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ESRF	**Experiment title:** Materials Science Applications of the 3DXRD Microscope	**Experiment number**: ME-268
Beamline:	**Date of experiment**: from: 1/9 2001 to: 1/9 2002	**Date of report**: 1/3/2002
Shifts:	**Local contact(s)**: S. Grigull, L. Margulies	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

H.F. Poulsen*, L. Margulies*, S. Schmidt*, E.M. Lauridsen*, S.F. Nielsen*, D. Juul Jensen, G. Winther, W. Pantleon, R.V. Martins*, Materials Research Dept., Dk-4000 Roskilde.

Report: We present a number of highlights based on work performed on the dedicated 3DXRD instrument. Much of this work has consisted of building on previous feasibility experiments toward collecting statistically significant data sets on the behaviour of individual grains during thermo-mechanical processing. In addition, initial studies have been performed towards increasing the spatial resolution of 3D grain mapping, as well as studying the dynamics of sub-grain structures.

In-Situ Measurement of Grain Rotation during Deformation of Polycrystals using a Conical Slit

Experimental data on the rotation pathway of individual bulk grains during plastic deformation is critical in guiding current modelling efforts in polycrystalline deformation. Recently we reported on the first such measurements on the rotation of grains embedded in the bulk of an Al polycrystal during tensile deformation [1]. These results on 4 imbedded grains showed that neither the classical Taylor nor Sachs models could adequately account for the observed rotations. In this first experiment, diffraction spots were recorded which originated from grains throughout the entire intersected cross section between the sample and incident beam. It was found that due to broadening of reflections at increasing strain, a maximum cross section of 10 grain lengths was allowable to prevent significant spot overlap at strains approaching 10%. Hence, a significant percentage of the reflections belonged to grains at or close to the surface, which had to be rejected due to their non-representative nature. This restriction also set a lower limit on the grain size which could be examined.

Here we report on an extension of the above technique which alleviates these limitations by adding an extra optical element, a conical slit. The conical slit is aligned between the sample and detector and acts to define a gauge volume within the bulk of the sample, which will give rise to diffraction spots on the detector. Diffraction from volumes outside this gauge volume will be rejected by the slit. In this way we can provide a three dimensionally resolved reference volume within the sample, and limit the spot overlap problem which would otherwise be faced when examining thicker or finer grained samples. The conical slit is used in conjunction with a point-focused beam which leads to a gauge volume of approximately 5x5x250 μm^3. By

assuring that the gauge volume is aligned on the center of rotation, and that the sample is probed far from the surface, we can increase the number of valid grains measured at each position. In this way we can scale up the efficiency of the technique in order to provide the statistics needed for model development.

The data presented here were taken on a sample of 99.5% percent pure Al with a thickness of 4 mm and an average grain size of 75 µm. Data was collected at 0, 2, 4, and 6 % strain. The indexing program GRAINDEX[3] was used to sort the reflections by grain and calculate their orientation. Fig 1 shows the rotation of the tensile axis for 95 embedded grains measured on a single sample. This analysis resulted in subdivision of the triangle into four different regions, each having a certain main rotation trend:

- Grains in the <110> corner rotate systematically towards the <100>-<111> line
- Grains at the <100>-<111> line rotate along this line towards the <111> corner but with more scatter than observed in the <110> corner
- Grains half way up the <110>-<111> line rotate directly towards the <111> corner without much scatter
- Grains in the <100> corner of the triangle rotate in an apparently random manner

It is therefore concluded that at least at low strains, the rotations are dominated by the lattice orientation and not by grain interaction. Grain interaction may, however, be the origin of the smaller variations found within each of these regions. These results represent the first experimental data containing both sufficient detail and statistics to distinguish between the existing plasticity models.

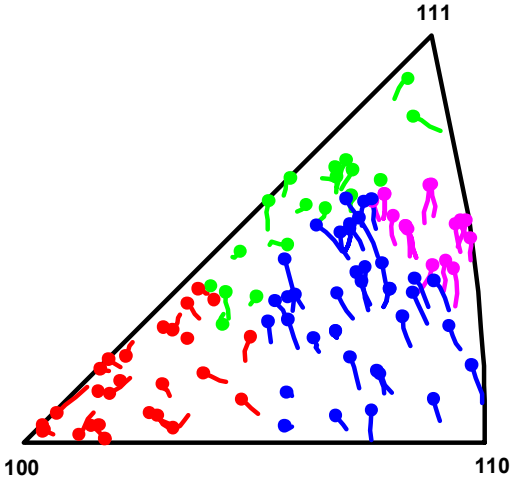

Fig. 1. Stereographic triangles showing the rotation of the tensile axis of individual bulk grains during tensile deformation measured in three experiments on different fcc metals. The circles mark the final orientation of the grains[14].

Orientation Dependence of the Recrystallization Kinetics of Commercial Purity Aluminium

For fcc metals of medium to high stacking fault energy, it is well known that during recrystallization the volume fraction of grains with the *cube* orientation increases significantly, and in some cases completely dominate the final recrystallization texture. We have used the 3DXRD microscope for *in-situ* studies of the recrystallization kinetics of a 90% cold-rolled commercial purity aluminium alloy (AA1050) in order to examine the dynamics of such recyrstallized texture in more detail [6]. In total the growth curves and crystallographic orientation of 244 individual grains were analysed, resulting in 14 *cube* grains, 124 grains belonging to the so-called *rolling* texture components, and 106 grains having other orientations (also referred to as *random* orientations).

From each of the 244 growth curves, we can determine fundamental recrystallization parameters such as the nucleation time, the initial growth rate and the final grain size. The distribution of nucleation times and initial growth rates are shown in Fig 2 and 3, respectively, in both cases classified with respect to the three texture components. In Fig 2, the majority of the nuclei are seen to form at the very beginning of the annealing period in agreement with the idea of site saturation normally assumed for this type of alloy. There are, however, a number of nuclei with significantly longer nucleation times. Within error the three distributions are identical.

In fig. 3, each of the distributions are seen to be broad, exibiting a range of growth rates over an order of magnitude. Such a distribution is inconsistent with all conventional Avrami-type recyrstallization models. In this case, there are clear differences in the shape of the 3 distributions, with the *cube* grains and *rolling* grains on average having a higher initial growth rate than *random* grains.

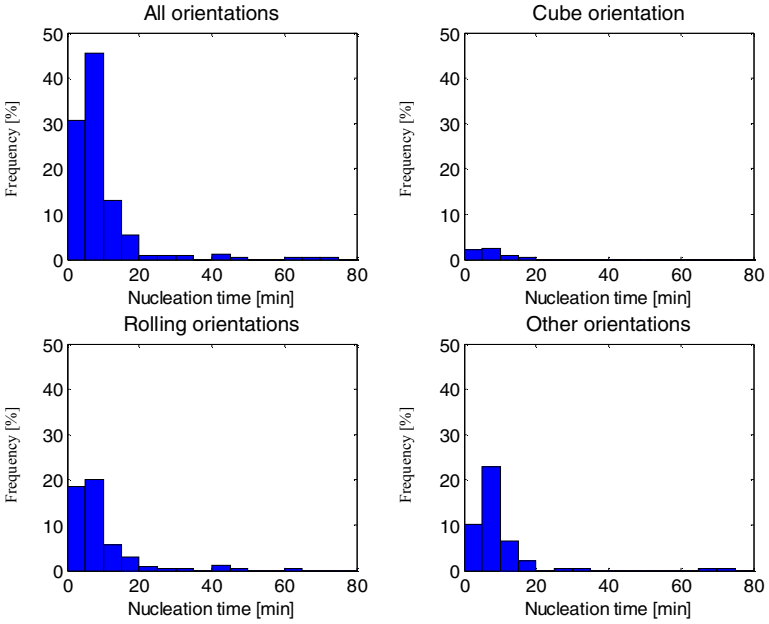

Fig. 2. Histograms of nucleation times (in minutes). The frequencies based on all grains is shown in the upper, left corner. Next, the grains are sorted into three texture groups; cube, rolling and other. The respective fractional frequencies are shown. The result of a Kolmogorov-Smirnof test is that the 4 distributions are identical within error.

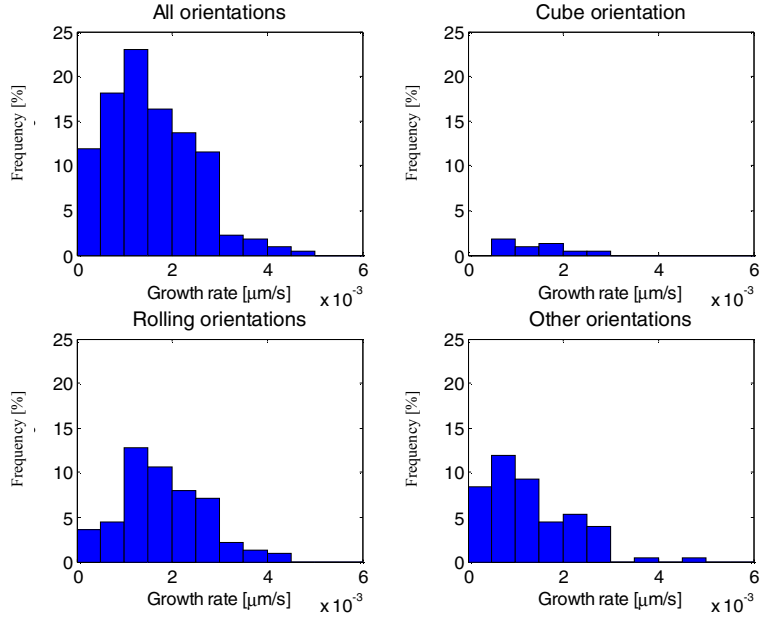

Fig. 3. Histograms of initial growth rates in µm/s (for t = 600s). The frequencies based on all grains is shown in the upper, left corner. Next, the grains are sorted into three texture groups; cube, rolling and other. The respective fractional frequencies are shown. Grains having cube and rolling orientations are seen on average to have a higher initial growth rate than grains with random orientations.

Development of Reconstruction for 3D Grain Maps

The spatial resolution of 3D grain maps is limited by both the resolution of the detector and the robustness of the reconstruction algorithms applied. Recently new optics have been added to the high resolution detector which has increased the spatial resolution of the camera to 6 microns. In order to take full advantage of this resolution new software has been developped for the generation of 3D maps. We here report on the establishment of a mathematical formalism needed for developing a general-purpose reconstruction algorithm [15]. A six-dimensional space is introduced, defined as the product of direct space and orientations space, parameterised by Rodrigues vectors. The task is to derive the density of this space from a set of two-dimensional images. This task is similar to reconstruction problems in absorption or emission tomography. However, it is substantially more complex, due to the reconstruction space being a 6-dimensional curved space and the sampling being not on a regular grid, but defined by the crystal symmetry operations. Furthermore, discretization is an issue. With orientation elements of $(0.5 \text{ degree})^3$ over 100x100x10 positions in a sample a set of $\sim 10^{11}$ 6D-voxels is required.

The projection surface in the 6D space has been derived. The projection surface is a manifold, defined by the set of points that can contribute to the intensity in a given pixel at a given diffractometer setting. It can be characterised as a combination of conical sections. In direct space it comprises a set of cones, a mirror image of the Debye-Scherrer cones for a hypothetical powder placed at the pixel position. In Rodrigues space it comprises a set of straight lines embedded in a curved envelope surface.

A formulation in terms of a constrained set of linear equations is found to be ideal. It applies to the geometry at hand and can deal with additional constraints such as detector point-spread-functions. With an iterative solution, such as a version of the ART routine, it can be up-scaled. Furthermore corrections for absorption can be included. Importantly, it can be extended to handle combined diffraction and absorption contrast tomography data.

As a feasibility study an ad-hoc MATLAB program was made based on the ART algorithm. This was applied to data obtained with the 3DXRD microscope. Again the geometry was simplified by investigating an un-deformed polycrystal, where the grains had a negligible mosaic spread. In this case the incoming beam was focused in one direction such that it defined a layer in the specimen. Each layer was therefore reconstructed independently. Furthermore there was only limited overlap between diffraction spots. Using the indexing program GRAINDEX [3] the spots could be sorted according to grain of origin. The reconstruction can then be performed grain by grain.

The experiment was performed on Al using a monochromatic 50 keV beam. The pixel size and the FWHM of the point-spread function were 2.3 μm and 6 μm, respectively. The results for 4 neighbouring grains in a randomly chosen layer are shown in Figure 4. From the amount of overlap and voids between grains we infer a spatial resolution of ≤ 5 μm. 3D maps are presently being generated.

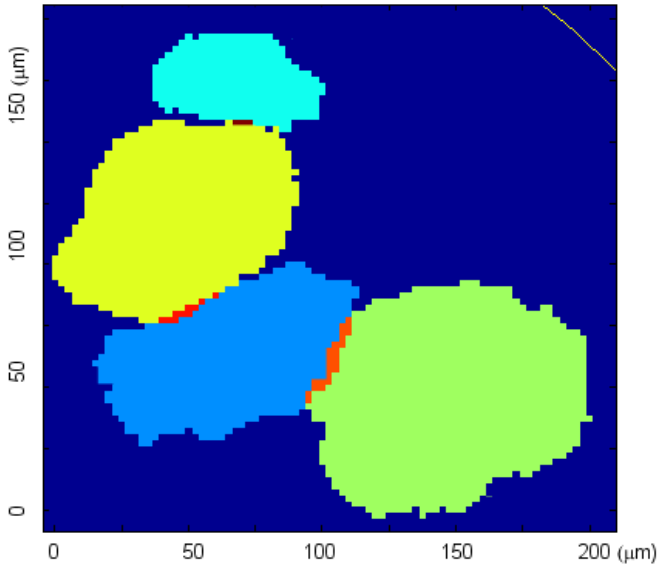

Fig. 4.
Independent reconstruction of 4 neighbouring grains marked by different colours. Regions of overlap between the grains are marked by red. The surface of the polycrystal is indicated by a solid yellow curve.

We plan to pursue the generation of a reconstruction program for the general case in collaboration with the group of Herman, N.Y., who for the last 30 years has been active in developing the methodology behind medical scanners.

Highlights from other work:
The data analysis on this work has still not been completed

- First ever study on the 3D morphological changes occurring to a single nucleus as it grows during recrystallization. The observed behavior sheds new light on the nature of the interaction between nuclei and the deformed matrix.
- First ever study on the bulk dynamics of dislocation structures during recovery.
- First ever study on the coarsening of sub-micron grains produced by severe plastic deformation on annealing.

- First study ever on the dynamics of grains during dynamic recrystallization, for the case of tensile deformation of Pb.
- A statistical study of the dynamics of the evolution of the elastic strain tensor for between 10 and 100 grains as a function of load. This data set is complementary to the rotation data presented above.
- A feasibility study showing that true 3D grain size distributions can be determined with 3DXRD. This is in contrast to surface techniques, which only produce 2D information. A potential is inferred for applied work.

Risø 3DXRD Publications 2001-2002

[1] L. Margulies, G. Winther, H.F. Poulsen. *In situ Measurement of Grain Rotation During deformation of Polycrystals.* Science, **291**, 2392-2394 (2001).

[2] H.F. Poulsen, D. Juul Jensen, T. Tschentscher, L. Wcislak, E.M. Lauridsen, L. Margulies, S. Schmidt. *Quantifiaction of minor texture components by hard x-rays.* Textures & Microstructures, **35**, 1, 39-54 (2001).

[3] E.M. Lauridsen, S.Schmidt, R.M. Suter, H.F. Poulsen. *Tracking: a method for structural characterization of grains in powders or polycrystals.* J. Appl. Crys. **34**, 6, 744-750 (2001).

[4] H.F. Poulsen, S.F. Nielsen, E.M. Lauridsen, S. Schmidt, R.M. Suter, U. Lienert, L.Margulies, T. Lorentzen, D. Juul Jensen. *Three-dimensional maps of grain boundaries and the stress state of individual grains in polycrystals and powders.* J. Appl. Cryst. **34**, 6, 751-756 (2001).

[5] L. Margulies, T. Lorentzen, H.F. Poulsen, and T. Leffers. *Strain tensor development in a single grain in the bulk of a polycrystal under loading.* Acta Mat. **50**, 1771-1779 (2002).

[6] E.M. Lauridsen, S. Schmidt, L. Margulies, H.F. Poulsen, and D. Juul Jensen. *Growth Kinetics of Individual Cube Grains as Studied by the 3D X-Ray Diffrction Microscope.* Proc. of 1st Int. Conf on Recrystallisation and Grain Growth. Eds. G. Gottstein and D.A. Molodov, Springer-Verlag 2001, **1**, 589-594 (2001).

[7] D. Juul Jensen. *Experimental Measurements of Nucleation and Growth Rates*. Proc. of 1st Int. Conf on Recrystallisation and Grain Growth. Eds. G. Gottstein and D.A. Molodov, Springer-Verlag 2001, **1**, 73-86 (2001).

[8] S.F. Nielsen, E.M. Lauridsen, D. Juul Jensen, and H.F. Poulsen. *A three-dimensional X-ray diffraction microscope for deformation studies of polycrystals*. Mater. Sci. Eng. A **319-321**, 179-181 (2001).

[9] H.F. Poulsen and D. Juul Jensen. Mat. *From 2D to 3D microtexture investigations*. Science Forum **408-412**, 49-66 (2002).

[10] G. Winther, L. Margulies, H.F. Poulsen, S. Schmidt, A.W. Larsen, E.M. Lauridsen, S.F. Nielsen and A. Terry. *Lattice rotations of individual bulk grains during deformation.* Mat. Science Forum **408-412**, 287-292 (2002).

[11] S. Schmidt, H.F. Poulsen, G.B.M. Vaughan, *Structural refinement of the individual grains within polycrystals and powders.* J. Appl. Cryst, In Press.

[12] H.F. Poulsen, E.M. Lauridsen, S. Schmidt, L. Margulies, J.H. Driver. *3D-Characterisation of microstructure evolution during annealing of a deformed aluminium single crystal*. Acta Mat. In print.

[13] S.F. Nielsen, S. Schmidt, E.M. Lauridsen, H. Yiu, J. Savoie, M. Zeng and D. Juul Jensen. *Growth kinetics of individual grains during recrystallization with an intermediate cooling cycle*. Scripta. Mater. In print.

[14] H.F. Poulsen, L. Margulies, S. Schmidt, G. Winther. *Lattice rotations of individual bulk grains. Part I: 3D X-ray Characterization*, submitted.

[15] H.F. Poulsen, X. Fu. *Generation of grain boundary maps by an algebraic reconstruction technique*. J. Appl. Cryst., in print.