iJRASET
International Journal For Research in
Applied Science and Engineering Technology

INTERNATIONAL JOURNAL FOR RESEARCH

IN APPLIED SCIENCE & ENGINEERING TECHNOLOGY

Volume: 6 **Issue:** XII **Month of publication:** December 2018

DOI:

International Journal for Research in Applied Science & Engineering Technology (IJRASET)
ISSN: 2321-9653; IC Value: 45.98; SJ Impact Factor: 6.887
Volume 6 Issue XII, Dec 2018- Available at www.ijraset.com

AC Conductivity and Piezoelectric Properties of(1-X)$PbMg1_{/3}Nb_{2/3}O_3$-(X)$K_{0.5}Bi_{0.5}TiO_3$ Ceramics

Kebede Legesse[1], P. Vijaya Bhaskar Rao[2]
[1, 2]Department of physics, college of natural and computational science, Wollega University, Post Box No: 395, Nekemte, Ethiopia

***Abstract:** AC conductivity and piezoelectric properties of (1-x) PbMg1/ 3Nb2/ 3O3-(x) K0.5 Bi0. 5TiO3 ceramics are important for investigating viability of these relaxor ferroelectric materials for sensor and actuator applications. Hence no studies seem to be available on PMN-KBTcomposition ceramics systems, in this paper, we report detailed study of ac conductivity and piezoelectric property of (1-x)PbMg1/3Nb2/3O3-(x)K0.5Bi0.5TiO3 ceramic with composition x=0.15, 0.25 and 0.35.The polycrystalline ceramic samples of ((1-x)PbMg1/3Nb2/3O3(PMN)-(x)K0.5Bi0.5TiO3 (KBT)) (x = 0.15, 0.25, and 0.35) were prepared by solid-state reaction technique at high temperature. AC conductivity and piezoelectric properties of the materials have been investigated within a wide range of temperature and frequency using complex impedance spectroscopy (CIS) technique using computer controlled impedance analyzer. It is observed that ac conductivity (σac) increases with increase in temperature showing negative temperature coefficient of resistance (NTCR) behavior. The ac conductivity (σac) shows an increasing trend with the increasing frequency and temperature. The study also showed that, Activation energies decreased with increase of the amo-unt of KBT in all samples.*

Frequency dependent ac conductivity at different temperatures indicates that the conduction process is thermally activated. The frequency dispersion spectrum obtained in this study shows two well defined frequency regions in which the conductivity follows a double power law behavior. Longitudinal piezoelectric charge coefficients of the samples poled under a dc electric field of ~ 9kV/mm at 150 °C in a silicone oil bath were found to be equal to 58 pC/N , 67pC/N, and 79.4 pC/N for the composition of 0.15,0.25 and 0.35 respectively. The electromechanical coefficients of the samples were also found to be 0.63, 0.66 and 0.68 for the composition of 0.15, 0.25 and 0.35 respectively.

Keywords. AC conductivity; piezoelectric properties; lead magnesium niobate- Potassium bismuth titanate.

I. INTRODUCTION

Perovskite type oxides have gained much attention as having many excellent physical properties such as ferroelectric, piezoelectric, magneto electric and electro optic effects [1]. 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 a ferroelectric crystal 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 characteristics, asymmetry and high dielectric constant or high permittivity [2-3]. Several ceramic materials with ferroelectric properties have been developed and utilized for a variety of applications. Perovskite ferroelectric ceramics can be applied to several electronic devices such as transducers, actuators and sensors [4]. Piezoelectricity is the ability of certain crystalline materials to develop an electrical charge proportional to a mechanical stress [5]. The piezoelectric effect is exhibited by a number of naturally-occurring crystals, for instance quartz, tourmaline and sodium potassium tartrate, and these have been used for many years as electromechanical transducers. Ferroelectric relaxors belong to a subgroup of ferroelectric materials. Ferroelectric relaxors belong to a subgroup of ferroelectric materials. Relaxors are characterized by unique dielectric, polarization, electromechanical and electro-optical properties. A piezoelectric material develops an internal electric field when strained/deformed. On the contrary, a piezoelectric material experiences strain when an electrical field is applied to it. These reactions, electrical field and mechanical behavior can be in either direction. Meaning that depending on the material, an electrical field in one direction can lead to a mechanical reaction in any direction.

Among the many ferroelectric and piezoelectric materials, lead magnesium niobate (PMN) and potassium bismuth titanate (KBT) are attractive because of their excellent electrical properties [6-7]. Pioneering studies by Smolenskii [8-9] demonstrated that some of the complex perovskites with mixed B-site 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 [10]. The

$Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN) ceramic, a prototype relaxor ferroelectric with perovskite structure demonstrates a quite high maximum dielectric constant around -10^0C with diffuse phase transition phenomena [11-13]. Lead magnesium niobate ferroelectric relaxors (PMN) exhibit desirable dielectric properties, low lossy, a wide dielectric peak, non-hysteric behavior and low sintering temperature [14-16]. 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 [17]. 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 [18]. 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 [19]. 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 [20]. The physical properties of relaxor ferroelectric PMN can be enhanced by the addition of ferroelectric materials like $PbTiO_3$ (PT), lead zirconate titanate $PbZr_xTi_{1-x}O_3$(PZT), etc. Despite their excellent ferroelectric & piezoelectric properties, these materials contain a large amount of lead (> 60 wt. %) which is toxic[21]. 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[22]. Potassium Bismuth titanate $(K_{½}Bi_{½})TiO_3$) (KBT) is one of the few A-site complex perovskites with ferroelectricity and has a promising potential for lead-free piezoelectric materials[23]. As Hiruma [24] 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. But there are also 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 (ε_r)[25-27]. It has high curie temperature which result in being unsuitable for working at low temperatures ,hence Usually many applications require that T_C close to ambient temperature. Therefore, there is a general interest to reduce the T_C of KBT ceramics to optimize their uses [28].

Therefore, one can expect the composite of PMN and KBT to have the advantage of both. Hence no studies seem to be available on PMN-KBT systems, in this paper, we report the detailed study of ac conductivity and piezoelectric properties of ((1-x)$PbMg1_{/3}Nb_{2/3}O_3$(PMN)-(x)$K_{0.5}Bi_{0.5}TiO_3$ (KBT)) ceramic with x=0.15, 0.25 and 0.35.

II. EXPERIMENTAL PROCEDURES

$Pb(Mg_{1/3}Nb_{2/3})O_3$(PMN)- $K_{1/2}Bi_{1/2}TiO_3$(KBT) ceramics with x=0.15, 0.25 and 0.35 were prepared by conventional solid state reaction route. Analytical reagent grade Bi_2O_3, Nb_2O_5,, MgO, TiO_2, K_2CO_3 and PbO of purity ≥ 99.9% were used as raw materials. Perovskite-phase PMN powders were obtained via a well-known columbite method [29]. In this method, the magnesium niobate powder was first prepared by mixing starting MgO and Nb_2O_5 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 800^0C to form a so-called columbite powder ($MgNb_2O_6$). This synthesized $MgNb_2O_6$ columbite was thoroughly mixed using agate mortar and pestle with reactant PbO according to the stoichiometry to obtain fine powder of PMN. The powder was calcined in an alumina crucible at 800◦C for 4h. Then, the calcined powders were checked for phase purity using x-ray diffractometer. KBT powder was prepared in similar way by a conventional mixed-oxide method from K_2CO_3, Bi_2O_3, and TiO_2 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 800^0C to obtain fine powder of KBT. Then, the calcined powders were checked for phase purity using x-ray diffractometer.

The (1-x)$(Mg_{1/3}Nb_{2/3})O_3$- (x)$K_{1/2}Bi_{1/2}TiO_3$ (when x = 0.15, 0.25 and 0.35) ceramic systems were prepared from 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 800^0C to obtain fine powder of PMN-KBT. Using polyvinyl alcohol (PVA), the calcined powders were cold pressed into cylindrical pellets of 10mm diameter and 1.5mm thickness under optimized load of 12.5 kN using a hydraulic press. PVA was used as binder to reduce the brittleness of the pellet, which burnt out during high temperature sintering. Then, the pellets were sintered at an optimized temperature of 1000 ◦C for 4 h so as to get nearly 97% of theoretical density.

For conductivity measurement, Complex impedance data on coated (1-x)PMN-(x)KBT samples was recorded using computer controlled impedance analyzer (AUTOLAB) in the temperature range of room temperature to 500^oC in the frequency range of 100Hz-1MHz. For d_{33} measurement the samples were coated with silver paste and dried under infrared lamp. Electrical poling of the sample immersed in a silicon oil bath was done at 90kV/cm, at a temperature 150 ^{O}C for 30 minutes. The samples were cooled to room temperature in the presence of the field and the d_{33} was measured using piezometer (d_{33}-meter). For the measurement of electromechanical coefficient (K_p) the point contacts were made on poled samples using silver paste using a thin copper wire. The sample was then hanged in the furnace and the copper wires were through high alumina narrow bored tubes and two terminals were fixed to two terminals on a plate fixed at the top of the Faraday cage(a metal box).The entire sample holder was shielded to avoid any possible stray pickups. The admittance measurements were performed using impedance analyzer as a function of frequency at room temperature.

III. RESULTS AND DISCUSSION

A. AC Conductivity Analysis

The motion of charges in the dielectric gives rise to the conduction current and additionally polarizes the dielectric. Study of electrical conductivity in ferroelectric materials is very important since properties like piezoelectric, pyroelectric, etc. depend on it. The ac conductivity (σ_{ac}) of the material depends on the dielectric nature of the sample. There is a strong correlation between the frequency response and the temperature dependence of electrical conductivity of the materials. The temperature dependence of σ_{ac} shows the presence of single or multiple relaxations in the material. In bulk materials two types of ac conductivity phenomena occurs; (a) long range ac conductivity which is due to the fact that at low frequencies the overall conductivity is due to the mobility / transportation of charge carriers over long distance and (b) localized transport oxygen vacancies which is due to relaxation / orientational mechanism in which case the charge mobility / transportation is restricted only to the nearest neighboring lattice sites [30]. In polar materials the conduction mechanism can be ionic and/or electronic in nature. Depending upon the purity and temperature of the materials the proportion of ionic to electronic conduction in the materials varies. Usually polar dielectric materials show an increasing trend in conductivity with rise in temperature. The variation of electrical conductivity with temperature is explained by the equation

$$\sigma = Aexp(\frac{-E_a}{K_BT}) + Bexp(\frac{-E_b}{K_BT}) \qquad (3.1)$$

Where E_a and E_b are the activation energy for the intrinsic and extrinsic conduction process respectively and A and B are constants. At higher temperatures the intrinsic conduction process dominates, and hence the eqn. (3.1) reduces to:

$$\sigma = Aexp(\frac{-E_a}{K_BT}) \qquad (3.2)$$

At low temperature, conductivity is dominated by mobility of extrinsic defects were carrier concentration is fixed by doping. At High temperature, conductivity is due to thermally formed (intrinsic) defects in which the carrier concentration varies with temperature. Most metal oxides are electronic conductors at high temperatures. For many of these oxides the conductivity increases with increasing temperature and as the conductivity at the same time is much smaller than in metals, this type of conductivity is termed semi conductivity. The principal reason for the increasing conductivity is that the number of electronic defects increases with increasing temperature. Electrical conductivity is an important experiment tool to probe the structural defects and internal purity of semi-insulating crystalline solids because unlike metals and semiconductors where electrons are the charge carriers, in semi-insulators the prime factors contributing to their electrical conductivity are the crystal defects. The electrical conductivity studies of an electro ceramics are complex. The real part of AC conductivity is given by:

$$\sigma'_{ac} = \omega_i \varepsilon_o \varepsilon'' \qquad (3.3)$$

Where $w = 2\pi f$, (f being the frequency used), ε_o is the permittivity of vacuum (8.854×10-12 F/m) and ε'' is the dielectric loss factor. Thus, σ'_{ac} is directly related to the dielectric properties of the material. Alternatively, the real part of the dominant bulk conductivity may be evaluated from the impedance spectrum using the relation:

$$\sigma'ac \approx \frac{t}{Z' \times A} \qquad (3, 4)$$

$$\sigma'ac \approx y' \times \frac{t}{A} \qquad (3.5)$$

Where σ'_{ac} is real part of conductivity, *Z'* is the real part of complex impedance (intersection of semicircle on the real-axis in *Z"* vs. *Z'* plot); *t* the thickness, and *A* the surface area of the sample, t is thickness of sample and A is the area of the sample and Y′ is real part of the admittance.

The variation of the real part of the ac conductivity with 1000/T is shown in the (Figs. 3.1a-c), at different frequencies. At lower temperatures, the complexes formed are immobile and the orientation effect is suppressed and doesn't contribute much to the conductivity. Even though, there is electronic and ionic hopping in ceramic conduction, at low temperatures, the electronic hopping conduction dominates the conductivity and ionic hopping makes little contribution [31]. At low frequencies the overall conductivity is due to the mobility / transportation of charge carriers over long distance rather than from relaxation / orientation mechanism in which case the charge mobility / transportation is restricted only to the nearest neighboring lattice sites [32].

As the temperature increase, impurity defect complex start to dissociate and there by contribute to conductivity. Because at low temperatures the conductivity shows weak temperature dependence and the values of conductivity are higher for higher frequencies, hence at low temperature the conductivity is frequency dependent. It is observed that σ_{ac} increases with increase in temperature showing negative temperature coefficient of resistance (NTCR) behavior. Generally, the σ_{ac} shows an increasing trend with the increasing frequency and temperature. The increase in the σ_{ac} with the increasing temperature can be attributed to the impurities or dislocations at metal semiconductor interface. These impurities lie below the bottom of the conduction band and thus it has small activation energy. (Fig3.1a-c) also shows that, increasing the composition(x value) increased the samples polarization conductivity. The nature of variation shows almost linear over wider range temperature regions and obeys the Arrhenius relationship. The ac conductivity (σ_{ac}) is related to the activation energy and the inverse absolute temperature, using an empirical relation:

$$\sigma_{ac} = \sigma_o e^{-E_a/k_B T} \qquad (3.6)$$

Where σ_o is the composite constant or the pre exponential factor, E_a the activation energy of the Conduction mechanism, k_B the Boltzmann constant and T is the temperature in Kelvin [33].The Arrhenius plots (lnσ_{ac} vs 1000/T) of ac conductivity at different frequencies are shown in Fig. 3.1. The activation energy (E_a) was calculated from the straight line fitting of plots shown in Fig. 3.1 and the values are presented in (table 3.1). As shown in (table3.1), the E_a decreases with the increasing frequency. The increase in the applied field enhances the charge carrier's jumps between the localized states (hopping), consequently, the activation energy decreases with the increasing frequency. Also, the obtained low values of activation energy suggest that the conduction mechanism may be due to the hopping of electrons [34-36].

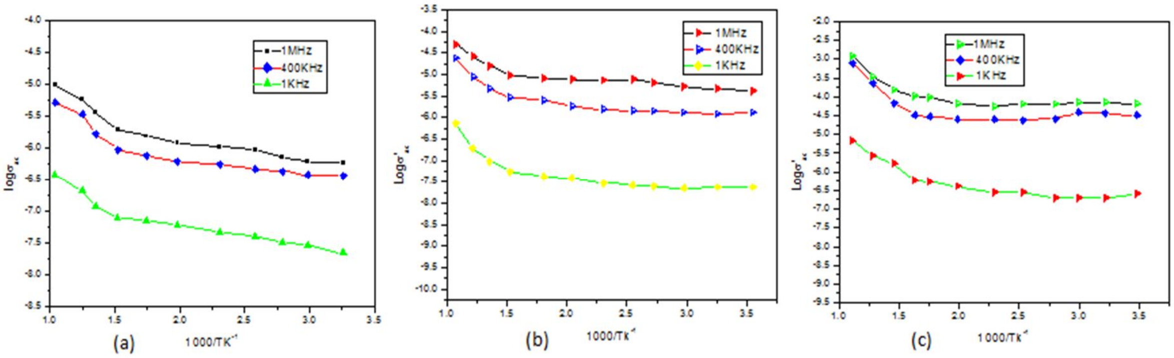

Fig.3.1 Arrhenius plots of ac conductivity at different frequencies for (a) 0.85PMN-0.15KBT, (b) 0.75PMN-0.25KBT and (c) 0.65PMN-0.35KBT compositions

Activation energies are decreased with increase of the amount of KBT in all samples.This may due to the disordering cations between the neighboring sites which promote the growth of polarization conduction (ferroelectric phase). The higher concentration of KBT substituted for PMN distorts the unit cell, changes the dipolar moment, and induces strain in the lattice [37]. The high activation energies at high temperature suggest that the conduction mechanism of the compounds may be due to hopping of charge carriers. The activation energy of the compound decreases on increasing frequency. So that, in higher temperature region the activation energies are found to decrease with increase in frequency.

Table3.1: Temperature regions and associated activation energies For PMN- KBT ceramics

Composition (x)	Activation Energy (eV)		
	At (1kHz) (400-500^{O}C)	At(400KHz) (400-500^{O}C)	At(1MHz) (400-500^{O}C)
0.15	0.74	0.50	0.41
0.25	0.52	0.35	0.30
0.35	0.28	0.22	0.19

(Figure3.2) shows logσ_{ac} versus logf plots of frequency dependent ac conductivity of (1-x)PMN-xKBT compositions at various temperatures. A typical frequency dependence of conductivity exhibits three distinguished regions: (a) low frequency dispersion (b) an intermediate frequency plateau (frequency independent dc conductivity) and (c) an extended dispersion at high frequency [38]. The variation of conductivity in the low frequency is attributed to the polarization effects at the electrode and electrolyte interface. As the frequency reduces, more and more accumulation occurs at the electrode & electrolyte interface and hence drops in conductivity. In the intermediate frequency plateau, conductivity is almost found to be frequency independent and is equal to dc conductivity σ_{dc}. In the high frequency region, the conductivity increases with increase in frequency. The frequency dependence of conductivity or so-called universal dynamic response (UDR) of ionic conductivity is related by a simple expression given by Jonsche′s power law [39].

$$\sigma(\omega) = \sigma_{dc} + \sigma'_{ac} = \sigma_{dc} + A\omega^{n} \qquad (3.7)$$

Where $\sigma(\omega)$ is the total conductivity and $A\omega^{n}$ is called the *ac* conductivity

The first term is the temperature-dependent (frequency-independent) dc conductivity due to the excess of electrons in the conduction band, and is related to the drift mobility of the electric charge carriers. The second term represents the frequency and temperature dependent ac conductivity (σ_{ac}) and is attributed to the dielectric relaxation caused by the localized electric charge carriers and where A is ac constant for a particular temperature and n is a power exponent which represents the degree of interaction between mobile ions and the environment surrounding them which generally varies between 0 and 1[40]. In the present study, the frequency dependence of ac conductivity does not seem to follow the simple Jonscher’s power law. Rather it seen to follow a double power law [41-45] given as:

$$\sigma(\omega) = \sigma(o) + A_1\omega^{n_1} + A_2\omega^{n_2} \qquad (3.8)$$

Where σ_0 is the frequency independent (electronic or DC) part of AC conductivity. The exponent $n_1(0 \le n_1 \le 1)$ characterizes the low frequency region i.e., it corresponds to the grain-boundary conductivity corresponding to the translational ion hopping whereas the exponent n_2 $(0 < n_2 < 2)$ characterizes the high frequency region i.e., to the grain conductivity indicating the existence of well localized relaxation/re-orientational process [46]. It is evident that when $\omega = \omega_p$, both terms in above equation are equal to the dc conductivity where from the figure, it is noticed that in the low frequency region, the second term in the above equation makes no contribution to the ac conductivity while the first term doesn’t contribute to the conductivity at high frequencies. Taking in account the dc contributions in both frequency regimes, the following expressions hold for the experimental data:

$$\sigma(\omega) \approx \sigma(o) + A_1\omega^{n_1} \quad , \omega < \omega_p \qquad (3.9)$$

$$\sigma(\omega) \approx \sigma(o) + A_2\omega^{n_2} \quad , \omega > \omega_p \qquad (3.10)$$

Where $n_2=0$ for $\omega < \omega_p$ and $n_1=0$ for $\omega > \omega_p$, where ω_p is the frequency at which slope change occurs and shifts to lower frequencies with increase in temperature and this frequency at which the slope changes is known as hopping frequency.

As indicated above, the frequency dispersion spectrum obtained in this study shows two well defined frequency regions in which the conductivity follows a power law behavior with different exponent’s n_1 and n_2 for low and high frequency regions, respectively. Schematic plot of the conductivity data exhibiting both low and high frequency dispersion is given by fig.3, 2 below.

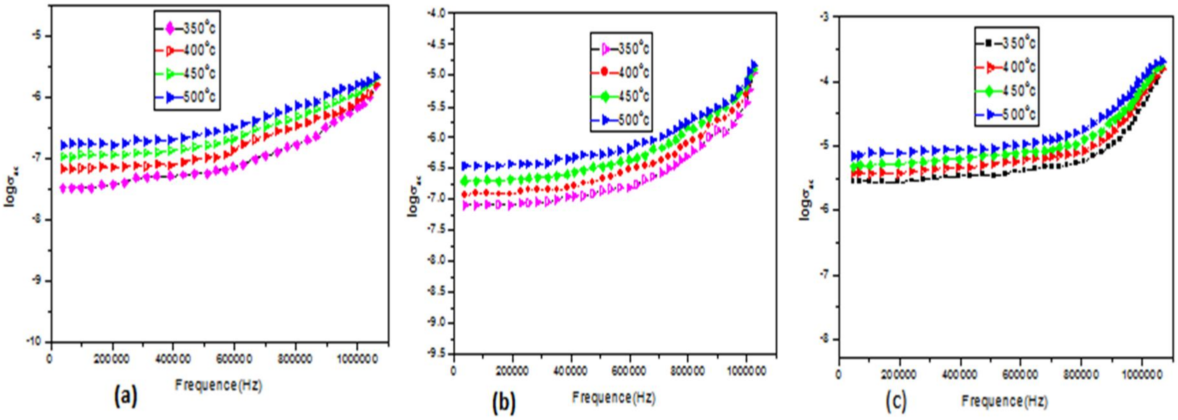

Fig.3.2 Frequency dependence of the ac conductivity at different temperatures for (a) 0.85PMN- 0.15KBT, (b) 0.75PMN-0.25KBT, (c) 0.65PMN-0.35KBT compositions

B. Piezoelectric Property

Piezoelectric materials have the ability to provide desired transformation from mechanical to electrical energy and vice versa. When a mechanical force is applied to the piezoelectric material an electrical voltage is generated and when an electrical voltage is applied to the piezoelectric material it gets strained or deformed. Owing to these characteristics piezoelectric materials can be used as a sensor, an actuator, as well as a power generation unit. Among many piezoelectric materials studied, the best material is modified $Pb(Ti,Zr)O_3$ whose coupling coefficient values range in between 0.5-0.7. Rochelle salt (Potassium sodium tartrate tetrahydrate) ($KNaC_4H_4O_6 \cdot 4H_2O$) at its Curie point has a coupling coefficient as much as 0.9 [26]. The piezoelectric properties of (1-x)PMN–xKBT solid solutions are summarized in table 3.2. The piezoelectric constant (d_{33}, induced polarization in direction 3 (parallel to direction in which ceramic element is polarized) per unit stress applied in direction 3 or induced strain in direction 3 per unit electric field applied in direction 3) value has been measured for the poled (1-x)PMN-(x)KBT samples. The poling of piezoelectric ceramics after fabrication is an important stage in the results obtained for the samples. Because of the possibility of dielectric break down under high-applied fields, the objective in all poling situations is to induce the maximum degree of domain re-orientation for the lowest applied field and in the shortest possible time. In this study, Poling treatment was carried out in the temperature range from room temperature to 150^{O}C and in the electrical field range from 9kV/mm. The piezoelectric constant (d_{33}) obtained is 70.6pC/N, 74.3pC/N,79.4pC/N for compositions (x) 0.15, 0.25 and 0.35 respectively. As it is given in the table3.2 below, the d_{33} values were measured for (1-x)PMN-(x)KBT compositions (x= 0.15, 0.25 and 0.35). It is observed that the ceramic x=0.35 composition has shown the maximum d_{33} =79.4pC/N, when compared with other compositions.

The admittance measurements as a function of frequency in the radial mode of thin (1-x)PMN-(x)KBT samples yields resonance and anti- resonance peaks at room temperature as shown in figure 3.3 a,b and c. The resonance and anti resonance peak frequencies are obtained from the plots and the planar coupling coefficient is computed. The values are given in the table3.2. The difference in resonance and anti-resonance frequencies yields a large planar coupling coefficient K_P that is 0.68 in the (1-x)PMN-(x)KBT ceramic samples for the composition x = 0.35. These values are higher than those provided by lead free potassium bismuth titanate(KBT) and sodium potasiom bismuth titanate(NKBT)based piezoceramics. For instance, according to Weda [47], where the density of the KBT ceramics was 92% of the theoretical one, k_p = 0.136. Hiruma[24] report the piezoelectric properties of hot pressed KBT ceramics (K_{33}=0.28 and d_{33}=69.8pC/N for hot pressed method KBT prepared at 1080^{O}C. Rao and sankaram[26] found that, the piezoelectric constant d_{33} = 56.6pC/N and planar coupling coefficient K_P = 0.59 for KBT ceramic samples and K_p = 0.56, d33 = 0. 72pC/N for NBT-KBT(NBT) systems. But still these values obtained are less than reported values for lead based lead magnesium niobate (PMN) and lead magnesium niobate-lead titanate(PMN-PT), lead zirconate titanate (PZT).for instance, Single crystal of PMN-PT with compositions near the MPB have been reported to display very high piezoelectric coefficients (d_{33} > 2500 pC/N) and very high electromechanical coupling factors (k_{33}>92%), lead zirconate titanate (PZT) based piezoceramics (600 pC/N to 700 pC/N and 0.17%) [48-49]. The 0.65PMN-0.35PT ceramic sintered at 1300°c with density of 7.76g/cm^3 showed d_{33} of 515 pC/N and K_p of 0.56 [50]. Some compositions of PMN-PT single crystals exhibit a very high piezoelectric coefficient (d_{33}~1240 pC/N) and electromechanical coupling coefficients (k_{33}~0.923) [51-52]. Recently, researchers have also reported that PMN-PT single crystals have highest piezoelectric coefficient (d33 ~ 1500 pC/N) and a high electrochemical coupling coefficient (k_{33} ~ 0.82) for <112> grain-oriented PMN–PT ceramics [53].

Table3.2 Electro mechanical coupling and piezoelectric coefficients for NKBT system

composition	F_r(Hz)	F_a(Hz)	K_p	d_{33} pC/N
x=0.15	9956434.34	12925946.10	0.63	58
x=0.25	9685034.38	12748556.80	0.66	67
x=0.35	11590935.13	15878722.04	0.68	79.4

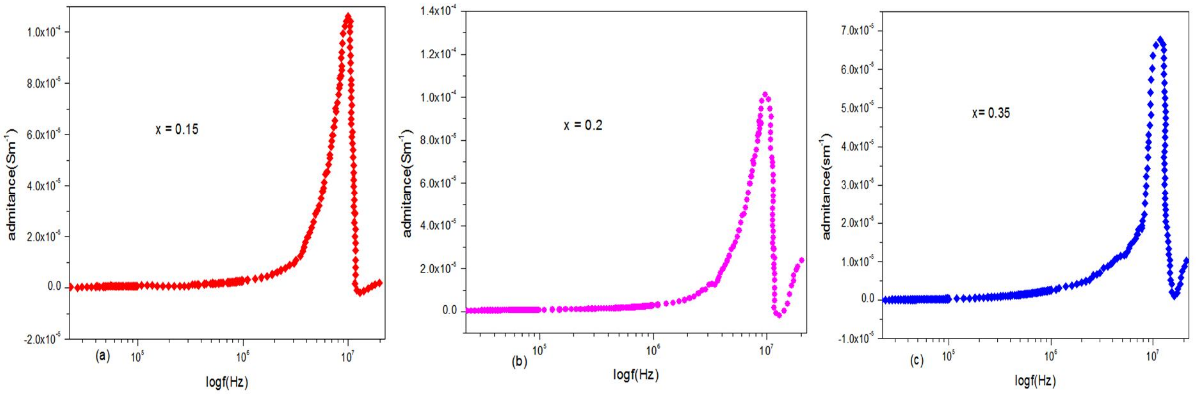

Figure3.3 Resonance and anti-resonance plots for (a) 0.85PMN- 0.15KBT, (b) 0.75PMN-0.25KBT, (c) 0.65PMN-0.35KBT compositions

IV. CONCLUSIONS

Polycrystalline **(**1-x) $PbMg1_{/3}Nb_{2/3}O_3$ - (x) $K_{\frac{1}{2}}Bi_{\frac{1}{2}}TiO_3$ with composition x = 0.15, 0.25 and 0.35 prepared through a high temperature solid state reaction technique was found to have a single phase perovskite type structure. The frequency dependent ac conductivity at different temperatures indicated that the conduction process is thermally activated process. The (I-x)PMN-xPT materials show great promise for sensor and actuator applications. They possess very high electromechanical properties which can be greatly varied by composition and ac bias field. A further understanding of the atomic level structure of PMN-KBT and the Relationship to processing and electrical properties will be of great significance for transducer applications including medical ultrasonic.

V. ACKNOWLEDGMENT

This research has been supported by Wollega University, Ethiopia. The authors also would like to thank prof. G.Prasad, department of physics, osmania University, Hyderabad, India for his help with the property measurements and preparation of samples.

REFERENCES

[1] A.Shukla, R. N. P.Choudhary ,Ferroelectric phase transitionandconductivityanalysisof La_3+/Mn^{4+} modified $PbTiO_3$ nanoceramics, Physica B, 405, 2508–2515,2010.
[2] D. W. Richerson, Modern ceramic engineering: Properties, processing and use in design Second Edition, Marcel Dekker, Inc, 1992.
[3] N. Nikulin , Fundamentals of electrical materials Mir Publishers, Moscow,1988.
[4] A. J. Moulson , J. M. Herbert, Electroceramics Chapman and Hall, London,1990.
[5] D.Segal, Chemical synthesis of advanced ceramic materials.Cambridge University Press,1991.
[6] Y. S.Kim ,N. K.Kim ,J. Ko , Processing and dielectric properties of (Pb,Bi)(Mg,Nb,Ti)O3 ceramics,Ceramics International,1083–1086, 2007.
[7] Z. Li , C.Wang, W. Zhong, J. Appl.Phys.94, 2548, 2003.
[8] G.Smolenskii ,A. Agranovskaya, Dielectric polarization of a number of complexes comounds Sov Phys Solid State 1: 1429, 1959.
[9] G. Smolenskii, V.Isupov & A.Agranovskaya et al., Ferroelectrics with diffuse phase Transitions Sov Phys Solid State 2: 2584, 1961.
[10] M. E.Lines , A .M.Glass, principles and applications of ferroelectrics and related materials Oxford,Clarendon, 1977.
[11] Z.Chen , J.Hu, Piezoelectric and dielectric properties of $(Bi_{0.5}Na_{0.5})_{0.94}$-$Ba_{0.06}TiO_3$–Ba $(Zr_{0.04}Ti_{0.96})O_3$ lead-free piezoelectric ceramics, Ceramics International 35, 111–115, 2009.
[12] Y. C. Liou , J. H. Chen , PMN ceramics produced by a simplified columbite route, Ceramics International 30, 17–22, 2004.
[13] P.Kumar ,C.Prakash ,O. P.Thakur eta l., Dielectric,ferroelectric and pyroelectric properties of PMNT ceramics, Physica B371, 313–316, 2006.
[14] L. E. Cross , Relaxor ferroelectrics: an over view, ferroelectrics, 151,305-320, 1994.
[15] R.Yimnirun , S.Ananta ,R.Tipakontitiku, Perovskite phase transformation in 0.65Pb Mg1/3Nb2/3)O3–0.35PbTiO3 nanoparticles derived by sol-gel, J.Appl.Phys, 6, 307-311, 2006.
[16] R. C. Buchanan, Ceramic materials for electronics 3rd ed., Marcel Dekker inc. NewYork, 2004.
[17] A.Izadpanahi , S.Baghshahi ,A.Shalbaf zadeh, A quick route for the synthesis of PMN-PZT powders. Iranian journal of material science and Enginaring,13(3), 2016.DOI:10.2 2068/ijmse.133.54,Accessed on may 19th 201
[18] H. Ogihara , C. A. Randall, T. M. Susan eta l., Ceram, Soc., 90(1), 110–118, 2009.
[19] D. R. Juberg, Lead and Human Health(2nd ed.), American Council on Science an Health, New York, 2000
[20] N.Jaitanong ,W. C.Vittayakorn , A.Chaipanich , Phase development and dielectric responses in PMN-BNT ceramics, Ceramics International, 36, 1479–1483, 2010.
[21] M. Demaeder , D. Damjanovic & N. Setter , Lead-free piezoelectric Materials, Journal of Electroceramics, 13, 385–392, 2004

[22] M.Dunce , Phase transitions and physical properties of na1/2 bi1/2tio3-based solid solutions(Unpublished PhD thesis),University of Latvia, institute of solid state physics, 2014.
[23] M.Hagiwara, and S.Fujihara, Appl. Phys. Lett.107, 012903, 2015.
[24] Y. Hiruma , Jpn. J.Appl. Phys.,Part I. 44, 5040, 2005.
[25] X. Li , C. Chen , H. Deng, the Growth and Properties of Lead-Free Ferroelectric Single Crystals, Review Paper, crystals , 5, 172-192; 2015. doi:10.339
[26] P. b. Rao ,T. s.sankaram, Studies on A-site modified $A'_{1-x}A_x''BO_3$ based electro ceramics (published PhD thesis), 2009,Osmania University
[27] V. A. ISUPOV , Ferroelectric Na0.5Bi0.5TiO3 and K0.5Bi0.5TiO3 Perovskites and Their Solid Solutions Review paper, 2005, DOI: 10.1080/00150199091027
[28] R.Yimnirun , E.Meechoowas , S. Ananta, Mechanical Properties of PMN-PZT ,Ceramic Systems CMU,3(2), 147, 2004.
[29] S. L. Swartz, T. R.Shrout, Fabrication of perovskite lead magnesium niobate.Mater,1982.
[30] A.K. Roy ,K.Prasad, A.Prasad , Piezoelectric, impedance, electric modulus and AC conductivity studies on $(Bi_{0.5}Na_{0.5})_{0.95}Ba_{0.05}$ TiO_3 ceramic, Processing and Application of Ceramics, 7 (2), 81–91, 2013
[31] N.Sahu,Study of Crystal Structure and Electrical Properties on Lead Titanate and Lead Zirconate Titanate Based Ceramic Oxides (unpublished PhD thesis),2011, department of physics National Institute Of Technology,Rourkela, Odisha-India.
[32] A.Chandran , M. Soosen Samuel , J. Koshy et al., Dielectric relaxation behavior of CdS nanoparticles and nanowires, J. Mater. Sci., 46, 4646–4653, 2011.
[33] A. Tataroğlu , Dielectric Permittivity, AC Conductivity and Electric Modulus Properties of Metal/Ferroelectric/Semiconductor (MFS) Structures, GU J Sci, 26(3), 501-508, 2013.
[34] A. M. Farid, H. E. Atyia, N. A. Hegab , Vacuum,80,284, 2005.
[35] B. Louati, F. Hlel, K. J. Guidara, Alloys Compd.486, 299, 2009.
[36] A. Hegab, M. A. Afifi, H. E. Atyia, , A. S. Farid, J.Alloys Compd.477, 925, 2009.
[37] G. A. Smolenskii, Physical phenomenon in ferroelectrics with diffuse phase transition, J. Phys. Soc. 28, 26-37, 1970.
[38] L.L.Hench,J. K. West Principles of Electronic Ceramics, 1990, John Wiley & Sons, Inc. Sinapore
[39] A. K. Jonscher, Nature (Lond). 254,673, 1977
[40] A. K. Jonscher, Dielectric relaxation in solids.Chelsea Dielectrics,London, 1983
[41] A. K. Roy ,A.Singh ,K.Kumari et al., Electrical properties and ac conductivity of (Bi0.5Na0.5)0.94Ba0.06TiO3 ceramic, ISRN Ceramics, 1–10,2012.
[42] K. Funke ,Jump relaxation in solid electrolytes,Prog. Solid State Chem, 22 (2), 111–195, 1993
[43] D.P.Almond , C. R. Bowen, Anomalous power law dispersions in ac conductivity and permittivity shown to be characteristic of microstructural electrical networks, Phys. Rev. Lett., 92 (15), 5, 2004
[44] G.Bator , Ac and dc conductivity around the ferroelectric phase transition in $(CH_3NH_3)_3Bi2Br_9$ (MABB) crystal, Ferroelectrics, 200, 1–4, 1997
[45] S. R. Elliot, AC conduction in amorphous chalcogenide and pnictide semiconductors, Adv. Phys., 36, 135– 217, 1987 .
[46] A.Pelaiz-Barranco ,M. P.Gutierrez-Amador , A.Huanosta ,R.Valenzuela , Phase transitions in ferromagnetic and ferroelectric ceramics by ac measurements, Appl.Phys.Lett.,73 (14), 2039-3, 1998
[47] T. Weda , A. Fukui and T. Matauo, Japan J.Appl.Phys.41(11B), 7025,2002.
[48] S. E. Park and T. R. Shrout, Ultrahigh strain and piezo- electric behavior in relaxor based ferroelectric single crystals, J. Appl. Phys., vol. 82, no. 4, 1804-1811, 1997.
[49] T.R.Shrout,Z.Chang,P.N.Kim et al.,Dielectric behavior of single crystals near the lead magnesium niobate-lead titanate $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$-$(x)PbTiO_3$) morphotropic phase boundary,Ferroelectrics Lett. Sect., vol. 12, no. 3, 63-69,1990.
[50] M.Ghasemifard , S. M.Hosseini and H. Ghasemifard , Piezoelectric Study of $0.65Pb(Mg_{1/3}Nb_{2/3})O_3 0.35PTO_3$Nanopwoder Ceramics ,Journal of Electronic Science and technology, Vol. 8, No. 1, 2010.
[51] S. E.Park ,T. R.Shrout , Characteristics of relaxor-based piezoelectric single crystals for ultrasonic transducers, IEEE Transactions on Ultrasonics, Ferroelectrics and Frequency Control 44, 1140-1147, 1997.ISSN 0885-3010.
[52] D.Viehland ,Piezoelectric instability in <011>oriented $Pb(B1/3IB2/3II)O_3$-$PbTiO_3$ crystals, Applied Physics Letters 79, 1006-1009, 2001,ISSN 1077-3118.
[53] S. Sun, Fabrication and electrical properties of grain-oriented 0.7Pb(Mg1/3Nb2/3)O3-0.3PbTiO3 ceramics, Applied Physics Letters 84, 574-576, 2004,ISSN1077-3118.