

Magnetic Properties and Applications of Glass-coated Ferromagnetic Microwires

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Abstract—The impact of post-processing on soft magnetic properties and the Giant Magneto-Impedance (GMI) effect of Fe- and Co-based glass-coated microwires is evaluated. A remarkable improvement of magnetic softness and GMI effect is observed in Fe-rich glass-coated microwires subjected to stress annealing. Frequency dependence of GMI ratio of stress-annealed Fe-rich microwires has been discussed considering frequency dependence of the skin penetration depth, δ , as well as magnetic anisotropy distribution within the metallic nucleus. Annealed and stress-annealed Co-rich microwires present rectangular hysteresis loop and single and fast domain wall propagation. However, Co-based stress-annealed microwires present high magnetoimpedance ratio. Observed stress-induced anisotropy and related changes of magnetic properties are discussed considering internal stresses relaxation and “back-stresses”.

Keywords- Giant Magneto-Impedance effect; magnetic microwires; magnetic softness; domain wall propagation.

I. INTRODUCTION

Development of magnetic sensors is focused on the miniaturization of their size, improvement of their features and on finding of new materials. Among new magnetic materials, a family of thin wire with reduced dimensions recently gained considerable attention [1]. Glass-coated magnetic microwires prepared using the Taylor-Ulitovsky technique with thin metallic nucleus (typically with diameters 0.5 to 50 μm) covered by flexible, insulating and biocompatible glass are therefore quite interesting for sensor applications [2]. This technique allows preparation of the thinnest rapidly quenched wires with amorphous or crystalline structure of metallic nucleus. Good magnetic properties can be observed either in crystalline or in amorphous magnetic wires, but amorphous magnetic wires present several advantages, such as superior mechanical properties, the absence of microstructure defects (grain boundaries, crystalline texture, dislocations, point defects, etc.) [2] and hence precise post-processing is not required. Particularly, amorphous microwires can present Giant Magneto-Impedance (GMI) or magnetic bistability. In the

case of glass-coated microwires the magnetoelastic anisotropy contribution becomes relevant since the preparation process involves not only the rapid quenching itself, but also simultaneous solidification of the metallic nucleus surrounded by non-magnetic glass-coating with rather different thermal expansion coefficients [2].

The purpose of this paper is to present the most recent results on tailoring of soft magnetic properties and GMI effect in glass-coated microwires paying special attention to achievement of high GMI effect and on optimization of domain wall dynamics. We also provide several examples of sensor applications of such glass-coated microwires. The rest of the paper is structured as follows. In Section II, we present the experimental methods, while in Section III we describe the results on the dependencies of hysteresis loops, domain wall dynamics and GMI effect of the studied microwires on annealing conditions.

II. EXPERIMENTAL DETAILS

We studied glass-coated amorphous microwires prepared by the Taylor-Ulitovsky method. The manufacturing technique is described elsewhere [3][4]. It involves melt quenching of the metallic alloy inside a glass capillary and winding of the solidified glass-coated microwires onto a rotating bobbin.

We measured the hysteresis loops, dynamics of domain wall propagation and GMI effect. Hysteresis loops were measured by the fluxmetric method using low frequency (100 Hz) AC magnetic field, as described elsewhere [5].

Sample impedance, Z , was determined from the reflection coefficient, S_{11} , using a vector network analyzer, as described in detail elsewhere [6]. From Z , measured at different magnetic fields, H , we evaluated the GMI ratio, $\Delta Z/Z$, as [2][7]:

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

where H_{max} – is the maximum applied DC magnetic field.

In the microwires with rectangular hysteresis loop, we studied the Domain Wall (DW) dynamics using modified

Sixtus-Tonks previously described elsewhere [8][9]. The main difference of used method with respect to classical Sixtus-Tonks [10] technique is that to ensure a single DW propagation we leave one end of the studied sample outside the magnetization coil. Additionally, we employed three pick-up coils in order to avoid the multiple DW propagation [8][9]. The DW velocity, v , can be estimated as [8][9]:

$$v = \frac{l}{\Delta t} \quad (2)$$

where l is the distance between pick-up coils and Δt is the time difference between the ElectroMotive Force (EMF) peaks originated by moving DW in the pick-up coils [8][9].

III. EXPERIMENTAL RESULTS AND DISCUSSION

As already reported previously [2][4], the character of the hysteresis loops is linked to the sign and value of the magnetostriction coefficient, λ_s . Therefore, studied Co-rich and Fe-rich microwires present rather different magnetic properties and hence GMI effect: Fe-rich microwires with $\lambda_s > 0$ present perfectly rectangular hysteresis loops exhibiting magnetic bistability and coercivity, H_c , of the order of 70 A/m (see Figure 1a). In contrast, Co-rich microwires ($\lambda_s \approx 0$) present

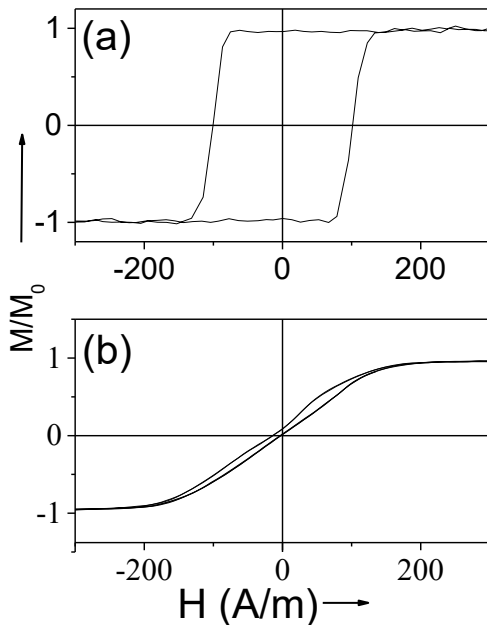


Figure 1. Hysteresis loops of amorphous magnetic microwires $\text{Fe}_{75}\text{B}_9\text{Si}_{12}\text{C}_4$ ($\lambda_s > 0$) (a), and $\text{Co}_{69.2}\text{Fe}_{3.6}\text{Ni}_1\text{B}_{12.5}\text{Si}_{11}\text{Mo}_{1.5}\text{C}_{1.2}$ ($\lambda_s \approx 0$) (b) microwires.

linear hysteresis loops with an order of magnitude lower H_c (see Figure 1b).

As experimentally shown in dozens of previous publications [2][8][9], the remagnetization of Fe-rich microwires with rectangular hysteresis loops runs by fast domain wall, DW, propagation.

The magnetic properties of amorphous materials are mostly determined by the magnetoelastic anisotropy. The common way to reduce the magnetoelastic anisotropy is annealing. For maintaining the amorphous structure of the samples the annealing temperatures, T_{ann} , must be selected below the crystallization temperature. Usually the crystallization of amorphous Fe- and Co-rich alloys is observed for $T_{ann} \geq 500$ °C [11]. The crystallization usually deteriorates mechanical properties of amorphous materials [11][12]. Therefore, we tried to anneal our samples at $T_{ann} < 500$ °C

As shown in Figure 2, it is evident that conventional furnace annealing (without stress) does not modify the hysteresis loop character of Fe-rich microwires: both as-prepared and annealed $\text{Fe}_{75}\text{B}_9\text{Si}_{12}\text{C}_4$ samples present similar

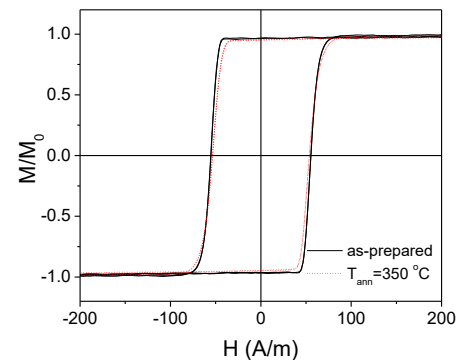


Figure 2. Hysteresis loops of as-prepared and annealed at $T_{ann} = 350$ °C $\text{Fe}_{75}\text{B}_9\text{Si}_{12}\text{C}_4$ microwire.

character of hysteresis loops. Only a slight decrease in H_c is observed after annealing (see Figure 2).

Roughly linear $v(H)$ dependencies are observed in studied Fe-rich microwires in the magnetic field region $H \geq H_c$ (see Figure 3a)

Linear $v(H)$ dependencies have been discussed in terms of the viscous DW motion [8][9]. The DW propagates with a velocity, v , given as:

$$v = S(H - H_0) \quad (3)$$

where H_0 is the critical propagation field and S is the DW mobility given by [8][9]

$$S = 2\mu_0 M_s / \beta \quad (4)$$

where μ_0 is magnetic permeability of vacuum, M_s -saturation magnetization and β is the viscous damping coefficient.

A substantial DW dynamics improvement in Fe-rich microwires after annealing is achieved (see Figure 3).

The origin of such improvement of DW dynamics, reflected by increase in v and S values is discussed elsewhere considering the dependence of DW dynamics on magnetoelastic anisotropy, K_{me} , given by [8][9][13]:

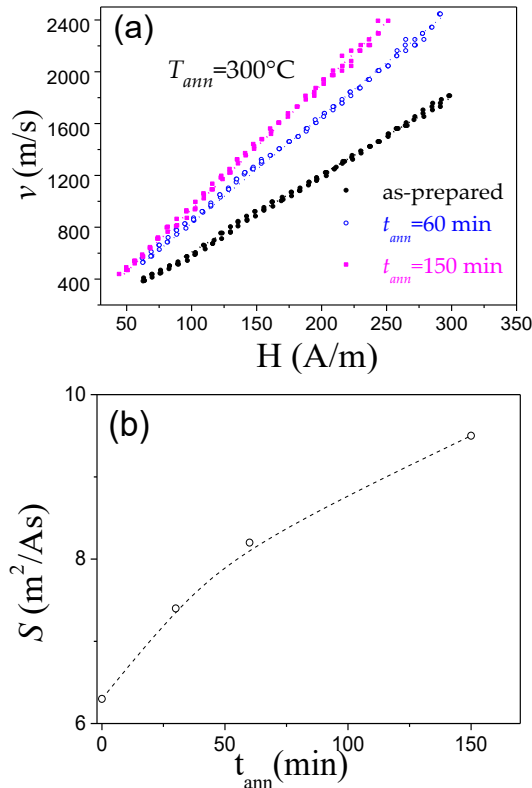


Figure 3. $v(H)$ dependencies, measured in as-prepared and annealed for different annealing time, t_{ann} , $Fe_{74}B_{13}Si_{11}C_2$ microwires (a), and $S(t_{ann})$ for the same microwires annealed at 300 °C (b).

$$K_{me} = 3/2 \lambda_s \sigma, \tag{5}$$

being σ —is the stresses value. In the absence of applied stresses, the internal stresses, σ_i , magnitude (and, hence, the degree of the σ_i relaxation) are essentially relevant for the DW dynamics improvement.

The magnetic relaxation damping, β_r , is considered elsewhere as the main factor affecting the DW dynamics at least in amorphous microwires [8]. The β_r is related to a delayed rotation of electron spins and inversely proportional to the domain wall width and given as [7][8]:

$$\beta_r \approx 2M_s \pi^{-1} (K_{me}/A)^{1/2} \tag{6}$$

where A is the exchange stiffness constant.

Consequently, K_{me} can affect S magnitude. Accordingly, the observed change in S -values can be qualitatively explained considering σ_i relaxation [7][8].

On the other hand, upon annealing the hysteresis loop of Co-rich microwires becomes rectangular presenting considerable magnetic hardening (see Figure 4). Such unexpected behavior was explained considering two different

phenomena. First of all, circular magnetic anisotropy and the remagnetization process by magnetization rotation of Co-rich microwires is associated to low negative λ_s -values and axial character of the internal stresses induced mostly by the difference in thermal expansion coefficients of metallic alloys and glass coating. Accordingly, the relaxation of internal stresses allows the contribution of magnetoelastic anisotropy to be reduced, while the contribution of shape anisotropy increases, facilitating axial magnetic anisotropy. Additionally, a change in the magnetostriction coefficient upon annealing is experimentally observed in several Co-rich microwires [10].

Accordingly, the hysteresis loops of annealed Co-rich microwires becomes similar to as-prepared Fe-rich

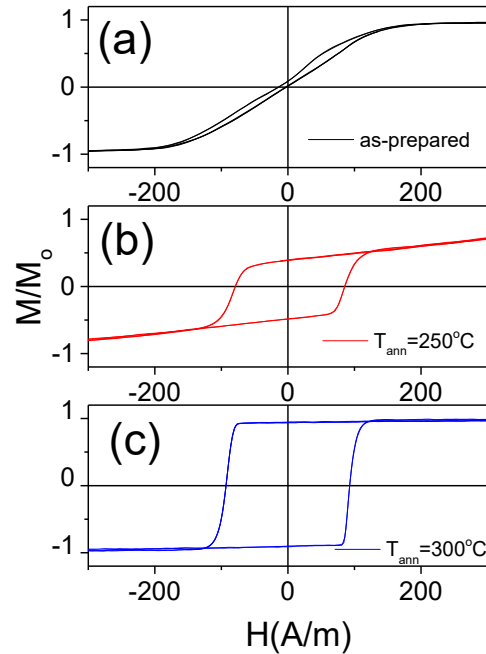


Figure 4. Hysteresis loop of as-prepared (a) and annealed at $T_{ann}=250^\circ C$ (b) and $300^\circ C$ (c) $Co_{69.2}Fe_{3.6}Ni_1B_{12.5}Si_{11}Mo_{0.15}C_{1.2}$ microwires.

microwires (see Figure 4). Such character of hysteresis loops is usually associated with the remagnetization process through the DW propagation.

Consequently, we measured the DW propagation in the Co-rich microwires with magnetic bistability induced by annealing. Obtained $v(H)$ dependence are presented in Figure 5a. Observed in studied Co-rich microwire v -values (above 2.5 km/s) are even higher than in Fe-rich microwires (see Figure 3). This difference can be attributed to lower λ_s -values of Co-rich alloy.

Surprisingly, a more pronounced GMI effect is observed in annealed Co-rich microwires with higher H_c and rectangular hysteresis loops (see Figure 5b): at a fixed frequency f , an increase in $\Delta Z/Z$ from approximately 100% to 140% is observed (see Figure 5b). Another important feature of the observed change in the $\Delta Z/Z(H)$ dependence after annealing is its modification from a double-peak to a single-

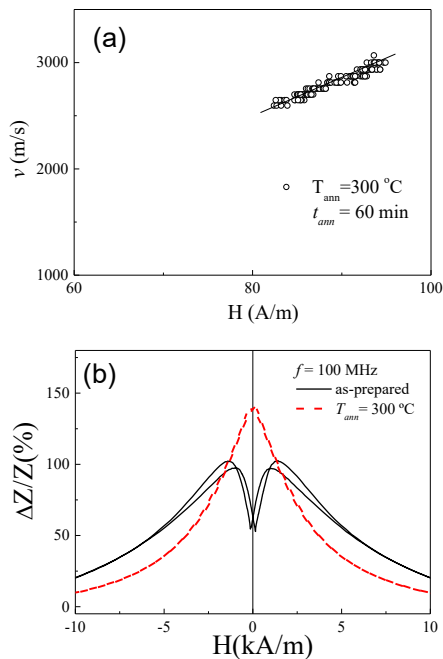


Figure 5. $v(H)$ dependence in annealed $\text{Co}_{69.2}\text{Fe}_{3.6}\text{Ni}_{1.1}\text{B}_{12.5}\text{Si}_{11}\text{Mo}_{1.5}\text{C}_{1.2}$ microwire (a) and effect of annealing on $\Delta Z/Z(H)$ dependences of $\text{Co}_{69.2}\text{Fe}_{3.6}\text{Ni}_{1.1}\text{B}_{12.5}\text{Si}_{11}\text{Mo}_{1.5}\text{C}_{1.2}$ microwires.

peak (see Figure 5b). It is worth noting that a double-peak $\Delta Z/Z(H)$ dependence is predicted for wires with transverse magnetic anisotropy, whereas a single-peak $\Delta Z/Z(H)$ dependence is typical for magnetic wires with axial magnetic anisotropy [14]. Accordingly, the observed change in the nature of the $\Delta Z/Z(H)$ dependence correlates fairly well with the observed change in the hysteresis loops after annealing.

Consequently, annealing of Co-rich microwires allows achievement of unique combination of magnetic properties. Annealed Co-rich microwires can simultaneously present single and fast Domain Wall (DW) propagation and high GMI effects.

Such combinations of magnetic properties can be suitable for various applications, such as magnetic sensors, smart composites with magnetic wire inclusions or magnetic memory devices [15]-[22]. Additionally, annealing can provide better time and temperature stability of properties of magnetic microwires.

For the results presented above, the annealing time was always 1 h, as is commonly used in most published works devoted to the effect of annealing on the magnetic properties of amorphous materials [1][5]. Such annealing time selection allows better comparison of obtained results with existing knowledge. However, annealing time is also another relevant parameter. Thus, it has been shown previously [23] that isothermal annealing with annealing time as a parameter allows for finer tuning of the hysteresis loops of Co-rich microwires. On the other hand, annealing at elevated

temperatures and/or for extended times can cause crystallization of amorphous materials, resulting in a significant deterioration of their magnetic and mechanical properties [12]. Therefore, annealing conditions should be chosen with caution.

IV. CONCLUSION AND FUTURE WORK

We demonstrated that magnetic properties, GMI effect and domain wall dynamics are substantially affected by annealing.

Obtained experimental results yield new and important insights suitable for development of material with unique combination of magnetic properties suitable for high performance magnetic sensors and devices.

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REFERENCES

- [1] G. Herzer, *Amorphous and Nanocrystalline Materials*, in: *Encyclopedia of Materials: Science and Technology*, pp. 149–157, Elsevier Science Ltd. (2001) ISBN: 0-08-0431526.
- [2] V. Zhukova et al., “Development of Magnetically Soft Amorphous Microwires for Technological Applications”, *Chemosensors*, vol. 10, p. 26, 2022.
- [3] H. Chiriac, S. Corodeanu, M. Lostun, G. Ababei, and T.-A. Óvári, “Rapidly solidified amorphous nanowires”, *J. Appl. Phys.*, vol. 107, 09A301, 2010.
- [4] T. Goto, M. Nagano, and N. Wehara, “Mechanical properties of amorphous Fe80P16C3B1 filament produced by glass-coated melt spinning”, *Trans. JIM*, vol. 18, pp. 759-764, 1997.
- [5] L. Gonzalez-Legarreta et al., “Optimization of magnetic properties and GMI effect of Thin Co-rich Microwires for GMI Microsensors,” *Sensors*, vol. 20, p.1558, 2020.
- [6] A. Zhukov et al., “Routes for Optimization of Giant Magnetoimpedance Effect in Magnetic Microwires”, *IEEE Instrumentation & Measurement Magazine*, vol. 23, no. 1, pp. 56-63, 2020, doi: 10.1109/MIM.2020.8979525.
- [7] L. V. Panina and K. Mohri, “Magneto-impedance effect in amorphous wires”, *Appl Phys Lett*. vol. 65, pp. 1189-1191, 1994.
- [8] V. Zhukova, J. M. Blanco, M. Ipatov, and A. Zhukov, Magnetoelastic contribution in domain wall dynamics of amorphous microwires, *Physica B* vol. 407, 1450-1454, 2012.
- [9] V. Zhukova et al., “Review of Domain Wall Dynamics Engineering in Magnetic Microwires”, *Nanomaterials* vol. 10, p.2407, 2020, doi: 10.3390/nano10122407.
- [10] K. J. Sixtus and L. Tonks, “Propagation of large Barkhausen discontinuities II”, *Phys. Rev.*, vol. 42, pp. 419-435, 1932.
- [11] L. Gonzalez-Legarreta et al., “Route of magnetoimpedance and domain walls dynamics optimization in Co-based microwires”, *J. Alloys Compound*. vol. 830, 154576, 2020, doi: <https://doi.org/10.1016/j.jallcom.2020.154576>.

- [12] V. Zhukova et al., "Correlation between magnetic and mechanical properties of devitrified glass-coated Fe_{71.8}Cu₁Nb_{3.1}Si₁₅B_{9.1} microwires", *J. Magn. Magn. Mater.* vol. 249, P1-II, 79-84, 2002.
- [13] A. Zhukov, J. M. Blanco, M. Ipatov, and V. Zhukova, "Fast magnetization switching in thin wires: Magnetoelastic and defects contributions". *Sens. Lett.* vol. 11 (1), pp. 170-176, 2013, doi: 10.1166/sl.2013.2771.
- [14] N. A. Usov, A. S. Antonov, and A. N. Lagar'kov, "Theory of giant magneto-impedance effect in amorphous wires with different types of magnetic anisotropy", *J. Magn. Magn. Mater.*, vol. 185, pp. 159-173, 1998.
- [15] Y. Honkura, "Development of amorphous wire type MI sensors for automobile use", *J. Magn. Magn. Mater.* vol. 249, pp. 375-381, 2002.
- [16] T. Uchiyama, K. Mohri, and Sh. Nakayama, "Measurement of Spontaneous Oscillatory Magnetic Field of Guinea-Pig Smooth Muscle Preparation Using Pico-Tesla Resolution Amorphous Wire Magneto-Impedance Sensor", *IEEE Trans. Magn.*, vol. 47, pp. 3070-3073, 2011.
- [17] D. Makhnovskiy, A. Zhukov, V. Zhukova, and J. Gonzalez, "Tunable and self-sensing microwave composite materials incorporating ferromagnetic microwires", *Advances in Science and Technology*, vol. 54, pp. 201-210, 2008.
- [18] D. Praslička et al., "Possibilities of Measuring Stress and Health Monitoring in Materials Using Contact-Less Sensor Based on Magnetic Microwires", *IEEE Trans. Magn.*, vol. 49(1), pp. 128-131, 2013.
- [19] A. Allue et al., "Smart composites with embedded magnetic microwire inclusions allowing non-contact stresses and temperature monitoring", *Compos.Part A Appl.*, vol. 120, pp. 12-20, 2019, DOI: 10.1016/j.compositesa.2019.02.014
- [20] V. Zhukova et al., "Free Space Microwave Sensing of Carbon Fiber Composites With Ferromagnetic Microwire Inclusions", *IEEE Sens. Lett.*, vol. 8, No. 1, p. 2500104, 2024.
- [21] D. A. Allwood, G. Xiong, C. C. Faulkner, D. Atkinson, D. Petit, and R.P. Cowburn, "Magnetic domain-wall logic", *Science*, vol. 309, pp. 1688-1692, 2005.
- [22] S. Parkin, S.-H. Yang, Memory on the racetrack. *Nat. Nanotechnol.*, vol. 10, pp. 195-198, 2015.
- [23] A. Zhukov, A. Talaat, J. M. Blanco, M. Ipatov, and V. Zhukova, "Tuning of magnetic properties and GMI effect of Co-based amorphous microwires by annealing", *J. Electr. Mater.*, vol. 43(12), pp. 4532-4539, 2014, doi: 10.1007/s11664-014-3348-2.