Recent Development and Techniques in PHYSICAL SCIENCES

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Chapter - 7

Dielectric Studies of PSN Mixed System

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Chapter - 7

Dielectric Studies of PSN Mixed System

Manish Uniyal, S.C. Bhatt, Sidharth Kashyap, Aditya Joshi and Richa Saxena

Abstract

Dielectric and Electrical properties of $K_{1-x}Na_xNbO_3$ (X=0, 0.2, 0.4) ceramics have been investigated in the temperature range 35^0C-250^0C at different frequencies varying from 0.1 MHz to 1MHz. The samples have been prepared by the conventional solid-state reaction method and sintering process. It is observed that dielectric constant, loss tangent and electrical conductivity increases with increasing temperature. Near the transition temperature dielectric constant, loss tangent and electrical conductivity of these samples show anomalous behavior. All the prepared samples show orthorhombic structure at room temperature.

Keywords: Transition temperature, dielectric constant, loss tangent, electrical conductivity.

1. Introduction

The class of materials known, as perovskite is of considerable technological importance, particularly with respect to their physical properties such as pyro-and piezo-electricity, dielectric susceptibility, linear & nonlinear electro-optic etc. Many of these properties vary enormously from one perovskite to another. The change in physical properties is particularly large when the external conditions, such as temperature, pressure, electric-field etc. are altered. Such effects occur in connection with the simultaneous presence of phase transition in the system, where the atomic structure of the perovskite changes either discontinuously or continuously into another form [1]. The dielectric properties perovskite structure $K_{1-x}Na_x$ NbO_3 for some of the compositions has been extensively studied at high temperature [2] and it shows a number of ferroelectric phases with high spontaneous polarization. The phase transition in perovskite crystal is generally assumed to be due to the instability of the temperature dependent low frequency optical phonons at transition temperature [3,4]. Potassium Sodium Niobate ceramic ($K_{1-x}Na_xNbO_3$) with perovskite structures are widely used for transducer applications with broad ranges of technologically important dielectric, piezoelectric, ferroelectric and electro-optic properties. The structure at room temperature, which, basically is of the orthorhombic type. $K_{1-x}Na_xNbO_3$ (x=0), is a particularly promising ferroelectric with its combination of relatively low dielectric constant ($\varepsilon 1 = 155, \varepsilon 2 = 44, and \varepsilon 3 = 980)$ with extremely high electro-optic coefficients (r_{42}=380pm/V, r_{33}=64pm/V). This makes $KNbO_3$ very attractive for opto-electronic devices, including high-speed electro-optic switches, modulators, and frequency doublers [5-7]. Because of the interest in this material for high frequency, electro-optical devices, investigation and analysis of dielectric properties have been performed on $KNbO_3$ pellets as well as mixed ceramics [8-12]. Dielectric measurements on this material were first reported by Matthias and Remeika [13] and then by Shirane *et al.* [14] and observed well defined ferroelectric hysteresis loops from room temperature to 400^0C, for compositions from about 10 to 100% (mol) of K in $K_{1-x}Na_xNbO_3$. Cross [15] has predicted, theoretically, the phase diagram of $KNbO_3$-$NaNbO_3$ mixture from phenomenological arguments. Dielectric properties were reported by Narayan Murty *et al.* [16] and Egerton & Dillon [17]. $NaNbO_3$ which is antiferroelectric at room temperature, when mixed with, small amount of $KNbO_3$, becomes ferroelectric, which creates interest in the present investigation to investigate this composition in a varying composition and temperature range.

In this paper, we report the results of investigation on dielectric constant, tangent loss and electrical conductivity of the ceramic system $K_{1-x}Na_xNbO_3$ (x=0, 0.2 & 0.4) in temperature range 35^0C-250^0C.

2. Preparation

The raw materials used for preparing the compositions from this system are sodium carbonate, potassium carbonate and niobium penta oxide. The two carbonates were both reagent-grade products of the Qualigens Fine Chemicals and the niobium pentaoxide was 99.9% purity from Fluka Chemie AG CH-9471 Buchs. The starting material was dried at 200^0C for one hour to remove absorbed moisture. Different compositions of $K_{1-x}Na_xNbO_3$ for (x=0, 0.2 & 0.4) were prepared by weighing the sodium carbonate, potassium carbonate and niobium penta-oxide (starting materials) in proper stoichiometric proportions. The mixture was calcined in the platinum crucible, in air, at 950^0C for 2h for carbonate removal. After cooling, in dry air, the calcined mixtures were weighed to ensure complete carbonate removal.

The pre-sintered mixture was ground and pressed into pellets of 10mm diameter. All the pellets were placed on a platinum crucible and sintered, in air, at 1050^{0}C for 26 h. The sintered pellets were electroded using air-drying silver paste for dielectric measurements.

3. Characterization

X-ray powder studies were performed at room temperature with a SEIFERT 3000P. X-ray diffractometer using filtered Cu Kα 1 radiation of 1.540598^0 A wavelength, in which Ni is used as filter. The instrument is well calibrated with silicon standard samples and the lines obtained are matching with the standard lines. At room temperature all PSN ceramics exhibit the orthorhombic symmetry. The subcell parameters were obtained using the auto-X computer software and were compatible with those obtained earlier for ceramics [16, 18-28]. The results are listed in table 1 given below.

Table 1: Lattice parameters of $K_{1-x}Na_xNbO_3$ at room temperature

Composition	Lattice parameter (A°)		
	a	**b**	**c**
$KNbO_3$	4.027	4.057	3.958
$K_{0.8}Na_{0.2}NbO_3$	4.002	4.04	3.946
$K_{0.6}Na_{0.4}NbO_3$	3.998	4.011	3.919

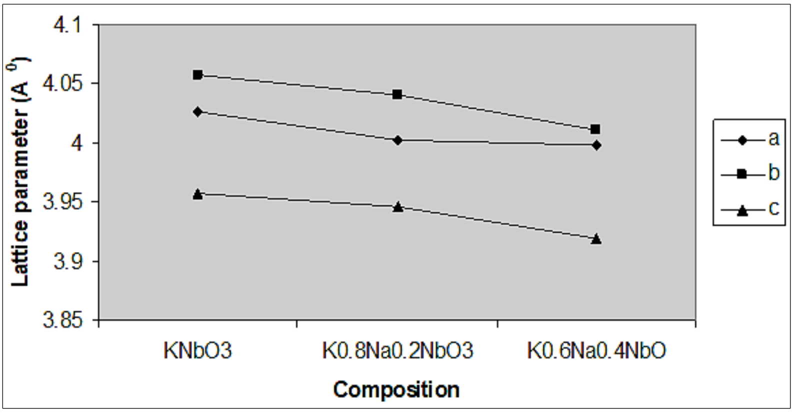

Fig 1: Composition dependence of lattice parameters

4. Measurements

The variation of dielectric constant, loss tangent and electrical conductivity at frequency 0.1 MHz and 1MHz in the temperature range 35^{0}C-250^{0}C have been plotted in Fig. 2 & 3, 4 &5, 6 & 7 respectively. These measurements were performed on RCL meter model FLUKE PM 6306.

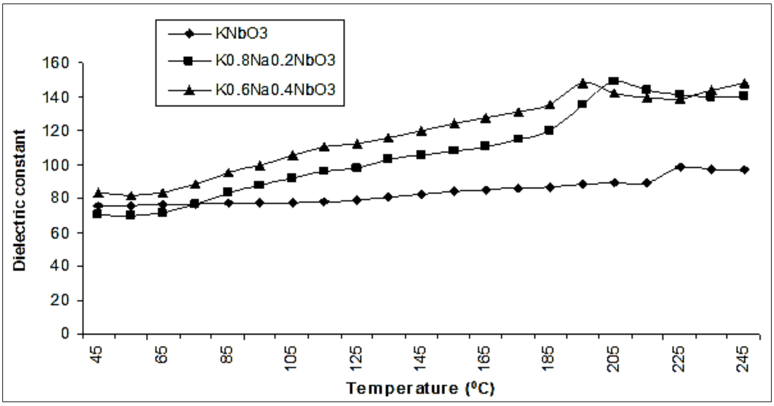

Fig 2: Variation of dielectric constant with temperature for $K_{1-X}Na_xNbO_3$ at 1 MHz

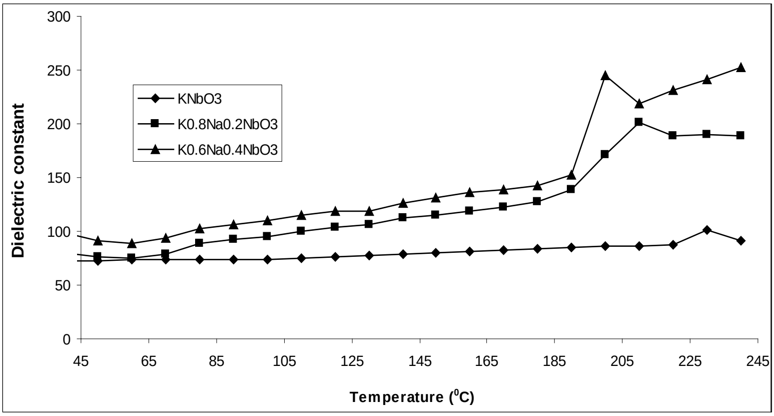

Fig 3: Variation of dielectric constant with temperature for $K_{1-X}Na_xNbO_3$ system at 100 KHz

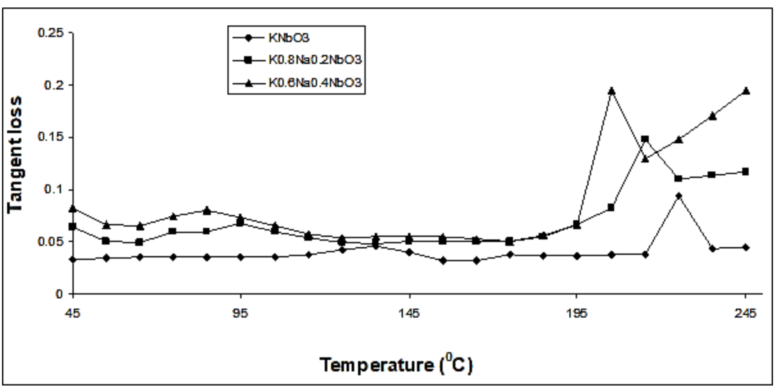

Fig 4: Variation of tangent loss with temperature for $K_{1-X}Na_xNbO_3$ system at 1MHz

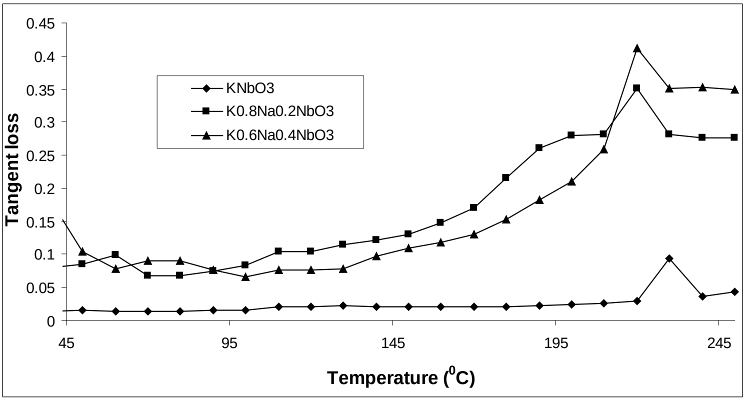

Fig 5: Variation of tangent loss with temperature for K1-X NaxNbO3 at 100 KHz

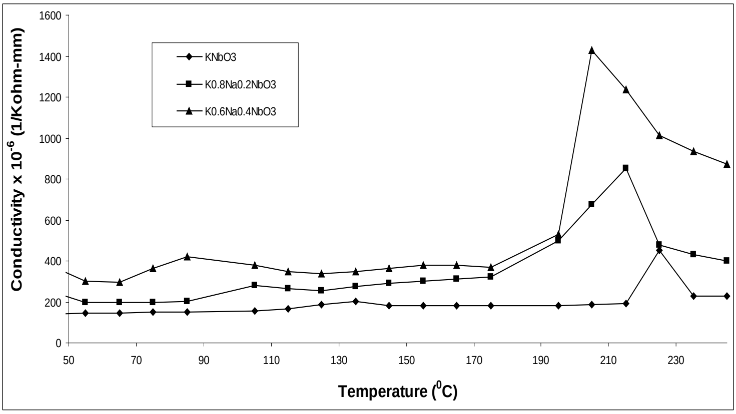

Fig 6: Variation of electrical conductivity with temperature for K1-X NaxNbO3 system at 1 MHz

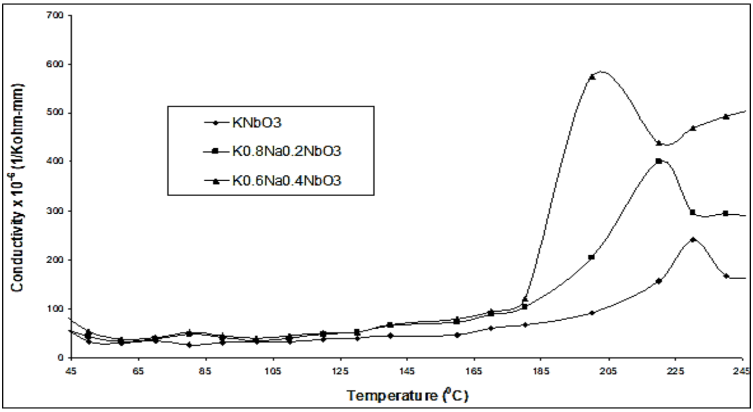

Fig 7: Variation of conductivity with temperature for K1-X NaxNbO3 system at 100 KHz

5. Result and discussions

We have measured dielectric constant, loss tangent of $K_{0.6}Na_{0.4}NbO_3$, $K_{0.8}Na_{0.2}NbO_3$ and $KNbO_3$ ceramic pellets using HP Impedance Analyzer and LCR meter PM 6304 at the temperatures from 35^0C to 250^0C. The effect of mixing of Na in $KNbO_3$, i.e, $K_{1-x}Na_xNbO_3$ on the lattice parameters is shown in Fig. 1. All the lattice parameters, ie., a, b & c show similar nature of variation. The value of the lattice parameters, increases with increasing the % of Na in $K_xNa_{1-x}NbO_3$ up to approximately 2% of Na, thereafter it is decreases, thereby showing a compositional transition with anomaly at 2%, which may be due to the difference in size of atom of K & Na. The temperature dependence of dielectric constant, loss tangent and conductivity at 1MHz and 100 KHz is shown in Figs. 2 & 3,4 & 5 and 6 & 7 respectively. From these figures it is observed that the mixed system of $K_{1-x}Na_xNbO_3$ has a transition from orthorhombic to tetragonal at about 220^0C. It is 225^0C for $KNbO_3$ 215^0C for $K_{0.8}Na_{0.2}NbO_3$ and 205^0C for $K_{0.6}Na_{0.4}NbO_3$, which shows that transition temperature from orthorhombic to tetragonal shifts towards lower temperature as we increase the quantity of anti-ferroelectric material sodium niobate in the mixed system, which is in agreement with previous observations [2, 29].

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