

## MAGNETIC PROPERTIES AND APPLICATIONS OF GLASS-COATED FERROMAGNETIC MICROWIRES

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**Keywords:** Magnetic microwires, amorphous wires, GMI effect, domain wall dynamics

### ABSTRACT

Magnetic wires have attracted considerable attention due to their rather attractive magnetic properties such as giant magneto-impedance (GMI) effect or magnetic bistability, potentially suitable for several prospective applications (magnetic and magnetoelastic sensors, magnetic memory and logic, electronic surveillance, etc.) [1,2]. Glass-coated magnetic microwires prepared using the Taylor-Ulitovsky technique with thin metallic nucleus (typically with diameters 0.1 to 100  $\mu\text{m}$ ) covered by flexible, insulating and biocompatible glass are therefore quite interesting for sensor applications [2]. This technique allows preparation of magnetic wires with amorphous or crystalline structure of metallic nucleus. 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 [3].

The purpose of this paper is present last 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.

The impact of post-processing on soft magnetic properties and the giant magnetoimpedance (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. 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 higher magnetoimpedance ratio. Observed stress-induced anisotropy and related changes of magnetic properties are discussed considering internal stresses relaxation and “back-stresses”.

Consequently, stress annealing of ferromagnetic microwires allows achievement of interesting combination of magnetic properties.

## INTRODUCTION

Scientific and practical interest in the studies of various types of magnetic wires have increased significantly over the past three decades [1-4]. One of the main reasons for such growing interest in magnetic wires is the development of amorphous magnetic materials with unique combination of soft magnetic properties and improved mechanical and anticorrosive properties [1,4]. Among the most remarkable magnetic properties of amorphous materials with cylindrical symmetry are magnetic bistability and the Giant Magnetoimpedance (GMI) effect [1-4].

The aforementioned magnetic bistability is associated with ultrafast magnetization switching through a large and single Barkhausen jump [3,4]. The mechanism of such a unique magnetization reversal process is the fast propagation of a single domain wall (DW) through a single internal domain with an axial magnetization orientation. The hysteresis loop of such magnetic wires presents perfectly rectangular shape.

The magnetic bistability observed in various types of amorphous wires is usually closely related to a peculiar domain structure consisting of an inner single-domain axially magnetized core surrounded by an outer domain shell [3,4].

The magnetic bistability of amorphous wires is proposed for several applications, such as magnetic and magnetoelastic sensors or electronic article surveillance [3-6]. On the other hand, controllable DW propagation is proposed for magnetic logics and memory applications [7,8].

On the other hand, the aforementioned GMI effect is intrinsically related to high magnetic permeability of magnetically soft amorphous wires, which can present high circumferential magnetic permeability,  $\mu_\phi$  [2,9,10]. The main advantages of the magnetic field sensors involving GMI effect is their high sensitivity allowing detection of even pT magnetic fields [9,11].

All aforementioned magnetic properties are related to both perfectly cylindrical shape of amorphous wires, peculiar magnetoelastic anisotropy as well as to amorphous structure [1-4]. The combination of such factors results in the peculiar domain structure of amorphous wires consisting of an inner axially magnetized core surrounded by an outer domain shell [1,3, 12-14].

The aforementioned applications demand high quality magnetic wires. Thus, most industrial applications require diameter miniaturization, robust mechanical properties and improved corrosion properties. These properties, along with biocompatibility, are provided by glass-coated amorphous microwires. Such glass-coated magnetic microwires can be prepared using the Taylor-Ulitovsky technique allowing the preparation of microwires with metallic nucleus diameters ranging from 0.1 to 100  $\mu\text{m}$  covered by insulating, flexible and biocompatible glass coating [1,4, 15-17].

The performance of proposed applications essentially linked to aforementioned magnetic properties: DW propagation velocity, GMI ratio magnitude, tenability of the switching field of

magnetically bistable microwires etc. Therefore, finding appropriate post-processing allowing the optimization of aforementioned magnetic properties becomes a topic of extensive research. The most common post-processing consists of annealing under carefully selected conditions aiming to improve the magnetic properties and GMI effect of glass-coated microwires.

Accordingly, the purpose of this paper is to present our latest results on effect of post-processing on magnetic softness, GMI effect and DW dynamics of glass-coated microwires.

## EXPERIMENTAL METHODS AND MATERIALS

We prepared Fe-Co-Ni based glass-coated microwires with either positive or negative magnetostriction coefficient,  $\lambda_s$ , and with different diameters of the metallic nucleus,  $d$ , and total diameters,  $D$  using the Taylor-Ulitovsky method described elsewhere [4, 15-17].

The axial hysteresis loops of the studied samples have been measured using the fluxmetric method, as described earlier [18]. For correct comparison of the samples with different chemical compositions, geometry and subjected to heat treatments, we plotted the hysteresis loops as the normalized magnetization,  $M/M_0$ , versus the applied magnetic field,  $H$ . Here,  $M$  is the magnetic moment at a given  $H$ -value, and  $M_0$ - the sample's magnetic moment at the maximum applied field.

The DW propagation has been studied using the modified Sixtus-Tonks-like technique described elsewhere [19]. The experimental setup consists of three pick-up coils mounted on the non-magnetic capillary in which studied 12 cm long microwire is inserted spaced by a 27 mm distance.

The experimental setup consists of three pick-up coils separated by a distance of 27 mm, mounted on a non-magnetic capillary into which the studied 12 cm long microwire is inserted. A uniform axial magnetic field,  $H$ , is generated by a 140 mm long solenoid using a square wave signal with a frequency of 10 Hz. The DW velocity,  $v$ , is then estimated from the time difference,  $\Delta t$ , in the electromotive force,  $emf$ , peaks induced in the pick-up coils, separated by a distance  $l$ , of the moving DW and defined as [19]:

$$v = \frac{l}{\Delta t} \quad (1)$$

The impedance,  $Z$ , and  $Z(H)$  dependencies have been evaluated from the reflection coefficient,  $S_{11}$ , measured using the vector network analyzer in the frequency,  $f$ , range up to 1 GHz, as previously described [20]. The magnetic field dependencies of the GMI ratio,  $\Delta Z/Z$ , is then evaluated from  $Z(H)$  dependencies using the commonly accepted definition given as [2,4,9-11]:

$$\Delta Z/Z = [Z(H) - Z(H_{max})] / Z(H_{max}) \cdot 100 \quad (2)$$

where  $H_{max}$  - the maximum applied DC magnetic field (up to 18 kA/m).

The magnetostriction coefficient,  $\lambda_s$ , of studied samples has been evaluated by the so-called small angle magnetization rotation (SAMR) method using specially designed setup for characterization of magnetic microwires [21].

We studied as-prepared and annealed microwires. Samples have been annealed in a conventional furnace generally at temperature,  $T_{ann}$ , below 400 °C. When the stress,  $\sigma$ , was applied during the annealing, different young modulus of the metallic nucleus and glass were considered to evaluate correctly  $\sigma$  –value, as described elsewhere [9].

## EXPERIMENTAL RESULTS AND DISCUSSION

### AS-PREPARED SAMPLES: EFFECT OF THE CHEMICAL COMPOSITION AND GEOMETRY.

Typically, the magnetic bistability is observed in as-prepared Fe-rich glass-coated microwires with high and positive  $\lambda_s$  (see Fig.1a), the best magnetic softness is observed for Co-rich compositions with vanishing  $\lambda_s$  (see Fig.1b), while almost unhysteretic loops with rather high magnetic anisotropy field Co-rich microwires with negative  $\lambda_s$ . Consequently, magnetostriction coefficient,  $\lambda_s$ , is one of the main parameters affecting the hysteresis loops of as-prepared glass-coated microwires.

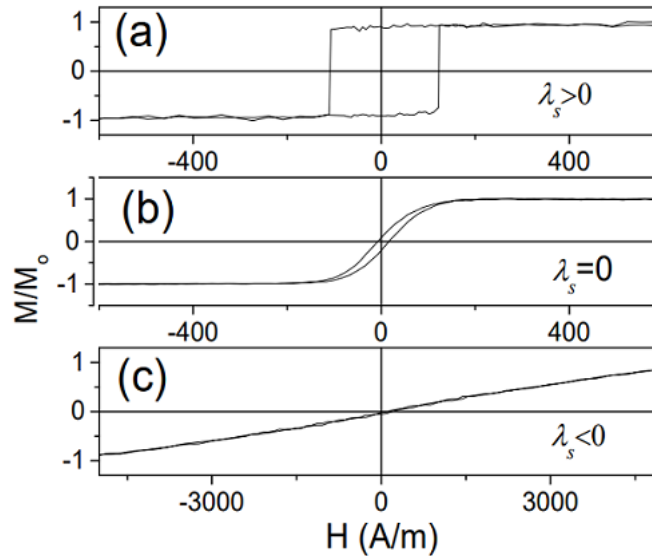


Figure 1: The hysteresis loops of glass-coated microwires of different chemical compositions and with different  $\lambda_s$ -values: (a) Fe-rich (Fe70B15Si10C5,  $\lambda_s > 0$ ), (b) Co-rich (Co67.1Fe3.8Ni1.4Si14.5B11.5Mo1.7,  $\lambda_s \approx 0$ ) and (c) Co-rich (Co77.5Si15B7.5,  $\lambda_s < 0$ ).

The peculiarity of glass-coated microwires with respect to the other families of amorphous magnetic wires is rather different character of hysteresis loops of Co-rich microwires with negative (of the order of  $10^{-6}$ )  $\lambda_s$  –values. As can be observed in Fig.1, such microwires have almost unhysteretic loops, while Co-rich conventional wires exhibit rectangular hysteresis loop.

This difference is commonly attributed to the strong internal stresses associated to the presence of non-magnetic glass-coating: the different thermal expansion of glass and metallic alloy is the main source of such internal stresses [4,15,22]. The magnitude of the internal stresses,  $\sigma_i$ , associated to difference in thermal expansion coefficients is directly to the  $\rho$ -ratio defined as  $d/D$ :  $\sigma_i$  increase with decreasing of the  $\rho$ -ratio [4,15]. Therefore, the internal stresses can be controlled through the  $\rho$ -ratio during the glass-coated microwires. Accordingly, the hysteresis loops of both Fe-rich and Co-rich microwires can be effectively tuned by changing the  $\rho$ -ratio. Two examples for Fe70B15Si10C5 and Co67.1Fe3.8Ni1.4Si14.5B11.5Mo1.7 amorphous glass-coated microwires are provided in Fig. 2.

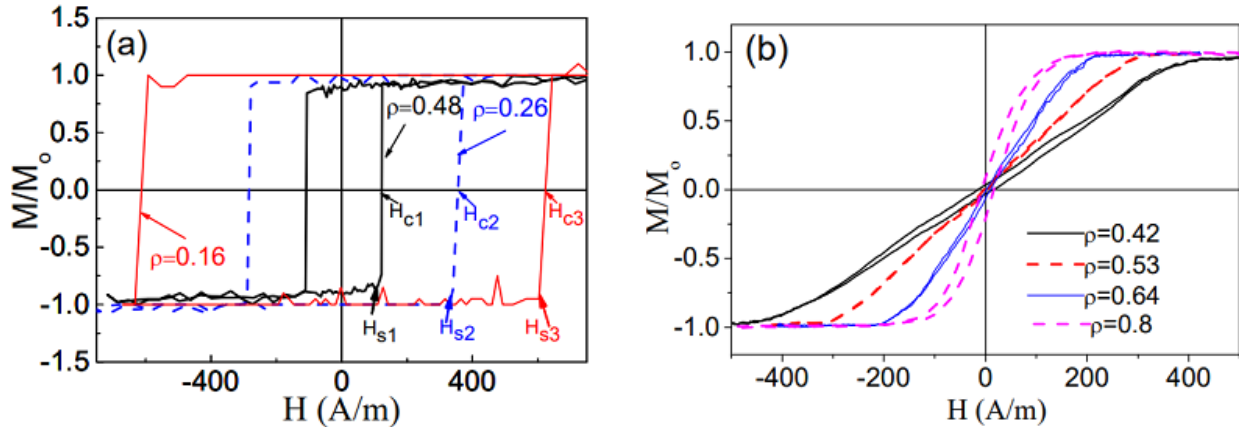


Figure 2: Effect of  $\rho$ -ratio on hysteresis loops of Fe70B15Si10C5 (a) Co67.1Fe3.8Ni1.4Si14.5B11.5Mo1.7 (b) amorphous glass-coated microwires.

Although the overall character of the hysteresis loops of both studied microwires remains almost the same, several parameters, such as switching field,  $H_s$ , of Fe-rich microwires or magnetic anisotropy field,  $H_k$ , of Co-rich microwires.

The possibility to tune the switching field values by changing the  $\rho$ -ratio has been proposed for electronic surveillance applications: such microwires are suitable for multi-bit magnetic tags design [6].

Regarding the GMI effect of as-prepared microwires: as expected, higher  $\Delta Z/Z$  -values are observed in Co-rich microwires due to their better magnetic softness. Thus, at fixed measurement conditions ( $f=100$  MHz), two orders of magnitude higher maximum GMI ratio,  $\Delta Z/Z_m$ , is observed in Co-rich microwires with vanishing  $\lambda_s$  (see Fig. 3). In this particular case  $\Delta Z/Z_m > 100\%$  is obtained for Co-rich microwires, while in carefully selected microwires even higher  $\Delta Z/Z_m$  -values can be observed [9].

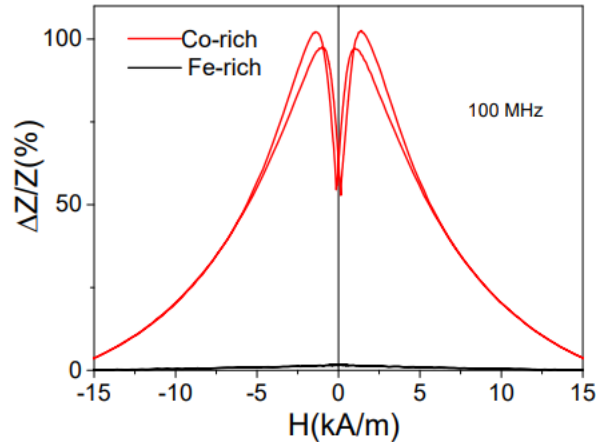


Figure 3:  $\Delta Z/Z$  (H) dependencies of Fe<sub>75</sub>B<sub>9</sub>Si<sub>12</sub>C<sub>4</sub> and Co<sub>69.2</sub>Fe<sub>3.6</sub>Ni<sub>1</sub>B<sub>12.5</sub>Si<sub>11</sub>Mo<sub>1.5</sub>C<sub>1.2</sub> microwires measured at 100 MHz.

### TAILORING OF MAGNETIC PROPERTIES OF AMORPHOUS MICROWIRES BY POST-PROCESSING

Given the high magnitude of internal stresses, annealing can be quite efficient in controlling the magnetic properties of glass-coated microwires. As shown in Fig. 4, the hysteresis loops of Co-rich microwires are quite sensitive to annealing: a transformation of almost unhysteretic loops into rectangular is observed upon annealing. Such change in hysteresis loops shape is observed for various Co-rich glass-coated microwires with vanishing  $\lambda_s$  –values [23]. The origin of such unexpected magnetic hardening upon annealing is explaining considering effect of annealing on  $\lambda_s$  value and even sign and the competition of shape and magnetoelastic anisotropies in magnetic microwires [4,23].

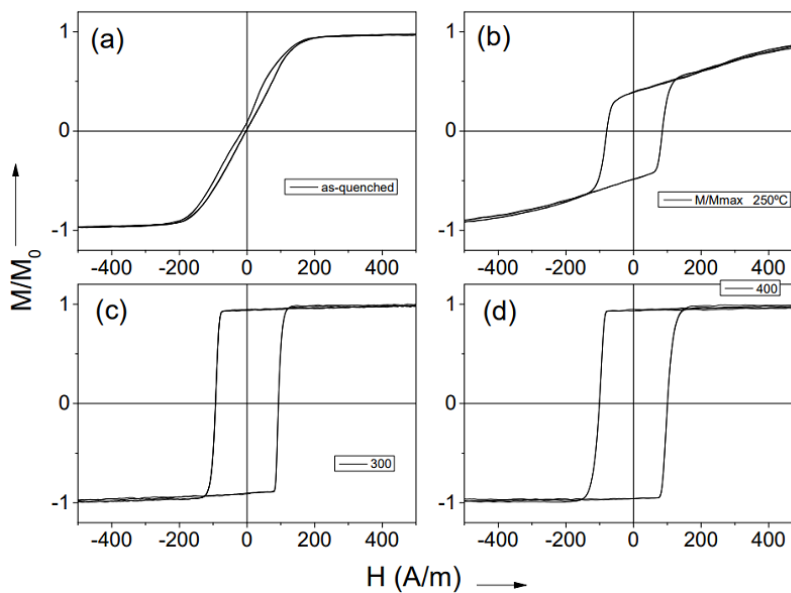


Figure 4: Hysteresis loops of as-prepared (a) and annealed for 60 min at  $T_{\text{ann}}=250$  oC (b), 300 oC (c) and 400 oC (d) Co<sub>69.2</sub>Fe<sub>3.6</sub>Ni<sub>1</sub>B<sub>12.5</sub>Si<sub>11</sub>Mo<sub>1.5</sub>C<sub>1.2</sub> microwires.

In contrast to Co-rich microwires, the hysteresis loops of Fe-rich glass-coated microwires are less affected by annealing: the rectangular character of hysteresis loops is maintained after annealing (see Fig.5). However, recently it was observed that the magnetic bistability of amorphous Fe-rich glass-coated microwires is lost upon heating above 100 oC, while such change of hysteresis loops is almost completely reversible: after cooling to ambient temperature the magnetic bistability recovers [24].

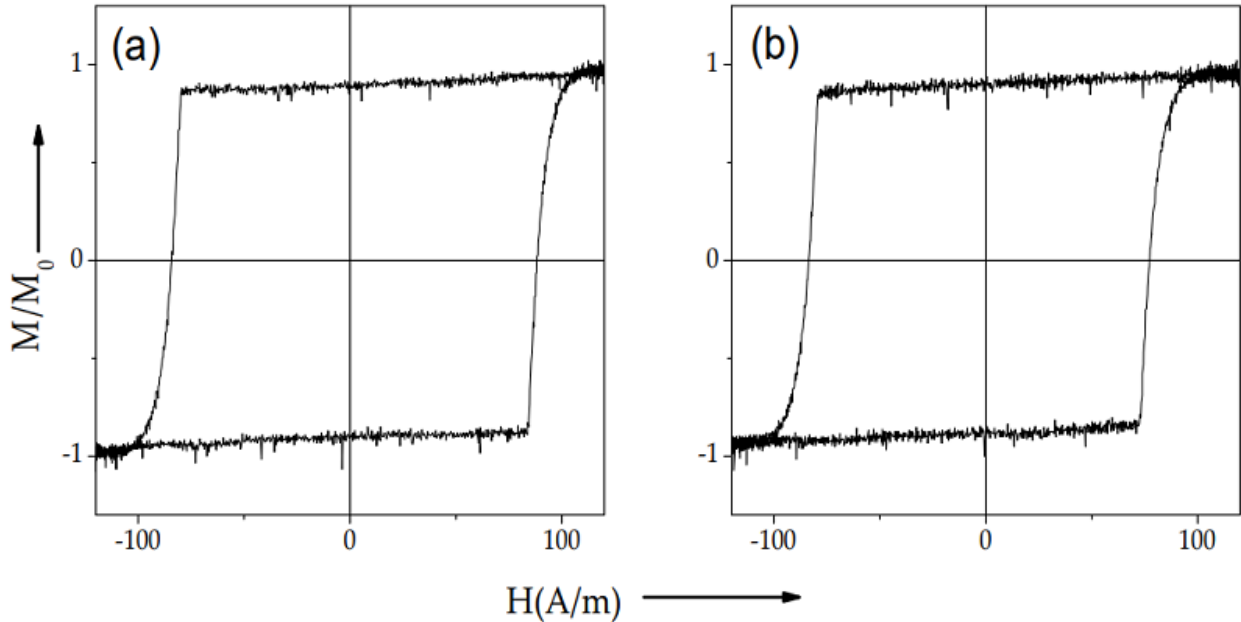


Figure 5: Hysteresis loops of as-prepared (a) and annealed for 120 min at  $T_{ann}=400$  oC (b) Fe75B9Si12C4 microwires.

As expected from perfectly rectangular shape of hysteresis loops of both as-prepared and annealed Fe-rich microwires (see Fig.5) and annealed Co-rich microwires (see Fig.4), the remagnetization process of such microwires runs by fast propagation of single DW. The  $v(H)$  dependencies measure in these microwires are provided in Fig. 6. In all the cases,  $v$  –values above 1 km/s have been obtained. Additionally,  $v$  –values and  $v(H)$  dependence of Fe-rich microwire are substantially affected by annealing. The remarkable improvement of  $v$  –values upon annealing of Fe-rich microwires was reported and discussed in terms of the effect of magnetoelastic anisotropy,  $K_{me}$ , on the DW mobility, S [19, 25].

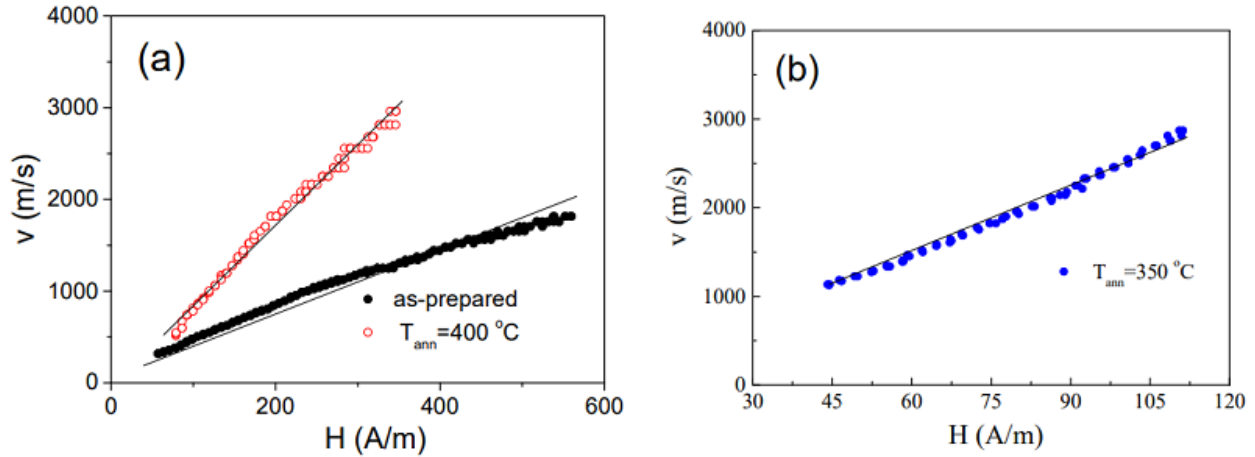


Figure 6:  $v(H)$  dependences measured in as-prepared and annealed for at  $T_{ann} = 400$  oC Fe77.5Si7.5B15 microwires (a) and in annealed at  $T_{ann} = 400$  oC Co69.2Fe3.6Ni1B12.5Si11Mo1.5C1.2 microwires.

Observed DW dynamics can be described by  $v(H)$  linear dependence according to the expression:

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

where  $H_0$  is the critical propagation field, below which the domain wall propagation is not possible.

The domain wall mobility,  $S$ , is commonly expressed as [19,25]:

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

where  $H_0$  is magnetic permeability of vacuum,  $M_s$  -saturation magnetization and  $\beta$  is the viscous damping coefficient, which is substantially affected by the  $K_m$ .

The observed quite fast DW propagation in annealed Fe-rich and Co-rich amorphous microwires, characterized by high  $v$  (up to 3 km/s) and  $S$ -values, can be suitable for the proposed applications using controllable DW propagation [7,8].

As regarding the GMI effect, magnetic microwires with spontaneous or annealing-induced magnetic bistability generally do not present a high GMI ratio. Recently, it was reported that annealing under stress can suppress magnetic bistability in both Fe-rich and Co-rich microwires [4]. The modification of the hysteresis loops upon stress-annealing of Fe-rich microwire is demonstrated by Fig. 7. At high enough  $\sigma$ -value a substantial transverse magnetic anisotropy is induced.

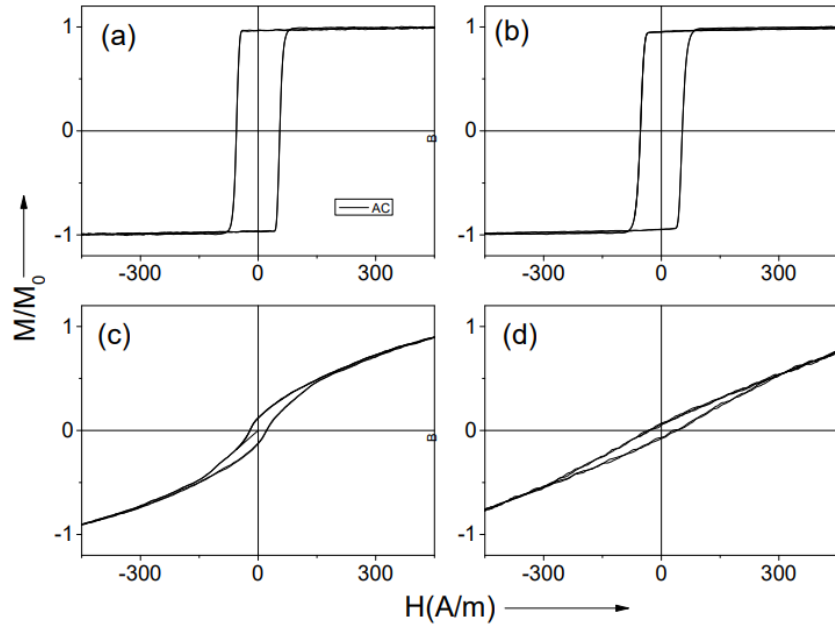


Figure 7: The hysteresis loops of as-prepared (a) annealed at 350 oC for  $\sigma=0$  (b)  $\sigma=190$  MPa (c) and  $\sigma=380$  MPa (d) Fe75B9Si12C4 microwires.

As shown in Fig. 8, a remarkable (an order of magnitude) increase in GMI effect is observed in Fe-rich microwires with such transverse magnetic anisotropy induced by stress-annealing.

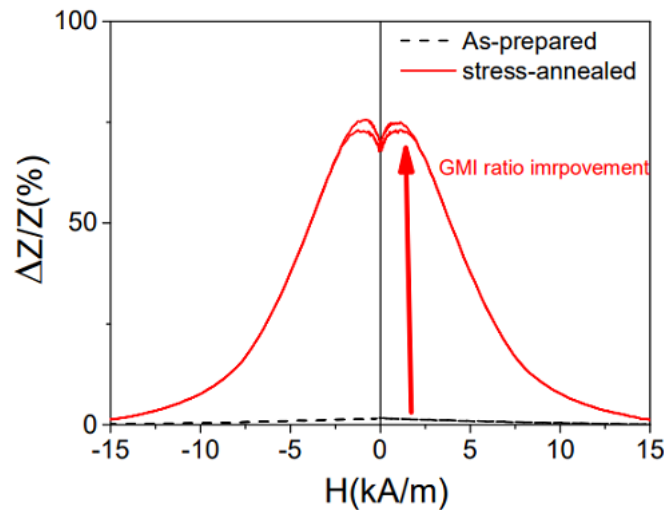


Figure 8:  $\Delta Z/Z(H)$  dependencies observed of as-prepared and stress-annealed at  $T_{ann}=350$  oC (for 1 hour and  $\sigma=380$  MPa) Fe75B9Si12C4 microwires measured at 100 MHz.

Likewise, with a high enough stress applied during annealing, magnetic bistability can be suppressed in Co-rich microwires. As can be seen in Fig. 9, even better magnetic softness can be obtained after stress-annealing if the  $\sigma$ -value is large enough. Accordingly, GMI performance can be substantially improved by appropriate stress-annealing, as shown in Fig. 10.

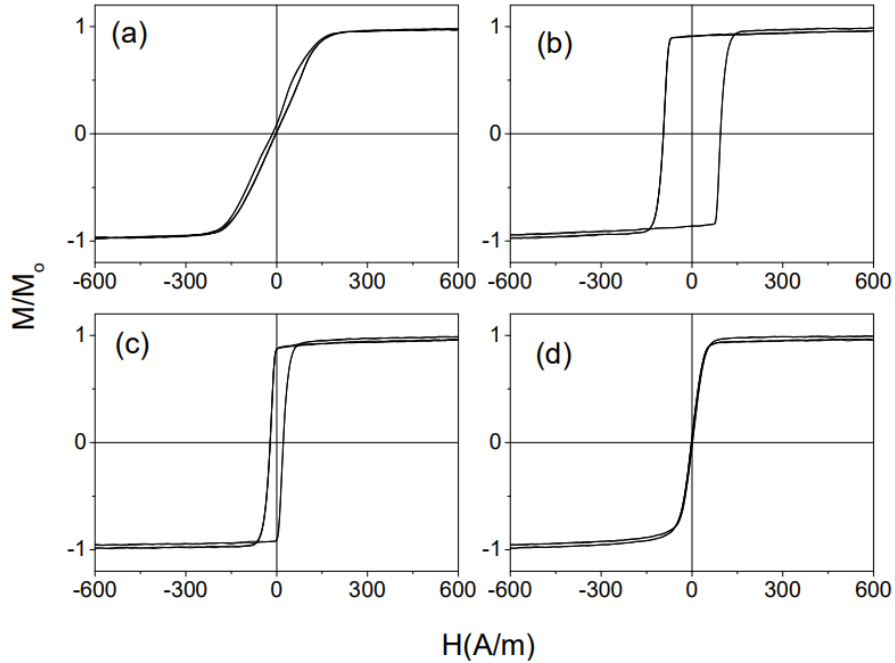


Figure 9: The hysteresis loops of as-prepared (a) annealed at 350 oC for  $\sigma=0$  (b)  $\sigma=354$  MPa (c) and  $\sigma=472$  MPa (d)  $\text{Co}_{69.2}\text{Fe}_{3.6}\text{Ni}_{1\text{B}12.5}\text{Si}_{1\text{M}o1.5}\text{C}_{1.2}$  microwires.

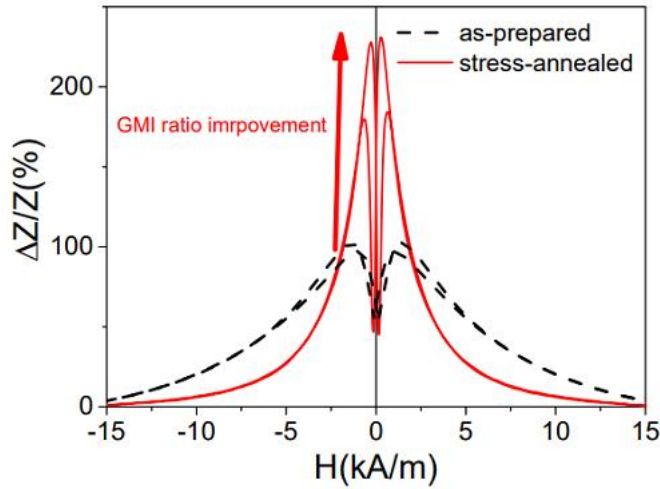


Figure 10:  $\Delta Z/Z(H)$  dependencies observed of as-prepared and stress-annealed at  $T_{\text{ann}}=350$  oC (for 1 hour and  $\sigma=472$  MPa)  $\text{Co}_{69.2}\text{Fe}_{3.6}\text{Ni}_{1\text{B}12.5}\text{Si}_{1\text{M}o1.5}\text{C}_{1.2}$  microwires measured at 100 MHz.

The origin of observed stress-annealing induced anisotropy in amorphous materials is discussed since 70-s [26,27]. Stress and magnetic field induced magnetic anisotropy in amorphous materials was discussed in terms of compositional and topological short-range ordering and atomic pair ordering upon annealing [26-28]. On the other hand, the presence of the glass-coated can be the origin of “back” stresses arising during the cooling and subsequent redistribution of the internal stresses after stress-annealing [29]. In the present case the transverse magnetic anisotropy was induced in both Fe-rich and Co-Fe-rich microwires. Therefore, the atomic pair ordering mechanism of such stress-annealing induced anisotropy looks less convincing.

## CONCLUSION

Summarizing the obtained experimental results, we showed that carefully selected postprocessing of both Fe-rich and Co-rich glass-coated microwires including annealing or stress-annealing can substantially improve magnetic properties of both families of studied microwires. Particularly, although commonly less-expensive Fe-rich microwires were not considered for GMI applications, magnetic softness and GMI effect of Fe-rich glass-coated microwires can be remarkably improved by stress-annealing. Additionally, magnetic properties and the GMI ratio of as-prepared microwires can be tailored by the appropriate post-processing.

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