Applied Innovative Research
Vol. 2, March 2020, pp. 18-21

Structural and dielectric properties of 0.75PMN-0.25PT relaxor ferroelectrics with different frequencies at room temperature

Sidharth Kashyap[a]*, S C Bhatt[a], Manish Uniyal[a] & Gambheer Singh Kathait[b]

[a]Department of Physics, Hemvati Nandan Bahuguna Garhwal University, Srinagar Garhwal, 246 174, India
[b]Department of Instrumentation Engineering (USIC), Hemvati Nandan Bahuguna Garhwal University, Srinagar Garhwal, 246 174, India

Received 31 January 2020

In the present work, the composition of 0.75PMN-0.25PT solid solution was synthesized by columbite precursor method with multiple heat treatments at 920°C and 1200 °C. XRD data shows the perovskite structure with minimized pyrochlore phase and intense peaks. It has been observed that the pyrochlore phase can be reduced by increase in sintering temperature. SEM report shows the polygonal, dense and compact grain boundaries. Dielectric study shows the effect of variation in frequency to the dielectric constant and electrical conductivity at room temperature.

Keywords: Pyrochlore phase, Sintering, Grain boundaries, Dielectric constant

1 Introduction

Lead Magnesium Niobate- Lead Titanate is a relaxor ferroelectric material which is widely studied due to its great performance, significance and wide application in various medical devices, sensors, actuators and many other electronic devices like Multi-Layer capacitors. These all important and unique properties of PMN-PT make them attractive for dielectric, piezoelectric and electrostrictive applications. Generally Lead based relaxor ferroelectrics are $Pb(B_1B_2)O_3$ type ceramics, where B_1 is divalent or trivalent metal ions like Mg^{2+}, Zn^{2+}, Fe^{3+} etc. and B_2 is penta valent ions like Nb^{5+}, Ta^{5+}, etc.[1-5]. The well-known example of $Pb(B_1B_2)O_3$ type relaxor ferroelectric material is Lead Magnesium Niobate (PMN) i.e., $Pb(Mg_{1/3}Nb_{2/3})O_3$. It is describedthat electric properties of Lead Magnesium Niobate can be improved by the proper mixing of Lead Titanate, $PbTiO_3$ (PT)[2-10]. The systemLead Magnesium Niobate- Lead Titanate (1-x)PMN-xPT is an solid solution of PbTiO3 and$Pb(Mg_{1/3}Nb_{2/3})O_3$ which shows interesting electrical and piezoelectric properties. At morphotropic phase boundary which is located between X=0.30 to 0.40[11], the compositions shows some extraordinary properties and coexistence of multiple phases like rhombohedral, tetragonal and cubic[2-10,12]which make them applicable for various industrial and scientific areas. From the literature review about relaxor ferroelectrics Kelly *et al.* have reported that the piezoelectric properties can be increased with increase in PT content[2]. Further study about this type of relaxor ferroelectrics shows that relaxor behaviour of PMN increased with increase in PT content up to X=0.25 and decreased as we increase the value of X greater than 0.25. It is also reported that a composition with small amount of Ti content behaves like a relaxor ferroelectrics and as we increase amount of Ti content in composition, the ceramic becomes a normal ferroelectrics[13]. It is an complicated task to prepare single phase (1-x)PMN-(x)PT composition because of volatile nature of PbO at higher temperature. There may be formation of pyrochlore phase during the reaction. Due to pyrochlore phase within the composition, it can be minimize the electrical and piezoelectric properties of ceramics. So excess amount of PbO and MgO were added to sample in this way that stoichiometry of whole compositions should be maintained. The electrical properties of the material is directly depends upon the morphology and density of prepared material. This densification is maintained by preparation process and sintering temperature. Density of PMN-PT can be increased by the heat treatment within the temperature range 1200°C to 1300°C. So we can say the sintering temperature, density, perovskite phase, microstructure and preparation method are the important parameters

*Corresponding author (E-mail: sid.kashyap1@gmail.com)

which can affect the electrical and piezoelectric properties of ceramics.

In the present work, the composition 0.75PMN-0.25PT was synthesized by columbite method with multiple heat treatments at 920°C and 1200°C. The perovskite phase, microstructure and dielectric properties of the prepared sample have been studied and investigated

2 Experimental Details

2.1 Sample preparation

Sample (1-x)PMN-(x)PT for X=0.25 was prepared via columbite precursor method [14] using high purity Lead Oxide (PbO), Titanium Oxide (TiO_2), Magnesium Oxide (MgO) and Niobium Pent-oxide (Nb_2O_5). The starting oxides were dried at 300 °C to remove the moisture and other volatile impurities. All the oxides were weighed and mixed in stoichiometric proportion for X=0.25 composition using agate mortar and pestle in ethanol medium. In the present study, proper amount of Nb_2O_5 and MgO were mixed in ethanol medium for 2 h and then calcined at 1110°C for 5 h. Now the resultant product $MgNb_2O_6$ was again mixed with Lead Oxide and Titanium Oxide in proper amount for 2 h in ethanol medium. The mixed sample was calcined at 920°C for 2 h in closed alumina crucible. The calcined sample was pelletize under uniaxial pressure without adding any binder and then sintered at 1200 °C for 6 h. The silver paste was used to make electrodes on both sides of pellets for electrical measurements.

2.2 XRD analysis

XRD patterns of 0.75PMN-0.25PT sample calcined, sintered at 920°C and 1200°C respectively for 2 h is given in the following figures (Fig.1). It is clear from the comparison of XRD data and figure that prepared sample is in perovskite phase with small amount of pyrochlore peaks. The pyrochlore peaks can be seen due to volatile nature of Lead Oxide at higher temperature and presence of pyrochlore phase make them less useful in various applications because it minimizes the electrical properties. From the XRD analysis it was investigated that pyrochlore phase can be reduced by increase in sintering temperature, which specifies that the dielectric constant and grain growth are function of sintering temperature[15]. Figure 1 shows the XRD pattern of calcined and sintered 0.75PMN-0.25PT sample at 920 °C and 1200°C respectively. The diffraction intensity was measured at different theta values ranging from 20° to 80° at room temperature. The composition 0.75PMN-0.25PT has rhombohedral (pseudocubic) crystal structure at room temperature.

2.3 SEM analysis

The scanning electron microscope was used to study the grain size, crystalline size and surface morphology of the prepared sample. Scanning electron micrographs of 0.75PMN-0.25PT at different scales are shown in the Fig. 2. It is clear from the following micrographs given below; the grains are arranged in compact form with sharp edges and they are polygonal in shape. The grain size of prepared sample is between ~2 μm to ~9.5 μm. The grain size and density of prepared sample are given in the following Table 1.

3 Dielectric Measurements

The FLUKE PM6306 programmable Automatic RCL meter was used for precise measurements of

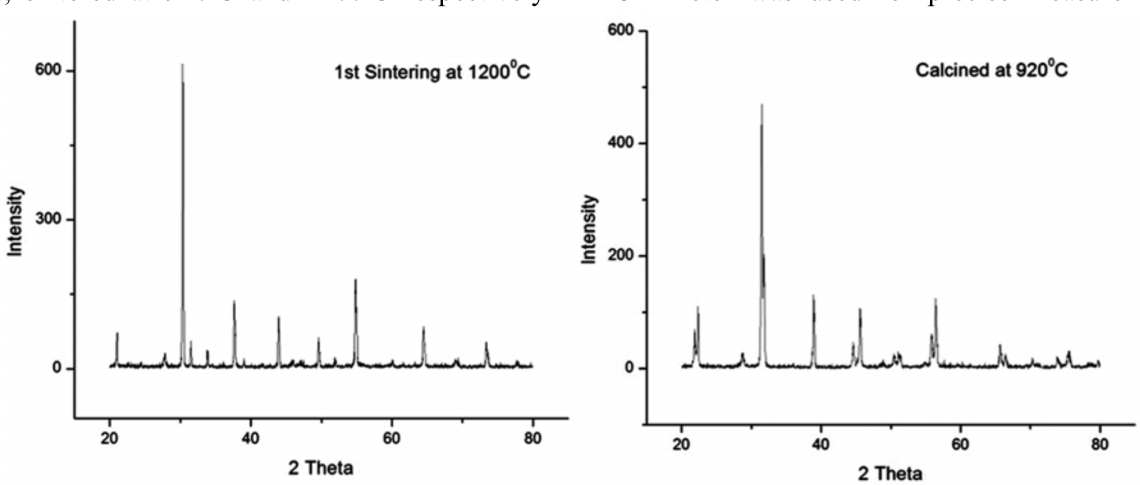

Fig. 1 − XRD patterns of the 0.75PMN-0.25PT sample calcined at 920°C and sintered at 1200°C with reduced pyrochlore phases.

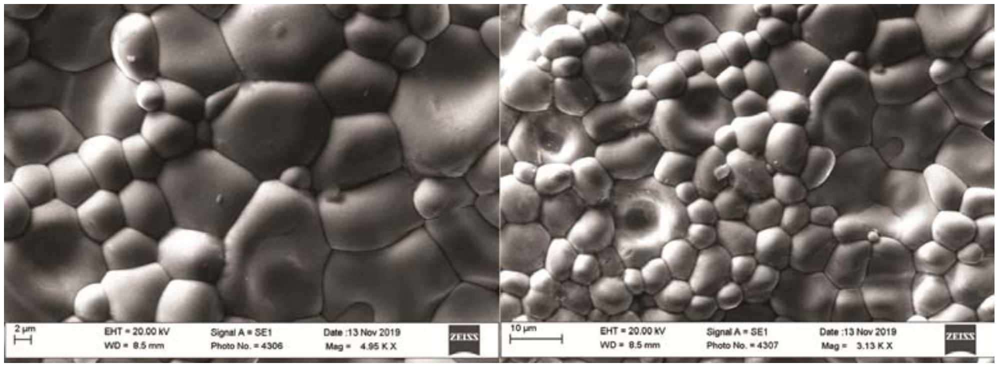

Fig. 2 − SEM Micrographs of 0.75PMN-0.25PT sample at different scale.

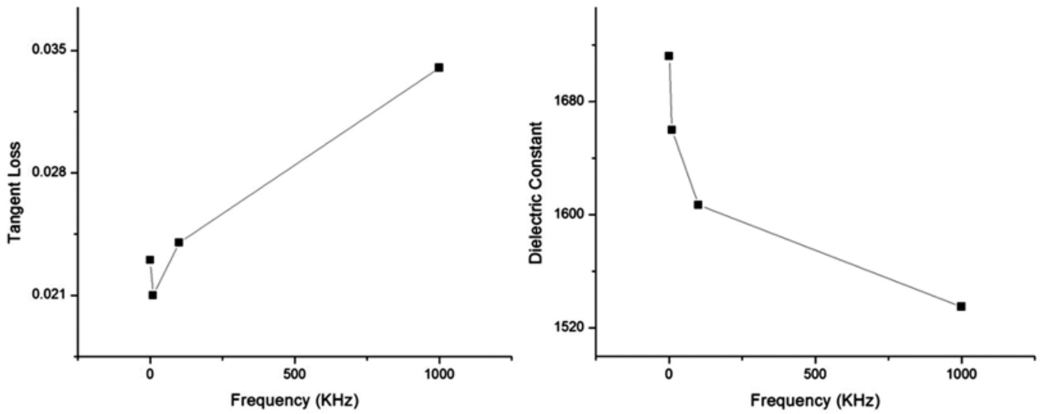

Fig. 3 − Variation of dielectric constant and loss tangent as a function of frequency at room temperature for 0.75PMN-0.25PT sample.

resistance, capacitance and inductance. In the present study, variation of Dielectric Constant, Tangent Loss and Dielectric Conductivity of 0.75PMN-0.25PT at different frequencies ranging from 1 KHz to 1000 KHz has been studied at room temperature. From the dielectric study of given sample, it can be seen that the dielectric constant of 0.75PMN-0.25PT sample gradually decreases with increase in frequency (Fig. 3). The variation in dielectric constant at different frequency may be due to disorder in alignment of dipoles with alternating electric field[15]. The frequency dependence nature of Dielectric Constant and Loss Tangent of composition 0.75PMN-0.25PT have plotted in graphs and also tabulated in the following Table 2.

Table 1 – Sintering temperature, Calcination temperature, grain size and densities.

Sample	Sintering Temp.	Calcination Temp.	Grain Size	Density
0.75PMN-0.25PT	1200°C	920°C	~2.5 µm	9.2 gm/cm^3

Table – 2 Dielectric Constant, Tangent Loss and Electrical Conductivity of 0.75PMN-0.25PT with different frequencies at room temperature.

Sample	Frequency (KHz)	Dielectric Constant (ε_{RT})	Loss Tangent (tan δ)	Electrical Conductivity (ohm-cm)$^{-1}$
0.75PMN-0.25PT	1	1712	0.023	2.19×10^{-8}
	10	1660	0.021	1.93×10^{-7}
	100	1607	0.024	2.14×10^{-6}
	1000	1535	0.034	2.89×10^{-5}

4 Results and Discussion

The single phase 0.75PMN-0.25PT was prepared via columbite precursor method with multiple heat treatments. It is clear from the XRD analysis, the sample is in perovskite phase with reduced pyrochlore phase. The pyrochlore phase can be reduced by increase in sintering temperature. The SEM graphs show the dense and compact boundaries. The dielectric constant of 0.75PMN-0.25PT shows the frequency dependent nature at room temperature. As we increase frequency 1 KHz to 1MHz, the Dielectric Constant and Tangent Loss will also decrease due to disorder in alignment of dipoles with alternating electric field. It is also investigated that the dielectric constant is directly correlated with density of grains and sintering temperature.

Acknowledgements

Authors are thankful to Dept. of Instrumentation (USIC), H N B Garhwal University, Srinagar Garhwal for availing electrical measurements and characterization facility. Authors are also thankful to all the research scholars, staff of physics department for their valuable help.

References

1 Singh S P, Singh A K, Pandey D, Sharma H & Prakash O, *J Mater Res*, 18 (2003) 2677.
2 Kelly J, Leonard M, Tantigate C & Safar A, *J Am Ceram Soc*, 80 (1997) 957.
3 Singh K & Band S, *Bull Mater Sci*, 20 (1997) 265.
4 Suh D, Lee D & Kim N, *J Eur Ceram Soc*, 22 (2002) 219.
5 Park J, Kim Y & Park S, *J Korean Phys*, 32 (1998) 967.
6 Feng Z, Zhao X & Luo H, *J Appl Phys*, 100 (2006) 24104.
7 Katayama K, Abe M, Akiba T & Yanagida H, *J Eur Ceram Soc*, 5 (1989) 183.
8 Alguero M, Alemany C, Jimenez B, Holc J, Kosec M & Pardo L, *J Eur Ceram Soc*, 24 (2004) 937.
9 Choi S, Shrout R, Jang S, Bhalla A, *Ferroelectrics*, 100 (1989) 29.
10 Ktunjak Z, Petzelt J & Blinc R, *Nature*, 441 (2006) 956.
11 Benayad A, Sebald G, Guiffard B, Lebrun L, Guyomar D & Pleska E, *J Phys IV Proc*, 126 (2005) 53.
12 Noblanc O, Gaucher P & Calvarin G, *J Appl Phys*, 79 (1996) 4291.
13 Huang J, Chateen N D & Fitzgerald J J, *Chem Mater*, 10 (1998) 3848.
14 Swartz S L & Shrout T R, *Mater Res Bull*, 17 (1982) 1245.
15 Tarale A N, Premkumar S & Reddy V R, *Ferroelectrics*, 516 (2017) 8.