Materials Science Forum Vol. 482 (2005) pp. 207-211
online at http://www.scientific.net

Positron annihilation studies of microstructure of ultra fine grained metals prepared by severe plastic deformation *

J. Čížek[1], I. Procházka[1,a], B. Smola[1], I. Stulíková[1], R. Kužel[1], M. Cieslar[1], Z. Matěj[1], V. Cherkaska[1], G. Brauer[2], W. Anwand[2], R.K. Islamgaliev[3], O. Kulyasova[3]

[1] Faculty of Mathematics and Physics, Charles University, V Holešovičkách 2, CZ-180 00 Praha 8, Czech Republic

[2] Institut für Ionenstrahlphysik und Materialforschung, Forschungszentrum Rossendorf, Postfach 510119, D-01314, Dresden, Germany

[3] Institute of Physics of Advanced Materials, Ufa State Aviation Technical University, Ufa 450 000, Russia

[a]ivanp@mbox.troja.mff.cuni.cz

Keywords: severe plastic deformation, positron annihilation, dislocations, microvoids.

Abstract. In the present work, positron annihilation spectroscopy (PAS) is employed for microstructure investigations of various ultra fine grained (UFG) metals (Cu, Ni, Fe) prepared by severe plastic deformation (SPD), namely high-pressure torsion (HPT) and equal channel angular pressing (ECAP). Generally, UFG metals prepared using both the techniques exhibit two kinds of defects introduced by SPD: dislocations and small microvoids. The size of the microvoids is determined from the PAS data. Significantly larger microvoids are found in HPT deformed Fe and Ni compared to HPT deformed Cu. The microstructure of UFG Cu prepared by HPT and ECAP is compared and the spatial distribution of defects in UFG Cu samples is characterized. In addition, the microstructure of a pure UFG Cu prepared by HPT and HPT deformed Cu+Al_2O_3 nano-composite (GlidCop) is compared.

Introduction

Bulk ultra fine grained (UFG) metals with no porosity can be prepared by methods based on severe plastic deformation (SPD) – for a review see Ref.[1] and references therein. The smallest grain sizes (typically around 100 nm) were achieved by high-pressure torsion (HPT). Samples deformed by HPT usually exhibit a homogeneous structure and only a weak texture. Disk shaped samples with a diameter of typically ~10 mm and a thickness of 0.2-0.5 mm can be prepared by HPT. More massive UFG specimens are produced by equal channel angular pressing (ECAP). It makes this technique more promising for possible future industrial applications. Nevertheless, the grain size of UFG samples prepared by ECAP is larger as a rule.

Contrary to conventional polycrystals, the volume fraction of grain boundaries (GB) becomes significant in UFG metals. This, together with a huge amount of defects (dislocations, vacancies, GBs) introduced by SPD, results in a highly non-equilibrium structure and a number of unusual physical properties [1]. Obviously, defects play a key role in the formation of UFG structure and are responsible for the extraordinary physical properties of these materials. Detailed defect studies thus become extremely important in order to understand microstructure of UFG metals.

Positron annihilation spectroscopy (PAS) is a non-destructive technique exhibiting a high sensitivity to open-volume defects [2]. Two PAS methods were employed in the present work: (i) positron lifetime (PL) spectroscopy for identification of defect type and evaluation of defect densities, and (ii) slow positron implantation spectroscopy (SPIS) with Doppler broadening measurements for identification of the spatial distribution of defects. Results of PAS investigations of various UFG metals prepared by HPT and ECAP are presented in this work.

Experimental

Specimens. The specimens of UFG metals were prepared by HPT at room temperature (RT). Torsion was performed up to a true logarithmic strain $\varepsilon = 7$ under high pressure of 6 GPa [1]. One set of UFG Cu samples was prepared using a pressure of 3 GPa. The HPT deformed samples were disk shaped with a diameter of 10-12 mm and a thickness of 0.2-0.4 mm. In addition, specimens of UFG Cu prepared by ECAP (route Bc, 12 passes, RT) were investigated.

Experimental Techniques. A PL spectrometer, similar to that described in [3], exhibiting timing resolution of 170 ps (FWHM for ^{22}Na), was employed in the present work. A positron source of 1.5 MBq ^{22}Na sealed between 2 μm mylar foils was used. At least 10^7 counts were collected in each PL spectrum. Decomposition of PL spectra into exponential components was performed using a maximum likelihood procedure [4]. SPIS was performed on the magnetically guided system "SPONSOR" at FZ Rossendorf [5]. Energy spectra of annihilation γ-rays were measured with a HPGe detector having an energy resolution (FWHM) of 1.09 keV at 511 keV. The diameter of the beam spot was ≈ 4 mm. The dependencies of annihilation lineshape parameter S on positron energy E were measured in the interval E = 0.03-35 keV and fitted by the program VEPFIT [6].

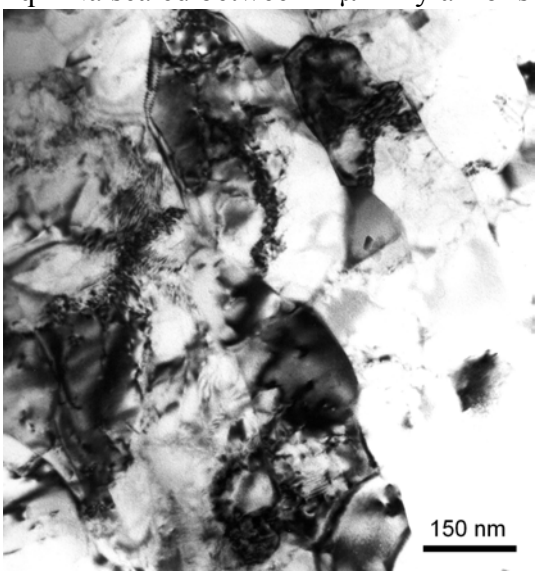

Fig.1: Bright field TEM image of HPT deformed Ni.

Results and Discussion

Defects in As-Deformed UFG Specimens. PL spectra of all samples studied could be resolved into two exponential components (except for the source contribution) and results are listed in Table 1. Lifetimes of both components are substantially higher than the respective bulk positron lifetimes τ_B. This clearly indicates that both components represent contributions of positrons trapped at defects. Virtually all positrons get trapped at defects prior to annihilation, and hence very high densities of defects occur in the as-deformed samples.

Table 1.Positron lifetimes τ_i and relative intensities I_i (i=1,2) observed in as-deformed UFG samples. Size of microvoids deduced from τ_2 is represented in the last column as corresponding number of vacancies n_V (see text for details). The errors (one standard deviation) are given in parentheses in units of the last significant digit.

Sample	τ_1 (ps)	I_1 (%)	τ_2 (ps)	I_2 (%)	n_V
HPT deformed Cu, p = 6 GPa	161(3)	64(4)	249(5)	36(4)	4.8(3)
HPT deformed Cu, p = 3 GPa	164(1)	83(4)	255(4)	17(3)	5.2(3)
HPT deformed Cu + 0.5 wt.% Al_2O_3 p = 6 GPa	161(3)	60.4(5)	257(1)	39.6(5)	5.31(7)
ECAP deformed Cu, route Bc, 12 passes	164(3)	80(6)	240(10)	20(6)	4.2(6)
HPT deformed Ni, p = 6 GPa	157(1)	88.9(6)	336(8)	11.1(6)	13.1(6)
HPT deformed Fe, p = 6 GPa	150.9(4)	90.6(3)	352(6)	9.5(3)	13.2(5)

The lifetime τ_1 of the shorter component with a dominant intensity I_1 is about 20 ps lower than the lifetime of positrons trapped at monovacancies in the corresponding material and typical for positron trapping at dislocations. A high density of dislocations introduced by SPD was observed by TEM and a typical image of HPT deformed Ni is shown in Fig. 1. The visible strongly non-uniform spatial distribution of dislocation, i.e. distorted regions along GBs and dislocation-free

grain interiors, was found by TEM in UFG Cu and Fe as well. Thus, we conclude that the τ_1-component in the materials under study comes from positrons trapped at dislocations situated in the distorted regions along GBs. On the other hand, a different kind of microstructure can be formed in other UFG materials. For example, an uniform distribution of dislocations (close to GBs as well as inside grains) was found in HPT deformed Mg-10wt.%Gd alloy [7].

The second component with lifetime τ_2 (and intensity I_2) represents a contribution of positrons trapped at clusters of vacancies called here microvoids. A large number of vacancies is created during SPD. As vacancies are mobile in the studied materials at RT [8] they diffuse to sinks at GBs, are anchored in the elastic field of dislocations, or form small vacancy clusters. The latter process leads to the creation of microvoids. This interpretation is supported by the fact that no microvoids were found in an HPT deformed Mg-10wt.%Gd alloy with homogeneously distributed dislocations [7]. A presence of such an array of closely spaced dislocations in the whole volume enhances the diffusion of vacancies to sinks (pipe diffusion), or the formation of vacancies bound to a dislocation line. As a consequence, the formation of microvoids is suppressed. The lifetime of positrons trapped in a microvoid consisting of n_V vacancies can be determined by ab-initio theoretical calculations based on the density functional theory and is available for Cu [9], Ni [10] and Fe [11]. The size (open volume) of microvoids present in the studied sample can be determined from comparison of the measured lifetime τ_2 with those calculated theoretically for various vacancy clusters. The obtained size of the microvoids in the studied samples is given in the last column of Table 1. Certainly, there is most probably a size distribution of microvoids in the studied samples and the values given (Table 1) represent a mean value. The results for Cu samples (including Cu+Al_2O_3 nano-composite, which exhibits the smallest grain size [12]) show a similar size of the microvoids (corresponding to 4-5 vacancies) independently of the method of deformation. On the other hand, substantially larger microvoids were formed in HPT deformed Ni and Fe. Hence, the size of microvoids seems to depend mainly on the mobility of vacancies, i.e. on the material in question.

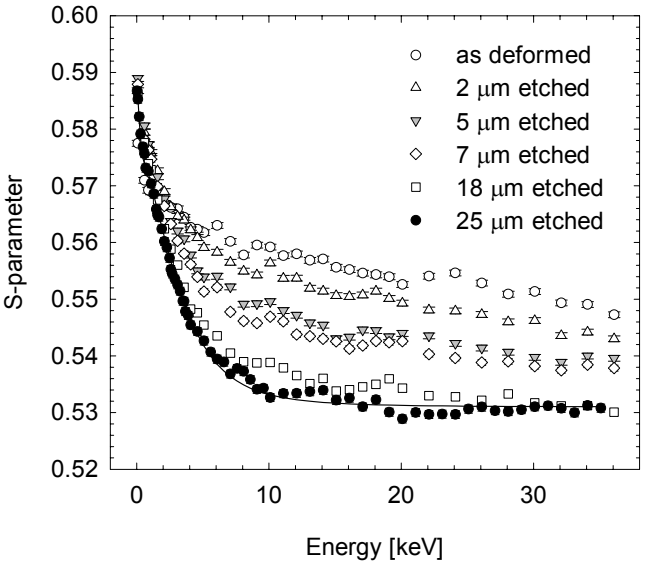

Fig.2: Dependence of annihilation lineshape parameter S on positron energy E for HPT deformed Cu (prepared using a pressure of 6GPa). The plotted curves show the effect of chemical etching. The thickness of the removed layer is given in the legend. The solid curve represents a fit by VEPFIT.

Defect Depth Profile. The S-E dependence for HPT deformed Cu is plotted in Fig. 2. The curve below ~10 keV represents a superposition of contributions from positrons annihilating inside the sample and those which diffuse back and annihilate from a surface state. Above 10 keV, almost all positrons annihilate in the bulk of the sample. One can see that S gradually decreases with E even above 10 keV. Such a behavior can be explained only by a decrease of the defect density in the studied sample with depth. The mean positron penetration depth into Cu at E = 35 keV is about 1.4 μm. In order to obtain information about the depth dependence of the defect density at a larger scale, the sample was subjected to controlled chemical etching. The S-E curves measured after each etching step, together with the thickness of the removed layer, are also shown in Fig. 2. The S value corresponding to the bulk decreases with depth thus clearly indicating a corresponding decrease of the defect density. After removal of an 18 μm thick layer, no further change of the defect density with depth is observed. It is testified not only by an insignificant change of S after the next etching

step, but also by the typical plateau-like behavior of S now observable at high values of E. A constant density of defects allows to reasonably fit the measured S-E dependence (see Fig.2). Hence, the concentration of defects decreases with depth in a surface layer of ~18 μm thickness. At higher depths the defect concentration becomes constant. This result is supported by XRD studies [10] which showed an increase of coherently scattering domain size with depth, i.e. a decrease of the volume fraction of the distorted regions with a high dislocation density.

Summary

Lattice defects created by SPD in various UFG metals were characterized. Dislocations represent a dominant type of defects. Distorted regions with high dislocation density are formed along GBs, while grain interiors are almost free of dislocations. Microvoids (small vacancy clusters) are formed inside grains. Size of microvoids depends mainly on material, but not on the way of deformation. Number of defects in HPT deformed sample decreases with depth in certain surface layer. At higher depths it becomes constant.

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* This work was financially supported by The Grant Agency of Czech Republic (contract 106/01/D049) and The Ministry of Education, Youth and Sports of Czech Republic (projects COST OC523.50 and 1K03025).