Supporting Information

Enhanced Dielectric Properties from Space Charge-Induced Interfacial Polarization in Multilayer Polymer Films

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1. Soft Touch Electrode for High Temperature Breakdown Study

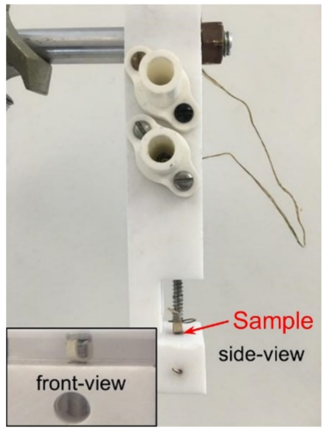

Fig. S1. Side- and front-view micrographs of the soft touch electrode used for high temperature Weibull breakdown strength analysis. Samples are put between upper soft electrode and the lower electrode. Here, the upper soft electrode is made by a silver-filled fluorosilicone (which cannot be swollen by the silicone oil and has a volume resistivity of ca. 0.002 Ω·cm). Part of the fixture with film samples were immersed in silicone oil to avoid corona breakdown.

2. Edge-on SAXS and WAXD Patterns for the PC/PVDF 50/50 32L and 256L Films

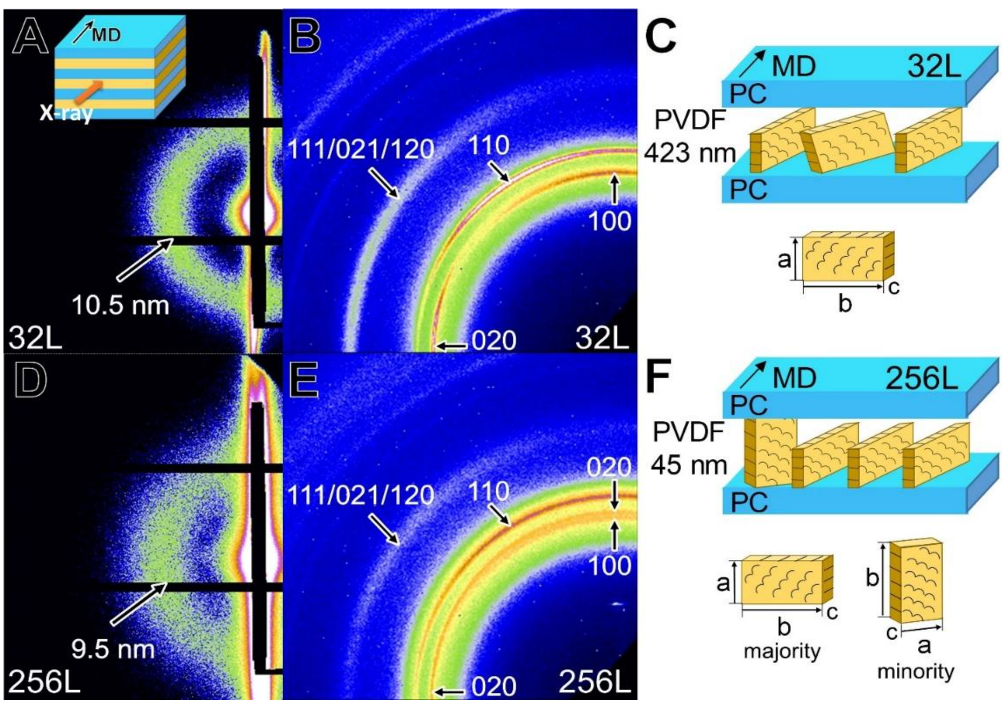

Fig. S2. (A,D) Small- and (B,E) wide-angle X-ray scattering (SAXS and WAXD) patterns for the PC/PVDF 50/50 (A,B) 32L and (D,E) 256L films when the X-ray is directed parallel to the machine direction (MD) of the film. Schematic lamellar and crystal orientation for confined PVDF in the 32L and 256L films are shown in (C) and (F), respectively. Lamellar thicknesses and crystalline reflections of PVDF α phase are indicated in SAXS and WAXD patterns, respectively.

PVDF crystal orientation in nanoconfined spaces (i.e., in the PC/PVDF 50/50 32L and 256L films) was studied by simultaneous SAXS and WAXD techniques. The edge-on 2D SAXS and WAXD patterns for both 32L and 256L films are shown in Figure S2. From the edge-on WAXD pattern in Figure S2B, the α-phase (100) and (020) reflections were on both vertical and horizontal directions, respectively. The (110) reflection was located at ca. 40° from the vertical direction. This 2D WAXD pattern suggested that the PVDF α phase crystals oriented with their *c*- and *b*-axes parallel to the film (thus, the *a*-axes oriented in the vertical direction). Similar orientation was also reported for nanoconfined PVDF.[S1] From the 2D SAXS pattern in Figure S2A, the lamellar crystals showed a stacking spacing of 10.5 nm with the maximum intensity oriented in the horizontal direction. This is consistent with the crystal orientation of the *c*-axes parallel to the film. Figure S2C shows the schematic crystal and lamellar orientations in the confined PVDF layers (ca. 423 nm) in the 32L film. Similar crystal and lamellar orientations were observed for the 256L film, except that the lamellar stacking spacing was 9.5 nm (Figure S2D). In addition, a minor crystal orientation was seen from the WAXD pattern in Figure S2E, because a weak/broad (020) reflection arc was also located in the vertical direction in addition to the sharp (020) reflection in the horizontal direction. Figure S2F shows schematic PVDF crystal and lamellar orientations confined in the 256 L film (the PVDF layer was ca. 45 nm thick). Currently, it is not clear why both majority (*b*-axes parallel to the film) and minority (*b*-axes perpendicular to the film) crystal orientation took place in the 45 nm confined space. More research is needed in the future to understand this observation.

3. Leakage Currents for the PC/PVDF 50/50 32L Film at Various Temperatures

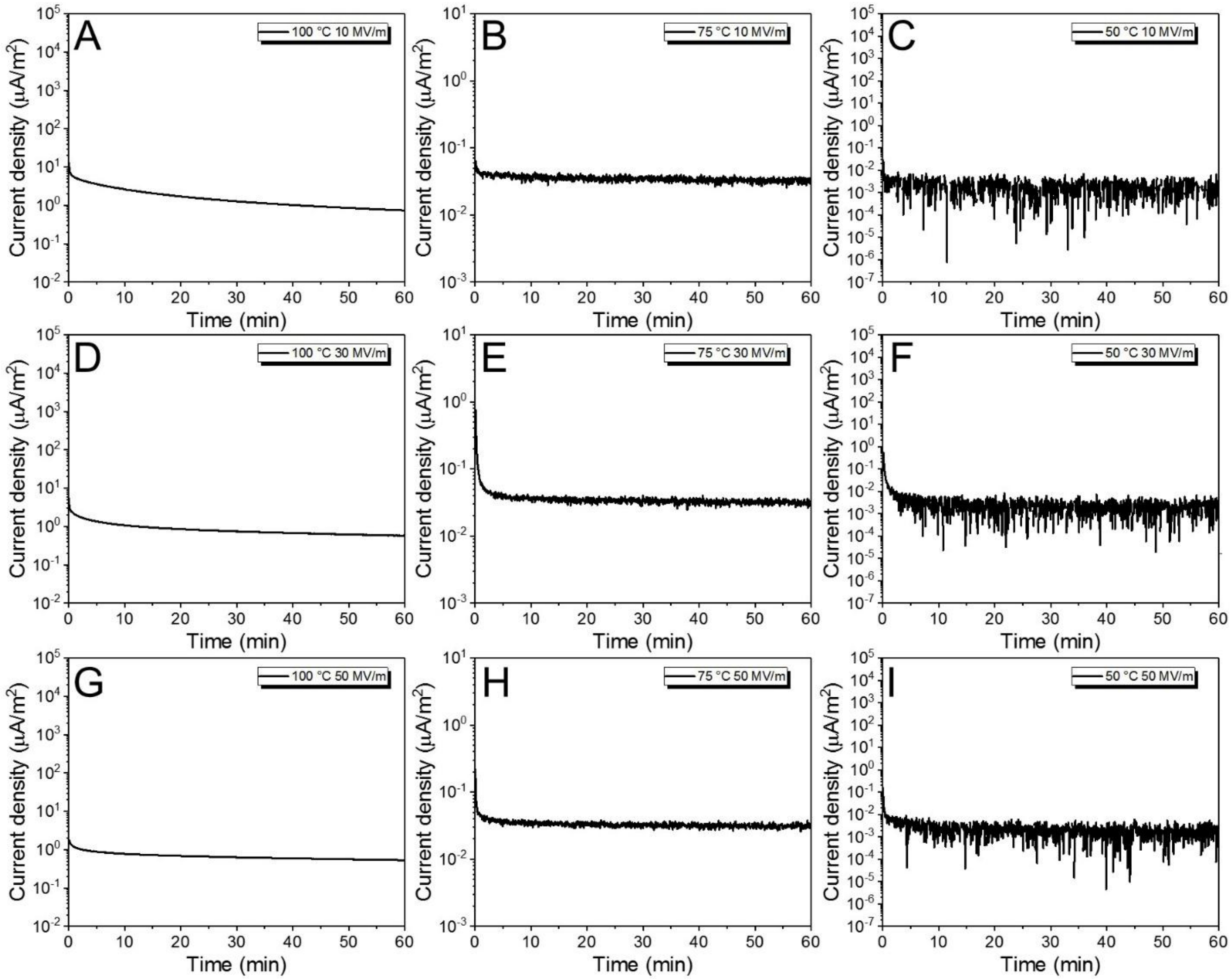

Fig. S3. Leakage current measurements for the PC/PVDF 50/50 32L film at (A,D,G) 100 °C, (B,E,H) 75 °C, and (C,F,I) 50 °C with the DC electric field being (A,B,C) 10 MV/m, (D,E,F) 30 MV/m, and (G,H,I) 50 MV/m, respectively.

4. Leakage Currents for the PC/PVDF 50/50 256L Film at Various Temperatures

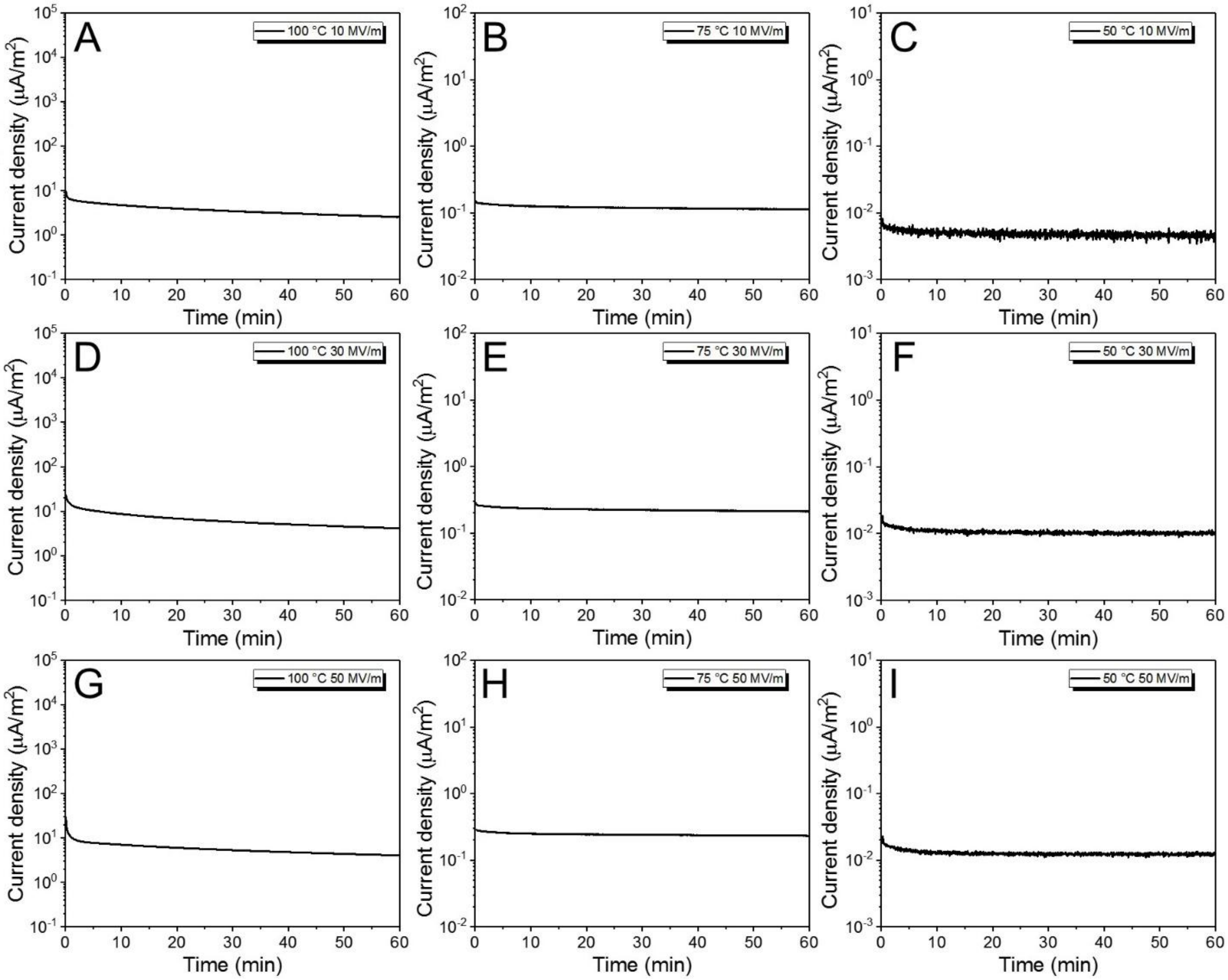

Fig. S4. Leakage current measurements for the PC/PVDF 50/50 256L film at (A,D,G) 100 °C, (B,E,H) 75 °C, and (C,F,I) 50 °C with the DC electric field being (A,B,C) 10 MV/m, (D,E,F) 30 MV/m, and (G,H,I) 50 MV/m, respectively.

5. Temperature-Scan BDS Results for PC/PVDF 50/50 32L and 256L Films

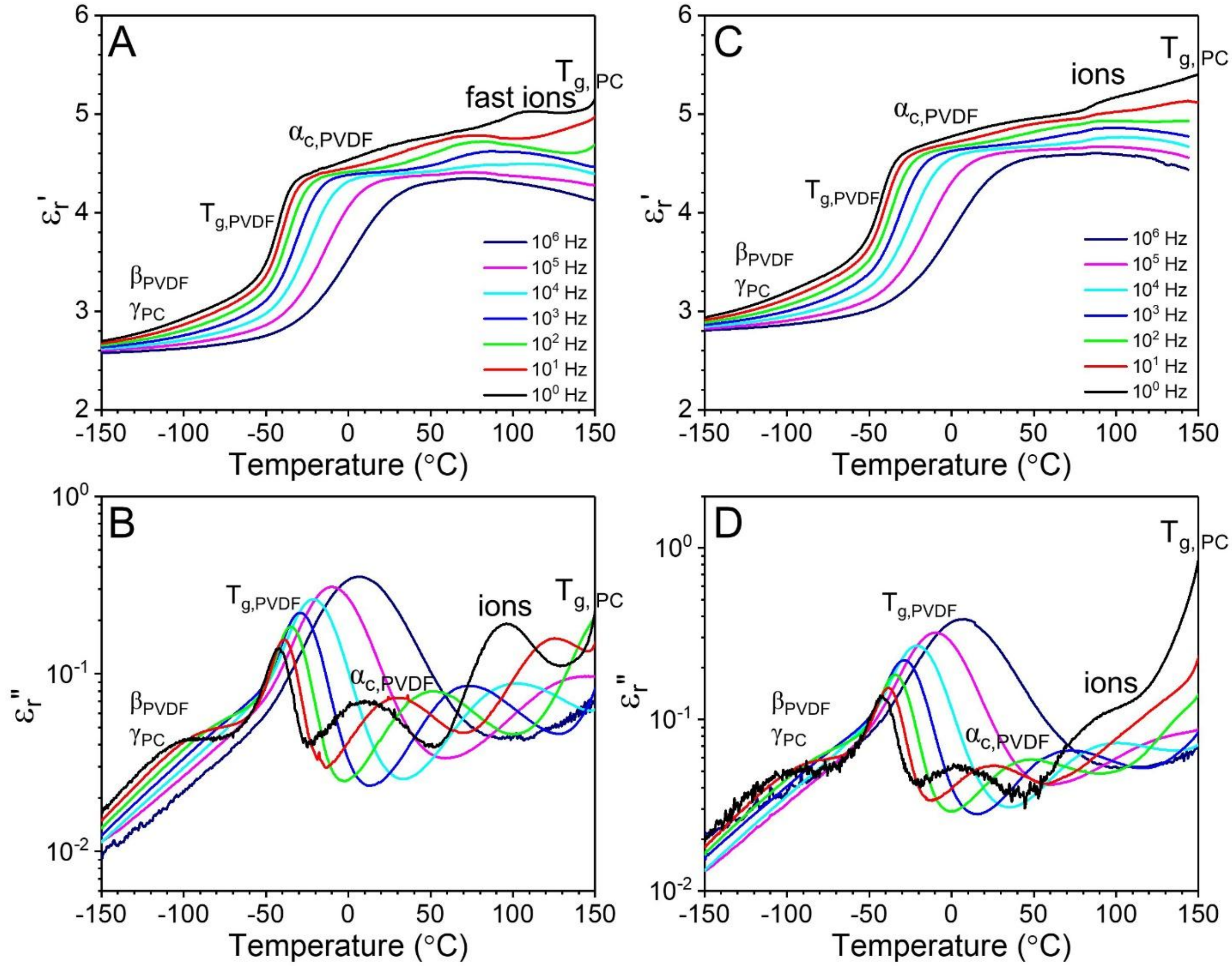

Fig. S5. (A,C) Real (ε_r') and (B,D) imaginary (ε_r'') relative permittivities as a function of temperature at different frequencies for (A,B) 32L and (C,D) 256L PC/PVDF 50/50 films. Different phase transitions for the PC and PVDF layers are labeled in the plots.

References

S1. M. Mackey, D. E. Schuele, L. Zhu, E. Baer, *J. Appl. Phys.* **2012,** *111*, 113702.