



Structural and magnetic properties of praseodymium substituted barium-based spinel ferrites



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ARTICLE INFO

Keywords:

Spinel ferrites
X-Ray diffraction
Sem
Susceptibility
Y-K angles

ABSTRACT

In this research work $\text{BaPr}_x\text{Fe}_{2-x}\text{O}_4$ ($x = 0.0, 0.025, 0.05, 0.075, 0.10$) spinel ferrites have been synthesized successfully by sol-gel technique. X-ray diffraction analysis confirmed the fcc spinel phase of the synthesized samples. All the samples showed inhomogeneous grain size distribution observed through scanning electron microscopy (SEM). Temperature dependence normalized AC susceptibility and Curie temperature studies revealed that BaFe_2O_4 exhibited multidomain (MD) structure with high Curie temperature. On the other hand, multidomain to single domain (SD) transitions occurred when the praseodymium is substituted into barium spinel ferrites. Narrow loops showed the soft nature of ferrites and it was confirmed from magnetic properties of the prepared samples. Decreasing trend of saturation magnetization and remanence was observed with the substitution of praseodymium contents. Coercivity and anisotropy constant both enhanced with the praseodymium concentration. The above-mentioned parameters revealed that the synthesized spinel ferrites might be useful for the of high-density magnetic recording applications.

1. Introduction

Based on the structural properties, ferrites or ferrimagnetic oxides are mainly divided into spinel, garnet and hexagonal ferrites. Spinel ferrite is the most widely studied structure of the ferrites family. Spinel ferrites have many potential applications, including power conditioning, electronics, bio processing, and magnetic resonance imaging enhancement [1]. High values of electrical resistivity and low eddy current losses make spinel ferrites usable for microwave frequency application [2]. Generally, the chemical formula of a cubic spinel ferrite is written as MFe_2O_4 , where M represents the divalent metal ion, such as Ba^{2+} , Zn^{2+} , Fe^{2+} , Mg^{2+} , Ni^{2+} , Cd^{2+} , and Cu^{2+} [3–6]. Spinel ferrite has face centered cubic structure (fcc) with eight formula units for each unit cell. Oxygen ions or anion arrangement in the lattice helps to determine the spinel crystal structure of ferrite.

Barium spinel ferrites have extensive applications in a variety of fields, including microwave and switching devices [7]. These properties can be enhanced by substituting specific rare-earth (RE) ions into the interstitial sites of lattices. Many researchers reported the change in

structural and magnetic properties with the substitution of these RE elements. RE materials normally change the strains and structural spins of the ferrite material [8]. Therefore, a minor addition of RE cations, alters the structural as well as electromagnetic properties [9]. Previous studies have also revealed the change in ferrites properties with the substitution of Pr, Tb, Dy ions [10–12]. It is evident that Fe–Fe interaction (spin coupling effect of 3d electrons) can affect the resistivity and magnetic properties of ferromagnetic oxides. Therefore by incorporating RE ions into the spinel lattice or RE–Fe interactions (3d–4fcoupling) would also leads to change in both the electrical and magnetic behavior of ferrites. Consequently, it is established that different RE metals have different effects on ferrites.

In the present work, Sol-gel method has been used to determine the effect of minor substitution of Pr^{3+} on structural and magnetic properties of BaFe_2O_4 spinel ferrites.

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2. Experimental procedure

2.1. Samples preparation

Spinel ferrites with composition $\text{BaPr}_x\text{Fe}_{2-x}\text{O}_4$ ($x = 0.0, 0.025, 0.05, 0.075, 0.10$) were successfully synthesized by Sol-gel process followed by auto-combustion technique. Initially, measured quantities of analytical grade $\text{Ba}(\text{NO}_3)_2$ and Fe_3Cl_2 were dissolved in 100 ml of de-ionized water. The Pr_2O_3 (99.99% pure) was first dissolved in HCl to get praseodymium chloride and then mixed with the solution. In this process citric acid was used as a chelating agent. Homogeneity of the precursor's solution was achieved by stirring and heating at $\approx 80^\circ\text{C}$. One molar NH_3 solution was added drop wise into the main solution so the pH is maintained at 7. After that a viscous gel is achieved by 8 h continuous stirring of the solution. The product was then subjected to self-combustion at 370°C for 3 h. This leads towards a fluffy product that was grounded and then sintered in a furnace at 700°C for 5 h. A load of 30 kn of Paul-Otto Weber Hydraulic Press was used to obtain fine pellets of 0.13 cm diameter and 0.14 cm to 0.26 cm thickness. Polyvinyl alcohol (3–5 wt.%) was used for the binding purpose of powdered ferrite material. The binding material was evaporated when the pellet samples were annealed at 250°C for 1 h, followed by 7 h sintering at 950°C .

2.2. Characterization of the samples

The structural characterization of the synthesized materials were done by using XRD diffractometer (Bruker axis D8 operating at 40 kV and at 30 mA) in the 2θ range of 15° to 80° . $\text{CuK}\alpha$ as a radiation source was used. Scanning Electron Microscopy (SEM) (Hitachi S4160) was used to observe morphology and microstructure of the spinel ferrites. Room temperature magnetic properties like the saturation magnetization M_s , remanence magnetization M_r and the coercivity H_c were measured by using the Vibrating Sample Magnetometer (VSM) (Model BHV-50 of Riken Danish Company Limited Japan). The applied magnetic field H ranged from 0.0 kOe to 2.0 kOe. Other parameters like magnetic anisotropy constant K_1 , squareness ratio M_r/M_s and Y-K angles are calculated on the basis of the observed data from hysteresis loop.

3. Results and discussions

3.1. X-Ray diffraction analysis

The X-ray diffraction patterns of $\text{BaPr}_x\text{Fe}_{2-x}\text{O}_4$ ($x = 0.0, 0.025, 0.05, 0.075, 0.10$) spinel ferrites are shown in Fig. 1. It can be observed from Fig. 1, that all the diffraction peaks for samples ($x \leq 0.05$) correspond to cubic spinel structure. Peaks become lower and broader with the

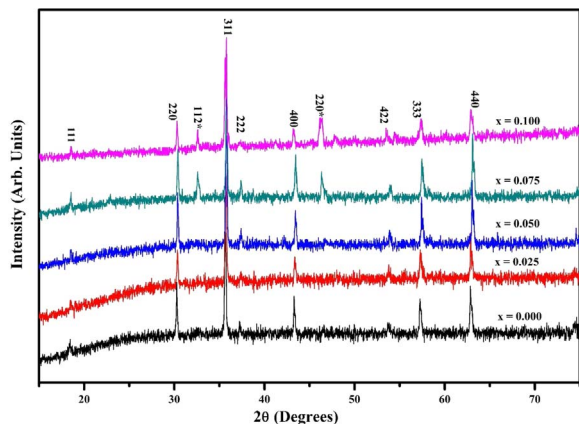


Fig. 1. X-Ray Diffraction patterns of $\text{BaPr}_x\text{Fe}_{2-x}\text{O}_4$ ferrites.

Table 1

Summary of results of various properties of $\text{BaPr}_x\text{Fe}_{2-x}\text{O}_4$ ($x = 0.0, 0.025, 0.05, 0.075, 0.10$): lattice constant (a), volume (V), measured density (ρ_m), X-ray density (ρ_x), porosity (P), average ionic radii of A-site and B-site r_A and r_B , Bond Lengths A-O, B-O. Jump Lengths (L_A and L_B) and Crystallite size D.

Composition	x = 0.000	x = 0.025	x = 0.050	x = 0.075	x = 0.100
Lattice constant 'a' (Å)	8.351	8.373	8.417	8.436	8.449
Volume (Å ³)	582.392	587.007	596.309	600.357	603.137
D_m (gm/cm ³)	5.37	5.415	5.48	5.52	5.58
D_x (gm/cm ³)	4.28	4.3	4.33	4.35	4.37
P (%)	20.29	20.59	20.98	21.19	21.68
r_A (Å)	0.45804	0.46281	0.47233	0.47644	0.47926
r_B (Å)	0.73775	0.74325	0.75425	0.759	0.76225
A-O (Å)	1.80804	1.81280	1.82233	1.82644	1.82926
B-O (Å)	2.0878	2.09325	2.10425	2.109	2.11225
Jump Length (L_A) (Å)	3.61609	3.62562	3.64467	3.65289	3.65852
Jump Length (L_B) (Å)	2.95252	2.96029	2.97585	2.98257	2.98716
D (311) nm	50.43	47.24	43.9	38.98	37.86

substitution of praseodymium ions. The diffraction peaks become lower and broader with the substitution of Pr^{3+} ions. It may be due to the poor crystallization accompanied the slower growth of crystallite. The appearance of the second phase (PrFeO_3) is observed for $x \geq 0.075$. The peaks of secondary phase correspond to $2\theta = 32.347^\circ$ and 46.384° with hkl values (112) and (220). These peaks were indexed by JCPDS card no. ICDD# 00-019-1012 for PrFeO_3 . The appearance of the secondary phase is mainly due to the high reactivity of Fe^{3+} ions with Pr^{3+} ions on the grain boundary. The second phase is orthorhombic and having insulating behavior [11,12].

The XRD data has also been used to calculate the crystallite size (D), bulk density (ρ_b), X-ray density (ρ_x), lattice constant (a) of the spinel ferrite and values are listed in Table 1. By putting the lattice constant 'a' in Standley's equations, ionic radii of A-site and B-site (r_A , r_B), as well as bond lengths of tetrahedral (A-O) and octahedral (B-O) sites, were calculated.

$$r_A = \left(\mu - \frac{1}{4} \right) a \sqrt{3} - r \quad (1-a)$$

$$r_B = \left(\frac{5}{8} - \mu \right) a - r \quad (1-b)$$

$$A - O = \left(\mu - \frac{1}{4} \right) a \sqrt{3} \quad (2-a)$$

$$B - O = \left(\frac{5}{8} - \mu \right) a \quad (2-b)$$

Where 'a' is the lattice constant; r (O^{2-}) is the radius of oxygen ion (1.35 Å); μ is the oxygen ion parameter, for ideal spinel ferrite $\mu = \frac{3}{8}$ [13]. Hopping lengths in tetrahedral sites (L_A) and in octahedral sites (L_B) which are nothing but the distance between the magnetic ions have been calculated using the following relations [13]

$$L_A = a \frac{\sqrt{3}}{4} \quad (3-a)$$

$$L_B = a \frac{\sqrt{2}}{4} \quad (3-b)$$

All the above mentioned calculated quantities are also given in Table 1.

A gradual increase in the lattice constant "a" with the increment of Pr^{3+} ion is shown in Fig. 2. This increase in the lattice constant can be credited to the larger ionic radius of Pr^{3+} ions as compared to that of Fe^{3+} ions. The larger Pr^{3+} ions (ionic radius ≈ 1.013 Å) partially

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