Increase in Regularity by Polymer Addition during Chaotic Mixing in Two-Dimensional Flows

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Experiments show that the addition of small amounts of polymer to a Newtonian fluid undergoing chaotic advection decreases the amount of chaos; regular or unmixed regions increase in size and squeeze out the regions of chaotic behavior while preserving underlying symmetries. The size of the islands increases with increasing elasticity and rate of stirring. Large effects are present at relatively low Deborah numbers (relaxation time of the fluid to the time scale of the flow).

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Since the first studies of chaotic advection in two-dimensional time-periodic flows,[1] all experimental studies of chaotic mixing to date[2-4] have been carried out using Newtonian fluids in low Reynolds number flows. The purpose of this Letter is to report results using a series of viscoelastic fluids with different relaxation times. Such a problem is more complex than the Newtonian counterpart—mostly due to the presence of an additional time constant inherent to the fluid itself—and it is relevant to processing of polymers[5] and to problems in geophysics.[6,7] Experiments with passive tracers conducted in carefully controlled two-dimensional time-periodic flows—namely the *journal bearing flow*[4,8] and the *cavity flow*[9,10]—reveal a morphological structure consisting of poorly mixed (or regular) islands, and well-mixed (or chaotic) regions. The separation between nearby particles increases in the regular regions increasing linearly in time. On the other hand, in chaotic regions, the separation increases exponentially in time and a passive tracer follows closely the regions traced out by the unstable manifolds.[8] Where manifolds do not invade, islands form. However, there is order amidst the chaos: For example, if the instantaneous streamline portraits are symmetric, it is possible to show that the placement of the islands in the flow region becomes symmetric at periodic intervals of time.[11]

How is this picture changed if the fluid is elastic? Here we present results obtained in a cavity flow apparatus; similar experiments in other flow fields seem to indicate that the phenomenon is largely independent of the flow field itself. The flow region is rectangular with width W (10.35 cm) and height H (6.2 cm) (for details of this apparatus, see Ref. 10). Two types of time-periodic wall motions and four different fluids were studied. The motions are either discontinuous or continuous and the fluids consist of a Newtonian fluid (glycerine, denoted N, viscosity $\mu = 7.5$ P, density $\rho = 1.26$ g/cm^3 at 25 °C), and three viscoelastic solutions made of polyacrylamide (PAA, Scientific Polymer Products) dissolved in glycerine. The concentrations are $C_1 = 60$ ppm, $C_2 = 125$ ppm, and $C_3 = 160$ ppm. (These solutions are an example of the so-called "Boger fluids,"[12] i.e., fluids exhibiting viscoelastic behavior and having a constant shear viscosity.) The rheological properties of the fluids are shown in Fig. 1; the largest relaxation time, τ, was estimated by means of the apparent extensional viscosity.[13] The two time-periodic flows studied are corotational; i.e., the top wall and bottom walls of the cavity move in opposite directions; the two other walls are stationary. In the *discontinuous* flow the top wall moves first, for a time $\frac{1}{2}T$ and with speed U, and then the bottom wall moves second, for a time $\frac{1}{2}T$ and with speed U. In the

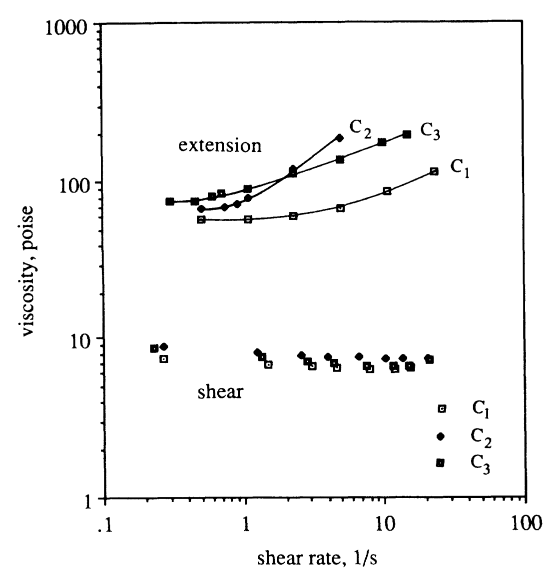

FIG. 1. The rheological properties of the viscoelastic fluids were measured using steady shear and extensional flows at 25 °C. The steady shear tests were conducted in a parallel-plate configuration and indicate that the fluids have approximately a constant shear viscosity; the extensional tests were conducted using a method of opposing jets (the theoretical ratio between the two viscosities at zero shear rate should be exactly 4). The relaxation times are obtained from the Newtonian plateau: $\tau_1 = 0.6$ s for C_1, $\tau_2 = 1.4$ s for C_2, and $\tau_3 = 2.8$ s for C_3.

experiments reported here the two walls move at the same speed, 1.9 cm/s, and there is a 5-s pause between each $\frac{1}{2}T$ cycle. In the *continuous* (sinusoidal flow) the velocity varies according to

$$v_{\rm top}=U\sin^2(\pi t/T+\pi/2),\quad v_{\rm bot}=-U\sin^2(\pi t/T)\,, \tag{1}$$

where $v_{\rm top}$ and $v_{\rm bot}$ are the top and bottom wall velocities, respectively, and U is 2.69 cm/s. In both flows we regard the period T as the governing parameter or, equivalently, the dimensionless wall displacement, $D=(|d_{\rm top}|+|d_{\rm bot}|)/W$, where

$$d_{\rm top}=\int_0^T v_{\rm top}(t)dt,\quad d_{\rm bot}=\int_0^T v_{\rm bot}(t)dt\,, \tag{2}$$

and W is the cavity width. The magnitude of inertial effects is given by the Reynolds number ($\mathrm{Re}=\rho UH^2/\mu W$) and the Strouhal number ($\mathrm{Sr}=TH^2/UW$). The range of Re is 0.5–1.7 and the range of Sr is 0.10–0.20; therefore the inertial effects are small (for experimental verification, see Ref. 12). The relative importance of the viscoelastic effects is quantified in terms of the Deborah number: the ratio of the relaxation time of the fluid τ to the time scale of the flow.[14] In time-periodic flows it is possible to select two different times scales for the flow; these give rise to two different Deborah numbers: One possibility (De′) is to use the inverse of the shear rate (H/U), another (De″) is to use the time period T. However, if the experiments are carried out at constant wall displacement D, both numbers are proportional to each other. The Deborah numbers can be changed by either varying the relaxation time of the fluid τ or by changing the rate of stirring, i.e., U or T. Several variations come to mind and only a limited range of possibilities is explored here.

The main observations are as follows. The steady streamlines, Fig. 2, of all four fluids are nearly indistinguishable and are symmetric with respect to the vertical axis. The dye patterns, which reveal large-scale regions of chaos, indicate island structures with vertical symmetry as well (at the precise moment when the photograph is taken; for an example involving Newtonian fluids, see Ref. 15). Figure 3 illustrates the effects of varying De′ by increasing the relaxation time. The flow corresponds to the sinusoidal flow with $D=5.19$. The macroscopic structures (folds) of the dye pattern in the four systems

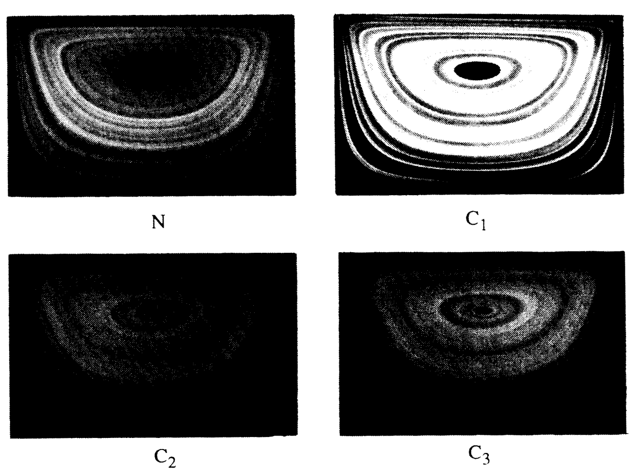

FIG. 2. Steady-state streamlines corresponding to the Newtonian fluid (N) and the three viscoelastic fluids of Fig. 1; in all cases the Reynolds number is 1.2.

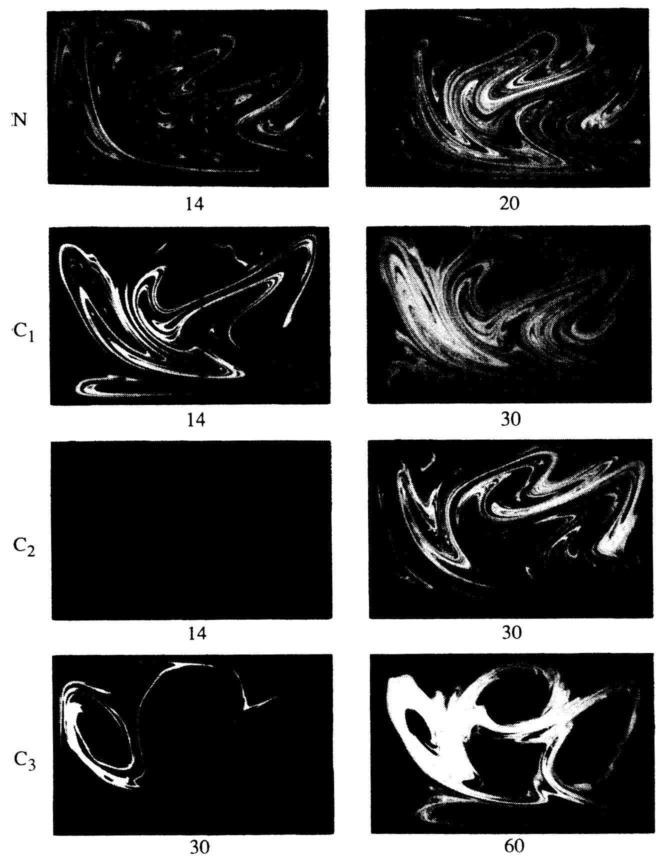

FIG. 3. To study the mixing process, we follow the deformation of a passive blob of tracer injected in chaotic regions of the flow. The tracer is a fluorescent dye dissolved in the bulk fluid and diffusion effects are negligible during the time scale of the experiment. The flow field corresponds to Eq. (1) with $D=5.19$. The pictures are taken when the top wall and bottom wall are moving at maximum and minimum (zero) speeds, respectively. The first row corresponds to the Newtonian fluid (N), the second row to C_1, the third row to C_2, and the fourth row to C_3. The numbers represent the number of periods. The best mixing corresponds to the Newtonian system, and islands grow with increasing PAA concentration and decrease with increasing number of periods (the photographs in the right column will not change appreciably if the number of periods is increased). Note the similarity of macroscopic structures (large-scale folds) in all experiments. The island structure corresponding to sixty periods ($\mathrm{De}'=0.85$) is symmetric with respect to the vertical axis; traces of symmetry are clear in the other experiments as well.

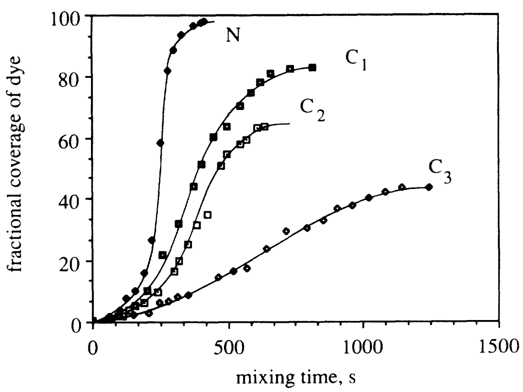

FIG. 4. Time evolution of the systems of Fig. 2 in terms of area coverage measured by image analysis. The Newtonian system approaches complete area coverage while the asymptotic coverage in the viscoelastic systems decreases with increasing PAA concentration. For coverages up to 0.5 the area grows as $A = A_0\exp(\sigma t)$, where $\sigma_N = 0.018\ \mathrm{s}^{-1}$, $\sigma_{C_1} = 0.013\ \mathrm{s}^{-1}$, $\sigma_{C_2} = 0.011\ \mathrm{s}^{-1}$, and $\sigma_{C_3} = 0.004\ \mathrm{s}^{-1}$.

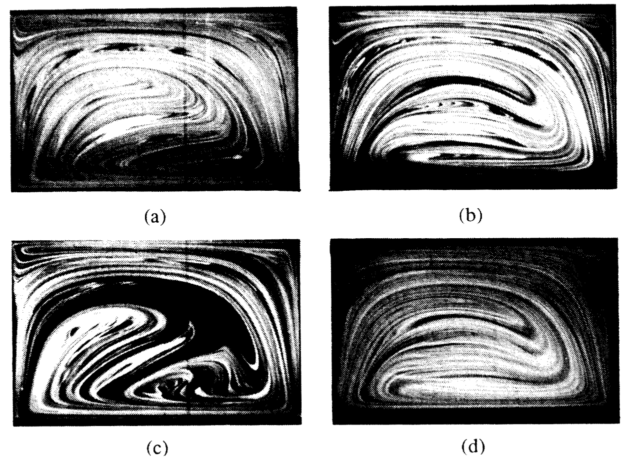

FIG. 5. Similar to Fig. 2 except that the mixing is produced by the discontinuous flow with $D = 12.85$ for eight periods (Re = 1.2). The experiments are conducted with two different fluids: (a) a Newtonian fluid and (b)-(d) the PAA solution. (b) and (d) correspond to a wall velocity $U' = 0.5$ cm/s, while (a) and (c) correspond to $U'' = 3.0$ cm/s. (d) shows what happens when (c) is stirred for two additional periods at speed U'. At low shear rates the viscoelastic system (b) behaves like the Newtonian system, but at high shear rate an unmixed region appears (c). The unmixed region disappears when the system is mixed further at a lower shear rate (d).

are strikingly similar; however, the islands grow with increasing relaxation time and squeeze out the regions of chaos. The islands persist even after very long mixing times and the phenomenon occurs in both continuous and discontinuous flows. The time evolution of the state of mixing is shown in Fig. 4 in terms of the fractional coverage of the dye versus the mixing time (image analysis studies[12] show that area coverage is proportional to the perimeter between the dye and the clear fluid for area coverages of less than 50%). An increase in De′ causes a decrease in asymptotic coverage and a slower rate of stretching (slope decreases). Figure 5 illustrates the effects of varying De by varying the shear rate. The experiments are conducted with a Newtonian fluid and the viscoelastic fluid C_2 under the discontinuous flow with a constant value of $D = 6.4$ for a total of eight periods. Figure 5(a) corresponds to the Newtonian system; Fig. 5(b) corresponds to the viscoelastic system with wall velocity $U' = 0.5$ cm/s (the characteristic shear rate is $\gamma' = U'/H = 0.08\ \mathrm{s}^{-1}$, and $\mathrm{De}' = \tau/\gamma' = 0.11$). Figure 5(c) corresponds to the viscoelastic system conducted at a higher wall velocity $U'' = 3.0$ cm/s ($\mathrm{De}' = 0.66$), whereas Fig. 5(d) is a continuation of 5(c), conducted at speed U' for two additional periods. At low shear rates the viscoelastic system [Fig. 5(b)] is indistinguishable from the Newtonian system, but at high shear rates the viscoelastic effects become significant and a large unmixed region appears [Fig. 5(c)]. However, if the shear rate is decreased, the unmixed region can be mixed with the rest of the fluid [Fig. 5(d)]. The conclusion is that the faster the "stirring" the worse the mixing. It is important to stress that the results seem to be rather insensitive to the rheological characteristics of the blob itself. For example, experiments conducted with a blob of PAA solution in pure glycerine do not show any significant differences with respect to pure glycerine for concentrations in the range 70–349 ppm and T in the range 34–70 s under the discontinuous mode of operation.

Can these results be modeled? Let us record a few observations. The results of Fig. 3 do not seem to be due to *asymmetry* in the streamline patterns; in fact, the symmetry apparent in the most viscoelastic case and for the

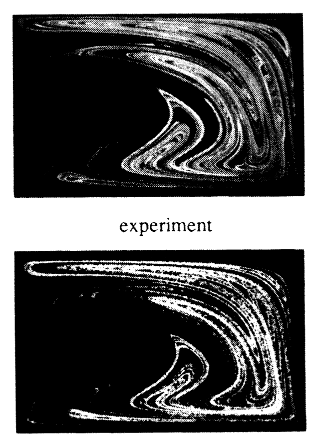

FIG. 6. Typical comparison between a computer simulation (finite difference) and an experimental result for a Newtonian fluid. The results correspond to a discontinuous flow with $D = 6.24$ and eight periods.

longest mixing time (Fig. 3, C_3-60) reinforces the belief that the instantaneous streamlines are symmetric in all cases. A small deviation in the *shape* of the streamlines—with respect to the Newtonian case—cannot account for the results either. Even though a computed streamline is inherently an approximation, simulations involving Newtonian fluids produce remarkable agreement with experimental results (see Fig. 6). Therefore, we are of the opinion that the differences in dye patterns are due to small variations of speed *along* the streamlines due to variations in the stress fields. The modeling of the results of Figs. 3 and 5 might present some difficulties. As is well known the computation of the viscoelastic velocity fields at moderate and high Deborah numbers is difficult and most studies to date have been restricted to steady flows.[16,17] Two other issues complicate the picture. The first has to do with *rate*: In the Newtonian case, and as long as inertial effects are unimportant, rate is of no importance and basically once the calculations have been carried out for one complete period the evolution of the tracer can be solved by iterations regardless of the speed of the boundaries. On the other hand, in the case of viscoelastic fluids, the mapping itself is a function of the speed. A second issue is agreeing on a suitable constitutive equation. To the extent that chaotic flows magnify rheological differences—witness Fig. 3—this aspect of the problem might be even more important than in steady flows. It is apparent that chaotic mixing of viscoelastic fluids deserves further computational and experimental scrutiny involving well characterized fluids.

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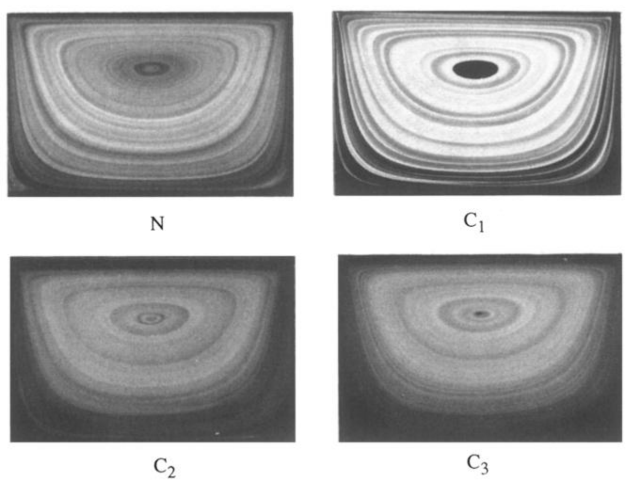

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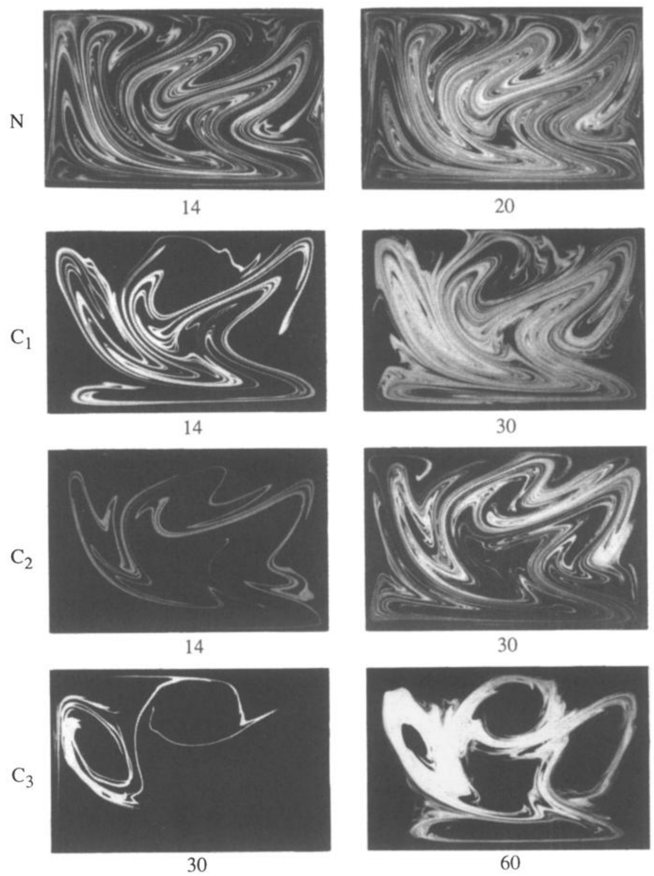

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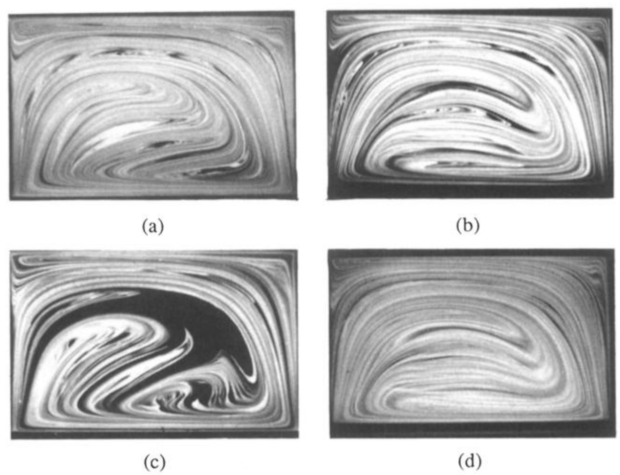

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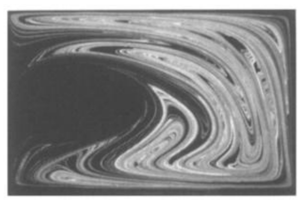
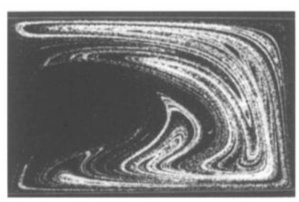


experiment



computation

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