## **Electronic Supporting Information**

## Optimizing Electric Field Distribution via Tuning Cross-linked Points Size for Improving Dielectric Properties of Polymer Nanocomposites

Jie Liu<sup>a,b</sup>, Yu Zhang<sup>a</sup>, Zhaoyang Wang<sup>a</sup>, Jiale Ding<sup>a</sup>, Shuhui Yu<sup>b</sup>, Yunhe Zhang<sup>a\*</sup> Zhenhua Jiang<sup>a</sup>

<sup>a</sup>Engineering Research Center of Super Engineering Plastics, Ministry of Education, College of Chemistry, Jilin University, Changchun, 130012, P. R. China.

<sup>b</sup>Shenzhen Institute of Advanced Electronic Materials, Shenzhen Institutes of Advanced Technology, Chinese Academy of Sciences, Shenzhen 518055, P. R. China

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\*Corresponding Authors: E-mail: zhangyunhe@jlu.edu.cn (Yunhe Zhang); E-mail:

sh.yu@siat.ac.cn (Shuhui Yu).



Fig. S1. FT-IR spectra of core-shell BT-BCB nanoparticles.

**Fig. S1** shows FT-IR spectra of BT, BT-MPS, and BT-BCB in the 500-4000 cm<sup>-1</sup> region. Compared with pure nano-BT, the new absorption bands that appeared at 2925 and 2859 cm<sup>-1</sup> in the spectrum of BT-MPS nanoparticles are mainly attributed to the bending vibration of -CH<sub>3</sub> and -CH<sub>2</sub> stretching vibration in the coupled agent of MPS, respectively. Moreover, the peaks at about 990 cm<sup>-1</sup> in the spectrum of BT-MPS nanoparticles are assigned as the characteristic absorptions of vinyl groups. In addition to the characteristic peak of MPS, the absorption bands located at about 1475 cm<sup>-1</sup> are attributed to the in-plane ring stretching vibration of C-H in the strained four-membered ring in the spectrum of BT-MPS nanoparticles, which suggests that the successful coating of BCB groups onto nano-BT surfaces.

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Fig. S2. TGA curves of 60 nm, 100 nm and 500 nm BT-BCB nanoparticles.



**Fig. S3.** SEM images of 60 nm BT (a), 100 nm BT (b) and 500 nm BT (c); XRD data of 60 nm, 100 nm and 500 nm nano-BT.



**Fig. S4.** Compared XRD data of 60 nm nano-BT and BT-BCB nanoparticles (a), 100 nm nano-BT and BT-BCB nanoparticles (b), and 500 nm nano-BT and BT-BCB nanoparticles (c).



**Fig. S5.** FT-IR spectra of DPAES and BT-BCB@DPAES composites with 60 nm, 100 nm and 500 nm BT-BCB nanoparticles.



Fig. S6. SEM images of BT/DPAES with 60 nm (a,d), 100 nm (b,e) and 500 nm (c,f) nano-BT.



**Fig. S7.** XRD data of DPAES and BT-BCB@DPAES composites with 60 nm, 100 nm and 500 nm BT-BCB nanoparticles.



**Fig. S8.** TGA curves of DPAES and BT-BCB@DPAES composites with 60 nm, 100 nm and 500 nm BT-BCB nanoparticles.

**Tab. S1** The scale parameter ( $\alpha$ ) and the shape parameter ( $\beta$ ) from the Weibull statistics of DPAES and the BT-BCB@DPAES nanocomposites with varied diameters nanoparticles at RT and 150 °C.

	RT ª		150 °C <sup>b</sup>	
	α (MV m⁻¹)	β	α (MV m <sup>-1</sup> )	β
DPAES	472	11.9	412	7.5
60 nm BT-BCB@DPAES	405	10.0	362	8.2
100 nm BT-BCB@DPAES	457	12.9	442	11.2
500 nm BT-BCB@DPAES	370	8.2	348	6.2

**a**: the data of  $\alpha$  and  $\beta$  at room temperature obtained by the linear fit to **Figure 7a**;

**b**: the data of  $\alpha$  and  $\beta$  at 150 °C obtained by the linear fit to **Figure 7b**.



**Fig. S9.** The percent change of breakdown strength of DPAES and BT-BCB@DPAES composites with 60 nm, 100 nm and 500 nm BT-BCB nanoparticles from room temperature to 150 °C.



**Fig. S10.** 3D distribution of local electric field in the BT-BCB@DPAES nanocomposites with 60 nm (a), 100 nm (b), and 500 nm (c) BT-BCB nanopartcles. The corresponding probability distribution function of the normalized local electric field in the nanocomposites at room temperature (d). The normalized local electric field is defined as the ratio of the computed local electric field to the applied electric field.



**Fig. S11.** Discharged energy density (a) and charge-discharge efficiency (b) of BT-BCB@DPAES nanocomposites with different BT-BCB nanoparticles at room temperature, respectively.



Fig. S12. Leakage current density (a) and DC electric resistivity (b) of the BT-BCB@DPAES nanocomposites with 60 nm, 100 nm and 500 nm BT-BCB nanoparticles at 150  $^{\circ}$ C and 50 MV m<sup>-1</sup>.