Supporting Information

Ultrahigh Breakdown Strength and Discharge Energy Density of Newly Designed Asymmetric Sandwich-Structured PVDF-based Nanocomposite Film

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Source of raw materials

Barium titanate (BaTiO₃) with average particle size of 100 nm were purchased from Sinocera Co., Ltd. Boron nitride powder (BN) with average particle size of 300 nm was purchased from Sigma Aldrich Co., Ltd. Poly(vinylidene fluoride) (PVDF) and Polyvinylidene fluoride-hexafluoropropylene copolymer (PVDF-HFP) were provided by Arkema, Kynars Co., Ltd. Isopropanol and N, N-dimethylformamide (DMF) were bought from Sinopharm Chemical Reagent Co., Ltd.

Formula and parameters for electrical breakdown model

The breakdown process was studied according to the following formula:

$$p(i,k \to i',k') = A \frac{(\phi_{i',k'})^{\prime\prime}}{\sum (\phi_{i',k'})^{\prime\prime}} + B \frac{\phi_{i',k'}}{\phi_0} + C$$
(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 asymmetric sandwich-structured nanocomposite PVDF-based films could be adjusted quantitatively by parameter A, B, C and ϕ_0 . In this work, ϕ_0 , A, B, and C are 0.0016, 1.0, 0.00002, and 0.61 for 2D BN nanosheets, respectively. For PVDF, ϕ_0 , A, B, and C are 0.12, 1.0, 0.0003, and 0.306,

respectively. For P(VDF-HFP), ϕ_0 , A, B, and C are 0.12, 1.0, 0.00024, and 0.34, respectively. For 0D BT powders, ϕ_0 , A, B, and C are 9, 1.0, 0.004, and 0.6, respectively.



Fig. S1. (a) *P-E* loops and (b) energy storage performance of the asymmetric sandwich-structure nanocomposite film containing 5 vol% 2D BN nanosheets in the breakdown cut-off layer.