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DEVELOPMENT OF MAGNETICALLY SOFT CO-RICH AMORPHOUS MICROWIRES WITH HIGH GIANT MAGNETOIMPEDANCE EFFECT

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ABSTRACT

Amorphous magnetic wires can exhibit unique magnetic properties, such as magnetic bistability [1] and/or Giant Magneto-Impedance, GMI, effect associated with excellent magnetic softness [2]. Additionally, amorphous materials are also characterized by superior mechanical and corrosion properties [3]. Such combination of physical properties makes the amorphous wires attractive for a variety of industrial applications, such as magnetic and magnetoelastic sensors or tunable metamaterials [2,4]. One of the latest trends in the development of amorphous magnetic wires is to reduce their size and expand their functionality through protective coatings. Among the most effective solutions for the production of thin amorphous magnetic wires is the so-called Taylor-Ulitovsky method, allowing the preparation of microwires with rather extended diameters range from 100 nm to 100 µm coated with an insulating, flexible and biocompatible glass coating [4]. The performance of GMI effect-based sensors and devices can be significantly improved by using materials with higher GMI effect. Typically, the highest GMI ratio of about 200-300% is observed in Co-rich magnetic wires with vanishing magnetostriction coefficients, λ_s [2]. While, in carefully processed magnetic microwires, GMI ratios of up to 650% have been obtained [4]. However, the reported GMI ratios are still below the theoretically predicted 3000% [2]

Consequently, in this paper we provide our latest attempt on optimization of the magnetic softness and GMI effect in Co-rich glass-coated magnetic microwires. We studied the effect of annealing on the hysteresis loops and the GMI ratio of Co-rich microwires. Surprisingly, after conventional annealing, in most of Co-rich microwires, magnetic hardening and transformation of a linear hysteresis loop into a rectangular one with a higher coercive force are observed. However, stress-annealing allows preventing magnetic hardening and remarkably improve GMI ratio. Properly stress-annealed samples present almost unhysteretic loops with coercivity about 2 A/m and magnetic anisotropy field about 35A/m. A remarkable GMI ratio improvement up to 735% is observed after annealing of Co-rich microwires at appropriate conditions. Observed magnetic softening and GMI ratio improvement have been discussed considering the internal stresses

relaxation, induced magnetic anisotropy and a change in the magnetostriction coefficient sign and values with increasing of annealing temperature.

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 critically 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 main advantages of amorphous magnetic wires are excellent magnetic softness that can be obtained directly after preparation. 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 [87,]:

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

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) dependencies of amorphous magnetic wires are the essentially relevant factors for achievements of high GMI effect [7-9].

Typically, the highest $\Delta Z/Z$ –values about 200-300% are observed in Co-rich magnetic wires with nearly-zero 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, reported $\Delta Z/Z$ –values are still below of the theoretically predicted $\Delta Z/Z \approx 3000\%$ [11].

There are several families of amorphous magnetic wires. The common feature of all of them is that the fabrication processes involve rapid melt quenching. The studies of the GMI effect are performed mostly in the following types of magnetic wires:

i) Conventional amorphous wires prepared using “in-rotating water” method (with diameters between 100 and 120 µm) [4,12]. The relatively thick diameters of such amorphous wires do not allow the production of low-dimensional sensors and devices. The cold-drawing can be used for the diameter reduction [12]. However, such cold-drawn can damage the surface and induce complex internal stresses.

ii) Melt extracted amorphous microwires (with diameters between 30 and 60 µm) have been developed at the beginning of 90-s[13,14]. Such microwires are prepared by extraction of the microwire from the melt using a rapidly rotating sharpened wheel. Accordingly, the cross section of such microwires is not perfectly round. This may affect the magnetic properties and hence the GMI effect.

iii) Glass-coated microwires (metallic nucleus diameters from 0.1 to 100 µm) can be prepared using so-called modified Taylor-Ulitovsky (also known as quenching-and-drawing method), actually known since 60-s and intensively studied since 90-s [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 clear that the features of sensors and devices utilizing the GMI effect is critically affected by the GMI ratio value. In several applications, such as wireless monitoring of composites for aircraft industry made from carbon fibers with magnetic wire inclusions, the value of the GMI ratio of the ferromagnetic glass-coated microwires inclusions is essentially relevant [18].The most common way to improve the magnetic softness of amorphous materials is appropriate post-processing (involving annealing) [7].

Consequently, in this paper we provide our latest attempt on optimization of the magnetic softness and GMI effect in Co-rich glass-coated magnetic microwires.

EXPERIMENTAL METHODS AND SAMPLES

We have studied the magnetic properties GMI effect of Co-rich glass-coated amorphous microwires with two different compositions and metallic nucleus diameters, *d*, ranging from 22 up to 40 µm and a total diameter, *D*, up to 45 µm: $Co_{64.6}Fe_5B_{16}Si_{11}Cr_{3.4}$ and $Co_{72}Fe_4B_{13}Si_{11}$ 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].

Axial hysteresis loops were measured using the fluxmetric method, developed for studies of soft magnetic microwires with reduced diameters [20]. The hysteresis loops represented as the normalized magnetization M/M_o versus applied magnetic field, H (being M_o -the magnetic moment of the samples at maximum amplitude H_o of magnetic field) allows better comparison of magnetic properties of studied microwires with different chemical compositions and diameters. All prepared microwires present rather soft magnetic properties with coercivities, H_c, below 20 A/m and magnetic anisotropy fields, H_k, below 200 A/m (see Fig.1).

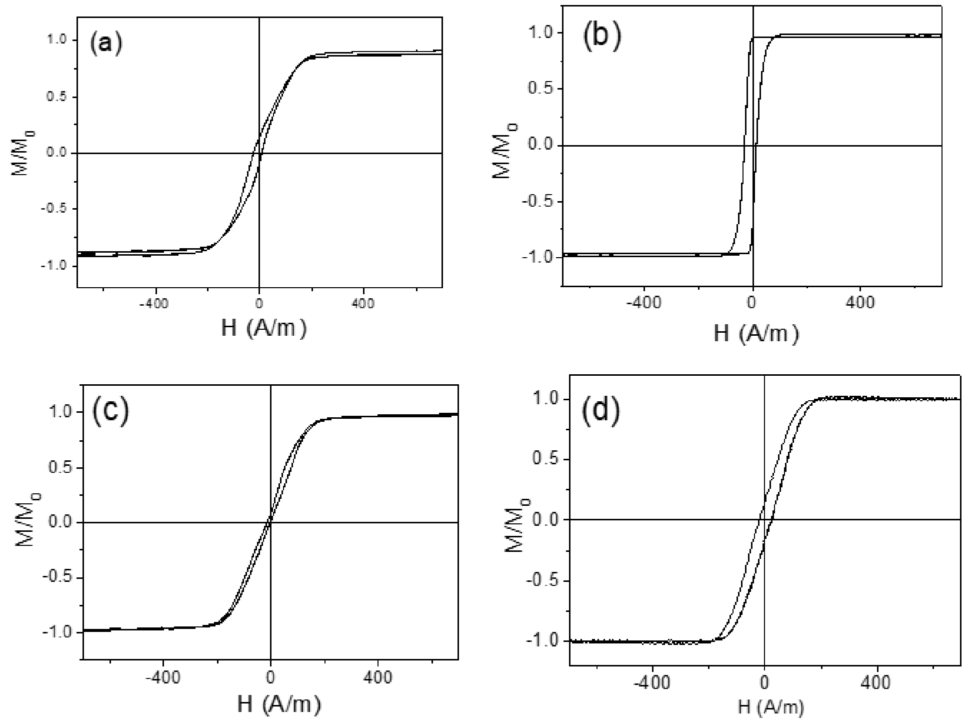


Figure 1: Hysteresis loops of Co64.6Fe5.0B16.0Si11.0Cr3.4 microwires (a,b,c) with d≈22 µm, D≈24µm; d=38 µm, D=43.5 µm and d≈22.8 µm, D≈23.2µm respectively and Co72Fe4B13Si11 microwires with d=40µm, D=45 µm (d).

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

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

A recently developed setup designed to evaluate the λ_s of magnetic microwires using the so-called small angle magnetization rotation (SAMR) method was used for λ_s measurements [19].

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

EXPERIMENTAL RESULTS AND DISCUSSION

A substantial GMI ratio is observed even in all as-prepared samples, as can be seen from the ΔZ/Z(H) dependencies, measured at $100 \leq f \leq 400$ MHz, (see Fig. 2). The highest GMI ratio, ΔZ/Zmax, is observed at f=200 MHz, in Co72Fe4B13Si11 microwire where ΔZ/Zmax ≈400 % (see Fig.2d).

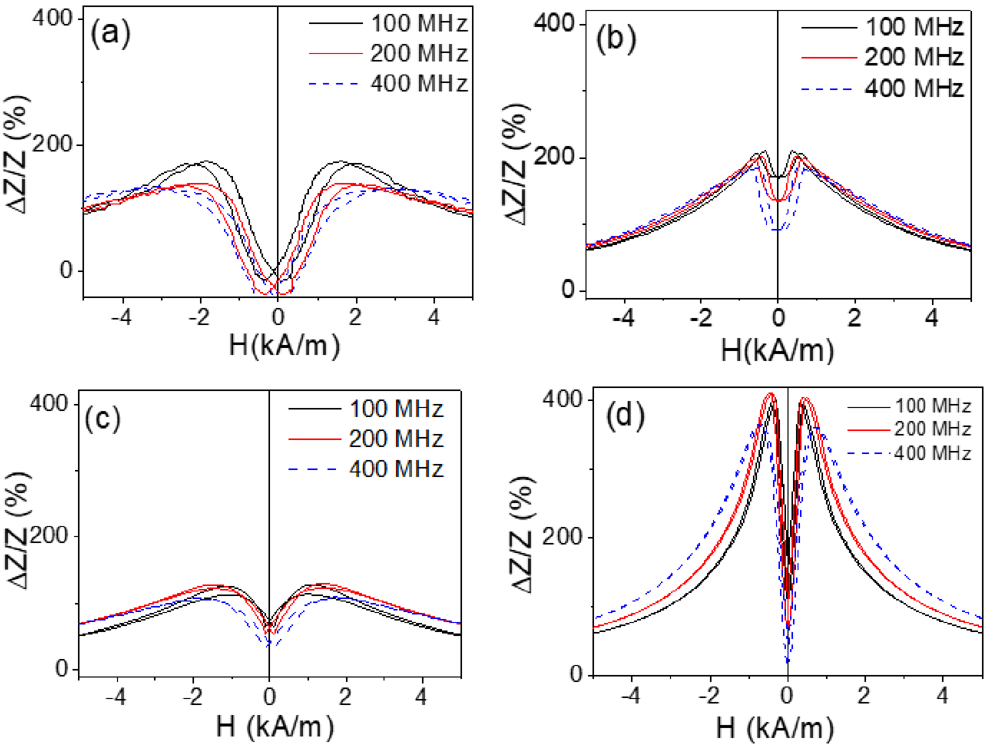


Figure 2: ΔZ/Z(H) dependencies measured in Co64.6Fe5.0B16.0Si11.0Cr3.4 microwires with d≈22 µm (a), d=38 µm (b) d≈23.2 µm (c) and in Co72Fe4B13Si11 with d≈40 µm microwires (d).

A double-peak ΔZ/Z(H) dependencies are observed in all studied samples for all measured frequencies (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 ΔZ/Zmax -values and double-peak ΔZ/Z(H) dependencies correlate with an inclined bulk hysteresis loop with low Hc and Hk -values.

To evaluate the effect of heat treatment on the magnetic properties and the GMI effect, we chose the Co72Fe4B13Si11 microwire with the highest ΔZ/Zmax –values.

The evolution of the hysteresis loops of Co72Fe4B13Si11 microwire after the annealing at Tann =275 ºC, 300 °C and 350 °C (60 min) is provided in Fig.3. The main change of the hysteresis loops consists of a decrease in Hk –value up to Hk≈75 A/m, while the Hc remains almost unchanged (Hc≈24 A/m).

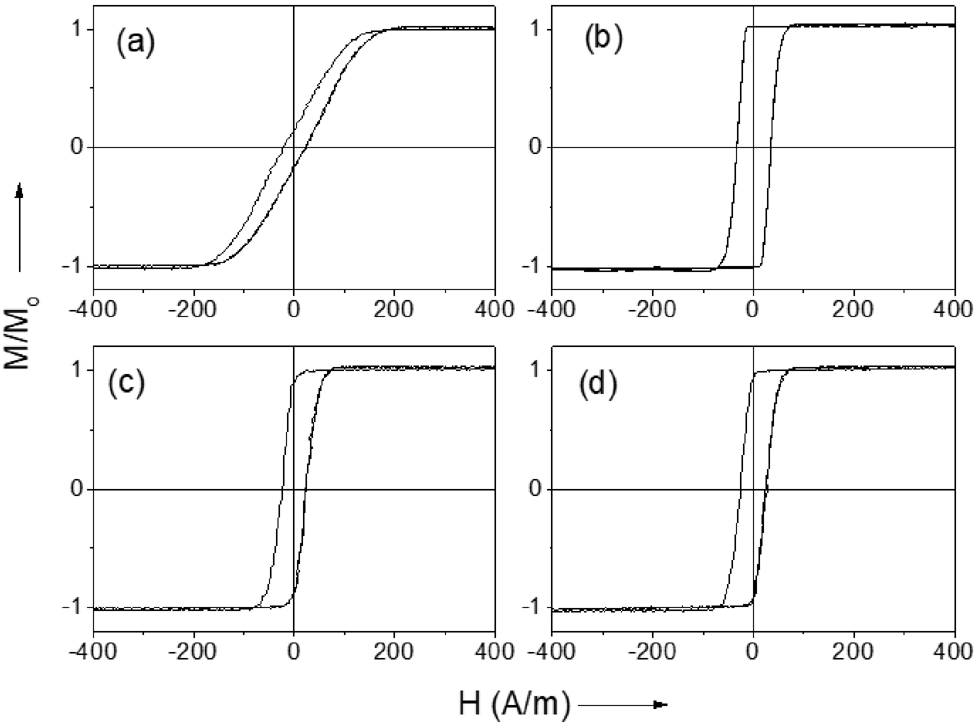


Figure 3: Hysteresis loops of as-prepared (a) and annealed at Tann= 275 oC (b), Tann= 300 oC (c) and Tann= 350 oC (d) Co72Fe4B13Si11 sample.

The ΔZ/Z(H) dependencies measured in Co72Fe4B13Si11 microwire annealed at 275 oC ≤ Tann ≤350 °C are provided in Fig.4. Compared to the as-prepared Co72Fe4B13Si11 microwire, a substantial increase in ΔZ/Zmax –values is observed for all Tann. The highest ΔZ/Zmax –values are observed for the Co72Fe4B13Si11 microwire annealed at Tann=300 °C, where ΔZ/Zmax ≈ 720 % (see Fig.4b).

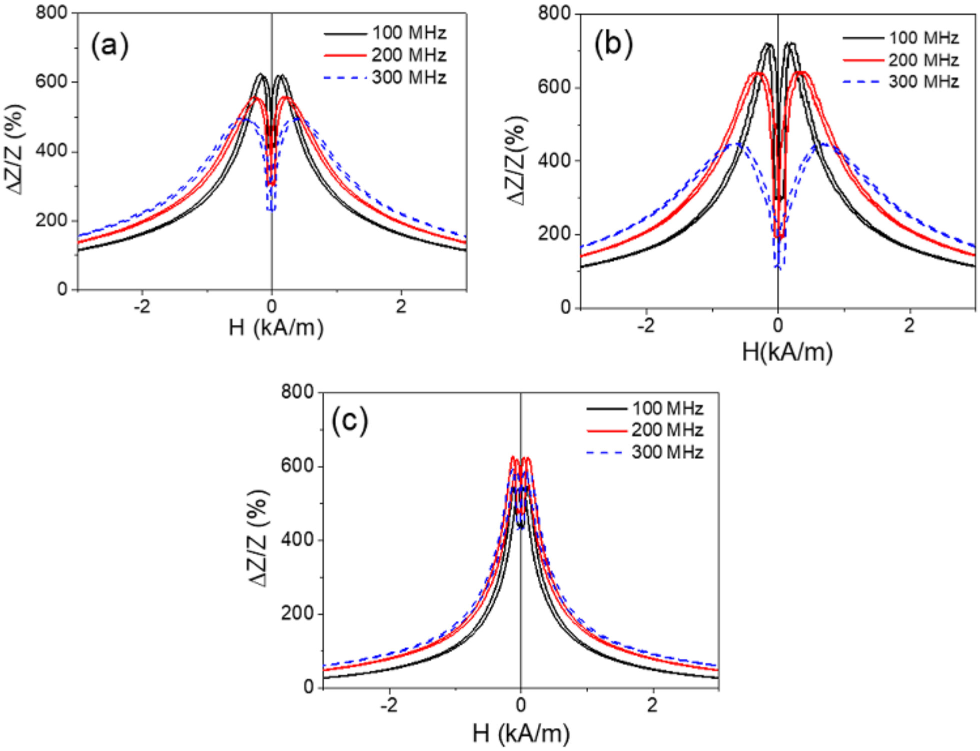


Figure 4: ΔZ/Z(H) dependencies measured in annealed at 350 oC (a) and 275 oC (b) Co72Fe4B13Si11 microwires at frequencies 10-300 MHz.

As in the as-prepared Co72Fe4B13Si11 sample, the ΔZ/Z(H) dependences of all annealed Co72Fe4B13Si11 microwires have a two-peak character. However, the difference is that the magnetic field Hm, at which such a maximum in the ΔZ/Z(H) dependences is observed, becomes lower. Thus, for f=100 MHz Hm decreases from 0.35kA/m (for as-prepared sample) to 0.15 kA/m (for all annealed Co72Fe4B13Si11 samples). The magnetic field of maximum, Hm, in ΔZ/Z(H) dependencies is commonly associated with the magnetic anisotropy field [8,22]. Therefore, observed change in in ΔZ/Z(H) dependencies after annealing correlates with the evolution of the hysteresis loops upon annealing. While higher ΔZ /Zmax –values, observed in annealed Co72Fe4B13Si11 samples must be related with a decrease in magnetic anisotropy field after annealing.

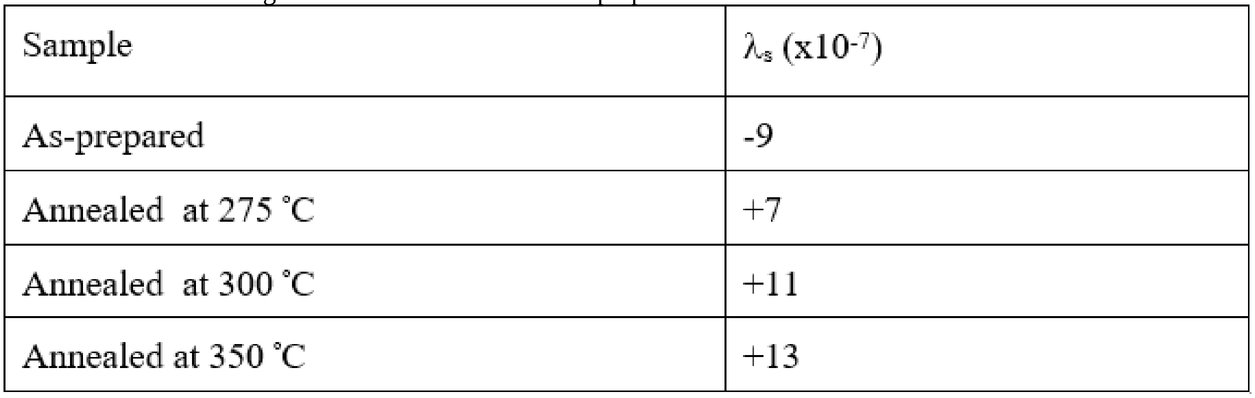
Observed in Fig.3 modification of the hysteresis loops Co72Fe4B13Si11 microwires towards increase in the remanent magnetization, Mr/M0 after annealing is similar to those previously reported for various Co-rich microwires [23]. Such change in hysteresis loops shape was explained in terms of the relationship between the λs-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 λs-value together with axial character of internal stresses result in transverse character of magnetic anisotropy of Co-rich microwires with low and vanishing λs-value, as shown in Fig.3.

Our measurements demonstrate that indeed low λs-value (λs-≈-9x10-7) are observed for as-prepared Co72Fe4B13Si11 microwires (see Table 1). Upon annealing a change in λs-value from low negative to low positive are observed. Such changes in λs-value can explain the modification in the hysteresis loops and in ΔZ/Z(H) dependencies. Observed modification in λs-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 Co72Fe4B13Si11 microwires (d ≈40 μm) microwire a relatively low coercivity values (Hc≈20-25 A/m) are obtained (see Fig. 3).

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, μϕ, 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 Co72Fe4B13Si11 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 Co72Fe4B13Si11 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].

Table 1: The magnetostriction coefficient of as-prepared and annealed Co72Fe4B13Si11 microwires.



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

It is worth mentioning that the improvement of the GMI ratio by annealing of Co72Fe4B13Si11 microwires in observed in quite extended frequency range. As illustrated by the Fig. 5, the highest ΔZ/Zmax –values in the Co72Fe4B13Si11 microwires annealed Tann=300 °C is obtained at f ≈ 80 MHz, when ΔZ/Zmax ≈735% is achieved [30].

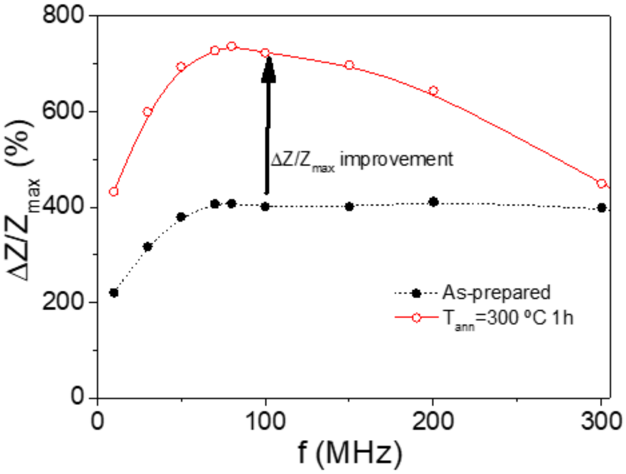


Figure 5: ΔZ/Zmax(f) dependences evaluated for as-prepared and annealed at 300 oC Co72Fe4B13Si11 microwires.

Such value of the optimal frequency for the highest ΔZ/Zmax is lower than that typically observed for thinner (15µm≤d≤ 20µm) Co-rich microwires where the highest ΔZ/Zmax 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 ΔZ/Zmax[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].

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.

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