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Particle-size distribution modified effective medium theory and validation by magneto-dielectric Co-Ti substituted BaM ferrite composites

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ABSTRACT

This letter reports an extended effective medium theory (EMT) including particle-size distribution functions to maximize the magnetic properties of magneto-dielectric composites. It is experimentally verified by Co-Ti substituted barium ferrite ($BaCo_xTi_xFe_{12-2x}O_{19}$)/wax composites with specifically designed particle-size distributions. In the form of an integral equation, the extended EMT formula essentially takes the size-dependent parameters of magnetic particle fillers into account. It predicts the effective permeability of magneto-dielectric composites with various particle-size distributions, indicating an optimal distribution for a population of magnetic particles. The improvement of the optimized effective permeability is significant concerning magnetic particles whose properties are strongly size dependent.

1. Introduction

Magneto-dielectric composites, in the form of magnetic particles dispersed in an electrically insulating matrix, are candidate materials for next generation microwave communication devices [1–4]. Such magneto-dielectric materials require high permeability and low loss, which are closely related to the particle size of magnetic fillers. Size effects on intrinsic magnetic properties, such as saturation magnetization, permeability and coercivity, of both metallic particles [5,6] and ferrite particles [7–9] have been extensively studied. In order to improve the magnetic performance of magneto-dielectric composites, optimization of the particle size and its distribution of magnetic fillers is necessary, especially in the nanoscale range where the magnetic properties of the particles are strongly size-dependent [10–12].

2. Theory

Developed from averaging the microscopic physical quantities of constituents, the effective medium theory (EMT) provides descriptions of the macroscopic properties of composite materials. Traditional EMT models like the Maxwell Garnett formula [13] and the Bruggeman formula [14] are not able to incorporate size-dependent parameters into account. According to the original Bruggeman formula, if N isotropic phases, with spherical geometry and permeability μ_i, form a mixture, each occupying a total volume fraction F_i ($\sum_{i=1}^{N} F_i = 1$), the effective permeability μ_{eff} is given by

$$\sum_{i=1}^{N} F_i \frac{\mu_i - \mu_{eff}}{\mu_i + 2\mu_{eff}} = 0. \quad (1)$$

This summation formula assumes a size-independent permeability or a particle-size distribution in the form of the Dirac delta function for each kind of inclusion. If the inclusion i has a nonnegligible size-dependent permeability $\mu_i(D)$ and a particle-size distribution function $p_i(D)$ in terms of particle diameter D, the volume-fraction distribution function $f_i(D)$ of the inclusion i is calculated as

$$f_i(D) = F_i \frac{p_i(D) D^3}{\int_0^{+\infty} p_i(D) D^3 dD}. \quad (2)$$

By replacing the total volume fraction F_i with the volume-fraction distribution function $f_i(D)$ for each kind of inclusion, the modified Bruggeman formula including the particle-size distribution function, which is applicable to a wide range of inclusion volume fraction, is given in an integral form as

$$\sum_{i=1}^{N} \int_0^{+\infty} f_i(D) \frac{\mu_i(D) - \mu_{eff}}{\mu_i(D) + 2\mu_{eff}} dD = 0. \quad (3)$$

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Log-normal distribution is often used as a good approximation for particles produced by attrition by random impacts, such as crushing, grinding and milling. In terms of particle size D, the log-normal distribution $p_{LN}(D)$ is given by

$$p_{LN}(D) = \frac{1}{D\sqrt{2\pi\sigma^2}} e^{-\frac{(\ln D - \varepsilon)^2}{2\sigma^2}}. \quad (4)$$

The two parameters ε and σ can be easily evaluated from the median $M = D_{50}$ and the span $S = (D_{90} - D_{10})/D_{50}$, where D_x stands for the diameter below which x% of the particle diameters are found [15], i.e.,

$$\varepsilon = \ln(M),\ \sigma = \frac{1}{1.2816} \ln\left(\frac{S + \sqrt{S^2 + 4}}{2}\right). \quad (5)$$

Based on the extended Bruggeman formula, the effective permeability of magneto-dielectric composites with log-normal particle-size distributions can be predicted relying on the median and the span of the distributions.

3. Experimental

Experimental verification adopted Co-Ti substituted BaM hexaferrite, i.e., $BaCo_xTi_xFe_{12-2x}O_{19}$ (x = 1.15), synthesized from reagent grade $BaCO_3$, Co_3O_4, TiO_2, and Fe_2O_3 by solid-state reaction method. The starting reagents were mixed thoroughly for 4 h in deionized water (DI water) in a ball mill. The mixture was dry-pressed in a stainless steel die, and calcined for 5 h at 1200 °C in air. The samples were then pulverized and milled for 7 h. The resultant powders were uniaxially dry-pressed again and sintered for 4 h at 1200 °C in air. Finally, the samples was crushed and sorted into different particle-size distributions by sieves denoted by #200, #230, #270, #325, #400, #450, #500, and #635 mesh sizes. A series of composite samples were prepared by introducing the powders for each particle-size distribution in a wax matrix at 40% volume concentration. The mixtures were then pressed into toroids with an outer diameter of 17.86 mm, an inner diameter of 10.59 mm, and a thickness of ~3 mm.

The microstructure of the sorted particles was observed by a scanning electron microscope (FEI Scios DualBeam). The particle-size distributions were measured by a laser diffraction particle size analyzer (HORIBA LA-960). The permeability spectra of the composite samples were measured by an impedance analyzer (Keysight E4991B) from 100 MHz to 1 GHz with the use of a 16454A test fixture.

4. Results and discussion

Fig. 1 shows the SEM micrographs of the sieved $BaCo_{1.15}Ti_{1.15}Fe_{9.7}O_{19}$ particles at each particle-size distribution. A $-x/+y$ notation is used to define the group of particles which pass through the #x mesh screen but retained by the #y mesh screen. Decreasing average diameter and narrowly distributed particle size are observed. More precise particle-size distributions are measured by the laser diffraction method. The distribution curves are plotted on a logarithmic scale of particle diameter as Fig. 2, indicating that a suitable classification into different particle-size distributions was achieved. The median of the sorted particles varies from 7.0 to 84.1 μm.

Fig. 3(a) shows the permeability spectra of the $BaCo_{1.15}Ti_{1.15}Fe_{9.7}O_{19}$/wax composite samples measured by the impedance analyzer from 100 MHz to 1 GHz. Using the inversion of the Bruggeman formula, the intrinsic permeability of the $BaCo_{1.15}Ti_{1.15}Fe_{9.7}O_{19}$ particles are obtained as a function of frequency and plotted as Fig. 3(b). The intrinsic permeability increases monotonically with particle diameter below 850 MHz, but no longer at higher frequencies, and it reaches a maximum at about 500 MHz for all finely sorted groups of particles expect for the −635 group.

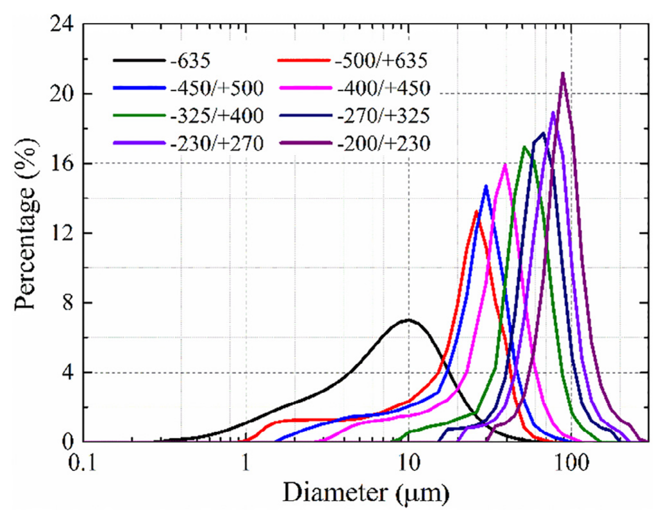

Fig. 2. Particle-size distributions of the $BaCo_{1.15}Ti_{1.15}Fe_{9.7}O_{19}$ powders.

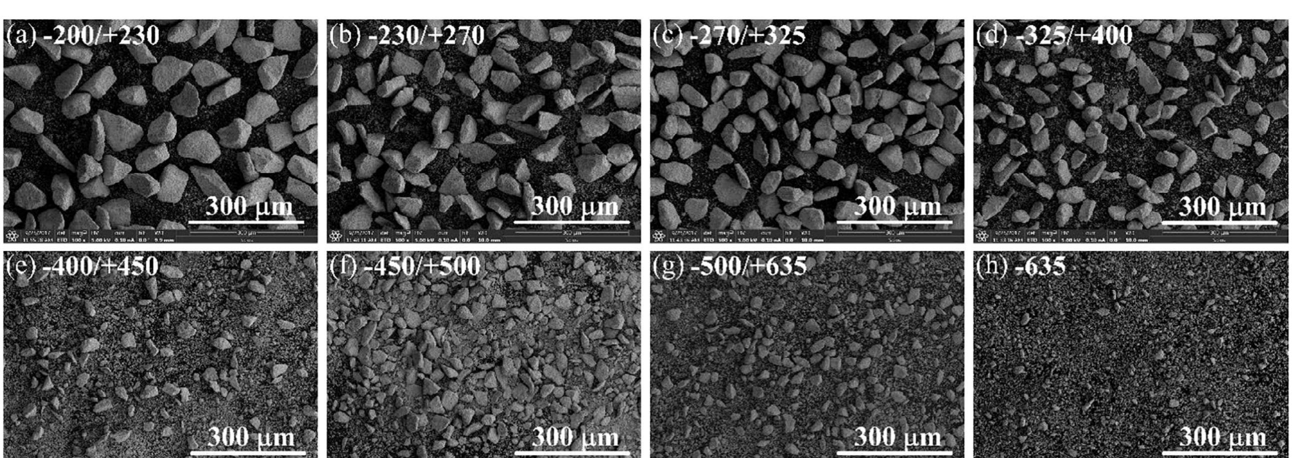

Fig. 1. SEM micrographs of the $BaCo_{1.15}Ti_{1.15}Fe_{9.7}O_{19}$ particles sorted by the mesh size of (a) −200/+230, (b) −230/+270, (c) −270/+325, (d) −325/+400, (e) −400/+450, (f) −450/+500, (g) −500/+635, and (h) −635.