Theoretical Analysis of 3D, Transient Convection and Segregation in Microgravity Bridgman Crystal Growth

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Introduction and Objectives

Key to the advancement of crystal growth processing is a better understanding of the dominant influence of buoyancy-driven convection on segregation and morphological stability during crystal growth on earth. Since these effects in turn play a critical role in establishing the structure and properties of grown materials, considerable interest has developed for their study by use of the microgravity environment provided by space flight. However, space flight experiments are still subject to accelerations that rapidly vary in magnitude and direction, and such variations complicate experiments by the introduction of significant transient and three-dimensional effects. We propose to develop rigorous process models that account for such three-dimensional, transient phenomena during melt crystal growth under microgravity conditions.

With this model, we will perform fundamental studies of transient acceleration and g-jitter effects – significant advances in the understanding of segregation phenomena are to be expected. In addition, there is an immediate and continuing need of the microgravity flight experiment program for three-dimensional growth models coupled with realistic heat transfer models that describe current NASA flight furnaces, such as the Advanced Automated Directional Solidification Furnace (AADSF) and Crystal Growth Furnace (CGF). The codes developed here will enable the unambiguous interpretation of flight experiments, and will, perhaps even more importantly, provide a tool to design the conditions needed in future flight experiments to best study the effects of microgravity on segregation theory.

Relationship to Microgravity Research

There has long been interest in employing the microgravity environment of space to process advanced materials. Growing crystals in space has the alluring potential to reduce convective flow velocities to the point where diffusion-controlled growth is possible, thus promoting the growth of higher quality crystals and enabling the detailed study of segregation phenomena. Early space experiments confirmed that microgravity processing could significantly reduce the level of melt convection. Uniform doping was not achieved, however, indicating that convection was not completely damped in the melt. Recent experiments conducted on the First United States Microgravity Laboratory (USML-1) also indicated that convection was not totally damped. Clearly, reducing the magnitude of the gravitation force alone was not sufficient to achieve diffusion-controlled growth in these experiments. Other effects can also complicate the microgravity environment, especially those associated with time-dependent changes in the direction and magnitude of the gravity vector (g-jitter). These effects induce flow in the melt that is three-dimensional and time-dependent.

Another three-dimensional effect that arises during microgravity crystal growth experiments concerns the dewetting of the melt from the ampoule walls, resulting in bubbles or free surfaces. Such events can lead to significant Marangoni flows driven by gradients in surface tension along the free surface of the melt. Of interest in these situations are the effects of these three-dimensional flows on segregation and the effects of the free surface on reducing mechanical interactions between the solidifying charge and the ampoule. Growth experiments of $Cd_{0.96}Zn_{0.04}Te$ aboard USML-1 resulted in material that was far superior in structural perfection compared to earth-grown material under similar conditions. It was speculated that the near absence of hydrostatic pressure allowed for the melt to solidify with minimal wall contact, thereby eliminating deleterious wall interactions. To truly quantify the conditions needed to promote dewetting and explain the resultant phenomena associated with the presence of the melt free surface requires a fully three-dimensional modeling capability.

Previous attempts to model the effects of g-jitter and dewetting have relied on an assumption of 2D, axisymmetric behavior that greatly simplifies analysis. The proposed work will directly address the limitations of these models. The rigorous simulation of three-dimensional, transient effects will allow, for the first time, quantitative analyses of segregation phenomena in microgravity systems. This capability will be extremely important for the unambiguous interpretation of flight experiments, and, perhaps more importantly, will provide a tool to design the conditions needed in future flight experiments to best study the effects of microgravity on segregation theory.

Methodology and Results

We employ the finite element method coupled with a fully implicit time-integration technique to solve the transient, three-dimensional momentum, continuity, and transport equations. The Newton–Raphson iterative scheme is used to solve the resulting equation set at each time step, and the GMRES (Generalized Minimal Residual) iterative scheme is used to solve the linear systems arising in the Newton–Raphson iterations. We have implemented this algorithm on the Cray T3E, a massively parallel, distributed memory supercomputer, at the University of Minnesota Army High Performance Computing Research Center.

While we have made substantial progress in our capability to compute three-dimensional, transient flows, there are several areas in which our software needs to be extended to enable the detailed modeling of Bridgman crystal growth under microgravity. Two extensions have already been completed. In order to accurately represent effects caused by the ampoule and container, we have modified the code to solve for heat transfer through multiple domains with unequal physical properties. We have also implemented a rigorous, self-consistent methodology to solve for the location and shape of the moving melt/crystal interface location. These extensions have allowed us to compute the flow, temperature distribution, and interface location in a prototype problem of steady-state solidification in a solid-liquid system.

Several additional extensions are required to complete our code development. One extension is to solve for coupled, time-dependent mass transport, to enable segregation studies throughout a growth run. Another is to provide for realistic thermal boundary conditions by coupling our code with an appropriate representation of global (furnace) heat transfer. Also to be implemented are provisions to represent free-surface traction boundary conditions caused by surface tension gradients, so that three-dimensional Marangoni flows can be calculated. This approach will be needed to consider imperfections caused by bubbles or melt dewetting. We anticipate that implementing the above code modifications will be straightforward and have confidence in the current capabilities of the underlying algorithm and implementation.