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Fundamental Studies of Fluid Mechanics
and Stability in Porous Media

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Progress Report and Continuation Request

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1. Summary of Activities

This is a summary of activities on the stated grant over the last grant period, December, 1991 - present. We have been active in four areas:

Numerical and analytical studies of viscous fingering in miscible displacements, including *non-monotonic* mobility profiles.

Numerical and analytical studies of the effect of non-Newtonian fluid characteristics on instabilities.

Experimental studies of instabilities of moving contact lines for Newtonian and non-Newtonian fluids.

Studies of the natural convective energy transport due to time-dependent body forces.

Progress in each of these areas is detailed in the sections below.

Students and Collaborators

Four PhD students and a post-doctoral fellow have been supported or partially supported over the last grant period. Our visitor, Dr. Amberg, was supported through his home institution while collaborating on the work reported in publication no. 6 below.

Students

Dr. William Zimmerman - Dr. Zimmerman completed his PhD thesis in October, 1991, and began a NATO Postdoctoral Fellowship for study with Prof. Manuel Velarde, Madrid, Spain.

Mr. Jalel Azaeiz - PhD candidate - Mr. Azaeiz is a fourth year PhD student who has completed all his coursework and passed the PhD qualifying examination. He will complete his thesis in mid 1993.

Mr. Olagappan Manickam - PhD candidate - Mr. Manickam is a fifth year PhD student who has completed all his coursework and passed the PhD qualifying examination. He will complete his thesis in February, 1993.

Mr. Ahmed Farooq - PhD candidate - Mr. Farooq is a second year PhD student who has passed his PhD qualifying examination and has begun his thesis research.

Postdoctoral Fellow

Dr. Nathalie Fraysse - Dr. Fraysse received her PhD in Physics from the College de France

Sabbatical Visitor

Prof. Gustav Amberg, Royal Institute of Technology, Stockholm, Sweden - Prof. Amberg was on leave in our group for six months.

Publications acknowledging DOE support

A. Paper accepted or appearing in print during the last grant period:

1. C.-T. Tan & G. M. Homsy, Viscous fingering with permeability heterogeneity, *Phys. Fluids A* **4**, 1099, 1992.
2. W. B. Zimmerman & G. M. Homsy, Three dimensional viscous fingering: A numerical study, to appear in *Phys. Fluids A*, **4**, September, 1992.

B. Manuscripts under review:

3. W.B. Zimmerman & G.M. Homsy, Viscous fingering in miscible displacements: Unification of effects of viscosity contrast, anisotropic dispersion, and velocity dependence of dispersion on non-linear finger propagation, reviewed favorably and revision re-submitted to *Phys. Fluids A*.
4. J. Azaiez & G. M. Homsy, Linear stability of free shear flow of viscoelastic liquids, submitted to *J. Fluid Mech.*
5. A. Farooq & G. M. Homsy, A study of g-jitter induced streaming motion, submitted to *J. Fluid Mech.*
6. G. Amberg & G. M. Homsy, Nonlinear analysis of buoyant convection in binary solidification with application to channel formation, submitted to *J. Fluid Mech.*

C. Manuscript in preparation

7. O. Manickam & G. M. Homsy, Fingering instabilities with non-monotonic mobility profiles, to be submitted to *Phys. Fluids A*.

Invited Talks - G.M. Homsy

"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.

Other Presentations

O. Manickam & G. M. Homsy "Effect of Non-monotonic Viscosity Profiles on the Stability of Miscible Displacements in Porous Media" presented at the 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" presented at the 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" presented at the 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.

2. Work Accomplished

Viscous Fingering in Miscible Displacements (W. Zimmerman, O. Manickam)

We consider the miscible displacement process illustrated below.

A fluid of viscosity μ_1 displaces a fluid of viscosity μ_2 at a uniform base velocity U . Miscible displacement is of much practical interest due to its applications in enhanced oil recovery, channelling in packed beds, and fixed bed reactor regeneration. Much past work has dealt with the effects of adverse viscosity ratio and isotropic dispersion on the stability and nonlinear evolution of viscous fingers. Largely ignored have been the effects of the tensor character of dispersion, its velocity dependence, the issue of whether two-dimensional simulations can represent three-dimensional fingering, and the possibility that mobility profiles might be *non-monotonic* in space.

The PhD thesis of W. Zimmerman focused on the first two of these issues. In the first of two papers, we demonstrated that anisotropic dispersion leads to new finger interaction mechanisms at lower Peclet numbers than with isotropic dispersion: Zimmerman & Homsy (1991). Through extensive simulations reported in [3], we then demonstrated that the effects of an anisotropic, velocity-dependent dispersion tensor, mobility ratio, and isotropic dispersion at extremely high Peclet number could be unified in terms of a small discrete set of finger interactions. Finally, we discovered an intriguing enhancement of the rate of finger growth when the velocity dependence of dispersion is sufficiently strong. This effect was also recently observed experimentally by Salin and co-workers in Paris (Bacri, et al (1991)).

A third paper [2] dealt with the important problem of fingering in three dimensions. There had previously been no reliable numerical simulations of fingering at high Pe in three dimensions. We developed a new vorticity formulation that allowed efficient use of spectral methods in three dimensions. In the region of parameter space accessible by our study, we found little change in the average profiles from the corresponding 2-D model. Similarly, we found that the interaction mechanisms of coalescence, shielding, spreading, and tip-splitting differed little from their 2-D counterparts. This leads to the important engineering conclusion that the *averaged* properties of 3-D fingering may be accurately predicted by 2-D models. The conclusion was related to the fact that there are no *new* mechanisms of vorticity production or transport in 3-D.

We have also continued our studies of fingering with non-monotonic mobility profiles. For a monotonic dependence of viscosity on concentration, $\mu(c)$, it is known that an unfavourable viscosity contrast ($\mu_1 < \mu_2$) results in instability. This same conclusion doesn't necessarily hold if $\mu(c)$ is non-monotonic, and leads to the possibility of instability when the overall mobility ratio is *favorable*, and vice-a-versa. The objective of the present work is to make precise the conditions for instability and to investigate the nature of non-linear fingering in such circumstances.

Linear stability analyses have been carried out to answer the first question. It is possible to obtain analytical results for the case of a sharp jump in concentration, corresponding to zero axial dispersion. The results indicate that the stability of the interface depends not on the end-point viscosities but on the end-point derivatives of viscosity with respect to concentration. The criteria for instability is $(d\mu/dc)_{\infty 1} + (d\mu/dc)_{\infty 0} < 0$, giving the following interesting possibility. A non-monotonic variation of viscosity with concentration $\mu(c)$ can yield a stable displacement even if the viscosity contrast is unfavourable ($\mu_1 < \mu_2$). Asymptotic expansions for short time shows that the diffusion of the base state does not always mitigate the instability. These analytical studies have been augmented by numerical results in the case of diffuse profiles (due to finite axial dispersion). It is shown that dispersion does not always stabilize the process: we have discovered some curious counterexamples in which a sharp front might be predicted to be stable, but a diffuse front is not (for the same parameters). A manuscript describing this work is in preparation.

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 those flows, as well as the observed increase in pressure drop through porous materials.

As a first step, we are studying these interactions in a well-defined flow, namely a free shear layer, a sketch of which is given below. The free shear layer represent an important prototype problem that is reasonably well-understood for *Newtonian* fluids (see e.g. Corcos & Lin (1984), Metcalfe, Orszag *et al* (1987), Moser & Rogers (1991)). Important understanding of the mechanisms of vorticity production, subharmonic (pairing) instabilities, and vortex stretching has come from such studies.

Schematic of a free shear layer

The goal of our research is to study the question of 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. We have begun a study of these issues by: (1) analyzing the linear stability problem and (2) developing codes for 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 Instability 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.

Oldroyd-B model

The Oldroyd-B equation, which can be derived from a molecular model in which the polymer molecule is idealized as a Hookean spring connecting two Brownian beads, gives acceptable qualitative predictions for dilute solutions. It predicts no shear thinning, a constant first normal stress coefficient and a zero second normal stress coefficient. As a result, it is limited to moderate shear and strain rates.

Since we are interested in the inviscid modes, we examined the limit of $Re \rightarrow \infty$, $We \rightarrow \infty$, We/Re fixed. In this distinguished limit, there is a coupling between vorticity and normal stresses even in the inviscid limit. This coupling is characterized by an *elasticity* parameter,

$$G \equiv We / Re = \lambda v / \mathcal{E}$$

representing the ratio of elastic to viscous relaxation times within the fluid. We find that viscoelasticity has a stabilizing effect on the *inviscid* modes which is due to the enhanced resistance of polymers to the plane strain associated with the Kelvin-Helmholtz instability. The spectrum of wave numbers for which the flow is unstable is shifted towards longer waves and the maximum growth rate is reduced in a similar manner. We speculate that the mechanisms of roll-up and pairing are reduced if not totally suppressed by the presence of small amounts of viscoelastic additives. These predictions are in qualitative agreement with experimental results.

Giesekus model

This model is based on the concept of a deformation-dependent tensorial mobility, allowing an improved description of polymeric solutions. It enables a description of a number of well-known properties of viscoelastic fluids, namely shear thinning, non-zero secondary normal stress coefficient and stress overshoot in transient shear flows. This model is characterized by an extra dimensionless parameter known as the mobility factor. Linear theory calculations based on a modified Orr-Sommerfeld equation show that the behavior for the Oldroyd B fluid is robust to changes in the constitutive model, and that similar stabilizing effects occur over ranges of We and the mobility factor.

2. Direct Numerical Simulation

Our current work is focused on the simulation of the roll up of the viscoelastic mixing layer, with particular attention paid to the effect of viscoelasticity on the non-linear dynamics. The linear theory indicates a shift to longer wave lengths and a diminishing of the temporal growth rate, but that should not be taken to mean that the non-linear problem is simpler than the Newtonian counterpart. In fact, it is much more difficult, since the stress evolution equation must be solved simultaneously with Cauchy's laws of motion. The current status is that we have a functioning computer code based on spectral methods for the viscoelastic problem that converges to quasi-equilibrated vortices for very moderate values of We and Re , an example of which is given in the following figure.

$Re=100$. $We= 1$. $ALPHA=0.44$ $K=0.50$

Example of the roll up of a single vortex for modest levels of viscoelasticity

Numerical instabilities have kept us from extending the range of parameters. Once these problems are resolved, the calculations will be extended to include an analysis of the secondary instabilities of both streamwise pairing and spanwise instability of rows of vortices. The latter will be especially instructive in interpreting the effect of polymers on transition to turbulence.

Thermal Convection with Time-Dependent Body Forces (A. Farooq)

The problem of energy transfer in natural convection is important in many technologies, including thermal insulation, extraction of thermal energy from porous formations, heat pipes, etc. We are interested in the changes that occur in thermal energy transfer when the body force is time dependent, such as might be the case when a system is subject to forced oscillations. There is almost no information available on this issue. Accordingly, we have begun by considering a model problem of a cavity heated in the horizontal and oscillated about the vertical axis, producing a periodic component to the effective gravity.

A semi-analytical method was used to examine this problem initially. The solution was expanded in a series expansion in the amplitude of the modulation, ϵ , taken to be a small parameter. The resulting set of equations was then be solved sequentially. The parameters are the Rayleigh number, Ra , the Prandtl number, and the dimensionless frequency of oscillation. The $O(1)$ equations are just those for steady thermal convection with a steady body force. The $O(\epsilon)$ equations describe the periodic response to periodic forcing, while the $O(\epsilon^2)$ equations have Reynolds-stress type terms that produce a steady streaming flow. These equations have been solved in the quasi-static limit to find the dependence of the streaming flow and the resulting energy transfer on Ra . The streaming flow is small at low Ra , as expected, and becomes appreciable at higher Ra , leading to an $O(\epsilon^2)$ correction to the Nusselt number. This correction exhibits an interesting and surprising change of sign as Ra increases, which is explained in terms of the convective effects of the streaming flow on the steady isotherms. These results are described in more detail in the manuscript ref. [5] above.

Viscoelastic Moving Contact Line Problems (N. Fraysse)

The flow of viscoelastic liquids near free boundaries and interfaces in a generic situation that arises in many diverse technologies. These include spin coating of non-Newtonian dispersions and polymers onto solid substrates for magnetic recording and lubrication applications, enhanced oil recovery involving the displacement of oil by polymer solutions in mixed wettability reservoirs, break-up of liquid threads into droplets, and a variety of printing applications. All of these applications involve moving contact lines between two fluids (a liquid and another fluid) and a solid. When the location of the contact line is particularly simple, steady and/or stable flows may result. On the other hand, an imbalance of surface forces may lead to interfacial instabilities that couple to bulk motions, leading to more complex dynamics.

We have begun a study of how viscoelasticity can influence contact line motion and contact line instabilities in spin coating of drops. In this regard, the use of centrifugal force is a convenience: the understanding obtained can be generalized to other situations. In the spin coating process, liquid is deposited at the center of the solid substrate, which is then rotated. Centrifugal forces cause a radial outward flow with an accompanying movement of the contact line. This process is not always stable. The figures below show photographs taken in our laboratory of the rivulet formation resulting from an instability of the motion of the contact line. (A viscous, Newtonian fluid was used in this particular experiment.) Such contact line instabilities prevent the achievement of a uniform coating of constant thickness and are important in the microscopic displacement of non-wetting fluids in porous media.

Sequence of photographs showing the contact line instability during spin coating of silicone oil on glass: the rotational rate is 700 RPM and the frames are at times of 0s, 30s, 50s, and 100s.

Our efforts to date have been devoted to the construction of a special spin coating apparatus the provides the following capabilities:

- (i) a range of rotational frequencies
- (ii) a circumferential belt drive, allowing the use of digital imaging of the entire drop
- (iii) interfacing with a stroboscope, a small lab computer, and our Silicon Graphics Personal Iris workstation for digital image analysis.
- (iv) techniques for centering the drop on the axis of rotation to within 0.1 mm have been perfected.

3. Proposed Work

During the next grant period we intend to pursue three projects: fingering with either non-monotonic mobility profiles or competing viscous/gravity effects, energy transfer with modulated body forces, and viscoelastic contact line instabilities.

Viscous Fingering in Miscible Displacements (O. Manickam)

Linear stability analysis gives the possibility of a stable displacement even under an unfavorable end-point viscosity contrast. But this holds true only for small disturbances, and it is not clear whether the displacement will remain stable when the disturbances are of finite amplitude.

Furthermore we suspect that the non-monotonicity of $\mu(c)$ will give rise to "penetrative fingering", in which the spatial extent of the fingering will be limited to 'unfavorable' parts of the mobility profile, and therefore will not propagate indefinitely as occurs with monotonic profiles.

In addition, we are interested in the problem of non-linear fingering when both buoyant and viscous forces are present, as sketched below. Because of the dependence of both density and viscosity on concentration, buoyancy and viscous forces both contribute to the dynamics. The linear theory for combined gravity/viscous fingering is classical, and is due to Hill (1952). He showed that a combined parameter determined stability or instability, but these results do not extend into the non-linear regime. Recently, Prof. Dominique Salin and his co-workers in Paris have obtained experimental data on the averaged properties of front propagation for these cases. They find significant differences in unstable propagation, depending on whether gravity is stabilizing or destabilizing. Such differences remain unexplained and influence the development of both understanding and engineering models to describe these situations.

We will study both of these non-linear problems using large scale simulations. We have made modifications to the Hartley transform based numerical scheme of Zimmerman & Homsy (1991) that has resulted in a two-fold increase in the speed of the code, which is currently being run on the San Diego Supercomputer Center. These studies will comprise the PhD thesis work of Mr. Manickam.

Thermal Convection with Time-Dependent Body Forces (A. Farooq)

Current work is involved in relaxing the assumption of quasistatic modulations. We expect to discover near resonances between the harmonic forcing and naturally occurring oscillatory modes of convection, dependent on the dimensionless frequency and the Prandtl number. This study is being pursued using the same finite-difference numerical techniques that we developed for the quasistatic case.

Longer range objectives involve the relaxation of two assumptions. First, other degrees of freedom such as rocking motions and oscillations out of the plane of the temperature gradient will be analyzed using the same semi-analytical approach. Second, the assumption of small modulations will be relaxed with the idea that the response of a nonlinear system to harmonic forcing may become complex and chaotic when the forcing is large. We will pursue this idea

initially by using the temperature and velocity fields obtained numerically as trial functions in a truncated Galerkin expansion. In this way we can derive a low-order set of amplitude equations whose bifurcation structure will be analyzed using the techniques of dynamical systems theory. Regimes of interesting dynamical behavior will be validated by a full numerical simulation of the governing partial differential equations.

Contact Line Instabilities (N. Fraysse)

Our apparatus has been built and several experiments with Newtonian fluids have been conducted and have been validated against existing theory and experiment for Newtonian fluids. We are currently planning a suite of experiments with viscoelastic fluids with significant relaxation times in order to probe the viscoelastic response at significant values of the Weissenberg number. We will then follow this with analyses (based on lubrication approximations) that will elucidate the mechanisms by which the combination of significant normal stresses and finite relaxation times influence the instability.

4. References

Bacri, J.-C., D. Salin & R. Woumeni, "Three dimensional miscible viscous fingering in porous media", *Phys. Rev. Lett.* **67**, 2005, 1991.

Corcos, G. M. & S. J. Lin "The mixing layer: deterministic models of a turbulent flow: Part 2. The origin of the three-dimensional motion" *J. Fluid Mech.* **130**, 67, 1984.

Hill, S. "Channeling in packed columns" *Chem. Engr. Sci.* **1**, 247, 1952.

Metcalf, R.W., S.A. Orszag, M.E. Brachet, S. Menon & J.J. Riley "Secondary instability of a temporally growing mixing layer" *J. Fluid Mech.* **184**, 207, 1987.

Moser, R. D. & M. M. Rogers "Mixing transition and the cascade to small scales in a plane mixing layer" *Phys. Fluids A*, **3**, 1128, 1991.

Zimmerman, W. & G. M. Homsy "Nonlinear viscous fingering in miscible displacement with anisotropic dispersion" *Phys. Fluids A* **3**, 1859, 1991.

5. Budget Justification

The proposed budget contains salaries for two PhD students, partial support for Dr. Fraysse, and one month salary for the PI. In addition, funds for incidental supplies and communications are requested. The requested travel funds are intended to cover costs for the PI to attend and present a paper at one major meeting, and travel costs for advanced students to attend either the AIChE or the APS/DFD annual meeting and present their work. The majority of the proposed work for the next grant period is computational in nature, although some funds must be budgeted for expendables related to the contact line experiments .

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