



Entropy-driven local structure regulation enables high energy density in NaNbO₃-based multilayer capacitors

Yipeng Zheng^{a,b}, Minghao Liu^b, Yujie Gao^b, Liangzhu Zhang^{a,*}, Deqiang Wang^{a,*},
Zhen Liu^b, Genshui Wang^{b,c,*}

^a School of Materials Science and Engineering, East China University of Science and Technology, Shanghai, 200237, China

^b Key Laboratory of Inorganic Functional Materials and Devices, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai, 200050, China

^c The State Key Laboratory of High Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai, 200050, China

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ABSTRACT

Multilayer ceramic capacitors (MLCCs) with high energy storage performance and robust reliability are urgently needed for miniaturized pulse power systems. However, the concurrent optimization of polarization response, hysteresis loss, and breakdown strength in lead-free dielectric ceramics remains challenging. Here, an entropy-driven local structure regulation strategy is proposed in NaNbO₃-based ceramics by introducing Ca (Mg_{0.2}Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2})O₃ as a high-entropy modifier. The resulting lattice distortion and antiferrodistortive oxygen-octahedral tilting reconstruct heterogeneous local polarization configurations and promote a relaxor-ferroelectric state with polar nanoregions. As a result, polarization saturation is delayed, hysteresis loss is suppressed, and breakdown strength is enhanced. The resulting MLCC achieves a high recoverable energy density of 14.60 J/cm³ with an efficiency of 85%, along with excellent thermal stability, fatigue endurance, and fast charge-discharge capability. This work highlights the effectiveness of entropy-driven local structure regulation strategy in optimizing energy storage performance and provides a practical route toward high performance MLCCs for advanced sustainable pulse power applications.

1. Introduction

Multilayer ceramic capacitors (MLCCs) are attractive energy storage components because of their ultrafast charge-discharge speed, high power density, and excellent reliability [1–3]. With the rapid development of advanced pulse power systems, there is a growing demand for ceramic capacitors with superior energy storage performance (ESP) [4,5]. The currently utilized energy storage dielectrics are lead-based ceramics (e.g. PbZrO₃-based), which exhibit superior ESP but inherently possess lead toxicity [6,7]. However, the relatively limited ESP of lead-free ceramics remains a major obstacle to their implementation in miniaturized and integrated electronic systems [8–10]. The recoverable energy density (W_{rec}) and efficiency (η) can be calculated from the formulas: $W_{\text{rec}} = -\int_{P_{\text{r}}}^{P_{\text{r}}} E dp$, $W_{\text{st}} = \int_0^{P_{\text{max}}} E dp$, $\eta = \frac{W_{\text{rec}}}{W_{\text{st}}} \times 100\%$ [11], where P_{max} and P_{r} represent the maximum polarization and remnant

polarization, respectively. Therefore, developing novel lead-free ceramics with large ΔP ($P_{\text{max}} - P_{\text{r}}$) and high breakdown electric field (E_{b}) is essential for promoting the energy storage applications of environmental-friendly high ESP MLCC in advanced pulse power systems [12,13].

NaNbO₃ (NN) is a representative lead-free ferroelectric/antiferroelectric (FE/AFE) system with a wide bandgap (~ 3.45 eV), high polarization strength ($\sim 40 \mu\text{C}/\text{cm}^2$) and low bulk density ($\sim 4.55 \text{ g}/\text{cm}^3$), thus exhibiting significant potential for energy storage applications [12]. Its ferroic behavior mainly originates from the long-range ordered displacement of Nb-centered dipoles together with cooperative oxygen-octahedral tilting, which gives rise to rich FE/AFE polymorphs and strong structural responsiveness to external perturbations [14]. Meanwhile, the structural diversity of NN and its highly tunable octahedral-tilting framework make it an attractive platform for local-structure

* Corresponding authors.

** Correspondence to: G. Wang, The State Key Laboratory of High Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai, 200050, China.

E-mail addresses: zhangliangzhu@ecust.edu.cn (L. Zhang), derek_wang@ecust.edu.cn (D. Wang), zhenliu@mail.sic.ac.cn (Z. Liu), genshuiwang@mail.sic.ac.cn (G. Wang).

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