

## Structural, Transport and dielectric property studies of $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$ ceramic

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### ABSTRACT

A New ceramic compound  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  has been synthesized by solid state reaction method. Preliminary structural analysis (XRD) shows that there is formation of single phase compound. SEM study with different magnifications provided an insight of uniform grain size distribution of the sample. Electrical and dielectric properties have been measured in the frequency range 42Hz – 5MHz in the temperature region 27-450 °C.

**Keywords:** Dielectric properties, microstructure, X - ray diffraction.

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### INTRODUCTION:

Ceramics, which exhibit very specific properties and cannot be replaced by other materials, play a significant role in the performance of numerous devices and equipments. Although there has been considerable amount of work done in the area of dielectric oxide in the past fifty years, dielectric materials continue to remain a hot area of scientific research due to their wide ranging application.

Since the discovery of ferroelectric materials and its related properties in  $\text{BaTiO}_3$  (1), a large number of oxides in different forms have been studied using various experimental techniques for the search of new materials for devices application. The material with a perovskite structure of general formula  $\text{ABO}_3$  (where A = mono or divalent ions, B = tri, tetra and penta-valent ions) have been found to be very useful and interesting for various solid state device (2-4) By making suitable substitution at A site and B site of  $\text{ABO}_3$  structure, a large number of charge neutral or deficient compound have been prepared (5-7) which have been found to be very suitable and useful for many industrial applications such as computer memories, display sensors, electro optical modulator etc. Studies of ferroelectric properties of Sn modified  $\text{BaTiO}_3$  (i.e.  $\text{BaSn}_{0.5}\text{Ti}_{0.95}\text{O}_3$ ) (8) showed that it has phase transition above room temperature with a high dielectric constant. It is well known that a suitable substitution at different atomic sites of any ferroelectric structure usually has dramatic effect on physical properties of material. Substitution

of different lanthanides doped material at Pb sites and zirconium at Ti site (Double doping) in PZT (9) has provided many solid solutions with interesting properties for wide industrial applications.

In 2006 Xiao-Bing Chen and co-worker [10] investigated ferroelectric and dielectric properties of bismuth layered structural  $\text{Sr}_2\text{Bi}_{4-x}\text{Ln}_x\text{Ti}_5\text{O}_{18}$  (Ln = La, Nd, Sm and Dy) ceramics, they reported that the ferroelectric property of  $\text{Sr}_2\text{Bi}_4\text{Ti}_5\text{O}_{18}$  is obviously improved by La, Nd, Sm, or Dy doping with appropriate content  $\text{Ca}_4\text{Bi}_2\text{Ti}_4\text{Nb}_6\text{O}_{30}$  ceramic also showed this type of behavior reported by H. Sharma, and co-workers [11] Neurgaonkar and their groups [12] reported structural and dielectric properties of  $\text{Pb}_{1-2x}\text{K}_x\text{M}_{x+3}\text{Nb}_2\text{O}_{30}$  (M= La or Bi) prepared by a solid-state reaction technique. Low temperature phase transition was observed in an orthorhombic tungsten bronze ferroelectric  $\text{Sr}_2\text{Ca}_2\text{NaNb}_5\text{O}_{15}$  at 75 K by Zhong and co-workers [13]. In view of this, in the present paper we have synthesized and studied the preliminary structural, transport and dielectric properties of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic compound.

## EXPERIMENTAL

Polycrystalline Sample of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  were prepared by a high temperature solid state reaction technique using pure carbonates and oxides, AR grade (>99.9 % pure) chemical ( $\text{CaCO}_3$ ,  $\text{Ce}_2\text{O}_3$ ,  $\text{Bi}_2\text{O}_3$  and  $\text{TiO}_2$ ) in suitable stoichiometric ratio. The constituent compounds were mixed thoroughly using agate mortar and pestal for 1 h. The mixed material was then calcined in a platinum crucible at  $1000^\circ\text{C}$  for 6 h. Requisite amount (5%) of polyvinyl alcohol was added as a binder before making it to final stage of pellets. The process of grinding and calcinations was repeated till a homogeneous fine powder of the desired compound was obtained. The calcined fine powder was pressed in to a cylindrical pellet using a hydraulic press at pressure of  $3\text{ tons/cm}^2$  to form pellets of diameter  $1.38\text{ cm}^2$  and thickness  $0.22\text{ cm}$ . The pellet were sintered in air atmospheric at  $1000^\circ\text{C}$  for 10 h. The formation of single phase compound was checked by XRD.

The X-ray diffraction of the compound were recorded at room temperature using X-ray diffractometer ( Rigaku , miniflex ) with  $\text{Cu K } \alpha$  radiation ( $\lambda = 1.54\text{ \AA}$ ) in wide range of Bragg angle  $2\theta$  ( $20^\circ < 2\theta < 90^\circ$ ) at a scanning rate of  $0.02^\circ / \text{sec}$ . The XRD pattern was indexed using a program called TEROR from the CRYSFIRE. To measure the electrical properties of compounds, air drying silver paint was applied on both the large faces of the samples to serve as electrodes. Electrical and dielectric studies as Dielectric constant ( $\epsilon$ ), tangent loss ( $\delta$ ) and

conductivity ( $\sigma$ ) were measured as a function of frequency (42 Hz - 5 MHz) at different temperatures (27-450 °C.) using a HIOKI- LCR Bridge (model no. 3532 -50) Hi-Tester, Japan.

## RESULT AND DISCUSSION

### X-Ray study

The XRD pattern of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic obtained using  $\text{Cu K}\alpha$  radiation as shown in fig (1). A good agreement between the observed and calculated interspacing ( $d_{OBS}$  and  $d_{CAL}$ ) is seen. The sharp and single diffraction peak of each compound which are quite different from those of ingredient oxide and carbonate confirm the formation of single phase compound. All the peaks of XRD pattern were indexed and cell parameter were determine with a standard computer programmed crysfire using these experimental (d) value in different crystal structure and unit cell configuration. Using the value of 12 strong peaks of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic as shown in Table (1).

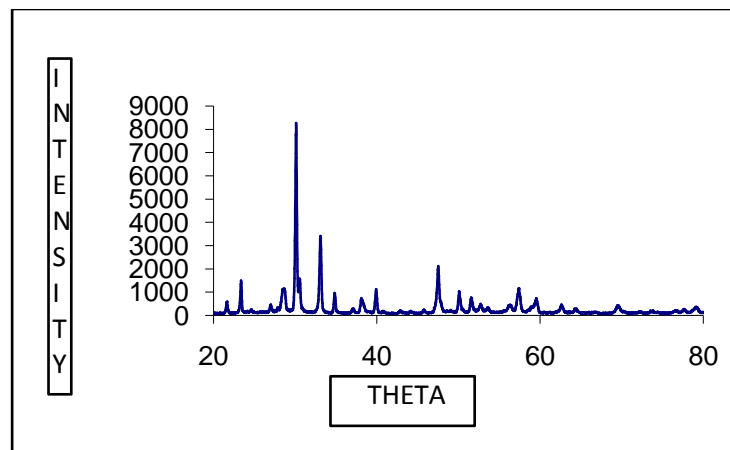


Fig (1) X- ray powder diffraction pattern of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  at room temperature

Table (1):- Indexed powder diffraction data of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic

hkl	d (observed)	d (calculated)	Intensity <sub>100</sub>
201	4.0959	4.0958	5
211	3.7986	3.7985	17
400	3.1080	3.1079	12
410	2.9646	2.9646	100
401	2.7026	2.7026	41
112	2.5750	2.5750	4
240	2.3482	2.3481	14
501	2.2576	2.2576	25
620	1.9164	1.9164	10
251	1.8193	1.8192	7
113	1.7699	1.7698	12
640	1.6040	1.6040	12
413	1.5500	1.5490	7
043	1.4785	1.4780	4

Figure (2) shows SEM micrograph of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$ , The compound show the uniform distribution of grains on the entire surface in the SEM micrograph. The shape of the grains are less spherical. The grain size evaluated from the micrographs by linear intersection methods and the measured grain size was found.

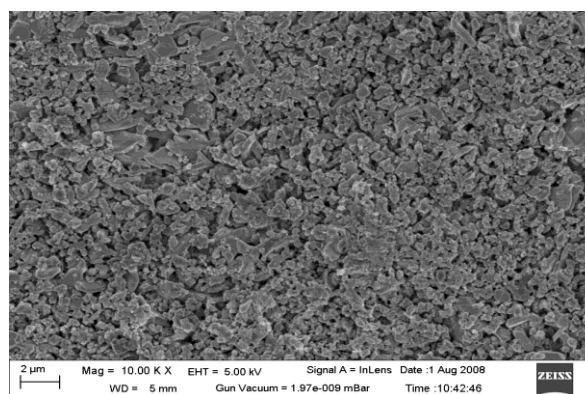


Fig (2) SEM micrograph of the  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  at room temperature

### Dielectric Studies

Figure 3 (a) and (b) show the variation of dielectric constant ( $\epsilon$ ) and tangent loss ( $\tan \delta$ ) of the sample as a function of frequency at room temperature. A decrease in dielectric constant ( $\epsilon$ ) and tangent loss ( $\tan \delta$ ) with frequency is observed which is a typical characteristic of normal dielectrics. The higher value of dielectric constant ( $\epsilon$ ) at lower frequency is due to the presence of various types of polarizations. (i.e. electronic, dipolar, interfacial, orientational etc.) in compound at room temperature.

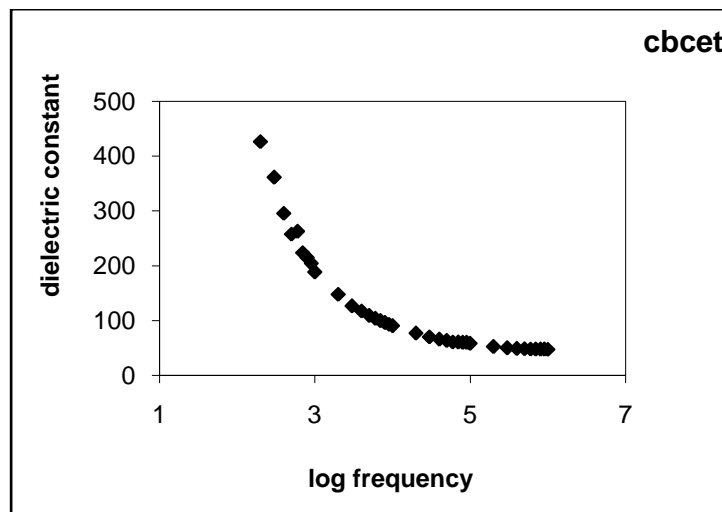


Fig. 3(a): Variation of dielectric constant ( $\epsilon$ ) of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic as a function of Frequency at room temperature

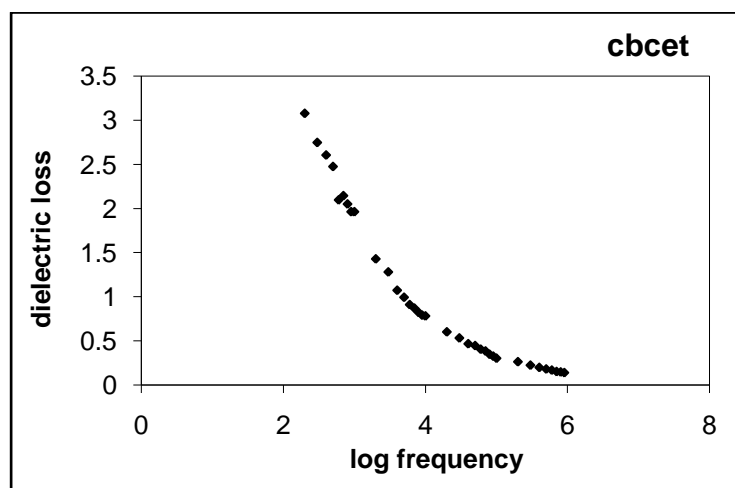


Fig. 3(b) Variation of tangent loss ( $\tan \delta$ ) of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic as a function of frequency at room temperature

As the frequency is increased, some of the polarization becomes ineffective, the electronic polarization dominates and thus the value of  $\epsilon$  decreases. The loss tangent ( $\tan \delta$ ) also decreases

with increasing frequency. This trend could be due to parallel conduction ( $\tan\delta = 1/\omega CR$ , where  $\omega$  is angular frequency,  $c$  is the capacitance and  $R$  is the resistance) probably due to connected porosity. It is observed that at higher frequency these parameters become almost frequency independent.

Figure 4 (a) and (b) shows The variation of dielectric constant ( $\epsilon$ ) and dielectric loss ( $\tan\delta$ ) with temperature at different frequency for  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic.

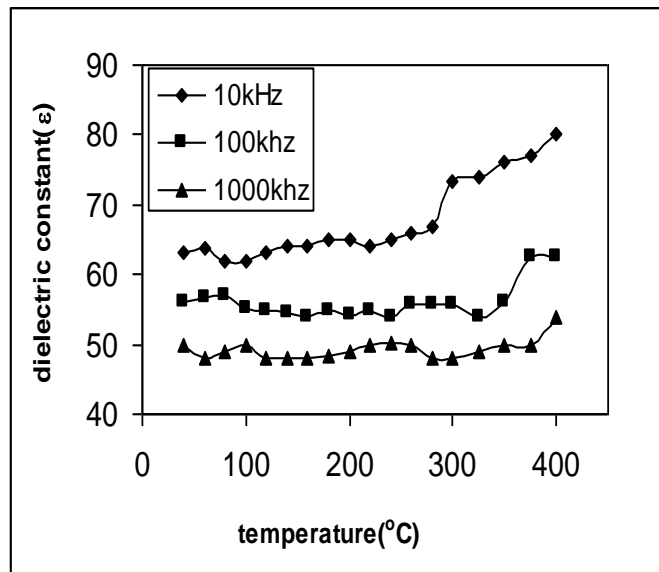


Fig. 4(a) Variation of dielectric constant ( $\epsilon$ ) of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic as a function of temperature at different frequency

No dielectric anomaly was found and temperature dependence plots of the dielectric constant were rather flat within the investigated temperature range measured at, 10 kHz, 100 kHz and 1000kHz frequency.

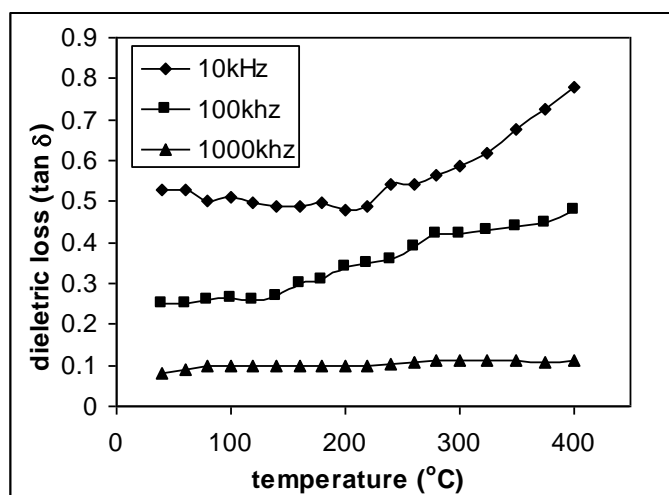


Fig. 4(b) Variation of dielectric loss ( $\tan\delta$ ) of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  Ceramic as a function of temperature at different frequencies.

As in ferroelectric, the dielectric constant ( $\epsilon$ ) increases with rise in temperature up to its maximum value at Curie temperature and then decreases with further increases in temperature. It is observed that the compounds undergo a phase transition from ferroelectric to paraelectric at particular temperature called Curie temperature. It is observed that the compounds have the maximum value of the dielectric constant at particular temperature for all the three frequencies (10, 100, 1000 kHz). Thus we can conclude that they do not exhibit relaxor behavior. Figure 4(b) shows the temperature dependence of dielectric loss of present ceramic at different frequency. As obvious from figure that dielectric loss also shows similar nature as dielectric constant.

### Conductivity analysis

Figure 5(a) shows the variation of ac electrical conductivity with inverse of absolute temperature ( $1/T$ ). ac conductivity also increases at different measured frequencies 10 kHz, 100 kHz and 1000 kHz. This infers that variation of ac conductivity as a function of temperature and frequency are analogous to each other in all respects.

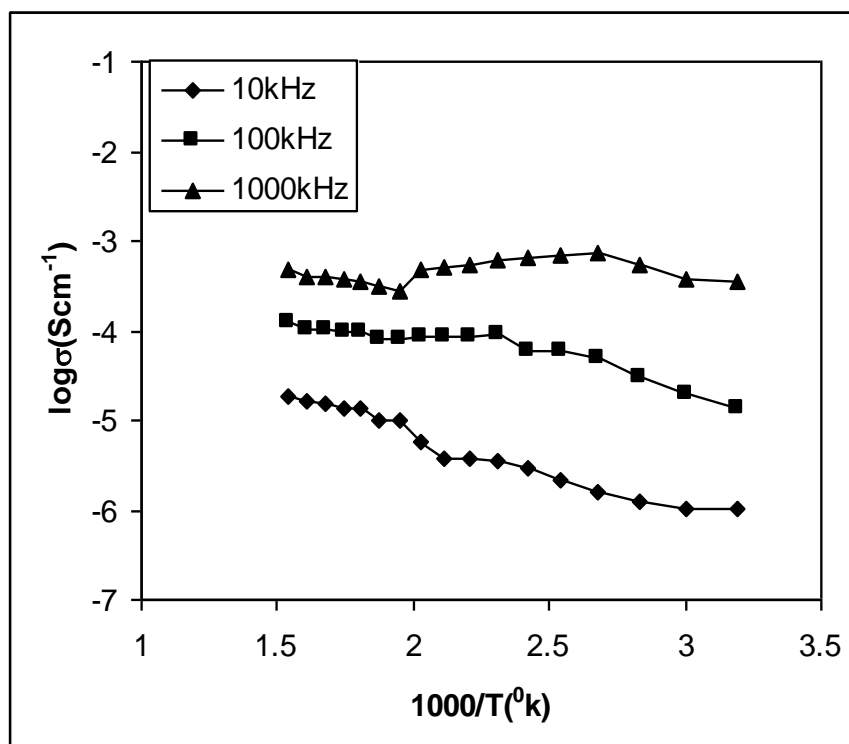


Fig.5 (a): Variation of ac conductivity ( $\sigma_{ac} = t/AZ'$ ) of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  Ceramic with  $10^3/T$  for 10, 100 & 1000 kHz

The low value of conductivity makes these ceramic suitable for capacitor application (to illustrate a few, in output section of amplifier circuit where it eliminates the need of parallel resistance, in multi element arrays where it eliminates leakage between adjacent elements etc.).

## CONCLUSION

From the preliminary structural investigation of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  ceramic, it is found that this compound is single phase with tetragonal structure. SEM micrograph of  $\text{Ca}_2\text{Bi}_3\text{CeTi}_5\text{O}_{18}$  show the uniform distribution of grains on the entire surface and shape of grains are less spherical. Dielectric studies showed that dielectric constant ( $\epsilon$ ) and tangent loss ( $\tan \delta$ ) decreases with increases frequency. The value of conductivity found to decreases with increases in value of frequency as well as temperature.

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