Supporting Information

PEI-based All-organic Composite Film with Simultaneous Excellent Energy

Storage Density and High Efficiency

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Fig. S1. *P-E* loops of PVDF/PEI bilayer composite films under different electric fields.



Fig. S2. Electric field simulation of the bilayer composite films cross-section by finite element analysis.

Source of raw materials

Analytical-grade Polyetherimide (PEI) was provided by SABIC, Co., Ltd. Poly (vinylidene fluoride) (PVDF) was supplied by Arkema Co., LLC. N-methyl-2-pyrrolidone (NMP) were bought from Sinopharm Chemical Reagent Co., Ltd. Other chemicals and reagents were obtained from Tianjin Fuyu Fine Chemical Co., Ltd.

Electrical breakdown model in bilayer structure PVDF/PEI composite films

The breakdown process was studied according to the following formula:

$$P(i, k \to i', k') = A \frac{(\phi_{i,k'})^{\eta}}{\sum (\phi_{i,k'})^{\eta}} + B \frac{\phi_{i,k'}}{\phi_0} + C$$
(S1)

where ϕ is the electric potential for all the lattice points, *i*, *k* and *i'*, *k'* represent the discrete lattice coordinates, ϕ_0 is the threshold electric potential; η is the fractal dimension, which depicted the relationship between the local field and probability. The above equations describe the growth direction of the electric trees, the difficulty in growing the electric tree, and the dielectric properties of the materials. The coefficients of *A*, *B* and *C* determine the weight of each term of the equation.

The growth of electric trees grows at adjacent grid points with the probability of *P* (*i*, $k \rightarrow i'$, k'), which depicts the relationship between local field and probability. The breakdown strength of the bilayer structure PVDF/PEI composite films can be adjusted quantitatively by parameter *A*, *B*, *C* and ϕ_0 . In this work, ϕ_0 , *A*, *B*, and *C* are 0.016, 1.0, 0.00021, and 0.34 for PVDF, respectively. For PEI, ϕ_0 , *A*, *B*, and *C* are 0.00265, 1.0, 0.0016, and 0.474, respectively.