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In-situ Observations of Recrystallisation in AA3103 using FEG-SEM and EBSD

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Abstract

Four different samples of AA3103 have been prepared with different secondary phase particles and Mn solute level and hot deformed in plane strain compression to simulate hot rolling. In-situ observations of recrystallisation have been followed in a Field Emission Gun Scanning Electron Microscope equipped with hot stage. Details of single recrystallising grains and grain boundaries have been isolated and analysed. Each grain was found to have its own incubation time for recrystallisation, velocity and boundary mobility. Evidence of particles and solutes influencing the grain boundary movement is analysed and commented upon.

1. Introduction

It has been well documented in past and recent years [1-3] that the recrystallisation behaviour and final grain size of alloys are strongly influenced by secondary phase particles present either in the deformed state or precipitating during annealing. Coarse precipitates accelerate the recrystallisation process by enhancing nucleation rate, while small and finely dispersed particles retard recrystallisation by pinning the grain boundaries (GB).

Many models have also been proposed to describe the recrystallisation of an alloy based on assumptions and experimental data that use average values of GB velocity, mobility and kinetics. The result of these models is that after nucleation, the grains are assumed to recrystallise as a sphere growing at constant speed [4].

In-situ observations of recrystallisation have proved these assumptions to be inaccurate. The recrystallising grains all seem to follow their own nucleation kinetics, GB velocity, and mobility. The secondary phases in the alloy pin the recrystallising GBs giving rise to a jerky movement of the boundary.

Several experimental techniques [5,6] have lately been used to get more detailed information on single grains, but still many questions remain without an answer.

Presented here are the first observations of in-situ annealing of AA3103 done in an electron microscope. The details of the behaviour of single grains are described and commented.

2. Experimental

The Aluminium alloy examined is a AA3103 with composition of roughly 1wt% Mn, 0.5wt% Fe, 0.06wt% Si and Al to balance. The samples were taken from an industrial processing after casting and after breakdown rolling. Different heat treatments (table 1) on the material resulted in four different initial populations of secondary phase particles and Mn solute contents (measured with Eddy current).

All the samples contain precipitates before the annealing procedure. The larger particles (more than 1 μm) are constituent particles of β-Al_6Mn that form during casting and cannot be dissolved during further heat treatments. Samples A, B and D also contain some secondary phase α-$Al_{12}Mn_3Si$ dispersoids and thin plate-like particles. These precipitates are randomly distributed within the grain and the subgrains.

Table 1: Scheme of sample preparation and resulting secondary particle and solute levels.

Name	Initial condition	Heat treatment	Secondary particles	Mn solute level (wt%)
A	As cast	500°C, 500 min + 550°C, 1 week	Dispersoids (big) and plates	0.48
B	As cast	570°C, 300 min + 550°C, 1 week	Dispersoids (small) and plates	0.48
C	Breakdown rolled	630°C, 24 hours	None visible in FEG-SEM	0.68
D	Breakdown rolled	630°C, 24 hours + 550°C, 1 week	Dispersoids	0.47

After heat treatment the material was deformed to a strain of 0.7 with a strain rate of 10 s^{-1} in plane strain compression at 450°C to recreate industrial hot rolling conditions.

The samples were cut to a size of 1cm x 5mm x 1mm and observed in the plane perpendicular to the transverse direction (in the images in this paper the rolling direction is always top to bottom). After mechanical polishing to ¼ μm diamond paste samples B and D were electropolished for 30 seconds at –30°C with a current of 12 volts in a solution of 70% CH_3OH and 30% HNO_3, while A and C were polished with OPU silica suspension.

The samples were observed and annealed in a CamScan X500 Field Emission Gun Scanning Electron Microscope (FEG-SEM) equipped with hot stage. The particular conformation of this microscope with the column inclined 70° from the vertical allows Electron BackScattered Diffraction (EBSD) measurements on a horizontal sample placed in the hot stage [7].

Separate experiments were carried out in a salt bath to find the temperature at which recrystallisation started after about 30 minutes of annealing and lasted for at least 1 hour. For conditions A and B it was found to be 400°C, for condition C 440°C and for D it was 370°C. These temperatures were used as indication for the annealing in the hot chamber of the microscope.

During annealing the recrystallisation velocity was kept relatively low by adjusting the temperature of the sample accordingly. On average the time necessary for the whole sample to recrystallise was about 2 hours. This speed gave sufficient time to be able to focus and record the movement of several grain boundaries in different areas of a sample. EBSD maps were done in detail (step sizes between 0,2 and 0,4 μm) on chosen areas of the samples on the deformed structure, the recrystallised one and also, in some cases,

during annealing. Grain boundaries with misorientation between 5 and 10° were defined as low angle grain boundaries (LAGB), those between 10 and 15° medium (MAGB) and above 15° misorientation were considered high (HAGB). In the present work LAGBs will be outlined in white, MAGBs with a thin black line and HAGBs with a thick black line.

The temperature control and measurement turned out to be rather complex for this configuration but is considered to be accurate within ±1.5°C.

3. Observations

EBSD areas on the deformed samples were selected and mapped for all conditions. Conditions A and C that were mechanically polished had a perfectly flat and polished surface but did not show any orientation contrast (OC) in the backscattered images and the quality of the patterns was poor. Conditions B and D that were finished with electropolishing had very good OC and high quality EBSD patterns, but were is some cases severely etched to the point that particles had fallen out of the sample. Also the surface of these sample was not flat but wavy and full of topography effects.

In spite of the different surface quality and the different solute and secondary particle content of the four conditions the samples all seemed to behave qualitatively in a similar way. The images in this paper are all taken from condition D but the observations reported are to be considered valid for all samples.

The EBSD maps showed that the deformed grains are relatively large (mean size of a 100 µm thick and up to a millimetre long) and elongated along the rolling direction, the subgrains are more equiaxed, as elsewhere reported [8], with a mean diameter of 3.4 µm.

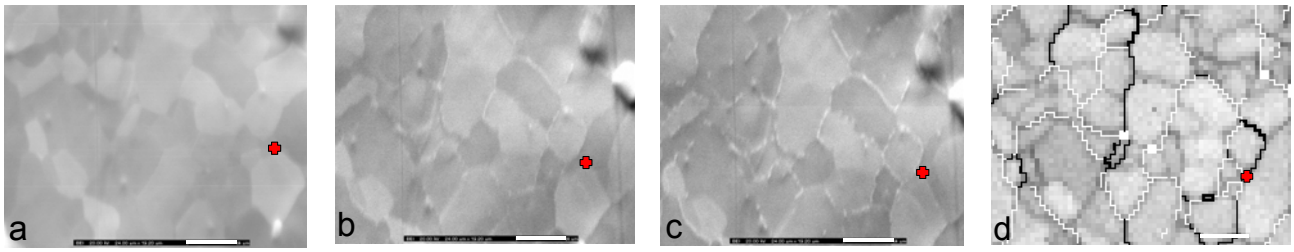

Figure 1: Electron backscattered FEG-SEM images of condition D after different annealing times: *a* 0 min (T=room temperature), *b* 37 min (T=380°C), *c* 86 min (T=397°C), *d* is the EBSD band contrast map. The spots indicate the same point in each image and also in fig 4. White scale bar represents 4 µm.

3.1. Precipitation

During annealing and before recrystallisation fine precipitates start to appear on the subgrain boundaries. In the present system the precipitation tends to start simultaneously on all subgrain boundaries regardless of their misorientation (figure 1) and continues to a great extent until the grain starts recrystallising. The observations may be influenced by a surface effect and a further investigation in this direction will be made in the future.

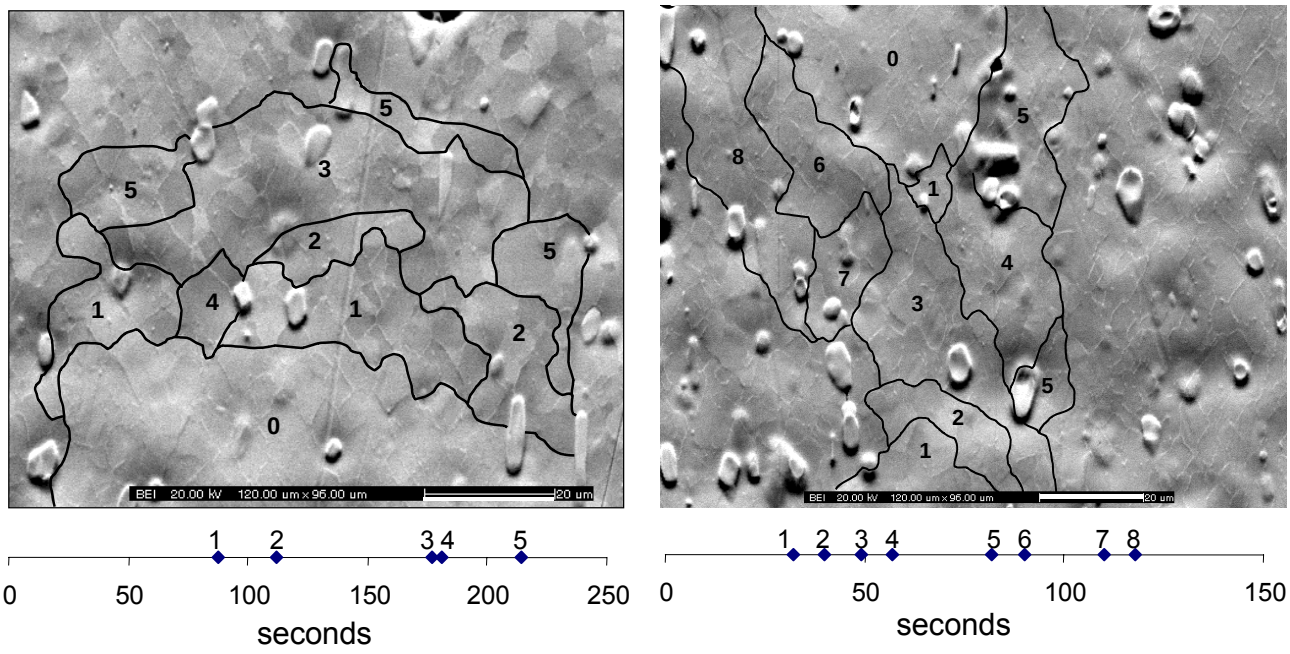

Figure 2: Scheme of recrystallising GB movement with time scale. The boundaries show the limits to which GBs have migrated at given times. After time 1 only the areas labelled '1' are recrystallised. After time 2 only the areas labelled '1' and '2' are recrystallised, etc. On the left the grain recrystallises from bottom to top. On the right the grain starts on the top and is then joined by the same grain appearing from bottom. White scale bar indicates 20 μm.

3.2 Recrystallisation

By observing the samples at low magnification it was found that the recrystallisation process starts uniformly within the samples. The first nuclei of recrystallising grains appeared both along the edges and in the bulk of the specimen at the same time, but not all grains started growing together. While some grains had already reached their final size and shape many areas remained deformed and new grains were appearing. It seems that different grains have different incubation times before they start recrystallising, as also documented in [5]. In some cases very small islands of deformed structure remain completely isolated until they finally join the surrounding recrystallised grain or recrystallise separately usually with an orientation very similar to the surrounding grain. The very last deformed areas recrystallised only after raising the temperature sometimes even 20°C higher than when the recrystallisation had began in other parts of the same sample.

Several grains were observed and followed throughout their recrystallisation process. It was seen that the grains do not grow regularly. The movement is characterised by jerkiness: GBs located along a pre-existing subgrain boundary (or a series of linked subgrain boundaries) jump to a location along another subgrain boundary. The GB velocity is faster than the refresh rate of the SEM image (a few seconds) as the GB sweeps through the interior of subgrains. Then the GB is pinned for varying times at a subgrain boundary before it starts moving again. This effect has also been reported in [5].

Figure 2 shows this jerky GB motion with two specific examples. The example on the left shows a grain recrystallising from the bottom to the top of the image. The movement is not continuous and some areas are recrystallised before others. For example the area that recrystallises in the third part of the sequence leaves a small isolated subgrain (area 4) that recrystallises only later. The example on the right shows that in reality the GB motion must be considered in 3-dimension: a grain that has begun to recrystallise at the top of the image emerges from beneath the surface at the bottom of the image (it is thought to be the

same grain because it has the same orientation), before the intervening material at the surface is recrystallised. Eventually the two isolated surface outcrops join.

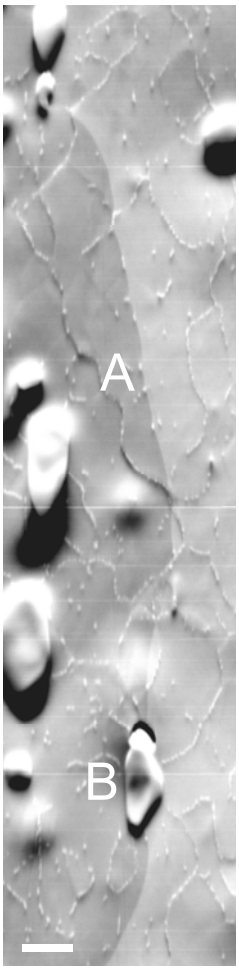

Figure 3: Detail of a recrystallised grain boundary. Area A shows the boundary pinned by dispersoids, area B shows pinning by a constituent particle. Scale bar indicates 4 µm.

Although the recrystallising grain boundaries are stopped and pinned it is not exactly clear how. During these in-situ observations no clear proof of particle pinning was found. In figure 3 a part of the boundary between two recrystallised grains is shown. The evidence on this GB shows both particles pinning the boundaries and areas in which the GB is completely unrelated to the surrounding precipitates. For example next to mark B of figure 3, and also from other observations it is clear that the constituent particles and the larger plate-like particles pin the GB, but the dispersoids do not offer clear proof of their pinning power. In the area marked A in figure 3 there seems to be dispersoid pinning, but just above or below the area the GB is clearly free of any secondary particle

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The grains grow elongated in the rolling direction following the same trend as the deformed grains but eventually becoming much larger (few millimetres in size) and with irregular and jagged shorter edges, typical for particle impeded GB migration [2]. Evidence has been found that the recrystallised grains not only grow preferentially in the rolling direction but actually follow the boundaries of the deformed grains as reported also in [6].

The result is that in most cases the grain boundaries of the recrystallised grains coincide with those of an old deformed grain. An example is given in figure 4. The deformed structure is shown in *a* and the recrystallised in *c*; *b* shows the EBSD band contrast map and grain boundaries (LAGB white line, MAGB thin black line, HAGB thick black line) of the deformed area. The crosses are placed in the same points. The HAGB of the deformed structure visible on the left of fig 4*b* going from top to bottom is in the same position as the recrystallised grain boundary in *c*. This ‘memory effect’ has also been documented in reference [9].

4. Discussion

Annealing in salt bath was carried out on the samples to determine the temperature at which, for each condition, recrystallisation would start after 30 minutes of annealing and last long enough to be able to record the movement of several grain boundaries. The temperatures found were used as an indication for the annealing in the hot stage. Due to the slower heating rates in the hot stage than in the salt bath the samples had more time at high temperature to recover and to precipitate dispersoids on the subgrain boundaries. These effects reflected on the temperature at which recrystallisation began: during the experiment it was 10 to 30°C higher than in the salt bath.

It is interesting to point out that unlike data found in literature [1] the sample containing no secondary particles recrystallised, with roughly the same kinetics, at higher temperature than the samples containing dispersoids and plates. It is not clear yet if the higher solute level of Mn in solid solution for condition C (table 1) gives solute drag effect or maybe

recrystallisation is retarded because of a very fine and pinning efficient precipitation on the substructure. Also condition D with only dispersoids but almost the same Mn solute level as conditions A and B, that contain both dispersoids and plates, recrystallised at lower temperature. This may be an indication of different pinning efficiency between dispersoids and plates.

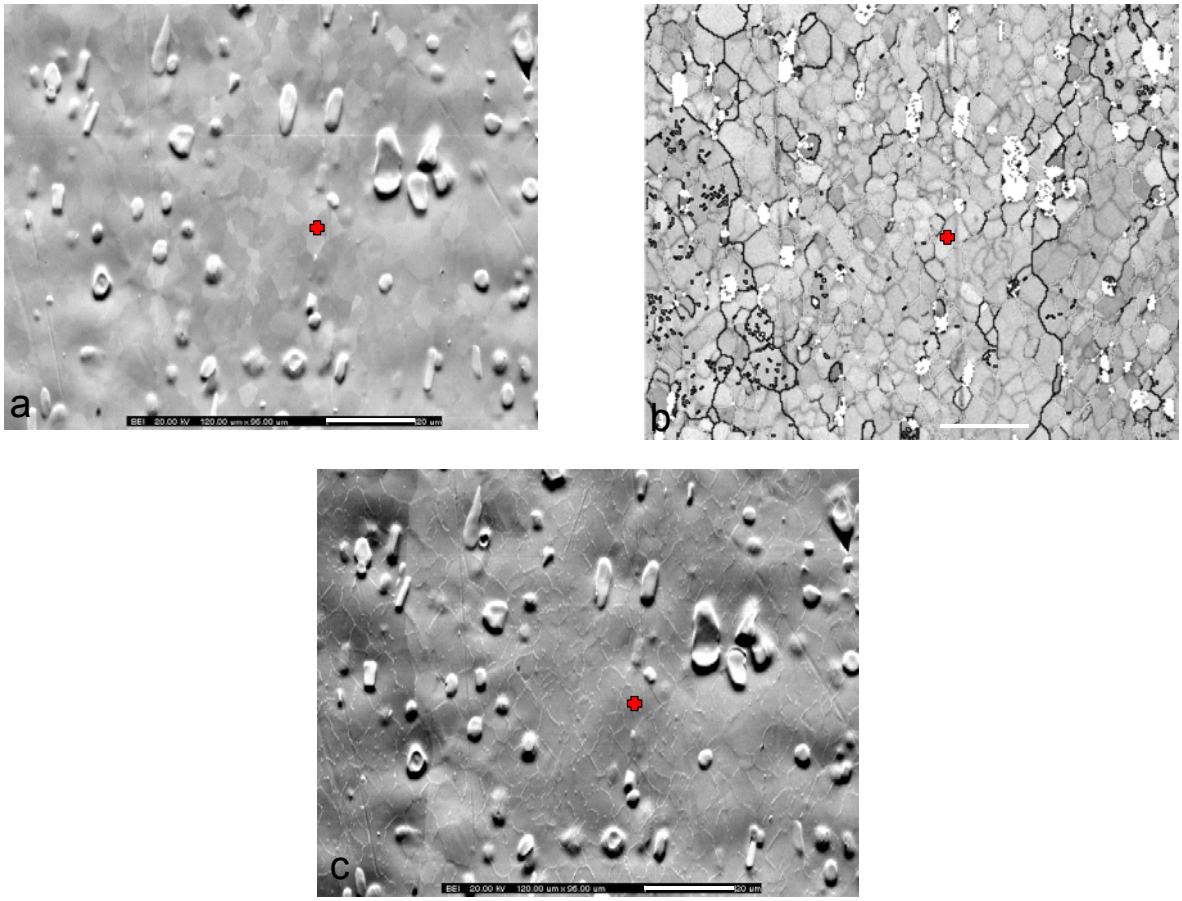

Figure 4: Electron backscatter images of condition D in *a* deformed and *c* recrystallised state. Image *b* is the EBSD band contrast map. The spots indicate the same point in each image and also in figure 1. White scale bar represents 20 μm.

As reported above, during annealing, before recrystallisation, precipitates start forming on the subgrain boundaries. This phenomenon is well documented in literature but it was found, on a similar alloy at higher deformation, that it seemed to be initially localised on HAGBs [3]. In this work the precipitation was found on all GBs at the same extent. The discrepancies may be due to the differences during deformation.

Literature suggests [1-3] that the precipitates that form during annealing before the recrystallisation have a role in pinning the boundaries, but there is proof [5,9] that similar behaviour has been found for grains recrystallising in materials without secondary precipitates. The explanation of the jerky movement of the boundaries cannot then be understood by only considering the pinning forces of the precipitates but also orientation differences between grains and subgrains must be considered.

The observations on the movement of the GBs and their final size seem to confirm that recrystallising grains grow preferentially in certain directions, according to the deformed state of the matrix and the misorientation of the moving GB. Also preferential pinning of certain GBs can contribute to the final recrystallised grain shape.

The mechanism controlling pinning and unpinning of GBs is not clearly understood yet and remains one the most important questions without an answer. Further experiments will be carried out in the future to try and clarify this point. So far also no model has been reported in literature that incorporates GB behaviour as reported in these observations. It may be necessary in the future to consider these behaviours in order to improve the existing models.

5. Conclusions

Detailed observations were made on single recrystallising grains of AA3103 with different secondary phase populations and Mn solute levels. It was seen that:

- The sample with higher Mn solute level and no secondary phase population visible in the FEG-SEM recrystallised at higher temperature. The sample with only dispersoids recrystallised at lower temperature than the sample with also plate-like particles.
- The GB movement is jerky and jumps from one subgrain to the next. The GBs are clearly pinned but the role of the particles on the boundaries is not always clear.
- The recrystallised grains are elongated in the rolling direction and the boundaries tend to coincide with boundaries of the old deformed grains.
- Every recrystallising grain has its own incubation time for nucleation and independent GB velocity and mobility.

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