Effect of Grain Boundary Structure on Misorientation Change of Pure Copper Bicrystals Pressed by One-Pass Equal-Channel Angular Pressing

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Following a previous paper, (Materials Transactions, Vol. 53 (2012), p. 1858), which reports on the dynamic misorientation change absorbed by the grain boundaries themselves during one-pass equal-channel angular pressing (ECAP), the effect of grain boundary structure on the misorientation change during ECAP was examined as the next step. Two bicrystals with a Σ3[111] grain boundary and a random one were subjected to ECAP for one pass, and compared. Both the bicrystals were designed to have a similar slip pattern where slip activity was concentrated on one slip plane. Direct evidence of misorientation changes absorbed by grain boundary themselves was obtained again, and its marked dependency on grain boundary structure became evident. Namely, in Σ3 grain boundary, misorientation was partially carried by the grain interior in its vicinity, which stems from the dislocation accumulation. On the other hand, in the random grain boundary, most of the misorientation was carried by the boundary itself. These differences are discussed in terms of the capability as a sink of lattice dislocations during severe plastic deformation. [doi:10.2320/matertrans.M2013076]

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1. Introduction

Severe plastic deformation (SPD) is now widely acknowledged as a process to fabricate bulk-nanocrystalline metallic materials,1) and the mechanism of dynamic formation of grain boundaries and grain fragmentation into nano- or submicron regions during SPD has been of primary concern.2-4) These reports seem to support, explicitly or implicitly, the grain subdivision as the grain refining mechanism, where the subboundaries emerge inside the grains and increase their misorientation with increasing strain.5) During SPD processing, grain boundaries (GBs) are believed to absorb lattice dislocations and change their misorientation at room temperature, which has been widely demonstrated at intermediate to high temperatures under moderate deformation such as tensile straining.

Misorientation change of GBs accompanied by the evolution of a UFG structure has been mainly evaluated using diffraction images of transmission electron microscopy (TEM) by observing an approximate tendency from the discrete collection of spots toward continuously distributed rings.4,6) The recent advent of high resolution electron backscattered diffraction (EBSD) provides the capability to obtain more quantitative information concerning distributional change of misorientation angles with an increase of strain.7) However, the question of how, and to what extent a grain boundary can change its misorientation remains to be addressed since clarifying the question requires tracking of a specified grain boundary with well-defined characteristics and this is technically difficult in studies using polycrystalline metals. Bicrystals with a well-defined grain boundary offer a very effective approach in this respect. In a previous study,8) we pressed pure copper bicrystals for one pass by ECAP, and obtained direct evidence of dynamic misorientation change of the GB by as high as 15° during ECAP. This change shows a marked dependency on slip geometry in terms of the slip plane of ECAP.

The capacity of GBs as a sink of lattice dislocations depends on GB structure, namely, high-energy GBs with incoherent structures have a high capacity to absorb dislocations and high tendency to misorientation change.9) We examined the effect of grain boundary structure on the dynamic misorientation change during ECAP using two bicrystals with different GB structures but with similar slip geometries.

2. Experimental Procedures

Copper bicrystals of commercial purity were grown by the Bridgeman method. Crystallographic orientation was controlled so that the GB plane became parallel to (111) with a common (112) direction on the plane. One bicrystal has a deviation of 0.2° from the ideal Σ3 relationship, and the other has 12.3°, thus the GB of the former was determined as the Σ3 boundary and that of the latter as random GB according to Brandon’s criterion.10) Billets with square cross section for ECAP were cut from the bicrystal by a spark-erosion machine so that the GB was inclined by 45° in relation to the longitudinal axis. Stereographic projections of the initial orientation and the geometrical relationship among the GB plane, the shear plane (SP), the shear direction (SD) of ECAP and the primary slip plane (PSP) is shown in Fig. 1. Definitions of SP and SD in terms of inserting and extruding directions of ECAP are shown in Fig. 2. The slip distribution among the 12 slip systems were determined by the shear factor,11) expressed by cosαβ, where α is the angle between the shear plane normal and a slip plane normal, and β is the angle between the shear direction and a slip direction as shown in Fig. 3. The shear factor of 12 slip systems in both the component crystals in Σ3 and random bicrystals are summarized in Table 1. As shown in the table, the shear factors of two slip systems in a slip plane are considerably higher than those of the other slip systems in both the

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bicrystals. Thus, we can assume the slip is concentrated in the slip plane. ECAP was conducted at a pressing speed of 10 mm/s at room temperature using MoS2 lubricant. After ECAP for one pass, microstructures were examined by an optical microscope, and field-emission scanning electron microscope (FE-SEM, JEOL JSM 7000F) equipped with EBSD (Oxford Instruments). The orientation image maps by EBSD were obtained by 0.1 µm step.

3. Experimental Results

3.1 Macroscopic morphology after one-pass ECAP

Optical micrographs and orientation image maps obtained by EBSD are shown in Fig. 4. In both the bicrystals, the GBs were rotated toward the ED to an inclination of 20° as

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**Fig. 1** Stereographic projections of initial crystallographic orientation in the component crystals with (a) random and (b) Σ3 grain boundaries. The upper and lower crystals are termed crystals 1 and 2, respectively.

**Fig. 2** Schematic diagram of ECAP and definition of coordinates. SP and SD represent the normal directions of the shear plane and the shear direction, respectively.

**Fig. 3** Geometric relationship between the shear plane normal, the shear direction, a slip plane normal and a slip direction. Shear factor is defined as \( \cos \alpha \cos \beta \).
shown in Figs. 4(a) and 4(d). This GB inclination is consistent with the material flow rule as shown in Fig. 5.12) In Fig. 5(a), the points A and B flow to A via M, and N, following the rule, AM + MA = BN + NB′. Thus, the grain boundary with initial 45° inclination should be changed to 18°. In both the bicrystals, inhomogeneous deformation structures, such as deformation bands or shear bands that may also carry misorientation, were not observed in meso- and micro-scale orientation image maps as shown in Figs. 4(b), 4(c) and 4(e), 4(f). In the smaller micro-scale, both the grain boundaries are wavy, and suggest dislocation accumulation near the GBs, leaving the GB in a non-equilibrium state. The Σ3 GB, particularly, shows an obviously wavy morphology, suggesting higher dislocation accumulation in its vicinity. This difference will be discussed with regards to the interaction between lattice dislocations and GBs in the discussion section. In Fig. 6, {100} pole-figures indicate the orientations of crystals 1 and 2 in the immediate vicinity of the GB before and after ECAP. In the random GB, crystal 2 rotated about 35° whereas crystal 1 rotated about 15° around the TD axis, resulting in a decrease of misorientation by 20° between the two crystals. In Σ3 GB, crystals 1 and 2 rotated about 22 and 30° around the TD axis resulting in a decrease by 8°. Since inhomogeneous structures, such as deformation bands or shear bands that may carry large misorientation, were not observed in EBSD as shown in Fig. 4, most misorientation change was indeed absorbed by the GBs themselves in both the bicrystals.
3.2 Misorientation distribution near grain boundaries

The line profiles of orientation change across the GB at the several points were obtained by EBSD along the GBs before and after ECAP. Figure 7 shows an orientation map and the corresponding orientation change across the Σ3 GB before ECAP. There is essentially no orientation change in the vicinity of the GB. Note that the steep gradients on the GBs are the result of interpolating the nearest two points across the GBs, and are not attributed to the dislocations. Figure 8 shows the orientation change after ECAP. The length and direction of line profiles A–D correspond to those of arrows A–D in the orientation maps in Figs. 8(a) and 8(b). The misorientation between the two component crystals was mostly accommodated by the GBs in both the random and Σ3 boundaries. In particular, in the random GB, there is little orientation change near the GB, and most misorientation change is carried by the GB itself (Fig. 8(c)). However, in Σ3 boundaries, misorientation change is observed in its vicinity; there is a positive change in crystal 2 while there is a negative change in crystal 1 (Fig. 8(d)). These misorientation distributions stem from accumulation of the lattice dislocations in the vicinity of GB, and suggest that higher density of dislocations accumulated near the Σ3 GBs. It becomes clear again by comparing with the result before ECAP in Fig. 7 that the steep gradients on the GBs are the result of interpolating the nearest two points across the GBs, and are not attributed to the dislocation accumulation. Considering that both the bicrystals have similar slip geometries and, hence, the similar slip patterns, it is reasonable to consider that the number of impinged lattice dislocations during ECAP is of little difference, and therefore the random GB has absorbed more lattice dislocations, resulting in low dislocation accumulation in its vicinity. This difference and associated crystal rotations are illustrated in Figs. 8(e) and 8(f). Since the GBs were sheared nearly in a perpendicular direction, the dislocations of positive and negative edge components should impinge on the GB from two neighboring crystals. The crystal rotation is assumed as a simple rotation around the axis perpendicular to the paper, and the counter-clockwise rotation is defined as the positive direction. The misorientation is defined as 0 at an arbitrary location. With the increasing distance x from the arbitrary position in crystal 2, the misorientation increases due to a positive rotation by the pile-up of positive edge dislocations in crystal 2, whereas it decreases due to a negative rotation by the pile-up of negative edge dislocations in crystal 1.

4. Discussion

It is known that a dislocation which impinges on a GB can interact with it in three different ways. First, a dislocation can cross the GB and slip into the neighboring grain if the GB is a small angle boundary with misorientation smaller than 10°–15°. It was once considered that the coherent Σ3 twin boundary does not act as an obstacle to impinging dislocation, and the question of whether it should be counted to estimate the average grain size in the Hall-Petch relation has been discussed. The Σ3 GB becomes transparent to dislocations in the very limited case where the primary slip plane is parallel to the boundary. Second, dislocations form a pile-up array and local stress concentration. This is the most prevalent case, and is the cause of Hall-Petch strengthening. Third, they are absorbed in the GBs. This tends to occur mainly in medium to high temperatures. In SPD processing, such as in the present study, a certain fraction of dislocations accumulated near the GBs was absorbed in the GB at room temperature.

It is known that GBs can change their misorientation by absorbing lattice dislocations at relatively high temperature.
under moderate straining.\textsuperscript{15) The lattice dislocations which impinge on the GB are trapped by the GB, and then dissociated into several GB dislocations making the GB reach a non-equilibrium state. These GB dislocations are dissipated and rearranged by climbing assisted by thermal activation, recovering the equilibrium GB structure.\textsuperscript{16) These reaction processes between lattice dislocations and GBs were directly observed by a transmission electron microscope.\textsuperscript{17} These so-called ‘extrinsic’ GB dislocations have a Burgers vector defined by the displacement shift complete (DSC) lattice as the intrinsic dislocations\textsuperscript{25,26) and the dissociation is expressed as follows:\textsuperscript{25)  

\[
b_{\text{latt}} = \sum_{i=1}^{n} b_{\text{DSC}}^{i}
\]  

where \(b_{\text{latt}}\) and \(b_{\text{DSC}}^{i}\) are the Burgers vectors of lattice dislocations and GB dislocations, respectively. The DSC lattice is the reciprocal lattice of CSL, and thus, \(b_{\text{DSC}}^{i}\) is inversely proportional to the \(\Sigma\)-value.\textsuperscript{27–29} With increasing \(\Sigma\)-value, lattice dislocations are dissociated into increasing number of GB dislocations.\textsuperscript{25) For example, a lattice dislocation is dissociated into three GB dislocations in a \(\Sigma 3\) GB, whereas it is considered to be dissociated into nearly 100 dislocations in a random GB.\textsuperscript{30}) Assuming that the dislocation energy is proportional to the square of the Burgers vector, energy reduction associated with the dissociation of a lattice dislocation is expressed as follows:\textsuperscript{31)  

\[
\Delta E_{\text{dis}} = A \left[ (b_{\text{latt}}^2 - \sum_{i=1}^{n} (b_{\text{DSC}}^{i})^2) \right]
\]  

Hence, the driving force for the dissociation of an impinged dislocation is higher in high-\(\Sigma\) GB. As shown in Fig. 8, there are misorientation distributions stemming from dislocation accumulation in the vicinity of \(\Sigma 3\) GB, whereas it was negligibly small in the random GB.

The above discussion concerns the tendency for dislocation dissociation into the GBs. We have to consider the other aspect of the GBs as a sink of dislocations, namely, the capacity for extrinsic GB dislocations. This can be
determined by the ability of the GBs to dissipate and consume the extrinsic GB dislocations by dynamic recovery. The GBs that absorbed extrinsic dislocations were in the non-equilibrium state, and this ability can be estimated by the time required to return to the equilibrium state by thermal activation as is expressed below. Namely, the capacity can be considered to be higher with a shorter recovery time.

\[ t_s = 0.036 \frac{kT_0 s^3}{\mu D^J \delta} \exp \left( \frac{Q}{RT_s} \right) \]

Where \( t_s \) is the time for complete relaxation, \( k \) is the Boltzmann constant, \( T_0 \) is temperature, \( s \) is dislocation interspace in the final structure, \( \mu \) is the shear modulus, \( D^J \) is the grain boundary diffusion coefficient, \( \delta \) is the grain boundary width, \( Q \) is the activation energy for diffusion. Since the \( D^J \) and \( Q \) differ in the random and \( \Sigma 3 \) GBs, relaxation time should be different. Qualitatively, extrinsic dislocations can dissipate faster in the random GBs and continue to be absorbed.\(^{34,35}\) Priester estimated the relaxation time for an average grain boundary of high purity copper (99.9998%) at room temperature to return to a complete equilibrium state to be approximately 6h.\(^{36}\) Therefore, in order for most GBs with CSL and random structure in copper to keep acting as a sink of dislocations, the saturation of the ever-increasing GB dislocations during SPD must be realized by redistribution and reduction of extrinsic grain boundary dislocations through grain rotation and misorientation change.

5. Conclusions

Two types of bicrystals with different grain boundary structures, namely, a random and \( \Sigma 3 \{111\} \) grain boundaries, were pressed by ECAP for one pass. Both the bicrystals have similar slip geometries and slip patterns in terms of ECAP, and the effect of grain boundary structure on the deformation and misorientation change was examined. As a result, both the bicrystals exhibited misorientation change of about 15° in the random and \( \Sigma 3 \) grain boundaries, respectively, during one-pass ECAP. Most misorientation change was absorbed by the grain boundaries themselves, and not by deformation bands or shear bands in the vicinity. The difference of the misorientation change may stem from the grain boundary structure, but the effect of slip pattern cannot be neglected. Most importantly, both the grain boundaries showed marked difference in misorientation distribution in the vicinity, which stemmed from the difference in the capacity as a dislocation sink.

REFERENCES

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