An additive manufacturing combining dielectrophoresis approach to 3D-structured flexible lead-free piezoelectric composites for electromechanical energy conversion

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Experimental Section

Materials

$BaTiO_3$ nanoparticles with a mean particle size of 200 nm were obtained from Shandong Sinocera Functional Material Co., Ltd, China. PDMS material SE 1700 and Sylgard 184 were both purchased from Dow Corning. 3D printing inks were synthesized by blending PDMS ink and $BaTiO_3$ nanoparticles. For silicone ink system, PDMS inks were produced by mixing 50 wt. % PDMS material SE 1700 and 50 wt. % Sylgard 184. Both two silicone-based materials were mixed with the catalyst in a 15:1 weight ratio before immixing. $BaTiO_3$-PDMS inks with different volume ratios were formulated by mixing varying ratios of $BaTiO_3$ nanoparticles to PDMS inks. The triple roller mills mixed the ink commixture for 2 hours. The mixture of $BaTiO_3$-PDMS inks was filled into the printing syringe and degassed for 3 hours at room temperature. Afterward, the ink was centrifuged to remove any air gas bubbles.

Additive manufacturing

The print-heads were designed using 3D computer-aided design software. The print-heads were attached to a set of ink-filled syringes fixed on a three-axis air-bearing linear-motion controller. The pressure was supplied to the syringes by digital pressure regulators. The inks were printed onto the aluminum foil using a nozzle (diameter, 260 or 410 μm) at a stable printing speed and extrusion pressure of 160 mm min^{-1}, and the gas pressure was in the range of 0.8 to 1.0 MPa.

Characterization

The microscopy and composition were conducted through scanning electron microscopy SEM (TESCAN) operated at 10 kV. Samples were detected by X-ray diffraction (XRD) with CuKα radiation (D8 Advance, Bruker). The polarization electric field hysteresis (*P*-*E*) loops of samples were obtained with precision multiferroic II (RADIANT technologies. INC). All mechanical properties of specimens were tested by a universal testing machine (UTM, INSTRON 5583) at room temperature. Rheological measurements were performed using a Rheometer HAAKE MARS III. The electromechanical performance was tested using a quasi-static piezoelectric meter (ZJ-3D, Institute of Acoustics, Beijing, China). The capacitance and impedance were characterized using an impedance analyzer (E4980A, KEYSIGHT Technology, USA). The piezoelectric energy harvesters' output voltages were measured with a digital storage oscilloscope (KEYSIGHT MSOX4024A). A digital force gauge was used to measure the dynamic pressing force applied to the energy harvester (SBT Co., Ltd, Guangzhou, China). An analog accelerometer (CT1005L, CHENGTEC Co., Ltd, Shanghai, China) was embedded with the sample holder's conditioning circuit and used for reference.

The 3D printed $BaTiO_3$-PDMS composites were polarized in a DC electric field of 100 kV cm^{-1}, in a high temperature (~ 110 °C) for 12 hours immerging in silicone oil and then taken out when the oil cooled down to room temperature. The 3D printed $BaTiO_3$-PDMS composites device was fabricated into strip with the dimension of 20 mm × 20 mm × 10 mm and assembled with copper as a piezoelectric energy harvester.

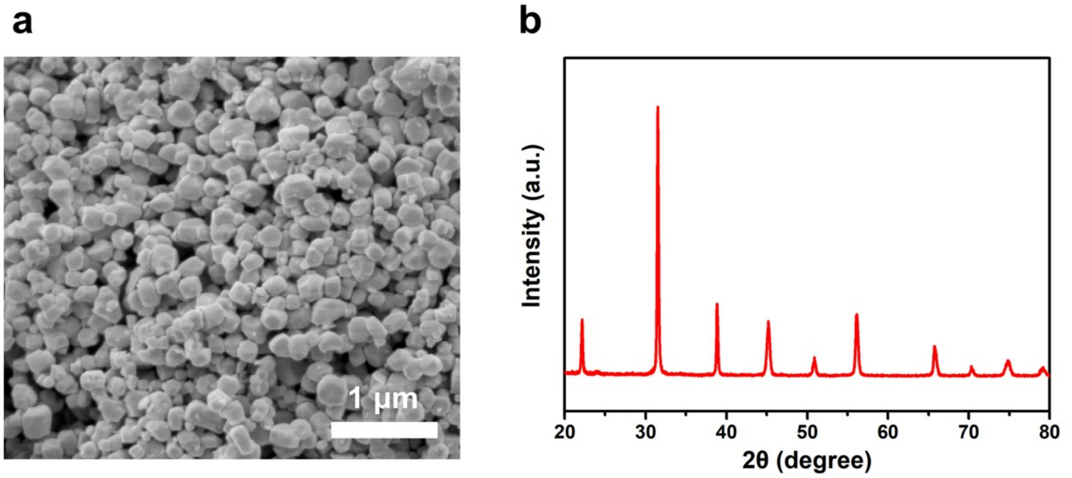

Fig. S1. (a) Cross-section scanning electron microscopy image of $BaTiO_3$ nanoparticles. (b) X-ray diffraction patterns of $BaTiO_3$ nanoparticles.

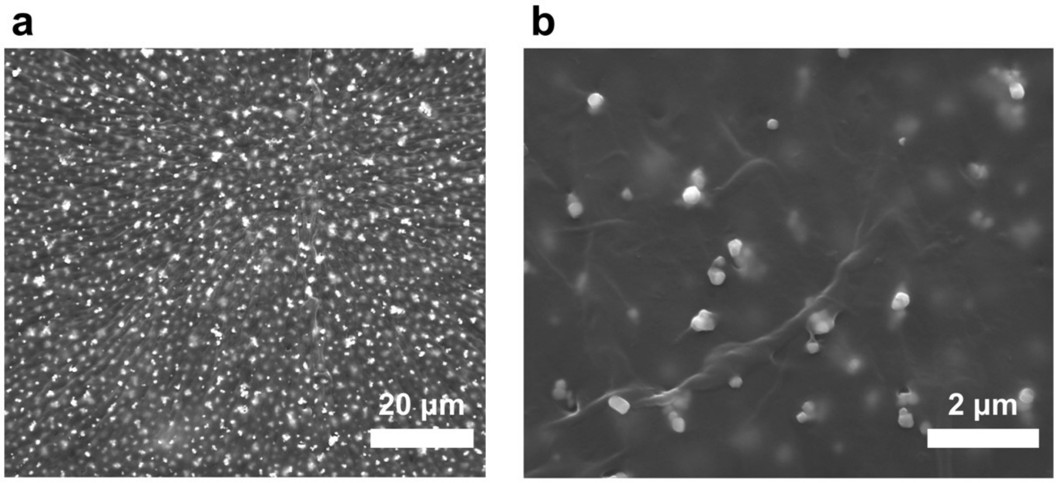

Fig. S2. Cross-section scanning electron microscopy image of 3D printed 0-3 structured $BaTiO_3$-PDMS composite.

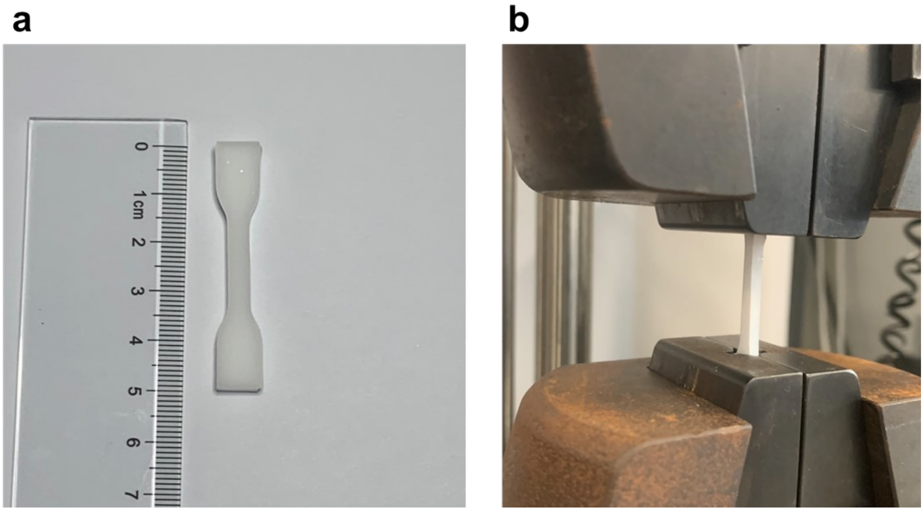

Fig. S3. (a) Dogbone-shaped 3D printed 1-3 structured $BaTiO_3$-PDMS composites. (b) Optical image of printed $BaTiO_3$-PDMS composite for tensile demonstrations.

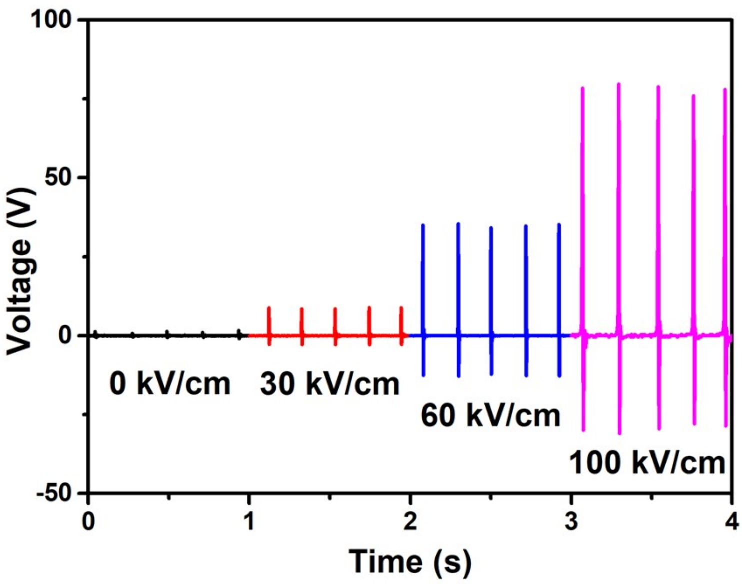

Fig. S4 The output voltage of the $BaTiO_3$-PDMS composites (15 vol. %) with different poling electric field.

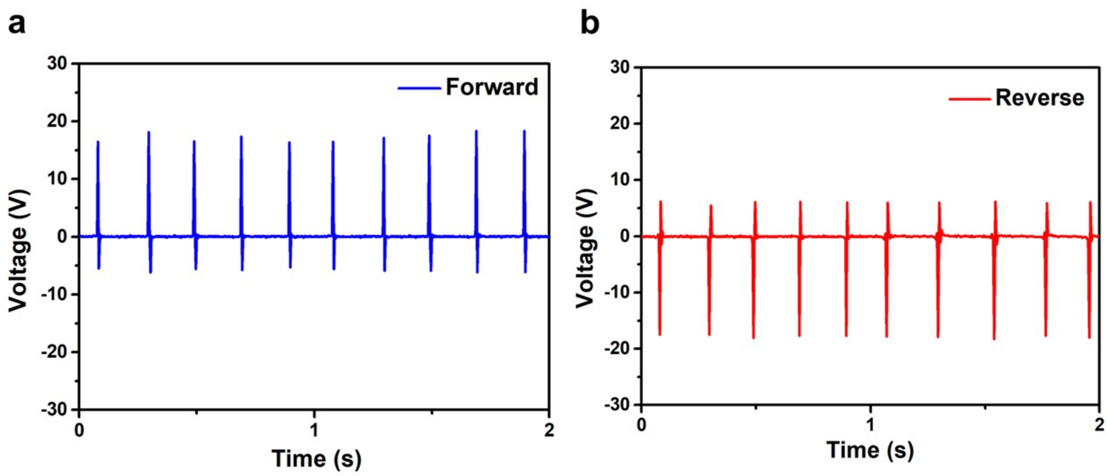

Fig. S5 Output voltage of forward (a) and reverse (b) connection respectively, which confirm that the output is due to piezo response.

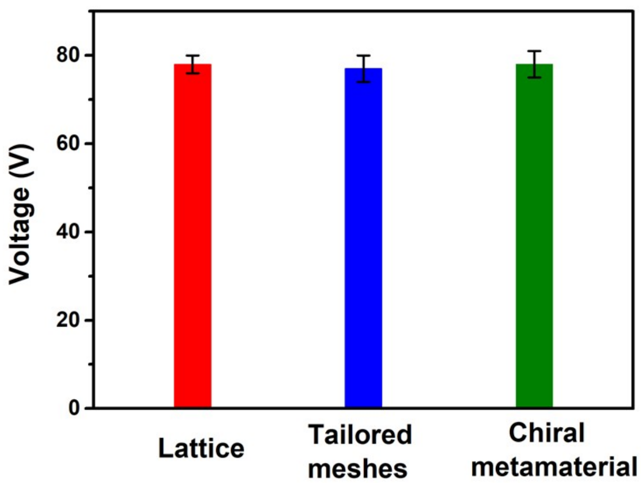

Fig. S6. Comparison of piezoelectric performance for different dielectrophoretically aligned 3D printed shapes (15 vol. %) under compressive stress of 0.1 MPa.

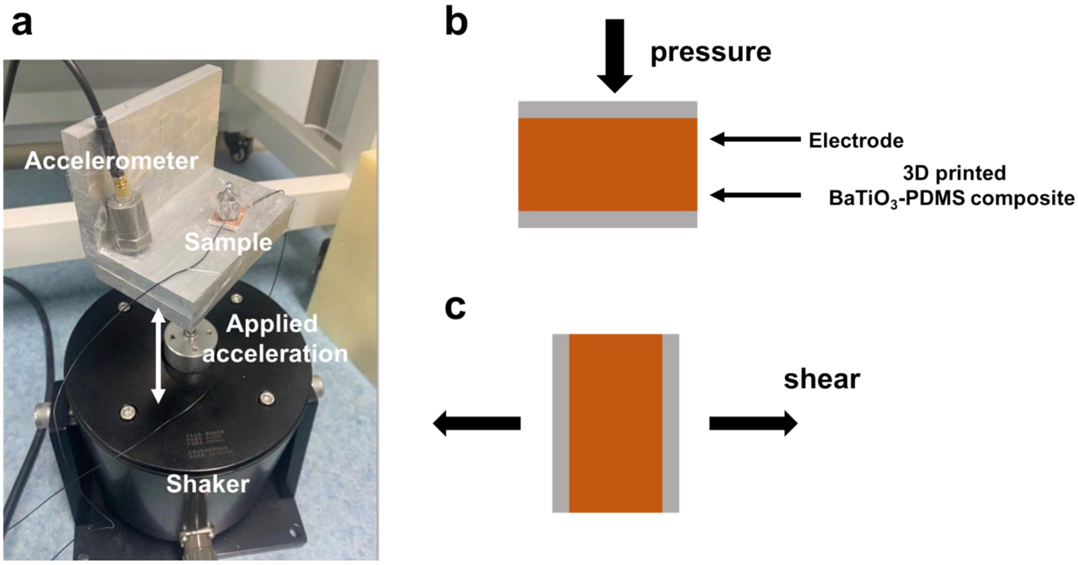

Fig. S7. (a) Optical image of the longitudinal sensor sensitivity experimental measurement (vertical mount of L holder for shear sensor measurements). Schematic of the (b) longitudinal [d_{33} mode] sensor and (c) shear [d_{15} mode] sensor to shaker acceleration

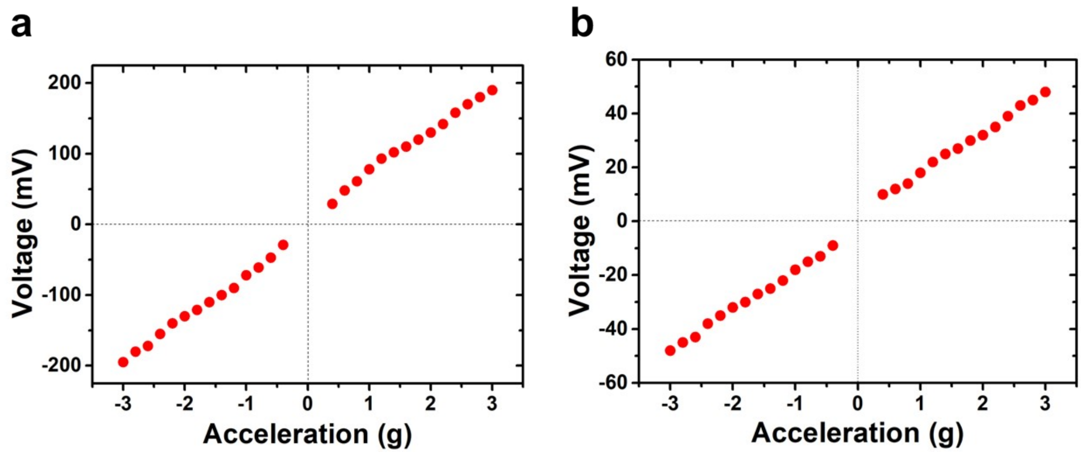

Fig. S8. Voltage response of the (a) d_{33} sensor and (b) d_{15} sensor to shaker acceleration.