Fundamental Studies of Solidification in Microgravity using Real-Time X-Ray Microscopy

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Objective

The objective is to obtain real-time dynamic data by utilization of X-ray Transmission Microscopy (XTM) to provide direct measure of the solute profile in the liquid, phase coalescence and growth in the liquid, and the detailed interface morphology (e.g., dendrites and cells) during solidification of metal alloys. We are also enhancing the XTM data with precise solid-liquid interfacial temperature and the thermal gradient measurement techniques, and working on the application of this technology to the study of the fundamentals of solidification in microgravity.

Relevance to Microgravity Research

There has been much interest in the study of solidification processes under the reduced sedimentation and convection environment available in low-gravity. This research applies a state-of-the-art X-ray Transmission Microscopy to image (with resolutions up to 3 micrometers) the solidification of metallic or semiconductor alloys in real-time. We have successfully imaged in real-time: interfacial morphologies, phase growth, coalescence, incorporation of phases into the growing interface, and the solute boundary layer in the liquid at the solid-liquid interface. We have also measured true local growth rates and can evaluate segregation structures in the solid. One example of a still unresolved microgravity finding is that directional solidification of on-eutectic alloy in low gravity often changes the eutectic interphase spacing, as well as eutectic grain size in equiaxed eutectic growth. During this study, the growth of eutectic and monotectic secondary phase fibers has been imaged in real-time during solidification for the first time in bulk optically opaque metal alloys. The ability to image these features in real-time will enable more fundamental and detailed understanding of solidification dynamics in microgravity than has previously been possible, thus allowing the full benefits of microgravity experiments to be applied towards rigorous testing of critical solidification models.

Significant Results

High resolution real-time X-ray Transmission Microscopy has been applied to obtain information fundamental to solidification of optically opaque metallic systems. We have reported the measurement of solute profile in the liquid phase growth, and detailed solid-liquid interfacial morphology of aluminum-based alloys with exposure times less than 2 seconds. *In-situ* X-ray imaging of Al-Pb and Al-In monotectic alloys showed, for the first time, that the isoconcentration lines of the solute boundary layer in metals are not necessarily parallel to the growth interface as has been assumed in some theories. Further, it was observed that striations in the solidified crystal may not decorate the interface position and shape. It was also shown that metal monotectic alloys at the monotectic composition do not necessary grow in a coupled manner. The ability of the XTM to integrate cross sectional microstructural features was utilized to study the detailed, previously

unreported formation of striations in Al-Pb. The process of morphological instability and cellular growth was imaged for Al-Cu and Al-Ag alloys. The dynamics of solid-liquid interface shape evolution near an insoluble particle was studied with XTM, showing the limits to the applicability of some analytical models. The results discussed above indicated that limitations of classical quench methods could be overcome, using XTM, providing more precise study of the dynamics.

The application of XTM for the study of solidification fundamentals was initially limited by a demonstrated resolution limit of 25 microns for real-time imaging. The study of the dynamics of formation of secondary eutectic and monotectic droplets and fibers requires a resolution on the order of 5 microns. This year we have accomplished advances in XTM furnace design that have provided the increase in real-time magnification (during solidification) for the XTM from 40X to 160X. The increased magnification has enabled, for the first time, XTM imaging of real-time growth of fibers and particles with diameters of 3-8 micrometers. We have applied this high resolution system to the study of the kinetics of formation and morphological evolution of secondary fibers and particles in Al-Bi monotectic alloys. We observed a previously unreported velocity dependent thermocapillary depletion mechanism for Bi-rich liquid which can penetrate many fiber diameters (over 100 microns) into the solid-liquid interface.

We also made what we believe is the first real-time observations of bulk solidification for optically opaque eutectic alloy Au-Al_2Au with 5 micron resolution. For this study we selected the on-eutectic composition of Al-7.4 atomic % Au. The alloy solidifies with Al_2Au fibers or plates (depending on growth rate) in an Al matrix. The high X-ray absorption difference between Au and Al provides excellent contrast for XTM. The tendency of the intermetallic phase to form ahead of the isotherm leads to an interesting observation that could only have been made by *in-situ* X-ray microscopy. The Al_2Au phase fibers remain extending into liquid from the solid-liquid interface after translation was set to zero velocity for a couple of minutes. We noted thickening of the Al_2Au phase fibers. Some fibers were about 25 µm thick and extended about 100 µm into the melt. It was observed over time (several minutes) that the coarsened, extended portions of the intermetallic phase loosened from the matrix and Stokes forces caused them to settle to the bottom of the crucible. The existence of such a process and subsequently its kinetics would be nearly impossible to study with conventional solidification and quench techniques. We are not aware that this phenomena has been previously reported.