

## International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Website: [www.ijirset.com](http://www.ijirset.com)

Vol. 6, Issue 2, February 2017

# A Ferroelectric Study on the Effect of Lead Barium Niobate (PBN) Ceramics

Vemuri Raghavendra<sup>1\*</sup>, B.Suryanarayana<sup>2</sup>, K.Chandra mouli<sup>3</sup>, P.V.Achary<sup>4</sup>

Research Scholar, Dept. of Engineering Physics, Andhra University, Visakhapatnam, A.P, India<sup>1</sup>

Teaching Assistant, Dept. of Engineering Physics, Andhra University, Visakhapatnam, A.P, India<sup>2</sup>

Professor, Dept. of Engineering Physics, Andhra University, Visakhapatnam, A.P, India<sup>3</sup>

Senior Assistant Professor, ANITS Engineering College, Sangivalasa, Visakhapatnam, A.P, India<sup>4</sup>

**ABSTRACT:** Lead barium niobate ceramics (PBN) were synthesized by high temperature solid state reaction technique with the stoichiometric formula  $Pb_{1-x}Ba_xNb_2O_6$  where  $x = 0.35$  kept constant. X-ray diffraction analysis reports single phase tetragonal structure. The lattice parameters obtained are  $a = 12.505 \text{ \AA}$  and  $c = 3.984 \text{ \AA}$ . The Transition temperature ( $T_C$ ) of PBN-1 is  $350^\circ\text{C}$ . The variation of dielectric constant with temperature as shown in fig.3 and it reports that the dielectric constant value linearly increases with increase of temperature upto a certain temperature and then gradually decreased. The transition temperature ( $T_C$ ) shows to be one of the primary qualities of ferroelectricity and antiferroelectricity. The sintering temperature increases the density of the material also increases to a maximum value. The density of the material is found to be more than 95% of theoretical density. The  $d_{33}$  value is 95pC/N and mechanical quality factor indicates low loss.

**KEYWORDS:** Solid state reaction, XRD, Dielectric, Piezoelectric, Lattice parameters.

## I. INTRODUCTION

Lead metaniobate ( $PbNb_2O_6$ , PN) with tungsten-bronze structure is one of the piezoelectric ceramics with high Curie temperature, which is highly desirable to construct transducers that can be operated at high temperatures.[1]. Goldman discovered ferroelectricity in tungsten bronze type niobates is  $PbNb_2O_6$ . It is having large pyroelectric, piezoelectric and electrooptic properties. Joseph valsek [2, 3] observed the phenomenon of ferroelectricity in Rochelle salt from the measurement of dielectric properties. Another discovery of similar phenomenon in a series of phosphates and arsenates among which is the best example is  $KH_2PO_4$  known as KDP [4]. The work in theoretical experimental studies increased with the inclusion of barium titanate [5] in the list of ferroelectric materials. The ceramics of the lead barium niobate  $Pb_xBa_{1-x}Nb_2O_6$  series had been studied in early years. The ferroelectric tungsten bronze can be classified by the general formula  $AB_2O_6$  where A is a divalent material and B is neither niobium or tantalum. By using Czochralski method Li and Na doped PBN single crystals studied by Xu et al. [6]. Low  $Q_m$  piezoelectric ceramics are suitable for wide band ultrasonic transducer applications. The piezoelectric effect of ferroelectric ceramics has vast potential as electromechanical transducers. The piezoelectric transducers are limited to device involving only very small mechanical displacements and small amounts of electrical charge per cycle. In this paper we briefly studied the structure, dielectric and piezoelectric properties of ceramics by solid state reaction method.

## II. EXPERIMENTAL PROCEDURE

High purity (99.9 %)  $PbO$ ,  $BaCO_3$  and  $Nb_2O_5$  were used for the preparation of the composition by solid state reaction method[7]. The general formula of the composition is  $Pb_{1-x}Ba_xNb_2O_6$  where  $x = 0.35$  then it can be written as  $Pb_{0.65}Ba_{0.35}Nb_2O_6$  (PBN-1). Stoichiometric quantities of the reagents were first dry mixed in an agate mortar. The

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Website: [www.ijirset.com](http://www.ijirset.com)

Vol. 6, Issue 2, February 2017

composition was further wet-mixed for 2 h with suitable organic solvent media (Methanol). The well-grounded pale yellowish powder was taken in alumina crucibles covered with lid and then calcined at 900<sup>o</sup>C for 3h for materials. This calcination process was repeated twice for each of the composition to increase homogeneity. An excess of Pbo of 5-6 wt% was added to composition to compensate the loss during high temperature treatment. The conventional ceramic method of dry pressing in a die punch assembly using a hydraulic press was employed to compress the powder into disc shape. To ensure good flow of the powder and a reproducible filling of the die, the powder was sieved and granulated using 5% polyvinyl alcohol (PVA).The granule were then pressed into discs at a pressure of 3-4 tonnes per sq.inch. The pressed discs were then placed on a platinum foil and closed with a crucible and the sintered in a silicon carbide heated furnace in air for a sintering at 1250<sup>o</sup>C for 2 hours for the preparation of different ceramic composition. The phase formation was confirmed by X-ray diffraction (XRD) studies of calcined materials. The X-ray diffractograms of the sample were taken using Cu K $\alpha$  radiation. For the dielectric characterization, the sintered pellets were polished and then coated with a conducting silver paste for making electrical contacts. Then the capacitance measurements were taken at 1khz from room temperature to 450<sup>o</sup>C. Piezoelectric coupling coefficients of ferroelectric ceramics are measured by the resonance and anti resonance technique [8].This was accomplished by using Hewlett Packard LF impedance analyser model 4192A.Probe fixture model 16095A was used as analyzer through BNC adapter.

## III. RESULTS AND DISCUSSION

### A. X-RAY DIFFRACTION

Powder x- ray diffraction technique is most commonly and frequently used method-ray diffraction pattern comprising both position and intensity are helpful in identification of the phase, lattice parameters, crystal symmetry and structural analysis. Powder x-ray diffraction was carried out by Philips X-ray diffractometer PW-1710 using CuK $\alpha$  radiation with Ni filter at room temperature and 2 $\theta$  ranging from 10<sup>o</sup> to 70<sup>o</sup> and 2<sup>o</sup>/ min step scan was used for determination of phase genesis, crystal structure and the average of particles.

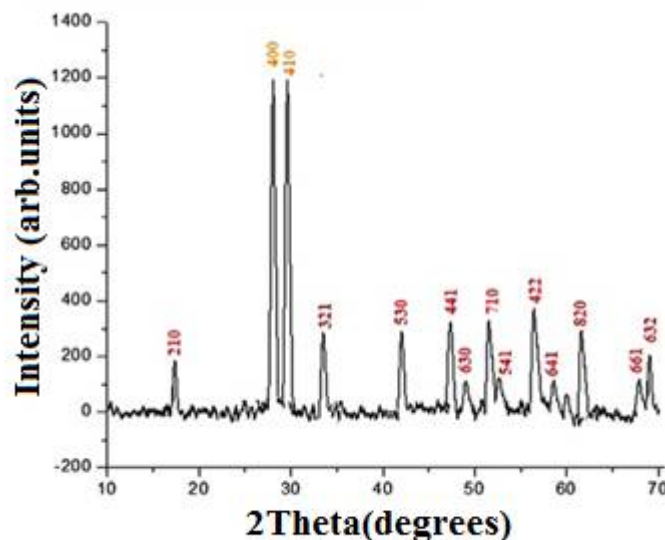


Fig.1: X-Ray diffraction patterns of Pb<sub>0.65</sub>Ba<sub>0.35</sub>Nb<sub>2</sub>O<sub>6</sub>

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Website: [www.ijirset.com](http://www.ijirset.com)

Vol. 6, Issue 2, February 2017

From the Fig.1 it is evident that confirm single phase tetragonal structure. The lattice constants for PBN-1 is  $a=12.505 \text{ \AA}$  and  $c=3.984 \text{ \AA}$  are well matched with reported values [9,10].The values of lattice parameter,  $V \text{ \AA}^3$ , density, density percentage and axial ratio are tabulated in table 1. The density of the ceramic was measured by Archimedes method. The axial ratio is very small indicates that the distortion in crystal lattice is also very small. The axial ratio relates to the nature of distortion of the  $\text{NbO}_6$  octahedron in TB structure.From fig.2 it is observed the variation of sintering temperature with density was reported.

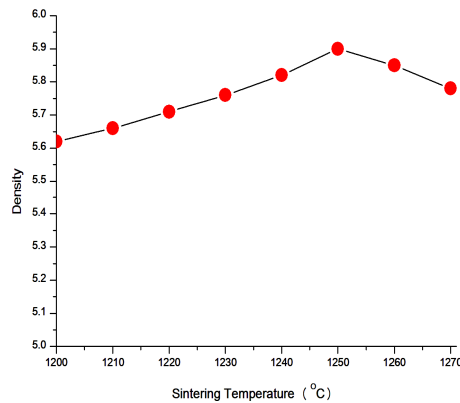


Fig.2: Variation of sintering temperature with density

Table 1: XRD data  $\text{Pb}_{0.65}\text{Ba}_{0.35}\text{Nb}_2\text{O}_6$

Composition	Lattice Parameter		$V \text{ \AA}^3$	$\rho_{\text{expt}} \text{ gm/cm}^3$	$\rho_{\text{cal}} \text{ gm/cm}^3$	$\sqrt{10} \frac{c}{a}$	Density Percentage
	$a \text{ \AA}$	$c \text{ \AA}$					
$\text{Pb}_{0.65}\text{Ba}_{0.35}\text{Nb}_2\text{O}_6$ (PBN-1)	12.505	3.984	629.279	5.90	6.15	1.007	96

## B. DIELECTRIC PROPERTIES

Dielectric materials are superior electrical insulators. They do not have free carriers due to the strong bounds between their valence electrons and atoms. Under control of an external electric field, dielectrics respond by redistributing the charge balance inside the material. The studies on the prepared materials have been performed Hewlett Packard LF impedance analyser model 4192A through an intermediary accessory model 16095A. Then the capacitance measurements were taken at 1 KHz frequency from room temperature to  $450^\circ\text{C}$ . The Delta design model 2300 is a precision environmental test chamber in which the temperature with accuracy of  $\pm 0.1^\circ\text{C}$  can be obtained.

In this model the temperature is controlled by means of Delta design's solid state controller which can be operated either manually or through an external programmer. The data has been analyzed by using the statistical program software package, "Origin 7.5", Northampton, USA.

Ferroelectricity in ceramics can easily implicit due to dielectric property of ferroelectric compositions over an extensive varying of temperature. The Transition temperature ( $T_c$ ) of PBN-1 is  $350^\circ\text{C}$  is well matched with literature values [11].

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Website: [www.ijirset.com](http://www.ijirset.com)

Vol. 6, Issue 2, February 2017

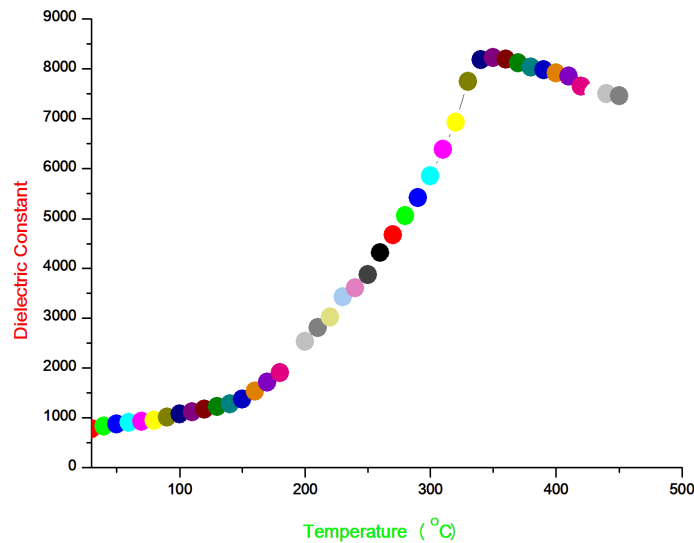


Fig.3: Variation of dielectric constant with temperature

The variation of dielectric constant with temperature as shown in fig.3 and it reports that the dielectric constant value linearly increases with increase of temperature upto a certain temperature and then gradually decreased. The transition temperature ( $T_C$ ) shows to be one of the primary qualities of ferroelectricity and antiferroelectricity. This measurement gives the beginning of the spontaneous polarized and it considered being one of the significant factors for piezoelectric material characterisation. The values of density,  $\epsilon_{RT}$ ,  $\epsilon_{Tc}$ ,  $T_C$  and  $\text{Tan}\delta$  of  $\text{Pb}_{0.65}\text{Ba}_{0.35}\text{Nb}_2\text{O}_6$  are presented in table 2.

Table 2: Dielectric data of  $\text{Pb}_{0.65}\text{Ba}_{0.35}\text{Nb}_2\text{O}_6$

Composition	Density gm/cm <sup>3</sup>	$\epsilon_{RT}$	$\epsilon_{Tc}$	$T_C$ (°C)	$\text{Tan}\delta$
$\text{Pb}_{0.65}\text{Ba}_{0.35}\text{Nb}_2\text{O}_6$	5.90	774	8226	350	0.05

### C. PIEZOELECTRIC PROPERTIES

Piezoelectricity is the capability of some materials or ceramics, to prepare an electrical potential when set under a mechanical stress analogous to the pyroelectric effect, when a material generates a electric potential because of the opposition with a temperature change. Lead barium niobate (PBN) are prepared by solid state reaction technique. The enhancement in the piezoelectric properties due to additions of rare earth ions and barium may be attributed to high mobility of ions on the site owing to the presence of the ions in the site. The present studies are primarily aimed to explore the possibility of enhance piezoelectric properties in PBN. The values of Planar coupling coefficient ( $k_p$ ), Thickness coupling coefficient ( $k_t$ ), Piezoelectric charge coefficients ( $d_{31}, d_{33}$ ), Piezoelectric voltage coefficients ( $g_{31}$ ) and mechanical quality factor ( $Q_m$ ) of  $\text{Sm}^{3+}$  doped PBN and unmodified PBN were presented in table 3.

## International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Website: [www.ijirset.com](http://www.ijirset.com)

Vol. 6, Issue 2, February 2017

**Table 3:** Piezoelectric coefficients data on  $Pb_{0.65}Ba_{0.35}Nb_2O_6$

Composition	Density $gm/cm^3$	$k_p$	$k_t$	$d_{31}$ pC/N	$d_{33}$ pC/N	$g_{31} \times 10^{-3}$ Vm/N	$Q_m$
$Pb_{0.65}Ba_{0.35}Nb_2O_6$	5.90	0.39	0.41	46	95	4.45	85

From the table it is evident that  $d_{33}$  value is 95pC/N and mechanical quality factor indicates low loss. The same trend have been reported by R.R.Neurgaonkar et.al [12] for hot pressed PBN:60 and PBLN:60. Hence in our study of enhance piezoelectric properties are found. The  $k_p$  and  $k_t$  values indicates that materials are useful for actuator applications.

### IV.CONCLUSIONS

Lead barium niobate ceramics (PBN) were synthesized by high temperature solid state reaction technique with the stoichiometric formula  $Pb_{1-x}Ba_xNb_2O_6$  where  $x = 0.35$  kept constant. X-ray diffraction analysis reports single phase tetragonal structure. The lattice parameters obtained are  $a = 12.505 \text{ \AA}$  and  $c = 3.984 \text{ \AA}$ . The Transition temperature ( $T_C$ ) of PBN-1 is  $350^\circ\text{C}$ . The variation of dielectric constant with temperature as shown in fig.3 and it reports that the dielectric constant value linearly increases with increase of temperature upto a certain temperature and then gradually decreased. The transition temperature ( $T_C$ ) shows to be one of the primary qualities of ferroelectricity and antiferroelectricity. The sintering temperature increases the density of the material also increases to a maximum value. The density of the material is found to be more than 95% of theoretical density. The  $d_{33}$  value is 95pC/N and mechanical quality factor indicates low loss

### REFERENCES

- [1] Jing-jing Yuan, Xiao-ming Chen, Han-li Lian, Peng Liu "Effects of the doping of  $W^{6+}$  ions on the structure and electrical properties of  $Pb_{0.95}Ba_{0.05}Nb_2O_6$  piezoelectric ceramics" *Ceramics International* 41 (2015) S662-S667.
- [2] J. Valasek, "Piezo-Electric and Allied Phenomena in Rochelle Salt" *Phys.Rev.* 15 (1920) 537.
- [3] J. Valasek, "Piezo-Electric and Allied Phenomena in Rochelle Salt" *Phys.Rev.* 17 (1921) 475.
- [4] S. D. ROSS "The vibrational spectra of lithium niobate, barium sodium niobate and barium sodium tantalate" *J. Phys. C: Solid St. Phys.*, 1970, Vol. 3.
- [5] B. Wul and I.M. Goldman, *CR. Acad.Sci* 6 (1949) 46.
- [6] Y. Xu, C. Tinachen, J.D. Mackenzie, "Dielectric and piezoelectric properties of  $Pb_{1-x}Ba_xNb_2O_6$  ferroelectric tungsten bronze crystals" *Ferroelectrics*, 74 (1987) 317.
- [7] Vemuri Raghavendra, Y. Ramesh Babu, B. Suryanarayana, K. Ramanjaneyulu and K. Chandra Mouli, "SYNTHESIS AND STUDIES ON TUNGSTEN BRONZE LEAD BASED CERAMIC MATERIALS", *IJMER*, Vol 4, 11(5), Nov 2015.
- [8] IRE Standards on Piezoelectric crystals *Proc. IRE*, 46 (1958) 764.
- [9] R.R. Neurgaonkar, J.R. Oliver and L.E. Cross, "Ferroelectric properties of tetragonal tungsten bronze single crystals" *Ferroelectrics* 56, 31 (1984).
- [10] K.S. Rao and N. Vallisnath "Influence of Rare-Earth Ion on Piezoelectric and Pyroelectric Properties of PBN System" *Ferroelectrics*, 325:15-24, 2005.
- [11] P. Baxter and N.J. Hellicar, "Electrical Properties of Lead-Barium Niobates and Associated Materials" *J. Am. Ceram. Soc.*, 43(11), 578 (1960).
- [12] R.R. Neurgaonkar, J.R. Oliver, J.G. Nelsom and L.E. Cross, "Piezoelectric and ferroelectric properties of La-modified and unmodified tungsten bronze  $Pb_{0.6}Ba_{0.4}Nb_2O_6$  dense ceramics" *Mat. Res. Bull.*, 26, 771 (1991).