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Supporting Information

Boost up Dielectric Constant and Push down Dielectric Loss of Carbon Nanotube/Cyanate Ester Composites through Gradient and Layered Structure Design

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S1. Dielectric properites of MWCNT/CE composites

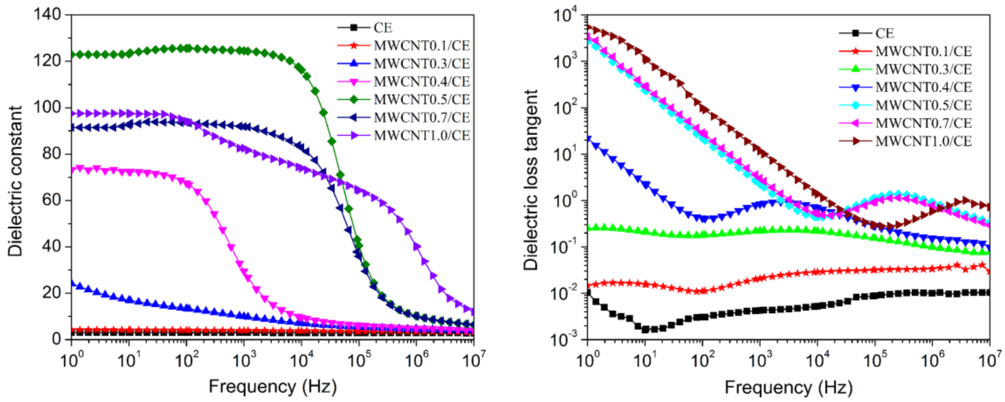

Figure S1. The real dielectric constant and loss tangent as a function of frequency for CE resin and MWCNT/CE composites with different loadings of MWCNTs

S2. The percolation threshold of MWCNT/CE composites

According to the percolation theory, there is a universal relationship between the AC conductivity (σ) and the loading of conductor (f) as shown in **Equation S1**.

$$\sigma \propto (f - f_c)^t, f > f_c \tag{S1}$$

where f_c is the percolation threshold, t is the critical conductivity exponent.

Using a least-squares fit for repeated experiments as shown in the inset plot of Figure S2b, the f_c value of MWCNT/CE composites was calculated to be 0.39 wt %.

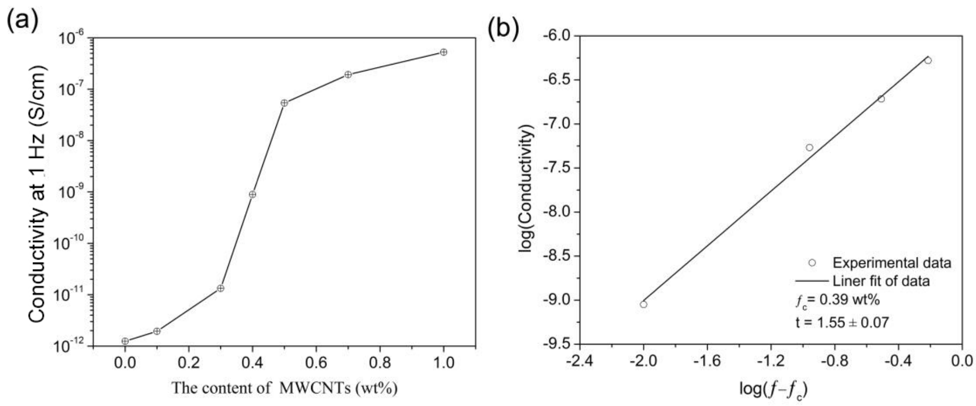

Figure S2. (a) The AC conductivity at 1 Hz as a function of MWCNTs concentration up to 1.0 wt% of MWCNT/CE composites. (b) The experimental data and linear fit of $\log(\text{Conductivity})$ vs $\log(f - f_c)$ for MWCNT/CE composites.

S3. Conductivity

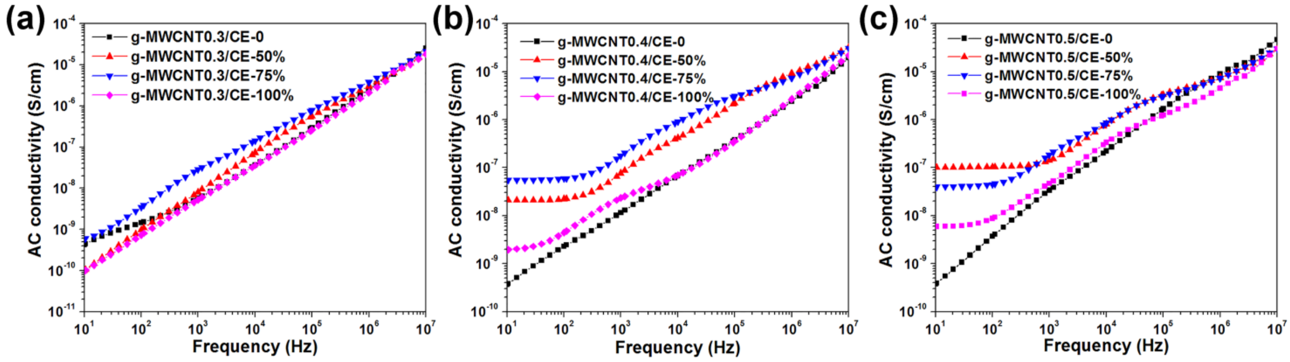

Figure S3. The AC conductivities of (a) g-MWCNT0.3/CE-x%, (b) g-MWCNT0.4/CE-x% and (c) g-MWCNT0.5/CE-x% nanocompoistes over a wide frequency range.

S4. SEM images

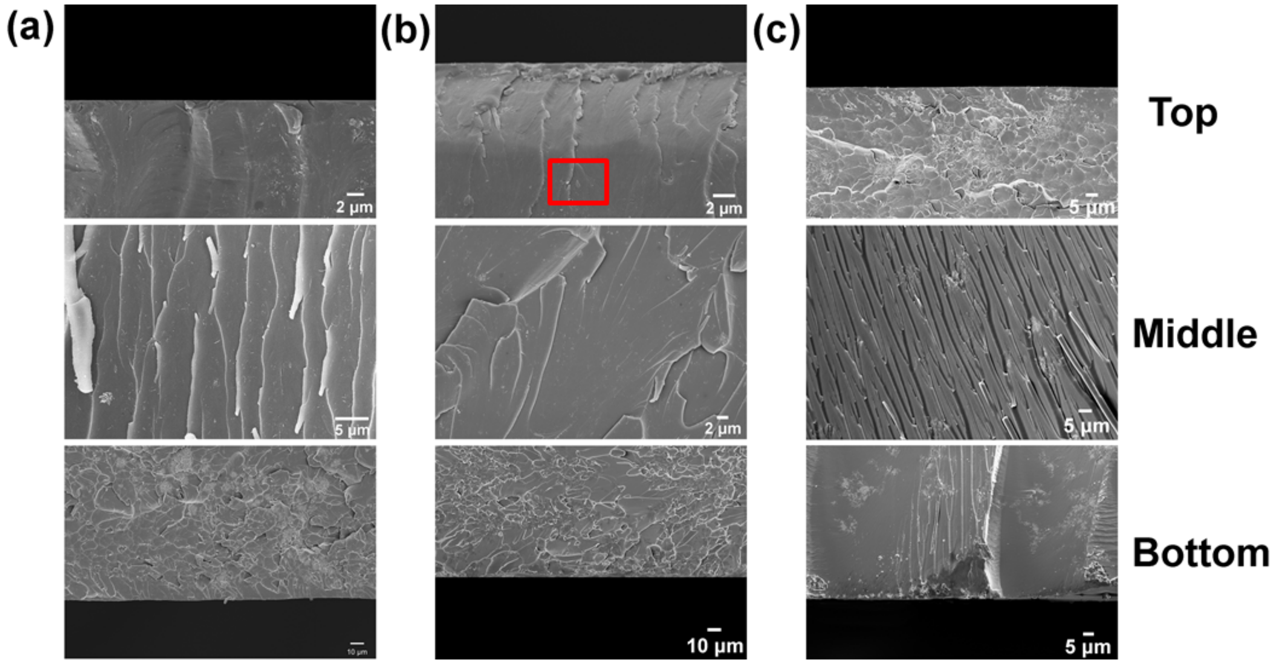

Fig. S4 The SEM images of the top, middle, and bottom parts of the cross-sections along the thickness direction of (a) g-MWCNT0.5/CE-0, (b) g-MWCNT0.5/CE-75% and (c) g-MWCNT0.5/CE-100% composites.

S5. Schematic diagrams and SEM images

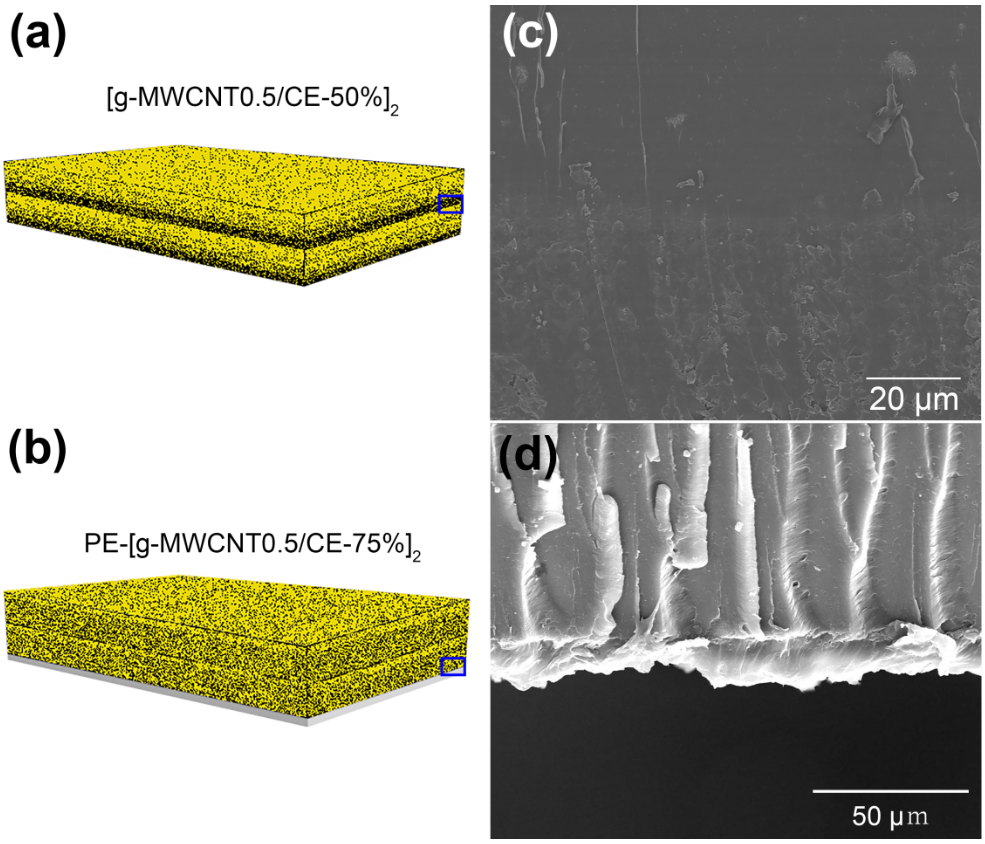

Figure S5. The schematic diagrams of [g-MWCNT0.5/CE-50%]$_2$ (a) and PE-[g-MWCNT0.5/CE-75%]$_2$ (b) materials. (c) and (d) are the SEM images of the interfaces between two layers labelled by blue squares in (a) and (b), respectively. The PE layer has a thickness of ~7 μm.

Table S1. The parameters derived from TG and DTG curves.

Sample	T_{di} (°C)	T_{max} (°C)	
		T_{max1} (°C)	T_{max2} (°C)
Top of MWCNT0.3/CE-0	339.0	427.0	602.6
Bottom of MWCNT0.3/CE-0	395.9	433.5	606.4
Top of MWCNT0.4/CE-0	331.0	427.7	605.6
Bottom of MWCNT0.4/CE-0	402.9	434.8	611.9
Top of MWCNT0.5/CE-0	350.3	431.0	608.9
Bottom of MWCNT0.5/CE-0	407.7	434.1	620.3
MWCNTs	468.3	562.9	

Table S2. Key parameters of high-*k* polymer composites with low dielectric loss. a)

Filler b)	Filler loading	Polymer matrix c)	$\varepsilon_{p,\max}$ ($\tan\delta_p$)	$\varepsilon_{c,\max}$	$\tan\delta_c$	η d)	Reference
Hydrazine reduced GO	1.7 vol%	P(VDF-TrFE-CFE)	57 (0.05)	10000	2	4.39	[S1]
Graphene-TiO_2 sheets	10.9 vol%	PS	2.7 (0.004)	1741	0.39	6.61	[S2]
MWCNTs	0.5 wt%	CE	3.1 (0.004)	306	0.21	4.7	[S3]
Chlorination reduced GO	0.5 wt%	CEP	17 (0.04)	169	0.05	7.95	[S4]
MWCNTs	0.5 wt%	CE	3.1 (0.004)	168	0.006	36.13	[S5]
Rutile rods	36.9 vol%	PS	2.0 (0.008)	80	0.14	2.29	[S6]
Graphene	0.17wt%	CEP	17 (0.09)	77	0.16	2.55	[S7]
Graphene nanosheets	4.0 vol%	P(VDF-TrFE-DB)	12 (0.04)	74	0.08	3.08	[S8]
Carbon nanofibers	2.5 wt%	PVDF	13 (0.13)	68	0.06	11.33	[S9]
MWCNTs	25 vol%	PSF	5 (0.02)	58	0.05	4.64	[S10]
PPy coated MWCNTs	10 vol%	PS	2.7 (0.004)	44	0.07	0.93	[S11]
rGO-MWCNTs	0.062 wt%	CEP	15 (0.036)	32	0.051	1.61	[S12]
$BaTiO_3$	10.8 vol%	PVDF-TrFE	14 (0.02)	30	0.03	1.43	[S13]
MWCNTs	0.34 vol%	PP	2.4 (0.002)	30	0.06	0.42	[S14]
GO	5.0 wt%	PDMS	3.1 (0.01)	8	0.005	5.16	[S15]
MWCNTs	0.5 wt%	CE	3.1 (0.004)	1027	0.017	77.95	This work

a) The data are arranged with decreasing dielectric constant of the composites. Some parameters not reported directly in the references are derived from the corresponding curves. $\varepsilon_{p,\max}$: The maximum dielectric constant of polymer matrix. $\tan\delta_p$: dielectric loss tangent of polymer matrix at corresponding frequencies. $\varepsilon_{c,\max}$: The maximum dielectric constant of the composites. $\tan\delta_c$: dielectric loss tangent of the composites at corresponding frequencies.

b) MWCNTs: Multi-wall carbon nanotubes, PPy: Polypyrrole, GO: Graphene oxide, $BaTiO_3$: Barium titanate.
c) P(VDF-TrFE-CFE): Poly(vinylidene fluoride-co-trifluoroethylene- co-chlorofluoroethylene); PS: Polystyrene; CE: Cyanate ester; CEP: Cyanoethyl pullulan polymer; P(VDF-TrFE-DB): Poly-(vinylidenefluoride-co-trifluorethylene) with double bonds; PVDF: Poly(vinylidene fluoride); PSF: Polysulfone; PVDF-TrFE: Poly(vinylidene fluoride-co-trifluoroethylene); PP: Polypropylene; PDMS: Poly(dimethyl siloxane).
d) η is defined as the proportion of the variation of dielectric constant to the variation of dielectric loss tangent, shown in **Equation S2**.

$$\eta = \varepsilon_{c,\max} \tan\delta_p / \varepsilon_{p.\max} \tan\delta_c \qquad \text{(S2)}$$

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