Effect of Different Strontium Content on Dielectric Properties of Barium Strontium Titanate Ceramic

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***Abstract*: This paper discusses the effect of different Strontium content on the transition of Barium Strontium Titanate solid solutions. Doping of ceramics is one way to enhance the electrical properties. BST, PZT and other ferroelectrics of interest are already studied and considered for random access memories. Shifting of curie temperature is related to the strontium content in the BST ceramic [1]. Samples were prepared for x = 0.10, 0.15, 0.20 and 0.22. Solid state reaction method is used for preparing different compositions. Pellets with diameter 8 mm using dia piston were prepared. For these pellets, the dielectric properties were observed between room temperature and 126 ^{o}C, for different Sr content. X-Ray powder diffraction study shows the structural and crystal symmetry for different strontium content. Transition of curie point, towards room temperature is observed with increasing strontium content. Low and fairly constant dielectric loss, near room temperature is observed, which is suitable for dynamic random access memory (DRAM) cell. Furthermore, composition dependent dielectric conductivity plot shows different region of conduction process. Study shows BST ceramics, with increasing Sr content are very important candidate for a wide range of applications.**

***Keywords*: Barium Strontium Titanate, Electrical properties, Ceramics, Ferroelectric transitions, Solid state reaction.**

I. INTRODUCTION

Ferroelectric materials of $BaTiO_3$-based solid solutions with perovskite structure are primarily utilized in the electronic industry [2, 3]. There are several other important ferroelectric materials which were studied such as $PbTiO_3$, $SrBiTaO_3$, $Pb(MgNb)O_3$ and $BaTiO_3$, which is the origin of $(BaSr)TiO_3$ [4]. BST is a material with high degree of crystallinity, high dielectric constant, high charge storage capacity and low leakage current at curie's temperature [5, 6]. In particular, BST ceramics are considered most suitable dielectric material for high density capacitors in DRAMs [7, 8]. Most of the microwave application requires material with low dielectric loss and dependence on temperature, of the dielectric constant. It is already reported that BST material exhibit high dielectric loss in microwave range. For the BaO-SrO-TiO_2 system, a 50% medium line in the phase diagram shows the unlimited solubility between $BaTiO_3$ and $SrTiO_3$ [1]. Therefore different Sr content BST materials are required to be investigated. The transition temperature of $BaTiO_3$ is 120 ^{o}C [9]. It is reported that the doping of strontium in $BaTiO_3$ ceramic has a great effect on the transition temperature [1]. When strontium atoms were introduced to A site in perovskite barium titanate matrix to replace barium atoms, the phase transition temperature of paraelectric to ferroelectric decreases. The curie temperature of BST can be varied for different strontium contents, that changes, the phase of the BST ceramics [10]. It is observed that, size of Ba and Sr also have major impact on the electrical properties of BST ceramic [1, 11]. So many methods have been focused on the processing methods as precipitation [12], sol-gel [13, 14], hydro thermal [15] and conventional methods.

II. EXPERIMENTAL

Barium Carbonate ($BaCO_3$) and Strontium Carbonate (Sr_2CO_3) were taken in the molar ratio 1-x : x, milled with Titanium Dioxide (TiO_2), where Sr content is varied for x= 0.1 to x=0.22. Conventional solid state reaction method was used to prepare different compositions. Reagent grade barium carbonate, strontium carbonate and Titanate of high purity 99.0% were used as starting materials. The raw material was dried at 150 ^{o}C for one hour, weighed in stoichiometric proportions, manual grinding in acetone for 2 hr (wet), 4 hr (dry), and then calcinated at 1000 ^{o}C for 4 hr. After calcinations the obtained powder was taken for XRD analysis. XRD analysis was obtained for intensity versus angle (2θ, in the range between 20 to 70). Pellets were prepared at a pressure of 2GPascal, with diameter 8 mm dia piston. These pellets were sintered at temperature 1250 ^{o}C for 4 hrs. Sintered pellets were crushed into powder and again the crystalline orientation was measured by X-ray diffraction (XRD), on a X-Pert PANalytical diffractometer using a fixed divergence1/2^{o} slit, a Cu W/Si mirror , a solar slit and a 10 mm mask as incident optics and a parallel plate collimator, a solar slit and a proportional detector as diffracted beam optics. The result from the machine indicates that only pure perovskite structure exists for all BST ceramics with different Sr content. Sintered pellet was coated both side, with silver paste. The coated pellet was dried for 2 hr at room temperature (25 ^{o}C). A computer controlled system, including RCL meter (make: Fluke, model: PM6306) was used to measure temperature and frequency dependence of dielectric properties. Both low frequency (1 KHz) and high frequency

(1MHz) dielectric measurement were performed using this programmable RCL meter.

III. RESULTS AND DISCUSSIONS

XRD ANALYSIS

Crystallinity determined using a X-RAY diffractometer (XPERT-PRO, PW40) with a CuKα radiation source (λ=1.54059 A^O), operated at a voltage of 45kV and a current of 40 mA. XRD patterns of BST for Sr content x = 0.10, 0.15, 0.20 and 0.22 are shown in Fig. 1. It can be observed from the figure that, the diffraction peaks are (1 0 1), (1 1 1), (0 0 2) and (2 1 1) within the 2θ range from 20^o to 66^o. As depicted in figure 1, the θ/2θ X-ray diagram shows that the deposited BST ceramics is crystalline in perovskite phase. The pattern for all Sr content x=0.10, 0.15, 0.20 and 0.22 is crystalline in nature and well oriented. The crystal structure exhibits tetragonal symmetry for all Sr content, that are under observation [16]. In all of them a single phase perovskite solid solutions, is observed.

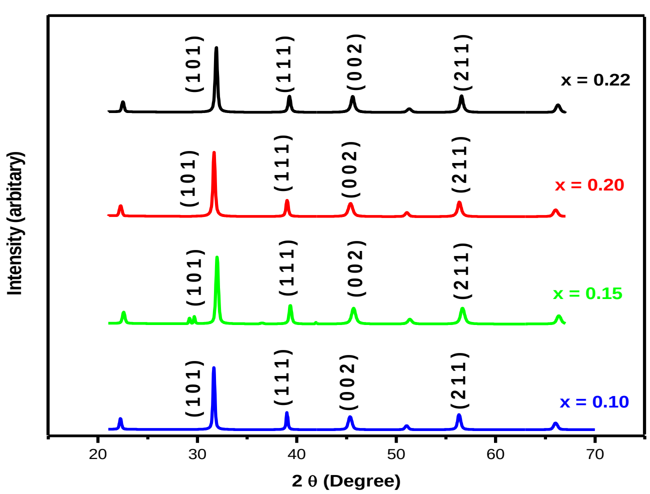


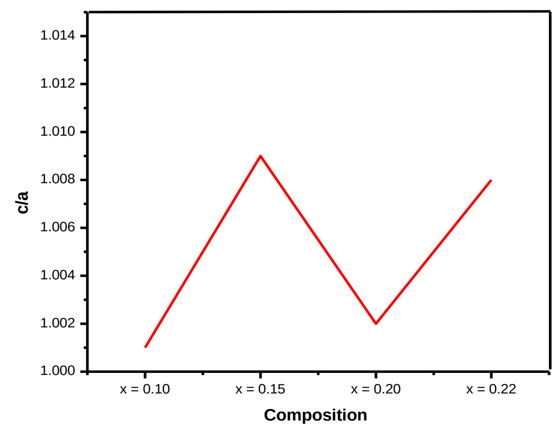
Fig. 1. XRD pattern for different strontium content (x = 0.10, 0.15, 0.20 and 0.22)

Table - 1

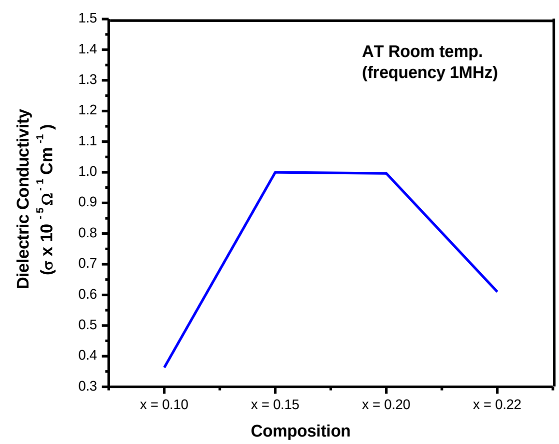
Lattice parameter of BST ceramics, for Sr content (x = 0.10, 0.15, 0.20 and 0.22).

Sample	a(A°)	c (A°)	c/a	Structure phase
BST x = 0.10	3.9932	3.9996	1.001	Tetragonal
BST x = 0.15	3.9390	3.9780	1.009	Tetragonal
BST x = 0.20	3.9937	4.0034	1.002	Tetragonal
BST x = 0.22	3.9825	3.9504	1.008	Tetragonal

Table-1 depicts that the c/a ratio is larger than 1(the c-axis lattice constant is larger than the a-axis lattice constant). The analysis indicates that the crystalline structure is tetragonal at room temperature for different Sr contents (x = 0.10, 0.15, 0.20 and 0.22). Fig. 2(a) indicates that c/a ratio is changing with the composition. The change in the c/a ratio have a direct effect on dielectric constant value.



(a)



(b)

Fig. 2. Variation of (a) c/a and (b) dielectric conductivity versus composition for different content at room temperature

ELECTRICAL MEASUREMENTS

Fig. 2(b) is the observation for variation of dielectric conductivity for different Sr content, for frequency 1MHz, at room temperature. In order to characterize the hooping dynamics of the charge carrier/ions, the frequency dependent conductivity is observed as shown in Figure 3. Conductivity is usually calculated by the relation given below [17].

$$\sigma = \epsilon_0 \omega K tan\delta$$

Where ε_o is the permittivity of free space, $\tan\delta$ is dielectric loss and $\omega = 2\pi f$, f is the applied frequency. It is observed that at lower frequencies the conductivity is frequency independent. In the mid region the conductivity varies linearly with the frequency. The conduction behavior is related to translational hooping motion [18, 19]. At higher frequency also conductivity increases linearly with frequency. Which indicate that the conduction mechanism in high frequency range related to the well-localized hooping motion [18, 19].

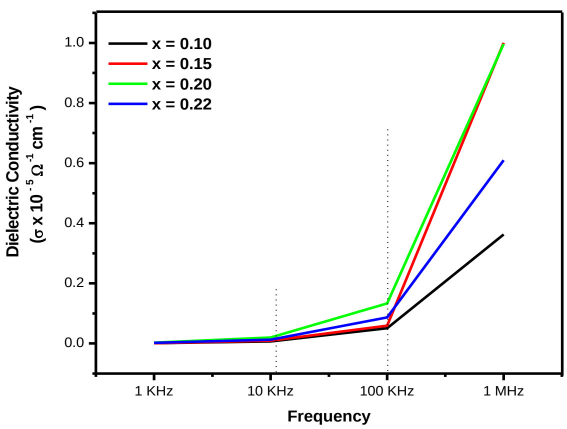


Fig. 3. Conductivity (σ) as a function of frequency for different Sr content at room temperature

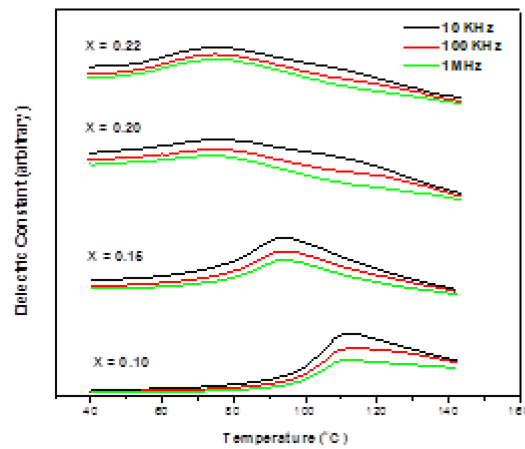
Fig. 4 shows the dielectric constant curve with temperature from 40 ^oC to 140 ^oC, for different Strontium contents. It is observed that the curie temperature is shifted towards the room temperature as the strontium content is increased as x = 0.10, 0.15, 0.20 and 0.22 (Table 2). This is an important behavior of BST ceramic which makes its utilization near room temperature. Sensors for applications, where the thermal energy and other parameters are varying near room temperature, can be developed.

Table 2

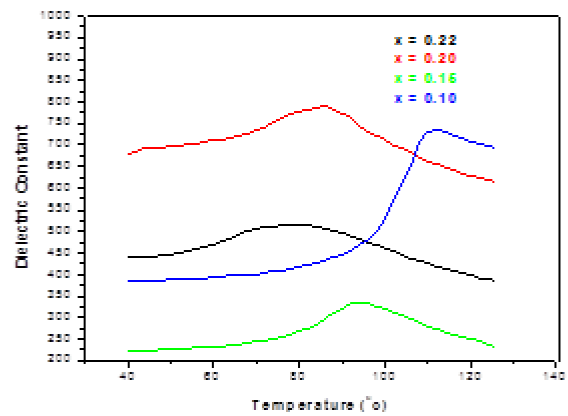
Transition of curie temperature for different Sr content

Sample	Curie Temp. (^oC) ϵ_r	Peak value	Room
BST x = 0.10	112	728	378
BST x = 0.15	94	338	225
BST x = 0.20	86	790	680
BST x = 0.22	78	522	437

Fig. 5 indicates the loss tangent for different compositions in the temperature range 40 ^oC to 126 ^oC. It is observed that, near room temperature the loss tangent is minimum for x = 0.22. This implies that the leakage current will be small, that can enhance its application in memory applications. The dielectric loss decreases with the strontium content, which increase the stability of the BST ceramic.



(a)



(b)

Fig.4. Dielectric Constant with Temperature for different (a) frequency and (b) compositions

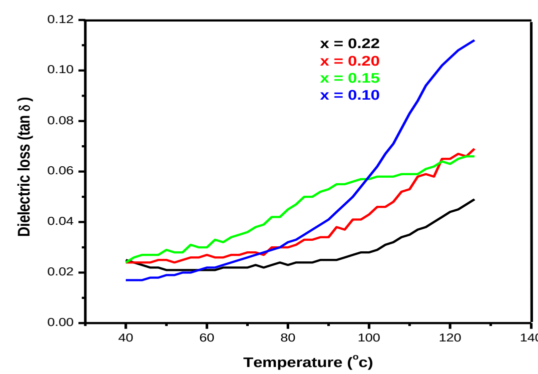


Fig. 5. Dielectric loss with temperature for different compositions

Fig. 6 shows the dielectric constant value for different strontium compositions from low frequency (1 KHz) to high frequency (1 MHz) range. It is observed that the dielectric constant remains nearly constant for entire frequency range. Fig. 7 shows the loss tangent variation for different strontium composition in the low frequency (1 KHz) to high frequency (1 MHz) range. It is evident from the figure that at lower frequency for all compositions the loss tangent is higher but at higher frequency range the loss tangent is small. From all the observations it is observed that the increased strontium content makes BST a promising ceramic candidate, near room temperature for various applications.

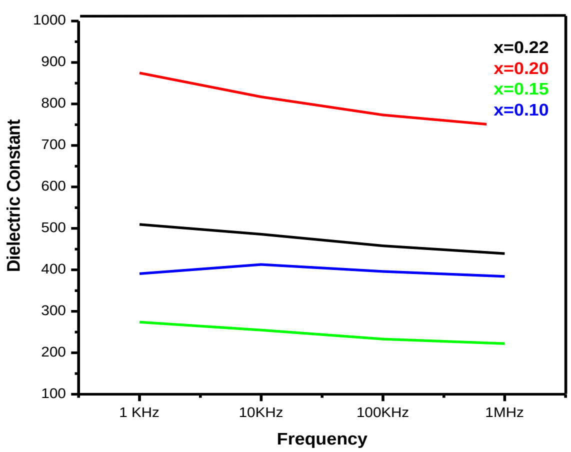


Fig. 6. Dielectric Constant with Frequency for different compositions

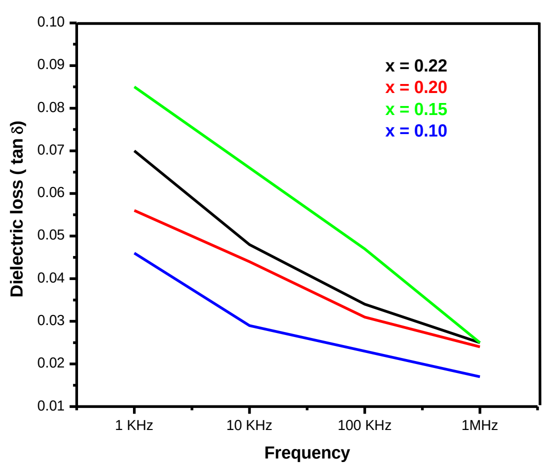


Fig. 7. Dielectric loss with frequency for different compositions

IV. CONCLUSION

Perovskite-type $Ba_{1-x}Sr_xTiO_3$ ceramics with different x values (x = 0.10, 0.15, 0.20 and 0.22) have been prepared using solid state reaction method from raw materials. XRD patterns for all Sr content x = 0.10, 0.15, 0.20 and 0.22 are found crystalline in nature and well oriented. The crystal structure exhibits tetragonal symmetry for all Sr content. In all structure a single phase perovskite solid solutions, is observed. Electrical measurement confirms the transition of curie temperature towards room temperature for increasing Sr content. Curie temperature near room temperature makes BST a promising candidate for sensor application. $Ba_{1-x}Sr_xTiO_3$ material with Sr content x = 0.22, indicates low and nearly constant loss tangent near room temperature, which makes it suitable for many applications.

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