Development of Free Space Microwave Sensing of Carbon Fiber Composites with Ferromagnetic Microwire Inclusions

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Abstract-In this work, we provide new experimental results on studies of composites with glass-coated ferromagnetic microwires aligned with the requirements of carbon composites. This work focuses on the free space microwave measurements of composites made from carbon fibers and ferromagnetic microwires inclusion focusing on the electromagnetic properties. We prepared and measured hysteresis loops and the magnetoimpedance effect of several microwires and selected Co-rich microwires with better magnetic softness and higher magnetoimpedance effect. We observed that, by using a low frequency modulating magnetic field allows us to distinguish the microwave signals originated by ferromagnetic microwires inclusions from that generated by the carbon fibers. The location of carbon fibers near magnetic microwires has a critical effect on the response signals (parameters S amplitude) obtained from such composites.

Keywords-magnetic microwires; magnetic softness; carbon fiber composite; magnetoimpedance effect.

I. INTRODUCTION

Amorphous soft magnetic materials, prepared by rapid melt quenching, can present excellent magnetic softness together with superior mechanical properties [1]-[5]. Thus, abrupt deterioration of the magnetic softness and mechanical properties (such as tensile yield) upon the devitrification of amorphous precursor is previously reported [4]. Additionally, the fabrication process of amorphous materials involving rapid melt quenching is fast and inexpensive [1]-[5]. Accordingly, amorphous soft magnetic materials are useful for numerous industrial applications (mostly for design of magnetic devices and magnetoelastic sensors) [8]-[12]. The development of novel applications of amorphous materials requires new functionalities, i.e., reduced dimensions, enhanced corrosion resistance or biocompatibility of the sensing material [11][13]. Therefore, great attention has been paid to development of alternative fabrication methods allowing preparation of amorphous materials at micro-nano scale involving melt quenching [11][13].

Glass-coated microwires prepared by the Taylor-Ulitovsky technique fit most of the requirements: such magnetic microwires have micro-nanometric diameters (between 0.5 and 100 μ m), covered with thin, insulating, biocompatible and flexible glass-coating and can present excellent magnetic softness or magnetic bistability [5] [11] [13]. Such features of glass-coated microwires allow development of exciting new applications in various magnetic sensors, as well as in smart composites with tunable magnetic permittivity [6][11][13]-[20]. One more advantage of glass-coated microwires is their excellent mechanical properties [4] [5].

Recently, the stress dependence of hysteresis loops and Giant Magnetoimpedance, GMI, effect have been proposed for the mechanical stresses monitoring in Fiber Reinforced Composites (FRC) containing microwires inclusions or using magnetoelastic sensors based on stress dependence of various magnetic properties [10] [20] [21].

One of the common problems in composite materials is monitoring of stresses and temperature. Usually, composite stress monitoring is performed by different sensors, like the pressure transducers and dielectric sensors [21]. However, these employed sensors are not wireless [21]. One of the proposed solutions for non-destructive FRC monitoring is by using of piezoelectric fibers with diameters of 10 to 100 μ m [22]. However, this solution requires electrodes to supply an electrical field, occupying a significant amount of space.

Among the promising solutions, addressing the problem of non-destructive FRC monitoring is a new sensing method involving free space microwave spectroscopy using inclusions of ferromagnetic microwire presenting the high frequency impedance quite sensitive to applied stress and magnetic field [21]. The aforementioned glass-coated microwires with metallic nucleus diameters of $0.2 - 100 \mu m$ can present excellent mechanical and corrosive properties (if produced with an amorphous structure), and hence perfectly suitable for the requirements of this technique, making it suitable for remote stresses and temperature monitoring in FRCs [18]-[21].

For the proposed application involving the nondestructive FRC monitoring glass-coated microwires must present good magnetic softness and high magnetoimpedance, MI, effect [18] [21]. Magnetic softness of amorphous microwires is substantially affected by the chemical composition of metallic nucleus: better magnetic softness and higher MI effect are reported for Co-rich microwires with vanishing magnetostriction [1] [5].

Accordingly, in this paper, we present our latest results on studies of magnetic properties of glass-coated Co-rich microwires and on our attempts to wirelessly health monitoring of composites containing both carbon fibers and ferromagnetic glass-coated microwires.

This paper is organized as follows. In Section 2, the experimental methods as well as the microwires characteristics analyzed in this paper are provided. Section 3 deals with experimental results dealing with free space microwave measurements of composites containing both carbon fibers and ferromagnetic glass-coated microwires. Finally, we conclude the paper in Section 4.

II. EXPERIMENTAL SYSTEM DETAILS

Generally, we prepared and analyzed two different types of magnetic amorphous microwires: i) amorphous microwires with high positive magnetostriction coefficients, λ_s , (Fe-rich) and ii) amorphous microwires with vanishing λ_s (Co-Fe-based microwires). We studied microwires with metallic nucleus diameters, *d*, ranging from 22 up to 38 µm and a total diameter, D, up to 45 µm, prepared using the modified Taylor-Ulitovsky method described elsewhere [11] [17]. The Taylor-Ulitovsky method allows the preparation of metallic microwires (with typical diameters of the order of 0.1 to 100 µm) covered with an insulating glass coating [11] [17].

Magnetic hysteresis loops of studied microwires have been measured using the fluxmetric method, previously described in detail elsewhere [16]. The hysteresis loops were represented as the dependence of normalized magnetization, M/M_0 (where *M* is the magnetic moment at a given magnetic field and M_0 is the magnetic moment of the sample at the maximum magnetic field amplitude almost at magnetic saturation) versus magnetic field, *H*. Such format of hysteresis loops allows better comparison of microwires with different chemical composition and diameters. The homogeneous axial magnetic field was produced by a long solenoid (about 1 cm in diameter and 12 cm in length). All the measurements were performed at low magnetic field frequencies (100 Hz).

The sample impedance, Z, in extended frequency range has been evaluated using the micro-strip sample holder from the reflection coefficient, S_{11} , obtained using Vector Network Analyzer (VNA), as previously described [23]. Such micro-strip holder with sample has been placed inside a long solenoid generating a homogeneous magnetic field, *H*. The GMI ratio, $\Delta Z/Z$, is obtained from Z(H) dependence as:

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

where H and H_{max} are given and maximum applied fields respectively.

The composites containing both carbon fibers and ferromagnetic glass-coated microwires were manufactured in the INFINITE project (Horizon Europe) at IDEKO's facilities (see Figure 1).



Figure 1. Image of the carbon fiber composite with magnetic microwire inclusions (the vertical lighter fibres) with 5 mm spacing.

The amorphous structure of all the microwires has been



Figure 2. Sketch of the free-space setup.

proved by the X-ray Diffraction (XRD) method. Typically, the crystallization of amorphous microwires was observed at Tann \geq 500 °C [16].

For wireless measurements we used the free space measurement setup (see Figure 2) consisting of two broadband horn antennas (1-17 GHz) fixed to the anechoic chamber and a vector network analyzer, previously employed for the characterization of the composites with magnetic wire inclusions [18,21]. Such setup allows to characterize the composite of $20 \times 20 \text{ cm}^2$.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Previous studies have demonstrated that the magnetostriction coefficient, λs , is primary affected by the composition of the microwires. Vanishing λs –value ($\lambda s \approx 10^{-7}$) is predicted in (Co_{1-x}Fe_x)_{1-y}(Si-B-C)_y amorphous alloys at 0.05 \leq x \leq 0.1 and 0.15 \leq y \leq 0.30 [17] [24]. Therefore, we prepared Co_{64.6}Fe₅B₁₆Si₁₁Cr_{3.4} glass-coated microwires with metallic nucleus diameter, *d*, of 22 and 38 µm, which previously showed high MI effect [17].

For comparison, we also prepared and measured magnetic properties of Fe-rich microwires (Fe_{77.5}B₁₅Si_{7.5}) with d=23 μ m, D=37 μ m and high and positive λs ($\lambda s \approx 40 \times 10^{-6}$).

The hysteresis loops of studied microwires are provided in Figure 3. As observed from Figure 3, both Co-rich microwires show good magnetic softness: a coercivity, H_c , about 16-20 A/m and a magnetic anisotropy field, H_k , about 150 A/m. In contrast, a rectangular hysteresis loops and H_c ≈ 100 A/m are observed for Fe_{77.5}B₁₅Si_{7.5} glass-coated microwires (d=23 µm, D=37 µm) (see Figure 3c).

Figure 3 shows the results on GMI effect of these microwires. As evidenced from Figure 4, both Co-rich microwires present high MI effect (maximum $\Delta Z/Z$ up to 220 % at 100 MHz, see Figures 4 a,b). However, for Fe_{77.5}B₁₅Si_{7.5} microwire the observed MI effect is rather low: up to 2% at the same frequency (100 MHz) (see Figure 4c). Consequently, Co-rich microwires with better MI effect have been selected for the composite preparation.

As reported elsewhere [25] [26], magnetic properties and MI effect of amorphous ferromagnetic microwires are substantially affected by applied stress and by heating. Therefore, the main advantage of utilizing of magnetic microwires inclusions in carbon fiber composites is the possibility for stress and/or temperature monitoring. Very few previous publications have reported attempts to prepare such composites, while Fe-rich microwire inclusions were used and the carbon fiber content was rather low [27].

The expected problem with composites containing conductive carbon fibers is that they can substantially



Figure 3. Hysteresis loops of $Co_{64.6}Fe_{5.0}B_{16.0}Si_{11.0}Cr_{3.4}$ microwires (a,b) with $d\approx 22 \ \mu$ m, $D\approx 24 \ \mu$ m and $d= 38 \ \mu$ m, $D= 43.5 \ \mu$ m respectively and $Fe_{77.5}B_{15}Si_{1.5}$ microwires with $d= 23 \ \mu$ m, $D= 37 \ \mu$ m (c).

interfere with the microwave signal from the magnetic microwire inclusions [27]. Therefore, we propose to apply a low frequency modulating magnetic field to distinguish the microwave signals from magnetic microwires from that originated by conductive carbon fibers, since only the magnetic microwires responds to the modulating field.



Figure 4. $\Delta Z/Z(H)$ dependencies measured in Co_{64.6}Fe_{5.0}B_{16.0}Si_{11.0}Cr_{3.4} microwires with d≈22 µm (a), d=38 µm (b) and in Fe_{77.5}B₁₅Si_{7.5} microwires (c).

In Figure. 5a, the microwave signals are shown (S parameters) measured at 2 GHz. As observed, the signals measured under these conditions are comparable to the noise level. In order to separate the microwave signal from ferromagnetic microwires, we used an external low frequency modulated magnetic field. As shown in Figure 5b, application of such modulated magnetic field (80 Hz) allows a sensitive and stable extraction of the response signal (R and T coefficients) from the ferromagnetic microwires inclusions.

However, the position of carbon fibers in vicinity of magnetic microwires critically affect the signals (S parameters) obtained from such composites.

In Figure 6 are provided the examples of the influence of thin insulating plastic layer (30 μ m thick) between the ordered microwires and carbon fiber composite on S₁₁ parameters



Figure 5. Microwave signals from the composites measured at 2 GHz (a) and the same signal with an external low frequency modulated magnetic field (80 Hz) (b). Line colors: yellow-S₁₁; green-S₂₂, blue and magenta: S₁₂ and S₂₁ respectively.



Figure 6. Effect of insulating plastic layer between the microwires and Carbon fibre composite on S_{11} parameter. Measurements with plastic layer (a) and without plastic layer (b).

As observed, the presence of even such thin insulating layer allows affecting substantially the amplitude of the S_{11} parameter signal originated by magnetic microwires.

The aforementioned examples provide the routes for development of the composites made from the carbon fibers and magnetically soft amorphous glass-coated microwires inclusions.

The key results are that use of a low frequency modulating magnetic field allows to distinguish the microwave signals originated by ferromagnetic microwires inclusions from that generated by the carbon fibers. However, the position of carbon fibers in vicinity of magnetic microwires critically affect the signals obtained from such composites.

IV. CONCLUSIONS

We have explored the feasibility of developing composites containing carbon fibers and glass-coated magnetic microwires inclusions using the free space microwave spectroscopy aligned with the requirements of carbon composites. For the preparation of such composites, we selected Co-rich microwires with better magnetic softness and higher magnetoimpedance effect. We experimentally demonstrated that the application of low frequency magnetic field allows to distinguish the microwave signals originated by ferromagnetic microwires inclusions from the signal generated by the carbon fibers. However, the location of carbon fibers near magnetic microwires has a critical effect on the signals (parameter S) obtained from such composites.

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REFERENCES

- P. Corte-Leon et al., "Magnetic Microwires with Unique Combination of Magnetic Properties Suitable for Various Magnetic Sensor Applications", Sensors, vol. 20, p. 7203, 2020.
- [2] J. Durand, "Magnetic Properties of Metallic Glasses" in Topics in Applied Physics, vol. 53, Glassy Metals II. Atomic Structure and Dynamics, Electronic Structure, Magnetic Properties, Editors: H. Beck and H.-J. Giintherodt, Springer-Verlag, Berlin Heidelberg New York Tokyo, pp. 343-386, 1983
- [3] 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, 1977.
- [4] V. Zhukova et al., "Correlation between magnetic and mechanical properties of devitrified glass-coated

Fe71.8Cu1Nb3.1Si15B9.1 microwires", J. Magn. Magn. Mater., vol. 249, pp. 79–84, 2002.

- [5] A. Zhukov et al., "Giant magnetoimpedance in rapidly quenched materials", J. Alloys Compound., vol. 814, pp. 152225, 2020.
- [6] V. Zhukova et al., "Electronic Surveillance and Security Applications of Magnetic Microwires", Chemosensors, vol. 9, p.100, 2021.
- [7] K. Mohri, T. Uchiyama, L. P. Shen, C. M. Cai, and L. V. Panina, "Amorphous wire and CMOS IC-based sensitive micro-magnetic sensors (MI sensor and SI sensor) for intelligent measurements and controls", J. Magn. Magn. Mater., vol. 249, pp. 351-356, 2001.
- [8] 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.
- [9] Y. Honkura, "Development of amorphous wire type MI sensors for automobile use", J. Magn. Magn. Mater., vol. 249, pp. 375-381, 2002.
- [10] A. Zhukov et al., "Magnetoelastic sensor of level of the liquid based on magnetoelastic properties of Co-rich microwires", Sens. Actuat. A Phys., vol. 81, no. 1-3, pp.129-133, 2000.
- [11] V. Zhukova et al., "Development of Magnetically Soft Amorphous Microwires for Technological Applications", Chemosensors, vol. 10, p. 26, 2022
- [12] L. Ding, S. Saez, C. Dolabdjian, L. G. C. Melo, A. Yelon, and D. Ménard, "Development of a high sensitivity Giant Magneto-Impedance magnetometer: comparison with a commercial Flux-Gate", IEEE Sensors, vol. 9, no.2, pp. 159-168, 2009.
- [13] D. Kozejova et al., "Biomedical applications of glass-coated microwires", J. Magn. Magn. Mater., vol. 470, pp. 2-5, 2019.
- [14] D. Makhnovskiy, N Fry, and A. Zhukov, "On different tag reader architectures for bistable microwires", Sens. Actuat. A Phys., vol. 166, pp. 133-140, 2011.
- [15] S. Gudoshnikov et al., "Evaluation of use of magnetically bistable microwires for magnetic labels", Phys. Stat. Sol. (a), vol. 208, no. 3, pp. 526–529, 2011.
- [16] 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.
- [17] A. Zhukov et al., "Advanced functional magnetic microwires for technological applications", J. Phys. D: Appl. Phys., vol. 55, p. 253003, 2022.
- [18] 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.
- [19] F. Qin and H.X. Peng, "Ferromagnetic microwires enabled multifunctional composite materials", Prog. Mater. Sci., vol. 58, no. 2, pp.183-259, 2013.
- [20] M. Churyukanova et al., Non-contact method for stress monitoring based on stress dependence of magnetic properties of Fe-based microwires", J. Alloys Compd., 748, no.5, pp. 199-205, 2018.
- [21] 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
- [22] L. J. Nelson, "Smart piezoelectric Fibre composites", Mat. Sci. and Tech., vol. 18, pp.1245-1256, 2002.
- [23] A. Zhukov, M. Ipatov, P. Corte-Leon, J. M. Blanco, L. González-Legarreta, and V. Zhukova, "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

- [24] M. Churyukanova et al., "Magnetostriction investigation of soft magnetic microwires", Phys. Stat. Sol. A, vol. 213, no. 2, pp. 363-367, 2016.
- [25] P. Corte-Leon et al., "Stress dependence of the magnetic properties of glass-coated amorphous microwires" J. Alloys Compound, vol. 789, pp. 201-208, 2019, doi: 10.1016/j.jallcom.2019.03.044
- [26] P. Corte -Leon et al., "Effect of temperature on magnetic properties and magnetoimpedance effect in Fe -rich microwires", J. Alloys Compound. vol. 946, 169419, 2023, doi: https://doi.org/10.1016/j.jallcom.2023.169419
- [27] Y. Luo et al., "Left-handed metacomposites containing carbon fibers and ferromagnetic microwires", AIP Advances vol. 7, 056110, 2017.