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Effect of temperature on structure and dielectric, electrical properties of Al substituted BaY, Y-type Hexaferrites

U.V.Rathod[1], S.V.Soni[2], C.L.Khobaragade[3]

[1]Department of Applied Physics, Dr.BabasahebAmbedkar College of Engineering and Research, Nagpur ,
uvr123rathod@gmail.com
[2]Department Applied Physics, Yeshwantrao Chavan College of Engineering, Nagpur
s_sanjaysoni@rediffmail.com
[3]Department Applied Physics, Govindrao Wanjari College of Engineering and Technology, Nagpur,
chanduparulk@gmail.com

Abstract — *In this paper structural, dielectric and electrical studies of Y-type hexaferrite (BaY) with Al substitution, obtained by procedure of chemical co-precipitation method annealed at temperatures 1100°C for 4 and 6hrs and 1000°C for 6hrs, is presented. The X-ray diffraction (XRD) analysis of the prepared sample confirmed the formation of single-phase Y-type hexaferrites with space group R-3m having a parameter of hexagonal crystal structure. SEM micrograph of the material shows a uniform distribution of nano sized grains throughout the surface of the samples. Detailed study of the Electrical and dielectric properties of BaY, investigated in a wide frequency range (100 Hz to 10 MHz) at room temperature by Wayn Kerr Impendence Precision Analyzer 6500B, showed that these properties are strongly dependent on frequency and on the added level of the impurity.*

The effect of temperatures and time on the properties of BaY hexaferrite was also studied.

Keywords*- Y type hexaferrites; Bulk density; X-ray density; Porosity; Chemical co-precipitation technique; XRD.*

I. INTRODUCTION

Ferrite has attracted considerable attention of the researchers and industries because of its broad technological application in a wide range of frequencies extending from radio frequency to microwave range [1]. Low cost, easy manufacturing, and interesting electrical and magnetic properties make polycrystalline hexaferrites one of the most demanding materials today [2]. Polycrystalline hexaferrites have very good dielectric and electrical properties that depend on several factors such as processing conditions, sintering temperature and time, chemical composition and substitution of different ions [3,4]. Y-type ferrite is constructed from basic units of hexagonal barium M ferrite [5] and cubic spinel ferrites, which retain a hexagonal structure, usually with the direction of magnetization parallel to the c-axis. The classical chemical co-precipitation method for preparing $BaMe_2Fe_{12}O_{22}$ (where Me selected as trivalent substitute) ferrite requires a high calcination temperature, which induces aggregation of the particles. Many physical properties of polycrystalline ferrites are very sensitive to the microstructure. The bulk (grain) and grain boundary are the two main components that determine the microstructure. Thus, the information about the associated physical parameters of the components that constitute the microstructure is important in understanding the overall properties of the materials [6]. The spinal ferrites can be used only upto 3 GHz frequency range, but the hexaferrittes can be used in the whole region due to their intrinsic uniaxial anisotropic property. In this work, the dielectric and electrical properties were studied over a wide range of frequency (100 Hz to 10 MHz) at room temperature by the Wayn Kerr Impendence Precision Analyzer 6500B on BaY hexaferrite added with Al substitution annealed at temperatures 1100°C for 4 and 6hrs and 1000°C for 6hrs. The study of the dielectric and electrical properties in the broad range of frequency is important in view of its applications for components in microwave and RF circuits.

II. EXPERIMENTAL METHODS

Y-type hexagonal ferrite powder with composition of $BaAl_2Fe_{12}O_{22}$(BaY), was prepared by the procedure of chemical co-precipitation method. The nitrates with analytical reagent grade were used as starting material. For the preparation of BaAl hexaferrites, stoichiometric amounts of Barium Nitrate $Ba(NO_3)_2.6H_2O$ (99.9%, Sigma Aldrich, Germany), Aluminum Nitrate $Al(NO_3)_3.9H_2O$, Ferric Nitrate($Fe(NO_3)_3.9H_2O$ (99.9%, Sigma Aldrich, USA), were taken and dissolved one by one in 100 ml of de-ionized water . Ammonia solution (30% w/v) was added slowly in the mixture to adjust $_P$H of 8 and verified by µpH-system (362, Systronics). The mixed solution was stirred by the magnetic stirrer (REMI– speed 2MLH) for two hours at room temperature and kept for aging up to 24 hours. The Barium Aluminum hexaferrites precipitates were separated in a centrifuge machine rotated at 2500 rpm for 20 minute .The prepared precipitates were washed in 1:1 mixture of methanol and acetone followed by 100% de-ionized water to remove the effective impurities. The precipitates were dried at 100^oC for 24 hours in oven and then calcinated at 1100^oC for 6 hours, 4 hours and then at 1000^oC for 6 hours separately with increasing rate of heating 100^oC per hours into the furnace and cooled down to room temperature at the same rate. Before to get desired final products having divalent and trivalent valency $BaAl_2Fe_{12}O_{22}$ hexaferrites, the resultant material were grinded for several hours in pestle mortal and obtained homogeneous nano-sized powder of the synthesized material [7]. Polyvinyl acetate (PVA, 10 vol.%) used as binder to

reduce the brittleness and by applying a pressure of 10 tones psi using hydraulic press for 15 to 20 minutes, pellets were prepared using dye punch of 16.3mm diameter. These pellets are slowly heated in an air atmosphere in the furnace at 300^{o}C for about 3 hours and slowly cooled at a rate of 5°/min to room temperature to remove the binder concentration. The pellets were weighed and densities were measured.
To characterize the phase structure, the resulting powders were investigated by X-ray diffraction (XRD) technique at room temperature with a powder diffractometer PW3071/60, XPERT-PRO system with Cu-Kα radiation (λ=1.54060Å) with scan step of 0.013^0/s which was operated at 40kV and at 30mA. The 2θ Bragg angles were scanned over a range of 10.01^o to 119.98^o. The single phase formation of the hexagonal structure is confirmed from the study of powder X-ray diffraction (XRD). The output data extracted from Rietveld refinement [8] was used to calculate the average crystalline particle size. The analysis of the crystallite size (d_{hkl}) was determined using the Debye-Scherrer's equation (Eq. (1)),

$$d_{hkl}=k.\lambda/[\beta.\cos(\theta)] \tag{1}$$

where K is the shape coefficient (value between 0.9 and 1.0), λ is the wave length (1.5406 Å), β is the full width at half maximum (FWHM) in radians of each phase and θ is the diffraction angle.
The surface morphology of the sintered pellets was studied using a scanning electron microscope (SEM) (ZEISS ULTRA 55 FESEM). The pellets were polished using fine emery paper to make their faces smooth and parallel. The pellets were finally coated with conductive silver painted and dried at 150°C for 2 h. The dielectric and electrical properties were measured using the data generated from a computer-controlled impedance analyzer (Wayn Kerr Impendence Precision Analyzer 6500B) as a function of frequency (100 Hz to 10 MHz) at room temperature (T = 300 K).The value of dielectric constant (ε') has been calculated using the formula:

$$\varepsilon'=Cp.t/(\varepsilon_o.A) \tag{2}$$

where $\varepsilon_o=8.85 \times 10^{-12}$ F/m, known as permittivity of free space, t is the thickness of pellets, A is the area of cross section of pellets, and Cp as capacitance of the pellets. The complex dielectric loss has been calculated from the relation:

$$\varepsilon''=\varepsilon'.\tan\delta \tag{3}$$

The dielectric loss tangent (tanδ) has been calculated from the relation:

$$\tan\delta=1/2\Pi f.\varepsilon_o.\varepsilon'.\rho \tag{4}$$

where ρ is electrical resistivity calculated from the observed data.
The ac part of the electrical conductivity has been calculated from the relation

$$\sigma_{ac} = \varepsilon_o.\varepsilon'\omega.\ \tan\delta \tag{5}$$

where ω is angular frequency.

III. RESULTS AND DISCUSSION

A. X-ray Analysis

Fig. 1(a, b,c) shows the XRD pattern for the calcined powder at1100^{o}C for 6 hours, 4 hours and then at 1000^{o}C for 6 hours of $BaAl_2Fe_{12}O_{22}$ Hexaferrites. The nature of XRD patterns shows the formation of the isolated phase BaY. All the diffraction peaks were indexed by applying a hexagonal crystal system and space group *R-3m*. Planes (0 0 12), (1 1 0), (1 0 14), (1 1 6), (1 1 8), (2 0 6), (0 2 10), (0 0 22), (2 1 7), (3 0 0), (2 1 13), (2 1 15), (2 1 20) have been identified in agreement with the reported hexagonal crystal structure [9,10]. The Rietveld refinement confirmed the formation of a hexagonal crystal structure. Lattice constant ***a*** and ***c*** of Barium Aluminum Hexaferrites has been calculated using equation (6) as shown below.

$$1/d^2_{(hkl)} = [4(h^2+hk+k^2)/(3a^2)] + l^2/(c^2) \tag{6}$$

where h, k, l are miller indices and d is inter planer distance. The unit cell volume V of Hexagonal crystal is obtained from equation (7) as shown below.

$$V = a^2c\ \sin(120^0) \quad . \tag{7}$$

Bulk density plays a key role in analysing the ferrite materials and in the present study it was calculated by using equation (8) as shown below.

$$d_B = m/(\Pi.r^2h). \tag{8}$$

where m, r and h are the mass, radius and thickness of the pellets respectively. The values of bulk density for all the sample are listed in Table 1.It is seen that bulk density shows a varied trend by changing temperature and the time in Y-type hexaferrites. X-ray density of all the samples were also measured by using equation (9) as shown below [11].

$$d_x = 3.M/(N_A.V). \tag{9}$$

where Z is 3 because the hexagonal unit cell in Y-type hexaferrite is composed of three overlapping T and S blocks [3(TS)] and of 18 oxygen layers [12], M is molar mass, N_A is Avogadro number and V is the volume of unit cell.
The values of X-ray density are listed in Table 1. It is clear that X-ray density is varied trend by the change in temperature and time. It can also be observed that the values of bulk density were found to be in general less than those of X-ray density d*x*, which was expected due to the presence of unavoidable pores created during sintering process [13]. Moreover the porosity (P) of all the samples was calculated by using equation (10) as shown below the relation [14].

$$P = 1-(d_B/d_x). \quad (5)$$

The values of porosity for the samples are listed in Table1. It is clear that values of porosity changes with temperature and time. The increase in porosity can be attributed to the decrease in bulk density [15]. The XRD pattern of the prepared samples is shown in Fig. 1(a,b,c) respectively. The average crystallite size was found to be varied about 129 nm to 59 nm. The X-ray diffraction of the samples calcinated at 1100^{o}C for 6 hours, 4 hours and then at 1000^{o}C for 6 hours presents the same behavior, without much of varied diffraction peak.

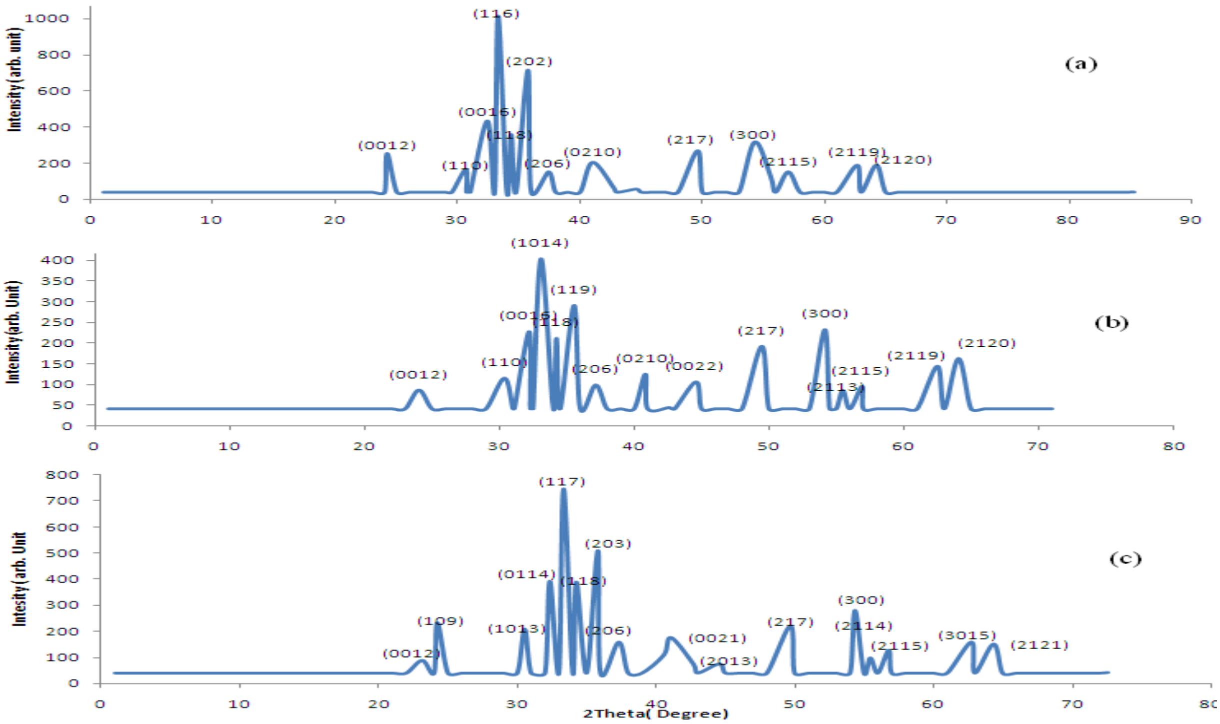

Figure 1. XRD pattern of $BaAl_2Fe_{12}O_{22}$ calcinated at (a) 1100^{0}C for 6 hrs (b) 1100^{0}C for 4 hrs (c) 1000^{0}C for 6 hrs

Table 1. Lattice parameter (a,c), Cell volume(V), Particle size(S), Bulk density(d_B), X-ray density(dx), Porosity of $BaAl_2Fe_{12}O_{22}$

Sample	**Calcinated Temp (^{0}C)**	**Time (Hrs)**	**Lattice Constant**			**Volume**	**Particle size**	**Bulk density**	**X-ray Density**	**Porosity**
			***a* (Å)**	***c* (Å)**	***c/a***	**V(Å^3)**	**S (nm)**	**d_B**	**d_x**	**(Fraction)**
$BaAl_2Fe_{12}O_{22}$	1100	6	5.8544	44.131	7.538	1309.87	129.77	2.469	4.6143	0.465
$BaAl_2Fe_{12}O_{22}$	1100	4	5.8674	44.445	7.574	1325.05	65	2.095	4.4614	0.5304
$BaAl_2Fe_{12}O_{22}$	1000	6	5.8467	46.119	7.888	1365.27	59.13	2.045	4.4270	0.538

B. SEM ANALYSIS

Fig. 2(a,b, c) shows SEM micrographs at room temperature of the samples: BaY cancinated at 1100^{o}C for 6 hrs, 4hrs and at 1000^{o}C for 6 hrs., respectively. For the sample BaY cancinated at 1100^{o}C for 6 hrs (Fig. 2(a)) we observe that the grains are uniformly and densely distributed over the surface of the sample and grain size of the compound is found to be in the range of 129nm. Fig. 2(b) shows the effect of the time of calcination temperature, the grain size of morphology is varied and found varying from 129nm to 65nm. Fig. 2(c) shows the effect of temperature of calcination, the grain size is varied from 129nm to 59 nm. The SEM micrographs shows that the grain size of the samples decreased distinctly and the porosity increased as the calcinated temperature and time decreased.

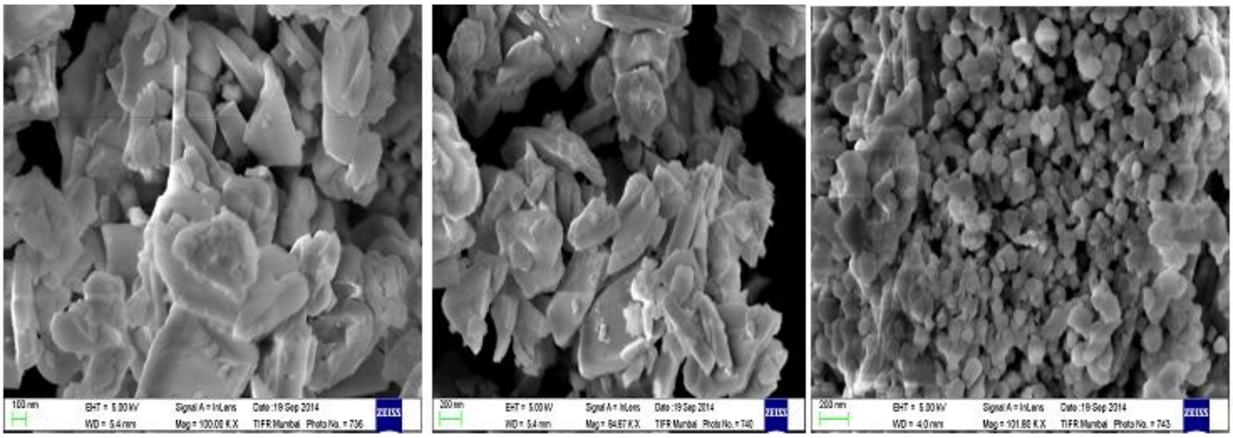

Figure 2. SEM micrographs of $BaAl_2Fe_{12}O_{22}$ $BaAl_2Fe_{12}O_{22}$ calcinated at (a)1100^0C for 6 hrs (b)1100^0C for 4 hrs (c)1000^0C for 6 hrs

C. DIELECTRIC ANALYSIS

The dielectric measurements were carried out over the frequency range of 100 Hz to 10 MHz at room temperature. The variation of dielectric constant ε', complex dielectric loss ε'' and dielectric loss tangent (tan δ) with frequency for sample $BaAl_2Fe_{12}O_{22}$ hexaferrite calcinated at 1100°C for 6 hrs, 4 hrs and at 1000°C for 6 hrs, are shown in figure (4a),(4b) and figure 4(c)respectively. It is clear from figure 4(a, b, c); that the dielectric constant (ε'), complex dielectric loss ε'' as well as dielectric loss tangent(tan δ)decreases with increasing measuring frequency. This decrease behavior of (ε'), (ε'') and (tan δ) with log frequency can be explained on the basis of the assumption that the mechanism of the polarization process in ferrites [16, 17].

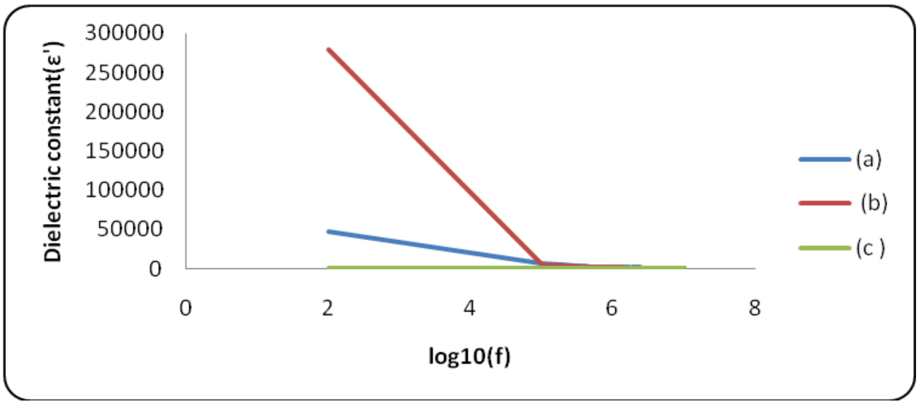

Figure 4.(a) Frequency dependence of dielectric constant(ε') for $BaAl_2Fe_{12}O_{22}$ calcinated at (a) 1100^0C for 6 hrs (b) 1100^0C for 4 hrs (c) 1000^0C for 6 hrs

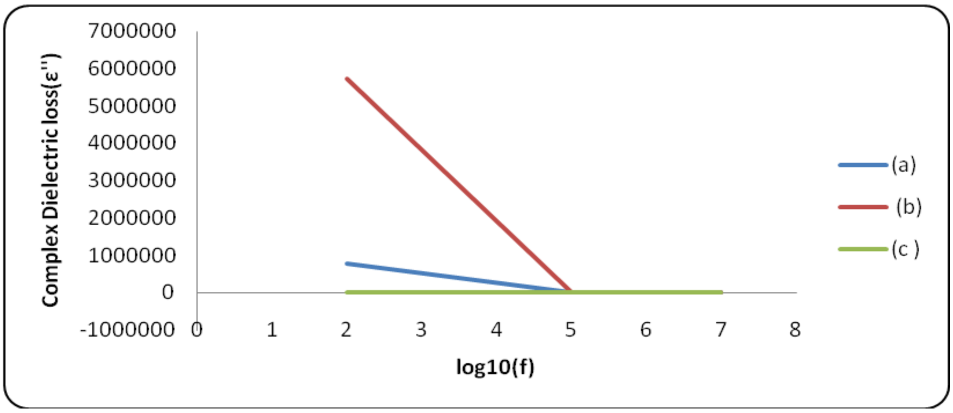

Figure 4.(b) Frequency dependence of Complex dielectric loss(ε'') for $BaAl_2Fe_{12}O_{22}$ calcinated at (a)1100^0C for 6 hrs (b)1100^0C for 4 hrs (c)1000^0C for 6 hrs

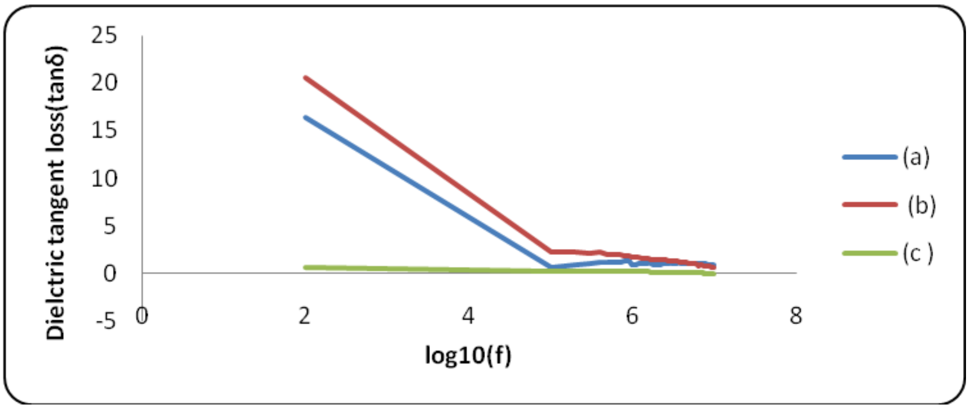

Figure 4.(c) Frequency dependence of Complex dielectric loss(ε'') for $BaAl_2Fe_{12}O_{22}$ calcinated at (a) 1100^0C for 6 hrs (b) 1100^0C for 4 hrs (c) 1000^0C for 6 hrs

D. AC CONDUCTIVITY ANALYSIS

The AC electrical conductivity (σac) was calculated from the dielectric data using an empirical relation of equation (5). Fig.5 shows the variation of AC conductivity with frequency (100 Hz to 10 MHz) at room temperature for all the samples. In general the results show dispersion in σ_{ac} with respect to frequency. For all the samples there is an increase of σ_{ac} value with increase in the frequency value. The dispersion in the AC electrical conductivity of polycrystalline ferrites was explained on the basis of interfacial polarization that formed due to the inhomogeneous structure of the ferrite material. According to Maxwell and Wagner model and Koops phenomenological theory [18], the ferrites are imagined to act as a multilayer capacitor in which the ferrite samples are characterized by a microstructure consisting of conductive thick layers (grains) with separated by resistive thin layers (grain boundaries) [19].

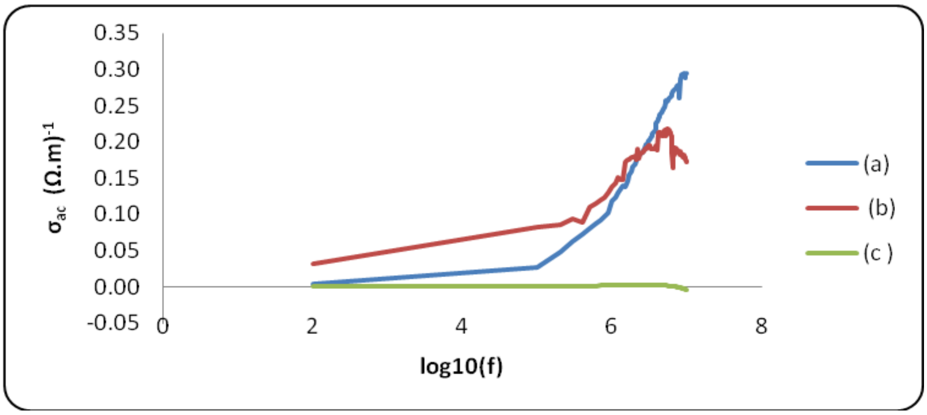

Figure 5. Frequency dependence of Complex dielectric loss(ε'') for $BaAl_2Fe_{12}O_{22}$ calcinated at (a)1100^0C for 6 hrs (b)1100^0C for 4 hrs (c)1000^0C for 6 hrs

IV. CONCLUSIONS

The Y-type hexaferrite $BaAl_2Fe_{12}O_{22}$ calcinated at 1100°C for 6 hrs, 4 hrs and at 1000°C for 6 hrs were prepared by a procedure of chemical co-precipitation method. The analysis by X-ray diffraction (XRD) of the Y sample using the Rietveld refinement confirmed the formation of single-phase compound with a parameter of hexagonal crystal structure. The surface morphology of the samples, studied by SEM, gives the grain size in the order of 129 nm to 59 nm for the Y sample indicates that grains are uniformly and densely distributed over the surface of the sample. The effect of temperature and time of calcination changes the morphology of the studied species. The grain size of the samples decreased distinctly and the porosity increased.

The dielectric constant ε', complex dielectric loss ε'' and dielectric loss tangent (tan δ) decreased with the increase of the frequency. The dispersion of AC conductivity was observed for all the samples calcinated at 1100°C for 6 hrs, 4 hrs and at 1000°C for 6 hrs. For all the samples there is an increase of σ_{ac} value with the increase of frequency.

The study of the dielectric and electrical properties is important in view of applications of the hexaferrite $BaAl_2Fe_{12}O_{22}$ in RF components and circuits.

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