

## Analytic Study of phase shift crystal structure and temperature dependent dielectric properties of modified synthesis one system ceramics using BNT-BZT

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**ABSTRACT :-**In this paper, the study of phase shift crystal structure and temperature dependent dielectric properties of modified one system ceramics using BNT-BZT. LF (Lead-free) BNT-BZT ceramics were successfully prepared using the CCP (chemical coprecipitation) method. A crest shift in XRD (X-ray diffraction) patterns, with the loss of numerous hkl reflections, indicated some significant crystal structure changes in these materials. Initial crystal-structure examination shows the existence of a rhombohedral to an orthorhombic structure phase transformation. The phase transition in dielectric and ferroelectric properties was also found and correlate.

**KEYWORDS:** ceramics; XRD; dielectric properties; ferroelectric properties; phase shift.

### INTRODUCTION

In last 10 years, the actuator and sensor manufacture by mostly used LF PBT – PBZ (PbTiO<sub>3</sub>-PbZrO<sub>3</sub>) material because of their excellent ferroelectric and piezoelectric properties. Now the Zr/Ti show the morphotropic phase boundary between tetragonal and a rhombohedral phase using the ratio of 52/48, where improved polarizability and finest domain orientation were observed. On the other hand, lead-based materials are increasing environmental toxic waste with extra problems of recycling and waste disposal and vital drawback of PZT is that it contains more than 65% lead (Pb) by weight. Now required to build up new LF smart materials in order to substitute the lead-based ones. BT (Bismuth Titanate) is an example of the most commonly used LF material for capacitors and actuators due to its innate ferroelectric nature. However, its main drawback is the narrow working temperature; therefore, the use of a BT-BZ solid solution with the addition of Zr up to 30% mole was investigated. The Induced phase transition find out the exhibit composition of material from normal to relaxor ferroelectric with higher dielectric constants with PZT and BT. Now the using over a border temperature range of this permitted material. Next, to these studies, this paper was aimed to study BNT-BZT CCP method with the addition of a Zr different concentration mole fractions. The correlation between the phase, crystal structure, and electrical properties is investigated and discussed.

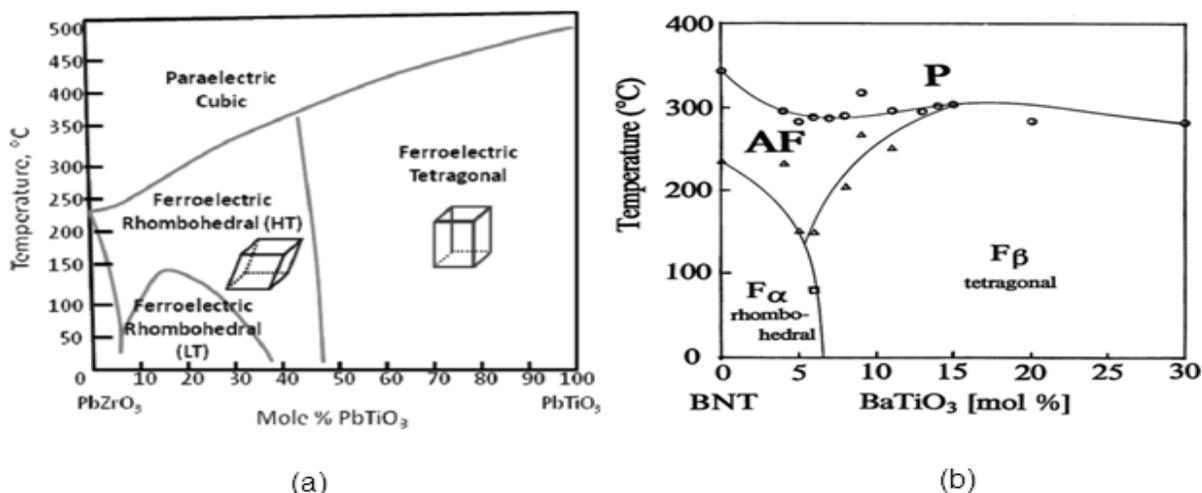
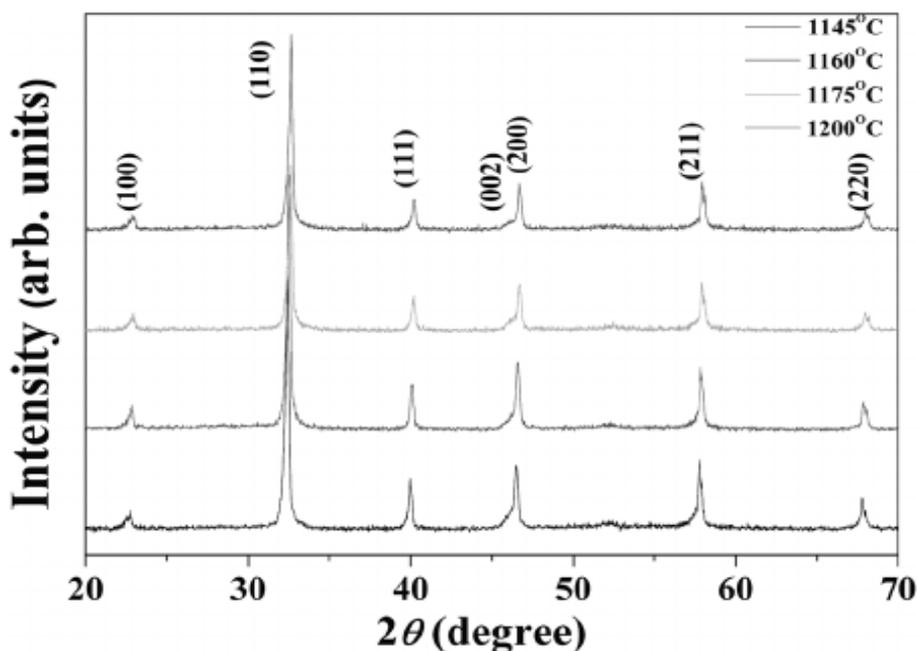


Figure 1 Morphotropic phase boundary in (a) PZT and (b) BNT

### EXPERIMENTAL METHODS

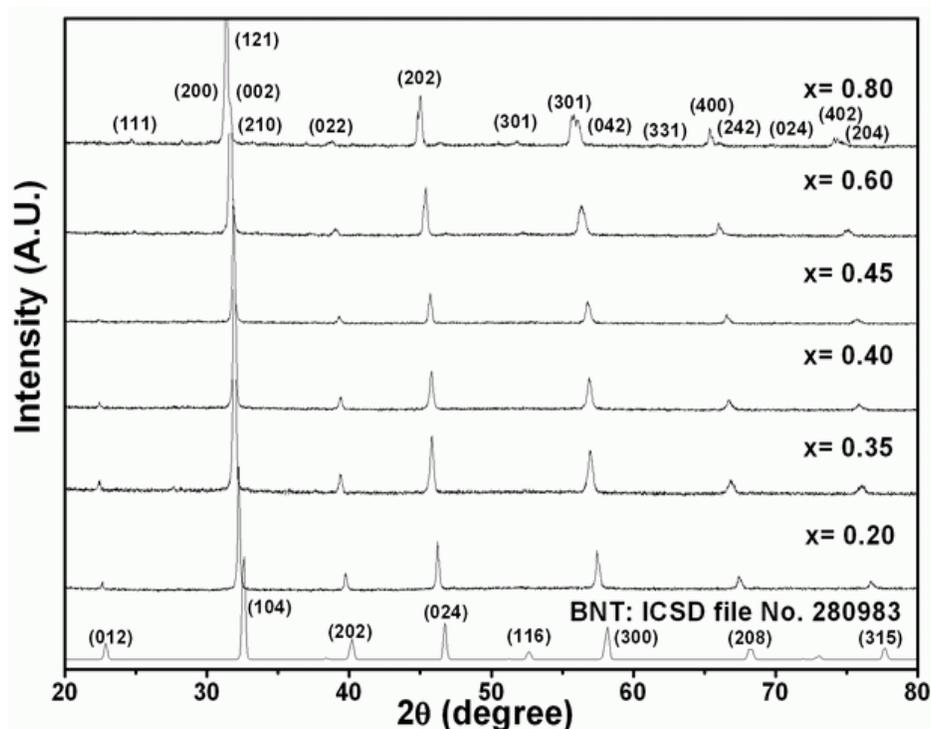
BNT-BZT compositions were prepared using the CCP method. BNT – BZT were synthesized via CCP technique by taking BiCl<sub>3</sub>, NaCl, TiCl<sub>3</sub>, BaCl<sub>2</sub>, and ZrCl<sub>3</sub> as starting chemicals. Metal salts were taken in stoichiometric ratio 2:5 and their

homogeneous solutions were prepared in distilled water using magnetic stirring [1]. Oleate was added to avoid a cluster of particles and to defend the particles from atmospheric oxygen. Ammonia solution was added dropwise under constant stirring for the precipitation of simple perovskites [2]. Pressed under the pressure of 5.543 MPa with some drops 3.0 Wt % PVA (polyvinyl alcohol) using the binding of perovskite ceramics of BNT-BZT powder on the disk of 10 mm Dia and 1 – 15 mm thickness [3]. The disks were the slag at 890°C for 2.30 hrs, except for the sample with 0.20-mole fractions Zr which was slag at 940°C for 2.30 hrs, in the air. The initial crystal structure details were calculated using the PC (Powder Cell) program, which is based on the XRD pattern of LF BNT-BZT ceramics. The bulk volume and area of the slag ceramics were computing using Archimedes' method. The theoretical density (volume and area) was approximated from the unit cell size and its component ions. Scanning electron microscopy [SEM] was used to observe the microstructure of the ceramics. To ready the SEM samples, they were well-coated and thermally carve for 15 min at 750°C. The average grain size was then evaluated from these SEM images. Figure 2 shows the XRD patterns of BNT and results and discussion on the basis. In rhombohedral space group R3c, the BNT – BZT phase might be same by pure BNT. All reflection peaks systematically shifted to lower than  $2\theta$  angles for the existence of Zr. Additional the shift, intensities of some diffraction peaks such as (012) and (202) were reduced, indicating that lattice distortion alongside unit cell expansion has occurred [4]. The enhancement of the XRD patterns was conceded out.



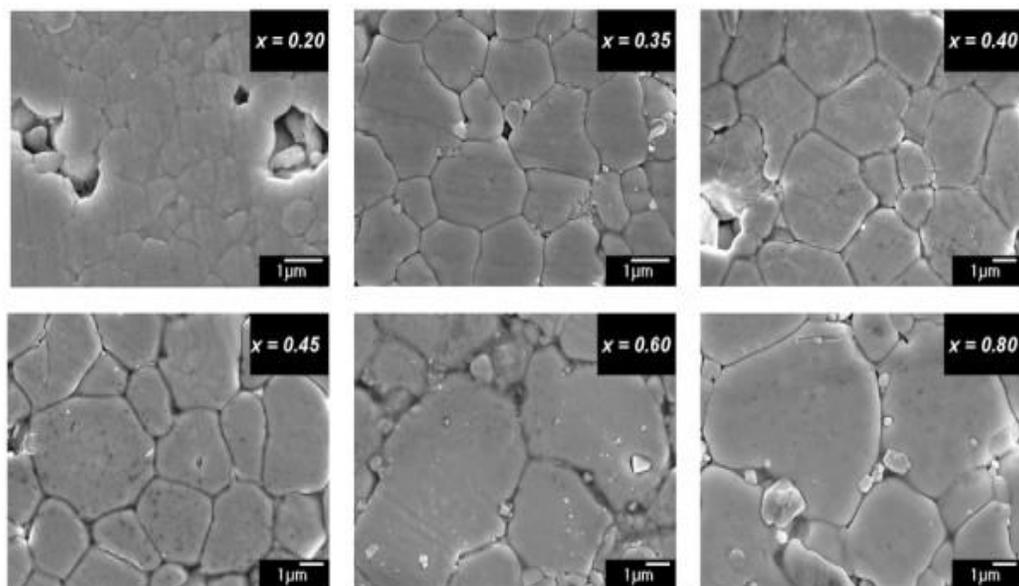
**Figure 2 X-ray diffraction patterns of BNT**

Figure 3 shows the refined pattern for the Zr composition equal to 0.2 – 0.8 in XRD patterns. BNT-BZT ceramics containing Zr from 0.2 – 0.6 overcome a rhombohedral structure with improved lattice parameters [4]. The rise in the value of interaxial angle caused the structure to be close to cubic, which resulted in the vanishing of certain reflections. For Zr = 0.8, Figure 3 showed an apparent splitting of the (104) and (300) peaks in the original rhombohedral structure [5]. This finding was quite in partial agreement with the orthorhombic structure earlier obtained for BNT-BZT. Therefore, for this BNT-BZT solid solution ceramic system, the structure changed from rhombohedral to orthorhombic when the Zr concentration exceeded 0.6-mole fraction [5]. The exact phase-transition composition is currently being investigated [4]. The entire BNT-BZT ceramics had trial density values in the range of 5.8 – 6.11g/cm<sup>3</sup>. The difference in sintering behavior could also be noted from the microstructure of BNT-BZT ceramics. The enhanced ability for ionic diffusion in BNT-BZT ceramics seemed to support the possible lowering of the melting point of these solid solutions. In general, increasing Zr concentration in BNT-BZT ceramics caused a gradual drop off in dielectric constant with a slight drop off in dielectric loss. This behavior was in agreement with other systems with isovalent additives [4]. In addition, the replacement of larger Zr ions may also cause the dipoles to be poorly induced due to limited ionic movement. This decreasing trend was observed for the sample with a composition of Zr = 0.8, whose structure was orthorhombic. It seemed that the effect of ionic size and limited ionic movement in the perovskite structure of this compound had a greater influence on the dielectric properties than the change in the crystal structure in their unit-all dimensions.[4][5].



**Figure 3 X-ray diffraction patterns of Bi<sub>0.5</sub>Na<sub>0.5</sub>Ti<sub>1-x</sub>Zr<sub>x</sub>O<sub>3</sub> ceramics. Where x = 0.20, 0.35, 0.40, 0.45, 0.60, and 0.80 mole fraction [4].**

The hysteresis loops were finding at the maximum applied electric field of  $20 \times 10^3$  V/cm and a frequency of 50 Hz and some degree of domain reorientation might also be the cause of unfortunate hysteresis loops for these compositions [6]. For samples with Zr = 0.6 and 0.8, the loops showed elevated values of remnant polarization however they were at rest unsaturated [5]. This seemed to show the approximate transition point between the rhombohedral and orthorhombic structures [4]. This was supported by an increase in the breakdown field strength for the Zr = 0.8 composition, which was partly due to the effect of a different crystal structure in this series of materials. Hence, this study showed that the observed dielectric and ferroelectric properties of BNT-BZT ceramics largely depended on compositional and crystal structure changes. Optimization of these properties could be achieved by fine-tuning the composition for specific applications.



**Figure 4 SEM image of BNT-BZT ceramics [4]**

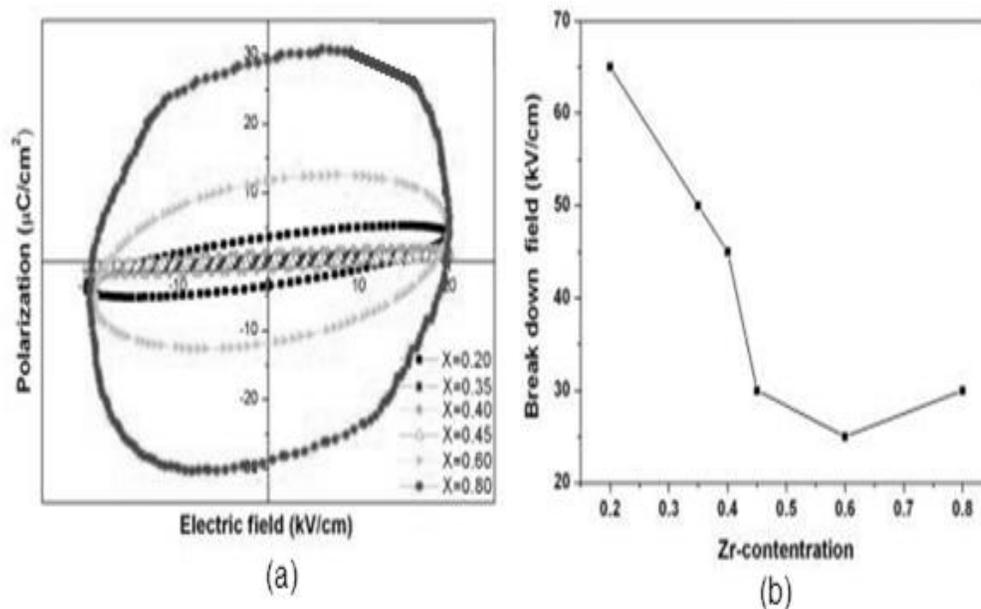


Figure 5 P-E hysteresis loops (a) and the breakdown field (b) of  $\text{Bi}_{0.5}\text{Na}_{0.5}\text{Ti}_{1-x}\text{Zr}_x\text{O}_3$  ceramics. Where  $x = 0.20, 0.35, 0.40, 0.45, 0.60,$  and  $0.80$  mole fraction.

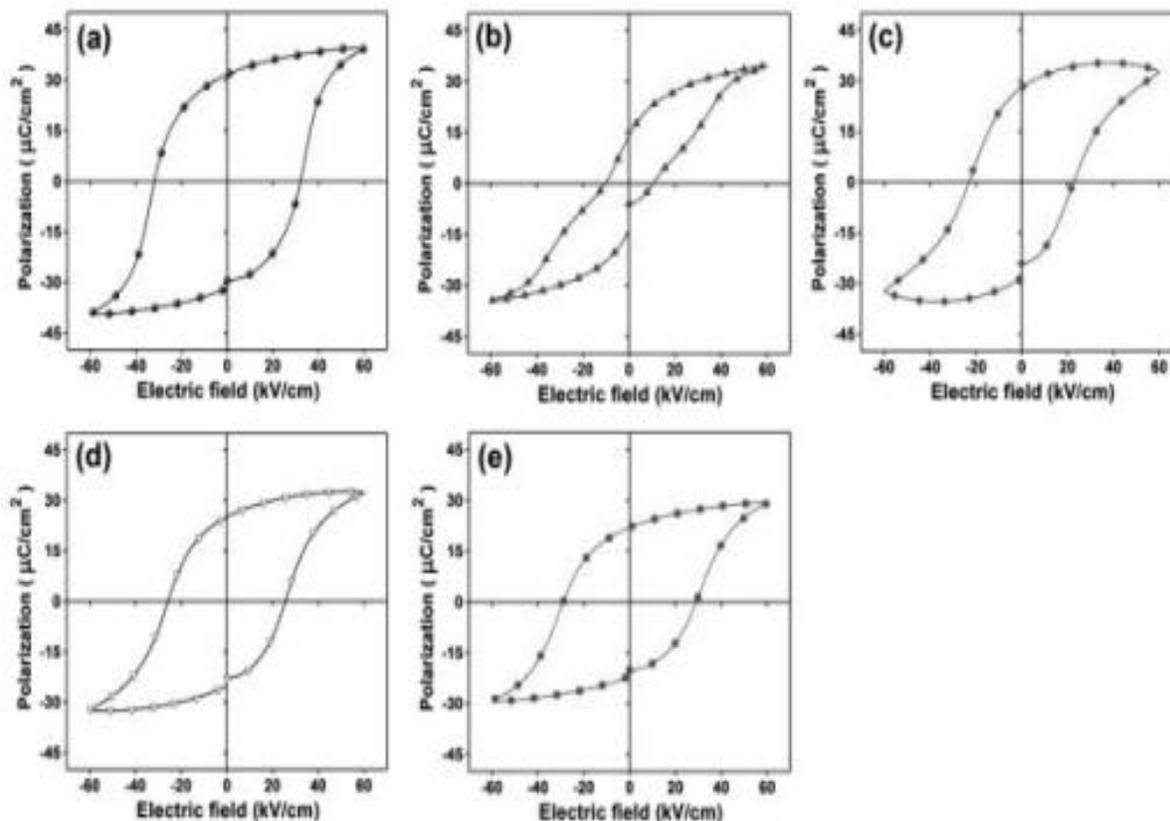
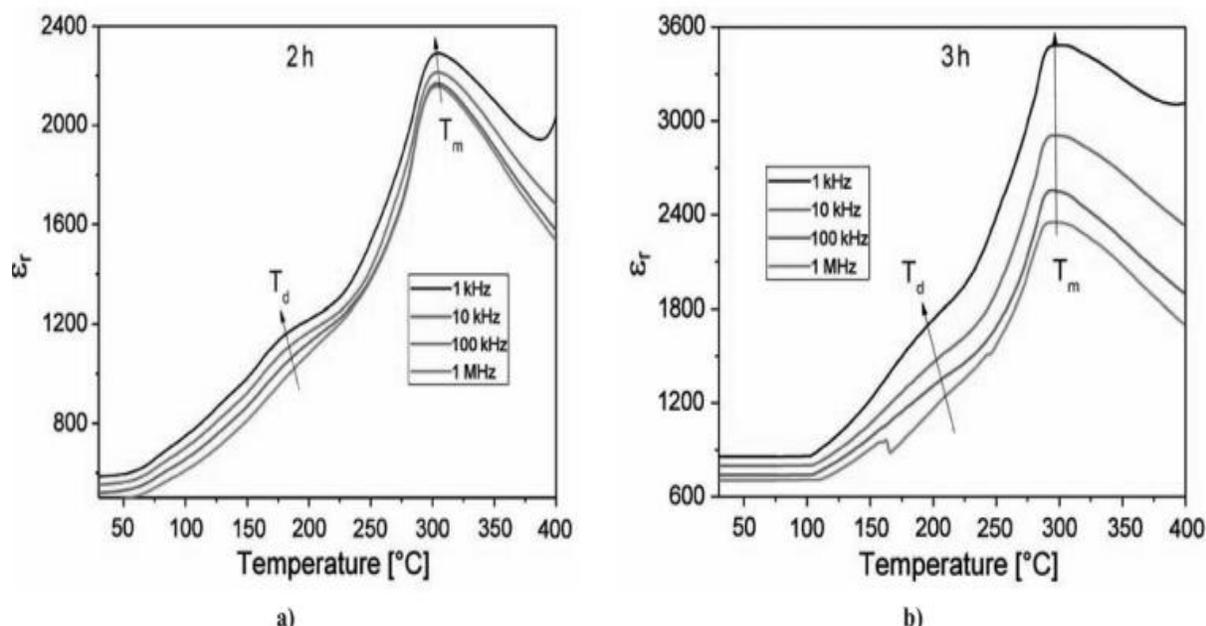


Figure 6 Plots of polarization as an electric field function of  $(1-x)\text{BNT}-x\text{BZT}$  ceramics. The samples were sintered at  $1,125^\circ\text{C}$ . (a)  $x = 0$ , (b)  $x = 0.05$ , (c)  $x = 0.10$ , (d)  $x = 0.15$ , and (e)  $x = 0.20$ .



**Figure 7** Temperature and frequency dependent dielectric study of BNT ceramics sintered at 1100 °C for: a) 2 and b) 3 hours

## RESULT DISCUSSION

The ceramic made by any one method, results are almost similar find. Last few decades there have been many discoveries and progress in the field of LF piezoelectric materials. In this work, LF BNT-BZT ceramics were successfully fabricated. X-ray diffraction patterns showed phase transition from rhombohedral to an orthorhombic structure [5]. The addition of Zr concentration caused lattice expansion in agreement with ionic size consideration [4]. All ceramic samples were dense with definite grain structures. The dielectric constant was found to drop off with increasing Zr content due to the larger sized ionic substitution that limited dipole movement. Ferroelectric properties also showed compositional dependence due to the variation in domain reorientation ability. This study showed that electrical properties of BNT-BZT ceramics could be further improved by fine-tuning their composition for certain applications. The LF BNT-BZT ceramic pellets were sintered at 1100 °C for two different times. The SEM of the sintered pellet showed well developed rectangular grains and the increased density of the pellet with the increase in the sintering time. The temperature dependent dielectric study showed that two types of phase transitions occurred in both samples. The dielectric analysis also showed that the transition temperature increases with frequency, thus confirming a diffuse phase transition behavior.

## CONCLUSION

The relative reactivity of the formation of one system ceramic by the CPP method shows the conspicuous difference in their SEM images. The images suggest that diffusing different species in BNT – BZT is one of the ways to transfer/displace Zr from Zirconium Titanate which is obvious from surface layers of the material into the titanate particles. So the porous material may find useful for devices dealing with thermal conductivity and temperature dependent dielectric properties.

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