ESRF	Timeresolved X-ray imaging of dendritic growth in binary alloys	**Experiment number:** MI-416
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Report:

Dendritic growth in two different alloys of the AlCu system were studied *in-situ* by direct beam timeresolved X-ray imaging at ID22 EH2 using the super-FReLoN CCD detector in 4-channel read-out mode with a 3.5 μm transparent luminescent screen. For controlling the solidification process a special rig, consisting of two furnaces, independently controllable in temperature and position, and a sample translation device was used. A more detailed description of this rig can be found elsewhere [1]. Experiments were conducted with a $\sim 1 \times 1\text{mm}^2$, 20 keV monochromatic beam, approximately 60 m from the source, with sample-detector distance fixed at 60cm. Temperatures in the range 940-1120 and 670-800 K were employed for the hot and cold furnaces, respectively. The super-FReLoN array was read exclusively in the 2×2 binning mode, giving a constant readout time ~ 0.2 s/frame, while exposure times were varied between 0.05-0.5 s/frame. Only the 10× magnifying lens was employed in the experiment, leaving the resulting spatial resolution to typically $\sim 1\mu$m, aberrations excluded.

Alloys of Al30%wtCu and Al3%wtCu were placed in rectangular $25 \times 15\text{mm}^2$ quartz glass containers - alloy thicknesses $\sim$150 μm $\parallel$ X-ray beam. To prevent Al from being oxidized from contact with the the quartz, the alloys were pre-oxidized by heat treatment to form a sub-μm thick protective inert layer of Al_2O_3 at the glass-alloy interface. Some samples were made with internal thermocouples to monitor local temperatures in the melt/solid during the growth processes. These particular samples were made by splitting the alloy in two 7.5 mm wide pieces, inserting the thermocouple into a narrow column between these two halves, and directly in contact with the oxide layer, all encapsulated by the glass. The thermocouple-oxide contact surface was made as small as possible, $r_{\text{contact}} \sim 60\mu$m, to minimize heat leakage to the thermocouple from the solidifying system.

In total 28 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. Samples with internal thermocouples functioned as intended. In Figure 1 a typical time-series of the solidification process in Al30%wtCu is illustrated. The images have been flat-field corrected and filtered with a nearest-neighbour equalizer.

During the experiments we encountered several problems that combined to prevent optimal control over the experiment. At an early stage, a short-circuit failure in one of the heating elements of the hot furnace made it necessary to improvise on the setup. The temperature control became significantly restricted – and as a result the solidification studies had to be performed in temperature gradients larger than desired, especially for the Al3%wtCu. The large gradients gave considerable thermal convection, making equiaxed dendrites rotate and translate during exposure. In several cases this prevented thorough studies of the spatiotemporal evolution of the solid-liquid interface. Yet, on the other hand the observation of convective currents illustrates the eminence of *in-situ* studies, as such effects although known to occur has not been observed previously. Another difficulty occurred at 0.05-0.15 s exposure times, where the incident beam was seen to oscillate in position. This corresponds to the $\sim 20Hz$ flickering of the ESRF beam. It prevents full advantage to be taken from the time resolution offered by the 4-channel read-out/direct memory storage, since a proper flat-field correction becomes impossible. The beam oscillation amplitude and a full beam cross-section field-of-view leave too much unaccountable beam structure in the images to use them for quantitative analysis. With a 1×1 mm^2 field-of-view, exposure times shorter than 0.15 s do not provide the image quality necessary for assessment of physical parameters from image-supported simulations of the growth processes.

Another unexpected observation was the formation and growth of gas bubbles in the liquid phase. Oxygen from water vapor present during the process may form Alumina, leaving H_2 gas that segregates during solidification, and causes porous alloy microstructures.

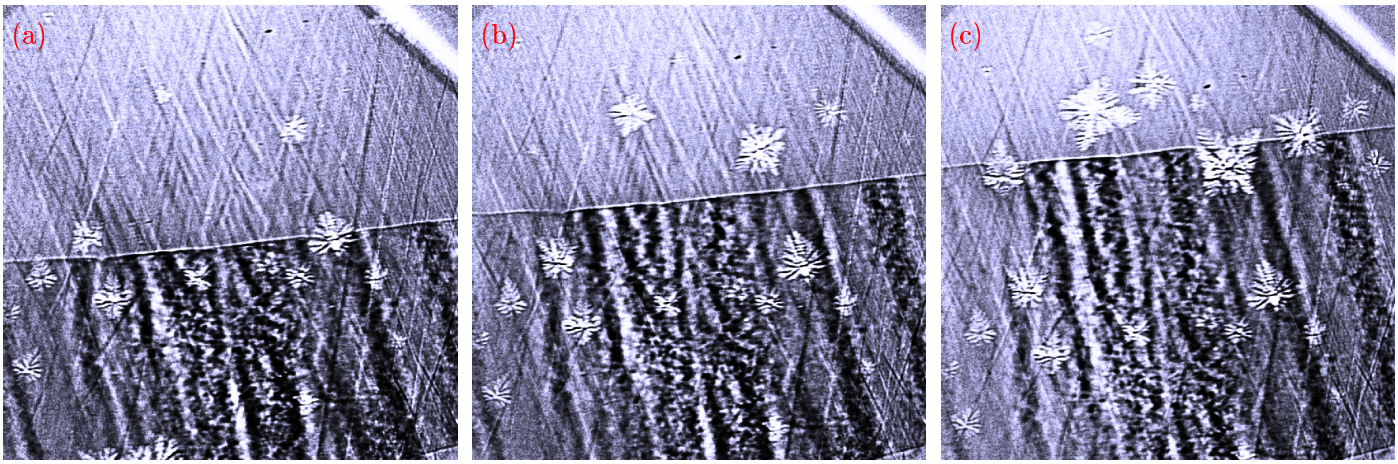

Figure 1. Equiaxed dendritic growth in the Al30%wtCu alloy. (a) t_0, (b) $t_0 + 1.05$s, (c) $t_0 + 2.1$s. Temp. gradient $(\partial T/\partial z) \simeq 30$ K/mm, exp. time 0.15 s/frame, sample trans. velocity $v_{\|z} \simeq 12\mu$m/s. A planar eutectic solid-liquid interface is present in the images, succeeding the dendritic front. The resulting eutectic microstructure is segregated with Cu-enriched columns. The eutectic growth occurs at thermal equilibrium, while the equiaxed dendrites grow from thermal diffusion in a super-cooled liquid. Other structures apparent in the images, originates from wrinkles in the protective oxide layer and from the beam shutter.

The data are still being processed, but it is evident that some of the measurements have sufficient spatiotemporal resolutions and are otherwise of adequate quality to serve as input for numerical simulations of dendritic growth processes. A manuscript for a publication on the experimental observations is currently under preparation.

References

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