

# Polymeric filament thinning and breakup in microchannels

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The effects of elasticity on filament thinning and breakup are investigated in microchannel cross flow. When a viscous solution is stretched by an external immiscible fluid, a low 100 ppm polymer concentration strongly affects the breakup process, compared to the Newtonian case. Qualitatively, polymeric filaments show much slower evolution, and their morphology features multiple connected drops. Measurements of filament thickness show two main temporal regimes: flow- and capillary-driven. At early times both polymeric and Newtonian fluids are flow-driven, and filament thinning is exponential. At later times, Newtonian filament thinning crosses over to a capillary-driven regime, in which the decay is algebraic. By contrast, the polymeric fluid first crosses over to a second type of flow-driven behavior, in which viscoelastic stresses inside the filament become important and the decay is exponential. Finally, the polymeric filament becomes capillary-driven at late times with algebraic decay. We show that the exponential flow thinning behavior allows a novel measurement of the extensional viscosities of both Newtonian and polymeric fluids.

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## I. INTRODUCTION

The progressive breakup of an initially stable fluid thread into small drops or bubbles is a rich phenomenon of great interest [1]. For example, flow focussing in microfluidic devices can continuously produce drops or bubbles whose sizes are controlled by the relative flow rate of the two immiscible fluids [2–7]. While most such work concerns Newtonian fluids, many fluids of interest for lab-on-a-chip applications are likely to exhibit complex micro-structure and non-Newtonian behavior, such as viscoelasticity. Furthermore, viscoelastic effects, which can be quantified by the Elasticity number  $El = \lambda\eta/(\rho L^2)$ , scale inversely with the square of the device length scale ( $L$ ), and are likely to be accentuated in microfluidic devices. Here,  $\lambda$  is the fluid relaxation time,  $\eta$  is viscosity, and  $\rho$  is density. For polymeric drop breakup in macroscopic flow, elasticity can give rise to breakup behavior that is markedly different from that of Newtonian fluids [8–11]. For example, a viscoelastic filament driven by gravity in a quiescent bath [12] undergoes an initial linear viscous decrease in the filament diameter, followed by a slower thinning process in which capillary forces are balanced by the fluid elastic stresses.

Recently, a numerical investigation in a flow-focusing device [13] showed qualitative differences with respect to Newtonian fluids such as prolonged thinning of the fluid filament and delay of drop pinch-off. No measurements of thinning rates or breakup times were presented. An experimental investigation in a ‘T’ shaped geometry using a low viscosity, elastic fluid [14] also found prolonged thinning of the fluid filament. The authors observed a linear decrease in filament diameter followed by a ‘self-thinning’ exponential regime, which was argued to have a rate inversely proportional to the fluid relaxation time ( $\lambda$ ). However,  $\lambda$  was found to vary over an order of magnitude with shear rate, though it should remain

constant. While both investigations found similar qualitative trends, no quantitative connection has yet been made to the extensional flow within the filament during thinning and breakup.

In this paper, we compare the filament thinning and breakup of Newtonian and viscoelastic fluids of equal shear viscosity in a microchannel cross-slot geometry. Here, the outer Newtonian fluid stretches the inner Newtonian or polymeric fluid into a thin filament until it eventually breaks up into drops. This geometry allows for very fine control of the flows over a broad range of shear rates. Measurements of filament thickness show two temporal regimes: (i) a flow-driven regime in which the filament thins exponentially and (ii) a capillary-driven regime in which the filament thins algebraically. Our analysis leads to a novel method of measuring the extensional viscosities of both Newtonian and polymeric fluids. The thinning behavior allows comparison with a microscopic model relating polymer extension to rheology.

## II. METHODS

The experimental configuration is a cross slot microchannel,  $W = 50 \mu\text{m}$  wide and  $L = 30 \mu\text{m}$  deep, molded in poly(dimethylsiloxane) (PDMS, Dow Sylgard 184) using standard soft-lithography methods [15, 16]. Channels are sealed with a glass cover slip after exposure to an oxygen plasma. In order to keep the microchannel wetting properties uniform, the glass cover slip is coated with a thin layer of PDMS prior to the exposure. The assembled channels are then baked for 12 hrs at 100 °C in order to obtain hydrophobic walls wetted by the continuous liquid phase.

The continuous phase is mineral oil containing 0.1% by weight of surfactant (SPAN 80, Fluka). Two types of dispersed phases are used: a Newtonian fluid and a

polymeric fluid. The Newtonian fluid is a 90%-glycerin aqueous solution. The polymeric fluid is made by adding 100 ppm of high molecular weight polyacrylamide (PAA,  $M_W = 18 \times 10^6$ , 15% polydispersity), which has a flexible backbone, to a Newtonian 85%-glycerin aqueous solution with a measured shear viscosity of 0.2 Pa s; the water/glycerin mixture is used as a solvent for the polymer. It is dilute, below the overlap concentration of approximately 350 ppm. The interfacial tension between the continuous and dispersed phases is  $\sigma = 10$  mN/m. Shear rheology is characterized with a stress-controlled rheometer at 25 °C. As shown in Fig. 1, the shear viscosities of the oil and Newtonian fluids are nearly identical and independent of shear rate:  $\eta_s \approx 0.24$  Pa s. Also as shown, the viscoelastic polymeric fluid exhibits nearly constant shear viscosity (power law index=0.97) and a first normal stress difference  $N_1$ , which increases with shear rate.

We fit the polymeric fluid shear rheology data to the widely-used FENE-P model (finite extensibility nonlinear elastic with Peterlin's closure) [17]. This model is well adapted for dilute (and semidilute) high molecular weight polymeric solutions, and has been used previously to analyze filament thinning of polymeric fluids in macroscopic experiments [9]. A fluid described by the FENE-P model possesses the same dynamical properties as a fluid described by the much simpler Oldroyd-b model [18], which assumes that polymers can be modeled as Hookean springs. The main difference is that the Oldroyd-b model allows for infinite extension of polymer molecules, while the FENE-P model uses a spring-force law in which the polymer molecules can be stretched only by a finite amount in the flow field [17, 18].

A simultaneous fit of the polymeric fluid  $\eta_s$  and  $N_1$  data to the FENE-P model provides the fluid relaxation time  $\lambda = 0.45$  s and a dimensionless finite extensibility parameter  $b = 4500$ , which are the only two adjustable parameters (Fig. 1). To perform the fit, we also need values of the water/glycerin solvent viscosity (0.2 Pa s) and the number density of polymer molecules in the solution ( $4.4 \times 10^{12}$  cm<sup>-3</sup>); both quantities are constants. Details on the equations and methods used to fit the FENE-P model to the shear rheology can be found elsewhere [19].

The dispersed and continuous phases are injected into the central and side arms of the cross-channel, respectively, using syringe pumps (Harvard Instruments). Experiments are performed for flow rate ratios,  $q = Q_{oil}/Q_{aq}$ , ranging from 10 to 200. In all cases, the aqueous flow rate is kept constant at  $Q_{aq} = 0.01$  l/min. This is low enough that the behavior is quasi-static, such that the periodicity -but not the morphology- depends  $Q_{aq}$ . For this range of parameters, the Reynolds number is less than 0.01; therefore viscous forces are much larger than inertial forces. Similarly the capillary number ranges from 0.02 to 0.8; therefore, viscous forces are also larger than surface forces. Under these conditions an aqueous filament is formed and stretched by the flow of the surrounding oil. The thinning and breakup of the filament are imaged using an inverted microscope and a fast video

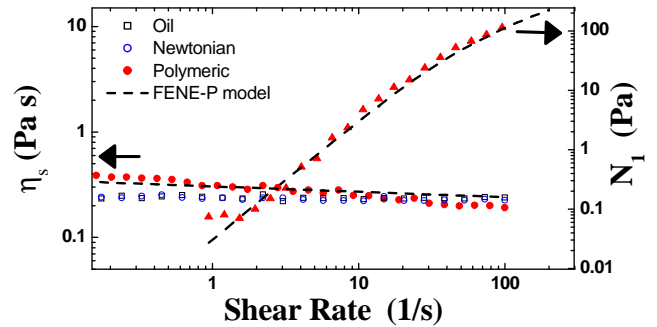


FIG. 1: (Color Online) Fluid rheological characterization. (Left y-axis) Shear viscosity vs shear rate for all fluids; oil=mineral oil; Newtonian=water/glycerin mixture; polymeric=PAA in water/glycerin mixture. The shear viscosity is nearly constant even for the polymeric solution  $\eta_s \approx 0.24$  Pa s. (Right y-axis) First normal stress difference for the polymeric solution vs shear rate. Dashed curves represent fits using the FENE-P model with parameters  $\lambda = 0.45$  s and  $b = 4500$ .

camera, with frame rates between 1 and 10 kHz.

### III. RESULTS

#### A. Qualitative Behavior

Sample frames from video data are shown in Fig. 2, for both Newtonian and polymeric fluids, at a flow rate ratio of  $q = 60$ . The Newtonian case, shown in the *left-column*, displays typical filament thinning and drop formation. The aqueous phase is drawn into the cross-slot channel (a), and begins to elongate and collapse (b-d), forming a primary drop connected by a very thin filament; later (e) the filament thins at a faster rate and breaks into a large primary drop and small satellite droplets.

The polymeric case, shown in the *right-column* of Fig. 2, displays very different behavior. Initially (a), we observe a morphology that is similar to that of the Newtonian fluid, i.e. viscoelasticity is negligible at first. As the thinning progresses, the polymeric fluid develops a longer neck with a drop attached to it (b). This filament elongates while thinning at a slower rate than in the Newtonian case (c). Near the breakup event, the polymeric fluid shows multiple beads ('beads-on-a-string') attached to the filament (d) [8, 10, 20]. After breakup, there are many satellite drops (e).

#### B. Quantitative Behavior

Filament thinning is quantified by the decrease in diameter,  $h(t)$ , as a function of time. To accomplish this, we fit a third-order polynomial equation to the interface contour in the cross slot region. We assume that the interface is symmetric across the centerline and only half of the contour is fitted with the polynomial. We then

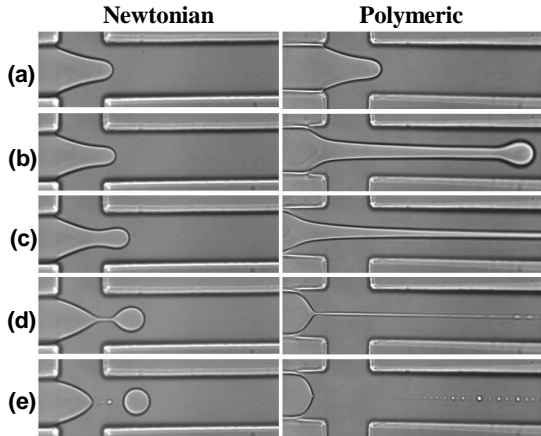


FIG. 2: Evolution of the thinning process for Newtonian (left column) and polymeric fluids (right column), for a flow rate ratio  $q=Q_{oil}/Q_{aq}=60$ , where  $Q_{oil}/Q_{aq}$  corresponds to the oil and aqueous phase flow rates, respectively. Oil is the continuous (outer) phase while the aqueous phase is either Newtonian or polymeric. (a) Initial regime; (b)  $t/t_b = 0.15$ , where  $t_b$  is breakup time; (c)  $t/t_b = 0.45$ ; (d)  $t/t_b = 0.95$ ; (e) after breakup. Values of  $t_b$  for the Newtonian and polymeric cases are 11.5 ms and 245 ms, respectively. Note the appearance of satellite droplets in the Newtonian case and multiple beads attached to the filament in the polymeric case (d,e). The channel width and depth are 50  $\mu\text{m}$  and 30  $\mu\text{m}$ , respectively.

locate the minimum in the polynomial first derivative. The filament diameter is measured at the point where the minimum in the first derivative is located; this minimum is usually located in the cross-slot region. Example results are shown in Fig. 3(a) for three flow rate ratios,  $q = 10, 30,$  and  $60$ . At short times, the Newtonian and polymeric fluids exhibit identical initial thinning, which is indicative of their common  $\eta_s$ . But at longer times, the two diverge with the polymeric filament lasting at least an order of magnitude longer before breakup. We also note shorter breakup times as  $q$  is increased. This trend is also found in other flow-focusing experiments [2, 21] and in a numerical investigation [22] using Newtonian fluids.

The filament strain rate is computed from  $h(t)$  data by assuming a homogeneous uniaxial extensional flow inside the filament:  $\dot{\epsilon} = -(2/h)dh/dt$  [23, 24]. While this method gives a good indication of  $\dot{\epsilon}$  when the filament thread is axisymmetric, it may break down at very early stages of the thinning process when the filament may be confined by the channel walls. However, we are confident in our measurements and methods since we are able to recover nearly the exact theoretical value of extensional viscosity expected for the Newtonian fluid (cf. Fig. 5).

Results for the same three flow rate ratios are given in Fig. 3(b). For the Newtonian fluid,  $\dot{\epsilon}$  is initially independent of time; therefore, in this regime,  $h(t)$  decreases exponentially with time. For the polymeric fluid,  $\dot{\epsilon}$  is initially equal to the same constant as for the Newtonian fluid. But it soon departs and, after a transient interval,

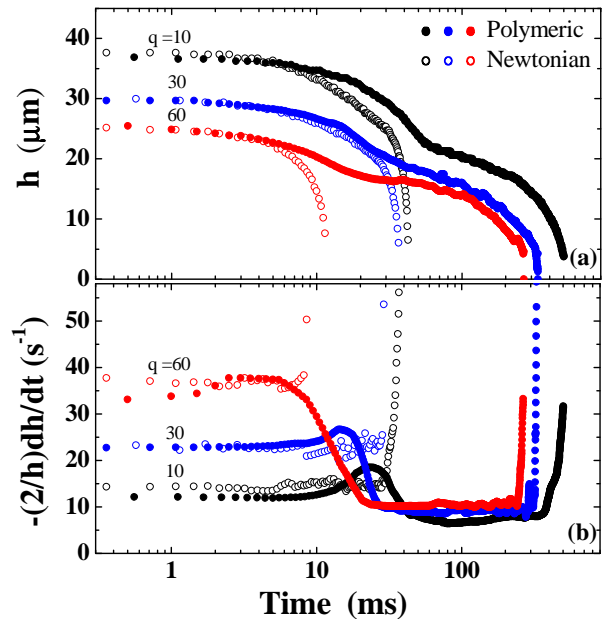


FIG. 3: (Color Online) (a) Filament thickness  $h(t)$  for both Newtonian and polymeric fluids for  $q=10, 30,$  and  $60$ . (b) Filament extensional strain rate  $\dot{\epsilon}=-(2/h)dh/dt$  for the same fluids. Both viscous and elastic regimes are characterized by constant  $\dot{\epsilon}$ .

settles down to smaller constant value, indicating a second regime of slower exponential thinning. For all fluids at the very latest times, close to breakup, the final decrease of  $h(t)$  to zero gives an apparent divergence of  $\dot{\epsilon}$ . We show in the following discussion that the data just before breakup are consistent with a linear decrease in filament diameter,  $h(t) \propto (t - t_b)$  where  $t_b$  is the breakup time.

### C. Flow-Driven Regime

To model the exponential decrease of filament diameter, we assume that (1) filament thinning is driven mainly by the outer fluid extensional flow in the cross-slot region and (2) the shear flow that develops is relatively far downstream from the cross-slot region and should have no implications on the local stress balance. These are reasonable assumptions since shear stresses tangential to the filament do not contribute to the thinning (or squeezing) of the filament; filament thinning is driven by viscous stresses normal to the filament.

Starting from an assumption of stress balance inside and outside the interface, and applying the definition of extensional viscosity [25], we obtain the condition  $\eta_e \dot{\epsilon} = \eta_{e,oil} \dot{\epsilon}_{oil}$ . Here, the left and right sides are the extensional viscosity multiplied by the extensional strain rate for the aqueous filament and continuous oil phases, respectively. As noted above, the strain rate in the filament is  $\dot{\epsilon} = -(2/h)dh/dt$ . The strain rate for oil in

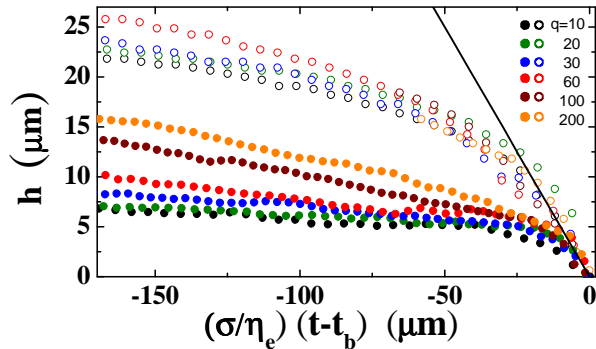


FIG. 4: (Color Online) Capillary driven breakup regime. At very late times, the filament thins roughly linearly in time with a speed proportional to  $\sigma/\eta_e$  for both Newtonian (open circles) and polymeric (filled circles) fluids. The flow ratio ( $q$ ) is color-coded in the legend for both cases. The solid line represents slope  $= -1/2$ .

the cross-slot region is  $\dot{\epsilon}_{oil} \approx Q_{oil}/(W^2L)$ , as verified by particle-tracking methods [26]. Lastly, since the oil is Newtonian, its extensional viscosity is  $\eta_{e,oil} = 3\eta_{s,oil}$  [25, 27]. Therefore, also assuming that  $\eta_e$  is independent of time, the filament diameter thins exponentially according to

$$h(t) = h_o \exp[-(3/2)(\eta_{s,oil}/\eta_e)\dot{\epsilon}_{oil}t]. \quad (1)$$

where  $h_o$  is an integration constant. In such *flow-driven* regimes, Eq. (1) may be used to deduce  $\eta_e$  from  $h(t)$  data.

We note that the quantity  $\dot{\epsilon}_{oil}$  is measured in the cross-slot region, where the flow is extensional and where pinching from the ‘mother drop’ occurs. To this end, we have checked that  $\dot{\epsilon}_{oil}$  remains constant during the filament thinning and breakup event; the average velocity of the oil in the cross-slot region is constant.

#### D. Capillary-Driven Regime

The linear decrease of the filament thickness near the final breakup can also be modeled by stress balance, now by incorporating surface tension effects. Specifically, the Rayleigh-Plateau instability eventually sets in so that capillary forces cause beading and ultimately breakup. Equating radial stress with the Laplace pressure gives  $\eta_e\dot{\epsilon} = \sigma/h$  [7, 28, 29]. Therefore, the filament diameter thins linearly with time according to

$$h(t) = -(1/2)(\sigma/\eta_e)(t - t_b), \quad (2)$$

where  $t_b$  is the breakup time. In such *capillary-driven* regimes, Eq. (2) may be used to deduce  $\eta_e$  from  $h(t)$  data. Equation 2 shows that, near the singularity,  $h(t) \sim \sigma/\eta_e$ , which has been observed numerically [30] in Stokes regime, except that, in the numerical work, shear rather than extensional viscosity is used in the denominator.

To demonstrate the consistency of extensional viscosity results from the flow- and capillary-driven regimes,

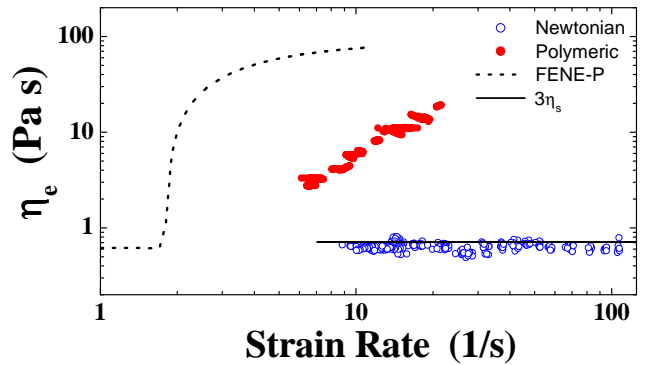


FIG. 5: (Color Online) Extensional viscosities of both Newtonian and polymeric fluids, derived from the filament thinning measurements and Eq. (1), as a function of the extensional strain rate  $\dot{\epsilon} = -2/h(dh/dt)$ . The polymeric fluid extensional viscosity shows strain hardening and increases with a power law exponent of approximately 1.0. The theoretical Trouton ratio of a Newtonian fluid is 3.0 (solid line). The FENE-P model prediction is also shown, but is far from the measurements.

we plot data for  $h(t)$  vs  $(\sigma/\eta_e)(t - t_b)$  in Fig. 4. There, the value of  $\eta_e$  is taken from analysis of the flow-driven regime using Eq. (1). To within apparently random deviations,  $h(t)$  data vanish linearly vs  $(\sigma/\eta_e)(t - t_b)$  with slope  $-1/2$ , in accord with Eq. (2). Note however that the dynamic range is limited, since the imaging resolution is about  $2 \mu\text{m}$ . Therefore, the capillary-driven regime is consistent with the flow-driven regime, but the latter gives more accurate values of extensional viscosity  $\eta_e$ .

## IV. DISCUSSION

The extensional properties of polymeric fluids are important for applications such as turbulent drag reduction and splash suppression [25, 31]; however, measurement of  $\eta_e$  has remained a difficult task [32]. In what follows, we show that high-quality data on the values of steady extensional viscosity for both polymeric and Newtonian fluids can be obtained using our method.

Final results for  $\eta_e$  based on Eq. (1) are plotted in Fig. 5 vs extensional strain rate. Here each point represents a different fixed flow-rate ratio,  $q$ . For the Newtonian fluid,  $\eta_e$  is independent of strain rate and nearly equals  $3\eta_s$  as expected [25, 27]. This agreement serves as a second check, complementary to Fig. 4. For the polymeric fluid at early times, in the first flow-driven regime, the behavior is the same as for the Newtonian fluid (not shown). At later times, in the second flow-driven regime, the extensional strain rate of the filament is lower and  $\eta_e$  is higher. This ‘strain hardening’ behavior is due to the stretching of the polymer molecules in the extensional flow of the thinning filament, and it has been observed in other macroscopic experiments [23, 33].

In Fig. 1, the FENE-P model properly describes both

the  $\eta_s$  and  $N_1$  versus shear rate with two adjustable parameters, which are  $\lambda=0.45$  s and  $b=4500$ . Using these values and the solvent shear viscosity value (0.2 Pa s), the FENE-P prediction for  $\eta_e$  is plotted in Fig. 5. It exhibits strain-hardening behavior, which saturates at high strain rates by accounting for the finite extensibility of the polymer molecules. However, by comparison with our data, the predicted strain hardening sets in too soon and too abruptly. A possible source of error may be polymer dispersivity ( $\sim 15\%$  in  $M_W$ ), which can smear out the sharp rise in  $\eta_e$  [9]. It cannot, however, account for such early transition to strain hardening behavior since  $\lambda \sim M_W^{3/2}$ .

Other sources of error may be the inherent limitations of the FENE-P model such as the pre-averaging force connectors in the dumb-bell model originally proposed by Peterlin [17]. This averaging is known to lead to unexpectedly large polymeric stresses compare to the non-averaged FENE model [34]. Another limitation is that while real polymeric fluids have a spectrum of  $\lambda$ , the FENE-P model, as used here, is described by a single  $\lambda$ . Therefore, we should expect some type of failure of predictions of  $\eta_e$  based on the FENE-P model.

## V. CONCLUSION

In conclusion, small amounts of flexible polymer can dramatically affect filament thinning and breakup in micro-channel extensional flow. In contrast to macroscopic observations, we find both a *flow-driven* regime in which the filament thins followed by a *capillary-driven* regime responsible for filament breakup. For a Newtonian fluid, the filament thins exponentially with time until onset of capillary surface tension-induced breakup.

For a polymeric fluid, with the same shear viscosity - nearly independent of shear rate, there is an intermediate regime in which the filament thins exponentially at a much slower rate. Furthermore the capillary regime features generation of a series of small droplets along the filament. These differences may be attributed solely to extensional viscosity, and its increase with extensional strain rate, since this is the only rheological difference between the Newtonian and polymeric fluids. For thinner filaments and faster thinning, the polymer molecules stretch and cause an increase in extensional viscosity without significant change in shear viscosity.

Measurements of the exponential rate of thinning can thus be used to determine extensional viscosity, an elusive quantity to measure. For the Newtonian case,  $\eta_e \approx 3\eta_s$ ; for the polymeric case, the results increase with extensional strain rate but much less slowly than predicted by the FENE-P model. This suggests the need for a better understanding of both the molecule-scale behavior of polymers in extensional flows as well as its connection to macroscopic rheology. Filament thinning in microchannels, and its variations with polymer molecular weight, may be a promising approach.

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