Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.
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Supplementary Information

Ultrahigh Enhancement Rate of Energy Density of Flexible Polymer Nanocomposites by Core-Shell $BaTiO_3$@MgO Structures as Fillers

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Experimental Section

1.1 Materials

Barium titanate powders were purchased from Shandong Guoci Functional Materials Co., Ltd, China, owning an average particle size of 80 nm. The P(VDF-HFP) (Arkema, Kynar Flex 3120) was provided by Arkema, France. Other reagents were purchased from Sinopharm Chemical Reagent Co. Ltd, China. All the materials were used as received. All solvents used in this paper are AR grade.

1.2 Synthesis of core-shell BT@MO nanoparticles

The core-shell BT@MO structure is fabricated by precipitation coating method. We prepared suspension slurries of BT by means of ultrasonic dispersion. The magnesium acetate tetrahydrate (6 wt%) was then added into the BT suspension slurries. Core-shell BT@MO structure with various thicknesses of MgO layers was obtained by controlling the contents with designed mass fractions of MgO ranging from 6 wt%. The PH value of the mixed slurry was adjusted to >9 by adding ammonia solution, triggering precipitation, and the doping elements were present as hydroxide coats on the surface of the BT particles. The coated BT powders were obtained by drying the total slurry, then calcining the residues at 700℃ for 2 h in air to transform them and the doping elements into oxides.

1.3 Fabrication of the P(VDF-HFP)-based nanocomposites

For the preparation of nanocomposites, the BT@MO nanoparticles was first dispersed in *N*,*N*-dimethylformamide (DMF) solvent through ultrasonication and magnetic stirring, and followed by the addition of P(VDF-HFP), then the mixture was stirred for 18 h to make it stable and homogenous. The nanocomposite films were cast from solutions on glass substrates with a scraper. The thickness of the films was controlled with scrapers. The films coated on glasses were dried under 60℃ for 12 hours and then the temperature was elevated to 200℃ for 5 min. After that, the films were quenched in cold water, followed by drying at 60℃ for overnight to evaporate residual water.

1.4 Characterization

The High Resolution-transmission electron microscopy (HR-TEM) micrographs were obtained by using Talos F200X (FEI) with an acceleration voltage of 200 kV. Scanning electron microscope (SEM) was conducted by using Quanta F250, FEI at 20 kV. Rigaku D/MAX-2400 X-ray diffractometer, (Tokyo, Japan) with Cu K_α radiation (λ = 0.15406 nm) operating at 40 kV and 100 mA. Young's modulus is measured using nanoindentation. Five samples for each composition were tested. The energy storage property was acquired using a ferroelectric test system (Premier II, Radiant) ferroelectric test system. During the measurement, the samples were immersed in silicone oil. The size of all the samples for energy storage property testing is 2.0 mm in diameter. The measurement of permittivity and loss tangent was performed with an Agilent 4990A impedance analyzer in the frequency range from 100 Hz to 10 MHz at room temperature. Electric displacement–electric field loops were measured at 10 Hz with a multiferroic ferroelectric test system (Premier II, Radiant Technologies, Inc.) at room temperature, the DC leakage current densities (in A/cm^2) were also analyzed by $\nabla D \cdot$area/∇t with this ferroelectric test system. The leakage current density and electrical tree propagation status were simulated by using COMSOL and MATLEB.

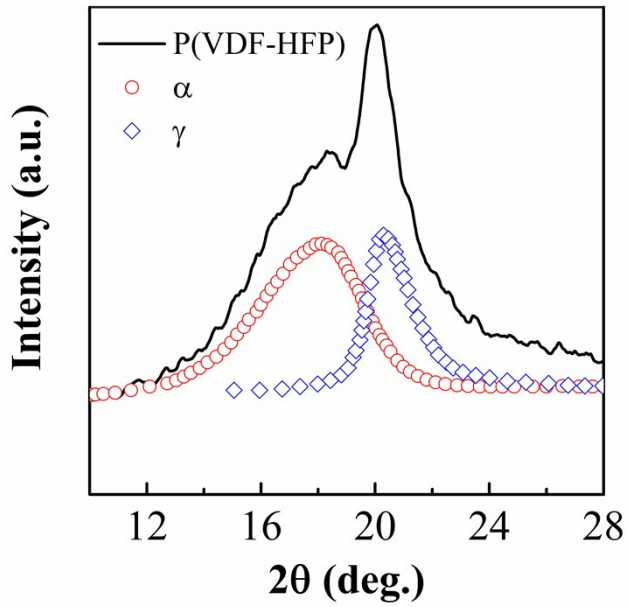

Fig. S1 Peak fitting results of XRD pattern from the 10-28° for α-phase and γ-phase of P(VDF-HFP).

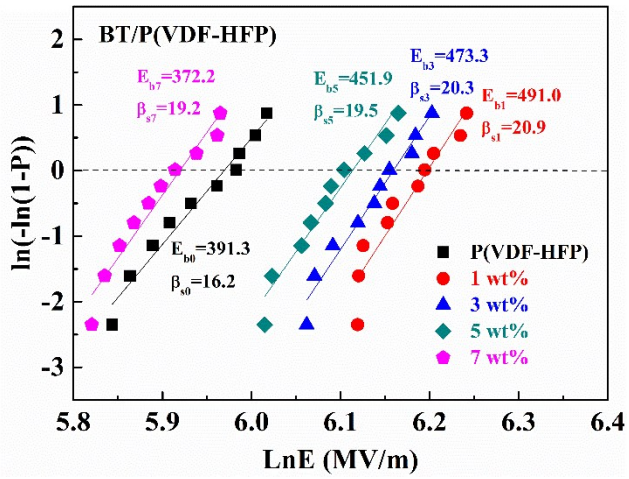

Fig. S2 Weibull distribution for BT/P(VDF-HFP) nanocomposites.

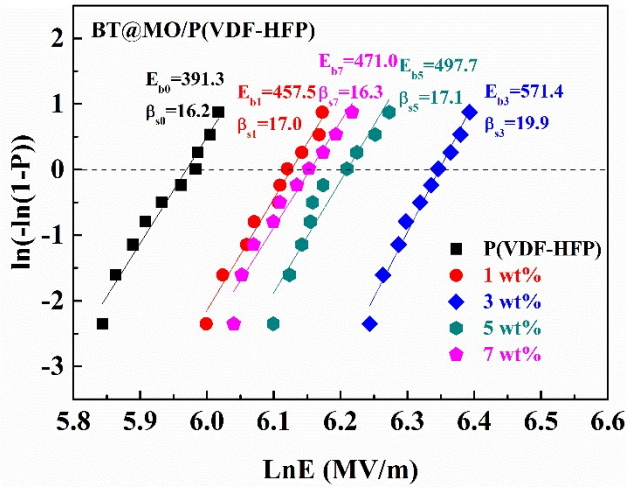

Fig. S3 Weibull distribution for the BT@MO/PVDF-HFP nanocomposites.

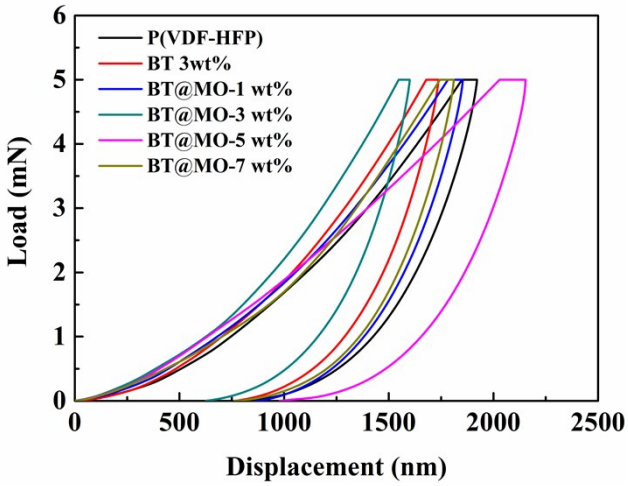

Fig. S4 Out-of-plane mechanical behaviors analyzed with nanoindentation**,** the load-displacement curves for polymer nanocomposites with different content.

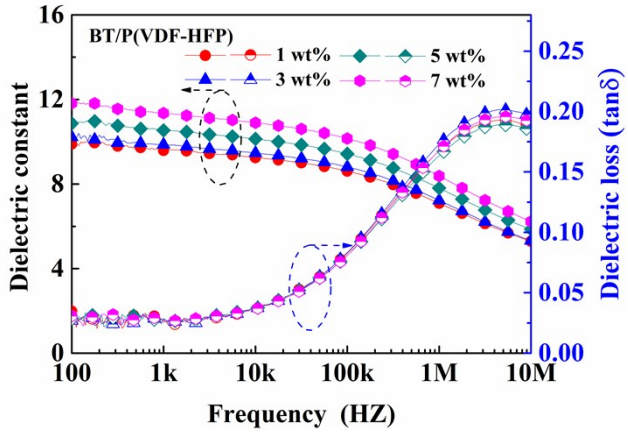

Fig. S5 The frequency-dependent dielectric constant (solid) and dielectric loss (half up) of P(VDF-HFP) nanocomposites filled with BT.