## **Supporting information**

## Ultrahigh energy harvesting properties in textured lead-free piezoelectric composites

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## Synthesis of 0.94(Ba<sub>0.979</sub>Ca<sub>0.021</sub>)(Ti<sub>0.957</sub>Sn<sub>0.043</sub>)O<sub>3</sub>-0.06BaTiO<sub>3</sub> control samples

Randomly oriented  $0.94(Ba_{0.979}Ca_{0.021})(Ti_{0.957}Sn_{0.043})O_3$ -0.06BaTiO<sub>3</sub> (0.94BCTS-0.06BT) ceramics were prepared by conventional mixed-oxide method using BaCO<sub>3</sub> (99.95%), CaCO<sub>3</sub> (99.99%), nano-TiO<sub>2</sub> (99.8%), and nano-SnO<sub>2</sub> (99.9%) raw materials. The raw materials were weighed according to the stoichiometry of 0.94BCTS-0.06BT, and then ball-milled for 96 h in ethanol. The dried powder mixture was calcined at 1200 °C for 3 h and then tape cast using a nonaqueous formulation.<sup>1</sup> Dried green tapes were diced, stacked and laminated under 20 MPa and 75 °C to fabricate green compacts. After binder burnout at 600 °C and cold-isostatic pressing at 200 MPa, samples were heated at 5 °C/min to 1450 °C and then held for 4 h.



Fig. S1 (a) XRD patterns and (b, c) SEM images of BCTS matrix powder and BT microplatelet template particles.



Fig. S2 Schematic diagrams of cantilever-type energy harvester and test system with load resistance.



Fig. S3 Element distributions (Ba, Ca, Ti and Sn) of 0.94BCTS-0.06BT ceramics.



Fig. S4 Output power density as a function of load resistance for non-textured counterpart and textured BCTS/BT energy harvesters at 1 *g* acceleration. According to the schematic diagram of the equivalent circuit with load resistance  $R_{\text{Load}}$  (Fig. S2), there is an optimum  $R_{\text{Load}}$  value that matches with internal impedance  $R_{\text{I}}$  of the harvester, resulting in the maximum output power density. Here the optimum  $R_{\text{Load}}$  value was found to be 450 k $\Omega$  for both non-textured and textured energy harvesters.



Fig. S5 Open circuit peak-to-peak voltage ( $V_{oc}$ ) as a function of frequency for non-textured counterpart and textured BCTS/BT energy harvesters at 1 g acceleration. The  $V_{oc}$  value shows a strong frequency dependence, and the maximum  $V_{oc}$  occurs at the resonance frequency of the cantilever beam, at which maximum strain on the piezoelectric samples can be generated. Here the resonance frequencies were found to be 92 Hz and 90 Hz for the non-textured and textured energy harvesters, respectively.

## Reference

1. Y. F. Chang, S. F. Poterala, Z. P. Yang, S. Trolier-McKinstry and G. L. Messing, *Appl. Phys. Lett.*, 2009, **95**, 232905.