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Sample synthesis and characterization of $SrBi_{2-x}La_xNb_2O_9$ (x=0, 0.5) Aurivillius-type layered oxides

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ABSTRACT: The various Aurivillius oxides, SrBi2-xLaxNb2O9 (x=0, 0.5), were synthesized by using the conventional solider state reaction method. Phase analyses were performed by X-ray diffraction (XRD) analyses and Raman spectroscopy. Microstructural morphology was investigated by scanning electron microscopy (SEM). The Curie temperature, maximum permittivity and activation energy decrease with the increase in La content.

KEYWORDS: Solid state, XRD, Raman, SEM. Dielectric proprieties.

1 INTRODUCTION

Aurivillius phase compounds, as bismuth layer-structured ferroelectrics are generally formulated $(Bi_2O_2)^{2+}(A_{m-1}B_mO_{3m+1})^{2-}$ where A is a mono, bi or trivalent ion, B a tetra, penta or hexavalent ion, and m the number of BO_6 octahedral in each pseudo-perovskite block (m=1 to 5) [1]. Bismuth layer-structured ferroelectric materials have attracted an increasing attention for non-volatile Ferroelectric Random Access Memory (FeRAM) applications [2] [3].

$SrBi_2Nb_2O_9$ (SBN) and their solid solution have attracted much scientific attention due to their fatigue-free properties that make them potentially suitable candidates for application in the FeRAM [4]. But, this material suffers from the formation of oxygen vacancies. Whoever, to remain this problematic, many works suggest doping with rare earth, such as La^{3+},Nd^{3+}, Pr^{3+} etc., can decrease the vacancies concentration, hence effectively modified the electrical properties [5, 6, 7].

The aim of this study is about synthesis, structural and dielectric properties of Lanthanum doped $SrBi_2Nb_2O_9$ ceramics. $SrBi_2Nb_2O_9$ (SBN) and $SrBi_{1.5}La_{0.5}Nb_2O_9$ (SBL0.5N) were prepared by solid-state reaction.

2 EXPERIMENTAL

Starting materials $SrCO_3$, Bi_2O_3, La_2O_3 and Nb_2O_5 were weighed, mixed and calcined at 100 and 1200 °C during 12 hours for undoped and doped samples respectively. The calcined powders were then pressed into pellets, and sintered at 1150 and 1250 °C during 8 hours for undoped and doped pellets respectively. X-ray diffraction (XRD) was used to determine the crystal structure of the powders. Raman spectroscopy informs us about ion substitution. The microstructure of ceramics was observed by Scanning Electron Microscope (SEM). Temperature dependence of dielectric properties of $SrBi_{2-x}La_xNb_2O_9$ ceramics were investigated from the room temperature to 500°C at various frequencies 10 kHz, 100 kHz and 1 MHz.

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3 RESULTS AND DISCUSSION

XRD (Fig. 1) analysis reveals that all patterns show a single Aurivillius-type crystal, with the following lattice parameters a= 5.4114 Å, b=5.4449 Å, c=25.9013 Å for SBN and a=5.5995 Å, b=5.4338 Å, c=25.1060 Å for SBL0.5N. The highest peak occurred at 29.4° is corresponded to (1 1 5) orientation, which is consistent with the (1 1 2m + 1) highest diffraction peak in bismuth layered structured ferroelectrics systems [8].

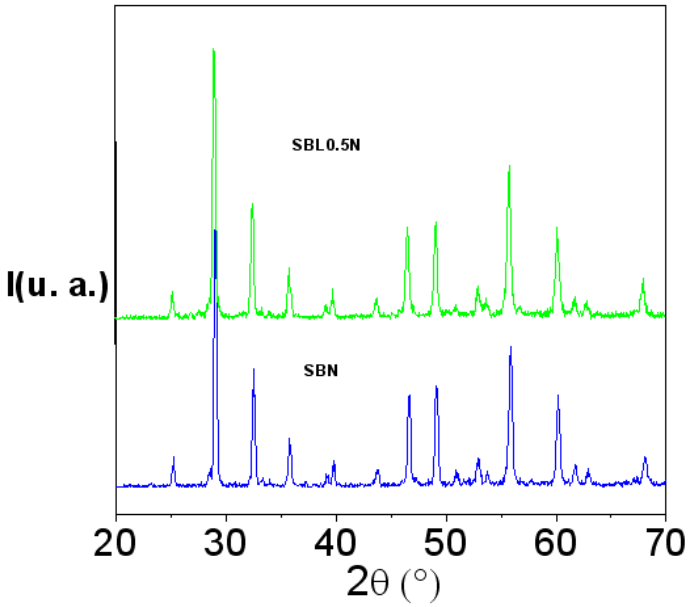

Fig. 1 X-ray diffraction patterns of SBN and SBL0.5N powders

Room temperature Raman spectra of SBL0.5N (Fig. 2) shows phonon modes around 60, 97, 175, 205, 575, and 838 cm^{-1}. These modes are related to the vibrational on two-layer Aurivillius phase. In the range 100–250 cm^{-1}, the bonds are overlapped for specimen that can be attributed to the disordered structure [9] [10].

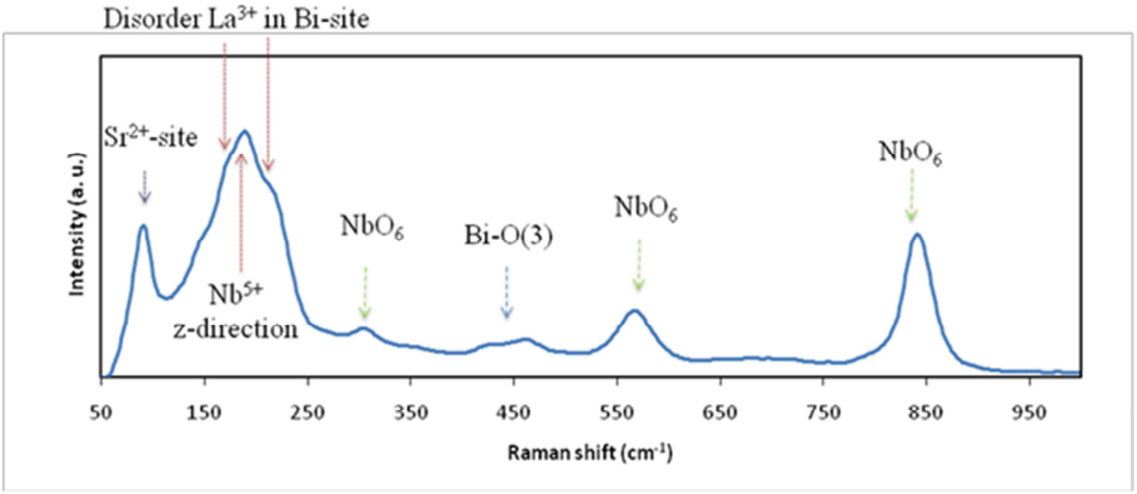

Fig. 2 Raman scattering of SBL0.5N

The SEM photos (Fig. 3) show that the SBL0.5N ceramic has an anisotropic plate-like microstructure, typically characteristic of Aurivillius ceramics [11].

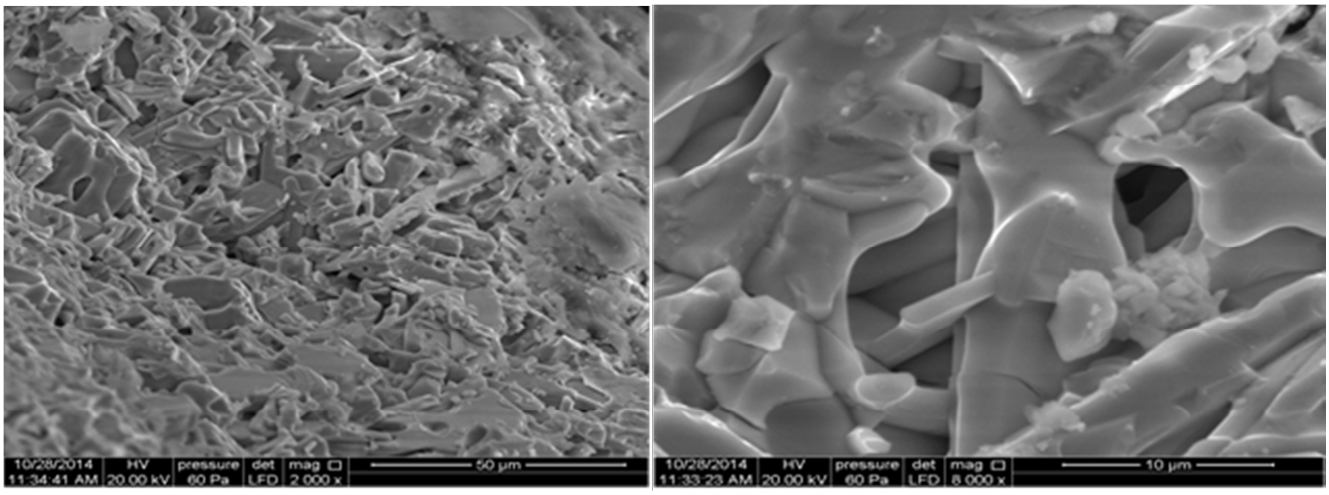

Fig. 3 Cross-sectional SEM photos of SBL0.5N.

Temperature dependence of dielectric properties of $SrBi_{2-x}La_xNb_2O_9$ ceramics were investigated from the room temperature to 500 °C at various frequencies 10 kHz, 100 kHz and 1 MHz (Fig. 4). The dielectric permittivity shows a sharp peak at the phase transition temperature for all frequencies. This indicates that SBN and SBL0.5N have a classical ferroelectric behavior. It was also observed for both ceramics, a strong frequency dispersion at lower frequencies and higher temperatures. The Curie temperature (T_c) decreases from 410 °C for SBN to 360 °C for SBL0.5N. This is probably due to La substitution which decreases the tilting of NbO_6 octahedral [12]. The dielectric permittivity peak of the ceramics decreases with La doping increasing, mainly due to the polarizability of Bi (lone pair of electrons) [13].

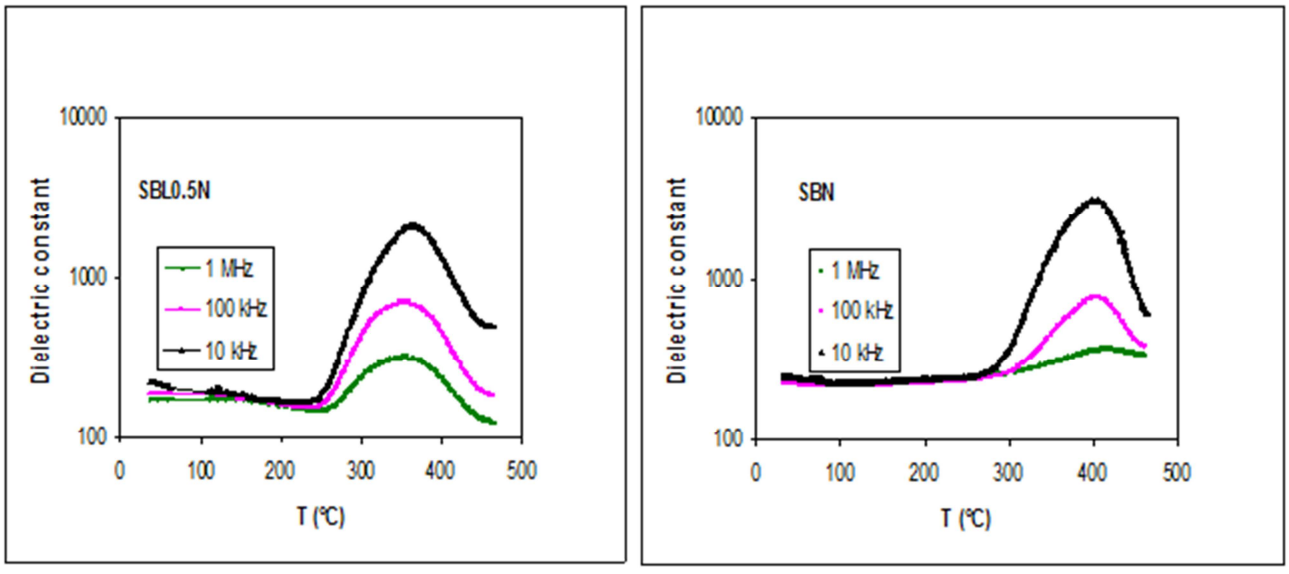

Fig. 4 Temperature dependence of the dielectric constant at various frequencies

The activation energy (E_a) of the ceramics in the high temperature region is calculated from the conductivity slope according to the reciprocal temperature (Fig. 5). The activation energy is about 2.052 eV for SBN and 1.189 eV for SBL0.5N. A

decrease in E_a may be due to chemical bond strengths with oxygen, knowing that the La–O (799 kJ/mole) bond is stronger than the Bi–O one (343 kJ/mole) [14].

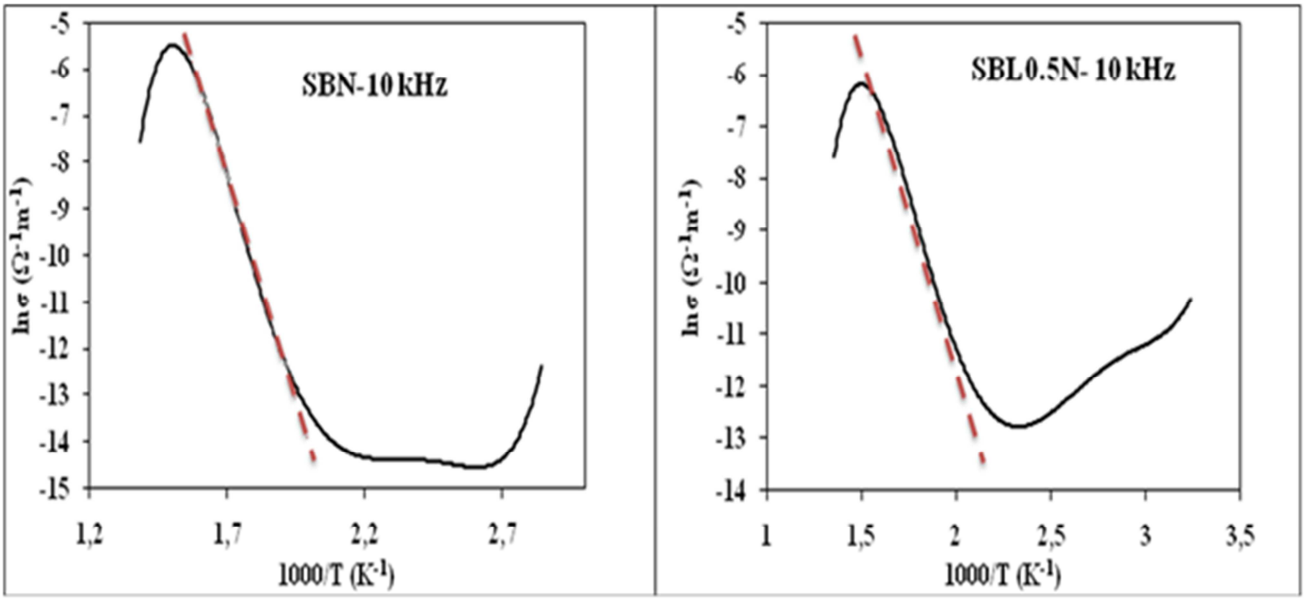

Fig. 5 Temperature dependence of ac conductivity

4 CONCLUSION

In summary, the effects of La^{3+} doping on the structure, dielectric proprieties have been investigated. A single phase of Bi-layered perovskite was confirmed for both compositions by XRD. The dielectric proprieties of the obtained ceramics were strongly dependent on the La.

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