

Supporting information

High-Strength, Ultra-Tough and Recyclable MXene-Composited Organohydrogels with Integrated Multi-Functions for Wearable Sensor

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1. Experimental Section

1.1 Mechanical tests

The mechanical properties of all gel samples were tested multi-functional mechanical tester (Shimadzu AGS-X, Japan) with a 100 N load cell. For tensile test, the sample was made into a dumbbell shape with a width of 2 mm and a thickness of 1 mm and the stretching rate was fixed at 50 mm/min. For compression tests, the gel samples were made into cylinders with diameter of 8 mm and height of 7 mm, and a speed of 5 mm/min was applied. For tearing test, the gel samples were proportionally cut into trouser shape (1/2 sized, width: 7.5 mm, length: 50 mm, notch length: 20 mm) and the rate of extension was fixed at 5 mm/min. Each sample was tested at least 3 times in parallel.

1.2 Fabrication and characterization of the organohydrogel-based sensing performances

In order to test the strain-sensing performance, a rectangular organohydrogel with a thickness of 1 mm and a width of 5 mm was stretched by a multi-functional mechanical tester, and the dynamical electrical signal was synchronously recorded by LCR meter (E4980AL, Keysight). For the practical application, the two ends of the organohydrogel were connected by wires encapsulated with transparent acrylic tapes, which was attached to the human body to monitor the human body movement.

1.3 Environmental stability measurement

The gel sample was placed at in an environment with a temperature of 23 °C and a relative humidity of about 40-50% to test the long-term stability. The weight of the sample at the same time interval was recorded and the retained weight was calculated. After the storage of 20 d, the tensile properties of the gel sample and sensing application were evaluated.

Differential scanning calorimetry (DSC) was used to analyze freezing resistance of the gels. The sample was first equilibrated at 25 °C, then cooled to -50° C for keeping 30 minutes, finally heated to the initial 25 °C, and the rate of temperature change was 2 °C/min. The tensile properties of the gel at low temperature were tested with a

multifunctional testing machine (Shimadzu AGS-X, Japan) equipped with a thermostatic control box. All samples are pre-cooling for 30 minutes at the test temperature and each sample was tested at least 3 times in parallel. The sensing properties of the gel was tested in low-temperature environment.

1.4 Recycling performance test

To investigate retrievability of the PTM organohydrogel, the samples were first cut into pieces using scissors. Then, the fractured fragments were put into a syringe and placed at 110°C for 1 hour to obtain a homogeneous solution. The solution was then transferred into designed molds. After the temperature dropped to room temperature, the recycled organohydrogel can be obtained. The mechanical properties and sensing performance of the recycled organohydrogel was estimated.

1.5 UV protection properties

The transmittance of the samples was measured in the wavelength range of 200–800 nm using UV-vis spectroscopy (Thermo EVOLUTION 201). All experimental samples sizes were 20 mm × 10 mm × 1 mm. The UV-shielding test was conducted on the back skin of male SD rats, which were purchased from Yishengyuan (Tianjin) Biotechnology Co., Ltd, China. The procedures for performing the animal test were approved by the Ethics Committee of Yanshan University. Specifically, three SD rats with a weight of 25–30 g were used for the test. First, the dorsal skin surface was shaved and cleaned using the razor prior to testing. The back skin of each rat was divided into two sections: (1) PTM organohydrogel-treated area (area 20 mm*20 mm, thickness 1 mm) and (2) PVA organohydrogel-treated area (area 20 mm*20 mm, thickness 1 mm). After The rats were anaesthetized by receiving an intraperitoneal injection of pentobarbital, the two sections were exposed to ultraviolet light (40 mW cm⁻², 365 nm) for 1 h. For histopathological examination, these skin tissues were collected and fixed in formalin solution (10%). The samples were stained with H&E and the images were observed using an optical microscope (BX51, Olympus, Japan).

2. Supporting Figures

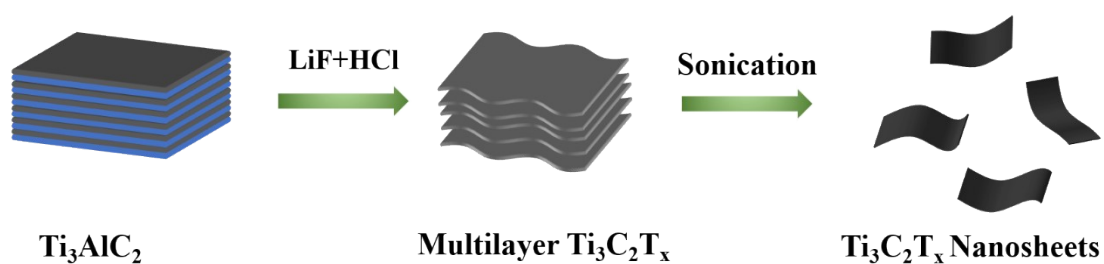


Figure S1. Schematic synthesis of $\text{Ti}_3\text{C}_2\text{T}_x$ (MXene) nanosheets.

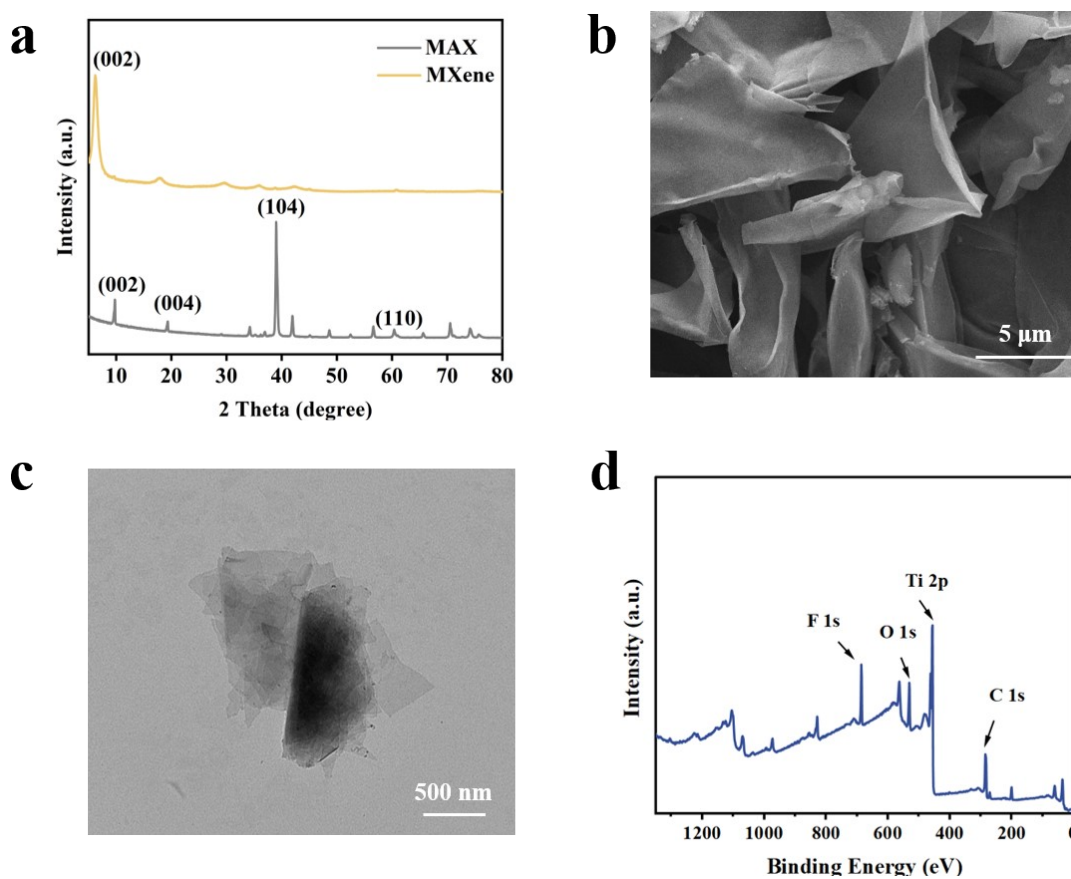


Figure S2. (a) XRD spectra of Ti_3AlC_2 MAX and $\text{Ti}_3\text{C}_2\text{T}_x$ MXene. (b) SEM and (c) TEM images of MXene nanosheets. (d) XPS survey spectrum of as-prepared MXene nanosheets with characteristic peaks of F 1s, O 1s, Ti 2p and C 1s.

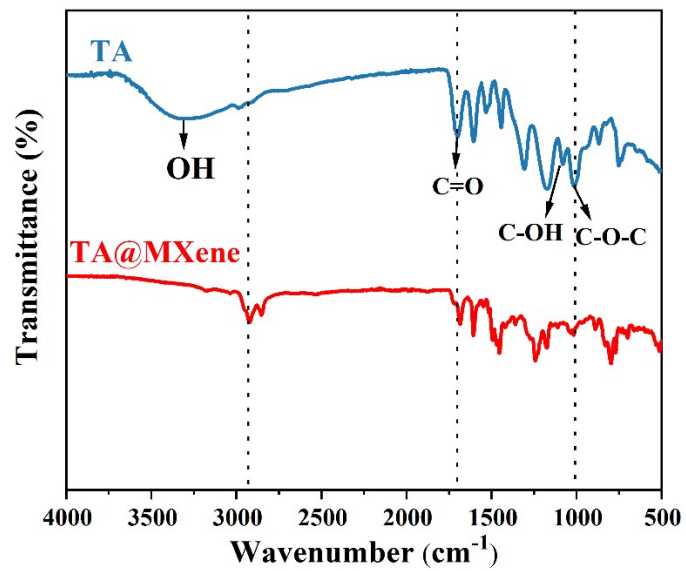


Figure S3. FTIR spectra of TA, and TA@MXene.

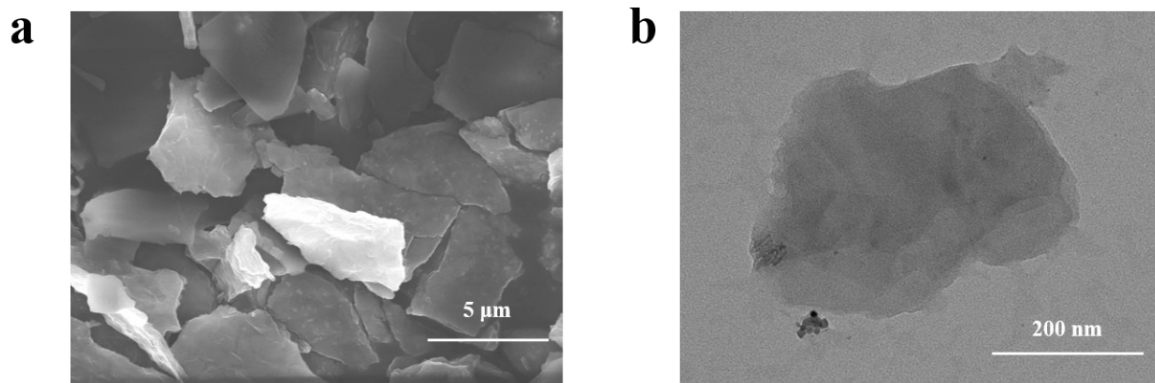


Figure S4. (a) SEM and (b) TEM images of TA@MXene nanosheets.

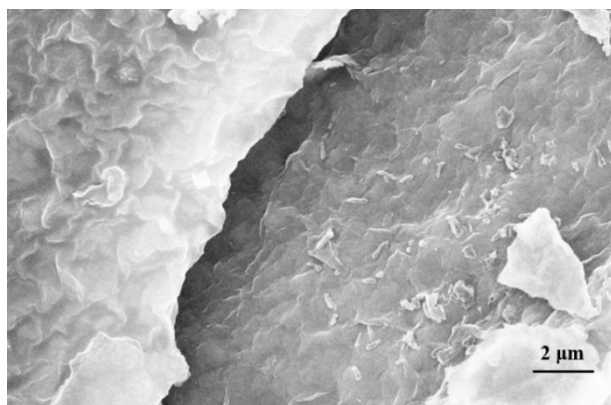


Figure S5. SEM image of TA@MXene after being stored for 65 days

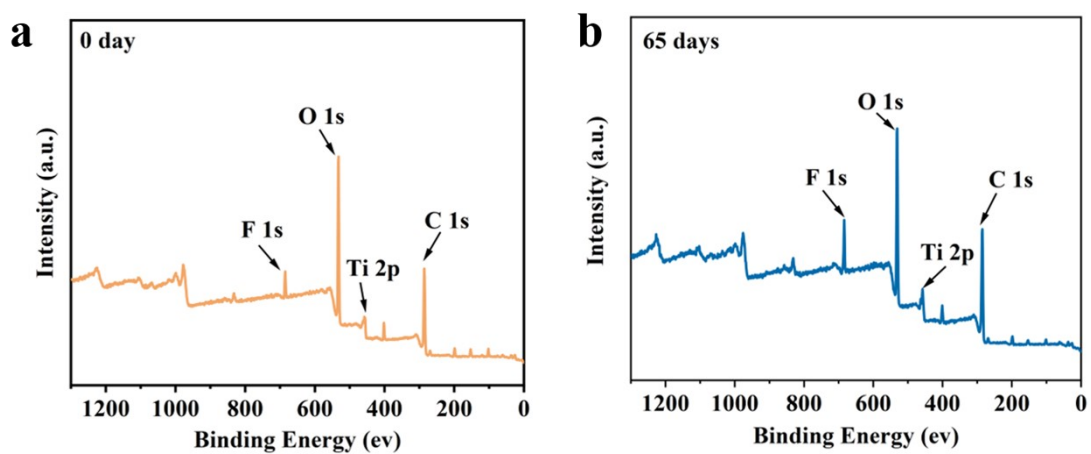


Figure S6. XPS survey spectrum of TA@MXene before and after being stored for 65 days with characteristic peaks of F 1s, O 1s, Ti 2p and C 1s.

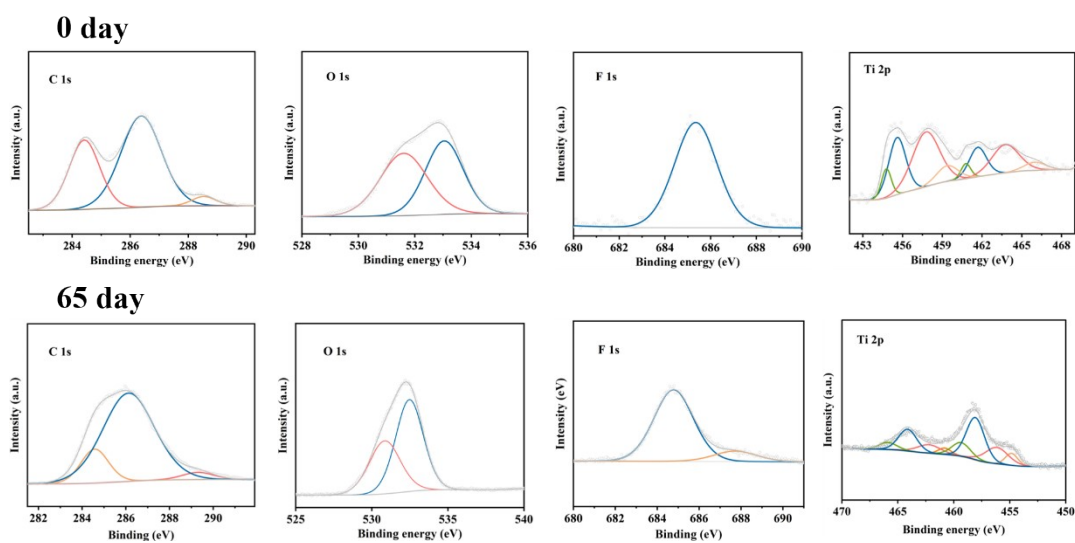


Figure S7. XPS spectra of O1s, C1s, F 1s and Ti 2p for TA@MXene being stored in aqueous dispersion before and after 65 days, respectively.

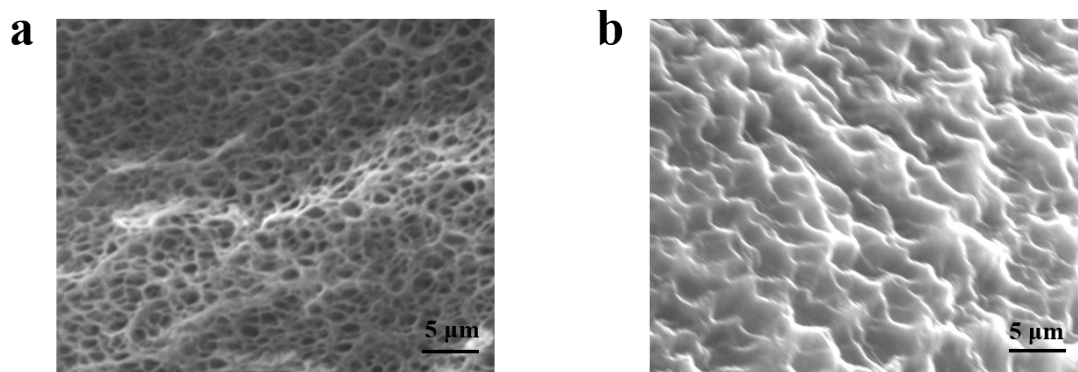


Figure S8. SEM images of (a) PVA hydrogel and (b) organohydrogel.

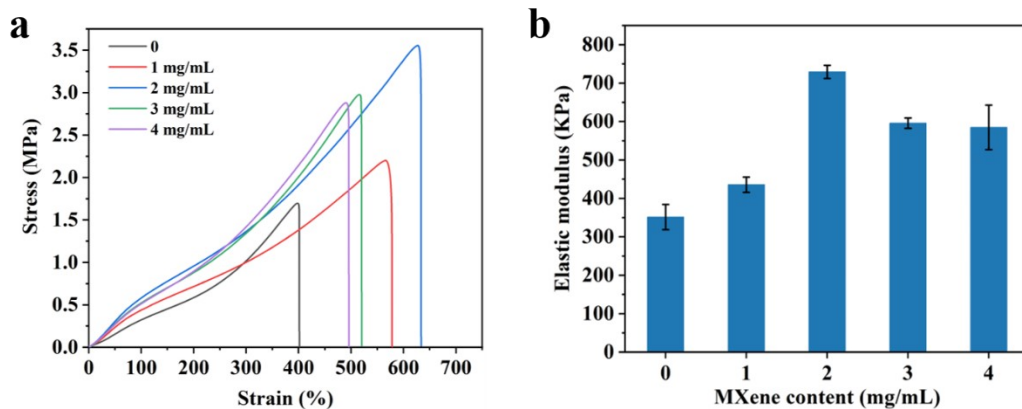


Figure S9. (a) Tensile stress-strain curves and (b) the corresponding elastic modulus of the PTM organohydrogels with different TA@MXene concentration.

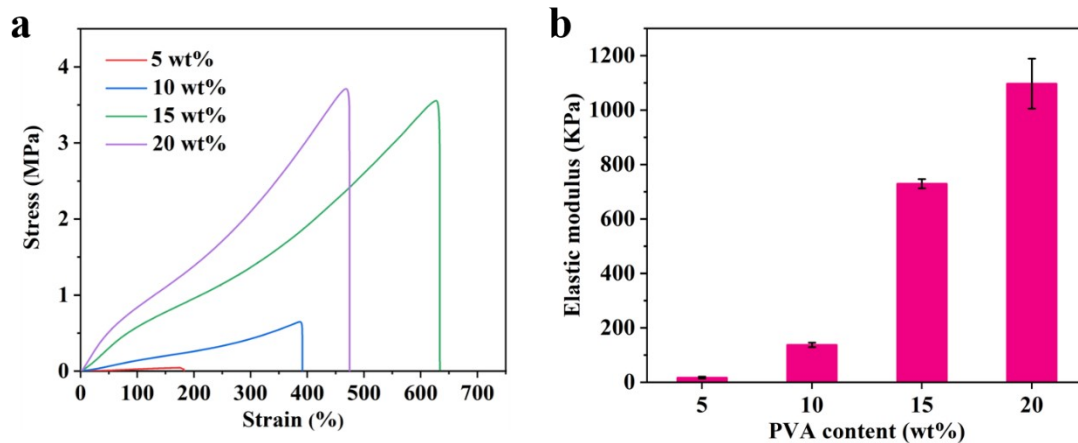


Figure S10. (a) Tensile stress-strain curves and (b) the corresponding elastic modulus of the PTM organohydrogels with different PVA content.

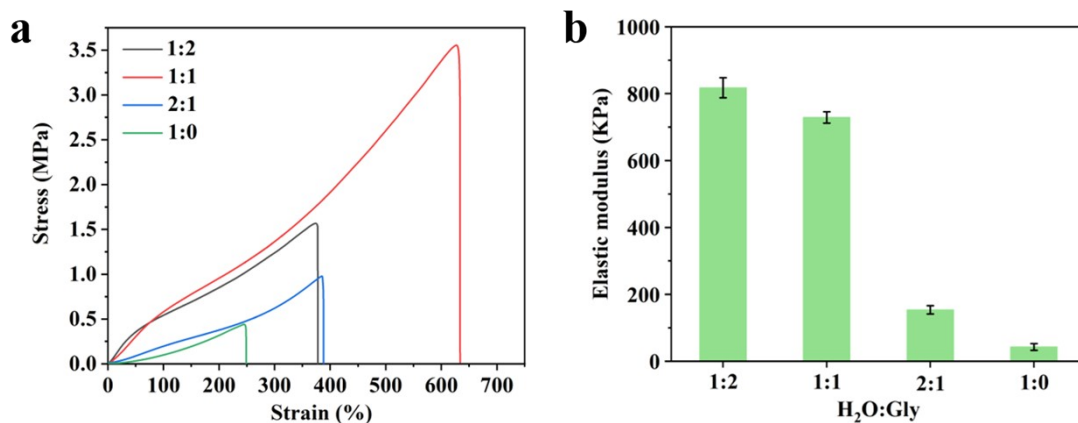


Figure S11. (a) Tensile stress-strain curves and (b) the corresponding elastic modulus of the PTM organohydrogel with water/glycerol ratios.

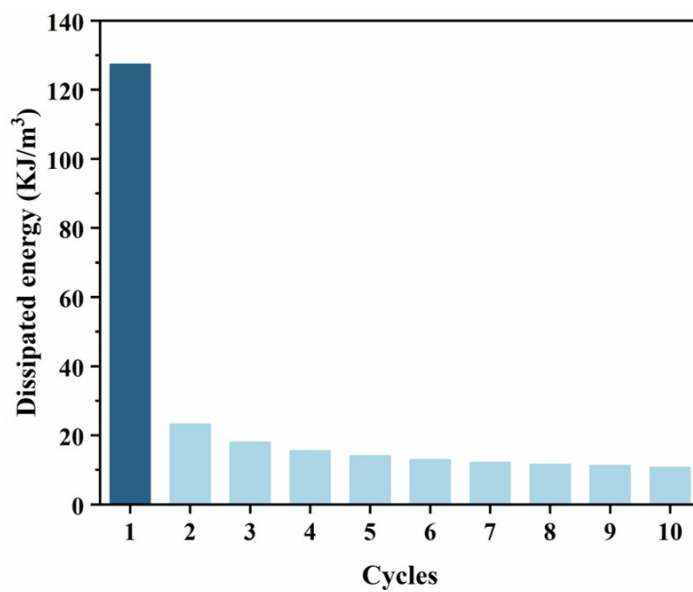


Figure S12. The dissipated energy of the PTM organohydrogel during ten successive loading-unloading process at maximum strain of 100%.

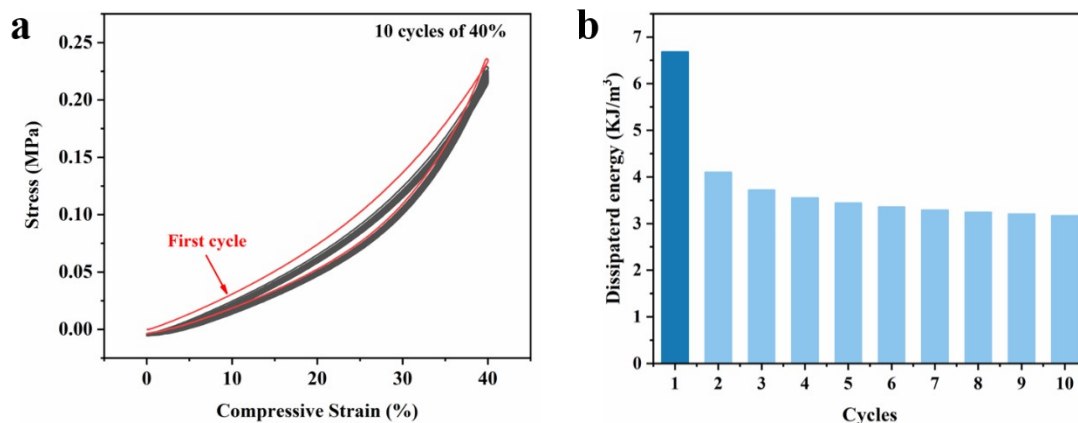


Figure S13. (a) Ten successive cyclic compressive curves and (b) the corresponding a dissipated energy of the PTM organohydrogel at maximum strain of 40%.

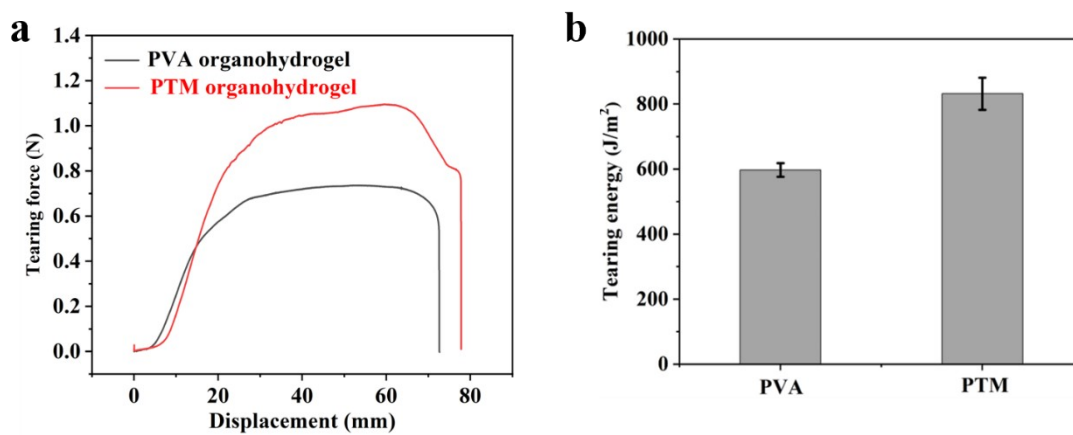


Figure S14. (a) Tearing curves and (b) the corresponding tearing energy of PVA and PTM organohydrogels.

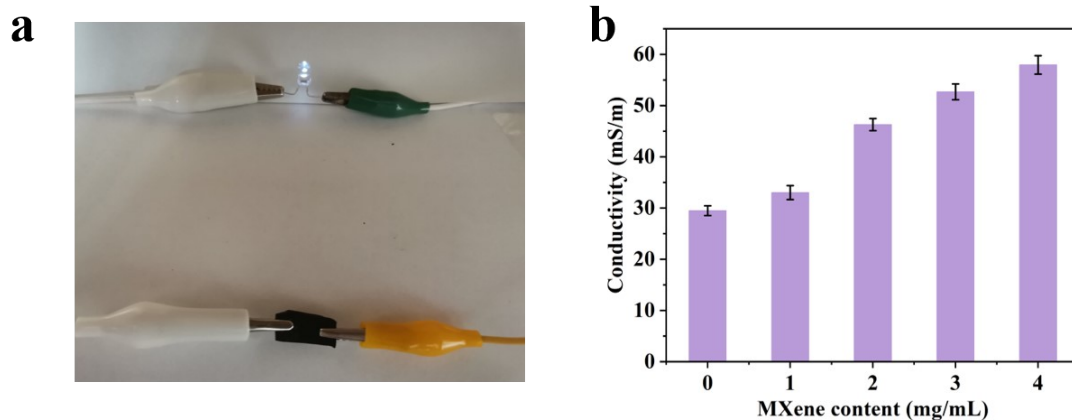


Figure S15. (a) Photo showing the luminance of LEDs using the PTM organohydrogel as a conductor in series circuits. (b) Conductivity of PTM organohydrogel with different MXene concentration.

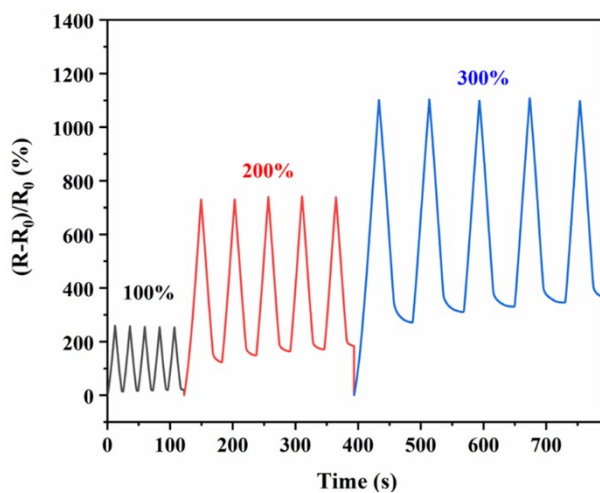


Figure S16. Relative resistance changes ($\Delta R/R_0$) of PTM organohydrogel under cyclic stretching–releasing at large strains (50, 100, and 200%).

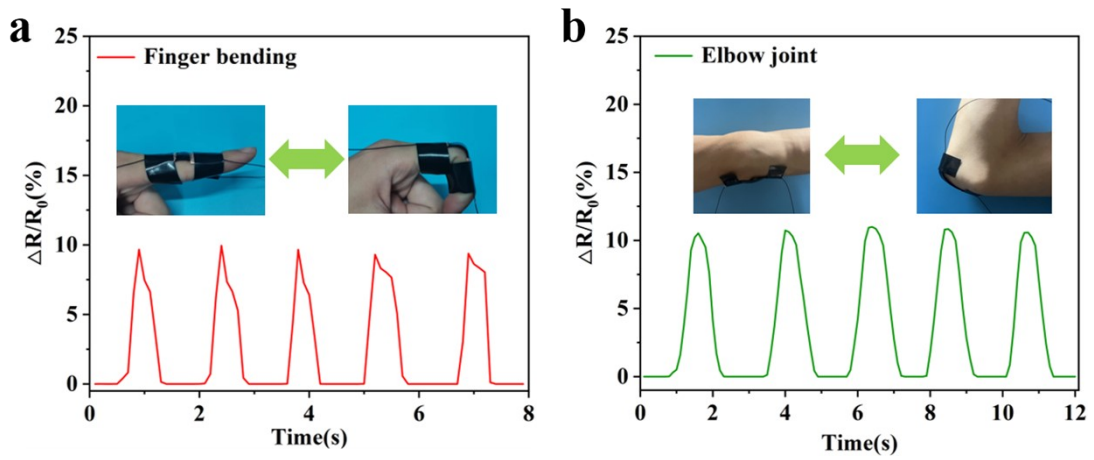


Figure S17. Relative resistance changes of the PTM organohydrogel sensor in response to (a) repetitive finger bending and (b) elbow movement.

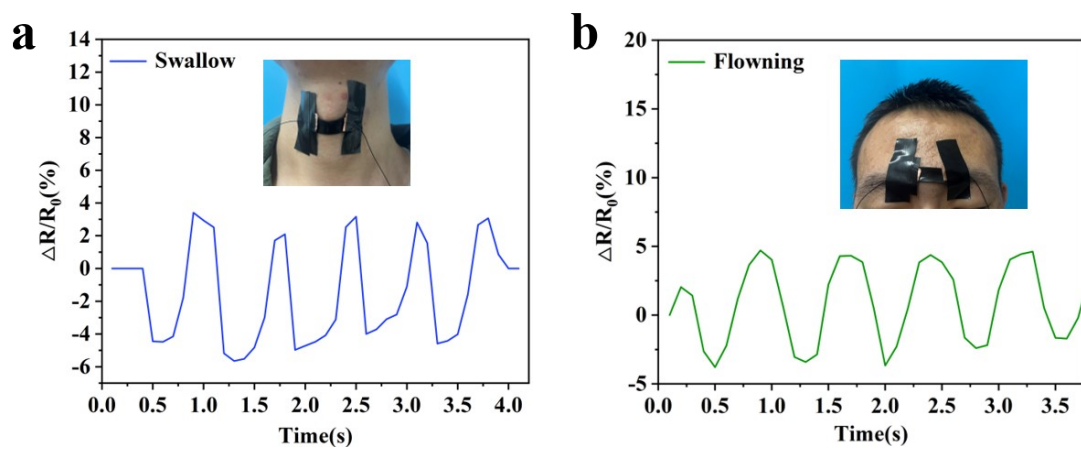


Figure S18. Continuous monitoring the subtle action of throat during (a) swallowing and (b) flowing.

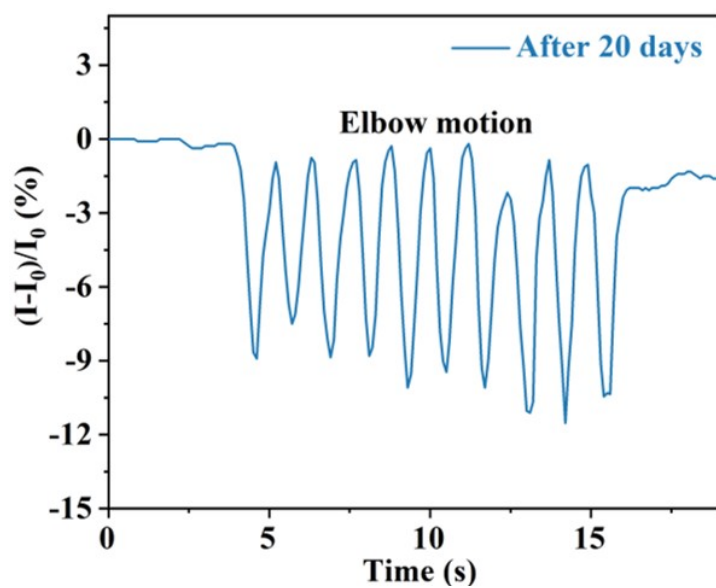


Figure S19. Sensing test of the PTM organohydrogel sensor after storing for 7 days.

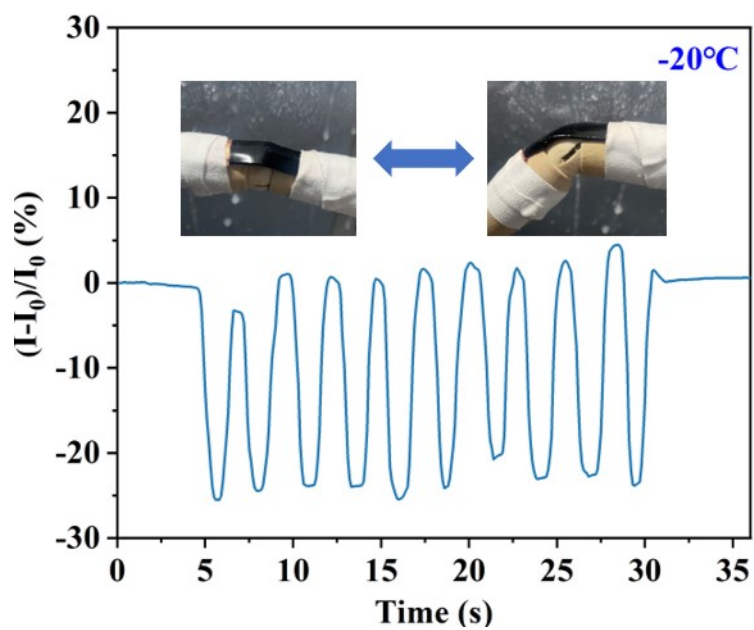


Figure S20. Sensing test of the PTM organohydrogel sensor at -20 °C.