Monitoring Al Solidification with High-energy X-rays

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Introduction

Approximately 68% of the aluminum produced in the United States is first cast into ingot prior to further processing into sheet, plate, extrusions, or foil. The primary casting process for these ingots has been direct chill (DC) semicontinuous casting. Though the DC process is, in principle, straightforward, the interaction of process parameters with heat extraction, microstructural evolution, and development of solidification stresses is too complex to analyze by intuition or practical experience. The *Aluminum Industry Technology Roadmap* has recognized the challenges inherent in DC casting and selected the development of "fundamental information on solidification of alloys to predict microstructure, surface properties and stresses and strains" as a high-priority need and "insufficient understanding of the aluminum solidification process; difficult to model" as a technology barrier in the aluminum casting process [1].

In order to address these challenges, a project team has been developed by Secat, a consortium of aluminum companies in the South and Midwest regions [2], with experimental and modeling efforts being carried out by national laboratory and university partners. As a participating member of the project, Argonne National Laboratory is developing techniques to directly probe microstructural and stress/strain evolution during aluminum solidification by using x-ray techniques at the APS. Here we report on the initial experiments carried out for this purpose.

Methods and Materials

We investigated three compositions of Al (pure Al, Al-4 at % Cu, and Al 3004 alloy) in the form of cylindrical ingots either 6 or 15 mm in diameter. (We only report on the Al-Cu, 15-mm-diameter sample here.) These ingots were placed in step-shaped graphite molds and pre-melted to flow into the step shape. This mold design was selected because of its well-known ability to generate tensile stresses in the aluminum alloy during solidification. During pre-melting, a thermocouple well was placed inside the ingots, providing temperature measurement along the sample length at ~25 mm intervals. Heating wires were wrapped around the top and bottom lengths of the graphite molds, with independent power sources, allowing formation of a controlled thermal gradient across the axial length of the sample.

These samples were investigated at the 1-ID line at APS by using an 80.72-keV monochromatic beam in transmission geometry (Fig 1). The 2-D diffracted intensity was recorded by using a Mar345 on-line image plate (3450×3450 pixels and 0.1-mm^2 pixel size) located 1365 mm from the samples. The use of high energies was important for two reasons: it allowed (1) sufficient penetration through the 15-mm sample diameter and furnace assembly and (2) forward compression of the Debye rings so that sufficient microstructural information could be captured with the 2-D detector (d_{min} of ~1A). Slits were used to define a transverse beam of 0.3×0.3 mm^2 without diffracted beam slits so that the entire longitudinal sample length of 15 mm was probed. Sample absorption was determined by measuring the beam intensity before and after the sample with an ion chamber and Si photodiode, respectively, and the samples were placed on xyz translation. Typical x-ray exposure times were 10 s, with about 100 s for detector readout.

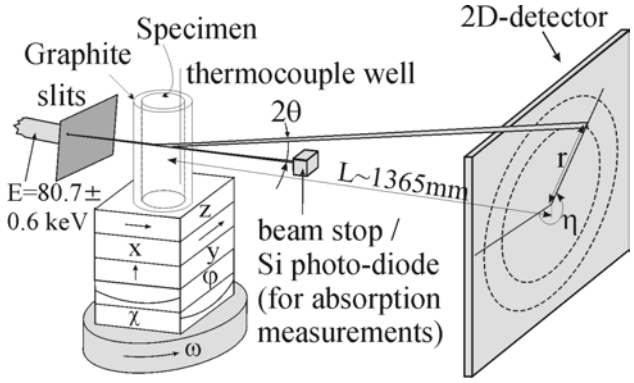

FIG 1. Experimental setup.

Results and Discussion

The furnace assembly was found to produce a stable temperature gradient, so that the liquidus and solidus boundaries could be probed by simply translating the sample along the cylinder axis (y-direction). Signature diffraction images of the Al-4 at % Cu sample taken across phase-field boundaries (after fully melting the

sample) are illustrated in Fig. 2, along with the partial phase diagram of the Al-Cu system. Diffraction spots from solidified Al and Al_2Cu phases are spotty, indicating that they arise from single grains. (We also smoothed the Debye rings by rotating the sample about the cylinder axis during exposure, but these images are not shown here.)

The liquid fraction of the sample as a function of temperature was also probed by using radiography (Fig. 3), which was permitted by density differences between the solid and liquid states. For these measurements, the sample volume was kept constant, and the temperature was steadily decreased as the transmission was measured in 10-s intervals. The solid fraction is seen to increase continually with decreasing temperature, over the temperature range $660 > T > 550°C$, showing the utility of this method to independently monitor solidification behavior.

Diffraction peak shifts were used to investigate thermal expansion coefficients (TECs) of the constituent phases. During heating, thermal expansion produces hydrostatic (direction-independent) peak shifts, which can be obtained by averaging the peak positions over the entire 2π azimuth measured. The peak positions for graphite (002) and Al (220) reflections were thus determined and converted to average lattice spacings a_{Al} and $c_{graphite}$ and are plotted as a function of temperature in Fig 4. While

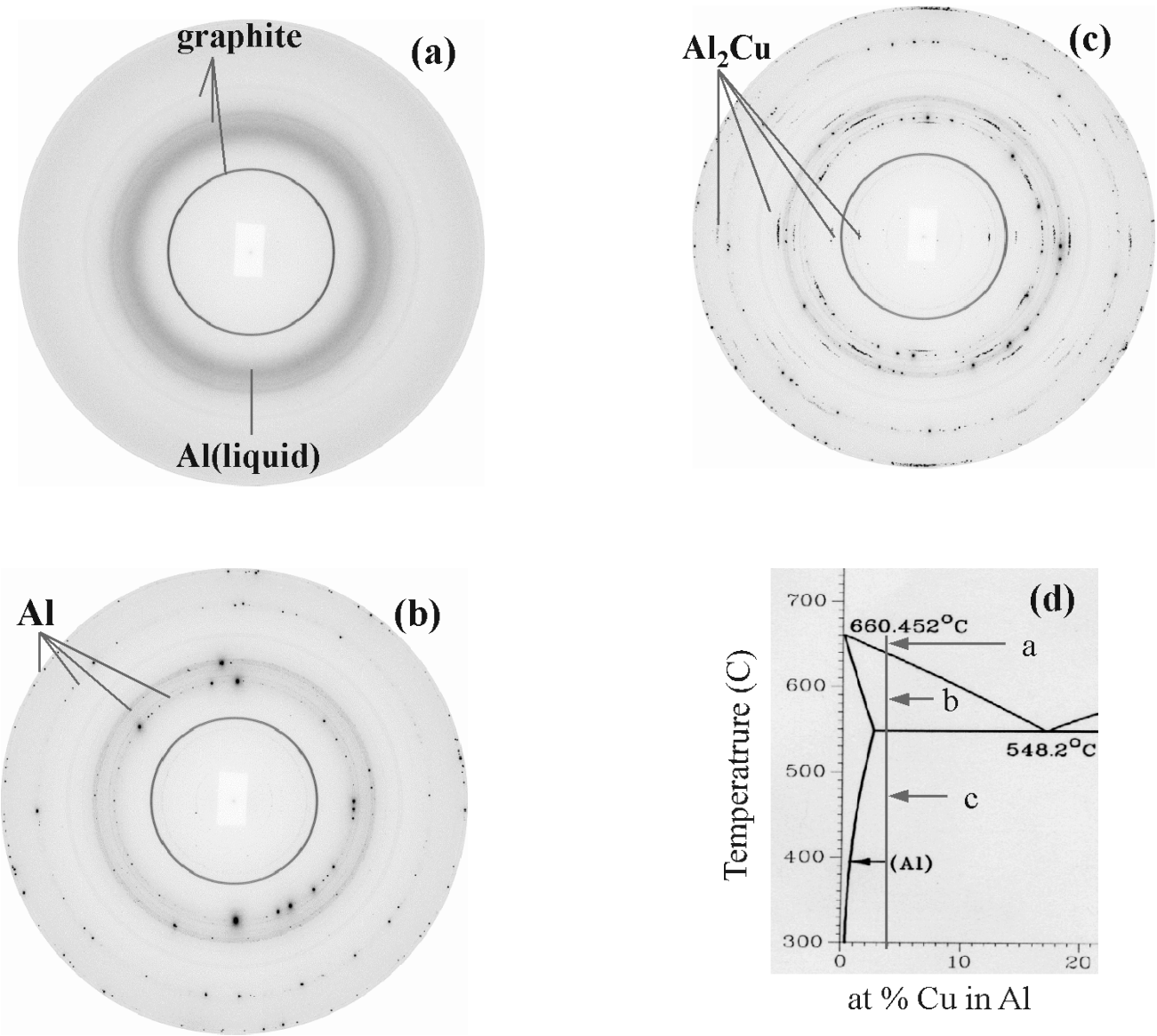

FIG 2. (a-c) Diffraction images during cooling and (d) partial phase diagram of the Al-Cu system showing composition and temperatures probed.

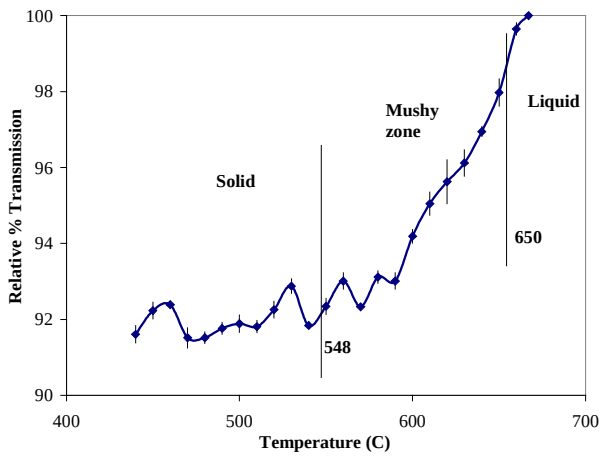

FIG 3. Transmission recorded during cooling across the 15 mm sample probe length.

$c_{graphite}$ behaves linearly up to 600°C, a_{Al} increases nonlinearly for $T > 480$°C and can be roughly divided into two regions, as shown in the figure. Linear fits provide $\alpha_{graphite} = 2.8 \times 10^{-6}$ K^{-1} and $\alpha_{Al} = 19.4$ and 49×10^{-6} K^{-1} below and above the solidus temperature, respectively. The TEC increases in the Al alloy are consistent with reported values for alloy AA1201 of 23.2 and 38.4 at $T < 500$°C and $T = 630$°C (solidus value), respectively [3].

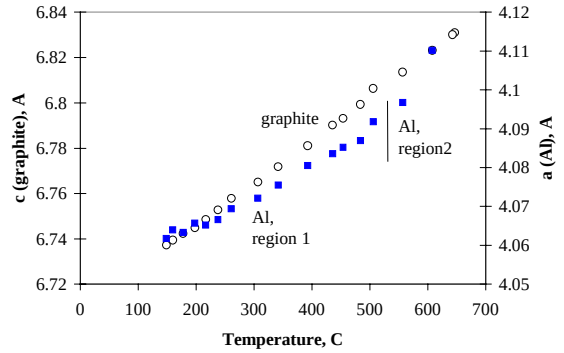

FIG 4. Lattice parameters derived from mean positions of graphite (002) reflections (open circles) and Al (220) reflections (solid squares) vs. temperature. While the graphite behaves linearly, the TEC of Al increases with temperature with a transition near $T_{solidus}$, as shown.

In closing, we mention a second set of experiments planned to more directly investigate strain and microstructural behavior in Al near solidification temperatures. These experiments would rely on three new developments: (1) use of a thermomechanical device, providing a controlled uniaxial stress (along the cylinder axis) at temperatures near the solidus; (2) incorporation of small-angle scattering (also at high energies) to investigate possible void and crack (hot tear) formation; and (3) use of a large-area charged-coupled device (CCD) (160 mm in diameter with 80-μm pixel size) for diffraction measurements, which should increase temporal resolution by an order of magnitude. The applied stress would be compared to measured internal strains/stresses in each phase (conversion of strains to stresses requires precise thermomechanical data, which are being determined by other project team members), providing information about stress transfer at elevated temperatures. In addition, we plan to use radiography to measure the nonequilibrium liquid fraction in these rapidly solidifying samples as a function of temperature.

Acknowledgments

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References

[1] Aluminum Association, Inc., *Aluminum Industry Technology Roadmap* (May 1997).
[2] For more information, visit www.secat.net.
[3] J.-M. Drezet and M. Rappaz, Metall. Trans. A **27A**, 3214 (1996).