Perspectives and challenges for lead-free energy-storage multilayer ceramic capacitors

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Abstract: The growing demand for high-power-density electric and electronic systems has encouraged the development of energy-storage capacitors with attributes such as high energy density, high capacitance density, high voltage and frequency, low weight, high-temperature operability, and environmental friendliness. Compared with their electrolytic and film counterparts, energy-storage multilayer ceramic capacitors (MLCCs) stand out for their extremely low equivalent series resistance and equivalent series inductance, high current handling capability, and high-temperature stability. These characteristics are important for applications including fast-switching third-generation wide-bandgap semiconductors in electric vehicles, 5G base stations, clean energy generation, and smart grids. There have been numerous reports on state-of-the-art MLCC energy-storage solutions. However, lead-free capacitors generally have a low-energy density, and high-energy density capacitors frequently contain lead, which is a key issue that hinders their broad application. In this review, we present perspectives and challenges for lead-free energy-storage MLCCs. Initially, the energy-storage mechanism and device characterization are introduced; then, dielectric ceramics for energy-storage applications with aspects of composition and structural optimization are summarized. Progress on state-of-the-art energy-storage MLCCs is discussed after elaboration of the fabrication process and structural design of the electrode. Emerging applications of energy-storage MLCCs are then discussed in terms of advanced pulsed power sources and high-density power converters from a theoretical and technological point of view. Finally, the challenges and future prospects for industrialization of lab-scale lead-free energy-storage MLCCs are discussed.

Keywords: multilayer ceramic capacitors (MLCCs); lead-free dielectric ceramics; energy storage; high power density

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1 Introduction

In recent decades, the consumption of energy and natural resources has increased alongside our growing population and social and technical advancements. However, there are significant challenges associated with the sustainability of using coal, oil, and natural gas as our primary sources of energy. First, the consumption of fossil fuels has caused increasingly serious environmental pollution and ecological destruction, and second, non-renewable fossil energy is a limited resource. Therefore, clean and renewable energy sources, such as solar and wind energy, have developed rapidly and become major contributors. Nevertheless, the supply of these energy sources is periodic and unstable because of natural environmental variations, and hence, energy-storage and conversion devices become a topic of intense research interest [1], these including lithium batteries, electrochemical capacitors, and dielectric capacitors. For practical applications, energy-storage and conversion devices are expected to have high energy density, high power density, high conversion efficiency, wide operating temperature range, environmental friendliness, and low cost.

The energy and power density requirements for common energy-storage devices are shown in Fig. 1 [2]. Compared with other energy-storage devices, dielectric capacitors have ultra-high power densities $> 10 \text{ kW} \cdot \text{kg}^{-1}$, and hence, they have ultra-high charging and discharging speed, which can release stored energy in the microsecond or nanosecond time scale, enabling an extremely high pulse power. Additionally, they have positive attributes including ultra-long cycle life, safety, and reliability. Thus, dielectric capacitors play a very important role in high-power energy-storage and pulse-

power systems, such as consumer electronics, advanced medical device, electric/hybrid energy vehicles, smart grids, and electromagnetic weapons [3,4]; these devices are also key to the application and development of third-generation semiconductor devices. However, currently used commercial polymer dielectric capacitors have a comparatively low energy density of $< 0.1 \text{ Wh} \cdot \text{kg}^{-1}$, which results in relatively large and heavy energy-storage and pulse-power systems. For example, the capacitors in high-voltage direct-current transmission converter valves occupy $\sim 50\%$ of the device volume and 60% of its weight, and the weight of the capacitors under each carriage of a high-speed train exceeds 50 kg . The large volume would also introduce a large equivalent series inductance (ESL), which may result in damage or even failure of semiconductor devices when quickly switched. This means that these capacitors cannot currently meet the requirements for application in electronic devices and systems that require compact and light integrated capacitors [5,6]. Here, the energy-storage properties of some commercial MLCCs are listed in Table 1.

At present, the dielectric material of the dielectric energy-storage capacitor, which is used in the main inverter of an electric vehicle, is biaxially-oriented polypropylene (BOPP). For this type of linear dielectric, its energy-storage density is $< 0.2 \text{ J} \cdot \text{cm}^{-3}$, its maximum operating temperature is $< 105 \text{ }^\circ\text{C}$, and the temperature of its long-term working environment should be $< 70 \text{ }^\circ\text{C}$ [9]. The relatively low stable working temperature severely limits the application of polymer capacitors. Hence, inorganic dielectric films have gained significant research attention in recent years. Methods such as polymorphic nanodomain design [10] and strain engineering [11] have been demonstrated to effectively

Table 1 Energy-storage properties of some commercial MLCCs

Commercial MLCCs	Energy	$E_b \text{ (MV} \cdot \text{m}^{-1}\text{)}$	$\eta \text{ (\%)}$
PLZT-based (B5803115105 M002) [7]	$93.0 \text{ mJ@500 V (} 0.69 \text{ J} \cdot \text{cm}^{-3}\text{)}$	—	89.5
BT-based (C1812C334K CRACTU) [7]	$10.7 \text{ mJ@500 V (} 0.37 \text{ J} \cdot \text{cm}^{-3}\text{)}$	—	62.9
C16 (C0G) [8]	0.88 J@1 kV	65	—
C11 (Z5U) [8]	0.02 J@1 kV	13.2	—
PMN-a (Z5U) [8]	0.02 J@1 kV	10	—
PMN-b (Z5U) [8]	0.02 J@1 kV	11	—
PLZT (X7T) [8]	0.71 J@1 kV	16	—
C08 (X7R) [8]	1.40 J@1 kV	30	—
C20 (X7R) [8]	1.34 J@1 kV	40	—
Exp (X7R) [8]	4.82 J@1 kV	90	—

Fig. 1 Relationship between energy density and power density among common energy-storage devices. Reproduced with permission from Ref. [2], © Elsevier Ltd. 2015.

increase the energy-storage density of dielectric films. Pan *et al.* [10] introduced multi-morphic nano-domains in BiFeO₃-system films, enabling an energy-storage density $> 112 J \cdot cm^{-3}$, which demonstrates the excellent potential of inorganic dielectric energy-storage materials. However, the low volume of the films leads to a low total energy storage, and hence, their application prospects are limited. Comparatively, thinner ceramic dielectric layers in multilayer ceramic capacitors (MLCCs) can realize larger electric capacitance and dielectric breakdown strength. The favorable structure endows the MLCC with an ultra-low equivalent series inductance. Additionally, the ceramic dielectric can withstand high temperatures, and therefore, the MLCC is considered as the most promising energy-storage dielectric capacitor.

With the goal of optimizing the composition of dielectric materials, the manufacturing process, and the structure of materials and devices, many researchers have made great effort to develop highly reliable MLCC devices with high energy densities, high energy conversion efficiencies, high power densities, high capacitances, and broad operational temperature ranges via simulations and experiments. The successful development of such MLCCs would ultimately broaden their application in the field of energy storage.

2 Energy-storage mechanism and characterizations

The storage of electric energy is the functional basis of capacitors in electronic devices. The energy-storage capability is called capacitance, which is determined by the physical geometry of the capacitor and the permittivity of the dielectric materials. When dielectric capacitors are used under an external voltage, charges with opposite sign and equal magnitude accumulate at the electrodes, referred to as the charging process (Fig. 2(a)). The charges form an internal electric field with a direction opposite to the external electric field. The internal electric field increases as the accumulated charge increases. The charging process will end when the internal electric field induced by the accumulated charges is equal to the external field. During the charging process, the charges drift under the externally applied electric field, and electrostatic energy is stored in the dielectric [12].

One of the most important parameters for evaluating the energy-storage properties of dielectric capacitors is the energy-storage density (W). The W of dielectric capacitors can be characterized in two ways. The most commonly used method is integrating the area between the polarization axis and the polarization

Fig. 2 (a) Schematic of a dielectric between two electrodes before and after applying an external voltage. Reproduced with permission from Ref. [13], © Elsevier Ltd. 2021. (b) Calculation of the energy-storage performance of a dielectric.

versus electric field curves (also named P – E loops), i.e., the blue curve shown in Fig. 2(b). The green shaded area is equal to the recoverable energy density (W_{rec}), which means the released energy density during the discharging process. The red shaded area is equal to the energy loss (W_{loss}) during the charge–discharge process. The total energy density (W_{total}) stored in the charging process is equal to the sum of W_{rec} and W_{loss} . Therefore, the parameters of the energy-storage properties can be calculated by the following equations:

$$W_{total} = \int_0^{P_{max}} E dP$$

$$W_{rec} = \int_{P_r}^{P_{max}} E dP$$

$$\eta = \frac{W_{rec}}{W_{total}} = \frac{W_{rec}}{W_{rec} + W_{loss}}$$

where P_{max} and P_r are the saturation and remnant polarizations under the applied electric field (E), respectively, and η is the energy efficiency. The above equations indicate that high saturation polarization, low remnant polarization, and high dielectric breakdown field (E_b) facilitate a higher energy density and energy efficiency in the dielectric materials. The dielectric material should have low dielectric loss, low leakage current, and high insulation to sustain the high applied electric field. Especially, the E_b depends on both internal factors (including the dielectric constant, band gap (E_g), density, defects, grain size, and grain boundary) and external factors (such as the sample area, sample

thickness and electrode shape size, temperature, humidity, type of voltage, and test duration) [12]. The thickness of dielectrics has a significant effect on E_b because of the fewer defects in thinner dielectric layer. The E_b exponentially increases as the thickness (t) of the single dielectric layer decreases [12,14–17], and it roughly follows this relationship: $E_b \propto 1/t^{0.5}$. Hence, there would be more stored energy in the MLCC with thinner dielectric and more layers.

The W_{rec} can also be obtained via charge–discharge measurements using a discharge resistor and a capacitance load circuit, as shown in Fig. 3. The dielectric capacitor is charged by the external power source. Then, the stored energy is discharged to a load resistor (R) in series with the capacitor. The current (I) passing through the resistor is then recorded as $I(t)$, where t is the time. The W_{rec} can be calculated using the following equation:

$$W_{rec} = \frac{\int I(t)^2 R dt}{V}$$

where V is the volume of the dielectric capacitor. It is worth noting that R should be much larger than the equivalent series resistor of the capacitor, so that most of the stored energy will be delivered to the load resistor and the measured energy density will be nearly the same as the stored energy density [12]. The W_{rec} calculated from P – E loops is usually higher than that calculated by the discharge current, which may be attributed to the different characterization mechanisms of the quasistatic P – E loops (1–100 Hz, near second scale) and the dynamic charge–discharge method (near microsecond or sub-microsecond scale) with different frequencies.

Fig. 3 (a) Schematic diagram of the charge–discharge circuit. (b) Discharge current and corresponding calculated W_{rec} . Reproduced with permission from Ref. [12], © Elsevier Ltd. 2018.

3 Dielectric ceramics with excellent energy-storage properties

To obtain dielectric ceramics with excellent energy-storage properties, composition modification and structure optimization have been widely used in recent research to effectively increase the P_{max} and E_b and decrease the P_r of dielectrics, leading to high W_{rec} and high η .

3.1 Composition optimization

Based on the different polarization responses to the applied electric field, energy-storage dielectric materials can be divided into linear dielectric, ferroelectric, relaxor ferroelectric, and antiferroelectric. The relationship between the polarization and electric field is shown in Fig. 4. Linear dielectric features in high breakdown strength but low dielectric permittivity. Simple oxides or nitrides and zirconates or hafnates, such as Al_2O_3 , Si_3N_4 , $CaZrO_3$, and $CaHfO_3$, respectively, are always linear dielectrics. Ferroelectric ceramics possess a high dielectric permittivity because of their high spontaneous polarization, which are widely used as dielectrics in MLCCs. However, their high remnant polarization and low breakdown strength lead to a relatively low recoverable energy density, which is not suitable for energy-storage applications. A common approach for ferroelectric ceramic modification is to disturb the long-range order of the domains, resulting in the formation of polar nano regions (PNRs), which is referred to as relaxor ferroelectrics. The PNR is highly activated, resulting in “slim” hysteresis loops with a high recoverable energy density and a relatively low energy loss, and these characteristics are suitable for energy-storage applications. Antiferroelectric features in double hysteresis loops with low remnant polarization

have recently drawn significant attention for energy-storage applications. However, there are few lead-free antiferroelectric ceramic systems, and thus, further in depth studies on antiferroelectric ceramics are needed.

The lead-based ceramics usually exhibit excellent energy storage properties by constructing field-induced multiphase transitions and decreasing grain size. Wang *et al.* [18] studied the energy-storage properties of $(Pb_{0.98}La_{0.02})(Zr_{0.55}Sn_{0.45})_{0.995}O_3$ (PLZS) antiferroelectric ceramics. An ultra-high W_{rec} of $10.4 \text{ J}\cdot\text{cm}^{-3}$ and a high η of 87% were achieved at $40 \text{ MV}\cdot\text{m}^{-1}$. Similarly, Liu *et al.* [19] studied the $(Pb_{0.98-x}La_{0.02}Sr_x)(Zr_{0.9}Sn_{0.1})_{0.995}O_3$ (PLSZS) antiferroelectric ceramics, which possessed an ultra-high W_{rec} of $11.18 \text{ J}\cdot\text{cm}^{-3}$ and a high η of 82.2% under $39.5 \text{ MV}\cdot\text{m}^{-1}$. These lead-based materials could possess both ultrahigh W_{rec} and high η , which was difficult to be achieved in other lead-free energy storage ceramics. However, the lead-based ceramic would cause serious environmental pollution due to toxicity of lead element. Hence, it is urgent to develop the lead-free dielectric materials. This section mainly introduces the compositional optimization of several common lead-free nonlinear dielectric ceramics, such as $BaTiO_3$ -based, $K_{0.5}Na_{0.5}NbO_3$ -based, $BiFeO_3$ -based, $Bi_{0.5}Na_{0.5}TiO_3$ -based, $AgNbO_3$ -based, and $NaNbO_3$ -based energy-storage ceramics, covering ferroelectric, relaxor ferroelectric, and antiferroelectric ceramics.

3.1.1 $BaTiO_3$ -based

Barium titanate, $BaTiO_3$, is one of the most studied and widely used lead-free dielectric ceramics, and it exhibits a paraelectric cubic phase above the Currie temperature (T_C) of 125°C and a ferroelectric tetragonal phase near room temperature. The macrodomains and typical ferroelectricity of $BaTiO_3$ ceramics lead to a high P_r and very low η . Most studies on $BaTiO_3$ -based bulk

Fig. 4 Schematic diagram of the hysteresis loops (polarization as a function of electric field) for: (a) linear dielectrics, (b) ferroelectrics, (c) relaxor ferroelectrics, and (d) antiferroelectrics.

ceramics attempted to improve the E_b and decrease the P_r via reducing grain size, coating dielectrics with low permittivity and high insulation, or doping the A/B site ions in a perovskite lattice. Grain size reduction and grain core-shell structure construction will be introduced in detail in Section 3.2 “Structure optimization”.

A/B-site ion doped BaTiO_3 ceramics are usually obtained via the addition of La, Ca, Zr, Sr, or BiMeO_3 (Me symbolizes trivalent or average trivalent metallic ions). Generally, the compositions of $(\text{Ba},\text{La})\text{TiO}_3$, $(\text{Ba},\text{Ca})\text{TiO}_3$, $\text{Ba}(\text{Zr},\text{Ti})\text{O}_3$, and $(\text{Ba},\text{Ca},\text{Sr})(\text{Ti},\text{Zr})\text{O}_3$ cannot yield an appreciable W_{rec} of $> 1 \text{ J} \cdot \text{cm}^{-3}$ [20–24]. The addition of BiMeO_3 could destroy the long-range ferroelectric order in BaTiO_3 and form PNRs with short-range order in the solid solutions. Ogihara *et al.* [25,26] studied the relaxor characteristics of the $\text{BaTiO}_3\text{--BiScO}_3$ (BT–BS) ceramics. The activation energy for relaxation for BT–BS ceramics is very high, and this type of relaxor ferroelectric is termed as a “weakly coupled relaxor”. The PNRs in weakly coupled relaxor ferroelectrics are isolated and randomly located in the matrix, and the interactions between the PNRs are very weak. Therefore, the PNRs possess fast responses under the applied electric field, resulting in limited spontaneous polarization, lower P_r , narrower hysteresis loops, and higher η . The 0.7BT–0.3BS ceramics had a W_{rec} of $2.3 \text{ J} \cdot \text{cm}^{-3}$ at an E_b of $22.5 \text{ MV} \cdot \text{m}^{-1}$. Shen

et al. [27] studied $\text{BaTiO}_3\text{--BiYbO}_3$ ceramics, and they observed that with increasing BiYbO_3 content, there was a gradual change from a normal ferroelectric behavior to highly diffusive and dispersive relaxor-like characteristics, and the nonlinearity of the $(1-x)\text{BaTiO}_3\text{--}x\text{BiYbO}_3$ (BT–BY) ceramics was markedly weakened. The 0.91BT–0.09BY ceramics exhibited a maximum W_{rec} of $0.71 \text{ J} \cdot \text{cm}^{-3}$ with a η of 82.6% under $9.3 \text{ MV} \cdot \text{m}^{-1}$.

Many other $\text{BaTiO}_3\text{--BiMeO}_3$ solid solutions are also weakly coupled relaxor ferroelectrics, where Me represents average trivalent B-sites for two or more cations, such as $\text{BaTiO}_3\text{--Bi}(\text{Zn}_{2/3}\text{Nb}_{1/3})\text{O}_3$ (BT–BZN) [28–30], $\text{BaTiO}_3\text{--Bi}(\text{Zn}_{0.5}\text{Zr}_{0.5})\text{O}_3$ (BT–BZZ) [31], $\text{BaTiO}_3\text{--Bi}(\text{Mg}_{0.5}\text{Zr}_{0.5})\text{O}_3$ (BT–BMZ) [32,33], $\text{BaTiO}_3\text{--Bi}(\text{Mg}_{2/3}\text{Nb}_{1/3})\text{O}_3$ (BT–BMN) [34,35], $\text{BaTiO}_3\text{--Bi}(\text{Zn}_{0.5}\text{Sn}_{0.5})\text{O}_3$ (BT–BZS) [36], $\text{BaTiO}_3\text{--Bi}(\text{Zn}_{2/3}(\text{Nb},\text{Ta})_{1/3})\text{O}_3$ (BT–BZNT) [37], and $\text{BaTiO}_3\text{--Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ (BT–BMT) [38,39]. The energy-storage properties of $\text{BaTiO}_3\text{--Bi}(\text{Zn}_{2/3}\text{Nb}_{1/3})\text{O}_3$ (BT–BZN) ceramics were studied, and a maximum W_{rec} of $0.79 \text{ J} \cdot \text{cm}^{-3}$ with a η of 93.5% under $13.1 \text{ MV} \cdot \text{m}^{-1}$ was achieved for the 0.85BT–0.15BZN ceramic. The $\text{BaTiO}_3\text{--Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$ (BT–BMT) ceramics were prepared via nano-scale polarization mismatch and reconstruction [38], as shown in Fig. 5. With increasing BMT concentration, the displacement

of Bi^{3+} cations could be sufficiently established and lead to an enhanced $P_{\max}$, which was because of the large polarizability of the Bi^{3+} ion caused by the 6s lone-pair electrons [12]. Additionally, there are numerous Ti-rich and Bi-rich PNRs that coexist in the matrix, which can respond quickly to an electric field and yield a high η . This enabled the realization of an ultra-high W_{rec} of $4.49 \text{ J}\cdot\text{cm}^{-3}$ with a η of 93% under $34 \text{ MV}\cdot\text{m}^{-1}$ for the 0.6BT–0.4BMT ceramic. However, the average ceramic grain size in this system was much larger at $> 8 \mu\text{m}$, which was unfavorable for miniaturization of the MLCCs.

Yuan *et al.* [33] studied $\text{BaTiO}_3\text{--Bi}(\text{Mg}_{0.5}\text{Zr}_{0.5})\text{O}_3$ (BT–BMZ) ceramics. They determined that the dynamic PNRs along with the decreased grain size, increased bulk density, and consequently enhanced E_b were responsible for the enhanced energy-storage properties of 0.85BT–0.15BMZ ceramic, leading to a high W_{rec} of $2.9 \text{ J}\cdot\text{cm}^{-3}$ with a η of 86.8% under $30.14 \text{ MV}\cdot\text{m}^{-1}$.

Subsequently, bioinspired raspberry-like hierarchically structured nanocomposites have been prepared by enclosing micro-sized BT–BMZ relaxor ferroelectrics using core–shell BT–BMZ@ SiO_2 nanoparticles [33], as shown in Fig. 6. The synergistic effects of the bioinspired hierarchical structure and the insulating SiO_2 resulted in a significantly improved dielectric breakdown strength and a sustained large polarization, which could be corroborated via experimental characterizations and theoretical simulations. As a result, a high W_{rec} of $3.41 \text{ J}\cdot\text{cm}^{-3}$ and a η of 85.1% were achieved in 93wt%BT–BMZ/7wt%BT–BMZ@ SiO_2 (BTBMZ–7BTBMZS) ceramic under $34.5 \text{ MV}\cdot\text{m}^{-1}$, along with good thermal stability.

It is worth noting that to decrease the MLCC cost, BaTiO_3 -based dielectric ceramics must have anti-reduction properties, so that they can be co-fired with the Ni internal electrodes in a reducing atmosphere. To sinter in a reducing atmosphere, the BaTiO_3 powders

Fig. 5 (a–g) Schematic diagram of the nano-scale polarization mismatch and reconstruction strategy. (h) SEM image for 0.6BT–0.4BMT ceramic and the grain size distribution. Reproduced with permission from Ref. [38], © Elsevier Ltd. 2019.

Fig. 6 Schematic of BT–BMZ-based raspberry-like hierarchically structured ceramics. (a) P – E loops and (b) energy-storage properties at room temperature for ceramics with various BT–BMZ@ SiO_2 contents. Reproduced with permission from Ref. [33], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2020.

are usually doped with MgO, MnO₂, or rare earth oxides. Zhao *et al.* [40] studied the influence of Al₂O₃ on the energy-storage properties of anti-reduction BaTiO₃-based ceramics prepared via a uniform chemical coating method. Coating with Al₂O₃ achieved not only a smaller grain size (140 nm) and a narrower distribution but also higher E_b , W_{rec} , and η at ambient temperature, as shown in Fig. 7. The BaTiO₃-based ceramics coated with 1.2 mol% Al₂O₃ had an enhanced W_{rec} of 0.51 J·cm⁻³ and a η of 79.6% under 9.1 MV·m⁻¹. Although the W_{rec} value was relatively low,

the anti-reduction properties allow this BaTiO₃-based ceramic to have practical significance for future application in energy-storage Ni-electrode MLCCs.

In summary, BaTiO₃-based ceramics can synergistically achieve both a high W_{rec} and a high η , which are promising for energy-storage capacitors. Of particular significance is that the BaTiO₃-BiMeO₃ dielectric ceramics exhibit relatively high permittivity and energy density over the temperature range from -50 to 150 °C with good temperature stability because of the diffuse phase transition around the dielectric maxima. Specifically,

Fig. 7 (a) Al, Mn, and Ti element distribution in the BaTiO₃-based ceramics with Al₂O₃ coating. (b) Energy-storage properties of the BaTiO₃-based ceramics without and with an Al₂O₃ coating. Reproduced with permission from Ref. [40], © The American Ceramic Society 2017.

Table 2 Energy-storage properties of BT-based bulk ceramics

Dielectric ceramic	W_{rec} (J·cm ⁻³)	E_b (MV·m ⁻¹)	η (%)
0.7BT-0.3BS [25]	2.3	22.5	—
0.91BT-0.09BY [27]	0.71	9.3	82.6
0.85BT-0.15BZN [28]	0.79	13.1	93.5
0.87BT-0.13BZNT [37]	1.44	21.8	92.5
0.85BT-0.15BMZ [32]	2.9	30.14	86.8
BTBMZ-7 wt% BTBMZ@SiO ₂ [33]	3.41	34.5	85.1
0.9BT-0.1BMN [35]	1.13	14.35	95.8
0.88BT-0.12Bi(Ni,Nb)O ₃ [41]	2.09	20	95.9
0.9BT-0.1Bi(Zn,Zr)O ₃ [31]	2.46	26.4	86.8
0.9BT-0.1Bi(Mg,Nb)O ₃ [42]	1.7	21	90
0.85BT-0.15Bi(Zn,Sn)O ₃ [36]	2.41	23	91.6
0.9BT-0.1BiInO ₃ [43]	0.753	~14	89.4
0.6BT-0.4BMT [38]	4.49	34	93
0.88BT-0.12BMT [39]	1.81	22.4	~88
0.88BT-0.12Bi(Li,Nb)O ₃ [44]	2.032	27	88
0.9BT-0.1Bi(Li,Ta)O ₃ [45]	2.2	28	88.1
0.86BT-0.14Bi(Zn,Ti)O ₃ [46]	0.81	12	94
BT-based@Al ₂ O ₃ [40]	0.51	9.1	79.6

BaTiO₃-based dielectric ceramics with anti-reduction properties can co-fire with base metal internal electrodes (Ni, Cu), which would significantly reduce the cost of multilayer ceramic capacitors and be suitable for practical power electronics and pulsed power technology applications.

3.1.2 *K_{0.5}Na_{0.5}NbO₃-based*

Potassium–sodium niobate, K_{0.5}Na_{0.5}NbO₃ (KNN), is a solid solution formed between ferroelectric KNbO₃ and antiferroelectric NaNbO₃. With increasing temperature, KNN ceramics show several phase transitions, i.e., the rhombohedral phase (R, < −190 °C), the orthorhombic phase (O, −190–220 °C), the tetragonal phase (T, 220–420 °C), and the cubic phase (C, > 420 °C) [47]. KNN-based ceramics usually have a large ferroelectricity with good piezoelectric properties, which are not suitable for energy-storage applications. The addition of SrMO₃ (*M* is tetravalent or average tetravalent metallic ions, such as Ti or Sc_{0.5}Nb_{0.5}) or BiMeO₃ into KNN would form a KNN-based solid solution [48–53]. With increasing content of the second component, there are a shift of *T_C* towards low temperature and a broadening of the dielectric permittivity peak, which indicate that the relaxor behavior of KNN-based solid solutions is strengthened. Through compositional modification and grain size engineering, the breakdown strength (*E_b*) and the response relationship between the polarization (*P*) and external electric field (*E*) of

KNN-based relaxor ferroelectrics would be improved, which leads to better energy-storage properties.

The addition of Sr(Zn_{1/3}Nb_{2/3})O₃ (SZN) into KNN leads to diffuse phase transitions and a typical relaxor-like behavior [52]. A uniform grain size < 200 nm and a dense microstructure were prepared via a conventional sintering method. The 0.94KNN–0.06SZN possesses a *W_{rec}* of 1.5 J·cm^{−3} with a *η* of 50% under 18 MV·m^{−1}, and a high optical transparency. Similarly, a high *W_{rec}* of 2.02 J·cm^{−3} with a *η* of 81.4% under 29.5 MV·m^{−1}, and a high optical transparency were achieved for the 0.8KNN–0.2Sr(Sc_{0.5}Nb_{0.5})O₃ (0.8KNN–0.2SSN) ceramic with a uniform grain size of ~500 nm [49]. Introducing ZnO into the 0.8KNN–0.2SSN ceramic could form a liquid phase to promote densification, which reduced the sintering temperature by ~100–200 °C and led to lower porosity and a higher *E_b* [54]. An enhanced *W_{rec}* of 2.6 J·cm^{−3} with a *η* of 73.2% and *E_b* of 40 MV·m^{−1} was achieved in 0.8KNN–0.2SSN–0.5 mol% ZnO ceramic, as shown in Figs. 8(a)–8(c). The (1−*x*)KNN–*x*SrTiO₃ (KNN–ST) ceramics were prepared via a strategy where the composition drives the grain size to the submicrometer scale [55]. With increasing ST content, the average grain size decreased from 2.26 μm for pure KNN to 0.35 μm for 0.85KNN–0.15ST and 0.28 μm for 0.8KNN–0.2ST, and the ferroelectric properties of the (1−*x*)KNN–*x*ST ceramics were also dramatically reduced. The fine nanograin led to a markedly large *E_b* of 40 MV·m^{−1}.

Fig. 8 *P*–*E* loops and energy-storage properties at room temperature of (a–c) 0.8KNN–0.2SSN–0.5%ZnO and (d–f) 0.85KNN–0.15ST. Reproduced with permission from Refs. [54,55], © The Royal Society of Chemistry 2016.

A large W_{rec} of $4.03 \text{ J}\cdot\text{cm}^{-3}$ with a η of 52% was obtained for 0.85KNN–0.15ST (Figs. 8(d)–8(f)), and a large W_{rec} of $3.67 \text{ J}\cdot\text{cm}^{-3}$ with a high η of 72.1% was obtained for 0.8KNN–0.2ST.

The addition of BiFeO_3 (BF) into KNN also reduced the grain size and enhanced the relaxation behavior [48]. The 0.9KNN–0.1BF ceramic with an average grain size of 150 nm possesses a W_{rec} of $2.0 \text{ J}\cdot\text{cm}^{-3}$ with a η of 61% under E_b of $20.6 \text{ MV}\cdot\text{m}^{-1}$. Moreover, the addition of BF also enhanced the bending strength, elastic modulus, and compression strength of 0.9KNN–0.1BF ceramic by $\sim 200\%$ compared with the pure KNN ceramics, which benefit the lifetime and reliability of the capacitors. The addition of $\text{Bi}(\text{Mg}_{2/3}\text{Nb}_{1/3})\text{O}_3$ (BMN) into KNN ceramics was also studied [51]. Through grain size engineering and hybridization between the Bi 6s and O 2p orbitals, the E_b and

saturation polarization (P_s) of the KNN–BMN ceramics were enhanced. A large P_s of $41 \mu\text{C}\cdot\text{cm}^{-2}$ and E_b of $30 \text{ MV}\cdot\text{m}^{-1}$ were obtained in the 0.9KNN–0.1BMN ceramic, leading to a large W_{rec} of $4.08 \text{ J}\cdot\text{cm}^{-3}$ with a η of 62.7%. Similarly, CuO was used as a sintering aid in the 0.9KNN–0.1BMN ceramic to assist in densification through the liquid phase during sintering [50], which could reduce the sintering temperature from 1150 to 940°C . A large W_{rec} of $4.02 \text{ J}\cdot\text{cm}^{-3}$ and a η of 57.3% under an E_b of $40 \text{ MV}\cdot\text{m}^{-1}$ were obtained in the 0.9KNN–0.1BMN–1 mol% CuO ceramic.

The two-step sintering reported by Chen and Wang [56] is widely used to control the grain size and prepare nanosized ceramics. In this work, by optimizing the grain size and grain boundary morphology via the two-step sintering method (Figs. 9(a)–9(e)), excellent

Fig. 9 (a) Schematic diagram of the two-step sintering method and corresponding ceramic morphologies. SEM and high-resolution TEM images of the ceramics after grinding for (b, d) 8 h and (c, e) 20 h. (f) Weibull distribution, (g) P – E loops, and (h, i) energy-storage properties at room temperature for the 0.975KNN–0.075BZTN ceramic with various grinding durations. Reproduced with permission from Ref. [53], © The Chinese Ceramic Society 2020.

energy-storage properties with both a large W_{rec} of $4.02 \text{ J}\cdot\text{cm}^{-3}$ and a high η of 87.4% under an E_b of $30.7 \text{ MV}\cdot\text{m}^{-1}$ were obtained for the 0.925KNN–0.075Bi(Zn_{2/3}(Ta_{0.5}Nb_{0.5})_{1/3})O₃ (0.975KNN–0.075BZTN) ceramic with an average grain size of 250 nm [53] (Figs. 9(h) and 9(i)).

In summary, most KNN-based relaxor ferroelectrics usually possess a relatively high E_b ($> 30 \text{ MV}\cdot\text{m}^{-1}$) and large W_{rec} ($2\text{--}4 \text{ J}\cdot\text{cm}^{-3}$). However, the strong ferroelectricity and high remnant polarization (P_r) of most KNN-based ceramics lead to their low η ($< 75\%$), which is not suitable for energy-storage capacitors. Reducing the P_r and simultaneously achieving high energy-storage density and high energy efficiency in KNN-based relaxor ferroelectric ceramics are key challenges for practical capacitor applications.

3.1.3 BiFeO₃-based

BiFeO₃-based (BF) ceramics with a giant $P_s \approx 100 \mu\text{C}\cdot\text{cm}^{-2}$, high Curie temperature, relative low sintering temperature, and a similar electron configuration for Bi³⁺ and Pb²⁺ are considered to be desirable candidates for energy-storage applications. However, there are still several drawbacks that BF-based ceramics need to overcome, such as large remnant polarization (P_r), undesirable breakdown strength (E_b), relatively large dielectric loss, and high conductivity that derives from the volatilization of Bi and the reduction of Fe³⁺ during the sintering process. To address these issues, numerous strategies for optimizing the energy-storage properties have been investigated, and they mainly focused on introducing multivalent elements to promote polar nanoregions and reducing defects to obtain an improved E_b , and most of the modified BF-based

ceramics are presented in the form of BF–BT-based ceramics.

Enhanced energy-storage properties have been achieved in modified BF-based relaxor ferroelectric ceramics. Qi *et al.* [57] proposed a nanodomain-engineering strategy to integrate a high-spontaneous-polarization gene, a wide band gap, and a heterogeneous nanodomain structure. Excellent comprehensive properties that include a giant W_{rec} of $8.12 \text{ J}\cdot\text{cm}^{-3}$ with a high η of 90% at $36 \text{ MV}\cdot\text{m}^{-1}$, along with an excellent thermal stability ($\pm 10\%$, $-50\text{--}250^\circ\text{C}$) were achieved in 0.57BiFeO₃–0.33BaTiO₃–0.1NaNbO₃ (0.57BF–0.33BT–0.1NN). The energy-storage properties and nanodomain characterizations are shown in Fig. 10. Similarly, Yan *et al.* [58] designed a $(1-x)(0.67\text{BiFeO}_3\text{--}0.33\text{BaTiO}_3)\text{--}x\text{Na}_{0.73}\text{Bi}_{0.09}\text{NbO}_3$ ((1– x)BFBT– x NBN) ceramic system, taking the advantage of the large band gap of NN to improve the E_b and disturbed the long-range ferroelectric order via substitution with low valence ions at the A-sites and high valence ions at the B-site to optimize the ΔP ($P_{\text{max}} - P_r$). They delivered a large W_{rec} ($> 5 \text{ J}\cdot\text{cm}^{-3}$) and a considerable η of $\sim 80\%$ with a high E_b ($\geq 38 \text{ MV}\cdot\text{m}^{-1}$) for the (1– x)BFBT– x NBN ceramics with a broad composition range of $x = 0.10\text{--}0.14$.

For some applications under a low electric field ($< 20 \text{ MV}\cdot\text{m}^{-1}$), BF-based ceramics are ideal candidates because of their giant polarization. Ji *et al.* [59] introduced BT–BiMeO₃ perovskites to frustrate the long-range ferroelectric order and improve the poor electrical insulation of BF. Consequently, optimal energy-storage properties with a high W_{rec} of $3.06 \text{ J}\cdot\text{cm}^{-3}$ and an excellent η of 92% were obtained at a relatively low electric field of $18.5 \text{ MV}\cdot\text{m}^{-1}$ in 0.35BiFeO₃–0.65(0.85BaTiO₃–0.15Bi(Sn_{0.5}Zn_{0.5})O₃). The details of various BF-based relaxor ferroelectric energy-storage ceramics are listed in Table 4. These results indicate that BF-based relaxor ceramics have excellent potential for high-energy-storage applications serving both low and high electric fields.

3.1.4 Bi_{0.5}Na_{0.5}TiO₃-based

Bi_{0.5}Na_{0.5}TiO₃ (BNT) is a conventional perovskite ceramic system that has been extensively studied since its discovery in the 1960s. Early research mainly focused on its piezoelectric properties; in recent years, it has also demonstrated excellent potential for dielectric energy-storage applications and attracted significant research attention. BNT ceramics have a relatively high

Table 3 Energy-storage properties of KNN-based bulk ceramics

Dielectric ceramic	W_{rec} ($\text{J}\cdot\text{cm}^{-3}$)	E_b ($\text{MV}\cdot\text{m}^{-1}$)	η (%)
0.94KNN–0.06SZN [52]	1.5	18	50
0.8KNN–0.2SSN [49]	2.02	29.5	81.4
0.8KNN–0.2SSN–0.5 mol% ZnO [54]	2.6	40	73.2
0.85KNN–0.15ST [55]	4.03	40	52
0.8KNN–0.2ST [55]	3.67	40	72.1
0.9KNN–0.1BF [48]	2.0	20.6	61
0.9KNN–0.1BMN [51]	4.08	30	62.7
0.9KNN–0.1BMN–1 mol% CuO [50]	4.02	40	57.3
0.975KNN–0.075BZTN [53]	4.02	30.7	87.4

Fig. 10 (a–f) Nanodomain structures of 0.57BF–0.33BT–0.1NN ceramic. (g) P – E loops and dP/dE – E loops at 10 Hz. (h) Recoverable energy density and efficiency as functions of the applied electric field. Reproduced with permission from Ref. [57], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019.

Table 4 Energy-storage properties of BF-based relaxor ferroelectric bulk ceramics

Dielectric ceramic	W_{rec} ($\text{J}\cdot\text{cm}^{-3}$)	E_b ($\text{MV}\cdot\text{m}^{-1}$)	η (%)
0.88BFBT–0.12NBN [58]	5.57	41	80
0.35BF–0.65BTBSZ [59]	3.06	18.5	92
0.57BF–0.33BT–0.1NN [57]	8.12	36	90
0.56BF–0.3BT–0.14AN [60]	2.11	19.5	84
BF–SBT [61]	2.01	18	75
BLF–BT–Mn [62]	1.08	13	49
0.75BNF–0.25BT [63]	1.82	18	45
BF–BZT [64]	2.56	16	75
BBFTO–SAN [65]	1.75	15.5	81
BF–BT–0.12SBT [66]	1.74	15	74
BF–BST–LMN [67]	3.38	23	59

Note: 0.75BNF–0.25BT: $0.75(\text{Bi}_{1-x}\text{Nd}_x)\text{FeO}_3$ – 0.25BaTiO_3 ; BZT: $\text{Ba}(\text{Zn}_{1/2}\text{Ta}_{2/3})\text{O}_3$.

Curie temperature (320°C) and they are ferroelectric materials with a rhombic symmetry at room temperature. More importantly, the similar lone pair electronic $6s^2$ configuration of the Bi^{3+} ion and the distorted perovskite structure is beneficial for simultaneously delivering a large polarization along with macroscopic relaxor characteristics. However, the low E_b and large P_r

of the pure BNT ceramics make it difficult to obtain high W_{rec} and η . To solve these issues, numerous studies have focused on constructing a morphotropic phase boundary (MPB) or doping with other elements, and various ceramic systems have been developed, such as the BNT– $\text{Sr}(\text{Ti}/\text{Zr})\text{O}_3$ -based (BNT–ST/Z), BNT– NaNbO_3 -based (BNT–NN), and BNT– BaTiO_3 -based (BNT–BT) systems.

It is well known that BNT has a relaxor feature between 200°C (T_s) and 320°C (Curie temperature, T_m). Therefore, by decreasing the T_s of BNT below room temperature, its P_r can be reduced and W_{rec} can be improved. Qiao *et al.* [68] introduced La^{3+} into the Sr^{2+} site of the BNT–ST ceramics: this is because La^{3+} doping plays a role in decreasing the grain size and delaying saturation of the polarization electric field in BNT-based ceramics, as shown in Fig. 11(a). With increasing $\text{Sr}_{0.7}\text{La}_{0.2}\text{TiO}_3$ (SLT) content, the crystal structure transforms from a rhombohedral phase to a weakly polarized pseudo-cubic phase, and the T_s shifts from 200°C to approximately room temperature, with enhanced relaxation behavior and improved E_b . The sample with $x = 0.45$ had a large W_{rec} of $4.14\text{ J}\cdot\text{cm}^{-3}$, a high η of 92.2%, excellent frequency stability, and excellent fatigue resistance.

Fig. 11 (a) Schematic diagram for simultaneously achieving large W_{rec} and high η : the E_b increased via decreasing grain size and increasing band gap; the relaxation behavior was enhanced by driving T_s to room temperature; and the electric field of polarization saturation was delayed by reducing the tolerance factor. Reproduced with permission from Ref. [68], © Elsevier B.V. 2020. (b) Schematic diagram of the energy-storage properties for various types of dielectric ceramics: from the P - E loops for purely linear nonpolar dielectric, relaxor ferroelectric, and normal antiferroelectric ceramics (left), to the P - E loops expected for a relaxor antiferroelectric ceramic (right). Reproduced with permission from Ref. [69], © The Royal Society of Chemistry 2019.

Additionally, defect control is another effective approach to improve the comprehensive energy-storage properties. Yan *et al.* [70] adopted a strategy via A-site defect engineering with Bi-excess and Na-deficiency to reduce the generation of oxygen vacancies, designing a ceramic with a composition of $0.75\text{Bi}_{(0.5+x)}\text{Na}_{(0.5-x)}\text{TiO}_3-0.25\text{SrTiO}_3$ (BNST- x). Bi plays a major role in the volatilized component, with the volatilization temperature of Bi_2O_3 ($\sim 825^\circ\text{C}$) during the sintering process being much lower than that of Na_2O ($\sim 1132^\circ\text{C}$) in BNT-based ceramics. Defect control via moderating the Bi/Na ratio is beneficial for inhibiting grain growth and improving E_b . An ultra-high W_{rec} of $5.63\text{ J}\cdot\text{cm}^{-3}$ and an outstanding η of 94% were simultaneously achieved at a relatively high electric field of $53.5\text{ MV}\cdot\text{m}^{-1}$ in the BNST-0.08 ceramic.

Furthermore, temperature stability is crucial for reliable operation of dielectric capacitors in complex environments. Notably, the BNT-based relaxor ceramics are considered to be promising for thermally-stable dielectric capacitor applications because of their diffuse phase transition behavior. Zhou *et al.* [71] developed an ergodic relaxor ferroelectric composed of

$0.8\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3-0.2\text{NaTaO}_3$ (0.8BNT-0.2NT), exhibiting the superior temperature stability of W_{rec} with a variation of $< 10\%$ over an ultrawide temperature range from -50 to 300°C . Zhu *et al.* [72] chose NN to modify $0.95\text{BNT}-0.05\text{SZ}$ and realized coexistence of the rhombohedral (R) and tetragonal (T) phases. The diffuse phase transition from the R phase to the T phase was responsible for the superior temperature stability of both the energy-storage and dielectric properties. A large W_{rec} of $3.14\text{ J}\cdot\text{cm}^{-3}$ was obtained in the $0.85\text{BNTSZ}-0.15\text{NN}$ ceramic at a high temperature of 120°C under $23\text{ MV}\cdot\text{m}^{-1}$, with a variation in the W_{rec} of $< \pm 4\%$ in the temperature range from 25 to 180°C under $12\text{ MV}\cdot\text{m}^{-1}$. Additionally, variation of the dielectric permittivity of the $0.8\text{BNTSZ}-0.2\text{NN}$ ceramic was $< \pm 15\%$ over the temperature range from -55 to 545°C , which is a record-high upper operating temperature, with a high room-temperature dielectric permittivity of 1170.

Recently, BNT-based relaxor antiferroelectrics have drawn significant attention because of their large spontaneous polarization, small hysteresis, and high breakdown strength. Qi and Zuo [69] introduced NaNbO_3

(NN), which is in the antiferroelectric orthorhombic phase at room temperature, into BNT to stabilize the high-temperature antiferroelectric phase, forming relaxor antiferroelectric BNT-based ceramics, as seen in Fig. 11(b). As a result, a high W_{rec} of $7.02 \text{ J}\cdot\text{cm}^{-3}$ and a desirable energy efficiency of 85% were obtained under an applied field of $39 \text{ MV}\cdot\text{m}^{-1}$, which is superior to most BNT-based ceramics. Table 5 summarizes the energy-storage properties of recently reported BNT-based ceramics.

In summary, the main constraint on the energy-storage properties in existing BNT-based ceramics can be attributed to their low breakdown strength. Modifying BNT-based ceramics to an antiferroelectric relaxor is one of the most promising approaches to simultaneously achieve high energy-storage density and good energy efficiency owing to the resulting large spontaneous polarization, small hysteresis, and high breakdown strength.

3.1.5 AgNbO₃-based

Recently, AgNbO₃ (AN) ceramics have received widespread attention as lead-free antiferroelectric materials for energy-storage applications. Through doping Bi or rare-earth elements at A-site, doping Ta and W at B-site, and adding MnO₂ as sintering aid could improve the energy storage properties of AN ceramics.

Table 5 Energy-storage properties of BNT-based relaxor ferroelectric bulk ceramics

Dielectric ceramic	W_{rec} ($\text{J}\cdot\text{cm}^{-3}$)	E_b ($\text{MV}\cdot\text{m}^{-1}$)	η (%)
0.9NBT–0.1LT [73]	3.1	20	74.2
NBT–BH [74]	2.1	17.5	68.1
BNT–BT–BZN [75]	2.83	18	67
BNST–0.08 [70]	5.63	53.5	94
BNT–SLT [68]	4.14	31.5	92.2
0.6BNT–0.4SST [76]	3.52	26	84.2
0.65(NBT–BKT)–0.35SBT [77]	4.06	35	87.3
BNT–SBT–4NN [78]	3.08	22	81.4
0.94(BNT–BST)–0.06KNN [79]	2.65	18	84.6
BNTSZNN [72]	2.93	23	72
BNT–NT [71]	4.21	38	77.8
BNT–0.4ST–0.02FN [80]	3.36	17	81
BNTST–CuO [81]	2.2	23	73.4
BLNKTANZ [82]	3.24	20	82
BNT–NN [69]	7.02	39	85

Zhao *et al.* [83] studied the Ag(Nb_{1–x}Ta_x)O₃ (ANT) solid solutions, and the Ta addition would effectively decrease the grain size and enhance the antiferroelectricity in ANT materials. As a result, a high W_{rec} of $4.2 \text{ J}\cdot\text{cm}^{-3}$ and a η of 69% were obtained under an applied field of $23 \text{ MV}\cdot\text{m}^{-1}$ in ANT15 ceramics, which was 260% enhancement over the pure AN ceramics (Figs. 12(a) and 12(b)). Gao *et al.* [84] also studied the La-doped AN ceramics. The antiferroelectric phase stability would be enhanced by the La doping, and the average grain size decreased from 5.2 to $3.3 \mu\text{m}$ with increasing La³⁺ content, leading to a higher E_b . The 2 mol% La-doped AN (ANL2) ceramics possessed the optimal energy storage properties with a high W_{rec} of $4.4 \text{ J}\cdot\text{cm}^{-3}$ and a η of 73% under an applied field of $27.3 \text{ MV}\cdot\text{m}^{-1}$ (Figs. 12(c) and 12(d)).

AgO is sensitive to light and heat, and it needs to be stored in a dark environment. Because AgO will decompose into Ag and O₂ at high temperature, the AN should be calcined and sintered in an O₂ atmosphere. These features of AN ceramics make that the AN MLCCs can only use precious metals as inner electrodes, which will greatly increase the cost of the MLCCs. Moreover, the energy efficiency of AN ceramic is usually less than 75%.

3.1.6 NaNbO₃-based

The discovery of promising relaxor or antiferroelectric properties in some derivate solid solutions, such as NaNbO₃–SrTiO₃, NaNbO₃–BiMeO₃, and NaNbO₃–Bi_{0.5}Na_{0.5}TiO₃ has resulted in NaNbO₃-based ceramics emerging as an important functional dielectric material for energy storage [89–93].

Zhou *et al.* [89] introduced Bi₂O₃ into NaNbO₃ ceramics to increase the E_b , P_{max} and decrease the P_r . The novel Na_{0.7}Bi_{0.1}NbO₃ (NBN) ceramics displayed a relaxor-like frequency dispersion near room temperature, simultaneously exhibiting a high W_{rec} of $4.03 \text{ J}\cdot\text{cm}^{-3}$ with a η of 85.4% at an E_b of $25 \text{ MV}\cdot\text{m}^{-1}$ (Figs. 13(a) and 13(b)). Zhou *et al.* [91] studied the 0.8NaNbO₃–0.2SrTiO₃ (0.8NN–0.2ST) ceramic. Because of its small grain size and dense microstructure, 0.8NN–0.2ST ceramic exhibited a W_{rec} of $3.02 \text{ J}\cdot\text{cm}^{-3}$ with a η of 80.7% at an E_b of $32.3 \text{ MV}\cdot\text{m}^{-1}$, and it had excellent energy-storage properties related to its frequency, temperature, and fatigue resistance. Similarly, BaZrO₃ and CaZrO₃ doped NaNbO₃-based antiferroelectric ceramics were also obtained [94]. The

Fig. 12 P - E loops and energy-storage properties at room temperature of (a, b) ANT [83] and (c, d) ANL [84] ceramics. Reproduced with permission from Ref. [83], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2017; Ref. [84], © The Royal Society of Chemistry 2019.

Table 6 Energy-storage properties for AN-based bulk ceramics

Dielectric ceramic	W_{rec} ($\text{J}\cdot\text{cm}^{-3}$)	E_b ($\text{MV}\cdot\text{m}^{-1}$)	η (%)
AN [83]	2.1	14	40
ANT15 [83]	4.2	23	69
ANL2 [84]	4.4	27.3	73
$\text{Ag}_{0.91}\text{Sm}_{0.01}\text{NbO}_3$ [85]	5.2	29	68
AN-0.1 wt% WO_3 [86]	3.3	20	50
$\text{Ag}_{0.97}\text{Bi}_{0.01}\text{NbO}_3$ [87]	2.6	17.5	55
$\text{Ag}_{0.88}\text{Gd}_{0.04}\text{NbO}_3$ [88]	4.5	29	64

0.9NaNbO₃-0.06BaZrO₃-0.04CaZrO₃ (0.9NN-0.06BZ-0.04CZ) ceramic possessed a W_{rec} of $1.59 \text{ J}\cdot\text{cm}^{-3}$ at an E_b of $23 \text{ MV}\cdot\text{m}^{-1}$, but the η was only ~30%. Wei *et al.* [95] chose the relaxor ferroelectric Sr_{0.7}Bi_{0.2}TiO₃ (SBT) to partially replace the NaNbO₃ ceramics, which introduced nanodomains and hindered the generation of field-induced ferroelectric phases, allowing the material to combine a large P_{max} with a near-zero P_r . This led to the realization of a large W_{rec} of $4.5 \text{ J}\cdot\text{cm}^{-3}$ and an ultra-high η of 90.3% in the 0.8NN-0.2SBT ceramic at an E_b of $28.8 \text{ MV}\cdot\text{m}^{-1}$, as shown in Figs. 13(c) and 13(d).

The NaNbO₃-BiMeO₃ ceramics have also been extensively studied, including NaNbO₃-Bi(Mg_{2/3}Ta_{1/3})O₃ (NN-BMT) [92], NaNbO₃-Bi(Zn_{0.5}Ti_{0.5})O₃ (NN-BZT) [96,97], NaNbO₃-Bi(Mg_{0.5}Zr_{0.5})O₃ (NN-BMZ) [98],

NaNbO₃-Bi(Mg_{2/3}Nb_{1/3})O₃ (NN-BMN) [99,100], NaNbO₃-Bi(Ni_{0.5}Zr_{0.5})O₃ (NN-BNZ) [101], NaNbO₃-Bi(Ni_{0.67}Ta_{0.33})O₃ (NN-BNT) [102], and NaNbO₃-Bi(Ni_{2/3}Nb_{1/3})O₃ (NN-BNN) [103]. With the addition of BiMeO₃, the long-range order in the NaNbO₃-BiMeO₃ ceramic was destroyed, leading to a random local field, polar nanoregions (PNRs), and decreased P_r . Hence, a high E_b and high energy-storage characteristics could be simultaneously obtained. The 0.85NN-0.15BNT ceramic [102] had a very high W_{rec} of $5.53 \text{ J}\cdot\text{cm}^{-3}$ and η of 82% at $57.5 \text{ MV}\cdot\text{m}^{-1}$, and the 0.78NN-0.22BMT [92] ceramic had a high W_{rec} of $5.01 \text{ J}\cdot\text{cm}^{-3}$ and η of 86.1% at $62.7 \text{ MV}\cdot\text{m}^{-1}$, as shown in Figs. 14(a) and 14(b).

Qi *et al.* [90] studied 0.76NaNbO₃-0.24Bi_{0.5}Na_{0.5}O₃ (0.76NN-0.24BNT) ceramic. The introduction of BNT disrupted the long-range ordered antiferroelectric domains, which not only enhanced the driving electric field for the antiferroelectric to ferroelectric phase transition, but also decreased the polarization hysteresis between the loading and unloading. Additionally, smaller phase switching currents would also improve the E_b . The orthorhombic $Pnma$ symmetry antiferroelectric phase and relaxor characteristics of the 0.76NN-0.24BNT ceramic contributed to the record W_{rec} of $12.2 \text{ J}\cdot\text{cm}^{-3}$ with an acceptable η of 69% at $68 \text{ MV}\cdot\text{m}^{-1}$ (Figs. 14(c) and 14(d)), as well as the good thermal

stability of the W_{rec} ($> 7.4 \text{ J} \cdot \text{cm}^{-3}$) and η ($> 73\%$) values at $45 \text{ MV} \cdot \text{m}^{-1}$ up to a temperature of 200°C .

Overall, the energy-storage properties of some recently reported NaNbO₃-based ceramics are listed in Table 7.

Introducing the appropriate relaxor end member into the NaNbO₃ ceramics could lead to the formation of relaxor ferroelectrics or antiferroelectrics with an ultrahigh E_b , W_{rec} , and acceptable η .

Fig. 13 (a) P - E loops and (b) energy-storage properties of NBN ceramic at room temperature. Reproduced with permission from Ref. [89], © The Royal Society of Chemistry 2018. (c) P - E loops and (d) energy-storage properties of 0.8NN-0.2SBT ceramic at room temperature. Reproduced with permission from Ref. [95], © Elsevier Ltd and Techna Group S.r.l. 2020.

Fig. 14 (a) P - E loops and (b) energy-storage properties of 0.85NN-0.15BNT ceramic at room temperature. Reproduced with permission from Ref. [102], © American Chemical Society 2020. (c) P - E loops, current-electric field curve under $68 \text{ MV} \cdot \text{m}^{-1}$, and (d) energy-storage properties of 0.85NN-0.15BNT ceramic at room temperature. Reproduced with permission from Ref. [90], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019.

The energy-storage properties of BaTiO₃-based, K_{0.5}Na_{0.5}NbO₃-based, BiFeO₃-based, Bi_{0.5}Na_{0.5}TiO₃-based, and NaNbO₃-based ceramics are shown in Fig. 15. Overall, the BaTiO₃-based ceramics, especially BaTiO₃-BiMeO₃ solid solutions, usually possess a very high η with acceptable W_{rec} . The K_{0.5}Na_{0.5}NbO₃-based ceramics possess a high W_{rec} , but the η was relatively low. BiFeO₃-based and Bi_{0.5}Na_{0.5}TiO₃-based ceramics show a very high W_{rec} , which can be attributed to the very high polarization caused by the high content of Bi³⁺. NaNbO₃-based ceramics usually possess an ultra-high W_{rec} . Introducing a relaxor member into Bi_{0.5}Na_{0.5}TiO₃-based and NaNbO₃-based ceramics could lead to the formation of relaxor antiferroelectric solid solutions, which also possess a relatively high η .

Table 7 Energy-storage properties of NaNbO₃-based bulk ceramics

Dielectric ceramic	W_{rec} (J·cm ⁻³)	E_b (MV·m ⁻¹)	η (%)
Na _{0.7} Bi _{0.1} NbO ₃ (NBN) [89]	4.03	25	85.4
0.8NN–0.2ST [91]	3.02	32.3	80.7
0.9NN–0.06BZ–0.04CZ [94]	1.59	23	30
0.8NN–0.2SBT [95]	4.5	28.8	90.3
0.93NN–0.07BMZ [98]	2.31	25.5	80.2
0.91NN–0.09BZT [96]	2.2	25	62.7
0.9NN–0.1BMN [99]	2.8	30	82
0.88NN–0.12BNZ [101]	4.9	50	~73
0.91NN–0.09BZT [97]	2.1	20	76
0.85NN–0.15BNN [103]	3.31	44	80.9
0.78NN–0.22BMN [100]	3.51	51	87
0.85NN–0.15BNT [102]	5.53	57.5	82
0.78NN–0.22BMT [92]	5.01	62.7	86.1
0.76NN–0.24BNT [90]	12.2	68	69

Fig. 15 Comparison of the energy-storage properties at room temperature between various lead-free non-linear dielectric ceramics [25,27,28,31–33,35–39,40–46,48–55, 57–92,94–103].

3.2 Structure optimization

Apart from composition modification, structural optimization is also an important and effective approach for enhancing the energy-storage properties of ceramic dielectrics, especially for increasing the E_b . In this section, common approaches to structure optimization, such as grain size engineering, core-shell structure construction, and layer-by-layer engineering, are reviewed to guide the structural design of new ceramic material systems.

3.2.1 Grain size engineering

The main obstacle that limits the energy-storage properties of ceramic dielectrics is the low E_b . Generally, the E_b of a ceramic dielectric is highly related to the grain size (G), and it is considered to fit the inverse power law ($E_b \propto G^{-1/2}$) [48,104]. Significant effort has been made toward realizing finer grains and thus enhancing the E_b of dielectric ceramics.

Preparing nanosized raw ceramic materials is one effective approach to obtain dielectric ceramics with fine grains. In our previous study, BT particles with an average size of 30–80 nm were fabricated via sand milling [105]. After chemical coating, dense sintered ceramics with various grain size range from 49±17 nm (BT50) to 105±44 nm (BT100) were obtained, as shown in Fig. 16. Owing to the decreasing grain size, the E_b of BT50 reached 18.5 MV·m⁻¹, which is 42% higher than that of BT100.

The grain size effect on the E_b of dielectric ceramics is modeled using a phase-field model as well [106]. Four nanostructured ferroelectric ceramic samples with different grain sizes from 50 to 125 nm were constructed via Voronoi tessellations. The final patterns of the breakdown path are presented in Fig. 17. The nominal breakdown strength of a ferroelectric ceramic with a grain size of 50 nm is almost 50% higher than for a grain size of 125 nm, and these results are in good agreement with the experimental work. As the grain size reduced, the volume fraction of the grain boundaries gradually increased. The grain boundary is the “strong” part of a ceramic under an applied electric field, and it is difficult to damage because of its higher breakdown energy. Similarly, the ceramic grain is the “weak” part and can easily be broken down compared with the grain boundary. Therefore, the enhanced E_b of the ferroelectric ceramic with smaller grains is aided by the larger number of grain boundaries.

Fig. 16 Microstructure of BaTiO₃-based ceramics with various grain sizes: (a) 49±17 nm, (b) 67±28 nm, (c) 77±34 nm, and (d) 105±44 nm. (e) Dielectric breakdown strength as a function of the grain size with the standard deviations. (f) Maximum recoverable energy density and the corresponding efficiency of the BaTiO₃-based ceramics as a function of grain size. Reproduced with permission from Ref. [105], © The American Ceramic Society 2020.

Fig. 17 Breakdown path featured for different grain sizes: (a) 50 nm, (b) 75 nm, (c) 100 nm, and (d) 125 nm. The contour lines are the equipotential lines of the then-current electric field. (e) Relation of the nominal breakdown strength against grain size. (f) Relative local electric field distribution of these ferroelectric ceramics at various grain sizes under a given applied electric field of 30 MV·m⁻¹. Reproduced with permission from Ref. [106], © The American Ceramic Society 2018.

Element doping is another effective approach to prevent the diffusion of the grain boundary, and thus, inhibit grain growth. By introducing BiFeO₃ to K_{0.5}Na_{0.5}NbO₃ ceramics, Yang *et al.* [48] found that the grain size was significantly reduced from 5.4 to 0.15 μm . Finally, a large recoverable energy density of 2 J·cm⁻³ was obtained in the 0.9K_{0.5}Na_{0.5}NbO₃–0.1BiFeO₃ ceramic at 20.6 MV·m⁻¹. Similarly, Ren *et al.* [107] introduced SrZrO₃ into the K_{0.5}Na_{0.5}NbO₃ ceramic to engineer a fine grain size. The grain size could be sharply reduced from 4.88 to 190 nm, as shown in Fig. 18. Interestingly, the obtained submicron grains not only improved the transparency of the ceramics, but also increased the E_b , and thus the recoverable energy density.

Apart from the grain size, the distribution of the grain size, or rather the grain size uniformity, can have a strong influence on the dielectric breakdown strength of the dielectric ceramics, because breakdown often occurs at the weakest point with the largest breakdown probability. The grain size distribution effect of the E_b has been studied based on the phase-field breakdown model [108]. Figures 19(a)–19(d) show four typical samples with $\alpha = 0.1, 0.3, 0.5$, and 0.7 (α is a parameter used to quantify the regularity of the 2D Voronoi tessellation) and the corresponding standard deviations (SD, representing the non-uniformity degree) of 83.7, 81.9, 80.1, and 78.7, respectively. We

can see that with increasing α , the grain distribution becomes more uniform. This conclusion can further be drawn by the distribution fittings shown in Fig. 19(e), which shows that an increase of α results in a narrower distribution of the grain size. The nominal breakdown strength of these four ceramic samples as a function of the SD is plotted in Fig. 19(f). It is clear that as the SD decreases, the distribution of the grain size becomes more uniform and the breakdown strength is distinctly enhanced. Specifically, the dielectric breakdown strength of the ceramic with an SD = 78.7 is nearly 35% higher than that of the sample with an SD = 83.7.

3.2.2 Core-shell structure

Due to the high breakdown energy, SiO₂, Al₂O₃, and other materials are often used for microstructure modification of ferroelectric ceramics to build a “core-shell” structure in the ferroelectric ceramics. The shell layer can effectively improve the E_b of dielectric ceramics. Chemical coating is a quite effective and simple method to modify the surface of ceramic particles and to form the core-shell structures [109,110]. First, ceramic particles are dispersed in aqueous or organic solutions to form uniform slurries. Then, the ceramic particles are coated by adding the doping elements into the uniform slurry in the form of inorganic solutions or organic solutions under an appropriate pH value

Fig. 18 SEM images and grain size distribution of the $(1-x)\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3-x\text{SrZrO}_3$ ceramics: (a) $x = 0$, (b) $x = 0.03$, (c) $x = 0.05$, (d) $x = 0.07$, (e) $x = 0.09$, and (f) $x = 0.11$. (g) Dependence of the mean grain size and bulk density on the composition. (h) Comparison of E_b and grain size between $0.91\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3-0.09\text{SrZrO}_3$ and other typical lead-free ceramics. Reproduced with permission from Ref. [107], © Elsevier B.V. 2020.

Fig. 19 Simulated samples generated with different α : (a) $\alpha = 0.1$, (b) $\alpha = 0.3$, (c) $\alpha = 0.5$, and (d) $\alpha = 0.7$. (e) Lognormal distribution fittings for the samples with various α . (f) Nominal electric field as a function of the standard deviation. Reproduced with permission from Ref. [108], © Author(s) 2020.

through chemical coprecipitation. After drying and calcining at a certain temperature, the ceramic powders with core-shell structure will be obtained. The key issue of chemical coating is the uniformity of the coating layer, which will influence element diffusion and local distribution of the electric field. The two-step sintering method is quite effective for controlling the grain size and grain uniformity, and this enhances the breakdown strength and reliability of the core-shell structured ceramics [111].

Zhao *et al.* [40] coated the grain surface of BaTiO_3 -based ceramics with a 2 nm-thick layer of Al_2O_3 , which allowed the E_b of the BaTiO_3 -based ceramics to increase from 7.88 to 10.85 $\text{MV} \cdot \text{m}^{-1}$, and the corresponding recoverable energy density was increased by 42%. Ma *et al.* [112] coated SiO_2 on $\text{BaTiO}_3@ \text{La}_2\text{O}_3$, and the breakdown field strength was increased by 62%. Wang *et al.* [113] co-coated SiO_2 and Al_2O_3 on $\text{Ba}_{0.4}\text{Sr}_{0.6}\text{TiO}_3$ ceramics, and the breakdown field strength was increased from 8.6 to 49.3 $\text{MV} \cdot \text{m}^{-1}$.

3.2.3 Layer-by-layer engineering

Layer-by-layer engineering is a flexible modification approach for dielectric ceramics, which can combine the advantages of two different layers and enable the

realization of comprehensive energy-storage properties. For example, in our previous study, a layer-by-layer structured ceramic was developed by combining the advantages of composition modification and structure modification of ferroelectric ceramics, and it enabled the attainment of a superimposed effect [114].

Next, a cross-section scanning electron microscopy (SEM) image of the layer-by-layer ceramic is presented (Fig. 21(c)), in which the $0.87\text{BaTiO}_3\text{--}0.13\text{Bi}(\text{Zn}_{2/3}(\text{Nb}_{0.85}\text{Ta}_{0.15})_{1/3})\text{O}_3$ (BTBZNT) layers, $\text{BaTiO}_3@3 \text{ wt}\% \text{ Al}_2\text{O}_3$, 1 wt% SiO_2 (BTAS) layers, and the interfaces can be clearly identified. The layers can also be confirmed via line scanning of the Al and Si elements in Fig. 21(b). Notably, the interface regions exhibit less porosity compared with the BTAS and BTBNZT layers, which may be caused by interdiffusion of multiple elements. The phase structure and morphology of the two surfaces are shown in Figs. 21(a), 21(d), and 21(e), respectively. The top BTBZNT layer shows a pure perovskite phase, while a second phase of BaAl_2O_4 can be observed in the bottom BTAS layer, which is in agreement with previous work [115]. The surface SEM images of the top and the bottom layers indicate the high density of sample S2, and the grain size of BTBZNT layer ($\sim 500 \text{ nm}$) is larger than that of the

Fig. 20 FE-TEM images of (a, b) $\text{BaTiO}_3@\text{La}_2\text{O}_3@\text{SiO}_2$ particles [112], (c, d) BST-2AlS particles [113], and (e, f) BaTiO_3 particles without and with Al_2O_3 [40]. Reproduced with permission from Ref. [112], © Elsevier B.V. 2016; Ref. [113], © Springer Science+Business Media New York 2013; Ref. [40], © The American Ceramic Society 2017.

Fig. 21 Microstructure characterization of the layer-by-layer ceramics with four BTBZNT layers and four BTAS layers alternately arranged in parallel. The cross-section SEM image is shown in the middle (c), in which the BTBZNT layers, BTAS layers, and the interfaces can be clearly identified. The results can be further confirmed via line scanning of the Al and Si elements in (b). The XRD patterns and surface SEM images of each end of the BTBZNT and BTAS layers are presented in (a), (d), and (e), respectively. Reproduced with permission from Ref. [114], © The Royal Society of Chemistry 2019.

BTAS layer (~ 100 nm).

By combining the complementary advantages of the two different layers and the interface effects, a record-high dielectric breakdown strength of $79 \text{ MV} \cdot \text{m}^{-1}$ was obtained in layer-by-layer structured bulk ceramic when four BTBZNT layers and four BTAS layers arrange alternately in parallel. The corresponding recoverable energy density was $5.04 \text{ J} \cdot \text{cm}^{-3}$, which was clearly much higher than that of the BaTiO_3 -based ceramics ($\sim 1\text{--}2 \text{ J} \cdot \text{cm}^{-3}$).

Recently, Yan *et al.* [116] proposed layer-by-layer engineering that combined high-polarization ferroelectric ceramics with a large grain size and the high breakdown strength of linear-like ceramics with a small grain size (Fig. 22). Based on this strategy, Yan *et al.* took advantage of the tape casting technique to design and fabricate layer-by-layer structured ceramics, and they investigated the relationship between the structure and properties in detail. The E_b could be gradually enhanced by increasing the thickness of the linear-like

Fig. 22 (a) Schematic diagram of the prepared lead-free ceramics with sandwich structures; (b) photograph of the prepared ceramic samples; (c) XRD patterns for the samples BNKSTT (S0) and BNT-SNA (S5); cross-section images of the BNKSTT layer, the BNT-SNA layer, and S3 are presented in (d), (e), and (f), respectively; and (g) line scanning of the Bi and Sr elements for S3 in (f). Reproduced with permission from Ref. [116], © The Royal Society of Chemistry 2020.

ceramic layer. Interestingly, the designed layer-by-layer ceramic reached an ultra-high recoverable energy density of $6.78 \text{ J} \cdot \text{cm}^{-3}$ and a very high efficiency of 89.7% at a high electric field of $57.2 \text{ MV} \cdot \text{m}^{-1}$.

Layer-by-layer engineering is one of the most effective approaches for the design of energy-storage ceramics because of flexible choice of the two different layers. Thus, one can combine the high-polarization layer and the high-breakdown-resistance layer together to achieve comprehensive energy-storage properties. However, the co-sintering of different layers is a major challenge and two-step sintering may be the optimal solution.

4 Energy-storage MLCCs

In this section, we introduce the preparation process and design of the inner electrode structure for energy-storage MLCCs, together with the state-of-the-art lead-free energy-storage MLCCs.

4.1 Preparation process

The detailed structure and fabrication process for the MLCCs are shown in Fig. 23 [117]. The MLCCs consist of many ceramic dielectric layers and internal

electrodes, which are alternately stacked in parallel, as shown in the center of Fig. 23. After co-sintering, metal with a low melting point was coated on both ends as external electrodes. The MLCCs were generally fabricated via the following process flow, which includes preparation of the ceramic powders, preparation of the ceramic slurry, the tape-casting process, screen printing of the internal electrodes, stacking and lamination, cutting, binder burnout and sintering, and dipping with external electrodes.

A ceramic slurry with good fluidity was prepared via ball-milling of the ceramic powders with solvent, dispersant, binder, and plasticizer in the appropriate proportion. The quality of the organic compound added into the ceramic slurry was related to the micro morphology, specific surface area, particle size distribution, and surface potential of the ceramic powders. The solvent, dispersant, and binder benefit distribution of the ceramic powders, and the binder would aid in the realization of ceramic tapes with a uniform thickness. Increasing the content of the ceramic powder and plasticizer would benefit the strength of the ceramic tapes. Ceramic powders with a narrow particle size distribution would enable the formation of flat and smooth ceramic tapes.

The ceramic slurry was coated on the polyethylene

Fig. 23 Schematic diagram of MLCCs and their fabrication process. Reproduced with permission from Ref. [117], © The Royal Society of Chemistry 2019.

terephthalate tape via a doctor blade or a casting head to form the ceramic tapes. Good ceramic tapes must be flat and have a smooth surface, a uniform distribution, a high areal density of the ceramic powder, and no macroscopic pore defects, which can increase the adaptability of the subsequent preparation process flow and ensure MLCCs with good performance.

The internal electrodes are screen printed on one side of the ceramic tapes. Common internal electrode materials can be divided into two categories, including the base metals Ni and Cu, and precious metals Pd, Pt, and Ag/Pd alloy. For practical applications, the internal electrodes need to be selected according to the ceramic sintering process and the cost. It is worth noting that the Ni and Cu internal electrodes need to be sintered in a reducing atmosphere, and the corresponding dielectric ceramics must have anti-reduction properties. The thickness of the screen-printed internal electrode layer should be adjusted according to the thickness of the ceramic tapes. This ensures that the internal electrodes in the MLCCs are complete and continuous after sintering. Additionally, the shrinkage of the dielectric ceramics and the electrodes must be matched, which would decrease internal stress in the MLCCs and improve their mechanical strength and reliability.

After lamination and warm isostatic pressing, the

stacked tapes will be cut into individual MLCC green bodies. To ensure the integrity of the MLCCs, a burnout process is required to remove the binder and prevent delamination and cracking of the MLCCs caused by rapid volatilization of the organic compounds at a high temperature. Then, the MLCC green bodies are co-fired at the ceramic sintering temperature. The heating rate, temperature, and duration of the burnout and sintering process have an important influence on the energy-storage performance of the MLCCs. After sintering and tumbling to expose the internal electrodes, an external electrode paste will be used to terminate the opposite ends of the MLCCs for electrical measurements and applications.

4.2 Electrode structure design

The energy-storage density of the ceramic dielectrics mentioned above is limited by their low E_b . The utilization of MLCCs for energy-storage applications has drawn increasing attention because the thickness of the dielectric layer is much smaller than that of bulk ceramics, which would significantly enhance the E_b of the MLCCs, and therefore, improve the energy-storage performance.

Marked efforts have been made to improve the energy-storage properties of ceramic dielectrics, while

the structure of the internal electrodes has generally been ignored, which may strongly affect the E_b of the MLCCs. Notably, the E_b plays a dominant role in the energy-storage performance.

MLCCs are fabricated with tens of thin dielectric layers arranged in parallel between internal electrodes. When an electric field is applied, the local electric field is concentrated around the tip of the internal electrodes, which favors the initiation and propagation of breakdown path inside the MLCCs and finally greatly reduces the E_b . The internal electrode structure, i.e., the margin length, would affect the distribution of the local electric field in the MLCCs. Different margin lengths would lead to different degrees of local field concentration around the electrode tips. Therefore, the design of the margin length is crucial for the MLCCs to achieve a high dielectric breakdown strength and a high energy density.

A finite element method (FEM) is widely used to model and design the microstructure of materials and devices at the macroscale. Wang *et al.* [118] calculated the distribution of the electric field via two-dimensional FEM by varying some geometric parameters of the MLCCs. The optimal geometric parameters were suggested after analysis of the concentration of the local electric field. Additionally, we calculated the

distribution of the electric field inside the MLCCs based on a thermal–mechanical–electrical coupled FEM for more precise simulation results [119].

At the mesoscopic scale, a phase-field electromechanical breakdown model was developed in our previous work to simulate the dielectric breakdown behavior in MLCCs [120–122]. The final patterns of the breakdown path inside MLCCs with various margin lengths are shown in Figs. 24(a)–24(c). Generally, the breakdown path all display similar branch-like tree patterns. The breakdown path initiates at the end of the internal electrode tip and the entire breakdown occurs when the electrodes are bridged by the breakdown pathway. Figures 24(d) and 24(e) present the nominal field–charge–density relation and the nominal breakdown strength as a function of the margin length, respectively. It is clear that as the margin length increases, the nominal breakdown strength of the MLCCs would be significantly enhanced.

The advantages of this model are that the evolution of polarization vectors and the breakdown path can be visualized and the nominal breakdown strength can be calculated as well. Further, the phase-field electromechanical breakdown model can be used to analyze the thickness-dependent, stress-dependent, and defect-dependent breakdown behavior in the MLCCs.

Fig. 24 Final patterns of the breakdown path for the selected region of MLCCs with various nominal margin lengths of (a) 50, (b) 75, and (c) 100. The shading represents the damage variable s and the arrows indicate the polarization vectors P . (d) Nominal field–charge–density relation of MLCCs with various margin lengths. (e) Nominal breakdown strength as a function of the MLCC margin length. Reproduced with permission from Ref. [121], © Author(s) 2019.

Apart from simulation of the electrical properties, Jiang *et al.* [123], Park *et al.* [124], Chen *et al.* [125], Huang *et al.* [126], and Franken *et al.* [127] independently calculated the distribution of stress in MLCCs with different internal electrode structures, obtained via various processes, including sintering, soldering, and the board flex test. In addition to leading to mechanical failure of the MLCCs, these stresses are related to the non-uniformity of electric field in the MLCCs.

The margin length dependent energy-storage performance was confirmed experimentally [121]. The 1210-type MLCC with internal 60Ag/40Pd electrodes were fabricated via the tape-casting technique. Various margin lengths of 100, 200, and 400 μm were adopted. From Figs. 25(a) and 25(b), it is clear that the hysteresis loops for these MLCCs under the same applied field were almost the same because the dielectric layer materials were totally identical. However, because of the difference in the breakdown strength, the maximum polarization measured was quite different. As the margin length increased from 100 to 400 μm , the E_b was enhanced from 78.3 to 89.5 $\text{MV}\cdot\text{m}^{-1}$ as seen in Fig. 25(c). The calculated recoverable energy density and

energy efficiency of MLCCs with various margin lengths as a function of the applied electric field is plotted in Fig. 25(d). The recoverable energy density and efficiency of various MLCCs are almost the same below an applied electric field of 64 $\text{MV}\cdot\text{m}^{-1}$. The observation that the MLCCs fabricated in our lab display a stable and reliable performance is quite positive. A large W_{rec} of 7.8 $\text{J}\cdot\text{cm}^{-3}$ can be achieved under an applied electric field of 79 $\text{MV}\cdot\text{m}^{-1}$ in the MLCC with a margin length of 400 μm , which is 30% higher than that for a margin length of 100 μm because of the enhanced breakdown strength. The experimental results clearly demonstrate the importance of the internal electrode structure to realize high-performance energy-storage MLCCs.

4.3 State-of-the-art energy-storage MLCCs

Over the last decade, energy-storage MLCCs that can realize higher energy densities because of their thin dielectric ceramic layers have received significant attention. Current reports on lead-free energy-storage MLCCs are all prototype devices and mainly divided into three systems, including BaTiO_3 -based, BiFeO_3 -based, and $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ -based MLCCs.

Fig. 25 (a) Hysteresis loops of MLCCs with various margin lengths measured under an applied electric field of 60 $\text{MV}\cdot\text{m}^{-1}$ at 1 Hz. (b) Unipolar hysteresis loops of the MLCCs with various margin lengths under their own AC breakdown strength at 1 Hz. (c) Weibull plots of the DC dielectric breakdown strength for MLCCs with various margin lengths. (d) Calculated recoverable energy density and energy efficiency of MLCCs with various margin lengths as a function of the applied electric field, measured at 1 Hz. Reproduced with permission from Ref. [121], © Author(s) 2019.

Ogihara *et al.* [25] first studied the weakly-coupled relaxor ferroelectric $0.7\text{BaTiO}_3\text{--}0.3\text{BiScO}_3$ (BT–BS) for single-layer ceramic capacitors. The BT–BS capacitors used Pt as the internal electrodes and the thickness of the dielectric ceramic layer was $15\text{ }\mu\text{m}$, which had a W_{rec} of $6.1\text{ J}\cdot\text{cm}^{-3}$ at an E_b of $73\text{ MV}\cdot\text{m}^{-1}$ (Fig. 26). It provided a good theoretical and experimental basis for the development of weakly coupled relaxor ferroelectric BT–BMO systems and energy-storage MLCCs. Then, Shay *et al.* [128] reported linear dielectric $0.8\text{CaTiO}_3\text{--}0.2\text{CaHfO}_3$ (CT–CH) single-layer ceramic capacitors, which had Pt internal electrodes and a dielectric ceramic layer of $9\text{ }\mu\text{m}$. The linear dielectric usually has a high dielectric breakdown strength and energy efficiency, and this enabled CT–CH capacitors with a W_{rec} of $9\text{ J}\cdot\text{cm}^{-3}$ and a η of 93% at an E_b of $120\text{ MV}\cdot\text{m}^{-1}$. The addition of MnO_2 could effectively decrease the dielectric loss, which led to an enhanced W_{rec} of $9.6\text{ J}\cdot\text{cm}^{-3}$ with a η of 96% at an E_b of $130\text{ MV}\cdot\text{m}^{-1}$ using the 0.5 mol% Mn-doped $0.8\text{CaTiO}_3\text{--}0.2\text{CaHfO}_3$ (CT–CH–Mn) single-layer ceramic capacitor.

Kumar *et al.* [129] studied the $\text{BaTiO}_3\text{--Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$ (BT–BZT) MLCCs with 30 $29\text{-}\mu\text{m}$ -thick dielectric

ceramic layers. The BT–BZN MLCCs with internal electrodes composed of the 40Pt/40Au/20Pd alloy had a W_{rec} of $2.8\text{ J}\cdot\text{cm}^{-3}$ at an E_b of $33\text{ MV}\cdot\text{m}^{-1}$ (Fig. 27(a)). When the MLCCs used the 70Ag/30Pd alloy for the internal electrodes, the W_{rec} was still greater than $2\text{ J}\cdot\text{cm}^{-3}$ (Fig. 27(c)). The cross-section SEM image of the MLCCs demonstrated that there were no obvious reactions between the Ag/Pd electrodes and the ceramic layers (Fig. 27(b)), proving the feasibility of the Ag/Pd alloy as internal electrodes for energy-storage MLCC applications.

$\text{BaTiO}_3\text{--Bi}(\text{Li}_{0.5}\text{Ta}_{0.5})\text{O}_3$ (BT–BLT) and $\text{BaTiO}_3\text{--Bi}(\text{Li}_{0.5}\text{Nb}_{0.5})\text{O}_3$ (BT–BLN) MLCCs with Pt internal electrodes and 13 $30\text{-}\mu\text{m}$ -thick dielectric ceramic layers were reported [46,130], and they had similar energy-storage performance. A W_{rec} of $4.05\text{ J}\cdot\text{cm}^{-3}$ with a η of 95.5% at an E_b of $45\text{ MV}\cdot\text{m}^{-1}$ and a W_{rec} of $4.5\text{ J}\cdot\text{cm}^{-3}$ with a η of 91.5% at an E_b of $46.6\text{ MV}\cdot\text{m}^{-1}$ were obtained, respectively, and the maximum operating temperature was $160\text{ }^\circ\text{C}$. Similarly, $0.94\text{BaTiO}_3\text{--}0.06\text{Bi}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (BT–BMN) MLCC with Pt internal electrodes and nine $9\text{-}\mu\text{m}$ -thick dielectric ceramic layers was reported [131], which had a W_{rec}

Fig. 26 (a) P - E loop and (b) energy density of BT–BS capacitors. Reproduced with permission from Ref. [25], © The American Ceramic Society 2009.

Fig. 27 Energy density of the BT–BZT MLCCs with (a) Pt alloy and (c) Ag/Pd alloy as the internal electrodes at various temperatures. (b) Cross-section SEM image of the MLCCs with Ag/Pd alloy as internal electrodes. Reproduced with permission from Ref. [129], © AIP Publishing LLC 2015.

of $6.88 \text{ J}\cdot\text{cm}^{-3}$ with a η of 90% at an E_b of $82 \text{ MV}\cdot\text{m}^{-1}$. As part of work carried out in our Lab, Chen *et al.* [132] studied multifunctional $0.9\text{BaTiO}_3\text{--}0.1\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{--Nb--Mn}$ (BT–BNT) MLCCs, which satisfied the EIA X9R specification (temperature coefficient of capacitance $< \pm 15\%$ within a temperature range from -55 to 200°C , Fig. 28(a)). The BT–BNT MLCCs formed using $10\text{--}30\text{-}\mu\text{m}$ -thick dielectric ceramic layers and internal electrodes composed of $60\text{Ag}/40\text{Pd}$ had excellent energy-storage performance at high temperature, with a W_{rec} of $3.33 \text{ J}\cdot\text{cm}^{-3}$ and η of 81.6% at an E_b of $48 \text{ MV}\cdot\text{m}^{-1}$ below 175°C (Fig. 28(b)).

We also prepared and investigated weakly-coupled relaxor ferroelectric $0.87\text{BaTiO}_3\text{--}0.13\text{Bi}(\text{Zn}_{2/3}(\text{Nb}_{0.85}\text{Ta}_{0.15})_{1/3})\text{O}_3$ (BTBZNT) energy-storage MLCCs using $60\text{Ag}/40\text{Pd}$ for internal electrodes [37]. The MLCCs with two dielectric ceramic layers of $5\text{ }\mu\text{m}$ each exhibited an excellent energy-storage performance with a W_{rec} of $10.12 \text{ J}\cdot\text{cm}^{-3}$ and η of 89.4% at an E_b of $104.7 \text{ MV}\cdot\text{m}^{-1}$, as well as a high temperature stability. Based on this work, we proposed a multiscale optimization strategy (including atomic scale, grain scale, and device scale designs) to increase the breakdown field strength and decrease the leakage current of the MLCCs [133]. On the atomic scale, the addition of $\text{Bi}(\text{Zn}_{2/3}(\text{Nb}_{0.85}\text{Ta}_{0.15})_{1/3})\text{O}_3$ to the BaTiO_3 system breaks the long-range polar ordering of the typical ferroelectric and generated local polar nanoregions (PNRs) of the relaxor ferroelectric (Fig. 29(c)); it exhibited high electric-field-induced polarization, low P_r , high E_b , and high η [27]. On the grain scale, introducing a SiO_2 coating layer on the BTBZNT particles to form perfect grain core–shell structures after sintering would cause the breakdown path to

dissipate more energy across the insulating SiO_2 layer with a high breakdown energy, leading to a higher E_b [134]. Through theoretical simulations via a phase-field breakdown model, it was found that the nominal breakdown strength of BTBZNT@ SiO_2 would be enhanced by 100% compared with the BTBZNT ceramics. On the device scale, the SiO_2 barrier layers were introduced at the interfaces between the electrodes and dielectrics (Fig. 29(b)), which would raise the Schottky barrier height for electron injection and decrease the leakage current and electric field concentration; hence, a marked improvement in the E_b , η , and temperature stability could be achieved. From theoretical simulations using a charge transport model related to the Schottky barrier for electron injection, the results show that the distortion of electric field and space charge density were much lower in the BTBZNT@ SiO_2 MLCC than in the BTBZNT MLCC (Figs. 29(f) and 29(g)). Furthermore, the energy-storage MLCCs would benefit from a thin dielectric layer that has a lower porosity, finer grain size, and a large capacity [16,37,135]. Based on the above multiscale optimization strategy, the relaxor ferroelectric BTBZNT@ SiO_2 MLCCs exhibited superior energy-storage performance with a large W_{rec} of $18.24 \text{ J}\cdot\text{cm}^{-3}$ and an ultra-high η of 94.5% under a high E_b of $175.5 \text{ MV}\cdot\text{m}^{-1}$, along with excellent temperature stability (variation of W_{rec} was $< 10\%$ in the temperature range from 25 to 190°C under $50 \text{ MV}\cdot\text{m}^{-1}$) and good cycling stability. Compared with the BTBZNT MLCCs without the SiO_2 coating, the W_{rec} was enhanced by 80% (Fig. 29(h)). The multiscale optimization strategy should provide a universal approach to comprehensively improve the energy-storage performance of dielectric ceramic multilayer capacitors.

Fig. 28 (a) Temperature coefficient of capacitance (TCC) and (b) W_{rec} of the BT–BNT MLCCs. Reproduced with permission from Ref. [132], © The American Ceramic Society 2018.

Fig. 29 (a) Low-magnification TEM dark-field image of BTBZNT@SiO₂ MLCCs. (b) High-resolution TEM bright-field image of the interface between the electrode and dielectric for BTBZNT@SiO₂ MLCCs. (c) Domain structure for BTBZNT@SiO₂ ceramics. (d, e) EDS analysis results of the Si and Pd distribution along the points from 1 to 24 in (d). The change in electric field concentration (f) and space charge density simulations (g) of the BTBZNT@SiO₂ and BTBZNT MLCC from 0.1 to 0.6 s after applying an electric field of 100 MV·m⁻¹. (h) Energy-storage performance of the BTBZNT and BTBZNT@SiO₂ MLCCs at room temperature. (i) Temperature-dependent energy storage properties of BTBZNT@SiO₂ MLCCs under 50 MV·m⁻¹. Reproduced with permission from Ref. [37], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2019; Ref. [133], © The Royal Society of Chemistry 2020.

Zhao *et al.* [136] prepared Ag(Nb_{0.85}Ta_{0.15})O₃–0.25 wt% MnO₂ (ANT–Mn) MLCCs. The Mn doping effectively reduced the leakage current and improved the E_b and η of the MLCCs. Moreover, the role of strain engineering in enhancing the P_s and $P_s - P_r$ values was demonstrated by the MLCCs with different

thickness of active and inactive layers (Figs. 30(a) and 30(b)). The optimal ANT–Mn MLCCs with Pt internal electrodes and 15- μ m-thick dielectric ceramic layers had a W_{rec} of 7.9 J·cm⁻³ with a η of 71% at an E_b of 102 MV·m⁻¹ (Fig. 30(c)).

Lu *et al.* [137] and Wang *et al.* [138–141] studied

Fig. 30 (a) Effect of the thickness of inactive layer (t_{inactive}) on the P - E loops and the finite-element simulations of the stress of the active layers with different t_{inactive} . (b) Effect of the thickness of active layer (t_{active}) on the P - E loops and the finite-element simulations of the stress of the active layers with different t_{active} . (c) P - E loops and energy-storage performance of ANT-Mn MLCCs at various temperatures. Reproduced with permission from Ref. [136], © The Royal Society of Chemistry 2021.

the energy-storage performance of a series of BiFeO_3 -based MLCCs. Among them, the BiFeO_3 - SrTiO_3 - 0.03Nb - $0.1\text{Bi}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (BF-ST-Nb-BMN) single-layer ceramic capacitor had the largest W_{rec} [137]. Nb doping could eliminate hole conduction and promote electrical uniformity, thereby enhancing the insulating properties of the dielectric materials. BMN-doping could reduce long range polar coupling in dielectric materials without decreasing the average ionic polarizability and enhance the relaxation properties. The capacitors used Pt as the internal electrode, and the thickness of the dielectric layer was $\sim 8 \mu\text{m}$, and it had a large W_{rec} of $15.8 \text{ J}\cdot\text{cm}^{-3}$ with a η of 75.2% at an E_b of $100 \text{ MV}\cdot\text{m}^{-1}$ (Figs. 31(a) and 31(b)). The $(0.7-x)\text{BiFeO}_3$ - 0.3BaTiO_3 - $x\text{Nd}(\text{Zn}_{0.5}\text{Zr}_{0.5})\text{O}_3$ (BF-

BT-NZZ) MLCC had the best thermal stability [139]. The BF-BT-NZZ MLCC with Pt internal electrodes and 7 16- μm -thick dielectric ceramic layers had a W_{rec} of $10.5 \text{ J}\cdot\text{cm}^{-3}$ with a η of 87% at an E_b of $70 \text{ MV}\cdot\text{m}^{-1}$. The maximum stable working temperature of the BF-BT-NZZ MLCCs could reach 150°C (Figs. 31(e) and 31(f)).

Li *et al.* [135,142] introduced a relaxor end-member into the antiferroelectrics and developed a new relaxor antiferroelectric $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ - $x\text{Sr}_{0.7}\text{Bi}_{0.2}\text{TiO}_3$ (NBT-SBT) MLCC (Fig. 32), which demonstrated excellent characteristics including a high E_b , high polarization strength, high W_{rec} , and high η . The NBT-0.45SBT MLCCs with Pt internal electrodes and 10 20- μm -thick dielectric ceramic layers had a W_{rec} of $9.5 \text{ J}\cdot\text{cm}^{-3}$ with

Fig. 31 (a) P - E loops and (b) energy-storage performance of the BF-ST-Nb-BMN MLCCs at room temperature. (c) P - E loops and (d) energy-storage performance of the BF-BT-NZZ MLCCs at room temperature. (e) P - E loops and (f) energy-storage performance of BF-BT-NZZ MLCCs at various temperatures. Reproduced with permission from Ref. [137], © The Royal Society of Chemistry 2020; Ref. [139], © The Royal Society of Chemistry 2019.

a η of 92% at an E_b of $72 \text{ MV} \cdot \text{m}^{-1}$. The NBT-0.45SBT MLCCs could operate stably in the temperature range from -60 to 120°C , and the W_{rec} was found to remain stable after 10^6 cycles at 100°C . Subsequently, Li *et al.* used $\langle 111 \rangle$ -oriented SrTiO_3 as templates to prepare $\langle 111 \rangle$ -textured NBT-0.35SBT MLCCs. Compared with nontextured samples, the $\langle 111 \rangle$ -textured samples could greatly reduce the strain of dielectric ceramics and MLCCs, thereby reducing the probability of electromechanical breakdown and enabling a higher E_b . The $\langle 111 \rangle$ -textured NBT-0.35SBT MLCCs with Pt internal electrodes and 10–20- μm -thick dielectric ceramic layers possessed the record-high W_{rec} of $21.5 \text{ J} \cdot \text{cm}^{-3}$ with a η of 80% at an E_b of $100 \text{ MV} \cdot \text{m}^{-1}$. After 10^6 cycles at 150°C , the energy-storage performance remained stable, demonstrating the excellent thermal and cycling stability. However, the preparation of the texture would result in a more complex fabrication process and greatly increase the cost of the energy-storage MLCCs. Recently, Ji *et al.* [143] introduced $\text{Bi}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ into NBT-SBT and obtained the $0.62\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3-0.3\text{Sr}_{0.7}\text{Bi}_{0.2}\text{TiO}_3-0.08\text{BiMg}_{2/3}\text{Nb}_{1/3}\text{O}_3$ (NBT-SBT-0.08BMN) ceramic. The W_{rec} of the NBT-SBT-0.08BMN ceramic reached up to $7.5 \text{ J} \cdot \text{cm}^{-3}$ with a η of 92%. Based on the NBT-SBT-0.08BMN ceramic, single-layer ceramic

capacitors were prepared with Pt internal electrodes and a dielectric ceramic layer of $7 \mu\text{m}$ (average grain size of $3 \mu\text{m}$), which had a W_{rec} of $18 \text{ J} \cdot \text{cm}^{-3}$ with a η of 93% at an E_b of $101.3 \text{ MV} \cdot \text{m}^{-1}$ (Fig. 33). Additionally, excellent energy-storage performance was also achieved with good thermal stability (up to 160°C), frequency stability, and cycling stability.

The energy-storage performance of various reported MLCCs is shown in Fig. 34 and Table 8. Most of these MLCCs possess more excellent energy-storage properties than the commercial MLCCs, such as PLZT-based (B58031I5105 M002) and BT-based (C1812C334K CRACTU) in Table 1. The BiFeO_3 -based energy-storage MLCCs usually have high maximum energy densities and low energy efficiencies, while the BaTiO_3 - BiMeO_3 system and $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ -based energy-storage MLCCs can realize both high energy densities and high energy efficiencies. However, the Bi element can easily diffuse during sintering due to its low melting point, leading to large grains in the micrometer scale [38,135,142]. Hence, the thickness of the dielectric ceramic layers of the Bi-rich MLCCs has some limits, which is not conducive for realizing miniaturized capacitors with a high capacity. Additionally, the Bi element would easily react with the Ag/Pd alloy internal electrode during the high-temperature sintering process [144] and Bi^{3+} would be

Fig. 32 (a) Schematic diagram for the preparation of relaxor antiferroelectrics. The energy-storage performance as a function of (b) the electric field, (c) the temperature, and (d) the cycles for NBT-0.45SBT MLCCs. (e–g) Cross-section SEM images, strain-electric field curves, and energy-storage performance of nontextured and <111>-textured NBT-0.35SBT MLCCs at room temperature. Reproduced with permission from Ref. [135], © WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim 2018; Ref. [142], © The Author(s), under exclusive licence to Springer Nature Limited 2020.

Fig. 33 Energy-storage performance of NBT-SBT-0.08BMN capacitors at (a) room temperature and (b) various temperatures. Reproduced with permission from Ref. [143], © Published by Elsevier B.V. 2021.

easily reduced in a reducing atmosphere [145]; therefore, most reported MLCCs use pure Pt for the internal electrodes. However, the expensive Pt significantly increases the cost of the MLCCs, which is not suitable for commercialization and practical application. There

is an urgent demand to develop energy-storage ceramic dielectric materials with anti-reduction properties and can be co-fired with the base metal electrode. In summary, current research work and the development of energy-storage MLCCs are still at the laboratory stage

and not yet ready for large-scale industrial application. Characterization and mechanism analysis related to insulator resistance degradation, fatigue characteristics, and the reliability of the MLCCs needs to be further investigated.

5 Emerging applications

The energy-storage MLCCs storage electric energy via the accumulation of charge, with functions including filtering, smoothing, coupling, decoupling, and snubbing in circuits [150]. The main applications that urgently require the miniaturization of energy-storage MLCCs are pulsed power sources and power converters. Before addressing these specific areas, we first look at how MLCCs behave in real circuits. The equivalent circuit model for MLCCs that is usually employed is described in Fig. 35(a). The capacitor is equivalently connected in series with a resistor and an inductor, with the values termed as the equivalent series resistance (ESR) and

equivalent series inductance (ESL), respectively. For ideal cases, the capacitors exhibit only capacitance with no losses; nevertheless, under practical conditions, parasitic effects linked with the resistance and inductance

Fig. 34 Comprehensive energy-storage performance of various reported MLCCs [37,46,130–133,135,137–142].

Table 8 Energy-storage performance of various MLCCs

Dielectric ceramic	Internal electrode	<i>N</i>	<i>t</i> (μm)	<i>W</i> _{rec} (J·cm ⁻³)	<i>E</i> _b (MV·m ⁻¹)	<i>η</i> (%)	Maximum operating temperature (°C)
BT-BS [25]	Pt	1	15	6.1	73	—	—
BT-BZT [129]	Pt/Au/Pd	30	29	2.8	33	—	—
BT-BNT [132]	Ag/Pd	10	30	2.76	45	84.3	175
BT-BZNT [37]	Ag/Pd	2	5	10.12	104.7	89.4	150
BT-BZNT [146]	Pt	2	5	14.08	150	69.7	150
BT-BZNT [147]	Ag/Pd	10	11	8.13	75	95	170
BTBZNT@SiO ₂ [133]	Ag/Pd	2	5	18.24	175.5	94.5	190
BTAS5 [15]	Ag/Pd	2	19	4.00	65.1	73.6	150
BT-BLT [45]	Pt	13	30	4.05	46.6	95.5	160
BT-BLN [130]	Pt	13	30	4.5	45	91.5	160
BT-BMN [131]	Pt	9	9	6.88	82	90	85
CT-CH [128]	Pt	1	9	9	120	93	—
CT-CH-Mn [128]	Pt	1	9	9.6	130	96	—
ANT-Mn [136]	Pt	—	15	7.9	102	71	100
NN-CZ-BNT [148]	Pt	—	19	3.7	42	82.1	120
BNF-BT [138]	Pt	9	32	6.74	54	77	125
0.4BF-0.6ST [149]	Pd/Ag	—	11	6	42	—	—
0.2BF-0.8ST [149]	Pd/Ag	—	11	2.7	32	—	200
BF-BST-LMN [140]	Pt	1	7	10	73	72	120
BF-BT-BLN [141]	Pt	1	8	13.8	95	81	100
BF-BT-NZZ [139]	Pt	7	16	10.5	70	87	150
BF-ST-Nb-BMN [137]	Pt	1	8	15.8	100	75.2	—
NBT-SBT-BMN [143]	Pt	1	7	18	100	93	160
NBT-0.35SBT [142]	Pt	10	20	21.5	100	80	150
NBT-0.45SBT [135]	Pt	10	20	9.5	72	92	120

Note: *N*—number of active dielectric layers; *t*—thickness of dielectric layer.

of MLCCs are also of concern because the ESR dissipates energy and the ESL may cause detrimental resonance, and these should be as small as possible. The ESR of MLCCs primarily originates from the loss of the dielectrics and the resistance of the inner electrodes and the outer terminals, while the ESL is closely related to the inner electrodes and the outer terminals. Because the ESR and ESL are in series form, an increased number of active layers and a smaller case size would lead to lower ESR and ESL, and hence, multi-layer and miniaturized MLCCs should be promoted. Additionally, the ESR and ESL are functions of the electric field, frequency, and temperature, and these conditions should be carefully considered for high-performance and high-reliability applications.

Pulsed power refers to devices that can provide high peak power by accumulating energy over a period of time and releasing it very quickly. Energy-storage MLCCs, which store electric charge at the electrodes, have an intrinsic ability to generate high power. MLCCs used in pulsed power sources can be roughly simplified according to the schematic shown in Fig. 35(a). The MLCCs can be charged under voltage-constant or current-constant modes with the switch open. Without controlling the discharge, MLCCs deliver energy to the load R via the inductor L +ESL. The discharge waveform can be underdamped, damped, or overdamped depending on the values of C , ESR, ESL, L , and R . For very high-power applications under near short discharge, the L is minimized under a very tiny load R , so the ESR causes energy dissipation and the ESL increases the impedance, which have the effect of limiting the peak power to the load. The ESR and ESL are electric field and frequency dependent, and the RCL charging–discharging circuit currently applied in the research community can somewhat mimic the behavior of the MLCC under real pulse conditions. The energy-storage MLCCs in pulsed power applications are widely used in missile detonators (military service), defibrillators (medical application), and camera flashers (consumer use). The Knowles Capacitors company developed a type of high-energy-density MLCC–Novacap [151] for pulsed power based on lead lanthanum zirconate titanate (PLZT) antiferroelectric materials (Fig. 35(b)). However, the lead-free high-energy-density MLCCs need to be pursued to alleviate the healthy and environmental issues associated with lead-containing materials.

Power converters deal with the voltage and current

Fig. 35 (a) Simplified schematic of a pulsed power circuit using MLCC as the energy-storage element. (b) High-energy-density Novacap MLCCs for pulsed power based on PLZT antiferroelectric materials developed by the Knowles Capacitors company. Reproduced with permission from Ref. [151], © Knowles Precision Devices 2018.

conversions, such as alternating current to direct current (AC–DC), direct current to direct current (DC–DC), and direct current to alternating current (DC–AC) by means of power diodes and transistors. Capacitors in power converters can be found in electric vehicles, 5G base stations, clean energy generation, and smart grids; here, the most distinctive role played by the capacitor is the DC-Link, as shown in Fig. 36(a). A typical demonstration of DC-Link capacitors in three-phase motor inverters is shown in Fig. 36(b). Crucially, the DC-Link capacitors absorb the ripple current, maintain the power imbalance, and restrict over-voltage between the DC battery and the AC motor [152,153], which ensures smooth operation of the motor, decreases energy loss, and extends the life of the battery and inverter [154]. In practical designs, the minimum capacitance is generally determined by the maximum ripple current that the capacitors can handle rather than simply the circuit topology, and this value is related to heat generation stemming from the ESR and thermal dissipation of the capacitors. Therefore, a low ESR and thermal resistance are often required. A large ESL for the capacitors leads to over voltage stress on the semiconductors under a high di/dt on- and off-switching process, which should be minimized via inductance cancellation design. It should be pointed out that the

DC-Link capacitors work at a large DC-biased voltage with small to medium AC superpositions (inset in Fig. 36(b)), and the capacitive response and energy-storage behavior around the DC-biased working point is more important than the overall energy-storage behavior evaluated based on the area enclosed by the P – E loops. Energy-storage performance evaluation should be extended to a large DC-bias with AC superposition fields for frequencies up to the order of megahertz; here, the capacitive response and the energy-storage behavior should be examined at points with higher fields with higher frequencies. This differs from the current measurements reported in literature for both the zero-to-maximum field reversals in P – E loops at low frequencies (typically ≤ 100 Hz) and the RCL charging–discharging method.

As shown in Fig. 37(a), the DC-Link capacitor occupies almost 50% of the volume of the traction inverter in the 1st Gen Toyota Prius [155] and Tesla Model 3 [156] electric vehicles. The capacitors have conventionally been electrolytic and film based owing to the huge capacitance needed for semiconductors operating at lower frequencies (16,500 μF for an electrolytic capacitor or 420 μF for a film capacitor is required for

a 80 kW motor inverter operating at 20 kHz [157]), where the utilization of MLCCs would appear to be technologically and economically inappropriate. The state-of-the-art DC-Link capacitors fall far short of the growing demands for compact, lightweight, and energy-efficient capacitors for power conversion applications, in which high-capacitance-density, high-temperature, and high-ripple-current solutions are desirable. This is particularly true for high-temperature fast-switching wide-bandgap third-generation semiconductors. With increasing switching frequency, the total capacitance needed rapidly decreases without sacrificing performance, as illustrated in Fig. 37(b). Here, MLCCs are in a desirable position because of their extremely low ESR and ESL, high-capacitance-density, high-temperature, and high-ripple-current characteristics compared with their electrolytic and film counterparts [158].

Based on a partial view of the latest industrial offerings, two leading companies have recently announced their MLCC solutions for the third-generation semiconductors, i.e., the KC-LINK series by KEMET [159] and the Ceralink series by TDK [160], as shown in Figs. 38(a) and 38(b). The KC-LINK MLCCs are C0G type with lead-free CaZrO_3 -based linear dielectric materials that

Fig. 36 (a) Role of DC-Link capacitor in the AC–DC, DC–DC, and DC–AC converters. (b) DC-Link capacitor in a three-phase motor inverter. The insets show the voltage and current waveforms of the capacitor and motor, respectively.

Fig. 37 (a) DC-Link capacitors in the traction inverters of the 1st Gen Toyota Prius and Tesla Model 3 [155,156]. (b) Trends and future requirements for DC-Link capacitors [158].

Fig. 38 (a) KEMET KC-LINK series DC-Link MLCCs [159]; (b) TDK Ceralink series DC-Link MLCCs [160]; (c) a high-power-density 200 kW DC–DC converter by Fraunhofer based on full DC-Link MLCCs and third-generation SiC MOSFETs [161]; and (d) a compact cooling pump power converter by AB Mikroelektronik GmbH for electric vehicles based on Ceralink MLCCs [162].

can function up to 200 °C, while the Ceralink MLCCs are based on PLZT antiferroelectrics that can work properly

under temperatures briefly up to 150 °C. Compared with the KC-LINK, the Ceralink has a higher energy density.

Nevertheless, the KC-LINK is lead-free and more temperature stable. Both the MLCCs are manufactured using cost-effective base-metal electrodes, i.e., nickel for the KC-LINK and copper for the Ceralink. Industrialization of lead-free MLCCs with a high energy density and good temperature stability is still ongoing. For the representative utilization of MLCCs for DC-Link applications, Fraunhofer demonstrated a high-power-density 200 kW DC–DC converter based on full DC-Link MLCCs and third-generation SiC MOSFETs (Fig. 38(c)) [161], which to the best of our knowledge is one of the most high-power-density DC–DC converters. AB Mikroelektronik GmbH developed a compact cooling pump power converter for electric vehicles based on the Ceralink MLCCs to replace the former electrolytic ones (Fig. 38(d)) [162].

6 Challenges and future prospects

Lead-free high-energy-density MLCCs that meet the practical requirements of current applications are of urgent demand to resolve the lead-free but low-energy-density problems or the high-energy-density but lead-containing issues. Despite the breakthroughs achieved over the past decade, lab-scale lead-free high-energy-density MLCCs still face challenges, which include:

(1) Scale-up capability. It has to be determined whether the costly gradients and fine-tuning method used for small-scale energy-storage MLCCs can be scaled up for industrialization, and fundamental studies need to be performed to determine how the material and device fabrication conditions can be effectively controlled when scaled up.

(2) Compatibility with the base metal electrode. State-of-the-art lead-free high-energy-density MLCCs utilize precious metals as internal electrode, which need to be replaced by base metals to significantly lower the cost. The compatibility of the base metal electrode requires the dielectric materials to be anti-reducible or low-fired. In addition, other issues related to maintaining a high energy-storage performance under extreme conditions, such as high electric field, high frequency, and high temperature will need to be addressed.

(3) High reliability. High reliability determines long-term service capability towards high-quality industrial applications. The performance degradation and failure under the influence of electric–thermal–mechanical–humid multiphysics should be unraveled. This requires

effective theories, models, methods, experimental platforms, and simulation techniques of accelerated degradation.

(4) Application-oriented measurement and evaluation. Present measurement and evaluation of the energy-storage performance deviate from real-world conditions for the specific applications previously mentioned. Hence, the developed materials and devices cannot be directly applied to practical applications. Proper standards should be set up based on the cooperation between researchers and manufacturers.

Despite these obstacles and challenges, industrialized lead-free high-energy-density MLCCs are anticipated in the future based on intensive research and development. With industrialization, a new era of pulsed power and power converter solutions may arrive, which can enable a more energy-efficient and eco-friendly world.

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