

Effect of Marangoni Convection Generated by Voids on Segregation during Low-G and 1-G Solidification

M. Kassemi¹, A. Fripp², N. Rashidnia¹, and H. de Groh³, ¹NCMR, NASA LeRC, Cleveland OH, ²NASA LaRC, Hampton, VA, ³NASA LeRC, Cleveland OH

1. Introduction and Objective

Solidification experiments, especially microgravity solidification experiments are often hampered by the evolution of unwanted voids or bubbles in the melt. Although these voids and/or bubbles are highly undesirable, there are currently no effective means of preventing their formation or eliminating their adverse effects, particularly, during low-g experiments. Marangoni Convection caused by these voids can drastically change the transport processes in the melt and, therefore, introduce enormous difficulties in interpreting the results of the space investigations. Recent microgravity experiments by Matthiesen (Study of Dopant Segregation Behavior During the Growth of GaAs in Microgravity, 1994), Andrews (Coupled Growth in Aluminum-Indium HyperMonotectics, 1997) and Fripp (Lead-Tin-Telluride, Growth in a Low-G Environment, 1997) are all good examples of how the presence of voids and bubbles affect the outcome of costly space experiments and significantly increase the level of difficulty in interpreting their results.

In this work we examine mixing caused by Marangoni convection generated by voids and bubbles in the melt during both 1-g and low-g solidification experiments. The objective of the research is to perform a detailed and comprehensive combined numerical-experimental study of Marangoni convection caused by voids during the solidification process and to show how it can affect segregation and growth conditions by modifying the flow, temperature, and species concentration fields in the melt.

While Marangoni convection generated by bubbles and voids in the melt can lead to rapid mixing that would negate the benefits of microgravity processing, it could be exploited in some terrestrial processing to ensure effective communication between a melt/solid interface and a gas phase stoichiometry control zone. Thus we hope that this study will not only aid us in interpreting the results of microgravity solidification experiments hampered by voids and bubbles but to guide us in devising possible means of minimizing the adverse effects of Marangoni convection in future space experiments or of exploiting its beneficial mixing features in ground-based solidification.

2. Background and Microgravity Relevance

The results of the PbSnTe solidification experiment on USMP3 (Fripp, 1997) together with the preliminary numerical simulations of thermocapillary convection generated by bubbles in both 1g and low-g environments (Kassemi and Rashidnia, 1996 and 1997) are good examples of how creation of voids or bubbles in the melt during the solidification process can affect the outcome of costly space experiments.

2.1 Lead-Tin-Telluride Low-G Growth Experiment

In the Lead-Tin-Telluride low-g growth experiment, crystals were grown using the Bridgman technique in AADSF facility. During the flight, three separate crystals were processed in a single,

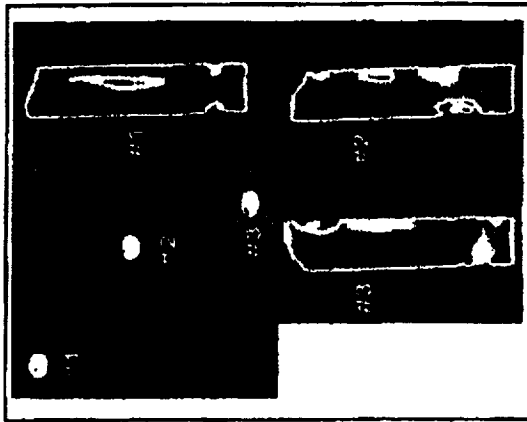


Figure 1. CAT Images of Void Formation in The Three Lead-Tin-Telluride Samples.

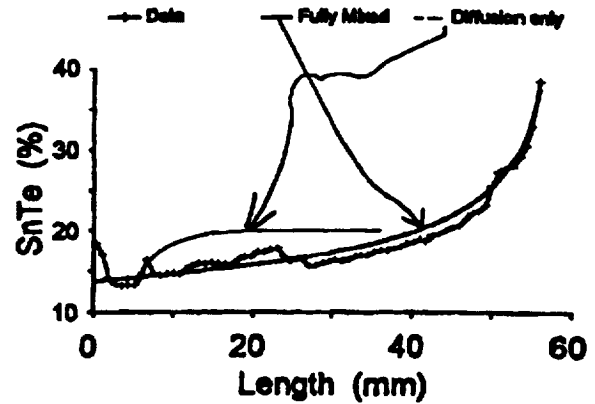


Figure 2. Fripp's Experimental Composition Profile for Solidification of PbSnTe in Cell #1 (Top-Heated). The Fully-Mixed and Diffusion-Only Theoretical Profiles Are Also Shown.

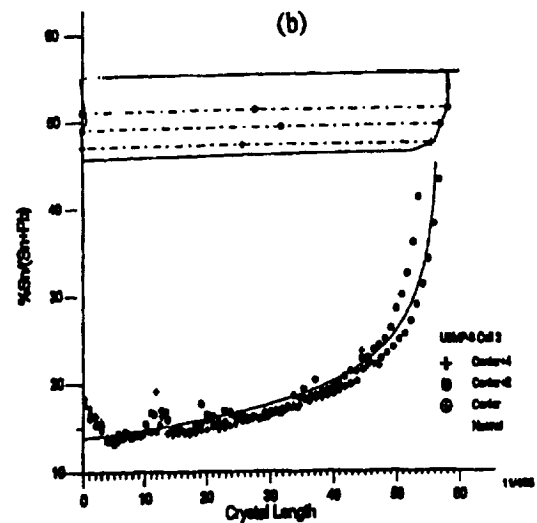
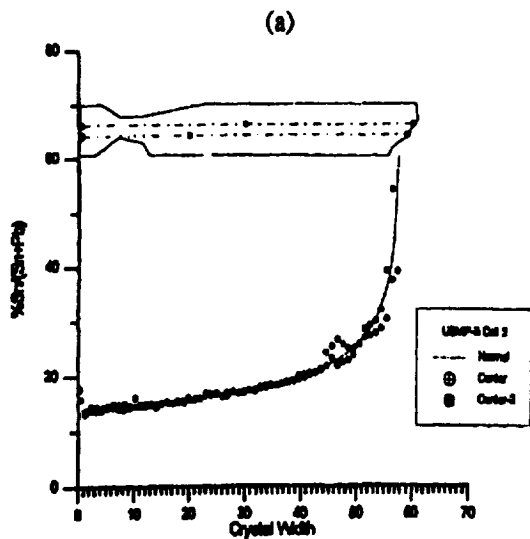


Figure 3. Fripp's Experimental Composition Profiles for Directional Solidification of PbSnTe in a) Cells #2 (Top-Cold) and b) #3 (Horizontal).

segmented ampoule. The crystals were grown in series, each in one of the three primary orientations with respect to the residual gravity vector. The growths were roughly analogous to hot-on-top, cold-on-top, and horizontal configuration. While the immediate objective of the experiment was to grow PbSnTe and establish its fundamental growth properties, another, more important objective was to gain a better understanding of the mechanisms involved in the generalized crystal growth process, particularly those affected by gravity-driven convection caused by temperature and/or solutal gradients.

The flight sample was retrieved from the AADSF at the Kennedy Space Center (KSC) in April 1996. The samples were examined, still in the Inconel cartridge, with a computer aided tomography (CAT) unit available in the KSC non-destructive test laboratory. These high voltage x-rays are capable of penetrating the PbSnTe samples and exposing any voids, large pits, and bubbles.

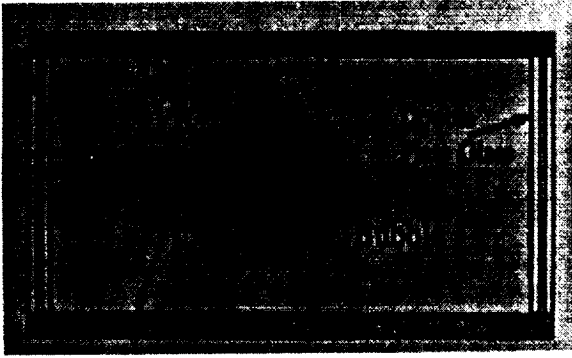


Figure 4. Schematic of The Test Enclosure

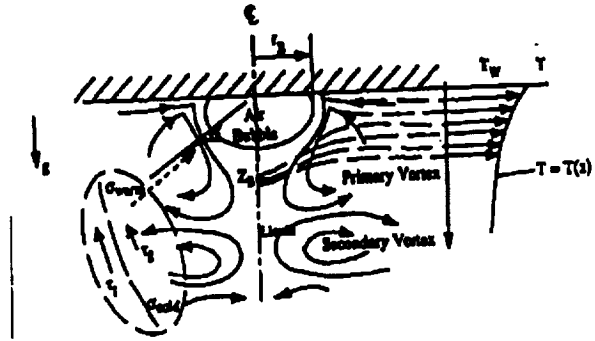


Figure 5. Bubble-Induced Thermocapillary and Natural Convective Flows

The results were totally unexpected; the crystals were cratered with large voids and riddled with meandering channels as shown in Fig. 1.

The primary objective of this flight experiment was to examine the effect of the direction of the microgravity vector on the convective mixing in the liquid during directional solidification. An excellent measurable physical parameter and a sensitive monitor of mixing in the liquid is the compositional profile in the solidified crystal. The axial compositional profiles for cells #1 and #2 and #3 are given in Figs. 2 and 3 respectively. Each cell shows evidence of considerable mixing. Fig. 2 shows the axial compositional distribution of cell #1. The anticipated spike of SnTe shows at the recalescence area at the left side of the plot, then the data flattens for what may be growth during thermal stabilization after the release of the latent heat. At 5 mm of growth, approximately the length of the produced seed after the solutal diffusion time, the composition shows signs of trying to go to diffusion growth but not making it. At approximately 25 mm of growth the curve closely approximates the fully mixed case. The axial compositional profile of the crystal in cell #2, the nominally hot-on-bottom orientation, follows the completely mixed curve for the first 45 mm of growth (after recalescence) and then exhibits deviations (see Fig. 3) that are, as yet, unexplained. Similar deviations occur in the axial compositional profile of the crystal grown in cell #3, but over a longer section (see Fig. 3). As yet, no acceleration perturbations have been related to these compositional variations. Unfortunately, any differences in convective mixing due to the alignment of the acceleration vector were also indistinguishable due to the *vigorous mixing brought about by Marangoni convection which is independent of orientation with respect to the gravity vector.*

2.2 Thermocapillary Convection Caused by a Bubble

To get a preliminary assessment of Marangoni convection caused by a bubble or void in the surrounding fluid consider an enclosure containing a liquid (silicone oil) with a bubble placed on the inside of the top wall as shown in Fig. 4. The side walls are insulated and the temperature of the top and bottom walls are uniformly maintained at T_h and T_c , respectively. Therefore, a thermally stratified state is established in the enclosure before the bubble is introduced. Once the bubble is positioned and the interface between the air and test liquid is formed, surface tension forces created by the temperature gradient along the interface will drive a thermocapillary convective flow as sketched in Fig. 4.

1-G Results: Numerical simulations shown in Fig. 6 indicate that in 1-g, this thermocapillary flow will disrupt the thermal stratification near the bubble resulting in significant temperature gradients near the bubble surface. As the thermocapillary convection gains strength, it will move the hot flu-

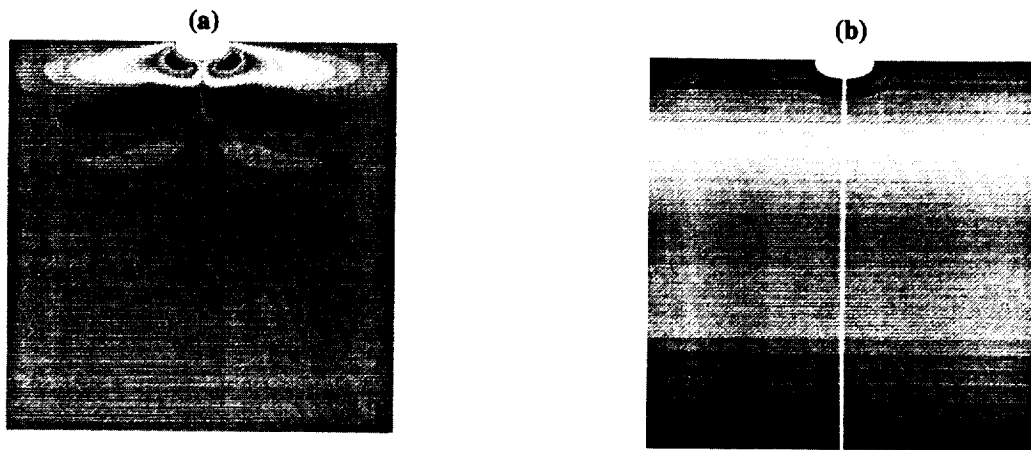


Figure 6. 1-G Numerical Predictions of Streamlines (a), Temperature Contours (b), and Bubble Shape for $Ma=2440$, $Ra=500$, $Pr= 122$.

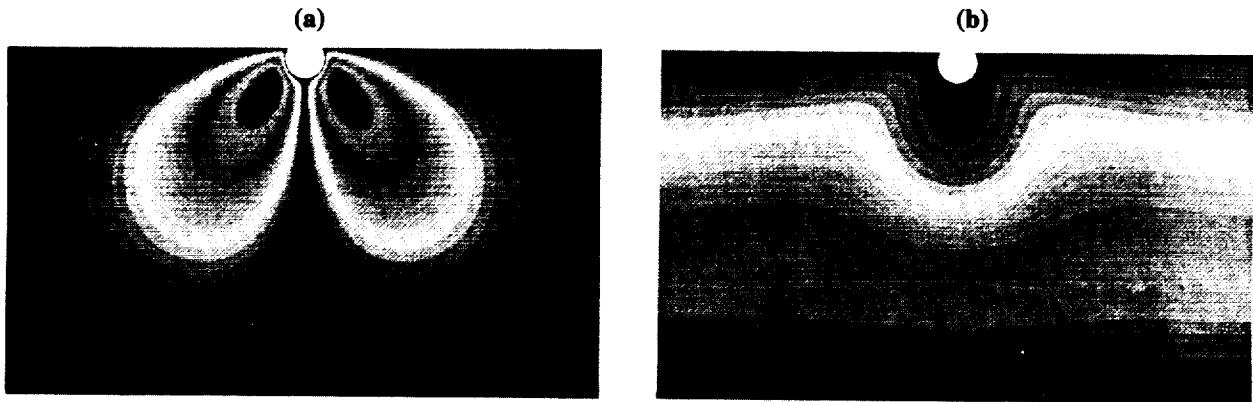


Figure 7. Low-G Numerical Predictions of Streamlines (a), Temperature Contours (b), and Bubble Shape for $Ma=1830$, $Ra=0.25$, $Pr= 122$.

id from the top of the cavity into the colder region below the bubble, thereby, modifying the temperature field in the lower section of the enclosure and resulting in horizontal temperature and density gradients. On earth, these density gradients will give rise to buoyancy-driven convection which will actually counteract the Marangoni flow. For small imposed vertical temperature differences, a steady-state condition will finally prevail where the vortices generated by the two counteracting forces co-exist and deform the temperature contours near the bubble as shown in Fig 6a and 6b. For larger temperature differences the fluid flow and temperature fields can go through a series of oscillatory modes which have been discussed in Kassemi and Rashidnia (1996 and 1997)

Low-G Results: Since, a low gravity environment is characterized by a reduced buoyancy force, decreased hydrostatic head, and negligible natural convection, the low-g shape of the bubble and the resulting thermocapillary flow and temperature fields will be also significantly different as shown in Fig. 7. In this case, just as in the previous situation, a vigorous thermocapillary flow is generated next to the bubble surface. This strong flow will drastically modify the temperature profiles in the enclosure. But in contrast to the terrestrial examples presented in Fig. 6, this time, a natural convective flow will not ensue due to the reduced buoyancy force. As a result, the recircu-

lating thermocapillary vortex will grow unopposed until it nearly fills the entire enclosure at steady-state. The streamlines of Fig. 7a clearly show that the microgravity flow pattern resembles a jet-like flow emanating from around the bubble and flowing downwards into the enclosure. As a result of this intense recirculating flow, the temperature field is greatly altered as depicted in Fig 7b. *Thus, in microgravity, the shape of the bubble, and the temperature and fluid flow fields are all drastically different from their terrestrial counterparts and the unrestricted thermocapillary flow fills the entire enclosure.* In a typical solidification experiment, similar thermocapillary convection generated by a void or bubble can easily modify the temperature and velocity fields and the segregation pattern in the melt.

3. Proposed Work

The previous experimental and numerical results included in section 2.1 and 2.2 clearly underscore the following facts:

- Diffusion-Controlled growth was not possible in microgravity solidification of lead-tin-telluride and gallium arsenide when voids formed in the melt during the solidification process. This has been attributed to significant levels of mixing brought about by Marangoni convection generated by the voids.
- Preliminary work with stationary bubbles in a fluid with an imposed temperature gradient indicates that a vigorous Marangoni flow prevails in both low-g and 1-g environments. This modifies the flow structure and the temperature gradients drastically.
- The nature of convection in both 1-g and low-g depends on the magnitude of the Ma number (dictated by void size, temperature and/or concentration gradients, melt Thermophysical properties). At high Ma numbers the thermocapillary flow is time-dependent in both low-g and 1-g environments and could lead to periodic symmetric and asymmetric oscillatory modes through interactions with buoyancy.
- In the absence of buoyancy force, the Marangoni flow can grow unrestricted to affect the transport process in the entire domain.
- There is a serious gap in our knowledge with regard to bubble-induced steady and oscillatory flows and their effect on temperature and concentration fields in the melt and interface stability during 1-g and low-g solidification.
- Preliminary numerical predictions indicate that the conclusions obtained from ground-based experiments with regard to oscillatory convection generated by bubbles and its possible effects on solidification cannot be directly extrapolated to predict the behavior and effect of the Marangoni flows in space and future microgravity experiments to directly study this phenomenon are necessary.

In the context of these preliminary findings and observations, the present research program consists of two phases. The first phase has four major components:

1. Solidification experiments with SCN to directly investigate: a) the effects of Marangoni convection on the temperature, velocity, and concentration fields in the melt; and b) their impact on growth conditions and stability of the solidification front.
2. Post growth characterization of the solid to determine levels of segregation and extent of mixing.

3. Controlled flow and temperature visualization experiments with different grades of silicone oils to determine the Pr number effect which is needed for extrapolating the results of the SCN experiment to growth of low Pr (high conductivity) number materials e.g. metals and semiconductors.
4. Development of a comprehensive numerical model and simulation of the entire solidification process.

The second phase of this work is concerned with space experiments and has two major components which are as follows:

1. Numerical simulation of recent space experiments (Fripp and Matthiessen) so that existing data can be re-interpreted in terms of the separate roles played by surface tension-driven convection and buoyancy-driven convection due to residual acceleration.
2. Refine the experimental conditions, set-up, and procedures in order to design the *appropriate* space experiment and define the *optimum* parametric space for studying the effects of bubbles and voids on microgravity solidification.

4. Closure

The study of Marangoni convection generated by voids or bubbles is not only warranted because of its fundamental scientific importance in understanding the behavior of fluid flow in space, but also because of its practical significance to solidification and materials processing experiments in both 1-g and low-g environments. During the two phases of this research, an experimental set-up and procedure will be devised to study the effects of Marangoni number on the temperature velocity and concentration fields. A comprehensive numerical model will be developed and tested against the ground-based results. The verified numerical model will be used to simulate previous low-g experiments and interpret their results. The numerical model will be also used to precisely identify the parametric range of interest for a future microgravity investigation. Finally, based on the knowledge accumulated during this ground-based research, a future long duration space experiment will be designed.

5. References

- Andrews, J. B., Hayes, L.J., Arikawa, Y., and, Coriell, S.R., *Microgravity Solidification of Al-In Alloys*, 35th Aerospace Sciences Meeting, AIAA, Reno, NV, January 6-10, 1997.
- Fripp, A.L., and et al., *The Effect of Microgravity Direction on the Growth of PbSnTe*, AIAA paper AIAA 97-0676, 1997
- Kassemi, M., and Rashidnia, N., *Steady and Oscillatory Thermocapillary Flows Generated by a Bubble in 1-G and Low-G Environments*, AIAA 97-0924, presented in the 35th AIAA Aerospace Conference, 1997.
- Kassemi, M., and Rashidnia, N., *Thermocapillary and Natural Convective Flows Generated by a Bubble in 1-G and Low-G Environments*, AIAA 96-0734, presented in the 34th AIAA Aerospace Conference 1996.
- Matthiessen D.H., and Majewski, J.A., *The Study of Dopant Segregation Behavior During the Growth of GaAs in Microgravity*, NASA Conference Publication 3272, Vol. 1, p 223, May 1994; Joint Launch + One Year Review of USML-1 and USMP-1 with the Microgravity Measurement Group