Advanced characterization of microstructural changes during recrystallization in aluminum alloy 6013

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Abstract. Aluminum alloy 6013 was chosen as an example of a material that, after thermal treatment, possesses a relatively uniform and stable bimodal distribution of fine (<<1 μm) and coarse (>1 μm) particles. Samples of this alloy were subjected to plastic deformation by cold rolling. The presence of large and small particles has an influence on the behavior of this material during the recrystallization process. A complex investigation of the microstructural changes during annealing were carried out by means of advanced SEM and TEM techniques. Orientation mapping (OM), i.e., automatic determination of the topography of the crystallographic orientations, was performed using electron backscatter diffraction (EBSD) in the SEM and microdiffraction in the TEM experiments. These techniques were combined with in-situ heating experiments in the TEM and SEM experiments. The quantitative description of the microstructure at each step of recrystallization is presented. Changes in the microstructure of the investigated material during annealing reveal the role and impact of both types of particles on recrystallization and grain growth. The obtained results are in agreement with parallel calorimetric studies.

1. Introduction

Advanced microstructural characterization of changes in a material under rising temperature is still a challenge and many in-situ devices have been developed for annealing a material using TEM or SEM. However, in-situ experiments in electron microscopy were considered difficult and unreliable, mainly because of different experimental conditions, sample preparations and observations which all can influence the behaviour of materials. Existence of one (in SEM) or two (in TEM) free surfaces can introduce differences in processes occurring in microscopic samples with respect to bulk material. [1, 2]. Fortunately, by taking into account many factors influencing in situ measurements, proper conditions can be assured, making them source of scientific observation [3]. Moreover, in order to avoid artificial results and confirm observations of in situ annealing, supplementary calorimetric studies should be performed.

New benefits of in situ experiments come from development of orientation microscopy, both in TEM and SEM. To obtain the most comprehensive data, not only a heating stage but also the technique of acquisition of crystallographic data should be used.

With these techniques, one can compare the changes in texture or orientation between the grains at each annealing step.

In this paper, some examples of combined in situ heating and orientation mapping study applied to aluminum alloy 6013 is presented. These results focus mainly on in-situ SEM and EBSD results. In situ TEM and calorimetry results have been presented previously [4-6].

2. Material

Aluminum alloys 6xxx series are especially interesting because of their very good properties

[7]. Aluminum alloy 6013 [table 1] was chosen because of the characteristic bimodal distribution of the second phase particles, which plays a particular role during the recrystallization process.
Sample of the as-received alloy was supersaturated (1 h - 530°C) and then aged (5 days - 165°C). As a result, the alloy contains a characteristic structure with stable secondary phase particles, which are both small (<<1 μm) and large (>1 μm). After proper heat treatment, the alloy was deformed up to 90% by cold rolling.

Table 1. Chemical composition of 6013 aluminum alloy (% wt.).

Mg	Si	Cu	Mn	Fe	others	Al
1.15	1.0	1.1	0.3	0.5	0.15	remainder

3. Experimental methods

To determine the changes in the material under annealing, a parallel study by means of scanning electron microscopy (SEM), transmission electron microscopy (TEM) and calorimetry was performed.
Experiments using TEM [4-6] were conducted using a heated sample holder. While the thin foil was annealed inside the holder, changes in the microstructure were observed and orientation maps were recorded using microdiffraction. Microdiffraction was used because of the very small grain size in the vicinity of the large particles, which are called deformation zones (DZ). However, to ensure that the observed changes are at least close to those occurring in bulk samples, it is necessary to maintain proper conditions during the experiment [5].
For the scanning electron microscope measurements, the limited spatial resolution is offset by the larger observable area of the sample, which allows for the investigation of bulk materials [3]. In addition, by using a dedicated, special heating stage for simultaneous heating and EBSD mapping (pretilted or with the ability to tilt to 70 ° depending on the model), it is possible to observe the nucleation and grain growth, or phase transformations [2,8-12].
The investigations in the SEM were performed using two heating stages, the H1002D heating stage module for SEM for EBSD applications (Universite Paris-Sud) and the Gatan Murano 525 (Institute of Metallurgy and Materials Science PAS, Krakow). EBSD analysis was performed with the TSL EDAX OIM system. The results obtained by both heating stages were comparable and repeatable.
The results from the TEM and SEM were confirmed by a calorimetric study [4-6].

4. Results and discussion

The microstructure of the deformed aluminum alloy is composed of grains elongated in the rolling direction (RD). The grain size of the matrix in the normal direction (ND) does not exceed 0.5 μm (fig. 1 and 2 - temperature 23°), and the boundaries between them were characterized as HAGBs (high angle grain boundaries). In the areas near the large secondary phase particles, deformation zones (DZ) were identified. This area is composed of grains of approximately 50-200 nm, which could not be examined using the SEM EBSD system. In fig. 1 and 2, areas of large particles are marked by an Image Quality (IQ) map and are surrounded by points with low confidence index (CI), which means that diffraction images in this area were not indexed or were indexed incorrectly.
Global deformation textures are typical for FCC metals with high stacking fault energies and contain two fibers, α with components S {123}<634> and Cu {112}<111> and β with components S {123}<634> and Cu {112}<111>.
After acquiring the orientation maps in the deformed state, the samples were heated inside the SEM to the temperatures represented by peaks in the calorimetric diagram (fig. 1). This allowed for an observation of changes in the microstructure.

At the temperature of the first calorimetric peak (maps from temperatures 270°, 290° and 310° in fig. 1 and fig. 2), nuclei/new grains in the areas of the DZ (fig. 2) are found, and only small changes in the matrix areas can be observed as the arrangements of the low angle grain boundaries (LAGB) with no migration of the HAGBs. In addition, the phenomenon of particle stimulated nucleation PSN [2] was identified.

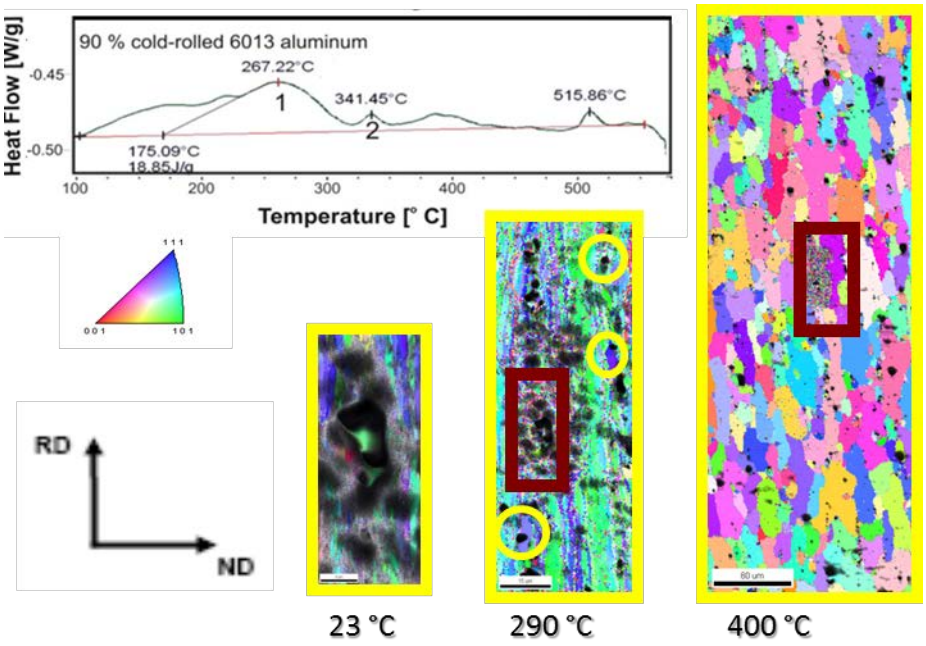

Fig. 1. Heat flow representing the release of stored energy from the 6013 aluminum alloy after 90% cold-rolling with two peaks marked, and EBSD maps with IQ+IPF from the TSL OIM 6.2 system presented before (23°C) and after annealing (290 and 400°C) at the temperatures indicated for each peak. The area of the PSN is marked near the deformation zones (DZ) (yellow circles).

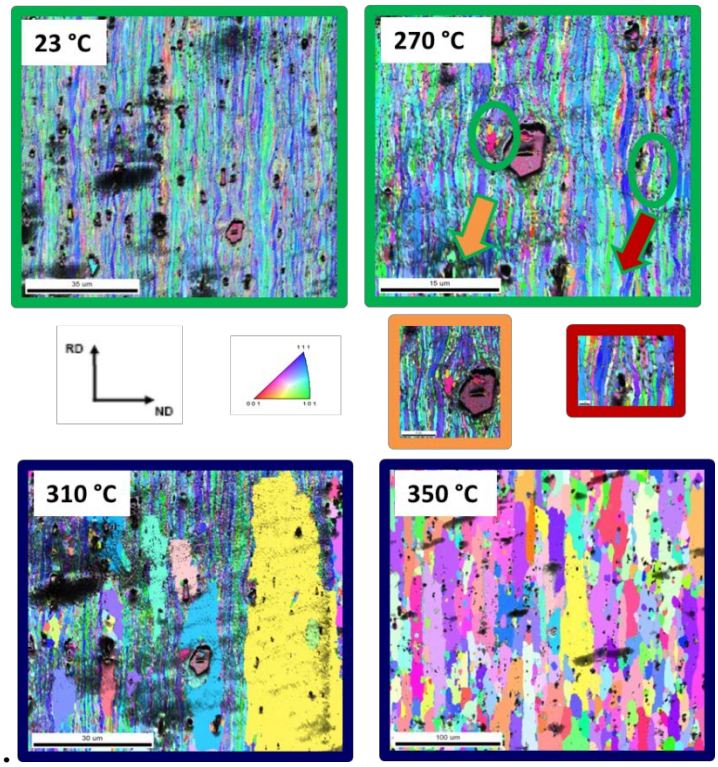

Fig. 2. EBSD maps (IQ+IPF) at each step of the recrystallization with the DZs marked.

At the temperature of the second peak, the migration of the HAGB in the matrix and further growth of the nuclei are observed, but the growth is limited in the direction parallel to the ND.
The relationship of the orientation between grains in the deformed state and between the new grains that appeared at the same location after annealing is characterized by a random distribution of misorientations that suggests that there was no favorable grain growth inside the DZs [5].
To confirm the results from the in-situ studies, parallel calorimetric research was also conducted. Samples were heated in a calorimeter to the temperature of each peak, and orientation mapping was performed [5]. The results obtained were identical to those determined from the in situ studies.

5. Conclusions

Complex investigations of the microstructural changes during annealing of the material after deformation were carried out by means of advanced scanning electron microscope (SEM) techniques such as EBSD and in situ annealing. The experiments were performed in parallel with a calorimetric study. The applied methods, such as combined in situ studies and orientation mapping in the SEM, are powerful tools in providing complex information about the material behavior during annealing.
These investigations confirm previous results from TEM analysis, which found that the recrystallization process in aluminum alloy 6013 is composed of several overlapping processes. The influence of the secondary phase particles is significant. The large particles act as sites for particle stimulated nucleation (PSN).

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References

[1] Mullins W W 1958 Acta Metall. **6** pp.414-427
[2] Humphreys F J and Hatherly M 2002 Recrystallization and Related Annealing Phenomena (Oxford: Pergamon Press)
[3] Sztwiertnia K, Haessner F 1994 Mater. Sci. Forum **157-162** pp.1069-1075
[4] Sztwiertnia K, Bieda M and Korneva A 2012 Application of rientation Mapping in TEM and SEM for Study of Microstructural Evolution during Annealing (Recrystallization) ed K Sztwiertnia (Croatia: InTech) chapter 3 pp 43-58
[5] Bieda M, Sztwiertnia K, Korneva A, Czeppe T, Orlicki R 2010 Solid State Phenom. **16** pp 13-18
[6] Sztwiertnia K, Bieda M, Korneva A 2013 Mater. Sci. Forum **753** pp 221 -224.
[7] Hirsch J, Al-Samman T 2013 Acta Mater. **61** pp. 818–843
[8] Humphreys F J, Ferry M 1996 Mater. Sci. Forum **529** pp 217–222
[9] Hurley P J, Humphreys F J 2004 J. Microsc. **213** 225
[10] Lens A, Maurice C, Driver J H 2005 Mater. Sci. Eng. A **403** 144
[11] Wright S and Nowell M 2009 Review of In Situ EBSD Studies (Electron Backscatter Diffraction in Materials Science) eds A J Schwartz et al. (Springer) chapter 24 pp 329-337
[12] Brisset F, Helbert A-L, Baudin T 2013 Microsc. and Microanal. **19** pp 969-977