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THE STRUCTURAL RELAXATION AND ITS INFLUENCE ON HIGH FIELD MAGNETIZATION PROCESSES

RELAKSACJA STRUKTURALNA I JEJ WPŁYW NA PROCESY MAGNESOWANIA W SILNYCH POLACH MAGNETYCZNYCH

In this paper the results of the structural and magnetic investigation of $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ alloy after solidification and isothermal annealing was presented. The isothermical annealing was carried out at 700 K for 1 h and 770 K for 3.5 h. For the structural investigation was performed by X-ray diffractometer equipped with a copper lamp. The results of (XRD) measurements showed the material in the state after the solidification and heat treatment is amorphous. Static hysteresis loops and initial magnetization curve was measured using vibrating magnetometer (VSM). The quality and quantity of structural defects in the sample after heat treatment was determined by indirect method using analyze the initial magnetization curve in accordance with the theory of Kronmüllera. These studies have shown that the annealing process has big influence to change significantly quantity of defects in amorphous structure as a result, there are changes of magnetic parameters such as saturation magnetization and field $\mu_0 M_s$ coercivity H_c.

Keywords: High magnetic fields, metallic glasses, magnetization, structural defects, relaxation

W pracy przedstawiono wyniki badań strukturalnych i magnetycznych stopu $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ w stanie po zestaleniu oraz po izotermicznym wygrzewaniu w temperaturze 700 K przez 1h i 770 K przez 3,5 h. Badania struktury wykonano przy użyciu dyfraktometru rentgenowskiego wyposażonego w lampę miedzianą. Wynik pomiarów (XRD) wykazał, że materiał w stanie po zestaleniu i obróbce termicznej jest amorficzny. Statyczne pętle histerezy i krzywą pierwotnego namagnesowania zmierzono za pomocą magnetometru wibracyjnego (VSM). Analizując krzywą pierwotnego namagnesowania i wykorzystując pośrednią metodę wyznaczania defektów strukturalnych zgodnie z teorią H. Kronmüllera wyznaczono jakość i ilość tych defektów w próbce w stanie po zestaleniu i po izotermicznym wygrzewaniu. Badania te wykazały że proces wygrzewania istotnie wypływa na zmiany zdefektowania struktury amorficznej w wyniku czego zachodzą zmiany parametrów magnetycznych takich jak magnetyzacja nasycenia $\mu_0 M_s$ i pole koercji H_c.

1. Introduction

Metallic glasses based on iron, showing the so-called soft magnetic properties are studied for over 30 years [1]. These materials are characterized by a much better soft magnetic properties, then their crystalline counterparts with the same chemical compositions [2-4]. They represent an interesting group of functional materials with great possibilities of application, eg in the electrical industry on energy-efficient transformer cores.

Amorphous alloys are obtained by rapid cooling of liquid metal. Undercooling of liquid in the production process leads to a disorder in the distribution of atoms in structure. As a result, we obtain a metastable material, characterized by short range interactions between atoms. The production process of metallic glasses causes by itself the formation of disorder in the structure of amorphous and leads to the formation of structural defects such as free volume and quasi dislocation dipoles [5, 6].

In high magnetic fields, above the anisotropy field ($H > 2K/\mu_0 M_s$: where K is effective anisotropy constant), the domain structure is not longer observed and material is not yet uniformly magnetized. Magnetization in the sample does not reach the maximum value, called the saturation of the magnetization, on what have a significant influence presence of structural defects, i.e. point defects and two-dimensional quasi dislocation dipoles. Through the analysis of the reduced saturation of the magnetization in the area known as the Ewing's "knee" [7] it can be determined the type, size and density of structural defects occurring in the volume of test material [8-10].

The aim of this study is to determine the effect of isothermal annealing on structural relaxation processes occurring in the amorphous materials in the form of tapes with a thickness of 30 μm. The aim of the work will be achieved by performing analysis of the reduced saturation of the magnetization curves according to the H. Kronmüller theory for the $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ alloy in the as-cast state and after thermal treatment.

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2. Research material and studies methodology

Research materials in the form of ribbons with a nominal chemical composition $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ were prepared of high purity elements (Fe, Co, Y, Zr = 99.99%), boron was added in the form of an alloy of known composition $Fe_{45.6}B_{54.4}$. Alloy ingots were prepared in an arc furnace in a protective gas atmosphere. The ingots were remelted several times in order to homogenise the polycrystalline structure and thorough mixing of the components. Tapes with a thickness of 30 μm and a width of 5 mm were prepared by melt-spinning technique, also in a protective argon atmosphere.

Samples in the as-cast state were heat-treated at temperatures of 700 K for 1 h and 770 K for 3.5 h. Annealing of samples took place in a quartz tubes from which the air was pumped out, in order to secure samples from oxidation during the heating process. The structure of the investigated material in the as-cast state and after isothermal annealing was studied using "BRUKER" X-ray diffractometer equipped with a copper lamp of radiation CuK_α and characteristic wavelength $\lambda = 1.54056$ Å.

Types of defects and their influence on magnetization processes in strong magnetic fields was determined using the micromagnetism theory, proposed by H. Kronmuller, called "approach to ferromagnetic saturation" [8]. Magnetization near the ferromagnetic saturation can be described by the relation [8, 9, 10, 11]:

$$M(H) = M_s \left[1 - \frac{a_{1/2}}{(\mu_0 H)^{1/2}} - \frac{a_1}{(\mu_0 H)^1} - \frac{a_2}{(\mu_0 H)^2} \right] + b(\mu_0 H)^{1/2} \tag{1}$$

where:

M_S – the saturation of magnetization, μ_0 – is the permeability constant (magnetic permeability of free-space), H – the strength of magnetic field, a_i (i = $^1/_2$, 1, 2) – coefficients of the linear fit, correspond to free volumes and linear defects, the b-coefficients of the linear fit corresponding to the suppression of spin waves by a magnetic field of high intensity - Holstein-Primakoffa paraprocess [12, 13].

3. Results and discussion

Fig. 1 shows the X-ray diffraction patterns of the sample in the as-cast state and after isothermal annealing at 700 K for 1 h and 770 K for 3.5 h.
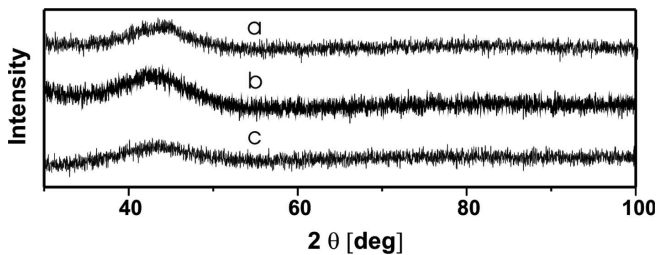

Fig. 1. X-ray diffraction patterns for samples in the as-cast state (a) and after annealing at 700 K (b) 770 K (c)

On all the diffraction patterns it is not observed the presence of narrow sharp peaks corresponding to reflections originating from crystallographic planes. Can be specified only the broad diffuse diffraction maxima corresponding to the amorphous matrix.

Hysteresis loops measured for as-cast and heat-treated at 700 K for 1 h and 770 K for 3.5 h samples are shown in Fig. 2. From the analysis of the hysteresis loops for all samples shows that they show relatively good soft magnetic properties, such as: low coercivity field (H_c) and high saturation of the magnetization ($\mu_0 M_s$). Good soft magnetic characteristics determine the application of the material in the electrical industry to build energy-efficient, low-power transformers. The parameters determined on the basis of the courses of static hysteresis loop are given in Table 1.
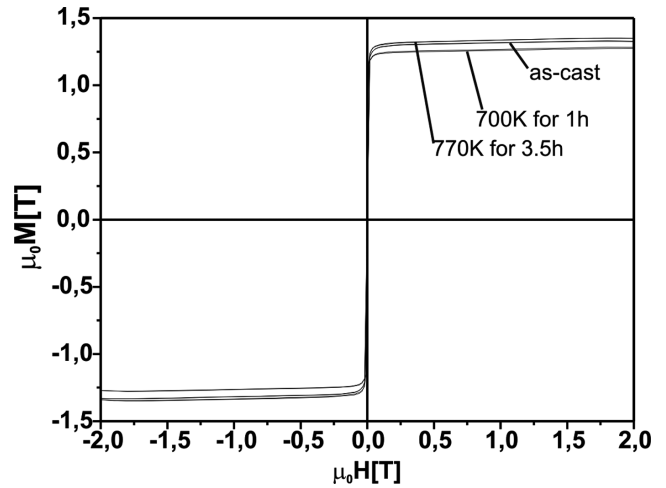

Fig. 2. Static hysteresis loops of $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ alloy in the as-cast state and after heat treatmen

Using the "approach to ferromagnetic saturation" theory and by analyzing the reduced curves of initial magnetization M/M_s as a function of magnetic field strength ($\mu_0 H$), it was observed fulfillment of rights only for $(\mu_0 H)^{-1/2}$, $(\mu_0 H)^{-1}$, $(\mu_0 H)^{1/2}$.
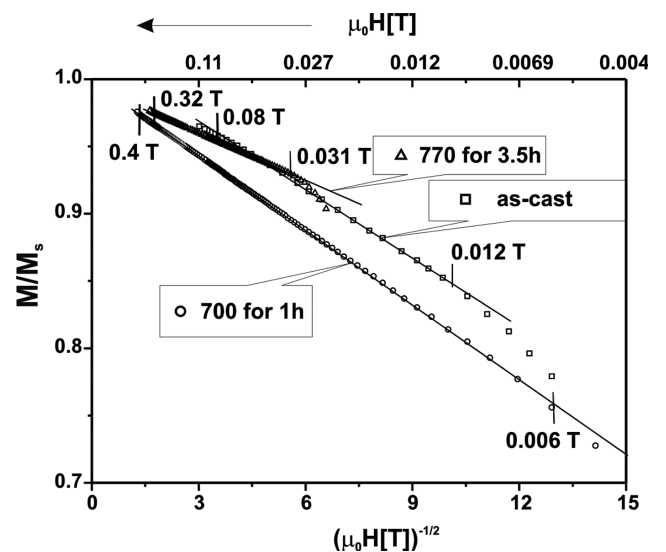

Fig. 3. Reduced magnetization M/M_S of the $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ alloy, in the as-cast state and after heat treatment as a function of $(\mu_0 H)^{-1/2}$

In the extent of magnetic fields from 0.012 T to 0.08 T, for the sample in the as-cast state, was observed that the linear fit meet the right $(\mu_0 H)^{-1/2}$ (Fig. 3). That indicates,

that the process of magnetization in this range of magnetic fields is done by rotation of magnetic moments within the free volumes.

At higher magnetic fields from 0.08 T to 0.27 T for the sample in the initial state was observed meeting law $(\mu_0 H)^{-1}$ (Fig. 4), which is associated with presence of linear defects, in the form of quasidislocalised dipoles. These defects are a source of short-range structural stresses, which cause heterogeneous distribution of magnetization within the free space, which in turn significantly affects the value of the coercivity field and saturation of the magnetization.

For samples subjected to isothermal annealing , there was no linear match, fulfilling the approach to ferromagnetic saturation law for $(\mu_0 H)^{-1}$, which indicates that there was a decay of unstable linear structural defects called quasidislocalised dipoles at point defects. In this case the magnetization process takes place as a result of rotation of magnetic moments within the free volumes and the right approach to ferromagnetic saturation is met for $(\mu_0 H)^{-1/2}$.
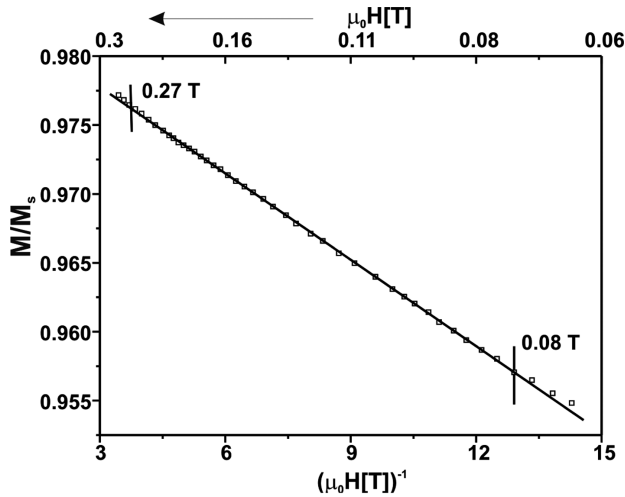
Fig. 4. Reduced magnetization M/M$_S$ sample of $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ alloy in the as-cast state as a function of $(\mu_0 H)^{-1}$

In high magnetic fields near the magnetic saturation (Fig. 5), i.e. above 0.28 T for samples in the as-cast state, 0.4 T and 0.33 T, respectively for samples subjected to annealing at 700K for 1h and at 770K for 3.5 h, was observed Holstein – Primakoff paraprocess, which is associated with suppression of thermally excited spin waves [12, 13].

The results obtained from the analysis of the reduced saturation of the magnetization in the magnetic field strength functions in powers of: – 1/2, 1/2 and -1 were collected in Table 1.

By analyzing the data shown in Table 1, it was found that annealing at a temperature of 700K led to the disintegration of an unstable linear defects in a more stable point defects, which means that the process of magnetization of the sample was held by a collective rearrangement of spins within the free volumes. Increasing the value of the transition field for Holstein – Primakoff paraprocess, contributed to reduction of the exchange distance of magnetic interactions, which resulted in an increase in coercivity field H_c.
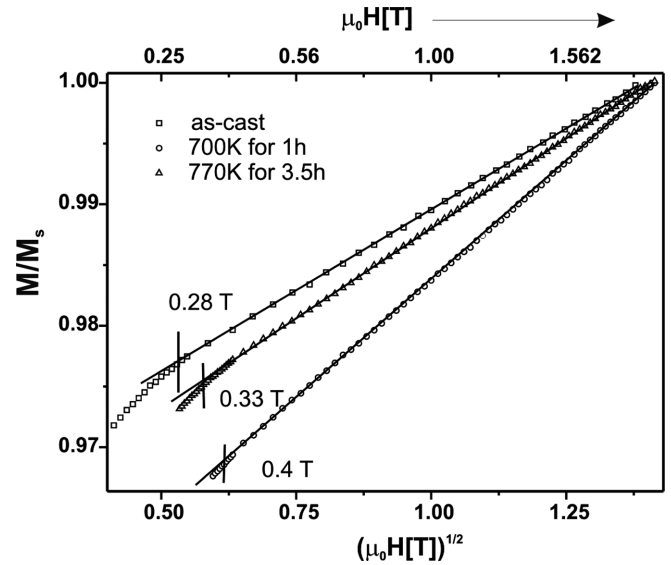
Fig. 5. Reduced magnetization M/M$_S$ of the $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ alloy, in the as-cast state and after heat treatment as a function of $(\mu_0 H)^{1/2}$

In such a case, subsequent annealing of the material at a temperature of 770 K for 3.5 h caused a reduction in density of point defects N, what may be related to diffusion of free volumes to the surface of the sample. The second annealing process has also affected the growth D_{sp}, A_{ex} and l_H, which according to posted data influenced the increase in the value of saturation of the magnetization and lowered the coercivity field values.

4. Conclusions

Using theory approach to ferromagnetic saturation, an attempt to explain the influence of the structural relaxation process that occurs during the isothermal annealing on the magnetic properties of the investigated alloy.

TABLE 1

The results obtained from the analysis of the reduced of magnetization as a function of magnetic field in the powers of - 1/2, -1, -2, or 1/2. D_{sp} – spin wave stiffness parameter, A_{ex} – exchange constant, l_h – exchange distance, N – the density of point defects, N_{dip} – density of quasidislocalised dipoles

	$a_{1/2}$ 10^{-2}	a_1 10^{-2}	b 10^{-2}	D_{sp} $[10^{-2}$ meV nm$^2]$	A_{ex} $[10^{-12}$ J m$^{-1}]$	l_H $[10^{-9}$ nm]	N_{dip} $[10^{16}$ nm$^{-2}]$	N $[10^{25}$ nm$^{-3}]$	$\mu_0 M_s$ [T]	H_c [A/m]
				$Fe_{61}Co_{10}Y_8Zr_1B_{20}$						
As - cast	1.70	0.21	2.67	74.27	3.23	4.67	4.58	0.98	1.33	35
700 K for 1h	1.84	-	3.88	57.97	2.41	3.45	-	2.43	1.27	54
770 K for 3.5 h	1.16	-	2.96	69.43	3.07	4.16	-	1.39	1.35	28

During the manufacturing process of amorphous glasses, comes to the formation of inhomogeneities of amorphous structure in the form of structural defects called: point and quasidislocalised dipoles. The formation of this type of structural defects caused by the freezing of free volumes during ultra fast quenching of alloy onto rotating copper drum. Defects occurring in the material are a source of internal mechanical stresses which give rise to non-uniform distribution of magnetic moments.

Investigated alloys both in the as-cast state and after heat treatment were amorphous.

From an analysis of initial magnetization curve for the sample in the as-cast state, the fulfillment of the approach to ferromagnetic saturation rights for $(\mu_0 H)^{-1/2}$ and $(\mu_0 H)^{-1}$ was shown. On the basis of these results it was found that two types of structural defects in this material are present. That in turn indicates, that the process of magnetization of the sample in the as-cast sample is done by ordering unevenly distributed spins due to their rotation within the free volumes and quasidislocalised dipoles.

After the first stage of the heating process, which was carried out at 700 K for 1 h occurred to disintegration of the structural defects in the form of quasidislocalised dipoles into smaller more stable point defects, which in turn contributed to increase the maximum excess volume. As a result of the second stage of heat treatment, which was performed at a higher temperature (770 K) and for a longer time (3.5 h), came to the annihilation of free volumes through migration to the surface of the material, which was the reason for reducing the N density.

Isothermal annealing of the material at different temperatures and at different time affects, among other things the change of correlation distance in the case of ferromagnetic interactions between magnetic Fe and Co atoms and change of the basic functional parameters of the investigated $Fe_{61}Co_{10}Y_8Zr_1B_{20}$ alloy.

Acknowledgements

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