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BaTiO₃-Bi(Li_{0.5}Ta_{0.5})O₃, Lead-Free Ceramics and Multilayers with High Energy Storage Density and Efficiency

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KEYWORDS: BaTiO₃, ceramics, multilayers, dielectrics, energy storage, capacitors.

ABSTRACT

BaTiO₃-based materials show great promise for energy storage capacitor but their low breakdown strength and high remnant polarization currently result in relatively low energy density. Here, we report a novel (1-x)BaTiO₃-xBi(Li_{0.5}Ta_{0.5})O₃ (0.06 ≤ x ≤ 0.12, BT-xBLT) lead-free ceramic with electric field (E) ~ 280 kV cm⁻¹, discharge energy density (W_e) ~ 2.2 J cm⁻³, charge-discharge efficiency (η) >89% that is thermally stable up to 160 °C and with a fast discharge time (≤ 0.5 μs). Multilayers of compositions with x = 0.1 also exhibited high $W_e = 4.05$ J cm⁻³ and $\eta = 95.5\%$, demonstrating their potential for energy storage.

1. Introduction

Capacitors with ultrahigh power density, the ultrafast discharge speed, and a wide operating temperature range have been used for energy storage applications.¹⁻⁵ They are regarded as promising energy storage devices in hybrid electric vehicles to control ripple currents.⁶⁻⁸ Multilayer ceramic capacitors (MLCC) based on BaTiO₃ are produced in vast quantities for use as filters in computers, tablets and smart phones but their breakdown strength (E_b) and W_e are too low for applications.⁹ Mass produced, low cost capacitors with high W_e would decrease volume of device.¹⁰ High η was demanded for energy storage applications to make less waste heat causes potential dissipation problems.¹¹⁻¹³ High charge-discharge efficiency is important in such applications to mitigate heat dissipation problems.^{14,15}

The energy storage properties of multilayers mainly depends on the defect chemistry of the dielectric.¹⁶⁻¹⁸ Improving breakdown or dielectric strength, W_e and η has thus been the focus of research in this area, culminating in commercial devices based on antiferroelectric (AFE) La doped Pb(Zr,Ti)O₃ (PZT).^{19,20} Jo *et al.*¹⁹ reported the Pb-based PLZT compositions with W_{rec} of

3.04 J cm⁻³ with η of 92% at 170 kV cm⁻¹ but *Zhang et al.*²⁰ further improved properties by co-doping with Sr and Sn to give PSLZST compositions (5.56 J cm⁻³ at 350 kV cm⁻¹). However, PbO is environmentally unfriendly and is toxic to humans. Therefore, lead-free equivalents to PZT based compositions are required for high energy density storage, culminating in a number of systems being investigated such as BaTiO₃ (BT), (Ba,Sr)TiO₃ (BST), Na_{0.5}Bi_{0.5}TiO₃-based ceramics.²¹⁻²³ BT-based ceramics have a moderate dielectric constant are potential for energy storage capacitors. However, their low discharge energy density coupled with high remnant polarization (P_r) hinder practical applications. For BT-based ceramics dielectrics, energy density is illustrated as:

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

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

$$\eta = \frac{W_{rec}}{W} \times 100\% \quad (3)$$

where E is electric field. BT-based ceramics for energy storage application are thus required with a relatively high E , large P_{max} and low P_r . Therefore, optimising E and maximizing the values of ($P_{max} - P_r$) enhance the W_e of BT-based samples.²⁴⁻²⁸ Oxides which are known to have intrinsically high dielectric strength such as Al₂O₃, SiO₂ and MgO have been introduced into the BT-based ceramics to enhance the breakdown voltage and energy storage properties.²⁴⁻²⁶ BT based ceramics formed from Al₂O₃- and SiO₂-coated powders show higher breakdown strength (190 kV cm⁻¹) which gives rise to higher $W_e = 0.725$ J cm⁻³ and greater $\eta \sim 80\%$.²³ BaSrTiO₃-MgO composites were prepared by using a new preparation method (SPS) to obtain breakdown

strength 300 kV cm^{-1} , $W_e \sim 1.5 \text{ J cm}^{-3}$ and $\eta \sim 88.5\%$.²⁵ Recent studies have shown that Bi^{3+} modified BaTiO_3 ceramics own high W_e due to their large P_{max} and small P_r . BaTiO_3 - BiMeO_3 systems have been studied widely with 0.7BaTiO_3 - 0.3BiScO_3 capacitors exhibiting remarkably high W_e of 6.1 J cm^{-3} but under extremely high electric field, 730 kV cm^{-1} .²⁶ $0.75(\text{Bi}_{0.75}\text{Nd}_{0.15})\text{FeO}_3$ - 0.25BaTiO_3 - $0.1\text{wt}\%$ MnO_2 ceramics have been shown to exhibit high W_{rec} (1.81 J cm^{-3}) and low η (41.3%) but multilayers of the same composition demonstrated significant improvement in performance.²⁷ In contrast, Shen *et al.* reported 0.91BaTiO_3 - 0.09BiYbO_3 ceramics with high $W_e \sim 0.71 \text{ J cm}^{-3}$ and further work on relaxor compounds in the solid solutions BaTiO_3 - $\text{BiB}^{3+}\text{O}_3$ ($\text{B} = \text{Y}, \text{Sc}, \text{Ga}, \text{Al}, \text{In}$), BaTiO_3 - $\text{Bi}(\text{Mg}_{2/3}\text{Nb}_{1/3})\text{O}_3$ and BaTiO_3 - $\text{Bi}(\text{Mg}_{1/2}\text{Ti}_{1/2})\text{O}_3$ revealed W_e typically limited to $<1.85 \text{ J cm}^{-3}$.²⁸⁻³² Despite the disappointing values of W_e , BT based relaxor-ferroelectrics hold promise for energy storage and inspired the design and synthesis of 0.88BT - 0.12BLN ceramic, which exhibited one of the highest energy density (2.013 J cm^{-3} under 270 kV cm^{-1}) in lead free ceramics.³³ Most importantly, it indicated that Bi^{3+} ion inter into the Ba site could optimize the W_e by maximizing P_{max} and minimising P_r .^{34,35} In this contribution, a further relaxor ferroelectrics BT - $x\text{BLT}$ ($0.06 \leq x \leq 0.12$) was designed with enhanced $E \sim 280 \text{ kV cm}^{-1}$, $W_e \sim 2 \text{ J cm}^{-3}$ and $\eta \sim 88\%$. Multilayers of the ceramic compositions also exhibited promising properties with $E \sim 466 \text{ kV cm}^{-1}$, $W_e \sim 4.05 \text{ J cm}^{-3}$ and $\eta \sim 95.5\%$.

2. Results and discussion

Figure 1a shows the XRD traces of BT - $x\text{BLT}$ ($0.06 \leq x \leq 0.12$) samples. All peaks of BT - $x\text{BLT}$ ceramics are indexed according to a cubic perovskite phase, indicating solid solubility up to at least $x = 0.12$. Fig. 1b shows the variation of cell parameters and the volume of the BT - $x\text{BLT}$ ceramics and Rietveld refinement fitting of 0.90BT - 0.10BLT , respectively. From Fig. 1b,

cell parameters and the volume increase from 4.0117(1) to 4.0152(2) Å and 64.563(5) to 64.736(5) Å³ with the *x* values increase from 0.06 to 0.12, respectively. According to the Shannon's ionic radii (*R*), *R* of Ba²⁺ (1.61 Å, 12 fold coordination) in the ABO₃ perovskite structure is larger than Bi³⁺ (1.35 Å).³⁶ The average ionic radii of (Li_{0.5}Ta_{0.5})³⁺ is 0.70 Å³⁶ and similar to but slightly larger than Ti⁴⁺ (0.605 Å). We propose therefore that Bi³⁺ and (Li_{0.5}Ta_{0.5})³⁺ enter the Ba and Ti site, respectively. XRD patterns were refined with space group Pm-3m (No. 221). Fig. S1 exhibited the results of fitted XRD patterns of BT-*x*BLT. The results of refined crystal structure and related parameters are also listed in Table S1. Fig. 1c shows Rietveld refinement fitting of 0.90BT-0.10BLT ceramic with all reflections indexed and with a satisfactory 'goodness of fit' (*R_p*= 6.74%, *R_{wp}*= 9.02% and *R_{exp}*= 5.95%). Refined fractional coordinates of Ba²⁺, Bi³⁺, Li⁺, Ta⁵⁺, Ti⁴⁺ ions are shown in Table 1 with *a* = *b* = *c* = 4.0139 (2) Å, (Ba²⁺ and Bi³⁺) and (Li⁺, Ta⁵⁺ and Ti⁴⁺) randomly distributed at the 1a Wyckoff position 1b respectively. Meantime, oxygen occupying 3c. A <110> SAED pattern of 0.90BT-0.10BLT is also shown in the inset of Fig. 1c which confirms the absence of superstructure reflections, consistent with Pm-3m symmetry.

Figure 2 a, c, e and g show SEM images of thermal etching surfaces, respectively for 0.94BT-0.06BLT, 0.92BT-0.08BLT, 0.90BT-0.10BLT and 0.88BT-0.12BLT samples. All samples exhibit a dense microstructure and no porosity. Distribution histograms from BT-*x*BLT (Fig 2b, d, f and h) show an average grain size of 0.6, 0.8, 1.2, and 1.6 for *x* = 0.06, 0.08, 0.10, and 0.12, respectively.

Figure 3 exhibits the location distribution of ions in 0.90BT-0.10BLT ceramic by using the elemental mapping. In Fig. 3a, it shows that these ions are uniformly distributed on top of the

grain and do not aggregate at the grain boundary. The ions of 0.90BT-0.10BLT ceramic are similar molar ratios to the batched 0.90BT-0.10BLT composition, Fig. 3b.

Figure 4 shows dielectric constant (ϵ_r) and dielectric loss ($\tan\delta$) of BT-BLT samples in temperature range -90 to 110 °C from 100 Hz to 1 MHz. ϵ_r decreased from 1980 for 0.94BT-0.06BLT to 760 for 0.88BT-0.12BLT ceramics accompanied by a low $\tan\delta$ (< 0.01) at room temperature. Previous studies have reported that $\text{BiM}_1\text{M}_2\text{O}_3$ substitution in BaTiO_3 mitigates sintering temperature and dielectric loss.²⁴⁻²⁶ with BT-BLT ceramics following similar trends. Fig. S2 exhibits Frequency dependence of ϵ_r and $\tan\delta$ of the BT-BLT ceramics at 25 °C is shown in. ϵ_r decreases as frequency increases from 1 kHz ~ 10 MHz consistent with a dielectric relaxation widely observed in related BaTiO_3 - BiMeO_3 systems.²⁴⁻²⁶

From Fig. 5a, the electrical resistivity of BT-BLT increased with increasing BLT concentration to maximum at $x = 0.10$. Breakdown strength (E_b) is key to optimizing energy storage density and therefore it is critical to have a robust methodology to establish this metric. To this end, the results of E_b are analysed by using the Weibull distribution function in Fig. 5b. From Fig. 5b, it can be seen all results of E_b fit well with the Weibull distribution for the samples. The characteristic E_b of 0.94BT-0.06BLT, 0.92BT-0.08BLT, 0.90BT-0.10BLT and 0.88BT-0.12BLT samples are therefore 290, 330, 335, and 332 kV cm⁻¹, respectively. The highest characteristic E_b can be achieved in the 0.9BT-0.1BLT ceramic with high resistivity about $3.53 \times 10^{12} \Omega \text{ cm}$. Compared with other BaTiO_3 -based samples, the values of E_b are significantly improved.²⁸⁻³⁶ Fig. S3 shows polarization-electric field (P-E) hysteresis of BT-xBLT ceramics measured with unipolar triangle signals at 10 Hz near the breakdown voltage. The P-E hysteresis of BT-xBLT samples transform gradually from a saturated loop to slim loops as x increased. From Fig. S3, breakdown strength of BT-BLT ceramics increases from 175 to

280 kV cm⁻¹ but then reduces to 225 kV cm⁻¹ with increase in BLT concentration and is significantly greater than most BT-based bulk ceramics.²⁸⁻³⁶ The W_e of the BT-BLT samples was measured by using P-E hysteresis loops in Fig. S3a. Fig. 5c illustrates the W_e and η as a function of BLT concentration. Although the W_e increases and reaches a maximum but then decreases with increase in BLT concentration, the η continuously increase from $0.06 \leq x \leq 0.12$. Outstanding energy storage performance of 0.90BT-0.10BLT is about $W_e \sim 2.2 \text{ J cm}^{-3}$ and $\eta \sim 88.1\%$ under 280 kV cm⁻¹. Fig. S3b shows P-E hysteresis loops of the 0.90BT-0.10BLT ceramic under various E at 25 °C. Based on P-E hysteresis loops of 0.90BT-0.10BLT sample in Fig. S3b, the result of the high performance of ceramic is exhibited in Fig. 5d. When electric field adds from 40 to 280 kV cm⁻¹, the W_e increases from 0.21 J cm⁻³ to 2.2 J cm⁻³ while the η is consistently > 88%.

For the capacitors in practical use, the discharge speed is required as fast as possible for pulsed power applications. It usually defined the $\tau_{0.9}$ as the discharge time to evaluate discharge speed. The $\tau_{0.9}$ means the time it takes to release 90% of energy value in the circuit.⁵¹⁻⁵³ It is important to study the discharge process by using a capacitor discharge circuit (Fig. S4). In this study, the discharge processes of the BT-xBLT capacitors are measured by load resistor of 205 Ω . Fig. 6 shows the discharging rate of BT-xBLT ceramics at 150 kV cm⁻¹. Fig. 6 exhibits the variation of discharge current waveforms and W_e on discharge time. The $\tau_{0.9}$ of the 0.94BT-0.06BLT, 0.92BT-0.08BLT, 0.90BT-0.10BLT and 0.88BT-0.12BLT ceramics are about 0.25 μs , 0.28 μs , 0.32 μs , 0.34 μs in the discharge processes.

In general, the ambient temperature of electronic device is up to 160 °C.³² So thermal stability is very important for electronic materials.³³ Fig. S5 shows P - E loops of BT-xBLT from 25 to 160 °C under same electric field of 150 kV cm⁻¹ with BT-xBLT ceramics generally

exhibiting slim P-E loops. Fig. 7 shows P_r , P_{max} and energy storage properties of BT-xBLT ceramics in from 25 to 160 °C. P_r and P_{max} decreased only slightly at 160 °C, compared with room temperature, as shown in Fig. 7a, indicating that polarization response is almost independent of temperatures. Fig. 7b exhibits the variation of W_e and η with temperature. Only slight variation in W_e coupled with a high η is observed up 160 °C.

To further illustrate the potential of the BT-xBLT system for energy storage applications, 0.90BT-0.10BLT multilayers with Pt internal electrode were prepared by the tape-casting (see Supporting Information). Fig. 8a shows SEM images of a 0.90BT-0.10BLT multilayer with a dielectric layer thickness of $\sim 30 \mu\text{m}$. Fig. 8b exhibits P - E hysteresis of the 0.90BT-0.10BLT multilayer under varying E at 25 °C. High $E_b \sim 466 \text{ kV cm}^{-1}$ is observed, favourable for high W_e which along with the slim P-E loops is expected to give rise to high energy storage density. The studies have been shown that high η is demanded of multilayers in energy storage applications as waste heat causes potential dissipation problems in the charge-discharge progress.³⁵⁻³⁹ Fig. 8c shows the W_e and η of 0.90BT-0.10BLT multilayers at various electric fields. The W and W_e of 0.90BT-0.10BLT multilayers increased from 0.59 and 0.63 to 4.05 and 4.24 J cm^{-3} , respectively, with the E enhanced from 120 to 466 kV cm^{-1} . In addition, high η (95.5%) was obtained for the 0.90BT-0.10BLT multilayers at high breakdown voltages (466 kV cm^{-1}). High η mitigates potential heat dissipation problems.¹³⁻¹⁶ Finally, Fig. 8d compares the W_e , E_b , and η of this work with other outstanding lead free systems which illustrates that 0.90BT-0.10BLT ceramics and multilayers have a combination of energy density, breakdown strength, and efficiency superior to other lead free systems.

3. Conclusion

In summary, we reported novel high E_b and W_e of the BT-xBLT ceramics and multilayers. This work significantly improved energy storage capacity of BT-BLT system ($W_e \sim 4.05 \text{ J cm}^{-3}$ and $\eta \sim 95.5\%$) than other BaTiO₃-based systems ($W_e < 2 \text{ J cm}^{-3}$ and $\eta \sim 80\%$). All samples densified at 1200 ~ 1260 °C with XRD demonstrated that a solid solution had been formed. The 0.90BT-0.10BLT ceramics had high $E_b \sim 280 \text{ kV cm}^{-1}$, $W_e \sim 2.2 \text{ J cm}^{-3}$, $\eta \sim 88\%$, temperature stability from ~25 °C to ~ 160 °C with a fast release speed ($\tau_{0.9} \leq 0.50 \text{ } \mu\text{s}$). Meanwhile, the 0.90BT-0.10BLT multilayers were fabricated with ultra-high energy density ~ 4.05 J cm⁻³ and $\eta \sim 95.5\%$ under 466 kV/cm, which confirmed that lead-free BT-BLT systems are promising alternative for applications in high energy density storage.

Figure Captions:

Figure 1. (a) XRD patterns of the BT-xBLT ceramics. (b) Variation cell parameters of the BT-xBLT ceramics. (c) Rietveld profile fitting of 0.90BT-0.10BLT. The open circles show experimental and the continuous curve shows the best fit to the XRD pattern with a cubic Pm-3m space group. Insets give the perovskite structure of ABO₃ and SAED.

Figure 2. SEM images of thermally etched surfaces and grain size distribution histograms for the BT-xBLT ceramics: (a)-(b) 0.94BT-0.06BLT, (c)-(d) 0.92BT-0.08BLT, (e)-(f) 0.90BT-0.10BLT, (g)-(h) 0.88BT-0.12BLT.

Figure 3. (a) Element mapping images of 0.90BT-0.10BLT sintered at optimum temperature and (b) EDX chemical mapping: blue, O-K; cyan, Bi-M; olive, Ba-L; green, Ti-K; and red, Ta-L.

Figure 4. Relative permittivity and loss tangent of BT-xBLT ceramics as a function of temperature (-90 °C to 110 °C) from 100 Hz to 1 MHz: (a) 0.94BT-0.06BLT, (b) 0.92BT-0.08BLT, (c) 0.90BT-0.10BLT, (d) 0.88BT-0.12BLT.

Figure 5. (a) Electrical resistivity of BT-xBLT ceramics. (b) Weibull distributions of E_b of BT-xBLT ceramics. (c) W_e and η of BT-xBLT ceramics near their breakdown strength. (d) The W_e and η of 0.90BT-0.10BLT ceramic at various electric fields.

Figure 6. (a) Pulsed discharging current of BT-xBLT samples at same electric field of 150 kV cm^{-1} . (b) The W_e of BT-xBLT samples depending on discharging time.

Figure 7. (a) The P_r and P_{max} for BT-xBLT ceramics at different temperatures. (b) The W_e and η of BT-xBLT ceramics depending on temperatures.

Figure 8. (a) SEM images of 0.90BT-0.10BLT MLCC samples. (b) The P-E hysteresis of 0.90BT-0.10BLT MLCC samples at 466 kV cm^{-1} at 25 °C. (c) Variation of the W_e and η of 0.90BT-0.10BLT MLCC samples depending on electric field. (d) Comparison of the W_e and η of 0.90BT-0.10BLT MLCC samples and other lead-free ceramic systems: BT-BLN ($\text{BaTiO}_3\text{-Bi}(\text{Li}_{1/2}\text{Nb}_{1/2})\text{O}_3$)³³; BT-BMT ($\text{BaTiO}_3\text{-Bi}(\text{Mg}_{1/2}\text{Ti}_{1/2})\text{O}_3$)²⁶; BST ($\text{Ba}_{0.4}\text{Sr}_{0.6}\text{TiO}_3$)²⁰; BT-BMN ($\text{BaTiO}_3\text{-Bi}(\text{Mg}_{2/3}\text{Nb}_{1/3})\text{O}_3$)²⁵; BT-BI ($\text{BaTiO}_3\text{-BiInO}_3$)³³; BT-BZN ($\text{BaTiO}_3\text{-Bi}(\text{Zn}_{2/3}\text{Nb}_{1/3})\text{O}_3$)²³; BT-BZT ($\text{BaTiO}_3\text{-Bi}(\text{Zn}_{1/2}\text{Ti}_{1/2})\text{O}_3$)³³; BT-BS ($\text{BaTiO}_3\text{@BiScO}_3$)³⁷; BT-BYb ($\text{BaTiO}_3\text{-BiYbO}_3$)²²; BT-BY ($\text{BaTiO}_3\text{-BiYO}_3$)³⁷; BLNLTZ-NBN ($\text{Na}_{0.73}\text{Bi}_{0.09}\text{NbO}_3\text{-Bi}_{0.48}\text{La}_{0.02}\text{Na}_{0.48}\text{Li}_{0.02}\text{Ti}_{0.98}\text{Zr}_{0.02}\text{O}_3$)⁴²; BT-NBT-NBN ($\text{BaTiO}_3\text{-Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-Na}_{0.73}\text{Bi}_{0.09}\text{NbO}_3$)³⁵; NBBT-BMT ($(\text{Na}_{0.5}\text{Bi}_{0.5})_{0.92}\text{Ba}_{0.08}\text{TiO}_3\text{-Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$)³⁴; NBT-KN ($\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3\text{-KNbO}_3$)⁴⁶; NBBT-KN ($\text{Na}_{0.47}\text{Bi}_{0.47}\text{Ba}_{0.06}\text{TiO}_3\text{-KNbO}_3$)⁴⁶; NBT-BT-SZ-NN ($\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-BaTiO}_3\text{-SrZrO}_3\text{-NaNbO}_3$)³⁶; NBABT ($\text{Na}_{0.42}\text{Bi}_{0.44}\text{Al}_{0.06}\text{Ba}_{0.08}\text{TiO}_3$)³⁸; AN (AgNbO_3)⁵⁰. Red and blue bars correspond to energy density and electric field.

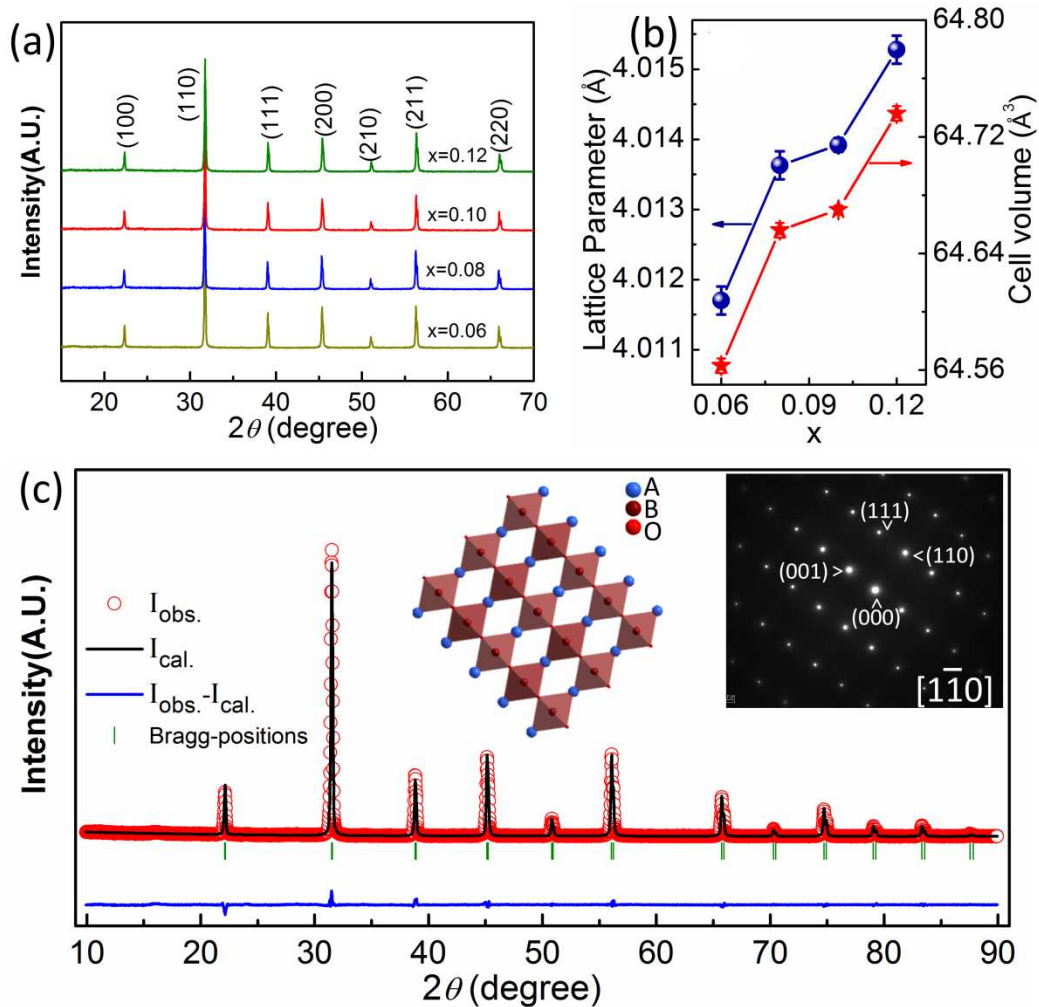


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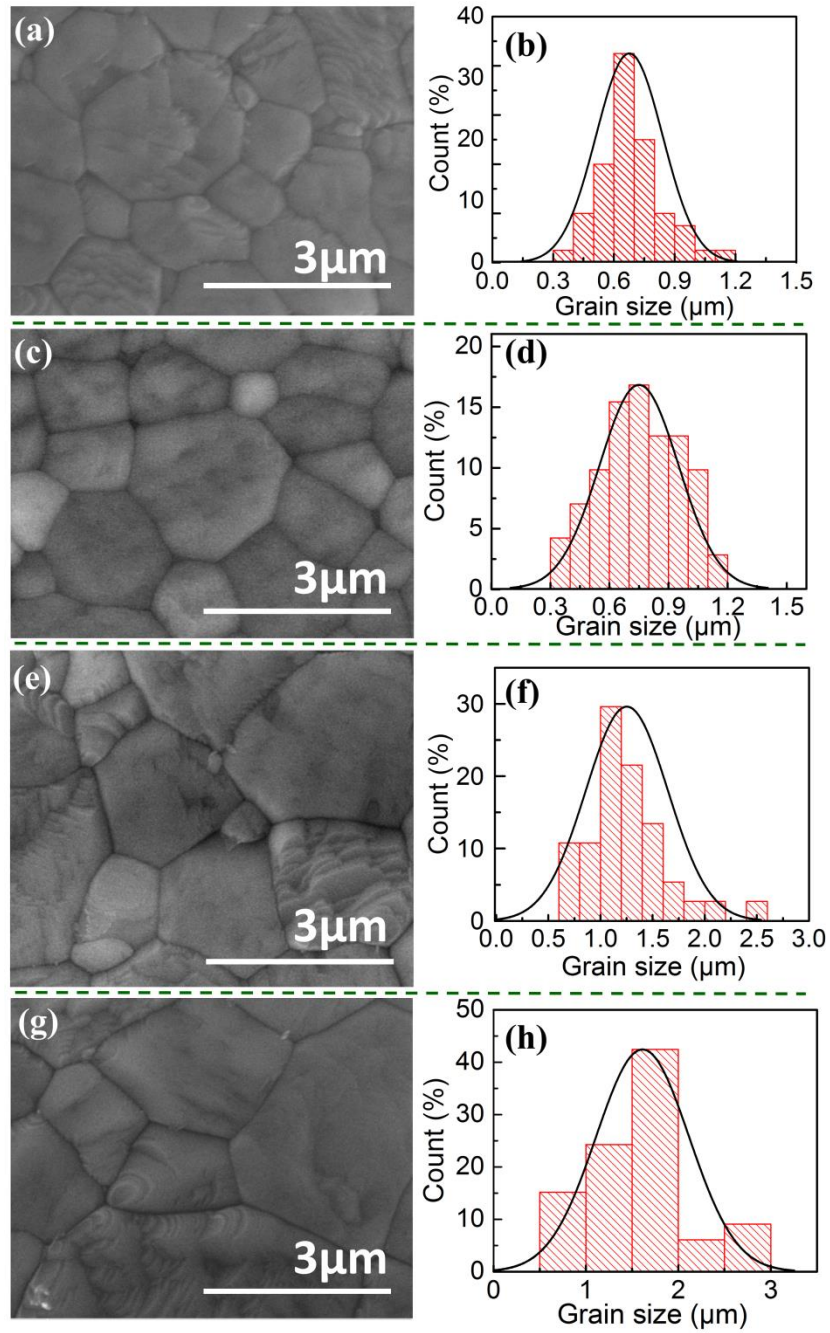


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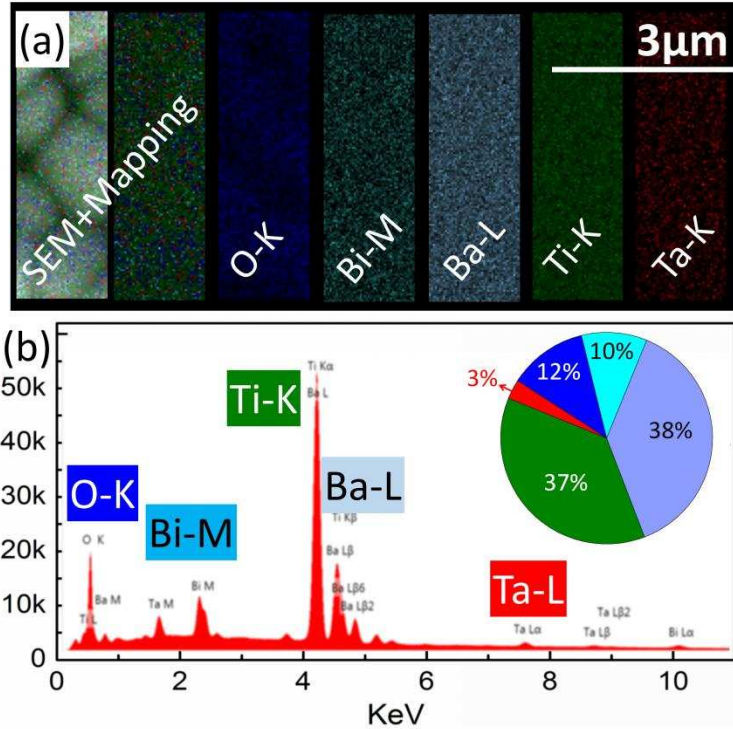


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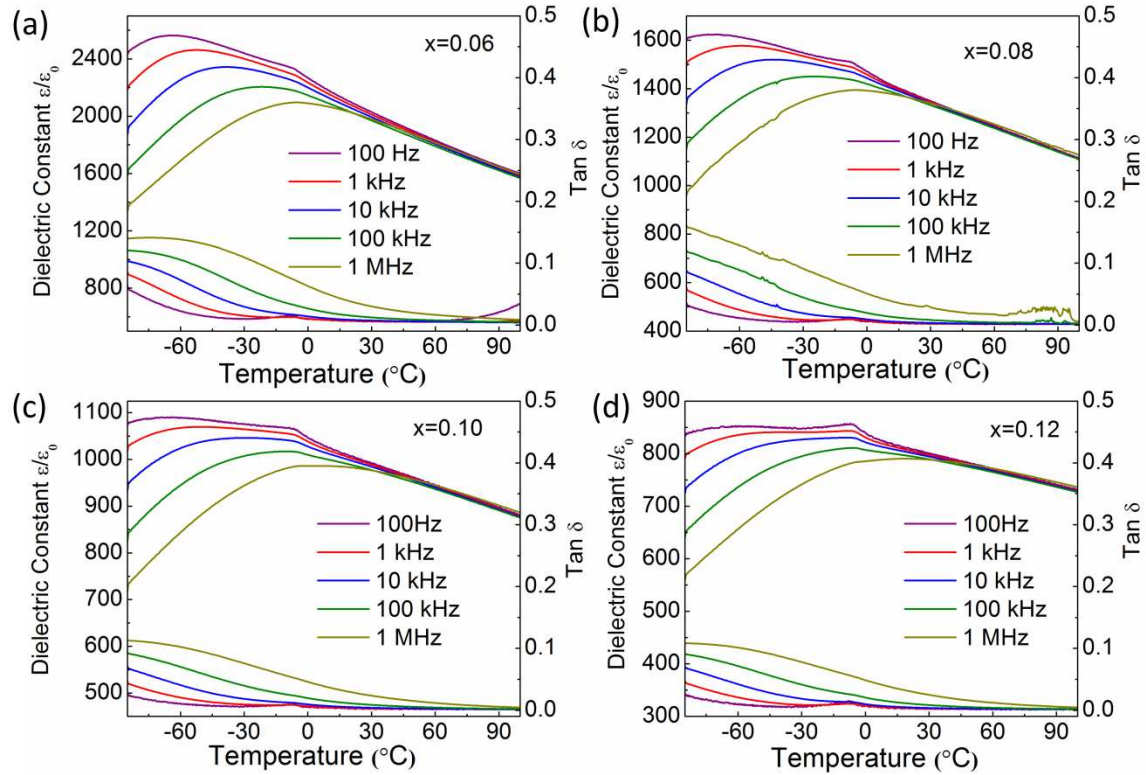


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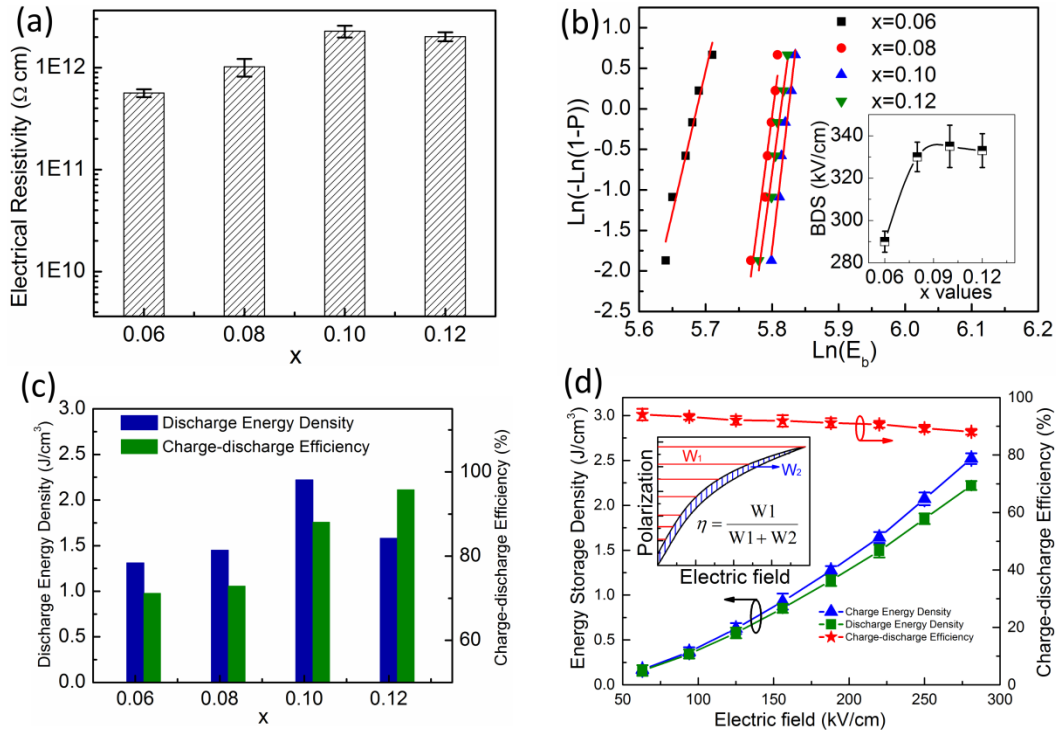


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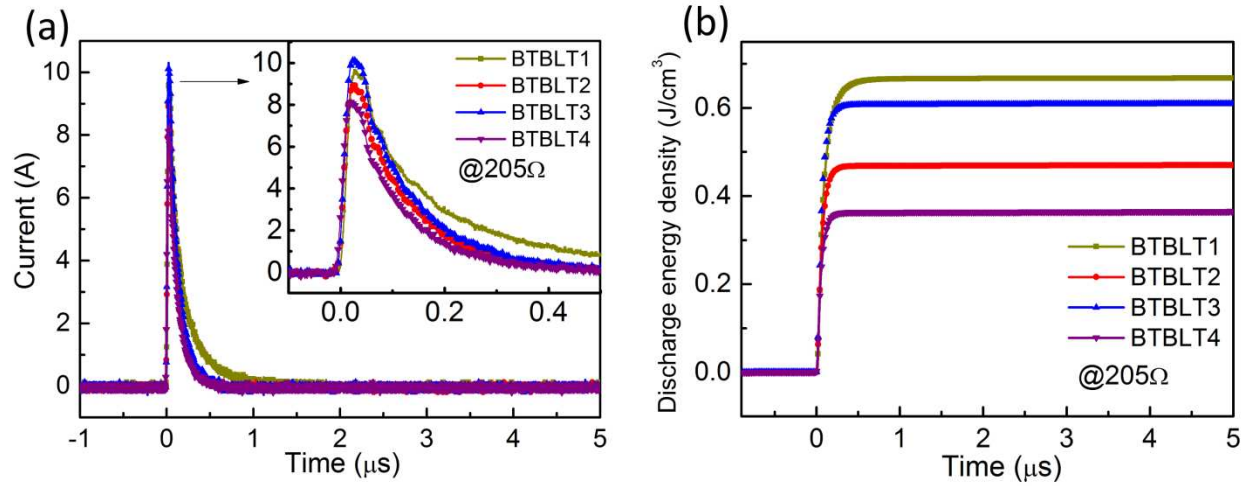


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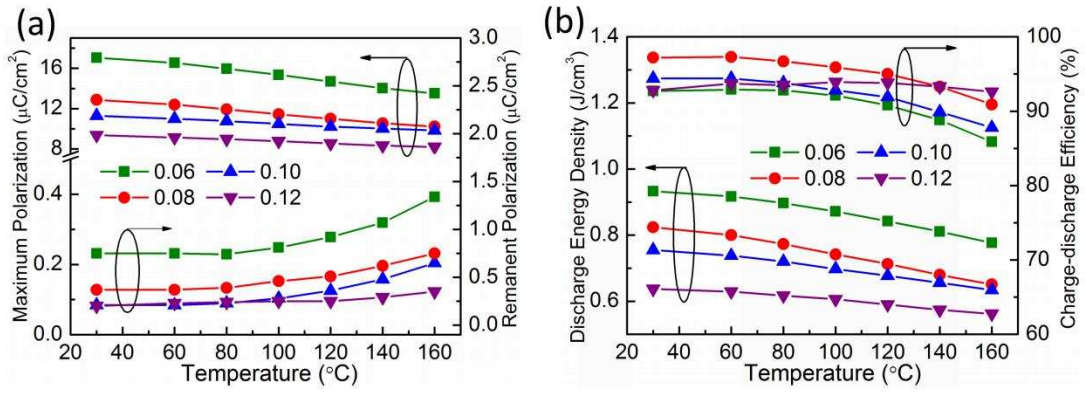


Figure 7. (a) The P_r and P_{max} for BT-xBTLT ceramics at different temperatures. (b) The W_e and η of BT-xBTLT ceramics depending on temperatures.

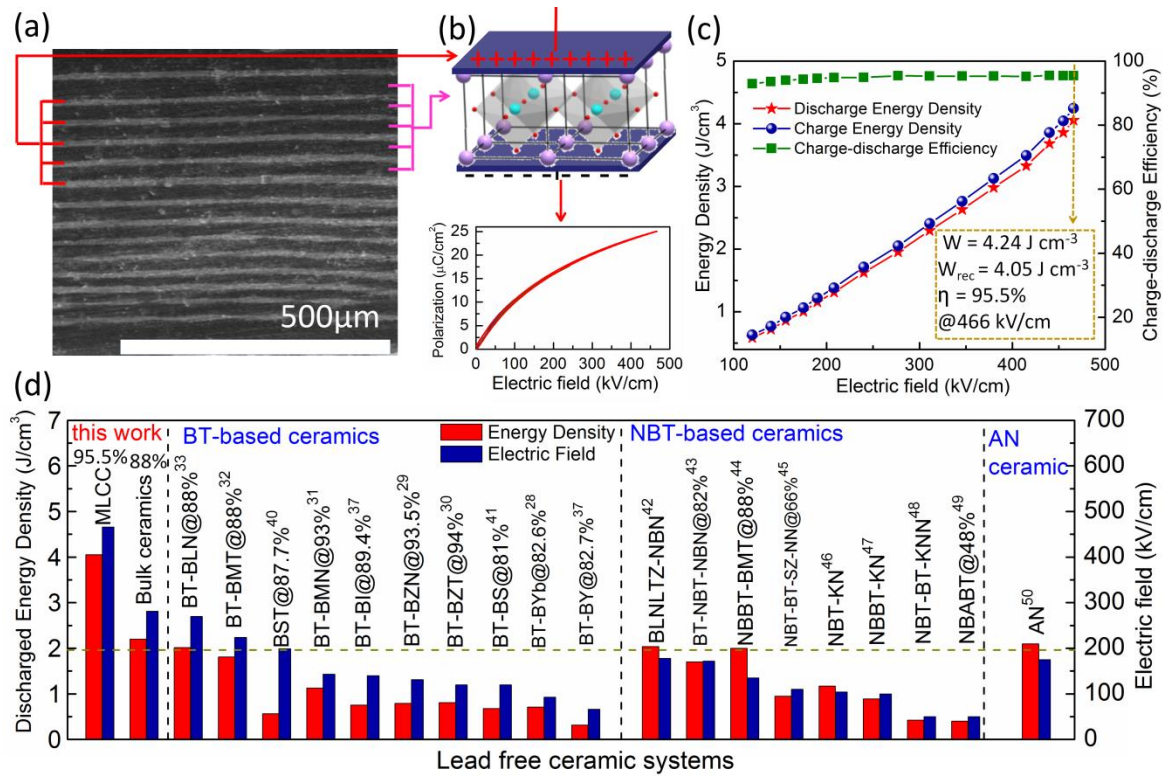


Figure 8. (a) SEM images of 0.90BT-0.10BLT MLCC samples. (b) The P-E hysteresis of 0.90BT-0.10BLT MLCC samples at 466 kV cm^{-1} at 25°C . (c) Variation of the W , W_e and η of 0.90BT-0.10BLT MLCC samples depending on electric field. (d) Comparison of the W_e and η of 0.90BT-0.10BLT MLCC samples and other lead-free ceramic systems: BT-BLN ($\text{BaTiO}_3\text{-Bi}(\text{Li}_{1/2}\text{Nb}_{1/2})\text{O}_3$)³³; BT-BMT ($\text{BaTiO}_3\text{-Bi}(\text{Mg}_{1/2}\text{Ti}_{1/2})\text{O}_3$)³²; BST ($\text{Ba}_{0.4}\text{Sr}_{0.6}\text{TiO}_3$)⁴⁰; BT-BMN ($\text{BaTiO}_3\text{-Bi}(\text{Mg}_{2/3}\text{Nb}_{1/3})\text{O}_3$)³¹; BT-BI ($\text{BaTiO}_3\text{-BiInO}_3$)³⁷; BT-BZN ($\text{BaTiO}_3\text{-Bi}(\text{Zn}_{2/3}\text{Nb}_{1/3})\text{O}_3$)²⁹; BT-BZT ($\text{BaTiO}_3\text{-Bi}(\text{Zn}_{1/2}\text{Ti}_{1/2})\text{O}_3$)³⁰; BT-BS ($\text{BaTiO}_3\text{@BiScO}_3$)⁴¹; BT-BYb ($\text{BaTiO}_3\text{-BiYbO}_3$)²⁸; BT-BY ($\text{BaTiO}_3\text{-BiYO}_3$)³⁷; BLNLTZ-NBN ($\text{Na}_{0.73}\text{Bi}_{0.09}\text{NbO}_3\text{-Bi}_{0.48}\text{La}_{0.02}\text{Na}_{0.48}\text{Li}_{0.02}\text{Ti}_{0.98}\text{Zr}_{0.02}\text{O}_3$)⁴²; BT-NBT-NBN ($\text{BaTiO}_3\text{-Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-Na}_{0.73}\text{Bi}_{0.09}\text{NbO}_3$)⁴³; NBBT-BMT ($(\text{Na}_{0.5}\text{Bi}_{0.5})_{0.92}\text{Ba}_{0.08}\text{TiO}_3\text{-Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$)⁴⁴; NBT-KN ($\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3\text{-KNbO}_3$)⁴⁶; NBBT-KN ($\text{Na}_{0.47}\text{Bi}_{0.47}\text{Ba}_{0.06}\text{TiO}_3\text{-KNbO}_3$)⁴⁷; NBT-BT-SZ-NN ($\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-BaTiO}_3\text{-SrZrO}_3\text{-NaNbO}_3$)⁴⁵; NBABT ($\text{Na}_{0.42}\text{Bi}_{0.44}\text{Al}_{0.06}\text{Ba}_{0.08}\text{TiO}_3$)⁴⁹; AN (AgNbO_3)⁵⁰. Red and blue bars correspond to energy density and electric field.

TABLES.

Table 1. Refined structural parameters from XRD Data for the 0.90BT-0.10BLT Sample.

Atom	Site	Occ.	x	y	z	Biso.
Ba1	1a	0.300	0.0000	0.0000	0.0000	1.5073(8)
Bi1	1a	0.033	0.0000	0.0000	0.0000	1.5073(8)
Ti1	1b	0.300	0.5000	0.5000	0.5000	0.0986(1)
Li1	1b	0.017	0.5000	0.5000	0.5000	0.0986(1)
Ta1	1b	0.017	0.5000	0.5000	0.5000	0.0986(1)
O1	3c	1.000	0.5000	0.5000	0.0000	0.0921(1)

$a = b = c = 4.0139 (2) \text{ \AA}$. $R_p \sim 6.74$, $R_{wp} \sim 9.02$, $R_{exp} \sim 5.95$

ASSOCIATED CONTENT

Supporting Information.

Experimental Section; Preparation of the MLCC samples; crystal and refinement parameters for BT-xBLT; rietveld profile fitting of BT-xBLT for $x = 0.06, 0.08, 0.10$ and 0.12 ; the P-E hysteresis of BT-xBLT ceramics near their breakdown strength; The P-E hysteresis of 0.90BT-0.10BLT ceramic at room temperatures as a function of electric fields; the P-E hysteresis of BT-xBLT ceramics at RT to $160\text{ }^{\circ}\text{C}$; the schematic diagram for pulsed discharging current.

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Notes

The authors declare no competing financial interest.

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