A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S
Volume 57 2012 Issue 1
DOI: 10.2478/v10172-012-0014-7

M. NABIAŁEK*, P. PIETRUSIEWICZ*, M. SZOTA**, M. DOŚPIAŁ* J. JĘDRYKA**, K. SZOTA***, S. LESZ****

EVALUATION OF THE MICROSTRUCTURE AND MAGNETIC PROPERTIES OF $Fe_{73}Me_5Y_3B_{19}$ (WHERE Me = Ti OR Nb) AMORPHOUS ALLOYS

OCENA STRUKTURY ORAZ WŁAŚCIWOŚCI MAGNETYCZNYCH STOPÓW AMORFICZNYCH $Fe_{73}Me_5Y_3B_{19}$ (GDZIE Me = Ti LUB Nb)

The results of microstructure and magnetic properties studies of the amorphous $Fe_{73}Me_5Y_3B_{19}$ (where Me = Ti or Nb) alloys are presented The samples of the investigated alloys were produced in the form of ribbons with thickness of approximately 30 μm by unidirectional cooling of the liquid material on a rotating copper wheel. Both investigated alloys, in the as-quenched state, were fully amorphous which was verified using a 'Bruker' X-ray diffractometer. Static hysteresis loops, measured using 'LakeShore' vibrating sample magnetometer (VSM), were typical as for soft magnetic ferromagnets. The $Fe_{73}Nb_5Y_3B_{19}$ and $Fe_{73}Ti_5Y_3B_{19}$ alloys were characterized by relatively high values of saturation of the magnetization (1.25 T and 1.26 T, respectively) and low coercivity field (16 A/m and 47 A/m, respectively). The core losses obtained for the investigated alloys were significantly lower than for commercially used FeSi transformer steels. Both alloys also exhibited excellent time and temperature stability of the magnetic properties (within the investigated temperature range), as confirmed by measurements of magnetic susceptibility and its disaccommodation.
Keywords: amorphous alloys, core losses, magnetic susceptibility, magnetic disaccommodation

Celem pracy było zbadanie mikrostruktury oraz właściwości magnetycznych czteroskładnikowych stopów $Fe_{73}Me_5Y_3B_{19}$ (gdzie Me = Ti lub Nb) o strukturze amorficznej, otrzymanych techniką ultraszybkiego zestalania ciekłego stopu na miedzianym obracającym się bębnie. Strukturę odlanych taśm badano za pomocą dyfraktometru rentgenowskiego firmy „Bruker". Z pomiarów tych wynika, że obydwa czteroskładnikowe stopy w stanie po odlaniu były amorficzne. Statyczne pętle histerezy zmierzone przy użyciu magnetometru wibracyjnego (VSM) firmy „LeakeSchore", były typowe jak dla ferromagnetyków magnetycznie miękkich, a wytworzone stopy $Fe_{73}Nb_5Y_3B_{19}$ i $Fe_{73}Ti_5Y_3B_{19}$ cechowały się względnie wysoką magnetyzacją nasycenia (odpowiednio: 1.25 T i 1.26 T) i małym polem koercji (odpowiednio: 16 A/m i 47 A/m). Straty energii potrzebnej na jeden cykl przemagnesowania dla obydwu badanych stopów były znacznie mniejsze niż dla komercyjnie produkowanych stali transformatorowych FeSi. Wytworzone stopy wykazywały dobrą stabilność czasową oraz temperaturową właściwości magnetycznych (w wyznaczonym zakresie temperatur), którą określono na podstawie pomiarów podatności magnetycznej i jej dezakomodacji.

1. Introduction

The first amorphous materials, in the form of a ribbon were obtained in the 1960s by three research groups, led by Pond and Maddin, Chen, Miller and Masumoto, and again Maddin [1, 2]. The fabricated material, in the form of a ribbon with thickness of a few micrometres started a revolution in the basic research of amorphous materials exhibiting soft magnetic properties. The main technique used in production relies on the rapid quenching of liquid material ejected from a quartz capillary onto a rapidly rotating copper wheel [3, 4]. Currently, amorphous materials in the form of ribbons are manufactured by experts in many companies and research institutes.

Materials with non-periodic crystalline structure are characterized by a lack of magnetocrystalline anisotropy, what causes very low values of coercivity field. Additionally, due to low thickness and resistivity of ribbons,

* INSTITUTE OF PHYSICS, CZĘSTOCHOWA UNIVERSITY OF TECHNOLOGY, 42-200 CZĘSTOCHOWA, 19 ARMII KRAJOWEJ AV., POLAND
** INSTITUTE OF MATERIAL SCIENCE ENGINEERING, CZĘSTOCHOWA UNIVERSITY OF TECHNOLOGY, 42-200 CZĘSTOCHOWA, 19 ARMII KRAJOWEJ AV., POLAND
*** INSTITUTE OF MATHEMATICS AND COMPUTER SCIENCE, FACULTY OF MECHANICAL ENGINEERING AND COMPUTER SCIENCE, CZĘSTOCHOWA UNIVERSITY OF TECHNOLOGY, 42-200 CZĘSTOCHOWA, 19 ARMII KRAJOWEJ AV., POLAND
**** INSTITUTE OF MECHANICAL ENGINEERING MATERIALS AND BIOMATERIALS, FACULTY OF MECHANICAL ENGINEERING, SILESIAN UNIVERSITY OF TECHNOLOGY, 44-100 GLIWICE, 18A KONARSKIEGO AV., POLAND

the values of core losses generated in these materials during the magnetization cycle are also low [5]. The other benefit of amorphous alloys is the possibility of the regulation of the Curie temperature, through changes in their chemical composition. This group of materials is also characterized by very good time and temperature stability of magnetic properties. All of the aforementioned properties make these alloys very interesting, as materials for use in transformer cores working under harsh conditions [6, 7].

In this paper, studies are presented of the microstructure and magnetic properties of $Fe_{73}Me_5Y_3B_{19}$ (where Me = Ti or Nb) amorphous alloys. The production process is relatively cost-effective given the resulting excellent magnetic properties.

2. Materials and methodology

The samples used in the investigations were obtained by the unidirectional melt-spinning method, in which liquid alloy was quenched onto a rotating, copper wheel. The components used in the production process were of high-purities: Fe - 99.98; Y - 99.98; Ti = 99.999 and Nb - 99.999. Due to difficulties in the melting process of pure boron, this component was added to the alloy in the form of an FeB ingot with known composition. All ingots were arc-melted several times in order to ensure the homogeneity of the finished alloy. In the next stage ingots were crushed and placed in quartz capillaries. The alloy was induction-melted and then ejected under the pressure of neutral gas onto the copper wheel, rotating with a linear velocity of about 35 m/s. The finished samples of the investigated $Fe_{73}Me_5Y_3B_{19}$ (where Me = Ti or Nb) alloys were in the form of ribbons with approximate sizes: thickness equal to 30 μm and width of 3 mm. All parts of the production process were performed under a protective argon atmosphere. The microstructure of the samples was studied by means of a Brucker X-ray diffractometer equipped with a copper cathode (1.5418 Å). The static hysteresis loops were measured using a 'LakeShore' vibrating sample magnetometer. The core losses were derived from measurements performed using a 'Ferrotester' ferrometer. The maximal magnetic permeability values were measured using the transformer method in a fully automatic set-up. The Curie temperatures (T_C) were evaluated from the temperature dependence of saturation magnetization measured using a force magnetometer.

3. Results and Discussion

X-ray diffraction patterns obtained for the investigated samples of $Fe_{73}Me_5Y_3B_{19}$ (where Me = Ti or Nb) amorphous alloys are shown in Fig.1. These measurements were performed on powdered samples in order to incorporate information from the entire volume of the investigated materials. The diffraction patterns obtained for the two investigated alloys were found to be similar, consisting only of broad maxima, which is typical for amorphous alloys. Conversely, in crystalline alloys, exists translational symmetry in the atomic order, and it can be observed in X-ray diffraction patterns as a presence of narrow maxima. These maxima can be used to determine distance between atomic layers.

The static hysteresis loops obtained for the investigated samples are shown in Fig. 2.

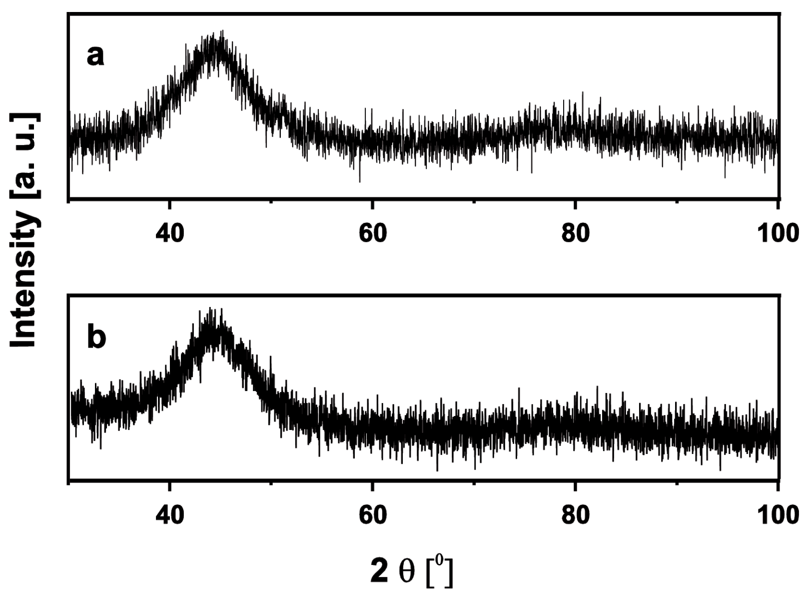

Fig. 1. X-ray diffraction patterns obtained for the $Fe_{73}Me_5Y_3B_{19}$ alloys in ribbon form in the as-quenched state: Me = Ti (a) and Nb(b)

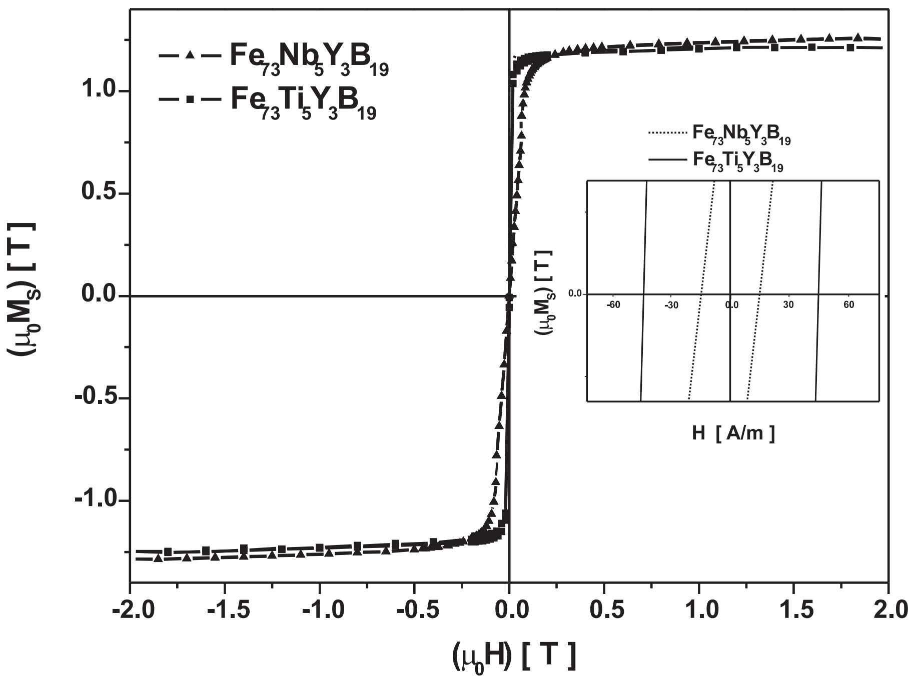

Fig. 2. The static hysteresis loops measured for the $Fe_{73}Me_5Y_3B_{19}$ alloys in ribbon form in the as-quenched state

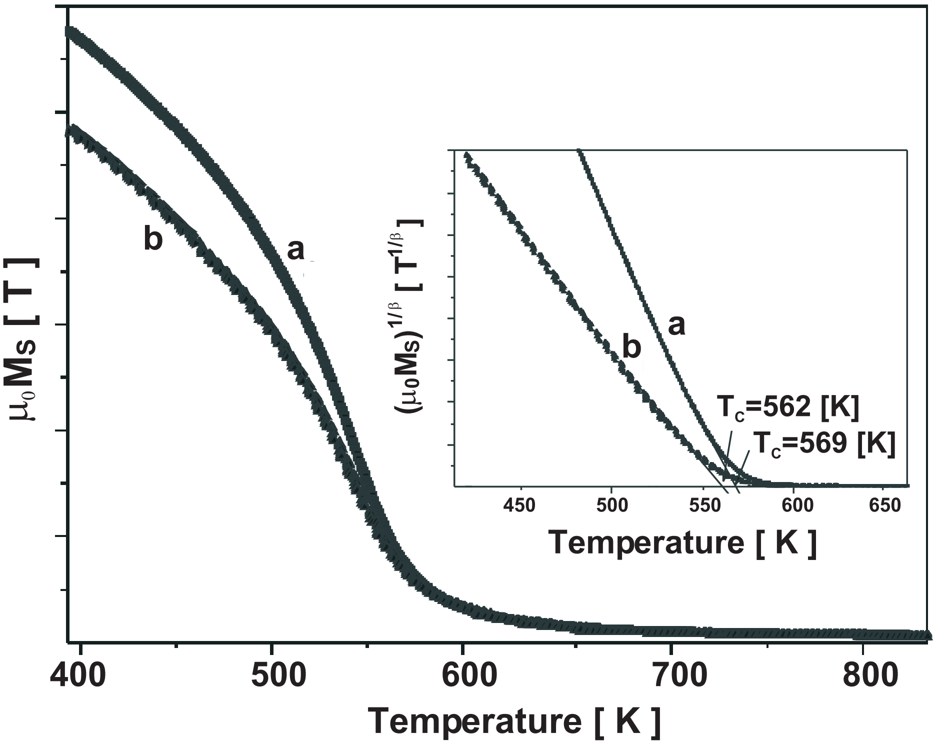

Fig. 3. Thermomagnetic curves of $Fe_{73}Me_5Y_3B_{19}$ alloys in the as-quenched state: Me = Ti (a) and Nb (b). In the inset, the relationship of $(\mu_0 M_s)^{1/\beta}$ with temperature is shown together with the Curie temperatures

The analysis of the static hysteresis loops indicates that both of the investigated alloys exhibit low coercivity and high values of saturation of the magnetization. The values of saturation of the magnetization and coercivity for $Fe_{73}Nb_5Y_3B_{19}$ amorphous ribbons were found to be 1.25 T and 16 A/m, respectively. The corresponding values for $Fe_{73}Ti_5Y_3B_{19}$ were 1.26 T and 47 A/m, respectively.

The commonly used materials for transformer steels in the electrotechnical industry are working at frequencies of approximately 50 to 60 Hz. These materials exhibit high values of core losses which are manifested as undesirable heat in the transformer core. Moreover, transformers equipped with classical FeSi cores are noisy in operation; this noise is in the form of a 'hum'. These unwanted features are caused by magnetostriction and losses in commercially used transformer cores. In comparison, the investigated in this paper alloys are characterized by much lower values of core losses. For both investigated alloys, was observed typical dependence of core losses i.e. increasing with increasing measurement frequency.

The Curie temperature is one of the most important parameters determining the suitability of materials for application as ferromagnetic alloys. Using a force magnetometer, the saturation of the magnetization $\mu_0 M_S$ as a function of temperature, was measured. The values of T_C for the studied samples were determined from the equation $\mu_0 M_S = \mu_0 M_0 (1 - T/T_c)^{\beta}$, where $\mu_0 M_0$ is the saturation magnetization at T=0 K and the factor β (equal to 0.36 for the Heisenberg ferromagnet) is the critical exponent in this equation[8].

The evaluated Curie temperatures for both investigated alloys were found to be similar, with values of approximately 570 K. The determination of a discrete value of Curie temperature is hindered due to the metastable structure of amorphous alloys.

The time-stability of magnetic susceptibility can be calculated from the isochronal curves describing disaccommodation of magnetic susceptibility phenomenon (Fig. 4). This phenomenon is observed during the magnetization cycle of the ferromagnetic material in magnetic fields between $0.1H_C < H < 0.4H_C$ (the so-called Rayleigh region).

The isochronal disaccommodation curves measured for the investigated alloys have shape as typically observed in the single-phase amorphous alloy (i.e. consisting of one broad maximum). It can be clearly seen, that the value of the disaccommodation for the alloy containing Nb is much lower than that for the alloy containing the addition of Ti. This can be associated with the lower number of 'free volumes' created during the production process. The density of niobium is equal to double density of titanium, what causes an increase in the density of the final alloy. On the basis of the results conducted by H. Kronmüller and his co-workers, it can be stated that the observed disaccommodation of magnetic susceptibility phenomenon in amorphous alloys is connected with the ordering of atomic pairs in the vicinity of 'free volumes' [9-14].

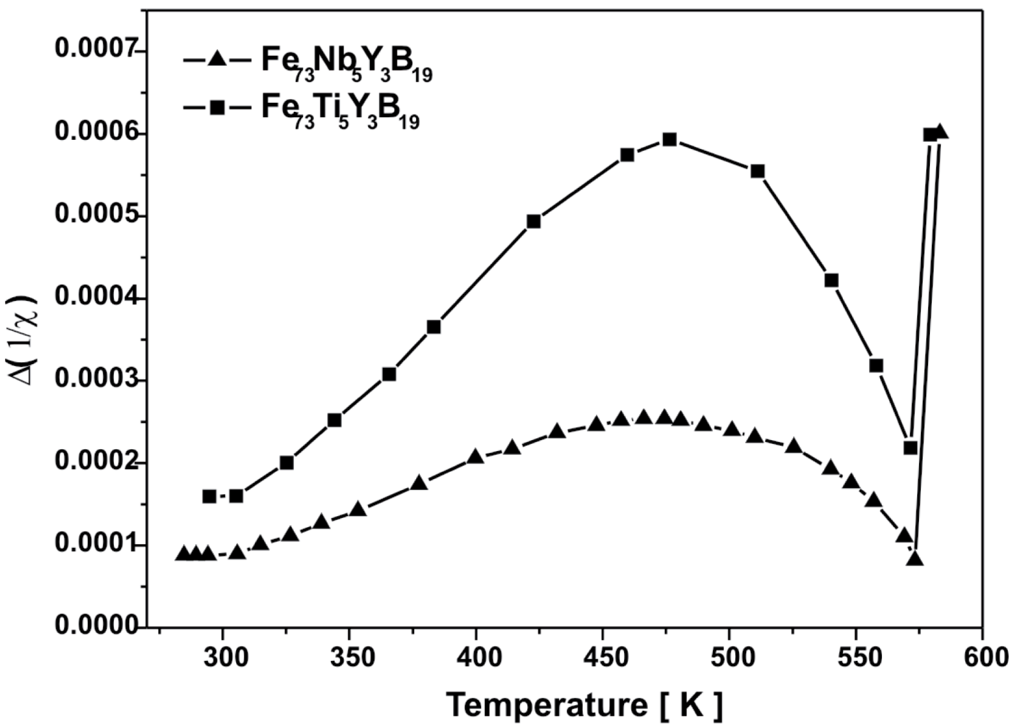

Fig. 4. Isochronal disaccommodation curves $\Delta(1/\chi) = f(T)$ for $Fe_{73}Me_5Y_3B_{19}$ alloys (where Me = Ti and Nb)

4. Conclusions

It is possible to obtain fully amorphous $Fe_{73}Me_5Y_3B_{19}$ (where Me = Ti or Nb),alloys in the form of ribbons with thickness of approximately 30 μm, using the rapid solidification method (the melt-spinning method).

The Curie temperatures of the investigated alloys had similar values, which indicates that substitution of a few atomic percent of Nb by Ti has no significant influence on the stability of the ferromagnetic state. This was confirmed by measurement of the thermal dependency of saturation magnetization, initial magnetic susceptibility and its disaccommodation. The saturation magnetization exhibited by the investigated alloys was found to be relatively high – approximately 1.25 T.

The substitution of Ti by Nb in the manufactured amorphous ribbons was found to have had a significant influence on values of the alloy coercivity (a decrease from 47 A/m to 16 A/m), the initial magnetic susceptibility and its disaccommodation (an increase in the former and decrease in the latter).

The core losses per single magnetization reversal cycle were found to be higher for the sample containing titanium.

Summarizing, from evaluations of the investigated alloys, superior soft magnetic properties were achieved for $Fe_{73}Nb_5Y_3B_{19}$, which as a result is considered the better alternative for electrotechnical applications.

REFERENCES

[1] R.B. Jr. Pond, R. Maddin, A method of producing rapidly solidified filamentary castings. Trans. Metall. Soc. AIME **245**, 2475 (1969).

[2] H.S. Chen, C.E. Miller, A rapid quenching for the preparation of thin uniform films of amorphous solids. Rev. Sci. Instrum. **41**, 1237 (1970).

[3] D.S. Easton, C.T. Liu, J.A. Horton, E.P. George, J.J. Campbell, Processing and characterization of Ni-Al-Fb-B shape memory alloys wires produced by rapid solidification. Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, Tn 37831-6115.

[4] W. Qin, F. Xu, K. Peng, W.L. Jiang, G. Ni, S.L. Tang, Y.W. Du, Effect of the slight surface oxidation of ribbons on the dynamic magnetization of nanocrystalline soft magnetic ribbons. Journal of Non-Crystalline Solid **316**, 398 (2003).

[5] A. Makino, T. Bitoh, A. Kojima, A. Inoue, T. Masumoto, Low core losses of nanocrystalline Fe-Zr-Nb-B soft magnetic alloys with high magnetic flux density. Material Science and Engineering **A304 306**, 1083 (2001).

[6] M. Nabiałek, J. Zbroszczyk, W. Ciurzyńska, J. Olszewski, S. Lesz, P. Brągiel, J. Gondro, K. Sobczyk, A. Łukiewska, J. Świerczek, P. Pietrusiewicz, Microstructure, some magnetic and mechanical properties of amorphous $Fe_{61}Co_{10}Zr_{2.5}W_2Hf_{2.5}Y_2B_{20}$ plates, Archives of Metallurgy and Materials **55**, 349-357 (2010).

[7] A. Łukiewska, J. Zbroszczyk, M. Nabiałek, J. Olszewski, J. Świerczek, W. Ciurzyńska, K. Sobczyk, M. Dośpiał, Low and high magnetic field properties of nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{20}$ rods, Archives of Metallurgy and Materials **53**, 881-885 (2008).

[8] Z. Stokłosa, P. Kwapuliński, J. Rasek, G. Badura, G. Haneczok, L. Pająk, L. Lelątko, Structural relaxation, crystallization and improvement of magnetic properties in FeXSiB (X = Cr, Nb) – type amorphous alloys. Journal of Magnetism and Magnetic Materials **320**, e762 (2008).

[9] H. Kronmüller, N. Moser, Magnetic after-effects and the hysteresis loop, in Amorphous Metallic Alloys, ed. F. E. Luborsky, Butterworth, 341, London (1983).

[10] H. Kronmüller, N. Moser, R. Rettenmeier, Micromagnetic analysis of magnetic after-effects in amorphous alloys by two-level system. IEEE Trans. Magn. Magn. p. 1338 (1984).

[11] H. Kronmüller, Magnetic aftereffects, induced anisotropics, and the role of two-level system in amorphous alloys. J. Magn. Magn. Mater. **41**, 366 (1984).

[12] N. Moser, H. Kronmüller, R. Rettenmeier, Reversible and irreversible structural relaxations in ferromagnetic amorphous alloys. In: Rapidly quenched metals, Proc. Fifth International Conference on Rapidly Quenched Metals, ed. S. Steeb, H. Walimont, Elsevier Science Publishers B. V. Amsterdam North Holland 1195 (1985).

[13] R. Rettenmeier, H. Kronmüller, Reversible and irreversible structural relaxations in soft ferromagnetic amorphous alloys. Phys. Stat. Sol. (a) **93**, 221 (1986).

[14] H. Kronmüller, The role of two-level system in amorphous metallic alloys. Phys. Stat. Sol (b) **127**, 531 (1985).

Received: 10 January 2011.