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# Microstructural Studies of $AgNbO_3$ Ceramic by Using Complex Impedance Spectroscopy

K. Gangaprasad, T. Durga Rao, Manish K. Niranjan, and Saket Asthana*

*Advanced Functional Materials Laboratory, Department of Physics,*
*Indian Institute of Technology Hyderabad-502205, India*
**E-mail: asthanas@iith.ac.in*

**Abstract.** Lead-free piezoelectric silver niobate ceramic was synthesized by conventional solid state route. Room temperature X-ray diffraction pattern revealed that the sample crystallizes in single phase orthorhombic perovskite structure. Scanning electron micrographs of $AgNbO_3$ ceramic showed that the average grain size is in the range 2-3 μm. The electrical properties were investigated by using impedance spectroscopy. Appearance of single semicircular arc in the Nyquist plot indicated the presence of grain contribution in the sample. Single *RC* parallel circuit model was employed to extract bulk capacitance ($C_b$), resistance ($R_b$) and electrical conductivity ($\sigma_b$). The activation energy calculated from impedance and modulus data indicate that same types of charge carriers (oxygen vacancy movements) are responsible for conduction and relaxation.

**Keywords:** $AgNbO_3$ ceramics, impedance, activation energy.
**PACS: 72.20.-i**

## INTRODUCTION

The perovskite $AgNbO_3$ (AN) is one of the most promising ferroelectric material for both fundamental and applied research. Silver niobate and its solid solutions with other compounds have been found very promising materials for lead free piezoelectric with large electromechanical response[1]. For environmental concern, there is growing interest in the development of high performance lead free piezoelectrics[2].The AN is a ferroelectric at room temperature and it has small spontaneous polarization (~0.041μC/cm$^2$) [3] and it can be switched to large polarization (52 μC/cm$^2$)[4] at room temperature under the application of electric field 220 kV/cm which could be suggested due to the strong displacement of $Ag^+$ in the perovskite structure. Such amazing ability of polarization is of great significance for the design of lead free piezoelectric materials. There are many reports focused on structural, dielectric properties of $AgNbO_3$ [5-8], however, the electrical studies on this compounds are very few. In order to study the electrical properties, we have extensively carried out impedance studies using complex impedance spectroscopy (CIS).

## EXPERIMENTAL DETAILS

$AgNbO_3$ polycrystalline ceramics were prepared by solid state reaction method under controlled $O_2$ atmosphere using high purity chemicals. The powders were calcined at 900$^o$C for 6 hours and finally sintered at 1050 $^o$C for 6 hours in $O_2$ atmosphere. The oxygen atmosphere can prevent the decomposition of silver oxide to silver at high temperatures. The x-ray diffraction technique was used to determine the structure and lattice parameters. The microstructural studies were performed using field emission scanning electron microscopy (Carl Zeiss, Supra40 Germany). Dielectric measurements were carried out using Wayne Kerr 6500B impedance analyzer with temperature and frequency variation.

## RESULTS AND DISCUSSIONS

### Structural Studies

Figure. 1 shows the X-ray diffraction (XRD) pattern for $AgNbO_3$ at room temperature. It crystallized in orthorhombic structure and indexed with *Pbcm* (JCPDS card no: 52-405). Inset of Figure. 1 depicts the FE-SEM micrograph of the $AgNbO_3$ ceramic. Uniform

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grain growth has been observed with small degree of porosity. The average grain size of the compound is found to be ~2-3 μm.

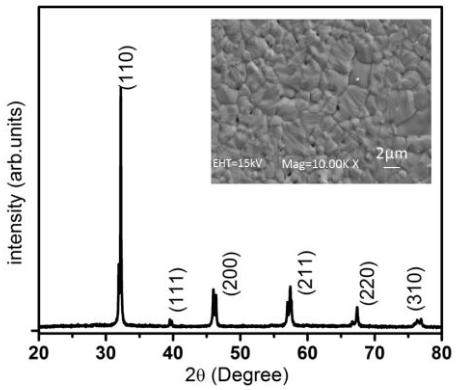


**FIGURE 1**. X-ray diffraction pattern of $AgNbO_3$ at room temperature. Inset shows SEM micrograph of $AgNbO_3$ ceramics.

## Impedance Studies

The complex impedance spectroscopy (CIS) is a technique to analyze the electrical properties of a polycrystalline sample in a wide range of frequencies and temperatures. The variation of real versus imaginary part of the impedance at different temperatures (633K-673K) is shown in Figure. 2 (Nyquist plot). The depressed semicircular indicates that the relaxations are of non-Debye type. Observation of single semicircle reveals the grains contributions only and semicircle arc can be modelled with single *RC* parallel circuit. *R* and *C* represent the bulk resistance ($R_b$) and bulk capacitance ($C_b$) of the material. The values of $R_b$, $C_b$ and relaxation frequency of the $AgNbO_3$ ceramics are given in Table I. On increasing temperature, the bulk resistance reduces due to increase in conductivity of the material.

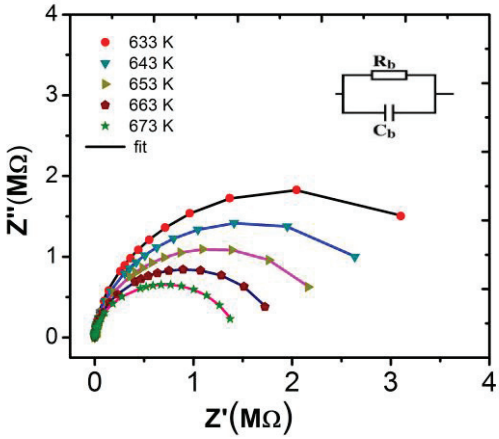


**FIGURE 2.** Experimental and calculated (solid line) of complex impedance data at different temperatures.

**TABLE I.** Bulk Resistance ($R_b$), bulk capacitance ($C_b$), Relaxation time ($\tau$) and Bulk Conductivity ($\sigma_b$) of $AgNbO_3$ ceramics

| T(K) | $R_b$ (MΩ) | $C_b$(pF) | τ(ms) | $\sigma_b(\Omega^{-1}m^{-1})$ |
|---|---|---|---|---|
| 633 | 4.0080 | 0.520 | 0.20 | $1.2213 \times 10^{-5}$ |
| 643 | 3.1211 | 0.517 | 0.16 | $1.5684 \times 10^{-5}$ |
| 653 | 2.4041 | 0.506 | 0.12 | $2.0362 \times 10^{-5}$ |
| 663 | 1.8438 | 0.496 | 0.091 | $2.6549 \times 10^{-5}$ |
| 673 | 1.4317 | 0.480 | 0.068 | $3.4192 \times 10^{-5}$ |

## Modulus Studies

The electric modulus can be calculated from the following equation:

$$M^* = M' + jM'' = j\omega C_o Z \quad (1)$$

where $Z = Z' - jZ''$, $M' = \omega C_o Z''$ and $M'' = \omega C_o Z'$, $\omega$ is the angular frequency and $C_o$ is the geometrical capacitance of the pellet. Figure .3 shows the real and imaginary parts of electric modulus spectra as a function of frequency at different temperatures. It is observed that $M''$ shows dispersion in the low frequency region and shows saturation at the high frequency region. It reveals that short range mobility of charge carriers is involved in conduction process [9, 10]. The imaginary part of electric modulus ($M''$) exhibits a peak at a frequency corresponding to the maximum of $M''$ ($M''_{max}$), which shifts towards higher frequency with temperature. The relaxation time (τ) and activation energy have been estimated by using the equations:

$$\tau\omega_{max} = 1, \omega_{max} = 1/2\pi f_{max} \quad (2)$$

where, $f_{max}$ is the relaxation frequency.

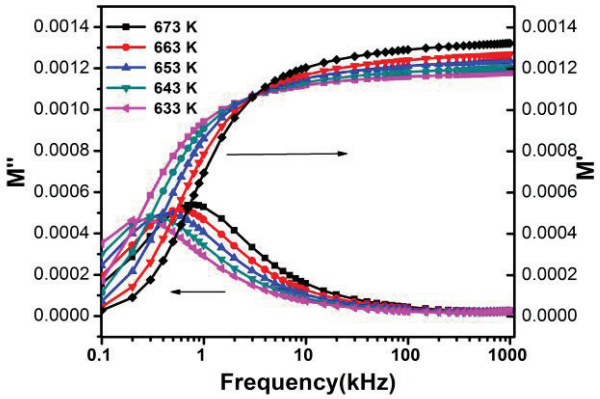


**FIGURE 3**. Real and imaginary part of the electrical modulus for $AgNbO_3$ ceramics.

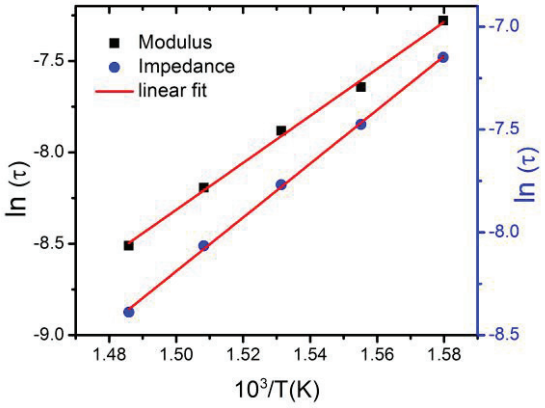


**FIGURE 4**. Arrhenius plot of Impedance and Modulus of $AgNbO_3$ ceramics

Figure.4 shows the variation of relaxation time with temperature which follows Arrhenius law. The activation energies calculated by using the relation:

$$\tau = \tau_0 exp(\frac{E_a}{k_B T}) \qquad (3)$$

where $\tau_o$ is the pre-exponential factor, $E_a$ is the activation energy for the relaxation process and $k_B$ is the Boltzmann constant. The activation energies are evaluated from the slope of the log ($\tau$) against $10^3/T$ linear fit and found to be 1.12 eV (from impedance data), 1.10eV (modulus spectrum).

## CONCLUSIONS

A detailed investigation on electrical properties of $AgNbO_3$ has been addressed through CIS analysis. Appearance of a semicircle indicates that grain relaxations present in the measured temperature and frequency range. The activation energy calculated from impedance and modulus data indicate that same type of charge carriers are responsible for conduction and relaxation. The estimated activation energy values reveals oxygen vacancy movements are responsible for conduction in $AgNbO_3$.

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