Supporting Information

Mxene Reinforced Organohydrogel with Ultra-stable, High Sensitivity and Anti-freezing Ability for Flexible Strain Sensors

Sheng-Ji Wang ^{a, b}, Zhuo Chen ^{a, b}, Xiangshu Hu ^{a, b}, Jian Zou ^{a, b}, Zhihui Xie ^{a, b},

Hao-Yang Mi^{a, b, c}, Zi-Hao Liu^{a, b}, Zhi Zhang^d, Yinghui Shang^d, Xin Jing^{a, b*}

*Affiliation:

^a National & Local Joint Engineering Research Center for Advanced Packaging

Material and Technology, Hunan University of Technology, Zhuzhou 412007, PR

China.

^b Key Laboratory of Advanced Packaging Materials and Technology of Hunan Province, Hunan University of Technology, Zhuzhou, 412007, China.

° National Engineering Research Center for Advanced Polymer Processing

Technology, Zhengzhou University, Zhengzhou, 450000, China

^d Shenzhen Weijian Wuyou Technology Co., Ltd., Shenzhen 518102, Guangdong,

China.



Figure S1. SEM images of (a) MAX and (b) MXene, the scale bar is 2 μ m and

200nm. EDS element mapping of (c) MAX. FTIR of (d) MXene.



Figure S2. (a) Mechanical properties of PAM. Dissipate energy of (b) different hydrogels strain at 100% and (c) PPMOH sensor at 20% compression.



Figure S3. Peeling process between Ecoflex and PPMOH.

Based on Owens-Wendt method, the surface energy of Ecoflex, PPM and PPMOH can be calculated from the contact angle with the following equation ¹:

$$\gamma^{l}\left(1+\cos\theta\right) = 2\left(\gamma_{s}^{d}\gamma_{l}^{d}\right)^{1/2} + 2\left(\gamma_{s}^{p}\gamma_{l}^{p}\right)^{1/2}$$
(1)

Meanwhile, the solid surface energy can be expressed by equation $(2)^2$.

$$\gamma^s = \gamma_s^{\ d} + \gamma_s^{\ p} \tag{2}$$

where, θ is the contact angle on the flat surface, γ^{s} is the solid surface tension, γ^{l} is the liquid surface tension, γ_{s}^{d} and γ_{l}^{d} are the dispersive component of solid and liquid, γ_{s}^{p} and γ_{l}^{p} are the polar component of solid and liquid.

Choose water ($\gamma^{l} = 72.8 \text{ mJ/m}^{2}$, $\gamma_{l}^{p} = 51.0 \text{ mJ/m}^{2}$, $\gamma_{l}^{d} = 21.8 \text{ mJ/m}^{2}$) and ethylene glycol ($\gamma^{l} = 48.0 \text{ mJ/m}^{2}$, $\gamma_{l}^{p} = 19.0 \text{ mJ/m}^{2}$, $\gamma_{l}^{d} = 29.0 \text{ mJ/m}^{2}$) of known γ_{l}^{d} and γ_{l}^{p} , dropped them onto the solid surface to obtained contact angle respectively, then combined equation (1) and (2) to calculate surface energy γ^{s} by substituting contact angle, γ_{l}^{d} and γ_{l}^{p} .



Figure S4. The contact angle of water to (a) Ecoflex, (c) PPMOH, (e) PPM and Eg to (b) Ecoflex, (d) PPMOH, (f) PPM. (g) FTIR spectra of the Ecoflex formed on PS plate and the Ecoflex formed on PPMOH.



Figure S5. Morphology of (a) PPM and (b) PPMOH under -20 °C for 0 h, 2 h, 4 h, 8

h, 24 h.



Figure S6. Monitoring response of current changes to different model parts movements: (a) finger, (b) elbow, (c) knee, (d) wrist.



Figure S7. The weight loss of PPMOH sensor in 90 days.



Figure S8. 200 load-unload cycles under 20 % compress after immersing in

water for 24 h.

Table S1. Comparison in the properties of hydrogel-based strain sensors based on

| Flexible sensor composition | Long-term stability | Strain (%) | Response time (ms) | Sensitivity | Ref. |
|--|------------------------|---------------|-----------------------|--|--|
| PAM/PEDOT:PSS/MX ene organohydrogel | 90 days | 891 | 100 | 0-300%, 4.71 300-500%, 10.69 | This work |
| PVA/CNF organohydrogel | 30 days | 696 | 130 | 0-100%, 0.96 100-300%, 1.57 | Carbohyd. Polym, 2022 ³ |
| TA@CNF/PAAm/MXe ne organohydrogel | 7 days | 1500 | - | 0-250%, 4.15 250-500%, 8.21 | Adv. Funct. Mater, 2020 ⁴ |
| PDA–rGO/ SA/PAM organohydrogel | - | 312 | 200 | 0-250%, 2.09 | J. Mater. Chem. C, 2021 ⁵ |
| PVA/gelatin/TA@CNC -Al ³⁺ organohydrogel | 20 days | 519.7 | 240 | 0-100%, 1.86 100-250%, 2.64 250-400%, 4.23 | Compos Part B- Eng, 2021 ⁶ |
| PVA/MXene/ PEDOT:PSS/PDA | - | 650 | 630 | 0-500%, 2.55 | J. Mater. Chem. A, 2021 ⁷ |
| PVA/ MXene/Zn ²⁺ organohydrogel | - | 247 | - | 0-50%, 3.42 50-100%, 4.77 100-180%, 5.82 | Adv. Funct. Mater, 2021 ⁸ |

different materials.

References

- X. Lu, Z. Zhao and Y. Leng, *Materials Science and Engineering: C*, 2007, 27, 700-708.
- A. Kozbial, Z. Li, C. Conaway, R. McGinley, S. Dhingra, V. Vahdat, F. Zhou,
 B. D'Urso, H. Liu and L. Li, *Langmuir : the ACS journal of surfaces and colloids*, 2014, 30, 8598-8606.
- 3. M. Li, D. Chen, X. Sun, Z. Xu, Y. Yang, Y. Song and F. Jiang, *Carbohydrate polymers*, 2022, **284**, 119199.
- Y. Wei, L. Xiang, H. Ou, F. Li, Y. Zhang, Y. Qian, L. Hao, J. Diao, M. Zhang,
 P. Zhu, Y. Liu, Y. Kuang and G. Chen, *Advanced Functional Materials*, 2020,
 30, 2005135.
- Z. Xie, H. Li, H.-Y. Mi, P.-Y. Feng, Y. Liu and X. Jing, J. Mater. Chem. C, 2021, 9, 10127-10137.
- 6. J. Yin, C. Lu, C. Li, Z. Yu, C. Shen, Y. Yang, X. Jiang and Y. Zhang, Composites Part B: Engineering, 2022, 230, 109528.
- W. Zhao, D. Zhang, Y. Yang, C. Du and B. Zhang, *Journal of Materials Chemistry A*, 2021, 9, 22082-22094.
- Y. Feng, H. Liu, W. Zhu, L. Guan, X. Yang, Andrei V. Zvyagin, Y. Zhao, C.
 Shen, B. Yang and Q. Lin, *Adv. Funct. Mater*, 2021, **31**, 2105264.