Idiosyncratic behaviour of $(\text{Na}_{0.495}\text{K}_{0.455}\text{Li}_{0.05})(\text{Nb}_{0.95}\text{Ta}_{0.05})\text{O}_3\text{--La}_2\text{O}_3$ ceramics: Synergistically improved thermal stability, ageing, and fatigue properties

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Abstract: La_2O_3 doped $(\text{Na}_{0.495}\text{K}_{0.455}\text{Li}_{0.05})(\text{Nb}_{0.95}\text{Ta}_{0.05})\text{O}_3$ ceramics are prepared using modified milling process, and the influences of La_2O_3 on ferroelectric behaviour, ageing characteristics, thermal stability, electrical stability, crystal structure, microstructure, dielectric and piezoelectric properties were reported. La_2O_3 addition improved the ferroelectric characteristic substantially, and obtained remnant polarization (P_r) and maximum strain ($S_{\max}$) around 34.3 C/cm^2 and 0.13% respectively. La_2O_3 doped ceramics improved the thermal stability and were stable up to 180°C compared to undoped ceramics (120°C). The Rietveld refinement along with the high-temperature X-ray diffraction studies suggested the presence of monoclinic phase in La doped compositions, which is responsible for their idiosyncratic behaviour. The maximum values were obtained around 179 pC/N and 0.385 for piezoelectric constant (d_{33}) and electromechanical coupling factor (k_p) respectively in La_2O_3 doped samples ($0.02 \text{ wt}\%$), which also exhibited the lowest ageing rate and stable electrical fatigue behaviour.

Keywords: dielectric; fatigue; piezoelectric; thermal stability

1 Introduction

The ferroelectric piezoceramics are widely used in various applications such as FeRAM, hydrophones, SONARs, filters, medical diagnostic devices, ultrasonic motors, and buzzers [1]. In particular, FeRAM applications are desired to have high polarization, low coercive field, low ageing rate, and good electrical fatigue behaviour along with high thermal stability. Mostly, lead-based

piezoceramics are used for these applications. However, these ceramics contained more than 60% lead and resulted in toxicity to the environment and human health. Therefore, lead-free piezoceramics are explored worldwide as an alternative to replace lead-based piezoceramics with excellent piezoelectric properties.

Among various lead-free ceramic materials, sodium potassium niobate $(\text{Na,K})\text{NbO}_3$ (NKN) based ceramic is emerged as an important alternative piezoceramic. Also, major breakthroughs of piezoelectric properties were accomplished in textured NKN based ceramics by Saito *et al.* [2] using RTTG method. It drew a lot of attention and motivated researchers to study the NKN

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system more extensively. Lithium antimony (V) oxide (LiSbO_3) [3,4], lithium niobate (LiNbO_3) [5], lithium tantalum oxide (LiTaO_3) [6,7], and their combinations [8] are commonly used to enhance piezoelectric properties. Mostly, lithium (Li) and tantalum (Ta) co-doped compositions were explored in NKN ceramics and reasonably good properties were achieved, which were often modified with other dopants to further improve the properties of interest [9–18]. Similarly, lanthanum (La) modified lead zirconium titanate (PZT) based composition was reported and La was considered as a good A-site donor dopant [19–21]. However, elaborate studies on the effect of La in NKN, and Li and Ta co-doped NKN system have not conceded. Only a few studies are available on La doping [22–25]. Therefore, we attempted to fill that void by extensively exploring the ferroelectric, crystal structure, and piezoelectric properties by adding La in Li and Ta co-doped NKN system.

In this article, La_2O_3 (0–0.04 wt%) doped $(\text{Na}_{0.495}\text{K}_{0.455}\text{Li}_{0.05})(\text{Nb}_{0.95}\text{Ta}_{0.05})\text{O}_3$ ceramics are prepared using modified milling process. The impacts of La_2O_3 addition on ferroelectric, ageing characteristics, thermal stability, electrical stability, and piezoelectric properties are investigated thoroughly. Ferroelectric characteristic was measured and substantial improvement was shown in remnant polarization ($P_r \approx 34.3 \text{ C/cm}^2$) and maximum strain ($S_{\text{max}} \approx 0.13\%$). The thermal stability study was carried out and the improvement was observed at around 60°C . High-temperature X-ray diffraction and their Rietveld refinement were conducted and suggested the presence of monoclinic phase in La_2O_3 doped compositions, which is responsible for idiosyncratic behaviour.

2 Experimental

Ceramics with the composition $(\text{Na}_{0.495}\text{K}_{0.455}\text{Li}_{0.05})(\text{Nb}_{0.95}\text{Ta}_{0.05})\text{O}_3 + x\text{wt}\%\text{La}_2\text{O}_3$ ($x = 0.00, 0.01, 0.02, 0.03, 0.04$), henceforth referred to as L0, L1, L2, L3, and L4, respectively, were prepared by conventional mixed oxide method. Stoichiometric amounts of La_2O_3 , Na_2CO_3 , K_2CO_3 , Li_2CO_3 , Nb_2O_5 , and Ta_2O_5 were mixed with ethanol in polyurethane jar, milled for 7 h in the conventional roller mill using the zirconia media, and then vibrated for 1 h by keeping in horizontal position on the vibration bench. The cycle was repeated for three times. Slurry was dried and calcined at 860°C

for 5 h. Calcined powder was re-milled, dried and granulated using polyvinyl alcohol as binder and compacted into disc shape. Discs were sintered at 1140°C (air atmosphere) for 3 h using inverted crucible technique. Sintered discs were lapped (diameter 11 mm and thickness 1.1 mm) and electroded with silver paste using screen-printing process. The electroded samples were poled under 3 kV/mm DC field using silicon oil bath at 120°C for 1 h.

The crystal structure of sintered samples at different temperatures was determined using Bruker AXS D8 advance X-ray diffractometer (XRD). The Rietveld refinement method was employed using TOPAS 4.2 analysis tool for phase determination and structural refinement. Dielectric properties as a function of temperature were measured at frequency 1 kHz using broadband dielectric spectrometer (Novocontrol). d_{33} meter (Piezotest PM300) and LCR meter (Hioki 3532-50 LCR Hi-tester) were used to measure the piezoelectric constant (d_{33} and d_{31}) and electromechanical properties (k_p and Q_m) respectively. The microstructural studies (fractured surface of ceramic disc) were carried out by field emission scanning electron microscope (Carl Zeiss). Also, transmission electron microscopy (TEM) images were captured using FEI Tecnai G² 30 (USA). The room-temperature polarized Raman spectra were measured using an Ar laser (300 mW, wavelength 514 nm) as excitation source (inVia Reflex, Renishaw, UK). The piezoelectric coefficients of poled samples were measured at desired time and specified temperature for ageing and thermal stability studies respectively. Also, poled samples were subjected to cyclic loading for electrical fatigue test using AixACT Tf analyzer 2000. The same equipment was used for measuring the ferroelectric properties of ceramics.

3 Results and discussion

3.1 Crystal structure analysis

XRD patterns of L0, L1, L2, L3, and L4 samples are shown in Fig. 1. From the XRD profiles, we observed three facts: (i) perovskite nature of all the ceramics, (ii) shift in peaks towards lower values, and (iii) appearance of additional peaks with La doping. Nonexistence of secondary phase confirmed that La is incorporated in the lattice. Also L1, L2, and L3 samples showed higher d values than L0 sample. However, L4 sample's peaks shifted again to nearly original position. This can be

Fig. 1 X-ray diffraction spectra of NKLNT- x La₂O₃ samples (L0, L1, L2, L3, and L4). Inset shows enlarged image of $2\theta = 43^\circ$ – 58° .

explained by considering the liquid phase sintering phenomena, where volatilization of A-site cations (Li^+ , Na^+ , and K^+) generally occurred above 1100°C . Among A-site cations, Li^+ vacancies will be formed readily due to most volatile nature of Li. Thus, Li^+ ($0.92\text{ \AA}$) vacancies were occupied by La^{3+} ($1.36\text{ \AA}$) which increased the volume of lattice and shifted XRD peaks towards lower angle at lower concentration. But at higher concentration of La, Na^+ ($1.39\text{ \AA}$) and K^+ ($1.64\text{ \AA}$) [26] sites were also occupied by La^{3+} and shifted the peaks again towards higher angle. The XRD pattern suggested the phase co-existence, where orthorhombic phase exists along with the tetragonal phase in L0 sample. However, distortion in lattice structure occurred as La got incorporated in the lattice in La doped samples due to unmatched ionic radii. Therefore, existence of two or more phases cannot be ruled out at room temperature.

3.2 Temperature dependent dielectric studies and structural refinement

La doped samples exhibited abnormal dielectric constant with respect to temperature as shown in Figs. 2(a) and 2(b). Three dielectric anomalies are noticed in La doped NKN systems. In general, NKN-based piezoceramics show two phases such as orthorhombic to tetragonal transition ($T_{\text{O-T}}$) at around 200 – 213°C and tetragonal to cubic transition (T_{C}) at around 390 – 400°C [27,28]. However, the existence of three dielectric peaks centred around 27 , 260 , and 400°C in La doped NKLNT reconfirmed the presence of an additional phase in the developed sample. In the case of L0, only two dielectric transitions are present due to orthorhombic to tetragonal (labelled as T1) and tetragonal to cubic (labelled as T_{C}) phase transitions. In general, intrinsic and extrinsic factors determine the profile of the temperature dependent dielectric plot. More intense dielectric transition may be appeared at room temperature due to the collective contribution of the number of phases, increase in number of charge carriers (non-stoichiometric composition), donor dopant nature of La (impart soft characteristics to the ceramic), and decrease in grain size (increases number of grain boundaries). In addition, increase of T_{C} suggested that the addition of lanthanum (La^{3+}) boosted ferroelectric coupling by reducing number of vacancies, which stabilizes the ferroelectric phase [29].

Further, Rietveld analysis was performed using the combination of $\text{Amm}2$ space group (ICSD No. 261355) for orthorhombic structure and $\text{P}4\text{mm}$ space group (ICSD No. 261356) for tetragonal structure. The results of structural refinement are shown in Table 1. The Rietveld

Fig. 2 (a) Plot of temperature dependent dielectric constant and (b) dependence of phase transition on composition for NKLNT- x La₂O₃ ceramics.

Table 1 Rietveld analysis data

L0 $R_{wp} = 6.36$, GOF = 3.06						
	R_{Bragg}	Phase (%)	a (Å)	b (Å)	c (Å)	Vol (Å) ³
<i>Amm2</i>	3.966	55.93	3.96158	5.67741	5.64215	126.9005
<i>P4mm</i>	2.442	44.07	3.946494	—	4.028737	62.747
L1 $R_{wp} = 7.55$, GOF = 3.55						
	R_{Bragg}	Phase (%)	a (Å)	b (Å)	c (Å)	Vol (Å) ³
<i>Amm2</i>	5.320	72.7	4.03995	5.58188	5.58146	125.8647
<i>P4mm</i>	2.210	16.2	3.9527	—	4.0233	62.858
<i>Pm</i>	5.146	11.1	3.9403	4.01331	3.97858	62.916 $\beta = 89.845$
L2 $R_{wp} = 5.74$, GOF = 2.75						
	R_{Bragg}	Phase (%)	a (Å)	b (Å)	c (Å)	Vol (Å) ³
<i>Amm2</i>	2.568	27.5	4.03258	5.5764	5.5707	125.268
<i>P4mm</i>	1.866	28.7	3.94461	—	4.0362	62.803
<i>Pm</i>	2.228	43.8	3.9461	4.0157	3.97892	63.050 $\beta = 90.03$
L3 $R_{wp} = 5.43$, GOF = 2.56						
	R_{Bragg}	Phase (%)	a (Å)	b (Å)	c (Å)	Vol (Å) ³
<i>Amm2</i>	4.116	45.7	4.03812	5.58109	5.58090	125.78
<i>Pm</i>	2.330	54.3	3.94841	4.01118	3.99059	63.202 $\beta = 90.196$
L4 $R_{wp} = 4.46$, GOF = 2.14						
	R_{Bragg}	Phase (%)	a (Å)	b (Å)	c (Å)	Vol (Å) ³
<i>Amm2</i>	3.296	27.3	4.02915	5.57290	5.57114	125.095
<i>Pm</i>	2.375	72.7	3.94448	4.01407	3.98253	63.057 $\beta = 89.99$

refinement of the room-temperature XRD data of L0 sample suggested a polymorphic phase boundary (PPB) between orthorhombic (*Amm2*) and tetragonal (*P4mm*) having 56% and 44% phase fractions respectively, and validated the assumption. In the case of L1 to L4, initial two peaks are labelled as T1 and T2. The third peak is appeared due to transition between ferroelectric tetragonal and paraelectric cubic phase (T_C), which is a norm for NKN-based system. The structural refinement indicated the presence of monoclinic (*Pm*) phase [30] in La doped samples. L1 and L2 samples showed three phases at room temperature, where monoclinic (*Pm*) phase co-existed with orthorhombic (*Amm2*) and tetragonal (*P4mm*) phases. However, the percentage of tetragonal phase decreased gradually by forming a new room temperature PPB between monoclinic and orthorhombic phases. The fraction of monoclinic phase increased from 11% in L1 to 72% in L4 sample due to the incorporation of La, which induced distortion in lattice structure. Further, polymorphic phase transitions at T1 and T2 are corroborated by conducting the temperature dependent X-ray diffraction study.

3.3 Temperature dependent XRD study

The X-ray profiles are collected near to polymorphic phase transition temperatures for L3 sample and shown

in Fig. 3. At 30 °C, the L3 composition had a PPB between monoclinic and orthorhombic phases (Table 1). However, the XRD profile at 60 °C showed the existence of PPB between orthorhombic and tetragonal phases. Further, the X-ray profile collected at 260 °C corresponds to tetragonal symmetry [31]. Therefore, the dielectric transition near to 260 °C (T2) attributed as orthorhombic to tetragonal phase transition. Hence, relating current findings of Rietveld results with dielectric studies, it can be concluded that the presence of monoclinic to tetragonal ferroelectric phase transition is near room temperature (T1). It explained clearly the order of phase transition in La doped NKLNT (M–T, O–T, and T–C) with respect to temperature.

3.4 Microstructural and EDX studies

Surface morphologies of the fractured surface of ceramics sintered at 1140 °C are shown in Fig. 4. Addition of La significantly inhibited the formation of larger grains and promoted the growth of smaller grains in NKLNT system. Similar type of observation was also noticed in PZT-based systems [20,32]. Bi-modal grain distribution was observed in all La doped samples having smaller grains ranging from 0.3 to 1.0 μm and larger grains ranging from 1.1 to 3.2 μm compared to un-doped one, where grain sizes were viewed in the range of

Fig. 3 Temperature dependent X-ray diffraction spectra of L3.

11–13 μm . The uneven grain growth might appear due to the combined effect of liquid phase sintering (promoting densification and large grain growth) and A-site cation vacancy formation (promoting small grain growth and inhibits densification) [22]. Also, elemental study (EDX) measured the elemental composition of the samples and is given in Table 2. The composition percentage of La was not appeared in L1 and L2 samples due to

very few amount of La present.

Figure 5 illustrates the TEM and HRTEM images of L0 and L2 samples. Inset in the image showed the calculated d -spacing of the samples and it corresponded to $\{001\}$ set of plane. Also, increase of d -spacing was observed in L2, which supported the XRD results. However, it is not possible to deduce the correct size of particles due to agglomeration.

3.5 Dielectric and piezoelectric properties

Figure 6(a) shows the dependence of piezoelectric constants (d_{33} and d_{31}) and dielectric constant (K^T_{33}), and Fig. 6(b) shows electromechanical coupling constant (k_p) and mechanical quality factor (Q_m) as a function of lanthanum content. L2 sample exhibited the highest value of k_p , d_{33} , and d_{31} . Dielectric constant shows a marked increase from 710 to 967 with lanthanum addition due to decreased grain size and increased charge carriers. However, addition of La showed slight improvement in d_{33} , which was also seen in previous studies [22,24].

Fig. 4 SEM micrographs of samples sintered at 1140 °C: (a) L0, (b) L1, (c) L2, (d) L3, and (e) L4.

Table 2 Elemental composition of NKLNT- $x\text{La}_2\text{O}_3$ samples

Element	L0		L1		L2		L3		L4	
	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)
O K	30.72	64.82	31.46	65.67	30.86	65.01	30.25	64.40	30.59	64.63
Na K	5.83	8.56	5.59	8.12	5.67	8.31	5.62	8.33	5.88	8.65
K K	7.95	6.86	8.06	6.89	8.10	6.99	8.28	7.22	8.03	6.94
Nb L	53.20	19.33	52.63	18.91	53.14	19.28	53.46	19.60	53.15	19.34
La L	—	—	0.01	0.00	0.02	0.00	0.05	0.01	0.07	0.02
Ta M	2.30	0.43	2.26	0.42	2.21	0.41	2.34	0.44	2.28	0.43
Total	100.00		100.00		100.00		100.00		100.00

Fig. 5 (a) TEM and (b) HRTEM images of L0, and (c) TEM and (d) HRTEM images of L2.

Fig. 6 Dielectric and piezoelectric properties of NKLNT- x La₂O₃ ceramics as a function of lanthanum content: (a) d_{33} , K_{31}^T , and d_{31} , and (b) k_p and Q_m .

3.6 Ferroelectric properties

The ferroelectric loops of the samples are shown in Fig. 7(a). Addition of La has affected the ferroelectric properties significantly in NKLNT samples. As shown in Fig. 7(b), the remnant polarization (P_r) value increased gradually from 21.5 $\mu\text{C}/\text{cm}^2$ in L0 sample to 34.3 $\mu\text{C}/\text{cm}^2$ in L2. As La acted as donor dopant, it prevented the formation of oxygen vacancies which reduced the pinning effect and increased the ferroelastic nature of domains. Thus, La addition made the domain alignment along the poling direction easier. At higher La doping, the decrease of P_r value indicated the loss of ferroelectric characteristic which was also reflected in d_{33} value. It can be attributed to the distortion of lattice structure which reduced the overlap

between oxygen p-orbital and B-site cation d-orbital [33]. However, L4 sample displayed lossy capacitor type behaviour and the loop was not saturated due to the presence of higher number of charge carriers, which resulted in higher electrical conductivity and leakage characteristic (Fig. 8).

3.7 Raman studies

ABO₃ type perovskite is formed by covalently bonded BO₆ octahedra and A-site cations at the corner of a cubic lattice. The region below wavenumber 200 cm^{-1} involves translational modes of ionic A-site cations and rotational modes of BO₆ octahedra, and above 200 cm^{-1} holds the stretching and bending modes of BO₆ octahedra [5,34]. In the case of NKN-based system, the vibrations of the BO₆ octahedra consist of $\nu_1(1A_{1g}) +$

Fig. 7 (a) Ferroelectric hysteresis loop and (b) remnant polarization (P_r) and coercive field (E_c).

Fig. 8 Voltage and leakage current characteristics as a function of La content for NKLNT- x La₂O₃ ceramics.

$\nu_2(1E_g) + \nu_3$ and $\nu_4(2F_{1u}) + \nu_5(F_{2g}) + \nu_6(F_{2u})$. Out of these vibrations $\nu_1(1A_{1g}) + \nu_2(1E_g) + \nu_3(F_{1u})$ are arisen due to the stretching modes and $\nu_4(F_{1u}) + \nu_5(F_{2g}) + \nu_6(F_{2u})$ are appeared due to the bending modes. In Raman spectroscopy, scattering intensity is proportional to the square of the induced dipole moment. In Fig. 9, it can be seen that the incorporation of La^{3+} significantly affected bands below 200 cm^{-1} , which is assigned as translational modes of A-site cations and rotational modes of the BO_6 octahedra. As La^{3+} has very high atomic mass as compared to $\text{Li}^+/\text{K}^+/\text{Na}^+$ cations, the intensity of band below 200 cm^{-1} decreased rapidly and the band almost vanished in L4. The frequency bands above 200 cm^{-1} are dominated by modes arising due to BO_6 octahedra and expected to be unaffected by

Fig. 9 Raman spectra as a function of La content for NKLNT- x La₂O₃ ceramics.

the type of A-site cations. However, the nature of A-site cations influences these bands by the other indirect factors such as steric hindrances and bonding between atoms. Also, the intensities of bands above 200 cm^{-1} reduced gradually from L0 to L4 due to the distortion in octahedral symmetry which altered the polarizability of BO_6 octahedra. Broadness of bands occurred due to the disorderness at A-site, random orientation of grains, and overlapping of Raman bands [35]. In addition, incorporation of La^{3+} noticeable shifted the stretching vibration $\nu_1(1A_{1g})$ and bending vibration $\nu_5(F_{2g})$ modes towards lower frequency side due to weakening of bonding strength.

3.8 Ageing, thermal stability, and electrical fatigue studies

Ageing is defined as the degradation of properties with time without influence of any external factors. But, degradation in switchable polarization when subjected

to bipolar electrical switching cycles is referred as fatigue. The behavior of the developed ceramics on ageing and under cyclic electrical loading is shown in Fig. 10. The underlying mechanism for both ageing and fatigue is dependent on the presence of space charge and oxygen vacancies. Scott *et al.* [36] proposed the migration of oxygen from the lattice lead to accumulation of space charge which is the primary reason for fatigue. Also, Nuffer *et al.* [37,38] demonstrated the formation of microscopic defect agglomerates during bipolar switching cycles which reduces the mobility of domain wall using pinning mechanism. Further reduction of remnant polarization occurred due to the increase in concentration of oxygen vacancies [39]. It can be seen that the low concentration of La stabilized the polarization state and reduced the rate of ageing. Again, the samples with lower ageing rates are shown stable under cyclic electrical loading. This can be explained by considering the dependency nature of both ageing and electrical fatigue on the number of charge carriers in the sample. In both the processes, redistribution of charge carriers occurred, which pinned the domain walls and reduced the switchable polarization

[40]. Therefore, higher number of charge carriers lowered the polarizability and boosted the ageing rate. At lower La concentration number of oxygen vacancies reduced which weakened the pinning of domains, and resulted increase in the fatigue resistance and ageing characteristics. Further increase in the La content raised number of free charge carriers due to the presence of uncompensated charges. Therefore, L3 and L4 samples were found to have inferior fatigue and ageing behavior.

In addition, the thermal stability of the developed ceramics was investigated and shown in Fig. 11. The addition of La improved the thermal stability of the samples. L0 sample showed stability only up to 120 °C, whereas La doped samples (L1 and L2) exhibited stable piezoelectric property even at 180 °C. The temperature dependent ferroelectric behaviour also showed similar characteristics as shown in Fig. 12. Strain values of 0.17% at 3 kV/mm for NKN-based fibers, 0.13% at 3 kV/mm and 0.16% at 6 kV/mm for NKN-based ceramics are reported in literatures [41–43]. The maximum strain of 0.13% was achieved in L2 sample at 3.5 kV/mm and 0.06% in L0. It can be correlated to the co-existence of multiple phases and

Fig. 10 (a) Ageing and (b) fatigue behaviour of NKLNT- x La₂O₃ ceramics.

Fig. 11 Thermal stability studies of NKLNT- x La₂O₃ ceramics.

ease switching and back-switching of non-180° domains. However, thermal instability of the domains and ferroelastic behaviour almost vanished in the L4 sample at elevated temperature. The unsymmetrical nature of the loops may be arisen due to the superposition of applied field with the localized internal field at grain boundaries which enhanced strain in one particular direction and restricted the movement of domain wall in other direction [44]. The asymmetry factor “ γ ” can be calculated as [45]:

$$\gamma = \frac{S_{33}^+ - S_{33}^-}{S_{33}^+ + S_{33}^-}$$

Fig. 12 (a–e) Temperature dependent ferroelastic properties and (f) asymmetry factor γ of NKLNT- x La₂O₃ ceramics.

where S_{33}^+ and S_{33}^- represented the strain values of positive and negative electric field side respectively in the ferroelastic loop. The “ γ ” values for L0 to L3 samples at different temperatures are illustrated in Fig. 12(f). The value of γ near to zero depicts symmetrical structure of strain loop, while negative and positive values represent corresponding bias. The L0 sample displayed symmetrical loop with slight positive bias while La doped NKN samples exhibited asymmetrical negative biased behaviour.

4 Conclusions

Ceramics with composition (Na_{0.495}K_{0.455}Li_{0.05})(Nb_{0.95}Ta_{0.05})O₃+ x wt%La₂O₃ were synthesized successfully using solid state reaction method. The La doped samples showed enhanced dielectric and ferroelectric properties. X-ray diffraction studies confirmed that the doping induced monoclinic phase in the samples and tri-phasic system existed at room temperature in La doped composition ($x = 0.02$). Our studies revealed that even addition of very few percentage of La₂O₃ improved dielectric, thermal stability, ageing and fatigue characteristic of NKLNT system. However, piezoelectric properties are not significantly improved by doping. Therefore, the achieved characteristics of La₂O₃ doped NKLNT ceramics made it more advantageous for FeRAM applications.

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