The Dynamics of Miscible Interfaces: Simulations

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Final Report: 3-28-05

Abstract

This research project focuses on the dynamics of interfacial regions between miscible fluids. While much attention has focused on immiscible interfaces in the past, miscible interfaces have been explored to a much lesser degree, so that there are many open questions regarding their dynamics at this time. Among the more pressing issues is the role that nonconventional stresses can play in such interfacial regions. Such stresses are typically not accounted for in efforts to model the dynamics of miscible flows. Our research aims to clarify under which circumstances these stresses do have to be taken into account, and what quantitative approaches are most suitable in this regard. In order to address these issues, we have focused on conducting linear stability analyses and nonlinear simulations for capillary tube and Hele-Shaw flows, and to compare the results with corresponding experiments performed in the labs of our co-investigators Prof. Maxworthy at USC, and Dr. Balasubramaniam at NASA. Over the duration of the project we have, among other things, focused on the effects of variable diffusion coefficients in such flows, and specifically on their influence in the growth of instabilities. Furthermore, our three-dimensional spectral element simulations have made good progress, so that we have come to a point where we can conduct more detailed comparisons with experimental observations. We are currently focusing our efforts on reproducing the tip-splitting instability observed by Maxworthy. Finally, we have discovered a new core-annular flow instability in the Stokes flow regime during the last year. This represents a significant finding, as this instability does not have an immiscible counterpart.

Research objectives

While it is generally accepted that the Navier-Stokes equations accurately describe the motion of uniform fluids, there still exists uncertainty about the correct form of the equations governing miscible fluid flows in the presence of concentration gradients. Already more than a century ago, Korteweg01 pointed out that significant additional stresses may exist in regions of large concentration gradients, which can give rise to surface tension like phenomena. Throughout the following decades, anecdotal observations of such an `effective surface tension' were reported in the experimental literature, such as miscible drops whose properties resemble those of immiscible drops with surface tension.

A more fundamental investigation of this issue did not begin until the late 1980s, and a consistent derivation from first principles is still lacking even at this time. Among the first theoretical investigations is the work by Davis, who employs arguments from molecular theory to estimate the magnitude of these stresses as a function of the

concentration layer thickness. His arguments show the stresses to scale with the square of the concentration gradient. Most macroscopic, continuum based attempts directed at a quantitative description employ this scaling as well. However, since the magnitude of the material coefficients in the expressions for these stresses is unknown, their influence on the dynamics of miscible fluid flows still cannot be predicted with any degree of certainty.

One potentially promising way to evaluate the strength of the Korteweg stresses relies on a detailed comparison between theoretical predictions and experimental measurements for a well defined flow field. Ideally such an analysis should be performed for a flow in which surface tension like stresses have a significant influence on phenomena that can be accurately measured experimentally. The experimental data can then be compared with theoretical predictions derived on the basis of a specific Korteweg stress model, in order to extract information about the magnitude of the stress coefficients.

Miscible capillary Hele-Shaw flows represent strong candidates for a combined experimental/theoretical investigation along the above lines, although other flow types have been explored in this regard as well. They give rise to well known, competing instability mechanisms driven by density and/or viscosity gradients, as first seen in the classical work of Wooding. In these flows, interfacial stresses strongly affect such features of the dispersion relations as the cutoff and most amplified wavenumbers, along with the maximum growth rate.

Traditional analyses are based on the gap-averaged Hele-Shaw equations, whose applicability to variable density and viscosity flows is limited to fairly thick interfaces and small values of a suitably defined Rayleigh or Peclet number. For this reason, one cannot hope to extract quantitative information about Korteweg stress coefficients by comparing the results based on the Hele-Shaw equations with experiments. The limited applicability of the Hele-Shaw equations was clearly demonstrated in our recent work by Graf et al., who investigated density-driven instabilities between constant viscosity, miscible fluids in a vertically oriented Hele-Shaw cell. There we performed a detailed linear stability analysis based on the conventional three-dimensional Stokes equations (without Korteweg stresses), and they compared the results with corresponding predictions based on the Hele-Shaw equations. As expected, the respective results were seen to agree only under conditions for which the velocity and concentration gradient components in the cross-gap direction are small. On the other hand, the three-dimensional Stokes flow stability results were seen to compare well overall with corresponding experimentally measured dispersion relations by \cite{Fernandezetal01}. Nevertheless, there appeared to be certain moderate but systematic discrepancies. For example, in the experimental relation of the maximum growth rate as a function of the Rayleigh number, an overshoot was observed for moderate values of Ra, which could not be explained theoretically on the basis of the Stokes equations. A similar overshoot was found to exist for the most amplified wavelength as a function of Ra.

The situation in capillary tubes is conceptually quite similar, so that we also focus a substantial part of our efforts on this class of flows.

Accomplishments over the duration of the project
Over the duration of the project, we have pursued the following three
directions in our research on miscible interfaces:

*1. Hydrodynamic stability of miscible fluids, where a heavier fluid is
placed above a lighter one in a vertically arranged capillary tube or
Hele-Shaw cell*

Here we have been analyzing the influence of a concentration dependent
diffusion coefficient on the density driven instability in vertically
arranged Hele-Shaw cells and capillary tubes. We have performed the
major part of a detailed linear stability analysis based on the Stokes
equations without main flow, and by employing the Stokes-Einstein
relation between the viscosity and the diffusion coefficient. We have
begun the interpretation of the eigenfunctions and dispersion relations
for different Rayleigh numbers, mobility ratios and interface
thicknesses. When interpreting the results, we find that it is helpful
to distinguish between the influence of the variable diffusion
coefficient on the base concentration profile, and its influence on the
growth of the perturbations themselves. The variable diffusion
coefficient leads to an asymmetric base concentration profile with a
long tail in the less viscous fluid. Hence there is a competition
between two mechanisms: On one hand, the base profile is steeper in the
high viscosity region, which can drive the instability. On the other
hand, the perturbations can grow more easily in the low viscosity
region, due to the locally higher mobility. This competition leads to
interesting growth dynamics and 'optimal behavior' for various
parameter regimes.

*2. Three-dimensional, spectral element simulations of displacements in
capillary tubes*

We have begun to conduct highly resolved, three-dimensional spectral
element simulations of variable viscosity and density displacements in
horizontal capillary tubes. We have been able to reproduce some of the
results observed by Petitjeans and Maxworthy (1996), in that the finger
becomes non-axisymmetric due to density differences in the
gravitational field. While the finger tip stays close to the tube's
axis, the trailing sections of the finger are displaced in the upward
direction due to the buoyancy forces acting on the finger. We have not
yet been able to reproduce the 'tip-splitting' results observed by
Petitjeans and Maxworthy. This may be due to the fact that our
parameter combination (Pe, F, At) is not yet in the appropriate region,
but it could also be a result of the fact that we are currently
employing a diffusion coefficient that does not vary with the
concentration. We plan to conduct a more systematic exploration of the
parameter regime, in order to pursue these issues in more detail.

3. Miscible core-annular flow instability in the Stokes regime

The stability of variable density and viscosity, miscible core-annular
flows in the Stokes regime is investigated. Such flows are found to be
linearly unstable towards both short and long wave disturbances, and
for a wide range of the governing parameters. This is in contrast to
immiscible core-annular flows, which are known to be stable in the

Stokes regime. A close inspection of the base flow properties along with the perturbation eigenfunctions provides insight into the physical mechanisms driving the instability. These mechanisms are distinct from those responsible for immiscible core-annular flow instabilities at non-zero Reynolds numbers, although similarities do exist. A prerequisite for the unstable nature of miscible Stokes flows of core-annular type is the finite thickness of the interfacial region, which allows for the emergence of a phase shift among the concentration contours, and between the perturbation concentration and the perturbation vorticity. This phase shift is caused either by a vortex within the interfacial region (for viscosity-driven instabilities), or by perturbation vorticity in the external flow (for density-driven instabilities). The phase enables the velocity field to amplify the initial interfacial disturbance, thus resulting in unstable behavior. The growth rate is seen to approach zero as the interface thickness of the base flow is reduced, which confirms the importance of the finite width of the interfacial region. A parametric study provide information on the influence of the various governing parameters on the linear instability modes.

Publications

Density-Driven Instabilities of Miscible Fluids in a Capillary Tube: Linear Stability Analysis, with S. H. Vanaparthy and D. Wilhelm, **J. Fluid Mech. 497**, 99 (2003).

Three-Dimensional Spectral Element Simulations of Variable Density and Viscosity, Miscible Displacements in a Capillary Tube, with D. Wilhelm, **Comp. Fluids 33**, 485 (2004).

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Unstable Density Stratification of Miscible Fluids in a Vertical Hele-Shaw Cell: Influence of Variable Viscosity on the Linear Stability, with N. Goyal, **J. Fluid Mech. 516**, 211 (2004).

Density-Driven Instabilities of Variable Viscosity Miscible Fluids in a Capillary Tube, with S.H. Vanaparthy, M. Payr and D. Wilhelm, to appear in the **Ann. New York Acad. Sci.** Also: <u>Proc. of the Microgravity Transport Processes in Fluid, Thermal, Materials, and Biological Sciences Conference III</u>, Davos, Switzerland, September 2003.

Influence of Variable Viscosity on Density-Driven Instabilities in Capillary Tubes, with M. Payr and S.H. Vanaparthy, to appear in **J. Fluid Mech.**

The Influence of Korteweg Stresses on the Density-Driven Instability between Miscible Fluids in a Vertical Hele-Shaw Cell, with U. Gerke, submitted to **J. Fluid Mech.**

Miscible Core-Annular Flows: Linear Instability in the Stokes Regime, with H. Koberg and M. Payr, submitted to **J. Fluid Mech.**