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LOW AND HIGH MAGNETIC FIELD PROPERTIES OF NANOCRYSTALLINE $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ RODS

NISKO I WYSOKOPOLOWE WŁAŚCIWOŚCI MAGNETYCZNE NANOKRYSTALICZNO/AMORFICZNYCH PRĘTÓW ZE STOPU $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$

Microstructure, low magnetic field properties i.e. initial magnetic susceptibility, its disaccommodation, core losses, and approach to ferromagnetic saturation studies for the bulk nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ material are presented. The nanocrystalline samples in the form of rods were obtained directly by a rapid solidification of the molten alloy. The nanocrystalline material consists of α-FeCo crystalline grains embedded and a heterogeneous amorphous matrix. The amorphous matrix exhibits two well separated urie temperatures which confirms existing of two amorphous phases. The investigated material exhibits good soft magnetic properties i.e. large initial magnetic susceptibility and very low disaccommodation in the temperature range from 150 K up to 400 K. The structure of the sample is relaxed after the solidification and only small quasi-dislocation dipoles with the width of 2.38 nm are present. Moreover, the $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ sample shows lower core losses than conventional crystalline materials.

Keywords: bulk nanocrystalline ferromagnets, magnetic properties, Mössbauer spectroscopy, approach to magnetic saturation

W pracy są przedstawione wyniki badań mikrostruktury i właściwości magnetycznych, takich jak początkowa podatność magnetyczna, jej dezakomodacja i straty na przemagnesowanie w słabych polach magnetycznych oraz podejście do ferromagnetycznego nasycenia dla masywnego, nanokrystalizowanego stopu $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$.

Próbki nanokrystaliczne w postaci prętów były otrzymywane bezpośrednio podczas szybkiego zestalania roztopionego stopu w miedzianej formie. Stop nanokrystaliczny składa się z ziaren fazy α-FeCo, i niejednorodnej matrycy amorficznej, charakteryzującej się dwiema różniącymi się wartościami temperatury Curie. Badany stop wykazuje dobre tzw. miękkie właściwości magnetyczne tj. dużą początkową podatność magnetyczną i bardzo małe natężenie dezakomodacji w zakresie temperatur od 150 K do 400 K. W próbce odprężonej w procesie produkcji są obserwowane pseudo-dyslokacyjne dipole o niewielkiej szerokości 2,38 nm. Dodatkowo, stop wykazuje mniejsze straty na przemagnesowanie niż konwencjonalne materiały krystaliczne.

1. Introduction

Nanocrystalline iron – based alloys are interesting materials due to their good soft magnetic properties with potential application in industry [1, 2]. Until now soft magnetic nanocrystalline alloys have been obtained by the proper annealing of amorphous ribbons. It is well known that the presence of nano-sized crystalline grains (with the volume fraction of 0.6-0.7) in the amorphous matrix improves the magnetic properties of these materials in comparison with their amorphous precursors [3, 4]. Recently, the amorphous alloys can be produced in the form of thick ribbons [5], rods, plates and tubes [6, 7]. Partial crystallization of these alloys enables us to obtain bulk nanocrystalline materials.

The goal of this paper is to study the microstructure, low and high magnetic field properties for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy.

2. Experimental procedure

The nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy in the form of rods 1 mm in diameter and 2 cm long was directly prepared by a rapid solidification of molten material in a copper mold cooled with water (suction- casting method).

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The microstructure of the powdered samples was investigated by Mössbauer spectroscopy and X-ray diffractometry. From X-ray diffraction studies the average diameter of the crystalline grains was evaluated using Scherrer's method [8].

The magnetic properties i.e. initial magnetic susceptibility and core losses were measured by a transformer method. The amplitude and frequency of the magnetic field during measurements of the magnetic susceptibility were equal to 0.32 A/m and 2 kHz, respectively. Those investigations were carried out after demagnetization of the sample. The magnetic susceptibility studies were performed in the temperature range from 150 K up to 540 K. Isochronal disaccommodation curves $\Delta(1/\chi) = f(T)$ were constructed according to the formula:

$$\Delta\left(\frac{1}{\chi}\right) = \frac{1}{\chi_2} - \frac{1}{\chi_1} = f(T) \qquad (1)$$

where χ_1 and χ_2 were magnetic susceptibilities measured at t_1=2 s and t_2=120 s after demagnetization of the sample, respectively. The high field magnetization was studied at room temperature using a vibrating sample magnetometer (VSM) for 5 mm long samples cut out from the rods. Moreover, thermomagnetic curves were measured using a force magnetometer in the magnetic field induction equal to B=0.7 T.

3. Results and discussion

In Fig. 1 the Mössbauer spectrum and hyperfine field distribution for $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy after solidification are presented. In the Mössbauer spectrum (Fig. 1 a) we can distinguish the Zeeman sextet with narrow lines superimposed on the broad line feature. The observed spectrum is characteristic of the partially crystallized amorphous alloy. From Mössbauer spectra analysis we have found that Zeeman sextet corresponds to α-FeCo crystalline phase with Co concentration equal to 12 at. %. The volume fraction of this phase evaluated from Mössbauer spectra equals to 0.07. From X-ray diffraction studies we have found that the crystalline phase consists of fine grains with the average diameter of 25 nm. However, in the as-quenched samples obtained at the lower quenching rate and consequently containing the larger amount of the crystalline phase we have stated the presence of the grains with the average diameter of 1 μm.

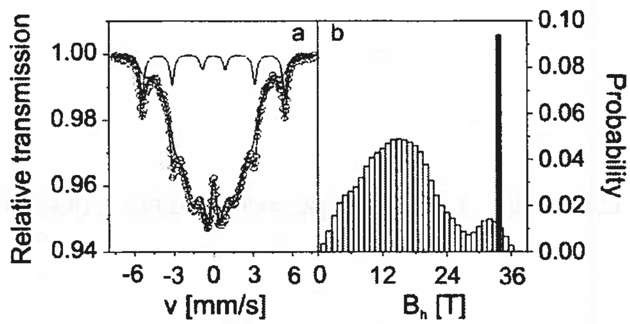

Fig. 1. Mössbauer spectrum (a) and hyperfine field distribution (b) for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy

The hyperfine field distribution obtained from the Mössbauer spectrum shows bimodal character and one can see low and high field components (Fig. 1 b). The obtained results indicate that the amorphous matrix in this alloy is inhomogeneous and regions with different iron concentration in this phase are present. In Fig. 2 the magnetic polarization μ_0M at the magnetizing field induction of 0.7 T versus temperature is depicted. The curve of $\mu_0M(T)$ shows two kinks at the temperature of 550 K and 750 K indicating the presence of at least two different ferromagnetic amorphous phases. The values of the Curie temperature of these phases, obtained from critical behaviour of the magnetic polarization (inset in Fig. 2), are equal to 529 K and 748 K, respectively. The existence of two phases leads to the bimodal hyperfine field distribution (Fig. 1 b). As can be seen from Fig. 2 the magnetic polarization does not reach zero at the temperature region of about 800 K due to the presence of small amount of the crystalline α-FeCo phase with much higher Curie temperature.

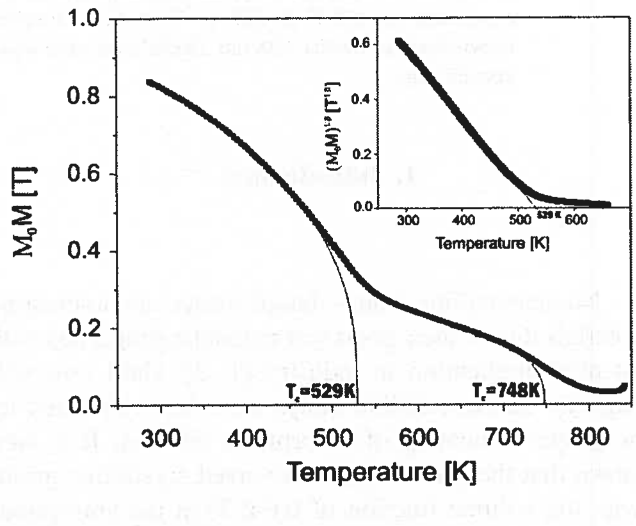

Fig. 2. Magnetic polarization, μ_0M, versus temperature for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy. Determination of Curie temperature by critical behaviour is shown in the inset

The initial magnetic susceptibility, χ_1, measured at t=2 s after demagnetization of the sample versus temperature, T, is presented in Fig 3. In the temperature range from 150 K to 400 K only a very broad maximum in the χ_1(T) curve followed by the sudden drop at higher temperature is observed. In the above mentioned temperature range the initial susceptibility exhibits good thermal stability. It is worth noticing that the magnetic susceptibility disaccommodation, $\Delta(1/\chi)$, as a measure of the time stability of the susceptibility in the temperature range 150 K – 400 K is very low (Fig. 4). The increase of $\Delta(1/\chi)$ at higher temperatures is connected with the ferro- to paramagnetic transition of the amorphous phase. Moreover, one can state that the phenomenon of the magnetic susceptibility disaccommodation in partially crystallized alloy is connected with relaxation processes occurring in the amorphous phase. The contribution of α-FeCo phase to this phenomenon is not evident.

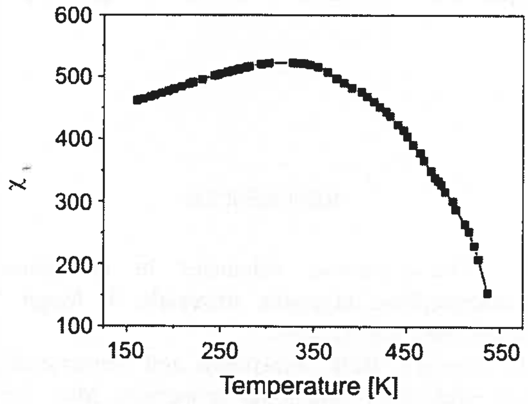

Fig. 3. Initial magnetic susceptibility, χ_1, as a function of temperature for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy

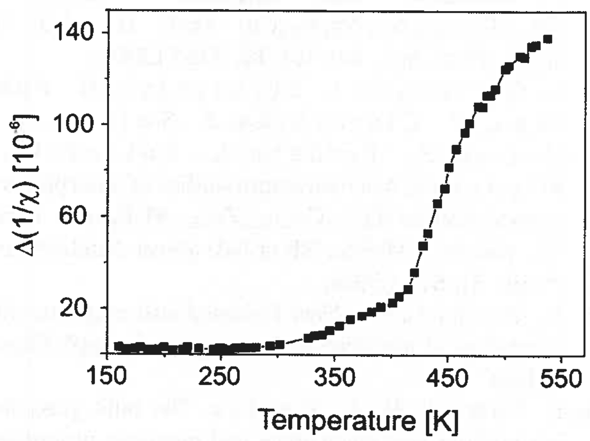

Fig. 4. Temperature dependence of the magnetic susceptibility disaccommodation, $\Delta(1/\chi)$, for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy

The total core losses versus maximum induction of the magnetizing field in the frequency range from 50 Hz to 1000 Hz are shown in Fig. 5. Taking into account the values of the core losses one can state that the investigated alloy is magnetically soft and at low frequencies the total losses are comparable with those observed in classical silicon-iron alloys [9]. However, at higher frequencies the losses are smaller than in those crystalline alloys.

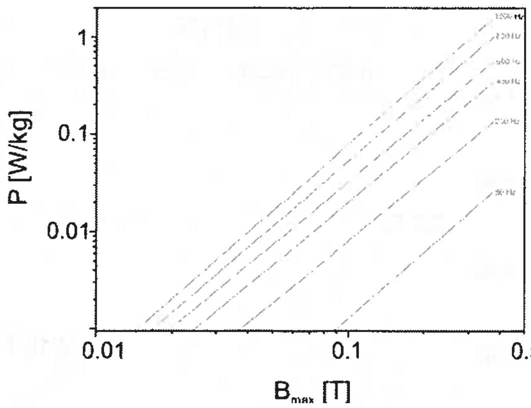

Fig. 5. Total core losses, P, versus the maximum induction, B_{max}, in the frequency range from 50 Hz to 1000 Hz of the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy

When the domain structure is not present in the sample the magnetic polarization ($\mu_0 M$) versus the magnetizing field (H) is described by the equation [10]:

$$\mu_0 M(H) = \mu_0 M_S \left(1 - \sum_i \frac{a_i}{(\mu_0 H)^i}\right) + b(\mu_0 H)^{\frac{1}{2}} \quad (2)$$

where $\mu_0 M_S$ is the magnetic saturation polarization at a given temperature, terms $\frac{a_i}{(\mu_0 H)^i}$ are connected with different kind of defects: i = $^1/_2$ for point like defects, i = 1 and 2 for linear defects called quasi-dislocation dipoles, the last term is ascribed to the Holstein-Primakoff paraprocess. In Fig. 6 the relative magnetization versus $\frac{1}{\mu_0 H}$ is shown. In the field range from 0.18 T to 0.80 T the linear dependence $\frac{M}{M_S} = f\left(\frac{1}{\mu_0 H}\right)$ is observed. According to the Kronmüller's theory [10], it indicates that in the sample the linear defects called quasi-dislocation dipoles are present and the rule

$$D_{dip} * l_H^{-1} < 1 \quad (3)$$

is fulfilled, where D_{dip} is the dipole width and l_H is the exchange length described by:

$$l_H = \sqrt{\frac{2A_{ex}}{\mu_0 H M_S}} \quad (4)$$

where A_{ex} is the exchange constant. At higher magnetizing field the Holstein-Primakoff paraprocess takes place and the linear dependence $\frac{M}{M_S} = f\left(\sqrt{\mu_0 H}\right)$ is observed

(Fig. 7). The validity of the relation (3) indicates that the quasi-dislocation dipoles are rather small and using the expressions (3) and (4) [10] the upper limit of the quasi-dislocation dipole width of 2.38 nm is obtained. The obtained results indicate that the atom packing density in the investigated alloy is relatively large.

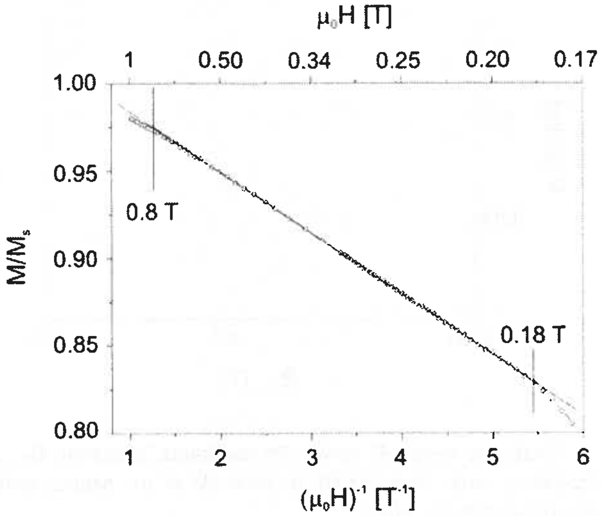

Fig. 6. Relative magnetization, M/M_s, versus $(\mu_0 H)^{-1}$ for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy

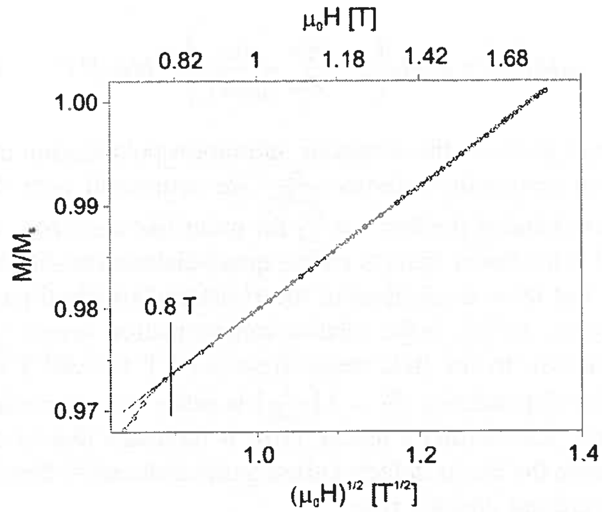

Fig. 7. Relative magnetization, M/M_s, versus $(\mu_0 H)^{1/2}$ for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy

4. Conclusions

- The nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy with small amount of a crystalline phase can be prepared directly by a rapid quenching using the suction casting method. The low quenching rate during the sample preparation enables structure relaxation which leads to the high density of atoms and only small structural defects in the form of quasi-dislocation dipoles 2.38 nm wide are present.
- the crystalline grains of the α-FeCo phase are small (25 nm) and their presence does not deteriorate the soft magnetic properties of the alloy.
- The initial magnetic susceptibility shows only a very broad maximum in the temperature range from 150 K up to 400 K and its disaccommodation in this temperature range is very low.
- The amorphous matrix is heterogeneous and exhibits two distincly separated Curie temperatures.
- The total losses for the nanocrystalline $Fe_{59}Co_{15}Zr_2Y_4Nb_5B_{15}$ alloy are comparable to those in the conventional Fe-Si crystalline alloys at low frequencies but they are smaller at high frequencies.

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