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Structural and Electrical Analysis of Strontium Substituted Barium Titanate

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***Abstract*— Barium strontium titanate ferroelectric with chemical formula $Ba_{1-x}Sr_xTiO_3$ with (x=0.0, 0.1, 0.2 and 0.3) were prepared by standard double sintering ceramic method. The structural, morphological, electrical and dielectric properties of pure and doped BST samples were investigated. Perovskite structure of polycrystalline BST ceramics was determined by X-ray diffraction. The surface morphology of prepared samples was studied to find out average grain size. DC resistivity were determined by two probe method. The DC resistivity against temperature plot shows semiconducting nature. The dielectric properties such as dielectric constant (έ) and dielectric loss (tan□) were studied as function of frequency as well as temperature within frequency 100Hz-1MHz. Temperature dependent dielectric constant were determined for four fixed frequencies 1 kHz, 10 kHz, 100 kHz and 1 MHz. The purpose of this research article is to synthesis the ferroelectric materials which gives high resistivity due to addition of strontium.**

***Keywords*— XRD; SEM; EDAX; Electric properties; Dielectric properties.**

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I.INTRODUCTION

ABO_3 types of ferroelectric materials have been widely used due to their high dielectric constant and low leakage current. Among the materials, the most important examples of ferroelectrics are $Ba_{1-x}Sr_xTiO_3$ (BST) which is developed from $BaTiO_3$ (BT). Due to its pyroelectric, ferroelectric and piezoelectric properties, it has been widely used in technological application, such as high permeability capacitors, ferroelectric memories, pyroelectric sensors, piezoelectric sensors, piezoelectric transducers, PTC thermistors.

When strontium atoms were introduced to A site, it replace barium atoms and phase transition temperature decreases. In case of paraelectric phase, these materials have considerably high dielectric constant and low losses at microwave frequencies.

$Ba_{1-x}Sr_xTiO_3$ (BST) Ceramics possesses high dielectric constant, low leakage current and good thermal conductivity due to these properties, BST have been widely used as dielectric capacitor, PTC, random access memory, infrared detector, gas sensor, humidity sensors [1-7] These BST ceramics are also used to developed microwave devices such as tunable filters, phase shifters, tunable oscillators and delay lines [8-9].

In the present research work barium strontium titanate is chosen ferroelectric phase to make magnetoelectric composite composites as it has high coupling coefficient and piezoelectric coefficients. Consequence of ME composite has any application as detectors, sensors and converters.

I. MATERIALS AND METHOD

The sample of $Ba_{1-x}Sr_xTiO_3$ with x=0.0, 0.1, 0.2, 0.3 were prepared by standard double sintering ceramic method by using A.R grade $BaCO_3$, $SrCO_3$ and TiO_2. In powder form materials were weighed in stoichiometric proportions and thoroughly mixed in agate mortar to form homogenous mixture. This homogenous mixture was presintered at 950^{o}C for 12 hours. The synthesized powder was milled again with polyvinyl alcohol (PVA) as a binder and pressed to form pellets of 10mm-15mm diameter and thickness of 2-3 mm using a hydraulic press. These palletized samples and remaining powder were finally sintered at 1200 ^{o}C for 12 hours in a muffle furnace.

Prepared samples was characterized by X-Ray Diffractometer (Brucker D8 Advance) using Cu–Kα radiation (λ=1.5418 Å) within the 2θ range 20^o-80^o. The lattice parameters were determined by X'pert High Score Plus software. The purity of compositions in ME composite is confirmed by EDAX spectrum (JEOL, Model JED-2300). D.C. resistivity (ρ) measurements were carried out by using a two probe method at different temperature (300^{o}K-1000^{o}K) by step of 5^{o}K. Capacitance (C_p) and loss tangent (tanδ) of sample were measured by Hioki 3532-50 LCR Hi Tester and as a function of temperature from which the dielectric constant (έ) was calculated at fixed frequencies.

II. RESULT AND DISCUSSION

Fig.1 shows the X-Ray Diffraction Patterns of $Ba_{1-x}Sr_xTiO_3$ ceramics with x=0.0, 0.1, 0.2 and 0.3. It exhibits all the characteristic peaks of ferroelectric material without any impurity peak and the most intense peak was observed at (110). The other planes observed are (100), (111), (200), (210), (211), (220), (221), (301). The XRD pattern confirms the formation of pervoskite tetragonal structure of $Ba_{1-x}Sr_xTiO_3$ ceramics without any impurity peaks. The lattice parameters of the sample are calculated

using X'pert high score plus software and recorded in Table.1. It is also observed that with increase in Sr content X-ray density decreases (fig.3.). The crystallite sizes were calculated by using Scherrer formula and are represented in table.1.

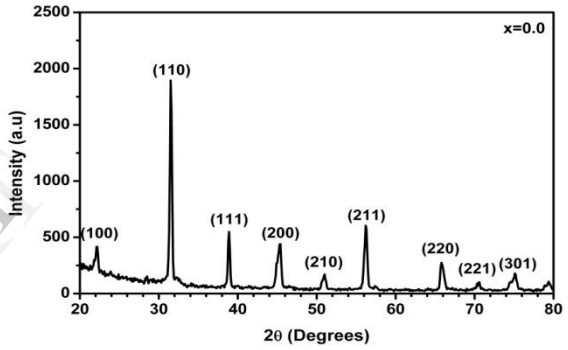
Fig.2.Shows SEM micrographs of $Ba_{1-x}Sr_xTiO_3$ ceramics. The average grain sizes of all the samples were calculated using Cottrell's method and recorded in table 2. It is found that with increase in Sr content the average grain size of sample increases. The variation of average grain size with Sr content is shown in fig.3.

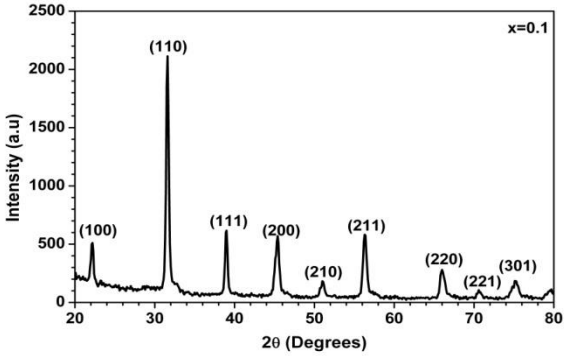
Fig.4. shows EDAX spectrum of $Ba_{0.9}Sr_{0.1}TiO_3$ ceramics. Purity of sample is confirmed by EDAX spectrum. EDAX spectrum shows all the elements (Ba, Sr, Ti and O) without any impurity.

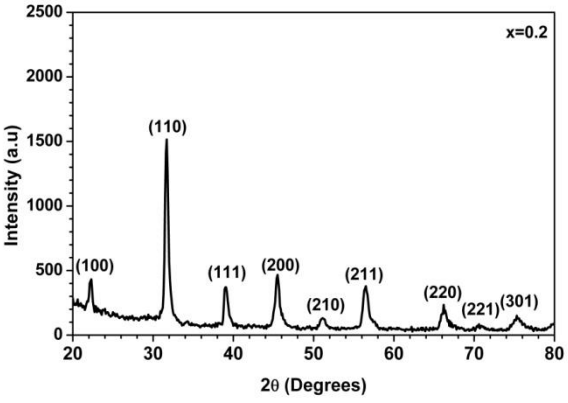
Fig.5 shows the variation of DC resistivity with temperature. DC resistivity graph shows two regions. The first region at low temperature the rate of decreases of resistivity is slow and in second region at high temperature the rate of decreases rapidly showing semiconducting nature of the samples. The first region is attributed to the ordered state of ferroelectric phase and the second region is attributed to the disordered paraelectric state. The electric conductivity observed in BST ceramic may be due to the electron hopping between Ti^{2+} to Ti^{3+} and hole hopping between Ba^{3+} to Ba^{2+} and Sr^{3+} to Sr^{2+}. The decrease in resistivity with the increase in temperature is may be due to the thermally activated drift mobility of charge carriers according to hopping conduction mechanism. For all sample graph shows semiconducting nature decreases rapidly showing semiconducting nature of the samples. The first region is attributed to the ordered state of ferroelectric phase and the second region is attributed to the disordered paraelectric state. The electric conductivity observed in BST ceramic may be due to the electron hopping between Ti^{2+} to Ti^{3+} and hole hopping between Ba^{3+} to Ba^{2+} and Sr^{3+} to Sr^{2+}. The decrease in resistivity with the increase in temperature is may be due to the thermally activated drift mobility of charge carriers according to hopping conduction mechanism. For all sample graph show semiconducting nature.

Fig.6a) shows the variation of dielectric constant (ε) with the frequency. It is observed that with the increasing frequency the dielectric constant decreases rapidly and remains constant at higher frequencies. The high value of dielectric constant observed at low frequencies is explained on the basis of space charge polarization due to inhomogeneous structure i.e. porosity, grain structure and impurities. At higher frequencies dielectric constant remains independent of frequency due to inability of dipoles to follow the applied field. The presence of Ba^{3+} and Ba^{2+} along with Sr^{3+} and Sr^{2+} ions in ferroelectric gives rise to p-type carriers and electronic exchange between Ti^{4+} and Ti^{3+} ions give rise to n-type carriers, however mobility of p-type carrier is smaller than that of n-type carriers. It is also observed that with increase in Sr content dielectric constant increases. From Fig.6b) shows that loss tangent decreases with the increase in frequency. At higher frequencies dielectric loss is reduced and the dipole contributes to the polarization.

Fig.7 shows the variation of dielectric constant with temperature. It is observed that with the increase in temperature, the dielectric constant goes on increasing and reaches the maximum value and then it decreases which indicates the phase transition from ferroelectric state to the paraelectric state. The maximum value of dielectric constant (ε_{max}) corresponds to Curie temperature. This is in good agreement with the results obtained by many workers. With increase in temperature the mobility of charge carriers increases that leads to increase in the conductivity and polarization of the sample and hence increase in dielectric constant. The transition is obtained at a fixed temperature. It is observed that the transition at 1 kHz frequency is very sharp than at other frequencies. It is also observed that with increase in Sr content Curie temperature decreases, similar results are obtained to researchers [10].







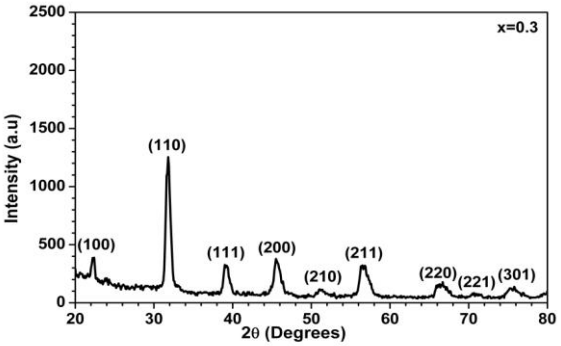


Fig.1. XRD patterns of $Ba_{1-x}Sr_xTiO_3$ ceramics.

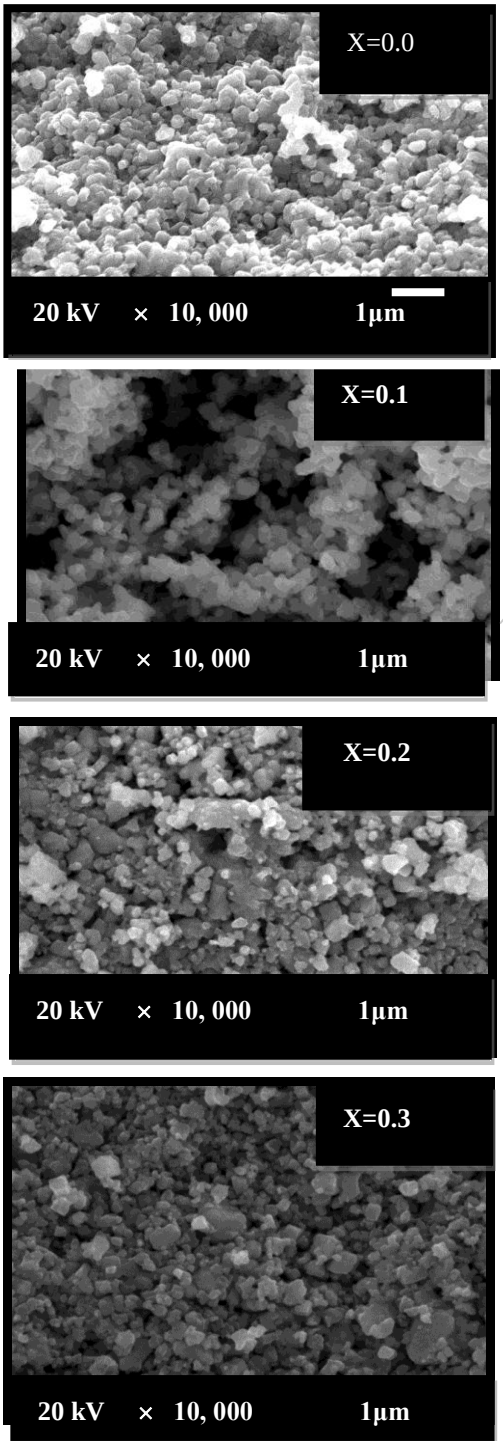


Fig.2. SEM Micrographs of $Ba_{1-x}Sr_xTiO_3$ ceramics.

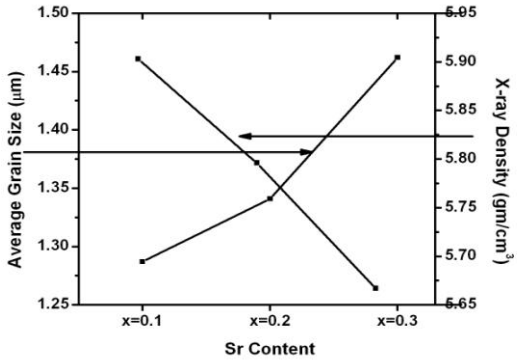


Fig.3. Variation of average grain size and X-ray density with Sr content of $Ba_{1-x}Sr_xTiO_3$ ceramics.

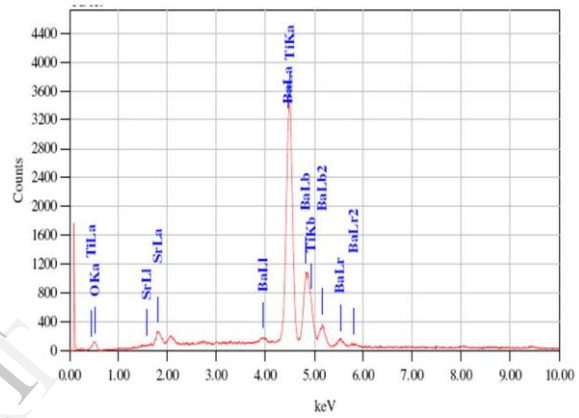


Fig.4. EDAX spectrum of pattern of $Ba_{0.9}Sr_{0.1}TiO_3$

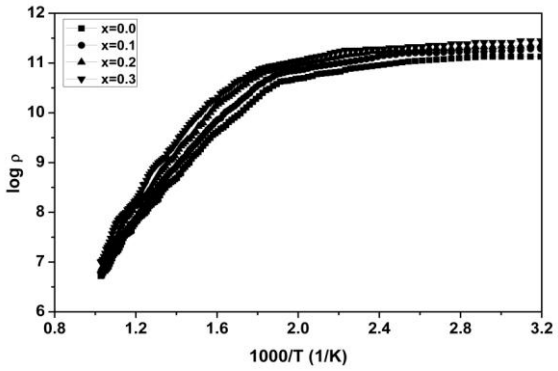


Fig.5.Variation DC Resistivity with temperature for $Ba_{1-x}Sr_xTiO_3$ ceramics.

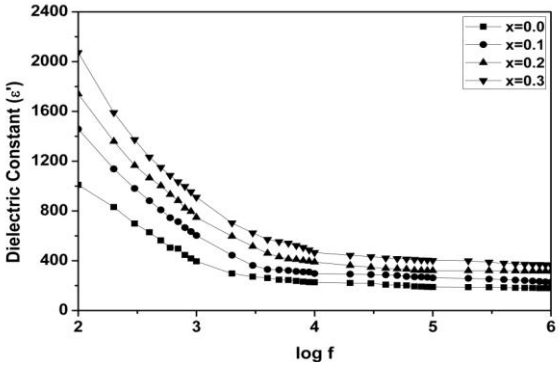


Fig.6a) Variation of dielectric constant (ε) with frequency for $Ba_{1-x}Sr_xTiO_3$ ceramics.

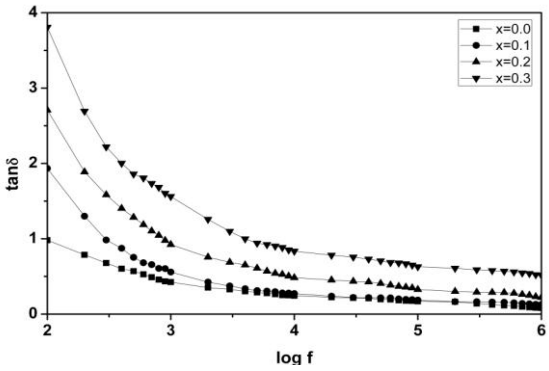


Fig.6b) Variation of dielectric loss (tan□) with frequency for $Ba_{1-x}Sr_xTiO_3$.

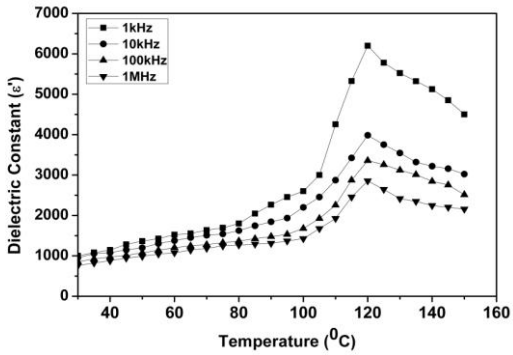


Fig.7a) Variation of dielectric constant (έ) with Temperature for $BaTiO_3$.

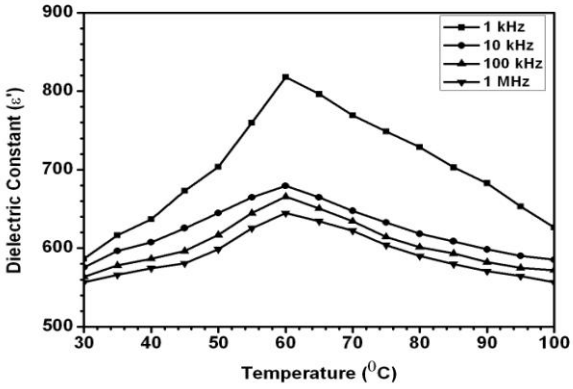


Fig.7b) Variation of dielectric constant (έ) with Temperature for $Ba_{0.9}Sr_{0.1}TiO_3$.

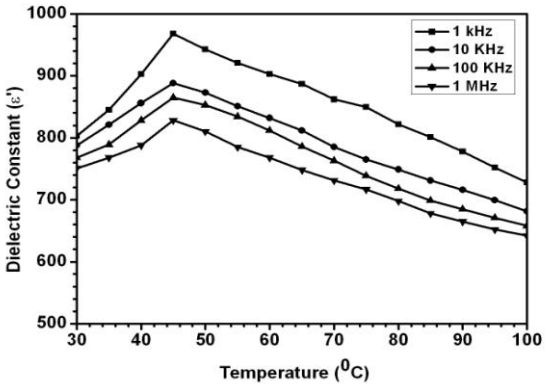


Fig.7c) Variation of dielectric constant □□') with

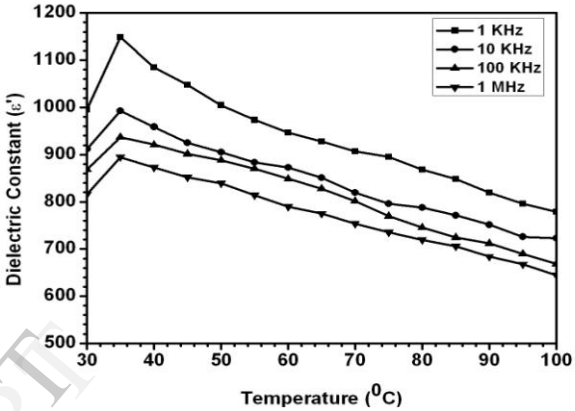


Fig.7d) Variation of dielectric constant □□') with Temperature for $Ba_{0.7}Sr_{0.3}TiO_3$.

Table.1. Lattice parameters of $Ba_{1-x}Sr_xTiO_3$.

Composition	Lattice Parameter			X-ray Density (gm/cm^3)
	c	a	c/a	
X=0.0	4.039	3.998	1.010	5.998
X=0.1	4.027	3.993	1.009	5.903
X=0.2	4.024	3.987	1.009	5.796
X=0.3	4.022	3.988	1.009	5.667

Table.2. Electrical and dielectric data of $Ba_{1-x}Sr_xTiO_3$.

Compo-sition	Crystallite Size (nm)	Average Grain Size (μm)	$\rho_{Tc} \times 10^{11}$ (Ω cm)	T_C (^oC)	$\acute{\varepsilon}_{Tc}$ 1kHz
X=0.0	55.89	0.870	1.34	120	6201
X=0.1	41.99	1.287	1.92	60	817
X=0.2	21.00	1.341	2.40	45	967
X=0.3	21.01	1.462	2.78	35	1148

I. CONCLUSION

The XRD pattern show well defined peaks without any impurity. The sharpness and intensity of XRD pattern indicate that prepared BST sample is crystalline with the formation of pervoskite tetragonal structure. Average grain size was found to be increase with increase in Sr content. EDAX spectrum confirms purity of prepared sample. The resistivity decreases linearly with temperature, however a break in the linear variation is observed for all the samples. The break corresponds to respective Curie temperature indicating a slope change. The change in the slope indicates a change in activation energy which suggests conduction is due to hopping of electrons. It is also observed that with increase in Sr content resistivity of BST increases. The dielectric constant is very large at low frequencies while it is independent of frequency beyond 10 kHz due to inability of dipoles to follow the applied field. The large values of dielectric constant at lower frequencies are explained on the basis of space charge polarization due to inhomogeneous dielectric structure and resistivity of the samples. Increase in Sr content shifts the Curie temperature towards room temperature.

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