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Recent advances in energy storage performance of barium titanate-based relaxor ferroelectric ceramics

Meng Wang*

School of Undergraduate Education, Shenzhen Polytechnic University, Shenzhen, 518055, China

Abstract: *Dielectric energy storage materials have drawn universal research attention for their potential application in a great number of electrical and electronic devices. They are becoming increasingly viewed as core components for the next-generation AI and IT circuits. This study briefly reviews the four dominant families of energy storage ceramic materials, and their respective underlying mechanisms and current research development. At the same time, extensive research has been conducted on the energy storage performance of relaxor ferroelectric (RFE) ceramics derived from barium titanate ($BaTiO_3$, BT). Given the limited energy storage capability of pure BT, four primary enhancement strategies have been explored: Doping modification, re-shell structure, core-shell structure, and grain engineering are comprehensively detailed based on their application for enhanced dielectric energy storage performance. By integrating artificial intelligence techniques, BT-based RFE ceramic development has vast potential for high-speed development.*

Keywords: *Barium titanate; energy storage; application*

1. Introduction

Dielectric energy storage materials are featured with high power density, ultra-fast charge/discharge rates, ultra-long service times, and high stability in general and are key components in electronic devices and pulsed power systems. As environmental friendliness and device miniaturization are becoming increasingly important on the agenda, enhancing the energy storage efficiency of dielectric energy storage materials has become a matter of great urgency. Lead-free barium titanate ($BaTiO_3$, BT) has a high dielectric coefficient, ultra-low dielectric loss, and ultra-high thermal and mechanical stability, and hence represents a most promising dielectric energy storage material. Despite these ideal properties, intrinsic defects in simple BT ceramies make them unfavorable for direct application for energy storage. Against such a backdrop, researchers have thus given close attention to strategies such as elemental doping, third-element incorporation, and architectural design to enhance the performance level of relaxor ferroelectric (RFE) ceramies on the basis of BT. As a result, the present paper provides an overview on classification for dielectric ceramic energy storage materials, critiques the latest findings on systems based on BT, and addresses advanced development within the field.

2. Overview of dielectric ceramic types used in energy storage

The energy storage materials in the form of ceramics are thus covered under two broad categories on the basis of the characteristic storage: linear dielectrics (LD) and nonlinear dielectric materials. The nonlinear one is further divided into ferroelectric (FE), relaxor ferroelectric (RFE), and antiferroelectric (AFE) material categories. All the above mentioned four categories possess characteristic and distinct energy storage properties and are thus usable for different application purposes.

2.1 Linear dielectrics (LD)

LD are those for which polarization is proportional to the applied electric field. There is no spontaneous polarization and domain structure for such materials. Thus, their polarization-electric field (P-E) response is a straight line without a hysteresis loop and provides high efficiency for energy storage. Unfortunately, it is normally difficult for a material to be both high in dielectric constant and high in breakdown strength [1]. Though LD are likely to possess high breakdown fields and low dielectric losses, low dielectric constants for these dielectrics render their polarization strength low and thus reduce their energy storage density. Al_2O_3, TiO_2, $CaTiO_3$, and $SrTiO_3$ are typical LD materials that

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are commonly encountered. Among them, calcium titanate ($CaTiO_3$) is a typical inorganic dielectric ceramic material that possesses excellent dielectric, thermal, mechanical, and optical properties and has been widely adopted for use in fields such as those for ceramic capacitors, positive temperature coefficient (PTC) thermistors, microwave devices, and filters. Nevertheless, $CaTiO_3$ has a quite small dielectric constant, usually below 300, and a limited polarization strength at 2.6 μC/cm^2, literally limiting its energy storage density and rendering its application requirements difficult to be satisfied. Recently, a number of studies have advanced the energy storage density of LD by virtue of doping modification..Polarization strength for $CaTiO_3$ was strongly augmented by the incorporation of $BiScO_3$ and could be up to 12 μC/cm^2 under high electric field stress. The modification was also witnessed to lead to an energy storage density of 1.56 J/cm³ with a storage efficiency of 90%. As part of another study, a storage density and breakdown strength were documented as high as 1.2 J/cm^3 and 280 kV/cm, respectively, for $SrTiO_3$ with a Sr/Ti ratio of 1. The grain size for $SrTiO_3$ was also optimized upon incorporation of SiO_2, and consequently, the material was able to exhibit a storage density as high as 1.2 J/cm^3 when a high electric field was supplied. All these developments notwithstanding, while performance has been boosted for LD, the cumulative energy storage for these dielectrics is yet low for mass production for applicability in a wide range of commercial applications, and hence augmentation is yet necessary [2].

2.2 Ferroelectric (FE)

In certain crystalline materials, spontaneous polarization can occur internally, and this polarization can be reversed when subjected to an external electric field—a phenomenon known as ferroelectricity. Materials exhibiting this behavior are termed FE crystals. Within these materials, regions that share the same polarization direction are referred to as FE domains, analogous to magnetic domains in ferromagnetic materials. In the absence of an external field, these domains are randomly oriented, resulting in a disordered domain structure. When an external electric field is applied, the domains begin to reorient and grow, gradually aligning in the direction of the field. However, this reorientation process lags behind the electric field, creating hysteresis loops in the P-E response. Because of the domain structures, FE materials typically exhibit high polarization strengths. Yet, after the external field is removed, some domains do not fully return to their original states, leaving behind a large remnant polarization, which significantly reduces energy storage efficiency. As a result, FE materials tend to suffer from high energy loss and are generally considered less ideal for energy storage applications. Additionally, these materials undergo a phase transition from the FE to the paraelectric state at a characteristic temperature known as the Curie temperature (T_c).

Currently, research in the field of dielectric ceramics is primarily focused on several lead-free systems, including BT, bismuth ferrite ($BiFeO_3$, BF), sodium bismuth titanate ($Na_{0.5}Bi_{0.5}TiO_3$, NBT), and potassium sodium niobate ($K_{0.5}Na_{0.5}NbO_3$, KNN). BT-based ceramics are integral components of lead-free FE ceramic systems and serve as the matrix material in this study. Their physical properties and recent research developments will be discussed in detail in subsequent sections. BF-derived ceramics exhibit high polarization strength, and some studies have reported strengths near 100 μC/cm^2. This is mainly ascribed to Bi^{3+} ions and their individual electron pairs responsible for greater ferroelectricity for the material. Consequently, BF-derived ceramics have gained a lot of research focus. The development of purely BF-derived ceramics is, however, very tricky, primarily at the stage of sintering, where volatilization of bismuth and valency changes for the iron ions may be induced. The resultant problems above often lead to a high dielectric loss and leakage current, hence limiting their application. Due to these drawbacks, newer research has pointed out solid solution methods and particularly by combining BF with other perovskite-phase materials. Among those systems, one that has been considerably researched is (1-x) BF-xBT. Separately, for x=0.33, the quasi-homogeneous phase boundary for the BF-BT system exhibits higher dielectric constant and polarization responses. The energy storage capability for BF-BT composites, nonetheless, is still short of being commercially feasible. The current emphasis is on boosting the energy storage ability of BF-BT FEs by the incorporation of dopants to produce relaxor character and thereby convert them into high-performance RFEs. At the same time, KNN-related compounds, a solid solution between FE potassium niobate ($KNbO_3$) and AFE sodium niobate ($NaNbO_3$), exhibit remarkable piezoelectric performance. The greater majority of KNN system research, however, has been directed at enhancing their piezoelectric performance and has not placed a great amount of emphasis on their energy storage performance. Conversely, NBT has also been endowed with a promising outlook for dielectric energy storage use due to its strong ferroelectricity. The characteristic for purely perovskite-structured NBT is that the A-site is composed of two valently distinct ions, leading to a compex and partly controversial crystal structure, particularly at room temperature. Pure NBT ceramics are, nonetheless, limited by low breakdown

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strength, high coercivity field, and high remnant polarization, which are unfavorable for energy storage performance. All these shortcomings can be offset by forming solid solutions with other perovskite-phase ceramics for better dielectric and energy storage performance[3].

2.3 Relaxor ferroelectric (RFE)

RFE materials have attracted great attention in the research community for dielectric energy storage in recent years. RFEs are a distinct subclass of FEs and are distinguished by the intrinsic features within FE materials (i.e., spontaneous polarization and domain structure). The RFEs are differentiated from conventional FEs by certain intrinsic properties. The RFEs exhibit extremely low residual polarization and low coercive electric field. The P-E hysteresis loops are therefore significantly narrow for RFEs. The innovation of high-capacity dielectric energy storage devices greatly relies on this feature. Therefore, RFEs are considered to be promising materials for dielectric energy storage devices. The study on RFEs has acquired great vigor in recent years, and hundreds of reports and research have been reported. Presently, the phenomenon of relaxation in RFEs primarily expresses in the following fields:

(1) Diffuse phase transition phenomenon

Diffuse phase transition is a gradual transition from the paraelectric to the FE state for a dielectric material across a broad temperature range, as compared to a sharp, well-defined transition character in polarization. Unlike normal FEs, RFEs are devoid of a sharp, definitive Tc. The dielectric temperature response is significantly broadened in their place, and the response is characteristic in the absence of sharp dielectric peaks that are typified in normal FE material.

(2) Frequency dispersion phenomenon

In conventional FE materials, the dielectric constant remains unaffected by frequency, resulting in dielectric temperature spectra at different frequencies that approximately overlap. With the increase in external field frequency, the maximum dielectric constant and dielectric loss for RFEs shift towards higher temperature [4]. The maximum dielectric constant also reduces at higher frequency, a situation that is normally followed by a slight increase in dielectric loss.

(3) Composite modified Curie-Weiss law

The diffused nature of the phase transitions in RFEs makes the classical Curie-Weiss law invalid for the temperature and dielectric constant relation. Instead, for temperatures above the temperature at the maximum dielectric constant (Tm), the temperature and dielectric constant relation is expressed by a modified Curie-Weiss law as follows [5]:

$$\frac{1}{\varepsilon}-\frac{1}{\varepsilon_m}=\frac{(T-T_m)^\gamma}{C}$$

where ε_m is the peak dielectric constant; T_m is temperature at the maximum dielectric constant; and γ is the relaxation degree of RFE materials.

γ is a critical parameter used to quantify the extent of relaxation behavior in FE materials, typically ranging from 1 to 2. For conventional FEs, γ equals 1, while for ideal RFEs, γ reaches 2.

REFs are quite well known for their high energy storage performance, and many high-performance compositions have actually been synthesized. As a prototype, a high-entropy RFE thin film with chemical composition $(Bi_{0.7}Na_{0.67}Li_{0.03})_{0.5}Sr_{0.3}TiO_3$, augmented by manganese doping against the impact of oxygen vacancies, exhibited great energy storage density up to 47 J/cm^3 and storage efficiency up to 69%, and ideal thermal, frequency, and fatigue stability. Hussain et al. [6] synthesized a set of RFEs through a high-entropy approach by A-site doping. Among them, the $(Ba_{0.2}Na_{0.2}Ca_{0.2}Sm_{0.2}Bi_{0.2})TiO_3$ showed 1.0 J/cm^3 energy storage density and 81% energy storage efficiency. $(Bi_{1/2}K1_{/2})_{0.5}Sr_{0.5}TiO_3$ was fabricated by a joint solid-state and hydrothermal approach with fine grain size and 2.25 J/cm^3 energy storage density at 240 kV/cm electric field. By adopting a solid-state method, Chen [7] prepared RFE material $(1-x)(0.99Na_{0.5}Bi_{0.5}TiO_3\text{-}0.01BiYbO_3)\text{-}xSrTiO_3$, achieving an energy storage density of 2.5 J/cm^3 and an energy storage efficiency of 81%. Additionally, by doping the rare-earth element neodymium into $Sr_{0.7}B_{i0.15}Nd_{0.05}TiO_3$, the energy storage density was further improved to 2.34 J/cm^3, with an enhanced storage efficiency of 92%. Wen et al. [8] introduced 0.15 wt.% $MnCO_3$ into the ternary solid solution $(0.67-x)BiFeO_3\text{-}xBi_{0.1}Na_{0.7}NbO_3\text{-}0.3BaTiO_3$, resulting in an enhanced energy storage performance with an energy density of 3.06 J/cm^3 and an energy storage efficiency of 81%.

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2.4 Antiferroelectric(AFE)

The dipoles are aligned in parallel and opposing directions in AFE compounds. There is no net spontaneous polarization and no hysteresis loop for these compounds. The AFE phase transforms into the FE phase with an external stimulus, for instance, an electric field. The development of a double hysteresis loop is an outcome that describes the transition. From the perspective of energy storage, the AFE materials are considered promising materials due to their high value for maximum polarization and remnant polarization, which facilitate effective charge-discharge cycles. The practicality of the AFEs for use, however, is always spoiled by low breakdown strength, being a direct effect of high internal stress and transient current that arises while experiencing the electric field-driven phase transition on the AFE-FE state. Moreover, the characteristic double hysteresis curves observed in the AFE materials lead to relatively low energy storage efficiency. Among the reported AFEs, the $AgNbO_3$ (AN) and $NaNbO_3$ (NN) have been the most studied. The present research is primarily concerned with improving the breakdown strength and resultant energy storage performance of the AFEs by virtue of doping modifications and structural designing. One approach that has been quite remarkable includes the development of relaxor-type AFEs that are designed to soften the AFE-FE phase transition, thereby enhancing both the energy density and efficiency. For example, an energy storage density value of 11 J/cm^3 and 91% efficiency was reported for a $0.9NaNbO_3$–$0.1Bi(Ni_{2/3}Nb_{1/3})O_3$ composition due to the tailoring of the microstructure for local polarization enhancement. A lead-free relaxor AFE material with chemical formula $0.94(Na_{0.88}Sm_{0.04}NbO_3)$–$0.06(BiFeO_3)$ exhibited an energy storage density value of 4.00 J/cm³ and an energy storage efficiency value of 80% [9].

3. Current Research status of BT based dielectric ceramics

BT is a widely studied FE material featuring a typical ABO_3 perovskite crystal structure. Since its introduction in the 1940s, BT-based ceramics have attracted significant attention due to their high dielectric constant and low dielectric loss, and have become the dominant class of lead-free FE materials, particularly favored for environmental reasons. At room temperature, BT adopts a tetragonal structure, where the A-site is occupied by Ba^{2+} ions located at the corners of the unit cell, each with a coordination number of 12 and an ionic radius of 1.61 Å. The B-site is occupied by Ti^{4+} ions, positioned at the center of the unit cell with a coordination number of 6 and an ionic radius of 0.605 Å. These Ti^{4+} ions form a TiO_6 octahedral configuration with surrounding oxygen atoms within the tetragonal lattice.

There are different forms of crystals for BT in relation to temperature. It has a cubic structure at temperatures lower than -90 °C. As temperature increases, the compound goes through an orthorhombic phase close to 5 °C and assumes a tetragonal structure at room conditions. Ti^{4+} ions are likely to shift along the c-axis without an external field, and thus crystal symmetry is lost and spontaneous polarization results, giving sharp ferroelectricity at room temperature. As temperature increases above T_c, a transition occurs from a tetragonal phase to a cubic phase and Ti^{4+} ions reoccupy the central symmetric lattice site and depress the spontaneous polarization and render the material paraelectric. When the temperature exceeds 146 °C, BT assumes a hexagonal shape.

Despite a high dielectric constant and high polarization, the dielectric response of BT is heavily influenced by structural phase transitions. The near-Tc for BT has a pronounced maximum in dielectric constant due to drastic symmetry changes. As a result of its intrinsic shortcomings—such as large domain structures, sluggish switching kinetics of the domain, and broad hysteresis loop with high remanent polarization—it is limited for high-efficiency energy storage. Therefore, undoped BT ceramic is inferior for direct use in advanced dielectric energy storage.

Currently, strategies to enhance the energy storage properties of BT ceramics primarily concentrate on developing BT-based RFEs to improve their relaxation behavior. This approach aims to modulate the phase transition temperature and polarization characteristics of BT. The principal methods employed at present include:

(1) Doping modification

Currently, doping modification represents one of the most widely adopted strategies for tuning the energy storage performance of BT-based ceramic materials. The energy storage capability of the ABO_3 compounds may significantly be enlarged by means of doping substitutions at the A- or at the B-sites with ions with same or disparate valence states. The most recent findings have substantiated that dopant substitutions in BT-derived ceramics effect relaxation conversions and thus improve their energy

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storage efficiency. The latest studies suggested that ionic substitution is an effective tool in terms of tuning the material properties of BT ceramics. For one, substitutions with Sr^{2+} at the A-site lowered Tc, while those with Pb^{2+} could raise the latter. Substitutions at the same site with Ca^{2+} lead to higher stability for the tetragonal phase and thus higher thermal stability. Substitution at the same site with Co^{2+} has also been discovered to lower dielectric loss significantly, particularly at higher temperatures. In the recent years, spectacular development in the enhancement in energy storage abilities of $BaTiO_3$-based RFE materials was presented by utilizing the simple doping technique. For example, a work presented a remarakable energy storage density of 3.13 J/cm^3 and efficiency of 91.71% by constructing $(Ba_{0.98-x}Li_{0.02}Bi_x)(Mg_{0\cdot04}Ti_{0.96})O_3$ RFEs. Ren et al. [10] prepared $(1-x)(0.88BaTiO_3–0.12Bi(Li_{0.5}Nb_{0.5})O_3)$-$x(0.8BaTiO_3–0.2SrTiO_3)$ series ceramics, achieving an energy storage density of 2.51 J/cm^3 and an energy storage efficiency of 86.81% under an electric field of 320 kV/cm. The composition $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$-$x$BiFeO$_3$ was developed by co-doping BT with calcium and zirconium ions, along with the incorporation of BF. This system achieved an energy storage density of 0.57 J/cm^3 and a high energy efficiency of 91%. Ma et al. [11] enhanced the energy storage performance of BT-based ceramics by incorporating bismuth-based compounds. Specifically, the composition $0.85BaTiO_3$-$0.15Bi(Zn_{0.5}Zr_{0.5})O_3$ achieved an energy storage density of 3.58 J/cm^3 and an energy storage efficiency of 90%.

(2) Core-shell structure

Structure-property relationship is a core feature in dictating the functional performance of material systems, particularly in energy storage ceramics. Parameters such as microstructure, interface structure, defect distribution, and phase composition all dictate the dielectric and energy storage performance in tandem. There are a vast number of interfacial structures that are inherent within the same systems of ceramics, particularly at the grain and grain boundary interfaces. The structural inhomogeneity between the two regions significantly affects both polarization performance as well as breakdown strength due to the tendency of grain boundary interfaces to be barriers/channels that suppress or facilitate domain motion and charge build-up. The phase interface between individual phase structures has a profound effect on the movement and reorientation of internal domain structures in ceramics. The addition of a core-shell structure has a high regulating effect on the interface and thus increases energy storage performance. The traditional solid-state sintering technique is a common technique for fabricating ceramics. The technique involves a stoichiometric mixing of starting materials followed by ball mill and pre-sintering for the formation of the required crystal phase and finally sintering at a suitable temperature for the formation of desired ceramic material. Although conventional solid-state sintering allows for multi-elemental doping and multiphase composites, controlling interfaces in the fields of ceramic material is a limiting factor. To compensate for the shortcoming, preparation of chemical-coated ceramic powders has been undertaken by researchers for the construction of core-shell architectural ceramics. Chemical coating not only offers a means for regulating interfaces, and moreover, reduces the effect of impurities on the properties of ceramic material. For example, Chen et al. prepared a core-shell structure by co-doping $(1-x)BiFeO_3$-$xBaTiO_3$ with neodymium and varying the amount of BT, and a storage energy density between 2.5 and 2.7 J/cm^3 was achieved for the compositional range x = 0.2 to 0.45. The sol-gel technique was used by Xu et al. [12] for synthesizing the series (BNTBT-KN), and a storage energy density was achieved that was 1.73 J/cm^3. Other research has adopted the use of the sol-gel technique for preparation for the production of La^{3+}-doped BT-based ceramics, allowing for structural transformations in the phases.

(3) Domain Engineering

The domain configuration within FE materials plays a crucial role in determining their energy storage behavior. Optimizing the domain structure in FE materials can effectively enhance relaxation behavior, thereby improving their energy storage properties. Additionally, the formation of multiple phase structures in ceramic materials can be facilitated through solid solution formation between ceramics or by doping with other elements. The integration of multiphase structures in FE ceramics has the capability to disrupt the long-range domain ordering and lead to short-range ordered polar nano-regions. The localized polar regions are inherently fast responding to external electric fields and thereby strongly diminish residual polarization and constrict hysteresis loops, ultimately providing high energy storage density and efficiency. Currently, researchers have developed numerous RFEs exhibiting superior performance by designing domain structures. Wang et al. [13] controlled the relaxation behavior of $Na_{0.5}Bi_{0.5}TiO_3$ through domain engineering, achieving an ultra-high polarization intensity of 52.4 μC/cm^2 at relatively low electric fields.

(4) Grain design

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The design of grain size and morphology in ceramic materials can enhance their energy storage capabilities. Recent studies demonstrated that optimizing grain size can increase the dielectric constant of ceramic materials, while simultaneously decreasing Tc and coercive electric field. Reducing grain size for ceramic materials leads to a higher grain boundary density that is able to impede charge carrier movement efficiently, serving to lower leakage current and raise breakdown field strength. Therefore, a tried and tested means to improve the energy storage capacity for ceramics is to develop them with finer grained structures. For the test material sample BT ceramics, grain size has a direct impact on the domain structure. Above the micrometer level for grain size, a reduction in grain size leads to a reduction in 90° domain number. The reduction increases interfacial activity between domains for an electric field. Considering that the dielectric constant for BT is greatly determined by motion for its internal 90° domains, any modification in grain size accordingly affects its dielectric response. However, while small grained structures are apt to generate better performance, fine grained structures are apt to destabilize domain structures, indicating an optimum range for grained size and suggesting that a mere "smaller is better" condition does not exist. At excessively small grain sizes, the domain structure may become unstable as a result of gradient compensation of grain boundary charges and polarization. Currently, grain size control can be accomplished through doping modifications, the development of multi-component solid solutions, the incorporation of sintering aids, and the optimization of sintering processes. Many studies investigated BT-based ceramics with varying grain sizes and reported that samples with an average grain size of approximately 70 nm exhibited an energy storage density of 0.83 J/cm^3 under an applied electric field of 170 kV/cm.

(5) Other cutting edge research

The latest development on BT-based ceramics has been to develop high-performance dielectrics by compositional and structural tailoring. Wang et al. [14] prepared the (1–x)(0.67$BiFeO_3$–0.33$BaTiO_3$)–x($Sr_{0.7}Nd_{0.2}$)TiO_3 by conventional methods. The material showed recoverable energy storage density (W_{rec}) value of 2.57 J/cm^3 and high storage efficiency of 79% at low electric fields and showed noteworthy frequency stability and charge-discharge functionality. Similarly, the (1–x)(0.67$BiFeO_3$–0.3$BaTiO_3$)–x($Sr_{0.7}Bi_{0.2}$)($Mg_{1/3}Ta_{2/3}$)O_3 composition that decomposes to a quasi-cubic perovskite phase at room temperature showed recoverable energy density of 2.52 J/cm^3 and storage efficiency of 77.3% at low fields. This system also exhibited outstanding electrical responsiveness across a range of frequencies and robust charge-discharge characteristics, indicating its strong potential for future cutting-edge energy storage applications.

4. Conclusion

The preceding discussion demonstrates that BT-based RFE ceramics exhibit outstanding energy storage performance. Despite core-shell structuring, doping modification, and grain tailoring having considerably enhanced energy storage performance for dielectric ceramics, progress in development is to be guided by theory-oriented researches on both the domain level and the microstructure. Integrating artificial intelligence is another promising avenue for enhancing design and optimization for the ensuing-generation energy storage materials.

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