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Influence of stacking fault energy on the minimum grain size achieved in severe plastic deformation

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Abstract

Samples of pure Cu and a Cu–10% Zn alloy were processed by high-pressure torsion and by high-pressure torsion followed by cold-rolling to a reduction of ~75%. The grain sizes in these two conditions were measured by transmission electron microscopy and by X-ray diffraction. The experimental results show the average grain size and the width of the grain size distribution are both smaller in the Cu–10% Zn alloy by comparison with pure Cu. This difference is due to the lower stacking fault energy of the Cu–10% Zn alloy. An analysis shows all of the experimental results are consistent with a theoretical model predicting the minimum grain size produced by milling.

Keywords: Copper alloys; Grain size distributions; Milling; Severe plastic deformation; Stacking fault energy

1. Introduction

The stacking fault energy, γ, plays an important role in determining the mechanical properties of face-centered cubic (fcc) metals. In practice, the width of the stacking fault ribbon between the two Shockley partials in crystalline solids is a significant feature controlling the rates of dislocation cross-slip and climb. A review of the literature shows that the importance of γ was recognized in a very early and extensive review of high temperature creep co-authored by Professor Amiya K. Mukherjee. Thus, in a classic review, presented at a conference in Haifa in 1969, Bird et al. [1] analyzed the available creep data for a wide range of fcc metals, they showed the value of the stress exponent, n, varied for different metals within the range from ~4.4 to ~7.0, and they further demonstrated the measured values of n increased with the dimensionless quantity Gb/γ where G is the shear modulus and b is the magnitude of the Burgers vector for each separate metal. Later, an alternative procedure was developed based on this earlier approach and it was shown that the normalized creep rate ($\dot{\varepsilon}kT/DGb$) varies with $(\gamma/Gb)^3$ for a range of fcc metals within the dislocation climb regime, where $\dot{\varepsilon}$ is the creep rate, k the Boltzmann's constant, T the absolute temperature and D is the coefficient for self-diffusion [2].

Following this early work on creep, Professor Mukherjee made important contributions in several areas of research including in superplasticity [3,4], in the superplastic deformation of ultrafine-grained materials [5] and, most recently, in the deformation processes occurring in nanocrystalline solids modeled using molecular-dynamics simulation [6–8]. Based on a consideration of these various contributions over a period of many years, it is appropriate that the present paper, honoring Professor Mukherjee in this special symposium, should be an evaluation of the significance of the stacking-fault energy in determining the minimum grain size attained in metals processed using severe plastic deformation (SPD).

Processing by SPD is now a well-established procedure for achieving remarkable grain refinement in polycrystalline metals [9,10]. In the present investigation, two different metals, copper and bronze (Cu–10 wt.% Zn), were selected as model materials because they have the same fcc single-phase crystal structure but significantly different stacking fault energies of 78 [11,12] and 35 mJ m^{-2} [11,13], respectively. Samples of these two materials were subjected to SPD processing using high-pressure torsion (HPT) and the grain sizes were measured both after HPT and

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after HPT followed by cold-rolling (CR). The variation of the grain size with the SFE was compared directly with a recent theoretical model of Mohamed [14] describing the minimum grain size obtainable by conventional ball milling operations. As will be demonstrated, the experimental data show good agreement with the predictions of the theoretical model.

2. Experimental materials and procedures

The experiments were conducted using commercial purity copper (99.9%) and bronze (Cu–10 wt.% Zn). The Cu was received as a rod with a diameter of 10 mm and the bronze was received as a plate with a thickness of 6 mm. Both materials were sliced into disks with thicknesses of 0.8 mm and diameters of 10 mm. These disks were processed by HPT using a conventional processing facility [15]. Each disk was subjected to a total of five revolutions at room temperature using a rotation speed of 1 rotation/min with an imposed pressure of 6 GPa. It is reasonable to anticipate, based on an earlier report for pure Cu processed by HPT [16], that this processing condition is sufficient to achieve a reasonably steady-state microstructure. For simplicity, and following a procedure suggested earlier [17], the strain is reported in terms of the numbers of revolutions imposed on the sample but in practice the strain across the disk may be estimated using conventional relationships [18] or finite element procedures [19]. For the processed condition after HPT without any additional working, these disks are designated the HPT samples.

To provide information on the effect of combining HPT with subsequent cold-rolling, some of the disks were cold-rolled into thin ribbons with thicknesses of ∼200 μm. The total thickness reduction for these samples was ∼75% after multiple rolling passes with a thickness reduction of ∼10% imposed in each separate pass. These specimens are designated the HPT + CR samples.

Following HPT, the microstructures of the specimens were examined using transmission electron microscopy (TEM) and X-ray diffraction (XRD).

For inspection by TEM, the specimens were thinned to a thickness of ∼10 μm by mechanically grinding both sides of the disks using diamond-lapping films in the order of 30, 6, 1 and 0.1 μm. Final thinning to a thickness suitable for electron transparency was performed using a Gatan Dual Ion Milling System with an Ar^+ accelerating voltage of 4 kV and liquid nitrogen for specimen cooling. Samples were inspected using a Philips CM30 microscope operating at 300 kV and all observations were made using top views of the disks and rolled ribbons. Individual grain sizes were recorded for approximately 300 grains in each sample using the linear intercept method. It is important to note that, depending on the experimental conditions, the grain sizes measured in samples processed by HPT may vary across the diameter of the disk [20–24]. In the present investigation, the HPT samples were examined near the edges of the disks where the average grain sizes may be smaller than in the whole disk. For the HPT + CR samples, the TEM observations were made in the centers of the rolled ribbons where the grain sizes may be larger than the average values.

Quantitative X-ray diffraction was performed using a Scintag X-ray diffractometer equipped with a Cu target operating at 1.8 kW. Pure Al powder (99.999%) was annealed at 200 °C in Ar and used as an XRD peak-broadening reference for estimates of the grain sizes. The XRD measured peaks were fitted using a Pearson VII function. For the HPT sample, the reported XRD values of the grain sizes represent average values obtained from the cross-section of the whole disk after processing by HPT and for the HPT + CR sample the XRD results represent microstructural information obtained along the cross-sectional directions of the HPT + CR processed ribbon.

3. Experimental results

Inspection of the HPT samples by TEM showed that both the Cu and Cu–10% Zn disks contained a reasonably equiaxed distribution of grains having random orientations. Examples of the microstructures for these two materials are shown in Fig. 1 where it is apparent that the grains are well defined in both metals. Some very limited twinning was visible in the Cu–10% Zn alloy and an example of twinning is marked in Fig. 1(b).

Fig. 2 shows the statistical grain size distributions recorded in these two materials after HPT using measurements taken from approximately 300 individual grains in each sample. Two conclusions may be reached from inspection of Fig. 2. First, the average grain size, $\bar{d}$, is 75 nm in pure Cu and 50 nm in the Cu–10% Zn alloy thereby demonstrating there is a decrease in the average grain size produced by HPT with decreasing SFE. Second, the width of the grain size distribution is also smaller in the Cu–10% Zn sample so that in practice the microstructure becomes more uniform with decreasing SFE. The general appearance of these distributions are similar to those reported earlier for samples of high-purity nickel processed by HPT [25].

The microstructures after HPT + CR are shown for the two materials in Fig. 3 where again the grains are well defined and essentially equiaxed despite the addition of cold-rolling. Some very limited twinning was also again visible in the Cu–10% Zn alloy, as indicated in Fig. 3(b). The grain size distributions for the samples subjected to HPT + CR are given in Fig. 4. For these samples, the histograms show there are increases in both the average grain sizes and the widths of the distributions. For the HPT + CR condition, the average grain sizes were determined as 180 nm and 110 nm in Cu and Cu–10% Zn, respectively.

The average grain sizes were also determined using X-ray diffraction and the measured values were 84 nm in Cu and 54 nm in the Cu–10% Zn alloy after HPT and 70 nm in Cu and 50 nm in Cu–10% Zn after HPT + CR. These values also confirm that the average grain size decreases with decreasing SFE. However, the values obtained from XRD for the HPT + CR samples are significantly smaller than those obtained on the same two samples using TEM. There are two reasons for this discrepancy. First, the grain sizes measured by TEM are often larger than those measured by XRD because the X-ray measurements relate to the size of the cell or subgrain structure and are taken over the whole disk [26]. Second, the X-ray measurements reflect a bulk three-dimensional distribution of grains whereas the TEM observations for the HPT + CR sample were taken from observations

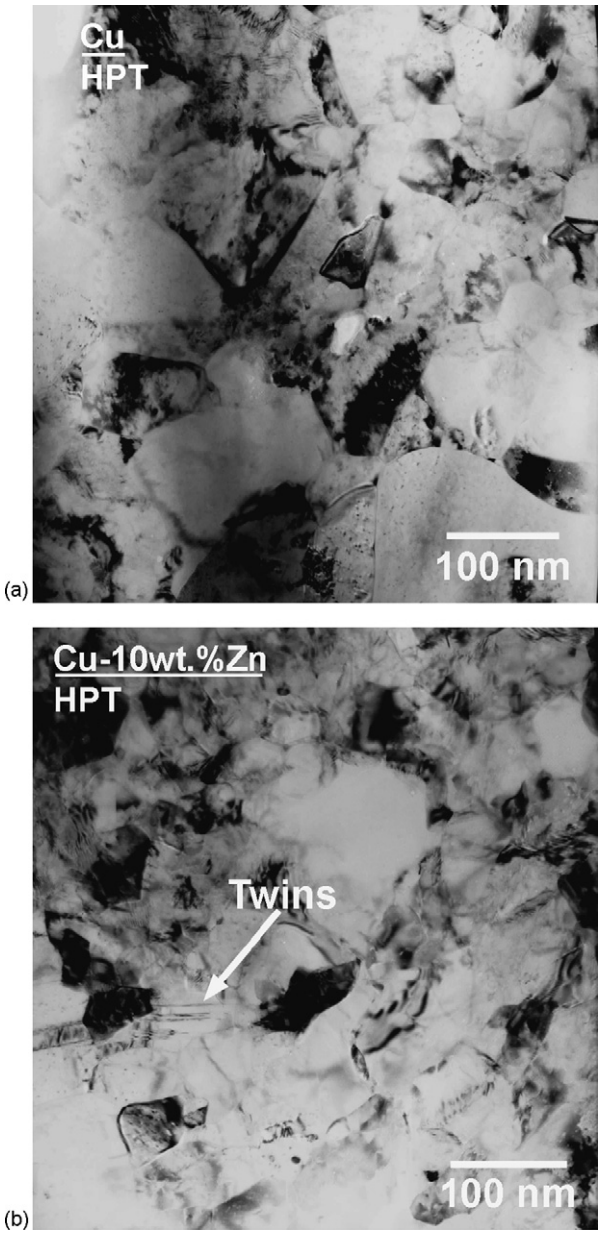

Fig. 1. Examples of the microstructures after HPT in: (a) Cu and (b) Cu–10% Zn.

in the plane of rolling. It is reasonable to anticipate the dimensions of the grains may be smaller when viewed perpendicular to the plane of rolling [27].

4. Discussion

The results from these measurements confirm the importance of the SFE. When the SFE decreases, as in the Cu–10% Zn alloy, grain refinement occurs more easily and the measured grain sizes are smaller. This is consistent with results on pure metals where the grain sizes produced in SPD processing are significantly larger in pure Al [28,29] than in pure Cu [30].

The grain sizes measured in this research are also exceptionally small by comparison with some other experiments on the SPD processing of Cu samples. For example, the average

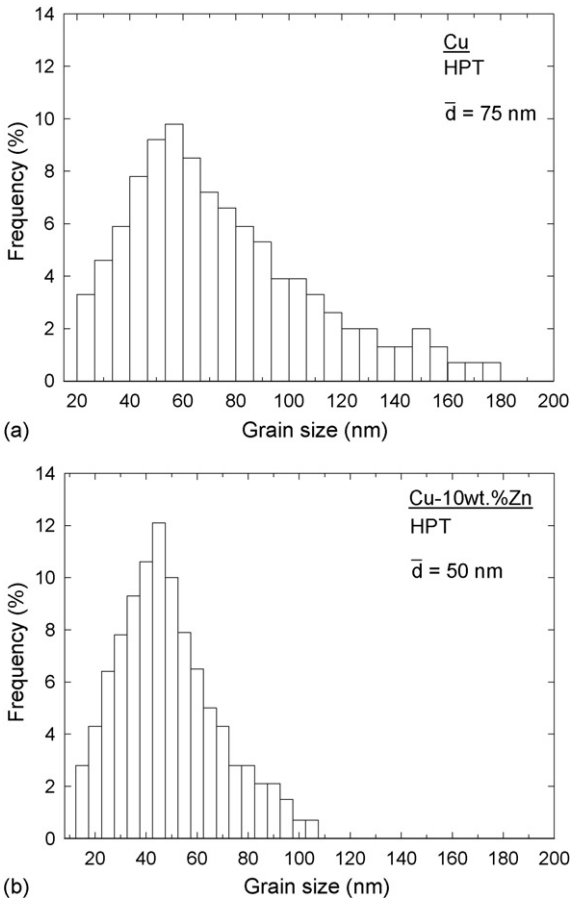

Fig. 2. Grain size distributions after HPT in: (a) Cu and (b) Cu–10% Zn.

grain size in pure (99.96%) Cu after processing by equal-channel angular pressing (ECAP) was measured by TEM as ∼270 nm [30] whereas the average grain size in the present experiments after HPT was ∼75 nm as shown in Fig. 2(a). This confirms the earlier observation that processing by HPT is more effective for producing grain refinement than processing by ECAP [20]. In addition, the average grain sizes measured in the present research are smaller than the values reported in an earlier study of pure (99.96%) Cu where, after five rotations at room temperature with an imposed pressure of 6 GPa, the grain sizes were recorded as ∼300 nm in the central region of the disk and ∼140 nm near the outer edge [31]. However, the present results are reasonably consistent with a study on high-purity (99.99%) Cu where, after HPT through five revolutions at room temperature using a slightly higher imposed pressure of 7 GPa, the grain sizes were measured as ∼100–200 nm near the center of the disk and ∼10–20 nm near the edge [32].

The effect of cold-rolling of the HPT samples is significant. A comparison of the grain size distributions in Figs. 2 and 4 shows that the introduction of CR after HPT broadens the distribution and increases the average grain size by a factor of slightly more than 2. However, these larger sizes are not reflected by the X-ray diffraction measurements where similar grain sizes were recorded after both HPT and HPT + CR. This difference is probably because the measurements for the HPT + CR samples were taken in the centers of the rolled ribbons. It is important to

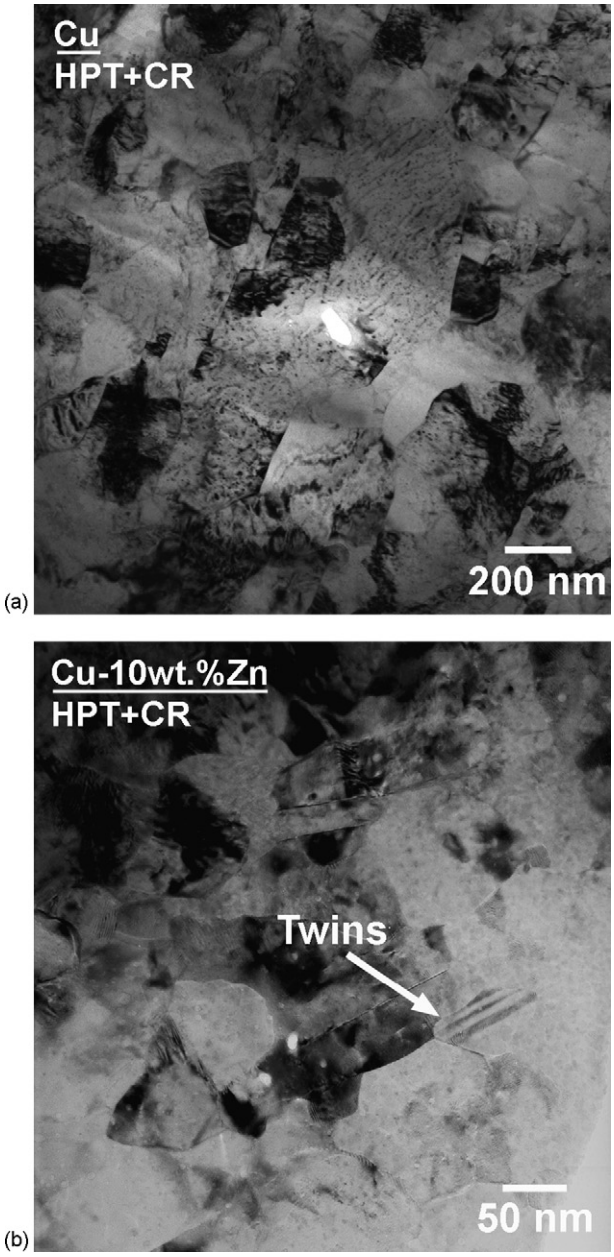

Fig. 3. Examples of the microstructures after HPT + CR in: (a) Cu and (b) Cu–10% Zn.

note that results obtained with an Al–Mg–Sc alloy after ECAP and ECAP + CR showed that the tensile properties, including the overall strains to failure, were essentially identical in both conditions [33].

The theoretical model developed by Mohamed [14] predicts the minimum grain size which may be achieved in the synthesis of bulk nanocrystalline materials by ball milling. Basically, the model characterizes the microstructure through the structural decomposition of a large-grained structure as a result of the imposition of severe plastic deformation. This means that the implications of the model should be equally applicable to other SPD processing routes including processing by HPT. Accordingly, it is worthwhile examining the precise implications of the model.

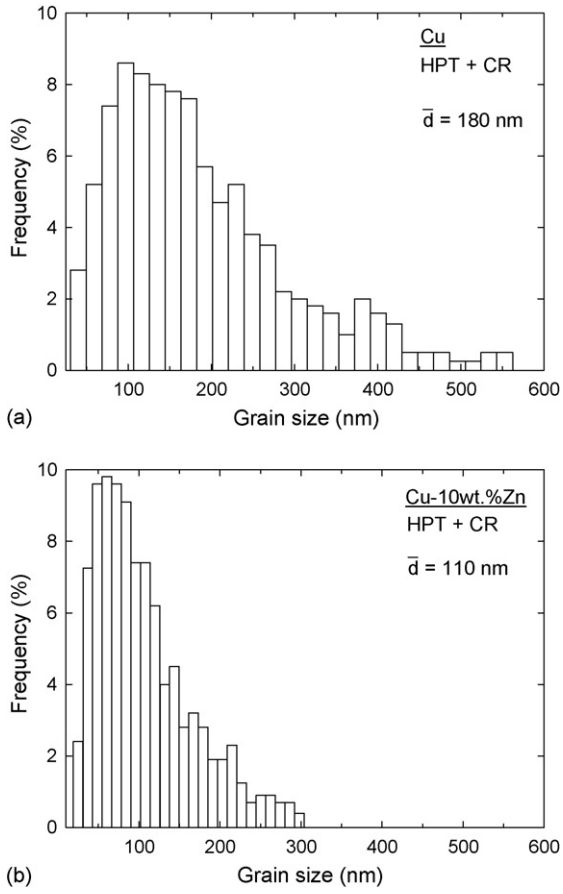

Fig. 4. Grain size distributions after HPT + CR in: (a) Cu and (b) Cu–10% Zn.

According to Mohamed [14], the measured minimum grain size, $d_{\min}$, is related to the SFE through an expression of the form

$$\frac{d_{\min}}{b} = A\left(\frac{\gamma}{Gb}\right)^{q} \tag{1}$$

where, according to the theory, the value of the exponent of the normalized SFE is $q = 0.5$ and A is a dimensionless constant. Taking extensive experimental data from ball milling for 11 different metals, Mohamed [14] showed that a logarithmic plot of $d_{\min}/b$ against γ/Gb yielded a straight line having a slope of $q \approx 0.65$ which is reasonably consistent with the theoretical prediction in Eq. (1).

To check on the validity of this approach in the present investigation, Fig. 5 shows a similar plot using the average values of the grain sizes obtained from the TEM and XRD measurements, d_{TEM} and d_{XRD}, for the two processing routes of HPT and HPT + CR. It is apparent that the present results, although based on data obtained for materials having only two different SFE, lead to exponents for the normalized SFE term within the range of ~0.53–0.64. These values of q are therefore similar to the earlier result reported by Mohamed [14] and they are also supportive of the theoretical model. It is reasonable to conclude from this analysis that the model of Mohamed [14] is applicable also in predicting the grain sizes in bulk solids fabricated using other SPD procedures.

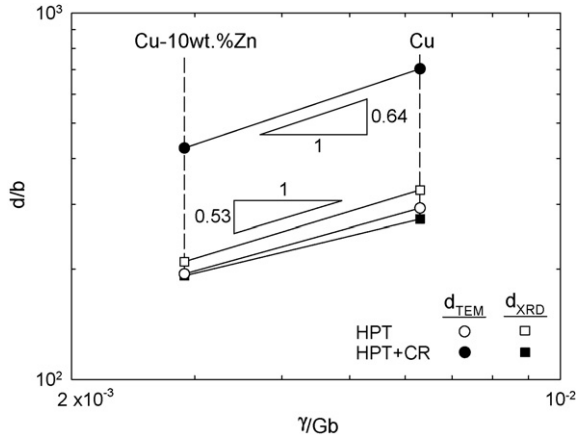

Fig. 5. Normalized grain size, d/b, plotted against the normalized stacking fault energy, γ/Gb, showing datum points obtained from TEM and XRD for the HPT and HPT + CR conditions.

Finally, it is necessary to inject a word of caution. An earlier study included work with a Cu–30% Zn alloy [34] where this material has a much lower SFE (14 mJ m^{-2} [11,13]) and the results showed a general lack of agreement with the model of Mohamed [14]. The failure to obtain agreement when incorporating the Cu–30% Zn alloy into the analysis probably reflects the very extensive twinning occurring in this material and consequently the inherent change that takes place in the deformation mechanism. By contrast, only limited lamellar twinning was noted earlier in samples of pure Cu and bronze [34] and this is confirmed by the present observations recorded in Figs. 1 and 3 using TEM. It is reasonable to conclude that, provided there is an absence of extensive twinning, there is generally good agreement between the experimental data and the theoretical model.

5. Summary and conclusions

(i) Disks of pure Cu and a Cu–10% Zn alloy were processed through five revolutions in high-pressure torsion under an imposed pressure of 6 GPa. These materials were selected because the stacking fault energies vary from 78 mJ m^{-2} in Cu to 35 mJ m^{-2} in Cu–10% Zn. Grain sizes were measured by transmission electron microscopy and by X-ray diffraction both after HPT and after a combination of HPT and cold-rolling to a reduction of ∼75%.

(ii) The grain sizes produced in HPT are smaller than the grain sizes reported after processing by equal-channel angular pressing. By comparison with pure Cu, the measured average grain size in the Cu–10% Zn alloy is smaller and the width of the grain size distribution is smaller because of the lower stacking fault energy.

(iii) All of the results, after both HPT and HPT + CR, are generally consistent with a detailed theoretical model predicting the minimum grain sizes produced by ball milling.

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