



Analytical and Hysteresis Loop Studies of Ferroelectric Al- Doped KNbO₃ Single Crystal

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Abstract

The doped KNbO₃ single crystal have been prepared using flux method by taking K₂CO₃ and Nb₂O₅ as starting materials, in the molar ratio of 1.2:1 with an impurity of Al₂O₃ (200mg). The good quality single crystals were obtained by this method. The sample are partially transparent suitable for optical properties. The detailed study of structural parameters, dielectric and Hysteresis loop studies were carried out in this paper. The prepared sample has been characterized by X-Ray diffraction method to determine lattice parameters. The material is orthorhombic with lattice parameters as: $a= 5.69399\text{\AA}$, $b=3.96801\text{\AA}$, and $c= 5.71852\text{\AA}$. The dielectric property shows the possibility of phase transition at high temperature. The material has high value of dielectric constant at Curie temperature of about 435°C. The hysteresis curve observed at room temperature to determine various parameters particularly, the spontaneous polarization (Ps), remanent polarization (Pr) and coercive field (Ec). The value of spontaneous polarization is determined from the hysteresis curve $9.632 \times 10^{-5} \mu\text{C}/\text{cm}^2$ at $E = 45.6\text{V}/\text{cm}$. The other parameters i.e. remanent polarization (Pr) is equal to $1.60 \times 10^{-5} \mu\text{C}/\text{cm}^2$, and coercive field (Ec) is 24.85 V/cm.

Keywords: KNbO₃; Single crystal, Dielectric properties, Hysteresis loop.

Introduction

Ferroelectric materials are the important class of electronic materials in electronic and optical industry¹. The potassium niobate crystal (KN) received substantial interest because of its multiple applications as a ferroelectric as well as a photorefractive material both from academic and commercial viewpoint². Higher dielectric constant of KN makes it a capacitor material³.

The particular attention has been focused on the phase transition⁴ because KN is the only ferroelectric crystal that exhibits same physical symmetry and the same sequence of transition⁵ as BaTiO₃. Therefore, it is very interesting to compare their physical properties and find out the similarities and differences⁶. KNbO₃ exists in an orthorhombic symmetry at room temperature that undergoes phase transitions at -10, 225, and 425°C from rhombohedral → orthorhombic → tetragonal → cubic, respectively⁴. These properties have been studied by using different impurities such as Fe, Al, Cu, Co, Mn etc. Now a days the role of impurities in ferroelectric crystal is important. Because, the addition of external impurities, affects domain structure and phase transition temperature⁷. This paper reports, the role of impurities to study the dielectric constant with temperature at different frequencies. Also, the hysteresis loop studies were carried out to know the basic parameters of the KNbO₃ single crystal.

Methodology

Crystal structure determination: The XRD studies of the KNbO₃ sample were carried out to characterize the crystal symmetry. The XRD pattern of Al- doped KNbO₃ single crystal with different concentration as shown in the Figure-1. X-ray structure analysis was done using programmed software such as powder-x, check cell, cell151 etc.

Dielectric and phase transition studies: The grown crystals were cut in small chips for dielectric studies in to a few (mm)² areas with thickness which is about 0.2 mm to 0.5 mm. The experiments to determine the dielectric constant were carried out w. r. to temperature at different frequencies i.e. 1 kHz. 10 kHz

Hysteresis Loop Studies: Ferroelectric hysteresis loop was observed using TF-Analyzer testing system at IIT, Mumbai. The hysteresis loop studies used for the measurement, of various parameter of ferroelectric hysteresis loop such as spontaneous polarization (Ps), remanent polarization (Pr), coercive field (Ec) etc.

Results and Discussion

XRD Analysis: Figure-1 shows the XRD pattern for Al- doped KNbO₃ single crystals. The XRD data was analyzed by powder-x programme. It has been observed that it exist in a single phase

perovskite structure since no trace of any secondary phase is detected. The calculated lattice parameters of Al- doped KNbO_3 single crystals are: $a = 5.69399 \text{ \AA}$, $b = 3.96801 \text{ \AA}$ and $c = 5.71852 \text{ \AA}$. It shows orthorhombic structure at room temperature.

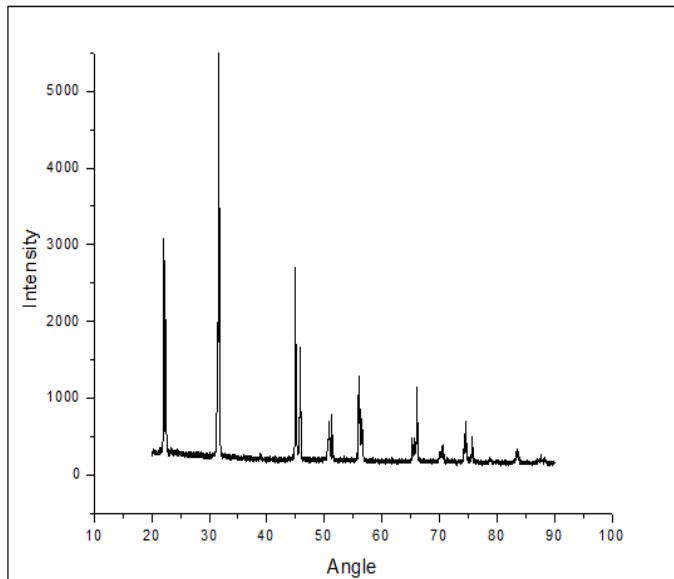
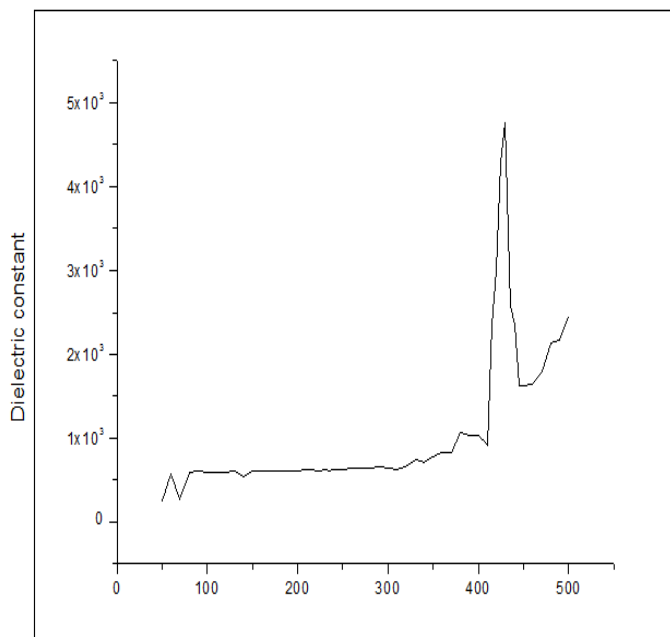


Figure-1
 XRD pattern of Al_2O_3 (200 mg) doped KNbO_3 single crystal

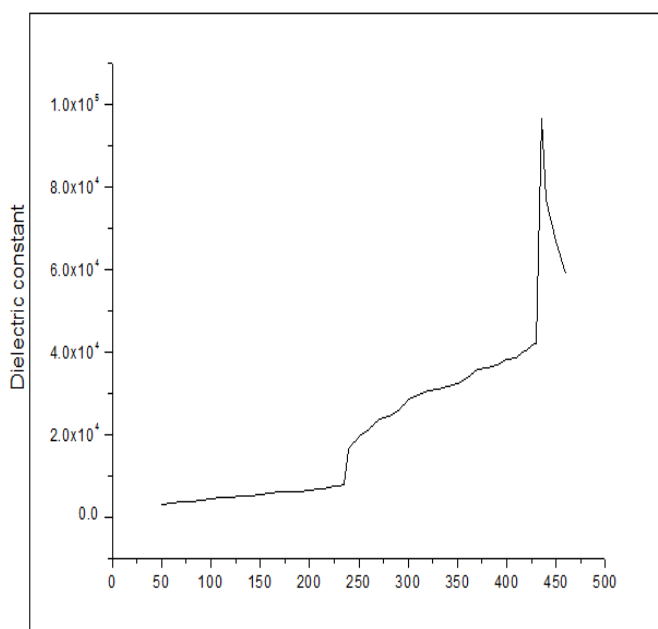
Dielectric constant: The variation of dielectric constant with respect to temperature at different frequencies is shown in Figure-2 (a and b). The high value of dielectric constant observed is 4.76×10^3 at 1 kHz and 9.76×10^4 at 10 kHz. The Curie temperature (T_c) was estimated as 435°C that is in agreement with that of KN single crystals indicating that addition dopant does not influence the Curie temperature⁸. The Al doped KN single crystal followed the same trend of enhancement of dielectric constant with temperature just like that of pristine KN crystal⁹. The frequency has insignificant influence on the Curie temperature¹⁰ as expected shown in Figure-2.

Figure-3 (a and b) shows the variation of loss tangent with temperature at two different frequencies. In both frequencies, the loss tangent values gradually increased with temperature up to Curie temperature, shoot up maximum at Curie temperature and then dropped to lower value. However, two breaks in smooth curve was observed at high frequency, indicating the possibility of phase transitions.

Hysteresis study: The hysteresis curve between electric field and electric polarization (P-E curve) is shown in Figure-4. The hysteresis measurements of sample were carried out at room temperature at 100 Hz. The measured value of remanent polarization (P_r) was found to be $1.60 \times 10^{-5} \mu\text{C}/\text{cm}^2$ and spontaneous polarization (P_s) is $9.62 \times 10^{-2} \mu\text{C}/\text{cm}^2$ at $E = 45.6 \text{ V}/\text{cm}$. The coercive electric field is $24.85 \text{ V}/\text{cm}$. The value of P_s shows the good stability and good storage capacity.



a



b

Figure-2
 Variation of dielectric constant w.r.to temperature at (a)1 kHz and (b) 10 kHz.

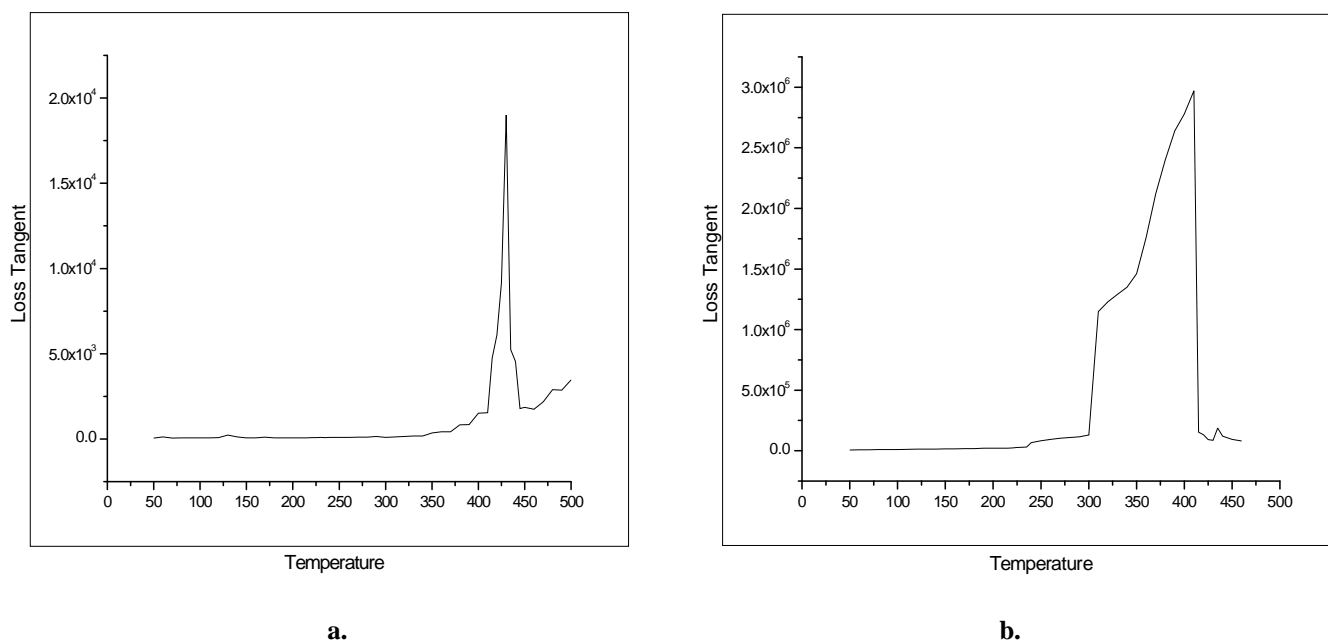


Figure-3
 Variation of loss tangent w.r.to temperature at (a) 1 kHz and (b) 10 kHz

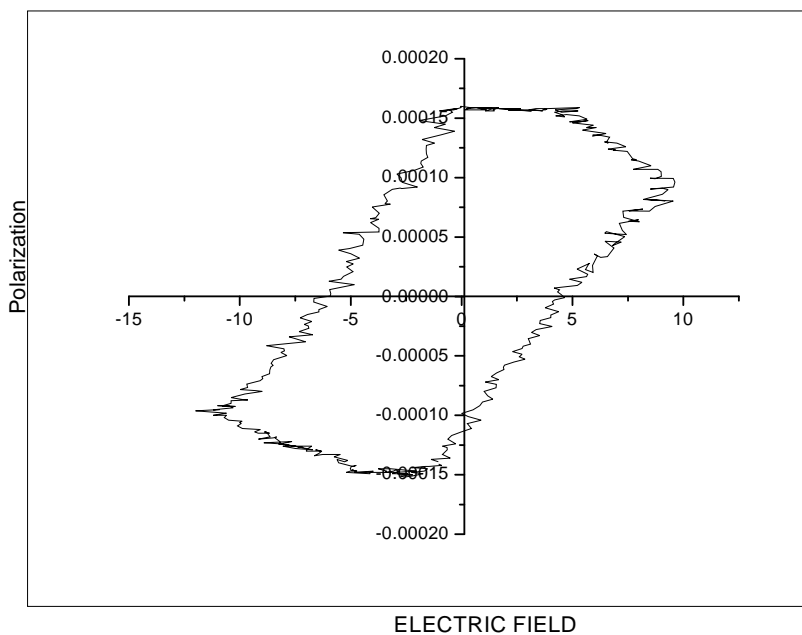


Figure-4
 P-E hysteresis loops of Al₂O₃ [200mg] doped KNbO₃ Single crystal

Conclusion

Al – doped KNbO₃ single crystal has been successfully grown by flux method. A good quality, partially transparent large single crystal has been obtained. The grown crystal has orthorhombic structure at room temperature with lattice

parameters: $a = 5.69399 \text{ \AA}$, $b = 3.96801 \text{ \AA}$ and $c = 5.71852 \text{ \AA}$. The dielectric studies shows high dielectric constant at the Curie temperature of about 435°C. The spontaneous polarization (P_s) is calculated from the hysteresis loop which is equal to $9.62 \times 10^{-5} \mu\text{C}/\text{cm}^2$ at $E = 45.6 \text{ V}/\text{cm}$.

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