Full length article

Microstructural evolution, strengthening and thermal stability of an ultrafine-grained Al–Cu–Mg alloy

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ABSTRACT

To gain insight into the origin of the ultra-high strength of ultrafine-grained (UFG) alloys, the solute clustering, precipitation phenomena, and microstructural evolutions were studied in an UFG Al-4.63Cu-1.51 Mg (wt.%) alloy (AA2024) processed by high-pressure torsion (HPT). The thermal analysis was performed using differential scanning calorimetry. The microstructures, internal microstrains and hardness following heating at a constant rate were characterised at room temperature using X-ray diffraction (XRD), transmission electron microscopy (TEM) and atom probe tomography (APT). The microhardness of the HPT processed sample initially increases following heating to 140 °C, and then remains unchanged on further heating to 210 °C. As the temperature increases up to 210 °C, the crystallite size calculated from XRD line broadening remains about 60–70 nm, while the dislocation densities remain in excess of 2×10^{14} m^{-2}. A multimechanistic model is established to describe the strengthening due to grain refinement, dislocation accumulation, solid solution, precipitation, solute clusters and their segregation. The analysis reveals that solute clusters and lattice defects are key factors in HPT-induced strengthening of alloys, and illustrates the interactions between alloying elements, dislocations and grain boundaries enhance strength and stabilize ultrafine microstructures. Furthermore, for an HPT sample heated beyond 210 °C, the formation of nano-precipitates also contributes to hardness increment. The multimechanistic model for hardness contribution indicates the short-range order strengthening due to cluster-defect complexes is the dominant mechanism, which accounts for more than 40% of overall hardness.

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1. Introduction

Severe plastic deformation (SPD) methods have attracted wide attentions as effective techniques to improve the mechanical properties of metals and alloys without changing their overall dimensions [1]. The high-pressure torsion (HPT) process has advantages over other SPD techniques due to its high efficiency in inducing a large density of dislocations and its good capability for continuous deformation. Over 20 years of research in HPT has demonstrated that the hardness and the strength of materials increase and often evolve to saturation with large straining [2–4].

A number of strengthening models have been developed to incorporate work hardening and grain refinement hardening [5–11]. A model predicting dislocation generation due to non-shearable particles was first proposed by Ashby [5]. The concept of dislocations generated and annihilated in grain boundaries then was incorporated in the models developed by Estrin and Mecking [11] and further refinements [7,8]. Further modified models have been applied to predict strength of SPD-processed aluminium and its alloys [9,10]. However, in these strengthening models, the alloying atom effects have not received much attention. The study by Edalati et al. [12] indicates that the solute atoms lead to extra grain refinement, causing an extra increase in strength. Recent advances in atom probe tomography (APT) have revealed solute atoms significantly segregate to grain boundaries in 7xxx and 6xxx Al alloys [13–15] and some pre-existing precipitates are fragmented or partially dissolved in Al–Cu alloys [16–20]. In their

studies on Al alloys [13–20], the increase of hardness can be partially attributed to the solute segregation and precipitate fragmentation.

In a precipitation-strengthening Al–Cu–Mg alloy, nanosized Cu–Mg clusters and/or uniformly dispersed precipitates are presented within the Al matrix after ageing heat treatment [21–23]. The investigation indicates both Cu–Mg clusters and S-phase contribute to overall strengthening in Al–Cu–Mg alloy during the heat treatment processes [24]. A specific question comes out - what will be the solute clusters and dispersed precipitates behaviour in heating processes, if the samples contain a large number of dislocations. Obviously, a large amount of energy is stored in the lattice defects as dislocations accumulate to grain boundaries and the grains become ultrafine [25–28]. At the same time, alloying elements are able to reduce the grain-boundary energy and enhance the thermal stability of nanostructured alloys [13,29,30]. It is essential to develop a deep understanding of the mechanisms that govern the redistribution of solute clusters, precipitates and dislocations.

More recently, the studies on the precise determination of the underlying strengthening mechanisms for precipitation-strengthening Al alloys processed by SPD were discussed [31–33]. It was suggested that the dramatic increase of strength after SPD process was attributed to the multiple mechanisms which comprised strengthening due to grain boundaries, dislocations, solid solution and precipitates [31,32]. However, most of recent predictions do not consider the co-cluster effects in the origin stage of HPT-processed Al–Cu–Mg alloys. Our recent studies [33] indicate the defect−cluster complexes are formed in ternary Al alloys when the number of dislocations and vacancies reach its saturation after HPT processes. The defect−cluster complexes are thermally stable and contribute to a primary part in strengthening. It is of great importance to formulate a quantitative insight into strengthening mechanisms, providing a direct guide of the microstructural modifications for industrial heat treatment.

The previous analysis [34–37] on small Cu–Mg clusters and dispersed precipitates were trying to reveal the essence of the strength increase in the heat-treated Al–Cu–Mg alloys. To the best of our knowledge, the quantitative predictions of strengthening at an increasing temperature have not been reported for UFG Al–Cu–Mg alloys, and this is the first time to address the multiple strengthening mechanisms that comprised cluster-defect complexes strengthening. The aim of this work is to investigate solute redistribution, evolution of dislocation density and nanoscale precipitation during linear heating of HPT-processed AA2024 alloy, mainly using APT and X-ray diffraction line broadening.

2. Experimental procedures

UFG Al-4.63Cu-1.51Mg (*wt.* %) (AA2024) samples were processed by HPT for five revolutions (5r-HPT) at room temperature, under a pressure of 6 GPa with a speed of 1 rpm. The composition of the alloy is shown in Table 1, and the as-received specimens were in the T351 temper. Prior to HPT, the samples were cut to discs with a diameter of 9.8 mm and mechanically ground and polished to 0.83−0.85 mm in thickness. Details of the HPT processing were explained previously [9,38].

Thermal analysis was performed using a differential scanning calorimeter (Perkin−Elmer DSC1) at heating rate of 10 °C/min in a

Table 1
Composition (wt.%) of the Al−Cu−Mg alloy studied.

Cu	Fe	Mg	Mn	Si	Zn	Total others	Al
4.63	0.15	1.51	0.66	0.08	0.05	<0.05	balance

flowing N_2 atmosphere from ambient temperature to 540 °C. The samples were put in copper pans under tight-fitted inverted lids, with an empty copper pan as the reference. Prior to DSC experiments, the near disk shaped samples were punched from the HPT disk at 4 mm from the centre using a 5 mm diameter punch. Each sample is approximately 20 mg. The DSC thermograms were corrected by subtracting a baseline run with an empty copper pan and a further baseline correction as described in Ref. [39] was applied.

Additionally, HPT processed discs were heated and held at temperature T_{iso} in a furnace. The hold time was chosen such that the heat treatment is equivalent to heating at the DSC heating rate, β, to the final temperature, T_f. The equivalent time t_{eq} of an isothermal heat treatment at T_{iso} has been approximated by using the theory for thermally activated reactions [39], which provides:

$$t_{eq} \cong 0.786 T_f \big/ \beta \left(RT_f \big/ E \right)^{0.95} exp\left(-E \big/ RT_f \right) \left[exp\left(-E / RT_{iso} \right) \right]^{-1}$$

(1)

where E is the activation energy of the reaction, taken as 107 kJ/mol [40]. Thus, the HPT samples were heat treated for the calculated equivalent time t_{eq} at the same temperature in the DSC curve ($T_{iso} = T_f$), e.g. a sample isothermal annealed at 210 °C for 2 min equivalent to that DSC linear heated up to 210 °C with a constant heating rate, 10 °C/min. These equivalent heat treatments were applied on the samples for hardness, XRD, TEM and APT tests.

Vickers hardness was measured on T351 and 5r-HPT processed samples after heating to selected temperatures in the range 100−400 °C and with different periods of holding time to produce the equivalent treatment ($T_{iso} = T_f$) defined by Eq. (1), followed by rapid cooling (cooling rate ~ 50 K/min). The hardness tests were conducted at room temperature immediately after completion of the cooling. Each sample was prepared by grinding to 4000-grid SiC paper and polishing to a mirror-like surface. The hardness tests were conducted using a load of 500 g for a dwell time of 15 s. Each reported value is the mean of 6 indentations all made at a distance of 4 mm from the centre of the disc.

XRD was carried out using a Siemens D5000 diffractometer equipped with a graphite monochromator using Cu $K\alpha$ radiation at 50 steps per degree and a count time of 1s per step. The Maud software [41–43] was used to analyse the microstrains and the crystallite sizes of HPT-processed samples and those under isothermal heat treatment. The analysis of XRD peak broadening is based on the full peak X-ray profile refinement (Rietveld method) [42,44,45].

TEM was performed on HPT-processed samples and heat-treated samples using a JEOL 3100 TEM operated at 300 kV. The samples were first ground to ~150 μm in thickness, and disc shaped samples of 3 mm in diameter were punched out at about 4 mm from the centre of a disk. Subsequently, the samples were thinned to perforation using twin-jet electropolishing at −30 °C with an electrolyte solution of HNO_3: methanol = 1:3 (in volume). The statistic average grain sizes were obtained based on 20 TEM images. Selected area diffraction (SAD) patterns were taken with an aperture of ~1 μm².

APT specimens in the form of sharp needles with an end radius of less than 100 nm were prepared from blanks with dimensions of $0.5 \times 0.5 \times 10$ mm³ using a standard two-stage electro polishing technique. The first stage used a solution of 25% perchloric acid in acetic acid at 15 V, whereas the second stage used an electrolyte of 5% perchloric acid in 2-butoxyethanol at 20 V. APT analysis was carried out under an ultrahigh vacuum (~1 × 10^{-8} Pa), UV laser pulsing energy of 40 pJ at the pulse repetition rate of 200 kHz and a specimen temperature of ~20−25 K using a local electrode atom probe (LEAP4000X SI®). Reconstruction and visualization of APT