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Optimization of electromagnetic matching of carbonyl iron/$BaTiO_3$ composites for microwave absorption

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ABSTRACT

Microwave absorbing materials filled with $BaTiO_3$ and carbonyl iron (CI) particles with various weight fractions ($BaTiO_3$/CI particles = 100/0 to 0/100) are investigated. The dielectric and magnetic properties of the absorbers can be tuned by changing the weight ratio of $BaTiO_3$/CI particles in the frequency range of 2–18 GHz. Numerical simulations are also performed to design a single-layer and double-layer absorber. The minimum reflection loss of the composite filled with 20 wt% $BaTiO_3$ and 60 wt% CI particles at 2.0 mm thickness can be reached to −42 dB at 4.1 GHz. With the weight ratio of CI particles in the composite increased, the microwave absorption peak shifted to the lower frequency region. By using a double-layer absorber structure, the microwave absorption performance of the absorber is enhanced. The result shows that the total thickness of the absorber can be reduced below 1.4 mm by using a matching layer filled with 50 wt% $BaTiO_3$, and an absorption layer filled with 60 wt% $BaTiO_3$ and 20 wt% CI particles, whereas the reflection loss below −10 dB can be obtained in the frequency range of 10.8–14.8 GHz and the minimum reflection loss of −59 dB can be obtained at 12.5 GHz.

1. Introduction

Recently, the demand for various kinds of microwave absorbers has increased in the frequency range of 2–18 GHz for their twofold use in the commercial and military applications [1]. Microwave absorbing materials are generally prepared by adding magnetic and/or dielectric fillers into a matrix to meet the demand of low reflection over a broad frequency range. Commonly, by controlling the characteristic and content of the fillers, the configuration, internal fractal structure and thickness of the composites, the absorption performance of these microwave absorbing materials can be optimized [2–7].

Among the magnetic candidates for microwave absorbers, carbonyl iron (CI) particles with thin plate shape whose thickness is controlled to be below skin depth which are particularly interesting [8]. With the advantages of higher saturation magnetization than ferrites and low eddy current loss coming from particle shape effect, a large value of magnetic permeability can be obtained in gigahertz frequencies above the Snoek's limit. Therefore, the CI particles filled polymer or rubber composites are widely being used for various applications, such as, magnetic memories, flexible magnets, microwave absorbers, and a multitude of other useful devices [9–12].

Barium titanate ($BaTiO_3$) as one of the perovskite-type structure and is widely used in multi-layer capacitors, optoelectronic devices and positive temperature coefficient, due to its high dielectric constant, good ferroelectric properties and nonlinear optical properties [13]. However, there have been few investigations concerning the $BaTiO_3$ used as microwave absorber. Abbas et al. [14] prepared $BaTiO_3$ filled polyaniline microwave absorbing composites, and the results showed a maximum reflection loss of −25 dB at 11.2 GHz in a composites thickness of 2.5 mm. Jing et al. [15] prepared the flake-shaped $BaTiO_3$ by a modified sol–gel method, and a maximal reflection loss of $BaTiO_3$/paraffin composite can be obtained −29.6 dB at 12 GHz with a matching thickness of 4 mm. These electromagnetic results indicate that although $BaTiO_3$ has a relatively high dielectric constant above 1000, the effective dielectric constant of the composite with high $BaTiO_3$ content still remains at a relatively low value of complex permittivity due to lower dielectric constant of the polymer matrix, and results in low microwave absorption or a larger matching thickness.

In spite of extensive research efforts, the prepared microwave absorber with high absorption in a broad frequency range still remains challenging. However, a few studies on the microwave absorbers made from both the $BaTiO_3$ and CI particles have been carried out. Here, we report the electromagnetic property of the microwave absorbing materials with a frequency range of 2–18 GHz, based on different weight ratios of $BaTiO_3$ and CI particles in epoxy matrix. It has also been indicated that the design of the microwave absorbers with multi-layer structures has a great effect on the absorption properties, and thus can be used to

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improve their microwave absorption [16,17]. A suitable matching layer, which can achieve a good impedance matching with the free space and allow an incident electromagnetic wave enter into the absorbing layer, is often employed for designing microwave absorber with a good absorption performance [18]. Therefore, the reflection loss of the microwave absorber with a different absorber weight content and composite thickness has been theoretically predicted based on the model of single-layer and double-layer absorbers backed by a perfect conductor.

2. Experimental

2.1. Materials

The CI particles used in this study were fabricated by the decomposition of $Fe(CO)_5$ and purchased from Xinghua Chemical Co. Ltd, Shaanxi Province, China. The main characteristics of the CI particles are: the content of α-iron >99.5 wt%, thin flakes of 1–5 μm in diameter and blow 1 μm in thickness, and a polycrystalline microstructure. Tetragonal-phase $BaTiO_3$ powders with a size smaller than 1 μm and a Ba/Ti mol ratio of 1.001 were synthesized using the standard solid state reaction technique and provided by Nantong Ao xin Electronic Technology Co. Ltd., JiangSu, China. The matrix used in this work is an epoxy resin and the cure agent is polyamide resin, which were supplied by XI AN Leeo Technological Co.

2.2. Sample preparation

In order to know the characteristic of the composite materials, samples containing different contents of $BaTiO_3$ and CI particles were manufactured and tested. We divided the composites by $BaTiO_3$/CI particles' weight fraction ratio that the electromagnetic property was changed from a dielectric material ($BaTiO_3$/CI particles = 100/0) to a magnetic material ($BaTiO_3$/CI particles = 0/100). A detailed list of samples is reported in Table 1. The $BaTiO_3$ and CI particles were separately dispersed into ethanol solutions via a high energy ultrasonic treatment for 30 min to yield homogeneous suspensions. Afterwards, the two suspensions were mixed together and treated via ultrasonic vibration for another 30 min to form a uniform suspension of the two fillers. Then the resin and hardener were added into the mixtures, followed by stirring at 2000 rpm for 10 min. The mixture was then left to dry at 70 °C to eliminate the ethanol solvent until the desired viscosity was reached. Finally, the mixtures were pre-cured at 90 °C for 30 min and then post-cured at 120 °C for 2 h.

2.3. Measurements

The complex permittivity $\varepsilon(f)$ and permeability $\mu(f)$ of the composites were measured using the *T*/*R* coaxial line method in the frequency range of 2–18 GHz by a network analyzer (Agilent Technologies E8362B: 10 MHz–20 GHz). The testing specimens had a cylindrical toroidal specimens: with an outer diameter of 7.0 mm and an inner diameter of 3.03 mm. The microwave absorption characteristics of the single-layer and double-layer were measured using a network analyzer (Agilent Technologies E8362B) by comparing between the signals transmitted by the samples, and reflected from its input. The antennas used for the measurements were ridged wideband horns covering the microwave region 2–18 GHz. First, each specimen was fabricated according to the experimental procedure. Second, the fabricated specimens were cut to the designed thickness. The specimen dimensions were 180 mm long × 180 mm wide with designed thickness, that was adhered to a 2 mm thick aluminum substrate.

3. Results and discussion

3.1. Electromagnetic properties of the microwave absorbing coatings

Interactions between absorber filled composites and electromagnetic fields in the microwave frequency band can be expressed by the complex permittivity ($\varepsilon^* = \varepsilon' - j\varepsilon''$) and complex permeability ($\mu^* = \mu' - j\mu''$). For the complex permittivity of the absorber, the real part of complex permittivity (ε') is proportional to the quantity of charge stored on the surface when the composite is under an applied electric field. The imaginary part of complex permittivity (ε'') accounts for the loss energy dissipative mechanisms in the composites [19–21]. Fig. 1 shows the complex permittivity of the Samples 1–6 as a function of frequency. For the Samples 1 and 2, as the $BaTiO_3$ weight content increased from 50% to 80%, both real and imaginary part of the complex permittivity increased in the frequency range of 2–18 GHz. The ε' of both Samples 1 and 2 remains almost invariable in the measured frequency range. The ε'' of the Sample 1 is almost lower and that of Sample 2 shows little change with frequency until 8 GHz and then increases with increase in frequency. The ε'' of both Samples 1 and 2 demonstrated that the dielectric loss of $BaTiO_3$ filled composite is almost low for the used microwave absorber, especially in the lower frequency range. Compared with Sample 2, the ε' of the Sample 3 is decreased when 20 wt% $BaTiO_3$ is replaced by 20 wt% CI particles. The results can contribute to the small $BaTiO_3$ particles filled composites that can form more interface between the filler/matrix at the same weight filler's content, and thus the higher complex permittivity can be obtained [22]. As the weight of $BaTiO_3$ decreased and the weight of CI particles increased in the composite, both the ε' and ε'' increased, and obtained a maximum values when the weight ratios of the $BaTiO_3$/CI particles is 0/100. As we know, when the filling fraction reaches the closest packing fraction, the high complex permittivity can be obtained due to the enhancement of the interfacial polarization, and the interactions between the fillers [23]. Obviously, both the $BaTiO_3$ and CI particles were included in an insulating resin matrix to constitute heterogeneous composites. The complex permittivity of the composite mainly resulted from the characteristic and content of both $BaTiO_3$ and CI particles. $BaTiO_3$ is a ferroelectric material exhibiting high resistivity, and CI particles is a ferromagnetic metal material. As the content of the CI particles with metallic behavior increased, the enhancement of

Table 1
Composition and notation of the manufactured and tested samples.

Specimen	$BaTiO_3$ (wt%)	CI particles (wt%)	Epoxy resin (wt%)	Polyamide resin (wt%)
1	50	0	40	10
2	80	0	16	4
3	60	20	16	4
4	40	40	16	4
5	20	60	16	4
6	0	80	16	4