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Stability of ultrafine-grained microstructure in fcc metals processed by severe plastic deformation

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Abstract. The thermal stability of ultrafine-grained (UFG) microstructure in face centered cubic metals processed by severe plastic deformation (SPD) was studied. The influence of the SPD procedure on the stability was investigated for Cu samples processed by Equal-Channel Angular Pressing (ECAP), High-Pressure Torsion (HPT), Multi-Directional Forging and Twist Extrusion at room temperature (RT). It is found that HPT results in the lowest thermal stability due to the very high dislocation density. Furthermore, the effect of the low stacking fault energy of Ag on the stability is also investigated. It is revealed that the UFG microstructure produced in Ag by ECAP is recovered and recrystallized during storage at room temperature. The driving force for this unusual recovery and recrystallization is the high dislocation density developed during ECAP due to the high degree of dislocation dissociation caused by the very low stacking fault energy of Ag.

Introduction

Severe plastic deformation (SPD) procedures are attractive methods for producing ultrafine-grained (UFG) metals in bulk form [1]. In the last decades, several methods such as Equal-Channel Angular Pressing (ECAP), High-Pressure Torsion (HPT), Multi-Directional Forging (MDF) and Twist Extrusion (TE) were developed for introducing severe plastic deformation into metals and alloys [2]. These procedures are quite different both in the physical principle of their operations and in the sizes of the workable pieces. Because of their practical importance, UFG face centered cubic (fcc) metals processed by different SPD procedures have been studied extensively (e.g. in Ref. [3]). The evolution of the microstructure as a function of strain, the characteristic parameters of microstructure in the saturation state and the correlation between the microstructure and the mechanical behavior have been established in detail. The thermal stability of the UFG microstructure processed by SPD is of great importance from the point of view of practical applications of these materials. Thus, if the fine grains become coarsened during their service life-time, their unique properties including their high strength will be lost. In the present work, the stability of the SPD-processed UFG microstructure in fcc metals was studied. First, the effect of the applied SPD method on the thermal stability of UFG Cu is investigated. Secondly, the influence of the low stacking fault energy (SFE) on the stability of the UFG microstructure at room temperature (RT) is studied.

Experimental materials and procedures

Oxygen-free copper (99.98% purity) samples were processed by 15 passes of TE, 20 cycles of MDF and 25 passes of ECAP at RT which correspond to equivalent strain values of about 14, 50 and 29, respectively, in the applied configurations (not presented here). According to former experiments (e.g. in Ref. [3]) these strains are above the threshold values corresponding to the saturation microstructure. An additional sample was processed by 25 revolutions of HPT on a disk having 10 mm in diameter and 0.6 mm in initial thickness. After 25 revolutions at a pressure of 4 GPa the thickness was reduced to 0.2 mm due to the unconstrained conditions of HPT. The HPT-processed disk was studied at the half-radius where the equivalent strain was about 567 assuming a mean thickness of 0.4 mm. The hardness of the SPD-processed Cu samples was measured after annealing for 1 h at different temperatures between 293 and 623 K. The thermal stability was also monitored by differential scanning calorimetry (DSC) at a heating rate of 40 K/min.

High-purity 99.99% Ag billets having lengths of ~70 mm and diameters of ~10 mm were pressed through totals of 1, 4, 8 and 16 passes of ECAP using route B_C at RT in a die having an internal channel angle of 90° and an outer arc of curvature of ~20° at the intersection of the two parts of the channel. In this configuration, one pass corresponds to an equivalent strain of ~1 [4]. Following ECAP, the billets were stored at RT and the microstructures and hardness were examined as a function of the time of storage for periods up to a total of four months.

The microstructures of Cu and Ag samples were investigated by transmission electron microscopy (TEM) using a JEM–100CX microscope and by X-ray line profile analysis. The X-ray line profiles were measured by a special high-resolution diffractometer (Nonius FR591) using $CuK\alpha_1$ radiation (λ = 0.15406 nm). The line profiles were evaluated by the extended Convolutional Multiple Whole Profile (eCMWP) fitting procedure described elsewhere [5]. This method gives both the dislocation density and the twin boundary frequency with good statistics, where the twin boundary frequency is defined as the fraction of the twin boundaries among {111} planes along their normal vector.

Results and discussion

The average grain size determined by TEM for Cu samples are listed in Table 1. All the grain size values are slightly above 200 nm, except for the HPT-processed specimen having a grain size of about 160 nm. The dislocation densities obtained by X-ray line profile analysis are also listed in Table 1. The lowest dislocation densities were measured after MDF and TE processes while the largest value was obtained after HPT. The twin boundary frequency for all the studied samples was relatively low (0.1±0.1 %), close to the detection limit of twin boundary frequency of X-ray line profile analysis for the applied experimental setup. The extremely high dislocation density after HPT is in good agreement with the values obtained by other authors [6]. The very high dislocation density can be attributed to the high pressure applied during HPT as this pressure increases the formation and migration enthalpy of vacancies [6] thereby hindering the annihilation of edge dislocations by climb. Moreover, the high applied pressure also increases the stresses which can assit or hinder the cross-slip of screw dislocations depending on their orientation relative to the line and Burgers vectors [7]. Due to the medium value of SFE for Cu, without stresses the degree of dislocation dissociation is not very large therefore the assisting stresses have only a minor promotion effect on cross-slip. However, the hindering stresses can increase effectively the degree of dislocation dissociation resulting in a difficult cross-slip and therefore an obstruction of dislocation annihilation during HPT which also contributes to the very high dislocation density.

During annealing of the SPD-processed samples in DSC an exothermic peak evolved which corresponds to the recovery and recrystallization of the microstructure as was shown in previous studies [8]. Table 1 shows that the lower onset temperature of recovery/recrystallization is related to the higher dislocation density due to the higher stored strain energy, therefore the samples produced by MDF and TE show the highest thermal stability while the HPT-processed specimen has the lowest stability. This result is also supported by the change of hardness as a function of annealing temperature presented in Fig. 1. The hardness of the sample processed by HPT starts to decrease at the lowest temperature compared to other specimens while the hardness of the MDF and TE samples remains unchanged up to relatively high temperatures.

Table 1: The average grain size (D_{TEM}) determined by TEM, the dislocation density (ρ) obtained from X-ray line profile analysis and the onset temperature of recovery/recrystallization (T_{onset}) measured by DSC at a heating rate of 40 K/min.

	D_{TEM} [nm]	ρ [10^{14} m^{-2}]	T_{onset} [K]
20 MDF	225	7±1	581±3
15 TE	225	10±1	572±3
25 ECAP	215	15±1	469±2
25 HPT	160	37±4	419±2

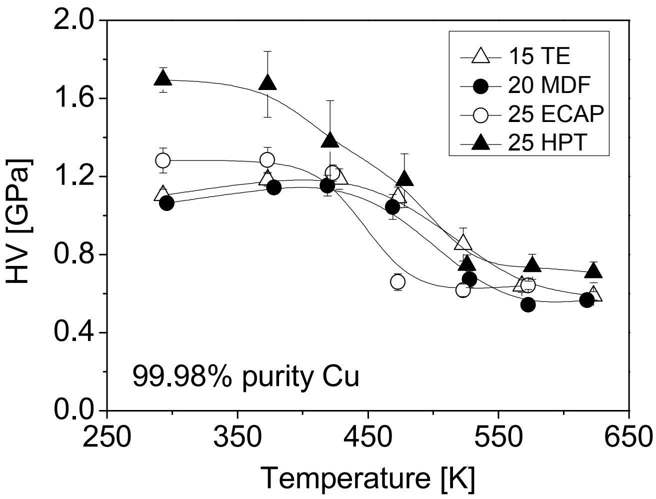
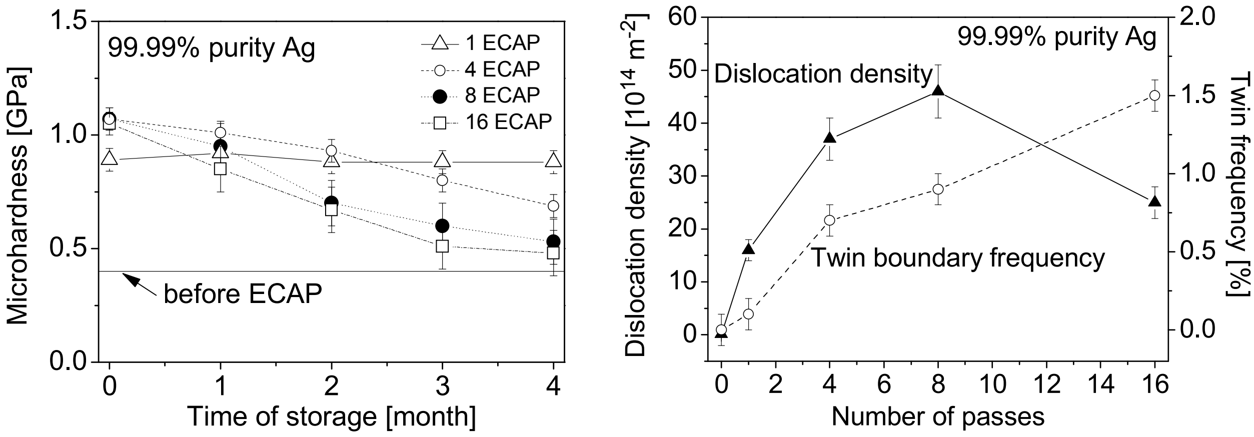


Figure 1: The hardness as a function of annealing temperature for 99.98% purity Cu samples processed by 15 passes of TE, 20 cycles of MDF, 25 passes of ECAP and 25 revolutions of HPT.

Figure 2a shows the microhardness of Ag samples processed by different numbers of ECAP passes as a function of the time of storage at room temperature, where the lower horizontal line denotes the hardness of the initial sample. It is apparent that the hardness after 1 pass remains unchanged within experimental error even after storage for 4 months. By contrast, the hardness gradually decreases with increasing storage time for the samples processed by 4, 8 and 16 passes due to recovery and recrystallization during long-term storage at room temperature. The recovery and recrystallization during storage were also confirmed in earlier microstructure investigations [7]. The higher the number of passes, the lowest the thermal stability of the microstructure. Figure 2b shows the dislocation density and the twin boundary frequency as a function of the number of ECAP passes. The extremely large dislocation density above 4 passes can be attributed to the hindered non-conservative motion of dislocations due to their high degree of dissociation resulting from the extremely low SFE of Ag [7]. Calculations have shown recently [7] that the high stresses caused by the large dislocation density after 4 passes may result in a delayed cross-slip of screw dislocations several months after SPD leading to the experimentally observed self-annealing of ECAP-processed Ag samples. It is worth noting that the microstructure obtained after 16 passes shows the lowest stability despite its smaller dislocation density compared with the samples processed by 4 and 8 passes. This apparent dichotomy can be explained by the relatively large twin boundary frequency after 16 passes (see Fig. 2b) as in the volumes where twins formed at the expense of dislocations, the stored energy decreased. These volumes may then act as the embryos for the new grains formed by recrystallization thereby reducing the time required for grain nucleation.



(a) (b)

Figure 2: (a) The microhardness as a function of time of storage at RT for Ag processed by ECAP for different number of passes. (b) The variation of dislocation density and the twin boundary frequency with the number of ECAP passes for Ag.

Summary

It was found for Cu samples processed by different methods of SPD that the thermal stability decreased in the order of MDF, TE, ECAP and HPT which correlates to the increase of dislocation density. The very low SFE of pure Ag resulted in an extremely low thermal stability due to the high degree of dislocation dissociation that yielded an extremely high value for the dislocation density. The high stresses evolved in this microstructure led to dislocation annihilation several months after ECAP. The low SFE also resulted in a large twin boundary frequency at high numbers of passes which facilitates the recrystallization.

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References

[1] R.Z. Valiev, R.K. Islamgaliev and I.V. Alexandrov: Prog. Mater. Sci. Vol. 45 (2000), p. 103

[2] R. Z. Valiev, Y. Estrin, Z. Horita, T. G. Langdon, M. Zehetbauer and Y. T. Zhu: JOM Vol. 58(4) (2006), p. 33

[3] N.Q. Chinh, J. Gubicza and T.G. Langdon: J. Mater. Sci. Vol. 42 (2007), p. 1594

[4] Y. Iwahashi, J. Wang, Z. Horita, M. Nemoto and T.G. Langdon: Scripta Mater. Vol. 35 (1996), p. 143

[5] L. Balogh, G. Ribárik and T. Ungár: J. Appl. Phys. Vol. 100 (2006), p. 023512

[6] E. Schafler: Scripta Mater. Vol. 62 (2010), p. 423

[7] J. Gubicza, N. Q. Chinh, J. L. Lábár, Z. Hegedűs and T. G. Langdon: Mater. Sci. Eng. A. Vol. 527 (2010), p. 752

[8] N. Gao, M.J. Starink and T.G. Langdon: Mater. Sci. Technology Vol. 25 (2009), p. 687