Dependence of Thermal Stability of Ultra Fine Grained Metals on Grain Size

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1 Introduction

High pressure torsion (HPT) is a technique based on severe plastic deformation (see [1] for review). The initial coarse-grained sample is deformed by torsion and simultaneously by high pressure of several GPa is applied. Ultra fine grained (UFG) metals with grain size around 100 nm can be produced by HPT. The mean grain size of HPT prepared sample depends on material and parameters of the deformation. Different grain size can be obtained, e.g. by varying the applied hydrostatic pressure.

UFG structure of as-deformed samples represents highly non-equilibrium kind of material structure from thermodynamic point of view, therefore, processes of its recovery towards more equilibrium one can be observed with increasing temperature. In the present work we studied how grain size of as-deformed UFG structure influences its thermal recovery. UFG microstructure and its evolution with increasing temperature were studied by positron lifetime (PL) spectroscopy, which represents non-destructive technique with very high sensitivity to open-volume defects such as vacancies, vacancy clusters, dislocations etc [2]. PL spectroscopy was combined with X-ray diffraction (XRD) and transmission electron microscopy (TEM).

2 Experimental Details

Two samples of UFG Cu (purity 99.99 %) denoted as A and B were prepared by HPT. In order to obtain different grain sizes the sample A was made using hydrostatic pressure p = 3 GPa, while the sample B using p = 6 GPa. Microstructure of the as-deformed samples was characterized. Subsequently, the samples were isochronally annealed with effective heating rate 1 K/min. The heating was carried out in silicon oil bath up to 250 °C and in vertical furnace with protective argon atmosphere above this temperature. Each annealing step was finished by rapid quenching into water of room temperature. A PL spectrometer with timing resolution of 150 ps (FWHM ^{22}Na) at coincidence count rate of 80 counts/s was used. See [3,4] for its detailed description. XRD studies were carried out with the aid of XRD7 and HZG4 (Seifert-FPM) powder diffractometers using Cu K_α radiation. TEM observations were performed on the JEOL 2000 FX electron microscope operating at 200 kV.

3 Results and Discussion

3.1 As-Deformed Samples

The mean grain sizes d determined by TEM for samples A and B are shown in Table 1. Clearly, the sample A prepared using lower pressure exhibits larger grain size.

Table 1: Structure properties of the as-deformed UFG Cu samples. Hydrostatic pressure p used in HPT is given in the second column. The mean grain size determined by TEM and domain size obtained from XRD and PL spectroscopy, respectively, are given in the next columns. The values in parenthesis represent one standard deviation.

Sample	p [GPa]	grain size [nm]	Domain size [nm]	
		TEM	XRD	PL
A	3	150(10)	120(20)	90(5)
B	6	105(10)	80(20)	70(10)

Bright field TEM image of sample A is shown in Fig. 1. One can see that it contains relatively high number of dislocations. Spatial distribution of the dislocations is strongly non-homogeneous. Grain interiors (non-distorted regions) almost free of dislocations are separated by distorted layers with high dislocation density. The distorted layers with thickness around 10 nm are situated along grain boundaries (GBs). Rather "diffusive" contrast testifies non-equilibrium state of majority of GBs. Microstructure of the sample B exhibits the same features as that of sample A. The only difference is smaller grain size.

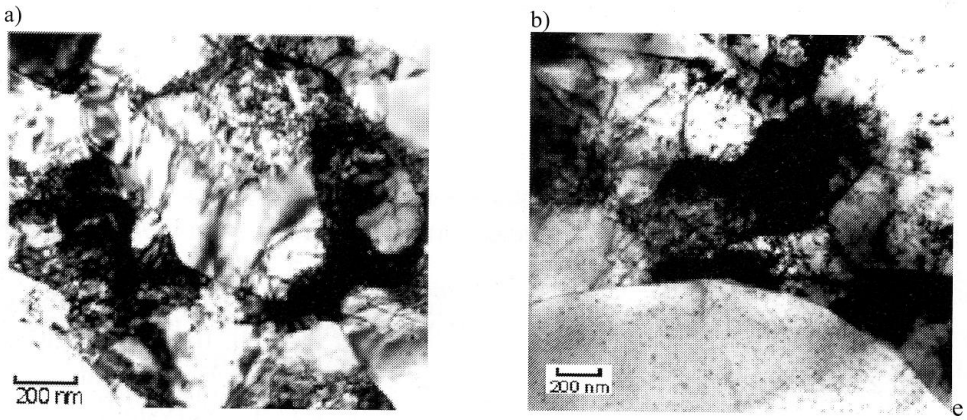

Figure 1: Bright field TEM image of sample A a) as-deformed state b) after annealing up to 250 °C

Broadening of X-ray diffraction profiles with characteristic anisotropy of the type $\beta_{h00} < \beta_{hhh}$ was observed in both samples. It can be explained by dislocation-induced line broadening. We used a simple procedure described in [5] to obtain coherent domain size from analysis of x-ray diffraction profiles. This procedure is similar to more complex procedure introduced by Ungár et al. [6,7]. The mean coherent domain sizes for both samples are given in Table 1.

PL spectra of both samples can be well fitted by two components. Lifetimes and relative intensities of these components are listed in Table 2. Detailed discussion of PL results for sample A can be found in [8]. Therefore, only brief description is given here and we will focus on differences in thermal recovery of both samples. The first component with lifetime $\tau_1 \approx 163$ ps comes from positrons trapped at dislocations inside the distorted regions. The longer component can be attributed to positrons trapped in microvoids, i.e. point defects with size of 4–5 vacancies [8]. No free positron component was found. It means that all positrons are trapped at defects in both samples. Sample B exhibits higher concentration of microvoids compared to sample A.

Table 2: Lifetimes and relative intensities of components resolved in PL spectra of as-deformed samples A and B. The values in parenthesis represent one standard deviation.

Sample	τ_1 [ps]	I_1 [%]	τ_2 [ps]	I_2 [%]
A	164(1)	83(4)	255(4)	17(3)
B	161(3)	64(4)	249(2)	36(5)

We developed a diffusion trapping model, which allows determination of size of the non-distorted regions (grain interiors) and volume fraction of the distorted regions from experimental PL spectra. The model is based on positron diffusion model introduced by Dupasquier et al. [9] modified for the structure of UFG metals. Detailed description of the model is out of scope of this paper and can be found in [8]. Size of the non-distorted regions obtained using the model is given in Table 1. One can see that the size of the non-distorted regions agrees well with the domain size determined by XRD, as both quantities represents size of dislocation-free grain interiors. Both these values are slightly lower than grain size obtained from TEM. It is because the former quantities are closely related to dislocation density, while grain size determined by TEM is connected with change of contrast between grain and GB. For detailed discussion see [8].

3.2 Thermal Recovery

Recovery of UFG structure is realized by similar processes in both samples. Therefore, we will illustrate them on an example of sample A. No change of microstructure of sample A was detected by TEM up to 190 °C. In temperature range from 190 °C to 250 °C isolated recrystallized grains with size ≈ 3 μm appeared in virtually unchanged deformed matrix. This so called abnormal grain growth was observed on UFG Cu also by Islamgaliev et al. [10]. Interface between the recrystallized grain and the deformed matrix is shown in Fig. 1b), which represents TEM image of sample A annealed up to 250 °C. Finally, from 280 °C to 400 °C, recrystallization in whole volume of sample takes place. Bright field TEM image of sample A annealed up to 400 °C is shown in Fig. 2a). Clearly, the material is fully recrystallized and exhibits mean grain size around 3 μm.

Temperature dependence of the mean positron lifetime is plotted in Fig. 2b). Two stages of recovery can be distinguished: (i) a drop in temperature interval 190–250 °C, which corresponds to the abnormal grain growth and (ii) radical decrease of the mean lifetime in temperature range 280-400 °C, which reflects the recrystallization.

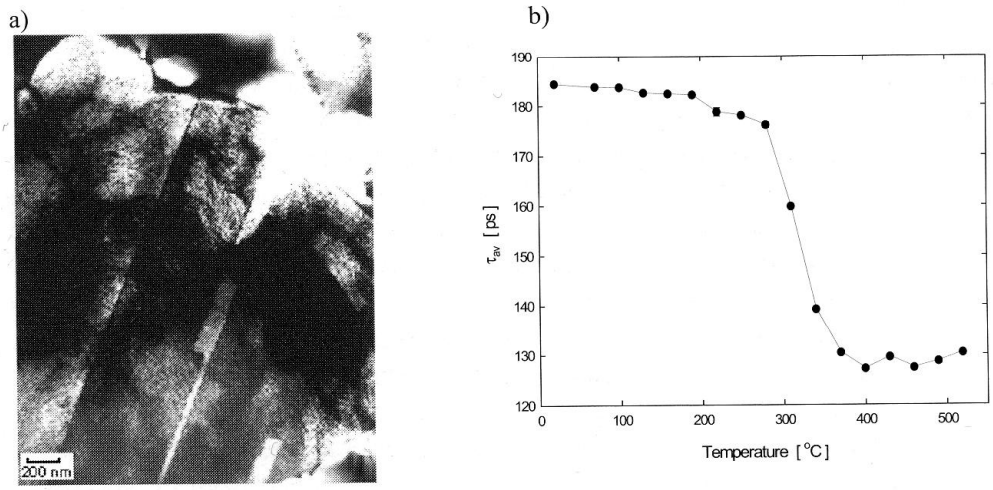

Figure 2: a) Bright field TEM image of sample A after annealing up to 400 °C. b) Mean positron lifetime for sample A as a function of annealing temperature

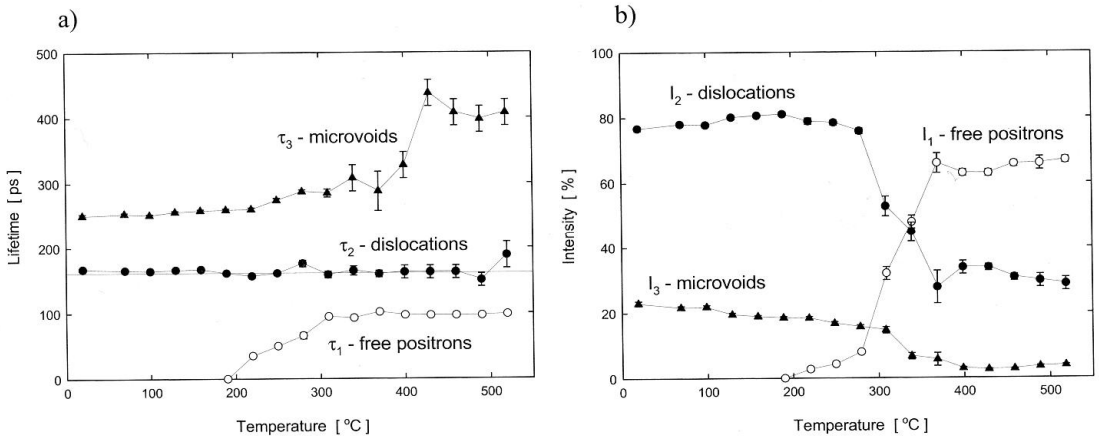

Figure 3: Temperature dependence of a) positron lifetimes and b) relative intensities of individual components for sample A

Temperature dependences of lifetimes and corresponding relative intensities for the sample A are shown in Fig 3 a) and b), respectively. Free positron component appeared in PL spectra from 190 °C when the abnormal grain growth occurs. It represents contribution of free positrons, which annihilate inside the recrystallized grains. Intensity of the free positron component radically increases during recrystalization (temperature interval 280–400 °C). It is accompanied by dramatic decrease of intensity I_2 of positrons trapped at dislocations inside the distorted regions. It reflects the situation when the distorted regions with high dislocation density are consumed by dislocation-free recrystallized grains. Lifetime τ_3 of microvoids increases during recrystallization, while intensity I_3 of this component decreases. It indicates that small microvoids become mobile and are annealed out or cluster to larger ones.

The volume fraction η of the distorted regions obtained by application of the diffusion model on PL spectra of isochronally annealed samples A and B is plotted in Fig 4a) as a function of annealing temperature. The volume fraction η exhibits abrupt decrease during the recrystallization

because the distorted regions are replaced by the recrystallized grains. It is clear from Fig. 4a) that in the case of sample A the abrupt decrease of η occurs at temperature interval 300–400 °C. It means in the temperature range when the recrystallization was observed in this sample by TEM, as was shown above. On the other hand, the abrupt decrease of η takes place from 190 °C to 250 °C in sample B. It means that the recrystallization is shifted to significantly lower temperatures in sample B, i.e. the sample with smaller grain size.

The mean size of the non-distorted regions (which is equivalent to domain size) obtained from PL results using the diffusion model is shown in Fig. 4b). As one can see in the figure the domain size substantially increases during the recrystallization. The increase of domain size starts around 300 °C in sample A in concordance with start of the recrystallization. On the other hand, in sample B the domain size begins to increase already from 190 °C. It clearly indicates that the recrystallization takes place at significantly lower temperatures in sample B due to smaller initial grain size. The domain size determined from XRD for sample B is also shown in Fig. 4b). It agrees well with the size of the non-distorted regions obtained from PL spectra. It was not possible to determine domain size at 200 °C and higher temperatures by XRD because the peak broadening was too small. Thus, domain size at 200 °C is higher than ≈ 300 nm, i.e. maximum domain size, which can be determined with resolution of the diffractometer used. It agrees well with the onset of the recrystallization at 190 °C determined by PL spectroscopy. Temperature dependence of the mean grain size obtained by TEM for sample B is shown in Fig. 4b) as well. TEM results are in reasonable agreement with PL spectroscopy and XRD. The mean grain size starts to significantly increase from 190 °C when the recrystallization occurs in sample B. Thus, grain growth takes place at about of 100 °C lower temperatures in sample B with smaller grain size of 105 nm compared to the sample A with grain size of 150 nm. We can, therefore, conclude that smaller initial grain size leads to lower thermal stability of corresponding UFG microstructure.

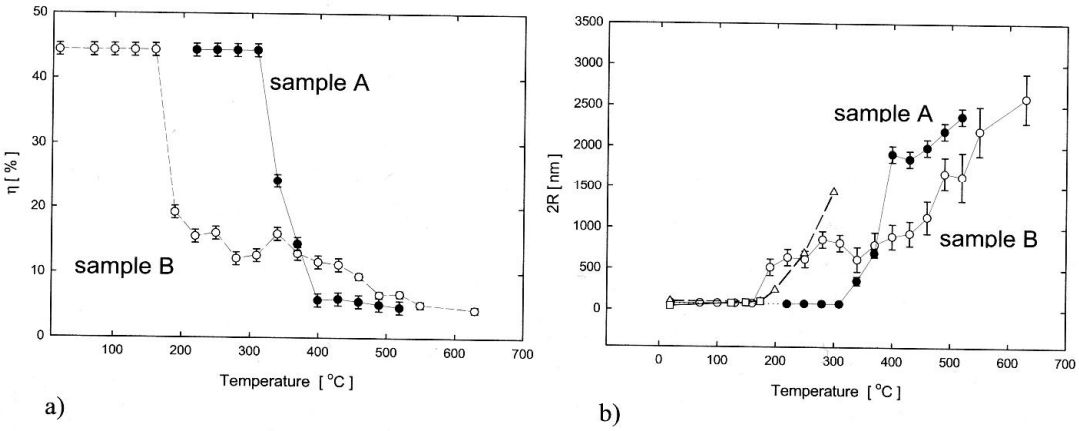

Figure 4: a) Temperature dependence of volume fraction η of the distorted regions obtained from PL spectra using the positron diffusion model. Full circles-sample A, open circles-sample B. b) Mean grain/domain size as a function of annealing temperature. Full circles – size of dislocation free grain interiors obtained from PL spectra of sample A, Open circles – size of the grain interiors obtained from PL spectra of sample B, Open triangles – grain size determined by TEM for sample B, Open squares – domain size determined from XRD for sample B.

4 Conclusions

Thermal stability of two samples of UFG Cu prepared by HPT with initial grain size of 150 nm and of 105 nm was compared in the present work. Recovery of the UFG structure is realized by the same processes in both samples. The abnormal grain growth when isolated recrystallized grains appeared in virtually unchanged deformed matrix is followed at higher temperatures by recrystallization in whole volume of sample. Temperature range when the recrystallization takes place is shifted to about of 100 °C lower temperatures in the sample with smaller grain size of 105 nm compared to that with grain size 150 nm. Hence, we can conclude that smaller initial grain size leads to lower thermal stability of UFG structure.

5 Acknowledgement

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6 References

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