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Influence of ceramic nanoparticles on grain growth in ultra fine grained copper prepared by high pressure torsion

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Bulk ultra-fine grained (UFG) materials with no residual porosity can be produced by high pressure torsion (HPT). A number of UFG metals exhibit improved mechanical properties consisting in a favourable combination of very high strength and a reasonable ductility. Unfortunately, the thermal stability of UFG structure is rather low. Recrystallization takes place at elevated temperatures and the superior mechanical properties are lost. Thus, it is highly desirable to improve the thermal stability of UFG structure. In the present work we studied the thermal stability of UFG Cu containing Al_2O_3 nanoparticles. The aim of this work was to check whether the ceramic nanoparticles can inhibit grain growth and, thereby, to extend the thermal stability of UFG structure. We have found that the HPT deformed samples exhibit UFG structure with grain size around 150 nm and a high density of dislocations situated mainly in distorted layers along grain boundaries. The grain growth in the samples which contain 0.5 wt.% of the Al_2O_3 nanoparticles is shifted to a significantly higher temperatures compared to the pure UFG Cu. Thus, we can conclude that the addition of the Al_2O_3 nanoparticles (concentration at least 0.5 wt.%) leads to a significant improvement of the thermal stability of UFG structure.

1 Introduction Bulk UFG materials with no residual porosity and grain size ≈ 100 nm can be produced by HPT [1]. A number of UFG metals exhibit improved mechanical properties consisting in a favourable combination of a very high strength and a sufficient ductility [1]. The superior mechanical properties are due to very small grain size, which results in a significant volume fraction of grain boundaries (GB's). Unfortunately, at elevated temperatures grain growth occurs and the advantageous properties are lost. Ceramic particles are stable up to very high temperatures. Moreover, if finely dispersed, they provide effective obstacles for movement of dislocations and GB's. Hence, one can expect that addition of the ceramic nanoparticles into a 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 the microstructure of such nanocomposites was characterized. Subsequently, thermal stability of UFG Cu with Al_2O_3 nanoparticles was compared with the thermal stability of pure UFG Cu in order to check whether the ceramic nanoparticles can inhibit grain growth.

2 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. The

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samples of pure UFG Cu and UFG Cu containing 0.5wt.% of Al_2O_3 nanoparticles were studied. The UFG samples were disk shaped with diameter of ≈ 10 mm and thickness of ≈ 0.3 mm. After microstructure characterizations the as-deformed samples were subjected to step-by–step isochronnal annealing with the 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 [2] with timing resolution of 160 ps (FWHM ^{22}Na) at the coincidence count rate of 120 s^{-1} was employed. Diameter of the positron source spot was ≈ 4 mm. PL measurements were taken in the centre 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.

3 Results and discussion

3.1 As deformed structure The HPT deformed pure UFG Cu and UFG Cu with Al_2O_3 nanoparticles exhibit very similar microstructure which is shown in Fig. 1a. The samples exhibit mean grain size ≈ 150 nm, mostly high-angle type GB's and a high density of dislocations. The spatial distribution of dislocations is highly non-uniform: they are situated mainly at the distorted layers around GB's, while the grain interiors are almost free of dislocations. Two kinds of Al_2O_3 particles were observed: (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.

PL spectra of the as-deformed samples are well fitted by two components. Majority of positrons are trapped at dislocations inside the distorted layers along GB's and contribute to the shorter component with lifetime ≈164 ps. A longer component with lifetime in the range 260-300 ps comes from positrons trapped at small vacancy clusters (so called microvoids) situated inside grains. From the lifetime of this component, one can deduce that size of microvoids corresponds approximately to 4-7 vacancies [2].

In torsion deformation strain increases from the centre of the sample towards margin. As a consequence defect density may depend on the radial distance r from the centre of the sample. 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. 2a. An increase of HV with r indicates an increase of dislocation density from the centre of the sample towards the margin. The centre 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.

3.2 Thermal stability of UFG structure Temperature dependence of the lifetimes τ_i and the relative intensities I_i of the components resolved in PL spectra of the UFG Cu and the UFG Cu with 0.5 wt.% of Al_2O_3 are in Fig. 3a,b. The lifetimes τ_2 and τ_3 of positrons trapped at dislocation and microvoids, respectively, remain essentially the same during annealing indicating that the nature of the defects does not change and only their concentrations vary with temperature. The recrystallization leads to a strong decrease in I_2 because the distorted regions with high dislocation density are replaced by the dislocation-free recrystallized grains. Moreover, the free positron component with lifetime τ_1 appears in the PL spectrum. It is a contribution of free positron annihilations in the recrystallized grains. In the pure UFG Cu the recrystallization starts at 190 °C, see Fig. 3b. Indeed, an intensive grain growth was observed by TEM in the pure UFG Cu annealed up to 220 °C. 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 (Fig. 3b), 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 350 °C as demonstrated by behaviour of I_2 in Fig. 3b. Selected HV(r) curves for various temperatures for UFG Cu with 0.5 wt.% of Al_2O_3 are plotted in Fig. 2b. No change in HV was seen up to 350°C in agreement with PL results. One can see in Fig. 2b that at 370 °C there is an abrupt decrease in HV at the margin. It clearly indicates that the recrystallization starts from the edge of the sample. With increasing temperature the recrystallization propagates towards the centre of the sample, see Fig. 2b. It can be understood taking into account that the strain increases from centre of the sample towards margin. Thus, there is more stored deformation energy at the margins than in the centre of the sample. As a consequence, the driving force for recovery of the UFG structure is higher at the margins and recrystalization at the margins starts, therefore, at lower temperature. A TEM image from the margin of the UFG Cu with 0.5 wt.% of Al_2O_3 annealed up to 400 ^oC is shown in Fig. 1b. It shows clearly that recrystallization has already started, while the centre of the sample still shows virtually unchanged UFG structure. Figs. 1c,d show the microstructure in the centre and at the margin, respectively, of the UFG Cu with 0.5 wt.% of Al_2O_3 annealed up to 490 ^oC. The grain growth can be now seen also in the centre of the sample. Nevertheless, the centre still exhibits only a partially recrystallized structure testifying that the process of recrystallization is still in progress. On the other hand, fully recrystallized structure with the annealing twins is visible at the margin (Fig. 1d).

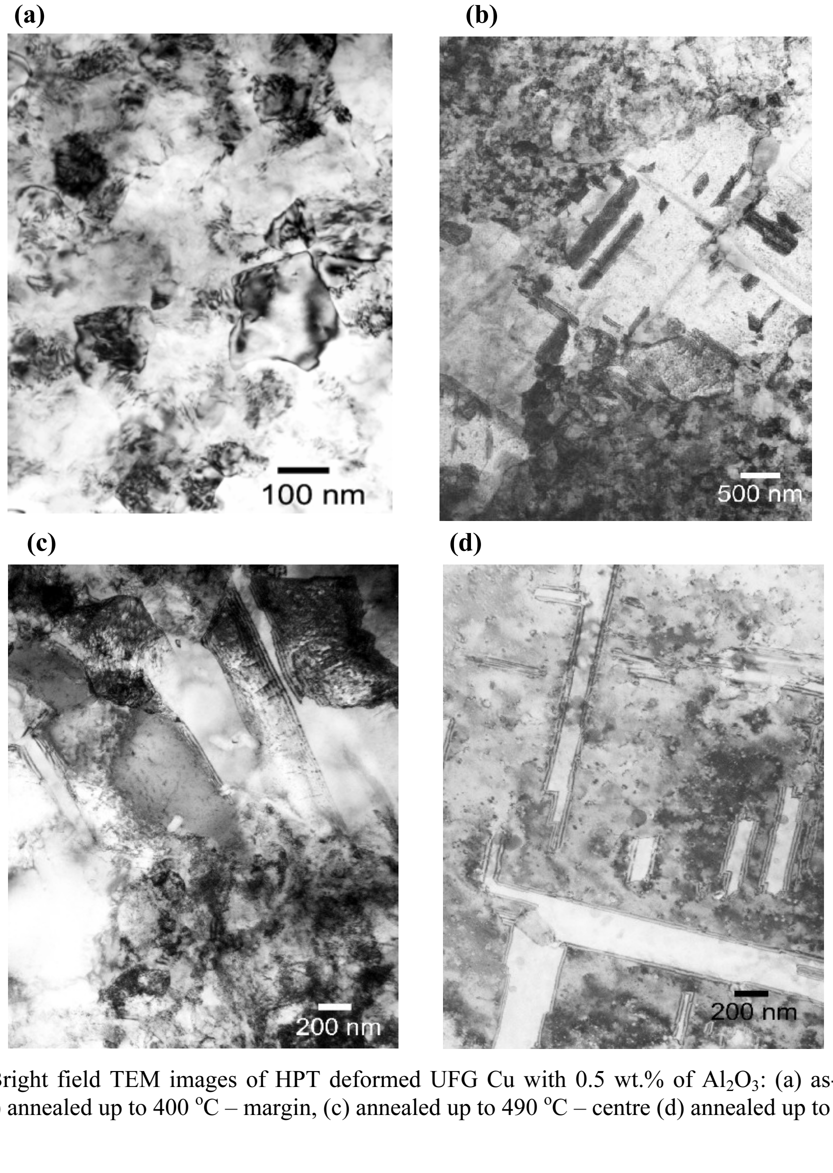

Fig. 1 Bright field TEM images of HPT deformed UFG Cu with 0.5 wt.% of Al_2O_3: (a) as-deformed sample – centre, (b) annealed up to 400 ^oC – margin, (c) annealed up to 490 ^oC – centre (d) annealed up to 490 ^oC – margin.

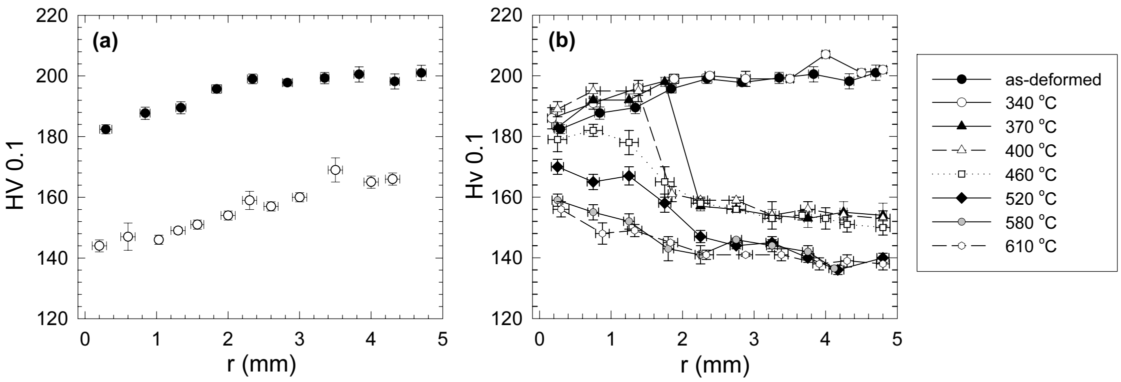

Fig. 2 (a) Dependence of microhardness HV on the radial distance *r* from the centre of the sample: open circles pure UFG Cu, full circles UFG Cu with 0.5 wt.% of Al_2O_3, (b) selected HV(*r*) curves at various temperatures for the UFG Cu with 0.5 wt.% of Al_2O_3.

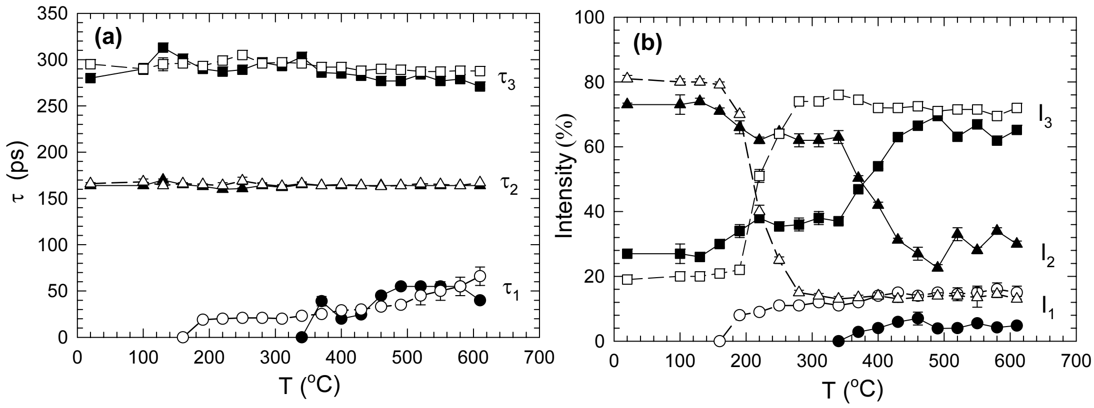

Fig. 3 Temperature dependence of the lifetimes (a) and the relative intensities (b) of the components resolved in PL spectra of HPT deformed pure Cu (open symbols) and Cu with 0.5 wt.% Al_2O_3 (full symbols).

4 Conclusions The UFG Cu and UFG Cu with 0.5 wt.% of Al_2O_3 nanoparticles were prepared by HPT. The samples exhibit UFG structure with grain size around 150 nm. It was shown that the sample with the Al_2O_3 nanoparticles exhibits significantly enhanced thermal stability of UFG structure compared to pure UFG Cu. It shows that the Al_2O_3 nanoparticles stabilize the UFG structure and suppress grain growth. It was found that the recrystallization starts from the margin of the sample.

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