Microtomographic PIV measurements of viscoelastic instabilities in a 3D micro-contraction

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Viscoelastic flow through an abrupt planar contraction geometry above a certain Weissenberg number \( Wi \) is well known to become unstable upstream of the contraction plane via a central jet separating from the walls and forming vortices in the salient corners. Here, for the first time we consider three-dimensional (3D) viscoelastic contraction flows in a microfabricated glass square-square contraction geometry. We employ state-of-the-art microtomographic particle image velocimetry to produce time-resolved and volumetric quantification of the 3D viscoelastic instabilities arising in a dilute polymer solution driven through the geometry over a wide range of \( Wi \) but at negligible Reynolds number. Based on our observations, we describe new insights into the growth, propagation, and transient dynamics of an elastic vortex formed upstream of the 3D micro-contraction due to flow jetting towards the contraction. At low \( Wi \) we observe vortex growth for increasing \( Wi \), followed by a previously unreported vortex growth plateau region. In the plateau region, the vortex circulates around the jet with a period that decreases with \( Wi \) but an amplitude that is independent of \( Wi \). In addition, we report new out-of-plane asymmetric jetting behaviour with a phase-wise dependence on \( Wi \). Finally, we resolve the rate-of-strain tensor \( \mathbf{D} \) and ascribe local gradients in \( \mathbf{D} \) as the underlying driver of circulation via strain-hardening of the fluid in the wake of the jet.

Key words:

1. Introduction

Entry flow has historically received attention as a canonical case for non-Newtonian fluid dynamics (Boger (1987); White et al. (1987)), and as a benchmark for developing computational models capable of studying highly elastic flows (Afonso et al. (2011); Pimenta & Alves (2017)). Under negligible inertia (i.e., Reynolds numbers \( Re \ll 1 \)), for Weissenberg numbers \( Wi = \lambda \dot{\gamma} \), where \( \lambda \) is the fluid relaxation time and \( \dot{\gamma} \) the shear rate) beyond a critical value \( Wi_c \approx 0.5 \), pipe flow moving towards a contraction becomes sufficiently elastic that it separates from the upstream walls, forming a central ‘jet’ that enters the constriction and vortices around the mouth of the constriction (McKinley et al. (1991); Rothstein & McKinley (1999)). Initially the corner vortices are static in their placement as \( Wi \) is increased, but eventually grow in size until a Hopf bifurcation characterized by a periodic fluctuation of the vortex separation point occurs. For \( Wi > Wi_p \approx Wi_c \), the vortices become increasingly unsteady for increasing \( Wi \) (McKinley et al. (1991); Rothstein & McKinley (1999, 2001)), and may lead to a period-doubling route to chaos (McKinley et al. (1991)). The onset and subsequent dynamics of this elastic flow instability are highly sensitive to the

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contraction geometry and fluid rheology. Indeed, for certain contraction ratios ($\beta = w_0/w_c$, the length scale ratio of uniform channel $w_0$ to the contraction width $w_c$) a lip vortex may form for $Wi < Wi_c$ (Giesekus (1968)), as summarized by Rothstein & McKinley (2001). Rothstein & McKinley (2001) also showed that the appearance of lip vortices in contraction flow is accompanied by a greater contribution of shear flow compared to extension-dominated flow where corner vortices manifest. More recently, viscoelastic contraction flow has received attention at the microscale, whereby inertia can be neglected and elastic effects are dominant (see the thorough review provided by Rodd et al. (2007)). For the negligible $Re$ regime, there is an apparent void of knowledge regarding three-dimensional (3D) flow at moderate to high $Wi$. Due to the difficulty of resolving 3D flows at the microscale, and the immense computational burden of solving a transient 3D elastic flow numerically, this flow type has not been fully detailed either experimentally or numerically.

To date, elastic contraction flow has been studied primarily via localized or planar measurements such as particle image velocimetry (PIV), laser doppler velocimetry (LDV), or streak imagery. Global pressure measurements have also been employed to provide insights into drag reduction. In recent years, tomographic PIV (TPIV) has received increasing attention as a method whereby 3D flow volumes can be resolved via the reconstruction of a particle-laden flow from overlapping lines of sight, followed by cross-correlation between subsequent particle volumes (Elsinga et al. (2006a)). This method can also be applied at the microscale ($\mu$-TPIV), with multiple lines of sight provided by stereomicroscopy. Holographic PIV (HPIV) has also shown success in taking volumetric viscoelastic flow measurements in microscale geometries (Qin et al. (2019, 2020)), reporting a bistable negative wake ahead of a cylinder and out-of-plane instability modes along the flow separatrix of a cross-channel. However, HPIV is quite limited in terms of volume depth and reconstruction resolution compared to $\mu$-TPIV (Schäfer & Schröder (2011)). Nonetheless, the novel results from Qin et al. (2019, 2020) suggest that, despite decades of research on fundamental viscoelastic flows, deep insights are still yet to be elucidated once out-of-plane dynamics are captured.

Figure 1: (a) A sketch and micro-CT scan of the glass square-sectioned contraction-expansion channel. (b, c) Shear and extensional rheology of the polyacrylamide test solution. (d) A diagram of the microtomographic PIV apparatus.
Here, using a dilute solution of a high molecular weight polymer, we report the first investigations of 3D viscoelastic contraction flow at the microscale using the μ-TPIV method. We focus on the range of $Wi$ encompassing the transition from the vortex growth regime (which is accompanied by the growth of a steady central jet) to the onset of periodic vortex procession (which is accompanied by the circulation of the jet). We demonstrate that the circulation of the jet has phase-wise asymmetry dependence on the nominal $Wi$. By fully resolving the 3D velocity field, we can assess the true velocity gradient tensor and thus the rate-of-strain tensor. We show that the procession of the corner vortex is driven by the central jet continuously retreating from regions of increased rate-of-strain, and hypothesize that the underlying driving mechanism is localized strain-hardening of the polymer solution.

2. Experimental set-up

2.1. Flow cell and viscoelastic fluid

The experiments were conducted in a square-sectioned contraction-expansion flow cell (figure 1(a)) fabricated from fused-silica glass via selective laser-induced etching (Gottmann et al. (2012)) using a commercial LightFab 3D printer (LightFab GmbH). This process can resolve features on the micron scale, with a surface r.m.s. of approximately 1 μm (Pimenta et al. (2020)). We measured the channel width and height from an x-ray microtomography scan (figure 1(a)) as $w_0 = 860 \pm 10 \, \mu m$ outside the contraction, and $w_c = 255 \pm 5 \, \mu m$ inside the contraction, yielding a contraction ratio $\kappa = 3.4$. Figure 1(a) displays our dimensionless coordinate system, where each component is reduced by $w_0/2$ (e.g., $x^* = 2x/w_c$). Flow was stepped to each velocity by two syringe pumps in a push-pull configuration.

The viscoelastic test fluid was a polymeric solution composed of 107 parts-per-million (ppm) partially hydrolyzed polyacrylamide (HPAA, $M_W = 18$ MDa, Polysciences Inc., U.S.A.), in a solvent of 85 wt% glycerol and 15 wt% deionized water. The refractive index of the fluid is closely matched to that of the fused-silica flow cell. An Anton-Paar MCR 502 stress-controlled rheometer was used with a cone and plate geometry (50 mm diameter, 1° angle) to characterize the shear viscosity of the fluid under steady shear. Figure 1(b) shows that the fluid is weakly shear thinning and has a zero shear viscosity of $\eta_0 = 184 \, \text{mPa} \cdot \text{s}$. The relaxation time of the fluid was measured as $\lambda = 0.65 \, \text{s}$ using a capillary breakup extensional rheometer (Haake CaBER, Thermo Fisher Scientific, see Anna & McKinley (2001)) fitted with $d_0 = 6 \, \text{mm}$ diameter endplates. We plot the ratio of the apparent extensional viscosity $\eta_{E,app}$ to the zero shear viscosity $\eta_0$, against the accumulated Hencky strain $\epsilon_H = 2\ln(\kappa)$ in figure 1(c). The fluid exhibits strong strain-hardening with $\eta_{E,app} \approx 400\eta_0$ at high strains. The zero shear viscosity $\eta_0$ and the characteristic length scale $w_c/2$ are used to calculate $Re = \rho u_c w_c/2\eta_0$ and $Wi = 2\lambda u_c/w_c$. In the present work, $Re \lesssim 10^{-2}$ and as such is considered negligible.

2.2. Microtomographic PIV (μ-TPIV)

Volumetric flow measurement can be achieved by the TPIV method, which is termed μ-TPIV when conducted via stereomicroscopy. As implemented in a LaVision FlowMaster system (LaVision GmbH), μ-TPIV uses a stereomicroscope (SteREO V20, Zeiss AG, Germany) with dual high speed cameras (Phantom VEO 410, 1280 x 800 pixels) imaging a fluid volume illuminated by a coaxial Nd:YLF laser (dual-pulsed, 527 nm wavelength), see figure 1(d). The fluid was seeded with 2μm diameter fluorescent particles (PS-FluoRed, Microparticles GmbH, Germany) to a visual concentration of 0.04 particles-per-pixel.

The flow was recorded as double-frame images captured at 12 Hz, with a flow-rate
dependent time interval between laser pulses $\Delta t$ such that no particle moved more than 8 pixels. Frames were pre-processed with local background subtraction and Gaussian smoothing at $3 \times 3$ pixels. 3D calibration was performed by capturing reference images of a micro-grid at the planes $z = \pm 450 \, \mu m$ and $z = 0 \, \mu m$, fully encompassing the depth of the flow cell ($w_0$), and a coordinate system was interpolated between these planes using a third-order polynomial. Particle positions in 3D were reconstructed from the images using four iterations of the Fast MART (Multiplicative Algebraic Reconstruction Technique) algorithm implemented in the commercial PIV software DaVis 10.1.2 (Lavision GmbH). Fast MART initializes the particle volume using the multiplicative line-of-sight routine (MLOS Worth & Nickels (2008); Atkinson & Soria (2009)), followed by iterations of Sequential MART (SMART Atkinson & Soria (2009)). We concluded the algorithm with five iterations of the Motion Tracking Enhancement (MTE) method (Novara et al. (2010); Lynch & Scarano (2015)) to reduce spurious "ghost" particles which arise from randomly overlapping lines of sight (Elsinga et al. (2006b)), and thus do not correlate in time. Volume self-calibration (Wieneke (2008)) was employed to improve the accuracy of reconstruction. Particle displacements between particle volumes were obtained using a multi-grid iterative cross-correlation technique, with the final pass at $32 \times 32 \times 32$ voxels with 75% overlap for a vector grid of 31 $\mu m$. To reduce measurement noise the vector field was spatiotemporally filtered with a second-order polynomial regression across neighborhoods of $5^3$ vectors in space and extended through five increments in time for a total kernel size of $5^4$ points. This polynomial regression visibly reduced measurement noise while predominantly preserving space-time resolution owing to the small kernel size: the filtering wavelength is substantially smaller than the flow dynamics reported in this work. This is a common approach to de-noise TPIV data (Scarano & Poelma (2009); Elsinga et al. (2010); Schneiders et al. (2017)). Ultimately we resolved 1600 flow volumes per recording: a duration of 133 s (2004).

Uncertainty quantification for TPIV is a topic of ongoing research (Atkinson et al. (2011); Sciacchitano (2019)), but a priori comparisons of experimental velocity fields to direct numerical solutions or synthetic reconstructions have yielded a TPIV uncertainty on the order of 0.1–0.3 pixels (Atkinson et al. (2011)). As an a posteriori assessment of our measurement quality, we validated conservation of mass for time-averaged flow volumes (Zhang et al. (1997)). Flow divergence $\nabla \cdot \mathbf{u}$ was calculated at each vector, and the error assessed relative to an assumption of incompressible flow (relative error $\zeta = (\nabla \cdot \mathbf{u})^2/\text{tr}(\nabla \mathbf{u} \cdot \nabla \mathbf{u})$). The value $\zeta$ averaged 0.25 for $3 \leq Wi \leq 87$. Divergence error relative to the magnitude of vorticity was 0.14 at $Wi = 87$, in good agreement with a value of 0.2 from a three-camera TPIV experiment by Kempaiah et al. (2020).

3. Results and discussion

Measurements were taken over a range of flow velocities encompassing $3 \leq Wi \leq 87$ for a region of interest upstream of the contraction. Note that the downstream (expansion) side was imaged in a separate series of experiments, but flow remained steady across the $Wi$ range investigated. Throughout the discussion of the flow field kinematics we nondimensionalize lengths by $w_c/2$ and velocities by $U_c$ (the average flow velocity in the contraction). Deformation rates are hence reduced by $2U_c/w_c$. Times are nondimensionalized either by the fluid relaxation time $\lambda$ or by the fundamental period of circulation in the case of periodic flows. All nondimensional quantities are indicated by a superscript $^{**}$.

3.1. Steady flow at low $Wi$

As flow approaches the contraction at $Wi \leq 5$, a steady separation point forms where the flow separates from the walls of the channel and the flow passes as central jet through the
constriction. Figure 2 presents (a) average isosurfaces and (b) a midplane $x^*-y^*$ slice of the streamwise velocity $u_x^*$ for $Wi = 5$. The pink isosurface shows $u_x^* = 1/3$ (i.e., the central jet), while the grey surface marks $u_x^* = 0$ (i.e., the edges of the recirculating regions). Flow separation follows the upstream downzero crossing of $u_x^*$; the central jet is characterized by positive $u_x^*$, while the corner vortices drive negative $u_x^*$ backflow along the walls towards the separation point. We did not observe any lip vortices here, which agrees with indications from the literature whereby lip vortices are unlikely for abrupt contractions with $\beta > 2$ (Rothstein & McKinley (2001)), signaling that extensional flow is dominant over shear flow.

3.2. The onset of periodic instability

For increasing $Wi$, the corner vortex propagates upstream but remains steady, until, for $Wi > Wi_p$, the flow transitions to a periodic instability characterised by axial fluctuation of the upstream separation point and a circumferential procession of the central jet (see movie 1 in the supplemental material for animations of the jet for $5 \leq Wi \leq 44$). Figure 3(a-e) present space-time diagrams depicting the streamwise flow velocity $u_x^*(x^*)$ along the line $(y^*, z^*) = (-w_0/w_c, 0)$ (dashed red line in figure 2(a)) for five representative values of $3 \leq Wi \leq 87$. The dimensionless vortex length $L_v^*$ is determined by the zero-crossing of $u_x^*$ (as indicated on figure 3(a)). The average value of $L_v^*$ ($L_{v,avg}^*$) and the range of oscillation between $L_{v,max}^*$ and $L_{v,min}^*$ are plotted as a function of $Wi$ in figure 3(f), indicating a transition from steady vortex growth at lower $Wi$ to a regime of oscillation whereby $L_v^*$ apparently no longer scales directly with $Wi$. The inset of figure 3(f) shows the period of oscillation ($T^* = T/\lambda$) determined by FFT of the $L_v^*(t)$ signals. Above the critical value $5 < Wi_p < 11$ for the onset of oscillation, the range of oscillation reaches a local maximum by $Wi = 16$, and the mean separation point actually moves downstream (i.e., $L_v^*$ reduces) for $22 \leq Wi \leq 44$. Interestingly, the reduction in $L_v^*$ does not affect the frequency of the instability, with the period of oscillation continuously decreasing from $T^* = 26$ at $Wi = 11$ to $T^* = 7$ at $Wi = 44$ (figure 3(f)). Our results expand on prior experimental observations made in axisymmetric abrupt contractions which saw the vortex size increase monotonically for increasing $Wi$, although at a lower $Wi$ range than probed here due to larger length scales involved (e.g., $Wi < 5$ for McKinley et al. (1991) and $Wi < 8$, for Rothstein & McKinley (1999)). In planar microfluidic contraction flow experiments, Rodd et al. (2007) reported a decrease in $L_v^*$ for a single data point at the maximum $Wi = 24$ achieved, but were unable to extrapolate the trend. Numerical work by Comminal et al. (2016) observed an $L_v^*$ plateau.
accompanied by periodic vortex annihilation for $Wi > 14$ in a 2D contraction, which they attributed to an accumulation of elastic strain upstream of the contraction. However, they acknowledged that their use of the Oldroyd-B constitutive model (Oldroyd (1950)) lacks physical mechanisms (such as finite-extensibility) which would otherwise limit elastic stress.

### 3.3. Out of plane jet dynamics

As shown in the $x^* - t^*$ space-time diagrams in figure 3(a–e), the flow instability is strongly periodic for $Wi \geq 11$. Thus, to collapse the dataset and further reduce noise we deploy time synchronous averaging (TSA) to reduce the time dimension into a single average cycle. This method is further discussed in Bechhoefer & Kingsley (2009). We isolated the time series of velocity magnitude at a single point in the volume, then used the local maxima in the time series at that point to segregate each period at all points in the volume. The cycles are mapped to a normalized phase time $\phi^*$ ranging from 0 to 1 (i.e., 1 cycle), and averaged into $f_s T$ bins where $f_s$ is the sampling frequency (12 Hz) and $T = T^*/\lambda$ is the average period ($T^*$ shown in figure 3(f)). Henceforth, phase averaged quantities will be marked by $\langle \rangle$.

We used the phase-binned data to investigate the 3D trajectory of the central jet passing through the toroidal corner vortex. We found the maximum flow velocity in each $y^*-z^*$ plane along the $x^*$ axis for $-L_{v,r}^* \leq x^* \leq 0$. Thus, we extract $x^*-y^*-z^*$ trajectories of the circulating inner jet as it approaches the contraction. To quantify fluctuations in the jet velocity throughout phase time, we calculate the normalized fluctuating velocity along $x^*$ as $\langle \hat{U} \rangle (\phi^*) = (1/n_{x^*}) \sum_{x^*=-L_v^*}^{x^*=0} ((U)^*(x^*, \phi^*) - \langle \overline{U} \rangle^*(x^*)) / \langle \overline{U} \rangle^*(x^*)_{\text{max}}$. In this way $\langle \hat{U} \rangle$ is not affected by the accelerating flow velocity as the jet approaches the contraction. Data of $\langle \hat{U} \rangle$ are presented in figure 4(a) for $Wi \geq 5$, along with snapshots of the location of the jet in figure 4(b, c) for $Wi = 44$ and $Wi = 87$. We observe a phase-wise asymmetry of the jet in both the fluctuating component of velocity and the jet location. For each $Wi$, the jet starts at the same place in the channel at $\phi^* = 0$, but with an opposite directionality about $x^*$ for $Wi < 16$ (the direction of circulation is seemingly random between experiments). Maxima and minima in $\langle \hat{U} \rangle$ are evident for $Wi = 11$ near $\phi^* = 0.18, 0.88$ and 0.56, respectively, and apparent but slightly degraded by $Wi = 22$. The fact that the velocity fluctuation is small (< 5%) and almost symmetric about $\phi^* = 0.5$ indicates that the cyclical fluctuation in the $11 \leq Wi \leq 22$ regime is likely geometric in origin as the channel has an $\approx 5 \mu m$ (or $\approx 0.02 w_e$) variation between the width and height. At higher $Wi$ (e.g., $Wi = 44$ and $Wi = 87$), the jet had the

![Figure 3](image-url)
Dilute solutions of high molecular weight polymers are known to shear-thin under shear flow, and strain-harden under extensional deformations (Tirtaatmadja & Sridhar (1993); Solomon & Muller (1996)), as we observed for the HPAA fluid used in our work (figure 1(b, c)). Furthermore, as summarized in Rothstein & McKinley (2001), the lack of lip vortices observed in our experiments implies that extensional flow is dominant over shear flow. We can gain qualitative insight into the relevance of strain-hardening on the dynamics of our micro-contraction flow by considering the local rate-of-strain tensor \( D = (\nabla \mathbf{u} + \nabla \mathbf{u}^\top) / 2 \). Here we rely solely on the \( \mu \)-TPIV measurements to obtain the velocity vector field before calculating \( D \). We take the magnitude of \( D \) as \( \langle \dot{\gamma} \rangle = \sqrt{2(D : D)} \) (reduced as \( \langle \dot{\gamma} \rangle^* = \langle \dot{\gamma} \rangle w_c / (2U_c) \)) to highlight regions of high rate-of-strain where strain-hardening of the HPAA is more likely. We extracted \( y^* - z^* \) slices at an arbitrary \( x^* = -8 \) for \( Wi = 87 \) to show a simplified perspective on relationship between 0.5\( \langle \dot{\gamma} \rangle^* \) and the location of the jet (coloured contours of 0.5\( \langle U \rangle^* \), \( \langle U \rangle^* = \langle U \rangle / U_c \)) in figure 5(a-d). The subplots progress along phase time \( \phi^* \), with each plane including the 0.5\( \langle U \rangle^* \) contour from the following phase time. Two trends are noted: first that
while the jet (the solid coloured contour) stays centered about \( \langle \gamma \rangle^* \), the jet location forward in time is always towards the exterior of the \( \langle \gamma \rangle \) contour. Secondly, a phase-wise asymmetry manifests in the distribution of \( \langle \gamma \rangle^* \). \( \phi^* = 0.12 \) has low asymmetry in \( \langle \dot{U} \rangle \) (figure 5(b)), as well as in the distribution of \( \langle \gamma \rangle \). By \( \phi^* = 0.62 \), \( \langle \dot{U} \rangle \) is highly deviated and this aligns with a strong mismatch in \( \langle \gamma \rangle^* \) about the circumference of the jet. Therefore, it appears that a mismatch in rate-of-strain about the jet can strongly influence the phase-wise progression of the jet as it circulates, with positive and negative fluctuations in \( \langle \dot{U} \rangle \) accompanied by an unbalanced distribution of \( \langle \gamma \rangle^* \).

In figure 6, we present isosurfaces of 0.5\( \langle \gamma \rangle^* \) and 0.5\( \langle \dot{U} \rangle^* \), the maximum rate-of-strain and flow velocity, for \( Wi = 87 \). Figure 6(a-d) show 4 timesteps throughout a circulation, where the volume of high rate-of-strain forms a band about the circumference of the core of the jet (the pink isosurface). Moreover, the \( \langle \gamma \rangle^* \) isosurface extends further upstream on one side of the jet for all timesteps, i.e., the rate-of-strain is greater along one side of the jet. An animated loop of the jet circulating with the rate-of-strain volume is shown in supplemental movie 2.

We directly compare the rate-of-strain forward in time in figure 6(e) as projections of \( \langle \gamma \rangle^* \) from (a-d) from the \(-x^*\) direction. Moving clockwise from the \( \phi^* = 0.12 \) isosurface, each surface forward in time presents a decrease in the rate-of-strain in the clockwise direction. In other words, a perpetual retreat of the central jet from regions of increased rate-of-strain. The dynamics of elastic contraction flow has been reported to be sensitive to the extensional rheology (Rothstein & McKinley (2001)), and since our HP AA solution strain-hardens (figure 1(c)), we can infer from the flow kinematics that the central jet moves about the contraction driven by gradients of strain-hardening HPAA, which will tend to follow the regions of increased rate-of-strain. This sheds new light onto the likely significance of extensional rheology for local dynamics in viscoelastic contraction flow, as experimentally describing flow topology via directly resolving the rate-of-strain tensor has not been achieved hitherto for elastic flow instability.

### 4. Conclusions

Using \( \mu \)-TPIV, we have experimentally resolved for the first time the highly 3D dynamics of viscoelastic flow through a square-square micro-contraction at low \( Re \) and high \( Wi \). We captured steady and periodic flow instability for a central jet of fluid passing through a toroidal vortex pinned about the contraction entrance, and observed a new vortex growth plateau whereby the period of instability decreases with \( Wi \) but the enveloped vortex volume stagnates. For low \( 11 \leq Wi \leq 22 \), the jet circulates with a symmetric cyclical velocity fluctuation likely originating from geometric imperfections. This region coincides with the vortex growth plateau. At higher \( Wi \geq 44 \), a strong asymmetry in the jet forms which...
Figure 6: (a-d) Phase-averaged isosurfaces of $0.5(\dot{\gamma})^*$ and $0.5(U)^*$ for $Wi = 87$. (e) The $-x^*$ projection of the $\langle \dot{\gamma} \rangle^*$ surfaces from (a-d), coloured by their normalized phase time $\phi^*$. For advancing time $\phi^*$, the inner jet displaces towards decreasing rate-of-strain.

corresponds to exiting the growth plateau. This would indicate that the asymmetric mode provides a preferable route to vortex growth. We determined the first experimental mapping of the full rate-of-strain tensor to transient dynamics for viscoelastic flow instabilities. We relate regions of increased rate-of-strain and the extensional rheology of the fluid to the directionality of the circulating jet. Gradients of strain-hardening in the fluid provide a likely circulation mechanism as the jet retreats from locally strain hardened regions of the flowing viscoelastic polymer solution: a new insight on the significance of extensional rheology to local dynamics of viscoelastic contraction flow.

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