ESRF	**Experiment title:** Microtomographic determination of the intercrystalline size-correlation function in a polycrystalline metal	**Experiment number:** ME-142
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Report:

The goal of experiment ME-142 was to quantify the intercrystalline grain-size correlations in a real, three-dimensional polycrystalline material using microtomography. Recent theoretical considerations and the results of large-scale computer simulations indicate that nearest-neighbor size correlations are an important—albeit heretofore largely neglected—factor governing the kinetics of the technologically important process of grain growth in polycrystalline solids. Until now, these correlations could be evaluated only in microstructures generated by computer simulation. In real materials, the local grain-size correlations cannot be studied using the standard characterization techniques of optical and scanning electron microscopy, because two-dimensional slices through a three-dimensional structure lack the necessary local size information. As a volumetric imaging technique, microtomography potentially offers direct access to the three-dimensional network of grain boundaries in a polycrystalline sample. A map of this network would reveal the size of each grain and the contact areas between neighboring grains, thereby providing the information needed to calculate the desired correlation function.

In order to image grain boundaries by microtomography, it would suffice to mark them with atoms that absorb x-ray radiation much more strongly than the atoms in the neighboring grains. The classic example for such a system is aluminum alloyed with a small amount of tin. Aluminum and Sn are immiscible in the solid state, and Sn is known to segregate to the grain boundaries of Al upon solidification of a melt containing the two elements. Since Sn has a much higher x-ray attenuation coefficient than Al, it should be possible to measure the spatial distribution of Sn in such a sample using absorption-contrast microtomography.

Ingots of Al-Sn with 1, 2 and 3 at.% Sn were prepared by annealing pieces of pure Al and Sn under vacuum at 750°C in an alumina crucible and then allowing the melt to cool slowly (over ~ 2 h) to room temperature. Pieces cut from the ingots were then remelted and cooled at a much faster rate (less than 1 min) in order to suppress grain growth. Scanning-electron-microscopy images of slices through the ingots revealed that the Sn atoms segregated completely to the boundaries of the Al grains, regardless of the cooling rate. The average grain size of the "slow-cool" specimens was about 150 μm, whereas that of the "fast-cool" samples was ~ 50 μm. Furthermore, the grain size was found to be uniform throughout the entire volume of each ingot, and the size was independent of the overall Sn concentration. In preparation for the tomographic measurements, samples of approximate dimension

$0.5 \times 0.5 \times 5$ mm^3 were cut from the ingots with a wire saw and glued to the ends of stainless steel posts (1 mm in diameter), which were then inserted into wax-filled goniometer sleeves.

Microtomographic measurements were carried out at beamline ID22 at energies between 15 and 25 keV with a combination of scintillator, magnification and rebin parameters chosen to yield an effective detector pixel dimension of 1.4 μm and a field of view exceeding 1.4 mm. Typically, 625 projections were recorded with exposure times between 3 and 10 sec, and then the sample was shifted along its long axis by 1 mm and the process was repeated up to four times. In this manner, we hoped to be able to reconstruct contiguous volumes much larger than that obtained from a single tomographic measurement.

Figure 1 illustrates the tomographic reconstruction of a section of a "slow-cool" sample of $Al_{98}Sn_2$. The threshold was set to reveal the spatial distribution of Sn segregated to the grain boundaries and triple junctions (intersections of three grain boundaries). Although the individual Al grains are clearly evident as regions devoid of absorption contrast, the coverage of grain boundaries by Sn atoms is rather nonuniform. The presence of gaps in the boundaries greatly complicates the determination of grain sizes and shapes in such images using an automated routine. The tomographic reconstructions of the "fast-cool" specimens are qualitatively similar to those of the "slow-cool" samples.

Owing to the large amount of computational time needed to generate a tomographic reconstruction, it was not until February 2001 that we were able to finish reconstructing the data generated in experiment ME-142. Therefore, we can submit only a preliminary report of our experimental findings at this time. Determination of the intercrystalline grain-size correlation function from tomographic reconstructions like that of Fig. 1 requires finding a way to "vectorize" the network of grain boundaries made visible by the segregated Sn. This task would be much easier if the unevenly marked grain boundaries could be removed from the image, leaving only the network of triple junctions. By filtering the reconstructed image with a Fermi function, we found that the range of image intensities associated with the Sn atoms can be "stretched" such that a threshold value can be set to distinguish between Sn in the grain boundaries and that in the triple junctions (Fig. 2). Once the positions and connectivities of the vertices of the triple-junction network have been determined, we can calculate the entire grain-boundary network using a surface-energy-minimization routine [1]. The resulting vectorized structure contains all of the information necessary for computing the desired intercrystalline size-correlation function.

[1] C. Monnereau and M. Vignes-Adler, *Phys. Rev. Lett.* **80** (1998) 5228.

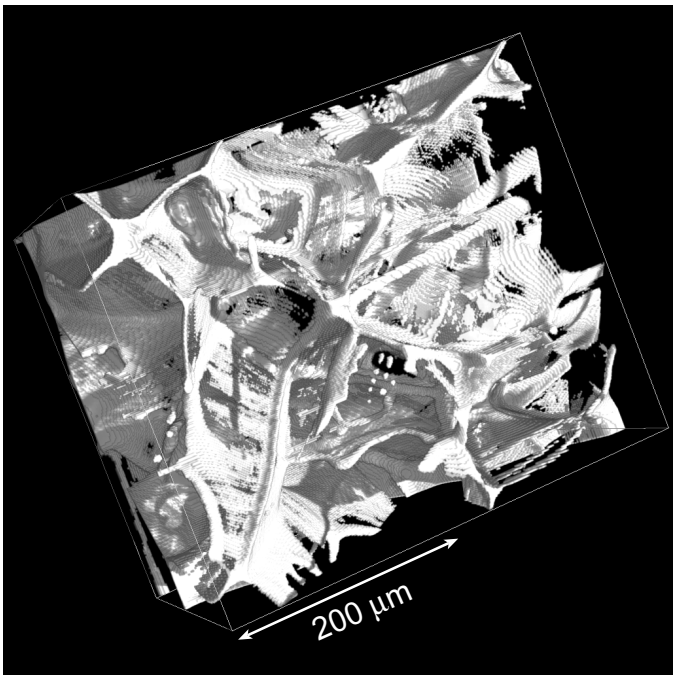

FIG. 1: Tomographic reconstruction of the absorption-contrast distribution in "slow-cool" $Al_{98}Sn_2$. The holes evident in several grain boundaries arise from regions of low Sn segregation.

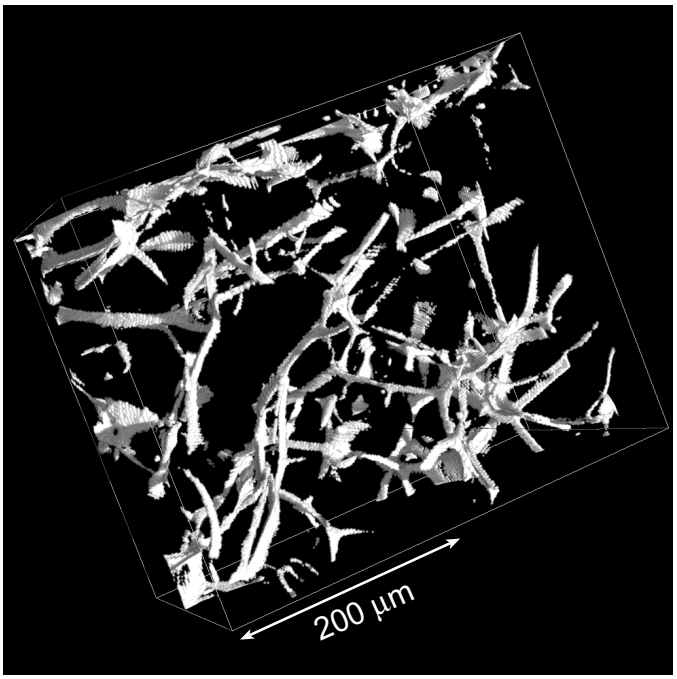

FIG. 2: Same region as in Fig. 1, processed to image only the triple junctions. In principle, the network of grain boundaries can be reconstructed from the vertices of the triple junctions and their connectivities.