Supporting Information

Reducing dielectric loss and enhancing electrical insulation for multilayer polymer films by nanoconfined ion transport under high poling electric fields

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I. Melt Viscosities of HTPC and PVDF as a Function of Temperature

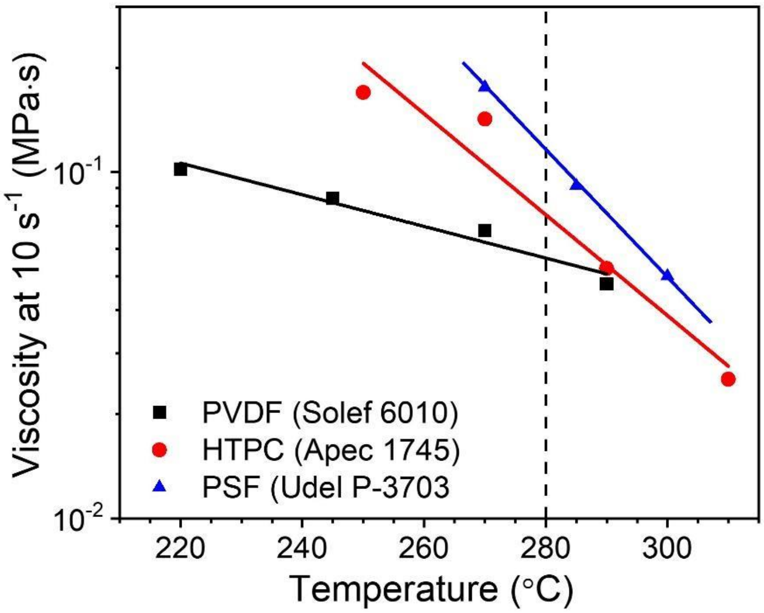

Figure S1. Melt viscosity at 10 s^{-1} as a function of temperature for HTPC and PVDF. This plot was used to determine the coextrusion temperature, which was around 280 °C. A higher processing temperature (> 290 °C) should be avoided, because PVDF tends to thermally decompose to emit the poisonous HF gas.

II. HN-Deconvolution of HV-BDS Curves

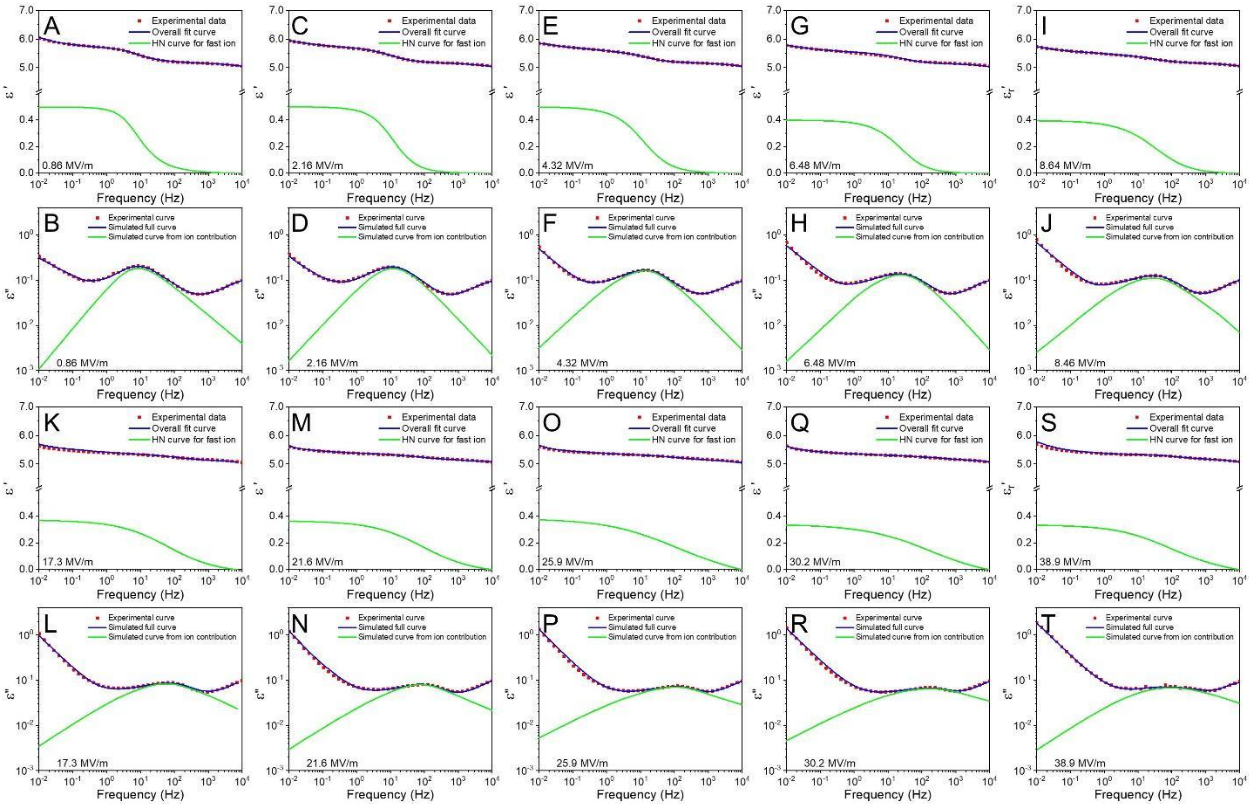

Figure S2. Deconvoluted HN ε_r' and ε_r'' curves for fast ions in the HTPC/PVDF 50/50 33L film (11.6 μm thickness) at (A,B) 0.86, (C,D) 2.16, (E,F) 4.32, (G,H) 6.48, (I,J) 8.64, (K,L) 17.3, (M,N) 21.6, (O,P) 25.9, (Q,R) 30.2, and (S,T) 38.9 MV/m, respectively. Solid symbols: experimental data, blue curves: overall fit curves, and green curves: HN curves for fast ions. HN fit curves for the $\alpha_{c,PVDF}$ relaxation and slow-ion conduction are not shown for clarity. Measurements were performed at 130 °C.

III. Determination of Optimal Electric Poling Conditions to Lock Impurity Ions in the HTPC Layers

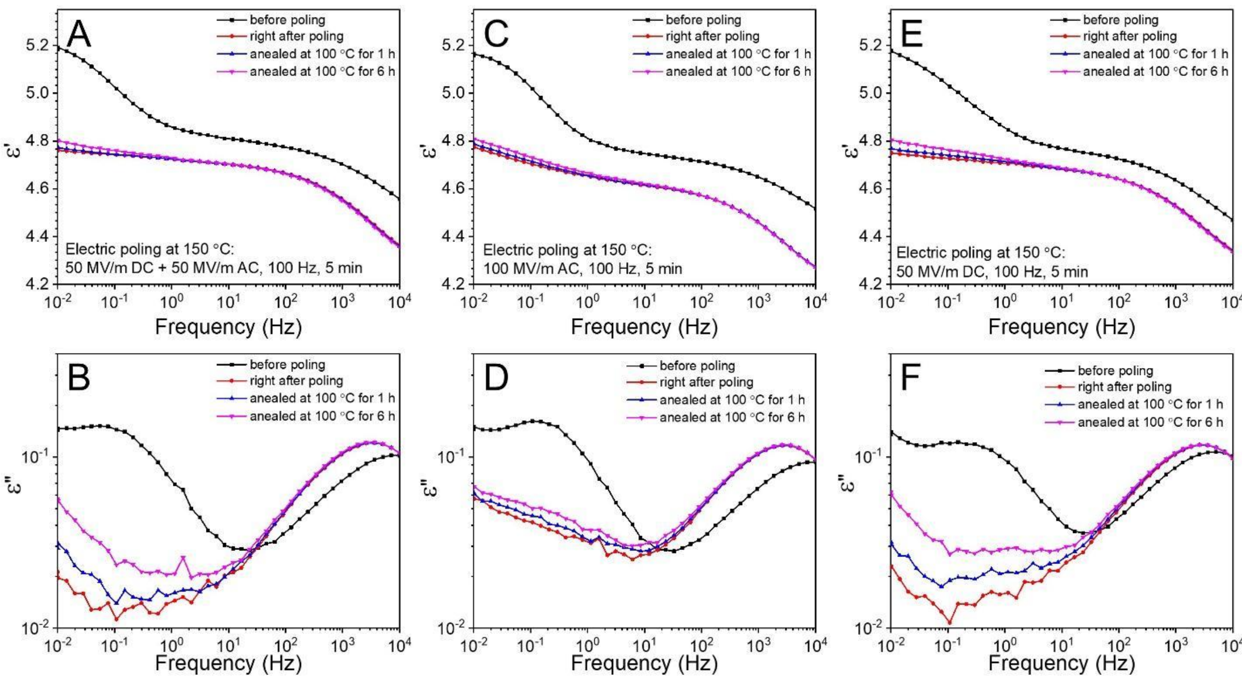

Figure S3. Comparison of different electric poling methods, i.e., (A,B) DC+AC, (C,D) AC, and (E,F) DC, on the ion conduction loss at low frequencies using HV-BDS. The poling time was 5 min at 150 °C. The poling fields are shown in the plots. Suppression of the fast ion conduction peak around 0.2 Hz is the worst for the AC poling method. This is understandable because the AC poling only drives the ion motion back and forth. Comparing the DC+AC and DC methods, the DC+AC method shows slightly better performance, i.e., lower ε″ around 1 Hz. Therefore, we consider that the DC+AC method should be adopted for efficient electric poling.

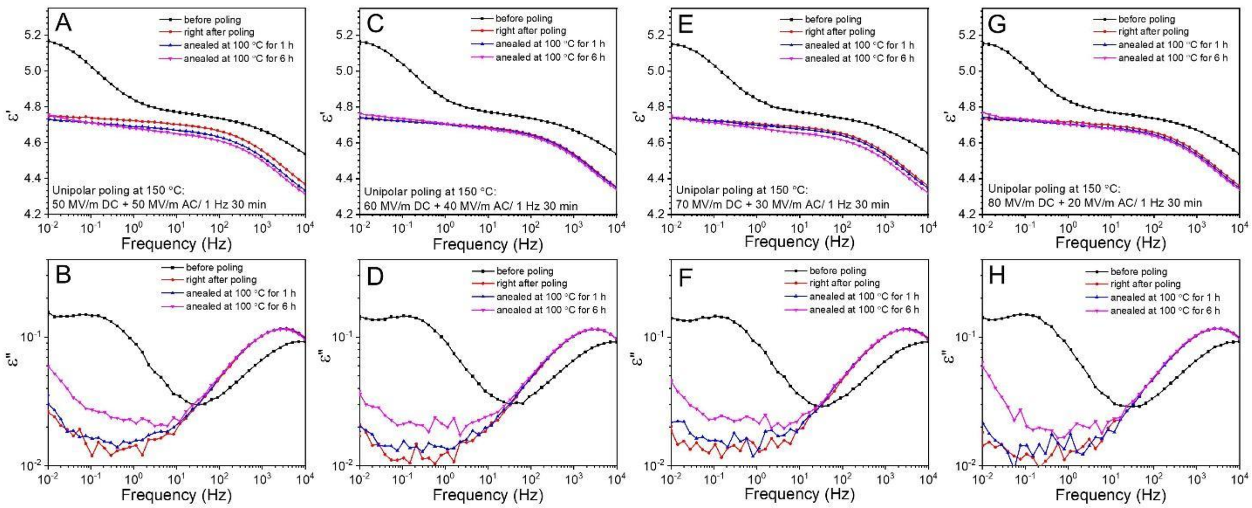

Figure S4. Comparison of different combinations of DC and AC electric fields for the DC+AC poling method. (A,B) 50 MV/m DC + 50 MV/m AC, (C,D) 60 MV/m DC + 40 MV/m, (E,F) 70 MV/m DC + 30 MV/m, and (G,H) 80 MV/m DC + 20 MV/m AC. The poling frequency was 1 Hz, and the poling time was 30 min at 150 °C. To avoid easy electric breakdown, we chose the condition of 60 MV/m DC + 40 MV/m at 1 Hz.

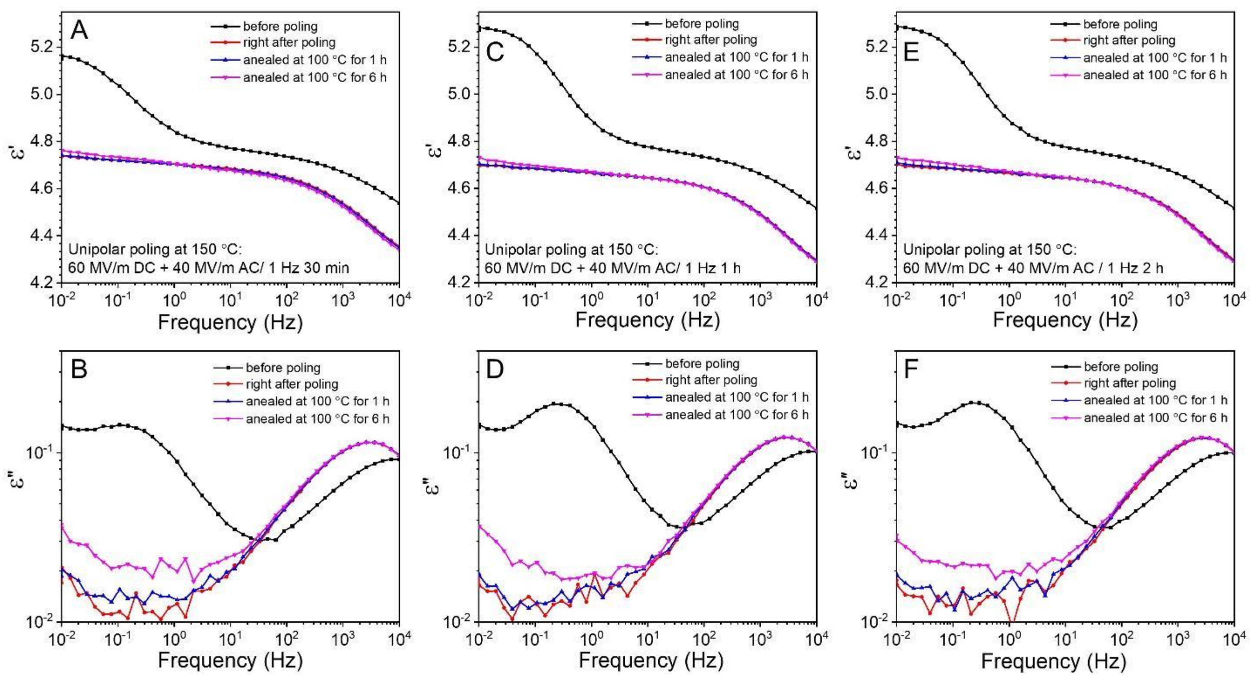

Figure S5. Comparison of different poling times for the DC+AC poling method. (A,B) 30 min, (C,D) 1 h, and (E,F) 2 h. The poling frequency was 1 Hz at 150 °C. Finally, 2 h poling gives the lowest fast ion conduction loss.

From Figure S3-S5, the optimal electric poling conditions were determined to be: 60 MV/m DC + 40 MV/m AC, 1 Hz, and 2 h at 150 °C.

IV. Determination of PVDF Crystal Polymorphism and Orientation Before and After Electric Poling

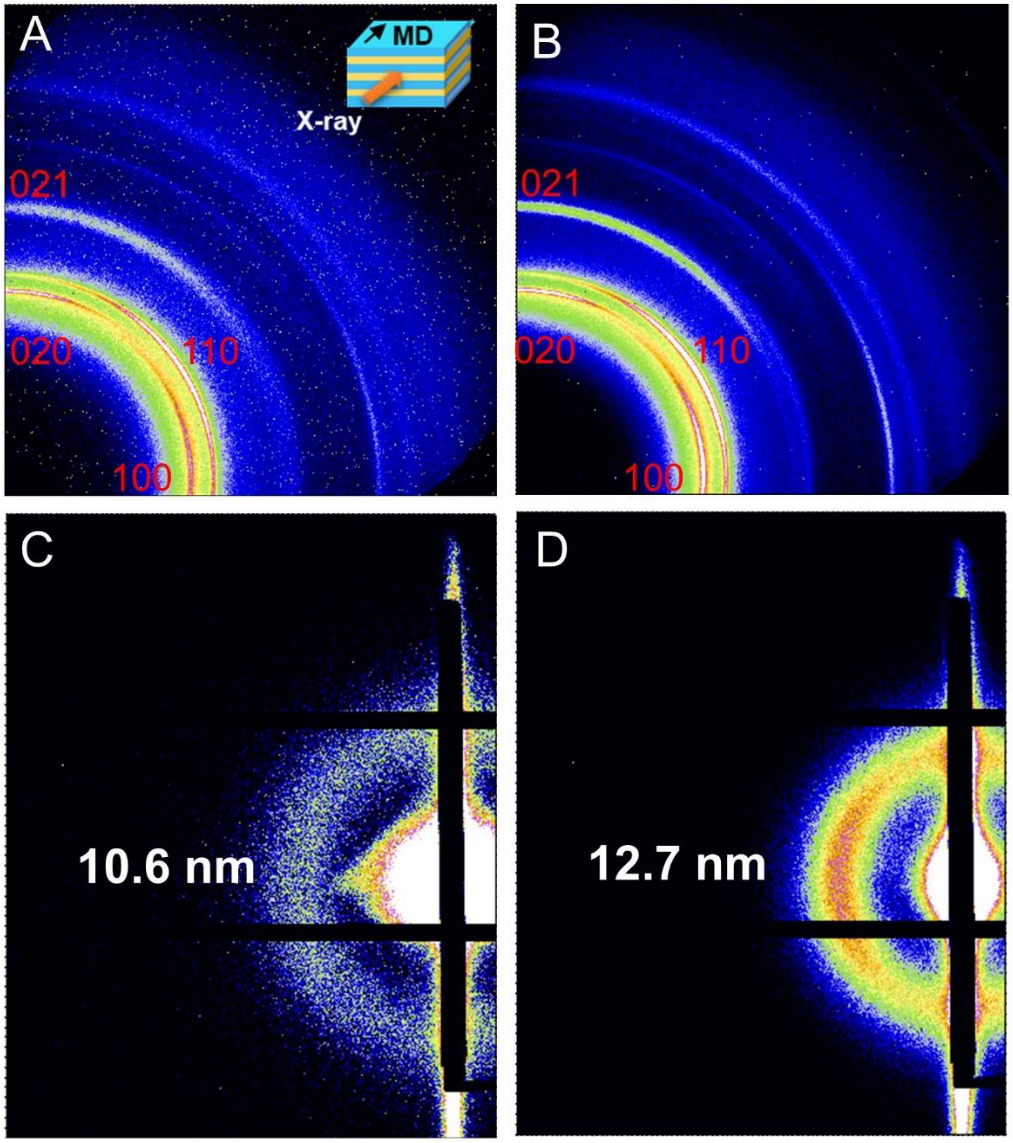

Figure S6. 2D (A,B) WAXD and (C,D) SAXS patterns for the HTPC/PVDF 50/50 33L films. (A,C) Before and (B,D) after electric poling (60 MV/m DC + 40 MV/m AC at 1 Hz and 150 °C for 2 h). The X-ray beam was directed along the machine direction (MD) of the films, which are placed in the horizontal direction. Based on the oriented WAXD patterns, edge-on α crystal of PVDF are determined.