Development of Amorphous Microwires with Improved Magnetic Softness and High Giant Magnetoimpedance Effect

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Abstract: - We provide our last results on studies of the annealing influence on the magnetic properties and Giant Magneto-Impedance, GMI, effect of Co-rich microwires. After appropriate annealing, we observed a remarkable GMI ratio improvement up to 735%. Observed high GMI ratio and magnetic softening of studied microwires is discussed in terms of the internal stresses relaxation and the effect of annealing on the magnetostriction coefficient sign and value.

Key-Words: - magnetic wire, amorphous materials, Giant Magneto-Impedance, magnetic softness, hysteresis loops, magnetic anisotropy.

Received: April 6, 2024. Revised: November 15, 2024. Accepted: December 21, 2025. Published: March 26, 2025.

1 Introduction

Magnetic wires with crystalline structure and soft magnetic properties have attracted substantial attention along many years [1]. However, soft magnetic properties of crystalline materials are substantially affected by the crystalline structure, such as grain size, crystalline texture, grain boundaries, dislocations density etc. [2]. While, amorphous wires are characterized by the liquid-like structure without such defects [2-4].

The principle advantage of amorphous magnetic wires is good magnetic softness, which typically observed even in as-prepared samples. In addition, superior mechanical and corrosion properties are observed in amorphous materials [4,5]. Therefore, studies of amorphous wires have attracted substantial attention owing to several promising applications [3,6,7]. Aforementioned magnetic softness of amorphous wires is intrinsically related to the Giant Magneto-Impedance, GMI, effect, consisting of a substantial impedance, Z, modification under the applied magnetic field, H [7,8]. The GMI effect is commonly described in terms of the GMI ratio, $\Delta Z/Z$, defined as [7,8]:

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

where H_{max} – is the maximum applied DC magnetic field (usually below a few kA/m).

A change of the skin depth, δ, of a magnetically soft conductor under effect of magnetic field, *H,* is assumed as the origin of the GMI effect. The relationship δ and circumferential magnetic permeability, μ_Φ, of magnetic wire is given as [8]:

$$\delta = \frac{1}{\sqrt{\pi\sigma\mu_\Phi f}} \qquad (2)$$

being σ -the electrical conductivity.

High μ_Φ and substantial μ_Φ(H) dependence observed in amorphous magnetic wires are essentially important factors for achievement of high GMI effect [7-9].

The main advantage of GMI eff ect is its unusually high magnetic field sensitivity (impedance change by several hundred percent with a weak external magnetic field). Currently, the most common technologies used for the magnetic field sensors are Magnetoresistance, Hall-eff ect or Fluxgate techniques. Quite recently Giant magneto-impedance eff ect (GMI) technology has been developed and proposed to achieve extremely high magnetic field sensitivity for various magnetic sensors applications [8-10,12]. The latest generations of magnetic field sensors involving GMI effect show pT magnetic field sensitivity [12]. These advanced characteristics of GMI sensors make them suitable for the biomagnetic field detection in small musculature samples with spontaneous electric activity, using a GMI sensor with the pT sensitivity.

Typically, the highest GMI ratio values (about 300%) are reported for Co-rich magnetic wires with vanishing magnetostriction coefficients, λ_s [8-10]. After appropriate post-processing $\Delta Z/Z$ –values up to 650% were achieved in Co-rich glass-coated [8].

However, the observed $\Delta Z/Z$ –values are remain below the predicted $\Delta Z/Z$ –value of about 3000% [11].

There are several fabrication technique allowing to produce amorphous magnetic wires, such as "in-rotating-water" [4,12], melt extraction [13,14] or Taylor-Ulitovsky technique [15-17]. The common feature of all of them is that the fabrication processes involves rapid melt quenching. However, the glass-coated microwires, prepared using the Taylor-Ulitovsky (also known as quenching-and-drawing) method allows to produce amorphous microwires with the most extended diameters range (metallic nucleus diameters from 0.1 to 100 μm) [7, 15-17]. Briefly, the fabrication method consists of rapid solidification from the melt of the metallic alloys inside the glass capillary. This method covers the widest diameters range. Additionally, the presence of insulating flexible glass allows to extend the applications possibilities. However, the presence of the glass-coating also associated with the strong internal stresses. Therefore, post-processing of such microwires can be useful for the improvement of magnetic softness and GMI effect [7].

It is obvious that the sensors and devices characteristics are critically influenced by the $\Delta Z/Z$ –values. In a number of applications, such as contactless monitoring of carbon fibers composites with magnetic wire inclusions (typically used in aircraft industry), the $\Delta Z/Z$ –values of magnetic wires inclusions are of significant importance [18].The most common route to improve the soft magnetic properties of amorphous materials involves annealing at appropriate conditions [7]. In fact, improved magnetic softness of amorphous materials is attributed to glassy-like structure characterized by the absence of the defects (dislocations, twins, grain boundaries, etc.) limiting the soft magnetic properties of the crystalline materials [3,6,7]. Therefore, one of the main sources of magnetic anisotropy of amorphous is the magnetoelastic anisotropy, affected by the mechanical stresses (applied and internal) and the magnetostriction coefficient, λ_s [3,6,7]. Accordingly, the internal stresses relaxation upon annealing usually affects the magnetoelastic anisotropy and hence expect to improve magnetic softness of amorphous materials.

Thus, we present our last results on improvement of the soft magnetic properties and GMI effect of Co-rich glass-coated magnetic microwires.

2 Materials and Methods

We have studied the magnetic properties GMI effect of Co-rich ($Co_{72}Fe_4B_{13}Si_{11}$) glass-coated amorphous microwires with the metallic nucleus diameter, *d*, of about 40 μm and a total diameter, *D*, of 45 μm prepared by the modified Taylor-Ulitovsky method [7, 16,17]. The compositions of studied Co-rich microwires are selected considering vanishing magnetostriction coefficient, λ_s ($\lambda_s \approx -10^{-7}$) [19].

We used the fluxmetric method for measurement of the hysteresis loops. This method has been developed for characterization of soft magnetically soft thin microwires [20]. For better comparison of the magnetic properties of the studied microwires after different treatments, the hysteresis loops are represented as a dependence of the normalized magnetization M/M_o on the applied magnetic field, *H* (where M_o is magnetic moment of the samples at the maximum magnetic field amplitude H_o). The as-prepared sample presents rather good magnetic softness with coercivities, H_c, below 20 A/m and magnetic anisotropy fields, H_k, below 200 A/m (see Fig.1).

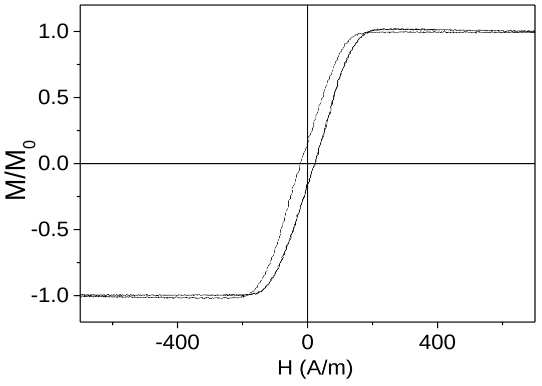

Fig. 1. Hysteresis loops of studied as-prepared

The GMI ratio, $\Delta Z/Z$, was defined using eq. (1) from the *Z(H)* dependence, evaluated using a vector network analyzer from the reflection coefficient S_{11}, as described elsewhere [21].

The amorphous structure of all studied samples has been confirmed by a broad halo observed in the X-ray spectra obtained using BRUKER (D8 Advance) X-ray diffractometer with CuK$_\alpha$ (λ=1.54 Å) radiation.

The λ_s of studied microwires has been evaluated by the so-called small angle magnetization rotation (SAMR) method using recently developed setup [19].

The microwires have been annealed in conventional furnace at temperatures, T_{ann}, up to and 350 °C for 60 min.

3 Experimental Results and Discussion

As observed from the *ΔZ/Z(H)* dependencies, measured at frequencies, *f* up to 600 MHz, (see Fig. 2), a substantial GMI ratio is observed even in an as-prepared microwire. A double-peak *ΔZ/Z(H)* dependencies are observed in studied sample for all *f* (see Fig. 2). At each *f*, a maximum GMI ratio, $\Delta Z/Z_{max}$, is observed at a certain magnetic field value, H_m. The highest GMI ratio, $\Delta Z/Z_{max}$, is observed at *f*=200-300 MHz, where $\Delta Z/Z_{max}$ ≈560 % (see Fig.2).

Such double-peak *ΔZ/Z(H)* dependencies were predicted [22] and obtained in magnetic wires with circumferential magnetic anisotropy [9]. The observed high $\Delta Z/Z_{max}$ -values and double-peak *ΔZ/Z(H)* dependencies correlate with an inclined bulk hysteresis loop with low H_c and H_k -values.

The evolution of the hysteresis loops of studied samples after the annealing at T_{ann} =275 ºC, 300 °C

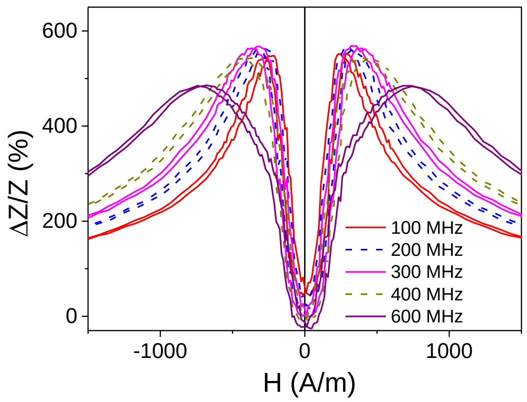

Fig.2. *ΔZ/Z(H)* dependencies measured at different frequencies in studied as- prepared sample.

and 350 °C (60 min) is provided in Fig.3. After annealing, a decrease in the H_k –value up to H_k≈75 A/m is observed, while H_c remains almost the same (H_c≈24 A/m).

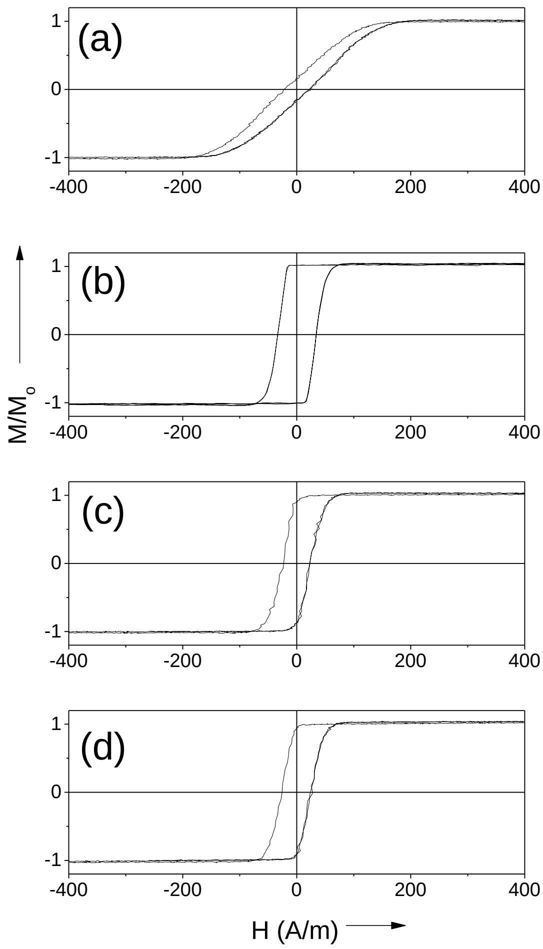

Fig. 3. Hysteresis loops of as-prepared (a) and annealed at T_{ann}= 275 ºC (b), T_{ann}= 300 ºC (c) and T_{ann}= 350 ºC (d) $Co_{72}Fe_4B_{13}Si_{11}$ sample.

The *ΔZ/Z(H)* dependencies measured in $Co_{72}Fe_4B_{13}Si_{11}$ microwire annealed at 275 ºC ≤ T_{ann} ≤350 °C are provided in Fig.4. Compared to the as-prepared $Co_{72}Fe_4B_{13}Si_{11}$ microwire, a substantial increase in $\Delta Z/Z_{max}$ –values is observed for all T_{ann}. The highest $\Delta Z/Z_{max}$ –values are observed for the $Co_{72}Fe_4B_{13}Si_{11}$ microwire annealed at T_{ann}=300 °C, where $\Delta Z/Z_{max}$ ≈ 720 % (see Fig.4b).

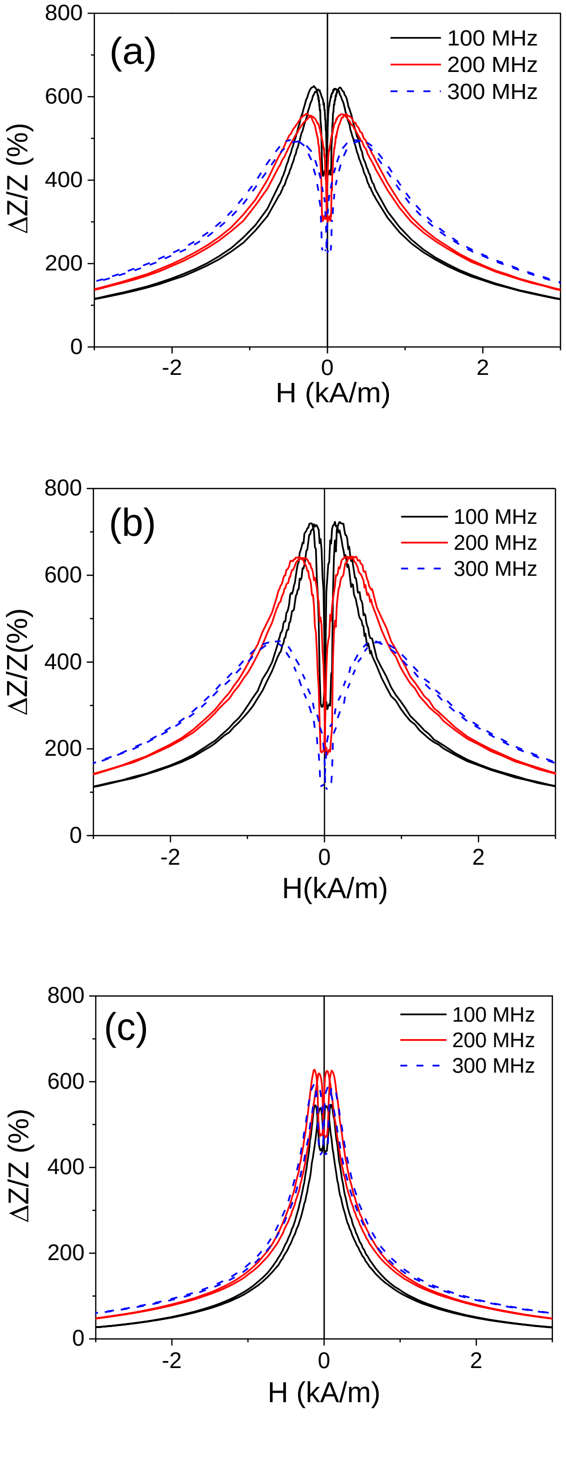

Fig. 4. *ΔZ/Z(H)* dependencies measured in

As in the as-prepared $Co_{72}Fe_4B_{13}Si_{11}$ sample, the *ΔZ/Z(H)* dependences of all annealed $Co_{72}Fe_4B_{13}Si_{11}$ microwires have a two-peak character. However, the difference is that the magnetic field H_m, at which such a maximum in the *ΔZ/Z(H)* dependences is observed, becomes lower. Thus, for f=100 MHz H_m decreases from 0.35kA/m (for as-prepared sample) to 0.15 kA/m (for all annealed $Co_{72}Fe_4B_{13}Si_{11}$ samples). The magnetic field at which a maximum is observed in the *ΔZ/Z(H)* dependencies, H_m, is commonly associated with the magnetic anisotropy field [8,22]. Therefore, observed change in *ΔZ/Z(H)* dependencies after annealing correlates with the evolution of the hysteresis loops upon annealing. While higher $\Delta Z/Z_{max}$ –values, observed in annealed $Co_{72}Fe_4B_{13}Si_{11}$ samples must be related with a decrease in magnetic anisotropy field after annealing.

Observed in Fig.3 modification of the hysteresis loops $Co_{72}Fe_4B_{13}Si_{11}$ microwires towards increase in the remanent magnetization, M_r/M_0 after annealing is similar to those previously reported for various Co-rich microwires [23]. Such change in hysteresis loops shape was explained in terms of the relationship between the λ_s-value, the internal stresses value and the structural relaxation [23-26]. In fact, the peculiarity of glass-coated microwires is related to the strong internal stresses induced during the preparation process involving the rapid melt quenching of the metallic alloy inside the glass tube [7,16,27]. Although such stresses have a complex character, theoretically predicted and experimentally (by glass-coating etching) demonstrated that the largest component of such internal stresses related to the difference in thermal expansion coefficients of the metallic alloy and glass is the axial one [7,16,27,28]. Therefore, negative λ_s-value together with axial character of internal stresses result in transverse character of magnetic anisotropy of Co-rich microwires with low and vanishing λ_s-value, as shown in Fig.3.

Our measurements demonstrate that indeed low λ_s-value (λ_s-≈-9x10^{-7}) are observed for as-prepared $Co_{72}Fe_4B_{13}Si_{11}$ microwires (see Table 1). Upon annealing a change in λ_s-value from low negative to low positive are observed. Such changes in λ_s-value can explain the modification in the hysteresis loops and in *ΔZ/Z(H)* dependencies. Observed modification in λ_s-value after annealing are similar to those recently reported in other Co-rich microwires with thinner metallic nucleus diameters (about 15-25 µm) [26]. However, in contrast to early studied thinner Co-rich amorphous microwires, in the case of studied thicker $Co_{72}Fe_4B_{13}Si_{11}$ microwires (d ≈40 µm) microwire a relatively low coercivity values (H_c≈20-25 A/m) are obtained (see Fig. 3).

Table 1. The magnetostriction coefficient of as-prepared and annealed $Co_{72}Fe_4B_{13}Si_{11}$ microwires.

Sample	λ_s (x10^{-7})
As-prepared	-9
Annealed at 275 °C	+7
Annealed at 300 °C	+11
Annealed at 350 °C	+13

As mentioned above and discussed elsewhere [8], the GMI effect is associated with the influence of the applied field, *H*, on the skin depth, *δ*, of an AC current flowing through the wire at frequency, *f*. Such *δ(H)* substantial dependence is intrinsically related to high value of the circumferential magnetic permeability, μ_ϕ, of amorphous magnetic wires and its substantial magnetic field dependence, as described in eq. (2). A double-peak *ΔZ/Z(H)* dependencies observed for all annealed $Co_{72}Fe_4B_{13}Si_{11}$ samples (see Figs. 4a-c) suggest the presence of the transverse magnetic anisotropy in the surface layer of the studied microwires at high enough *f*-values.

The origin of the weak transverse anisotropy in the surface of studied $Co_{72}Fe_4B_{13}Si_{11}$ microwires can be explained by the presence of the interface layer between the metallic nucleus and the glass-coating and hence different chemical composition in thin surface interface layer, as previously experimentally observed elsewhere [29].

It is worth mentioning that the improvement of the GMI ratio by annealing of $Co_{72}Fe_4B_{13}Si_{11}$ microwires in observed in quite extended frequency range. As illustrated by the Fig. 5, the highest $\Delta Z/Z_{max}$ –values in the $Co_{72}Fe_4B_{13}Si_{11}$ microwires annealed T_{ann}=300 °C is obtained at $f \approx 80$ MHz, when $\Delta Z/Z_{max} \approx 735\%$ is achieved [30]. Such value of the optimal frequency for the highest $\Delta Z/Z_{max}$ is lower than that typically observed for thinner (15µm≤d≤20µm) Co-rich microwires where the highest $\Delta Z/Z_{max}$ is typically reported at 100-200 MHz [7]. As discussed elsewhere, there is a relationship between the optimal frequency, *f*, and the diameter of the magnetic wire: a decrease in wire diameter is related with higher optimal frequency for the highest $\Delta Z/Z_{max}$ [31]. Thus for thicker magnetic wires (*d*=55 µm and 120 µm) the optimum frequency, *f*, is between 20 MHz and 1 MHz [32,33].

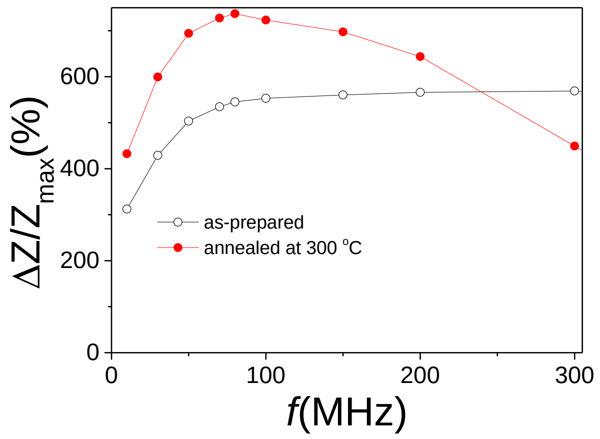

Fig.5. $\Delta Z/Z_{max}(f)$ dependences evaluated for as-prepared and annealed at 300 °C $Co_{72}Fe_4B_{13}Si_{11}$ microwires.

Observed high $\Delta Z/Z_{max}$ –values and remarkable improvement of GMI effect after annealing open possibilities for design of more sensitive and efficient magnetic field sensors and devices for applications in various fields, including civil engineering, aircraft or automobile industries or medicine.

4 Conclusion

The main conclusion that can be summarized from obtained experimental results are that the appropriate annealing of Co-rich microwires can substantially improve the GMI ratio. Modification of hysteresis loops and ΔZ/Z(H) dependencies after annealing can be attributed to the change in the sign and values of the magnetostriction coefficient and the internal stresses relaxation.

Acknowledgement:

The authors thank the technical and human support provided by SGIker of UPV/EHU (Medidas Magneticas Gipuzkoa) and European funding (ERDF and ESF). J.O. thanks the Ministry of Higher Education, Science and Technology (MESCYT) of the Dominican Republic.

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Contribution of Individual Authors to the Creation of a Scientific Article (Ghostwriting Policy)

The authors equally contributed in the present research, at all stages from the formulation of the problem to the final findings and solution.

Sources of Funding for Research Presented in a Scientific Article or Scientific Article Itself

This work was supported by EU under “INFINITE” (HORIZON-CL5-2021-D5-01-06) project and “Harmony” (HORIZON-CL4-2023-RESILIENCE-01) projects, by the Spanish MICIN, under PID2022-141373NB-I00 project, by the Government of the Basque Country under Elkartek (MOSINCO and ATLANTIS) projects and under the scheme of “Ayuda a Grupos Consolidados” (ref. IT1670-22).

Conflict of Interest

The authors have no conflicts of interest to declare that are relevant to the content of this article.

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