Electronic Supplementary Material (ESI) for Materials Horizons.

Modulating piezoelectricity and mechanical strength via three-dimensional gradient structure for piezoelectric composites

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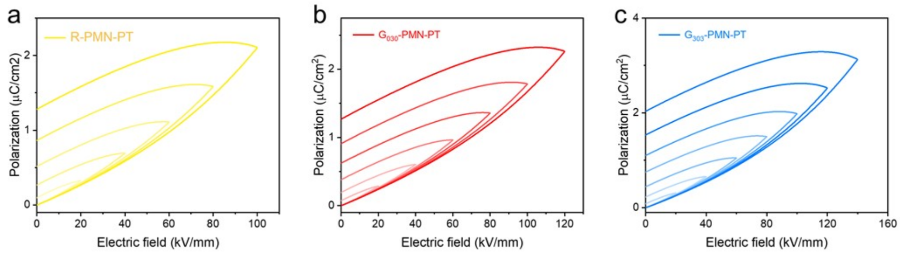

Figure S1. Polarization-electric (P-E) curves of R-PMN-PT (a), G_{030}-PMN-PT (b) and G_{303}-PMN-PT (c) under different electric fields.

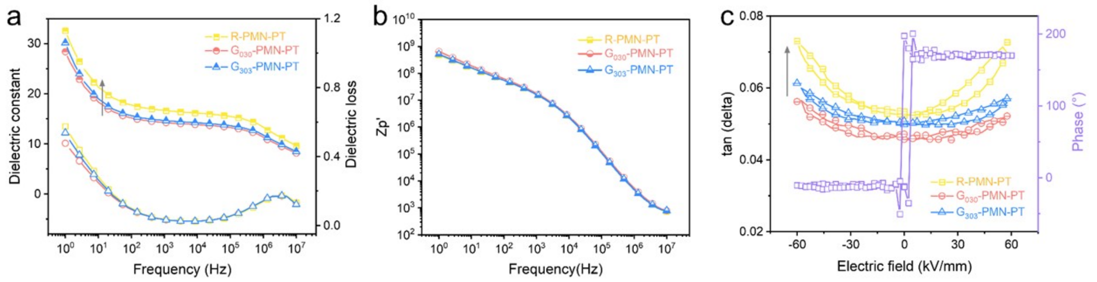

Figure S2. Dielectric constant, dielectric loss (a) and impedance (b) of the gradient structure film with frequencies from 10^0 to 10^7 Hz. (c) Tan (delta) and phase image of the gradient films under AC triangular waves at 1 kHz.

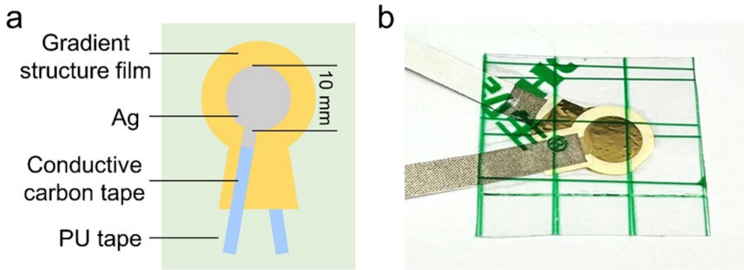

Figure S3. Structure and photograph of the prepared flexible pressure sensor. a) Structure of the flexible device. b) Photograph of the fabricated flexible device.

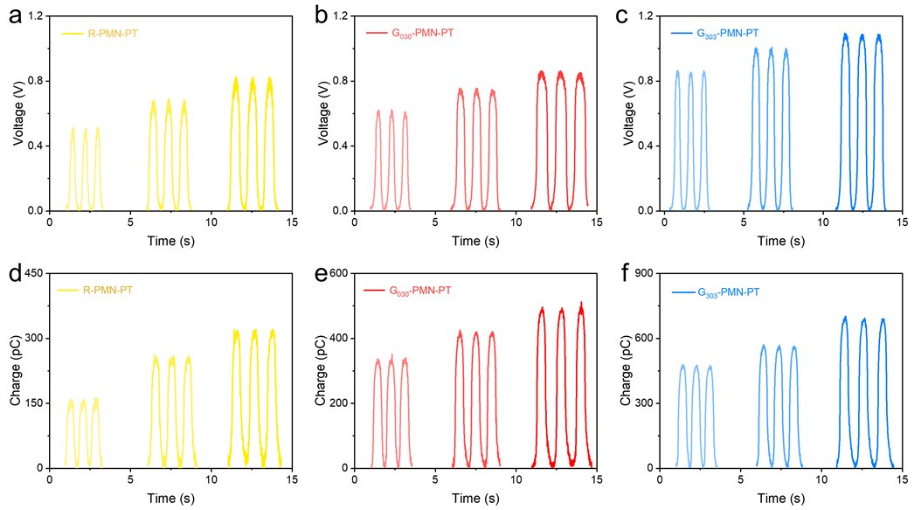

Figure S4. Output voltage (a-c) and charge (d-f) of the gradient structure films under different pressures at the same frequency.

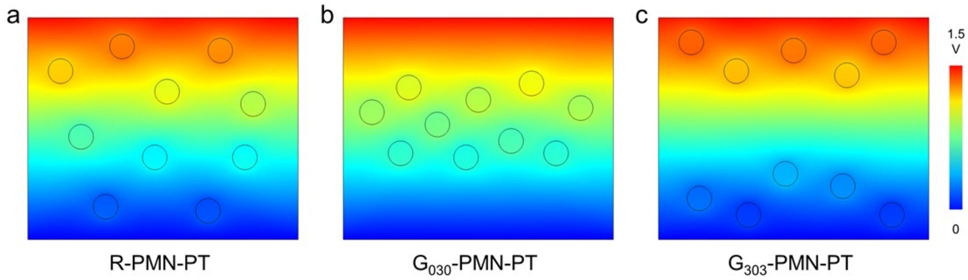

Figure S5. Simulation results of output voltage for R-PMN-PT (a), G_{030}-PMN-PT (b) and G_{303}-PMN-PT (c) under the same pressure.

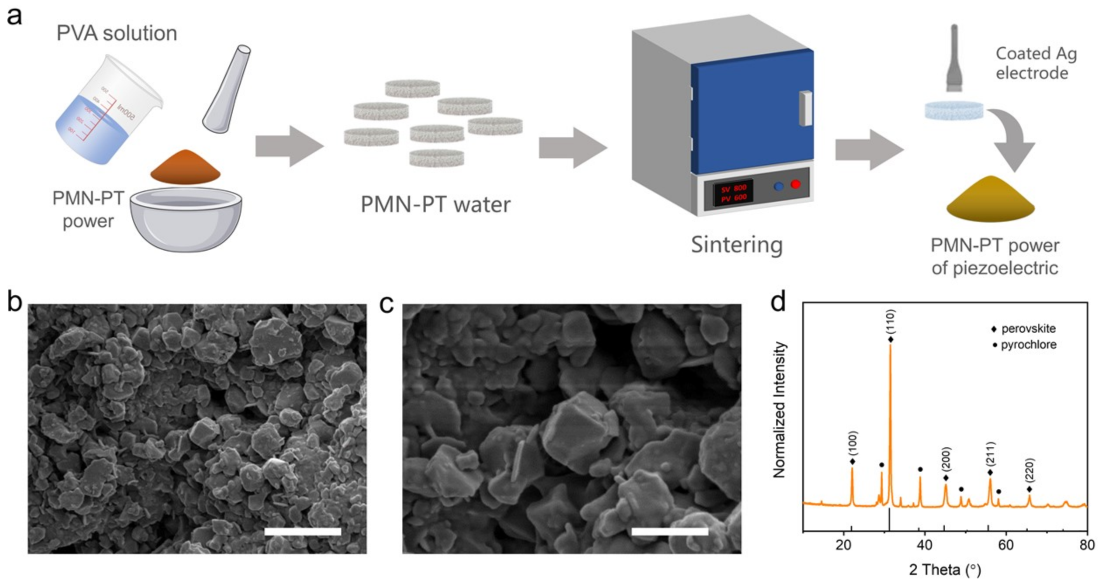

Figure S6. a) Schematic diagram of the fabrication process of PMN-PT nanoparticles. b-c) SEM images of the PMN-PT nanoparticles. Scale bars are 3 μm and 1 μm. d) XRD pattern of PMN-PT ceramic with perovskite phase.

Table 1. Comparison of the mechanical strength and piezoelectricity with different tough piezoelectric materials.

Materials	Tensile modulus (MPa)	Tensile strength (MPa)	Piezoelectric coefficient (pm/V)	References
BT-PVDF composites	368	12	9.4	1
CNT@3-3-3 composites	28	×	120	2
Piezoelectric metamaterials	340	7	150	3
Piezoceramic textile	350	5	190	4
Piezoelectric gel	13.5	12	62	5
Heterostructure glycine-PVA film	4000	10	5.3	6
Gradient structure	830	55	15.5	This work

References:

1. T. Kowalchik, F. Khan, K. Le, P. Leland, S. Roundy and R. Warren, *Nano Energy*, 2023, **109**, 108276.
2. F. Yang, J. Li, Y. Long, Z. Zhang, L. Wang, J. Sui, Y. Dong, Y. Wang, R. Taylor, D. Ni, W. Cai, P. Wang, T. Hacker and X. Wang, *Science*, 2021, **373**, 337-342.
3. T. Tang, Z. Shen, J. Wang, S. Xu, J. Jiang, J. Chang, M. Guo, Y. Fan, Y. Xiao, Z. Dong, H. Huang, X. Li, Y. Zhang, D. Wang, L-Q. Chen, K. Wang, S. Zhang, C-W. Nan and Y. Shen, Natl. Sci. Rev., *2023*, nwas177.
4. Y. Hong, B. Wang, Z. Long, Z. Zhang, Q. Pan, S. Liu, X. Luo and Z. Yang, *Adv. Funct. Mater.*, 2021, **31**, 2104737.
5. Q. Li, L. Chen, M. Guo and Z. Hu, *Adv. Mater. Technol.*, 2022, **7**, 2101371.
6. J. Li, F. Yang, Y. Long, Y. Dong, Y. Wang and X. Wang, *ACS Nano*, 2021, **15**, 14903-14914.