	Experiment title: **Recovery and recrystallisation kinetics in ultra fine grained Ti processed by severe plastic deformation**	**Experiment number**: MA693
Beamline: **BM01A**	**Date of experiment**: from:15.04.09 to:20.04.09	**Date of report**: 30.04.10
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Names and affiliations of applicants (* indicates experimentalists):

Li, Yanjun.*, SINTEF Materials and Chemistry, Material Physics , Hogskoleringen 5, N-7465 Trondheim, Norway

Mathiesen, Ragnvald H*, Dept. of Physics, NTNU, N-7491 Trondheim, Norway.

Meier Vivienne *, Dept. of Physics, NTNU, N-7491 Trondheim, Norway.

Chen, Yongjun*, Dept. of Materials Science and Engineering, NTNU, N-7491 Trondheim, Norway.

Report:

The mechanical strength in commercial purity Ti (CP-Ti) can be improved greatly by so-called severe plastic deformation (SPD) processing, by which a so-called ultra-fine grained (UFG) material with grain sizes in the 100-500 nm regieme can be achieved. Such a material could be an ideal material for medical implants by being both light and strong and less toxic than some of the alloyed variants used today. To realise this or any similar commercial potential, however, the ductility of the UFG material has to be further improved. A complication to the practical SPD processing of Ti is that it, in contrast to many other metals, has to be carried out at elevated temperatures, e.g. by Equal Channel Angular Pressing (ECAP) at ~ 700 K. Therefore, in order to arrive at the ultimate Ti material it is necessary to understand and be able to control the nucelation mechanisms and kinetics of the thermomechancial softening processes, such as recovery and recrystallisation. For nano-structured metals in general, the high strain together with the considerably high energy stored at grain boundaries give rise to a substantial driving force for nucleation and a dense and highly heterogeneous distribution of nucleation sites. At elevated temperatures these circumstances can combine to yield growth and softening dynamics far away from the conventional near-equilibrium growth and softening models, where early-stage processing could be dominated by competing kinetics and therefore decisive to the final outcome of the thermomechancial treatment. The studies reported here are to our knowledge the first attempts made with XRD to study recovery- and recrystallisation-kinetics in SPD processesed UFG materials in situ at elevated temperatures.

In the study, CP-Ti UFG materials which were prepared by ECAP in 4 different batches, yielding different thermomechanical states, by a combination of deformation strains (1,4,8) and deformation temperatures (350 and 450°C). For each batch, 150 μm thick rectangular samples were prepared by spark-erosion followed by electro polishing, in an attempt to minimize sample preparation impact on the as-ECAPed thermomechanical state of the material. For each of the 4 initial states, in-situ X-ray diffraction was carried out during isothermal annealing at 5-6 different temperatures ranging from 300-525°C. A few samples were measured twice with the same initial state and the same isothermal annealing temperature to verify that reproducible results were obtained and that the data coverage with each sample was adequate to obtain a reliable statistics. The experiments were carried out in transmission geometry with the sample mounted inside a custom built resistance furnace. Furnace and sample temperatures can be controlled and monitored in situ, and allows for the tiny samples to be heated from room temperature to the operating isotherm in less than a second, with isotherm stability ~0.3 K.

It turned out to be a bit more challenging than anticipated to obtain good conditions for XRD experiments in transmission mode at BM 01A with 150 μm thick Ti. In preparing for the experiment we had expressed a need for 22-25 keV to reduce absorption. The beamline optics had not been operated at energies considerably above 17-18 keV for a very long time, and in the end it turned out to be practically impossible to reach the desired X-ray energy. Through a considerable effort of step-by-step optimizing of the optics by the beamline responsible we managed to reach 21.2 keV with a reasonably stable and uniform cross-section incident beam. Another (but foreseen) limitation with the BM01A setup concerns the performance of the detector systems available, i.e. an Oxford Diffraction Onyx-CCD and a MAR 345, both with camera read-outs from tens of seconds to minutes. Consequentially the experiments had to be limited to rather slow softening or growth kinetics happening over several tens of minutes to hours, practically limiting the upper isothermal annealing temperatures. A 1° sample oscillation was carried out during each exposure to secure proper intensity integration for the new ultra-fine recrystallised grains.

All in all the whole study comprised 23 different annealing experiments, each taken with continuous exposures and read outs over a sequence duration from 1 – 6 hours. The long duration sequences were taken at low temperatures where recovery alone is responsible for softening. The as-deformed initial grain structure gave rise to a textured powder diffraction pattern, for which recovery result in relaxation of the Ti-lattice and the reduction of the texture band width which can be followed continuously during annealing. At temperatures of 450 °C and above, a relatively large number of new recrystallized grains and their subsequent grain growth could be monitored for each sample. The growth of recrystallised grains into the neighboring grains composed of a high dislocation densities and sub grains is therefore accompanied by a reduction of the diffraction intensity in the parent material texture components.

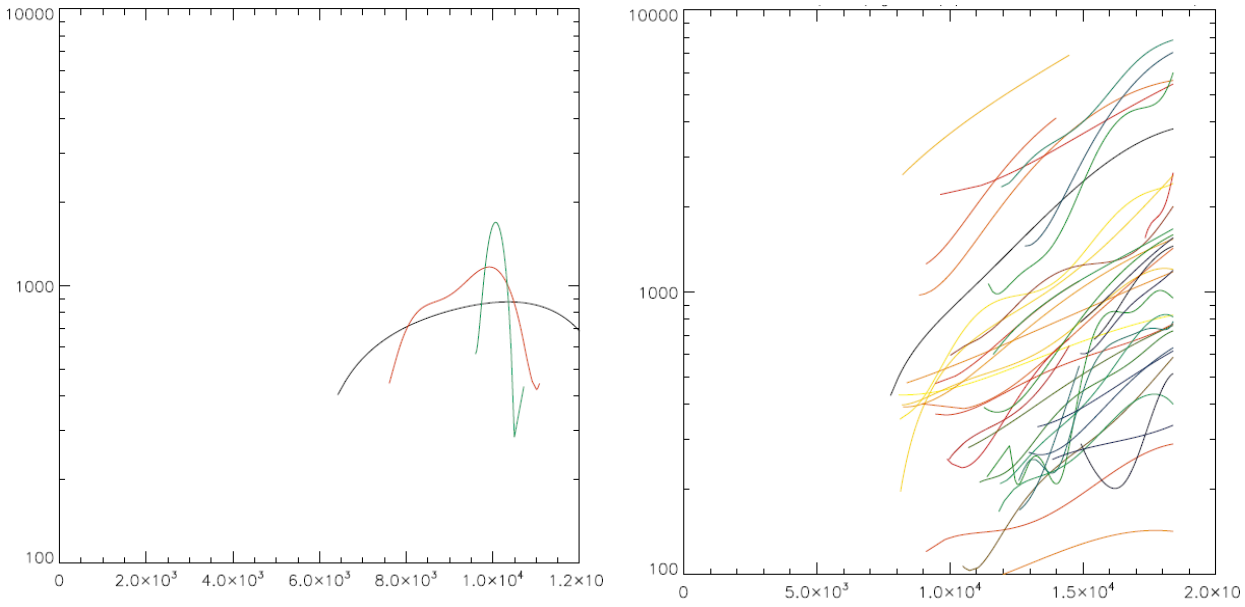
Figure 1 and 2 show growth curves for individual recrystallized grains as a function of annealing time. As can be seen, different newly-recrystallized grains can have totally different shapes of growth curves. This is because the newly nucleated grains have different orientations and surrounding matrix with different deformation energy, therefore different growth rates. The competing growth of neighboring recrystallized grains will also influence the growth curves of individual grains.

As expected, the recrystallization kinetics of the ECAPed CP-Ti was found to depend very strongly on the initial thermomechanical state. Recrsytallistion appeared at the lowest annealing temperature in the sample ECAPed at 350 °C for 6 passes. As shown in Fig.1(a), a large number of recrystallized grains were detected in the ECAP-6P@350°C sample during annealing at 450°C. However, very few recrystallized grains could be detected in the ECAP-4P@ 450°C sample during annealing at 485°C. ECAP-6P@350°C sample also showed a higher density of recrystallised grains than all the other samples ECAPed at 450°C during annealing at temperatures above 450°C . This is because a higher density of dislocations and smaller sub

grains can be formed, and therefore a much higher deformation energy can be stored in the sample during ECAP at 350 °C than at 450°C.
Likewise, for the samples deformed at 450°C, as expected, the driving force for recrystallization increased with increasing deformation strain, and whereas growth of 260 individual grains could be followed in the ECAP-8P sample during annealing at 525°C, only 1 recrystallized grain was detected during 5h of annealing at the same temperature with the ECAP-1P sample. Fig. 2 shows the evolution of the diffraction intensity of recrystallized grains in the ECAP-4P@450°C sample (Fig. 2a and c) and ECAP-8P@450°C sample (Fig. 2 b and d) during annealing at 525°C. As can be seen, the incubation time for recrystallization of grains is shorter while the density of the recrystallized grains is higher in the 8P sample than in the 4P sample. There are also much more recrystallized grains in the 8P sample showing the annihilation behavior during annealing than in the 4P sample (Fig. 2 c and d).

For the 6P@350°C, the 4P and 8P@450°C samples the total number of recrystallised grains monitored during isothermal annealing at 450°C and above exceeds 1300. Obviously, it is relatively straight forward to asses directly from the experimental results the differences in driving force for recrystallisation as a function of the initial thermomechanical state and annealing temperature. It is however also possible to obtain a lot more detailed information on eventual differences in the growth mechanisms that dominate with different initial states and processing parameters by performing a classification of the individual growth curves for each of the samples. The classification scheme is designed in such a way that each category is associated with a particular growth restriction/growth behaviour model. The classification scheme allows an overall comparison between the different sequences both in terms of the frequency at which different growth and annihilation mechanisms appear. It is also convenient to compare results overall and for subcategories by quantitative data such as nucleation times and frequencies, growth rates.

At present the results from MA693 are still being analysed in light of the classification scheme. We are quite convinced that the results obtained contain necessary information to promote new insight into several aspects of recrystallisation and recovery in SPD materials, in particular on early-stage competing growth kinetics, both by quantitative assessment from a sound statistics, and by several intricate details discovered in individual growth curves, such as step-wise growth temporarily arrested at plateaus, etc. We expect the ongoing analysis work to result in at least two journal papers.



(a) **(b)**

Fig. 1 Evlution of the diffraction intensity (vertical axis) of individual recrystallizd grains as a function of annealing time (horrizonal axis) to show the influence of ECAP temperature on the recrystallization behavior of CP-Ti. (a) sample ECAP-4P@450°C, during annealing at 485°C , (b) sample ECAP-6P@350°C , during annealing at 450°C .

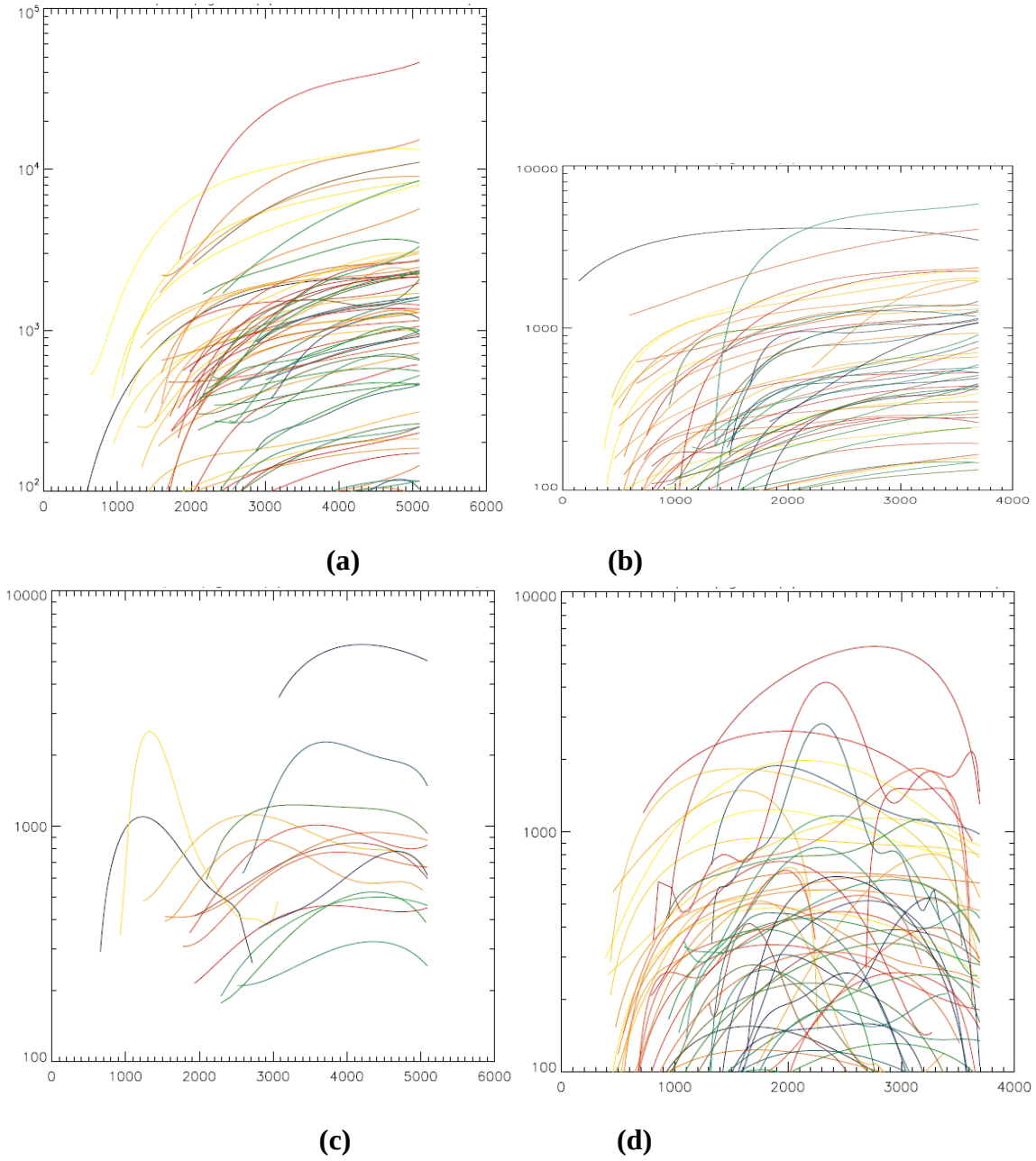


Fig. 2 The growth curves of individual recrystallized grains during during iothermal annealing at 525 °C. (a), (c) sample ECAP-4P@450°C; (b), (d) sample ECAP-8P@450°C