

## Supporting Information

### **Crosslinking Modification and Hydrogen Bonding Synergy to Achieve High Breakdown Strength and Energy Density of PMMA-co-GMA/PVDF Dielectric Composite Films**

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## **Characterization**

Fourier transform infrared spectroscopy (FTIR, Thermo Nicolet Avatar-360 spectrometer, ATR mode) was carried out to analyze the reaction between the amino group of PEA and epoxy group of MG and multiple hydrogen bonding interactions by the ATR mode over a range of 700~2000  $\text{cm}^{-1}$  with a  $32\text{cm}^{-1}$  resolution. The elemental analysis of the specimens was characterized by X-ray photoelectron spectroscopy (XPS) (Thermo Fisher Scientific K-Alpha, USA). The high-resolution XPS spectra was analyzed by the software of “Avantage”. Scanning electron microscopy (SEM) images were acquired on a JSM6510 instrument. The cross-section of the sample was obtained by freeze fracture in liquid nitrogen and tested with a thin layer of gold applied to the fracture surface. The dielectric properties were measured by using the LCR Meter (E4980A, Keysight Technologies, USA) under a frequency range of  $10^2$ - $10^6$  Hz. All the samples were coated with gold electrodes on both surfaces before measuring. The specimens were placed in the specimen chamber of thermally stimulated depolarization current (TSDC, Polyk) for the TSDC measurement. The discharge energy density, efficiency, leakage current, and Polarization-electric field (P-E) hysteresis loops of all the samples were measured by a ferroelectric test system (CPE1901, Polyk) at 1000 Hz at room temperature. The breakdown strength measurements were used by the same equipment to measure the P–E loops at a direct-current voltage ramp of 500 V/s.

## **DFT Calculation**

The density functional theory (DFT) is calculated by the Materials Studio 2020

software at the Dmol3 code. All models require structural optimization before the calculations. To simplify the calculation, the molecular model of PEA contains only one repeat unit with two amino groups and the molecular model of MG was defined as containing one MMA unit and one GMA unit. The molecular model of PEA/MG contains one PEA model and two MG models after the reaction.