



Finite control of dielectric constant with magnetic field in Sm-doped Ba-Co U-type hexaferrites

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Abstract

Emergence of memory devices based on magneto-electric coupling leads us to investigate the dielectric and magneto-dielectric properties of U-type hexaferrites. $(\text{Ba}_{1-3x}\text{Sm}_{2x})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$ ceramics, where $x = 0.0-0.25$, with the interval of 0.05, have been produced via auto combustion method and sintering at 1200 °C. Every sample has an $R\bar{3}m$ symmetric hexagonal structure, according to the XRD study. The samples with lower Sm content, i.e. $0.0 < x < 0.15$ have better magnetic properties than the samples having $0.15 < x < 0.25$. Impedance spectroscopy has been used to study dielectric characteristics. The sample with $x = 0.15$ at 1.2 T and 1000 Hz showed maximum magnetization and a magneto-electric coupling accompanied by a magnetic coefficient of about 104.9%, which is very high in case of hexaferrites. Therefore, these results will be fruitful from the application point of view.

Keywords: U-type hexaferrites, magnetic properties, impedance spectroscopy, magneto-dielectric coupling

1. Introduction

Magnetoelectric (ME) multiferroics are exciting materials because it is quite easy to manipulate their magnetic states electrically. A set of iron oxides called hexaferrites, which have hexagonal crystal structures, appear attractive due to their high operating temperatures [1–4]. Due to their intricate magnetic configurations, their extreme sensitivity to chemical composition and weak external magnetic field, these materials exhibit enormous ME effects [1,5,6]. Hexaferrites are categorized as M, Y, Z and U type crystals according to the arrangement of the hexagonal axis' basic crystallographic building pieces [1,7]. Due to their intricacy, the magnetic patterns revealed by neutron diffraction are typically characterized by means of several magnetic blocks that are oriented along the axis of the hexagon and designated as L and S, respectively [5,8–10]. The magnetic mo-

ments are collinearly aligned within each block [1]. A non-collinear alignment of spins is frequently produced by the magnetic disturbance brought on by the conflicting superexchange activity between the L and S blocks boundary. This spin structure, known as the Conical transverse spin arrangement, then generates an electric dipole moment. The Katsura-Nagaosa-Balatsky model and the inverse Dzyaloshinskii-Moriya (iDM) interaction can both explain this behaviour [11,12].

Zhang *et al.* [13] has investigated the magneto-dielectric effect in Z-type $\text{Sr}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ at various frequencies and temperatures and found a fairly large negative magneto-dielectric response. Later on, Abdullah *et al.* [14] explored magneto-dielectricity of Y-type $\text{Ba}_2\text{Mg}_2\text{Fe}_{12}\text{O}_{22}$ which has shown 30% of magneto-dielectric response. Zhau and Tang [15] have studied the magneto-dielectric effects in M-type $\text{BaFe}_{10.2}\text{Sc}_{1.8}\text{O}_{19}$ hexaferrite thin films. However, there is much less literature present for the study of magneto-dielectric effects in U-type hexaferrites. This leads us to investigate the

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magneto-dielectric response of Ba-based U-type hexaferrite.

Therefore, the present work deals with the preparation and characterization of Sm-doped $\text{Ba}_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$ U-type hexaferrite. After structural analyses, electrical, magnetic and magneto-dielectric properties were studied.

II. Experimental

$(\text{Ba}_{1-3x}\text{Sm}_{2x})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$ powders, where $x = 0.0$ – 0.25 with the interval of 0.05 , were fabricated through the auto-combustion method. The raw components, such as $\text{Ba}(\text{NO}_3)_2$, $\text{Sm}(\text{NO}_3)_3$, $\text{Co}(\text{NO}_3)_2$ and $\text{Fe}(\text{NO}_3)_3$, in stoichiometric ratio were initially soaked in de-ionized water (~ 20 g in 100 ml) with citric acid at ambient temperature. Thereafter, a gel-based, complex sol was created after 6 h of uninterrupted boiling at 200°C . The gel was burned for 2 h at 600°C to turn it into powder. After that, the obtained powders were calcined in a high-temperature furnace at 1000°C for 12 h. The produced powders were then combined with PVA glue to create pellets. The pellets were made by utilizing a hydraulic press with pressure 3.5 MPa (pressing at higher pressure caused breaking of the pellets) and sintered at 1200°C .

All of the sintered specimens' X-ray diffraction (XRD) data were collected between 20° and 80° using a $2^\circ/\text{min}$ scan rate on the Shimadzu Maxima 7000 (Japan). EDAX and field emission scanning electron microscope (FESEM) from JEOL USA were used for the elemental and surface studies, respectively. The density was determined utilizing the Archimedes principle. The magnetic hysteresis loops were obtained using vibrating sample magnetometer from Microsense (USA). The impedance analyser E4990A from Keysight was used to collect data for evaluation of electrical and magneto-capacitance properties. The sample was mounted on the sample holder attached to impedance analyser and situated in between the poles of the high power magnet (1.5 T) from Marine (India).

III. Results and discussion

3.1. Structural analysis

Figure 1 displays all of the sintered samples' X-ray diffractograms at ambient temperature. Each sample has distinct, strong peaks that demonstrate their

crystalline nature. Additionally, there are no additional peaks, demonstrating that each of the samples belongs to the same phase and does not contain an impurity phase.

In comparison to the undoped $\text{Ba}_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$ ceramics, the samples with $x = 0.05$ and 0.10 have different intensities of some characteristic XRD peaks. However, with the increase in doping concentration the intensity of the peak near 31° decreases and becomes similar to that corresponding to the sample with $x = 0.0$, because, the samples again settled down into the basic structure at provided conditions, e.g. calcination and sintering temperature. This can be explained with the fact that U-type hexaferrite has complex structure and a very small variation in atomic position can lead to the variation in XRD pattern. XRD patterns were also refined by the FullProf software using hexagonal phase with $R\bar{3}m$ space group. Table 1 lists the parameters' values that were determined by refining. The goodness of fit (*GOF*) score for each sample is <5 , indicating that there is high agreement between the computed and observed values. However, the discrepancy in the refined and observed data can be expected due to the structural complexity of the U-type hexaferrite.

Figure 2 depicts fluctuation in the lattice parameters a and c . The chart clearly demonstrates that the values of a and c are decreased with the x amount, which results in a reduction of the volume of the associated unit cell. The variation in the lattice parameters is basically due to the difference in the ionic radii of Ba^{2+} (132 pm)

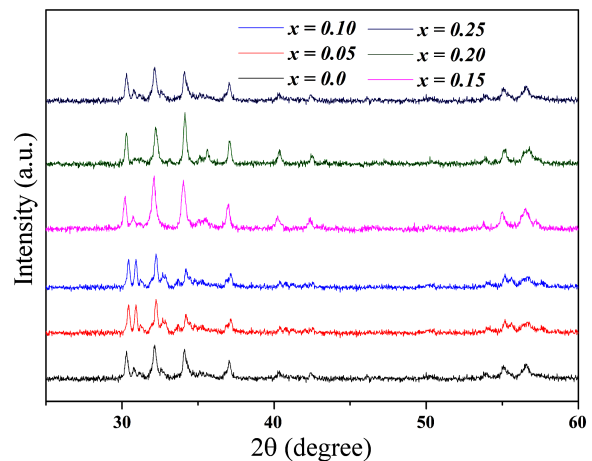


Figure 1. X-ray diffractograms of sintered samples having different Sm content

Table 1. Lattice parameters and goodness of fit (*GOF*) acquired from the refinement

Composition, x	a [Å]	c [Å]	c/a	Volume [Å ³]	<i>GOF</i>
0.0	5.79	112.25	19.38	3763.08	3.79
0.05	5.77	112.10	19.42	3732.13	3.55
0.10	5.75	112.07	19.49	3705.31	3.42
0.15	5.75	112.00	19.47	3703.00	2.21
0.20	5.74	112.00	19.51	3690.13	3.29
0.25	5.74	111.92	19.50	3687.49	3.90

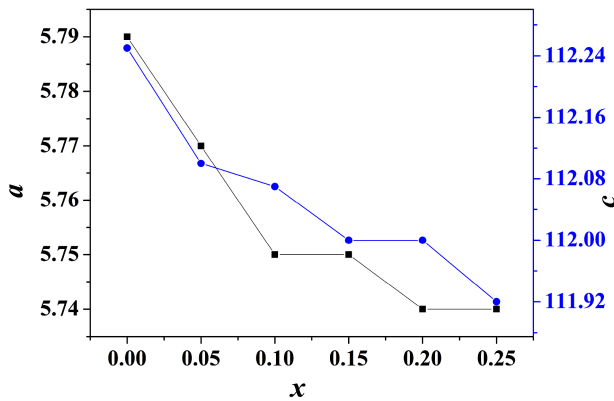


Figure 2. Fluctuation of lattice parameters a and c with Sm content

and Sm^{3+} (109.8 pm). The smaller size of Sm^{3+} leads to the discrepancy in lattice and causes distortion which further advances into the variation in lattice parameters with increased doping concentration.

3.2. Microstructure and density

The surface morphology of each sample was examined using FESEM (Fig. 3). Images showed that the well-defined and evenly spaced grains have a hexagonal shape. Employing the ImageJ software, each sample's grain size was determined. A visible change in the microstructure appeared when x changes from 0.05 to 0.10. The combined effect of dopant concentration and sintering temperature has strong influence on microstructure of the samples. Thus, for the sample with $x = 0.10$, under used sintering conditions, change from uniaxial to plate like structure can be recognised. Therefore, plate like structure can be seen for all higher doped samples. Moreover, a melted plate like structure appeared for the ceramics with the highest Sm-doping which may be due to the inappropriate sintering temperature for this particular composition. Elemental mapping for the prepared ceramics was validated by EDAX and confirmed uniform distribution of element. Figure 4 displays the mapping of the sample with $x = 0.05$ as one example. Ad-

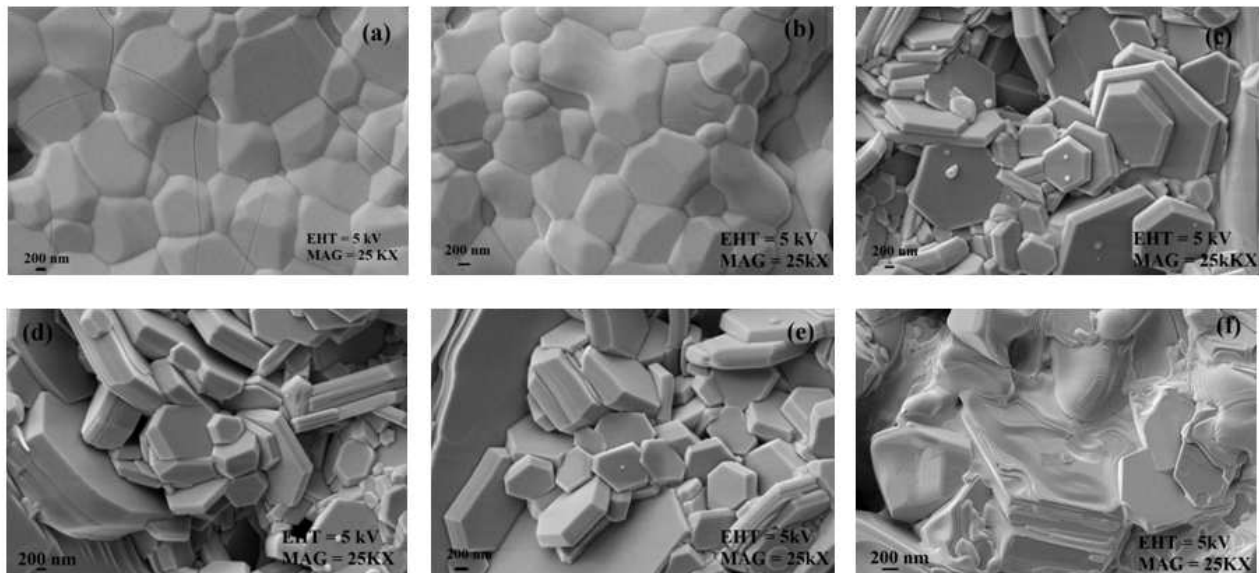


Figure 3. SEM cross-section images of the sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

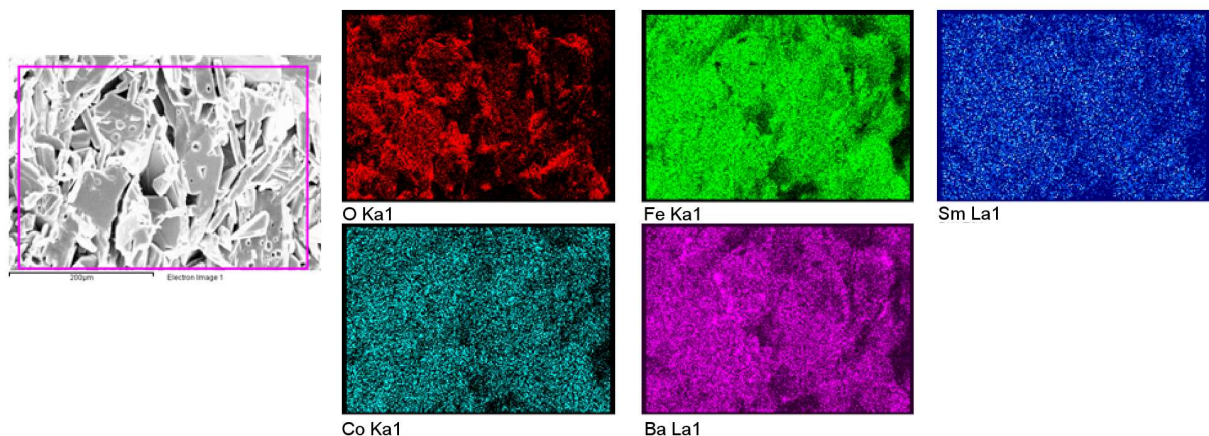


Figure 4. Elemental mapping of the sintered ceramics with $x = 0.05$

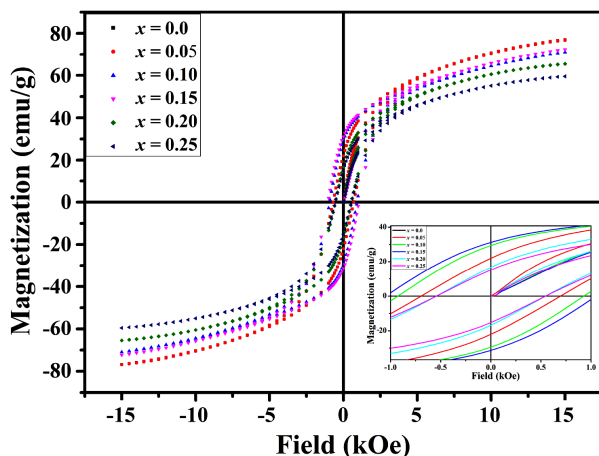
Table 2. Densities and grain sizes (with standard deviation) of the sintered samples

x	Grain size [μm]	Density [g/cm^3]
0.0	9.20 ± 0.011	5.16
0.05	9.11 ± 0.017	5.12
0.10	8.99 ± 0.022	5.01
0.15	8.87 ± 0.015	4.88
0.20	8.56 ± 0.016	4.61
0.25	9.12 ± 0.021	4.75

ditionally, the Archimedes principle has been used to measure the experimental densities. Table 2 contains the computed grain sizes, grain size standard deviations and densities. Clear reduction in the grain size was observed with the increase of the doping amount up to $x = 0.20$, which was corroborated by density and XRD data results.

3.3. Magnetic properties

Figure 5 displays the magnetization versus magnetic field at ambient temperature for the produced samples. It can be seen that remnant magnetization rises with Sm^{3+} ion doping at the Ba-site for $x \leq 0.15$ and then drops. Thus, remnant magnetization increases from 21.66 emu/g for $x = 0.0$ to 31.39 emu/g for $x = 0.15$ and subsequently declines to 14.88 emu/g for $x = 0.25$ (Table 3). The massive Ba^{2+} cations acquire the position in the oxygen lattice in the hexaferrite U-type unit cell, whereas five separate crystallographic locations have various distributions of Co^{2+} and Fe^{3+} cations, including three octahedral sites, one tetrahedral site and one trigonal-bipyramidal site [16,17]. Furthermore, exchange contact via O^{2-} ions couples the opposing spins together [16]. Table 1 shows that the smaller ionic radius of Sm^{3+} compared to Ba^{2+} causes the c/a ratio to progressively decline. The superexchange interaction in $\text{Fe}^{3+}-\text{O}-\text{Fe}^{3+}$ is strengthened by this drop in the c/a ratio, which raises the magnetization value [16–18]. A decrease in remnant magnetization values results from further doping, which may be influenced by Fe^{3+} ions be-

**Figure 5.** Magnetization versus magnetic field at ambient temperature**Table 3.** Coercivity (H_c), remnant magnetization (M_r) and saturation magnetization (M_s) of the sintered samples

x	M_s [emu/g]	M_r [emu/g]	H_c [kOe]
0.0	76.40	21.66	0.69
0.05	77.09	21.96	0.70
0.10	70.24	29.33	0.93
0.15	72.29	31.39	1.05
0.20	64.75	16.65	0.55
0.25	59.27	14.88	0.53

Table 4. Literature data of magnetization strengths for various materials

Material	M_s [emu/g]	H_c [kOe]	Source
$\text{Ba}_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	51.5	0.590	[48]
$\text{Ba}_4\text{Zn}_2\text{Fe}_{36}\text{O}_{60}$	59	0.182	[49]
$\text{Ba}_4\text{Ni}_2\text{Fe}_{36}\text{O}_{60}$	46	0.380	[50]
$\text{Ba}_4\text{Fe}_2\text{Fe}_{36}\text{O}_{60}$	67	0.47	[16]
$\text{Ba}_4\text{Cu}_2\text{Fe}_{36}\text{O}_{60}$	70	0.30	[16]
$(\text{Ba}_{0.7}\text{Bi}_{0.2})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	63.5	0.056	[51]
$(\text{Ba}_{0.55}\text{Bi}_{0.3})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	55.3	0.302	[51]
$(\text{Ba}_{0.7}\text{La}_{0.2})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	73.27	0.740	[52]
$(\text{Ba}_{0.55}\text{La}_{0.3})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	73.31	1.77	[52]
$(\text{Ba}_{0.7}\text{Sm}_{0.2})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	70.24	0.93	This work
$(\text{Ba}_{0.7}\text{Sm}_{0.2})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	72.29	1.05	This work

ing magnetically diluted into Fe^{2+} ions by reduction at octahedral sites [16,17,19,20]. Table 4 compares magnetization data for various Co and Ni-based Ba hexaferrite ceramics gathered from the literature. It is very interesting to observe that with increasing concentration of rare earth doping, such as Bi, La and Sm, the magnetization increases.

3.4. Complex impedance spectroscopy

Figures 6 and 7 demonstrate the temperature-dependent changes in the real and imaginary components (Z' and Z'') of impedance, respectively. The selected temperature range of 250–300 °C was completely based upon the relaxation peaks in impedance in the selected frequency region and the presence of semicircles in the Nyquist plots. Increased temperature reduces both Z' and Z'' magnitudes at low frequencies, but at all temperatures there is a convergence in the values of Z' at higher frequencies. The drop in Z' at lower frequencies may be caused by an increase in AC conductivity with temperature. At higher frequency, convergence of Z' curves measured at different temperatures could be due to space charge release with a steadily declining potential barrier [21–24]. Additionally, the aggregation in Z'' at higher frequencies exhibits a decrease in space charge along inter-granular interfaces, pointing to a thermally driven relaxation reaction [21,25]. The shift of peaks observed in the impedance spectrum of base hexaferrite compared to those samples doped with Sm^{3+} , can be attributed to the influence of dopant on electrical and structural properties of the ma-

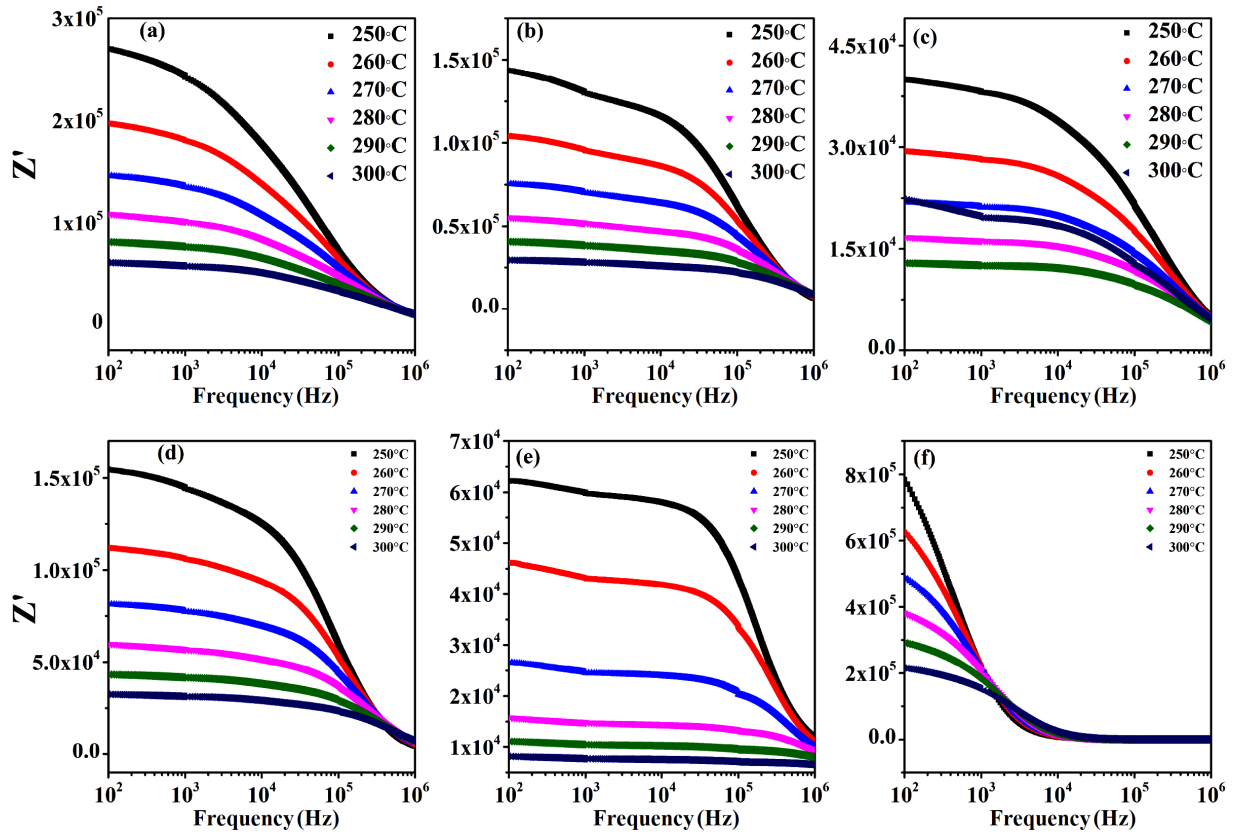


Figure 6. Z' versus frequency for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

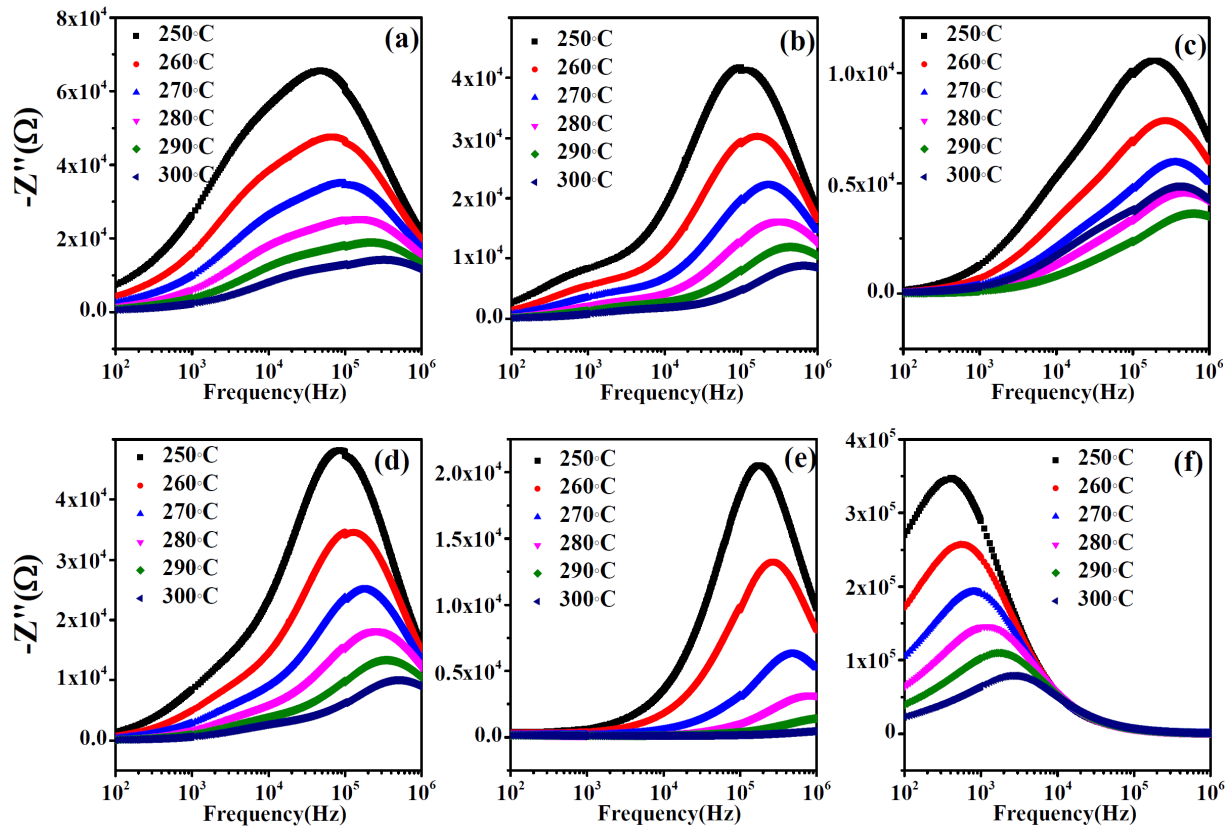


Figure 7. Z'' versus frequency for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

terial. The introduction of dopant alters the crystal lattice structure and creates defects, resulting in localized electronic states that affect the movement of charge carriers. Therefore, the value of Z' of the doped hexaferrite is lower than that for the pure one. The sample having $x = 0.10$ has the lowest impedance value. At specific frequencies corresponding to the characteristic relaxation times of the defects, the movement of charge carriers becomes impeded, leading to the increased resistance and enhanced imaginary impedance, which manifests as peaks in the Z'' component of impedance. The shifting of peak heights towards higher frequencies is caused by the modification of dopant concentrations, which affects the density and distribution of defects within the material. Varying dopant ratios change the energy barrier heights and relaxation times associated with charge carrier movement, ultimately influencing the position of the peaks in Z'' component. Experimental techniques such as impedance spectroscopy, XRD and electron microscopy can be utilized to analyse the structure and electrical properties of doped hexaferrite and confirm the relationship between dopants, defect structure and impedance response. Literature studies have also provided supporting evidence for the observed behaviour of peaks in the Z' and Z'' components and the structural modifications induced by dopants in ferrites [26].

Figure 8 shows the Nyquist diagrams (Z' versus Z'') across a wide frequency range (100 Hz–1 MHz) and various temperatures (250–300 °C). The centroid of two semicircle arcs below the real axis within the chosen temperature range is clearly an evidence of non-Debye relaxation time [27,28]. Such behaviour may be connected to a number of elements, such as grains, grain

boundaries, the distribution of atomic flaws and the stress-strain phenomenon. Higher frequency semicircles are related to bulk (grain) contributions, while lower frequency ones are related to grain border contributions. The circuit comprising two constant phases elements (CPE) and two resistances (R) operating in tandem was used to mimic the impedance data. CPE is employed in place of capacitance to represent how the grain and grain boundary regions behave in order to get beyond the non-linearity and dispersion, as well as for better fitting [29]. One can determine the impedance of CPE by using $Z_{CPE} = 1/((i \cdot \omega)\beta \cdot CPE)$ where $\beta \leq 1$. R_1 and CPE_1 represent the circuit equivalent to the grain, whilst the R_2 and CPE_2 simulate the circuit related to the grain boundary. One way to display the semicircles is by $Z^*(\omega) = Z' + i \cdot Z''$ where:

$$Z' = \frac{R_1}{1 + (\omega_1 \cdot R_1 \cdot C_1)^2} + \frac{R_2}{1 + (\omega_2 \cdot R_2 \cdot C_2)^2} \quad (1)$$

$$Z'' = \frac{\omega_1 \cdot R_1^2 \cdot C_1}{1 + (\omega_1 \cdot R_1 \cdot C_1)^2} + \frac{\omega_2 \cdot R_2^2 \cdot C_2}{1 + (\omega_2 \cdot R_2 \cdot C_2)^2} \quad (2)$$

where (R_1, C_1, ω_1) and (R_2, C_2, ω_2) demonstrate the grain and grain boundary semicircles' resistance, capacitance and peak frequency. Resistance is measured by the Z' axis intercept, while capacitance is determined using the following formula:

$$C_1 = \frac{1}{\omega_1 \cdot R_1} \quad (3)$$

$$C_2 = \frac{1}{\omega_2 \cdot R_2} \quad (4)$$

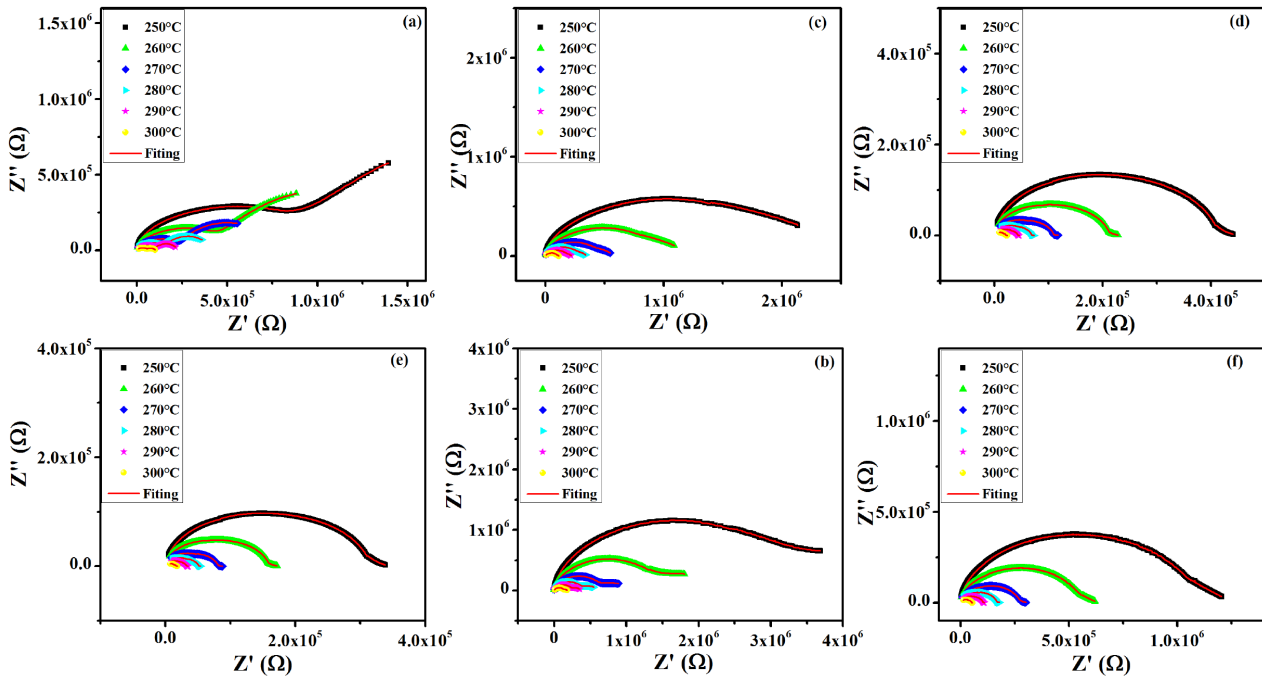


Figure 8. Z' versus Z'' at various temperatures for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

3.5. Study of electrical modulus

The modulus analysis is an effective method for identifying the dielectric relaxation mechanism in materials with low capacitance values. By using modulus analysis, the non-conducting materials' dielectric response can be estimated. In addition, the complex modulus offers a different method for examining the electrical behaviour of materials, and it has been employed by researchers to examine relaxor activities in ceramic materials and ionic substances [30]. The ambiguity caused by the grain or grain boundary impact at elevated temperature, which complex impedance charts might not be able to show, is also confirmed by this notion. When describing the complex modulus, the following equation is used:

$$M^*(\omega) = M'(\omega) + iM''(\omega) \quad (5)$$

Figure 9 illustrates temperature and frequency dependence of M' for all samples. It is evident from the diagram that M' reaches 0 in the lower frequency band, at all temperatures. However, M' increases in the higher frequency range. Such behaviour may be explained by the lack of a restoring force that would normally regulate charge carriers' movement when subjected to an electrical output field [23,31]. This natural occurrence supports the conduction phenomena of charge carriers within narrow range mobility.

Figure 10 illustrates the change of the imaginary component of temperature-dependent electrical modulus (M'') in respect to frequency. The maximum of

M'' curves for the same sample shifts towards higher frequency with the increase of temperature, detailing the hopping process of electrical conduction across all samples. The uneven modulus peak's evolution exhibits a non-Debye form of relaxation [23,28]. The peak's location indicates how long-range mobility relaxes to short-range mobility as frequency rises. Theoretically, the data collected were matched with the Bergman-modified function of the Kohlrausch-William-Watts (KWW) function [32,33]:

$$M'' = \frac{M''_{max}}{(1 - \beta) + \frac{\beta}{1 + \beta} \left[\beta \left(\frac{\omega_{max}}{\omega} + \left(\frac{\omega}{\omega_{max}} \right)^\beta \right) \right]} \quad (6)$$

The frequency during which the peak appears is known as ω_{max} , β is the fitting constant and M''_{max} is the highest value in M'' . Without a doubt, Fig. 10 demonstrates how well theoretical and experimental evidence are consistent.

3.6. Dielectric dispersion

The fluctuations in the real (ϵ') and imaginary (ϵ'') portions of the dielectric permittivity as a frequency variation at varying temperatures are shown in Figs. 11 and 12, respectively. Both ϵ' and ϵ'' drop quickly in the low-frequency region and gradually in the range of higher frequencies. The phenomena of dipole relaxation can be used to explain this kind of behaviour [34]. This means that several polarizations, including interfacial,

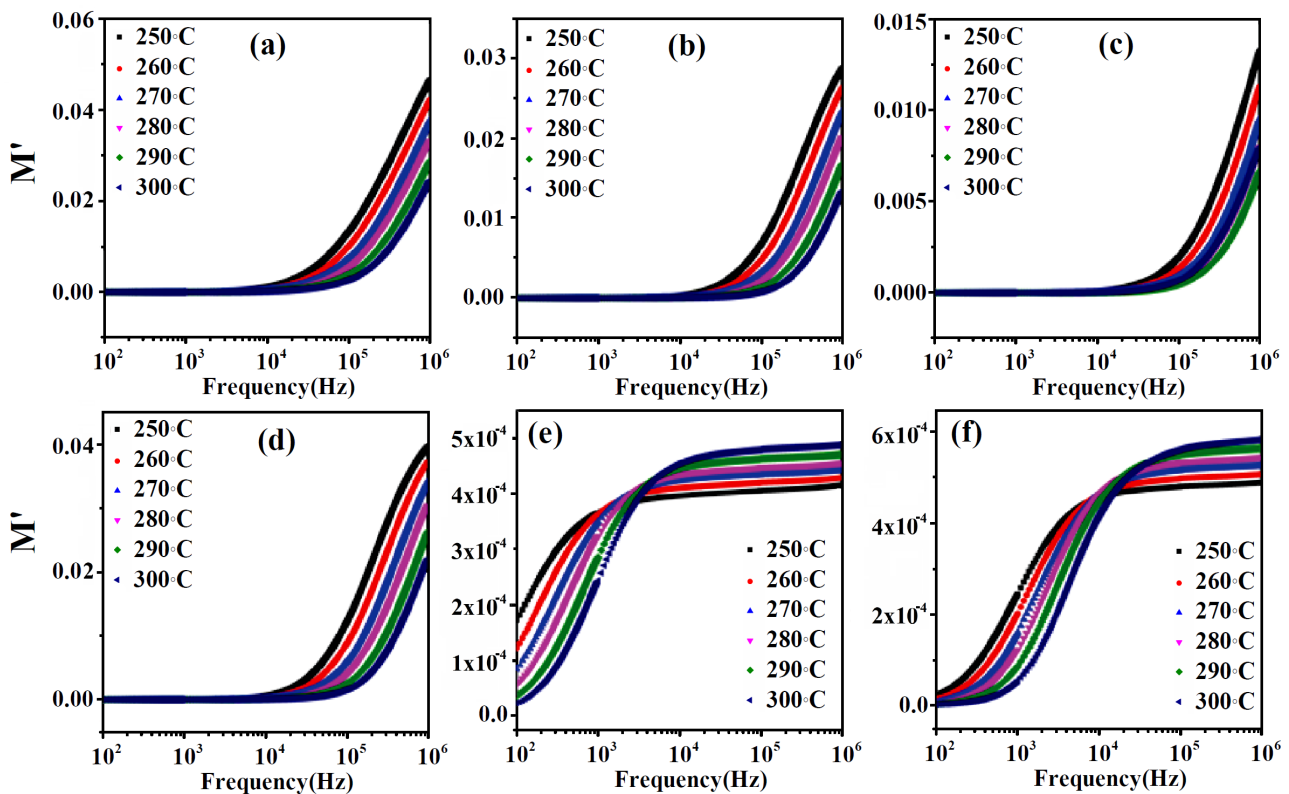


Figure 9. The correlation between frequency and M' at various temperatures for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

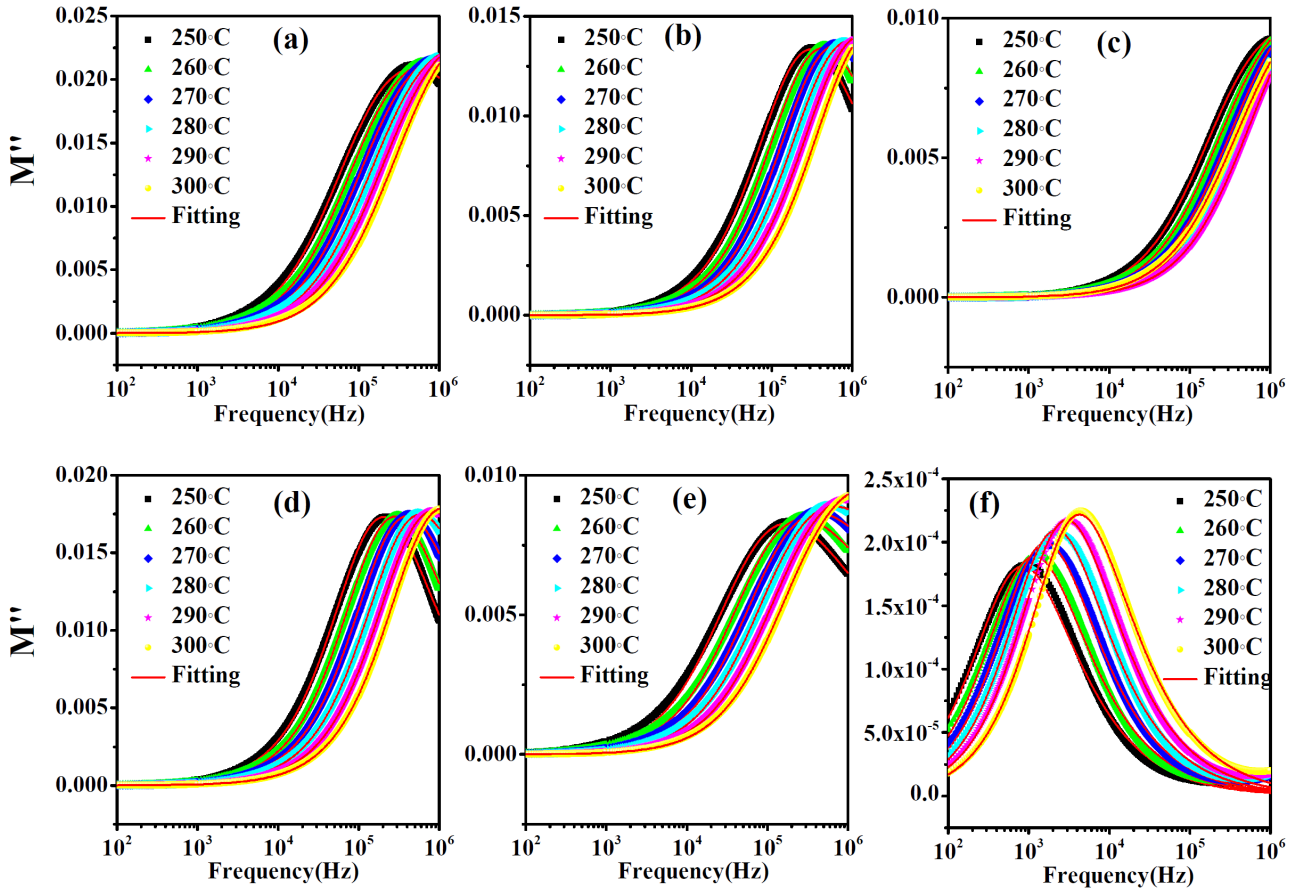


Figure 10. The correlation between frequency and M'' at various temperatures for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

electronic, ionic and dipolar polarization, are added to determine the degree to which a dielectric material is polarized. The low frequency change in electric field has an advantageous effect on all of them, leading to a high reading for both ϵ' and ϵ'' and greater levels for total polarization. It was observed that the high frequency relaxation process can be avoided in a measured frequency window with Sm^{3+} doping in base ferrite which shifts the relaxation frequency towards higher value. This is due to the failure of electric dipole to follow fast varying electric field that increases the friction between them due to the presence of all type of polarization in these samples [35–37]. The dielectric constant of a material determines the amount of energy that a capacitor can store when voltage is applied. Therefore, the static nature of dielectric constants in a specific frequency window favours the constant amount of energy storage within this frequency range [38]. Moreover, the resistance and capacitance properties are interlinked with the microstructure. At lower frequencies, grain boundaries with high resistance lead to space charge polarization where electrons accumulate, resulting in an increase in the dielectric constant. However, at higher frequencies, the dielectric constant decreases due to a reduction in charge accumulation at grain boundaries [39]. It is clear from the Table 2 that the sample with $x = 0.20$ has exhibited the lowest grain size which means it has the highest

amount of grain boundaries. Hence, this sample has the highest value of dielectric constant among all the samples.

Low-frequency band dielectric constant increases with temperature from 250 to 300 °C. With a rise in temperature, this finding demonstrates improved charged carrier movement. All produced samples' relaxation behaviour is explained by the Havriliak-Negami relaxation model [40]. All prepared samples' relaxation characteristics are accounted by the Havriliak-Negami relaxation model [40] in line with this model:

$$\epsilon' = \epsilon_{\infty} + \frac{\epsilon_s - \epsilon_{\infty}}{(1 + (\omega \cdot \tau)^{2\alpha})^{\beta}} \quad (7)$$

$$\epsilon'' = \frac{(\epsilon_s - \epsilon_{\infty})\omega \cdot \tau}{(1 + (\omega \cdot \tau)^{2\alpha})^{\beta}} \quad (8)$$

where ϵ_{∞} denotes the dielectric permittivity measured at the high frequency limit, ϵ_s denotes the dielectric permittivity measured at low frequency, ω denotes the external field's angular frequency and τ denotes the medium's average relaxation time. The irregularity and broadness of the dispersion curves are defined by the exponents α and β , respectively. The Cole-Cole and Cole-Davidson equations are the products of the Havriliak-Negami equation for $\beta = 1$ and $\alpha = 1$, respectively. This model also provides estimation for the Cole-Cole

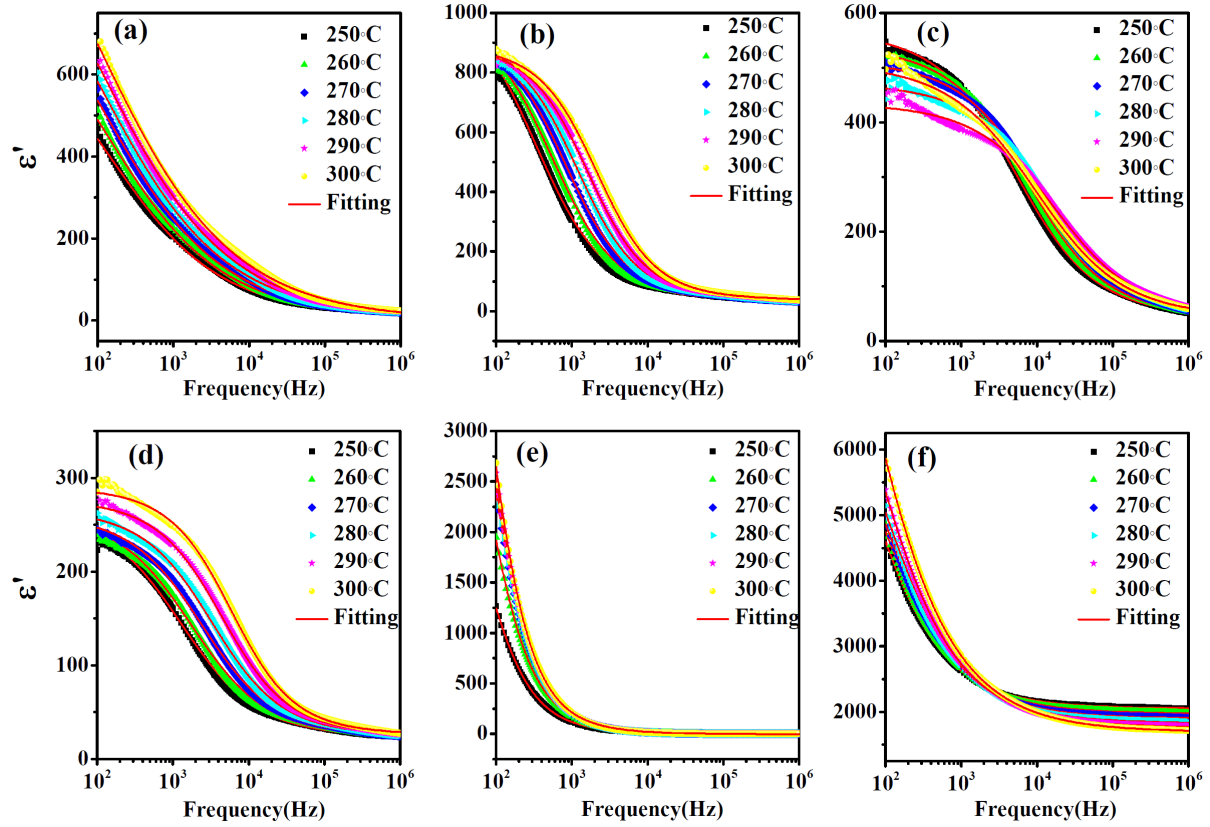


Figure 11. ϵ' versus frequency at various temperatures for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

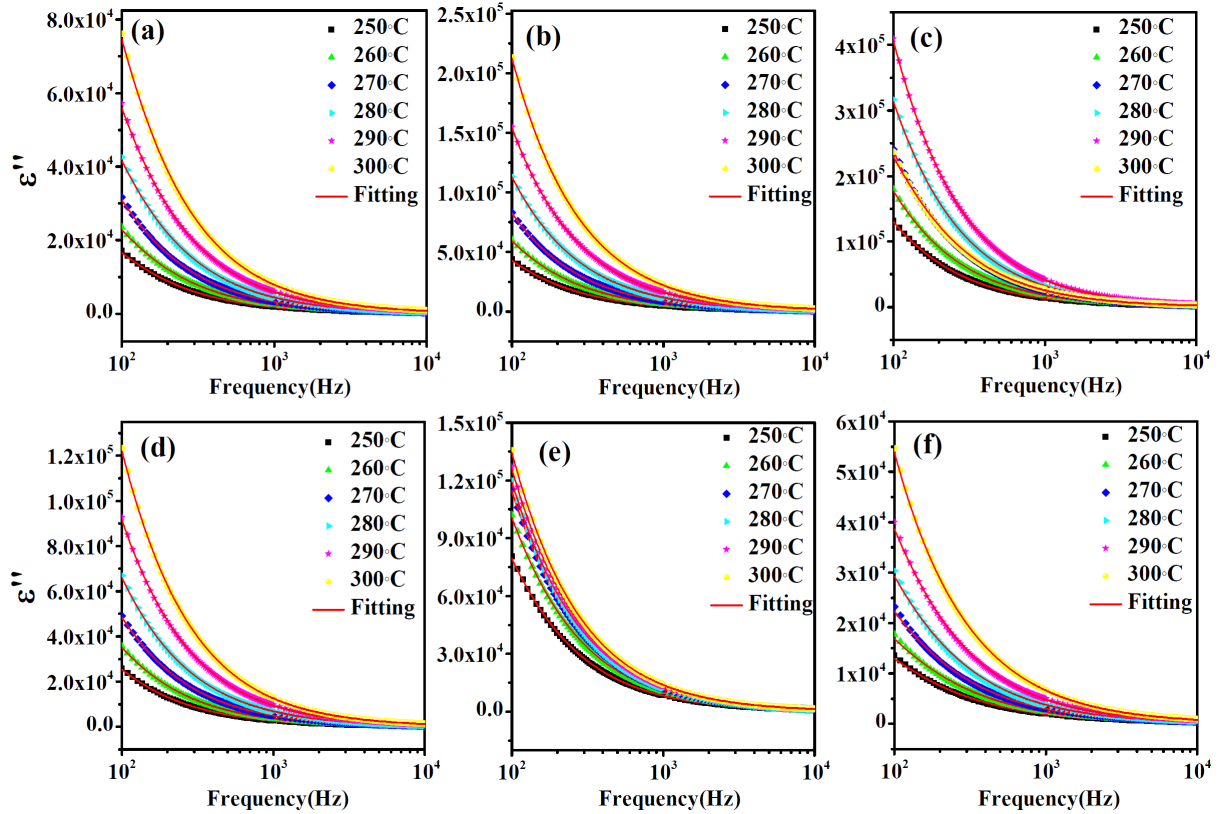


Figure 12. ϵ'' versus frequency at various temperatures for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

Table 5. Values of α obtained from different modes of fitting

Temperature [°C]	α $x = 0.0$			α $x = 0.05$			α $x = 0.10$			α $x = 0.15$			α $x = 0.20$			α $x = 0.25$		
	ϵ'	ϵ''	M''	ϵ'	ϵ''	M''	ϵ'	ϵ''	M''	ϵ'	ϵ''	M''	ϵ'	ϵ''	M''	ϵ'	ϵ''	M''
250	0.88	0.90	0.93	0.91	0.86	0.72	0.9	0.93	0.88	0.9	0.87	0.86	0.86	0.72	0.66	0.88	0.87	0.71
260	0.88	0.89	0.94	0.90	0.87	0.70	0.9	0.92	0.89	0.89	0.88	0.87	0.87	0.70	0.64	0.89	0.88	0.72
270	0.89	0.87	0.96	0.85	0.85	0.77	0.91	0.9	0.91	0.84	0.86	0.85	0.85	0.77	0.62	0.91	0.86	0.71
280	0.87	0.89	0.99	0.86	0.87	0.75	0.89	0.92	0.94	0.85	0.88	0.87	0.87	0.75	0.69	0.94	0.88	0.74
290	0.91	0.92	0.95	0.86	0.88	0.75	0.93	0.95	0.9	0.85	0.89	0.88	0.88	0.75	0.66	0.9	0.89	0.73
300	0.92	0.95	0.95	0.85	0.85	0.73	0.94	0.98	0.9	0.84	0.85	0.84	0.85	0.73	0.61	0.9	0.85	0.71

and Cole-Davidson models. The values of α (at constant β value of 0.95) are listed in Table 5. Including all temperatures, it is obvious that the α values are higher than 0, demonstrating that the relaxations are elongated. As a result, the relaxations are much wider than the ideal Debye relaxation, indicating that the sample displays non-Debye relaxation [41].

3.7. AC and DC conductivities

The variation in electrical conductivity versus frequency is depicted in Fig. 13. The frequency dependence of AC conductivity was examined using known the “global law of conductivity”, the Jonscher’s power law [42]:

$$\sigma_{AC} = \sigma_{DC} + A \cdot \omega^n \quad (9)$$

where the non-dimensional frequency coefficient is denoted by n and A stands for the dispersion parameter (A and n stand for the degree of polarizability and how mobile ions interact with the lattice surrounding them, respectively [43]). The longitudinal movement of electric charge with sudden hopping is represented by $n < 1$ and localized hopping of electric charge without departing the local area by $n > 1$ [44].

The plateau and dispersion zone are two separate regimes shown in Fig. 13 as frequency dependent conductivity charts. In the plateau region, it was discovered that the conductivity is frequency independent. On the other hand, in the dispersion area, a gain in conductivity as frequency increases in high-frequency region is observed. Accordingly, the dispersion zones correlate to AC conductivity (σ_{AC}) and the plateau region to DC

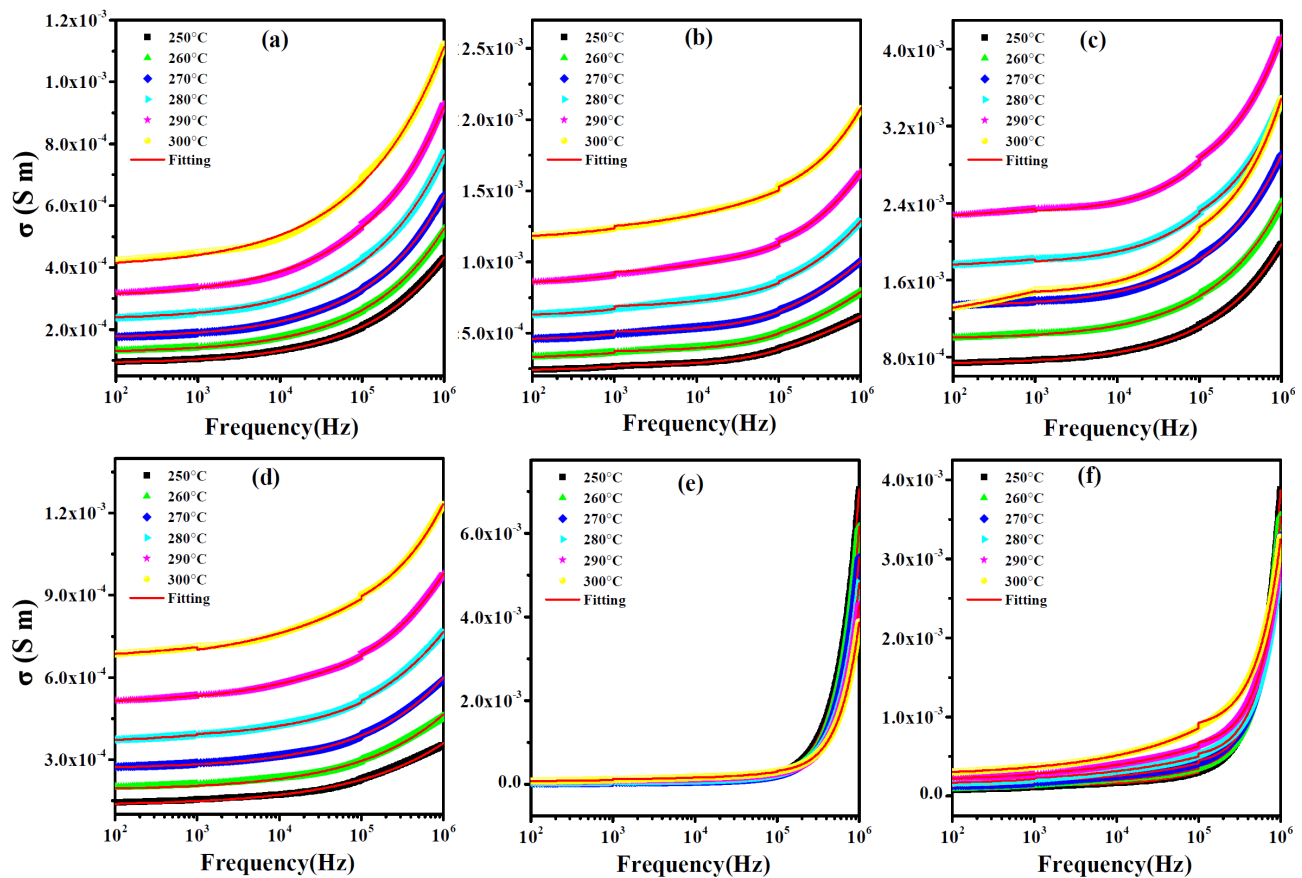


Figure 13. Frequency-dependent change of electrical conductivity at various temperatures for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

Table 6. List of constant n values and σ_{DC} at different temperatures

Temperature [°C]	$x = 0.0$		$x = 0.05$		$x = 0.10$		$x = 0.15$		$x = 0.20$		$x = 0.25$	
	σ_{AC} [10 ⁻⁵ S·m]	n	σ_{AC} [10 ⁻⁵ S·m]	n	σ_{AC} [10 ⁻⁵ S·m]	n	σ_{AC} [10 ⁻⁵ S·m]	n	σ_{AC} [10 ⁻⁵ S·m]	n	σ_{AC} [10 ⁻⁵ S·m]	n
250	1.59	0.17	1.55	0.69	0.86	0.20	5.80	0.43	4.05	0.42	1.26	0.30
260	2.39	0.21	2.45	0.51	1.86	0.21	11.8	0.42	7.93	0.42	2.88	0.35
270	4.68	0.25	3.85	0.43	3.63	0.26	22.3	0.45	16.1	0.45	6.63	0.41
280	6.61	0.27	6.08	0.42	5.73	0.30	36.0	0.49	26.6	0.48	11.6	0.47
290	11.69	0.31	9.32	0.42	9.37	0.35	59.3	0.53	42.0	0.52	19.6	0.52
300	24.84	0.37	18.01	0.44	17.79	0.43	118.1	0.55	81.7	0.54	40.4	0.57

conductivity (σ_{DC}). Table 6 lists the values of n and DC conductivity for each sample upon fitting. The table unequivocally demonstrates that the value of n is below 1. This indicates that a sudden hopping phenomenon with carrier translational motion occurs. Moreover, the value of conductivity increases with the rise in doping concentration, which is primarily due to the availability of more charge carriers and oxygen vacancies generated by the Sm^{3+} substituting Ba^{2+} [43]. Therefore, the sample which has shown the lowest value of impedance (i.e. $x = 0.10$) has exhibited maximum conductivity of all samples.

3.8. Magneto-dielectric response

Frequency dependent dielectric measurements at room temperature were performed for all samples in order to assess the magneto-dielectric reaction. Figure 14 shows the frequency dependence of ε' at different magnetic fields from 0–1.2 T. The image makes it evident that ε' decreases with the increase in magnetic strength to 1.2 T value. Such results demonstrate that the external magnetic field can easily change the dielectric constant,

which is important to note for the magneto-dielectric interaction at specific temperatures. The polarization of space charges may be caused by the change in the magnetic field [45]. The following formula was used to determine the magnitude of the magneto-dielectric response:

$$MDR = \frac{\varepsilon'(H) - \varepsilon'(0)}{\varepsilon'(0)} \cdot 100 \quad (10)$$

where $\varepsilon'(0)$ and $\varepsilon'(H)$ are dielectric constants at zero field and magnetic field H , respectively. Table 7 contains MDR values that were calculated using the formula above. Therefore, it is obvious that the sample with the strongest reaction is the sample $x = 0.15$. As previously discussed, the sample $x = 0.15$ produced the highest magnetization. The strain phenomena caused by a magnetic field is a good explanation for this behaviour. The material experiences strain as a result of the magnetization that the applied magnetic field causes. This induced strain causes tension and a little electric field within the material, which alters its dielectric characteristics [46].

Since the sample with $x = 0.15$ has the highest mag-

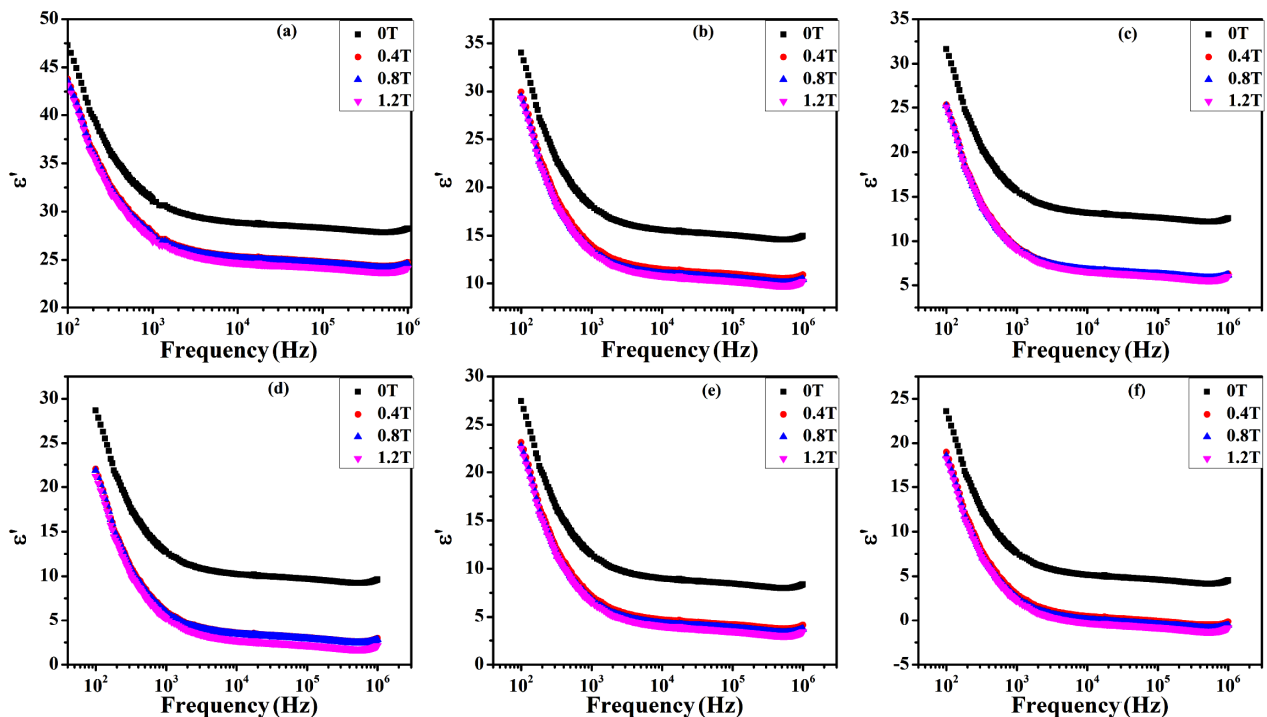


Figure 14. ε' versus frequency at different magnetic fields at room temperature for sintered ceramics with different dopant content: a) $x = 0.0$, b) $x = 0.05$, c) $x = 0.10$, d) $x = 0.15$, e) $x = 0.20$ and f) $x = 0.25$

Table 7. MDR values for each sample collected at 1000 Hz

x	MDR [%] at 0.4 T	MDR [%] at 0.8 T	MDR [%] at 1.2 T
0	12.13	12.82	14.31
0.05	26.11	29.20	30.60
0.10	65.02	66.55	73.12
0.15	90.72	98.74	104.9
0.20	47.61	52.16	55.18
0.25	47.79	48.69	49.95

Table 8. A comparison of the magneto-dielectric response produced by several different hexaferrite types

Composition	MDR [%]	Field [T]	Reference
$\text{Sr}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$	32.2	0.0125	[53]
$\text{BaFe}_{10.2}\text{Sc}_{1.8}\text{O}_{19}$	10	2.5	[54]
$\text{Sr}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$	10	0.15	[55]
$(\text{Ba}_{0.55}\text{La}_{0.3})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	54.18	1.5	[52]
$(\text{Ba}_{0.4}\text{La}_{0.4})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	50.21	1.5	[52]
$(\text{Ba}_{0.55}\text{Sm}_{0.3})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	104.9	1.2	This work
$(\text{Ba}_{0.7}\text{Sm}_{0.2})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$	73.12	1.2	This work

netism, it follows that this sample will exhibit the highest magneto-dielectric response. Table 8 compares the magneto-dielectric response derived from several hexaferrite types. It is obvious that the compositions suggested in this work have a relatively high MDR value. It is worth mentioning here that the value of dielectric constant shown in Fig. 14 at room temperature at 0 T is very low as compared to the one at 250 °C as given in Fig. 12 in the absence of magnetic field. The increase in the value of $\varepsilon'(0)$ is due to the rise in the mobility of charge carriers at high temperature. The temperature has a similar impact on the dielectric constant as frequency does. The dielectric constant rises with temperature because polar sites are more mobile at higher temperatures [47].

IV. Conclusions

By using auto combustion method, U-type hexaferrites $(\text{Ba}_{1-3x}\text{Sm}_{2x})_4\text{Co}_2\text{Fe}_{36}\text{O}_{60}$ powders, where $x = 0.0$ – 0.25 with the interval of 0.05 , were prepared and the corresponding ceramics were obtained after sintering at 1200°C . XRD analysis of the obtained samples revealed a $R\bar{3}m$ symmetric hexagonal structure without the impurity phases. In contrast to the sample with $x = 0.0$, the samples where $0.0 < x \leq 0.15$ have shown enhanced magnetic characteristics, while the samples having doping content $0.15 < x \leq 0.25$ showed decreasing magnetization. Dielectric properties were investigated using impedance spectroscopy. Maximum magnetization and a magneto-electric reaction accompanied by a magnetic coefficient of roughly 104.9% were both displayed by sample with $x = 0.15$ composition at 1.2 T at 1000 Hz.

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