

**Supporting Information:**

**Novel crosslinkable high-k copolymer dielectrics for high-energy-density capacitors and organic field-effect transistors applications**

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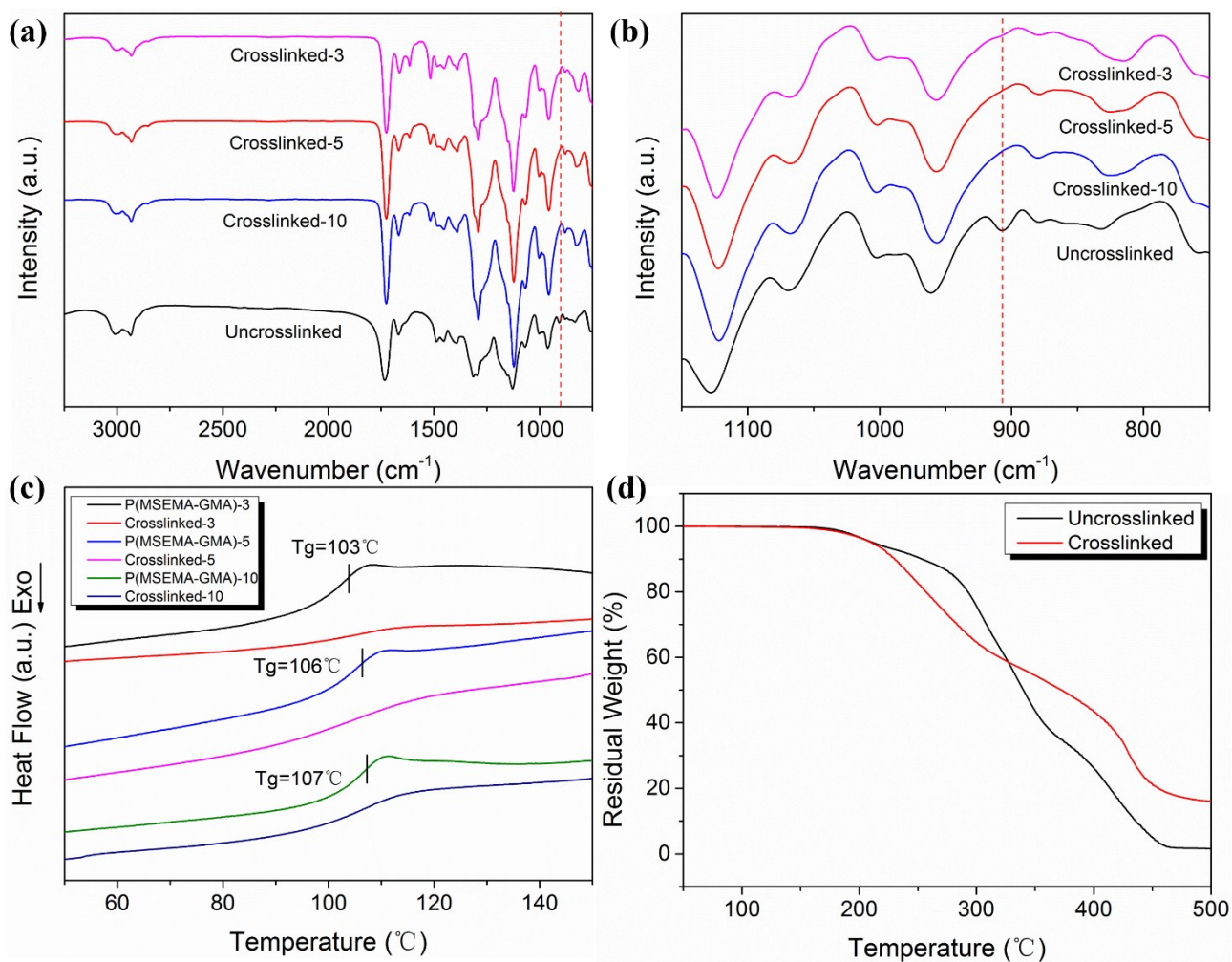
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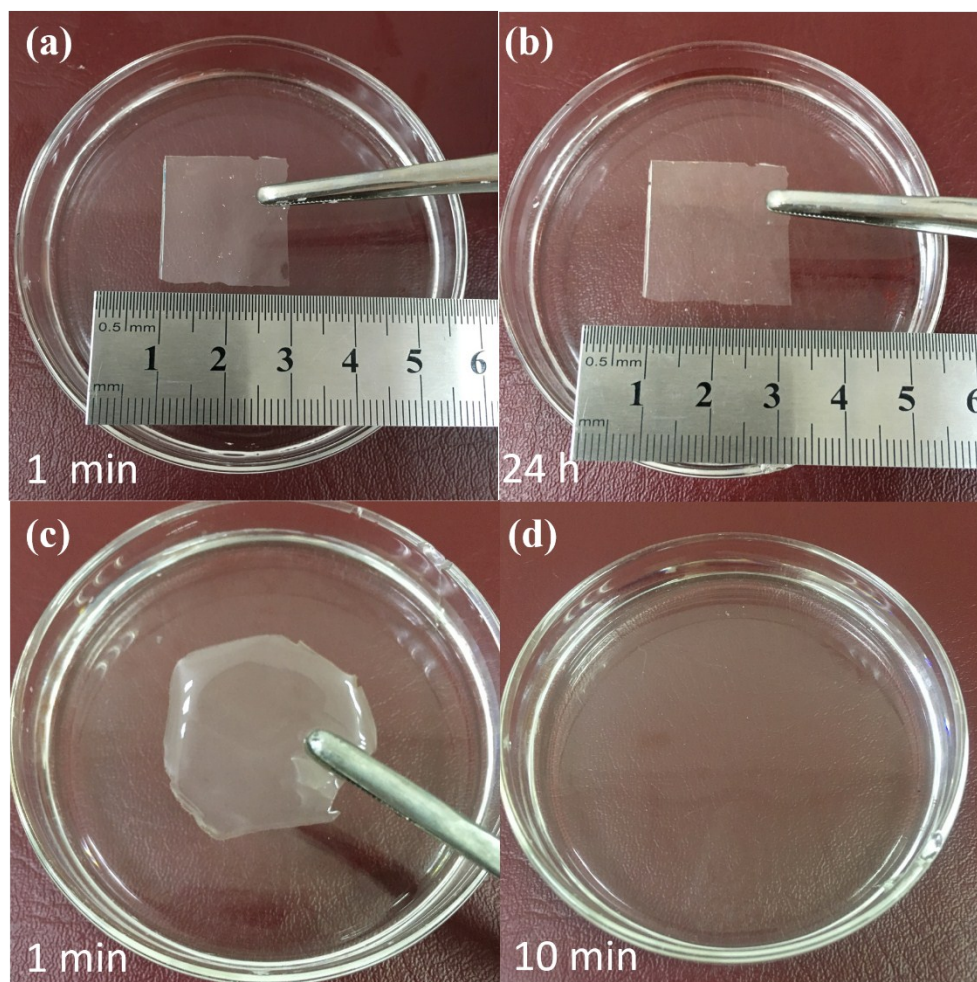
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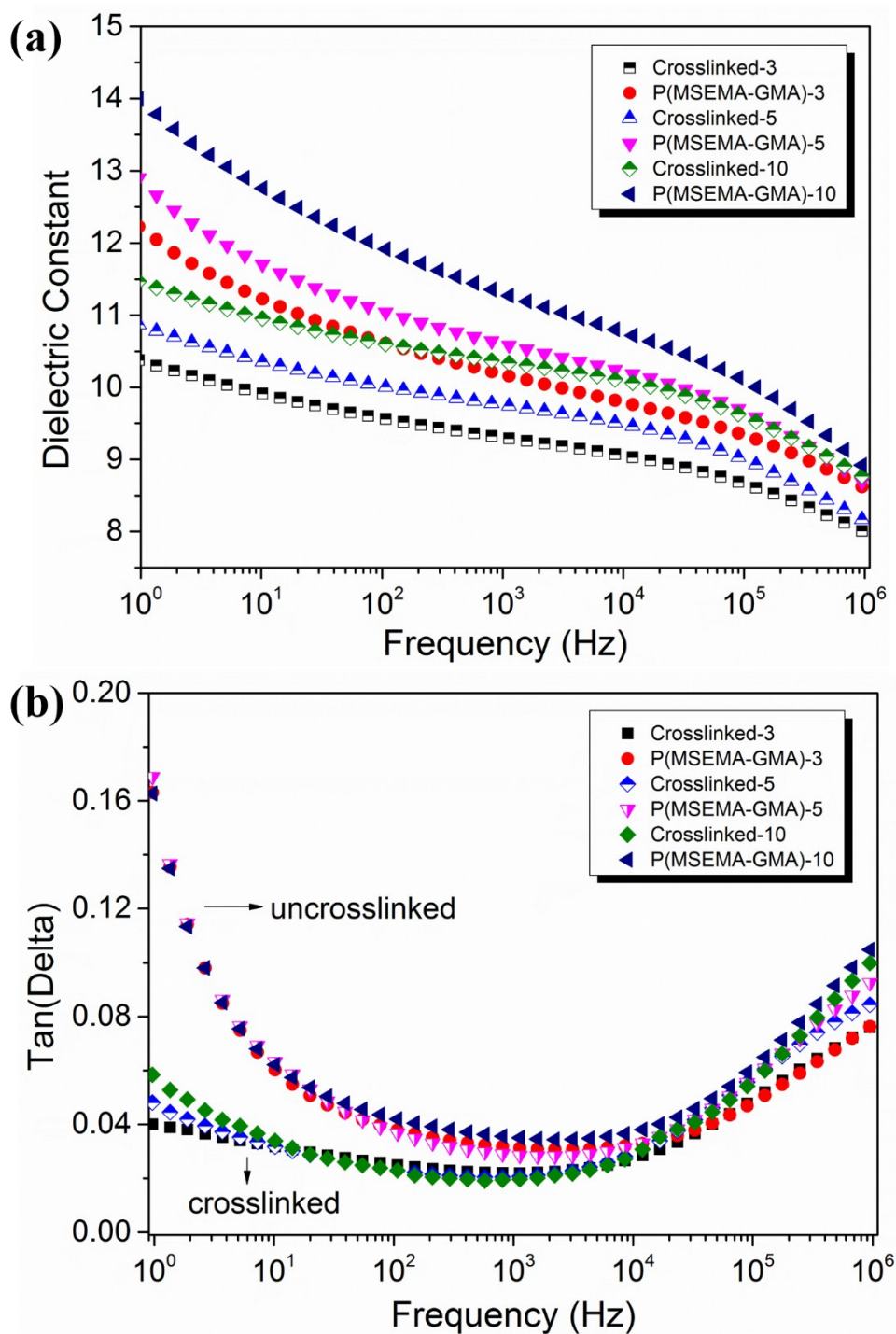
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**Figure S1.** (a) FT-IR spectra of copolymers before and after crosslinking and (b) the amplified region of the epoxy groups. (c) DSC curves and (d) TGA curves of polymers before and after crosslinking.

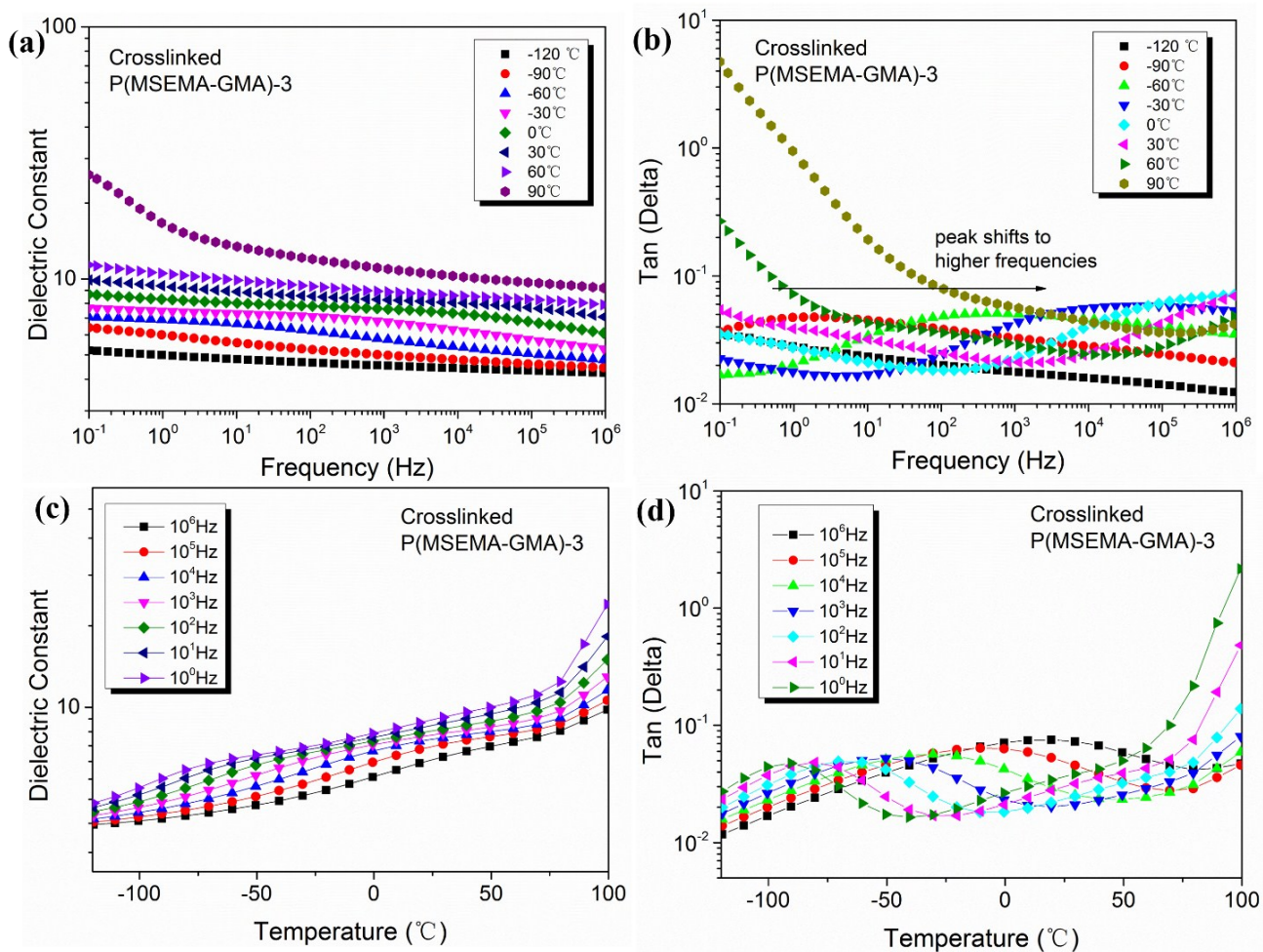


**Figure S2.** Optical images of crosslinked (a)-(b) and uncrosslinked (c)-(d) P(MSEMA-*co*-GMA)-3 films after soak in DMF for different time period. The crosslinked polymer showed a virtually unchanged image and slight swelling after the 24-h soak, while the uncrosslinked polymer turned white with shape-change after 1 min, and dissolved after 10 min.

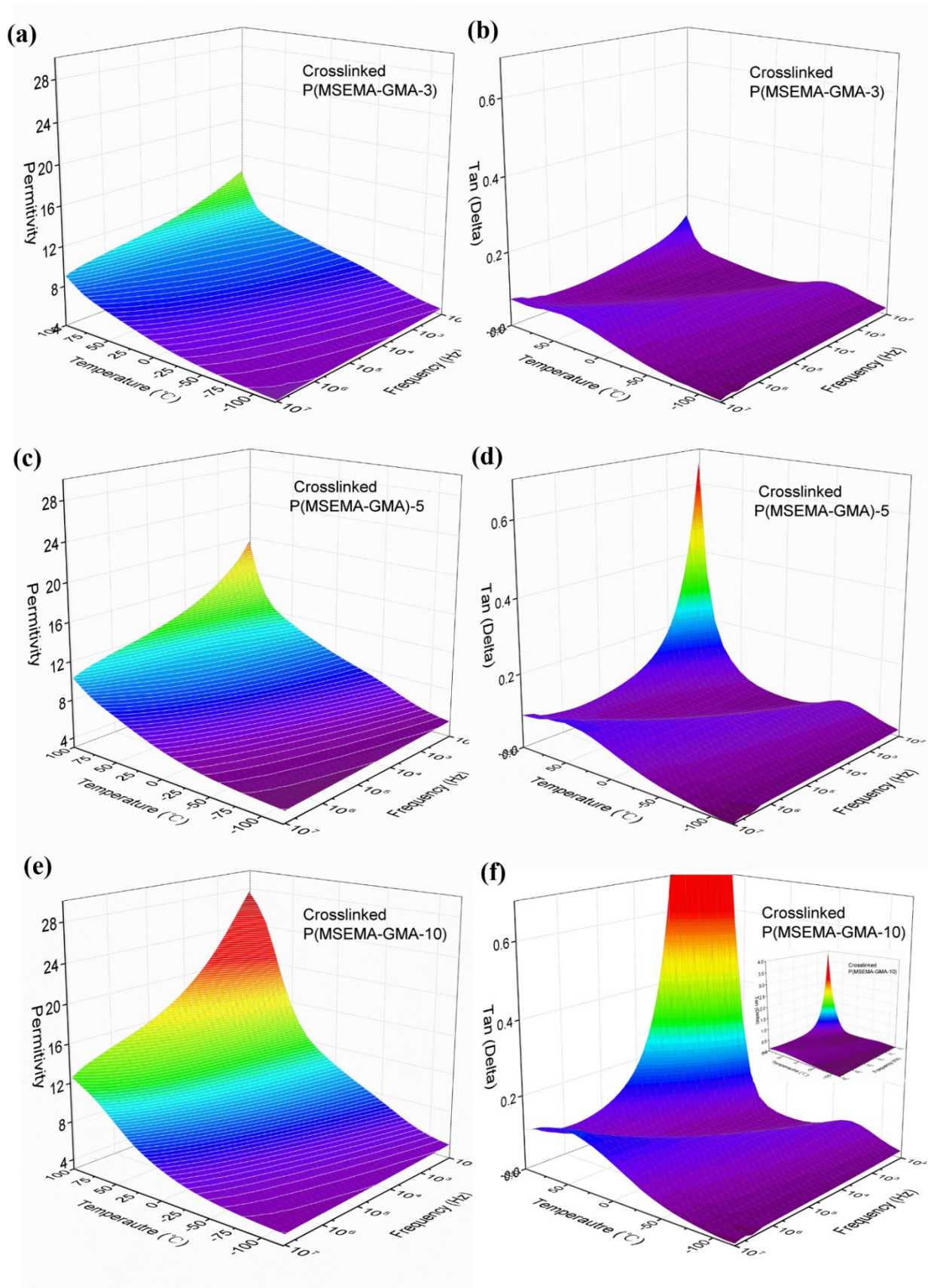


**Figure S3.** (a) Frequency-dependence of dielectric constant and (b) Frequency-dependence of dielectric loss of the three copolymers before and after crosslinking at room temperature.



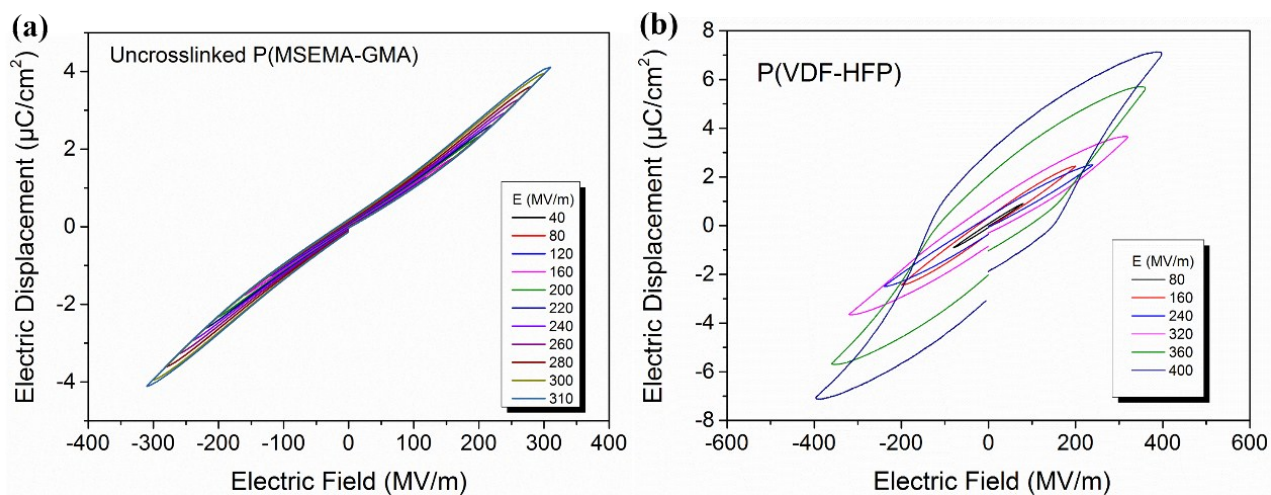


**Figure S4.** (a) Frequency dependence of dielectric constant and (b) Frequency dependence of dielectric loss under different temperatures for crosslinked P(MSEMA-GMA)-3. (c) Temperature dependence of dielectric constant and (d) Temperature dependence of dielectric loss of crosslinked P(MSEMA-GMA)-3 under selected frequencies.

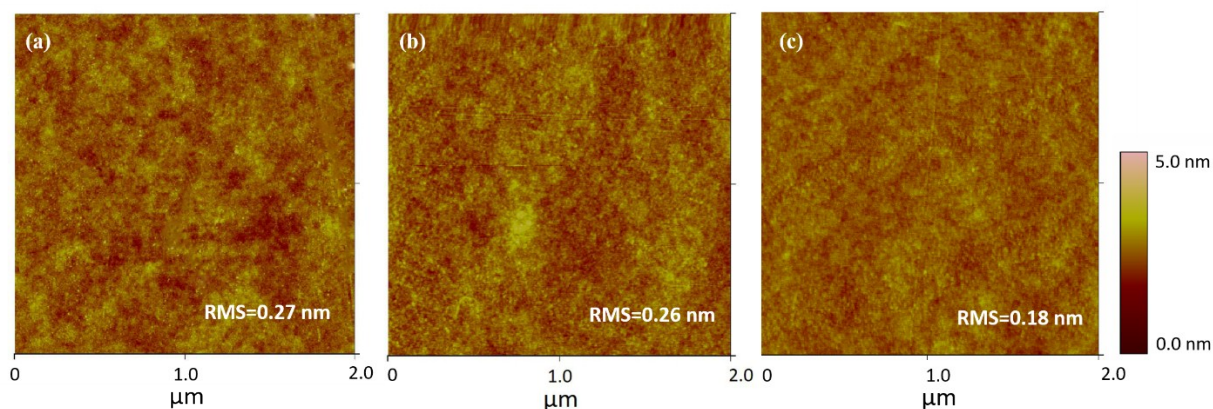


**Figure S5. (a)-(f)** Frequency and temperature dependent dielectric spectra of the three crosslinked copolymers.

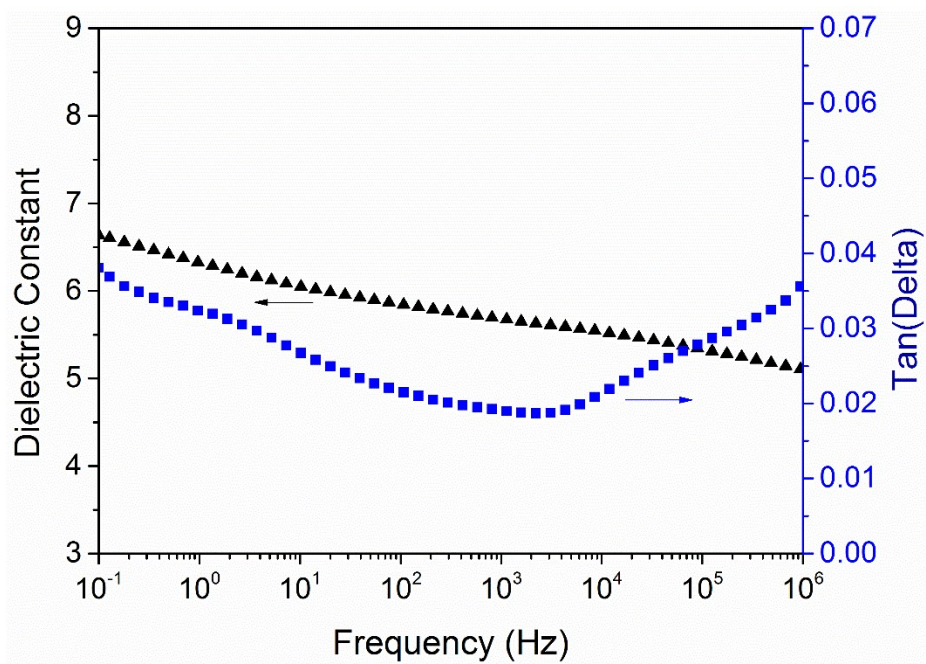




**Figure S6.** *D-E* loops of (a) uncrosslinked P(MSEMA-GMA) and (b) P(VDF-HFP) at 100 Hz, 25°C.



**Figure S7.** The atomic force microscopy (AFM) images (2  $\mu\text{m} \times 2 \mu\text{m}$  scan area) of the (a) P(MSEMA-GMA)-3, (b) P(MSEMA-GMA)-5 and (c) P(MSEMA-GMA)-10.



**Figure S8.** Frequency-dependence of dielectric constant and dielectric loss of poly(glycidyl methacrylate) (PGMA).