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Velocity fluctuations of fracturelike disruptions of associating polymer solutions

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Velocity has been measured as a function of time for propagating crack tips as water is injected into solutions of end-capped associating polymers in a rectangular Hele-Shaw cell. Measurements were performed for flows with different values of cell gap, channel width, polymer molecular weight, and polymer concentration. The condition for the onset of fracturelike behavior is well described by a Deborah number which uses the shear-thinning shear rate of the polymer solution as a characteristic frequency for network relaxation. At low molecular weight, the onset of fracturelike pattern evolution is accompanied by an abrupt jump in tip velocity, followed by a lower and approximately constant acceleration. At high molecular weight, the transition to fracturelike behavior involves passing through a regime that may be understood in terms of stick-slip dynamics. The crack-tip wanders from side to side and fluctuates (in both speed and velocity along the channel) with a characteristic frequency which depends linearly on the invading fluid injection rate.

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In a previous study of radial viscous fingering patterns in Hele-Shaw cells filled with viscoelastic aqueous solutions of end-capped associating polymer, we observed a transition from viscous fingering to fracturelike behavior [1]. The fracturelike behavior was easy to distinguish from viscous fingering when the full time series of patterns was observed because the large angle branches, which were obvious in any one stage of development of the pattern, could be observed to grow from a branch at very large distances behind the propagating crack tip, a clear departure from viscous fingering where the (generally much smaller angle) sidebranches always result from tip splitting. Further examination of the fracturelike phenomenon showed that a Deborah number could be defined as a ratio of the polymer network relaxation time (taken to be the inverse shear-thinning shear rate) to the inverse external shear rate (or viscous flow characteristic time), that is,

$$\mathrm{De} = \frac{\tau_r}{\tau_f} \ , \quad \tau_f \sim \frac{b^2}{Q} \ , \quad \tau_r = \frac{1}{\dot{\gamma}_0} \ , \tag{1}$$

with b being the cell gap, Q the volumetric injection rate, and $\dot{\gamma}_0$ the shear-thinning shear rate of the polymer solution. τ_f would also contain a characteristic length of the radial cell, which is irrelevant if the same cell is used in all the experiments. The transition to fracturelike behavior appeared at roughly the same Deborah number under variations of polymer molecular weight or polymer concentration (rheological properties) and variations of cell gap (change in cell geometry). That is, the water injection rate needed to produce a fracture could be predicted rather well for arbitrary values of the other control parameters using the empirically determined critical value of the Deborah number.

By returning to the simpler problem of flow in a Hele-Shaw channel, one can restore the basic viscous fingering problem to having a steady state rather than asymptotic behavior characterized by continuous tip splitting with a decreasing average tip velocity. In analogy to the measurements of Fineberg *et al.* for fracture in brittle solids [2], the study of the tip velocity evolution and particularly its departure from the constant velocity in the fingering regime should give us some insight into the dynamics of this transition into fracturelike behavior. In this paper we report the results of measurements of the onset and development of fracturelike behavior in a Hele-Shaw channel.

Our Hele-Shaw cell had a channel of length 22 cm with Teflon spacers, which both set the sidewall spacing at values between 1.0 and 2.0 cm and determined the gap between the cell plates. Gaps were studied in the range 0.05–0.16 cm. The glass plates of the cell were 1/4 in. thick and the calculated maximum flexion [3] was in no case greater than 0.5% of the gap. Polymers of molecular weights 17 400, 50 700, and 100 400 were studied at concentrations in the range 2–8 %. Flows were run at constant and controlled volumetric injection rate by injecting dyed water with a syringe pump [4] over a range from 0.5 ml/min to about 10 ml/min (±10%). Resulting patterns were observed with a charge coupled device videocamera and recorded on an enhanced S-VHS videocassette recorder. Subsequent digital analysis was performed with the assistance of the image-processing software package OPTIMAS [5]. In addition to analyzing the shapes of patterns, the recorded time series could be digitized to give tip positions at 1/30-s intervals, as the tip of the pattern progressed through the cell. From this time

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series, velocities averaged over 1/30-s intervals could be constructed (electronic shutter speeds of 1/2000 s were used to sharpen images and define temporal averaging intervals).

As in Ref. [1], the model associative polymers used consisted of a linear water-soluble poly(oxyethylene) backbone terminated by hydrophobic hexadecyl end groups [6,7]. Polymers of each molecular weight differed only in the length of the backbones. Once in aqueous solution, the hydrophobic end groups of different molecules would form an associating network. Published values of linear viscoelastic properties are available for these solutions at or near the concentrations we have studied and for all molecular weights [6].

In all cases it was possible to inject the water sufficiently slowly to produce a viscous fingering steady state. The width and the tip curvature of this steady state pattern always conformed to the relation expected of a Saffman-Taylor solution even though there is no reason to believe that this solution must survive the use of a viscoelastic material. Even though the fluids are miscible, such systems are known to exhibit an effective surface tension and if this is large enough it might be expected to make the finger width follow the Saffman-Taylor width as a function of inverse capillary number [8]. However, our fingers always have a width very close to half the channel, suggesting a sufficiently small dynamic surface tension that our effective inverse capillary number is always large enough to give this width. We would have to measure extremely low velocities to hope to see much wider fingers.

For the same molecular weights that were studied in the radial geometry (17 400 and 50 700), the transition from viscous fingering to a propagating crack tip appeared at a Deborah number quite consistent with the previous results. (While the channel-flow crack tip does not develop the rich array of sidebranches that were observed in the radial case for Deborah numbers far above threshold, we assume that this arises from side-wall effects and that the propagating crack tip observed in the present channel-flow case is closely related to the fracturelike patterns in the radial cases.) That is, taking $\tau_f = b^2 w/Q$, where w is half the width of the cell (i.e., the finger width in the fingering regime), instead of (1), the Deborah numbers for the transition can be brought into agreement by dividing the radial flow results by an arbitrary factor of 4.0 (which can be justified by the omission of scale factors in the definition of τ_f for both geometries). Figure 1 shows the range of Deborah number within which the fracturelike instability appears for several different combinations of polymer molecular weight, polymer concentration, cell-gap thickness, and (in the rectangular cell experiments only) channel width. Five of these cases involve flow in the channel geometry and four are radial flow realizations.

At injection rates above the threshold for fracturelike behavior, using the two lower molecular weight polymers, the pattern in the channel shows a very narrow, rather straight crack, which we have measured (comparing the rate of growth of the area with the known volumetric injection rate) to fill about one-sixth of the gap thickness occupied by the previous viscous finger. This can be noted in the lighter color (transparency) of this pattern. An example is shown in Fig. 2. Note that the fracturelike structure emerges cleanly from the Saffman-Taylor finger and that the tip velocity fluctuates about a small constant rms value in the Saffman-Taylor regime, exhibiting an abrupt increase followed by a gentler, almost linear increase in the rms value after the transition into the fracture regime. Fracturelike patterns typically progress at sufficiently high speeds that not many independent measurements of velocity are possible before the tip reaches the end of the cell. An example of measured

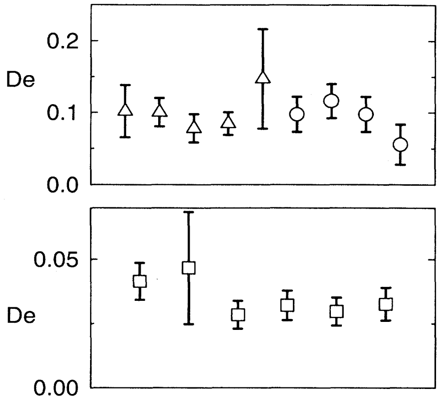

FIG. 1. Comparing ranges of Deborah number De for the onset of fracture for nine different fracturelike flows (top) and six different meandering flows (bottom). Top: 17 400 and 50 700 molecular weight polymers; △, experiments in the channel geometry; ○, experiments in the radial geometry. De as defined in the text, where both geometries are brought into agreement by the scale factor 4.0. Bottom: □, channel geometry results for the onset of meandering flow in solutions of the 100 400 molecular weight polymer.

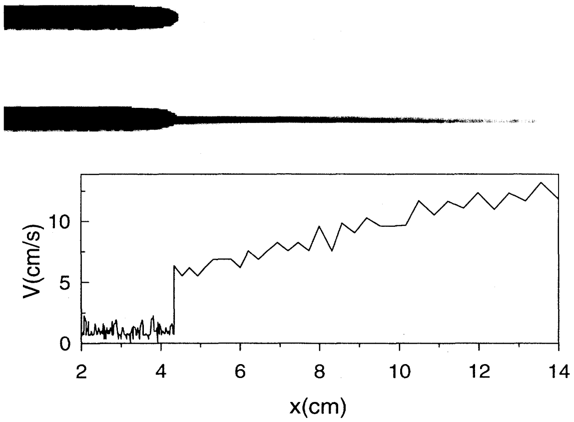

FIG. 2. Fracture using the polymer of molecular weight 17 400. Top: viscous finger at early times. The shape is that of a Saffman-Taylor finger. Middle: the fracturelike pattern emerges cleanly from the unperturbed finger. Note the lighter color of the fracture, indicating that it is much thinner across the cell gap than the parent finger. Bottom: tip velocity as a function of tip position during the growth.

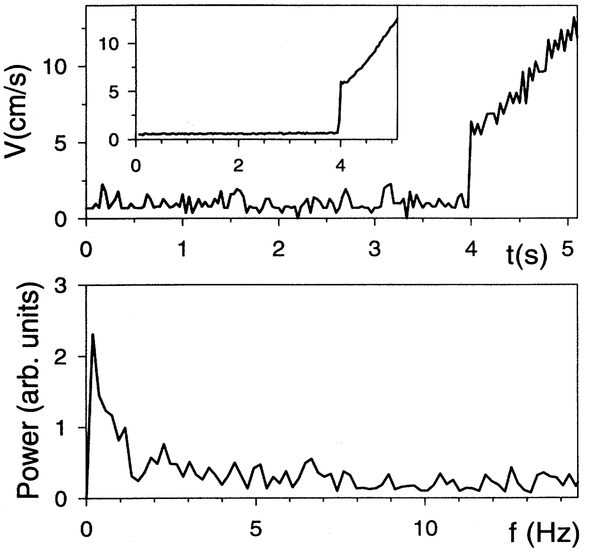

FIG. 3. Fracture using the polymer of molecular weight 17 400. Top: time evolution of the tip velocity for the same flow realization shown in Fig. 2. Note the sharp transition in tip velocity from the viscous fingering regime into the fracturelike regime. The inset shows the same velocity evolution after a low-pass filter is applied. Bottom: frequency spectrum for the same velocity distribution.

tip velocity vs time and a Fourier decomposition of the velocity vs time sequence for the same flow realization are shown in Fig. 3. While there are apparent fluctuations in velocity, they are small fluctuations superimposed on a dominant acceleration of the tip and their Fourier spectrum shows no trend that is reproducible (not surprising considering the severe limitations in the finite range of data). Note that the increase in the tip velocity following the transition is very sharp, less than 1/30-s of duration (our time resolution).

The highest molecular weight polymer shows behavior significantly different from the others. When molecular weight 100 400 is used, long before the injection rate reaches the Deborah number needed for the onset of fracturelike behavior (as found for the low molecular weights), the Saffman-Taylor finger becomes wrinkled and breaks up. Figure 1 shows also that the Deborah number scaling works successfully for the threshold of this instability, but the range of Deborah numbers involved are less than half those for the lower molecular weights, which translates into a lower threshold Q for the onset of this instability. After the Saffman-Taylor finger breaks up, the much narrower, much more ramified "finger" that results meanders down the cell, always filling much more (about three times more) of the gap thickness than in the fracturelike regime defined above and with a significant velocity component normal to the channel axis even though the dominant velocity component is that parallel to the channel axis. At high injection rates and at later times, this meandering phase can turn into a fracture pattern as defined above, but these higher molecular weight solutions always show at least a stage of meandering. It is difficult to define the exact point of onset of the fracturelike behavior in the presence of this meandering phase, but qualitatively one can say that there is no inconsistency in assuming that it sets in at the same Deborah number as expected from the lower molecular weights. That is, it is always possible to find a point in the evolution within the meandering regime where the tip velocity has reached a value which gives a Deborah number in agreement with the one expected for the fracture transition from the lower molecular weights. While the transition between the meandering regime and the fast fracture regime is not clear at all, it is never far from this point at which the Deborah number expected from the low molecular weight results is reached. Figure 4 shows an example of a flow pattern with a meandering phase, along with the measured velocity as a function of position in the cell. It is interesting to note that only a small region close to the advancing tip evolves with time. Sidebranches that appear during the flow quickly stop growing once the main tip leaves them behind. This is a clear departure from the fracturelike behavior in soft condensed matter reported in the literature [1,9], where sidebranches grow far behind the advancing tips at angles close to 90°. It may result from damping by channel walls or it may indicate that the meandering regime is dominated by a dynamics different from the fracturelike dynamics seen at lower molecular weights. Another possible mechanism for meandering is for stick-slip dynamics to carry the system back and forth between fracture and fingering with the individual fracture events (which give the directional changes) quenched too rapidly for the process to be clearly discernible within the time resolution

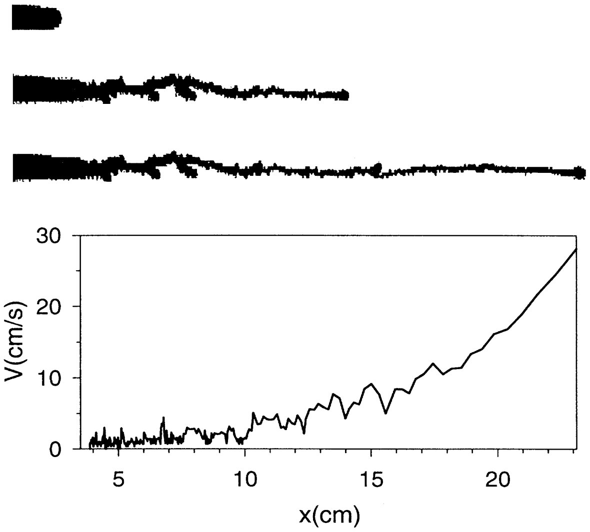

FIG. 4. Fracture using the polymer of molecular weight 100 400. Top: viscous finger at early times. The shape is that of a Saffman-Taylor finger. Second row: a meandering regime emerges from the unstable finger. Third row: the later stages correspond to a fast increase of velocity straight along the channel. Bottom: tip velocity as a function of tip position during the growth.

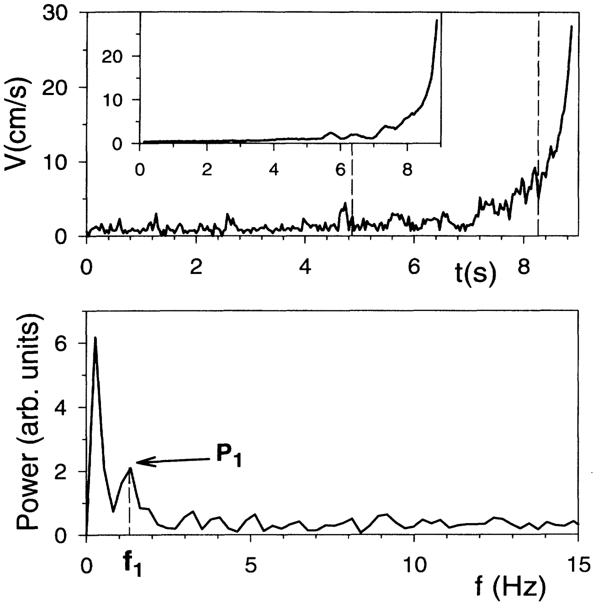

FIG. 5. Fracture using the polymer of molecular weight 100 400. Top: time evolution of tip velocity for the same flow realization shown in Fig. 4. Boundaries between the viscous finger regime, the meandering regime, and the fast fracture regime are marked. The inset shows the same velocity evolution after a low-pass filter is applied. Bottom: frequency spectrum of the last 4 s of the velocity evolution. A peak P_1 at frequency f_1 can be reproducibly observed. The peak extending over the first three frequency channels is an artifact of the analysis.

of the experiment.

Figure 5 shows the tip speed as a function of time and its Fourier transform for the same flow realization shown in Fig. 4. The first three frequency channels of the Fourier transform show a large peak, which is uninteresting because it is an artifact of the numerical windowing used to reduce the effect of the jump discontinuity in the time series. Beyond the third channel, we can see a smaller peak P_1 at frequency f_1 and then a succession of still smaller peaks at higher frequencies. Of all the meaningful peaks in the Fourier spectrum from a given flow realization, P_1 is always the largest and is the only reproducible peak. All these properties are true whether we Fourier analyze the tip speed or the component of the tip velocity in the channel direction.

The characteristic frequency f_1 can be detected in the time series of the meandering regime alone or in the combination of the meandering and the fast-fracturing regime. However, it cannot be distinguished in the fast-fracturing regime alone, possibly because of a lack of experimental resolution. Figure 6 shows f_1 as a function of Deborah number for flows at several different concentrations of the high molecular weight polymer and at different Hele-Shaw channel dimensions. The figure shows that, in the range of Deborah numbers attained, each polymer solution shows a linear dependence of the characteristic frequency on the Deborah number (or on the injection rate). We can also see that the geometric definition of the Deborah number successfully scales two experiments with the same polymer concentration (5.0%), but different channel dimensions. Similarly, data measured at concentrations of 5.0% and 6.4% are collapsed by the Deborah number scaling, even though their characteristic relaxation times [6] differ by a factor of 1.4. This collapse is not extended to the data for the 8.0% polymer concentration whose estimated relaxation time [6] is roughly twice the one for the 5.0% solution. The estimated relaxation times were determined by extrapolation from the available data [6] on shear rates for the onset of shear thinning at concentrations of 5% and less. The extrapolation certainly introduces extra uncertainty in the shear-thinning shear rates, which are themselves arbitrarily chosen from the measured shear-rate dependences of the solution viscosities.

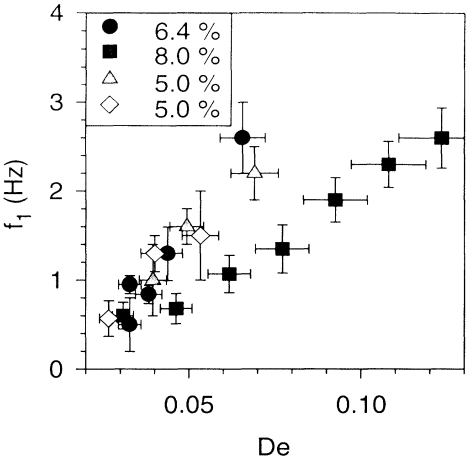

FIG. 6. Dependence of characteristic meandering frequency f_1 on Deborah number. The Deborah number has been constructed using the shear thinning shear rate as the polymer network characteristic time De= $Q/(wb^2\dot{\gamma}_0)$.

While the Deborah number defined above uses the shear-thinning shear rate to set the characteristic relaxation time of the polymer solution, it is of interest to consider an alternative characteristic shear rate ω_T, the frequency at which the storage and loss moduli of the polymer solution take on the same value. While this frequency is a very plausible candidate for representing the point at which fracturelike behavior might set in, our channel-geometry results agree well with the radial-geometry results of Ref. [1] in finding that the Deborah number is a much more reliable prediction of fracture onset if $\dot{\gamma}_0$ is used to represent the polymer relaxation rate. (The consistency of the thresholds shown in Fig. 1 is lost if ω_T is used to define De.) Figure 7 shows frequency f_1 as a function of a Deborah number De$'$, based on ω_T for the same data shown in Fig. 6. While the attempt to scale the data with this alternate Deborah number is not a total failure, it collapses the data less effectively than the analysis of Fig. 6.

It would clearly be of interest to learn whether the characteristic frequency we have detected in the pres-

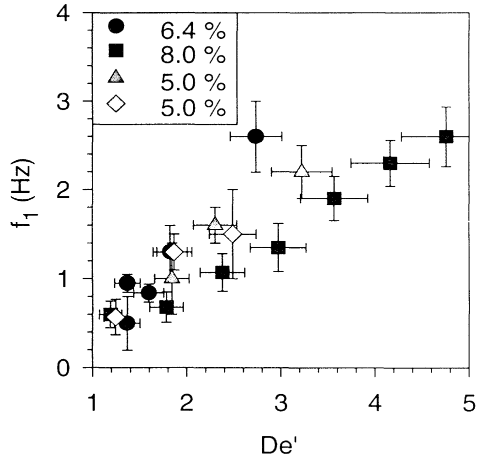

FIG. 7. Dependence of the characteristic meandering frequency f_1 on an alternative Deborah number [$\mathrm{De}' = Q/(wb^2\omega_T)$, discussed in the text] whose characteristic polymer relaxation time is defined by the excitation frequency at which the solution's storage and loss muduli have the same magnitude.

ence of noise in the meandering regime has an analog in the fracture regime. As mentioned above, no reproducible preferred frequency has been detected in the very noisy velocity-fluctuation spectra from the fracture regime. However, the spectra in the fracturelike regime are limited by our 1/30-s frame-to-frame time interval in the small number of observing frames measured during the rather short time for propagation of the crack tip through the cell.

The linear relation between the characteristic meandering frequency and the fluid injection rate (Fig. 6) is reminiscent of a very similar linear relation between the stick-slip frequency and the imposed crack velocity in the peeling of adhesive tape [10]. In the latter case, the frequency is attributed to a Hopf bifurcation, which can be at least qualitatively analyzed in terms of known stress-strain relations of the adhesive material. Stick-slip oscillatory behavior also disappears at higher tape peeling velocity and this is not inconsistent with our observations of fracturelike flows. Oscillatory solutions have also been predicted [11–13] as rather general features of quasistatic crack propagation, always in a restricted range of driving velocity reminiscent of the results of this experiment.

Kurtze and Hong [14] have explicitly considered fracturelike behavior in soft condensed matter and have predicted an oscillatory velocity for the advancing crack tip as long as the injection rate is such that the system evolution is dominated by stick-slip dynamics. They speculated that the frequency of these oscillations should increase with invading fluid injection rate. Our results are consistent with their expectations.

While the meandering regime shown by the highest molecular weight solutions has similarities to quasistatic crack-tip propagation discussed above, the characteristic behavior of the pattern evolution of lower-molecular-weight solutions (shown in Figs. 2 and 3) is very reminiscent of the brittle-solid fracture results of Fineberg *et al.* [2]. As in that work, we observe a sudden increase in velocity at the onset of fracturelike behavior, followed by small fluctuations superimposed on a rather constant value of $\Delta V/\Delta x$ at later stages of the crack propagation. Fineberg *et al.* have measured the jump of velocity at the onset to be continuous in Δx, with a mean value of $\Delta V/\Delta x$, which we have determined (from their published data) to be about 30 times higher than $\Delta V/\Delta x$ in the subsequent regime. The jump we observe occurs in one time step. If we assume that our observed jumps occur linearly thoughout the one averaging interval Δx, we estimate, for the example shown in Fig. 2, that $\Delta V/\Delta x$ at the onset of fracture is of the order of 30 times that of $\Delta V/\Delta x$ in the subsequent regime. This is consistent with the results of Fineberg *et al.* and we cannot compare time scales directly since we cannot confidently define a dimensionless time. However, Fineberg *et al.* report a third crack propagation stage characterized by a still lower value of $\Delta V/\Delta x$, which we have not observed.

In summary, we have measured crack-tip velocities for the injection of water into end-capped associating polymer solutions in rectangular Hele-Shaw cells. The condition for the the onset of fracturelike behavior can be defined for all observed values of the cell-gap, the polymer molecular weight, and the polymer concentration by a Deborah number which uses the shear-thinning shear rate as a characteristic polymer-solution relaxation frequency. For a low molecular weight polymer, the onset of fracturelike pattern evolution is accompained by an abrupt jump in the tip velocity, followed by a constant and rather low acceleration. At high molecular weight, the transition to fracturelike behavior is blurred by a regime in which the crack tip meanders from side to side and fluctuates in both overall speed and channel velocity with a characteristic frequency which depends linearly on fluid injection rate. It is plausible that stick-slip dynamics dominates this meandering regime which, at later times, would lead to a fracture regime. The abrupt transition observed in this experiment between viscous fingering and a fast fracture at the lower molecular weights might be expected to reveal a region were stick-slip dynamics dominates the evolution if observations could be made at shorter length scales and smaller time scales.

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