USE OF MICROGRAVITY TO CONTROL THE MICROSTRUCTURE OF EUTECTICS

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Introduction

The long term goal of this project is to be able to control the microstructure of directionally solidified eutectic alloys, through an improved understanding of the influence of convection.

Prior experimental results on the influence of microgravity on the microstructure of fibrous eutectics have been contradictory [1,2]. Theoretical work at Clarkson University showed that buoyancy-driven convection in the vertical Bridgman configuration is not vigorous enough to alter the concentration field in the melt sufficiently to cause a measurable change in microstructure when the eutectic grows at minimum supercooling. Currently, there are four other hypotheses that might explain the observed changes in microstructure of fibrous eutectics caused by convection:

1. Disturbance of the concentration boundary layer arising from an off-eutectic melt composition and growth at the extremum.
2. Disturbance of the concentration boundary layer of a habit-modifying impurity.
3. Disturbance of the concentration boundary layer arising from an off-eutectic interfacial composition due to non-extremum growth.
4. A fluctuating freezing rate combined with differences in the kinetics of fiber termination and fiber formation.

We favor the last of these hypotheses. Thus, the primary objective of the present grant is to determine experimentally and theoretically the influence of a periodically varying freezing rate on eutectic solidification. A secondary objective is to determine the influence of convection on the microstructure of at least one other eutectic alloy that might be suitable for flight experiments.

Influence of Electrical Current Pulsing on the Microstructure of MnBi-Bi Eutectic (Fengcui Li)

Previously, we had performed a few ground-based experiments using electric current pulses to deliberately create an oscillatory freezing rate [1,2]. Although the MnBi rod spacing appeared to increase with increasing current, we were not confident of the results because the freezing rate or pulsing conditions were varied during each experiment. With such experiments, it is difficult to be certain of the conditions under which each portion of the resulting ingot froze. Thus, we are currently performing a more thorough study in which the conditions are held constant for the duration of each experiment. New image analysis techniques were developed to yield detailed statistical information on the microstructure from numerous scanning electron micrographs. Thus far, 15 ingots have been solidified and analyzed under these conditions. The microstructures without current pulsing and at the lowest current consisted of fine, quasi-regular, semi-triangular MnBi rods. As the current was increased, increasing portions of each cross sectional slice showed other areas of less regular fibers, broken lamellae, large irregular MnBi, or completely absent of MnBi. We have confined our analysis of the microstructure to the areas showing the quasi-regular rods. The nearest neighbor distance was determined for each rod. With one exception, the standard deviation ranged from 33% to 38% of the mean nearest neighbor distance λ. The 95% confidence limits were very small compared to λ. The kurtosis and skewness were appreciable compared to the standard deviation, indicating moderate deviations from the usual bell-shaped normal distribution. Without current pulsing, the value of λ^2V was nearly the same for three different ampoule lowering velocities V. Figure 1 shows λ versus current amplitude for two different freezing rates and three different current pulsing conditions. For all conditions, λ decreased as current was increased. Note that a constant current (without pulsing) caused no significant change in λ. An alternate method of characterizing fiber spacing is to measure the fiber density ρ, i.e. the number of fibers per unit area. The values of $\rho^{-1/2}$ parallel the results shown in Figure 1, but are about 50% larger. Figure 2 shows the average percent of the cross sectional area that is MnBi. This appears to increase slightly with increasing current. (The run showing the exceptionally high value of %MnBi also exhibited an exceptionally large standard deviation for nearest neighbor distance, about 56% of the mean.)

Flight Experiments (Fengcui Li)

Originally, we had planned to fly experiments on Mir using QUELD, the Canadian gradient-freeze furnace that is installed in the Microgravity Isolation Mount (MIM). The MIM is capable of introducing controlled vibrations into an apparatus, as well as actively reducing vibrations. We had planned to take advantage of this capability. Unfortunately, because of the problems with Mir these experiments have been delayed, probably permanently. Twenty-four ampoules containing MnBi-Bi eutectic were prepared for these experiments. Several of these ampoules will be solidified at Queen's University in a ground-based copy of QUELD. We plan to determine the influence of growth rate perturbations, either by periodic mechanical shocks or heater power variations.

Influence of Accelerated Crucible Rotation on Al-Si Microstructure (Ram Ramanathan)

Experiments are underway on directional solidification of the Al-Si eutectic at different freezing rates, with and without application of accelerated crucible rotation to induce convection.

Theory of Eutectic Solidification with an Oscillatory Freezing Rate (Dimitri Popov)

For the first time, theoretical methods were developed to analyze eutectic solidification with an oscillatory freezing rate. Both a classical sharp-interface model and a phase field model are being used. A paper demonstrating the application of phase field methods to periodic structures was submitted for publication [3].

One-sided sharp-interface method. This method is based on the solution of heat and mass transfer equations separately in each phase. These solutions are connected at the freezing interface using the boundary conditions for flux conservation and continuity of temperature and concentration. At steady state with a constant freezing rate, the composition field near the interface depends on the distance d by which one phase leads the other. For both lamellar and rod eutectics, as d increases, the composition along the interface moves farther from the eutectic for both leading and trailing phases. The composition near the trailing phase becomes more uniform as d increases.

We imposed temperature fluctuations in the melt in order to generate an oscillatory freezing rate. Under some conditions there is an amplification of the amplitude of the freezing rate oscillations due to oscillations of temperature, especially if the kinetic coefficient is high. The spatially averaged composition at the interface of each phase was calculated and integrated over one period of freezing rate oscillations after the initial transient had decayed. The difference between this composition and that for a steady freezing rate was calculated and represents the excess amount of rejected component ahead of the growing phase due to freezing rate oscillations. The frequency dependence of the spatially averaged composition reveals the same high-frequency cut-off for all values of d. With increasing d, the difference in composition from the eutectic increases, as can be seen in Figures 3 and 4. That is, there is a higher supersaturation ahead of each growing phase. We believe that this high degree of supersaturation may provoke nucleation of the other phase. Therefore, a large d would be favorable for development of a finer microstructure due to freezing rate oscillations. Figure 5 shows how the concentration difference from the eutectic would diminish after nucleation of new rods or lamellae that make λ smaller.

Phase-field method. In order to be able to track the interface dynamics, particularly where the three phases (melt, α solid and β solid) come together, we initially chose the phase-field method. The governing equations were formulated using two phase-field parameters, functions of temperature and concentration. First, the necessary accuracy in the calculations of the concentration in the bulk of the phases was achieved for one dimension and one solid phase, providing the correct solution for the interfacial region. A simple lamellar eutectic structure was calculated at a constant freezing rate. As shown in Figure 6, the interface shape and the composition field ahead of the interface agree with the classic model of Jackson and Hunt. A fluctuating freezing rate and concentration ahead of the interface were obtained as a response to temperature fluctuations in the melt. The concentration change lags behind the interface velocity fluctuation, as in the one-sided model. With an oscillatory freezing rate, the majority phase tends to grow at the expense of the minority phase, and under some conditions even chokes it off and makes λ larger.

Thus we have mechanisms that can both increase and decrease λ. The challenge remains to incorporate those mechanisms in a single computational model.

Acknowledgment

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References

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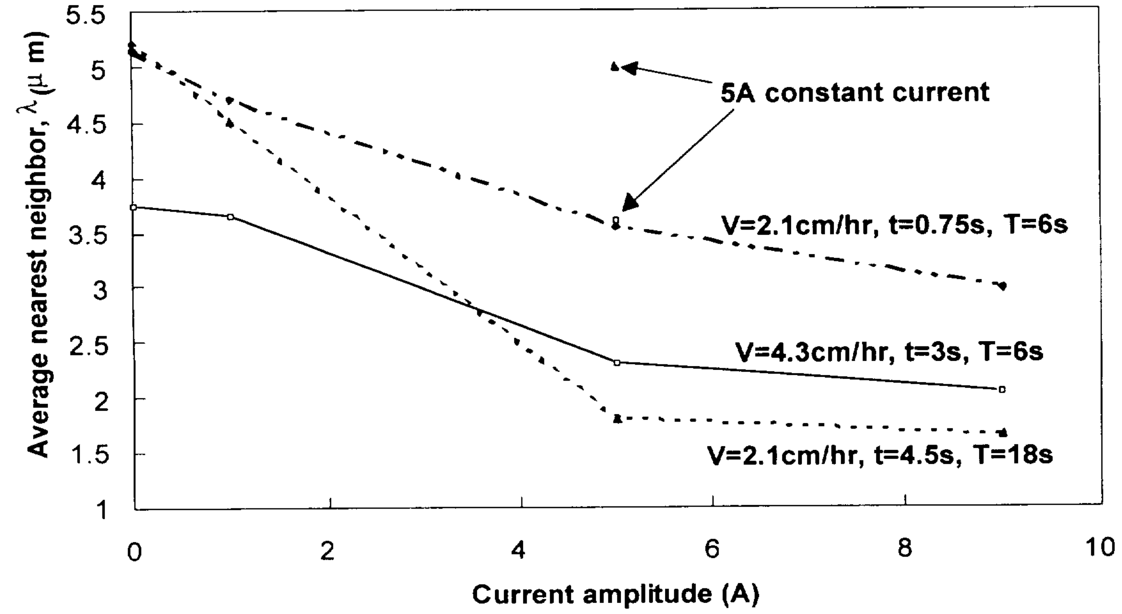

Figure 1. Average nearest neighbor distance λ of quasi-regular MnBi rods versus current amplitude, ampoule translation rate V, time t during which current is passed through the sample, and period T of current pulses. Note the two data points for a constant current of 5A (no pulsing).

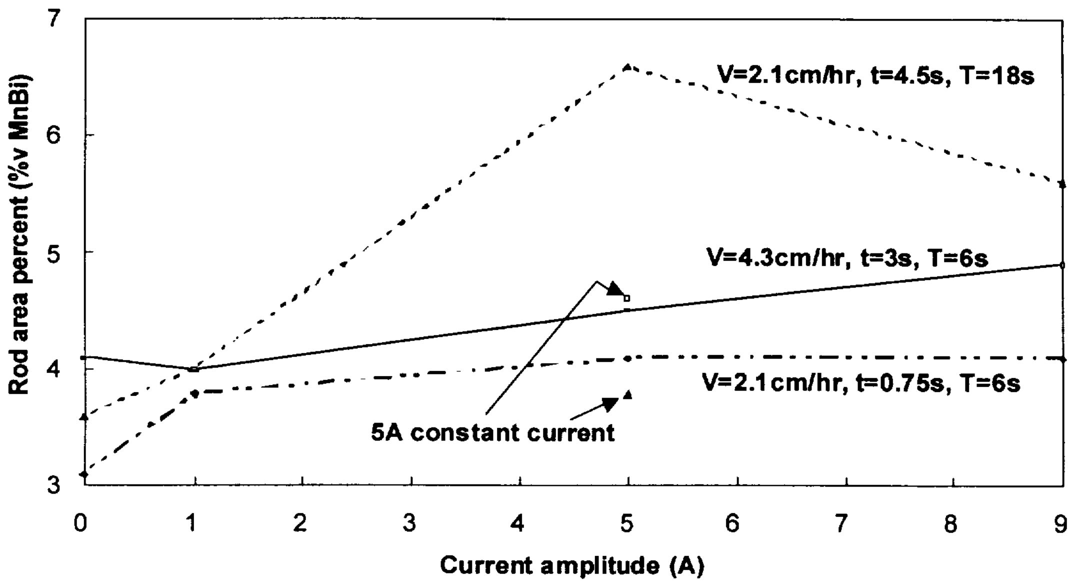

Figure 2. Average percent of the SEM quasi-regular areas that is MnBi fibers. Here V is the translation rate of the ampoule, time t during which current is passed through the sample, and period T of current pulses. Note the two data points for a constant current of 5A (no pulsing).

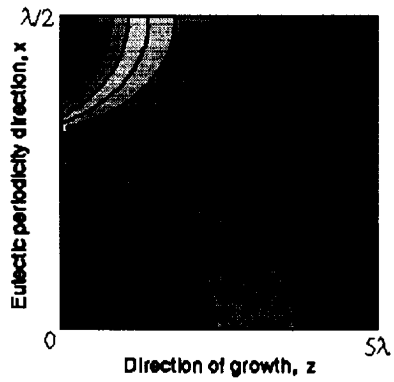

Figure 3. Composition field at an instant of time near the planar interface of a lamellar eutectic, freezing with an oscillatory rate. This solution was obtained using a sharp interface model.

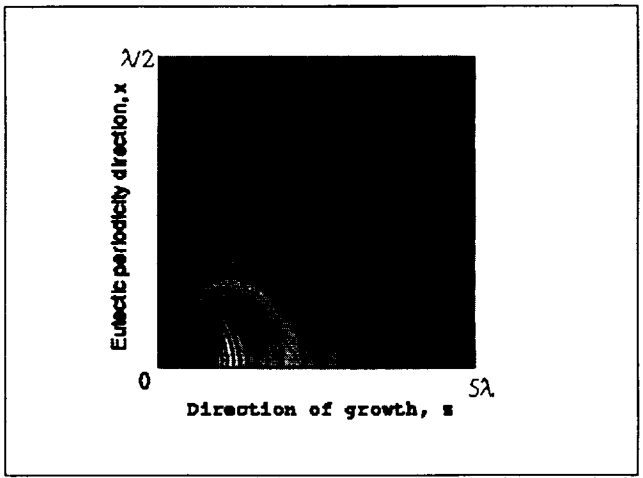

Figure 4. Composition field at one instant of time near a stepped eutectic interface, freezing with an oscillatory rate, from the sharp interface model.

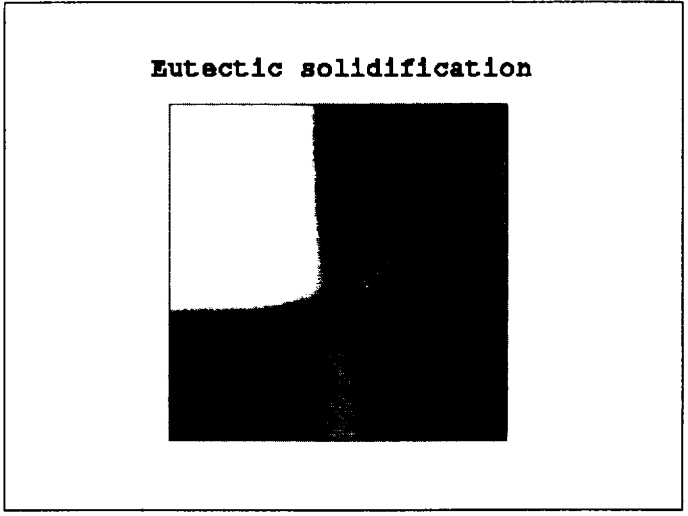

Figure 6. Eutectic solidification at constant rate, modeled with the phase-field method. The α-phase has a large concentration of A (white), β has a low concentration of A (black), and the melt is at the eutectic (intermediate) concentration. Note the zones of rejection of the alien components (A rejected by β, and B rejected by α).

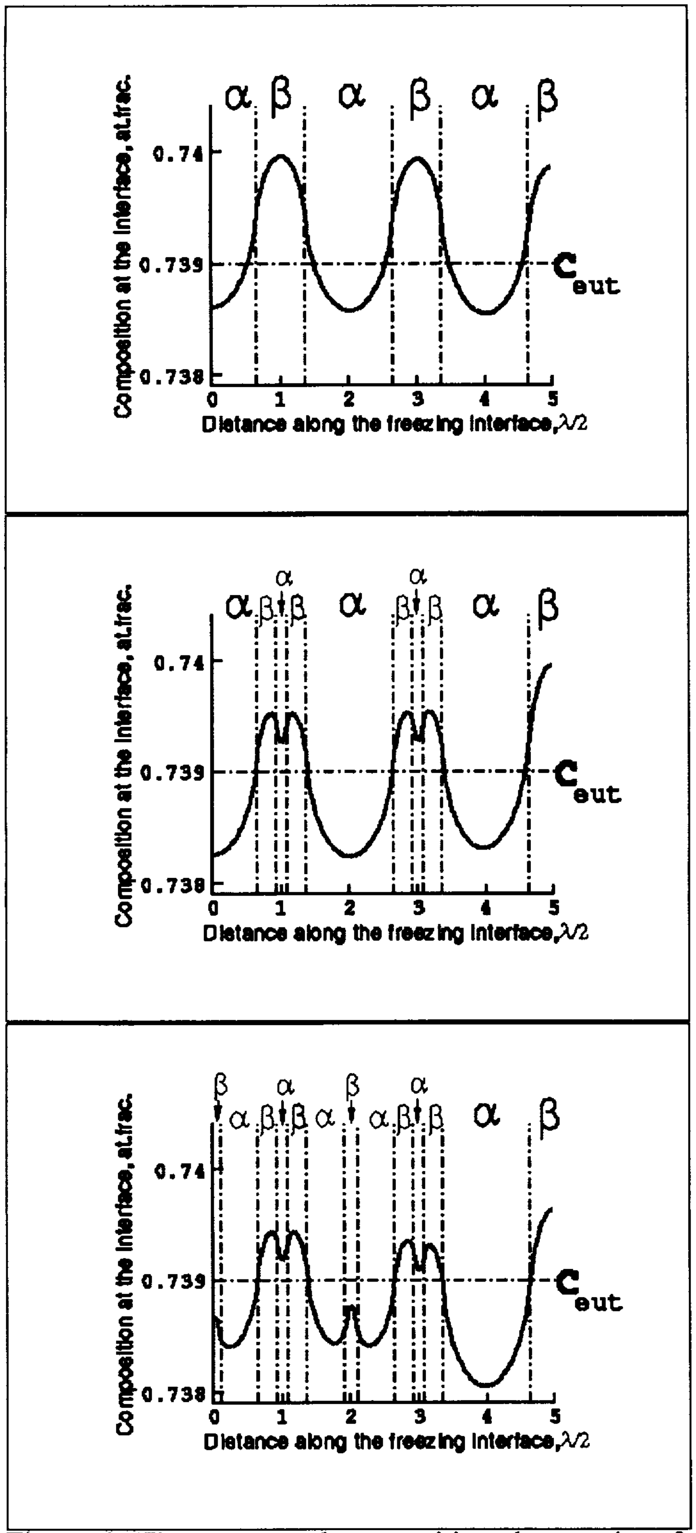

Figure 5. Time-averaged composition along an interface with an oscillatory freezing rate.
(a) – Initial state, before nucleation
(b) – After nucleation of new β
(c) – After the additional nucleation of new α