Gradient structured all-organic dielectrics by electrospinning for Enhanced energy storage performance

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Table S1. The solution flow rates (v) and the corresponding distribution functions of PVDF volume fraction (f_1) in all-organic composites with different topological structures

Topological configurations	v（ml/h）	The relationship between f_1 and d
Homogeneous structure	v_1=0.6 (0<t≤T) v_2=0.4 (0<t≤T)	f_1=60% (0<d ≤ 1)
Trilayer structure	v_1=1;v_2=0 (0<t≤3T/5) v_1=0;v_2=1 (0<t≤2T/5)	f_1=100% (0<d ≤ 1/3) f_1=0 (1/3<d ≤ 2/3) f_1=100% (2/3<d ≤ 1)
Linear gradient structure	v_1=1-0.8t/T (0<t≤T) v_2=0.8t/T (0<t≤T)	f_1=1-1.6d (0 ≤ d ≤ 0.5) f_1=1.6d-0.6 (0.5<d ≤ 1)
Nonlinear gradient structure	v_1=1.2-1.2t/T (0<t≤T) v_2=0.8t/T (0<t≤T)	$f_1 = 3 - \frac{6}{\sqrt{9 - 10d}}$ (0 ≤ d ≤ 0.5) $f_1 = 3 - \frac{6}{\sqrt{10d - 1}}$ (0.5 < d ≤ 1)
Rev-nonlinear gradient structure	v_1=1.2t/T (0<t≤T) v_2=0.8-0.8t/T (0<t≤T)	$f_1 = 3 - \frac{6}{\sqrt{10d + 4}}$ (0 ≤ d ≤ 0.5) $f_1 = 3 - \frac{6}{\sqrt{14 - 10d}}$ (0.5 < d ≤ 1)

* The subscripts of 1 and 2 represent PVDF and P(VDF-TrFE-CTFE), respectively.

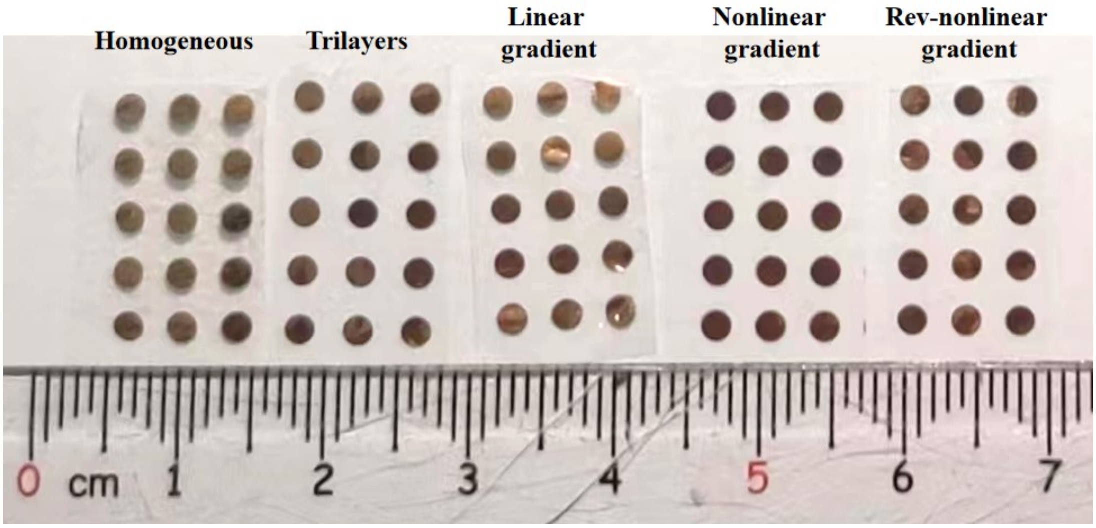
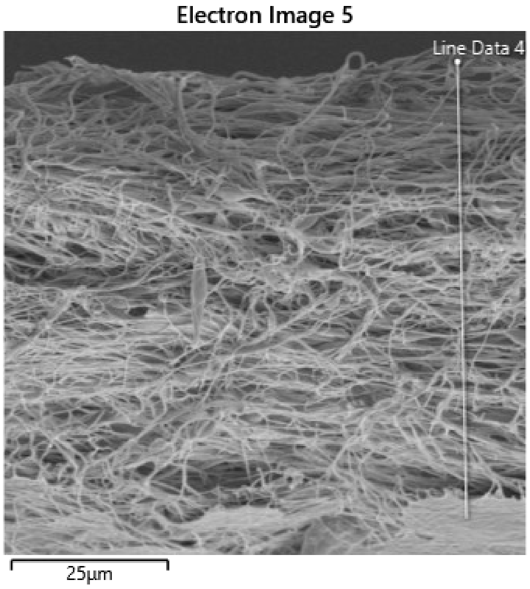


Figure S1. Photographs of composites with different topological structures that have been sputtered by gold electrodes



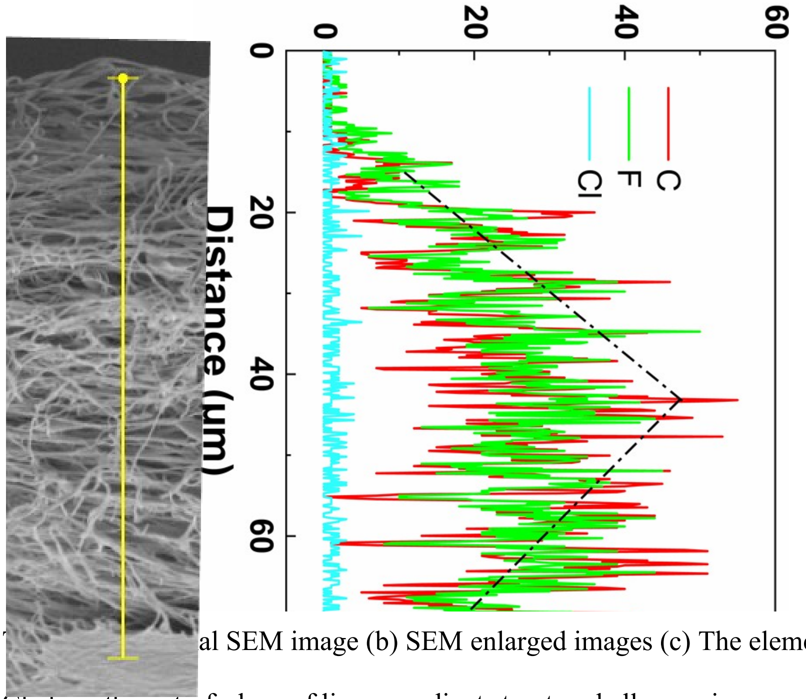


Figure S2. (a) T al SEM image (b) SEM enlarged images (c) The element mapping of C,F,Cl along the out-of-plane of linear gradient structured all-organic composite without hot pressing by electrospinning

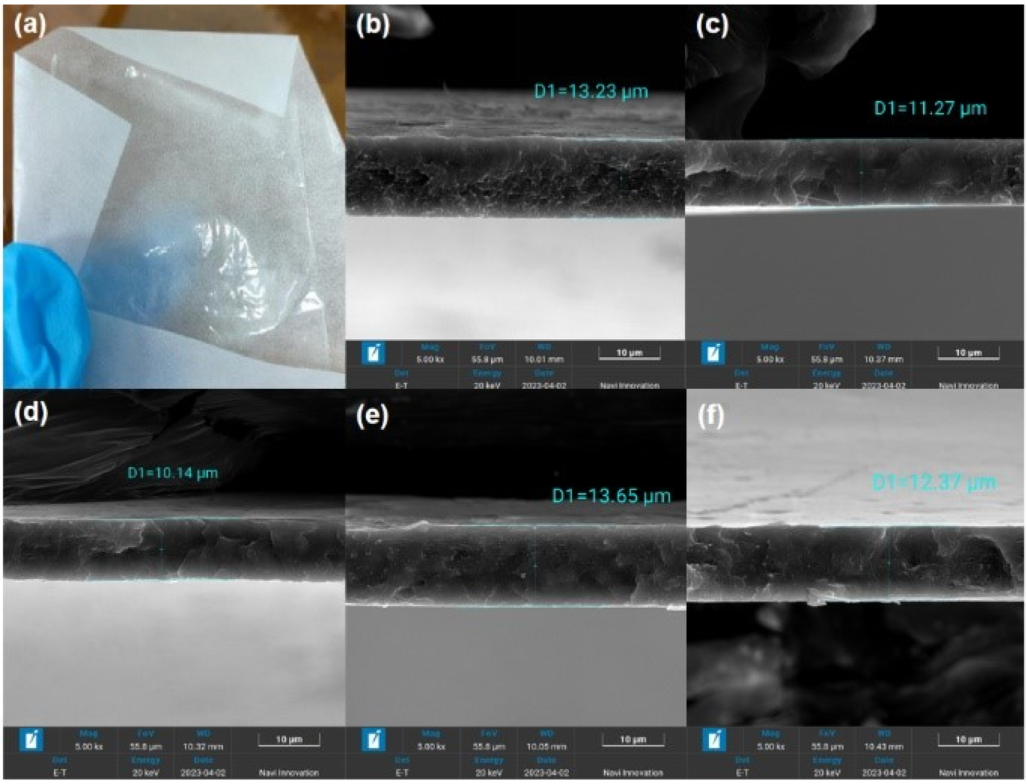


Figure S3. (a) Picture of transparent all-organic linear gradient structured composite obtained by hot-pressing; SEM images of composites cross-sections with different topological structures: (b) homogeneous structure, (c) trilayer structure, (d) linear gradient structure, (e) nonlinear gradient structure, (f) rev-nonlinear gradient structure

Table S2. The thermal properties and crystallinity of all-organic composites with different topological structures

T_{m1}(℃)	T_{m2}(℃)	$\triangle T_{m1}$(K)	$\triangle T_{m2}$(K)	$\triangle H_{m1}$(J/g)	$\triangle H_{m2}$(J/g)	χ_1(%)	χ_2(%)
167.99	122.99	441.25	396.14	24.94206	11.22575	39.78	10.69
167.89	124.89	440.65	398.04	23.06965	10.17609	36.79	9.69
166.89	123.89	440.05	397.04	23.21318	10.58457	37.02	10.08
167.86	125.86	441.05	399.01	23.49802	10.09086	37.48	9.61
168.86	124.86	442.45	398.01	23.68177	10.67103	37.77	10.16

* The subscripts of 1 and 2 represent PVDF and P(VDF-TrFE-CTFE), respectively.

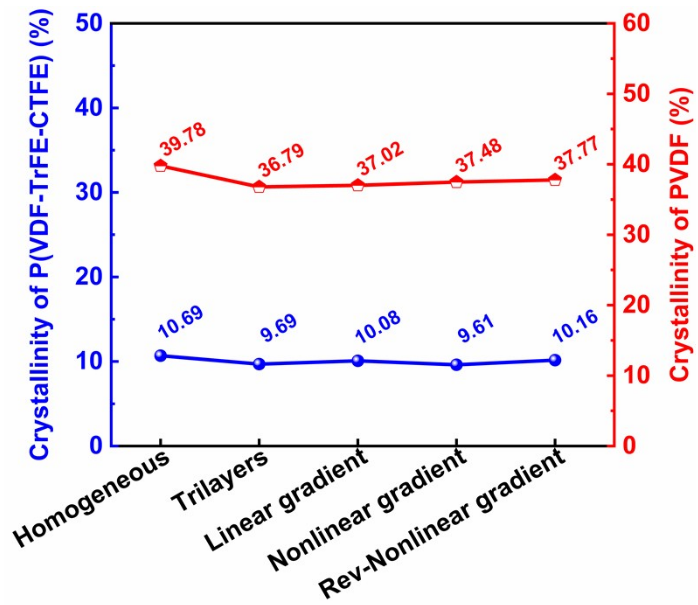


Figure S4. Crystallinity of P(VDF-TrFE-CTFE) and PVDF in polymer films with different topological structures

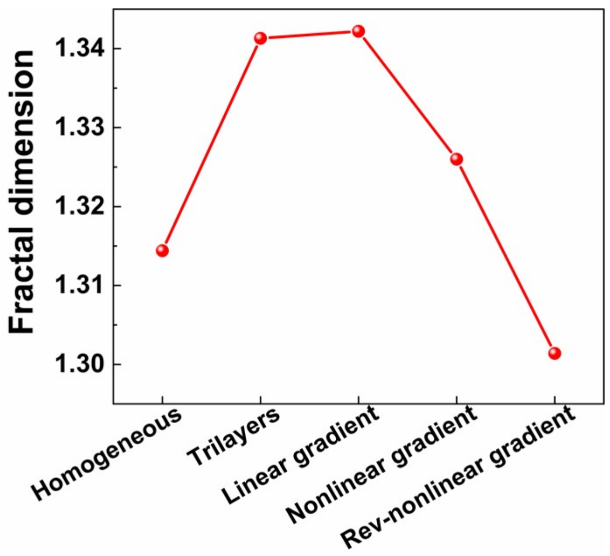


Figure S5. The fractal dimension of electric tree branches of composites with different topological structures

Table S3. Table of properties summary of all-organic composites with different topological structures

	Homogeneous	**Trilayers**	**Linear gradient**	**Nonlinear gradient**	**Rev-nonlinear gradient**
Dielectric constant @1 kHz	19.00	20.43	19.59	19.61	19.74
Dielectric loss @1 kHz	0.0396	0.0378	0.0385	0.0402	0.0415
D_{max} @390 kV/mm	7.34	7.97	7.62	7.67	7.79
D_r @390 kV/mm	1.83	1.14	0.96	1.15	1.30
D_{max}-D_r	5.51	6.83	6.66	6.52	6.49
E_{max}	390	410	540	420	400
E_b	366.5	376.5	477.8	395.2	354.2
β	13.1	13.5	10.6	13.3	4.8
Young's modulus	1.05	1.12	1.30	1.20	0.95
U_{dis}	8.12	11.47	17.75	11.51	10.26
η	57.67	72.26	70.26	70.85	66.82