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Magnetic Properties of Nanocrystalline $Fe_{83}Sn_5B_{12}$ Alloys after Electron Irradiation of Precursor

J. Sitek[a,*], D. Holková[a], B. Butvinová[b], P. Butvin[b] and J. Dekan[a]

[a]Institute of Nuclear and Physical Engineering, Faculty of Electrical Engineering and Information Technology, Slovak University of Technology, Ilkovičova 3, 812 19 Bratislava, Slovakia

[b]Institute of Physics, Slovak Academy of Science, Dúbravská cesta 9, 845 11 Bratislava, Slovakia

Precursor of nanocrystalline $Fe_{83}Sn_5B_{12}$ alloy was irradiated by electron beams of doses up to 4 MGy. Irradiation affected the magnetic microstructure of the amorphous and nanocrystalline alloy. This was manifested as a change in the direction of the net magnetic moment, intensity of the internal magnetic field, and volumetric fraction of the constituent phases. All these parameters were determined from the measured Mössbauer spectra. The direction of the net magnetic moment was the most sensitive parameter. Structural changes were identified by Mössbauer spectroscopy as the ratio of the amorphous to the crystalline component. Irradiation of the amorphous precursor has been reflected also at the shape and parameters of hysteresis loop. The results indicates the changes of the microscopic magnetic parameters due to electron irradiation.

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1. Introduction

In the recent years, nanocrystalline alloys have become attractive for many applications. The most prominent FINEMET, NANOPERM, and HITPERM-type alloys have been frequently investigated, because they exhibit excellent soft magnetic properties. It has been already shown [1, 2] that some physical properties of the nanocrystalline materials can be (more or less) affected by neutron irradiation. The particle bombardment produces defects that may cause a realignment of magnetic domains implying a reorientation of the magnetic moments. Changes in the local neighbourhoods of atoms affect the average hyperfine magnetic field, as well as the shape of the hyperfine field distributions. Recently, soft magnetic alloys NANOMET were developed as FeSiBCu and FeSiBPCu alloys [3]. It was also found that substitution of Co for Fe improved the soft magnetic properties and thermal stability of the nanocrystalline alloys [4, 5].

It is know that the crystallization process of the amorphous precursor is controlled by thermal annealing. It has already been shown that nanocrystalline structure could be (more or less) affected by different external factors [6], among them also electron irradiation is used [7].

Amorphous $Fe_{95-x}Sn_5B_x$ ribbons, $(x = 15, 17, 20)$, and formation of crystalline phases and their morphology were analyzed by X-ray diffraction and transmission electron microscopy [8]. The results indicated the existence of critical composition for the transformation kinetics. The relation between the magnetic state of the material and the kinetics of crystallization was also analyzed.

In turn, the effect of electron irradiation on the crystallization of NANOMET alloys was studied in [9, 10]. In this case final properties of nanocrystal were dependent on the former irradiation of amorphous precursor by electrons. Therefore, our work is focused on tracing structural and local magnetic modifications induced in alloys by electrons irradiation. The method of Mössbauer spectroscopy and magnetic measurements allowed to inspect the changes in the orientation of local magnetization in the value of the magnetic hyperfine field, as well as in the volumetric fraction of the crystalline, and the amorphous components of the nanocrystalline alloy after irradiation of $Fe_{83}Sn_5B_{12}$ amorphous precursor.

2. Experimental technique

Ribbon-shaped specimens of the master alloy were prepared by planar flow casting method. The ribbons were about 22 μm thick and 7 mm wide. Electron irradiation of amorphous precursor was performed at Slovak Medical University Trenčín using a linear accelerator with a dose up to 4 MGy and energy of 5 MeV. The applied dose density of irradiation corresponds to 1.746×10^{16} e/cm^2. Annealing was carried out in vacuum furnace for 30 min at 400 °C. Mössbauer spectra were measured in transmission geometry using a standard constant acceleration spectrometer with a ^{57}Co (Rh) source. All spectra were recorded at room temperature and evaluated using the CONFIT program [11], hence enabling simultaneous treatment of crystalline components and residual amorphous phases using individual lines and distributions of

*corresponding author; e-mail: jozef.sitek@stuba.sk

hyperfine parameters. Hysteresis loops were recorded using a digitizing hysteresis graph at standard ac (frequency 21 Hz) sinusoidal field H excitation in Helmholtz drive coils in longitudinal direction on 10 cm long strips.

3. Results and discussion

Mössbauer spectra of amorphous sample were fitted with two distribution functions corresponding to magnetic structure composed of high and low field component. Mössbauer spectra of the nanocrystalline samples were evaluated using a fitting model comprising two component groups, i.e., the first one consists of narrow lines attributed to the Fe-atoms situated in the bulk of nanocrystalline grains, and the second group describes the amorphous rest with two magnetic distributions of hyperfine magnetic field (low-field and high-field distributions). Mössbauer spectrum of the amorphous on-irradiated sample recorded at room temperature is shown in Fig. 1a. Qualitatively, the Mössbauer spectrum of irradiated amorphous sample did not change much, although changes in some parameters given in Table I could be observed. In fact, the previous measurements on samples after electron irradiation [9, 10] reported changes in the mean orientation of local magnetization, value of the average hyperfine magnetic field of the amorphous and crystalline components and of their volumetric fraction. Electron irradiation has an influence on the distribution of internal magnetic field described by $p(B)$ distribution function, as shown in Fig.1b. Mean orientation of local magnetization is reflected by the ratio of the second (and fifth) to the third (and fourth) line intensities of a sextet in Mössbauer spectrum (A_{23}). This parameter achieves its maximum value if the mean local magnetization lays in the ribbon plane. However, when A_{23} is oriented perpendicular to the ribbon plane, its value is the minimal.

An irradiation by electrons leads to redistribution of atoms in amorphous matrix. In our case, parameter A_{23} is the most sensitive, which behavior points on some changes of the mean orientation of local magnetization. The value of A_{23} parameter obtained for investigated samples are listed in Table I.

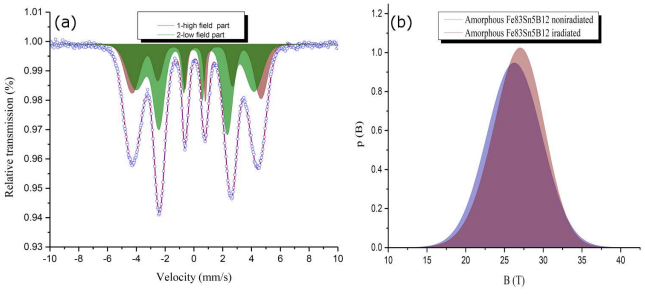

Fig. 1. (a) Mössbauer spectrum of non-irradiated amorphous alloy of $Fe_{83}Sn_5B_{12}$. (b) Distribution $p(B)$ of internal magnetic field non-irradiated and irradiated amorphous alloy.

TABLE I

Parameters of Mössbauer spectra: parameter A_{23}, internal magnetic induction of amorphous component B_{am}, concentration of amorphous component A_{am}, internal magnetic induction of crystalline component B_{cr}, concentration of crystalline component A_{cr}.

Stage	A_{23}	B_{am} [T]	A_{am} [%]	B_{cr} [T]	A_{cr} [%]
amorphous non-irrad	2.40	26^h 25^l	68 32		
amorphous irrad	3.61	27^h 25^l	56 44		
nanocryst. non-irrad	1.80	24	88	33.1	12
nanocryst. irrad	2.30	26	68	32.4	32

^{h}high field part, ^{l}low field part

Our measurements showed that the mean orientation of local magnetization turns to the ribbon plane. Internal magnetic field of amorphous component consists of low and high field part. We observed also relative changes of these two components as is given in Table I.

Mössbauer spectra of non-irradiated and irradiated amorphous precursor by electrons after thermal treatment recorded at room temperature are shown in Fig. 2. After thermal treatment of non-irradiated sample, the alloy transformed in nanocrystalline state in a standard composition. This crystalline component was identified according to value of internal magnetic field as a metallic iron. Error of the concentration and the internal magnetic field was estimated at 3% and at 0.5 T, respectively.

After thermal treatment of irradiated samples, the alloy transformed to nanocrystalline state. We observed quantitative difference in the relative amount of amorphous and crystalline components, see Table I. Our measurements show that crystalline component increased about 20% and the mean orientation of local magnetization turns to the ribbon plane. These results confirm that irradiation of the amorphous precursor before thermal treatment has an influence at the final nanocrystalline state.

Electron irradiation of amorphous precursor has reflected in hysteresis loop, as shown in Fig. 3.

The effect of electron irradiation on the magnetic properties of the studied ribbons was investigated by measuring hysteresis loops, from which we evaluated the values of coercivity H_c, and maximum magnetic polarization J_s. The exciting field that we used turned to be not enough to saturate the samples. The amorphous sample showed a relatively low coercivity value of 50 A/m, that dropped to 48 A/m after irradiation.

The coercivity increased by more than 10 times after annealing at 400 °C. This is related to the change in microstructure and the formation of medium nanosized grains ($\sim$ 35 nm by the Scherrer formula of XRD). The shape of the hysteresis loop after irradiation in the amorphous state exhibited a slight reduction of

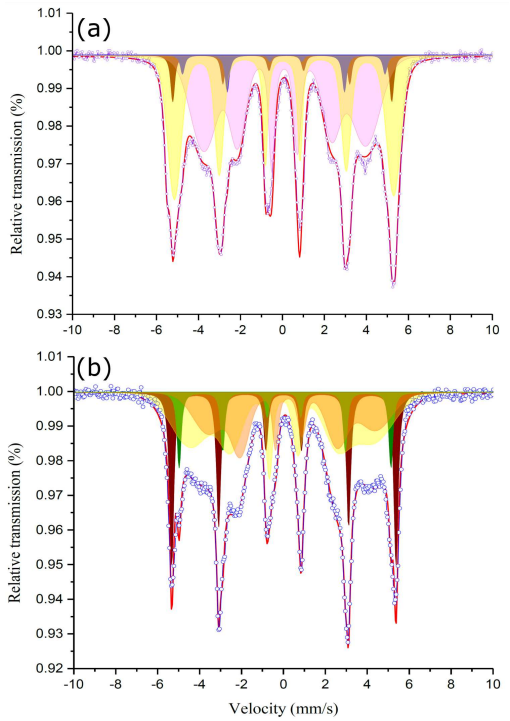

Fig. 2. (a) Mössbauer spectrum of non-irradiated, and (b) irradiated nanocrystalline of $Fe_{83}Sn_5B_{12}$.

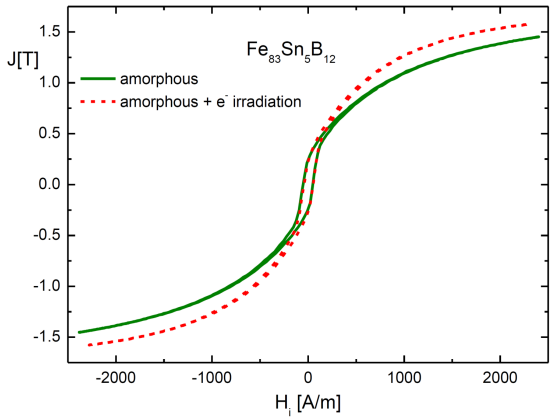

Fig. 3. Hysteresis loops of amorphous sample before and after irradiation.

off-axis anisotropy, and thus better approach to saturation (J_s increases from 1.44 T to 1.58 T). We attribute this effect to structural relaxation, a decrease in internal stresses in the ribbon which is reflected by the decrease in the magnetic anisotropy, and the orientation of the easy direction of magnetization along the ribbon axis. The fact that parameter A_{23} increased after irradiation, also indicates a change in the orientation of local magnetization to the ribbon plane. The nanocrystallization was manifested in the loops: although the coercivity increases, approach to saturation significantly improves, reaching $J_s = 1.72$ T of non-irradiated, as well as of irradiated sample.

4. Conclusions

Nanocrystalline $Fe_{83}Sn_5B_{12}$ alloy after electron irradiation of amorphous precursor of the doses up to 4 MGy has been modified, as indicated by the mean orientation of local magnetization, volumetric fraction of amorphous and crystalline component, and induction of internal magnetic field. Electron irradiation caused small structural changes that were reflected in magnetic structure parameters. In the case of nanocrystalline alloys, which consists of crystalline nanograins embedded in an amorphous intergranular matrix, an irradiation by electrons led to redistribution of atoms in the amorphous matrix, disturbance of regular atomic ordering of the crystal lattice, and atom exchange between the amorphous and crystalline component. These phenomena were also reflected in the parameters of hysteresis loop.

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References

[1] J. Sitek, J. Dekan, M. Pavlovič, *Acta Phys. Pol. A* **126(1)**, 84 (2014).

[2] T. Nagase, Y. Umakoshi, N. Sumida, *Sci. Technol. of Adv. Mater.* **3**, 119 (2002).

[3] A. Makino, H. Men, T. Kubota, K. Yubuta, A. Inoue, *Mater. Trans.* **50**, 204 (2009).

[4] R. Xiang, S. Zhou, B. Dong, G. Zhang, Z. Li, Y.Wang, Ch. Chang, *Prog. Nat. Sci. Mater. Int.* **24**, 649 (2014).

[5] I. Janotová, J. Zigo, P. Švec, I. Maťko, D. Janičkovič, P. Švec Sr., *J. Alloys Compd.* **643**, 265 (2015).

[6] J. Sitek, J. Degmová, K. Sedlačková, J. Dekan, *J. Mod. Phys.* **3**, 274 (2012).

[7] Y-Q. Chang, Q. Guo, J. Zhang, L. Chen, Y. Long, F-R. Wan, *Front. Mater. Sci.* **7(2)**, 143 (2013).

[8] E.Ileková, I.Maťko, P.Švec, P.Švec jr., D. Janíčkovič, *J. Alloys Compd.* **5005**, 546 (2011).

[9] K. Shimizu, M. Nishijima, A. Takeuchi, T. Nagase, H. Yasuda, A. Makimo, *J. Jpn. Inst. Metals* **78**, 364 (2014).

[10] J. Sitek, D. Holková, P. Novák, J. Dekan, *Acta Phys. Pol. A* **131(4)**, 708 (2017).

[11] T. Žák, Y. Jirásková, *Surf. Interface Anal.* **38**, 710 (2006).