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**Fundamental Studies of Fluid Mechanics
and Stability in Porous Media**

DE-FG03-87ER13673

Progress Report

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I. INTRODUCTION

This progress report covers our work over the last grant period (1990-1993). Support over this period has been in the form of graduate student assistantships for PhD students, partial support for postdoctoral fellows, maintenance of computer hardware, and construction of new experimental apparatus for the study of unstable drop spreading.

The report comprises summaries of important results we have obtained in various problem areas over the last three years. Much of the work has appeared in published papers, and in those cases, we do not go into extensive detail. However, more detail is provided for work in progress, especially when it relates to proposed further studies, discussed in the accompanying renewal proposal.

II. PROGRESS REPORT

1. Summary

We have been active in several areas over the last grant period. These include:

- o Semi - analytical studies of enhanced energy transport in natural convection due to time-dependent body forces (A. Farooq)
- o Large scale simulations of non-linear instabilities in porous media flow for situations of interest in enhanced oil recovery (O. Manickam, W. Zimmerman)
- o Analytical studies of 'chimney' formation in unstable freezing of mixtures (G. Amberg)
- o Analytical and experimental studies of contact line dynamics for Newtonian and non-Newtonian fluids (N. Fraysse, M. Spaid)
- o Large scale numerical simulations of shear instabilities of viscoelastic fluids (J. Azaeiz)

During the last grant period, we have worked with a number of students, postdoctoral fellows, and sabbatical visitors.

Past PhD Student

Dr. W. B. Zimmerman, PhD - Dr. Zimmerman completed a NATO Postdoctoral Fellowship in Madrid, Spain, and is now a Research Associate in Applied Mathematics, Sheffield University.

Past Postdoctoral Students

Dr. N. Ramanan - Dr. Ramanan is with Fluid Dynamics International, Evanston IL

Dr. Nathalie Fraysse - Dr. Fraysse is currently with the CNRS, Nice, France

Sabbatical Visitor

Prof. Gustav Amberg - Prof. Amberg is on the faculty at the Royal Institute of Technology, Stockholm, Sweden.

PhD Students

Mr. Jalel Azaiez - Mr. Azaiez has successfully defended his PhD thesis and will receive his degree at the end of the 1993 summer quarter.

Mr. O. Manickam - Mr. Manickam will defend his thesis and receive his degree in mid-September, 1993.

Mr. Michael Spaid - Mr. Spaid is entering his third year of study, has passed the PhD qualifying examination and has completed all his course requirements.

Ms. Jean Ro - Ms. Ro joined the group in Spring, 1993, and will take her PhD qualifying examination in October, 1993.

Postdoctoral Fellow

Dr. Anne DeWit - Dr. DeWit received her PhD from Universite Libre de Bruxelles, Belgium, and will join the group in October, 1993.

Published Papers

A. Papers appearing over the last grant period acknowledging DOE support:

1. "High Marangoni Number Convection in a Square Cavity. Part II". with B. Carpenter, Phys. Fluids A, 2, 137 (1990).
2. "Nonlinear Viscous Fingering in Miscible Displacement with Anisotropic Dispersion", with W.B. Zimmerman, Phys. Fluids A, 3(8), 1859 (1991).
3. "Viscous Fingering with Permeability Heterogeneity", with C.-T. Tan, Phys. Fluids A, 4, 1099 (1992).
4. "Three Dimensional Viscous Fingering: A Numerical Study", with W.B. Zimmerman, Phys. Fluids A, 4, 1901 (1992).
5. "Viscous Fingering in Miscible Displacements: Unification of Effects of Viscosity Contrast, Anisotropic Dispersion, and Velocity Dependence of Dispersion on Non-Linear Finger Propagation", with W.B. Zimmerman, Phys. Fluids A, 4, 2348 (1992).
6. "Nonlinear Analysis of Buoyant Convection in Binary Solidification with Application to Channel Formation", with G. Amberg, in press, J. Fluid Mech.
7. "Stability of miscible Displacements in Porous Media with Non-Monotonic Viscosity Profiles", with O. Manickam, Physics of Fluids A 6, 1356 (1993).

B. Papers acknowledging DOE support submitted and in the review process:

8. "Linear Stability of Free Shear Flow of Viscoelastic Liquids", with J. Azaiez, has been favorably reviewed: revision submitted to J. Fluid Mech. March, 1993.
9. "A Study of g-jitter Induced Streaming Motion", with A. Farooq, has been favorably reviewed: revision submitted to J. Fluid Mech. May, 1993.

10. "Simulation of Viscous Fingering in Miscible Displacements with non-Monotonic Viscosity Profiles", with O. Manickam, has been favorably reviewed: revision submitted to Physics of Fluids A August, 1993.

11. "Fingering Instability of Rotating Drops: Effect of Contact Angle and Fluid Elasticity", with N. Fraysse, submitted to Phys. Fluids A, September, 1993.

C. Papers in preparation for which the research was supported by DOE and is complete:

12. "Spin Coating of a Viscoelastic Liquid", with M. Spaid, to be submitted to J. Non-Newtonian Fluid Mechanics, October 1993.

13. "Gravity / Viscous Fingering by Numerical Simulation", with O. Manickam, to be submitted to Phys. Fluids A, October, 1993.

14. "Roll-up and Pairing of the Viscoelastic Mixing Layer", with J. Azaiez, to be submitted to J. Non-Newtonian Fluid Mechanics, October, 1993.

Invited Talks, Seminars and Meeting Presentations

Major Invited Talks and Plenary Lectures

"Fingers, Dendrites and Cracks: Modelling Unstable Growth Processes".
SIAM Annual Meeting, July 1990, Chicago IL

"Fingering instabilities in porous media: large scale simulations", Oberwolfach Conference on Porous Media, Oberwolfach, Germany, June, 1992.

"Fingering instabilities in porous media: large scale simulations", Gordon Conference on Porous Media, Plymouth, NH, August, 1992.

Presentations

G. M. Homsy, "Fingers, Dendrites and Cracks: Modelling Unstable Growth Processes". Departmental Seminar, Dept. Chemical Engineering, Univ. Florida, March, 1991.

G. M. Homsy, "Fingers, Dendrites and Cracks: Modelling Unstable Growth Processes". Applied Mathematics Colloquium, Stanford University, April, 1991.

O. Manickam & G. M. Homsy, "Effect of Non-monotonic Viscosity Profiles on the Stability of Miscible Displacements in Porous Media" Annual Meeting of the APS Division of Fluid Dynamics, Tempe AZ, November 1991.

J. Azaiez & G. M. Homsy, "Linear Stability of Inviscid non-Newtonian Free Shear Layers" Annual Meeting of the APS Division of Fluid Dynamics, Tempe AZ, November 1991.

G. M. Homsy & W. B. Zimmerman, "Viscous Fingering in 3D Porous Media: A Numerical Study" Annual Meeting of the APS Division of Fluid Dynamics, Tempe AZ, November 1991.

G. M. Homsy, "Numerical Simulation of Non-Linear Viscous Fingering in Miscible Displacements", Research seminars presented at ARCO Exploration and Production Technology *and* Mobil Research and Development Co., Dallas TX, Nov., 1991.

G.M. Homsy, "Recent Results in Viscous Fingering", seminar presented in Department of Applied Mathematics and Engineering Sciences, Northwestern University, April, 1992.

G. M. Homsy, "Surface Tension Driven Flows", seminar presented in Department of Chemical Engineering, Notre Dame University, April, 1992.

G. M. Homsy, series of seminars delivered to ESPCI, Nov.-Dec., 1992

"Viscous Fingering in Two and Three Dimensions"

"Surface Tension Driven Flows"

"Pattern Selection in Unstable Freezing of Binary Mixtures: A Porous Media Model"

G. M. Homsy, two seminars delivered at the University of Bordeaux, Nov., 1992.

"Viscous Fingering in Two and Three Dimensions" *and*

"Surface Tension Driven Flows"

J. Azaeiz & G. M. Homsy, "Linear Stability of Free Shear Flow of Viscoelastic Liquids", 64th Annual Meeting, Society of Rheology, Santa Barbara, CA, February, 1993.

G. M. Homsy, "Viscous Fingering in Two and Three Dimensions", Oxford Center for Industrial and Applied Mathematics, Oxford University, April 1993, *and* Dept. Theoretical and Applied Mathematics, Cambridge University, April 1993.

G. M. Homsy, "Pattern Selection and Bifurcation Theory for Unstable Directional Solidification of Mixtures", Seminar in Mechanics, Oxford University, *and* Department of Applied Mathematics, Sheffield University, April, 1993.

M. Martin & G. M. Homsy "Influence of Viscous Fingering on Zone Broadening in Liquid Chromatography", Symposium on Liquid Chromatography, Hamburg Germany May, 1993.

O. Manickam & G. M. Homsy, "Reverse Fingering in Miscible Displacements with Non-monotonic Viscosity Profiles", Annual Meeting of the American Physical Society Division of Fluid Dynamics meeting, Albuquerque, NM, Nov. 1993.

M. Spaid & G. M. Homsy, "Spin Coating of Viscoelastic Fluids", Annual Meeting of the American Physical Society Division of Fluid Dynamics meeting, Albuquerque, NM, Nov. 1993.

A. Farooq & G. M. Homsy, "G-jitter Induced Streaming in Natural Convection", Annual Meeting of the American Physical Society Division of Fluid Dynamics meeting, Albuquerque, NM, Nov. 1993.

G. M. Homsy & N. Fraysse, "Fingering Instability of Rotating Drops: Effect of Contact Angle and Fluid Viscoelasticity", Annual Meeting of the American Physical Society Division of Fluid Dynamics meeting, Albuquerque, NM, Nov. 1993.

2. Work Accomplished

A. Viscously Driven Instabilities in Porous Media (W. Zimmerman and O. Manickam)

We consider the miscible displacement process shown in fig.1. A fluid of viscosity μ_1 displaces a fluid of viscosity μ_2 . The two fluids are miscible and the viscosity is a function of the local composition of the fluid. We have carried out extensive studies for the cases when the viscosity-concentration relationship is monotonic, the permeability is constant, the dispersion characteristics are either isotropic or anisotropic, and the fingering is in either two or three dimensions. All of this work has been published [1,2,3] and has shown the important result that fingering involves no new mechanisms in three dimensions and that the separate effects of viscosity contrast, levels of dispersion, and dimensionality can be unified [2].

Our current work focuses on the effects on viscous fingering of viscosity profiles that are *non-monotonic*. When the viscosity μ varies non-monotonically with the local fluid composition c , the flow develops a potentially unstable region followed downstream by a potentially stable region as the two fluids disperse into each other as illustrated in fig.2. The stable region on the downstream side will act as a barrier to the growth of the viscous fingers, thereby providing a mechanism to control the growth rate of the fingered zone. Thus a fundamental understanding of the finger propagation in flows with non-monotonic viscosity profiles may serve as a building block to develop technologies aimed at controlling the growth of viscous fingers. We expect that the physical insights obtained through our study are extendable to viscous fingering against stable barriers achieved through other means such as the injected concentration field as in Water-Alternate-Gas (WAG) schemes, and naturally occurring heterogeneities in the porous media.

We have published our work on non-monotonic viscosity profiles in two parts [4,5] - the first part describes the results of a linear stability analysis and the second part presents the results of direct numerical simulations.

For flows with monotonic viscosity profiles, it is known that a favorable viscosity contrast results in a stable flow while an unfavorable viscosity contrast leads to instabilities. This conclusion does not necessarily hold when $\mu(c)$ is non-monotonic. Our linear analysis [4] shows that all flows with non-monotonic viscosity profiles, no matter what their endpoint viscosities are, will become unstable if diffusion is given sufficient time to act on the flow. Through asymptotic expansions in small wavenumber, we have established that $(d\mu/dc)_{c=0} + (d\mu/dc)_{c=1} < 0$ is a sufficient condition for the flow to remain unstable at all times. On the other hand, flows with viscosity profiles such that this quantity is positive are stable when the interface between the two fluids is sharp but as the fluids disperse into each other the flow will eventually become unstable. We have obtained the "critical" time at which a stable flow turns unstable as a function of the parameters of the problem. Diffusion is usually expected to stabilize a flow but here we have shown that on certain profiles diffusion acts to destabilize. We have put forth a physical mechanism to explain this surprising effect of diffusion.

The second part of the work [5] extends the linear analysis into the nonlinear regime by direct numerical simulations. The primary objective of this part of the work is to study the non-linear fingering mechanisms in displacements with non-monotonic viscosity profiles and compare them with those observed in flows with monotonic viscosity profiles. The striking contrast between the two is the direction of fluid penetration. The non-monotonicity in the viscosity profile gives rise to a new phenomena of "reverse" fingering in which the displaced fluid fingers through the displacing fluid more readily than vice-a-versa. One such case is shown in

Figure 1. Schematic of the miscible displacement process, showing the potentially unstable displacement of a higher viscosity fluid by one of lower viscosity.

Figure 2. (a) A schematic of the non-monotonic viscosity profile as a function of concentration and (b) its spatial variation under stable conditions.

Fig.3. The time sequence of the concentration contours are for a non-monotonic profile with an endpoint viscosity $\alpha = 0.5$, maximum viscosity $\mu_m = 2$ and the maximum located at $c_m = 0.75$. The concentration contours shown are as seen from a reference frame moving with the injection velocity. In the last frame the initial location of the interface is shown by a vertical line. Clearly, in a moving reference frame, the fingers have spread farther in the upstream than in the downstream direction, indicating reverse fingering.

We have quantitatively studied reverse fingering by comparing the growth rates of the mixing lengths in the forward and reverse directions which, at large times, grow linearly in time. In most of our simulations with non-monotonic viscosity profiles, the rate of growth of the reverse mixing length is larger than that of the forward mixing length. Detailed studies of isolated single fingers help us understand the differences in the mechanisms that control finger growth in flows with monotonic and non-monotonic viscosity profiles. In Fig. 4, the evolution of an isolated finger when the viscosity profile is non-monotonic is compared with that when the profile is monotonic. As seen in Fig. 4, the single finger grows rapidly when the profile is exponential whereas it is choked and its growth is stunted when the profile is non-monotonic. The primary reason for the difference is the mechanism of coalescence. In the exponential case, the emerging fingerlets coalesce with the main finger and supplies it with the necessary low viscosity fluid to sustain its growth. In the non-monotonic case, the strong reverse flow prevents the fingerlets from coalescing and the finger eventually fades away.

Current work is focusing on fingering driven by either viscosity or buoyancy differences, so that fingering can be initiated by unfavorable pressure forces and mitigated by gravity, or vice-a-versa. Somewhat surprisingly, we have made a connection between this problem and that of fingering with non-monotonic mobility profiles in the absence of gravity. In the usual case where the viscosity and density have different functional dependencies on the concentration of the solute, dispersion and the two forces of pressure and gravity can conspire to yield similar behavior in the fingering characteristics to that of the non-monotonic mobility case alone. Simulations are being completed and a manuscript is in preparation.

B. Contact Line Dynamics and Instabilities (M. Spaid and N. Fraysse)

Moving contact lines occur in a wide variety of energy related technologies, including manufacturing processes in which high speed coating takes place and immiscible displacements of oil by water in porous media. In many of these applications, non-Newtonian as well as Newtonian fluids are involved. We have begun a combined theoretical and experimental investigation of the flow and instabilities of spin coating of viscoelastic drops, a moving contact line problem. This situation is of particular interest in manufacturing processes in which thin liquid films of uniform and well-controlled thickness are desired, as in the manufacture of magnetic storage disks. In the so-called spin coating process, a viscous liquid drop is deposited on a dry solid substrate, then spread by spinning the disk. Once the leading edge of the drop becomes unstable, the liquid flows preferentially through the rivulets. This results in severe inefficiencies of the coating process. It has been shown that this instability is of hydrodynamic origin and is related to the existence of a capillary ridge of liquid near the contact line. It is thus of practical interest to understand the instability mechanism involved. In addition to being an important technological problem in and of itself, spin coating provides a generic setting in which the motion of the contact line is driven by an applied (centrifugal) force that can be varied over a wide range: the knowledge gained in this problem has immediate applicability to moving contact line problems in which the driving force may be gravity or an applied pressure gradient, such as occur in porous media applications.

Figure 3. Concentration fields at various times for the non-monotonic viscosity profile with an endpoint viscosity ratio $\alpha=0.5$, maximum viscosity $\mu_m=2$ located at $c_m=0.75$. The concentration contours span from $c=0.1$ to $c=0.9$ with equal increments and are shown in a moving reference frame. The computational domain is 1000 diffusive lengths wide with an aspect ratio of 2.

(a)

(b)

Figure 4. Evolution of isolated fingers. (a) for an exponential viscosity profile with an endpoint viscosity ratio of 10. The domain is of width 1500 and an aspect ratio of 2. (b) for a non-monotonic profile with an endpoint viscosity ratio $\alpha=8$, maximum viscosity $\mu_m=10$ located at $c_m=0.25$. The domain is of width 1000 diffusive lengths and the aspect ratio is 1.

Theoretical Studies

In our theoretical studies, the goals are to determine the effects of elasticity on a variety of aspects of the problem, i.e. the rate of drop spreading, the change in the shape of the free surface, and potential differences in the rivulet instability. Spreading of a Newtonian drop was studied by Melo *et al.* [6], and a stability analysis for the analogous problem of flow down an inclined plate was presented by Troian *et al.* [7]. These results may be used as a basis for comparison with the present theoretical development.

The spinning drop problem is of matched asymptotic expansion type in which there are two distinct regions, each having its own characteristic scalings. The first of these regions is the central part of the spinning drop, or region I in Fig. 5. In this region the dominant forces are the centrifugal force which tends to push fluid radially outward, and a viscous force which has an opposing effect. After some time, the free surface of the central part of the drop becomes flat, thinning uniformly in time. Near the contact line, surface tension competes with the centrifugal and viscous forces, and smooths the free surface as the fluid comes in contact with the spinning plate (region II). As the drop spins, a bulge develops near the moving contact line, and the shape of the bulge is instrumental in determining the stability of the advancing contact line. The third region in Fig. 5 is a small area of rapid variation near the actual point where the fluid contacts the plate, and arises due to complications with the no-slip condition near the moving contact line.

We chose to study the problem using lubrication theory, which is valid when the characteristic radial dimension R of the drop is much larger than the characteristic vertical dimension, h^* and when the contact angle is small. The Oldroyd-B constitutive equation is used to model the viscoelastic stresses present in the fluid. This model is derivable from kinetic theory by considering the forces acting on two beads attached by a linear spring. For the central part of the spinning drop, it is well known that for Newtonian fluids, the free surface $h(t)$ thins uniformly in time, with

$h(t) \sim \sqrt{\frac{1}{t}}$ for long times. We have shown that with the Oldroyd-B constitutive equation there *also* exists a solution in which the free surface thins *uniformly* in time. Perturbation theory was used to determine the effect of viscoelasticity on the thinning rate of the drop, considering small viscoelastic effects.

The first order perturbation result, which is valid over a few characteristic relaxation times of the fluid, gives the following equation for the free surface:

$$\frac{dh}{dt} = -\frac{2}{3} h^3 - \frac{16}{15} \epsilon We(1-S) h^5$$

where $\epsilon We(1-S)$ is the perturbation parameter which governs the magnitude of the viscoelastic terms in the constitutive equation. Neglecting the $\epsilon We(1-S)$ term gives the corresponding

Newtonian equation for the free surface, in which $h(t) \sim \sqrt{\frac{1}{t}}$ as mentioned earlier. It is evident from the equation that the first order viscoelastic effects tend to *increase* the thinning rate of the free surface. Analysis of this result indicates three dominant contributions which combine to form this enhanced thinning rate. The finite relaxation time of the fluid tends to decrease the thinning rate, but this effect is overwhelmed by normal stress gradients acting to increase the thinning rate.

The problem is rescaled near the contact line to include the effects of surface tension, and with suitable rescalings it is possible to derive a quasi-static equation governing the free surface in this region. Figure 6 shows the solution to the Newtonian free surface equation for several

Figure 5. Schematic of the free surface profile for a spinning drop, showing the three regions of interest in the theoretical development. The drop is spinning about a vertical axis with angular velocity ω . The sketch shows only half the drop, from the axis to the contact line region.

values of the parameter b , the precursor film thickness. (Matching the free surface to a precursor film near the contact line is one method that may be employed to relieve the contact line singularity.) For all values of the precursor film thickness, the free surface profile is characterized by a capillary ridge. Other methods, such as introducing a slip boundary near the moving contact line, may be used to relieve the contact line singularity. We have found that the essentials of the profile are *independent* of the way in which one chooses to relieve the contact line singularity.

We have also determined the correction to the free surface caused by viscoelasticity by a corresponding perturbation theory in the vicinity of the contact line. As before, this involved expanding the governing equations in the relevant parameter which determines the magnitude of the viscoelastic terms and deriving an equation for the free surface correction. The non-Newtonian free surface correction h_1 is shown in figure 6 for two values of the slip parameter b . The correction predicts an enhancement of the capillary ridge near the contact line. This change in the free surface shape suggests possible differences in the stability mechanism, since the capillary ridge plays a key role in the stability problem.

A manuscript describing this theoretical study is currently in preparation, and will be submitted to the J. Non-Newtonian Fluid Mechanics.

Experimental Studies

We have carried out an experimental studies to observe and understand the onset and evolution of centrifugally driven rivulets, with a special interest on the influence of the experimental parameters (drop volume and rotational frequency), the wetting properties of the liquid (surface tension and contact angle) and the fluid elasticity.

Experimental details

Our spin coating apparatus was designed to provide a range of rotational frequencies and to allow continuous observation of the entire drop. This is achieved by making use of dyed liquids and recording the absorption of light as it passes through the drop. Each experiment is video-taped using a CCD video camera placed above the set-up. Digital images are also taken by using the electronic shutter of the camera, achieving exposure times as short as $1/10,000$ s. These digitized pictures from the tape are then transferred to our Silicon Graphics Personal Iris workstation for image analysis.

We have studied three liquids so as to vary the contact angle on glass and introduce elasticity:

1. Light silicone oils polydimethylsiloxane (PDMS) which have a Newtonian behavior and are available in a wide range of viscosity. Those oils have a low surface tension ($21.5 \cdot 10^{-3}$ dyne.cm⁻¹) and a zero contact angle on glass.
2. A mixture of polybutene (PB) and kerosene which is the Newtonian solvent of the viscoelastic fluid we prepared. Its surface tension and contact angle on glass are larger than those for PDMS and are expected to be very close to those of the viscoelastic fluid.
3. A solution of polyisobutylene (PIB) (1000 ppm) in that solvent whose rheology was characterized on a Rheometrics Dynamic Analyzer II rheometer. Small amounts of a high molecular weight polymer in a viscous Newtonian solvent lead to a fluid which steady shear viscosity remains nearly constant up to large shear rates and which is highly elastic at room temperature (such fluids are often referred to as "Boger fluids"). The steady shear viscosity of our solution is 46 P and its relaxation time is about 1 s.

Substrates consisted of glass plates that were put through a cleaning protocol that ensured both a homogeneous surface and reproducibility of the flow results.

Figure 6a. Newtonian free surface profiles in the capillary region for three values of the dimensionless precursor film thickness, b . The vertical coordinate is scaled to the instantaneous film thickness in the outer region I (refer to Figure 5). For small ξ , the film reaches the prescribed precursor film thickness.

Figure 6b. Free surface corrections to the profiles in Figure 6a above due to non-Newtonian dynamics in region II for two different values of the precursor film thickness. Viscoelasticity raises the capillary ridge relative to the Newtonian solution by an amount proportional to the fluid relaxation time.

Experimental results

Figure 7 gives the results of a typical experiment. The initial spherical capped drop (Figure 7a) rapidly evolves into a drop profile which is nearly flat, except near the contact line where a bulge of liquid develops (Figure 7b). The contact line remains circular for a while, after which the drop circumference becomes angular, approximately polygonal (Figure 7c). An azimuthal modulation in the bulge height is faintly visible on the pictures as alternatively lighter and darker parts on the modulated ridge. The thickest parts are located at the polygon vertices which eventually give rise to rivulets. Those rivulets assume the shape of fingers (Figure 7d) as they grow and spread to the edge of the disk. Similar qualitative results were reported by Melo et al [6], but with only one substrate/fluid pair.

Using digital image analysis, we can characterize the drop shape and spreading rate, and in turn by computing the deformation of the contact line compared to a circle, we have access to three important features of the instability: the faster growing modes, their growth rate and an accurate determination of the critical time, and thus of the critical radius, at the onset of instability.

A linear stability analysis has been carried out by Troian and coworkers [7] for Newtonian liquids, using lubrication and quasi-steady approximations. The fastest growing mode observed experimentally is in very good agreement with the most unstable mode predicted theoretically as long as the critical radius is taken from the experiment itself, even for the viscoelastic fluid. Table 1 summarizes the results for a few experiments run with the various liquids.

We also compared the growth rate β (s^{-1}) of the fastest growing mode to that predicted by theory. In order to do so, we made the experimental growth rate dimensionless using a time scale t_0 adapted to the local behavior of the capillary ridge responsible for the instability. The dimensionless growth rates are equal for all experiments to a very good precision (Table 1), which confirms the choice of the time scale t_0 . The value of the numerical constant is in good agreement with the value at the maximum of the dispersion curve computed numerically by Troian et al [7]. The results we report here constitute the first confirmation of the validity of the linear stability analysis, with quantitative agreement on the whole set of instability features. An important consequence is that one is able to predict the wavelength as well as the growth rate, that is to say the exact way the instability develops in the linear regime, as long as one knows the critical time.

Experiments performed in identical conditions, with the Boger fluid solvent and a PDMS of same kinematic viscosity, show that a change in contact angle and/or surface tension significantly modifies the critical radius, which in turn affects the number of fingers (cf. Table 1 and Figures 8a,b which display the time-evolution of the contact line in both cases). This result is in contrast with conventional wisdom inferred from previous, less accurate experimental studies on gravity driven flows [8,9] for which no wavelength dependence on the wetting properties of the liquid could be detected.

No qualitative nor quantitative difference in behavior has been observed between the Boger fluid we prepared and its Newtonian solvent in the experimental conditions considered. As discussed above, a theoretical study of viscoelastic spreading is in progress in our group. We find that the key dimensionless parameter measuring the viscoelastic effect varies in our experiments from 6.10^{-3} to 2.10^{-1} . These low values are consistent with our experimental finding of negligible viscoelastic effects, at least over the parameter range studied.

A manuscript describing these experiments has been submitted to Phys. Fluids A.

Figure 7. Experimental visualizations showing the different stages of drop spreading, including axisymmetric spreading, initial development of the instability, and subsequent non-linear rivulet formation and propagation.

Exp. number	Number of Fingers N_f	Theoretical value of N_f	Growth Rate $\beta\tau_0$	R_c/R_1	t_c/T_1 ($\times 10^{-3}$)
PDMS 1	6	8	0.47	3.6	2.7
PDMS 2	7	10	0.56	3.9	2.8
PDMS 3	10	10	0.50	4	3.2
PDMS 4	9	9	0.43	3.75	2.7
PDMS 5	6	9	0.63	3.6	2.2
Solvent	4	5	0.55	2.7	0.68
Boger 1	4	4	0.46	2.6	0.78
Boger 2	3	3	0.44	2.2	0.13
Boger 3	4	4	0.53	2.6	<0.16

Table I . Comparison of the azimuthal wave-number N_f with the linear stability analysis predictions, and experimental values of the growth rate made dimensionless using a time scale τ_0 adapted to the local behavior of the inner region. Note that a constant value is obtained for $\beta\tau_0$, which is in agreement with the value of 0.4-0.6 predicted by the linear stability analysis. For a given liquid, the number of fingers is independent of the conditions of drop volume and rotational frequency within the experimental error. This yields a scaling whose characteristic time and radius are noted T_1 and R_1 . The value of the corresponding dimensionless critical radius depends on the liquid only.

Figure 8. Digital images of drop spreading and instability: (a) spreading of PDMS, (wetting fluid) showing the development of many rivulets: (b) spreading of the Boger fluid solvent, (finite contact angle) showing the earlier development of fewer rivulets.

C. Nonlinear Chimney Formation During Solidification of Binary Alloys (G. Amberg)

The problem of redistribution of chemical components during rapid solidification of mixtures is an important one in the manufacturing of alloys and castings, as well as in geophysical applications. Our work concerns the regime of rapid solidification which is well above the threshold for instability of planar freezing, and is characterized by the growth of dendritic structures, leading to a two phase region containing both solid dendrites and an interstitial fluid. One of the most interesting aspects of this problem is the occurrence, under certain circumstances, of 'chimneys' of clear fluid within the dendritic forest. These chimneys appear spontaneously and contain a component mixture which is buoyant relative to the surroundings and, when frozen, appear as 'freckles' of differing composition in the solid casting. Our work is a theoretical study to explain the origin of chimneys and freckles from a fluid mechanical viewpoint. We treat the two phase mixture as a saturated porous medium or 'mushy zone' and examine the nonlinear buoyancy driven convection therein during conditions of freezing. We are led to an effective porous medium convection problem, which we solve using techniques of weak non-linear bifurcation theory. The results clearly indicate that chimneys evolve from the coupling between buoyant convection, compositional melting of the mushy zone, and the resulting decrease in the resistance to flow. The bifurcation to two dimensional rolls may be either super- or sub-critical depending on the strength of the coupling between composition and permeability, and is trans-critical to hexagonal convection in three dimensions. These results are the first to establish an unequivocal connection between convection and chimneys, and are in good agreement with recent experimental observations of hexagonal planforms during freezing. The work is described in more detail in [10].

D. Polymer Flow Interactions in Free Shear Layers of Viscoelastic Fluids (J. Azaiez)

The problem of polymer conformation and interactions with flow is key in understanding the use of polymer additives to stabilize displacements against fingering instabilities. Due to the geometrical complexity of porous media, however, the local flow field is not well characterized, even in a statistical sense. Our long-range interest is in understanding how the large resistance of dissolved polymers to extensional flows leads to a modification of these flows, as well as the observed increase in pressure drop through porous materials.

As a first step, we are studying the interactions in a well-defined flow, namely a free shear layer, a sketch of which is given in Figure 9. The free shear layer represents an important prototype problem that is reasonably well-understood for Newtonian fluids. Important understanding of the mechanisms of vorticity production, subharmonic (pairing) instabilities, and vortex stretching has come from such studies.

Our goal is to study how dissolved polymers may influence stability and transition in the mixing layer. We are in particular interested in the possibility of viscoelasticity affecting known inviscid instability modes. To do so, we proceeded in two steps, first we examined the linear stability problem and then studied the full nonlinear problem through large scale simulations.

The choice of the constitutive equation describing the rheology of the fluid is a crucial factor in the study of non-Newtonian flows. For our study we adopted different models of progressive complexity and realism known to satisfactorily describe either dilute polymer solutions or melts over restricted ranges of shear and strain rates.

1. Linear Stability Analysis

When Cauchy's laws of motion and the rheological stress evolution equations are scaled, the flow problem is characterized by at least two dimensionless groups: the Reynolds number Re and the Weissenberg number We . These equations are analyzed by standard techniques. Details are presented in ref[4] above.

Our interest in the inviscid modes lead us to examine the special limit $Re \rightarrow \infty$, $We \rightarrow \infty$, We/Re fixed. In this distinguished limit, there is a coupling between vorticity and normal stresses leading to a *less unstable* flow, but viscoelasticity cannot completely stabilize such an inviscid flow. It is only in this distinguished limit that there is any significant modification of the inviscid vortex dynamics.

2. Numerical Simulations

We developed a code based on spectral method that we have validated by reproducing the well-known structures of the Newtonian mixing layer. In early calculations, we used the Oldroyd-B model to represent the viscoelastic fluid and we showed that this model is inadequate to simulate high shear flows due to an unlimited growth of the first normal stress difference ($a_{11} - a_{22}$).

Using the FENE-P model, which limits the extension of the polymer chain, we were able to run simulations at large elasticity number, $E = We/Re$. These simulations showed that the inertial times for the roll-up of the flow are not significantly affected by viscoelasticity, and that the global vortex structure is unchanged from the Newtonian case. The local distribution of the vorticity is however changed by the presence of viscoelasticity with a tendency for intense gradients to appear in the mid-braids, i.e the regions connecting contiguous vortices, as well as in the vortex core (Fig. 10). During the roll-up of the non-Newtonian mixing layer, more vorticity tend to remain in the braid than what is usually observed in the case of the Newtonian flow. The intensification of the vorticity gradients is spatially correlated with the build-up of the first normal stress difference ($a_{11} - a_{22}$). Similar results have been obtained for the subharmonic instability where the pairing of two viscoelastic vortices occurs virtually at the same time as for the Newtonian fluid. However, the parent vortices tend to rotate faster in the case of the viscoelastic fluid resulting in a more compact vortex structure with a larger absolute value of the minimal vorticity (Figure 11).

A manuscript describing the simulations is in preparation and will be submitted to the J. Non-Newtonian Fluid Mechanics.

E. Natural Convection with Time Dependent Body Forces (A. Farooq)

Natural convection is of interest for many technological applications, including thermal insulation, heat pipes, metallurgical technologies etc. We have focussed on natural convection induced by time dependent buoyant forces, which may arise due to vibrations produced by moving parts for example, and the effect that these forces have on the transport of scalars such as heat, which we have considered and others, as species concentration etc. A model problem has been selected: a square cavity, where a lateral temperature gradient interacts with a constant gravity field modulated by small harmonic oscillations of order ϵ , Fig.[12]. Invoking the Boussinesq approximation, the equations are expanded by regular perturbation in powers of ϵ , and the $O(\epsilon^2)$ equations contain Reynolds stress type terms that cause streaming. The resulting hierarchy of equations are solved by finite differences to investigate the $O(\epsilon)$ and $O(\epsilon^2)$ fields and their parametric dependence on the Rayleigh number Ra , Prandtl number, Pr and forcing

Figure 9. Schematic of the mixing layer, showing the roll-up into organized two dimensional vortices.

Figure 10. Roll-up of a single viscoelastic vortex, showing contours of both vorticity and primary normal stress difference. The vorticity field is characterized by a higher degree of striation and gradients than in the Newtonian case, which is spatially correlated with the build up of the normal stresses. The parameter values give the Reynolds number, Re , the elasticity number, E , and a parameter b associated with the FENE model.

T = 40.0
Minimum Contour= -3.300E-01
Maximum Contour= -7.000E-02

T = 40.0
Minimum Contour= -3.900E-01
Maximum Contour= -7.000E-02

T = 60.0
Minimum Contour= -2.900E-01
Maximum Contour= -7.000E-02

T = 60.0
Minimum Contour= -3.400E-01
Maximum Contour= -7.000E-02

T = 80.0
Minimum Contour= -2.600E-01
Maximum Contour= -7.000E-02

T = 80.0
Minimum Contour= -3.000E-01
Maximum Contour= -7.000E-02

T = 100.0
Minimum Contour= -2.400E-01
Maximum Contour= -7.000E-02
NEWTONIAN FLUID, Re=50

T = 100.0
Minimum Contour= -2.700E-01
Maximum Contour= -7.000E-02
FENE-P MODEL Re=50, E=1, b=5

Figure 11. Pairing of two viscoelastic vortices. Pairing occurs more quickly for viscoelastic vortices than the Newtonian counterpart due to the higher levels of vorticity remaining in the braid region during roll-up.

Figure 12. Schematic of the modulated heated square box. A horizontal temperature gradient is imposed on an enclosed Boussinesq fluid, which is then modulated in the vertical direction, leading to a modulation of the effective body force, $g(t)$.

frequency, ω . Since the problem is governed by no fewer than four parameters and there are a number of physical mechanisms operative in various regimes of this parameter space, the semi-analytic approach adopted here, while limited to small ϵ , yields insight and understanding that would be difficult to obtain otherwise. Our initial interest is in streaming flows caused by forcings that are periodic and aligned with the steady gravitational field and a temperature gradient that is normal to the direction of the gravitational field.

The $O(1)$ perturbation in the above hierarchy is the well studied case of natural convection of a heated cavity which we call the base state. The higher order time dependent perturbations of the base flow represent forced linear systems. One of the concerns in the present study is: Can the applied forcing interact with the natural modes of instability of the base flow? It is known that the base state shows two modes of instability: boundary layer waves associated with the instability of the boundary layers that form on the lateral walls of the cavity at high Ra and neutral gravity wave modes associated with density stratification in the core of the cavity, also found at high Ra . In our investigation (see [12] for details) we found that for high Ra when the core is stratified, application of a forcing of the same frequency as the Brunt-Vaisala frequency of the neutral gravity waves leads to strong resonance with the Ψ_1 field. In Fig.13, we plot the maximum value (over the cavity) of Ψ_1 , the $O(\epsilon)$ correction to the base flow (which is harmonic in time) as Pr and ω are varied, having fixed Ra at 10^4 . We comment on the following results. (i) For low frequencies all the curves are relatively flat and asymptote to a quasi-static limit. (ii) Curves for different Pr collapse for high ω confirming a Pr/ω scaling for Ψ_1 which can readily be derived from the equations in the asymptotic limit of large ω . (iii) For $Pr > 1$ the streamfunction exhibits a mild resonance; for $Pr \ll 1$ no resonance can be seen. The flow shows a broad resonant interaction and we call the frequency corresponding to the peak of the curve the resonant frequency. In Fig.14 we show contour plots of Ψ_1 for half a cycle of the forcing frequency. The time sequence shows the formation of a standing internal gravity wave in the center of the cavity, and follows its evolution as it is finally engulfed by the boundary layers.

The streaming flow, which is driven by the non-linear self interaction of the above mentioned stability modes also shows resonant behaviour at the Brunt-Vaisala frequency, Fig.15. However this resonant interaction is found only for Prandtl number near unity. Another interesting effect that is observed is that the $O(\epsilon^2)$ correction to the Nusselt number goes to zero and changes sign at the resonant frequency. This can easily be explained as being due to the formation of a standing internal gravity wave in the cavity.

Figure 13. Log scale plot of the magnitude of the first order harmonic response, Ψ_1 as a function of dimensionless modulation frequency. The Rayleigh number is fixed at 10^4 and the parameter is the Prandtl number. The curves show a mild resonance for $Pr > 1$ at a frequency corresponding to the Brunt-Vaisala frequency.

Figure 14. Contour plots of the harmonic response stream function Ψ_1 as the phase is varied, showing the formation and destruction of standing internal gravity waves. As the time sequence progresses, the disturbances originating in the corners interfere both constructively and destructively, leading to the formation of a standing wave. Later in the sequence, the vertical boundary layers become dominant and damp the wave, leading to a repetition of the cycle.

Figure 15. Log scale plot of the magnitude of the steady streaming field, Ψ_2 , as a function of dimensionless frequency, for $Pr = 1.0$. The parameter is the Rayleigh number, and there are only mild resonances at the Brunt Vaisala frequency.

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