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:

$$d_{33} = \frac{V^{PZT} \ d_{33}^{PZT} \varepsilon_{33}^{pc} + V^{pc} \ d_{33}^{pc} \ \varepsilon_{33}^{PZT}}{V^{PZT} \varepsilon_{33}^{pc} + V^{pc} \ \varepsilon_{33}^{PZT}}$$

(S1)

$$d_{31} = \frac{V^{PZT} \ d_{31}^{PZT} \ \varepsilon_{33}^{pc} \ s_{11}^{pc} + V^{pc} \ d_{31}^{pc} \ \varepsilon_{33}^{PZT} \ s_{11}^{PZT}}{\left(V^{PZT} \varepsilon_{33}^{pc} + V^{pc} \ \varepsilon_{33}^{PZT}\right) \left(V^{PZT} \ s_{11}^{pc} + V^{pc} \ s_{11}^{PZT}\right)}$$
(S2)

For parallel connection by Equations S3 and S4:

$$d_{33} = \frac{V^{PZT} \ d_{33}^{PZT} \ s_{33}^{pc} + V^{pc} \ d_{33}^{pc} \ s_{33}^{PZT}}{V^{PZT} \ s_{33}^{pc} + V^{pc} \ s_{33}^{PZT}}$$
(S3)

$$d_{31} = V^{PZT} \ d_{31}^{PZT} + V^{pc} \ d_{31}^{pc}$$
(S4)

3.4 Pyroelectric properties

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

For series connection:

р

$$= \frac{V^{PZT}p^{PZT} \varepsilon_{33}^{pc} + V^{pc} p^{pc} \varepsilon_{33}^{PZT}}{V^{PZT} \varepsilon_{33}^{pc} + V^{pc} \varepsilon_{33}^{PZT}} + \frac{2 V^{PZT} V^{pc} (\varepsilon_{33}^{pc} d_{33}^{PZT} - \varepsilon_{33}^{PZT} d_{33}^{pc}) (\alpha}{(V^{PZT} \varepsilon_{33}^{pc} + V^{pc} \varepsilon_{33}^{PZT}) [V^{PZT} (s_{11}^{pc} + s_{12}^{pc}) + (S5)]}$$

For parallel connection:

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$$p = V^{PZT} p^{PZT} + V^{pc} p^{pc} + \frac{V^{PZT} V^{pc} p^{PZT} (\alpha^{pc} - \alpha^{PZT}) (d^{PZT}_{33} - d^{pc}_{33})}{V^{PZT} s^{pc}_{33} + V^{pc} s^{PZT}_{33}}$$
(S6)

where α is thermal expansion coefficient.



Figure S3 Dielectric loss of the aligned porous PZT with (A) Parallel-connected and (a. Department of Mechanical Engineering, University of Bath, BA2 7AY, UK. E-mail: <u>C.R.Bowen@bath.ac.uk</u> b. State Key Laboratory of Powder Metallurgy, Central South University, 410083, China. E-mail: <u>dzhang@csu.edu.cn</u>

B) Series-connected modes.

Table S1 Model fitting parameters of the piezoelectric coefficients of the freeze-cast porous

d_{33}		d_{31}	
Series- connected	Parallel- connected	Series-modelled	Parallel- connected
m	n	m, n	N/A
6.012	0.781	m=6.012, n=0.781	-
6.019	0.762	m=6.019, n=0.762	-
6.021	0.759	m=6.021, n=0.759	-
6.032	0.757	m=6.032, n= 0.757	-
6.089	0.755	m=6.089, n= 0.755	-
	d Series- connected m 6.012 6.019 6.021 6.032 6.089	d ₃₃ Series-connected Parallel-connected m n 6.012 0.781 6.019 0.762 6.021 0.759 6.032 0.757 6.089 0.755	d_{33} d_{3l} Series- connectedParallel- connectedSeries-modelledmnm, n 6.012 0.781 $m=6.012, n=0.781$ 6.019 0.762 $m=6.019, n=0.762$ 6.021 0.759 $m=6.021, n=0.759$ 6.032 0.757 $m=6.032, n=0.757$ 6.089 0.755 $m=6.089, n=0.755$

PZT with both series and parallel connectivities.