Supplementary Information (SI) for Journal of Materials Chemistry C.

Scalable all-polymer dielectric films with aligned structures for high-temperature energy storage applications

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Table. S1 Definition of parameters used in this simulation.

Parameter	Values	Units
Schottky barrier	1.25	eV
Trap depth	0.2	eV
Charge mobility of PEI	1×10^{-12}	$cm^2/(V \cdot s)$
Charge mobility of PVDF	1×10^{-8}	$cm^2/(V \cdot s)$
Electric field	100	MV/m
Temperature	423	K

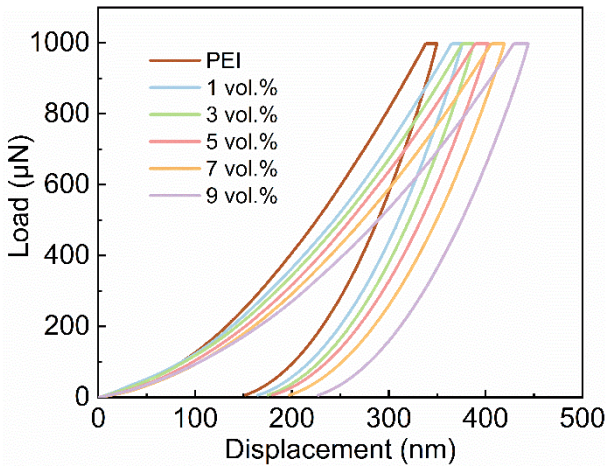

Fig. S1 The load-displacement curves for all-organic composite films with different PEI/PVDF components.

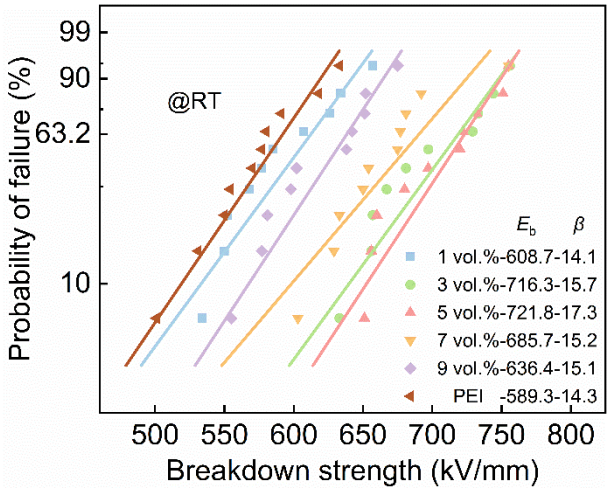

Fig. S2 Weibull breakdown characteristics of all-organic composite films with different PEI/PVDF components at room temperature.

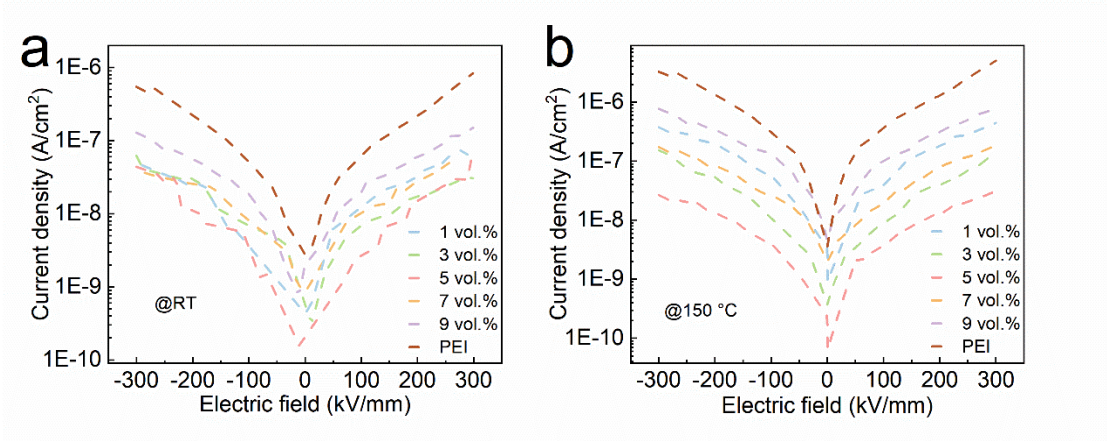

Fig. S3 Variation of leakage current density with the applied electric field at a) room temperature and b) 150 °C.

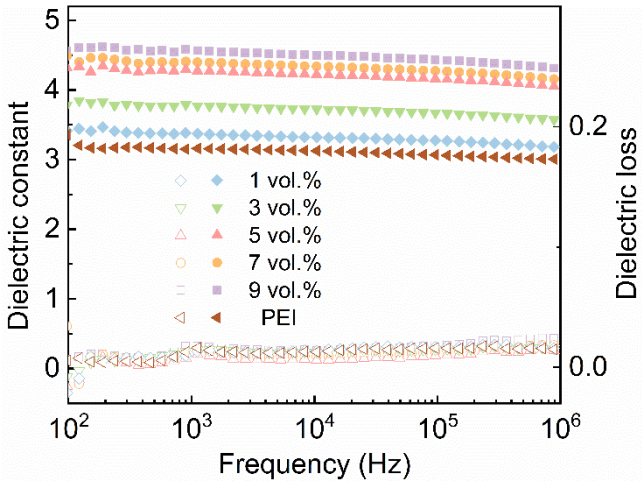

Fig. S4 The dependence of dielectric constant-loss of all-organic composite films with different PEI/PVDF components on frequency.

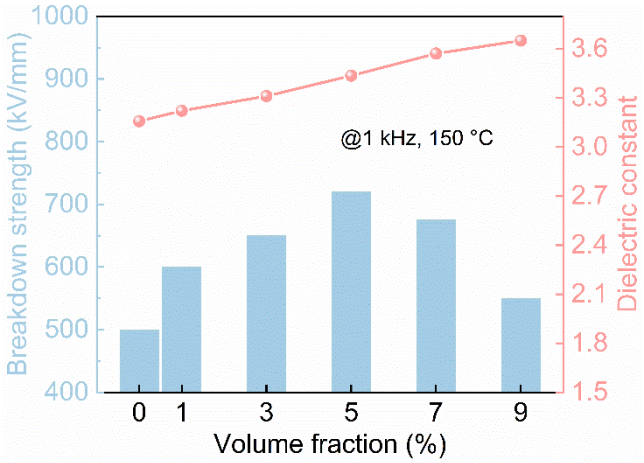

Fig. S5 Variation of dielectric constant and breakdown strength as a function of different PEI/PVDF components at 1 kHz and 150 °C.

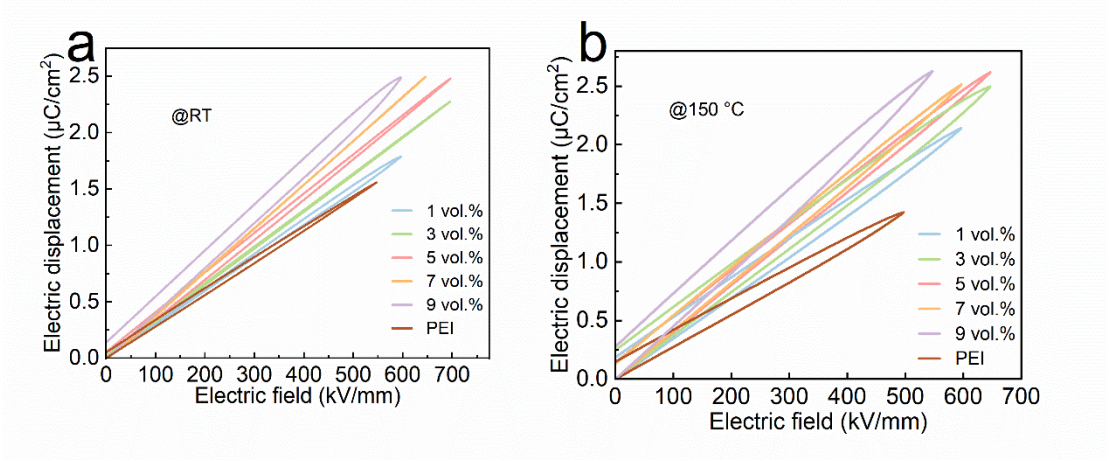

Fig. S6 *D-E* loops of all-organic composite films with different PEI/PVDF components at a) room temperature and b) 150 °C.

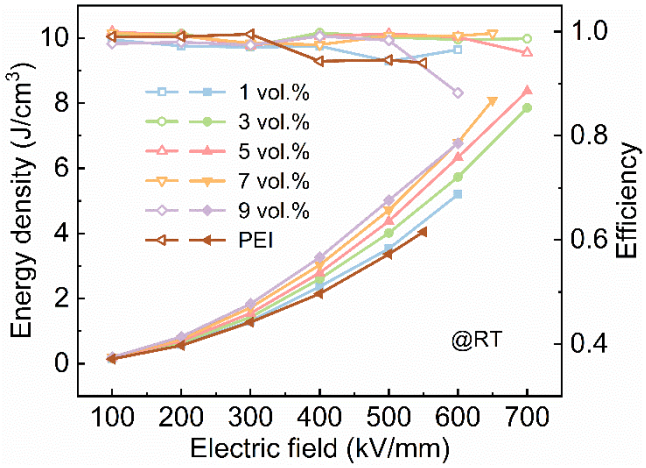

Fig. S7 Variation of energy storage density and charge-discharge efficiency with the applied external electric field at room temperature.

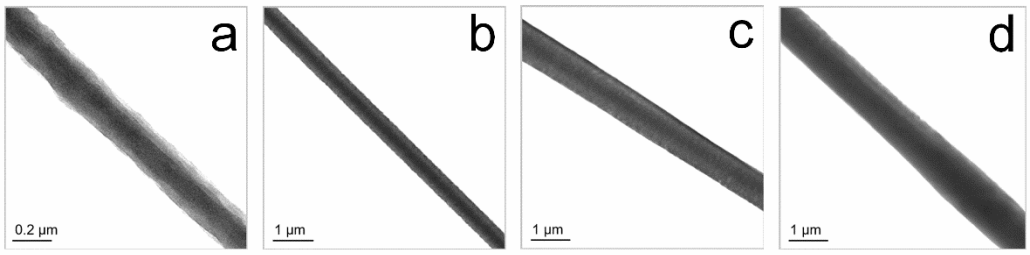

Fig. S8 The SEM images with fibers of diameters a) 200 nm, b) 500 nm, c) 1 μm and d) 1.5 μm

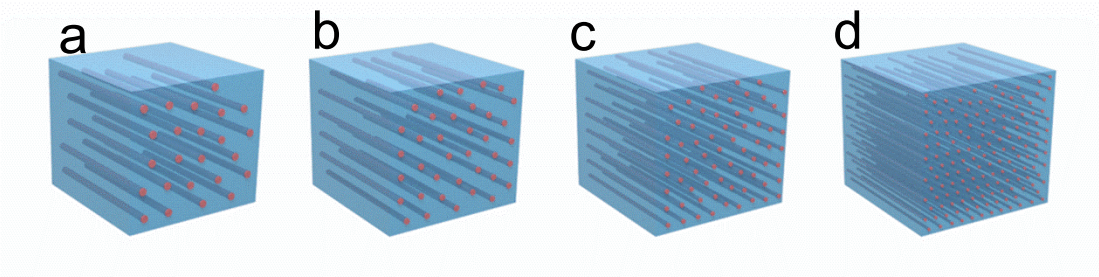

Fig. S9 Schematic illustration of films with different fiber diameters 1.5 μm, b) 1 μm, c) 500 nm and d) 200 nm

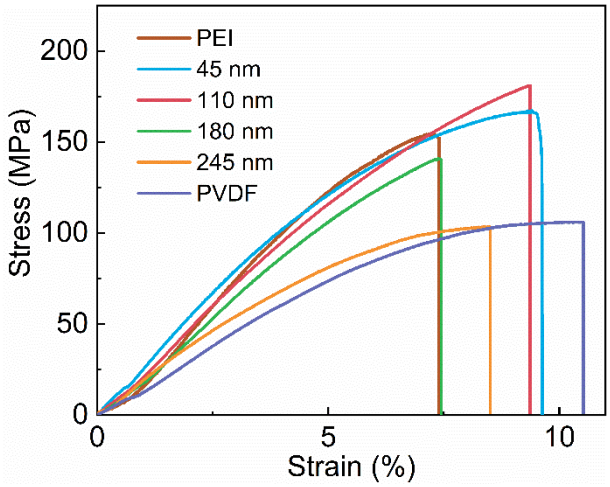

Fig. S10 Stress-strain curves of composite films loaded with different fiber diameters of 5 vol.% PEI/PVDF

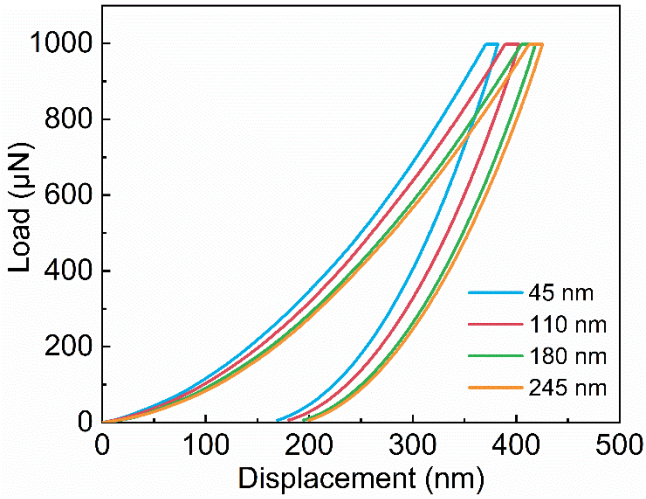

Fig. S11 The load-displacement curves for different fiber diameters of 5 vol.% PEI/PVDF

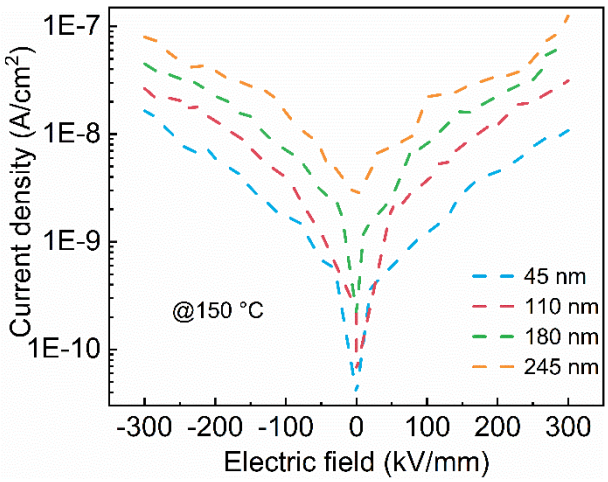

Fig. S12 Variation of leakage current density with the applied electric field at 150 °C

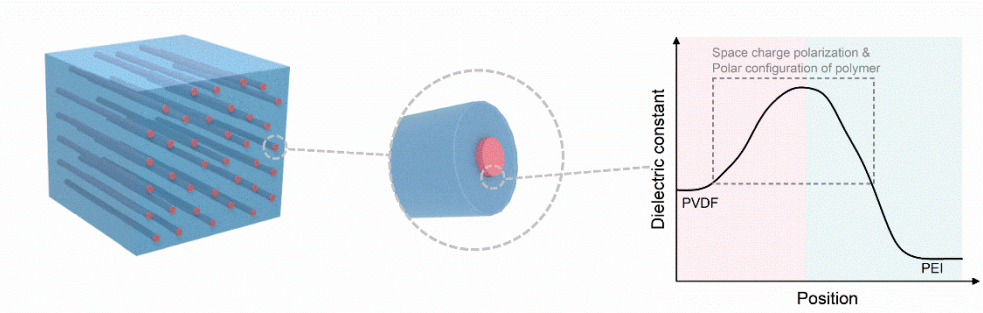

Fig. S13 Schematic illustration of the interfacial polarization mechanism

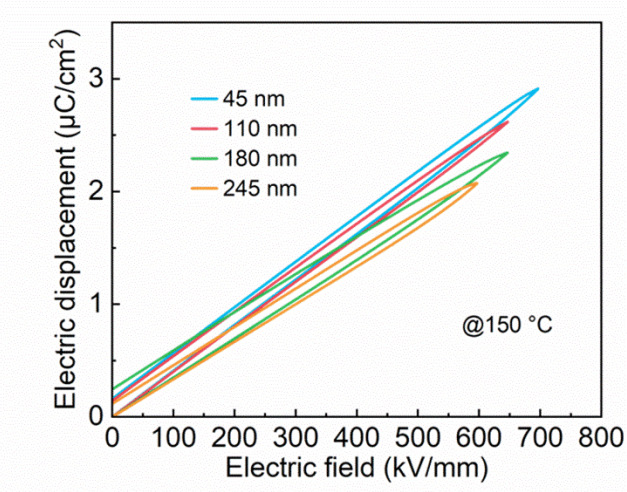

Fig. S14 *D-E* loops of films with different fiber diameters of 5 vol.% PEI/PVDF at 150 °C

Table. S2 The dielectric and energy storage properties at 150 °C of previously published polymer-based dielectrics and the PEI/PVDF composite in this work.

Composites	U_d (J/cm³)	η (%)	E_b (kV/mm)	ε_r
PEI/PVDF (this work)	9.39	>90	711	4.44
PEI/PCBM[1]	4.5	>90	~550	~3.5
POFNB[2]	5.7	~77	~686	2.5
ITIC/PEI[3]	6.37	>90	560	3.2
o-POFNB[4]	8.3	~83	~800	~2.88
CS-ODA[5]	7.02	>90	641.7	3.53
PC-BN-SiO_2[6]	5.22	~76	610	~3.0
PEI-Al_2O_3[7]	3.5	>90	636.4	~3.1
F-PI/PCBM[8]	6.39	>90	815	2.99
PI-PAA/BNNS[9]	7.4	~47	527	4.05
PEEU-Al_2O_3[10]	5	90	600	7.4
NTCDA/PEI[11]	5.1	90	630	~3.2
ITIC-PI@PEI[12]	4	~63	505	~3.27

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