Supplementary Information for

Scalable all polymer dielectrics with self-assembled nanoscale multiboundary exhibiting superior high temperature capacitive performance

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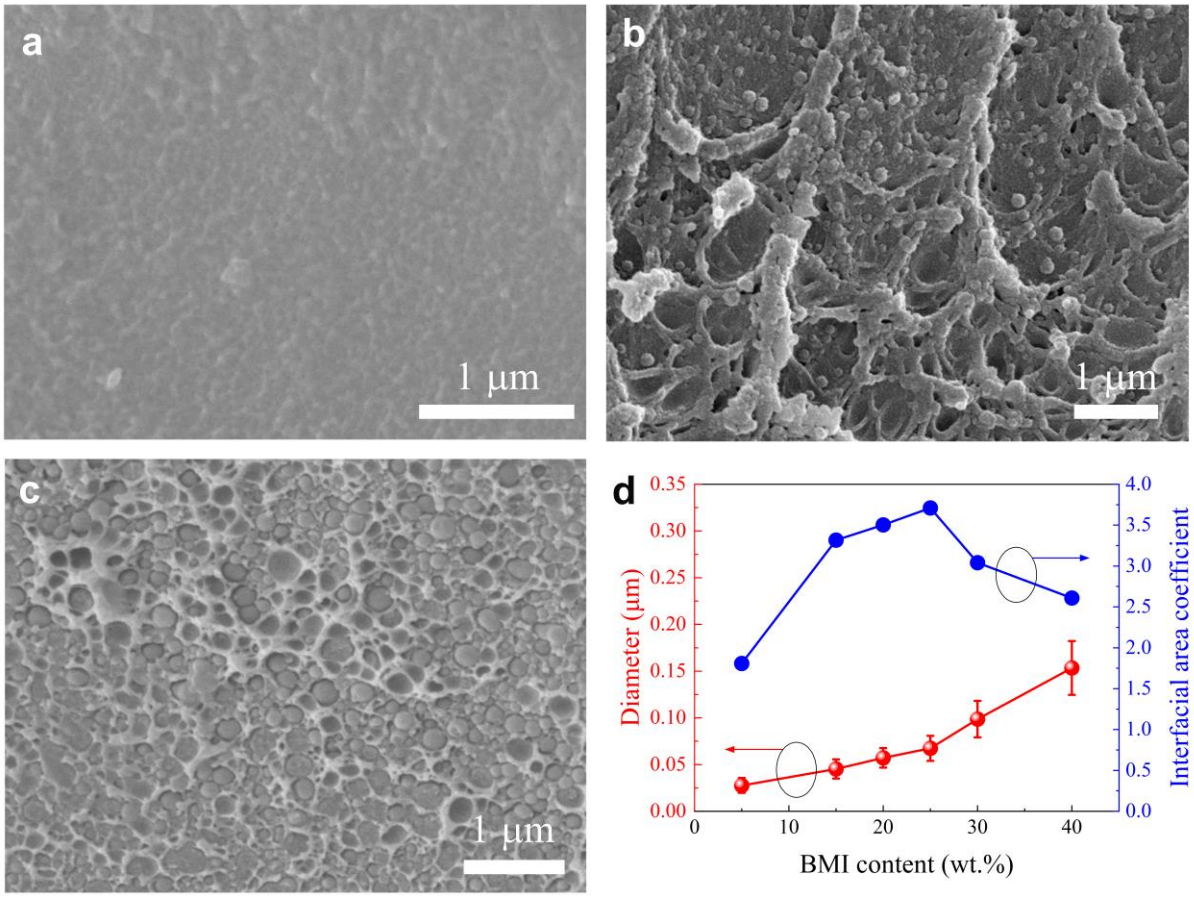

Figure S1. Scanning electron microscope (SEM) images of (a) Pristine PEI, (b) PEI/BMI 85/15, (c) PEI/BMI 60/40, and (d) Variation in average particle size of BMI domains and interfacial area coefficient with BMI content in PEI/BMI PNDs, with error bars representing standard deviation.

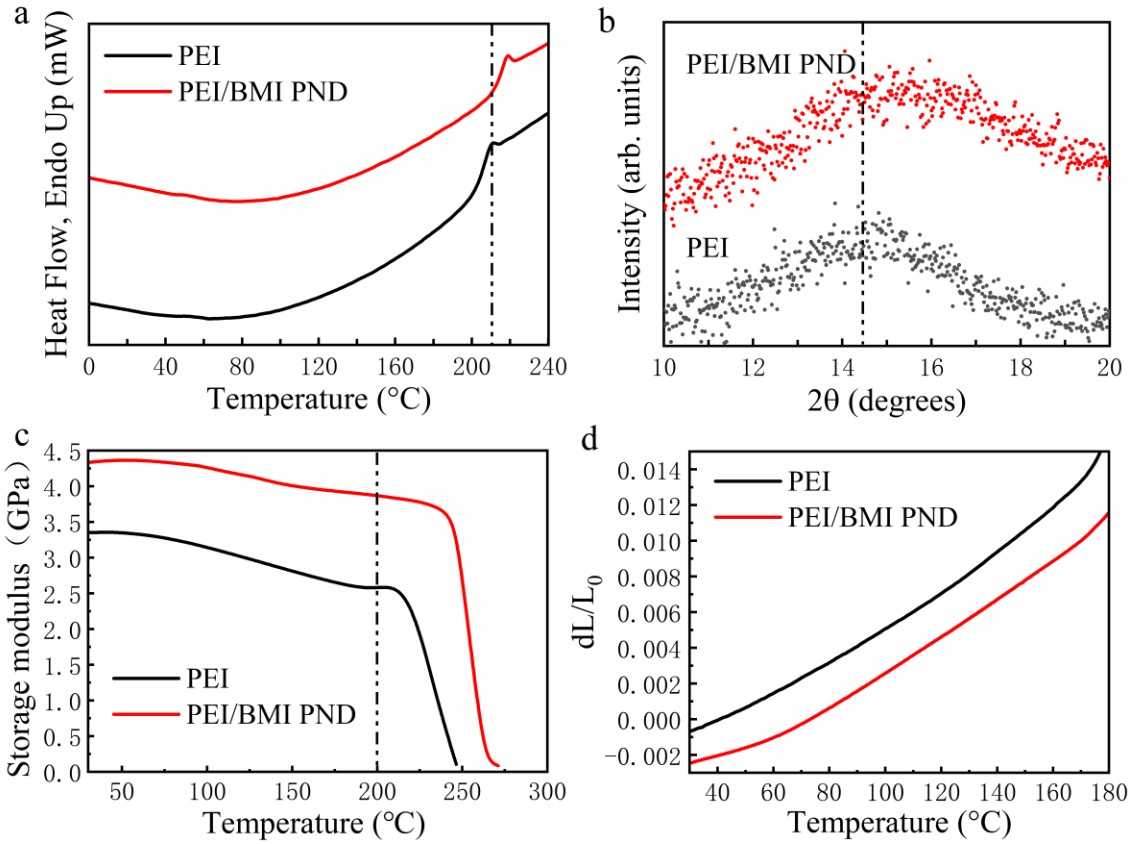

Figure S2. Structural characterization of PND films. (a) Differential scanning calorimetry (DSC) heating curves, (b) wide-angle X-ray diffraction data, (c) storage modulus as a function of temperature, and (d) typical TMA curves of pristine PEI and PEI/BMI 75/25 based PND films.

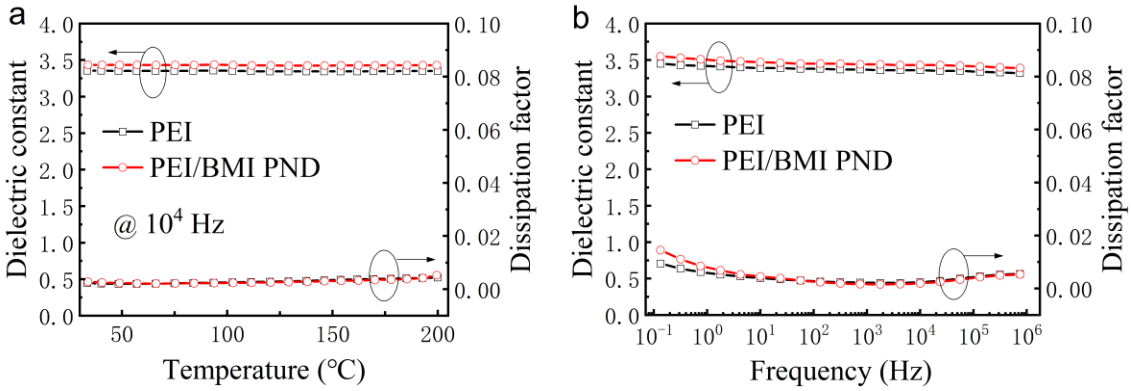

Figure S3. Dielectric properties of PND films. (a) Temperature dependence and (b)frequency dependence of dielectric constant and dissipation factor of PEI/BMI 75/25 based PND and compared with pristine PEI.

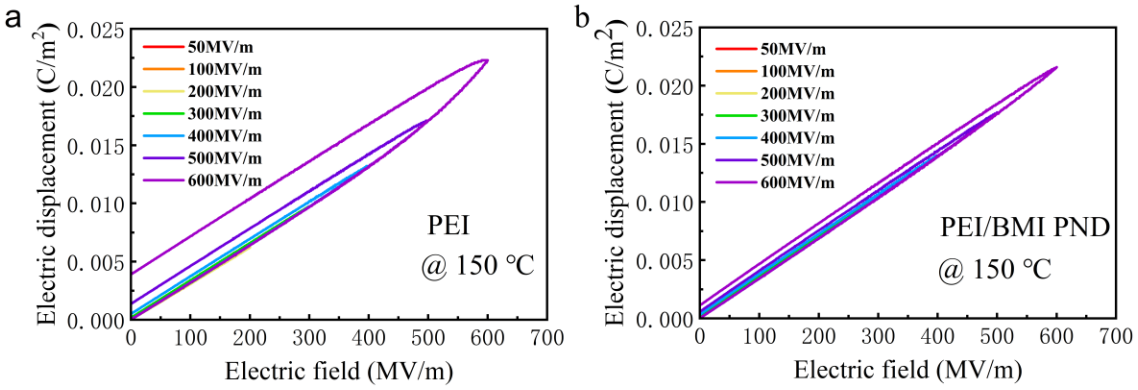

Figure S4. P-E loops of pristine PEI (a) and PEI/BMI 75/25 based PND (b) at 150 °C.

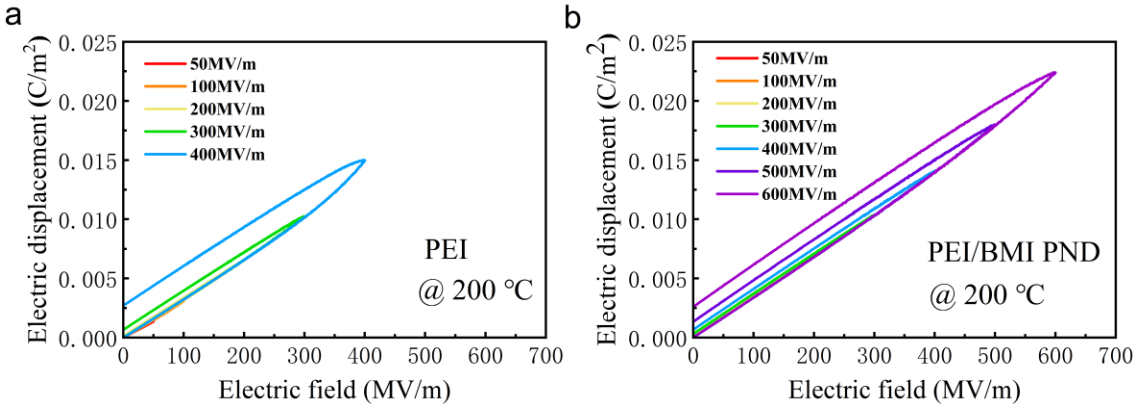

Figure S5. P-E loops of pristine PEI (a) and PEI/BMI 75/25 based PND (b) at 200 °C.

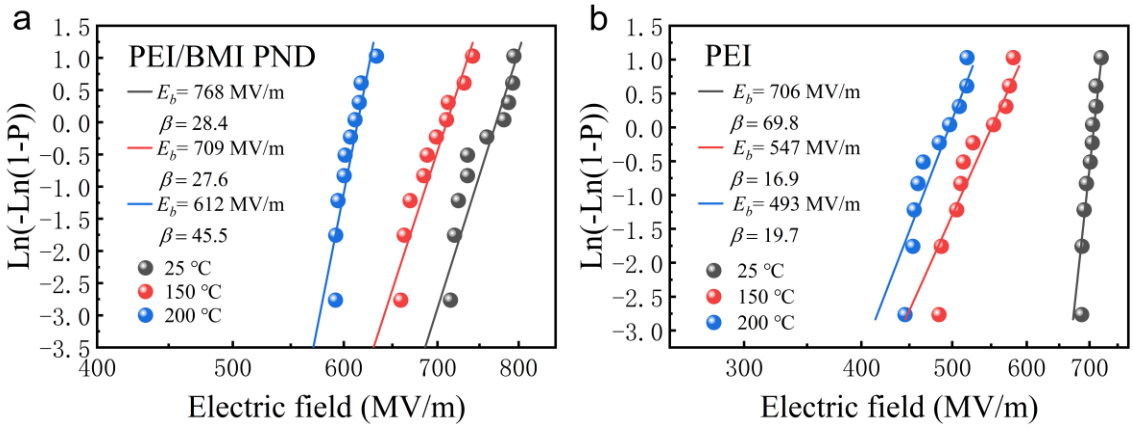

Figure S6. Two-parameter Weibull distribution analysis of the characteristic breakdown strength of PEI/BMI 75/25 based PND (a) and pristine PEI (b) measured at 25 °C, 150 °C and 200 °C.

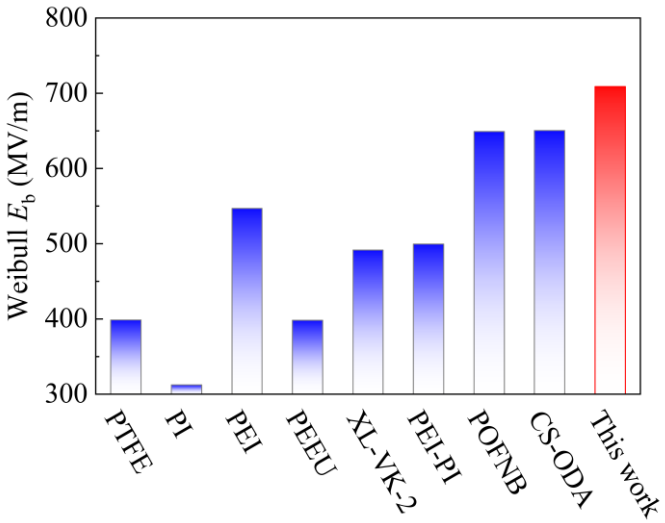

Figure S7. Comparison of the Weibull breakdown strength between PND and the current high-temperature neat dielectric polymers at 150 °C.

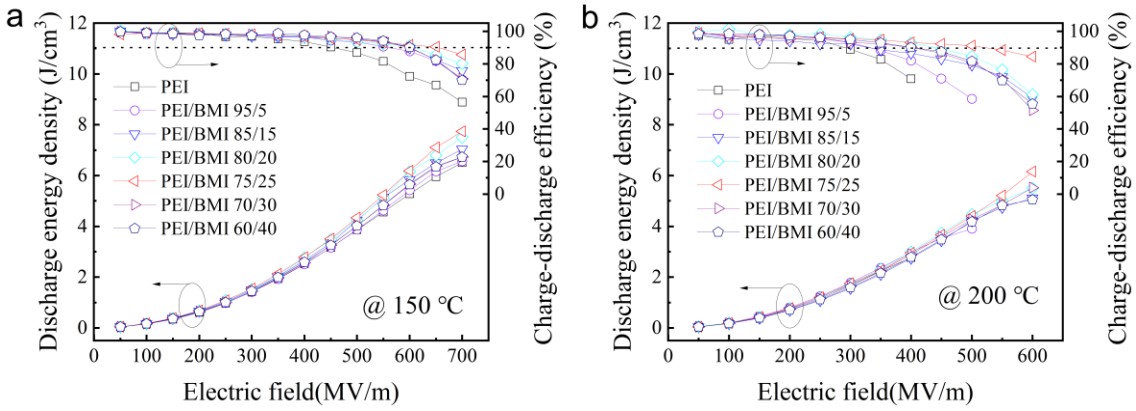

Figure S8. Discharged energy density and charge–discharge efficiency of PEI/BMI-based PNDs with different content of BMI at 150 °C (a) and 200 °C (b).

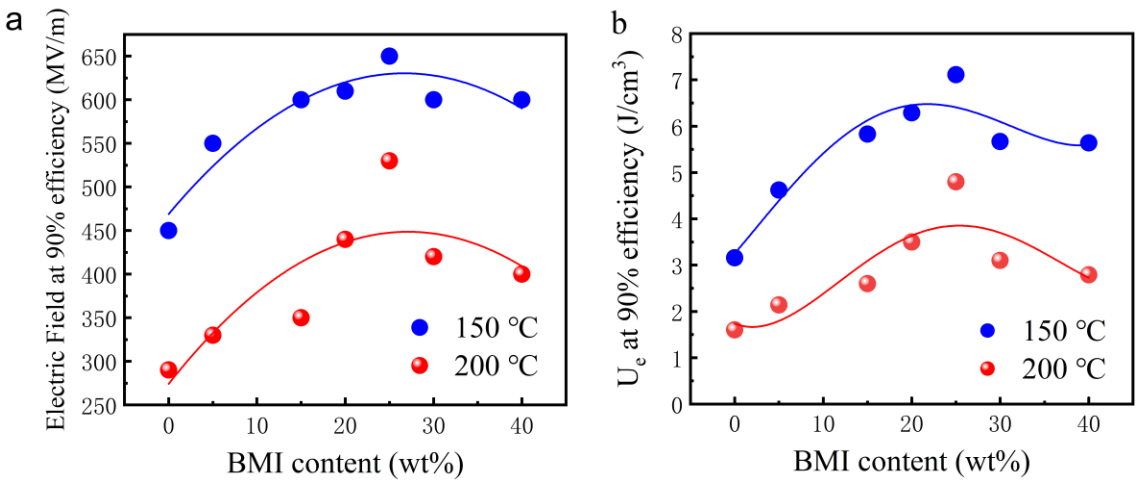

Figure S9. (a) Electric field and (b) discharged energy density at 90% charge–discharge efficiency of PEI/BMI-based PNDs with different content of BMI.

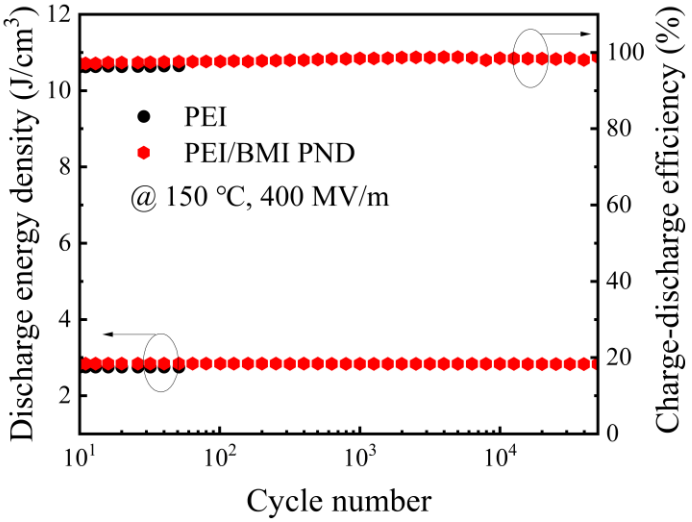

Figure S10. Cyclic performance of pristine PEI and PEI/BMI 75/25 based PNDs at 150 ℃ and 400 MV/m.

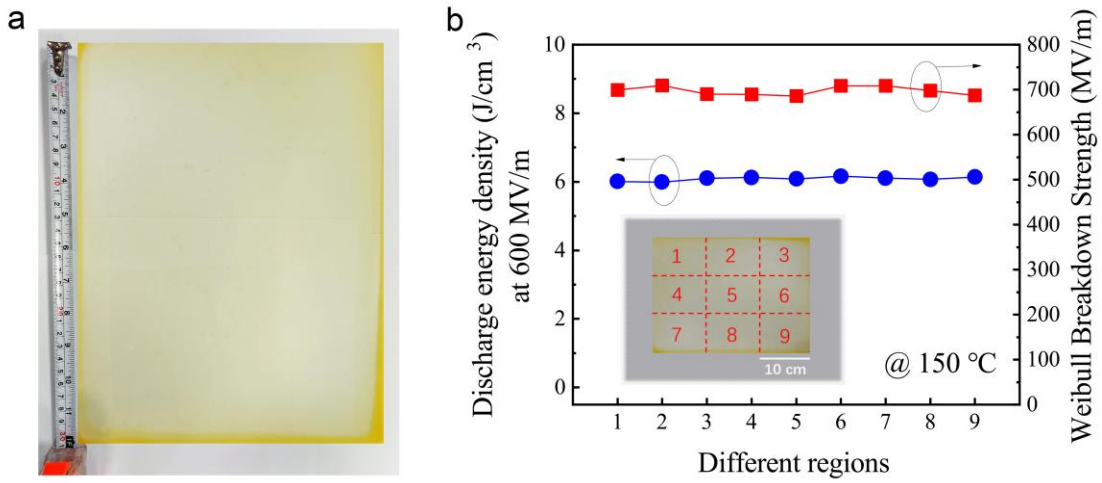

Figure S11. (a) Photograph of a PND film with composition of PEI/BMI 75/25 and an area of 20 × 30 cm^2. (b) Discharged energy density and Weibull breakdown strength tested at different regions (as marked in the inset photograph) of the PND film in (a) at 150 °C.

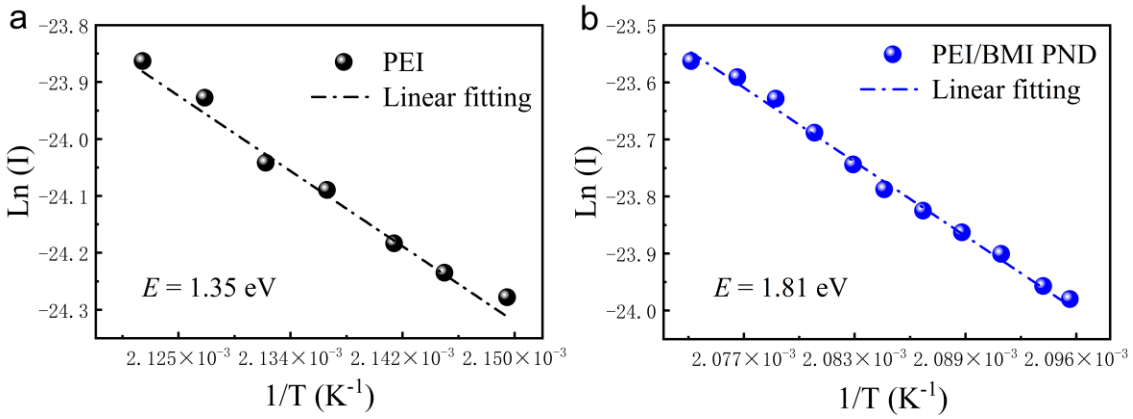

Figure S12. Fitting curves of TSDC data. (a) Pristine PEI and (b) the PND with composition of PEI/BMI 75/25.

Table S1. The peak temperature and trap level of PEI, PEI/BMI 75/25 in the TSDC measurement.

Dielectric	T_{peak} (°C)	Trap level (eV)
PEI	207.2	1.35
PEI/BMI	217.5	1.81