Materials Science & Engineering A 585 (2013) 178–189

Contents lists available at ScienceDirect

Materials Science & Engineering A

journal homepage: www.elsevier.com/locate/msea

Annealing behavior of cryogenically-rolled copper

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ARTICLE INFO

Article history:
Received 8 May 2013
Received in revised form
30 June 2013
Accepted 4 July 2013
Available online 31 July 2013

Keywords:
EBSD
Nanostructured materials
Thermomechanical processing
Recrystallization
Grain growth

ABSTRACT

The static annealing behavior of cryogenically-rolled copper over a wide temperature range (50–950 °C) was established. At temperatures below 350 °C ($\sim 0.5T_m$), microstructure and texture evolution were interpreted in terms of discontinuous recrystallization. Grains having orientations close to (55;30/60;0), {236}⟨385⟩ (Brass-R), and {4;4;11}⟨11;11;8⟩ (Dillamore) were shown to recover rapidly and thus exhibited preferential growth during subsequent static recrystallization. At temperatures of 350 °C and higher, annealing behavior was dominated by abnormal grain growth. The abnormal character of this process was attributed to the relatively large spread in grain sizes produced during preceding recrystallization.

1. Introduction

There is significant commercial interest in the development of materials with ultrafine grain structures for structural applications. This interest is mainly driven by a substantial improvement in strength and ductility as well as a good balance of these properties. In addition, the control of the mechanical properties by processing may be an attractive alternative to expensive alloying. This could result in the use of fewer and simpler industrial alloys and would lead to economic benefits as well as improved recyclability.

Techniques for the production of fine-grain alloys are thus of considerable commercial interest. Of particular importance are cost-effective methods that can be used to obtain large quantities of such materials. In this regard, an approach involving large deformation at cryogenic temperatures has recently attracted significant attention. It is believed that low temperatures may suppress dynamic recovery and stimulate mechanical twinning (e.g., [1, 2]) thereby enhancing the grain-refinement effect. This may decrease the level of strain necessary to achieve an ultra-fine microstructure and thus enable the application of conventional working processes such as rolling to produce such materials.

To date, the majority of research in the field of cryogenic working has focused on aluminum and copper alloys [e.g., 1–7], most likely because of the superior ductility of these materials. It has been established that the key mechanism governing grain-structure evolution in both materials at cryogenic temperatures is the geometrical effect of strain per se. In other words, grains change their shape in proportion to the imposed strain, and noticeable grain subdivision and mechanical twinning are not observed [3,6]. By this means, a reasonably homogeneous grain structure, dominated by heavily elongated grains aligned with the direction of macroscopic material flow, is developed. Such grain structures typically contain a significant proportion of low-angle boundaries [3,6] and, in the case of copper, a high density of free dislocations [6]. The limited formation of deformation-induced boundaries during cryogenic deformation is believed to be partially associated with suppression of cross-slip at low temperatures [6]. This effect is also responsible for the strengthening of the {110}⟨112⟩ Brass texture in cryo-rolled materials [3,6]. Despite these characteristics, cryogenic deformation has been shown to be able to produce ultra-fine grain structures in both materials in some cases [3,6].

Not surprisingly, one important requirement to obtain and maintain a fine-grain microstructure is a high degree of microstructural stability within the expected range of use temperature and time. Hence, the annealing behavior of such materials is of interest. For cryo-deformed aluminum, this issue has been studied systematically by Zahid et al. [8]. For example, it was found that the fraction of low-angle boundaries increased progressively with

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http://dx.doi.org/10.1016/j.msea.2013.07.042

annealing temperature, leading eventually to discontinuous recrystallization. At intermediate temperatures, such behavior resulted in a bimodal structure comprising bands of coarse grains and fine subgrains. This unusual behavior has also been related to a strong texture in deformed material giving rise to the so-called "orientation pinning" effect [9] during subsequent grain growth.

In contrast to aluminum, relatively little attention has been paid to the annealing behavior of cryo-deformed copper. It has been demonstrated that this material is often unstable at low and ambient temperatures; i.e., it is prone to discontinuous recrystallization or abnormal grain growth [6,10–13]. However, a comprehensive understanding of the effect of annealing temperature on microstructure and texture evolution of cryo-deformed copper is still lacking. The objective of the present work was to fill this gap in knowledge.

2. Material and experimental procedures

The material used in the present work consisted of 99.9 wt pct. pure copper supplied as a hot-rolled bar. The as-received material was preconditioned by severe "abc" deformation [14] and then cryogenically rolled to 90 pct. overall thickness reduction (true strain = −2.3). The total thickness reduction was achieved using multiple passes of ~10 pct. each. In order to provide cryogenic-deformation conditions, the rolling perform and work rolls were soaked in liquid nitrogen prior to each pass and held for 20 min. Immediately after the pass, the workpiece was re-inserted into liquid nitrogen. The total time of each pass (i.e., the exposure time of the specimen under ambient conditions) was only a few seconds. Heat transfer calculations revealed that the warming of the rolls and copper specimens prior to rolling due to free convection in air was small, resulting in temperature increases of the order only ~1–4 °C. Additional details of the cryo-rolling process are described elsewhere [6].

To investigate the subsequent annealing behavior of the cryo-rolled material, samples were furnace annealed over a range of temperatures from 50 to 950 °C for 1 h (+10 min for heatup), as well as isothermally for various times at 150 and 450 °C. For the isothermal tests, the specimen temperature was continuously monitored by a thermocouple. Following heat treatment, each specimen was quenched in water. To preserve the microstructures developed during each thermomechanical treatment, the cryo-rolled as well as annealed samples were stored in a freezer at ~ −20 °C prior to examination.

To obtain insight into the three-dimensional nature of microstructure development, metallurgical observations for the as-rolled and the rolled-and-annealed samples were made in the plane containing the RD and ND (i.e., the longitudinal plane) as well as in the plane containing the RD and TD (i.e., the rolling plane). (Per the typical flat-rolling convention, RD denotes the rolling direction, TD the transverse direction, and ND the normal direction of the rolled sheet.) Specifically, microstructure and texture were determined using the electron backscatter diffraction (EBSD) technique. For this purpose, samples were prepared using conventional metallographic techniques followed by electro-polishing in a solution of 70 pct. orthophosphoric acid in water at ambient temperature with an applied potential of 5 V. In the longitudinal plane, all observations were made at the mid-thickness of the rolled sheet.

High-resolution EBSD analysis was conducted using a Hitachi S-4300SE field-emission gun, scanning-electron microscope equipped with a TSL OIM™ EBSD system. Depending on the particular microstructure, the EBSD scan step size ranged from 0.1 to 5 μm. To improve the reliability of the EBSD data, small grains comprising three or fewer pixels were automatically removed from the maps using the grain-dilation option in the TSL software in the TSL software. Furthermore, to eliminate spurious boundaries caused by orientation noise, a lower limit boundary-misorientation cutoff of 2° was used. A 15° criterion was used to differentiate low-angle boundaries (LABs) and high-angle boundaries (HABs).

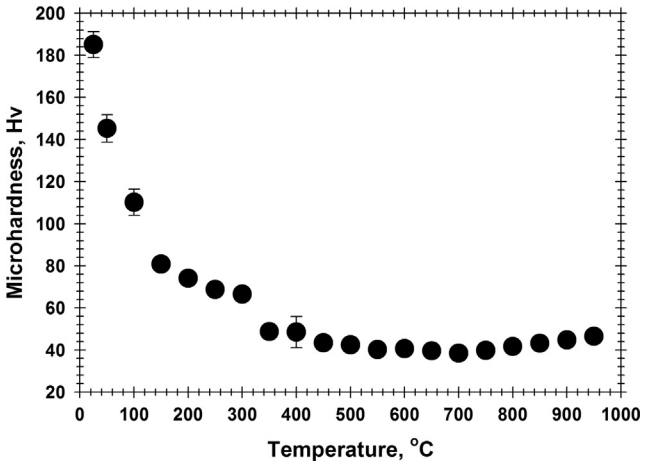

Fig. 1. Effect of annealing temperature on microhardness. Error bars show standard deviation of the measurements.

Because the microstructures developed during large deformation are frequently characterized by a complex mixture of LABs and HABs, there is often confusion regarding the definition of grains. To avoid ambiguity, the term "grain" in the present work refers to a crystallite bordered by a continuous boundary having a misorientation of greater than 15°. For the cryo-rolled and partially-recrystallized microstructures at temperatures of 150 °C and below, the grain size was determined by the linear intercept method along the ND (i.e., "grain thickness"). At higher annealing temperatures, essentially equiaxed grain structures had developed, and the grain size in these cases was quantified by measurement of the grain area (ignoring annealing twin boundaries) and calculation of the circle-equivalent diameter (i.e., the so-called grain-reconstruction method [15]).

To obtain a broader view of underlying microstructure changes, the Vickers microhardness was also measured on each sample using a load of 50 g for 10 s. At least 10 measurements were made in each case to obtain an average value.

3. Results

3.1. General trends of microstructure evolution

The broad aspects of the annealing behavior in terms of the evolution of microhardness, microstructure, and texture during *isochronal* annealing in the range of 50–950 °C are summarized in this section.

3.1.1. Microhardness

The effect of annealing temperature on microhardness is illustrated in Fig. 1. Four different temperature regimes were noted as follows:

(i) At temperatures below 150 °C, the microhardness decreased rapidly with increasing temperature.
(ii) In the temperature range of 150–300 °C, the hardness tended to saturate.