Effect of doping on structural and dielectric properties of Barium hexaferrite

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Abstract

Pr-Co doped barium hexaferrites, having the general formula $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3) have been synthesized by citrate precursor technique. Powder X-ray diffractometer, Scanning electron microscope (SEM) and diectric measurement instrument were used for the characterization of the prepared samples. X-ray diffraction confirms all the samples exhibits single hexagonal phase with space group $P6_3/mmc$. . It is observed that the lattice parameter (a) and (c) decreases with the substitution. The unit cell volume also shows the same pattern. The average grains size was found to vary with Pr-Co substitution. The dielectric constant and dielectric loss were examined as a function of frequency. There is decrease in dielectric constant at lower frequencies and constant behavior at higher frequencies which is a general trend of ferrites.

Keywords: Hexaferrite, SEM, Dielectric constant

1. Introduction

The field of ferrite materials, due to their potential applications and interesting physics involved in it, substituted with different cations and prepared by different techniques have recently attracted the focus of condense matter community.. In particular, hexaferrites show interesting magnetic and dielectric properties in its nanocrystalline form compared to those of micron sized bulk particles. Among the hexaferrites, M-type hexaferrite ($AFe_{12}O_{19}$, A = Ba, Sr, Pb) have been a subject of interest for several decades [1]. . Hexaferrites, due to their low cost, easy manufacturing and efficient significance in the area of electronics and telecommunication industry are useful in radiofrequency circuits, reflection coils, high quality filters, antennas, transformers etc [2-3]. Thus, it is important to study the dielectric behaviour at different frequencies in order to get valuable information of various kind and amount of additives required to obtain high quality materials for practical applications [4].. The dielectric properties of hexaferrites may

effectively vary in wide ranges by doping with cations that define their use in various devices and instruments. Dielectric properties can be improved by elemental substitutions to Ba^{+2} or Fe^{+3} sites or both. . Brahma et al. [5] worked on the Sb_2O_3 doped $BaFe_{12}O_{19}$ and observed that real and imaginary part of dielectric constant are varied by the substitution of Sb_2O_3.

Keeping all the previous studies in mind, we in the present study, made an attempt to investigate the effect of Pr-Co substitution on the structural and dielectric properties of barium hexaferrite. The obvious relevance of the study is to analyse whether the dielectric constant of these materials can be increased compared with the pristine. In addition, to the best of our knowledge, no systematic information is available in the literature on the dielectric properties of Pr-Co substituted barium hexaferrite. Therefore, such an investigation is expected to contribute significantly to our current understanding of the Pr-Co substitution in barium hexaferrite. The observed results are presented, and the effect of Pr-Co substitution on barium hexaferrite dielectric properties is presented in this paper.

2. Experimental details

M-type Pr-Co substituted barium hexaferrite material have been prepared by citrate-precursor method using analytical grade chemicals namely ferric nitrate nona-hydrate , prasodimium nitrate hexa-hydrate, Cobalt nitrate hexa-hydrate, barium nitrate, citric acid anhydrous. Metal salts and citric acid were dissolved in deionised water separately and then mixed together at room temperature with continuously constant stirring. Citric acid anhydrous acts as a fuel. The pH of the solution was adjusted at 6.5 by using ammonia (25%). The obtained solution was heated at 90 °C till a gel solution was obtained. The gel thus obtained was heated till combustion, ultimately leaving only loose ashes. These ashes were grinded by a motor pestle arrangement for about 30 min. The whole powder was then heated at 500°C to remove the organic moiety. Finally the powder was calcinated at 950 °C for 3 h.

The crystalline phase of the synthesised samples were determined by the D8 Advance Bruker X-ray diffractometer with CuKα (λ=1.5406 Å) radiation. The morphological studies of these samples were carried out by scanning electron microscope (SEM) (JOEL scanning Microscope, Model JSM-6490LV) operating at voltage of 25 kV. The dielectric measurements of the prepared samples was made by using Agilent 4285A precision LCR meter as a function of frequency of the applied ac field in the range of 20 Hz to 1 MHz

3. Results and Discussions

3.1 XRD Analysis:

The X-ray powder diffraction patterns for the $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x=y=0, 0.1, 0.3) are shown in the Fig.1. The diffraction peaks corresponding to planes (110), (107), (114), (203), (205), (217), (2011), (220) match well with the standard pattern for M-type barium hexaferrite. This suggests that the prepared material belongs to hexagonal structure with the space group $P6_3/mmc$. The lattice parameters of all the prepared samples were calculated and are shown in Table 1. It is observed that the lattice parameter (a) and (c) decreases with the substitution. The unit cell volume also shows the same pattern.
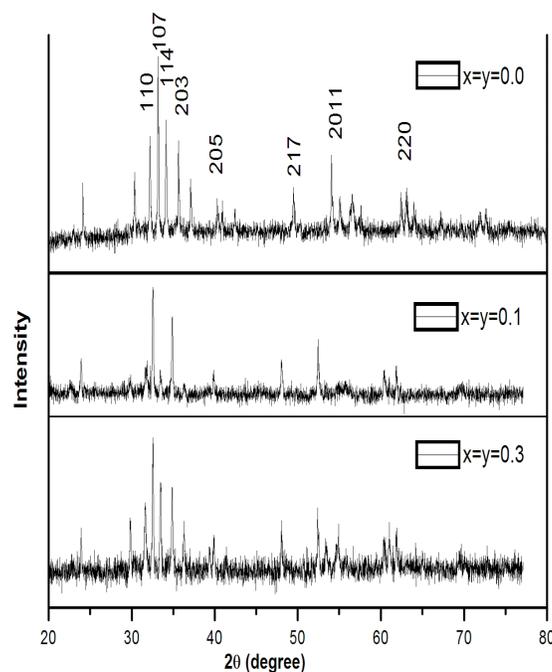

Fig 1.X-ray powder diffraction patterns for the $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3)

Table I: Lattice parameters, unit cell volume and grain size of $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3) samples

Composition $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$	a (Å)	c (Å)	$V_{cell}(Å^3)$	Grain size (nm)
x=y=0	5.86	23.16	795.30	104.65
x=y=0.1	5.85	23.10	790.53	236
x=y=0.3	5.84	23.08	787.15	176.37

3.2 SEM analysis

Fig.2. shows SEM images of $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3) compounds and the morphological shows the well-defined grains for $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3) compounds; however, moderately agglomerated particles are present in the Pr-Co doped $BaFe_{12}O_{19}$ samples. The average grains size was found to vary with Pr-Co substitution. The average grain size measured for $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3) Compounds are shown in table 1.
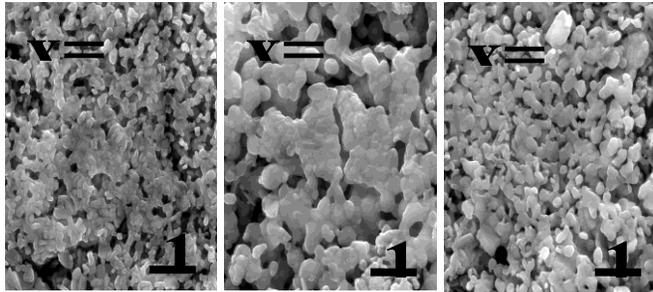

Fig.2.   SEM images of $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3)

3.2 Dielectric Studies

The dielectric constant is in the complex form in an ac field and is given by

$$\varepsilon = \varepsilon' - j\varepsilon''$$

Where ε' is the real par and ε'' is imaginary part designating the stored and dissipated energy respectively. Fig. 3 shows the frequency dependence of real part of dielectric constant of $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3) in an ac field ranging from 20 Hz to 1MHz. In fig.3. there is decrease in dielectric constant at lower frequencies and constant behavior at higher frequencies. This behavior may be attributed to the changes in valence states of cations and space charge polarization resulting from dipoles creation and higher dielectric constant at lower frequencies is associated with heterogeneous conduction in composites [6]. The constant behavior in dielectric constant at higher frequencies indicates the inadequacy of electric dipoles to follow the variation in frequencies due to alternating applied electric field [7].

From Fig. 3 it is observed that dielectric constant decreases with the substitution of Pr-Co at any particular frequency. The possible reason for this behaviour can be attributed to the fact that there is an increase in the resistance of grain thereby decreasing the probability of electrons reaching the grain boundary.
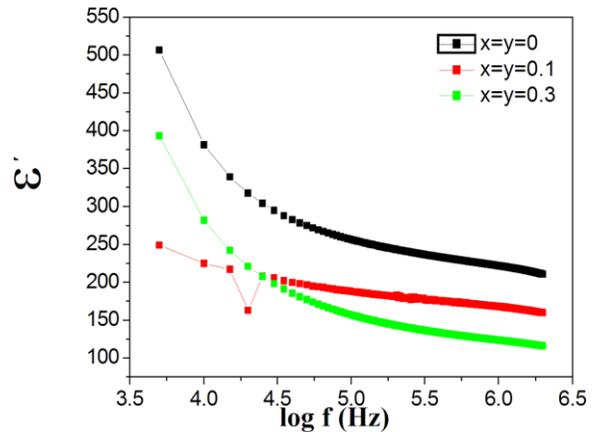

Fig.3. The frequency dependence of dielectric constant of $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3)

The dielectric loss is a measure of lag in the polarization with respect to the applied alternating field. Variation of dielectric loss of $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3) as a function of frequency is shown in Fig. 4. It is observed that for a particular concentration of dopant, dielectric loss decreases with the increasing frequency and is described using Koop's model [8]. The low frequency domain consequences the higher resistivity (due to grain boundaries), therefore acquisition of higher energy forwards the mobility of electrons between ions resulting higher energy loss. Similarly the higher frequency region corresponds to low resistivity (due to grain) and smaller energy loss takes place.
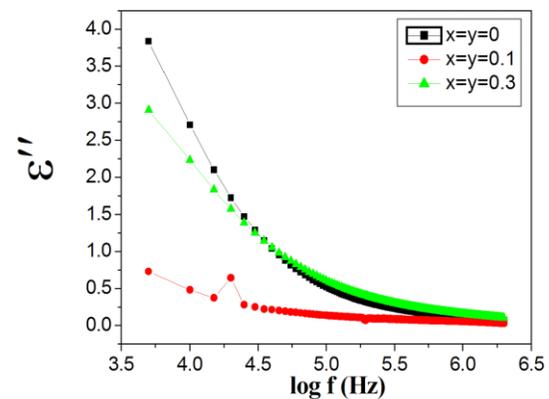

Fig.4. The frequency dependence of dielectric loss of $Ba_{1-x}Pr_xFe_{12-y}Co_yO_{19}$ (x,y = 0, 0.1,0.3)

The composition x = 0.1 exhibit the loss peak. The peaking nature occurs when the jumping frequency of electrons between Fe^{2+} and Fe^{3+} is equal to the frequency of the applied ac field

4. Conclusions

Pr-Co doped barium hexaferrite were synthesized by citrate precursor method. The effect of Pr-Co doping on the structural and dielectric properties of the prepared material were examined.. Phase formation was confirmed by X-ray diffraction. The lattice parameter (a) and (c) decreases with the substitution. The unit cell volume also shows the same pattern. The average grains size was found to vary with Pr-Co substitution. The dielectric constant and dielectric loss decreases with doping. The measurements of dielectric constant and dielectric loss with frequency suggest that the conduction in materials is similar to the conduction in ferrites and occurs due to polaron hopping.

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