Supporting Information

Significantly enhanced permittivity and energy density in dielectric composites

with aligned BaTiO₃ lamellar structures

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1. The theoretical model ^[1-4] for predicting effective permittivity:

Parallel model: $\varepsilon_{eff} = v_1 \varepsilon_1 + v_2 \varepsilon_2$ eq.S1

$$\varepsilon_{eff} = \frac{\varepsilon_1 \ \varepsilon_2}{\nu_1 \varepsilon_2 + \nu_2 \varepsilon_1}$$

Series model:

Lichtenecker model: $\varepsilon_{eff}^{k} = v_1 \varepsilon_1^{k} + v_2 \varepsilon_2^{k}$ eq.S3

$$\varepsilon_{eff} = \frac{\nu_2 \varepsilon_2 + \nu_1 \varepsilon_1 \frac{3\varepsilon_2}{\varepsilon_1 + \varepsilon_2} [1 + \frac{3\nu_1(\varepsilon_1 - \varepsilon_2)}{\varepsilon_1 + \varepsilon_2}]}{\nu_2 + \nu_1 \frac{3\varepsilon_2}{\varepsilon_1 + \varepsilon_2} [1 + \frac{3\nu_1(\varepsilon_1 - \varepsilon_2)}{\varepsilon_1 + \varepsilon_2}]}$$
eq

eq.S2

.S4

Jayasundere-Smith model:

$$\varepsilon_{eff} = \varepsilon_2 \frac{(2\nu_1 + 1)\varepsilon_1 + 2(1 - \nu_1)\varepsilon_2}{(1 - \nu_1)\varepsilon_1 + (2 + \nu_1)\varepsilon_2}$$
eq.S5

Maxwell-Garnett model:

Where ε_1 and ε_2 are the permittivity of fillers and polymer matrices; v_1 and v_2 are the volume fraction of fillers and polymer matrices, respectively. ε_{eff} is the effective permittivity of the composites.

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- [3]. S. Luo, Y. Shen, S. Yu, Y. J. Wan, W. H. Liao, R. Sun, C. Wong, Energy & Environmental Science, 2017, 10, 137-144.
- [4]. S. K. Dong, C. Baek, H. J. Ma, D. K. Kim, Ceramics International, 2016, 42, 7141-7147.

The structure and composition information of $BaTiO_3$ were shown in Fig.S1 (a) that $BaTiO_3$ had the single (111) diffraction peak, split (002) / (200) peak. It is a typical tetragonal structure with space group P4mm and the PDF order of $BaTiO_3$ is #05-0626.



Figure S1. (a) The XRD patterns of raw BaTiO₃ powder about structure and composition information. (b) Comparison of XRD patterns of epoxy resin, raw BaTiO₃ powder, sintered porous BaTiO₃ ceramics and BaTiO₃/epoxy composites.



Figure S2. SEM images of the $BaTiO_3$ porous ceramics in the parallel freezing direction with varying $BaTiO_3$ volume fractions, (a) 24 vol.%, (b) 40 vol.%, (c) 59 vol.%, respectively.



Figure S3. The SEM of composites with BaTiO₃ particles randomly distributed in

epoxy resin as comparison.



Figure S4. The histograms of porosity and relative density of the porous BaTiO₃ ceramics measured by Archimedes method corresponding to the BT volume fraction.

The dielectric properties as a function of temperature for the BaTiO₃/epoxy composites were shown in Fig S5. Temperature dependence of permittivity have been measured from 25°C to 150°C. It is observed in Fig. S5(a) that the curie temperature of BaTiO₃ ceramic is about 125°C, and at this temperature, the permittivity of BaTiO₃ reaches the maximum value; As shown in Fig. S5 (b), the permittivity of the resin appears a hump in the temperature ranges. For BaTiO₃/epoxy composites, the permittivity-temperature curve combines the change trend of ceramic and epoxy resin. The permittivity of the composites remains relatively high in the whole temperature range, and which reaches the largest value at the curie temperature.



Figure S5. Temperature dependence of permittivity measured from 25°C to 150°C of (a) BaTiO₃ ceramic; (b) Epoxy resin; and BaTiO₃/Epoxy composite with (c) BT-Random-59, (d) BT- \pm -59, (e) BT-//-59.

As shown in Fig S6. The bending strength of composite BT-random-59, BT-//-59, BT- \perp -59 are 55.79, 68.21, 75.45MPa, respectively, which were higher than that of the BaTiO₃ ceramic and epoxy resin of 30.12 and 44.60 MPa, respectively. This result may be attributed to the deformation space of the interface between BaTiO₃ and epoxy resin layer in the laminar structure under external loading, compare with the single ceramic



or epoxy resin.

Figure S6. Comparison of bending strength of $BaTiO_3$ ceramic, epoxy resin and $BaTiO_3$ /epoxy composite with BT-Random-59, BT- \perp -59, and BT-//-59.