Supporting Information

High loading BaTiO3 nanoparticles chemically bonded with fluorinated silicone rubber for largely enhanced dielectric properties of polymer nanocomposites

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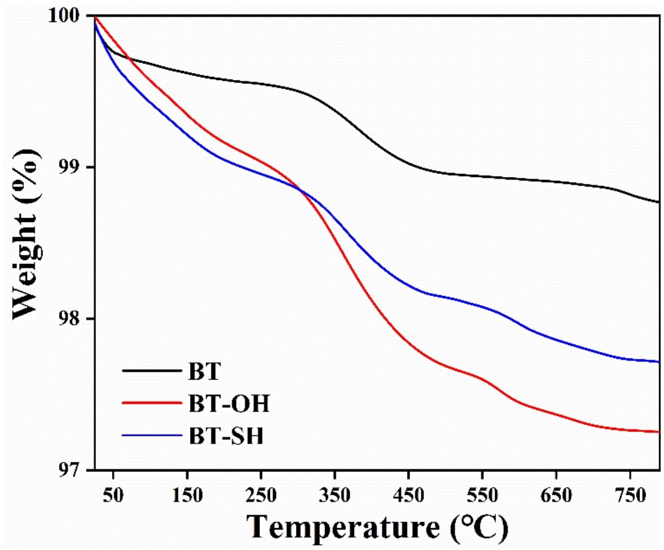

Figure S1. TGA spectrum of the BT, BT-OH and BT-SH nanoparticles.

Table S1 TG data and grafting density of BT. BT-OH and BT-SH

	Weight loss	**Grafting ratio/ (group nm^{-2})**
BT	**1.2%**	**-**
BT-OH	**2.1%**	**32.6**
BT-SH	**2.7%**	**10.3**

Weight loss of BT, BT-OH and BT-SH nanoparticles are determined by TGA, as shown in **Figure S1** and **Table S1**. According to the method in literature [3], the grafting density of BT-OH and BT-SH nanoparticles are calculated based on the weight loss and molar mass of grafting groups. Considering each nanoparticle as a sphere with d=100nm, and product of KH580 after decomposition as neat SiO_2. The molar mass of hydroxyl groups, KH580, SiO_2 and $BaTiO_3$ are 17 g mol^{-1}, 238 g mol^{-1}, 60 g mol^{-1} and 233 g mol^{-1}, respectively. The structure of BT unit cell is cubic with a=b=c=0.395nm.

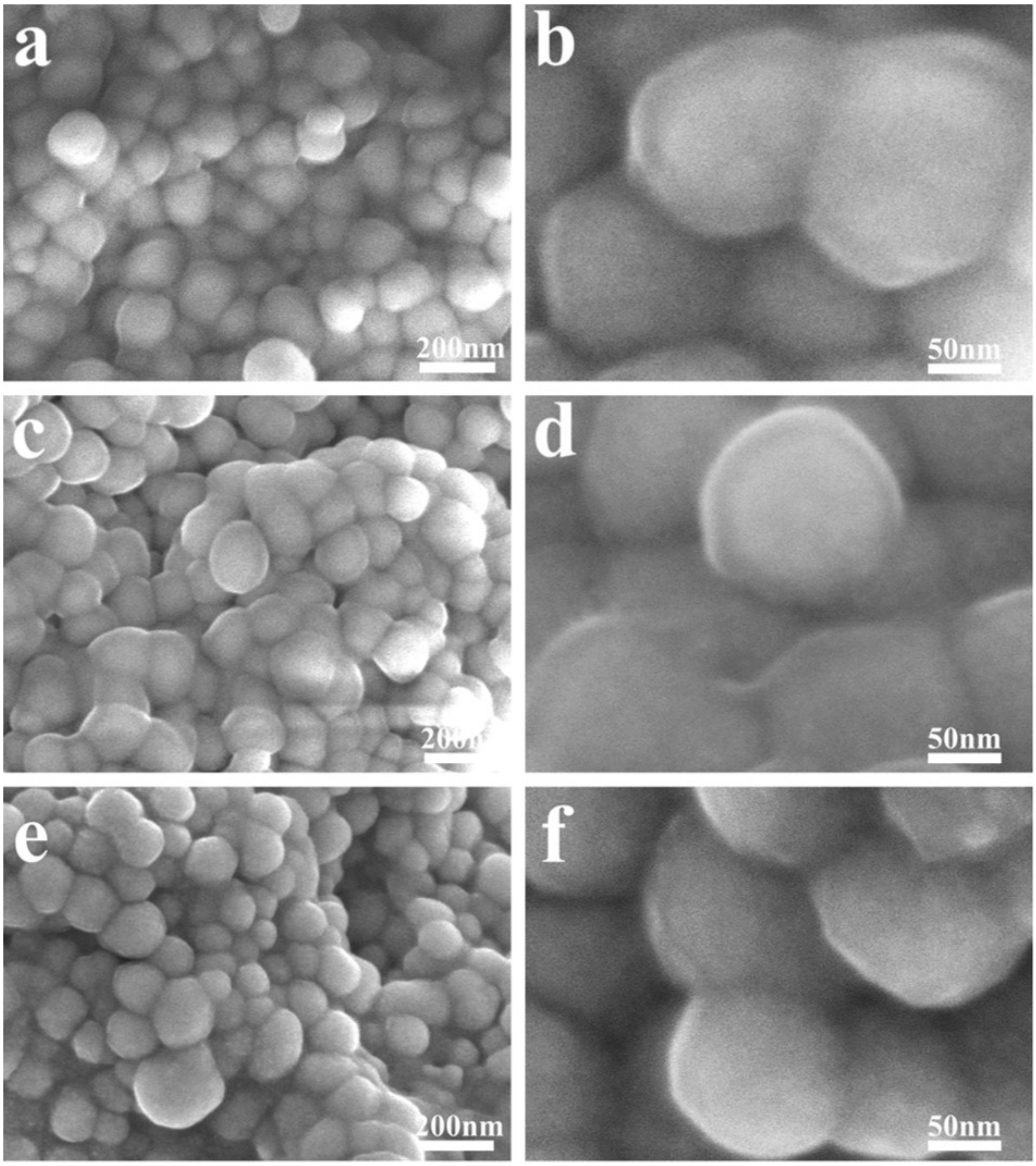

Figure S2 SEM images of the BT/FSR nanocomposites with the loading of 82 wt.% (a, b), 86 wt.% (c, d) and 90 wt.% (e, f).

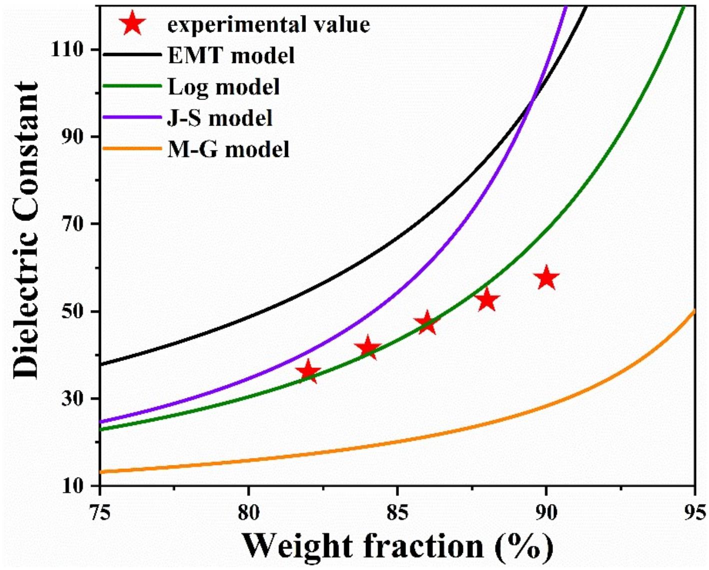

Figure S3. Plots of dielectric constants versus weight fraction (*ρ_{BT}=6.08g/cm^3*, *ρ_{FSR}=1.2g/cm^3*) with various models for polymer-based composites. For calculation, dielectric constants of BT (*ε=300*) and FSR (*ε=5*) were applied as dielectric constants of filler and matrix. The shape factor (*n*=0.09) in EMT model was obtained from BT/VSR composites in former study.

Table S2 Theoretical models for predicting the dielectric constants of polymer composites

Models	Equations
EMT model	$\varepsilon = \varepsilon_M \left[1 + \frac{vol_F\% \, (\varepsilon_F - \varepsilon_M)}{\varepsilon_M + n \, vol_M\% \, (\varepsilon_F - \varepsilon_M)} \right]$ [1]
Log model	$\varepsilon = e^{vol_F\% \ln \varepsilon_F + vol_M\% \ln \varepsilon_M}$ [2]
J-S model	$\varepsilon = \frac{\varepsilon_M (1 - vol_F\%) + \varepsilon_F vol_F\% \frac{3\varepsilon_M}{\varepsilon_F + \varepsilon_M} \left[1 + \frac{3vol_F\%(\varepsilon}{\varepsilon_F +} \right.}{(1 - vol_F\%) + vol_F\% \frac{3\varepsilon_M}{\varepsilon_F + \varepsilon_M} \left[1 + \frac{3vol_F\%(\varepsilon_F}{\varepsilon_F + \varepsilon_M} \right.}$ [2]
M-G model	$\varepsilon = \varepsilon_M \frac{\varepsilon_F (2vol_F\% + 1) + 2\varepsilon_M (1 - vol_F\%)}{\varepsilon_F (1 - vol_F\%) + \varepsilon_M (2 + vol_F\%)}$ [2]

Figure S3 shows the experimental dielectric constants of BT/FSR nanocomposites attached with expectation value of a series of theoretical models. A phenomenon can be noticed that the EMT model fails to match with the experimental value, while the Log and J-S model show good agreement.

Reference

[1] C. P. Wong, T. Marinis, Q. Jianmin and R. Yang, IEEE Transactions on Components and Packaging Technologies, 2000, 23, 680-683.

[2] S. Luo, Y. Shen, S. Yu, Y. Wan, W.-H. Liao, R. Sun and C.-P. Wong, Energy & Environmental Science, 2017, 10, 137-144.

[3] K. Yang, X. Huang, M. Zhu, L. Xie, T. Tanaka and P. Jiang, ACS Appl. Mater. Interfaces, 2014, 6, 1812-1822.