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INFLUENCE OF THE INHOMOGENEITIES ON MAGNETIC PROPERTIES OF GLASS-COATED MICROWIRES

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Summary: *We observed that most relevant magnetic properties, like GMI effect and DW propagation velocity are limited by the microwires technology, i.e. by the interfacial layer and inhomogeneities. Using direct method we observed gas bubbles within the glass coating with volume content of about 8-10 %. The sizes of the bubbles were between 1 and 15 μm. The existence of such bubbles might be the origin of the inhomogeneities in the internal stresses distribution. We observed the existence of the interfacial layer and obtained the elements distribution between the glass coating and metallic nucleus. This allowed us to estimate that the thickness of the interfacial layer is about 0.5 μm.*

1 INTRODUCTION

Magnetically soft composite microwires consisting of amorphous metallic nucleus covered by glass coating attracted great attention owing to a number of outstanding magnetic properties such as magnetic bistability and giant magneto-impedance, GMI, effect [1, 2].

Generally magnetic properties of amorphous ferromagnetic microwires depend on the composition of the metallic nucleus as well as on the composition and thickness of the glass coating. For conventional amorphous materials main compositional dependence is related with stresses arising from the rapid solidification process, when the metallic alloy solidifying from the surface. Stronger dependence of magnetic properties of glass-coated microwires on metallic nucleus composition is originated to the additional internal stresses arising from the glass-coating. Main feature of the fabrication technique of glass-coated microwires is that it involves the simultaneous solidification of composite microwire consisting of ferromagnetic nucleus surrounded by glass coating. Quite different thermal expansion coefficients of the glass and the metallic alloys introduce considerable internal stresses inside the ferromagnetic nucleus during simultaneous fast solidification of the composite microwire [1- 3]. Strength of these internal stresses depends on ρ-ratio defined as the $\rho=d/D$, where d is the metallic nucleus diameter and D-total microwire diameter. The estimated values of the internal stresses in these glass coated microwires arising from the difference in the thermal expansion coefficients of metallic nucleus and glass coating are of the order of 100-1000 MPa, depending strongly on the ratio between the glass coating thickness and metallic core diameter [2,4], increasing with the glass coating thickness.

Consequently, tailoring of the magnetoelastic energy, K_{me}, is essentially important for optimization of magnetic properties of glass-coated microwires [1, 2]. Considerable attention has been paid to studies of correlation between the magnetoelastic energy, K_{me}, induced by the stresses and magnetic properties.

On the other hand, the glass-coating technique involves complex metallurgical processes, such as the effect of electromagnetic field of inductor on alloy ingot and the interaction between the alloy and the glass coating at elevated temperatures. These problems are less studied [2]. Additionally the technology has been initially developed for the non-magnetic alloys [4]. The character of interaction depends on chemical composition of the ingot as well as on type of glass used for the casting [2,4].

Generally, small particles of the glass can be involved by the molten metal ingot moving under the effect of the high-frequency magnetic field of the inductor. Therefore the chemical composition of the ingot at the beginning of the casting process can be different from the alloy composition from that at the end of casting process.

Additionally, for the case of non-magnetic alloys the interfacial layer between the metallic nucleus and glass coating has been observed and studied [4]. As regarding the origin and the properties of the interfacial layer, there are few possibilities described for the non-magnetic alloys. Thus, among the origins of the interfacial layer the following considerations have been discussed:

1. Formation of the series of solid solutions. In this case the thickness of the interfacial layer is about few μm.
2. Uncompensated molecular forces on the interface between the glass and the metallic nucleus. In this case the thickness of the interfacial layer should be less than 0,1 μm.
3. Formation of stable chemical compounds with the structure different (from crystallographic point of view) from that of the interacting materials. The thickness of the interfacial layer is of the same order (few μm) as in the case of formation of solid state solutions, but the interface layer is more visible, like it was observed by microscopy in few cases [4].

Taking into existence of the interfacial layer we must consider, that glass-coated microwire consist of metallic nucleus, glass insulating shell and interfacial layer.

From the point of view of high-frequency magnetic properties (like the Giant Magneto Impedance, GMI, effect) the interfacial layer between the metallic nucleus and glass coating is especially relevant [5,6]. At higher frequencies the current flows closer to the surface, then the effective anisotropy field and permeability dispersion can change with frequency [5,6].

The other source of instability of properties of cast microwire is related with gas content inside the microwire. The sources of the gas are: the atmosphere, the gas impurities in the alloy and the glass. Some content of oxygen and/or hydrogen (about 5 cm^3/100 g each other) and even nitrogen has been detected. Gaseous precipitations can cause the metallic nucleus deformation, cracks and even discontinuities. Chemical reactions of hydrogen with the oxides of the metals can result in appearance of water bubbles inside the metallic nucleus as well as in the glass coating.

Recently we showed that the microwires's inhomogeneities sufficiently affect the remagnetization process of magnetically bistable microwires limiting single domain wall, DW, propagation regime as well as the domain wall propagation velocity, v [7,8].

We observed the correlation of the domain wall dynamics with the distribution of the nucleation fields.

On the other hand, we observed that there is a correlation between the distribution of the local nucleation field along the length and the domain wall, DW, dynamics. Thus if external magnetic field, H, is below some critical value we observed single DW propagation along the wire axis, manifested as linear $v(H)$ dependence. Measuring the local nucleation fields, H_n, along the microwire length, L, we observed considerable oscillations and even dip holes on the $H_n(L)$ dependences identified as the positions of localized defects existing within the microwire [9]. The minimum value of local nucleation field, H_{nmin}, determines the threshold between single and multiple DW propagation regimes. Thus, we observed the spontaneous DW nucleation on local defects, which limits the single DW propagation regime in magnetically bistable microwires [8,9]. Below H_{nmin} there is a linear dependence of DW velocity, v, on the applied magnetic field, H, for all microwires with positive sign of the magnetostriction constant. Above H_{nmin} we observed abrupt jumps on the $v(H)$ dependences associated with multiple DW propagation regime.

Consequently, the microwires inhomogeneities sufficiently affect the remagnetization process [1,2,8,9]. The origin of the defects is unclear, and might be related to stress inhomogeneities, shape irregularities, oxides etc. It is assumed, that at least some of these defects might have a magnetoelastic origin and, therefore, might be affected by heat treatment.

In this paper we present our last results on studies of the metallurgical processes during the microwires fabrication on structure and magnetic properties (DW dynamics and GMI effect) of glass-coated microwires. We are paying special attention to studies of the interfacial layer between the metallic nucleus and glass coating and studies of the inhomogeneities related with fabrication process of thin ferromagnetic microwires.

2 EXPERIMENTAL METHOD

We studied amorphous Co rich and Fe-rich glass-coated microwires with different metallic nucleus diameter, d, and total microwire diameter, D, were produced by modified Taylor-Ulitovsky method [1-3]. It is worth mentioning, that the strength of internal stresses is determined by ratio ρ [3, 4, 10]. Therefore, controllable change of the ρ–ratio allowed us to control the residual stresses.

Hysteresis loops have been determined by fluxmetric method, as described elsewhere [4].

We have measured dependences of the diagonal Z_{zz} and off-diagonal $Z_{\varphi z}$ impedance components and GMI ratio, $\Delta Z/Z$, on external axial magnetic field H in microwires, as described elsewhere [5, 6, 11].

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

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

An axial *DC*-field with intensity up to 8 kA/m was supplied by a magnetization coils. The dependence of the magnetoimpedance ratio, $\Delta Z/Z$, on the axial field, H, at the driving *AC*-current, I, ranging from 0.75 up to 5 mA of the frequency, f, in the range 1 - 30 MHz, both treated as the parameters, have been investigated.

We used specially designed microstrip cell. One wire end was connected to the inner conductor of a coaxial line through a matched microstrip line while the other was connected to the ground plane. The components Z_{zz} and $Z_{\varphi z}$ were measured simultaneously using vector network analyzer. The diagonal impedance of the sample $Z_w = Z_{zz}l$, where l is the wire length, was obtained from reflection coefficient S_{11} and the off-diagonal impedance $Z_{\varphi z}$ was measured as transmission coefficient S_{21} as a voltage induced in a 2-mm long pick-up coil wounded over the wire. The static bias field H_B was created by the dc current I_B applied to sample through the bias-tee element. The other experimental details are given in Ref. 4. The frequency range for the off-diagonal component $Z_{\varphi z}$ was 10 – 300 MHz, while diagonal impedance component has been measured till 7 GHz.

We studied the DW dynamics of the microwires using a Sixtus–Tonks-like technique, using three pick-up coils instead of two or one (see, for example, [3,14]). Consequently we were able to measure DW velocities between the 1st and the 2nd pick-up coils, V_{1-2}, between the 1st and the 3d pick-up coils, V_{1-3}, and between the 2nd and the 3d pick-up coils, V_{2-3}. The distribution of the nucleation fields, $H_N(L)$, was measured using a technique described previously [8,9].

We used the microscope Axio Scope is A1 for the metallographic analysis and defects studies of glass-coated microwires (in reflected and transmitted light). Microscope Axio Scope A1 is a modular system that allows studying the material using several methods of contrast: transmitted light, polarization, phase contrast, dark or light field, reflected light-polarization, luminescence.

For studies of the surface of microwires we used a light field mode when using lenses with a numerical aperture of 0.8 and 0.5. The size of the obtained images of the microstructure of microwire: using lens 50x/0.8 HD: the length of the picture is 0.26 mm, the height is 0.2 mm. When using a 20x/0.5 HD lens: the length of the picture is 0.66 mm, height-0.485 mm.

To determine the composition of the sample and study the surface has been used scanning electron microscope JEOL JSM-6610. Scanning electron microscope allows obtaining images of the surface with high spatial resolution (a few nanometres), as well as to determine the composition, structure and properties of surface layers of materials. Analysis of the particles reflected from the surface provides information on relief of the surface of the phase composition and the crystalline structure of subsurface layers. Analysis of x-ray radiation from electron beam interaction with the sample allows to characterize qualitatively and quantitatively the chemical composition of the surface layers.

3 STUDIES OF THE INTERFACE LAYER AND DEFECTS IN GLASS-COATED MICROWIRES

Effect of geometric parameters on hysteresis loops of Fe and Co-rich microwires is shown in Fig.1. As previously discussed, geometric parameters significantly affect hysteresis

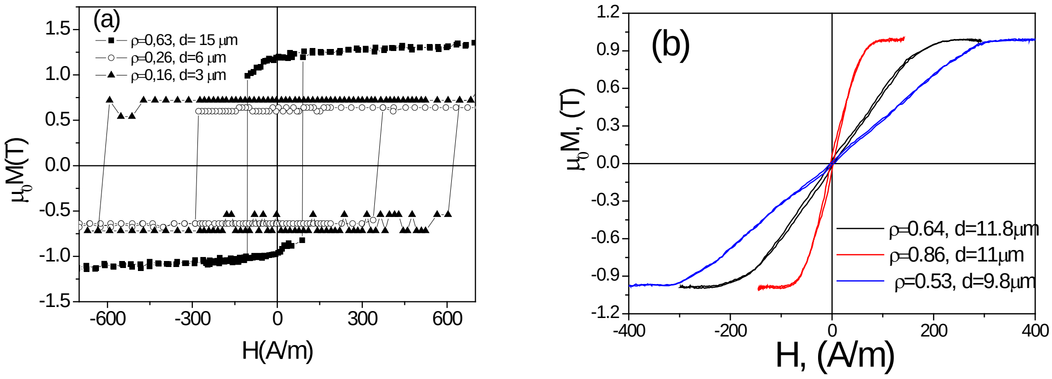


Figure 1. Hysteresis loops of amorphous $Fe_{70}B_{15}Si_{10}C_5$ (a) and $Co_{69.12}Fe_{4.01}Ni_{1.46}B_{11.63}Si_{10.83}Mo_{1.46}C_{1.49}$ (b) microwires with the same sample length and different metallic nucleus diameter d and total ρ – ratios.

loops of both compositions.

Considerable increasing of switching filed (from about 80 A/m up to 500 A/m) is of Fe-rich microwires is observed when ferromagnetic metallic nucleus diameter decreases from 15 to 3 μm (i.e. 5 times). At the same time, rectangular character of hysteresis loop shape is maintained even for smallest microwires diameters. Previously similar increasing of coercivity with decreasing the metallic nucleus diameters have been attributed to enhanced magnetoelastic energy arising from enhanced internal stresses when ρ– ratio is small [3]. Consequently, one of relevant parameters affecting strength of internal stresses and the magnetoelastic energy is ρ-ratio.

Additionally increasing of the magnetic anisotropy field with decreasing of the ρ– ratio observed in Co-rich microwires (Fig.1b) must be attributed to the increasing of the strength of internal stresses, when ρ– ratio decreases. But decreasing of the remanent magnetization observed in Fig.1a with decreasing of the ρ–ratio is unclear considering only the magnetoelastic contribution. Previously similar dependence has been reported in ref (12). Length of the samples was the same, so effect of demagnetizing factor cannot be considered for explanation of observed dependences (even the effect must the opposite).

For explanation of observed dependence of remanent magnetization of Fe-rich microwirs on ρ– ratio we can consider, that for lower metallic diameter the ratio between the surface and volume of metallic nucleus increases. Consequently this dependence might be explained if the surface layer of ferromagnetic metallic nucleus near the interface between the glass coating and metallic nucleus has lower magnetization. The reason for lower magnetization can be existence of the interfacial layer between the glass coating and metallic nucleus.

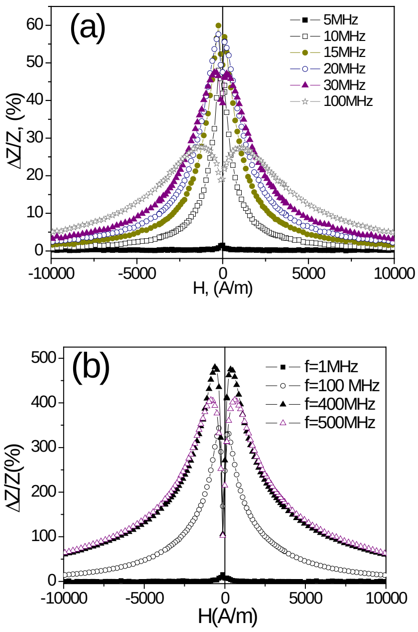


Figure 2. *ΔZ/Z(H)* dependences of $Co_{69,4}Fe_{4.3}Ni_{1.5}B_{11.7}Si_{11.9}Mo_{1.2}$ microwires with different ρ–ratios: $\rho \approx 0{,}59$; d=6; D=10,2μm (a), $\rho \approx 0{,}75$; d=8,2; D=11μm (b) and $\rho \approx 0{,}92$; d=21; D=22,8 μm (c).

From analysis of the magnetic field dependence of GMI ratio measured in $Co_{69.12}Fe_{4.01}Ni_{1.46}B_{11.63}Si_{10.83}Mo_{1.46}C_{1.49}$ microwires

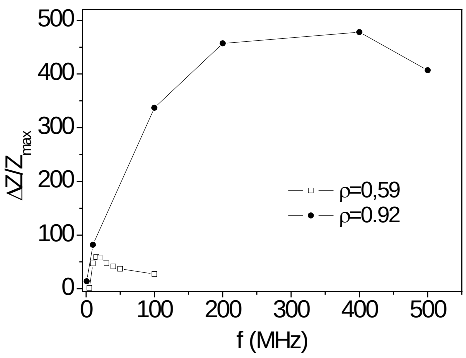


Figure3. Frequency dependence of $Co_{69.4}Fe_{4.3}Ni_{1.5}B_{11.7}Si_{11.9}Mo_{1.2}$ microwires with different geometric parameters (ρ-ratio).

at different frequencies we can deduce that all the exhibit optimum frequency at which highest GMI ratio is observed (see Fig.2). Moreover all $\Delta Z/Z$ present double-peak dependence with a maximum of $\Delta Z/Z$, $\Delta Z/Z_{max}$ at certain magnetic field. The field of maximum, H_{max} increases with frequency f;

For comparison we plotted $\Delta Z/Z_{max}(f)$ dependences for measured samples (see Fig.3). The origin of the maximum on $\Delta Z/Z_{max}(f)$ dependences might be again explained considering different magnetic anisotropy in the surface layer. Consequently existence of the interfacial layer might be one of the reasons for observed dependences.

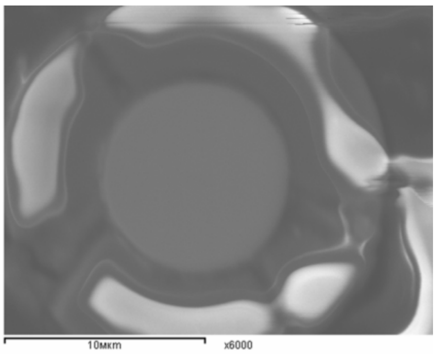
For studies of the interfacial layer we used scanning microscopy and studied cross section of the glass-coated mcrowires. At high enough magnification (x14000) we got the image of the interfacial layer (Fig.4a) and obtained the elements distribution in different point of the cross section (Fig.4b).

The existence of the interfacial layer is well observed in Fig.4.
This allowed us to estimate the thickness of the interfacial layer, t_{il}. For both Fe- and Co-rich microwires $t_{il} \approx 0.5$ µm.

Consequently we can assume that for submicrometric diameters of the metallic nucleus the interfacial layer may considerable affect magnetic properties and GMI effect.

As mentioned above, the remagnetization of Fe-rich samples exhibiting magnetic bistability effect is related with the depinning and fast domain wall, DW, propagation along the microwires [8, 9]. Recently using Sixtus-Tonks like method and local nucleation field distribution a correlation between the samples inhomogeneities (defects) and magnetic field dependence of DW velocity, *v* is observed.

Strong correlation between the type of the *v(H)* dependence and the distribution of the local nucleation fields can be observed in Fig.5. Even measuring *v(H)* dependence of the same microwire choosing different samples we can observe abrupt jumps on *v(H)* dependences at different fields (Fig. 5b). We observed that the magnetic field value, corresponding to such jump on *v(H)* dependences, correlates with the minimum nucleation field (see Fig.5a and b). At different part of the same sample the minimum nucleation field might be different due to the defects (see Fig.5a). Consequently defects rising in microwires determines threshold between single and multiple domain wall propagation regimes limiting maximum DW velocity that



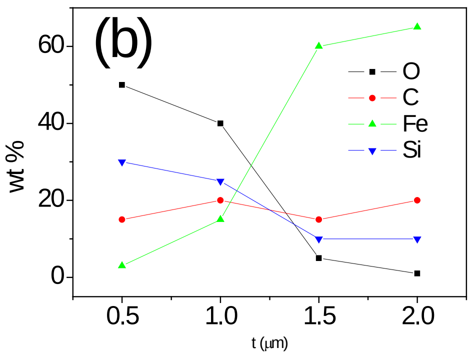


Figure 4. Image of the interfacial layer (a) and elements distribution (b) near interfacial layer in Fe-rich microwire

can be achieved in microwires (see Fig.5b).

To study the defects we employed the optical microscopy. Figure 6 shows the images of the Fe-Co-based glass-coated microwires. We can observe the bubbles inside glass (shown by the arrows in the image) forming during the fabrication process of the microwire.

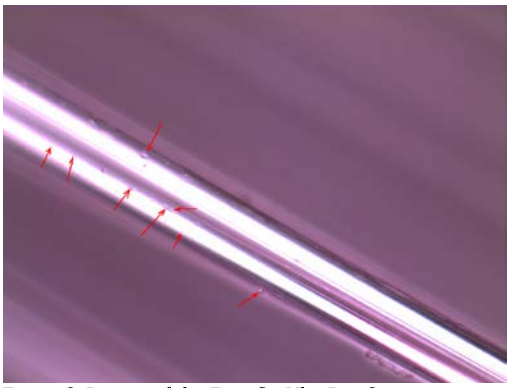


Figure 6. Images of the $Fe_{70.8}Cu_1Nb_{3.1}B_{10.6}Si_{14.5}$ microwires obtained using Axio Scope.

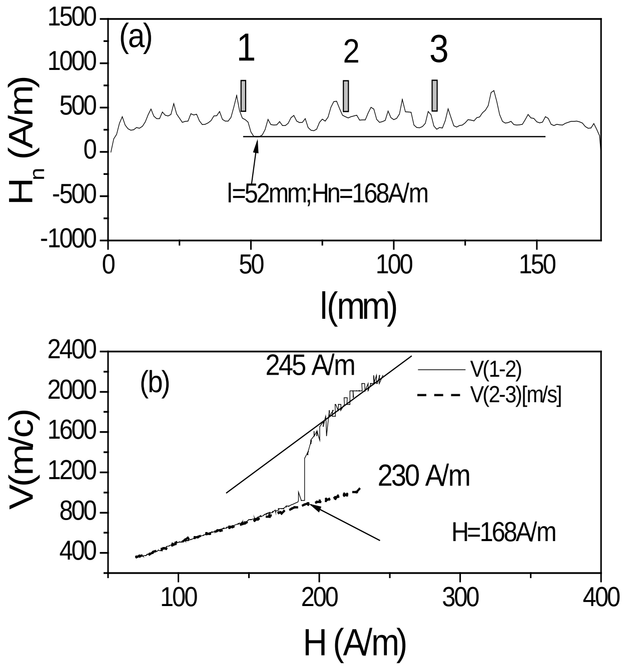


Figure 5. Correlation of local nucleation fields distribution (a) and *v(H)* dependences in magnetically bistable amorphous $Fe_{74}B_{13}Si_{11}C_2$ microwire. 1, 2, 3 are the positions of the pick-up coils.

These bubbles can potentially serve as places of origin of cracks when bending the wire. Additionally these bubbles may result in inhomogeneous stresses distribution along the sample length. The average sizes of bubbles and their volume fraction is different for studied samples (see table 1).

Table 1. Average size and volume content of the bubbles in Co- and Fe-rich microwires

Sample	Size range of bubbles, μm	Volume fraction of bubbles, %
Co-rich	1÷15	10±2
Fe-rich	3÷5	8±0,5

Volume fraction of bubbles is probably depends on the chemical composition of metallic nucleus as well as on the technology of amorphous microwires. Most likely, the volume fraction of such defects depends on the drawing speed of composite wire, composition of the metallic alloy, thermal conditions of the rapid quenching during the microwires fabrication.

Indeed, recently we observed considerable dependence of the local nucleation fields distribution and maximum DW velocity on heat treatment [13]. The annealing of microwires enhances the DW velocity by extending the field range for single DW propagation, as well as enhancing DW velocity at a given magnetic field due to internal stress relaxation. A correlation between the annealing influence on the local nucleation field distribution, and a change of magnetic field dependence of DW velocity, in amorphous Fe- based microwires was observed. Consequently, the nature of local defects limiting the single DW propagation regime and damping the DW might be related with the internal stress distribution near the gas bubbles in the glass-coating.

4 CONCLUSIONS

We observed that most relevant magnetic properties, like GMI effect and DW propagation velocity are limited by the microwires technology, i.e. by the interfacial layer and inhomogeneities.

We observed gas bubbles within the glass coating with volume content of about 8-10 %. The sizes of the bubbles were between 1 and 15 μm. The existence of such bubbles might be the origin of the inhomogeneities in the internal stresses distribution.

Using scanning electron microscope JEOL JSM-6610 we obtained the image of the interfacial layer and the elements distribution within the glass coating and metallic nucleus. This allowed us to estimate that the thickness of the interfacial layer is about 0.5 μm. Further understanding of the origins of the interfacial layer and defects may help for improvement of the existing technology for thin composite wires fabrication and enhance their magnetic properties.

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