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Thermal Stability of Ultra Fine Grained Cu with Al_2O_3 Nanoparticles Prepared by High Pressure Torsion

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Abstract. Bulk ultra fine grained (UFG) metals prepared by severe plastic deformation exhibit often improved mechanical properties. However, at elevated temperatures grain growth occurs and the advantageous properties are lost. In the present work we examined whether the thermal stability of UFG Cu can be improved by addition of Al_2O_3 nanoparticles. Bulk UFG Cu samples with Al_2O_3 nanoparticles were prepared by high-pressure torsion (HPT). Thermal stability of UFG Cu with Al_2O_3 nanoparticles was compared with thermal stability of pure UFG Cu prepared by the same procedure. Development of microstructure and recovery of defects with increasing temperature was studied by positron annihilation spectroscopy combined with TEM. We have found that presence of Al_2O_3 nanoparticles leads indeed to improved thermal stability of UFG structure.

Introduction

Bulk UFG materials with no residual porosity and grain size ≈ 100 nm can be produced by high pressure torsion [1]. Number of UFG metals exhibit improved mechanical properties consisting in a favorable combination of very high strength and sufficient ductility [1]. It makes them highly attractive for future industrial applications. The superior mechanical properties are due to very small grain size, which results in a significant volume fraction of grain boundaries (GBs). At elevated temperatures grain growth occurs and the advantageous properties are lost. Development of microstructure of UFG Cu with temperature was studied in details in [2,3]. It was found that recovery of UFG structure is realized by abnormal grain growth, when isolated recrystallized grains appear in virtually unchanged deformed matrix, followed by recrystallization, i.e. grain growth in the whole volume of sample. Reduction of grain size leads to a shift of recrystallization to lower temperatures. For example, the onset of recrystallization in HPT deformed UFG Cu sample with grain size of 150 nm was found at 280°C, while recrystallization in UFG Cu with grain size of 110 nm starts already at 190°C. It is clear that relatively low thermal stability of UFG structure represents a serious limitation of usability of UFG materials. Thus, it is highly desirable to improve thermal stability of UFG structure.

Ceramic particles are stable up to very high temperatures. Moreover, if finely dispersed, they provide effective obstacles for movement of dislocations and GBs. Hence, one can expect that addition of ceramic nanoparticles into metal matrix could improve thermal stability of UFG structure. In the present work we examined this hypothesis. We prepared UFG Cu with Al_2O_3

nanoparticles. At first microstructure of such nanocomposites was characterized. Subsequently, thermal stability of UFG Cu with Al_2O_3 nanoparticles was compared with thermal stability of pure UFG Cu to check whether the ceramic nanoparticles can inhibit grain growth and, thereby, extend thermal stability of the UFG structure.

The UFG samples exhibit high density of defects introduced by severe plastic deformation. Characterization of these defects and their recovery is an important task in microstructure investigations of UFG materials. In the present work we employed positron annihilation spectroscopy (PAS) for defect studies. It is known that PAS represents a well established technique with very high sensitivity to open volume defects like vacancies, vacancy clusters, dislocations etc. [4]. PAS has been successfully used many times for defect studies of UFG materials, see e.g. [2,3,5]. In the present work we combined PAS investigations with direct observation of microstructure by TEM. Changes of mechanical properties were monitored by measurements of microhardness.

Experimental

In order to fabricate the UFG structure, the initial material of pure Cu (99.99%) and Cu with Al_2O_3 (GlidCop) were deformed by HPT at room temperature using pressure of 6 GPa. Samples of pure UFG Cu and UFG Cu with 0.5wt.% of Al_2O_3 nanoparticles were studied. The UFG samples were disk shaped with diameter ≈ 10 mm and thickness of ≈ 0.3 mm. After microstruture characterization the as-deformed samples were subjected to step-by–step isochronnal annealing with effective heating rate 1°C/min. Each annealing step was finished by rapid quenching and mictrostructure investigations at room temperature. A fast-fast positron lifetime (PL) spectrometer [6] with excellent timing resolution of 160 ps (FWHM ^{22}Na) at coincidence count rate of 120 s^{-1} was employed. Diameter of positron source spot was ≈ 4 mm. PL measurements were taken in the center of the sample. TEM observations were performed on the JEOL 2000 FX electron microscope operating at 200 kV. The microhardness HV was measured by the Vickers method with a load of 100 g applied for 10 s using the LECO M-400-A hardness tester.

Results and Discussion

As deformed structure. Typical TEM images of as-deformed pure UFG Cu are shown in Figs. 1a and 1b. The sample exhibits mean grain size ≈ 150 nm, mostly high-angle type GBs and high density of dislocations. Spatial distribution of dislocations is highly non-uniform: they are situated mainly at distorted layers around GBs, while grain interiors are almost free of dislocations. Bright-field TEM images from center and margin of HPT deformed UFG Cu with 0.5 wt.% of Al_2O_3 are shown in Figs. 2a and 2b, respectively. The sample exhibits virtually the same microstructure as pure UFG Cu. No differences between the central region and the margin were observed by TEM. Two kinds of Al_2O_3 particles were observed in the sample: (i) isolated coarse particles with size around 100 nm, i.e. comparable with grain size, and (ii) clumps of very fine Al_2O_3 nanoparticles with diameter less than 10 nm.

Lifetimes and relative intensities of the exponential components resolved in PL spectra of the studied samples are listed in Table 1. Majority of positrons are trapped at dislocations inside the distorted layers along GBs and contribute to the component with lifetime $\tau_1 \approx 164$ ps. A longer component with lifetime τ_2 comes from positrons trapped at small vacancy clusters so called microvoids situated inside grains. From the lifetime τ_2, one can deduce that size of microvoids corresponds approximately to 4-5 vacancies [2].

Table 1. Lifetimes and relative intensities of the components resolved in positron lifetime spectra of studied UFG samples.

HPT deformed sample	τ_1 [ps]	I_1 [%]	τ_2 [ps]	I_2 [%]
pure UFG Cu	164 ± 1	83 ± 5	255 ± 4	17 ± 3
UFG Cu with 0.5 wt.% Al_2O_3	165 ± 1	71 ± 1	301 ± 4	29 ± 1

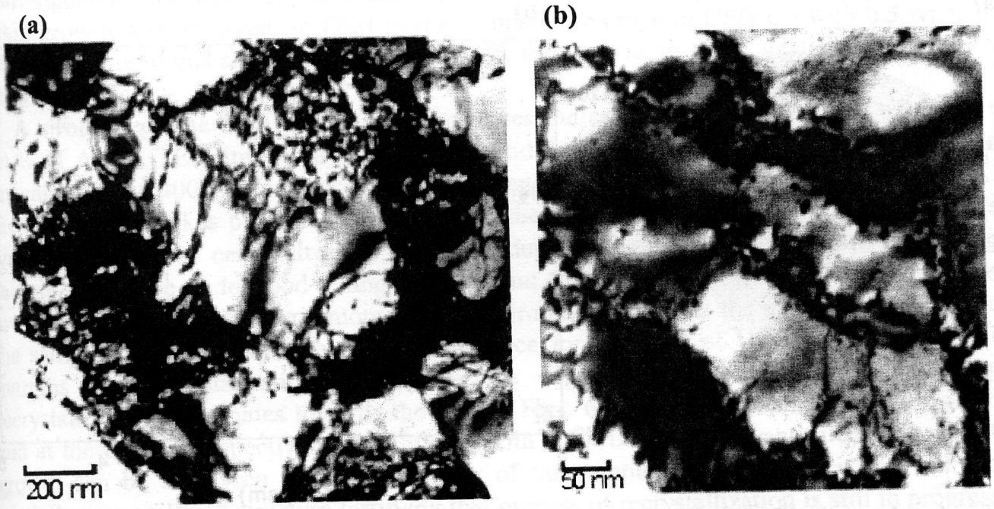

Fig. 1 (a) A bright field TEM image of HPT deformed pure UFG Cu, (b) the same sample in a higher magnification.

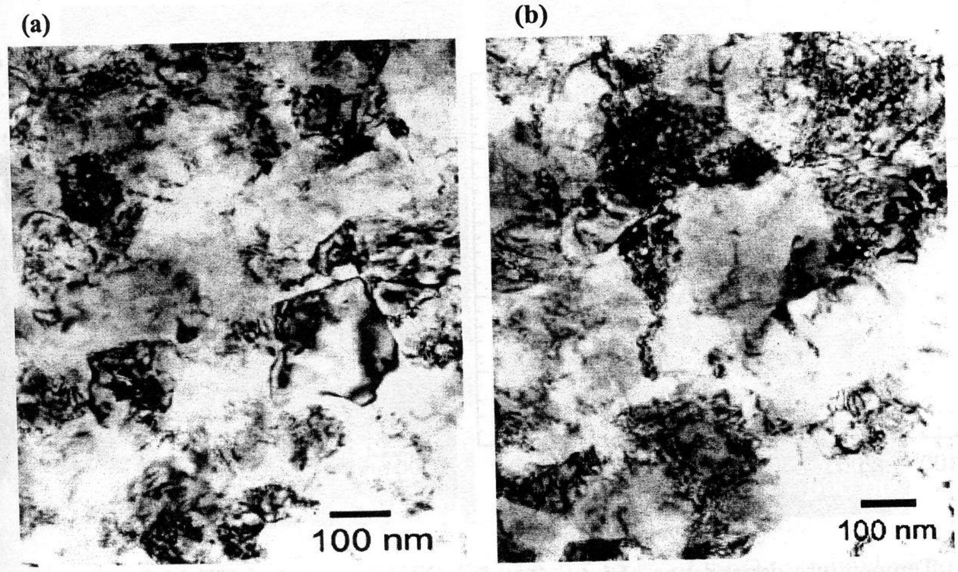

Fig. 2 Bright field TEM images of HPT deformed UFG Cu with 0.5 wt.% of Al_2O_3: (a) center of the sample, (b) margin.

In torsion deformation strain increases from the center of the sample towards margin. As a consequence defect density may depend on the radial distance r from the center of the sample, see Fig. 3a. Microhardness, HV, as a function of r for pure UFG Cu and UFG Cu with 0.5 wt.% of Al_2O_3 is plotted in Fig. 3b. An increase of HV with r indicates an increase of dislocation density from the center of the sample towards the margin. The center of the sample exhibits the lowest dislocation density, while highest number of dislocations can be found at the margin. Such spatial distribution of dislocations seems to be typical for HPT deformed samples.

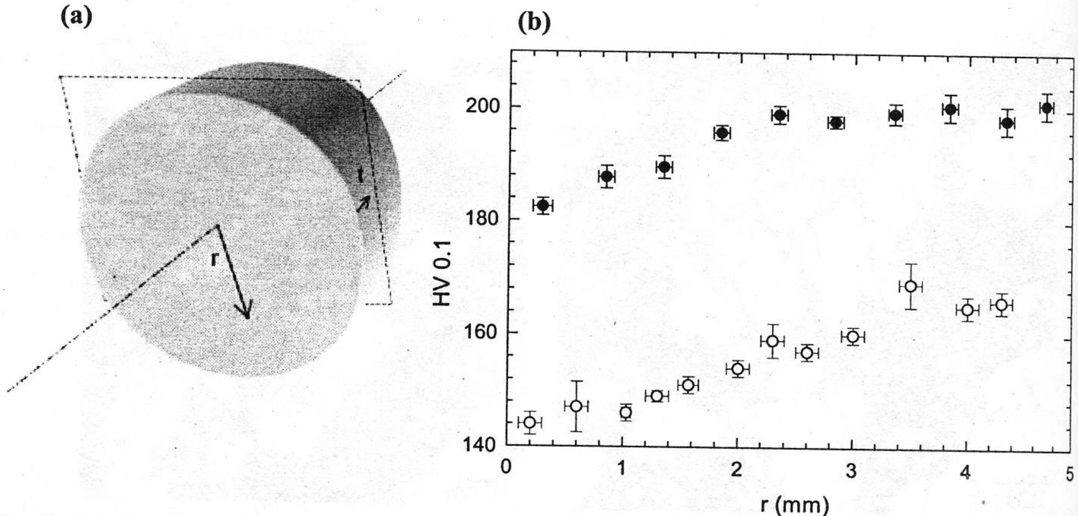

Fig. 3. (a) A schematic picture of UFG specimen, (b) dependence of microhardness HV on the radial distance r from the center of the sample: open circles pure UFG Cu, full circles UFG Cu with 0.5 wt.% of Al_2O_3,.

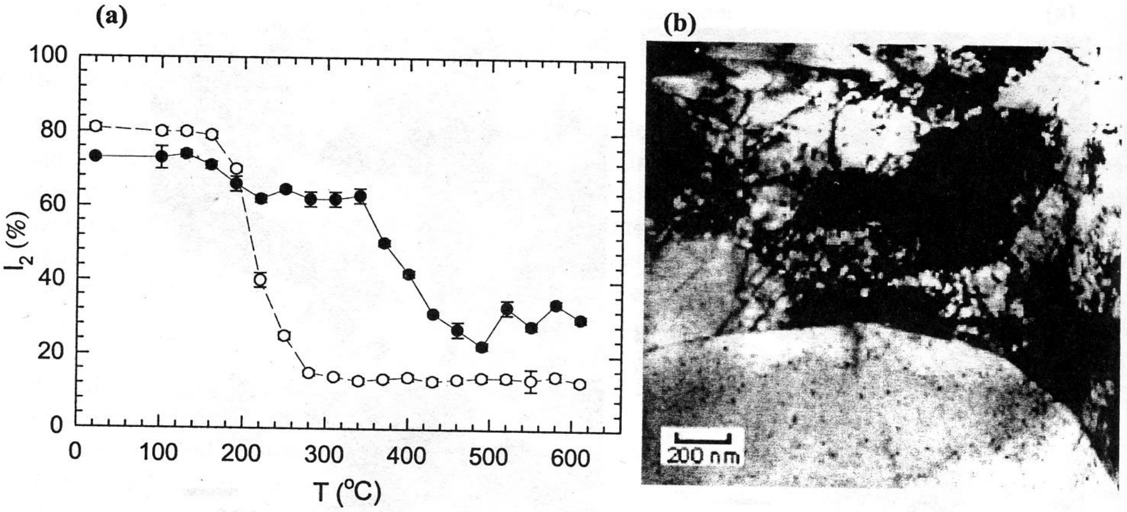

Fig. 4. (a) Temperature dependence of intensity of positrons trapped at dislocations: open circles pure UFG Cu, full circles UFG Cu with 0.5 wt.% of Al_2O_3, (b) a bright field TEM image of pure UFG Cu annealed up to 220°C.

Thermal Stability of UFG Structure. Temperature dependence of intensity I_2 of positrons trapped at dislocations is plotted in Fig. 4a. Recrystallization leads to a strong decrease in I_2 because the distorted regions with high dislocation density are replaced by recrystallized grains virtually free of dislocations. Pure UFG Cu exhibits a drastic decrease of I_2 starting at 190°C, see Fig. 4a. Indeed, intensive grain growth was observed by TEM in pure UFG Cu annealed up to 220°C. A boundary between a recrystallized and an UFG region is shown in Fig. 4b. On the other hand, UFG Cu with 0.5 wt.% of Al_2O_3 exhibits only a slight drop of I_2 around 200°C, which is likely due to some rearrangement and/or partial annihilation of dislocations and sharpening of the distorted regions. No grain growth was observed by TEM in this temperature range in UFG Cu with 0.5 wt.% of Al_2O_3. Microstructure of this sample remains essentially the same up to 380°C as demonstrated by behavior of I_2 in Fig. 4a.

A strong decrease of I_2 above 380°C indicates the onset of grain growth in UFG Cu with 0.5 wt.% of Al_2O_3. TEM images from the center and the margin of UFG Cu with 0.5 wt.% of Al_2O_3 annealed up to 400°C are shown in Figs. 5a and 5b, respectively. One can see in Fig. 5b that recrystallization takes place at the margin. On the other hand, basically unchanged UFG structure was observed in the center. It gives a clear evidence that recrystallization starts at the edge of the sample. It can be understood taking into account that strain increases from center of the sample towards margin. Thus, there is more stored deformation energy at the margins than in the center of the sample. As a consequence, the driving force for recovery of UFG structure is higher at the margins and recrystalization starts from the edge of the sample. At higher temperatures recrystallization propagates towards the center. Figs. 6a and 6b show microstructure in the center and at the margin, respectively, of UFG Cu with 0.5 wt.% of Al_2O_3 annealed up to 490°C. Grain growth can be now seen also in the center of the sample. Nevertheless, it still exhibits only a partially recrystallized structure testifying that process of recrystallization is still in progress. On the other hand, fully recrystallized structure was found at the margin, see Fig. 6b.

(a) (b)

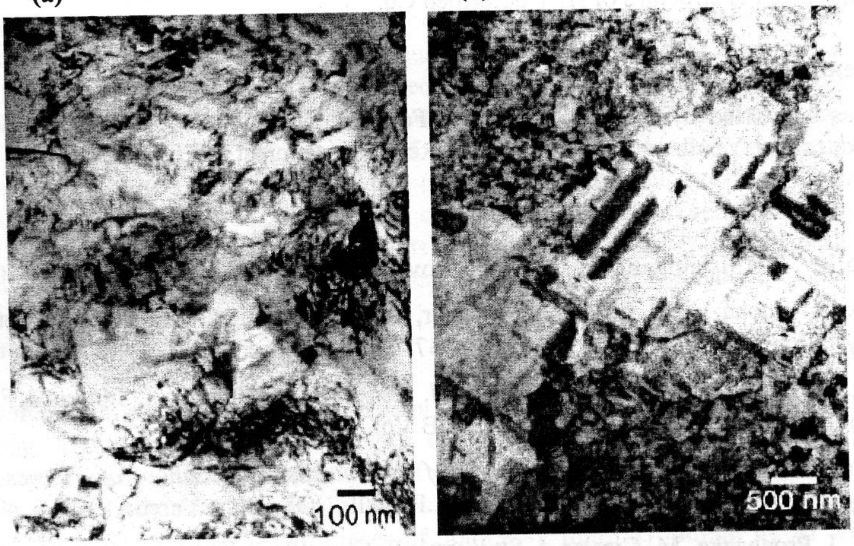

Fig. 5. Bright field TEM images of UFG Cu with 0.5 wt.% of Al_2O_3 annealed up to 400°C (a) center, (b) margin .

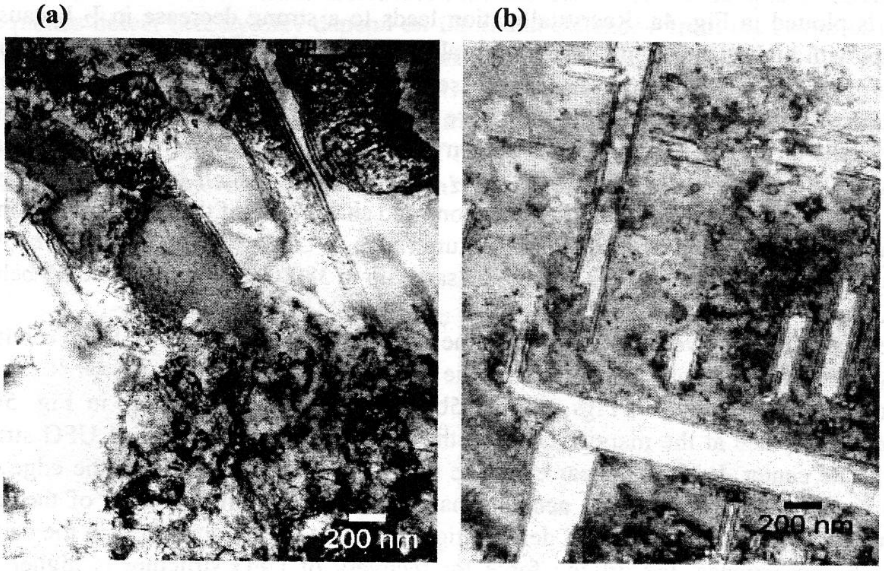

Fig. 6. Bright field TEM images of UFG Cu with 0.5 wt.% of Al_2O_3 annealed up to 490°C (a) center, (b) margin .

Conclusions

We have found that addition of 0.5 wt.% of Al_2O_3 nanoparticles leads to significantly enhanced thermal stability of UFG structure of HPT deformed Cu.

Acknowledgements

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