Polymer Dielectric Films Exhibiting Superior High-Temperature Capacitive Performance by Utilizing an Inorganic Insulation Interlayer

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Finite-element simulations

In order to qualitatively understand the charge carrier behaviors in nanocomposites, a unipolar electron injection and transport model is established. Here, we describe electrons injected from the metal electrode into the dielectric by Schottky thermionic emission, which is expressed as

$$J_e = AT^2 \exp\left(-\frac{\varphi_i}{k_B T}\right) \exp\left(\frac{e}{k_B T}\sqrt{\frac{eE}{4\pi\varepsilon_0\varepsilon_r}}\right) \quad \text{(S1)}$$

where J is the current density, A is the Richardson constant, T is the temperature, φ_i is the injection potential barrier, k_B is the Boltzmann constant, e is the elementary charge, E is the electric field, ε_0 is the vacuum permittivity, ε_r is the relative permittivity, respectively.

The behavior of charge within the films is governed by the following equations:

Poisson's equation,

$$\frac{\partial \psi(r,t)}{\partial r_i \partial r_j} = \frac{\rho(r,t)}{\varepsilon_0 \varepsilon_r} \quad (2)$$

Current continuity equation,

$$\frac{\partial n(r,t)}{\partial t} + \frac{\partial J(r,t)}{\partial r} = s \quad (3)$$

Charge transport equation,

$$J(\mathrm{r},\mathrm{t}) = \mu(\mathrm{r},\mathrm{t})n(\mathrm{r},\mathrm{t})E(\mathrm{r},\mathrm{t}) \quad (4)$$

where ψ the electrical potential, ρ the total charge density (here equal n(r,t)), s the source term (s=0 in this simulation), μ the mobility of charge.

Table 1

Definition of parameters used in this simulation.

Parameter	Values	Units
Charge mobility of PEI	1×10^{-10}	$cm^2/(V \cdot s)$
Charge mobility of MgO	1×10^{-13}	$cm^2/(V \cdot s)$
Dielectric constant of PEI	3.15	---
Dielectric constant of MgO	9.8	---
Electric field	100	MV/m
Temperature	423	K
Schottky barrier	1.55	eV

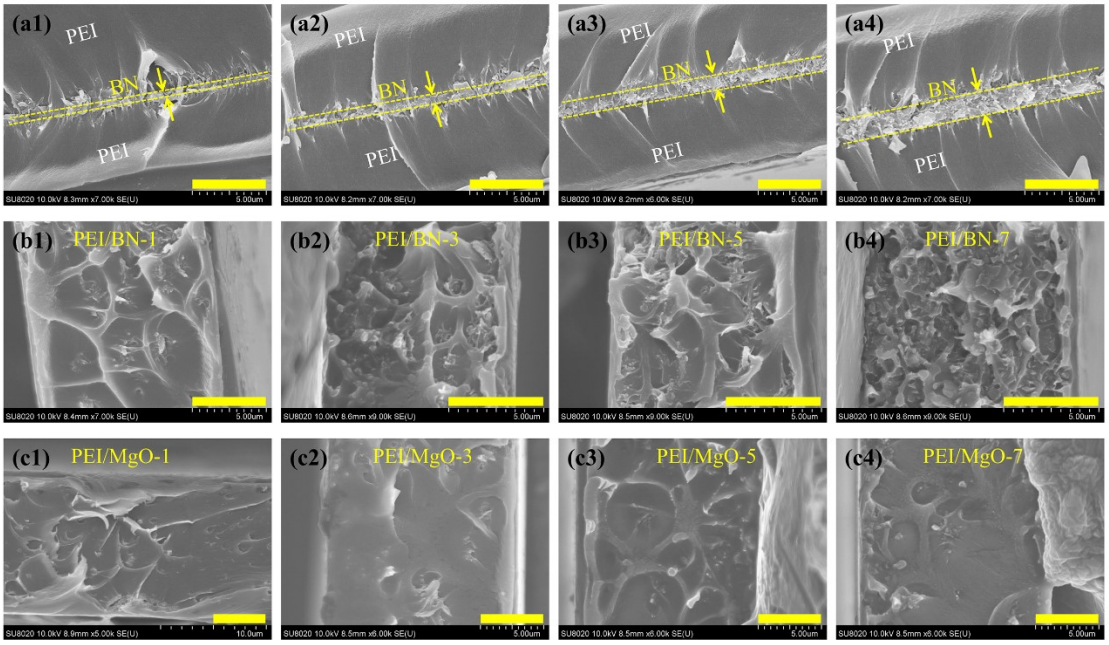

Figure S1 Cross-sectional structure of SEM images of (a) PBP films. (b) PEI/BN films. (c) PEI/MgO films.

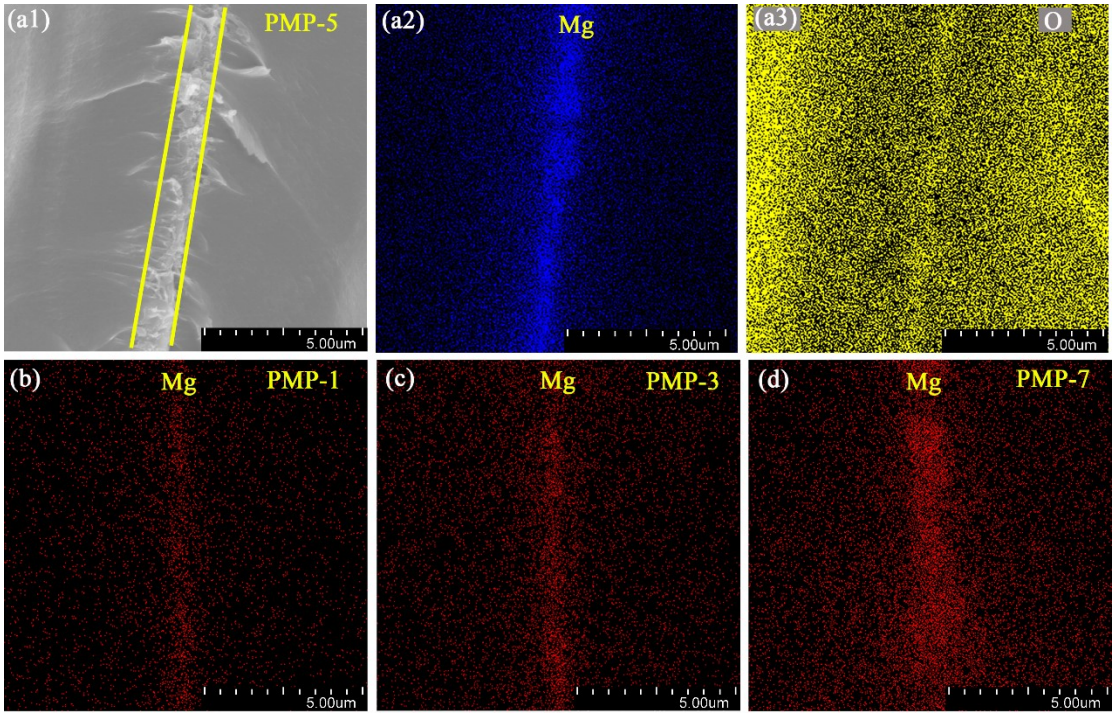

Figure S2 SEM-EDX mapping of PMP films.

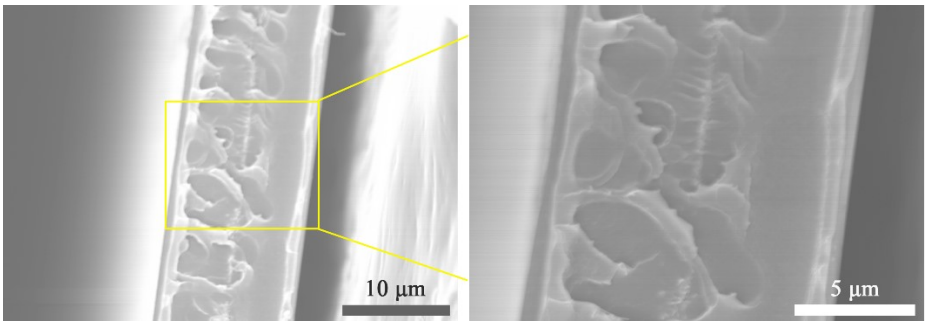

Figure S3 Representative low-magnification of SEM image of PMP films.

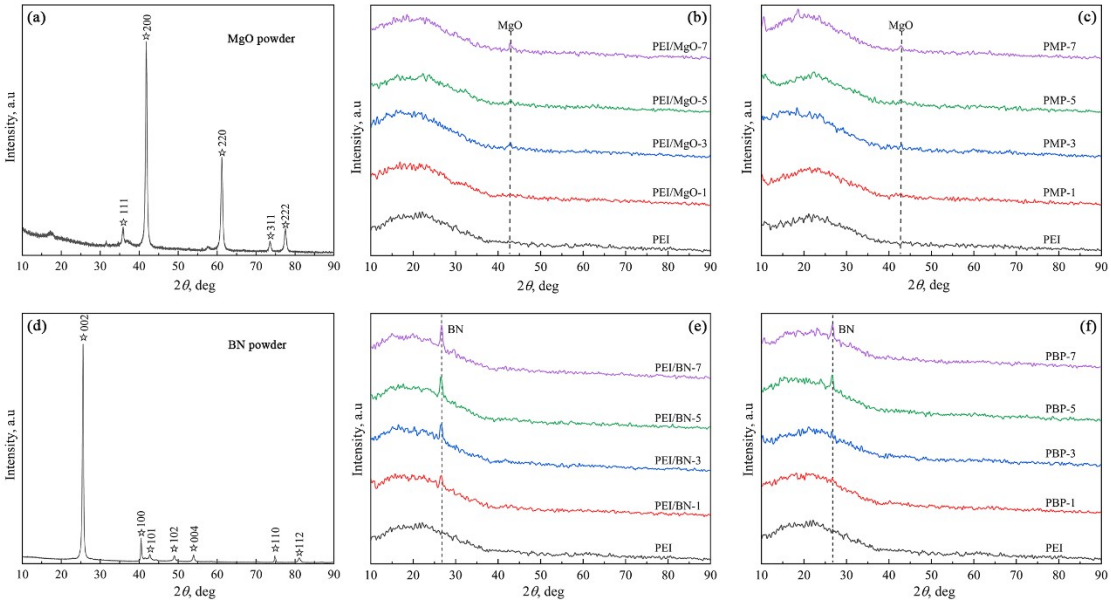

Figure S4 XRD patterns of (a) MgO powder. (b) PEI/MgO films. (c) PMP films. (d) BN powder. (e) PEI/BN films. (f) PBP films.

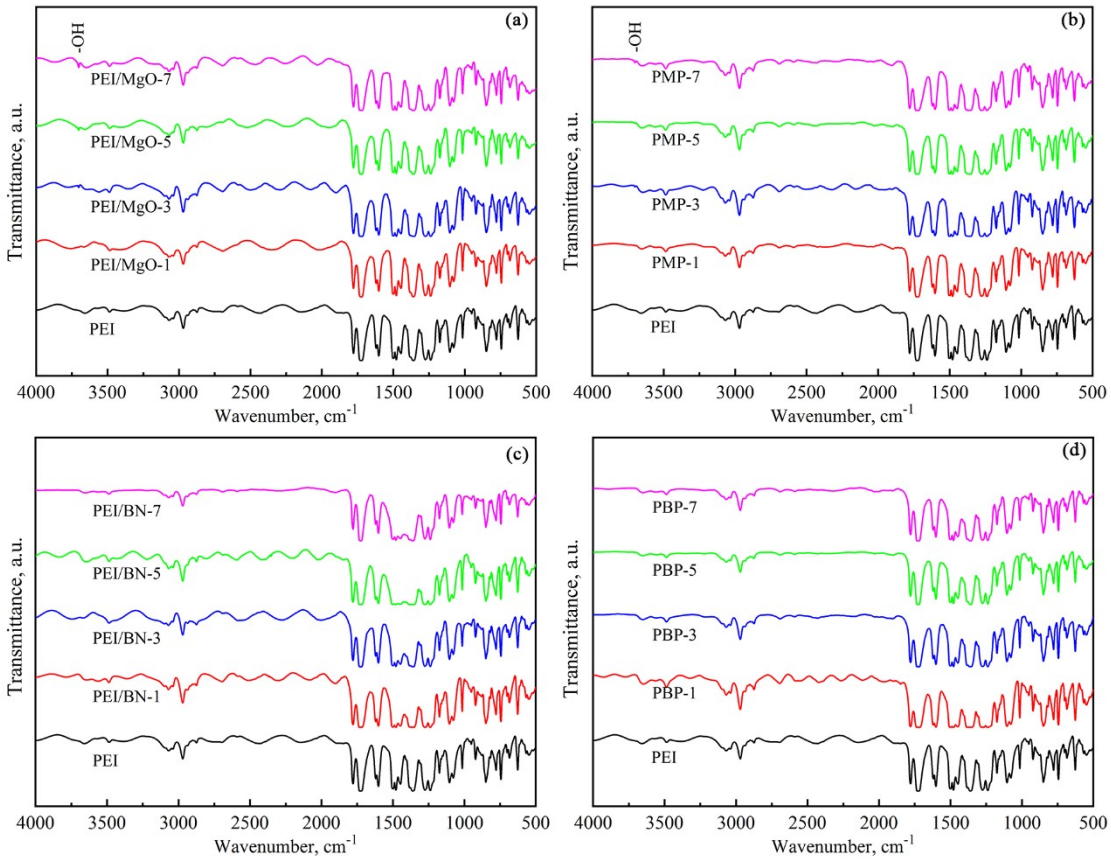

Figure S5 FTIR spectra of (a) PEI/MgO films. (b) PMP films. (c) PEI/BN films. (d) PBP films.

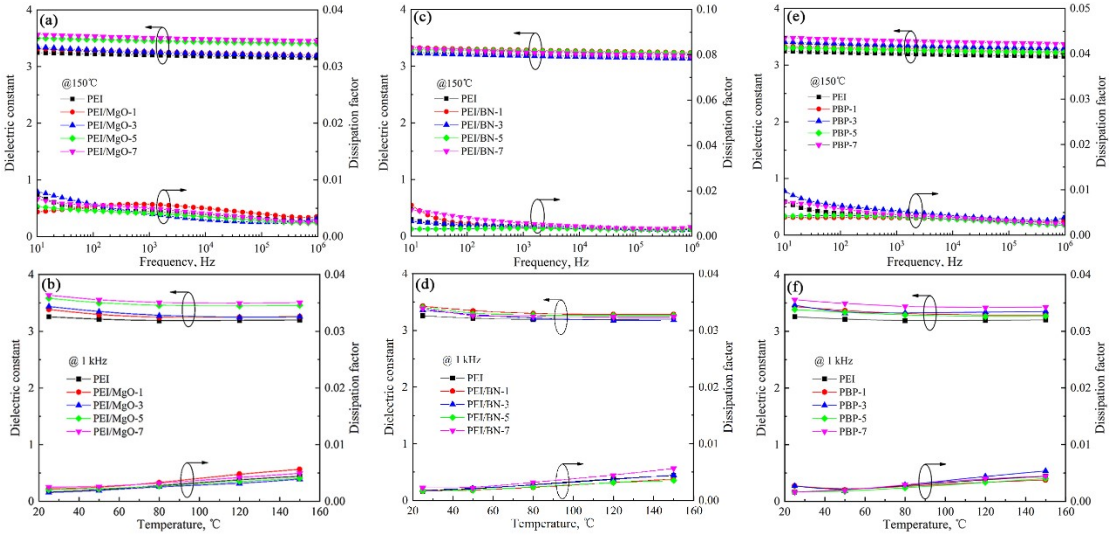

Figure S6 Frequency-dependent and temperature-dependent dielectric properties measured at 150°C and 1 kHz, respectively. (a) (b) PEI/MgO films. (c) (d) PEI/BN films. (e) (f) PBP films.

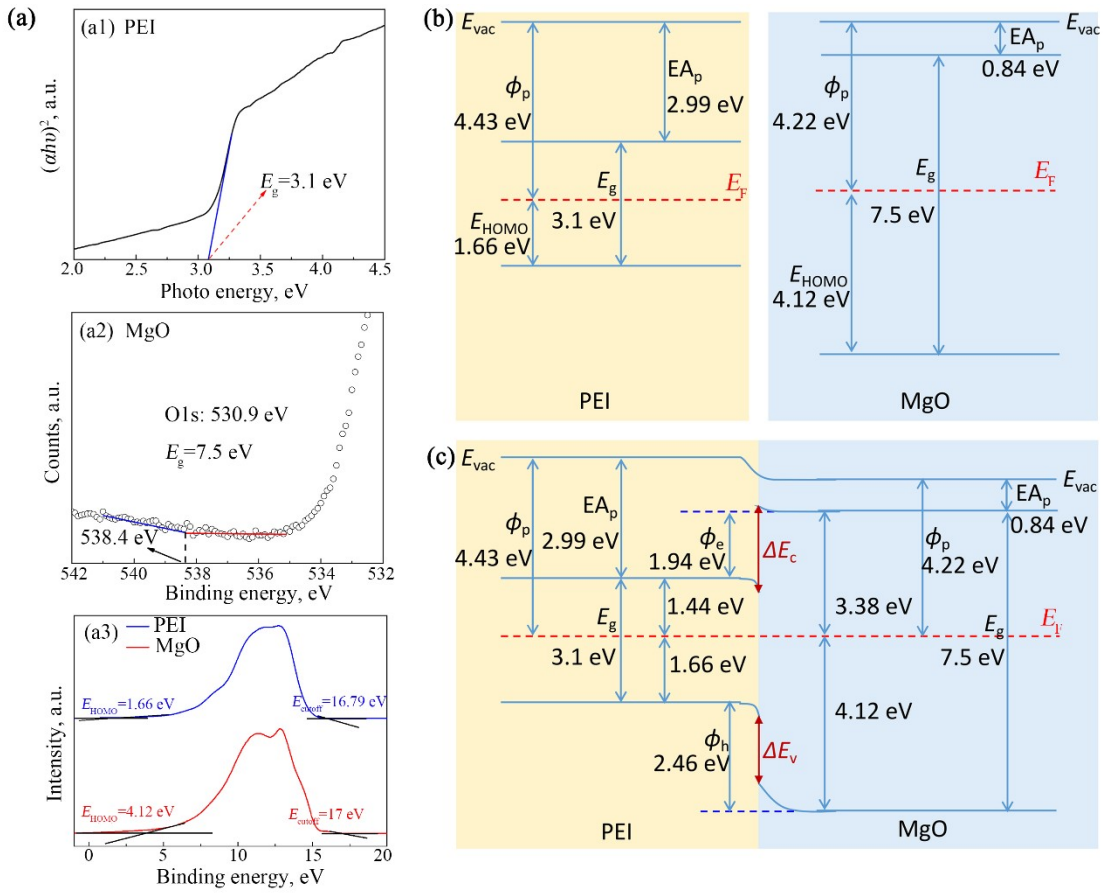

Figure S7 Electronic structure of (a1) PEI, (a2) MgO, (a3) PEI and MgO. Band diagrams at PEI/MgO interface. (b) PEI and MgO interlayer before contact. (c) PEI and MgO interlayer after contact. Herein, E_g is bandgap, E_F is Fermi levels, E_{HOMO} stands for the highest occupied molecular orbital of the dielectric, E_{cutoff} represents the energy of secondary electron cutoff. ϕ_p is the work function, EA_p is the electron affinity, E_{vac} is the vacuum energy level. ϕ_e is the potential barrier for electrons, and ϕ_h is the potential barrier for holes.

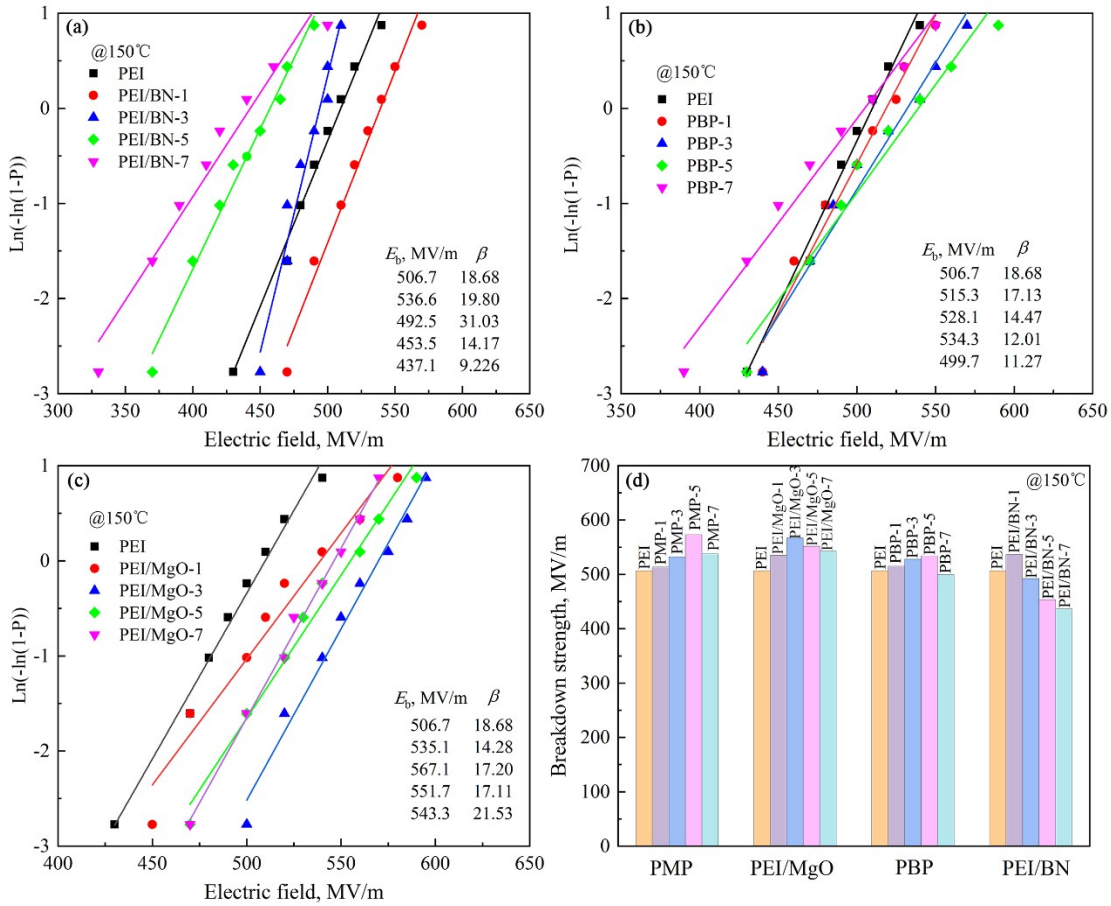

Figure S8 Weibull distribution of (a) PEI/BN films. (b) PBP films. (c) PEI/MgO films measured at 150°C. (d) Comparison of breakdown strength of resultant films in this work measured at 150°C.

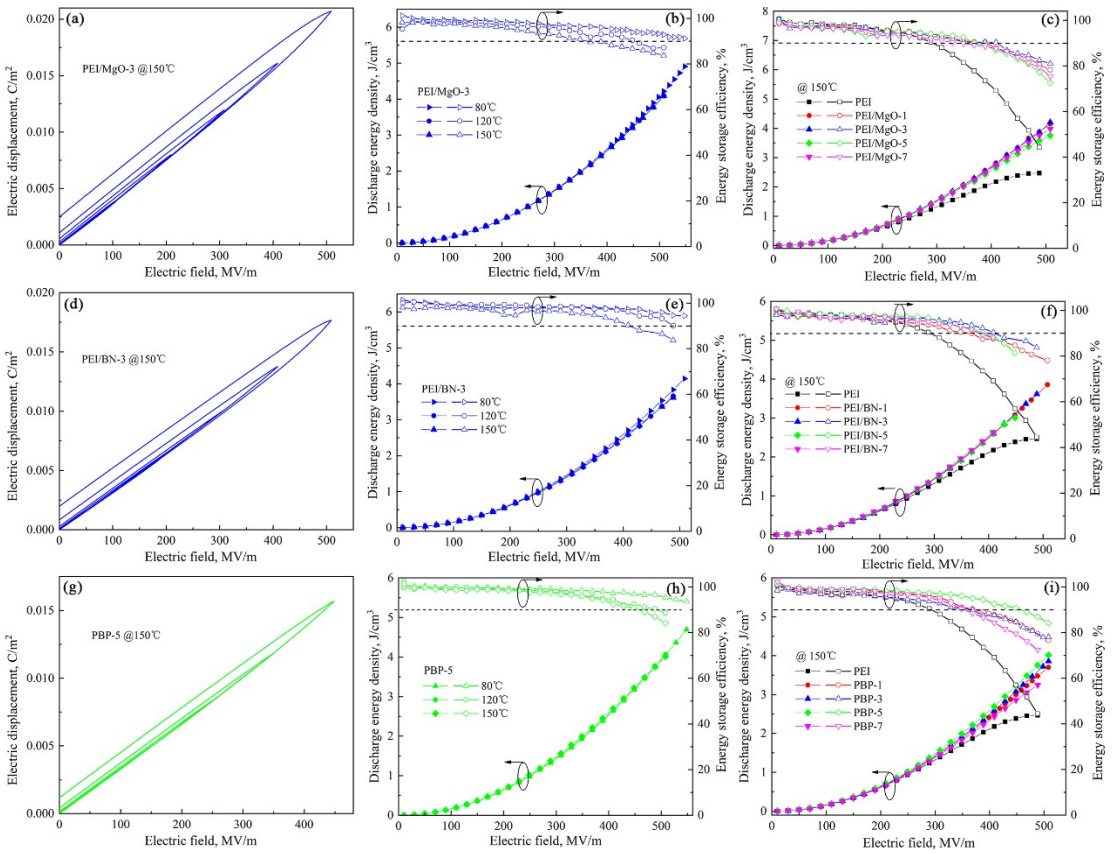

Figure S9 Hysteresis loops measured at 150°C of (a) PEI/MgO-3 film. (d) PEI/BN-3 film. (g) PBP-5 film. Discharged energy density and efficiency of (b) PEI/MgO-3 film. (e) PEI/BN-3 film. (h) PBP-5 film at various temperatures. Discharged energy density and efficiency of (c) PEI/MgO films. (f) PEI/BN films. (i) PBP films measured at 150°C.

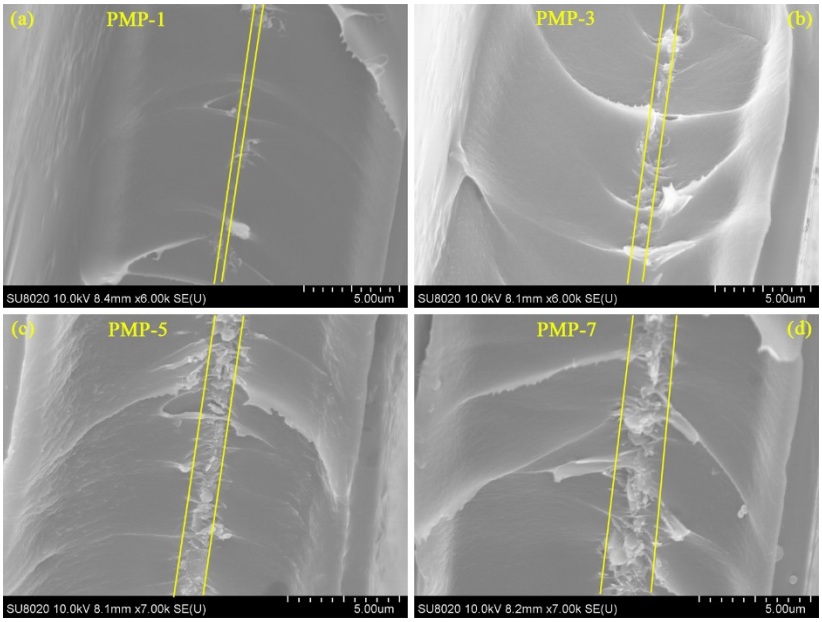

Figure S10 Cross-sectional structure of SEM images of PMP films after 50000 charge/discharge cycles.