Supporting Information

Synergistically Enhanced Discharged Energy Density and Efficiency Achieved in Designed Polyetherimide-based Composites via Asymmetrical Interlayer Structure Induced Optimized Interface Effectiveness

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Source of raw materials

Analytical-grade powders of $NaCO_3$ (>99.8%), TiO_2 (>99.8%), Bi_2O_3 (>99.8%) and NaCl (>99%) were purchased from Sinopharm Chemical Reagent Co., Ltd. Boron nitride (BN) (>99.8%) powder with average particle size of 300 nm was purchased from Sigma Aldrich Co., Ltd. Polyetherimide (PEI) (>99%) was provided by SABIC, Co., Ltd. Analytical-grade Isopropanol and N-Methylpyrrolidone (NMP) were bought from Sinopharm Chemical Reagent Co., Ltd.

Equation and parameters for electrical breakdown model

The breakdown process was studied according to the following eqn:

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

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 $P\ (i, k \to i', k')$, which depicts the relationship between local field and probability. The PEI-based composite films with different structures could be adjusted quantitatively by parameter ϕ_0, A, B, and C. In this work, ϕ_0, A, B, and C are 9, 1.0, 0.0012, and 0.29 for $Na_{0.5}Bi_{0.5}TiO_3$ (NBT) ceramic fibers, respectively. For PEI, ϕ_0, A, B, and C are 0.12, 1.0, 0.0004, and 0.336, respectively. For BN nanosheets, ϕ_0, A, B, and C are 0.0016, 1.0, 0.00002, and 0.61, respectively.

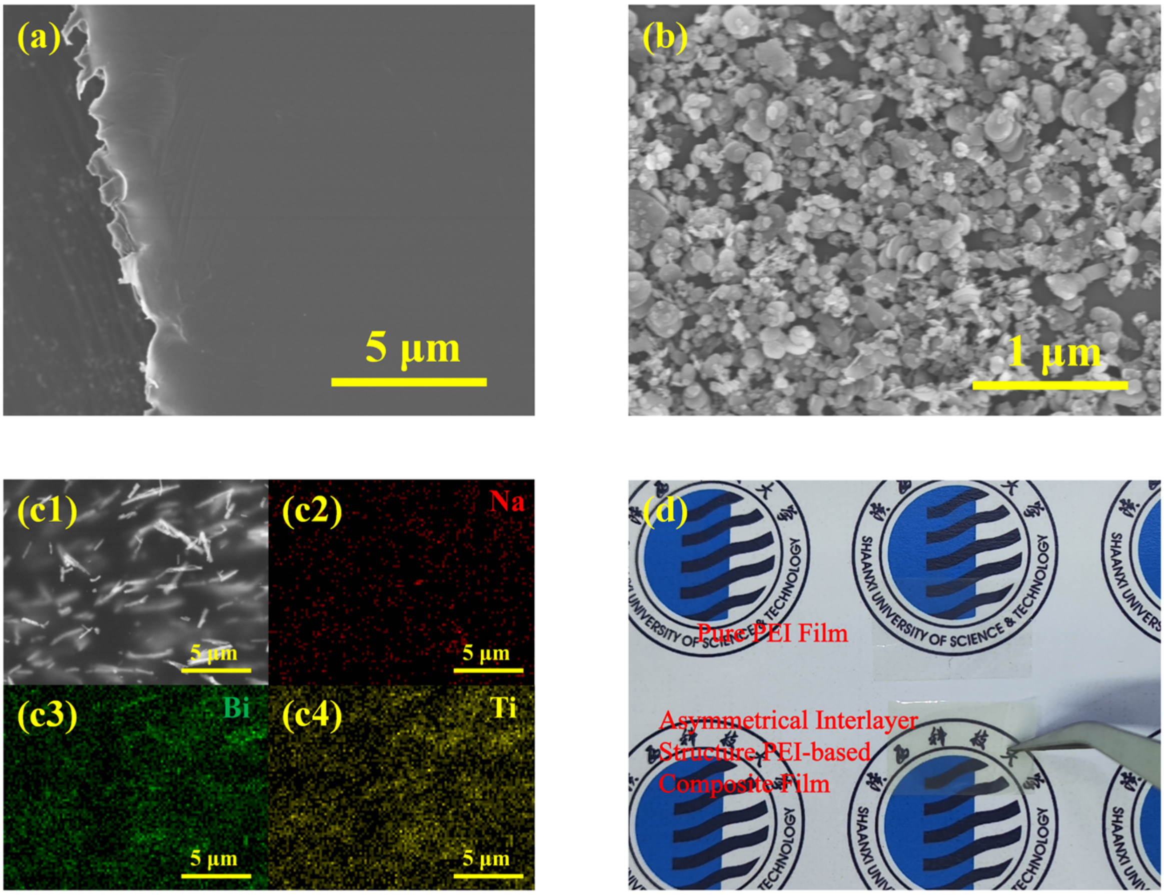

Fig. S1. Surface SEM images of (a) pure PEI layer, (b) BN pure inorganic layer and (c) NBT/PEI composite layer (including elemental mapping analysis for Na, Bi, and Ti) of asymmetrical interlayer structure PEI-based composite film. (d) Shadowless lamp photograph of the asymmetrical interlayer structure PEI-based composite film and pure PEI film.

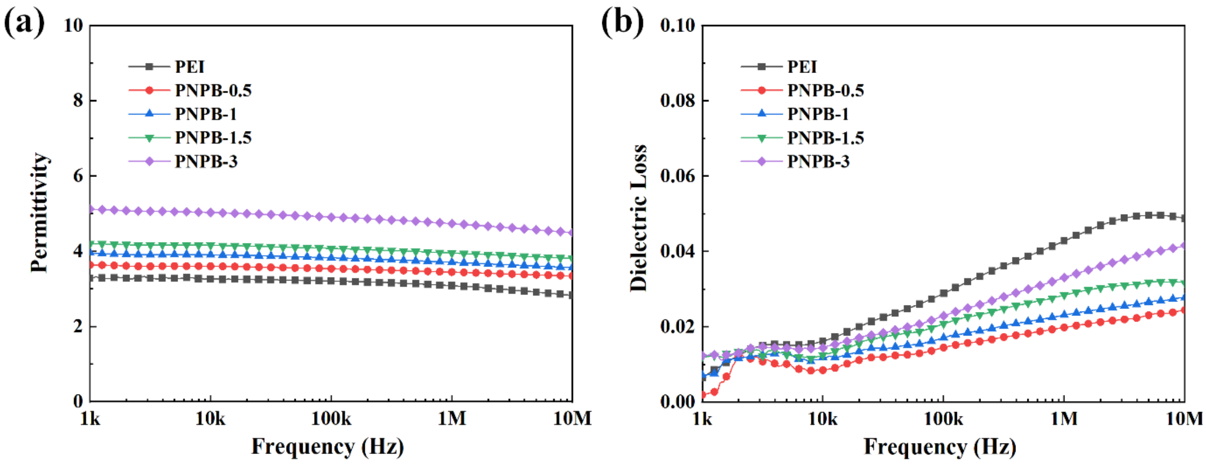

Fig. S2. Frequency dependencies of (a) permittivity and (b) dielectric loss of the asymmetrical interlayer structure PEI-based composite films and pure PEI films at ambient temperature.

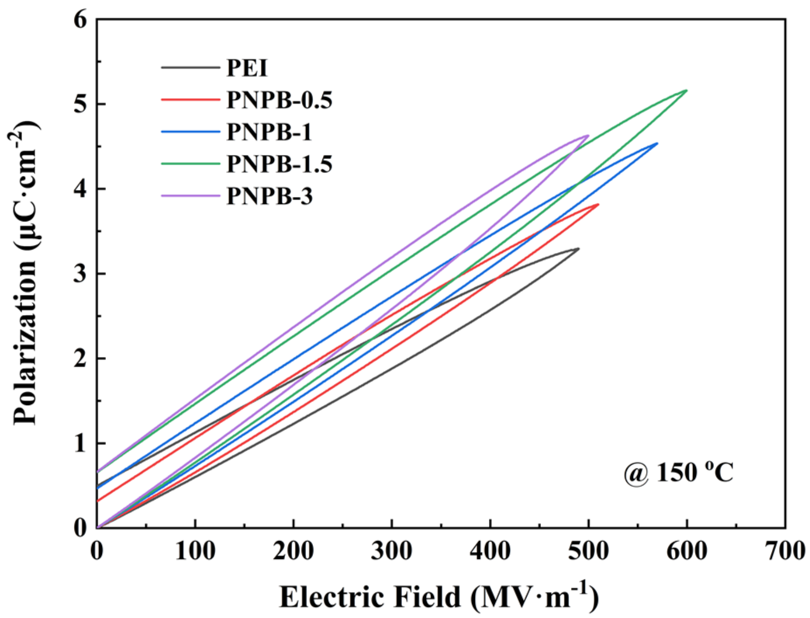

Fig. S3. *P-E* loops at the maximum breakdown electric field of asymmetrical interlayer structure PEI-based composite films and pure PEI film at 150 ^{o}C.

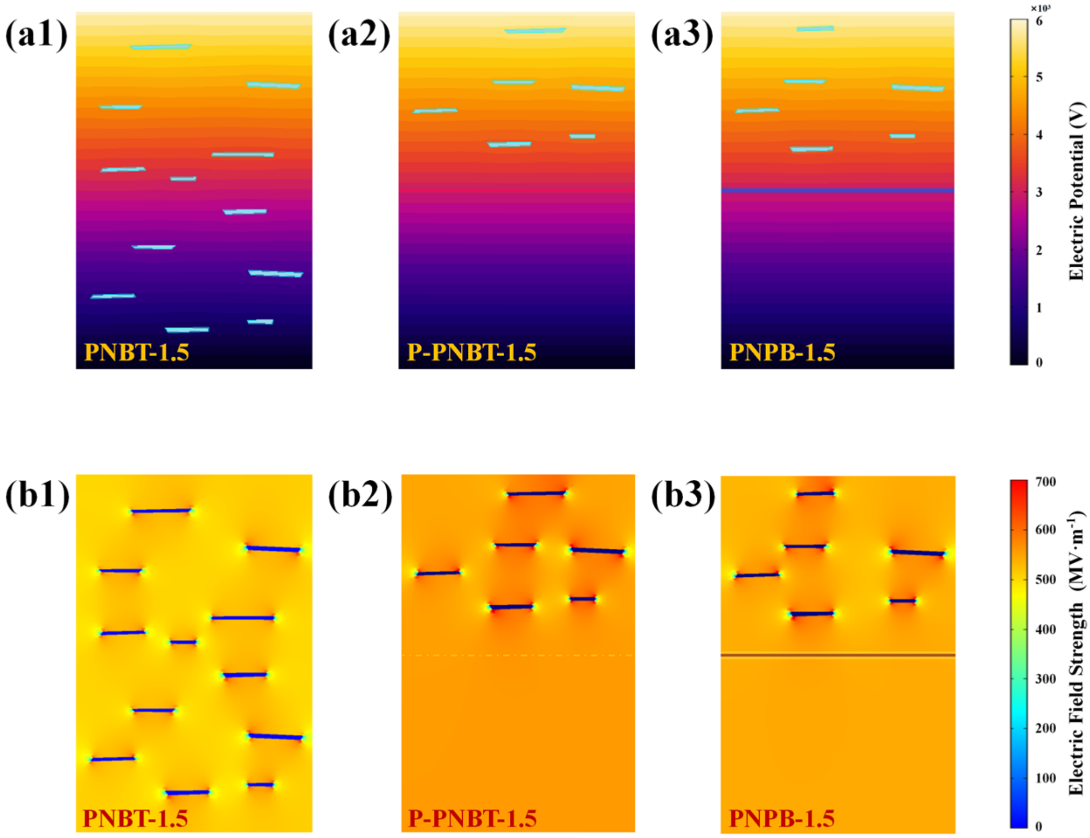

Fig. S4. (a) Potential distribution simulation and (b) electric field distribution simulation of the PEI-based composite films with different structures.