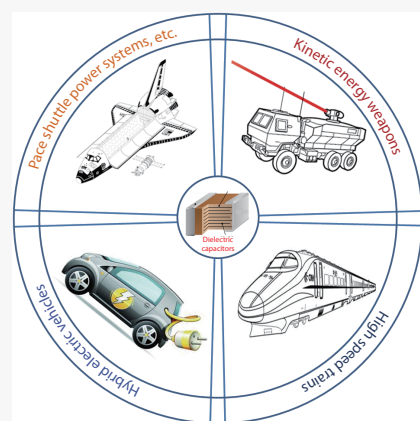


Antiferroelectric capacitor for energy storage: a review from the development and perspective

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With the fast development of the power electronics, dielectric materials with large power densities, low loss, good temperature stability and fast charge and discharge rates are eagerly desired for the potential application in advanced pulsed power-storage system. Especially, antiferroelectric (AFE) capacitors which have been considered as a great potential for electric device applications with high energy density and output power are widely concentrated recently. To propel the development of dielectric capacitors marketization, in this view, we comprehensively summarized the development process of energy storage density and efficiency, improving strategy, raw materials cost and thermal stability of the typical AFE capacitors, including $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$, AgNbO_3 , $(\text{Bi}, \text{Na})\text{TiO}_3$, and NaNbO_3 AFE systems. Moreover, the advantages and disadvantages of these AFE energy-storage ceramics are compared and discussed, which lay the foundation for the AFE energy storage capacitor early realization of marketization.



With the rapid development of electronics industry, the demand for dielectric energy storage devices is becoming more and more urgent, such as hybrid electric vehicles, laser weapons, space vehicle power systems, and cardiac defibrillators as shown in Fig. 1^[1-3], because they have fast charging/discharging speed, high power density and excellent fatigue resistance^[4]. Various types of dielectric materials can be potential candidates for energy storage, including antiferroelectrics (AFEs)^[5-7], relaxor ferroelectrics (RFEs)^[8,9], normal ferroelectrics (FEs)^[10], and linear nonpolar dielectric materials^[11]. Among these dielectrics, AFE dielectrics, characterized by a double hysteresis loop, are favored for energy storage due to their relatively high maximum polarization (P_{max}) and particularly low remanent polarization (P_r) compared with other types of dielectrics. In this paper, we summarized the energy storage density (U_{rec}) and efficiency (η) of AFE dielectrics systems in recent 10 years as shown in Fig. 2, and reviewed their industrialization application potential from the properties, economics, and environment.

Lead-based antiferroelectrics

Lead-based AFE ceramics that possess excellent U_{rec} and η , like $(\text{Pb}, \text{La})(\text{Zr}, \text{Ti})\text{O}_3$ system^[5,6,12-14], have been the mainstay

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energy storage materials. Even so, the U_{rec} of lead-based AFE ceramics is just about 0.55 J cm^{-3} in the early days, and increases to 13.9 J cm^{-3} at present, along with $\eta > 90\%$. From the whole development process as shown in Fig. 2a^[15-26], the improvement of energy storage performances can be summarized in the following three aspects. Firstly, increasing the electric field of AFE-FE phase transition, such as phase transition electric field of AFE-to-FE (E_F) and FE-to-AFE (E_A)^[27]; Secondly, designing a core shell^[28] or laminated composite structure^[26] to improve the breakdown strength (BDS); Thirdly, building a multiple phase transition^[22,29,30]. All these measures not only improve the U_{rec} of lead-based AFE ceramics, but also make their η significantly increased. Except for above bulk ceramics, multilayer capacitors (MLCCs) technique also is a great way to achieve high U_{rec} and η . For example, Hao et al.^[31,32] achieved a variety of ultrahigh $U_{\text{rec}} > 15 \text{ J cm}^{-3}$ and excellent $\eta > 90\%$ in lead-based MLCCs. However, with increasing environmental concerns, the adverse effect of lead oxide on the environment and human health will gain more and more attention, and the development of lead-free AFE energy storage capacitors have necessitated.

$\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ based antiferroelectrics

Lead-free antiferroelectric materials have attracted increasing attention for environmentally friendly energy-storage applications in recent years^[33]. As one of the earliest studied lead-free energy storage ceramics $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ (BNT)-based AFE have a series of attractive features, including relatively large and tunable spontaneous polarization and high Curie

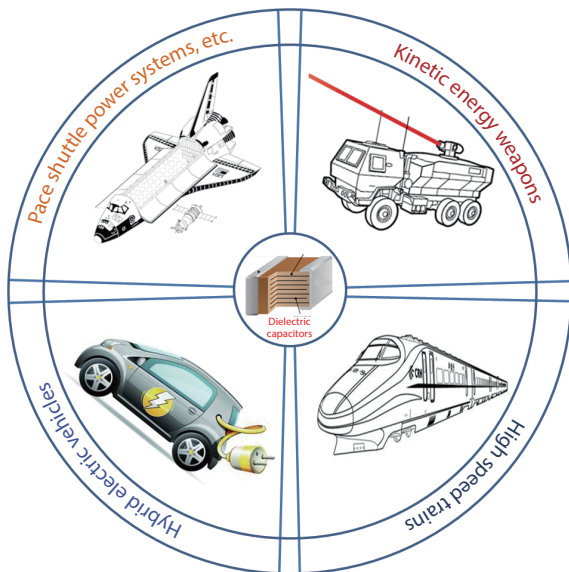


Fig. 1 The application of dielectric energy storage materials in pulsed-discharge and power conditioning electronic devices.

temperature, which enables BNT to maintain large polarization over a wide temperature range. Yet, the U_{rec} of pure BNT ceramic is limited because of their high P_r and relatively low BDS. Therefore, from 2011 to 2020, researchers have been working on reducing the P_r and increasing the BDS by increasing relaxor behavior and decreasing the grain size [34–37]. Especially the strategies, such as defect engineering, [38] field-induced structure transition [39], multiscale polymorphic domains [41], were adopted in recent years, making U_{rec} and η of

BNT system markedly increased as shown in Fig. 2b [35,38–44]. Besides, the feature of thin dielectric layer thickness also makes BNT based MLCCs possess an ultrahigh BDS and U_{rec} , especially for the <111>-textured $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3\text{-Sr}_{0.7}\text{Bi}_{0.2}\text{TiO}_3$ (NBT-SBT) MLCCs, which possess an ultrahigh BDS $\sim 103 \text{ MV m}^{-1}$ and $U_{\text{rec}} \sim 21.5 \text{ J cm}^{-3}$ because it has a greatly low tensile electro-strain [45]. However, it is still not mature to be used to marketization, due to the complicated chemistry structure and high process cost for the texture.

AgNbO₃ based antiferroelectrics

The characteristic of double P-E hysteresis loops for Ag-NbO₃ (AN) was first reported in 2007, suggesting the practical possibility of its application in energy storage [46]. However, the U_{rec} of pure AN ceramic is only about 2 J cm^{-3} [47,48], far lower than that of lead-based AFE systems [45,49,50]. The main reasons are that there have a non-zero P_r value and poor breakdown strength (BDS) at room temperature for pure AN ceramic. Enormous efforts [51–58] have been made as to solve these questions as following three aspects. One is using oxide dopants for compositional modification to suppress the ferroelectricity characteristic and boost the AFE one. Typical examples include AN+0.1wt%MnO₂ [51] and AN+0.1wt%WO₃ [47] systems, where the U_{rec} reaches 2.5 J cm^{-3} and 3.3 J cm^{-3} , respectively. Another is ion substitutions, e.g., replacement of Ag⁺ by La³⁺ [48], Sm³⁺ [59], Ba²⁺ [60], Lu³⁺ [61], Gd³⁺ [62], etc., and/or Nb⁵⁺ by Ta⁵⁺ [52]. The U_{rec} can be effectively increased to 3.2 J cm^{-3} in Ag_{1-3x}La_xNbO₃ system at $x=0.02$ [48], 4.5 J cm^{-3} in Ag_{1-3x}Sm_xNbO₃ system at $x=0.02$ [59], 2.3 J cm^{-3} in Ag_{1-2x}Ba_xNbO₃ system at $x=0.02$ [60], and 4.2 J cm^{-3} in Ag(Nb_{1-x}Ta_x)O₃ system at $x=0.15$ [52], and so on. Reducing

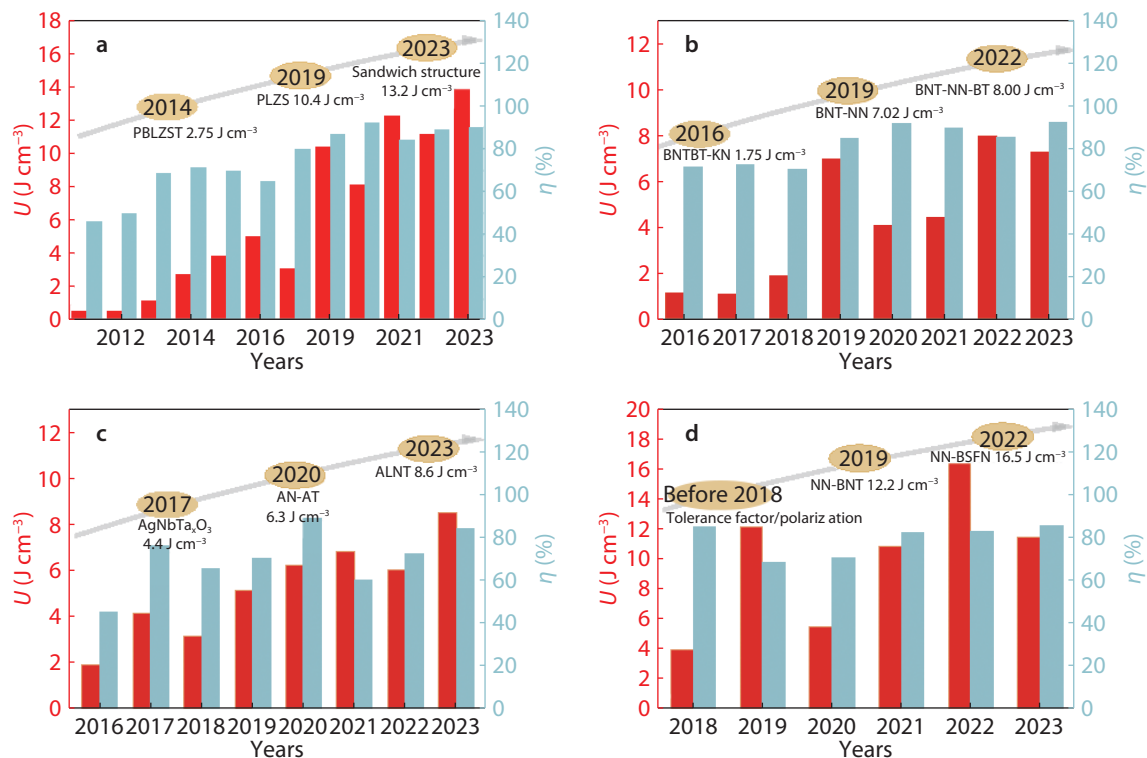


Fig. 2 Comparison of energy-storage properties among four typical anti-ferroelectric ceramics in recent 10 years.

the thickness of the dielectric layer is the other efficacious strategy to enhance the BDS and U_{rec} of the AN system. For example, Zhu et al.^[63] indicates that an ultrahigh $U_{rec} > 14 \text{ J cm}^{-3}$ and excellent $\eta > 85 \%$ were achieved in $\text{Sm}_{0.05}\text{Ag}_{0.85}\text{Nb}_{0.7}\text{Ta}_{0.3}$ MLCCs. All these results suggest that AN AFE materials have a great application potential in pulsed-discharge and power conditioning electronic devices. However, it is difficult to meet the low-cost market requirement because of its high price for raw materials in AN system.

NaNbO₃ based antiferroelectrics

Sodium niobate (NaNbO_3 , NN) system has also been attracted intensive interest because it has similar with AN system's AFE perovskite structure. Different with the AN system, lead-free NN AFE ceramics has a raw material with low cost, simple sintering process, high band gap, and so on, which was considered the most promising dielectric capacitors. However, it is difficult to get double P-E hysteresis loops at room temperature, because its metastable FE phase (P21ma) and AFE phase (Pbma) can coexist at room temperature and the electric field induced FE phase can still be preserved after the electric field is removed, exhibiting a ferroelectric P-E hysteresis loop^[64-71]. An idea which was to stabilize the antiferroelectric phase and exploits its potential for AFE energy storage was achieved in $\text{NaNbO}_3\text{-CaZrO}_3$ ^[72,73], $\text{NaNbO}_3\text{-CaHfO}_3$ ^[74], and $\text{NaNbO}_3\text{-CaSnO}_3$ ^[75] systems, and so on with applying the strategy of reducing the tolerance factor and average B-site polarization. However, their U_{rec} are still very low because of low BDS and large P_r . For example, the U_{rec} and η of AFE

$\text{NaNbO}_3\text{-CaZrO}_3$ system are just about 0.55 J cm^{-3} and 63%^[76]. To further improve the U_{rec} and η , many of the strategies were adopted, such as breaking long-range ferroelectric order which is the benefit to build relaxation characteristics and improve BDS, decreasing the grain size or reducing the dielectric properties to enhance the breakdown field, and soon^[77-87]. The U_{rec} and η of NN-based ceramics have gotten an obviously improvement as shown in Fig. 2d^[76-79,84,88-90]. For example Qi et al.^[77] have reported that an $0.76\text{NaNbO}_3\text{-}0.24(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ relaxor antiferroelectric (AFE) ceramic was designed, which shown an ultrahigh energy-storage density $U_{rec} = 12.2 \text{ J cm}^{-3}$, which is higher that of other lead-free system and most of lead ceramic. Xie et al.^[78] have shown that the fine grain for $0.83\text{NaNbO}_3\text{-}0.17\text{SrTiO}_3$ ceramics prepared by the two-step sintering method is the benefit to improve the U_{rec} and η , which are 1.60 J cm^{-3} and 50%, respectively. Besides, Shi et al.^[48] have reported that a PC-phase $0.78\text{NN}\text{-}0.22\text{Bi}(\text{Mg}_{2/3}\text{Ta}_{1/3})\text{O}_3$ ceramics has ultrahigh breakdown field about 627 kV cm^{-1} , which conduce to achieving high $U_{re}=5.01 \text{ J cm}^{-3}$. Although the U_{rec} and η of NN-based ceramics have achieved a big breakthrough, they still have some shortcomings, such as the volatilization of Na ions at high temperature, complex phase transitions and sample poor stability, which is not conducive to its practical application.

Other properties

Except the U_{rec} and η , the cost of raw materials for dielectric capacitors is also another critical factor in whether it can be used to industrialization application. Herein, the prices of

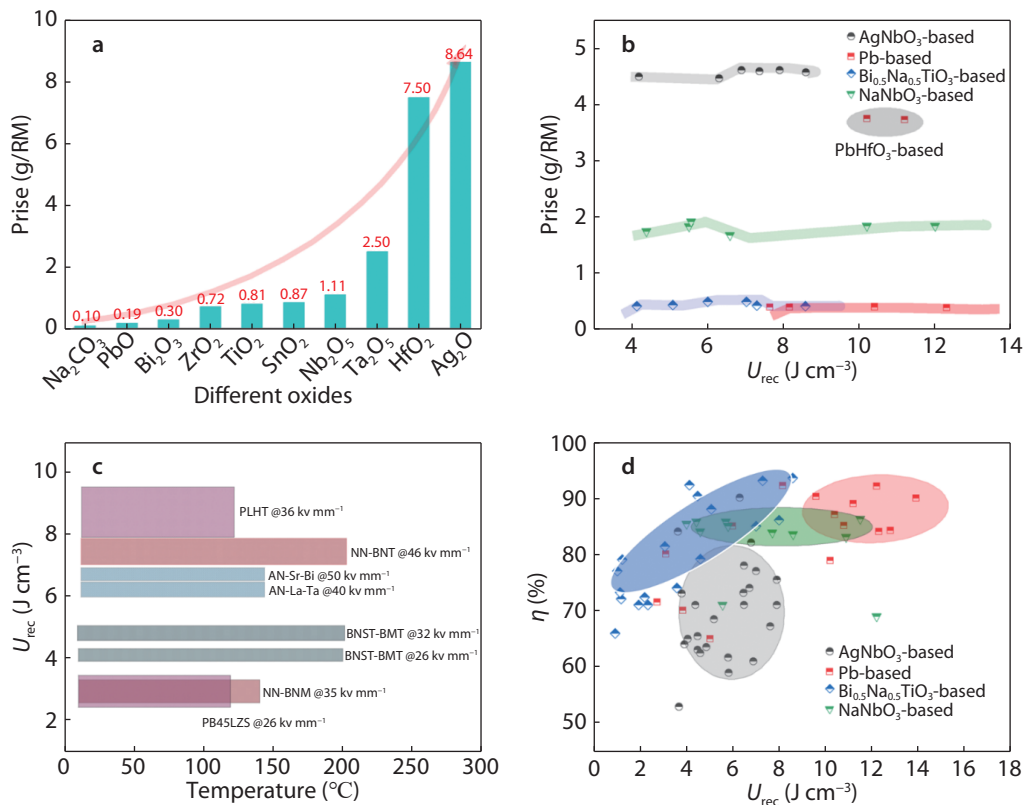


Fig. 3 a The prices of commonly used oxides. Comparison of b raw material price, c temperature stability and d energy storage performance of four typical antiferroelectric ceramics reported recently.

main four typical antiferroelectric raw materials (data from McLean and Aladdin website) are listed in Fig. 3a. Ag₂O and HfO₂ are the most expensive than others in these raw materials, which lead to the AgNbO₃-based and PbHfO₃-based AFE ceramics exhibited high raw material cost as shown in Fig. 3b [22–24,32,37,40,43,55,56,77,91–95] that is not conducive to their practical application. Besides, the temperature stability is also the crucial parameters to make sure the practical application of dielectric capacitor. The temperature stability of Bi_{0.5}Na_{0.5}TiO₃-based and AgNbO₃-based systems is better than that of NaNbO₃-based and Pb-based capacitors as shown in Fig. 3c [30,40,58,92,94,96,97]. This result suggests that Bi_{0.5}Na_{0.5}TiO₃-based and AgNbO₃-based ceramics can be used in more complex environments conditions. Fig. 3d presents the comparison of U_{rec} and η of these typical AFE ceramics [19–24,26,27,29,30,35,37,39,40,48,54–56,59,77,88–90,92,94,98–127]. The Pb-based capacitor possesses the highest U_{rec} than others, and an excellent η . In addition, the U_{rec} and η of Bi_{0.5}Na_{0.5}TiO₃-based and NaNbO₃ ceramics are higher than that of AgNbO₃-based systems.

Table 1. The advantages and disadvantages of four typical antiferroelectric systems.

	Pb-based	Bi _{0.5} Na _{0.5} TiO ₃ -based	AgNbO ₃ -based	NaNbO ₃ -based
U_{rec}	excellent	poor	well	well
η	well	excellent	poor	well
Materials price	low	low	high	midium
Human health	poor	excellent	excellent	well

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

The manuscript was drafted by Dong Liu and revised by Ting Tang and Prof. Li-Feng Zhu. All authors had approved the final version of the manuscript.

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Outlook and perspective

To meet the practical applications, the energy storage capacitors are needed to possess not only the high U_{rec} and η values, but also the excellent temperature stability and the low cost as well as environment friendly, and so on. The advantages and disadvantages of four typical antiferroelectric systems were shown in Table 1. Although the Pb-based AFE capacitors have a significant advantage in U_{rec} and η , its widespread adoption will be restricted because the hazardous element exists. In addition, AgNbO₃-based ceramic has an ultrahigh BDS, high U_{rec} and η , as well as excellent temperature stability, but it is also difficult to be used to marketization due to high cost for raw materials. However, for NaNbO₃-based and Bi_{0.5}Na_{0.5}TiO₃-based AFE systems, due to low cost for the raw materials, eco-friendly, high $U_{rec} > 10 \text{ J cm}^{-3}$ and $\eta > 85 \%$, as well as excellent temperature stability, they have a great potential in pulsed-discharge and power conditioning electronic devices as the energy storage capacitors.

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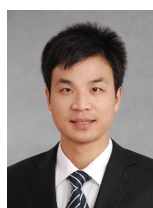


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