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Modification of Precipitate Microstructure in 6060 Al Alloy by Equal-Channel Angular Pressing

Gang Sha[1,2], Xiaozhou Liao[3], Rimma Lapovok[4] and Simon P. Ringer[1,2]

[1]ARC Centre of Excellence for Design in Light Metals;

[2]Australian Centre for Microscopy & Microanalysis, The University of Sydney, NSW, 2006, Australia

[3]School of Aerospace, Mechanical & Mechatronic Engineering, The University of Sydney, NSW 2006, Australia

[4]Department of Materials Engineering, Monash University, Clayton, Vic 3800 Australia

An Al-6060 alloy containing elongated rod-like β′ and β precipitates was processed by equal-channel angular pressing (ECAP) for up to 8 passes at room temperature and 180 °C. Transmission electron microscopy and atom probe tomography are employed to characterise the microstructure of precipitates and to measure the compositional evolution of the precipitates and the matrix of the alloy. This investigation demonstrates that ECAP processing significantly fragments elongated β′ and β precipitates. Moreover, ECAP processing at 180 °C drives further decomposition of the alloy and the formation of large, spherical β precipitates. In contrast, ECAP processing at room temperature does not drive further decomposition of the alloy but leads to precipitates coarsening through Ostward ripening.

Keywords: *ECAP, 6060 Al alloy, precipitate microstructure, APT, TEM.*

1. Introduction

Equal-Channel Angular Pressing (ECAP) is a very attractive technique to produce ultrafine-grained materials with superior mechanical properties in a wide range of metals and alloys [1,2]. Application of ECAP to modify the microstructure of precipitates is a relatively new research area and has recently drawn much research attention because of the formation of novel precipitation microstructures and the potential to further improve material properties [3]. Our recent work has demonstrated that ECAP has a complicated and striking effect on the precipitation process, including enabling new precipitate nucleation and growth process in an Al alloy and modifying the precipitate dispersion including the crystallographic orientation, size and morphology [4]. A better understanding of the evolution of precipitate microstructure is essential for establishing the optimum ECAP processing parameters.

It is known that long-term natural ageing and artificial ageing treatments will lead to the precipitation strengthening from β′′ and β′ phases in 6xxx series Al alloys [5,6,7]. These precipitates are often in needle or lathe morphology with well-defined orientation relationships with the matrix and have their long axis always parallel to $<100>_{\alpha Al}$ [5-7]. ECAP processing on the alloys can significantly change precipitation microstructure including their size, morphology, and orientations in the matrix. Three mechanisms have been reported to be responsible for the modification of precipitate microstructure in alloys during ECAP processing. One is the *fragmentation* of pre-existing precipitates due to a high strain applied by ECAP processing [8, 9]. The second mechanism is *accelerated precipitation,* which is observed in Al alloys processed by ECAP at temperatures such > 100 °C [4,10]. The third mechanism is *precipitates dissolution,* which has been reported during ECAP processing at low/ambient temperatures [11]. Controversy remains in the literatures on the precise effect of processing conditions on the dissolution of precipitates. For example, one study suggests that dissolution of precipitates occurs during RT ECAP processing a 6xxx series Al alloy [11], while another proposes that dynamic precipitation occurs in a similar alloy during RT ECAP processing [10]. The objective of the present research is to combine atom probe tomography (APT) and transmission electron microscopy (TEM) to investigate changes in the precipitate microstructure and the corresponding matrix chemistry in samples processed by ECAP, in order to understand the effect of ECAP processing on Al-6060 alloy.

2. Experimental

A commercial 6060 Al alloy (0.47 at% Si, 0.55 at% Mg and Al in balance) was provided as exuded rods of 10 mm in diameter. The as-received materials contains long precipitates (likely β′ and β) of ~100 nm in length and ~ 6 nm in diameter, as shown in Fig. 1. The as-received rods were cut into billets with a length of 35 mm for ECAP processing. The ECAP experiments were conducted at room temperature and 180°C (with isothermal condition), respectively, for up to 8 passes using a hydraulic press of 100 tonnes capacity operating at a ram speed of approximately 2 mm/s. The ECAP processing used a die with two channels intersecting at 90° and route B_C [12]. The duration time of each pass was estimated to be 10-15 seconds. The time to remove the sample from the die was ~ 1 min. The samples were quenched after high-temperature ECAP processing.

TEM characterizations were performed using Philips CM12 operating at 120 kV. Specimens for TEM investigations were cut from bulk specimens using a diamond saw, followed by mechanical wet-grinding to about 50 μm, and then electro-polishing in a solution of 33% nitric acid in methanol at -20°C. Atom probe samples were prepared using a two-step electro-polishing procedure with thin bars having a cross-section of 0.5x0.5 mm^2 [13]. The first step used an electrolyte of 25% perchloric acid in acetic acid at 15 V at room temperature and the second step used an electrolyte of 4% percholric acid in 2-butoxethynal at 20V. The atom probe analysis was performed using a local electrode atom probe at a specimen temperature of 20K under laser pulse energy of 0.5 nJ and with a target evaporation rate of 1%.

3. Results

3.1 Precipitate microstructure of the as-received material

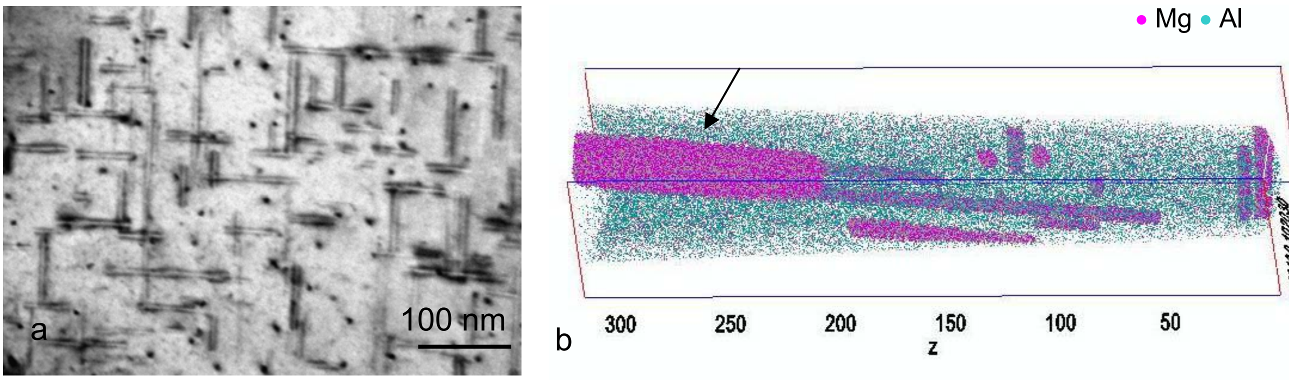

Fig. 1 Precipitate microstructure of AA6060 samples in as-received conditions (a) bright filed TEM micrograph close to a <001>$_{\alpha\text{-Al}}$ zone axis, and (b) a reconstructed atom map showing the distribution of Al and Mg atoms.

The precipitation sequence of Al-Mg-Si alloys during artifitial ageing treatment is generally considered as: GP zone →β″→β′→β [5]. GP zones are known to be spherical solute-enriched features and coherent to the Al matrix. β″ and β′ are needles and rods with their long axis aligned to <001>$_{\alpha\text{-Al}}$. Figure 1a is TEM image from the as-recieved Al-6060 alloy material, and indicates that most of the precipitates possessed lengths > 50 nm, diameters of approximately 6 nm and the usual orientation relationship with the Al matrix. Atom probe analysis indicated that these rod-shaped precipitates were rich in Mg and Si and using a selected cylinder of analysis (diameter 2 nm) their composition was measured to be ~ 20 at % Al with the balance as Mg and Si in a ratio of ~1.9. The Mg/Si ratio is significamtly higher than a ratio of 1.2 of β′′ precipitates [7], but is much closer to 1.8 of β′ precipitate [14, 15], indicating that these rods are β′ phase. Interestingly, a coarse precipitate, marked with an arrow in Fig. 1b, was measured having a low level of Al = 2 at% with a Mg/(Si+Al) ratio of 2. Consequently, the precipitate was considered as an equalibrium β (Mg_2Si) phase.

3.2 Precipitate microstructure of samples processed by room temperature ECAP

It is known that room-temperature ECAP processing introduced high-density dislocations into the microstructure of the material [1,2]. The mean grain size of an Al-6060 alloy sample processed after 8-pass ECAP is 411± 104 nm. A TEM micrograph, as shown in Fig. 2a, confirmed that dislocations and localized strain effect were present in the sample processed by 8-pass ECAP. Unlike β′ precipitates in the as-received sample, no precipitates in a sample processed by 8-pass ECAP at room temperature were visible in the bright field (BF) image in a <001> zone axis of the Al matrix (Fig. 2a). There has been a suggestion that ECAP processing at room temperature causes the dissolution of precipitates and formation of supersaturated solid solution in 6xxx series Al alloy [11]. In fact, the presence of dislocations, residual local strain and the orientation change of precipitates in the Al matrix can make these precipitates invisible in BF images in a low index zone axis such as $<001>_{\alpha\text{-Al}}$. Without detailed chemical information of the microstructure of the alloy processed by ECAP, simple TEM image itself may not be sufficient to answer if the ECAP processing at room temperature has induced the dissolution of precipitates in the alloy.

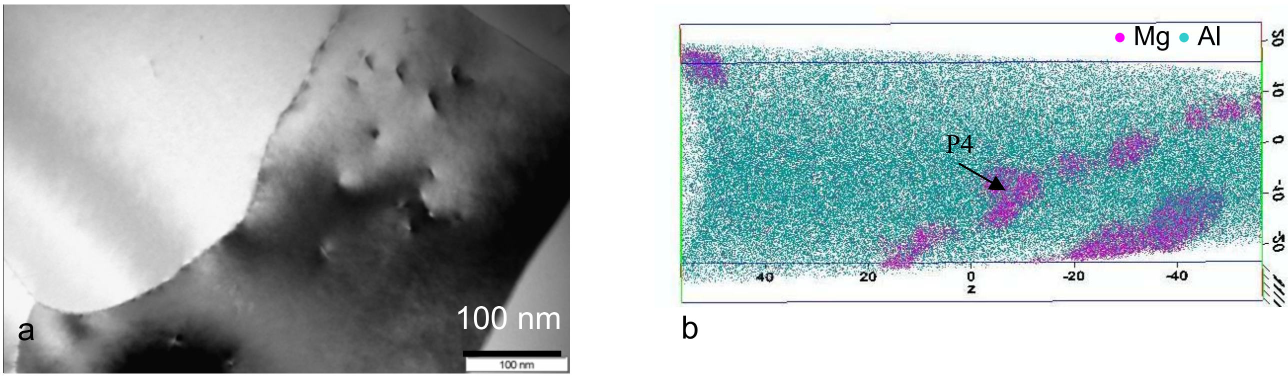

Fig. 2 Precipitate microstructures of Al-6060 alloy samples processed by 8-pass ECAP at room temperature (a) bright filed TEM micrograph along a $<001>_{\alpha\text{-Al}}$ zone axis and (b) a reconstructed three-dimensional atom map showing Al and Mg distributions

Reconstructed atom maps of an Al-6060 alloy sample processed by 8-pass ECAP at room temperature clearly shows the presence of β′ precipitate fragments enriched with Mg , as shown in Fig. 2b, and Si (in a Si map not shown here) in the Al matrix. Some of these small precipitate fragments were approximately 5-10 nm in diameter and distributed linearly. They were likely broken pieces of a single β′ rod caused by the ECAP processing. Small fragments were measured having a different composition from a larger one, marked by P4 in Fig. 2b. The core of the particle P4 was measured having an Al content of 13±2 at%, lower than 24±3 at% of its smaller neighbor fragments. The Mg/Si ratio of the P4 particle is 2.16, higher than 1.84-1.90 of other smaller neighbor fragments. Because the matrix solute concentrations of the samples processed by 8-pass ECAP at room temperature, as shown in Fig. 3, are similar to that of the as-received sample, it is reasonable to believe that the ECAP processing did not cause any significant dissolution of β′ precipitates for formation of a supersaturated solid solution. The high Mg/Si ratio and a low Al content of the larger fragment may suggest that the large fragment was in the process of growth, while its neighbor small fragments were in the processing of dissolution. The overall precipitate microstructural evolution in the alloy was likely in a coarsening regime during ECAP processing. The chemistry change of these precipitate fragments is an essential process for further phase transformation of metastable β′ fragments into the equilibrium β phase in the alloy.

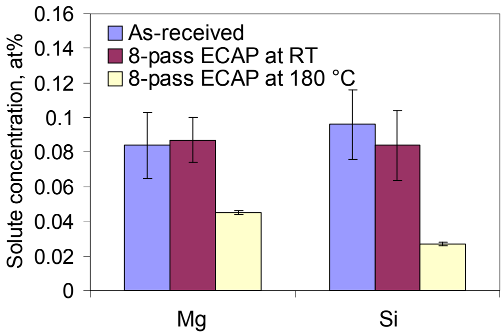

Fig. 3 Solute concentrations of the matrix measured from Al-6060 alloy processed under different conditions

3.3 Precipitate microstructure of samples processed by ECAP at 180 °C

The mean grain size of Al-6060 alloy sample processed after 8-pass ECAP at 180 °C was 571± 128 nm. The sizes of these precipitate are in bimodal distributions. Coarse spherical precipitates of about 50 nm in diameter, as marked by black arrows in Fig. 4a, are loosely distributed in the Al matrix. In addition, smaller spherical precipitates of 5-18 nm in diameter are present in the Al matrix. After processing at 180 °C, dislocation segments pined by precipitates are visible, as pointed by large hollow arrows in Fig. 4a. These dislocations serve as fast diffusion paths of solutes to accelerate precipitate microstructure evolution. Because of a low number density of these precipitates, they were hard to capture using atom probe analysis, and no precipitates were observed in a small analyzed volume shown in Fig. 4b. The Mg and Si concentrations in the matrix of the sample processed after 8-pass ECAP at 180°C, as shown in Fig. 3, are significantly lower than these of as-received samples. This indicated that further decomposition occurred during the multiple-pass ECAP processing at 180°C. All precipitates were measured with very low level of Al concentration of ~ 2 at%, but a high Mg/Si close to 2. This suggested that ECAP processing at 180°C have accelerated precipitation and promoted the formation of equilibrium β. The exceptional low Si concentration in the matrix and high Mg/Si ratio of the β precipitates may suggest that there is precipitation of Si-rich phase in the alloy processed by ECAP. Further TEM characterization will aim to identify these Si-rich phases.

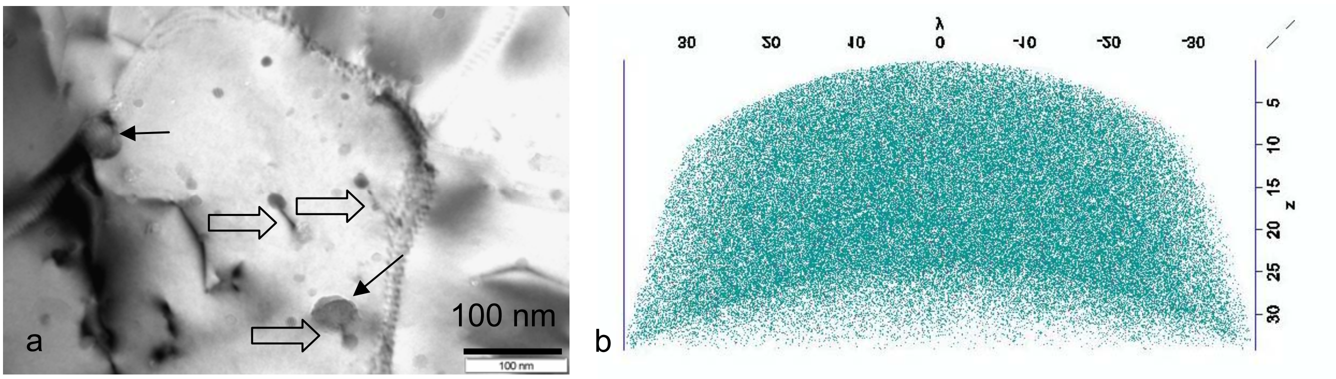

Fig. 4 Precipitate microstructure of Al-6060 alloy samples processed by 8-pass ECAP at room temperature (a) bright filed TEM micrograph along a <001>$_{\alpha\text{-Al}}$ zone axis and (b) a reconstructed atom map showing Al and Mg distributions

4. Conclusions

A combination of TEM and APT was used to investigate the effect of ECAP processing on the evolution of precipitate microstructure in Al-6060 alloy. We summarise that:

1. ECAP processing of AA6060 containing a dispersion of pre-existing β′ precipitates results in extensive fragmentation of these rods.

2. ECAP processing at 180 °C triggered the precipitation of spherical β phase particles, resulting in a significant depletion of the solute in the matrix.

3. ECAP processing at room-temperature after 8 passes caused β′ precipitate fragment coarsening through Ostward ripening i.e. certain precipitate fragments increase their sizes while other small fragments shrink and the solute concentrations of the matrix are unchanged in comparison with that of the as-received material. These larger fragments from individual rod β′ precipitates developed a new chemistry that was richer in Mg (with an Mg/Si ratio of 2.16) after 8-passes of ECAP processing.

Acknowledgments

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References

[1] RZ Valiev, RK Islamgaliev, IV Alexandrov. Prog Mater Sci 2000;45:103
[2] RZ Valiev, TG Langdon. Prog Mater Sci 2006;51:881
[3] Murrayama M, Horita Z, Hono K. Acta Mater 2001;49:21.
[4] G. Sha, Y.B. Wang, X.Z. Liao, Z.C. Duan, S.P. Ringer, T.G. Langdon, Acta Mater. 2009, **57**:3123.
[5] I. Dutta and S. M. Allen, J. Mater. Sci. Lett., 1991,10, 323.
[6] M. Murayama and K. Hono, Acta mater. 1999; 47:pp. 1537-1548.
[7] G A Edwards, K Stiller, G L Dunlop, and M J Couper, Acta mater., 1998, 46: 3893.
[8] M Murayama, Z Horita, K Hono. Acta Mater 2001;49:21.
[9] C Xu, M Furukawa, Z Horita, TG Langdon. Acta Mater 2005;53:749.
[10] HJ Roven, M Liu, JC Werenskiold, Mater. Sci Eng.A. 2008;483-484:54.
[11] I Gutierrez-Urrutia, MA Munoz-Morris, D.G Morris, Mater. Sci. Eng A. 2005; 394:399.
[12] K Oh-Ishi, Z Horita, M Furukawa, M Nemoto and T G Langdon, Metall. Mater. Trans. A 1998; 29:2011.
[13] MK Miller, A Cerezo, MG Hetherington and GDW Smith, Atom probe field ion microscopy. Oxford: Clarendon Press; 1996, p476.
[14] K. Marsuda, Y Sakaguchi, Y Miyama, Y Uetani, T Sato, A Kamio and S Ikeno, J Mater. Sci. 2000; 35:179.
[15] CD Marioara, SJ Andersen, HW Zandbergen, and R Holmestad, Metall. Mater. Trans. A 2005; 36:691.