

## Supplementary Information

### Machine Learning-Enabled Planar Interdigitated Thermogalvanic Hydrogel for Synergistic Thermal and Strain Sensing

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## **Inventory of Supplementary Information:**

### **Supplementary Figures and tables**

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| <b>Supplementary Fig. S3</b>  | <b>Ti 2p XPS spectra of MXene electrodes immediately after printing and after ambient storage for three days.</b>  |
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| <b>Supplementary Fig. S6</b>  | <b>Time-dependent voltage response to various temperature gradients for (a) freshly prepared and (b) after 8 hours of ambient exposed PITH device. Thermopower fitting of the PITH device: (c) freshly prepared device and (d) device after 8 h of ambient exposure.</b> |
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| <b>Supplementary Fig. S8</b>  | <b>(a) Photograph of the electrical conductivity measurement using a four-point probe. (b) Variation of the electrical resistance (R) of the MXene electrode as a function of bending cycles.</b>  |
| <b>Supplementary Fig. S9</b>  | <b>Boxplot of voltage responses of the PITH under transient touch and prolonged touch.</b>   |
| <b>Supplementary Fig. S10</b> | <b>Photographs demonstrating the PITH device attached to a gloved finger during the handwriting of different letters.</b>  |
| <b>Supplementary Fig. S11</b> | <b>Voltage response curves of handwritten digit “0” under (a) no-temperature-gradient and (b) temperature-gradient</b>   |

conditions. (c) Comparison of recognition accuracies for digit handwriting with and without a temperature gradient.

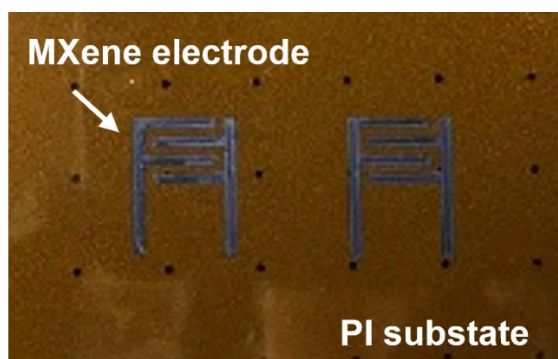
**Supplementary Table S1** Resistance of the printed MXene electrodes under different bending cycles.

**Supplementary Table S2** Comparison of the sensing performance between our PITH device and previously reported self-powered thermogalvanic sensors.

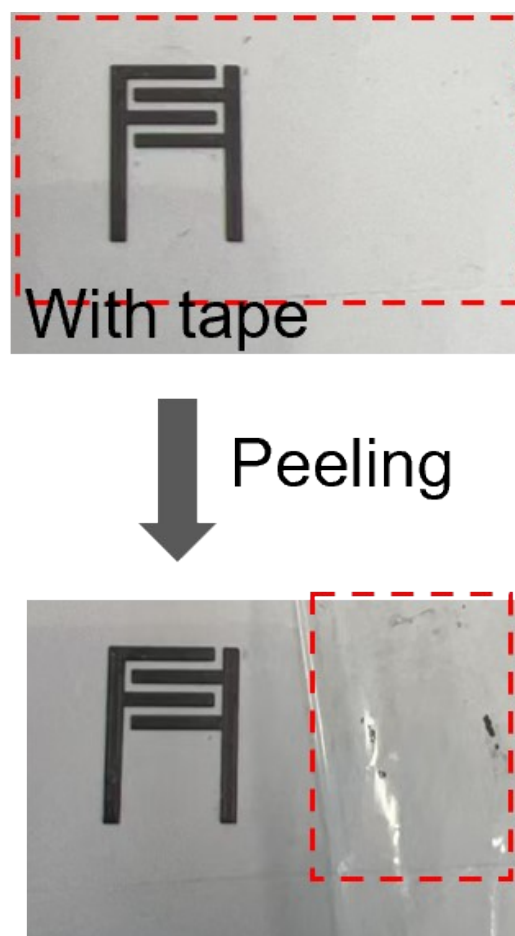
**Video S1.** Direct ink writing (DIW) printing process of interdigital MXene electrodes on a flexible PET substrate, demonstrating the continuous extrusion and pattern fidelity of the MXene ink during electrode fabrication.

**Video S2.** Appearance and fluidity of the MXene ink, illustrating its homogeneous dispersion and suitable rheological properties for direct ink writing.

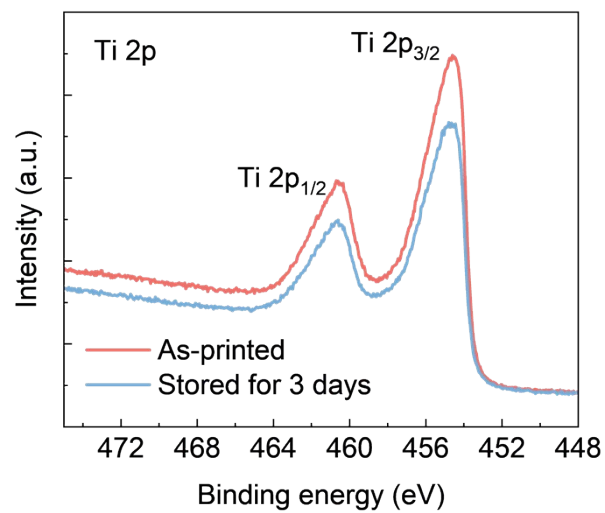
**Video S3.** Manual stretching and mechanical deformation of the PAAm hydrogel, demonstrating its high flexibility, elasticity, and mechanical robustness under tensile strain.



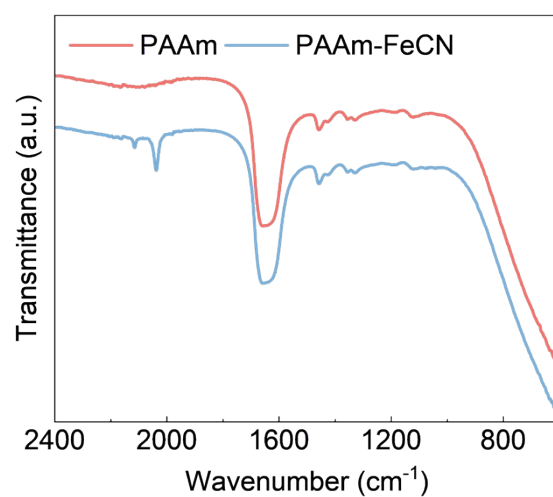
**Figure S1.** Photograph of the MXene electrode directly printed on a polyimide (PI) substrate.



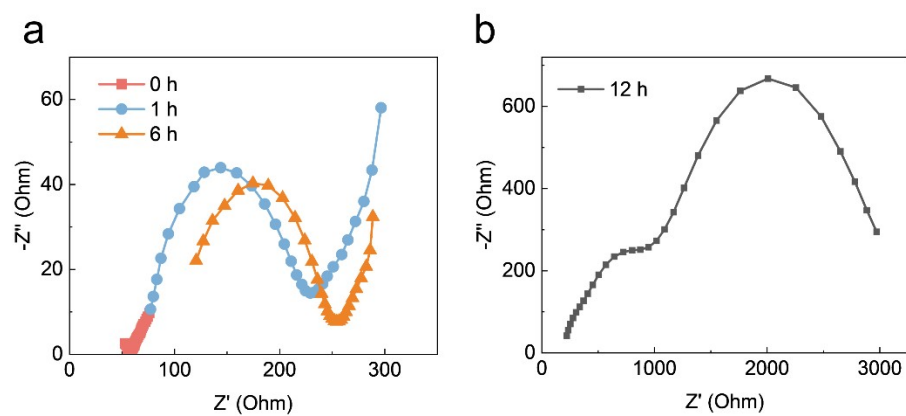
**Figure S2.** Adhesion test of the MXene electrode on the PET substrate using the tape-peeling method.



**Figure S3.** Ti 2p XPS spectra of MXene electrodes immediately after printing and after ambient storage for three days.

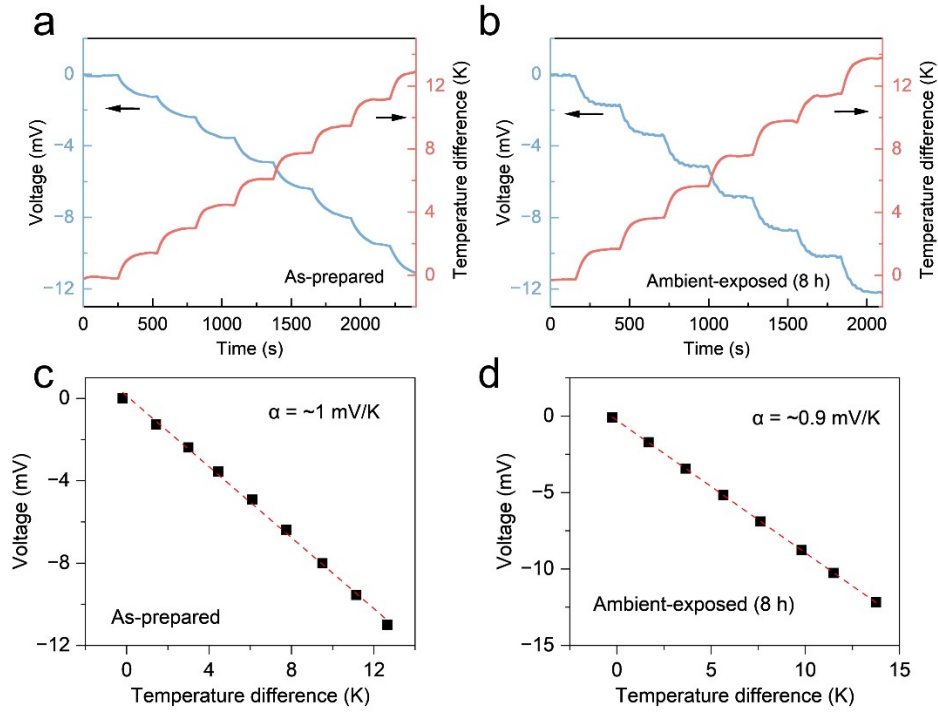


**Figure S4.** FTIR spectra of pure PAAm hydrogel and PAAm hydrogel containing redox ion pairs.

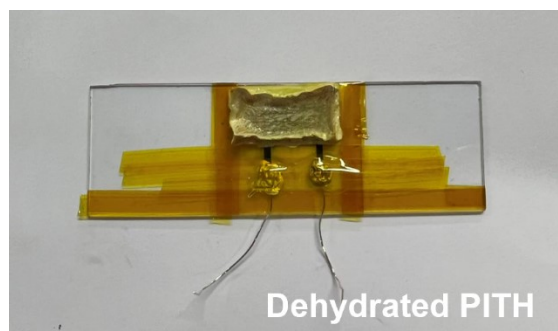


**Figure S5.** EIS spectra of the device at 0, 1, and 6 h. (b) EIS spectrum at 12 h.

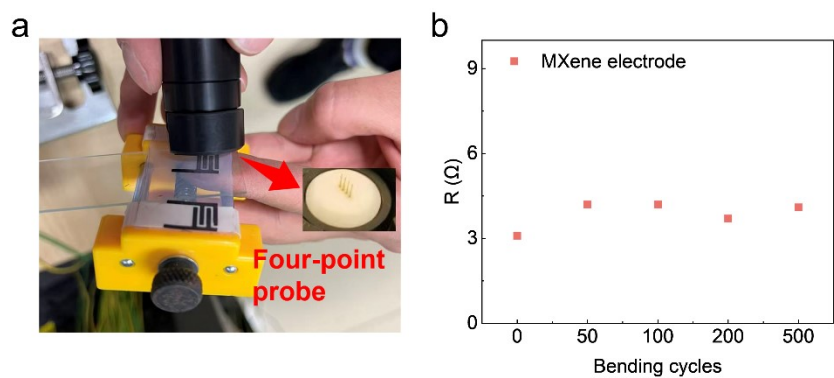




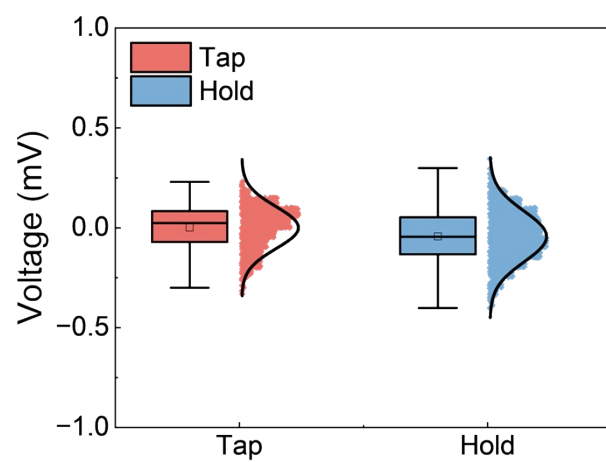
**Figure S6.** Time-dependent voltage response to various temperature gradients for (a) freshly prepared and (b) after 8 hours of ambient exposed PITH device. Thermopower fitting of the PITH device: (c) freshly prepared device and (d) device after 8 h of ambient exposure.



**Figure S7.** Photograph of the fully dehydrated PITH device after storage under ambient conditions for four days.



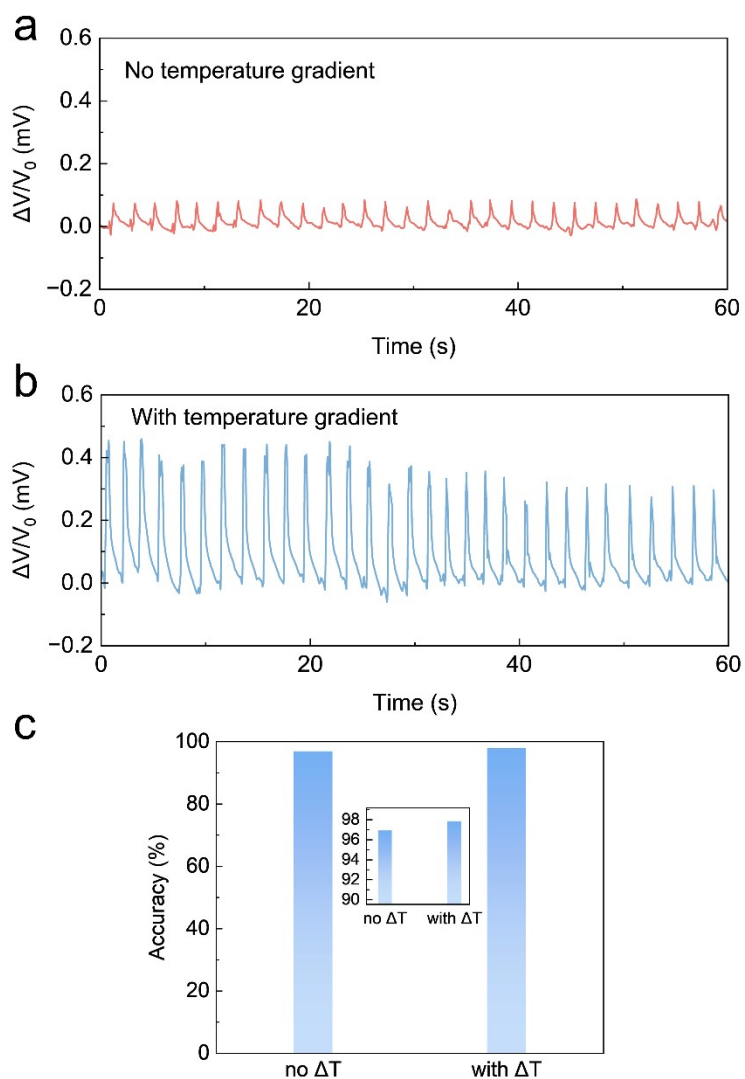
**Figure S8.** (a) Photograph of the electrical conductivity measurement using a four-point probe. (b) Variation of the electrical resistance ( $R$ ) of the MXene electrode as a function of bending cycles.



**Figure S9.** Boxplot of voltage responses of the PITH under transient touch and prolonged touch.



**Figure S10.** Photographs demonstrating the PITH device attached to a gloved finger during the handwriting of different letters.



**Figure S11.** Voltage response curves of handwritten digit "0" under (a) no-temperature-gradient and (b) temperature-gradient conditions. (c) Comparison of recognition accuracies for digit handwriting with and without a temperature gradient.

**Table S1.** Resistance of the printed MXene electrodes under different bending cycles.

| Bending cycles | R ( $\Omega$ ) | $\sigma$ (S cm <sup>-1</sup> ) |
|----------------|----------------|--------------------------------|
| 0              | 3.1            | 2932.6                         |
| 50             | 4.2            | 2164.5                         |
| 100            | 4.2            | 2164.5                         |
| 200            | 3.7            | 2457.0                         |
| 500            | 4.1            | 2217.3                         |

**Table S2.** Comparison of the sensing performance between our PITH device and previously reported self-powered thermogalvanic sensors.

| Device             | Materials  | Sensing modality          | Device configuration                  | Response time (ms) | thermopower (mV K <sup>-1</sup> ) | Ref.      |
|--------------------|--|---------------------------|---------------------------------------|--------------------|-----------------------------------|-----------|
| Gel Patch          | PVA/PDMS + Fe <sup>2+/3+</sup>                     | Photo-thermal-electric    | Flexible PTE gel patch                | 500                | 1.46                              | 1         |
| Antifreezing Gel   | PVA/EG + Fe(CN) <sub>6</sub> <sup>3-/4-</sup>      | Thermal energy harvesting | Sandwich structure                    | N/A                | 1.43                              | 2         |
| Fingertip Receptor | PVA + Fe(CN) <sub>6</sub> <sup>3-/4-</sup>         | Pressure & Thermal        | Micropatterned and gradient structure | 80                 | 1.89                              | 3         |
| TGH E-Skin         | PAM + LiCl + Fe(CN) <sub>6</sub> <sup>3-/4-</sup>  | Thermal (Signature)       | Sandwich structure                    | 230                | 1.82                              | 4         |
| Smart Pen          | PVA/Gelatin + Fe(CN) <sub>6</sub> <sup>3-/4-</sup> | Thermal & Piezoresistive  | Gel-wrapped pen structure             | 130                | 2.05                              | 5         |
| TGH Array          | PVA/Agar + Fe(CN) <sub>6</sub> <sup>3-/4-</sup>    | Thermal (Biometric)       | Concave-arranged array                | N/A                | 1.50                              | 6         |
| PITH               | PAAm + Fe(CN) <sub>6</sub> <sup>3-/4-</sup>        | Thermal-strain coupled    | Planar interdigitated                 | 390                | 1.44                              | This work |

\*The corresponding full name for abbreviation in the table as follows.

PVA: poly(vinyl alcohol), PDMS: polydimethylsiloxane, EG: ethylene glycol, PAM: polyacrylamide, TGH: thermogalvanic hydrogel



## References

- (1) Yang, H.; Ahmed Khan, S.; Li, N.; Fang, R.; Huang, Z.; Zhang, H. Thermogalvanic gel patch for self-powered human motion recognition enabled by photo-thermal-electric conversion. *Chem. Eng. J.* 2023, 473, 145247.
- (2) Zhang, D.; Zhou, Y.; Mao, Y.; Li, Q.; Liu, L.; Bai, P.; Ma, R. Highly antifreezing thermogalvanic hydrogels for human heat harvesting in ultralow temperature environments. *Nano Lett.* 2023, 23 (23), 11272-11279.
- (3) Li, N.; Wang, Z.; Niu, Y.; Li, Y.; Wen, S.; Zhang, H.; Lin, Z. H. Bionic Multimodal Augmented Somatosensory Receptor Enabled by Thermogalvanic Hydrogel. *Adv. Sci.* 2025, e05873.
- (4) Li, N.; Wang, Z.; Yang, X.; Zhang, Z.; Zhang, W.; Sang, S.; Zhang, H. Deep - learning - assisted thermogalvanic hydrogel e - skin for self - powered signature recognition and biometric authentication. *Adv. Funct. Mater.* 2024, 34 (18), 2314419.
- (5) Sang, S.; Bai, C.; Wang, W.; Khan, S. A.; Wang, Z.; Yang, X.; Zhang, Z.; Zhang, H. Finger temperature-driven thermogalvanic gel-based smart pen: Utilized for identity recognition, stroke analysis, and grip posture assessment. *Nano Energy* 2024, 123, 109366.
- (6) Zhang, D.; Xu, Z.; Wang, Z.; Cai, H.; Wang, J.; Li, K. Machine-learning-assisted wearable PVA/Acrylic fluorescent layer-based triboelectric sensor for motion, gait and individual recognition. *Chem. Eng. J.* 2023, 478, 147075.