Proceedings of the 16th Czech and Slovak Conference on Magnetism, Košice, Slovakia, June 13–17, 2016

# Thickness Dependence of Mössbauer Parameters for Fe<sub>78</sub>Si<sub>9</sub>B<sub>13</sub> Metallic Glass Ribbons

N. AMINI<sup>*a,b,\**</sup>, M. MIGLIERINI<sup>*a,c*</sup>, M. HASIAK<sup>*d*</sup>, J. DEKAN<sup>*a*</sup> AND S. HABIBI<sup>*b*</sup>

<sup>a</sup>Slovak University of Technology in Bratislava, Faculty of Electrical Engineering and Information Technology,

Institute of Nuclear and Physical Engineering, Ilkovičova 3, 812 19 Bratislava, Slovakia

<sup>b</sup>Department of Physics, Bu-Ali Sina University, 65174-4161, Hamedan, Iran

<sup>c</sup>Department of Nuclear Reactors, Czech Technical University in Prague,

V Holešovičkách 2, 180 00 Prague, Czech Republic

<sup>d</sup>Department of Mechanics and Materials Science, Wrocław University of Science and Technology,

M. Smoluchowskiego 25, 50-370 Wrocław, Poland

Changes in magnetic microstructure of  $Fe_{78}Si_9B_{13}$  ribbons were investigated by the Mössbauer spectrometry as a function of their thickness. The latter decreases inversely with the quenching wheel velocity. Amorphicity of the samples was checked by X-ray diffraction and the Mössbauer spectrometry. Average values of hyperfine magnetic fields do not appreciably differ with the production velocity. On the other hand, notable deviations are observed in the position of a net magnetic moment. After annealing with moderate temperature, the magnetic anisotropy improves as indicated by an increase of the average value of hyperfine magnetic field. However, magnetic moments turn out of the ribbon plane and this process continues towards higher production velocity, i.e., smaller thickness of the ribbons. No appreciable deviations in the investigated Mössbauer parameters were revealed after annealing during different time intervals.

DOI: 10.12693/APhysPolA.131.666

PACS/topics: 75.50.Kj, 75.60.Ej, 75.75.-c, 76.80.+y

#### 1. Introduction

Iron-based metallic glasses (MGs) obtained by meltspinning technique exhibit interesting soft magnetic properties that are studied already for decades [1–4]. Ternary Fe–Si–B metallic glass (MG) systems have a special position among other MGs due to their excellent magnetic, electrical and mechanical properties that still attract attention of the researchers [5]. In the investigation of MGs, the Mössbauer spectrometry is frequently applied [1–6]. This method was used also to study correlations between quenching wheel velocity and structural properties of several types of MGs [7, 8]. Other methods of investigations including theoretical calculations were also employed [9, 10].

Continuing interest in a classical  $Fe_{78}Si_9B_{13}$  MG can be documented for example by a recent work [11] in which this system was prepared under different quenching conditions and investigated from the point of view of its microstructure, magnetism, and electrochemical properties employing various techniques. Unlike for other MGs, the effects of quenching conditions upon the Mössbauer hyperfine parameters were, to our knowledge, not reported so far for this system.

In this paper, we present the results of the Mössbauer spectrometry investigations of  $Fe_{78}Si_9B_{13}$  MG ribbons. We concentrate on the effect of quenching velocity upon hyperfine parameters obtained from as-quenched alloys

as well as from ribbons annealed at a moderated temperature (i.e., below the onset of crystallization) for different time intervals. In particular, deviations in magnetic microstructure and spin texture are discussed. The obtained data were correlated with the results of magnetic measurements. These are, however, not reported here.

## 2. Experimental details

Iron-based MG with the nominal composition of  $Fe_{78}Si_9B_{13}$  was fabricated by a melt spinning method in argon atmosphere in a form of ribbons using various tangential disk velocities namely 15, 20, 25, 30, 35, and 40 m/s. The resulting thickness of the samples was measured by a micrometre with the accuracy of 0.001 mm. Structural properties of the samples were checked by Xray diffraction (XRD) using Bruker D8 Discover diffractometer employing filtered Cu  $K_{\alpha}$  radiation.

In order to study spin texture in the obtained ribbons the Mössbauer measurements have been used. <sup>57</sup>Fe Mössbauer spectra were collected at room temperature in transmission geometry using a conventional constantacceleration spectrometer equipped with a <sup>57</sup>Co  $\gamma$ -ray source in a rhodium matrix. The spectra were analysed by a least-square fitting procedure using the NORMOS fitting software. They were evaluated by Lorentzian line sextets assuming model independent distributions of hyperfine magnetic fields P(B). Linewidths of the individual sextets were fixed to the value obtained from calibration and their relative line ratios were let free during the fitting.

Annealing of the prepared samples was carried out at  $450 \,^{\circ}$ C for 60 min in Ar protective atmosphere. The sam-

<sup>\*</sup>corresponding author; e-mail: phn.amini@basu.ac.ir

ple prepared with the highest velocity was annealed also for different time intervals.

### 3. Results and discussion

The effect of quenching disk velocity upon the thickness of the produced MG ribbons is depicted in Fig. 1. The thickness (d) decreases monotonically as the disk velocity (v) increases. It satisfactorily obeys the d = 1/v formula.



Fig. 1. Thickness of the MG ribbons plotted against velocity of the quenching wheel.



Fig. 2. Mössbauer spectrum of as-quenched ribbon prepared with the velocity of 15 m/s (a) and corresponding P(B) distribution (b).

Amorphicity of the samples was checked by XRD and the Mössbauer spectrometry measurements and no signs of crystallites were revealed. All samples exhibit Mössbauer patterns that are typical for amorphous ferromagnetic material as seen in Fig. 2 where a spectrum obtained for as-quenched ribbon produced with the velocity of 15 m/s is shown as an example. All spectra of asquenched ribbons are characterized by very similar broadened sextuplets of absorption lines. They were refined by distributions of hyperfine magnetic fields P(B). The obtained average values of P(B) distributions  $\langle B \rangle$  and line intensity ratios  $A_{23}$  of the spectra are discussed below.

Transmission Mössbauer spectra in Fig. 3 were recorded at room temperature after annealing the ribbons at 450 °C for 60 min. They show dramatic decrease of the second and the fifth line intensities towards higher preparation velocities. It is obvious that during the annealing process, amorphous ferromagnetic state is still conserved.

Parameters derived from the Mössbauer spectra of asquenched and annealed ribbons are plotted in Fig. 4 as a function of production velocity. They show dependences of average hyperfine magnetic fields  $\langle B \rangle$  obtained from P(B) distributions and  $A_{23}$  parameters. The latter represents a ratio of the second and fifth to the third and fourth Mössbauer spectral line intensities (areas). They can acquire values between 0 and 4.  $A_{23} = 0$  indicates that the magnetic moments of the resonant <sup>57</sup>Fe nuclei are positioned perpendicular to the plane of a ribbonshaped sample while  $A_{23} = 4$  means that all magnetic moments are situated in the ribbon plane. For random orientation of the spins  $A_{23} = 2$ .



Fig. 3. Transmission Mössbauer spectra (a) and corresponding distributions of hyperfine magnetic fields P(B) (b) of the samples annealed at 450 °C for 60 min. The particular preparation velocities (in m/s) are indicated in the figure.

Average hyperfine magnetic fields  $\langle B \rangle$  derived from P(B) distributions of the as-quenched samples in Fig. 4a are practically independent of the production velocity within the error range. On the other hand, the parameter  $A_{23}$  in Fig. 4b exhibits significant deviations. In the 15 m/s sample,  $A_{23} = 2.49$  implies not far off random spin arrangement. In the very next sample namely 20 m/s, the magnetic moments are already positioned close to the ribbon plane  $(A_{23} = 3.53)$ . Further on,  $A_{23}$  systematically decreases towards higher velocities which indicates that the magnetic moments tend to acquire again random orientations. So even though the average hyperfine magnetic field values do not substantially alter with respect to the production velocity or the ribbon thickness, positions of local magnetic moments are obviously governed by fluctuations in microstructure of the as-quenched ribbons due to quenched-in internal stresses [12].

After annealing, internal stresses are partially removed and structural relaxation takes place in the amorphous alloy. This leads to re-arrangement of the constituent atoms. As a consequence, magnetic behaviour of the annealed ribbons is affected. This is documented by  $\langle B \rangle$  which exhibits slight increase in its mean value from (24.9±0.2) T in the as-quenched state up to (25.4±0.2) T after annealing.



Fig. 4. Average hyperfine magnetic field  $\langle B \rangle$  (a) and  $A_{23}$  (b), plotted against the production velocity.

As far as the  $A_{23}$  parameter is concerned, it exhibits significant differences in comparison with the asquenched state as seen in Fig. 4b. The spins are highly textured showing predominant orientations out of the ribbon plane. With increasing velocity (decreasing thickness of the ribbons) they approach close-to-perpendicular positions. This is presumably caused by changes in the microstructural arrangement of the resonant atoms caused by annealing. As a consequence, the 2nd and the 5th Mössbauer lines in Fig. 3a nearly disappear.

The sample produced with the highest velocity of 40 m/s was annealed at 450 °C for different time intervals. The obtained Mössbauer spectra and their P(B) distributions are illustrated in Fig. 5. The obtained values of  $\langle B \rangle$  and  $A_{23}$  are of 25.5, 25.5, 25.4 T and 1.0, 0.8, 0.8, respectively, as derived from the Mössbauer spectra of the samples annealed for 60, 90, and 120 min, correspondingly. Their close similarities demonstrate that duration of the annealing time does not significantly affect these Mössbauer parameters.



Fig. 5. Mössbauer spectra (a) of the  $Fe_{78}Si_9B_{13}$  ribbon produced with the velocity of 40 m/s and annealed at 450 °C for the indicated times (in min) with the corresponding hyperfine field distributions P(B) (b).

We have performed also magnetic measurements including temperature dependences of magnetization under different external magnetic fields and cooling regimes. They have indicated changes in the selected magnetic parameters including the Curie temperature as a function of the ribbon thickness. The obtained results correlate well with the findings of the Mössbauer spectrometry and will be presented elsewhere.

## 4. Conclusions

The effect of quenching wheel velocity in the melt spinning method upon parameters of the Mössbauer spectra of  $Fe_{78}Si_9B_{13}$  MG was investigated. The sample's thickness decreases drastically as the disk velocity increases. XRD and the Mössbauer spectrometry confirmed that all ribbons are amorphous in the as-quenched state as well as after annealing at a temperature that is below the onset of crystallization. Average hyperfine magnetic fields of the as-quenched samples are independent of the production velocity even though the orientation of particular magnetic moments of the resonant Fe atoms varies.

After annealing, similar trends are observed as far as average hyperfine magnetic fields are concerned. They are slightly higher than in the as-quenched state which indicates modifications in the microstructural arrangement. The latter is demonstrated also by highly textured spin structure that was revealed after annealing. Individual magnetic moments are positioned almost perpendicular to the ribbon plane while they have acquired nearly random arrangement in the as-quenched ribbons.

The obtained alterations of the Mössbauer parameters suggest complex anisotropy distribution caused by the internal stresses induced during the quenching process as well as their variation after annealing. Extension of the time of annealing had no significant impact upon the investigated Mössbauer parameters in the thinnest ribbon.

#### Acknowledgments

This work was supported by the grants GACR 14-12449S and VEGA 1/0182/16. We thank M. Pavúk (Bratislava) for providing the thickness measurements.

#### References

- [1] H.N. Ok, A.H. Morrish, *Phys. Rev. B* **22**, 3471 (1980).
- [2] A.K. Bhatnagar, *Hyperfine Interact.* **25**, 637 (1985).
- [3] R. Hasegawa, J. Magn. Magn. Mater. 215, 240 (2000).
- [4] E. Kuzmann, S. Stichleutner, A. Sápi, L.K. Varga, K. Havancsák, V. Skuratov, Z. Homonnay, A. Vértes, *Hyperfine Interact.* 207, 73 (2012).
- [5] J.M. Cadogan, S.J. Campbell, J. Jing, C.P. Foley, P. Kater, Y.W. Mai, *Hyperfine Interact.* **226**, 7 (2014).
- [6] S.N. Kane, A. Gupta, L.K. Varga, J. Magn. Magn. Mater. 254-255, 501 (2003).
- [7] G. Roulin, J. Teillet, A. Fnidiki, B. Labulle, P. Ochin, *Hyperfine Interact.* 88, 65 (1994).
- [8] M.G. del Muro, R. Zquiak, X. Battle, J. Magn. Magn. Mater. 171, 315 (1997).
- [9] L. Gránásy, *Trans. JIM* **27**, 51 (1986).
- [10] L. Gránásy, A. Ludwig, *Mater. Sci. Eng. A* 133, 751 (1991).
- [11] L.L. Meng, X.Y. Li, J. Pang, L. Wang, B. An, L.J. Yin, K.K. Song, W.M. Wang, *J. Metal. Mater. Trans. A* 44, 5122 (2013).
- [12] S.N. Kane, N. Bhagat, A. Gupta, L.K. Varga, J. Magn. Magn. Mater. 167, 241 (1997).