

**In situ microwave characterization of microwire composites under mechanical stress**

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We present results of an experimental characterization of the dielectric properties and microwave absorption of rubber composite samples containing Fe0.4Co68.7Ni1B13Si11Mo2.3 amorphous microwires which are submitted to a low uniaxial tension. Measurements of the dielectric loss and microwave absorption as a function of strain over the frequency range of 300 MHz-6 GHz reveal that the uniaxial elongation randomly breaks wires at about 2.8% strain and this has for effect to decrease the loss factor for larger strain. Two possible mechanisms are identified to account for our observations, namely, the stress and shape effects. The ability to control this stretch breaking phenomenon will be instrumental to developing stress tunable microwire composites for sensing applications. © 2011 American Institute of Physics. [doi:10.1063/1.3668109]

Multifunctional micro- and nanostructures exploiting microwave relaxation and/or resonance phenomena are important in many technological applications from reconfigurable microwave devices1 to microwave non-destructive testing.2 Whether generated by the application of a magnetic field, a mechanical deformation, or even a temperature gradient, questions surrounding the coupling between the structural, electromagnetic, and mechanical properties of such materials continue to pose many theoretical and experimental challenges.

The correlation between the materials parameters (effective permittivity and magnetic permeability) and stress in filled polymers, e.g., carbon black filled cross-linked rubber and plasto-ferrites, has been experimentally established.3 For example, microwave measurements indicated large decreases in the real and imaginary parts of the effective permittivity of axially elongated samples.4,5 This set of experiments illustrates that the microwave permittivity data scale as a power law in frequency, where the exponent is extremely sensitive to stress. In addition, the effective permittivity measurements under stress can be explained in terms of the Gaussian molecular network model in the limit of low stress.6

From a different perspective, we recently considered a class of multifunctional microwire polymer composites for structural health monitoring.7,8 While the magnetic field response of these materials has been intensively studied,9,10 definite models for the stress effect remain debatable. It is well established11 that the application of an external stress modifies the magnetic anisotropy and domain structure of the wires. This has the effect to change the ferromagnetic resonance and to modify the eddy current loss, which in turn will eventually affect the effective permittivity. We have also observed that the permittivity spectra of such composites can be modulated by the application of a mechanical constraint.6,11,12

Here, we focus on the dielectric response of microwire polymer composites and approach this problem through a combination of well-controlled experiments of microwave characterization under mechanical stress. First, we perform tests on microwire filled rubber composites which we regard as a model material. Our experiments also show that the uniaxial elongation randomly breaks wires at about 2.8% strain, and this has the effect to decrease the loss factor for larger strains. This stretch breaking phenomenon suggests the possibility of controlling the dielectric properties and microwave absorption of microwire polymer composites.

Fe0.4Co68.7Ni1B13Si11Mo2.3 glass-coated magnetic microwires having diameter of 12.8 μm (MFTI, Moldova) and silicone rubber were used for the fabrication of composite materials. The wires were chosen due to their strong magnetoimpedance properties making them suitable for designing complex composite media for microwave-frequency operations.7,8 Twelve continuous microwires with length of 70 mm (wire length is comparable with the wavelength of the exciting wave) were embedded in a parallel manner with lattice constant of 1 mm into the silicone rubber matrix. The resultant composite has a dimension of 70 mm × 13 mm × 1.8 mm. The volume fraction of inclusions is calculated by the ratio of the wires volume to the composite sample volume.

We measured the room temperature effective complex (relative) permittivity $\varepsilon$ and magnetic permeability $\mu$ spectra of these samples which are submitted to a tensile strain up to 4.5%, using a home-built experimental setup over the 0.3–6 GHz frequency range. Our experiments consist of measuring the transmission and reflection coefficients of an asymmetric microstrip transmission line containing the sample during elongation.13 The electromagnetic measurement was carried out with a 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. A vector network analyzer (Agilent, model H8753ES) with SOLT calibration is used to measure the S parameters of the cell containing the sample under test. Using the Nicolson-Ross procedure for the transformation of the load impedance by a transmission line, the strain of the material subjected to a uniaxial tension is defined as $\lambda = (\ell - \ell_0) / \ell_0$, where $\ell$ and $\ell_0$ denote the actual and initial lengths of a sample, respectively. The accuracy, reproducibility, and analysis of our technique were tested by measurements on a number of heterogeneous soft materials.

As was discussed in Ref. 13, these measurements require an accurate measure of the residual air-gap between the sample and the transmission line walls. Additional tests of the tensile properties of the microwires were carried out by employing the Instron 1466 testing machine equipped with a load cell of 1 kN.

In Fig. 1, the real and imaginary parts of the effective complex permittivity spectra are presented with respect to tensile strain up to 4.5%. The stress-strain curve of a single wire is given in the inset. It is important to note that the linear behavior is characteristic of a typical brittle material without yielding region. Two important features are noted here. (1) Significant variation of $\varepsilon'$ for the stressed samples compared to the unstressed state is visible for $\lambda = 1.4\%$ and 4.5% at frequencies larger than 4 GHz. The stress effect becomes insignificant at low frequency. The bandwidth of anomalous dispersion of $\varepsilon'$ remains independent of stress, i.e., typically between 3.5 and 4.4 GHz. (2) The maximum absorption corresponding to the peak of the $\varepsilon''$ spectrum increases as strain increased from 0 to 2.8% and shifted to a higher frequency. With a further increase of strain, the absorption maximum decreases and shifts to a lower frequency. This is clearly seen in Fig. 2, where the stress dependence of the permittivity is plotted for frequencies of 4.1 and 4.7 GHz. The former is close to the absorption maximum while the latter is far away from the peak.

As the frequency is increased from 4.1 to 4.7 GHz, both $\varepsilon'$ are decreased. It is worth noting that the evolution of the dielectric loss $\varepsilon''$ with strain is very similar for these two frequencies. We further include the stress dependence of the loss factor in the inset of Fig. 2. The impact of stress remains identical to that shown on $\varepsilon''$. It should be noted that a strain larger than 7% annihilates the strain effect.

We now turn our attention to microwave absorption coefficient defined as $A = 1 - |S_{11}|^2 - |S_{21}|^2$. Comparison of the dielectric loss factor (Fig. 3(a)) and the absorption spectra (Fig. 3(b)) is meaningful for the various values of the frequency range explored. It is noted that this result differs from the observations reported by Marín et al.,16 who found that the absorption of microwire composites for a large wire concentration (0.5 vol. %) is dominated by magnetic loss and associated with the ferromagnetic resonance (FMR) of the wires. These observations do not contradict with the current results since we are dealing with more dilute composite systems, i.e., the wire volume fraction is close to 0.005 vol. %. The FMR
frequency $f_c$ of the planar wire composites is determined by
\[
f_c = \gamma \sqrt{\frac{4\pi M_s (H_s + H_0)}{n}} ,
\]
where $\gamma$ is the gyromagnetic factor, $M_s$ is the saturation magnetization, $H_s$ is the anisotropy field, and $H_0 \propto i M_s (a/l)^2$ is the dipole-dipole interaction field for all neighboring wires of diameter $2a$, length $l$, and number $l$. It is instructive to examine the wire content dependence of $f_{MR}$. Unless sufficient wires are embedded into the matrix, ensuring a large contribution of the anisotropy and dipole-dipole interaction fields, one cannot expect a resonance induced by the magnetic loss in the range of GHz frequencies. Although the absorption performance is smaller for dilute composites, low losses are desirable for the design of ferromagnetic tunable composites and metamaterials based on ferromagnetic microwires.

From now on, we focus our discussion on several possibilities that may shed light on our understanding of the origin of the observed difference in the stress dependence of the permittivity spectra. The wires are brittle materials with very low ductility (as seen from the inset of Fig. 1). At relatively small strain before fracture arises, the stress effect on the dielectric response of the composite can be described by $\varepsilon = \varepsilon_m + 4\pi p \langle x \rangle$, where $\varepsilon_m$ denotes the permittivity of the matrix, $p$ is the wire concentration, and $\langle x \rangle$ the effective (averaged) polarisability. When the strain exceeds 2.8%, breakdown of some wires occurs. Due to nonuniform interfacial conditions, each single wire experiences a different stress state resulting in partial but not total fracture of all wires, as shown in Fig. 4(b). Following Makhnovskiy et al., $\langle x \rangle$ can be written as
\[
\langle x \rangle = \frac{1}{2\pi n (l/a) K} \left( \frac{\pi}{2} \tan(K/2) - 1 \right),
\]
with $K = k \left( 1 + \frac{\varepsilon_m}{\varepsilon_\infty} \right)^{1/2}$, and $k = \omega \sqrt{\varepsilon_m / c}$, where $\omega$ is the angular frequency, $c$ is the velocity of the electromagnetic wave in vacuum, and $\varepsilon_\infty$ is the wire surface impedance. Consequently, the wire length plays an important role in determining the effective permittivity. A recent study by Ipatov et al. showed that the reduction of the wire length can significantly decrease the permittivity and lead to a blueshift of the resonance frequency. This explains why the present results differ from those reported previously. Figure 4(a) shows an image of the elongated but not fractured wires for low strain, i.e., $\lambda = 1.4\%$. Thus, the observation here of broken wires as strain is over 2.8% (see Fig. 4(b)) is consistent with the decrease of the dielectric loss with stress of composites containing a smaller number of strained continuous wires. Some of the possible mechanisms which are involved to explain this observation are the stress effect, i.e., the stress changes the current distribution in the wires and induces a higher dielectric loss and the shape effect, i.e., wires can be broken in shorter pieces and the anisotropy field of the wire becomes non-uniform, resulting in the reduction and the broadening of the resonance bandwidth. In addition, it is noted that the relevant relaxation mechanisms attributed to dipolar relaxation and Maxwell-Wagner-Sillars interfacial polarization due to the boundaries separating microwires are expected to change due to wire fracture. Thus, the local properties of wires and the composite mesostructure are significantly altered. From an engineering perspective, we note that our work and recent results by other groups indicate that stretch-broken fibres systems achieve the formability of discontinuous reinforced systems without large performance knockdowns of continuous fibre prepreg which have restricted its use in automated manufacturing processes.

In summary, we have performed a detailed study of the stress effect on the microwave properties of magnetic microwire polymer composites over a broad frequency range. The study of the permittivity spectrum demonstrates a nonlinear tunable stress effect over a broad range of strain. The analyses of permittivity and the strain sensitivity afford to identify two plausible mechanisms arising when a fraction of wires are broken, namely, the stress and shape effects. The effect of wire concentration has also been highlighted. This study has significant implications for research in the emerging field of stress tunable microwire composites for sensing applications.

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