Stress tunable properties of ferromagnetic microwires and their multifunctional composites

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We report the results of a systematic study on stress tunable absorption of glass-coated amorphous $Co_{68,7}Fe_4Ni_1B_{13}Si_{11}Mo_{2,3}$ microwires and their composites. The magnetic microwires possess good stress-impedance properties and yield a stress dependence of absorption at gigahertz frequencies. The stress compensates the reverse effect of magnetic field on absorption. There exist strong stress dependences of the effective permittivity and transmission parameters. Composite failure due to the wire damage results in a dramatic change of the sign and magnitude of effective permittivity. The double peak is identified in the stress dependence of field tunability, in contrast to the single peak for the magnetic field tunability. All these results indicate that the present composites are very promising for detecting the ambient stress levels and interrogating the structural integrity. \bigcirc 2011 American Institute of Physics. [doi:10.1063/1.3535553]

Much interest has been aroused in the study of engineered multiphase composites containing microscale or nanoscale fillers in terms of their electromagnetic properties for microwave applications such as fiber reinforced composite structures,¹ tunable microwave devices,² remote interrogated sensors,³ etc. A key question of achieving effective design and application of these composites is to understand how the wave transport properties are modified as it interacts with the composite material in the presence of external stimuli such as temperature and/or stress. In the latter case, to decode the effect of stress on the polarization and magnetization mechanism and the way to control them are the basis of developing multifunctional composites for domestic and industrial applications. Especially, we focus on the dielectric response induced by the external magnetic field or stress, which are often associated with the relaxation and resonance characteristics in the spectra.⁴

Most recently, a smart microwire polymer composite has been devised to realize multifunctionalities for a spectrum of applications such as structural health monitoring. The magnetic field effect on the effective permittivity has been systematically presented in previous reports.⁵ As a step forward, the present study targets the stress effect. The rationale is briefly described as follows. For a composite containing ferromagnetic wires exhibiting giant magnetoimpedance effects at microwave frequencies,⁵ the effective permittivity may depend on a dc magnetic field via the corresponding dependence of the surface impedance. The surface impedance can also be changed by applying an external stress which modifies the magnetic anisotropy and domain structure in these wires. Thus, the effective

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permittivity may also depend on the external stress or strain. Corresponding to the stress tunable theory proposed by Panina *et al.*,² we focus on the technological aspects of developing multifunctional composites for structural interrogation applications.

The glass-coated amorphous microwires of Co68.7Fe4 $Ni_1B_{13}Si_{11}Mo_{2,3}$ with a metallic core diameter of 8.0 μ m and a glass coat's thickness of 2.4 μ m were fabricated by a modified Taylor–Ulitovskiy process.⁶ The wire-composites were prepared by arranging 500 mm long microwires periodically (9 mm spacing between the wires) into silicone rubber sheets, which were then adhered by silicon resin casting. The composite was cured at ambient temperature and the resultant dimensions are 520 mm \times 500 mm \times 1.5 mm. Glass-fiber composite tabs were adhered on both sides of the specimen normal to the length direction using Redux 810A/B. 1/4 in. diameter holes were drilled on both ends of tabs for load bearing. The freespace measurements on the composites were conducted at 1-18 GHz as described elsewhere.⁷ The microwave absorption of microwires was carried out using a modified scalar network analyzer assembly (SNA) at the X-band. In both measurements, loads were applied along the axis of the microwires.

Figure 1(a) depicts the field dependence of absorption of the wire at varying stresses up to 233.3 MPa at 9 GHz. The absorption shows strong magnetic field dependence and the applied magnetic field increases the absorption until saturation in analogy to the valvelike GMI profiles.⁸ With increasing stress, the sensitivity of absorption to magnetic fields is declined. In another aspect, the influence of stress on the absorption increases with increasing magnetic field. In the derivative absorption profiles [Fig. 1(b)], the absorption peaks are found to become wider and shift to a higher magnetic field with increasing applied tensile stress.





FIG. 1. (Color online) Axial magnetic field dependence of absorption (a) and derivative absorption profiles (b) in the presence of varying stresses at 9 GHz for the microwire.

Figure 2 shows the complex permittivity spectra for the as-prepared intact composite after being damaged with wire breakage under tensile loads. There are pronounced changes for both the real part ε' and imaginary part ε'' of the effective permittivity. Particularly, a drastic change of ε' is seen with a reversal of sign from negative to positive when the damage happened to the wires.

A stress tunable behavior parallel to the field tunable behavior is observed in the transmission spectra (Fig. 3). Interestingly, the composite shows a similar response to stress and magnetic field, although the evolution of transmission with magnetic field is steadier than that with stress. To gain a deeper insight into such stress tunable characteristics, we plot in Fig. 4 the calculated stress tunability n_{s21}^s (respectively, magnetic field tenability n_{s21}^f) versus stress (respectively, magnetic field)

FIG. 2. (Color online) The effective permittivity spectra of (a) as-received composite and (b) after damage by tensile stress.

at frequencies of 1, 4.8, and 8 GHz, which are lower than, equal to, and higher than the plasma frequency (f_p), respectively. Here the tunability is defined as the ratio of the induced transmission change to the corresponding magnetic field or stress change. Overall the evolution of tunability remains the same at all three frequencies. In comparison to the single peak feature displayed in the magnetic field dependence of tunability [Fig. 4(b)], both a maximum and minimum appears in the stress dependence of tunability [Fig. 4(a)].

The absorption constant of the microwire, by definition, can be expressed as the reciprocal of skin depth associated with electrical resistivity (ρ), circumferential magnetic permeability (μ), and frequency (f) via the following formula:⁹

$$\alpha = \frac{1}{\delta} = \sqrt{\frac{\pi f \mu}{\rho}},\tag{1}$$



FIG. 3. (Color online) Transmission spectra of microwire composite under tensile stresses (left panel) and magnetic fields (right panel).



where μ is dependent on the susceptibility (χ) and the angel (θ) between the magnetization vector and the wire axis, formulated as⁸

$$\mu = 1 + 4\pi \cos^2(\theta) \chi. \tag{2}$$

For the case of Co-rich microwires, the application of the magnetic field along the wire axis increases the circumferential magnetic permeability by rotating the magnetization vector toward the wire axis and hence the absorption constant. On the other hand, when a stress is applied along the microwires with a negative magnetostriction, the magnetization vector rotates away from the axis direction.¹⁰ As a result, the circumferential magnetic permeability is decreased and hence the absorption is reduced. It is expected, therefore, that the application of a longitudinal stress will compensate the effect of the magnetic field. This can be further proved by the stress impedance (SI) ratio derived from absorption profiles in the presence of varying magnetic fields (not shown here), where the SI ratio was observed to be optimal when the external magnetic field takes the value of 10 Oe, i.e., the anisotropy field of unstressed wire.

As mentioned above, the mechanism of stress tunable effects for microwire composites lies in the dependence of the effective permittivity of the composite on the external stress via the stress-impedance effect the microwires possess. Microwire parameters, such as composition, domain structure, and geometry,¹¹ and the mesostructure constituted by the microwires are critical to determine the stress tunable sensitivity of the microwire wire composite. The wire failure dramatically changes the properties of microwires as well as the periodic topology in the composite mesostructure, thereby resulting in the observed significant change of collective dielectric response.

As reported in our previous work,⁵ the single peak presented in $n_{s21}^{f}(H)$ is associated with the anisotropy field. In contrast, a more complex relationship of $n_{s21}^{s}(\sigma)$ merits more discussion. As discussed above, the circumferential magnetic permeability and surface impedance is reduced upon the application of a longitudinal stress. This explains the maximum occurred at around 13 kPa [Fig. 4(a)]. Afterwards, with increasing stress, the well defined circumferential anisotropy may remain unchanged and hence the surface impedance shows very little variation to the incremental stress, giving rise to a minimum of tunability at 40 kPa. Higher stress than 40 kPa may depreciate the circumferential anisotropy and increase the surface impedance, which accounts for the recovery of the stress tunability.

FIG. 4. (Color online) Stress (a) and magnetic field (b) dependences of tunability at 1, 4.8, and 8 GHz. (plasma frequency $f_p = 4.8$ GHz; the ordinate profiles are normalized by 10^{-4} and 10^{-5} , respectively.)

It should be noted that tens of kPa imposed on the composite yields hundreds of MPa on each wire according to a simple calculation as follows. As the present case meets the isostrain condition, the following equation holds:

$$\varepsilon_c = \frac{\sigma_c}{E_c} = \varepsilon_w = \frac{\sigma_w}{E_w},\tag{3}$$

where ε_c and ε_w denote the strain for composite and microwires, respectively; σ_c and σ_w the stress exerted on the composite and microwires, respectively; and E_c and E_w the Young's modulus of the composite and microwires, respectively. Using the law of mixture, the stress each microwire experiences is given by

$$\sigma_w = \frac{\sigma_c}{(E_m/E_w)f_m + f_w},\tag{4}$$

where f_m and f_w are the volume fraction of the matrix and microwires, respectively. Due to the significant difference between the Young's modulus of rubber matrix (2 MPa) and of microwires (100 GPa), 10 kPa on the composite can result in 500 MPa on the microwires. This is within the reasonable stress range, as commonly discussed in the literature, in terms of the stress effect on GMI properties of microwires.¹²

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