Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Synergistically Ultrahigh Energy Storage Density and Efficiency in Designed Sandwich-structured Poly(vinylidene fluoride)-based Flexible Composite Films Induced by Doping $Na_{0.5}Bi_{0.5}TiO_3$ Whiskers

Ying Lin,[1a] Yongjing Zhang,[1a] Shili Zhan,[b] Chuang Sun,[a] Guangliang Hu,[a]

Haibo Yang*[a] and Qibin Yuan*[b]

[a]*Shaanxi 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*

[b]*School of Electronic Information and Artificial Intelligence, Shaanxi University of Science and Technology, Xi'an, Shaanxi 710021, China.*

*Corresponding authors

(Haibo Yang*[a]) E-mail: yanghaibo@sust.edu.cn

(Qibin Yuan*[b]) E-mail: yuanqibin-sust@163.com

[1]Author Contributions

Ying Lin and Yongjing Zhang contributed equally to this paper.

The dielectric constant of NBT ceramics was used to approximate the dielectric constant of NBT whiskers. We prepared the NBT ceramic sheet by isostatic pressing and tested its dielectric properties via E4990A, as shown in Fig. S1.

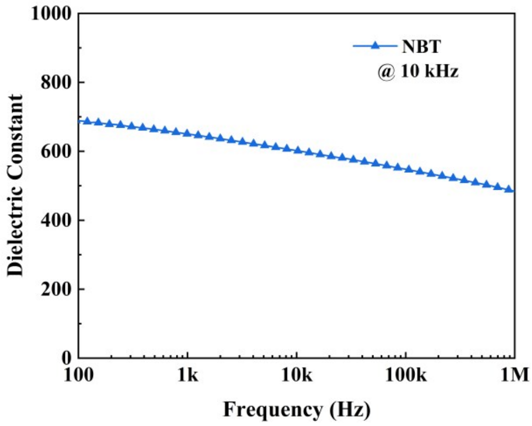

Fig. S1. Frequency dependency of dielectric constant of NBT ceramic.

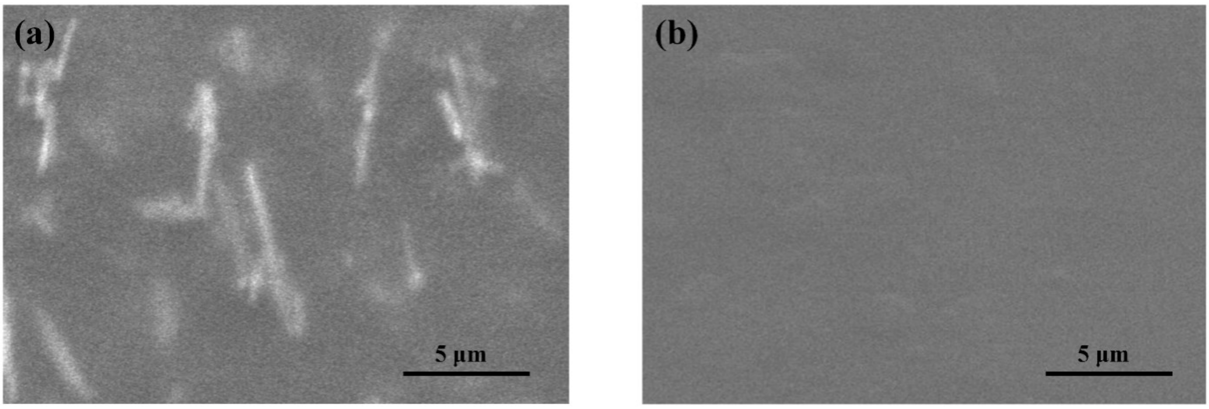

Fig. S2. SEM images of the surfaces of (a) the inner layer and (b) the outer layer of sandwich-structured NBT/PVDF composite films.

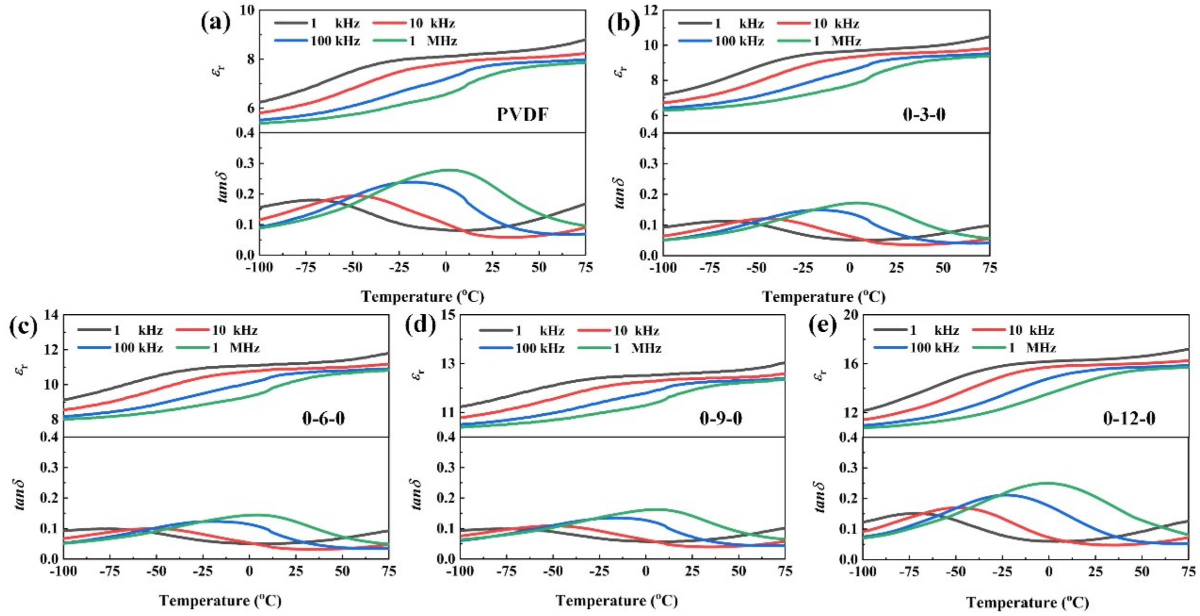

Fig. S3. Dielectric properties of the pure PVDF films, 0-3-0 composites, 0-6-0 composites, 0-9-0 composites and 0-12-0 composites as functions of temperature at different frequencies.

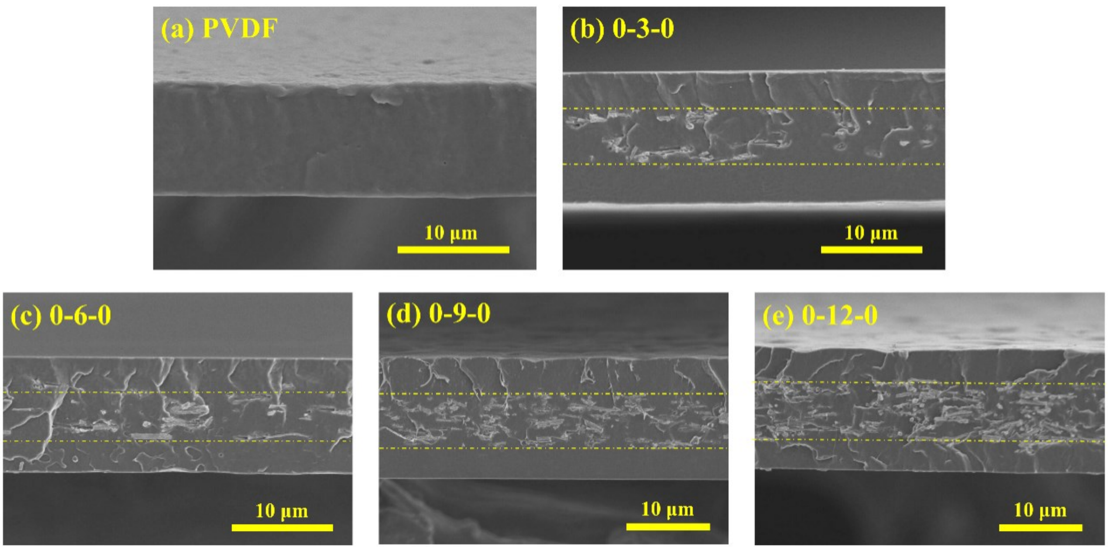

Fig. S4. Cross-section SEM of the pure PVDF films, 0-3-0 composites, 0-6-0 composites, 0-9-0 composites and 0-12-0 composites.

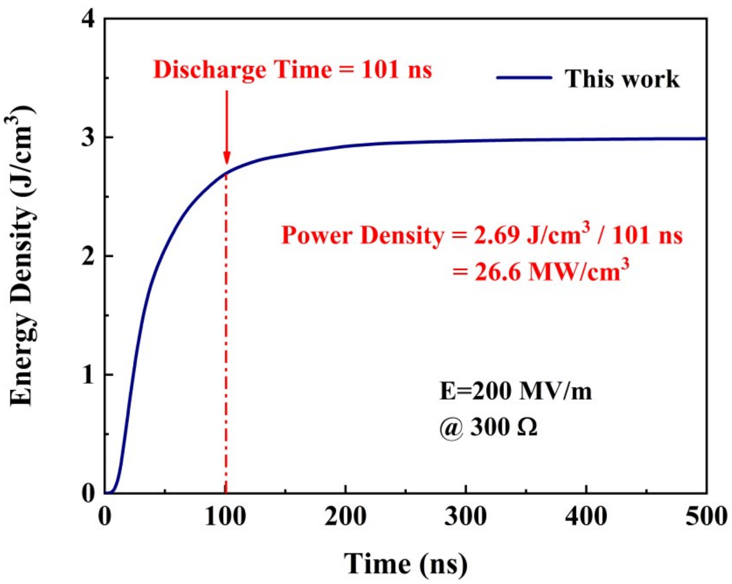

Fig. S5. Discharged energy density as a function of time of the sandwich-structured NBT/PVDF composite films under an electric field of 200 MV/m.

Electrical breakdown model in sandwich-structured NBT/PVDF composite films

The breakdown process was studied according to the following formula:

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

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 depicts 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 sandwich-structured NBT/PVDF composite films can be adjusted quantitatively by parameter A, B, C and ϕ_0. In this work, ϕ_0, A, B, and C are 0.08, 1.0, 0.0016, and 0.61 for NBT whiskers, respectively. For PVDF, ϕ_0, A, B, and C are 6, 1.0, 0.003, and 0.34, respectively.

Algorithm of average electric field in different layers of the composite films

The average electric field in different layers of sandwich-structured NBT/PVDF composite films was calculated by the following equations:

$$E_s = V/(2d_s + d_h\varepsilon_s/\varepsilon_h) \tag{S2}$$

$$E_h = V/(d_h + 2d_s\varepsilon_h/\varepsilon_s) \tag{S3}$$

where V is the applied electric voltage on the sandwich-structured NBT/PVDF composite films, E_s is the applied electric field on the outer layer, E_h is that on the inner layer; d_s is the thickness of the outer layer, d_h is that of the inner layer; ε_s is dielectric constant of the outer layer, and ε_h is that of the inner layer.