

Supporting Information

Functionally graded lead-free piezoelectric composites for enhanced ultrasound energy harvesting

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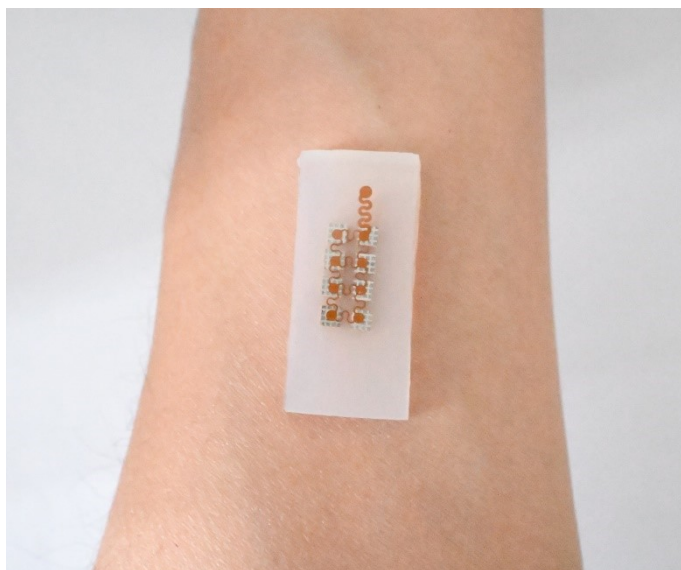


Figure S1 The PUEH attached to the arm.

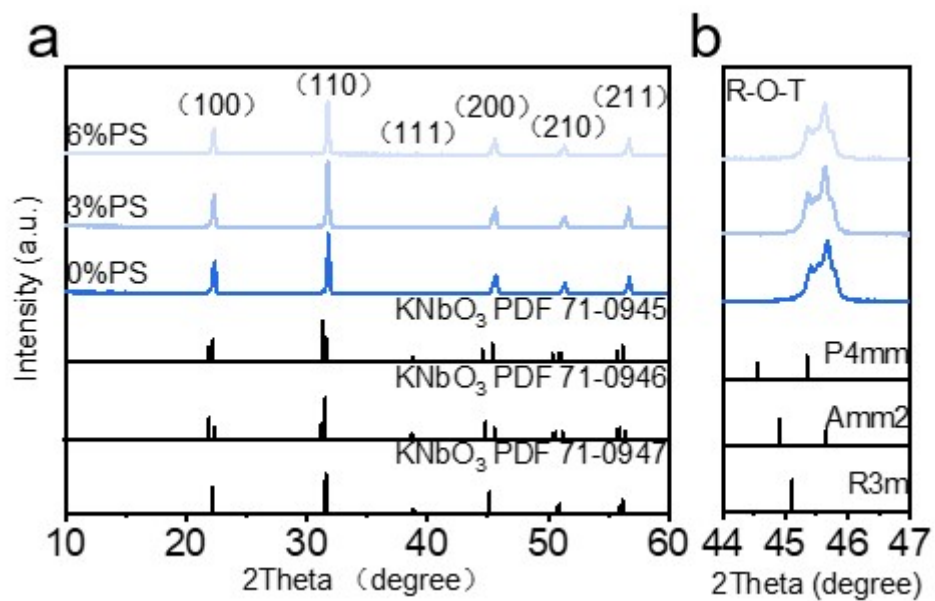


Figure S2 XRD patterns of the piezoelectric composites with different PS contents. a Patterns for samples doped with 6 wt.%, 3 wt.%, and undoped with PS. b An enlarged view of the XRD patterns.

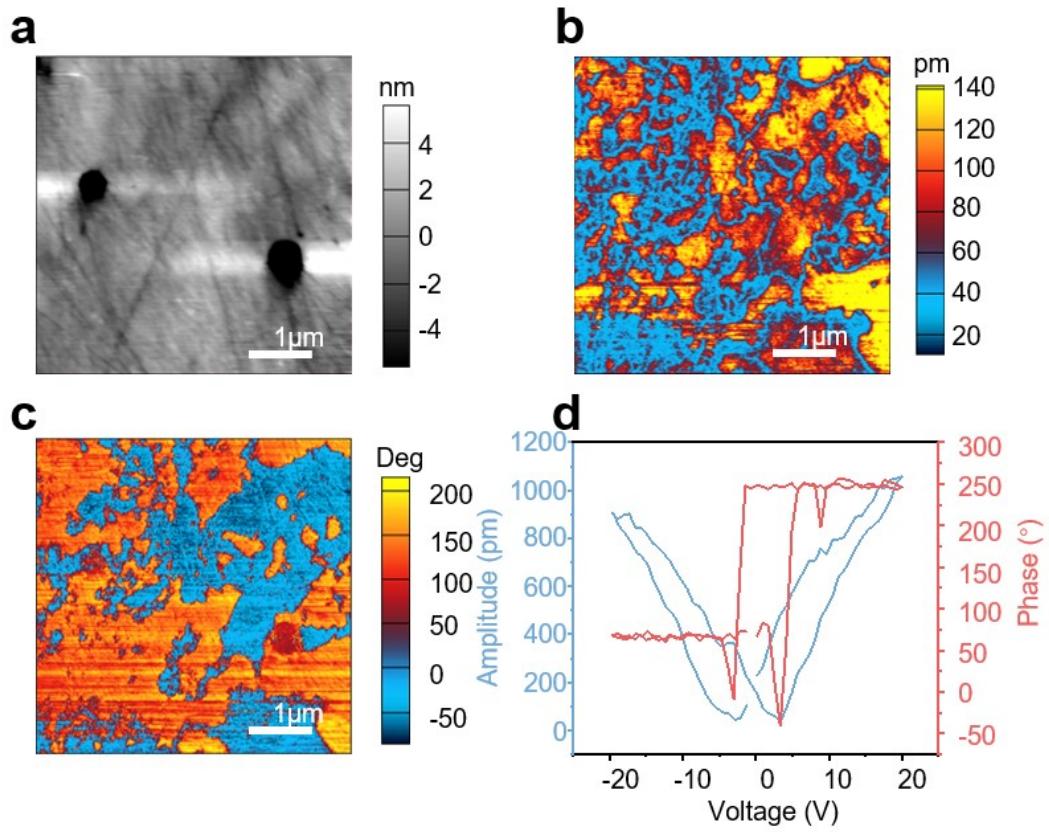


Figure S3 PFM characterization of dense KNNS-NS piezoelectric ceramics: a schematic illustration, b amplitude image, c phase image, and d SS-PFM loop.

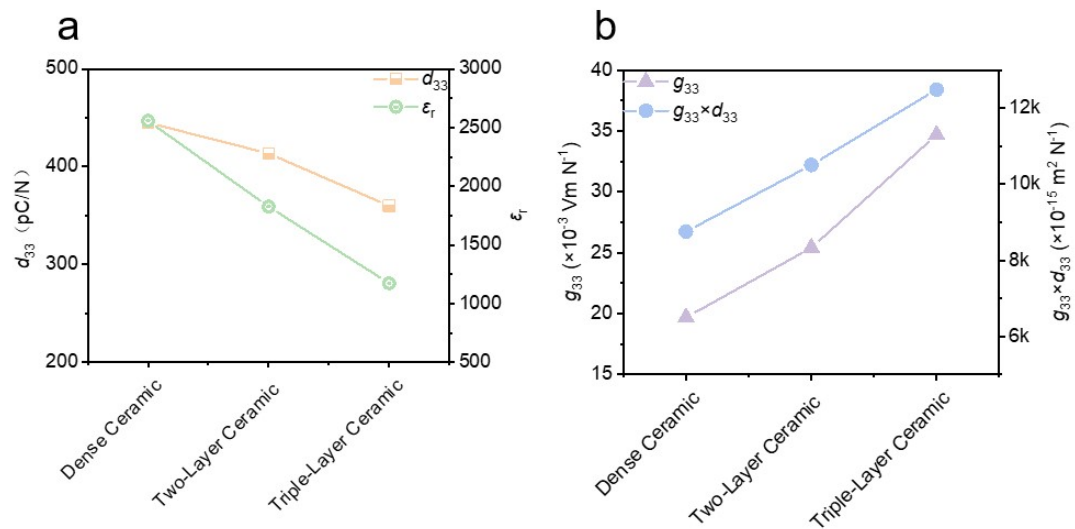


Figure S4 d_{33} , ϵ_r , g_{33} and $d_{33} \times g_{33}$ as a function of the number of gradient layers.

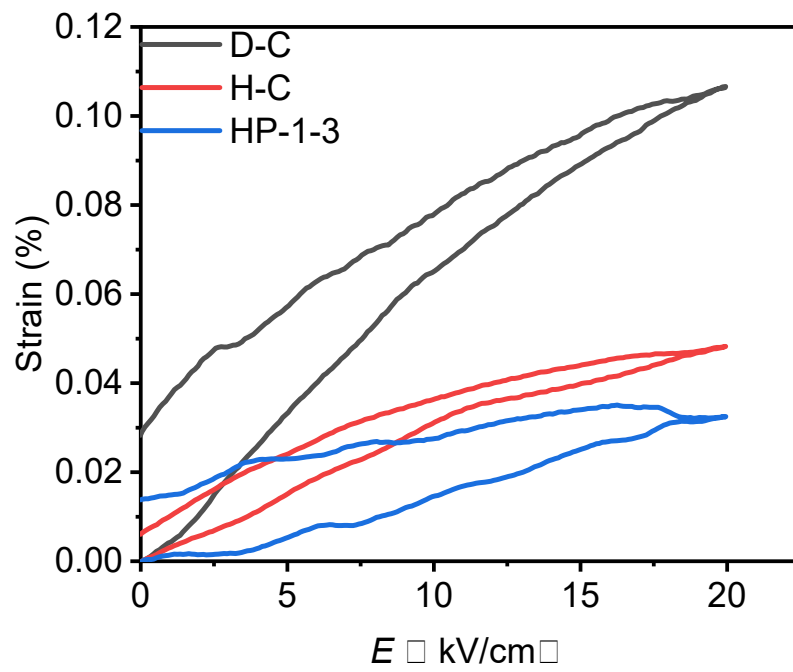


Figure S5 Unipolar strain curves of the composite materials with three different structures.

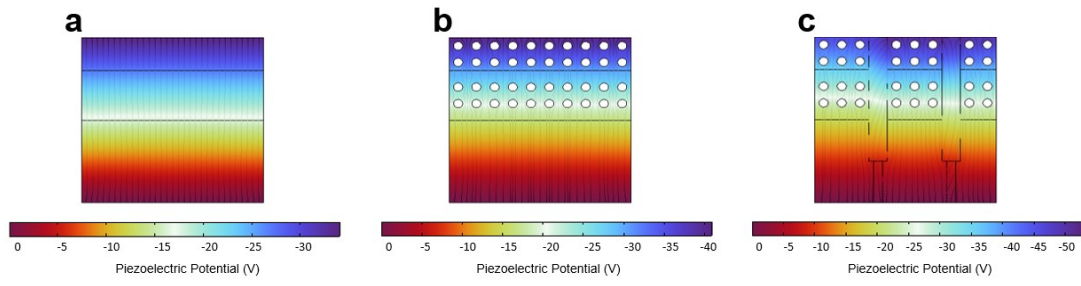


Figure S6 Finite element analysis of three piezoelectric composite structures under identical mechanical loading. a D-C, b H-C, c HP-1-3.

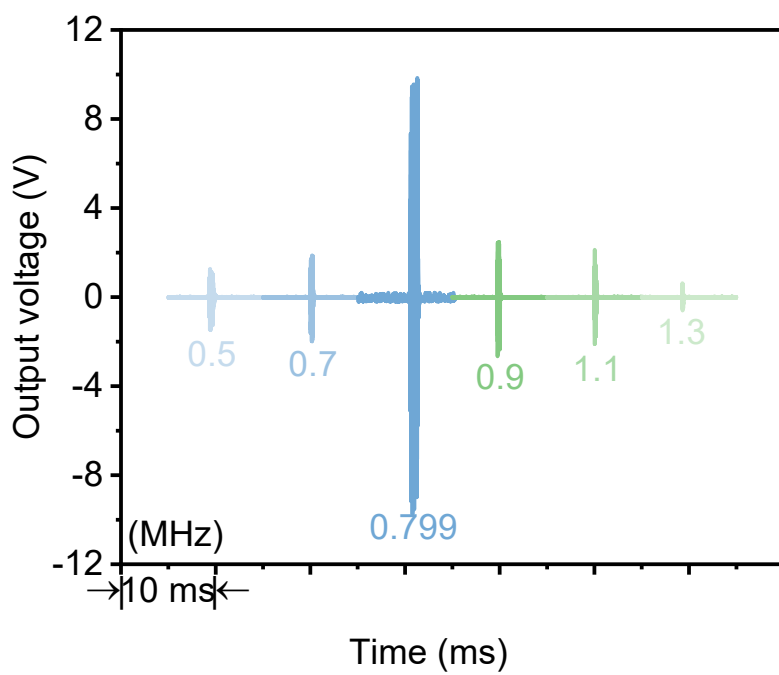


Figure S7 Dependence of the output voltage on frequency.

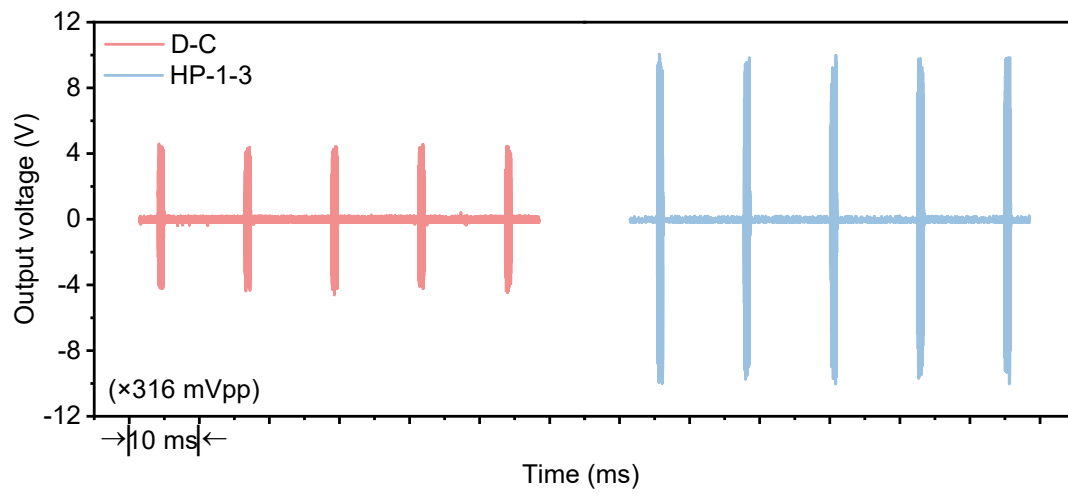


Figure S8 Comparison of the output signals between D-C and HP-1-3 composites under identical conditions.

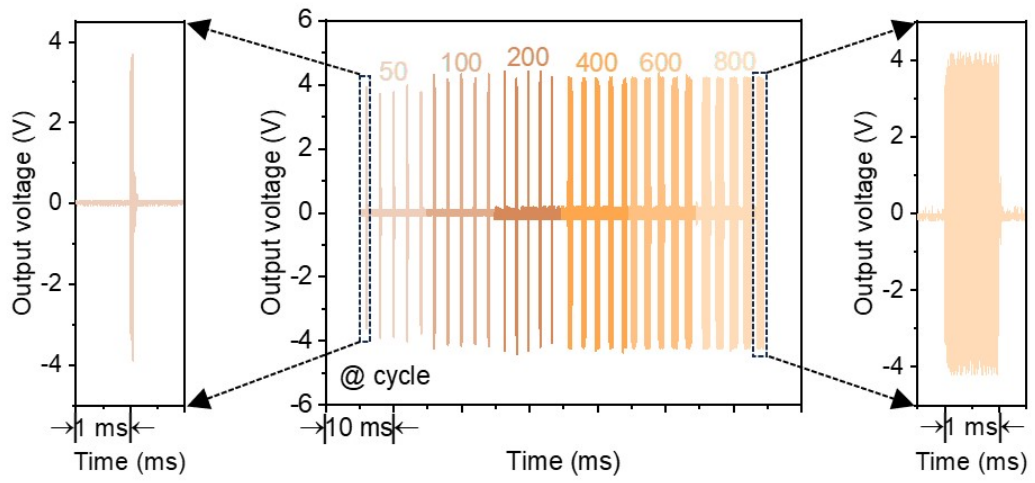


Figure S9 Output voltages under the excitation of trigger signals with different cycles.

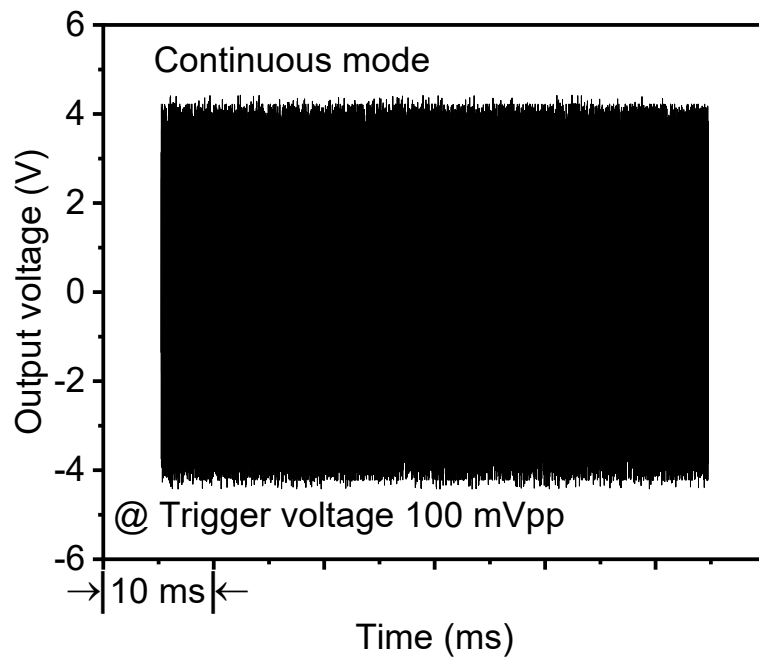
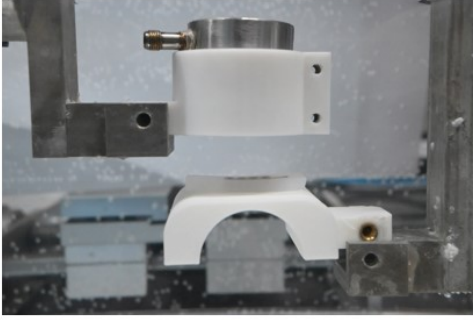


Figure S10 Output voltages of the sample measured in a continuous mode.

a



b

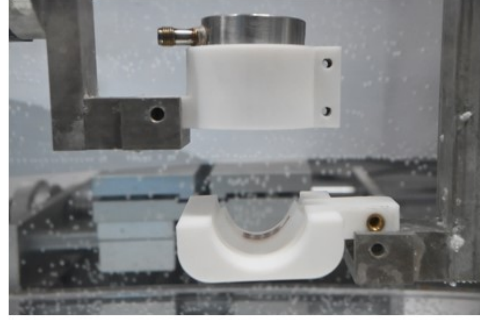


Figure S11 Schematic of the water tank test for the PUEH on different surfaces. a Flat surface, b curved surface.

Table S1. Comparison of piezoelectric properties between the proposed ceramic formulation and conventional lead-free/lead-based counterparts

Materials	d_{33}
KNNS-NS	480 pC/N
KNNS-BNZ-Fe ¹	450 pC/N
KNN-0.045BNZ ²	401 pC/N
KNNS-CZ ³	430 pC/N
PZ-PT ⁴	405 pC/N
BS-PT ⁵	490 pC/N
PZT-4 ⁶	612 pC/N

Table S2 Comparison of acoustic impedance values

PS x wt. %	Z (MRayl)
0	33.75
3	23.45
6	16.7

Reference

1. C. Zhu, H. Xue, Q. Zhou, L. Jiang and J. Wu, *Journal of Materials Chemistry A*, 2024, **12**, 16116-16128.
2. Z. Dong, J. Yang, H. Liu, Y. Zhou, Y. Ai, J. Song and J. Jiao, *Ceramics International*, 2025, DOI: 10.1016/j.ceramint.2025.06.087.
3. H. Yang, L. Tang, S. Han, Y. Hua, J. Lin, J. Qian, B. Shen, G. Jiang and J. Zhai, *Advanced Functional Materials*, 2025, DOI: 10.1002/adfm.202507702.
4. Z. Xue, D. Jiang, Y. Li, H. Yang, L. Xing and Z. Li, *Journal of Alloys and Compounds*, 2025, **1036**.
5. Y. Dong, Z. Zhou, R. Liang and X. Dong, *Journal of Materiomics*, 2022, **8**, 319-326.
6. H. Liu, Y. Luo, H. Chen, W. Zhuang, W. Wang and J. Zhu, *Journal of the American Ceramic Society*, 2025, **108**.