Journal of Magnetism and Magnetic Materials 324 (2012) 519–523

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Extrinsic permeability of Fe-based flake composites from intrinsic parameters: A comparison between the aligned and random cases

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ARTICLE INFO

Article history:
Received 31 December 2009
Received in revised form
13 May 2011
Available online 27 August 2011

Keywords:
Effective permeability
Landau–Lifshitz–Gilbert equation
Maxwell–Garnett mixing rule
Aligned orientation
Random orientation

ABSTRACT

Composites consisting of aligned ferromagnetic flakes embedded in a nonmagnetic matrix are the focus of attention as a kind of promising microwave material. In this paper, the effective permeability for composites consisting of nanocrystalline Fe-based flakes with aligned and random spatial orientation distribution of inclusions for comparison is measured in gigahertz. Flakes were coated with thin silica layer to reduce permittivity and improve impedance match. It is found that the magnetic loss in flakes is mainly caused by the natural ferromagnetic resonance and exchange resonance. To explore the unsolved problem of calculating effective permeability for the aligned case, the combination of Landau–Lifshitz–Gilbert equation and Maxwell–Garnett mixing rule with important modification is used to predict the effective permeability spectrum of composites. An excellent agreement with the experiment results indicates the validity of proposed theoretical approaches.

1. Introduction

Magnetic composites consisting of magnetic particles embedded in nonmagnetic matrix have been widely used as absorbing materials. Among these materials, nanocrystalline ferromagnets, such as iron, nickel, cobalt and their alloy particles show very promising microwave absorption owing to high saturation magnetization and Snoek limit [1,2].

Nowadays, absorption application potential of magnetic composites has been improved by flake shaping, and aligned orientation, due to enhanced permeability such as K.S. Lee's work [3]. The surface of flakes is coated with dielectric materials to drop the permittivity and improve impedance match. He has successfully fabricated flexible magnetic composite sheets containing aligned $Fe_{85}Si_{9.5}Al_{5.5}$ flake particles coated with resistive film of 377 Ω/sq. On the one hand, the thickness of absorber resulting from high permeability and permittivity can be reduced. On the other hand, due to wave impedance matching, the reflection loss can be reduced apparently. X.K. Zhang et al. have carried out the relevant research [4]. From these points of view, it is necessary for us to pay attention to the theoretical calculation of high frequency of composites complex permittivity and permeability with shape and alignment considerations.

However, in recent theoretical calculations, attention was focused on the effective permeability of composites consisting of random flakes. The theoretical model of the effective permeability for the case of aligned orientation has been rarely reported so far. Difficulties include the following: 1. the lack of proper term in effective medium equations to present the effect of alignment and 2. errors or incorrect model introduced during shaping the intrinsic spectrum of inclusion. Therefore, it is important to calculate the extrinsic effective permeability from the intrinsic parameters of constituent materials, a more reasonable methodology to effective parameter prediction.

In the present paper, both the aligned and random case have been involved to test the calculation techniques of the intrinsic and effective permeabilities. The Landau–Lifshitz–Gilbert equation has been widely used to calculate the permeability spectra of magnetic materials in gigahertz [5]. Some modifications on the Maxwell–Garnett mixing rule have been made by taking into account of the percolation threshold (f_c) and the interactions between particles to give a more accurate result of effective permeability. The good agreement between theoretical results and experimental data demonstrates the validity of our proposed approaches.

2. Experiment

The preparation of Fe-based nanocrystalline flake particles has been described in Ref. [6]. These particles are coated with a very thin amorphous silica layer using the sol–gel technique. The composites consisting of aligned flakes are made by the following process: (1) Fe-based flakes are mixed with rubber matrix at volume fraction 35%; (2) the mixture is heated to the softening temperature and (3) they are extruded in a two-screw extruder. The third step can be omitted for the operation procedures of the

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doi:10.1016/j.jmmm.2011.08.032

random oriented flake composites. Finally, the mixture is pressed into toroidal shape in a mold with an outer diameter of 7 mm, inner diameter of 3.04 mm and thickness less than 4 mm for microwave measurement.

Microstructure and composition of powders are characterized by Philips X Pert Pro x-ray diffractometry (XRD). According to the Scherrer formula, the average crystal size of Fe-based flakes is determined to be about 15 nm. The magnetic hysteresis loop is measured using a vibrating sample magnetometer (VSM) at room temperature. Saturation magnetization (M_s) is 190 emu/g and coercive force (H_c) is 156 Oe. The real and imaginary values of the effective permeability and permittivity of composite sample are examined over the frequency range of 2–18 GHz by the reflection and transmission method using an Agilent 8720ET vector network analyzer.

The morphology, size and orientation of samples are analyzed by transmission electron microscopy (TEM) and scanning electron microscopy (SEM), as shown in Fig. 1. Fig. 1(a) exhibits the flakes coated by thin silica layer with thickness around 5 nm. The fracture surface and cross section images of flake/rubber composites with toroidal shape clearly demonstrate that most of the flakes are aligned according to Fig. 1(b) and (c). The flakes have a width limited to the range from 1 to 8 μm [Fig. 1(c)]. As exemplified in Fig. 1(d), the flakes embedded in the mixture are randomly oriented and the thickness is around 0.2–0.3 μm, which is less than the skin depth reported to be about 1–2 μm [7]. We have randomly chosen 100 flakes with different thicknesses and particle sizes in order to derive the average values of width and thickness, respectively, by virtue of “smile view” software, which is provide by JEOL. It is found that the average value of particle size is about 3.6 μm; the mean thickness of particles is estimated to be near 0.24 μm. Accordingly, the average aspect ratio of particles is 15.

3. Theoretical calculation

3.1. Theoretical formula of intrinsic permeability

The microwave magnetic loss of ferromagnetic materials mainly originates from eddy current, natural ferromagnetic resonance and exchange resonance [2]. Since the thickness of as-prepared particle is far less than the skin depth of Fe-based particulates, the eddy current loss effect can be neglected [8]. It means that the magnetic loss mechanism of our flakes is mainly natural resonance and exchange resonance in the range of 2–18 GHz. Hence, the Landau–Lifshitz–Gilbert equation can be used to calculate the intrinsic permeability spectra [5,9]:

$$\frac{d\overline{M}}{dt} = -\gamma(\overline{M}\times\overline{H}) + \frac{\alpha}{M_s}\overline{M}\times\frac{d\overline{M}}{dt}, \quad (1)$$

where $\overline{M}$ represents the magnetization vector; γ is the gyromagnetic factor; α is the damping coefficient, which needs to be determined from experimental data; M_s is the saturation magnetization and $\overline{H}$ is the total magnetic field vector.

Aharoni’s exchange resonance modes were evidenced to be very effective for nanocrystalline particles [2]. Therefore, the total magnetic field includes in-plane magnetocrystalline anisotropy field (H_a), exchange field (H_0), demagnetization field and microwave magnetic field (h) for flakes. Here, we set the non-geometrical effective anisotropy magnetic fields H_{eff} as $H_{eff}=H_a+H_0$. Although materials of two different compositions are used in Ref. [5] and our paper, they are all ferromagnetic nanocrystalline materials with similar microstructure and size. Hence, the single domains (flakes) are considered in the calculation of effective permeability.

A simplified diagram shown in Fig. 2 represents the action between h and effective in-plane anisotropy field (H_e) in a single

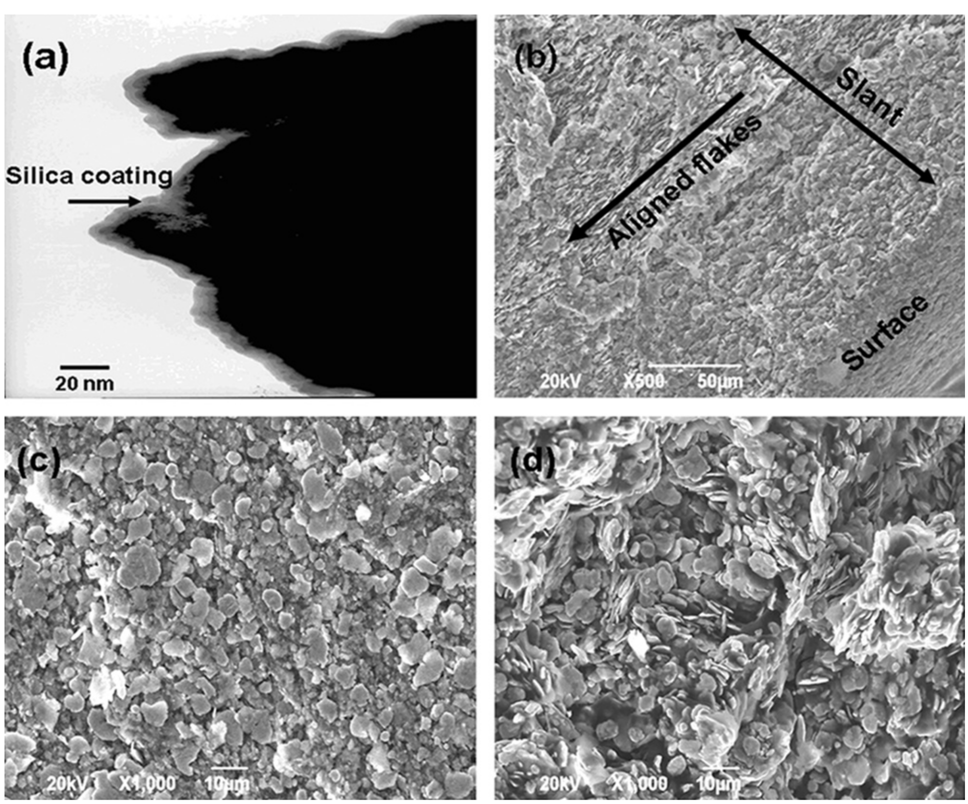

Fig. 1. (a) TEM graph of single flake-shaped particle with the silica coating layer, (b) SEM micrographs observation of fracture surface of toroidal composite consisting of aligned flakes, (c) SEM micrographs observation of cross section of toroidal composite consisting of aligned flakes and (d) SEM micrographs observation of cross section of toroidal composite consisting of random flakes.