FLUID MECHANICS OF DIRECTIONAL SOLIDIFICATION

AT REDUCED GRAVITY

1. ABSTRACT

The primary objective of the proposed research is to provide additional ground-based support for the flight experiment "Casting and Solidification Technology" (CAST), with principal investigators Drs. M. H. McCay and T. D. McCay of UTSI. This experiment is to be performed in IML-1, scheduled to be flown in a space shuttle mission scheduled for 1992. In particular, we will provide data on the convective motion and freckle formation during directional solidification of NH_4Cl from its aqueous solution at simulated parameter ranges equivalent to reducing the gravity from the sea-level value down to 0.1 g or lower. The secondary objectives of the proposed research are to examine the stability phenomena associated with the onset of freckles and the mechanisms for their subsequent growth and decline (to eventual demise of some) by state-of-the-art imaging techniques and to formulate mathematical models for the prediction of the observed phenomena.

This project was funded by NASA-MSAD starting 1 April 1991 at $75,000 per year for three years. We have made excellent progress during the first grant year, and we are requesting continuing support for the second year.

2. PROJECT DESCRIPTION

During the first year of the grant period, we accomplished a number of the objectives stated in the original proposal and examined a number of interesting problems related to material processing in space. We will summarize our research results first, then present a description of the tasks to be accomplished in the second year.

2.1. *Progress to Date*

2.1.1 *Comprehensive Experimental Results on the Directional Solidification of NH_4Cl Solution*

This work was started under the previous grant, NAG 3-723. The results are presented in Chen and Chen (1991), which appeared in June. A solution of 26% NH_4Cl-H_2O was directionally solidified by cooling it from below with a constant-temperature surface ranging from -31.5^0C to $+11.9^oC$. It was found that finger convection occurred in the fluid region just above the mushy layer in all experiments. Plume convection with associated chimneys in the mush occurred in experiments with bottom temperatures as high as $+11.0^oC$. However, when the bottom temperature was raised to $+11.9^oC$, no plume convection was observed, although finger convection continued as usual. These results indicate that plume convection could occur at $10^{-1}g$, but finger convection could occur at $10^{-4}g$, the residual gravity level in orbiting space shuttles. A method has been devised to determine the porosity of the mush by computed tomography. Using the mean value of the porosity across the mush layer and the permeability calculated by the Kozeny-Carman relationship, the critical Rayleigh number across the mush layer for onset of plume convection was estimated to be between 200 and 250.

2.1.2 *Flow Visualization Study of Convection in the Mush*

During the months of June and July of 1991, Dr. Chen was a Visiting Fellow at the Research School of Earth Sciences, Australian National University. While there, he carried out experiments on directional solidification of NH_4Cl in the Geophysical Fluid Dynamics Laboratory of Professor J. S. Turner. Using flow visualization techniques and time-lapse motion pictures, he was able to record the convective motion in the mushy zone before and after the onset of plume convection. The results show that prior to the onset of plume convection, there was no convective motion within the mushy zone, even

through there was vigorous finger convection in the liquid region right above the mush. After the onset of plume convection, the convection in the mush consisted of a general downward motion to the bottom of the tank and ejection of the fluid through the plumes. At the fully developed plume convection state, the downward velocity was measured to be 0.44 mm/min.

A directional solidification experiment with NH_4Cl solution was also done in a Hele-Shaw cell, 150 × 220 × 1 mm. The solidification process is similar to that in a three-dimensional tank but at a slower pace. Because of the small thickness of the tank, the formation of the chimney and the flow in the chimney are clearly visible. From visual observations and from a time-lapse film made (at 3 sec/frame) during the experiment, the following results were obtained: (i) At the early stage of the mushy growth, the upward-convecting fluid from the mush contains tiny crystals of NH_4Cl. These crystals become nuclei for further crystallization. Eventually, they become heavy enough to drop back to the mush. This phenomenon was subsequently observed in a three-dimensional tank. (ii) Tiny crystals are also present in the upward-flowing plumes in the chimneys. Because of remelting, there are pieces of crystals, broken off the chimney wall, descending in the plume. Some of these became lodged in the crevices in the wall, making the periphery of the chimney more densely packed, as seen in the CT scan (Chen and Chen 1991). (iii) Chimneys are established from the top of the mush downward.

To investigate the effect of increased viscosity on the directional solidification of NH_4Cl, small amounts of "Natrosol" (hydroxyethylcellulose) were added to the solution. According to Tait and Jaupart (1989), this would only increase the viscosity of the solution and would not affect its phase diagram. The results show that as the viscosity is increased, convection becomes less vigorous. When the viscosity is increased 30-fold, the dendrites are very sparsely packed and there is no chimney. The results are summarized in a poster entitled "Viscosity Effects on the Directional Solidification

of NH_4Cl Solution in a Hele-Shaw Cell," which was exhibited in the Gallery of Fluid Motions at the 44th Annual Meeting of the Division of Fluid Dynamics, American Physical Society, held in Scottsdale, Arizona, November 24-26, 1991. This poster was awarded a prize, which consists of publication in the journal of *Physics of Fluids A* in September 1992.

2.1.3 *Imaging Technique*

During the past few months, Dr. Chen has been working with Dr. Art Guitro of the Magnetic Resonance Imaging Facility, Arizona Health Sciences Center, to obtain MRI scans of the mushy zone resulting from directional solidification of NH_4Cl solution. There were two reasons for switching from CAT scans to MRI scans. One was that a research MRI facility that serves no clinical function was available. Therefore, it was easier to schedule scans of the solidifying sample. The other was that by using MRI, the motion of the fluid within the mush might be visualized. After a number of trials, it was determined that due to the high conductivity of the 26% NH_4Cl solution, the resulting images are not sharp enough to obtain any quantitative information. This approach has been abandoned, and we will be concentrating on CAT scans.

2.1.4 *Effect of Gravity Modulation on the Stability of a Double-Diffusive Layer*

The effect of gravity modulation on the onset of instabilities in a double-diffusive layer with cross-diffusion has been analyzed using linear stability analysis with Floquet theory. Results show fundamentally different features in the topology of neutral curves and stability boundaries not found in singly diffusive layers under g-jitter. Among the more important findings are: (1) The existence of neutrally stable periodic solutions that are neither synchronous nor subharmonic. (2) Neutral curves exhibiting multiple bifurcation points connecting three different classes of neutral solutions. (3) The existence of two onset incommensurate frequencies at two different wave numbers for

the same Rayleigh number. This work is part of the Ph.D. dissertation by Dr. Guillermo Terrones, with Dr. Chen as his thesis advisor. A paper on this subject will be presented by Dr. Terrones at the AIAA Aerospace Sciences Meeting in January 1992.

2.1.5 *Effect of Surface Tension on the Stability of a Double-Diffusive Layer*

Since salt-finger convection is likely to occur at gravity levels as low as 10^{-4} g, it is important for us to study the effect of the interaction between surface tension and the double-diffusive phenomenon. Such a study has been carried out using linear stability analysis. The major conclusions are: (i) A low level of gravity may damp out oscillatory instability or may alter its dynamics so that the instability appears as steady convection. (ii) The onset of salt-finger instability may be in the overstable mode due to the surface tension effects. (iii) The Lewis and Prandtl numbers of the material have a profound effect on the stability boundaries and onset characteristics of the system. This work will be presented by Dr. Chen at the AIAA Aerospace Sciences Meeting in January 1992.

2.1.6 *Salt-Finger Convection Under a Gravity Gradient*

It has been shown that salt-finger convection under a constant gravity gradient is exactly analogous to the small-gap Taylor-Couette instability. This analogy was used to obtain the marginal stability conditions for such a system. The results will be published in the *Physics of Fluids A* in February 1992.

2.2. *Publications and Presentations Acknowledging the Current Grant*

Chen, C. F., "Directional Solidification of NH_4Cl Solution in a Hele-Shaw Cell," 44th Annual Meeting of the Division of Fluid Dynamics, American Physical Society, Scottsdale, Arizona, November 1991.

Chen, C. F., "Surface Tension Effects on the Onset of Double-Diffusive Convection," IUTAM Symposium on Microgravity Fluid Mechanics, Bremen, Germany, September 2-6, 1991 (to appear in the *Proceedings*).

Chen, C. F., "Viscosity Effects on the Directional Solidification of NH_4Cl Solution in a Hele-Shaw Cell," Prize-Winning Poster, 44th Annual Meeting of the Division of Fluid Dynamics, American Physical Society, Scottsdale, Arizona, November 1991.

Chen, C. F. and F. Chen, "Onset of Salt-Finger Convection in a Gravity-Gradient," *Physics of Fluids* (to appear Feb. 1992).

Chen, C. F. and T. F. Su, "Stability Analysis of Surface Tension Effects on a Double-Diffusive Layer," AIAA Paper 92-0605, AIAA 30th Aerospace Sciences Meeting, January 6-9, 1992.

Terrones, G., "Gravity Modulation and Cross-Diffusion Effects on the Onset of Multicomponent Convection," Ph.D. dissertation, The University of Arizona, Tucson, November 1991.

Terrones, G. and C. F. Chen, "G-Jitter and Soret Effect on the Onset of Convection in a Doubly Diffusive System," AIAA Paper 92-0246, AIAA 30th Aerospace Sciences Meeting, January 6-9, 1992.

2.3. *Work to Be Performed During the Second Year*

2.3.1 *Detailed Examination of the Mushy Zone by CAT Scan*

The mushy zone will be scanned at frequent intervals during the directional solidification process. This will yield information on the spatial and temporal evolution of the porosity distribution within the mush. The condition of the mushy zone just prior to the onset of plume convection

2.3.2 *Experiments of Directional Solidification with Initial Temperature Gradient*

The CAST experiment is conducted under a high initial temperature gradient in the NH_4Cl solution (McCay et al. 1988). It is important to obtain further ground-based data, especially on the onset conditions for plume convection.

2.3.3 *Directional Solidification in a Hele-Shaw Cell*

The preliminary experimental results of directional solidification in a Hele-Shaw cell showed that much can be learned from this simple apparatus. We plan to carry out a series of experiments with systematic variations of the bottom temperature. The effect of such variations on the onset characteristics of chimney and plume convection will be examined.

2.4. *References*

Chen, C. F. and Chen, F., 1991, "Experimental Study of Directional Solidification of Aqueous Ammonium Chloride Solution," *J. Fluid Mech.*, 227, 567-586.

McCay, T. D., McCay, M. H., Lowry, S. A., and Smith, L. M., 1989, "Convective Instabilities During Directional Solidification," *J. Thermophysics and Heat Transfer*, 3, 345-350.

Tait, S. and Jaupart, C., 1989, "Compositional Convection in Viscous Melts," *Nature*, 338, 571-574.

3. PERSONNEL

The Principal Investigator of this project is:

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A short biographical sketch is presented in the following.

Education

University of Illinois, B.S.M.E., 1953, Class Salutatorian
University of Illinois, M.S.M.E., 1954
Brown University, Ph.D., 1960; Dissertation Title: "Theoretical and Experimental Study of Configurations Composed of a Body Under a Lifting Wing in Supersonic Flow"

Experience

University of Arizona, Department of Aerospace and Mechanical Engineering
Professor, 1980–present
Head of Department, 1980–1989

The Australian National University, Institute of Advanced Study, Research School of Earth Sciences
Visiting Fellow, Summer 1978
Research: Crystallization in double-diffusive systems
Visiting Fellow, Summer 1991
Research: Convection in the mushy zone

University of Cambridge, Department of Applied Mathematics and Theoretical Physics
Senior Visitor, 1971–72
Research: Double-diffusive phenomena
Senior Visitor and Visiting Fellow, Clare Hall, Michaelmas term, 1987
Research: Freckle formation in directionally solidified binary alloys

Rutgers University, Department of Mechanical, Industrial, and Aerospace Engineering
Department Chairman, 1976–80
Professor II, 1973–80
Professor I, 1969–73
Associate Professor, 1966–69
Assistant Professor, 1963–66

Hydronautics, Incorporated, Laurel, Maryland
Assistant to the Chief Engineer, 1962–63
Senior Research Scientist, 1961–62
Research Scientist, 1960–61
Theoretical Research: Flows about supercavitating hydrofoils and propellors, water entry phenomena, stratified fluids

Brown University, Research Assistant in Engineering, 1955–60

University of Illinois, Research Assistant in Mechanical Engineering, 1953–55

Publications (since 1986)

"Stability Analysis on the Convection of a Variable Viscosity Fluid in an Infinite Vertical Slot," with S. Thangam, *Phys. Fluids* 29:1367–1372, 1986.

"Stability of Thermal Convection in a Vertical Porous Layer," with L. P. Kwok, *J. Heat Transfer* 109:889–893, 1987.

"Double-Diffusive Effects During Solidification" (Invited), pp. 527–540 in *Interdisciplinary Issues in Materials Processing and Manufacturing*, Samanta et al. (eds.), Vol. 2, ASME, 1987.

"Onset of Finger Convection in a Horizontal Porous Layer Underlying a Fluid Layer," with Falin Chen, *J. Heat Transfer* 110:403–409, 1988.

"Double-Diffusive Convection in a Porous Medium," with B. T. Murray, *J. Fluid Mechanics* 201:147-166, 1989.

"Experimental Investigation of Convective Instability in a Superposed Fluid and Porous Layer When Heated From Below," with Falin Chen, *J. Fluid Mech.* 207:311-321, 1989.

"Salt Finger Convection Under Reduced Gravity," AIAA Paper 90-0122.

"Double-Diffusive Convection and Its Effects Under Reduced Gravity," in *Low Gravity Fluid Dynamics and Transport Phenomena* (Koster and Sani, Eds.), pp. 355-366, AIAA, Washington, D.C., 1990.

"Convective Instability in Superposed Fluid and Anisotropic Porous Layers," with Falin Chen and A. J. Pearlstein, *Phys. Fluids A* 3:556-565, 1991.

"Experimental Study of Directional Solidification of Aqueous Ammonium Chloride Solution," with Falin Chen, *J. Fluid Mech.* 227:567-586, 1991.

"Surface Tension Effects on the Onset of Double-Diffusive Convection," *Microgravity Sci. and Tech.* 4:108-109, 1991.

"Convection in Superposed Fluid and Porous Layers," with Falin Chen, *J. Fluid Mech.* 234:97-119, 1992.

"Stability Analysis of Surface Tension Effects on a Double-Diffusive Layer," with T.-F. Su, AIAA Paper 92-0605.

"Effects of G-Jitter and Cross Diffusion on the Onset of Convection in Doubly Diffusive Systems," with G. Terrones, AIAA Paper 92-0246.

"Onset of Salt Finger Convection in a Gravity Gradient," with Falin Chen, *Phys. Fluids A* (in press).

4. FACILITIES AND EQUIPMENT

In the Double-Diffusive Convection Laboratory, there are two Lauda constant-temperature circulators, one of which can maintain a low temperature of -40°C, and one Haake constant-temperature circulator. There is a Fluke Datalogger with 60 input channels for temperature reading. The Department has a modern machine shop capable of constructing test cells for the proposed experiments.

There are two CAT scan machines in the Medical Imagining Laboratory of the Arizona Health Sciences Center.

5. OTHER SUPPORT

Dr. Chen is the Principal Investigator of NASA Grant NAG3-1268, "Fluid Mechanics of Directional Solidification at Reduced Gravity, for three years, April 1, 1991 through March 31, 1994, at $75,000/year. Dr. Chen is supported for two summer months each year.

Dr. Chen is the Principal Investigator of a proposal in response to NRA-91-OSSA-17 entitled "Marangoni and Double-Diffusive Convection in a Fluid Layer Under Mirogravity" with Dr. Cho Lik Chan as Co-Investigator. The budget is $301,120 for three years.

6. BUDGET

PROPOSED BUDGET - NASA
April 1, 1992 - March 31, 1993
Year 2

Direct Labor

C. F. Chen, Principal Investigator		
2.0 Months Summer		$ 20,529
Graduate Research Assistant		
50.0% Academic Year		12,915
1.5 Months Summer		4,308
	Total Direct Labor	$ 37,752

Employee Related Expenses

18.9% Faculty		$ 3,880
1.5% Student		258
	Total ERE	$ 4,138

Operations

Expendable Supplies		$ 2,245
Publications		1,000
Communications		500
CT Scan - 110 hours @ $20/hour		2,200
	Total Operations	$ 5,945

Travel

Domestic Meetings and one Trip to UTSI		$ 2,500
	Total Travel	$ 2,500
	Total Direct Costs	$ 50,335

Indirect Costs

49.0% MTDC	Total Indirect Costs	$ 24,665
	Total Direct and Indirect Costs Year 2	**$ 75,000**