3 the dynamic shear data measured under small amplitude oscillatory shear (SAOS) flow, namely the dynamic shear viscosity (η') and $2G'/\omega^2$ vs. ω . Since a single-mode model is not able to fit G' and η' accurately over the whole range of measured frequencies, instead a multi-mode model is used. The details of linear viscoelastic spectrum determined at $T_0 = 293.2 \text{ K}$ are listed in Table 3.3. The predictions using the three-mode model are overlapped with the experiments in Figure 3.3 (solid lines) and it is clear that, within the measured frequency range, a three-mode model is adequate to represent the linear viscoelastic behavior of the fluid.

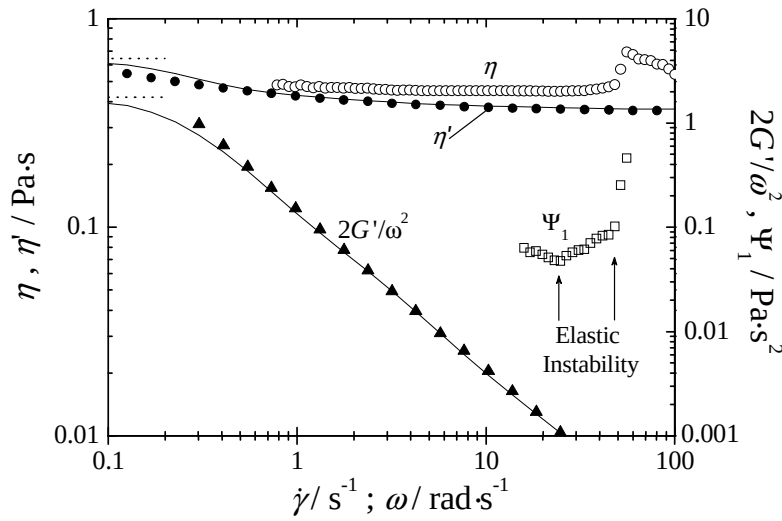


Figure 3.3. Material functions for Boger fluid at reference temperature (293.2 K) in steady (open symbols) and dynamic (solid symbols) shear flow.

The value of the solvent viscosity ($\eta_{\text{solvent}} = 0.367 \text{ Pa s}$) of the three-mode model was chosen to match the shear-viscosity of the N91 Newtonian fluid used by Alves *et al.* (2005), which has a composition equivalent to the Boger fluid used here except, obviously, the N91 fluid contains no PAA.

Table 3.3. Linear viscoelastic spectra for the Boger fluid at the reference temperature ($T_0 = 293.2$ K).

Mode k	λ_k / s	η_k / Pa s
1	4.0	0.225
2	0.4	0.04
3	0.04	0.014
Solvent	-	0.367

Using the parameters of the three-mode model presented in Table 3.3, an average relaxation time can be calculated as:

$$\lambda = \frac{1}{\eta_p} \sum_{k \neq \text{solvent}} \eta_k \lambda_k, \quad (3.4)$$

where $\eta_p = \sum_{k \neq \text{solvent}} \eta_k$, leading to $\lambda = 3.29$ s. This average relaxation time will be used throughout this work in the determination of the Deborah number.

3.4. Numerical method and governing equations

The governing equations for steady, laminar flow of an incompressible fluid are those expressing the conservation of mass and momentum (Bird *et al.*, 1987):

$$\nabla \cdot \mathbf{u} = 0, \quad (3.5)$$

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right] = -\nabla p + \nabla \cdot \boldsymbol{\tau}, \quad (3.6)$$

where $\mathbf{u}$ is the velocity vector, ρ the density of the fluid, t the time, p the pressure and $\boldsymbol{\tau}$ the extra stress tensor, defined as the sum of a Newtonian solvent and a polymeric solute contribution, ($\boldsymbol{\tau} = \boldsymbol{\tau}_s + \boldsymbol{\tau}_p$).

For Newtonian fluids, $\boldsymbol{\tau}_p = \mathbf{0}$ and the Newtonian solvent component $\boldsymbol{\tau}_s$ is expressed by:

$$\boldsymbol{\tau}_s = \eta_s (\nabla \mathbf{u} + \nabla \mathbf{u}^T) = 2\eta_s \mathbf{D}, \quad (3.7)$$

where η_s is the constant solvent viscosity.

The set of governing equations, i.e. the mass and momentum conservation given by Eqs. (3.5) and (3.6), together with the constitutive equation for Newtonian fluids (Eq. (3.7)) are solved numerically using a fully-implicit finite volume method as explained in detail by Oliveira *et al.* (1998).

The governing equations are discretized in time over a small time step (δt) and in space by integration over control volumes (CV) used to divide the computational flow domain. The time derivative is discretized with an implicit first-order Euler scheme, while the diffusive terms are discretized with central differences. For accurate discretization of the advective terms we use the CUBISTA high-resolution scheme (Alves *et al.*, 2003).

All numerical calculations were carried out using computational meshes representing the full wall-to-wall geometries. The overall dimensions of the square-square contractions are identical to those used experimentally (cf. Section 3.2.1), except the inlet and outlet planes that were located away from the region of interest (at $x/(2H_1) = -25$ and $x/(2H_1) = +25$, respectively), so that fully-developed flow conditions were enforced.

Table 3.4. Characteristics of the two meshes used for each contraction ratio studied.

	CR = 2.4		CR = 4		CR = 8	
	M40	M80	M40	M80	M40	M80
NC	82 000	656 000	51 000	408 000	83 300	652 800
$\Delta x_{min}/2H_1$	2.08×10^{-2}	1.03×10^{-2}	1.31×10^{-2}	6.25×10^{-3}	7.50×10^{-3}	3.75×10^{-3}
$\Delta y_{min}/2H_1 = \Delta z_{min}/2H_1$	1.99×10^{-2}	9.93×10^{-3}	1.25×10^{-2}	6.25×10^{-3}	7.50×10^{-3}	3.75×10^{-3}

The computational domain was structured in orthogonal blocks composed by non-uniform cells. For each contraction ratio studied (2.4:1, 4:1, 8:1), two meshes were used to map the square-square contraction in order to assess the accuracy of the numerical solutions. The coarse mesh (M40) has 40 cells in the largest duct along each of the two directions normal to the flow. The refined mesh (M80) has twice more cells in each of the three directions. Table 3.4

summarizes the characteristics of the meshes: the total number of cells (NC) and the dimensions of the smaller cell in the mesh, normalized by the width of the upstream duct, $2H_1$, for the range of contraction ratios studied.

Figure 3.4 shows a three-dimensional representation of the coarse mesh (M40) used to simulate the 4:1 contraction ratio geometry. Clustering of cells near the re-entrant corners (cf. Figure 3.4) was imposed in order to achieve higher accuracy in these regions, where the velocity and stress gradients are high. We have also included in Figure 3.4 some important variables and notation used throughout the text.

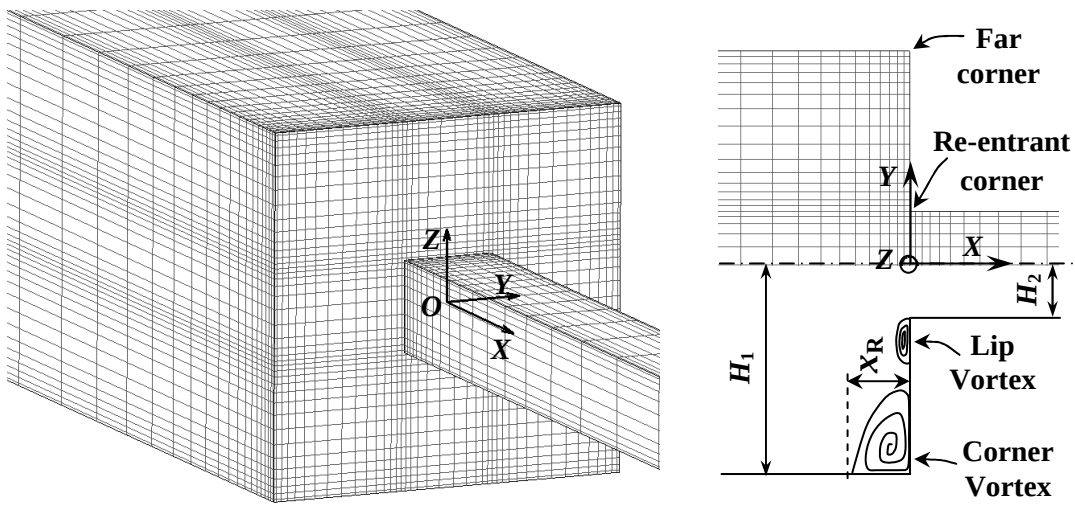


Figure 3.4. Mesh M40 used in the numerical simulations of the flow through 4:1 contraction and identification of the coordinate system.

3.5. Newtonian fluid flow

The study of the flow of a Newtonian fluid through 3D square-square contractions was carried out for three different contraction ratios: $CR = 2.4, 4$ and 8 . In this section we present the experimental and numerical results of flow patterns, vortex size and velocity field. The results were obtained experimentally using the Newtonian fluid and were simulated numerically using the computational meshes presented in Section 3.4.

3.5.1. Flow patterns and vortex length

The patterns generated by the Newtonian fluid flowing through the square-square contractions are presented in Figure 3.5. For each CR , the streak lines obtained experimentally and the corresponding numerical predictions are shown at the center plane ($y = 0$ or $z = 0$) for two different Reynolds numbers, here defined in terms of downstream variables:

$$Re_2 = \frac{\rho U_2 (2H_2)}{\eta}. \quad (3.8)$$

For the range of contraction ratios and Re_2 studied, we observe the formation of a vortical structure upstream of the contraction near the far corner, i.e. corner vortex (cf. schematics in Figure 3.4), which is in agreement with previous studies in square-square contractions (Xue *et al.*, 1998a,b; Alves *et al.*, 2005).

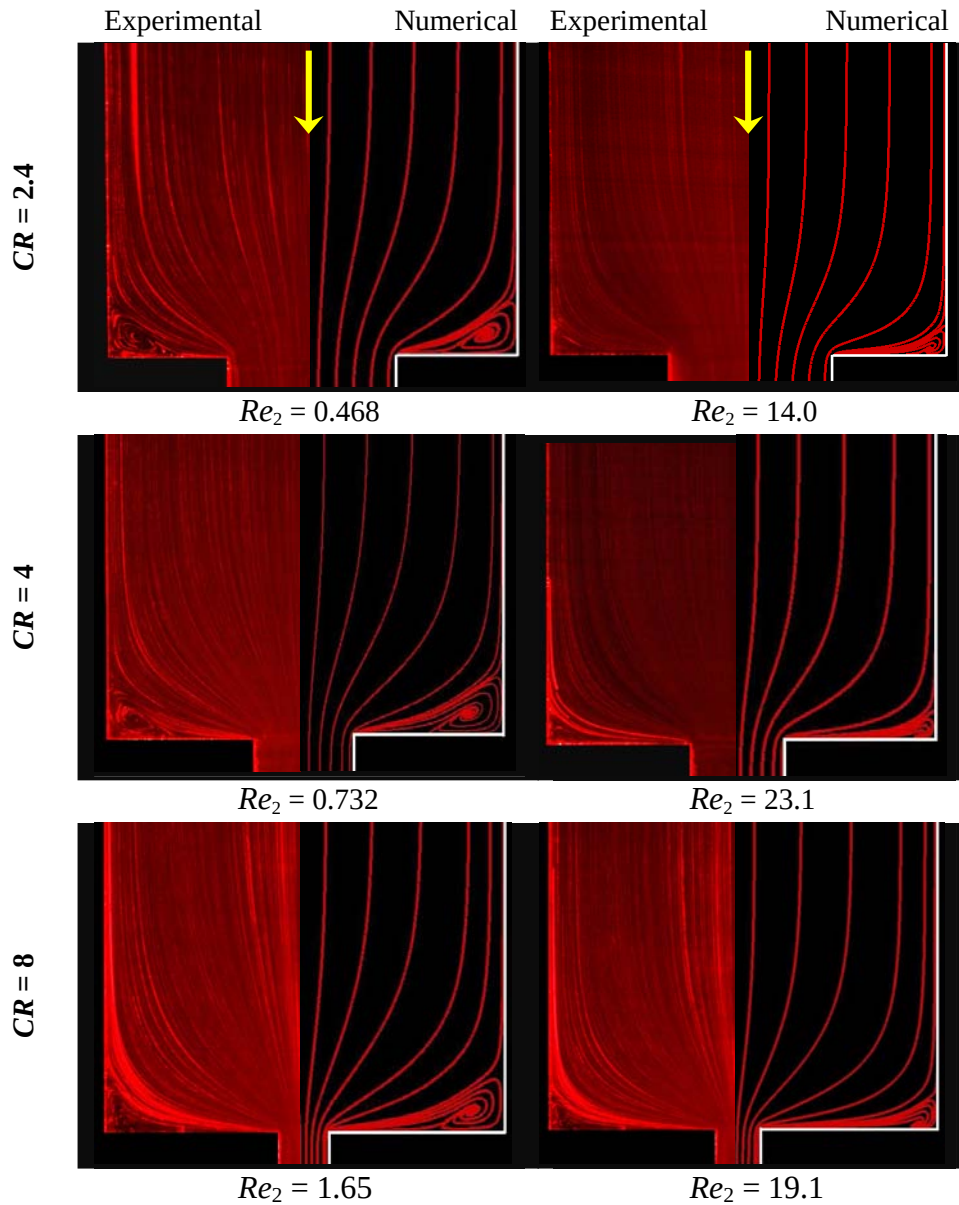


Figure 3.5. Experimental and numerical streak lines at the center plane of the square/square contraction for different contraction ratios and Reynolds numbers.

In Figure 3.6 we show the variation of the vortex length, x_R , defined as the distance between the contraction plane and the top of the recirculation (cf. sketch in Figure 3.4), with the Reynolds number for the three contraction ratios studied. The vortex length was measured at the center plane and its value was normalized using the width of the upstream duct, $2H_1$. Note that the characteristic dimension used in the Reynolds number is the width of the downstream channel, $2H_2$, which is different for each contraction ratio. We see that, for all CR , the experimental results are well predicted by the numerical simulations (cf. Figure 3.6).

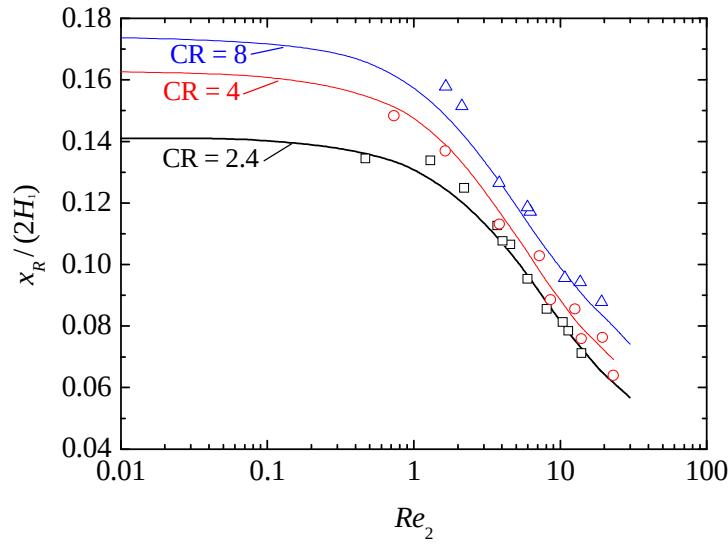


Figure 3.6. Effect of the Reynolds number on the vortex length at the center plane of the square/square contraction for $CR = 2.4$ ($\square$), $CR = 4$ ($\circ$) and $CR = 8$ ($\triangle$). Comparison with numerical calculations in mesh M80 (solid lines).

For low Reynolds numbers ($Re_2 \leq 0.5$), the dimensionless vortex length, $x_R/(2H_1)$, determined numerically asymptotes to a plateau which is slightly dependent on the CR : $x_R/(2H_1) = 0.142$ for $CR = 2.4$; $x_R/(2H_1) = 0.163$ for $CR = 4$ (in agreement with Alves *et al.*, 2005); $x_R/(2H_1) = 0.174$ for $CR = 8$. The dependence of the plateau value of the vortex length on the contraction ratio was found to be non-linear, analogously with the findings of Alves *et al.* (2004) and Oliveira *et al.* (2007a) for planar and axisymmetric contractions, respectively. However, under creeping flow conditions the value of $x_R/(2H_1)$ will become independent of CR for very high contraction ratios. For higher Reynolds number ($Re_2 \geq 0.5$), the length of the vortex decreases with inertia for the three contraction ratios investigated.

Even though the flow inside the corner vortex looks bi-dimensional, in reality it exhibits a complex behavior, showing a 3D open vortical structure upstream of the contraction. This type of open 3D recirculation has been observed previously for a square-square contraction with $CR = 4$ by Alves *et al.* (2005) and for 3D planar expansions by Schreck *et al.* (2000) and Oliveira *et al.* (2008). To better illustrate the flow inside the vortical structure, streak-line images were taken at different planes. Figure 3.7 shows the experimental and numerical streak-line projections in four different planes at several locations between the center plane and the wall plane for $CR = 8$. Due to the three-dimensionality of the flow within the contraction, the streak-line images obtained experimentally, with the exception of that at center plane, correspond to the projected path lines of the PVC particles on the illuminated plane. Additionally, in Figure 3.8 we compare the streak lines at the center plane and at the diagonal plane predicted numerically.

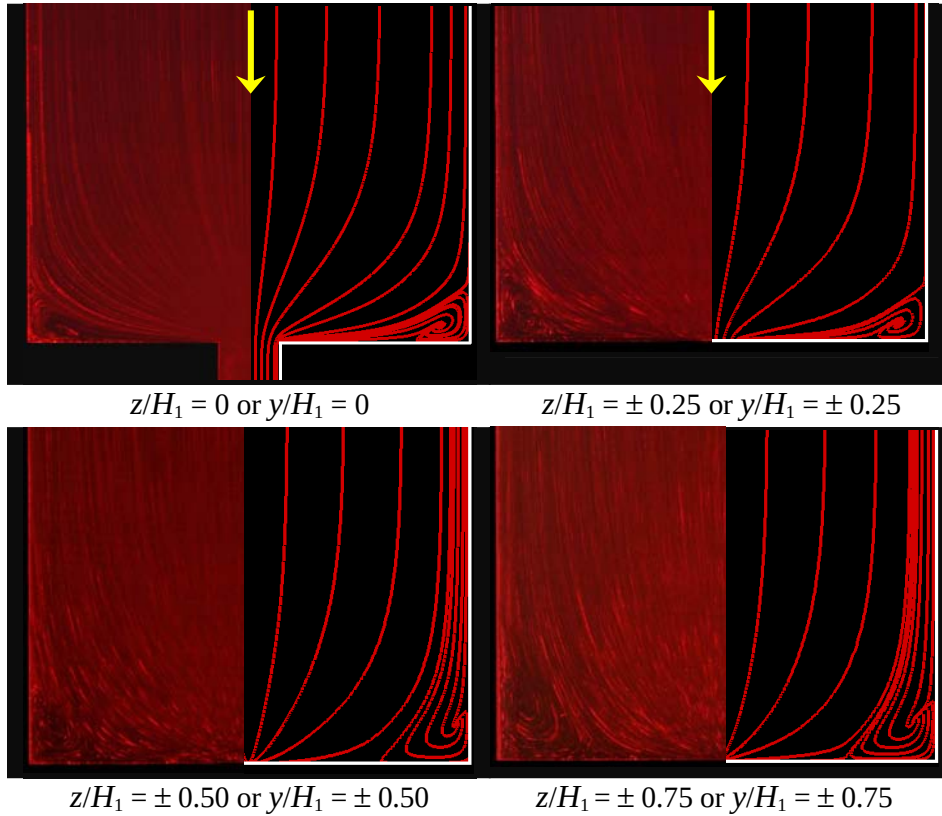


Figure 3.7. Experimental (left side) and numerical (right side) projected streak lines at different planes, for $Re_2 = 3.82$ of the 8:1 square/square contraction.

The streak-line projections in a number of planes demonstrate that the particles in the upstream square duct enter the flow recirculation at the diagonal plane and exit at the center plane close to the re-entrant corner where they proceed into the downstream duct. The structure of these recirculations is open and three-dimensional, similar to that observed experimentally and

predicted numerically for Newtonian fluid flows through 3D sudden expansions (Chiang *et al.*, 2000; Schreck *et al.*, 2000; Oliveira *et al.* 2008). Furthermore, the flow is symmetric relative to the two center planes ($y = 0$ and $z = 0$) and to the two diagonal planes ($y = \pm z$) due to the symmetry of the geometry. Figure 3.8(c) (adapted from Alves *et al.*, 2005) shows an overview of the 3D particle trajectories of a Newtonian fluid under low Reynolds number flow conditions. Under steady-state flow conditions, the recirculating vortex structures develop and the particles flowing close to the wall along the diagonal plane (ABCD) enter the corner vortex, rotate toward its center, and then drift to the center plane (EFGH). After rotating toward the vortex periphery at the center plane, the particles exit toward the downstream duct, near the re-entrant corner as described in detail by Alves *et al.* (2005).

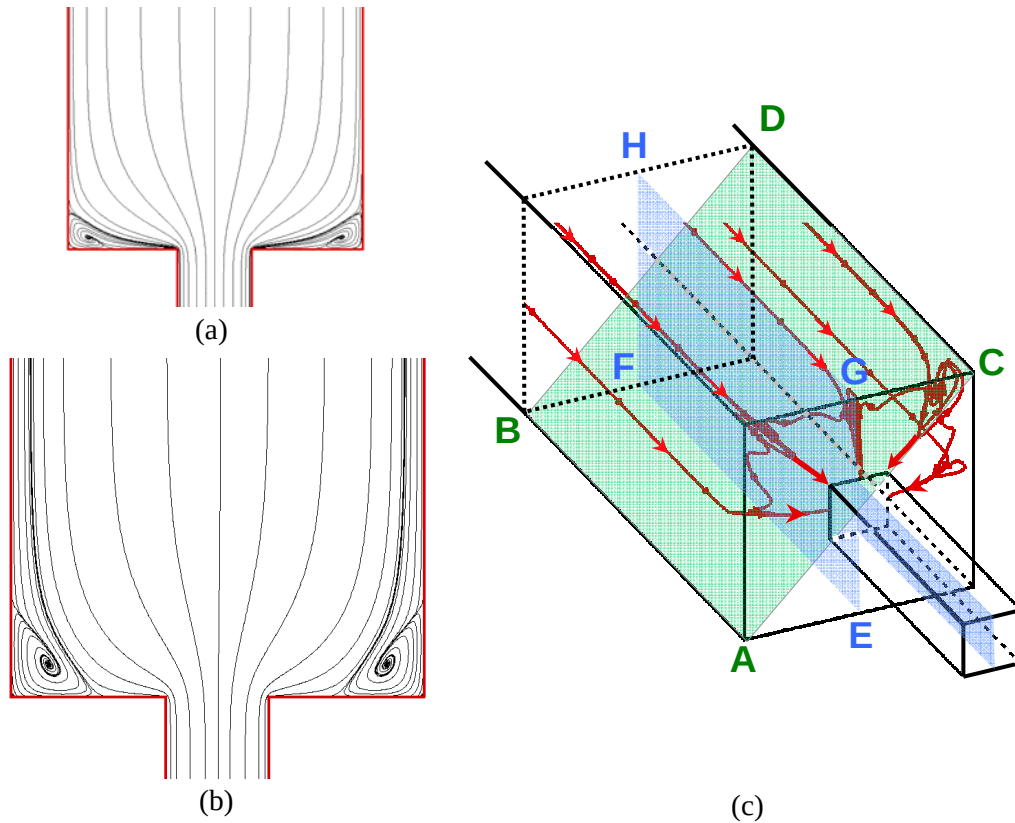


Figure 3.8. Numerically predicted streak lines for $Re_2 = 2.36$ at: (a) the center plane (EFGH) and (b) diagonal plane (ABCD). In (c) some path lines are illustrated to better demonstrate the three-dimensionality of the flow.

3.5.2. Velocity field

In Figure 3.9, we show the axial velocity profiles along the spanwise direction measured downstream and upstream of the 4:1 contraction. The position represented in the abscissa is normalized using the width of corresponding duct and the axial velocity was scaled with the

relevant average velocity. For clarity, we also show as inset a diagram of the location where the profiles were taken.

In the downstream channel close to the centerline ($y/(2H_1) = 0$ in Figure 3.9(a)), where the maximum velocity is reached, the coarse mesh (M40) describes the experimental results with a maximum relative error of approximately 5%, but the agreement is significantly improved by using the refined mesh (M80). In this case, the relative error between the numerical and experimental values becomes negligible, within the experimental error. In the upstream channel (Figure 3.9(b)), the average velocity is sixteen times smaller than that in the downstream channel. In this case, the two meshes used in the numerical calculations provide similar results, and the experimental velocity profile is well captured. Since the maximum relative error incurred when using the coarse mesh is small, mesh M40 might be useful when simulation time is a constraint, because the CPU time is substantially smaller than when using the M80 mesh. Note that the M80 mesh has eight times more cells than mesh M40 and therefore CPU times are about one order of magnitude higher. The small differences observed between the experimental (or numerical) results and the analytical solution in Figure 3.9(a) are due to the fact that at that axial location the fully-developed velocity profile is not yet observed since it is too close to the entrance of the contraction. This is evident in Figure 3.10(b), where we can observe that at $x/(2H_1) = 0.15$ ($x/(2H_2) = 0.60$) the centerline velocity is still below the fully-developed value.

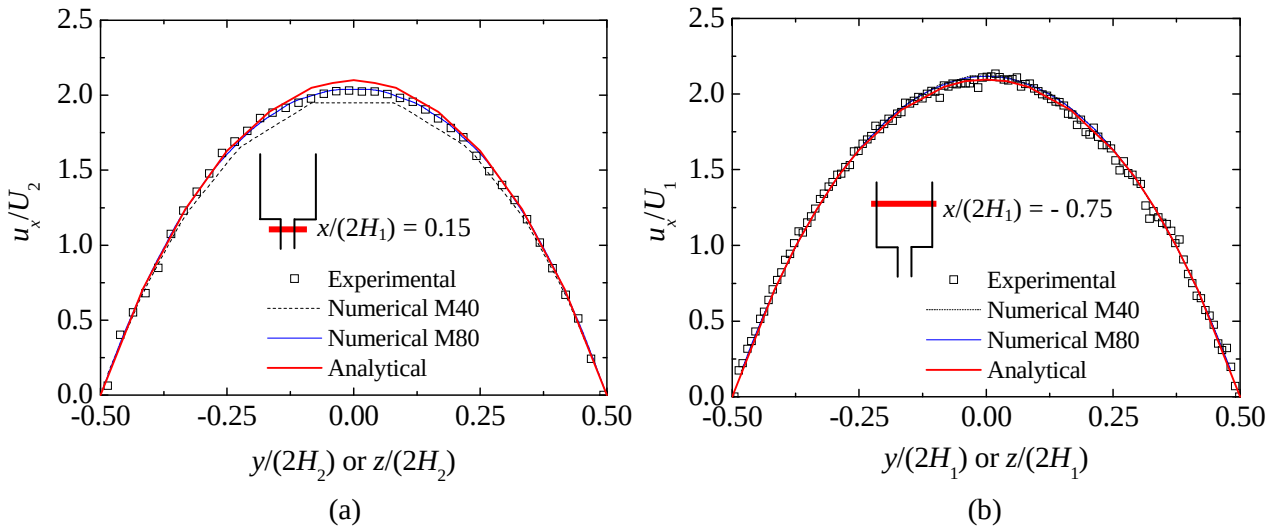
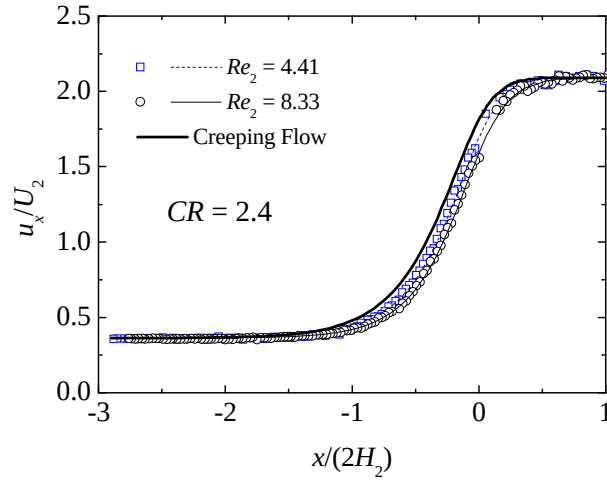
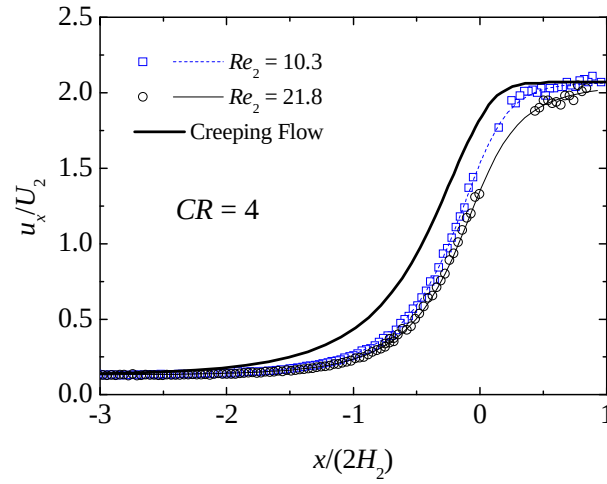


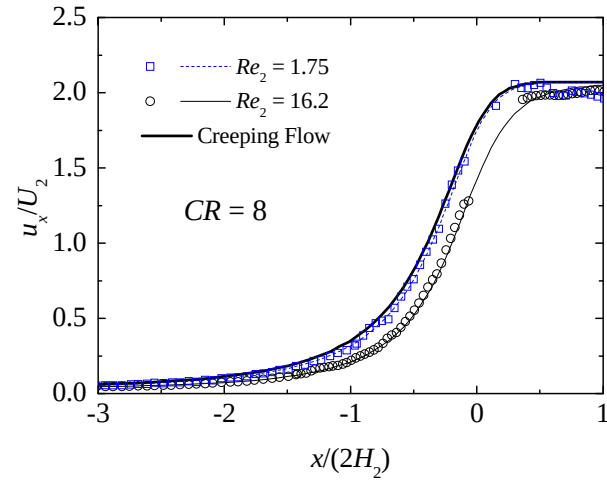
Figure 3.9. Axial velocity profiles at the center plane for $CR = 4$ and $Re_2 = 10.3$ at: (a) $x/(2H_1) = 0.15$ in the downstream channel and (b) $x/(2H_1) = -0.75$ in the upstream channel.



(a)



(b)



(c)

Figure 3.10. Experimental axial velocity profiles (symbols) along the centerline at the center plane for: (a) $CR = 2.4$, (b) $CR = 4$ and (c) $CR = 8$ and comparison with numerical predictions (lines) in mesh M80.

Figure 3.10 depicts the effect of inertia on the axial velocity profile along the centerline ($y = 0, z = 0$) for three values of CR . These experimental profiles display some scatter in the vicinity of the contraction plane, due to a shade that forms at the contraction plane when the laser light sheet illuminates the channel, which reduces the visibility of tracer particles, therefore leading to a reduction of the precision of the PIV results in this region of the flow. Nevertheless, we can see that the fluid accelerates as it approaches the contraction and an increase in Re_2 leads to a delay in the increase of the dimensionless velocity along the centerline and it takes longer for the flow to fully develop downstream of the contraction plane. The agreement between the experimental and numerical results is again very good for all CR .

The axial velocity profile along the streamwise direction for various lines $y = 0$ and $z = kH_1$ (or identically for $z = 0$ and $y = kH_1$) where $k = 0, 0.25, 0.50, 0.75$ and 0.92 , is shown in Figure 3.11.

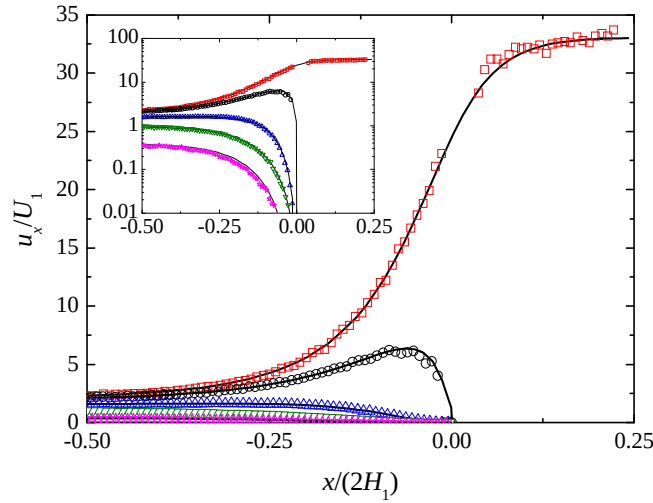


Figure 3.11. Experimental (symbols) and numerical (lines) profiles of the axial velocity for $CR = 4$ and $Re_2 = 10.3$ at: $y = 0$ and $z = \pm kH_1$ (or $z = 0$ and $y = \pm kH_1$), where ($\square$) $k = 0$; ($\circ$) $k = 0.25$; ($\triangle$) $k = 0.50$; (∇) $k = 0.75$ and ($*$) $k = 0.92$.

As expected, the magnitude of the axial velocity component increases as we move from the wall to the center plane. The three profiles closer to the wall show a velocity decrease along the streamwise direction as the fluid approaches the contraction region due to the presence of the contraction plane wall at $x = 0$. At the center plane, the fluid accelerates towards the contraction and the maximum strain rate is attained. For the plane $z/H_1 = 0.25$, which is coincident with the downstream channel wall, the profile displays an interesting behavior: there is a significant

acceleration toward the contraction, followed by an abrupt decrease as we move towards the wall at the contraction plane. The velocity profiles obtained at planes away from the center plane and in particular at the plane $z/H_1 = 0.25$ reveal the flow structure in the recirculation zones. Again good quantitative agreement between experiments and numerical simulations was found, thus demonstrating the good accuracy of the experimental technique, which will also be applied to the flow of Boger fluids in the next section.

3.6. Boger fluid flow

The study of the flow of a Boger fluid through 3D square-square contractions was carried out for four different contraction ratios, $CR = 2.4, 4, 8, 12$. The effect of Deborah number and contraction ratio on the flow patterns is examined in detail and, when appropriate, the corresponding vortex size is quantified. Furthermore, the velocity fields are measured at the center plane and at additional parallel planes closer to the wall. In this section we also present the experimental results of pressure drop across the contraction.

3.6.1. Flow patterns and velocity field

The effect of Deborah number on the flow patterns of the Boger fluid are illustrated in Figure 3.12-3.15 for a range of contraction ratios. The photographs were taken at the center plane using the streak-line photography technique described in Section 3.2.2.1.

To quantify the elastic effects we define the Deborah number based on downstream flow conditions and on the average relaxation time obtained by shear rheology as quantified in Section 3.3 ($\lambda = 3.29$ s), $De_2 = \lambda U_2/H_2$. Experimentally, De_2 was changed by varying the flow rate in the range $1.02 \times 10^{-8} \text{ m}^3/\text{s} \leq Q \leq 1.43 \times 10^{-5} \text{ m}^3/\text{s}$. Overall, fluid elasticity enhances the formation of complex structures upstream of the contraction and as the flow rate increases, a number of different flow type regions can be identified, such as: Newtonian-like flow; lip vortex activity, vortex growth and diverging flow; unstable flow.

At low Deborah number flows, viscous effects predominate and Newtonian-like patterns are observed, with concave-shaped corner vortices forming upstream of the contraction (Figure 3.12(a), 3.13(a), 3.14(a), 3.15(a)). Initially, as De_2 increases, the vortex shape and size do not seem to change significantly but as De_2 is increased further, elastic effects become significant.

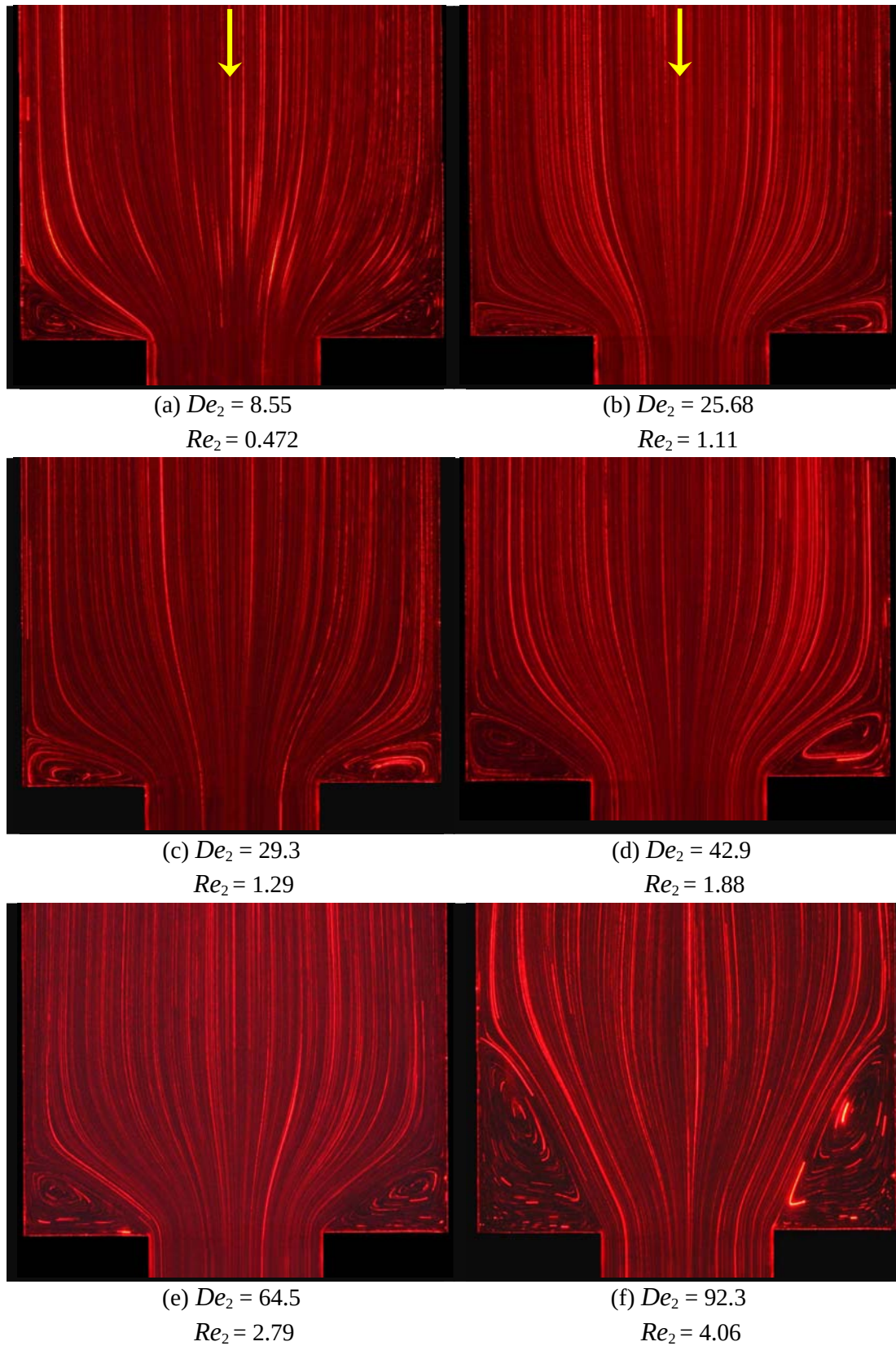


Figure 3.12. Effect of elasticity on the flow patterns at the center plane for $CR = 2.4$.

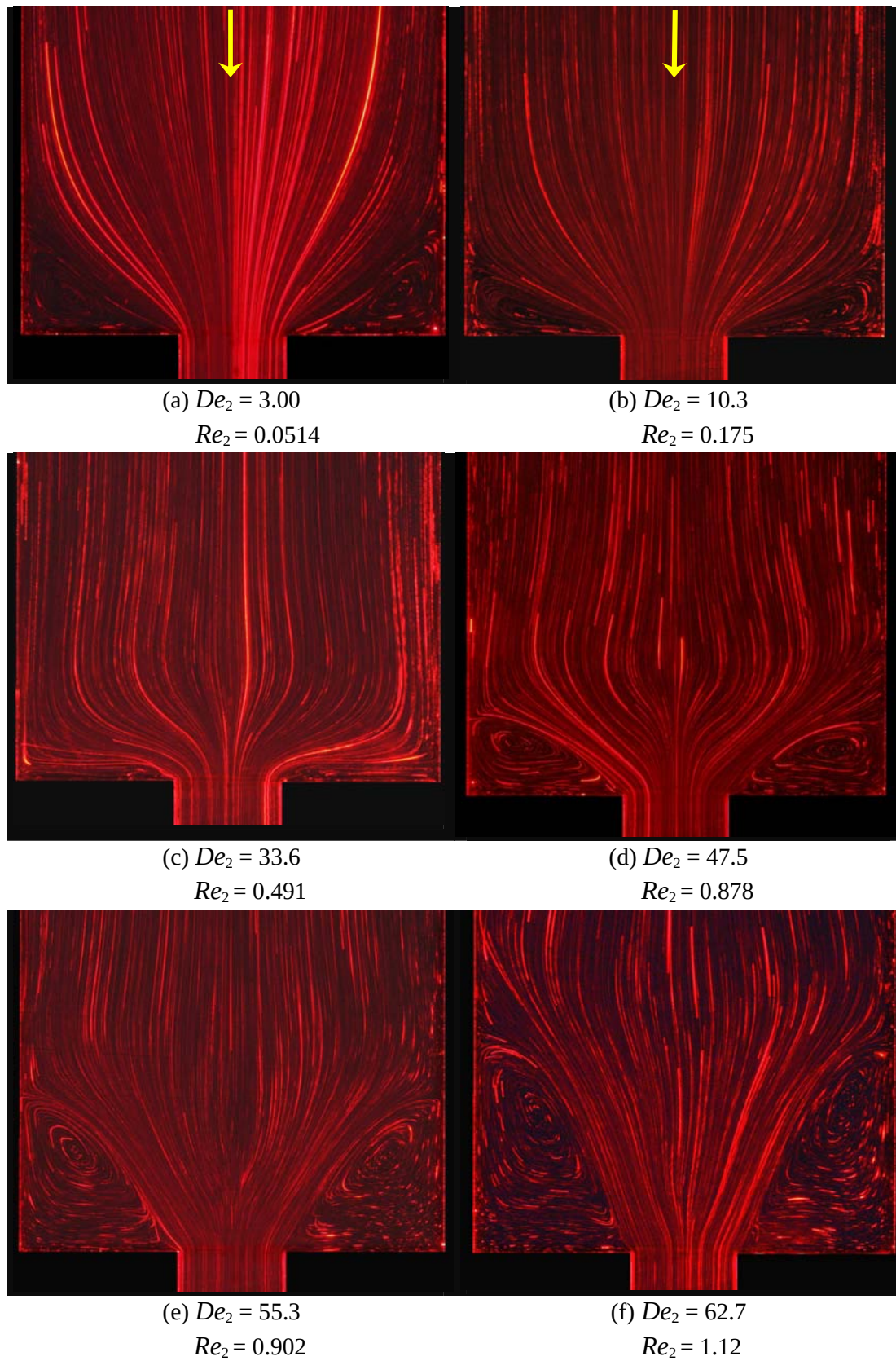


Figure 3.13. Effect of elasticity on the flow patterns at the center plane for $CR = 4$.

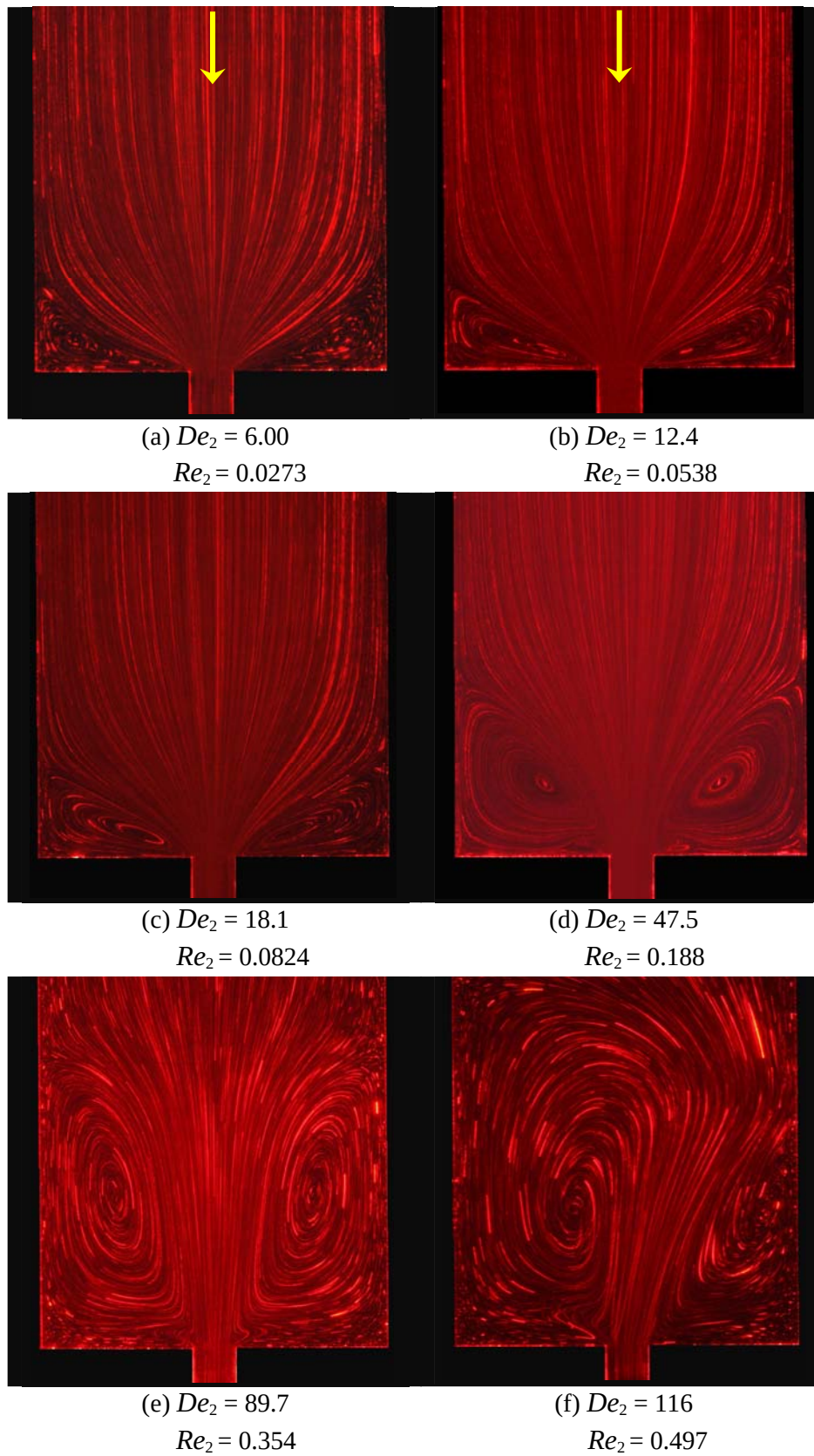


Figure 3.14. Effect of elasticity on the flow patterns at the center plane for $CR = 8$.

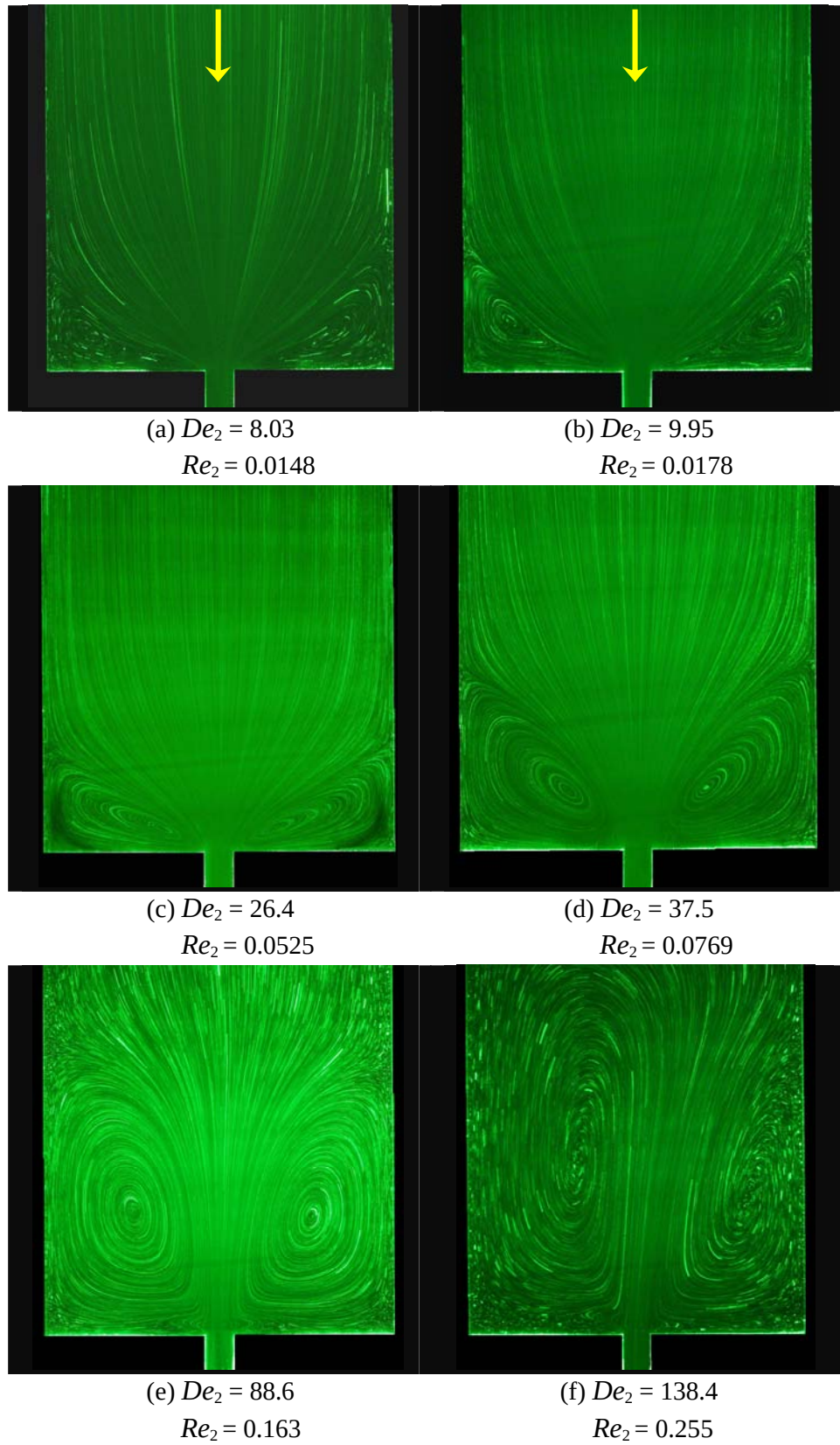


Figure 3.15. Effect of elasticity on the flow patterns at the center plane for $CR = 12$.

In addition to the corner vortex a weak lip vortex (cf. sketch in Figure 3.4) develops for $CR = 4, 8, 12$ at a Deborah number ($De_2 \sim 10$) which is approximately independent of the contraction ratio (Figure 3.13(b), 3.14(b), 3.15(b)). The formation of lip vortices close to the re-entrant corner has also been discussed by Alves *et al.* (2005) and Xue *et al.* (1998a, b) for flows of Boger fluids in 4:1 square-square contractions.

At high flow rates (Deborah numbers), elastic effects are enhanced and eventually lead to a change of the flow direction in the recirculations, i.e., the direction of fluid motion inside the vortices is the opposite of what is seen for lower flow rates (i.e. lower Deborah numbers) and for Newtonian fluids. This is observed for $De_2 \gtrsim 35, 15, 25, 30$ for $CR = 2.4, 4, 8, 12$, respectively.

In Figure 3.16, we show the streak-line images obtained at different planes by varying the distance from the center plane to the wall plane for $CR = 8$ and $De_2 = 67.9$. We can see that the tracer particles enter the corner vortex through the center plane contrary to the observations with the Newtonian fluid in which the particles enter the recirculation through the diagonal plane and exit through the central plane, as shown in Figure 3.8. The reversal of the flow pattern was not reported for the Boger fluid studied by Alves *et al.* (2005) but was observed and predicted numerically by Alves *et al.* (2008) for shear-thinning fluids. Therefore, we conclude that shear-thinning behavior is not necessary to produce reversal of the flow in the recirculation and that the flow inversion is due to strong elastic effects and in particular due to the high values of the extensional viscosity, as discussed by Alves *et al.* (2008).

The size of the corner-vortex structure is found to depend non-monotonically on the Deborah number, in resemblance with previous observations in 4:1 axisymmetric contractions (McKinley *et al.* 1991) and 4:1 square-square contractions (Alves *et al.*, 2005). Since in the region just upstream of the contraction the flow is highly extensional, we can define a Weissenberg number based on the relaxation time and on the estimated strain rate along the centerline, $Wi = \lambda \dot{\epsilon} \cong \lambda (U_{2,c} - U_{1,c}) / H_1 \cong 2.1 \left[(1 - 1/CR^2) / CR \right] \lambda U_2 / H_2$ (note that for a square channel the centerline velocity of a fluid with constant shear-viscosity is nearly 2.1 times the average velocity in the channel: $U_{2,c} / U_2 = U_{1,c} / U_1 \approx 2.1$), where $U_{1,c}$ and $U_{2,c}$ are the centerline velocities on the upstream and downstream channels, respectively. For square channels we can easily convert the Deborah number defined earlier in this section into a Weissenberg number: $Wi = 2.1 \left[(1 - 1/CR^2) / CR \right] De_2$.

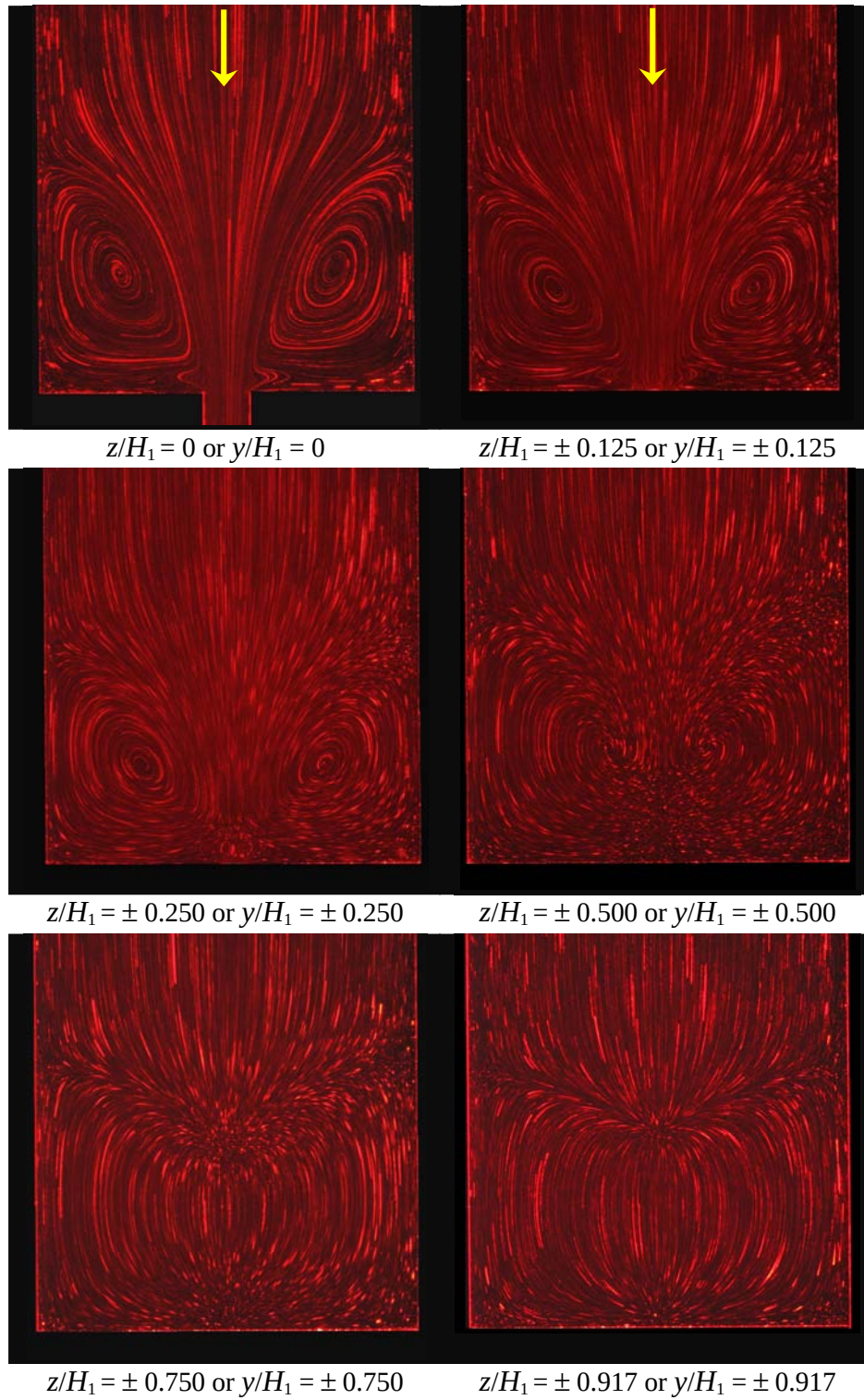


Figure 3.16. Streak line images at different planes of the square-square contraction for $De_2 = 7.9$, $Re_2 = 0.264$ and $CR = 8$.

The variation of the dimensionless vortex length (normalized with the upstream channel width, $2H_1$) as function of Wi is shown in Figure 3.17 for the range of contraction ratios tested. The vortex features observed in low ($CR = 2.4$ and 4) and high ($CR = 8, 12$) contraction ratio geometries are distinct.

The curves for high contraction ratios overlap when plotted as a function of Wi (or De_2/CR), in accordance with the scaling results of Alves *et al.* (2004) and Oliveira *et al.* (2007a) for planar and axisymmetric geometries, respectively. In these cases, De_2 enhances vortex growth and the flow features in the far corner area are only affected by upstream events (Oliveira *et al.*, 2007a). Thus, the behavior for different CR is similar and the curves coincide for high CR . For the low contraction ratios, the vortex growth regime is preceded by a decrease in vortex size until a minimum size is reached for $De_2 \sim 30$ (cf. inset in Figure 3.17). As this reduction takes place, the shape of the vortex changes becoming “convex” (cf. Figure 3.12(c), 3.12(d), 3.13(e)).

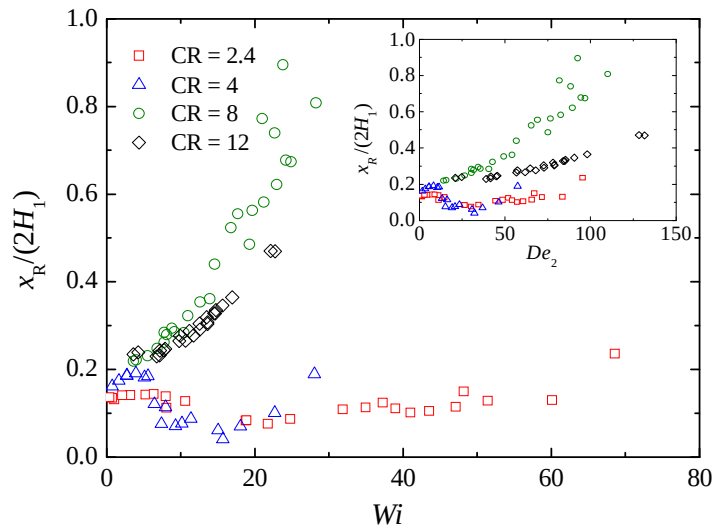


Figure 3.17. Normalized vortex length as function of Wi for all contraction ratios studied. In the inset, the normalized vortex length is presented as a function of Deborah number.

In the vortex growth regime, the flow behavior in low and high contraction ratio geometries is distinct as pointed out previously. For low contraction ratios ($CR = 2.4, 4$), an increase in the flow rate causes diverging streamlines to appear upstream of the contraction (cf. Figure 3.13(d)). This counterintuitive feature, which is related to the extensional behavior of the flow, has also been observed for viscoelastic fluids (both for shear thinning and Boger fluids) in converging flows with different geometric characteristics (e.g. Evans and Walters, 1986, 1989; Boger and

Binnington, 1990; Boger and Walters, 1993; McKinley *et al.*, 1991). The interested readers are referred to the work of Cable and Boger (Cable and Boger, 1978a, 1978b, 1979) for a comprehensive illustration of this flow regime and to the work of Alves and Poole (2007) for a discussion of this phenomenon and the conditions under which it arises. This diverging pattern of the flow streamlines is accompanied by a significant increase of the vortex length for $De_2 \gtrsim 30$ ($CR = 4$) and $De_2 \gtrsim 50$ ($CR = 2.4$) (cf. inset of Figure 3.17).

For higher contraction ratios ($CR = 8, 12$), we observe the development of a large lip vortex (as a result of the engulfment of the corner vortex by the lip vortex) with a convex shape that occupies the whole wall from the re-entrant corner to the far corner. The diverging streamlines seen for low CR are not observed here. However, concurrently with the vortex growth regime, the flow exhibits another remarkable feature ($De_2 \gtrsim 30$): the streak lines beneath the main vortical structure close to the re-entrant corner are locally strongly divergent and exhibit a bent-elbow shape (cf. Figure 3.14(e) and Figure 3.15(e)) almost as if the fluid had to detour around some small “invisible obstacle”. Such sharp divergent streamlines are not observed for lower CR , at least not for the range of flow conditions studied.

In Figure 3.18, we show a zoomed view of this flow feature at the center plane, which includes both results of flow visualization and PIV. Figure 3.18(a) and 3.18(b) show the velocity field in two different forms, as a contour plot and as a vector plot, respectively. The technique employed is able to capture the flow feature in detail within a large area of interest. To further attest the validity of the PIV measurements we compare the streak lines obtained by streak-line photography (Figure 3.18(c)) and those obtained from integration of the velocity field obtained using PIV (Figure 3.18(d)). Even though the streak lines obtained using the PIV technique are not as smooth near the contraction plane, good agreement is observed between the two techniques.

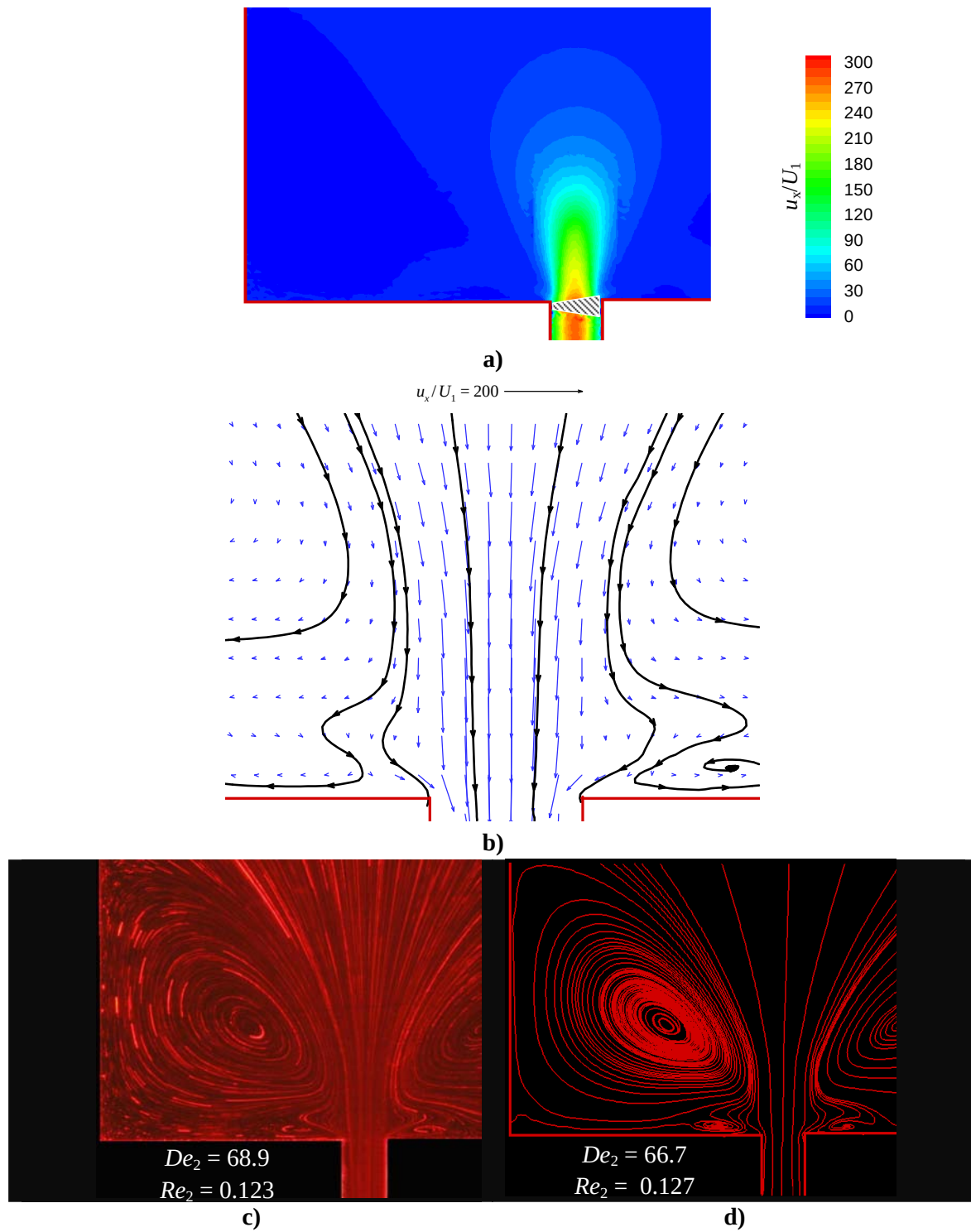


Figure 3.18. Normalized axial velocity contour plot (a) and vector map (b) for $De_2 = 66.7$, $Re_2 = 0.127$ and $CR = 12$ measured experimentally using PIV. Experimental streak line image (c) and streamlines obtained by integration of the velocity field (d) for the same flow conditions.

In Figure 3.19, we show the axial velocity profile along the streamwise direction in five different lines ($y = 0, z = kH_1$ or $z = 0, y = kH_1$ where $k = 0, 0.125, 0.25, 0.5, 0.75$) to highlight the sudden axial velocity increase as the fluid moves very close toward the contraction plane ($x/2H_1 \gtrsim -0.05$) for the planes $z/H_1 = 0.125$ and 0.25 (cf. inset Figure 3.19). This characteristic of the velocity profiles, which is not observed for Newtonian-like flow, is a consequence of the presence of localized diverging-converging streamlines in this region. In addition, we wish to highlight the three-dimensionality and symmetry of the flow under these conditions – the profiles for different z -planes shown here are in all similar to the profiles in the corresponding y -planes.

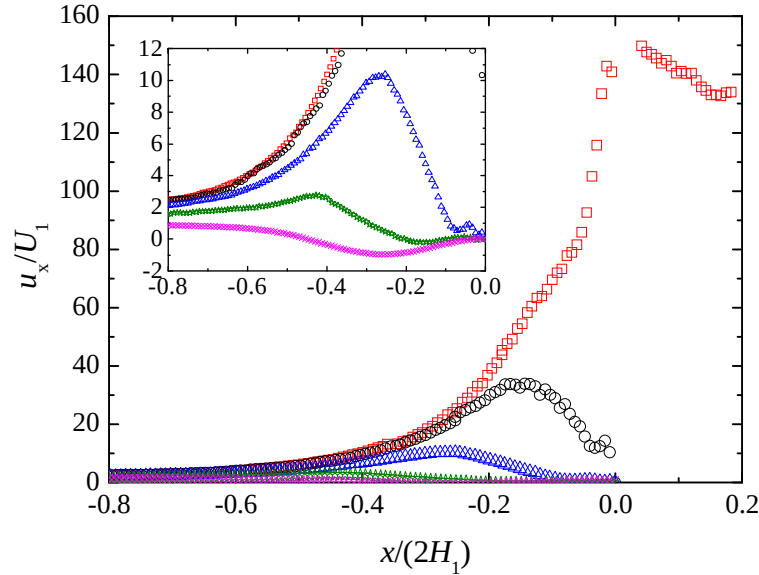


Figure 3.19. Axial velocity profiles along the streamwise direction for $De_2 = 62.3$, $Re_2 = 0.254$ and $CR = 8$, at: $y = 0$ and $z = \pm kH_1$ (or $z = 0$ and $y = \pm kH_1$), where ($\square$) $k = 0$; ($\circ$) $k = 0.125$; ($\triangle$) $k = 0.250$; ($\ast$) $k = 0.500$ and ($\times$) $k = 0.750$.

The effect of the Deborah number on the profiles of the streamwise velocity along the centerline ($y = 0, z = 0$) is shown in Figure 3.20 for the four contraction ratios studied. At low De_2 , the profiles resemble those obtained with a Newtonian fluid under creeping flow conditions. Upstream of the contraction, the fluid is seen to accelerate as it approaches the contraction ($x/2H_1 \gtrsim -0.2$) and the profiles may exhibit a velocity overshoot in the region of the contraction plane ($x/2H_1 \approx 0$), that increases significantly with increasing De_2 . For the lower contraction ratios ($CR = 2.4, 4$), when divergent streamlines are observed a velocity undershoot

upstream of the contraction ($-0.6 \lesssim x/2H_1 \lesssim 0$) is clearly visible, in agreement with the findings of Alves and Poole (2007) and Poole and Alves (2009).

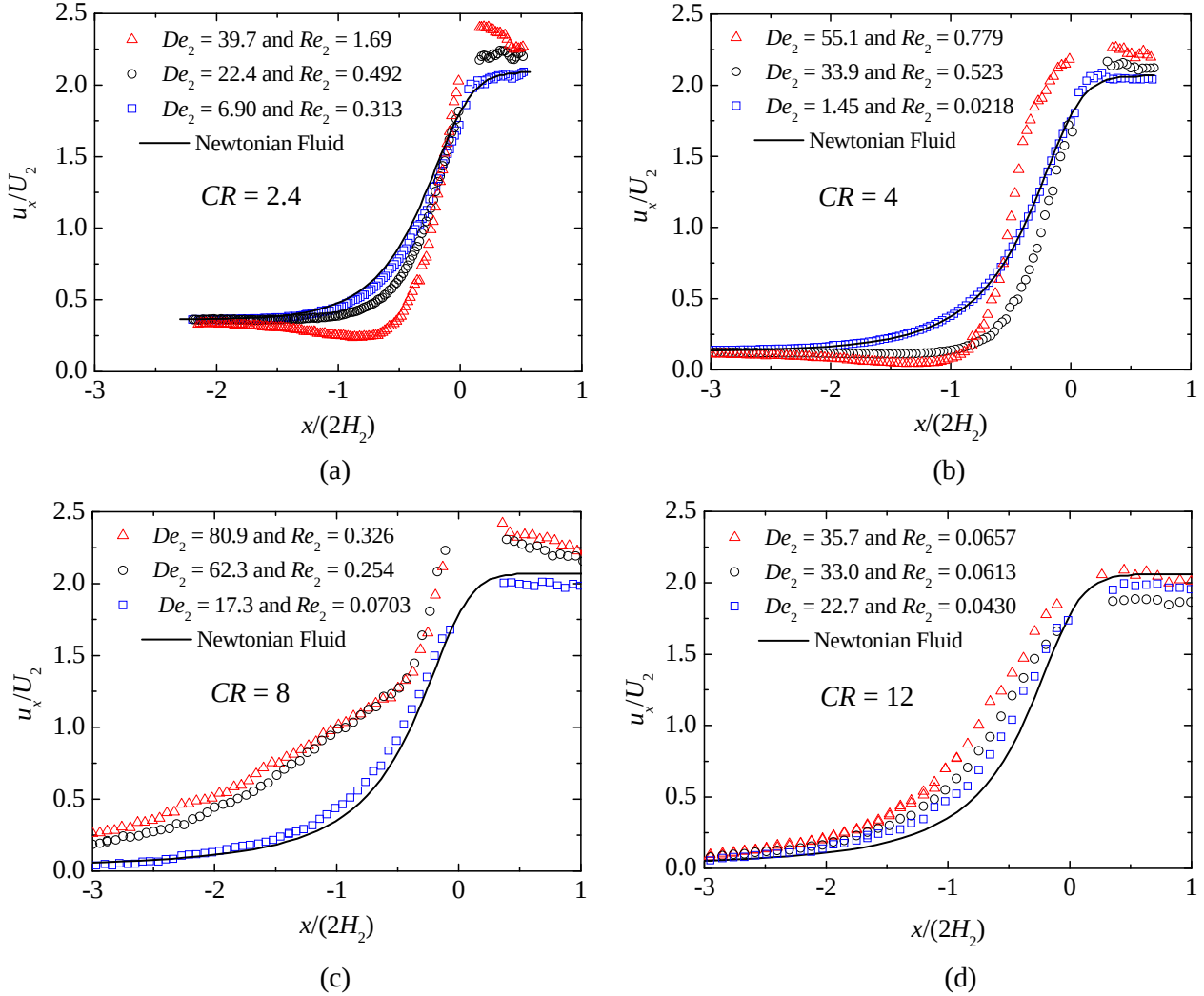


Figure 3.20. Axial velocity profiles along the centerline for (a) $CR = 2.4$, (b) $CR = 4$, (c) $CR = 8$ and (d) $CR = 12$. The symbols refer to PIV data, while the curves are from simulations for a Newtonian fluid under creeping conditions.

In Figure 3.20 we also include the velocity profile obtained numerically for the Newtonian fluid under creeping flow conditions to highlight that larger De_2 and lower CR strengthen the diverging flow and as a result the undershoot effect on the velocity profile is more pronounced.

As De_2 increases even further the flow eventually becomes unstable due to an elastic instability (cf. Figure 3.13(f), Figure 3.14(f), Figure 3.15(f)). In this regime, the flow is asymmetric and the vortical structures as a whole are rotating periodically in the azimuthal direction. Additionally, the degree of asymmetry is seen to increase with increasing Deborah

numbers. Figure 3.21(a) shows a sequence of streak-line images at the center plane within a cycle of flow oscillation. From films taken with a video camera at a known frame rate we were able to measure the frequency of oscillation, f , for different flow rates within the periodic regime. The normalized frequency of this process ($\lambda f / CR$) was found to be approximately independent of the Deborah number and of the contraction ratio for high CR , as shown in Figure 3.21(b).

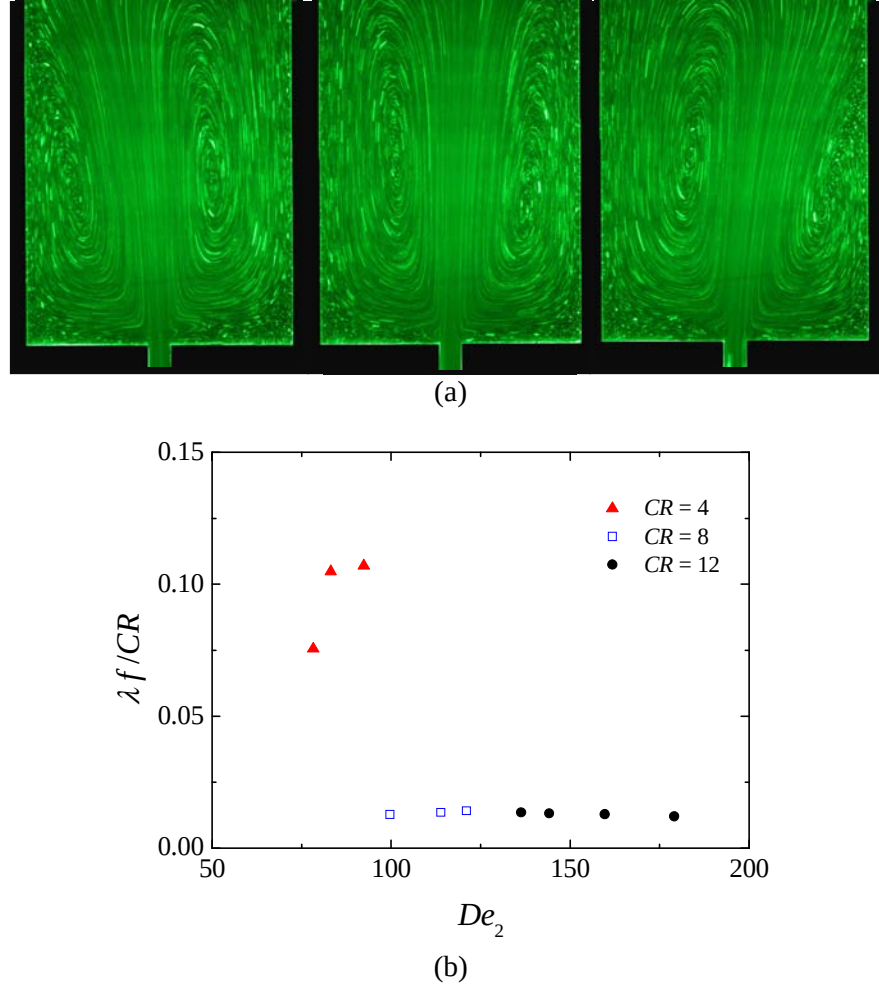


Figure 3.21. (a) Streak line images of the unstable flow taken at the center plane of the 12:1 square/square contraction for $De_2 = 138$ and $Re_2 = 0.255$; (b) normalized frequency of oscillation scaled as a function of the Deborah number for $CR = 4$, $CR = 8$ and $CR = 12$.

In this regime, the diverging streamlines with a bent-elbow shape are still present even though they are now skewed by the asymmetry of the flow (cf. Figure 3.21(a)). Analogously to the vortex size results, the frequency results for high CR are consistent with each other but are different from those obtained for lower CR ($CR = 4$).

To summarize, Figure 3.22 displays a map of flow patterns and their location on the two-dimensional $CR - De_2$ parameter space. This map is a result of extensive experiments for various contraction ratios and increasing Deborah numbers. The identified flow features are classified as: Newtonian-like flow (1) (Figure 3.12(a), 3.13(a), 3.14(a), 3.15(a)); vortex decrease (2) (Figure 3.12(b) and 3.13(c)); vortex growth and upstream diverging streamlines (3) (Figure 3.12(e), 3.12(f), 3.13(d), 3.13(e)); simultaneous lip and corner vortices (4) (Figure 3.13(b), 3.14(b), 3.15(b)); large lip vortex (5) (Figure 3.14(c), 3.15(c)); large vortex with localized diverging streamlines at the contraction entrance (6) (Figure 3.14(e), 3.15(e)); unstable flow (7) (Figure 3.13(f), 3.14(f), 3.15(f)).

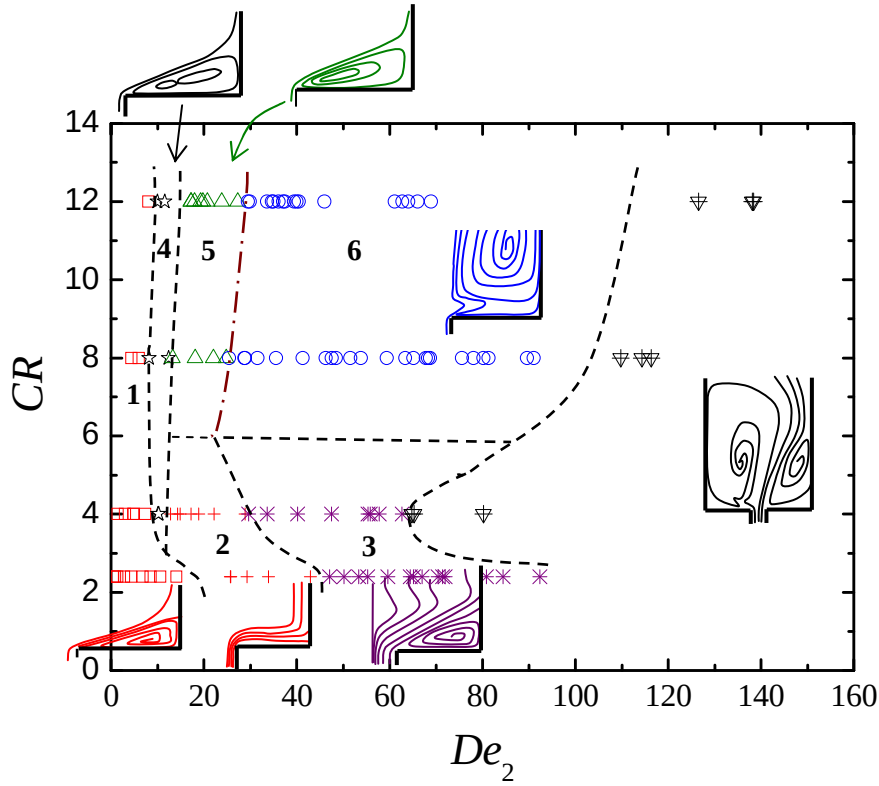


Figure 3.22. Flow pattern map. 1 - Newtonian-like flow; 2 - vortex decrease; 3 - vortex growth and upstream diverging streamlines; 4 - simultaneous lip and corner vortices; 5 - large lip vortex; 6 - large vortex and localized diverging streamlines at the contraction entrance and 7 - unstable flow.

3.6.2. Pressure drop

The pressure drop between the reference port upstream of the contraction and each of the downstream pressure ports were measured experimentally as a function of the flow rate. These results are shown in Figure 3.23 and can be useful for benchmarking purposes. The pressure drop vs. flow rate curves display similar characteristics for all CR studied, with the pressure drop

increasing non-linearly with the flow rate. Also shown in Figure 3.23 is the extra pressure drop (Δp_{extra}) caused by the extensional flow in the contraction. The extra pressure drop is usually associated with vortex enhancement in contraction flows and is calculated by subtracting the pressure drop due to fully developed (fd) Poiseuille flow in the duct upstream ($\Delta p_{\text{fd,u}}$) and downstream ($\Delta p_{\text{fd,d}}$) of the contraction from the total pressure drop (Δp_{Total}) across the contraction. In our case:

$$\Delta p_{\text{extra}} = \Delta p_{\text{Total}} - \Delta p_{\text{fd,u}} - \Delta p_{\text{fd,d}} = \Delta p_3 - \left| \frac{dp}{dx} \right|_{\text{fd,u}} |\Delta x_0| - \left| \frac{dp}{dx} \right|_{\text{fd,d}} |\Delta x_3|, \quad (3.9)$$

where the subscripts u and d refer to upstream and downstream ducts, respectively.

We have estimated dp/dx in two different ways, one considering the theoretical values of dp/dx calculated analytically for fully-developed Poiseuille flow (White, 2003) and the other using an experimental value of dp/dx determined from measurements at locations 2 and 3: $dp/dx = (\Delta p_3 - \Delta p_2) / (\Delta x_3 - \Delta x_2)$. For $CR = 2.4$ and 4, we observe a considerable discrepancy between the curves obtained using the two approaches referred above: while the experimental values of Δp_{extra} become negative, which is representative of a local pressure recovery as the fluid goes through the contraction, the estimated extra pressure drop from analytical values remain positive and increase with increasing flow rate. In fact, for such CR the extra pressure drop is very small compared to the total pressure drop and therefore a large degree of uncertainty is associated with the estimation of Δp_{extra} since the extra pressure drop is estimated from extrapolation of the ideal (fully-developed) pressure profile starting at a distant pressure location (to guarantee that fully developed conditions are observed). Additionally, at the locations considered for the measurements the flow might not yet be fully developed (cf. Figure 3.20(a), 20(b)). For higher contraction ratios ($CR = 8$ and 12), the differences between the two approaches diminish significantly and in particular for $CR = 12$ the results obtained using the two methods are nearly identical, an indication that for this CR the determination of the entry pressure drop has reasonable accuracy.

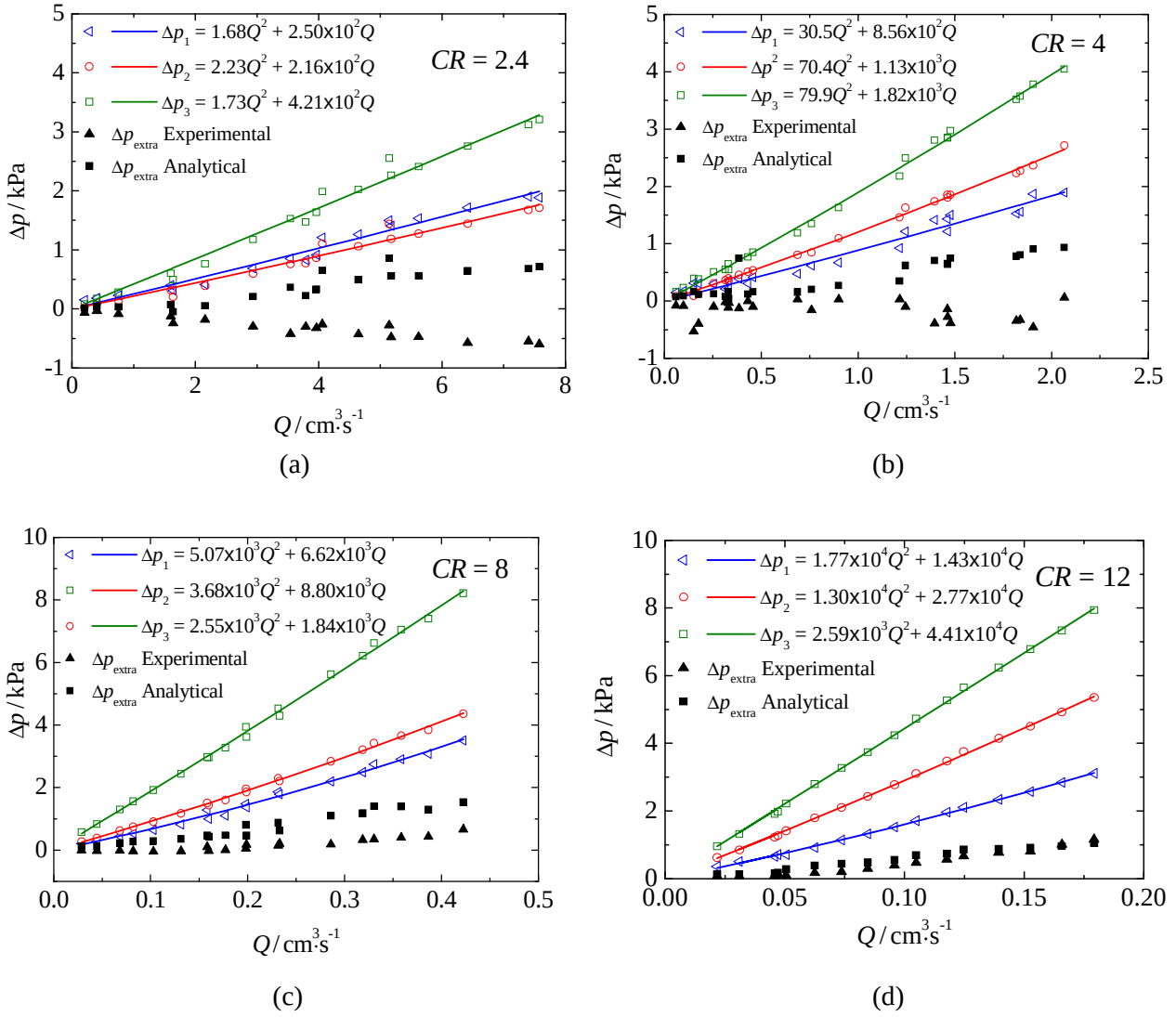


Figure 3.23. Pressure drop across the contraction measured with the Boger fluid for a range of flow rates and contraction ratios: (a) $CR = 2.4$; (b) $CR = 4$; (c) $CR = 8$ and (d) $CR = 12$. The open symbols represent the experimental data and the solid lines the corresponding fit. The extra pressure drop obtained from the experimental data and from analytical calculations (White, 2003) are shown as solid symbols.

The extra pressure drop is typically reported in dimensionless form as a Couette correction, defined as $C = \Delta p_{\text{extra}} / 2\tau_w$, where τ_w is the average downstream fully-developed wall shear stress, which for a square duct can be estimated as $(-dp/dx)_{\text{fd,d}} H_2/2$. The evolution of the Couette correction with the Deborah number in the range $60 < De_2 < 120$ is shown in Figure 3.24 for the higher contraction ratios studied ($CR = 8, 12$), illustrating a significant increase of the Couette correction with De_2 . Similarly to what was found for the vortex length, the results for $CR = 8$ and 12 are identical. We should emphasize that this parameter is difficult to estimate

experimentally since it requires the flow to be fully developed, the measurements have to be performed far downstream of the contraction and therefore any small error on the estimation of the slope $(dp/dx)_{fd,d}$ will have a strong impact on the extrapolated pressure values and consequently on the extra-pressure drop and Couette correction.

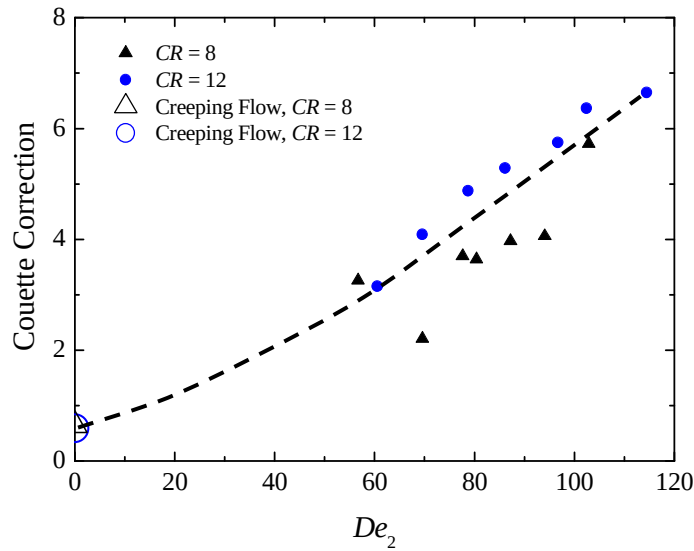


Figure 3.24. Couette correction as a function of Deborah number for $CR = 8$ and $CR = 12$. Also shown are the values of the Couette correction obtained numerically for a Newtonian fluid under creeping conditions ($De_2 \rightarrow 0$). The dashed curve is a guide to the eye.

3.7. Conclusions

The flow through sudden square-square contractions was characterized experimentally using streak-line photography, pressure drop measurements and PIV, which can be valuable for benchmarking purposes. Two fluids with different rheological properties were used: a Newtonian fluid and a Boger fluid. Additionally, the effect of the contraction ratio was investigated ($CR = 2.4, 4, 8, 12$). For the Newtonian fluid, in addition to the experimental measurements, numerical simulations were carried out using a finite volume code. The experimental results of flow pattern and velocity field were found to be in good agreement with the numerical results for the whole range of conditions tested, validating the experimental techniques.

By taking advantage of the visualizations of the flow patterns and quantification of the velocity field at various planes along the spanwise directions (y or z) covering the whole distance from the wall to the center plane, we have shown that the flow is highly three-dimensional with

open vortex structures forming upstream of the contraction. Furthermore, we have demonstrated that the dynamics inside the open recirculations for the Boger fluid at high Deborah numbers are reversed relative to Newtonian fluid flow. For the viscoelastic fluid, tracer particles that enter the recirculations from the upstream duct at the center plane exit at the diagonal plane towards the downstream duct while for the Newtonian fluid, the opposite process takes place with fluid elements entering through the diagonal plane and exiting at the center plane close to the re-entrant corner. For most conditions tested, the flow was found to be symmetrical relative to the diagonal planes ($y = \pm z$) as well as to the y -plane and z -plane. However, in the case of the Boger fluid, the flow becomes time dependent for high Deborah numbers and in this regime, the flow loses its overall symmetry.

For the Newtonian fluid, the size of the corner vortex that forms upstream of the contraction was found to depend not only on the Reynolds number but also on the contraction ratio of the geometry. Under low inertial flow conditions, the size of the corner vortex is nearly independent of Re but as Re is increased, a reduction of vortex size is observed. For the Boger fluid, the vortex size was seen to depend on De_2 and CR in a complex way, showing two distinct behaviors according to the contraction ratio:

- For low contraction ratios, the vortex growth regime is preceded by a decrease in vortex size until a minimum size is reached. Approximately beyond this minimum, we observe the onset of divergent streamlines upstream of the contraction, displaying a bell-shaped form in the region just upstream of the corner vortex similarly to observations in other converging geometries. This diverging flow pattern is strengthened for larger De_2 and lower CR and results in an undershoot in the axial velocity profiles along the centerline upstream of the contraction entrance. Furthermore, the fluid at the centerline is seen to accelerate and the streamwise velocity profiles exhibit a velocity overshoot in the vicinity to the contraction plane, which is enhanced for increasing De_2 .

- For high CR , a monotonous vortex growth was observed, and the size of the corner vortex is dictated by the scaled Deborah number, De_2/CR . For $De_2 \gtrsim 30$, an atypical type of divergent flow is observed in which the streamlines are strongly divergent, revealing a bent-elbow shape near the re-entrant corner just beneath the large corner vortex. This atypical divergent flow also causes an overshoot in the axial velocity profiles at the centerline, which becomes more pronounced for higher CR . In addition, as a consequence of the locally divergent streamlines, an undershoot is observed in the axial velocity profiles along the spanwise direction at an axial

position which intersects the region where the flow is strongly divergent. The extra pressure drop was found to increase significantly with the Deborah number, in line with previous works with axisymmetric contractions.

In summary, when elastic effects become important as De_2 is increased, the flow becomes significantly different from that of a Newtonian fluid and we have identified a number of distinct flow type regions which we have mapped in the $CR-De_2$ parameter space. These regions include, amongst others, a region in which lip and corner vortices coexist, two distinct regions of diverging flow which are associated with vortex growth but exhibit different characteristics for low and high contraction ratios, and unstable flow in which the vortex size varies periodically in time.

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EFFECT OF THE CONTRACTION RATIO UPON VISCOELASTIC FLUID FLOW IN THREE-DIMENSIONAL SQUARE-SQUARE CONTRACTIONS

Abstract

In this work we investigate the laminar flow through square-square sudden contractions with various contraction ratios ($CR = 2.4, 4, 8$ and 12), using a Newtonian fluid and a shear-thinning viscoelastic fluid. Visualizations of the flow patterns were carried out using streak line photography and detailed velocity field measurements were performed using particle image velocimetry. The experimental results are compared with numerical predictions obtained using a finite-volume method. For the Newtonian fluid, a corner vortex is found upstream of the contraction and increasing flow inertia leads to a reduction of the vortex size. Good agreement is observed between experiments and numerical simulations. For the shear-thinning fluid flow a corner vortex is also observed upstream of the contraction independently of the contraction ratio. Increasing the elasticity of the flow, while still maintaining low inertia flow conditions, leads to a strong increase of the vortex size, until an elastic instability sets in and the flow becomes time-dependent at $De \approx 200, 300, 70$ and 450 for $CR = 2.4, 4, 8$ and 12 , respectively. At low contraction ratios, viscoelasticity brings out an anomalous divergent flow upstream of the contraction. For both fluids studied the flow presents a complex three-dimensional helical vortex structure which is well predicted by numerical simulations. However, for the viscoelastic fluid flow the maximum Deborah number achieved in the numerical simulations is about one order of magnitude lower than the critical Deborah number for the onset of the elastic instability found in the experiments.

4.1. Introduction

Non-Newtonian fluids have been extensively studied since the 1940s, when investigations carried out during the Second World War showed several interesting phenomena and stimulated numerous industrial applications (Denn, 2004). Thereafter, flows of viscoelastic fluids and consequently the impact of their rheological properties on the flow patterns were the aim of several research studies. In this context, special attention was given to entry-flow problems, and in particular to contraction flows (e.g. Cable and Boger, 1978a,b, 1979; White and Baird, 1988; Quinzani *et al.*, 1995; Rothstein and McKinley, 1999; Alves *et al.*, 2004; Lu *et al.*, 2006; Oliveira *et al.*, 2007; Walters *et al.*, 2009; Chung *et al.*, 2009).

Several authors have investigated the Newtonian and non-Newtonian fluid flow through sudden contractions with circular and planar geometrical configurations, with particular emphasis on the understanding of the flow patterns developed near the re-entrant and salient corners (e.g. Boger *et al.*, 1986; Nigen and Walters, 2002).

As described by Boger (1987) in his review, the viscoelastic fluid flow through axisymmetric contractions presents a rich variety of flow patterns which depend on the contraction ratio, the fluid rheology and the flow conditions. For instance, large corner vortices develop upstream of the contraction plane, which are normally preceded by the formation of lip vortices near the re-entrant corner, as the Deborah number increases. In order to investigate the influence of inertial effects on the flow of non-Newtonian fluids through axisymmetric contractions, Cable and Boger (1978a, 1978b, 1979) analyzed the flow of aqueous solutions of polyacrylamide (PAA) in 2:1 and 4:1 axisymmetric contractions. At low flow rates, i.e. in the absence of significant inertial and elastic effects, the flow behavior is similar to that found for a Newtonian fluid. As the flow rate is increased, two different flow regimes were found, namely the vortex growth regime and the onset of diverging streamlines. Concerning the viscoelastic fluid flow, Evans and Walters (1986, 1989) and Walters and Webster (1982), amongst others, also studied experimentally the flow of PAA solutions through planar contractions. Later, Purnode and Crochet (1996) simulated numerically the experimental results obtained previously by Evans and Walters (1986, 1989) using the FENE-P constitutive equation. The effect of the polymer concentration, the contraction ratio and the shape of the re-entrant corner on the appearance and enhancement of the vortices were established. The authors found that the observed corner vortices increase in strength with polymer concentration and a lip vortex appears for low-viscosity solutions. Additionally, the lip vortex sets in for the contraction ratio of 4 in the presence of inertial effects, while for higher contraction ratios inertia is not essential.

Most of the previous numerical works focused on the development of robust and stable numerical methods able to simulate the flow of complex fluids with viscoelastic behavior, usually described using a differential constitutive equation. Furthermore, two-dimensional (2D) steady-state numerical simulations have been more exploited than three-dimensional ones (3D), primarily due to the limited computer resources. However, the non-Newtonian fluid flow in 3D geometries leads, in some cases, to a complex and time-dependent flow behavior and therefore only time-dependent 3D numerical calculations can accurately predict the characteristics of the flow. Most of the 3D viscoelastic fluid flows investigated numerically are for planar contractions with ending walls separated by a finite distance, usually defining a large aspect ratio. For example, in the work of Mompean and Deville (1997) the flow of an Oldroyd-B fluid was simulated numerically using 2D and quasi-2D calculations. The authors found that the results obtained from 3D calculations are substantially different from those obtained assuming a 2D approximation. Xue *et al.* (1998) also studied the viscoelastic fluid flow in the benchmark 4:1 planar contraction, using a three-dimensional finite-volume method. The 3D results were also compared with 2D calculations using the upper-convected Maxwell (UCM) and the Phan-Thien-Tanner (PTT) models, in order to describe Boger and shear-thinning fluids, respectively, and the 2D simulations were a good representation of the flow.

For the numerical simulation of viscoelastic fluid flow, it is very important to select an adequate constitutive equation and the corresponding rheological parameters in order to reproduce accurately the shear viscosity, normal stress and extensional properties, since these material functions have a direct impact on the flow behavior leading consequently to the onset of different flow patterns depending on the selection of the parameters.

The square-square contraction is a simple geometrical arrangement that leads to complex 3D flow patterns which can be useful as a benchmark solution for the development of 3D numerical codes in computational rheology. Sirakov *et al.* (2005) studied the flow of a viscoelastic fluid from a square channel to a circular pipe with smaller cross-sectional area. The authors reported the formation of an open vortical structure in the flow through the contraction. This three-dimensional behavior observed experimentally was successfully captured in the numerical results obtained using the eXtended Pom-Pom model. Alves *et al.* (2005) investigated the Newtonian and non-Newtonian fluid flow through a 4:1 square-square contraction. Two different Newtonian fluids were used in the experiments and the corresponding flow patterns were simulated numerically. Moreover, the flow of a viscoelastic fluid with a nearly constant shear viscosity (Boger fluid) was also studied experimentally. More recently, Sousa *et al.* (2009) (cf. Chapter 3) investigated the effect of the contraction ratio on flow through square-square

contractions of a Newtonian fluid and the Boger fluid used by Alves *et al.* (2005). Contraction ratios of 2.4, 4, 8 and 12 were explored and for the Newtonian fluid the corner vortex formed in the upstream channel was found to decrease in size monotonically as the Reynolds number increases. On the other hand, the viscoelastic fluid presented a different flow behavior that depends strongly on the contraction ratio. For the lower contraction ratios, diverging streamlines upstream of the contraction plane were observed with an initial decrease in the vortex size followed by an increase as the Deborah number (or the flow rate) increases. For the higher contraction ratios, vortex enhancement was found with localized diverging streamlines occurring near the re-entrant corner. Furthermore, at high Deborah number flows, an elastic instability sets in and the flow eventually becomes time-dependent. The flow through 3D square-square contractions using a viscoelastic fluid with a shear-thinning rheological behavior was also investigated experimentally and numerically by Alves *et al.* (2008) using a 4:1 contraction ratio. The authors found a strong vortex enhancement, with the flow eventually becoming unsteady as the Deborah number increases. The correct representation of the extensional behavior of the fluid was shown to be important in order to obtain accurate flow predictions.

In this work, we investigate the flow of a shear-thinning fluid through 3D square-square contractions in more detail and assess the influence of the contraction ratio upon the flow patterns and the velocity field. For that purpose, we show the flow patterns visualized using streak line photography and quantify the vortex activity in terms of the vortex length. Moreover, we present the velocity field obtained from particle image velocimetry (PIV) measurements. The experiments were performed with a Newtonian and a shear-thinning viscoelastic fluid and numerical simulations of the flow were performed using a finite volume method (Oliveira *et al.*, 1998). The PTT model (Phan-Thien and Tanner, 1977) was used to describe the rheology of the shear-thinning viscoelastic fluid.

The remainder of this paper is organized as follows: in Section 4.2 the experimental set-up is described, as well as the experimental techniques used in the characterization of the flow. The fluids used and the rheological characterization is presented in Section 4.3. The numerical method employed for the simulation of the Newtonian and non-Newtonian fluid flow as well as the characteristics of the computational meshes used are described in Section 4.4. In Sections 4.5, and 4.6 we present and discuss the results regarding the flow patterns and velocity field, respectively. Finally, in Section 4.7 the main conclusions are summarized.

4.2. Experimental set-up and techniques

The test rig was made of transparent acrylic and placed in a dark room for flow visualizations and particle image velocimetry measurements. Figure 4.1 shows schematically the experimental set-up.

The main duct is composed of two parts: an upstream channel with a fixed width of $2H_1 = 24$ mm and an interchangeable square duct with a smaller inner side length, which fits precisely in the larger square duct. To assess the effect of the contraction ratio, this downstream side length can be set to $2H_2 = 10.0, 6.0, 3.0$ or 2.0 mm, corresponding to contraction ratios of 2.4, 4, 8 and 12, respectively. We define the contraction ratio, CR , as the ratio between the upstream and the downstream side lengths of the square ducts, $CR = H_1/H_2$. The upstream and downstream sections are denoted by subscripts 1 and 2, respectively. The flow rate is set by varying the height between the two free surfaces of the reservoirs or using a vacuum pump (KNF Loboport N811- KT.18) that applies vacuum to the outlet reservoir. For both cases, the selection of the diameter of the pipe that connects the duct to this outlet reservoir is also important to set the desired flow rate. To avoid degradation of the viscoelastic fluids used, flow regulating valves were not used in the experimental set-up. Further details on the experimental set-up can be found in Sousa *et al.* (2009) (cf. Chapter 3).

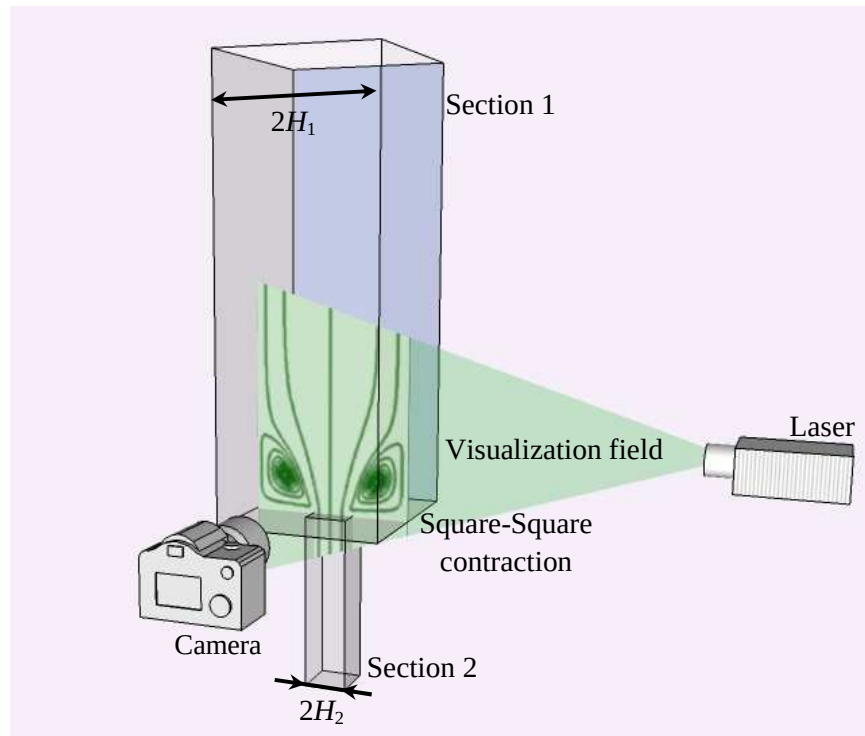


Figure 4.1. Three-dimensional schematic representation of the experimental set-up used for flow visualization and particle image velocimetry measurements.

The flow patterns were visualized using long time exposure streak line photography, while detailed velocity field measurements were performed using PIV. For this purpose, the working fluids were seeded with a low concentration of 10 μm PVC tracer particles. In order to illuminate the plane of the flow under study, we used either a 3 mW 532 nm laser diode (Imatronic, model LLM115) or a 5 mW 635 nm laser diode (Vector, model 5200-20), both fitted with a cylindrical lens to create a light sheet. The streak line images were captured using a Canon EOS 30D digital camera equipped with a macro lens (Canon EF100mm, f/2.8) and the exposure time was varied from about 1 s to 10^3 s, depending on the flow rates.

For the PIV measurements, and for the flow visualizations at high flow rates, a double pulsed Nd:YAG laser with a maximum energy of 50 mJ (Solo PIV III, New Wave Research) was used to illuminate the channel and a digital CCD camera (Flow Sense 2M from Dantec Dynamics coupled with a Nikon AF Micro 60 mm lens), which was placed perpendicularly to the light sheet, was used to record the images. Time interval between pulses was varied in the range of $10^{-4} \leq \Delta t / \text{s} \leq 1$ depending on the flow rates. The images were acquired and post-processed by FlowManager v4.60 software (Dantec Dynamics) using adaptive correlation on interrogation windows ranging between 64 by 64 pixels to 16 by 16 pixels, with 50% overlap.

4.3. Fluid characterization

A Newtonian fluid and a viscoelastic fluid with a shear-thinning rheological behavior were used in this study. The Newtonian fluid, used for comparison purposes, is an aqueous solution of glycerol (85 % w/w) and the non-Newtonian fluid is a solution composed of 40.0 % (w/w) of glycerol, 59.9 % (w/w) of water and PAA at a weight concentration of 600 ppm. In order to minimize bacteriological growth in the fluids, and consequently to prevent its degradation, a biocide (Kathon LXE, Rohm and Haas) at a weight concentration of 25 ppm was added to all solutions. The density (ρ) of the Newtonian and viscoelastic fluids, measured at 293.2 K using a hydrometer (readability of 0.001 kg/m^3 ; range 1 000–1 300 kg m^{-3}), were 1 221 kg m^{-3} and 1 156 kg m^{-3} , respectively.

The fluids were characterized rheologically using a shear rheometer (Physica MCR301, Anton Paar) under shear rate control. The flow curves were measured at different temperatures (T) ranging from 283.2 K to 303.2 K. The effect of the temperature on the shear viscosity can be described using an Arrhenius equation for the shift factor, a_T :

$$\ln(a_T) = \frac{\Delta H}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right), \quad (4.1)$$

where ΔH is the activation energy for flow, R the universal gas constant and T_0 the reference temperature. We selected $T_0 = 293.2$ K, the average temperature at which the experiments were carried out, since the temperature of the measurements varied from 293.0 K to 294.7 K. The shift factor is generally defined as (Dealy and Plazek, 2009):

$$a_T = \frac{\eta(T)}{\eta(T_0)} \frac{T_0}{T} \frac{\rho_0}{\rho}, \quad (4.2)$$

in which $\eta(T_0)$ is the shear viscosity at the reference temperature, $\eta(T)$ is the shear viscosity at a given temperature T and ρ_0 and ρ are the fluid densities at the reference temperature, and at temperature T , respectively. Nevertheless, in the range of measurements performed, the temperature variation is small and consequently the fluid density does not change significantly. As a result, the shift factor can be described as:

$$a_T = \frac{\eta(T)}{\eta(T_0)}. \quad (4.3)$$

For the Newtonian fluid, $\Delta H/R = 5580$ K and $\eta_0 = 0.0982$ Pa s at the reference temperature. For the viscoelastic fluid, the flow curve was measured also at various temperatures, ranging from 283.2 K to 298.2 K, using a cone-plate with a 50 mm in diameter and 2° angle. Subsequently, the time-temperature superposition principle (Dealy and Plazek, 2009) was used to obtain the master curve at the reference temperature ($T_0 = 293.2$ K). In this case, a ratio of $\Delta H/R = 2703$ K was obtained. Figure 4.2 shows the resulting steady shear viscosity master curve as well as the corresponding fit of a simplified Phan-Thien-Tanner (PTT) model (Phan-Thien and Tanner, 1977) with a solvent viscosity contribution. A detailed description of this viscoelastic model is given in Section 4.4.

The parameters of the PTT model used in the fit presented in Figure 4.2 are: extensibility parameter, $\varepsilon = 0.06$; polymer shear viscosity coefficient, $\eta_p = 1.62$ Pa s; solvent shear viscosity, $\eta_s = 0.03$ Pa s; relaxation time, $\lambda = 32$ s.

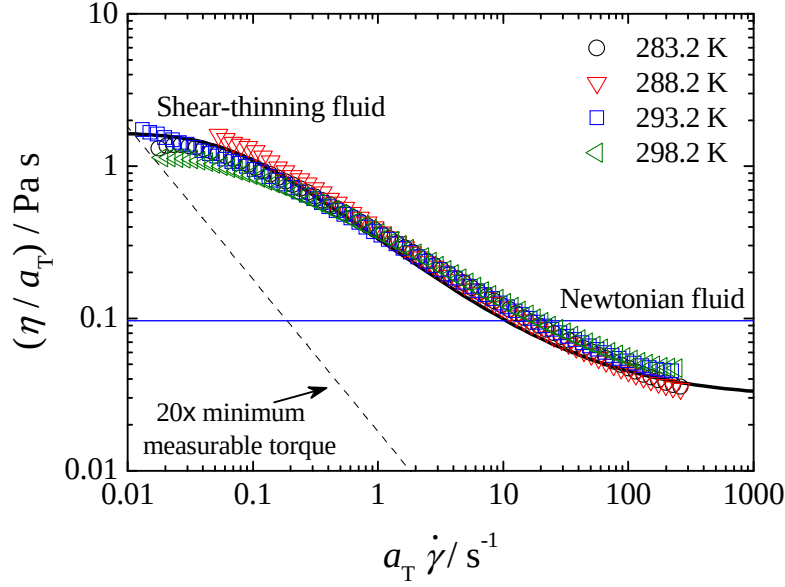


Figure 4.2. Steady shear viscosity of the Newtonian and the shear-thinning fluid. For the shear-thinning fluid, the symbols represent the experimental data measured at different temperatures and the thick solid line the PTT model predictions. For the Newtonian fluid, the thin solid line represents the shear viscosity of the fluid at 293.2 K. The minimum measurable shear viscosity based on 20 times the minimum resolvable torque is represented by the dashed line.

4.4. Numerical method and computational meshes

In order to describe the isothermal and laminar flow of Newtonian and non-Newtonian incompressible fluids through square-square contractions, the following equations need to be solved: the conservation of mass,

$$\nabla \cdot \mathbf{u} = 0, \quad (4.4)$$

the conservation of momentum,

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + \nabla \cdot (\mathbf{u} \mathbf{u}) \right] = -\nabla p + \eta_s \nabla^2 \mathbf{u} + \nabla \cdot \boldsymbol{\tau}, \quad (4.5)$$

and an adequate constitutive equation for the polymeric contribution ($\boldsymbol{\tau}$) to the extra stress tensor. In these equations, $\mathbf{u}$ is the velocity vector, t the time, p the pressure and η_s the Newtonian solvent viscosity. The solvent contribution to the extra stress tensor, $\boldsymbol{\tau}_s = \eta_s (\nabla \mathbf{u} + \nabla \mathbf{u}^T)$, is

added explicitly in the momentum equation since this diffusive term ($\nabla \cdot \boldsymbol{\tau}_s = \eta_s \nabla^2 \mathbf{u}$) promotes numerical stability.

The rheological model used to fit the experimental shear data of the viscoelastic fluid was the simplified form of the Phan-Thien-Tanner model (cf. Section 4.3) and consequently, the polymeric contribution to the extra-stress tensor is given by (Phan-Thien and Tanner, 1977):

$$f(\text{Tr } \boldsymbol{\tau})\boldsymbol{\tau} + \lambda \left[\frac{\partial \boldsymbol{\tau}}{\partial t} + \nabla \cdot \mathbf{u} \boldsymbol{\tau} \right] = \eta_p (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \lambda (\boldsymbol{\tau} \cdot \nabla \mathbf{u} + \nabla \mathbf{u}^T \cdot \boldsymbol{\tau}). \quad (4.6)$$

The linear form of the PTT model was used, with the stress function given by (Phan-Thien and Tanner, 1977):

$$f(\text{Tr } \boldsymbol{\tau}) = 1 + \frac{\lambda \varepsilon}{\eta_p} \text{Tr}(\boldsymbol{\tau}), \quad (4.7)$$

where $\text{Tr}(\boldsymbol{\tau})$ represents the trace of the extra stress tensor. In the numerical calculations for the polymer stress tensor we used the log-conformation methodology (Fattal and Kupferman, 2004) described in detail by Afonso *et al.* (2009). This transformation is used in order to improve the stability of the numerical method and to reduce the High-Weissenberg Number Problem (HWNP) and attain converged solutions at higher Deborah (or Weissenberg) numbers (Hulsen *et al.*, 2005).

The set of governing equations are solved sequentially using a finite volume method (FVM) with a time marching pressure-correction algorithm, as described in detail in Oliveira *et al.* (1998) and Afonso *et al.* (2009). Regarding the accuracy of the calculations, an implicit first-order Euler scheme is used in the discretization of the time derivative, second-order central differences are used to discretize the diffusive terms and the CUBISTA high-resolution scheme (Alves *et al.*, 2003) is used in the discretization of the advective terms in the momentum and constitutive equations.

The simulations were performed on three-dimensional meshes covering the whole wall-to-wall geometries, i.e., no symmetry at the central and/or diagonal planes was enforced. The meshes consist of orthogonal blocks which are composed of non-uniform cells. The inlets and outlets were positioned far from the expansion plane so that fully developed flow conditions were attained. No-slip boundary conditions were imposed at the solid walls and a uniform

velocity profile and null stress components were imposed at the inlet boundary. The outflow boundary condition involves vanishing stream wise gradients of velocity and stress components and linear extrapolation of pressure from the two upstream computational cells.

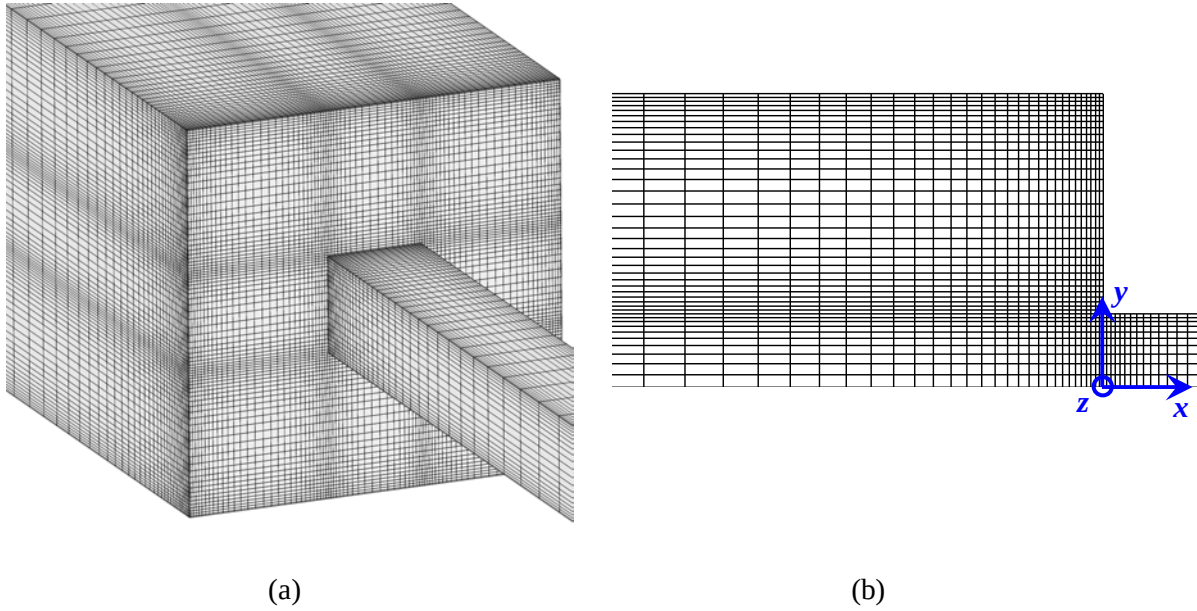


Figure 4.3. Zoomed view near the contraction plane of the refined mesh (M80) used in the numerical simulations of the flow through the 4:1 contraction. (a) 3D view; (b) central plane, $z = 0$ (only half of the plane is shown).

Table 4.1. Characteristics of the computational meshes used.

CR	Mesh	NC	$\Delta x_{\min}/2H_1$	$\Delta y_{\min}/2H_1 = \Delta z_{\min}/2H_1$
2.4	M40	164 000	2.08×10^{-2}	1.99×10^{-2}
	M64	419 840	1.30×10^{-2}	1.25×10^{-2}
	M80	656 000	1.03×10^{-2}	9.93×10^{-3}
4	M40	51 000	1.31×10^{-2}	1.25×10^{-2}
	M64	130 560	8.20×10^{-3}	8.16×10^{-3}
	M80	408 000	6.26×10^{-3}	6.25×10^{-3}
8	M40	163 200	7.50×10^{-3}	7.50×10^{-3}
	M64	417 792	4.69×10^{-3}	4.55×10^{-3}
	M80	652 800	3.75×10^{-3}	3.75×10^{-3}
12	M48	113 664	1.07×10^{-2}	9.52×10^{-3}
	M60	177 600	8.61×10^{-3}	8.33×10^{-3}
	M96	909 312	4.04×10^{-3}	3.64×10^{-3}

Figure 4.3 shows a zoomed view, near the contraction plane, of the refined mesh used in the simulation of the flow through the 4:1 square-square contraction. To assess numerical accuracy, three different computational meshes were used for each CR studied. For $CR = 2.4, 4$ and 8 , we used mesh M40, mesh M64 and a more refined mesh, M80. For $CR = 12$, meshes M48, M60 and M96 were used. Details of the total number of cells (NC) as well as the cell smallest dimensions ($\Delta x_{\min}/(2H_1)$, $\Delta y_{\min}/(2H_1)$ and $\Delta z_{\min}/(2H_1)$) for each mesh are described in Table 4.1. The reference number identifying each mesh corresponds to the number of cells along each transverse direction in the upstream channel.

4.5. Flow patterns and vortex length

4.5.1. Newtonian fluid

A detailed discussion of the Newtonian fluid flow through square-square contractions with different contraction ratios was documented in a previous work (Sousa *et al.*, 2009) (cf. Chapter 3). Here, we focus only on the main results with the purpose of comparing the flow of the Newtonian and the shear-thinning fluids.

In order to characterize the Newtonian fluid flow we use the Reynolds number, here defined as $Re = \rho U_2(2H_2)/\eta$, where U_2 is the average velocity in the downstream channel. In Figure 4.4 we show the flow patterns obtained for the Newtonian fluid flow through the 8:1 square-square contraction for two Reynolds numbers ($Re = 2.13$ and 13.7). Moreover, in Figure 4.4 we compare the experimental results with numerical predictions highlighting the variation of the vortex length with increasing flow inertia. The vortex length, x_R , is here defined as the distance from the separation point to the contraction plane, as shown in Figure 4.4(a).

The Newtonian fluid flow behavior is similar for all contraction ratios studied ($CR = 2.4, 4$ and 8). A corner vortex appears upstream of the contraction plane which is typical of flows through abrupt contractions (cf. Oliveira *et al.*, 2007 for the axisymmetric arrangement and Alves *et al.*, 2003b for the planar configuration) and agrees with the findings of Sirakov *et al.* (2005) and Alves *et al.* (2005) for 3D geometries. Furthermore, the size of the upstream vortex decreases when the Reynolds number is increased. The comparison between the experimental and numerical results demonstrates that there is an excellent agreement between both approaches.

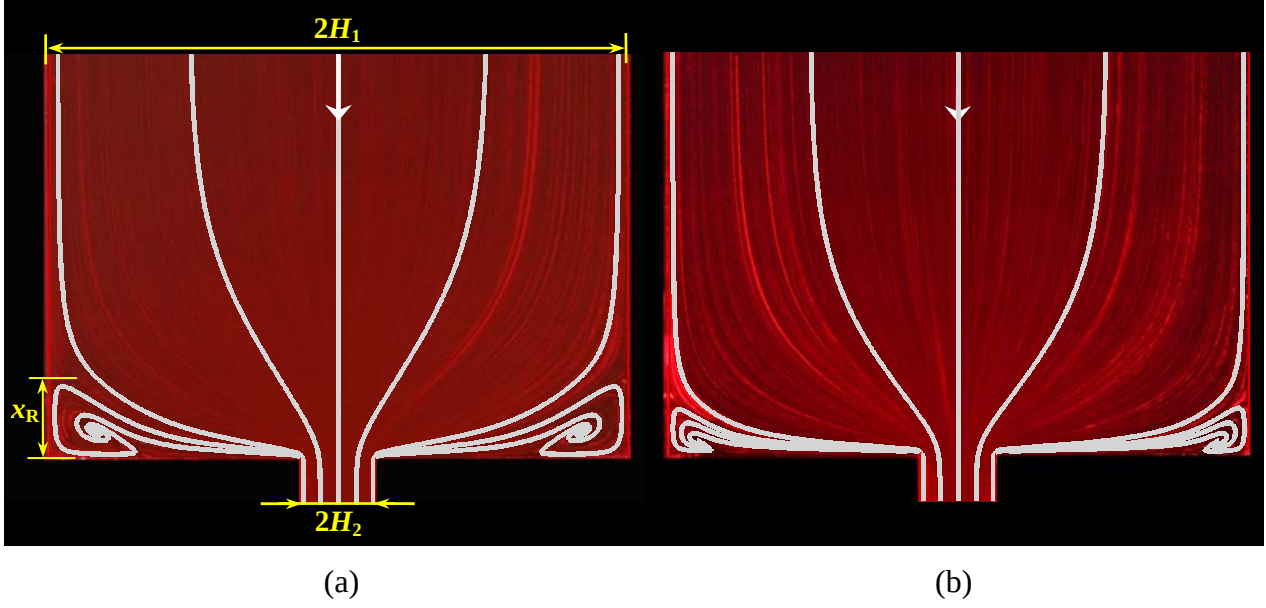


Figure 4.4. Experimental and numerical streamlines obtained at the center plane for the Newtonian fluid flow through the 8:1 square-square contraction at (a) $Re = 2.13$ and (b) $Re = 13.7$.

4.5.2. Shear-thinning viscoelastic fluid

Besides the contraction ratio and the Reynolds number, an important dimensionless number that should be taken into account for the characterization of the viscoelastic fluid flow is the Deborah number, which is here defined as $De = \lambda U_2 / H_2$. Due to the shear-thinning behavior of the viscoelastic fluid, the Reynolds number is now defined as $Re = \rho U_2 (2H_2) / \eta(\dot{\gamma})$, where the shear viscosity is calculated using the PTT model at the characteristic shear rate $\dot{\gamma} = U_2 / H_2$.

Figure 4.5 shows the flow patterns for a low and a high contraction ratio ($CR = 2.4$ and 12) for different Deborah numbers, which were varied by changing the flow rate. The Reynolds number also varies with the flow rate, but in all cases inertial effects are not important because Re is small for the range of flow conditions studied with the viscoelastic fluid.

For all CR studied, at low flow rates (or De), a Newtonian-like behavior is observed (cf. Figure 4.5(a₁)). Increasing the Deborah number, a strong vortex enhancement is observed (cf. Figure 4.5(a₂)–4.5(a₄) and 4.5(b₁)–4.5(b₄)). For the lower contraction ratios investigated, $CR = 2.4$ and 4 , this vortex enhancement is accompanied by the onset of diverging streamlines upstream of the contraction plane, which appear at $De \gtrsim 10$ for $CR = 2.4$ and at $De \gtrsim 20$ for $CR = 4$. Divergent streamlines are often observed in viscoelastic contraction flows (Cable and Boger, 1978a, 1978b, 1979; Evans and Walters, 1989, McKinley *et al.*, 2007) and are usually enhanced at lower contraction ratios (Alves and Poole, 2007), as confirmed here.

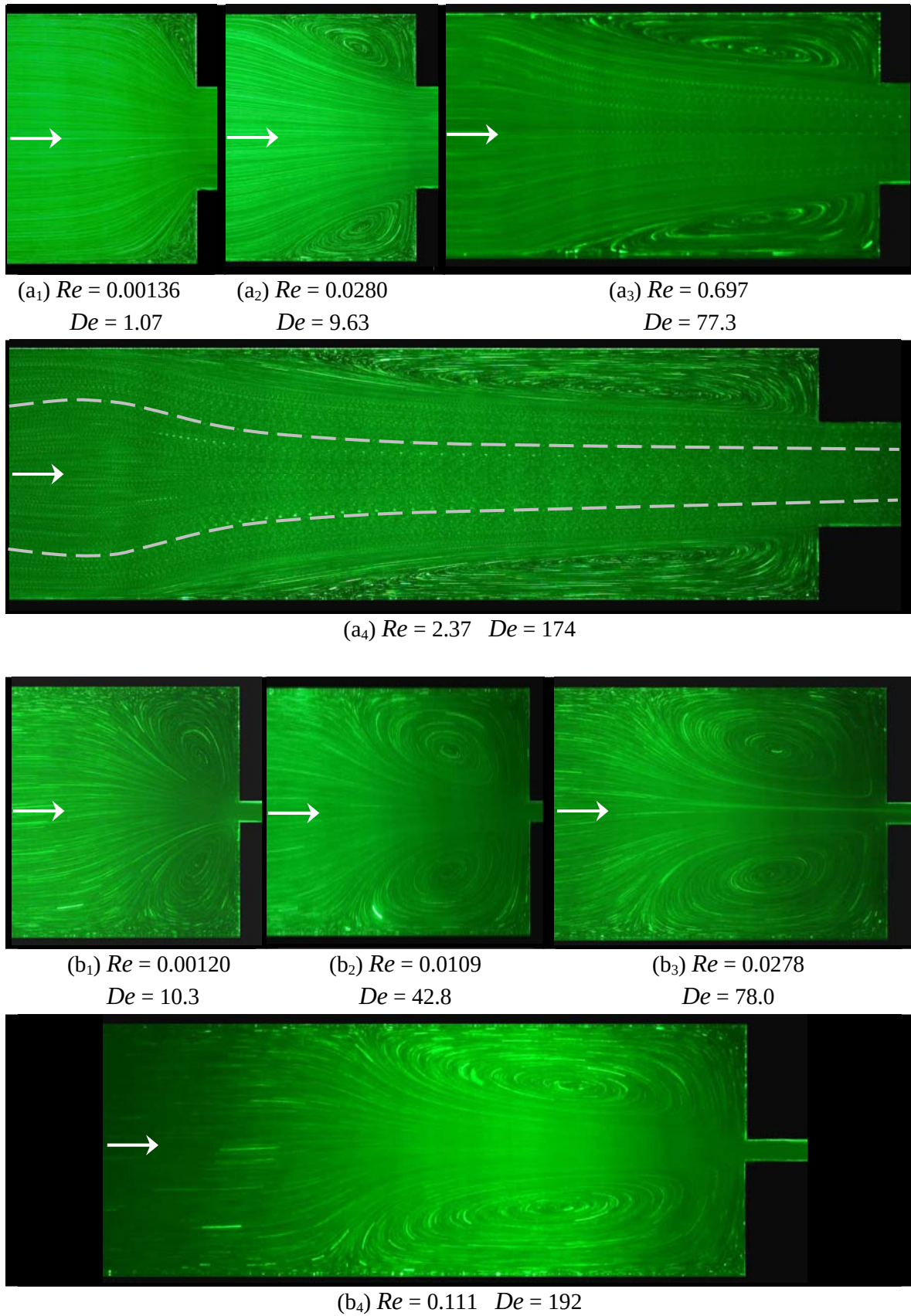


Figure 4.5. Flow patterns obtained with the viscoelastic fluid at the center plane for (a) $CR = 2.4$ and (b) $CR = 12$. The diverging streamlines which appear for $CR = 2.4$ are highlighted by dashed lines (a₄).

In fact, in the present work divergent flow was not observed for the higher contraction ratios, $CR = 8$ and 12 . Furthermore, the small values of the Reynolds numbers used in this work suggest that inertia is not a fundamental condition for the appearance of diverging flow, in agreement with the findings of Alves and Poole (2007).

The vortex length at the center plane of the square channel, x_R , was measured for all flow rates and contraction ratios studied, using the streak line images captured experimentally as well the velocity field predicted from numerical simulations. Figure 4.6 shows the variation of the dimensionless vortex length at the center plane ($x_R/(2H_1)$) as a function of the Deborah number for all the CR investigated.

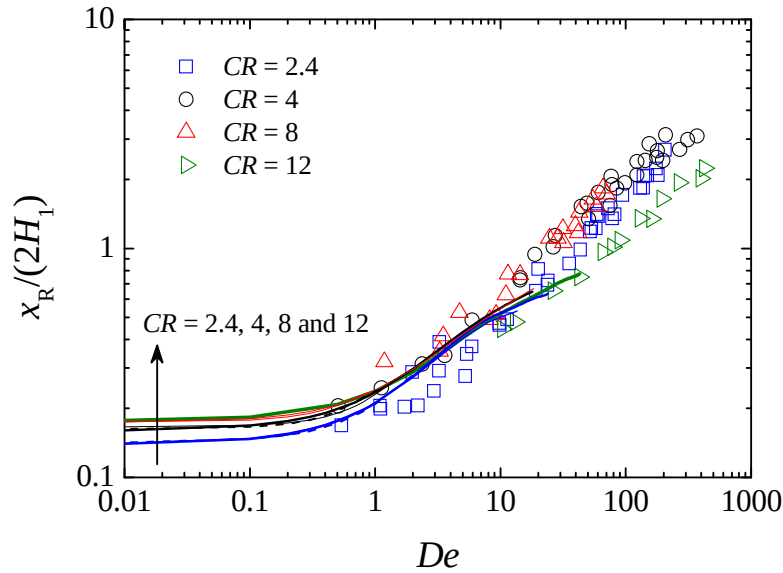


Figure 4.6. Dimensionless vortex length at the center plane as a function of the Deborah number for all contraction ratios studied. The symbols represent the experimental data, the thick solid lines represent the numerical predictions using mesh M40, the thin solid lines the predictions using mesh M64 and the dashed lines the predictions using mesh M80.

As can be seen, the recirculations that appear upstream of the contraction, increase in size with an increase in the flow rate (or Deborah number), in contrast with Newtonian fluid flow, in which the vortex length decreases slightly with the flow rate, as shown in Section 4.5.1. For all CR , but particularly for the lower contraction ratios studied in this work ($CR = 2.4$ and 4), the vortex length increases considerably, exceeding three times the width of the upstream channel. This considerable vortex enhancement observed in the flow of a shear-thinning fluid through contractions was previously documented in the works of Alves *et al.* (2008) for a 4:1 square-

square contraction and also by Evans and Walters (1986) for an axisymmetric geometry. For $CR = 2.4$ and 4, the numerical results presented in Figure 4.6 were obtained using the three meshes described in Section 4.4 (M40, M64 and M80). For the range of De where steady solutions were obtained in the numerical simulations, the vortex lengths predicted numerically are in good agreement with the experimental values. Moreover, the differences between meshes M64 and M80 are small (relative deviation of about 1%). For this reason and since numerical simulation of the viscoelastic fluid flow using the more refined mesh is time-consuming due to the very high relaxation time of the fluid and the large number of control volumes of those meshes, for the higher contraction ratios, $CR = 8$ and 12, we only show the numerical results obtained using mesh M40 or M48 and M64 or M60, respectively.

In the numerical calculations using the PTT model we are only able to probe a range of Deborah numbers much smaller than in the experiments, as a consequence of the well known High-Weissenberg Number Problem (Fattal and Kupferman, 2004) and the limitations of the PTT model. Nevertheless, we are able to predict numerically the observed vortex enhancement as De increases with good accuracy. To further attest this, in Figure 4.7 we show the numerical streamlines for the shear-thinning fluid flow through the 2.4:1 square-square contraction and a comparison with the flow visualization at the high De to illustrate the good agreement between experiments and numerical predictions.

As explained previously, at low Deborah numbers the flow is Newtonian-like (compare Figure 4.7(a) and Figure 4.5(a₁) for the viscoelastic fluid to Figure 4.4(a) corresponding to the Newtonian fluid) and the upstream vortices are slightly concave-shaped. At $De = 0.231$, the numerical results show the presence of small lip vortices near the re-entrant corners which were not observed in the experimental results because visualizations were restricted to $De \gtrsim 0.5$ due to experimental difficulties of operation at such low flow rates. Increasing De , the recirculations increase in size and become convex in shape (cf. Figure 4.7(b) and 4.7(c)) in contrast with those obtained for Newtonian fluid flow. At a certain value of the Deborah number, there is a reversal of the flow direction within the 3D recirculation which is predicted numerically as can be seen in Figure 4.7(c) and 4.7(d) where the fluid is clearly entering the recirculation in the middle plane, in contrast with the behavior of low De or Newtonian fluid flows (cf. Figure 4.4), where the fluid exits the recirculation at the middle plane vortex.

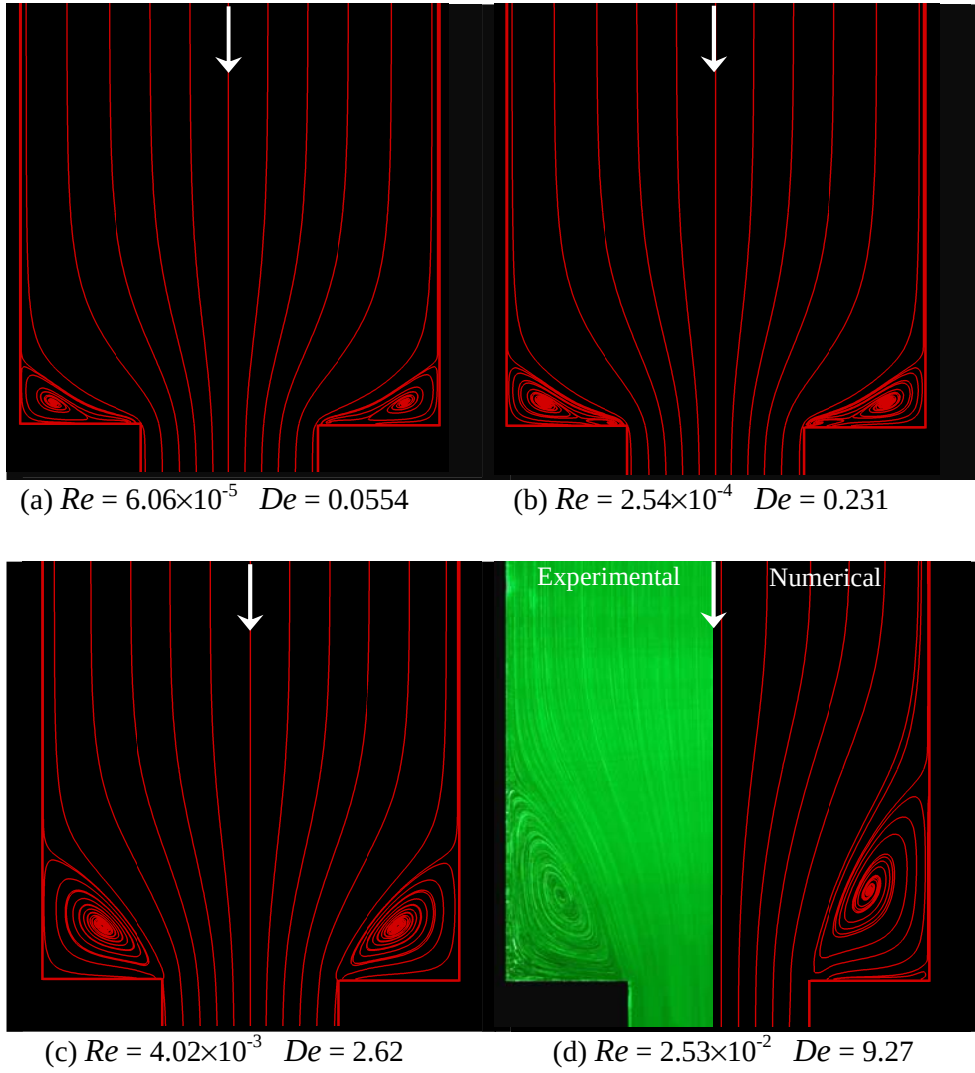


Figure 4.7. Viscoelastic fluid flow patterns predicted numerically for $CR = 2.4$ at the center plane at different flow conditions and comparison with experimental results (d).

For the lower CR , within the experimental range of flow rates measured, it is possible to establish that the inversion of the flow direction occurs at $De \approx 3$ and $De \approx 2$ for $CR = 2.4$ and 4 , respectively. For higher CR we are not able to determine experimentally the value of De at which flow reversal takes place due to the absence of experimental data at those flow conditions. From numerical results we predict that this phenomenon occurs at $De \approx 1.5$ for all CR . The same phenomenon was reported in the works of Alves *et al.* (2008) and of Sousa *et al.* (2009) (cf. Chapter 3) for the flow through square-square contractions of a PAA-based shear-thinning and a Boger fluid, respectively. Additionally, flow reversal was also observed in square-square expansions (Sousa *et al.*, 2010) (cf. Chapter 5) although in this case it only happened for Newtonian fluids, when inertial effects become important. The reported vortex enhancement due to elasticity in contraction flows, and due to inertia in expansion flows, seems to be linked with the flow reversal phenomenon.

Figure 4.8 shows the projected streamlines at different parallel planes of the square channel, in order to illustrate the trajectory of the fluid within the recirculation for high De flows in the 2.4:1 square-square contraction. It is interesting to note that the recirculation structure is complex and that the eye of the recirculation moves upstream as the wall plane is approached.

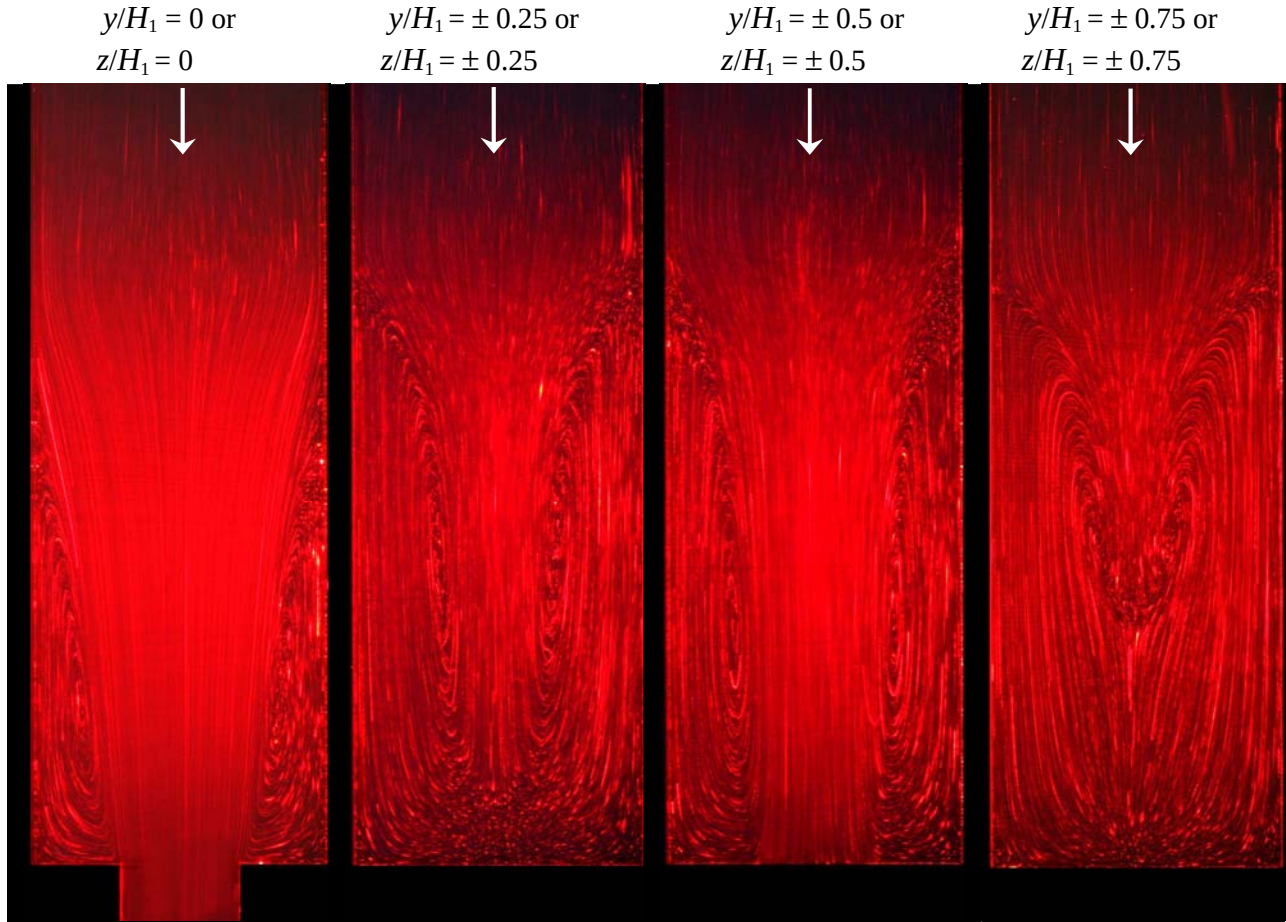


Figure 4.8. Projected streamlines obtained for the viscoelastic fluid at different parallel planes of the 2.4:1 square-square contraction for $Re = 0.930$ and $De = 92.0$.

When the flow rate (or De) is increased further, an elastic instability occurs and the flow becomes time-dependent, with the size of the recirculations varying in time in a complex way, which eventually becomes chaotic at larger flow rates. In Figure 4.9 we show a sequence of *instantaneous* pathlines visualized using the shear-thinning fluid flow for a Deborah number at which the flow is unstable, clearly showing that the vortices vary in size and shape with time. To further attest to the time dependency and the three-dimensional structure of the flow we also show in Figure 4.9 *instantaneous* projections of pathlines at a plane located at middle-distance between the center plane and the wall of the square channel ($y/H_2 = \pm 0.50$ or $z/H_2 = \pm 0.50$).

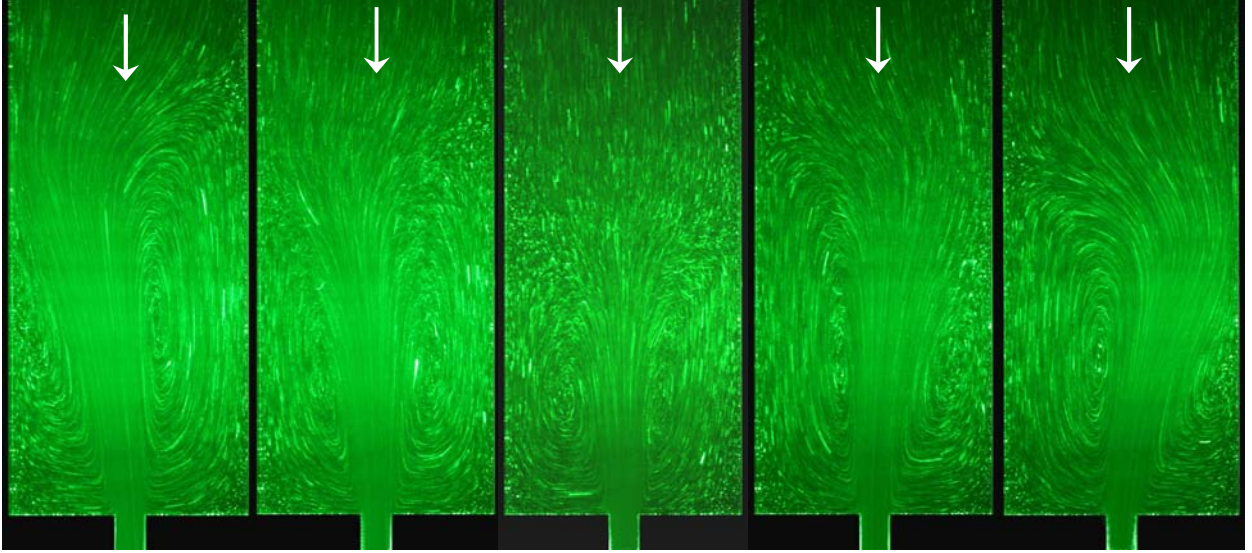
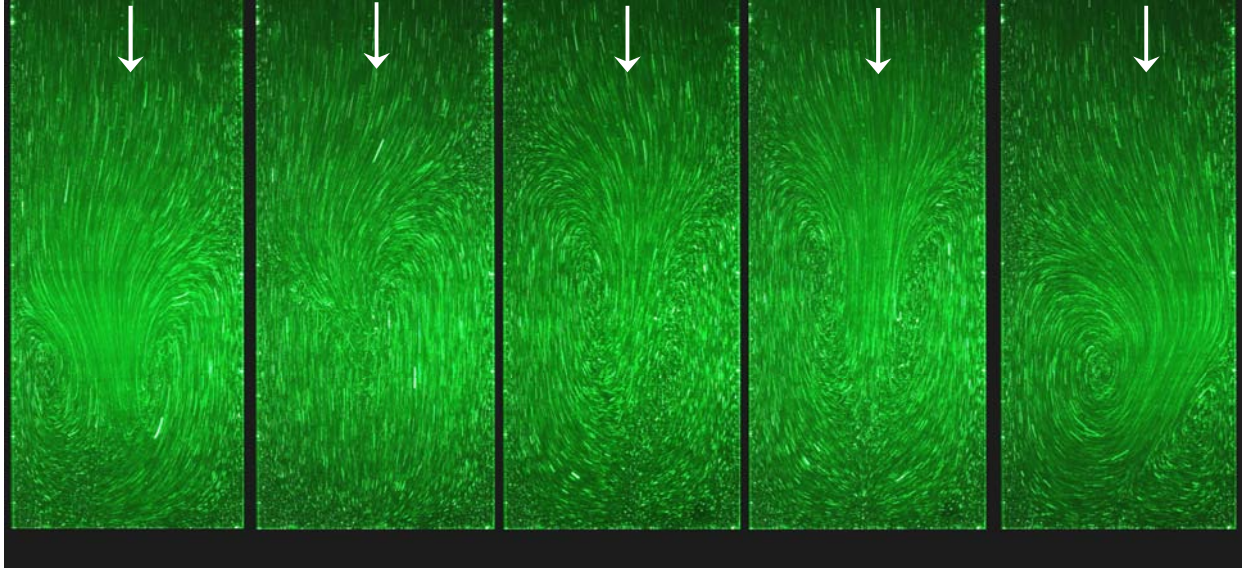
(a) $y/H_2 = 0$ or $z/H_1 = 0$ (b) $y/H_2 = \pm 0.50$ or $z/H_2 = \pm 0.50$

Figure 4.9. Streak line images of the viscoelastic fluid flow through the 8:1 square-square contraction acquired at different moments of the oscillating cycle for $Re = 0.181$ and $De = 152$: (a) center plane; (b) middle plane between the wall and the center plane.

As can be seen, the flow becomes asymmetric oscillating periodically, i.e. during a cycle of oscillation, one region of the vortex size decreases while the other part is increasing. From Figure 4.9(b), it is possible to conclude that the entire 3D vortical structure is moving. The normalized frequency of oscillation, λf , is plotted in Figure 4.10 as a function of the ratio De/CR , together with results reported in a previous work (Sousa *et al.*, 2009) (cf. Chapter 3) for a Boger fluid flowing through square-square contractions with $CR = 4, 8$ and 12 . A general trend can be identified with λf varying approximately linearly with $\log(De/CR)$.

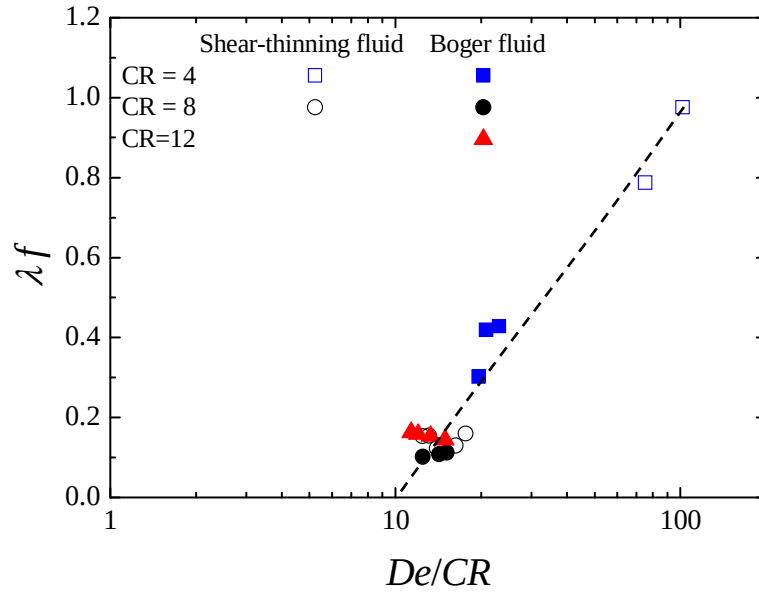


Figure 4.10. Normalized frequency of oscillation as a function of De/CR for $CR = 4, 8$ and 12 . The open symbols represent experimental data for the shear-thinning fluid and the filled symbols represent experimental data obtained by Sousa *et al.* (2009) (cf. Chapter 3) for the Boger fluid flow through similar square-square contractions. The dashed line is a guide to the eye.

4.5.3. Flow pattern map

The different flow phenomena identified for the viscoelastic fluid flow through square-square contractions can be summarized in a $CR - De$ map, as shown in Figure 4.11.

Each flow regime is bounded between an upper and lower limit that are marked as dashed lines in Figure 4.11. The distinct flow regimes observed with the shear-thinning fluid were categorized as:

Newtonian-like flow for conditions of negligible elastic effects, in which the upstream vortices increase in size only slightly as the Deborah number increases;

Vortex enhancement, in which the vortex increases significantly due to elastic effects;

Diverging streamlines, which occurs for the lower contraction ratios ($CR = 2.4$ and 4) when the Deborah number (and consequently the elastic effects) is further increased. We note that this flow regime occurs simultaneously with vortex enhancement;

Unsteady flow that occurs at higher Deborah numbers, in which the entire flow field oscillates periodically.

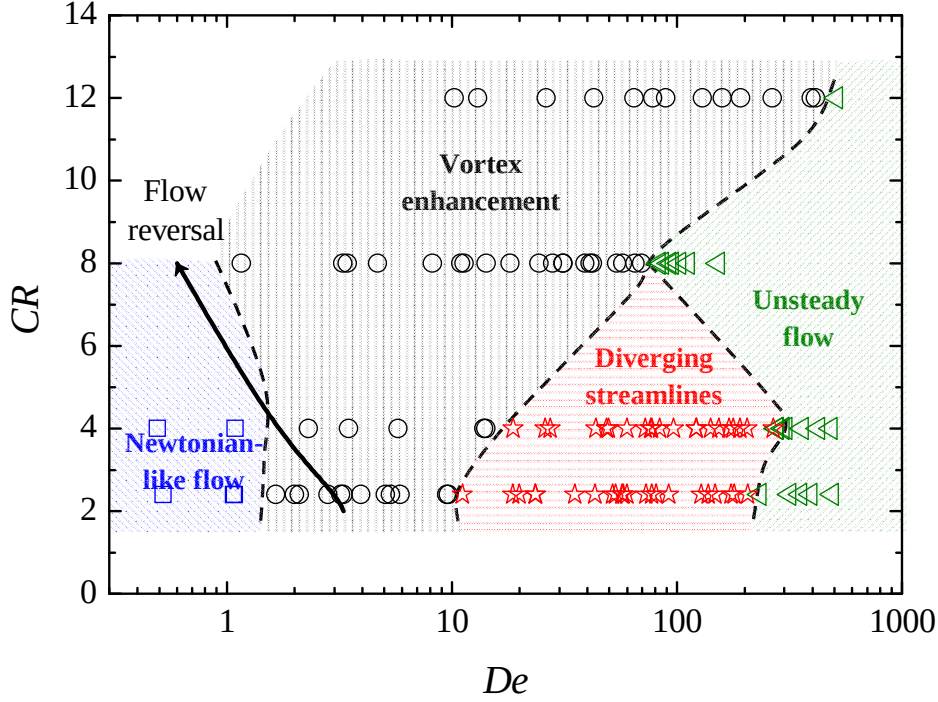


Figure 4.11. Summary of all flow patterns identified in the $CR - De$ parameter space for the shear-thinning viscoelastic fluid.

4.6. Velocity field

In order to highlight the effect of elasticity on the velocity field, in Figure 4.12 we show dimensionless axial velocity profiles along the centerline ($y = z = 0$) for three contraction ratios at different Deborah numbers. In addition, the numerical prediction of the axial velocity profile for a Newtonian fluid flow under creeping flow conditions is also presented in Figure 4.12 for comparison purposes, since this is the limiting case that would be observed with the viscoelastic fluid at very low flow rates. For the range of high Deborah numbers at which the experimental velocity profiles were measured using PIV, we are unable to obtain the velocity field numerically using the PTT model due to the high Weissenberg number problem. However, we have performed numerical calculations using a generalized Newtonian fluid (GNF), which does not account for elasticity, but presents a rheological behavior in steady shear flow similar to that of the viscoelastic fluid used in the experiments. For this purpose, a Carreau-Yasuda model (Bird *et al.*, 1987) was used to fit the shear viscosity data of the GNF model:

$$\eta = \eta_s + \frac{\eta_0 - \eta_s}{\left[1 + (\Lambda \dot{\gamma})^a\right]^{(1-n)/a}}, \quad (4.8)$$

where the fitted parameters are $\eta_0 = 1.65 \text{ Pa s}$, $\eta_s = 0.03 \text{ Pa s}$, $\Lambda = 10 \text{ s}$, $n = 0.36$ and $a = 0.9$.

For the inertialess Newtonian fluid flow the behavior is similar for all CR : upstream of the contraction the fluid experiences a constant axial velocity along the centerline, up to a location near the contraction plane, when the fluid accelerates and enters the downstream channel eventually reaching the conditions of developed flow, with $u/U_2 = 2.096$ at the centerline of a square channel (Sousa *et al.*, 2009) (cf. Chapter 3). For the viscoelastic fluid flow, the influence of elasticity leads to a different, and more complex, variation of the axial velocity along the centerline. At a location far upstream from the contraction plane, the value of the dimensionless axial velocity is similar to that found for the Newtonian fluid. However, for all contraction ratios studied the location at which the fluid starts to accelerate, due to entry effects, shifts significantly upstream with an increase with the Deborah number. For the lower contraction ratios ($CR = 2.4$ and 4) the presence of diverging streamlines leads to a velocity undershoot immediately before the fluid starts to accelerate. Concurrently, for the same CR , the velocity profiles reveal an overshoot when the fluid flows through the contraction plane and enters the downstream duct (in accordance with the findings of Alves and Poole (2007), Poole and Alves (2009) and Sousa *et al.* (2009) (cf. Chapter 3)) particularly for the lower CR . The enhancement of the velocity undershoot and subsequent velocity overshoot in contraction flows of viscoelastic fluids was investigated by Alves and Poole (2007) for smooth planar contractions and shown to be pronounced at low contraction ratios, in agreement with our observation in a square-square contraction flow.

In Figure 4.12 we also include the numerical results obtained with the GNF model in order to demonstrate that the significant changes observed in the velocity profiles as De increases, as well as the onset of undershoots and overshoots in the centerline velocity profile are a consequence of elastic effects. For clarity, only the predicted velocity profiles for the higher flow rates are illustrated in Figure 4.12 for each CR . The overshoot on the axial velocity profiles for the lower CR is not observed in the numerical predictions using the GNF model, showing clearly that this is an effect of elasticity. The upstream shift of the velocity profile is also not captured in the GNF simulations. On the other hand, the GNF model is able to accurately predict the axial velocity in the downstream channel for fully-developed flow conditions for $CR = 8$. For $CR = 2.4$ and 4 , due to the large velocity overshoots, the fully-developed flow conditions downstream of the contraction plane were not achieved in the PIV measurements, but eventually the velocity profile will fall down to the values predicted by the GNF model. We note that the dimensionless velocity at the centerline under fully developed flow conditions is lower than that found for a Newtonian fluid, due to the shear-thinning rheological behavior of the GNF fluid.

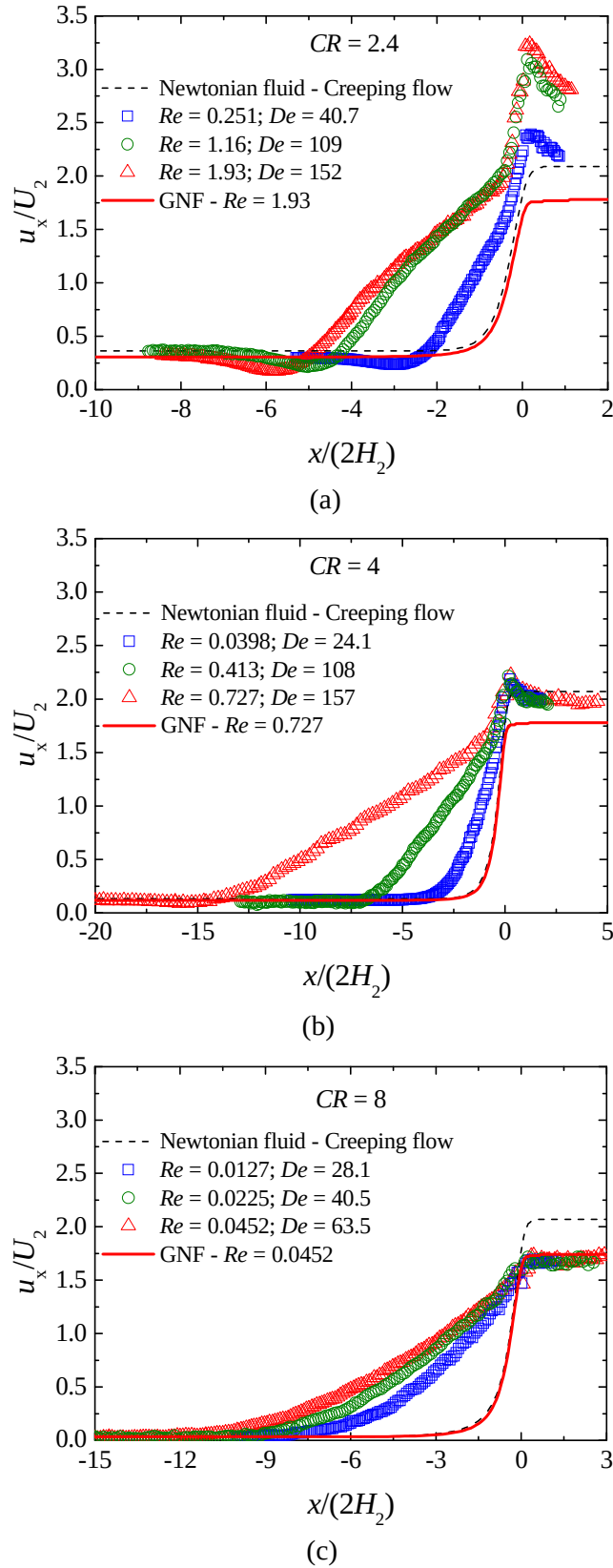


Figure 4.12. Axial velocity profiles at the channel centerline for $CR = 2.4$ (a), $CR = 4$ (b) and $CR = 8$ (c). The symbols represent the experimental data obtained with the viscoelastic fluid, the dashed line represents the numerical predictions for a Newtonian fluid under creeping flow conditions and the solid line represents the numerical predictions for the flow of a generalized Newtonian fluid.

Figure 4.13 shows a contour plot of the velocity magnitude measured using PIV at the center plane of the square duct ($y = 0$ or $z = 0$) for the flow of the viscoelastic fluid at $De = 76.3$ and $CR = 2.4$. The velocity magnitude is scaled with the downstream average axial velocity, U_2 .

As can be seen, the velocity magnitude increases significantly near the contraction plane, which corresponds to the overshoot in the axial velocity profile at that location (cf. Figure 4.12(a)). Moreover, we can also observe a large region with low velocities near the corners, a confirmation of the large recirculations observed at high De (cf. Figure 4.5), and the existence of off-center maximum of the velocity in the upstream region, a consequence of divergent flow.

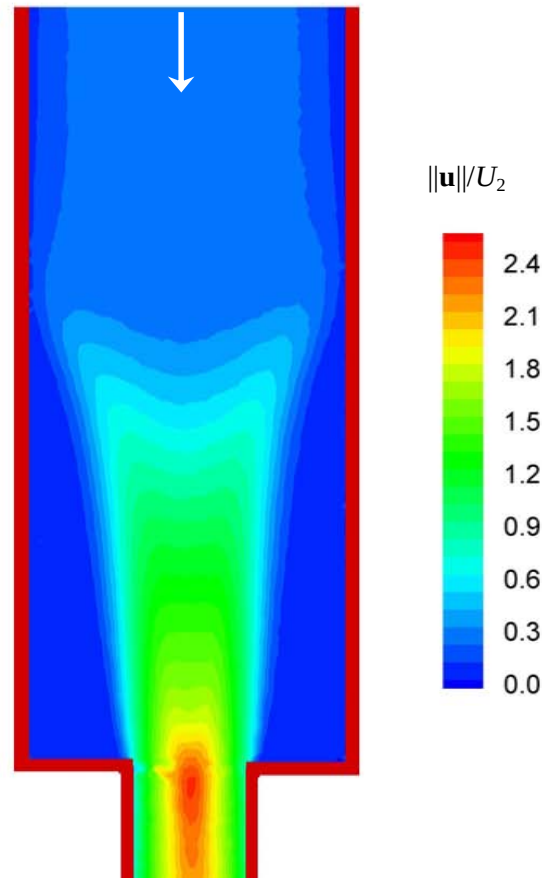


Figure 4.13. Contour plot of the normalized velocity magnitude for the shear-thinning viscoelastic fluid measure with PIV at $De = 76.3$, $Re = 0.670$ and $CR = 2.4$.

4.7. Conclusions

The flow of a viscoelastic fluid with a shear-thinning rheological behavior, through square-square contractions was studied experimental and numerically and compared with the flow of a Newtonian fluid. In order to assess the effect of the contraction ratio on the fluid flow and with the purpose of establishing benchmark solutions of the non-Newtonian fluid flow through

contractions with a square-square geometrical arrangement, different contraction ratios were investigated ($CR = 2.4, 4, 8$ and 12).

For the Newtonian fluid and for the range of contraction ratios studied, a vortex appears upstream of the contraction plane, which decreases in size with an increase of the flow inertia.

For the viscoelastic fluid flow a complex 3D corner vortex also appears. At low Deborah numbers, the flow is similar to that found for the Newtonian fluid but when the flow rate (or the Deborah number) is increased, the vortex length increases significantly for all contraction ratios studied. Furthermore, when elastic effects are present, the increase of the vortex length is accompanied by a reversal of the flow direction within the 3D open vortical structure. For the lower contraction ratios, $CR = 2.4$ and 4 , diverging streamlines appear when the Deborah number is increased. However, these diverging streamlines are not present in the flow through the $8:1$ and $12:1$ square-square contractions. Independently of CR , when elasticity is increased further, the flow eventually becomes time-dependent, with the size of the vortices varying in time.

Acknowledgements

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LAMINAR FLOW IN THREE-DIMENSIONAL SQUARE-SQUARE EXPANSIONS

Abstract

In this work we investigate the three-dimensional laminar flow of Newtonian and viscoelastic fluids through square-square expansions. The experimental results obtained in this simple geometry provide useful data for benchmarking purposes in complex three-dimensional flows. Visualizations of the flow patterns were performed using streak line photography, the velocity field of the flow was measured in detail using Particle Image Velocimetry and additionally, pressure drop measurements were carried out. The Newtonian fluid flow was investigated for the expansion ratios of 1:2.4, 1:4 and 1:8 and the experimental results were compared with numerical predictions. For all expansion ratios studied, a corner vortex is observed downstream of the expansion and an increase of the flow inertia leads to an enhancement of the vortex size. Good agreement is found between experimental and numerical results. The flow of the two non-Newtonian fluids studied, a Boger fluid and a shear-thinning viscoelastic fluid, was investigated experimental and numerically for expansion ratios of 1:2.4, 1:4, 1:8 and 1:12. For both viscoelastic fluids, a corner vortex appears downstream of the expansion, which decreases in size and strength when the elasticity of the flow is increased. For all fluids and expansion ratios studied, the recirculations that are formed downstream of the square-square expansion exhibit a three-dimensional structure evidenced by a helical flow, which is also well predicted in the numerical simulations.

5.1. Introduction

The flow of Newtonian and non-Newtonian fluids in channels with variable cross-section, which include contractions and expansions, is a classical fluid mechanics problem, which has been addressed in a number of experimental and numerical studies published in the literature (e.g. Boger, 1987; Rothstein and McKinley, 1999; Mompean and Deville, 2002; Nigen and Walters, 2002; Alves *et al.*, 2004; Rodd *et al.*, 2005; Oliveira *et al.*, 2007). The investigation of these flows is essential not only from a fundamental point of view but also due to their importance in practical applications, namely in the polymer processing industry. The flow through sudden contractions is a benchmark problem that has received broader attention than the flow through abrupt expansions, possibly due to the wide and interesting nature of the flow patterns that occur in contraction flows, particularly with viscoelastic fluids (e.g. vortex enhancement, divergent and unsteady flow) (Sousa *et al.*, 2009) (cf. Chapter 3), and its applicability to estimate the extensional viscosity in entry-flows (e.g. Cogswell, 1978; Binding, 1988). Nevertheless, expansion flows can also be used to understand entry-flow problems and to validate two and three-dimensional numerical codes.

Early experimental studies on expansion flows focused on Newtonian fluids in two-dimensional (2D) geometries (Hung and Macagno, 1966; Macagno and Hung, 1967; Durst *et al.*, 1974). It was found that Moffatt vortices (Moffatt, 1964) appear downstream of the expansion and that flow inertia promotes the enhancement of these recirculations. Later on, Acrivos and Schrader (1982) and Milos *et al.* (1987) investigated numerically the flow of Newtonian fluids at high values of the Reynolds number (Re) and they found that above a critical value the flow becomes unsteady. Furthermore, according to the experiments of Townsend and Walters (1994) and the numerical predictions of Baloch *et al.* (1995), a pair of lip vortices develops close to the re-entrant corner for high expansion ratios. These vortices then expand to the downstream wall increasing in size with the Reynolds number.

Townsend and Walters (1994) have also studied the flow behaviour of viscoelastic fluids through expansions, including the flow of a 0.15% aqueous solution of polyacrylamide (PAA) through 3:40 and 1:80 planar expansions, and the flow of a 0.1% aqueous solution of xanthan gum and glass fibers through three-dimensional (3D) axisymmetric expansions (expansion ratio of 3:40). For the viscoelastic fluids, recirculations also appear downstream of the expansion, but unlike the Newtonian case, these recirculations decrease in size when the elasticity of the flow is

increased. Baloch *et al.* (1996) used the linear form of the Phan-Thien and Tanner (PTT) model (1977, 1978) to numerically simulate a number of viscoelastic flows in planar (expansion ratio of 3:40 and 1:80) and axisymmetric (expansion ratio of 3:3:40) geometries. In agreement with the experimental work of Townsend and Walters (1994), the numerical results of Baloch *et al.* (1996) reported a decrease in the vortex activity due to viscoelasticity, which pushes the recirculations against the downstream corners as previously described by Halmos and Boger (1976). In particular, the viscoelastic fluid flow through planar expansions shows a reduction of the vortex size and strength as the Deborah number (De) is increased. Eventually, at high Deborah numbers, the recirculations downstream of the expansion seem to disappear altogether. This suppression mechanism has been compared to the extrudate-swell phenomenon, in which the polymer molecules passing through an expansion and entering a larger cross-section reservoir relax the elastic stresses by expanding the main flow stream tube (Poole *et al.*, 2007). Another interesting feature worth mentioning is the onset of a lip vortex in the inlet channel upstream of the expansion observed for Upper-Convected Maxwell (UCM) and Oldroyd-B fluids flowing through a 1:3 planar expansion under creeping flow conditions (Poole *et al.*, 2007). More recently, the same authors investigated the effect of the expansion ratio on the flow patterns of a UCM fluid (Poole *et al.*, 2007). For the lower expansion ratios, a non-monotonic variation of vortex length with De was predicted, with vortex reduction at low De , followed by vortex enhancement at higher De . For high expansion ratios ($ER > 3$), the vortex size was found to decrease monotonically as De increases.

There are several works that report the development of asymmetric flow in planar expansions for Newtonian and non-Newtonian fluids (e.g. Cherdron *et al.*, 1978; Fearn *et al.*, 1990; Oliveira, 2003; Manica and Bortoli, 2004; Neofytou, 2006; Rocha *et al.*, 2007; Oliveira *et al.*, 2008). Cherdron *et al.* (1978) used flow visualization and laser-Doppler anemometry techniques to quantify the steady asymmetry that develops for the Newtonian flow in symmetric planar sudden-expansion geometries, and concluded that the intensity of fluctuating energy measured in such low Reynolds number flows can even be larger than that observed in corresponding turbulent flows. Fearn *et al.* (1990) investigated experimental and numerically the Newtonian fluid flow through a symmetric sudden planar expansion ($ER = 3$) and found that the flow becomes asymmetric and time-dependent for high Reynolds numbers, and related this effect with the 3D structure that develops under those conditions. For generalized Newtonian fluids there are also a number of works that investigate the effect of shear-thinning and shear-thickening effects on the critical conditions for development of flow asymmetries in planar sudden expansion flows. Manica and Bortoli (2004) used a power-law model to study numerically the laminar flow

in a 1:3 planar expansion, and concluded that for shear thinning fluids the critical Reynolds number for onset of steady flow asymmetry increases as the power-law index (n) is reduced, while the opposite happens to shear thickening fluids as n increases. More recently, Neofytou (2006) used Casson and power-law rheological models to investigate the laminar flow in a 1:2 planar expansion. For both non-Newtonian fluids the numerical results showed a linear relation between the inverse dimensionless inlet wall shear stress, at the critical flow conditions for onset of asymmetry flow, and the corresponding dimensionless wall shear rate for the range of Power-Law index and Bingham numbers investigated.

There are also a few studies investigating the stabilizing effect of viscoelasticity on the critical conditions for onset of flow asymmetries in planar expansion flows. Oliveira (2003) studied numerically the flow of constant shear viscosity viscoelastic fluids (Boger fluids) through 1:3 planar expansions. The working fluids were a viscoelastic fluid that was described using the modified FENE-CR constitutive equation and a Newtonian fluid that was used with the purpose of validating the method and for comparison purposes. The numerical results showed that the flow becomes asymmetric for both fluids, however, for the viscoelastic case, the elasticity tends to stabilize the flow and the critical Reynolds number is higher than that found for a Newtonian fluid. More recently, Rocha *et al.* (2007) also investigated the onset of asymmetries in viscoelastic fluid flow through a 1:4 planar sudden expansion. The authors determined the critical Re at which the bifurcation phenomenon occurs, and concluded that it is lower than that obtained for lower expansion ratios (Oliveira, 2003). Asymmetries in the mean axial velocity profile of a shear-thinning fluid flowing through an axisymmetric abrupt expansion were also found in the work of Dales *et al.* (2005) in which the fluid was an aqueous solution of PAA and the flow regime was turbulent. However, in this case, the Newtonian fluid (water) flow did not show any asymmetries. More recently, another experimental study (Mullin *et al.*, 2009) showed that for a 1:2 axisymmetric expansion the Newtonian fluid flow also becomes asymmetric (and steady), under laminar flow conditions, above $Re = 1139$. For $Re > 1400$ the flow eventually becomes time-dependent.

In most of the planar expansion flows referenced above, three-dimensional effects are negligible and less demanding 2D numerical simulations can be used to predict the flow correctly. Configurations where three-dimensional effects are important are more challenging to analyse experimentally and numerically and as such, the number of published works concerning this topic is reduced. Two important exceptions are the works of Burgos and Alexandrou (1999) and Alexandrou *et al.* (2001) in which the flow of a generalized non-Newtonian fluid (Herschel-

Bulkley) through three-dimensional 1:2 and 1:4 expansions was investigated. In the first work (Burgos and Alexandrou, 1999), the unsteady flow in a 1:2 sudden expansion was studied and in the latter (Alexandrou *et al.*, 2001), the steady flow of Herschel-Bulkley fluids was investigated. The authors showed that the development of yielded and unyielded regions in this type of 3D geometries depends on the expansion ratio and on Bingham and Reynolds numbers.

In this work, we focus on the flow through a three-dimensional square-square expansion, with different expansion ratios ($ER = 2.4, 4, 8$ and 12 , defined as the ratio between the side lengths of the upstream and downstream square ducts). We use Newtonian and viscoelastic fluids with different rheological characteristics: a Boger fluid, for which the shear viscosity is nearly independent of the shear rate and a shear-thinning viscoelastic fluid. Visualizations were undertaken in order to assess the flow patterns and detailed particle image velocimetry (PIV) measurements were performed to quantify the velocity field. For the Boger fluid, pressure drop measurements are also presented and discussed. Moreover, 3D numerical simulations were carried out in order to predict the Newtonian and non-Newtonian fluid flow through square-square expansions.

5.2. Experimental Techniques

5.2.1. Experimental Set-up

A scheme of the experimental set-up is shown in Figure 5.1. The main test section in the experimental rig has a square cross-section of length $L = 1.75$ m, which is composed of a downstream square channel, with a side length of $2H_2 = 24.0$ mm, and an upstream interchangeable square channel which fits inside the larger one and has a smaller side length. The internal side length of the inner square part can be set to $2H_1 = 10.0$ mm, 6 mm, 3 mm or 2 mm in order to obtain the desired expansion ratios, $ER = H_2/H_1 = 2.4, 4, 8$ or 12 .

The upstream and downstream sections are denoted by subscripts 1 and 2, respectively. For optical access, the channels are made of transparent perspex.

The flow rate is set by adjusting the difference between the liquid levels in the two reservoirs, applying vacuum to the left-hand side reservoir using a vacuum pump (KNF Loboport N811-KT.18) and selecting the diameter of the pipe that connects the channel to the outflow reservoir. We do not use valves to regulate the flow rate to avoid degradation of the viscoelastic fluids in the narrow passage of the valve. The right-hand side reservoir was used to supply the fluid that

flowed through the column, and it was placed on a weighting scale (KERN DS 16k0.2; with readout of 0.2 g and maximum range of 16 kg) in order to measure the mass flow rate ($Q_m \equiv \Delta \text{Weight} / \Delta \text{Time}$). More details of the experimental set-up can be found in (Sousa *et al.*, 2009) (cf. Chapter 3).

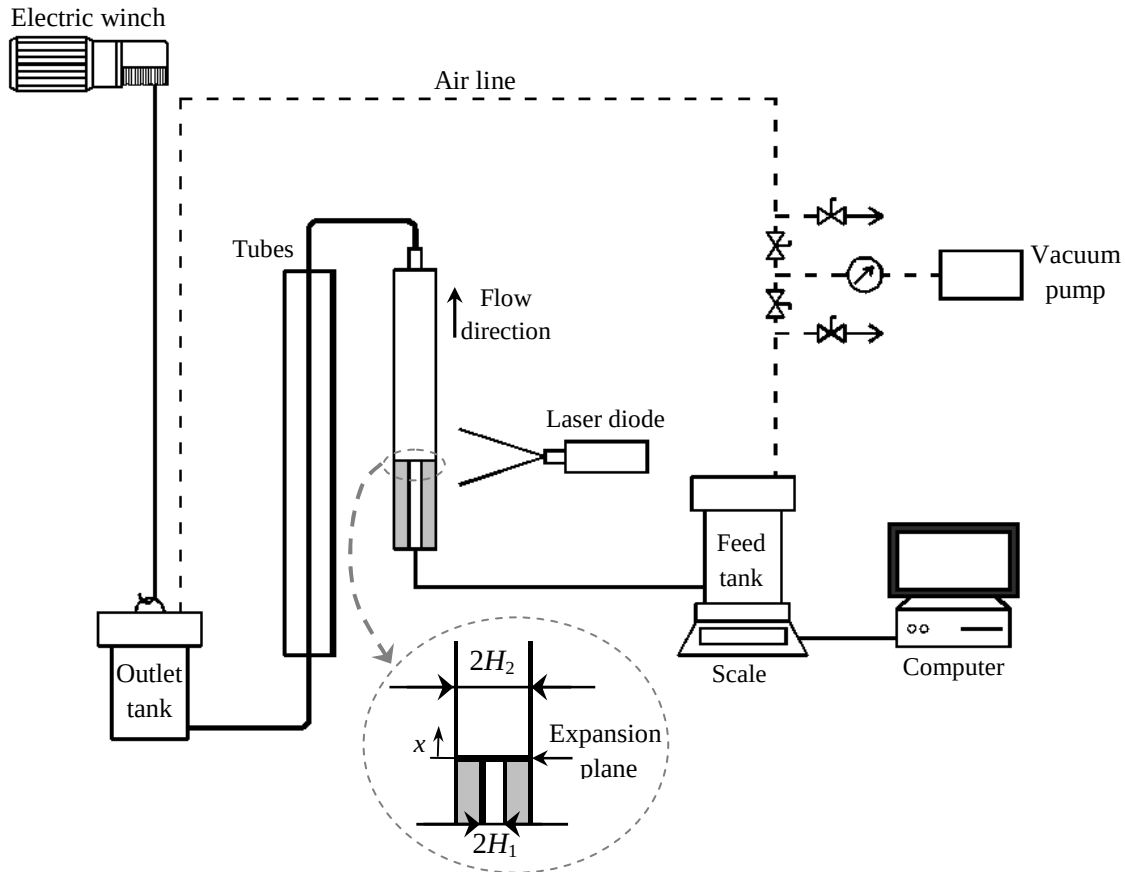


Figure 5.1. Schematic diagram of the experimental set-up.

5.2.2. Measurement Techniques

In this study, two different optical techniques were used in order to characterize the flow field: streak line photography and particle image velocimetry. For this purpose, the fluids used in the experiments were seeded with 10 μm PVC tracer particles.

Streak line photography uses long time exposures to obtain a visual fingerprint of the flow patterns. For flow illumination, we used a 365 nm 5 mW laser diode (Vector, model 5200-20) or a 532 nm 3mW laser diode (Imatronic, model LLM115). In both cases the diode also has a cylindrical lens to generate a sheet of light. The streak line images were captured using a digital camera (Canon EOS 30 D) with a macro lens (Canon EF100mm, f/2.8), placed perpendicularly to the light sheet.

For the measurement of the velocity field, a doubled pulsed Nd:YAG laser, with a maximum energy of 50 mJ (Solo PIV III from New Wave Research) was used. The laser was combined with appropriate optical components, producing a planar light sheet that illuminates the plane under study. The images were recorded using a digital CCD camera (Flow Sense 2M from Dantec Dynamics equipped with a Nikon AF Micro Nikkor 60 mm f/2.8D lens) which was placed perpendicularly to the light sheet. The images were processed using FlowManager v4.60 software (Dantec Dynamics) and the velocity vector map for each pair of images was determined using a cross-correlation technique. A detailed discussion of the technique employed can be found in (Keane and Adrian, 1992).

The flow visualizations and the velocity field measurements using PIV were both undertaken at different parallel planes of the flow in the square channel. Therefore, it was necessary to translate simultaneously the light source and the camera utilized for each optical technique using for this purpose a x-y traverse and a dial comparator (readout of ± 0.01 mm).

Pressure drop measurements across the expansion were carried out using differential pressure sensors (Honeywell, model 26PC series), previously calibrated using a hydrostatic column of water, which are able to cover pressures up to 6.9 kPa. The experiments were performed by determining the pressure drop (Δp) across the expansion, with one pressure port located upstream (p_1) and another port (p_2) located downstream of the expansion plane. The precise locations of the pressure ports are listed in Table 5.1 for different ER . For each flow rate studied, the output signal of the pressure transducer was recorded using LabView v7.1 software, until the steady-state was reached.

Table 5.1. Pressure port locations in the x-direction (streamwise) for each expansion ratio. The expansion plane is located at $x = 0$.

	x_1 / mm	x_2 / mm
$ER = 4$	-99	152
$ER = 8$	-48	153
$ER = 12$	-29	154

5.3. Rheological Characterization

A Newtonian and two different non-Newtonian fluids were used in the experiments. Table 5.2 summarizes the composition and density (ρ) of the fluids, measured at 293.2 K using a hydrometer (readability of 0.001 kg/m³; range 1200-1300 kg/m³). A biocide (Kathon LXE, Rohm and Haas) was added to the solutions, at a weight concentration of 25 ppm, in order to minimize bacterial growth and degradation of the fluids.

Furthermore, the working fluids were characterized rheologically at different temperatures in the range of $283.2 \leq T / \text{K} \leq 303.2$ using a shear rheometer (MCR301, Anton Paar) and the time-temperature superposition method was used to obtain a master flow curve.

Table 5.2. Composition by weight and density of the fluids measured at 293.2 K.

Fluid	PAA [ppm]	Glycerin [%]	Water [%]	Kathon [ppm]	NaCl [%]	ρ [kg m ⁻³]
Newtonian	-	84.99	15.01	25	-	1221
Boger	100	90.96	7.52	25	1.5	1249
Shear-Thinning	600	59.94	40.00	25	-	1156

The variation of the viscosity with temperature can be described using an Arrhenius equation (Bird *et al.*, 1987):

$$\ln(a_T) = \frac{\Delta H}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right), \quad (5.1)$$

where a_T is the shift factor, ΔH is an activation energy, R is the universal gas constant, T is the absolute temperature of the measurement and T_0 is the absolute reference temperature, which is set here as $T_0 = 293.2$ K, the temperature at which the experiments in the square-square expansion were performed. The shift factor is defined as (Bird *et al.*, 1987):

$$a_T = \frac{\eta(T)}{\eta(T_0)} \frac{T_0}{T} \frac{\rho_0}{\rho}, \quad (5.2)$$

where $\eta(T)$ and ρ are the shear viscosity and the fluid density at temperature T and $\eta(T_0)$ and ρ_0 are the shear viscosity and the fluid density at the reference temperature T_0 . Since the range of temperatures of the measurements is small, the shift factor can be simplified to:

$$a_T = \frac{\eta(T)}{\eta(T_0)}. \quad (5.3)$$

For the Newtonian fluid, the shear viscosity at the reference temperature is $\eta(T_0) = 0.0982$ Pa s and $\Delta H/R = 5580$ K. For the non-Newtonian fluids we present in Figure 5.2 the master curves measured for steady shear flow.

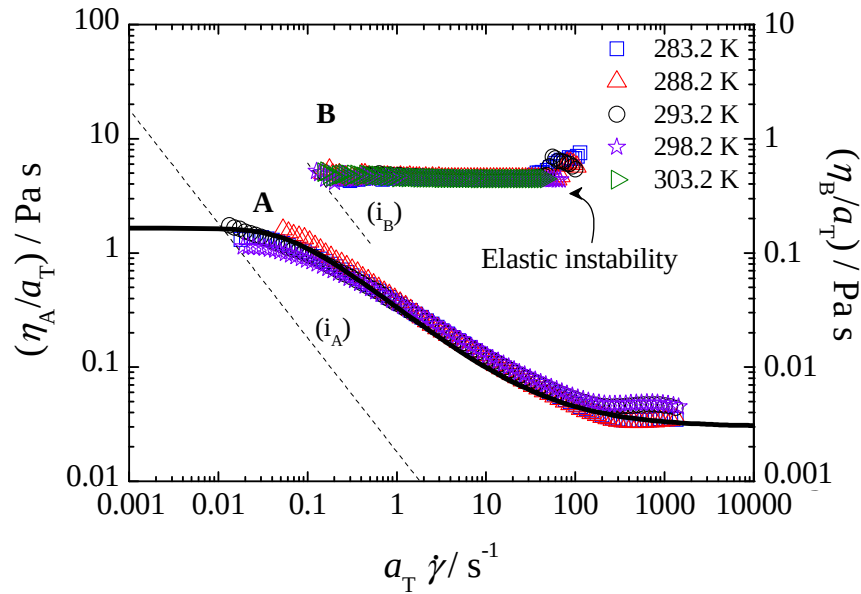


Figure 5.2. Master curves of the steady shear viscosity for the Boger (B) and shear-thinning (A) fluids used (symbols). The solid line represents the fit of the PTT model to the experimental data of the shear-thinning fluid. The minimum measurable shear viscosity, determined from 20× the minimum measurable torque of the rheometer, is shown by dashed lines for the shear-thinning fluid (i_A) and the Boger fluid (i_B).

For the Boger fluid, we obtained $\Delta H/R = 6780$ K for the range of temperatures between 283.2 and 303.2 K. Furthermore, the experimental shear data was fitted using a three-mode model with a solvent contribution based on measurements under low amplitude oscillatory shear flow (cf. Sousa *et al.* (2009) [Chapter 3] for details). At the reference temperature, $T_0 = 293.2$ K, the values of the relaxation time of each mode are $\lambda_i = 4, 0.4$ and 0.04 s and the values of the corresponding shear viscosities are $\eta_i = 0.225, 0.04$ and 0.014 Pa s, for $i = 1, 2$ and 3 . In order to

determine an average relaxation time, the Oldroyd-B model was used: the polymeric contribution to the shear viscosity is $\eta_p = 0.279 \text{ Pa s}$ ($\eta_p = \sum_{i=1}^3 \eta_i$) and the solvent viscosity is $\eta_s = 0.367 \text{ Pa s}$.

The resulting average relaxation time is $\lambda = 3.29 \text{ s}$ ($\lambda = \sum_{i=1}^3 \eta_i \lambda_i / \eta_p$). More details on the characterization of the Boger fluid can be found in (Sousa *et al.*, 2009) (cf. Chapter 3).

For the shear-thinning fluid, we fitted the experimental shear data using a simplified Phan-Thien-Tanner (PTT) model with a solvent contribution (Phan-Thien and Tanner, 1977; Phan-Thien, 1978) which is also shown in Figure 5.2. The parameters of the PTT model are: zero-shear polymer viscosity, $\eta_p = 1.62 \text{ Pa s}$; solvent viscosity, $\eta_s = 0.03 \text{ Pa s}$; relaxation time, $\lambda = 32 \text{ s}$; extensibility parameter, $\varepsilon = 0.06$. A description of the PTT model used in the numerical simulations can be found in Section 5.4.

5.4. Numerical Method and computational meshes

The flow through square-square expansions with different expansion ratios ($ER = 2.4, 4, 8$ and 12) was simulated numerically using a fully-implicit finite-volume method with a time marching pressure-correction algorithm (Oliveira *et al.*, 1998). The governing equations that describe an isothermal, laminar and incompressible fluid flow are those of conservation of mass and momentum:

$$\nabla \cdot \mathbf{u} = 0, \quad (5.4)$$

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + \nabla \cdot \mathbf{u} \mathbf{u} \right] = -\nabla p + \eta_s \nabla^2 \mathbf{u} + \nabla \cdot \boldsymbol{\tau}, \quad (5.5)$$

where $\mathbf{u}$ is the velocity vector, t the time, p the pressure and η_s the Newtonian solvent viscosity.

The extra-stress tensor ($\boldsymbol{\tau}_t$) is described by the sum of a Newtonian ($\boldsymbol{\tau}_s = \eta_s (\nabla \mathbf{u} + \nabla \mathbf{u}^T)$) and a polymeric solute contribution ($\boldsymbol{\tau}$). In the numerical simulations of the Newtonian fluid flow, the polymeric contribution to the extra stress tensor is null ($\nabla \cdot \boldsymbol{\tau} = \mathbf{0}$) and only the Newtonian component remains in Eq. (5.5). On the other hand, for the numerical simulations of the viscoelastic fluid flow, the polymeric contribution is included and is described using an appropriate rheological constitutive equation. For a PTT fluid, the polymeric contribution to the extra-stress tensor is given by (Phan-Thien and Tanner, 1977; Phan-Thien, 1978):

$$f(\text{Tr } \boldsymbol{\tau}) \boldsymbol{\tau} + \lambda \left(\frac{\partial \boldsymbol{\tau}}{\partial t} + \nabla \cdot \mathbf{u} \boldsymbol{\tau} \right) = \eta_p (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \lambda (\boldsymbol{\tau} \cdot \nabla \mathbf{u} + \nabla \mathbf{u}^T \cdot \boldsymbol{\tau}). \quad (5.6)$$

Here, the stress function $f(\text{Tr } \boldsymbol{\tau})$ takes the linear form (Phan-Thien and Tanner, 1977):

$$f(\text{Tr } \boldsymbol{\tau}) = 1 + \frac{\lambda \varepsilon}{\eta_p} \text{Tr}(\boldsymbol{\tau}), \quad (5.7)$$

where λ is the relaxation time, η_p is the zero-shear viscosity of the polymer, ε is the extensibility parameter and $\text{Tr}(\boldsymbol{\tau})$ represents the trace of tensor $\boldsymbol{\tau}$. The Oldroyd-B model is obtained setting $\varepsilon = 0$ (i.e. $f(\text{Tr } \boldsymbol{\tau}) = 1$).

In this study, the Boger fluid flow was simulated using the Oldroyd-B model, with $\varepsilon = 0$, $\lambda = 3.29$ s, $\eta_p = 0.279$ Pa s and $\eta_s = 0.367$ Pa s as described in Section 5.3. For the shear-thinning fluid flow, the numerical simulations were performed using the PTT model with one mode and a solvent contribution ($\eta_p = 1.62$ Pa s; $\eta_s = 0.03$ Pa s; $\lambda = 32$ s; $\varepsilon = 0.06$).

Eqs (5.4) - (5.6) are integrated in space over the computational cells of the mesh and in time over a small time step (δt). The time derivative is discretized with an implicit first-order Euler scheme, the diffusive terms are discretized with second-order central differences and the discretization of the advective terms, both in the momentum and constitutive equations, is done using the CUBISTA high-resolution scheme (Alves *et al.*, 2003).

The computational meshes used in this study for the numerical simulation of the flow through square-square expansions are composed of three-dimensional orthogonal blocks and non-uniform cells that follow a geometrical progression within each direction. For the expansion ratios of 2.4, 4 and 8, the numerical simulations were carried out using three different meshes: mesh M40 which has 40 cells in y and z directions of the downstream channel, mesh M64 with 64 cells in y and z directions and a more refined mesh, M80 which has 80 cells in each of the three orthogonal directions. For the expansion ratio of 12, simulations were performed using three meshes with a slight different number of cells: mesh M40, M60 and M96, which have 40, 60 and 96 cells in the y and z directions of the downstream channel, respectively. The total number of cells (NC) and the dimensionless minimum cell size ($\Delta x_{\min}/(2H_2)$, $\Delta y_{\min}/(2H_2)$ and $\Delta z_{\min}/(2H_2)$) of the meshes used for all the ER are presented in Table 5.3. In Figure 5.3 we show two meshes used in the numerical simulations of the flow through the 1:8 square-square expansion.

In spite of the geometrical symmetry relative to the central planes ($y = 0$ and $z = 0$) and diagonal planes ($z = \pm y$), all simulations were performed on meshes covering the whole wall-to-wall geometries.

Regarding the boundary conditions, no-slip condition at the solid walls was imposed and the inlets and outlets were positioned far from the expansion plane so that fully developed flow conditions were enforced. At the outlet, we impose Neumann boundary conditions for velocity and stresses, while pressure is linearly extrapolated from the two upstream cell values.

Table 5.3. Characteristics of the computational meshes used (NC: number of computational cells).

ER	Mesh	NC	$\Delta x_{\min}/(2H_2)$	$\Delta y_{\min}/(2H_2) = \Delta z_{\min}/(2H_2)$
2.4	M40	164000	2.08×10^{-2}	2.04×10^{-2}
	M64	419840	1.39×10^{-2}	1.25×10^{-2}
	M80	656000	1.03×10^{-2}	9.93×10^{-3}
4	M40	51000	1.31×10^{-2}	1.45×10^{-2}
	M64	130560	8.20×10^{-3}	8.16×10^{-3}
	M80	408000	6.26×10^{-3}	6.25×10^{-3}
8	M40	163200	7.50×10^{-3}	8.69×10^{-3}
	M64	417792	4.69×10^{-3}	4.55×10^{-3}
	M80	652800	3.75×10^{-3}	3.75×10^{-3}
12	M48	113664	1.08×10^{-2}	9.52×10^{-3}
	M60	177600	8.61×10^{-3}	8.33×10^{-3}
	M96	909312	1.09×10^{-3}	1.35×10^{-3}

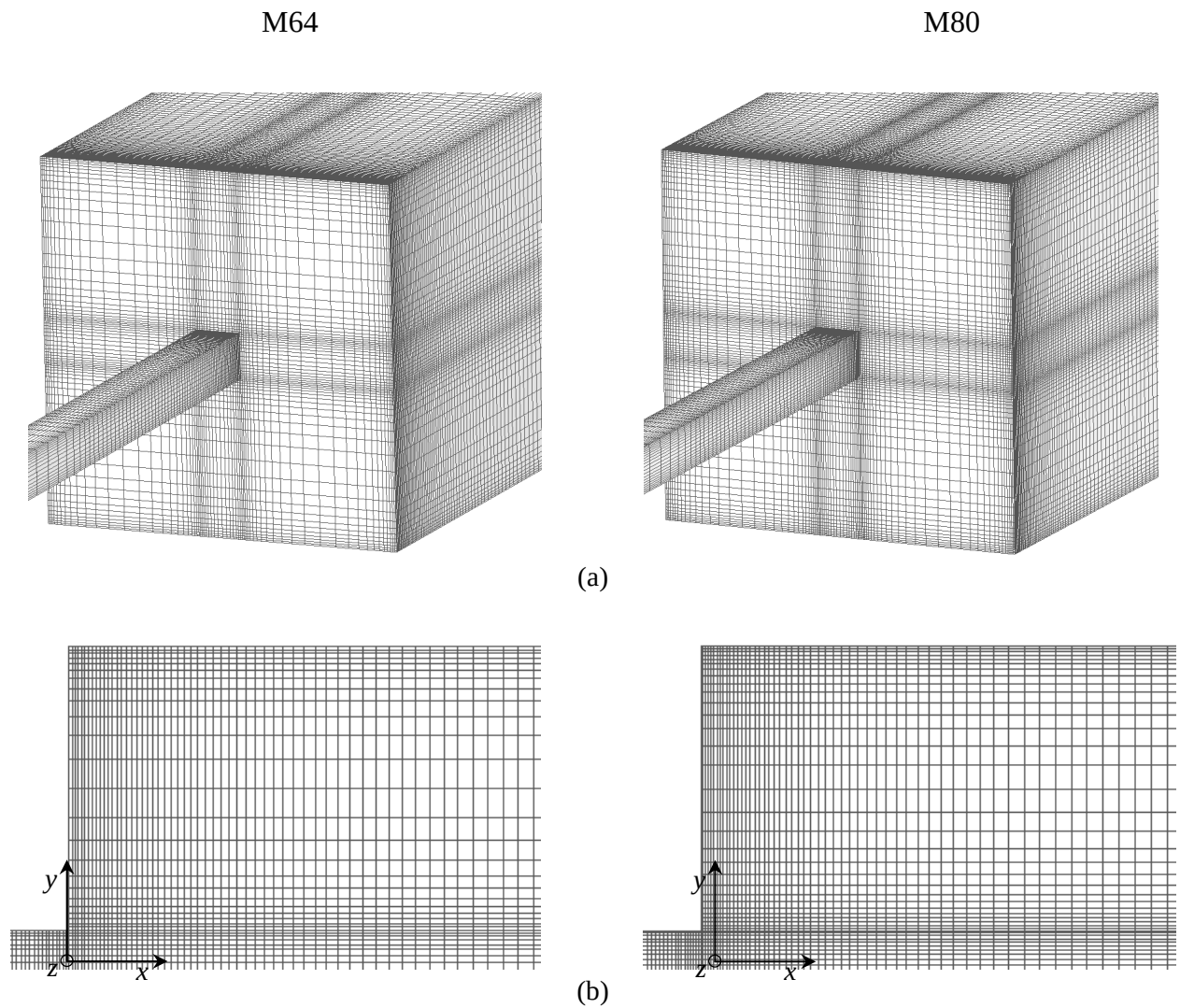


Figure 5.3. Zoomed view of the meshes M64 and M80 used in the numerical simulations of the flow through the 1:8 square-square abrupt expansion. (a) 3D view; (b) xy centerplane ($z = 0$).

5.5. Flow patterns and vortex length

The flow of the Newtonian and viscoelastic fluids through square-square expansions, with different expansion ratios, was investigated in terms of the vortex length and flow patterns for a wide range of flow rates. For this purpose, different parallel planes of the channel were investigated using flow visualization.

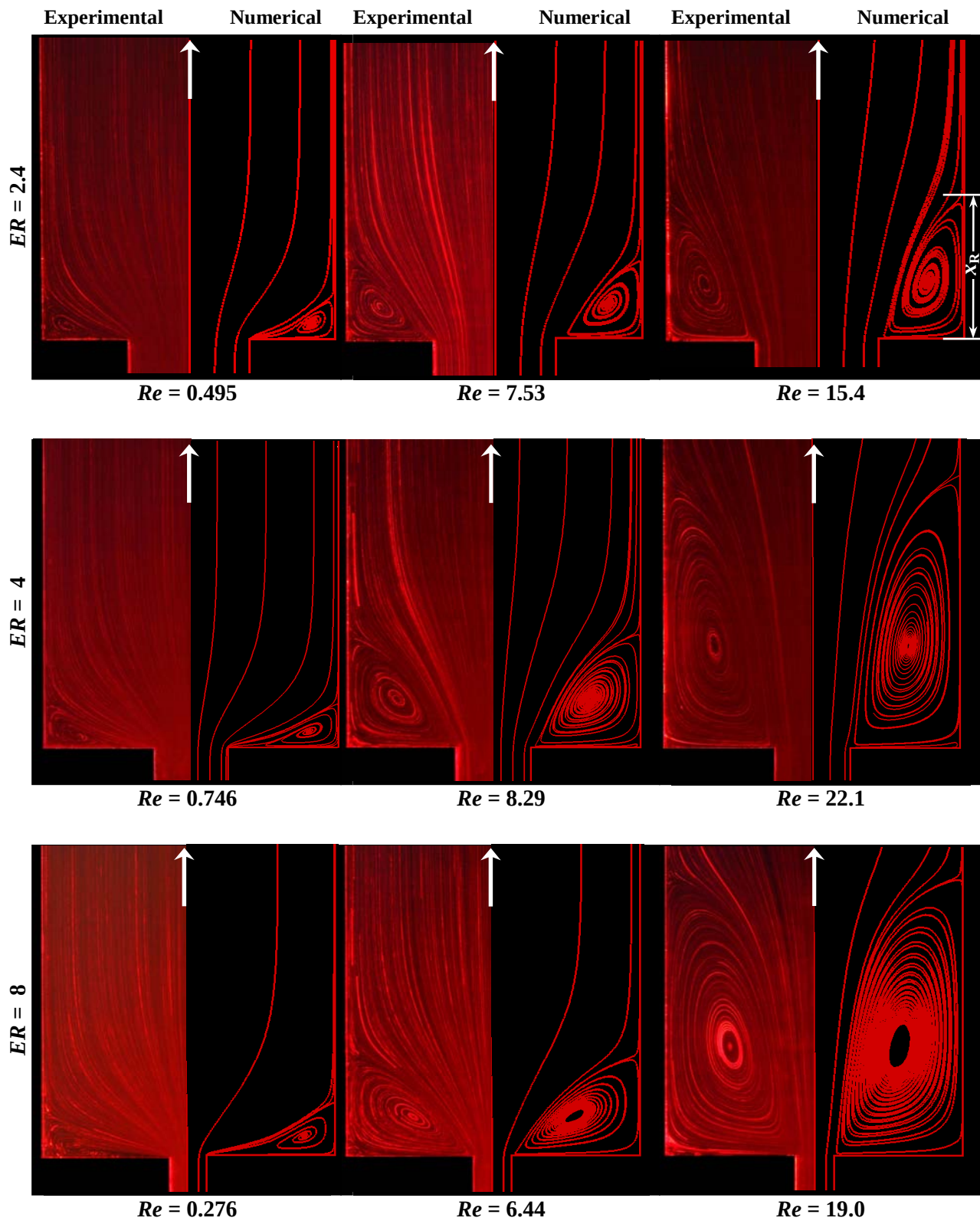


Figure 5.4. Experimental and numerical flow patterns at the middle plane ($y = 0$ or $z = 0$) for the Newtonian fluid at different Reynolds number and expansion ratios.

5.5.1. Inertial effects

In order to study the vortex growth dynamics of the Newtonian fluid with inertia, visualizations of the flow patterns near the expansion plane were performed for a wide range of flow rates ($Re < 20$) and various expansion ratios ($ER = 2.4, 4$ and 8). Here, the Reynolds number is based on upstream flow characteristics and defined as $Re = \rho U_1(2H_1)/\eta$, where U_1 is the average velocity in the upstream channel. For the shear-thinning fluid, the viscosity is calculated using the PTT model at a characteristic shear-rate ($\dot{\gamma} = U_1/H_1$).

In Figure 5.4 we compare the experimental and numerical flow patterns obtained at the center plane for all expansion ratios studied. The numerical predictions were carried out with the refined mesh (M80) in order to ensure high accuracy.

As can be observed in Figure 5.4, a Moffatt vortex (Moffatt, 1964) is formed downstream of the expansion plane, for all expansion ratios. Increasing the flow inertia leads to an increase of the vortex size and an excellent agreement between the experimental results and numerical predictions is found. For Newtonian fluids, the formation and dynamics of the recirculations that appear downstream of an abrupt expansion is well-documented in the literature (e.g. Cherdron *et al.*, 1978; Townsend and Walters, 1994; Baloch *et al.*, 1995; Poole *et al.*, 2009).

In Figure 5.5 we analyze the effect of flow inertia on the measured normalized vortex length, $x_R/(2H_2)$ (c.f. Figure 5.4 for $Re = 15.4$ and $ER = 4$) for the Newtonian fluid. Moreover, we also present the numerical predictions obtained with the three meshes (M40, M64 and M80) used for each expansion ratio. For all expansion ratios studied, the vortex size increases monotonically when the Reynolds number is increased. For the whole range of Re studied, the numerical results obtained using meshes M40, M64 and M80 are in good agreement with the experimental results. Furthermore, the differences between the three meshes are small and the highest deviation between the results obtained is below 2 %, demonstrating the good accuracy of the numerical method. For creeping flow conditions (i.e. in the limit when $Re \rightarrow 0$) the numerical simulations predict the following vortex dimensions: $x_R/(2H_1) = 0.141$ for $ER = 2.4$; $x_R/(2H_1) = 0.163$ for $ER = 4$; $x_R/(2H_1) = 0.174$ for $ER = 8$. These results are in agreement with the predictions for square-square contractions flows under creeping flow conditions (cf. Alves *et al.*, 2005; Alves *et al.*, 2008; Sousa *et al.*, 2009 [Chapter 3]) as consequence of the reversibility of Newtonian inertialess flows, i.e. in the limit when $Re \rightarrow 0$ the flow patterns for expansion and contractions flows are indistinguishable.

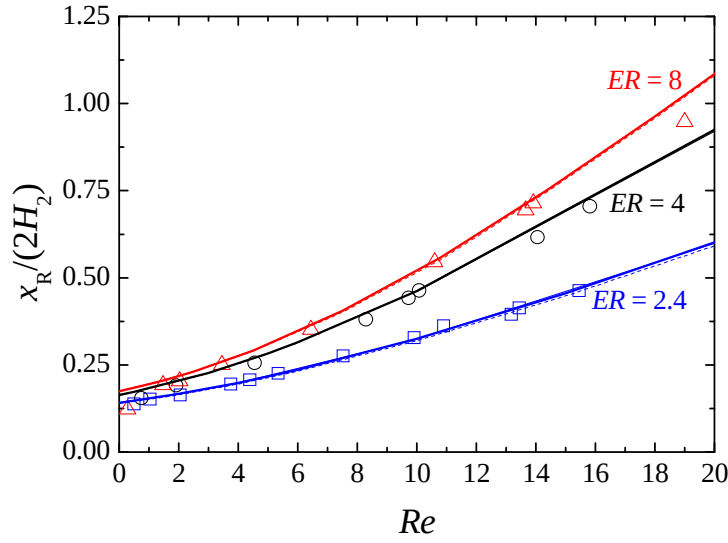


Figure 5.5. Dimensionless vortex length as a function of the Reynolds number for $ER = 2.4$, 4 and 8. The symbols represent the experimental data, the dashed lines represent the numerical predictions performed using mesh M40, the thin solid lines the numerical results obtained with mesh M64 and the thick solid lines the predictions using the more refined mesh, M80.

5.5.2. Elastic effects

In order to quantify the effect of viscoelasticity, we use the Deborah number, here defined based on upstream flow conditions, $De = \lambda U_1/H_1$. Figure 5.6 shows the flow patterns of both viscoelastic fluids obtained experimentally at the middle plane ($y = 0$ or $z = 0$), for a range of Deborah numbers (or flow rates), using the Boger and the shear-thinning fluids flowing through the square-square expansions for the range of ER studied.

Comparing the flow patterns of both viscoelastic fluids, several similarities can be identified. For all expansion ratios studied, the flow of the two viscoelastic fluids presents a corner vortex downstream of the expansion plane and, in general, increasing the Deborah number leads to a decrease of the corner vortex length.

The dependence of the vortex length on the Deborah number, for all expansion ratios studied, is quantified in Figure 5.7(a) for the Boger fluid and in Figure 5.7(b) for the shear-thinning fluid. The vortex length is scaled with the downstream width of the channel ($2H_2$). In addition, in Figure 5.7 we also show the predictions obtained from the numerical simulations.

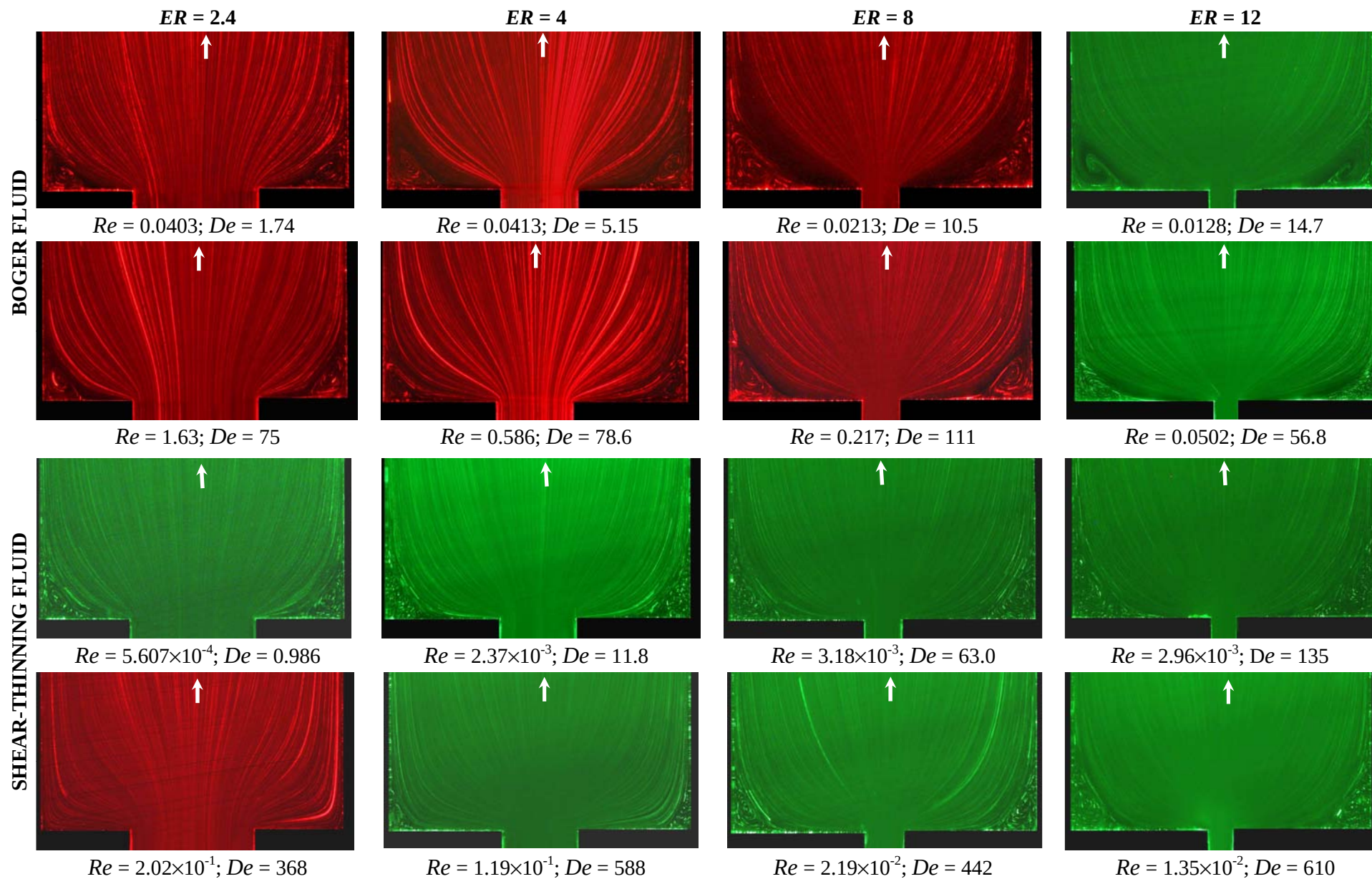


Figure 5.6. Effect of elasticity on the Boger and shear-thinning fluid flow patterns at the middle plane for different expansion ratios.

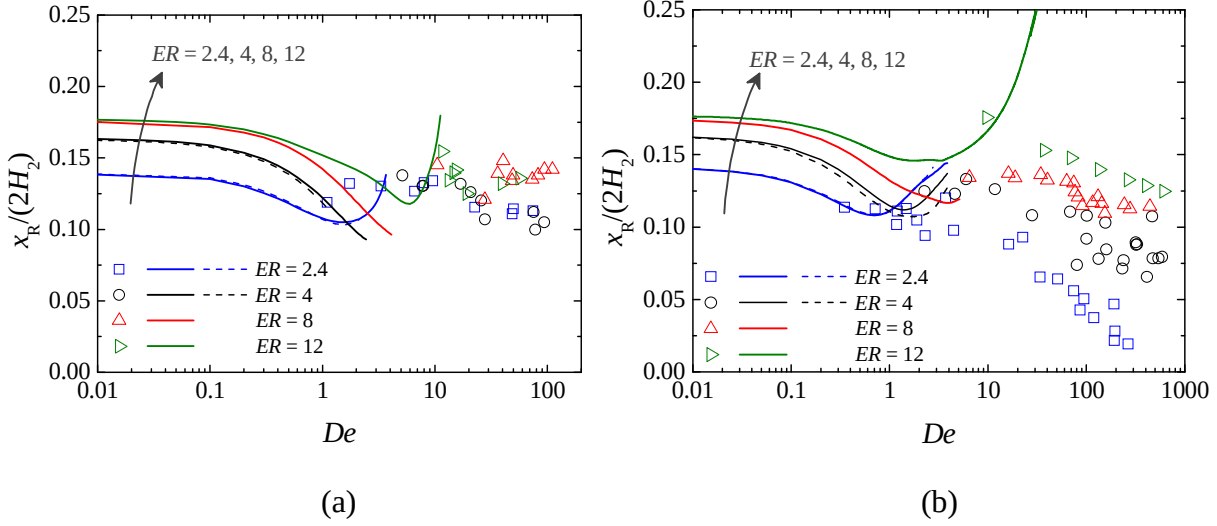


Figure 5.7. Dimensionless vortex length as a function of the Deborah number for the Boger (a) and shear-thinning (b) fluid flows for all expansion ratios studied. The symbols represent experimental data, the solid lines represent the numerical predictions using mesh M64 for $ER = 2.4, 4$ and 8 or mesh M60 for $ER = 12$ and the dashed lines represent the numerical predictions using mesh M80 for $ER = 2.4$ and 4 .

For the lower expansion ratios, $ER = 2.4$ and 4 , the numerical predictions of the viscoelastic fluid flow were obtained using meshes M64 and M80 and there is a good agreement between the numerical results using both computational meshes. For this reason, and since the computational time required for the runs with more refined meshes is substantially higher, for the simulations of the viscoelastic fluid flow through the 1:8 and 1:12 square-square expansions we used only mesh M64 or M60, respectively. Note that besides the high relaxation times used, specially for the shear-thinning fluid, the enlargement of the number of cells for the refined meshes (e.g. for $ER = 12$, the more refined mesh, M96, has five times more cells than mesh M60) renders the simulations of the viscoelastic fluid flow through 3D expansions with a high expansion ratio unfeasible in practice due to the very large CPU times involved (of the order of weeks for the highest De).

At low Deborah numbers the numerical simulations predict accurately the creeping flow Newtonian plateaus for all expansion ratios ($x_R/(2H_1) = 0.141$ for $ER = 2.4$; $x_R/(2H_1) = 0.163$ for $ER = 4$; $x_R/(2H_1) = 0.174$ for $ER = 8$; $x_R/(2H_1) = 0.177$ for $ER = 12$), which are not evident from the experimental results. Increasing De , the numerical predictions show a decrease followed by an increase of the vortex length. Above a critical Deborah number, which increases with ER , we are no longer able to obtain converged numerical simulations, in contrast with the experimental results that are steady up to much higher De , and show a progressive decrease of the vortex

length as the flow rate (or De) is increased for both viscoelastic fluids. However, for the shear-thinning fluid, the decrease of the vortex size with De is more pronounced, in particular for the lower expansion ratios (cf. Figure 5.7(b) for $ER = 2.4$). The limitations of the viscoelastic models used, and in particular the numerically stringent Oldroyd-B model, are revealed by the significantly lower De numbers attained in the simulations, and by the inaccuracy that is obtained at the largest De where steady solutions are obtained numerically. Still, the simulations are able to predict qualitatively the initial trend of vortex suppression due to elasticity.

5.5.3. Three-dimensionality of the flow

The previous investigations regarding the flow of Newtonian and viscoelastic fluids, through square-square contractions (Alves *et al.*, 2005; Alves *et al.*, 2008; Sousa *et al.*, 2009 (cf. Chapter 3)), demonstrated that the flow in these geometries is highly three-dimensional. In order to study the 3D nature of the flow through square-square expansions, a detailed investigation at several parallel planes was carried out. Figure 5.8 shows the visualized path lines and the corresponding numerical predictions for the Newtonian fluid flow at different planes of the 1:8 square-square expansion, ranging from the center plane ($y/H_2 = 0$ or $z/H_2 = 0$) to a plane near the wall of the channel ($y/H_2 = \pm 0.875$ or $z/H_2 = \pm 0.875$). At the center plane the pathlines illustrated are real, while for the other planes the visualized flow shows the projections of the pathlines at the visualized plane. Again, we observe an excellent agreement between the visualizations and the numerical predictions.

For the range of flow rates studied, the flow is symmetric relative to the two center planes ($y = 0$ and $z = 0$) and to the two diagonal planes ($y = \pm z$). To further document the complex flow behavior in square-square expansions, in Figure 5.9 we show a three-dimensional view of some streamlines, showing the open vortical structures predicted numerically with the refined mesh (M80) for the Newtonian fluid at creeping flow conditions ($Re \rightarrow 0$ and $CR = 4$).

To simulate creeping flow conditions we neglect the convective term in the momentum equation, but keep the transient term ($\rho \partial \mathbf{u} / \partial t$) and use a pseudo-time marching algorithm to achieve steady flow conditions. When steady-state is achieved, the transient term vanishes, and we obtain exactly creeping flow conditions ($Re = 0$).

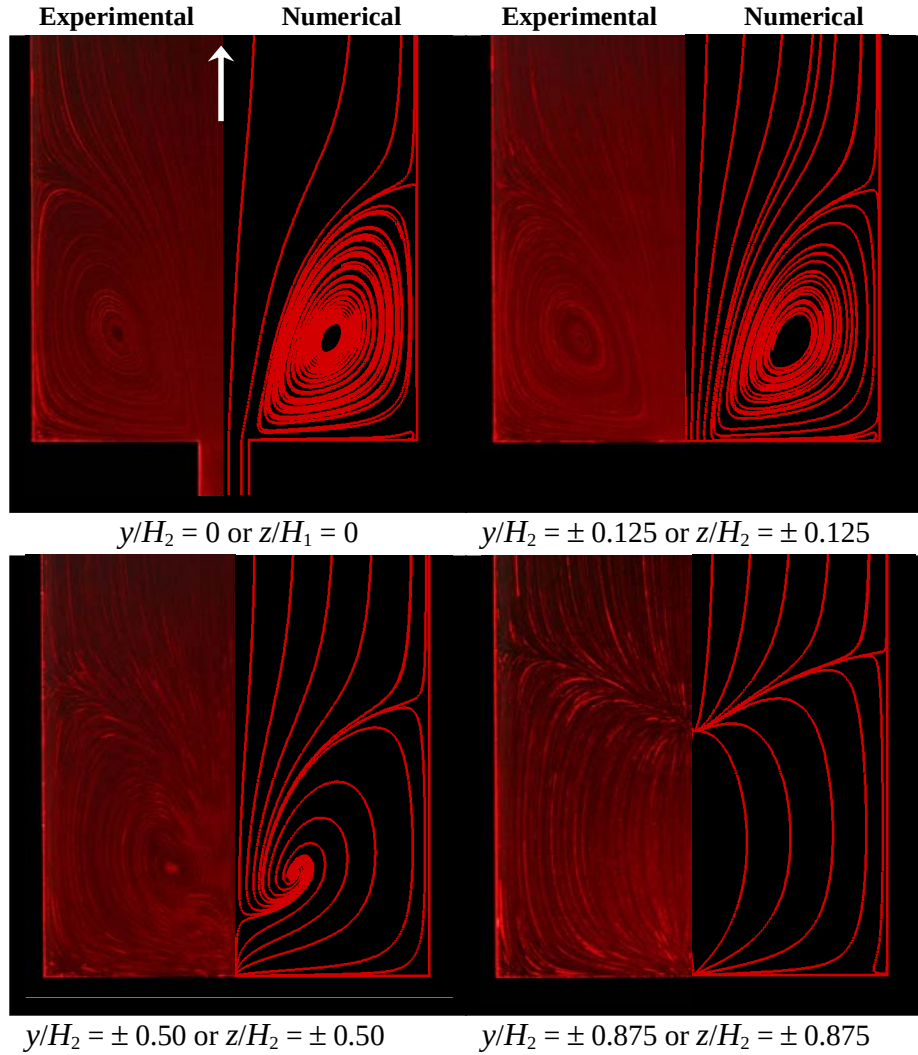


Figure 5.8. Experimental and numerical projected pathlines of the Newtonian fluid flow at different planes of the 1:8 square-square expansion for $Re = 13.9$.

Under creeping flow, the streamlines generated by the Newtonian fluid flowing through a square-square expansion are coincident with those documented previously for the flow through square-square contractions (Alves *et al.*, 2005; Alves *et al.*, 2008; Sousa *et al.*, 2009 (cf. Chapter 3)) due to the reversibility of inertialess flows. However, in the present configuration the flow occurs in the opposite direction, causing the fluid that is flowing near the center plane wall of the upstream channel to pass through the expansion plane and enter the recirculation in the center plane (plane EFGH in Figure 5.9). Once there, the fluid rotates around the center of the recirculation and follows a helical trajectory toward the diagonal plane (ABCD), where it turns now to the periphery and exits the recirculation moving toward the exit of the downstream channel close to the diagonal plane wall, as illustrated in Figure 5.9(c). The streamlines in the

center and diagonal planes are shown in Figure 5.9(a) and 5.9(b), respectively, to better illustrate the dynamics of the secondary flow in the symmetry planes.

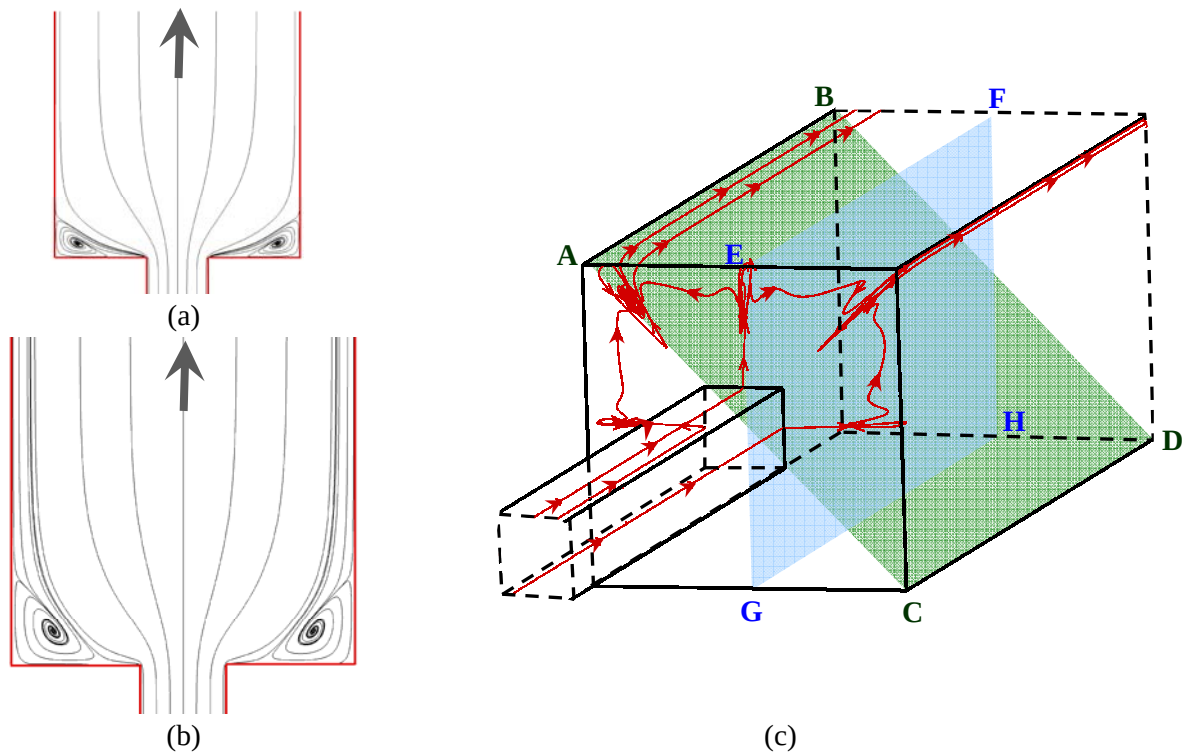


Figure 5.9. Streamlines predicted numerically for the Newtonian fluid flow through the 1:4 square-square expansion under creeping flow conditions. (a) Streamlines at the center plane (EFGH); (b) streamlines at the diagonal plane (ABCD); (c) three-dimensional view of some streamlines. Note that in the center planes (a) the fluid enters the recirculation while in the diagonal planes (b) the fluid exits the recirculation.

Interestingly, when inertial effects are important, besides the increase of the recirculation documented previously (cf. Figure 5.4 and 5.5) we observe a reversal in the flow direction within the recirculation. This can be observed in the streamlines plotted in Figure 5.10 at $Re = 10$ and $CR = 4$. In this case, the recirculating flow occurs from the diagonal planes, where the fluid is sucked in (cf. Figure 5.10(b)), to the center planes where the fluid is ejected (cf. Figure 5.10(a)). This flow reversal was previously documented in square-square contractions, but due to elastic effects (Alves *et al.*, 2005; Alves *et al.*, 2008; Sousa *et al.*, 2009 (cf. Chapter 3)). The present results suggest that flow reversal in square-square contractions or expansions is not a fingerprint of elastic effects, but seems to occur concomitantly with vortex enhancement.

In Figure 5.11 we show streak line projections of the Boger and shear-thinning fluid flows at different parallel planes of the 1:2.4 square-square expansion and identical Deborah numbers, illustrating the complex and highly three-dimensional flow behavior particularly near the walls.

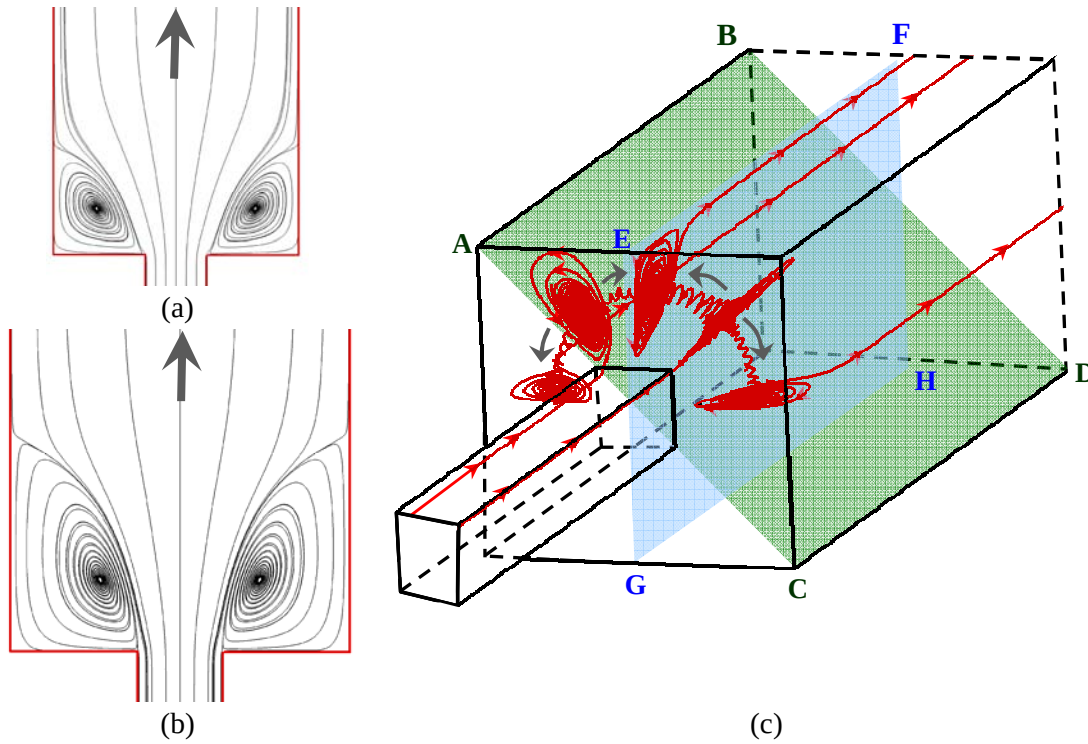


Figure 5.10. Streamlines predicted numerically for the Newtonian fluid flow through the 1:4 square-square expansion at $Re = 10$. (a) Streamlines at the center plane (EFGH); (b) streamlines at the diagonal plane (ABCD); (c) three-dimensional view of some streamlines. Note that in the diagonal planes (b) the fluid enters the recirculation while in the central planes (a) the fluid exits the recirculation.

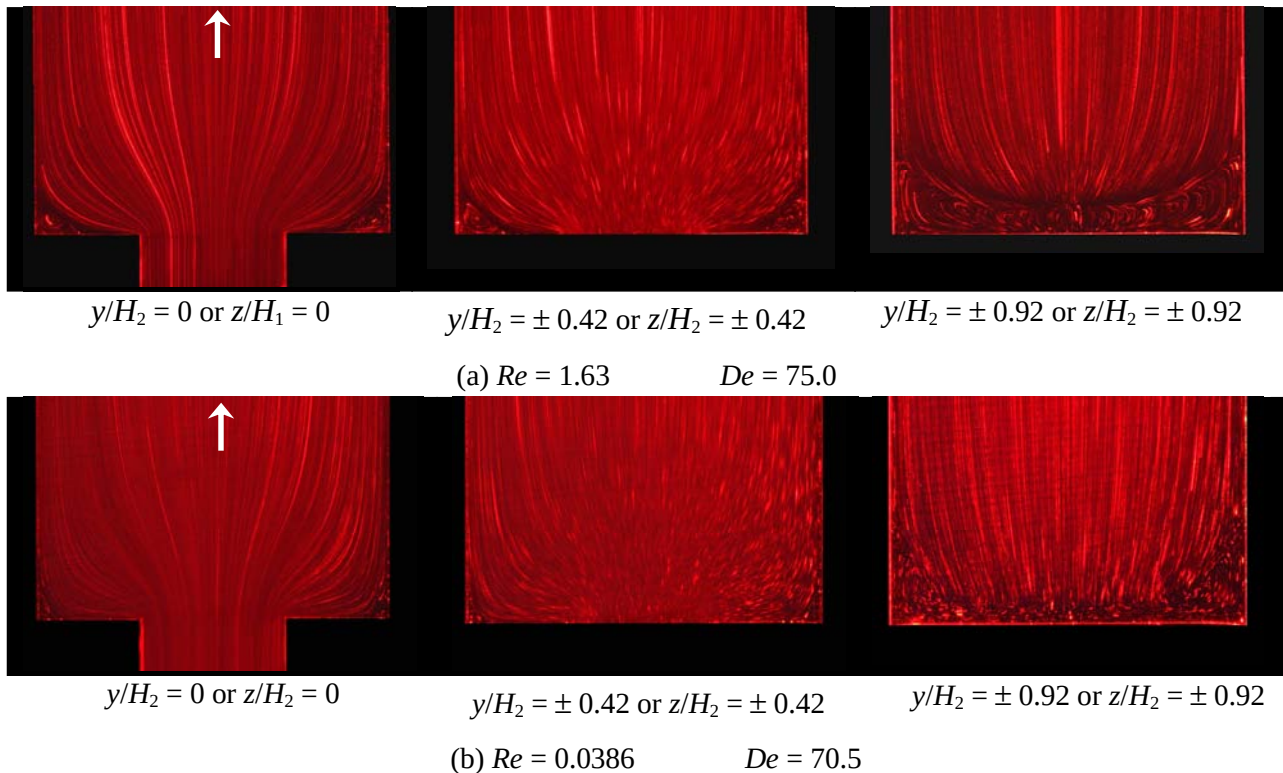


Figure 5.11. Projections of path lines at different parallel planes for the Boger (a) and shear-thinning (b) fluid flow through the square-square expansion ($ER = 2.4$).

5.6. Velocity field

5.6.1. Newtonian fluid

In order to highlight the inertial effects on the velocity field, in Figure 5.12 we show axial velocity profiles along the centerline for two expansion ratios ($ER = 2.4$ and $ER = 8$), at different Reynolds numbers. The axial velocity profile predicted numerically for negligible inertial flow conditions (creeping flow) is also shown for comparison purposes. Moreover, we compare the experimental results with numerical predictions for each value of Re .

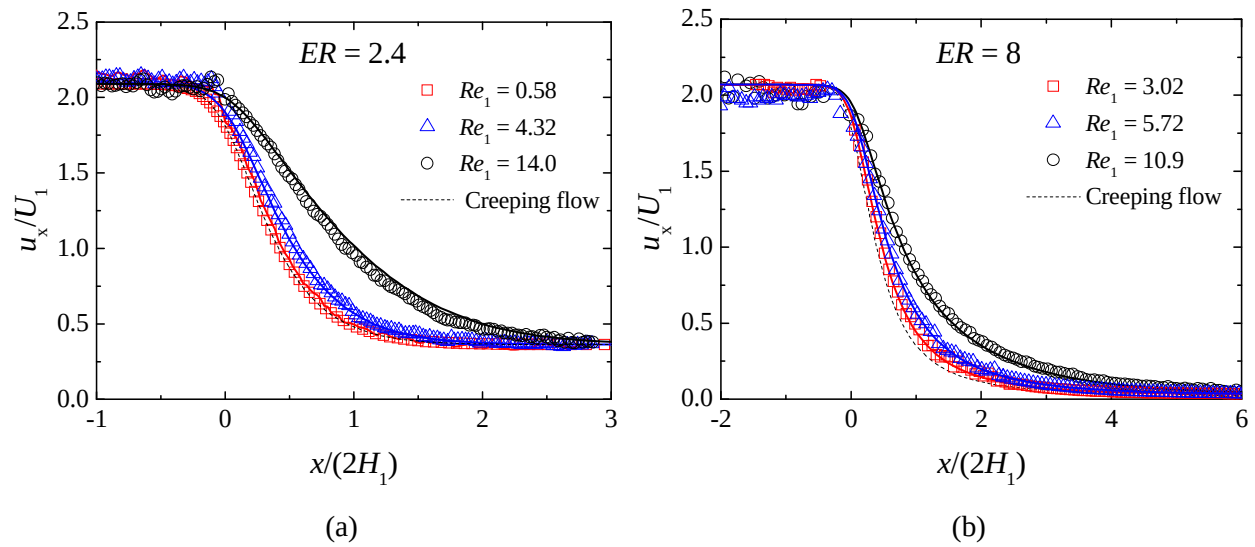


Figure 5.12. Experimental (symbols) and numerical (lines) axial velocity profiles at the centerline for the Newtonian fluid flow at different Re : (a) $ER = 2.4$; (b) $ER = 8$.

In all cases, the dimensionless velocity is plotted from locations upstream ($x < 0$) to downstream ($x > 0$) of the expansion plane. Due to the symmetry of the geometry, the velocity profiles in the x - y plane are identical to those in the x - z plane. As can be seen, the dimensionless velocity gradient downstream of the expansion plane decreases when the Reynolds number increases. As inertia increases, entrance effects also become more pronounced and the size of the recirculations, formed downstream of the expansion plane, also increases. For $ER = 2.4$, the normalized velocity profile measured experimentally at low Re is similar to that obtained numerically for creeping flow. For all axial velocity profiles shown in Figure 5.12, the experimental results are in good agreement with those predicted numerically, for both $ER = 2.4$ and 8, thus validating the PIV measurements.

A comparison between experimental and numerical axial velocity profiles along the spanwise direction is presented in Figure 5.13. The velocity profiles were taken at fixed x -positions from a location upstream ($x = -2H_1$) to downstream ($x = 12H_1$) of the expansion plane as shown in the inset scheme. The velocity profile at position $x = 3.25H_1$ was selected because it crosses the center of the recirculation. When the Newtonian fluid flows through the expansion, the fluid decelerates abruptly and for the positions at which the velocity profiles presented in Figure 5.13 were taken, the fluid experiences a decrease on the velocity of more than forty times (note that comparing the velocity for fully-developed conditions of the flow at positions well upstream and downstream of the expansion, the total decrease on the velocity is sixty four times).

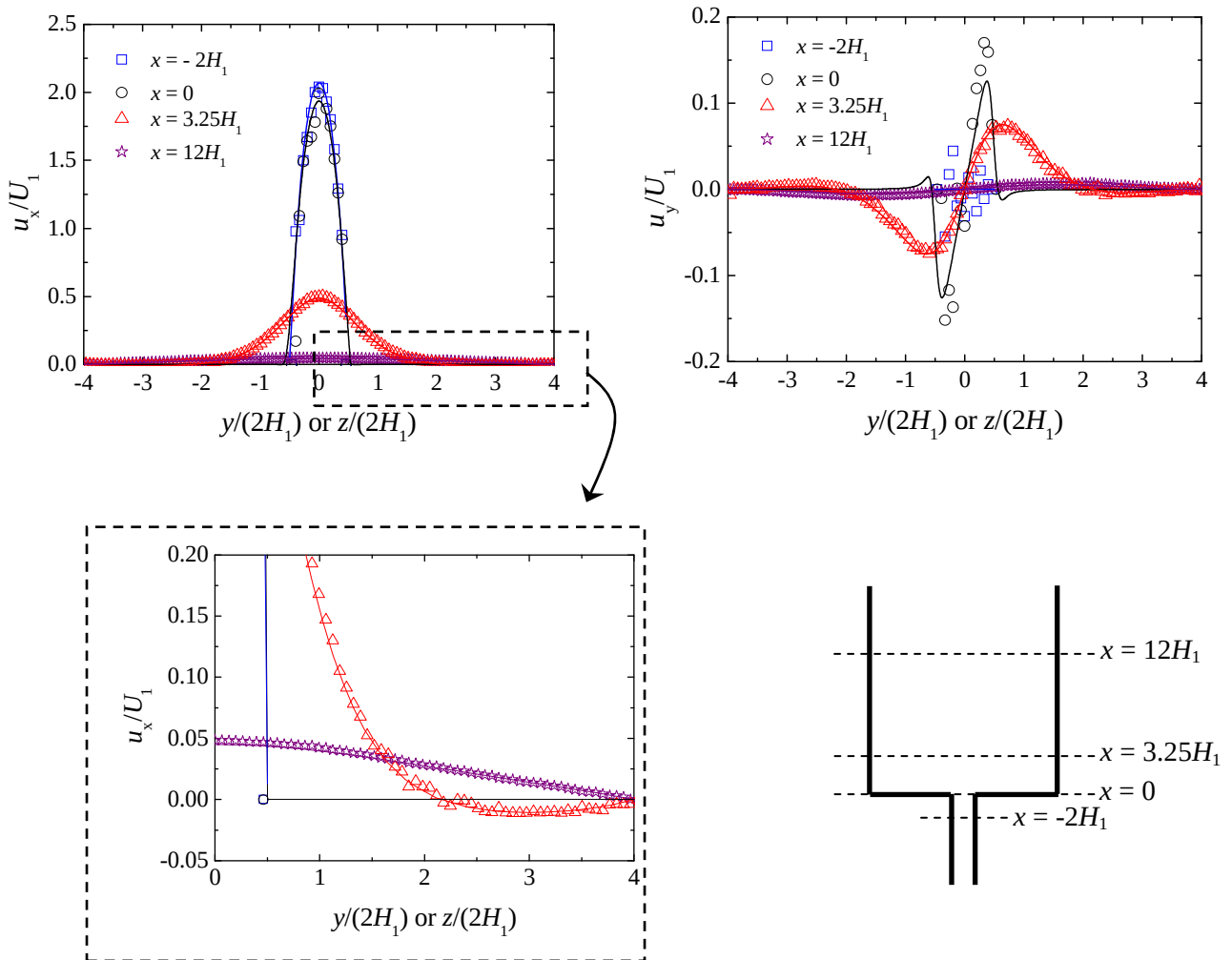


Figure 5.13. Dimensionless velocity profiles taken along the span wise direction of the square channel for the Newtonian fluid at $Re = 0.9$ and $ER = 8$. The numerical predictions are shown as lines.

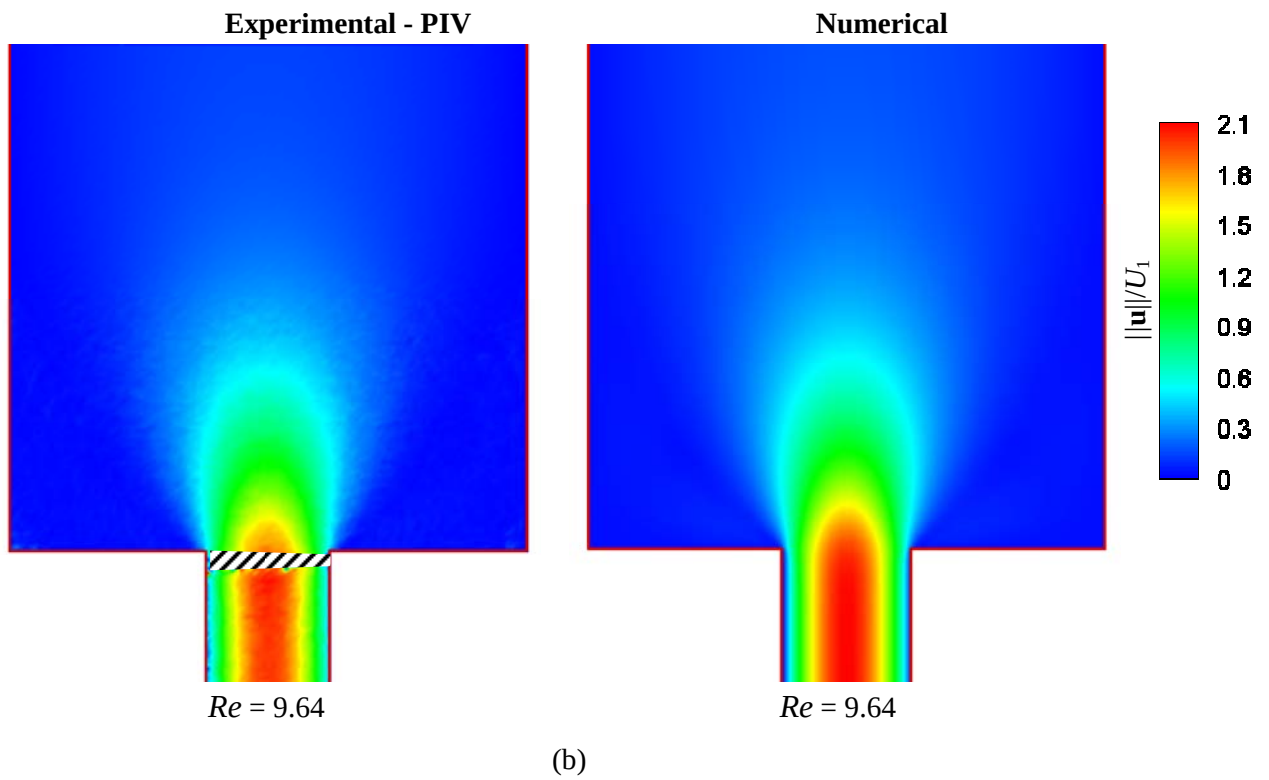
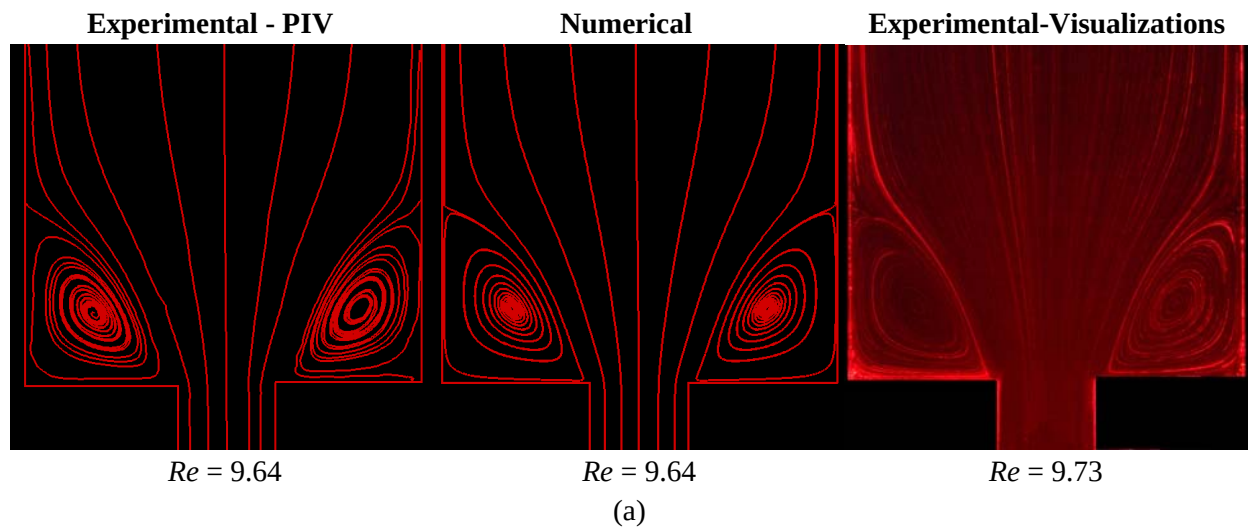


Figure 5.14. Comparison between experimental and numerical results: (a) flow patterns (obtained experimentally from integration of the velocity field measured with PIV; obtained from numerical calculations; obtained using long time exposure streak line photography); (b) normalized velocity magnitude contour plots.

Figure 5.13 demonstrates that the experimental results have a good accuracy and that we are able to measure the low values of the velocity in the recirculation region (cf. profile taken at location $x = 3.25H_1$ for $2 \leq |y/(2H_1)| \leq 4$ and $2 \leq |z/(2H_1)| \leq 4$ in the inset). In general, good agreement between experimental and numerical results is observed. However, for profiles taken near the expansion region ($x/(2H_1) \approx 0$), there is a deviation in the experimental spanwise component of the velocity (u_y) from the numerical predictions which is due to the reduced visibility of tracer particles in that region. Consequently, the precision of the PIV results near this region of the flow is reduced.

To further attest the good agreement between experimental and numerical results, in Figure 5.14(a) we present a comparison between the middle plane pathlines obtained using three different approaches: flow visualizations using streak line photography; integration of the measured velocity field using PIV; numerical predictions. The example shown corresponds to Newtonian fluid flow through the 1:4 square-square expansion at similar Reynolds numbers ($Re \approx 10$). In Figure 5.14(b) we compare the normalized axial velocity contour plots obtained from the PIV measurements and numerical simulations. Again, the experimental results are in excellent agreement with the numerical predictions.

5.6.2. Viscoelastic fluid

Figure 5.15 shows normalized axial velocity profiles taken along the centerline for the Boger fluid flowing through the 1:4 and 1:12 square expansions (Figure 5.15(a)) and for the shear-thinning fluid flowing through square-square expansions with expansion ratios of 2.4 and 8 (Figure 5.15(b)). Predictions of velocity profiles for a Newtonian fluid flowing at creeping flow conditions are also shown in Figure 5.15 to highlight the strong influence of elasticity on the dimensionless velocity profiles.

Once again, the axial velocity profiles are plotted from positions far upstream to far downstream of the expansion plane ($-2 \lesssim x/(2H_1) \lesssim 5$). For a square channel, the theoretical maximum velocity or centerline velocity is 2.096 times the average velocity for a constant viscosity fluid (either Newtonian or a Boger fluid) (Alves *et al.*, 2008). Therefore, for the Boger fluid, the axial velocity far upstream of the expansion should be $u_x/U_1 = 2.096$ as observed for the creeping flow velocity profile computed. In the limit $De \rightarrow 0$, i.e. for very low flow rates, the axial velocity is similar to that of a Newtonian fluid under creeping flow conditions. An increase

in the flow rate (or De) leads to the appearance of a local maximum of the axial velocity near the expansion region.

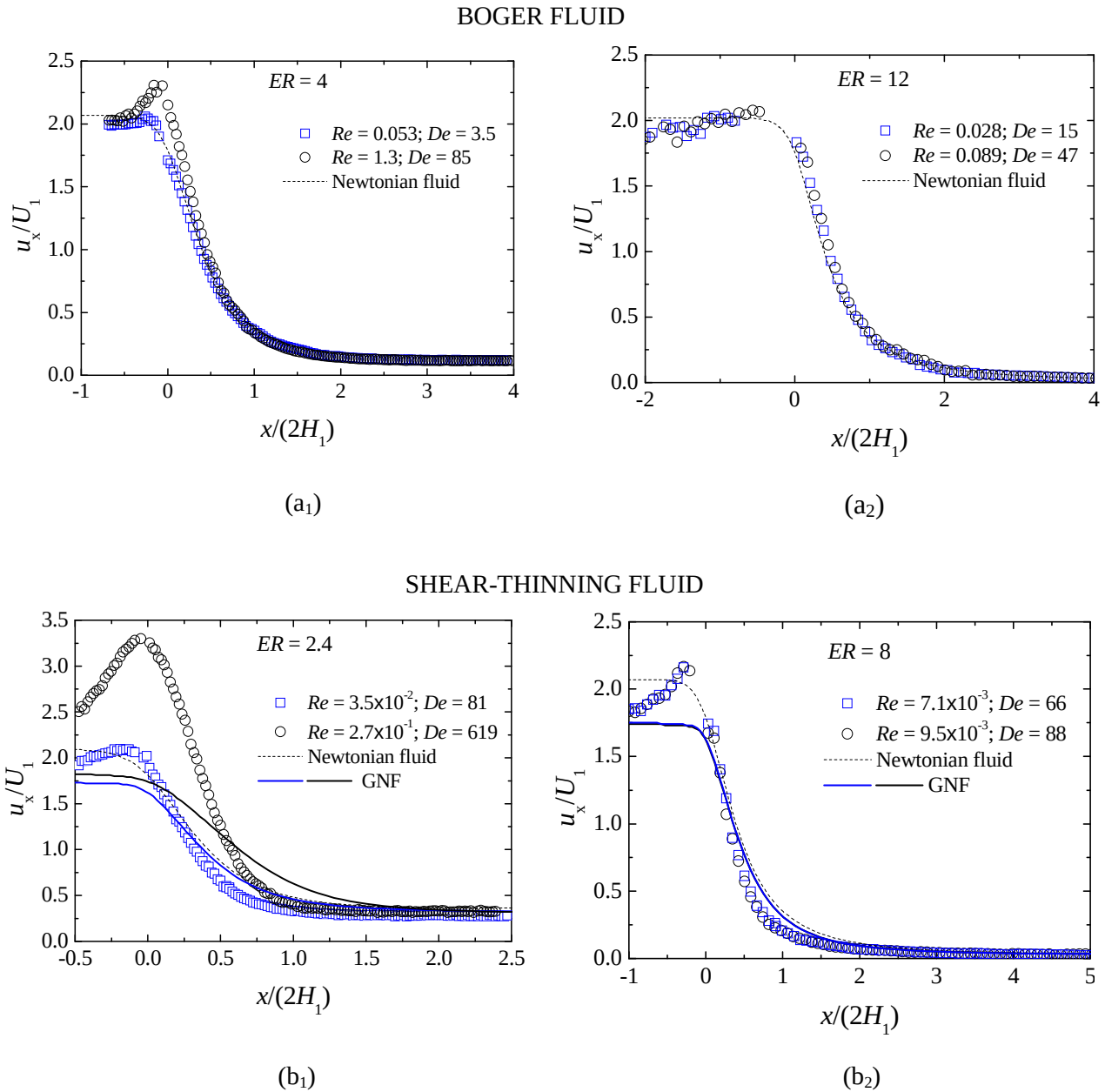


Figure 5.15. Dimensionless axial velocity profiles at the centerline of the square-square expansion for the Boger ((a₁) and (a₂)) the shear-thinning fluids ((b₁) and (b₂)) at different expansion ratios. The symbols represent the experimental results, the dashed lines represent the numerical predictions for the Newtonian fluid under creeping flow conditions and the solid lines represent the numerical predictions for a generalized Newtonian fluid with rheological properties similar to those of the viscoelastic fluids used in this work, flowing at the same flow rate as in the experiments.

For the Boger and the shear-thinning fluids studied, at high De , a significant overshoot on the axial velocity along the centerline is clearly visible with the maximum axial velocity reaching

values well above the fully-developed value, which means that the fluid experiences an acceleration as it is approaching the expansion plane (converging streamlines) leading to a higher rate of decay of the velocity when it enters the channel with larger cross-section. The same phenomenon was observed experimentally by Rothstein and McKinley (2001) for Boger fluid flow through an axisymmetric contraction-expansion and numerically by Oliveira (2003) for a FENE-CR fluid and by Poole *et al.* (2007) for UCM and PTT fluids flowing through planar expansions, although to a smaller extent. Eventually, the centerline velocity downstream of the expansion reaches the theoretical value corresponding to fully-developed flow conditions. However, as the flow rate is increased, fully-developed flow is only achieved progressively farther away from the expansion plane and the overshoot on the velocity exhibits a higher magnitude. On the other hand, for the shear-thinning fluid, since the viscosity depends strongly on the shear rate, the ratio of the maximum velocity achieved in the upstream channel to the average velocity, depends on the flow rate. Thus, in order to attain the value of the axial velocity for fully-developed conditions, which was not measurable experimentally, we performed numerical simulations using a generalized Newtonian fluid (GNF) with a rheological behavior in steady shear flow similar to that found for the shear-thinning viscoelastic fluid used. For this, the experimental rheological data (cf. Section 5.3) was fitted using a Carreau-Yasuda model (Bird *et al.*, 1987),

$$\eta = \eta_s + \frac{\eta_0 - \eta_s}{\left[1 + (\Lambda \dot{\gamma})^a\right]^{(1-n)/a}}, \quad (5.8)$$

where the parameters of the fitted model are: $\eta_0 = 1.65$ Pa s; $\eta_s = 0.03$ Pa s; $\Lambda = 10$ s; $n = 0.36$ and $a = 0.9$. The numerical predictions using this rheological model are also shown in Figure 5.15(b) for the shear-thinning fluid. As can be seen, the fully developed normalized axial velocity depends on the flow rate and its value is smaller than that for a constant viscosity fluid. Moreover it is possible to further attest that the overshoot present on the axial velocity profile is a consequence of the elastic effects, since for a GNF no overshoot is observed.

In Figure 5.16 we present the profiles of the x and y velocity components taken in the spanwise direction at different x -locations. In addition, we also compare the upstream axial velocity profiles with a fully-developed velocity profile obtained for a Newtonian fluid in Figure 5.16(a1) and with the GNF fluid in Figure 5.16(b1). It is also clear that the flow remains symmetric for both the Boger and the shear-thinning fluid.

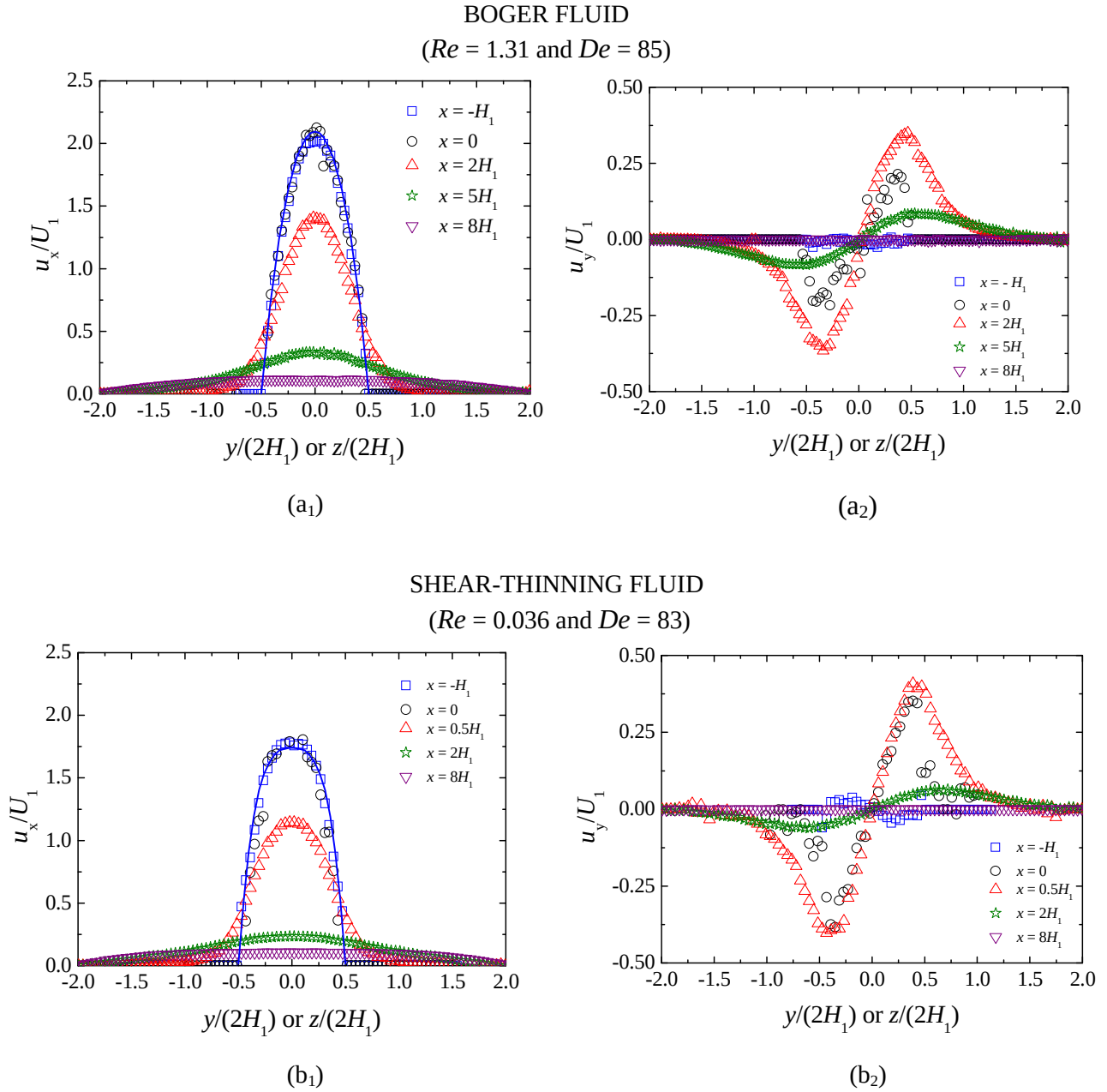


Figure 5.16. Dimensionless velocity profiles taken along the span wise direction of the 1:4 square-square expansion for the Boger (a₁ and a₂) and the shear-thinning fluids (b₁ and b₂). The symbols represent the experimental data and the solid lines represent the theoretical predictions for a Newtonian fluid (a₁) and a GNF fluid (b₁) under fully-developed flow conditions.

From the velocity profiles taken at discrete x -positions it is possible to infer about the evolution of the velocity along the channel. For the Boger fluid (cf. Figure 5.16(a)), the velocity profile at location $x = -H_1$ shows that the fluid is under fully-developed conditions, since the transverse profile of the axial velocity shows good agreement with the theoretical profile for a Newtonian fluid under fully developed flow conditions (which is coincident with the profile of any constant viscosity fluid, such as a Boger fluid). In the channel with a larger cross-section, the

x -component of the velocity decreases progressively while the y -component experiences an increase near its entrance and up to $x = 2H_1$, gradually decreasing for locations farther downstream. For the shear-thinning fluid, the flow behavior is similar to that observed for the Boger fluid, except for the shape of the velocity profile. In this case, the profile becomes more like a plug flow profile, especially upstream of the expansion, a typical behavior of shear-thinning fluids under fully-developed flow conditions. Good agreement with the fully developed flow velocity profile predicted with the GNF fluid is observed for the shear-thinning fluid, as shown in Figure 5.16(b1) at the upstream location ($x = -H_1$).

5.7. Pressure drop

Pressure drop measurements for viscoelastic fluid flows through expansions are scarce, particularly if the flow occurs in channels with a 3D geometrical arrangement. The results obtained in this study for the Boger fluid flow through square-square expansions with different ER can be useful as benchmark data for validation of numerical results.

Figure 5.17 shows the pressure drop measured with the Boger fluid (cf. Section 5.2.2 for pressure ports locations) as a function of the flow rate for $ER = 4, 8$ and 12 . Since the pressure drop increases linearly with the flow rate for all ER studied, we also present a linear fit to the experimental results.

A linear increase of the pressure drop with the flow rate indicates that elastic effects are not significant in the flow rate range measured. Nevertheless, in order to further investigate the presence of elastic effects in the experimental data, numerical simulations were performed considering two different cases: (i) a Newtonian fluid with a shear viscosity equal to the total shear viscosity of the Oldroyd-B model used to fit the rheology of the Boger fluid ($\eta_0 = 0.646$ Pa s); (ii) a Newtonian fluid with a shear viscosity equal to the solvent contribution of the shear viscosity of the Oldroyd-B model ($\eta_s = 0.367$ Pa s). Note that an ideal Boger fluid has a shear viscosity independent of the shear rate but, in practice, there is always a slight decrease of the viscosity with the shear rate. In Figure 5.17 we show the numerical predictions of the pressure drop across the expansion plane for these Newtonian fluids to compare with the experimental data obtained with the Boger fluid used in the experiments. As can be seen, the relation between experimental and numerical results is analogous for all expansion ratios studied and the experimental data lies in-between the numerical predictions of the Newtonian fluids. From these results we can conclude that there is no significant enhancement of pressure drop due to elastic effects, in contrast with the results obtained with the same fluid in 3D square-square

contractions where a significant increase of the entry pressure drop due to elastic effects was found for high contraction ratios (Sousa *et al.*, 2009) (cf. Chapter 3).

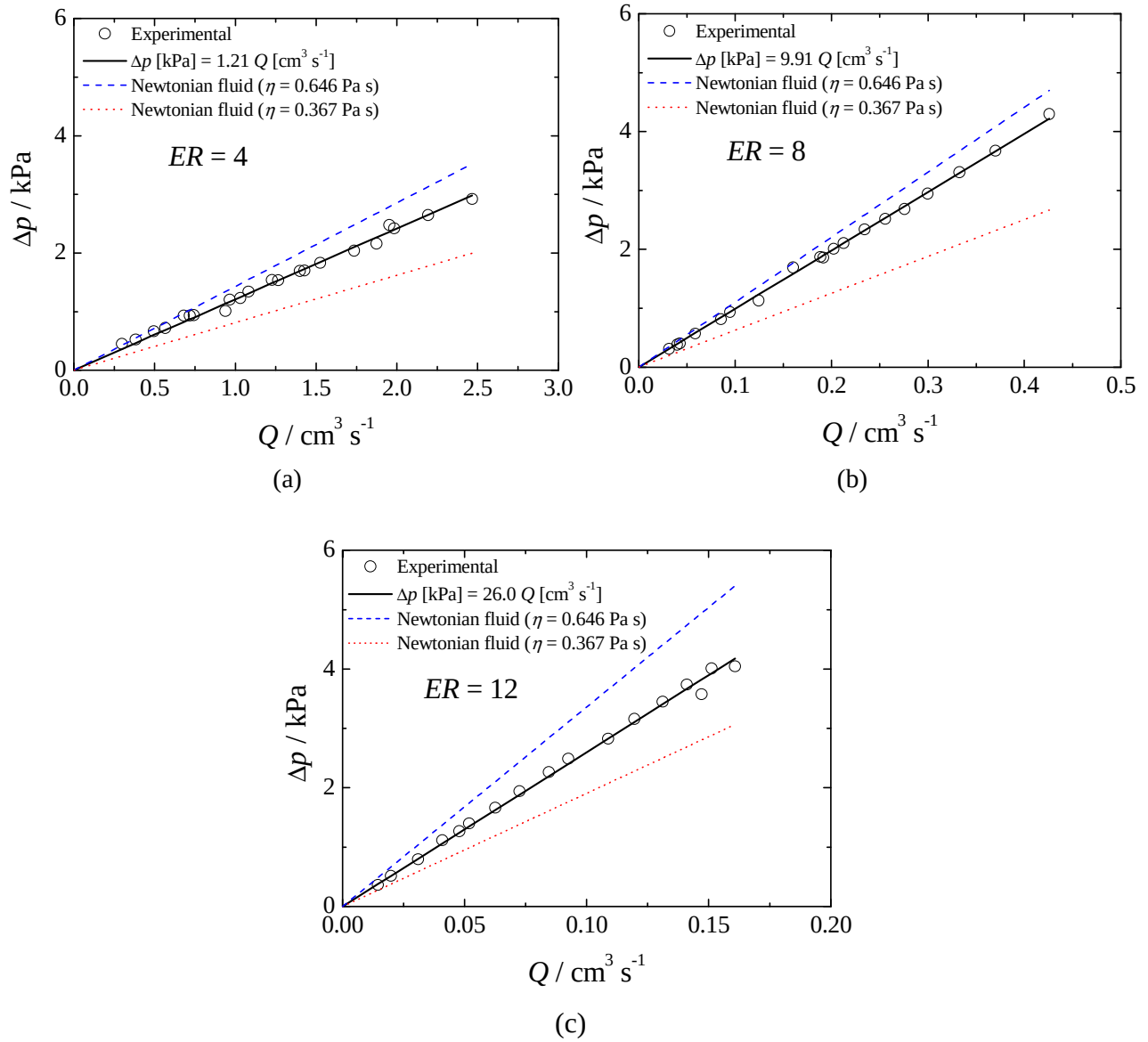


Figure 5.17. Pressure drop as a function of the flow rate for $ER = 4$ (a), $ER = 8$ (b) and $ER = 12$ (c). The symbols represent the experimental data, the thick solid lines the corresponding fit and the dashed lines represent the numerical predictions for Newtonian fluids.

5.8. Conclusions

The three-dimensional flow of a Newtonian and two viscoelastic fluids through square/square expansions with expansion ratios of 2.4, 4, 8 and 12 was investigated experimentally and numerically. In addition to the characterization of the flow through square-square expansions, this work also intends to provide useful data for benchmarking in a complex 3D flow.

Three-dimensional numerical simulations of the Newtonian and non-Newtonian fluid flow were performed using a finite volume method. The Newtonian fluid flow presents a Moffatt corner vortex downstream of the expansion plane and the effect of inertia on the flow behavior is similar for all expansion ratios studied: increasing the Reynolds number leads to an increase of the vortex length and intensity and a reversal in the flow direction inside the recirculation. The viscoelastic fluid flow behavior is also analogous for all expansion ratios studied: a corner vortex is observed downstream of the expansion plane and increasing the Deborah number leads, in general, to a decrease of the corner vortex length, which is more marked for the shear-thinning fluid. A complex helicoidal flow within the vortical structure is observed experimentally for all fluids studied and confirmed by the numerical simulations. The numerical results capture very well the flow characteristics obtained experimentally for the whole range of conditions for Newtonian the fluid. For the viscoelastic fluid flow the numerical predictions also predict a decrease of vortex activity at low Deborah number flows, followed by a vortex enhancement at higher De , a phenomenon not observed in the experiments. For the Boger fluid, the pressure drop across the square-square expansion increases linearly with the flow rate and does not reveal an enhancement of the extra pressure drop due to elasticity.

Acknowledgements

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PART II

Entry flow of viscoelastic fluids at micro-scale

EFFICIENT MICROFLUIDIC RECTIFIERS FOR VISCOELASTIC FLUID FLOW

Abstract

In this work we propose a new type of microfluidic rectifier, which is able to operate efficiently under creeping flow conditions. The flow of Newtonian and non-Newtonian fluids was investigated experimentally in different microchannels with triangular (nozzle/diffuser) and hyperbolic shapes in order to achieve high anisotropic flow resistance between the two flow directions. The Newtonian fluid used was de-ionized water and the viscoelastic fluids were aqueous solutions of polyacrylamide and polyethylene oxide with different molecular weights. Pressure drop measurements were performed in addition to visualizations of the flow patterns by streak line photography for a wide range of flow rates. For the Newtonian flows, inertia leads to the appearance of recirculations for both flow directions, but no significant rectification effects appear. For the viscoelastic fluids, two distinct behaviors are identified: at low flow rates, the pressure drops are similar in both flow directions; above a critical flow rate (or Deborah number), the flow patterns become quite different, leading to different flow rates in the forward and backward flow directions for the same pressure drop, i.e., rectification effects emerge. In particular, the viscoelastic fluid flow becomes unsteady in the forward direction, due to the presence of elastic instabilities, which leads to a significant increase in the flow resistance. Flow resistance ratios greater than three were achieved for the hyperbolic rectifier, clearly in excess of the value for the triangular-shaped rectifier and for other geometries proposed in the literature for operation in creeping flow conditions. This high diodicity is associated with the distinct nature of the extensional flows in the forward and backward directions of the hyperbolic-type micro-geometry.

6.1. Introduction

Today, the lab-on-a-chip concept has widespread application in science and technology, as in biology, medicine and engineering. Miniaturized systems for dispensing therapeutic agents, analysis of drugs or DNA molecules, microchemical reactors or inkjet printing heads are just a few examples of microfluidics applications. These devices handle small-sized samples, making portability a relevant characteristic of these miniaturized laboratories. In micro total analysis systems (μ TAS), small and precise fluid volumes with well defined fluid properties and flow characteristics (i.e. viscosity, density, pH, temperature, etc) must be pumped, transported, controlled or handled using external devices such as pumps, valves or other actuators.

Positive displacement pumps are the primary selection in microfluidics given their typical low Reynolds numbers flows. These devices require valves at the inlet and outlet and also between chambers in multi-chamber pumps, which poses a complex challenge in the miniaturization of pumping systems (Forster *et al.*, 1995) and an increase in their effective cost and risk of failure (Laser and Santiago, 2004). These valves may also have detrimental effect upon cells, molecules and other entities sensitive to damage, which in some cases must be avoided. Therefore, and as an alternative to the use of active or passive valves, microchannels with a fixed geometry and direction-dependent flow resistance are used. These so-called flow rectifiers are the fluid mechanics equivalent of electronic diodes; pumps equipped with such devices are frequently encountered in microfluidics, where they are called valveless micropumps also known as no-moving parts (NMP) or fixed geometry micropumps and have received broad attention and various optimized geometries have been developed and tested when operating with Newtonian fluids (Nabavi, 2009). Note also that in some micropumps the actuator is placed in the middle of the flow rectifier.

One of the earliest reciprocating displacement micropumps was developed by Jan Smits in the 1980s for controlled delivery of insulin to diabetics, as an alternative to classical delivery systems (Smits, 1990). Since then, several micropump designs have been developed and investigated (e.g. Laser and Santiago, 2004; Koch *et al.*, 1997; Unger *et al.*, 2000; Jeong and Yang, 2000; Cui *et al.*, 2008) and the vast majority do contain some inlet/outlet valves or NMP flow rectifiers.

The first flow rectifier, named “Valvular conduit”, was proposed and patented by Nikola Tesla (Tesla, 1920) and consisted of an open main duct connected to a series of side loop

channels at sharp angles. Its rectification effects were found for Newtonian fluids at moderate Reynolds numbers (Re), when nonlinearities due to inertial effects became important. Stemme and Stemme (1993) proposed a nozzle/diffuser micropump with a fixed geometry, which consists of a channel with opening angles that work as flow-directing elements. When Newtonian fluids flow in the diverging wall direction (diffuser element), enhanced flow-directing properties are achieved and the pressure drop becomes dependent on the flow rate. This nozzle/diffuser geometry became a very popular flow rectifier design. In order to optimize the corresponding valveless micropumps, Olsson *et al.* (1995) investigated, from a theoretical and experimental point of view, the planar nozzle/diffuser structure of a double chamber brass based micropump. Other contributions to the optimization of the nozzle/diffuser performance have been carried out numerically and experimentally over the last decade (e.g. Cui *et al.*, 2008; Olsson *et al.*, 1996; Jiang *et al.*, 1998; Pan *et al.*, 2003; Yang *et al.*, 2004; Singhal *et al.*, 2004; Oh and Ahn, 2006; Sun and Han, 2007; Tanaka *et al.*, 2008; Hwang *et al.*, 2008; Wang *et al.*, 2009; Nabavi and Mongeau, 2009). Gerlach and Wurmus (1995) studied a piezoelectrically driven micropump in which a nozzle/diffuser valve was incorporated and concluded that the device could be used as a mixer/reactor chamber, in addition to operating as a micropump, due to the turbulence that was generated during its operation. Forster *et al.* (1995) tested and compared the efficiency of different NMP valves, a simple diffuser and a valvular conduit (Tesla-type). In the end, the Tesla-type valve was found to have the highest difference between forward and backward flow resistance, an important flow property called diodicity.

Three-dimensional (3D) nozzle/diffuser microvalves etched in silicon were investigated by Heschel *et al.* (1997). Devices with different dimensions and shapes were tested and it was found that the flow behavior within 3D nozzle/diffuser microstructures depends only weakly on the shape of the geometry, with higher improvements in efficiency brought by rounding the throat. Olsson *et al.* (2000) reported a numerical and experimental investigation of nozzle/diffuser elements for valveless micropumps. The sharpness of the throat corner edges was explored in their study and the flow-directing capability of the geometry was demonstrated. At low Reynolds numbers, the 3D simulations predicted well the difference between the pressure drops for each flow direction, whereas at high Reynolds numbers good results could only be achieved using a two-dimensional numerical code. Nozzle/diffuser geometries were also used in valveless micropumps embedded as asymmetric obstacles within a piezo-electrical micropump (Sheen *et al.*, 2008; Lee *et al.*, 2009). The Tesla-type valves were used in several studies as a NMP microdevice or coupled in micropumps, similar to the device used by Forster *et al.* (1995). More

recently, the geometry of these valves has been optimized for high Reynolds number Newtonian flow (Bardell *et al.*, 1997; Feldt and Chew, 2002; Gamboa *et al.*, 2005).

Micropumps, valves and flow rectifiers can be fabricated in diverse materials using straightforward and inexpensive methods (Yamahata *et al.*, 2005). Photocurable polymers, such as hydrogels that can be photopolymerized *in situ*, were explored (Carrozza *et al.*, 1995; Liu *et al.*, 2002). Loverich *et al.* (2006) reported a polymer-based micropump fabricated with transparent (for optical access), fluid-compatible and economical materials. This micropump integrated an elastomeric actuator and flap check valves, made up of PDMS (polydimethylsiloxane), that conditioned the flow behavior generating a dependency between the pressure drop across the microvalve and the flow rate. More recently, Loverich *et al.* (2007) reported an improved flap check-valve for operation at low Reynolds number flow having a maximum pressure drop ratio of 4.6 between the two flow directions and for the same flow rate. In medical or biological μ TAS chips, it is very important to preserve the characteristics of the fluid that is being pumped (e.g. biological fluids) and passive or active check-valves frequently are not the best choices. Instead, NMP valves have the lowest detrimental effect upon the biological entities in the fluid. A valveless micropump for pumping fluids, containing beads and living cells, without damage was developed by Andersson *et al.* (2001).

All investigations discussed so far refer to Newtonian fluid flow, usually using water. For Newtonian fluids the rectification effect of NMP devices is a consequence of the flow inertia. However, microfluidic flows typically occur under low Reynolds number flow conditions and as such, the flow resistance is only weakly dependent, or even independent, of the flow direction due to the reversibility of creeping flows of Newtonian fluids (i.e. rectification effects are small). Nevertheless, anisotropic flow resistance can be achieved under creeping flow conditions providing other nonlinear mechanisms come to play a role. One possible alternative relies on adding small amounts of additives that confer viscoelastic rheological properties to the fluid (Groisman and Quake, 2004). Since viscoelastic nonlinearities can be enhanced at the microscale (Squires and Quake, 2005), rectification effects can be achieved even with dilute polymeric fluids. The elasticity number, which represents the relative importance of elastic to inertial effects and is given by the product of relaxation time and kinematic viscosity divided by the length scale squared, increases as the size of the geometry is reduced, thus demonstrating the importance of elastic effects in microfluidics as compared to the *macroscale*. A pioneering investigation on viscoelastic flows at the microscale was undertaken by Groisman *et al.* (2003).

The flow of dilute aqueous polyacrylamide (PAA) solutions in a complex nonlinear resistor was studied, demonstrating the potential for use as a flow stabilizer (the flow rate across the microfluidic device is approximately constant for a large range of applied pressure drops), due to the elastic nature of the working fluid.

Somewhat surprisingly, studies that describe microfluidic rectifiers with non-Newtonian fluid flows are scarce. Groisman and Quake (2004) investigated the flow of a 0.01 wt. % aqueous solution of a high molecular weight PAA, through a microchannel with a nozzle/diffuser structure composed of a consecutive number of similar triangles (cf. Figure 6.1(b)). Anisotropic flow resistance was observed and a maximum flow rate ratio of almost 2 was achieved for the same pressure gradient applied in both flow directions. Nguyen *et al.* (2008) also investigated the rectification effects at low Re in a similar nozzle/diffuser structure using an aqueous solution of polyethylene oxide (PEO) at 0.1 wt. %. They studied the effect of the opening angle of the triangular structures on the flow resistance anisotropy and found a maximum rectification effect characterized by a flow rate ratio of about 1.8.

A list summarizing relevant investigations on flows of Newtonian and non-Newtonian fluids in systems including valveless micropumps or flow rectifiers is presented in Table 6.1, where the above referenced works are also listed. This table includes information on the type of investigation, materials used, flow geometry, maximum pressure drop across the device, Reynolds number range and the achieved diodicity, in addition to some relevant comments.

It is clear that there is much to be explored in this area of flow rectifiers for creeping flows operating with polymer additives and this paper is one such contribution. Here, we investigate the performance of microfluidic no-moving-parts rectifiers having a hyperbolic-like shape, which are able to operate efficiently under creeping flow conditions using dilute viscoelastic polymeric solutions. The main objective is to design an efficient microgeometry with high rectification effects. Using Newtonian and non-Newtonian fluids, the performance of the optimized geometry was compared with that of the nozzle/diffuser structure of Groisman and Quake (2004) and Nguyen *et al.* (2008) by means of flow rate and pressure drop measurements, in addition to visualizations of the flow patterns. The microfluidic rectifiers were fabricated in PDMS using a low cost soft lithography technique and are composed of a succession of contractions/expansions with different hyperbolic and triangular shapes.

Table 6.1. Summary of previous studies regarding no-moving parts micropumps.

Ref.	Valve Material / Type	Application	Exp/Num	Fluid	$\Delta P_{\max}$ kPa	Diodicity/ efficiency	$Re_{\max}$	Notes
Forster <i>et al.</i> , 1995	Silicon-glass Diffuser/Nozzle Tesla-type	Valv. micr. (PZT actuator)	Exp/Num	Newtonian (water)	30.4	1.2	n.r.	2-D and 3-D simulations; Best diodicity for diffuser valve.
Jeong and Yang, 2000	Silicon-glass Diffuser/Nozzle	Valv. micr. (thermopneumatic actuator)	Exp	Newtonian (water)	2.5	n.a.	n.r.	The maximum flow rate of the micropump is 0.014 ml/min.
Cui <i>et al.</i> , 2008	n.a. Diffuser/Nozzle	Valv. micr. (PZT actuator)	Num	Newtonian (Water)	n.r.	n.r.	n.r.	Theoretical analysis and numerical simulations; Maximum pump flow rate of 0.08 ml/min.
Stemme and Stemme, 1993	Brass Diffuser/Nozzle	Valv. micr.	Exp	Newtonian (water)	19.6	2	10^3	Maximum pump flow rate of 16 ml/min.
Olsson <i>et al.</i> , 1995	Brass Diffuser/Nozzle	Valv. micr. (PZT actuator)	Exp	Newtonian (Water)	16	2	n.r.	Double chamber pump; Comparison between experimental and theoretical results; Maximum net flow rate of 16ml/min.
Olsson <i>et al.</i> , 1996	Silicon-glass Diffuser/Nozzle	Valv. micr.	Exp	Newtonian (Methanol/water)	100	1.45	~ 200	Different open angles and different dimensions were studied; Maximum efficiency for methanol; Experimental and analytical results are compared.
Jiang <i>et al.</i> , 1998	Silicon Diffuser/Nozzle	Valv. micr.	Exp/num	Newtonian (Water)	1	1.5	10^5	Different open angles and conical structures were studied; Maximum net flow rate of 0.028 ml/min.

Table 6.1. Summary of previous studies regarding no-moving parts micropumps (continued).

Ref.	Valve Material / Type	Application	Exp/Num	Fluid	$\Delta P_{\max}$ kPa	Diodicity/ efficiency	$Re_{\max}$	Notes
Pan et al., 2003	n.a. Diffuser/Nozzle	Valv. micr.	Analytical	Newtonian	n.a.	n.a.	n.a.	Derivation of a model for the fluid flow; Dimensionless analysis.
Yang et al., 2004	Silicon Diffuser/Nozzle	Valve	Exp	Newtonian (Water)	n.r.	1.725	400	Different opening angles and depths were studied; Max. diodicity for an open angle of 30°; Fixed flow rate in all experiments.
Sighal et al., 2004	n.a. Diffuser/Nozzle	Valv. micr.	Num	Newtonian (water/methaol)	n.r.	1.43	103	Planar, pyramidal and conical geometries studied.
Sun and Han, 2007	Silicon Diffuser/Nozzle	Valve	Exp	Newtonian (Water)	0.6	1.5	22	Best efficiency obtained at an open angle of 30°.
Tanaka et al., 2008	Stainless steel Diffuser/Nozzle	Valv. micr. (PZT actuator)	Exp	Newtonian (Water/ water+glycerol)	1.4	n.r.	103	Different opening angles were studied.
Hwang et al., 2008	Epoxy-glass Diffuser/Nozzle	Valv. micr. (PZT actuator)	Exp/num	Newtonian (Water)	n.r.	n.r.	n.r.	Comparison between experimental and theoretical results.
Wang et al., 2009	Alluminium Diffuser/Nozzle	Valv. micr.	Exp/num	Newtonian (Water)	n.r.	2	2×103	Studied different opening angles (best efficiency at 40°).
Nabavi and Mongeau, 2009	n.a. Diffuser/Nozzle	Valv. micr. acoustic	Num	Newtonian (Water)	10	~1.3	51	Pulsating flow; Studied different angles; Maximum rectification effect at 60°.
Gerlach and Wurmus, 1995	Silicon-glass Diffuser/Nozzle	Valv. micr. (PZT actuator)	Exp	Newtonian (water/methanol)	7	n.r.	104	Maximum pressure drop for methanol.

Table 6.1. Summary of previous studies regarding no-moving parts micropumps (continued).

Ref.	Valve Material / Type	Application	Exp/Num	Fluid	$\Delta P_{\max}$ kPa	Diodicity/ efficiency	$Re_{\max}$	Notes
Heschel et al., 1997	Silicon Diffuser/Nozzle	Valve	Exp	Newtonian (Water)	80	Diffuser/ Nozzle 29%	n.r.	3D structures; Different open angles and different dimensions were studied.
Olsson et al., 2000	Micromachined Diffuser/Nozzle	Valv. micr.	Exp/num	Newtonian (water)	~200	1.59 (Exp) 3.20 (2D Num)	1350 (Num) 650 (Exp)	Structures with round inlets and sharp outlets; 2-D and 3-D simulations; Experimental study of various geometrical dimensions.
Lee et al., 2009	Silicon Diffuser/Nozzle	Valv. micr. (PZT actuator)	Exp	Newtonian (Water)	1.2	n.r.	n.r.	Diffuser/nozzle structures used as obstacles in a micropump.
Feldt and Chew, 2002	n.a. Tesla	Valv. micr.	Analytical	Newtonian	n.a.	n.a.	n.a.	Geometry-based model for analysis of no-moving-parts microvalves; Experimental data reported in the literature is used.
Gamboa et al., 2005	Acrylic Tesla	Valv. micr.	Exp/num	Newtonian	20	1.5	2×103	Shape was optimized using CFD tools; Experiments carried out using the optimized geometry.
Yamahata et al., 2005	PMMA Diffuser/Nozzle	Valv. micr. (electromagnetic actuator)	Exp/num	Newtonian (water/air)	2	2	27	Studied sharp and round corners; Pump flow rate of 0.4 ml/min and 12 mbar.
Loverich et al., 2006	PDMS Flap	Valv. micr.	Exp	Newtonian (Water)	8	3.9	n.r.	Flap valve that works as a rectifier.
Loverich et al., 2007	PDMS Flap	Valve	Exp.	Newtonian (Water)	14	4.6	30	Different valve heights were studied.

Table 6.1. Summary of previous studies regarding no-moving parts micropumps (continued).

Ref.	Valve Material / Type	Application	Exp/Num	Fluid	$\Delta P_{\max}$ kPa	Diodicity/ efficiency	$Re_{\max}$	Notes
Andersson et al., 2001	Silicon-glass Diffuser/Nozzle	Valv. micr. (PZT actuator)	Exp	Newtonian fluids used in biochemistry	n.r.	n.r.	n.r.	Tested different fluids that cannot be pumped in other micropumps, such as high ionic character fluids and cell suspensions; Maximum flow rate of 0.014 ml/min.
Groisman and Quake, 2004	PDMS Diffuser/Nozzle	Valve	Exp	Viscoelastic (PAA)	11	2	1.4	Rectifier with 43 similar elements in series.
Nguyen et al., 2008	Silicon-glass Diffuser/Nozzle	Valve	Exp	Viscoelastic (PEO)	100	1.8	20	Rectifier with 10 similar elements; Different dimensions and angles of the walls were studied.

Valv. micr. – valveless micropump; PZT – piezoelectric; CFD – computational fluid dynamics; Exp. – Experimental; Num. – Numerical; n.a. – not applicable; n.r. – not reported; PDMS – polydimethylsiloxane; PEO – polyethylene oxide; PMMA – polymethylmethacrylate.

The remainder of this paper is organized as follows: Section 0 describes the flow geometries, their fabrication and the experimental techniques used. In Section 6.3 the working fluids and their rheology are presented, followed by a discussion of the dimensionless groups that characterize the flow to set the stage for the experiments, the results of which are presented and discussed in Section 6.4. The paper closes with conclusions in Section 6.5.

6.2. Experimental Techniques

6.2.1. Microchannels geometry, fabrication and experimental set-up

The microchannels used in the experiments were fabricated in PDMS using standard soft lithography techniques (McDonal *et al.*, 2000) and SU-8 photo-resist molds. PDMS elastomer has been widely used for the fabrication of microfluidic devices because of its characteristics such as transparency, mechanical behavior, biocompatibility, rapid prototyping and low cost. In this work, two different types of planar microgeometries were used: one composed of triangular elements, based on the nozzle/diffuser structure reported in the literature (Olsson *et al.*, 2000; Groisman and Quake, 2004; Nguyen *et al.*, 2008) and another type of microchannels with hyperbolic shaped elements. Figure 6.1 illustrates the layout of two typical microchannels used in this work.

All microgeometries used consist of a sequence of 42 similar elements aligned in series. The hyperbolic shape was selected in order to achieve fluid flows that are primarily extensional, with a nearly constant strain rate along the centerline, as discussed by Oliveira *et al.* (2007). We define the forward direction when the fluid flows along smooth contractions and abrupt expansions (left to right in Figure 6.1) and backward direction as the opposite one. For the forward flow the fluid is stretched at a nearly constant extensional rate along the centerline, resulting in a high flow resistance for fluids that have large extensional viscosities, such as solutions of high molecular weight polymers, whereas in the backward flow direction the fluid can relax also at a nearly constant extensional rate. The devices used in the present investigation are composed of two inlets/outlets located at the extremes of the microchannels, followed by pressure taps on each side of the test section, which is positioned at the central part of the microchannel, as shown in Figure 6.1(c). The pressure taps allow for the measurement of the pressure drop (ΔP) across the 42 repeating elements of the test section.

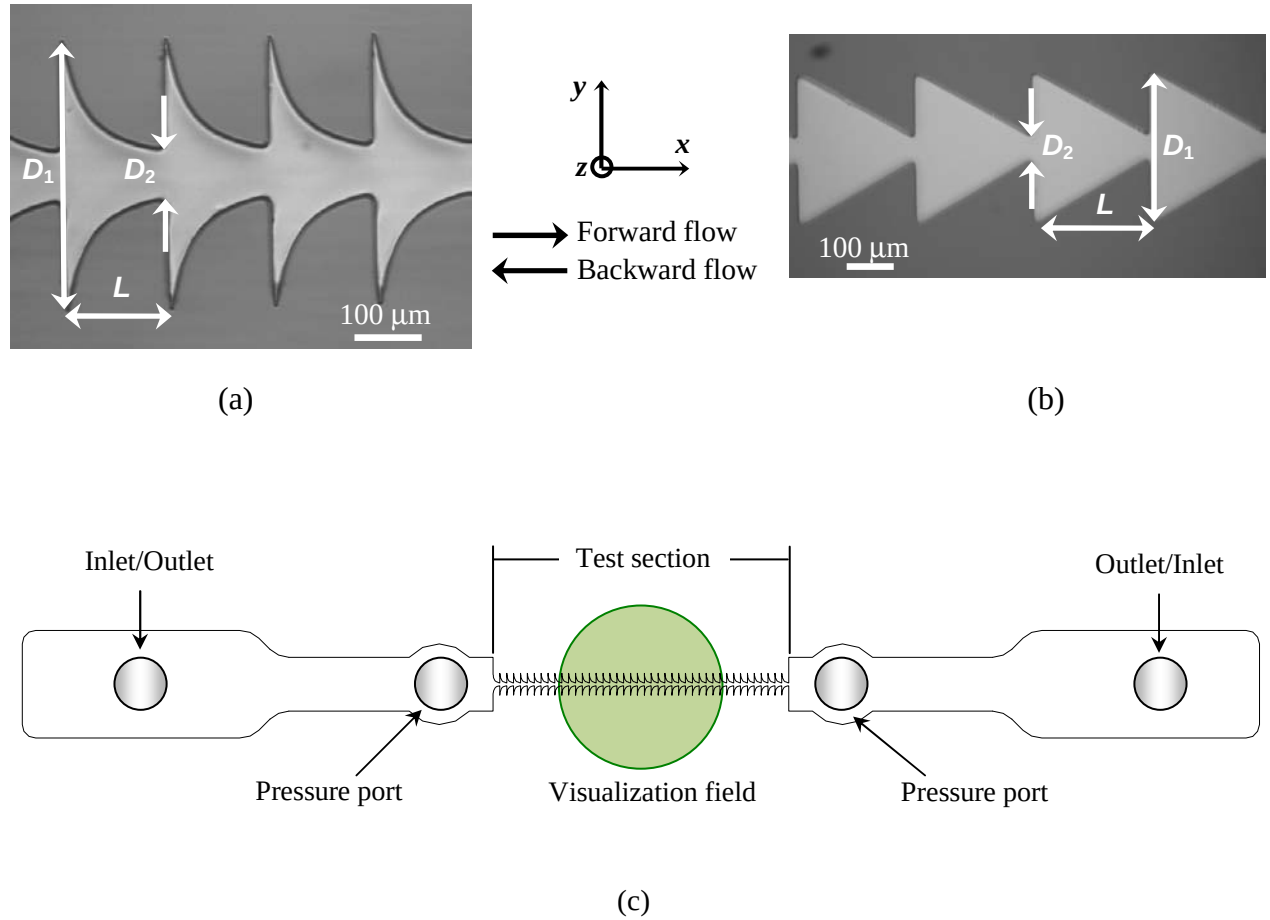


Figure 6.1. Micrograph of the (a) hyperbolic and (b) triangular shaped microfluidic rectifiers. (c) Top view of the microchannel used in the experiments.

Table 6.2 summarizes the dimensions of the different microgeometries used in the experiments, as well as the values of the Hencky strain (total deformation) experienced by the fluid, here defined as $\varepsilon_H = \ln(D_1/D_2)$. The parameters of the hyperbolic function that describes the shape of the microchannel elements, $y = \pm a/(x + b)$, where $0 \leq x \leq L$, are also listed in Table 6.2. At the widest and narrowest parts, the widths of the microgeometries are D_1 and D_2 , respectively, and the length of each element is L , as sketched in Figure 6.1. The depth of the channels is uniform and was kept constant for all micro-devices studied, $h = 50 \mu\text{m}$.

In order to make a direct comparison between the results obtained with the hyperbolic rectifier proposed in this work and the triangular rectifier geometry investigated by Groisman and Quake (2004) it is necessary to use microgeometries with similar aspect ratios. For that purpose the triangular rectifier and the C_2 hyperbolic rectifier used in this work have similar dimensions.

The flow behavior of Newtonian and non-Newtonian fluids was studied at room temperature in the forward and backward directions (cf. Figure 6.1) for a wide range of flow rates, which

were imposed using a syringe pump (PHD2000, Harvard Apparatus). Syringes (Hamilton) with volumes ranging from 50 μl to 10 ml were used according to the desired flow rate and connected to the microchannels via Tygon tubing of 0.44 mm internal diameter. The outlet of the microchannels was connected to Tygon tubing that directs the fluid to a reservoir open to the atmosphere where the fluid is collected.

Table 6.2. Geometrical characteristics of the microchannels studied.

Channel	D_1 [μm]	D_2 [μm]	L [μm]	Hencky strain, ε_H	a [μm ²]	b [μm]	
Triangular, T	330	37	230	2.19	-	-	
Hyperbolic	C_1	400	54	128	2.00	4000	20
	C_2	326	37	230	2.18	4800	29
	C_3	390	54	382	1.98	11972	61
	C_4	400	20	382	3.00	4000	20

6.2.2. Flow visualization

Visualizations of the flow patterns relied on streak line photography. For this purpose, the fluids were seeded with 1 μm fluorescent tracer particles (Nile Red, Molecular Probes, Invitrogen, Ex/Em: 520/580 nm) and sodium dodecyl sulfate (0.1 wt. %, Sigma-Aldrich) was added to the solutions in order to reduce the adhesion of fluorescent particles to the channel walls. Rheological measurements confirmed that the rheology of the fluid was not changed by using such low concentrations of this surfactant agent. The optical setup is shown schematically in Figure 6.2. It consists of an inverted epi-fluorescence microscope (Leica Microsystems GmbH, DMI 5000M) equipped with a CCD camera (Leica Microsystems GmbH, DFC350 FX), a filter cube (Leica Microsystems GmbH, excitation BP 530-545 nm, dichroic 565 nm, barrier filter 610-675 nm) and a 100 W mercury lamp light source.

The microgeometries were continuously illuminated and path line images were acquired using a 10 $\times$ (NA = 0.25) microscope objective (Leica Microsystems GmbH) and *long* exposures times (~ 1 s) in order to obtain a visual fingerprint of the flow patterns in the focused center-plane.

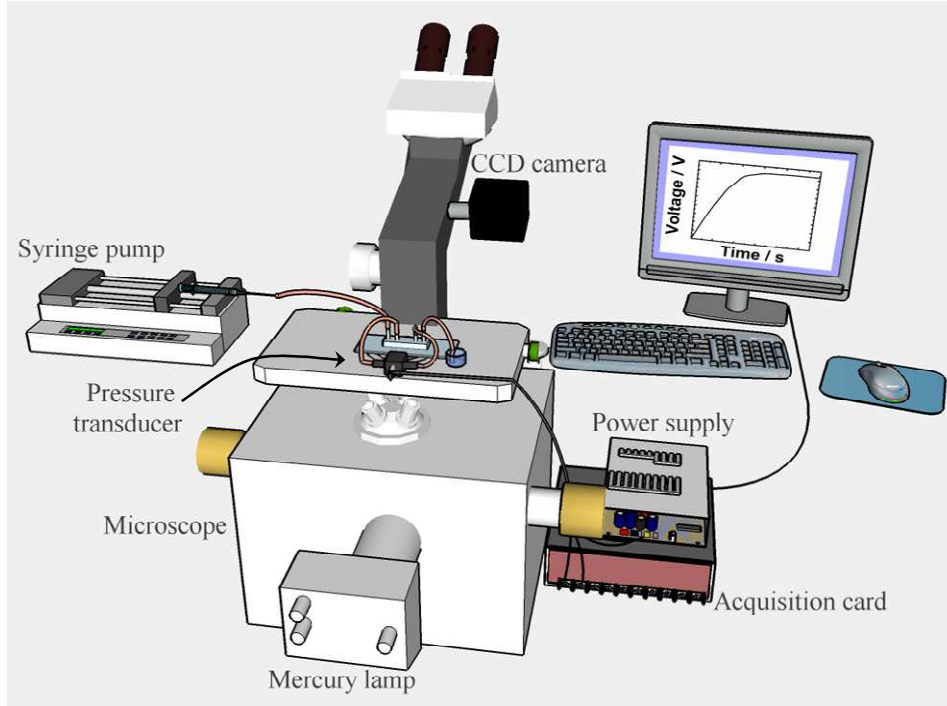


Figure 6.2. Schematic view of the experimental set-up.

The depth of field (DOF) for an optical system can be calculated as (Meinhart *et al.*, 2000):

$$\delta z = \frac{n\lambda_0}{(\text{NA})^2} + \frac{ne}{(\text{NA})M}, \quad (6.1)$$

where n is the refractive index, λ_0 is the wavelength of the light (in vacuum), NA is the numerical aperture of the objective, e is the minimum detectable size and M is the total magnification. For our optical set-up, $\delta z = 12 \mu\text{m}$ and $e/M = 0.65 \mu\text{m}$ which is a value smaller than the particle diameter, d_p .

6.2.3. Pressure drop measurements

Pressure drop measurements were performed using Honeywell differential pressure sensors (model 26PC series) covering values up to $\Delta P = 200 \text{ kPa}$. The pressure sensors were calibrated using a static column of water for pressures up to $\Delta P = 34 \text{ kPa}$ and using a compressed air line and a manometer (Wika Instrument Corporation, model 332.50) with an accuracy of $\pm 2 \text{ kPa}$ for sensors that are able to measure higher differential pressures, up to 200 kPa .

The ports of the pressure transducer were connected to two pressure taps, located upstream and downstream of the test section (cf. Figure 6.1(c)), respectively. A 12 V DC power supply

(Lascar electronics, PSU 206) was used to power the pressure sensors that were also connected to a computer via a data acquisition card (NI USB-6218, National Instruments) in order to record the output data using LabView v7.1. The transient response of the pressure sensors was continuously recorded until steady-state was reached.

6.3. Fluids and nondimensional numbers

6.3.1. Fluid composition and rheology

A Newtonian fluid (de-ionized water) and various viscoelastic fluids with different rheological behavior were used in the experiments. The polymers used to prepare the viscoelastic fluids were polyacrylamide with a molecular weight $M_w = 18 \times 10^6 \text{ g mol}^{-1}$ (Polysciences) and polyethylene oxide (PEO) with two different molecular weights: $M_w = 2 \times 10^6 \text{ g mol}^{-1}$ (PEO-2M) and $M_w = 8 \times 10^6 \text{ g mol}^{-1}$ (PEO-8M), both supplied by Sigma-Aldrich. Solutions were prepared by mixing the polymer into the solvent (de-ionized water, de-ionized water/glycerol or de-ionized water/sucrose solutions) at different concentrations, utilizing magnetic stirrers at low speeds, in order to avoid mechanical degradation of the polymer molecules. To prevent chemical degradation, all solutions were kept in a refrigerator and additionally PEO solutions were also protected from light using dark bottles (Hassouna *et al.*, 2007). The density of all solutions prepared was measured at 293.2 K using calibrated 5 cm³ density flasks.

The composition of all fluids by weight, fluid density (ρ) and polymer molecular weight are given in Table 6.3.

Table 6.3. Properties of the fluids used.

Fluid	M_w [g mol ⁻¹]	Polymer [ppm]	Water [%]	Sucrose [%]	NaCl [%]	Glycerol [%]	ρ [kg m ⁻³]
Newtonian	-	-	100	-	-	-	998
PEO-2M	2×10^6	1000	99.90	-	-	-	998
PEO-8M	8×10^6	1000	99.90	-	-	-	998
PEO-8M+Glyc.	8×10^6	1000	59.90	-	-	40.00	1099
PEO-8M 100	8×10^6	100	99.99	-	-	-	999
PAA	18×10^6	100	85.91	13.06	1.02	-	1059

The rheology of the fluids was measured in both extensional and shear flows. The extensional flow characterization used a capillary break-up extensional rheometer (Haake CaBER 1, Thermo

Scientific) and the characteristic relaxation time of each fluid was measured. These measurements were performed using circular plates with a diameter $D_p = 6$ mm. The initial separation gap between the two plates was set to h_i , which corresponds to an aspect ratio, $A_i = h_i/D_p$. The liquid bridge confined between the two plates is stretched as the top plate moves linearly ($-50 \text{ ms} \leq t \leq 0$) to a final height (h_f), which leads to a final aspect ratio, $A_f = h_f/D_p$. In the ensuing liquid filament thinning the relaxation time, λ , was determined by fitting the experimental data of $\log[D(t)]$ vs time in the linear region to the equation resulting from the elasto-capillary balance (Entov and Hinch, 1997): $D(t)/D_0 \propto \exp[-t/(3\lambda)]$, where D_0 is the radius of the filament at time $t = 0$. In Table 6.4, we present the relaxation times obtained as well as the initial and final aspect ratios used for each viscoelastic fluid tested.

Table 6.4. Initial and final aspect ratios used in CaBER experiments and measured relaxation times ($T = 293.2$ K).

Fluid	A_i	A_f	λ /ms
PEO-2M	0.33	1.34	8.2±0.8
PEO-8M	0.50	1.39	73.9±1.0
PEO-8M+Glyc.	0.50	1.63	17.9±0.6
PEO-8M 100	0.33	1.19	2.5±0.2
PAA	0.50	1.50	13.2±0.2

The critical overlap concentration, c^* , or the concentration at which adjacent polymer coils overlap, is calculated according to Graessley (1980) as $c^* = 0.77/[\eta]$, where $[\eta]$ is the intrinsic viscosity.

The intrinsic viscosity for the PEO solutions can be estimated using the Mark-Houwink-Sakurada (MHS) equation, $[\eta] = 0.072 M_w^{0.65}$ with $[\eta]$ expressed in cm^3/g (Tirtaatmadja *et al.*, 2006). The PEO-2M solution has an intrinsic viscosity, $[\eta] = 897 \text{ cm}^3/\text{g}$, while for the other PEO solutions with a molecular weight of $8 \times 10^6 \text{ g mol}^{-1}$ the intrinsic viscosity is $[\eta] = 2210 \text{ cm}^3 \text{ g}^{-1}$. The intrinsic viscosity of the PAA solution was determined experimentally using a Cannon-Fenske viscosimeter (Coneccta SA, model 1464, size 50) and various dilute solutions with the same solvent and different polymer concentrations. The intrinsic viscosity was determined by means of an extrapolating method, namely using the Huggins equation, $(\eta - \eta_s)/(\eta_s c) = [\eta] + K_H [\eta]^2 c$, where η is the solution viscosity, η_s is the solvent viscosity, c is the concentration and K_H is the Huggins constant that measures the interaction

between polymer and solvent (Brandrup *et al.*, 1999). The fit to the experimental data yields $[\eta] = 1504 \text{ cm}^3/\text{g}$ and $K_H = 8.7^\dagger$.

For a polymer chain in a good solvent, the longest relaxation time can be calculated according to the Zimm theory (Doi and Edwards, 1986) as:

$$\lambda_{\text{Zimm}} = \frac{1}{\zeta(3\nu)} \frac{[\eta] M_w \eta_s}{N_A k_B T}, \quad (6.2)$$

where N_A is Avogadro's number, k_B the Boltzmann constant, T the absolute temperature and $\zeta(3\nu) = \sum_{i=1}^{\infty} (1/i^{3\nu})$ represents the sum of individual modal contributions to the relaxation time. For the PEO solutions in water or in water/glycerol, the solvent quality exponent, ν , and the prefactor, $1/\zeta(3\nu)$ are 0.55 and 0.46 respectively, as reported elsewhere (Entov and Hinch, 1997). On the other hand, to the best of our knowledge, the solvent quality exponent reported in the literature for PAA solutions considers only water as the solvent, rather than water/sucrose. However, using these values we can obtain a crude estimate of the longest relaxation time (λ_{Zimm}) for comparative purposes. As such, considering water as the main solvent component, the exponent varies between $\nu = 0.61$ (for $4 \times 10^4 \leq M_w / \text{g mol}^{-1} \leq 1.47 \times 10^6$) and $\nu = 0.57$ (for $4.9 \times 10^5 \leq M_w / \text{g mol}^{-1} \leq 3.2 \times 10^6$ and a NaCl concentration of 1N) (Brandrup *et al.*, 1999). Hence, the prefactor $1/\zeta(3\nu)$ becomes 0.54 for $\nu = 0.61$, and 0.49 for $\nu = 0.57$. Consequently, the Zimm relaxation time varies in the range between 9.8 and 10.8 ms, which is similar to the value found from the CaBER experiments for the same fluid. For quantitative purposes, in the calculation of the dimensionless numbers, we will use in all cases the CaBER relaxation time.

Table 6.5 provides the coil overlap concentration (c^*), the dimensionless concentration (c/c^*) and the Zimm relaxation time (λ_{Zimm}) obtained at the reference temperature (293.2 K), for each fluid used in the experiments.

The PEO-8M 100 and PAA solutions are diluted and therefore the Zimm relaxation time is in reasonable agreement with the measured values. All the other viscoelastic fluids used in this work are semi-dilute solutions. For the PEO-2M and PEO-8M fluids in particular, λ_{Zimm} is smaller than the relaxation time found under capillary break-up extensional measurements which is probably due to molecular chain interactions during the CaBER experiments as described in Rodd *et al.* (2005) and Soulages *et al.* (2009).

[†] Appendix D

Table 6.5. Coil overlap concentration, dimensionless concentration and Zimm relaxation times for each fluid used in the experiments.

Fluid	$c^* / \text{g cm}^{-3}$	c/c^*	$\lambda_{\text{Zimm}} / \text{ms}$
PEO-2M	8.58×10^{-4}	1.16	0.63
PEO-8M	3.48×10^{-4}	2.86	10.1
PEO-8M+Glyc.	3.48×10^{-4}	3.15	21.8
PEO-8M 100	3.48×10^{-4}	0.29	3.36
PAA	5.12×10^{-4}	0.21	10.3 ± 0.5

For the shear flow rheological characterization, a shear rheometer (Anton Paar, model Physica MCR301) was used with a cone-plate geometry (75 mm diameter and 1° angle) operating in the shear rate range of $1 \leq \dot{\gamma} / \text{s}^{-1} \leq 7 \times 10^3$. For all polymer solutions, the shear viscosity (η) was measured at different temperatures ranging from 283.2 K to 303.2 K and the time-temperature superposition principle was used in order to obtain a master curve. The corresponding shift factor is defined as (Bird *et al.*, 1987):

$$a_T = \frac{\eta(T)}{\eta(T_0)} \frac{T_0}{T} \frac{\rho_0}{\rho}, \quad (6.3)$$

where $\eta(T)$ is the shear viscosity at a given absolute temperature T , $\eta(T_0)$ is the shear viscosity at the reference absolute temperature T_0 (293.2 K), whereas ρ_0 and ρ are the fluid densities at the reference temperature and at temperature T , respectively. For the range of temperatures used in the measurements, the variation of the fluid density is negligibly small, hence the shift factor can be calculated through the following expression (Bird *et al.*, 1987):

$$a_T = \frac{\eta(T)}{\eta(T_0)}. \quad (6.4)$$

The dependency of the shift factor on the temperature can be described by an Arrhenius equation:

$$\ln(a_T) = \frac{\Delta H}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right), \quad (6.5)$$

where ΔH represents the activation energy for flow and R the universal gas constant.

In order to determine the master curve, the shear viscosity and the shear rate need to be calculated in the reduced form or transformed to the reference temperature according to:

$$\eta_r = \eta(T_0) = \eta(T) / a_T, \quad (6.6)$$

$$\gamma_r = \dot{\gamma}(T_0) = a_T \dot{\gamma}. \quad (6.7)$$

The master curves for steady shear flow are shown in Figure 6.3(a) for the PEO solutions and in Figure 6.3(b) for the PAA solution. In addition, the fit of the simplified Phan-Thien-Tanner (sPTT) model (Phan-Thien and Tanner, 1977), with a solvent contribution, which predicts a shear-thinning behavior is also shown. More details about this viscoelastic model can be found in (Soulages *et al.*, 2009) or on the original reference (Phan-Thien and Tanner, 1977).

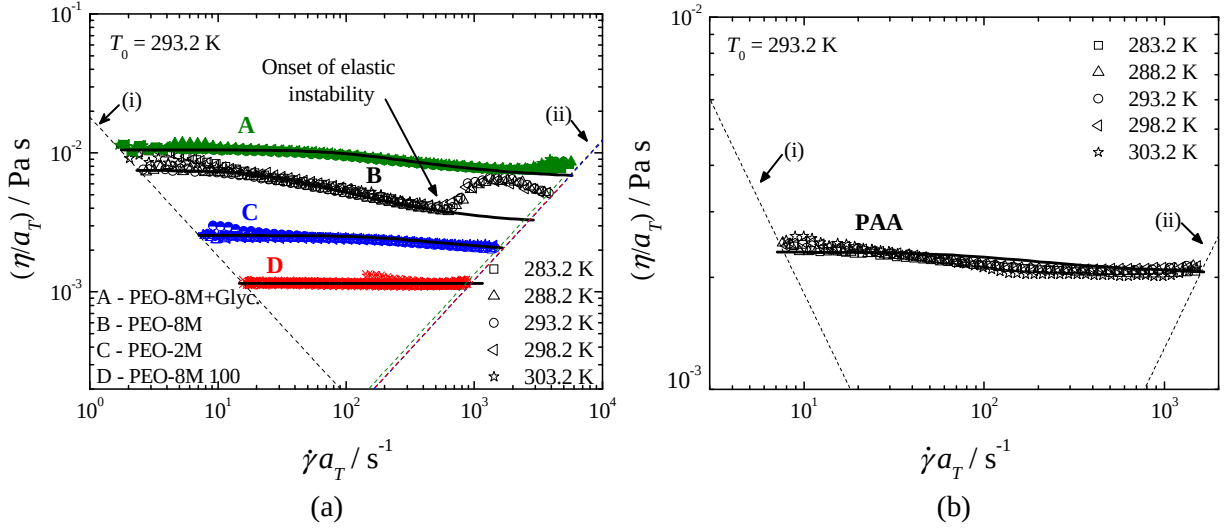


Figure 6.3. Master curves of the steady shear viscosity: (a) PEO solutions; (b) PAA solution. The solid lines represent the PTT model fittings for each fluid. (i) Minimum measurable shear viscosity calculated from $20\times$ the minimum measurable torque of the rheometer; (ii) onset of secondary flow due to Taylor instabilities (two distinct lines are drawn in (a) corresponding to different fluid densities).

In the PTT model, ε is an extensibility parameter that influences the extensional viscosity, η_p is the zero-shear viscosity of the polymer and η_s is the solvent viscosity. The zero-shear viscosity of the model in shear flow is simply given by: $\eta_0 = \eta_p + \eta_s$ and the solvent viscosity ratio is given by $\beta = \eta_s/\eta_0$. Here the parameter of the model related to the second normal stress difference, ξ , was kept equal to zero. Table 6.6 presents the rheological parameters of the sPTT model used in this work, including the Arrhenius equation parameter $\Delta H / R$ (cf. Eq. (6.5)).

In the fitting of the sPTT model we used the relaxation time obtained in the CaBER measurements (cf. Table 6.4). We fitted the rheological data to this model and report the

corresponding values in Table 6.6, in order to be possible to perform numerical simulations of the experiments here presented in future works.

Table 6.6. Rheological parameters of the PTT model at the reference temperature ($T_0 = 293.2$ K), and parameter $\Delta H / R$ for the fluids used. The relaxation times are presented in Table 6.4.

Fluid	η_0 / mPa s	ηS / mPa s	ε	$(\Delta H / R) / K$
PEO-2M	2.55	1.9	0.05	2.84×10^3
PEO-8M	7.50	3.0	0.04	2.98×10^3
PEO-8M+Glyc.	10.5	6.5	0.04	2.18×10^3
PEO-8M 100	1.15	1.0	0	2.31×10^3
PAA	2.34	2.0	0.1	1.76×10^3

6.3.2. Relevant dimensionless numbers

In addition to the Hencky strain, defined in Section 6.2.1 as $\varepsilon_H = \ln(D_1 / D_2)$, other dimensionless numbers are relevant for this work. The Reynolds number is defined here as:

$$Re = \frac{\rho V_2 D_2}{\eta_0}, \quad (6.8)$$

where V_2 is the mean velocity at the channel throat (with dimension D_2) (cf. Figure 6.1).

The flow of the viscoelastic fluids is also characterized using a Deborah (De) or a Weissenberg (Wi) number:

$$De = \lambda \dot{\gamma} = \frac{\lambda V_2}{D_2 / 2}, \quad (6.9)$$

$$Wi = \lambda \dot{\varepsilon} = \lambda \frac{V_2 - V_1}{L}, \quad (6.10)$$

in which the characteristic shear rate is estimated using the narrow dimension of the channels $\dot{\gamma} = V_2 / (D_2 / 2)$. The Weissenberg number is expressed in terms of the strain rate ($\dot{\varepsilon}$) along the microchannel, where V_2 and V_1 correspond to the average velocity at positions corresponding to D_2 and D_1 , respectively, separated by a distance L . The corresponding elasticity number is independent of flow rate and is defined as $El = De / Re = 2\lambda\eta_0 / \rho D_2^2$, assuming a constant value for each fluid/geometry combination.

The diodicity of the microchannels can be evaluated at a constant flow rate value, here defined as:

$$Di|_Q = \frac{\Delta P_{Forward}}{\Delta P_{Backward}}, \quad (6.11)$$

where $\Delta P_{Forward}$ is the pressure drop in the forward direction and $\Delta P_{Backward}$ is the pressure drop in the backward direction for the same flow rate Q . Alternatively, rectification effects can also be calculated at a constant pressure drop value:

$$Di|_{\Delta P} = \frac{Q_{Backward}}{Q_{Forward}}, \quad (6.12)$$

where $Q_{Backward}$ and $Q_{Forward}$ are the flow rates in the backward and forward flow directions for the same total pressure drop, ΔP , respectively.

6.4. Results and Discussion

Initially, a preliminary investigation was carried out on the flow of a single viscoelastic fluid in all hyperbolic microchannels in order to identify the most efficient rectifier under negligible inertia. Subsequently, the best geometry is investigated more extensively with all Newtonian and non-Newtonian fluids described in Section 6.3.1. Finally, the performance of the chosen geometry is compared with that of the triangular nozzle/diffuser rectifier proposed by Groisman and Quake (2004).

6.4.1. Preliminary study

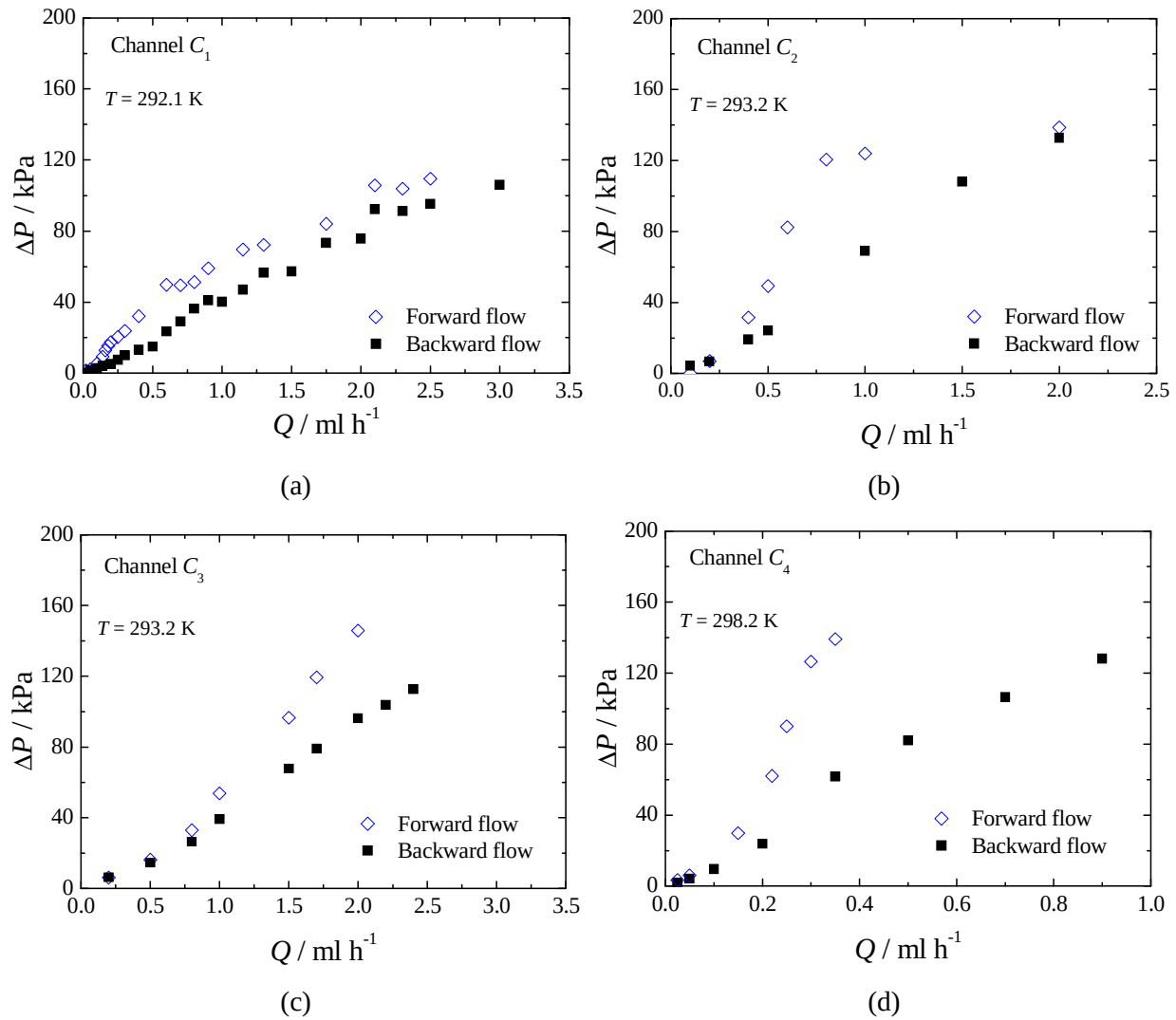


Figure 6.4. Pressure drop measured with the PEO-8M fluid in the forward and backward flow directions for a range of flow rates in different hyperbolic microchannels: (a) C_1 at 292.1 K; (b) C_2 at 293.2 K; (c) C_3 at 293.2 K; (d) C_4 at 298.2 K.

The flow of a single viscoelastic fluid was studied in the four hyperbolic microchannels C_1 , C_2 , C_3 and C_4 (cf. section 6.2.1) with the aim of establishing the most efficient microfluidic rectifier. The non-Newtonian fluid used for comparison purposes was the 0.1 wt.% aqueous solution of PEO-8M. Figure 6.4 shows the pressure drop measured as a function of the flow rate for both flow directions.

The results show a similar qualitative behavior in all microgeometries. At low flow rates the flow is quasi-Newtonian and reversible (negligible diodicity) and the pressure drop increases linearly with the flow rate. However, above a critical flow rate value (Q_{crit}), which is geometry-

dependent, the pressure drop increases nonlinearly with Q (or De). The critical conditions are $Q_{\text{crit}} \sim 0.05 \text{ ml h}^{-1}$ for channel C_1 , $Q_{\text{crit}} \sim 0.3 \text{ ml h}^{-1}$ for channel C_2 , $Q_{\text{crit}} \sim 0.6 \text{ ml h}^{-1}$ for channel C_3 and $Q_{\text{crit}} \sim 0.1 \text{ ml h}^{-1}$ for channel C_4 . This is a typical behavior of viscoelastic fluid flows through contractions at micro or macro-scales (e.g. Rodd *et al.*, 2005; Gulati *et al.*, 2008; Sousa *et al.*, 2009 (cf. Chapter 3); McKinley *et al.*, 2007). Furthermore, the flow resistance is different in the two flow directions and is higher when the fluid flows along the series of hyperbolic contractions in the forward direction, i.e. when the fluid flows in a quasi-homogeneous stretching extensional flow, than when it flows in the backward direction, in a quasi-homogeneous relaxing extensional flow. As a consequence of this different flow resistance, rectification effects emerge. The corresponding variation of diodicity is shown in Figure 6.5 as a function of flow rate for all micro-channels studied.

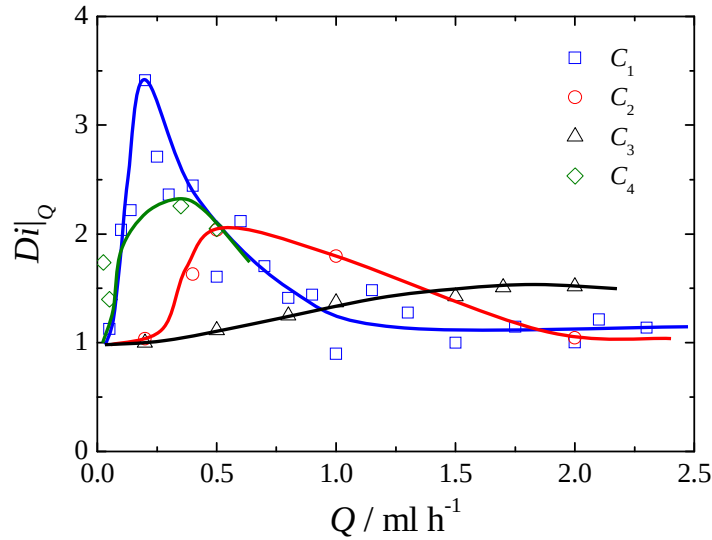


Figure 6.5. Diodicity obtained for the flow rate range studied in the four microchannels with hyperbolic shaped elements using PEO-8M fluid. The lines are a guide to the eye.

Channels C_1 and C_4 present the highest diodicities at low flow rates, namely at $Q \sim 0.2 \text{ ml h}^{-1}$ and $Q \sim 0.35 \text{ ml h}^{-1}$, respectively. The maximum diodicities ($Di|_Q$) are in excess of 3 and 2 for channels C_1 and C_4 , respectively. For channel C_2 , the maximum diodicity is about 2 (at a flow rate, $Q \sim 0.5 \text{ ml h}^{-1}$) while for channel C_3 , which has the lowest total Hencky strain, the measured diodicity is the lowest (maximum diodicity about 1.5 at $Q \sim 1.7 \text{ ml h}^{-1}$).

We anticipated that the highest rectification effects would be observed for the channel with the highest Hencky strain (channel C_4). However, this trend was not observed in the experiments, presumably because for geometry C_4 shear effects become important in the narrow part of the channel, with the boundary layer of the shear flow occupying a significant portion of the duct.

This apparent increase in shear effects counteracts the extensional effects that are necessary for the increase of the local pressure drop in the forward direction, with the corresponding increase of rectification effect. Thus, the channel with the best performance corresponds to a compromise between high extension and low shear and it is microgeometry C_1 , which exhibits the highest diodicity. We shall focus on this microchannel over the next sections.

6.4.2. Newtonian fluid flow

Figure 6.6 shows the effect of inertia on the visualized pathlines for the Newtonian fluid flow in channel C_1 taken at the center plane of the microgeometry.

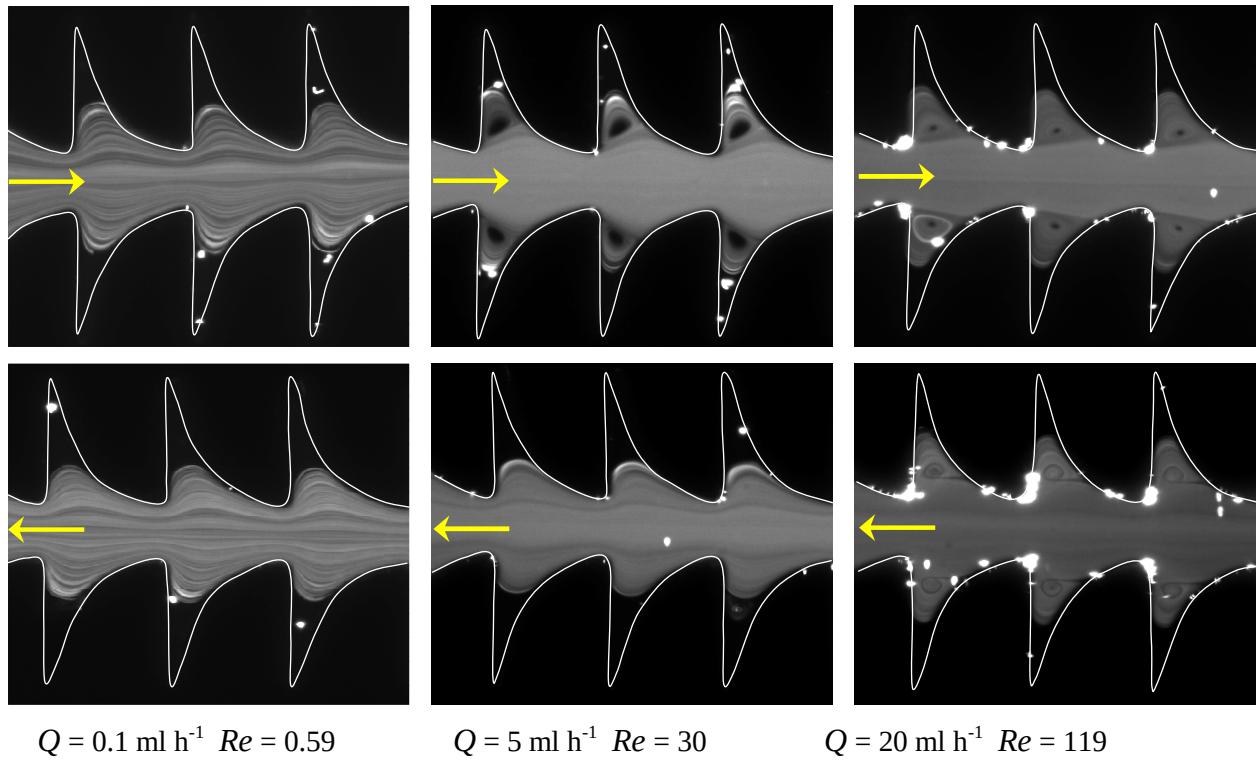


Figure 6.6. Flow patterns for Newtonian fluid (de-ionized water at $T = 296.2 \text{ K}$) flow. Top images correspond to forward flow and bottom images correspond to backward flow in microchannel C_1 .

At low flow rates ($Re \lesssim 0.6$), there is no significant flow separation and the flow patterns are similar for the two flow directions, due to the flow reversibility characteristic of Newtonian creeping flows. In the range $0.6 \lesssim Re \lesssim 6$ recirculations become visible between the expansion corners and the far corners and increase in size as Re increases for the forward flow direction. When the flow inertia increases further ($Re \gtrsim 6$), large recirculations are visible near the far corner in the forward direction, in agreement with the experimental results of Groisman *et al.*

(2004). Since the fluid is flowing through a sequence of abrupt expansions, inertia is responsible for the appearance and the subsequent growth of the recirculations, which is in agreement with the results found by Oliveira *et al.* (2007) and Rodd *et al.* (2005) for planar microchannels.

In the backward direction the recirculations also emerge and grow in size in the region between the corners, but these events occurred at Re higher than the corresponding Re in the forward flow direction. In all cases, the increase of the size of the recirculations with Re is limited by spatial confinement. For both flow directions, the fluid in the vicinity of the far corners is essentially stagnant. A similar behavior was also observed in the experiments of Groisman *et al.* (2004)

In order to assess the Newtonian fluid flow anisotropy, we conducted pressure drop measurements using de-ionized water in two geometries (the hyperbolic C_1 , and triangle T microchannels) and for both flow directions. The experiments were carried out for a wide range of flow rates, $Q \lesssim 45 \text{ ml h}^{-1}$ ($Re \lesssim 260$) for microchannel C_1 and $Q \lesssim 30 \text{ ml h}^{-1}$ ($Re \lesssim 180$) for microchannel T . The results are compiled and compared in Figure 6.7.

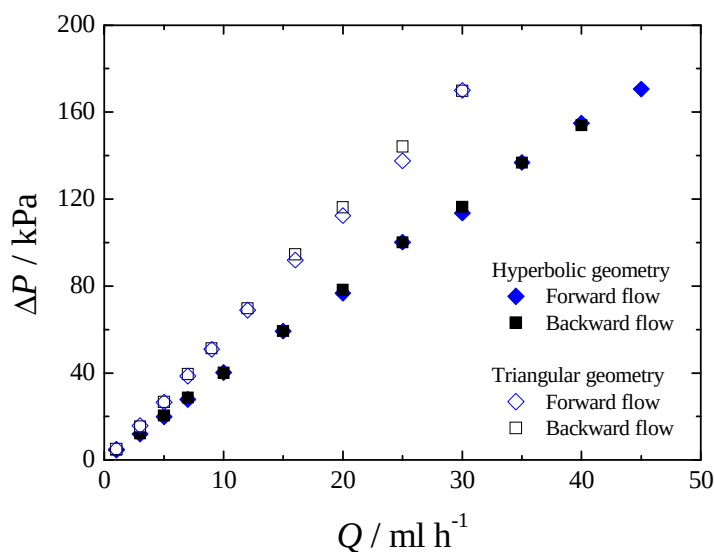


Figure 6.7. Pressure drop measured as a function of flow rate for the Newtonian fluid (de-ionized water) flow in the microchannel with a hyperbolic shape, C_1 (solid symbols) at 295.7 K and in the microchannel with a triangular shape, T (open symbols) at 298.5 K.

In the range of flow conditions studied, the pressure drop increases linearly with the flow rate for both microgeometries and flow directions. Furthermore, no rectification effects are found since the pressure drop in the two flow directions essentially coincide at the same flow rate, even at the highest Reynolds numbers measured, which is well inside the nonlinear range where inertial effects are important. Therefore, these microchannels are not efficient for operation with Newtonian fluids.

6.4.3. Viscoelastic fluid flow

6.4.3.1. Flow patterns

The flow patterns for all viscoelastic fluids are qualitatively similar as can be assessed from observation of Figure 6.8 to 6.12, where representative streak line images are shown to highlight the effects of the Deborah number and flow direction.

At low flow rates, viscous effects are dominant and the flow patterns (Figure 6.8(a), 6.9(a), 6.10(a), 6.11(a) and 6.12(a)) are similar to those obtained with the Newtonian fluid (Figure 6.6), i.e. they are identical for both flow directions rendering the flow fully reversible and rectification effects entirely absent.

Above a certain flow rate (or a critical Deborah number, De_{crit}), markedly different flow patterns are observed for the two flow directions. For the backward flow direction, regions of separated flow appear near the far corner in all elements of the microgeometry with the flow remaining symmetric and steady (Figure 6.8(b), 6.8(c), 6.9(b), 6.10(b), 6.10(c), 6.12(b) and 6.12(c) for backward flow). In this case, the fluid tends to enter inside the hyperbolic corner and the pathlines show a bent shape. The existence of upstream recirculations when a shear-thinning fluid flows through an abrupt contraction is a well-known phenomenon and the results obtained from flow visualizations are consistent with the numerical studies for a 2D channel presented by Alves *et al.* (2003) at the macroscale and the experimental results of McKinley *et al.* (2007) for a microchannel with a hyperbolic shape.

For the forward flow direction, recirculations also emerge near the far corner inside each microchannel element. However, in this case, elastic instabilities appear and the recirculations are only visible in some elements of the microgeometry (Figure 6.8(c), 6.8(d), 6.9(a), 6.9(c), 6.10(c), 6.12(b), 6.12(c) and 6.12(d) for forward flow direction), appearing and disappearing in time without a clearly discernible pattern. Consequently, due to the onset of this elastic instability, the flow is now asymmetric and unsteady.

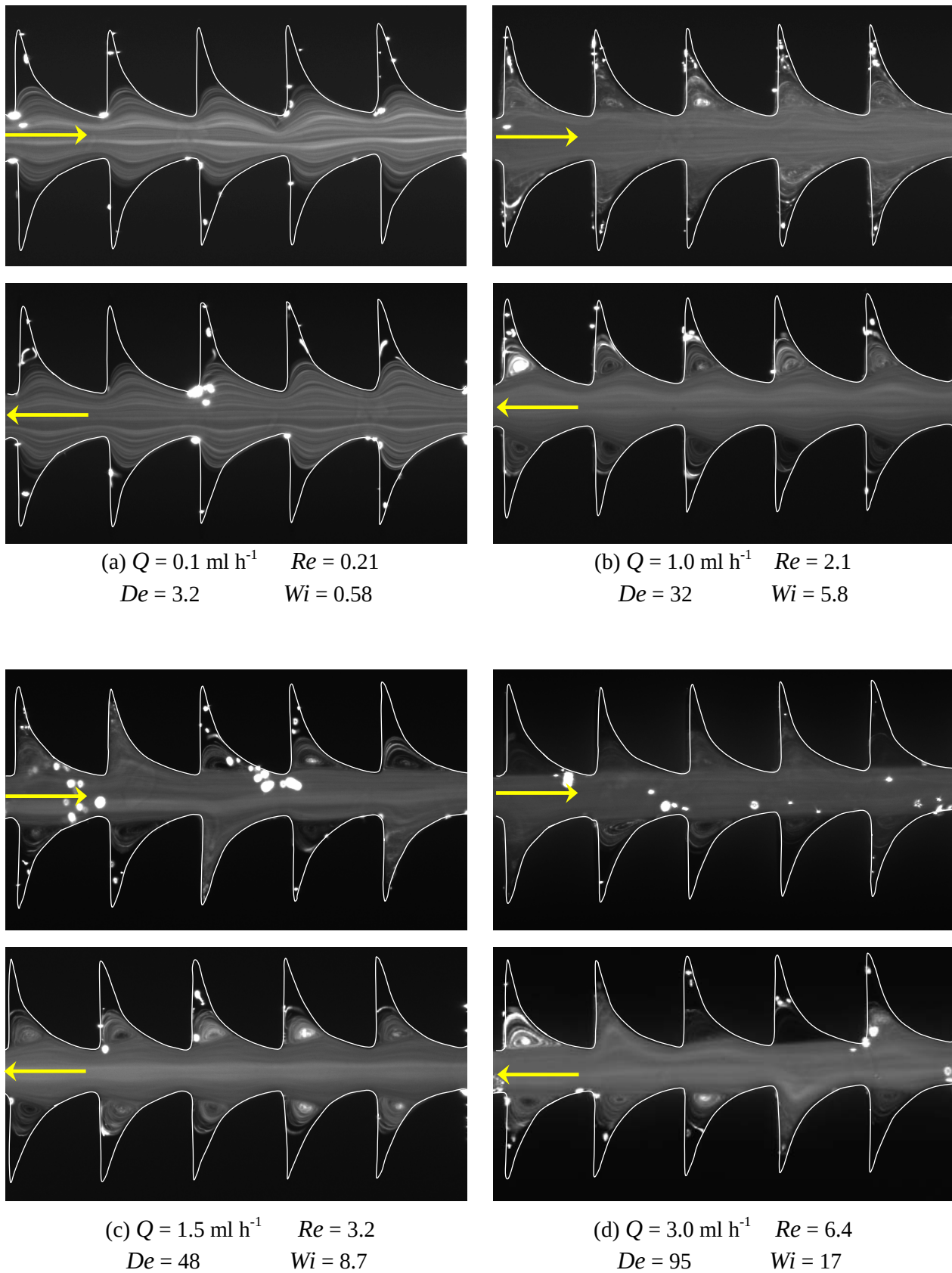


Figure 6.8. Effect of elasticity on the flow patterns of the PEO-2M fluid (microchannel C_1) at 292.7 K.

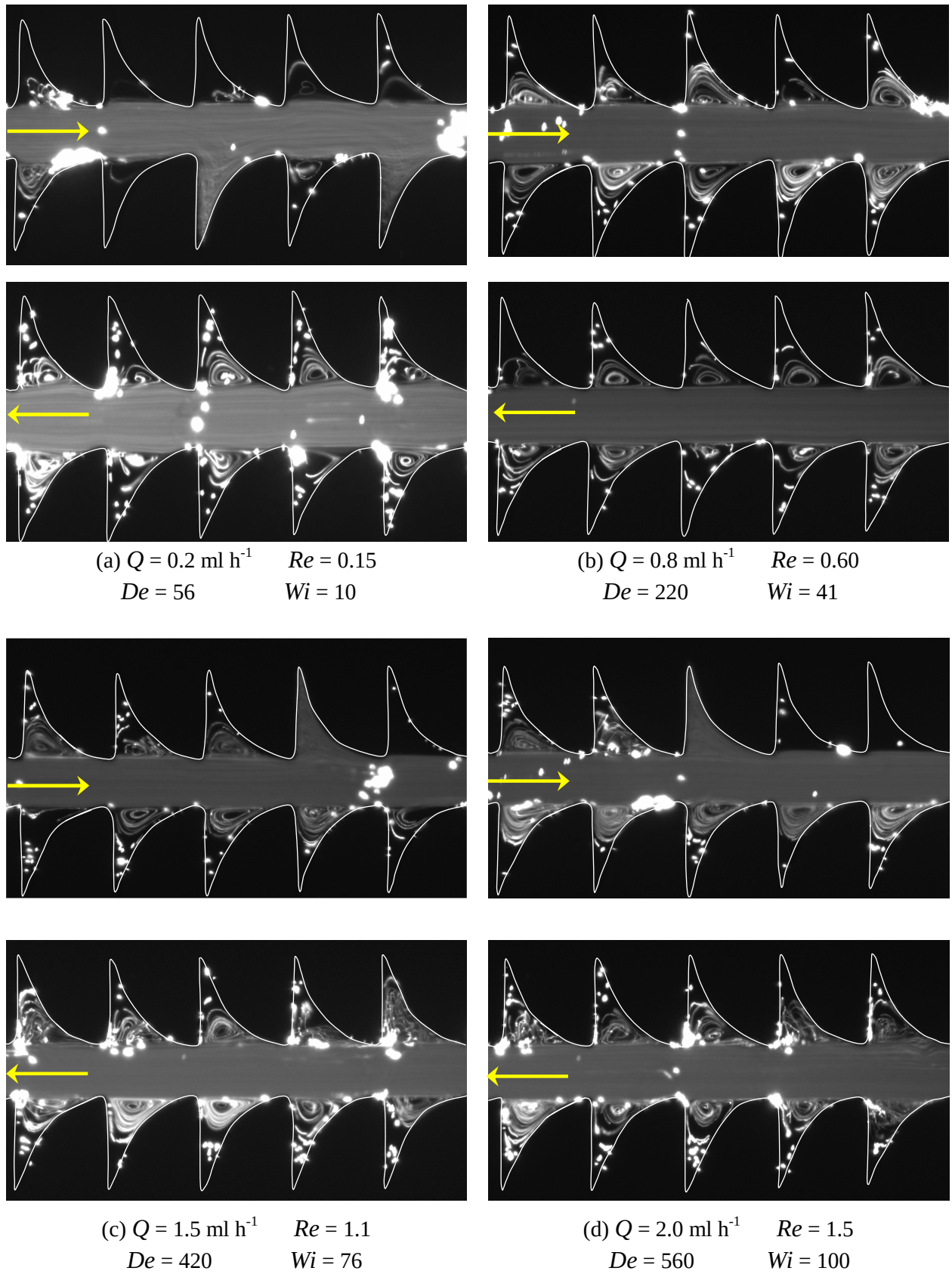


Figure 6.9. Effect of elasticity on the flow patterns of the PEO-8M fluid (microchannel C_1) at 293.5 K.

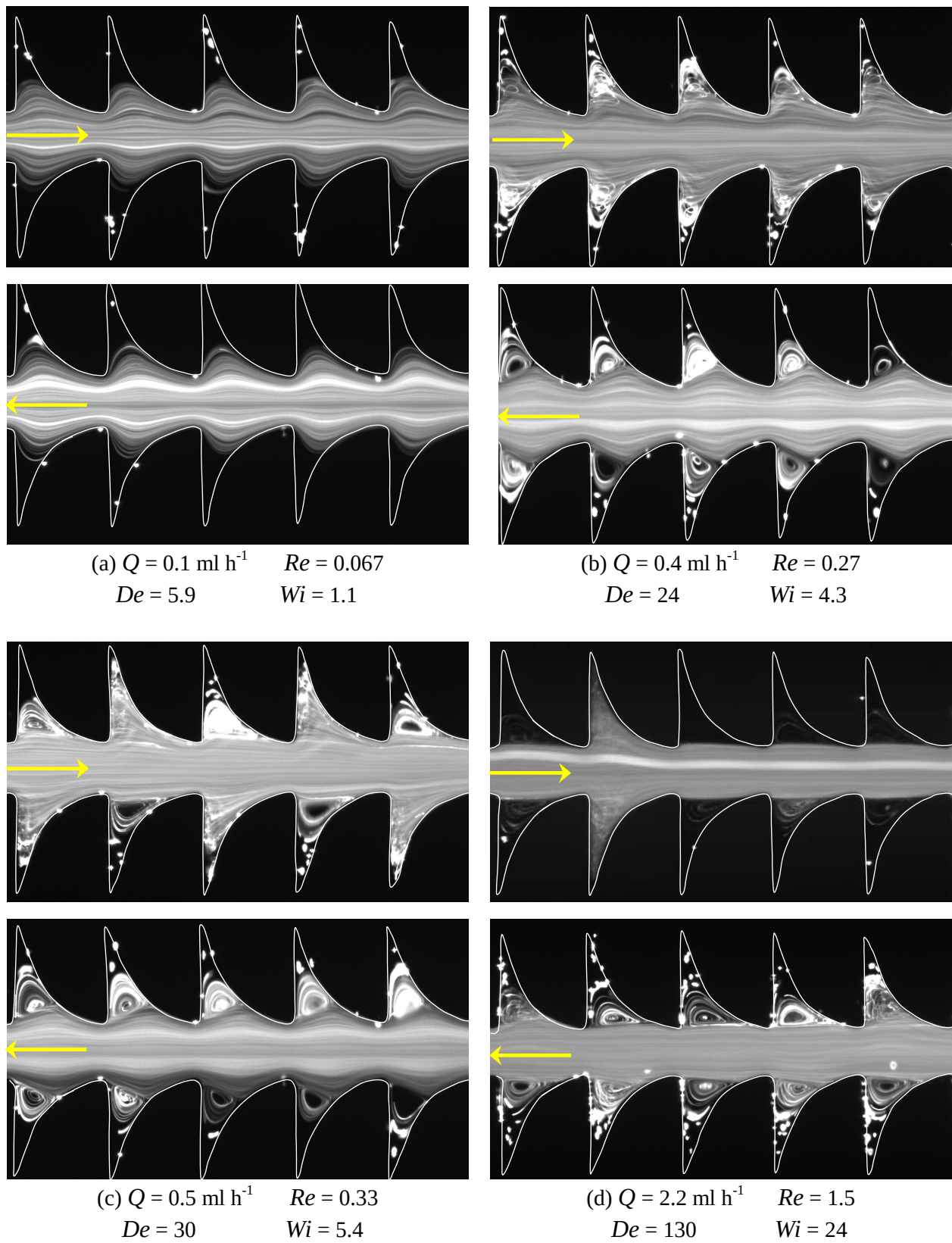


Figure 6.10. Effect of elasticity on the flow patterns of PEO-8M+Glyc fluid (microchannel C_1) at 298.8 K.

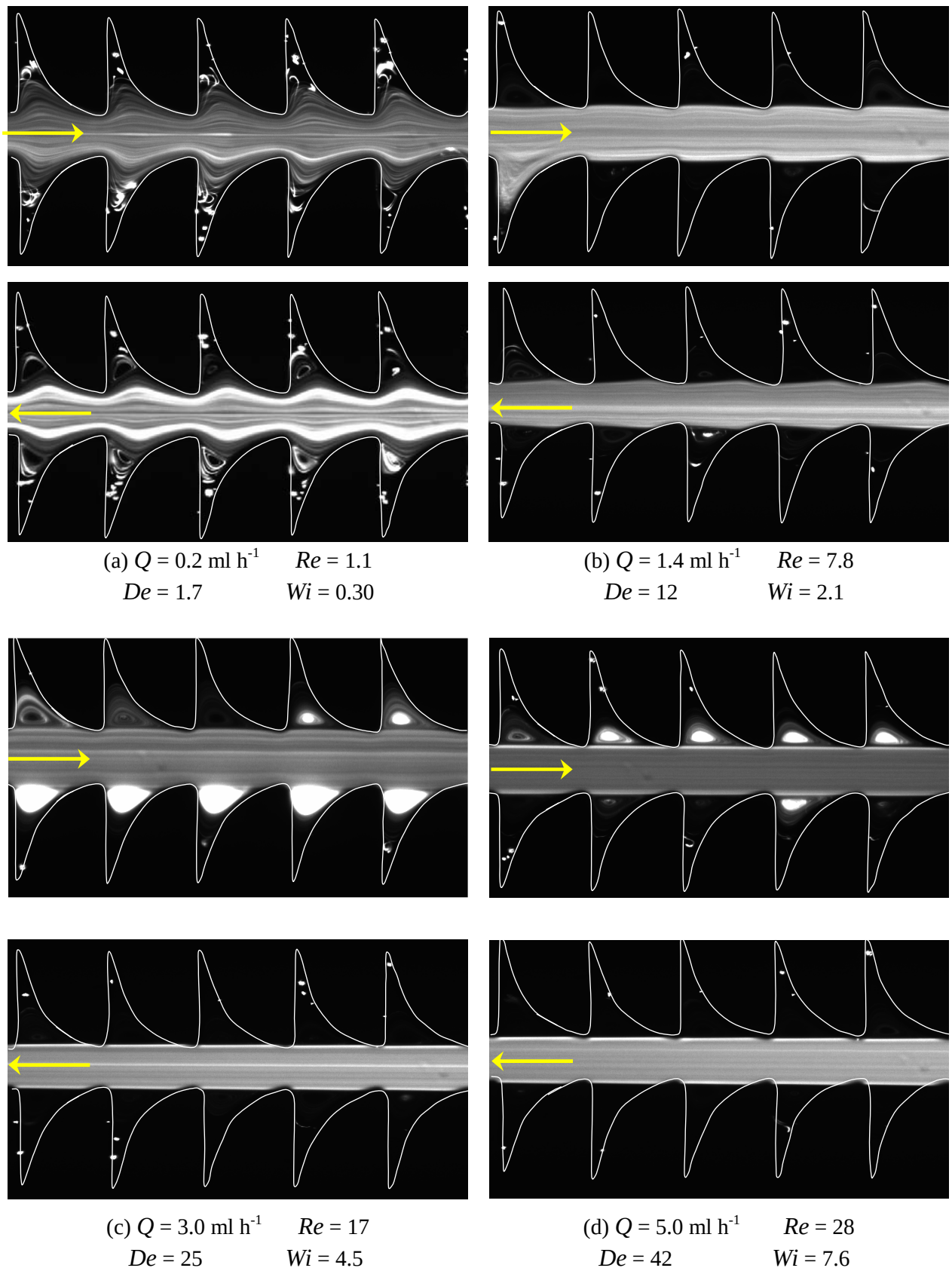


Figure 6.11. Effect of elasticity on the flow patterns of the PEO-8M 100 fluid (microchannel C_1) at 298.4 K.

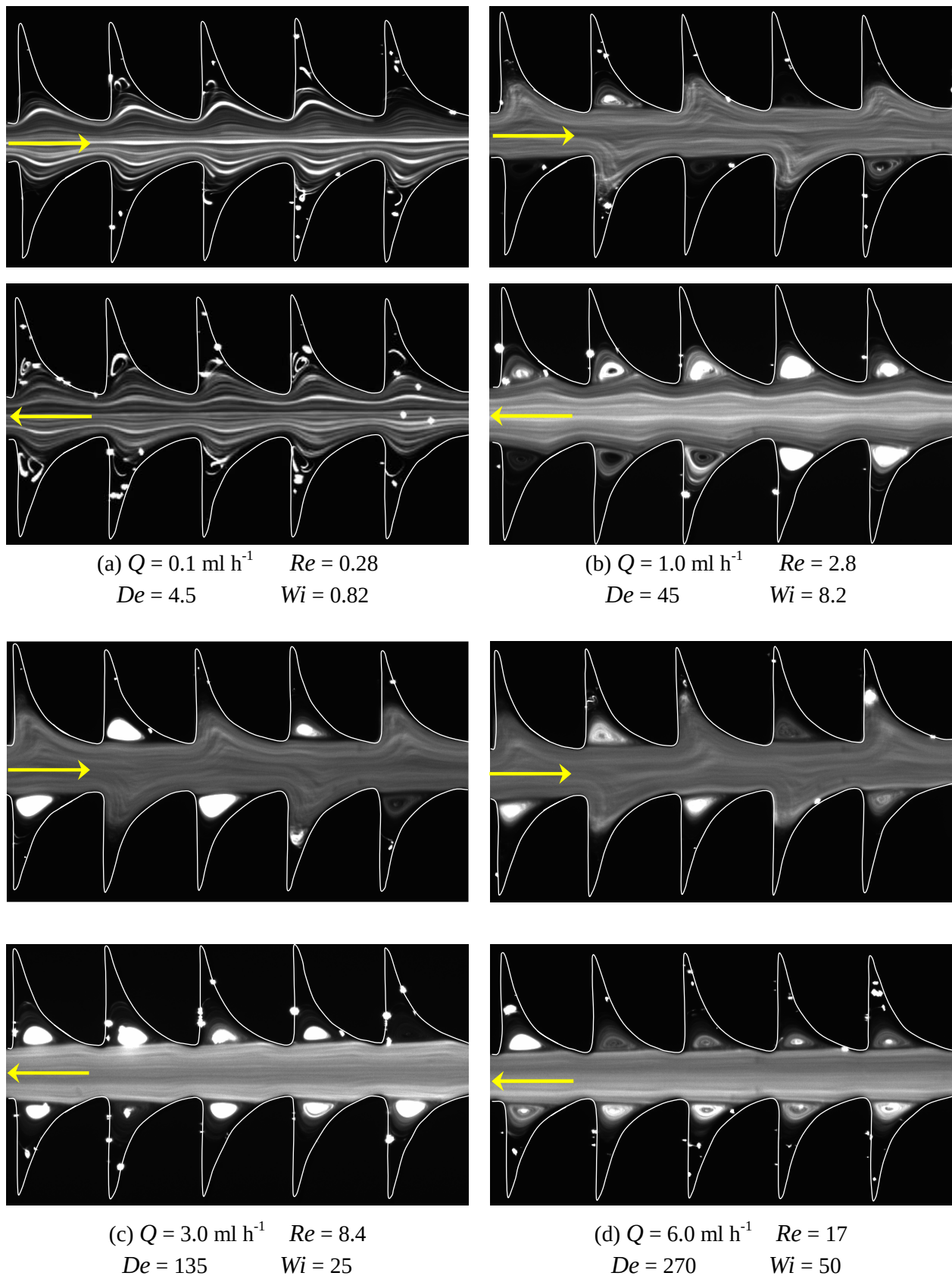


Figure 6.12. Effect of elasticity on the flow patterns of the PAA fluid (microchannel C_1) at 298.5 K.

These instabilities are elastic in nature since inertial effects are not important (low Reynolds number flows). Purely elastic instabilities in microchannels have been studied by Arratia *et al.* (2006) and Poole *et al.* (2007) in cross-slot microchannels, by Oliveira *et al.* (2009) in flow focusing geometries with 3 entrances and by Soulages *et al.* (2009) in T-shaped microgeometries. See also Morozov and van Saarloos (2007) and papers within for a discussion on elastic instabilities.

For the backward flow direction, we also observe the onset and enhancement of elastic instabilities (Figure 6.8(d) for backward flow), but these occur at higher flow rates. The nonlinear increase of the flow resistance also occurs above a critical flow rate, but to a smaller extent than those found in the forward flow direction, at least for the range of flow rates studied in our experiments.

Another interesting flow characteristic is that the fluids tend to flow mostly through the central region/portion of the microchannel and the streamlines are no longer curved (cf. Figure 6.8(a), 6.8(b), 6.10(a), 6.10(b), 6.11(a), 6.12(a)), becoming progressively rectilinear as the flow rate increases. This phenomenon is observed to some extent for all viscoelastic fluids studied, especially at high Deborah numbers (Figure 6.10(d), 6.11(b), 6.11(c), 6.11(d)).

Streak line images obtained in the triangular shaped microchannels (inspired by the work of Groisman and Quake (2004)) are shown in Figure 6.13 for the PEO-8M fluid and in Figure 6.14 for the PAA solution.

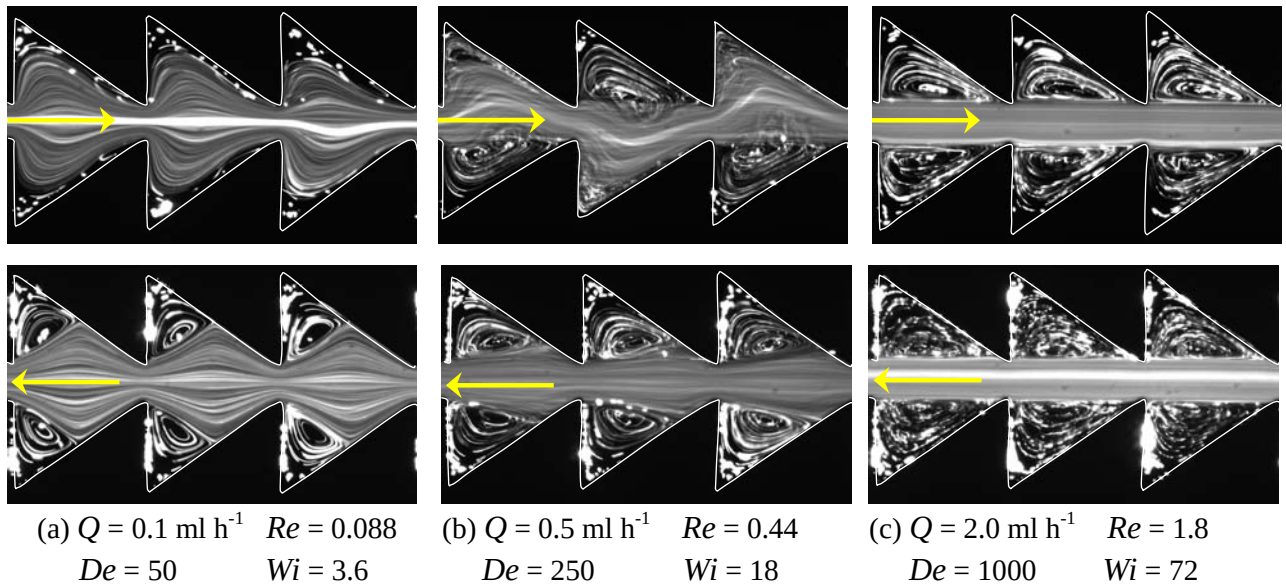


Figure 6.13. Flow patterns of the PEO-8M fluid flow in the microchannel with a triangular shape (T) at 298.4 K.

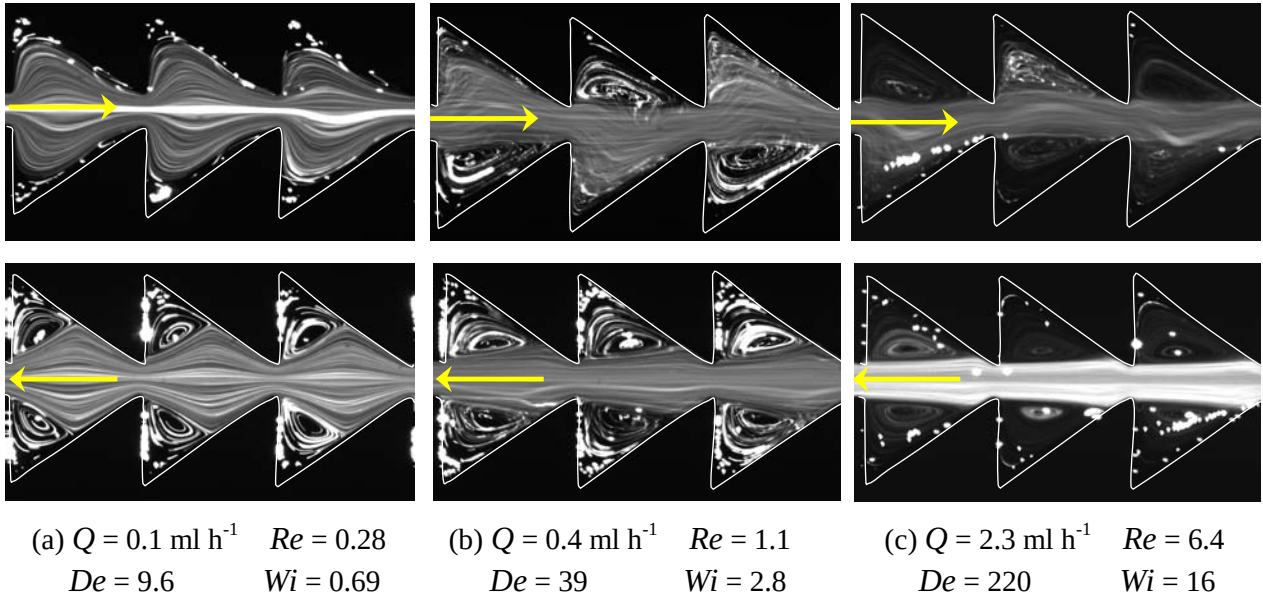


Figure 6.14. Flow patterns of the PAA fluid flow in the microchannel with a triangular shape (T) at 298.5 K.

The flow behavior observed for this microgeometry has some general similarities to that obtained with the hyperbolic geometries for all viscoelastic fluids studied, but also bears some important differences. The forward flows at low De exhibit recirculations near the far corner, and show small lip vortices emerging in the vicinity of the throat (Figures 6.13(a), and 6.14(a)), whereas in the backward direction, the recirculations become visible at lower flow rates than in the forward direction (Figure 6.13(a), and 6.14(a)). Increasing De , elastic effects become important, and instabilities appear in the forward flow direction (Figure 6.13(b), 6.14(b) and 6.14(c)) leading to unstable flow. When De is increased further, the pathlines become symmetric, steady and are similar for the two flow directions (Figure 6.13(c)). These results are in agreement with the findings of Groisman and Quake (2004) in a geometry with the same shape (the only difference here is the smaller depth of $50 \mu\text{m}$ as compared with $100 \mu\text{m}$ in their study).

6.4.3.2. Pressure drop

Figure 6.15 presents the variation of the dimensionless pressure drop with flow rate measured in channel C_1 for all non-Newtonian fluids considered. The flow resistance behavior in the two flow directions is analogous for all viscoelastic fluids and resembles the preliminary results presented in Section 6.4.1. At low De , the flow is quasi-Newtonian and reversible, but above a critical value (De_{crit}) the dimensionless pressure drop increases sharply with the flow rate and starts to differ in the forward and backward directions, being invariably larger in the former.

The flow rate at the transition point in the pressure drop profile is significantly different for all fluids studied, as illustrated in Table 6.7, which stems from the different fluid rheology. In Table 6.7 the corresponding critical Deborah numbers, calculated with the relaxation time at the temperature of the experiments, are included. These relaxation times were determined from the values measured with the CaBER at $T = 293.2$ K and then corrected to the temperature of the experiments using the time-temperature superposition principle (cf. Section 6.3.1). In Table 6.7 we also include the estimated critical Weissenberg number (Wi_{crit}), showing that the critical conditions occur for a Weissenberg number close to the coil-stretch transition, $\lambda\dot{\epsilon} = 0.5$ (Larson and Magda, 1989), in particular for the PAA and the lighter PEO, an indication suggesting that the significant increase in the pressure drop is related to the strong extensional behavior of the flow along the forward direction.

Cross analyzing Table 6.7 with the flow patterns reported in Section 6.4.3.1, we conclude that De_{crit} corresponds approximately to the conditions when the flow becomes unsteady in the forward flow direction, and enhanced flow resistance anisotropy sets in due to the elastic instability. When the flow is unsteady we report the average pressure drop, together with the maximum and minimum values measured. These extreme values are reported as horizontal bars (cf. Figure 6.15), while the symbols correspond to the time-averaged values.

Table 6.7. Critical conditions for the onset of diodicity in the C_1 hyperbolic microchannel, and average temperature of the experiments.

Fluid	$Q_{crit} / \text{ml h}^{-1}$	T / K	De_{crit}	Wi_{crit}
PEO-2M	0.13	293.1	4.1	0.74
PEO-8M	0.08	292.1	23	4.3
PEO-8M+Glyc.	0.35	295.4	23	4.1
PEO-8M 100	0.80	298.7	6.6	1.2
PAA	0.04	298.8	1.8	0.33

For the forward direction, the pressure drop variation with flow rate shows a sharp increase above De_{crit} . Thus, the onset of elastic instabilities leads to an enhancement of the flow resistance in the forward direction and as a consequence rectification effects (or diodicity) emerge, since the same flow rate generates significantly different pressure drops in the forward and backward flow directions. When the flow instabilities in the backward direction also emerge, at higher flow rates, the difference between the forward and backward pressure drops decreases, and the rectification effect is reduced.

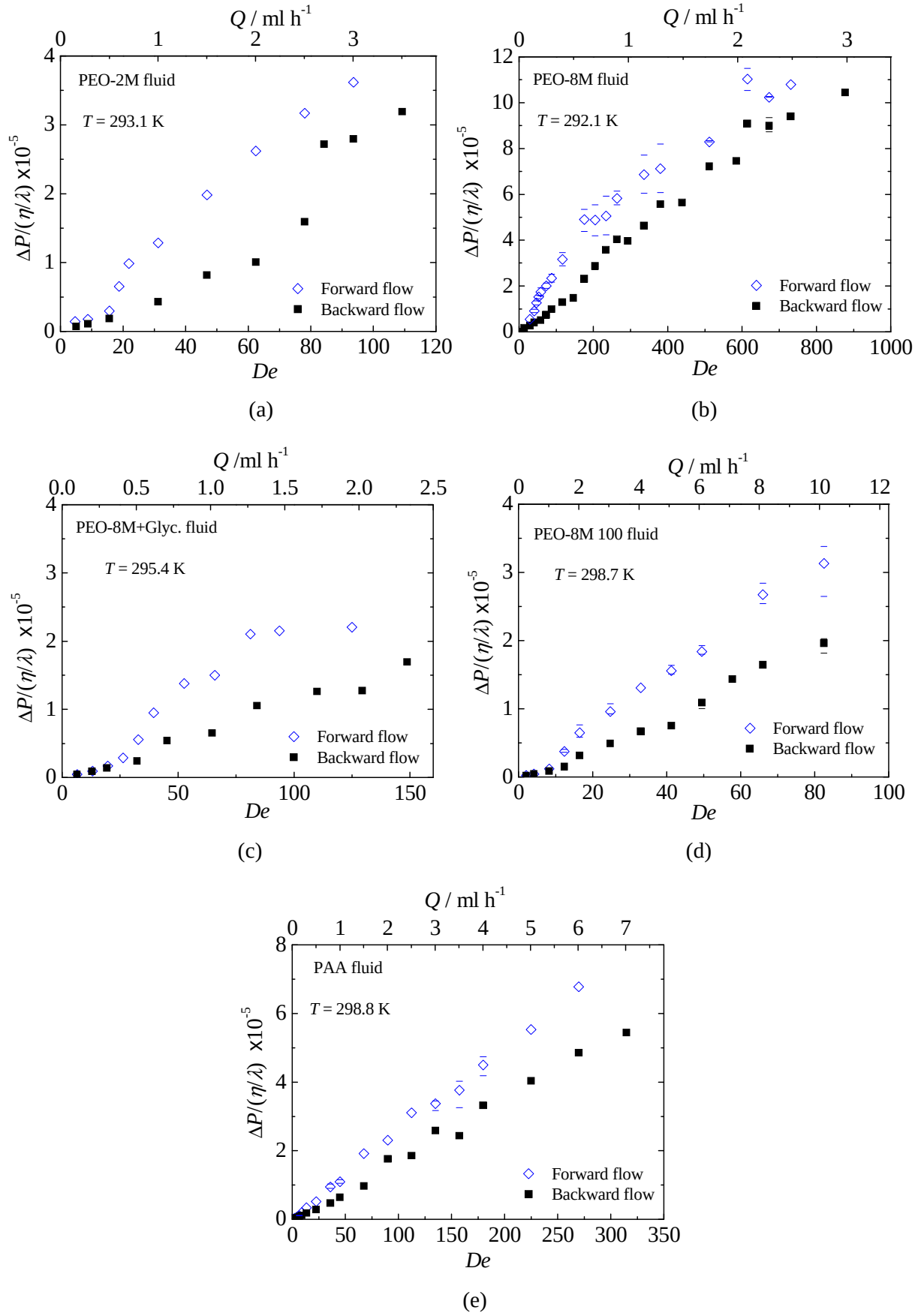


Figure 6.15. Dimensionless pressure drops in the forward and backward directions of microchannel C_1 , as a function of the flow rate and Deborah number for: (a) PEO-2M, (b) PEO-8M, (c) PEO-8M+Glyc., (d) PEO-8M 100 and (e) PAA.

To better understand the origin of the enhancement of the flow resistance we carried out a simple theoretical analysis which assumes that the main flow occurs primarily in the central region of the microchannel (as shown in Figure 6.9(b)) when elastic effects are important. Under this simplified scenario, the measured flow resistance can be compared with two limiting pure shear flow cases: (i) the flow confined in a rectangular channel with four walls, corresponding to no slip conditions at the walls and (ii) the flow between two parallel plates spaced $50\text{ }\mu\text{m}$ apart, which corresponds to the depth of the microchannel. Case (i) assumes a no-slip condition at the interface between the main flow and the recirculation, an assumption that is reasonable when the secondary flow in the recirculation is weak, and therefore the velocity of the dividing streamlines (assumed as straight lines) is small. This case corresponds to the flow in a rectangular channel, for which the velocity profile and pressure drop expressions can be found in White (2003) for a Newtonian fluid under fully developed flow conditions. The viscoelastic fluids tested have a nearly constant shear viscosity (cf. Figure 6.3(a)), hence these expressions for a Newtonian fluid are adequate (indeed, they are exact for fluids with constant shear viscosity and zero second-normal stress difference, either Newtonian or viscoelastic). The zero-shear viscosity of the viscoelastic fluids was used in the analytical solutions, which is a reasonable estimate due to the weak shear thinning behavior of the fluids. As an example, for the PAA fluid the decrease of the shear viscosity is only about 15 %, in a range of two decades of shear rates. Case (ii) assumes full slip at the dividing streamline, thus the flow field is assumed uniform in the x - y plane (cf. Figure 6.1). The bottom and top walls are planar and set $50\text{ }\mu\text{m}$ apart, thus this ideal case corresponds to the flow between two infinite parallel plates. Again, we assume fully developed flow conditions, and Newtonian behavior with a viscosity matching the zero-shear rate viscosity of the fluids. The predictions of these two ideal pure shear flows are shown as solid lines in Figure 6.16. When viscoelastic effects are important it is clear that the flow resistance measured in our microfluidic rectifier is higher than that predicted for the two limiting conditions, even higher than for the flow in a rectangular channel, which can be viewed as the upper limit of flow resistance if only shear effects were at play. This gives further credit to the hypothesis that enhanced flow resistance is due primarily to the strong extensional flow along the central region of the microchannel.

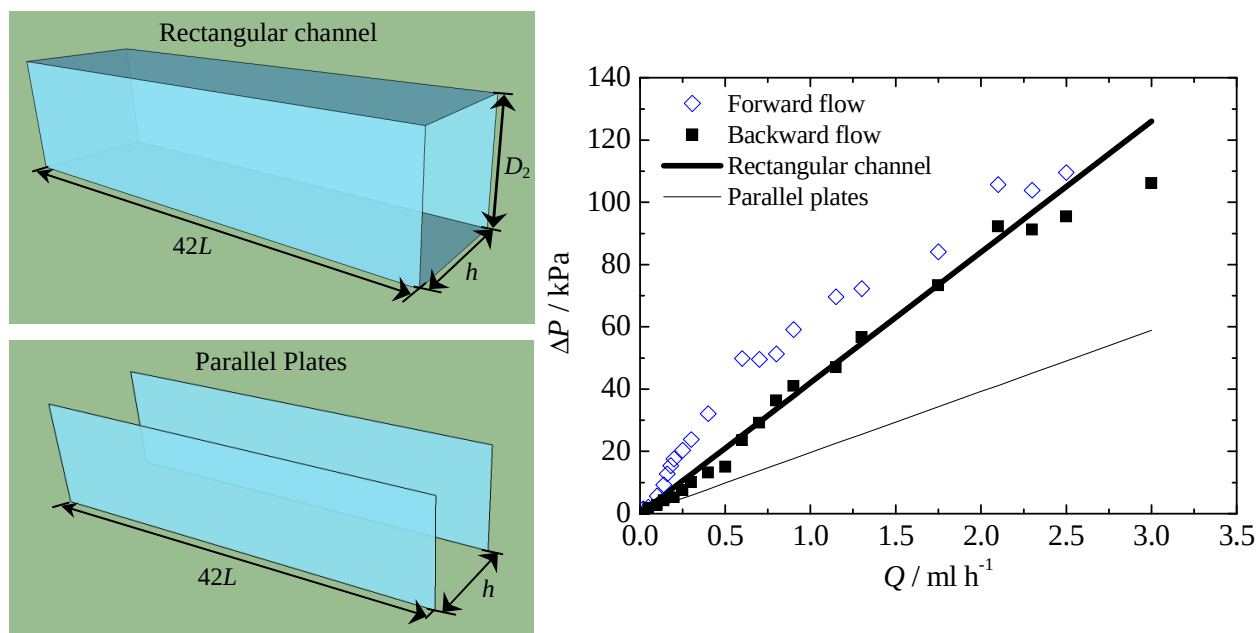


Figure 6.16. Pressure drop as a function of the flow rate for the PEO-8M fluid flow. Comparison with the pressure drop estimated for flow in a rectangular channel and between two parallel plates.

In order to compare the diodicity of the flows in the microfluidic device C_1 for the different viscoelastic fluids studied, we plot in Figure 6.17 the diodicity calculated in terms of the pressure drop ratio. As can be seen, the 0.1 wt.% aqueous solution of PEO-8M shows the highest diodicity, achieving a maximum value around 3.5. This fluid has the highest concentration of the heaviest polymer used here. The semi-dilute solution of PEO-2M presents a maximum diodicity of about 3 and the other fluids have rectification effect values in excess of 2, which are all significantly higher than values reported in other studies for viscoelastic fluids in triangular-based geometries (Groisman and Quake, 2004; Nguyen *et al.*, 2008). When plotted as function of De (or Wi), the diodicity follows a similar trend for all fluids, as shown in Figure 6.17(b), exhibiting less scatter than Figure 6.17(a).

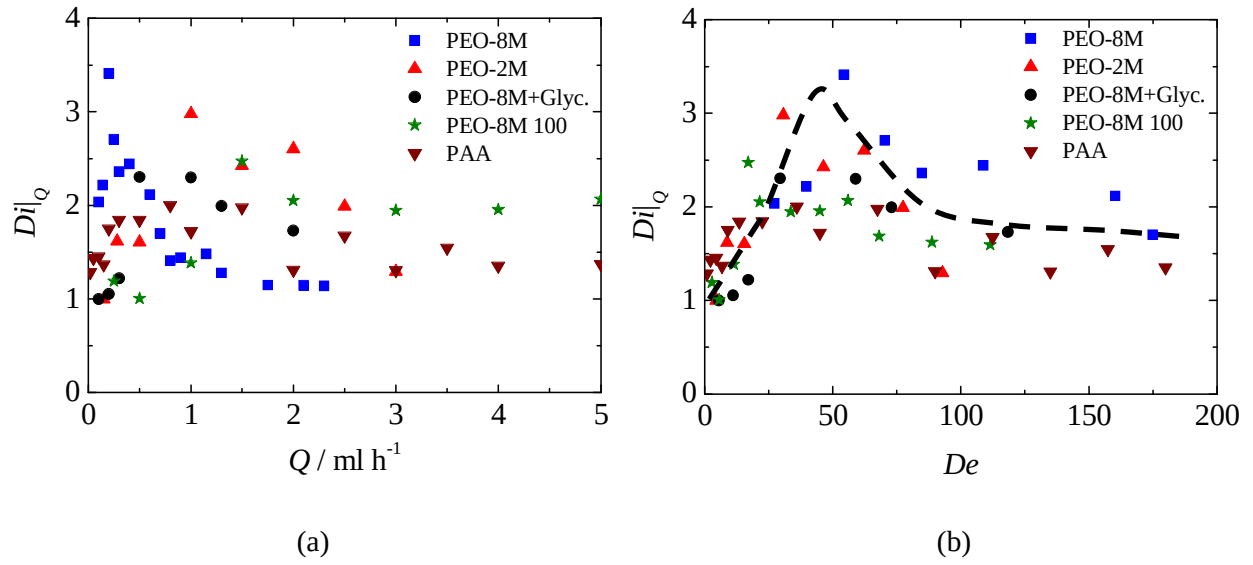
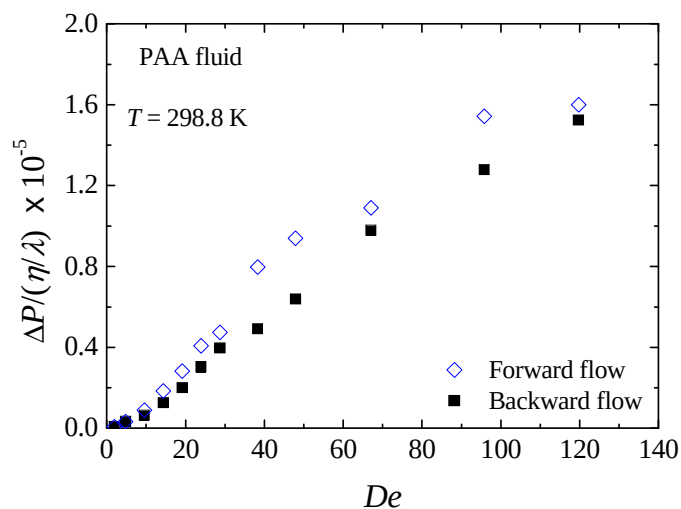


Figure 6.17. Diodicity (in terms of pressure drop ratio) as a function of flow rate (a) and Deborah number (b) for all viscoelastic fluids used. The dashed line in (b) is a guide to the eye.

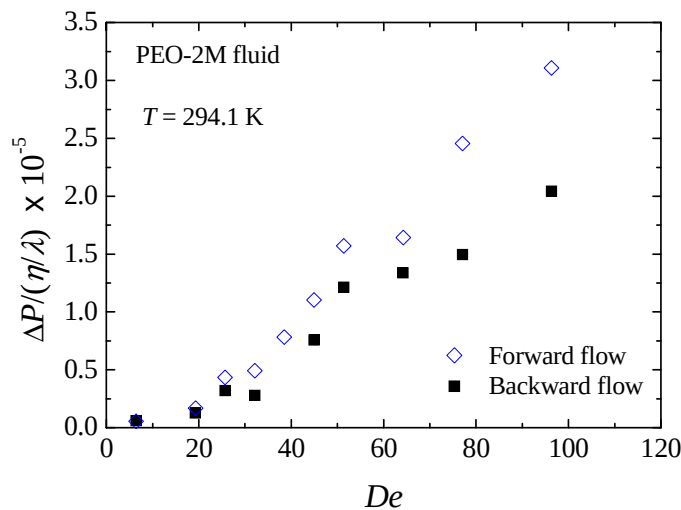
In order to make a direct comparison between the results obtained in the hyperbolic rectifier and the triangular geometry proposed by Groisman and Quake (2004) it is necessary to use microgeometries with similar aspect ratios and equal depth. This is achieved here by using the microchannel T of Table 6.2.

In Figure 6.18(a) we plot the dimensionless pressure drop as a function of De in the two flow directions for the T microrectifier for the PAA solution, which is similar to the fluid used by Groisman and Quake (2004). Figure 6.18(b) and 6.18(c) display the same quantities in the T microchannel, but for the solutions of PEO-2M and PEO-8M, respectively.

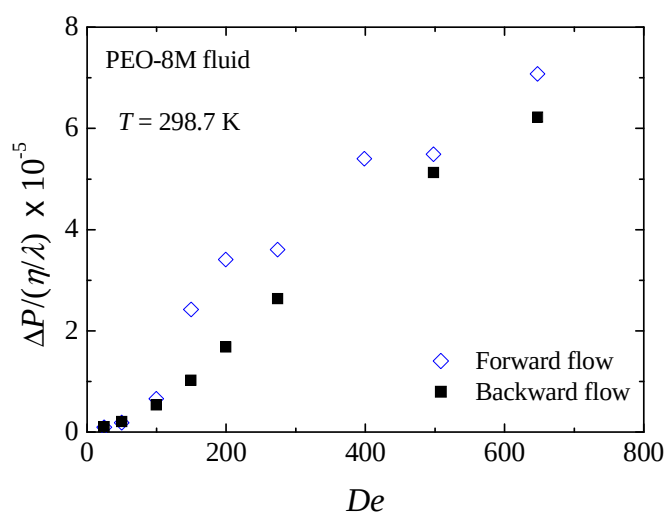
Qualitatively, the variations of the pressure drop with flow rate are similar to those seen for the hyperbolic rectifier, but the difference between the forward and backward pressure drops are clearly smaller. This is better seen in Figure 6.19, where the two diodicities ($Di|_{\Delta P}$ and $Di|_Q$) of all comparable cases are plotted.



(a)



(b)



(c)

Figure 6.18. Dimensionless pressure drop measured in the two flow directions of triangular shaped channel (T) as a function of the Deborah number for (a) PAA, (b) PEO-2M and (c) PEO-8M fluids.

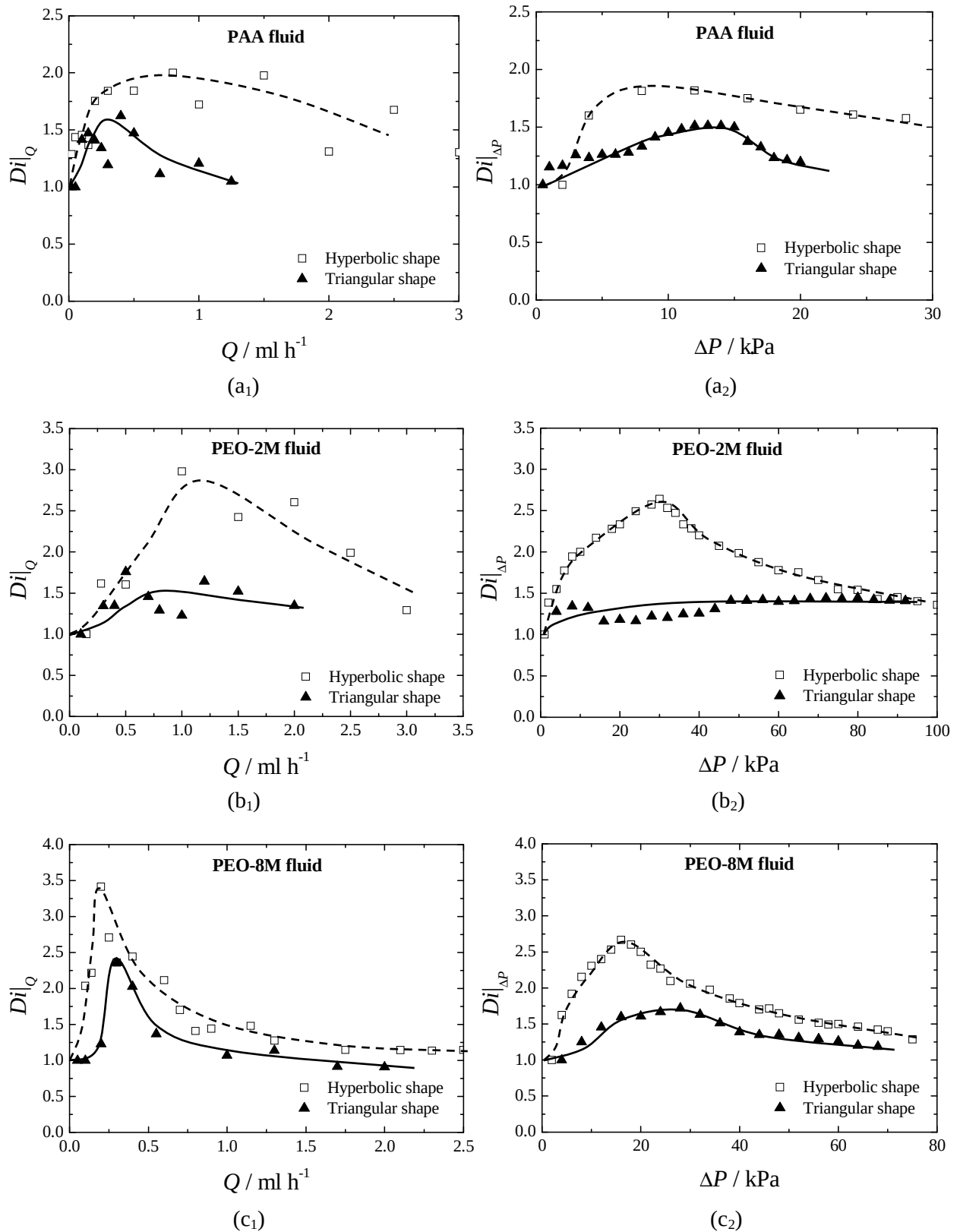


Figure 6.19. Comparison between the diodicity obtained with the hyperbolic and triangular microgeometries in terms of pressure drop ratio as a function of the flow rate (left-hand side column) and in terms of flow rate ratio as a function of the pressure drop (right-hand side column). The graphs (a₁) and (a₂) correspond to the fluid PAA, (b₁) and (b₂) to PEO-2M and (c₁) and (c₂) to PEO-8M. The curves are a guide to the eye.

Since in the experiments we imposed a value of the flow rate and the corresponding pressure drop was measured, we can directly determine the pressure drop ratio for all values of the flow rate considered ($Di|_Q$). On the other hand, to calculate the diodicity in terms of flow rate ratio, it was first necessary to fit the experimental data of ΔP vs Q , in order to allow for the interpolation of the data and the determination of the corresponding flow rate ratio. For this reason, the plot of the $Di|_Q$ vs Q has more scatter (provided from experimental results) than the smoother plot of $Di|_{\Delta P}$ vs ΔP calculated from the fits. In all cases there is a clear advantage of the rectifier with hyperbolic shaped elements both in terms of the ratio between the forward and backward pressure drop for a given flow rate, and also in terms of flow rate ratio, as used by Groisman and Quake (2004) in their seminal paper on elasticity driven flow anisotropy under creeping flow conditions.

For the PAA solution, a maximum value of $Di|_Q \approx 2$ is obtained for the hyperbolic geometry while for the triangular geometry a maximum value of approximately 1.6 is observed. In terms of $Di|_{\Delta P}$, the hyperbolic rectifier has a maximum diodicity of about 1.8, while the triangular geometry achieves a maximum value of about 1.5. However, we should emphasize that the value determined here is lower than the values reported by Groisman and Quake (2004), due to the difference in channel depth (and consequently of aspect ratio) between their microchannel (100 μm) and ours (50 μm). For the other viscoelastic fluids higher values of $Di|_Q$ and $Di|_{\Delta P}$ were obtained for both rectifiers, but always showing higher diodicity for the hyperbolic shape.

6.5. Conclusions

The flow of Newtonian and viscoelastic fluids through microfluidic channels that are able to operate as microfluidic rectifiers under creeping flow conditions, was studied. Two different shapes of the microchannels were compared: the nozzle/diffuser arrangement based on triangular elements used in the literature (Groisman and Quake, 2004) and a new rectifier based on elements with hyperbolic shape (Oliveira *et al.*, 2007) that are especially suitable to generate a nearly purely extensional flow.

For Newtonian fluid flow, the flow patterns for both rectifiers are qualitatively similar in the two flow directions and the flow is steady for the wide range of flow rates studied. The pressure drop through the microchannels is found to increase linearly with the flow rate and no noticeable rectification effects were observed even though the maximum Reynolds number achieved for both flow directions was of the order of 250. However, the secondary flow that is induced by

flow inertia is found to appear at lower Re for the forward direction, but it has a negligible influence on the pressure drop.

For the polymeric solutions, the pressure drop is also independent of the flow direction at low flow rates, when the flow is quasi-Newtonian. However, due to the viscoelastic nature of the fluids used, increasing the flow rate leads to an anisotropic flow resistance between the two flow directions. At the onset of rectification effects, elastic instabilities emerge in the forward flow direction as the flow becomes unsteady and the flow resistance increases sharply with the flow rate. Although this phenomenon was observed in both types of microgeometries, the hyperbolic shaped microchannels present a higher diodicity. For channels with the same depth (50 μm), the triangular shaped channel presents a maximum diodicity in terms of pressure drop ratio of about 2.5, whereas the hyperbolic shaped microchannel has a maximum diodicity of 3.5. The hyperbolic shape of the contraction region for forward flow leads to a quasi-ideal extension flow that generates significant increases in pressure drop, thus the maximum diodicity values found for the various viscoelastic fluids are significantly higher than those achieved with the nozzle/diffuser (triangular) geometry proposed by Groisman and Quake (2004), and also higher than those reported by Nguyen *et al.* (2008).

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VISCOELASTIC FLUID FLOW IN A MICROFLUIDIC RECTIFIER: WALL EFFECT ANALYSIS

Abstract

The flow of Newtonian and non-Newtonian fluids within microfluidic rectifiers with a hyperbolic shape was studied with the aim of understanding the effect of the bounding walls on the diodicity of the microfluidic device. For this, we employed two microchannels with different depths but similar geometrical configuration, which was selected in order to generate a strong extensional flow and attain high anisotropic flow resistance between the two flow directions. The Newtonian fluid, de-ionized water, was used as a reference fluid and for validation purposes. The viscoelastic fluid used was an aqueous solution of polyethylene oxide (0.1 % w/w) with high molecular weight ($M_w = 8 \times 10^6 \text{ g mol}^{-1}$). The flow patterns were visualized using streak line photography and the velocity field was investigated in detail using micro-particle image velocimetry. Moreover, pressure-drop measurements were performed in order to compare the diodicity achieved in the two microfluidic rectifiers. For the Newtonian fluid flow, the experimental results are also compared with numerical predictions obtained using a finite-volume method and good agreement was found between both techniques. For the viscoelastic fluid there is anisotropic resistance in both microchannels, with the different channel depth affecting both the onset and magnitude of diodicity. The diodicity is enhanced when the wall effect is reduced, i.e. when the channels are deeper, but its onset is also delayed to higher Deborah numbers and significantly higher flow rates. A maximum diodicity of almost seven was found for the deeper channel (higher aspect ratio ($AR = \text{depth}/\text{width}$)).

7.1. Introduction

Microfluidic devices are systems with integrated channels, whose dimensions are of the order of tens to hundreds of microns, which operate with small amounts of fluids. A current challenge of engineering science is to scale-down these systems (process intensification) in order to achieve integrated and portable “lab-on-a-chip” tools able to operate with high resolution and sensitivity, short analysis time, low cost and reduced volumes of reagents and waste (Whitesides, 2006). Microfluidic technology is applied across fields: in biology for cellular analysis (Gómez-Sjöberg *et al.*, 2007) or DNA separation (Tegenfeldt *et al.*, 2004); in medicine for clinical diagnostics (Verpoorte, 2002); in chemistry for undertaking chemical reactions (deMello, 2006).

The small length scales that characterize these microsystems lead typically to low Reynolds numbers (Re) and consequently laminar flow conditions. Furthermore, surface forces, which are often negligible in flows at the macroscale, may become significant in microfluidics due to the strong intermolecular forces (Stone and Kim, 2001) and the inherent higher ratio of surface area to fluid volume. The fluids of interest in such lab-on-a-chip applications have often non-Newtonian rheological behavior, as for example saliva (Helton and Yager, 2007), blood (Tovar-Lopez *et al.*, 2009), several suspensions or other fluids containing small amounts of polymers (Mall-Gleissle *et al.*, 2002; Oliveira and McKinley, 2005). The non-linear rheological properties of these fluids result in complex flow phenomena, which need to be better understood in order to optimize the design of microfluidic devices.

Viscoelastic fluid flow is prone to instabilities, which in microfluidics are often purely elastic as inertial effects are small. These purely elastic instabilities, as documented in Muller *et al.* (1989) and Larson *et al.* (1990) for Taylor-Couette flow, occur above a critical Deborah number (De) and result from the combination of curved streamlines and large normal stresses. Pakdel and McKinley (1996) developed a criterion that predicts the critical conditions for the onset of purely elastic instabilities. Utilizing a microfluidic cross-slot geometry, Arratia *et al.* (2006) found experimentally that a viscoelastic fluid under creeping flow conditions can undergo different types of instabilities. These authors reported the onset of a first instability, in which the flow becomes asymmetric but remains steady, followed by a second instability, in which the flow becomes unsteady, with the amplitude of oscillation increasing as the flow rate increases. Subsequently, Poole and Alves (2007) demonstrated that the steady asymmetry observed in the cross-slot geometry can be predicted numerically for creeping flow conditions, using the

simplest differential viscoelastic model, the upper-convected Maxwell (UCM) model. Broad attention has been paid recently to geometries in which elastic instabilities emerge in highly elongational flow fields, such as cross-slot (Pathak and Hudson, 2006), T-shaped (Soulages *et al.*, 2009) and flow-focusing devices (Oliveira *et al.*, 2008, 2009). Elastic instabilities can be useful for mixing purposes at low Reynolds numbers (Gan *et al.*, 2006). In fact, purely elastic instabilities stimulate a disordered state of the flow brought about by elastic stresses even at vanishingly small Re , giving rise to the concept of elastic turbulence (Groisman and Steinberg, 2000, 2001). With the fluid motion occurring over a wide range of temporal and spatial scales, elastic turbulence generates enhancement of flow resistance and increases the mixing performance (Groisman and Steinberg, 2004).

Taking advantage of elastic nonlinearities, Groisman and co-workers (Groisman *et al.*, 2003; Groisman and Quake, 2004) developed different microfluidic devices which operate as control and memory elements. The authors exploited the viscoelastic rheological properties of a dilute polymeric solution and found that increasing the applied pressure leads to complex flow behavior, due to the differences between coil stretching and relaxation, and consequently the variation of the flow rate was found to be small for a wide range of applied pressures (Groisman *et al.*, 2003). A microfluidic rectifier with no moving parts was also described using the nonlinearities brought about by elastic stresses in viscoelastic fluid flow (Groisman and Quake, 2004). In the proposed microfluidic device, consisting of a series of triangular cavities, the non-Newtonian fluid experiences a different deformation history when flowing in the two possible flow directions, leading to significant anisotropic flow resistance. The development of efficient flow rectifiers is an important area of research in microfluidics, with particular application in the development of piezoelectric-driven micropumps. The technology of microscale pumping has been the aim of several investigations with applications as diverse as drug delivery systems, fuel delivery in micro-engines and cooling in micro-electronics (Navabi, 2009). However, despite the interest of non-Newtonian fluid flows in microfluidics, in these studies, the flow of Newtonian fluids has been much more investigated and studies of microfluidic rectifiers using viscoelastic fluids are still scarce (Groisman and Quake, 2004; Nguyen *et al.*, 2008; Sousa *et al.*, 2010 (cf. Chapter 6)).

With the aim of designing a more efficient microfluidic rectifier for operation with viscoelastic fluids under creeping flow conditions, we presented in our previous work (Sousa *et al.*, 2010 (cf. Chapter 6)) a comparison between the efficiency of distinct geometries, namely a

triangular nozzle/diffuser rectifier proposed in the literature (Groisman and Quake, 2004; Nguyen *et al.*, 2008) and a new rectifier based on hyperbolic-shaped elements (Sousa *et al.*, 2010 (cf. Chapter 6)). A detailed investigation, in terms of pressure drop measurements and visualizations of the flow patterns for different viscoelastic fluids as well as for a Newtonian reference fluid, was carried out. In that work, a hyperbolic-shaped geometry with a Hencky strain $\varepsilon_H = 2$ was identified to exhibit the highest diodicity. Additionally, the importance of wall effects imposed by the shallow depth of the microchannels and its influence on the diodicity arose as an important topic for further research. Therefore, in the present study, we investigate the wall effects on the diodicity using the most efficient hyperbolic shaped microfluidic rectifier presented in the previous work (Sousa *et al.*, 2010 (cf. Chapter 6)). For this purpose, we use two microchannels with similar shape, but having different depths, $h = 46 \mu\text{m}$ and $h = 120 \mu\text{m}$. Pressure-drop measurements in addition to visualization of the flow patterns and micro-particle image velocimetry (μPIV) measurements were carried out for different flow rates. The non-Newtonian fluid used here is an aqueous solution of a polyethylene oxide (PEO) with high molecular weight ($M_w = 8 \times 10^6 \text{ g mol}^{-1}$), hereafter labeled as PEO-8M, which presented the highest rectification effect of all the fluids tested previously (Sousa *et al.*, 2010 (cf. Chapter 6)). Moreover, the flow of a Newtonian fluid (de-ionized water) is also investigated in order to validate the experimental techniques and to be used as reference. For the Newtonian fluids a set of numerical calculations are also carried out using an in-house code and its predictions agree very well with the experimental results and help understand them.

7.2. Experimental

7.2.1. Experimental techniques

Two types of microchannels were designed to have the same generic geometrical shape but different depths (h). The devices, fabricated in polydimethylsiloxane (PDMS) using standard soft lithography (Xia and Whitesides, 1998), include two inlets/outlets located at the extremes of the channels and two pressure taps on each side of the test section of the microgeometry. In Figure 7.1(a) we show an optical transmission microscope image of a typical microchannel used in the experiments. A scanning electron microscopy (SEM) image of the $46 \mu\text{m}$ deep microchannel is also shown in Figure 7.1(b) to illustrate the well-defined and quasi-vertical walls of the PDMS microchannels. The general hyperbolic function that was used to describe the shape of the channels is $y = \pm a / (1 + b x)$, with $a = 200 \mu\text{m}$ and $b = 0.05 \mu\text{m}^{-1}$; x is the axial position ($0 \leq x \leq L$);

L is the length of each hyperbolic element ($L = 128 \mu\text{m}$). The chrome mask used in the manufacturing process follows closely this hyperbolic shape and was used to make the molds from which both channels were produced, which in principle would only differ in depth. However, the actual molds are slightly different due to inaccuracies inherent to the photolithography fabrication procedure. The characteristic dimensions of the final PDMS channels are given in Table 7.1, including the total Hencky strain (ϵ_H), here defined as $\epsilon_H = \ln(D_1/D_2)$.

Two microchannels with the same geometrical shape but different depth (h) were used in this investigation. The devices, fabricated in polydimethylsiloxane (PDMS) using standard soft lithography (Xia and Whitesides, 1998), were equipped with two inlets/outlets located at the extremes of the channels and two pressure taps on each side of the microgeometry. In Figure 7.1 we show scanning electron microscopy (SEM) images of the microchannels used, which illustrate the well-defined walls of the PDMS microchannels. The hyperbolic function that describes the shape of the channel is $y = \pm a/(x + b)$, where x is the position in the range $0 \leq x \leq L$; a and b are the parameters of the function, which are described in Table 7.1.

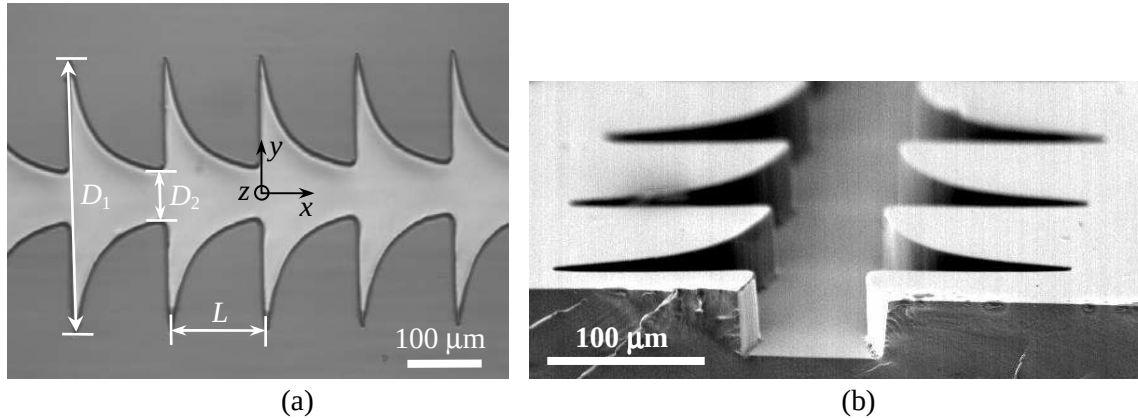


Figure 7.1. (a) Optical micrograph of the hyperbolic microfluidic rectifier A with $46 \mu\text{m}$ in depth. Microchannel B has a similar geometric shape, but has $120 \mu\text{m}$ in depth. (b) SEM image illustrating the verticality of the walls of microchannel A. Only part of the microchannels is shown.

Table 7.1. Dimensions of the microchannels, values of Hencky strain and parameters of the hyperbolic function used to describe the shape of the channels.

	Projected (chrome mask)			PDMS microchannel				
	$D_1 / \mu\text{m}$	$D_2 / \mu\text{m}$	ϵ_H	$L / \mu\text{m}$	$h / \mu\text{m}$	$D_1 / \mu\text{m}$	$D_2 / \mu\text{m}$	ϵ_H
<i>Channel A</i>	400	54	2.0	128	46	325.4	62.7	1.65
<i>Channel B</i>	400	54	2.0	128	120	326.0	69.6	1.54

The actual shape of the hyperbolic elements of both PDMS channels was imaged and using the software DataThief III (available as shareware at <http://www.datathief.org/>) the real shape of the hyperbolic walls of the PDMS channels was fitted to the following expressions:

Channel A ($h = 46 \mu\text{m}$)

$$\begin{cases} |y / \mu\text{m}| = 153.7 + 2.25\sqrt{4^2 - (x - 4)^2} & 0 < x / \mu\text{m} \leq 7.66 \\ |y / \mu\text{m}| = \frac{220}{1 + 0.052x} + 15e^{(x-128)/3} & 7.66 < x / \mu\text{m} < 128 \end{cases} \quad (7.1)$$

Channel B ($h = 120 \mu\text{m}$)

$$\begin{cases} |y / \mu\text{m}| = 155.5 + \sqrt{7.5^2 - (x - 7.5)^2} & 0 < x / \mu\text{m} \leq 14.9 \\ |y / \mu\text{m}| = \frac{380}{1 + 0.097x - 9.1 \times 10^{-5}x^2} + 10e^{(x-128)/4} & 14.9 < x / \mu\text{m} < 128 \end{cases} \quad (7.2)$$

The configuration of the experimental set-up used in this work is identical to that employed in a previous work (Sousa *et al.*, 2010 (cf. Chapter 6)). Flow is imposed by means of a syringe pump (PHD2000, Harvard Apparatus), used to feed the fluid to the channels and control the volumetric flow rate.

For pressure drop measurements, the pressure taps in the microchannel were connected to a pressure transducer (Honeywell, model 26PC series) in order to measure the differential pressure drop inside the test section. A 12 V DC power supply (Lascar electronics, PSU 206) was used to power the pressure sensors that were also connected to a computer, via a data acquisition card (NI USB-6218, National Instruments). The output data was recorded using LabView v7.1. The

pressure sensors were calibrated using a static column of water or using a compressed air line depending on the pressure range. A detailed description can be found in Sousa *et al.* (2010) (cf. Chapter 6).

Visualizations of the flow patterns and measurements of the velocity field were performed at the center-plane (i.e. mid-distance between top and bottom planar bounding walls) of the microchannel. The imaging experimental set-up is composed of an inverted epi-fluorescence microscope (DMI 5000M, Leica Microsystems GmbH) equipped with a filter cube (Leica Microsystems GmbH, excitation filter BP 530-545 nm, dichroic 565 nm and barrier filter 610-675 nm) as well as with a light source, objective and digital camera appropriate for each optical technique used.

For visualizations of the flow patterns, we employed streak line photography. For this, the light source used was a 100 W mercury lamp. The fluids were seeded with 1 μm fluorescent tracer particles (Nile Red, Molecular Probes, Invitrogen, Ex/Em: 520/580 nm) and the streak images were captured through a 10 $\times$ (NA = 0.25) objective lens using a CCD camera (DFC350 FX, Leica Microsystems GmbH).

For micro-particle image velocimetry measurements, the fluorescent particles used were 0.5 μm in diameter and the light source was a double-pulsed 532 nm Nd:YAG laser (Dual Power 65-15, Dantec Dynamics). Images were captured using a 20 $\times$ (NA = 0.4) microscope objective and a CCD digital camera (Flow Sense 4M, Dantec Dynamics), with a resolution of 2048 $\times$ 2048 pixels and running on double frame mode. The time interval between pulses was adjusted so that the displacement of the particles between two frames was about 25% of the width of the interrogation area. Interrogation areas of 64 $\times$ 32 pixels, with 50% overlap were used to create the vector maps. For steady flow, the vector maps were obtained by ensemble averaging 150 pairs of images. On the other hand, for unsteady flow, we present both instantaneous and ensemble-average vector maps attained from a set of 500 and 1000 pairs of images for the rectifiers A ($h = 46 \mu\text{m}$) and B ($h = 120 \mu\text{m}$), respectively.

The depth over which there is a contribution of unfocused particles to the velocity field determined by PIV is called the total measurement depth, δz_m , and can be calculated as the sum of three different effects: the effect due to diffraction, the effect due to geometrical optics and the

effect due to the finite size of the particle (Meinhart *et al.*, 2000), respectively the first, second and third terms on the right-hand-side of Eq. (7.3):

$$\delta z_m = \frac{3n\lambda_0}{(\text{NA})^2} + \frac{2.16d_p}{\tan \theta} + d_p, \quad (7.3)$$

where n is the refractive index, λ_0 is the wavelength of the light (in vacuum), NA is the numerical aperture of the microscope objective, d_p is the particle diameter. For the streak imaging optical set-up, the measurement depth is $\delta z_m = 37 \mu\text{m}$, whereas for the μPIV set-up, $\delta z_m = 14 \mu\text{m}$.

7.3. Fluid rheology

The fluids used in this investigation consist of a Newtonian fluid (de-ionized water) and a non-Newtonian fluid, which is an aqueous solution 0.1 wt.% of a high molecular weight polyethylene oxide (PEO, $M_w = 8 \times 10^6 \text{ g mol}^{-1}$, Sigma-Aldrich). The viscoelastic fluid density, measured at the reference temperature ($T_0 = 293.2 \text{ K}$), is $\rho = 998 \text{ kg m}^{-3}$.

The rheological properties of the viscoelastic solution were characterized in both steady shear and transient uniaxial extension flow and were described in detail in Sousa *et al.* (2010) (cf. Chapter 6), where a thorough description of the rheological characterization performed experimentally can be found.

The rheological measurements were fitted to a simplified form of the Phan-Thien-Tanner (sPTT) model (Phan-Thien and Tanner, 1977) with a Newtonian solvent contribution and the parameters obtained in the fitting of the master curve are: $\varepsilon = 0.04$; $\eta_p = 0.0045 \text{ Pa s}$; $\lambda = 73.9 \text{ ms}$. The relaxation time used in the sPTT fitting was determined from CaBER measurements and the Newtonian solvent viscosity is $\eta_s = 0.003 \text{ Pa s}$. More details can be found in Sousa *et al.* (2010) (cf. Chapter 6).

7.4. Numerical

7.4.1. Governing equations and numerical method

The Newtonian fluid flow was simulated numerically using a fully-implicit finite-volume method with a time marching pressure-correction algorithm (Oliveira *et al.*, 1998). The

governing equations that describe an isothermal, laminar and incompressible fluid flow are those of conservation of mass and momentum:

$$\nabla \cdot \mathbf{u} = 0, \quad (7.4)$$

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right] = -\nabla p + \nabla \cdot \boldsymbol{\tau}, \quad (7.5)$$

where p is the pressure and $\boldsymbol{\tau}$ is the extra-stress tensor defined as the sum of a Newtonian solvent and a polymeric solute contribution ($\boldsymbol{\tau} = \boldsymbol{\tau}_s + \boldsymbol{\tau}_p$). Since in this work we will restrict the numerical simulations to Newtonian fluid flows, the polymeric contribution to the extra-stress tensor is null ($\boldsymbol{\tau}_p = \mathbf{0}$) and the extra-stress tensor is calculated considering only the Newtonian solvent component:

$$\boldsymbol{\tau}_s = \eta_s (\nabla \mathbf{u} + \nabla \mathbf{u}^T), \quad (7.6)$$

In the numerical method, the governing equations ((7.4) - (7.6)) are integrated in time over a small time step (δt) and in space over the control volumes of the computational mesh. Regarding the accuracy of the calculations, the time derivative is discretized using an implicit first-order Euler scheme, the diffusive terms are discretized using second-order central differences and the CUBISTA high-resolution scheme (Alves *et al.*, 2003) is used for the discretization of the advective terms. More details of the numerical method used, can be found in Alves *et al.* (2003) and Oliveira *et al.* (2008).

7.4.2. Computational meshes and Dimensionless numbers

We performed numerical simulations of the Newtonian fluid flowing in the forward and backward flow directions of the hyperbolic shaped rectifiers. The meshes used to describe the computational domain are three-dimensional (3D), block-structured, and are composed of non-uniform cells. Due to symmetry of the Newtonian fluid flow under steady flow conditions, symmetry boundary conditions were imposed at the two center planes ($x = 0$ and $z = 0$), thus only a quarter of the total domain was used in the numerical simulations. The computational domain consists of a *long* straight channel used to describe the inlet and outlet channels and 10 hyperbolic elements which define the test section and have dimensions matching exactly those used in the experiments as described in Section 7.2.1. As explained previously in Section 7.2.1,

the fabricated microchannels present slight differences relative to the idealized shape mainly due to inaccuracies inherent to the fabrication procedure. In order to perform numerical simulations which are as meaningful as possible, the meshes used were adjusted to reflect such deviations as accurately as possible and the shapes described by Eqs. (7.1) and (7.2) were used. The outline of each mesh is shown in Figure 7.2 to highlight the deviations between the two devices corresponding to different depths and the idealized shape printed on the chrome mask. It is clear that the main differences are related to the difficulty in obtaining experimentally features with sharp corners, which in reality appear slightly rounded. Although, the differences are small, these can have a noticeable impact in quantitative terms.

We also note that the experimental geometries consist of 42 repeating hyperbolic units, while in the numerical simulations only 10 elements were used in order to reduce the size of the meshes and reduce the computational times. The numerical results confirm that for all flow conditions investigated numerically the flow field in the hyperbolic elements located in the middle of the computational domain is already fully-developed, and therefore using only 10 elements in the numerical simulations is enough to obtain accurate results, provided the comparisons are made in a normalized manner.

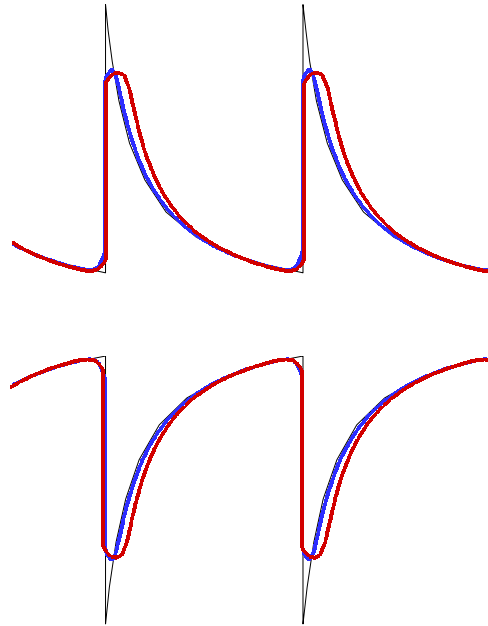


Figure 7.2. Comparison between the designed shape of the microchannel in the chrome mask (black line) and the real shapes of the final PDMS microchannels with 46 μm in depth (blue line) and 120 μm in depth (red line).

A zoomed view of the coarse mesh used for the simulations with channel A is illustrated in Figure 7.3. Moreover, the Newtonian fluid flow in channel A was simulated using two meshes with different refinements. For channel A mesh M1 has a total of 290 304 cells, while mesh M2

which is created by doubling the number of cells in each direction, has a total number of 2 322 432 cells. For channel *B* only one mesh (M1) was used, also consisting of 290 304 cells.

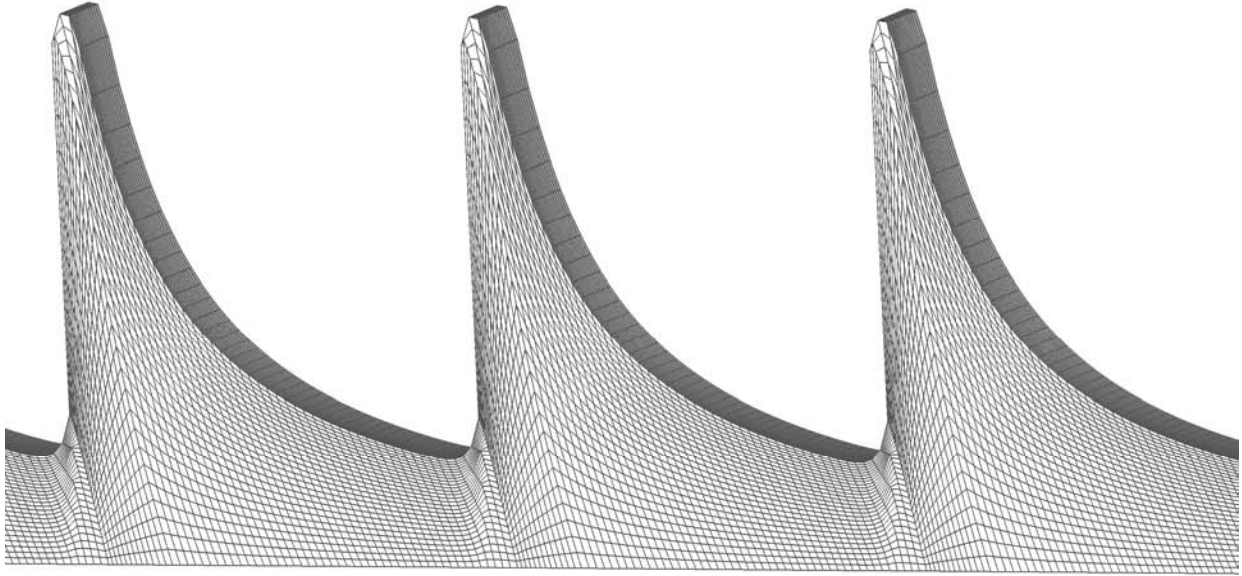


Figure 7.3. Zoomed view of the coarse mesh used in the numerical simulation of the Newtonian fluid flow in the forward direction.

Except for the (inlet/outlet) boundary conditions, the meshes used for the forward and backward flow directions are similar in their characteristics. The inlets and outlets were positioned far from the hyperbolic elements that define the test section allowing fully developed flow conditions to be attained well upstream and downstream of the hyperbolic elements. No-slip boundary conditions were imposed at the solid walls and at the inlet, a uniform velocity profile and null stress components were imposed. At the outflow boundary, we imposed a constant gradient of pressure and vanishing streamwise gradients of velocity and stress components.

Regarding the relevant dimensionless numbers, the aspect ratio of the channels is defined as $AR = h/D_2$. The Newtonian fluid flow is also characterized using the Reynolds number, based on the flow characteristics at the contraction throat (D_2) and defined as $Re = \rho U_2 D_2 / \eta_0$, where U_2 is the average velocity at the contraction throat, of width D_2 (cf. Figure 7.1), and η_0 is the viscosity. For the viscoelastic fluid the zero-shear rate viscosity is used in the definition of the Reynolds number.

For the characterization of the viscoelastic fluid flow we use the Deborah number (De), also based on the flow characteristics at the throat: $De = \lambda U_2 / (D_2/2)$.

The diodicity, which is used to characterize the rectification effects is defined as the ratio between the pressure drop in the forward direction, $\Delta P_{\text{Forward}}$, and the pressure drop in the backward direction, $\Delta P_{\text{Backward}}$, obtained for the same flow rate: $Di|_Q = \Delta P_{\text{Forward}} / \Delta P_{\text{Backward}}$.

7.5. Results and discussion

7.5.1. Flow patterns

The Newtonian fluid flow patterns observed in hyperbolic shaped rectifiers including rectifier A, were already discussed in detail in a previous work (Sousa *et al.*, 2010) (cf. Chapter 6) and are not presented here. For Newtonian fluid flow through rectifier B, the flow patterns observed are qualitatively similar to those in rectifier A and the diodicity is nearly unitary for the flow rates investigated (i.e. no rectification effects are observed). The flow phenomena observed in the PEO-8M fluid flow through the hyperbolic shaped microfluidic rectifiers A and B is qualitatively similar, as illustrated in Figure 7.4 and 7.5.

At low flow rates, Newtonian-like flow patterns are observed, with recirculations appearing inside the hyperbolic corner, for both flow directions. Increasing the flow rate, a distinct flow behavior is found for the two flow directions, as documented previously in Sousa *et al.* (2010) (cf. Chapter 6). For the forward direction, the flow becomes asymmetric and unsteady due to onset of elastic instabilities, and recirculations appear and disappear randomly within the hyperbolic elements. In our previous work with the 46 μm microfluidic rectifier (Sousa *et al.*, 2010) (cf. Chapter 6), we analyzed the onset of these purely elastic instabilities and identified a connection between these and the appearance of rectification effects. In that work, a nominal depth of 50 μm was assumed, however, subsequent SEM measurements of the microchannels showed that the correct depth is 46 μm , as correctly used in the present work. Also, in Sousa *et al.* (2010) (cf. Chapter 6) the throat width of the chrome mask was assumed in the calculations ($D_2 = 54 \mu\text{m}$) while in this work we use the actual minimum width of the final PDMS channel, which is $D_2 = 62.7 \mu\text{m}$ for the microchannel with $h = 46 \mu\text{m}$. These corrections in the microchannel depth and width leads to an increase of less than 10 % in the Re and a decrease of less than 20 % in the De values presented in the previous work.

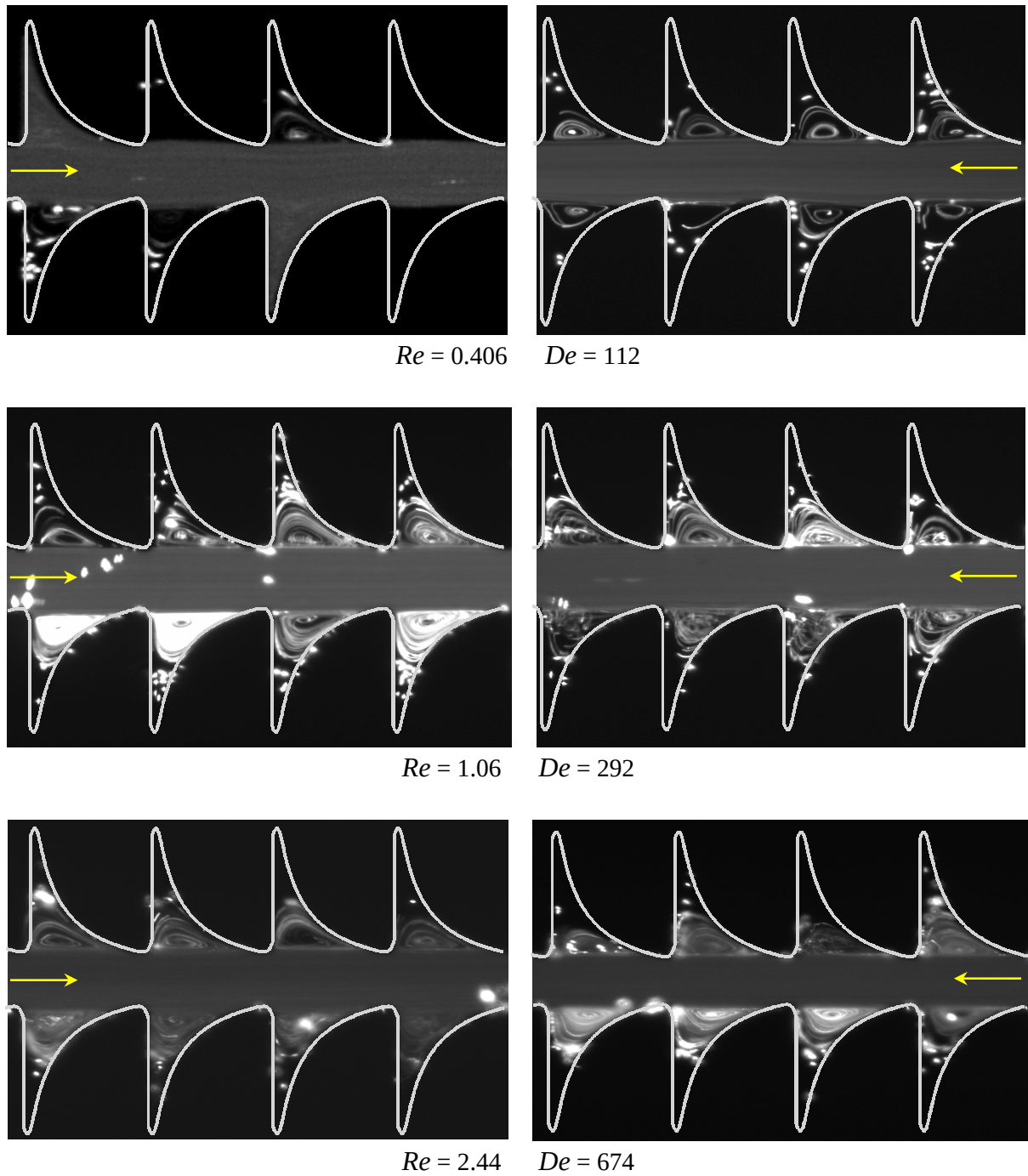


Figure 7.4. Streak line images of the viscoelastic fluid flow in the microfluidic rectifier with $h = 46 \mu\text{m}$ for the forward (left column) and backward (right column) directions at $T = 293.5 \text{ K}$.

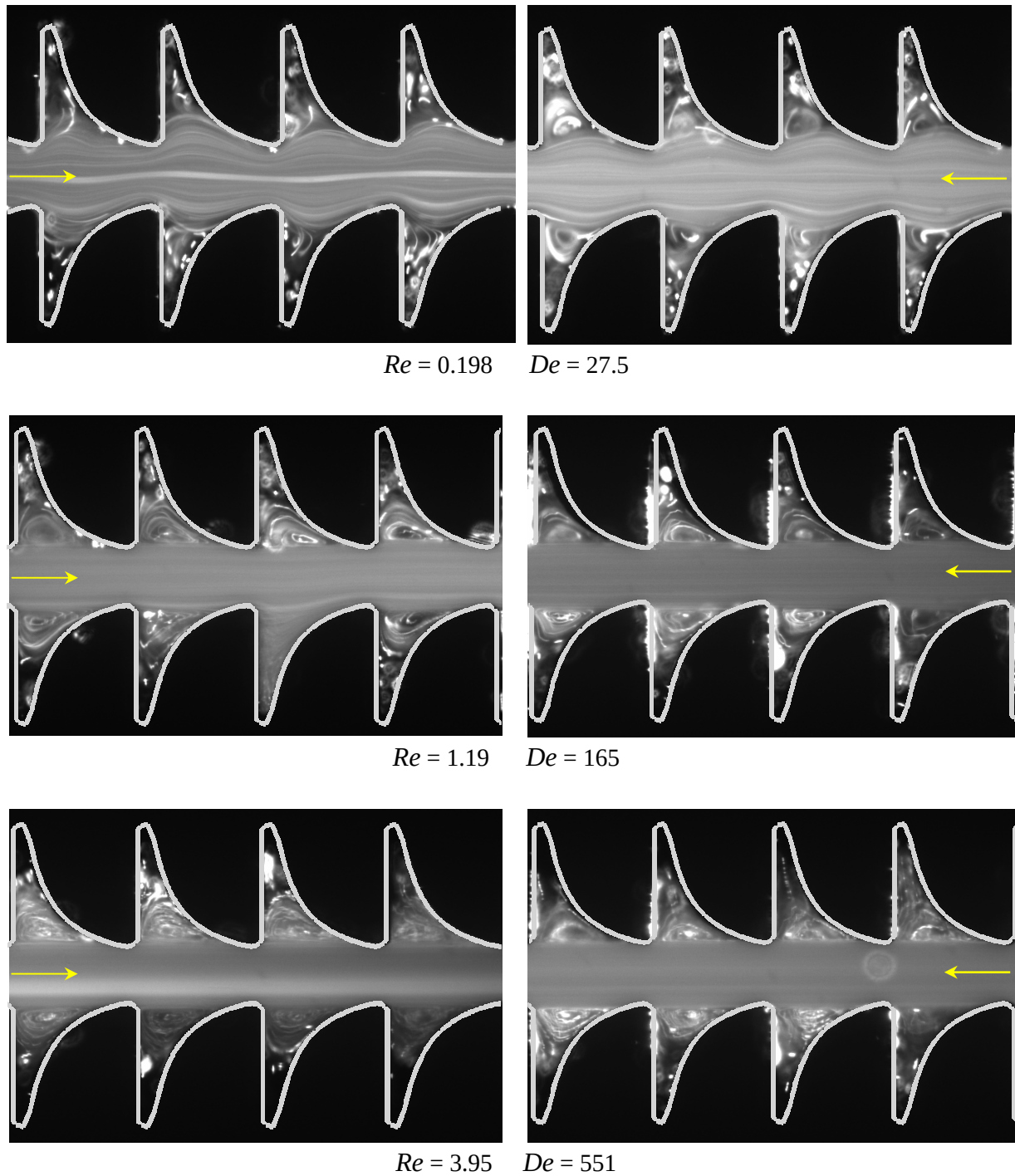


Figure 7.5. Streak line images of the viscoelastic fluid flow in the microfluidic rectifier with $120\ \mu\text{m}$ in depth for the forward (left column) and backward (right column) directions at $T = 300.6\ \text{K}$.

For the deeper microchannel ($h = 120 \text{ }\mu\text{m}$), rectification effects also emerge following the appearance of elastic instabilities (Figure 7.5) as will be explained in Section 7.5.3. For the backward flow direction, recirculations also appear near the hyperbolic corners. The flow remains steady as the flow rate (or De) is increased until a critical De is reached. The critical value for the appearance of elastic instabilities that lead to unsteady flow is significantly higher than for forward flow.

In summary, similar flow behavior is found for the two micro-rectifiers (A and B) but the onset of elastic instabilities occurs at significantly different Deborah numbers, with the critical De increasing with aspect ratio: while for the microchannel with $h = 46 \text{ }\mu\text{m}$ elastic instabilities are already evident at $De \approx 20$, for the deeper microchannel, similar flow patterns are found only at much higher Deborah numbers, $De \approx 90$.

7.5.2. Velocity field

7.5.2.1. Newtonian fluid flow – Experimental validation

The velocity field of the Newtonian fluid flow in both hyperbolic rectifiers was determined using μPIV . Figure 7.6 shows the dimensionless axial velocity profiles obtained at the centerline of the two microfluidic rectifiers for both flow directions. For microchannel with a depth of $46 \text{ }\mu\text{m}$, we compare the experimental results with numerical predictions using meshes M1 and M2 (Figure 7.6(a)), whereas for microchannel with $120 \text{ }\mu\text{m}$ of depth, we only compare the experimental and numerical results using only the coarse mesh M1 (Figure 7.6(b)).

As expected, the velocity profiles along the centerline present a periodic behavior caused by the geometrical shape of the microchannel, i.e. the sequence of the hyperbolic elements composed of contractions and expansions leads to a quasi-sinusoidal velocity profile. As such, the axial velocity varies from a minimum value at a position near the center of each element, to a maximum value near the throat region. At low flow rates, inertia is negligible and the axial velocity profiles for both flow directions are similar due to the reversibility of Newtonian creeping flows. When the flow rate is further increased, differences between the velocity profiles measured for the forward and backward flow directions become noticeable due to inertial effects.

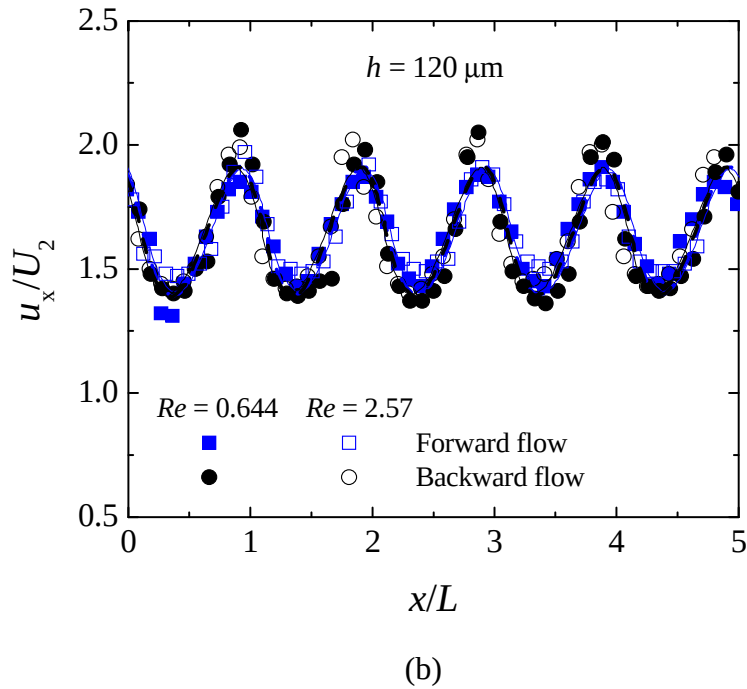
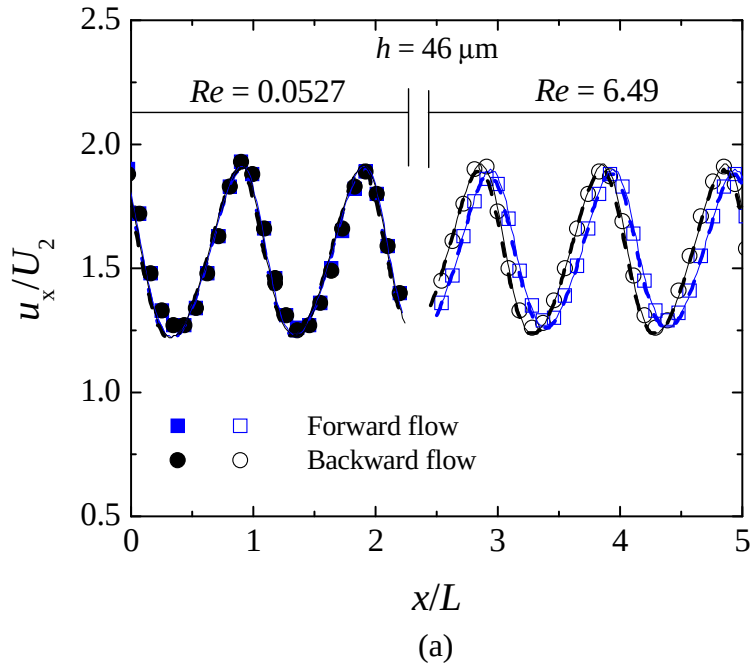


Figure 7.6. Dimensionless axial velocity profiles for Newtonian fluid flow along the centerline of the hyperbolic rectifier with (a) $h = 46 \mu\text{m}$ ($T = 296.1 \text{ K}$, for $Re = 0.527$ and $T = 295.5 \text{ K}$ for $Re = 6.49$) and (b) $h = 120 \mu\text{m}$ ($T = 298.0 \text{ K}$, for $Re = 0.644$ and $T = 297.8 \text{ K}$ for $Re = 2.57$). The symbols represent the experimental data and the lines represent the numerical predictions. In (a) the experimental results are compared with those obtained numerically using mesh M1 (solid thin lines) and mesh M2 (thick dashed lines). In (b), the thick dashed lines and the solid thin lines represent the numerical predictions for $Re = 0.646$ and $Re = 2.57$, respectively using only mesh M1.

Comparing the axial velocity profiles at the higher Re , for the two flow directions, it is possible to identify a lag between both profiles. For the forward flow direction inertia pushes the fluid against the hyperbolic (gradual) contraction and the value of the velocity is maximum at the throat. On the other hand, for the backward flow direction, the fluid experiences a sudden acceleration when flowing through the abrupt contraction and the maximum value of the axial velocity occurs immediately downstream of the throat. Moreover, it is possible to observe that the amplitude of the velocity oscillation (and consequently the maximum strain rate) in the backward direction is slightly higher than in the forward flow direction. The numerical results presented in Figure 7.6 were determined at the center plane of the computational domain. However, since the μ PIV measurements take into account not only the focused particles at the center plane but also unfocused particles at adjacent planes over the whole measurement depth (Meinhart *et al.*, 2000), as described in Section 7.2.1 a numerical weighted-average velocity in the measurement depth, δz_m , was calculated and a deviation from the centerline velocity field of less than 3% was found for rectifier A. For rectifier B, due to the larger depth, this deviation is significantly lower. Since the imparted difference in velocity is small and considering that the focused particles are responsible for the major contribution to the μ PIV correlation, we only present the numerical axial velocity taken only at the centerline to compare with the experimental results. As can be seen in Figure 7.6, the results obtained experimentally are in good agreement with the numerical predictions, thus validating the experimental procedure. Moreover, the predictions obtained using meshes M1 and M2 are very similar and deviations observed are smaller than 1%. For this reason, only mesh M1 will be used this point forwards to allow for smaller CPU computational time required to converge the three-dimensional simulations.

7.5.2.2. Viscoelastic fluid flow

In agreement with the findings of Sousa *et al.* (2010) (cf. Chapter 6), at low flow rates the flow is steady, but above a critical flow rate, elastic instabilities arise and the flow becomes unsteady, initially for the forward direction and then, at significantly higher flow rates, also in the backward direction.

a) Steady flow

In Figure 7.7 we show the axial velocity profiles obtained for the viscoelastic fluid flow in both microgeometries. The flow rates presented correspond to steady flow conditions corresponding to identical flow resistance between both flow directions. Moreover, in Figure 7.7 we show the

numerical predictions for a Newtonian fluid flowing at the same flow rate as the viscoelastic fluid.

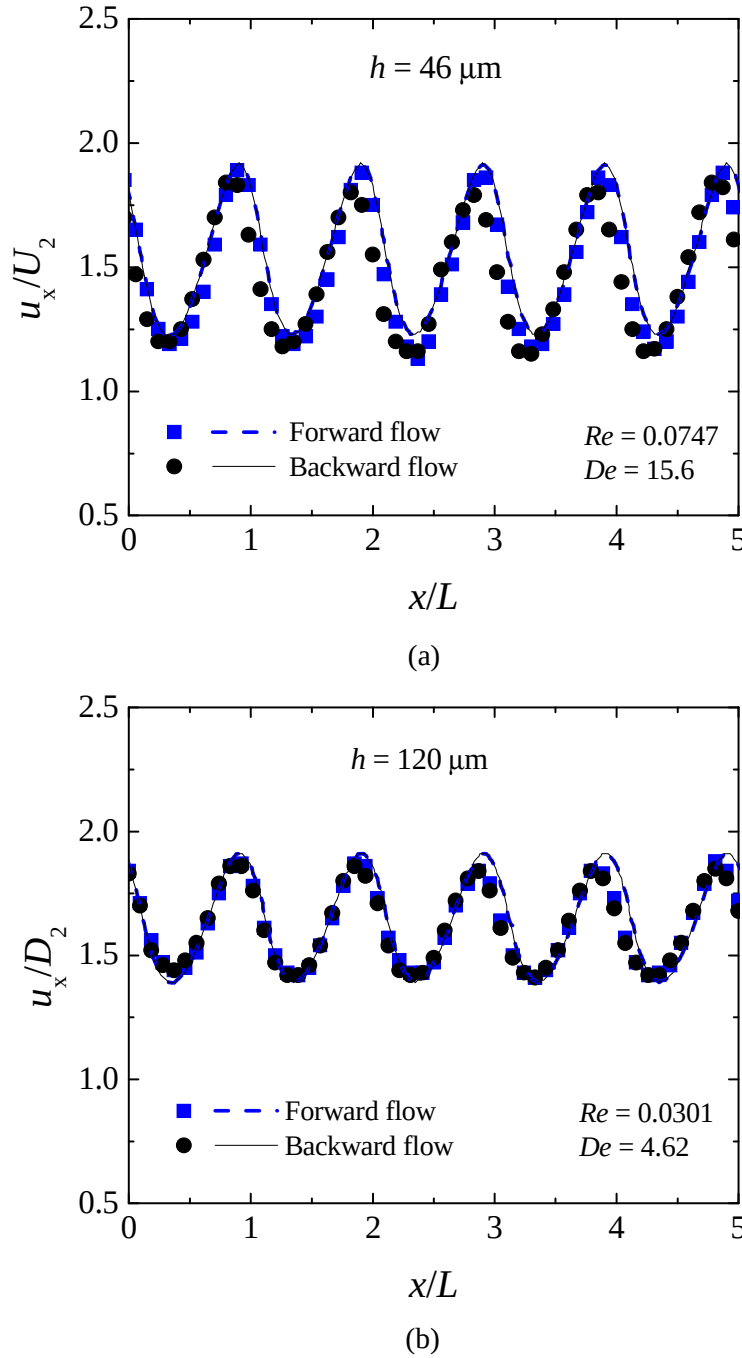


Figure 7.7. Dimensionless axial velocity profiles taken along the centerline of the hyperbolic rectifier with (a) $h = 46 \mu\text{m}$ ($T = 297.6 \text{ K}$) and (b) $h = 120 \mu\text{m}$ ($T = 299.1 \text{ K}$) for steady fluid flow. The symbols represent experimental data obtained for the viscoelastic fluid and the dashed lines represent the numerical predictions for a Newtonian fluid at the same flow conditions (Re) as the viscoelastic fluid.

The experimental results obtained for steady flow conditions, in which elastic effects are still small, present similarities with those predicted numerically for the Newtonian fluid flow at the same flow conditions, but already a lag is apparent between the experimental (viscoelastic) and numerical (Newtonian) data. The axial velocity profile exhibits the expected sinusoidal-like shape, with the profile for the forward direction showing a slightly higher amplitude. Moreover, there is also a second small lag between the forward and backward velocity, mostly due to elastic effects.

b) Unsteady flow

The velocity field was also measured using μ PIV at flow conditions for which the behavior in the two flow directions is markedly different i.e., when rectification effects exist. Two flow rates were investigated, which correspond to the following scenarios: (i) unsteady flow in the forward direction and steady flow conditions in the backward direction; (ii) unsteady flow in both forward and backward directions, corresponding to a higher flow rate.

When the flow becomes unsteady, due to elastic instabilities, the vortices appear and disappear along time in some elements of the rectifier and in this case instantaneous velocity measurements are more suitable to analyze the flow behavior. However, time-averaged measurements are also useful to establish the overall time-average flow field. In μ PIV measurements, the particle image density is generally low and high background noise is commonly observed, which means that using a single pair of images is generally not sufficient (Wereley, 2002), even under steady conditions, to obtain accurate velocity measurements. Therefore, a correlation function based on averaging a *higher* number of image pairs is usually used in order to improve the accuracy of the μ PIV measurements (Meinhart et al, 2000).

In Figure 7.8, we compare one instantaneous velocity profile (one image pair) with the average velocity profiles using 10, 150 and 500 pairs of images for a flow rate of $Q = 0.25$ ml/h, which corresponds to the first scenario described above (i.e. steady backward flow and unsteady forward flow). The unsteady nature of the forward flow is visible in the instantaneous velocity profile in Figure 7.8, obtained at a given instant in time, since a clearly cyclic velocity profile is not observed.

We note that the instantaneous velocity profile at different times is clearly different, due to the unsteady nature of the flow. On the other hand, the backward flow for the same flow rate is steady as can be attested from the instantaneous velocity profile. In this case, the velocity profile

determined from one pair of images acquired at any instant is similar to the average velocity obtained by averaging 150 pairs of images, with deviations smaller than 10%.

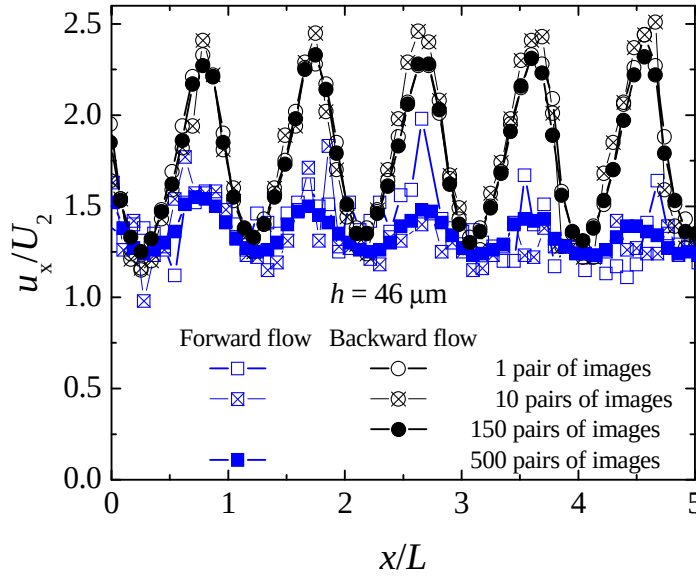


Figure 7.8. Average and instantaneous dimensionless axial velocity profiles at the centerline of the hyperbolic rectifier with $h = 46 \mu\text{m}$, for $Re = 0.244$ and $De = 46.7$ (case i) at $T = 298.9 \text{ K}$.

In the steady case, if the axial velocity profile is obtained from 10 pairs of images, the deviation decreases to less than 3% when compared to that obtained from 150 pairs of images. In the forward flow direction, the results obtained with 10 image pairs still present a disorderly pattern and we have used 500 images pairs when calculating the time-averaged measurements for establishing the time-averaged flow field along the centerline. It is also interesting to note that the dimensionless velocity profile shown in Figure 7.8 for the backward flow direction shows a significant overshoot in the throat region ($u_x/U_2 \approx 2.3$ as compared with $u_x/U_2 \approx 1.9$ for lower De) and a moderate velocity undershoot. This behavior is typical of highly elastic contraction and expansion flows, as discussed by Alves and Poole (2007).

For higher flow rates, when the flow becomes unsteady in both directions (case ii), a high number of images pairs were acquired for the characterization of the average velocity field. A total number of 500 and 1000 images pairs were acquired for the microfluidic rectifiers with the lower and higher depths, respectively.

In Figure 7.9 we show the average axial velocity profiles obtained at the centerline of the microchannel with $h = 46 \mu\text{m}$ (Figure 7.9 (a)) and $h = 120 \mu\text{m}$ (Figure 7.9 (b)) in order to establish the overall evolution of the flow field and compare low and high De data. For the lower

De presented in Figure 7.9(a) and 7.9(b), despite the unsteady flow behavior of the viscoelastic fluid in the forward direction, the flow is steady in the backward direction. In this case, one can see the differences in the amplitude of oscillation between the velocity profiles for both flow directions.

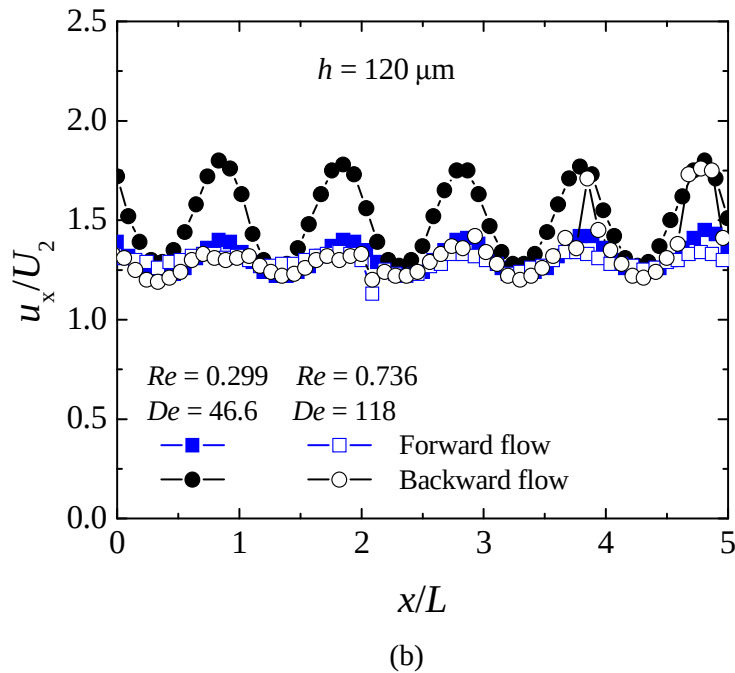
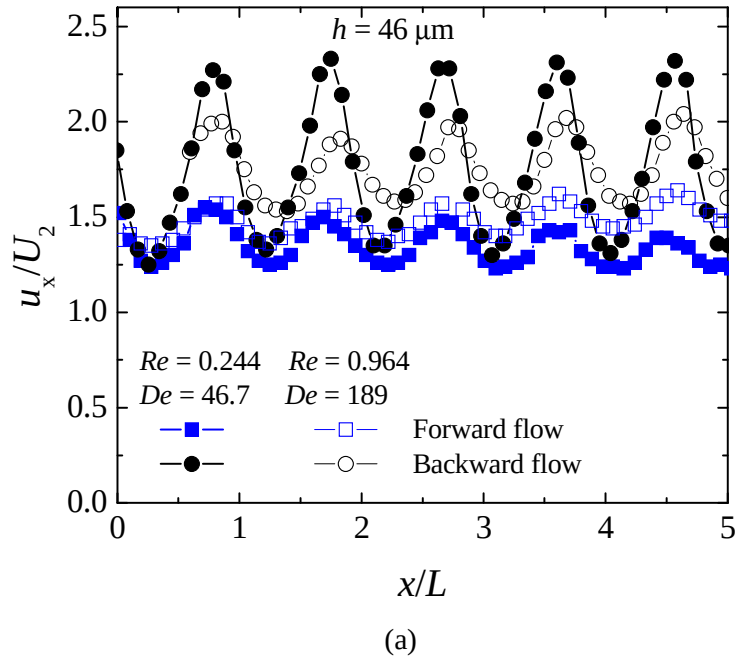


Figure 7.9. Dimensionless axial velocity profiles taken along the centerline of the hyperbolic rectifier with (a) $h = 46 \mu\text{m}$ ($T = 298.9 \text{ K}$, for $De = 46.7$ and $T = 298.5 \text{ K}$ for $De = 189$) and (b) $h = 120 \mu\text{m}$ ($T = 298.8 \text{ K}$, for $De = 46.6$ and $T = 298.4 \text{ K}$ for $De = 118$). At the lower De , the flow is unsteady in the forward flow direction (filled squares) and steady in the backward flow direction (filled circles). At the higher De , the flow is unsteady in both flow directions (open symbols).

For the backward direction, in which the flow remains steady, the velocity gradient at the centerline is higher than for the forward direction. Since we are averaging the pairs of images acquired during a long period of time, in which the flow behavior was varying due to the elastic instabilities, the velocity fluctuations are smoothed out. At the higher De (cf. Figure 7.9 (a) and 7.9(b)), when the flow is unsteady in the forward and backward directions, the amplitude of the velocity is lower than for lower elasticity flows and the differences between oscillating time-average forward and backward profiles are still visible, but smaller than at the lower De .

7.5.3. Wall effect on the diodicity

In Figure 7.10(a) we show the pressure drop measured experimentally as well as the numerical predictions for the Newtonian fluid flow through the hyperbolic rectifier with different depths and in Figure 7.10(b) we show the pressure drop measured experimentally for the viscoelastic fluid. For both cases, the results obtained for the two flow directions are presented allowing the determination of the diodicity corresponding to each microfluidic rectifier.

As expected, the pressure drop observed in the microchannel with a smaller depth is significantly higher than that found for the deeper microchannel. For the Newtonian fluid, the pressure drop increases quasi-linearly with the flow rate. Furthermore, in agreement with the findings of Sousa *et al.* (2010) (cf. Chapter 6), negligible rectifications effects are observed experimentally, i.e. for the same value of the flow rate, the corresponding pressure drop obtained for the forward and backward flow directions are very similar, within experimental uncertainty. A good agreement between experimental and numerical results confirms that the characteristics of the experimental microgeometries are well captured by the numerical meshes. At the highest flow rates, small deviations (smaller than 10%) are observed between the experimental data and numerical predictions, but this is primarily due to the progressive enlargement/deformation of the cross section area due to the high pressures required to obtain the higher flow rates (above 1.5 bar) and the corresponding deformability of PDMS.

For the non-Newtonian fluid flow, despite the considerably lower flow resistance measured in the deeper channel, the overall behavior is independent of the channel depth. At low flow rates, the pressure drop increases linearly with the flow rate indicating the reversibility of the quasi-Newtonian flow behavior. Increasing the flow rate (or De) beyond a critical value, the flow resistance becomes considerably different for the two possible flow directions, which is directly related with the onset of elastic instabilities (Groisman and Quake, 2004; Sousa *et al.*, 2010).

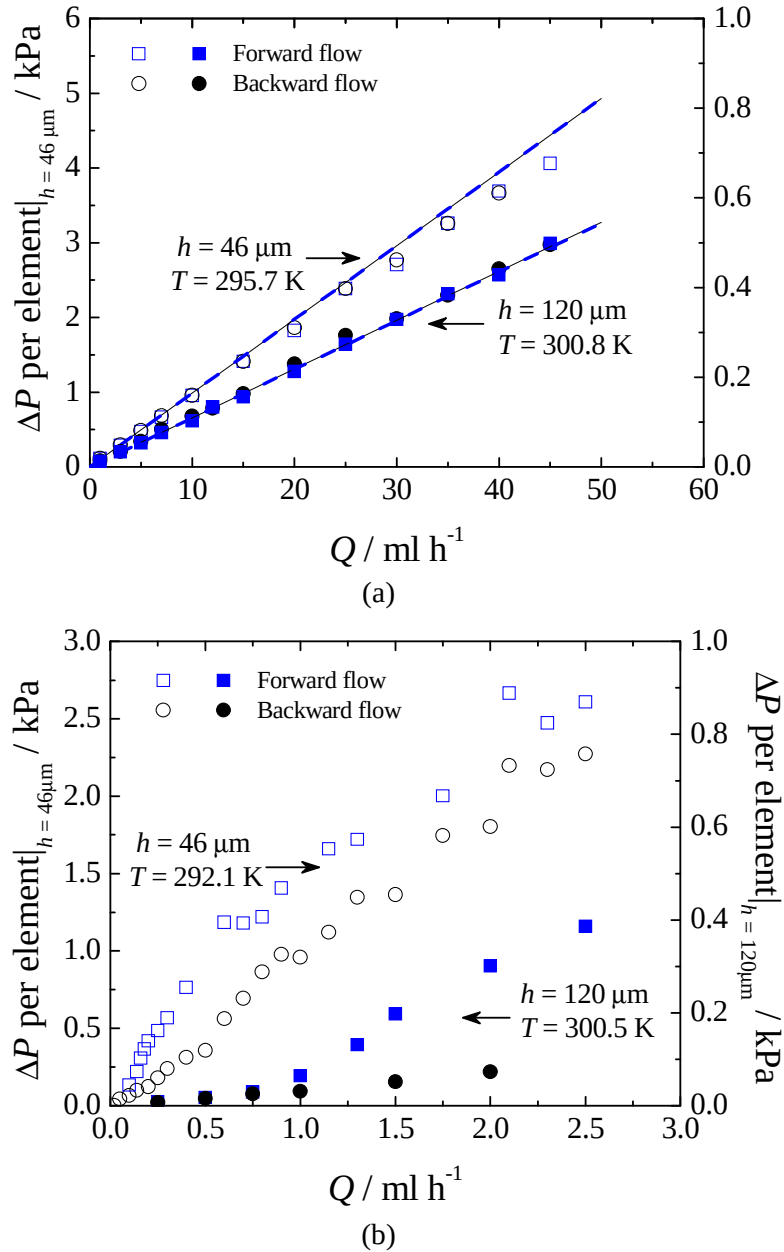


Figure 7.10. Pressure drop measured in the hyperbolic rectifier with different depths, as a function of the flow rate for the Newtonian and the viscoelastic fluids (b). For the Newtonian fluid, the experimental results (symbols) are compared with numerical predictions (solid lines).

In the forward flow direction, above the critical flow conditions, the flow resistance increases first abruptly and then moderately as the flow rate increases. On the other hand, in the backward flow direction, the more pronounced increase of the flow resistance with the flow rate occurs only at significantly higher flow rates. Since elastic instabilities in the backward flow direction appear also at these higher flow rates, it is possible to infer, once again, that the changes in flow dynamics triggered by the onset of elastic instabilities lead to a markedly different flow resistance behavior for both flow directions.

In order to analyze the effect of the channel walls on the diodicity, in Figure 7.11 we plot the diodicity, defined in terms of the pressure drop ratio for a constant flow rate, as a function of the De for the two channel depths. The maximum diodicity observed depends significantly on the aspect ratio of the channel. For the microchannel with lower aspect ratio ($AR = 0.73$; $h = 46 \mu\text{m}$), the maximum diodicity obtained is about 3.5, while for the channel with larger aspect ratio ($AR = 1.7$; $h = 120 \mu\text{m}$) the maximum diodicity is about 6.5, and occurs at a higher De . Thus, higher aspect ratios lead to higher rectification effects, indicating that deeper geometries, where wall effects (or shear effects) are less pronounced and extensional flow is enhanced, may be preferred in order to devise efficient microfluidic diodes. Simultaneously, higher aspect ratios delay the onset of rectification effects (or of elastic instabilities) to higher Deborah numbers and even higher flow rates. It would be desirable to further increase the depth of the microfluidic diode, to investigate if higher diodicity would be achieved. However, using PDMS only moderate aspect ratio geometries can be replicated, and other fabrication techniques need to be used to allow the precise fabrication of deeper microchannels.

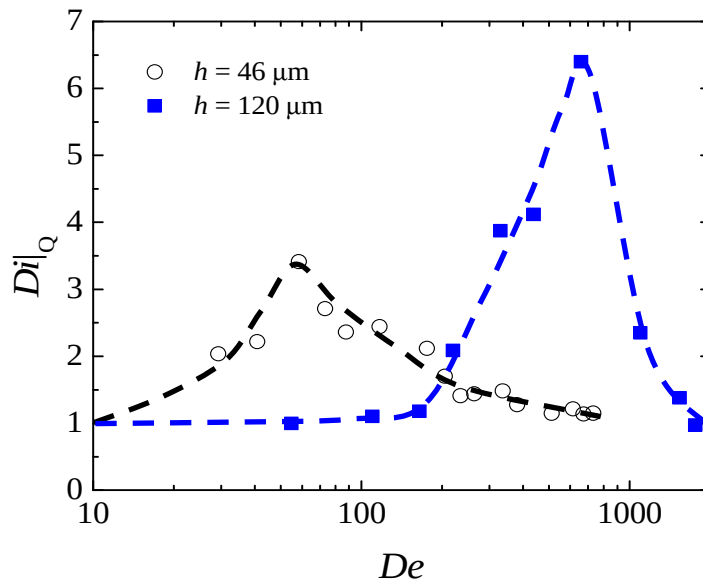


Figure 7.11. Diodicity as a function of Deborah number for the hyperbolic shaped rectifier with different depths.

7.6. Conclusions

The flow of a Newtonian fluid (de-ionized water) and a non-Newtonian fluid with viscoelastic behavior was investigated in microfluidic rectifiers with hyperbolic shape. Microchannels with two different depths were employed in order to assess the influence of bounding walls on the diodicity. Moreover, the Newtonian fluid flow was simulated numerically and a good agreement

between experimental and numerical results was found. The fluid flow behavior is qualitatively similar in the microgeometries with different depths. For the Newtonian fluid, the axial velocity at the centerline shows a periodic pattern due to geometric effects. At low inertial flow conditions, the velocity profile in the forward flow direction resembles that in the backward flow direction, while increasing the flow inertia leads to the appearance of an offset between both velocity profiles. For the viscoelastic fluid, different flow characteristics were found. At low De , the velocity profiles are Newtonian-like, but increasing the flow rate, due to the onset of elastic instabilities, the flow becomes unsteady and the axial velocity along the centerline of the microchannel varies in time.

The pressure drop within the hyperbolic rectifier shows a similar behavior for the two rectifiers. For the Newtonian fluid, the experimental results do not reveal noticeable rectification effects. For the viscoelastic fluid, an anisotropic flow resistance is observed independently on the aspect ratio of the microchannel. In terms of diodicity, the microrectifier with the higher aspect ratio presented a maximum diodicity of about 7, which is clearly in excess of others presented in the literature (Groisman and Quake, 2004; Nguyen *et al.*, 2008). Our results lead us to conclude that, to some extent, reduction of shear and enhancement of the extensional component of the flow field (achieved by decreasing the effect of bounding walls) leads to higher diodicities.

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EXTENSIONAL FLOW OF BLOOD ANALOGUE SOLUTIONS IN MICROFLUIDIC DEVICES

Abstract

In this study, we show the importance of extensional rheology, in addition to the shear rheology, in the choice of blood analogue solutions intended to be used in vitro for mimicking the microcirculatory system. For this purpose, we compare the flow of a Newtonian fluid and two well established viscoelastic blood analogue polymer solutions through microfluidic channels with a hyperbolic contraction followed by an abrupt expansion. The hyperbolic shape was selected in order to impose a nearly constant strain rate at the centerline of the microchannels and achieve a quasi-homogeneous and strong extensional flow, often found in features of the human microcirculatory system such as stenoses. The two blood analogue fluids used are aqueous solutions of a polyacrylamide (PAA, 125 ppm w/w) and of a xanthan gum (500 ppm w/w), which were characterized rheologically in steady-shear using a rotational rheometer and in extension using a capillary breakup extensional rheometer (CaBER). Both blood analogues exhibit a shear-thinning behavior similar to that of whole human blood, but their relaxation times, obtained from CaBER experiments, are substantially different (by one order of magnitude). Visualizations of the flow patterns using streak line photography, measurements of the velocity field using micro-particle image velocimetry and pressure-drop measurements were carried out experimentally for a wide range of flow rates. The experimental results were also compared with the numerical simulations of the flow of a Newtonian fluid with a constant viscosity and a generalized Newtonian fluid with shear-thinning behavior. Our results show that the flow patterns of the two blood analogue solutions are considerably different, despite their similar shear rheology. Furthermore, we demonstrate that the elastic properties of the fluid have a major impact on the flow characteristics, with the polyacrylamide solution exhibiting a much stronger elastic character.

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8.1. Introduction

The human blood is a multiphase complex fluid that carries vital substances to cell tissues and organs of the body. It consists of a suspension of cellular elements in an aqueous matrix, the plasma, of which 99% are erythrocytes (red blood cells, RBCs), less than 1% are leukocytes (white blood cells, WBCs), and the remainder are platelets, proteins and other solutes (Picart *et al.*, 1998). Blood viscosity depends on blood composition (e.g. hematocrit, Brookshier and Tarbell, 1991), temperature, shear rate, vessel diameter (e.g. Fåhræus- Lindqvist effect, Popel and Johnson, 2005), cell aggregation level, shape, deformation and orientation, in addition to plasma viscosity (Ylmaz and Gundogdu, 2008), and is an important parameter since it provides valuable information about human health. Even though plasma can be considered as a Newtonian fluid, with a shear viscosity of about 1.1 mPa s at 37 °C (Goldsmith and Turitto, 1986), whole blood is a non-homogeneous complex fluid, which exhibits non-Newtonian properties (Owens, 2006; Moyers-Gonzalez *et al.*, 2008a,b; Moyers-Gonzalez and Owens, 2008).

Early dynamic rheological measurements that showed a non-Newtonian behavior of human blood were performed by Thurston (1971, 1972) and by Lessner *et al.* (1971), using a cylindrical tube and a Couette geometry, respectively. Such non-Newtonian properties, like viscoelasticity, thixotropy and shear-thinning behavior, arise mainly as a consequence of the deformability of RBCs, as well as their tendency to interact with each other forming aggregates (Vlastos *et al.*, 1997; Owens, 2006). Indeed, at low shear rates, in a process that depends on the concentration of plasma proteins, red blood cells may form aggregate structures (rouleaux), causing an increase in blood viscosity (Thurston and Henderson, 2007). On the other extreme, when the deformable RBCs are flowing at high velocities (or high shear rates), these structures are broken and the individual RBCs tend to be elongated, leading to a decrease in the shear viscosity (Chien, 1970). In addition, rheological measurements show that blood exhibits a yield stress (Picart *et al.*, 1997). This property is notoriously difficult to quantify since it requires precise measurements at very low shear rates, and as a consequence there is a wide variability in reported values (Picart *et al.*, 1997). Nevertheless, the reported values for blood yield stress are very small, ranging from about 0.005 to 0.01 N/m² depending on the hematocrit – at high RBC concentration, the rouleaux structures are more cohesive and therefore the yield stress tends to be higher.

Despite the extensive evidence for the complex rheology of blood (Owens, 2006; Waite and Fine, 2007) the vast majority of research on hemodynamics assumes that blood behaves as a Newtonian fluid. When flowing in large vessels, such as arteries (diameter ~ 4 mm, Caro *et al.*, 1974), this assumption is usually reasonable. However, even in large vessels, non-Newtonian effects may become important due to the pulsatile (i.e., time-dependent) nature of the blood flow. More importantly, in small vessels typical of the microcirculation system, e.g. arterioles (diameter ~ 50 μm) and capillaries (diameter ~ 6 μm) (Caro *et al.*, 1974), where the characteristic times of the flow and the fluid become comparable, blood behavior is highly viscoelastic (Owens, 2006). Furthermore, non-Newtonian effects can be enhanced when the flow vessel presents geometric features such as contractions/expansions, taper or bifurcations. Two important physiological examples are stenoses, an abnormal narrowing in the blood vessel due to the accumulation of cells, usually fatty material, and aneurysms, which are local abnormal dilations of the blood vessels. An interesting study about the effect of the geometry on the hemodynamic variables is reported in Tovar-Lopez *et al.* (2010), in which blood itself is used and it is considered that it exhibits a Newtonian behavior. The authors studied platelets aggregation dynamics in blood flow through different contractions/expansions representing diseased blood vessels with different stenosis levels, in order to generate strain rate conditions similar to those in human vessels. Their flows are always characterized by Reynolds numbers on the order of hundreds.

Different constitutive equations, relating the stresses with the strain history, have been proposed and used to describe the rheological properties of human blood. To illustrate and model the inelastic shear-thinning behavior of the shear viscosity, equations such as the power-law (Neofytou and Tsangaris, 2006, Gray *et al.*, 2007), the Casson (Fung, 1993) and the Carreau model (Lee and Steinman, 2007) are among the most widely used in the context of generalized Newtonian constitutive equations. Nevertheless, other time-independent as well as time-dependent viscosity models have been used (e.g. Yilmaz and Gundogdu, 2008), but to a lesser extent.

A comprehensive characterization of blood rheology and its flow dynamics is indeed very important in order to predict cardiovascular diseases, to plan vascular surgeries, understand the transport of drugs through the circulatory system and for the development of cardiovascular equipments as for example blood pumps, heart valves valves and stents (Yilmaz and Gundogdu, 2008). However, the manipulation of whole blood is not a straightforward task and may not

always be practical, primarily due to safety reasons. As such, blood analogue solutions are widely used for *in vitro* experiments as they exhibit several advantageous characteristics such as non-toxicity, low cost and transparency (Thurston, 1996). These fluids present rheological characteristics similar to human blood and are typically based on polymer solutions. The earliest works that used blood analogues considered a suspension of 1 μm polystyrene spheres in water, Dextran 70 and calcium chloride to stimulate the aggregation process (Fukada *et al.*, 1989) and a suspension of disc-shaped, biconcave particles in Dextran 70 (Liepsch *et al.*, 1991). In addition, other analogues have been used, namely aqueous solutions of a polyacrylamide, PAA, and a xanthan gum, XG (Liepsch *et al.*, 1991; Thurston, 1996; Vlastos *et al.*, 1997; Gray *et al.*, 2007) in which the addition of glycerine was used to tune the blood rheology at different hematocrit levels (Brookshier and Tarbell, 1991, 1993). Gray *et al.* (2007) studied the flow through a simplified arterial junction corresponding to a distal anastomosis in a femorodistal bypass using a Newtonian and two different non-Newtonian xanthan gum based blood analogues (with and without glycerine). In their experiments, the authors considered a time-dependent flow to reproduce the pulsation of the cardiac cycle and showed that the wall shear stresses obtained for the Newtonian fluid flow are very different from those obtained when non-Newtonian fluids with a shear-thinning behavior are used. Lerche *et al.* (1993) and Vlastos *et al.* (1994) tested different concentrations at which PAA and XG solutions are blood-like and subsequently, Vlastos *et al.* (1997) performed a combination of steady and oscillatory shear tests and found a similar rheological behavior between the analogues and human blood, at concentrations of 125 ppm (w/w) for the PAA and 500 ppm (w/w) for XG, particularly at low shear rates while at high shear rates they tend to exhibit higher viscosity and elasticity than blood (Vlastos *et al.*, 1997).

Since blood rheology is extremely complex, it is difficult to develop analogue fluids that yield a complete description of all the properties of blood and these fluids are typically chosen based on their density and shear viscosity. In this work, we aim to show that elastic characteristics of the fluids (such as the relaxation time) should also be taken into account in particular when considering conditions equivalent to those found in microcirculation. At these small scales, the role of fluid elasticity is significantly enhanced beyond what can be achieved at the macroscale without being overwhelmed by inertial effects. For this purpose, we examine the flow of a Newtonian fluid and two well established viscoelastic blood analogue solutions through microfluidic channels containing contraction/expansion features in order to impose a highly extensional flow often found in features of the human circulatory system such as stenoses. The blood analogue fluids used are an aqueous solution of a polyacrylamide at a concentration of

125 ppm (w/w) and an aqueous solution of a xanthan gum at a concentration of 500 ppm (w/w) as proposed by Vlastos *et al.* (1997) and Thurston (1996). These fluids were characterized rheologically in steady-shear flow using a rotational rheometer and in extension using a capillary breakup extensional rheometer (CaBER). We will show that despite having very similar shear-rheology, the elastic character of the two solutions is substantially different, which will have a significant impact in flows at the microscale.

8.2. Experimental Techniques

8.2.1. Microchannels fabrication and geometry

The flow of the blood analogue solutions was investigated in microchannels with dimensions comparable to small human vessels, which include in their design contractions and expansions with hyperbolic and abrupt shapes. The hyperbolic shape was chosen in order to provide a nearly constant strain rate of the fluid flow along the centerline of the microgeometry, as proposed by Oliveira *et al.* (2007). The combination of geometry and reduced lengthscale characteristic of microfluidics, make it possible to study the response of the fluid under strong accelerations, in the absence of significant inertial effects, for controlled extensional flow conditions. The dimensions of the narrow gap of the microgeometries are typical of small vessels, such as arterioles (internal diameter $\sim 50 \mu\text{m}$), venules (internal diameter $\sim 40 \mu\text{m}$) and capillaries (internal diameter $\sim 6 \mu\text{m}$) (Caro *et al.*, 1974).

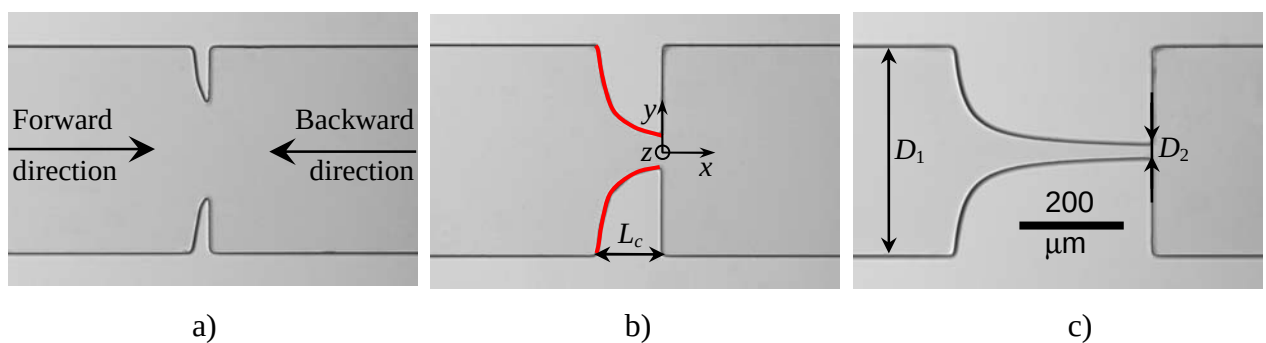


Figure 8.1. Micrograph of the microgeometries studied: (a) microchannel S_1 ; (b) microchannel S_2 ; (c) microchannel S_3 . In (b) the shape of the quasi-hyperbolic walls are marked in red.

The planar microgeometries were fabricated in polydimethylsiloxane, PDMS (Sylgard 184, Dow Corning), from an SU-8 reusable mold, using standard soft-lithography techniques (Xia and

Whitesides, 1998; Friend and Yeo, 2010). Three different configurations were used as shown in the microscopy images of Figure 8.1.

Independently of the configuration, the inlet/outlet width, D_1 of the microchannels were kept constant and equal to 400 μm . The minimum width of the contraction, D_2 , and the hyperbolic contraction length, L_c were varied in order to obtain different values of the total Hencky strain (ε_H). The total Hencky strain experienced by a fluid element moving along the centerline of a planar contraction, is given by $\varepsilon_H = \int_0^t \dot{\varepsilon} dt = \ln\left(\frac{D_1}{D_2}\right)$ (Rodd *et al.*, 2007). In Table 8.1 we present the values of D_2 and L_c for each geometry, as well as the corresponding value of the Hencky strain. The hyperbolic walls were designed assuming the hyperbolic function: $y(\mu\text{m}) = \pm 200/[1 + 0.05(x + L_c)]$, valid for $-L_c \leq x \leq 0$. The final PDMS micro channels obtained by replication in the SU-8 mold were imaged in the microscope set up, and the real shape of the microchannels was slightly different (maximum deviation of about 5-10 μm), and followed accurately the curve $y(\mu\text{m}) = \pm 200/[1 + 0.05(x + L^*) - 2.5 \times 10^{-5}(x + L^*)^2]$ with the characteristic length L^* presented in Table 8.1. The corner features of the final geometries were not as sharp as printed on the chrome mask, a limitation of the fabrication techniques used, but had a radius of curvature of about 5-10 μm . Moreover, the real value of the depth of the microchannels is actually lower than the projected value (as shown in Table 8.1).

Table 8.1. Values of the Hencky strain and dimensions of the microchannels used.

Projected (chrome mask)		PDMS microchannel					
Channel	ε_H	D_2 [μm]	L_c [μm]	ε_H	D_2 [μm]	L^* [μm]	h [μm]
S_1	1.0	147	34.4	0.76	187.1	33	47
S_2	2.0	54.1	128	1.86	62.4	123	45
S_3	3.0	19.9	382	2.75	25.5	373	44

8.2.2. Flow visualization

Visualizations of the flow patterns were carried out using streak line photography with exposure times of the order of 1 s. For this purpose, the fluids were seeded with 1 μm fluorescent polystyrene tracer particles (20 ppm w/w, Nile Red, Molecular Probes, Invitrogen, Ex/Em:

520/580 nm, density: 1.05 g/mL) and, in the case of the blood analogue solutions, sodium dodecyl sulfate (0.05 wt. %, Sigma-Aldrich) was added in order to reduce adhesion of the particles to the channels walls. A syringe pump (PHD2000, Harvard Apparatus) was used to inject the fluid and control the flow rate in the microchannels. Syringes with different volumes (50 μ L-10 mL) were used, according to the desired flow rate, and were connected to the microgeometries using Tygon tubing. The microgeometries were placed on an inverted epi-fluorescence microscope (DMI 5000M, Leica Microsystems GmbH) and were continuously illuminated by a 100 W mercury lamp. A filter cube (Leica Microsystems GmbH, excitation filter BP 530-545 nm, dichroic 565 nm and barrier filter 610-675 nm) was used to filter the multichromatic light into excitation light which reaches the microgeometries containing the seeded fluid via a microscope objective (10 $\times$, NA = 0.25). The light emitted by the fluorescent tracer particles was imaged through the same objective onto the CCD array of the camera (DFC350 FX, Leica Microsystems GmbH), in order to capture the particle trajectories. All streak line images presented here are centered at the mid-plane of the microchannel. The flow visualization experiments were carried out at room temperature ($T = 293.2 \pm 1.5$ K), for a wide range of flow rates ($1 \lesssim Q \lesssim 70$ mL h⁻¹ for de-ionized water and $0.001 \lesssim Q \lesssim 70$ mL h⁻¹ for blood analogues).

8.2.3. Particle image velocimetry

Measurements of the velocity field were carried out using micro-particle image velocimetry (μ PIV). For that purpose, the fluids were seeded using fluorescent Nile Red polystyrene microspheres 0.5 μ m in diameter (90 ppm w/w, Molecular Probes, Invitrogen, Ex/Em: 520/580 nm, density: 1.05 g/mL). The generic imaging set-up used is similar to that described in Section 8.2.2 except for the microscope objective and light source. In this case, two different objectives were employed: a 20 $\times$ objective (NA = 0.4) in order to capture the particle displacement in a wide field of the microchannel, and a 40 $\times$ objective (NA = 0.55) in order to improve the accuracy of the results within the contraction region, where the width is much smaller and the velocity is considerably higher. The light source used for μ PIV is a doubled pulsed Nd:YAG laser (Dual Power 65-15, Dantec Dynamics) with a wavelength of 532 nm. The time interval between pulses was adjusted in the range of $1 \leq \Delta t / \mu\text{s} \leq 10^4$, such that particle displacement was about 25% the size of the interrogation area, which was selected according to the velocity of the flow and the objective used. For each flow rate, 150 pairs of images were acquired using a digital CCD camera (Flow Sense 4M, Dantec Dynamics), with a resolution of 2048 $\times$ 2048 pixels and running in double frame mode. The images were post-processed to

obtain the velocity vector map using DynamicStudio v2.3 software (Dantec Dynamics) based on averaging the 150 pairs of images. All images were acquired at the mid-plane of the microchannel.

For the optical set-up used in microfluidics, it is well known that the depth of field (δz) underestimates the actual depth over which there is a contribution of unfocused particles to the velocity field determined by μ PIV and, thus, the measurement depth, calculated as:

$$\delta z_m = \frac{3n\lambda_0}{(\text{NA})^2} + \frac{2.16d_p}{\tan \theta} + d_p, \quad (8.1)$$

where n is the refractive index, λ_0 is the wavelength of the light (in vacuum), NA is the numerical aperture of the microscope objective, d_p is the particle diameter, provides a more correct estimate of the depth of the image plane (Meinhart *et al.*, 2000). Table 8.2 lists the depth of field and the total measurement depth for the range of objectives used with our optical set-up.

Table 8.2. Depth of field and total measurement depth for the present optical set-up.

Objective	δz (μm)	δz_m (μm)
20×	4.55	13.9
40×	2.25	7.89

8.2.4. Pressure drop measurements

Pressure-drop measurements were performed connecting the two pressure taps of the microchannels, which are located far upstream ($x = -2.8$ mm) and far downstream ($x = 2.8$ mm) of the test section, to a differential pressure transducer (Honeywell, model 26PC series). This pressure sensor was chosen based on the range of measurable differential pressure ($\Delta P_{\text{max}} = 6.89$ kPa). A 12 V DC power supply (Lascar electronics, PSU 206) was used to power the sensor, which in turn was connected to a data acquisition card (NI USB-6218, National Instruments) interfaced with LabView v7.1. The output voltage of the sensor was previously calibrated in terms of pressure difference using a static column of water.

8.3. Fluid composition and rheology

Two different viscoelastic fluids, which have a shear rheology behavior similar to that of human blood (Thurston, 1996; Vlastos *et al.*, 1997), were used. The blood analogue solutions are aqueous solutions of a xanthan gum (Sigma-Aldrich) with a concentration of 500 ppm (w/w) and of a polyacrylamide (Sigma-Aldrich; average molecular weight, $M_w = 1.8 \times 10^7 \text{ g mol}^{-1}$) at a concentration of 125 ppm (w/w), as suggested by Vlastos *et al.* (1994). A Newtonian fluid, de-ionized water, was also used for comparison purposes. The density (ρ) of the XG and PAA solutions at 293.2 K is $\rho = 998.0 \text{ kg m}^{-3}$ and $\rho = 998.2 \text{ kg m}^{-3}$, respectively.

The rheology of the fluids was characterized with a rotational rheometer (MCR301, Anton Paar) and a capillary break-up extensional rheometer (Haake CaBER 1, Thermo Scientific) was used to determine the relaxation time of the fluid in extensional flow. This property was measured at the reference temperature ($T_0 = 293.2 \text{ K}$) in extensional flow using two circular plates of 6 mm in diameter (D_p). Moreover, the geometrical configuration used for both fluids was such that the initial height was set to $h_i = 1.99 \text{ mm}$, which corresponds to an initial aspect ratio of $A_i = h_i/D_p = 0.33$. The final height was varied in a number of exploratory tests, and the best results were obtained using $h_f = 8.36 \text{ mm}$ and $h_f = 9.10 \text{ mm}$ for the XG and PAA solutions, respectively. Consequently, the final aspect ratio used in the measurements, which is defined as $A_f = h_f/D_p$, was $A_f = 1.39$ and $A_f = 1.52$ for the XG and PAA fluids, respectively. The evolution of the filament thinning monitored in the CaBER experiments is illustrated in Figure 8.2(a) as $\log[D(t)/D_0]$ vs time (where D_0 is the radius of the filament at time $t = 0$). The relaxation time (λ) was determined by fitting the experimental data in the linear region of the semi-log plot to the equation $D(t) / D_0 \propto \exp[-t / (3\lambda)]$ resulting from the elasto-capillary balance (Entov and Hinch, 1997). The relaxation time for the blood analogue solution composed of xanthan gum was 3.77 ms while for the blood analogue composed of PAA was 38.0 ms, which is one order of magnitude greater than the relaxation time of the XG solution.

It is appropriate here to refer that to the best of our knowledge there are no archival publications with data on the elongational properties of blood. However, we are aware of preliminary work by Poole *et al.* (2006), who have attempted the measurement of blood relaxation time using a CaBER. The relaxation times measured by Poole *et al.* (2006) were on the order of one hundred milliseconds for hematocrit $48\% < \text{Hct} < 52\%$, which is in excess of the relaxation time of both blood analogues used here. As such, we would expect a more pronounced elastic behavior (namely in terms of vortex size upstream of the contraction and pressure drop)

for real blood than what was observed here for either blood analogues. However, a complete comparison must be left until a more detailed study with blood is performed and/or published.

The steady shear rheology was measured in the temperature range of $283.2 \leq T / \text{K} \leq 303.2 \text{ K}$ using a cone-plate geometry (75 mm, 1° angle) under shear rate control ($0.1 \leq \dot{\gamma} / \text{s}^{-1} \leq 5 \times 10^3$) and the time-temperature superposition method (Dealy and Plazek, 2009) was used to obtain the master curve. The variation of the shift factor, a_T , with temperature can be described accurately by an Arrhenius equation (Dealy and Plazek, 2009):

$$\ln(a_T) = \frac{\Delta H}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right), \quad (8.2)$$

where ΔH is the activation energy for flow, R is the universal gas constant, T is the temperature of the measurement and T_0 is the reference temperature, which is set here as $T_0 = 293.2 \text{ K}$. The shift factor is defined as (Dealy and Plazek, 2009):

$$a_T = \frac{\eta(T)}{\eta(T_0)} \frac{T_0}{T} \frac{\rho_0}{\rho}, \quad (8.3)$$

where $\eta(T)$ and ρ are the shear viscosity and the fluid density at temperature T and $\eta(T_0)$ and ρ_0 are the shear viscosity and the fluid density at the reference temperature, T_0 , respectively. In the range of measurements performed the temperature variation is small, therefore the fluid density does not change significantly and the shift factor can be simplified to (Dealy and Plazek, 2009):

$$a_T = \frac{\eta(T)}{\eta(T_0)}. \quad (8.4)$$

For the Newtonian fluid, $\Delta H/R = 2.05 \times 10^3 \text{ K}$ and $\eta_0 = 1.00 \times 10^{-3} \text{ Pa s}$ at the reference temperature. For the viscoelastic fluids, a ratio $\Delta H/R$ of $2.79 \times 10^3 \text{ K}$ and $1.82 \times 10^3 \text{ K}$ was obtained for the PAA and for the XG solutions, respectively. Figure 8.2(b) shows the resulting master curves for both blood analogue solutions, as well as the predicted values for a modified Phan-Thien-Tanner (MPTT) model (Xue *et al.*, 1995), which corresponds to a standard PTT model (Phan-Thien and Tanner, 1977) having an explicit shear-thinning viscosity coefficient. This model was chosen because of the viscoelastic shear-thinning rheological behavior of the blood

analogue solution used. We note that human blood has a small yield stress, which is not reproduced using these blood analogues, neither by the MPTT model, which was chosen in order to reproduce accurately the shear rheology of the blood analogues.

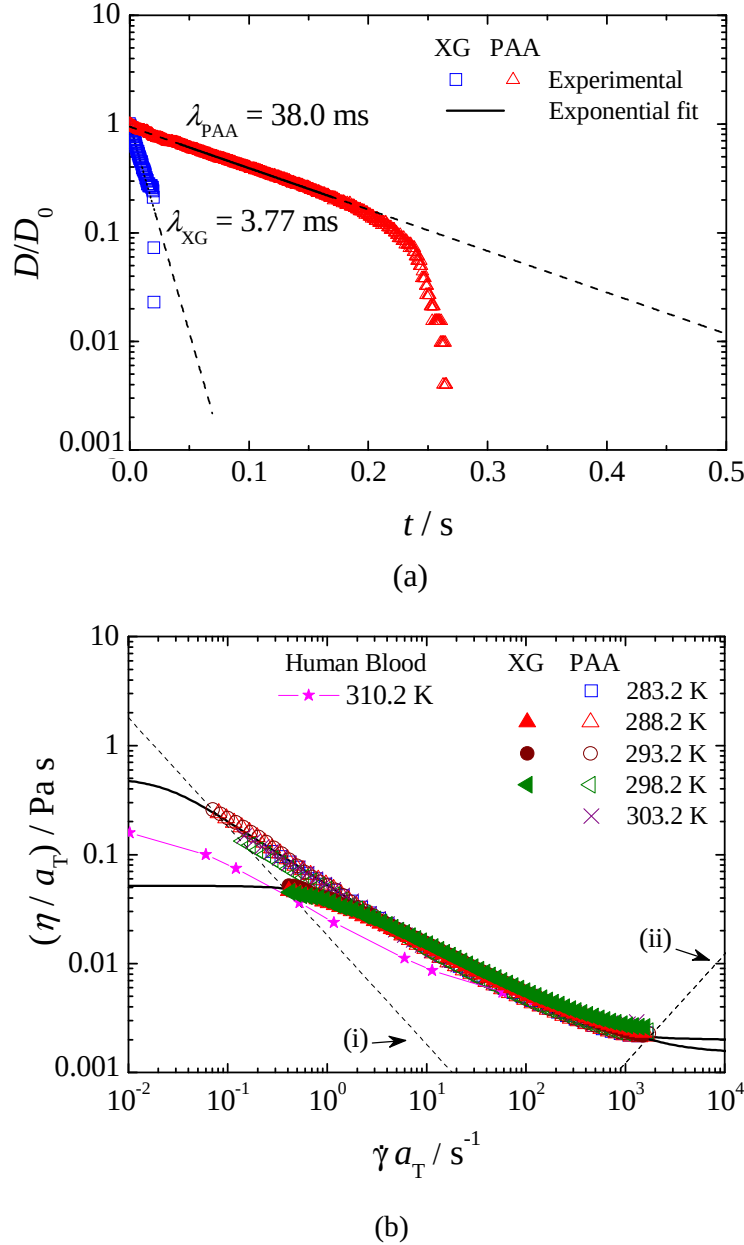


Figure 8.2. (a) Time evolution of the mid-point diameter of the fluid thread in a CaBER experiment at 20°C. The symbols represent the experimental data and the lines represent the exponential fit to the experimental data. (b) Master curves of the steady shear viscosity for the XG and PAA blood analogue solutions. The steady shear data for human blood at 310.2 K reported in Vicent (1982) is also shown for comparison purposes. (i) Minimum measurable shear viscosity determined from 20× the minimum measurable torque of the rheometer; (ii) onset of secondary flow due to Taylor instabilities for the 75 mm, 1° cone-plate geometry.

In general, the constitutive equation of the PTT model, which governs the polymer contribution to the extra-stress tensor ($\boldsymbol{\tau}$), is given by:

$$f(\text{Tr}(\boldsymbol{\tau})) \boldsymbol{\tau} + \lambda \left[\frac{\partial \boldsymbol{\tau}}{\partial t} + \mathbf{u} \cdot \nabla \boldsymbol{\tau} \right] = \eta_{\text{polymer}} (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \lambda (\boldsymbol{\tau} \cdot \nabla \mathbf{u} + \nabla \mathbf{u}^T \cdot \boldsymbol{\tau}), \quad (8.5)$$

where $\mathbf{u}$ is the velocity vector, t is the time, η_{polymer} is the shear-rate dependent viscosity coefficient of the model and $\text{Tr}(\boldsymbol{\tau})$ is the trace of the polymer extra-stress tensor. We use the linear form of the PTT model and therefore, the stress function is given by (Phan-Thien and Tanner, 1977):

$$f(\text{Tr}(\boldsymbol{\tau})) = 1 + \frac{\lambda \varepsilon}{\eta_{\text{polymer}}} \text{Tr}(\boldsymbol{\tau}). \quad (8.6)$$

Since we are using the modified form of the PTT model, the viscosity of the polymer is given by a Carreau-type equation (Xue *et al.*, 1995):

$$\eta_{\text{polymer}} = \frac{\eta_p}{\left[1 + (\Gamma \dot{\gamma})^2 \right]^{\frac{1-n}{2}}}, \quad (8.7)$$

where η_p is the zero-shear-rate polymer viscosity, Γ is a time constant, n is the power-law index and $\dot{\gamma}$ is the shear-rate, which represents the magnitude of the shear rate tensor, $\dot{\boldsymbol{\gamma}} = \nabla \mathbf{u} + \nabla \mathbf{u}^T$,

and can be calculated as $\dot{\gamma} = \sqrt{\frac{1}{2} II(\dot{\boldsymbol{\gamma}})} = \sqrt{\frac{1}{2} \sum_i \sum_j \dot{\gamma}_{ij}^2}$, where $II(\dot{\boldsymbol{\gamma}})$ is the second invariant of the shear rate tensor.

The parameters of the MPTT model used are: the extensibility parameter, ε ; the polymer shear viscosity coefficient, η_p ; the relaxation time, λ ; the time constant Γ ; the power-law index, n . The respective values are presented in Table 8.3 for each fluid. Additionally, a solvent viscosity (η_s) is also added, which is incorporated directly in the momentum equation.

Table 8.3. Parameters of the MPTT model at the reference temperature ($T_0 = 293.2$ K).

Fluid	ε	$\eta_p / \text{Pa s}$	$\eta_s / \text{Pa s}$	λ / ms	Γ / s	n
XG	0.05	0.05	0.0015	3.77	1.3	0.5
PAA	0.01	0.498	0.002	38.0	45	0.4

8.4. Numerical method and computational meshes

The flow of the reference Newtonian fluid (de-ionized water) was simulated numerically in order to compare the flow patterns and velocity field with the experimental numerical results and consequently validate the measurement technique. In addition, for the PAA solution we performed numerical simulations using a generalized Newtonian fluid (GNF) to probe for the effects of the shear-thinning viscosity in the absence of elasticity. Comparison of these numerical calculations with the experimental results allow us to assess the effect of elasticity on the observed experimental results. The MPTT model was not used in the numerical simulations, because of the high Deborah number flow conditions of the experiments that creates numerical problems due to the high-Weissenberg number problem (Owens and Phillips, 2002).

We assume that the flow is laminar, isothermal and incompressible. Therefore, the equations that need to be solved are those of conservation of mass and momentum,

$$\nabla \cdot \mathbf{u} = 0, \quad (8.8)$$

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \eta_s \nabla^2 \mathbf{u}, \quad (8.9)$$

where p is the pressure. For the calculations using the Newtonian fluid the polymeric extra stress tensor is neglected ($\boldsymbol{\tau} = \mathbf{0}$) while for the generalized Newtonian fluid calculations we use a Carreau model to describe the shear-thinning behavior of the shear viscosity:

$$\eta = \eta_s + \frac{\eta_0 - \eta_s}{[1 + (\Lambda \dot{\gamma})^2]^{(1-n)/2}}, \quad (8.10)$$

where η_0 is the zero shear-rate viscosity, Λ is a time constant and n is a power-law like index. The two last terms in Eq. (8.9) are replaced by $\nabla \cdot [\eta(\nabla \mathbf{u} + \nabla \mathbf{u}^T)]$.

For the PAA fluid, the parameters of the Carreau model are: $\eta_0 = 0.5 \text{ Pa s}$; $\eta_s = 0.002 \text{ Pa s}$; $A = 29 \text{ s}$; $n = 0.33$.

The governing equations were solved using a fully implicit finite-volume method with a time-marching pressure correction algorithm, which is described in detail in Oliveira *et al.* (1998), Alves *et al.* (2003) and, consequently, are not explained here.

The numerical simulations are three-dimensional (3D) and the meshes used to represent the computational domain are block-structured, composed of non-uniform control volumes. The mesh characteristics, such as the total number of cells (NC) and the normalized minimum cell spacing ($\Delta x_{\min}/D_2$, $\Delta y_{\min}/D_2$, $\Delta z_{\min}/D_2$), are listed in Table 8.3 Table 8.4 and an exemplifying mesh used in the simulations (corresponding to microchannel S_3) is shown in Figure 8.3. Symmetry was enforced at the two center planes ($x = 0$ and $z = 0$), which means that only a quarter of the full domain was mapped, reducing significantly the memory requirements and the computational times.

Table 8.4. Characteristics of the meshes used in the numerical simulations.

Mesh	NC	$\Delta x_{\min}/D_2$	$\Delta y_{\min}/D_2$	$\Delta z_{\min}/D_2$
MS ₁	49920	8.03×10^{-3}	7.29×10^{-3}	5.04×10^{-3}
MS ₂	50080	2.09×10^{-2}	2.04×10^{-2}	2.12×10^{-2}
MS ₃	66720	2.14×10^{-2}	2.50×10^{-2}	1.90×10^{-2}

Regarding the boundary conditions, we imposed a uniform velocity profile (average velocity U_1) at entrance, no-slip conditions at the solid walls and at the outlet vanishing gradients for velocity components and linear extrapolation of pressure are imposed. The inlets and outlets were positioned far from the constriction in order that fully-developed flow conditions were attained prior to the contraction region.

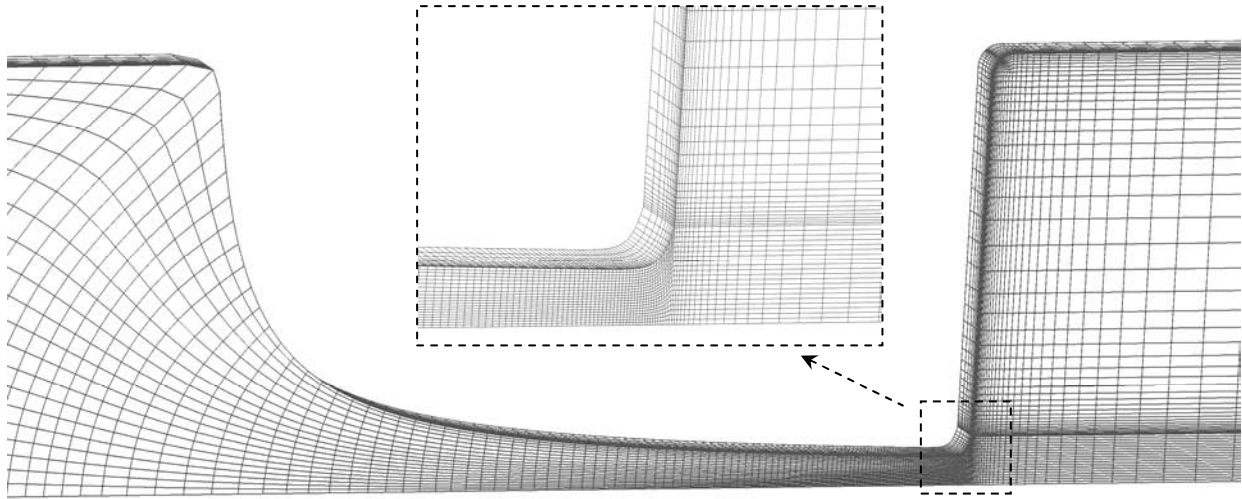


Figure 8.3. Zoomed view of the mesh used in the numerical simulations of the flow in microchannel S_3 .

8.5. Flow patterns

8.5.1. Newtonian Fluid

The flow of a Newtonian fluid (de-ionized water) was studied for all microgeometries in order to validate the experimental technique and simultaneously investigate the influence of inertia on the flow patterns in the absence of elasticity. The Reynolds number (Re) is defined as $Re = \rho U_2 D_2 / \eta$, where U_2 is the average velocity in the narrowest gap of the contraction, with width D_2 (cf. Figure 8.1(b)). The Reynolds number can also be expressed in terms of the flow rate, $Re = \rho Q / (\eta h)$.

Figure 8.4 compares the flow patterns predicted numerically and captured experimentally at the center plane of the microchannel S_2 for the two flow directions and at different Reynolds numbers in order to highlight the effect of inertia.

For both flow directions, the Newtonian fluid flow behavior is found to be qualitatively similar for all microgeometries studied. Moreover, the appearance of recirculations downstream of the expansion (either abrupt or smooth) brought about by inertia, occurs in an analogous way for the two flow directions, even though higher Reynolds numbers are required in the backward direction. At low Reynolds numbers, the fluid is pushed towards the centerline as it moves through the contraction and no flow separation is observed except near the far corners of the expansion, where small Moffatt vortices exist (cf. Figure 8.4(a₁), 8.4(a₂)).

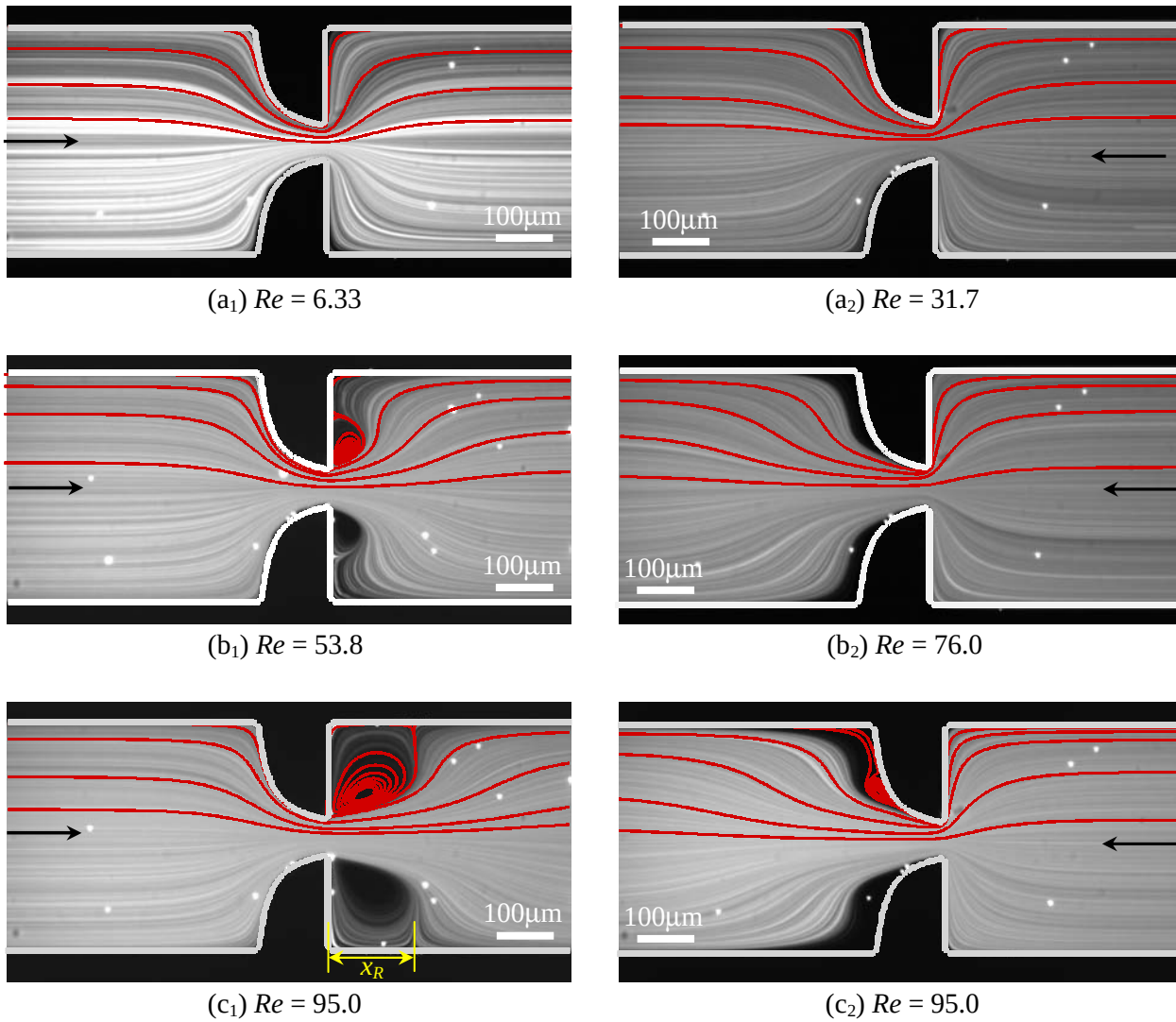


Figure 8.4. Effect of inertia on the Newtonian fluid flow patterns obtained experimentally (photograph) and predicted numerically (solid lines) for microchannel S_2 . The left-hand side column corresponds to flow through the hyperbolic contraction followed by the abrupt expansion (forward direction) and the right-hand side column corresponds to the flow through the abrupt contraction followed by the hyperbolic expansion (backward direction).

Increasing the flow rate leads to the onset of flow separation downstream of the expansion due to the flow being driven against an adverse pressure gradient, leading to flow separation. First, lip vortices appear close to the re-entrant corners (Figure 8.4(b₁) and 8.4(c₂)) and eventually these vortices enlarge to the far corner with increasing Re . As inertia is further increased, the vortices grow in size in the downstream direction (Figure 8.4(c₁)). With further increases in the flow rate (not attained in the experiments) the flow will eventually become asymmetric, with recirculations downstream of the expansion plane having different sizes, as

found by Oliveira *et al.* (2007) for abrupt microfluidic expansions and by Cherdron *et al.* (1978), among others, for planar expansions at macro-scale. The experimental flow features, including the onset of vortex formation and vortex growth, are well captured by the numerical predictions.

In Figure 8.5 we compare the dimensionless vortex length, x_R / D_1 (cf. Figure 8.4(c₁) for x_R definition), predicted numerically and measured experimentally from the flow visualizations for a range of Reynolds numbers, and for the three micro geometries investigated. For all cases, a vortex increase is observed as inertial effects are enhanced, and this effect is more intense for the higher Hencky strain geometries. Good agreement between experiments and numerical simulations are observed, and considering a deviation of 3 μm in the depth of the channel is enough to explain any of the small differences observed.

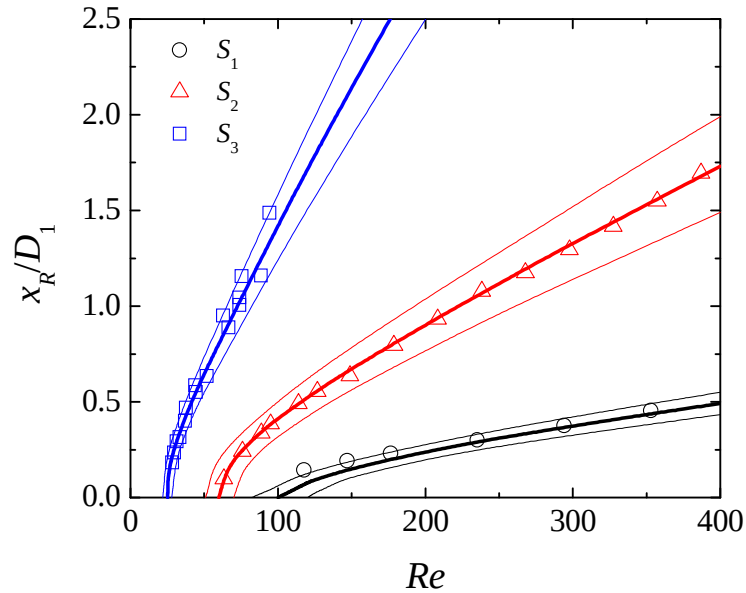


Figure 8.5. Influence of Reynolds number on the length of the recirculation formed downstream of the abrupt expansion for the Newtonian fluid (forward flow). The symbols represent experimental data obtained for channels S_1 , S_2 and S_3 and the thick solid lines represent the corresponding numerical predictions using the depth of the channel (cf. Table 8.1). The solid thin lines represent the numerical predictions when the depth of the channel is varied $\pm 3 \mu\text{m}$ (upper line: $h + 3 \mu\text{m}$; lower line: $h - 3 \mu\text{m}$).

The values of Re at which the different flow patterns arise depends on the geometry used. For geometries with higher Hencky strain, the onset of lip vortices (cf. Figure 8.4(b₁)) and subsequent development of corner vortices (cf. Figure 8.4(c₁)) occurs at lower Re , and the vortices exhibit a more elongated shape. For the backward flow direction, recirculations emerge downstream of the hyperbolic expansion at higher values of Re than for the corresponding

forward flow direction (cf. Figure 8.4). This behavior is expected since in the forward direction, the fluid is flowing through an abrupt expansion, in which the adverse pressure gradient is higher, as compared with the smooth expansion flow, where the adverse pressure gradient is weaker. For microchannel S_2 the numerical calculations underpredict the experimental vortex size, but assuming a deviation of $3\text{ }\mu\text{m}$ in the depth of the channel is enough to explain the observed difference.

8.5.2. Blood analogue fluids

In order to characterize the viscoelastic fluid flow, we use the Deborah number, here defined as $De = \lambda U_2 / (D_2 / 2)$. The Reynolds number definition presented in Section 8.5.1 is also used, but the shear viscosity, η , is evaluated at a characteristic shear rate $\dot{\gamma}_2 = U_2 / (D_2 / 2)$. The range of estimated shear rates achieved with the different geometries are shown in Table 8.5 for the range of flow rates tested experimentally.

Table 8.5. Range of flow rates and estimated shear rates used in the experiments with the blood analogues.

Channel	Min Q [ml h ⁻¹]		Max Q [ml h ⁻¹]		Min $\dot{\gamma}_2$ [s ⁻¹]		Max $\dot{\gamma}_2$ [s ⁻¹]	
	PAA	XG	PAA	XG	PAA	XG	PAA	XG
S_1	1.0×10^{-2}	1.0×10^{-2}	10	60	3.37	3.37	3.37×10^3	2.02×10^4
S_2	2.0×10^{-3}	5.0×10^{-2}	1.0	30	6.34	158	3.17×10^3	9.51×10^4
S_3	1.0×10^{-3}	1.0×10^{-2}	0.6	20	19.5	195	1.17×10^4	3.89×10^5

In Figure 8.6(a) we show the flow patterns obtained experimentally for the PAA blood analogue solution flowing in the forward flow direction of microgeometry S_2 .

At very low flow rates (and consequently low De), the flow patterns are Newtonian-like (cf. Figure 8.6(a₁)) and on increasing the flow elasticity, or in other words, on increasing De , symmetric lip vortices develop upstream of the contraction and grow to the far corners (cf. Figure 8.6(a₂)). The formation of corner vortices upstream of a microfluidic hyperbolic contraction has also been discussed by McKinley *et al.* (2007) for the flow of a 0.3 wt.% PEO aqueous solution. Increasing further the flow rate, and consequently the elastic effects, leads to a large increase of the vortex size (cf. Figure 8.6(a₃)) until a critical Deborah number is reached. Above this critical De the flow becomes unsteady and asymmetric, due to an elastic instability,

and the size of the upstream vortices varies in time (cf. Figure 8.6(a₄), where the *instantaneous* flow field is clearly asymmetric). An important characteristic to bear in mind is that inertial effects remain insignificant since the values of Re in these experiments are very small.

These results demonstrate that the PAA fluid flow is highly elastic, and viscoelasticity leads to the formation of strong upstream vortices, in contrast to flow inertia that leads to downstream separation. The PAA fluid flow behavior is similar for all microchannels studied and the results for the other geometries are not shown for conciseness.

In the case of the XG blood analogue solution, the elasticity-driven flow transitions upstream of the contraction are similar to those observed with the PAA solution, but the vortex growth is weaker and occurs at significantly higher flow rates due to the lower value of the XG fluid relaxation time. Consequently, the flow is markedly different from that observed with the PAA solution, upstream and downstream of the expansion, because the inertial effects become relevant as the flow rate is increased, as shown in the streak line images of Figure 8.6(b) acquired for the XG solution in the same microgeometry (S_2).

At low flow rates, Newtonian-like flow patterns are observed (not shown here) and increasing the flow rate leads to the appearance of symmetric vortices upstream of the contraction (cf. Figure 8.6(b₁)) that increase in size with the flow rate, due to the enhancement of elastic effects. When the flow rate is increased further, inertial effects also become important and symmetric vortices are observed downstream of the abrupt expansion (cf. Figure 8.6(b₄)), as for the Newtonian fluid flow. Small corner vortices and lip vortices appear near the reentrant corner of the expansion plane (cf. Figure 8.6(b₂)) which join together forming large corner vortices (cf. Figure 8.6(b₃)).

For high flow rates, the flow becomes unstable (cf. Figure 8.6(b₄)) with the corner vortices varying in size periodically.

Comparing the viscoelastic and the Newtonian fluid flow (Figure 8.4) at similar Re , we conclude that these downstream vortices are smaller for the viscoelastic fluid than for the Newtonian fluid, since elasticity is responsible for the development of normal stresses in shear, which are known to lead to jet swelling and consequently to a reduction of the size of downstream recirculations (Poole *et al.*, 2009).

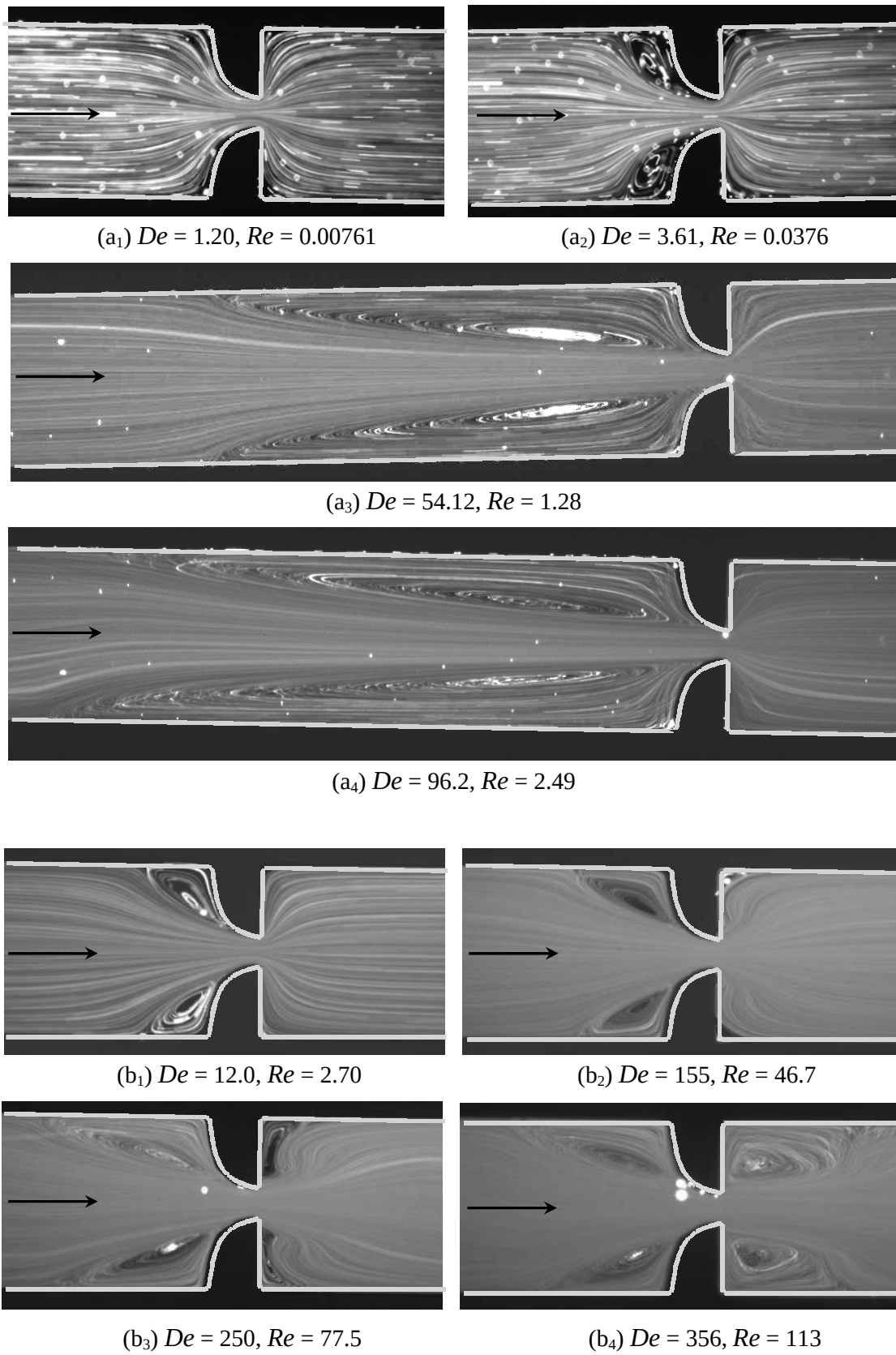


Figure 8.6. Streak line images taken at the center plane of the microgeometry S_2 for PAA (a) and XG (b) blood analogue solutions flowing in the forward flow direction.

In Figure 8.7 we present the flow patterns obtained experimentally for both fluids flowing in the backward direction in microgeometry S_2 . For the PAA fluid the flow features in the two flow directions are similar, but take place at different flow conditions (lower Q or De for the backward flow direction – cf. Figure 8.6(a) and 8.7(a)).

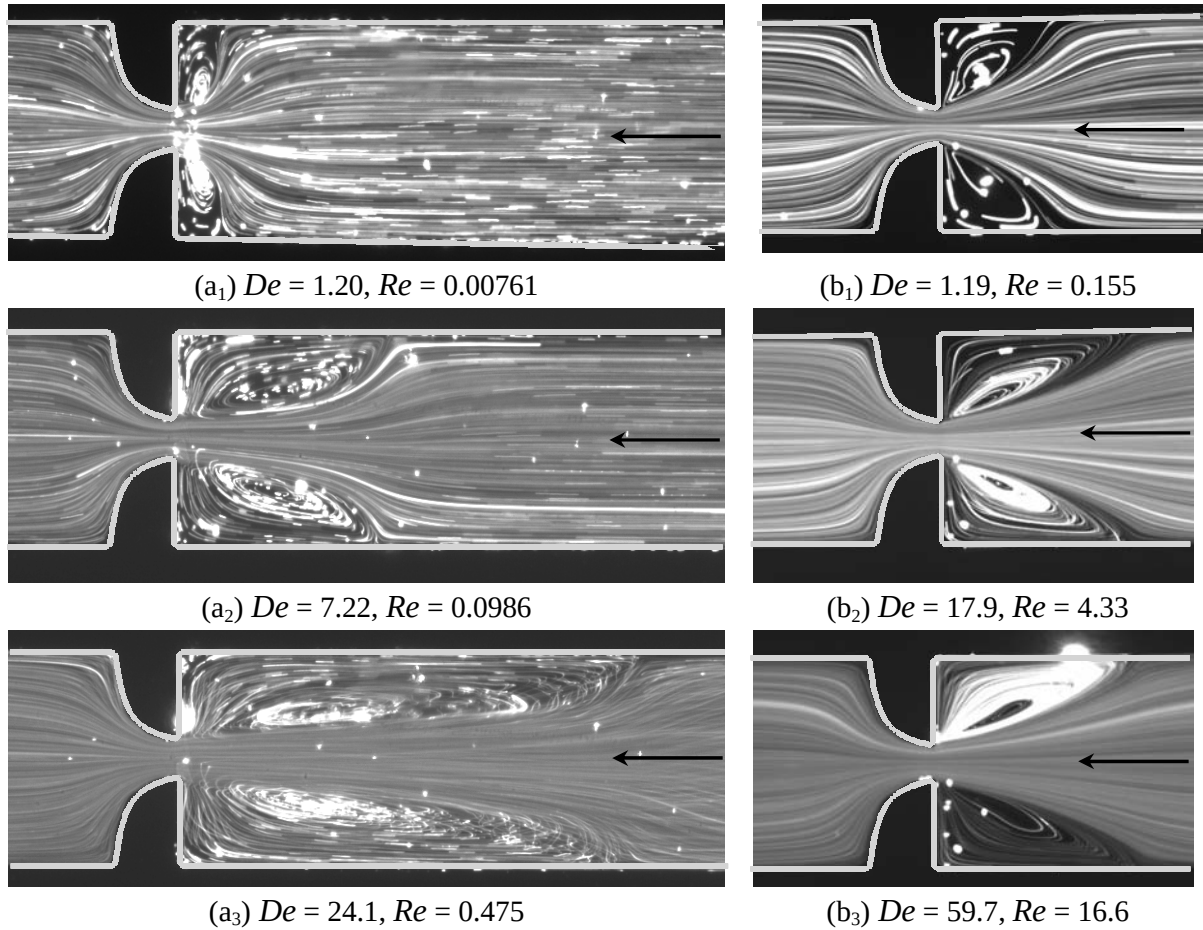


Figure 8.7. Streak line images taken at the center plane of the microgeometry S_2 for the PAA (a) and XG (b) blood analogue solutions flowing in the backward flow direction.

For the XG fluid flow in the backward direction, and for the range of flow rates measured, inertial effects were not observed (cf. Figure 8.7(b)) possibly due to the fact that the recirculations downstream of the smooth expansion appear at higher flow rates than in the opposite forward flow direction, as was seen for the Newtonian fluid flow (cf. Section 8.5.1).

The upstream (elastic) vortex length corresponding to geometries S_1 , S_2 and S_3 is plotted in Figure 8.8 as function of the Deborah number for the forward flow direction. For all cases the vortex size increases significantly with De , but much more intensively for the PAA solution. The increase of the vortex size with De is qualitatively different for the two polymers, with the

growth regime for XG being qualitatively similar to that observed by Gulati *et al.* (2008) for a DNA solution (of concentration $c = 4c^*$, with c^* being the overlap concentration) in flows through 2:1 abrupt contractions. Furthermore, despite the differences in geometry (Gulati *et al.* (2008) used a 2:1 abrupt contraction with a larger aspect ratio, i.e. a deeper micro-device) the range of vortex sizes reported by Gulati *et al.* (2008) is comparable to those obtained here with the XG solution. These similarities are rather interesting, and may be associated to the fact that both DNA and XG are rigid rod-like molecules.

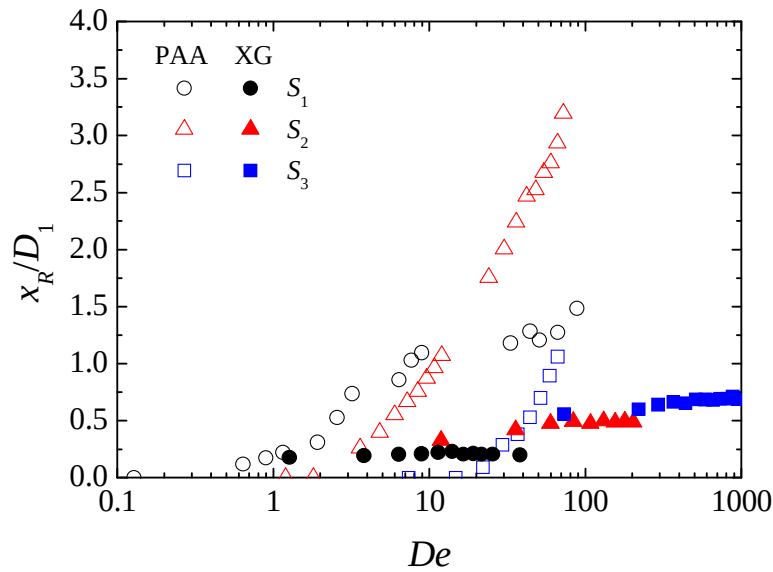


Figure 8.8. Upstream vortex size as function of De for PAA and XG fluid analogues (forward flow direction).

In summary, markedly different flow behavior was found for the two viscoelastic fluids in these obstructed ducts, with the PAA solution showing stronger viscoelastic effects, even though both fluids are considered blood analogues and exhibit similar rheological characteristics in steady shear flow.

8.6. Velocity field

The velocity field in the centre plane ($z = 0$) was measured using μ PIV for the flow of the Newtonian and the PAA blood analogue solution. These experiments were conducted using microchannel S_2 with the fluids flowing through the hyperbolic contraction followed by the sudden expansion (forward direction only). In addition, for validation purposes the experimental

results obtained for the Newtonian fluid case are compared with the corresponding numerical predictions.

8.6.1. Inertial effects

Figure 8.9 shows the dimensionless axial velocity profiles along the centerline measured for the Newtonian fluid at different flow rates. Moreover, a comparison between experimental and numerical results is presented in order to validate the μ PIV experimental measurements. The axial velocity profile predicted for the Newtonian fluid flowing at creeping flow conditions ($Re \rightarrow 0$) is also included in Figure 8.9.

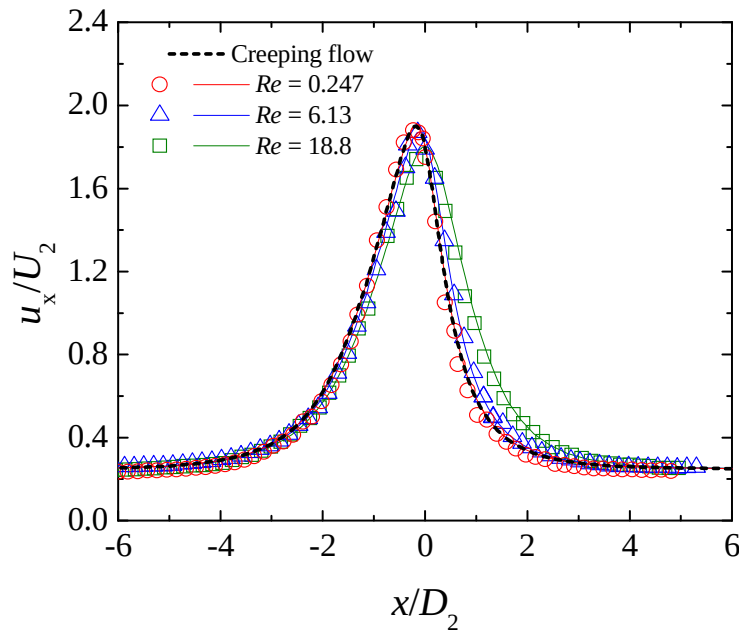


Figure 8.9. Experimental (symbols) and numerical (lines) dimensionless axial velocity profiles at the centerline of the microchannel S_2 for the Newtonian fluid flow. The solid lines represent the numerical predictions for each Re and the dashed line represents the numerical predictions for creeping flow.

It is clear that as the fluid approaches the hyperbolic contraction the axial velocity starts to increase due to entrance effects. After passing through the contraction, the fluid decelerates until the flow re-develops downstream of the abrupt expansion. The prediction of an axial velocity profile for the Newtonian fluid flowing at creeping-flow conditions is also included in Figure 8.9. For the lower flow rate, inertia is not important and the axial velocity profile coincides with that predicted numerically for creeping flow ($Re \rightarrow 0$). As the flow rate increases, fully-developed conditions are reached gradually farther downstream of the expansion due to inertial

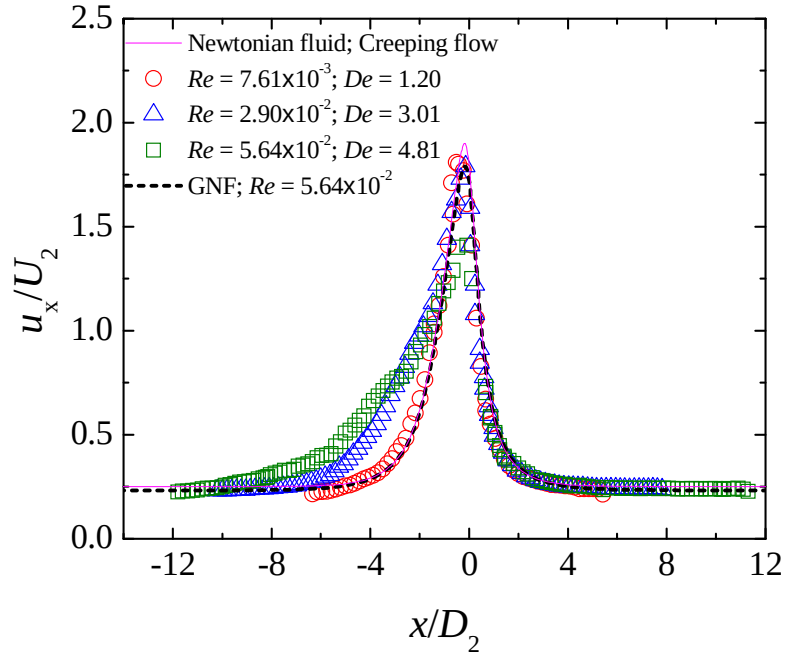
effects, in agreement with the results of Oliveira *et al.* (2007), and the profiles are pushed towards the downstream direction. In addition, it is clear from Figure 8.9 that the experimental results are in excellent agreement with those predicted numerically, thus validating the μ PIV measurements.

8.6.2. Elastic effects

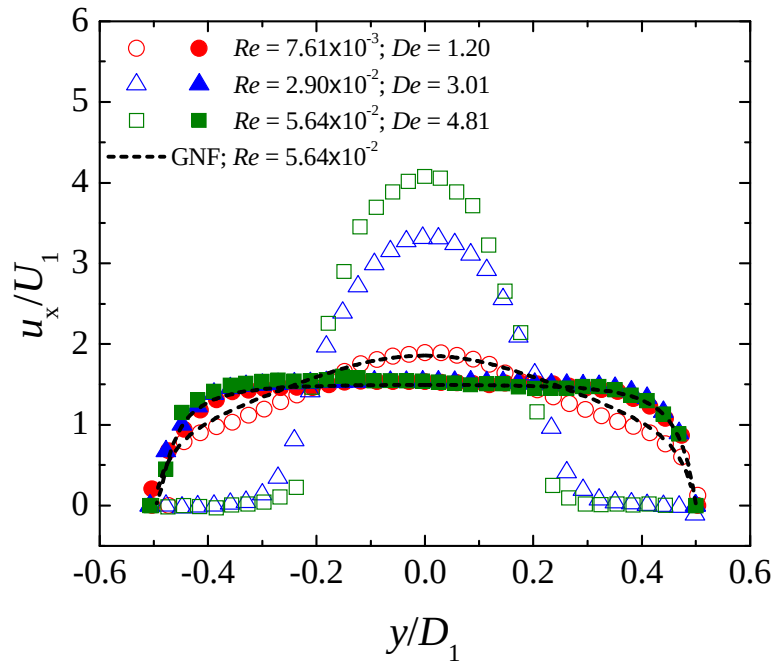
In order to highlight the effect of elasticity on the flow kinematics, Fig. 8.10 presents axial velocity profiles obtained at different flow rates (or Deborah numbers) for the PAA blood analogue solution in channel S2. As shown in Section 8.5.2, using the PAA solution we are able to probe for elastic effects while minimizing inertial effects. In Figure 8.10(a), we report the axial velocity profiles obtained along the centerline and in Fig. 8.10(b) those obtained along the spanwise direction (y -direction) for different axial locations corresponding to $x/D_1 = -0.63$ (just upstream of the contraction region) and $x/D_1 = 1.64$ (far downstream of the contraction region). Additionally, the axial velocity profile predicted numerically for a Newtonian fluid under creeping flow conditions, and for a GNF fluid described by the Carreau model fitted to the PAA shear rheology, are also shown in Figure 8.10 for comparison purposes. This GNF simulation corresponds to the highest flow rate achieved in the measurements with the PAA solution.

At the lowest flow rate (and De) for which μ PIV measurements were performed, the axial velocity profile along the centerline in Figure 8.10(a) is analogous to that obtained numerically for the Newtonian fluid under creeping flow conditions, which is the limiting case when both Re and De vanish.

Increasing the flow rate leads to significant changes of the normalized axial velocity profiles upstream and in the hyperbolic contraction region, as expected from the flow visualizations presented in Section 8.5.2. In fact, the upstream recirculations observed for higher De act as an extension of the contraction, and as a result, the fluid at the centerline starts to accelerate further upstream in the channel (Figure 8.10(a)), an effective way to reduce the strain-rate and consequently the flow resistance for strain-hardening fluids.



(a)



(b)

Figure 8.10. Dimensionless axial velocity profiles in the microchannel S_2 : (a) along the centreline ($y = 0$); (b) along the spanwise direction (for $x/D_1 = -0.63$ and $x/D_1 = 1.64$). The symbols represent experimental data obtained for the PAA blood analogue, the solid line represents the numerical predictions for the Newtonian fluid flow under creeping flow conditions and the dashed lines represent the numerical predictions for a generalized Newtonian fluid (Carreau model).

The dimensionless axial velocity profile along the centerline obtained using the GNF model is similar to that found for the Newtonian fluid, except in the microchannel throat, where its value is lower than for the Newtonian fluid due to shear-thinning effects (Figure 8.10(a)). The GNF prediction is also similar to the experiments at the lowest flow rate, where elasticity is at a minimum and only shear thinning matters (Figure 8.10(a) and 8.10(b)). Comparing the experimental results obtained for the viscoelastic fluid at the higher flow rates with the corresponding GNF predictions, it is possible to observe significant differences in the velocity profiles particularly upstream and in the contraction region [cf. profiles in Figure 8.10(a) and profiles at $x/D_1 = -0.63$ in Fig. 8.10(b)], revealing once again, the strong elastic character of the flow of the PAA solution through this type of microgeometry that is enhanced when strong velocity gradients are observed.

8.7. Pressure drop

We measured the pressure drop across the test sections using the PAA blood analogue, for microgeometries S_2 and S_3 . Figure 8.11 shows the pressure drop obtained as a function of the flow rate for the forward flow direction.

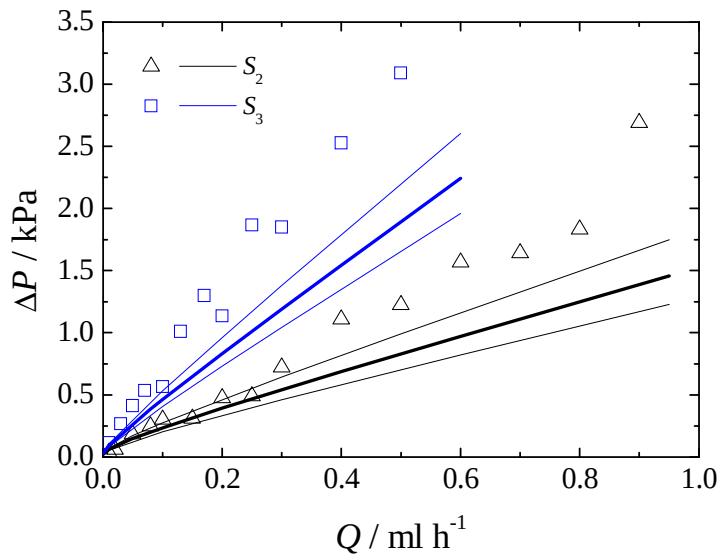


Figure 8.11. Pressure drop of the PAA blood analogue solution, measured for the forward flow direction, as a function of the flow rate for microchannels S_2 and S_3 . The symbols represent experimental data and the solid lines represent the numerical predictions using the Carreau model. The thick solid lines represent the numerical predictions using the depth of the channel and the thin solid lines represent the numerical predictions assuming a variation of $\pm 3 \mu\text{m}$ in the depth of the channels (upper line: $h - 3 \mu\text{m}$; lower line: $h + 3 \mu\text{m}$).

The pressure drop increases approximately linearly with the flow rate, or Deborah number, for all microgeometries studied. As expected, for the same value of the flow rate, the pressure drop is higher for channels with higher values of the total Hencky strain, which have longer and narrower contraction regions and produce stronger extensional flows. In Figure 8.11 we also show the corresponding values predicted numerically using the Carreau model. The solid thick lines represent the numerical predictions using the depth of the channel measured experimentally, while the thin lines represent the numerical predictions assuming a variation of $\pm 3\mu\text{m}$ in the channels depth.

As can be seen, at low flow rates, the pressure drop measured experimentally and predicted numerically using the GNF model are similar. However, above $Q \sim 0.1 - 0.2 \text{ ml h}^{-1}$, elastic effects become increasingly noticeable and the numerical predictions of the pressure drop using the GNF model, which consider the shear-thinning behavior but not the existence of elasticity, are substantially lower than the experimental results for the higher Hencky strains. We believe that the high extensional viscosity of the PAA fluid, that generates high normal stresses in the contraction region where a strong extensional flow is observed, is responsible for the distinct behavior, leading to the strong quasi-linear increase of the pressure drop with the flow rate for the geometries with the higher Hencky strains.

8.8. Conclusions

The flow of non-Newtonian fluids, commonly employed as blood analogues, through hyperbolic and abrupt contractions/expansions revealed a considerably different behavior under strong extensional flow. Our rheological analysis confirms that both blood analogues have a shearthinning behavior very similar to that of whole human blood but the relaxation time for both analogues differs by one order of magnitude. Moreover, the characteristics of the flow through obstructed ducts observed with the viscoelastic fluids are very different from those found with the Newtonian fluid. In the latter, inertia promotes the appearance of vortices downstream of the expansion plane. On the other hand, in the viscoelastic fluid flow case, complex flow patterns brought about by the elastic nature of the polymeric solutions are observed. When inertia is negligible, elastic-driven vortices appear upstream of the constriction, which increase in size when De is increased. Oppositely, when inertial effects are significant, downstream vortices appear, which increase with Re and result in a decrease of the size of the upstream vortices. The PAA blood analogue solution revealed a stronger elastic nature than the XG blood analogue, with the flow eventually becoming unsteady even in the absence of significant inertial effects. In contrast, in the flow of the XG blood analogue solution, both

elastic and inertial effects are observed before the flow becomes unsteady. The pressure drop measured with PAA solution in the forward direction was found to increase quasi-linearly with the flow rate due to the presence of elastic effects, which compensate for the shear-thinning effects that act to slightly reduce the pressure drop as shown by the GNF simulations. Hence, it is possible to conclude that comparing the flow of human blood with the Newtonian fluid flow at the microscale is a rough over-simplification. Moreover, taking into consideration only the shear viscosity in order to manufacture a blood analogue solution with a viscoelastic character is not adequate, as demonstrated by the markedly different results obtained in a strong extensional flow using two common blood analogue solutions.

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CONCLUSIONS

9.1. Thesis conclusions

The main objective of this thesis was the investigation of viscoelastic fluid flow within channels of simple geometries, in which strong extensional flow is generated. For this purpose, the study was divided into two main parts with specific objectives: firstly we used a macro-scale geometry with a square cross-section to investigate the laminar flow through three-dimensional square-square contractions and expansions; secondly, microfluidic channels composed of hyperbolic and abrupt contractions/expansions were employed.

Macro-scale studies

The selected square-square arrangement was found to be an excellent 3D test-case since it provides a good compromise between geometric simplicity and complex 3D flow structure. Sudden square-square contractions and expansions were used to perform a systematic investigation of the effect of inertia on the Newtonian fluid flow and the effect of elasticity on the viscoelastic fluid flow under creeping flow conditions. For this purpose, two viscoelastic fluids with different rheological behavior were employed, namely a Boger fluid and a shear-thinning viscoelastic fluid. Furthermore, the effect of contraction (or expansion) ratio was also analyzed.

For both contraction and expansion flows, at low Deborah number flow conditions, the viscoelastic fluid flow is similar to that found for creeping flow of a Newtonian fluid. Nevertheless, when elastic effects set in, the flow becomes significantly different and eventually at high flow rates elastic instabilities appear.

The Newtonian fluid flow through square-square contractions presents a Moffatt corner vortex and, for all contraction ratios investigated, increasing the Reynolds number leads to a monotonic decrease of the vortex length and intensity. On the other hand, for the Newtonian fluid flow through square-square expansions, increasing flow inertia leads to an increase of the vortex size for all CR analyzed.

The contraction flow of the two viscoelastic fluids revealed a different flow behavior with the Boger fluid presenting a more complex range of flow phenomena. For both fluids, the effect of the contraction ratio on the flow patterns results in two distinct behaviors that can be grouped in a region for low ($CR = 2.4$ and 4) and high ($CR = 8$ and 12) contraction ratios.

For the Boger fluid, at low CR , increasing the elasticity of the flow leads to a decrease of the vortex length until a minimum size is reached, followed by the onset of diverging streamlines upstream of the contraction. In terms of the velocity field, a significant undershoot in the axial velocity profile along the centerline is observed upstream of the contraction plane, as a result of the diverging streamline flow pattern. In addition, downstream of the contraction region, the fluid accelerates and the axial velocity profile exhibits an overshoot that increases with De . For high contraction ratios, a localized divergent flow near the re-entrant corner is observed, which contributes to the onset of an overshoot in the axial velocity profiles at the centerline. Elastic effects lead to the onset and enhancement of lip and corner vortices, respectively, and at high De the flow eventually becomes unsteady due to the onset of an elastic instability.

For the shear-thinning viscoelastic fluid, diverging streamlines also appear upstream of the contraction plane for low CR and a strong vortex enhancement as well as unsteady flow, are found independently of the contraction ratio.

The flow behavior of both viscoelastic fluids through square-square expansions is similar. A corner (Moffatt) vortex is observed downstream of the expansion plane, which decreases in size when elastic effects increase.

In summary, it was demonstrated that the flow through square-square contractions/expansions is highly three-dimensional with open spiraling vortex structures. Furthermore, when elastic effects become important, the dynamics of the viscoelastic fluid flow is opposite to that found for the Newtonian fluid flow. The numerical results showed a good agreement with experimental results. In addition, numerical predictions allowed a thorough investigation of the 3D flow characteristics and allowed the assessment of some flow features that were not noticeable

through experimental measurements. Moreover, the study at the macro-scale was particularly useful for implementation and validation of the experimental techniques, namely the PIV system, prior to the investigation at micro-scale.

Micro-scale studies

At the micro-scale, a significant part of the work was devoted to the development of an efficient rectifier, able to operate efficiently with dilute polymer solutions even under creeping flow conditions. Previous works concerning valveless or no-moving parts micropumps have focused on Newtonian fluid flows. In those cases, rectification effects are due exclusively to inertial effects. However, microfluidic flows typically occur under low Reynolds number flow conditions and future reductions in scale will further reduce the flow inertia. As such, anisotropic flow resistance in microfluidic devices can only be achieved using the non-linear properties of viscoelastic fluids.

Microchannels with different shapes based on triangular elements, as documented in the literature (Groisman and Quake, 2004; Nguyen *et al.*, 2008) and on hyperbolic shaped elements were investigated. For the Newtonian fluid flow, the pressure drop through the microchannels is found to increase linearly with the flow rate even up to moderate Reynolds number flows, and no rectification effects were observed in the range of Reynolds number measured ($Re \lesssim 250$). For the viscoelastic fluid flow, a different flow behavior was observed and significant anisotropic flow resistance was found even for dilute polymeric solutions. In this case, elasticity is enhanced due to the small length scales, and leads to a markedly different flow resistance for both flow directions and consequently to the onset of rectification effects. It was found that those rectification effects are enhanced when elastic instabilities emerge in the forward flow direction. Under those flow conditions, the forward flow becomes unsteady and the flow resistance increases significantly with the flow rate. Moreover, the hyperbolic shape leads to a quasi-ideal extensional flow that generates significant increases in the pressure drop for polymer solutions with high molecular weight macromolecules. The hyperbolic shaped microfluidic rectifiers, where a quasi-ideal extensional flow is observed, presented higher diodicities than the triangular shaped microchannels. Subsequently, the micro-rectifier with a hyperbolic shape, which showed the best rectification effect, was employed with different aspect ratios in order to assess the wall effects on the diodicity. For the higher aspect ratio, the shear component of the flow is reduced

and the extensional component becomes more important, leading to the improvement of the diodicity of the microchannel.

In addition, the Newtonian fluid flow was also simulated numerically and good agreement between experimental and numerical velocity profiles along the centerline was found, validating the μ PIV technique.

In summary, a microfluidic rectifier consisting of a series of hyperbolic contractions/abrupt expansions was proposed and demonstrated to operate efficiently under creeping flow conditions, with maximum achieved diodicities on the order of seven, which is significantly higher than any previously published work.

The last objective of this thesis was to investigate the extensional flow of well-established blood analogue fluids in microfabricated hyperbolic contractions followed by sudden expansions. The two viscoelastic fluids studied have shear rheology similar to that documented for human blood but different elasticity levels, and consequently revealed different flow patterns. For negligible inertial flow conditions, elastic-driven vortices appear upstream of the hyperbolic contraction, which increase in size with De . For the less elastic blood analogue, downstream vortices set in when inertial effects become significant, which grow in size as flow inertia increases, but to a lower extent than for the Newtonian fluid flow. At high De the flow becomes unsteady for both blood analogue fluids.

From the experimental observations it was possible to conclude that preparing a polymeric solution with a shear viscosity similar to that of human blood is not sufficient in order to create a good blood analogue. Extensional properties should also be taken into account, especially when the fluids are to be used in flows at small length-scales when characteristic times of the flow and fluid become comparable.

In summary, from the various studies undertaken at macro- and micro-scale, it was demonstrated that strong extensional flows can be used in a wide range of applications, and that contraction and expansion flows are useful for generating strong extensional flows, with the hyperbolic arrangement being particularly useful due to the quasi-homogeneous conditions generated.

9.2. Future work

The experimental results obtained in the investigation of the flow of viscoelastic fluids through square-square contractions and expansions provided valuable data that can be used for benchmark purposes in future works. Validation of three-dimensional numerical codes, as well as micro-scale studies of complex 3D flows, are two of the most relevant applications that shall benefit from the results of this thesis.

Follow-up work for blood analogue fluids would best be performed using real human blood for direct comparison and assessment of elastic behavior. Flow visualizations, pressure drop measurements and velocity field measurements will add important knowledge about the flow behavior of human blood, and comparing the observed flow patterns with those of the blood analogue solutions studied in this thesis will undoubtedly provide useful information about the extensional viscosity of real human blood. Also, the development of more reliable blood analogues should be envisioned, considering not only the shear rheological properties, but also the elasticity level and the extensional behavior of the blood analogues in canonical flows.

Further work on the improvement and final implementation of the in-situ photopolymerization technique should be pursued. This new technique revealed a great potential for rapid fabrication of microchannels, but was not explored in more detail due to the time constraints. This rapid prototyping technique proved to be very versatile, since it allows the manufacture of microfluidic devices without a photomask by using the moving stage microscope system. In this way, it is possible to test different designs and microchannels with different shapes in a few minutes. Nevertheless, more tests should be carried out to improve the cleaning process and to improve the verticality of the channel walls, if precise channels are required. Further experiments using these photopolymerized microchannels should also be done in order to ensure that they can be used efficiently as an alternative to PDMS microchannels fabricated using standard techniques such as photolithography and soft-lithography.

APPENDIX

APPENDIX A. PDMS MICROCHANNEL FABRICATION

The spincoated PDMS layer existing on top of the glass slide, which is used to seal the microchannels, should have an adequate thickness so that the entire depth of the channel is within the focal length of the microscope objective used to visualize the flow. Moreover, a uniform thickness along the glass slide is desired.

Different spincoating velocities were investigated in order to evaluate the thickness of the resulting PDMS layer. The spin velocities were tested following the same protocol: first, a 10 s velocity ramp was imposed, following a constant spin velocity during 50 s. The resulting glass slide with the PDMS layer was placed in the oven to cure for 24 h at 80°C. After that, the thickness of the PDMS layer, Δl , was measured using a digital caliper (Lineartools, resolution 10 μm). The spin velocities tested and corresponding thickness of the PDMS layer are presented in Figure A.1.

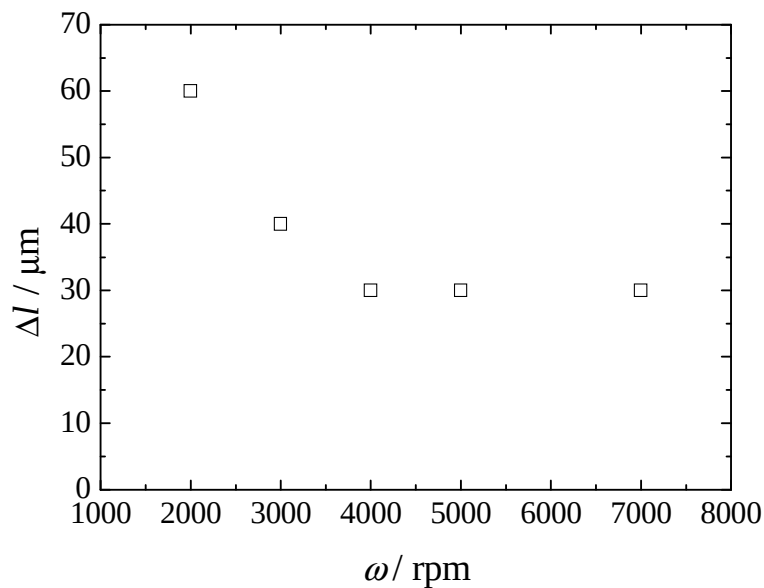


Figure A.1. Thickness of the PDMS layer as a function of the spincoating velocity.

It is possible to infer that using a spincoating speed of 4000 rpm or above, the thickness of the layer becomes nearly independent of the spin speed. We selected $\omega = 5000$ rpm for the glass slides coating, because the results at this velocity showed a uniform thickness over the whole extension of the glass slide.

APPENDIX B. PROCEDURE FOR MACRO-SCALE PARTICLE IMAGE VELOCIMETRY MEASUREMENTS

Useful tips:

- at least 5 particles should be focused on each interrogation area;
- the diameter of the seeding particles should have 2-3 pixels of the acquired image. If this criteria is not met it is useful to slightly defocus the camera;
- the particles presented in the acquired images should have a maximum displacement between frames of about one quarter of the width of the interrogation area defined.

Procedure to achieve a final vector map using adaptive correlation:

Step i – Image masking. The acquired images are masked. For that, we define first the mask by selecting the area of the image to be rejected. Once the mask is created, it is applied to all acquired images.

Step ii – Correlation. An adaptive correlation is applied to the set of masked images. The initial and final interrogation area sizes selected were 64 pixels $\times$ 64 pixels and 16 pixels $\times$ 16 pixels, respectively, with 50 % of overlap. In this case, the maximum displacement of the seeding particles may be less than one quarter of the initial interrogation area and more than one quarter of the final interrogation area. In the iterative procedure, it is possible to validate the intermediate vectors, which are created during the refinement of the interrogation areas. The validation methods used were the peak validation and the moving average validation.

The correlation peaks, resulting from fast Fourier transform (FFT) processing, are used to assess the quality of the measured vectors by analyzing the height and location of these correlation peaks in the correlation plane. In the peak validation, each vector is validated by comparing the height of the highest FFT peak with the height of the second highest FFT peak (peak 2) in the correlation plane. Keane and Adrian (1992)² demonstrated that a peak height ratio of 1.2 should be attained for this criterion, known as the detectability criterion. The position of the peak in the correlation plane corresponds to the average particle displacement within the

² Keane, R.D. and Adrian, R.J., 1992. Theory of cross-correlation analysis of PIV images. Applied Scientific Research 49, 191-215.

interrogation area under investigation and the height of the peak depends not only on the number of particles but also on the light intensity in the interrogation area.

In the moving average validation, a local neighborhood validation is made by comparing each vector with its neighbors in an area of 3×3 pixels. An average vector is calculated based on all vectors within the neighborhood and compared with each vector under investigation. The allowed deviation from the average vector to each vector is called the acceptance factor. If the acceptance factor is 1, all vectors are validated, whereas if the acceptance factor equals 0, all vectors are rejected. In this study, an acceptance factor of 0.1 was used.

In the calculations of the adaptive correlation, a window filter was also applied, namely a Gaussian filter. This filter reduces the problem of losing particles along the boundaries of the interrogation areas and improves the signal-to-noise ratio. The value of the parameter $1/k^2$ used was 0.75, where the constant, k , determines the width of the window.

When the adaptive correlation is applied, each pair of images originates a vector map.

Step iii – Vector map masking. The mask created in step (i) is applied to all vector maps in order to remove undesired noise.

Step iv – Validation. The vector maps created and masked are subjected to a validation step, in which some vectors are rejected. This validation process is the moving average validation previously explained in step (ii). An average vector is calculated based on all vectors within the averaging area.

Step v – Final vector map. The set of vector maps (already validated) is used to determine a mean velocity vector map. For the same position within the interrogation area, each vector component (u and v) is used to calculate a single average value and consequently create one vector map: the final vector map.

APPENDIX C. OPTIMIZATION OF MICRO-PARTICLE IMAGE VELOCIMETRY MEASUREMENTS USING A STRAIGHT MICROCHANNEL

Micro-particle image velocimetry measurements were performed in a straight channel, with a rectangular cross-section using a Newtonian fluid (de-ionized water) in order to improve the procedure used in the experiments. For this, the experimental results obtained were compared with the analytical solution for Poiseuille flow in a rectangular channel.

Considering a microchannel with a rectangular cross-section, in which the depth and width is h and D , respectively, the analytical solution for the axial velocity is given by (White, 2003)³:

$$u(y, z) = \frac{16(D/2)^2}{\mu\pi^3} \left(-\frac{dp}{dx} \right) \sum_{i=1,3,5,\dots}^{\infty} (-1)^{(i-1)/2} \left[1 - \frac{\cosh(i\pi z / D)}{\cosh(i\pi h / 2D)} \right] \frac{\cos(i\pi y / D)}{i^3} \quad (C.1)$$

where the pressure gradient can be related with the flow rate by:

$$\left(-\frac{dp}{dx} \right) = \frac{3Q\mu}{2h(D/2)^3 \left[1 - \frac{192D}{\pi^5 h} \sum_{i=1,3,5,\dots}^{\infty} \frac{\tanh(i\pi h / 2D)}{i^5} \right]} \quad (C.2)$$

Therefore, the axial velocity profile was calculated using the analytical solution for fully-developed flow conditions and the results were compared with those obtained experimentally. The parameters used, which lead to a better accuracy of the experimental results, are then selected for the subsequently μ PIV measurements. The parameters tested are listed below.

a) μ PIV correlations

Different correlations were applied to the set of double-frame images. For all, an overlap of 50% between surrounding interrogation areas was chosen.

³ White, F. M., 2003. Fluid mechanics. 5th ed., McGraw-Hill, Boston.

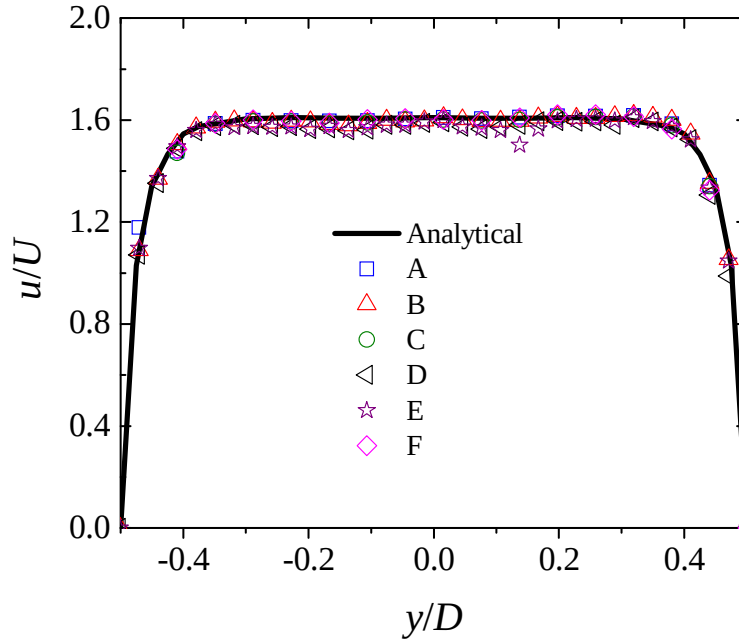


Figure C.1. Axial velocity profile at the centerplane of the straight channel. A–Average correlation 128 pixels $\times$ 64 pixels; B–Average correlation 64 pixels $\times$ 32 pixels; C–Average correlation 64 pixels $\times$ 64 pixels; D–Average correlation 32 pixels $\times$ 32 pixels; E–Adaptive correlation 128 pixels $\times$ 128 pixels to 32 pixels $\times$ 32 pixels; F–Adaptive correlation 256 pixels $\times$ 256 pixels to 64 pixels $\times$ 64 pixels.

The results obtained using the average correlation present a smaller deviation from the analytical solution than those obtained using the adaptive correlation. Moreover, interrogation areas with dimensions of the range from 128 pixels to 64 pixels lead to experimental results more close to the calculated analytical results, as illustrated in Figure C.1.

b) Average correlation with/without application of window filter

Using a set of 150 images, we tested the effect of applying a window filter on the resultant velocity field. For this, an average correlation without a filter and with a window filter was applied. The window filter used was a Gaussian filter, as for macro-PIV (cf. Appendix B), with $1/k^2 = 0.75$. Comparing the results obtained for the two conditions, it is possible to conclude that both are similar, as illustrated in Figure C.2.

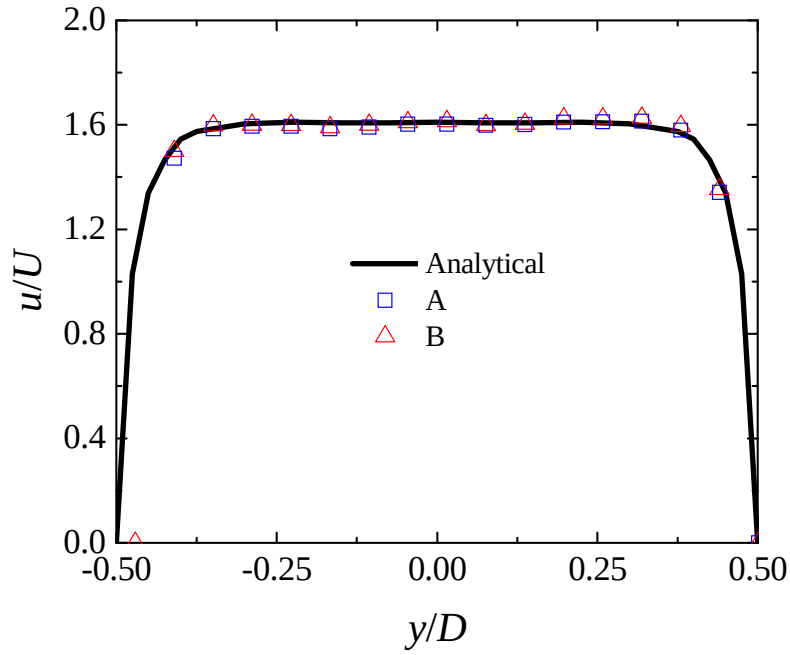


Figure C.2. Axial velocity profile at the centerplane of the straight channel. A–average correlation without window filter; B–average correlation with window filter.

c) Number of acquired images

The total number of double frame images acquired during the measurements was varied in order to assess the effect of the number of images on the accuracy of the velocity field. In Figure C.3 we compare the results obtained using 70, 100 and 150 pairs of images.

As can be seen, in spite of the differences between the experimental results being small, a higher number of images leads to a better accuracy of the results.

d) Repeatability of the results

Velocity measurements were performed at the same conditions in two different days in order to test the repeatability of the results. A set of 150 pairs of images was acquired and an average correlation was applied. The dimensions of the interrogation areas used are 64 pixels $\times$ 64 pixels and 50% of overlap was selected. The results are illustrated in Figure C.4, where it is possible to verify the repeatability of the results.

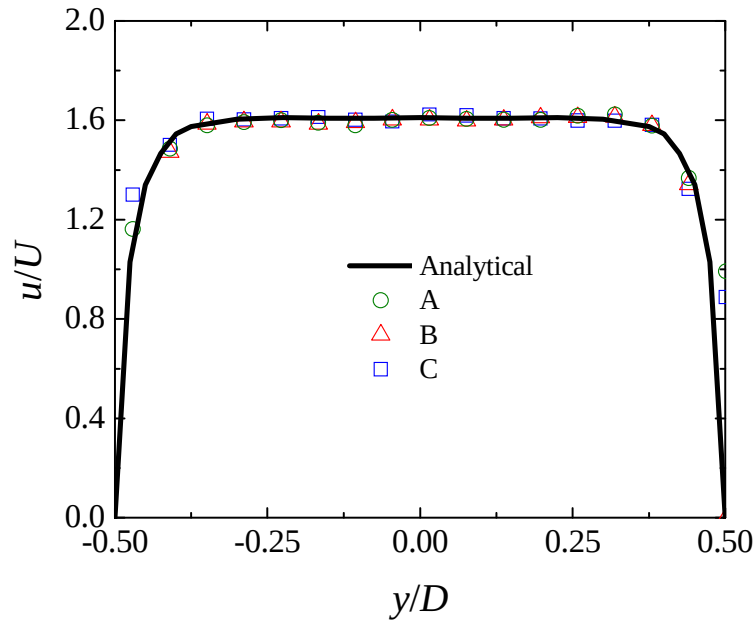


Figure C.3. Axial velocity profile obtained at the centerline of the straight channel using different number of pairs of images. A–70 pairs of images; B–100 pairs of images; C–150 pairs of images.

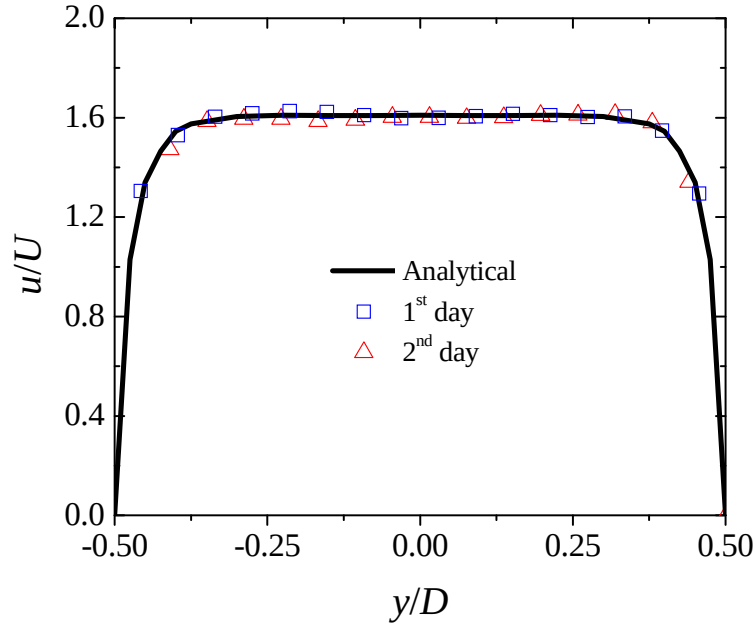


Figure C.4. Comparison between axial velocity profiles obtained experimentally in two different days with the analytical solution.

APPENDIX D. CALIBRATION OF A PRESSURE SENSOR

The pressure sensor (Honeywell, 26PCA FA6D), which covers a differential pressure range up to 6.9 kPa, was calibrated using a static column of water. For this, the ports of the pressure sensor were connected to two tubes having approximately 3 m of length, which were positioned vertically.

First, de-ionized water was added to both tubes and air bubbles were removed. After that, keeping constant the level of water in one tube, which was considered as the reference height, the calibration measurements were performed by adding de-ionized water to the other tube. After reaching the steady-state response of the sensor, an average voltage value was measured using the data acquisition card (NI USB-6218, National Instruments).

Since the pressure of the column of water can be calculated by:

$$\Delta p = \rho g \Delta h, \quad (\text{D.1})$$

where Δh is the height difference of the two levels of water in both tubes, it is possible to relate the output voltage value provided by the pressure sensor with the corresponding pressure difference. The same procedure was carried out for the two tubes. The values of the heights of water, the corresponding pressure difference as well as the sensor signal are illustrated in Table D.1. Furthermore, Figure D.1 shows the calibration data and the corresponding equation used to convert the output voltage signal of the pressure sensors into a pressure difference value.

Table D.1. Calibration data for a differential pressure sensor covering a range up to 6.9 kPa: heights of water in the tubes, pressure drop calculated using these heights and corresponding signal of the pressure sensor.

Δh / m	Δp / Pa	Signal /mV
-0.0515	-505.2	-0.969
-0.174	-1706.9	0.698
-0.331	-3247.1	2.747
-0.424	-4159.4	4.077
0.0680	667.1	-2.622
0.175	1716.8	-4.070
0.326	3193.2	-6.098

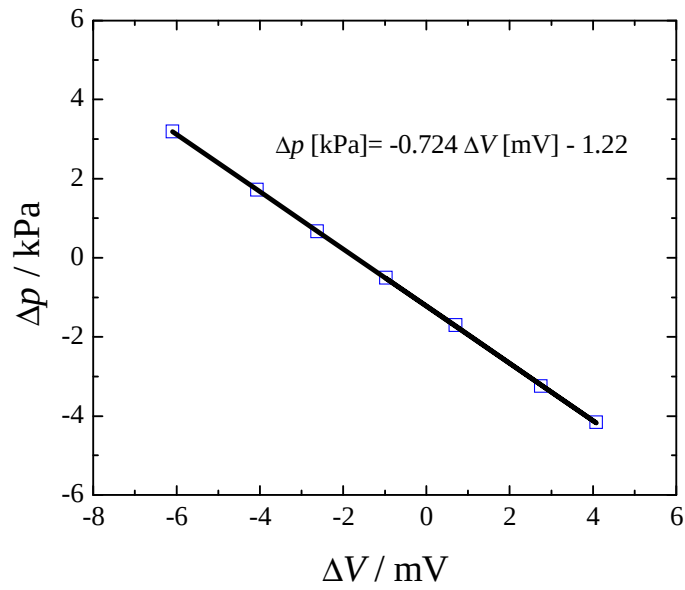


Figure D.1. Calibration data for a differential pressure sensor covering a range up to 6.9 kPa: pressure drop as a function of the output signal of the pressure sensor.

The pressure sensors that measure a differential pressure range up to 200 kPa were calibrated using a compressed air line and a manometer (Wika Instrument Corporation, model 332.50) by reading directly the value of the differential pressure.

APPENDIX E. DETERMINATION OF THE INTRINSIC VISCOSITY OF THE POLYACRYLAMIDE SOLUTION

In order to determine the intrinsic viscosity of the PAA fluid used for the investigation of the microfluidic rectifier, four dilute solutions were prepared by mixing different quantities of polyacrylamide in a similar solvent. Table E.1 presents the composition by weight of the referred solutions as well as the corresponding density.

Table E.1. Composition by weight and density of the solutions measured at 293.2 K.

Solution	PAA [ppm]	Sucrose [%]	NaCl [%]	Water [%]	ρ [kg m ⁻³]
Sol. A	34.9	13.0	1.0	86.0	1059
Sol. B	49.9	13.0	1.0	86.0	1059
Sol. C	65.0	13.0	1.0	86.0	1059
Sol. D	74.6	12.9	1.0	86.1	1059
Solvent		13.0	1.0	86.0	1059

The flow time of each solution using a Cannon-Fenske viscosimeter (Comecta SA, model 1464, size 50) was measured in order to determine the relative viscosity, η_{rel} , and the specific viscosity, η_{sp} . The relative viscosity is defined as the ratio between the solution viscosity, η , and the viscosity of the pure solvent, η_0 , which can also be defined, as the ratio of the flow time of the solution, t , and the flow time of the solvent, t_0 :

$$\eta_{rel} = \frac{\eta}{\eta_0} = \frac{t}{t_0}. \quad (E.1)$$

The specific viscosity is defined as the relative change in viscosity upon addition of polymer and is described as follows:

$$\eta_{sp} = \frac{\eta - \eta_0}{\eta_0} = \eta_{rel} - 1. \quad (E.2)$$

Table E.2 presents the average flow time measured for each solution used as well as the corresponding relative viscosity, specific viscosity and the reduced viscosity, which is defined by dividing the specific viscosity by the solution concentration ($\eta_{red} = \eta_{sp} / C$).

Table E.2. Polymer concentration, average flow time, relative viscosity and specific viscosity of the dilute solutions used for determination of the intrinsic viscosity.

Solution	Polymer concentration [g ml ⁻¹]	Average flow time [s]	η_{rel} [-]	η_{sp} [-]	η_{sp}/C [-]
Sol. A	3.70×10 ⁻⁵	357.91	1.08	0.0834	2257
Sol. B	5.29×10 ⁻⁵	374.80	1.13	0.135	2546
Sol. C	6.89×10 ⁻⁵	392.75	1.19	0.189	2744
Sol. D	7.90×10 ⁻⁵	412.39	1.25	0.248	3144
Solvent	-	330.34	-	-	-

The Huggins equation was employed to determine the intrinsic viscosity. This equation is defined as follows (Brandup *et al.*, 1999)^{iv}:

$$\frac{\eta_{sp}}{C} = [\eta] + K_H [\eta]^2 C, \quad (E.3)$$

where $[\eta]$ is the intrinsic viscosity and K_H is the Huggins coefficient. Figure E.1 shows the reduced viscosity as a function of the concentration of the solutions used as well as the parameters of the Huggins equation determined. Hence, the intrinsic viscosity of the PAA solutions in a solvent composed of water, sucrose and sodium chloride was found as $[\eta] = 1504 \text{ cm}^3/\text{g}$ and the Huggins constant is $K_H = 8.7$.

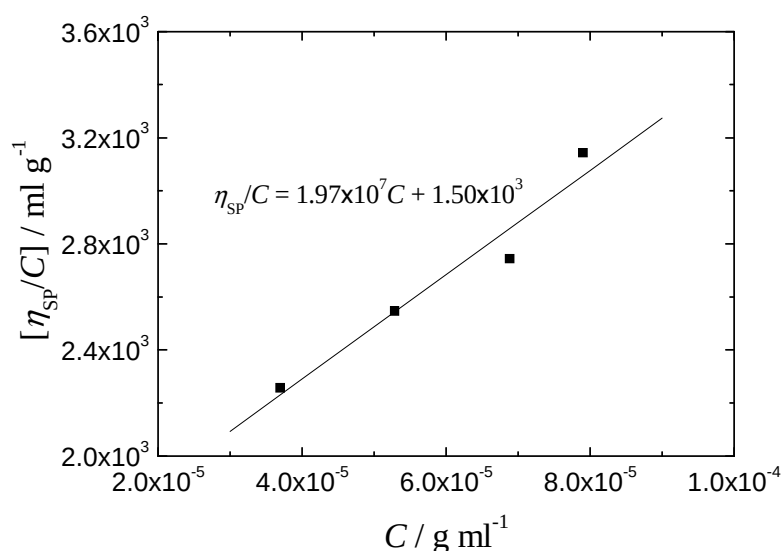


Figure E.1. Reduced viscosity as a function of the concentration of the solutions used to determine the intrinsic viscosity.

^{iv} Brandrup, J., Immergut, E.H. and Grulke, E.A., 1999. Polymer Handbook. 4th edition, John Wiley & Sons, New York

