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## MULTILAYER FLUID DYNAMICS OF IMMISCIBLE LIQUIDS

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## ABSTRACT

A comprehensive analytical, numerical and experimental study of convective flow in multilayer immiscible liquids has been performed. Studies include transparent high Prandtl number liquids as well as opaque low Prandtl metallic melts. A new radiosopic flow visualization has been developed for the latter studies.

## INTRODUCTION

The study of thermal convection in a double layer system of immiscible liquids is inspired by the development of liquid encapsulated crystal growth techniques for producing mono-crystalline electronic materials. Encapsulation of the electronic melt is used to control the melt's stoichiometry when the melt contains a volatile component. The encapsulant, typically a liquid glass, also has the potential for reducing or even eliminating convective flow in the melt. Convective flow in the melt, especially time-dependent flow, can lead to undesirable inhomogeneities in the solidified material. To reduce buoyancy driven thermal convection in the melt by taking advantage of the reduced gravity environment aboard a spacecraft, space based processing of GaAs with the liquid encapsulated float zone processing technique has been proposed.

Fluid dynamics of the melt float zone during the solidification process poses a formidable problem involving the transport of mass and heat along with phase change in one layer. To make the problem manageable we focus on simplified model situations. We consider the following basic multiple layer fluid dynamical systems [1-14]:

- I) High Prandtl number fluids
  - a) Natural convection featuring a liquid/liquid interface and a free upper surface
  - b) Rayleigh-Bénard convection featuring one interface
- II) Low Prandtl number fluids
  - a) Development of a radiosopic flow visualization capability
  - b) Single/double layer natural convection with solidification

The multi-layer problem is characterized by mechanical and thermal coupling across liquid/liquid interfaces. The liquid layers are mechanically coupled via transfer of momentum across the interfaces. Transfer of momentum results from the continuity of interface tangential velocity and balance of shear stress across the interface. Together these two conditions comprise the 'no-slip' condition at a liquid-liquid interface. Thermal coupling is achieved through continuity of temperature at the interface and the balance of heat transfer across the interface. It is these coupling conditions that distinguish the multi-layer problem from the single layer problem.

We also present results from an analytical study of thermocapillary flow in a horizontal liquid layer encapsulated by two immiscible liquids, featuring two liquid/liquid interfaces (*Figure 1*). The three layers are confined in a shallow rectangular cavity which is differentially heated. The differential heating produces a temperature gradient parallel to the two interfaces. These investigations are in support of a space flight experiment, to be performed aboard the International Microgravity Laboratory IML-2.

Recently, radiosopic techniques have been presented as viable methods to track liquid metal flows, melt/gas interfaces, and melt/crystal interfaces [15-23]. The potential of radiosopic techniques for fluid dynamics and solidification studies has been well delineated by these studies.

We report on new radiosopic studies on melting and solidification of mono-component gallium under natural convection conditions. The effect of natural convective flow on the shape of the melt front is discussed. This work seeks further understanding of liquid encapsulated convection flows by analytically and numerically modeling convective flows in encapsulated gallium layers as preparation to experiments planned in the near future. Our main emphasis is the velocity and density fields in both layers which will be the primary data in the forthcoming experiments.

Since X-ray absorption is a function of material density, it was proposed that the difference in absorption from the hot and cold liquids could provide sufficient variation in intensity levels to be detected by the radiosopic image processing system. The percent density difference which must be detected for visualization of constant density fields in liquid gallium with an applied temperature difference of 8° C is calculated to be about  $\Delta\rho = 0.2\%$ .

#### NUMERICAL SIMULATION AND ANALYTICAL STUDIES

The multilayer fluid systems are numerically simulated using the commercial finite-element computer code FIDAP. Simulation of the experimental system is restricted to a 2-D model. The natural convection cases were extensively studied with asymptotic methods.

Rayleigh-Bénard convection is considered to further analyze the coupling between immiscible liquids (*Figure 2*). By visualizing the temperature field in double layers confined in a narrow slot, we have observed the two modes of viscous and thermal coupling near the onset of convection, and beyond. Generally, the observations are found to be in remarkable agreement with 2-D numerical simulations. The experiments also showed that onset of convection was in the form of steady flow. Oscillatory convection at onset was not detected from the thermocouple signals.

Analytical and numerical models have been compared for flows in encapsulated gallium melts. The analytical model is a useful tool for quickly obtaining a basic understanding of the general flow field and assessing the underlying physics. The obtained data will serve as a basis for liquid metal flow studies in a layer of gallium encapsulated by glycerol. Liquid encapsulation of gallium reduces the flow velocities to the level of a rigidly contained gallium layer. Flow velocities in the viscous encapsulant are negligibly small. The

liquid encapsulant acts essentially as a solid; the flow velocity at the interface is close to zero. The superiority of glass lining of ampoules and encapsulation comes with the absence of corrosion of electronic melts, control of volatiles, and barrier to contaminants in the surrounding atmosphere.

Thermocapillary convection is studied analytically in a two-dimensional layer of GaAs which is symmetrically encapsulated by liquid  $B_2O_3$ . In a space based reduced gravity environment thermal convection is primarily due to thermocapillary stresses at free surfaces and interfaces. The free surface tension gradient of  $B_2O_3$  is parameterized and its influence along with the influence of varying encapsulant layer thickness on flow in the GaAs melt is investigated. The results are compared with those for rigidly contained encapsulant layers. The free surface and the interface are considered to be deformable. The analytical model is benchmarked with a numerical simulation which confirms the validity and accuracy of the model.

Within the bounds of validity of the analytical model, it is found that thermocapillary flow in the GaAs melt can be significantly controlled by the encapsulant liquid. A 'near-halt-condition' or 'quasi-no-flow' condition in the melt can be achieved if the ratio of the free surface tension gradient (of  $B_2O_3$ ) to the interface tension gradient (of  $B_2O_3$ /GaAs) is around 2.3. Experimental data on the surface tension gradient of boron oxide, when available, will show how closely  $B_2O_3$  encapsulation approaches the theoretically required surface tension gradient ratio for a no-flow condition. A thin encapsulant layer is found to be also helpful in reducing the flow in the melt layer. Also, it is observed that a free outer encapsulant surface is more effective in reducing convective flow in the electronic melt layer than a liquid glass lined ampoule with rigid containment.

Preliminary studies were done to model effects of density inversion on the flow pattern in convective situations. It is well known that some fluids exhibit a density inversion behavior in a certain temperature range. A common example is water which possess a maximum density at  $3.98^\circ\text{C}$  at standard conditions. Others include liquid helium in the range of temperature about 2.178 K and the pseudobinary alloy  $Hg_{1-x}Cd_xTe$  in the range of temperature 1028 K. The single layer studies are currently extended to double liquid layers with density inversion occurring in the lower layer.

## EXPERIMENTS

Experiments are performed in a two-layer system, such as 10 cSt silicone oil over a fluorinert FC-70 liquid. Other silicone oils and fluorinerts are also used. Both the natural convection cases and the Rayleigh-Bénard cases are of interest.

The flow field is visualized with the aid of tracer particles suspended in the fluids. A streakline, if the exposure time is not very long, depicts the local velocity vector at the particle location. Velocity measurements are performed with a Laser Doppler Velocimetry (LDV) system and particle displacement tracking velocimetry (PDTV). Real time holographic interferometry is used to obtain the temperature field. In addition to the visualization of the flow field, we also measured the surface tension of the two liquids under investigation, and

the interface tension between them as a function of temperature. A radiosopic facility has been developed to visualize melting and solidification of opaque metals. The system consists of a modified non-destructive testing facility with 160 kV tungsten X-ray source, image intensifier, image acquisition and processing system, temperature data acquisition system, and heaters and controllers.

*Figure 3* shows a visualization of melting gallium under natural convection. The lighter portion represents the solid gallium. Liquid is visualized by the dark area. Because solid gallium has a 3% lower density than liquid gallium, the solid is more transparent to X-rays. As the absorption is proportional to the density, density changes are visualized as intensity changes. Using background subtraction image processing techniques, the small density change can be visualized as a black and white transition with a steep linear gray scale covering the 3% density change across the interface line. This high resolution permits visualization of corrugated interfaces as a function of time.

The method has now been improved to permit coarse visualization of the density fields within liquid gallium. When a fluid is differentially heated, the density of the fluid varies as a function of temperature. Since X-ray absorption is a function of material density, the difference in absorption from the hot and cold liquids provides sufficient variation in intensity levels to be detected by the radiosopic image processing system. The resolution of 0.2% density change has been achieved to date (*Figure 4*). It is confirmed that natural convection has a major influence on the shape of the solid/liquid interface.

A vertical Bridgman furnace is currently in the final stages of development. Visualization of density fields, solidification fronts and flow patterns in InSb are planned for the near future. Further exploratory studies were made for natural convection in encapsulated liquids undergoing solidification, and with liquids that exhibit miscibility gaps.

## CONCLUSION

A comprehensive study has been made on the convective fluid dynamics of multiple immiscible liquid layers. Thermal coupling and viscous coupling have been demonstrated in several cases. Oscillatory coupling has not been observed so far. It is found that the argument of interface contamination has been used too deliberately in the past. New measuring techniques have been developed to study liquid metals fluid dynamics. Flow patterns and density fields can now be observed in opaque melts.

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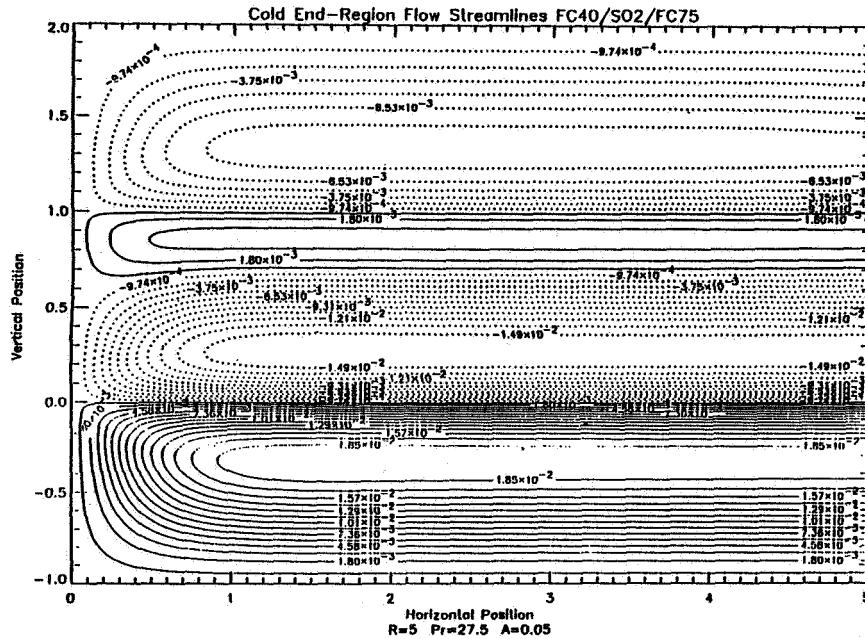


Figure 1: Spaceflight configuration: FC-40 /SO-2/FC-75

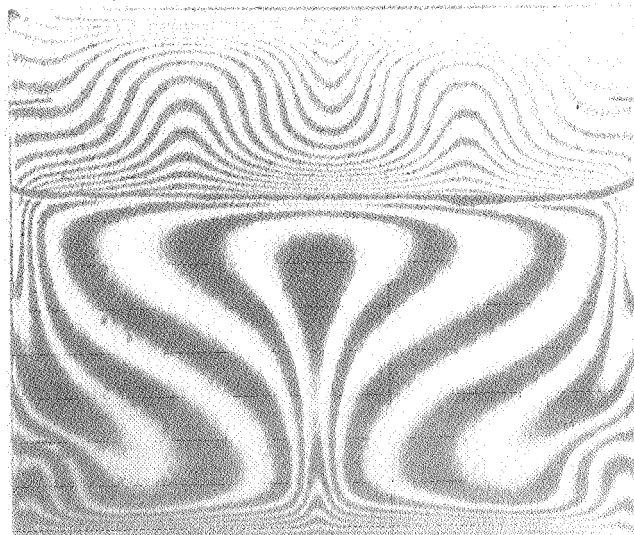


Figure 2 Silicone oil 100 cSt / Ethylene Glycol,  $\Delta T = 12$  K

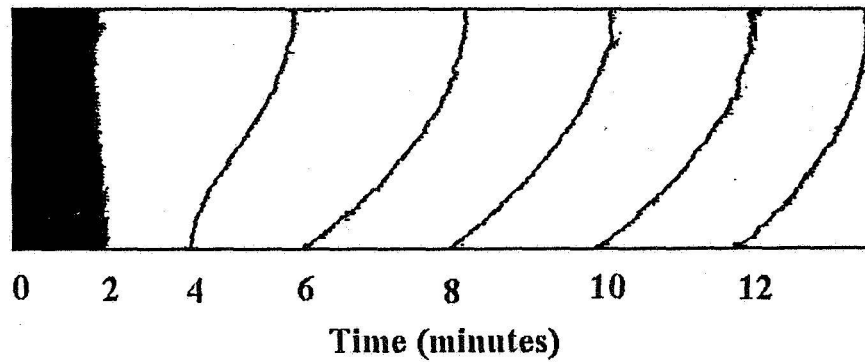


Figure 3: Natural convection in liquid gallium. Advancing melting front as a function of time. Grashof number  $Gr = 1.1 \times 10^6$

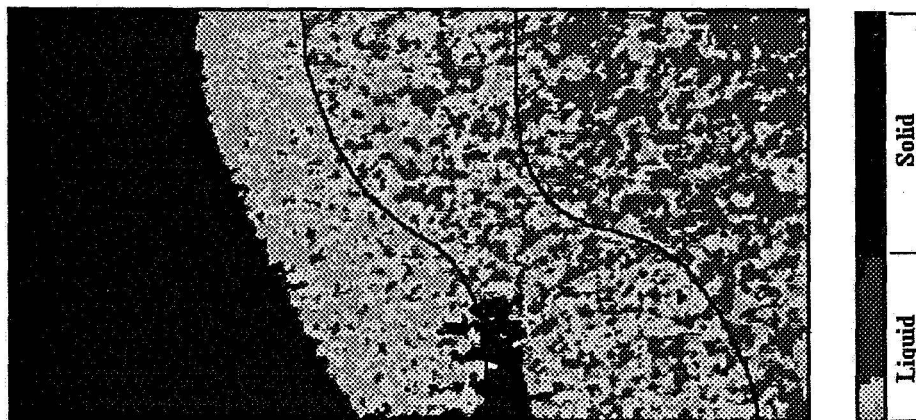


Figure 4: Isodensity fields in 1.25 cm layer of liquid Gallium subject to natural convection and solidification.