Available online at www.scholarsresearchlibrary.com

Scholars Research Library

Archives of Physics Research, 2016, 7(1):1-14
(http://scholarsresearchlibrary.com/archive.html)

Archives of Physics Research
Scholars Research Library
ISSN 0976-0970
USA CODEN: APRRC7

Synthesis, Characterization and Study of Dielectric Properties, AC Conductivity, and Surface Morphology of Ferroelectrics Ba1-xSrxTiO3 by Ceramic Method

Smita Patil*, R.B.Pujar, Bahubali Doddannavar

P.C. Jabin Science College, Hubli-580031, Karnataka, India

ABSTRACT

Ferroelectrics with general chemical formula $Ba_{1-x}Sr_xTiO_3$ (x=0.3,0.5,0.7,0.9) were synthesized by ceramic method and characterized by X-ray diffraction, study of surface morphology by scanning electron microscopy(SEM), porosity variation with different composition of x and same behavior is indicated by SEM micrographs. Dielectric constant studies found with dispersion is due to Maxwell Wagner type interfacial polarization. AC conductivity studies with frequency suggest that small poloron type of conduction mechanism.

Keywords: piezoelectric, ceramic, poloron, grain boundaries, dispersion

INTRODUCTION

The breakthrough in the research on ferroelectric materials came in the early 1950's with widespread use of BaTiO3 based ceramics in capacitor applications and piezoelectric transducer devices. (1) particle size and surface structure explain the distinct properties of the ferroelectrics (2).

In the present research work
In this present work the ferroelectric Ba1-xSrxTiO3 has been synthesized with x=0.3, 0.5, 0.7, 0.9 using ceramic method for better physical properties with high purity.

Experimental part
Precursors, BaCO3, SrCO3and TiO2in AR grade, were taken in Stoichiometry proportion for preparing 10 grams of Ba1-XSrXTiO3with x=0.3 ,0.5, 0.7 and 0.9. The required quantities of these raw materials were mixed thoroughly in acetone medium by milling for 3 to 4 hours to get fine powders. The ferroelectric powder was pre-sintered at 10000 C for 10 hours in an air medium to make the raw materials, if any to react partially to reduce the evolution of gas in the final sintering process. Then the powders were pressed in to pellet by adding a drop of PVA and pressed by KBr Press by applying a pressure of about 7 tons per square inch for 5 minutes by putting a powder of about 1gm in a di of 1 cm in diameter. The pellets were subjected to final sintering at about 12000C and furnace cooled.

Binder addition
The calcined sample at 1200 0C was chosen and mixed with polyvinyl alcohol that acts as a binder. The role of a binder is to provide mechanical strength to the pellets that are to be formed for sintering. Binder is made by mixing

2-3% of polyvinyl alcohol in powder form with distilled water and then stirring with the help of a magnetic stirrer. The binder vaporizes during sintering. After mixing the binder the sample is left for some time so that the mixture dries and then grinded again.

RESULT AND CONCLUSIONS

X ray characterization

X-Ray diffraction of the sample was obtained from Indian institute of science, Bangalore. Using filtered Cu.kα, radiation of wavelength 1.5418 A0. The interplanar distance of the cubic system was calculated by the relation

$$d = a/ (h2+k2+l2)1/2$$

Where, a = lattice constant
(h, k, l) = miller indices

While the relation calculated the lattice constant

$$a = \lambda (h2+k2+l2)/2\sin\theta$$

Where, λ = wavelength of the monochromatic X-ray used
θ = glancing angle

The diffract grams were indexed in the light of the crystal structure of natural spinel BaTiO3. The prominent line in the diffraction pattern of spinel corresponds to (110) plane. Knowing the values of θ, λ and (hkl), lattice constant was calculated. Knowing the value of lattice constant and miller indices, the value of interplanar distance was calculated for other planes.

Density measurement

The density and porosity were calculated as follows:

Actual density (da) = m/V

Where m= mass

V= Volume

X-Ray density (dx) = 8M/Na3

Where M- molecular weight
N- Avogadro's number
a-lattice constant

Porosity (Pa) = $\frac{dx-da}{dx}$ 100 =-------

Data on X-Ray Diffraction

Table 1: For X=0.3 a=8.1621A^0 λ=1.5418A^0

2Θ	Θ	Sin Θ	(h k l)	d (A^0) Calculate	d obs(A^0)
22.2426	11.1213	0.192886753	(0,0,1)	3.996645638	3.99684
31.6778	15.8389	0.272933465	(1,1,0)	2.824497905	2.82463
39.0354	19.5177	0.334098047	(1,1,1)	2.307406484	2.30752
45.4871	22.74355	0.386607143	(2,0,0)	1.994013856	1.99411
51.1842	25.5921	0.431961399	(2,1,0)	1.784650207	1.178474
57.5744	28.7872	0.481557893	(2,1,1)	1.600845944	1.60092

66.1869	33.09345	0.54600619	(2,1,0)	1.411888755	1.41196
70.7069	35.35345	0.57861873	(1,0,3)	1.332310829	1.33238
75.3387	37.66935	0.611103692	(3,0,1)	10261488043	1.26155
79.6232	39.8116	0.640265231	(2,2,2)	1.204032271	1.20409
83.8336	41.9168	0.66805077	(3,2,0)	1.153954212	1.15305

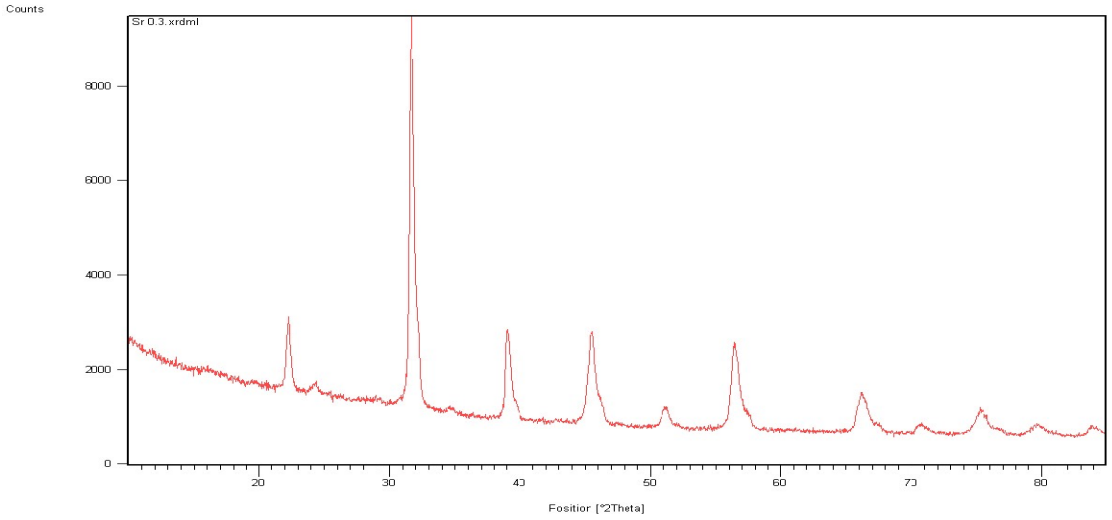

Figure 1: X-Ray diffraction pattern for X=0.3(Ba0.7Sr0.3 TiO3)

Table 2: For X=0.5 a=8.1851A0 λ=1.5418A0

2Θ	Θ	Sin Θ	(h k l)	d (A^0) Calculate	d obs(A^0)
22.3469	11.17345	0.19377977	(0,0,1)	3.978227	3.97842
32.2221	16.11105	0.277499942	(1,1,0)	2.77801	2.77815
39.7956	19.8978	0.340343445	(1,1,1)	20265064923	2.26518
46.0967	23.04835	0.391507772	(2,0,0)	1.969054142	1.96915
51.3630	25.6815	0.433368116	(2,1,0)	1.778857215	1.77894
57.5845	28.79225	0.481635137	(2,1,1)	1.6005892	1.60067
66.1760	33.088	0.545926497	(2,2,0)	1.4112094858	1.41216
67.3645	33.68225	0.554586664	(3,0,1)	1.390044242	1.9011
76.8363	38.41815	0.621396005	(3,1,1)	1.240593749	1.24065
84.0062	42.0031	0.669170813	(2,2,2)	1.152022749	1.15112

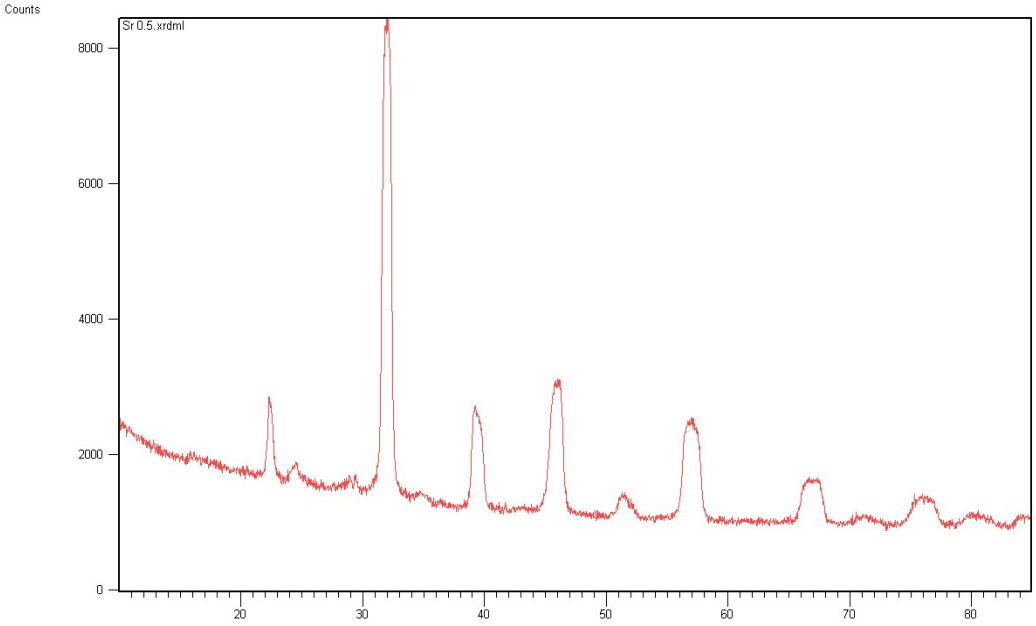

Figure 2: X-Ray diffraction pattern for X=0.5 (Ba0.5 Sr0.5 TiO3)

Table 3: For X=0.7 a=8.1610A0 λ=1.5418A0

2Θ	Θ	Sin Θ	(h k l)	d (A^0) Calculate	d obs(A^0)
22.5402	11.2701	0.19543438	(0,0,1)	3.944546493	3.94474
32.2479	16.12395	0.27771624	(1,1,0)	2.775854952	2.77599
39.7798	19.8899	0.340213792	(1,1,1)	2.265928122	2.26604
46.2977	23.14885	0.393121207	(2,0,0)	1.960972812	1.96107
52.0467	26.02335	0.438737399	(2,1,0)	1.757087498	1.75717
57.5855	28.79275	0.481642785	(2,1,1)	1.600563785	1.60064
67.6035	33.80175	0.556320996	(2,2,0)	1.385710777	1.38578
76.8726	38.4363	0.621644168	(3,0,1)	1.240098499	1.24016
81.4254	40.7127	0.652266433	(2,2,2)	1.181879	1.18096

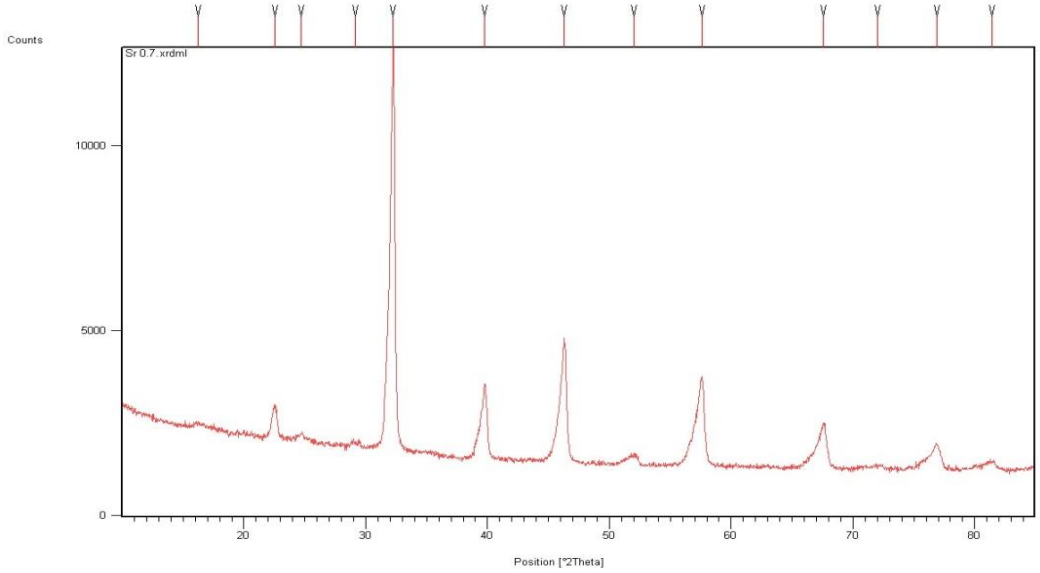

Figure 3: X-Ray diffraction pattern for X=0.7 (Ba0.3Sr0.7 TiO3)

Table 4: For X=0.9 a=8.1829A0 λ=1.5418A0

2Θ	Θ	Sin Θ	(h k l)	d (A^0) Calculate	d obs (A^0)
22.6975	11.34875	0.1967820427	(0,0,1)	3.197564418	3.91775
32.3564	16.1782	0.278625711	(1,1,0)	2.76679419	2.76693
39.9131	19.95655	0.341307433	(1,1,1)	2.258667479	2.25878
46.4251	23.21255	0.394143226	(2,0,0)	1.955887982	1.95598
52.2547	26.12735	0.440367791	(2,1,0)	1.750582163	1.75067
57.7232	28.8616	0.482695532	(2,1,1)	1.597072994	1.59715
67.7425	33.87125	0.557328553	(2,2,0)	1.383205644	1.38327
72.4703	36.23515	0.591100613	(1,0,3)	1.3404177297	1.30424
77.0870	38.5435	0.623108627	(3,0,1)	1.237183961	1.23724
81.6354	40.8177	0.653654426	(3,1,1)	1.179369357	1.17845

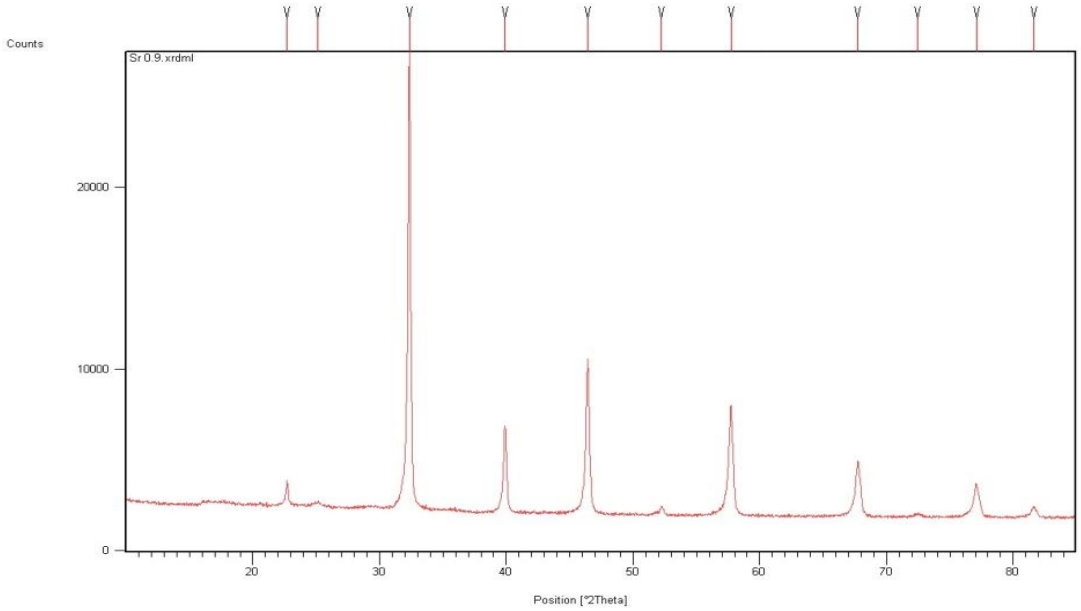

Figure 4: X-Ray diffraction pattern for X=0.9 (Ba0.1 Sr0.9 TiO3)

Table 5: Data on Porosity

Composition(X)	x-ray density gr/cm	Actual density gr /cc	Porosity P=dx-da/dx
0.3	4.54922	2.96566	34.80%
0.5	4.3636	2.7940	38.77%
0.7	4.3560	2.14256	50.81%
0.9	4.1785	3.7918	9.254%

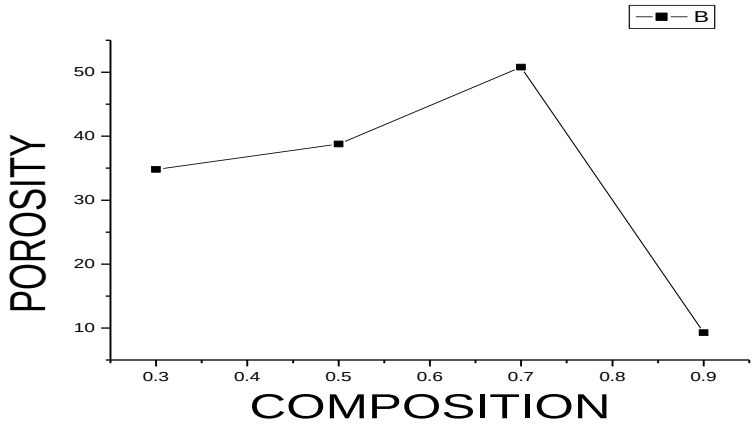

Figure 5: Porosity VS Composition

Table 6: Data on lattice parameters

composition(X)	lattice constant a(A^0)	C (A^0)	Tetragonality (c/a)
0.3	3.99463	3.99463	1.001
0.5	3.92889	3.92889	1.000
0.7	3.92584	3.92584	1.000
0.9	3.91302	3.91302	1.000

The diffraction pattern of ferroelectrics indicates tetragonal of perovskites structure without extra peaks this confirms the formation of single phase ferroelectrics.
The observed and estimated values of d for each plane are in good agreement with earlier reports (q) the tetragonality ratio (c/a) is constants the variation of lattice constants with composition is listed in table no (6). From the table, it is observed that lattice parameters decrease with increases "Sr" content it is attributed to the volume differences of Ionic radius Sr=219pm. Ti=176pm, Ba =25pm&O=48pm ions.

Porosity

Variation of porosity with composition is depicted in table (5) from the table it is noticed that porosity of all the samples lies in the tangle of 9 to 50 it indicates that porosity increases up to x=0.7 & reaches minimum x=0.9. The same behavior indicated by SEM Micrographs.

Experimental detail for SEM

The surface morphology of ferroelectric is carried out by scanning electron microscopy (SEM) from Indian institute of science, Bangalore. The average grain size of ferroelectric sample the average grain size was calculated by counting number of grain boundaries intercepted by a measured length of a random straight line drawn on micrographs [3].
Average grain size = length / number of particles =_____μm

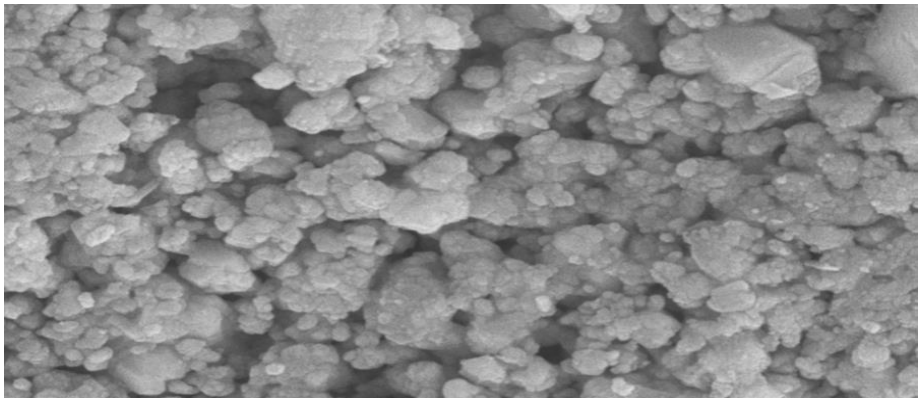

Photo 1: X=0.3 SEM for Ba0.7Sr0.3TiO3

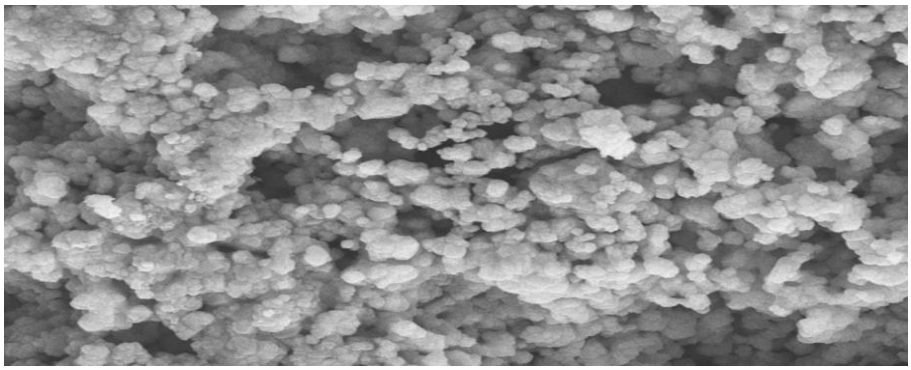

Photo 2: X=0.5 SEM for Ba0.5Sr0.5 TiO3

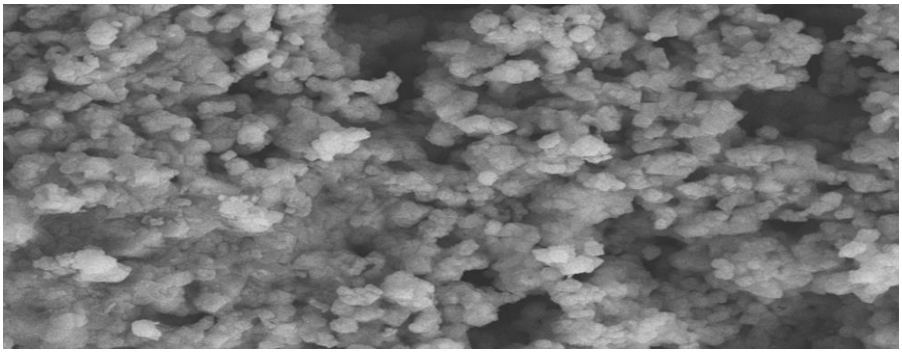

Photo 3: X=0.7SEM for $Ba_{0.3}Sr_{0.7}$ TiO_3

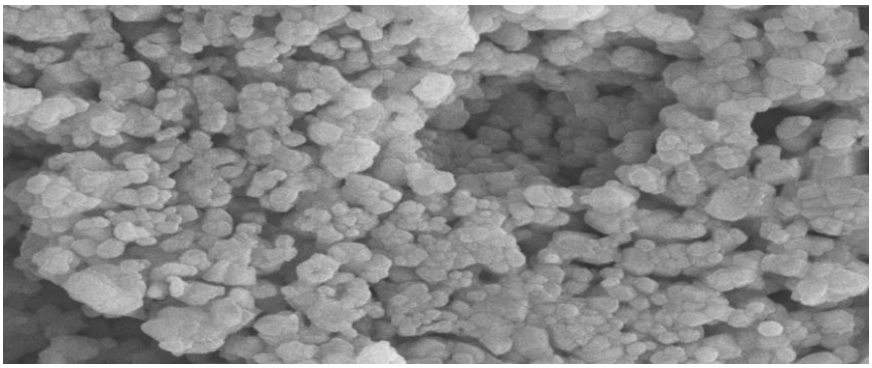

Photo 4: X=0.9 SEM for $Ba_{0.1}$ $Sr_{0.9}$ TiO_3

Table 7: Data on Average grain size

Composition(X)$BaTiO_3$	Average Grain Size
0.3	0.101 µm
0.5	0.214 µm
0.7	0.2303 µm
0.9	0.2283 µm

Above tabulation indicates that, as the composition of ferroelectrics increases, the average grain size also increases.

The SEM micrographs of ferroelectric samples with the four different compositions shown in the photos (1-4) respectively. It is observed that all the samples show fine particles without segregation of impurity and highly dense microstructure with the proper grain growth after sintering. The average grain size was calculated by counting number of grain boundaries intercepted by a measured length of a random straight line drawn on micrographs [3].

Experimental detail for dielectric properties and AC conductivity

a) Dielectric constant and dielectric loss

The final sintered pellet samples were well polished and coated with silver paste on both surfaces for good ohmic contact. This silver pasted pellet samples of ferroelectric are used to estimate the parallel capacitance (Cp) and dielectric loss tangent (tanQ) at room temperature in the frequency range of 0 Hz to1MHz.

The dielectric constant is calculated using the relation,

$$\varepsilon' = \frac{Cpt}{\varepsilon 0 A}$$

Where Cp - parallel capacitance,
t -thickness of the pellet,
A -cross sectional area of the pellet ($\pi r2 = nd2/4$),
ε0-permittivity of free space = 8.854 x 10-12 F/m.

Therefore

$$\varepsilon^{1} = \frac{4\,C\,p\,t}{8.854\times10^{-12}\ x3.142x\ d^{2}}$$

Therefore

$$\varepsilon 1 = \frac{14.38Cpt\ x10^{12}}{d2}$$

Above Equation is used to estimate dielectric constant of ferroelectric samples.

b) AC Conductivity

The AC conductivity (σac) is related to the dielectric relaxation caused by localized electric charge carriers. The frequency dependent AC conductivity is calculated from the dielectric constant (ε') and loss tangent (tanδ) i.e.

$$\sigma ac = \varepsilon' \varepsilon 0\ \omega \tan \delta$$

Where ε' - the relative dielectric constant of the material,
ε0- Permittivity of free space,
tanδ - loss tangent
ω-angular frequency

The frequency response of the dielectric behavior and AC conductivity is known as relaxation spectra or dispersion curve. This curve gives the valuable information about the type of conduction mechanism present in the polycrystalline sintered ferroelectric samples.

RESULTS AND DISCUSSION

Dielectric constant

The variation of dielectric constant with frequency of all the samples is shown in figure.6-17. The dielectric constant decreases rapidly up to 1 kHz due to usual dielectric diversion in low frequency region. Beyond 1khg it almost remains constant in high frequency region. The dispersion is due to Maxwell wages type interfacial polarization (4) in agreement with Koops phenomenon magical theory (5). The maximum dielectric constant at low frequency is due to space charge polarization at grain boundaries, interfacial dislocation pileups, oxygen. Vacancies, grain boundaries &defects (6) for the composition X=0.7 the dielectric constant is maximum it is attributed maximum space charge polarization at the grain boundaries. The Maximum grain size & minimum grain boundary area at X=0.7 results in the increase of mean free path of electron & hence dielectric constant becomes maximum at X=0.7.

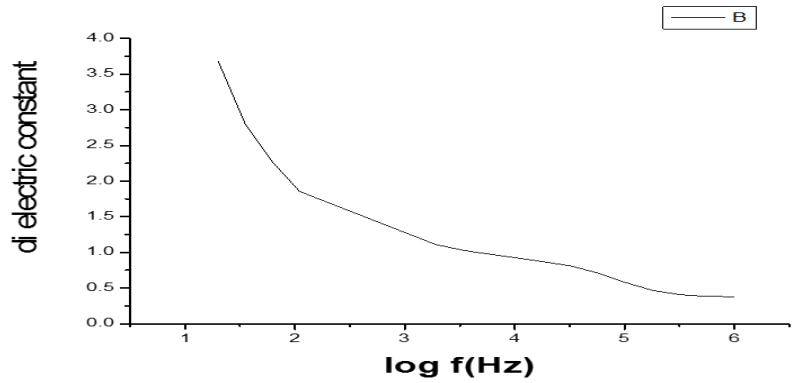

Figure 6: Dielectric constant VS Log frequency (f) for X=0.3 (Ba0.7Sr0.3TiO3)

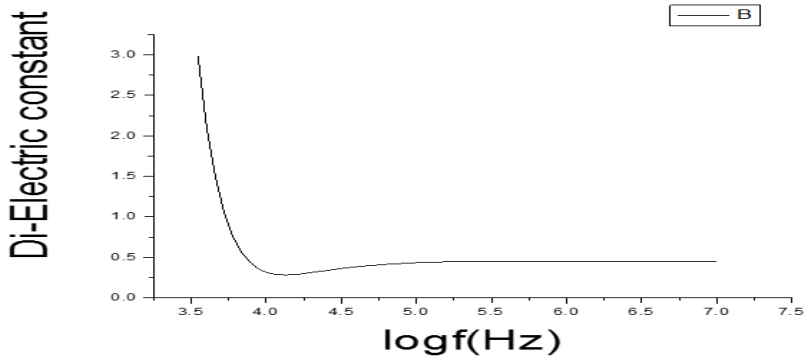

Figure 7: Dielectric constant VS Log frequency (f) for X=0.5 (Ba0.5Sr0.5 TiO3)

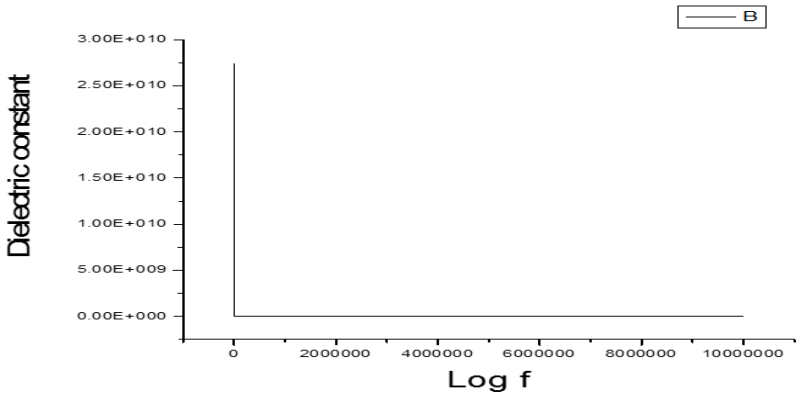

Figure 8: Dielectric constant VS Log frequency (f) for X=0.7 (Ba0.3Sr0.7 TiO3)

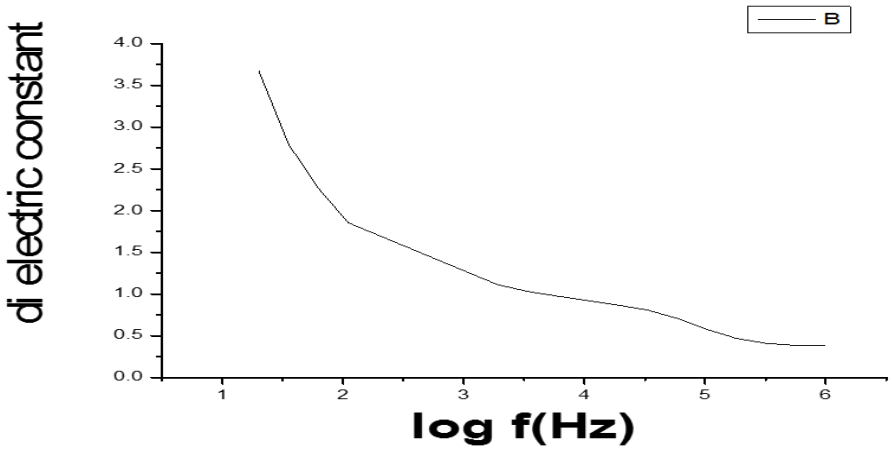

Figure 9: Dielectric constant VS Log frequency (f) for X=0.9 (Ba0.1 Sr0.9 TiO3)

The graph representation loss angle (tanδ) VS Log frequency (f)

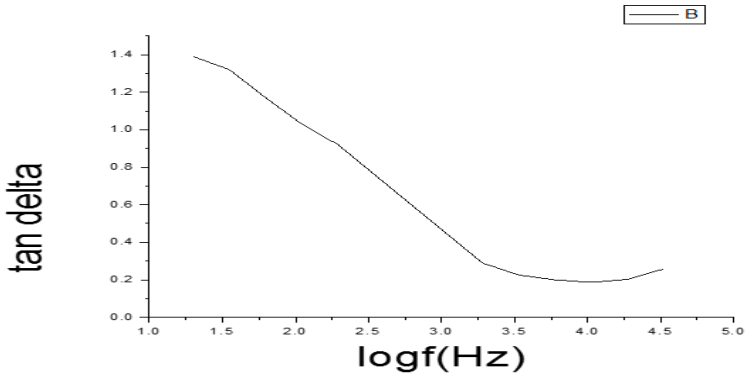

Figure 10: loss angle (tanδ) VS Log frequency (f) for X=0.3 ($Ba_{0.7}Sr_{0.3}$ TiO_3)

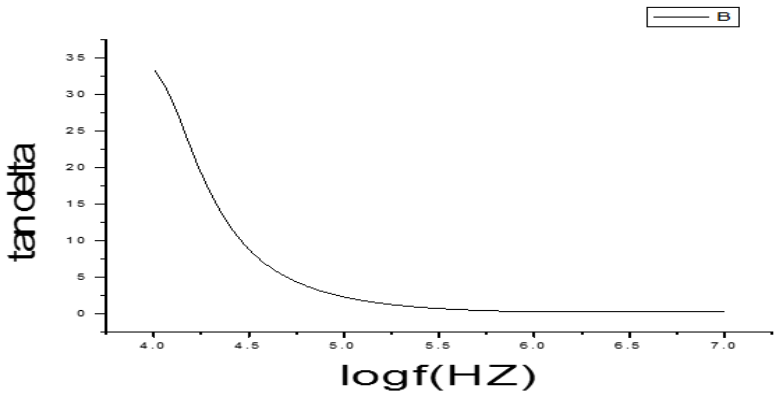

Figure 11: loss angle (tanδ) VS Log frequency (f) for X=0.5 (Ba0.5Sr0.5 TiO3)

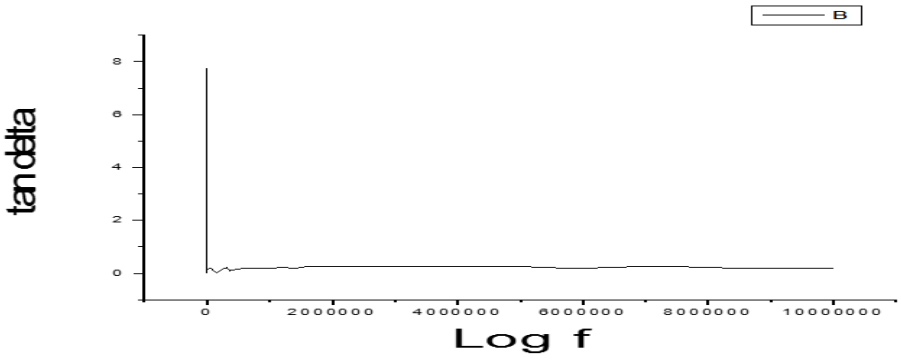

Figure 12: loss angle (tanδ) VS Log frequency (f) for X=0.7 (Ba0.3Sr0.7 TiO3)

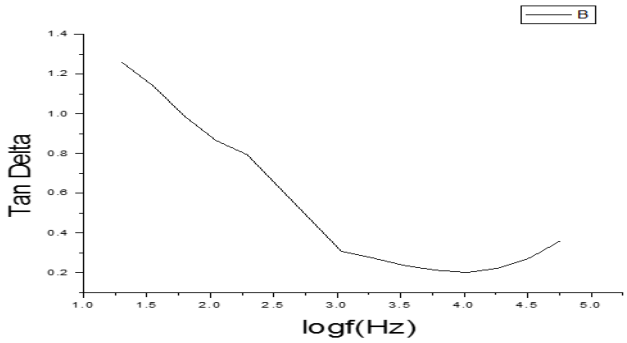

Figure 13: loss angle (tanδ) VS Log frequency (f) for X=0.9 (Ba0.1 Sr0.9 TiO3)

The graph representation AC-Conductivity Slog frequency (f)

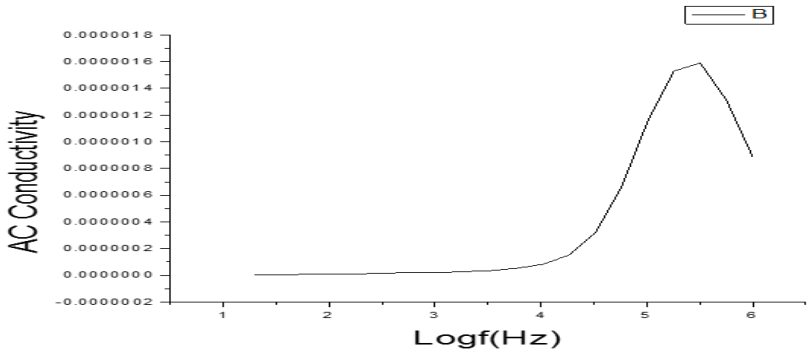

Figure 14: AC-Conductivity VS Log frequency (f) for X=0.3 (Ba0.7 Sr0.3 TiO3)

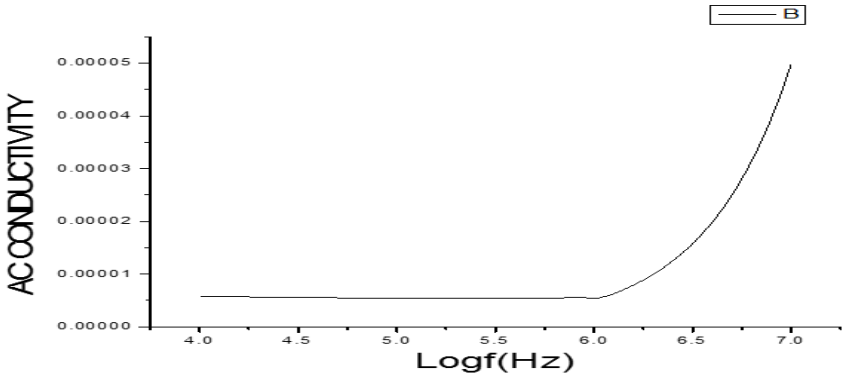

Figure 15: AC-Conductivity VS Log frequency (f) for X=0.5 (Ba0.5 Sr0.5 TiO3)

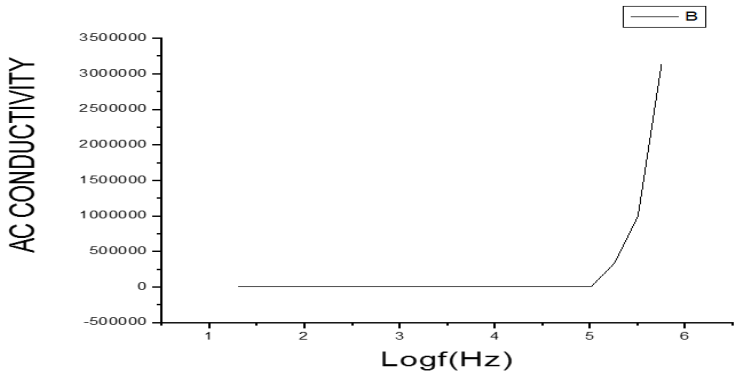

Figure 16: AC-Conductivity VS Log frequency (f)X=0.7 (Ba0.3 Sr0.7 TiO3)

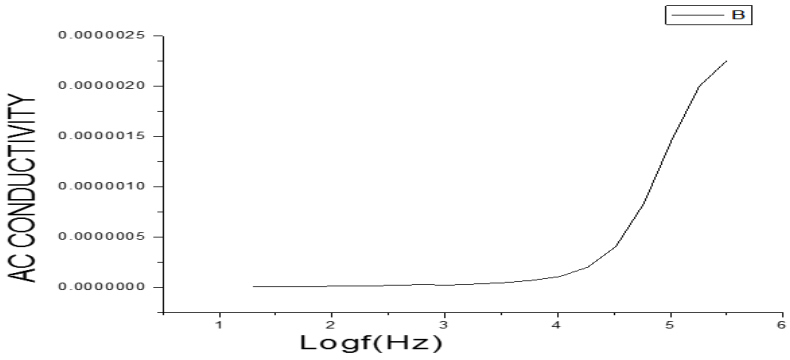

Figure 17: AC-Conductivity VS Log frequency (f) X=0.9 (Ba0.1 Sr0.9 TiO3)

REFERENCES

1. A Safari, R K Panda, V F Janas, Ferroelectric Ceramics: Processing, Properties & Applications.
2. B Jaffe, W Cook, H Jaffe, Piezoelectric Ceramics, Academic Press, London, **1971**.
3. Goel NC, thesis Delhi University, **1977**.
4. CG koops, *Phy Rev*,**1951**,83,121.

5. AJ Bosmon, HJ VanDwal, *Adv phys.*
6. KW Wagner, *Am J Phys*, **1993**,40,8176.