Journal of Magnetism and Magnetic Materials 375 (2015) 100–105

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Greatly enhanced microwave absorbing properties of planar anisotropy carbonyl-iron particle composites

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ARTICLE INFO

Article history:
Received 20 March 2014
Received in revised form
26 July 2014
Available online 16 September 2014

Keywords:
Carbonyl-iron particles
High frequency
Permeability spectra
Natural resonance

ABSTRACT

This paper investigates the high permeability and high resonance frequency characteristics of carbonyl-iron particle composites at microwave range. It finds that the key factor to keep both high microwave absorbing characteristics is the planar anisotropy. The effective permeability of planar anisotropy carbonyl-iron particles/nonmagnetic matrix composition in high frequency is measured and calculated. In contrast to the sphere shaped particles with no planar anisotropy, the permeability and resonance frequency of flake particles are greatly enhanced by introducing the planar anisotropy, and the permeability can be further enhanced by using a rotational orientation method to get higher planar anisotropy. As the use of the planar anisotropy, the flake soft magnetic particles increase the natural resonant frequencies so as to lead the higher real part and the imaginary part of the permeability in a broadband range. The resonance peak of flake particles is simulated by using the combination of the Landau–Lifshitz–Gilbert equation and Bruggeman's effective medium theory, considering the correction of shape factor. Our theory simulation agrees well with the experimental data.

1. Introduction

The microwave properties of the ferromagnetic metal-based materials have potential for microwave application [1,2]. A composite magnetic material with high permeability and high resonance frequency characteristics at microwave range is required for miniaturizing electronic components [3]. The connection between the permeability and resonance frequency has a long illustrative history which was starting with Snoek who showed that the product of the initial permeability and the resonance frequency was proportional to the saturation magnetization [4]:

$$(\mu_s - 1) f_r = \frac{4}{3}\gamma M_s \tag{1}$$

This is understood as the result of the precession of magnetization under uniaxial anisotropy field, the permeability is proportional with resonance frequency and inverse proportional with static permeability. Both the extent of such precession and the loss of energy are small, in Fig. 1(a). Base on the uniaxial anisotropy, planar anisotropy picture has been developed [5–7], in Fig. 1(b), the amplitude of precession is relatively bigger, which is ascribed to the correction of Snoek's constant with the increase of the ratio $(\sqrt{H_\theta/H_\phi})$. Smaller H_ϕ with larger H_ϕ can get higher permeability and higher resonance frequency, where H_ϕ is the out-of-plane anisotropic field and H_ϕ is the in-plane anisotropy field.

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

The planar anisotropy ferromagnetic particles have been attracting considerable interest in experiment. Two types of planar anisotropy can be identified: intrinsic and external. Intrinsic planar anisotropy includes Co_2Z planar hexaferrites and planar rare earth magnets [7–9]; external planar anisotropy includes flakes and thin film systems in which the term of planar anisotropy is used as the large demagnetization field of planar shape particle and the out-of-plane anisotropic field corresponds to the demagnetization field [10–12]. Walser et al. and Li Fashen et al. have suggested that the use of a planar anisotropy in oblate spheroid increases the natural ferromagnetic resonant frequencies (> 1 GHz) and exceeds the Snoek's limit [6,13,14]. Kim et al. and Zhang et al. have also found that the flake-shaped iron particles with planar anisotropy have higher permeability than sphere-shaped particles in the frequency range of 0.5–6 GHz and 2–18 GHz [15,16]. Li et al. have done a research about the permeability in microwave band by Maxwell-Garnett effective medium theory, their work indicates that there exists an upper-limit of real permeability which is related to the shape factor N_d. If the shape of particles is modified from spherical

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http://dx.doi.org/10.1016/j.jmmm.2014.09.015

to flaky, the permeability can be significantly increased due to the reduced N_d [17,18]. In the most previous works, attentions were focused on reducing shape factor to improve permeability. In order to improve the high frequency complex permeability and to understand the physical mechanism of intrinsic and external planar anisotropy, it is important to develop a uniform picture planar anisotropy to investigate permeability and resonance frequency at the same time.

In our present work, the frequency dependent permeability of the planar anisotropy carbonyl-iron particles (PACI)/nonmagnetic matrix composition is calculated by the combination of the Landau–Lifshitz–Gilbert equation and Bruggeman's effective medium theory. We make a choice of carbonyl-iron particles due to their higher magnetization. In comparison, the microwave properties of composites filled with the sphere shaped carbonyl-iron particles (SSCI) and orientation PACI are presented. The good agreement of theoretical calculations of the effective permeability and experimental results indicates the validity of our planar anisotropy approach.

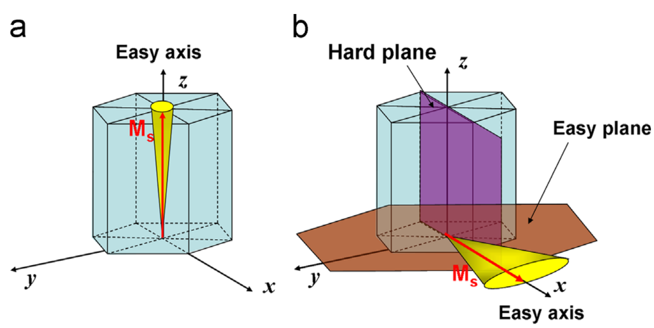

Fig. 1. Scheme of the precession of magnetization under the uniaxial anisotropy field (a) and planar anisotropy field (b).

2. Experimental details

In this study, the raw material was purchased from Tianyi super-fine metallic powder Co. Ltd. Jiangsu province. The raw powders were SSCI particles and milled on a planetary ball mill with 500 r/min for 8 h to obtain PACI particles. In ball milling process 50 ml *n*-hexane was added as the process control agent. The ball-to-powder weight ratio was 25:1.

The composite samples were prepared by mixing the particles with paraffin wax with 20% volume concentration of the particles. The mixture was then pressed into toroidal shape with an outer diameter of 7 mm, an inner diameter of 3.04 mm, and a length of 1.5–3.0 mm for microwave measurement. For the oriented PACI sample, the mixture was treated at a high temperature (80 °C) for 30 min in a disk-shaped compact. Then the compact was rotated around its axis in a perpendicular magnetic field of 1.5 T until the paraffin solidifies. Thus, the flake planes of PACI particles are parallel to each other.

The complex permeability of the composite samples was obtained using an Agilent E8363B vector network analyzer in the 0.1–18 GHz range. The magnetic properties of milled particles were measured using a vibrating sample magnetometer (VSM) (Lakeshore 7304 model) with a maximum magnetic field of 7 kOe. The morphology and particle size of samples was analyzed by scanning electron microscope (HITACHI S-4800) and Laser particle-size-distribution tester (JL-1177). The planar anisotropy property of the as-milled particles was characterized by room temperature transmission Mössbauer spectra. In the transmission geometry, the incident γ-ray is parallel to the axis of the oriented disk.

Fig. 2 shows the morphological difference of SSCI and PACI particles. The SSCI have a spherical shape with a size distribution of 1–10 μm, in Fig. 2(d) the volumetric mean diameter of the particles is found to be about 1.937 μm. With the ball milling, the

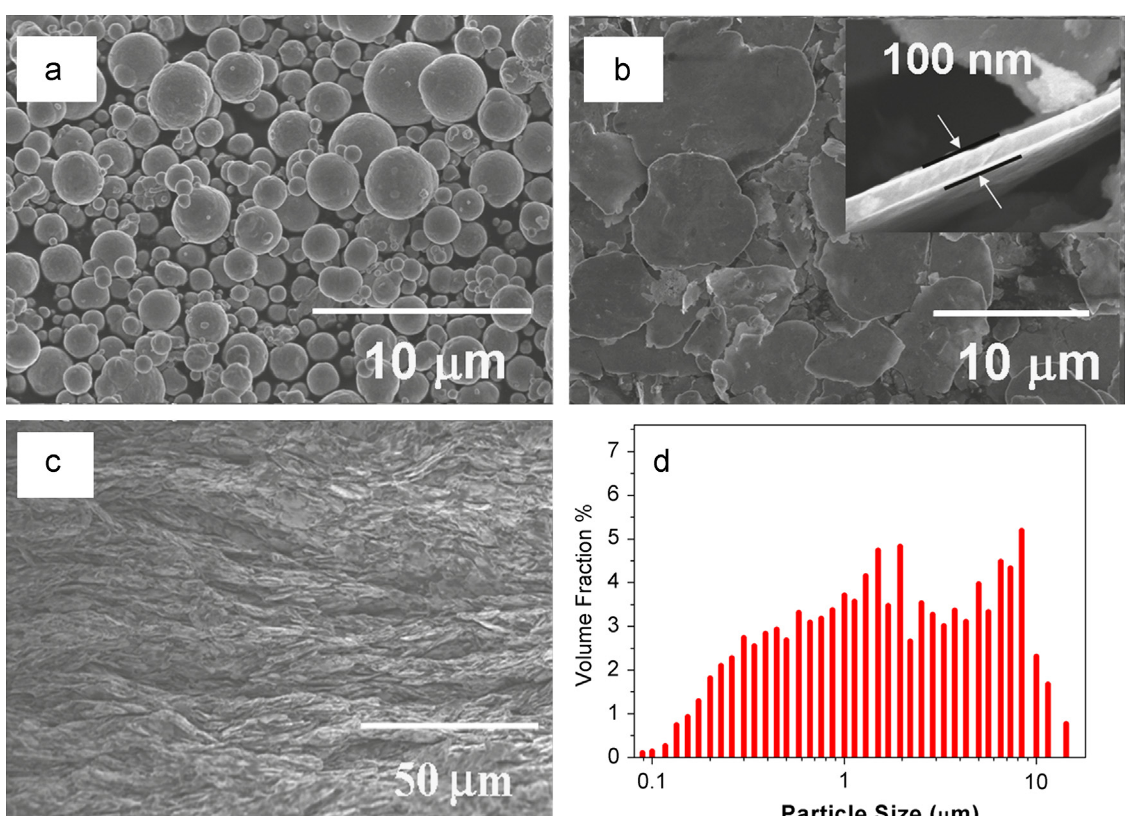

Fig. 2. SEM images of the SSCI (a), PACI (b), fractured cross-sections of the oriented PACI (c) and particle size distribution of SSCI (d).