Scripta METALLURGICA et MATERIALIA
Vol. 24, pp. 1303-1306, 1990
Printed in the U.S.A.

PREPARATION OF SOME SPECIFIC GRAIN BOUNDARIES IN ALUMINUM FOR HREM STUDIES BY COLD ROLLING AND ANNEALING

M. Shamsuzzoha and P.A. Deymier
Department of Materials Science and Engineering
University of Arizona, Tucson, AZ 85721

(Received April 2, 1990)
(Revised May 3, 1990)

Introduction

The preparation of bicrystals with an aim to study grain boundary structure by high resolution electron microscopy (HREM) requires a careful approach. Primary attention needs to be directed towards the orientations of the crystals forming a boundary so that atomic resolution images of the bicrystal can be achieved by HREM. Mader, Necker and Balluffi (1) have reviewed the limitations of present day high resolution microscopes in resolving grain boundary structures. According to these authors, the resolution limit (information resolution limit = 0.14 nm) of such microscopes now allows most f.c.c metallic crystals to reveal a projected point pattern along any of the <100>, <110>, <111> and <112> directions. Resolving a grain boundary structure requires simultaneous imaging of atomic columns in crystals on both sides of the boundary. For a very thin f.c.c. metallic bicrystal, a 1° or less misorientation between neighboring crystals is required to achieve a simultaneous imaging of atomic columns across the interface (2). The {200}, {220} and {111} projected line pattern of a f.c.c. metallic crystal can also be obtained with current microscopes. A misorientation of 1° to 3° between such planes of f.c.c. metallic bicrystal can provide atomic plane resolution across the interface.

Bicrystals meeting these criteria for HREM studies are usually prepared either by disposition of thin film on a suitable substrate (3) or by solidification of a required orientation seeded bicrystal either by the Bridgeman technique (4) or by the Czochralski technique (5). These techniques are rather expensive and require sophisticated instrumentations. In the present work we investigated a relatively simple and inexpensive cross-rolling and annealing technique to obtain bicrystals of aluminum for HREM studies of grain boundary structures. In this paper we give a report of such preparation techniques for certain specific aluminum bicrystals, which were found suitable for HREM imaging.

Experimental Procedures

The starting material consists of polycrystalline aluminum (with 99.999% stated purity) cylindrical bars of 10 mm in diameter by 50 mm in length. The bars were longitudinally strained to 1% reduction and then annealed at 600°C for one hour. The annealed bars typically revealed randomly oriented centimeter size grains. Single crystal slices of 15 mm length were cut from the annealed bars and then subjected to equal step pass cross-rolling to obtain ten different thicknesses with a total reduction ranging from 90 to 99%. The pass steps used were varied between 1.25 to 20% of the initial thickness. Heavily deformed specimens were given either a single cycle or a two cycle annealing. The single cycle annealing was performed at three different temperature ranges: (1) between 125 to 225°C, (2) between 250 to 350°C and (3) between 400 to 600°C. The two cycle annealing was comprised of a primary recrystallization annealing (at temperatures ranging either between 125 to 225°C or between 250 to 350°C) and a respective secondary recrystallization annealing (at temperatures ranging either between 250 to 350° or between 400 to 500°C).

For thin foil preparation, 3 mm diameter discs were spark-cut from the annealed samples and then electropolished at a voltage of 50 V at room temperature in a solution of 73% methanol, 25% nitric acid and 2% hydrochloric acid. The thin foil thus prepared was examined with 120 kV Hitachi and 400 kV JEM 4000EX electron microscopes. A minimum of 30 grain boundaries selected at random for each cold rolling and annealing

0036-9748/90 $3.00 + .00

condition were analyzed by TEM. In the electron microscopes, boundaries with neighboring crystals exhibiting any one of the <100>, <110>, <111> and <112> diffraction patterns were selected. Using the goniometer tilt in the microscope, corresponding Kikuchi line patterns in each crystal across such selected boundaries were centered; relative misorientation between crystals forming boundaries was measured by a method described elsewhere (6).

Results and Discussion

For all cold rolling and heat treatment conditions, the grains in the resulting specimens are generally found to be equiaxed and extending diametrically from micrometer to millimeter in size. Almost all crystals were found to exhibit either (100) or (110) texture, but with a wide variety of misorientations. A notable exception to this is the rare occurrence of (112) texture.

The analyses of all TEM data revealed that only those specimens which were reduced in the range of 94 to 96% of the initial thickness possess boundaries suitable for HREM studies. The suitable grain boundaries observed in this optimal range are:

A. $\Sigma 3$ twin boundary: Twin $\Sigma 3$ grain boundaries were observed in specimens produced with a large rolling pass step $\geq 7.5\%$ and a single cycle annealing at temperatures ranging between 400 to 600°C. The straight twin boundaries were readily recognized within the specimens containing mainly curved and ill defined boundaries. Approximately 5% of the existing boundaries were found to be twin boundaries. Figure 1 shows a composite mean diffraction pattern taken on a $\Sigma 3[\bar{1}10]/(111)$ bicrystal with perfect alignment of their <110> poles as well as a HRTEM bright field image of the twin boundary. HREM images of such twin boundaries are reported elsewhere (7).

B. Low angle <100> tilt boundaries: Low angle <100> tilt boundaries with tilt angle ranging between 0° and 10° were observed in specimens produced with a low rolling pass step ranging between 1.25 to 5% and two cycles of heat treatment. The heat treatment consists of a primary recrystallization annealing for one hour at a temperature ranging between 125 to 225°C, followed by a secondary recrystallization at a higher temperature ranging between 250 to 350°C for an additional hour. Analysis of a random set of 200 boundaries indicated that approximately 25% meet the projected line resolution criterion for HREM with only 5% meeting the projected point resolution criterion. An example of such a boundary is shown in Fig. 2. The 6° [001] tilt grain boundary meets the atomic resolution criteria and its structure has been reported elsewhere (8).

C. <100> 45° twist grain boundary: This grain boundary was observed in specimens produced under most of the studied conditions in which a two cycle heat treatment was applied. However, its occurrence is significantly increased in those specimens which were produced with a rolling pass step ranging between 5 to 7.5% and low temperature two cycle heat treatment. A random analysis on some 200 specimens revealed a 5% occupancy for such boundaries. Most of the studied [100] 45° twist boundaries were found to exhibit an additional tilt character with a rotation (rotation angle ranges between 0 and 17.5°) parallel to the common <100> and <110>. The composite diffraction pattern of a [001] 45° twist plus 17.5° tilt grain boundary presented in Fig. 3 meets the criterion of atomic resolution by HREM. HREM images of this grain boundary structure have been obtained and reported in reference (8).

Conclusion

In this paper we are reporting a method for the production of some specific grain boundaries in aluminum, whose atomic structure can be imaged with current high resolution microscopes. The method consists of a simple cross rolling of aluminum single crystals of arbitrary orientation followed by a heat treatment. By careful control of the rolling conditions and annealing temperatures, the probability of occurrence of well-defined perfect grain boundaries can be raised to a level sufficient for random selection of specimen. The probability of obtaining a perfect $\Sigma 3$ twin, a perfect <100> low angle tilt or a perfect <100> 45° twist plus tilt is found

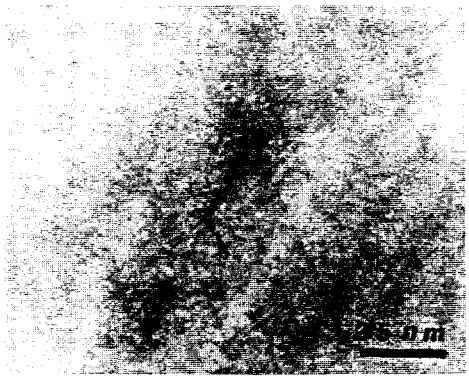
to be approximately 5%, which makes the method economically viable for the production of such grain boundaries suitable for HREM studies.

Acknowledgements

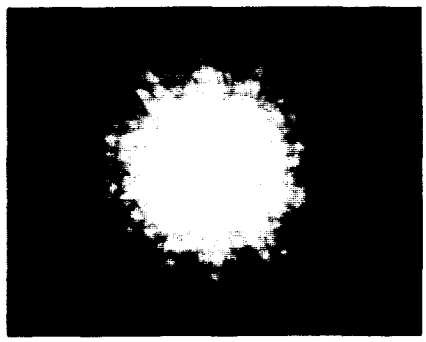
This research was supported in part by the Facility for High Resolution Electron Microscopy in the Center for Solid State Science at Arizona State University, established with support from the National Science Foundation (Grant No. DMR-86-11609). The authors acknowledge the receipt of financial support from the U.S. Department of Energy under Contract No. DE-FR02-87ER45285. The authors also express their thanks to Dr. David J. Smith of the Center for Solid State Science at Arizona State University for his assistance in high resolution microscopy.

References

1. W. Mader, G. Necker, and R. W. Balluffi, J. de Physique, C5-49, 233 (1988).
2. J. M. Penisson, J. de Physique, C5-49, 87 (1988).
3. T. Schober and R. W. Balluffi, Phil. Mag., 20, 511 (1969).
4. B. Chalmers, Canadian Journal of Physics, 31, 132 (1953).
5. J. Czochralski, Z. Phys. Chem., 92, 92 (1918).
6. J. I. Bennetch, in: Proc. 46th Annual EMSA Meeting, 1988, p. 830.
7. M. Shamsuzzoha, P. A. Deymier, and D. J. Smith, Phil Mag. submitted (1989).
8. M. Shamsuzzoha, P. A. Deymier, and D. J. Smith, submitted to 12th International Congress for Electron Microscopy (1990).



a



b

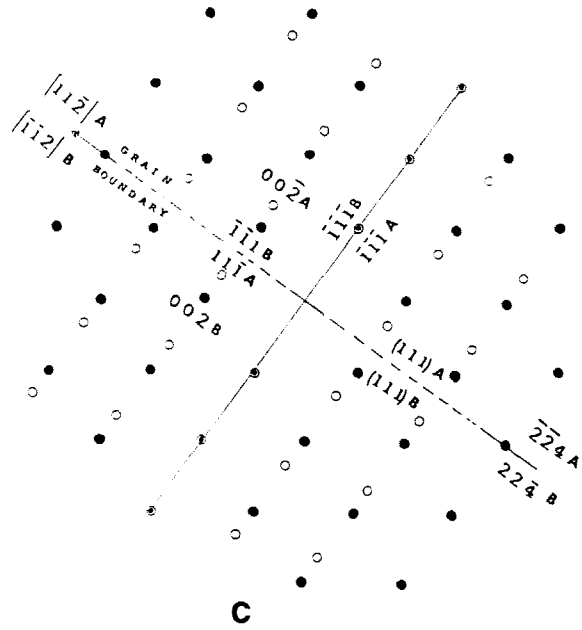
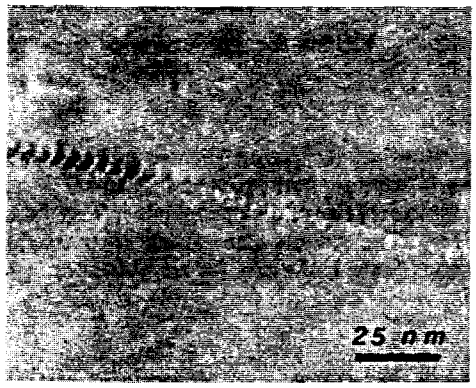
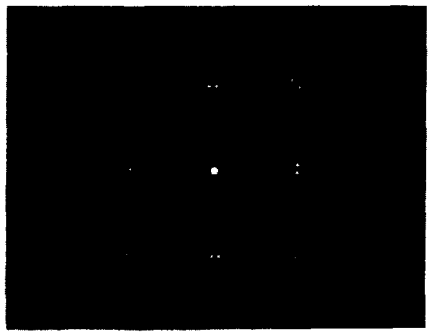


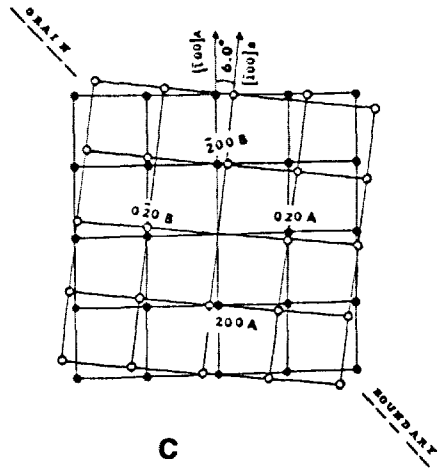
FIG. 1. (a) Bright field HRTEM image of a Σ3 twin boundary with incident beam parallel to {111} twinning plane. (b) Corresponding selected area composite microdiffraction pattern. (c) Indexing for bicrystal diffraction pattern.



a

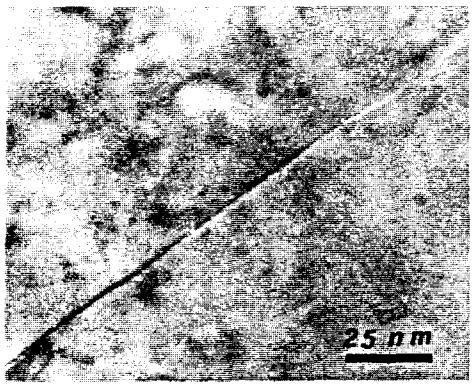


b

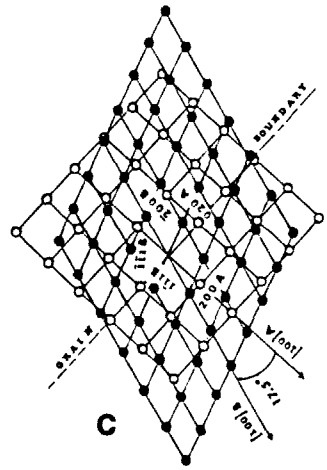


c

FIG. 2. (a) Bright field HRTEM image of a 6° [001] tilt boundary with incident beam parallel to {100} of each crystal across the boundary. (b) Corresponding selected area composite diffraction pattern. (c) Indexing of bicrystal diffraction pattern. The bicrystal misalignment of the common (001) planes is approximately 0.1°.

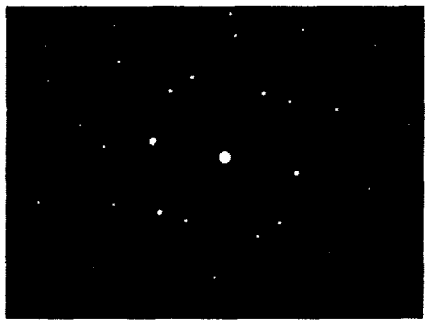


a



c

FIG. 3. (a) Bright field HRTEM image of a [001] 45° tilt plus 17.5° tilt boundary with incident beam parallel to {100} of crystal A and {110} of crystal B. (b) Corresponding selected area composite diffraction pattern. (c) Indexing for bicrystal diffraction pattern. The common (001) and ($\bar{1}$10) planes are misaligned by 0.2°.



b