Electronic Supplementary Material (ESI) for Soft Matter
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SUPPLEMENTARY INFORMATION for:

Extensional Rheometry and Elastic Instabilities of a Wormlike Micellar Solution in a Microfluidic Cross-Slot Device

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In this supplementary section we present additional results describing the nature of the flow field within the cross-slot geometry over a range of volume flow rates (*Q*) and Weissenberg numbers (*Wi*). We use micro-particle image velocimetry (μ-PIV) to briefly investigate how the re-entrant lip vortex that develops at the inlet channel of the cross-slot grows with increasing *Wi*. We also present movies constructed from time series of streakline images that provide a qualitative global impression of the flow field at different *Wi*, and complement the quantitative μ-PIV data presented in the main text.

Lip-vortex growth with increasing Weissenberg number

Twenty individual micro-particle image velocimetry (μ-PIV) images, performed as described in Section 2.5 of the main text, were averaged in order to obtain streamlines along the inlet channel of the cross-slot geometry over a range of volume flow rates and Weissenberg numbers. The results are presented in Fig. SI1a-e.

It is clear that with increasing *Wi*, the lip vortex at the re-entrant corner of the inlet channel (bottom-right corner in Fig. SI1a-d, bottom-left corner in Fig. SI1e) grows considerably in length, eventually extending beyond the field of view (Fig. SI1d,e). The vortex length L_v is defined as the distance from the nearest corner of the cross-slot to the point of detachment of the vortex from the inlet channel wall and is marked on Fig. SI1b,c for demonstration purposes. The vortex length L_v was normalized by the channel width (w = 200 μm) to obtain a dimensionless vortex length ($\chi = L_v / d$) as a function of *Wi*, which is shown in Fig. SI1f. The large error bars at the two highest Weissenberg numbers are due to the large extension and growth of the vortices beyond the field of view, which meant that the detachment point was determined by a linear extrapolation of the dividing streamline towards the channel wall. For the limited range of *Wi* examined, we find a functional form for the vortex growth of $\chi = 0.104(Wi - Wi_{vortex})$, where $Wi_{vortex} \approx$ 11, is the critical *Wi* for the appearance of the lip vortex.

Streakline imaging

The setup for visualization of streaklines in the CPyCl/NaSal solution consisted of a CCD camera (mvBlueFOX-120a, Matrix Vision GmbH), an inverted microscope (Nikon, Eclipse TE 2000-S) equipped with a G-2A filter cube and an external continuous light source (mercury lamp, illumination wavelength 532 nm). The wormlike micellar solution was seeded with 1.1 μm diameter fluorescent tracer particles (Nile Red, Molecular Probes, Invitrogen; Ex/Em: 520/580 nm; $c_p \approx$ 0.02 wt.%), which were illuminated by the light source and focussed through a 10× 0.25 NA microscope objective onto the CCD array. The CCD camera was operated at a frame rate of between 4 and 50 frames per second (fps), depending on the volume flow rate through the cross-slot. All images were recorded at the mid-plane of the microchannel.

Movie SI1 shows the streakline pattern for the 100 mM/60 mM CPyCl/NaSal solution at a volume flow rate of Q = 1 μL min^{-1} ($\dot{\varepsilon}_{nom}$ = 0.4 s^{-1}, *Wi* = 1). Here we observe a steady, symmetric, pseudo-Newtonian flow field. The movie is presented in real time.

Increasing the flow rate to Q = 1.5 μL min^{-1} ($\dot{\varepsilon}_{nom}$ = 0.6 s^{-1}, *Wi* = 1.5, Movie SI2) the flow is in the steady bifurcated regime. The majority of fluid entering via the bottom channel exits via the left-hand channel, and the majority of fluid entering via the top channel exits via the right-hand channel. Note the deformed shape of the central region of low velocity particles around the stagnation point. The movie is presented in real time.

At Q = 2 μL min^{-1} ($\dot{\varepsilon}_{nom}$ = 0.8 s^{-1}, Wi = 2, Movie SI3) the flow is in the fully bifurcated state with an asymmetry parameter, DQ = 1 (eqn (15), main text). All of the fluid entering via the bottom channel exits via the right-hand channel, and all the fluid entering via the top channel exits via the left-hand channel. The movie is presented in real time.

Still in the fully bifurcated state at Q = 50 μL min^{-1} ($\dot{\varepsilon}_{nom}$ = 20 s^{-1}, Wi = 50), Movie SI4 shows evidence of the development of lip vortices near the upstream re-entrant corners of the inlet channels. Observing the top-right and bottom-left sides of the inlet channels, it is clear that the fluid remains essentially stagnant whereas the fluid within the bulk of the inlet channels flows rapidly around the re-entrant corner of the cross-slot geometry. Occasionally, it is also possible to observe very low velocity particles trapped on the diagonal dividing streamline between the top-left and bottom-right corners. The movie is presented in real time.

The final movie, Movie SI5, was recorded at Q = 500 μL min^{-1} ($\dot{\varepsilon}_{nom}$ = 200 s^{-1}, Wi = 500) in the time-dependent flow regime. Due to the high flow velocity the highest possible frame rate was used (50 fps) to capture the dynamics as well as possible. The movie has been slowed down by 2.5× and clearly shows the unsteady nature of the flow field at this flow rate.

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FIGURES:

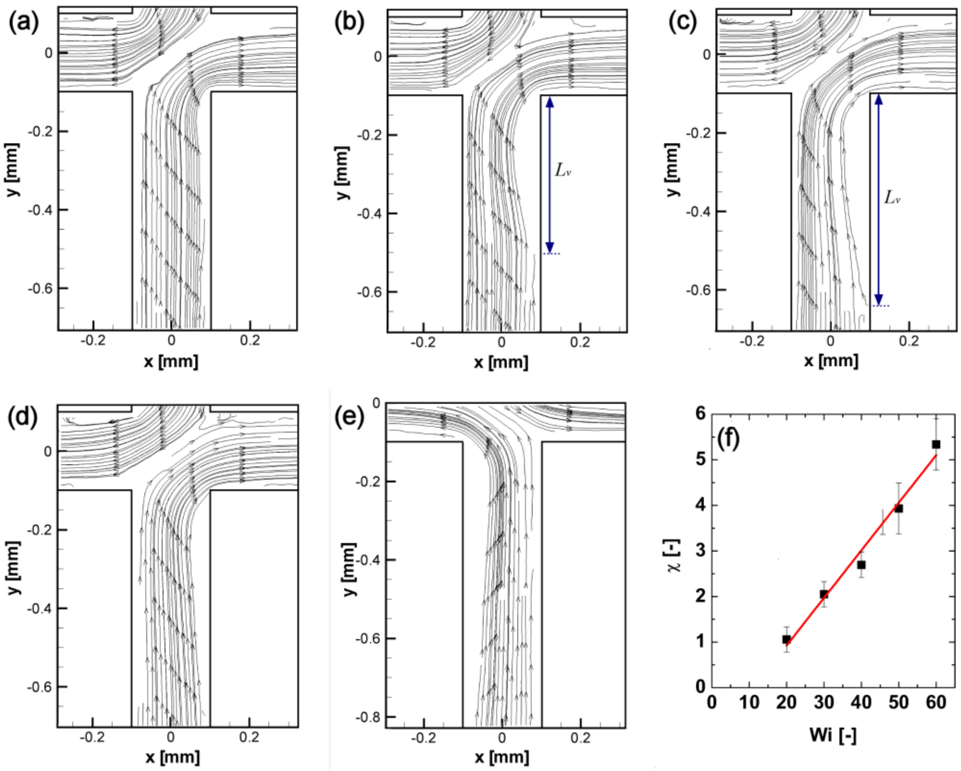

Fig SI1: (a) to (e) Streamlines demonstrating the growth of the lip-vortex with *Wi* along the inlet channel of the cross-slot: (a) Q = 20 μL min $^{-1}$, *Wi* = 20, (b) Q = 30 μL min $^{-1}$, *Wi* = 30, (c) Q = 40 μL min $^{-1}$, *Wi* = 40, (d) Q = 50 μL min $^{-1}$, *Wi* = 50, (e) Q = 60 μL min $^{-1}$, *Wi* = 60. (f) Dimensionless vortex length (χ) as a function of *Wi*, showing a linear relationship of the form $\chi = 0.104(Wi - Wi_{vortex})$, where $Wi_{vortex} \approx 11$.

MOVIE TITLES, DESCRIPTIONS AND KEYWORDS

- *Movie SI1: Cross-slot flow of CPyCl/NaSal solution at Q = 1 μL min^{-1}, Wi = 1.*

This movie demonstrates the steady, symmetric, pseudo-Newtonian nature of the flow field in the cross-slot for flow of 100 mM/60 mM CPyCl/NaSal solution at *Wi* = 1. Asymmetry parameter, $DQ = 0$.

KEYWORDS: Cross-slot, viscoelastic flow, extensional flow, flow field, microfluidics, wormlike micelles, elastic instabilities, flow bifurcation.

- *Movie SI2: Cross-slot flow of CPyCl/NaSal solution at Q = 1.5 μL min^{-1}, Wi = 1.5.*

This movie demonstrates the steady, asymmetric, nature of the flow field in the cross-slot for flow of 100 mM/60 mM CPyCl/NaSal solution at *Wi* = 1.5. Asymmetry parameter, $DQ \approx 0.25$.

KEYWORDS: Cross-slot, viscoelastic flow, extensional flow, flow field, microfluidics, wormlike micelles, elastic instabilities, flow bifurcation.

- *Movie SI3: Cross-slot flow of CPyCl/NaSal solution at Q = 2 μL min^{-1}, Wi = 2.*

This movie demonstrates the steady, fully asymmetric flow field in the cross-slot for flow of 100 mM/60 mM CPyCl/NaSal solution at *Wi* = 2. Asymmetry parameter, $DQ = 1$.

KEYWORDS: Cross-slot, viscoelastic flow, extensional flow, flow field, microfluidics, wormlike micelles, elastic instabilities, flow bifurcation.

- *Movie SI4: Cross-slot flow of CPyCl/NaSal solution at Q = 50 μL min^{-1}, Wi =50.*

This movie demonstrates the steady, fully asymmetric flow field in the cross-slot for flow of 100 mM/60 mM CPyCl/NaSal solution at *Wi* = 50. Asymmetry parameter, $DQ = 1$. Note presence of lip-vortices to top-right and bottom-left inlet channels.

KEYWORDS: Cross-slot, viscoelastic flow, extensional flow, flow field, microfluidics, wormlike micelles, elastic instabilities, flow bifurcation.

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- *Movie SI5: Cross-slot flow of CPyCl/NaSal solution at Q = 500 μL min*$^{-1}$, *Wi = 500.*

This movie demonstrates the unsteady, time-dependent, nature of the flow field in the cross-slot for flow of 100 mM/60 mM CPyCl/NaSal solution at *Wi* = 500.

KEYWORDS: Cross-slot, viscoelastic flow, extensional flow, flow field, microfluidics, wormlike micelles, elastic instabilities, flow bifurcation.