Supporting Information

Superior High-Temperature Energy Storage Performance in Multilayer PEI-Based Nanocomposites via Functional Fillers Integration

Xiangying Li,^a Haibo Yang,^{*a *b}Yanlong Ma^b and Ying Lin^{*b}

^aSchool of Electronic Information and Artificial Intelligence, Shaanxi University of Science and Technology, Xi'an, Shaanxi 710021, China. ^bShaanxi Key Laboratory of Green Preparation and Functionalization for Inorganic

Materials, School of Materials Science and Engineering, Shaanxi University of Science and Technology, 710021 Xi'an, China

*Corresponding authors:

- (Haibo Yang*a) E-mail: <u>yanghaibo@sust.edu.cn</u>
- (Ying Lin*a) E-mail: <u>linying@sust.edu.cn</u>



Fig. S1 (a) Fiber diameter distribution of BSTNFs. (b)SEM images of BNNSs.



Fig. S2 (a) SEM images of BSTNFs. Elemental mapping analysis for (b) O, (c) Ba, (d) Sr and (e) Ti of BSTNFs.



Fig. S3 (a) cross-section SEM images of NBN-1.(b) NBN-3.(c) NBN-7 nanocomposites.



Fig. S4 (a) Cross-section SEM image of the three-layer structured nanocomposites. Elemental mapping analysis for (b) C, (c) O, (d) N, (e) B, (f) Ba, (g) Sr, and (h) Ti of the three-layer structured nanocomposites.



Fig. S5 Temperature dependence of dielectric properties of NBN-5 nanocomposites.



Fig. S6 P-E loops of pure PEI and NBN-x three-layer structured nanocomposites at 350 MV/m.



Fig. S7 (a) $U_{\rm e}$, η values and (b) *P-E* loop of the NBN-5 nanocomposite under an applied electric field of 250 MV/m for fatigue cycles ranging from 1 to 10^5



Fig. S8 (a) Schematic structure of 5 vol%. (b) and 0-5-0.(c) SEM images of cross sections at 5 vol%. (d) and 0-5-0.



Fig. S9 (a) P-E loops of different structured nanocomposites at room temperature



Fig. S10 *P*-*E* loops of the three-layer structured nanocomposites under different electric fields.

Electrical breakdown model in three-layer structured PEI-based nanocomposites

The breakdown process was studied according to the following formula:

$$P(i,k \to i',k') = A \frac{(\varphi_{i,k'})^{\eta}}{\sum (\varphi_{i,k'})^{\eta}} + B \frac{\varphi_{i,k'}}{\varphi_0} + C$$
(S1)

where *i*, *k* and *i'*, *k'* represent the discrete lattice coordinates, φ denotes the electric potentials of all lattice points, φ_0 is the threshold electric potential, and η is the fractal dimension, which reflects the relationship between the local field and the probability. The above equations describe the growth direction of electric trees, the difficulty of electric trees growth and the dielectric properties of materials, respectively. *A*, *B* and *C* are the three coefficients in the equation, which identify the weight of each term.