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Effect of Magnetic Orientation on the Microwave Absorption Properties for Planar-Anisotropy $Ce_2Fe_{17}N_{3-\delta}$ Powders/Silicone Composite

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Abstract. The electromagnetic parameters of magnetic unoriented and oriented $Ce_2Fe_{17}N_{3-\delta}$ powders/Silicone composite with 15 vol% were investigated in the 0.1–18 GHz range. It was found that the initial permeability increased from 2.78 to 3.6 as well as the optimal RL reached –61.3 dB with a thickness of 1.91 mm at 9.01 GHz for the composite after orientation. The microwave absorbing properties of oriented composite were greatly enhanced resulting from the perfect impedance match condition improved by rotational magnetic orientation. This novel rotational magnetic orientation method might provide a new way to design high performance microwave absorbers.

Introduction

Recently, in order to solve serious electromagnetic interference (EMI) and compatibility (EMC) problems owing to the increasing usage of information technology and communication facilities in commercial and military applications, considerable attention has been paid to microwave absorbers [1, 2]. The perfect microwave absorbers should have the characteristics of light weight, thin thickness, broadband and strong reflection loss [3]. To satisfy those requirements, high permeability and high resonance frequency are demanded for microwave absorbers [4, 5]. Rozanov pointed out that the bandwidth of EM absorbers is directly proportional to their permeability [6]. Also, in the work carried out by Wang et al., high permeability is not only crucial for getting thin absorber matching thickness but also for improving impedance matching condition [1].

However, for traditional microwave absorbers, the permeability value of which are small in GHz frequency range owing to the snoek's limit [7], namely:

$$(\mu_0 - 1)f_r = \frac{2}{3}\gamma M_s \tag{1}$$

Where μ_0, f_r , γ, M_s are the initial permeability, the resonance frequency, the gyromagnetic ratio, the saturation magnetization, respectively. In order to improve the permeability of microwave absorbers, the magnetic materials with planar anisotropy have been drawing much attention [5, 8-10].The product of the static susceptibility ($\mu_0 - 1$) and resonance frequency f_r for these magnetic materials can be described by [11]

$$(\mu_0 - 1)f_r = \frac{2}{3}\gamma M_s\sqrt{\frac{H_\theta}{H_\phi}} \tag{2}$$

where H_θ and H_ϕ are the out-of-plane and in-plane anisotropy fields, respectively.

Consequently, the resonance frequency and permeability of those planar-anisotropy magnetic materials can be greatly enhanced compared with traditional microwave absorbers due to the large value of $\sqrt{\frac{H_\theta}{H_\phi}}$ and M_s [10, 12]. In this paper, planar-anisotropy $Ce_2Fe_{17}N_{3-\delta}$ compound was reported as a microwave absorber in view of its high permeability in the GHz range and high resonance frequency [2].

In order to further enhance the microwave absorption performance, higher complex permeability is necessary. It was reported that the permeability of the composite consisting of planar anisotropy magnetic powders can be improved by rotational orientation [2, 10]. In our work, the RL of $Ce_2Fe_{17}N_{3-\delta}$ powders/Silicone composite with 15 vol% before and after orientation have been studied in 0.1–18 GHz frequency range. As a result, the initial permeability increases from 2.79 to 3.6 after rotational orientation. Furthermore, the microwave absorbing properties of oriented composite is far more enhanced than the unoriented at the same absorber thickness, which indicates that impedance matching condition can be improved by orientation. Finally, the mechanism of the enhanced microwave absorption performance were also discussed in details.

Experimental

The alloy Ce_2Fe_{17} ingots were prepared from Ce (>99.9% in purity), Fe (>99.99% in purity) by induction-melting under Ar atmosphere. Extra 5% Ce was added for compensating the loss of Ce due to evaporation. A single-roller melt-spinning method in an Ar atmosphere was used to produce the rapidly solidified ribbons with a thickness about 100 μm. The as-casted ribbons were broken into micrometer-sized particles in an agate mortar and then sieved to produce fine powders. The nitrogenation reaction of the fine powders was performed in a stainless steel tube (pressure: 0.8 MPa, volume: 3000 cm^3) with high-purity nitrogen gas for 1 h at 743 K. The nitrided powders were milled for 12 h on a vibrating ball mill with the ball-to-powder weight ratio of 25:1 in absolute ethyl alcohol. The as-milled powders was naturally dried in room temperature.

The phases were examined by X-ray diffraction (XRD, Bruker D8 Advance) using Cu-Kα radiation. The morphology was analyzed by scanning electron microscope (SEM, FEI–QUANTA–250–FEG). The powders were mixed by Silicone with 15% volume concentrations. Afterward, a toroidal shape composite with outer diameter of 7.0 mm and inner diameter of 3.04 mm was fabricated by pressing the mixture in a mold. For preparing the oriented composite, the mold with mixture inside it was placed in a pulsed magnetic field with the maximum intensity of 1.7 T, and subsequently rotated around its axis in the perpendicular pulsed magnetic field until the composite was solidified. The electromagnetic parameters of unoriented and oriented composite samples were obtained using an Agilent N5234A vector network analyzer in the 0.1–18 GHz range.

Results and Discussion

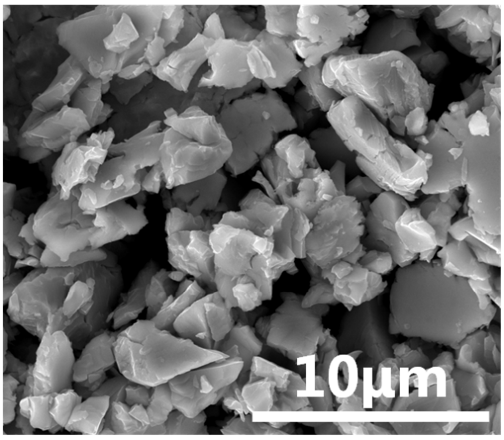

Fig. 1. SEM image of $Ce_2Fe_{17}N_{3-\delta}$ powders.

Microstructure and Phase Analysis. Fig. 1 shows the SEM image of $Ce_2Fe_{17}N_{3-\delta}$ powders after ball milling. It can be seen that the powders is of irregular shaped powders, ranging from 2 to 5 μm with a mean diameter of 4 μm.

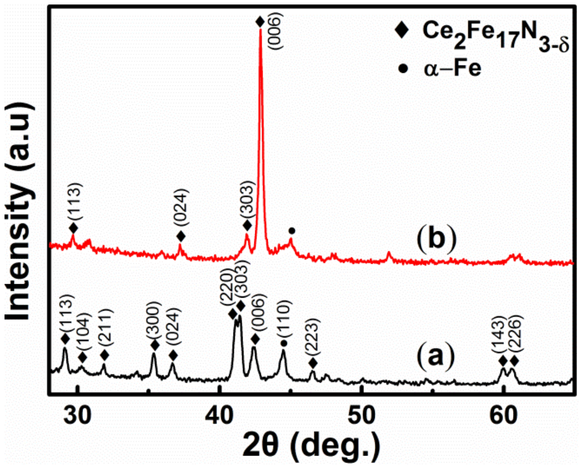

Fig. 2. XRD patterns of (a) $Ce_2Fe_{17}N_{3-\delta}$ free powders and (b) oriented $Ce_2Fe_{17}N_{3-\delta}$/Silicone composite.

Fig. 2 show the XRD patterns of $Ce_2Fe_{17}N_{3-\delta}$ free powders and oriented $Ce_2Fe_{17}N_{3-\delta}$/Silicone composite, respectively. It can be seen in Fig. 2(a) that all the diffraction peaks for the powders match well with standard powder diffraction data, which shows a rhombohedral Th_2Zn_{17}-type structure [13], and a small amount of α-Fe phase in the nitrides is observed. Compared with the unoriented powders, the peak intensity of the (006) crystalline plane is strongly enhanced and the other peaks of the powders almost disappear after orientation, these results clearly present that the easy magnetization direction is in the (006) plane (c-plane) for the $Ce_2Fe_{17}N_{3-\delta}$ powders.

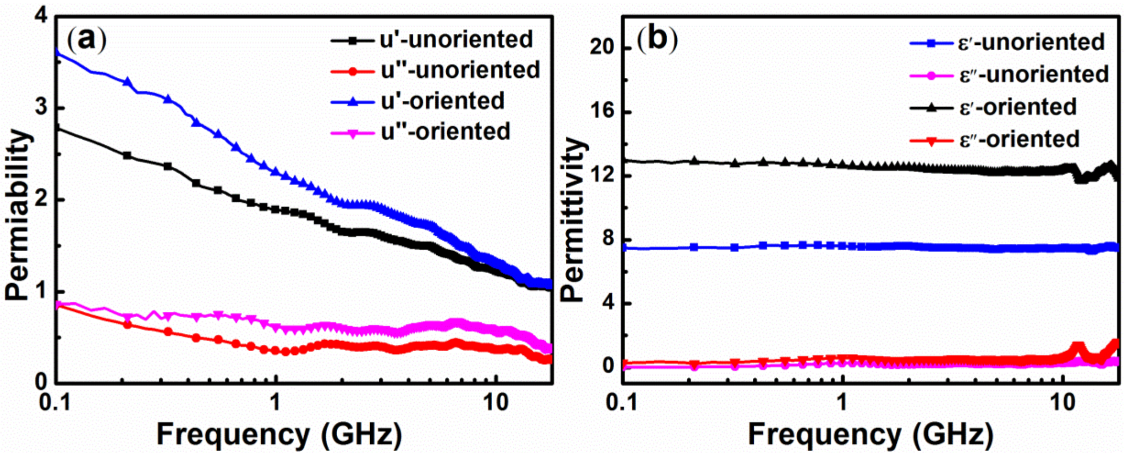

Fig. 3. Plots of (a) complex permeability and (b) permittivity vs. frequency for unoriented and oriented $Ce_2Fe_{17}N_{3-\delta}$ composites with 15 vol%.

Complex Permeability and Permittivity. Plots of (a) complex permeability (μ_r=$-j\mu''$) and (b) permittivity (ε_r= $\varepsilon' - j\varepsilon''$) vs. frequency for unoriented and oriented $Ce_2Fe_{17}N_{3-\delta}$ composites with 15 vol% are shown in Fig. 3. It is clearly in Fig. 3(a) that both theand μ'' were enhanced after orientation, which can be ascribed to the planar anisotropy of $Ce_2Fe_{17}N_{3-\delta}$ and orientation [14, 15]. The value of increases from 2.78 to 3.6 at 0.1 GHz after orientation, M. Matsumoto indicated that the value of the complex permeability of the oriented sample is 1.2–1.5 times larger than the unoriented one, the result in our paper agrees with his report [16]. The value of μ'' of those unoriented and oriented samples fluctuates around 0.5 in the measured frequency range.

As shown in Fig. 3(b) that the complex permittivity of both unoriented and oriented $Ce_2Fe_{17}N_{3-\delta}$ composites are almost independent on increasing frequency. The complex permittivity, represent the storage capacity of charge and the loss energy dissipative abilities, which associated with the space-charge polarization between $Ce_2Fe_{17}N_{3-\delta}$ powders isolated by silicone and the resistivity of the composite [17, 18]. Therefore, the value of ε' for the oriented sample greatly enhanced owing to the enhancement of the space-charge polarization. In addition, the electrical anisotropy was introduced by orientation, the conductivity of the oriented composite in the direction perpendicular to the orientation is higher than the unoriented [1].

Microwave Absorbing Properties. According to the transmission line theory, when the electromagnetic wave is normally incident on a single-layer absorber backed by a perfect conductor, the normalized input impedance (Z) and the RL of a microwave absorber with a certain thickness can be calculated from the complex permeability and permittivity via the following expressions [19]:

$$Z = \frac{Z_{in}}{Z_0} = \sqrt{\mu_r/\varepsilon_r}\tanh\left(j(2\pi f t/c)\sqrt{\mu_r\varepsilon_r}\right) \tag{3}$$

$$RL = -20\lg\left|\frac{Z_{in}-Z_0}{Z_{in}+Z_0}\right| \tag{4}$$

Where Z_{in} represents impedance of the composite, Z_0 is intrinsic impedance of free space, $Z_0 = (\mu_0/\varepsilon_0)^{1/2} = 377\ \Omega$, t represents the absorber thickness, c represents velocity of light in free space, f represents frequency, μ_r and ε_r are the complex permeability and complex permittivity.

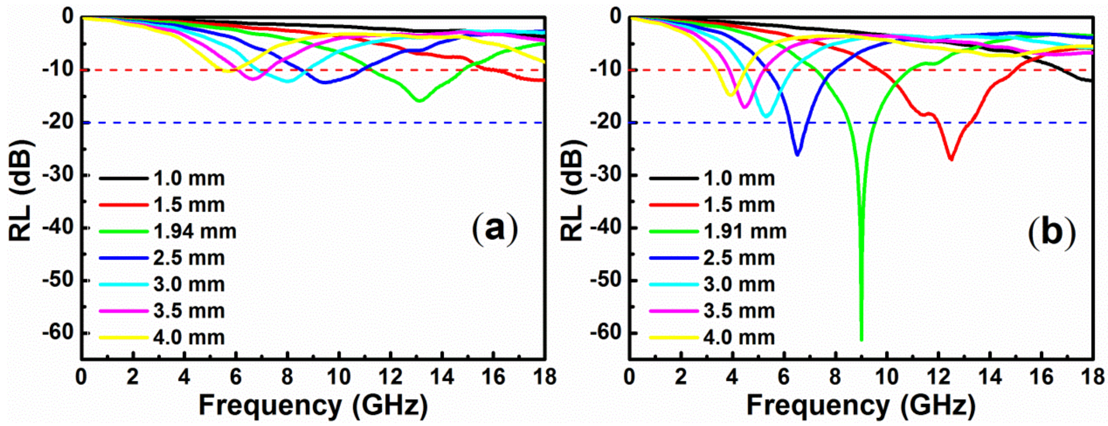

Fig. 4. Plots of RL against frequency for (a) unoriented and (b) oriented $Ce_2Fe_{17}N_{3-\delta}$/Silicone composites with 15 vol% at different absorber thicknesses.

Fig. 4 shows the frequency dependence of RL for the (a) unoriented and (b) oriented $Ce_2Fe_{17}N_{3-\delta}$ composites with 15 vol% at different absorber thicknesses. It can be seen that the RL peak appears at a frequency when an absorber thickness is given, and the peak position obviously shifts to a lower frequency with the increasing absorber thickness for both samples. These results can be clearly explained by interface-reflection model [20], namely:

$$t_m = \frac{n}{4}\frac{c}{f_m\sqrt{|\mu_r\varepsilon_r|}},\quad n = 1,3,5\ldots \tag{5}$$

Where t_m and f_m are matching thickness and matching frequency, respectively.

It can be concluded that t_m is in inverse proportion to f_m at the given product of $|\mu_r\varepsilon_r|$. Consequently, compared with the unoriented sample, RL peak shifts to a lower frequency at the same thickness for the oriented sample owing to the increasing product of $|\mu_r\varepsilon_r|$, as shown in Fig. 4(a) and (b). Furthermore, the optimal RL of the unoriented composite reaches –16 dB at 13.1 GHz with a thickness of 1.94 mm. However, the optimal RL of the oriented composite is as high as –61.3 dB at 9.01 GHz with the thickness of 1.91 mm, and the bandwidth with RL values exceeding –10 dB is 3.67 GHz, which almost covering the entire X-band frequency range, the performance of which outdistances most of previous reported ferrite composite [21-23]. On the other hand, the intensity of the RL peak greatly enhanced after orientation for a given thickness. These phenomena can be explained by impedance matching condition, as shown in Fig. 5.

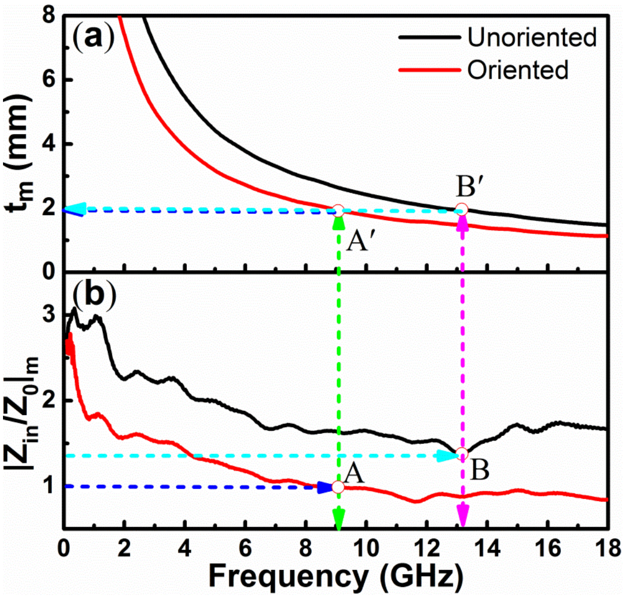

Fig. 5. Plots of (a) matching thickness t_m and (b) normalized input impedance on matching frequency f_m for unoriented and oriented $Ce_2Fe_{17}N_{3-\delta}$/Silicone composites.

Fig. 5(a) and (b) show the plots of matching thickness t_m and normalized input impedance $|Z_{in}/Z_0|_m$ on matching frequency f_m for unoriented and oriented $Ce_2Fe_{17}N_{3-\delta}$/Silicone composites with 15 vol%, respectively. The intensity of RL peak is determined by normalized input impedance (Z) from Eq. (4), and the optimal RL which corresponds to negative infinite RL value can be gained only when Z=1, in this situation, the corresponding absorber thickness and peak frequency are defined as perfect matching thickness t_{pm} and perfect matching frequency f_{pm}, respectively. Consequently, the corresponding parameters of microwave absorbing properties can be directly obtained from the crossover point A and A′ in Fig. 5. However, it is also worth noting that Z=1 can’t be found in the entire frequency range for unoriented sample, as the crossover point B and B′. Fig. 5(a) also indicates that thinner matching thickness could be obtained with the oriented sample due to the enhancement of the permeability and permittivity. By taking advantage of the intrinsic planar-anisotropy, the perfect matching point occurs and the perfect matching frequency is tuned to X-band (9.01 GHz) after orientation.

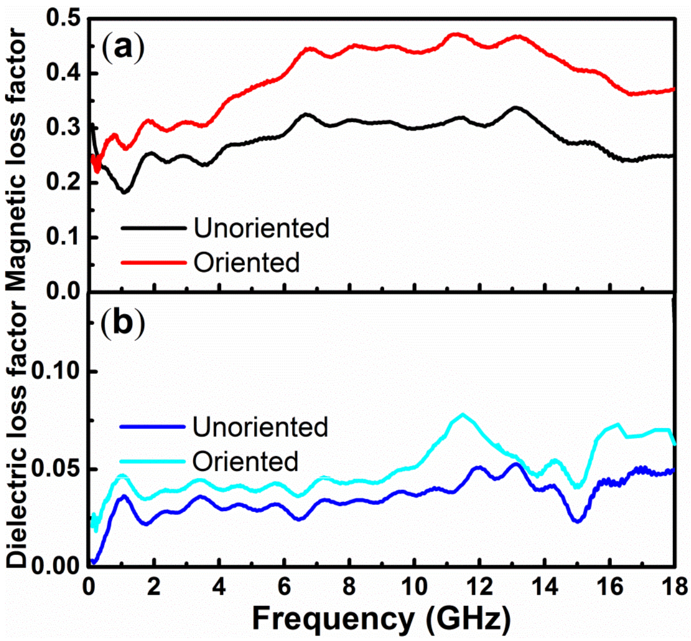

Fig. 6. Frequency dependence of (a) magnetic loss factor and (b) dielectric loss factor for the unoriented and oriented $Ce_2Fe_{17}N_{3-\delta}$/Silicone composites.

Dielectric loss and magnetic loss are two possible contributions for microwave absorption. Both magnetic loss factor ($\tan\delta_{\mu} = \mu''/\mu'$) and dielectric loss factor ($\tan\delta_{\varepsilon} = \varepsilon''/\varepsilon'$) based on the permeability and permittivity of the composite are measured as shown in Fig. 6(a) and (b), respectively. It is noticed in Fig. 6 that $\tan\delta_{\mu}$ was enhanced after orientation, so did the $\tan\delta_{\varepsilon}$. What's more, $\tan\delta_{\mu}$ is much larger than $\tan\delta_{\varepsilon}$ in the 0.1–18 GHz range, which indicates that magnetic loss plays a more important role in the $Ce_2Fe_{17}N_{3-\delta}$/Silicone composites in this frequency range.

Conclusions

In conclusion, the planar anisotropy $Ce_2Fe_{17}N_{3-\delta}$ powders has been successfully prepared. By employing the rotational orientation, the permeability and permittivity of the $Ce_2Fe_{17}N_{3-\delta}$ powders/Silicone composites were enhanced, resulting in the improvement of impedance matching condition. Moreover, there is no perfect matching for the unoriented composite with 15 vol%, and all the reflection loss (RL) can't exceed –20 dB at the measuring range. However, the optimal RL of this composite is –61.3 dB with a thickness of 1.91 mm at 9.01 GHz after orientation, and the bandwidth with RL less than –10 dB is 3.67 GHz. In a word, the microwave absorbing properties of oriented composite is far more enhanced than the unoriented at the same absorber thickness, which indicates that impedance matching condition can be improved by orientation. This novel rotational orientation method may provide a new way to design high performance microwave absorbers.

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