Contents lists available at ScienceDirect



Journal of Magnetism and Magnetic Materials

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



Effect of Mg²⁺ and Ti⁴⁺ dopants on the structural, magnetic and high-frequency ferromagnetic properties of barium hexaferrite



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ARTICLE INFO

Article history: Received 27 February 2015 Received in revised form 27 July 2015 Accepted 23 August 2015 Available online 8 September 2015

Keywords: Hexaferrite Mössbauer spectroscopy Magnetic properties Ferromagnetic resonance

ABSTRACT

The doped barium hexaferrite, $BaFe_{12-x}(Mg_{0.5}Ti_{0.5})_xO_{19}$ with $1 \le x \le 5$, is synthesized by a solid state ceramic method. Its crystalline structure, morphology, as well as static and dynamic magnetic properties are investigated by X-ray diffraction (XRD), scanning electron microscopy (SEM), vibrating sample magnetometry, and vector network analysis, respectively. The cation distribution of Mg^{2+} and Ti^{4+} in the hexagonal structure of $BaFe_{12-x}(Mg_{0.5}Ti_{0.5})_xO_{19}$ is investigated by ⁵⁷Fe Mössbauer spectroscopy. The effect of Mg^{2+} and Ti^{4+} dopants on static and high-frequency magnetic properties of the ferrite is studied.

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1. Introduction

Hexagonal ferrites have attracted the attention of researchers and engineers since they were discovered. These materials are widely utilized as magnetostatic and electromagnetic devices. The M-type ferrite, with the crystal structure similar to that of mineral Magnetoplumbite, and the composition $BaFe_{12}O_{19}$, is the best known ferrite [1]. It has a large magnetocrystalline anisotropy, stabilized by the strong exchange interactions among the Fe ions. By substituting Fe³⁺ with other ions, its magnetocrystalline anisotropy can be greatly reduced, and even switched from the *c*-axis to the basal plane. An extensive work has been done to modify the magnetic parameters of M-type hexaferrites by substituting Fe³⁺ with other cations or cation combinations such as Ce, Al, Ga, Cr, Co-Ti, Ru-Ti, Mn-Cd-Zr, Zn-Nb, Cr–Ga, Co–Zr, and so forth, as recently published [2–11]. It should be emphasized that in previous work on the Mg-Ti substituted barium hexaferrite [12–16], the effects of only individual concentrations of dopants (mostly in a narrow *x* range) have been studied. Therefore, this paper attempts to study in detail the structural and magnetic characteristics, and high-frequency ferromagnetic properties of $BaFe_{12-x}(Mg_{0.5}Ti_{0.5})_xO_{19}$ as a function of the substituted amount of dopants in a broad range $(1 \le x \le 5)$.

2. Experimental

2.1. Preparation of ferrite powders

BaFe_{12-x}(Mg_{0.5}Ti_{0.5})_xO₁₉ (x=0-5) powders were prepared from barium carbonate (BaCO₃), iron oxide (Fe₂O₃), magnesium oxide (MgO), and titanium oxide (TiO₂). The starting materials were mixed in a planetary mill for 5 h and sintered in air at 1250 °C for 1 h. Finally, the sintered ferrites were crushed again in a mill for 18 h to obtain powders.

2.2. The preparation of composite samples

To measure the complex permittivity and permeability using a vector network analyzer, the powdered ferrite was added to paraffin, dissolved in toluene and vibrated for 5 min in an ultrasonic bath. It was then kept at 80 °C for 10 h to remove the toluene from the mixture and after that, it was cooled to room temperature. The as-prepared composite samples (containing 70 wt% of the ferrite and 30 wt% of paraffin) were measured in the form of the standard rings (outer diameter: 7 mm, inner diameter: 3.04 mm, thickness: 2 mm) and rectangles (with two various sizes – length: 10.66 mm, width: 4.31 mm, thickness: 2 mm and length: 7.11 mm, width: 3.55 mm, thickness: 1.5 mm).

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2.3. Structural characterization (XRD, SEM)

XRD patterns of the powder samples were recorded in the range of 15–80 (2 θ) degrees using a X'Pert Pro MPD, PANalytical, diffractometer with Cu K α radiation (λ =1.5406 Å). The characteristic lattice parameters, *a* and *c*, were calculated according to the following formula [17]:

$$\frac{1}{d\frac{2}{hkl}} = \frac{4(h^2 + hk + k^2)}{3a^2} + \frac{l^2}{c^2},\tag{1}$$



Fig. 1. XRD patterns of the as-prepared doped barium ferrites $BaFe_{12-x}(Mg_{0.5}Ti_{0.5})$ (x=1-5).

where *h*, *k* and *l* are Miller indices and *d* is interplanar spacing as determined by the Bragg formula, $2d \sin \theta = n\lambda$. The unit cell volume was calculated from the lattice parameters *a* and *c* using the following formula [18]:

$$V_{\text{cell}} = \frac{\sqrt{3}}{2}a^2c.$$
 (2)

The X-ray density, ρ_x , of the material was calculated according to the relation:

$$\rho_x = \frac{2M}{N_a V_{\text{cell}}} \tag{3}$$

where M represents the molar mass of the sample, and N_a is Avogadro's number.

Electron micrographs were taken using a Philips XL30 scanning electron microscope (SEM) with the accelerating voltage of 25 kV. The particle size of the samples was determined from the SEM imaging.

2.4. Mössbauer spectroscopy

⁵⁷Fe Mössbauer spectra of the samples were taken in transmission geometry at 293 K. A ⁵⁷Co/Rh γ-ray source was used. The velocity scale was calibrated relative to ⁵⁷Fe in Rh. *Recoil* spectral analysis software [19] was used for the quantitative evaluation of the Mössbauer spectra. A Lorentzian line width of 0.274 mm/s, resulting from the fit of the spectrum of BaFe₁₂O₁₉ was chosen for the fit of the spectra of the substituted hexaferrite, BaFe_{12-x}(Mg_{0.5}Ti_{0.5})_xO₁₉ (*x*=1-5). The distribution of the ferric cations over the five non-equivalent cation sublattices provided by the hexagonal ferrite structure was calculated from the Mössbauer subspectral intensities, assuming the same recoilless fractions of the Fe³⁺ cations on all available crystal sites.



Fig. 2. a) XRD patterns of the Mg–Ti substituted barium hexaferrites in the 2θ range of $32-34.5^{\circ}$. b) The lattice parameter, *c*, and c) the volume of the unit cell, *V*, for BaFe_{12-x}(Mg_{0.5}Ti_{0.5})_xO₁₉ (x=1-5) vs. dopant content.

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