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A quantitative analysis of spatial extensional rate distribution in nonlinear viscoelastic flows

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ABSTRACT

A numerical method is proposed to calculate local extensional rate along arbitrary streamlines measured experimentally. It leads to the calculation of a "local Deborah number" (*De*) field, which characterizes heterogeneous flows of polymer solutions in a more insightful way than the commonly used nominal Weissenberg number (*Wi*), defined by an average shear rate. The spatial distribution of non-equilibrium polymer conformation across flow domain could be visualized through an image of the *De* field for flows of aqueous semi-dilute polyacrylamide (PAAm) solutions through the 4:1:4, 8:1:8 and 16:1:16 planar contraction geometries, over a range of *Wi* ($1.4 \leqslant Wi \leqslant 131.7$) and Elasticity numbers (*El*) ($13.3 \leqslant El \leqslant 476.8$). The time evolution of these *De* fields and their profiles near the contraction plane are presented. The correlation between the maximum local *De* and the nominal *Wi* is analyzed. The results show that the vortex formation near the contraction entry coincides with shifting the position of the maximum local *De* away from the centerline. Its actual location is sensitive to flow rate and strongly depends on dynamic flow regime.

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1. Introduction

The flow of aqueous polymer solutions through micro-fabricated planar flow channel with a sudden contraction exhibits intriguing nonlinear vortex dynamics [1,2]. A better understanding of the phenomena is of significance to a number of practical applications, such as ink-jet printing [3,4], high speed spinning, coating and painting process, subcutaneous injection of concentrated antibody solutions, oil recovery as well as rheometric characterization of low viscosity fluids at high deformation rate [5]. Nonlinear flows of several well-characterized polyethylene oxide (PEO) and polyacrylamide (PAAm) aqueous semi-dilute solutions through a range of micro-fabricated planar contraction geometries have also been quantitatively characterized using micro-particle image velocimetry (micro-PIV) and pressure drop measurements, in a range of *Wi* ($1.4 \leqslant Wi \leqslant 131.7$), *El* ($13.3 \leqslant El \leqslant 476.8$) and contraction ratios $4 \leqslant \beta \leqslant 16$ [6,7]. The further quantitative analysis of these experimental data could greatly enhance fundamental understanding of polymer dynamics in microscopic flows.

As it has been pointed out by Binding [8] and Binding and Walters [9], the vortex growth mechanisms observed in viscoelastic contraction flows are caused by the high extensional viscosity of polymer solution, especially so for semi-dilute polymer solutions with high Trouton's ratio. As fluid element approaches the contraction plane, polymer chains are extended and result in large elastic stresses. According to the theoretical analysis of de Gennes [10], to stretch a polymer chain under flow it is necessary to impose a sufficiently high extensional rate (to be greater than the inverse of the longest relaxation time of the molecular chain), and to accumulate a sufficient amount of strain on the molecule. Thus for a better understanding of the high *Wi* flows of polymer solutions through contraction geometries, it is desirable to estimate a degree of local extensional rate experienced by fluid elements from quantitative flow characterization. The extensional rate along the centerline of contraction flow could be easily calculated from the centerline velocity profile (see [2,6,7]). However in highly nonlinear flow regime, all streamlines become curved in the vicinity of the contraction throat. Moreover, high *Wi* flows are usually asymmetric and time-dependent. It is necessary to choose an appropriate reference frame to calculate local deformation rate, e.g. the streamline coordinate system, already used in simulations of complex viscoelastic fluid flows [11–13].

In the present work, a numerical method will be presented to calculate local extensional rate along arbitrary streamlines from experimental data measured by micro-PIV technique [7], and to visualize spatial distribution of non-equilibrium polymer conformation under complex nonlinear flows. Section 2 gives a summary

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Table 1
The properties of the PAAm solutions.

Fluid property	0.1% PAAm	0.25% PAAm	0.5% PAAm
η_0 [Pa s]	0.0024	0.0061	0.0174
ρ [g/cm^3]	0.998	0.994	0.89
c/c^*	3.3	8.3	16.6
λ [ms]	2.25	7.73	9.95

Table 2
The dimensions of the micro-fabricated flowcells. $D_h = \frac{2w_c h}{w_c+h}$ is the hydraulic diameter of the downstream narrow channel, $\beta = \frac{w_u}{w_c}$ is the contraction ratio, $\alpha = \frac{h}{w_u}$ is the aspect ratio of the upstream channel and $\varepsilon_H = \ln\frac{w_u}{w_c}$ is the Hencky strain.

	Flowcell 1	Flowcell 2	Flowcell 3
w_u [μm]	800	800	400
w_c [μm]	200	100	25
L_1 [mm]	20	20	20
L_2 [mm]		45	45
h [μm]	47	47	47
D_h [μm]	76.1	63.9	32.6
β	4:1:4	8:1:8	16:1:16
α	47/800	47/800	47/400
ε_H	1.38	2.07	2.77

Table 3
The flow rate Q, the nominal shear rates and the flow dimensionless numbers in three flowcells for the PAAm solutions. As in [7], the nominal shear rate in the downstream channel is $\dot{\gamma}_d = \frac{2Q}{hw_c^2}$, where Q is the flow rate, the Reynolds number is $Re = \frac{\rho\langle v_c\rangle D_h}{\eta_0} = \frac{2\rho Q}{\eta_0(h+w_c)}$, the nominal Weissenberg number is $Wi = \lambda\dot{\gamma}_d$ and the Elasticity number is $El = \frac{Wi}{Re} = \frac{\lambda\eta_0(w_c+h)}{\rho w_c^2 h}$.

	0.1% PAAm	0.25% PAAm	0.5% PAAm
Flowcell 1			
Q [ml/h]	\	\	2–35
$\dot{\gamma}$ [s^{-1}]	\	\	591–10,343
Re	\	\	0.05–4.02
Wi	\	\	1.4–102.9
El	\	\	25.5
Flowcell 2			
Q [ml/h]	\	0.5–13	05–11
$\dot{\gamma}$ [s^{-1}]	\	591–15,366	591–13,002
Re	\	0.31–6.8	0.1–2.12
Wi	\	4.5–100.5	5.9–129.4
El	\	14.8	60.8
Flowcell 3			
Q [ml/h]	0.025–2.8	\	0.05–0.7
$\dot{\gamma}$ [s^{-1}]	473–52,955	\	945–13,239
Re	0.08–8.95	\	0.02–0.27
Wi	1.06–119.1	\	9.4–131.7
El	14.7	\	476.8

of the materials and the experimental techniques, and method for quantitative analysis of micro-PIV data. In Section 3, the local Deborah number fields for flow of aqueous polyacrylamide (PAAm) solutions through planar micro-contractions with contraction ratios $\beta = 4, 8$ and 16 are presented. The correlation of the nominal *Wi* with the maximum local *De* for various *El* and contraction ratios is also analyzed and discussed. Conclusions are drawn in Section 4.

2. Experimental methodology and data analysis

2.1. Materials and rheometric characterization

1 wt.% (5 M) PAAm aqueous solution was purchased from Polysciences Europe GmbH and used as received. The average molecular weight M_w and the polydispersity index M_w/M_n as measured by standard GPC (TSK PW 6000 GPC column, an ERC 751A differential refractometer and a Waters 510 pump) are $M_w = 5.7$ M and $M_w/M_n = 34.4$. The radius of gyration was calculated as $R_g = 1578$ Å [14], and the overlap concentration was estimated as $c^* = 0.031$ wt%, using a simple cubic packing assumption. The 1 wt.% PAAm solution was subsequentely diluted with deionised water to prepare 0.1 wt.%, 0.25 wt.% and 0.5 wt.% PAAm aqueous solutions, ranging from $3.3c^*$ to $16.6c^*$, respectively. The rheometric properties of the PAAm solutions were characterized using a range of experimental techniques, including an ARES rotational shear rheometer (TA Instruments) with a double wall Couette geometry (cup OD = 34 mm, cup ID = 27.95 mm, bob OD = 32 mm, bob ID = 29.5 mm), and a Capillary Breakup Extensional Rheometer (CaBER, Thermo Fisher Scientific). The density ρ, the zero-shear viscosity η_0 and the CaBER relaxation time λ of the PAAm solutions are listed in Table 1. The error over 10 repeated measurements of λ was estimated as $\epsilon_{CaBER} \cong 7\%$. Other rheometric properties of the PAAm solutions can be found in [7].

2.2. Micro-channel geometry

The characteristic dimensions of contraction–expansion flow cells with contraction ratio $\beta = 4, 8$ and 16 (labeled as Flowcell 1, 2, and 3 respectively) are given in Table 2, and a schematic diagram is presented in Fig. 1. The microfluidic chips are made of poly (methyl methacrylate) (PMMA) and aromatic epoxy, and were fabricated by standard soft lithographic method (Epigem Ltd., UK). The internal surfaces were treated by oxygen plasma to make them hydrophilic. The length L of the narrow channel was set to be 20 mm so that the ratio L/w_c is much larger than unity for each

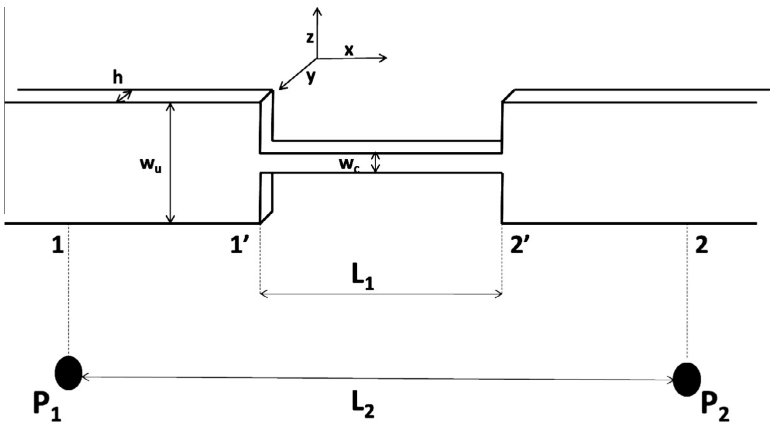

Fig. 1. Schematic diagram of microfluidic flow geometry. 1 and 2 are the positions of the pressure taps.

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