Influence of precipitate size and morphology on grain refinement in nickel aluminium bronze

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Abstract. Nickel aluminium bronze (NAB) was subjected to equal channel angular pressing (ECAP) using routes B_A and C at 400\degree C to investigate the effect of precipitate size and morphology on grain refinement in low stacking fault energy alloys. Both routes produced dynamically recrystallised grains of ~550 nm in size although only route B_A was able to create a uniform distribution of the refined grains. The large unrefined regions in NAB processed via route C was thought to arise from its inability to redistribute the various precipitate phases, as recrystallisation was enhanced around the coarse $\kappa_{II}$ rosettes and refined $\kappa_{III}$ lamellae but reduced in the areas containing fine $\kappa_{IV}$ precipitates.

1. Introduction
The interactions between dislocations and precipitates are well known for their ability to increase strength and work hardening rates [1, 2]. The dislocation structures resulting from these interactions are highly dependent on the size and morphology of the precipitate phase [3-8] and can have either a beneficial [3, 4] or detrimental [5] effect on recrystallisation and grain refinement. Understanding how best to utilise these effects is of great importance to ultrafine grained (UFG) materials produced through severe plastic deformation (SPD), as grain refinement could be completed at lower strains compared to their single phase counterparts. Previous studies have analysed the effect of precipitate size in aluminium following equal channel angular pressing (ECAP) [3-5]. Being a high stacking fault energy (SFE) material, aluminium undergoes grain refinement through the recovery of tangled dislocations into low angle grain boundary (LAGB) cells and the subdivision of the original grains by high angle grain boundaries (HAGBs) originating from dense dislocation walls and microbands [9-15]. Introducing very fine precipitates (~20 nm) was seen to inhibit recovery and the formation of LAGB cells as well as to suppress microband formation through homogenising the slip [5]. In contrast, matrix rotations around coarse precipitates (~2 $\mu$m) led to the rapid production of HAGBs which subdivided the original grains to a greater extent than in the single phase material for the same strain [3, 4], thus accelerating the transformation from a coarse-grained to a UFG microstructure.

While these results are useful for high SFE materials, the limited cross-slip and recovery in low SFE materials result in new grain refinement processes based on twinning [16-18], and potential changes to the influence of precipitate size and morphology on grain refinement. However, little attention has been paid to the effect of precipitates on low SFE materials processed by SPD. Nickel aluminium bronze (NAB) is an ideal material to investigate such changes, as its matrix is similar to the well researched single phase low SFE Al-bronzes [17, 18] while also containing coarse, fine and...
lamellar precipitates [19-21]. The influence of the lamellar phase is of particular interest as it has been shown to break down into fine fragments following ECAP in route B_A [22] whereas a near lamellar structure is kept in route C [22, 23]. This paper aims to use electron backscatter diffraction (EBSD) to examine the final NAB microstructures following ECAP and highlight the role of routes and precipitate size and morphology on grain refinement.

2. Experimental materials and procedures
A cast NAB plate with a composition of Cu–8.8 Al–4.4 Fe–5.2 Ni–1.1 Mn (wt.%) was supplied by VEEM Australia, after heat treatment at 675°C for 6 h to transform any retained β’ phase into α + κ_{III} [21]. The plate was machined into bars of 8 × 8 × 100 mm, which were lubricated using graphite before being inserted into a 90° ECAP die with a cross section of 9 × 9 mm. The die was heated to 400°C and after the temperature stabilised, pressing was conducted at 5 mm/min. In addition to a forward plunger in the entrance channel, a back plunger was placed in the exit channel to provide a constant back pressure of 50 MPa during ECAP. Samples were pressed for 4 passes via route B_A and 8 passes via route C, with samples quenched upon retrieval from the die. Microstructures were examined using scanning electron microscopy (SEM, backscattered electron imaging at an accelerating voltage of 15 kV) on samples prepared to 0.5 μm diamond polish before finishing with colloidal silica. Grain structures were then analysed using electron backscatter diffraction (EBSD), using an accelerating voltage of 12 kV and step sizes of 1500, 100 and 50 nm for the as-received, route C and route B_A samples, respectively, prepared using electropolishing at 30 V in a solution of 30% nitric acid in methanol at –32°C.

3. Results

Figure 1. (a) SEM and (b) EBSD microstructures of the as-received NAB, with different colours denoting different boundary types including HAGBs (black), LAGBs (grey), twin boundaries (red) and various coincidence site lattice boundaries (other colours).

The SEM and EBSD microstructures for the as-received NAB are shown in figures 1a and 1b, respectively. The κ_{II} rosettes, lamellar κ_{III} and κ_{IV} precipitates [19, 20] were all present in the α matrix (arrowed, figure 1a), although the larger κ_1 rosettes were not seen. Due to the evolution of the various phases during cooling [21], the lamellar regions can be easily identified, and the κ_I phase is only found in the areas where κ_{III} is present. From the EBSD mapping (figure 1b), the grains were largely equiaxed with an average size of ~40 μm, with many containing a single twin and very few LAGBs.
The microstructures following 4 passes via route B_A are shown in figure 2. In contrast to the as-received one, the route B_A microstructure is homogeneous, with little distinction between the k_{IV} and the refined k_{III} (figure 2a). While most of the k_{III} had broken down into fine fragments similar in size to the k_{IV} (~350 nm), some had transformed into coarser spheroids (~1 μm). The EBSD mapping (figure 2b) also shows a uniform grain structure consisting mostly of recrystallised grains (~550 nm in size) with annealing twins, as well as a few small pockets dense with LAGBs.

Figure 2. (a) SEM and (b) EBSD microstructures following 4 passes of ECAP at 400°C via route B_A, showing uniform precipitate and grain structures with a few small pockets dense with LAGBs (arrowed). The different coloured lines have the same meanings as in figure 1, with the dark areas indicating the presence of precipitates.

The microstructures following 8 passes of ECAP at 400°C via route C. Both grain and precipitate structures are inhomogeneous, with fine grains found only in the areas containing k_{III} (arrowed 1) while the areas containing k_{IV} remain unrefined (arrow 2). The different coloured lines have the same meanings as in figure 1, with the dark areas indicating the presence of precipitates.

Figure 3. (a) SEM and (b) EBSD microstructures following 8 passes of ECAP at 400°C via route C. Both grain and precipitate structures are inhomogeneous, with fine grains found only in the areas containing k_{III} (arrowed 1) while the areas containing k_{IV} remain unrefined (arrow 2). The different coloured lines have the same meanings as in figure 1, with the dark areas indicating the presence of precipitates.
The microstructures following 8 passes via route C are shown in figure 3. Unlike route B, route C is unable to fully break down the lamellae (figure 3a) even after twice the number of passes. This is due to the restorative action of redundant strain on each even numbered pass, which prevents the redistribution of the $\kappa_{III}$ phase and maintains its lamellar morphology [22]. The grain structure shown by EBSD mapping (figure 3b) is also vastly different, with distinct areas of refined and unrefined grains. While the recrystallised grains were of similar size to those via route $B_A$, they could only be found in the vicinity of the $\kappa_{II}$ rosettes and refined $\kappa_{IV}$. In contrast, the areas of $\alpha + \kappa_{IV}$ remained unrefined and were found to have a cellular structure of LAGBs. Such cells are uncommon for low SFE materials in which linear arrays of stacking faults or twins along a preferred crystal orientation usually form [16].

4. Discussion

4.1. Influence of precipitates on grain refinement

The above results indicate that grain refinement in NAB at 400°C is governed by dynamic recrystallisation and any enhancement of grain refinement must stem from an increase in the recrystallisation sites. Precipitates larger than 1 $\mu$m, such as the $\kappa_{II}$ rosettes, are particularly well suited to this task as they rapidly produce 'deformation zones' with locally high dislocation densities and lattice rotations necessary for recrystallisation. This process is known as particle stimulated nucleation (PSN) [8] and is not seen for finer precipitates in which deformation is more readily accommodated through the punching of prismatic dislocation loops [6, 7]. This may explain the distinct lack of recrystallised grains in the vicinity of the $\kappa_{IV}$ particles for route C, as the precipitates are too small to cause PSN. In the absence of precipitates, a critical boundary misorientation is required before recrystallisation can take place [8]. While a high concentration of LAGBs exists around the $\kappa_{IV}$ particles, these boundaries are insufficient to act as nucleation sites. It remains to be seen whether these LAGB structures are an improvement over those in the single phase material.

The unchanging nature of the $\kappa_{II}$ and $\kappa_{IV}$ makes their effect on recrystallisation easier to interpret, unlike the $\kappa_{III}$ which is seen to evolve from the original lamellae to either fragments or spheroids. Regardless of the final morphology, recrystallisation is promoted around all the refined $\kappa_{III}$. This is to be expected around the spheroidised $\kappa_{III}$, as the transformation coarsens them to $\sim 1 \mu$m when PSN becomes active. The same process, however, cannot apply to the fragmented $\kappa_{III}$ as it remains the size of the $\kappa_{IV}$. It is therefore believed that the enhanced nucleation originates at an earlier stage where the lamellae are still intact. Further EBSD studies are required to analyse the developing grain structures during the initial few passes of ECAP to pinpoint this mechanism.

4.2 Effect of ECAP routes on grain refinement

Although PSN is seen as the main contributor to the enhanced grain refinement following ECAP at 400°C, recrystallisation through PSN is limited to the deformation zones around each precipitate [8]. This can be clearly seen in route C with recrystallised grains solely in the vicinity of the $\kappa_{II}$ or $\kappa_{III}$ particles, despite the high LAGB density in the neighbouring $\kappa_{IV}$ regions. At the same time, the microstructure via route $B_A$ consists almost entirely of recrystallised grains, complementing its homogeneous distribution of precipitate phases. The homogeneity in precipitate distribution is attained through the non-redundant strain applied in route $B_A$, which spreads the various phases throughout the microstructure. As the $\kappa_{III}$ and $\kappa_{IV}$ are forced into increasingly thin layers, the deformation zones around the $\kappa_{III}$ are more likely to overlap with the $\kappa_{IV}$ regions, thus allowing the whole microstructure to become recrystallised. In route C, strain is reversed on even numbered passes and this limits the extent of redistribution and the interior of the $\kappa_{IV}$ regions remain unaffected.

Another contributing factor leading to more efficient grain refinement via route $B_A$ may lie in its four unique shear planes compared to the single shear plane in route C [24] upon which the shear direction reverses on each pass. It has been shown that processes with strain reversals were only 65%
as effective in promoting recrystallisation as those in which strain is applied in a single direction [8]. The effect of this redundant strain must be significant, as the route C sample has had twice the total strain as route B yet the $\kappa_{IV}$ areas show no sign of recrystallisation.

5. Conclusions

(1) Grain refinement in NAB following ECAP at 400°C takes place through dynamic recrystallisation, producing equiaxed grains of ~550 nm in size.

(2) ECAP routes have a significant influence on the extent of grain refinement, with route B producing a microstructure consisting mostly of refined grains but route C giving rise to large unrefined regions.

(3) The nucleation of recrystallised grains is dependent on the size and morphology of the surrounding precipitate phases. The coarse $\kappa_{III}$ rosettes enhance the nucleation of new grains through particle stimulated nucleation. Nucleation is also enhanced around the refined $\kappa_{III}$, although the changing morphology makes the precise mechanism difficult to identify.

(4) The fine $\kappa_{IV}$ precipitates limit nucleation, leading to the associated unrefined areas via route C.

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