Exceptional electromagnetic interference shielding properties of ferromagnetic microwires enabled polymer composites

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We present systematic studies of the electromagnetic interference (EMI) shielding and microwave properties of a new class of shielding material, i.e., the ferromagnetic microwires-embedded polymer composites. We show that at 1–2 GHz the shielding effectiveness (SE) of the continuous-wire composite reaches a high value of 18 dB (98.4% attenuation) for a very low filler loading of 0.024% and a thickness of 0.64 mm. The normalized SE of this new composite is about 70 times higher than that of the bucky paper-based composite and is two to four orders of magnitude higher than those of other shielding candidate materials. Complex permeability, permittivity, and impedance experiments reveal that the absorption of electromagnetic radiation is a dominant mechanism for EMI shielding of the studied composites. The advantages of high shielding efficiency, good physical integrity, low fabrication costs, and multifunctionalities make them an attractive candidate material for a variety of technological applications. © 2010 American Institute of Physics. [doi:10.1063/1.3471816]

I. INTRODUCTION

Electromagnetic interference (EMI) is the electromagnetic (EM) radiation emitted by electrical and electronic equipments. EMI among electronic devices such as computers, mobile phones, radios, and airplane navigators can degrade device performance.1–3 It also causes potential health hazards such as insomnia and cancer. With the rapid proliferation of electronic and telecommunication devices, control over EMI becomes imperative but remains a challenge. To control the increasing EMI, it is vital to develop new materials with enhanced EMI shielding capacity. According to the EM theory, high frequency EM radiation only occurs in the surface layer of an electrical conductor. This is well known as the skin effect characterized by the skin depth (δ), which is a measure of a drop in the electrical field of a plane wave penetrating into the conductor, associated with electrical resistivity (ρ), magnetic permeability (μ), and frequency (f) via the following formula:4

$$\delta = \sqrt{\frac{\rho}{\pi f \mu}}. \hspace{1cm} (1)$$

Since a smaller skin depth results in larger attenuation, lower resistivity and higher permeability are required for achieving a better shielding performance. In this context, a wide range of candidate materials have been suggested for EMI shielding applications. The most typical material among them is the polymer composite containing metallic fillers such as steel, nickel, silver in the form of powder fiber or flake.5–8 It was reported that the shielding effectiveness (SE) exceeded 30 dB for steel fiber reinforced composites with a filler loading of 6 wt % and a thickness of 1.5 mm at 1–2 GHz.7 The SE of more than 50 dB was also reported for the nickel powder-filled composites with a filler loading of 40 wt % and a thickness of 2.85 mm.5 While these fillers benefit the shielding properties of the resultant composites, the large density, physical rigidity, and low corrosion resistance of the metallic phase are disadvantageous. In addition, a relatively high loading of these fillers is required for achieving low resistivity and high SE. Recently, carbon fiber9–12 and carbon nanotubes (CNTs) (Refs. 13–20) have been shown to be useful for EMI shielding applications. In the case of carbon fibers, it would be ideal if they could yield large SE as these materials are widely used in aerospace and automobile industry. However, when compared with the metallic fillers, the higher resistivity of the carbon fibers imposes a stringent requirement of a larger loading of carbon fibers in order to achieve the same SE, thus raising the manufacture cost. While some efforts had been made to improve the conductivity of the fibers by coating them with a thin metallic layer, the coating itself could be easily damaged during the coating processing.9 For the case of the CNTs-based composites, Li et al.13 reported the SE of more than 25 dB for multwall carbon nanotube (MWCNT) polyacrylate composites with 10 wt % CNTs and a 1.5 mm thickness at X-band (8–12 GHz). Park et al.14 found that a 15 μm thin CNT bucky paper (BP) containing more than 50 wt % CNTs reached the SE of 22 dB at 2–18 GHz. By catalyzing the CNT with magnetic particles such as Fe, Co, Ni, and their compounds, the SE of the CNTs-based composites were significantly en-

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enhanced due to the increased absorption as a result of the introduction of magnetic permeability. However, the difficulty in dispersing nanosized fillers and high fabrication costs hinder the CNTs-based composites from commercial applications. Some conductive polymers have recently been developed for EMI shielding applications, but their poor mechanical properties make them undesirable for such applications.

Soft ferromagnetic amorphous glass-coated microwires (AGCMs) with giant magnetoimpedance (GMI) or giant stress-impedance (GSI) effect are excellent candidate materials for making highly sensitive magnetic and stress sensors. Recently, we have developed a new concept of incorporating AGCMs with GMI/GSI effect into fiber-reinforced composites as means of sensing stress and external magnetic fields through changes in impedance, which offers an alternative to optical fibers for self-monitoring composites.

We have demonstrated the remarkable field tunable properties of the microwire composite and the retained mechanical properties of the resultant composite with the embedded wires. These studies open up new opportunities to develop smart structural composites with self-monitoring properties for a wide range of engineering applications. Since AGCMs possess their small dimension (1–30 μm in diameter), high electrical conductivity (~6 × 10^3 S/cm), high magnetic permeability (~10^4), and high mechanical strength (~10^3 MPa), they can be incorporated into polymer-based composites for creating high-performance EMI shielding composite materials at low filler loading.

We report the first study of the EMI shielding properties of the composites containing soft ferromagnetic AGCMs embedded in a polymer matrix. These new composites exhibit a high SE of 18 dB (98.4% attenuation) for a very low filler loading of 0.024% and a thickness of 0.64 mm. The EMI shielding properties can be enhanced by reducing wire spacing and/or tailoring the soft magnetic properties of the microwires. The systematic analyses of complex permeability, permittivity and impedance reveal that the absorption of EM radiation is a dominant mechanism for EMI shielding of the studied composites.

II. EXPERIMENTAL

Glass covered amorphous microwires of Co_{68.7}Fe_{3.85}Ni_{1.44}B_{11.53}Si_{14.47}Mo_{1.66} (No. 1) and Co_{68.7}Fe_{3.85}Ni_{1.44}B_{11.53}Si_{14.47}Mo_{1.66} (No. 2) were fabricated by the Taylor–Ulitovsky method and were supplied by the Microfiber Technology Industry (MFTI). The information on the dimension and the magnetic parameters of the microwires are summarized in Table I. Four layers of E-glass prepreg 913 (500 × 500 mm^2) were used to prepare the composites containing the microwires in a periodic manner following the procedure detailed in our previous paper. Two wire configurations were designed in these wire-composite systems: the continuous-wire composites [the wire length of 50 cm and the wire spacing (b) of 3, 7, 9 mm] and the short-wire composites (the wire length of 5 cm and the wire spacing of 5 mm and 30 mm in perpendicular and parallel direction, respectively). For convenience of discussion, we denote the composites containing continuous No. 1 and No. 2 microwires as C1 and C2, respectively. The short-wire composites containing No. 1 microwires are denoted as S1. The microwave properties and SE were studied at S-band (1–4 GHz) by free-space measurements of the transmission (S21) and reflection spectra (S11). The detailed description of the free-space measurement system can be found in Ref. 35. Effective complex permittivity, magnetic permeability, and fractional impedance were extracted from the obtained transmission/reflection profiles. As compared with the coaxial transmission line method, the free-space method provides a higher degree of accuracy in measured S-parameters and hence the permittivity and permeability precisely computed from these data.

III. RESULTS

Figure 1 shows the frequency dependence of SE for the composite samples with different wire spacing (b). As one can see clearly from this figure, SE decreases with increasing frequency for all the samples, and it increases as the wire spacing is reduced. Notably, a large improvement of SE of more than 10 dB is achieved as the wire spacing decreases.

![Graph showing frequency dependence of SE for different wire spacings](image-url)

FIG. 1. (Color online) Frequency dependence of the SE of the wire-composites with varying wire spacing b at 1–4 GHz. The inset shows the fractional change in impedance of the same samples at 1–4 GHz.
from 7 to 3 mm corresponding to a volume fraction of 0.024%. A similar trend has been observed for the fractional change in impedance as shown in the inset of Fig. 1.

The EMI attenuation can arise from three different sources, including reflection (R), absorption (A), and multiple reflection (M), which is given by:

\[ SE = -10 \log \frac{P_{out}}{P_{in}} = SE_R + SE_A + SE_M, \tag{2} \]

where \( P_{out} \) and \( P_{in} \) are the incident power and transmitted power, respectively. The reflection SE, \( SE_R \), can be approximated as:

\[ SE_R = -10 \log(1-R). \]

Since the multiple reflection is only considered in the presence of large surface areas as in a porous or foam material, it is not relevant in the present case. The absorption SE, \( SE_A \), can thus be approximated as:

\[ SE_A = -10 \log[T/(1-R)], \]

where \( T \) stands for transmission. The wire volume fraction dependencies of SE, \( SE_A \), and \( SE_R \) at 1–2 GHz are shown in Fig. 2. It is readily shown that both the EM reflection and absorption contribute to the shielding effect but the absorption of EM radiation is a dominant mechanism for EMI shielding of the composites. Furthermore, it is noted that reducing the wire spacing enhances both \( SE_A \) and \( SE_R \) and hence SE. From an EMI shielding application perspective, it is very interesting to note that, with merely 0.024 vol %, SE reaches approximately 18 dB, which corresponds to an attenuation of 98.4%.

Figure 3 shows the complex permittivity spectra for the composites with \( b=7 \) and 3 mm. Note that the values of dielectric constants discussed here are absolute values. It can be observed that both the real and imaginary parts of complex permittivity decrease as the frequency is increased. The real part of the permittivity is larger than the imaginary part for both the samples. At 1–2 GHz, \( \varepsilon'' \) varies from 13 to 85 for the composite sample with \( b=7 \) mm and from 2 to 6 for the composite sample with \( b=3 \) mm. It is clear that the permittivity strongly increases with decreasing wire spacing. This dependence is correlated with frequency evolution of SE (Fig. 1) and the fractional change in impedance (the inset of Fig. 1).

Finally, we performed a comparative study of the magnetic and shielding properties of the composites having the same wire spacing of 7 mm but using different microwires (Co$_{68.7}$Fe$_{4}$Ni$_{13.5}$Si$_{14.4}$Mo$_{2.3}$ and Co$_{67.05}$Fe$_{3.85}$Ni$_{14.5}$Si$_{14.4}$Mo$_{1.66}$) as described in Table I. The results are presented in Fig. 4. It can be seen that \( C_2 \) possesses a higher SE, larger complex permittivity and permeability than \( C_1 \). The initial dc permeability calculated from the hysteresis loops for \( C_1 \) and \( C_2 \) are 1.054 and 1.048, respectively, showing a negligible difference. But significant variations in the ac permeability and permeability spectra are observed for these samples. At \( f=1 \) GHz, for instance, \( \varepsilon =15–12.5i, \mu=0.84–0.7i, \) and SE=9 dB for \( C_1 \), while \( \varepsilon =23.5–20i, \mu=1.33–1.14i, \) and SE=10.4 dB for \( C_2 \). In the case of the short-wire composites, a small attenuation of around 4 dB is obtained. When compared with the continuous-wire composites, the short-wire composites possess larger dielectric and magnetic losses. To put our results in the context of the EMI shielding application, we summarized in Table II the SE and normalized SE of the continuous-wire composites with other shielding candidate materials. The filler loading and sample thickness are also given for an appraisal of shielding efficiency.

IV. DISCUSSIONS

The magnetic microwires are periodically dispersed in a polymer matrix in a parallel fashion. The EM wave passes through the space between the microwires by scattering, resulting in a power attenuation. Due to the ordered filler architecture of the composite, its SE can be modeled as:

\[ SE \approx -20 \log(b \cdot f), \tag{3} \]

where \( b \) denotes the wire spacing and \( f \) is the frequency. Equation (3) explains an inverse relationship between the SE and wire spacing or frequency as presented in Fig. 1. Since the decrease in the wire spacing decreases electrical resistivity and increases magnetic permeability of the composites, according to Eq. (1) the skin depth \( \delta \) will be decreased and consequently the absorption SE\(_A\) will be increased as SE\(_A\) \( \propto \frac{1}{f \mu \rho} \), where \( t \) is the thickness of the composite. Since SE\(_R\) is proportional to \( \log(1/f \mu \rho) \), the increase in SE\(_R\) is relatively small (see Fig. 2) which implies a dominant role of
On the underlying physical mechanism for the EMI shielding of the ferromagnetic microwire composites, we first note that the real and imaginary parts of the permittivity are associated with storage and loss effects. Larger $\varepsilon'$ than $\varepsilon''$ suggests a stronger absorption than reflection. The strong polarization can be inferred from the high values of $\varepsilon'$ and $\varepsilon''$ for the composite with $b=3$ mm. With increasing wire spacing, the polarization is massively reduced, because there exists fewer elements to induce polarization and scattering with free charges. The periodical arrangement of the microwires helps utilize the resonance absorption of each microwire resulting from GMI effect and dipolar interactions between the domains induced by magnetic flux closure in a multiwire system. Such effects will be strengthened in the composites with decreased wire spacing. Note that $\varepsilon''$ can be approximated as $1/(2\pi\varepsilon_0\mu_0f)$ according to free electron theory. A higher $\varepsilon''$ of the composite with narrower wire spacing also indicates a lower resistivity.

Furthermore, within the framework of the EM theory, the domination of absorption over reflection suggests a serious impedance mismatch between input impedance ($Z_{in}$) and free-space impedance ($Z_0$) in view of the reflection loss (RL) that can be given by:

$$RL = 20 \log \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right|,$$

where $\mu_r$ and $\varepsilon_r$ are the relative complex permeability and permittivity of an absorber. $c$ denotes the light velocity and $t$ is the thickness of the absorber. The greater increase in magnetic permeability than that of permittivity (related to resistivity) with decreasing wire spacing narrows the impedance mismatch and thus improves the absorption and reflection, and the total SE is consequently enhanced. The ac permittivity and permeability play crucial roles in determining the shielding effect. From Table I and Fig. 4, it seems that the continuous microwire with softer magnetic property does not necessarily make it favorable for EMI shielding. The short-wire pieces have proved to be inferior to the continuous wires as the shielding functional fillers probably owing to the sparse dispersion of them lowering the total polarization. A previous study has revealed a significant influence of the

The increased permeability. This well agrees with the wire spacing and frequency dependencies of impedance (cf. the inset of Fig. 1).

**FIG. 4.** (Color online) (a) Frequency dependence of the SE of the composites containing Co$_{63.8}$Fe$_{23}$Ni$_{12}$Si$_{1.5}$Mo$_{2.5}$ (C$_1$), Co$_{63.8}$Fe$_{23}$Ni$_{12}$Si$_{1.5}$Mo$_{2.5}$ (C$_2$), and short-wire composites containing No. 1 wire (S$_1$). The magnetic parameters of the microwires are listed in Table I. The inset in (a) shows the hysteresis loops of both the microwires; (b) the complex permittivity, and (c) permeability spectra of their composites.
aspect ratio of carbon fibers on the SE of the carbon fiber-based composites. This would lead to a similar expectation for the case of the ferromagnetic microwire-based composites. A plausible explanation for our composites is the following: the shape effect is critical in determining the magnetic and dielectric response of the materials; a larger aspect ratio will greatly decrease the demagnetizing field and accordingly increase the field efficiency and material response. From the permittivity spectra, the larger value of the imaginary part of permittivity implies a reflection-dominant shielding mechanism for $S_1$, which can be related to the emergence of current in wire pieces which enhanced the surface impedance.

A further analysis of the shielding effect with respect to filler loading and sample thickness indicates the superior shielding properties of the ferromagnetic microwires-based composites. A wire loading as low as 0.026 vol % (0.09 wt %) suffice to yield 18 dB (98.4% attenuation). As one can see readily in Table II, quantitatively, the normalized SE with respect to that in the present work at 1–2 GHz is about 70 times (respectively, 50 times) larger than that of the BP (respectively, BP composites), a well known composite possessing excellent shielding properties. In comparison with other materials, the normalized SE of the ferromagnetic microwires-based composite exceeds by two to four orders of magnitude. It should be noted that although the influence of differing measurement techniques may compromise such a sharp contrast, the microwire composite shows great potential in the shielding application. From an application point of view, 10 dB is the minimum required value, and 60–90 dB is a sharp contrast, the microwire composite shows great potential in the shielding application.

V. CONCLUSIONS

We have studied systematically the EMI shielding and microwave properties of the short and continuous ferromagnetic microwire-embedded polymer composites. It is shown that the composite with 0.024 vol % microwires is able to provide 18 dB protection at 1–2 GHz. The shielding effect is demonstrated to be dominated by the absorption of the EM radiation, which depends on the wire spacing and microwave properties. The advantages of high shielding efficiency, good physical integrity, low fabrication costs, and multifunctionalities make the proposed composites an attractive candidate material for a variety of technological applications.

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