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Tunable electromagnetic properties and enhanced microwave absorption ability of flaky graphite/cobalt zinc ferrite composites

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ABSTRACT

In this study, the flaky graphite/cobalt zinc ferrite composites were synthesized by the coprecipitation method. The XRD results showed that the flaky graphite/cobalt zinc ferrite composites can be obtained and there is no impurity when the graphite weight ratio is less than 20 wt%. The FE-SEM results exhibited that the flaky graphite/cobalt zinc ferrite composites possessed flake-like shape with high aspect ratio. Moreover, the electromagnetic properties could be tuned by varying the graphite weight ratio. In the series of as-prepared flaky graphite/cobalt zinc ferrite composites, the sample with 10 wt% graphite possessed excellent impedance matching performance. As a result, the it exhibited the best microwave absorption properties. The reflection loss was less than −10 dB in frequency range of 10.3 −13.5 GHz and the maximum reflection loss was reach to −33.85 dB at 11.7 GHz when the coating thickness was 2.5 mm. Moreover, the electromagnetic analysis demonstrated that the electromagnetic loss properties and the electromagnetic impedance matching performance should be both satisfied to obtain the excellent microwave absorption properties. This study was meaningful to design the ferrite/carbon composites based microwave absorbers.

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1. Introduction

Microwave absorption materials (MAMs) have received worldwide interests due to their important roles on solving electromagnetic pollution problems in human daily life [1,2]. According to the microwave absorption mechanism, MAMs can be divided into magnetic loss type microwave absorbers and electric loss type microwave absorbers. Among these candidates, ferrites, including spinel ferrites and magnetoplumbite ferrites, are typical magnetic loss type microwave absorbers. They exhibit high complex permeability values because of their excellent nature resonance loss and magnetic hysteresis loss [3–5]. Meanwhile, carbon materials, including graphite, graphene, carbon fiber and carbon nanotube, are typical electric loss type microwave absorbers and present high complex permittivity values owing to their superior electric conductive loss and relaxation loss [6–8].

However, single microwave absorber is difficult to achieve well electromagnetic impedance matching, which needs a balance between complex permeability and complex permittivity. As a result, recent studies are focused on composite microwave absorbers, such as RGO/$CoFe_2O_4$ composites [9], Co_2Z hexaferrite-$BaTiO_3$ composites [10], $SrFe_{12}O_{19}/\alpha$-Fe composites [11], carbon nanotube-copper phthalocyanine/magnetite [12], (Z-type barium ferrite/silica) @polypyrrole composites [13], and so on. Most of them are concerned on the preparation method and the final microwave absorption properties. To further investigate the design principle and the microwave absorption mechanisms of electric/magnetic composite microwave absorbers, the flaky graphite/cobalt zinc ferrite composites were prepared in this study. The electromagnetic properties in the frequency range of 2–18 GHz were analyzed.

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Furthermore, the electromagnetic impedance matching performance, the microwave attenuation ability and the electromagnetic cancellation effect were discussed.

2. Experiment

2.1. Materials preparation

The flaky graphite/cobalt zinc ferrite composites were prepared by the coprecipitation method. Firstly, the necessary amount of flake-like graphite and 0.5 g sodium dodecyl benzene sulfonate were added into 200 ml deionized water. Then, the homogeneous graphite solution was obtained after ultrasonic dispersing for 1 h. Secondly, the necessary amount of $Zn(NO_3)_2 \cdot 6H_2O$, $Co(NO_3)_2 \cdot 6H_2O$ and $Fe(NO_3)_3 \cdot 9H_2O$ with the mole ratio of 1:1:4 were dissolved into the graphite solution. After magnetic stirring for 0.5 h, the as-prepared solution was moved into the water bath with the temperature of 70 °C. The precipitant NaOH was then dropped into the solution and the pH value was adjusted to about 9.0. The coprecipitation reaction occurred and the precipitate was formed after ageing for about 6 h. Then, the precipitate was washed twice by deionized water and ethanol. Finally, the as-prepared precipitate was dried at drying oven for 3 h and then calcined in muffle furnace at 700 °C for 2 h under nitrogen atmosphere. The heating rate was 3 °C/min. The flaky graphite/cobalt zinc ferrite composites were obtained after cooling to room temperature.

2.2. Measurement

The structures were characterized by X-ray diffraction (XRD, Rigaku D/Max-2500) and field emission scanning electron microscopy (FE-SEM, HITACHI S-4800). The electromagnetic parameters (ε', ε'', μ', μ'') in the frequency range of 2–18 GHz were measured by the Network analyzer (Agilent PNA 8363B). During this process, 40 wt% of paraffin and 60 wt% of flaky graphite/cobalt zinc ferrite composites were uniformly mixed, and then the as-prepared composites were pressed in a cylindrical mould to form the sample for the electromagnetic parameters test.

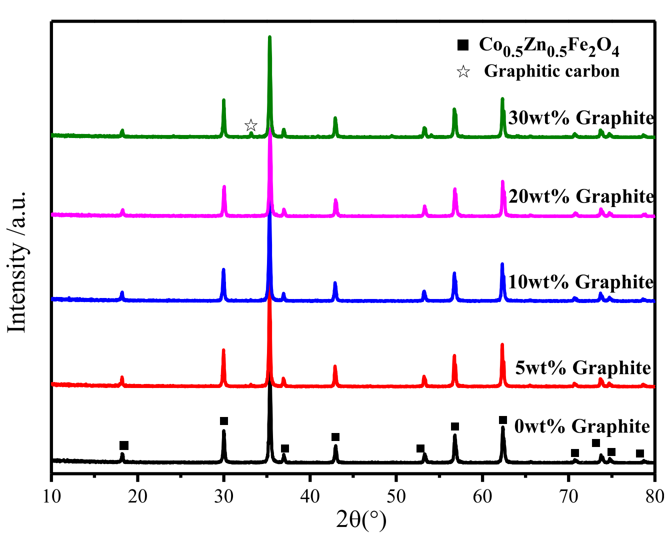

Fig. 1. XRD patterns of graphite/cobalt zinc ferrite composites with different graphite weight ratios.

3. Results and discussion

Fig. 1 shows the XRD patterns of as-prepared flaky graphite/cobalt zinc ferrite composites with different graphite weight ratios. It can be found that the samples showed pure cobalt zinc ferrite phase when the graphite weight ratio was less than 20 wt%. It is demonstrated that the cobalt zinc ferrite is well coated on the surface of flaky graphite because the X-ray can not transmit through the thick cobalt zinc ferrite shell-layer. However, the impurity graphitic carbon phase appeared at 33.2° when the graphite weight ratio is increased to 30 wt%. Similar graphitic carbon phase was also detected in the studies by Luo et al. [14] and Yu et al. [15]. It is because that the thickness of the cobalt zinc ferrite shell-layer will dramatically decrease with the increase of graphite weight ratio. Once the cobalt zinc ferrite can not completely coat the flaky graphite, the excessive graphite will be exposed outside and formed the typical graphitic carbon phase. Therefore, the optimal graphite weight ratio should be controlled to be less than 20 wt% to obtain the pure graphite/cobalt zinc ferrite composites.

The morphologies of the pure flaky graphite, pure cobalt zinc ferrite and graphite/cobalt zinc ferrite composites with different graphite weight ratios are measured by FE-SEM. As shown in Fig. 2 (a), the flake size of pure graphite is 5–20 μm and the thickness is about 200 nm. For the pure cobalt zinc ferrite, it can be found from Fig. 2 (b) that the particle size is about 70 nm and the particles are seriously agglomerated. After the flaky graphite is compounded with the cobalt zinc ferrite, it can be seen from Fig. 2(c) to Fig. 2(e) that the ferrite particles are assembled on the surface of flaky graphite. All the graphite/cobalt zinc ferrite composites exhibit flake-like shape. However, for the composite with 5 wt% graphite, most ferrite particles can not homogeneously distribute and then agglomerate on the surface of flaky graphite. As a result, it shows high flake thickness. Then, the ferrite particles can be uniformly distributed on the flaky graphite with the increasing graphite ratio. For the composite with 20 wt% graphite, the sample exhibits high aspect ratio. The flake size is about 5–20 μm and the thickness is about 300 nm.

Fig. 3 shows the electromagnetic parameters of as-prepared flaky graphite/cobalt zinc ferrite composites with different graphite weight ratios in the frequency range of 2–18 GHz. It can be seen from Fig. 3(a) and (b) that the real part and the imaginary part of complex permittivity are both dramatically increased with the increase of graphite weight ratio. It can be ascribed to the enhanced electrical loss properties of flaky graphite. It is well known that the carbon materials, including the flaky graphite and graphene, can conduct the conductive and dielectric loss [16–18]. As shown in Fig. 3(c) and (d), the real part and the imaginary part of complex permeability are nearly decreased with the increasing graphite weight ratio. It is because that the saturation magnetization from the cobalt zinc ferrite will reduce and then the complex permeability is firstly decreased. However, the imaginary part of complex permeability is increased when the graphite weight ratio is 20 wt%. It is known that the natural resonance frequency (f_r) can be expressed as $f_r = (\gamma H_a)/2\pi$, where γ is the gyromagnetic factor, H_a is the effective anisotropy field [19]. Deng et al. [20] and Ma et al. [21] suggested using the coercive force H_c to replace the effective anisotropic field. For the graphite/cobalt zinc ferrite composites with 20 wt% graphite, the coercivity is reinforced due to the introduce of second phase. Then, the higher coercivity is helpful to improve the natural resonance loss and then causes the increase of complex permeability when the graphite weight ratio is 20 wt%.

Then, the reflection loss can be calculated according to the transmission line theory [22].

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