Supporting Information

High and thermal-stable piezoelectricity in relaxor-based ferroelectrics for mechanical energy harvesting

Xiaole Yu, Yudong Hou,* Mupeng Zheng, Mankang Zhu

Faculty of Materials and Manufacturing, Key Laboratory of Advanced Functional

Materials, Education Ministry of China, Beijing University of Technology, Beijing

100124, China

*Corresponding author

E-mail: ydhou@bjut.edu.cn



Fig. S1. (a) The piezoelectric charge coefficient d_{33} of xPZN-(1-x)PH $_y$ T $_{(1-y)}$ ceramics as a function of PH $_y$ T $_{(1-y)}$ content. (b) Temperature dependence of dielectric permittivity ε_r and dielectric loss tan δ for different MPB ceramic samples measured at 1 kHz.



Fig. S2. The temperature dependence of the dielectric permittivity ε_r and dielectric loss $\tan \delta$ for the MPB-PZT (PZN-PZT) and MPB-PHT (PZN-PHT) ceramics measured at 100 Hz. The insert shows the fitted relaxor behavior by a modified Curie-Weiss law $(1/\varepsilon_r - 1/\varepsilon_{max} = (T - T_{max})^{\gamma}/C).^1$



Fig. S3. The frequency dependence of the output open-circuit voltage for the MPB-PHT PEH under 1*g* acceleration excitation at different temperatures: (a) 25 °C, (b) 150 °C, (c) 250 °C. (d) The resonate frequencies as a function of temperature for the MPB-

PHT PEH under 1g acceleration excitation.

Fig. S3a-c show that the open circuit voltage of MPB-PHT piezoelectric energy harvester (PEH) with varying frequency and temperature under the fixed vibration excitation of 1 g acceleration. It can be seen that the output voltage at different temperatures increases first and then decreases as the frequency increases, and reaches the maximum value at the resonance frequency of cantilever beam.² The resonance frequencies at different temperatures are given in Fig. S3d.



Fig. S4. Durability test of the MPB-PHT harvester at 250 °C.

No obvious electrical loss of the output voltage of MPB-PHT PEH after about 200000 cycles at 250 °C.

| Samples | Т | Phase | Lattice parameters | | | Reliability factors | | |
|---------|------|-------|--------------------|--------|---------|---------------------|-----------------|----------|
| | (°C) | | a (Å) | c (Å) | β (°) | $R_{\rm wp}$ (%) | $R_{\rm p}(\%)$ | χ^2 |
| MPB-PHT | 150 | P4mm | 4.0458 | 4.0994 | 90.0000 | 5.29 | 3.89 | 2.61 |
| | | R3mr | 4.0808 | 4.0808 | 89.8889 | | | |
| | 250 | P4mm | 4.0518 | 4.0817 | 90.0000 | 6.32 | 4.45 | 3.11 |
| | | R3mr | 4.0716 | 4.0716 | 89.8071 | | | |
| MPB-PZT | 150 | P4mm | 4.0489 | 4.1137 | 90.0000 | 7.88 | 5.72 | 4.21 |
| | | R3mr | 4.0800 | 4.0800 | 89.9524 | | | |
| | 250 | P4mm | 4.0562 | 4.0991 | 90.0000 | 8.98 | 6.31 | 4.77 |
| | | R3mr | 4.0709 | 4.0709 | 89.9896 | | | |

Table S1. Calculated lattice parameters of the poled MPB-PHT and MPB-PZT

 ceramics by refinement at different temperatures.

Notes: *P4mm*: $a = b \neq c$, $\alpha = \beta = \gamma = 90^{\circ}$; *R3mr*: a = b = c, $\alpha = \beta = \gamma < 120^{\circ} \neq 90^{\circ}$.

References

- 1. K. Uchino and S. Nomura, *Ferroelectrics*, 2011, 44, 55-61.
- 2. X. Yu, Y. Hou, M. Zheng and M. Zhu, ACS Appl. Mater. Interfaces, 2021, 13,

17800-17808.