

Tip-splitting instabilities in the channel Saffman-Taylor flow of constant viscosity elastic fluids

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Boger fluids are used to study viscous fingering growth in viscoelastic fluids in channel Hele-Shaw flow. We have found that the viscous finger growing in the Boger fluid is unstable to tip splitting at high velocities, in a regime where a Newtonian viscous finger is stable. No fracturelike instabilities were observed. We show that the viscoelastic normal stress differences arising in shear and extensional flow reach very high values at shear and extensional rates comparable to those achieved at the tip of the finger at the onset of tip splitting, and the fluid becomes highly anisotropic. The viscoelastic stress could affect the dynamics of the finger and induce the tip-splitting instability.

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I. INTRODUCTION

The viscous fingering (VF) problem [1], which arises when a less viscous fluid is forced into a more viscous fluid, has been the object of extensive studies during the past forty years, due to its application to the study of many industrial processes, e.g., crude oil recovery and flow through porous media. The highly nonlinear Saffman-Taylor problem, in which the two fluids are flowing in a channel formed by parallel plates separated by a small gap (Hele-Shaw cell) is now well understood [1]. More recently, a large number of studies have focused on the Saffman-Taylor flow in non-Newtonian fluids [2–5]. These studies are only at their beginnings, despite their direct involvement in important industrial processes [6] such as air-assisted injection molding, production of hollow fiber membranes, and coating of ceramic monoliths for the manufacture of catalytic converters.

Experiments in homopolymer [2] and associating polymer solutions [3], clay dispersions [4,5] and foams [7] have all shown rich classes of patterns. Some non-Newtonian fluids (associating polymer solutions, clay dispersions) show a dramatic fracturelike behavior in VF experiments. Experimentally, the onset of fracture has been determined to depend on a characteristic (Deborah) number, constructed by dividing a characteristic time of the non-Newtonian fluid by the time of the flow [3,5]. Unfortunately, the complexity of the rheology of the real fluids makes the viscous fingering problem in such fluids extremely difficult to analyze. Real fluids can simultaneously display many non-Newtonian properties, such as shear thinning or thickening, viscoelasticity, yield stress, etc. For example, viscoelastic fluids are usually shear thinning. This adds complexity to the problem and creates difficulties in understanding the impact of the rheological properties of the fluid on the dynamics of the viscous fingers. In a recent study [8], in which we simultaneously measured the stress and the shear rate in fracture experiments in associating polymer solutions, we inferred that the fracturelike instabilities are probably determined by the extreme shear thinning of these solutions.

Recent theoretical and experimental papers discuss the effect of viscoelasticity on viscous fingering. The Saffman-Taylor problem for an Oldroyd-B fluid was discussed in Ref. [9] and it was found that the growth rate of interface pertur-

bations is enhanced by viscoelasticity. Other studies [10,6,11] have focused on the cross-sectional area of an air bubble growing into tubes filled with viscoelastic fluids.

By engineering special non-Newtonian fluids, it is possible to separate the viscoelastic effects from the other non-Newtonian effects, especially those due to shear thinning. A common method is to use constant viscosity elastic fluids (also known as Boger fluids) [12], which are obtained by dissolving small amounts of high-molecular-weight polymers in viscous Newtonian solvents. Typically, in Boger fluids the Newtonian solvent contribution to the viscosity is larger than 90%, thus ensuring a nearly constant viscosity for the fluid. The high-molecular-weight polymer chains stretch in shear and extensional flow, making the fluid elastic, but they do not significantly change the viscosity of the base Newtonian fluid.

Allen and Boger performed some preliminary measurements on viscous fingering in radial geometry when Newtonian fluids displaced Boger fluids [13]. Their patterns did not depart qualitatively from those seen in viscous fingering in Newtonian fluids, but they cautioned that they had not tested an adequate range in Deborah numbers. In this paper we report the results of experiments on such model fluids wherein we have formed viscous fingers in the fluids in linear Hele-Shaw cells and measured the effects of isolated non-Newtonian properties.

II. EXPERIMENTS

A. Characterization of the fluids

As a first step in the preparation of the Boger fluids, stock solutions were prepared by dissolving small amounts of high molecular weight polyisobutylene (PIB) of molecular weight 1.2×10^6 (Exxon Vistanex L-120) in kerosene. Then the Boger fluids were obtained by adding amounts of stock solution to low molecular weight (MW=600) polybutene (PB). A description of the rheological properties of the PIB/PB Boger fluids used in this study is presented in Ref. [14].

The strength of viscoelasticity is tuned by the amount of PIB added, which is usually in the ppm range. We prepared two Boger fluids (B1 and B2) and one control, Newtonian fluid (B0). The same amount of PB and kerosene was used in the preparation of all three fluids. The concentration of PIB

TABLE I. Chemical composition of the fluids.

polyisobutylene Solution	polybutene ppm	kerosene (%)	(%)
B1	514	92.18	7.77
B2	257	92.18	7.79
B0	0	92.18	7.82

in B1 is 514 ppm (same concentration as in the fluid studied in [14]) and 257 ppm in B2, one-half of the PIB concentration in B1. The control fluid B0 contains only PB and kerosene. A description of the composition of the fluids is presented in Table I.

The three fluids have similar viscosities and surface tensions. The viscosities, measured with a Brookfield Rheometer, and the surface tensions, measured with a CSC-DuNouy Tensiometer, are shown in Table II.

The rheological properties of fluid B1 were reported in Ref. [14]. Viscosity measurements at various shear rates indicate the absence of a significant shear thinning, variations of viscosity being confined to 9% of the value of viscosity at low shear rate. The solution exhibits strong viscoelastic behavior. The first normal stress difference (N_1) is found to be proportional to the second power of the shear rate over a reasonably large range of $\dot{\gamma}$, $N_1 = 0.137\dot{\gamma}^{2.013}$ [14].

The relaxation time of the fluid λ [15] provides a measure for the strength of the viscoelastic behavior of the fluid. It can be calculated from N_1

$$\lambda = \frac{N_1}{2\eta\dot{\gamma}^2}. \quad (1)$$

For liquid B1 the relaxation time, calculated from the measurements of N_1 reported in Ref. [14], is 0.055 sec. The relaxation time for the fluid B2 can be extrapolated from the B1 relaxation time. When the intrinsic elasticity of the solvent can be neglected, and for dilute PIB solutions with a concentration smaller than the critical concentration $c^* = 0.11\%$ [16], λ is proportional with the PIB concentration. By using this proportionality, we obtain a relaxation time for B2 of 0.028 sec.

Reference [14] also reports rheological measurements for the PB/kerosene fluid B0. The measured relaxation time was of the order of 10^{-4} sec, two orders of magnitude smaller than the relaxation time for B1. Due to the smallness of its relaxation time, for all practical purposes we can neglect the viscoelasticity of the fluid B0, and we can consider B0 to be Newtonian. The values for the relaxation time for the fluids are included in Table II.

TABLE II. Properties of the fluids: viscosity, relaxation time, and surface tension.

Solution	η (Poise)	λ (sec)	σ (dyne/cm)
B1	9.3+/-0.7	0.055	27+/-3
B2	9.4+/-0.4	0.028	30+/-3
B0	9.0+/-0.4	$\sim 10^{-4}$	30+/-3

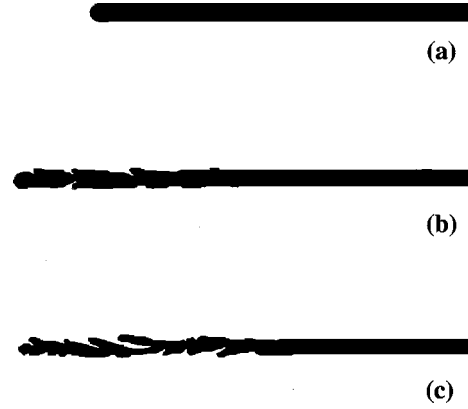


FIG. 1. Viscous fingering patterns obtained for Boger fluid B1. The width of the channel was $w = 1$ cm. (a) The gap between the glass plates of the Hele-Shaw channel $b = 1.5$ mm and the injection pressure $P = 4$ PSI. (b) $b = 0.75$ mm and $P = 8$ PSI, (c) $b = 0.38$ mm and $P = 12$ PSI.

B. Experimental procedure

The Hele-Shaw channel used in our experiments had a length $L = 22.5$ cm and width $w = 1$ cm. The gap between the plates b was varied between 0.22 to 1.5 mm. Air at constant pressure was injected into the channel previously filled with one of the fluids described above. The growth of the air finger was recorded on videotape. An image-processing software package allowed us to extract the position of the tip of the finger. The velocity of the finger v_f was constructed from the time series of the finger position. The pressure of the air injected into the channel was also measured with a pressure transducer.

C. Observations and results

In experiments with the Boger fluids, at low injection pressure we observe smooth viscous fingers growing into the channel. The fingers are similar to the fingers normally observed in Newtonian viscous fluids. But as the injection pressure is increased, the tip of the viscous finger starts splitting. The experiments being conducted at constant pressure, the pressure gradient increases when the finger moves closer to the open end on the channel, and the finger is accelerating. The tip-splitting instability appears when the velocity of the tip of the finger increases above some threshold velocity v_f^0 . In the range of pressure and b values we studied for the Boger fluids we did not observe any other types of instabilities, e.g., fracture [3].

Three sample patterns for fluid B1 are shown in Fig. 1. In Fig. 1(a) the gap between the plates is $b = 1.5$ mm and the injection pressure is $P = 4$ PSI. The pattern is a smooth viscous finger. Figure 1(b) corresponds to a gap of 0.75 mm and a larger injection pressure of 8 PSI. When the finger starts growing its velocity is small, and the finger grows smoothly. The first tip splitting is observed when the finger tip velocity reaches 5.7 cm/sec. In the experiment presented in Fig. 1(c) the gap between the plates is reduced to 0.38 mm and the pressure is increased to 12 PSI. The tip-splitting instability starts developing earlier, at a threshold velocity of 2.4 cm/sec.

TABLE III. Finger tip velocity at the onset of the tip-splitting instability.

b (cm)	v_f^0 (cm/sec) for B1	v_f^0 (cm/sec) for B2
0.022	0.79 ± 0.24	1.31 ± 0.14
0.038	2.19 ± 0.54	2.96 ± 0.38
0.049	3.19 ± 0.21	4.5 ± 0.42
0.06	4.41 ± 0.39	6.63 ± 1.07
0.075	5.47 ± 0.48	10.4 ± 1.7
0.086	7.41 ± 0.66	14.9 ± 0.6
0.113	14.5 ± 2.4	26.3 ± 2.6
0.15	22.1 ± 3.2	not measured

The Newtonian control fluid B0 shows no tip-splitting fingers, even at very high injection pressure and values of the finger tip velocities. For a gap between the plates of 0.38 mm we reached values of the tip velocity of up to 24.3 cm/sec, about 10 times larger than the threshold for tip splitting for B1, and still we did not observe a tip-splitting finger.

For each value of b and for each Boger fluid we performed between 5 and 7 experiments, each experiment corresponding to a different injection pressure. The value of the threshold velocity for tip splitting, v_f^0 was found to be dependent on b and on the fluid used, see Table III, but independent on the injection pressure. When the injection pressure is smaller tip splitting starts closer to the open end of the cell than it starts when the pressure is larger. This can be understood by realizing that the dynamics of the viscous finger is determined by the magnitude of the pressure gradient, instead of the pressure. The finger tip velocity is related to the pressure gradient through relations similar to Darcy's law, which can be rather complicated for non-Newtonian fluids [17,8].

The experimental data from Table III show that v_f^0 increases with the aspect ratio of the cell, b/W and that v_f^0 is larger for the fluid with the smaller relaxation time (B2). A plot for v_f^0 vs b/W for the two Boger fluids is shown in Fig. 2. In the range of b/W we studied, the data can be fitted reasonably well with a power law. The result of the fit is

$$v_f^0 = 537(b/W)^{1.7} \quad (2)$$

for fluid B1, and

$$v_f^0 = 1285(b/W)^{1.84} \quad (3)$$

for fluid B2. The exponents of the power-law fit are similar for the two Boger fluids, but the prefactors are significantly different.

D. Viscoelasticity and tip splitting

A generally used viscoelasticity measure, which characterizes the ratio of elastic to viscous forces for the fluid, is the Weissenberg (or Deborah) number Wi . Wi can be defined as

$$Wi = \lambda \dot{\gamma}. \quad (4)$$

Elastic effects are small at low Wi , but become observable when Wi is of the order of 1 and larger.

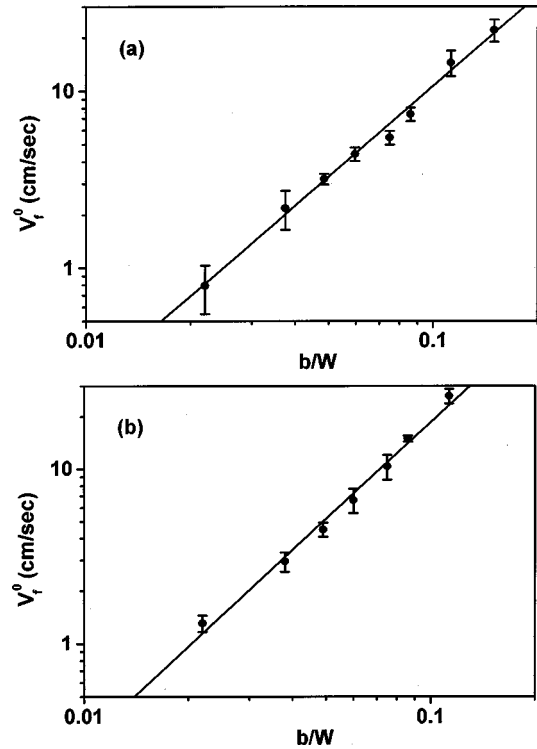


FIG. 2. Finger tip velocity at the threshold of the tip-splitting instabilities vs the aspect ratio of the cell for (a) fluid B1 and (b) fluid B2.

In order to estimate the magnitude of Wi , and therefore the strength of the viscoelastic effects, we need an estimate for the shear rate. The shear rate in the neighborhood of the tip of the finger can have a very complex spatial dependence, due to the complexity of the flow near the air-liquid boundary. However, a measure for the magnitude of the shear rate can be determined by dividing the tip velocity by the smallest length scale which constrains the flow. The fluid elements in front of the tip of the air finger, equally distanced from the upper and lower glass plates, move with a velocity equal to the finger velocity. Assuming no-slip boundary conditions at the glass plates, the velocity near the glass plates should be zero. Therefore, a measure for the shear rate in the liquid, in front of the moving air finger is

$$\dot{\gamma}_f = v_f / b' \quad (5)$$

with $b' = b/2$.

From Eqs. (4) and (5), Wi for the fluid in the gap in front of the moving finger can then be written as

$$Wi_f = \lambda v_f / b'. \quad (6)$$

The Weissenberg numbers at the onset of tip splitting Wi_f^0 can be estimated by using Eq. (6) in conjunction with the measured values for the onset finger tip velocities, v_f^0 from Table III. The results are presented in Table IV. The dependence of Wi_f^0 on the aspect ratio of the cell is

$$Wi_f^0 = 59(b/W)^{0.7} \quad (7)$$

for fluid B1 and

TABLE IV. Weissenberg number at the onset of tip splitting.

b (cm)	Wi_f^0 for B1	Wi_f^0 for B2
0.022	3.96 ± 1.22	3.28 ± 0.35
0.038	6.41 ± 1.58	4.28 ± 0.54
0.049	7.23 ± 0.49	5.05 ± 0.47
0.06	8.15 ± 0.73	6.08 ± 0.98
0.075	8.02 ± 0.70	7.60 ± 1.23
0.086	9.48 ± 0.84	9.55 ± 0.38
0.113	14.2 ± 2.3	12.8 ± 1.2
0.15	16.2 ± 2.3	not measured

$$Wi_f^0 = 71(b/W)^{0.84} \quad (8)$$

for fluid B2.

The threshold Weissenberg numbers are similar for the two Boger fluids, suggesting that the onset of tip-splitting is determined by Wi_f^0 . This result, in conjunction with the observation that the instability is absent for the Newtonian control fluid in the range of shear rates (injection pressures) studied, provides a strong suggestion that tip splitting has a viscoelastic origin.

A common regression for both B1 and B2 has the following result:

$$Wi_f^0 = 66(b/W)^{0.78}. \quad (9)$$

The complexity of the viscous fingering problem in non-Newtonian fluids prevents us from providing a rigorous explanation of how viscoelasticity induces tip splitting. A naive explanation involves the viscoelastic normal stresses arising in shear and extensional flow in front of the moving finger. It is well known that the stress tensor in a viscoelastic fluid undergoing a shear or extensional flow is no longer isotropic (see, e.g., Ref. [18]), as it is in the flow of a Newtonian fluid, where the diagonal elements of the stress tensor are all the same and can be identified with the pressure. The magnitude of the anisotropy is captured through the first and the second normal stress difference.

As the finger advances into the channel, its tip shears and stretches the fluid while splitting it. The viscoelastic normal stresses, which oppose the finger growth, should be larger in front of the tip of the finger, where the shearing and stretching is the largest. The finger can then be viewed as avoiding the local stiff region in front of its tip by tip splitting and growing around that region.

E. Estimate of the magnitude of the normal stress difference at the onset of tip splitting

The Boger fluids are reasonably well described by the Oldroyd-B model [15], which is a quasilinear combination of Newtonian (for solvent) and upper convected Maxwell (for polymer) constitutive equations. The stress tensor for the fluid is the sum of the stress contributions from the Newtonian solvent (s) and from the elastic polymer (p)

$$\tau = \tau_s + \tau_p. \quad (10)$$

1. Shear flow

In the Oldroyd-B model, in simple shear flow the viscosity of the fluid is the sum of the viscosities of the solvent and of the polymer

$$\eta = \eta_s + \eta_p. \quad (11)$$

For a fluid element flowing in the x direction (direction of the finger growth in our experiments), and for a velocity gradient in the z direction (direction perpendicular to the glass plates in the Hele-Shaw cell), the first normal stress difference from Eq. (1) is

$$N_1 \equiv \tau_{xx} - \tau_{zz} = 2\eta\lambda \dot{\gamma}^2. \quad (12)$$

In the Oldroyd-B model the second normal stress difference is zero.

Due to its proportionality to the second power of the shear rate, at large shear rate N_1 can take very high values. If we compare N_1 with the shear stress, τ_{xz} we obtain

$$N_1 / \tau_{xz} = 2\lambda \dot{\gamma} = 2Wi. \quad (13)$$

As a result, in the Oldroyd-B model the magnitude of N_1 relative to τ_{xz} increases with the Weissenberg number, and therefore with the finger tip velocity. Based on our experimental determinations for the Weissenberg numbers, N_1 can reach values up to 30 times larger than the shear stress at the threshold of the tip-splitting instabilities.

2. Extensional flow

The normal stress difference arising in extensional flow could be even larger than the estimate of N_1 in shear flow. As we mentioned above, the advancing finger stretches and splits the fluid elements near its tip. The main stretching is in the y direction, perpendicular to the axis of the channel and parallel to the glass plates.

The resistance of the viscoelastic fluid to the extensional motion is measured by the normal stress difference

$$\tau_{yy} - \tau_{xx} = \dot{\epsilon} \bar{\eta}, \quad (14)$$

where $\dot{\epsilon}$ is the extensional rate, $\dot{\epsilon} = \partial v_y / \partial y$, and $\bar{\eta}$ is the extensional viscosity. For polymer solutions the extensional viscosity can have very large values, much larger than the shear viscosity η . By definition, the Trouton ratio is

$$\text{Tr} = \bar{\eta} / \eta. \quad (15)$$

Tr values of the order of 10 000 are not unusual for polymer solutions.

In order to estimate the magnitude of the normal stresses generated in extensional flow in viscous fingering experiments, we need an estimate of the extensional rate and of the Trouton ratio. An estimate for the extensional rate due to the lateral stretching motion of the polymer fluid in front of the tip of the finger is

$$\dot{\epsilon} = v_f / w_f, \quad (16)$$

where w_f is the width of the finger, which is approximately equal to one half the width of the channel. In the conditions

of our experimental studies for the two Boger fluids, the magnitude of $\dot{\epsilon}$ at the onset of tip splitting varies between 1 and 20.

The Trouton ratio for PIB-based Boger fluids was measured experimentally [19] and it was investigated theoretically via Brownian dynamic simulations of flexible bead-rod chains [20]. One of the Boger fluids investigated in Ref. [19] was a solution of 0.31% PIB of molecular weight 1.2×10^6 (same PIB as in our fluids) in a PB-tetradecane Newtonian solvent, and it had a Trouton ratio of the order of 3200 to 3500 for strain rates between 2.1 and 4 s^{-1} . The other fluid was a solution of 0.185% PIB of MW = 2.4×10^6 in a PB/kerosene base fluid. Its measured Trouton ratio was 2000 to 2200 for similar strain rates. The simulations from Ref. [20] compare favorably to the experimental data for these fluids. Although the fluids studied experimentally are not identical to our Boger fluids, they should have comparable properties. The strain rates achieved at tip-splitting in our experiments are also comparable to those in the study from Ref. [19]. We estimate that the Trouton ratio for fluids B1 and B2 should be of the order of few thousands at the onset of tip splitting. With these estimates we can compare the extensional normal stress difference with the shear stress at the tip of the finger

$$(\tau_{yy} - \tau_{xx})/\tau_{xz} \approx \frac{b'}{w_f} \text{Tr}, \quad (17)$$

which should be of the order of few hundreds at the onset of tip splitting. These estimates show that in conditions of shear and extension similar to those achieved at the threshold of tip splitting in our experimental studies the material becomes a highly anisotropic liquid, and the normal stress differences can have very high values, orders of magnitude larger than the shear stress.

III. CONCLUSIONS

We have investigated the effect of viscoelasticity on the dynamics of the viscous fingers in the channel Hele-Shaw flow. In order to distinguish the effects due to viscoelasticity from those produced by other non-Newtonian properties of real liquids, we have used two constant viscosity elastic (Boger) fluids, which are dilute solutions of a rubber polymer (polyisobutylene) in a Newtonian viscous solvent. The strength of the viscoelastic response was adjusted by changing the concentration of the elastic polymer. A control Newtonian fluid, of properties similar to those of the Boger fluids, except for the negligible viscoelasticity, was used for comparison reasons.

When the finger velocity exceeded some threshold value,

a tip-splitting instability was observed for the Boger fluids. The threshold was dependent on the aspect ratio of the cell and it is smaller for the fluid with the larger relaxation time. The control Newtonian fluid showed no tip splitting in conditions similar to those in which the experiments with the Boger fluids were performed, not even at finger tip velocities much higher than the threshold velocities for tip splitting for the elastic fluids. This strongly suggests that the observed tip splitting in Boger fluids has a viscoelastic origin. In addition, the onset for viscoelastic tip splitting was determined to occur at similar Weissenberg numbers for the two Boger fluids.

Tip-splitting viscous fingers were observed in previous experiments with non-Newtonian fluids [21] and non-Newtonian colloid suspensions of clay particles in water [22]. The origin of the instability in Newtonian fluids [21] is determined by the small nonuniformities of the cell. In the experiments in Refs. [21,22] tip splitting occurred at values of $1/B$ of the order of $10^3 - 10^4$, but should be dependent on the uniformity of the cell. For our experiments in our cell with the fluid B1, the value of $1/B$ at the onset of tip splitting was between 4000 and 6400, while the threshold for fluid B2 was 7000 to 10000, and the viscous finger in the Newtonian control fluid B0 remained stable even at $1/B$ exceeding 50000, reinforcing our conclusion that our observed tip splitting in Boger fluids is due to viscoelasticity, and not to cell inhomogeneities.

The Boger fluids showed no fracture, in agreement with the inference [8] that fracturelike instabilities observed in associating polymer solutions [3,8] are determined by extreme shear thinning, and not by viscoelasticity. An analysis of the normal stress differences arising in viscoelastic fluids in shear and extensional flow show that these stresses can become very large at Weissenberg numbers similar to those involved near the tip of the finger. At large shear and/or extension rate the polymer chains are being stretched and the fluid becomes a highly anisotropic fluid, with the highest stresses developing near the tip of the finger, where the local shear and extension is the largest. The strong elastic forces associated with the normal stress components (which oppose the finger growth) provide a plausible source of the observed growth on either side of the finger tip, resulting in tip splitting.

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