Ultrahigh Dielectric Strength and Energy Density in Ultrathin Polymer Films via Confinement and Interface Engineering

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Ultrathin polymer dielectrics are a promising platform for nanoelectronics, enabling capacitor scaling with low loss and mechanical compliance.[1-3] Yet a key question remains: are the extraordinary breakdown fields in confined films unique to stiff, glassy polymers, or a more universal interface-driven effect that can be engineered into reliable nanocapacitors?[4] Here we show that confinement plus interface control yields record breakdown strength and device-level energy storage in both glassy PMMA and soft elastomeric PDMS (Figure 1). Spin-coated films (≈14 nm-μm) were measured using a non-destructive eutectic Ga-In (EGaIn) droplet electrode to avoid sputter-induced metal infiltration that prematurely fails sub-500 nm films. Weibull analysis reveals a sharp upturn in breakdown strength below ~0.5-1 μm, reaching ≈2.0 GV m^{-1} for ~14 nm PMMA and ≈0.96 GV m^{-1} for ~15 nm PDMS, demonstrating that even low-modulus dielectrics can access the ultrathin high-E_{BD} regime relevant to nanoelectronic architectures. These films translate into linear nanocapacitors delivering 85.2 J cm^{-3} (PMMA) and 78 J cm^{-3} (PDMS) with 75-90% efficiency, stable over 10^6 charge-discharge cycles, approaching ferroelectric-class energy densities while retaining low-loss behavior. A simple interfacial-series model, $E_{\mathrm{eff}} \propto V_{\mathrm{int}}/t + C$ ($R^2 \approx 0.99$ for PMMA), identifies a dominant interfacial voltage drop V_{int} in the ultrathin limit, supported by independent adhesion/surface-energy trends and tunable via polymer-brush interlayers for thermal stabilization. Together, these results establish an interface-engineered, scalable route to high-field, low-loss polymer nanodielectrics and compact nanocapacitors for next-generation nanoelectronics systems.

References

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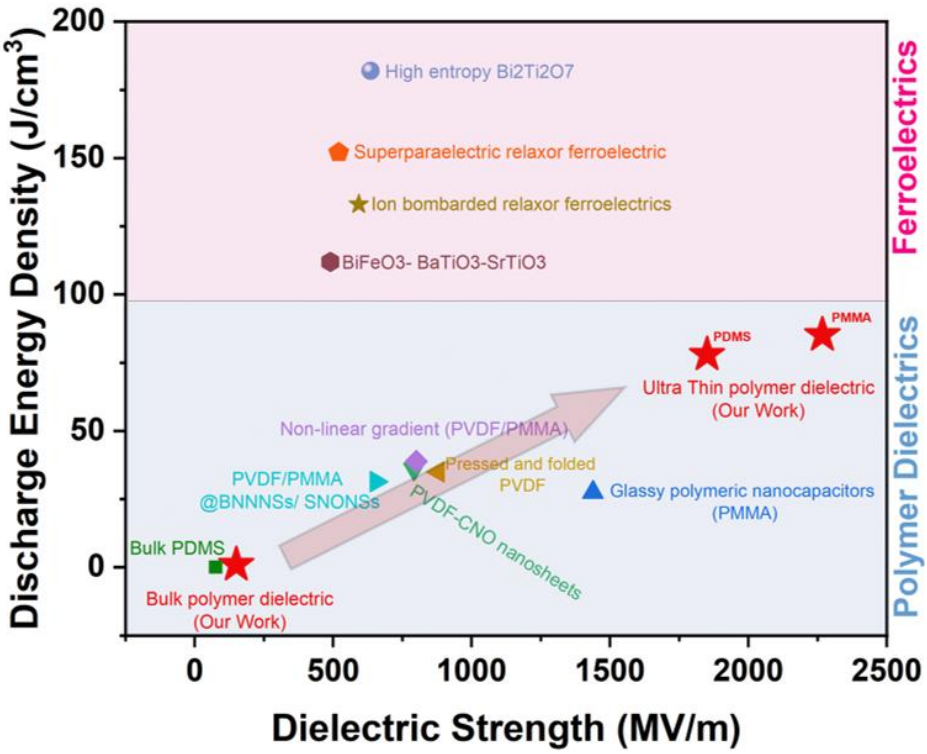

Figure 1. plot showing ultra thin polymer dielectric performance in comparison to dielectrics available literature. The performance map compares discharge energy density against dielectric strength for representative dielectric systems. Our ultrathin polymer dielectrics (red stars; PDMS and PMMA) occupy the extreme upper-right of the polymer-dielectric domain, reflecting the quadratic leverage of breakdown strength on energy density and validating the interfacial/thickness engineering developed in this work. Notably, the achieved energy densities approach those reported for advanced ferroelectric and relaxor systems, yet our devices are linear dielectrics that maintain high efficiency with low loss, avoiding the heating and hysteresis penalties typical of ferroelectrics.