Use of Microgravity to Control the Microstructure of Eutectics

William R. Wilcox and Liya L. Regel
Clarkson University
Potsdam NY 13699-5814
E-mail: wilcox@agent.clarkson.edu)

and

Reginald W. Smith
Queen's University
Kingston, Ontario

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. The primary objective of the present projects is to test hypotheses for the reported influence of microgravity on the microstructure of three fibrous eutectics (MnBi-Bi, InSb-NiSb, Al3Ni-Al). A secondary objective is to determine the influence of convection on the microstructure of other eutectic alloys.

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 three hypotheses that might explain the observed changes in microstructure of fibrous eutectics caused by convection:

1. A fluctuating freezing rate, combined with different kinetics for fiber termination and branching.
2. Off-eutectic composition, either in the bulk melt or at the freezing interface as a consequence of operation off the minimum supercooling condition.
3. Presence of a strong habit modifying impurity.

We favor the first of these hypotheses. Previously, we had performed ground-based experiments using electric current pulses to deliberately create an oscillatory freezing rate.[1,2] Although the current pulsing appeared to have coarsened the microstructure of MnBi-Bi eutectic as predicted, it may also have introduced localized convection. For the current grant, image analysis techniques were developed to obtain the distribution of MnBi fiber spacings and area fraction. Experiments in which the freezing rate or pulsing conditions were varied during solidification, as in references 1 and 2, proved impossible to interpret unambiguously. Consequently, 10 ingots were solidified under different conditions, but with the ampoule translation rate and pulsing held constant during each run. Samples from the same positions in all ingots are being compared. Thus far, there is no evidence for an increase in fiber spacing caused by current pulsing.

Twenty-four ampoules containing MnBi-Bi eutectic were prepared for use in the gradient freeze furnace QUELD, developed at Queen's University for use in microgravity. Some of these ampoules will be solidified at Queen's in a ground-based model. We plan to determine the influence of growth rate perturbations, including periodic vibrations. We hope to solidify other samples in the QUELD that is mounted on the Canadian Microgravity Isolation Mount on *Mir.*

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.

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 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 diffusion equations separately in each phase and sewing the solutions at the interface by appropriate boundary conditions resulting from flux conservation. When one phase leads the other by distance *d*, the composition field near the interface changes. We calculated the steady state composition distribution versus *d*, both for lamellar and for rod eutectics. As *d* increases, the composition at the interface moves farther from the eutectic for both leading and trailing phases. The composition along the interface of the trailing phase becomes more uniform, and is almost constant for high *d*.

We imposed temperature fluctuations in the melt in order to generate an oscillatory freezing rate. We assumed that the response of the freezing rate is linear to a change in the interface temperature. Thus there should be no phase lag between the temperature and freezing rate oscillations. Because of diffusion-controlled growth, at particular frequencies we found an amplification of the amplitude of the freezing rate oscillations due to oscillations of temperature. This amplification is considerable 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 solely due to freezing rate oscillations. The value of this difference approaches a constant value after several periods of oscillation. The number of periods necessary to reach a stationary solution is less for lower frequencies of 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 concentration difference increases. 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.

We are now developing a model in which nucleation of the other phase occurs when the local supersaturation at the interface exceeds a certain level. In this model, the volume fraction can change in time due to growth of the new phase and the subsequent redistribution of the composition in the melt near the interface.

Phase-field method

In order to be able to track the interface dynamics, and particularly the region of connection of the three phases (liquid and two solid phases), 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 in one dimension and one solid phase, providing the correct solution for the interfacial region. A simple lamellar eutectic structure was recovered in a constant temperature gradient. The interface shape and the composition field ahead of the interface are in agreement with Jackson-Hunt. A fluctuating freezing rate and concentration ahead of the interface were obtained as a response to a temperature fluctuation in the melt. The concentration change lags behind the interface velocity fluctuation, as in the one-sided model. We are now trying to find phase-field parameters that give the most sensitivity of the tri-junction to variations in the temperature.

References

1. W.R. Wilcox and L.L. Regel, *Microgravity Quarterly*, 4, 147-156 (1994).
2. W.R. Wilcox and L.L. Regel, *Acta Astronaut.*, 38, 511-516 (1996).
3. D.I. Popov, L.L. Regel, and W.R. Wilcox, "Fourier Collocation and Fourier Galerkin Methods applied to the Phase-Field Model of Two-Dimensional Phase Transition Problems," *J. Physics D: Applied Physics* (submitted).