## Achieving High Breakdown Strength and Energy Density in All-Organic Sandwich Structured Dielectrics via Introducing

## **Polyacrylate Elastomers**

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## Characterization

The morphology of films were characterized by scanning electron microscopy (SEM, JSM6390). Differential scanning calorimetry (DSC) traces of the films were obtained using a TA Q10 DSC instrument at a heating rates of 10°C/min during the first heating process. 1D WAXD experiments were performed on a BRUKER AXS D8 advance diffractometer with a 40 kV FL tubes as the X-ray source (Cu Ka) and a LYNXEYE\_XE detector. The scanning speed of 1D WAXD was 2°/min. Gold electrodes with a thickness of 100 nm and a diameter of 3.14 mm were sputtered on two sides of films. The dielectric performance was measured using an Agilent 4294A LCR meter with a frequency range from 10<sup>3</sup> Hz to 10<sup>7</sup> Hz at room temperature. The polarization-electric field hysteresis loops and fatigue test of charge-discharge cycling of films were performed at 200 Hz by a TF analyzer 2000 ferroelectric polarization tester (aixACT, Germany). The discharge rate of films were performed by dielectric material charge measurement system DCQ-20A.



Figure S1. Cross-section SEM images of positive-sandwich structured film M-15-M with stretching ratio (a) 0, (b) 1, (c) 2, (d) 3 and (e) 4, respectively.



Figure S2. (a) The XRD and (b) DSC of positive-sandwich structured M-15-M films with different stretching ratio.



Figure S3. The frequency dependence dielectric constant of 20 wt%-PMMA/PVTC blending single layer film and DE/PVTC blending single layer films with the 0 wt%, 5 wt%, 15wt% and 20 wt% DE content, respectively.