| ESRF | Time resolved X-ray Imaging of Dendritic Growth in Binary Alloys | number: HS-1332 |
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| **Beamline:** ID22 | **Date of experiment:** from: 15.11.00 to: 20.11.00 | **Date of report:** 03.04.01 |
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Report:

Dendritic growth in 6 different alloys of the AlCu system were studied *in-situ* by direct beam time resolved X-ray imaging at ID22 EH2 using the super-FReLoN CCD detector in 4-channel read-out mode with a 11.0 $\mu$m transparent luminescent screen. The *in-situ* solidification conditions were controlled with a special rig, consisting of two furnaces, independently controllable in temperature and position, and a sample translation device. An outline of the working principles of the rig can be found elsewhere [1], however the version used for the studies reported here has been modified to overcome problems encountered in prior experiments (MI-416). Measurements were carried out with $\sim 1 \times 1$mm$^2$, monochromatic beams between 12-15 KeV, at sample pos. $\sim$ 60 m from the source. The sample-detector distance was fixed at 55cm. Temperatures in the range 840-1000 and 720-870 K were employed for the hot and cold furnaces, respectively. The super-FReLoN array was read both in unbinned and $2 \times 2$ binning mode, readout times of $\sim 0.75$ and $\sim 0.2$ s/frame, respectively. Exposure times were between 0.7-0.8 and 0.1-0.35 s/frame for unbinned and binned read-outs, respectively. A $10\times$ magnifying lens was employed throughout the experiment, leaving the resulting spatial resolution for unbinned images to typically $\sim 1\mu$m, aberrations excluded.

Alloys of Al30%wtCu, Al20%wtCu, Al10%wtCu and Al6%wtCu, the latter two made in two batches with and without $TiB_2$ grain refiners added. All samples were placed in rectangular $25 \times 15$mm$^2$ quartz glass containers - alloy thicknesses $\sim$150 $\mu$m $\parallel$ X-ray beam. About 50% of the samples were equipped with internal thermocouples that registered local temperatures in the melt/solid at 2 Hz with an accuracy of $\pm$0.5K. In experiment MI-416 we succeeded in preventing Al from oxidizing in contact with the the quartz by pre-oxidation the samples through heat treatment to form a sub-$\mu$m thick protective inert layer of $Al_2O_3$ at the glass-alloy interface. However, this time the oxide did not prevent such reactions to take place. During the first 3-4 shifts of HS-1332 we destroyed several samples without locating one that could sustain the 2-3 time series required for alignment of a semi-stationary solidification front in the field of view. In search for better alternatives, one of the remaining samples was taken out of its glass container, polished slightly, spray coated with Boron nitride (BN) before being reinserted in to the container. Subsequent tests revealed that the BN-coating prevented quartz-Al reactions, and just as important - the coating did not form structures giving rise to additional X-ray transmission contrast.

In total 113 time-series of solidification were acquired. Dendritic and eutectic interfaces were studied at various growth rates by simultaneous adjustments of temperatures and inter spacing for the furnaces as well as the sample translation velocity.
From the different alloy compositions investigated, only the Cu-enriched ($\geq$ 10%wt) solidified with the quasi-2D phase boundary structure required for measurements to be applicable for modeling and quantitative extraction. In the 6%wtCu-alloys, the Cu-consentration was inadequate to form a quasi-2D solid-liquid interface in the plane orthogonal to the beam. The samples of 10%wtCu with grain refiners added were also unsuccessful due to the formation of a 3D equiaxed interface, but as a result of too large a number of nucleation sites rather than of an inadequate Cu concentration. The studies with BN coated alloys at Cu concentrations $\geq$ 10%wt were all successful. In total 64 time series of solidification fronts were acquired with these samples, with image qualities and spatiotemporal resolutions limited by the FReLoN setup (readout/storage time) and the ESRF beam stability (20 Hz flickering), rather than by our samples and/or control over temperature- and interface velocity.

In *ab-initio* 2- and 3D modelling of dendritic phase front propagation the input diffusion field is 1D, directed normal to the interface at each point from which the time-space coordinates of the boundary are calculated numerically. Such a model restricts the liquid undercooling to be small *wrt.* the numerical resolution, as each grid of the interface must diffuse latent heat into separate liquid segments. In this framework, interactions among nearby interfacial features cannot be accounted for. At larger undercoolings a vector field description of the diffusion is neccessary.
Image processing of the high-quality time-series followed by a sobel-based edge detection algorithm has been used to extract coordinates for the propagating dendrite-liquid and eutectic phase boundaries. We are currently developing an algorithm for joining split segments of the phase front, subsequently to be followed by a smoothing before final coordinates are extracted for modeling of a 2D diffusion field. With the extraction of a 2D diffusion field based on experimental phase front coordinates we aim at developing a consistent model accounting for interactions between interfacial features. Apart from the phase boundary propagation, the series uncover several other details, such as convection and $H_2$ gas porosity formation. A publication summarizing the experimental details is in preparation. Further, we anticipate a potential for several separate publications to emerge from a new model. At a later stage, more beam time may be required to fill in eventual gaps. In Figure 1 typical images of a time-series is shown.

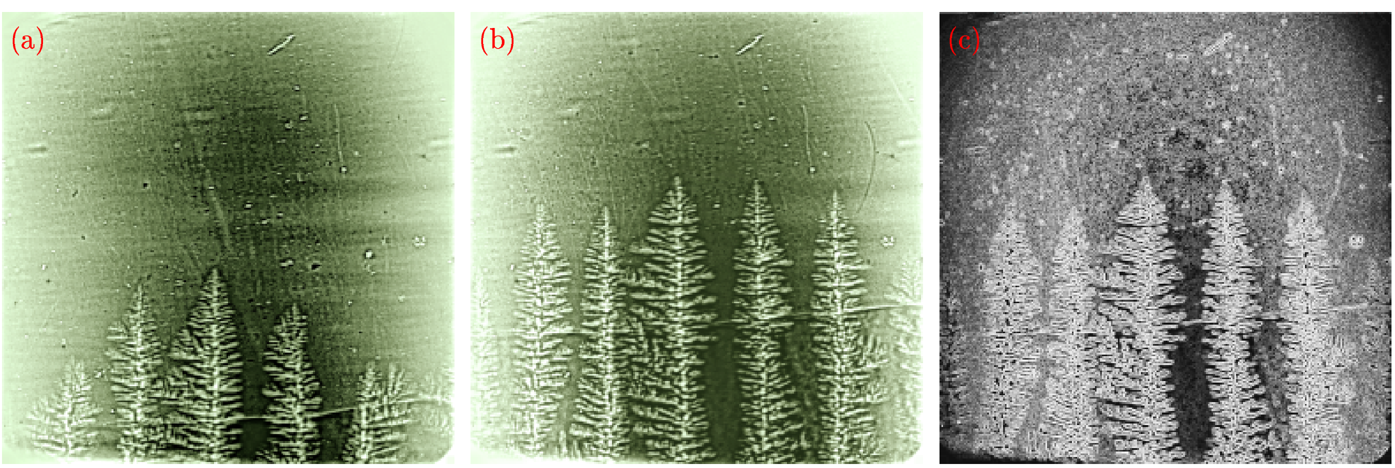


**Figure 1.** Directional dendritic growth in the Al30%wtCu alloy. Temp. gradient $\simeq$ 28 K/mm, exp. time 0.4 s/frame, sample trans. velocity $v_{||z} \simeq 6.4\mu$m/s. A propagating dendritic front at (a) $t_0$ and (b) 40 frames/56 s later. (c) shows the edges detected from image (b).

## References

[1] R.H. Mathiesen *et al.* *Phys. Rev. Lett.* **83** , 5062 (1999).