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Ultra-soft magnetic properties and giant magneto-impedance of $Co_{68}Fe_{4.5}Si_{12.5}B_{15}$

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ABSTRACT

We have prepared an amorphous $Co_{68}Fe_{4.5}Si_{12.5}B_{15}$ alloy, annealed it in the temperature range of 200–580 °C and carried out a detailed study of the effect of crystallization on its magnetic properties. When annealed in an optimized condition, a very high value of initial permeability of the order of ~10^4 has been attained in association with a drastic decrease of the relative loss factor. This change of properties has been attributed due to the formation of nanograins of fcc Co and Co_3B, as identified by X-ray diffraction and differential thermal analysis. The activation energy of crystallization is 4.18 eV. Hysteresis loop parameters were then extensively studied for the samples annealed at various temperatures. Finally, a very high value of giant magneto-impedance (GMI)—which is a characteristic property of Co-based amorphous alloys derived from well defined anisotropy axis (around 375) has been observed for a sample annealed at 380 °C.

1. Introduction

Thermal treatments of amorphous materials below their crystallization temperature relax the amorphous structure, giving rise to ultra-soft magnetic properties. Changes in the pre-existing anisotropy directions have been observed in some cases due to the rearrangement of the atom pairs during heat treatments, even in the absence of external magnetic field and applied stresses [1–3]. In recent studies, it has been found that heat treatments below the crystallization temperature in the Co-based amorphous alloy of different composition gave rise to the formation of nanocrystalline phase with grain size of about 2 nm, which has been revealed by Transmission Electron Microscopy (TEM) [1,3]. According to ref. [4], the formation of the nanocrystalline phase in the Fe-based alloy containing Cu annealed above the crystallization temperature, gives rise to reduction of the effective anisotropy because the random oriented anisotropies are averaged out by exchange interaction. Similar effect is believed to exist in the Co-based alloy in the early state of crystallization [5–7].

In this work, the aim is to analyze the possible formation of nanocrystalline phases and the relative softening of the magnetic properties in the amorphous alloy by appropriate thermal treatment. In order to study magnetic softness at different stages of annealing condition, nature of the hysteresis loops, coercivity and losses of Co-rich amorphous alloy have been studied. Further, geometry and annealing effect on magneto-impedance in amorphous relaxed state at current driving frequency of 4.5 MHz have also been investigated.

2. Experimental

The alloy with composition of $Co_{68}Fe_{4.5}Si_{12.5}B_{15}$ has been prepared by using Fe (99.98%), Si (99.9%) and B (99.5%) of Johnson Mathey and Co (99.8%) of Chempur Feinchemlkalien. The alloy in the form of ribbon with a thickness of 25 μm was prepared by the rapid quenching method. The samples were annealed at 13 different temperatures up to 530 °C for 30 min for studying the annealing effect on permeability. The permeability has been measured on toroidal shaped samples using HP 4192A impedance analyzer with an applied AC field of 10^{-3} Oe. The GMI effect is measured by using the GMI measuring setup with the applied magnetic field ranging from −300 to +300 Oe and operating frequencies of 4.5 MHz. Kinetics of crystallization of $Co_{68}Fe_{4.5}Si_{12.5}B_{15}$ has been studied by differential thermal analysis (DTA) in nitrogen atmosphere with continuous heating rate of 10–50 °C/min. Hysteresis loops have been measured using B–H loop tracer with the toroidal shaped samples.

3. Results

DTA curves presented in Fig. 1 show exothermic peaks which represent the formation of crystalline phases. For the heating rate of 10 °C/min, the initiation of crystallization is around 510 °C and the process of crystallization is completed at 542 °C while the peak temperature is 522 °C. Activation energy of crystallization has been obtained by using Kissinger's plot of $\ln\beta/T_p^2$ vs. $1/T_p$ presented in Fig. 2

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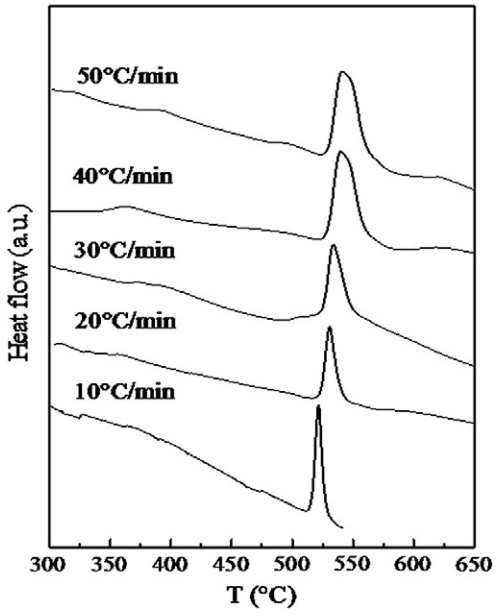

Fig. 1. Differential thermal analysis traces of as-cast sample for different heating rates.

and found as 4.18 eV. In this expression, β is the heating rate and T_p is the peak temperatures of the DTA curves.

In an attempt to identify crystalline phases, X-ray diffraction studies have been performed for samples annealed at different temperatures and the patterns are presented in Fig. 3. The phases are identified by using standard software and found as fcc-Co and Co_3B. Similar phases have been identified in ref. [8] by using TEM.

In Fig. 4, the real part of the complex initial permeability, μ′ vs. temperature, T curves have been presented for as-cast sample in order to determine the Curie temperature of amorphous sample. It has been found from Fig. 4 that μ′ decreases gradually and then there is a sharp increase of μ′ near the Curie temperature which is a characteristic feature of the Hopkinson effect. The Curie temperature has been estimated to be 343 °C for this alloy composition in the as-cast condition.

In Fig. 5, the real part of the complex initial permeability μ′ up to $f = 500$ kHz has been measured and presented for as-cast and annealed samples. The results refer to isothermal annealing time of 30 min. The measurement of permeability was performed with toroidal shaped samples at room temperature with an AC field of amplitude 10^{-3} Oe. In Fig. 5, it can be noted that the real part of permeability μ′, is fairly constant up to some frequency range and then falls rapidly.

In Fig. 6, annealing temperature dependence of the real part of permeability μ′ and relative loss factor $(\tan\delta)/\mu'$ have been presented for the frequency of 1 kHz in order to demonstrate the evolution of magnetic softness with annealing. The initial permeability value increases with the annealing temperature up to 480 °C and attains a maximum of 18,000. At this annealing temperature, relative loss factor $(\tan\delta)/\mu'$ is of the order of 10^{-6}.

Fig. 7 shows field dependence of the GMI ratio for as-cast and annealed samples of 0.1, 0.3, 0.5, 1, and 2 mm in width for 1 h at the frequency of 4.5 MHz. Maximum value of GMI has been observed as 240 for 0.3 mm width. In Fig. 8, field dependence of the GMI ratio has been presented for as-cast samples of 2, 3, 4, 5, 6, 7 and 8 mm in length for 1 h at the frequency of 4.5 MHz. Maximum value of GMI is 260 for 5 mm length.

In Fig. 9, field dependence of magneto-impedance ratio (MIR) has been presented for as-cast and conventional annealed samples at 300, 350 and 380 °C for 1 h at the frequency of 4.5 MHz. The sample dimension has been chosen as follows: length = 5 mm and width = 0.3 mm. MIR is defined as MIR (%) = 1 − Z (H_{DC})/Z (H_{max}), where H_{max} is the external DC magnetic field for obtaining the saturation value of magneto-impedance. For the present work the value of H_{max} is 300 Oe.

In Fig. 10, hysteresis loops of annealed samples have been presented for a current driving frequency of 1000 Hz. It can be observed from Fig. 10 that the shape of the hysteresis loops changes to a great extent at different stages of annealing. Variation of coercivity, H_c for the sample annealed at different temperatures is presented in Fig. 11 derived from the hysteresis loops presented in Fig. 10. For the sample annealed at 300 °C, H_c has been obtained as 25.2 A/m. Further increase of the annealing temperature to 480 °C, the H_c decreases to 9 A/m.

4. Discussions

It can be observed from Fig. 1 that the initiation of crystallization is around 510 °C for a continuous heating rate of 10 °C/min. For higher heating rates the phase transition temperature shifts towards higher values. From Kissinger's plot presented in Fig. 2, activation energy of crystallization has been found as 4.18 eV, which is higher than the activation energy of crystallization of the first primary peak of Fe-based alloy of FINEMET, which is normally 2.5 to 3.5 eV.

Crystalline phases that evolved through heat treatment have been studied by X-ray diffraction and presented in Fig. 3. In Fig. 3(f), XRD pattern has been presented for the annealing temperature of 550 °C where the crystallization is in the advanced stage. Nucleation of these crystalline phases has been initiated at much lower temperature and for the annealing temperature of 480 °C some of the similar phases just appeared with very small grain size. Buttino et al. [1] have carried out similar experiments in which they have annealed the samples just below the crystallization temperature and revealed devitrification of an amorphous matrix where grain size of about 2 nm was determined by TEM. From the information revealed by DTA traces presented in Fig. 1 and from the X-ray diffraction patterns presented in Fig. 3(a) and (b), it can be understood that for the annealing temperature of 450 °C or below the samples are in the amorphous relaxed states with broad diffused patterns which is characteristic to the amorphous materials.

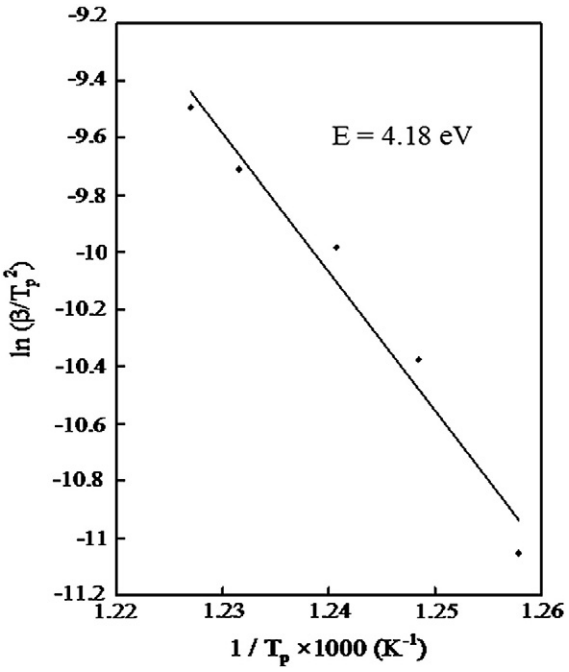

Fig. 2. Kissinger's plot for the determination of activation energy of crystallization.