# The Study of Magnetization in Strong Magnetic Fields for $Fe_{62-x}Co_{10}Nb_xY_8B_{20}$ (X = 0, 1, 2) Alloys

# KONRAD GRUSZKA<sup>1</sup>\*, MARCIN NABIALEK<sup>1</sup>, MICHAL SZOTA<sup>2</sup>, PETRICA VIZUREANU<sup>3,4</sup>, MOHD MUSTAFA AL BAKRI ABDULLAH<sup>3,4</sup>, KASIA BLOCH<sup>1</sup>, ANDREI VICTOR SANDU<sup>4,5\*</sup>

<sup>1</sup> Institute of Physics, Faculty of Production Engineering and Materials Technology, Czestochowa University of Technology, 19 Armii Krajowej Str., 42-200 Czêstochowa, Poland

<sup>2</sup>Institute of Materials Science and Engineering. Department of Materials Processing Technology and Applied Physics, Czêstochowa University of Technology, 19 Armii Krajowej Str., 42-200 Czestochowa

<sup>3</sup>Gheorghe Asachi Technical University of Iasi, Faculty of Materials Science and Engineering, 41 D. Mangeron Blvd., 70050, Iasi, Romania 3 Center of Excellence

<sup>4</sup> Geopolymer & Green Technology (CeGeoGTech), School of Material Engineering, Universiti Malaysia Perlis (UniMAP), 01000 Kangar, Perlis Malaysia

<sup>5</sup> Romanian Inventors Forum, 3 Sf. P. Movila Str., Iasi, 700089, Romania

The magnetization process in the area called the approach to ferromagnetic saturation for  $\operatorname{Fe}_{g_2,x} \operatorname{Co}_{10} \operatorname{Nb}_x Y_s B_{20}$ (x = 0, 1, 2) bulk alloys was analyzed. The research were performed using the LakeShore vibrating magnetometer in the fields range of 0 T to 2 T. In the studied alloys, in strong magnetic fields, the structural defects affect magnetization process and the type of these defects was designated – on the basis of the Kronmüller theory. The article shows that the addition of niobium instead of the iron slightly decreases the saturation magnetization and decreases the value of the coercivity field by filling structure voids leading to material increase in homogenisation.

Keywords: Amorphous materials, metals and alloys, Bulk metallic glasses, microstructure, X-ray diffraction

Metallic amorphous materials despite their topological and chemical disorder very often show magnetic ordering [1-7]. Years of research have shown that many of these materials are characterized by excellent ferromagnetic properties, frequently finding their application in electrical industry mainly as magnetic cores for transformers, chokes and electric motors [8, 9]. Constant search for materials with better properties is born from the need to develop more efficient ways of energy transmission and generally in costs lowering. Such approach enforces the need of better understanding of the fundamental properties and the details of production processes that allow for their achievement.

Magnetic amorphous iron base alloys are well known for their good soft magnetic properties such as low magnetostriction, low core loses, high magnetic saturation and low coercivity field. Among them, the ternary Fe-Co-B type alloys exhibit good magnetic properties simultaneously showing quite strong response to various doping, thus being a good base for further improvements. In our previous works [1] we show that in case of FeCoB alloys yttrium is the one of the additives positively influencing both structural properties and glass forming ability (GFA), if its content does not exceed at. 8%. In comparison to the rest of components yttrium has biggest atomic radius satisfying one of A. Inoue empirical law of amorphous glass preparation [10].

For the magnetization process apart from main material parameters like magnetic saturation and coercivity field also important is how the process occurs. A significant impact on its route have also structural parameters of material. In case of crystalline structure where high topological order is present the magnetic structure of material is commonly following this order. Deviations from the ideal arrangement of atoms making the unit cell are typically anchor points breaking smooth domain wall movement. This kind of structure disorder are known as structural defects. However, in amorphous materials the lack of topological order makes it very hard to define such defects. The literature has adopted that counterparts of single crystalline vacancies are known as point defects when dealing with amorphous material. The two dimensional aggregate of point defects are called pseudodislocational dipoles. Both, as in the case of crystalline materials have great impact on domain walls movement and magnetization process. The indirect analysis of those defects is possible, by studying primary magnetization curve with respect to the Kronmüller theory [11-18].

In this work we study the relatively small addition of niobium at the expense of iron atoms. This element shows relatively low solubility in iron (both  $\alpha$ -Fe and  $\gamma$ -Fe) and it is usually used in corrosion resistant steels (due to low chemical activity) where it binds with carbon to form NbC and allows to harden the material. The addition of niobium could also improve alloy ductility.

The main aim of this paper is to examine low concentration (up to 2 at. %) of niobium on magnetic properties of  $Fe_{62}$ ,  $Co_{10}Nb_xY_8B_{20}$  bulk amorphous alloy.

# **Experimental part**

The samples used in the investigations were made using high-purity component elements: Fe – 99.98 at %, Co – 99.98 at %, W- 99.9999 at %, Y- 99.98 at %, Mo – 99.9999 at %, Nb – 99.98 at %. The element boron was added as an alloy of known composition, i.e.Fe<sub>45.4</sub>B<sub>54.6</sub>. The liquid alloy was solidified using two respective methods: in the copper die using the injection-casting method (radial cooling) and on the copper plate (unidirectional cooling).

The samples in form of square plates (10x10 mm, 5 mm thick) were prepared using high purity elements (Fe: 99.95%, rest of elements 99.99%). Initially the ingots were

<sup>\*</sup> email: kgruszka@wip.pcz.pl ; sav@tuiasi.ro

melted several times ensuring even distribution of ingredients using arc-melting under protective atmosphere of argon. Next, thus produced ingots were placed in quartz capillaries inside of induction furnace (also under protective atmosphere of argon), and by using injection casting method under 1000 hPa pressure samples were injected into water-cooled copper mold having temperature of 320 K.

The XRD studies were conducted using BRUKER D8 Advance diffractometer equipped with Cu-K $\alpha$  lamp ( $\lambda$ =1.54056 Å) in the 2 $\Theta$  angle ranging in 30° to 100°, with step size 0.02° on powdered samples. Magnetic measurements were conducted using LakeShore vibrating sample magnetometer (VSM) in magnetic field strength up to 2T. The initial magnetization curves were measured with increased measuring points density providing a good quality base for defects studies in the approach to ferromagnetic saturation area. For the reference also sample without niobium addition was prepared.

### **Results and discussions**

After material preparation the structure of samples were investigated. Figure 1 presents XRD diffraction patterns for two samples with niobium substitution and reference  $Fe_{62}Co_{10}Y_8B_{20}$  alloy. As can be seen all three diffraction patterns show only broad maximum located between approximately 35° - 50° 2 $\Theta$  angle, which is typical for amorphous structure. As expected, the small addition of niobium did not influence long range ordering and there are no obvious crystallization peaks revealed.

Next, static magnetic hysteresis loops for all samples were measured. Results of those measurements are presented in figure 2.

The hysteresis loop shapes for all three samples are typical for soft magnetic materials (high magnetization and relatively low coercivity). Data resulting from the analysis of figure 2. are presented in table 1. As can be seen the magnetization saturation decreases with increasing niobium content. At the same time coercive field drops down drastically from 204 [A/m] for sample



without niobium to 25 [A/m] for sample with 2 at. % which is more than 8 times lower.

Table 1 BASIC MAGNETIC PROPERTIES

Sample	μ₀M₅ [T]	H <sub>C</sub> [A/m]
Fe62C010Y8B20	1.34	204
Fe61C010Nb1Y8B20	1.20	57
Fe60C010Nb2Y8B20	1.14	25

The coercive field is inextricably linked to the anchoring of domain walls which makes their movement difficult in the magnetization process. As it is known, structural defects may become such anchoring points causing magnetic moment between nearest neighbours harder to exchange. For this reason, a detailed analysis of primary magnetization curve was conducted on the basis of Kronmüller theory, which can provide indirect insight into materials structure.

## Approach to ferromagnetic saturation area

Data obtained from primary magnetization curve analysis were subjected to the Kronmüller theory [15]. The initial magnetization curve in the approach to ferromagnetic saturation area can be analyzed using following relationship:

$$\mu_{\mathbf{0}}M(H) = \mu_{\mathbf{0}}M_{s}\left[1 - \frac{a_{1/2}}{(\mu_{\mathbf{0}}H)^{1/2}} - \frac{a_{1}}{(\mu_{\mathbf{0}}H)^{1}} - \frac{a_{2}}{(\mu_{\mathbf{0}}H)^{2}}\right] + b(\mu_{\mathbf{0}}H)^{1/2}$$

where and are coefficients of linear fits responsible for the occurrence of defects  $(a_{1/2}, a_1, a_2)$  and for Holsteina-Primakoff process (b). According to this approach following figures presenting fits were prepared (fig. 3 - fig. 10).



Fig.3. High-field magnetization curve for Fe<sub>62</sub>Co<sub>10</sub>Y<sub>8</sub>B<sub>20</sub> sample responsible for a<sub>1</sub> coefficient



responsible for  $a_{1/2}$  coefficient



Fig.5. High-field magnetization curve for  $Fe_{e2}Co_{10}Y_8B_{20}$  sample responsible for Holstein-Primakoff process



Fig.6. High-field magnetization curve for  $Fe_{61}Co_{10}Nb_1Y_8B_{20}$  sample responsible for  $a_{1/2}$  coefficient



Fig.7. High-field magnetization curve for  $Fe_{61}Co_{10}Nb_1Y_8B_{20}$  sample responsible for Holstein-Primakoff process

The coefficient  $a_{1/2}$  is responsible for occurrence of point defects,  $a_1$  and  $a_2$  are connected with emergence of two dimensional conglomerates of point defects. The parameters obtained from detailed analysis of fits are presented in table 2 and table 3.

As can be seen by analyzing data from table 2, in case of sample  $Fe_{62}Co_{10}Y_8B_{20}$  the high value of  $a_{1/2}$  parameter indicates, that in the structure there is relatively lot of point defects. The presence of non zero a, coefficient suggests small, but finite presence of pseudo-dislocational dipoles. However in comparison with other two samples with niobium, the value of a, is comparable with magnitude of point defects, so it's importance can't be neglected. In case of samples  $Fe_{61}Co_{10}Nb_1Y_8B_{20}$ , and  $Fe_{60}Co_{10}Nb_2Y_8B_{20}$  there is no  $a_1$  or  $a_2$  coefficients indicating lack of linear defects influence on magnetization. The relatively small (in comparison with sample without niobium) value of  $a_{1/2}$ parameters near 10 times lower in case of sample with 1 at. % (and about 4 times for 2 at. %) times of niobium suggests a much smaller number of point defects in those samples, but comparable with the value of linear defects. Interestingly, the b parameter responsible for Holstein-



Fig.8. High-field magnetization curve for  $Fe_{60}Co_{10}Nb_2Y_8B_{20}$  sample responsible for  $a_{1/2}$  coefficient



Fig.9. High-field magnetization curve for  $Fe_{60}Co_{10}Nb_2Y_8B_{20}$  sample responsible for Holstein-Primakoff process



Fig.10. High-field magnetization curve for  $Fe_{60}Co_{10}Nb_2Y_8B_{20}$  sample responsible for  $a_{1/2}$  coefficient

Primakoff process (damping of thermally excited spin waves) is decreasing with the niobium addition (figs. 5,7,9). The calculations showed, that transition field for this process is similar in case of samples  $Fe_{62}Co_{10}Y_{8}B_{20}$  and  $Fe_{61}Co_{10}Nb_{1}Y_{8}B_{20}$  (0.66 [T] and 0.68 [T] respectively) while for the last sample its value increased up to 1.06 [T].

In the same time, the  $D_{spf}$  parameter (located in table 2) responsible for spin wave stiffness acts similar in case of sample without niobium and sample with 1 at. % of Nb while one can see about 1/3 increase in its value for sample with 2 at. % of Nb. Both transition field value and D<sub>sof</sub> are indicating that much more energy should be delivered to the system to change neighboring spin vectors. This can suggest, that Nb addition is acting like paramagnetic inclusion between magnetic atoms. The A<sub>av</sub> (exchange constant) parameters follow this behavior, increasing in case of  $Fe_{60}Co_{10}Nb_2Y_8B_{20}$ . The  $l_b$  parameter responsible for exchange distance is similar in all samples, denoting that niobium did not change magnetic influence distance. Basing on above, it can be concluded that Nb atoms filled the voids created by both point defects and their conglomerates. This resulted in complete linear defects removal, and significantly lowered number of point defects.

Sample	a <sub>/2</sub> 10 <sup>-2</sup> T	a,s	$\begin{bmatrix} a_1 \\ 10^{-2}T^{-1} \end{bmatrix}$	م 10-1	r-1]		b 10 <sup>-2</sup> <i>T</i> -1/2	Table 2	
Fe <sub>62</sub> Co <sub>10</sub> Y <sub>8</sub> B <sub>20</sub>	20.5	54	2.97	2.97		-		DATA OBTAINED FROM THE	
$Fe_{61}Co_{10}Nb_{1}Y_{8}B_{20}$	2.5	6	-				5.92	MAGNETIZATION CURVE	
$Fe_{60}Co_{10}Nb_2Y_8B_{20}$	4.7	0	-				3.31		
Sample	$D_{spf}$ $\left[meV \cdot nm^2\right]$	l <sub>h</sub> [nm]	] [10 <sup>-12</sup>	$\left[\frac{J}{m}\right]$	$\begin{bmatrix} N_{max} \\ 10^{24} m^{-3} \end{bmatrix}$		$N_{dip} = 10^{16} m^{-2}$	Table 3	
Fe <sub>62</sub> Co <sub>10</sub> Y <sub>8</sub> B <sub>20</sub>	40.19	2.23	3 1.1	76	-		19.96	ANALYSIS OF INITIAL	
Fe61Co10Nb1Y8B20	43.73	2.29	9   1.1	72	19.8	0	_	MAGNETIZATION CURVES	
Fe60Co10Nb2Y8B20	64.49	2.23	3 2.4	46	21.3	3	-		

The two last parameters presented in table 3 show point defect density  $(N_{max})$  and linear defects density  $(N_{dip})$ . Although in the reference sample there are point defects, its density can't be established because according to the Kronmüller theory it is possible only when  $l_{i}^{-1} < 1$ .

# Conclusions

In this work we studied the influence of niobium substitution (at the expense of iron) on the structural and magnetic parameters of Fe<sub>62-x</sub>Co<sub>10</sub>Nb Y<sub>8</sub>B<sub>20</sub> amorphous alloy samples. The small addition of this element significantly lowered the coercive field while only slightly decreased magnetization saturation. At the same time it seems that niobium atoms filled voids present in the reference sample. This resulted in improvement in samples homogeneity and caused reduction of number of anchoring sites, which may explain lowering of coercivity field. Because niobium is paramagnetic, it caused small decrease in magnetization saturation. It was also possible to determine the kind of defects that influence the magnetization process in all cases. The Kronmüller analysis shows, that in case of Fe<sub>62</sub>Co<sub>10</sub>Y<sub>8</sub>B<sub>20</sub> sample both point and linear defects had influence on magnetization process, while in samples with Nb only point defects are involved.

Acknowledgment : The authors would like to thank Dr eng. Sebastian Garus for the part of the material reaserch results.

### **References**

1.GRUSZKA, K., NABIALEK, M., BLOCH, K., WALTERS, S., Int. J. Mater. Res. **106**(7) 2015, p. 689.

2.GRUSZKA, K., NABIALEK, M., BLOCH, K., OLSZEWSKI, J., Nukleonika 60, 2015 p. 23.

3.NABIALEK M., PIETRUSIEWICZ, P., SZOTA, M., ABDULLAH, M.M.A.B., SANDU, A.V., Rev. Chim. (Bucharest), **68**, no. 1, 2017, p. 22.

4.BLOCH, K., NABIA£EK, M., SZOTA, M., Rev. Chim. (Bucharest), 68, no. 1, 2017, p. 18.

5.K. GUSZKA, M. NABIALEK, K. BLOCH, Arch. Metall. Mater. 61(2), 2016, p. 499–502

6.K. BLOCH, J. Magn. Magn. Mater., 390,2015, p. 118-122

7.NABIALEK M., J. Alloys and Compd., **642**, 2015, p. 98-103 8.MCHENRY M., WILLARD M., LAUGHLIN D., Prog. Mater Sci. **44**, 1999, p. 291-433

9.DRABOLD D.A., Eur. Phys. J. B 68,2009, p. 1-21.

10.INOUE A., Acta Mater. 48, 2000, p. 279-306.

11.KRONMULLER H., Phil. Mag. B 48, 1983, p. 127-150

12.KRONMULLER H., J. Magn. Magn. Mater. 41, 1984, p. 366-375

13.KRONMULLER H., MOSER N., R. Rettenmeier, IEEE Trans. Mag. MAG-20, (1984) p. 1388-1390

14.KRONMÜLLER H., Phys. Stat. Sol. (b) 127, 1985, p. 531-541

15.KRONMULLER H., MOSER N., Amorphous Metallic Alloys, Buttenworth, London, 1983, p. 341

16.BORROUS M., MOSER N., KRONMULLER H., Phys. Stat. Sol. (a) **112**, 1989, p. 181-188

17.POIANA, M., DOBROMIR, M., SANDU, A.V., GEORGESCU, V., J. Superconductivity Novel Magn., **25**(7), 2012, p. 2377.

18.POIANA, M., VLAD, L., PASCARIU, P., SANDU, A.V., NICA, V., GEORGESCU, V., Optoelectron Adv Mater-RC, **6**(3-4), 2012, p. 434.

Manuscript received: 15.10.2016