Enhanced structural refinement via intermediate annealing in cold rolled Al-2%Cu

Du, Keming; Huang, Tianlin; Wu, Guilin; Hansen, Niels; Huang, Xiaoxu

Published in: Proceedings of the 40th Risø International Symposium on Materials Science: Metal Microstructures in 2D, 3D and 4D

Publication date: 2019

Document Version Peer reviewed version

Link back to DTU Orbit


General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.
Enhanced structural refinement via intermediate annealing in cold rolled Al-2%Cu

Keming Du1, Tianlin Huang1,2, Guilin Wu1, Niels Hansen1,3 and Xiaoxu Huang1,4
1 International Joint Laboratory for Light Alloys (MOE), College of Materials Science and Engineering, Chongqing University, Chongqing 400044, China
2 Electron Microscopy Center of Chongqing University, Chongqing University, Chongqing 400044, China
3 Risø Campus, Technical University of Denmark, DK-4000 Roskilde, Denmark
4 Department of Mechanical Engineering, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

E-mail: huangtl@cqu.edu.cn

Abstract. Fine scale lamellar structures develop during cold rolling of metals to high strain. However a mechanically assisted triple junction motion process occurs during cold rolling, which removes lamellar boundaries and balances the decrease in lamellar boundary spacing. Segregation and precipitation at the lamellar boundaries are expected to suppress the triple junction motion and therefore enhance the structural refinement. In this study an Al-2wt.%Cu alloy has been cold rolled in two steps with an intermediate annealing: first cold rolled to a 75% thickness reduction forming a microstructure of lamellar morphology, followed by an annealing at 100℃ for 30 minutes, and then further cold rolled to the final thickness reduction of 98%. An average lamellar boundary spacing of 113nm has been achieved, which is much smaller than the value of 139 nm obtained in the sample cold rolled to the same strain but without intermediate annealing.

1. Introduction

It is well-established that plastic deformation of metals refines the microstructure and after large strain the microstructure typically shows a lamellar morphology with finely spaced lamellar boundaries connected by triple junctions. Previous observations showed the lamellar boundary spacing decreases at a very low arte at high strains when compared with the externally imposed shape change [1,2,3,4,5]. In a recent study of cold rolled Al to true strains up to 5.5, Yu et al [6] identified that mechanically assisted triple junction motion occurs along the rolling direction and each event of this triple junction motion replaces two boundaries by one while maintaining the structural morphology. This triple junction motion mechanism causes the approach to a dynamic equilibrium of structural refinement at large strains [1-5]. It is therefore expected that an enhancement of structural refinement may be achieved if the triple junction motion can be suppressed.

Recently, we found that severe boundary segregation of Cu takes place in an Al-0.3%Cu alloy cold rolled to a thickness reduction of 98% [7], which stabilized the microstructure and led to the formation of a well-developed lamellar structure with an average lamellar boundary spacing of 200 nm. This structural refinement is not possible without the addition of the 0.3% Cu [7] and the pinning effect of
Cu segregation on the triple junction motion is considered to play a role in the structural refinement. In this study, we investigate the evolution of deformation microstructure of an Al-2%Cu alloy during cold rolling. An intermediate annealing is introduced during cold rolling to facilitate the segregation of Cu at boundaries with a purpose to enhance the structural refinement.

2. Experimental
The starting material used was a 50 mm thick Al-2%Cu slab, which was produced by combining ultra-high purity (99.9996%) Al with 2 wt.% OFHC Cu. The slab was solution-treated at 490°C for 2 hours and water-quenched, and then cut into two pieces of sample for cold rolling. One piece of the sample was cold rolled by multiple passes to 98% thickness reduction, producing a 1 mm thick sheet (hereafter referred to as DR-98). The other piece of the sample was cold rolled at two steps with an intermediate annealing included between the two rolling steps. At the first step, the sample was cold rolled to 75%, which is a strain at which a microstructure with lamellar morphology in general develops. The rolled sample is then annealed at 100°C for 30 minutes before the second rolling step. After this intermediate recovery annealing, and the sample is further cold rolled to the same total thickness reduction of 98% as that applied to the first sample (referred to as RAR-98).

The microstructure of the DR-98 and RAR-98 samples were characterized by transmission electron microscopy (TEM) with a JEOL 2000FX electron microscope operating at 200 kV. The distribution of Cu was analyzed with a ChemiSTEM system equipped on an FEI Titan G2 60-300 electron microscope. Thin foil samples for TEM observations were prepared from the longitudinal section (containing the normal direction, ND and the rolling direction, RD).

To validate the microstructural results the strength of the two deformed samples was measured by tensile testing, which was conducted with a SHIMAZU AG-X10 electronic universal testing machine at an initial strain rate of $2 \times 10^{-4}$/s at room temperature.

3. Results and discussion
Fig. 1 shows the microstructure of the DR-98 and RAR-98 samples. In both cases, lamellar structures (fig. 1a and b) are formed that contain a high density of dislocations in the volume between the lamellar boundaries. Note that the lamellar boundaries in the DR-98 sample in some regions (fig. 1a) seem to be not so straight as in the RAR-98 sample (fig. 1b).

![Fig. 1 TEM images showing the lamellar structures formed in (a) DR-98 and (b) RAR-98 samples.](image-url)
The lamellar boundary spacings were measured from TEM images by intersecting method along the ND. More than 500 boundary spacings were measured for each sample and the distributions of the measured spacings are shown in fig. 2. The spread and average value of the lamellar boundary spacings are larger in DR-98 than in RAR-98. The average spacings were determined to be 139 nm and 113 nm, respectively, for the RAR-98 and RAR samples, showing clearly an enhancement of structural refinement due to the intermediate annealing.

![Fig. 2 Distributions of lamellar boundary spacings. (a) DR-98; (b) RAR-98](image)

To study the effect of the intermediate annealing on the structural refinement, the microstructures of the 75% cold rolled sample and the sample after subsequent intermediate annealing were characterized. The majority of the microstructure in the 75% cold rolled state is composed of lamellar structures although some of the boundaries have relatively larger inclination angles to the RD (fig. 3a). After recovery annealing at 100 ℃ for 30 minutes, the lamellar morphology shows no change (fig. 3b), but many fine particles are formed. Fig. 3c shows a higher magnification TEM image of the annealed sample. A high density of very fine particles can be seen. A ChemiSTEM map of the area in fig. 3c was obtained and is shown in fig. 3d. The ChemiSTEM map reveals that the fine particles have high Cu contents. Many of these Cu-enriched particles are formed at the triple junctions and lamellar boundaries (as indicated by arrows). An early study by Silcock et al. [8] showed that without pre-deformation the precipitation kinetics is very slow in a solution-treated Al-2%Cu alloy at low temperatures; there was almost no precipitation at 130 ℃ for aging up to 10 days. Nevertheless, the high density of dislocations and boundaries after 75% cold rolling are expected to accelerate the formation of fine particles. It should be pointed out that the boundary segregation of Cu is not clearly seen on the ChemiSTEM maps. One of the reasons may be that the ratio between the Cu concentration at the grain boundaries and in the matrix is not so high due to the high percentage of the Cu content in the matrix. In recent studies, we observed high tendency of Cu segregation at boundaries after 98% cold rolling in a Al-0.3%Cu alloy [7,9] where the Cu concentration at the grain boundaries is 20 times higher than in the matrix. In a nanostructured Al-2.5%Cu alloy [10], the segregation of Cu was clearly manifested by three dimensional atom probe (3DAP). 3DAP analysis is undertaken for the present Al-2%Cu alloy to quantify of the Cu segregation at grain boundaries. It is believed that the intermediate annealing has enhanced precipitation and segregation at the triple junctions and boundaries, which stabilize the boundaries and prevent the triple junction motion during the subsequent deformation, and therefore facilitated the formation of a finer lamellar structure.

The microhardness of the 75% cold rolled sample was measured to be 90 HV, which slightly decreased to 87 HV after the intermediate annealing at 100 ℃ 30 minutes. This heat treatment has
concurrent effects on recovery of the deformed microstructure (e.g. dislocation annihilation) and precipitation, which are expected to cause softening and hardening, respectively. The small decrease in hardness indicates that the softening effect is slightly stronger than the hardening effect.

Recently, Azad and Borhani [11] investigated the pre-aging effect on the structural refinement in an Al-2%Cu during accumulative roll bonding (ARB). After solid solution treatment and pre-aging at 190 °C for 10 and 30 minutes, they deformed the two pre-aged samples by six-cycle ARB processing that accumulated a total true strain of 4.8. They characterized the deformed microstructures by electron backscatter diffraction and found that lamellar structures are formed in the two samples with different pre-aging times but the average lamellar boundary spacings are 420 nm and 400 nm, respectively, which are much coarser than the present results obtained in the DR-98 and RAR-98 samples. The EBSD technique they used may not allow them to reveal the low angle lamellar boundaries, however, the coarser microstructures are also suggested by the much lower yield stress they measured, which is to be discussed later in the following.

Fig. 3 The microstructure of (a) sample cold rolled to 75%; (b) sample cold rolled to 75% and annealed at 100°C for 30 minutes; (c) sample same as (b) at a higher magnification. d) ChemiSTEM map of the region shown in (c).
Fig. 4 shows the tensile stress-strain curves of the DR-98 and RAR-98 samples. The tensile stress-strain curve of a coarse grained sample, which was obtained by annealing the 98% cold rolled sample at 300℃ for 30 minutes, is included for comparison. The coarse-grained material has a yield strength of 23.0 MPa and a UTS of 91.3 MPa. The low yield strength obtained in this coarse-grained sample suggests that the strengthening of Cu in solid solution is rather small in the well-annealed state. The yield strength and the UTS of the DR-98 sample are 289.5MPa and 315.8MPa, respectively. For the RAR-98 sample, the yield strength and UTS are 343.4MPa and 352.3MPa, respectively, showing a clear improvement as compared to the DR-98 sample. This improvement is considered to be mainly related to the structural refinement due to the intermediate annealing. Note that the yield strength of the RAR-98 sample is much higher than that values (200 – 220 MPa) reported by Azad and Borhani in the pre-aged and 6 cycles ARB processed Al-2%Cu alloy [11]. The much finer lamellar boundary spacing and the much higher yield strength obtained in the present RAR sample as compared with the pre-aged and ARB processed similar alloy suggests that the intermediate annealing is more effective than the pre-aging for the structural refinement in Al-Cu alloys.

**Fig. 4 Tensile stress-strain curves of the Al-2%Cu alloy after different deformation and annealing**

4. Conclusions

Al-2wt.%Cu alloy with ultrahigh purity matrix has been cold rolled to a reduction of 98% by two procedures. One is a direct cold rolling by multiple passes to 98% (DR-98) and the other is a two-step cold rolling with an intermediate annealing: first cold roll to 75%, followed by an annealing at 100℃ for 30 minutes, and then cold rolled to the final thickness (RAR-98). An average lamellar boundary spacing of 113nm has been achieved in the RAR-98 sample, which is much smaller than the value of 139 nm obtained in the DR-98 sample. Segregation and precipitation at the lamellar boundaries during the intermediate annealing is considered to be the key reason for the enhanced structural refinement due to their effects on the suppression of triple junction motion during the second step cold rolling.

**Acknowledgments**

The authors wish to thank the financial support of the State Key Research and Development Program of MOST of China (2016YFB0700400), Natural Science Foundation of China (NSFC, Grants No. 51421001). NH thanks the support of the 111 Project (B16007) by the Ministry of Education and the State Administration of Foreign Experts Affairs, China.

**References**