Supporting Information

Optimizing Ply Pattern and Composition of Layered Composites based on Cyanate Ester, Carbon Nanotube, and Boron Nitride: Toward Ultralow Dielectric Loss and High Energy Storage

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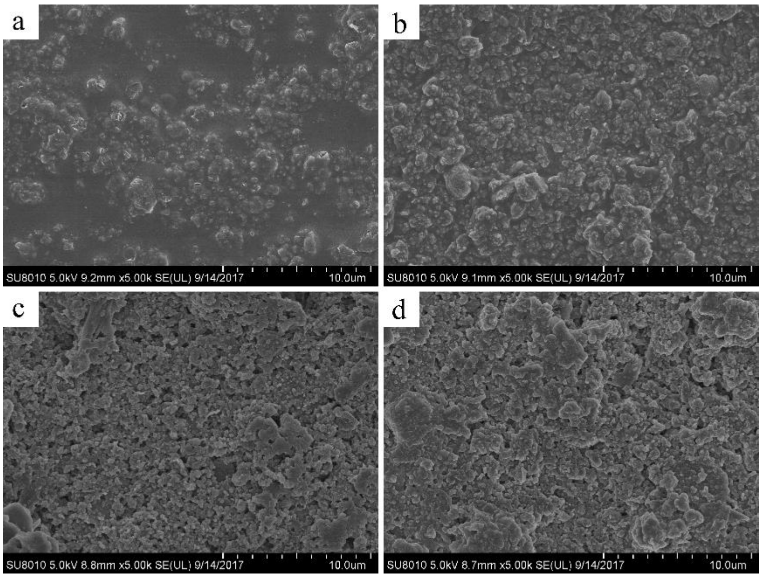

Figure S1 SEM pictures of B layer in multilayered structures (under magnification of 5k)

Table S1 Dielectric properties, breakdown strengths and energy densities of multilayered structure composites based on both conductor/polymer and insulating layers in literature. [a)]

Multi-layered structure [b)]	The loading of conductor (wt%) [c)]	Dielectric constant (100 Hz)	Dielectric loss (100 Hz)	Improvement of breakdown strength [d)]	Improvement of energy density [d)]	Ref
CNF/PVDF-PVDF-CNF/PVDF	3	~80	~0.2		/	S1
PE-CNT /CE	0.5	~170	~0.007	/	/	S2
$[PP\text{-}CB/PP]_{64}$	15	128	/	/	/	S3
$[PVDF\text{-}CB/PVDF]_{128}$	15	~67	0.05	/	/	S4
PVDF-CNT/PVDF-PVDF	2.2	32	0.08	/	/	S5
PI-CNT/PI-PI	10	~32	~0.0016	/		S6
GR/PVDF-BST/PVDF-GR/PVDF	20	~23	~0.08	~ -7.5%	~220%	S7
BT/PVDF-GR/PVDF-BT/PVDF	20.3	~92	~0.02	<28%	<650%	S8
CNT/CE-MP/CE-CNT/CE	0.6	219	0.4	~75%	~1550%	S9
CNT/CE-BN/CE-CNT/CE	0.4	323	0.049	55%	2410%	this work

a) Some properties not reported directly in the references are derived from the corresponding curves, so the symbol "~" was used to indicate the datum is an approximate value; the symbol "/" represents no data reported in the literature.

b) 64 and 128 are the repeat times of multilayered structure.
CNF: Carbon nanofiber.
CNT: Carbon nanotube.
CB: Carbon black.
GO: Graphene oxide.
GR: Graphite.
PVDF: Poly(vinylidene fluoride).
CE: Cyanate ester.
PP: Polypropylene.
PI: Polyimide.
BST: $Ba_{0.6}Sr_{0.4}TiO_3$.
BT: $BaTiO_3$.

c) The loading of conductor is the loading that multilayered structure obtained the highest dielectric constant at 100 Hz.

d) $$\text{The magnitude of improvement} = \frac{P_{\text{multi-layer structure}} - P_{\text{conductor/polymer}}}{P_{\text{conductor/polymer}}} \times 100\%$$
where, $P_{\text{multi-layer structure}}$ and $P_{\text{conductor/Polymer}}$ represent property (breakdown strength or energy density) of multi-layer structure and conductor/polymer, respectively.

References

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Table S2 Parameters of equivalent circuits for 0.4CNT/CE and BC composites.

	0.4CNT/CE	10BC	20BC	30BC	40BC
L (F)	1.47E-21	1.87E-22	9.43E-20	1.97E-17	1.05E-18
C (F)	7.36E-12	8.95E-11	4.62E-11	1.50E-19	5.32E-17
R_1 (Ω)	2.33E+05	1.15E+19	8.34E+05	1.46E+04	1.82E+04
CPE_1 (F)	1.01E-08	2.06E-05	2.70E-07	1.89E-10	3.82E-10
n_1	0.4889	0.9039	0.94529	0.9735	0.9611
R_2 (Ω)	2.01E+06	7.48E+12	1.23E+18	6.00E+18	8.19E+18
CPE_2 (F)	6.91E-11	4.06E-11	9.52E-11	9.72E-11	9.45E-11
n_2	0.9183	0.8091	0.8524	0.9021	0.9731
R_3 (Ω)	/	1143	4024	1454	1460
CPE_3 (F)	/	5.15E-10	6.52E-10	6.85E-10	2.74E-09
n_3	/	0.8128	0.785	0.8019	0.7223
R_t (Ω)	2.24E+06	1.15E+19	1.23E+18	6.00E+18	8.19E+18
C_t (F)	1.41E-11	1.69E-11	1.92E-11	1.89E-11	1.86E-11

Table S3 Parameters of equivalent circuits for BCB composites.

	10BCB	20BCB	30BCB	40BCB
L (F)	2.03E-16	4.73E-19	1.00E-20	1.50E-18
C (F)	1.73E-11	1.58E-11	2.17E-11	1.70E-11
CPE_1 (F)	3.84E-05	2.71E-05	1.03E-05	8.06E-05
n_1	0.1576	0.125	0.2295	0.2589
R_2 (Ω)	6.13E+21	5.34E+21	7.07E+21	9.00E+21
CPE_2 (F)	3.94E-11	7.95E-11	2.20E-11	2.90E-11
n_2	0.9885	0.9875	0.9902	0.9889
R_3 (Ω)	155.7	182.1	181.1	19.18
R_t (Ω)	6.13E+21	5.34E+21	7.07E+21	9.00E+21
C_t (F)	1.31E-11	1.39E-11	1.23E-11	1.18E-11

Table S4 Parameters of equivalent circuits for CBC composites.

	10CBC	20CBC	30CBC	40CBC
L (F)	1.87E-16	4.71E-15	1.06E-06	9.15E-07
C_1 (F)	1.46E-10	1.89E-17	1.34E-11	3.35E-09
R_1 (Ω)	1.62E+06	1.70E+05	3.23E+08	2.77E+06
CPE_1 (F)	1.07E-05	2.59E-10	0.0001801	1.40E-14
n_1	0.03089	0.9045	0.09782	0.1039
R_2 (Ω)	3577	3622	874.2	3673
CPE_2 (F)	8.15E-10	9.06E-09	1.83E+09	1.23E-10
n_2	0.8195	0.7438	0.9843	0.9708
R_3 (Ω)	3.27E+04	9.17E+05	4.06E+04	5.43E+04
CPE_3 (F)	4.94E-09	7.86E-09	1.58E-10	8.43E-11
n_3	0.771	1	0.9176	0.9718
R_4 (Ω)	3.70E+08	1.33E+09	2.18E+6	1.08E+08
C_2(F)	2.50E-10	3.71E-10	2.57E-10	2.99E-10
R_t (Ω)	3.71E+08	1.33E+09	3.25E+08	1.11E+08
C_t (F)	3.70E-11	5.61E-11	4.46E-11	3.01E-11