Synthesis and characterization of barium niobate and silver niobate solid solution

Cite as: AIP Conference Proceedings **2220**, 020146 (2020); https://doi.org/10.1063/5.0001743
Published Online: 05 May 2020

Isha S. Kapte, Sucharit Niyogi, Parvez J. Qureshi, C. Pavithra, W. Madhuri, and D. Zarena

Synthesis and characterization of barium niobate and silver niobate solid solution

Isha S. Kapte[1], Sucharit Niyogi[1], Parvez J. Qureshi[1], C. Pavithra[1], W. Madhuri [1, a)], D. Zarena [2, b)]

[1]*Department of Physics, SAS, VIT University, Vellore-14, TN, India*
[2]*Department of Physics, JNTUA College of Engineering, Anantapur -515002, AP, India.*

Corresponding Email: [a)]madhuriw12@gmail.com, [b)]zareenajntua@gmail.com

Abstract. Silver niobate and barium niobate solid solution ferroelectric material with the general formula AgNbO3+BaNb2O6 (ABN) is synthesized by conventional ceramic route. X-Ray diffraction studies of ABN revealed AgNbO3 and BaNb2O6 phases distinctly. The solid solution is characterized for ferroelectric and dielectric properties. After poling, the sample is scanned for P-E loop. The solid solution has resulted in a dielectric constant of 162 at room temperature when frequency scanned in the range of 100 Hz to 5 MHz is studied.

Keywords: XRD, Silver Niobate, P-E loop, Dielectric constant

INTRODUCTION

Lead zirconium titanate (PZT) has ruled the electronic industry as the best piezoelectric material with perovskite structure for over half a century. Piezo electrics have a wide range of applications as sensors, actuators, transmitters, transformers and resonators [1]. However recently, the environmental toxicity and hazardous nature of lead has resulted in development of lead free piezoelectric materials. Silver niobate is presently being investigated due to its high local polarization. Theoretical and experimental calculation of Ag-O bond lengths in AgO12 coordination has revealed a short bond (~ 2.44 Å) and a long bond (~3.10 Å). The short bond length is approximately 0.3 Å shorter than the sum of the ionic radii Ag+(1.28 Å) and O2-(1.40 Å) leading to large displacement of Ag in O-Ag-O bond. The off center position of Ag has resulted in a local polarization of as large as 52µC/cm2 in AgNbO3 [2]. On the other hand, Sr substituted Ba niobate is extensively studied for its applications as pyro electric, photo refractive and electro-optic devices [3-5]. However, little attention is paid to BaNb2O6 bulk ceramics and most of the literature is focused on it phase transformations. Barium niobate is receiving great attention for being tungsten bronze structure ferroelectric material [3]. In the present work, in order to modify the dielectric and piezoelectric properties of silver niobate a solid solution of AgNbO3 and BaNb2O6 at a proportion 75:25 is synthesized by conventional ceramic route.

EXPRIMENTAL

Synthesis technique

Reagent grade Ag2O, BaCO3 and Nb2O5 are weighed in the stoichiometric proportion of (1-x) AgNbO3+xBaNbO3 (ABN) with x = 0.25. The powders are well mixed and milled in Fritsche ball mill at 250 rpm for 3 hrs. The powders are made into thick cakes and are calcined at 1000oC for 30min. The cakes are again grinded

3rd International Conference on Condensed Matter and Applied Physics (ICC-2019)
AIP Conf. Proc. 2220, 020146-1–020146-5; https://doi.org/10.1063/5.0001743
Published by AIP Publishing. 978-0-7354-1976-6/$30.00

and milled for 6 hrs at 250 rpm. Thus obtained green powders are made into 1 cm diameter pellets and are sintered at 1150oC for 10 min. The pellet is painted with silver paste for good ohmic contacts. Thus obtained pellet is used for dielectric and ferroelectric characterization.

Characterization technique

The powder X-Ray diffraction pattern of prepared ABN is recorded using an XRD system (Bruker D8 Advance) and Cu Kα radiation ($\lambda = 1.5405$ Å). Dielectric measurements are performed at room temperature by using LCR meter (LCR HiTESTER 3532-50 HIOKI) which is computer interfaced. The pellet is polled to align the dipoles of the piezoelectric material at 1kV for 30 min. in silicon oil bath. The ferroelectric P-E loop is measured by Marine India instrument. The piezoelectric coefficient d33 is measured by sinocerapiezotronics (YE2730A).

RESULTS AND DISCUSSION

X-Ray diffraction analysis

Fig.1 shows powder X-ray diffraction (XRD) profile of the synthesized ABN powder sample. The data is plotted as a function of diffraction angle to X-ray intensity at different 2θ angles from 20o to 90o. In Fig.1 sharp peaks indicate the crystalline nature of material. In XRD data of ABN confirms the presence of two phases viz BaNb2O6 and AgNbO3 which are confirmed with PCPDFWIN PDF-742201 and PCPDFWIN PDF- 520405 respectively. The crystalline structure of the material found to be rhombohedral and the space group is (Pbmm). From the XRD it is deduced that the two phases are co-existing and are immiscible.

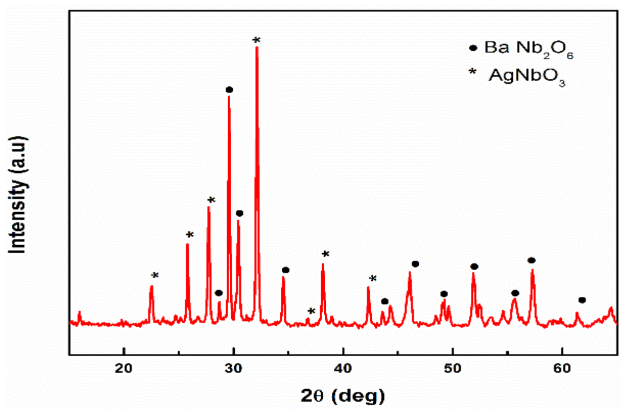
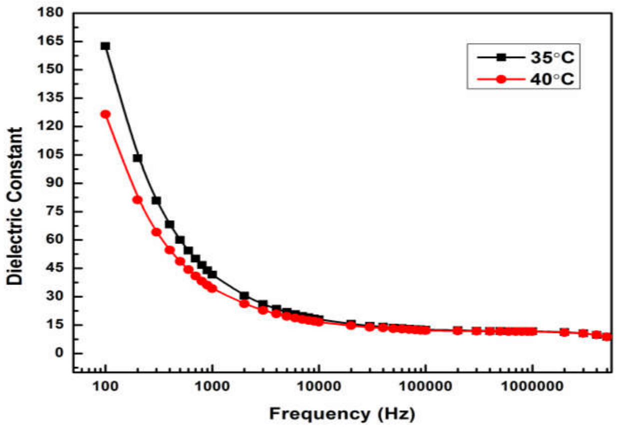


FIGURE 1: X-ray diffraction pattern of ABN ceramic sintered at 1150 °C.

Dielectric behavior of the sample

Fig.2 shows the frequency dependence of dielectric constant (Ɛr) at room temperature of ABN which is calculated using relation (1). Near room temperature(35°C) dielectric constant is found to be 162.50 at 100Hz. Low dielectric constant is attributed to immiscibility of the ABN solid solution. The value of dielectric constant decreases with the increase in frequency, which is a normal behavior of dielectric or ferroelectric materials.



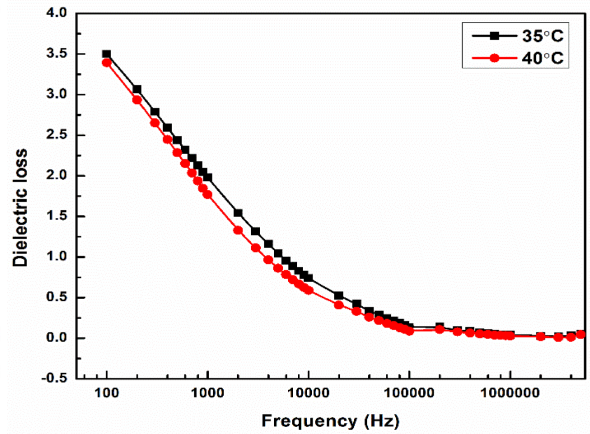


FIGURE 2: Frequency variation of dielectric constant

FIGURE 3: Frequency variation of dielectric loss

High dielectric constant at low frequencies is because of space charge and dipolar polarisation contributions. With increase in frequency, dielectric constant decreases gradually due to dipolar and other relaxation process.

$$\varepsilon_r = \frac{C \times t}{\varepsilon_{0} \times A} \qquad \text{.........(1)}$$

Fig.3 shows the frequency dependence of dielectric loss at room temperature of ABN sample and it is found to be 3.49. High dielectric loss is a representation of immiscibility of the ABN solid solution. At room temperature silver niobate is strongly anti-ferroelectric and a weak ferroelectric [2, 6] which seems to dominate the polarization process resulting in high dielectric loss and low dielectric constant. Another important point to be noticed here is that silver and barium niobate individually possess a dielectric constant of the order of 350 and 800 [6, 7] . But in the solid solution ABN the dielectric constant is a meager 160 indicating strong dipolar interaction of the two phases resulting in cancellation of net polarization in the solid solution.

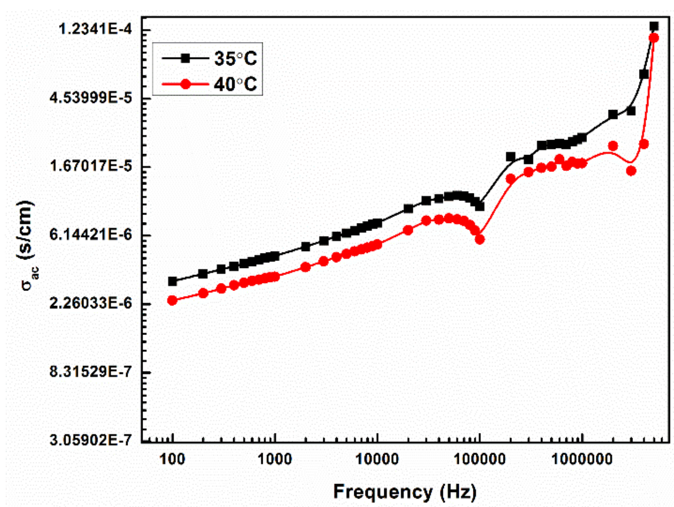


FIGURE 4: Frequency variation of ac conductivity at room temperature.

AC electrical conductivity as a function of frequency is depicted in Fig.4 at two different temperatures (35oC and 40oC). AC conductivity is found to be increasing with frequency linearly. Such a behavior of AC conductivity supports (small) polaron conduction. However, the slope change observed around 100 KHz and 3 MHz suggests change or existence of two different conduction mechanisms. Further analysis of AC conductivity will confirm the exact transport mechanism. Applied frequency is expressed as a power law relation (2) where, A and n are arbitrary constants depending on the composition and temperature.

$$\sigma_{ac}(\omega) = A\omega^{n} \quad \ldots\ldots\ldots\ldots (2)$$

Ferroelectric and piezoelectric studies

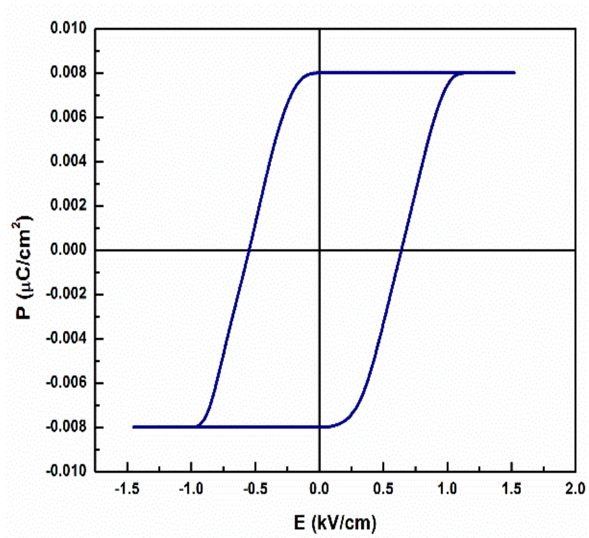


FIGURE 5: P-E loop for ABN ceramic at room temperature.

Fig.5 shows the polarization vs electric field (P–E) hysteresis loop for ABN sample at room temperature. The capacitor and resistor values are fixed at 1nF and 1 MΩ. It is noticed that ABN solid solution exhibits well defined square hysteresis behaviour of polarization response to applied electric fields. The coercivity Ec and remnance Pr are found to be 0.6 kV/cm and 8 nC/cm2. Silver niobate is reported to be weak ferroelectric and strong anti-ferroelectric at room temperature [2, 6]. Barium niobate synthesized via solid state reaction route is reported to possess 0.5 µC/cm2 and 12kV/cm Pr and Ec [7] . Further lower concentration of BaNb2O6 in the solid solution could not enhance the Pr and Ec of the solid solution ABN. This result also explains high dielectric loss and low dielectric constant of the solid solution at room temperature. The piezoelectric coefficient d_{33} is used to understand the basic mechanism of the external electric field. The value of piezoelectric coefficient d_{33} is measured as 104 pC/N. The piezoelectric voltage susceptibility (G_{33}) is calculated using the relation (3) and it is found to be 0.64 pm/V.

$$G_{33} = d_{33} / \varepsilon_{33} \quad \ldots\ldots(3)$$

where, d_{33} is the piezoelectric coefficient and ε_{33} is the dielectric constant at room temperature.

CONCLUSION

The silver and barium niobate solid solution is synthesized by conventional ceramic route followed by microwave double sintering. X-ray diffraction studies confirm the two phases and rhombohedral structure with Pbmm space group of the solid solution. Immiscibility and strong dipolar interaction between silver niobate and barium niobate is supposed to be the reason for low dielectric and ferroelectric properties.

ACKNOWLEDGEMENTS

The authors would like to thank SAS, VIT University for XRD facility. Special thanks to Dr. Kalainathan, Director, CCG, VIT University, Vellore, for ferroelectric and dielectric studies.

REFERENCES

[1] Y. Pu, P. Gao, T. Wu, X. Liu, and Z. Dong, J. Electron. Mater. **44**, 332–340 (2015).
[2] Fu, D., Endo, M., Taniguchi, H., Taniyama, T., Koshihara, S. Y., & Itoh, M. Applied Physics Letters, **92(17)**, 172905 (2008).
[3] R. Muduli, R. Pattanayak, S. Raut, P. Sahu, V. Senthil, S. Rath, P. Kumar, S. Panigrahi, and R. K. Panda, J. Alloys Compd. **664**, 715–725 (2016).
[4] R. Muduli, P. Kumar, R. K. Panda, and S. Panigrahi, . Materials Chemistry and Physics. **180**, 422-431 (2016).
[5] D. W. Kim, K. S. Hong, C. H. Kim, and K. Char, Appl. Phys. a-Materials Sci. Process. **79(3)**, 677–680 (2004).
[6] D. Fu, M. Endo, H. Taniguchi, T. Taniyama, M. Itoh, and S. Koshihara, J. Phys. Condens. Matter. **23(7),** 75901 (2011).
[7] S. R. Dhage, R. Pasricha, and V. Ravi, Mater. Lett. **59(14–15)**, 1929–1931 (2005).