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Effect of Annealing Temperature on the Permeability and Magneto-Impedance Behaviors of $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ Amorphous Alloy

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The effect of annealing temperature on the permeability and giant magneto-impedance (GMI) behaviors of $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ amorphous alloy has been systematically investigated. The nanocrystalline $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ alloys consisting of ultra-fine $(Fe,Mn)_3Si$ grains embedded in an amorphous matrix were obtained by annealing their precursor alloy at the temperature range from 500°C to 600°C for 1 hour in vacuum. The permeability and GMI profiles were measured as a function of external magnetic field. It was found that the increase of both the permeability and the GMI effect with increasing annealing temperature up to 535°C was observed and ascribed to the ultrasoft magnetic properties in the sample, whereas an opposite tendency was found when annealed at 600°C which is due to the microstructural changes caused by high-temperature annealing. The study of temperature dependence on the permeability and GMI effect showed some insights into the nature of the magnetic exchange coupling between nanocrystallized grains through the amorphous boundaries in nanocrystalline magnetic materials.

Key words : magnetoimpedance, amorphous, nanocrystalline materials, annealing, incremental permeability

1. Introduction

Recently, the nanocomposite materials represent a very interesting object for basic investigations and application researches because of their magnetically two-phase structure with a variety of macroscopic magnetic properties [1]. Among these nanocomposite ferromagnets, produced by primary crystallization of amorphous alloys, the widely studied $Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$ alloy commercially known as FINEMET [2], exhibits excellent magnetic properties. The optimum nanocrystallized state is obtained by isothermal annealing of the as-cast amorphous ribbon above its dynamic crystallization temperature, typically in the range from 773 to 818 K for 1 hour [1-3]. After such a heat treatment the material shows a uniform structure of ultrafine crystallites (bcc FeSi) with average diameter of 10-20 nm embedded in residual amorphous matrix. Structural changes, induced by annealing, modify the macroscopic magnetic behavior of the constituent material. The microstructure dependence of the magnetic properties was explained by the random anisotropy model, which is proposed for amorphous ferromagnets by Alben *et al.* [4] and developed by Herzer *et al.* [5]. According to this model, when the grain size is less than the ferromagnetic exchange length (L_{ex}), the exchange interaction dominates the anisotropy energy and forces the magnetization vectors to be parallel to each other over several grains. Under this condition, the effective anisotropy is averaged out and thus it leads to good soft magnetic properties, low coercivity and very high permeability. This indicates that the exchange interaction between the nanocrystalline and amorphous phase plays an important role in achieving good soft magnetic properties. However, the many questions concerning the nature of the magnetic exchange coupling in a nanocrystalline magnetic material remain to be addressed.

It has been well-established that the change of a magnetic permeability and impedance as a function of an external magnetic field has brought much interest in the

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basically physic understanding, the applications for magnetic recording heads and magnetic sensors, etc. Furthermore, a number of recent studies on the giant magneto-impedance (GMI) effect in Fe-based amorphous soft magnetic alloys subjected to heat treatment showed some insights into the nature of the magnetic exchange coupling between these grains through the amorphous boundaries in Fe-based nanocrystalline materials [15, 18-19].

Several continuing attempts have been made to improve the soft magnetic properties of Finemet-type alloys either by altering the average distance between the nanograins with the help of suitable heat treatments or by tailoring the Curie temperature of the amorphous phase through modifying the composition of the precursor alloy. Especially, some reports indicated an outstanding role of the Mn atoms, which is substituted for Fe in Finemet-type alloy, in the evolution of exchange coupling between the crystalline and residual amorphous phases [7, 16, 17]. Therefore, we selected the $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ amorphous alloy for the present study. However, the magnetic properties of this alloy are strongly dependent on thermal treatments (i.e., annealing temperature).

In present work, therefore, we have investigated the influence of annealing temperature on the permeability and magneto-impedance effect in $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ amorphous alloy.

2. Experiment

Amorphous ribbon with nominal composition $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ of about 8 mm in width and 20 μm in thickness were produced from ingots using the standard single copper wheel melt spinning technique. The $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ nanocrystalline samples consisting of ultrafine grains embedded in an amorphous matrix were obtained by annealing their amorphous alloys at the temperature range from 500°C to 600°C for 1 hour in vacuum. The schematic diagram of the vacuum annealing system where a high frequency driving current is applied to the longitudinal direction of the sample can be found elsewhere [8].

Magneto-impedance and permeability measurements were carried out along the ribbons axis under a dc longitudinal applied magnetic field. The samples with a length of about 15 mm were used for all measurements. A computer-controlled RF signal generator with its power amplifier was connected to the sample in series with a resistor for monitoring the driving ac current. The ac current and the voltage across the sample, for calculating the impedance, could be measured by using digital multimeters (DMM) with RF/V probes. The external dc field, applied by a solenoid, was swept through the entire cycle equally divided by 800 intervals from −300 Oe to 300 Oe. The frequency of the ac current, through the samples, was varied from 1 to 10 MHz, while its amplitude was varied from 10 to 30 mA. We have measured the change of the permeability as a function of external dc field using MI measurement system by replacing the MI probes with a set of primary and secondary coils located in the solenoid [9].

3. Results and Discussion

The permeability ratio (PR) can be defined as PR (H) = $\Delta\mu/\mu(H_{max})=1-|\mu(H)/\mu(H_{max})|$, where H_{max} is maximum applied magnetic field. In present experiment H_{max}=300 Oe. It is pointed out that the permeability of soft magnetic alloys is one of basic properties to estimate the magnetic softness. The incremental permeability (biased permeability) is the slope on a point in the B-H loop and responds sensitively to the dc magnetic field. Therefore, the incremental permeability can be changed drastically as a function of an external field in the ultrasoft magnetic material. Additionally, the behavior of the incremental permeability is directly related to the MI effect. The MI effect at high frequency can be explained in terms of an external magnetic field dependence of impedance as a result of the transverse magnetization with respect to the current direction flowing through the sample and the skin effect of an ac current. The MI is well-known to be proportional to $(\omega\mu_t)^{1/2}$ where μ_t is the magnetic permeability in the transverse direction and depends on dc magnetic fields and the magnetic anisotropy. Therefore, the MI effect has been observed only in very soft magnetic materials. In summary, it should be noted that the GMI effect can be achieved in such magnetic materials where the permeability (μ_t) for a planar magnetic film (i.e., a magnetic ribbon) is large and the magnetic penetration depth (δ_m) and the electric resistance (R_{dc}) are small.

First, we measured the PR curves as a function of longitudinal dc external magnetic field at various frequencies from 1 MHz up to 5 MHz for the as-quenched $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ alloy as displayed in Fig. 1. It can be seen that the PR curves exhibit a maximum peak at nearly zero field ($H_{dc}\sim 0$), the magnitude of PR decreases with increasing frequency while the shape of the PR curves becomes broader. For this, because the external magnetic field is a hard axis field with respect to the circumferential anisotropy, the magnetic field applied along the ribbon axis will suppress the circular magneti-

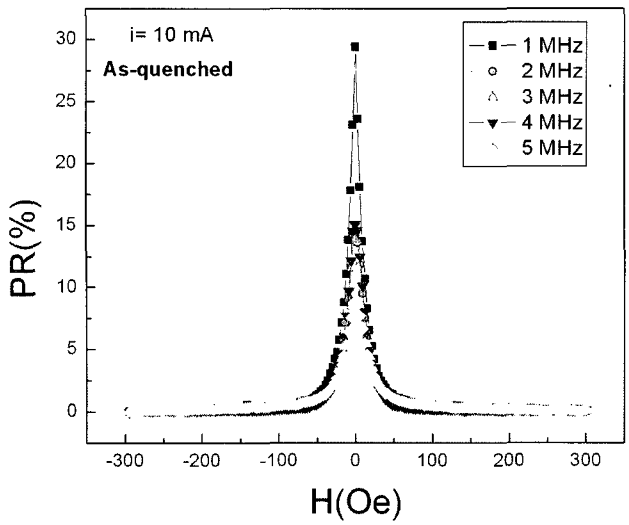

Fig. 1. PR curves measured as a function of the external magnetic field at various frequencies for the $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ as-quenched alloy.

zation by domain wall movements at low frequencies, or the motion of localized magnetic moments at high frequencies. Therefore, as the external magnetic field increases, the circumferential permeability or transverse permeability decreases rapidly. As a result, the maximum peaks of the PR curves are observed at around zero magnetic field [10]. Besides, some reports [8-10] indicated that the magnetic permeability changes sensitively with the annealing temperature and the measuring frequency. Figure 2 shows the PR curves measured at various frequencies for the $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ samples annealed at different temperatures between 500°C and 600°C. It was found that the maximum value of PR increases with increasing annealing temperature up to 535°C and then decreases at higher temperatures. This is likely ascribed to the microstructural variations as the annealing temperature is increased For a instance, the PR curve of the nanocrystalline $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ sample annealed at 535°C for 1 hour (see Fig. 2b) exhibit-

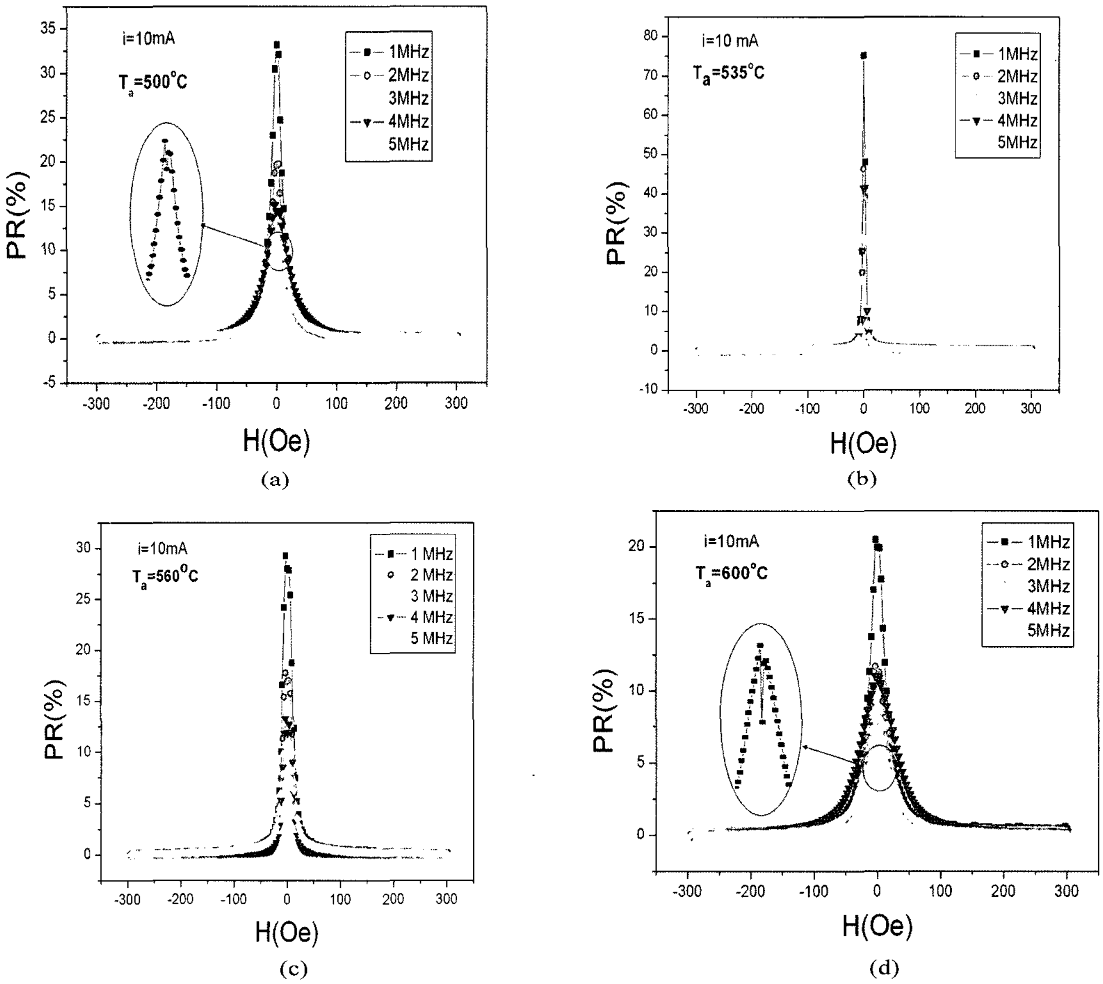

Fig. 2. PR curves measured as a function of the external magnetic field at various frequencies for the $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ alloys annealed at 500°C, 535°C, 560°C, and 600°C for 1 hour in vacuum, respectively.

ed the largest sharpness and reached the maximum PR value (~75%). This is mainly due to the improvement of soft magnetic properties in the sample which is caused by the appearance of ultra-fine $(Fe,Mn)_3Si$ nanograins (~7 nm from the XRD calculation), in which magnetocrystalline anisotropies are averaged out, therefore nanocrystalline grains are strongly coupled though magnetic exchange interactions. It should be noted that the growth of these nanoparticles to an optimum size and the volume fraction of nanoparticles led to their soft magnetic properties. Furthermore, the increase of permeability is simultaneously ascribed to the decrease in the magnetocrystalline anisotropy and the saturation magnetostriction upon a proper thermal treatment. These lead to the enhancement of soft magnetic properties in the sample [7]. When annealing temperature was relatively high over 535°C caused a rapid decrease of the permeability indicating a large degradation of the soft magnetic properties. This is likely related to the formation of strongly magnetocrystalline anisotropic phases likes Fe-borides (i.e., Fe_2B or Fe_3B) which deteriorate soft magnetic properties. These results are good in agreement with obtained results and reported *et al.* [6, 15]. When annealed at 600°C (see Fig. 2d), the PR curves become broader and the maximum PR value decreases drastically (~20%). Obviously, the PR profiles showed two-peak characteristics at $f = 5$ MHz as observed in Figs. 2a and 2d. This is due to the change in the switching and anisotropy field of the sample with increasing frequency [11]. The increase of the switching and the anisotropy field with frequency causes a two-peak behavior in PR profiles together with a shift of the maximum peak towards a higher value of H_{dc}. Additionally, the broadening of the PR curves with increasing frequency can be explained by adapting a model for a transversely biased permeability in thick ferromagnetic films where eddy current damping and the ripple field H_R incorporated with the anisotropy field H_k give rise to a permeability peak at an external field, as well as to the broadening of the PR curves at high frequencies [12].

The magnetoimpedance ratio (MIR) can be defined as MIR $(H) = \Delta Z/Z(H_{max}) = 1-|Z(H)/Z(H_{max})|$, where H_{max} is an external magnetic field sufficient for saturating the magnetoimpedance. In present experiment H_{max}=300 Oe. Based on results from permeability analyses, it has been shown that the nanocrystalline sample annealed at 535°C for 1 hour has the largest magnetic permeability. Consequently, this nanocrystalline sample is expected to have noticeable MI effect due to its excellent soft magnetic properties.

Figure 3 shows the external dc magnetic field dependence of the MIR at various frequencies up to $f = 10$ MHz.

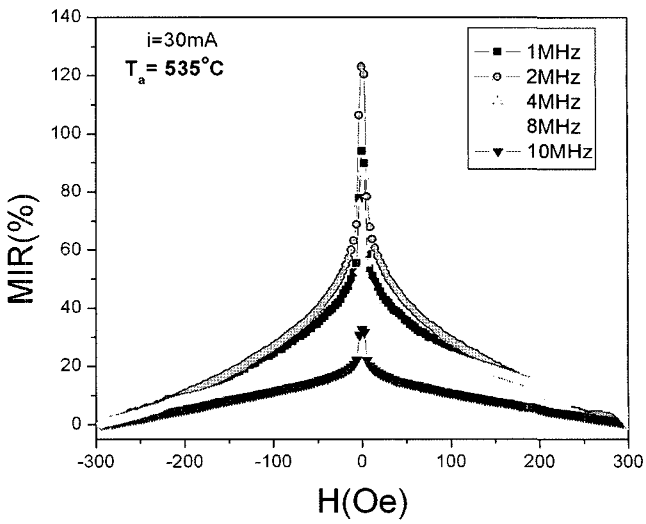

Fig. 3. MIR curves measured as a function of the external magnetic field at various frequencies for the nanocrystalline $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ sample annealed at 535°C for 1 hour.

As can be seen clearly in Fig. 3, the GMI profile had a single-peak feature at near zero field ($H_{dc} \sim 0$), and the maximum value of MIR increases with increasing frequency up to 2 MHz, and then decreases at higher frequencies. In this experiment, the maximum value of MIR reached the highest value of 130% at $f = 2$ MHz which is ideal for quick-response GMI sensor applications. Accordingly, the higher MIR value observed at $f = 2$ MHz is likely due to the presence of its special domain structure as transverse domains formed by a magnetomechanical coupling between internal stress and magnetostriction which increased the transverse magnetic permeability of the sample and hence GMI value [13]. In order to qualitatively interpret the frequency dependence of the impedance, a model of the skin effect for thin ribbons is adopted [14]. At frequencies below 1 MHz (i.e., $a < \delta_m$, where a is the thickness of the ribbon), the maximum MIR value was relatively low due to the contribution of the magneto-inductive voltage to MI which is mainly ascribed from the circular magnetization process. When 1 MHz $\leq f \leq$ 4 MHz ($a \sim \delta_m$), the skin effect becomes dominant, where both the domain wall displacements and the magnetization rotation contribute to the change in the circular permeability and hence skin effect, consequently a higher MIR value was found.

In order to assess the annealing-temperature dependence of GMI effect which is linked to the permeability, we investigated the variations of maximum MIR (denoted as MIR_{max}), measured at $f = 2$ MHz for all samples, as a function of annealing temperatures and displayed in Fig.

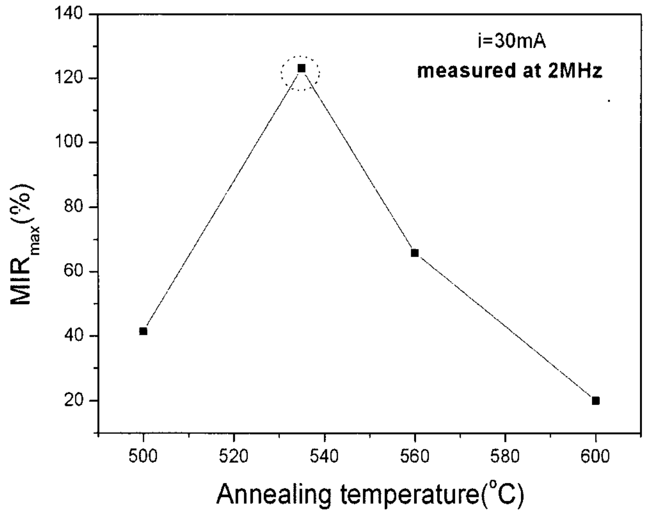

Fig. 4. The annealing temperature dependence of maximum values of MIR measured at $f = 2$ MHz for the $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ nanocomposites.

4. It is clear that, with increasing temperature up to 535°C, an increase of MIR_{max} was found but an opposite behavior was observed when the annealing temperature exceeded 535°C. For an example, when annealing at high-temperature ($T_a = 600$°C), besides the additional phases are formed like Fe-boride phases, an increase of mean size of $(Fe,Mn)_3Si$ grains (~13.7 nm) was found. These decreased the magnetic exchange coupling between the grains and accordingly the drastic decrease of both the permeability and the GMI effect was observed.

3. Conclusions

The influence of annealing temperature on the permeability and magneto-impedance behaviors of $Fe_{68.5}Mn_5Si_{13.5}B_9Nb_3Cu_1$ amorphous alloy has been investigated. The increase of both the permeability and the GMI effect with increasing annealing temperature up to 535°C was observed and ascribed to the microstructural changes caused by the proper heat treatments, whereas an opposite tendency was found when annealed at higher temperatures (over 535°C). It is worth mentioning that a large variation in the magnetic nature of the amorphous phase upon annealing temperature changed the intergrain exchange coupling, consequently altered both the permeability and the GMI features.

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