Supplementary Information

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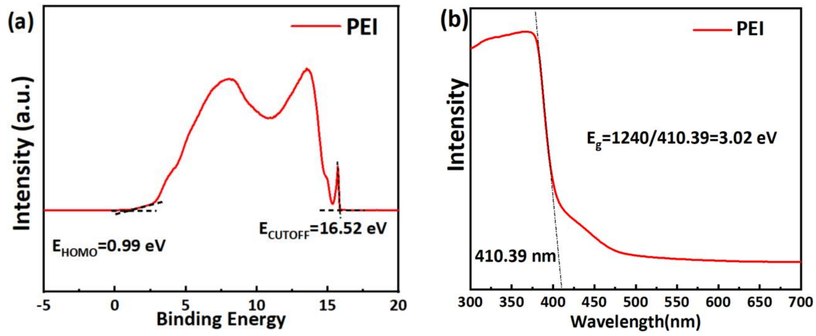

Figure S1 (a) UPS measurements in the secondary electron cutoff and HOMO regions of PEI. (b) UV-vis absorption spectra of PEI.

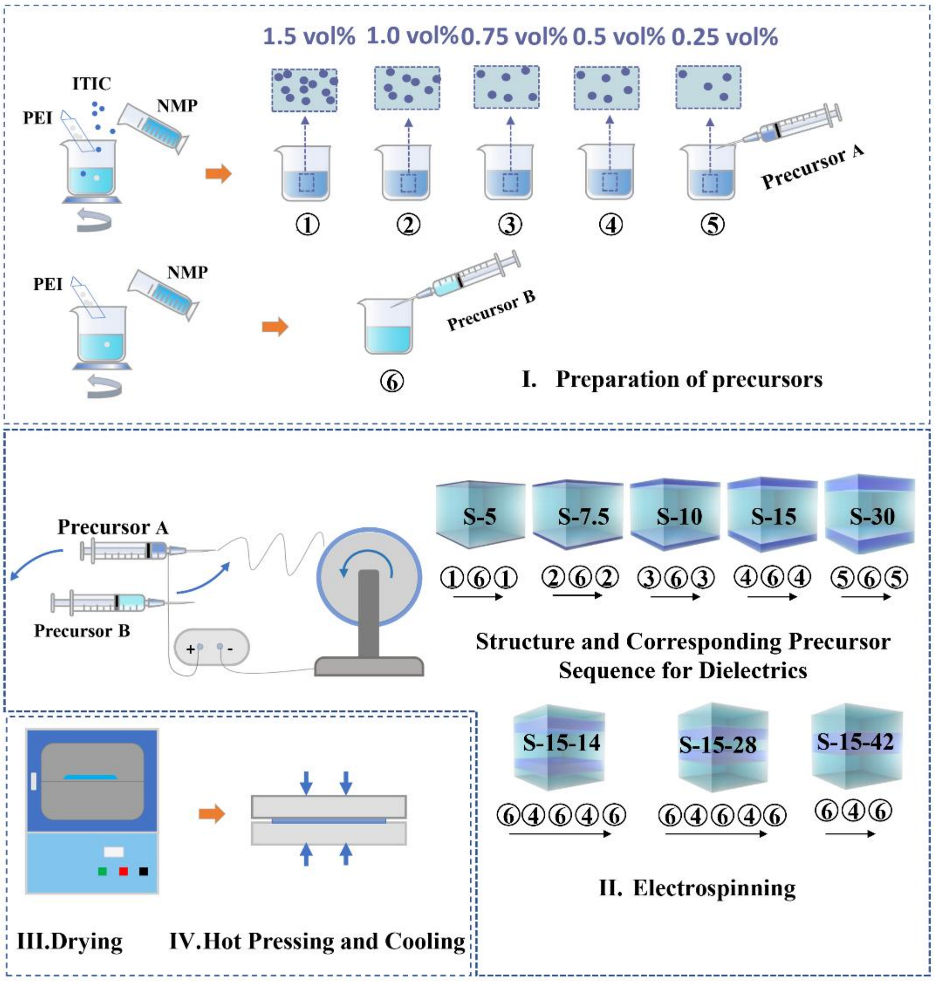

Figure. S2 Schematic of the preparation for trap-introduced PEI composite dielectrics.

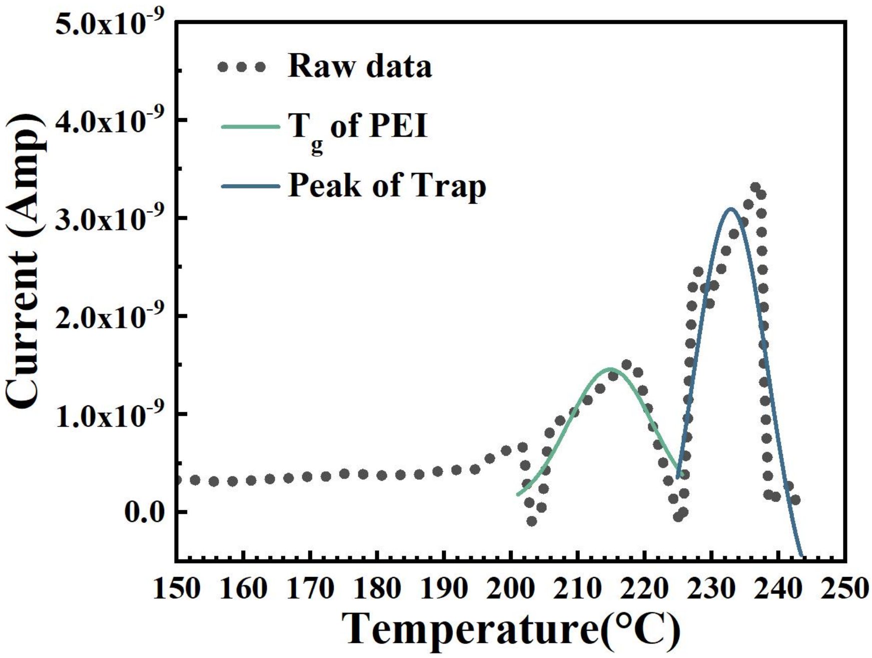

Figure S3. TSDC of S-15-28.

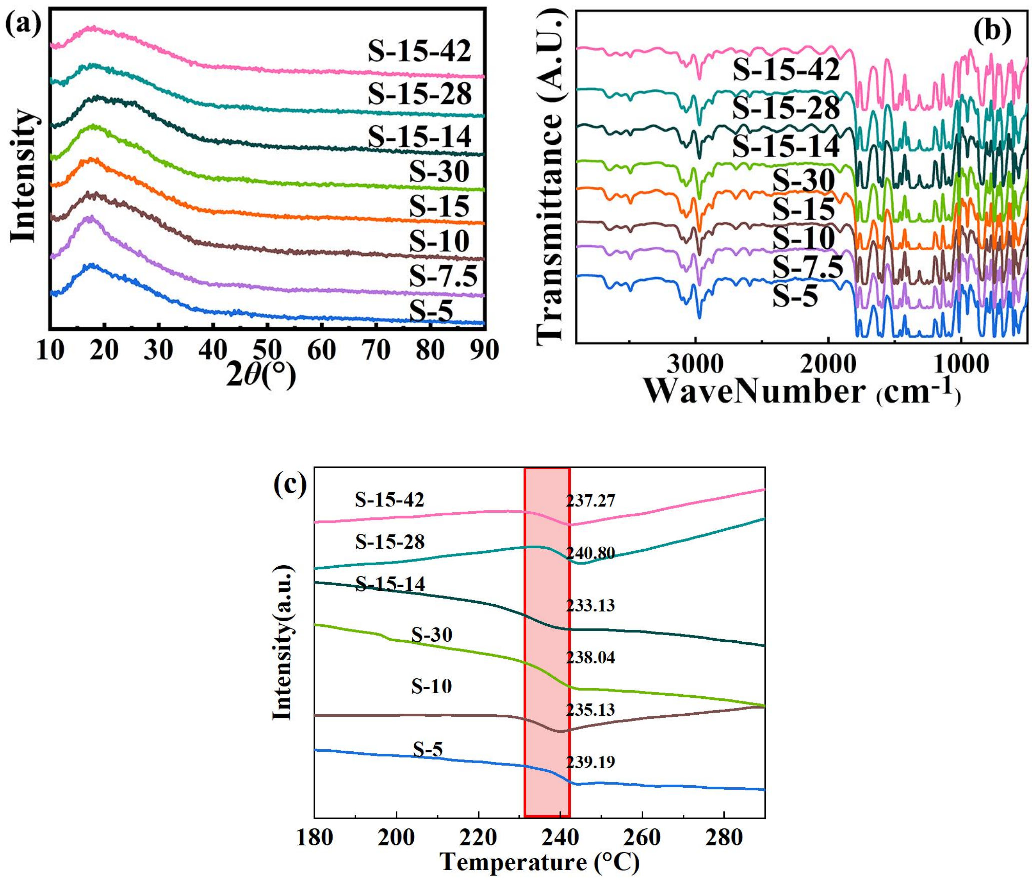

Figure S4. (a) XRD pattern and (b) FTIR spectrum (c) DSC curves for composite dielectrics.

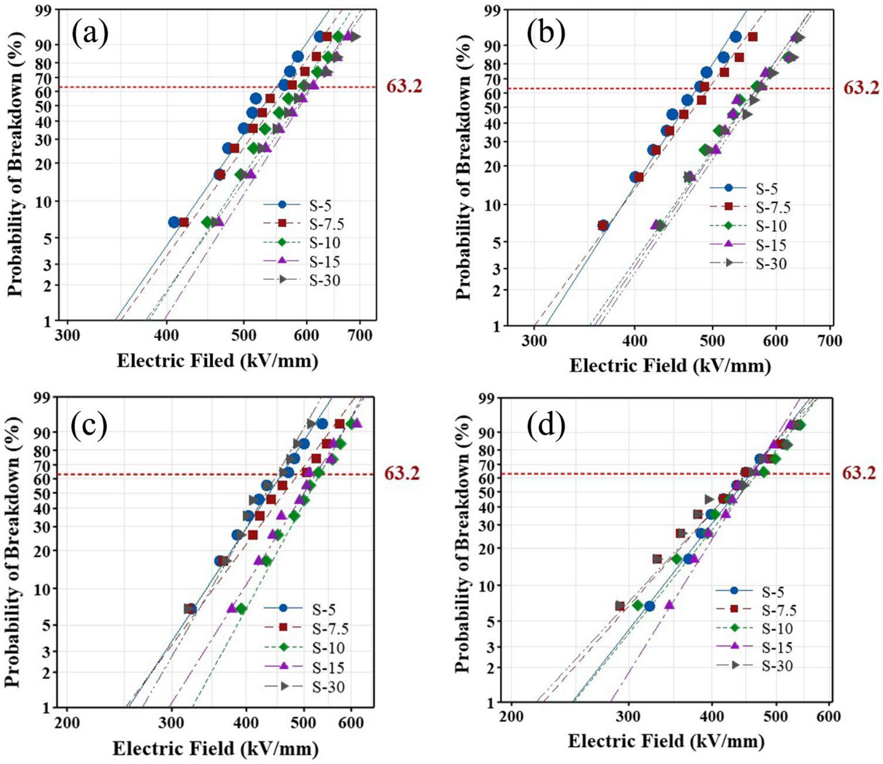

Figure S5. Weibull distribution of breakdown strength at (a) 20°C, (b) 100°C, (c) 150°C and (d) 180°C for composite dielectrics with different trap densities.

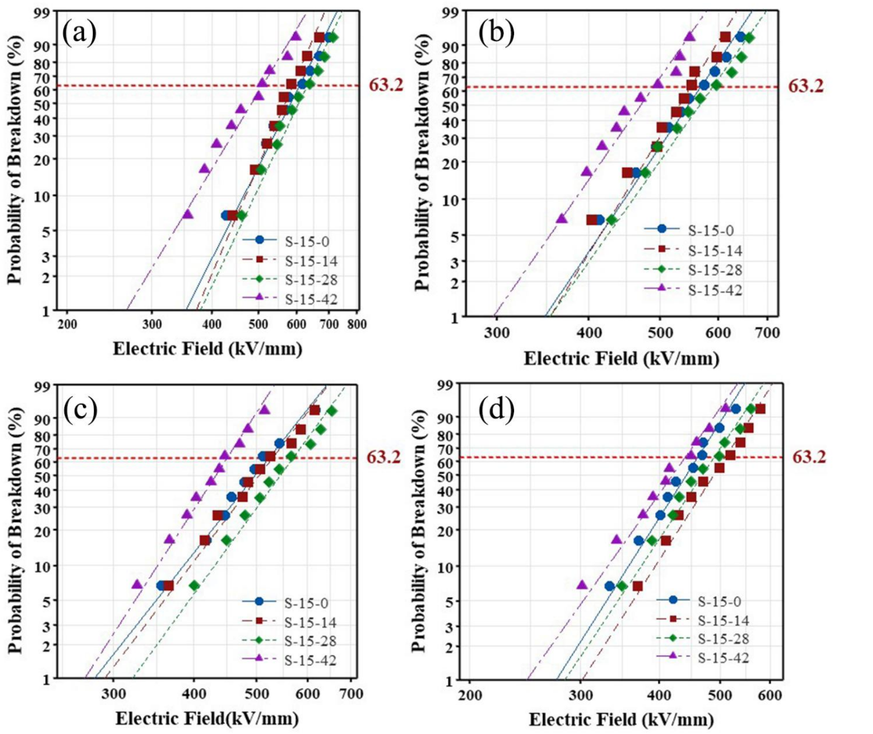

Figure S6. Weibull distribution of breakdown strength at (a) 20°C, (b) 100°C, (c) 150°C and (d) 180°C for composite dielectrics with different trap layer.

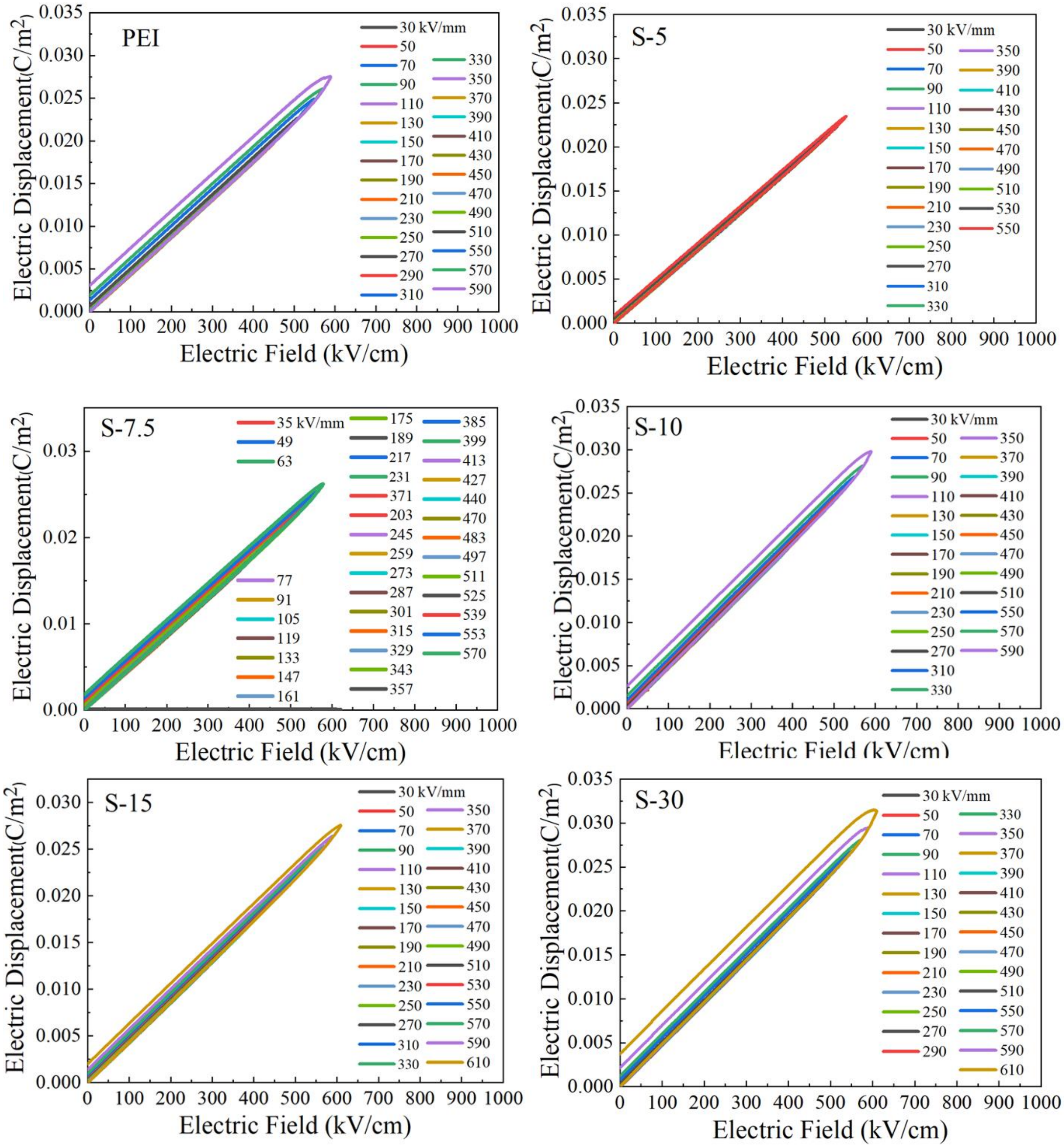

Figure S7. *D*-*E* loops for composite dielectrics with different trap density at 20°C.

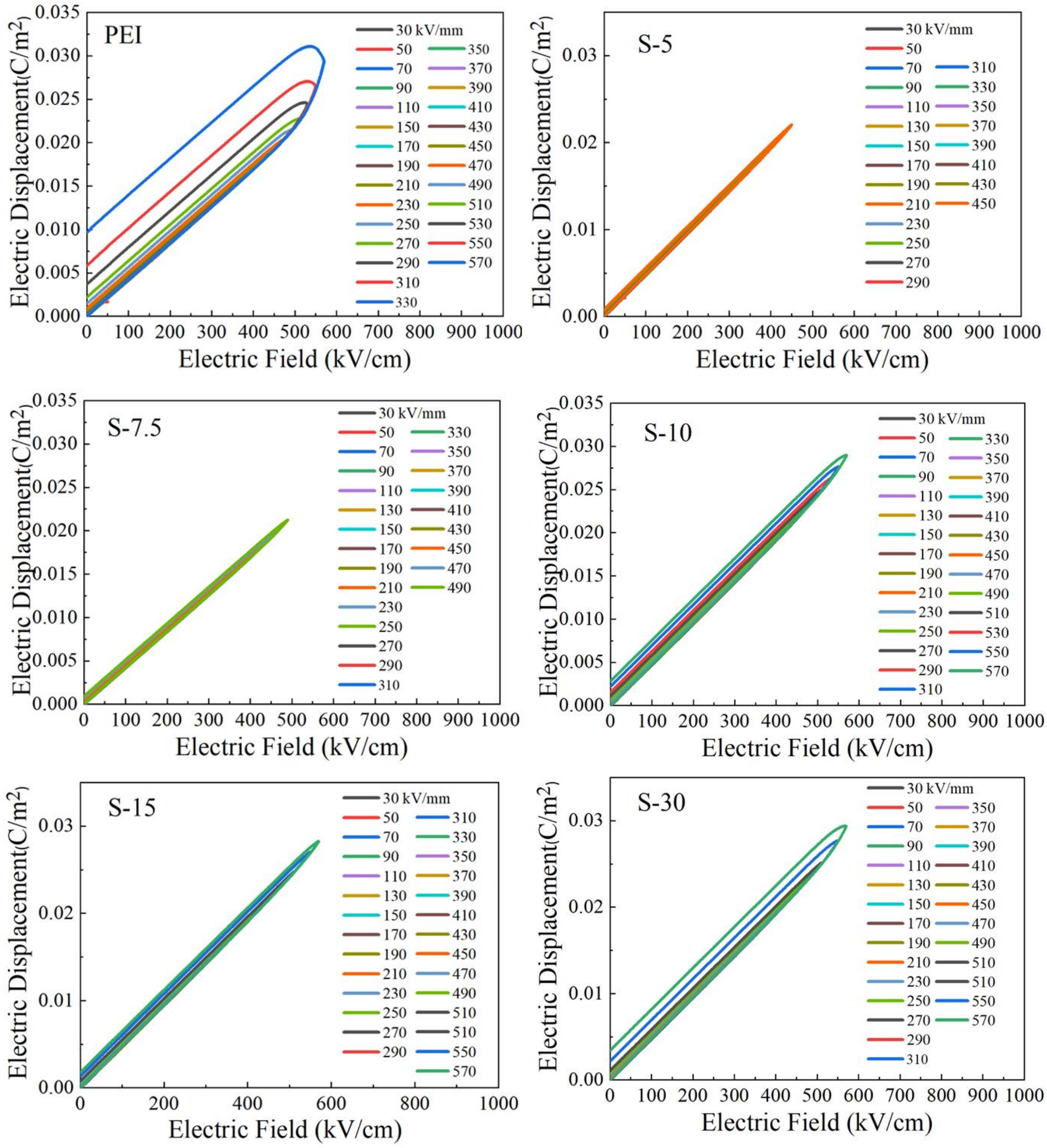

Figure S8. *D*-*E* loops for composite dielectrics with different trap density at 100°C

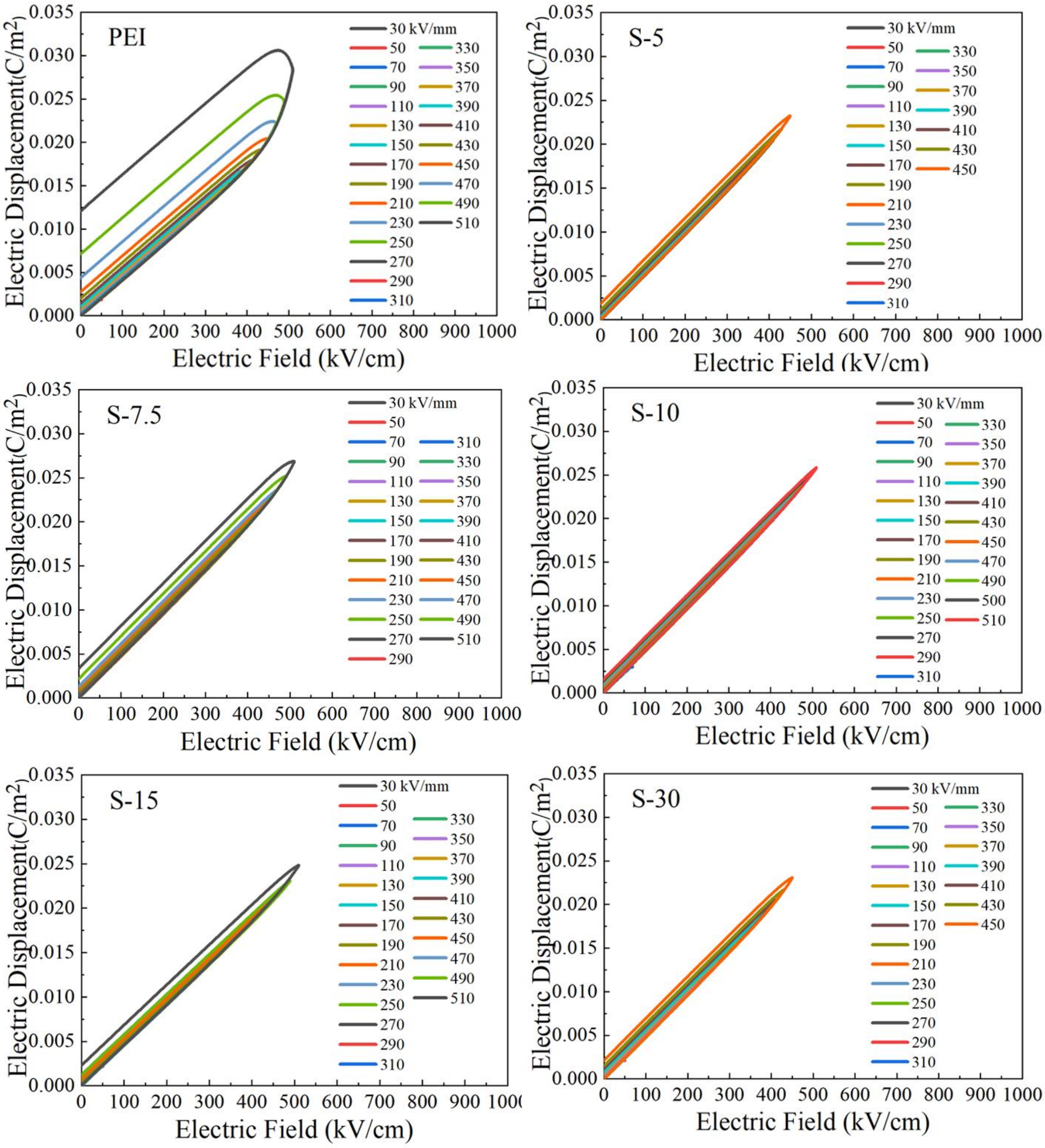

Figure S9. *D*-*E* loops for composite dielectrics with different trap density at 150°C

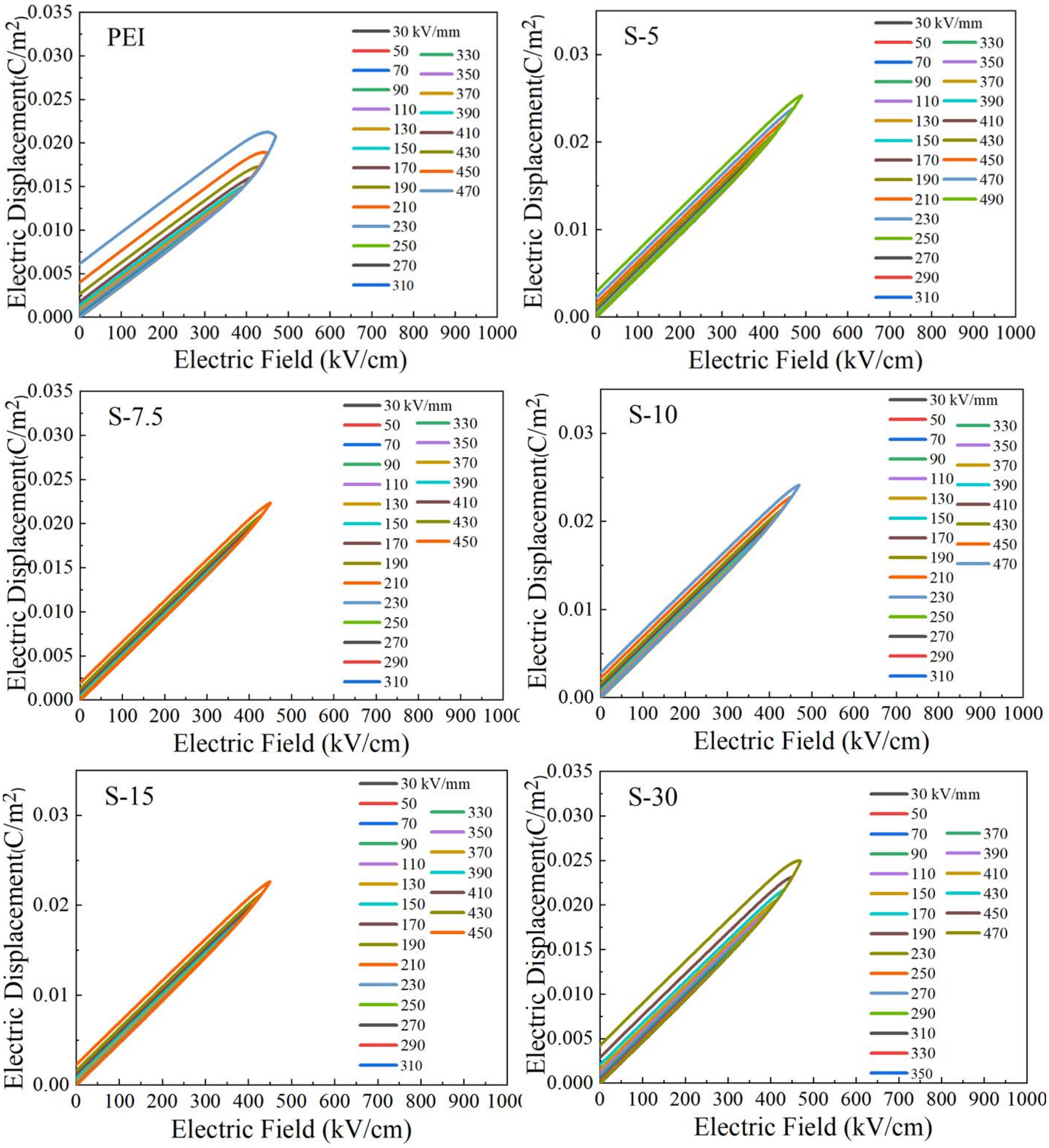

Figure S10 *D*-*E* loops for composite dielectrics with different trap density at 180°C

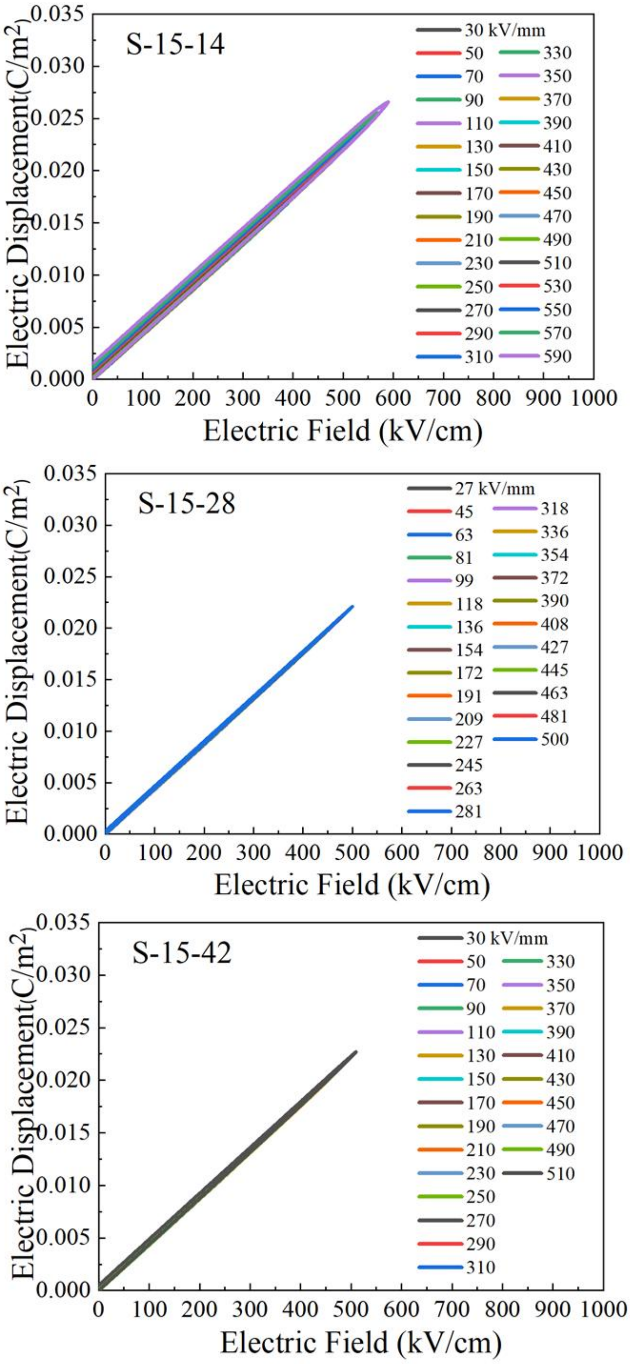

Figure S11 *D*-*E* loops for composite dielectrics with different trap layer locations at 20°C

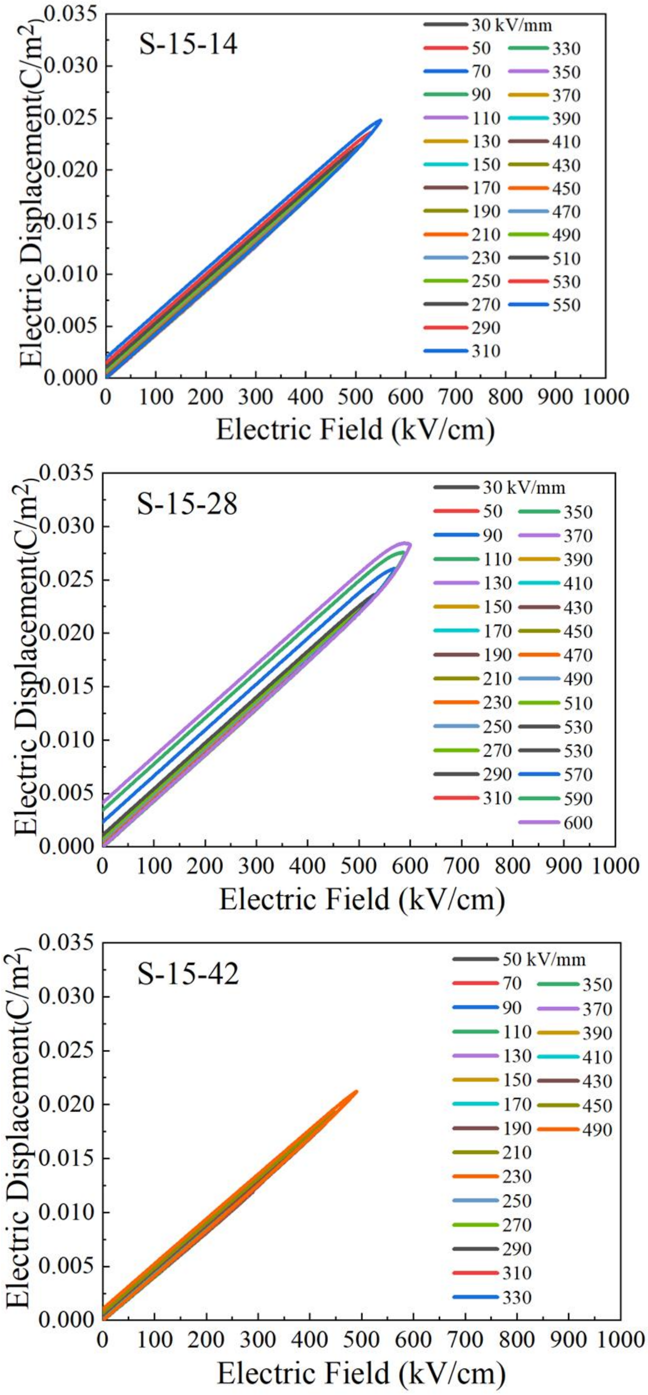

Figure S12 *D*-*E* loops for composite dielectrics with different trap layer locations at 100°C

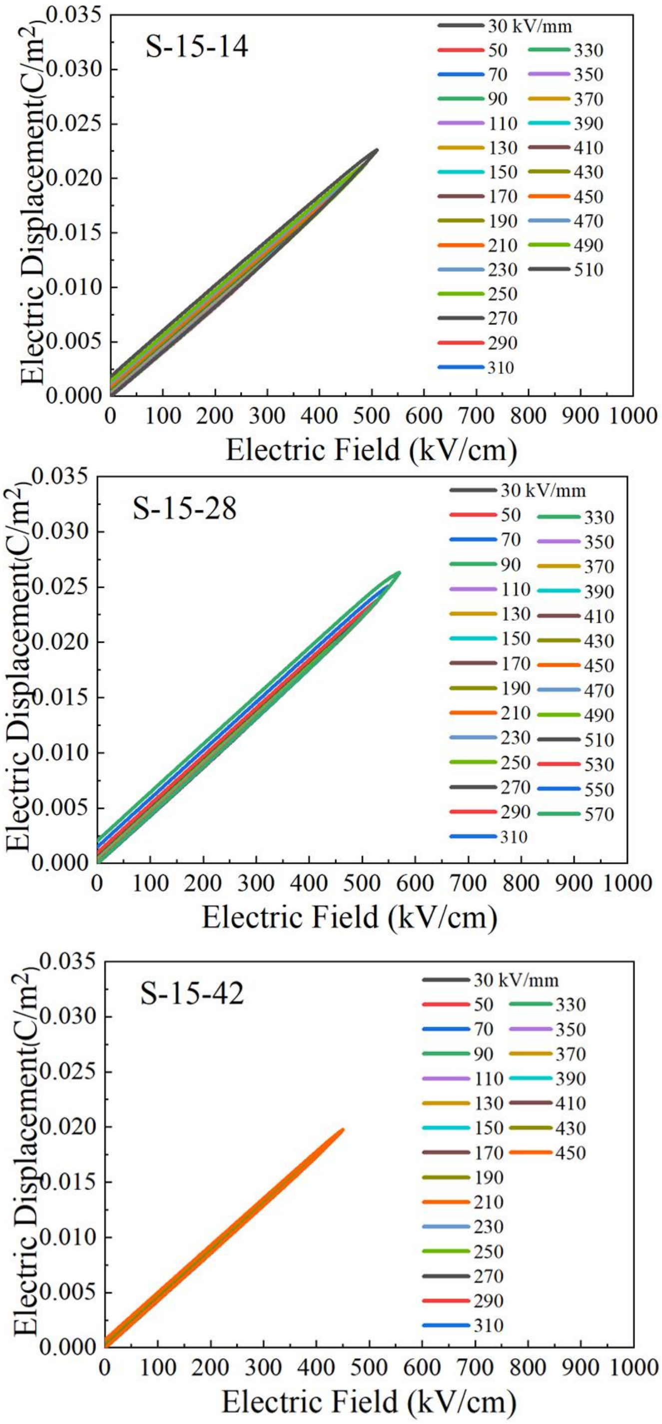

Figure S13 *D*-*E* loops for composite dielectrics with different trap layer locations at 150°C

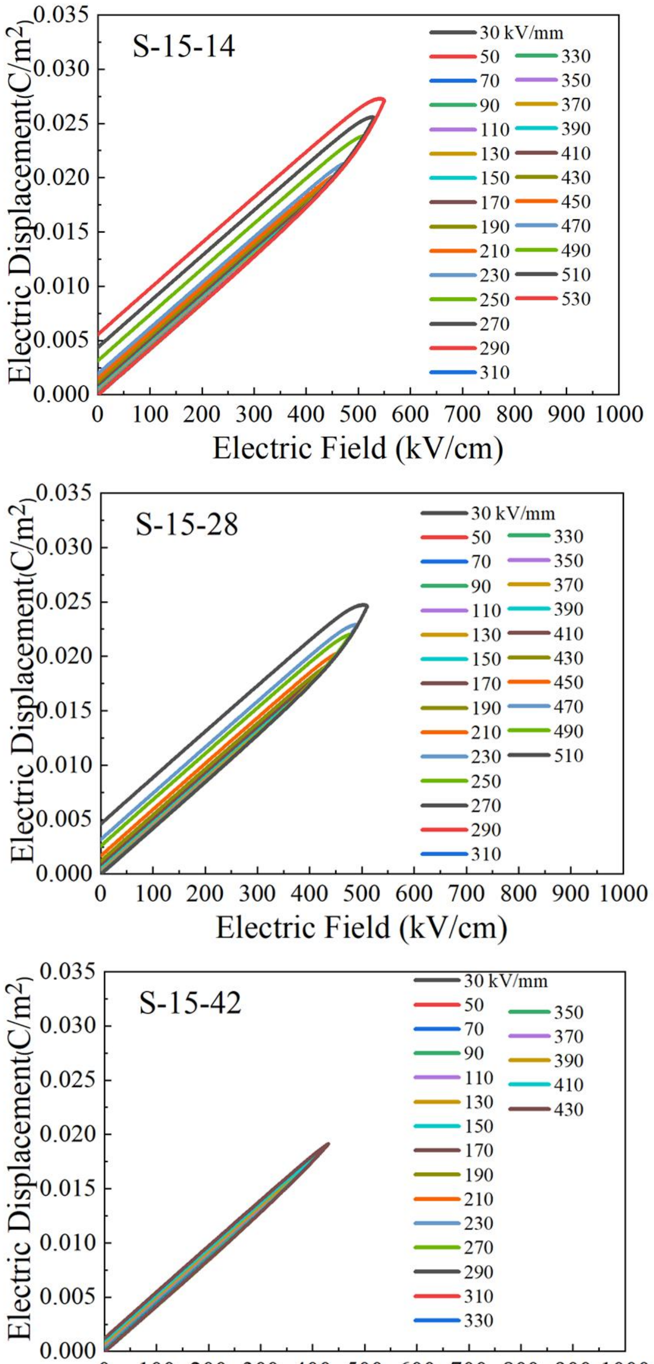

Figure S14 *D*-*E* loops for composite dielectrics with different trap layer locations at 180°C

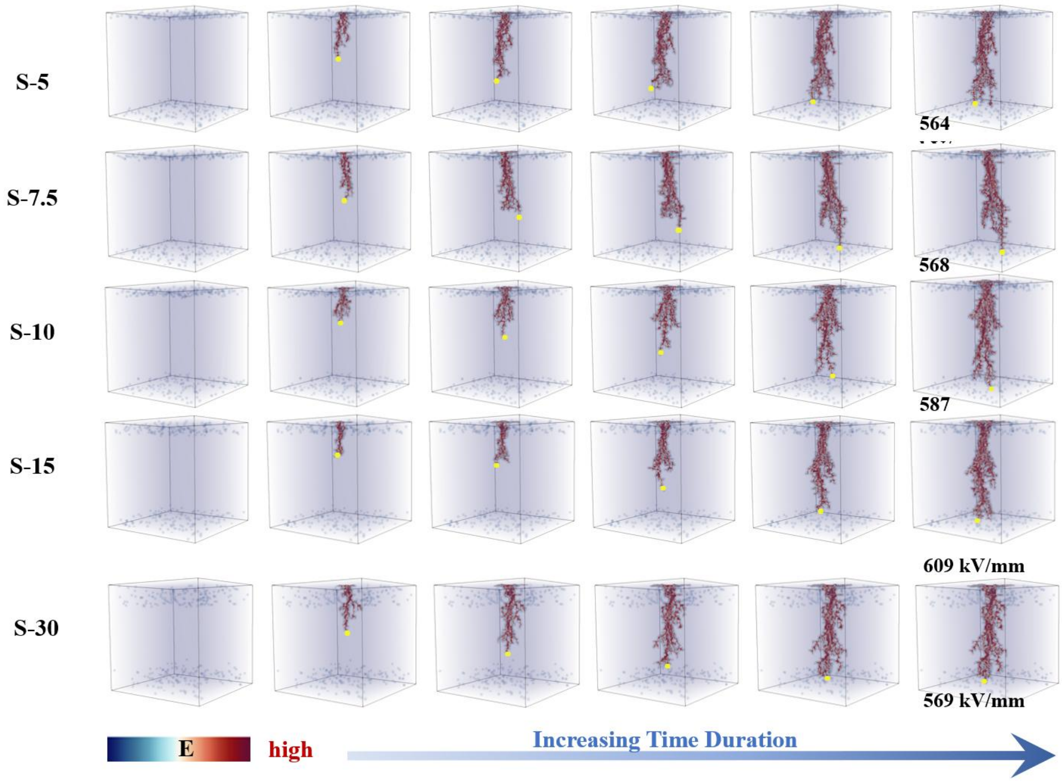

Figure S15 Phase-field simulation of breakdown process of composite dielectric with different trap densities at room temperature

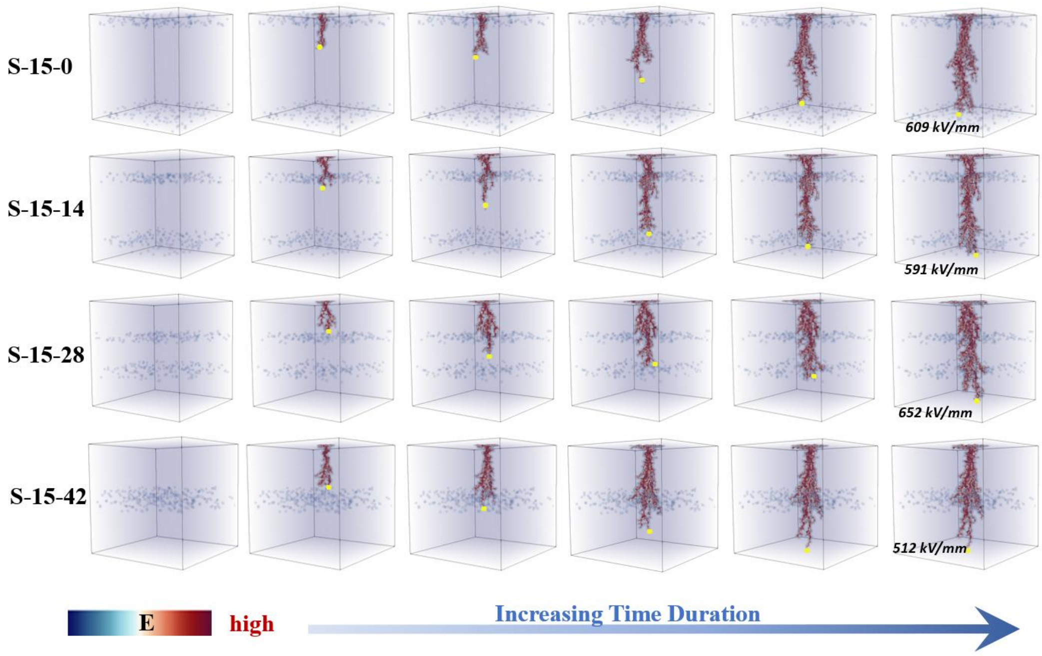

Figure S16 Phase-field simulation of breakdown process of composite dielectric with different trap layer locations at room temperature

Supplementary Notes:

Stochastic breakdown Model:

The stochastic model proposed by Niemeyer et al. was improved to study the microstructure effect on the breakdown strength.[1,2] In this model, the breakdown probability of a local point is defied by

$$P(r)=\frac{E(r)^2}{E_b(r)^2}\bigg/\sum\frac{E(r)^2}{E_b(r)^2}$$

where E(r) is the electric field of a local point determined by the externally applied voltage and the microstructure, and E_b(r) is the corresponding intrinsic breakdown strength determined by the materials, and the summation in the denominator is the sum over all points that the local electric field exceeds the breakdown strength.[3] The local electric field distribution is obtained by solving the electric equilibrium equation using an spectral iterative perturbation method.[4] The material parameters used in the simulation are listed in Table S1.

Table S1. Material parameters used in the stochastic modeling of breakdown.

Material	Relative Dielectric Constant	Intrinsic Breakdown Strength (kV/mm)
PEI	3.2	450
Doped Particle	-1	600

References

[1] L. Niemeyer, L. Pietronero, H. J. Wiesmann, Physical Review Letters 1984, 52, 1033.
[2] Y. Feng, J.-P. Xue, T.-D. Zhang, Q.-G. Chi, J.-L. Li, Q.-G. Chen, J.-J. Wang, L.-Q. Chen, Energy Storage Materials 2022, 44, 73.
[3] D. Yue, Y. Feng, X. X. Liu, J. H. Yin, W. C. Zhang, H. Guo, B. Su, Q. Q. Lei, Adv Sci 2022, e2105773.
[4] J. J. Wang, X. Q. Ma, Q. Li, J. Britson, L.-Q. Chen, Acta Materialia 2013, 61, 7591.