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
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INFLUENCE OF ANNEALING ON THE MICROSTRUCTURE AND MAGNETIC PROPERTIES IN AMORPHOUS ALLOYS

WPŁYW WYGRZEWANIA AMORFICZNEGO STOPU NA MIKRUSTRUKTURĘ ORAZ WŁAŚCIWOŚCI MAGNETYCZNE

The influence of isothermal annealing on the magnetisation process in strong magnetic fields of the amorphous $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy ribbons was investigated. Samples in the form of ribbons were produced by rapid quenching of liquid alloy on a rotating copper wheel. In order to study the relaxation process, the investigated $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ samples were subjected to annealing below the crystallisation temperature at 700 K for 1 h and then at 770 K for 3.5 h. The structure of the samples was examined by X-ray diffraction measurements (XRD). It was found, that all of measured samples in the as-cast state and after thermal treatment, were amorphous. On the basis of virgin magnetisation curve analysis, the type, size and density of structural defects occurring in the investigated samples were determined. It was found that after the first stage of annealing, decay of linear defects (pseudo-dislocation dipoles) into smaller, more thermodynamically stable, point defects occurs. The presence of point like defects was also confirmed after the second stage of annealing.

Keywords: magnetic properties, classical amorphous alloys, Kronmüller theory, point and linear structural defects, structural relaxation

W pracy badano wpływ izotermicznego wygrzewania na proces magnesowania w silnych polach magnetycznych amorficznego stopu $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ w postaci taśmy. Materiał do badań wytworzono metodą szybkiego chłodzenia ciekłego stopu na miedzianym wirującym bębnie. Próbki stopu $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ poddano dwukrotnemu wygrzewaniu: 1) w temperaturze 700 K przez 1 godzinę, 2) w temperaturze 770 K przez 3,5 godziny. Strukturę taśm w stanie po zestaleniu i po obróbce termicznej badano za pomocą dyfraktometru rentgenowskiego. Wyniki analizy rentgenowskiej wykazały, że badane taśmy były w pełni amorficzne. Pomiar krzywych M-H wykonano za pomocą magnetometru wibracyjnego. Na podstawie analizy krzywych pierwotnego namagnesowania określono rodzaj, wielkość i gęstość defektów strukturalnych występujących w badanej taśmie. Na ich podstawie stwierdzono, że po pierwszym etapie wygrzewania nastąpił rozpad defektów liniowych (pseudodyslokacyjnych dipoli) na mniejsze bardziej stabilne termodynamicznie defekty punktowe, których obecność stwierdzono również po drugim etapie wygrzewania.

1. Introduction

The iron-based metallic glasses, exhibiting so-called ‘soft’ magnetic properties, are one of the new classes of materials in use in the electrotechnical industries, [1]. This class of alloys possesses substantially-improved applicability over their crystalline counterparts, [2-4]. Their functional properties make them very suitable for use in the electrotechnical industries, for example as core materials in more energy-efficient transformers.

The amorphous alloys are made in a process of rapid solidification of liquid alloy which facilitates the omission of the crystallisation process. The limitation in the long-distance atomic diffusion, during the alloy solidification process, is caused by an increase in its viscosity, which in turn leads to a lack of long-range atomic order. As a result, the obtained material is thermodynamically metastable. Moreover, during the rapid solidification process, locations with higher and lower density are created in the alloy, which are considered to be structural defects, [5-6]. The structural defects strongly influence the magnetic properties, such as the saturation magnetisation (M_S) and the value of the coercive field (H_C); according to the H. Kronmüller theory, presence of these defects can be investigated indirectly from the M/M_S curve analysis as a function of μ_0H, in the values of power -1/2, -1 and -2. According to this theory, the type, size and density of the structural defects present in the volume of the investigated material can be found. Magnetisation in the vicinity of ferromagnetic saturation can be described by equation (1), [7-10]:

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

where:

M_S – saturation magnetisation, μ_0 – magnetic permeabil-

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ity of a vacuum, H – magnetic field, $a_{1/2}/(\mu_0 H)^{-1/2}$ is related to the presence of point-defects, and factors $a_1/(\mu_0 H)^{-1}$ and $a_2/(\mu_0 H)^{-2}$ are related to linear defects (so-called quasidislocation dipoles), b – coefficient in the factor $b(\mu_0 H)^{1/2}$, the latter describing Holstein-Primakoff paraprocess, [11-12].

The aim of this work was to investigate the influence of isothermal annealing on the microstructure and magnetic properties (such as M_S and H_C) of the $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy in the shape of ribbons of thickness 30 μm.

2. Methods and materials

The ingot of $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy was prepared from high-purity elements as follows: Fe - 99.99%, Co – 99.99%, Y – 99.99%, and Ni – 99.99%. The element Boron was added as FeB alloy of a known composition. The ingot was re-melted several times, using an arc-melting process, in order to ensure homogeneity. The amorphous ribbons, of thickness 30 μm and width 3 mm, were prepared by the melt spinning method which involved the rapid solidification of the molten alloy on a rotating copper wheel. The production processes of both the polycrystalline ingots and the amorphous ribbons were carried out under a protective argon atmosphere. Samples in the as-quenched state were subjected to a thermal treatment at a temperature of 700 K for one hour, then subsequently at a temperature of 770 K for 3.5 hours. Annealing of the samples was also performed under an inert atmosphere. The microstructure of the amorphous ribbons, in the as-quenched state and after isothermal annealing, was investigated by means of an X-ray diffractometer (XRD) equipped with a Cu lamp with K_α (λ =1.54056 Å). The static hysteresis loops were measured with a magnetometer with vibrating sample (VSM) with a magnetic field range of 0 – 2 T. In accordance with the H. Kronmüller theory describing 'the approach to ferromagnetic saturation', the size, type and density of the structural defects were found.

3. Results and discussion

In Fig. 1, the X-ray diffraction patterns of the investigated material are shown, both in the as-quenched state and after isothermal annealing. In the diffraction patterns for the investigated samples, in the as-quenched state and after isothermal annealing, only wide diffuse 'halos', characteristic for amorphous materials, can be seen.

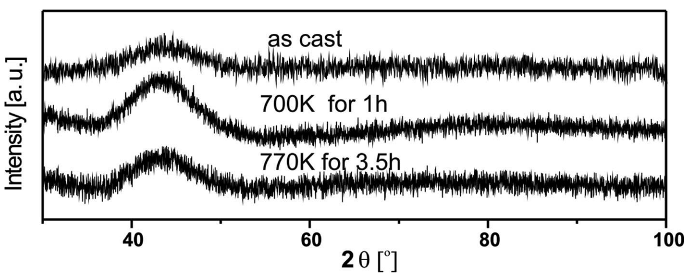

Fig. 1. The X-ray diffraction patterns of the investigated $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy in the as-quenched state and after isothermal annealing

The analysis of the static magnetic hysteresis loops (Fig. 2) revealed that the highest value of the saturation magnetisation (1.45 T) is exhibited by the sample in the as-quenched state.

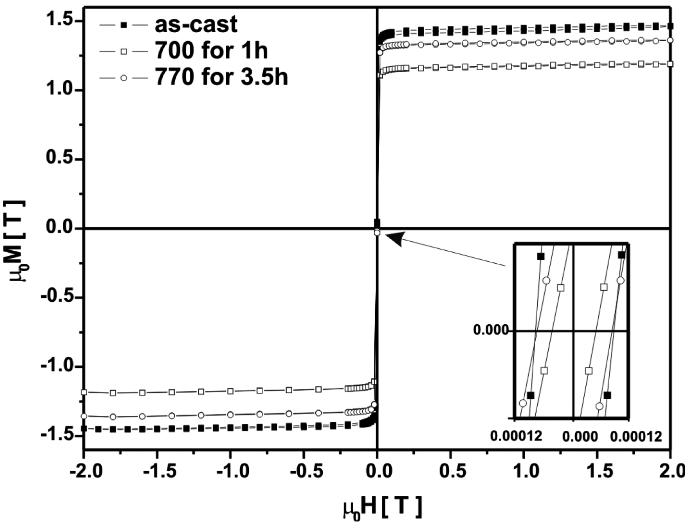

Fig. 2. The static hysteresis loops for the $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy in the as-quenched state and after isothermal annealing

With the help of the 'approach to ferromagnetic saturation' theory, the analysis of reduced magnetisation in the vicinity of ferromagnetic saturation was carried out. It is well – known that existing structural defects are a cause of internal stresses in materials; these internal stresses, through magnetoelastic interactions, influence the distribution of the magnetisation vectors. For the samples in the as-quenched state, the linear relationship of reduced saturation magnetisation, as a function of $(\mu_0 H)^{-1/2}$, in the field range of 0.12 T – 0.19 T, was found (Fig. 3). The existence of this kind of relationship means that the magnetisation process, in this magnetic field range, is related with the rotation of the magnetisation vector in the vicinity of free volumes. In higher magnetic fields, from 0.19 T -0.29 T, the relationship $M/M_S(\mu_0 H)^{-1}$ (Fig. 4) was observed which indicates the existence of linear structural defects called quasidislocational dipoles.

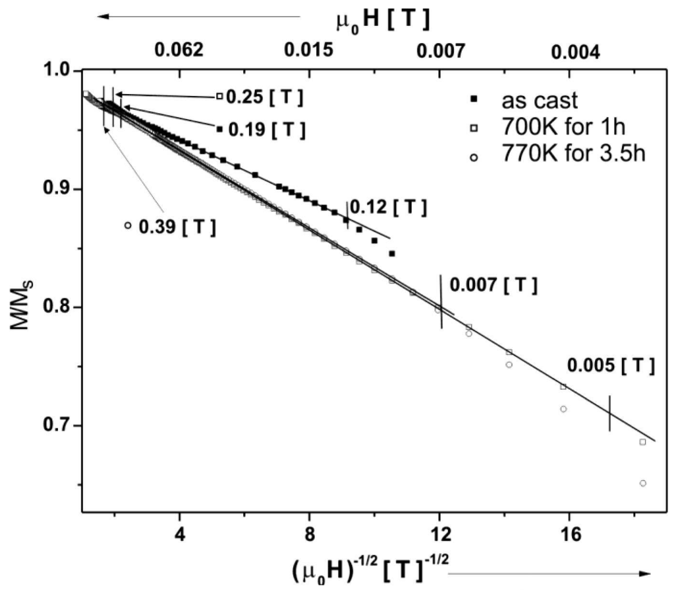

Fig. 3. Reduced magnetisation M/Ms versus $(\mu_0 H)^{-1/2}$ for the sample in the as-quenched state and after thermal annealing

TABLE 1

Data obtained from the analysis of the reduced magnetization as a function of the magnetic field in the powers of: -1/2, -1, -2 and $^{1}/_{2}$. D_{sp} – spin wave stiffness parameter, A_{ex} – exchange constant, l_H – exchange interactions length and D – density of the point defects

	$Fe_{61}Co_{10}Y_8Ni_1B_{20}$	$a_{1/2}$ 10^{-2} $[T^{1/2}]$	a_1 10^{-2} $[T^1]$	b 10^{-2} $[T^{1/2}]$	Dsp $[10^{-2}$ meV $nm^2]$	A_{ex} $[10^{-12}$ J $m^{-1}]$	l_H $[10^{-9}$ m]	N_{dip} $[10^{16}m^{-2}]$	N $[10^{25}m^{-3}]$	$\mu_0 M_s$ [T]	H_c [A/m]
1	As-cast	1.29	0.44	3.21	65.80	3.12	4.32	5.54	–	1.45	97
2	700K for1h	1.68	–	3.68	60.08	2.32	4.44	–	1.13	1.18	55
3	770K for 3.5h	1.65	–	3.25	65.16	2.90	3.70	–	1.96	1.36	83

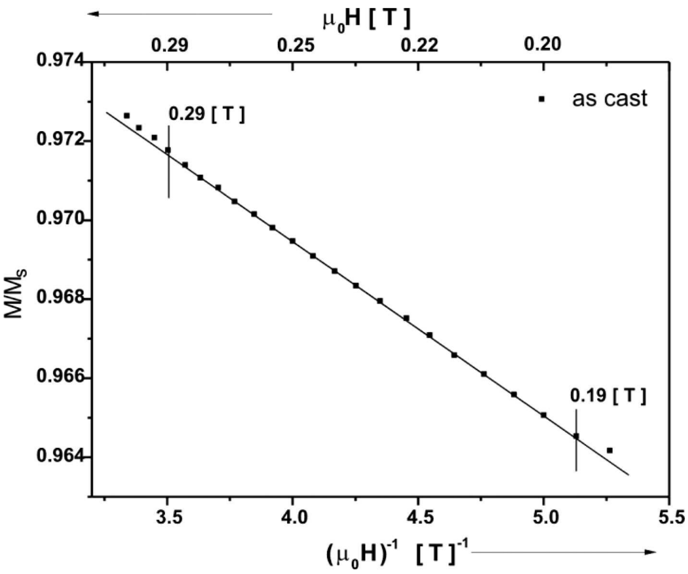

Fig. 4. Reduced magnetisation M/Ms as a function of $(\mu_0 H)^{-1}$ for the sample of $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy in the as-quenched state

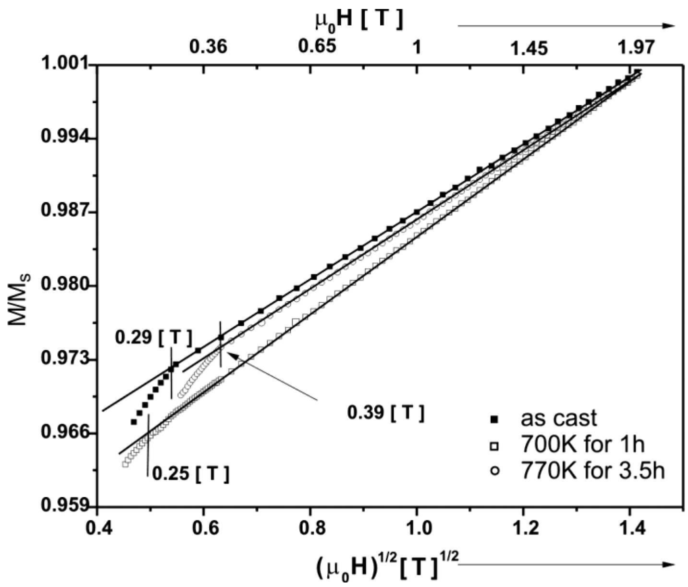

Fig. 5. Reduced magnetisation M/M_s as a function of $(\mu_0 H)^{1/2}$ for the sample of $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy in the as-quenched state and after thermal annealing

The Holstein-Primakoff paraprocess related with spin waves damping by magnetic field, [7-8], was observed in the magnetic field above: 0.29 T, 0.25 T, and 0.39 T for the sample in the as-quenched state, annealed at 700 K for 1 hour and 770 K for 3.5 hours, respectively. Parameters obtained from the magnetisation measurements are assembled in Table 1. On the basis of this data, it can be stated that during annealing of the investigated materials at the temperature of 700 K for one hour the decay of metastable linear defects to point defects occurred. This is confirmed by the linear relationship $M/M_S((\mu_0 H)^{-1/2})$ in the magnetic field range from 0.005 T - 0.25 T (Fig. 3). After the first stage of the annealing process, the beginning of the relationship $M/M_S((\mu_0 H)^{1/2})$, called the Holstein-Primakoff paraprocess, moved towards lower magnetic fields.

After the second annealing stage at the temperature of 770 K for 3.5 hours, the widening of the magnetic field range obeying the law of 'approach to ferromagnetic saturation' $M/M_S((\mu_0 H)^{-1/2})$ was observed (Fig. 3); the range of values was 0.007 T - 0.39 T.

In the case of the relationship $M/M_S((\mu_0 H)^{1/2})$ starting at the higher magnetic fields the exchange length, l_H, is decreasing (density of the point defects, N, is decreasing; please see Table 1). It is believed that this is caused by the rearrangement of the atoms in the vicinity of the free volumes during the thermal treatment.

4. Conclusions

The X-ray pattern analysis confirmed that the sample of the $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy in the shape of ribbons with thickness 30 μm were fully amorphous, both in the as-quenched state and after annealing at the temperature of 700 K for 1 hour and 770 K for 3.5 hours. The process of isothermal annealing of the ribbons of the $Fe_{61}Co_{10}Y_8Ni_1B_{20}$ alloy influenced the values of saturation magnetisation and coercive field (Table 1).

On the basis of the analysis of the reduced saturation magnetisation curves, as a function of the magnetic field in the powers of -1/2 and -1, it can be stated that after the first stage of the thermal treatment at the temperature of 700 K the decay of the linear defects (quasidislocational dipoles) to smaller, more thermodynamically stable, point defects occurred. After the second stage of thermal treatment, at the temperature of 770 K, the magnetisation process under high magnetic fields was also connected with the presence of point defects. It was noticed that the second stage of the annealing process caused an increase in the density of free volumes in the investigated alloy.

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