ORBITAL PROCESSING OF EUTECTIC ROD-LIKE ARRAYS

525-29
393400
358181
p8

David J. Larson, Jr.
Department of Materials Science & Engineering,
University at Stony Brook,
Stony Brook, NY 11794-2275
NASA Summer Faculty Fellow

Frank Szofran, Palmer Peters and Richard Grugel
Space Sciences Laboratory
George C. Marshall Space Flight Center 35812
NASA Colleagues

I. Introduction

The eutectic is one of only three solidification classes that exist. The others are isostructural and peritectic-class reactions, respectively. Simplistically, in a binaryeutectic phase diagram, a single liquid phase isothermally decomposes to two solid phases in a cooperative manner. The melting point minimum at the eutectic composition, isothermal solidification temperature, near-isocompositional solidification and refined solidification microstructure lend themselves naturally to such applications as brazing and soldering; industries that eutectic alloys dominate.

Interest in direct process control of microstructures has led, more recently, to in-situ eutectic directional solidification with applications in electro-magnetics and electro-optics. In these cases, controlled structural refinement and the high aspect ratio and regularity of the distributed eutectic phases is highly significant to the fabrication and application of these in-situ natural composites. The natural pattern formation and scaling of the dispersed phase on a sub-micron scale has enormous potential application, since fabricating bulk materials on this scale mechanically has proven to be particularly difficult. It is thus of obvious importance to understand the solidification of eutectic materials since they are of great commercial significance.

The dominant theory that describes eutectic solidification, Jackson and Hunt, was derived for diffusion-controlled growth of alloys where both solid eutectic phases solidify metallically, i.e. without faceting at the solidification interface. Both high volume fraction (lamellar) and low volume fraction (rod-like) regular metallic arrays are treated by this theory. Many of the useful solders and brazements, however, and most of the regular in-situ composites are characterized by solidification reactions that are faceted/non-faceted in nature, rather than doubly non-faceted (metallic). Further, diffusion-controlled growth conditions are atypical terrestrially since gravitationally-driven convection is pervasive. As a consequence, it is important to determine whether these faceted/non-faceted composites behave in the same manner as their doubly non-faceted counterparts, particularly in the presence of convection.

Prior analytical convective sensitivity testing of this theory predicted insensitivity, Verhoeven, Gibson and Homer. Prior experimental testing of this theory offered broad-based agreement between theory and experiment, though most results were for high volume fraction lamellar eutectics that solidified without faceting at the solidification interface. Directional solidification experiments of low volume fraction rod eutectics under damped (microgravity or magnetic field) conditions, however, have demonstrated significant sensitivity, challenging this fundamental theory. More recent theories have been proposed which introduce kinetic undercooling, faceting, fluid shear of the solute redistribution zone and the possibility that the interface composition is not the same as the bulk liquid composition. This program tests the established and proposed analytical theories and addresses the origins of discrepancies between the experimental and analytical results.

II. Experiment

II.1 Proposed Research Program

This program will directionally solidify low volume fraction regular eutectic structures in orbit (μ-g), under diffusion-controlled growth conditions. Complementary in-situ diagnostics will be applied to monitor and/or perturb the solidification interface velocity and to precisely monitor the solidification interface temperature (undercooling) and shape. Post-flight analyses will determine, quantitatively, the relations between the solidification interface velocity, solidification interface temperature (undercooling), and microstructural parameters (rod diameter, inter-rod spacing and volume fraction) that are enumerated in the eutectic theoretical relationships. Identical experiments will be conducted terrestrially, under damped (thermal, gravitational and applied magnetic field stabilization) and undamped conditions to provide a comprehensive quantitative one-g baseline for comparison.

The experiments enumerated above will develop critical data, under both diffusion-controlled and convection-dominated conditions. These data sets will be used to test analytical theories describing the solidification of regular, low volume fraction, eutectic alloys. Specific efforts will be made to quantitatively compare these results with the diffusion-controlled Jackson-Hunt theory and the Verhoeven, et al, modification which predicts convective insensitivity. These results will also be compared to the analytical formulation of Drevet, et al, which attempts to incorporate within the Jackson-Hunt formulation kinetic undercooling, faceting, convection and the possibility that the interfacial composition is not the same as the bulk liquid eutectic composition.

As a critical part of this task, a directional solidification model that describes current interface demarcation will be developed and validated experimentally. This model will predict interface shape and location during the current pulse and post-pulse transient, as well as eutectic rod-like structure within these regions and during steady state growth. This insight will allow us to optimize the microstructural observation, while maintaining steady-state growth conditions between pulses. This model will include thermoelectric contributions (Peltier, Thomson, Joule, and Seebeck), as well as solute redistribution (including Soret diffusion), current-induced magnetic field effects, applied magnetic field effects, and gravitationally-driven convection. This model will predict responses under damped and undamped conditions and will differentiate between the microgravity damping and the applied magnetic field damping.

II.2 Description Of Anticipated Summer Research

Previous work conducted by the Principal Investigator, under NASA contract, has established that gravitationally-dependent convection substantially impacts the scaling dimensions of the Bi/MnBi eutectic microstructures under investigation. This structural modification occurs on damping the natural convection, not on accelerating the convection. This has been demonstrated by damping gravitationally-dependent convection in microgravity as well as by applying transverse or longitudinal magnetic fields in unit gravity. This convective sensitivity is not predicted by the existing eutectic solidification theories noted above.

The proposed work is intended to investigate the microstructural refinement experimentally, under damped conditions. Previously used magnetic field facilities are no longer available to the Principal Investigator. The Applied Magnetic Field Facility housed at the Space Sciences Laboratory of the NASA George C. Marshall Space Flight Center is an ideal location at which to conduct this study. Dr., Frank Szofran of the NASA Space Sciences Laboratory graciously offered to act as the NASA Research Colleague.

Our plan was to directionally solidify Bi/MnBi eutectic samples with and without applied magnetic fields, under conditions of thermal and gravitational stabilization. Simultaneous in-situ temperature measurements will be made, using sub-miniature Chromel -Alumal sheathed thermocouples. Complementary current interface demarcation experiments will be conducted under identical growth conditions, and simultaneous current interface demarcation experiments will be attempted. In the latter case, the generation of large Lorentz forces, which might significantly disturb experiments of this type, will be a major concern. Failure of this approach will render the microgravity experiments unique. Quantitative microstructural measurements will be conducted on the processed samples.

II.3 Revised Research Plan, Results and Discussion

On arrival at the NASA/MSFC/SSL facilities, it became apparent that facilities other than the previously described Applied Magnetic Field facility would also be of great utility to the program. One of the difficult aspects of this experimental program is to fabricate homogeneous alloys of the correct eutectic composition. This is a problem because of the large difference in melting point between the two constituent species combined with their relatively large differences in density, which allows the non-melted Mn to separate from the liquid Bi by Stokes flow. A candidate solution to this problem was found at SSL in the form of a rocking furnace that resided in the laboratory. It was decided to utilize this furnace to fabricate homogeneous alloys for the investigations. Empirical tests demonstrated that 8 hours of mixing at 525°C was totally ineffective, but that 100 hours at 925°C was sufficient to create a homogeneous alloy. The latter conditions were adopted as the baseline. Evacuated ampoules of correct alloy composition were fabricated at the State University of New York and transported to SSL by the PI. Four boules have been homogenized and additional castings will be fabricated throughout the Fall on an as-needed basis.

Further discussions with SSL personnel indicated that a new program was being initiated at SSL studying precision temperature measurement using the Seebeck technique. This technique had been proposed in our original proposal as a candidate to measure the interfacial undercooling that are fundamental to the theories being investigated. This technique is particularly valuable since it uses the solid/liquid solidification interface as a thermocouple without employing intrusive hardware and can measure the interface temperature, in some cases, to with 0.1°C. This is an ideal technique for achieving our ends, but the technology had to be extended to the two phase eutectic case, since it had previously been demonstrated for single phase solidification. These complementary programmatic interests offered an opportunity to advance both programs by collaborating.

Discussions with Drs. Peters, Sen, and Kaukler suggested that the ideal sample geometry for their apparatus was a single crystal of the sample material approximately 0.1 cm in diameter and 15 to 20 cm long. This posed a serious practical problem of growing such crystals. The first steps were taken with the assistance of Dr. Grugel of SSL. He provided facilities to aspirate liquid Bi and Bi alloys into the requisite fused silica capillaries. Initial successful attempts were made using 0.999 Bi and subsequent samples were fabricated using 0.999999 Bi. This provided samples to initiate the Seebeck work and allowed additional time to fabricated Bi/MnBi eutectic alloys. The latter work is in progress.

Initial measurements on the 0.999999 Bi samples suggested the signal from the sample was of the correct order of magnitude (i.e. 8 to 10 microvolts) and we were encouraged to note that the signal noise was 0.1 to 0.2 microvolts, which was tolerable. Experiments are underway to measure the Seebeck Coefficient for pure Bi to compare with published values, and to make similar measurements on the Bi/MnBi eutectic alloy. Experiments on the eutectic alloy will determine whether Soret Transport is a problem with respect to the surface composition and output voltage of the reference thermocouple junction. Further experiments on the eutectic alloy will determine whether the distributed second phase inherent to the eutectic creates output noise that overwhelms the Seebeck signal or decreases the signal/noise ratio to the point where interpretation of the temperature unambiguously is impossible. These experiments are continuing.

Finally, the Applied Magnetic Field experiments were also undertaken. First homogeneous alloys were fabricated, as described previously. The next step was to calibrate the temperature field within the growth furnace. This was accomplished with the help of Mr. Curtis Bahr and the final value selected was a thermal gradient of 35°C/cm at the solidification temperature of 265°C. Experimental conditions selected were solidification velocities of 4, 9, and 16 cm/h with and without the applied fields. Success with this experimental matrix would cue the initiation of the current interface demarcation experiments with and without the applied fields. To date the 4.0 and 9.0 experiments, with and without the magnetic field have been completed. The remainder of the program will be conducted over the next several months.

III. Conclusions

This was a unique opportunity to advance the research program of the Summer Faculty Fellow. Results in homogeneous alloy fabrication were excellent. Similarly, the fabrication of Bi and Bi/MnBi samples with a very high aspect ratio were successfully accomplished. Initial Seebeck measurements were encouraging both with respect to output signal magnitude and signal cleanliness. Lastly, initial samples under damped and undamped growth conditions have been fabricated and are under analysis. More work is necessary, and collaboration in each of these areas will continue throughout the 1997/1998 academic year.