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OPTIMIZATION OF SOFT MAGNETIC PROPERTIES IN NANOCRYSTALLINE GLASS-COATED MICROWIRES

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Abstract

We have studied correlation of magnetic properties, structure and Giant Magnetoimpedance (GMI) effect, of Finemet-type FeCuNbSiB microwires. We have observed that GMI effect and magnetic softness of glass-coated microwires produced by the Taylor-Ulitovski technique can be tailored either controlling magnetoelastic anisotropy of as-prepared FeCuNbSiB microwires or controlling their structure by heat treatment. We have observed considerable magnetic softening of studied microwires after annealing. This magnetic softening correlates with the devitrification of amorphous samples. Amorphous Fe-rich microwires exhibit low GMI effect (GMI ratio below 1%). Considerable enhancement of the GMI effect (GMI ratio up to 90%) has been observed in heat treated microwires with nanocrystalline structure. We believe that FINEMET-type glass-coated microwires with higher saturation magnetization are good candidates for GMI sensor and metacomposites applications.

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1.Introduction

Glass-coated magnetically soft microwires are recognized as one of the most promising soft magnetic materials for applications in magnetic sensors, electronic surveillance, security, etc. [1-4]. Usually modified Taylor-Ulitovsky technique consisting of rapid quenching and drawing of composite microwire consisting of metallic nucleus (with diameter, d, from 1 up to 30 μm) and glass coating sheath (with thickness from 2 to 10 μm) is employed for glass-coated microwires preparation.

Like in other families of amorphous materials (ribbons and wires) the absence of magneto-crystalline anisotropy and defects typical for crystalline materials are considered as the main reasons of extremely soft magnetic properties [3-5]. Consequently magnetoelastic and shape anisotropies of amorphous materials in most case play the most important role in formation of their magnetic properties. Particularly if the microwire diameter is about 120 μm (typical for wires produced by the “in-rotating water”method), the sample length affects the hysteresis loop [4, 6]. The main advantages of glass-coated are small diameters, perfectly circular symmetry and almost continuous technological process (allowing producing continues microwires up to few kilometers long), the simple and cheap processes. But elevated internal stresses induced by the simultaneous rapid solidification of metallic nucleus surrounded by the glass coating produce additional magnetoelastic anisotropy. The origin of internal stresses is related with the difference between the thermal expansion coefficient of the metallic nucleus and the glass coating as well as from the solidification from the outer shell [1, 6, 7].

Less expensive Fe-rich microwires are preferable for applications, but amorphous Fe-rich materials exhibit rather high magnetostriction coefficient [4, 6]. Consequently Fe-rich amorphous microwires usually present rather low GMI effect [6, 8]. Enhancement of magnetic softness of Fe-rich amorphous materials and decreasing of the magnetostriction constant is possible by controllable crystallization of amorphous Fe-rich materials allowing the creation of two-phase nanocrystalline alloys [9-11]. This nanocrystallization has been achieved in conventional Fe-rich (FeSiB) amorphous alloys with small additions of Cu and Nb (so called FINEMET composition) after annealing in the range of 500-600^{o}C for 1 hour (i.e., at temperatures between the first and second crystallization peaks). After partial crystallization, such material consists of small (about 10 nm average grain size) nanocrystalites embedded in the residual amorphous matrix and exhibits excellent soft magnetic properties. Magnetic softness is usually explained considering the vanishing magnetocrystalline anisotropy and quite low magnetostriction coefficient values when the grain size approaches 10 nm [9, 10].

One of the most promising and relevant applications of soft magnetic amorphous materials is related to the Giant Magneto Impedance (GMI) effect consisting of a large change of the electrical impedance of soft magnetic conductor subjected to an axial *dc* magnetic field, H [3,11-13]. The origin of the GMI effect is well-explained from the dependence of the transverse magnetic permeability upon the *dc* magnetic field and skin effect. Large GMI effect (up to 600%) has been reported for Co-based amorphous glass-coated microwires with vanishing magnetostriction coefficient [14, 15]. Extremely high magnetic field resolution observed in amorphous wires with GMI effect attracts attention of engineers for creation of sensitive and cheap magnetic sensors and magnetometers [16-19]. For such applications glass-coated microwires with reduced dimensions are quite desirable. But only few reports deal with optimization of magnetic properties by nanocrystallization of Fe-rich microwires [20-23]. Previously reported GMI ratio values in FINEMET-type microwires are much lower than reported for amorphous microwires or

FINEMET-type ribbons [8, 11, 14]. Considerable GMI effect in nanocrystalline glass-coated microwires has been reported very recently [24, 25].

Consequently we report on our studies of annealing conditions on magnetic properties and GMI effect of FINEMET-type microwires.

2. Experimental details

For preparation of FINEMET-type glass-coated microwires we employed modified Taylor-Ulitovsky and/or quenching-and-drawing method described elsewhere [4, 8, 24]. This method essentially consists of a simultaneous drawing of the composite microwire (metallic nucleus inside the glass capillary) through the quenching liquid (water or oil) jet onto rotating bobbins. More detailed description of the fabrication method can be found elsewhere [4]. The microstructure of a microwire and hence, its properties, depends mainly on the cooling rate, which can be controlled by a cooling mechanism when the metal-filled capillary enters into a stream of cooling liquid water on its way to the receiving coil. Composition and geometries of the studied microwires are collected in Table 1.

We have measured dependences of both diagonal Z_{zz} and off-diagonal $Z_{\varphi z}$ impedance components and the GMI ratio, $\Delta Z/Z$, on external axial magnetic field *H*, as described elsewhere [26].

The magneto impedance ratio, $\Delta Z/Z$, has been defined as:

$$\Delta Z/Z = [Z(H) - Z(H_{max})]\cdot 100/Z(H_{max}), \qquad (1)$$

An axial *DC*-field with maximum value, H_{max}, up to 8 kA/m was supplied by magnetization coils.

We use a specially designed microstrip cell previously described elsewhere [26]. The components Z_{zz} and $Z_{\varphi z}$ were measured simultaneously using a vector network analyzer. The frequency range for the diagonal impedance component has been measured till 7 GHz.

Table 1. Composition of the metallic nucleus, geometry, and the total diameter of FINEMET glass-coated microwires

Sample Composition	Metallic nucleus diameter d (μm)	Total diameter D (μm)	Geometric ratio ρ- ratio
$Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$	14.2 μm	23.0 μm	0.62
$Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$	11.4 μm	15.2 μm	0.75
$Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$	12.8 μm	15.8 μm	0.81
$Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$	15.6 μm	21.8 μm	0.72
$Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$	11.8 μm	14.4 μm	0.81
$Fe_{72.3}Cu_1Nb_{3.1}Si_{14.5}B_{9.1}$	10.5 μm	27.8 μm	0.38
$Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$	11.4 μm	19,2 μm	0,59

Hysteresis loops have been determined by flux-metric method described elsewhere [4, 8]. Structure and phase composition have been checked using a BRUKER (D8 Advance) X-ray diffractometer with Cu K_α (λ=1.54 Å) radiation.

3 Experimental results and discussion

Most of studied as-prepared microwires present amorphous structure. Usually, the basic method to obtain nanocrystalline structure from the amorphous state is to control the crystallization kinetics by optimizing the heat treatment conditions (annealing temperature, annealing time, heating rate, etc.). The evolution of structural and magnetic properties has been studied for each sample at different annealing temperatures, T_{ann}, in the range between 450-550°C for 1 hour in order to investigate the devitrification process (Fig. 1). As-prepared and annealed at $T_{ann} \leq 450°C$, $Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$ microwires present amorphous structure. Increasing the T_{ann}, a noticeable growth of the intensity in X-ray diffraction, XRD, patterns is observed, which reveals the beginning of the crystallization at annealing of the sample. Starting from 550°C a main crystalline peak is appearing in the range between 42° to 45° which is correspond to the existence of α-Fe (Si) BCC crystal structure [9-11, 21-25].

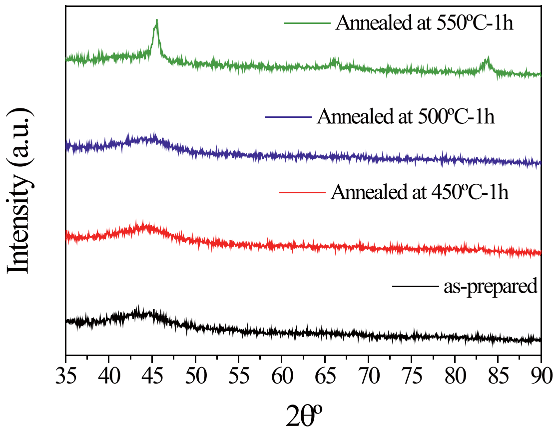

Fig. 1 XRD patterns of as- prepared and annealed $Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$ microwires with ρ = 0.59.

Strong dependence of the magnetic properties on the sample's geometry in as-prepared state has been observed for all compositions. As it can be seen from Fig. 2 (a), for the $Fe_{72.3}Cu_1Nb_{3.1}Si_{14.5}B_{9.1}$ sample the coercivity strongly increases as the ρ- ratio decreases. These results could be explained, taking into account, that the internal stresses acting into the metallic nucleus increase as the geometric ratio decreases because the thickness of the glass coating acts on the metallic nucleus to be responsible mainly of those internal stresses. Similarly to other Fe-rich amorphous microwires, all studied microwires in as-prepared state present rectangular hysteresis loop (Fig.2 (b)).

After annealing a considerable magnetic softening manifested as decreasing of the coercivity, H_c, is observed (see Fig.3). This magnetic softening correlates with the devitrification process observed by XRD.

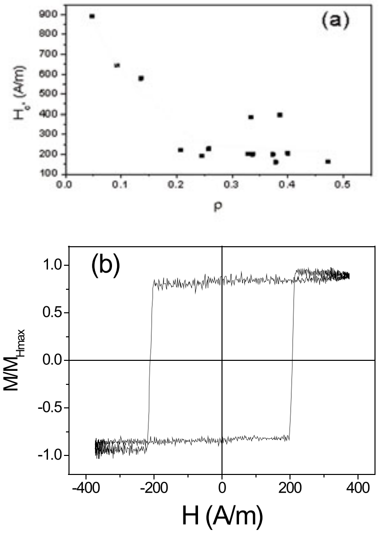

Fig. 2 Dependence of coercivity on ρ-ratio measured in as-prepared $Fe_{72.3}Cu_1Nb_{3.1}Si_{14.5}B_{9.1}$ microwires (a) and hysteresis loop of $Fe_{72.3}Cu_1Nb_{3.1}Si_{14.5}B_{9.1}$ microwire with ρ=0.38 (b).

As it was expected, samples with amorphous structure in as-prepared state exhibit rather small GMI (generally $\Delta Z/Z$ below 1-2%, see Figs. 4a, 4b) similarly to other Fe-based glass-coated microwires with positive magnetostriction [24, 25]. Low GMI effect of as-prepared amorphous Fe-based microwires must be related with high positive magnetostriction coefficient. Consequently the internal stresses distribution usually

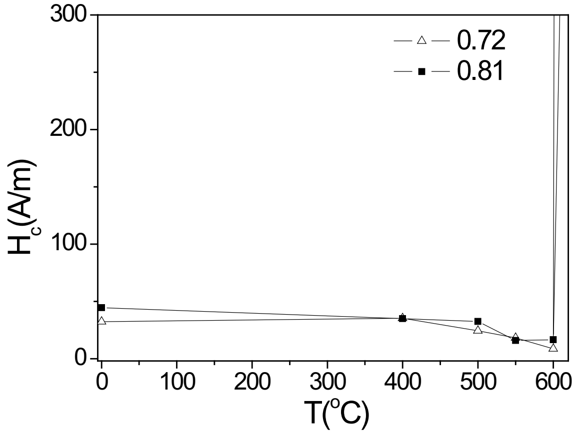

Fig. 3 Dependence of the coercivity of $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ glass-coated microwire with different ρ-ratio on annealing temperature.

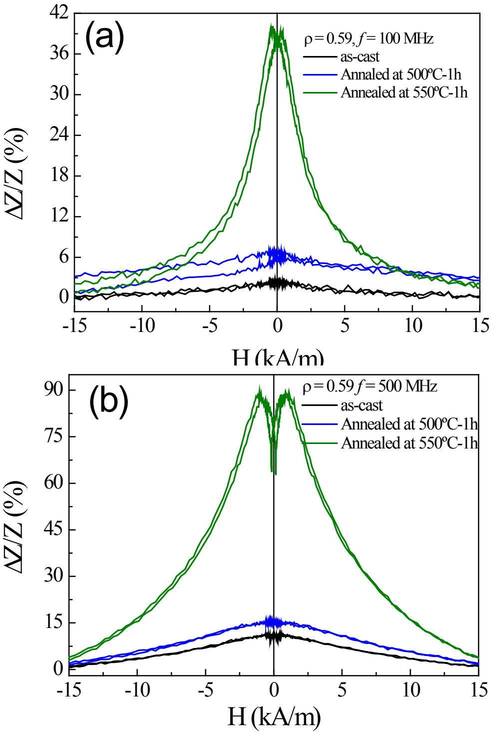

Fig.4 Effect of annealing at 550ºC for 1 hour on ΔZ/Z (H) dependence of $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ microwires with ρ = 0.59 measured at 100 MHz (a) and 500 MHz (b).

results in a longitudinal easy axis in the inner part of the metallic nucleus and radial easy axis in the outer shell of metallic nucleus. The sample with this domain structure usually exhibits a small GMI effect, because such domain structure presents low circumferential magnetic permeability. After annealing we observed considerable GMI effect improvement (see Figs 4). The highest GMI ratio has been observed in samples annealed at 550°C, where enhancement of the ΔZ/Z ratio (up to 90%) has been observed in case of $Fe_{70.8}Cu_1Nb_{3.1}Si_{14.5}B_{10.6}$ microwire with ρ = 0.59. Such enhancement must be related to its nanocrystalline structure that consists of α-Fe(Si) grains embedded in the amorphous matrix as discussed before.

It should be noted (as can be appreciated from Fig.4) that the maximum GMI ratio, $\Delta Z/Z_{max}$ is affected by the frequency and can be optimized choosing appropriate frequency [12, 13, 24, 25].

As mentioned above the skin effect of magnetic conductor is the main origin of the GMI effect [12, 13]. The penetration or skin depth, δ, is given by the well known expression;

$$\delta = (\pi\sigma\mu_0 f)^{-1/2} \qquad (2)$$

where, *f*, is the frequency of the current, σ is the electrical conductivity of the material, and μ_0 is the circular magnetic permeability.

One of the conditions for observation of the GMI effect is that the skin depth must be reduced below the wire radius. Despite this, the frequency, at which the maximum GMI value is obtained, depends strongly on the sample geometry, i.e, the ρ-ratio [27].

We have performed studies of frequency dependence of $\Delta Z/Z_{max}$. Comparative results obtained in $Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$ microwires with different ρ-ratios annealed at different conditions are displayed in Fig. 5. Similarly to amorphous Co-rich microwires one can conclude that ΔZ/Z and its frequency dependence are strongly affected by the magnetoelastic anisotropy determined by the ratio ρ.

Consequently we were able to improve GMI effect from few (1-2) % to 90% after devitrification of FINEMET-type FeCuNbSiB microwires. It is worth mentioning, that the obtained nanocrystalline microwires are more brittle than the amorphous microwires. Certainly this limits the applications of obtained samples. On the other hand there are few applications where cut microwires are used, i.e. tuneable metamaterials with embedded magnetically soft cut microwires [2, 28]. We believe that less expensive FINEMET-type glass-coated microwires with higher saturation magnetization are good candidates for tuneable metamaterials applications.

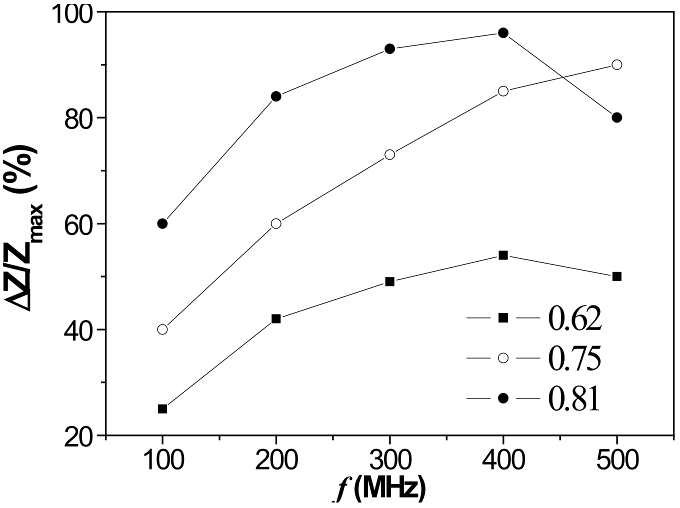

Fig. 5 Frequency dependence of $Fe_{70.8}Cu_1Nb_{3.1}Si_{16}B_{9.1}$ microwires with different geometric ratios ρ annealed at different conditions.

4. Conclusions

We have observed considerable increasing of the GMI effect after devitrification of FINEMET-type FeCuNbSiB glass-coated microwires. Moreover the magnetic properties of as-prepared microwires as well as the GMI ratio are affected by the magnetoelastic anisotropy related to internal stresses. A magnetic softening and a considerable enhancement of the GMI effect in FINEMET-type FeCuNbSiB with nanocrystalline structure have been observed. The frequency dependence of maximum GMI ratio, $\Delta Z/Z_{max}$, has been analyzed. We have observed the optimum frequency range for studied microwires with different ρ-ratios. We believe that FINEMET-type glass-coated microwires with higher saturation magnetization are good candidates for GMI sensors and metacomposites applications.

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