Supplementary Information (SI) for Nanoscale.
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Supporting Information

Polymer nanocomposites reinforced with Al_2O_3 nanoplates for high-temperature capacitive energy storage application

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Aluminum oxide particle size statistics:

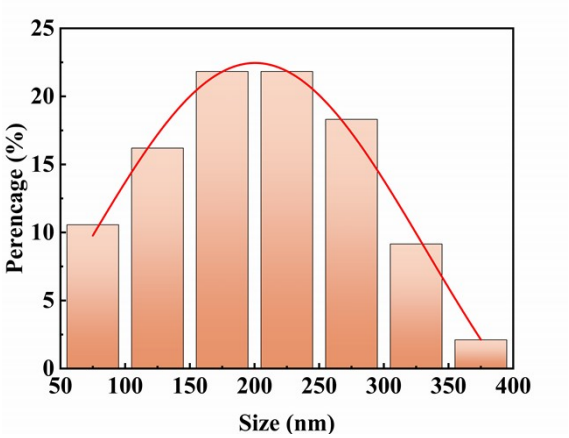

Fig. S1 Particle size statistics of alumina nanoplates.

Electric field intensity simulation:

In order to simplify the calculation, a two-dimensional model is established. The two-dimensional model diagram is shown in **Fig. S2**, and the length and width are set to 40 μm and 20 μm respectively, where **Fig. S2a** is the FPE model and **Fig. S2b** is the FPE/Al_2O_3 NPLs model.

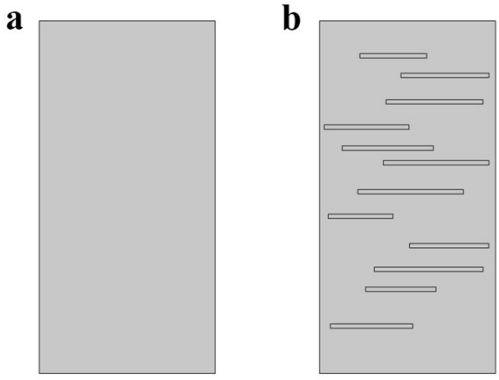

Fig. S2 Schematic diagram of the models of (a) FPE and (b) FPE/Al_2O_3 NPLs.

An electric field of 200 MV m^{-1} is vertically applied on two models from top to bottom, in which the bottom is set to be ground. In addition, the ambient temperature was set to 423.15 K. The relative dielectric constant (ε_r) of FPE and Al_2O_3 nanofillers are set to be (3.0, 9.5), respectively.[1] The electric field (E) can be expressed as:

$$E = -\nabla\varphi \tag{1}$$

where φ is the electric potential in V. The two-dimensional model was calculated using the finite element method.

Current density simulation:

The leakage current density simulation is based on the jump conduction model, and the current density (J) can be calculated:

$$J(E,T) = 2ne\lambda v * \exp(-\frac{W_a}{k_B T}) * \sinh(\frac{\lambda eE}{2k_B T}) \tag{2}$$

where n and e are carrier density and carrier charge, respectively, λ is the hopping distance, v is the attempt-to-escape frequency, W_a, k_B, T and E represent activation energy, Boltzmann constant, Kelvin temperature and electric field, respectively. Equation (2) can be simplified as follows:

$$J(E) = A * \sinh(B * E) \tag{3}$$

where A and B are two lumped parameters, which can be obtained by fitting the experimental J-value, and finally the current density is simulated by finite element method.

Dielectric breakdown process simulation:

To systematically investigate the influence of alumina nanoplates morphology on the dielectric breakdown strength of polymer nanocomposites, a comprehensive phase-field modeling approach was employed to simulate the breakdown process in nanocomposite with different nanoplates.[2, 3] A scalar phase field $s(x,t)$ was introduced to quantify the breakdown behavior in the polymer nanocomposites, where $s = 1$ corresponds to the initial state and $s = 0$ denotes the fully damaged state. The x and t are position and time, respectively. For an intermediary state, the ε for Al_2O_3 and FPE can be expressed as:

$$\varepsilon(s) = \frac{\varepsilon_{\text{ini}}}{f(s) + \eta} \tag{4}$$

where $f(s) = 4s^3 - 3s^4$.$\varepsilon_{\text{ini}}$is the initial dielectric constant and η is a constant of low value. Here, the ε_{ini} values of Al_2O_3 and FPE were set to 9.5 and 3, respectively, and η was set to 10^{-3}, From a thermodynamic perspective, dielectric breakdown occurs if the process lowers the total potential energy of the system:

$$\Pi[s, x_i] = \int_{\Omega} W_{\text{es}}(E, s) + W_{\text{d}}(s) + W_{\text{i}}(\nabla s)]dV \tag{5}$$

where $\Pi[s,x_i]$ is the total potential energy.

In the total potential energy expression (5), $W_{\text{es}}(E,s)$ is the complementary electrostatic potential energy per unit volume which can be expressed as follows:

$$W_{\text{es}}(E, s) = -\frac{\varepsilon}{2} E \times E \tag{6}$$

$W_{\text{d}}(s)$ is the damage energy which can be expressed as follows:

$$W_{\text{d}}(s) = W_{\text{c}}[1 - f(s)] \tag{7}$$

where W_{c} is the critical density of electrostatic energy.

$W_i(\nabla s)$ is a gradient energy term used to adjust the sharp phase boundaries. The gradient energy term can be expressed as follows:

$$W_{\text{i}}(\nabla s) = \frac{\Gamma}{4} \nabla s \times \nabla s \tag{8}$$

where Γ can be approximated as breakdown energy.

By assuming the linear dynamic equation:

$$\frac{\partial s}{\partial t} = -m \frac{\delta \prod}{\delta s} \tag{9}$$

where m is the mobility of the breakdown propagation. The final normalized governing

equations are written in dimensionless form as:

$$\overline{\nabla}\left[\frac{1}{f(s)+\eta}\overline{\nabla}\overline{\phi}\right]=0 \tag{10}$$

$$\frac{\partial s}{\partial t}=-\frac{f'(s)}{2\left[f(s)+\eta\right]^2}\overline{\nabla}\overline{\phi}\overline{\nabla}\overline{\phi}+f'(s)+\frac{1}{2}\overline{\nabla}^2 s \tag{11}$$

where the corresponding dimensionless quantities are symbolized with overbars.

Temperature distribution simulation:

To investigate the temperature distribution in dielectric film-based capacitors under high-temperature conditions, we conducted finite element simulations. The thermal behavior of both FPE and FPE/Al_2O_3 NPLs films was modeled using the following governing equations:[4]

$$\rho C\frac{\partial T}{\partial t}=k\nabla^2 T+Q \tag{12}$$

where ρ, C, T, t, k, and Q represent the density, specific heat capacity, temperature, time, thermal conductivity, and thermal power density, respectively. Q was estimated from the energy loss calculated based on the P–E loop with the equation $Q = f \times U_{loss}$. The energy loss (U_{loss}) was determined from the P–E loop measurements of the films under an electric field of 200 MV m^{-1}, while f represents the operational frequency. The capacitor was modeled as a cylindrical structure with both height and diameter of 20 mm. The simulation employed a convective heat flux boundary condition with an ambient temperature of 423.15 K and a heat transfer coefficient of 200 W m^{-2} K^{-1}.

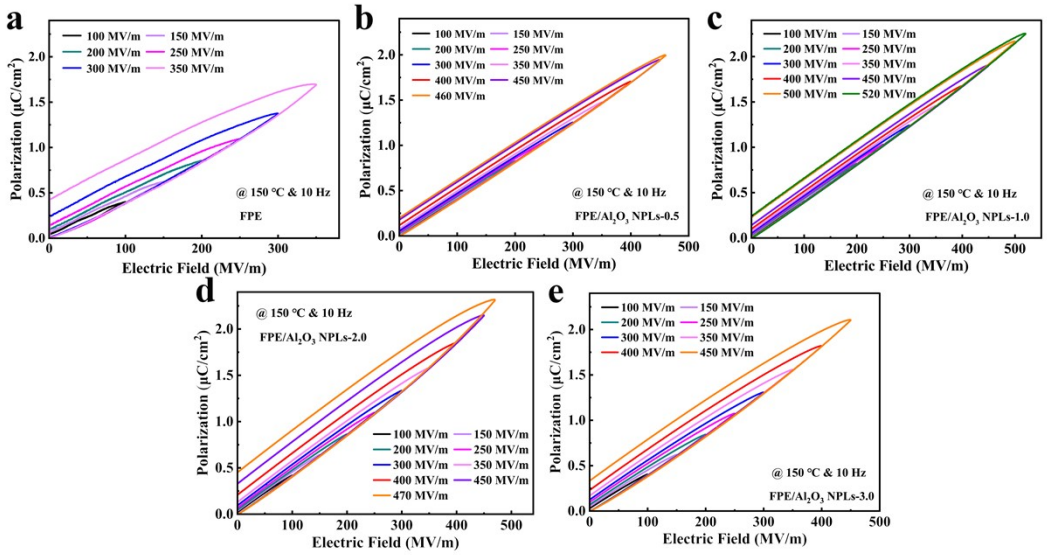

Fig. S3 *P-E* loops of (a) FPE and (b, c, d, e) FPE nanocomposite with 0.5, 1.0, 2.0, 3.0 wt% Al_2O_3-NPLs at 150 ℃ and 10 Hz.

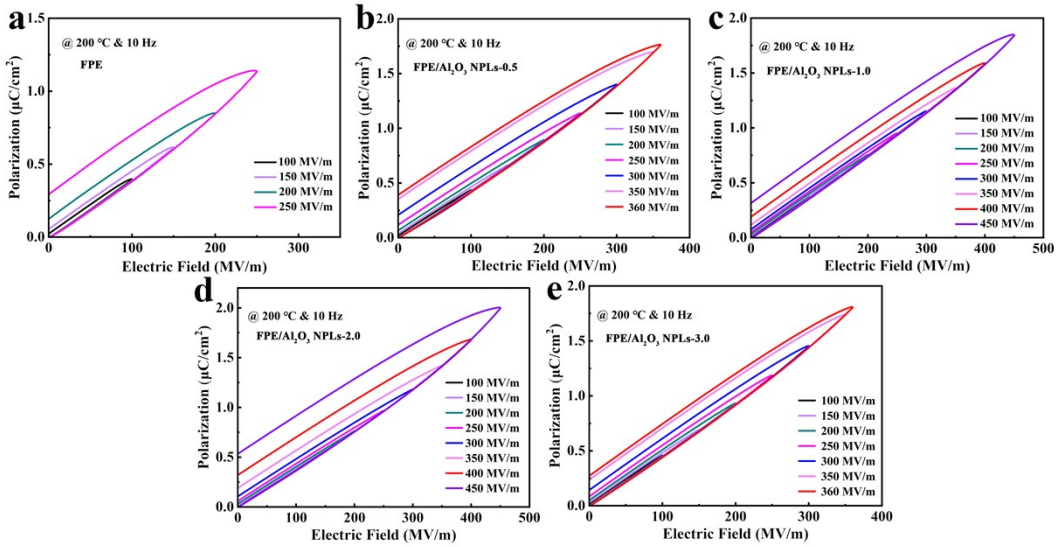

Fig. S4 *P-E* loops of (a) FPE and (b, c, d, e) FPE nanocomposite with 0.5, 1.0, 2.0, 3.0 wt% Al_2O_3-NPLs at 200 ℃ and 10 Hz.

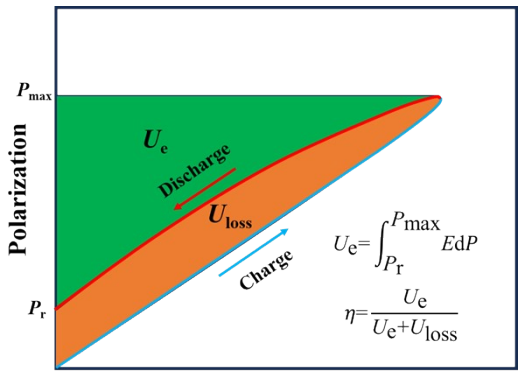

Fig. S5 Schematic polarization-electric field (*P*-*E*) loop of a dielectric material with the discharged energy density (U_e) represented by the area colored in green and energy loss (U_{loss}) indicated by the area painted orange. The total stored energy density (U_s) equals U_e plus U_{loss}, and the charge-discharge efficiency (η) is calculated by $\eta = U_e/U_s \times 100\%$.

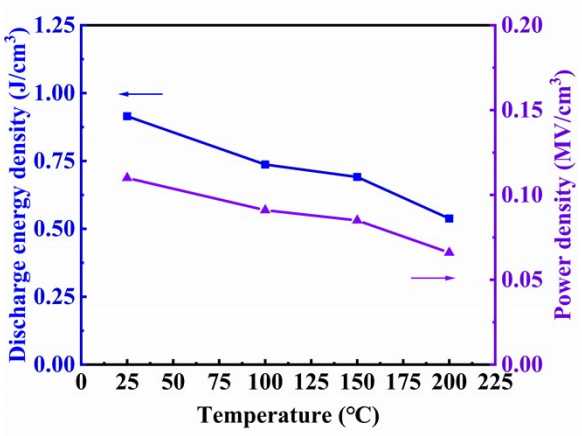

Fig. S6 Discharge energy density and power density as a function of temperature.

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2. M. Yang, H. Li, J. Wang, W. Shi, Q. Zhang, H. Xing, W. Ren, B. Sun, M. Guo, E. Xu, N. Sun, L. Zhou, Y. Xiao, M. Zhang, Z. Li, J. Pan, J. Jiang, Z. Shen, X. Li, L. Gu, C.-W. Nan, X. Wang and Y. Shen, *Nat. Energy*, 2024, **9**, 143-153.
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4. M. Yang, Z. Wang, Y. Zhao, Z. Liu, H. Pang and Z.-M. Dang, *Adv. Mater.*, 2024, **36**, 2309640.