14th Czech and Slovak Conference on Magnetism, Košice, Slovakia, July 6–9, 2010

Microstructural Study of Fe–Si(Ge)–Nb–Cu–B Finemet Alloys

S. Michalik[a,b,*], J. Bednarcik[b], K. Brzózka[c], P. Sovak[a] and B. Górka[c]

[a]Institute of Physics, P.J. Šafárik University, Park Angelinum 9, 041 54 Košice, Slovakia

[b]HASYLAB am DESY, Notkestraße 85, D-22503 Hamburg, Germany

[c]Department of Physics, Technical University of Radom, Krasickiego 54, 26-600 Radom, Poland

Structural changes in Finemet based alloys invoked by germanium replacement for silicon were investigated using the X-ray diffraction, the X-ray absorption spectroscopy and the Mössbauer spectroscopy. Ge substitution preserves a nanostructural character of annealed samples at temperature 550 °C; specifically the formation of a DO_3--type with about 19 at.% of (Si,Ge) was confirmed. The mean size of nanocrystals was estimated to be about 7 nm.

PACS numbers: 61.05.cp, 61.05.C−, 61.46.Hk

1. Introduction

Recently Ge has been extensively used for replacing Si in Finemet based alloys [1, 2]. Motivations is coming from the Corb model [3] which suggests that the magnetic moment of Fe atom in the bcc lattice with Ge content is greater than in the bcc lattice with the same amount of Si. In this work the authors present structural study of the influence of germanium replacement for silicon in Finemet based alloys. The study of their magnetic properties was also carried out and this one will be published later.

2. Experimental methods

Amorphous ribbons with the composition of $Fe_{73.5}Si_{13.5-x}Ge_xCu_1Nb_3B_9$ for $x = 1$, 5, 10 and 13.5, shortly referred as Ge1, Ge5, Ge10 and Ge13.5, were prepared by single-roller melt spinning technique. Samples were investigated in both as-prepared and nanocrystalline state (obtained after one hour of isothermal annealing in a vacuum at 550 °C) by means of the X-ray diffraction (XRD), the X-ray absorption spectroscopy (XAFS), the Mössbauer spectroscopy (MS) and high resolution transmission electron microscopy (HRTEM). XRD and XAFS experiments were performed using synchrotron radiation at experimental stations BW5 ($\lambda = 0.114$ Å) and X1 (at K absorption edge of Fe atoms) at HASYLAB/DESY. MS was carried out using a conventional spectrometer arranged in a vertical geometry with ^{57}Co(Rh) source.

3. Experimental results

The fully amorphous character of as-quenched samples was clearly confirmed by XRD and MS as well. From diffraction profiles of as-prepared samples structural factors were extracted and then the atomic pair distribution functions $g(r)$'s (not shown here) were calculated applying Fourier transformation [4]. The similarity of the all $g(r)$'s suggests a comparable average local atomic arrangement in all as-prepared samples. In order to obtain more precise information about local atomic structural changes due to Ge substitution we realized XAFS at Fe K edge and MS measurements. XAFS data confirmed expansion of average interatomic distances between Fe atoms and their nearest neighbours with increasing Ge content in the as-prepared samples, as it is documented in Fig. 1. It is seen that the peak position of the Fourier transformed XAFS signal is regularly shifted to higher values of interatomic distances r with Ge increase. Additionally, MS revealed 8% enhance of the mean magnetic hyperfine field (MHF) with increase of Ge content in amorphous samples (see Fig. 2).

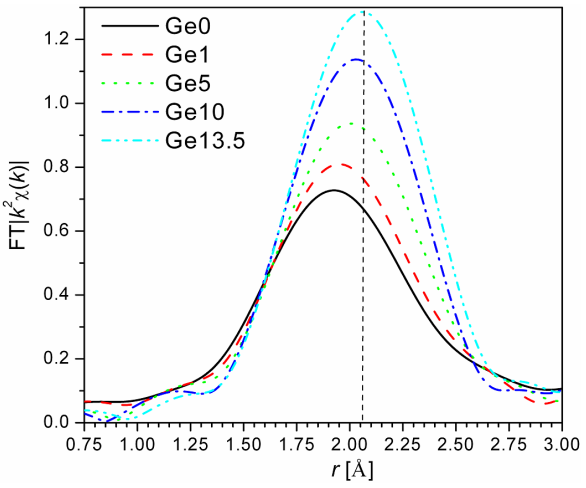

Fig. 1. Fourier transformed XAFS signals of as-quenched $Fe_{73.5}Si_{13.5-x}Ge_xCu_1Nb_3B_9$ alloys measured at the Fe K edge in a fluorescence mode.

* corresponding author; e-mail: stefan.michalik@upjs.sk

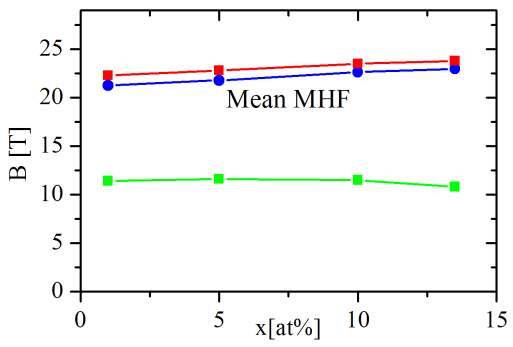

Fig. 2. Mean magnetic hyperfine field of: the whole MHF distribution (blue), the high field component (red) and the low field one (green).

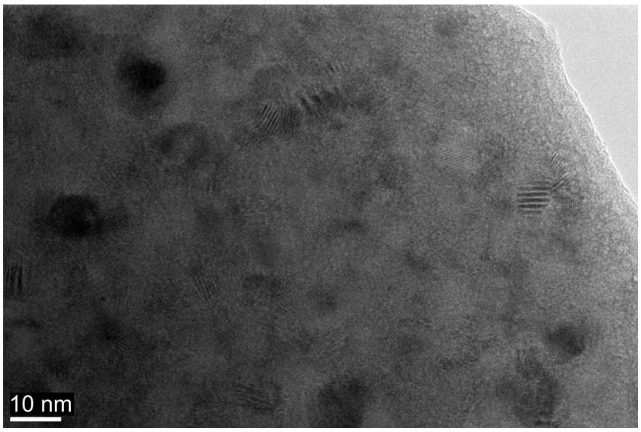

Fig. 3. Selected TEM micrograph obtained for Ge10 annealed sample.

For all samples annealed at 550 °C the XRD phase analysis confirmed the presence of the Fe_3Si phase with DO_3-type structure without any additional detection of borides. The average crystalline size of about 7 nm was determined using well-known Scherrer formula [5] and also confirmed by HRTEM (see Fig. 3). The Bragg peaks corresponding to the DO_3-type structure are progressively moved towards lower 2θ values with Ge increase. As a result the unit cell expansion is observed. This is shown in the inset of Fig. 4 (right upper corner). The lattice expansion is easily explained in the view of atomic radii as larger Ge atoms ($r_{\mathrm{Ge}} = 1.225$ Å) gradually replace smaller Si atoms ($r_{\mathrm{Si}} = 1.17$ Å) in the cubic Fe_3Si phase due to their higher concentration. The presence of the DO_3-type structure with about 19 at.% of (Si,Ge) was confirmed by MS as well. The Mössbauer spectra obtained from annealed samples show coexistence of amorphous and crystalline phases. The amorphous phase was reproduced by a continuous MHF distribution while the DO_3 structure was represented by a set of six discrete sextets corresponding to bcc–(Fe–Ge) structure (not shown here). Furthermore it was found that MHF

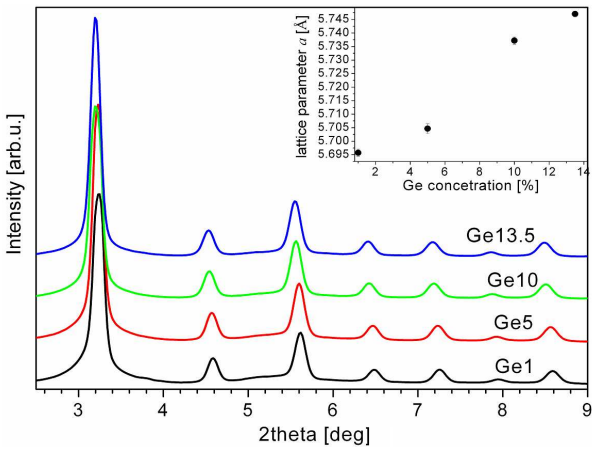

Fig. 4. Normalized diffraction patterns of annealed samples $Fe_{73.5}Si_{13.5-x}Ge_xCu_1Nb_3B_9$ at temperature 550 °C. In the upper right corner the inset documents the dependence of the lattice parameter a of $Fe_3(Ge,Si)$ phase on the Ge concentration.

of the amorphous remainder is considerably lower than the value obtained for the primary samples and slightly decreases with increasing Ge content.

4. Conclusion

XAFS, as an element sensitive method, suggests prolongation of the average interatomic distance between Fe atoms and their closest surrounding. Both, MS and XRD phase analysis, revealed the presence of the DO_3 structure with about 19 at.% of (Si,Ge) with the mean crystalline size of about 7 nm for all annealed investigated samples.

Acknowledgments

The paper was supported by the Slovak Grant Agency VEGA (1/0167/10) and it was done in the frame of the project "Centre of Excellence of progressive materials with nano- and submicron microstructure", with support of the Operational Program Research and Development financed from European Regional Development Fund.

References

[1] J.A. Moya, V.J. Cremaschi, H. Sirkin, *Physica B* **389**, 159 (2007).

[2] D. Muraca, V. Cremaschi, J. Moya, H. Sirkin, *J. Magn. Magn. Mater.* **320**, 1639 (2008).

[3] B.W. Corb, *Phys. Rev. B* **31**, 2522 (1985).

[4] S. Michalik, K. Saksl, P. Sovák, K. Csach, J.Z. Jiang, *J. Alloys Comp.* **478**, 441 (2009).

[5] P. Scherrer, *Göttinger Nachrichten Gesell.* **2**, 98 (1918).