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Temperature evolution of magnetic susceptibility during devitrification of Cu-free HITPERM alloy

M. Varga [a], R. Varga [a,*], E. Komova [b], K. Csach [c], P. Vojtanik [a]

[a] Institute of Physics, Faculty of Science, P.J. Safarik University, Park Angelinum 9, 041 54 Kosice, Slovakia
[b] Faculty of Aeronautics, Technical University, Rampova 7, 041 21 Kosice, Slovakia
[c] Institute of Experimental Physics SAS, Watsonova 47, 040 01 Kosice, Slovakia

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ABSTRACT

Development of structural and magnetic properties of the $Fe_{44.5}Co_{44.5}Zr_7B_4$ alloy during its successive devitrification from amorphous to nanocrystalline state was investigated. Temperature independence of the initial susceptibility in a wide temperature range (20–600 °C) was observed. The origin of such susceptibility behavior was described through the compensation of lowering exchange interaction between nanocrystalline grains and their magnetization at increased temperatures. The stabilization of the domain structure, due to atom pair directional ordering, was confirmed by measurements of the amplitude dependence of susceptibility in a wide field range, where the initial susceptibility remains constant (Perminvar effect).

1. Introduction

Technical applications require materials with good soft magnetic properties (low coercivity, high permeability, and saturation magnetization). Moreover, these should be stable within a wide temperature and time range. Nanocrystalline alloys, which consist of nano-sized ferromagnetic crystals embedded in a residual ferromagnetic amorphous matrix, execute these properties perfectly [1]. Their good,soft magnetic properties arise from the exchange interaction between the crystalline grains through the amorphous matrix, which decreases an effective anisotropy [2]. FINEMET (Fe–Si–Cu–Nb–B) [1] and NANOPERM (Fe–Cu–M–B; M=Zr, Nb, Hf) [3] alloys complete these properties sufficiently, but the exchange coupling of the grains deteriorates above the Curie temperature of the amorphous matrix. Therefore, these materials have a low thermal limitation [4]. A new family of the nanocrystalline soft magnetic materials composed of Fe–Co–M–Cu–B (M=Zr, Nb, Hf), also named HITPERMS, produced by Willard et al. [5], offers larger magnetic induction values and higher Curie temperatures of the crystalline and amorphous component phases, due to the presence of Co. This makes them excellent candidates for various high temperature applications of the soft magnetic materials. An alloy, with a chemical composition of $Fe_{44.5}Co_{44.5}Zr_7B_4$, also named Cu-free HITPERM, has in the nanocrystalline state lower values of the coercive field (H_C) [6] than those of the original Cu-containing HITPERM materials [5,7]. It was shown by Johnson et al. [8] that the random crystalline texture of the nanocrystalline grains was not affected by field annealing.

This article presents the temperature evolution of the initial susceptibility of the Cu-free HITPERM amorphous and nanocrystalline alloy. The initial susceptibility of the FeCoZrB alloy exhibits temperature independence in a wide temperature range from 20 to 600 °C only in the nanocrystalline state. The origin of such behavior is mainly due to the compensation between lowering of exchange interaction anisotropy among the ferromagnetic grains embedded in the residual amorphous matrix of the nanocrystalline alloy and lowering saturated magnetization at increasing temperature.

2. Experimental

All measurements were performed on the amorphous and nanocrystalline $Fe_{44.5}Co_{44.5}Zr_7B_4$ ribbons of 5 cm length, 2 mm width, and 45 μm thickness, which were prepared by the melt–spinning method. Amorphous nature of the samples was checked by XRD. The demagnetizing factor was neglected. The nanocrystalline state was obtained by the controlled heat treatment. Thermomagnetic analysis measurements (TMA) are based on the measurement of temperature dependence of the initial susceptibility and resistivity. The heating and cooling rate was 10 K/min and the susceptibility was measured by the induction method. The applied field, generated during the thermomagnetic analysis, was less than 1 A/m, aiming to measure the susceptibility in a range, where the magnetization processes run through reversible

* Corresponding author.
E-mail address: rvarga@upjs.sk (R. Varga).

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movements of the domain walls in their potential wells [9]. The frequency of the applied magnetic field was around 11 kHz to avoid eddy currents. The resistivity was measured by the classical four-points method. The amplitude dependences of susceptibility were measured at room temperature, after cooling from the previous annealing temperatures (room temperature −600 °C), and after 10 min-long stabilization of the sample-demagnetized state. The amplitude of the applied magnetic field changed from 0.5 to 1000 A/m and the field frequency was 334 Hz. The thermal treatment was provided in protective argon atmosphere to avoid the oxidation of the sample.

3. Results and discussion

Fig. 1 shows temperature evolution of a relative resistivity, $\rho(T)/\rho_o$ (where $\rho(T)$ is the resistivity at given temperature and ρ_o is the resistivity of untreated amorphous sample), for the as-cast Cu-free HITPERM alloy, from room temperature to 550 °C, during successive heating–cooling cycles to higher temperatures each time. At first, the resistivity increases up to 200 °C (curve 1), as a result of a thermal activation of an atomic structure. Above 200 °C, the $\rho(T)/\rho_o$ curve decreases, which is accompanied by the structural relaxation. When the temperature of 300 °C was reached, we started the cooling of the sample. The resistivity after the heating–cooling cycle, at room temperature, is evidently lower than that in the initial state. This is caused by the homogenization of the structure and relaxation of internal stresses induced during the manufacture of the amorphous sample. Subsequent heating–cooling cycles up to 350 and 400 °C, respectively (curves 2 and 3) led to further structural relaxation. However, this was almost complete at 400 °C. Another annealing up to 450 °C (curve 4) did not produce the resistivity change; the material had a stable amorphous structure. Heating up to 500 °C (curve 5) led to the rapid decrease of the resistivity, due to the onset of primary crystallization of α′-FeCo phase [7]. The crystallization was confirmed also by the DTA measurement (inset of Fig. 1). Further annealing up to 550 °C (curve 6) was followed by the complete first crystallization of the sample, which finished at the temperature T=523 °C. After this treatment, the sample did not exhibit other structural changes, and the temperature dependence of the relative resistivity remains linear down to room temperature.

Magnetic system reflects the structural changes provided by the thermal treatment. Fig. 2 shows temperature evolution of a relative initial susceptibility, $\chi_i(T)/\chi_{io}$ (where $\chi_i(T)$ is the initial susceptibility at given temperature and χ_{io} is the initial susceptibility of the untreated amorphous sample), for the $Fe_{44.5}Co_{44.5}Zr_7B_4$ as-cast sample, during the same successive heating–cooling cycles as those given for the resistivity. During the initial heating (curve 1) up to 50 °C, an increase of the susceptibility occurred due to the thermal activation of magnetic moments, which increased the domain walls mobility. An anomalous decrease of the $\chi_i(T)/\chi_{io}$ curve is seen above this temperature, which originates from the diffusion of mobile point defects on atomic scale—an atom pair directional ordering (magnetic relaxation) [9,10]. Due to the thermal activation, the mobile atoms started to diffuse and stabilize the local magnetic moments, which caused domain structure stabilization (locally induced magnetic anisotropy in domains as well as the stabilization of domain walls in their positions), and consequently the decrease of initial susceptibility. The $\chi_i(T)/\chi_{io}$ curve increases again above the temperature T=250 °C, due to the start of the structural relaxation, reducing magnetoelastic and the induced uniaxial anisotropies. Further heating–cooling cycles up to the temperatures 350, 400 and 450 °C, respectively (curves 2, 3 and 4)

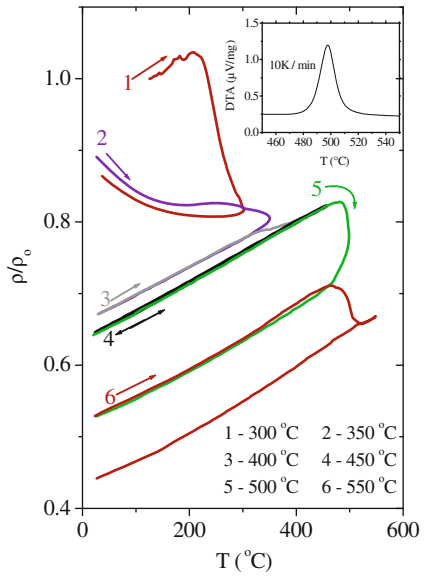

Fig. 1. Temperature dependences of relative resistivity for $Fe_{44.5}Co_{44.5}Zr_7B_4$ alloy. Inset shows DTA measurement at heating rate 10 K/min.

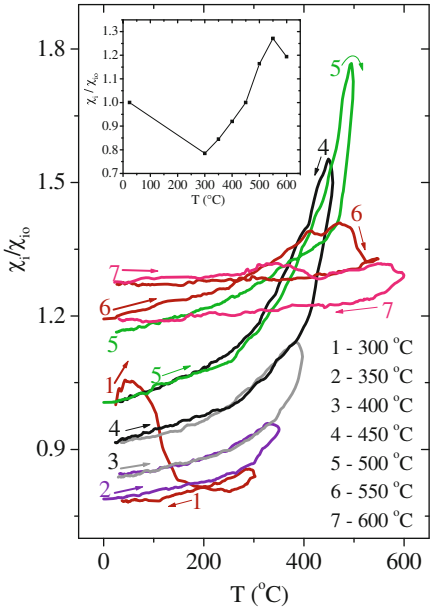

Fig. 2. Temperature dependences of relative initial susceptibility for $Fe_{44.5}Co_{44.5}Zr_7B_4$ alloy. Inset shows dependence of the relative initial susceptibility, measured at room temperature after successive heating–cooling cycles up to the temperature given in x-axis.

caused the onward structural relaxation, which is indicated by the further increase of the initial susceptibility after each cycle. The decrease of susceptibility during cooling is reversible and its temperature dependence is followed by the same curve in the subsequent heating. The susceptibility continuously increases up to the temperature T=495 °C (curve 5), at which the maximum value of $\chi_i(T)/\chi_{io}$ was obtained. The $\chi_i(T)/\chi_{io}$ curve, during further annealing up to 550 °C (curve 6), increases more linearly than in the previous cases. However, the curve begins to decrease at T=470 °C, due to the onset of the crystallization in the sample. On the other hand, the susceptibility temperature dependence during cooling is almost constant. The temperature independence of the $\chi_i(T)/\chi_{io}$ was not influenced by the subsequent heating up to