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Microstructure-controlled enhancement of magnetoimpedance in nanocrystalline FeSiNbBCu alloys

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ABSTRACT

Rapidly solidified amorphous $Fe_{68.5}Si_{18.5}Nb_3B_9Cu_1$ ribbon has been subjected to heat treatment at a temperature of 550 °C for different time periods. All the annealed ribbons show the precipitation of nanocrystalline Fe_3Si phase from the amorphous phase. The estimated crystallite size from X-ray diffraction peak analysis was in the range of 15–25 nm. While the surface studies confirm the presence of a distribution of spherical nanostructures in amorphous matrix. Both magnetoimpedance and longitudinal permeability ratios are found to increase with annealing time, and attain a maximum value for 60 min annealed ribbon and decrease on further increase in the annealing time. The enhanced magnetic properties and magnetoimpedance on suitable heat treatment is attributed to the change of magnetic parameters such as anisotropy and magnetostriction, due to change in microstructure. Analysis of permeability and impedance data taken under similar conditions suggests a strong correlation between them.

1. Introduction

Nanostructured Fe-based magnetic materials have received considerable attention not only because of their unusual structural, electrical, magnetic and optical properties which are different from corresponding non-crystalline precursor alloys, but also they have wide range of applications, such as, power transformers, inductors, components for data communication interface, etc. [1–3]. These nanostructured alloys are composed primarily of nanocrystalline phases embedded in a residual amorphous matrix and are produced by the partial devitrification of amorphous alloys [4]. It is observed that some selected dopants are needed to promote massive nucleation through the addition of insoluble atoms such as Cu, Ag, etc. and to inhibit grain growth by adding refractory elements like Nb, Ta, etc. [5]. FINEMET, based on Fe–Si–B–Nb–Cu [5]; NANOPERM, based on Fe–M–B–Cu [5]; and HITPERM, based on Co(Fe)–M–B–Cu [5] alloys (where M = Zr, Ti, Ta, Nb, Hf, etc.) are examples of such materials which exhibit soft magnetic properties and find applications in magnetic heads and sensors based on the magnetoimpedance (MI) effect. The MI effect is a classical electromagnetic phenomenon, which is characterized by a large change in the total impedance of a magnetic conductor under the application of low static magnetic field at relatively high frequency [6,7]. It is now well established that the soft magnetic materials with slightly negative/zero value of magnetostriction (λ) present large values of MI, which corresponds to the large value of transverse permeability [8]. The conventional Fe-based materials have relatively large positive λ and low resistivity, whereas Co-based materials have low negative λ and high resistivity. On the other hand, almost zero λ has been achieved for nanocrystalline FINEMET (Fe–Si–Nb–B–Cu) type of alloys, by nullifying the positive λ of the residual amorphous matrix ($\lambda_{amorph} = 20 \times 10^{-6}$) with the negative contribution of α-Fe(Si) nanocrystalline phase ($\lambda_{nc} = -6 \times 10^{-6}$) [9,10]. These observations suggest that the magnetic properties can be tuned through microstructure modifications. It is well known that through an optimum heat treatment, the microstructure can be controlled and the relative volume fractions of nanocrystalline and amorphous phases can be varied in such a way that the effective λ becomes zero. That is,

$$\lambda_{effective} = (1 - \nu)\lambda_{amorph} + \nu\lambda_{nc} = 0 \quad (1)$$

where ν is the volume fraction of nanocrystalline phase. This will lead to enhancement of soft magnetic properties [11], and hence higher value of MI can be expected. On the other hand random anisotropy model (RAM), as applied to soft nanocrystalline ferromagnets, requires that the nanocrystallites be randomly oriented and have grain sizes smaller than the exchange correlation length. When these constraints are observed, effective magnetocrystalline anisotropy (K_{eff}) replaces the bulk magnetocrystalline anisotropy (K). Using coherent rotation model the estimated coercivity takes the form $H_C \sim K_{eff}/M_s = (K^4D^6)/(A^3M_s)$,

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where D and A are average grain size and exchange stiffness constant, respectively. Therefore, for grain sizes smaller than the exchange correlation length, the coercivity is proportional to the grain size of the sixth power.

In this report, the MI effect on the nanocrystalline $Fe_{68.5}Si_{18.5}Nb_3B_9Cu_1$ alloys has been investigated. As-quenched (AQ) amorphous ribbons were subjected to optimum annealing condition to maximize the MI effect. Present results show the maximum MI value reached 62% for the sample annealed at 550 °C for 60 min and strongly related to the permeability.

2. Experimental

An alloy of composition $Fe_{68.5}Si_{18.5}Nb_3B_9Cu_1$ was prepared by melting the pure elements in a vacuum induction-melting furnace. Around 30–40-μm-thick and 9-mm-wide amorphous ribbons were obtained by rapid solidification using melt-spinning technique under argon atmosphere [12]. The thermal stability of the amorphous ribbons was determined using a differential thermal analysis (DTA) technique (PerkinElmer, Pyris Diamond TG/DTA) at a scanning rate of 20 °C/min. The amorphous ribbon was then heat treated at 550 °C for 30, 60 and 90 min under high vacuum conditions (~10^{-5} mbar). The structure of as-quenched and annealed samples has been investigated using X-ray diffractometer (Philips PW 1710) with Co-K_α ($\lambda = 0.17889$ nm) radiation. The surface morphology of the ribbons was examined by using a field emission scanning electron microscope (FESEM) (Carl Zeiss, Supra 40). Saturation magnetization of all the ribbons was measured using a homemade vibrating sample magnetometer (VSM). The dc resistivity measurements were performed using the four-probe method. The MI measurements were carried out on 5-cm-long ribbon samples with Cu-leads connected on to the sample by using pressure contacts. The sample was then placed in a homogeneous magnetic field produced through homemade Helmholtz coil. The impedance measurements were carried out in a frequency range of 100 kHz–20 MHz. The applied ac current flowing across the sample was kept constant at 10 mA. The external field (H) was generated by the Helmholtz coil with axis perpendicular to the Earth's magnetic field to allow field variation from 0 to 76 Oe. The percentage change of impedance (i.e., MI ratio) with applied magnetic field is expressed as

$$\mathrm{MI} = \left(\frac{\Delta Z}{Z}\right)\% = \left(\frac{Z(H) - Z(H_{max})}{Z(H_{max})}\right) \times 100 \tag{2}$$

where H_{max} is the maximum applied magnetic field which is 76 Oe in the present work. The initial permeability and longitudinal permeability ratio (LPR) were measured by placing the ribbon in a small rectangular pick-up coil and by passing 10 mA ac current through the pick-up coil. Similarly, the percentage change of longitudinal permeability with applied magnetic field is expressed as

$$\mathrm{LPR} = \left(\frac{\Delta\mu}{\mu}\right)\% = \left(\frac{\mu(H) - \mu(H_{max})}{\mu(H_{max})}\right) \times 100 \tag{3}$$

All the ac measurements were performed using Agilent impedance analyzer (4294A) with 42941A probe at room temperature.

3. Results and discussions

The X-ray diffraction (XRD) pattern of the as-quenched $Fe_{68.5}Si_{18.5}Nb_3B_9Cu_1$ ribbon is shown in Fig. 1 (inset). The diffractogram exhibits a broad diffused halo around $2\theta = 53^\circ$ indicating the amorphous nature of sample. From the DTA experiments, it is found that the crystallization temperature is about 550 °C. However, it has been shown earlier that the annealing of the as-quenched ribbon in the temperature range of 525–575 °C leads to the development of Fe_3Si nanocrystalline phase with crystallite size of 10–12 nm in the amorphous matrix. This gives rise to the improved soft magnetic properties with small coercivity values [12,13]. Therefore, in the present study all amorphous ribbons were heat treated at 550 °C for 30–90 min. Fig. 1 also shows a typical XRD pattern of the sample annealed at 550 °C for 60 min revealing the formation of Fe_3Si crystalline phase with crystallite size in the range of 15–25 nm [14]. Fig. 2 shows the FESEM images of as-quenched and annealed samples for different time durations. It can be seen that the as-quenched sample exhibits featureless uniform background indicating the absence of crystalline phase, while the annealed samples show the nanocrystalline precipitates of about 25–40 nm size embedded in the amorphous matrix.

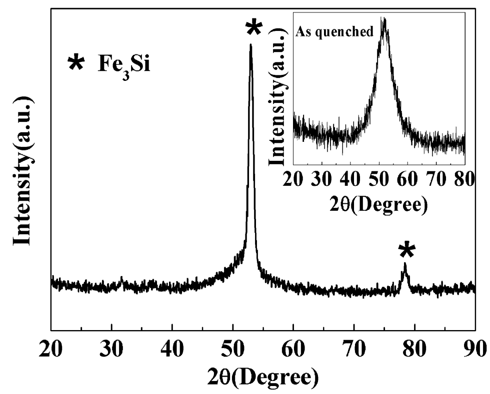

Fig. 1. X-ray diffraction pattern of as-quenched (inset) and 60 min annealed ribbons at 550 °C.

The initial permeability (i.e., permeability at zero fields) measurements have been performed by equivalent method and the real part of initial permeability for a wide spectrum of frequencies is shown in Fig. 3. It can be observed that the annealed ribbons show higher values of magnetic permeability than as-quenched samples in the measured frequency range. The increase in magnetic permeability indicates the enhancement of magnetic softness in the annealed ribbons, which can be attributed to the nucleation of nanocrystalline Fe_3Si grains. It is well known that if the size (D) of the nanocrystalline grains is less than the exchange correlation length (L_{ex}) (i.e., $D < L_{ex.}$), then the grains are magnetically coupled through exchange interaction with each other and the effective magnetocrystalline anisotropies are averaged out [15]. These observations are in good agreement with earlier observations [12].

The origin of magnetoimpedance is related to the permeability and its dependence on applied dc magnetic field. The special domain structure of the soft magnetic sample (ribbon) and different magnetization processes, domain wall motion and moment rotation cause the change in transverse magnetic permeability (μ_T) upon application of an external dc longitudinal magnetic field. At high frequency, μ_T affects the magnetic penetration depth (δ_m) through $\delta_m = (\rho/\pi\mu_T f)^{1/2}$, where ρ is the electrical resistivity and f is the frequency. It is worth noting that as the skin effect becomes dominant ($t/\delta_m \gg 1$, where t is the half thickness of the ribbon), the impedance $Z \propto (f\mu_T)^{1/2}$ [16]. However, it has been proposed that the change of $(\Delta Z/Z)\%$ is closely related to longitudinal permeability ratio $(\Delta\mu/\mu)\%$ in the high-frequency region [17]. Hence, the evolution of magnetoimpedance can be realized either by $\Delta Z/Z$ measurements or $\Delta\mu/\mu$ measurements.