

## Investigation of Dielectric and Piezoelectric studies on Sr doped PBN ceramics

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### ABSTRACT

The ceramic compositions of PBN are  $Pb_{1-x}Ba_xNb_2O_6$  ( $x=0.4$ ),  $Pb_{1-x-y}Ba_xSr_yNb_2O_6$  ( $x=0.4$ ;  $y=0.02, 0.04, 0.05$ ) were prepared by solid state reaction method. The general formula of TB type is  $[(A1)_2(A2)_4C_4] [(B1)_2(B2)_8] O_{30}$ . Dielectric studies of pure PBN and Sr doped PBN as a function of temperature were studied. Piezoelectric studies  $K_p$ ,  $K_t$ ,  $Q_M$  and charge coefficient are reported.

**Key words:** PBN, Dielectric studies, Piezoelectric studies, Tungsten Bronze.

### INTRODUCTION

Lead niobate ( $PbNb_2O_6$ ) is one of the first crystals of the tungsten bronze-type structure which was first reported as ferroelectric (G. Goodman, Text Book, 1953). It is well known that the tungsten bronze niobates generally indicate a relaxor ferroelectric nature. These materials crystallize in a variety of structures including tetragonal tungsten bronze (TTB), hexagonal tungsten bronze (HTB), and intergrowth tungsten bronze (ITB) (V. Hornebecq et.al, 2000). The synthesis and phase identification of the piezoelectric/ferroelectric phase of lead niobate is difficult, that a few competing phases and compounds tend to form during the preparation (M. Venet et al., 2005; H. S. Lee et al., 1998; J. Soejima et al., 2000; S. Ray et al., 2000). The general formula for tungsten bronze structure could be represented as follows:  $(A1)_2^{XII}(A2)_4^{XV} (C)_4^{IX} (B1)_2^{VI} (B2)_8^{VI} O_{30}^{VI}$ .

PBN has three distinct interstitial sites which are A1 = square, A2 = pentagon and C = triangle. The common chemical formula is  $(A1)_2(A2)_4(C)_4(B1)_2(B2)_8O_{30}$ , where positions A1, A2, B1, B2 and C will be filled by different valence cations or maybe partially empty. In a 4-mm unit cell of  $(Pb, Ba)_5Nb_{10}O_{30}$ , Pb ions prefer 12-coordinated square (A1-site) while Ba ions prefer 15-coordinated pentagon (A2-site) cations surrounded by 9-coordinated triangle (C-site) oxygen anions, and since there are only five  $Ba^{2+}$  and  $Pb^{2+}$  cations available for six A-sites in the unit cell, there is one vacancy distributed in the A-sites (C.A. Randall et al., 1993). Due to high Curie temperature and low-quality factor, the material is useful for fabrication of ultrasonic transducers for high-temperature applications where the PZT and other piezoelectric materials cannot be used (Y. M. Li et al., 2008). This paper reports the dielectric and piezoelectric properties of Sr:PBN oxide ceramics.

### RESULTS AND DISCUSSIONS

The polycrystalline samples of  $Pb_{1-x}Ba_xNb_2O_6$  ( $x=0.4$ ) &  $Pb_{1-x-y}Ba_xSr_yNb_2O_6$  ( $x=0.4$ ;  $y=0.02, 0.04, 0.05$ ) were prepared by solid state reaction method. The synthesized ceramic compositions are  $Pb_{0.6}Ba_{0.4}Nb_2O_6$ ,  $Pb_{0.58}Ba_{0.4}Sr_{0.02}Nb_2O_6$ ,  $Pb_{0.56}Ba_{0.4}Sr_{0.04}Nb_2O_6$ ,  $Pb_{0.55}Ba_{0.4}Sr_{0.05}Nb_2O_6$ .

The raw materials of oxides and carbonates  $PbO$ ,  $BaCO_3$ ,  $SrCO_3$  and  $Nb_2O_5$  are of AR grade. The physical mixture is weighed to give the compositions. They have been calcined around  $900^\circ C$  for 2 hours. Repeated calcinations and grinding facilitate lesser particle size and to give fresh surface to individual reactants which will enhance the rate of solid state reaction. The calcined powders are grounded with PVC binder and made pellets to disc shape. The pelletized sample has been sintered at the temperature of  $1300^\circ C$  for two hours. They have been electrode with air drying silver paste, cured at  $600^\circ C$  at 20 min and has been for the measurement of dielectric and piezoelectric properties. The procedure adopted to yield the following compositions has given in the flow chart.

The dielectric measurements are one of the characterization techniques to understand ferro-electricity of ferroelectric ceramics. The dielectric constant and  $\tan\delta$  of the material under study have been measured at 1KHZ by noting capacitance value using digital LCR meter type LCR-6. The dielectric measurements made in the temperature range from room temperature to  $300^\circ C$ . The temperature has been monitored by using Cr-A1 thermocouple. The e.m.f of thermocouple has been measured with the help of a temperature indicator type Masbus DTI 4001.

**The dielectric constant is calculated from the following formula:**  $K = cd/\epsilon_0 Af$

Where  $c$  is the capacitance,  $d$  the thickness,  $A$  the surface area of sample and  $\epsilon_0$  is the permittivity of free space ( $8.85 \times 10^{-12}$  F/m). Piezoelectric coupling coefficients of ferroelectric ceramics are measured by the resonance-antiresonance technique. This is accomplished by using HP impedance analyzer (Model No.4192A) probe fixter model 16095A was used as intermediary connection between the sample holder and impedance analyzer through BNC adopter. The piezoelectric material behaves capacitively below  $f_r$  and above  $F_a$  between  $f_r$  and  $F_a$  behaves inductively. The phase angle of the element also undergoes a sign change at the resonance and anti-resonance frequencies and therefore can also be used to determine  $f_r$  and  $F_a$ .

**Piezoelectric Coefficients:**  $K_p^2/1 - K_p^2 = f_a / f_a - f_r$   $K_p =$  planar coupling coefficient,  $f_r =$  resonance frequency,  $f_a =$  anti-resonance frequency. The electromechanical thickness coupling coefficient  $K_t$  is determined from overtone frequency using the formula  $K_t^2 = \pi/2 \{ [f_s/f_p] \tan [\pi/2(\Delta f/f_p)] \}$ ,  $f_s$  is series resonance frequency and  $f_p$  is parallel resonance frequency. Mechanical quality factor  $Q_M$  is calculated from the following.  $Q_M = 1/2\pi f_r Z_m C_o \{ f_a^2/f_a^2 - f_r^2 \}$ ,  $Z_m =$  minimum resonance impedance,  $C_o =$  capacitance of the sample at 1KHz. Piezoelectric charge coefficient is the ratio of electric charge generated per unit area to an applied force and is expressed in Coulomb/Newton.

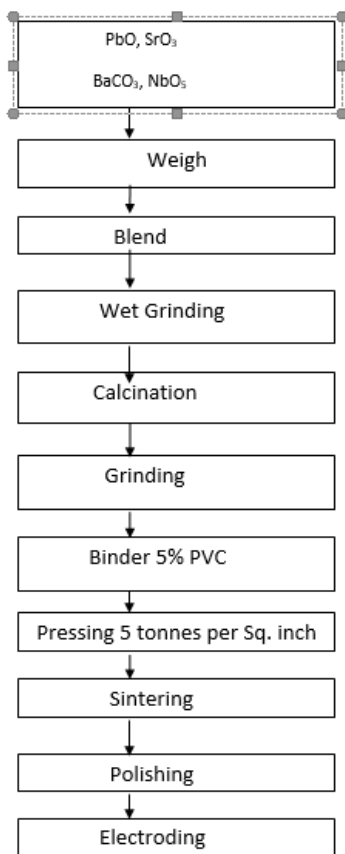


Figure.1.

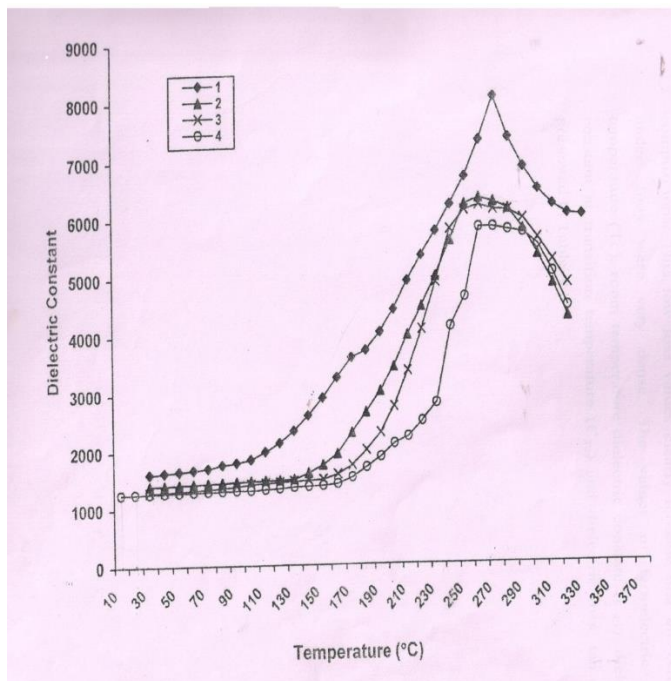


Figure.2. Variation of Dielectric Constant with temperature as a function of composition

Table.1. Dielectric Studies of pure PBN and Sr doped PBN as a function of temperature

PBN Compositions	Dielectric compositions at room temperature $\epsilon_{RT}$	Dielectric constant at transition temperature $\epsilon_{TC}$	Transition temperature $TC^{\circ} C$	Dielectric loss $\tan \delta$
$Pb_{0.6}Ba_{0.4}Nb_2O_6$	1605	8021	270	0.05
$Pb_{0.58}Ba_{0.4}Sr_{0.02}Nb_2O_6$	1410	6318	263	0.04
$Pb_{0.56}Ba_{0.4}Sr_{0.04}Nb_2O_6$	1296	6159	260	0.035
$Pb_{0.55}Ba_{0.4}Sr_{0.05}Nb_2O_6$	1253	5839	262	0.03

**RESULTS AND DISCUSSION**

**Dielectric studies:** The variation of dielectric constant with temperature measured at 1 KHz frequency. These studies are carried out for pure lead barium niobate (PBN) and Sr doped PBN compositions. The maximum dielectric constant is observed at a Curie temperature (Transition temperature) of 270<sup>0</sup>C in pure PBN. The obtained ferroelectric curie temperature T<sub>c</sub> and the maximum dielectric constant ε<sub>Tc</sub> are similar to reported value <sup>20</sup>The ferroelectric transition temperature T<sub>c</sub>,260<sup>0</sup>c has also been reported<sup>21</sup>.It is reported that variation of dielectric constant with temperature of pure PBN is sharp compared with Sr doped PBN. The dielectric curves are broadened with increase of Sr content. The T<sub>c</sub> slightly decreases with Sr dopant and further there is no significant change with increase of Strontium content. The very low loss tanδ ranging from 0.030to 0.050 at room temperature has been observed for all compositions. This low loss values clearly indicate that the ferroelectric curie temperature (T<sub>c</sub>), room temperature dielectric constant (ε<sub>RT</sub>), dielectric constant at transition temperature (ε<sub>Tc</sub>) and dielectric loss tanδ are given in Table.1.

**Piezoelectric studies:** The behaviour of piezoelectric constants such as planar coupling coefficients K<sub>p</sub>, mechanical quality Q<sub>M</sub> dependence on the Sr dopant in PBN at room temperature are given in Table 2.the piezoelectric constants are calculated from the above equation and the piezoelectric strain coefficients d<sub>33</sub> values are presented in Table 2.The variation of K<sub>p</sub> is found to be increases with dopant concentration. Initially K<sub>p</sub> is decreased with dopant and then slightly increase with dopant concentration. The thickness coupling coefficients K<sub>t</sub> is found to increase gradually with increase of dopant content .similar trend has been observed in the hot pressed Lanthanum doped PBN ceramics <sup>22</sup>The quality mechanical factor, Q<sub>M</sub> is found to be 83 for pure PBN.It is almost doubled with strontium dopant and further the Q<sub>M</sub> is slightly decreased with increase of Sr content The Piezoelectric strain coefficient d<sub>33</sub>is obtained as 89x10<sup>-12</sup>C/N for pure PBN which is well agreement with reported values.

**Table.2.Piezoelectric constants**

PBN compositions	Planar coupling coefficient K <sub>p</sub>	Thickness coupling coefficient K <sub>t</sub>	Quality Factor Q <sub>M</sub>	Piezoelectric charge coefficient d <sub>33</sub> x10 <sup>-12</sup> C/N
Pb <sub>0.6</sub> Ba <sub>0.4</sub> Nb <sub>2</sub> O <sub>6</sub>	0.28	0.15	83	89
Pb <sub>0.58</sub> Ba <sub>0.4</sub> Sr <sub>0.02</sub> Nb <sub>2</sub> O <sub>6</sub>	0.27	0.19	150	125
Pb <sub>0.56</sub> Ba <sub>0.4</sub> Sr <sub>0.04</sub> Nb <sub>2</sub> O <sub>6</sub>	0.27	0.19	147	137
Pb <sub>0.55</sub> Ba <sub>0.4</sub> Sr <sub>0.05</sub> Nb <sub>2</sub> O <sub>6</sub>	0.26	0.21	138	155

**CONCLUSIONS**

The maximum dielectric constant is observed at a Curie temperature (Transition temperature) of 270<sup>0</sup>C in pure PBN. It is reported that variation of dielectric constant with temperature of pure PBN is sharp compared with Sr doped PBN. The dielectric curves are broadened with increase of Sr content. The very low loss tanδ ranging from 0.030to 0.050 at room temperature has been observed for all compositions. The quality mechanical factor, Q<sub>M</sub> is found to be 83 for pure PBN. The Piezoelectric strain coefficient d<sub>33</sub>is obtained as 89x10<sup>-12</sup>C/N for pure PBN

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