Electrical characterization of perovskite nanocubes of $Na_{0.5}K_{0.5}NbO_3$ at low temperature

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Abstract

Due to the toxicity of lead, there is an urgent necessity to develop lead-free alternatives to replace the currently dominant lead-based piezoelectric such as Pb(Zr,Ti)O_3 (PZT). $Na_{0.5}K_{0.5}NbO_3$ based piezoelectric nanomaterial are promising because of their relatively high Curie temperatures as well as good piezoelectric coefficients among the non-lead piezoelectrics. We have successfully synthesized 3-dimensional perovskite nanocubes of functional ternary transition-metal oxide of sodium potassium niobate $Na_{0.5}K_{0.5}NbO_3$ by using novel synthesis method. We have carried out resistance and impedance spectroscopy studies at low temperature using two probe technique. For this electrical measurement, we have used CCR system. There has been much interest in the texturing of electro ceramics, due to the possibility of achieving enhanced electrical properties. A major purpose for this electrical characterization study is to examine how $Na_{0.5}K_{0.5}NbO_3$ (NKN) pellet behaves under low temperature. Copyright © 2017 VBRI Press.

Keywords: Electro ceramic, $Na_{0.5}K_{0.5}NbO_3$ Pellet, low temperature, impedance spectroscopy, two probe technique.

Introduction

The most successful synthesized and commercially in-use piezoelectric ceramics are based on lead zirconate titanate Pb(Zr,Ti)O_3 (PZT) in which 60% lead (Pb) is present in its corresponding lead oxide (PbO) form and even it can cause problems with health and learning abilities, and can even cause death. Environmental concerns over their lead content could disappear with the advent of a new ceramic that is lead-free **[1]**. The ceramics having general structure of perovskite ABO_3 has been reported in literature such as ternary transition metal oxide, in which sodium potassium niobate ($Na_{0.5}K_{0.5}NbO_3$) is one of the important oxide which open a new domain of both technologically and fundamentally interest **[2,3]**.

Therefore, the subject of lead-free material such as sodium potassium niobate ($Na_{0.5}K_{0.5}NbO_3$) is highly interested to many research scientists and motivate them to investigate for alternative to PZT. In view of this, we mainly focused on to the study of $Na_{0.5}K_{0.5}NbO_3$ ceramic pellet system electrical properties by two probe techniques experimentally **[4,5]**. In case of sodium potassium niobate ($Na_{0.5}K_{0.5}NbO_3$), consist of sodium niobate $NaNbO_3$ is one of the anti-ferroelectric material at wide range of temperature and potassium niobate $KNbO_3$ is ferroelectric material, but during mixing of Potassium (K) at sodium (Na) site in $NaNbO_3$ becomes ferroelectric in nature **[6,7]**. Nowadays, cube like nanostructure have attracted wide attention in their promising application in various field such as nanosensors, electronic nanodevices, functional nanomaterial etc **[8]**. Therefore, lots of interest generated to study such amazing $Na_{0.5}K_{0.5}NbO_3$ ceramic nanomaterial.

In addition to the scientific researcher community, these materials have also fascinated the more attention in electronic industries. The piezoelectric ceramics are widely used as probe for medical ultrasonography, piezoelectric transformers, surface acoustic wave (SAW) filters, ultrasonic motors, piezoelectric buzzers **[9-15].** The main aim of this study was to draw together the existing knowledge on various electrical properties related to the sintered $Na_{0.5}K_{0.5}NbO_3$ system based electro ceramics at low temperature **[16]**.

Experimental

Materials and method

The synthesis of $Na_{0.5}K_{0.5}NbO_3$ is done by various physical as well as chemical techniques **[17-22]**, but we prepared the $Na_{0.5}K_{0.5}NbO_3$ by novel method **[23]**. Initially, for electrical characterization of Sodium potassium niobate ($Na_{0.5}K_{0.5}NbO_3$) pellet system, we weigh about 0.5 gram of ultrafine nanopowder of sodium potassium niobate ($Na_{0.5}K_{0.5}NbO_3$). These powders are converted into circular shape pellet having thickness of about 2 mm and diameter of about 10 mm by applying 5 tone forces by using powder press pellet making machine. To achieve proper strength (such as mechanical) for $Na_{0.5}K_{0.5}NbO_3$ ceramic pellet, we have done sintering of the pellet at about 1000^0 C for 3 hours.

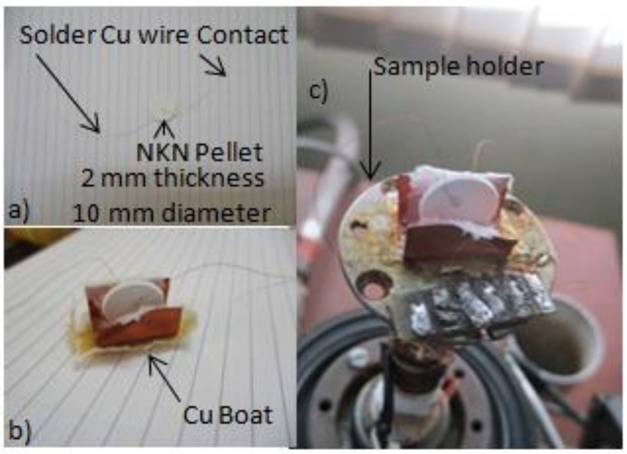

Fig. 1. (a) $Na_{0.5}K_{0.5}NbO_3$ Pellet (b) $Na_{0.5}K_{0.5}NbO_3$ Pellet placed on Cu boat (c) Cu boat placed on CCR system for Electrical Characterization

For making electrical contact with $Na_{0.5}K_{0.5}NbO_3$ ceramic pellet, we used silver paste on both the sides. Then soldered by using copper wire on both sides, which is shown in **Fig. 1(a)**. Afterwards the pellet is sticked with the help of glue and placed on copper boat, shown in **Fig. 1(b)**. This boat which contain $Na_{0.5}K_{0.5}NbO_3$ pellet is placed on sample holder which is shown in **Fig. 1(c)**. Then we closed the whole set up and start rotary pump to achieve high vacuum level around at 10^{-3} torr. In this experimental setup, re-circulating helium is used to get low temperature state. We carried out two probe electrical characteristic measurements with the help of CCR system.

Impedance spectroscopy (IS) is one of the relatively efficient and powerful technique for characterizing many of the electrical properties of electro ceramic nanomaterials (i.e. in pellet form). We have monitored the electrical characteristics of $Na_{0.5}K_{0.5}NbO_3$ (NKN) pellet during cooling as well as heating cycle. For cooling of $Na_{0.5}K_{0.5}NbO_3$ system experiments, we have cooled the sodium potassium niobate ($Na_{0.5}K_{0.5}NbO_3$) system from room temperature (300K) to low temperature (25K) and measure impedance of $Na_{0.5}K_{0.5}NbO_3$ system at constant frequency $1.00x10^{+6}$ Hz, the temperature drop down up to 25K. During the heating of $Na_{0.5}K_{0.5}NbO_3$ system from 25K to 300K, at about 25 K increment, we hold the temperature and measured the impedance of $Na_{0.5}K_{0.5}NbO_3$ from 1000 Hz to 2,000,000 Hz with frequency step size of 1000 Hz.

Characterizations

The synthesized nanomaterial surface morphology was studied by scanning electron microscopy (FE-SEM, FEI Company Inspect F50 model) with the help of secondary electron mode detection. For the electrical characterization of sodium potassium niobate ($Na_{0.5}K_{0.5}NbO_3$) pellet we used Impedance analyzer (Agilent E4980A Precision LCR Meter) and for low temperature we have used Closed–Cycle Helium Refrigerator (CCR) system.

Results and discussion

Surface morphology of powder material characterization

The synthesized $Na_{0.5}K_{0.5}NbO_3$ nanomaterials and its surface morphology is characterized using FE-SEM technique. The products are look like Nanocubes shape with the size in the range 90 nm to 100 nm. The nanocubes shape aggregates with distinct edges are observed in FE-SEM and it has been shown in **Fig. 2**.

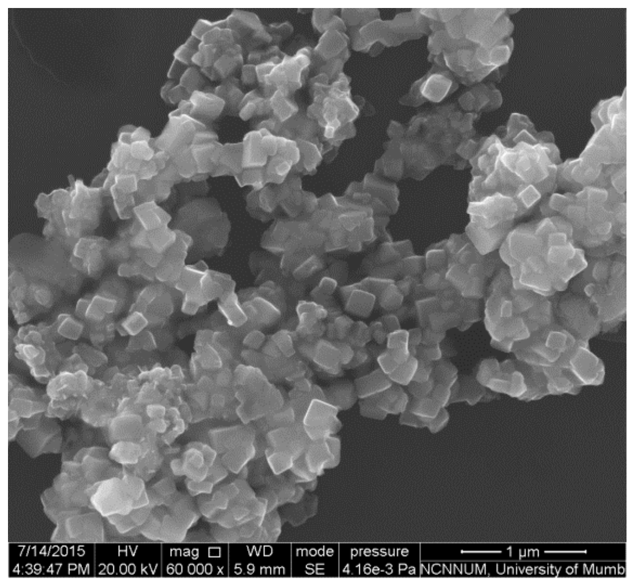

Fig. 2. FE-SEM image of $Na_{0.5}K_{0.5}NbO_3$ nanocubes.

Electrical characterization of $Na_{0.5}K_{0.5}NbO_3$system

Impedance Analysis

During impedance measurement of NKN system, in **Fig. 3(a)** shows that, at lower frequency, it is observed that the resistance of $Na_{0.5}K_{0.5}NbO_3$ is extremely high and varies arbitrarily, however at high frequency the resistance of $Na_{0.5}K_{0.5}NbO_3$ decreases linearly in log scale and reaches less than 100 Ω at high frequency. The graph in **Fig. 3(b)** shows that, frequency versus reactance curve. The reactance is increases rapidly at lower frequency and stable or constant above 10 kHz. Variation of impedance with frequency of sodium-potassium niobate ($Na_{0.5}K_{0.5}NbO_3$) system is shown in **Fig. 3(c)**. Impedance (denoted as Z) is an expression of the opposite that an electronic component in system offers to alternating electric current. It is seen from the **Fig. 3(c)**, that there is continues linear decrease in impedance at lower to higher frequency for both room temperature and low temperature.

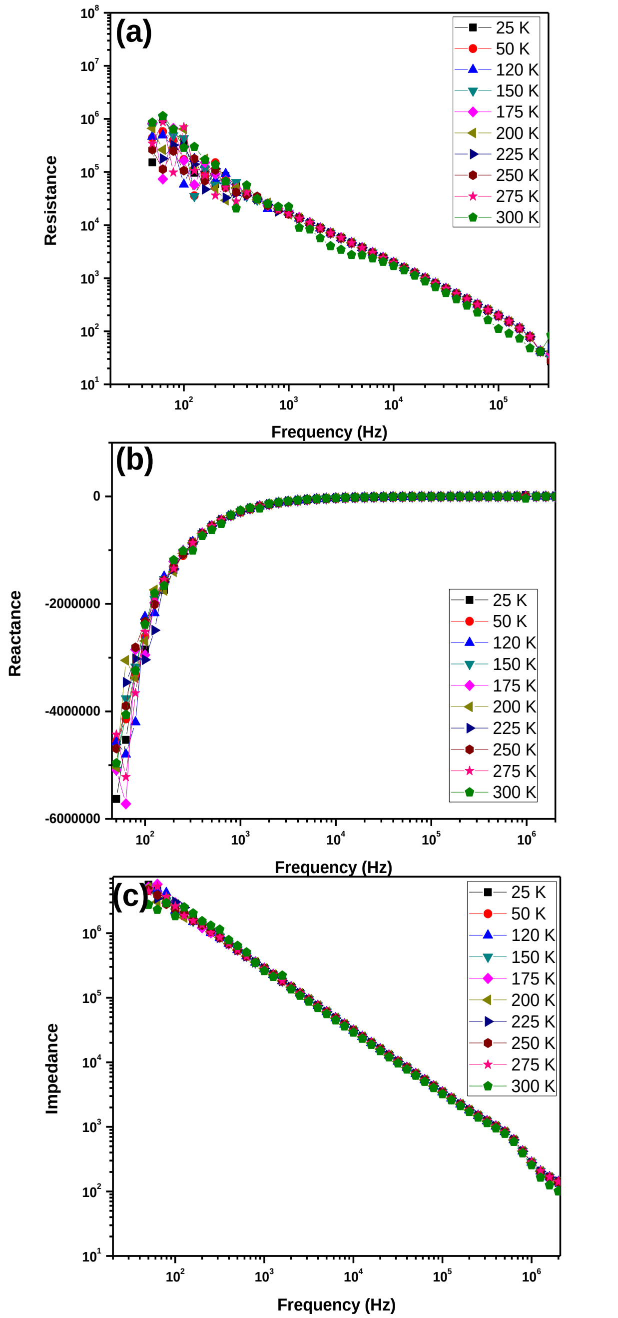

Fig. 3. Impedance analysis (3a) Variation of resistance of $Na_{0.5}K_{0.5}NbO_3$ system with frequency (3b) Variation of reactance of $Na_{0.5}K_{0.5}NbO_3$ system with frequency (3c) Variation of impedance of $Na_{0.5}K_{0.5}NbO_3$ system with frequency.

Admittance analysis

The admittance data presented, in **Fig. 4(a)** shows that at lower frequency the conductance of $Na_{0.5}K_{0.5}NbO_3$ ceramic pellet system is low, while as higher frequency range at room temperature (300 K) conductance increases from 1.54 $E^{-4}\Omega^{-1}$ to 71.16 $E^{-4}\Omega^{-1}$ and in case of low temperature (such as 275 K to 25 K) conductance increases from 2.00 $E^{-4}\Omega^{-1}$ to 12.07 $E^{-4}\Omega^{-1}$ and again decreases up to 9.04 $E^{-4}\Omega^{-1}$. While as in case of susceptance, at low frequency it is stable and then very slowly increase and at higher frequency susceptance is rapidly increases from 1.28 $E^{-4}\Omega^{-1}$ to 68.31 $E^{-4}\Omega^{-1}$ for both room temperature as well as low temperature, which is shown in **Fig. 4(b)** graph of Variation of susceptance (B) with frequency of sodium-potassium niobate (NKN) system. The resultant admittance of $Na_{0.5}K_{0.5}NbO_3$ system is very stable up to 100 kHz, then it increases at applied higher frequency range, for room temperature admittance is increase from 1.60 $E^{-4}\ \Omega^{-1}$ to 9.86 $E^{-4}\Omega^{-1}$, while as for low temperature admittance is increase from 1.87 $E^{-4}\Omega^{-1}$ to 7.06 $E^{-4}\ \Omega^{-1}$. The observed admittance profile is shown in **Fig. 4(c).**

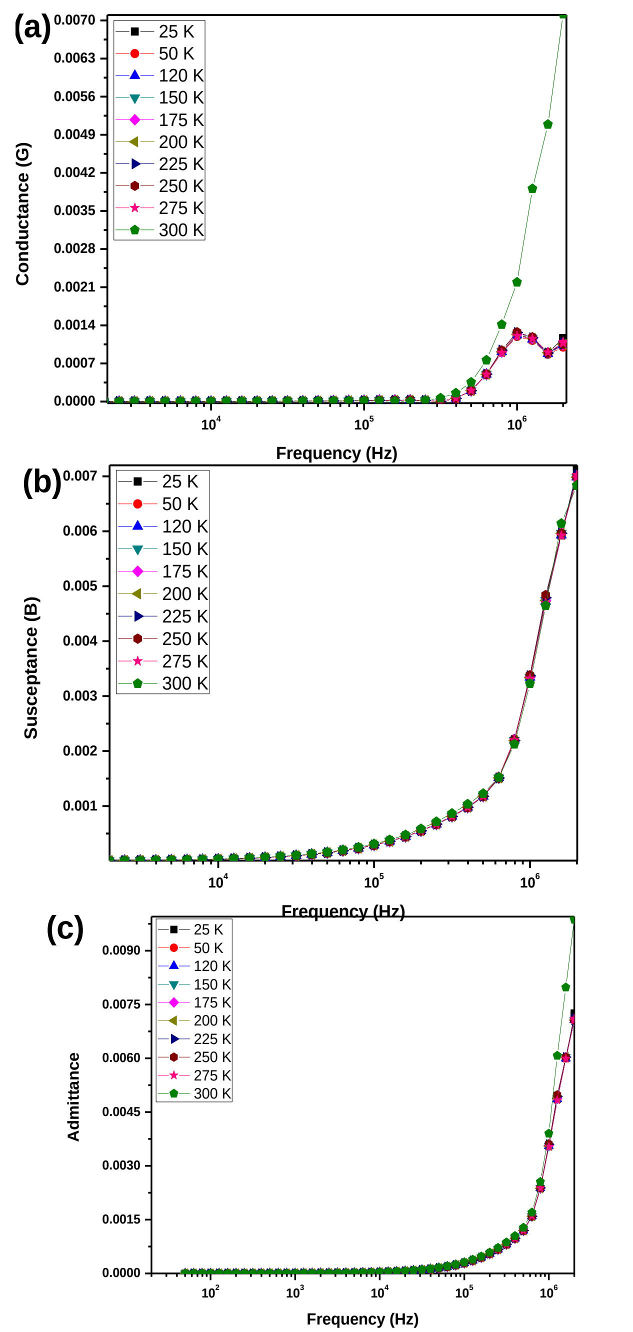

Fig. 4. Admittance analysis (4a) Variation of conductance (G) of $Na_{0.5}K_{0.5}NbO_3$ system with frequency (4b) Variation of susceptance (B) of $Na_{0.5}K_{0.5}NbO_3$ system with frequency (4c) Variation of admittance of $Na_{0.5}K_{0.5}NbO_3$ system with frequency.

D and Q factor analysis

Basically, dissipation factor (denoted by D) is reciprocal of quality factor therefore it can measure loss rate of energy during electrical characterization in a dissipative system. Dissipation factor represents quality (i.e. durability) of electro ceramic material during electrical measurement. The dissipation factor recorded for $Na_{0.5}K_{0.5}NbO_3$ **Fig 5(a)** at lower frequency for low temperature is slightly more as compare to room temperature, therefore the DF at room temperature having better efficiency. But at higher frequency range, the whole scenario changes such as, the dissipation factor at room temperature is more as compare to low temperature, therefore the DF at low temperature has better efficiency, which is seen from the above graph **Fig. 5(a).** The quality factor (denoted by Q) is dimensionless parameter. The quality factor observed for $Na_{0.5}K_{0.5}NbO_3$ **Fig. 5(b)** does not shows any drastic changes for room temperature at both high and low frequency, therefore it shows that higher rate of energy loss relatively to the store energy, but at low temperature for higher applied frequency at around 300 kHz, it shows drastic changes (in Q factor), which indicates a lower rate of energy loss relatively to the store energy, which is shown in **Fig. 5(b)**.

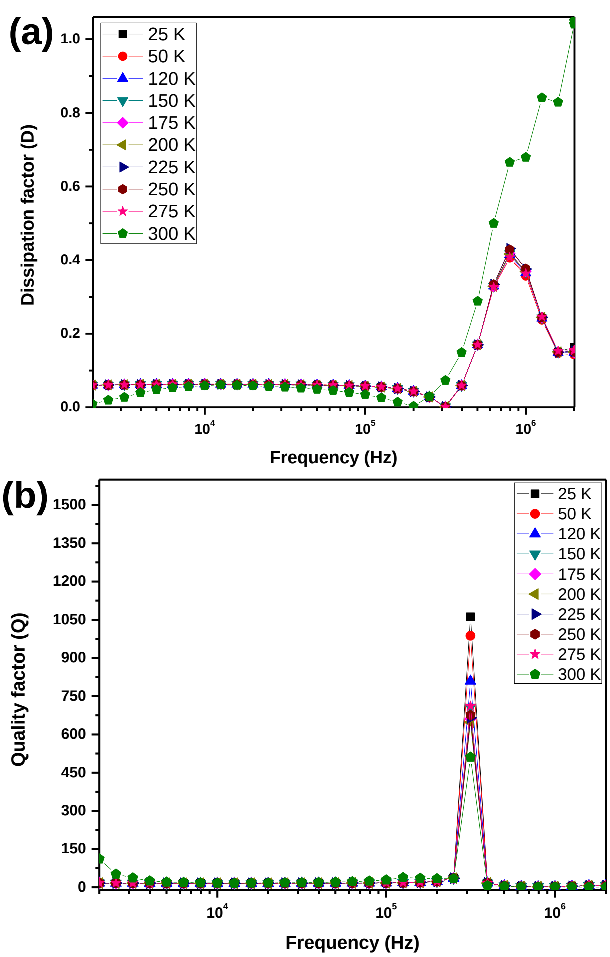

Fig. 5. Dissipation and Quality factor analysis (5a) Variation of Dissipation factor (D) of $Na_{0.5}K_{0.5}NbO_3$ system with frequency (5b) Variation of Quality factor (Q) of $Na_{0.5}K_{0.5}NbO_3$ system with frequency.

Phase angle and dielectric constant analysis

The phase angle at lower frequency for both low and room temperature does not show any significant changes but at higher frequency at around 200 – 300 kHz, the phase angle drastically increases for both low temperature and room temperature shown in **Fig. 6 (a)**. The dielectric material is nothing but electrical insulator and even it can be polarized by an applied electric field. In case of dielectric properties initially decreases up to 500 kHz for both low and room temperature, but at higher applied frequency (> 500 kHz) there is sudden increase, which is shown in the graph **Fig. 6 (b)** of variation of dielectric constant of $Na_{0.5}K_{0.5}NbO_3$ system with frequency.

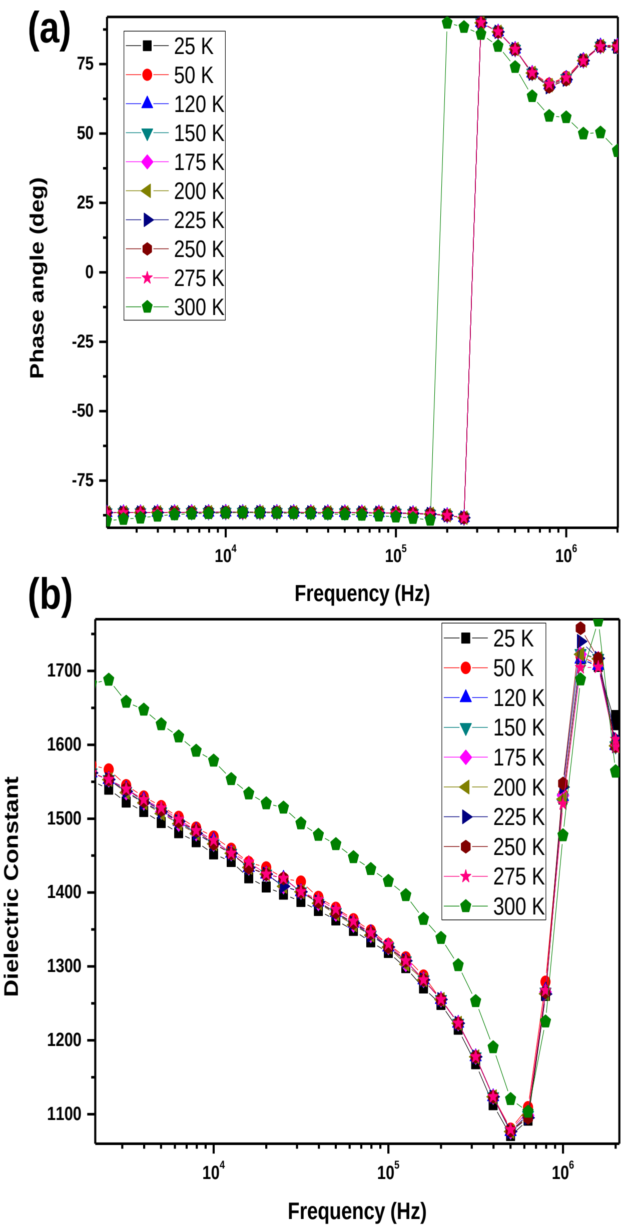

Fig. 6. Phase angle and dielectric constant analysis (6a) Variation of phase angle (deg) of $Na_{0.5}K_{0.5}NbO_3$ system with frequency (6b) Variation of dielectric constant of $Na_{0.5}K_{0.5}NbO_3$ system with frequency

Capacitance and inductance analysis

In case of variation of capacitance with frequency given in **Fig. 7(a)** shows that initially the capacity to store electric charge is decreases for lower frequency (up to 500 KHz) at both room and low temperature, but at higher frequency range (above 500 KHz) it is seen that the capacity to store electric charge is increase for both room and low temperature. It is seen from the graph **Fig. 7(b)** that there is continues decrease in inductance from lower frequency to higher frequency.

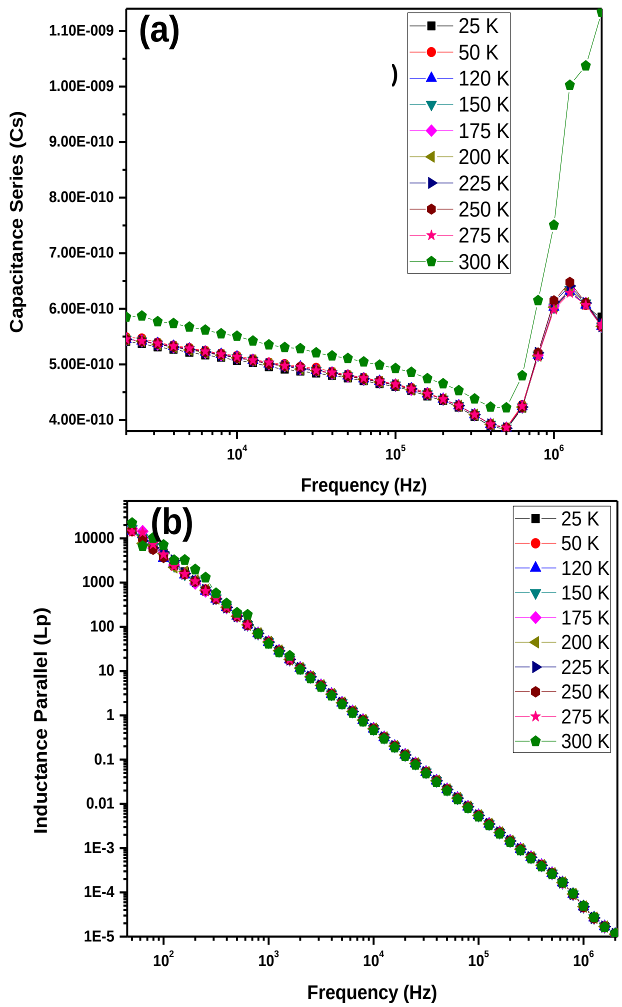

Fig. 7. Capacitance and inductance analysis (7a) Variation of capacitance series (Cs) of $Na_{0.5}K_{0.5}NbO_3$ system with frequency (7b) Variation of inductance parallel (Lp) of $Na_{0.5}K_{0.5}NbO_3$ system with frequency.

Conclusion

Electrical characterization has been carried out successfully over a range of frequencys from 1000 Hz to 2,000,000 Hz on sodium potassium niobate $Na_{0.5}K_{0.5}NbO_3$ ceramic pellet system at low temperature (25 K) to room temperature (300 K). The pervoskite nanocubes of $Na_{0.5}K_{0.5}NbO_3$ (NKN) used for present experiment is synthesized at our lab by using very low cost equipment and without introducing any seed material. The structural morphologies as well as average particle size were examined with scanning electron microscopy (FE-SEM) techniques and we got average partial size of Nanocubes in the range of 90 nm to 100 nm. Electrical characterization of $Na_{0.5}K_{0.5}NbO_3$ system at low temperature to make it applicable in cryogenic environment. Resistance (R) Vs reactance (X) based electrical characterization instruments provide a means for studying the impedance, dielectric properties of sodium potassium niobate $Na_{0.5}K_{0.5}NbO_3$ ceramic pellet. We have mainly focused on during heating (from 25 K to 300 K) electrical parameter measurements. The results of experiments show the various electrical characteristics properties of $Na_{0.5}K_{0.5}NbO_3$ system such as resistance, reactance, impedance, conductance, suceptance, admittance, inductance, capacitance, dissipation factor, quality factor, phase angle, dielectric constant etc.

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