Physical and Dielectric Properties of Silver Lithium Niobate Mixed Ceramic System

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Abstract
The perovskite niobates (ANbO₃) constitutes an interesting structural family. Silver lithium niobate Ag₁₋ₓLiₓNbO₃ (x = 0, 0.3, 0.5 and 0.7) mixed ceramic pellets were synthesized by the by solid-state reaction and sintering method. The lattice parameters of ceramic pellets were characterized by X-ray diffraction (XRD). The prepared samples show a perovskite structure and exhibit the orthorhombic symmetry at room temperature. Frequency dependent dielectric investigations, i.e., dielectric constant, loss tangent and electrical conductivity were carried out in the frequency range 10Hz-10MHz at room temperature.

Keywords: perovskite, silver lithium niobate, ceramic, X-ray diffraction (XRD), dielectric constant, loss tangent, electrical conductivity

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1. Introduction

The constituents of the silver lithium niobate (Ag₁₋ₓLiₓNbO₃) system are silver niobate (AgNbO₃) and lithium niobate (LiNbO₃). Silver niobate AgNbO₃ undergoes a sequence of phase transitions at 387°C orthorhombic to tetragonal and at 567°C, tetragonal to cubic [1,2]. Lithium niobate LiNbO₃ undergoes the phase transition at 1210°C, from the trigonal [Ferroelectric] to the trigonal [Paraelectric]. The non-linear acoustical properties of LiNbO₃ have led to the observation of a new physical effect. An acoustical tone burst stores energy within the crystal that is re-emitted at a later time of order of 70 μs. This effect can be characterized as an ‘acoustic memory’. This phenomenon is dependent on frequency and temperature [3]. X-ray investigations of Ag₁₋ₓLiₓNbO₃ solid solution ceramics (0 ≤ x ≤ 0.15) showed that small Li substitution causes a change of symmetry [4]. At room temperature, a phase boundary between orthorhombic and rhombohedral symmetry is observed for x = 0.05; this phase boundary is indicated also by dielectric properties. The Li- substitution leads to a gradual rise and shift of diffuse ε’ (T) maximum which is observed for (AN) as 227°C Instead of this diffuse maximum, a sharp one at 197°C is already observed for Ag₀.9₄Li₀.⁰₆NbO₃ [4].

In the present study pellets of Ag₁₋ₓLiₓNbO₃ (for x = 0, 0.3, 0.5 & 0.7) were prepared by conventional solid-state reaction method. Characterization of the samples was made by X-ray diffraction (XRD) and scanning electron microscopy (SEM). Dielectric measurements of all the prepared samples were carried out in the different frequency ranges 5Hz to 100Hz; 0.1KHz to 100KHz and 0.1MHz to 10MHz, at room temperature.

2. Experimental Details

The raw materials, used for preparing compositions, for present study, were silver oxide (Ag₂O), lithium carbonate (Li₂CO₃), and niobium pentaoxide (Nb₂O₅). Similar to preparation of silver sodium niobate [5,6,7] and silver potassium niobate [8,9,10], the sample of silver lithium niobate was also prepared by conventional sintering method, i.e., solid-state reaction method [11]. Prepared samples were characterized using XRD and SEM. The prepared and sintered pellets of all compositions were gold polished for Scanning Electron Micrographs (SEM) and electroded in metal-insulator-metal (MIM) configuration using air-drying silver paste for dielectric measurements.

X-ray diffraction (XRD) pattern of all the samples at room temperature have been obtained on Bruker’s D-8 ADVANCE X-ray diffractometer, using Cu-Kα filter radiation of 1.540598Å wavelength. Surface topography of the samples was studied by LEO-440 scanning electron microscope. The dielectric constant, loss tangent and conductivity of the prepared samples were measured and calculated with the help of ‘Solartron 1260 Impedance Gain Phase Analyzer’.

3. Results and Discussion

3.1. X-ray Diffraction Patterns
The X-ray diffraction patterns of Ag\textsubscript{1-x}Li\textsubscript{x}NbO\textsubscript{3} for x = 0, 0.3, 0.5 and 0.7 obtained from all the prepared samples have been shown in Figure 1, Figure 2, Figure 3, Figure 4. From X-ray patterns, it was found that at room temperature all the compositions show characteristic lines corresponding to the orthorhombic. Lattice parameters (Figure 5) also reveal the structures of present systems. From Scanning Electron Micrographs (SEM), the grain of different sizes with orthorhombic shape grows in the prepared samples of Ag\textsubscript{1-x}Li\textsubscript{x}NbO\textsubscript{3} [12]. Smaller grains occupy the space between the bigger grains, and thus reducing the porosity.
Figure 4. X-ray diffraction pattern of Ag0.3Li0.7NbO3 samples

Figure 5. Lattice parameters of different compositions of Ag1-xLi x NbO3 samples

Figure 6. Variation of dielectric constant (K) with frequency at room temperature for Ag1-xLi x NbO3 system in the high frequency range (0.1 MHz to 10 MHz)

Figure 7. Variation of dielectric constant (K) with frequency at room temperature for Ag1-xLi x NbO3 system in the frequency range 0.1 KHz to 100 KHz
3.2. Dielectric Properties

3.2.1. Dielectric Constant (K)

The variation of dielectric constant with frequencies, at room temperature, for different compositions of Ag\(_{1-x}\)Li\(_x\)NbO\(_3\) system, in the different frequency ranges 0.1 MHz to 10 MHz; 0.1 KHz to 100 KHz and 5 Hz to 100 Hz has been shown in Figure 6, Figure 7 & Figure 8 respectively.

From these figures, it has been observed that at room temperature, dielectric constant slightly decreased with increasing frequency except at higher frequencies, i.e., at 3-10 MHz, where dielectric constant slightly increased. However, dielectric constant decreases with increasing ‘Li’ content in Ag\(_{1-x}\)Li\(_x\)NbO\(_3\) in the measured frequency range (10 Hz to 10 MHz), except for x = 0, i.e., for AgNbO\(_3\), who show the anomalous behavior, in the frequency range 10 KHz to 10 MHz, where it decreased.

3.2.2. Tangent Loss (tan\(\delta\))

The variations of tangent loss (tan\(\delta\)) with frequency, at room temperature, have been shown in Figure 9, Figure 10, Figure 11, in the frequency range 5 Hz to 10 MHz. It has been observed that tangent loss very slightly decrease with increasing frequency and show a small increase for AgNbO\(_3\) at 3 KHz - 30 KHz. However, loss tangent decrease with increasing x-value, i.e., ‘Li’ contents in Ag\(_{1-x}\)Li\(_x\)NbO\(_3\), but magnitude of loss tangent for x = 0.7, is found higher than that for x = 0.3 and 0.5, in the frequency range 10 Hz to 100 KHz. Further, tan\(\delta\) very slightly increase with increasing frequency, and show a small decrease at 1.26 MHz. However, loss tangent decrease with increasing x-value, in Ag\(_{1-x}\)Li\(_x\)NbO\(_3\), in the frequency range 0.1MHz to 10MHz, but Ag\(_{0.7}\)Li\(_{0.3}\)NbO\(_3\) shows anomalous behavior above 6 MHz frequency, where it increases.
3.2.3. Electrical Conductivity ($\sigma$)

The variations of electrical conductivity ($\sigma$) with frequency, at room temperature, have been shown in Figure 12, Figure 13, Figure 14, in the frequency range 1 Hz to 10 MHz. It has been observed from these figures, that electrical conductivity increases with increasing frequency in all measured frequency range (10 Hz to 10 MHz), with a small decrease at 1.26 MHz. It has also been observed that electrical conductivity decrease as ‘Li’ content increase, in the frequency range 10 Hz – 2 MHz and Ag$_{0.7}$Li$_{0.3}$NbO$_3$ shows anomalous behavior, showing increase, in the frequency range 2 MHz to 10 MHz. The maximum values of electric conductivity have been observed $103.0 \times 10^{-6}$ $\Omega^{-1}$ cm$^{-1}$, $306.0 \times 10^{-6}$ $\Omega^{-1}$ cm$^{-1}$ and $91.3 \times 10^{-6}$ $\Omega^{-1}$ cm$^{-1}$ for $x = 0$, 0.3, and 0.5, i.e., for AgNbO$_3$, Ag$_{0.7}$Li$_{0.3}$NbO$_3$ and Ag$_{0.5}$Li$_{0.5}$NbO$_3$ respectively, at 10 MHz frequency.
4. Conclusions

In the present work, physical and dielectric properties of lithium doped silver niobate perovskite system, $\text{Ag}_{1-x}\text{Li}_x\text{NbO}_3$ for $x = 0, 0.3, 0.5$ and 0.7 have been investigated. From characterization of the prepared samples, it has been observed that at room temperature all the compositions are in orthorhombic phase. Frequency variations of dielectric constant, tangent loss and electrical conductivity were measured at room temperature in the frequency range 5Hz to 10 MHz.

The solid solution of perovskite silver lithium niobate ($\text{Ag}_{1-x}\text{Li}_x\text{NbO}_3$) can be formed over a whole composition range, and thus allowing a high degree of tailorability of physical properties to cover a broad range of technologically important dielectric, piezoelectric, ferroelectric, optoelectronic, optical, electrical, etc. properties. The observations in present study indicate that these systems have tremendous technological potential. Carrying out further careful and systematic studies, with varying composition and preparative conditions, appropriate materials for different industrial and technological applications can be developed out of these systems.

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