# Viscoelastic flow asymmetries in a helical static mixer and their impact on mixing performance.

T.P. John<sup>a,\*</sup>, R.J. Poole<sup>b</sup>, A. Kowalski<sup>c</sup>, C.P. Fonte<sup>a</sup>

<sup>a</sup>Department of Chemical Engineering; The University of Manchester; Oxford Road; Manchester; M13 9PL; UK <sup>b</sup>School of Engineering; The University of Liverpool; Brownlow Street; Liverpool; L69 3GH; UK <sup>c</sup>Unilever R&D; Port Sunlight Laboratory; Quarry Road East; Bebington; Wirral; CH63 3JW; UK

#### Abstract

Helical static mixers are often used during the processing of formulated products with complex rheological properties, such as viscoelasticity. Previous experimental studies have highlighted that increasing the viscoelasticity reduces the mixing performance of helical static mixers in the laminar flow regime. In this study, we use computational fluid dynamics to investigate the flow of a FENE-CR fluid in a helical static mixer. The results show clearly that the reduced mixing performance is caused by flow distribution asymmetries which develop at the mixer element intersections. The numerical results allow us to quantify the degree of asymmetry for the range of conditions studied, which is correlated with the quantified mixing performance for each simulation. The mixing is quantified using a Lagrangian particle tracking technique, and a new mixing index is defined based on the mean nearest distance between the two sets of tracked particles. The results show that the asymmetry parameter does not follow a pitchfork bifurcation, as it typically does for elastic instabilities in symmetrical geometries such as the cross-slot. For low values of the extensibility parameter,  $L^2$ , the flow remained (Eulerian) steady for all Reynolds Re and Weissenburg Wi numbers studied. At fixed Re and Wi, increasing  $L^2$  causes the flow to become transient and greatly increases the magnitude of the asymmetry. The results presented in this study greatly help us to understand the effects that viscoelasticity can cause in mixing processes.

<sup>\*</sup>Corresponding author

Email address: thomas.john@manchester.ac.uk (T.P. John )

## 1. Introduction

Static mixers are passive mixing devices which can be fixed into a pipeline in order to 1 facilitate mixing of two or more components. They are particularly useful for mixing in flows 2 in the laminar regime, where the naturally diffusive nature of inertially-driven turbulence 3 cannot be utilised to promote mixing. Mixing at the molecular scale (micro-mixing) is 4 essential for phenomena such as chemical reaction and is facilitated by molecular diffusion, 5 which acts over significantly longer time-scales compared to the time-scales associated with 6 turbulent dispersion. Thus, the primary function of a laminar mixing device is usually to 7 reduce the scale of segregation of the components to a point where molecular diffusion can 8 act to promote the micro-mixing in feasible time scales. The helical static mixer is one such 9 commonly used static mixer for the mixing of highly viscous materials [24], which consists of 10 a number of helical mixing elements arranged longditudinally in the pipe that split, rotate, 11 stretch and recombine the flow. At the end of each mixer element, the consecutive element 12 is twisted in the opposite direction and rotated 90 degrees around the pipe's longitudinal 13 axis. If two fluids are initially joined with a Y-junction (i.e. for two initial striations), the 14 number of striations should double, and hence the striation thickness should halve, every two 15 mixing elements. Each combination of two mixing elements can be denoted as a flow period, 16 since, across each combination two mixer elements, the flow is spatially periodic (provided 17 it remains laminar) [41]. 18

Many formulated products across various sectors such as personal care, home care, and food, exhibit complex rheological behaviour. In particular, many of these products exhibit viscoelasticity [6, 28]. Despite their frequent use for mixing of such rheologically complex materials, there have been only two previous studies (to the best of the authors' knowledge) regarding the mixing performance of a helical mixer for viscoelastic flows, both of which are experimental. Ramsay et al. [38] used Planar Laser Induced Fluorescence (PLIF) to visualise and quantify the mixing of a fluorescent dye in two different Boger fluids (water-glycerol

solutions with 0.01 and 0.02 wt% of polyacrylamide (PAM)). The PLIF results showed that, 26 for the two Boger fluids, the mixing performance after 6 elements of the mixer is drastically 27 reduced when compared to the mixing of a Newtonian fluid (Glycerol), which the authors 28 attribute to the generation of secondary structures in the flow by viscoelastic effects. The 29 pressure drop across the mixer was found to increase for the two Boger fluids when compared 30 to the Newtonian case. It is mentioned that this could be caused by the polymeric normal 31 stresses present in the Boger fluids. Both the pressure drop and the striation patterns were 32 strongly time-dependent for the Boger fluids but time-independent for the Newtonian fluid. 33 The Reynolds number range in this study was between 10 and 30, and so, although the 34 time-dependence of the flow is only observed for the Boger fluids, it can not be ascertained 35 whether the time-dependence is purely elastic in nature or inerto-elastic. 36

Migliozzi et al. [30] also used PLIF to study mixing of viscoelastic materials in a helical 37 static mixer. They used a Boger fluid (PAM dissolved in pure glycerol) and also two shear-38 thinning viscoelastic fluids (Xanthan Gum in water/glycerol solutions). They also found that 39 the striation patterns produced by the viscoelastic flows were different to those produced by 40 the Newtonian flows. They observe that increasing the viscoelasticity of the flow reduces the 41 number of striations after two mixing elements (and thus reduces the mixing performance). 42 and causes anomalous shapes to appear in the striation patterns. For moderate viscoelas-43 ticity, the striation patterns remain symmetric between the two halves of the cross-section. 44 This symmetry is lost for higher degrees of viscoelasticity however. Time-dependence of the 45 striation patterns was observed for flows beyond a critical value of the Deborah number, 46  $De = \lambda/t$ , where  $\lambda$  is the viscoelastic relaxation time and t is a characteristic time scale of 47 the flow. For the Boger fluids, the onset of transient fluctuations in the mixing patterns 48 occurred at approximately De > 3. This critical De was substantially lower for the Boger 49 fluid than it was for the shear-thinning viscoelastic fluids, indicating that the Boger fluid 50 is more prone to instabilities. It is explained by the authors that the PAM molecules in 51 the Boger fluid exhibit higher extensibility than the Xanthan Gum molecules. Therefore, in 52

flows with extension-dominated regions, the extensional stresses and viscosity grow larger 53 for the Boger fluid than they do for the Xanthan Gum. This is reported to be the cause of 54 the stronger time-dependence and lower critical De observed for the Boger fluids than for 55 the shear-thinning fluids. For the shear-thinning fluids investigated, the striation patterns 56 were still substantially different from the Newtonian ones, even though the time-dependence 57 of the mixing patterns was much weaker or even negligible in some cases, indicating that the 58 viscoelasticity can cause both steady and time-dependent changes in the flow and mixing 59 performance of the helical mixer. In both of these previous experimental studies, only the 60 mixing performance, quantified with the Coefficient of Variance (CoV), and pressure drop is 61 explicitly measured. Changes brought about to the flow pattern within the mixer are only 62 inferred from the changes in the striation patterns or pressure drop. 63

In the last few decades, there has been much research in the field of viscoelastic instabilities [13, 18, 42] and, in particular, flow asymmetries [15, 20, 21, 37, 39, 40, 43]. One of the first geometries to be studied in detail was the cross-slot geometry, now recognised as a bench-mark geometry for viscoelastic asymmetries [12]. In the cross-slot geometry, shown in Figure 1, the viscoelasticity causes a steady-state symmetry-breaking instability, which manifests as a super-critical pitchfork bifurcation [37] given by

$$\frac{\mathrm{d}\Delta}{\mathrm{d}t} = 0 = \Delta^3 - A(De - De_{\mathrm{crit}}), \tag{1}$$

where  $\Delta$  is the parameter quantifying the degree of asymmetry and A is a constant to 70 be determined empirically. Below the critical Deborah number  $De_{crit}$ , the flow bifurcates 71 symmetrically ( $\Delta = 0$ ) at the stagnation point in the center of the cross-slot. But then 72 above  $De_{\rm crit}$ , although reaching steady-state, the flow becomes asymmetric and the growth 73 of  $\Delta$  with increasing De follows a square-root trend. In bifurcation theory, in the case that 74 a bias exists due to, for example, a slight asymmetry or imperfection in the geometry, an 75 imperfection parameter h can be added to the right hand side of Equation (1). In this case, 76  $\Delta \neq 0$  for  $De < De_{crit}$ , and there is a gradual growth of  $|\Delta|$  as  $De \rightarrow De_{crit}$ , rather than an 77

<sup>78</sup> instantaneous change in  $\Delta$  at  $De_{crit}$ . This is highlighted in Figure 2. In the cross-slot, for <sup>79</sup> large enough De, the steady-state asymmetry transitions into a fully transient chaotic state <sup>80</sup> [10], which is often referred to as elastic-turbulence [19, 27].

In the earlier investigations of the steady viscoelastic flow asymmetries in the cross-slot 81 geometry, it was suggested that the driving mechanism for the asymmetry was related to 82 the extensional flow and stresses at the stagnation point. It is observed in the regular 83 cross-slot geometry that the onset of the asymmetry causes the flow near the stagnation 84 point to change from being extensionally dominated to being shear dominated [2]. Also, the 85 asymmetry causes a parameter named the "Couette correction" to drop with increasing De. 86 The Couette correction is the pressure drop across the geometry after taking into account the 87 pressure drop required for the viscoelastic channel flow in the absence of the cross-slot. As 88 such, it represents, in a way, the energy requirement to drive the flow in the cross-slot. The 89 drop in the value of the Couette correction with increasing De above the critical De indicates 90 that the underlying driving mechanism for the asymmetry might be related to a drop in the 91 energy requirement for the flow. However, the results from a more recent investigation by 92 Davoodi et al. [14] suggest that asymmetry in the cross-slot is instead related simply to a 93 classic "curved-pathlines" viscoelastic instability [33]. In the study of Davoodi et al. [14], a 94 cylinder is added to the center of the cross-slot geometry, which removes the free stagnation 95 point (and the associated strong extensional flow and stresses), however the viscoelastic 96 asymmetry still occurs beyond a critical degree of viscoelasticity, indicating the asymmetry 97 arises due to the curvature of the pathlines and high deformation rates near the channel 98 corners. 99

<sup>100</sup> Viscoelastic asymmetries have also been observed in a confined cylinder geometry, in <sup>101</sup> which flow in a channel bifurcates around an obstructing cylinder. It has been found that <sup>102</sup> increasing the Weissenberg number,  $Wi = \lambda \dot{\gamma}$ , where  $\dot{\gamma}$  is a characteristic rate of strain, again <sup>103</sup> led to a symmetry breaking instability, where the flow passed preferentially around one side <sup>104</sup> of the cylinder in the channel [20, 21, 43]. This was observed with both experimental and



Figure 1: Contours of |u| for the cross-slot geometry before (a) and after (b) viscoelastic asymmetry is observed. Fluid pathlines are superimposed as white solid lines.



Figure 2: Examples of perfect (red) and imperfect (blue and green) pitchfork bifurcations. Solid lines show stables solution branches and dashed lines show un-stable solution branches

numerical methods. With regards to the choice of representing the degree of viscoelasticity 105 with either De or Wi, De can be thought of as being related to the unsteadiness of the flow 106 in a Lagrangian sense, whilst Wi represents the ratio of elastic and viscous forces (taking 107  $[\tau_{11} - \tau_{22}]/\tau_{12}$  for the upper-convected Maxwell model under steady simple shear flow yields 108  $2\lambda\dot{\gamma}$ ). For flows which are Eulerian and Lagrangian steady, it is the case that De = 0, and 109 for geometries where the same length scale controls both the characteristic strain rate and 110 time-scale, it is the case that De = Wi and thus the choice for representing the degree of 111 viscoelasticity is arbitrary. For Lagrangian unsteady flows with more than one important 112 length scale, Wi and De are usually related simply via a geometric factor [36]. 113

The confined cylinder geometry is not so dissimilar from the helical static mixer, in the 114 sense that the flow in the helical mixer is also confined and bifurcates around the mixing 115 elements at each element intersection point. The primary differences are that the helical 116 mixer geometry twists the flow as it moves in the axial direction (hence the base flow is 117 not symmetric), and that the edges of the mixing elements (at least those in this investi-118 gation) are square rather than circular. Whist the majority of investigations of viscoelastic 119 flow asymmetries have employed symmetrical geometries, a recent numerical investigation 120 by Kumar and Ardekani [26] employed an asymmetrical geometry. The geometry consisted 121 of a channel with two confined cylinders, longitudinally arranged, where the front cylinder 122 was fixed in the center of the channel width and the spanwise position of the rear cylinder 123 was varied. When the rear cylinder is positioned centrally, the behaviour of the viscoelas-124 tic flow asymmetry follows closely that observed for other symmetrical geometries; the flow 125 distribution around the cylinders is practically symmetrical until a critical Wi is reached, 126 beyond which a sudden and sharp increase of the flow asymmetry is observed as Wi is in-127 creased. However, for the case where the rear cylinder is positioned off-center, the behaviour 128 of the flow asymmetry is much different; even for low Wi there is a clear flow asymmetry 129 around both cylinders and increasing Wi causes a more gradual increase in the degree of the 130 asymmetry, rather than a sharp sudden increase. This would seem similar to the addition 131

of an imperfection parameter h in the pitchfork bifurcation (see Figure 2), however this was not explored further in their study.

In this study, Computational Fluid Dynamics (CFD) is used, for the first time, to study 134 viscoelastic fluid flows in the helical static mixer geometry, and the effect of viscoelasticity on 135 the resulting mixing performance. In particular, we aim to understand and demonstrate how 136 viscoelasticity can affect the performance of industrial process equipment. Gaining a better 137 understanding of how viscoelasticity impacts the mixing quality in static mixers will allow 138 for better design of equipment, processes, and formulated products. Previously, suggestions 139 regarding the flow pattern of viscoelastic flows in the helical static mixer have been inferred 140 from the mixing striation patterns observed with PLIF. With CFD, we can study details 141 of the flow pattern inside the mixer and directly relate this to the change in the mixing 142 performance. 143

#### <sup>144</sup> 2. Materials and methods

Here, we present the numerical methodology for the simulations, followed by an expla nation of the methods used to quantify the mixing performance from the numerical results.

#### 147 2.1. Governing equations

The continuity and momentum equations for in-compressible flow in the absence of external body forces are given respectively as

$$\nabla \cdot \boldsymbol{u} = 0, \tag{2}$$

$$\rho \left( \frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u} \right) = \nabla \cdot \boldsymbol{\sigma},$$
(3)

where  $\sigma$  is the total stress tensor given as  $\sigma = -pI + \tau$ , p is the pressure and  $\tau$  is the extra-stress tensor. We employ the FENE-CR (Finitely Extensibile Non-linear Elastic with <sup>152</sup> Chilcott and Rallison modification) model, first presented by Chilcott and Rallison [11], is <sup>153</sup> used, which is given as

$$\boldsymbol{\tau} = \boldsymbol{\tau}_p + 2\eta_s \boldsymbol{D}, \tag{4}$$

$$\boldsymbol{\tau}_{p} + \lambda \left(\frac{\boldsymbol{\tau}_{p}}{f}\right) = 2\eta_{p}\boldsymbol{D}, \qquad (5)$$

where  $\boldsymbol{\tau}_p$  is the polymeric stress and  $\boldsymbol{D}$  is the rate-of-strain tensor given by  $\boldsymbol{D} = 1/2 (\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^{\mathrm{T}})$ .  $\eta_s$  and  $\eta_p$  are the solvent and polymeric viscosities, respectively. The symbol  $(\stackrel{\nabla}{})$  denotes the upper-convected time derivative, which, for a tensor  $\boldsymbol{\psi}$ , is given as

$$\vec{\psi} = \frac{\mathrm{D}\psi}{\mathrm{D}t} - \psi \cdot \nabla u - \nabla u^{\mathrm{T}} \cdot \psi, \qquad (6)$$

and represents the time rate of change written in a coordinate system which translates, rotates, and deforms with the material. It should be noted that the upper convected derivative in Equation (5) is acting on the whole term in the brackets, and not just on  $\tau_p$  as is the case in many other viscoelastic constitutive models. f is given by

$$f = \frac{L^2 + (\lambda/\eta_p) \operatorname{Tr}(\boldsymbol{\tau}_p)}{L^2 - 3},\tag{7}$$

where  $L^2$  is the extensibility parameter. We note that  $\lim_{L^2 \to \infty} f = 1$ , and in this limiting case the FENE-CR model reduces to the widely-used Oldroyd-B model.

In terms of material functions, the FENE-CR model has a constant shear viscosity in steady-shear, and so it can potentially be thought of as a Boger fluid [7] model. This is the primary feature of the FENE-CR model, which was modified empirically from the original FENE-P for this reason. The first normal stress difference,  $N_1 = \sigma_{11} - \sigma_{22}$  (again, in steadyshear), grows quadratically with shear rate in the linear viscoelastic regime, and there is shear thinning of  $N_1$  in the non-linear regime. In steady-state extensional flow, there is some

thickening of the extensional viscosity with increasing strain rates, after which a plateau 169 is reached. The value of the extensional viscosity at the plateau is proportional to  $L^2$ . 170 For the Oldroyd-B model  $(L^2 \rightarrow \infty)$ , there is no limiting of the extensional thickening and a 171 singularity occurs. This is a well known short-coming of the Oldroyd-B model [42]. It should 172 be noted here that since f is contained within the upper-convected derivative, the behaviour 173 of the FENE-CR model in Eulerian or Lagrangian unsteady flows will differ from that implied 174 from its steady material functions. This will be the case in the helical mixer geometry where 175 even an Eulerian steady flow is un-steady in a Lagrangian sense. Moreover, the flow in the 176 mixer section is complex (not pure shear, pure extension, or pure rotation), and it has been 177 recently highlighted [44] that even the simplest viscoelastic model, the Oldroyd-B model, 178 exhibits rheological behaviour which, in such flows, is more complex that that inferred from 179 its material functions. We point this out just to highlight that the observed behaviour of 180 the FENE-CR model in ideal steady flows can not necessarily be extrapolated to complex 181 flows such as those in the static mixers. 182

183 We now introduce the dimensionless variables

$$t^* = t\frac{\mathcal{U}}{\mathcal{L}}, \quad \boldsymbol{u}^* = \boldsymbol{u}\frac{1}{\mathcal{U}}, \quad \boldsymbol{\tau}^* = \boldsymbol{\tau}\frac{\mathcal{L}}{\mathcal{U}(\eta_s + \eta_p)}, \quad p^* = p\frac{\mathcal{L}}{\mathcal{U}(\eta_s + \eta_p)}, \quad \nabla^* = \nabla\mathcal{L}, \quad (8)$$

<sup>184</sup> and the following dimensionless groups

$$\beta = \frac{\eta_s}{\eta_s + \eta_p}, \quad Re = \frac{\rho \mathcal{UL}}{(\eta_s + \eta_p)}, \quad Wi = \frac{\lambda \mathcal{U}}{\mathcal{L}}, \tag{9}$$

where  $\mathcal{U}$  and  $\mathcal{L}$  are characteristic velocity and length scales, respectively. Substituting the dimensionless variables into the momentum and constitutive equation, and dropping the asterisks for brevity, gives

$$Re\left(\frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u}\right) = -\nabla p + \beta \nabla^2 \boldsymbol{u} + \nabla \cdot \boldsymbol{\tau}_p, \qquad (10)$$

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$$\boldsymbol{\tau}_{p} + Wi\left(\frac{\boldsymbol{\tau}_{p}}{f}\right) = (1 - \beta)(\boldsymbol{\nabla}\boldsymbol{u} + \boldsymbol{\nabla}\boldsymbol{u}^{\mathrm{T}}) \quad \text{where} \quad f = \frac{L^{2} + \left(\frac{Wi}{(1 - \beta)}\right) \mathrm{Tr}(\boldsymbol{\tau}_{p})}{L^{2} - 3}.$$
(11)

For the geometry used in this investigation, the helical static mixer, we define  $\mathcal{U}$  and  $\mathcal{L}$ 188 as the inlet velocity  $u_{in}$  and pipe diameter D, respectively. There are three dimensionless 189 parameters which, for a given geometry and value of  $L^2$ , can be varied, which are Re, Wi, 190 and  $\beta$ . In this investigation, we use a constant value for  $\beta$  of 1/817 and we systematically 191 vary Re and Wi for  $L^2 = 50$ , which is a typical value value of  $L^2$  used for simulations with 192 FENE constitutive models [31, 39]. The value of  $\beta$  chosen is close to that for typical fluids 193 processed with helical static mixers in the laminar flow regime. We note here that the 194 relatively low value of  $\beta$  is not representative of that for a "true" Boger fluid; the primary 195 reason for the use of the FENE-CR model in this study is to remove the effects of shear-196 thinning and second normal stress differences etc, and not to explicitly attempt to model a 197 Boger fluid. For  $L^2 = 50$ , Re was varied between 0.040 and 5.965, and Wi was varied between 198 0.010 and 1.500. Simulations were also run with increasing values of  $L^2$  (up to  $L^2 = 5000$ ) 199 for Re = 0.487 and Wi = 0.429 in order to assess the effect of extensibility on the flow. 200

#### 201 2.2. CFD simulations

All simulations were run using version 5.0 of RheoTool [34, 35], an open-source tool box 202 based on OpenFOAM<sup>®</sup> [32]. OpenFOAM<sup>®</sup> uses the finite volume method to discretise the 203 governing equations. The University of Manchester's Computational Shared Facility was 204 used to run the simulations. Simulations were run with between 80 and 200 processors and 205 took between a few hours to several days depending on the values of Re and Wi. In order to 206 avoid the infamous high Weissenberg number problem, in which the polynomial interpolation 207 fails to capture the exponential growth in stress, the log-conformation approach proposed by 208 Fattal and Kupferman [17] is employed in this study. In this approach, the constitutive model 209 is reformulated and the matrix logarithm of the conformation (or configuration) tensor  $\Theta$  is 210 solved for, rather than  $\boldsymbol{\tau}_p$ . Spacial gradients of  $\boldsymbol{\Theta}$  are better approximated by polynomials, 211

and  $\Theta$  is also always positive-definite [1], which helps to stabilise the simulations. In the (dimensionless) FENE-CR model,  $\Theta$  is related to  $\tau_p$  by  $\tau_p = [(1 - \beta)f_{\Theta}/Wi](e^{\Theta} - \mathbf{I})$  where  $f_{\Theta} = L^2/[L^2 - \operatorname{tr}(e^{\Theta})]$ . The FENE-CR model in log-conformation form can be found in the RheoTool user guide [34].

Gradient terms were discretised using the Gauss Linear scheme, whilst the convective 216 terms in both the momentum equation and FENE-CR model were discretised using the 217 high-order CUBISTA scheme proposed by Alves et al. [4]. Highly accurate interpolation 218 schemes are often needed for the disretisation of viscoelastic models, even for creeping flows, 219 due to the absence of a diffusion-like term. Lower order schemes such as first-order upwind 220 can introduce excessive numerical diffusion [5]. The Gauss Linear scheme with corrected 221 surface normal gradient scheme was used for discretisation of Laplacian terms. The Euler 222 scheme was used for time discretisation. 223

At the inlet of the pipe, a uniform and constant value for the fluid velocity and zero 224 normal gradient of the pressure were set as boundary conditions. All components of  $\Theta$ 225 were also specified as 0 at the inlet. At the outlet, the pressure was fixed at 0, and the 226 pressureInletOutletVelocity boundary condition available in OpenFOAM<sup>®</sup> was used for the 227 velocity, which is essentially a zero-gradient condition for purely outgoing flow. Although 228 the flow at the outlet was always out-going, it was found that the *pressureInletOutletVelocity* 229 condition helped to prevent convergence issues in some simulations. Zero normal gradient 230 was also used at the outlet for  $\Theta$ . At all solid walls, the no-slip condition was used for 231 velocity, the zero-gradient condition was used for  $\Theta$ , and the polymeric stress was linearly 232 extrapolated. The fluid was initialised with a value of 0 for all variables (considering  ${m au}_p$ 233 rather than  $\Theta$ ). 234

To solve the equations, the Geometric agglomerated Algebraic MultiGrid (GAMG) solver was used for pressure with the Gauss Seidel smoother, and the *smoothSolver* was used for both the velocity and  $\Theta$  also with the Gauss Seidel solver. The tolerance and relative tolerance for both solvers respectively were  $10^{-12}$  and  $10^{-3}$ . All simulations with  $L^2 = 50$ 

were run until steady-state had been reached. The two simulations with  $L^2 = 500$  and 239  $L^2 = 5000$  were unsteady and so were run for approximately  $13\lambda$ , which was long enough for 240 the unstable system to become fully-developed (i.e. by this point, fluctuations were clearly 241 fluctuating around a steady mean). The time step was auto-adjusted using the maximum 242 value of the Courant number. The Courant number is defined as  $Co = \Delta_t \mathcal{T}$  where  $\Delta_t$  is 243 the time-step and  $\mathcal{T}$  is a time scale based on the local cell flow scales. The maximum Co 244 was varied between 0.4 for lower Wi cases and 0.05 for higher Wi cases. The change in the 245 time-step should not affect any results for  $L^2 = 50$  since they all reached steady-state. For 246 the  $L^2 = 5000$  simulation, which was unsteady, a time-step sensitivity test confirmed that 247 the results were insensitive to the time-step at the maximum Co number we used. 248

#### 249 2.3. Geometry and mesh

The mixer investigated in this study consists of 8 mixing elements (or 4 flow elements/periods). The diameter and length of the blades were fixed at 25.5 mm and 34.5 mm respectively (aspect ratio of roughly 1.35) and the thickness of the blades was 1.5 mm. These dimensions were used in the investigation conducted by Migliozzi et al. [30]. The entire pipe section simulated was 1 m in length and 25.5 mm in diameter. The center of the mixer section was located in the center of the pipe. Thus, there was always sufficient length between the inlet and the first mixer element for the flow to become fully-developed [16].

The mesh for the simulations, shown in Figure 3, was generated using ANSYS Meshing. To mesh the elements of the mixer, the sweep method was used to ensure that cells were hexahedral. The final mesh used for the simulations consisted of 4.93 million hexahedral cells. The mesh was chosen after performing a mesh independence study, the results of which can be found in the Supplementary Material. The mesh generated by ANSYS Meshing was converted for use in OpenFOAM<sup>®</sup> with the *fluentMeshToFoam* command.



(c)

Figure 3: Mesh used for the simulations. (a) 2D longitudinal slice in the mixer section. (b) Circular slice in the pipe section. (c) 3D view of the mesh at the beginning of the mixer section.

#### 263 2.4. Quantification of mixing performance

In order to quantify the mixing performance, passive fluid elements were introduced to 264 the flow at the beginning of the mixer geometry and their pathlines were analysed throughout 265 the mixer. Starting at the entrance of the mixer geometry, roughly 50,000 elements were 266 tracked using MATLAB<sup>®</sup> 2022a v9.12 [29] and Paraview v5.10 [3]. The elements are split 267 into two groups, denoted by A and B, of roughly 25,000 elements each. The orientation of 268 the two groups of elements was rotated at 90 degrees with respect to the first mixer element 269 so that the elements did not bypass the first mixer element. This is shown in Figure 4. 270 Circular slices (normal to the longitudinal direction) of the pathlines were taken at each 271 element intersection point, and the mixing was then quantified. 272



Figure 4: Initial positions of the tracked passive fluid elements at  $n_e = 0$ . The white horizontal rectangle shows the solid wall of the start of the first mixer element.

There are a number of methods for quantifying mixing using discrete particles or elements. For example, one could use the CoV or the Shannon Entropy, both of which have been employed previously in similar CFD mixing studies [8, 9, 22]. However, during the processing in ParaView, a number of pathlines were terminated, leading to some small blank patches in the mixing patterns. In order to avoid this affecting the results, we introduce a new method for the quantification of the mixing on 2D y - z slices throughout the mixer geometry, implemented via MATLAB. We define the mixing index,  $I_m$ , as

$$I_m = \frac{1}{N_A} \sum_{i=1}^{N_A} \min[\mathbf{d}(\mathbf{P}_{A,i}, \mathbf{P}_B)], \qquad (12)$$

where  $N_A$  is the number of fluid elements belonging to group A and  $d(\mathbf{P}_{A,i},\mathbf{P}_B)$  is given by

$$\mathbf{d}(\mathbf{P}_{A,i},\mathbf{P}_B) = \sqrt{(\mathbf{y}_B - y_{A,i})^2 + (\mathbf{z}_B - z_{A,i})^2},\tag{13}$$

and represents an array consisting of the magnitudes of all of the 2D vectors in a y-z plane 281 pointing from the position **P** of particle *i* in group A (located in the 2D plane at  $y_{A,i}$  and  $z_{A,i}$ ) 282 to the position of each of the particles in group B. The arrays  $\mathbf{y}_B$  and  $\mathbf{z}_B$  contain respectively 283 the y and z positions of all elements in group B. As such, Equation (12) represents the mean 284 distance from a particle in A to its nearest neighbour particle in B. This should scale closely 285 with the striation thickness, which is commonly used to measure mixing performance in 286 laminar mixing devices such as the helical static mixer [25, 41], and should be less influenced 287 by the blank spaces caused by the termination of some of the pathlines. However, since 288 the mixing here is being quantified with discrete methods, the index of mixing  $I_m$  will not 289 decay to zero as the true mixing index in a continuous sense does, but will decay to a value 290 determined by the total number of particles or pathlines used. Although this should be fairly 291 obvious, it is noted here for clarity that  $\min[\mathbf{d}(\mathbf{P}_{A,i},\mathbf{P}_B)] = \min[\mathbf{d}(\mathbf{P}_{B,i},\mathbf{P}_A)]$  and so the 292 choice of which group of pathlines to index is arbitrary so long as  $N_A \approx N_B$ . Also, when 293 results for the decay of  $I_m$  are presented, they will be normalised by the value of  $I_m$  obtained 294 for the initial seeding of the pathlines,  $I_0$ , so that  $I_m/I_0$  always decays from 1 at the start of 295 the first mixing element. 296

#### <sup>297</sup> 3. Results and discussion

# <sup>298</sup> 3.1. Steady flow asymmetries for $L^2 = 50$

In this sub-section, the main CFD results regarding the flow patterns generated in the 299 mixer will be presented and discussed. The CFD results show that, as the viscoelasticity of 300 the flow is increased, the flow bifurcates asymmetrically at the element intersections. This 301 is shown for Re = 0.04 (i.e. negligible inertia) in Figure 5. For Wi = 0.01, the flow splits 302 around the vertical wall of the mixing element relatively evenly. However, for Wi = 1.5, 303 the flow clearly passes preferentially to one side of the vertical wall of the mixing element. 304 Contours of  $u_x$  approaching  $n_e = 5$  can be seen for Re = 0.04 and Re = 5.96 in Figures 6 305 and 7 respectively. For Re = 0.04 (Figure 6), the flow begins to preferentially flow below the 306 up-coming horizontal mixing element as Wi is increased from 0.01 to 0.122. However, for 307 Wi > 0.122, the flow distribution suddenly changes and the flow preferentially passes above 308 the mixing element. It appears that the maximum value of  $u_x$  increases in both the upper 309 and lower halves of the contour (above and below the up-coming horizontal intersection) as 310 Wi is increased from 0.01 to 0.122, however, although it is not immediately obvious to the 311 eye, the distribution of  $u_x$  in the upper half of the contour becomes slightly narrower and 312 appears to be pushed slightly in an anti-clockwise direction, which matches the direction of 313 the observed flow asymmetry for this range of Wi. For  $Wi \ge 0.429$ , the distribution of  $u_x$  in 314 the lower quadrant seems to be almost "cut" diagonally (from bottom-left to upper-right or 315 vice versa), where the flow above this diagonal line is forced above the up-coming horizontal 316 element. For Re = 5.96, this "cutting" effect for  $Wi \ge 0.429$  seems more pronounced and 317 there appear to be sharp spatial gradients of  $u_x$ . Also, for Re = 5.96, the flow seems to pass 318 preferentially above the horizontal mixing element as Wi is increased from 0.01 to 0.122, 319 which is in contrast to the behaviour observed for Re = 0.04. 320



Figure 5: Example of fluid pathlines around mixer elements 4 and 5 for (a) Wi = 0.01 and (b) Wi = 1.5. Re = 0.04 and  $L^2 = 50$  for both cases. Pathlines are coloured by |u|. Flow direction is from left to right. Pathlines are integrated (forward and backwards) from a line at the intersection of the two mixing elements.



(a) Wi = 0.010 (b) Wi = 0.035 (c) Wi = 0.122 (d) Wi = 0.429 (e) Wi = 1.500

Figure 6: Contours of  $u_x$  for various Wi. Re = 0.040 and  $L^2 = 50$ . Slices are positioned at 5 mm before  $n_e = 5$ . The twisting direction of the element is clockwise. Fluid is flowing in the positive x direction (into the page).



(a) Wi = 0.010 (b) Wi = 0.035 (c) Wi = 0.122 (d) Wi = 0.429 (e) Wi = 1.500

Figure 7: Contours of  $u_x$  for various Wi. Re = 5.95 and  $L^2 = 50$ . Slices are positioned at 5 mm before  $n_e = 5$ . The twisting direction of the element is clockwise. Fluid is flowing in the positive x direction (into the page).

In order to quantify the degree of asymmetry, the following parameter is defined

$$\Delta \equiv \frac{\max(u_x)_{s_i} - \max(u_x)_{s_j}}{\max(u_x)_{s_i} + \max(u_x)_{s_i}},\tag{14}$$

where  $\max(u_x)_{s_i}$  and  $\max(u_x)_{s_j}$  are the maximum longitudinal velocities on the clipped surfaces  $s_i$  and  $s_j$  respectively. As such,  $-1 < \Delta < 1$ . Considering the clipped surfaces shown in Figure 8, a number of definitions for  $\Delta$  can be introduced using adjacent surfaces. We introduce two of these and denote them as  $\Delta_1$  and  $\Delta_2$ , which are given as

$$\Delta_1 \equiv \frac{\max(u_x)_{s_1} - \max(u_x)_{s_2}}{\max(u_x)_{s_1} + \max(u_x)_{s_2}}, \quad \Delta_2 \equiv \frac{\max(u_x)_{s_3} - \max(u_x)_{s_4}}{\max(u_x)_{s_3} + \max(u_x)_{s_4}}.$$
(15)

For all simulations using  $L^2 = 50$ , we were able to avoid having to re-define  $\Delta$  for odd and even mixer elements, since it was apparent that  $\max(u_x)_{s_1} \approx \max(u_x)_{s_3}$  and  $\max(u_x)_{s_2} \approx$  $\max(u_x)_{s_4}$ . We checked  $|\Delta_1 - \Delta_2|$  at all mixing element intersections for all simulations with  $L^2 = 50$ . The maximum value of  $|\Delta_1 - \Delta_2|$  was found to be  $1.1 \times 10^{-3}$ , indicating that it is

fair to assume  $\Delta_1 = \Delta_2$ . This anti-symmetry across the mixer element was also observed 330 experimentally by Migliozzi et al. [30]. From here-on-in we will use  $\Delta \equiv \Delta_1$  and we will not use 331 the subscript when presenting results for  $\Delta$ . With this choice of the definition of  $\Delta$ , positive 332  $\Delta$  means the flow is predominantly passing through the quadrant positioned in the twisting 333 direction of the previous element, and negative  $\Delta$  means the flow is predominantly passing 334 through the quadrant positioned against the twisting direction of the previous element. For 335  $\Delta = 0$ , the bifurcation of the flow at the element intersections should be approximately 336 symmetrical. 337



Figure 8: Part of the helical static mixer geometry with the clipped surfaces  $s_1$ ,  $s_2$ ,  $s_3$ , and  $s_4$  which are used to define  $\Delta$ . Flow direction is in the x-direction.

Figure 9 shows  $\Delta$  as a function of Wi for each element number for Re = 0.040, 0.487, 5.965. 338 In Figures 9a and 9b,  $\Delta$  starts very close to zero for Wi = 0.010 (i.e. Newtonian) before 339 becoming negative and increasing in magnitude up to Wi = 0.122. Beyond Wi = 0.122, the 340 magnitude of  $\Delta$  increases further, however the sign of  $\Delta$  abruptly changes from negative 341 to positive, as is indicated by the velocity contours in Figure 6.  $\Delta$  is virtually constant for 342 all elements for the lower values of Wi, but there is some change of  $\Delta$  with the element 343 number, specifically for the first few elements, for higher values of Wi. There appears to be 344 no notable difference between the solutions for Re = 0.04 and Re = 0.487, indicating that the 345 solution is practically independent of *Re* in this range. 346



Figure 9:  $\Delta$  versus Wi for (a) Re = 0.040, (b) Re = 0.487, and (c) Re = 5.965. Red dashed line shows the fitting of a pitchfork-type bifurcation for  $n_e = 0.4$ 



Figure 10:  $\Delta$  at  $n_e = 4$  vs Wi for the extra simulations ran to probe more closely the behaviour of  $\Delta$ . (a) shows results between Wi = 0.01 and Wi = 0.035 (blue symbols) where the blue line represents a fitted imperfect pitchfork bifurcation. (b) shows the fitted imperfect pitchfork over the entire range of Wi, and extra simulations between Wi = 0.122 and Wi = 0.429 in green.



Figure 11: Results for simulation employing inlet velocity ramp. Red symbols show the steady-state simulations for Re = 0.04. Black symbols show the transient solution between Wi = 0.122 at t = 0 and Wi = 0.429 at  $t_f = 10\lambda$ . Filled symbols exhibit a downwards trend whilst hollow symbols follow the steady-state solution in an upwards trend.

There are multiple possible explanations for the behaviour observed in Figures 9a and 9b. 347 One is that a viscoelastic instability occurs between Wi = 0.01 and Wi = 0.035, and both the 348 downwards and upwards trends in  $\Delta$  represent the two respective stable solution branches of a 349 supercritical pitchfork bifurcation (either perfect or imperfect). To highlight this possibility, 350 we have fitted the solution for a perfect pitchfork bifurcation  $(\Delta = \pm \sqrt{A_1(Wi - Wi_{crit})})$ 351 to the results in Figure 9a, which is shown by the red dashed line. We fitted the curve 352 to the value of  $\Delta$  at  $n_e = 4$  and found  $A_1 = 0.031$  and  $Wi_{crit} = 0.029$ . Since there is not 353 enough data in Figure 9a to validate the onset of the square-root trend at the fitted value 354 of  $Wi_{crit}$ , we ran 9 extra simulations spaced linearly between Wi = 0.010 and Wi = 0.035. 355 The results for these simulations are displayed in Figure 10a. It is evident then that the 356 asymmetry parameter does not exhibit a perfect pitchfork bifurcation (the red solid line) 357 in the range of Wi investigated. This is highlighted by the gradual increase in  $|\Delta|$  between 358 Wi = 0.010 and Wi = 0.035 and also by the fact that  $\Delta$  appears to asymptote to a small but 359 finite value as  $Wi \rightarrow 0$ . As mentioned in Section 1, this gradual increase in the asymmetry 360 parameter was also observed by Kumar and Ardekani [26] for flow around an asymmetric 361 arrangement of two confined cylinders. Due to the asymmetric nature of the helical static 362 mixer geometry, it would seem plausible that the flow asymmetry exhibits, instead, an 363 imperfect pitchfork bifurcation. We have fitted the data shown in Figure 10a to an imperfect 364 pitchfork bifurcation, shown by the blue solid line. The data appears to fit well between 365 Wi = 0.01 and Wi = 0.035, however, in Figure 10b we show that  $\Delta$  does not fit to the same 366 imperfect pitchfork bifurcation for the entire range of Wi investigated. We also ran 3 extra 367 simulations in between Wi = 0.122 and Wi = 0.429 to probe the apparent sharp transition 368 from negative to positive  $\Delta$ . These are shown by the green symbols in Figure 10b. If the 369 asymmetry is indeed characterised by a pitchfork bifurcation, this transition from negative 370 to positive  $\Delta$  would likely just represent the CFD solver being able to access only the upper 371 stable solution for Wi > 0.122. In Figure 10b, however, it is observed that the transition 372 between negative and positive  $\Delta$  is gradual, indicating that the flow is likely not characterised 373

<sup>374</sup> by either the perfect or imperfect pitchfork bifurcations.

Another potential explanation for the observed trend in  $\Delta$  as Wi is increased is that 375 the initial downwards trend in  $\Delta$  for Wi < 0.122 represents some phenomenon related to 376 viscoelasticity and streamline curvature; potentially a hoop stress or a similar elastic stress 377 which can drive a change in the flow direction. In this case, the abrupt change in the sign of 378  $\Delta$ , and hence direction of the asymmetry, between Wi = 0.122 and Wi = 0.429 might then 379 represent the onset of an instability. To investigate this, we ran a simulation for Re = 0.04380 where the initial condition was the steady-state solution for Wi = 0.122, and the inlet velocity 381 was ramped such that Wi = 0.122 at time t = 0 and Wi = 0.429 at time  $t = t_f$ . We chose 382  $t_f = 10\lambda$  so that the system remains close to steady-state at each point in time. It should 383 be noted that, since the inlet velocity is increased, Re also increases from Re = 0.04 to 384 Re = 0.14. However, as has been demonstrated, the solution is essentially independent of 385 *Re* within this range. The results for this simulation are presented in Figure 11. At the 386 start of the velocity ramp, the asymmetry continues on a downwards path despite the fact 387 the steady-state solution starts to exhibit an upwards path. This indicates that beyond 388 approximately Wi = 0.122, the system might still exhibit two stable solution branches, as 389 a pitchfork-type bifurcation does. At approximately Wi = 0.2, the transient solution then 390 begins to follow the trend exhibited by the steady-state solution. Potentially, a second stable 391 steady-state solution path (downwards) exists beyond Wi = 0.122 but is just not accessible 392 by the CFD solver. 393

For Re = 5.96, the sign of  $\Delta$  is positive for all Wi studied, and for  $Wi \rightarrow 0$ ,  $|\Delta|$  tends to a larger value than it does for the lower Re cases. This shows that a degree of asymmetry is caused purely by inertia. Hobbs and Muzzio [23] simulated laminar Newtonian flows in a helical static mixer with CFD and also found that increasing Re led to an asymmetry at the element intersections. They also investigated the effect of the asymmetry on the mixing by using Lagrangian particle tracking to plot the Poincaré sections. They found that for intermediate Re (i.e. between creeping flow and transitional flow) the mixing was no longer

globally chaotic, due to the flow asymmetries, as it was for creeping flow. In Figure 9c, 401 for Re = 5.96, the maximum value of  $\Delta$  reached as Wi is increased is significantly lower 402 than for Re = 0.04 and 0.487, indicating that increasing inertia dampens these effects of 403 viscoelasticity at high Wi. It has been shown for the cross-slot geometry that increasing Re404 dampens the viscoelastic instability and asymmetry [37]. There is also more variation in  $\Delta$ 405 with the element number for Re = 5.96 than for Re = 0.04 and Re = 0.487, particularly in 406 the first 4 elements of the mixer. Figure 12 shows the contour of  $\Delta$  for  $L^2 = 50$  in the range 407 of Re and Wi studied.  $\Delta$  becomes most negative at moderate Wi (approximately 0.1) when 408  $Re \rightarrow 0$ , indicating that inertia acts to inhibit this effect of viscoelasticity in this region of 409 Re - Wi space. For  $Re \approx 2$ ,  $\Delta$  is no longer negative for the lowest values of Wi, however, 410 the magnitude of  $\Delta$  increases to a larger value (and more rapidly) with increasing Wi than 411 it does for the largest values of *Re*. This indicates a complicated transition between the 412 dominating effects of elasticity and inertia. 413



Figure 12: Contour of  $\Delta$  for the range of Re and Wi studied. Note the data for the extra simulations shown in Figures 10 and 11 is not used to create this contour.

#### 414 3.2. Effect of viscoelasticity on mixing performance

In this sub-section, we investigate the impact of the previously-discussed flow distribution asymmetries on the the mixing performance of the mixer for the range of Re and Wi investigated with  $L^2 = 50$ . As mentioned, the mixing performance is calculated using pathlines generated from the numerical flow-field results.

Figure 13 shows the mixing patterns predicted by the CFD simulations for Re = 0.04 at 419 the ends of mixer elements 2, 4, and 6 for various Wi. For Wi = 0.010 (essentially creeping 420 Newtonian conditions), the mixing patterns are very similar to the typical patterns generated 421 by the helical static mixer for laminar flows reported in the literature [23, 41]. After 6 mixing 422 elements, the striations are small and the distribution of striation thickness is narrow. As 423 the viscoelasticity of the flow is increased, however, the mixing patterns change significantly. 424 For Wi = 1.5, the pattern at the end of element 2 is changed slightly in shape from the lower 425 Wi patterns, although the mixing quality might not seem so different. In the upper-left 426 quadrant of plot, the striation of red particles furthest on the left appears to be squashed 427 downwards; much less of this striation exists above the horizontal wall of the up-coming 428 mixer element than it does for  $Wi \leq 0.122$ . Similarly, in the lower right quadrant, there is 429 only one striation of red particles for Wi = 1.5, whereas there are two for  $Wi \leq 0.122$ . The 430 direction of the movement of the striations as Wi is increased matches the direction of the 431 flow asymmetry (note the co-ordinate system here is different to that in Figures 6 and 7). 432 For the end of elements 4 and 6, there is a significant reduction in the mixing performance for 433 Wi = 1.5. There is a significantly broader distribution of striation shape and size, with some 434 very large striations (relatively speaking) remaining. We cannot infer that the direction of 435 movement of striations is correlated with the asymmetry direction here since the striation 436 patterns are much more complex. However, our results show that the significant change in 437 the qualitative mixing performance is caused by the flow asymmetries present at the element 438 intersection points discussed previously. 439



Figure 13: Mixing patterns predicted by CFD simulations at Re = 0.04 for various values of Wi. Patterns shown at the end of elements 2 (a-c), 4 (d-f) and 6 (g-i).

Figure 14 shows  $I_m/I_0$  versus  $n_e$  for the range of Wi and Re investigated with  $L^2 = 50$ . For both Re = 0.04 and Re = 0.487,  $I_m/I_0$  decays practically as  $c^{-x}$  for the first three

mixing elements when  $Wi \leq 0.122$ , which is natural since the mixing in the helical static 442 mixer involves splitting and recombination of the flow, meaning the mixing index (or striation 443 thickness etc) should ideally decay exponentially. As mentioned previously, the decay of 444  $I_m/I_0$  slows for higher  $n_e$  even for the (practically) Newtonian case (Wi = 0.010), which is 445 caused by the fact that the mixing is approaching the limit of that which can be quantified 446 with the number of pathlines used. This does not impact the analysis or findings of this 447 investigation since the effect of viscoelasticity on  $I_m/I_0$  is easily distinguishable from this 448 effect caused by using a finite number of pathlines. 449

For  $Wi \leq 0.122$ , the decay of  $I_m/I_0$  is practically the same for varying Wi, with the 450 exception that there is a small increase in  $I_m/I_0$  at large  $n_e$  for Wi = 0.122, which is explained 451 by the fact that the asymmetry grows in magnitude between Wi = 0.010 and Wi = 0.122. For 452  $Wi \ge 0.429$ , the decay of  $I_m/I_0$  seems relatively unaffected by the viscoelasticity of the fluid 453 for  $n_e \leq 2$ . However, for  $n_e > 2$  there is a sudden and sharp change in the decay of  $I_m/I_0$ , where 454  $I_m/I_0$  suddenly decays significantly slower for  $n_e > 2$  than for  $n_e \le 2$ . Since it has already 455 been shown in Figure 9a that, for  $Wi \ge 0.429$ , the viscoelastic flow asymmetry is observed 456 before  $n_e = 2$ , and is roughly constant for all  $n_e$ , the fact that  $I_m/I_0$  is relatively unaffected 457 by the increase in Wi before  $n_e = 2$  cannot be attributed to an onset of the flow asymmetries 458 at  $n_e = 2$ . Since the flow is symmetric (ie, not twisting) approaching the start of the first 459 mixing element  $(n_e = 0)$ , it is not be expected that the viscoelasticity of the flow would affect 460 the mixing caused by the first element, except in the case of exceptionally large Wi (far 461 beyond the range investigated in this study) where a purely symmetry-breaking instability 462 would be expected as the flow bifurcates over the flat-edge of the first mixing element. 463 This phenomenon would be similar in nature to the viscoelastic asymmetries observed for 464 symmetric confined cylinder geometries [20, 21]. It is possibly the case, since the striations 465 are relatively large at  $n_e = 1$ , that the observed flow asymmetry simply does not distort 466 the fluid enough to have a significant impact on the decay of  $I_m/I_0$  here. Similar results 467 were obtained by Migliozzi et al. [30], who found that the CoV, measured experimentally 468

using PLIF, was largely unaffected by the viscoelasticity of the fuid for the first two mixing elements, but then an increase in CoV (and hence reduction in mixing performance) was observed for increasing elasticity in the subsequent mixer elements. Since they used PLIF to investigate only the mixing performance, they were not able to explicitly relate the observed changes in the decay of the CoV to the changes in the flow kinematics induced by the viscoelasticity, however.



Figure 14: Normalised mixing index,  $I_m/I_0$ , defined in Equations (12) and (13) versus  $n_e$  for varying Wi and for (a) Re = 0.04 and (b) Re = 5.96. Solid black line shows  $c^{-x}$ , where c = 2.45.



Figure 15:  $I_m/I_0$  at  $n_e = 8$  (mixer outler) versus  $\Delta$  (computed at  $n_e = 4$ ) for all  $L^2 = 50$  simulations. Note that  $\Delta$  is virtually constant for each mixer element after the first couple of elements.

Figure 15 shows  $I_m/I_0$  at the outlet of the mixer section versus  $\Delta$  (computed at  $n_e = 4$ ) 475 for all simulations where  $L^2 = 50$  and the flow reached steady-state. There is a strong 476 proportionality between the quantification of the asymmetry  $\Delta$  and the mixing performance 477 quantified by  $I_m/I_0$ , indicating that the asymmetry in the flow is indeed the cause of the 478 change in the mixing performance, as expected. The non-zero y-intercept at  $\Delta = 0$  is caused 479 primarily by the fact a finite number of pathlines were used to quantify  $I_m$ , since the fitted 480 exponential decay for low  $n_e$  (2.45<sup>-x</sup>) gives a value of  $7.7 \times 10^{-4}$  for  $n_e = 8$ , which should be 481 the value of  $I_m/I_0$  in the case that  $N_A \rightarrow infty$ . 482

# 483 3.3. Effect of polymer extensibility

As mentioned previously,  $L^2$  characterises the limit of the extensibility in the FENE constitutive models. Reducing  $L^2$  in the FENE-CR model causes shear-thinning in  $N_1$  to occur at lower Wi, and also inhibits the extensional-thickening behaviour. Thus, reducing  $L^2$ should hypothetically lead to more stable viscoelastic flows for cases in which the extensibility causes large growths of polymeric stresses (e.g. flows with stagnation points).

In this study, we increase the value of  $L^2$  between 50 and 5000 for fixed values of Re = 0.49and Wi = 0.429. All results discussed previously were for cases in which  $L^2 = 50$ , and all of these simulations reached steady-state. It was observed in the helical static mixer that,

for  $L^2 > 50$ , the flow became time-dependent. Figure 16 shows  $\Delta$  as a function of time 492 for Re = 0.49 and Wi = 0.429 for the three values of  $L^2$  investigated. For  $L^2 = 500$  the 493 asymmetry, despite being time-dependent, seems to be fairly constant with respect to  $n_e$ 494 for the elements further down the mixer, with  $n_e = 4$  and  $n_e = 7$  both showing the same 495 behaviour in time. It should also be noted that for elements  $n_e = 4$  and  $n_e = 7$  the sign of  $\Delta$ 496 is opposite to those for  $L^2 = 50$ . For  $L^2 = 5000$ , the time dependence appears to be strong 497 for  $n_e = 7$ , with large time fluctuations in  $\Delta$  observed, particularly at  $t/\lambda \approx 3$  and  $t/\lambda \approx 11$ . 498 However, interestingly,  $\Delta$  at  $n_e$  = 4 appears to be fairly steady in time, albeit much larger in 499 magnitude. It is likely expected that for higher degrees of viscoelasticity, brought about by 500 either increasing  $L^2$  or Wi further, this time dependence will just grow stronger and more 501 complex since the system should transition into a chaotic state of elastic turbulence [19]. 502

Figures 17 and 18 show, respectively, contours of  $u_x$  at  $n_e = 5$  for  $L^2 = 500$  and  $L^2 = 5000$ . For  $L^2 = 5000$ , the asymmetry is significantly stronger than for  $L^2 = 50$  and  $L^2 = 500$ , with the flow almost totally bypassing the upcoming element intersection (starting horizontally). The previous element is twisting in a clockwise direction, and so the fluid is predominantly flowing against the twisting direction (i.e.  $\Delta < 0$ ).



Figure 16:  $\Delta$  versus  $t/\lambda$  at various  $n_e$  for various values of  $L^2$ . Simulations are initialised with a stationary field at t = 0.



Figure 17: Contours of  $u_x$  at  $n_e = 5$  for  $L^2 = 500$ . Flow direction is into the page, and the flow is just approaching the horizontal edge of the upcoming element.



Figure 18: Contours of  $u_x$  at  $n_e = 5$  for  $L^2 = 5000$ . Flow direction is into the page, and the flow is just approaching the horizontal edge of the upcoming element.

<sup>508</sup> Migliozzi et al. [30] report that the time-dependence of the mixing patterns, captured <sup>509</sup> by PLIF, is totally suppressed in the helical static mixer geometry for solutions of Xanthan

gum, whereas there is strong time-dependence for increasing elasticity with the PAA solutions 510 (Boger fluid). They attribute this to the fact that the Xanthan gum solution is expected 511 to have a near-constant extensional viscosity, whereas the PAA solution is expected to be 512 strain-thickening in extensional flows. Ramsay et al. [38] also reports time-dependence of the 513 flow of a PAA solution in the helical static mixer. Our results are in good agreement with 514 the previous experimental results, since, by increasing  $L^2$ , the strain-thickening behaviour 515 of the extensional viscosity is made more pronounced and the shear-thinning of  $N_1$  is made 516 less pronounced, giving rise to higher elastic normal stresses in complex flows (shear and 517 extensional) such as those observed in the static mixer geometry. 518

# 519 4. Conclusions

We have used CFD to model viscoelastic fluid flow in a helical static mixer for a range 520 of Reynolds and Weissenberg numbers. The results show that the viscoelasticity causes the 521 flow to bifurcate asymmetrically at the intersections of the mixer elements, which reduces 522 the mixing performance. Increasing viscoelasticity has been found to reduce the mixing 523 performance of a helical static mixer in previous experimental studies, and so the numerical 524 results from this study are in good agreement with experimental observations. However, with 525 the CFD results, we are able to uncover the exact driving mechanism for the reduced mixing 526 performance, having access to the necessary flow variables throughout the domain. The 527 results greatly help us to understand how the complex rheology affects the mixing process 528 in these flows. 529

The asymmetry at the element intersections has been quantified for the range of Re and Wi studied. It was shown that the sign of the asymmetry parameter (or the direction of the "bending" of the flow relative to the element twisting direction) exhibited complicated behaviour when varying Re and Wi. For low Re, the asymmetry parameter was negative and increased in magnitude with increasing Wi up until a critical Wi. Beyond this critical Wi, the sign of the asymmetry parameter changed abruptly, and the magnitude further

increased with increasing  $W_i$ , indicating the possible presence of a viscoelastic instability. 536 For high Re, the sign of the asymmetry parameter was always positive, and increased with 537 increasing Wi. However, the highest magnitudes of the asymmetry parameter (at high Wi) 538 were significantly lower for higher Re than for lower Re, indicating that inertia dampens the 539 mechanism responsible for the asymmetry. This dampening of the viscoelastic asymmetry 540 with increasing Re has been observed in simpler geometries previously. We show that the 541 asymmetry parameter does not follow either a perfect or an imperfect pitchfork bifurcation. 542 However, for low Re, and for Wi > 0.122, the results suggest that there may be two stable 543 solution branches for the asymmetry parameter. 544

Increasing the limit of extensibility in the viscoelastic constitutive model  $(L^2)$  caused 545 the simulation to change from steady to transient for intermediate values of Re = 0.49 and 546 Wi = 0.43. Previous experimental studies also showed that viscoelastic materials which are 547 expected to exhibit more extensibility showed stronger transient behaviour in the helical 548 static mixer than those expected to exhibit less extensibility. And so, again, the numerical 549 results agree with these experimental observations. The numerical results also show that 550 increasing  $L^2$  significantly increases the magnitude of the asymmetry, which is likely due to 551 the increased extensional viscosity and normal stresses. 552

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