Microstructure and properties of ultrafine grained structure of Cu-Zn-Si alloy fabricated by heavy cold rolling

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Abstract. Cu-18.2Zn-1.5Si-0.25Fe (mass%) alloy was heavily cold rolled. Ultrafine grained (UFGed) structure, containing a mixture of lamellar and mechanical twins, was easily and homogeneously formed. The average grain size was approximately 100 nm. The as-rolled sample showed quite high ultimate tensile strength (UTS) over 1 GPa. The UTS was higher than those obtained by multi directional forging. When the samples were annealed at relatively low temperatures between 553 K and 653 K, they showed slight hardening followed by large softening due to occurrence of static recrystallization (SRX). Annealing of UFGed structure at relatively low temperature of around 0.4 Tm caused extensive SRX that, in turn, induces ultrafine RXed grained structure. The grain size of the RXed sample was as fine as 200 nm. Although the annealing induced recovery of ductility while UTS gradually reduces, UTS over 1 GPa with ductility of 15 % were attained. The RXed grains mainly contained ultrafine annealing twins. Therefore, UFGed structure and superior mechanical properties could be achieved by a simple process of cold rolling, i.e., without severe plastic deformation.

Keywords: Mechanical and annealing twins, cold rolling, mechanical property

1. Introduction

Various kinds of processes for severe plastic deformation (SPD) are proposed to attain bulk ultrafine grained (UFGed) metallic materials and are actually produced [1-5]. It is being known that the UFGed materials possess specific properties in addition to high strength. The sizes of the UFGed materials processed by SPD methods are, however, in
general too small to employ as structural ones. A new method for the fabrication of bulky UFGed metallic materials is therefore highly desirable. The minimum grain size achieved by SPD methods seems to be around 0.2 µm independently of the process. Recently, Miura et al. [3] have reported that strain-induced grain refinement during multi-directional forging can be enhanced by mechanical twinning. The formation of numerous mechanical twins involves the subdivision and fragmentation of the initial grains. By employing this mechanism for the grain refinement during multi-directional forging of Cu-Zn alloy, a homogeneous microstructure of about 20 nm in diameter was produced. They applied the twinning induced grain refinement mechanism also to heavy cold rolling of a Cu-Zn alloy and attained UFGed structure where thin lamellar structure was subdivided by mechanical twins [6]. Furthermore, UFGed recrystallized (RXed) structure with an average grain size of 200 nm was achieved by additional annealing of the heavily cold rolled Cu-Zn alloy. Their results suggest that formation UFGed structure should be emphasized in materials with lower stacking fault energy (SFE) by the formation of more mechanical and annealing twins even through simple heavy cold rolling. This is the motivation of the present study.

2. Experimental

A Cu-18.2Zn-1.5Si-0.25Fe (mass%) alloy plate with 5.0 mm thick and having an initial grain size of 9.4 µm was cold rolled down to 0.7 mm or 1.8 mm. Si addition to a Cu-Zn alloy is expected to lower the SFE. Some of them were cryogenically rolled after soaking in liquid nitrogen for about 10 minutes. They were subsequently annealed at selected temperatures from 653 K down to 533K for various periods of time. The processes and the experimental procedure are schematically described in Fig. 1. The microstructures produced were examined in detail by transmission electron microscopy (TEM) at an accelerating voltage of 200 kV. The changes in hardness during annealing were examined using a Vickers micro-hardness tester. Tensile tests, samples with gage length 6 mm and

Fig. 1 Schematic representation of the processes and the experimental procedure.
width 3 mm, were carried out parallel to the rolling direction at ambient temperature at an initial strain rate of $1.5 \times 10^{-3}$ s$^{-1}$. In the C route, sample cracked at 64.4% reduction during rolling at 77 K. In the present paper, results of A and B routes are mainly reported.

3. Results and discussion
The change in the hardness was shown in Fig. 2. The hardness was first rapidly raised by onset of annealing, and then exhibit plateau or slight decrease followed by abrupt drop. The hardness initial increase is known as low temperature anneal-hardening. The hardening is believed to due to the effect of formation of Cottrell atmospheres and/or by the G.P Zones [7]. The combined effect of a high density of dislocations and widely expanded stacking faults likely to induce such quick and large increase in hardness. The expected dislocation density in such microstructure composed of lamellar structure further subdivided by mechanical twins formed by heavy cold rolling is much higher than $10^{15}$ m$^{-2}$ [6]. This promotes the formation of the atmosphere. As will be mentioned later, the high dislocation density contributed also to static RX at low temperatures around 533 K, i.e., 0.4Tm where Tm the melting temperature. The abrupt drop in the hardness is attributed to the extensive onset of RX, and therefore, indicates that recovery does not play a large role during annealing. As expected, the onset of RX is accelerated with increasing annealing temperature.

Typical TEM microstructures obtained by cold rolling following B route are displayed in Fig. 3. The coarse initial grains were changed to fine lamellar structure parallel to the

![Fig. 2 Hardness change during annealing at various temperatures of the as-rolled samples prepared by (a) A and (b) B routes, respectively.](image-url)
rolling direction (Fig. 3 (a)). The lamellar boundaries were composed of low angle boundaries and mechanical twin boundaries. In particular, it is obvious to see in Fig. 3 (b) that thin lamellar grains with width of about 100 nm were further subdivided by numerous mechanical twins. The mechanical twins are also lamellar in shape and appear in parallel arrays with spacing ranged from 10 to 20 nm. Therefore, UFGed structure with average gain size much less than 100 nm was fabricated by heavy cold rolling employing mechanical twinning in a low SFE material. With increasing amount of reduction, the twin density increases and the distribution becomes more homogeneous.

Figure 4 exhibits a typical TEM micrograph of the fully RXed sample after annealing. It is can be seen that new ultrafine grains developed by RX contain a profusion of

Fig. 4 TEM micrograph of the fully recrystallized B route sample after annealing for 5400 s at 563 K.
annealing twins. The rapid drop in hardness during annealing at 563 K observed in Fig. 2 is, therefore, confirmed to be due to the extensive occurrence of RX. The grains are ranging in size from about 150 nm to 500 nm. Nakao et al. have reported that the temperature for RX is reduced in UFGed structure; full RX at only 423 K in a SPDed Cu-30%Zn alloy [8]. They explained that the low annealing temperature impeded the coarsening of new grains, and still more, the low SFE affected to form ultrafine annealing twins which were thermally stable and hardly migrate [9]. In the present study, the formation of a high density ultrafine annealing twins thermally stabilized the UFGed structure in the similar manner.

The cold rolled samples followed by low temperature annealing were deformed in tension and the flow curves are exhibited in Fig. 5. In all the processes, the as-rolled samples showed quite high ultimate tensile strength (UTS) over 1 GPa. The strength of the as-rolled sample changed depending on the process. In particular the sample prepared by the C route showed UTS about 1.4 GPa, although the flow curve is not shown here. In the B route, the UTS was raised up to 1.3 GPa due to the effect of annealing hardening, while in the strength obtained through the route C rapidly decreased due to the earlier occurrence of RX. These UTS would be nice to have a value or to superimpose the related stress-strain curve obtained by multi-directional forging of Cu-Zn alloy [5]. The UTS gradually decreased with increasing annealing time regardless of the processing route. After full RX, the samples exhibited excellent

![Fig. 5 True stress vs. nominal strain curves obtained by tensile test at room temperature of the annealed samples of (a) A and (b) B routes for various period of time at 563 K.](image)

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mechanical properties of 800 MPa UTS and 40% ductility. The quite high strength in the as-rolled sample and the superior mechanical properties are presumably due to the high density of coherent Σ3 twins. It has been reported that coherent Σ3 twins are the strongest interfaces among all possible grain boundaries [10]. That is, crack initiation or fracture at twin boundaries are more difficult compared with general boundaries. Therefore, the coupled effects of numerous coherent Σ3 mechanical/annealing twins in addition to strain hardening and solid solution hardening effectively contributed to strengthen the alloy.

4. Summary
The evolution of ultrafine grained (UFGed) microstructure in a heavily cold rolled Cu-18.2Zn-1.5Si-0.25Fe (mass%) alloy followed by annealing was investigated. The results obtained are summarized below.
1) UFGed structure was easily formed by heavy cold rolling of a Si added Cu-Zn alloy. By cold rolling, lamellar structure of 200 nm thick was subdivided by dense formation of a high density of mechanical twins with 10 nm to 20 nm spacing.
2) UFGed structure composed of grains ranging from 150 nm to 500 nm was produced by low temperature annealing after cold rolling. The UFGed structure mainly composed of annealing twins appeared thermally stable with limited grain growth after prolonged annealing.
3) UFGed Cu-Zn-Si alloy exhibited quite high ultimate tensile strength of 1.4 GPa. By annealing at relatively low temperatures to form ultrafine annealing twins, excellent mechanical properties, 800 MPa UTS and 40% ductility, were attained.

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