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Review Articles

Microwave absorption properties of flake-shaped Co particles composites at elevated temperature (293–673 K) in X band

Guowu Wang, Xiling Li, Peng Wang, Junming Zhang, Dian Wang, Liang Qiao, Tao Wang *, Fashen Li

Key Laboratory for Magnetism and Magnetic Materials of MOE, Key Laboratory of Special Function Materials and Structure Design, Ministry of Education, Lanzhou University, Lanzhou 730000, People's Republic of China

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ABSTRACT

The complex permeability and permittivity of the easy-plane anisotropic Co/polyimide composite at high temperature (293–673 K) in X band were measured. The results show that both the complex permeability and permittivity increase with the increase of temperature in the measured temperature range. The calculated absorption properties display that the intensity of the reflection loss (RL) peak first increases and then decreases with the increase of temperature, and reaches the maximum (−52 dB) at 523 K. At each temperature, the composite can achieve the RL exceeding −10 dB in the whole X band. The composite can even work stably for more than 20 min with the excellent absorption performance under 673 K. In addition, the RL performance of the composite at high temperature is better than that at room temperature.

Contents

1. Introduction

Microwave absorbing materials have important applications in radar wave stealth and civil high frequency devices. The microwave absorption properties of absorbing materials are closely related to their electromagnetic parameter. Most of previous studies mainly focused on the electromagnetic parameter and microwave absorbing performance at room temperature. In recent years, some research groups have begun to pay attention to the electromagnetic parameter and microwave absorption performance of absorbing materials at elevated temperature [1–4]. According to the reported researches, most of research groups use pure dielectric materials with outstanding thermal stability, such as graphite, carbon nanosheets and SiC fiber, to prepare microwave absorbers and study their electromagnetic parameters and microwave absorption performance at high temperature [5–10]. It is reported that all the RL can exceed −10 dB in the whole X band at elevated temperature (573–873 K). However, the thickness of these absorbers is too thick and the minimum one is larger than 2.5 mm due to that the permeability of pure dielectric materials is equal to 1.

Previous researches have shown that magnetic microwave absorbing materials can process a wider absorption bandwidth and thinner thickness than pure dielectric materials. Therefore, In

* Corresponding author.
E-mail address: wtao@lzu.edu.cn (T. Wang).

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order to effectively reduce the thickness of the absorber applied at high temperature, the magnetic coating is advised to be used. However, the ferrite with excellent anti-oxidative properties is not suitable for high temperature application because of low Curie temperature. Recently, a research group reported that a kind of composite which is mixed by carbonyl iron and polyimide can achieve a good microwave absorbing performance at 573 K with a small thickness [11]. As we all know, iron can not be used at too high temperature due to the relatively low Curie temperature (1043 K) and instability at high temperature. Compared with Fe, magnetic Co is a more potential high-temperature absorbing material because of higher Curie temperature (1403 K) and better oxidation resistance [12,13]. In addition, a high permeability of Co particles composite is expected to be obtained to reduce the absorber thickness. Our previous work has shown that the high frequency properties of magnetic particles with easy magnetization plane comply with the following rule [14,15]:

$$f_r \cdot (\mu_i - 1) = \frac{\gamma}{4\pi} M_s \sqrt{\frac{H_\theta}{H_\varphi}}, \quad (1)$$

where H_θ and H_φ are out-of-plane and in-plane anisotropy field, respectively. It could be found that the $f_r \cdot (\mu_i - 1)$ can be much higher than conventional Snoek limit if $H_\theta \gg H_\varphi$ and this kind of material can keep a large permeability at GHz [16]. If the soft magnetic particles are transformed to thin flakes and the sample preparation process is properly controlled, the magnetic moments can lie in the flake plane and the easy magnetization plane can be obtained.

In this paper, irregular Co particles are chosen as the raw material and transformed into thin flakes with easy-plane anisotropy by ball milling. The composite mixed by ball-milled Co particles and polyimide is prepared and then the electromagnetic parameter and microwave absorption performance at high temperature are investigated.

2. Experiment

The irregular Co particles are chosen as the raw additive and converted into thin flakes by ball milling method in our laboratory. The prepared Co flakes are further oxidized with H_2O_2 to form a layer of cobalt oxide on the surface of Co flakes. The surfaced-oxidized Co powders were prepared by dispersing 4.0 g of Co powders into 40 mL of deionized water and then adding 15 mL of H_2O_2 with mechanical stirring [17]. The polyimide is chosen as binder and mixed with the surface-oxidized Co flakes under the condition of ultrasound. The mixed specimen is pressed to a rectangle flake with the length of 22.80 mm, width of 10.16 mm and thickness of 2 mm at 6 Mpa. Finally, it is heated to 513 K in argon environment and maintained for 2 h.

In order to prove that the prepared flaky Co particles are easy-plane anisotropy, the flaky Co particles/paraffin composite is prepared. Then it is heated to 353 K for 10 min, and oriented by a rotated external magnetic field in the melting state. The external magnetic field intensity is 1.5 T [18]. In addition, a composite with same volume concentration but without orientation is prepared as the contrast.

The phase and morphology of flake-shaped Co powder was analyzed by the X-ray diffractometer (XRD) and scanning electron microscope (SEM), respectively. The vibrating sample magnetometer (VSM) was used to measure the static magnetic properties of ball-milled Co powder at the elevated temperature. The electromagnetic parameters (complex permeability and permittivity) of the composite at high temperature environment were measured by means of the waveguide method using a vector network analyzer. The microwave absorption properties of the composite were calculated by the transmission line theory [19,20], which is described as follows:

$$Z = \sqrt{\frac{\mu_r}{\varepsilon_r}} tanh\left[j(2\pi ft/c)\sqrt{\mu_r \varepsilon_r}\right], \quad (2)$$

$$RL = 20\log\left|\frac{Z-1}{Z+1}\right|, \quad (3)$$

where RL is the reflection loss, μ_r and ε_r are the complex permeability and permittivity of composite, f is frequency of microwave, t is the thickness of absorber, c is the velocity of light.

3. Results and discussion

3.1. Morphology, phase and component

Fig. 1(a) shows the morphology of ball-milled Co particles prepared in our laboratory. After ball milling, the raw particles are converted into flakes with the diameter of 10 μm and thickness of about 1 μm. The XRD diffraction pattern of Co powder before and after ball milling is shown in Fig. 1(b). The results show that the raw Co powder is consisted of α-Co (hcp) and β-Co (fcc). After ball milling, a large number of defects are introduced into particles, and the crystallinity of the material decreases, causing the decrease in peak intensity. Fig. 1(c) shows the Co 2p photoelectron spectra from the surface-oxidized Co flakes. The results show that the surface of the powder is the oxide of Co and reveal that the oxidation layer is formed on the surface of the Co flake. Similar results have been found in our previous work [17].

3.2. The static magnetic properties at the elevated temperature

Fig. 2 shows the hysteresis loop of surfaced-oxidized Co powder measured at an argon atmosphere at different temperatures. At elevated temperature, the saturation magnetization are almost unaffected by temperature, which is mainly related to the high curie temperature of Co itself. In addition, Co exhibits a larger coercivity (*Hc*) compared with other soft magnetic materials because it has a large magnetocrystalline anisotropy for the hcp Co. As the temperature increases, we find the decrease of *Hc* shown in the inset of Fig. 2. This might come from the decreased magnetocrystalline anisotropy with the temperature increasing [21].

Fig. 3 shows the hysteresis loops of the surface-oxidized Co flakes/paraffin composite without and with rotated orientation. We know that the magnetic moment will be confined in the plane if the flake plane is easy magnetization plane. When this kind of material is oriented in an oriented magnetic field, the flakes will rotate and all flakes are parallel to each other and arrange along the direction parallel to the external magnetic field. For the orientation sample, the applied magnetic field is parallel to the oriented flake planes when the hysteresis loop is measured. From Fig. 3, it is obvious that the sample is more easily magnetized to saturation, demonstrating that the Co flake is an easy-plane anisotropy material.

As shown in Fig. 1(b), the ball-milled thin Co flakes consists of α-Co (hcp) and β-Co (fcc). β-Co has small magnetocrystalline anisotropy. The demagnetization field can spur the magnetic moment to lie in the flake plane. However, α-Co has a large axial magnetocrystalline anisotropy and the magnetic moments tends to array along the c axis. For Co particles with hcp structure, the magnetocrystalline anisotropy field is defined as:

$$H_k = \frac{2K_1}{Ms}, \quad (4)$$