E-ISSN: 2278–179X

JECET; September 2013 – November 2013; Vol.2.No.4, 1401-1406.

Journal of Environmental Science, Computer Science and Engineering & Technology

An International Peer Review E-3 Journal of Sciences and Technology

Available online at www.jecet.org

Engineerig & Technology

Research Article

Structural and Dielectric Behaviour of 0.77 $Ba(Fe_{0.5}Nb_{0.5})O_3$ -0.23$BaTiO_3$ Ceramics

Avinash Kumar[1], Pritam Kumar[2] and N. K. Singh[1,*]

[1]Department of Physics, V. K. S. University, Ara 802301 Bihar, India

[2]Department of Engineering Physics, R.I.T., Indore, (M. P.) India

Received: 2 November 2013**; Revised:** 25 November 2013; **Accepted:** 30 November 2013

Abstract: Complex perovskite oxides are widely used in various solid -state devices e.g., optical devices, multilayer capacitors, transducers, actuators, sensors etc. High dielectric constant materials such as barium iron niobate nowadays become more and more important in ceramics materials. However, Pb-based compound are somewhat toxic and therefore, as a substitution for more environment friendly materials in application, attention is being increasing focused on the preparation of Pb-free high–performance piezoelectric materials. Perovskite oxides of 0.77Ba $(Fe_{0.5}Nb_{0.5})O_3$-0.23$BaTiO_3$, (BFN-BT) was synthesized by the tradition solid-state reaction method and performed their preliminary X-ray diffraction (XRD) and Scanning electron micrograph (SEM) analysis and the temperature and frequency dependence of dielectric measurements of the BFN-BT ceramics. The XRD patterns of the BFN-BT at room temperature show a monoclinic phase. Above compounds are useful for high voltage capacitors and other applications, such as multilayer capacitors, sensors, detectors and actuators. Detailed studies of dielectric and electrical properties of the materials in a wide range of frequency (100Hz–1MHz) and temperatures (30-230°C) showed that these properties are strongly temperature and frequency dependent.

Keywords: Perovskite oxides; dielectric constant, X-ray diffraction, scanning electron microscope

INTRODUCTION

For the development of capacitors, many investigators have tried to synthesize and study the materials that exhibit high dielectric constant[1-3]. However, most high dielectric materials contain lead that inevitably causes environmental problems due to toxicity of lead oxide. Therefore it is necessary and urgent to search for lead free materials with excellent dielectric properties, which should be comparable with those found in the lead based ceramics. Recently a large number of lead-free electroceramics such as ($Na_{½}K_{½}NbO_3$, $Bi_{½}Na_{½}TiO_3$) and their solid solution with $BaTiO_3$ have been developed[4, 5]. Sodium bismuth titanate, $Bi_{½}Na_{½}TiO_3$ (BNT) discovered by Smolenskii et al.[6] in 1961, is considered to be an excellent candidate of lead-free piezoelectric ceramics with a large remnant polarization. This material has indeed high dielectric constant and electromechanical coupling co-efficient which can be tailored by suitable substitutions at the A and/or B sites of many perovskite compounds of general formula ABO_3 type (A = mono or divalent ions, B = tri or pentavalent ions) of the compounds[7, 8.] Interestingly, most of the perovskite oxides which are of the greatest technological interests are not simple system, but complex oxides with two different kinds of B atoms, such as $A(B'B'')O_3$. From a chemical point of view, two classes of compounds $A(B'B'')O_3$ can be distinguished: homovalent and hetrovalent compounds. In homovalent compounds, the two B-atoms belong to the same column of the periodic table and hence the $A(B_x'B_{1-x}'')O_3$ compounds can thus have a composition x continuously ranging from 0 to 1. A typical example of such homovalent compound is $Pb(Zr_x'Ti_{1-x}'')O_3$ [9, 10]. On the other hand, there is a unique stoichiometry in hetrovalent compounds since the two B-atoms do not belong to same column of the periodic table, Typical examples of hetrovalent compounds are $A(Fe_{0.5}Nb_{0.5})O_3$, where A = Ba, Sr and $BaAlNb_{0.5}O_3$. Complex perovskite oxides of type $A(B'B'')O_3$, are particularly attractive for various applications, such as microwave frequency resonators, multilayer capacitors, sensors, detectors and actuators[11]. Alternating current (AC) impedance spectroscopy is an appropriate method to study (i) the properties of the intragranular and interfacial regions and their interrelations, (ii) their temperature and frequency dependent phenomena in order to separate the individual contributions from the total impedance and (iii) their interfaces with electronically conducting electrodes[12–14]. $BaTiO_3$ (BT) with a perovskite structure is a strong dielectric material, which has far reaching applications in the electronics industry for transducers, actuators and high-k dielectrics, and is mostly used to make multilayer ceramic capacitor (MLCC) materials[15-17].In recent years, many workers have studied $BaFe_{0.5}Nb_{0.5}O_3$ (BFN) of certain compositions. They have reported that the BFN-based electroceramics exhibit a relaxor behavior by showing very attractive dielectric and electric properties but, however, there still exist considerable debates concerning the physical mechanisms governing their electrical behavior. Much attention has been given to diffuse phase transitions (DPT's) from the points of view of fundamental interest and of practical importance in the development of ferroelectric materials for, e.g., capacitor applications. Complex perovskite-type ferroelectrics with disordered cation arrangements show DPT's (commonly known as relaxor ferroelectrics) which are characterized by a broad maximum for the temperature dependence of the dielectric permittivity and the dielectric dispersion in the transition region[18]. To our best knowledge, structural and dielectric studies of titled perovskite oxides have not been reported so far. This has prompted us to examine the Structural and dielectric of above cited sample.

EXPERIMENTAL DETAILS

Powders of $BaCO_3$, (reagent grade), TiO_2, Fe_2O_3 and Nb_2O_5 (reagent grade) were taken in stoichiometric ratio, and mixed in the presence of acetone for 5 h. The mixture was calcined at 1200ºC in air for 6 h and pellets of sample were sintered at 1250ºC for 8 h. From the measurement, we have obtained dielectric data

of the samples as a function of frequency at different temperature (30°C–280°C) using a PSM (Comm.1735). The X-ray powder diffraction pattern of the sample is taken at room temperature using a X-ray powder diffractometer (Rigaku Miniflex, Japan) using Cuk_{α} radiation (λ = 1.5418 Å) in a wide range of Bragg angles 2θ (20° ≤ 2θ ≤ 80°) with scanning rate 2°/min. The micrographs are recorded using scanning electron microscopy JEOL-JSM-5800 to study the surface morphology/microstructure of the sintered pellets.

RESULTS & DISCUSSIONS

The X-ray diffraction (XRD) pattern of the sample taken at room temperature is shown in Fig. 1. All the peaks were indexed and the cell parameters were determined in different crystal systems by using a standard computer program POWDMULT [19]. The compound has monoclinic structure at room temperature.

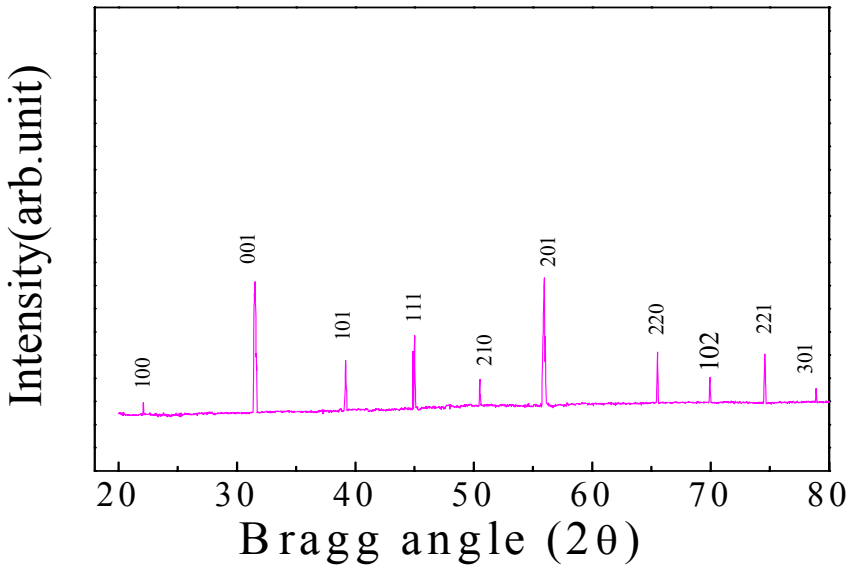

Fig.1:X-ray diffraction patterns of BFN-BT ceramics, at room temperature.

The microstructure of BFN–BT ceramics was investigated by scanning electron microscope (SEM). The surface micrograph of BFN-BT is shown in Fig. 2. The grain size of the pellet sample was found to be in the range of 1.86 µm. When the sintering process performed at 1250°C for 5 h, the movement of atoms or molecules is driven by differences in curvature between the particles in contact. In order to reduce surface free energy, atoms supposedly move from particles with smaller radius to those with larger radius. Particularly, for above mentioned samples it is possible that the matter transport between several aggregated particles and the high anisotropy in the grain boundary energies induced the formation of compact and irregular particles.

The frequency dependence of the dielectric constant (□') and loss tangent (tanδ) of BFN-BT ceramic at different temperatures is plotted in Fig. 3. Dielectric characteristics of ceramic materials are of increasing importance due to their various applications in the field of solid-state electronics. The main applications of ceramic dielectrics are as capacitive elements in electronic circuits and as electrical insulator. The dielectric constant, loss tangent and dielectric strength are the important characteristics of the dielectrics relevant to their suitability for application purpose.

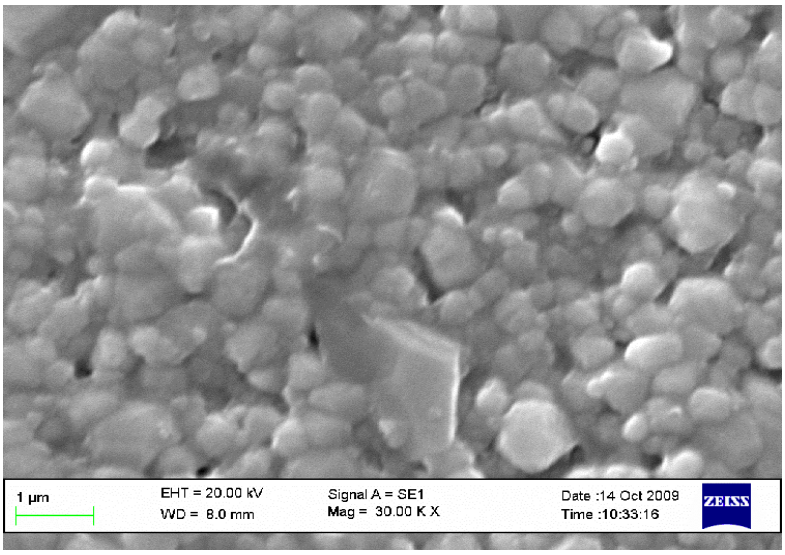

Fig.2: SEM of BFN-BT, ceramics at room temperature.

The nature of dielectric permittivity related to free dipoles oscillating in an alternating field may be described in the following way. At very low frequencies ($\omega<<1/\tau$, τ is the relaxation time), dipoles follow the field and $\varepsilon' \approx \varepsilon_s$ (value of the dielectric constant at quasi-static field). As the frequency increases (with $\omega<1/\tau$), dipoles begin to lag behind the field and ε' slightly decreases. This decrease is due to the release/clamping of space charge or space charge relaxation which is supposed to be dominated at lower frequencies. When frequency reaches the characteristic frequency ($\omega =1/\tau$), the dielectric constant drops (relaxation process). At very high frequencies ($\omega>>1/\tau$), dipoles can no longer follow the field and $\varepsilon' \approx \varepsilon_\infty$ (high-frequency value of ε'). Due to this space charge relaxation, relaxation mechanisms namely; orientation, ionic and electronic relaxation/ polarization are observed at higher frequency in **Fig. 3.** It is observed that with increase of frequency, generally tanδ decreases, exhibiting a typical characteristic of a normal dielectric.

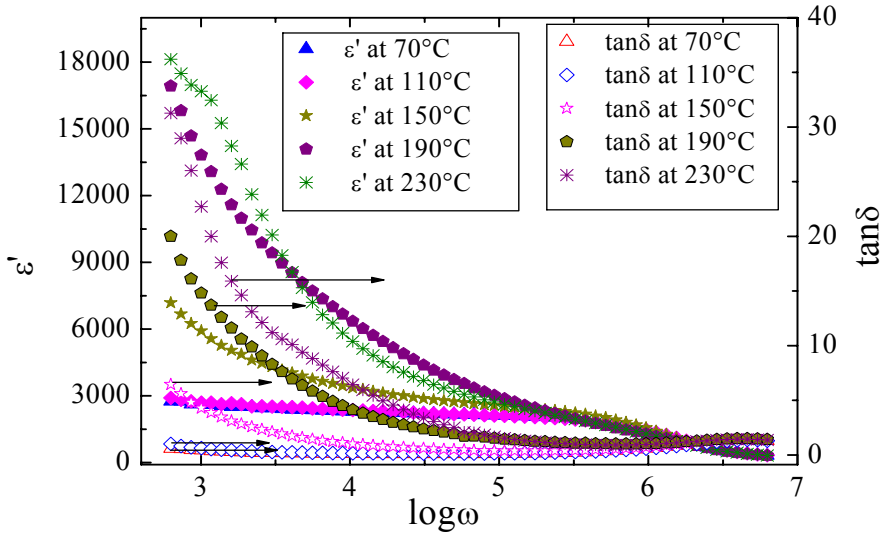

Fig. 3: frequency dependence of dielectric constant (ε') and tanδ of BFN-BT ceramic at different temperatures.

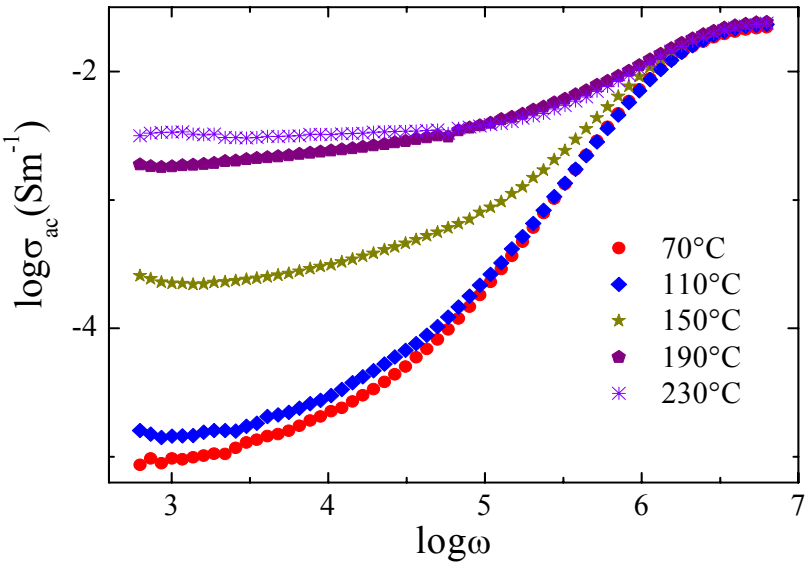

Fig.4: Frequency dependence of ac conductivity of BFN-BT ceramic at different temperatures.

Logarithmic angular frequency dependence of σ_{ac} of BFN-BT samples at various temperatures is shown in Fig. 4. Electrical conduction in dielectrics is due to the ordered motion of loosely bound charge particles due to the influence of an electric field. If we assumes that all dielectric loss in the temperature range studied are due to conductivity, the conductivity can be expressed as $\sigma(\omega) = \omega\varepsilon_0\varepsilon''$, here σ is the real part of the conductivity and ε'' is the imaginary part of complex dielectric permittivity (ε^*).For given samples the conductivity shows dispersion which shifts to higher frequency side with the increase of temperature. As the temperature rises, the conductivity spectrum shows low frequency plateau followed by high-frequency dispersion. The high-frequency conductivity dispersion may be attributed to ac conductivity whereas the frequency independent plateau region of the conductivity pattern corresponds to the dc conductivity (σ_{dc}) of the material.

CONCLUSIONS

0.77Ba $(Fe_{0.5}Nb_{0.5})O_3$-0.23$BaTiO_3$, (BFN-BT) ceramic is synthesized by solid state-reaction technique. Detailed studies of structural and dielectric properties of the sample in a wide range of frequency (100Hz–1MHz) and temperatures (30-230°C) showed that these properties are strongly temperature and frequency dependent.

ACKNOWLEDGEMENT

The authors are grateful to UGC, for providing financial assistance for this research work.

REFERENCES

1. C.C. Homes, T. Vogt, S. M. Shaprio, S. Wakimoto, M. A. Subramanian, A. P. Ramirez, *Phys. Rev. B*,2003, **67,** 092106.
2. M. A. Subramanian, D. Li, N. Duan, B. A. Reisner, A. W. Sleight, *J. Solid State Chem*.,2000, **151**, 323.

3. S. Saha, T. P. Sinha*, Phys. Rev. B.,* 2002, **65,** 134103.
4. S. Priya, A. Ando, Y. Sahabe, *J. Appl. Phys*. 2003,**94**, 1171.
5. A. Herabut, A. Sefari, *J. Am. Ceram. Soc,* 1997, **80,** 2954.
6. G. A. Smolenskii, V. A. Isupov, R. I. Agranovskaya, N. N. Kainik, *Sov. Phys.Sol. State*, 1961, **2**, 2651.
7. T. R. Shrout, A. Italliyal, *Am. Ceram. Soc. Bull.,* 1987, **66,**704.
8. D. Veiffland, N. Kim. Z. Xu. D. A. Payne. *Am. Ceram. Soc*.1995, **78**, 2481.
9. Tanaka, T.; *Ferroelectrics* 1982, **40**, 167.
10. Uchino, K.; Piezoelectric Actuators and Ultrasonic Motors, Kluwer Academic, Boston, 1996.
11. U. Intatha, S.Eitssayeam, J.Wang, T. Tunkasiri, *Curr. Appl. Phys*., 2010, **10,** 21.
12. J. R. Macdonald, Impedance Spectroscopy Emphasizing Solid Materials and Systems, John Wiley and Sons, Inc., New York, 1987.
13. D. C. Sinclair, A.R.West, *J. Appl. Phys*., 1989, **66,** 3850.
14. A. K. Jonscher, *Phys. Stat. Sol.(a),* 1975**, 32**, 665.
15. Y.B. Mao, S. Banerjee, S.S. Wong, *J. Am. Chem. Soc*.2003, **125**, 15718.
16. B. Cui, P.F. Yu, J. Tian, Z.G. Chang, *Mat. Sci. Eng. B,* 2006, **133**, 205.
17. Pengfei Yu, Xue Wang and Bin Cui, *Scripta Materialia,* 2007, **57**, 623.
18. F. Roulland, R. Terras, G. Allainmat, M. Pollet, S. Marinel, *J. Eur. Ceram. Soc*. 2004, **24,** 1019.
19. E. Wul, PowdMult, An interactive powder diffraction data interpretation and index program, version 2.1, school of Physical science, Flinders University of South Australia, Bedford Park, S.A. 5042, Australia

***Corresponding Author: N. K. Singh;** University Department of Physics, V. K. S. University, Ara 802301 Bihar, India

.