MICROWAVE TUNABLE COMPOSITES WITH MELT-EXTRACTED MAGNETIC MICROWIRES

F.X. Qin[1,2*], H.X. Peng[2], C. Brosseau[1], Huan Wang[3], Jianfei Sun[3]
[1]Lab-STICC, Université de Bretagne Occidentale
CS 93837, 6 Avenue Le Gorgeu, 29238 Brest Cedex 3, France
[2] Advanced Composite Center for Innovation and Science, Department of Aerospace Engineering, University of Bristol, University Walk, Bristol, BS8 1TR, UK
[3] School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, China
Corresponding author (faxiang.qin@gmail.com)

Keywords: *microwire composite, ferromagnetic microwires, structural health monitoring, microwave, permittivity*

1 General Introduction

A key to the realization of effective microwave tunable devices, among others, is to develop adaptive materials with superior adjustability of electromagnetic parameters. Since there are two fundamental electromagnetic parameters, namely, permittivity and permeability, the tunable property can then be readily approached by manipulating (i) permittivity directly via the electric field, such as in ferroelectrics and conducting polymer [1, 2], or (ii) permeability via magnetic field, such as in ferromagnetic materials [3], or (iii) permittivity (permeability) indirectly by magnetic field (electric field) such as in multiferroics [4].

A unique class of composite materials is considered here to realize such tunable properties: ferromagnetic microwires with excellent soft magnetic properties and giant magnetoimpedance effect [5,6] were introduced into a polymer matrix material so that an indirect control of permittivity can be tuned by the magnetic field. The principle is that the magnetic field can induce significant variation of the impedance via the skin effect at high frequency. The current distribution along the wire then changes accordingly and induces the dipolar polarization [7]. There have been a few relevant studies on this subject in the open literature. Panina et al. [8] and Acher et al. [9] used the microwire arrays to realize the left-hand materials and tunable properties in the presence of magnetic field. Starostenko et al. [10] gave an in-depth discussion of the magnetic bias effect on the permittivity for random wire composites and proved the feasibility of using the wire composites to control attenuation. Di et al. [11], Marin et al. [12] and Zhang et al. [13] have demonstrated the perspective of microwire composites as microwave absorbers.

In addition, our previous studies have shown that this kind of composite prove to be multifunctional for a wide range of potential engineering applications such as sensing and structural health monitoring (see e.g. [14-16]). The aim of this paper is to utilize instrumentation [17] dedicated to precise in-situ measurements of electromagnetic parameters to characterize the microwire composites over a broad frequency range and the field effect on the microwave behaviors of the wire composites for potential structural health monitoring (SHM) application.

2 Experimental

Soft magnetic microwires with composition of $Co_{68.15}Fe_{4.35}Si_{12.25}B_{15.25}$ and diameter of about 40μm were fabricated by a precision melt extraction facility [18]. The magnetization curve of wires was measured by using a conventional induction method. They possess a low coercivity (see Fig. 1a) and vanishing magnetostriction (~ -10^{-7}), which make them suitable for sensing applications [19, 20]. Also, an INSTRON machine with a load cell of 1kN was used to obtain the tensile stress-strain curves of single wires. These wires exhibits excellent mechanical properties (see Fig.1b), making them suitable as multifunctional fibres to be introduced into polymer matrices [21].

Fig. 1 HERE

Two types of dilute composites were prepared with different length of wires. For short 5mm wires, 50 mg of wires are first randomly dispersed in the silicon rubber after thorough mixing. Then they are cast into a mold of the size of 70 × 10 × 1.8 mm^3 and cured in the room temperature for 24 hours (Fig.2a). The resultant composite has a microwire concentration of 3 wt.%. The other type of composite is made from 70 mm microwires by aligning the microwires in a periodical manner with fixed wire spacing of 0.77 mm into the silicon rubber matrix sheets, which were bonded together using silicone resin. The resultant sample size remains the same as the short wire one (Fig.2b).

Fig. 2 HERE

Complex effective relative (omitted hereafter) magnetic permeability and relative (omitted hereafter) permittivity spectra were measured at room temperature using a modified microwave frequency-domain spectroscopy with/without a magnetic bias applied along the wire direction at a frequency range of 300 MHz-6 GHz, and then extracted by a built-in utility program, whereby the gap between the sample surface and microstrip is also taken into account. Briefly, our experiments consist of measuring the transmission and reflection coefficients of an asymmetric microstrip transmission line containing the sample in the presence of a magnetic bias as schematically shown in Fig.2. The electromagnetic measurement was carried out with the wave vector of the electromagnetic field perpendicular to the wires. The quasi-TEM transverse electromagnetic mode, which is the only mode that propagates in the structure, makes the analysis of the complex transmission and reflection coefficients relatively simple. Using the Nicolson-Ross procedure for the transformation of the load impedance by a transmission line, complex permittivity and permeability are determined by the transmission S21 and reflection S11 parameters. A vector network analyzer (Agilent, model H8753ES) with SOLT calibration (short, open, load and thru) was used to measure the S parameters of the cell containing the sample under test within the frequency range between 300 MHz and 5 GHz. Further details of the instrumentation were discussed elsewhere [17, 22].

3 Results and Discussion

Figure 3 displays the complex permittivity spectra in the presence of varying magnetic bias. At zero magnetic field, the composite simply presents a single relaxation line. When the external field of 100 *Oe* is applied, the permittivity is increased but its frequency dispersion remains unchanged. With further increase of magnetic field to 500 *Oe*, a dielectric absorption maximum is seen at 4.1 GHz. This maximum shifts to higher frequency of 4.6 and 4.7 GHz as the magnetic field is increased to 1000 and 1500 *Oe*, respectively, while the absorption linewidth is increased with the value of 1 GHz. Also, there appears an obvious anomalous dispersion of ε' from 500 *Oe* upwards. Quantitatively, for H_{dc} = 500 *Oe*, the anomalous dispersion spans from 3.5 to 4.3 GHz, and the maximum of dielectric absorption falls in this region. A so-called frequency response effect [23] can then be inferred herein and will be discussed in the context of microwave absorption in the next section. Also of particular interest is that the application of the magnetic field above 1000 *Oe* results in negative ε' at the frequency band from 4.7 GHz onwards.

Fig. 3 HERE

The magnetic bias dependence of permittivity is shown in Fig.4 at two feature frequencies, 2.5 and 4.5 GHz, the latter is in the resonance region. At a lower frequency band, the variation of permittivity $\Delta\varepsilon$ is small. In contrast, a rather pronounced change of permittivity is observed for higher frequencies. It can be also seen that, at 4.5 GHz, the application of magnetic field actually gives rise to a larger ε'' than ε', which is not seen in the lower frequency band.

Fig. 4 HERE

In the case of composite with longer wires in periodical manner, the permittivity spectra obtained is illustrated in Fig. 5. Two separate absorption maxima were shown at 1-2 GHz and 4-5 GHz, respectively. The bias dependency of permittivity, in the same manner as the composite with random wires, can be identified clearly in the resonance region; the second absorption maximum shows a significant rise with the magnetic field and becomes narrower [24,25].

Fig. 5 HERE

Now let us discuss these findings. For the composites containing short wire pieces, the microwires respond to the electromagnetic wave as electric dipoles. For the same matrix, the dipole resonance is defined by the length according to $f_{res} = c / 2l\sqrt{\varepsilon_m}$ [7], where c is light speed, l is the wire length, ε_m is the matrix permittivity. When the length is 5 mm, the resonance frequency is about 15 GHz, which is out of the measurement range in the present work. This explains the absence of the absorption maximum in the obtained spectra. With the application of magnetic field, the current induced resonance of circumferential permeability, *i.e*, magnetoimpedance resonance comes into play. As there is a strong dependence of impedance on the external magnetic field (GMI effect), the applied magnetic field can then vary the current distribution in the microwires through the skin effect, which is pronounced in the gigahertz frequency. This gives rise to a significant change of dielectric response (permittivity). As the ferromagnetic resonance frequency is below the dielectric resonance frequency, the skin effect dominates the absorption and increases the absorption linewidth [26]. The blueshift of resonance frequency ω_{FMR} with the field H_{dc} can be accounted by the Kittel equation [27]: $\omega_{FMR} = \mu_0\gamma\sqrt{(H_{dc} + H_k + M_s)(H_{dc} + H_k)}$, where μ_0 is the vacuum permeability, γ is the gyromagnetic parameter, M_s is the saturation magnetization, H_{dc} denotes the applied magnetic field, H_k is the anisotropy field.

The microwave behavior of interest of the prepared composite lies partly in its anomalous dispersion, which is often accompanied with a so-called frequency response entailing an opposite frequency dependence of ε'' and ε' . This is particularly useful for microwave absorption and indicates that the microwire composites could be exploited for designing a microwave absorber with sufficient absorption bandwidth. In addition, the anomalous dispersion is often associated with the negative permittivity [7], making the wire composite a metamaterial. Thus, a variety of unusual properties can be engineered to meet specific applications such as high-performance frequency selective surface (FSS).

The discrepancy of permittivity change with the magnetic field for different frequency can be attributed to the fact that, as the frequency approaches the ferromagnetic resonance frequency, the application of magnetic bias could achieve the minimal skin depth and hence the maximal variation of dielectric response. It should be noted that the magnetic bias is effective in tuning the permittivity, but has no visible effect on permeability (not shown here), in that the composite proves to be so dilute that the permeability is close to unity at such frequency range. For random wire composites, the wires perpendicular to the microwave magnetic field makes no contribution, whereas the parallel ones has no response to the field at gigahertz frequency range. Thus, the realization of such microwave tunable composites is focused on the dielectric response. And the sensitivity of permittivity to the frequency and magnetic field in the prepared composite make it very practical from the application point of view.

Finally, let us examine the composite with periodical wires of 70 mm. Comparing to the short wire composite, this sample has a much lower computed resonance frequency of 1.1 GHz. This again is confirmed in the permittivity spectra, the variation of this value to the experimental value can be attributed to the possible interface defects in the composite. As the relationship of dielectric resonance frequency and ferromagnetic resonance frequency reverses in relative to the previous type of composite, the linewidth also presents opposite trend with increasing field. In summary, the wire length plays an important role in defining the microwave response of the wire composites.

4. Conclusions

We have performed a detailed study on the rubber matrix composites containing amorphous CoFeSiB wires. The results obtained here show that there is a strong dependence of permittivity spectra on the external magnetic bias. For wire composites containing short wires of 5mm, with increasing magnetic bias, the dielectric constant increases and reaches the maximum response to the magnetic bias (the so-called tunability) in the vicinity of maximum absorption induced by the magnetoimpedance resonance. It is also shown that both absorption resonance frequency and linewdith can be conveniently tuned by the magnetic bias. For composites containing long wires (70 mm), the

interference of dipole resonance and magnetoimpedance resonance results in multi-resonance permittivity spectra; at the second resonance associated with magnetoimpedance a narrower linewidth is observed with increasing magnetic fields. All these results provide useful insights to the selection and optimization of these materials for design of microwave devices based on these materials. It is worth mentioning that, directly linked to the SHM application, the effect of stress on the composite containing these microwires with excellent stress-impedance sensitivity is in progress. Different from the case under magnetic field, the microwire composite under stress involves the shape change of the wires and the variation of non-uniform interfacial properties. More significant tunable effects are expected and will be reported elsewhere.

Acknowledgements
FXQ would like to thank Mr. Gregory Mignot and Christopher Prunier of Lab-STICC, UBO for their help with the microwave characterization as well as the financial support provided by Conseil Général du Finistère, France.

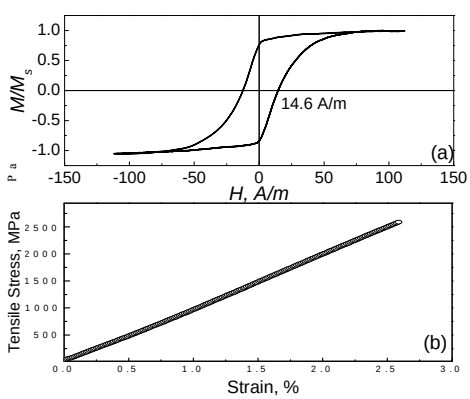

Fig. 1. (a) Magnetization curve of the CoFeSiB microwire. (b) Stress-strain curve of the microwire.

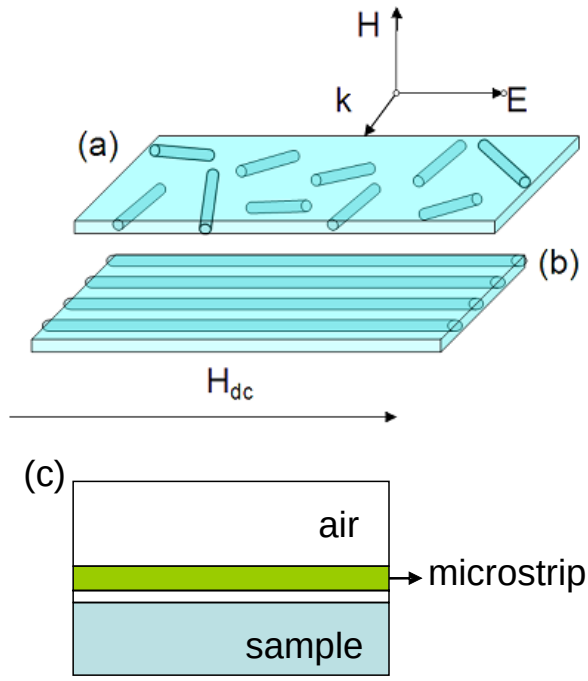

Fig. 2. Schematics of microwire composites with (a) 5 mm long wire in random patterns and (b) 70 mm long wire in periodical manner with fixed wire spacing of 0.77 mm, (c) Schematic cross-sectional view of the measuring cell.

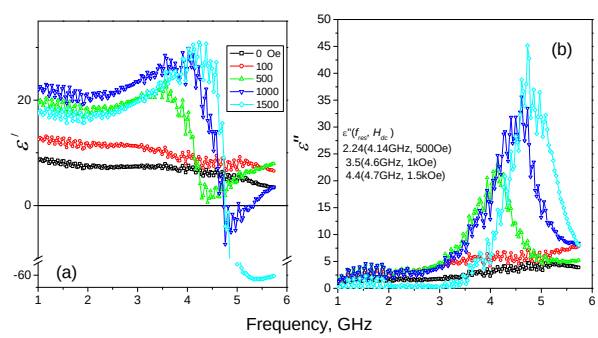

Fig. 3. Frequency dependence of (a) real part of permittivity and (b) imaginary part in the presence of varying magnetic bias for composites with 5 mm short wires.

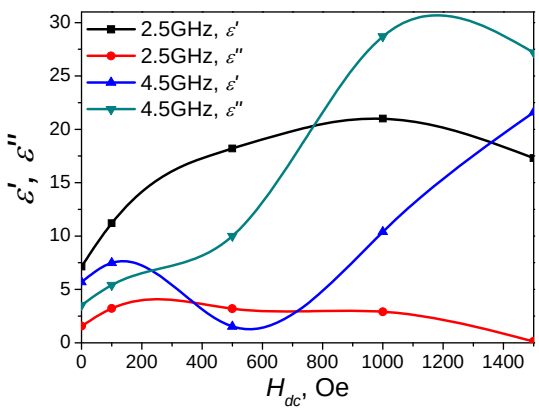

Fig. 4. Magnetic field dependence of complex permittivity at 2.5 and 4.5 GHz.

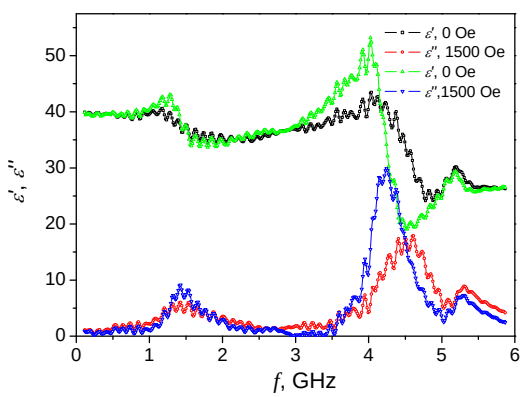

Fig. 5. Frequency dependence of complex permittivity for composites with 70 mm wires at zero magnetic field and 1500 *Oe*.

References

[1] P. Teo, K.A. Jose, Y.B. Gan, *et al*. "Beam scanning of array using ferroelectric phase shifters". *Electronics Letters*, Vol.36, No.19, pp. 1624-1626, 2000.

[2] R. Zhang, A. Barnes, K. Lee Ford, *et al.* "A new microwave 'smart window' based on a poly(3,4-ethylenedioxythiophene) composite". *Journal of Materials Chemistry*, Vol.13, No.1, pp. 16-20, 2003.

[3] E. Salahun, G. Tanne, and P. Queffelec. "Enhancement of design parameters for tunable ferromagnetic composite-based microwave devices: application to filtering devices. " in Microwave Symposium Digest, 2004 IEEE MTT-S International. 2004.

[4] V. Castel and C. Brosseau. "Magnetic field dependence of the effective permittivity in BaTiO3 /Ni nanocomposites observed via microwave spectroscopy". *Applied Physics Letter,*. Vol.92, No.23, pp. 233110-233110-3, 2008.

[5] M.-H. Phan and H.-X. Peng. "Giant magnetoimpedance materials: Fundamentals and applications". *Progress in Materials Science.* Vol.53, No.2: pp. 323--420, 2008.

[6] M. Knobel, M. Vazquez, and L. Kraus. *Giant magnetoimpedance*. Vol. 15. 2003: K.H. Buschow.

[7] D.P. Makhnovskiy, L.V. Panina, C. Garcia, *et al.* "Experimental demonstration of tunable scattering spectra at microwave frequencies in composite media containing CoFeCrSiB glass-coated amorphous ferromagnetic wires and comparison with theory". *Physical Review B: Condensed Matter*, Vol.74, No.6, pp. 064205, 2006.

[8] L.V. Panina, M. Ipatov, V. Zhukova, *et al.* "Magnetic field effects in artificial dielectrics with arrays of magnetic wires at microwaves". *Journal of Applied Physics*, Vol.109, No.5, pp. 053901, 2011.

[9] O. Acher, M. Ledieu, A.L. Adenot, *et al.* "Microwave properties of diluted composites made of magnetic wires with giant magneto-impedance effect". *Magnetics, IEEE Transactions on*, Vol.39, No.5, pp. 3085-3090, 2003.

[10] S.N. Starostenko and K.N. Rozanov. "Microwave screen with magnetically controlled attenuation". *Progress In Electromagnetics Research*, Vol.99, pp. 405-426, 2009.

[11] Y.J. Di, J.J. Jiang, G. Du, *et al.* "Magnetic and microwave properties of glass-coated amorphous ferromagnetic microwires". *Trans. Nonferrous Met. Soc. China,* Vol.17, No.6: pp. 1352-1357, 2007.

[12] P. Marin, D. Cortina, and A. Hernando. "Electromagnetic Wave Absorbing Material Based on Magnetic Microwires". *Magnetics, IEEE Transactions on*, Vol.44, No.11, pp. 3934-3937, 2008.

[13] Z. Zhang, C. Wang, Y. Zhang, *et al.* "Microwave absorbing properties of composites filled with glass-coated Fe69Co10Si8B13 amorphous microwire". *Materials Science and Engineering: B*, Vol.175, No.3, pp. 233-237, 2010.

[14] F.X. Qin, N. Pankratov, H.X. Peng, *et al.* "Novel magnetic microwires-embedded composites for structural health monitoring applications". *Journal of Applied Physics*. Vol.107, No.9, pp. 09A314-3, 2010.

[15] F. Qin, H.X. Peng, C. Prunier, *et al.* "Mechanical--electromagnetic coupling of microwire polymer composites at microwave frequencies". *Applied Physics Letters*, Vol.97, No.15, pp. 153502-3, 2010.

[16] H.X. Peng, F.X. Qin, M.H. Phan, *et al.* "Co-based magnetic microwire and field-tunable multifunctional macro-composites". *Journal of Non-Crystalline Solid,*. Vol.355, No.24-27: pp. 1380-1386, 2009.

[17] C. Brosseau and P. Talbot. "Instrumentation for microwave frequency-domain spectroscopy of filled

polymers under uniaxial tension". *Measurement Science and Technology*, Vol.16, No.9: pp. 1823, 2005.

[18] H. Wang, D. Xing, X. Wang, *et al.* "Fabrication and Characterization of Melt-Extracted Co-Based Amorphous Wires". *Metallurgical and Materials Transactions A*, Vol.42, No.4, pp. 1103-1108.

[19] V. Zhukova, M. Ipatov, and A. Zhukov. "Thin Magnetically Soft Wires for Magnetic Microsensors". *Sensors,* Vol.9, No.11, pp. 9216-9240, 2009.

[20] L.V. Panina, D.P. Makhnovskiy, and K. Mohri, "Magnetoimpedance in amorphous wires and multifunctional applications: from sensors to tunable artificial microwave materials". *Journal Of Magnetism And Magnetic Materials*, Vol.272-276, No.Part 2, pp. 1452--1459, 2004.

[21] F. Qin, H.-X. Peng, J. Tang, *et al.* "Ferromagnetic microwires enabled polymer composites for sensing applications". *Composites Part A: Applied Science and Manufacturing*, Vol.41, pp. 1823 -1828, 2010.

[22] E. Salahun, P. Queffelec, M. Le Floc'h, *et al.* "A broadband permeameter for "in situ" measurements of rectangular samples". *Magnetics, IEEE Transactions on,* Vol.37, No.4, pp. 2743-2745, 2001.

[23] T.C. Zou, N.Q. Zhao, and C. Shi. "Design of activiated carbon fibres / epoxy resin composites". *Journal of Functional Materials and Devices.* Vol.13, No.1, pp. 54-58, 2007.

[24] M.H. Phan, H. X Peng S.C. Yu, M. Vazquez, Optimized giant magnetoimpedance effect in amorphous and nanocrystalline materials Vol. 99(8), *JOURNAL OF APPLIED PHYSICS* 08C505-1-4, 2006.

[25] M.H. Phan, H. X Peng S.C. Yu, W.R. Wisnom, Large enhancement of GMI effect in polymer composites containing Co-based ferromagnetic microwires, *JOURNAL OF MAGNETISM AND MAGNETIC MATERIALS*, Vol.316(2), pp: E253-E256, 2007.

[26] S.N. Starostenko, K.N. Rozanov, and A.V. Osipov. "Microwave properties of composites with glass coated amorphous magnetic microwires". *Journal of Magnetism and Magnetic Materials*, Vol.298, No.1, pp. 56-64, 2006.

[27] H. García-Miquel, M.J. Esbri, J.M. Andres, *et al.* "Power absorption and ferromagnetic resonance in Co-rich metallic glasses". *Magnetics, IEEE Transactions on*, Vol.37, No.1, pp. 561-564, 2001.