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INFLUENCE OF DYSPROSIUM ON THE DIELECTRIC PROPERTIES OF BISMUTH LAYERED STRUCTURED PIEZOCERAMICS

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ABSTRACT
Piezoelectric ceramics are used for the applications of piezoelectric sensors, actuators and transducers. Bismuth Layered Structured Ferroelectric (BLSFs), a member of the Aurivillius family, has been studied for its application in FeRAMs (non-volatile Ferroelectric Random Access Memory). Besides the above usage, BLSFs also have their functions in electronics such as high-temperature sensors, resonators and other similar devices. In regards to its application as resonators, piezoelectric materials are used as inductors since BLSFs have higher operating temperatures as compared to lead-based materials such as PZT. Therefore BLSFs are ideal alternatives to PZT kind of lead-based materials for piezoelectric applications such as sensors at high temperatures. $SrBi_4Ti_4O_{15}$ (SBT) compounds, a four-layered member of the family of Aurivillius BLSFs are rare earth modified materials prepared by adding Dysprosium in A site ($Sr_{0.2}Na_{0.4}Dy_{0.4}Bi_4Ti_4O_{15}$) (SNDBT). They are prepared by a conventional double sintering method using planetary wet Ball Milling. XRD revealed the formation of a single phase with an orthorhombic structure. Scanning Electron Microscopy (SEM) is used for sample microstructure and grain size studies. The dielectric loss and dielectric constant at various temperatures and frequencies are studied.
Keywords: Piezoelectric, Bismuth Layered Structured Ferroelectrics (BLSFs), Curie temperature, dielectric constant

INTRODUCTION

$SrBi_4Ti_4O_{15}$ (SBT) is one of the examples of the BLSF family. The general formula of BLSF is $(Bi_2O_2)^{2+}(A_{m-1}B_mO_{3m1+1})^{2-}$, here $(A_{m-1}B_mO_{3m1+1})^{2-}$ is the perovskite layers and m denoting a number perovskite layers and A take place monovalent, divalent, trivalent ions , B takes place a Ti^{+4} tetravalent, Nb^{+5} pentavalent and W^{+6} hexavalent ions[1–3]. Their high transition temperature or Curie temperatures and low dielectric loss make BLSFs quantitatively usable. SBT has four perovskites of the same form TiO_6 (octahedral) cells sandwiched in between $(Bi_2O_2)^{2+}$layers. The compound possesses orthorhombic symmetry and the space group of the compound is $A2_1am$[4].

The changes in its different properties by the addition of oxides of rare-earths into another oxide compound are obviously understood with regard to the change in ionic radii. In electro-ceramic materials, for example, PZT (Lead Zirconate Titanate), the adding of a rare earth ion which maybe off-valent (e.g. La^{3+}) it observed affect of its electrical in that way physical properties. A few PZT doped with La (PLZT) structure has produced ceramic materials with excessive density and appropriate clearness, making them appropriate in order to electro-optic applications[5]. It is found that in $Bi_4Ti_3O_{12}$ (BIT) and $SrBi_4Ti_4O_{15}$ (SBT) doped with La, Sr^{2+} ions, that are greater stable act as La^{3+} and accordingly decreasing

the acceptability of La^{3+} into perovskite layers. The presence of Sr^{2+} ion at the A site of $SrBi_4Ti_4O_{15}$, can also result in the blending of $Dy3^{+}$into the Bi_2O_2 layers at low doping contents. For non-lead electro-ceramics, *Yamaji* et.al[6] has investigated the impact of doping Dy in $BaTiO_3$. It is discovered that in doped ceramics, the common grain sizes of are smaller, wherein as the dielectric constants are greatly that of un doped $BaTiO_3$ higher than an extensive variety of temperatures[7-10].This paper reports the Microstructure Dielectric constant, Curie temperature and Dielectric loss of the substitution of Na^{+1} and Dy^{+3} in place of Sr^{+2} in SBT.

EXPERIMENTAL

SNDBT ceramics are prepared from commercially available carbonates of Strontium and Sodium, oxides of Dy, Bi and Ti (Sigma Aldrich) using the solid state double sintering technique. The chemical ratios as per stoichiometry are accurately weighed and grounded by way of planetary wet ball mill in distilled water medium for 5h. The mixture is next calcined at 800°C up to 2 hours.[11-12.] The calcined chemical is once more grounded up to 5h. Then pellets are cast using a hydraulic press of 10Mpa pressure by adding 1% PVA (polyvinyl alcohol) as if binder. The thickness of the pellet is 0.1 cm and 1.10 cm diameter and final sintering temperature are around 1150°C for about 2h. The density of the pellet is estimated using the Archimedes principle. Using XRD (X-Ray Diffraction) (Philips X-pert PRO). The sintered powder is evaluated for its crystalline properties. The microstructure was found through a Scanning Electron Microscope (Carl ZEISS EVO18). Conductive gold paint is coated on both sides of the material and the dielectric loss and dielectric constant as if the function of frequency from room temperature to up to 700°C is measured using Wayne-Kerr 6500P high-frequency LCR meter.

RESULTS AND DISCUSSION

XRD Studies

SNDBT ceramic diffraction patterns matched with standard $SrBi_4Ti_4O_{15}$ phase with a slight shift in peaks. Single phase form with a orthorhombic structure is determined by XRD (Fig.-1).

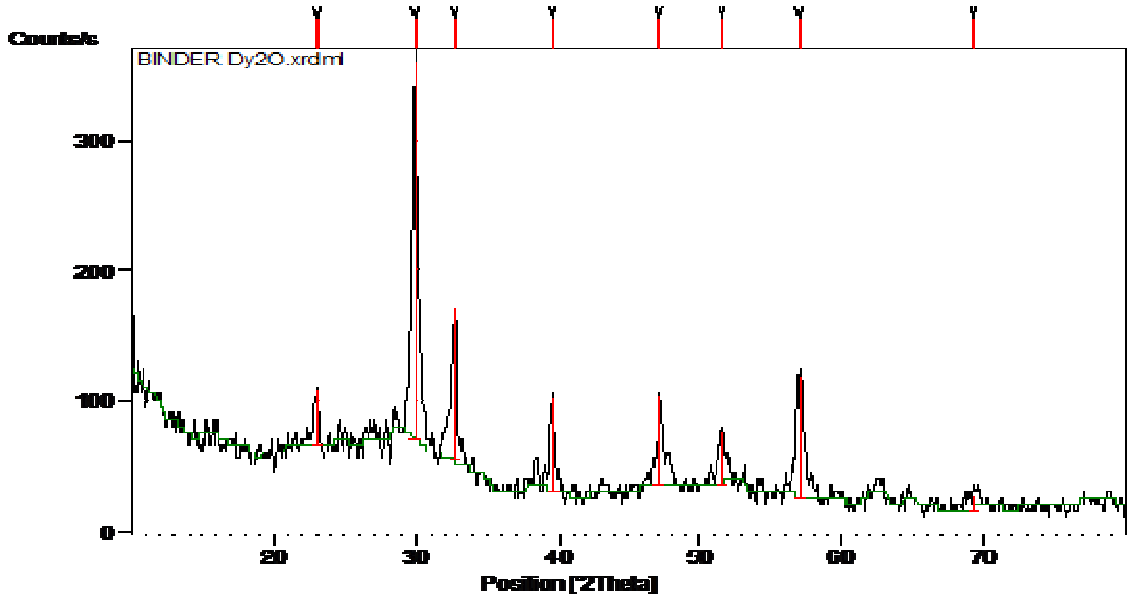

Fig.-1: The X-ray Diffraction Pattern of SNDBT Ceramics

Scanning Electron Microscopy Studies

Figure-2 is given the SEM micrograph of the fractured surface of SBT and SNDBT ceramics. Plate-like grains with grain sizes between 100 and 200 nm is observed. All samples possess a relatively dense structure. The samples have plate-like grains in random orientations. SEM pictures reveal that the compound ceramics has a good density.

The densities are calculated by the liquid displacement method based on the Archimedes principle. Distilled water is used as liquid media. Experimental density of 7.194 g/cm^3 is obtained.

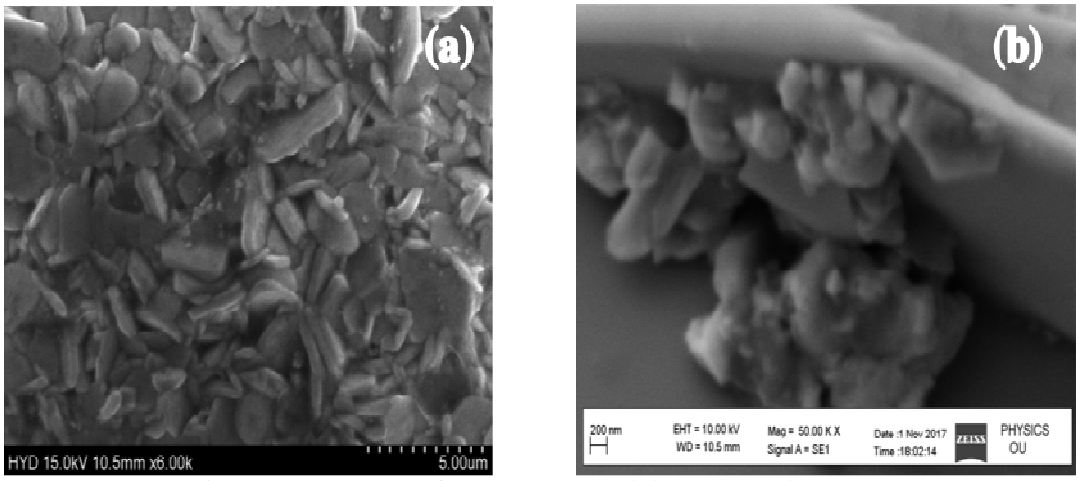

Fig.-2: SEM Images of (a) Pure SBT and (b) Dy-Doped SNDBT

Dielectric Properties

The dielectric constant of SNDBT ceramic as if the function of both temperature and frequencies (1 kHz and 1 MHz) is shown in Fig.-3. It can be seen as the dielectric constant value is increasing like the temperature increases and frequency decreases. A strong peak is observed at 671°C at all frequencies. This consistent with Curie temperature, T_c. For $SrBi_4Ti_4O_{15}$ (without any dopants), the Curie temperature is 535°C[13-14]. With the substitution of Na^{+1}Sodium and Dy^{+3} Dysprosium in place of Strontium, the Curie temperature was found to increase. The dielectric constant values at various frequencies are given in table1. The increased T_c confirms the distortion of Oxygen octahedral. The T_c depends on ionic displacements. This ionic displacement depends on ionic polarizability, ionic size, and tolerance factor. The degree of distortion can be evaluated by a tolerance factor (t) [15]. According to the *Goldsmith relation*[14], the *tolerance factor* (t) is:

$$t = R_A + R_O / \sqrt{2}(R_B + R_O) \quad (1)$$

Where, R_A is ionic radii of A-site ions and R_B is ionic radii of B- Site ions. The tolerance factor value (t) generally lies between 0 and 1.

The ionic radius of oxygen is denoted by R_O. The tolerance factor of pure SBT is 0.938.With the substitution of Dy^{3+}, the *tolerance factor* reduced. According to the theory of *Donaji*[16], there is an increase in the Curie temperature with a decrease in tolerance. Figure-3 shows the dielectric loss of $Sr_{0.2}Na_{0.4}Dy_{0.4}Bi_4Ti_4O_{15}$ ceramic as a function of temperature at various frequencies (1 kHz-1MHz).

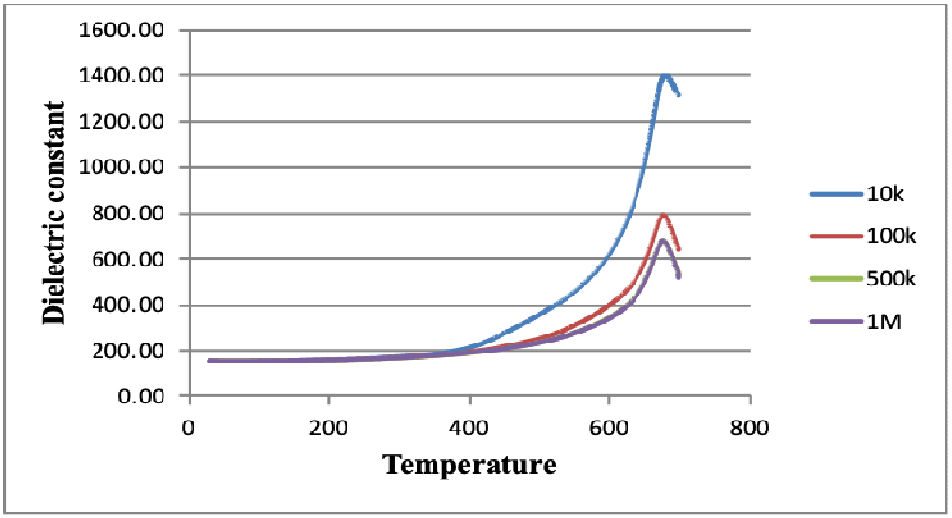

Fig.-3: Dielectric Constant (Ɛ) of SNDBT as a Function of Temperature at Different Frequencies

Table-1: Dielectric Constant at Different Frequencies

S. No.	Sample	Dielectric Constant at RT		Dielectric Constant at T_c		T_c	Dielectric loss
		At 10 kHz	At 100 kHz	At 10 kHz	At 100 kHz		
1	SBT	234	234	2668	1971	535	0.021
2	SNDBT	160	160	1390	785	676	0.013

The temperature and frequency dependent dielectric loss of SBT modified with Dysprosium is shown in Fig.-4. There is a slight decrease in the dielectric loss with the introduction of Dysprosium in SBT ceramics. The reduction is attributed to the cationic vacancies that are formed which in turn suppress the oxygen vacancy formation. This further leads to a reduction in space charges. Thus a lower dielectric loss of doped SBT ceramics is observed[17,18]. The dielectric loss is very low, almost nearing zero till 400°C. With temperature increasing beyond 400ºC, the dielectric loss increased.

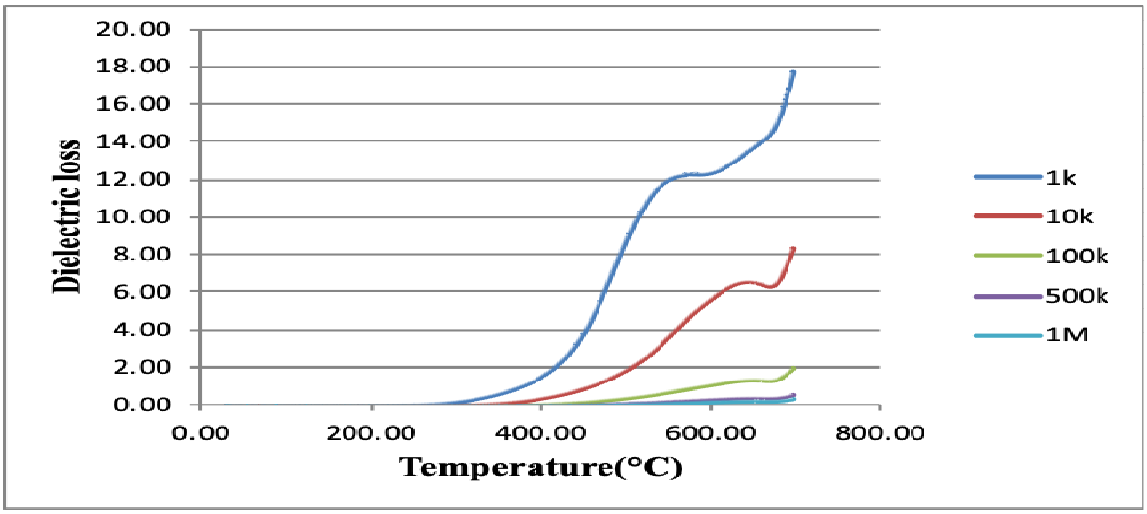

Fig.-4: Dielectric Loss as a Function of Temperature for SNDBT Ceramic

CONCLUSION

Polycrystalline $Sr_{0.2}Na_{0.4}Dy_{0.4}Bi_4Ti_4O_{15}$ ceramic pellets are synthesized following the solid state sintering technique by using planetary wet ball mill. Single Phase is formed which is confirmed by XRD. Curie temperature increased from 535°C for pure SBT to 671°C with modification of Na^{2+} and Dy^{3+} doping. The dielectric constant is 1350 at 10 kHz at the observed Curie temperature. The results obtained indicate that the as-synthesized materials are excellent ceramics for piezoelectric applications.

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