Investigation of SIBM driven recrystallization in alpha Zirconium based on EBSD data and Monte Carlo modeling

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Abstract. The work focuses on the influence of moderate plastic deformation on subsequent partial recrystallization of hexagonal zirconium (Zr702). In the considered case, strain induced boundary migration (SIBM) is assumed to be the dominating recrystallization mechanism. This hypothesis is analyzed and tested in detail using experimental EBSD-OIM data and Monte Carlo computer simulations. An EBSD investigation is performed on zirconium samples, which were channel-die compressed in two perpendicular directions: normal direction (ND) and transverse direction (TD) of the initial material sheet. The maximal applied strain was below 17%. Then, samples were briefly annealed in order to achieve a partly recrystallized state. Obtained EBSD data were analyzed in terms of texture evolution associated with a microstructural characterization, including: kernel average misorientation (KAM), grain orientation spread (GOS), twinning, grain size distributions, description of grain boundary regions. In parallel, Monte Carlo Potts model combined with experimental microstructures was employed in order to verify two main recrystallization scenarios: SIBM driven growth from deformed sub-grains and classical growth of recrystallization nuclei. It is concluded that simulation results provided by the SIBM model are in a good agreement with experimental data in terms of texture as well as microstructural evolution.

1. Introduction

Hexagonal zirconium and its alloys bring scientific attention, mainly due to their importance in the nuclear industry. In consequence, they have been investigated in detail in order to understand and control microstructural evolution induced by recrystallization and grain growth phenomena.

In particular, it has been shown by Zhu et al. [1] that the microstructure of Zr-2Hf alloy strained up to 55% by plane strain compression exhibits high heterogeneity, which is characterized by the coexistence of highly deformed and less deformed zones. During annealing the classical nucleation occurs in the highly deformed areas, especially at grain boundaries and/or nearby twin intersections, then further growth of new recrystallization grains is observed. On the other hand, recovery and related continuous recrystallization occur in the less deformed areas. In this case, in-situ TEM observations revealed that the sub-grain structure can be formed due to annihilation and rearrangements of dislocations and further transformed through sub-grain coalescence. This, in turn, can facilitate formation and growth of recrystallization grains.

The presented description of possible recrystallization mechanisms has been refined further based on examination of rolled Zr-2Hf alloy (50% and 90% of strain) [2]. Taking into account an evaluation...
of stored energy and detailed analysis of texture evolution during annealing, it has been noted that strain induced boundary migration (SIBM) can play an important role, as the process responsible for growth of sub-grains formed inside grains during recovery. In the cited paper SIBM has been also used to explain competition between two main texture components: R -\{hkil\}\langle11-20\rangle and D -\{hkil\}\langle10-10\rangle observed over the grain growth stage.

A similar observation has been reported for Zr702\(\alpha\) rolled by 80% of thickness reduction and further annealed, where SIBM facilitates consumption of large and slowly recrystallizing grains [3].

The main aim of this work is to contribute to the current understanding of recrystallization in zirconium. It is noted that all the above mentioned studies dealt with relatively high deformations (50% or higher) and its further influence on annealing behavior, which relies on a complex interplay between different mechanisms, classical nucleation in highly deformed zones and SIBM driven growth of sub-grains, for instance. Hence, it is difficult to separate each of them and analyze in detail.

However, in the opposite case of moderately deformed material (less than 20% of strain), SIBM driven recrystallization is expected to dominate [4], which gives an opportunity to focus on texture and microstructural evolution caused only by this mechanism.

Therefore, the hypothesis of SIBM driven recrystallization is analyzed and tested in the case of appropriately heat-treated zirconium. The investigation is based on EBSD analysis of experiments similar to the one conducted by Francillette et al. [5], which is then supported by computer simulations of a Monte Carlo Potts model.

2. Experimental procedure

Low alloyed zirconium (Zr702\(\alpha\)) was formed by rolling and annealing into a recrystallized sheet, which was afterwards used to prepare parallelepipedic samples of dimensions 8.12 mm, 6.30 mm, 6.30 mm for channel-die compression, with the longest dimension parallel to the initial rolling direction (RD) of the sheet. Two compression directions were chosen, the transverse direction (TD) and the normal direction (ND). In both cases, elongation was restricted only to RD and the surface subjected to the load had the same dimensions. Using an Instron 1195 testing machine, the final achieved strain was 17%. It was observed that compression along ND required a slightly higher force than compression along TD. Because of that, the samples deformed in ND are referred to as hard (H), whereas samples deformed in TD are called soft (S). Then, all the samples were briefly annealed (615 \({\degree}\)C, 15 minutes) in an air atmosphere to reach a partly-recrystallized state.

It has to be mentioned that this experiment is quite close to the one performed by Francillette et al. [5], which revealed a strong influence of the initial texture on the activated slip/twin systems, resulting hardening curve and level of macroscopic stress. They noticed that the soft samples correspond mainly to the activation of easy prismatic glide systems, which is related with a linear character of the hardening curve. Conversely, the hard samples are associated with the activation of prismatic glide and secondary hard systems, thus the hardening curve has a parabolic shape in this case.

Both, deformed and annealed states, were examined by the EBSD technique using a Cambridge S360 (WGUN) scanning electron microscope and Carl Zeiss Supra VP - Ultra High Resolution FEG-SEM, equipped with TSL/EDAX Data Acquisition software. The surface preparation procedure, required by EBSD, consisted of grinding with silicon carbide paper down to grade 4000, then grinding with an OPS solution from Struers, and finally electropolishing under 40 V with an A3 solution from Struers. EBSD topological maps were measured on the RD-TD plane for H samples and the RD-ND plane for S samples. The total area covered by EBSD maps was at least 1 mm\(^2\) for each investigated sample. All the obtained EBSD data were analyzed with the OIM Analysis v5.3 software from TSL.

3. EBSD investigation

EBSD grains (deformed and recrystallized) were determined using standard algorithm provided by the software that we used as structures comprising of at least 10 EBSD points (taking into account an EBSD lattice step equal to 1.2 \(\mu\)m), which satisfy the condition that a misorientation angle between the nearest-neighboring points is lower than 15\(\degree\). Then grain size was calculated using diameter and area
methods. Textures were analyzed using Euler angles ($\varphi_1$, $\Phi$, $\varphi_2$) and an Orientation Distribution Function (ODF) cross-section for $\varphi_1 = 0^\circ$. The crystal coordinate system was defined as: $X = [2\text{-}1\text{-}1\text{0}]$, $Y = [0\text{1}\text{-}1\text{0}]$, $Z = [0\text{0}0\text{1}]$. Calculations were conducted using harmonic method.

3.1. Deformed state.
The initial microstructure consists of completely recrystallized and equiaxed (in 3D) grains with an average area grain size of 240 $\mu$m$^2$ (or equivalent diameter of 16 $\mu$m).

Most of the grains have a c-axis located in the vicinity of ND. Further texture analysis reveals domination of a tilted \{0001\}<2\text{-}1\text{-}1\text{0}> component, which appears in the cross-section of the ODF as strong symmetrical maxima ($0^\circ$, $30^\circ$, $0^\circ$) and ($0^\circ$, $30^\circ$, $60^\circ$).

Figure 1 presents example EBSD maps of the deformed state of the H and S samples strained by 17% and 9%, respectively. First one is used to explain the role played by orientation spread, whereas the second shows activity of twins.

Orientation spread appears within the grains (visualized by the color gradient), because of the local misorientations, which are associated with the formation of intragranular dislocations during deformation. In consequence, opposite parts of the deformed grain can vary in terms of orientation.

![EBSD maps](image)

**Figure 1.** EBSD maps (cropped area) of the deformed state of the H sample strained by 17% (a) and S sample strained by 9% (b). High angle grain boundaries (HAGBs) are represented by black lines, while white lines fulfill an axis/angle condition for tensile twins boundaries.

At the same time, the overall shape of the deformed grains, being discussed, is not changed significantly in comparison to the initial equiaxed microstructure. This can be explained by the fact that grains undergo accumulation of local misorientations instead of heavy fragmentation. Concerning the latter, the presented maps show that twinning, which is usually responsible for structural refinement in hexagonal metals, is not very active in the investigated samples. However, some subtle difference between them can be noticed at the beginning of the plastic regime, namely tensile twinning is much more frequent at the early stages (9% of strain) of TD compression – figure 1b, figure 2.

Regarding orientation gradients and local misorientations, both are investigated using grain orientation spread (GOS) and kernel average misorientation (KAM) statistics. The averaged GOS and KAM values reveal the same trend, which is a faster increase in H samples. This is most probably a direct consequence of different operating deformation modes and resulting hardening curves in H and S samples.
The described differences in misorientation accumulation process and slight influence of twinning observed in S samples have together a visible reflection in the final average size of deformed grains - Table 1.

The expected decrease of grain size is only seen at the beginning of plastic deformation (9% of strain) in the S sample, which can be reasonably explained by the above mentioned presence of tensile twins. In the corresponding H sample (7% of strain) the average grain size is not changed at all. Then, further deformation results in an increased average grain size, especially in the H sample, which means that large grains have to be formed during compression. This surprising effect is a consequence of a grain coalescence process driven by a high density of local misorientations nearby grain boundaries and orientation spread within the grains.

Figure 3 presents example of an extremely big grain (purple color) determined by a grain reconstruction algorithm, which in fact is an area comprising of “partial-grains” delimited by low angle grain boundaries (LAGBs). The misorientation profile calculated for the trace marked on figure 3, shows that such large grain have a significant orientation spread, since misorientation from one side to another can reach 35° and despite that it is still treated as a single grain. Such examples were not observed in the initial state.

Therefore, it can be concluded that coalescence of deformed grains is caused by accumulation of local misorientations; they are responsible for transition from HAGB to LAGB, which in turn leads to sticking of the partial-grains. The described overall tendency of merging grains into larger areas is still valid, even if the threshold misorientation used for grain determination is changed to a lower value – 10° for instance. This mechanism is more pronounced in the H sample, thus it has more LAGBs and less HAGBs in comparison to the S sample. Moreover, it the latter case more HAGBs could be generated due to twinning at the beginning of the compression process.

### Table 1. Average grain size for Zr samples.

| Sample | Strain [%] | Area [μm²] | Diameter [μm] (Average/Std.Deviation) |
|--------|-----------|------------|----------------------------------------|
| Initial| 0         | 240        | 16.0 ± 8.3                              |
| H      | 7         | 240        | 15.2 ± 8.5                              |
| S      | 9         | 200        | 17.0 ± 10.6                             |
| S      | 17        | 245        | 15.5 ± 8.6                              |

Figure 2. MDF plot for tensile twin misorientation angle (85°) in the case of S sample after 9% strain.

Figure 3. Fragment of deformed microstructure (H sample, 17% of strain) containing an extremely large grain - purple color (left) - and corresponding IQ map presenting LAGB structure – red lines (right).
3.2. Annealed state
Recrystallization is not complete in both of the investigated samples, since deformation substructures still appear in some of the grains. Therefore, the recrystallized fraction has been extracted based on the GOS analysis.

It is noted that recrystallization kinetics depends on the direction of the applied compression – the recrystallized area fraction is higher in the S sample: 0.7 in comparison to 0.58 in the H sample. However, in both samples recrystallized grains have a similar size – 160 µm² (12.7 µm). It means that in the S sample a higher number of recrystallized grains emerged from the deformed microstructure.

![EBSD maps](image1)

**Figure 4.** EBSD maps (800 µm x 800 µm) of the recrystallized grains in annealed: H sample (a) and S sample (b).

An example EBSD map recrystallized partition extracted from the annealed state of H sample strained by 17% is shown in figure 4. It can be underlined that strain-free grains tend to gather in cluster-like structures instead of being randomly distributed in terms of spatial location.

3.3. Texture evolution
The performed texture analysis is shown only for H samples (figure 5), as the main motivation in this context is to compare results obtained with those reported in the literature for rolled zirconium, since both cases have similar strain geometry (compression direction parallel to ND).

Channel die compression results in the formation and strengthening of deformation \{hkl\}<10-10> component, which is mainly located along \( \phi_2 = 30^\circ \). Hence, the ODF cross-section contains a very broad maximum, which can be described as superposition of four main orientations: (0°, 30°, 30°), (0°, 45°, 30°) and two symmetrically located (0°, 30°,15°) - (0°, 30°, 45°).

After annealing textures have been separately calculated for recrystallized and still deformed partitions. Based on this comparison, it can be noted that most of the grains with an orientation close to the main deformation components, described above, have been consumed and only those with an orientation (0°, 30°, 45°) retained in the annealed microstructure.

Texture evolution in recrystallized partition is very interesting, as the recrystallization component (0°, 30°, 0°), i.e. tilted\{0001\}<2-1-10> is formed, despite the fact that recrystallization is not complete, while it is often underlined that such a component should appear as a result of an additional grain growth process [2, 3]. Therefore, other mechanisms responsible for such a development of recrystallization texture should be expected in the considered case.
Additional insight into the presented texture evolution can be provided by analysis of grain boundary regions extracted from deformed microstructures. For that reason, EBSD points are specially selected based on misorientation criteria, as depicted in [6, 7]. Obtained data contain information about the orientation distribution of grain boundary regions – figure 6. Textures of Intragranular Boundary (IB) and LAGB regions, are of particular interest in this context, since these boundaries can be related with additional energy stored inside grains. These grains, in turn, should be replaced by a recrystallization front due to increased gradient of the stored energy.

The texture of the HAGB area follows that of the whole sample. More interesting observations concern regions adjacent to LAGBs and IBs, since they have a main maximum located at (0°, 45°, 30°) and two slightly less pronounced centered at (0°, 30°, 30°) and (0°, 30°, 15°). Please note that the deformation component (0°, 30°, 45°) in not present in this case.

After annealing it is clearly demonstrated that grains having a high fraction of IBs and LAGBs are disappearing first. Based on that, it can be understood why the orientation (0°, 30°, 45°) has remained in the texture of still deformed grains – figure 5c.

4. SIBM model and Monte Carlo simulations
Taking EBSD observations into account, a following scenario is proposed in order to explain the observed texture and microstructural evolution. First, SIBM is considered as the main mechanism of growth of an almost strain-free sub-grain separated from a deformed grain by HAGB. Then, it is assumed that grains having a substantial fraction of local misorientations (IBs and LAGBs) and the related dislocation density exhibit a higher stored energy and thus are consumed first as a consequence of the SIBM process activated on the HAGB interface adjacent to the above mentioned sub-grain. From this point of view, the higher fraction of HAGBs surrounded by local misorientations in the deformed sample, the higher driving force for sub-grain growth and therefore the higher number of
recrystallized grains after annealing. This corresponds very well with the difference in recrystallized area fraction between annealed H and S samples. Moreover, this model can be used to explain texture evolution, if the main orientation of strain-free sub-grains is related with tilted \( \{0001\}<2-1-10> \) component.

In the next step, Monte Carlo Potts model [8, 9] has been employed in order to verify the suggested hypothesis. Simulations have been performed using EBSD microstructures of the deformed state (666×665 sites in the lattice) of H and S samples. However, results obtained are presented only for the first case. The stored energy (SE) distribution, which is a key parameter in simulations of recrystallization has been evaluated based on rescaled KAM parameter – SE \( \in [0, 4.5] \). Regarding boundary properties, well-known Read-Shockley and Huang-Humphreys formulas [10] have been applied in order to calculate the energy and mobility of IBs and LAGBs.

Several scenarios have been simulated, including the SIBM model and classical nucleation. In the latter it was assumed that spatial position of a nucleus is inside the HAGB structure, while its orientation was taken from the deformed sites located nearby or chosen randomly from the deformed texture. Simulations were stopped when a desired fraction of recrystallized area - 0.6 - was reached.

The recrystallized partition obtained from simulation with the SIBM model (figure 7a) consists of 2300 grains of an average size equal to 12.5 \( \mu \)m. These grains are not homogeneously distributed in terms of spatial location.

![SIBM model and classical nucleation](image)

**Figure 7.** Recrystallized grains obtained from simulation performed in the case of the H sample using: the SIBM model (a) or classical nucleation (b).

Also, a strong recrystallization component is developed and only orientation \( (0^\circ, 30^\circ, 45^\circ) \) remains in the texture of the deformed partition – figure 8.

![Recrystallized and Deformed Partition](image)

**Figure 8.** Texture (ODf cross-section for \( \phi_1 = 0^\circ \)) obtained from simulation of recrystallization in H sample. Maximum value of ODF function (red color) is equal to 8.5.
All these observations are in very good agreement with experimental results presented for the annealed state of the H sample.

In contrary, simulation of a classical scenario does not match the experimental data: recrystallized grains are more homogeneously distributed in the microstructure (figure 7b) and texture evolution is wrongly predicted.

Finally, it should be emphasized that SIBM driven simulations also gave correct predictions in the case of the annealed S sample.

5. Conclusions

Hexagonal zirconium was channel-die compressed to reach 17% of final strain and briefly annealed to achieve a partially recrystallized state. Compression in ND (hard samples, H) requires a higher force than in TD (soft samples, S).

Deformation in the hard samples is strongly connected with the accumulation of dislocation cells inside the grains, which has a reflection in local misorientations (higher values of KAM, GOS value) as well as faster development of IBs and LAGBs. This, in turn, leads to grain coalescence – large areas are created from closely oriented grains due to transition from HAGB to LAGB.

In the S samples, in turn, grain fragmentation is easier to achieve at the beginning of the deformation due to activity of tensile twins, thus the fraction of HAGBs increases much more, while the development of the LAGB population is less significant. This difference in the HAGB population between samples was linked with recrystallization kinetics, which is faster in the S sample.

Transition from the tilted \(\{0001\}<10\text{-}10>\) to the tilted \(\{0001\}<2\text{-}1\text{-}10>\) component is seen in the texture of the H sample after partial recrystallization, which indicates that in this case the mechanism of formation of recrystallization texture is different from the one already reported in the literature for annealing of much higher deformations. The evolution described can be associated with SIBM driven recrystallization. Moreover, analysis of textures of grain boundary regions revealed the important role played by local misorientations located nearby HAGBs. Clearly, orientations that are present simultaneously in the texture of HAGBs, IBs, and LAGBs, disappear first during annealing. Such behavior also suggests a domination of the SIBM mechanism.

A physical model was formulated. It assumes sub-grain growth, which is caused by a SIBM process of HAGB in regions containing a high density of local misorientations. The proposed hypothesis was positively tested using a Monte Carlo modeling approach. The simulation results show very good agreement with the experimental data in terms of texture as well as microstructural evolution.

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References
[1] Zhu K Y, Chaubet D, Bacroix B and Brisset F 2005 Acta Mater. 53 5131–40
[2] Zhu K Y, Bacroix B, Chauveau T, Chaubet D and Castelnau O 2009 Metall. Mater. Trans. A, 40 2423–34
[3] Dewobroto N, Bozzolo N, Barberis P and Wagner F 2006 Int. J. Mat. Res. 97 826–33
[4] Humphreys F and Hatherly M 2004 Recrystallization and Related Annealing Phenomena (New York, Pergamon)
[5] Franchillette H, Bacroix B, Gasperini M and Bechade J L 1998 Acta Mater. 46 4131–42
[6] Jedrychowski M, Tarasiuk J, Bacroix B and Wronska S 2013 J. Appl. Cryst. 46 483–92
[7] Jedrychowski M, Tarasiuk J, Bacroix B and Wronska S 2013 Mater. Sci. Forum 753 93–6
[8] Rollett A and Raabe D 2001 Comput. Mater. Sci. 21 69–78
[9] Miodownik M 2002 J. Light Met. 2 125–35
[10] Humphreys F J 1997 Acta Mater. 45 4231–4232