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BaTiO₃–(Na_{0.5}Bi_{0.5})TiO₃ Ceramic Materials Prepared via Multiple Design Strategies with Improved Energy Storage

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Abstract

The investigation of environmentally friendly, Pb-free ceramic dielectric materials with excellent energy storage capability represents a fundamental yet challenging research direction for the development of next-generation high-power capacitors. In this study, linear dielectric Ca_{0.7}La_{0.2}(Mg_{1/3}Nb_{2/3})O₃ was added into [0.65BaTiO₃–0.35(Na_{0.5}Bi_{0.5})TiO₃] to form a solid solution. The introduction of Ca_{0.7}La_{0.2}(Mg_{1/3}Nb_{2/3})O₃ modified the crystal structure, enhanced insulation performance and breakdown strength, and reduced hysteresis loss. These improvements collectively contributed to higher energy storage density and efficiency (η). The ceramic pellet with the optimal 10 mol% Ca_{0.7}La_{0.2}(Mg_{1/3}Nb_{2/3})O₃ demonstrated a higher retrievable energy density ($\sim 3.40 \text{ J cm}^{-3}$) and efficiency ($\sim 81\%$) at a breakdown strength of 340 kV cm^{-1} compared to BaTiO₃-based ferroelectric ceramics. The sample also exhibited good stability across a temperature range of 30–90 °C and a frequency range of 0.5–300 Hz. Thus, the as-prepared ceramics sample exhibited significant potential for pulsed power device applications.

Keywords: barium titanate; lead-free; ceramic capacitors; Ca_{0.7}La_{0.2}(Mg_{1/3}Nb_{2/3})O₃; energy storage



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1. Introduction

The rapid rise in global energy consumption over recent decades has driven the demand for renewable, efficient, and environmentally friendly energy storage devices [1–4]. Dielectric capacitors, as essential components of modern electronic systems, can store energy electrostatically and are widely employed for energy storage due to their ability to rapidly and reliably store and release energy [5,6]. When an electric field is applied, the positive and negative charges inside the material's atomic structure are slightly pulled apart. Energy is stored within this induced polarization. After the electric field is removed, the material releases this stored energy as it discharges. However, their relatively low energy storage density remains a significant limitation for pulsed power applications. Consequently, there is an urgent need for environmentally friendly (lead-free) energy

storage materials that have both a high recoverable energy storage density (W_{rec}) and high energy storage efficiency (η) [7,8]. Generally, the total energy storage density (W), W_{rec} , and η are expressed as Equations (1)–(3) [9]:

$$W = \int_0^{P_{\text{max}}} E dP \quad (1)$$

$$W_{\text{rec}} = \int_{P_r}^{P_{\text{max}}} E dP \quad (2)$$

$$\eta = \frac{W_{\text{rec}}}{W} \quad (3)$$

where P_{max} , P_r , and P indicate the maximum, remnant, and normal polarization, respectively; E denotes the applied electric field. W_{rec} can be improved by simultaneously achieving ΔP ($P_{\text{max}} - P_r$) and a high E .

Energy storage ceramic materials with dielectric properties are generally classified into four categories: ferroelectric (FEc), linear dielectric, antiferroelectric (AFEc), and relaxor ferroelectric (RFe) ceramics [10,11]. RFe and AFEc ceramics are regarded as excellent energy storage materials owing to their high P_{max} . However, many AFEc systems contain expensive noble metals such as silver or environmentally hazardous elements like lead [12–14]. RFe materials are particularly attractive because they contain polar nano-regions (PNRs). These PNRs enable rapid domain switching under an applied E , resulting in a small P_r . This, combined with their high P_{max} values, allows them to simultaneously achieve excellent W_{rec} and η . Consequently, lead-free RFe materials are gaining significant attention for both environmental and economic reasons.

Barium titanate (BaTiO_3 , BT), the first discovered perovskite-type piezoelectric material, has been extensively utilized in commercial multilayer ceramic capacitors [15]. However, the low intrinsic breakdown electric field (E_b) and high coercivity field (E_c) of these devices limit a desirable W value. The fast response of polar nanoregions (PNRs) reduces E_c in the presence of electric fields, which substantially improves the efficiency (η) and energy density (W) [16–19]. Lv et al. [20] added 40 mol% of strontium bismuth titanate ($\text{Sr}_{0.7}\text{Bi}_{0.2}\text{TiO}_3$) to a 0.6 [0.94 sodium bismuth titanate ($\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$, NBT)–0.06 BT] system. The macroscopic domain was disrupted and P_r and E_c were reduced, which enhanced W and η . In addition, Yuan et al. [21] enhanced the threshold field and decreased E_c for introducing a long-range order in a BT–bismuth magnesium zirconate [$\text{Bi}(\text{Mg}_{0.5}\text{Zr}_{0.5})\text{O}_3$] system by forming PNRs. While various modification strategies exist, a long-term challenge remains in further improving the energy storage capacity of these ceramics [16].

Sodium bismuth titanate ($\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$, NBT) is a lead-free ceramic material for energy storage. Its high P_{max} ($>40 \mu\text{C cm}^{-2}$) arises from the hybridization between the O^{2-} 2p orbitals and the Bi^{3+} 6s² lone pair electrons [22]. Incorporating NBT into BaTiO_3 effectively enhances polarization and stabilizes dielectric performance. This additional O 2p–Bi 6s orbital hybridization compensates for the polarization deficiency in BT [23]. As a result, 0.65BT–0.35NBT-based ceramics have been extensively researched for their excellent W . High energy storage performance has been reported for $(1-x)(0.65\text{BT}–0.35\text{NBT})–x\text{Bi}(\text{Mg}_{2/3}\text{Nb}_{1/3})\text{O}_3$ materials with η of 90.3% and W_{rec} of 1.6 J cm^{-3} [24]. Similarly, Dai et al. [25] et al. achieved η of 90.18% and W_{rec} of 2.02 J cm^{-3} at 206 kV cm^{-1} by adding $\text{Sr}(\text{Sc}_{0.5}\text{Nb}_{0.5})\text{O}_3$ into 0.65BT–0.35NBT ceramics.

Herein, 0.65BT–0.35NBT was selected as a basic component, while $\text{Ca}_{0.7}\text{La}_{0.2}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (CLMN), a linear dielectric material, was used for doping 0.65BT–0.35NBT. This is a rational, multifunctional design strategy. The effect of CLMN on the energy storage performance of 0.65BT–0.35NBT was investigated. Calcium titanate (CaTiO_3) is a linear dielectric material with a high E_b and negligible P_r . Incorporating CaTiO_3 into ferroelectric matrices to form

solid solutions can enhance W [26,27]. The addition of the rare earth element lanthanum (La) is known to refine grains [28], while the presence of magnesium oxide (MgO) and niobium pentoxide (Nb₂O₅) with their wide bandgaps (E_g , 7.8 eV and 3.4 eV, respectively) improves the material's insulation performance. These improved relaxation behavior, and reduced P_r in CLMN-doped 0.65BT–0.35NBT, optimizing its energy storage properties. Introducing 10 mol% of CLMN into the ceramic significantly enhanced its energy storage performance, with an E_b of 340 kV cm^{−1}, η of ~81%, and a W_{rec} of ~3.40 J cm^{−3}. The resulting ceramic also demonstrated good stability over a temperature range of 30–90 °C and a frequency range of 0.5–300 Hz. This approach provides an effective strategy to optimize the energy storage capability of BT-based ceramics, making them suitable for advanced power systems.

2. Experimental Procedure

(1− x)[0.65BaTiO₃–0.35(Na_{0.5}Bi_{0.5})TiO₃] $-x$ Ca_{0.7}La_{0.2}(Mg_{1/3}Nb_{2/3})O₃ (abbreviated as x CLMN, $x = 0.08, 0.10, 0.12, 0.14$ and 0.16) lead-free ceramic samples were synthesized using high-purity raw materials: bismuth oxide (Bi₂O₃, AR 99%), sodium carbonate (Na₂CO₃, AR 99.8%), barium carbonate (BaCO₃, AR 99%), calcium carbonate (CaCO₃, AR 99%), lanthanum oxide (La₂O₃, metal basis 99.9%), magnesium oxide (MgO, AR 98%), niobium oxide (Nb₂O₅, Metals basis 99.9%) and titanium oxide (TiO₂, AR 99%) using a standard solid-state method. The chemicals were procured from Alladin Scientific, Shanghai, China. The powders and ceramic pellets were prepared according to the procedure described in our previous work [29]. The powders were weighted according to the stoichiometric ratio and ball milled in ethanol for 24 h. After drying, the mixtures were calcined at 900 °C for 4 h in air. The calcined powders were milled again and pressed into disks with 12 mm in diameter and 1.5 mm in thickness using 6 wt% PVA aqueous solution as binder. The green pellets were heated to 600 °C for 6 h to burn out the binder. The pellets were sintered in a covered alumina crucible at 1050–1170 °C for 3 h to obtain high density.

The crystal structures were characterized using a Renishaw Raman spectrometer and X-ray diffraction (XRD) on SmartLab (Rigaku, Tokyo, Japan) instruments under the following conditions: (i) step size, 0.02°; (ii) 2 θ range, 20–80°; and (iii) scanning speed, 2° min^{−1}. Raman spectroscopy (Renishaw InVia Reflex) with a radiation of Ar⁺ laser ($\lambda = 514.5$ nm) was carried out to investigate the structural properties of ceramics at room temperature. The thermally etched surface morphology of the ceramic pellets was examined via scanning electron microscopy (SEM; VEGA3/XUM, TESCAN, Brno, Czechia), and elemental composition was analyzed using energy-dispersive X-ray spectroscopy (EDS) during SEM analysis. Grain size distribution of the samples was determined using Nano Measurer software 1.2. The ceramic samples were manually ground and polished to a thickness of 0.5 mm for electrode fabrication. Silver electrodes were applied and subsequently heated at 550 °C for 30 min. Impedance spectra and dielectric properties were measured using an impedance analyzer (Keysight E4990A, Santa Rosa, CA, USA) at a heating rate of 3 °C min^{−1} over a temperature range of 30–200 °C and at various frequencies. The ferroelectric properties were measured using unipolar triangular waveform at 1 Hz and under varying electric fields using a Premier II ferroelectric tester (Radiant Technologies Inc., New Mexico, NW, USA). The green pellets were polished to an appropriately 0.15 mm thickness prior to testing. Au electrodes with a 3.14 mm² area were sputtered.

3. Results and Discussion

The phase structure of x CLMN samples was analyzed via XRD, and the results are presented in Figure 1. The XRD patterns of the samples exhibited a pure perovskite structure. The absence of secondary phases indicated that Ca, La, Mg, and Nb ions were

successfully incorporated into the BT-NBT lattice, forming a solid solution. With increasing CLMN doping concentration, the symmetry of the (200) diffraction peak at the 2θ value of 45° increased (Figure 1b), suggesting a transition from the tetragonal phase (T , $P4mm$) to the pseudo-cubic phase (PC , $Pm-3m$) [30]. The presence of the K_{a2} reflection, often mistaken for non-cubic distortion, is a key observation [31]. This reflection is typically difficult to resolve in tetragonal or rhombohedral symmetries. The distinct appearance of the (200) peak at low diffraction angles ($<50^\circ$) indicates that non-cubic distortions were negligible within the detection limits of the X-ray apparatus. Further, with increasing CLMN content, the peaks shifted toward higher 2θ values, suggesting a reduction in the lattice constant and unit cell volume. This effect likely arises because the ionic radii of the dopants (Ca^{2+} , Mg^{2+} , Nb^{5+} , and La^{3+}) are smaller than those of the original A-site and B-site ions [23,30].

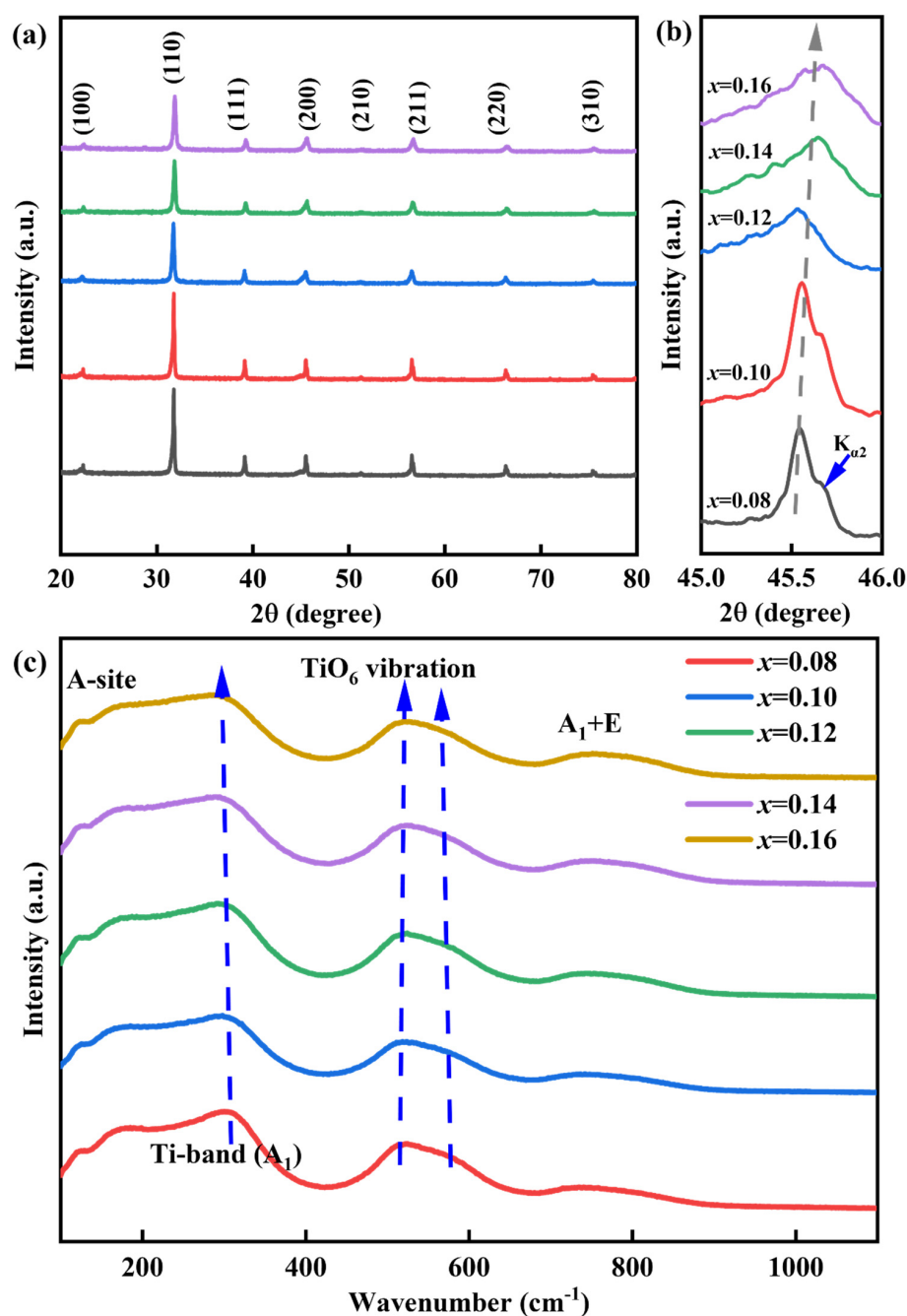


Figure 1. (a) Room-temperature XRD patterns of as-prepared ceramics; (b) the corresponding magnification around $2\theta \sim 45^\circ$; (c) Raman spectra of the x CLMN ceramics.

The structure and phase evolution of x CLMN samples were analyzed via Raman spectroscopy (Figure 1c). BT-based ferroelectric ceramics exhibit four main vibrational modes [30–33], which were also observed in our sample spectra: (i) A-site cations vibrations ($<200\text{ cm}^{-1}$), (ii) B–O bond vibrations ($200\text{--}400\text{ cm}^{-1}$), (iii) BO_6 octahedra vibrations ($400\text{--}650\text{ cm}^{-1}$), and (iv) $A_1 + E$ ($>700\text{ cm}^{-1}$) overlapping bands. The Raman bands at ~ 179 and 120 cm^{-1} correspond to A–O vibrations, confirming the presence of Na^+ , Ba^{2+} , Bi^{3+} , La^{3+} , and Ca^{2+} . Furthermore, the bending vibrational mode (A_1) of the Ti–O bond at $\sim 300\text{ cm}^{-1}$ shifted to a lower wavenumber. With increasing CLMN concentration, the Raman peaks near 300 cm^{-1} became broader and more diffuse. Multiple cations occupy the A-site causing a mismatch in the valency, which induces a high local disorder and reduced unit cell polarity [34,35]. It can be induced that CLMN doping progressively replaces the Ti^{4+} ions, which reduces the material's P [30]. The creation of local nanoregions is proved by the appearance of the Raman peaks at $\sim 750\text{ cm}^{-1}$ [36]. Consequently, adding CLMN breaks down the long-range ferroelectric order, leading to the formation of PNRs [37]. This makes the material exhibit a high P_{max} and a nearly negligible P_r during charging and discharging. The transformation of the Raman peaks from active modes to complex/diffused modes is similar to those of the XRD peaks. This indicates a phase transition from the tetragonal (ferroelectric) state to a pseudo-cubic (relaxor ferroelectric) state.

The thermally etched surface morphology of the ceramic samples was analyzed using SEM (Figure 2). The x CLMN ceramics exhibited a dense microstructure with well-defined grain boundaries and a few pores. Grain-size distributions were calculated using analytical software, and variations in grain size among the as-sintered samples were assessed (Figure 2a₁–f₁). The grain sizes of CLMN-doped ceramics decreased from $1.48\text{ }\mu\text{m}$ to $0.9\text{ }\mu\text{m}$. This reduction is crucial for achieving a high E_b and is closely related to grain-boundary migration and the sintering temperature. Moreover, the ceramic grain size was highly sensitive to the CLMN content. EDS mapping of 0.10CLMN (Figure 2g) revealed compositional homogeneity, with all elements uniformly distributed across the observed region, showing no signs of segregation or aggregation.

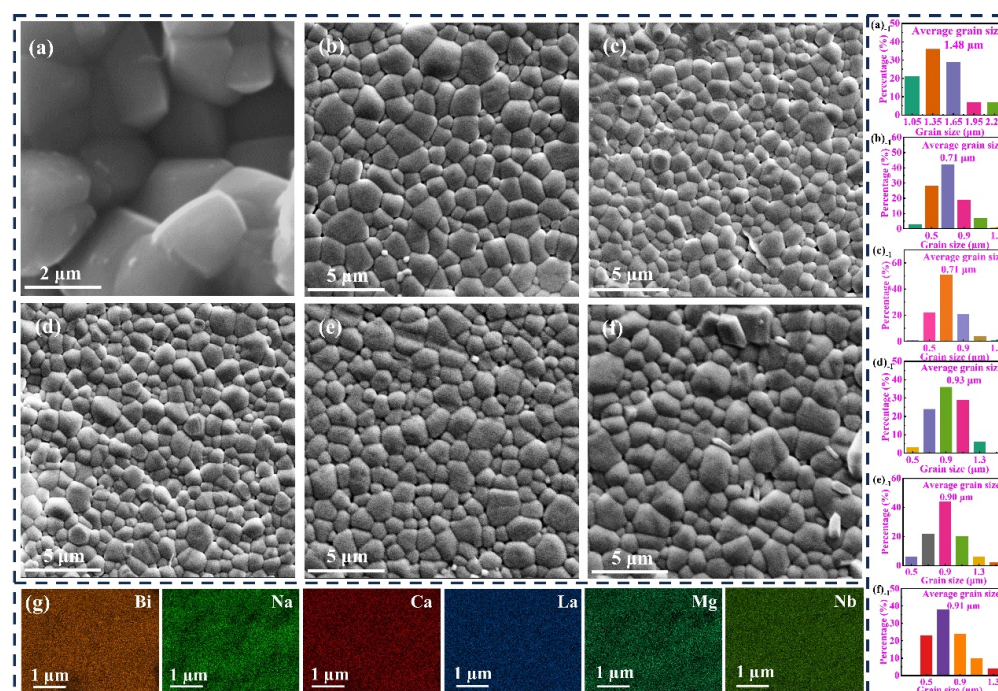


Figure 2. (a–f) SEM micrographs and grain size for ceramics sintered at optimal sintering temperature; (g) SEM-EDS images of 0.1CLMN ceramic.

The behavior of different polar components in a material under an external field with a low alternating current is indicated by the dielectric properties of the material. Based on this phenomenon, the relaxation properties and phase transition of ferroelectrics were investigated. The loss tangent ($\tan \delta$) and dielectric constant (ϵ) of x CLMN were measured as a function of temperature at various frequencies, and the results are shown in Figure 3. For ferroelectric materials, the sharp peaks representing dielectric components are independent of frequencies. However, an increase in the CLMN concentration gradually alters the sharp high ϵ -temperature peak (T_m) to a diffuse and broad peak. This phenomenon is attributed to frequency dispersion, highlighting the existence of pronounced relaxor characteristics [38]. Furthermore, T_m and the maximum dielectric constant (ϵ_m) decreased with increasing x amount. The reduction in ϵ_m is likely due to changes in the lattice constant and A-site ionic radii [39]. The presence of different cations at the A-site compresses the oxygen octahedra and reduces the mobility of B-site cations. Therefore, ϵ and P are lowered. The domain structure might also be involved. An increase in the x content diminishes the phase proportion of the T phase whereas that of the PC phase increases, forming a relaxor ferroelectric phase. The presence of these nanodomain structures might decrease ϵ_m [39]. Notably, T_m shifts to room temperature for samples with $x \geq 0.10$.

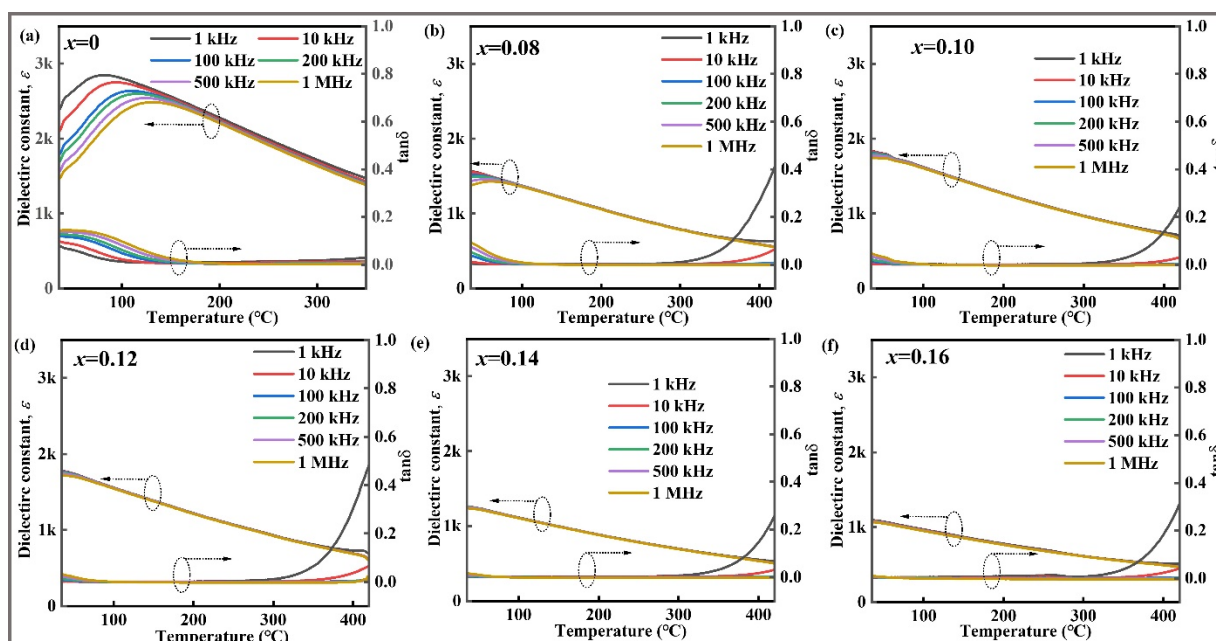


Figure 3. Temperature-dependent dielectric constant and $\tan \delta$ for all ceramics at different frequency: (a) $x = 0$; (b) $x = 0.08$; (c) $x = 0.10$; (d) $x = 0.12$; (e) $x = 0.14$; (f) $x = 0.16$.

The conductivity, dielectric behavior, and relaxation-type characteristics of x CLMN ceramic materials were investigated via impedance spectroscopy. The impedance spectra of the samples from 400 °C to 460 °C in the range of 20–10⁶ Hz are presented in Figure 4. The samples exhibited a semicircular feature in the examined temperature range, likely due to grain boundaries [40]. The impedance data were fitted using a single parallel resistor–capacitor equivalent circuit to extract reliable resistance values for the grain boundary phases. Grain boundary conductivity displayed an evident decrease in impedance at high temperatures, indicating a thermally activated conduction behavior [40]. Plotting the composition-dependent impedance spectra at 445 °C [Figure 4f] reveals a dramatic increase in the semicircle radius as the CLMN concentration rises. This clearly indicates a corresponding increase in insulating resistance. In addition, the Arrhenius

law was used to calculate the required activation energy (E_a) for electrical conductivity [Equations (4) and (5)] [17,40]:

$$\sigma = \sigma_0 \exp\left(-\frac{E_a}{k_B T}\right) \quad (4)$$

$$\sigma = \frac{l}{RS} \quad (5)$$

where k_B denotes the Boltzmann constant, σ_0 denotes a pre-exponential factor, and σ denotes electrical conductivity. S denotes the cross-sectional sample area, and R represents the real-part impedance (Z') extrapolated intercept. T and l denote the measured temperature and sample thickness, respectively. Figure 4g shows the plot of $\ln \rho$ against $1000/T$; The solid line is fit using Equation (4). The E_a of the x CLMN samples was ascertained by fitting the conductivity at high temperatures linearly (1.10–1.30 eV) [Figure 4h]. The B- and A-site values of cation transport are 14 and 4 eV, respectively, and the resulting E_a values of the samples are 1.11–1.25 eV, which is similar to E_a required for the migration of the oxygen vacancy ($E_a < 2$ eV) [41]. Therefore, oxygen vacancy dictates the conductivity of the samples at high temperatures, which is in agreement with reported BT-based ceramics [19,41,42]. It is noted that when the CLMN content rises, the E_a of the 0.10CLMN ceramics peaks at 1.25 eV.

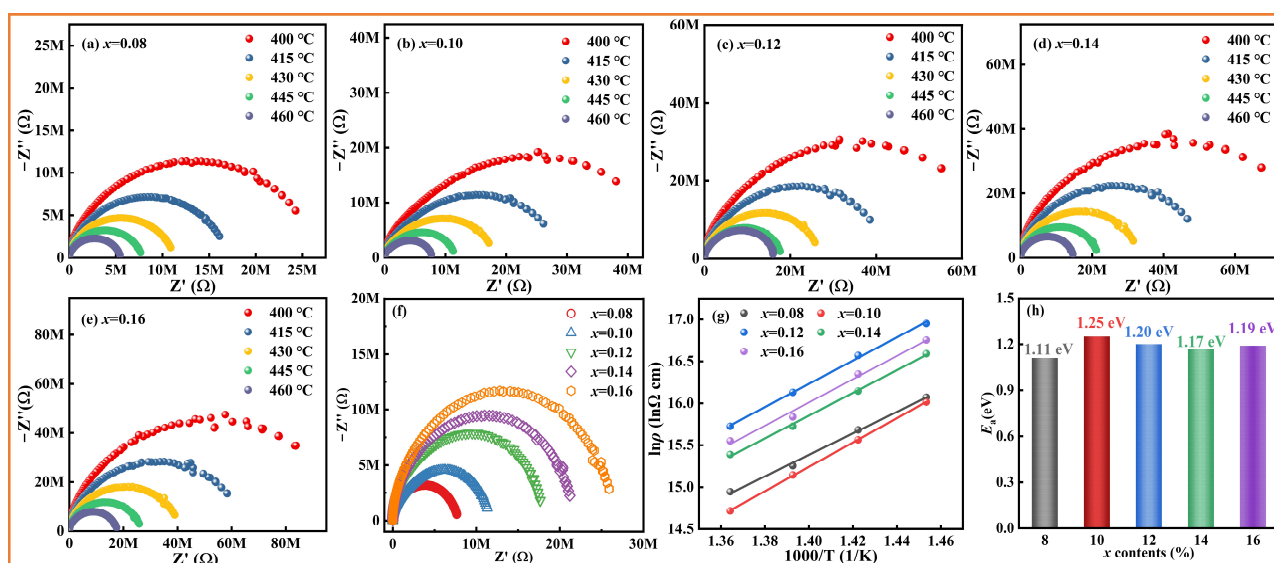


Figure 4. Impedance spectra of x CLMN ceramics measured at various temperatures: (a) $x = 0.08$; (b) $x = 0.10$; (c) $x = 0.12$; (d) $x = 0.14$; (e) $x = 0.16$; (f) composition-dependent impedance spectra of as-prepared ceramics at 445 °C; (g) Arrhenius-type plots of resistivity vs. $1000/T$ of as-prepared ceramics; (h) variation of E_a value with doping content).

The ferroelectric performance of x CLMN ceramics (Figure 5) is exhibited by their ferroelectric hysteresis P –electric field (E) loops under the applied electric field with a maximum value. With increasing CLMN content, the P – E loop gradually shifted from one with a large P_r and high E_c to a more linear shape, indicating that the long-range ferroelectric order was gradually disrupted, leading to the formation of nanodomains or PNRs [23]. The PNRs or nanodomains are dependent on the applied electric field. Under an external electric field, they exhibit a long-range ferroelectric state, while in the absence of the field, they revert to the original ergodic state. This behavior is consistent with the dielectric measurement results shown in Figure 3. A lower P_r and moderate P_{max} are obtained in 0.10CLMN ceramics, which has a peak value of E_b .

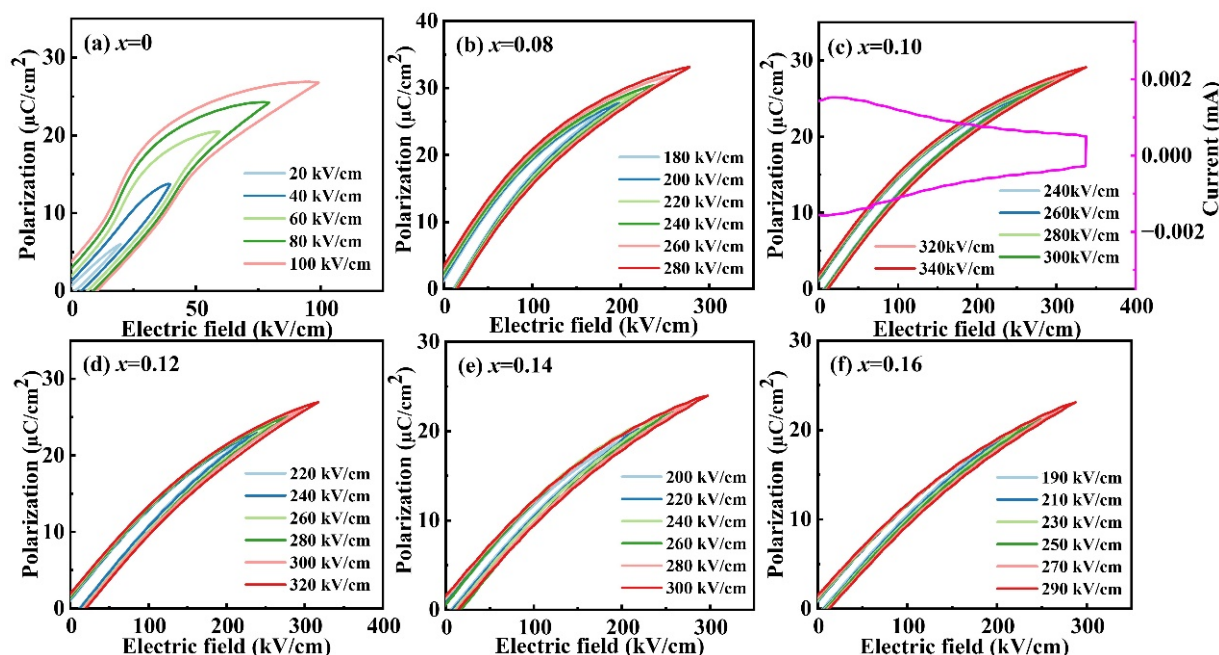


Figure 5. Unipolar P - E loops under different applied electric fields for x CLMN ceramics: (a) $x = 0$; (b) $x = 0.08$; (c) $x = 0.10$; (d) $x = 0.12$; (e) $x = 0.14$; (f) $x = 0.16$. (the pink line in (c): the current-electric field curve).

Figure 6a revealed the P - E loops of x CLMN ceramic materials determined at room temperature under a critical electric field. With increasing the CLMN content, the ferroelectric hysteresis loop of the ceramic samples narrowed, accompanied by a gradual decrease in the polarization (P) value. The observed phenomenon was attributed to the presence of the PC phase, which is consistent with the findings derived from the dielectric and XRD analyses. The energy storage properties of x CLMN ceramic materials are presented in Figure 6b,c, showing the critical values $P_{\max}$, P_r , W_{rec} , and W of P - E as a function of E . P_r demonstrated minimal variation with an increase in x , revealing the characteristic relaxor behavior. Upon removing the electric field, a transition occurred from a more coherent long-range ferroelectric ordering to PNRs. With an increase in electric field, the initially ordered short-range PNRs transition into long-range ferroelectric ordering, resulting in a continuous rise in $P_{\max}$. Thus, an increase in the external electric field led to a continuous increase in W_{rec} and W . The E_b values for 0.08CLMN, 0.10CLMN, 0.12CLMN, 0.14CLMN, and 0.16CLMN were determined to be 280, 340, 320, 300, and 290 kV cm^{-1} , respectively. The W_{rec} value of the pure sample demonstrated an initial increase from 0.65 J cm^{-3} to a maximum of 3.40 J cm^{-3} at $x = 0.10$. However, further doping leads to a decline in W_{rec} . The 0.10CLMN composition demonstrated the best energy storage performance, achieving a high value of W_{rec} (3.40 J cm^{-3}) and η (81%). Therefore, the addition of CLMN can substantially enhance the W of BT-NBT-based ceramics.

Figure 6d presents a summary of key energy storage parameters at approximately 350 kV cm^{-1} for several lead-free dielectric ceramic materials, including bismuth ferrite (BiFeO_3 , BF)-BT-based [43–45], BT-based [23,30,39], potassium sodium niobate [(K, Na) NbO_3 , KNN]-based [46–48], and NBT-based [49–53] ceramics. High η and W_{rec} of 0.10CLMN ceramic are achieved simultaneously, indicating it as a potential component for the application in energy storage devices. The energy storage properties of 0.10CLMN are comparable with most reported studies, with wide application prospects in dielectric capacitors. Table 1 presents a comparison of the energy storage characteristics of 0.10CLMN ceramic with those of other reported lead-free materials. The energy storage

properties of 0.10CLMN specimen were found to be analogous to those of other reported Pb-free ceramics.

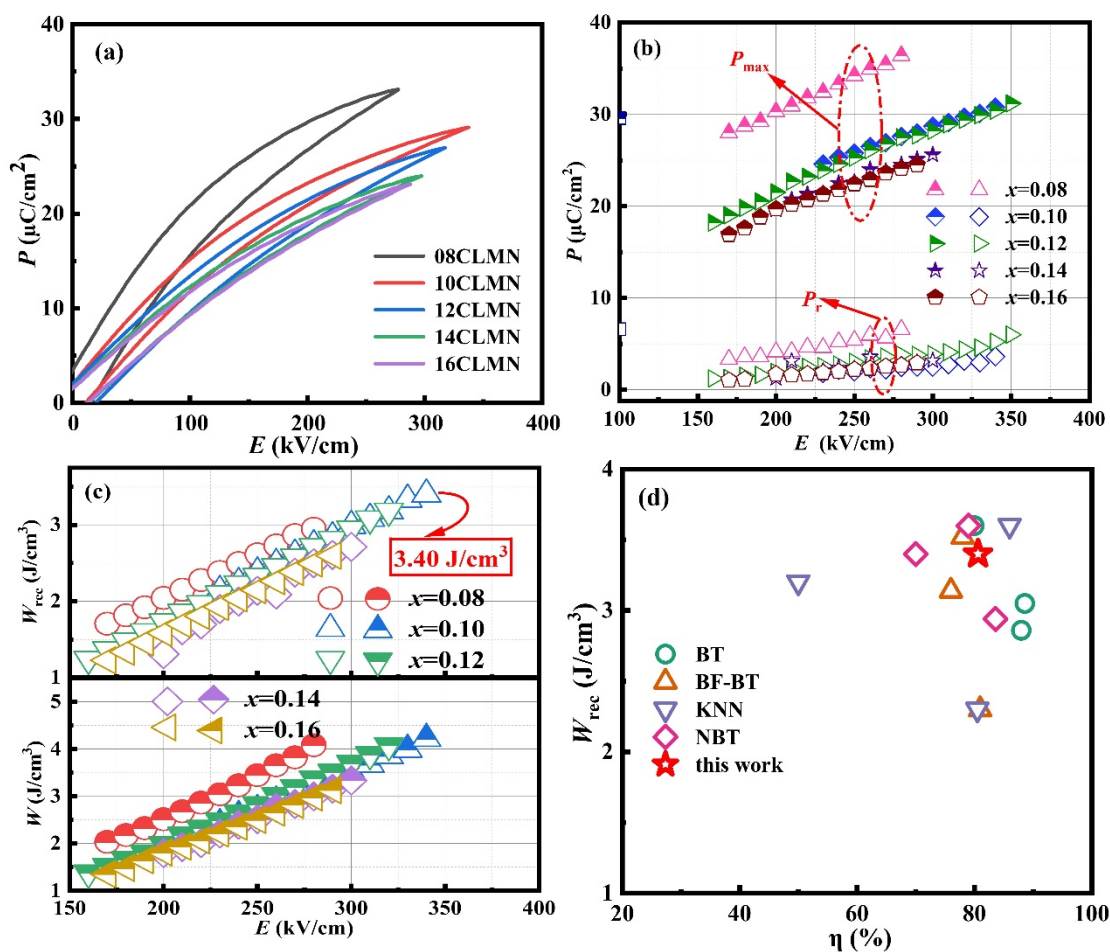


Figure 6. (a) Unipolar P - E loops prior to their breakdown electric fields; (b) $P_{\max}$, P_r and (c) W , W_{rec} change with E for x CLMN ceramics; (d) A comparison of W_{rec} and η in this study and reported in other lead-free bulk ceramics.

Table 1. Comparison of energy storage properties of 0.10CLMN ceramic with other ceramic capacitor systems.

Composition	W_{rec} (J cm^{-3})	η (%)	E_b (kV cm^{-1})	Refs.
BT-NBT-SBMT	7.12	90	720	[30]
BT-NBT-CZ	9.04	87.2	540	[19]
BT-BMH	3.38	87	240	[39]
BT-NBT-BZMASZ	3.74	82.2	273	[23]
BT-NBT-BMN	1.6	90.3	182	[24]
BF-BT-NNT	6.1	81.4	330	[54]
BF-BT-LA	5.71	80.19	270	[55]
BCT-BMZ	10.9	93	720	[56]
BNBSCTZZTN	7.3	90.6	530	[57]
KNN-STZ-BZTN	11.14	87.1	750	[46]
NBT-NN	8.04	85	630	[58]
BT-NBT-CLMN	3.40	81	340	This work

While dielectric capacitors demonstrated high W and η , maintaining stable energy storage performance is essential for their reliable use in electrostatic capacitor applications. Moreover, the capacitors must also demonstrate consistent reliability. Since 0.10CLMN

demonstrated high η and W_{rec} , its P - E loops with an E of 160 kV cm^{-1} were selected for an in-depth analysis of energy storage performance under different temperature and frequency conditions (Figure 7a,c). The values of P_{max} , P_r , W_{rec} , and η of 0.10CLMN, calculated using the methods described previously [8] are presented in Figure 7b,d. Remarkably, the P - E loops of 0.10CLMN demonstrated a narrow shape. According to Figure 7b, the P_{max} and P_r demonstrated a slight decrease with rising temperature, whereas the values of W_{rec} and η ranged from 1.46 – 1.50 J cm^{-3} and 89.28 – 90.17% , respectively. The observed fluctuations for these parameters ($\Delta W_{\text{rec}} < 2.67\%$ and $\Delta \eta < 0.82\%$) are small within this temperature range. Thus, the energy storage performance of 0.10CLMN might possess high temperature stability. Figure 7d shows that the fluctuation of P_{max} and P_r at 0.5 – 300 Hz is relatively small. Thus, W_{rec} remains relatively stable around 1.50 J cm^{-3} , with fluctuations not exceeding 2.63% across the frequency range of 0.5 to 300 Hz . The η of 0.10CLMN remained above 90% within this frequency range, demonstrating its potential for use across a broad spectrum of frequencies. These findings highlight the favorable energy storage performance of 0.10CLMN, characterized by its strong stability across both temperature and frequency variation.

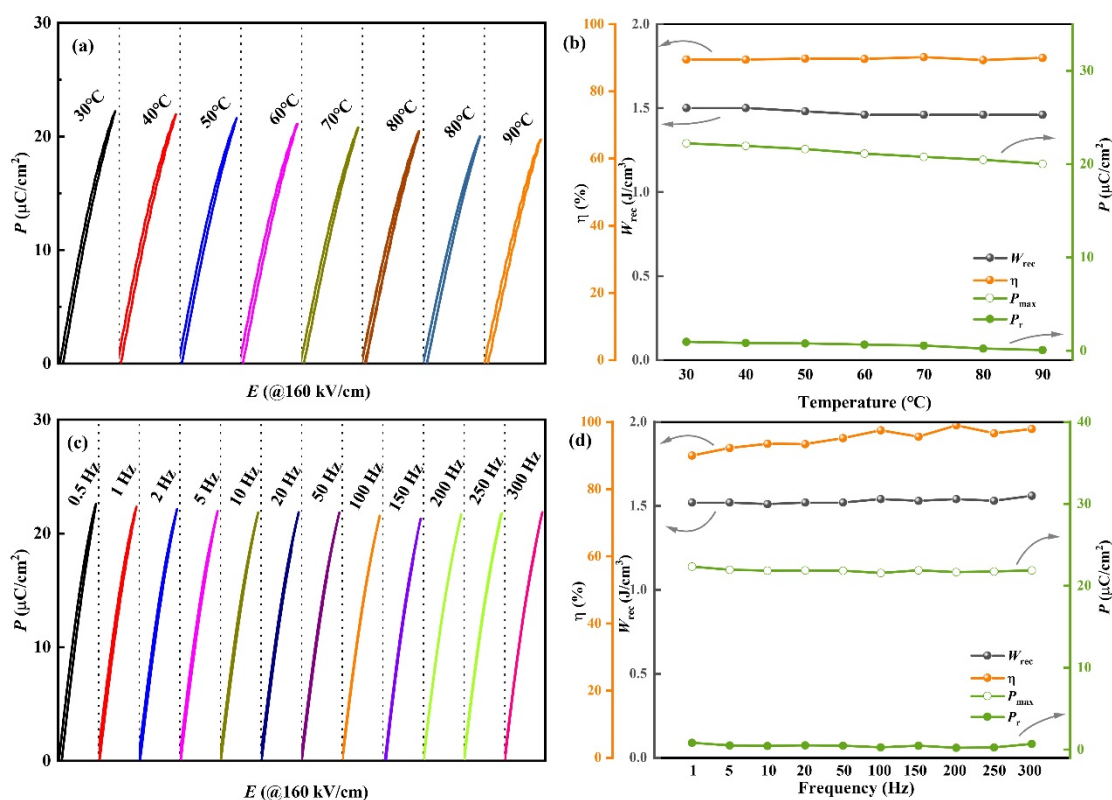


Figure 7. (a) Unipolar P - E curves (30 – 100°C at 50 Hz at 160 kV/cm); (c) Unipolar P - E curves (50 – 300 Hz at room temperature at 160 kV/cm); (b,d) show the energy storage temperature, and frequency stability of the 0.10CLMN ceramic at 160 kV/cm .

4. Conclusions

By doping CLMN, a linear dielectric material, into BT–NBT–based ceramic materials, a substantial improvement in energy storage capability was achieved. The performed modification enhanced the insulation performance and altered the relaxation behavior, optimizing the overall energy storage properties of the ceramics. These modifications significantly enhanced the breakdown strength while reducing hysteresis loss, leading to a synergistic increase in both W_{rec} and η . W_{rec} of 3.40 J cm^{-3} and η of 81% was achieved in the 0.10CLMN. The energy storage parameters demonstrated high thermal and fre-

quency stability, with W_{rec} fluctuating by less than 2.67% and 2.63% over the 30–90 °C and 0.5–300 Hz range, respectively. Thus, 0.10CLMN is a promising material for pulsed-power applications and demonstrates a linear additive approach for developing ceramic materials with high energy storage performance.

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