



Investigating the onset of a viscoelastic instability in the flow around a microfluidic cylinder

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Abstract

Recent efforts to characterise the haemodynamics around swimming microbots through the employment of weakly-elastic, shear-thinning blood analogues in a small-blood-vessel-like microfluidic channel with a centring circular cylinder, were confronted with what seemed to be a flow-asymmetry-inducing viscoelastic instability (Rodrigues et al., 2020). This work aimed to shed light on the possible instability by quantifying the velocity vector field for its onset. The microfluidic devices were fabricated with PolyDiMethylSiloxane (PDMS) and contained pressure taps equally distanced up and downstream of the cylinder that differed on their intake widths; the channels' defining geometries were the same, with a channel aspect ratio of 0.37 and a blockage ratio of 0.28. Two fluids were studied: a non-shear-thinning elastic fluid consisting 2000 ppm of 4MDa poly(ethylene oxide) (PEO) dissolved in a 50 wt% glycerol/distilled water mixture—PEO4, and a viscoelastic, shear-thinning fluid of 5000 ppm of 8 MDa PEO in pure water—PEO8. The flow was characterised via micro-Particle Image Velocimetry for a Weissenberg number range of 100 to 500. It was found, firstly, that the pressure taps with wider intakes heavily influenced the flow, which was not found so dramatically for the weakly elastic solutions of Rodrigues et al. (2020). Secondly, for the narrow tap intake the flow asymmetry was still found for the, more elastic, shear-thinning PEO8, while PEO4 presented a “wineglass” flow pattern. From the seemingly lack of influence that the narrow-intake tap had on PEO4, it could be drawn that the flow asymmetry found for PEO8 would be an elastic instability, however, the flow fields seem to point to the taps' influence on the flow.

Keywords: Microfluidics. Flow asymmetry. Elasticity. Shear-thinning. Microbot.

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Chapter 1

Introduction

Micro-robotics is a rapidly growing subject of interest. Among the many useful applications of microrobots, or “microbots”, their employment in medicine represents an enormous step towards innovative solutions on countless subjects of the field. From the reduction of recovery times and drug side effects (Soto et al., 2020), to cancer diagnosis and treatment (Schmidt et al., 2020), the possible benefits of the use of untethered microrobots in medicine are vast and so are their applications. Medical imaging, drug and cell delivery, and surgery are among the many possible functions of these tiny machines, whose incredible precision allows for localised, minimally invasive action (Soto et al., 2020). However, despite the many advances already made in the field, microbots still face many challenges. Control system and hardware issues pose as some of the main concerns for microbot technology development and application at this stage, which is no wonder since the reduced size of these devices requires its construction to be extremely compact and simple (Sun et al., 2021). Furthermore, for the useful application of microbots *in vivo*, they must be able to adapt to the environment they are subjected to, as the conditions throughout the body change.

Many studies have been made on different ways to move a microbot. Propulsion methods diverge between locally powered with built-in energy conversion systems (Soto et al., 2020), to externally powered through magnetic stimuli, among many other interesting techniques (Yu et al., 2021). Apart from power sources, the approaches to microbot movement itself present a truly fascinating part of its development; examples of interesting solutions can be found in the literature (Ishiyama et al., 2001; Edd et al., 2003; Y. Zhang et al., 2004; Yamazaki et al., 2004).

When it comes to *in vivo* application, it all depends on where the microbot will be applied. The digestive track is an appealing destination for microbot employment (Han et al., 2021), removing the need for endoscope inspection (which can be quite unpleasant for the patients), and, being the track a relatively large channel, the size constraint is reduced quite a bit. Other attractive targets for microbots being currently investigated include the eyes, ears, fetuses, central nervous system, urinary system, and, of course, the circulatory system (Nelson et al., 2010).

Besides the immense range the circulatory system has on the human body, and the advantages of having microbots travel through it, may it be for localised drug delivery or surgery, as mentioned before—there are issues within the blood vessels themselves that could be solved, or at least facilitated, by the employment of these devices. Cardiovascular diseases, or CVDs, namely ischaemic heart disease and stroke, are the leading

causes of death in the world, according to the World Health Organisation (WHO, 2020), which shows the true importance of this research. The usage of microbots to remove blood vessel blockages is being investigated (Xie et al., 2020), but many difficulties arise with the employment of these devices in blood vessels. Doutel et al. (2021) characterised the haemodynamic challenges that microbots face in the treatment of CVDs. Further in-depth considerations on the circulatory system's flow characteristics will be discussed in Section 1.2.

The motivation for this work comes from one of these challenges—characterisation of the haemodynamics around swimming microrobots, under the frame of the FCT project HEMOSwimmers - Hemodynamic optimization around 3D swimming microbots (PTDC/EME-EME/30764/2017). The effort to understand the behaviour of blood flow around obstacles has led Rodrigues et al. (2020) to investigate the flow of blood-like polymer solutions around microfluidic cylinders. They reported what appeared to be the onset of a viscoelastic instability upstream of one of the cylinders. The purpose of this work was to complement this recent study, to elucidate the origin for the observed flow behaviour, characterising the potential instability by experimentally quantifying the velocity-vector field around the same cylinder using a viscoelastic, shear-thinning fluid, and a non-shear-thinning elastic fluid.

This thesis is divided into four chapters:

Chapter 1 has begun with a quick mention of the motivation for this thesis and will continue with an introduction to microfluidics, a brief description of blood flows in the circulatory system, and an overview of non-Newtonian flows around cylinders, focusing on recent microfluidic flow studies.

In Chapter 2 we will delve into the rheological characterisation of our working fluids, the preparation of the microfluidic devices and the experimental setup for μ PIV measurements.

In Chapter 3 the obtained results will be shown and discussed.

In Chapter 4 the conclusions gathered from Chapter 3 will be presented and possibilities for future work will be discussed.

1.1 Microfluidics

Microfluidics may be defined as “the science and engineering of systems in which fluid behaviour differs from conventional flow theory primarily due to the small length scale of the system” (Nguyen & Wereley, 2006). Its birth came along with a trend of miniaturisation of mechanical and electronic components that began in the 1950s (Junk & Riess, 2006). These efforts to reduce device size were growing when Richard Feynman presented his famous speech “There’s Plenty of Room at the Bottom” in 1959 (Feynman, 1959)¹. Feynman’s intentions on creating a new branch of technology may be arguable (Junk & Riess, 2006), but the development of silicon-based microelectronics boomed after his speech and has presented an incredible growth ever since (Nguyen & Wereley, 2006). The advancement of micromachining led to the fabrication of the first movable micromechanical parts in 1987, and, in the coming years, microelectromechanical systems, or MEMS (Bao & Wang, 1996). These advances made possible the creation

¹It is noteworthy that Feynman also mentioned in his speech Albert R. Hibbs’s idea of “swallowing the surgeon”, referring to the possibility of “putting the mechanical surgeon inside the blood vessel”. The exact same curious thought that has led us here.

of several crucial devices such as microflow sensors, micropumps, and microvalves in the late 1980s, giving microfluidics a spotlight as a new discipline (Nguyen & Wereley, 2006). The ultimate goal of microfluidics was “to be able to detect biological molecules, and transport, mix and characterise a raw sample, all with one device” (Tabeling, 2005), the complete miniaturisation of an entire laboratory, as if Feynman’s Encyclopaedia (Feynman, 1959) was not ambitious enough. In truth, the expectations for miniaturisation may have risen, at one point, a “sceptical cloud” over the emerging subject, as there had been “outrageous predictions, like microrobots gathering up radioactive particles at toxic sites, and microsubmarines stalking and attacking viruses and cancerous cells while swimming through the bloodstream” (Bao & Wang, 1996). Indeed, quoting Arthur C. Clarke’s famous words, “any sufficiently advanced technology is indistinguishable from magic”. Despite the “microsubmarines” still being under investigation, lab-on-a-chip systems have been constructed and commercialised (Tabeling, 2005) with rather impressive figures (Nguyen & Wereley, 2006). Whitesides (2006) provides a brilliant review of current applications and future expectations for microfluidic systems.

1.1.1 Continuum hypothesis

Before heading to the characteristics of microflows one must wonder if the continuum hypothesis still holds. The continuum hypothesis, as the name suggests, consists on the assumption that the fluid may be treated as a continuum, and that all quantities associated with the fluid are defined everywhere in the flow and vary continuously from point to point (Nguyen & Wereley, 2006). Of course we know that, in truth, all matter is composed of particles and that the assumption is incorrect. Nevertheless, that does not make it invalid, as it yields acceptable results that have been confirmed experimentally and well documented into a massive amount of literature for practical and theoretical applications. This is, in fact, the biggest strength of the continuum hypothesis; but not the only one, as it also simplifies the calculations tremendously, reducing the computational efforts in quite a lot (Nguyen & Wereley, 2006). However, there are limits to the validity of the continuum hypothesis and, besides the issue of continuum breakdown, there is the problem of finding the limit itself. For gases it may be simple; the Knudsen number, Kn , is the ratio between the mean free path, λ' (the index is used here to differentiate from the relaxation time λ to be discussed later on), and the characteristic length scale, L , and it is a measure of flow rarefaction. It is also used to quantify the barrier of the continuum hypothesis. There are four intervals for the Knudsen number value that aid us in dealing with rarefied flow (Nguyen & Wereley, 2006):

- $Kn < 10^{-3}$ (Navier–Stokes equation and no-slip condition are valid).
- $10^{-3} < Kn < 10^{-1}$ (Navier–Stokes equation is valid but slip boundary conditions are required).
- $10^{-1} < Kn < 10^1$ (Transitional flow regime).
- $Kn > 10^1$ (Free molecular flow).

Thus, for Knudsen numbers lesser than 10^{-3} the unaltered continuum hypothesis is valid i.e., Navier–Stokes equation and no-slip. From then on, additional precautions must be taken to ensure the correct modelling.

Briefly attending to the no-slip boundary condition, it assumes that the velocity tangential to the fluid-solid interface is zero, as the fluid “sticks” to the wall. It has been widely used for its simplicity and agreeable results with macroscopic experiments (Nguyen & Wereley, 2006). However, it is only an assumption; Lauga et al. (2007), provide a rich review on slip, presenting several means of analysis that have been used for slip measurements, gathering experimental results for Newtonian fluids and reviewing physical parameters that influence slip. Experimental studies confirm that the no-slip boundary condition does hold for microflows in Newtonian fluids, but may breakdown at smaller scales (Lauga et al., 2007). For gases, however, slip must be considered, in accordance to the Knudsen number, and many microflows are often in slip and transitional regimes, forcing us to apply slip boundary conditions (W.-M. Zhang et al., 2012). Non-Newtonian fluid flows present another category where slip has been encountered (Lauga et al., 2007). The standard slip boundary condition used today was introduced by Navier in 1823 (W.-M. Zhang et al., 2012) and it consists on considering a relative velocity at which the fluid slips along the wall that is proportional to the velocity gradient, and, thus, to the shear stress (Nguyen & Wereley, 2006).

Returning to the validity of the continuum hypothesis, for liquids the definition of its limit is still unclear and the literature diverges on the use an equivalent Knudsen number. As liquid molecules do not have a mean free path, Sharp et al. (2000) propose the lattice spacing, δ , dependent on molar volume and Avogadro’s number, as a substitute, and keeps the value of 10^{-3} as the limit for the continuum hypothesis for liquids. The hypothesis for liquids may also breakdown from excessive shear, Nguyen & Wereley (2006) mention this issue but disregard its concerns at the microscale. Despite the different stands on the Knudsen number, or more clearly, its equivalent, it is generally accepted that the continuum hypothesis should breakdown by the nanoscale (Czerwinska, 2009). So we can assume that for the general microflow the continuum hypothesis is applicable (Nguyen & Wereley, 2006).

1.1.2 Dimensionless numbers

Dimensionless numbers are one of the biggest tools of a fluid mechanics analyst—they give us critical information about the flow at hand from very simple expressions relating fluid properties and flow characteristics. The most widely used dimensionless number in the usual macroscale is the Reynolds number, which has tremendous importance on macroscopic flows (White, 2006), but it also gives us important information on microflows. The Reynolds number conveys the ratio between inertial and viscous forces. It is used to determine the transition between laminar (low Reynolds numbers) and turbulent flow (high Reynolds numbers) (White, 2006) and is given by

$$Re_L = \frac{\rho V L}{\eta}, \quad (1.1)$$

where ρ and η represent the fluid density and viscosity, respectively, V is the flow velocity and L is the characteristic length scale.

Other dimensionless numbers grant us insight into non-Newtonian effects. Here we shall discuss the Deborah, Weissenberg and elasticity numbers. Starting from the first, the Deborah number, De , is the ratio between relaxation time, λ , and time of observation,

T (Galindo-Rosales et al., 2014) as seen in the following expression:

$$De = \frac{\lambda}{T}. \quad (1.2)$$

Its concept may be, at first sight, awkward to grasp, but Poole's examples of silly putty and steady flows grant an idea of its meaning (Poole, 2012). Reiner's original paper (Reiner, 1964) also presents a very interesting image—the “flowing mountains before the Lord” may take us aback momentarily, but it underlines perfectly the importance of time scales, as even solids may behave as fluids in a large enough time-scale, and fluids behave as solids in an equally short time-scale (Poole, 2012). This can be applied to our current use of the Deborah number, partaking in the quantification of non-Newtonian behaviour. Reduced values for the Deborah number translate into a purely viscous response of the fluid, while large values represent a viscoelastic reaction. However, the Deborah number alone is not sufficient to fully characterise effects due to viscoelasticity (Poole, 2012).

The Weissenberg number and the Deborah number are frequently used as synonyms, but they represent different effects (Poole, 2012). The Weissenberg number, Wi , represents the ratio of elastic to viscous forces and is given by the following expression (Galindo-Rosales et al., 2014):

$$Wi = \lambda \dot{\gamma} = \lambda \frac{V}{L}, \quad (1.3)$$

where λ remains the relaxation time and $\dot{\gamma}$ is the characteristic rate of deformation that can be estimated through the ratio of the flow velocity, V , to the characteristic length, L (Galindo-Rosales et al., 2014). The Deborah number is calculated from an observation time, while the Weissenberg number uses the deformation rate. Poole (2012) clarifies the distinction between the two dimensionless numbers by their derivation from dimensionless analysis for a viscoelastic flow and notes on the conditions for which the two numbers coincide. On a general note, both numbers are measures of the strength of the fluid's viscoelastic response in opposition to a Newtonian, viscous behaviour.

Lastly, the elasticity number, El , can be interpreted as the ratio of relaxation time to diffusion time or the ratio of elastic to inertial effects and is given by (Czerwinska, 2009; Poole, 2012)

$$El = \frac{\lambda \eta}{\rho L^2} = \frac{Wi}{Re}. \quad (1.4)$$

Interestingly, the elasticity number depends only on material and geometric properties; kinematic properties having, thus, no influence on it (Mckinley, 2005a).

The reduction of the length scale clearly shows its influence on the flow characteristics through dimensionless numbers. The Reynolds number scales proportionally with the length scale. Accordingly, for the majority of microdevices, $Re \ll 1$ (Czerwinska, 2009), which translates into viscous domination over inertial effects and, consequently,

typically laminar flows. Thus, microflows are generally considered as laminar, despite the existence of particular cases where turbulent regime may be achieved for a variety of Reynolds numbers (Nguyen & Wereley, 2006; Sharp et al., 2000). The Weissenberg and elasticity numbers scale with L^{-1} and L^{-2} , respectively. Thus, reducing L , as to the microscale, leads to large Wi and El , which translates into viscoelastic domination over inertial effects. So, fluids that tend to behave as almost Newtonian at the macroscale, may have significant non-Newtonian capabilities at the microscale. These particularities of microflows are actually quite useful for rheological characterisation of certain fluids. Mckinley (2005a) mentions “low viscosity” and “weakly elastic” fluids—dominated by inertial effects at the macroscale, their non-Newtonian behaviour is extremely hard to quantify by the usual rheological means, but having them in microflows allows inertia to be disregarded and, as viscoelastic effects gain sovereignty, we may proceed to the needed measurements.

1.2 Haemodynamics

The circulatory system consists in a huge and extremely complex network of interconnecting, size-variable vessels, in which a complex viscoelastic fluid flow is driven by an incredible bio-pump. The system is, if nothing else, brilliant, and moreover because the working principle is fairly simple. Explained very crudely: the heart pumps the blood into large-scale arteries, from which it is subdivided into progressively smaller-scale vessels, reducing its velocity and pressure in order to allow for cell-beneficiary exchanges with the tissues all over the body. From there, it follows the inverse path, flowing into progressively larger veins until it reaches the heart. This composes the systemic circulation, but the journey is not yet over; from the heart, the blood is pumped into the lungs for gas exchanges and comes back—the pulmonary circulation. From a fluid mechanics approach, we are looking at pulsatile flow (from the heart’s intermittent pumping) on multi-scale, non-rigid vessels and variable flow conditions.

Starting from the heart, it acts as a pressure pump, contracting its chambers to push the blood into the arteries (having valves within the cardiac chambers in order to prevent back-flow), and then relaxing; creating pressure and flow pulses as it contracts and eases. These pulses can be sensed all over the body (Doutel et al., 2021), however, further away, in the smaller vessels, the flow is almost constant (Martínez-Aranda et al., 2016; Rodrigues et al., 2020). Considering the blood vessels, they are generically non-rigid, being able to deform at high pressures and, even though it is a useful characteristic in order to deal with pressure variations, it may lead to grave complications, namely aneurysms (Campo-Deaño et al., 2015). Blood vessel diameter varies widely throughout the body, from 25 mm (aorta) to merely 8 μm (capillaries) (Müller et al., 2008). The flow conditions also vary within the vessels, from velocity and diameter differences, with Reynolds numbers ranging from 3000 in the aorta and vena cava, to 0.002 in the capillaries (Doutel et al., 2021). Concerning capillaries in particular, their extremely small diameters force us to take increasing precautions such as the need to consider particle flow (instead of a generally homogeneous solution as blood components may have almost the same length scale as the capillaries themselves, i.g., red blood cells) (Robertson et al., 2008), and the aggravation of non-Newtonian behaviours.

1.2.1 Whole blood properties

As has been mentioned before, blood is a complex, viscoelastic fluid. It is a suspension of cellular elements, such as thrombocytes (platelets), leukocytes (white blood cells, or WBCs) and erythrocytes (red blood cells, or RBCs), in plasma, a Newtonian aqueous solution (90-92 wt% water) of minerals, proteins, hormones, among others (Robertson et al., 2008; Campo-Deaño et al., 2015; Rodrigues et al., 2020).

Red blood cells are, as the name suggests, cells, filled with an aqueous solution of hemoglobin, which is the protein that gives blood its red colour and is responsible for the transport of oxygen and carbon dioxide. RBCs are highly flexible, and, at their undisturbed shape, which is a biconcave discoid, they have a diameter of 6 to 8 μm (around the size of the smallest capillaries). They have the largest concentration in blood, diverging for healthy women and men approximately at 40 and 45 %, respectively (being this volume concentration of RBCs named hematocrit - Hct) (Robertson et al., 2008). They are the main responsible party for blood's shear-thinning behaviour, as they aggregate at low shear rates, in rod-shaped structures called *rouleaux*, and deform and align with the flow at large shear rates. Both the aggregation into *rouleaux* and the deformation and alignment of the RBCs are reversible. This shear-thinning is not, however, all that straightforward in the human body. There is a large time-dependence of RBCs in order to aggregate and deform, and, therefore, shear-thinning may not be relevant in various regions of the body as flow properties vary widely (Robertson et al., 2008). This viscosity time-dependence, or thixotropy, is one of blood's defining traits, requiring special attention from either experimental work and numerical modelling (Horner et al., 2019; Beris et al., 2021). Moreover, blood viscosity is also dependant of several other factors such as temperature, blood component concentrations, plasma viscosity, subject diseased state and gender, among others (Robertson et al., 2008). In order for us to gain some insight into blood's viscous behaviour, in Figure 1.1 are shown whole blood viscosity values, η , (against shear rate, $\dot{\gamma}$), representing the mean of two data sets of healthy donors: a female (Hct = 0.36) and a male (Hct = 0.46), both at 37°C, from Valant et al. (2011). Additionally, error bands were plotted to represent the standard deviation of the mean values.

As mentioned before, blood is a viscoelastic fluid, and again, RBCs are the main responsible components for this behaviour, as their three-dimensional structures are able to store and release elastic energy. Individual RBCs also influence blood's viscoelasticity due to their individual deformations, but to a much lesser degree (Robertson et al., 2008). Fluid viscoelasticity is usually described through a storage modulus, G' , and a loss modulus, G'' . These moduli can be measured experimentally through specific rheological essays (usually Small Amplitude Oscillatory Shear, or SAOS) and represent the fluid's ability to store and release elastic energy, and are associated with elasticity and viscosity, respectively. G' represents the fluid response that is in phase with the strain rate, while G'' , the response that is out of phase. A Newtonian fluid has $G' = 0$ and $G'' = G$, while, on the other hand, an elastic solid, responding by Hook's law, has $G' = G$ and $G'' = 0$. A viscoelastic fluid has no null components, having both the capability to store and release elastic energy. Whole blood's G' and G'' obtained by Campo-Deaño et al. (2013) through one-point passive microrheology at 27°C can be observed also in Figure 1.1, plotted against rotational velocity, ω . The loss modulus' domination throughout the whole range corresponds to a typical liquid behaviour, however, blood's elasticity is

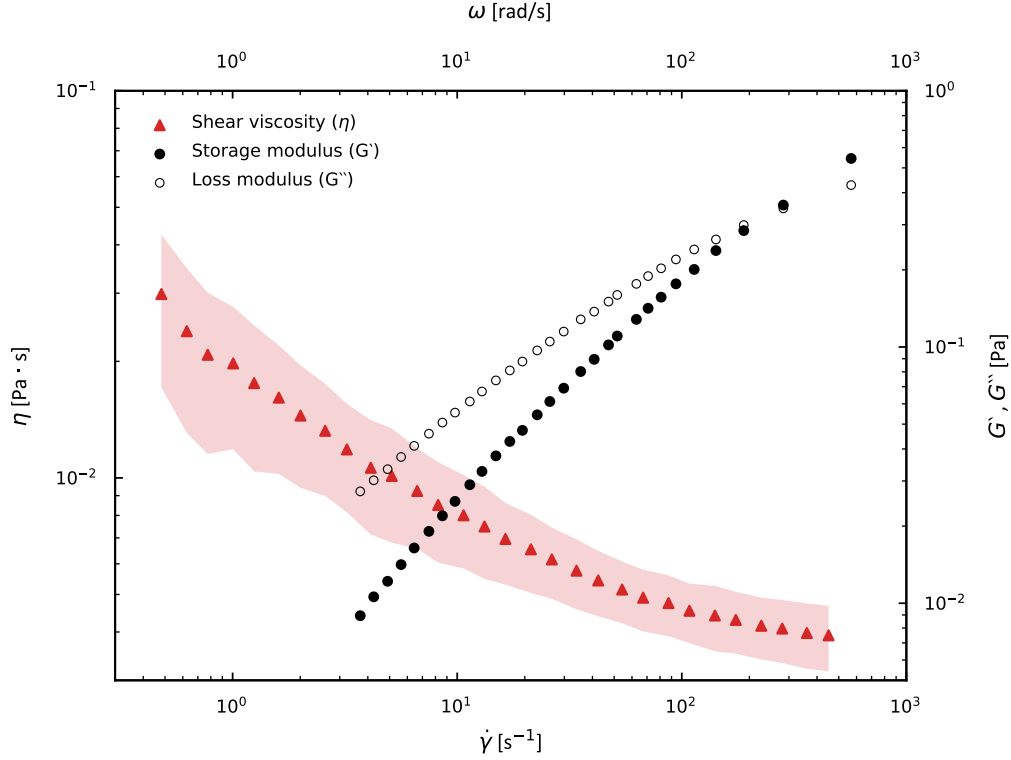


Figure 1.1: Whole blood rheological properties: Mean viscosity curve plotted from data from Valant et al. (2011); storage and loss moduli from Campo-Deaño et al. (2013).

of relevance, as the storage modulus is not negligible.

1.3 Non-Newtonian flows past confined macroscopic cylinders

Flow past a confined cylinder has long been a relevant problem due to its significance for several industrial applications. This specific flow has been studied extensively for Newtonian fluids, however, it gains particular significance for non-Newtonian fluids, such that it became a rheological benchmark problem, specifically for a blockage ratio of 0.5 (Brown & McKinley, 1994). Industrially, it can be found in a number of applications, such as flows in heat exchangers, polymer processing operations and mud flows, among others (Nejat et al., 2011). The need for such complex flows, apart from industrial motivations, advents from the lack of information yielded by the usual free shear flows in rheological analysis, which return unreliable results when considering more complex flows of non-Newtonian fluids (Baaijens et al., 1995). Baaijens et al. (1995) grant a brief review of the literature up until 1995 and claim that the effect of viscoelasticity on the velocity field is influenced by four key aspects: the position of the walls, the degree of elasticity in the flow, the importance of inertia and the rheological behaviour of the liquid in elongational flow.

Considering the actual flow, far upstream of the cylinder there is essentially plane Poiseuille flow and, when approaching the cylinder, the streamlines are forced to diverge into the gaps between the cylinder and the walls of the channel, compressing the fluid elements at the forward stagnation point. At the gaps, the velocity profiles are steeper, due to the no-slip boundary conditions at both the walls and the cylinder

face, leading to shear rates that may rise to very high values, depending on the flow rate and the blockage ratio (the ratio between the cylinder diameter and the channel width: $\beta = D/W$). After passing through the gap, the fluid elements are expanded and, at the downstream stagnation point, they are subjected to large residence times. For non-Newtonian fluids, elastic stresses may lead to viscoelastic or purely elastic instabilities, which are extremely important to the study of non-Newtonian flows, as they may massively diverge flow behaviour from the Newtonian case, creating interesting effects. Viscoelastic behaviour can aggravate or mitigate already existing flow instabilities occurring due to other phenomena, or create entirely new ones as consequences of viscoelastic forces, leading to unsteady flows when, for Newtonian fluids, the flow would be stable (Larson, 1992). Larson (1992) presents a review of viscoelastic instabilities in relevant flows. For flow past a cylinder, the author mentions the encounter of McKinley et al. (1991) with a three-dimensional instability in the wake of a cylinder for a viscous Boger fluid. A lot of work has been done on viscoelastic instabilities in flows around cylinders, McKinley et al. (1993) studied the onset of a purely elastic instability in the wake of the cylinder for several blockage ratios in the flow of an highly elastic Boger fluid, mentioning the large residence times of the polymer molecules in the two stagnation points, creating large molecule extensions and elastic stresses, responsible for discrepancies of the flow behaviour when compared to the Newtonian case. The authors also present a literature review on experimental and numerical studies. More specific effects of non-Newtonian characteristics on the flow will be discussed when reviewing the current literature on flows around microfluidic cylinders in the next Subsection.

The majority of viscoelastic flows are studied in low to moderate Reynolds numbers and creeping flow. However, there are some studies of Newtonian and non-Newtonian turbulent and transitional flow past confined cylinders—Richter & Naudascher (1976) experimentally studied the influence of wall effects on confined Newtonian flow around the critical Reynolds number, and Kanaris et al. (2015) numerically studied the transition to turbulent flow for a Bingham fluid. Usually, confined cylinder experiments and models consider two-dimensional, pressure driven flow, as a Poiseuille flow. Nevertheless, three-dimensional analysis gains importance in the study of wall effects, the influence of the channel's and the cylinder's aspect ratios ($\alpha = H/W$ and $\Lambda = \alpha/\beta = H/D$, respectively) and other three-dimensional phenomena such as turbulence. Ribeiro et al. (2014) conducted an experimental and numerical study for aspect ratios, Λ , of 16, 8 and 2 to examine the influence of three-dimensional effects on viscoelastic flows (an analogous study was also conducted for Newtonian flows by Ribeiro et al. (2012)). Kanaris et al. (2011) investigated the confinement effects on wake instabilities via two- and three-dimensional numerical simulations.

The aforementioned work of Ribeiro et al. (2014), was not conducted at the microscale, were we will be conducting our experiments, however, it is noteworthy and shall be discussed further in depth. As mentioned before, the authors studied three-dimensional effects on the flow by varying Λ . The test fluids were two Boger fluids, which consisted in aqueous solutions of glycerine (85 wt%) with 1% of NaCl and 200 and 400 ppm of polyacrylamide (PAA), and a shear-thinning fluid consisting in an aqueous solution of 1000 ppm of PAA. The flow was analysed from creeping flow to low Reynolds numbers ($0.008 < Re < 3.2$) and for a range of Deborah numbers: $0.025 < De < 0.99$. For the shear-thinning fluid, a steady asymmetric flow instability was reported upstream of the cylinders for all aspect ratios, however, the critical Deb-

orah number at which it onset, De_c , varied, increasing with the reduction of Λ due to viscous effects at the walls. Thus, the confinement of the flow had a dampening effect on the instability. Furthermore, a time-dependent flow was observed for low Reynolds numbers, and, therefore, was not an effect of inertia, but of elasticity of the fluid. Also, while maintaining fore-aft symmetry, up and downstream wakes, aggravating with De , were also reported when comparing to the Newtonian cases. For the Boger fluids, no asymmetries were found, being either fore-aft or lateral.

1.4 Recent literature review on non-Newtonian flow past confined microfluidic cylinders

Flow around microscopic cylinders was surely the next logical step in non-Newtonian fluid dynamics analysis. Besides the macroscopic analogue being the already established benchmark problem, bringing this flow into the microscale is not only advantageous due to the predominant laminar flow, but also due to the particular aggravations of non-Newtonian effects and the current importance of microfluidics. As so, it has gained popularity in the last few years and several studies have been made on non-Newtonian flows past confined microfluidic cylinders. In this Subsection, results obtained by some of the most recent studies are gathered and discussed.

Kenney et al. (2013) studied low-viscosity Boger fluids (composed of polyethylene oxide (PEO) dissolved in glycerol/water solutions in mass concentrations of 0.3, 0.1, 0.05 and 0.043 wt%) in a microfluidic geometry with an aspect ratio (α) of 0.35 and a blockage ratio (β) of 0.65, being the cylinder diameter of 65 μm . The shown results for the largest elasticity fluid ($El = 88$) ranged: $0.1 < Re < 20$ and $8.4 < De < 1672$. For $Re \sim 1$, a downstream instability onset with unsteady, disordered and crossing streamlines. The authors relate this instability with the purely elastic one found for creeping flow by McKinley et al. (1993). Increasing Re to approximately 3, a second instability, now upstream of the cylinder, causes a streamline “pinch” that aggravates even further with the increase of Re , incorporating more streamlines and reducing the width of the wineglass-shaped flow and disconnecting the forward stagnation point from the cylinder face. This instability was found to be cyclic, growing and collapsing back into the cylinder face with time and also responsible for a very significant increase of the downstream wake length. The authors ascribe the onset of both instabilities primarily to enhanced extensional stresses and the large confinement and blockage of the channel.

Galindo-Rosales et al. (2014) review experimental works on elastic instabilities for several flows with a strong extensional component, including flow around a confined cylinder, on which we will focus. The aspect ratio (Λ) was set for: 2.0, 1.0 and 0.55, and the blockage ratio (β): 0.25, 0.5 and 0.75, having thus nine distinct geometries with three cylinder diameters (158, 105, 55 μm). A Boger fluid was studied, composed of an aqueous solution of glycerol and 200 ppm of PAA with 1 % NaCl for a wide range of Wi . For creeping flow, a sequence of flow transitions was found as Wi increases: Newtonian-like flow, steady divergent streamlines, periodic flow and chaotic flow. The critical conditions for the transitions were found to be the aspect and the blockage ratios. As Wi increases, the divergent streamlines upstream become more pronounced until the onset of an elastic instability near the forward stagnation point. Observing the flow

pattern images provided by the authors for $\beta = 0.5$ and $Wi \sim 30$, we can observe a similar “pinching” effect to that reported by Kenney et al. (2013), being aggravated with decreasing aspect ratio.

Shi et al. (2015) employed water and glycerol solutions with dissolved PEO of different molecular weights, as low-viscosity Boger fluids on channels with aspect ratios: $\alpha = 0.175, 0.35, 0.5$ and 0.7 and blockage ratios: $\beta = 0.55, 0.65$ and 0.75 . Having six cylinders with diameters ranging from 110 to $300 \mu\text{m}$. The Reynolds number was varied from low to intermediate values ($0.001 \lesssim Re \lesssim 100$), the Deborah number ranged equally widely from low to high values ($0.001 \lesssim De \lesssim 1000$) and El took twenty four values ranging from 0.1 to 177 . The authors provide an analysis of a significant amount of experimental results and intended to provide a characterisation of the onset of a downstream instability, and a posterior upstream instability which provokes the already mentioned wineglass shaped flow encountered in their earlier work (Kenney et al., 2013). They found that a viscoelastic Mach number (representing the ratio of the local flow speed to the velocity of the viscoelastic waves), $Ma = \sqrt{ReDe} = 1$ appropriately described the onset of the first instability. The onset of the upstream instability, however, was found harder to characterise but it occurred at $Ma \sim 10$. The study’s results led to the conclusion that it was due to velocity perturbations caused by the downstream instability and there were connections to the aspect and blockage ratios and El . In fact, they found that decreasing the aspect and increasing the blockage ratios reduced the value of Ma at which the second instability occurred. Moreover, larger elasticity numbers increased this effect of the confinement ratios.

Nolan et al. (2016) analysed the flow of three weakly elastic, shear-thinning aqueous solutions of PAA of $25, 50$ and 100 ppm around a cylinder of $100 \mu\text{m}$ of diameter, over a Reynolds and Weissenberg numbers range of $5 < Re < 300$ and $20 < Wi < 10^3$. The channel’s width and height were 500 and $200 \mu\text{m}$, respectively; leading to $\Lambda = 2$ and $\beta = 0.2$. The authors reported long downstream wakes (with flow separation and vortex shedding suppression) and detachments of the forward stagnation points into further upstream. They mention the extended wake length being a consequence of fluid elasticity, which goes accordingly to the reports of Kenney et al. (2013) for Boger fluids. The detachment of the stagnation point was progressively suppressed by inertial effects for the 25 ppm solution, approximating the cylinder face with increasing Re . For the other two solutions, the stagnation point’s distance from the cylinder increased with Re before collapsing back into the cylinder. This drift of the stagnation point was accompanied by a buckling-type instability that was found immediately upstream of the cylinder and was concluded as a consequence of shear-thinning effects and viscosity fluctuations, being also enhanced by increasing El . A viscoelastic Mach number, $Ma = 1$, was found to be representative for the buckling onset.

Zhao et al. (2016) investigated the flow of a strong shear-thinning wormlike micellar solution (WLM) on geometries defined by $\alpha = 0.167$ and $\beta = 0.5, 0.67$ and 0.83 (attained by variation of the cylinder’s diameter; respectively: $300, 400$ and $500 \mu\text{m}$). The Weissenberg, Reynolds and elasticity numbers ranged, respectively: $1 \lesssim Wi \lesssim 10^5$, $10^{-7} \lesssim Re \lesssim 10$ and $10^4 \lesssim El \lesssim 10^7$. For the lowest blockage ratio, 0.5 , the flow remained Newtonian-like up until $Wi \sim 50$. Increasing the flow rate, divergences from the Newtonian flow began to appear, such as bending streamlines upstream of the cylinder, having a resemblance with the asymmetric flow division around the cylinder reported by Ribeiro et al. (2014) and the buckling flow of Nolan et al. (2016). Then, a recirculation

vortex upstream of the cylinder formed and shifted further upstream with increasing flow rate. Following came bending streamlines and lateral flow asymmetry in the wake of the cylinder. For even greater flow rates, the upstream vortex collapsed into unsteady, three-dimensional flow. For higher blockage ratios, these flow transitions occurred similarly in a well defined sequence, however, at lower critical Weissenberg numbers. The unitary viscoelastic Mach number did not prove useful for predicting the onset of instabilities, as here $Ma \ll 1$. The authors mention that the upstream instability appearing before the downstream one goes contrarily to the results obtained by Kenney et al. (2013) and Shi et al. (2015), who studied Boger fluids. However, similar results were attained by Ribeiro et al. (2014) and Nolan et al. (2016), as mentioned before. Zhao et al. attributed their found upstream instability to stresses as the fluid accelerates into the gaps, and the downstream instability to extensional deformations at the further stagnation point and influenced by the upstream unsteady flow.

Haward et al. (2018) fabricated two microchannels, both defined by $\alpha = 5$ and $\beta = 0.1$ with cylinder diameter of $40 \mu\text{m}$, being a good approximation to two-dimensional flow. One of the channels contained two axially-aligned cylinders and the other, a single cylinder. We will focus on the results attained for the latter. The fluid used by the authors was a solution of 6.9 MDa atactic polystyrene dissolved in dioctyl phthalate at a 0.07 wt% concentration—a practically non-shear-thinning fluid over the analysed range. The authors presented results for $1.35 \leq Wi \leq 54.2$, $Re \leq 0.023$ and $El \sim 2400$. The flow remained Newtonian-like for low Wi . As Wi increased, a downstream wake developed and grew with Wi up to an extremely long length (up until 300 cylinder radii for $Wi = 54.2$ from birefringence measurements). An upstream wake was also reported, as the velocity immediately behind the cylinder decreased, and around the cylinder increased. An upstream instability was also noted, particularly for higher Weissenberg numbers ($Wi = 54.2$), from the distortion of streamlines. Haward et al. mentioned that the stretching of the polymer around the cylinders surrounds it in viscosified fluid, practically enlarging its radius, shifting the stagnation point upstream with increasing Wi . Also, downstream of the cylinder, the extensional region of the flow is replaced by shear-dominated flow with increasing Wi .

Haward et al. (2019) studied the flow of a wormlike micellar solution (WLM), that exhibited strong shear-thinning, around a confined microfluidic cylinder on an identical geometry to the previous analysed article (Haward et al., 2018). Creeping flow was maintained, with $Re < 10^{-4}$ for the maximum flow rates, and Wi was varied from 1.9 up to 3750, with $El \sim 2 \times 10^8$. For low Wi the flow remained symmetric until the development of an extended wake downstream of the cylinder. For $Wi = 93.8$ a steady lateral asymmetry in the flow was developed, with the fluid flowing primarily over one side of the cylinder, being the preferential side chosen seemingly at random. The asymmetry is progressively aggravated with increasing Wi until virtually all the fluid passes through one side ($Wi = 150$). For the $Wi = 1875$ the flow partially regained symmetry. The authors utilise an asymmetry parameter, I , to quantify the intensity of the flow asymmetry. I remained zero up until a critical Weissenberg value (in this case $Wi_c \sim 61$); from there, it grew with Wi . For $Wi \gtrsim 130$, the flow was found to be time-dependent. Additionally, the authors also tested for hysteresis, finding no relevant divergences. They also presented the possibility of the onset of the asymmetric flow being due to a random fluctuation in the high elongational stress wake region, leading to an imbalance of flow rates, and thus shear rates which is then maintained and aggravated by the strong shear-

thinning behaviour of the fluid. The partial resymmetrization of the flow was thought to be a consequence of the breakage of WLM chains.

From the encounter with the axial flow asymmetry reported in the previous article, Haward et al. (2020) pursued to study it more in-depth through several solutions of high molecular weight (18 MDa) hydrolysed polyacrylamide dissolved in deionized water in different concentrations (ranging from 50 to 3000 ppm on the tested fluids), therefore, with different degrees of shear-thinning and different characteristic times. The geometry employed was the same as in the authors' previous studies (Haward et al., 2018, 2019). The Reynolds number in their reported results did not exceed 0.05 and the Weissenberg number was varied widely and the elasticity number: $10^3 \lesssim El \lesssim 10^8$. For the lowest concentrations (50 and 100 ppm) no asymmetry in the flow was found. With increasing Wi , an extended wake was reported, being a consequence of fluid elasticity, resulting in high elastic stresses and an increase in extensional viscosity. For an intermediate concentration of 200 ppm, flow asymmetry was found for middling Wi values, collapsing back into symmetric flow for large Wi . The same occurred for 500 ppm, but there was only a partial recovery of symmetry at high Wi . For 1000 ppm, the asymmetry onsets again for intermediate Wi and it remained intense up to the last Wi tested. These results go accordingly to the previously obtained (Haward et al., 2019). The same asymmetry parameter, I (Haward et al., 2019) was analysed. For the lowest concentrations (50 and 100 ppm) it remained low for all Wi . From 200 to 500 ppm, I reached a maximum value before dropping at higher Wi . For 1000 ppm, I reached a high maximum value before reducing very slightly. Finally, for 3000 ppm, it reached a very large value and did not drop, remaining high even to the largest Wi tested. The critical value of the Weissenberg number, Wi_c at which asymmetric flow onsets was found to decrease with polymer concentration. These results led to the conclusion that this flow asymmetry is not a particular result for WLMs but a more general phenomenon resulting of elasticity and shear-thinning, going accordingly to the studies of Ribeiro et al. (2014) and Nolan et al. (2016).

Varchanis et al. (2020) set out to compute numerically, and confirm experimentally, 2D simulations of creeping flow of three different fluid types. The first fluid was an inelastic, shear-thinning fluid—a solution of 0.1 wt.% of xanthan gum dissolved in a 82 wt.% mixture of glycerol and water (numerically modelled by a Carreau–Yasuda model). The second fluid was a non-shear-thinning, elastic fluid—a solution of 0.2 wt.% of PEO (4 MDa) dissolved in a 50 wt.% mixture of glycerol and water (modelled by a finitely extensible non-linear elastic dumbbell model). The final fluid was a both elastic and shear-thinning fluid—a solution of 0.5 wt.% of PEO (8MDa) dissolved in pure water (modelled through a linear Phan–Thien–Tanner model). The physical microfluidic geometry was characterised through $\alpha = 5$ and $\beta = 0.1$ (the same geometry employed in the studies of Haward et al.), which was utilised as it stands as a good approximation to two-dimensional flow in microchannels, particularly useful when comparing to numerical 2D simulations (Haward et al., 2018; Varchanis et al., 2020). The Reynolds number was kept at $Re \leq 0.25$. For the first test fluid, the flow remained essentially symmetric, either laterally or fore-aft for the entire test range. For the second fluid, extended wakes were found for higher Weissenberg numbers from both methods, but no relevant lateral asymmetry was found. For the final, elastic and shear-thinning fluid, there was a good agreement between the simulation and the experimental results, showing downstream wake formation for low Wi and lateral asymmetry for larger Wi . The

preferential path of choice around the cylinder was chosen by the fluid seemingly at random, as reported before by Haward et al. (2019). The numerical simulations reported an increasing bending of the downstream wake with Wi (bending towards the side opposite to the preferential path around the cylinder). The variation of the asymmetry parameter, I with Wi was also obtained through the numerical model, reasonably well fitting the experimental data. The authors also concluded numerically that the independent increase in shear-thinning and elasticity result similarly in a reduction of the critical Weissenberg number, Wi_c , for the asymmetric flow onset and also in an increase of $I_{\max}$, effectively aggravating the asymmetry, in accordance with the results obtained by Haward et al. (2020). However, both elasticity and shear-thinning were found to be necessary for the asymmetry to occur at all. Lastly, the authors found that increasing β led to a reduction of Wi_c and, for this Weissenberg value, it was found that the gap wall shear rate was similarly located in the middle of the shear-thinning curve, $\dot{\gamma} \sim 100 \text{ s}^{-1}$. Additionally, the authors plotted I against β in bifurcation diagrams for different values of Wi and found that increasing Wi enlarges the range of β for which there is flow asymmetry, reducing both the value of β for the asymmetry onset and the value at which there is resymmetrization of the flow (in accordance with the results obtained by Haward et al. (2020)). Also, the value of $I_{\max}$ is increased with Wi .

Rodrigues et al. (2020) experimentally investigated the flow of blood analogues—one Newtonian and five, weakly elastic shear-thinning fluids—through a geometry characterised by $\alpha \sim 0.37$ and $\beta \sim 0.28$, similarly for a circular and an elliptic (1:4 aspect ratio) cylinder. The test fluids consisted of xanthan gum in aqueous solutions of 52 wt.% dimethyl sulfoxide. The concentrations of xanthan gum were different for each fluid: 100, 250, 500, 750 and 1000 ppm, presenting different relaxation times and degrees of shear-thinning. The Reynolds number was varied over a range of $0.05 \lesssim Re \lesssim 100$, the Weissenberg number: $0.05 \lesssim Wi \lesssim 1500$ and the elasticity number ranged: $1.97 \lesssim El \lesssim 31.8$. They reported streamline divergence upstream and stronger streamline curvature downstream of both cylinders than for the Newtonian blood analogue, due to strong compressional flow leading to the forward stagnation point and extensional flow in the downstream wake, which was found to be increasingly long with elasticity. An absence of downstream flow separation and vorticity was noted for all viscoelastic fluids (similarly to what was reported by Nolan et al. (2016)) and, additionally, there was no drift of the leading stagnation point further upstream. A slight lateral flow asymmetry was found for $Re < 1$ (similar to the buckling reported by Ribeiro et al. (2014) and Haward et al. (2020), as the authors mention) and a viscoelastic Mach number near unity (corresponding to shear rates in the most shear-thinning range of the fluid), only for the most elastic flow, decaying for higher Re . This goes accordingly to the reported findings of Haward et al. (2020) and Varchanis et al. (2020). Extended downstream wakes and earlier flow deceleration upstream, aggravated by elasticity, were also reported.

Our goal is to study the aforementioned flow-asymmetry-inducing instability. From this literature review we gather that it is a result both of elasticity and shear-thinning and the reduction of one can be compensated by the enlargement of the other, always being necessary to have a degree of both (Varchanis et al., 2020), which is supported by having no reports of lateral flow asymmetry found for Boger fluids. The flow confinement is also a relevant parameter, as increasing channel confinement, represented by a decrease of the aspect ratio, has a dampening effect on the instability through viscous domination

at the walls (Ribeiro et al., 2014). The channel blockage provoked by the cylinder affects the degree of asymmetry as well, as increasing the blockage ratio leads to a reduction of the critical Weissenberg number at which asymmetry onsets and to a increase of the maximum asymmetry achievable (Varchanis et al., 2020). This instability seems to onset at approximately unitary values of the viscoelastic Mach number, when the speed of the flow starts to surpass the speed of elastic waves (Nolan et al., 2016; Rodrigues et al., 2020). However Zhao et al. (2016) reported a similar instability for very low values of Ma , believing that it was due to high stresses as the fluid accelerates into the gaps. Another possibility for the driving mechanism for the onset of the instability is a random fluctuation on the high extensional stress wake, leading to an imbalance of flow rates on either side of the cylinder, and, consequently, an imbalance of shear rates, then, the fluid's shear-thinning feeds the flow asymmetry by creating a low-flow-resistance path (by viscosity differences) on one of the cylinder sides (Haward et al., 2019; Varchanis et al., 2020; Rodrigues et al., 2020).

Chapter 2

Methodology

In this chapter, we shall discuss the experimental steps taken towards quantifying the velocity-vector fields. This does not only relate to μ PIV alone, but to fluid rheological characterisation, microdevice fabrication and flow parameters definition as well.

2.1 Test fluids

The first step was the preparation of our test fluids, which were, as mentioned before, a (essentially) non-shear-thinning elastic fluid and a viscoelastic shear-thinning fluid. We chose to use two poly(ethylene oxide) (PEO) solutions. The non-shear-thinning fluid (from now on named as PEO4) consisted in 2000 ppm of 4 MDa PEO dissolved in a mixture of 50 wt% glycerol in distilled water. The shear-thinning fluid (named PEO8) was composed of 5000 ppm of 8 MDa PEO dissolved in pure water. These are the same fluids used by Varchanis et al. (2020). Additionally, 50 ppm of sodium azide (SA) were added to both solutions, in order to guaranty their purity from microorganisms, not influencing the rheological properties of neither fluid at this concentration.

The masses of each fluid component were obtained as follows:

$$m_{\text{PEO}} = \frac{\text{ppm}_{\text{PEO}}}{10^6} m_{\text{Total}} , \quad (2.1)$$

$$m_{\text{SA}} = \frac{\text{ppm}_{\text{SA}}}{10^6} m_{\text{Total}} , \quad (2.2)$$

$$m_{\text{Total}} = m_{\text{PEO}} + m_{\text{SA}} + m_{\text{Solvent}} . \quad (2.3)$$

Whereas for PEO8 the solvent is pure water alone, for PEO4 it is a mixture of water and glycerol in equal mass proportions. For the latter, the solvent mass was computed as follows:

$$m_{\text{Solvent}}^{\text{PEO4}} = m_{\text{Water}}^{\text{PEO4}} + m_{\text{Glycerol}}^{\text{PEO4}} , \quad (2.4)$$

$$m_{\text{Water}}^{\text{PEO4}} = m_{\text{Glycerol}}^{\text{PEO4}}. \quad (2.5)$$

For both fluids we prepared samples with total mass of 120 g. The masses of each solution component, for both samples, are shown in Table 2.1:

Table 2.1: Component masses for each test fluid

Sample	Total mass [g]	PEO [g]	Water [g]	Glycerol [g]	SA [g]
PEO4	120.000	0.240	59.877	59.877	0.006
PEO8	120.000	0.600	119.394		0.006

The fabrication process was initially similar for both fluids, adding the respective masses of PEO and SA to water, and mixing gently in a magnetic stirrer for six days. At this stage, for PEO4, we added the glycerol and gently stirred again for another six days, being, at this stage, visibly homogeneous; PEO8 was kept mixing in the meanwhile. The whole process was conducted at room temperature ($\sim 23^\circ\text{C}$). Additionally, both fluids' densities were measured using a pycnometer of 9.929 ml. Two measurements were done for each fluid, resulting in the mean densities, and respective standard deviations, given in Table 2.2.

Table 2.2: Mean density of each test fluid

Sample	PEO4	PEO8
Measured masses [g]	11.1564	9.9108
	11.1812	9.9084
Calculated densities [kg/m^3]	1124	998.2
	1126	997.9
Mean density [kg/m^3]	1125 ± 1.249	998.1 ± 0.1209

2.1.1 Steady shear viscosity

The steady state viscosity curves were obtained by means of a stress-controlled rotational rheometer, Anton Paar Physica MCR 301 equipped with a parallel-plate geometry with 50 mm of diameter. The temperature was kept constant at 20°C and the parallel plate gap was set at $100\ \mu\text{m}$ in order to obtain reliable results at high shear rates (Ewoldt et al., 2015). Furthermore, all tests were performed using a solvent trap in order to minimise evaporation. These specifications were kept for all rotational rheometer measurements.

In order to guaranty that the viscosity measurements were taken at steady state, preliminary tests were done at constant shear to evaluate the minimum time period that each fluid takes to achieve steady viscosity values. This time-dependence is due to the

non-instantaneous alignment of polymer molecules and to the non-infinite acceleration of the rheometer. It is known that, as the shear-rate increases, it takes less time for the fluid to achieve steady viscosity, as the polymer chains orient themselves faster under faster shear. Nevertheless the time-dependent viscosity values were measured, for both fluids, at two different shear rates. The lower shear rate was 0.1 Pa·s, as it was expected to be within the first viscosity plateau (from previously reported results (Varchanis et al., 2020)). The largest rate was chosen as 100 Pa·s only as a representative value in order to address the time-dependence reduction with shear rate. For both fluids, the procedure was the same: 500 points were taken in 50 s (0.1 s intervals between measurements). The time range shown here, however, was reduced to 20 s from the original 50 for clarity, proving to be more than enough to attain stability. The results are shown in Figure 2.1.

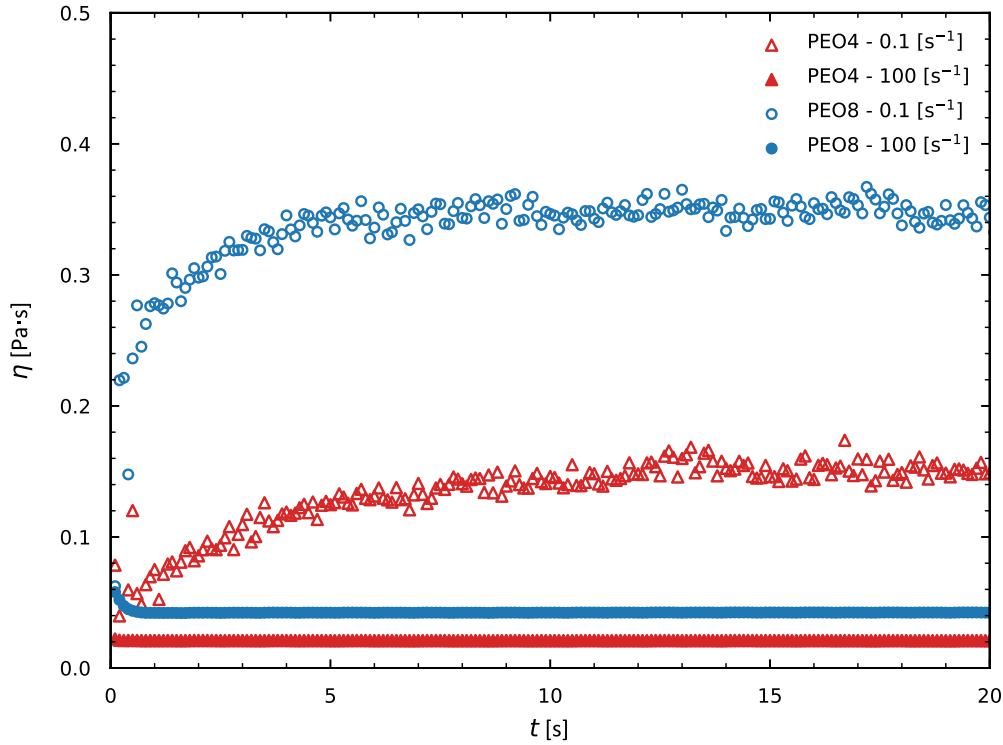


Figure 2.1: Time-dependent viscosity measurements, for PEO4 and PEO8, at constant shear rates: 0.1 and 100 s⁻¹.

From Figure 2.1, we can conclude that, indeed, for higher shear rates both fluids attain stability faster. Also, we can observe that at the lesser shear rate, PEO4 takes more time to attain stability than PEO8. We chose 5 s as the recovery time, even though it could be argued that, at this time, PEO4 isn't quite stable yet. But since we know that the resting time does decrease with shear rate, and that 0.1 Pa·s will be our minimum shear rate, it was thought as acceptable.

Once the required time to achieve the steady state of the viscosity is set, the viscosity curves were obtained varying the shear rate from 0.1 Pa·s to 10⁵ Pa·s. Each test was composed by 55 points and five tests were done for each fluid in order to ensure reproducibility. The results of the five datasets for each fluid are shown in Figure 2.2.

From the raw data collected from all tests, we then computed the mean viscosity curves and the error associated to each mean viscosity value within a 95% confidence

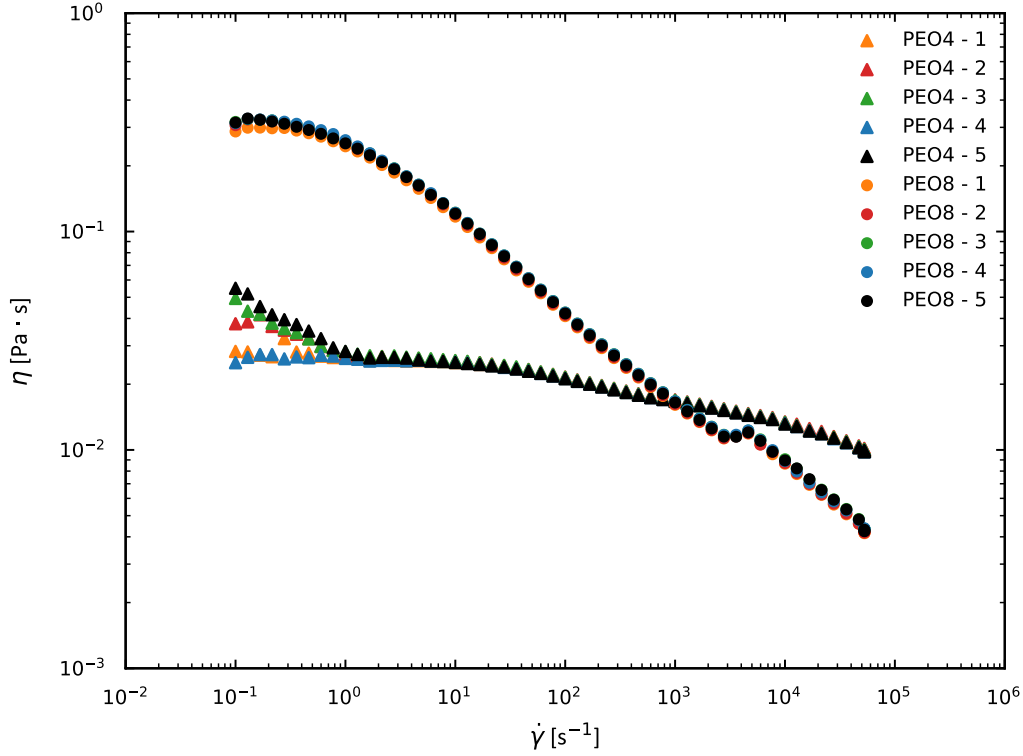


Figure 2.2: Gross data of all obtained viscosity curves for both fluids.

interval. To aid in visualisation, the 95% confidence intervals, which were calculated using Excel's Student's *t* function, were plotted in the form of error bands. These results can be seen in Figure 2.3.

Starting with PEO8, the results for all five series (see Figure 2.2) are very similar, having almost no discrepancies throughout the entire shear rate range. Accordingly, the error was never higher than 5% of the mean value (Figure 2.3). The fluid presents a clear shear-thinning behaviour, exactly as expected. However, at high shear rates ($\dot{\gamma} \sim 3000 \text{ s}^{-1}$) there is a slight increase in apparent viscosity followed by a reduction, similar to the initial shear-thinning behaviour.

It is known that shear-thinning fluids' viscosity tends to the solvent viscosity at increasing shear rates, due to the complete disentanglement and alignment of the polymer molecules. In this case $\eta_{\text{Solvent}} = \eta_{\text{Water}} \sim 10^{-3} \text{ Pa}\cdot\text{s}$ at 20°C , but it was thought that the presence of the polymer itself was raising the viscosity plateau to higher viscosity values. From this school of thought, there were possibilities for the reduction of viscosity after achieving the viscosity plateau, such as sample evaporation or sample loss in the geometry due to the high rotational speeds. But both these were disregarded. The former because water (the used solvent) evaporation at 20°C is negligible, and the latter because there was no presence of sample residues in both the solvent trap and the area between the geometry and the trap. Furthermore, none of these hypothesis explained the increment in viscosity immediately before. Raises in apparent viscosity at high shear rates can either be due to secondary flows, that were disregarded as we will discuss later on, or elastic instabilities (Ewoldt et al., 2015). But these theories also did not explain the second apparent shear-thinning.

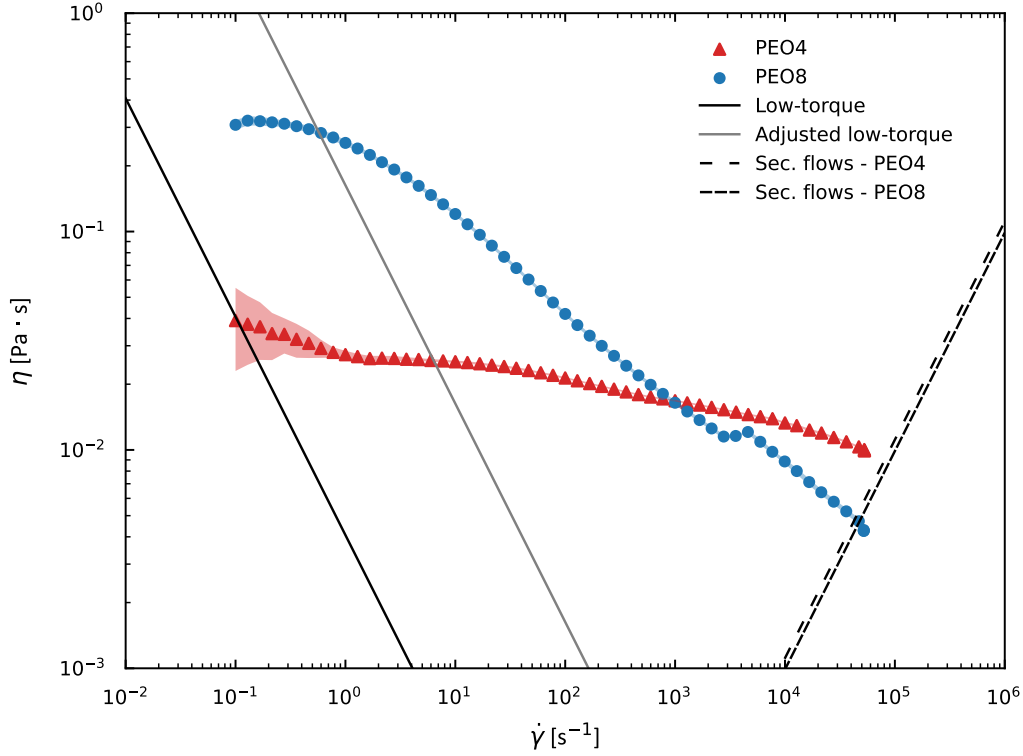


Figure 2.3: Mean-viscosity curves, associated 95% confidence interval bands and experimental limits.

We speculate now, as the most likely explanation, that the second plateau wasn't yet reached at $\eta \sim 1.15 \times 10^{-2}$, and, moreover, wasn't reached at all in our measurements, as the viscosity keeps decreasing, without stabilising, even at our largest achieved shear rates¹. The reason for the slight increase in viscosity is thought to be, indeed, due to an elastic instability and the reason that it doesn't keep increasing, in an apparent shear-thickening manner, would be due to the re-stabilisation of the flow. Elastic instabilities in rheometry are well documented in the literature (Öztekin & Brown, 1993; McKinley et al., 1995; Pakdel & McKinley, 1996), however, being out-of-scope for this work, we shall limit ourselves to quantifying its onset. Oliveira et al. (2006) reported a similar apparent viscosity increase when obtaining the viscosity curve of a solution of 2000 ppm of approximately 3.7 MDa PEO dissolved in a mixture of ethylene glycol and water. The authors also quantify their instability's onset through a Reynolds ($Re = 8.3$) and a modified Weissenberg numbers ($Wi_{Mod.} \sim 11$) for their utilised cone-plate geometry, defined as in the following expressions, respectively:

$$Re = \frac{\rho(\Omega R)(R\theta_0)}{\eta(\dot{\gamma})}, \quad (2.6)$$

¹The maximum shear rate input into the rheometer was $\dot{\gamma} = 10^5 \text{ s}^{-1}$, but our maximum measured value was of $\dot{\gamma} = 5.21 \times 10^4 \text{ s}^{-1}$. This was, most likely, due to the rheometer's mechanical limitations.

$$Wi_{\text{Mod.}} = Wi\sqrt{\theta_0} = \lambda\dot{\gamma}\sqrt{\theta_0}, \quad (2.7)$$

where ρ is the fluid density, Ω is the rotational speed, R is the geometry radius, θ_0 is the cone angle, $\eta(\dot{\gamma})$ is the shear viscosity, λ is the fluid's relaxation time (the relaxation times of both fluids were obtained experimentally and will be considered further in Section 2.1.2) and $\dot{\gamma}$ is the shear rate.

In order to have comparable results, we sought to compute both these dimensionless numbers for the onset of our instability ($\dot{\gamma} \sim 2770 \text{ s}^{-1}$ and $\eta \sim 0.01151 \text{ Pa}\cdot\text{s}$). Of course our parallel plate geometry has no cone angle, θ_0 , but for small values, it can be adjusted to our geometry as (Pakdel & McKinley, 1996): $\theta_0 \sim h/R$. Where h is our geometry gap (0.1 mm) and R is our geometry radius (25 mm). Additionally knowing that $\dot{\gamma} = R\Omega/h$ (Pakdel & McKinley, 1996), Equations 2.6 and 2.7 become, respectively:

$$Re = \frac{\rho h^2 \dot{\gamma}}{\eta(\dot{\gamma})}, \quad (2.8)$$

$$Wi_{\text{Mod.}} = Wi\sqrt{\theta_0} = \lambda\dot{\gamma}\sqrt{\frac{h}{R}}, \quad (2.9)$$

The Reynolds and modified Weissenberg numbers computed for the onset of our instability are shown in Table 2.3.

Table 2.3: Characterisation of the onset of the encountered elastic instability through the Reynolds and modified Weissenberg numbers.

$\dot{\gamma} [\text{s}^{-1}]$	$\eta [\text{Pa}\cdot\text{s}]$	$Re [-]$	$Wi_{\text{Mod.}} [-]$
2770	0.01151	2.40	23.30

As we can gather from Table 2.3, we have large a Weissenberg number for a reduced Reynolds number; however, we are nowhere near creeping flow as $Re > 1$, thus, both elastic and inertial effects seem to be important, just as Oliveira et al. concluded as their results are relatively similar to ours, only slightly larger Reynolds and lower Weissenberg numbers. In Figure 2.3 the onset of the instability is noted but there is no reason to disregard the viscosity data after it's dissipation, only additional considerations will be taken when adjusting the viscosity model, but that will be discussed further on.

Moving on to PEO4, it presents a slight shear-thinning behaviour, but it can be treated as a practically non-shear-thinning fluid in the range of this study (the shear rate ranges, to be discussed later in Section 3.2, are indicated in Figure 2.4 through vertical bands). The results for all five series were as coincident as for PEO8, showing good reproducibility, except for an evident discrepancy which can be observed in Figure 2.2 for lower shear rates, having up to a maximum error of 40% for the minimum shear rate (see Figure 2.3). These fluctuating values are well associated to the rheometer's minimum torque, below which the measured values are prone to large errors (Ewoldt et

al., 2015). In fact, we could also re-mention that PEO4 was not quite stable within 5 s of the rheometer's velocity shifts, as was discussed before, however, these fluctuations are more likely to be associated with the mechanical limitations of the rheometer than that issue. Thus, we shall discuss now the well known low-torque limit. For a parallel plate geometry, this limit is given by (Ewoldt et al., 2015)

$$\eta_{\min}^{\text{torque}} = \frac{F_{\tau} T_{\min}}{\dot{\gamma}} = \frac{2T_{\min}}{\pi R^3 \dot{\gamma}}, \quad (2.10)$$

where $T_{\min}$ is the minimum torque applied by the rheometer, which is, in this case, 0.1 $\mu\text{N}\cdot\text{m}$, from the machine's specifications; and R is the geometry radius (25 mm). Plotting this limit, as can be seen in Figure 2.3, it seems out-of-reach of the high-error values. Well, in fact this is a theoretical limit, and it is subjected to the rheometer's physical state. It was assumed that the non-ideal condition of the machine, as all real machines are, implies a shift of this limit to higher shear rates. Moreover, other effects, such as surface tension, may also shift the low-torque limit (Ewoldt et al., 2015). The shift can simply be done by multiplying the equation by an adjustment coefficient larger than one. From former work done with the rheometer in question, we found that a usual, conservative, value for this coefficient would be of around 40, thus, this was the value utilised in our analysis, even if seemingly over-conservative. The new, adjusted low-torque limit, given by Equation 2.11, can also be seen in Figure 2.3.

$$\eta_{\min}^{\text{adj.torque}} = 40 \times \eta_{\min}^{\text{torque}} \quad (2.11)$$

The final note for our data analysis is the already mentioned secondary flows. This phenomenon occurs at high shear rates due to fluid inertia and generates an apparent shear-thickening. Secondary flows for cone-plate and parallel plate geometries consist in the formation of an outward radial velocity component at the upper plate due to centrifugal effects and an inward velocity at the lower plate, from mass conservation (Ewoldt et al., 2015). The limit at which we can expect secondary flows, for a plate-plate geometry is given by (Ewoldt et al., 2015)

$$\eta_{\max}^{\text{sec}} = \frac{\rho h^3 \dot{\gamma}}{Re_{\text{crit}} R}, \quad (2.12)$$

where, ρ is the fluid density, R remains the geometry radius, given before, h is the gap dimension, already mentioned as 100 μm , and Re_{crit} is the critical Reynolds number above which we can expect secondary flows. Secondary flows apply to both PEO4 and PEO8, thus, limits for both fluids were computed and plotted, in spite of being quite similar as the densities are also very alike. The critical Reynolds number is obtained considering the ratio between the measured torque, aggravated by secondary flows, T , and the ideal torque generated only by free shear flow, T_0 (Ewoldt et al., 2015):

$$\frac{T}{T_0} = 1 + \frac{3}{4900} Re_{\text{crit}}^2. \quad (2.13)$$

For a torque error of 1% ($T/T_0 = 1.01$, similarly to the considered by Ewoldt et al. (2015)), the critical Reynolds number is of approximately 4, and the limits for both PEO4 and PEO8 are plotted in Figure 2.3. As we can observe, both limits are very close together, showing that fluid density variations, in the order of magnitude of our fluids, are not critical. Moreover, both limits are quite out-of-range of our apparent viscosity increase for PEO8 and, therefore, we disregarded the influence of secondary flows in our sample, as was mentioned before. From Figure 2.3, we can observe that some data points for PEO8, at the highest shear rates, are actually in range of secondary flows; however, at that range, the data's quality is no longer assured due to the rheometer's mechanical limitations.

Having all these boundaries set, we compare our mean viscosity curves to the literature, trimming only the viscosity values measured at the maximum measured shear rate, as they may be affected by systematic errors associated to the rheometer's limitations, and the data points beyond the low-torque limit. In Figure 2.4 our useful data is then shown and compared to the viscosity curves presented by Varchanis et al. (2020) for both fluids, and another PEO8 viscosity curve measured by Rodrigues et al. (2022). Fitting curves, corresponding to viscosity models, are also shown in Figure 2.4 and will be discussed later in this Section; additionally, shadowed areas, representing each fluid's experimental range, can also be seen, these will be discussed in Section 3.2 but were shown here to avoid repetition later on.

From Figure 2.4 we can conclude that our PEO4 sample agrees relatively well with the viscosity curve given by Varchanis et al. (2020). Our fluid presents slightly higher viscosity values and a moderately steeper shear-thinning behaviour, nevertheless, this shear-thinning can be considered irrelevant when comparing to PEO8, and we can conclude that our sample is fit to be studied. Our PEO8 sample also agrees well with other measurements, it's shear-thinning behaviour is indeed very similar to the ones reported by Varchanis et al. (2020) and Rodrigues et al. (2022), diverging only by presenting slightly lesser overall viscosities, and, therefore, this sample is also ready to move forward.

The next step is to fit viscosity models to our fluids; this will aid us later on, when developing viscosity charts from our μPIV results. For both fluids were employed Carreau-Yasuda (C-Y) models, very commonly utilised to describe shear-thinning fluids. Initially, for PEO4, a power-law model was considered, as the sample should represent a constant-viscosity fluid. Despite, indeed, acting as an almost non-shear-thinning fluid, especially when comparing to PEO8, a better correlation was obtained with a C-Y model, and noting that, in our case, a well-fitting model is quite relevant for μPIV data analysis, we kept the latter, which is given, generally, by Equation 2.14.

$$\eta_{\text{C-Y}} = \eta_{\infty} + (\eta_0 - \eta_{\infty}) \left[1 + \left(\frac{\dot{\gamma}}{\dot{\gamma}^*} \right)^a \right]^{\frac{n-1}{a}} \quad (2.14)$$

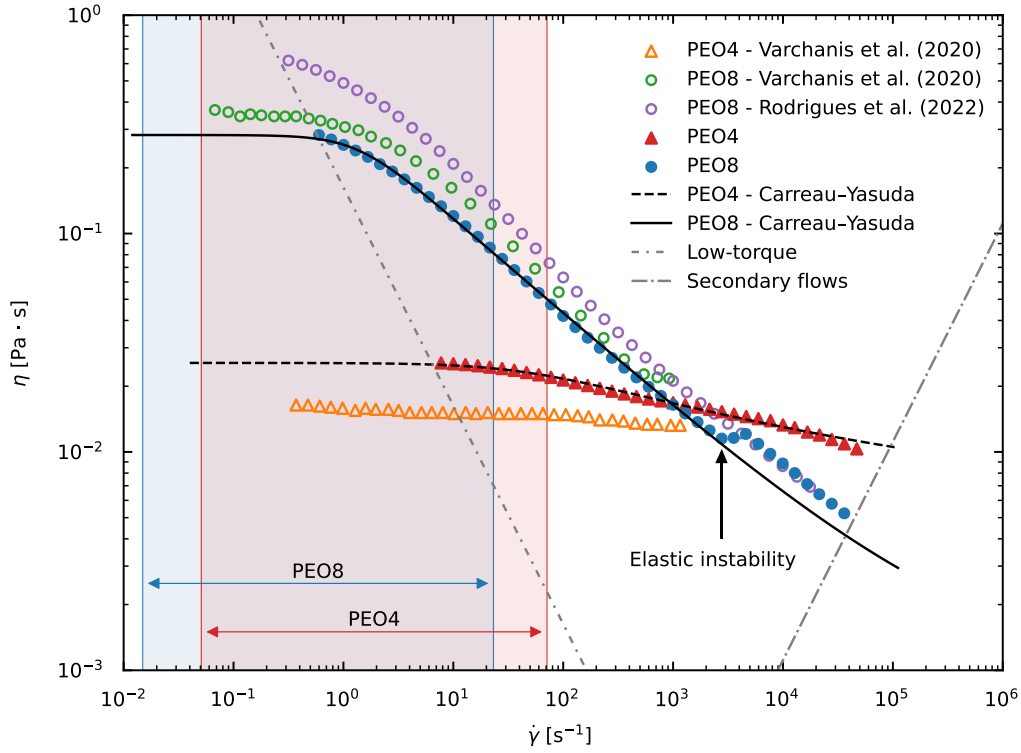


Figure 2.4: Trimmed mean-viscosity curves, comparative data from the literature, relevant experimental limits and experimental range of shear rates achieved in our work for each fluid.

Where η_∞ is the infinite-shear viscosity, usually similar to the fluid's solvent viscosity, η_0 is the zero-shear viscosity, $\dot{\gamma}^*$ is the characteristic shear rate for the onset of shear-thinning, $\dot{\gamma}$ is the shear rate, n is the dimensionless power-law index and a is a dimensionless exponent that describes the transition between the zero-shear plateau to the power-law region (Haward et al., 2020).

The fitting processes were carried out by iteratively changing some of the model's material constants in order to minimise the following error function (Escudier et al., 2001):

$$\sum_N \left(1 - \frac{\eta_M}{\eta_{\text{model}}} \right)^2, \quad (2.15)$$

where η_M and η_{model} are the measured and the modelled viscosities, respectively. And the quality of the fits were analysed through the Pearson correlation coefficient given by Excel as:

$$r = \frac{\sum (\eta_M - \bar{\eta}_M)(\eta_{\text{model}} - \bar{\eta}_{\text{model}})}{\sqrt{\sum (\eta_M - \bar{\eta}_M)^2 - \sum (\eta_{\text{model}} - \bar{\eta}_{\text{model}})^2}}. \quad (2.16)$$

As mentioned before, not all of the model's material constants were altered. For both fluids, η_0 and η_∞ were fixed in order to preserve rheological significance in the model's

mathematical formula. The values for η_0 were kept as the approximate values found experimentally, i.e., the first values of both trimmed viscosity curves, as the low-torque limit applied seems to cross our data grossly at the end of both low-shear viscosity plateaus (see Figure 2.3). The values for η_∞ were considered as the solvents' viscosities. For PEO8 this meant water viscosity, which, at 20°C, is approximately 10^{-3} Pa·s. For PEO4, the solvent is a 50%wt aqueous solution of glycerol, for which Shankar & Kumar (1994) provide kinematic viscosity data obtained experimentally (at 20°C: $\nu = 5.26 \times 10^{-6}$ m²/s). The dynamic viscosity is then given as: $\eta = \nu\rho \sim 5.918 \times 10^{-3}$ Pa·s. Going back to PEO8's elastic instability, it was thought pertinent to adjust the model only to the viscosity data before the instability's onset, as the model would adjust more tightly to the low-shear data and the Pearson coefficient, applied to the entire range, was acceptable. Both fitted Carreau–Yasuda models can be seen in Figure 2.4 and their respective material constants and Pearson correlation coefficients, applied to the whole range, can be seen in Table 2.4.

Table 2.4: PEO4 and PEO8's fitted Carreau–Yasuda viscosity models' material constants and respective Pearson correlation coefficient, r

Sample		PEO4	PEO8
Material constants	η_∞ [Pa·s]	0.0059	0.0010
	η_0 [Pa·s]	0.0256	0.2826
	$\dot{\gamma}^*$ [s ⁻¹]	40.7564	1.3363
	n [-]	0.8147	0.5608
	a [-]	1.0663	1.9402
r		0.9969	0.9995

Analysing the Pearson coefficients we can gather that both fluids are reasonably well fitted by the respective C–Y models as, according to Mustapha et al. (1999), a reasonable adjustment coefficient would be of 0.9975. The models' material constants are also reasonable. $\dot{\gamma}^*$ that, as mentioned before, corresponds to the characteristic shear rate at which we can expect a transition from the low-shear plateau to the power-law region, agrees with the experimental results. The power-law indexes, n , being lower than unity, correspond to shear-thinning fluids, being PEO8's lower value representative of its more accentuated viscosity decrease, also compatible with our data.

Now, that our fluids' viscous behaviours are modelled, it was found pertinent to obtain their relaxation times in order to calculate viscoelastic dimensionless numbers, namely the Weissenberg numbers, facilitating the comparison of our results with the literature. Moreover, we can then further compare our samples to the fluids prepared by Varchanis et al. (2020).

2.1.2 Capillary breakup extensional rheometry

The relaxation time of a fluid can be very simply attained using a Capillary Breakup Extensional Rheometer (CaBER). The working principle consists in placing a small fluid sample between two circular plates distanced by a small height. The upper plate is then rapidly separated from lower one until a final user-set height is achieved. The fluid

sample takes initially an hourglass shape, which progressively thins with time onto a filament with cylindrical shape, in the case of viscoelastic fluids—elasto-capillary regime with no inertial interference. The filament undergoes a thinning process, balancing the surface tension with the fluid's elastic properties (Mckinley, 2005b; Galindo-Rosales et al., 2015). In this elasto-capillary regime, the variation of the filament's diameter with time can be associated to the fluid's relaxation time through the following equation (Mckinley, 2005b):

$$\frac{D(t)}{D_0} \sim \left(\frac{D_0 G}{4\sigma} \right)^{1/3} \exp \left(-\frac{t}{3\lambda} \right), \quad (2.17)$$

where D_0 is the initial sample diameter, usually the plate's diameter; G , σ and λ are the fluid's elastic modulus, surface tension and relaxation time, respectively.

All tests were carried out on an HAAKE™ CaBER™ 1, Thermo Scientific, shown in Figure 2.5, at room temperature (approximately 24°C), and all stretch profiles were linear with a "strike time" of 20 ms.

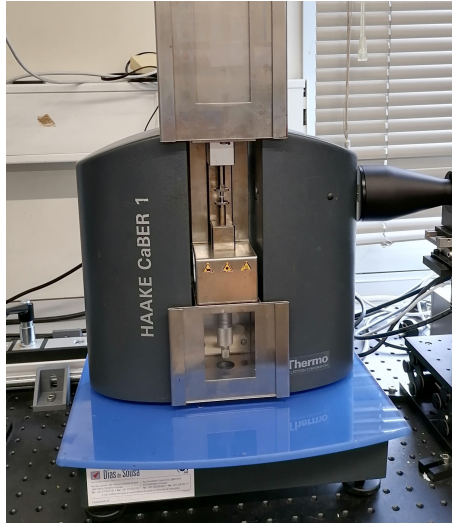


Figure 2.5: Capillary Break Up Extensional Rheometer (CaBER™ 1, HAAKE™ , Thermo Scientific).

In order to guaranty the reliability of our experimental results, two conditions were taken into account:

- Firstly, that the surface tension is capable of supporting the fluid filament against gravitational collapse. This is ensured by having a sufficiently small initial height so that it supports a static liquid bridge. The condition is set by the following expression (Galindo-Rosales et al., 2015; Rodd et al., 2005):

$$\frac{h_0}{l_{\text{cap}}} < 1, \quad (2.18)$$

where l_{cap} is the capillary length and is given by:

$$l_{\text{cap}} = \sqrt{\frac{\sigma}{\rho g}}. \quad (2.19)$$

- Secondly, there is an optimal range, found in numerical studies, for the initial aspect ratio, as described in the following expression (Galindo-Rosales et al., 2015):

$$0.5 < \left[\Lambda_0 = \frac{h_0}{D_0} \right] < 1. \quad (2.20)$$

Considering our first condition, the capillary lengths of both samples had to be calculated. The surface tensions of PEO solutions can be found in the literature. Arnolds et al. (2010) provide surface tension values attained experimentally for aqueous solutions of 1 and 2 MDa PEO with concentrations varying between 0.5 and 5 wt%. The surface tension values varied between 53.7 and 65.7 mN/m. Rodd et al. (2005) provide surface tension values for two solutions of 0.1 wt% of 2 MDa PEO; one aqueous solution and, the other, a mixture of 55% of glycerol in water, yielding surface tension values of 61 and 58 mN/m, respectively. We gather from these results that surface tension values for our PEO solutions can be considered, for a rough estimate of the initial height, of approximately 60 mN/m. Considering their respective densities, the maximum initial heights for each fluid were calculated from Equation 2.18, giving, respectively for PEO4 and PEO8, heights of 2.33 and 2.48 mm. Thus, an initial height was chosen for both fluids as $h_0 = 2$ mm. Reconsidering the approximation, surface tensions of 44 and 39 mN/m would be the lowest values for PEO4 and PEO8, respectively, for the condition set by Equation 2.18 to be broken when fixing h_0 to 2 mm, which fall below the usual range of surface tensions for PEO solutions.

For the second condition, having now $h_0 = 2$ mm, the initial aspect ratio inequalities of Equation 2.20 yield boundaries for the initial diameter as:

$$2 < D_0[\text{mm}] < 4. \quad (2.21)$$

Our available geometries had diameters of 4, 6 and 8 mm. Thus, the 4 mm geometry was selected.

The final aspect ratio, and therefore the final height, ought to have no effect on the measurement of the relaxation time (Galindo-Rosales et al., 2015); however, if the stretch is too high, amplification of inertial effects is expected, resulting in fluctuations of the filament's diameter that may compromise its correct measurement (Rodd et al., 2005). Initially a final height of approximately 6 mm was selected for both fluids; however, and also to verify that the variation of the parameter should not significantly affect the measurements, for PEO8 additional tests were conducted with a final height of around 9 mm.

In Figure 2.6 are shown the sample placement in the measurement geometry and the elasto-capillary regime (both for a PEO8 measurement).

Several tests were done for each fluid to guaranty a good statistical analysis and

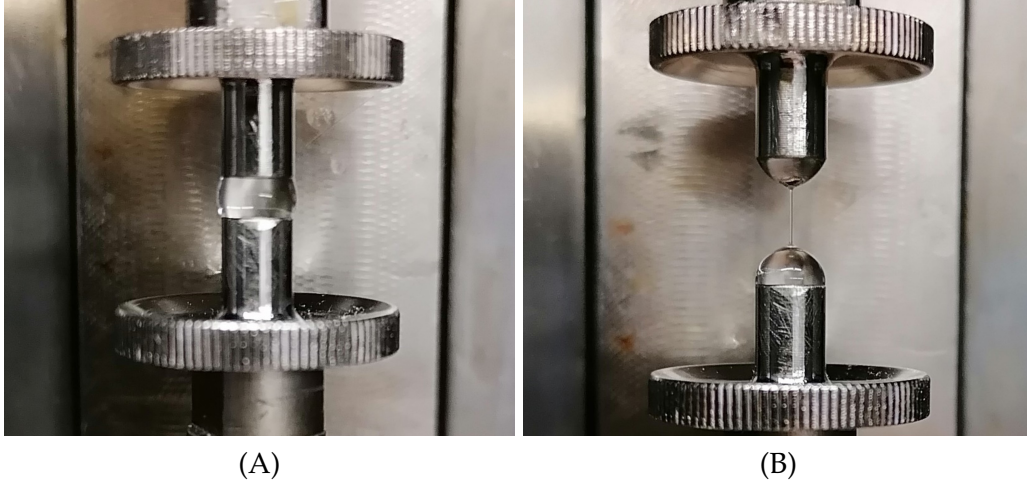


Figure 2.6: CaBER measurement: fluid sample placement within the employed measurement geometry (A) and fluid filament in the elasto-capillary regime (B).

measurement reproducibility. All tests were plotted individually showing the evolution of $D(t)/D_0$ with time until filament breakup. The relaxation times were attained by fitting exponential curves to a portion of the data corresponding to the elasto-capillary regime of each test and analysing the exponent of the fit, according to Equation 2.17. It was not found useful, nor pertinent to have all data sets and fitting curves shown here; however, to exemplify the measured data and the fitting process, in Figure 2.7 are shown two data sets (the first test on PEO4 and the first 9 mm final height PEO8 measurement) given by CaBER and their respective exponential fits.

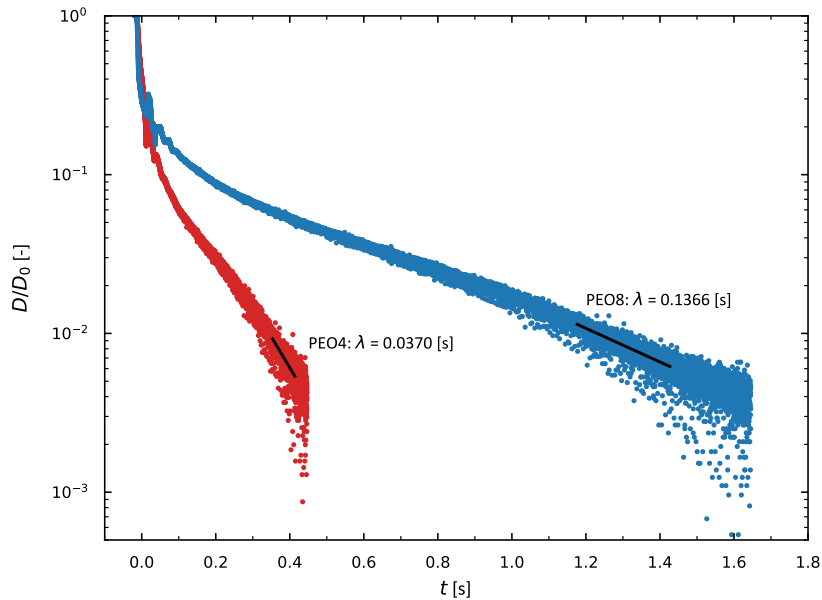


Figure 2.7: Sample dimensionless diameter variation with time, fitting exponential curve and respective relaxation time for each fluid.

All respective fluid tests were very similar, showing good reproducibility. Inertial-

driven oscillations can be seen at the initial stages of the tests, however, as they dampen before the analysed range, they do not influence the quality of our results. In fact, our fit data range was chosen specifically not to be affected by inertial effects. It is worth to note that our tests begin at $t = -20$ ms, this corresponds to our "strike time", such that our $t = 0$ s corresponds to the instant at which the final height is achieved. We consider in all tests the final height, h_f (which changed slightly from test to test), the relaxation time and the filament breakup time as relevant results. These were compiled, and treated statistically, calculating the mean values and standard deviations, for PEO4 and PEO8 (and also for different chosen final heights for PEO8), respectively shown in Tables 2.5 and 2.6.

From the collected data we can then gather the relaxation times: $\lambda_{\text{PEO4}} = 0.0383$ s and $\lambda_{\text{PEO8}} = 0.1330$ s. These results agree relatively well with the relaxation times obtained by Varchanis et al. (2020). Also, the filament breakup times are: $t_{\text{break}}^{\text{PEO4}} = 0.450$ s and $t_{\text{break}}^{\text{PEO8}} = 1.624$ s, which goes accordingly to PEO8's largest elasticity. Moreover, now considering the effect of the final height, it is relevant to note that no aggravations of inertial effects were noted for the increase in final height, from 6.62 to 9.06 mm, for PEO8; nor, from analysing Table 2.6, are the relaxation and breakup times affected by it. No data is shown in Figure 2.7 for PEO8's final height of 6.62 mm due to the scatter similarity to the largest height plots, making it irrelevant to try to visually discern between the two.

Table 2.5: Final heights, relaxation times, breakup times and respective mean values and standard deviations for all tests done in CaBER with PEO4

Sample	h_f [mm]	λ [s]	t_{break} [s]
PEO4	6.17	0.0370	0.446
	6.29	0.0441	0.463
	6.29	0.0431	0.463
	6.25	0.0399	0.462
	6.21	0.0328	0.441
	6.25	0.0363	0.460
	6.21	0.0386	0.431
	6.25	0.0362	0.434
	6.21	0.0400	0.449
	6.25	0.0348	0.453
PEO4: Mean values	6.24 ± 0.04	0.0383 ± 0.0034	0.450 ± 0.011

Table 2.6: Final heights, relaxation times, breakup times and respective mean values and standard deviations for all tests done in CaBER with PEO8

Sample	h_f [mm]	λ [s]	t_{break} [s]
PEO8	6.56	0.1304	1.606
	6.64	0.1349	1.599
	6.48	0.1420	1.685
	6.52	0.1318	1.592
	6.76	0.1230	1.579
	6.76	0.1257	1.614
	6.60	0.1241	1.491
	6.62 ± 0.10	0.1303 ± 0.0062	1.595 ± 0.053
	8.87	0.1366	1.645
	9.02	0.1419	1.643
	8.98	0.1500	1.683
	9.02	0.1396	1.750
	9.10	0.1350	1.685
	9.14	0.1256	1.634
	9.26	0.1219	1.531
	9.06 ± 0.12	0.1358 ± 0.0089	1.653 ± 0.062
	PEO8: Mean values		0.1330 ± 0.0082
			1.624 ± 0.064

2.2 Microfluidic devices

2.2.1 Design

Our microfluidic devices' geometry is a 10 mm long channel of rectangular cross-section, with half-width: $L_c = 135 \mu\text{m}$, and depth: $H = 100 \mu\text{m}$. A circular cylinder of $75 \mu\text{m}$ of diameter is placed at the half-width of the channel, distanced 7 mm from the channel's entrance, in order to avoid entrance effects, assuring fully developed flow. This geometry was designed by Rodrigues et al. (2019) in order to approximate a small blood vessel with a microbot within. The hydraulic diameter of the channel is given by Equation 2.22, whose value ($\sim 146 \mu\text{m}$) falls into the small blood vessel range (less than 1 mm of diameter), at which viscoelastic effects are enhanced, as was mentioned in Section 1.1.

$$D_h = \frac{4L_c H}{2L_c + H} \quad (2.22)$$

The diameter of the cylinder was chosen as 10 times the average size of a RBC, in order for real whole blood, if employed in the channel, to be considered a continuous

medium and not a suspension (Rodrigues et al., 2020). A schematic of the channel is shown in Figure 2.8.

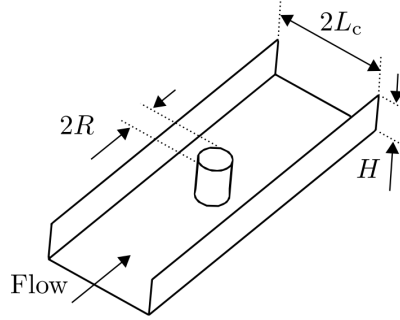


Figure 2.8: Schematic representation of the channel's geometry. Adapted from Rodrigues et al. (2020), with permission from Elsevier.

The microchannels also had pressure taps, designed by Rodrigues et al. (2019), up and downstream of the cylinder, both distancing $500\text{ }\mu\text{m}$ from the respective cylinder face. The design of the pressure taps is out-of-scope for this work as we will not be executing any pressure measurements. Our microfluidic chips contained three microchannels, all differing only on the pressure taps' design. Two of these had the same tap intake width (wide pressure taps, or WPT) and the other had a narrower width (narrow pressure tap, or NPT). The different widths of the pressure taps can be seen in Figure 2.9, where streamlines of obtained velocity fields were inserted only for reference of scale.

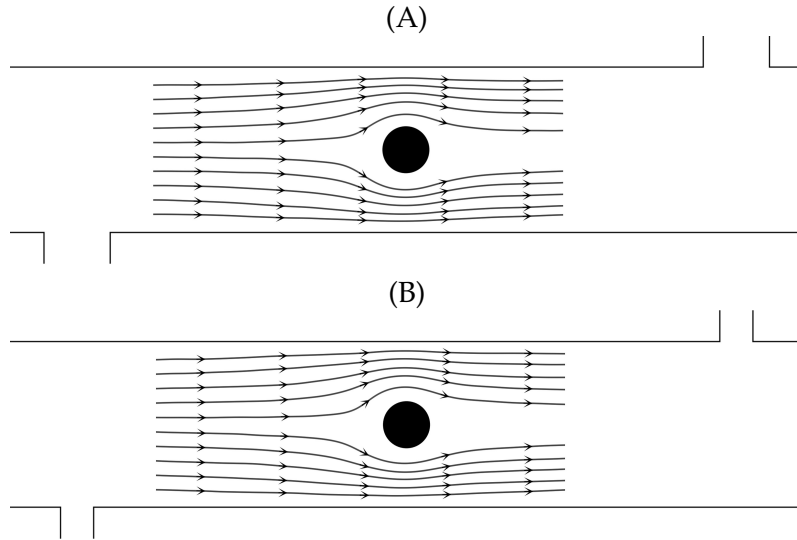


Figure 2.9: Schematics of the microchannels: (A) wide pressure tap intake (WPT), and (B) narrow pressure tap intake (NPT), with their respective PEO8, $Wi = 100$ streamlines. The streamlines' width and the arrow dimensions were enlarged, relative to the plotted results of Section 3.1 to aid in their visualisation.

The relevant geometric parameters for flow around a cylinder are the channel's and the cylinder's aspect ratios (α and Λ , respectively) and the blockage ratio (β), which have been mentioned before in Section 1.3. For our geometry they are given by the following expressions:

$$\alpha = \frac{H}{2L_c}, \quad (2.23)$$

$$\beta = \frac{2R}{2L_c}, \quad (2.24)$$

$$\Lambda = \frac{\alpha}{\beta} = \frac{H}{2R}. \quad (2.25)$$

In Table 2.7, the relevant geometric dimensions and parameters are shown.

Table 2.7: Microchannel's nominal geometric dimensions and parameters, from Rodrigues et al. (2020)

$2R$ [μm]	$2L_c$ [μm]	H [μm]	D_h [μm]	α [-]	β [-]	Λ [-]
75	270	100	146	0.37	0.28	1.33

2.2.2 Fabrication

The devices were fabricated with PolyDiMethylSiloxane (PDMS), using soft-lithography techniques from a SU-8 coated silicon mould containing the aforementioned WPT and NPT channels. Other techniques exist for the fabrication of microchannels, for instance, the studies of Haward et al. are conducted with large aspect ratio microchannels fabricated out of rigid, silica glass through selective laser-induced etching (SLE); however, even though it enables for higher flow rates, this process is more time- and cost-demanding than PDMS soft-lithography. Considering the moulds themselves, they were both fabricated on single square pieces of silicon through optical lithography from aluminium coated Corning[®] glass hard-mask, on which the channel designs were originally cut out through direct laser lithography. The microfluidic chips were fabricated using a two-part Sylgard[®] 184 PDMS polymer kit. The polymer kit and the mould are shown in Figure 2.10.

The PDMS was mixed thoroughly with the cross-linker in a 10:1 ratio for 5 minutes and was then inserted into a desiccator for 1 hour, in order to degas under vacuum, removing all air bubbles created in the mixing process. In the meanwhile, the SU-8 mould was subjected to a silanization process with TriMethylChloroSilane (TMCS) for also 1 hour, in order for it to become hydrophobic and facilitate the peeling of the PDMS layer. If this process were to be skipped, the PDMS would stick to the silicon mould, making the removal of the layer very difficult. When both processes were done, the PDMS was poured into the mould gently and was allowed, once again, to degas in the desiccator for another 30 minutes, to avoid any remaining air bubbles that may have formed in the pouring process. The silanization process and the utilised desiccator can be seen in Figure 2.11.

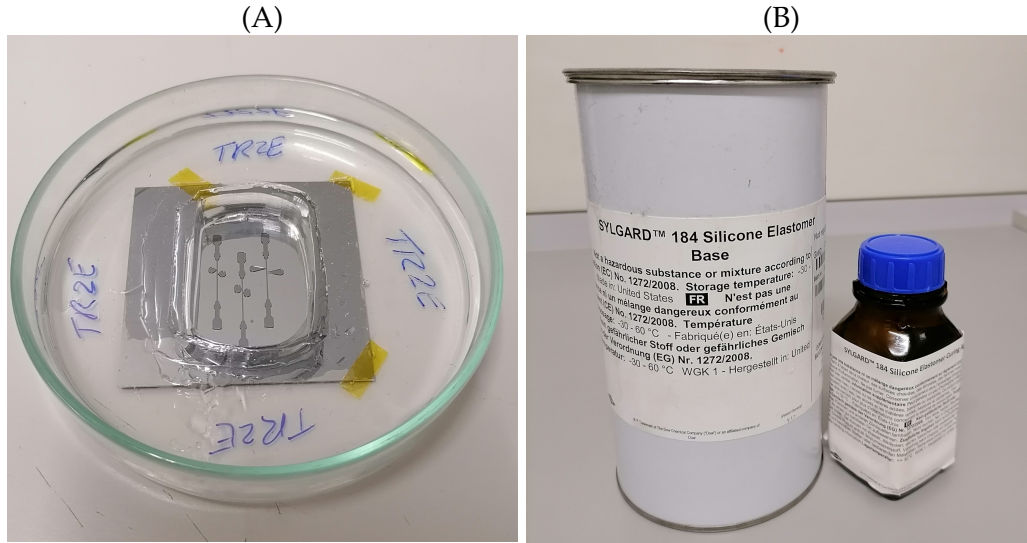


Figure 2.10: (A) SU-8 mould of the microchannels, and (B) the employed two-part Sylgard[®] 184 PDMS polymer kit.

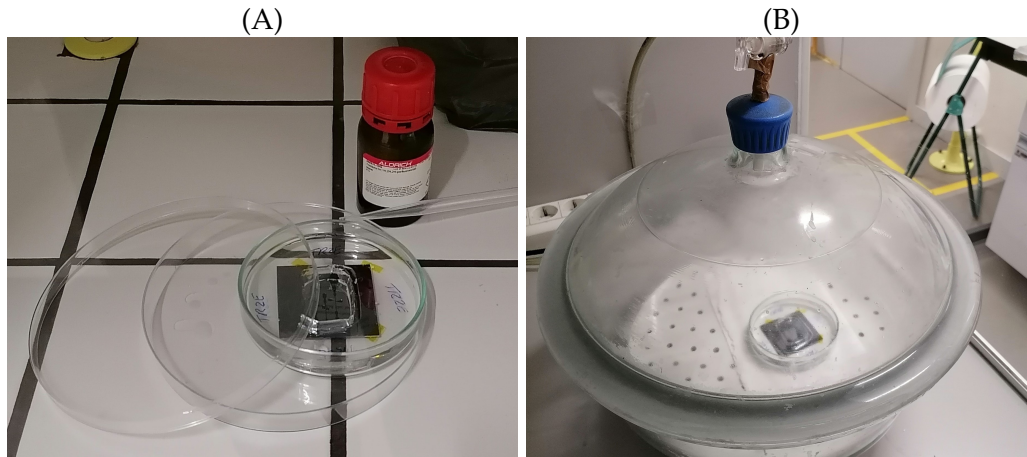


Figure 2.11: (A) silanization process of the SU-8 mould with TMCS and (B) PDMS-filled mould in the desiccator.

Then, the PDMS layer, yet within the mould, was cured in an oven at 80°C for 30 minutes to accelerate the cross-linkage while making the fully cured PDMS chip soft enough to be manipulated in the next steps. Then, the cured PDMS layer was cut with a scalpel, peeled off from the mould and the channels' inlets and outlets were punctured with a punch. It is relevant to notice that the pressure taps were not punctured, as they won't be used in our study. In Figure 2.12, the peeled and punctured layer can be seen.

The channels were closed with a glass microscope slide, which was, before-hand, cleaned in an ethanol bath in an ultrasonic cleaner for 8 minutes, and then cleansed with distilled water and thoroughly dried. The PDMS layer and the glass slide were plasma-bonded for 30 seconds in vacuum (1.4 mbar approximately). Figure 2.13 shows the glass slide in the ultrasonic cleaner and the plasma-bonding of the slide and the PDMS layer.

The final product was then allowed to rest overnight before usage for μ PIV testing. The final microfluidic chip can be seen in Figure 2.14.



Figure 2.12: Cut, peeled and punctured, cured PDMS layer.



Figure 2.13: (A) ultrasonic cleaning of the glass microscope slide and (B) plasma bonding of the slide and the PDMS layer.



Figure 2.14: Fabricated PDMS-glass microfluidic chip.

2.3 Flow control and dimensionless groups

A neMESYS low pressure syringe pump of 14:1 gear ratio (CETONI GmbH) was used to drive the flow into the microchannels. Considering our flow rate range (which will be discussed further on), a 5 ml Hamilton® Gastight syringe was used for all measurements. The syringe and the inlet of the microchannels were connected by a flexible Tygon® tube of 440 μm of inner diameter and a stainless steel precision tip of 8 μm of inner diameter (Nordson Corporation) was used to fix the tube to the PDMS microfluidic chip. The outlet of the channel was left open, but, in order to avoid cleaning it every few seconds, it was treated similarly to the inlet, using now a larger 16 μm tip to connect the channel with the tube, avoiding leaking in the outlet due to spacing between the tip and the PDMS. The tube was then lead into a plastic cup to dispose of the used fluid, which was not re-utilised to guarantee the quality of the results. The whole experimental setup can be seen in Figure 2.15.

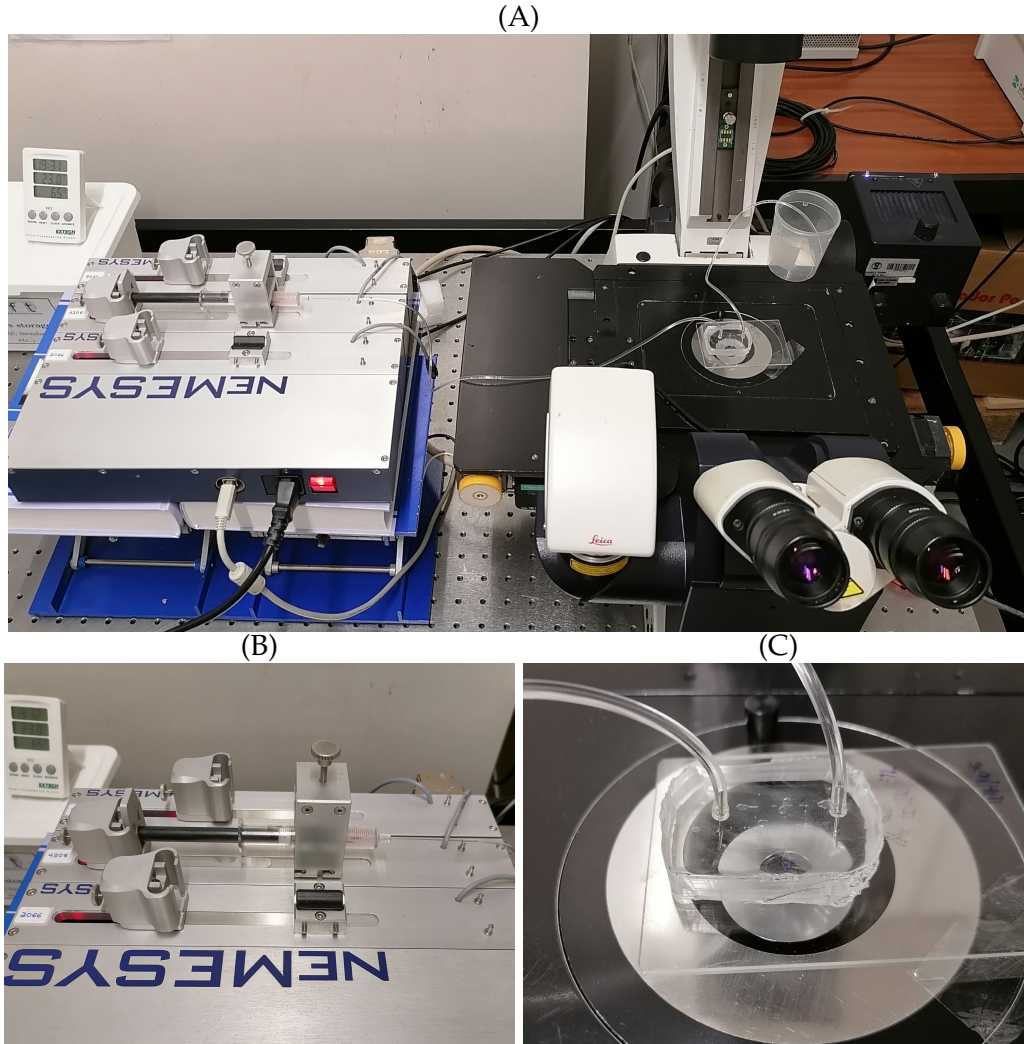


Figure 2.15: Experimental setup for μPIV : (A) general equipment, (B) syringe and syringe pump and (C) in-use microchannel.

As we knew we would be dealing with strongly elastic, low Reynolds flows, our flow conditions were ruled by the desired Weissenberg numbers, which were defined,

similarly to the work of Haward et al., and, from which, the mean velocity in the channel can be calculated as

$$U = \frac{WiR}{\lambda}. \quad (2.26)$$

The flow rate is then calculated by

$$Q = 2L_c H U. \quad (2.27)$$

The Reynolds number around the cylinder can also be obtained from the mean velocity, considering the fluid's characteristics:

$$Re = \frac{\rho U R}{\eta_0}, \quad (2.28)$$

where η_0 is the zero-shear viscosity obtained from the Carreau–Yasuda models. The elasticity number, constant for each fluid, is given by:

$$El = \frac{Wi}{Re} = \frac{\lambda \eta}{\rho R^2}. \quad (2.29)$$

The viscoelastic Mach number:

$$Ma = \sqrt{Wi Re}. \quad (2.30)$$

And finally, the average velocity in the contractions around the cylinder (Rodrigues et al., 2020):

$$U_c = U \frac{L_c}{L_c - R}. \quad (2.31)$$

Before the μ PIV measurements, preliminary visualisations with PEO8 were conducted in order to evaluate at which Weissenberg numbers a flow asymmetry could be observed. It was found, visually, that the asymmetry would onset between Wi of 100 and 200, and that it was kept until the cylinder dislocated from the microscope slide, at Wi of approximately 600. Thus, the studied flows were for Wi of 100, 200, 300, 400 and 500. It is worth noting that for $Wi = 500$, even though the cylinder did not dislocate from the glass slide, a very slight bending in the flow direction could be observed at the centre plane, returning to the original position when lowering the flow rate. This is due to the non-rigid nature of our channels. This elastic bending did not seem to affect the obtained results and, moreover, being a streamwise deformation, and, indeed, very

slight, it should not have any significant influence in the analysis of the axial asymmetry. The relevant flow parameters relative to these Weissenberg numbers, for both fluids, are shown in Table 2.8.

Table 2.8: Relevant flow parameters for μ PIV measurements, for both fluids

Sample	Wi [-]	U [mm/s]	Q [ml/h]	Re 10^{-3} [-]	Ma [-]	U_c [mm/s]	El [-]
PEO4	100	97.911	9.517	161.334	4.017	135.569	620
	200	195.823	19.034	322.667	8.033	271.139	
	300	293.734	28.551	484.001	12.050	406.708	
	400	391.645	38.068	645.334	16.067	542.27	
	500	587.467	57.102	968.002	24.100	813.416	
PEO8	100	28.195	2.741	3.298	0.574	39.040	30323
	200	56.391	5.481	6.596	1.149	78.080	
	300	84.586	8.222	9.894	1.723	117.120	
	400	112.782	10.962	13.192	2.297	156.160	
	500	140.977	13.703	16.489	2.871	195.200	

2.4 Micro-Particle Image Velocimetry

Particle image velocimetry, or PIV, consists on acquiring a fluid's velocity by following the motion of seeding particles. The working principle is fairly simple, the flow region to be analysed is illuminated, usually with a laser, and the seeding particles' movement is captured by taking photographs within a time-frame (Grant, 1997). The distance travelled by the particle, Δx , divided by the time between frames, Δt , gives the particle velocity, u . Having two-dimensional displacement data of a multitude of particles within the analysed flow area, gives us the flow velocity-vector field.

Micro-PIV, or μ PIV, varies slightly from standard, macroscopic PIV measurements. Wereley & Meinhart (2010) mention three main differences between the two. Firstly, in order for the flow not to be disturbed, the seeding particles must be sufficiently small. However, the particles' size must also be sufficiently large to scatter the light emitted by the laser. Thus, a compromise must be reached when considering particle size. Secondly, at such small scales, the effects of Brownian motion cannot be disregarded. Lastly, the illumination methods must be different. PIV measurements use a laser to create a planar sheet, defining the plane of measurement (Grant, 1997). The same cannot be successfully employed in μ PIV, and the whole volume of the flow is illuminated. Here, two techniques can be used to obtain quality measurements. One has the depth of focus out-scale the flow depth, focusing all particles in the flow. However, it then leads to significant post-processing efforts in order to analyse only the desired measurement plane. The other option, and the one used in our measurements, is to have a depth of

focus smaller than the flow depth². This leads to the particles in the desired depth of measurement to be within the focal plane, while all other particles are unfocused. There is one remaining, difference between standard PIV and μ PIV. The camera apparatus used to track the particles' movements must be coupled to a microscope for microscale measurements.

In this study, the μ PIV measurements were carried out similarly to the work of Rodrigues et al. (2020). Both fluids were seeded with 2 μ m of diameter, fluorescent tracing particles (Molecular ProbesTM FluoSpheres[®]; F8825 Nile red carboxylate-modified microspheres). For PEO4 a particle concentration of approximately 0.5 wt% was used, while for PEO8 it was 0.2 wt%. In order to avoid excessive adhesion of the particles to the PDMS or the microscope slide, sodium dodecyl sulphate (SDS) was added to the samples in a mass concentration of approximately 0.1 wt%. The samples were then placed in a roller/tilt mixer at low speed for a few hours before usage. Finally, the fluid samples were filtered with a Thermo ScientificTM NalgeneTM syringe filter of 10 μ m pore size to remove any fibres that may have been present, after, of course, verifying that the filtering process had no rheological impact on the samples. The entire sample preparation, and the μ PIV measurements, were carried out at a room temperature of around 23°C.

The setup used for all visualisations and measurements consisted of an inverted microscope (Leica Microsystems GmbH; DMI5000 M), equipped with a 20 $\times$ magnification lens (Leica Microsystems GmbH; HCX PL Fluotar with a numerical aperture of NA = 0.4). The laser used to illuminate the whole fluid volume was a double-pulsed Nd:YAG laser system, with 532 nm wavelength, and the images were taken with a CCD camera (Dantec Dynamics; FlowSense 4M MkII) of 2048 $\times$ 2048 px resolution at full frame (1 px = 7.4 μ m).

The channel's centre plane was identified by focusing its top and bottom and then averaging the two. This was done by illuminating the channel with a 100 W mercury lamp, and selecting the first and last depths at which fluorescent particles could be focused. When the centre plane was set, the scale factor needed to be confirmed³ and a mask applied, to later remove the values corresponding to the walls and cylinder boundaries. An example of the walls and cylinder masks can be seen in Figure 2.16.

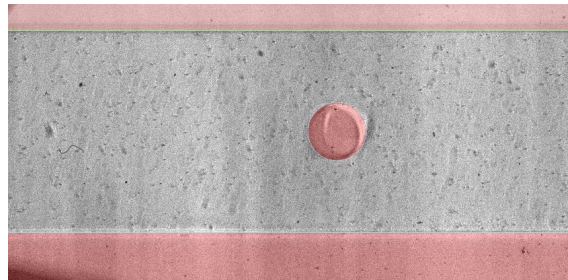


Figure 2.16: Defining the mask.

The μ PIV measurements were done by taking 100 image pairs and processing them

²Rodrigues et al. (2020) estimated a depth of focus $\delta_{z_m} \sim 30.7 \mu\text{m}$, for his experiments which correlated to approximately 30% of their flow depth (which was $H = 100 \mu\text{m}$, the same as ours). As the only difference between their study and ours are the fluids, a similar estimated depth of focus can be assumed for our case.

³The scale factor is simply the inverse of the magnification; since all visualisations were conducted at 20 $\times$ magnification, the scale factor was of 1/20.

using an average cross-correlation PIV algorithm (Dantec Dynamics; DynamicStudio v2.30). The pictures were taken with a trigger rate of 7.4 Hz, in double frame mode, synchronised with the laser pulses. The time between pulses was set for each measurement, depending on the flow rate; it was considered as the time that a particle would take to travel a fraction of the interrogation area (on our case, the whole measurement area was split into interrogation areas of 32×32 px), at the maximum streamwise velocity in the channel, i.e., the velocity in the contraction. It is, thus, given by the following expression:

$$\Delta t = f \frac{\Delta x}{U_C}, \quad (2.32)$$

where f is the chosen fraction (usually $1/4$) and Δx is the real length of the interrogation area, such that

$$\Delta x = 32[\text{px}] \times 7.4 \left[\frac{\mu\text{m}}{\text{px}} \right] \times \frac{1}{20}. \quad (2.33)$$

Thus, the typical displacement was $f\Delta x \sim 3 \mu\text{m}$.

The pictures were ensemble-averaged and masked into a single velocity vector field. Two vector fields were done, and later averaged, for each flow regime. Further image analysis, generation of contour plots and streamline traces is performed using Python.

A few steps, not as relevant to the understanding of the obtained results, were left out of this section for the sake of clarity. Although, a further in-depth description of the procedure we followed for the μPIV measurements and post-processing, that lead to the results shown in the next Chapter are described further in detail in Appendix A.

Chapter 3

Results and discussion

Having our data treated, we can analyse it. In the following Sections we will go through what information we want to gather, and how, and then analyse the plotted results, for both fluids, according to data-type. The experiments were carried out with two different microchannels which differ on the size of the pressure tap intake, so the results are presented and discussed as a function of the narrow pressure tap (NPT) and wide pressure tap (WPT).

3.1 Flow fields and velocity profiles

In order for our velocity fields to show the same magnitude range, the magnitudes of the velocity vectors were normalised by the maximum value found in the channel for each flow regime, thus the velocity fields show $0 \leq |\mathbf{v}|/|\mathbf{v}|_{\max} \leq 1$. This gives us a normalised look at the velocity magnitude throughout the channel. In order to analyse how the fluid actually flows around the cylinder, streamlines were also plotted, starting from equidistant points at the upstream boundary and travelling, without breaking, to the downstream end.

Valuable information can also be gathered from the axial velocity profiles at the cylinder centre plane. In order for us to attain consistent profiles, the axial velocity was normalised by its maximum value found at the centre plane, for each flow regime; thus, the profiles show $0 \leq u/u_{\max} \leq 1$. This already provides information about the axial asymmetry; nevertheless, in order to aid in visualisation of the "degree" of asymmetry, the asymmetry parameter was plotted against Wi . This parameter is given, as defined by Haward et al. (2019), through the following expression:

$$I = \frac{|u_2 - u_1|}{(u_2 + u_1)}, \quad (3.1)$$

where u_i denotes the axial velocities at the midpoints between the channel walls and the cylinder faces, at the centre plane ($x = 0$). We defined $y_1 = -(L_c + R)/2$ and $y_2 = +(L_c + R)/2$, but as our data takes discrete values, our u_i values were taken, not exactly at the midpoints, but at the nearer data points available to us. The asymmetry parameter may take values between $-1 \leq I \leq 1$, where perfect symmetry is represented by a value of 0.

In Figures 3.1 and 3.2 the velocity magnitude fields with streamlines and the axial velocity profiles for PEO4 are shown, respectively.

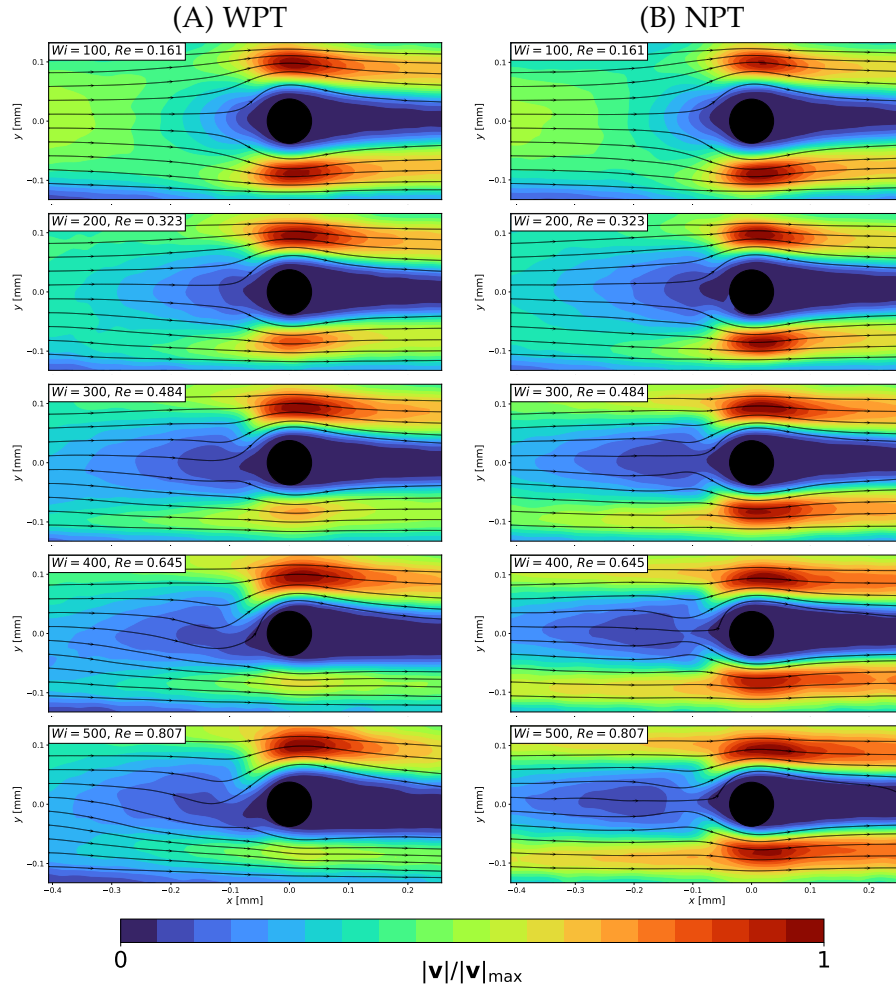


Figure 3.1: PEO4 velocity fields and streamlines obtained from microchannels with (A) wide and (B) narrow pressure taps.

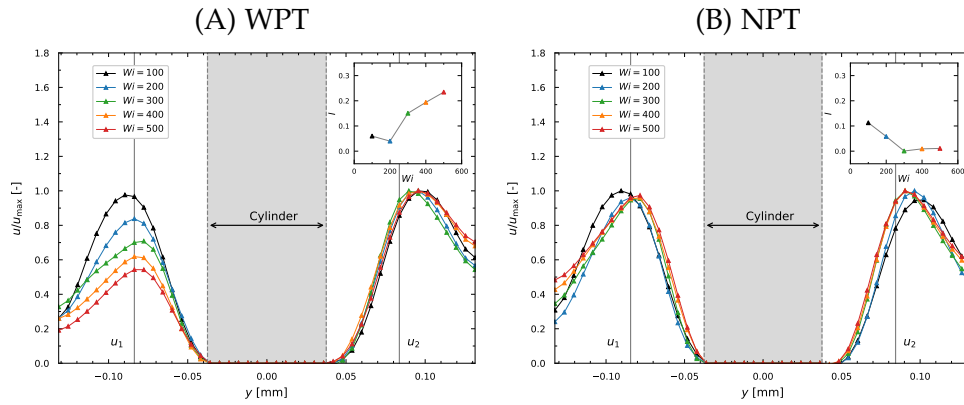


Figure 3.2: PEO4 velocity profiles at the cylinder centre plane and associated asymmetry coefficient, obtained from microchannels with (A) wide and (B) narrow pressure taps.

Looking firstly at the velocity fields for the wider tap intake (Figure 3.1A), for the initial Weissenberg of 100, the flow seems to be practically axially symmetric, revealing, however, lower velocities at the negative- y wall than at the positive one. Even at this Wi , already there is no fore-aft symmetry as an extended downstream wake and a slight upstream wake are already formed. For $Wi = 200$, a slight axial asymmetry seems to onset, with larger velocity magnitudes shown on the contraction opposite the upstream pressure tap. We cannot conclude on the length of the downstream wake region, as our data doesn't reach larger axial lengths, nevertheless, the upstream wake grows from $Wi = 100$ to 200. Further increasing Wi seems to aggravate the axial asymmetry and enlarges the downstream wake. The upstream wake also grows with Wi but, interestingly, it becomes "deformed", seemingly sprouting from the upstream pressure tap and bending upwards until it half contracts in a half-wineglass shape, also visible in the arching of the streamlines for Wi of 400 and 500. Analysing the velocity profiles at the centre plane (Figure 3.2A), the axial flow asymmetry is clearly growing with Wi , being the fluid velocity always larger on the positive- y contraction. The asymmetry parameter, I , however, shows a slight axial asymmetry for $Wi = 100$, but this is due to the positioning of the input velocities' (u_1 and u_2) coordinates, as the maximum velocities found in the curves are not always found at the contractions' midpoints. This also explains the reduced value for the asymmetry parameter for $Wi = 200$, as can clearly be seen that the flow profile is not symmetric. Despite these misleading values, I shows an obvious crescendo, reaching a maximum value of approximately 0.23 for $Wi = 500$. For non-shear-thinning fluids, no such axial asymmetry has been reported so far, neither has the half-wineglass shape of the upstream flow area, to the best of our knowledge.

Now looking at the narrower tap intake flows (Figure 3.1B), the flow remains essentially axially symmetric for all tested regimes, except, again, for the presence of lower velocities bound to the negative- y wall. For the initial Wi of 100, similarly to the WPT results, the flow presents fore-aft asymmetry due to down- and upstream wakes. From then on, increasing Wi creates a clear full wineglass profile, identifiable from the streamline curvature before the forward stagnation point, similar to the reported findings of Kenney et al. (2013), Galindo-Rosales et al. (2014) and Shi et al. (2015). However, the increase in streamline "pinch" and disconnection of the forward stagnation point reported by Kenney et al. (2013) were not found in our Wi range. The viscoelastic Mach number for the onset of our wineglass instability was between 8 and 12 (see Table 2.8), values similar to the critical Mach number reported by Shi et al. (2015) ($Ma = 10$). Varchanis et al. (2020) did not mention such instability for their PEO4 studies; however, their maximum Mach number¹, for PEO4 experiments, was approximately 1. Moreover, Shi et al. (2015) found that lowering the aspect ratio and raising the blockage ratio lead to a reduction of the critical Mach number for the onset of the instability, thus, for the channel used by Varchanis et al., the critical Mach number may be even higher than ours. Turning our eye now to the velocity profiles (3.2B), there is absolutely no noticeable axial asymmetry and the slight non-zero values for I are once again a product of a mispositioning of the maximum velocities found in either contractions.

Now moving onto PEO8's results, Figures 3.3 and 3.4 show, for both tap intakes, the velocity fields and profiles, respectively.

¹From Varchanis et al. (2020): $U_{\max}^{\text{PEO4}} = 0.02$ m/s that, inserting into our definition of the Reynolds number with their fluid and channel properties, corresponds to $Re_{\max}^{\text{PEO4}} \sim 0.0281$, and $Wi_{\max}^{\text{PEO4}} = 46$: $Ma = \sqrt{WiRe} \sim 1.14$.

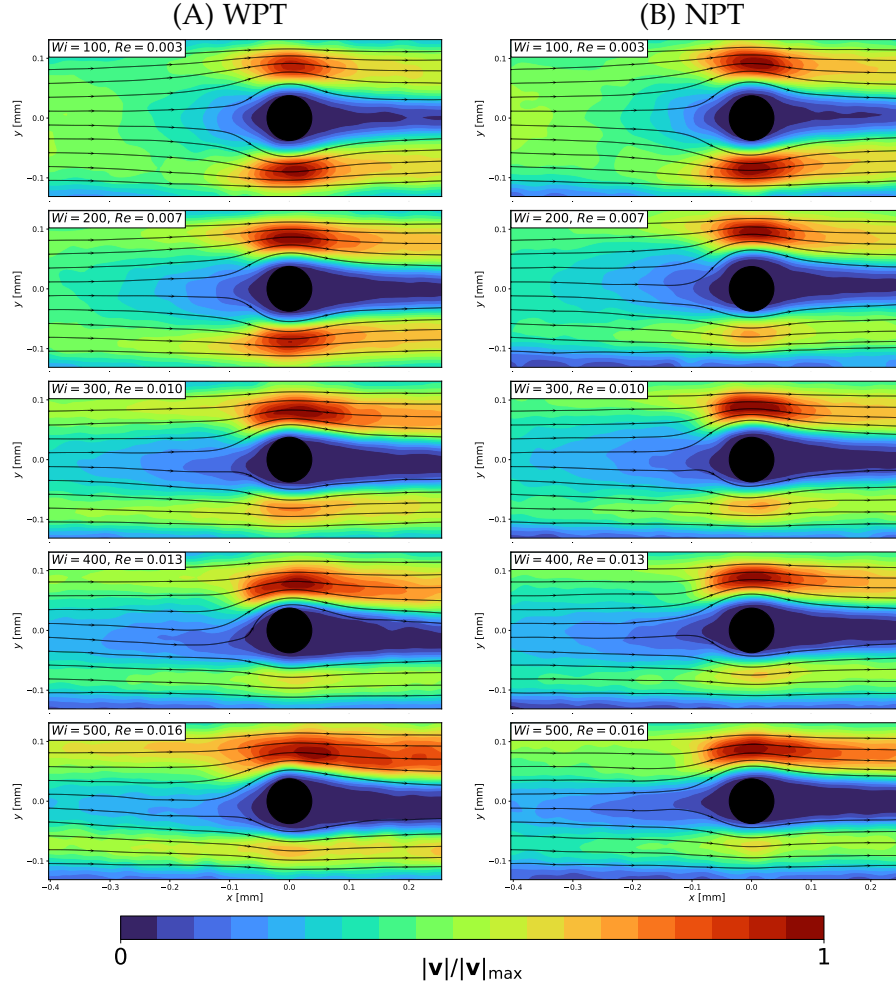


Figure 3.3: PEO8 velocity fields and streamlines obtained from microchannels with (A) wide and (B) narrow pressure taps.

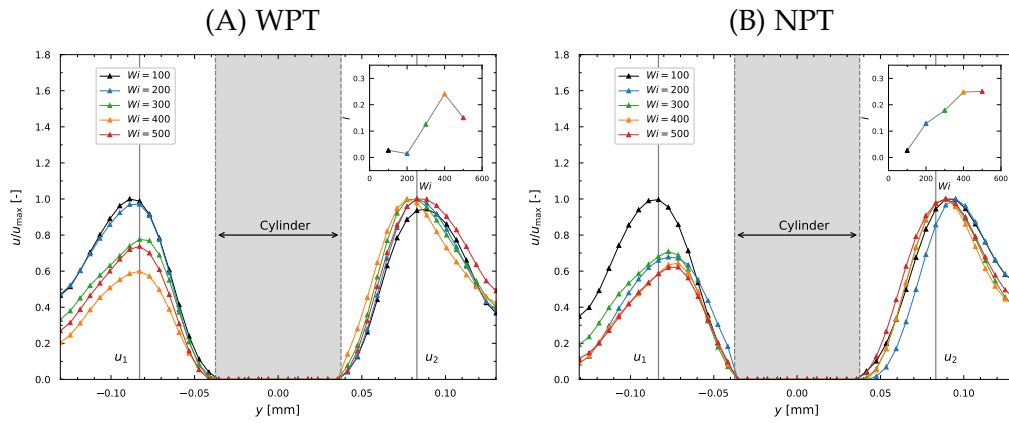


Figure 3.4: PEO8 velocity profiles at the cylinder centre plane and associated asymmetry coefficient, obtained from microchannels with (A) wide and (B) narrow pressure taps.

Starting again from WPT channel velocity fields (Figure 3.3A), for the initial Weissenberg of 100, the flow exhibits axial symmetry and wakes up and downstream from

the cylinder, being the latter significantly larger, similarly to both $Wi = 100$ flows for PEO4. Increasing Wi to 200 both wakes seem to grow. An axial asymmetry onsets only for $Wi = 300$ and grows with its further increase to 400, where it reaches a maximum value of 0.24 for the asymmetry parameter. For a Weissenberg of 500, the flow at the negative- y contraction seems to softly speed up, leading to a slight regain of symmetry. Little can be said with certainty about the evolution of downstream wake, as we have no further-downstream data, but the upstream wake seems to grow much slower than for PEO4, and doesn't seem to reach dimensions as large, it actually slightly decreases when Wi is increased from 400 to 500. Additionally, there is no relevant bending of the upstream wake as was the case for the same tap intake for PEO4 (Figure 3.1A). The velocity profiles (Figure 3.4) are regular and, together with the asymmetry factors, show the same behaviour as described above.

For the NPT flows, the velocity fields (Figure 3.3A) are quite similar to the former. Notably, the axial flow asymmetry now onset earlier, at $Wi = 200$, and aggravated with Wi until stabilising at an asymmetry parameter of 0.25, but without regaining symmetry. The upstream wake also does not seem to diminish for $Wi = 500$. For Wi of 400 and 500, there is also less streamline curvature upstream of the cylinder than for WPT measurements.

PEO8's behaviour is very similar for both pressure tap intake designs, showing extended downstream wakes and lateral flow asymmetry growing with Wi , as reported by Ribeiro et al. (2014), Nolan et al. (2016), and Haward et al. and coworkers. No evidence was found of the separation of the forward stagnation point nor recirculation vortexes anywhere in the channel, similarly to the findings of Rodrigues et al. (2020) who attributed this absence to the low blockage ratio of the channel and their fluids' low elasticity, despite this last not being as applicable to our case. Moreover, in accordance with the results obtained by Nolan et al. (2016) and Rodrigues et al. (2020), our axial flow asymmetry onsets at approximately unitary viscoelastic Mach numbers, which has been reported to be connected with elastic instabilities (Nolan et al., 2016). For WPT the asymmetric flow appears between $Ma = 1.149$ and 1.1723 ; while for NPT, it onsets between 0.574 and 1.149 , accordingly to the previously mentioned Weissenberg numbers (Table 2.8).

3.2 Local viscosity

Important information about the influence of viscosity, particularly shear-thinning, on the flow, can be gathered by creating viscosity fields, similarly to the work of Nolan et al. (2016). The fitting quality of our Carreau–Yasuda models now becomes vital, as we can use them to compute the local viscosities from local shear rate values. We shall use as input for our viscosity models the magnitude of the local shear rate, which can be calculated through the deformation rate tensor (Bird et al., 1987) as

$$\dot{\gamma} = \sqrt{(1/2)(\mathbf{D} : \mathbf{D})} = \sqrt{(1/2)\text{tr}(\mathbf{D}^T \mathbf{D})} = \sqrt{(1/2) \sum \mathbf{D}_{ij}^2}, \quad (3.2)$$

where the deformation rate tensor, $\mathbf{D}$, represents the symmetric component of the velocity gradient, whereas the vorticity tensor, $\mathbf{\Omega}$, to be used later on, the anti-symmetric

component, which are given by Equations 3.3 and 3.4, respectively.

$$\mathbf{D} = \frac{\nabla \mathbf{v} + \nabla \mathbf{v}^T}{2} \quad (3.3)$$

$$\mathbf{\Omega} = \frac{\nabla \mathbf{v} - \nabla \mathbf{v}^T}{2} \quad (3.4)$$

Additionally, from the local shear rates, we were able to gather both fluid's experimental ranges to evaluate our viscosity working range, namely, at which viscosity curve regions we are working. The shear rate ranges consist on the minimum and maximum shear rates found for each fluid, independently of the configuration of the microchannel (pressure tap size). These ranges can be seen in Figure 2.4 through shaded areas.

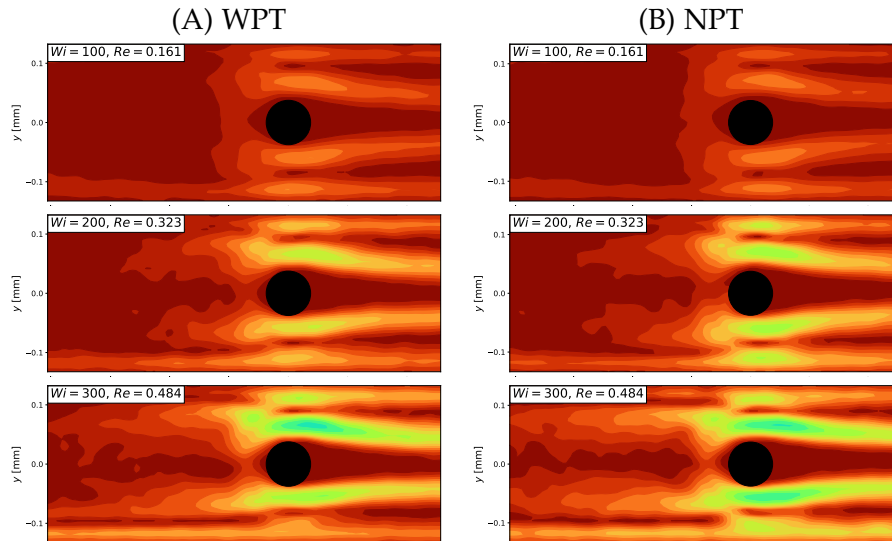
After gathering the local viscosities from the shear rates, we can normalise them between the viscosity plateaus using the models' η_0 and η_∞ as described in the following expression (Nolan et al., 2016):

$$\eta^* = \frac{\eta(\dot{\gamma}) - \eta_\infty}{\eta_0 - \eta_\infty}, \quad (3.5)$$

being η^* the normalised viscosity and ranging from 0 ($\eta = \eta_\infty$) and 1 ($\eta = \eta_0$). This gives us insight into the fluid's shear-thinning across the flow.

Before moving onto the plotted fields, it is noteworthy to say that, in order to better visualise the viscosity variations, the shown normalised viscosity range was set for each fluid. The ranges were defined by the minimum and maximum values, found for each fluid, rounded to the decimals. The minimum value was rounded down and the maximum rounded up, to guaranty that the values depicted in the viscosity fields were within range.

In Figure 3.5 the viscosity fields for PEO4 are shown for both pressure tap intakes.



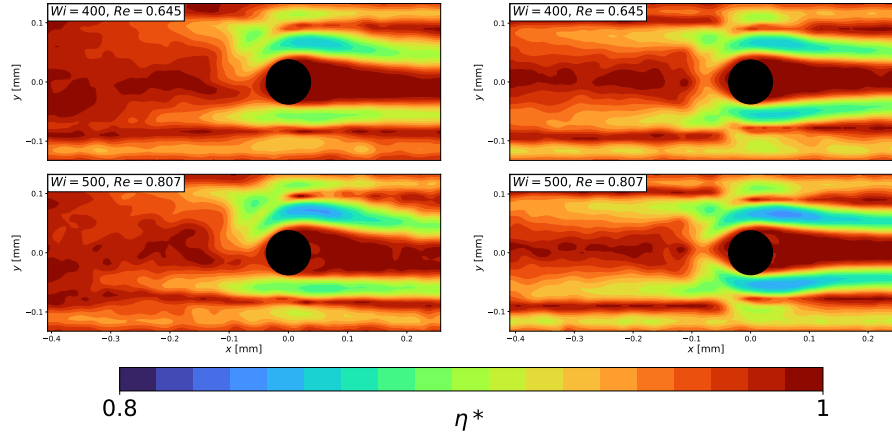


Figure 3.5: PEO4 viscosity contour plots obtained from microchannels with (A) wide and (B) narrow pressure taps.

PEO4's normalised viscosity ranges only between 0.8 and 1, meaning the local viscosities fall near the high-shear plateau (which could also be gathered from the intersection of the shear rate band with its Carreau–Yasuda model in Figure 2.4). Moreover, being PEO4 an almost non-shear-thinning fluid, this normalised viscosity range relates to a minimal viscosity variation within the flow ($0.0217 \leq \eta \leq 0.0256$ Pa·s), as was expected.

Analysing the WPT viscosity fields (Figure 3.5A), for $Wi = 100$ the flow displays high viscosity ($\eta^* = 1$) upstream of the cylinder, except for a thin slightly-lower viscosity region near the negative- y wall. The fluid suffers shear when approaching the cylinder, lowering its viscosity in the high velocity gradient areas, i.e., near the cylinder face and the walls. From there, the wall shear is more significant (still more so at the wall containing the upstream tap) and the interface between the downstream wake and the higher velocity region also leads to viscosity variations (due to higher velocity gradients) creating two axially symmetric sets of lower-viscosity bands that continue downstream. When raising Wi to 200, at the walls upstream of the cylinder the viscosity values begin to abandon the high-shear plateau, starting to resemble the shape of slug flow, as described by Nolan et al. (2016), with a slow, higher-viscosity central flow. Also, lower viscosity bands start to appear at the interface between the upstream wake and the rest of the surrounding flow. For $Wi = 300$, lower-viscosity spots start to appear before the positive- y contraction and noticeably lower viscosities are found at this contraction than at the opposite one, as the axial flow asymmetry onsets and the velocities at the contraction diametrical to the upstream tap are raised. Additionally, the upstream wake's bending towards the pressure tap also shows and a larger high-viscosity area can be seen at the positive- y half of the channel. Raising Wi even further, the cylinder-face low-viscosity band bends toward the forward stagnation point and the aforementioned spot grows as the fluid slithers in the half-wineglass shape seen in Figure 3.1A. As the aforementioned band begins to bend towards the stagnation point, the analogous, opposite band begins to straighten, tending to become parallel with the channel walls with Wi .

For the NPT flows, the behaviour is the same as the last up until $Wi = 300$, there, the appearance of the low-viscosity spot, and the following bending of the cylinder-face-shear band are mirrored axially, as the half-wineglass shape shown for WPT is replaced

by the full glass shape.

The minimum viscosities are reported in the contraction bands slightly downstream of the cylinder, where the high-velocity fluid abandoning the contractions meets the slow-moving fluid of the downstream wake.

Now for PEO8, the viscosity contour plots can be seen in Figure A.3.

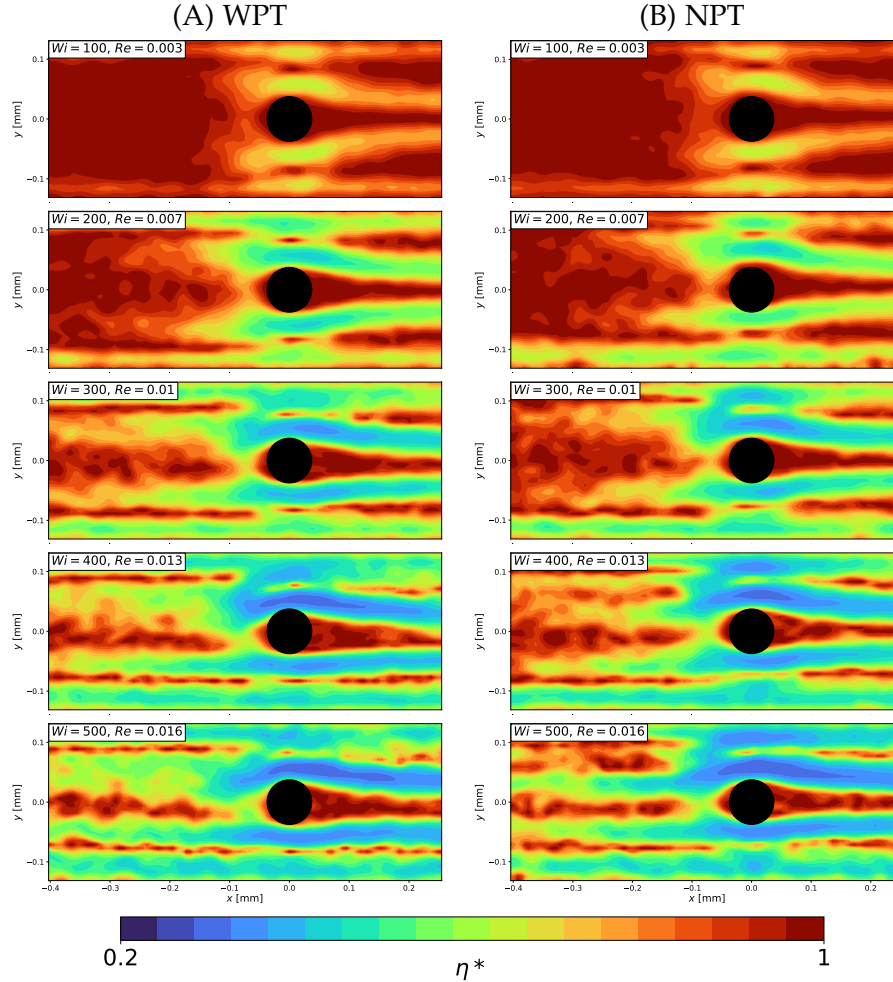


Figure 3.6: PEO8 viscosity contour plots obtained from microchannels with (A) wide and (B) narrow pressure taps.

Contrary to PEO4, PEO8's normalised viscosity varies significantly, from 0.2 to 1 in the experimental shear rate range (see again Figure 2.4). Furthermore, being PEO8 a shear-thinning fluid, this normalised viscosity variation relates to a substantial viscosity range: $0.0573 \leq \eta \leq 0.2826$ Pa·s. Compared to PEO4's viscosity range, PEO8's is almost 60 times larger.

Starting once more from the wider tap intake flows (Figure 3.5A), for the lowest Wi , upstream of the cylinder, the flow near the walls already presents lower viscosities ($\eta^* \sim 0.8$), again resembling slug flow. Reaching the contractions, the shear rate increases from the interaction with the cylinder face and the channel walls, leading, again, to the formation of axially-symmetric low-viscosity bands that further propagate downstream from the fast-moving fluid's interaction with the wake. At this Wi of 100, the minimum viscosity, which is always felt at the end of the contractions, was $\eta^* \sim 0.7$. As Wi is

increased to 200, the upstream wake becomes relevant, showing the beginning of low viscosity bands at its interaction with the faster-moving fluid. The minimum viscosity reaches, at this Wi , $\eta^* \sim 0.5$. Increasing Wi even further, the positive- y contraction begins to show lower viscosity values, either at the cylinder face or at wall, due to the flow asymmetry. Downstream, the wake-interaction low-viscosity band seems to be slightly larger than the negative- y , analogous band. This straightening of the contraction band creates a thin region of fast-moving, high-viscosity fluid, lubricated on both sides, from the up and downstream wakes' and the wall's low-viscosity regions. Upstream, the y -positive half of the channel shows a larger low-viscosity area as the streamlines bend towards the contraction. When Wi reaches 500, the viscosity at the contraction wall raises slightly and the negative- y contraction, low-viscosity bands seem to weakly grow in intensity.

For the NPT channels (Figure 3.5B) the differences between contraction viscosities are noticed sooner, at $Wi = 200$, in agreement with the earlier asymmetry onset. The flow, then, follows a similar trend as for the WPT case up until $Wi = 400$, where, upstream of the cylinder, the large, low-viscosity area, placed at the positive- y half of the channel for WPT, is replaced with two lesser stream-wise-oriented low-viscosity regions separated by thin, slightly undefined higher viscosity area. Increasing Wi to 500 reduces the size of the low-viscosity separator and the up and downstream data borders become "connected" with two extended low-viscosity bands, positioned, almost axially symmetric, on either side of the cylinder. The viscosity near the positive- y wall is also significantly lower than for the equivalent opposite wall.

3.3 Flowtype parameter

Lastly, we sought to compute the flowtype parameter fields. This parameter, ξ , gives information about the contributions of rotation, shear and extension in the flow (Nolan et al., 2016; Rodrigues et al., 2020). It can be locally calculated through Equation 3.6 with the magnitudes of the aforementioned, deformation rate tensor, $|\mathbf{D}|$, and vorticity tensor, $|\mathbf{\Omega}|$, which are respectively given by Equations 3.7 and 3.8 (Nolan et al., 2016; Rodrigues et al., 2020).

$$\xi = \frac{|\mathbf{D}| - |\mathbf{\Omega}|}{|\mathbf{D}| + |\mathbf{\Omega}|} \quad (3.6)$$

$$|\mathbf{D}| = \sqrt{2(\mathbf{D} : \mathbf{D})} = \sqrt{2 \text{tr}(\mathbf{D}^T \mathbf{D})} = \sqrt{2 \sum \mathbf{D}_{ij}^2} \quad (3.7)$$

$$|\mathbf{\Omega}| = \sqrt{2(\mathbf{\Omega} : \mathbf{\Omega})} = \sqrt{2 \text{tr}(\mathbf{\Omega}^T \mathbf{\Omega})} = \sqrt{2 \sum \mathbf{\Omega}_{ij}^2} \quad (3.8)$$

A flowtype parameter of -1, 0 and 1 relate, respectively, to pure rotational flow, pure shear flow and pure extensional flow.

Starting again with PEO4, in Figure 3.7 are shown the flowtype parameter fields for both pressure taps.

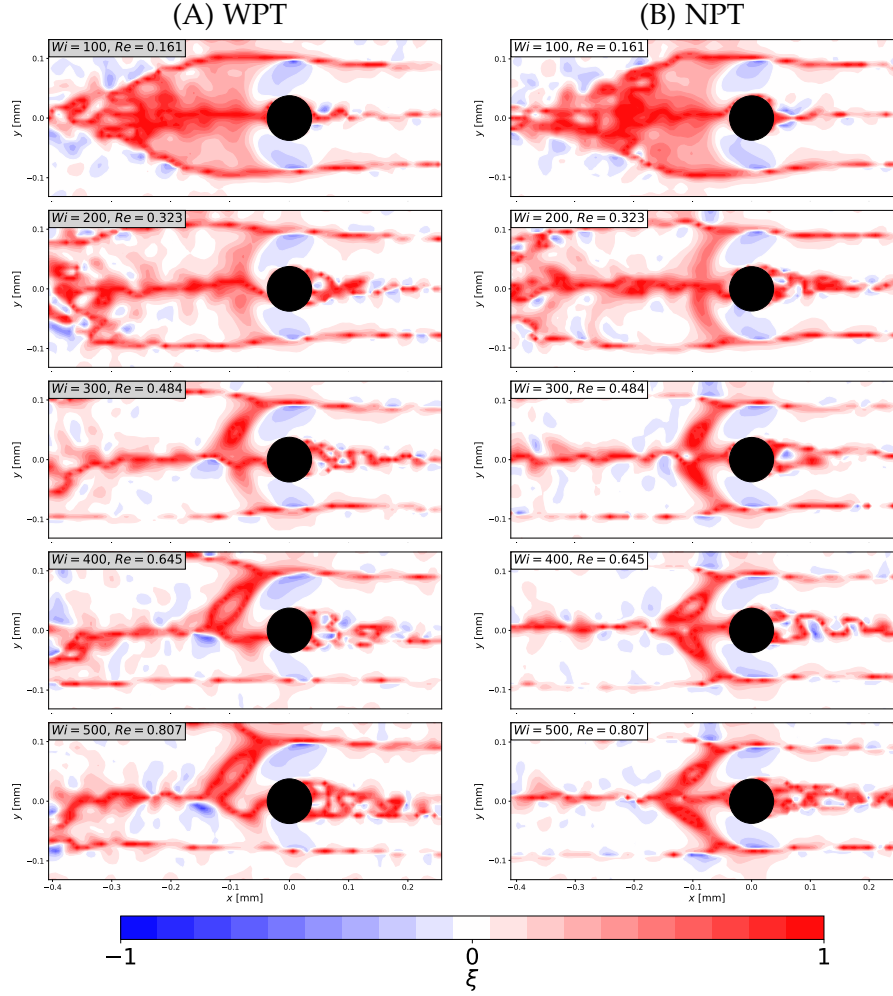


Figure 3.7: PEO4 flowtype parameter contour plots obtained from microchannels with narrow pressure taps (A) before and (B) after *smoothn*'s application.

Beginning, once more, with WPT (Figure 3.7A), for the initial Wi of 100, three streams of extensional flow can be seen up and downstream of the cylinder, which are related to the elongational stresses of the stagnation points and the extension suffered by the fluid as it stretches past the wall/cylinder gaps. Upstream of the cylinder, there is still a considerable area of extensional flow, which will be replaced with shear flow as Wi increases, due to the shear provoked by the upstream wake's velocity gradients. Near the forward stagnation point, though, extensional flow regions remain between the channel stream-wise axis and the contractions, as the fluid stretches along the bent streamlines towards the gaps. Downstream, the flow is shear-dominated as the wake is already formed. Near the top and bottom of the cylinder there can be seen rotational flow areas of identical shapes. When Wi is increased, and flow asymmetry onsets, the extensional flow region connecting the middling and positive- y streams grows in intensity, as the fluid flows progressively more through the gap opposite the upstream tap, speeding up to maintain the imposed flow rate. Additionally, with increasing Wi , the positive- y extensional stream progressively nears the wall upstream of the cylinder. This is due to the growth and bending of the wake (shear-dominated region), which causes the high velocity, extensional flow that is usually aligned with the centre of

the gap, to shift upwards. Moreover, another extensional flow region can be seen, for Weissenberg numbers from 300 to 500, connecting the middling and lower extensional streams near the upstream pressure tap intake. Nevertheless, it is unclear if this is connected to the pressure tap, or not, as we don't have access to further-upstream data.

Looking now at the NPT flows (Figure 3.7B), the flowtype fields are similar to the WPT ones up until the onset of flow asymmetry for the latter. From then, both extensional regions between the contractions and the mid-stream are enhanced as the fluid flows equally through both gaps. The contraction-bound extensional streams slightly bend before the gaps, possibly from streamline curvature induced by the wineglass instability.

Now for PEO8's results, in Figure 3.8 are presented the flowtype parameter fields, for both pressure tap intakes.

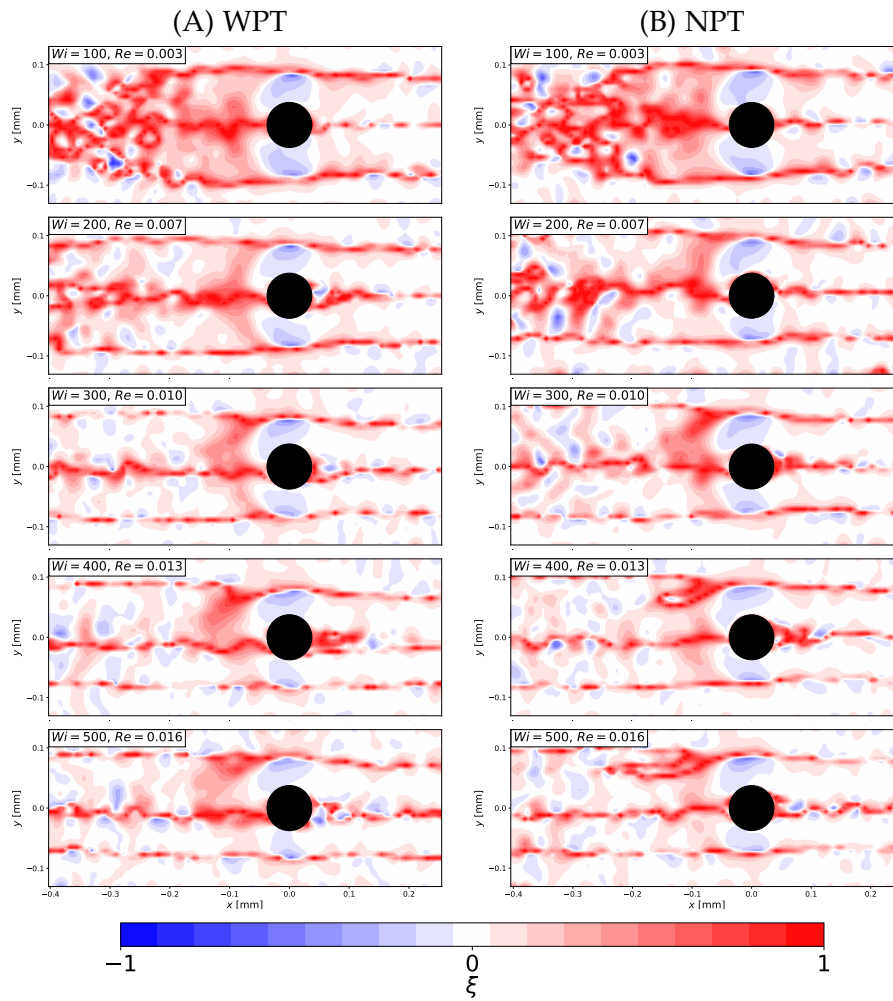


Figure 3.8: PEO8 flowtype parameter contour plots obtained from microchannels with narrow pressure taps (A) before and (B) after *smoothn*'s application.

For PEO8, the flowtype parameter fields for both taps are quite similar. They present the same similar rotational flow areas on the gaps, near the cylinder face, that become asymmetrical with Wi ; three extensional streams up and downstream of the cylinder; shear-dominated downstream areas for all Wi ; extension-dominated upstream areas for $Wi = 100$ but decaying into shear with increasing Wi , and slightly more intense ex-

tensional flow between the forward stagnation and the positive- y contraction than the negative one. The only noticeable difference between the two sets of flowtype fields is the shape of the extensional flow region between the middling and positive- y extensional streams. Observing the fields of $Wi = 400$ and 500 for both taps, one can see that for NPT the extensional region has a much more complex shape.

Chapter 4

Remarks and further work

In this work, velocity vector fields were obtained through micro-Particle Image Velocimetry, for the flow around a confined microfluidic cylinder of a practically non-shear-thinning, elastic fluid and a viscoelastic, shear-thinning fluid. The fluids were rheologically characterised in terms of shear viscosity and relaxation time, and the microfluidic devices were fabricated with PDMS, through standard soft-lithography techniques. The μ PIV vector fields were treated to compute velocity fields and profiles and local viscosities and flowtype parameters, granting us varied information about the flows, which was shown and discussed in Chapter 3.

From PEO4's results we can gather that the pressure taps with the widest intake influence the flow significantly; so substantially, in fact, that it leads to a flow asymmetry, with a velocity at one side of the cylinder almost 60% higher than at the other ($I = 0.23$ for $Wi = 500$), when the counterpart flow for the narrow taps intake was perfectly symmetric (recall Figure 3.2A). This conveys the notion that the flow asymmetry we witnessed for PEO4 should not be associated with an instability per se, but with a flow perturbation instead, as it is not a result of the rheological properties, but of the geometry. Interestingly, despite the tap intakes' influence, the fluid managed to partially retain the expected flow pattern, showing a half-wineglass shape, visible in Figure 3.1A for $Wi = 400$ and 500 . The viscosity variations were negligible ($0.0217 \leq \eta_{\text{PEO4}} \leq 0.0256$ Pa·s), as was expected since it is an almost non-shear-thinning fluid; thus, we can disregard any kind of viscosity variation influence on the asymmetry felt for PEO4. The viscoelastic Mach number for the onset of this asymmetry lies in the range between 4 and 8. The wineglass instability, observed for the narrow pressure tap intakes (Figure 3.1B), onset at a viscoelastic Mach number between 8 and 12, in agreement with previous studies (Shi et al., 2015; Kenney et al., 2013), and endured until the highest Wi , with no evidence of stagnation point separation from the cylinder face, nor upstream vortex formation. Shi et al. (2015) accredits the onset of his instability to flow perturbations, originary from a precursor, downstream instability. This downstream instability is identifiable from disordered and temporally varying streamlines downstream of the cylinder and seems to onset at $Ma = 1$. We have no data to evaluate the existence of this downstream instability, but, the streamline curvature and the upstream wake bend lead us to believe that the cause for this flow asymmetry is indeed the influence of the wide entrance pressure taps.

This flow asymmetry found for PEO4 raises questions about the nature of the PEO8 asymmetries. It was our original thought that PEO8's flow asymmetries were born of

downstream flow perturbations and triggered by an interplay between elasticity and shear-thinning. However, now that, at least for the WPT channels, the shear-thinning necessity may have been removed, we must wonder if the observed asymmetries with PEO8 were indeed elasticity-driven, or, similarly to PEO4, a product of a flow perturbation from the geometry. The first thing that would make us disregard PEO8's asymmetries as pressure-tap-driven is the absence of upstream-wake curvature towards the pressure tap, simply because that is the most striking visual clue that we can gather from PEO4's WPT velocity fields. Besides that, the fact that PEO4's NPT flows seem unaffected by the tap's intake could make us think that that is also the case for PEO8. However, there are still effects of the taps on the flow field, such as lower velocity values at the upstream-tap's wall, and more significative near the tap intake, and the fact that, for every measurement done the highest velocity value was found in the contraction opposite the upstream tap, even if it was mentioned in the literature that, for the reported instability, the fluid chooses its preferential path seemingly at random (Haward et al., 2019; Varchanis et al., 2020). Nevertheless, even if with small differences between the two PEO8 datasets, both look quite similar to the results on the instability in question present in the literature (Varchanis et al., 2020; Haward et al., 2021). When comparing our asymmetries to the results of Varchanis et al. (2020), our higher critical Weissenberg numbers for the onset, Wi_c , and the lower degrees of asymmetry, I , can all be related to the differences between the used channel dimensions. It is true that our higher blockage ratio should both lower the Wi_c and raise the maximum asymmetry parameter (Varchanis et al., 2020), however, our much higher channel confinement (lower α) leaves the flow susceptible to three-dimensional effects and should raise Wi_c and reduce the asymmetry intensity (Ribeiro et al., 2014). Moreover, the viscoelastic Mach numbers for the onsets of the asymmetries with PEO8 are near the critical reported value in the literature: $Ma_c \sim 1$. The local viscosity fields showed lower viscosity values on one of the two cylinder/wall gaps, but this could be both the driving mechanism, or a consequence of the flow asymmetry. Therefore, there is no definite proof about the nature of the flow asymmetries found for PEO8.

Further experiments would be needed in order to elucidate whether the effect of the pressure tap width is the responsible for the onset of the flow asymmetry encountered with PEO8. In this sense, the same microchannels, now without pressure taps, were designed and the SU-8 moulds were ordered, the results will be included in a future publication of this work. Furthermore, it would also be of interest to study the influence of single pressure tap up and downstream, as our work studied the combination of both. From our microchannels, though, more information about the flow around the pressure tap intakes could still be gathered. With the selected experimental setup, the pressure tap intakes were not visible and thus, it may prove elucidating to conduct the same analysis process, locally, specifically for each pressure tap intake, thus gathering the missing information about their effects on the encountered flows.

All in all, the effects of the pressure taps may actually be a very useful field of study. A previous work carried out by Rodrigues et al. (2019), who designed our microchannels, dealing with the determination of the optimal pressure tap design, concluded that our wide-intake pressure tap (WPT) was the best design for proper pressure measurements, with errors, for some configurations, lesser than 10%, when comparing to the analytical solution. In contrast, the narrow-intake pressure taps (NPT), performed sub-optimally, with errors, for some configurations, between 10% and 30%. Therefore, it

seems that a compromise can be achieved between the quality of the pressure measurements and the influence on the flow, opening new possibilities for future research.

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Appendices

Appendix A

Micro-PIV experimental procedure and postprocessing

This appendix serves as complementary reading to Section 2.4, with as little repetition as possible. Here we briefly describe the most practical steps taken in the μ PIV measurements, may them be experimental or in postprocessing.

Having the fluid sample homogeneous with fluorescent particles, with dissolved SDS and being filtered, the syringe was cleaned, filling it with water and soap three times, in order to remove any impurities and fibres. Then, the same was done with only water, to remove the soap. The syringe was then thoroughly and carefully dried with air and filled with the fluid sample very slowly, not to produce excessive extensional tensions. It was then placed and fixed in the syringe pump. The aforementioned Tygon® tubes and precision tips (recall the beginning of Section 2.3) were also cleaned by pumping water through them with a plastic syringe; this cleared the channel of any fibres or impurities that may have been within, and removed any remaining viscoelastic fluid that could have been ill-cleaned after the previous use. After all tubing was clean, it was connected to the syringe and small flow rate was applied to remove any water or air either from the syringe tip, or the tube. Then, the precision tip was connected to the inlet of the channel and the outlet was equally bound to a plastic cup for discharge. Then, the microchannel was positioned, and fixed to the microscope, in such a way that the camera output was horizontally aligned, facilitating the postprocessing to be done later. Furthermore, the channel was positioned so that the cylinder was visible, of course, and that we could observe a larger portion of the channel upstream from it, as we were interested in the flow behaviour upstream of the cylinder. Again, Figure 2.15 shows the experimental setup. Then, the centre plane was identified with a mercury lamp as was described in Section 2.4. The scale factor was then set, and the mask applied by taking a single-frame picture of the channel was taken and saved for calibration. From this image, the scale factor was confirmed by setting the distance between left-most upper and lower points of the channel walls for 0.27 mm. Then the channel walls and the cylinder were manually masked. After this preparation, the μ PIV measurements were done. After finishing the measurements, the syringe and Tygon® tubes were, again, thoroughly cleaned and then stored. The microchannels used in the session were marked and discarded not to compromise future measurements.

From the μ PIV software, the data was exported into CSV files and then loaded and treated with Python. Firstly, the data was trimmed, removing all values corresponding

to the channel walls, in order to guaranty that the analysed data related only to the flow inside the channel. This was done by disregarding all beginning and ending, constant-y lines that contained at least one cell with velocity magnitude of value zero ($|\mathbf{v}| = \sqrt{u^2 + v^2} = 0$). From the moment that a line with no zero values was found, the channel wall was defined. Then, the cylinder centre was located by finding the line (y) and column (x) that contained the maximum number of values of $|\mathbf{v}| \sim 0$. In fact, the data did not contain values of absolute zero in the area concerning the cylinder, thus, we treated values of $|\mathbf{v}| \lesssim 10^{-5}$ m/s as approximately zero (the minimum mean velocity in our channels is of 9.7911×10^{-2} m/s, as seen in Table 2.8). The reason that there still are velocity values in the area concerning the cylinder is, possibly, a product of the software's vector calculation and was, perhaps, aggravated by an ill fit of the initial mask, which was done manually (see Figure 2.16 - B), and by the fact that only the centre plane was focused, causing residual values from particles sliding along the cylinder face at other depths. To avoid having our code select the cylinder centre at the walls (as these also contain velocity values lower than 10^{-5} m/s), some of the first and last, constant-y lines were disregarded. Having found the cylinder centre, the x and y data was altered, to shift the origin¹ to it ($x = x - x_{\text{cyl}}$ and $y = y - y_{\text{cyl}}$). Moreover, now we could re-mask the cylinder with null values, which allowed for the plotted streamlines, to be discussed further on, not to be influenced by aforementioned residual values.

Having our new referential originating from the cylinder centre and knowing that the walls distance $L_c = 135 \mu\text{m}$ from it, the data was once again adjusted. The reason this last step was done was the same for the cylinder mask, avoiding residual velocity values originated from poor masking and particle depth, and also to guarantee that the channel's axial symmetry is preserved in our analysis.

Lastly, a penalised least squares based algorithm (*smoothn*) was used to treat the velocity vectors, yielding smoother results (Garcia, 2010). This was the same algorithm employed by Rodrigues et al. (2020) and was only applied to the velocity fields (u , v and $|\mathbf{v}|$), being the smoothing of the remaining properties only a result of their dependence on velocities. The algorithm allows for the user to input a smoothing parameter, s , which correlates to the "strength" of the smoothing effects. A parameter of $s = 1$ was used for all shown results, which relates to a gentle smoothing, except for the streamlines, which required s to take a value of two in order to obtain quality lines with no abrupt endings mid-channel.

We shall go over PEO8's results for narrow pressure tap channels to exemplify the algorithm's effects on the obtained results. In Figures A.1 to A.4 are shown, respectively, the axial velocity profiles, the velocity fields and streamlines, the local viscosity fields and the flowtype parameter plot are compared before and after *smoothn*'s application.

¹The initial origin was located manually at the upstream end of the lower channel wall (see Figure 2.16(A))

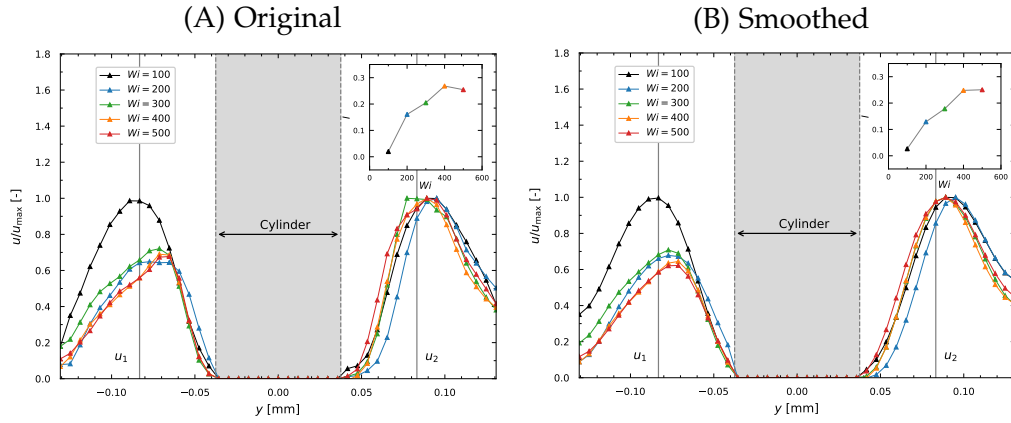


Figure A.1: PEO8 velocity profiles at the cylinder centre plane and associated asymmetry coefficient, obtained from microchannels with narrow pressure taps (A) before and (B) after *smoothn*'s application.

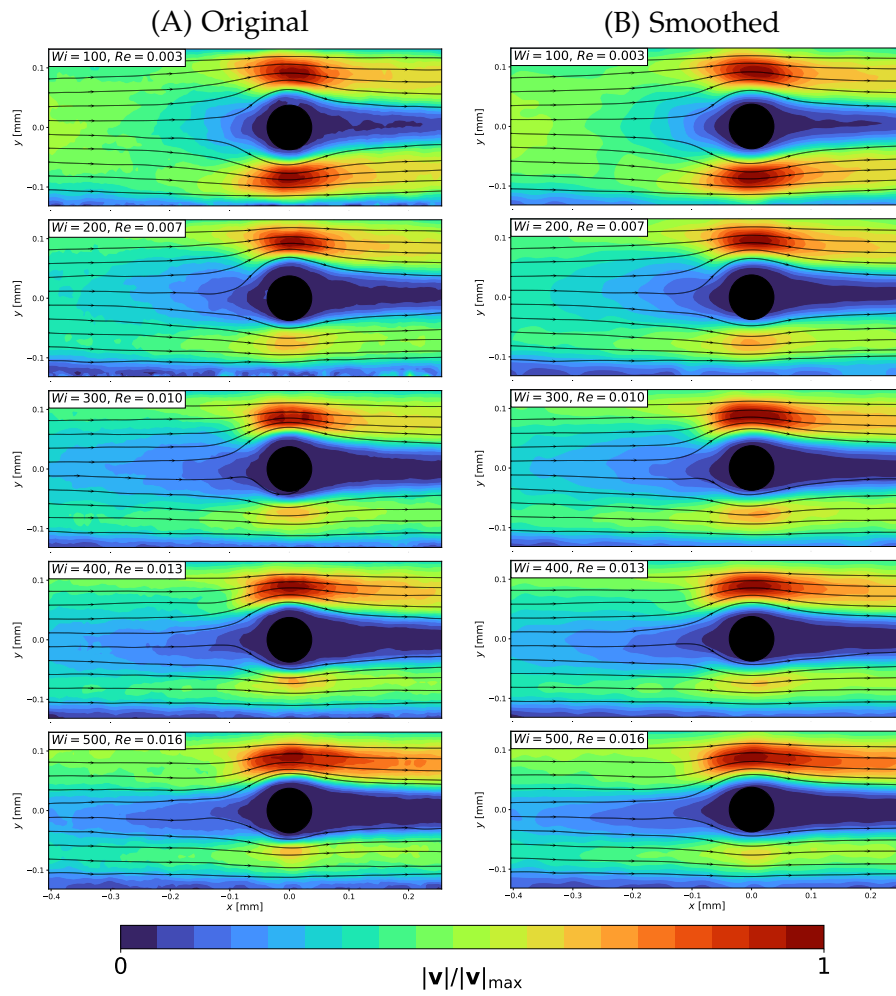


Figure A.2: PEO8 velocity fields and streamlines obtained from microchannels with narrow pressure taps (A) before and (B) after *smoothn*'s application.

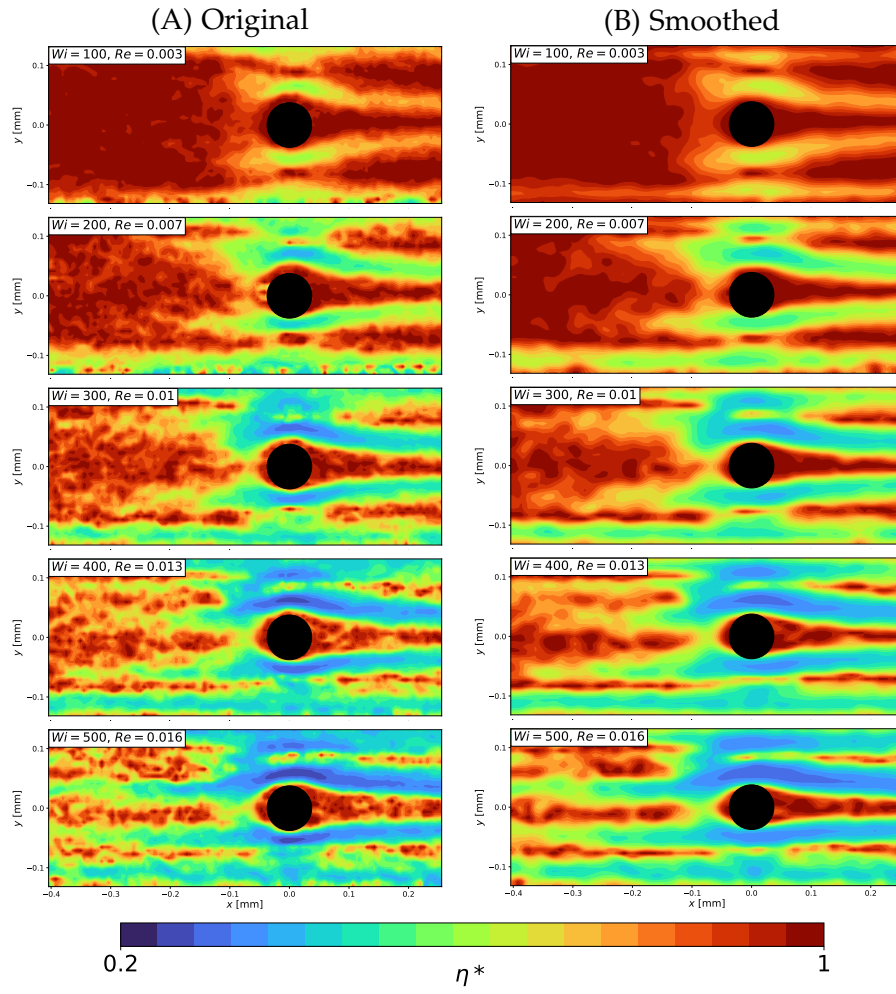
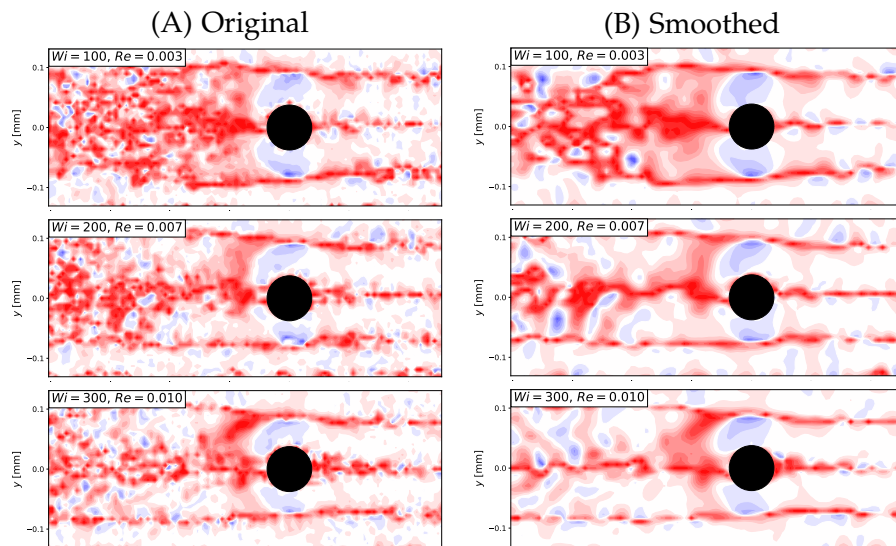


Figure A.3: PEO8 viscosity contour plots (A) before and (B) after *smoothn*'s application.



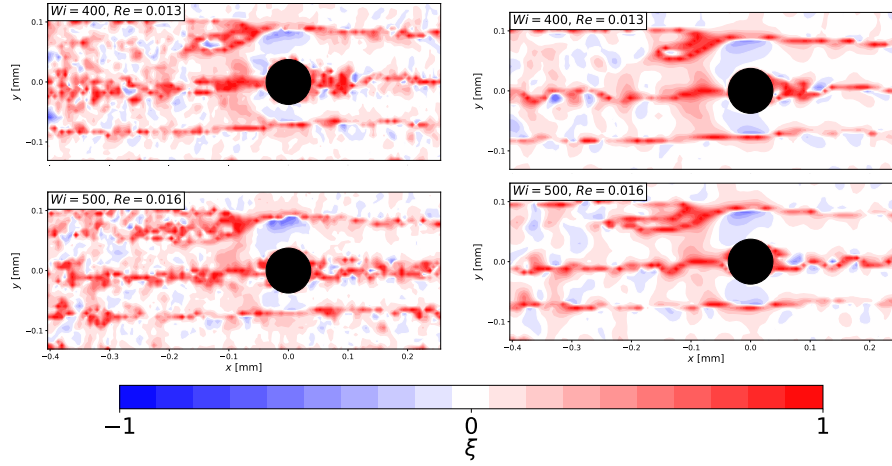


Figure A.4: PEO8 flowtype parameter contour plots (A) before and (B) after *smoothn*'s application.

Considering the velocity profiles first (Figure A.1), *smoothn*'s effects are clear, the curves are neatly smoothed into the usual expected shapes, affecting very slightly the asymmetry parameters. The algorithm may have a double-edged effect on our data, reducing potential errors from the μ PIV measurements while itself inducing error; nevertheless, looking at the velocity profiles plotted by Haward et al. (2019), it is clear that our smoothed results are much closer. Also, the smoothing parameter, s , was kept low, in a trade-off between visualisation aid and data compromise and, in truth, *smoothn*'s effects seem to mainly adjust data spikes.

Concerning the velocity fields (Figure A.2), the algorithm is quite useful in reducing noise and uniforming the results without significantly altering them. For the streamlines it was found critical, allowing us to create smooth, continuous lines from side to side without altering their density over the field, even when the original streamlines ended abruptly, which happened several times (as can be seen in Figure A.2 for $Wi=300$).

The viscosity and flowtype parameter fields (Figures A.3 and A.4) also benefited greatly from the smoothing, reducing noise and creating quite visually appealing maps without compromising its data significance.

