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# Phase development and dielectric responses in PMN–KBT solid solutions

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**Abstract :**  $(1-x)PbMg1_{3}Nb_{2/3}O_{3}(PMN)-(x)K_{0.5}Bi_{0.5}TiO_{3}$  (KBT)) materials( with x = 0.15,0.25 and 0.35) were prepared by double sintering conventional mixed-oxide method. X-ray powder diffraction patterns reveal that the crystal structure of PMN-KBT crystal changes from cubic to tetragonal symmetry with increasing amounts of KBT(x). The dielectric and ferroelectric properties of PMN-KBT crystals with different compositions near the morphotropic phase boundary (MPB) were studied systematically. The dielectric constant and dielectric loss tangent were measured as a function of both temperature and frequency from impedance data. With increasing KBT content, the frequency dependence of the transition temperature and the diffuseness of phase transition is observed in dielectric peak for sintered specimens. Highphase transition temperature  $(T_m)$  of 319°C was obtained at 1 kHz for the composition x = 0.35. The diffuse phase transition(DPT) of the samples were assessed and the broadest dielectric peak occurs at x = 0.35, with diffusivity value  $\gamma$  = 1.86, which leads to a morphotropic phase boundary in this system. In order to study ferroelectric property of (1-x) PMN-(x)KBT ceramics, polarization vs. electric field (P-E) hysteresis loops were traced at room temperature under different applied field. It is observed that the loop area is increased as the applied voltage has increased. The remnant polarization ( $P_r$ ) and coercive field ( $E_c$ ) are also increased as the applied voltage is increased.

**Keywords :** Ferroelectrics, lead magnesium niobate (PMN), Potassium bismuth titanate (KBT), PMN–KBT, sintering.

### Introduction

Never-ending thirst of man to improve his living kept him working for the advancement of technology <sup>1</sup>. These advanced technology materials needed are those which are more efficient and environmental-friendly materials. One of such smart material is Ferroelectric materials. Ferroelectrics refer to the group of dielectrics having the property of spontaneous polarization (i.e., they retain a dipole even after an applied voltage has been removed). The key characteristics of ferroelectric crystals are that the direction of the polarization can be reversed by application of an electric field and that a hysteresis loop results. Ferroelectrics mainly have two

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characteristics, asymmetry and high dielectric constant or high permittivity <sup>2, 3</sup>. Several ceramic materials with ferroelectric properties have been developed and utilized for a variety of applications. The perovskite family having a structure of the type ABO<sub>3</sub> is the most prominent type of ferroelectrics. Perovskite ferroelectric ceramics can be applied to several electronic devices such as transducers, actuators and sensors <sup>4</sup>. Among the many ferroelectric materials, lead magnesium niobate (PMN) and potassium bismuth titanate (KBT) are attractive because of their excellent electrical properties <sup>5, 6</sup>.

Pioneering studies by Smolenskii <sup>7,8</sup>demonstrated that some of the complex perovskites with mixed Bsite cations, such as  $PbMg_{1/3}Nb_{2/3}O_3$  (PMN) display a broad, high and strongly frequency dependent real part of the permittivity peak as a function of temperature. This behavior is different from observations on conventional ferroelectrics where a sharp and lower peak in the real part of the permittivity is associated with critical behavior evidenced at the phase transition temperature and the Curie-Weiss law is followed when close to this temperature <sup>9</sup>. The Pb( $Mg_{1/3}Nb_{2/3}$ )O<sub>3</sub> (PMN) ceramic, a prototype relaxor ferroelectric with perovskite structure demonstrates a quite high maximum dielectric constant around -10°C with diffuse phase transition phenomena <sup>10,11</sup>. Lead magnesium niobate ferroelectric relaxors (PMN) exhibit desirable dielectric properties, low lossy, a wide dielectric peak, non-hysteric behavior and low sintering temperature <sup>12-14</sup>. However, there are some disadvantages to PMN such as low electro-mechanical coupling, toxicity due to presence of lead in it and low working temperature<sup>15</sup>. Therefore, currently, much effort has been placed on the study of searching and developing the new lead-free or lead-reduced relaxor ferroelectrics for high temperature applications, such as automotive, aerospace, and related industrial applications <sup>16</sup>. So that, toxicity of the lead oxide and its high vapor pressure during processing has resulted in an increasing demand for the alternative materials with reduced toxicity <sup>17</sup>. Moreover, pure PMN perovskite is very difficult to obtain by solid-state reaction because of the unwanted pyrochlore phase. Thus, mixing PMN with other perovskite compound is expected to enhance the formation of more stabilized perovskite structure which means purer perovskite with lower amount of undesirable pyrochlore phases 18

The dielectric properties of relaxor ferroelectric PMN can be enhanced by the addition of ferroelectric materials like PbTiO<sub>3</sub> (PT), lead zirconate titanate PbZr<sub>x</sub>Ti<sub>1-x</sub>O<sub>3</sub>(PZT), etc. .Despite their excellent ferroelectric & piezoelectric properties these materials contain a large amount of lead (> 60 wt. %) which is toxic  $^{19}$ . considerations connected with protection of environment and health of the world's population stimulate intensive research of new alternative FE materials which would not contain lead or would contain it only in small concentration<sup>20</sup>. Potassium Bismuth titanate (( $K_{\frac{1}{2}}Bi_{\frac{1}{2}}$ )TiO<sub>3</sub>, KBT) is one of the few A-site complex perovskites with ferroelectricity and has a promising potential for lead-free piezoelectric materials<sup>21</sup>. As Hiruma and Li eta 1.<sup>22,6</sup> reported, KBT shows typical characteristics of a relaxor ferroelectric, i.e., a broad dielectric peak with a frequency dependent high dielectric maximum temperature ( $T_m$ ) around 380  $^{0}C$ . It is lead free perovskite ceramic so that it is environmental friendly. Also there are some shortcomings to KBT as well. These properties are still notably inferior to that of lead-based ones, for instance, in its electrical properties, it has low dielectric constant  $(\varepsilon_r)^{23-25}$ . It has high curie temperature which result in being unsuitable for working at low temperatures ,hence Usually many applications require that T<sub>C</sub> close to ambient temperature. Therefore, there is a general interest to reduce the  $T_{\rm C}$  of KBT ceramics to optimize their uses<sup>26</sup>. Therefore, one can expect the composite of PMN and KBT to have the advantage of both. Hence, PMN-KBT compositions under investigation were systematically characterized for density, structural, morphological, dielectric and ferroelectric properties. Finally, the relationships among these experimental results are discussed.

#### **Materials and Methods**

Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>(PMN)- K<sub>1/2</sub>Bi<sub>1/2</sub>TiO<sub>3</sub>(KBT) ceramics with x=0.15, 0.25 and 0.35 were prepared by conventional solid state ceramic route. Analytical reagent grade Bi2O3, Nb<sub>2</sub>O<sub>5,3</sub>MgO, TiO2, K<sub>2</sub>CO<sub>3</sub> and PbO of purity ≥ 99.9% were used as raw materials. Perovskite-phase PMN powders were obtained via a well-known columbite method<sup>27</sup>. In this method, the magnesium niobate powder was first prepared by mixing starting MgO and Nb<sub>2</sub>O<sub>5</sub> powders. Stoichiometric amount of these powders with acetone as mixing media were mixed and grinded for 6h using agate mortar and pestel. This mixture was dried and then calcined inside furnace for 6 h at optimized temperature of 800<sup>0</sup>C to form a so-called columbite powder (MgNb<sub>2</sub>O<sub>6</sub>). This synthesized MgNb<sub>2</sub>O<sub>6</sub> columbite was thoroughly mixed in agate mortar and pestle with reactant PbO according to the stoichiometry to obtain fine powder of PMN. Then, the powder was calcined in an alumina crucible at 800 °C for 4h. KBT powder was prepared in similar way by a conventional mixed-oxide method from K<sub>2</sub>CO<sub>3</sub>, Bi<sub>2</sub>O<sub>3</sub>, and TiO<sub>2</sub> starting powders. Stoichiometric amount of these powders with acetone as mixing media were mixed and grinded for 6h using agate mortar and pestel. This mixture was dried and then calcined inside furnace for 4hrs in an alumina crucible at optimized temperature of  $800^{\circ}$ Cto obtain fine powder of KBT. The (1-x)(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>- (x)K<sub>1/2</sub>Bi<sub>1/2</sub>TiO<sub>3</sub>( when x = 0.15, 0.25 and 0.35 ) ceramic systems were prepared from the starting PMN and KBT. Stoichiometric amount of the PMN and KBT powders with acetone as mixing media were mixed and grinded for 8hrs using agate mortar and pestel. This mixture was dried and then calcined inside furnace for 4hrs at optimized temperature of  $800^{\circ}$ C to obtain fine powder of PMN-KBT. These calcined powders were mixed with 2 wt% polyvinyl alcohol (PVA) solution as binder and then pressed to prepare pellets with a diameter of 10 mm and a thickness of 1.5mm under optimized load of 12.5 kN using a steel die of 10 mm diameter and then the samples were sintered at optimized temperature of  $1000^{\circ}$ c for 4hrs in sealed crucibles with heating/cooling rates of  $10^{\circ}$ C/min.

The densities of the sintered ceramics were measured by Archimedes method from the specimens weighed in air, in xylene and the density of xylene. The phase formations of the sintered specimens were studied by Pan Analytic X-ray diffractometer (Philips X'pert) by CuK<sub> $\alpha$ </sub> radiation ( $\lambda$ =1.54 Å) technique. The microstructure analyses were undertaken by a scanning electron microscopy (SEM). Grain size was determined from SEM micrographs by a linear intercept method. For the electrical measurement, silver paste was painted on both sides of the polished samples as electrodes. The dielectric properties of all sintered samples were measured with Impedance analyzer (Auto Lab)in connection with a temperature chamber and a sample holder capable of high temperature measurement. The dielectric constant and loss were measured as a function of temperature (in the range 25–500 °C) and frequency (in the range100 Hz–1MHz). The dielectric constant ( $\epsilon_r$ ) was then calculated using the geometric area and thickness of samples using the formula:

$$\varepsilon_{\rm r} = \frac{Z''}{2\pi f C_{\rm o} |Z|^2} \tag{1}$$

Where Z'' is the imaginary impedance is frequency, |Z| is the magnitude of total impedance, C<sub>0</sub> is the capacitance of the air gap between the electrodes for a separation equal to the thickness of the sample and is given by the geometry of the electrode system. It is given by:

$$C_{o} = \frac{\varepsilon_{o}A}{d}$$
(2)

Where A is area,d is thickness of the sample and  $\varepsilon_o$  dielectric permittivity in vacuum (or) absolute permittivity( $\varepsilon_o = 8.85*10^{-12}$ F/m). The ceramic pellet was immersed in silicon oil at 150°C and poled using a DC poling unit at 15kV/cm. The ferroelectric P–E hysteresis loops were traced with increasing temperature using an automatic P–E loop tracer.

#### **Results and Discussion**

XRD is a rapid analytical technique used for providing the information of unit cell dimensions. The structural analysis of the synthesized powders were characterized by X-ray diffractometer (Philips X'pert) by CuK<sub>a</sub> radiation ( $\lambda$ =1.54 Å) in the 20 range from 20° to 80° with a step size of 0.02 at a scan rate of 2°/min at room temperature for each PMN-KBT composition. Fig.3.1 shows the XRD pattern of the powdered (1x)PMN-xKBT ceramics calcined at 800°C and its sintered pellet at 1000°C. All the XRD patterns confirm the formation of single-phase perovskite structure of (1-x)PMN-xKBT ceramics for the composition x = 0.15, 0.25and 0.35. For the compositions with x = 0.15 (lower concentration of KBT), all the pseudo-cubic reflections shown in Fig.3.1 appear to be singlet. This characterizes a cubic structure with space group Pm3m for this composition <sup>28</sup>. Hence, PMN-KBT for x = 0.15 exhibits cubic crystal structure. The tetragonal structure is characterized by the splitting of (h00)-type reflection peaks <sup>29</sup>. In the present case, the splitting of peak in the vicinity of 45.57° into two peaks (doublet) (002) and (200) confirms the tetragonal nature of (1-x)PMN-(x)KBT ceramic. For the compositions with x=0.25 and 0.35, the examination of (200) pseudo-cubic XRD profile shown in Fig.3.1 suggest that the peaks corresponding to both the cubic and tetragonal phases are present indicating coexistence of these two phases. Thus the compositions with x = 0.25 and 0.35 correspond to the MPB region in which both the tetragonal and cubic phases coexist<sup>30, 31</sup>. With increasing KBT content, there is minute peak shift towards higher angle which could be due to the existence of strain results from planar stress very likely in thin layer peaks<sup>32</sup>. This has a contribution for the slight change of lattice parameters of PMN-KBT samples in the table 3.1 below.



Figure 1. XRD pattern of (1-x) PMN-(x)KBT ceramics

The optimized density, relative density and crystallite size of the sintered (1-x)PMN-xKBT ceramics are listed in Table1 below. The experimental densities of the sintered ceramics of different composition of PbMg1<sub>/3</sub>Nb<sub>2/3</sub>O<sub>3</sub>-K<sub>1/2</sub>Bi<sub>1/2</sub>TiO<sub>3</sub> were measured by Archimedes method from the specimens weighed in air and in xylene. The densities of PMN–KBT ceramics were within the range of 5.42 –6.61 g/cm 3. It should be noted here that the composition and densification of ceramic can influence the dielectric properties<sup>33</sup>. In the present work it was observed, the relative density varies between 95.76 and 96.82%, as shown in Table 3.1, which may play a significant role in the variation of dielectric properties. The crystallite size calculated from XRD datausing Debye Scherer's formula.

$$D = \frac{k\lambda}{\beta \cos\theta} \tag{3}$$

Where the D is crystallite size, k is Debye constant, equal to 0.9,  $\lambda$  is the wavelength of the x-ray

Used,  $\beta$  is the full width half maxima (FWHM),  $\theta$  is the Bragg angle.

Composit ions	Structu re	Lattice Parameters	x-ray density	Density (g/cm <sup>3</sup> )	Relative Density	Porosity %	Crystallite size
			$(g/cm^3)$		(%)		(XRD) nm
X = 0.15	Cubic	$a = 4.52A^{\circ}$	5.66	5.42	95.76	4.24%	17.92
X = 0.25	tetragon	$a = 3.98 A^{\circ}$	6.77	6.49	95.91	4.08%	28.63
	al	$c = 4.87 A^{\circ}$					
X =0.35	tetragon	$a = 3.97 A^{\circ}$	6.83	6.61	96.82	3.18%	28.67
	al	$c = 4.85 A^{\circ}$					

Table1. Structure, crystallite size and density of PMN-KBT compositions

SEM micrographs of (1-x)PMN-(x)KBT ceramics with composition x=0.15, 0.25 and 0.35 are shown in Figure 2 (a), (b) and (c) respectively. The average grain size for x=0.15, 0.25 and 0.35 is obtained to be ~ 0.150µm, 0.218µm and 0.246µm respectively, which suggests slightly increasing trend in the grain size with increasing KBT concentration. The microstructure reveals that the compound consists of well-defined grains. Few pores are observed, which is in agreement with the relative density which implies the existence of pores. It should be noted that some of the grains are observed to be in irregular shapes with few voids are seen in SEM images in all compositionswhich may be due to evaporation of slight Pb concentration from the surface of the sample<sup>33</sup>.This may not influence the properties of the samples.



Figure2.SEM micrographs for sintered pellets of different composition, (a) x = 0.15, (b) x = 0.25, (c) x = 0.35

Figure3 (a), (b) and (c) show the EDX spectra of (1-x)PMN-xKBTceramics. EDAX spectrum shows the X-ray peaks associated with individual elements. The X-ray spectrum obtained gives information on the elemental composition of the material under examination. The researcher observed the peaks corresponding to Pb, Mg, Nb, K, Bi, Ti and O elements. This technique is generally associated with scanning electron microscope (SEM). From the plot it is observed that the elements in the prepared compound are identified. It is clearly seen that the prepared (1-x)PMN-xKBT ceramics compound is only composition of Pb, Mg, Nb, K, Bi, Ti and O.



Figure3. Representation of crystal composition spectrum, (a) x = 0.15, (b) x = 0.25, (c) x = 0.35

The temperature dependence of dielectric constant ( $\varepsilon_r$ ) and dielectric loss (tan $\alpha$ ) at various frequencies for compositions with x = 0.15, 0.25 and 0.35 is shown in Figure4 (a),(b),(c) and Figure5(a),(b),(c) respectively. Temperature dependence of dielectric constant ( $\varepsilon_r$ ) was calculated at three fixed frequencies for 1kHz, 400KHz, 1MHz in the temperature range of RT to 500°C. The dielectric behavior is of diffuse type and the dielectric anomaly peak is broad spreading in the temperature range of 290°C-330°C and the maximum dielectric temperature is found to be 319°c for 0.65PMN-0.35KBT composition at 1 KHz. This implies that, When KBT is added to form the binary system with PMN, the maximum temperature is shifted towards that of KBT ceramic. The observed dielectric constant is frequency dependent. The maximum value of  $\varepsilon_r$  observed is 971 at 1kHz frequency. This is greater than that observed by Rao and sankarum<sup>24</sup>,Weda eta 1.<sup>34</sup> for KBT ceramics but this obtained value is much lower than that observed by Yimnirun<sup>33</sup>, Guha<sup>35</sup> and Park and Shrout<sup>36</sup> for PMN ceramic and this value also lower than that observed by Cao<sup>37</sup> and Yimnirun<sup>33</sup> for PMN-33%PT and 0.7PMN-0.3PZT respectively.With increasing KBT content, the maximum transition temperature (T<sub>m</sub>) shifts from 75°c monotonously to higher temperatures at 319°c and the maximum dielectric constant ( $\varepsilon_m$ ) value significantly rises after adding more KBT, which may be attributed to microstructure and density of material<sup>38</sup>. This behavior is similar to PMN-PT<sup>39</sup>, PMN-BNT<sup>18</sup> and PZT-PMN<sup>38</sup>.



Figure 4. Variation of dielectric constant ε as a function of temperature at 1kHz, 400kHz & 1MHz for (a) 0.85PMN-0.15KBT, (b) 0.75PMN -0.25KBT,(c) 0.65PMN- 0.35KBT

The broadest dielectric peak is found at the 0.7PMN–0.3KBT composition which indicates diffused phase transition behavior. However, it is known that PMN and KBT have relaxor characteristics and typical relaxor behavior is characterized by a diffused ferroelectric phase transition. Relaxor ferroelectrics are characterized by diffuse phase transition which has been interpreted as a result of short range order <sup>40</sup>. This behavior is explained in terms of a coexistence region between phases <sup>41</sup>.



Figure 5. Variation of loss tangent tanα as a function of temperature at 1kHz, 400kHz, and 1MHz for (a) 0.85PMN-0.15KBT, (b) 0.75PMN-0.25KBT, (c) 0.65PMN-0.35KBT

Figure5shows the variation of loss tangent tan $\alpha$  as a function of temperature for (1-x) PMN-(x)KBT compositions for x = 0.15, 0.25 and 0.35 respectively. From Figure5, it is observed that tan $\alpha$  passes through a peak just below temperature T<sub>m</sub>, which appears to be a common behavior for ferroelectric systems<sup>39</sup>. For temperature T>T<sub>m</sub>, tan $\alpha$  possess minimum values and remains almost constant with further increase in temperature. The observed values of  $\varepsilon$  and tan $\alpha$  at T<sub>m</sub> for (1-x)PMN-(x)PT are listed in the Table2.

Figure 6shows the variation of dielectric constant ( $\varepsilon$ ) as a function of frequency (log f) for (1-x)PMN-(x)KBT compositions with x = 0.15, 0.25 and 0.35 respectively. From the Figure6, it is observed that the dielectric constant  $\varepsilon$  slowly decreases with increase in applied frequency f. This can be explained on the basis of oscillation of free dipoles in presence of alternating field. At very low frequencies (f << 1/ $\tau$ ,  $\tau$  is the relaxation time), dipoles follow the electric field, while at very high frequencies (f >> 1/ $\tau$ ) dipoles begin to lag behind the field and  $\varepsilon$  slightly decreases <sup>42,44</sup>. It is also reported that the higher values of  $\varepsilon$  at low frequencies is due to space charge polarization or accumulation of charges at interfaces of sample and electrode <sup>44</sup>. From the Figure6, it is observed that the PMN-KBT for x = 0.35 exhibits higher value of  $\varepsilon$ , while it is moderately high for x = 0.25.



Figure 6. Variation of dielectric constant ( $\epsilon$ ) as a function of frequency (log f) for (1-x)PMN-xKBT ceramics With x = 0.15, 0.25 and 0.35

Figure7shows the variation of loss tangent tane as function of frequency f (from 100 Hz to 1MHz) for PMN-KBT compositions with x = 0.15, 0.25 and 0.35 respectively at 500<sup>o</sup>c. In present case 0.85PMN-0.15KBT posses higher values of tan $\alpha$  in the range of 100 Hz to 1MHz. The values of dielectric constant  $\epsilon$  and tan $\alpha$  at both T<sub>m</sub> and room temperature for a frequency of 1kHz for PMN-KBT compositions are given above in the Table 2.



Figure 7. Variation of loss tangent tan $\alpha$  as a function of frequency (log f) for (1-x) PMN-xKBT ceramics with x = 0.15, 0.25 and 0.35.

To assess the diffuse phase transition of relaxor ferroelectric, the degree of broadening or diffuseness in the observed dielectric variation could be estimated with the diffusivity ( $\gamma$ ) using the ln  $(\frac{1}{\epsilon} - \frac{1}{\epsilon_m})$  versus ln(T - T<sub>max</sub>). A material that shows a diffused phase transition (broad peak) deviates strongly from Curie–Weiss law. Hence, the degree of diffuseness for any material can be calculated by using the modified Curie Weiss law <sup>41</sup>as given in Eq. (4). If the local Curie temperature distribution is Gaussian, the reciprocal permittivity can be written in the form:

$$\frac{1}{\epsilon} = \frac{1}{\epsilon_m} + \frac{(T - T_m)^{\gamma}}{2\epsilon_m \delta^2} \tag{4}$$

Where  $\varepsilon_m$  is maximum dielectric constant,  $\gamma$  is diffusivity, and  $\delta$  is diffuseness parameter.  $\gamma$  and  $\delta$  can be estimated from the slope and intercept of the dielectric data shown in Figure 8, which should be a linear. The values of  $\gamma$  and  $\delta$  are both material constants depending on the composition and structure. In a composition with the diffused phase transition such as relaxor ferroelectric, the value of  $\gamma$  is expected to be 2<sup>44</sup>. For  $\gamma = 1$ , the system behaves as a normal ferroelectric system, while for  $\gamma = 2$  it depicts a relaxor system<sup>45</sup>. In order to demonstrate the relaxor behavior of PMN-KBT ceramic, a graph between  $\ln(1/\epsilon - 1/\epsilon_m)$  and  $\ln(T-T_m)$  shown in Figure8 is plotted at frequency 1 kHz. The slope of the graph gives the value of  $\gamma$ . The value of  $\gamma$  reported in Table 3.2 varies between 1.60 and 1.86 which confirms relaxor behaviors. However, the results show the degree

of diffuseness of the phase transition to be highest at the 0.65PMN–0.35KBT composition, which lead to a morphotropic phase boundary (MPB) in this system <sup>47</sup>. In the MPB regions, enhanced dielectric constants are obtained. It was attributed to easily switching of the polarization vector between all allowed polarizations orientations.

Table2. The values of dielectric constant at room temperature( $\epsilon_{RT}$ ) and at transition temperature( $T_m$ )( $\epsilon_{max}$ ), loss tangent tan $\alpha_{RT}$  and tan $\alpha_{Tm}$ , transition temperature and Diffusivity ( $\gamma$ ) for PMN-KBT compositions

Sample name	ε <sub>RT</sub>	$tan\alpha_{RT}$	$T_m(o_c)$	<b>E</b> max	$tan \alpha_{Tm}$	(γ)
0.85PMN-0.15KBT	125	0.2	75	130	0.1	1.66
0.75PMN-0.25KBT	200	0.15	222	320	0.05	1.60
0.65PMN-0.35KBT	581	0.05	319	971	0.03	1.86



Figure 8. Plot of  $ln(T-T_{max})$  versus  $ln((1/\epsilon_r) - (1/\epsilon_{max}))$  for (1-x)PMN-(x)KBT solid solutions

Hysteresis loop is the fingerprint of ferroelectricity. In order to study ferroelectric property of (1-x) PMN–xKBT ceramics, polarization versus electric field (P–E) hysteresis loops were traced at room temperature under different applied field conditions. Figure9, a, b and c shows ferroelectric hysteresis loop at room temperature. Room temperature spontaneous polarization, remnant polarization and coercive electric field values were found to be listed in table 3 which suggested its utility as switching devices. From the figures, It is observed that the loop area is increased as the applied voltage has been increased. The remnant polarization (P<sub>r</sub>) and coercive field (E<sub>C</sub>) are also increased as the applied voltage is increased.



Figure 9. Ferroelectric hysteresis loop of (1-x)PMN–xKBT ceramics at room temperature: (a) x = 0.15, (b) x = 0.25, (c) x = 0.35

The ferroelectric loops indicate the (1-x)PMN-xKBT samples have high loss in energy. From the table2 it was observed that for the composition of 0.35, the maximum obtained values of  $P_s$ ,  $P_r$ , and  $E_c$  are to be 0.393  $\mu$ C/cm<sup>2</sup>,0.246 $\mu$ C/cm<sup>2</sup> and 6.401kV/cm at an applied voltage of 7kV .When compared with the KBT values obtained by Li et al.<sup>6</sup>, the values are greater (For KBT,  $P_r = 0.058 \mu$ C/cm<sup>2</sup> at room temperature). But When compared with the PMN-PT values obtained by Hussain<sup>48</sup>, the values are less (The 0.64PMN–0.36PT room temperature spontaneous polarization, remnant polarization and coercive electric field were found to be 19.74 $\mu$ C/cm<sup>2</sup>, 13.27 $\mu$ C/cm<sup>2</sup> and 7.55kV/cm, respectively).However the PMN-KBT samples are good low lead based ferroelectric materials which are suitable for device applications.

Composition	Applied field(kv/cm)	$P_s(\mu C/cm^2)$	$P_r(\mu C/cm^2)$	E <sub>c</sub> (kv/cm)
	5	0.081	0.054	1.427
0.15	6	0.156	0.084	2.189
	7	0.190	0.117	3.200
	5	0.242	0.154	3.792
0.25	6	0.300	0.184	4.747
	7	0.342	0.214	5.808
	5	0.304	0.184	4.651
0.35	6	0.354	0.220	5.691
	7	0.393	0.246	6.401

Table3. Data obtained from P-E hysteresis loops at various applied fields

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