

# **Analysis of residual acceleration effects on transport and segregation during directional solidification of tin-bismuth in the MEPHISTO furnace facility**

**NAG3-1740**

## **Final Report**

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### **1.1 General**

The research accomplishments summarized in this Final Report refers to work funded under the grant NAG3-1740 *Analysis of residual acceleration effects on transport and segregation during directional solidification of tin-bismuth in the MEPHISTO furnace facility* during the period from 3/95 to 3/98, which included a 12 months no-cost extension granted at the end of the nominal 2 year period of performance. The report has 5 sections, in section 1 the objectives are presented, a task description is given and the background and significance of the work is outlined. In section 2 the research accomplishments are summarized. In section 3 publications and presentations are listed. Student participation is listed in 4. The work is summarized in section 5. and references for sections 1 and 2 are supplied in section 6.

### **1.2 Objectives:**

The object of this work, is to approach the problem of determining the transport conditions (and effects of residual acceleration) during the plane-front directional solidification of a tin-bismuth alloy under low gravity conditions. The work involved using a combination of 2- and 3-D numerical models, scaling analyses, 1D models and the results of ground-based and low-gravity experiments. The experiments conducted in the MEPHISTO furnace facility during the USMP-3 space flight which took place between February 22 through March 6, 1999). This experiment represents an unprecedented opportunity to make a quantitative correlation between residual accelerations and the response of an actual experimental solidification

### **1.3 Background and significance**

The MEPHISTO program is the result of a cooperative effort that involves the French nuclear and space agencies Commissariat à l'Energie Atomique, CEA - Centre National d'Etudes Spatiales, CNES) and the National Aeronautics and Space Administration (NASA). Six space flights on the USMP carrier were initially planned, with odd-numbered missions being the responsibility of the French scientific teams and the even-numbered missions being the responsibility of the US. During the first flight of MEPHISTO in October 1992, bismuth-doped tin samples (Sn:0.5 at.% Bi) were used and experiments were carried out both below and above

the morphological stability threshold. Exciting results were obtained [1] that opened new perspectives for research.

The first scientific objective of the USMP-3 flight was to investigate the g-jitter induced solutal segregation in planar front solidification. To do so, we requested well controlled gravity perturbations, mainly in the form of Primary Remote Control System (PRCS) burns. The other main scientific objective was to track the morphological stability threshold (i.e. the instability where the growth front goes from planar to cellular) with the finest accuracy possible. In comparison with the first flight, the USMP-3 alloys were slightly more concentrated (1.5% at. Bi), in order to check a possible soluto-convective effect. It should be noted that, in contrast, both the USMP-2 and USMP-4 MEPHISTO flights, (Professor Abbaschian, University of Florida, PI) featured bismuth based alloys with low tin concentrations. MEPHISTO is basically a directional solidification furnace, where three samples are simultaneously processed. An original point is that there are in fact two heating/cooling subsystems [2,3]; one of them is maintained at a fixed position to provide a reference interface, whereas the other is allowed to move for solidification and melting of the alloy. Sample #□1 is dedicated to a measurement of the Seebeck voltage between the two ends, meaning that the system acts as its own thermocouple, with a "cold" and a "hot" reference junction (respectively the moving and fixed interfaces). The Seebeck voltage is then a measure of the undercooling at the growth front, a key feature being that the signal is obtained in real time. It is thus possible to run many experimental cycles on the same sample, which in turn allows to check the reproducibility of the process and to ensure a better accuracy of the results. On sample #□2, the position and the velocity of the moving interface are obtained from an one line resistance measurement. At the end of the experimental cycle, a quench freezes the structure of the solid-liquid front. Peltier pulse marking performed on sample #□3 allow the determination of the shape of the interface at given time intervals. Moreover, thermocouples present in the liquid phase for both samples #□2 and #□3 are used to determine the temperature gradient and possible thermal fluctuations.

The MEPHISTO facility ran for 312 hours, including 216 hours dedicated to scientific operation. During that time period, 24 solidification/fusion cycles were carried out. Five growth rates were preprogrammed (1.7, 3.7, 5.7, 12 and 24 mm/h) before the flight, but, thanks to telepresence operations, we were able to different growth rates to track the morphological stability threshold. Controlled gravity perturbations were realized through planned thruster firings and maneuvers. In all 9 PRCS burns, ranging in duration from 10 to 25 seconds were carried out. In addition, an OMS burn and a 360° X-axis roll were also performed (see Table I).

**Table 1** MEPHISTO PRCS and OMS burns and rolls

<i>Velocity</i>	<i>MET</i>	<i>Duration [sec]</i>	<i>Type</i>	<i>Orientation</i>
V3	5/21:45	25	PRCS	+Z
V3	6/00:00	15	PRCS	+Z
V2	6/22:00	10	PRCS	+Z
V2	7/00:25	10	PRCS	+X
V1	10/12:30	15	PRCS	+Z
V3	12/12:25	15	PRCS	Z*
V3	12/14:25	15	PRCS	+Y
V3	12/14:28	15	PRCS	-Y
V3	12/16:28		360° X-axis roll	
V3	13/12:50	15	PRCS	-Z*

During the first flight of the MEPHISTO directional solidification experiment on NASA's USMP-1 mission in 1992, the impact of sudden effective gravity perturbations were clearly evidenced [1]. Real-time Seebeck voltage variations across a Sn-Bi melt showed a distinct variation that can be correlated with thruster firings (Fig. 1) [1]. The Seebeck voltage measurement is related to the response of the instantaneous average melt composition at the melt-solid interface [4]. This permitted a direct comparison of numerical simulations (and acceleration data) with the Seebeck signals obtained on USMP-1. Motivated by the results of the comparison, we used numerical simulations to predict the response of the Seebeck signal to thruster firings of various magnitudes and durations. The behavior of the signal is directly related to changes in interfacial composition caused by thruster-induced convective disturbances. Motivated by the observations made on USMP-1, one of the objectives of the USMP-3 MEPHISTO experiments was to quantitatively characterize g-jitter effects on an actual crystal growth experiment. To plan the USMP-3 MEPHISTO experiments, simulations were carried out for different solidification rates and g-jitter scenarios.

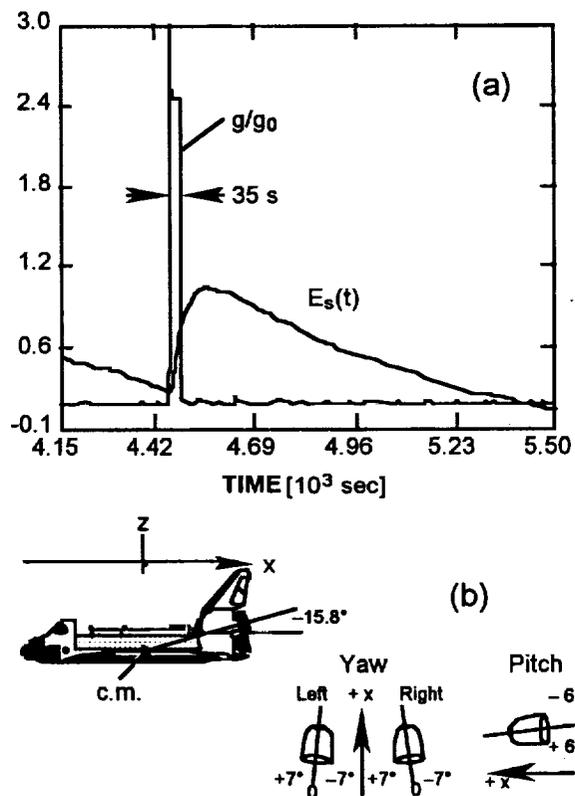


Fig. 1 (a) Acceleration vs. time for the OMS burn recorded on USMP-1 and the associated Seebeck signal ,  $E_s(t)$  [1]. (b) OMS engine alignment with the Orbiter.

There were several differences between the USMP-3 and USMP-1 experiments. First, a more concentrated alloy was solidified on USMP-3, and, second, Primary Reaction Control

System (PRCS) thruster burns were requested at particular times during four separate growth runs. The Seebeck signal was recorded continuously and down-linked in real-time to the MEPHISTO experiment team at NASA's Marshall Space Flight Center. This allowed for quantification of the effects of "g-jitter" on convective-diffusive transport in the melt through the changes in average interfacial composition obtained from the Seebeck measurement. In addition, guided by SAMS acceleration data, we carried out simulations for comparison with the recorded Seebeck signals. The effects of thruster firings on the average composition was monitored in six separate experiments and for eleven separate acceleration disturbance events. Selected results from the post-flight analysis are described below.

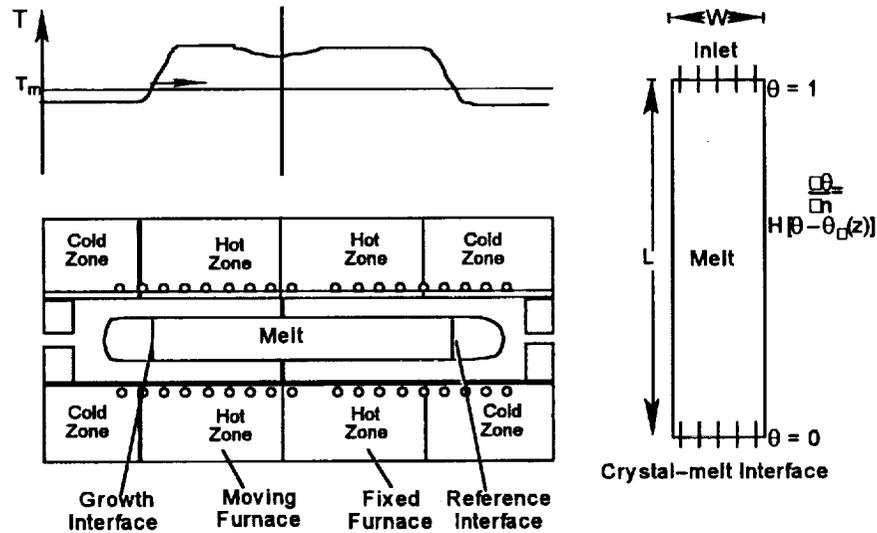


Fig. 2 The MEPHISTO set-up (bottom-left), temperature profile (top left) and computational model (right).  $T_m$  denotes the melting temperature.

#### 1.4 Approach

A sketch of the experiment set-up is shown in Fig. 2. There are two furnaces. One is fixed. The other is translated. The applied temperature profile shown in Fig. 2 leads to a central cylindrical melt volume bounded by a moving and a stationary (or reference) solid-liquid interface. The melt composition at the moving and the stationary reference interfaces is not the same. For Sn-Bi there is a dependence of melting temperature on concentration. Thus, it follows that the melting temperature at the two interfaces will also be different. The Seebeck effect gives rise to a small but measurable voltage difference between these two interfaces. Measurement of this voltage difference allows us to determine the average temperature and, thus, the average composition of at the growing interface. The MEPHISTO set-up and the Seebeck measurements are discussed in more detail in [4].

The numerical model used to simulate the response of the tin-bismuth melt to particular types of g-jitter has been described elsewhere [5-7]. The essential features are outlined below. Solidification takes place as the furnace is translated along the ampoule (see Fig. 2). Directional solidification due to the furnace translation is simulated by supplying a two-component melt of bulk composition  $c_0$  at a constant velocity  $V_g$  at the top of the computational space (inlet), and

withdrawing a solid of composition  $c_s = c_s(x,t)$  from the bottom (See Fig. 2). The crystal-melt interface is located at a distance  $L$  from the inlet; the width of the ampoule is  $W$ . The temperature at the interface is taken to be  $T_m$ , the melting temperature of the crystal, while the upper boundary is held at a higher temperature  $T_h$ . In the actual experiment, the temperature gradient along the ampoule wall ahead of the growing interface was essentially linear (195 K cm<sup>-1</sup>). Thus, we set a linear temperature gradient along the wall in the simulations. Furthermore, since we wish to confine our attention to compositional nonuniformities caused by buoyancy-driven convection, rather than variations resulting from non-planar crystal-melt interfaces, the interface is held flat. We expect that, given the large temperature gradient, changes in melting temperature due to compositional non-uniformity will not lead to significant changes in interface shape due to interfacial compositional inhomogeneity. Furthermore, because of the melt's low Prandtl number and the low magnitude accelerations, convection does not lead to significant deviations of the temperature from the conductive state. Thus, changes in the interface shape due to changes in the thermal field will be negligible. In an actual experiment, owing to the finite length of the ampoule, there is a gradual decrease in length of the melt zone during growth. In this model, transient effects related to the change in melt length are ignored. Since the MEPHISTO experiments involve melt lengths that far exceed the ampoule diameter, this does not preclude us from calculating the compositional transient. That is, we can start the calculations by solidifying from an initially uniform composition melt.

The dimensionless governing equations governing coupled convective-diffusive heat mass and species transfer in the melt were assumed to be those for a Boussinesq fluid with a linear dependence of density on temperature and composition. The boundary conditions imposed were those corresponding to plane front directional solidification at a translation rate  $Vg$  with a linear temperature gradient applied to the ampoule walls. Solute was preferentially rejected at the crystal-melt interface ( $k = 0.27$  for this Sn-Bi alloy). The equations were solved using a Chebyshev spectral method.

## 2. Selected Results

The PRCS firings produced an impulse acceleration with the largest component parallel to the solid liquid interface. Figure 3 shows the actual (uncorrected for drift) and predicted Seebeck signal for an experiment subject to two thruster burns (25 seconds and 10 seconds in duration) that produced an acceleration oriented parallel to the crystal-melt interface (i.e. perpendicular to the ampoule axis). Before the first burn, the Seebeck signal decreases monotonically. Immediately following the burn, the signal increases rapidly and reaches a maximum about 100 seconds after the termination of the burn. The signal then decreases slowly and eventually takes on almost the same slope that it had before the burn. The same behavior occurs following the second burn. After translation of the furnace was stopped. Solidification then ceases and the Seebeck signal increases as the average interfacial concentration decreases. Note that the actual Seebeck measurement shown has not been corrected for drift and, thus, the voltage does not return to its original value. For the response of the Seebeck signal predicted by computer simulation, there is an immediate rapid response to each of the firings. In addition, we see that the time taken for the Seebeck signal to reach a maximum value is approximately the same for the computed and measured signals.

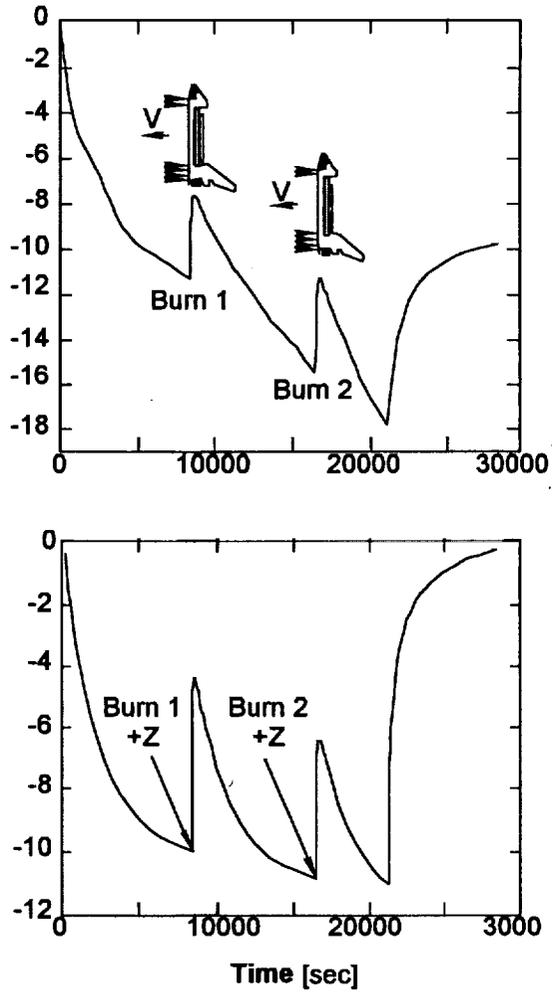


Fig. 3 (a) Actual and (b) predicted Seebeck signal for a 25 second and 15 second burn oriented parallel to the crystal-melt interface.

### **3. Publications and presentations arising from the funded research**

#### **3.1 Publications in refereed journals and books (4)**

- J.I.D. Alexander, J.P. Garandet, J.J. Favier and A. Lizée, "G-jitter effects on segregation during directional solidification of tin-bismuth in the MEPHISTO furnace facility", *J. Crystal Growth*, 178, 657-661, 1997.
- A. Lizée and J.I.D. Alexander, "Thermovibrational convection, parametric resonance and transitions to chaos", *Physical Review E*, 56, 4152-4156, 1997.
- J.P. Garandet, S. Corre, S. Gavaille, J.J. Favier and J.I.D. Alexander, "On the effect of gravity perturbations on composition profiles during Bridgman crystal growth in space", *J. Crystal Growth*, 165, 471-481, 1996.
- J.I.D. Alexander, "Response of crystal growth experiments to time-dependent residual acceleration", *Lecture Notes in Physics Vol. 464*, Eds. L. Ratke, H. Walter and B. Feuerbacher (Springer Verlag, Berlin, 1996) 95-105.

#### **3.2 Publications in Conference Proceedings**

- J. Iwan D. Alexander, Impulses, Vibrations and random disturbances: Consequences for convective diffusive transport and fluid interfaces in low gravity experiments AIAA Paper No. 96-2073, Proc. 27th AIAA Fluid Dynamics Conference, June 17-20, New Orleans, La., 1996
- A. Lizée and J.I.D. Alexander, "Transitions to chaos during vibrational forcing of a differentially heated cavity", *Proceedings of the 2nd Symposium on Fluids in Space*, Naples, Italy, April 22-26, 413-417, 1996.
- J.I.D. Alexander, J.-J. Favier, J.-P. Garandet, Study of Interesting Solidification Phenomena on the Ground and in Space (MEPHISTO), *Proceedings of the 1998 NASA Microgravity Materials Science Conference*, Von Braun Center, Huntsville, Alabama, USA, July 14-16, 1998.

#### **3.3 Presentations**

- "Quantitative experimental characterization of g-jitter effects on directional solidification," presented at the 35th AIAA Symposium, Reno, Nevada, January 6-10, 1997.
- "G-Jitter and chaos" (Invited), Gordon Research Conference on Gravitational Effects in Physico-Chemical Systems, Henniker, New Hampshire, June 29-July 4, 1997.
- "Study of interesting solidification phenomena on the ground and in space (MEPHISTO)", USML-2 and USMP-3 Launch-plus 1 Year Conference, National Academy of Science, Washington D.C., February 10-11, 1997.
- "Vibrations and random disturbances": Consequences for convective diffusive transport and fluid interfaces in low gravity experiments AIAA Paper No. 96-2073, Proc. 27th AIAA Fluid Dynamics Conference, June 17-20, New Orleans, La., 1996
- "Response of crystal growth experiments to time-dependent residual acceleration", Ninth European Symposium on Gravity Dependent Phenomena in Physical Sciences, Berlin 2-5 May, 1995

### **4. Students and Post-doctoral researchers supported under this grant\**

The grant supported one post-doctoral researcher. Dr. Arnaud Lizée

## 5. Summary

The USMP-3 MEPHISTO experiments permitted a quantitative correlation between well-characterized g-jitter recorded by spacecraft accelerometers and the response of a directionally solidifying alloy. Furthermore, the comparison of the results of the experiment with the predictions of numerical simulations carried out on the Alabama Research and Education Network's C-90 Supercomputer, allowed us to verify the degree to which such simulations can accurately predict experiment sensitivity to g-jitter accelerations. Without such quantitative experimental verification, predictions are of limited use and could lead to unnecessary design restrictions, an undesirable low gravity environment, or unsuitable experiment operating conditions.

Ongoing work beyond this proposal involves corrections of the raw Seebeck data, analysis of residual acceleration data measured by NASA's Space Acceleration Measurement System, estimation of the degree of melt homogeneity following remelting cycles and refinement of the computational simulations. This unique experiment showed use of simulations to assist in changes in experiment strategy greatly enhances the scientific return from limited opportunity spaceflight experiments. It is expected that ongoing work will yield benchmark comparisons between measured and predicted residual acceleration effects.

## 6. References

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