Supplementary information

Significantly increased energy density and discharge efficiency at high temperature in polyetherimide nanocomposite by a small amount of Al₂O₃ nanoparticles

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Fig. S1(a) presents the X-ray diffraction (XRD) patterns of the AO-nps, and all of the deflection peaks of the product are well indexed by the standard data of facecentered cubic γ -Al₂O₃ (PDF#29-0063), indicating the high purity of the nanoparticles.



Fig. S1. (a) X-ray diffraction (XRD) patterns of the nanostructured Al_2O_3 fillers. (b) TEM image of Al_2O_3 particles.



(g)



Fig. S2. Cross-sectional SEM images of (a) pure PEI film, (b) PEI nanocomposites with 0.5 vol% AO-nps, (c) PEI nanocomposites with 1 vol% AO-nps, (d) PEI nanocomposites with 2 vol% AO-nps, (e) PEI nanocomposites with 3 vol% AO-nps, (f) PEI nanocomposites with 5 vol% AO-nps, and (g) PEI nanocomposites with 7 vol% AO-nps.



Fig. S3. (a) DSC and (b) TGA curves of pristine PEI and the PEI/AO-nps nanocomposite films.



Fig. S4. Dielectric spectra of nanocomposites with varied frequency and temperature, (a) PEI, (b) 0.5vol%, (c) 1vol%, (d) 2 vol%, (e) 3 vol%, (f) 5 vol%, and (g) 7vol%. (h) Frequency-dependent dielectric properties of the PEI and PEI composite with 1vol% AO-nps at 150 °C.

| 成分 | 25 °C | | 50 °C | | 100 °C | | 150 ℃ | |
|----------|--------------------------|------|--------------------------|------|--------------------------|------|--------------------------|------|
| | E _b (MV/m) | β |
| PEI | 558.1 | 16.9 | 558.1 | 14.5 | 500.3 | 13.0 | 421.8 | 6.5 |
| 0.5 vol% | 567.7 | 15.7 | 562.6 | 17.3 | 516.0 | 13.8 | 445.3 | 8.0 |
| 1 vol% | 581.7 | 19.4 | 575.9 | 18.5 | 548.2 | 16.6 | 503.9 | 11.1 |
| 2 vol% | 571.3 | 20.1 | 566.1 | 20.1 | 527.2 | 19.3 | 456.8 | 7.4 |
| 3 vol% | 544.9 | 16.5 | 543.4 | 16.7 | 511.1 | 13.2 | 430.8 | 6.4 |
| 5 vol% | 428.7 | 13.0 | | | | | | |
| 7 vol% | 384.3 | 8.6 | | | | | | |

Table S1 Breakdown strength of PEI and nanocomposites with varied content measured at different temperature.



Fig. S5. Young's modulus of the nanocomposites with varied filler content measured at room temperature.



Fig. S6. Electric displacement-electric field (D-E) loops of nanocomposites with varied filler content measured at 25 °C, (a) PEI, (b) 0.5vol%, (c) 1vol%, (d) 2 vol%, (e) 3 vol%, (f) 5 vol%, and (g) 7vol%.



Fig. S7. Electric displacement-electric field (D-E) loops of nanocomposites with varied filler content measured at 50 °C, (a) PEI, (b) 0.5vol%, (c) 1vol%, (d) 2 vol%, and (e) 3 vol%.

Fig. S8. Electric displacement-electric field (D-E) loops of nanocomposites with varied filler content measured at 100 °C, (a) PEI, (b) 0.5vol%, (c) 1vol%, (d) 2 vol%, and (e) 3 vol%.

Fig. S9. Electric displacement-electric field (D-E) loops of nanocomposites with varied filler content measured at 150 °C, (a) PEI, (b) 0.5vol%, (c) 1vol%, (d) 2 vol%, and (e) 3 vol%.

Fig. S10. Maximum electric displacement-electric field (D-E) loops of nanocomposites measured at different temperature, (a) PEI, (b) 0.5vol%, (c) 1vol%, (d) 2 vol%, and (e) 3 vol%.

Fig. S11(a) shows the TSDC spectroscopies of the nanocomposites, which are collected under a poling field (E_p) of 15 MV/m for a poling time of 30 min at poling temperature (T_p) of 150 °C. There is only a peak at 180-200 °C, which is supposed to

be ascribed to the depolarization of the injected charges from electrodes, i.e., Schottky or thermionic emission. As shown in **Fig. S11**(b) The E_p -dependent TSDC test was performed for PEI with 1 vol% AO-nps to verify the hypothesis about injection charges. In general, for Schottky emission, the current density (*J*) scales as $\ln J \propto E_p^{1/2}$. At equilibrium state, the injected charges equal to the leakage charges (C), which can be obtained from the TSDC spectrum as $\ln C \propto E_p^{1/2}$. **Fig. S11**(c) establishes the linear relationship between $\ln C$ and $E_p^{1/2}$, which supports our prediction. The increase of maximum displacement and decrease of remnant displacement bring about considerable improvement in high-temperature electric energy storage.

Fig. S11. (a) TSDC curves of PEI and PEI/AO-nps under poling electric field of 15 MV/m at 150 °C for 30 min. (b) TSDC spectra for PEI/AO-nps film under different poling electric field at 150 °C for 30 min. (c) The relationship between lnC and $E_p^{1/2}$.

Fig. S12. Discharged energy density and charge-discharge efficiency of PEI/AO-nps nanocomposite films with varied filler contents measured at (a) 25 °C, (b) 50 °C, (c) 100 °C, and (d) 150 °C.

Fig. S13. Discharged energy density and charge-discharge efficiency of PEI/AO-nps nanocomposite films nanocomposites measured at different temperature, (a) PEI, (b) 0.5 vol%, (c) 1 vol%, (d) 2 vol%, and (e) 3 vol%.