Supplementary information

Enhanced pyroelectric and piezoelectric properties of PZT with aligned porosity for energy harvesting applications

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Figure S1 Compressive strengths of both parallel-connected and series-connected freeze-cast porous PZT. Data for conventional porous PZT with uniformly distributed porosity and dense PZT ceramics also shown.
Figure S2 Polarisation (P) - electric field (E) hysteresis loops of the dense PZT.
3.3 Piezoelectric and dielectric properties

Piezoelectric coefficients for the series and parallel connection can be calculated by the following equations:

For series connection by Equations S1 and S2:

\[
\begin{align*}
\frac{d_{33}}{V_{PT}^{PZT}d_{33}^{pc}\varepsilon_{33}^{pc} + V_{PC}d_{33}^{PC}\varepsilon_{33}^{PZT}} &= \frac{V_{PT}^{PZT}d_{31}^{pc}\varepsilon_{33}^{PC} + V_{PC}d_{31}^{PC}\varepsilon_{33}^{PZT}}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} \quad (S1) \\
\frac{d_{31}}{V_{PT}^{PZT}d_{31}^{pc}\varepsilon_{33}^{PC} + V_{PC}d_{31}^{PC}\varepsilon_{33}^{PZT}} &= \frac{V_{PT}^{PZT}s_{11}^{pc} + V_{PC}s_{11}^{PC}}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} \quad (S2)
\end{align*}
\]

For parallel connection by Equations S3 and S4:

\[
\begin{align*}
\frac{d_{33}}{V_{PT}^{PZT}d_{33}^{pc}\varepsilon_{33}^{PC} + V_{PC}d_{33}^{PC}\varepsilon_{33}^{PZT}} &= \frac{V_{PT}^{PZT}s_{33}^{pc} + V_{PC}s_{33}^{PC}}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} \quad (S3) \\
\frac{d_{31}}{V_{PT}^{PZT}d_{31}^{pc}\varepsilon_{33}^{PC} + V_{PC}d_{31}^{PC}\varepsilon_{33}^{PZT}} &= \frac{2V_{PT}^{PZT}V_{PC}(d_{33}^{PC} - d_{33}^{PZT})}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} + \frac{V_{PT}^{PZT}(s_{11}^{PC} + s_{12}^{PC})}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} \quad (S4)
\end{align*}
\]

3.4 Pyroelectric properties

Theoretical pyroelectric formulations for the series and parallel connections are given by Equations (S5) and (S6) respectively.

For series connection:

\[
p = \frac{V_{PT}^{PZT}d_{33}^{PC}\varepsilon_{33}^{PC} + V_{PC}d_{33}^{PC}\varepsilon_{33}^{PZT}}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} + \frac{2V_{PT}^{PZT}V_{PC}(d_{33}^{PC} - d_{33}^{PZT})}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} + \frac{V_{PT}^{PZT}(s_{11}^{PC} + s_{12}^{PC})}{V_{PT}^{PZT}\varepsilon_{33}^{PC} + V_{PC}\varepsilon_{33}^{PZT}} \quad (S5)
\]

For parallel connection:
\[ p = V^{PZT} p^{PZT} + V^{pc} p^{pc} + \frac{V^{PZT} V^{pc} p^{PZT} (\alpha^{pc} - \alpha^{PZT}) (d_{33}^{PZT} - d_{33}^{pc})}{V^{PZT} s_{33}^{pc} + V^{pc} s_{33}^{PZT}} \]

(S6)

where \( \alpha \) is thermal expansion coefficient.

Figure S3 Dielectric loss of the aligned porous PZT with (A) Parallel-connected and (B) Series-connected.
B) Series-connected modes.

Table S1 Model fitting parameters of the piezoelectric coefficients of the freeze-cast porous PZT with both series and parallel connectivities.

| Porosity (%) | $d_{33}$ | $d_{31}$ |
|--------------|----------|----------|
|              | Series-connected | Parallel-connected | Series-modelled | Parallel-connected |
|              | m    | n    | m, n        | N/A         |
| 20           | 6.012 | 0.781 | m=6.012, n=0.781 | -           |
| 30           | 6.019 | 0.762 | m=6.019, n=0.762 | -           |
| 40           | 6.021 | 0.759 | m=6.021, n=0.759 | -           |
| 50           | 6.032 | 0.757 | m=6.032, n=0.757 | -           |
| 60           | 6.089 | 0.755 | m=6.089, n=0.755 | -           |