Microwave Absorption of Structural Polymer Composites Containing Glass-Coated Amorphous Microwires

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A new kind of glass fiber reinforced polymers (GFRP) incorporating ferromagnetic microwires are developed for microwave absorption applications. Strikingly, an orientation preference to the absorption due to the polarization effect of glass fibers is observed for the unidirectional GFRP laminates; there also exists a remarkable dependence of absorption on the number of laminates. These observations are rationalized by an equivalent circuit model. With wires addition, we established here the correlation of the microwave absorption performance with the local properties of wires such as wire geometry, the mesostructure including inter-wire spacing, wire orientation, wire pattern (random or continuous) and wire concentration. All these observed effects are elucidated within the framework of microwave absorption theory and equivalent circuit models

Index Terms—Amorphous microwires, microwave absorption, microwire composites.

I. INTRODUCTION

MULTIFUNCTIONAL composites containing microscale or nanoscale fillers have been of much interest to the scientific community in terms of their electromagnetic properties for microwave applications such as tunable microwave devices [1] and remote interrogated sensors [2]. Particularly of interest is the possibility of manipulating the electromagnetic parameters of the composites through controlling the local properties of fillers at the fabrication stage or post-processing treatments and the mesostructure by varying the pattern design of filler topology.

Among a rich family of absorbing materials, ferrites and metallic ferromagnetic materials are widely used in view of their excellent magnetic and microwave properties. Compared with dielectric absorbents, these magnetic absorbents provide additional magnetic losses and achieve better impedance matching. As ferrites have large resistivity with insignificant amount of free electrons, they meet no issue of eddy current loss at high frequency; but they are limited by the Snoek's law [3]. Their intrinsic ferrimagnetic nature renders them inferior soft magnetic properties to the metallic ferromagnetic materials in term of permeability and saturation magnetization [4]. Metallic ferromagnetic materials are advantageous in having a large Snoek's product. But their issue lies in the large conductivity which could make it useless for high frequency applications owing to the eddy current loss, which is formulated as [5]

$$P_e = \frac{CB^2 f^2 d^2}{\rho} \quad (1)$$

where C is the proportional constant, B is the flux density, f is the frequency, d is the sample thickness. ρ is the resistivity. In

a composite material, the concentration p has to be considered, (1) can then be revised to

$$P_e = p \frac{CB^2 f^2 d^2}{\rho} \quad (2)$$

For the low resistivity material to be used in high frequency, the immediate approach as indicated by (2) is to reduce the thickness of the absorbent or their concentration in the composite absorber. This is de facto one of the basic rationales for our adopting dilute ferromagnetic microwires for microwave absorption at gigahertz.

Further, the as-cast magnetic microwire could have a strong magnetoelastic anisotropy which ensures its nature resonance occurs at high frequency. Its good conductivity can yield a large polarization loss at the preferable orientation. Also, the fiber-like shape offers the superior tailorability of electromagnetic properties through wire geometry and concentration. In addition, the microwires possess high mechanical strength ($\sim 10^3$ MPa) and thus would be very useful to make multifunctional composites for engineering applications, which requires a good structural performance for the composite component or coating [6]–[9].

We report here the microwave absorption properties of the composites containing thin Fe-based glass-covered ferromagnetic amorphous microwires embedded in an aerospace-grade E-glass/epoxy prepreg matrix for the first time. These new composites exhibit excellent absorption of up to 12 dB for an extremely low filler loading of up to 0.05 wt%.

II. EXPERIMENTAL DETAILS

Glass-covered microwires with nominal composition $\text{Fe}_{74}\text{Si}_{11}\text{B}_{13}\text{C}_2$ of total diameter $36.2 \mu\text{m}$, ($19.4 \mu\text{m}$, resp) and core diameter $33.8 \mu\text{m}$ ($14.2 \mu\text{m}$, resp) were used to be embedded into the polymer matrix. GFRP 950 E-glass prepreg with the in-plane size of $300 \text{ mm} \times 200 \text{ mm}$ and fiber direction of 0° (w.r.t. the 300 mm edge) were used to prepare microwire composite by hand lay-up and autoclave-cured at the usual cycles. As each laminate contains 10 layers of unidirectional prepreg, the resultant microwire composites end up with a

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Fig. 1. In-plane view of the short (a) and continuous wire composites (b) as examples, the black lines drawn highlight the microwires embedded in random and parallel pattern, respectively. For both samples presented, the size of the composite is $300 \times 200 \times 2 \text{ mm}^3$ and the wire concentration is 0.05 wt.%.

dimension of $300 \text{ mm} \times 200 \text{ mm} \times 2 \text{ mm}^3$. The sufficiently large size of each specimen is selected to minimize the edge diffraction effects.

Microwire composites containing two different microwire configurations were produced, short random microwires and long continuous microwires (Fig. 1), with various microwire contents. For microwave characterization, microwires were laid out or dispersed on the top surface of the composite laminate. Short random wires having length of 30 mm (wavelength $\lambda = 30 \text{ mm}$ at 10 GHz) were decided upon after consultation of Knott [10] while long continuous wire are along the fiber direction 0° w.r.t. the 300 mm edge in a parallel manner. Microwave characterization was performed within the X-band (8–12 GHz) frequency range, using a two-port HP8510 vector network analyzer (VNA), linked to a PC via GPIB with SOLT (short, open, load, thru) calibration. Software interface with the VNA was provided by a bespoke MATLAB code to capture the scattering parameters S_{11} and S_{21} , thus the absorption can be calculated by $A = 1 - |S_{11}|^2 - |S_{21}|^2$.

III. RESULTS AND DISCUSSION

Fig. 2(a) shows the absorption of the pure GFRP laminates. Two main observations are highlighted here. 1) Considering the GFRP is a dielectric material, the absorption displayed ($>10 \text{ dB}$) is significant. 2) The absorption of the GFRP is dependent on the orientation of the fibre. As the conductivity of glass fibre is low, which is expected to have little response to the incident electromagnetic wave, but the difference of absorption for composites containing different fibre orientation clearly originates from the glass fibre, suggesting the large volume fraction of glass fibres do have certain absorption of about maximum 4 dB, (39.8%). This is consistent as shown in a previous paper [11], showing that the tangent loss of the GFRP with the fibre along the electrical field vector is the maximum of all angles of fibre, and the minimum in the case of the fibre perpendicular to the electric field vector. Epoxy is non-conductive as well. Therefore, this absorption should be of a structural origin associated with: 1) a suitable thickness, 2) multiple laminate interfaces. To understand these two mechanisms, one can relate the laminate media to an equivalent circuit as shown in Fig. 3, the equivalent impedance of the whole composite (Z_{eq}) is then derived from the impedance of each layer in parallel connection, expressed as

$$\frac{1}{Z_{eq}} = \frac{1}{Z_1} + \frac{1}{Z_2} + \dots + \frac{1}{Z_n} \quad (3)$$

Fig. 2. (a) Absorption spectra for unidirectional GFRP with different fiber orientation: 0° and 90° with respect to the incident electrical field vector; (b) the number of laminates dependence of maximum absorption.

Fig. 3. (a) Schematic model of the laminates consisting of n layers with each layer of the size $w \times d \times t$, as indicated in the graph; (b) an equivalent circuit composed of parallel connection of capacitances (C) and resistances (R). Z represents the impedance.

After several steps of reduction, one can obtain

$$Z_i = \frac{\omega w t_i (\varepsilon_i'' + j \varepsilon_i')}{d} \quad (4)$$

where Z_i is the impedance of i -th layer, ω is the radian frequency of the electromagnetic wave w , t , d , are the geometrical parameters as indicated in Fig. 3, ε_i' and ε_i'' represent the real and imaginary part of the permittivity of i -th layer, respectively.

In analogy, one can extend (4) to the case of the bulk composites, written as

$$Z_{eq} = \frac{\omega w t_i (\varepsilon_{eq}'' + j \varepsilon_{eq}')}{d} \quad (5)$$

Thus, three primary factors can be identified that contribute to the impedance of each layer: (i) the possible air gap between

layers, of which impedance is determined by the geometry of the gap and the air permittivity [12]. Thus the air gap will create some local discontinuities and fluctuations of impedance and consequently induce higher reflections. (ii) The dielectric transient region between each layer. As the impedance is determined by the geometry of each layer, the thickness variance of each ply will result in a graded material in terms of impedance. This will also induce higher reflections at the transient region of each layer. (iii) Multiple scattering between layers. These structural reasons conspire to an appreciable absorption. Based on these analyses, with increasing the number of laminates, the probability of air gaps will increase and the reflections occurred at the interface will definitely increase. Indeed, this is substantiated in Fig. 2(b). It can also be seen that the materials absorption (dissipated into heat energy), i.e., difference between the absorption of 0° composite and of the 90° one is as small as no more than 2 dB. Previous studies demonstrate that absorption loss and multiple reflection loss counter each other [13], small materials absorption indicates more multiple reflection.

From the whole composite point of view, the equivalent (average) impedance depends on the geometry of the composite, it is well established that there exists a matching thickness of the absorber at which the maximum absorption is achieved due to the ‘geometrical’ effect, i.e., the reflected wave and incident wave cancel each other owing to the phase difference. As such, 2 mm thickness corresponds to a matching frequency at 8.8 GHz. According to the absorption theory, the matching thickness should be decreased for higher frequency applications [14].

Fig. 4(a) compares the short-wire composite and continuous-wire composites with 0° and 90° angle, respectively. A very interesting contrast is shown that the short-wire composite exhibits a slightly larger absorption than the continuous-wire one in the 90° mode while displays entirely the opposite trend in the 0° mode. This suggests the short wires align mostly along the 90° direction, but there indeed exist the component of 0° fibre direction, thus the short-wire composite shows higher absorption in the 90° mode. In the 0° manner, the continuous-wire composite, as expected, shows better absorption than the short-wire composite with the same concentration. In the continuous-wire composite with fixed inter-wire spacing b , reducing b will effectively increase absorption, which can be explained by an equivalent circuit. When an array of microwires is excited by a microwave with electric field E polarized along the microwire, the displacement current $\partial D/\partial t$ flows along both the microwires and air gaps in-between. Hence, the system can be considered as a parallel combination of lumped impedances, expressed as

$$Z_{wire} = \frac{i}{\omega \pi r^2 \varepsilon_{wire}} \quad (6a)$$

$$Z_{air} = \frac{i}{\omega (2rb - \pi r^2) \varepsilon_{air}} \quad (6b)$$

where ε_{wire} and ε_{air} are the permittivities of the studied microwire and air gaps; r denotes the radius of the wire. One can then calculate the equivalent impedance $Z_{eq-wire}$ for each microwire along with its neighbouring air gap in the form of

$$Z_{eq-wire} = \frac{Z_{wire} Z_{air}}{Z_{wire} + Z_{air}} \quad (7)$$

Fig. 4. (a) Absorption spectra for continuous and short microwire/GFRP composites with different wire orientation: 0° and 90° with respect to the incident electrical field vector, respectively; $r = 16.9 \mu\text{m}$, $b = 2 \text{ mm}$, $p = 0.1 \text{ wt\%}$; (b) Absorption spectra for continuous 0° microwire/GFRP composites with different wire gap b ; $r = 7.1 \mu\text{m}$ (c) Absorption spectra for 0° continuous microwire/GFRP composites with wires of different radius r ; $b = 10 \text{ mm}$.

With reference to the above equations, reducing b will give rise to large $Z_{eq-wire}$, consequently inducing a diminishing transmission and improve absorption. Two important aspects are highlighted here. (i) By analogy to the electronic circuits at radio frequency, the absorption (or transmission) spectra can be modulated by varying the equivalent impedance of the circuit, which can be exploited for developing effective filters. (ii) The impedance can be formulated by selecting different b and r that will be discussed later, which lead to the change of absorption maximum and shift of absorption. As shown in Fig. 4(b), the absorption frequency shifts slightly to lower frequency and the absorption bandwidth is enlarged as b is reduced. With further reduction of the wire gap, more desired absorption characteristics can be pursued. But when the wires spacing is reduced small enough to induce indirect dipolar interactions or even direct exchange interactions, the dielectric loss dominated absorption mechanism in the dilute composite system will give way to the magnetic loss dominated mechanism [8].

Subsequently, we examine the impact of the wire diameter. For wires of different diameter, although composites containing the same loading of thin wires have smaller wire spacing, the thick wires composite shows much larger absorption than thin wires composite (Fig. 4(c)). This can be explained by a theoretical model dealing with thin wires of radius smaller than $20 \mu\text{m}$: [15]

$$\varepsilon_{eff} = \varepsilon_0 - \frac{1}{\omega t b} \cdot \left(\frac{\omega L + X_W}{R_W^2} + j \frac{1}{R_W} \right) \quad (8)$$

which conveys that, with the fixed wire spacing b , the dielectric loss (imaginary part of effective permittivity ε_{eff}) of wire arrays is very much dependent on the dc resistance of the wire R_W

rather than inductance L or reactance X_W . As the thick wire has smaller resistance, its composite therefore shows better absorption. On the other hand, we can also understand this effect of wire diameter through its impact on the anisotropy field. As reported previously by Chiriac *et al.* [16] for $\text{Co}_{68.15}\text{Fe}_{4.35}\text{Si}_{12.5}\text{B}_{15}$ glass-coated microwires having the same glass thickness ($d = 5, 10$ and $20\ \mu\text{m}$), the circumferential anisotropy constant first increased with an increasing d from $5\ \mu\text{m}$ to $20\ \mu\text{m}$ and then decreased for $d > 20\ \mu\text{m}$. In this sense, for the present wire, with the increase of wire diameter, the effective anisotropy field should be increased. In consequence, higher natural ferromagnetic resonance could be excited. It is also worth mentioning that, the effect of wire diameter is split at $20\ \mu\text{m}$. Our previous work [17] has experimentally proved that the electromagnetic behaviour of thicker wires ($d > 20\ \mu\text{m}$) conforms to a different model that, in contrast to the case of thin wire ($d < 20\ \mu\text{m}$) as discussed in the present work, describes an opposite relationship between wire resistance and the effective permittivity; it teams with (8) to provide a robust theory in explaining the microwave properties of microwire composites.

IV. CONCLUSIONS

In conclusion, a detailed study of the microwave absorption of GFRP and microwire/GFRP composites is presented. In spite of the weak absorption from the glass fibre, the structural imperfections arising from the laminate composite fabrication contribute significantly to the absorption of GFRP. The composites containing continuous thin wires oriented along the electrical field vector are favorable for large absorption. The response of the wire composite can be modelled by an equivalent circuit parameterized by the equivalent impedance, which provides a deep understanding of the microwave absorption properties. From the absorption mechanism perspective, it has been found that dilute ferromagnetic microwire composite behaves as artificial dielectric material. The geometry of the microwires and their composite mesostructure have a profound impact on the absorption features.

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