Supporting Information

Significantly enhanced permittivity and energy density in dielectric composites with aligned BaTiO$_3$ lamellar structures

Ru Guo$^a$ $\triangleright$, James Roscow$^b$ $\triangleright$, Chris R. Bowen$^b$, Hang Luo$^a$*, Yujuan Huang$^a$, Yupeng Ma$^a$, Kechao Zhou$^a$ and Dou Zhang$^a$*

R. Guo, Dr. H. Luo, Y.J. Huang, Y.P Ma, Prof. K.C. Zhou and Prof. D. Zhang
State Key Laboratory of Powder Metallurgy,
Central South University,
Changsha 410083, Hunan Province, China
E-mail: dzhang@csu.edu.cn, hangluo@csu.edu.cn

Dr. J. Roscow, Prof. C. Bowen
Department of Mechanical Engineering
University of Bath
Claverton Down, Bath BA2 7AY, UK
1. The theoretical model [1-4] for predicting effective permittivity:

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

Series model:  \( \varepsilon_{\text{eff}} = \frac{\varepsilon_1 \varepsilon_2}{v_1 \varepsilon_2 + v_2 \varepsilon_1} \)  eq.S2

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

Jayasundere-Smith model:  
\[
\varepsilon_{\text{eff}} = \frac{v_2 \varepsilon_2 + v_1 \varepsilon_1}{v_2 + v_1} \frac{3 \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \left[ 1 + \frac{3 v_1 (\varepsilon_1 - \varepsilon_2)}{\varepsilon_1 + \varepsilon_2} \right]
\]

Maxwell-Garnett model:  \( \varepsilon_{\text{eff}} = \varepsilon_2 \frac{(2v_1 + 1) \varepsilon_1 + 2(1 - v_1) \varepsilon_2}{(1 - v_1) \varepsilon_1 + (2 + v_1) \varepsilon_2} \)  eq.S5

Where \( \varepsilon_1 \) and \( \varepsilon_2 \) are the permittivity of fillers and polymer matrices; \( v_1 \) and \( v_2 \) are the volume fraction of fillers and polymer matrices, respectively. \( \varepsilon_{\text{eff}} \) is the effective permittivity of the composites.

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](image_url)

**Figure S1.** (a) The XRD patterns of raw BaTiO$_3$ powder about structure and composition information. (b) Comparison of XRD patterns of epoxy resin, raw BaTiO$_3$ powder, sintered porous BaTiO$_3$ ceramics and BaTiO$_3$/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 BaTiO$_3$ particles randomly distributed in epoxy resin as comparison.
Figure S4. The histograms of porosity and relative density of the porous BaTiO$_3$ ceramics measured by Archimedes method corresponding to the BT volume fraction.
The dielectric properties as a function of temperature for the BaTiO$_3$/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 BaTiO$_3$ ceramic is about 125°C, and at this temperature, the permittivity of BaTiO$_3$ reaches the maximum value; As shown in Fig. S5 (b), the permittivity of the resin appears a hump in the temperature ranges. For BaTiO$_3$/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) BaTiO$_3$ ceramic; (b) Epoxy resin; and BaTiO$_3$/Epoxy composite with (c) BT-Random-59, (d) BT-⊥-59, (e) BT-∥-59.
As shown in Fig S6. The bending strength of composite BT-random-59, BT-//-59, BT-⊥-59 are 55.79, 68.21, 75.45 MPa, respectively, which were higher than that of the BaTiO$_3$ 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 BaTiO$_3$ 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-⊥-59, and BT-//-59.