

Composition-Dependent Dielectric and Piezoelectric Properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ Ceramics

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Sodium potassium niobate, $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$, is a ceramic material that exhibits dielectric and piezoelectric properties, which can be tuned by changing the composition of the material. The dielectric properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics are characterized by a high dielectric constant and low loss, which makes them useful for applications in capacitors, filters, and resonators. Pellets of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) were prepared by the solid-state reaction technique. The structural and morphological study was carried out on the prepared compositions at room temperature (RT). A Piezoelectric indirect constant (d_{33}^*) was obtained for the prepared compositions. Additionally, dielectric measurements were performed at frequencies 10-1000 kHz, from ambient temperature to 500°C. For the prepared compositions with $z = 0.175$, a break in the XRD peak shifting patterns was observed, and the average grain size was calculated at $2.044 \pm 0.3 \mu\text{m}$. Also, the maximum indirect piezoelectric constant (d_{33}^*) was observed for $z = 0.175$ among the prepared compositions. For the prepared compositions, compositions with $z = 0.140$ showed the highest dielectric constant (peak value 1486, 385°C). Among the prepared samples, dielectric constant (ϵ), loss tangent ($\tan \delta$) and electric conductivity (σ) were found to be minimum for the composition with $z = 0.175$ at all the measured frequencies. Anomalous dielectric, piezoelectric and structural properties are evident for the samples with $z = 0.175$ among the prepared samples. Overall, the composition-dependent dielectric and piezoelectric properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics make them promising materials for use in various electronic and sensing applications where high-performance dielectric and piezoelectric materials are required.

Keywords: Ceramics; Dielectric constant; Electric conductivity; Grain size; SEM; Ferroelectrics

1 Introduction

$\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics have attracted significant attention due to their unique composition-dependent dielectric and piezoelectric properties. These ceramics are a type of perovskite oxide with a complex crystal structure, and their properties can be optimized by adjusting the potassium content (z) in the material. The dielectric properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics refer to their ability to store electrical charge and are characterized by a high dielectric constant and low loss. The piezoelectric properties, on the other hand, refer to the ability of the material to convert mechanical energy into electrical energy and vice versa. The piezoelectric properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics can be optimized by adjusting the potassium content, which affects the crystal structure and polarization of the material. The ferroelectric oxide-based PZT [Lead zirconate titanate]

has been broadly used due to its large piezoelectric and dielectric properties^{1,2}. It may be observed due to its co-existence of two or more phases³. However, the well-known PZT compound has a large amount of lead. Considerable interest has been growing in developing lead-free materials due to the high toxicity of lead. The lead-free solid solutions exhibit significant electro-physical properties near MPB (morphotropic phase boundary) area. It might be explained by the volatility that developed in the polarization state or by sudden structural modifications at the MPB that changed the ferroelectric domains⁴. The ABO_3 -type perovskite materials have been investigated due to their large dielectric, piezoelectric, optoelectronic and ferroelectric properties⁵. The recent advancement of piezoelectric devices requires the urgent need for environmentally friendly materials that can replace the widely used PZT (lead zirconate titanate) system. Also, lead-free materials and compounds are widely used owing to their non-toxicity. One prominent lead-free alkali niobate- based

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compound is sodium potassium niobate ($\text{Na}_{1-z}\text{K}_z\text{NbO}_3$), which has a perovskite structure. Saito *et al.*⁶ observed a large piezoelectric constant ($d_{33} \sim 416$ pC/N) in Li-modified NKN-based ceramics with co-dopants of Ta and Sb⁶. Nonetheless, the piezoelectric coefficient of the lead-based system was much higher than NKN ceramics⁷. The mechanism underlying the improved electrical characteristics around the MPB is very important. An MPB could have multiple different phases. Also, MPB may have two different phases (with a change in lattice) with different symmetries. For the compositions close to MPB, combining these phases may lead to various polarization directions, which may maximize the dielectric and piezoelectric properties even in the presence of dopants. Large piezoelectricity in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics at the morphotropic phase boundary is caused by polarization extension and rotation, intermediate phases, and elastic softening of lattice⁸⁻¹⁰. The high piezoelectric constant ($d_{33} \sim 416$ pC/N) present in the solid solutions of ferroelectric lead-free $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics has thus been the subject of widespread research due to environmental concern. The development of numerous potential polarization directions at MPB, which may show extreme electrophysical features, has drawn much interest to the $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramic systems^{11,12}. The thickness of the coexisting area is also related to the particle size. In the field of sensors and actuators, electrical properties are essential factors. The piezoelectric effect is used in various applications, including ultrasonic transducers, accelerometers, and underwater sound sources¹³.

With the addition of Na at the K site, the transition temperature (T_c) has been changed, and a negligible change in structure may be observed^{14,15}. Also, improvement may be observed in their losses, densification and temperature stability *etc.*^{14,15}. To enhance the dielectric and ferroelectric properties, doping is the best possible choice. Ions like Li, Ta, and Sb *etc.*, can be used at A- site and B-site to improve the electrical properties of NKN ceramics¹⁵⁻¹⁹. Ferroelectric materials having a perovskite kind of structure may alter their property with the change in processing conditions. Also, the Li-modified compositions show fair piezoelectric properties among the lead-free piezoceramics¹⁹.

Further, because of their large electrical properties, the sodium-potassium niobate ceramics may be used for ultrasonic sensors, capacitors, modulators, and memory devices. In this study, the prepared compositions of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics have undergone a thorough analysis of their structural and dielectric properties.

Solid-state reaction was used to prepare the NKN ceramic compositions. At room temperature, XRD data of the prepared samples were taken. From RT to 500 °C temperature and at a frequency range 10-1000 kHz, dielectric properties (ϵ , $\tan \delta$ and σ) of the prepared compositions were measured. Also, for the prepared samples, the piezoelectric constant (d_{33}^*) was measured. The prepared samples were examined using SEM images at room temperature.

The study of composition-dependent dielectric and piezoelectric properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics is an important area of research with significant potential for the development of high-performance materials for a range of applications.

2 Experimental Details

Sodium carbonate (Na_2CO_3), potassium carbonate (K_2CO_3) and niobium pentoxide (Nb_2O_5) were used as raw materials. All the mentioned raw materials are 99.99% pure. In order to remove the absorbed moisture, the raw materials were dried at 200 °C for two hours. They were also accurately weighed in accordance with the stoichiometric ratio. The compositions were ground wet in acetone for the first two hours followed by dry wet. The grinding cycle was performed for 6 hrs. For grinding, an agate mullet mortar and pestle are used. To remove carbonate, compositions were calcined at 900 °C. To keep the mixture at a cool stage, dry air was used. They were further pressed into pellets (8 mm dia, ~ 2 mm thick), at 0.2 GPa pressure. The pellets of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics ($0.120 \leq z \leq 0.210$) were sintered at 1200 °C for 4 hours. The pellets were single sintered. The preparation methodology has been illustrated in the previous communications²⁰⁻²².

All the prepared powder samples of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) were taken for XRD measurements. The XRD patterns of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ are measured using an X-ray diffractometer (PANalytical, X'PERT PRO, with $\text{CuK}\alpha_1$ radiation of wavelength 1.5406) for structural measurement. The data is collected by the X-ray equipment at a scanning rate of 6°/min. To view the SEM images of the prepared samples, scanning electron microscopy is used with model number EVO 18 Special Edition (CARL ZEISS). At different frequencies, dielectric measurements were carried out using an LCRmeter (Fluke, PM 6306). Compositions were electroded, in a metal-insulator-metal (M-I-M) arrangement, with a conducting silver paste for dielectric measurements.

3 Results and Discussion

The XRD patterns are expected to show changes in the crystal structure of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics. As the potassium content increases, the crystal structure of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics undergoes changes from tetragonal to cubic. This is due to the effect of the potassium ions on the crystal structure, which leads to an increase in the polarization and a decrease in the symmetry of the crystal lattice. Fig. 1 shows the measured XRD patterns of the $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics ($0.120 \leq z \leq 0.210$). For this purpose, fine powder samples were collected. With the support of “Inorganic Crystal Structure Database (ICSD)” data, the peaks were indexed. The X-ray diffraction patterns show polycrystalline behavior. XRD data of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics are consistent with the code 01-074-2025 of the ICSD database. This was done with the help of the most intense peaks corresponding to (100), (110) and (200), *etc*²³.

A continuous shifting of the intense peaks (100), (110), and (200) to elevated angles was obtained in the measured compositional range ($0.120 \leq z \leq 0.210$), with an increase in potassium content (z value). For the composition $z=0.175$, the most intense peaks were measured moving to a lower angle, indicating a break in the peak shifting tendency, as shown in Fig. 2. A similar investigation has been reported in NKN ceramics, near $z = 0.475$ and 0.500 ²⁴⁻²⁶. The experimental break in the peak shifting patterns may be attributed to the lattice change.

The lattice parameters were calculated for the prepared $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ samples. Rietveld refinements

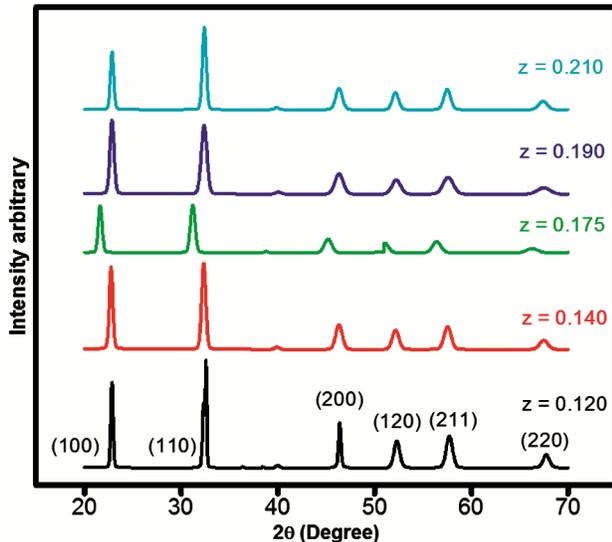


Fig. 1 — XRD patterns of the $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) ceramics.

have been performed using XRD data to evaluate lattice parameters of the prepared $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics. High Score Plus software has been used to refine the lattice parameters. The background was modeled using the Legendre polynomial and the peaks' profiles were refined using a pseudo-Voigt function^{27,28}. For the prepared compositions, the XRD patterns show monoclinic phase. The selected space group for the refinements was $Pm\bar{6}$ ^{23,29}. The variation of lattice parameters for the prepared NKN samples (z) has been shown in Fig. 3. The cell parameters show an abnormal nature for the compositions $z = 0.175$, among the prepared samples. For the prepared compositions, a rise was observed in c/a ratio plot for $z = 0.175$, which exhibits structural anomaly in this compositional region. The SEM images of the prepared compositions were shown in Fig. 4. The average grain size was calculated $2.044 \pm 0.3 \mu\text{m}$. The samples have fine grains. The images were obtained with the help of SEM machine (EVO 18, Special edition, Zeiss). The SEM images have same magnification ($10 \mu\text{m}$). For the prepared compositions, the variation of density and grain size with composition (z) were shown in Fig. 5. The density of NKN ceramics was found to increase with increasing K content (z), peaking at $z = 0.175$ and then decreasing with further increase in z ; however, the grain size was found to have the opposite variation, decreasing with increasing z , reaching minimum for $z = 0.175$ and increasing with further increase in z . Furthermore, the structural anomaly, at $z = 0.175$, may be the reason for the observed highest density, among the prepared samples²⁵. The composition- dependent dielectric properties has been

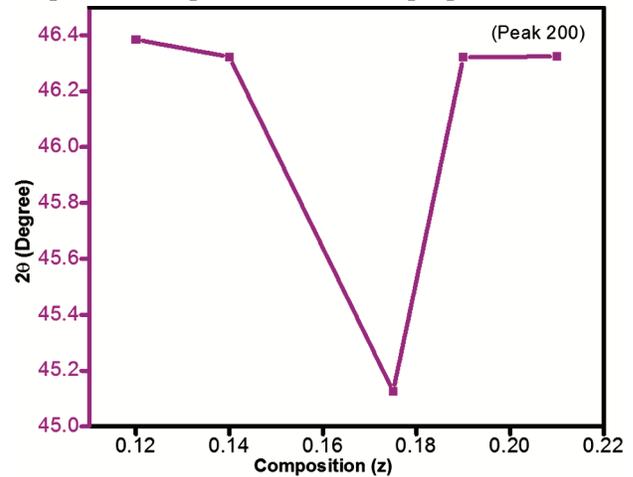


Fig. 2 — Peak (200) shifting tendency with sample (z), in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) ceramics.

discussed. The dielectric properties (ϵ , $\tan \delta$ and σ) have been increasing with an increase in potassium content (z value). They were found increasing up to the concentration $z = 0.140$. For composition $z = 0.175$, the measured values of ϵ , $\tan \delta$ and σ were

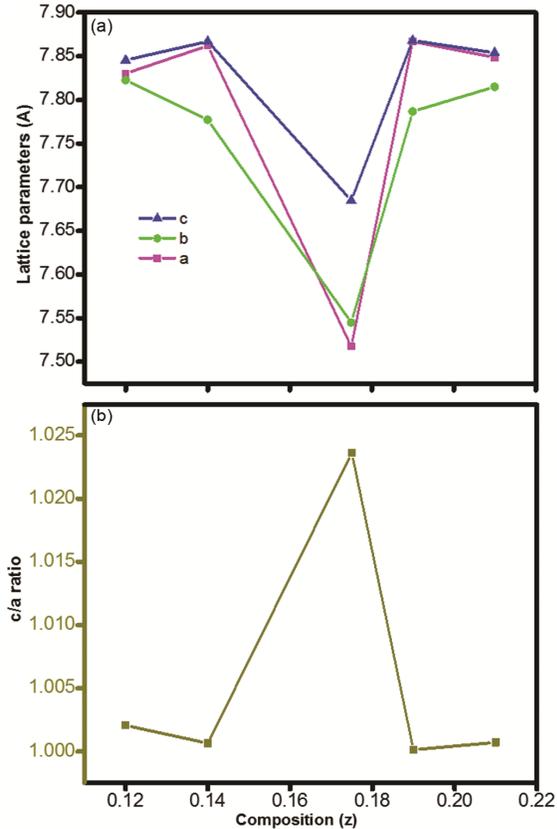


Fig. 3 — (a) Variation of Cell parameters with sample (z), and (b) variation of c/a ratio with sample (z), in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) ceramics.

found decreasing. At 1 MHz, the measured composition ($0.120 \leq z \leq 0.210$) dependence of ϵ , $\tan \delta$ and σ with different temperatures, were shown in Fig. 6(a-c). An anomalous change was seen in the dielectric properties (ϵ , $\tan \delta$, and σ) near $z = 0.175$, at all the observed temperatures with 1 MHz frequency. Furthermore, among the prepared compositions, an irregular change was found in the dielectric properties (ϵ , $\tan \delta$ and σ) near $z = 0.175$, at different frequencies with RT. Fig. 7(a-c) indicates the observed composition ($0.120 \leq z \leq 0.210$) dependence of ϵ , $\tan \delta$ and σ with different frequencies. The measured dielectric nature can be attributed to the change in polarization extension and rotation followed by samples, applied frequency, residual stress, porosity, grain size, thickness of grain boundary, and dielectric

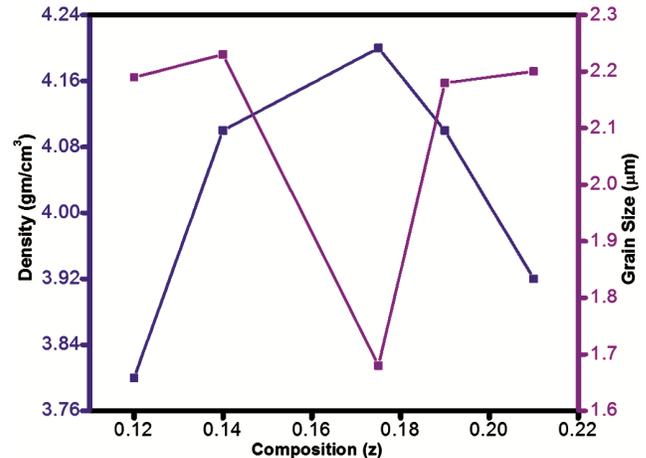


Fig. 5 — Variation of density and grain size with composition (z), in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) ceramics.

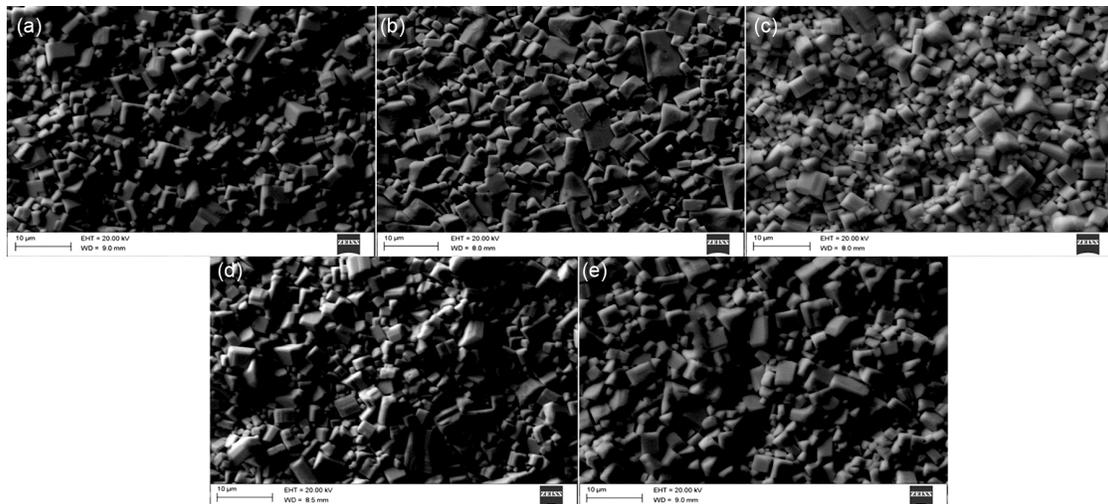


Fig. 4 — The SEM images of $(\text{Na}_{1-z}\text{K}_z\text{NbO}_3)$ ($0.120 \leq z \leq 0.210$) ceramics, (a) $z = 0.120$, (b) $z = 0.140$, (c) $z = 0.175$, (d) $z = 0.190$, (e) $z = 0.210$, respectively.

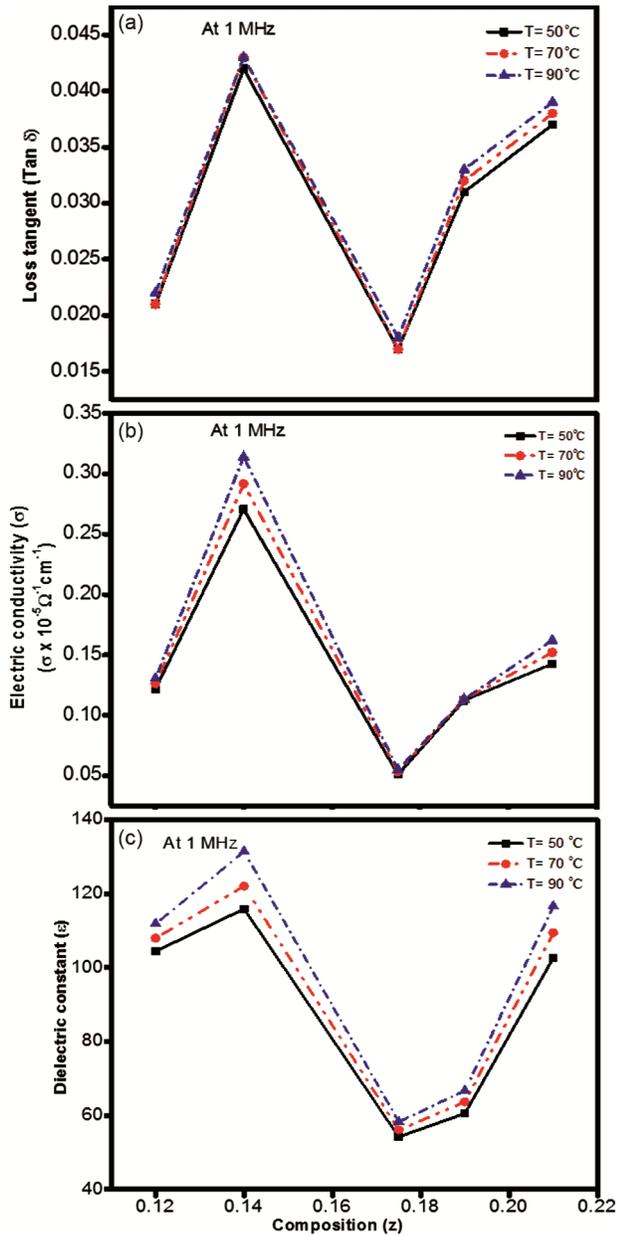


Fig. 6 — (a) Composition dependent loss tangent ($\tan \delta$) of NKN ceramics, at different temperatures. (b) Composition dependent electric conductivity (σ) of NKN ceramics, at different temperatures. (c) Composition dependent dielectric constant of NKN ceramics, at different temperatures.

thermal properties^{22,25,30}. The observed minimum values of dielectric properties at all the measured frequencies, and break in the X-ray diffraction peak shifting patterns of the prepared compositions show an anomalous behavior, near $z = 0.175$, in the compositional range of the $(\text{Na}_{1-z}\text{K}_z\text{NbO}_3)$ ($0.120 \leq z \leq 0.210$) ceramics. Additionally, smaller grains are related to lower electrical permittivity values. However, in this instance, the composition-dependent

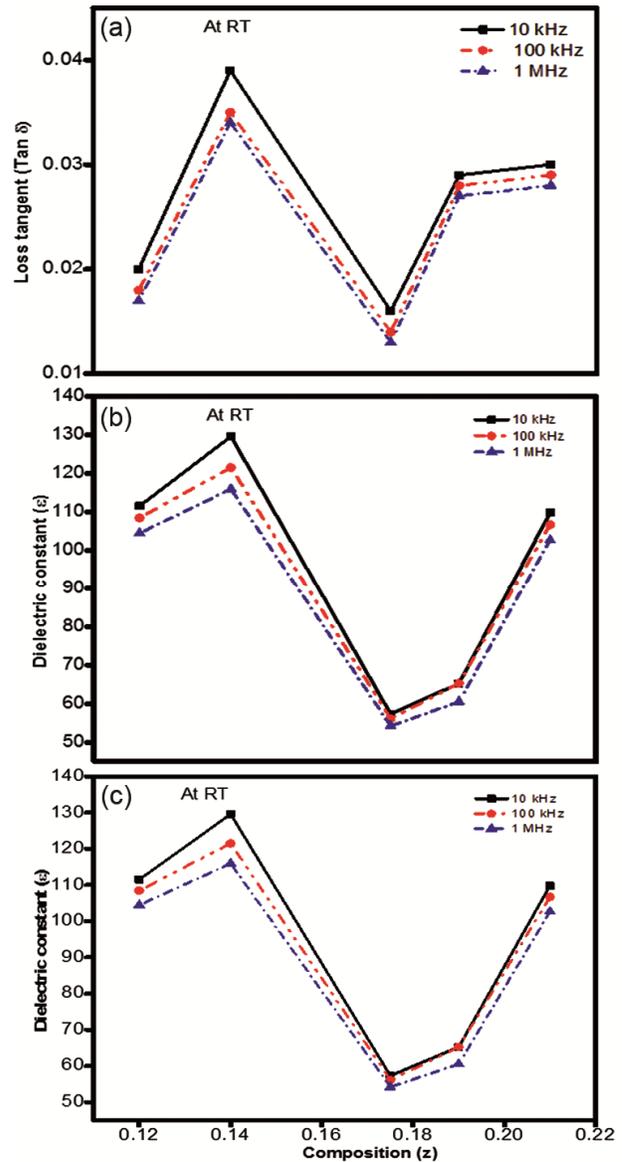


Fig. 7 — (a) Composition dependent loss tangent ($\tan \delta$) of NKN ceramics, at different frequencies. (b) Composition dependent electric conductivity (σ) of NKN ceramics, at different frequencies. (c) Composition dependent dielectric constant of NKN ceramics, at different frequencies.

anomalous behavior, close to $z = 0.175$, has a greater influence than grain size. The effect of MPB is confirmed from XRD data, SEM analysis and composition-dependent dielectric properties. For the prepared compositions with $z = 0.175$, the change in lattice size, lowest grain size, dielectric properties (ϵ , $\tan \delta$ and σ) and highest density followed by a rise in c/a ratio were exhibiting abnormal behavior which shows a morphotropic phase transitions.

For all the prepared compositions, P-E (polarization vs. electric field) hysteresis loops were

taken at RT (room temperature). The measurements were obtained with an input triangular wave, at an electric field 12.16 kV/cm. Fig. 8 shows the plots of P-E hysteresis loops for the prepared compositions of NKN ceramics. The S-E [strain (%) vs. electric field] plots of the prepared NKN samples have been shown in Fig. 9. The obtained butterfly loops exhibit good ferroelectric behavior of the prepared NKN ceramics³¹.

Maximum strain (%) was found for the sample with $z = 0.175$, among the prepared samples. With the help of the eq. $d_{33}^* = S_{\max}/E_{\max}$ ³², i.e., the ratio of strain and electric field, the piezoelectric constant was calculated for the prepared NKN samples. The maximum strain was calculated from the butterfly plot, and the applied electric field was taken at that corresponding strain. Fig. 10 exhibits the variation of the piezoelectric constant (d_{33}^*) with sample (z).

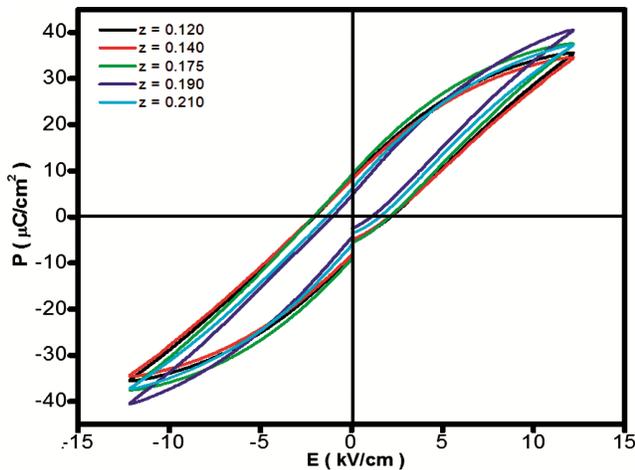


Fig. 8 — Polarization vs. electric field (P-E) hysteresis loops of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$, ($0.120 \leq z \leq 0.210$) ceramics.

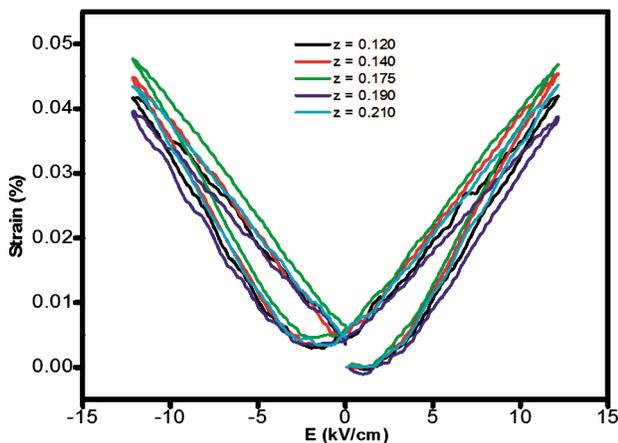


Fig. 9 — Plot of strain (%) with electric field, in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$, ($0.120 \leq z \leq 0.210$) ceramics.

Among the prepared compositions, the highest value (386 pm/V) of piezoelectric constant (d_{33}^*) was observed for the compositions with $z = 0.175$. Researchers found similar findings near the MPB region³³. At composition $z = 0.175$, the obtained distinct greater deformation in piezoelectric constant (d_{33}^*), polarization and strain (%), may be attributed to the change in lattice, near the MPB. Fig. 11 indicates the plots of coercive field (E_c) and remnant polarization (P_r) with variation in composition (z). For the compositions with $x = 0.175$, maximum polarization ($P_r = 9.73 \mu\text{C}/\text{cm}^2$) and coercive field ($E_c = 2.18 \text{ kV}/\text{cm}$) were observed, among the prepared compositions. These findings could be explained by the change in the lattice in the prepared NKN ceramics, at $x = 0.175$, close to the MPB^{34,35}. The processing conditions³⁶, as well as the purity and grain size of the starting materials^{22,37}, have a significant impact on the internal stress, defects, and morphology

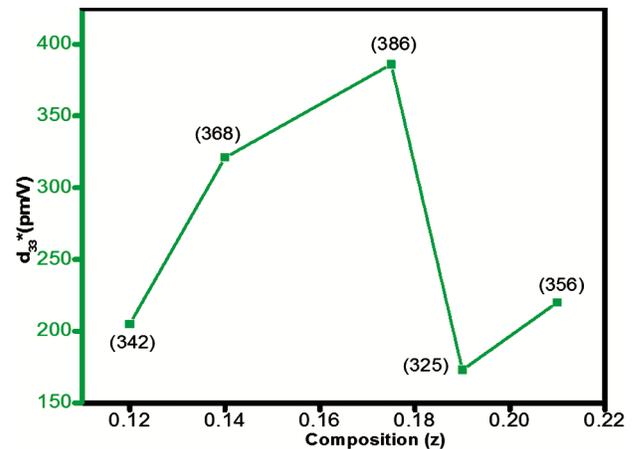


Fig. 10 — Plot of converse piezoelectric constant (d_{33}^*) with sample (z), in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$, ($0.120 \leq z \leq 0.210$) ceramics.

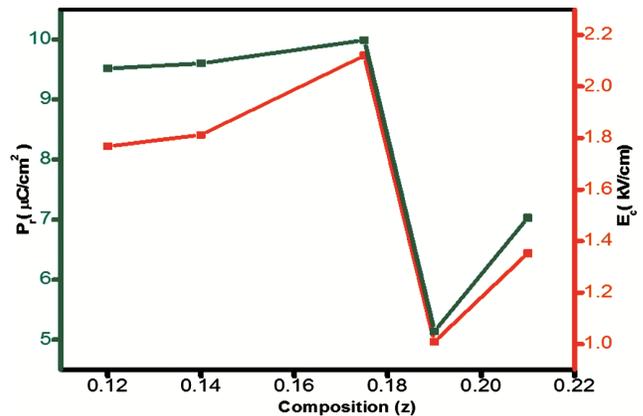


Fig. 11 — Variations of coercive field (E_c) and remnant polarization (P_r) with sample (z), in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$, ($0.120 \leq z \leq 0.210$) ceramics.

of ceramics. The crystallographic details, density and grain size of the prepared compositions can significantly influence the electrical properties^{22,38,39}. Low polarization in the ferroelectrics is a result of the grain boundary's low permittivity. Additionally, the space charges that are present at the grain boundary may contribute to the exclusion of polarization charges on the grain surface and result in polarization discontinuity. In the smaller grain ceramics, a depolarization field consequently manifests, and the polarization further decreases.

The variations of coercive field (E_c) and remnant polarization (P_r) with sample (z) in $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics depend on the crystal structure and polarization of the material, which are affected by the potassium content. As the potassium content (z) increases, the crystal structure of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics undergoes changes from tetragonal to cubic, which affects the polarization and domain structure of the material. The coercive field (E_c) and remnant polarization (P_r) are two important parameters that characterize the electric properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics. The density of the prepared NKN ceramics has been found to increase with increasing Na component (z), reaching its peak at $z = 0.175$ and decreasing with a further increase in z . In contrast, the grain size was found to have the opposite variation. It has been observed that grain size rises with rising in z , showing lowest at $z = 0.175$ and increasing with further increase in z . The grain boundaries can increase or decrease depending on the ceramics' grain size; smaller grains produce fewer grain boundaries and vice versa. For the composition with $z = 0.175$, the reduction in the polarization of the NKN ceramics would be caused by the increased number of grain boundaries which may be due to the observed minimum grain size. However, in the present case, it has been discovered that the sample with $x = 0.175$ had the highest observed remnant polarization (P_r) and coercive field (E_c), indicating that these parameters are composition-dependent rather than related to the grain size or density of NKN ceramics. In a previous communication, a similar result was reported regarding the dielectric and structural properties of NKN ceramics²¹.

4 Conclusions

In conclusion, structural and electrical properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) ceramics were observed. The composition-dependent dielectric and

piezoelectric properties of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ($0.120 \leq z \leq 0.210$) ceramics were discussed. For the prepared $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics, a continuous XRD peaks shifting pattern was observed with varying z , with a break for the sample with $z = 0.175$. The dielectric measurements show a minimal loss for all the prepared compositions of $\text{Na}_{1-z}\text{K}_z\text{NbO}_3$ ceramics. At RT, the dielectric constant was found to be lowest for the compositions with $z = 0.175$ at all the measured frequencies among the prepared compositions. Also, the maximum piezoelectric coefficient was for the sample with $z = 0.175$ among the prepared samples. Therefore, from the above discussion, anomalous structural and electrical properties were found for the sample with $z = 0.175$ among the prepared samples. The observed dielectric measurements of the NKN ceramics will extend the possibilities for using this compound in various energy storage applications.

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