Influence of a pre-deformation on the growth of titanium multicrystals

D Chaubet¹, W Beucia¹, P Franciosi² and B Bacroix²

¹Université Paris 13, LSPM CNRS, 93430 Villetaneuse, France
²LSPM CNRS UPR 3407, Université Paris 13, 93430 Villetaneuse, France

brigitte.bacroix@univ-paris13.fr

Abstract.
In the present study, we have developed a new elaboration process which facilitates the growth of titanium single crystals by combining plastic strain and thermal cycling around the transition temperature. It consists in introducing a small plastic deformation into annealed specimens (like in the critical strain hardening method) before carrying out annealing cycles of only a few days under high vacuum. This method has been applied to titanium of commercial purity (CP-Ti). At the end of each cycle, samples are examined in SEM / EBSD for the characterization of grain sizes and shapes, crystallographic orientations and crystalline quality.

Our first results demonstrate that we can obtain grains bigger than one square centimetre (thickness 2mm) after a preliminary plastic deformation of 3% followed by two 2 pairs of annealing cycles (total time: 96 hours). This enhanced growth is thought to be due to the combination of two driving forces associated (i) to the energy stored within the grains during plastic strain and (ii) to the phase transformation, which acts alone in the transus cycling process. These single crystals are intended to be used to study their mechanical behaviour with the small tensile machines developed to perform in situ mechanical tests into microscopes (SEM, AFM) or diffractometers.

1. Introduction
Titanium alloys are widely used in many industrial sectors because of excellent mechanical properties, combined with a high corrosion resistance and lightness. However, the basic plasticity mechanisms still remain insufficiently characterized because of the large variety of possible mechanisms in hcp structures and a lack of basic studies on single crystals, difficult to produce. Studies reported in a fairly old literature [1] have reported the development of multi-centimetre titanium "multicrystals" by means of very long thermal cycles (i.e. about 8 days) around the hcp – bcc transition temperature, under ultra-vacuum conditions required for a satisfactory crystal quality. This very long duration of such cycling methods was the reason for which we formerly choosed another way to carry on preparation of single crystals of another hcp metal, zirconium, by a strain-annealing method [2]. In these two solid ways for single crystal preparation, there is a first step that brings energy to the metal, and a second one which use this energy to produce limited nucleation or grain growth in order to get very large grain sizes. For the two ways, this second step generally lasts few days, or more. In the present study, we intend to develop a faster procedure, mixing pre-deformation and thermal cycling with short thermal cycles.
2. Experimental material and procedures

The starting material was a 2.0 mm-thick sheet of commercially pure titanium (Ti grade 2 according to the American Society for Testing and Materials standards). The initial microstructure was in an annealed state with an equiaxed grain shape. The EQVivalent Average Diameter (EQAD) of the grains, as calculated from EBSD measurements, was about 47 µm (surface weighted). Samples were cut in a rectangular shape of 14.0 cm x 2.0 cm for the grain growth procedure. For EBSD, the sample preparation consisted first in mechanical grinding with silicon carbide paper down to grade 400, followed by OPS (silica suspension) polishing. Then, a final electrochemical polishing was performed with A3 electrolyte of Struers, 35V for 30s.

The EBSD diagrams were performed within a ZEISS SUPRA40VP FEG- SEM equipped with the automatic Orientation Imaging Microscopy (OIM™) software from Tex-SEM laboratories and the processing of the raw data using the TSL OIM™ Analysis7 software. The orientation maps covered a typical area of 2.00 mm x 2.00 mm scanned with a step size varying from to 2µm to 3 µm. As most samples had β-transformed microstructures, we considered the colonies of α laths as grains. Such a choice led us to the following grain parameters for the analysis software: minimal grain boundaries misorientation of 2°, minimum grain size of 5 pixels, and multi-row shape. Grains of less than 5 pixels size were cleaned-up using the grain dilatation method provided by the software (with a 2° misorientation tolerance angle).

Tensile strain of the first step of thermo-mechanical treatment was performed with a Schenck/MTS testing machine at room temperature, with a strain rate of 10⁻⁴s⁻¹, tensile axis being parallel to the transverse direction (TD) of the as-received state. Gauge test length was 8.0 cm. Strain was measured with an extensometer, in order to obtain a final plastic strain of 3%.

High vacuum annealings were performed in a Nabertherm RHTC 80-710/15 high temperature tube furnace with a ceramic working chamber. Ramping of 5° per minute was used for heating and the beginning of cooling down to 550°C for each step of the procedure. High vacuum was obtain using turbomolecular pumping until a maximum gas pressure of 1.5·10⁻⁵mbar was reached.

3. Experimental results and discussion

We will first present the thermo-mechanical procedure that led to the growth of large titanium crystals, and then the investigations on the influence of pre-deformation, comparing the microstructure evolution during the whole process of no pre-deformed (0%) and pre-deformed (3%) samples.

3.1. Thermomechanical procedure leading to large titanium grains

The five steps thermo-mechanical procedure used is sketched in figure 1. First step was a 3% tensile deformation, followed by two annealing cycles. Each cycle consisted of two annealing steps: the first one of 24 hours in the β phase at 920°C, and the second one of 24 hours in the α phase at 860°C.

![Figure 1](image-url)

Figure 1. Schematic representation of the experimental thermo-mechanical procedure applied to produce large titanium crystals.
With this procedure some very large grains developed, so large that a complete view needed numerous backscattered SEM pictures to be constructed as can be seen in figure 2. For such large grains, IPF maps were plotted from EBSD measurement on both sides of the sample, showing the same uniform colour, which means that the orientation is the same on both sides, and that there is no substructure. We concluded that these grains had the thickness of the sheet and a good crystalline quality. As can be seen in figure 2, large grains are surrounded with much smaller grains, suggesting the occurrence of an Abnormal Grain Growth (AGG) mechanism.

3.2. Comparison of microstructure evolution during the procedure for 0 % and 3 % pre-deformed samples.

As seen in former section, our procedure is less time-consuming than the usual thermal-cycling method used by Jourdan [1]. In order to understand how pre-strain could influence the grain growth, we compared the microstructures at the end of steps 2 to 5 for no pre-deformation and 3% pre-deformation (step 1 of the procedure). At the end of each of these steps, the microstructure was characterized through SEM/EBSD. From OIM analysis, IPF maps and boundaries misorientation distributions were also examined, and the average equivalent grain size (EQAD) was calculated (surface weighted).

3.2.1. First cycle and first 24h β annealing (step 2)

During this step, some growth is expected during heating in the α phase, particularly for the pre-deformed sample where Strain Induced Boundary Migration (SIBM) is likely to occur. Then, samples undergo complete phase transformation at 920°C: it has been checked that in some samples which were just heated to 920°C and immediately cooled down, the α grains have indeed completely lost their initial equiaxed grain shape. During the following 24 hours within the β phase, we assume that the large β grains could grow, but unfortunately, we could not observe them directly.

The microstructures of the 0% and the 3% pre-deformed samples that can be examined at the end of this step are the ones produced by the subsequent β→α transformation during cooling. As can be seen in figure 3, the morphology is typical from transformation, and composed of colonies of laths. At the end of this step, the main difference between the two microstructures is a bigger size of the colonies (considered as “grains”) within the pre-deformed sample, as shown by the EQAD values given under [001] IPF maps.
3.2.2. First cycle and first 24h α annealing (step 3)
In classical cycling procedure this step is the “growth” step, for which the driving force is to be found in the reduction of interfacial boundary energies for all kinds of boundaries produced by the $\beta \rightarrow \alpha$ transformation, namely the inter-lath boundaries or inter-variants boundaries within the former $\beta$ grains. Actually, it can be observed that the EQAD value increases for the 0% pre-deformed sample, while it does not seem to vary for the pre-deformed sample in the areas in which no abnormal grain growth occurs. Indeed, the most noteworthy event of this step is the occurrence of abnormal growth of few grains as can be seen in figures 4 and 5.

**Figure 3**. [001] IPF map of microstructure after R1 (first $\beta$ anneal, step 2) showing bigger colonies in the pre-deformed sample.

**Figure 4**. [001] IPF map of microstructure after R2 (first $\alpha$ anneal, step3) showing abnormal grain growth in the pre-deformed sample.
Figure 5. Reconstructed picture of abnormally grown grains for 3% pre-deformed Ti, at the end of the first cycle (backscattered SEM)

3.2.3. Second thermal cycle (steps 4 and 5)
As in the first step of the second cycle (step 4), the samples went through a new run of $\alpha \rightarrow \beta \rightarrow \alpha$ transformations, it was surprising to observe that no needle or lath could be seen in the abnormally grown grains from the first cycle, as if no phase transformation occurred for them. So, apparently, these “metastable” $\alpha$ grains could take advantage of this step to continue their growth.

The comparison of the two microstructures at the end of the second $\alpha$ annealing is presented in figure 6. It is seen that the normal growth which took place in the 0% pre-deformed sample has led to grains with a diameter of few millimetres. At the same time, as the grains which have abnormally grown during the first cycle in the 3% deformed sample are only subjected to grain growth during the second cycle, their size can exceed 1 cm. No sub-structure could be detected on the maps within the very large grains (figure 6 right). Grains produced by this relatively short thermal process are thus of good crystalline quality. We can also notice that the orientations of different large grains are not all the same.

3.3. Why abnormal grain growth could be facilitated by pre-deformation?
It is likely that hardening due to the low plastic pre-deformation is eliminated during $\alpha$ heating (step 2), by a recrystallization mechanism implying limited nucleation and enhanced SIBM. As a consequence, we expected to see the influence of that prestrain simply on the average grain size after $\beta$ annealing (step 2, figure 3), but not on the initiation of abnormal grain growth during further $\alpha$ annealing (step 3, figure 4).

In fact, it is likely that the SIBM mechanism could have also changed the impurities distribution owing to possible interactions either with fast moving boundaries, or with residual dislocations in the lower energy growing grains. If the impurities distribution in solid solution or as precipitates is still different at the end of the first $\beta$ annealing step, it is then possible that some boundaries have become free of solute drag impurities or pinning particles, thus allowing the abnormal growth of some grains.

In this respect, it is also significant that abnormally grown grains seem to resist to phase transformation, as if no nucleation site was available within the large grains, or as if these grains contained sufficient solute elements capable of stabilizing the $\alpha$ phase.
Otherwise, if we simply assume that bigger $\alpha$ grains produced by SIBM (because of the 3% strain) on heating transform into bigger $\beta$ grains during $\beta$ annealing, we understand why the $\alpha$ colonies that grow on cooling from the reverse phase transformation are larger in size than those observed in the 0% pre-deformed sample, as seen in figure 3.

4. Conclusion

We proposed here a new thermo-mechanical procedure allowing to produce titanium large crystals, of good crystalline quality and of sizes greater than one centimeter. These crystals may be valuable materials for fundamental investigations on single crystals with small testing machines.

The procedure consists of 3% pre-deformation followed by two annealing cycles under high vacuum. Each thermal cycle consists of a first step of 24 hours $\beta$ annealing at 920°C followed by a second step of 24 hours $\alpha$ annealing.

We compared the microstructures obtained within the 0% and 3% predeformed samples at the end of each step of the procedure. A small pre-deformation allows to activate abnormal grain growth of few grains during the second step of the first thermal cycle. Experiments are under way in order to investigate the mechanism involved in such a rapid activation of abnormal growth.

References
[1] C Jourdan, D Rome-Talbot and J Gastaldi 1972 Philosophical Magazine 26:4 1053-55
[2] D Chaubet, J P Fondere and B Bacroix 2001 Materials Science and Engineering A30 245-53