NENAMAT

International Conference

Proceedings

NANO '05

NETWORK FOR NANOSTRUCTURED MATERIALS OF ACC

NENAMAT

November 8 - 10, 2005

Brno, Czech Republic

NANO ´05
International conference

Proceedings volume

Edited by: Pavel Šandera
Published by: Brno University of Technology
Faculty of Mechanical Engineering

First edition
December 2005

Number of copies : 200

Printed by: Tisk Auda, Bezručova 17a, Brno
ISBN 80-214-3085-0

Ultra Fine-grained Mg-Based Alloys Prepared by High-pressure Torsion: Positron Lifetime Study

Jakub Čížek[1], Ivan Procházka[1*], Bohumil Smola[1], Ivana Stulíková[1], Radomír Kužel[1], Zdeněk Matěj[1], Viktoriya Cherkaska[1], Rinat K. Islamgaliev[2] and Olya Kulyashova[2]

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

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

*ivanp@mbox.troja.mff.cuni.cz

Keywords: Ultra Fine-grained Metals, Magnesium-based Alloys, High-pressure Torsion, Dislocations, Positron Lifetime.

Abstract. Microstructure of the ultra fine-grained (UFG) Mg and Mg-9.33wt.%Gd (Mg10Gd) was investigated by means of positron lifetime spectroscopy combined with TEM and XRD. The specimens were prepared by high-pressure torsion (HPT). A binomial structure was identified in the HPT-deformed Mg, which contained (1) deformed regions with a UFG structure and a high density of dislocations and (2) recrystallised regions with a low dislocation density and a larger grain size. This indicates a dynamic recovery is taking place already during the HPT processing. A homogeneous UFG structure with mean grain size of ≈100 nm and a high density of homogeneously distributed dislocations inside grains was observed on the HPT-deformed Mg10Gd. The development of the microstructure and defects recovery with increasing temperature was investigated in isochronal annealing experiments. A recovery of dislocations is accompanied by grain growth in the HPT-deformed Mg, but not in the HPT-deformed Mg10Gd. The precipitation sequence in the HPT-deformed Mg10Gd was found to differ from that found in the coarse-grained alloy.

Introduction

Magnesium-gadolinium alloys are promising light hardenable materials showing a high creep resistance even at elevated temperatures [1]. Despite of a favourable strength and a good thermal stability, however, a potential of magnesium-based alloys for industrial applications is yet limited due to a low ductility of these alloys. On the other hand, a number of ultra fine-grained (UFG) metals are known to exhibit a very high strength combined with a significant ductility [2]. Bulk UFG metals with a mean grain size of ≈ 100 nm can be produced by means of high-pressure torsion (HPT) [2]. Contrary to the ordinary coarse-grained polycrystals, the volume fraction of grain boundaries (GB) becomes significant in the UFG metals. A huge amount of open-volume defects (e.g. vacancies, dislocations) is also introduced by HPT. These defects, obviously, play a key role in the formation of UFG structures. Detailed defect studies appear thus to be extremely important for understanding the microstructure of UFG materials.

The present work is focused on the microstructure investigation of the HPT deformed Mg and Mg-9.33wt.%Gd alloy (Mg10Gd) and its development with temperature. Since positron lifetime (PL) spectroscopy is a high-sensitive non-destructive technique enabling the characterisation of open-volume defects in metals [3], PL measurements were chosen as the principal experimental tool in the present work. The PL measurements were combined with complementary techniques: transmission electron microscopy (TEM) and X-ray diffraction (XRD). In this paper, PL results are stressed. A detailed description of experiments of Mg-based alloys and discussion of results is published elsewhere [4-6].

Experiment

Specimens of pure Mg (technical purity) and Mg10Gd were investigated in the present work. The Mg10Gd alloy was prepared by sqeeze casting from technically pure Mg. The as-cast alloy was subjected to a homogenising anneal (500°C for 6 hours) followed by a rapid quenching. UFG specimens were prepared from the as received Mg and the homogenised Mg10Gd alloy by HPT at room temperature. The specimens were deformed up to true logarithmic strain $\varepsilon = 7$ under high pressure of 6 GPa [2]. The HPT-deformed specimens were disc-shaped with diameter of 12 mm and thickness of 0.3 mm. Annealing was performed at the effective heating rate of 1°C/1 min in argon protective atmosphere. Each annealing step was followed by a rapid quenching.

Positron source for PL measurements was a drop of ≈1.5 MBq of carrier-free ^{22}Na carbonate (iThemba) dried and sealed between MylarC foils (DuPont) of 2.5 μm thickness. A fast-fast PL spectrometer similar to that described in Ref. [4] was employed in the present work. The spectrometer exhibited time resolution of 170 ps and coincidence count rate of 100 s^{-1} for the above positron source. PL spectra were decomposed into four exponential components using a maximum likelihood procedure [4]. The two of these components (10 % of total intensity) belonged to positron annihilations in the source.

The TEM observations were carried out on a JEOL 2000 FX electron microscope operating at 2000 kV. The XRD measurements were performed using XRD7 and HZG4 (Seifert-FPM) powder diffractometers with Cu K_α radiation.

Results and discussion

HPT-deformed Mg. TEM observations on HPT-deformed Mg are illustrated in Fig. 1, part A. Two different kinds of regions can be seen on the Figure: (1) deformed regions of UFG structure with grain size of 100÷300 nm and a high dislocation density and (2) recrystallised regions with substantially larger grains (1÷5 μm) and almost free of dislocations. A partial dynamic recovery of the UFG structure occurring already during HPT processing is indicated by the presence of the latter kind regions. PL spectrum of HPT-deformed Mg was composed of two exponential components given in Table 1. The shorter lifetime arises from annihilations of the free positrons, while the longer one should be attributed to annihilations of the positrons trapped at dislocations. Such an interpretation is based on the good agreement of the latter lifetime with that found in cold-rolled Mg for positrons trapped at dislocations [6]. Thus, one can conclude that positrons are trapped at dislocations inside the deformed regions.

HPT-deformed Mg10Gd alloy. TEM results obtained on HPT-deformed Mg10Gd alloy are shown in Fig. 1, part B. A uniform UFG microstructure was observed with grains of size of ≈100 nm, a high dislocation density distributed homogeneously inside grains and high-angle misorientation of neighbouring grains. Thus, contrary to the above pure Mg case, no dynamic recovery of the UFG structure took place during HPT processing. A high dislocation density could also be deduced from a significant broadening of the XRD profiles described in [6] in details. Similarly to the HPT-deformed Mg, two components were resolved in the PL spectrum of the HPT-deformed Mg10Gd, see Table 1. The shorter lifetime corresponds to the free positrons, while the

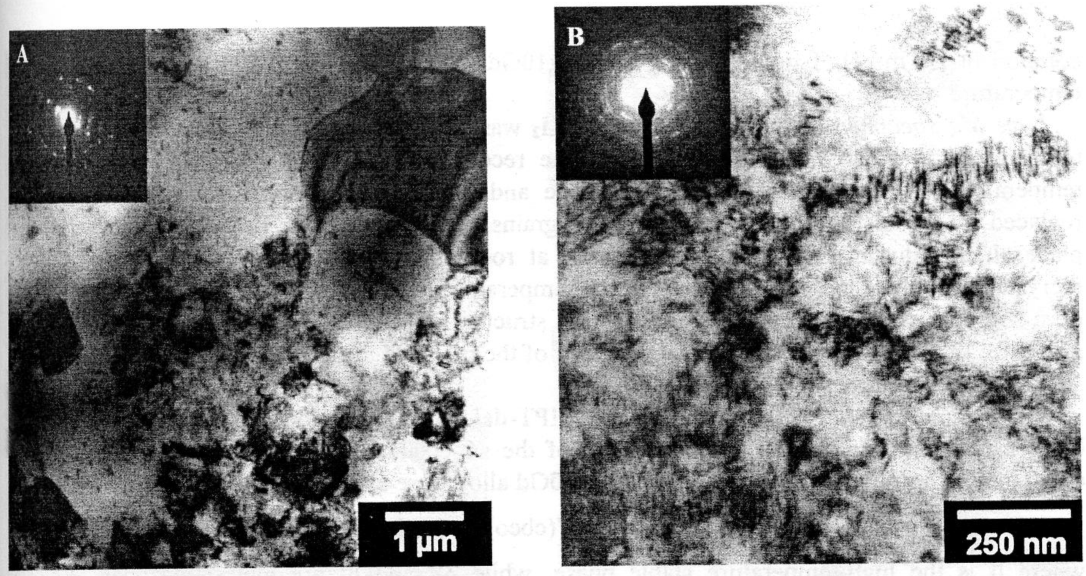

Fig. 1. Bright-field TEM images and electron diffraction patterns of the HPT-deformed specimens: A - pure Mg, B – Mg10Gd.

longer one originates in annihilations of the positrons trapped at dislocations. Compared to the HPT-deformed cubic metals Cu, Fe and Ni, see [8,9] and references therein, two differences should be pointed out in the UFG Mg10Gd alloy which shows the hexagonal structure. (1) Homogeneous distribution of dislocations over the grain interior appears in Mg10Gd, while in the cubic metals dislocations are concentrated in distorted regions along grain boundaries and grain interiors remain almost free of dislocations. (2) No microvoids could be detected inside grains in hexagonal Mg10Gd. Both these differences are obviously related to the hexagonal structure, which hinders dislocation movement towards grain boundaries. The dislocations remaining distributed in the grain interior then limit vacancy migration and, consequently, formation of microvoids.

Table 1. Lifetimes τ_i and relative intensities I_i (i=1,2) of the exponential components observed in PL spectra of Mg and Mg10Gd in the present work[a]

Sample	τ_1 (ps)	I_1 (%)	τ_2 (ps)	I_2 (%)
HPT deformed Mg	188(5)	39(1)	257(3)	61(1)
HPT deformed Mg-10Gd	210(3)	34(2)	256(3)	66(2)

[a] The errors (one standard deviation) are given in parentheses in units of the last significant digit. Normalisation $I_1+I_2=100\%$ is used.

Isochronal annealing of Mg and Mg10Gd specimens. PL spectra were measured after each annealing step. Two components corresponding to the free positrons and the positrons trapped at dislocations were resolved at all annealing temperatures. Since the dislocation lifetime did not exhibited meaningful changes with annealing temperature, the nature of positron traps remains unchanged during annealing. Therefore, dislocation lifetime was kept a constant independent of the annealing temperature in the evaluation of PL spectra. Relative intensity I_2 of the dislocation component for the HPT-deformed Mg and Mg10Gd is plotted in Fig. 2 as a function of annealing temperature.

Pure Mg specimen. A dramatic decrease in I_2 was observed for HPT-deformed Mg from room temperature to 220°C. This indicates that the recovery of dislocations starts already at room temperature. The regions with UFG structure and a high density of dislocations are gradually replaced by the dislocation-free recrystallised grains. As shown above, dislocation recovery takes place already during HPT deformation made at room temperature. The volume fraction of the recrystallised grains increases with increasing temperature and, eventually, at 200°C the specimen of pure Mg consists of a completely recrystallised structure with a low dislocation density and a mean grain size of ≈5 μm. The recrystallised structure of the pure Mg specimen after annealing at 200°C is shown in Figure 3, part A.

Mg10Gd specimen. Precipitation effects in HPT-deformed Mg10Gd alloy can be followed on the basis the data in Fig. 2. The decomposition of the supersaturated solution (sss) and precipitaion effects in the homogenised coarse-grained Mg10Gd alloy were established elsewhere [1,6] as

$$sss \rightarrow \beta''(D019) \rightarrow \beta'(cbco) \rightarrow \beta(Mg_5Gd, cubic),$$

where β is the high-temperature stable phase, while β'' and β' are metastable ones. A radical decrease in I_2 takes place in the temperature interval 100÷240°C (see Fig. 2), which indicates a significant recovery of dislocations in this temperature range. A slight local increase in I_2 around 100°C is tentatively due to formation of tiny β'' particles. A bright-field TEM image of HPT-deformed Mg10Gd specimen after annealing at 260°C is shown in Fig. 3, part B. An apparent

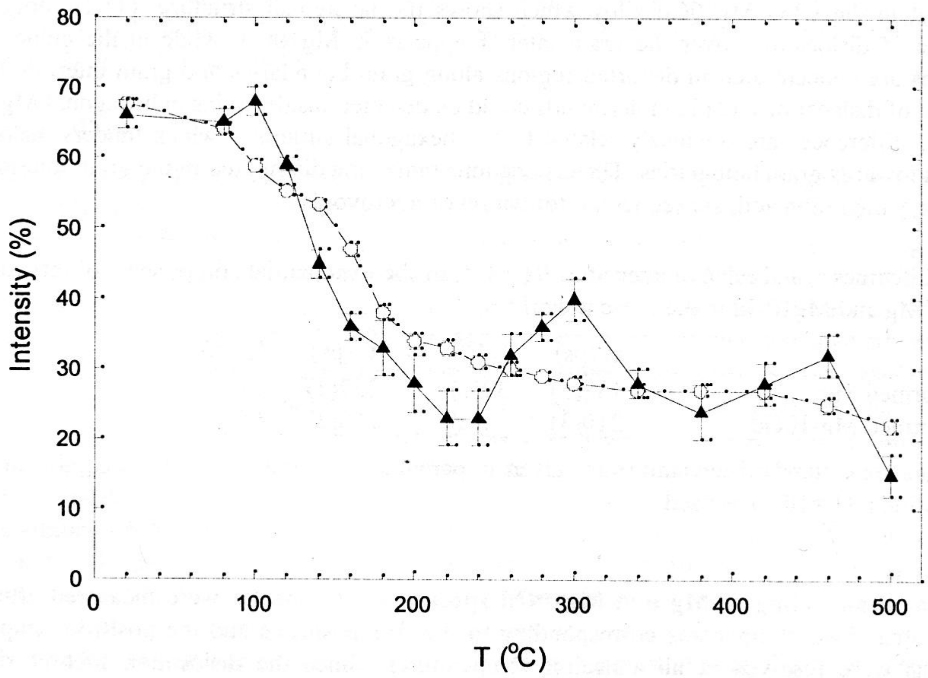

Fig. 2. The dependence of the relative intensity I_2 of positrons trapped at dislocations on annealing temperature T for HPT-deformed Mg (open circles) and HPT deformed Mg10Gd (full triangles).

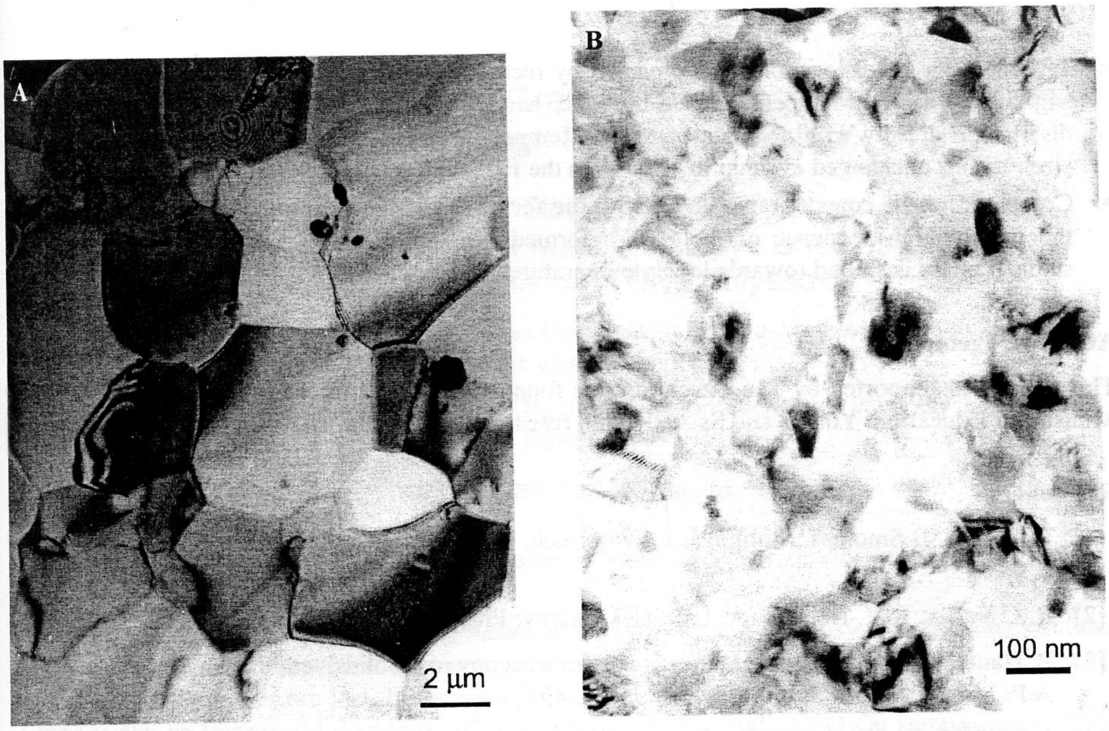

Fig. 3. Bright-field TEM images of annealed specimens: A – pure Mg annealed at 200°C, B – Mg10Gd annealed at 260°C.

decrease in dislocation density is seen in the Figure. On the other hand, the grain size remains to be ≈100 nm. It means that, opposite to the HPT-deformed Mg, the dislocation recovery is not accompanied by grain growth in the Mg10Gd. As shown by TEM, the mean grain size remains unchanged even up to ≈300°C in the HPT-deformed Mg10Gd. Thus, a very good thermal stability of the UFG structure in the HPT-deformed Mg10Gd is demonstrated. A local maximum of I_2 is observed at 300°C (Fig. 2). It can be attributed to the precipitation of the equilibrium β phase. Positrons are trapped at the misfit defects between the incoherent β phase particles and the matrix. The β phase particles were identified by TEM. Contrary to the coarse-grained alloy, therefore, precipitation of the metastable β' phase is absent in annealing of the HPT-deformed Mg10Gd alloy. The formation of the stable β phase starts at a significantly lower temperature. A subsequent decrease in I_2, seen in Fig. 2, is caused by a coarsening of the β phase particles. Above 450°C, β phase particles are dissolved and the solid solution is restored.

Summary

The present results of microstructure investigations of HPT-deformed Mg and Mg10Gd alloy and its development during isochronal annealing can be summarised as follows:

- An incomplete dynamic recovery of dislocations takes place during HPT processing of the Mg specimen, which leads to a binomial kind of structure.
- The HPT-deformed Mg10Gd exhibits a homogeneous UFG structure with a mean grain size of ≈100 nm and homogeneously distributed dislocations inside grains.

- The recovery of dislocations accompanied by recrystallisation is present in the HPT-deformed Mg already at room temperature. On the other hand, Mg10Gd exhibits a remarkable decrease in dislocation density with no grain growth in temperature range of 100÷240°C. The mean grain size remains unchanged even up to ≈300°C in the HPT-deformed Mg10Gd.
- Compared to the coarse-grained Mg10Gd, the formation of the metastable β' phase is absent in the precipitation sequence of the HPT-deformed Mg10Gd alloy and the precipitation of the stable β phase is shifted towards lower temperatures.

Acknowledgements

This work was supported by The Czech Science foundation under project 106/05/0073 and by The Ministry of Education, Youths and Sports within research plan MS 0021620834.

References

[1] P. Vostrý, B.Smola, I. Stulíková, F. von Buch, B.L. Mordike: phys. stat. sol. (a) 175 (1999), p. 491

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

[3] P. Hautojärvi, C. Corbel: in Positron Spectroscopy of Solids, ed. by A. Dupasquier and A.P. Mills, Jr. (IOS, Amsterodam 1995), p. 491

[4] J. Čížek, I. Procházka, B. Smola, I. Stulíková, R. Kužel, Z. Matěj, V. Cherkaska, R.K. Islamgaliev, O. Kulyasova: Mater. Sci. Forum Vol. 482 (2005), p. 183

[5] J. Čížek, I. Procházka, B. Smola, I. Stulíková, R. Kužel, Z. Matěj, V. Cherkaska, R.K. Islamgaliev, O. Kulyasova: Acta Phys. Polonica Vol. 107 (2005), p. 738

[6] J. Čížek, I. Procházka, F. Bečvář, B. Smola, I. Stulíková, R. Kužel, V. Cherkaska, R.K. Islamgaliev, O. Kulyasova: physica status solidi (a) (2005), submitted for publication

[7] F. Bečvář, J. Čížek, L. Lešťák, I. Novotný, I. Procházka, F. Šebesta: Nucl. Instr. Meth. A Vol.443 (2000) p. 557

[8] J. Čížek, I. Procházka, B. Smola, I. Stulíková, R. Kužel, M. Cieslar, Z. Matěj, V. Cherkaska, G. Brauer, W. Anwand, R.K. Islamgaliev, O. Kulyasova: Mater. Sci. Forum Vol. 482 (1992), p. 207

[9] J. Čížek, I. Procházka, B. Smola, I. Stulíková, R. Kužel, M. Cieslar, Z. Matěj, V. Cherkaska, G. Brauer, W. Anwand, R.K. Islamgaliev, O. Kulyasova: Acta Phys. Polonica Vol. 107 (2005), p. 745