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Dendritic nanopolar architectures: A paradigm for ultrahigh-density and radiation-hard dielectric energy storage

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Dielectric capacitors are re-emerging as essential energy-storage elements that bridge the gap between batteries and supercapacitors. Unlike typical electrochemical devices that rely on redox reactions, ferroelectric (FE) or dielectric capacitors store energy through reversible polarization of dipoles, offering ultrafast charge-discharge capabilities, high power density, and exceptional cycling stability. However, their relatively low energy density (U_e) and/or limited energy-storage efficiency remains a major bottleneck for advanced pulsed-power and microelectronic systems. This intrinsic limitation stems from a fundamental trade-off: large maximum polarization (P_m) is typically accompanied by high remanent polarization (P_r) and low breakdown strength (E_b). Achieving both large P_m and E_b with minimal hysteresis has long been an unresolved challenge. While chemical doping, defect engineering, and antiferroelectric conversions have offered incremental progress, these strategies often sacrifice either polarization strength or reliability.[1]

Vertically aligned nanocomposite (VAN) thin films with two epitaxial phases with perpendicularly oriented heterointerface, have been engineered to regulate ferroelectricity and piezoelectricity of oxides.[2] Their vertical heterointerfaces enable effective modulation of strain and depolarization fields, thereby enhancing ferroelectric and dielectric responses. However, in conventional VAN configurations, the applied electric field is typically oriented parallel to the interface planes, limiting their effectiveness in suppressing dielectric breakdown pathways (Figure 1A). As a result, while VAN structures may efficiently reduce P_r, achieving a simultaneous enhancement in E_b remains challenging.[3]

A recent breakthrough introduced a dendritic nanopolar (DNP) thin film architecture that redefines the classic polarization-breakdown trade-off. Using $PbZr_{0.53}Ti_{0.47}O_3$-MgO (PM) as a model system, Liu et al. realized a self-assembled dendritic network of ferroelectric domains separated by wide-bandgap insulating phase, rather than forming conventional VAN architecture (Figure 1A).[3] Thin films were grown by pulsed laser deposition (PLD) using mixed composite targets with varied PZT/MgO ratios, enabling precise control over film composition and stoichiometry (details refer to the Supplementary Materials).[3] The formation of dendritic architectures is closely tied to the growth conditions. Driven by growth kinetics, interphase surface energies, and phase-substrate interfacial energies,[2] adatoms from the PZT-MgO plume land on the substrate, diffuse, and migrate to energetically favorable sites, resulting in spontaneous phase separation and the development of the dendritic structure.

This distinctive dendritic topology forms a tortuous, branched network of FE-dielectric interfaces, analogous to a nanoscale "root system", that simultaneously enhances breakdown strength while maintaining high polarizability. The introduction of tilted and branched interfaces intersecting the electric field direction effectively elongates the breakdown pathway and homogenizes the local electric field distribution, thereby achieving superior energy-storage performance. This finding is also confirmed by the phase-field simulation results. As a result, the nanocomposite achieves an ultrahigh breakdown strength of 7.4 MV cm^{-1}, over three times compared to that of pure PZT. Simultaneously, the formation of nanoscale ferroelectric domains (1-3 nm) sustains strong polarization coupling across the MgO matrix, yielding high P_m (~ 100 μC cm^{-2}) with a drastically reduced P_r (~ 11 μC cm^{-2}). This optimized balance produces an exceptionally slim hysteresis loop and a remarkable U_e value of 215.8 J cm^{-3} with 80.7% efficiency, outperforming benchmark materials at comparable thickness, such as ion-irradiated $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (≈ 5.9 MV cm^{-1}, 133 J cm^{-3}), Sm-$BiFeO_3$-$BaTiO_3$ (≈ 5.3 MV cm^{-1}, 152 J cm^{-3}), and lead-free $Bi(Mg_{0.5}Ti_{0.5})O_3$-$SrTiO_3$ (≈ 7 MV cm^{-1}, 202 J cm^{-3}) (Figure 1B).[4] At a field of 4 MV cm^{-1}, U_e and η vary by less than 2% from -100°C to 150°C, indicating excellent thermal stability, while fatigue degradation after 10^{10} cycles remains < 6%. Importantly, this performance is achieved in a 100 nm-thick film, fully compatible with in-chip integration.

High-resolution microscopy analysis reveals atomically sharp but subtly inter-diffused PZT/MgO boundaries, where limited Mg^{2+} substitution induces local lattice distortions and mixed tetragonal-rhombohedral nanodomains, giving rise a relaxor-like state. This fine-grained disorder facilitates polarization rotation and domain-wall motion under high electric field while suppressing long-range ferroelectric coupling, precisely the condition required for large P_m, small P_r, and slim hysteresis. Furthermore, the bandgap disparity between PZT (3.4-3.5 eV) and MgO (7.8 eV) generates deep interfacial trap states that suppress leakage current, further stabilizing the high-field response. Hence, the DNP interfaces act as dual-function barriers: electrically blocking charge migration and mechanically confining nanodomains. This coupling of polar disorder and dielectric insulation underpins the unprecedented combination of high P_m, large E_b, and superior energy-storage efficiency in the DNP design.

Building upon the exceptional high-field stability of the PM films, Liu et al. exposed the DNP architecture to ionizing radiation environment-a frontier challenge in aerospace, nuclear, and deep-space electronics.[5] High-energy photons and particles typically induce displacement defects and charge traps within oxide dielectrics, leading to polarization fatigue, increased leakage current, and eventual dielectric failure. Remarkably, under γ-ray doses up to 20 Mrad, the PM films retain nearly all their polarization and energy density, that is U_e degradation ≤ 9%, far surpassing commercial polymer dielectrics such as the commercially available BOPP (breakdown ≤ 10 Mrad, U_e < 5 J cm^{-3}). Phase-field modeling and microstructural analyses attribute this extraordinary irradiation tolerance to the high density of FE-dielectric phase boundaries, which serve as efficient defect sinks (Figure 1C). Radiation-induced vacancies and interstitials are driven toward these interfaces, where they recombine and annihilate, effectively mitigating defect accumulation. Only at extreme exposure (40 Mrad), defect clusters accumulate at interfaces, inducing domain pinning and reduced polarization. The authors further demonstrate that these PM films maintain superb thermal endurance (-100°C to 170°C), ultrafast discharge ($\tau_{0.9}$ ≈ 3.4 μs), and fatigue resistance (> 10^{10} cycles) even under 20 Mrad exposure, underscoring their robustness under multi-extreme conditions.

The remarkable performance of the DNP films can be traced to a multi-scale interplay between nanoscale polarization dynamics and mesoscopic defect kinetics, as outlined below: (1) Polar-Nanoregion Formation: Local chemical fluctuations and interfacial strain fields induce nanoscale polar regions with randomized orientations, yielding relaxor-like behavior with high dielectric diffuseness (γ ≈ 1.8-1.9); (2) Domain Miniaturization: Confinement by MgO branches limits long-range order, reducing ferroelectric domain size from > 10 nm to 1-3 nm, lowering polarization switching barriers and minimizes hysteresis loss; (3) Field Redistribution: Branched MgO channels increase the tortuosity of conduction pathways, homogenizing local electric fields, mitigating hotspot formation, and effectively suppressing electrical treeing; (4) Defect Trapping and Self-Healing: Interface-related deep traps immobilize carriers and act as sinks for irradiation-induced defects, enabling

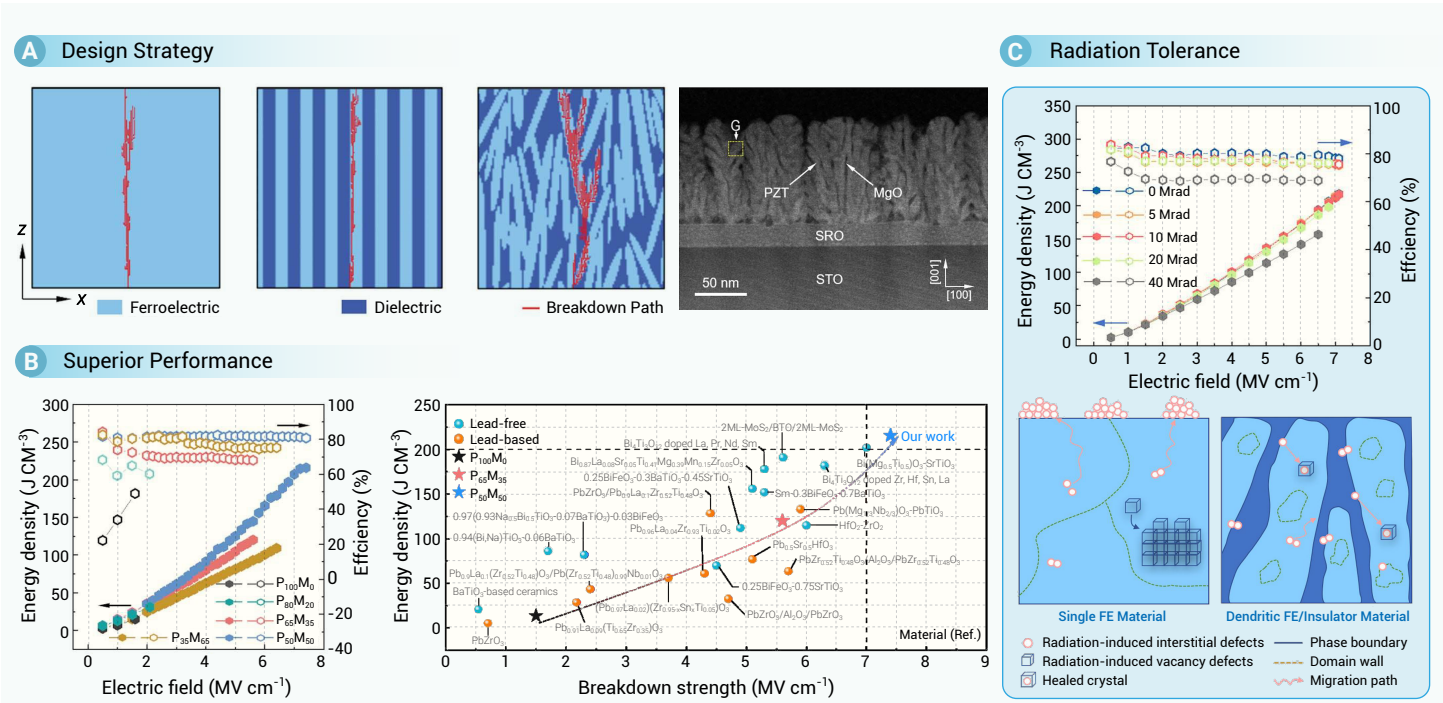

Figure 1. Design strategy, superior performance and radiation tolerance of the dendritic nanopolar thin films (A) Comparison of breakdown paths for a pure FE, a VAN structured FE/dielectric nanocomposite thin film, and a DNP structured FE/dielectric nanocomposite thin film; (B) Energy density and efficiency as functions of electric field up to E_b (left), and Comparison of the energy density and the breakdown strength of DNP films with the state-of-the-art dielectric films (right); (C) Energy density and efficiency at electric fields up to E_b (upper), and schematic illustration of possible self-healing evolution modes for conventional single FE film and DNP film with a high density of phase boundaries (lower).

radiation tolerance; (5) Elastic Compatibility: the relatively close lattice matching of PZT and MgO minimize misfit strain accumulation, might avoid premature breakdown from mechanical cracking. Collectively, these multiscale effects transform the energy-storage process from a simple charge-displacement cycle to a dynamic equilibrium among polarization rotation, defect migration, and interfacial charge stabilization.

Beyond its immediate achievements, the DNP framework establishes a materials-agnostic design principle. While demonstrated in Pb-based perovskites, the underlying DNP architecture could possibly be extended to lead-free systems, such as HfO_2-, $BaTiO_3$-, or $BiFeO_3$-based systems. In HfO_2-based ferroelectrics, for instance, incorporating a dendritic dielectric phase (e.g., Al_2O_3 or SiO_2) could stabilize non-centrosymmetric nanoregions and mitigate leakage at scaled thicknesses. Likewise, perovskite-oxide dendrites embedded in GaN or SiC semiconductors could yield ferroelectric-semiconductor hybrid capacitors with unprecedented charge modulation for low-power transistors or neuromorphic devices. Beyond functional electronics, the radiation-resilient DNP architecture holds compelling prospects for space-grade and aerospace applications, where materials must endure high electric fields, temperature swings, and intense particle irradiation. The observed self-healing behavior under γ-irradiation could be leveraged to achieve autonomous, defect-resistant oxide circuits-an emerging requirement for long-duration lunar or Martian missions.

Nevertheless, several challenges remain before DNP architectures transition from laboratory prototypes to practical microcapacitors: (1) Scalability: achieving controlled dendritic morphology across wafer scales demands precise growth kinetics or lithographic templated deposition; (2) Interface Quantification: mapping trap state distributions and band alignment requires correlative scanning-probe and *ab initio* methods; (3) Dynamic Self-Healing Kinetics: Real-time *in situ* irradiation or under bias experiments using TEM or synchrotron sources could elucidate defect-recovery kinetics; (4) Sustainability: lead-free candidates shall be prioritized; (5) Integration with Semiconductors: direct coupling with wide-bandgap semiconductors (e.g. GaN, SiC, or β-Ga_2O_3) may enable next-generation negative-capacitance or energy-recovery transistors, merging energy storage and logic functionality in a single platform. In perspective, mastering these aspects will determine whether DNP films evolve into a unifying platform for high-density energy storage and adaptive oxide electronics, bridging materials design, device integration, and space-resilient functionality.

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DECLARATION OF INTERESTS

The authors declare no competing interests.