

Supplementary Information for

Versatile MXene Gels Assisted by Brief and Low-Strength Centrifugation

Weiyan Yu^{1,2}, Yi Yang^{1,2}, Yunjing Wang², Lulin Hu^{1,2}, Jingcheng Hao^{2,3,*}, Lu Xu^{1,2,*} and Weimin Liu^{1,2}

¹ State Key Laboratory of Solid Lubrication, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, P. R. China

² Shandong Laboratory of Advanced Materials and Green Manufacturing at Yantai, Yantai 264006, P. R. China

³ Key Laboratory of Colloid and Interface Chemistry & Key Laboratory of Special Aggregated Materials, Shandong University, Jinan 250100, P. R. China

*Corresponding authors. E-mail: jhao@sdu.edu.cn (Jingcheng Hao); xulu@licp.cas.cn (Lu Xu)

Supplementary Figures and Tables

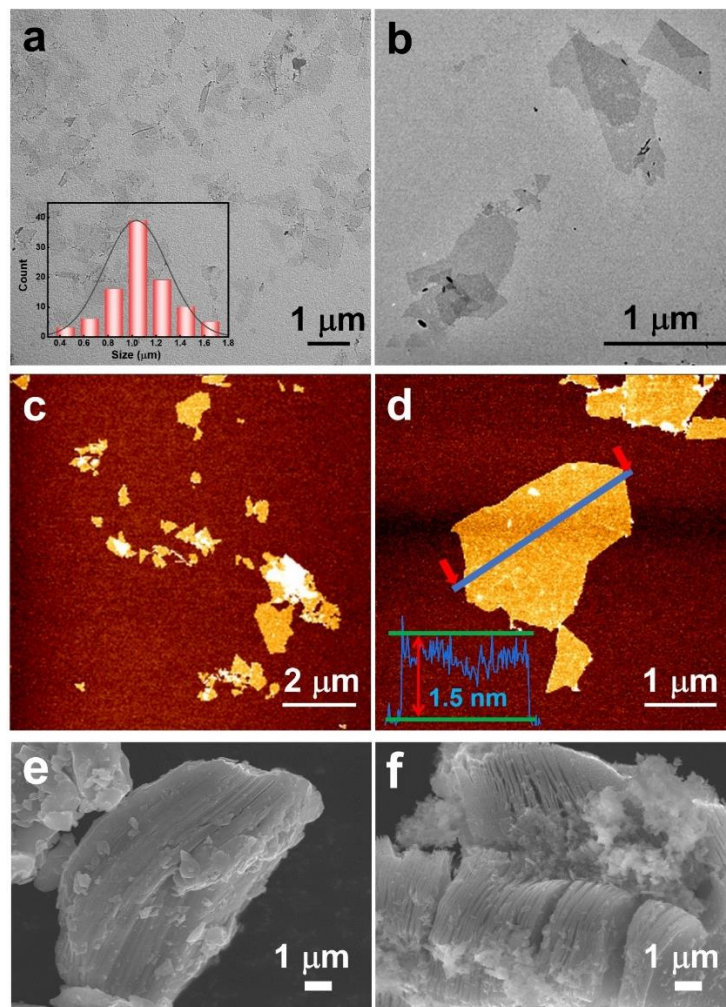


Fig. S1 **a, b** TEM and **c, d** AFM observations of exfoliated ultrathin $\text{Ti}_3\text{C}_2\text{T}_x$ MXene. SEM images of **e** pristine Ti_3AlC_2 MAX phase and **f** multilayered $\text{Ti}_3\text{C}_2\text{T}_x$ MXene obtained after etching. Insets in **a** and **d** are the particle size distribution and height profile of the MXene nanosheets

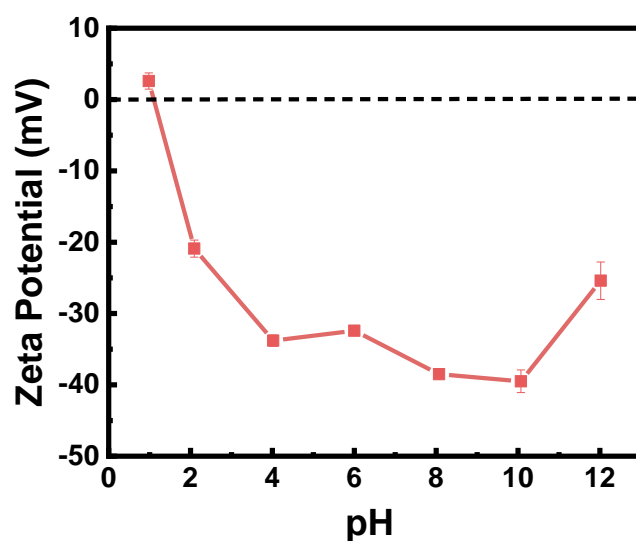


Fig. S2 Zeta potential measurements of $\text{Ti}_3\text{C}_2\text{T}_x$ dispersions at different pH values. $T = 25\text{ }^\circ\text{C}$

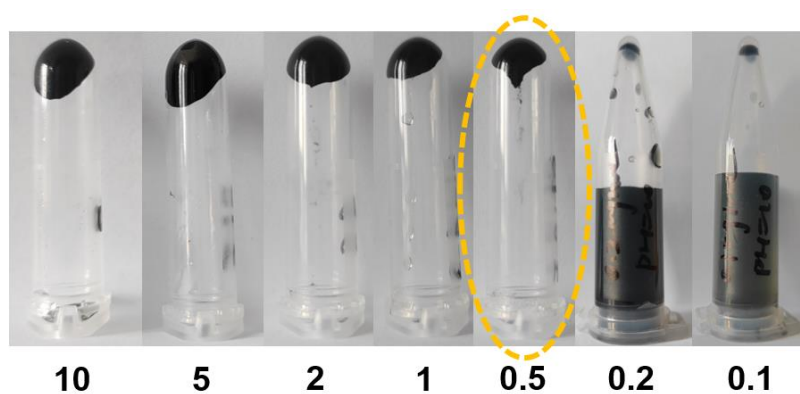


Fig. S3 Photographs of pH 10 $\text{Ti}_3\text{C}_2\text{T}_x$ dispersions at different particle concentrations (in mg mL^{-1}) after centrifugation at $10000 \times g$ for 30 s. $T = 25\text{ }^\circ\text{C}$

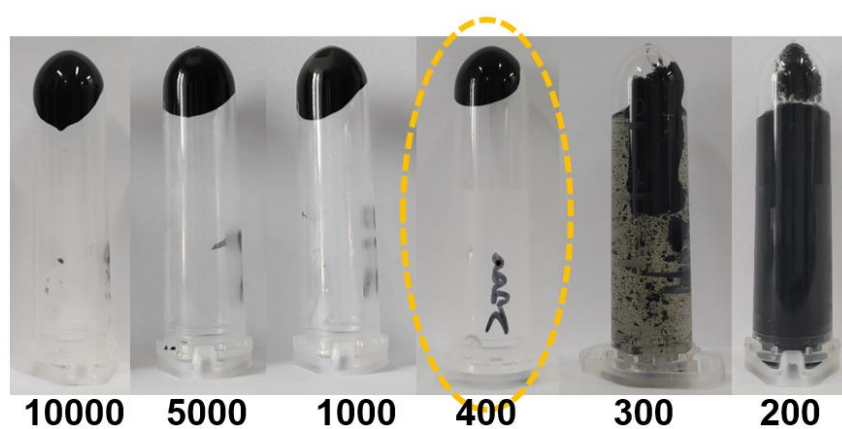


Fig. S4 Photographs of pH 10, 0.5 mg mL^{-1} $\text{Ti}_3\text{C}_2\text{T}_x$ dispersions after exposure to different relative centrifugal forces (RCFs, in $\times g$) for 30 s. $T = 25\text{ }^\circ\text{C}$

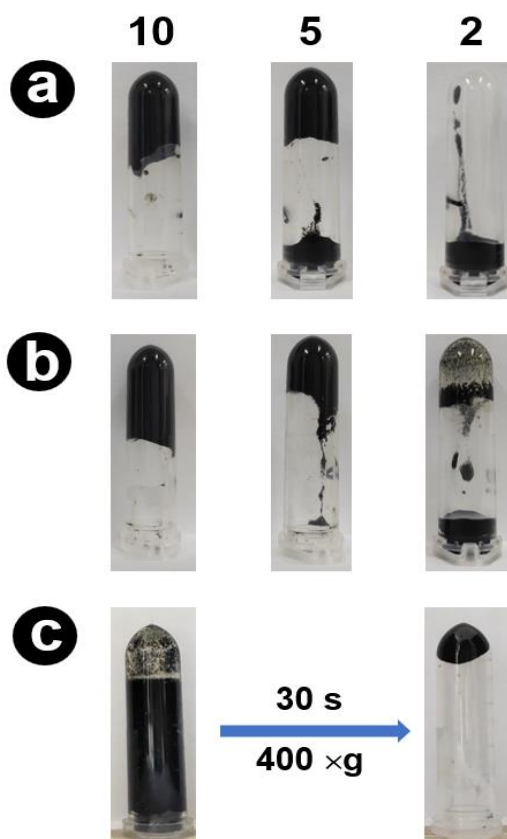


Fig. S5 Photographs of pH 10 $\text{Ti}_3\text{C}_2\text{T}_x$ dispersions at different particle concentrations (in mg mL^{-1}) after addition of **a** Fe^{2+} and **b** Zn^{2+} to a final mass ratio of 3:8 (metal chloride-to-MXene). **c** Photographs of an aqueous dispersion containing 2 mg mL^{-1} $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets and 5 mmol L^{-1} Fe^{2+} before and after exposure to $400 \times g$ centrifugation for 30 s. $T = 25 \text{ }^\circ\text{C}$

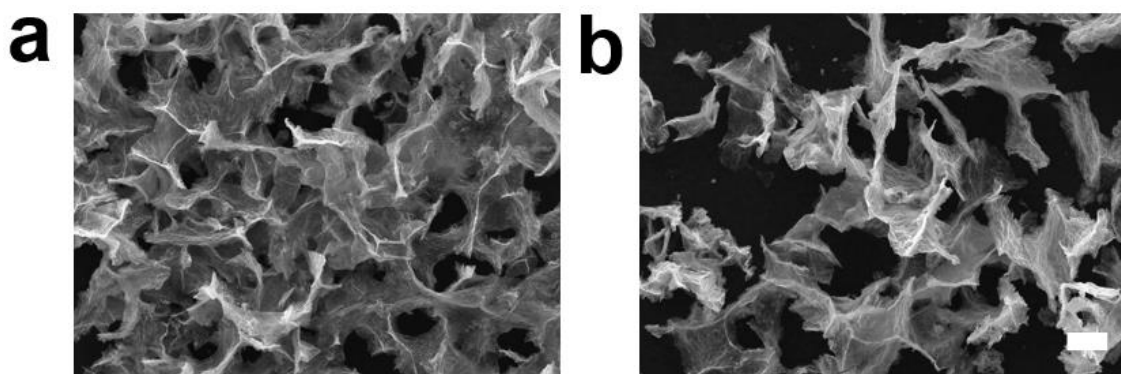


Fig. S6 SEM observations of freeze-dried pH 10 MXene gels **a** and dispersions **b**. Scale bar = $20 \text{ }\mu\text{m}$

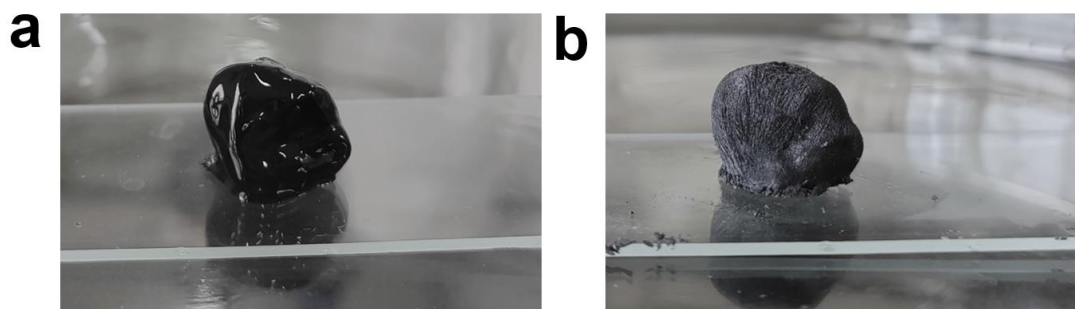


Fig. S7 3D macrostructures of a pH 10 MXene gel **a** before and **b** after complete lyophilization

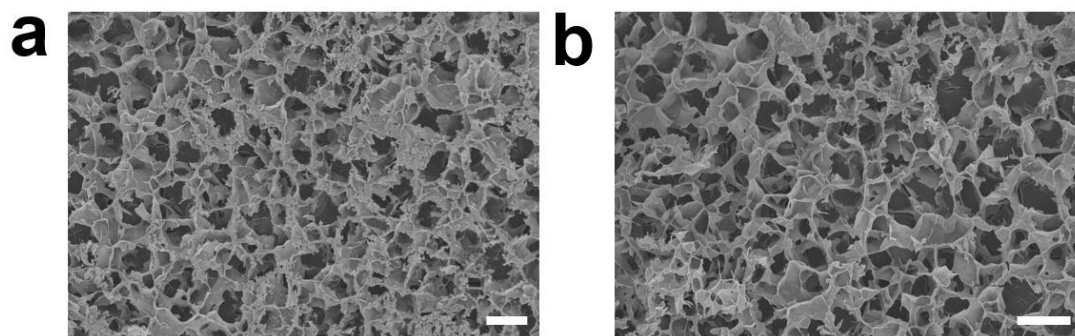


Fig. S8 Cryogenic SEM observations on a pristine pH 10 MXene gel. Scale bar = 5 μm

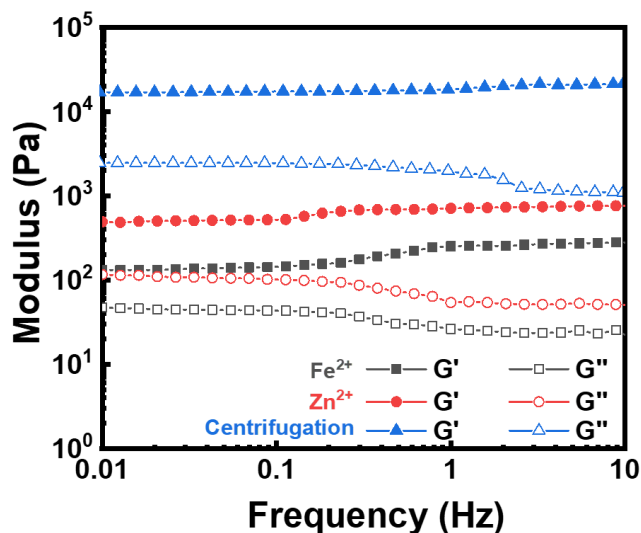


Fig. S9 Comparison in viscoelasticity between the $\text{Ti}_3\text{C}_2\text{T}_x$ gels with an identical water content of ~ 98 wt% triggered by centrifugation and divalent metal ions. $T = 25$ $^\circ\text{C}$

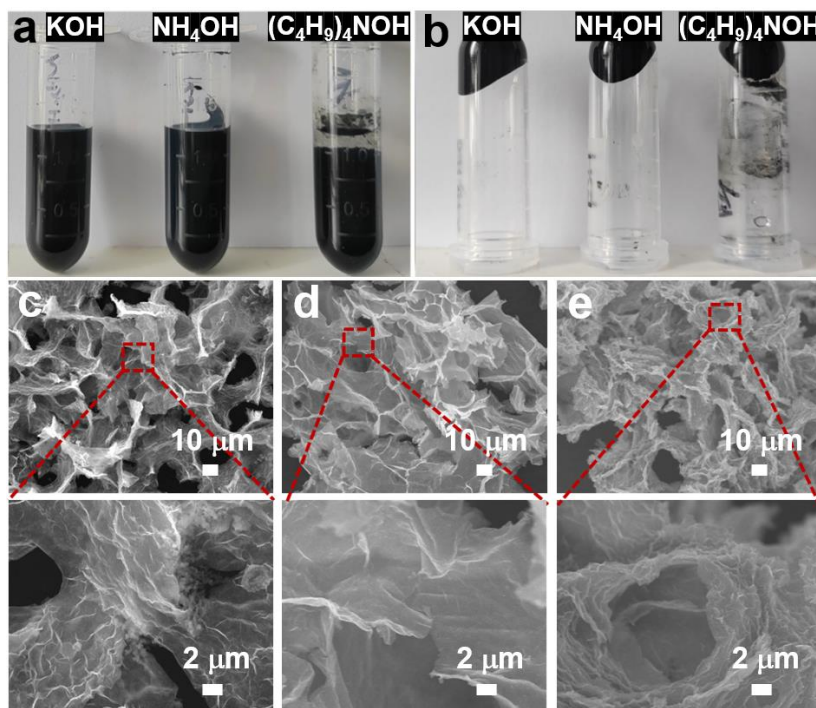


Fig. S10 Photographs of 10 mg mL^{-1} , pH 10 MXene dispersions prepared with different alkalis **a** before and **b** after centrifugation at $400 \times g$ for 30 s. SEM images of the centrifugation-assisted MXene gels prepared with **c** KOH, **d** NH_4OH and **e** $(\text{C}_4\text{H}_9)_4\text{NOH}$. $T = 25$ $^\circ\text{C}$

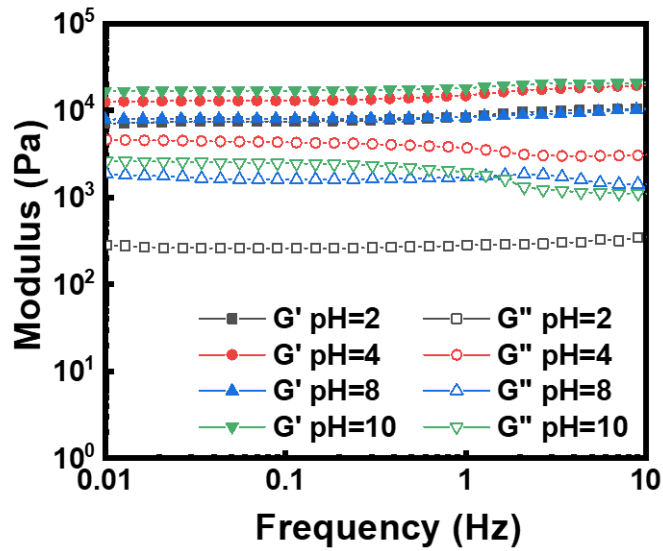


Fig. S11 Rheological property of the centrifugation-assisted MXene gel with different internal pH values. $T = 25\text{ }^\circ\text{C}$

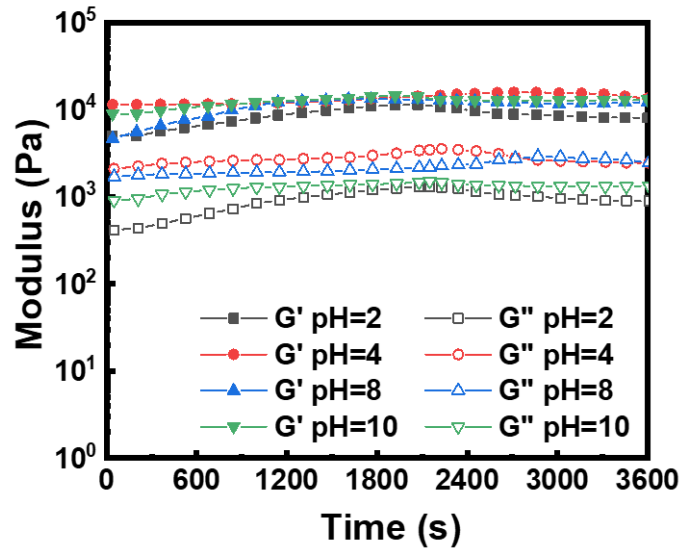


Fig. S12 Changes in viscoelasticity of the MXene gels over time at a constant shear frequency and strain of 1 Hz and 5%, respectively. $T = 25\text{ }^\circ\text{C}$

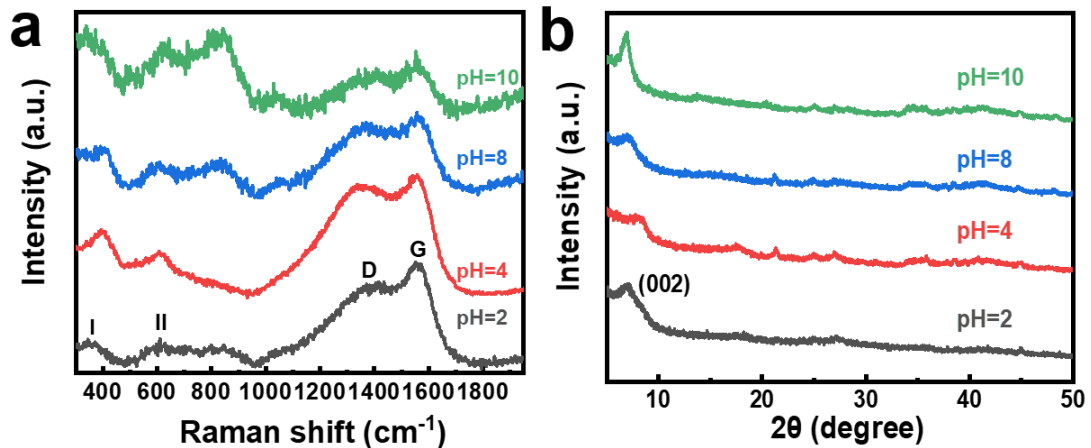


Fig. S13 **a** Raman and **b** XRD profiles of lyophilized MXene gels at different pH values. $T = 25\text{ }^\circ\text{C}$

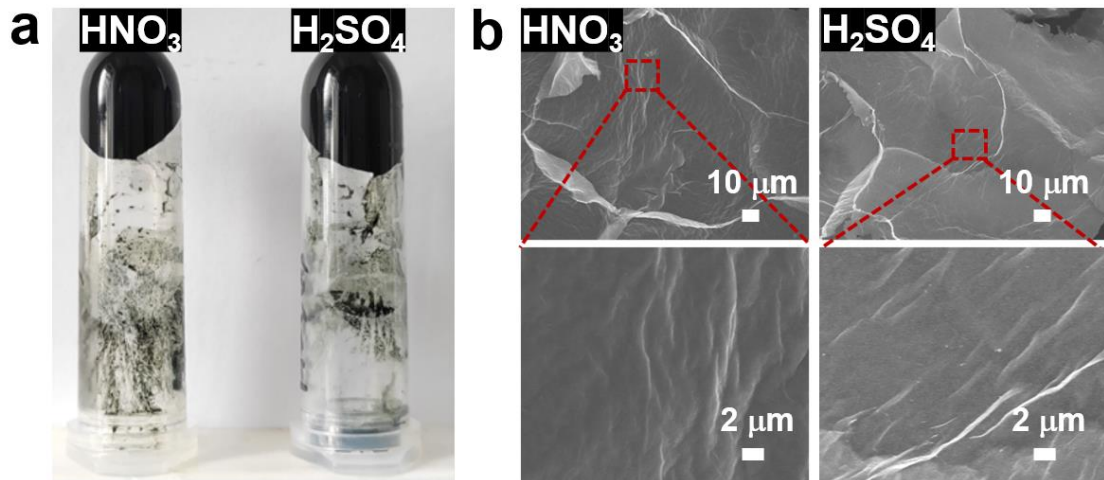


Fig. S14 **a** Photographs and **b** internal microstructures of pH 2 MXene gels prepared using H_2SO_4 and HNO_3 . $T = 25\text{ }^\circ\text{C}$

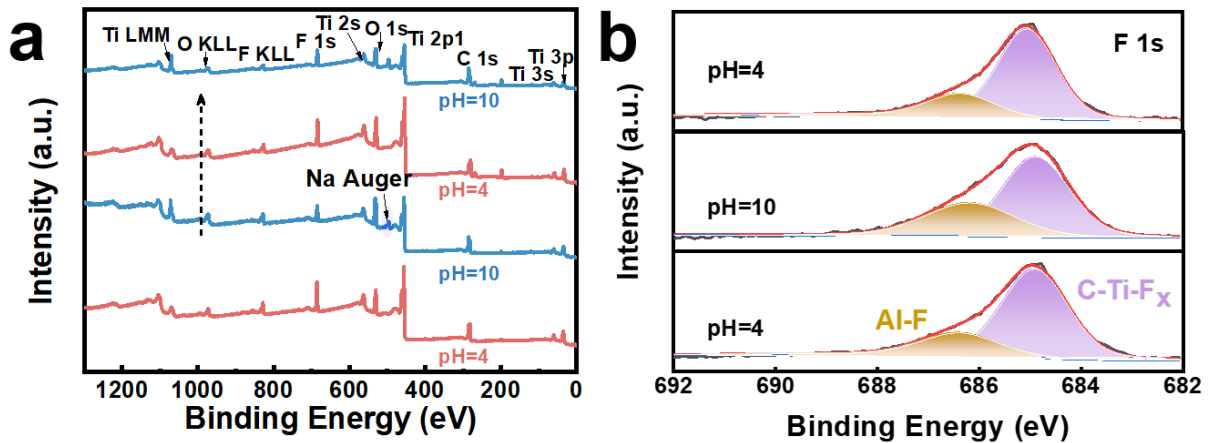


Fig. S15 **a** Full XPS and **b** XPS F 1s spectra of a lyophilized MXene gel after alternately changing pH between 4 and 10. $T = 25\text{ }^\circ\text{C}$

Table S1 Contents of each terminal group determined by the deconvolution and integration of corresponding band areas from the XPS profiles

| pH | -F | -O | -OH |
|-------|-------|-------|-------|
| pH=4 | 50.4% | 23.9% | 25.7% |
| pH=10 | 30.3% | 48.0% | 21.7% |
| pH=4 | 48.2% | 24.0% | 27.8% |

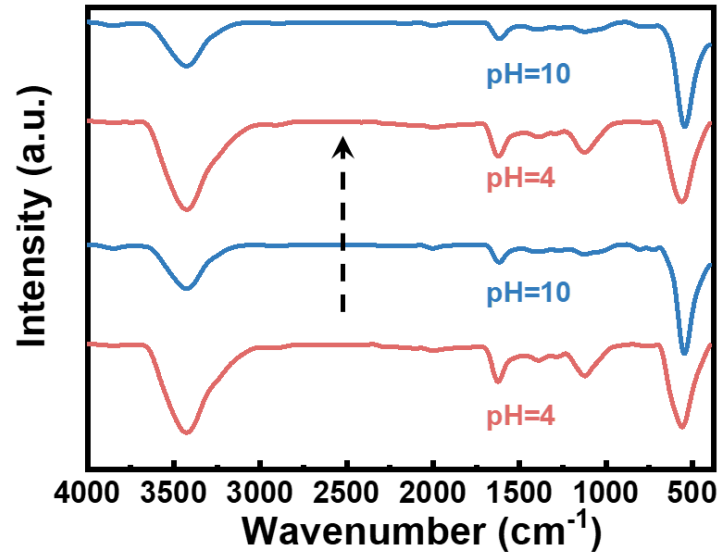


Fig. S16 FTIR profile of a freeze-dried MXene gel after alternately changing its internal pH value between 4 and 10. $T = 25\text{ }^{\circ}\text{C}$

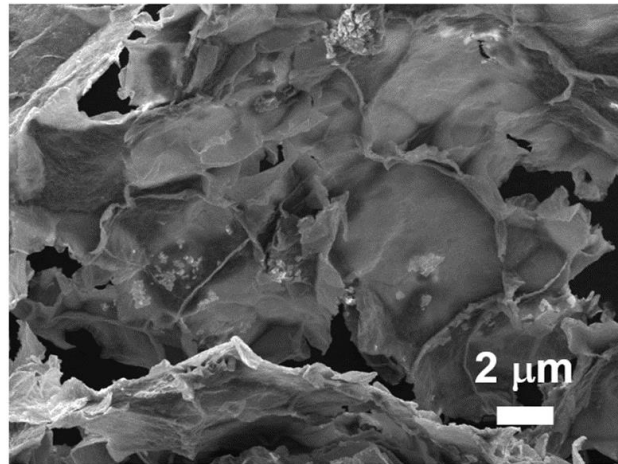


Fig. S17 Internal microstructure of a pH 4 gel after addition of an equal amount of NaCl instead of NaOH

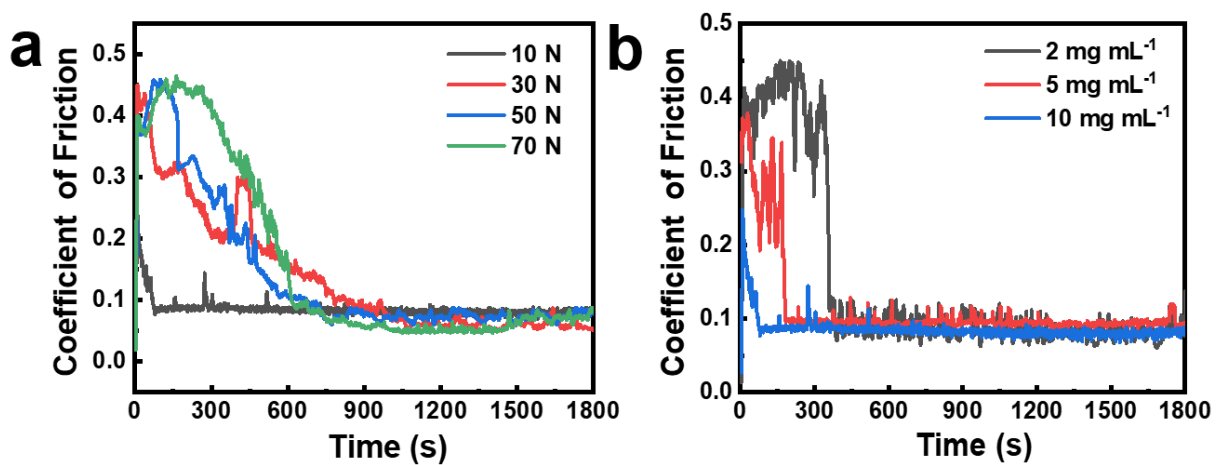


Fig. S18 Effect of **a** applied normal load and **b** initial particle concentration on the CoF of a pH 8 MXene gel. Sliding velocity = 10 mm s^{-1} . $T = 25\text{ }^{\circ}\text{C}$

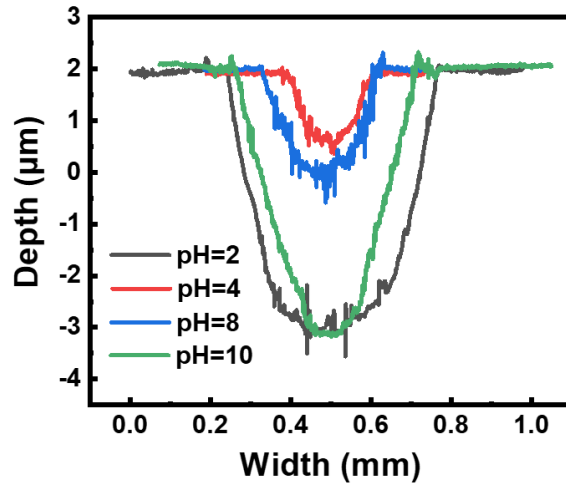


Fig. S19 Width and depth profiles of the wear scar on a steel substrate lubricated with MXene gels in different pH values

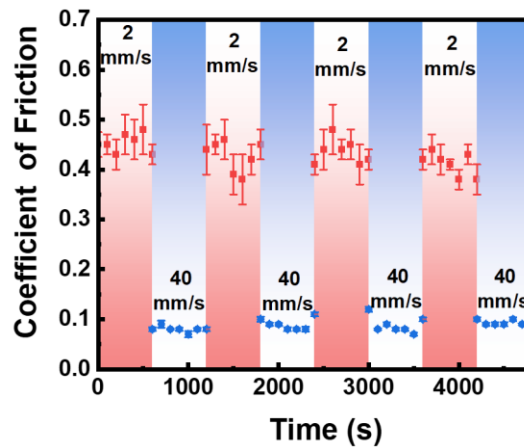


Fig. S20 Variations in the CoF of a pH 4 MXene gel upon alternately changing sliding velocity between 2 and 40 mm s⁻¹. T = 25 °C

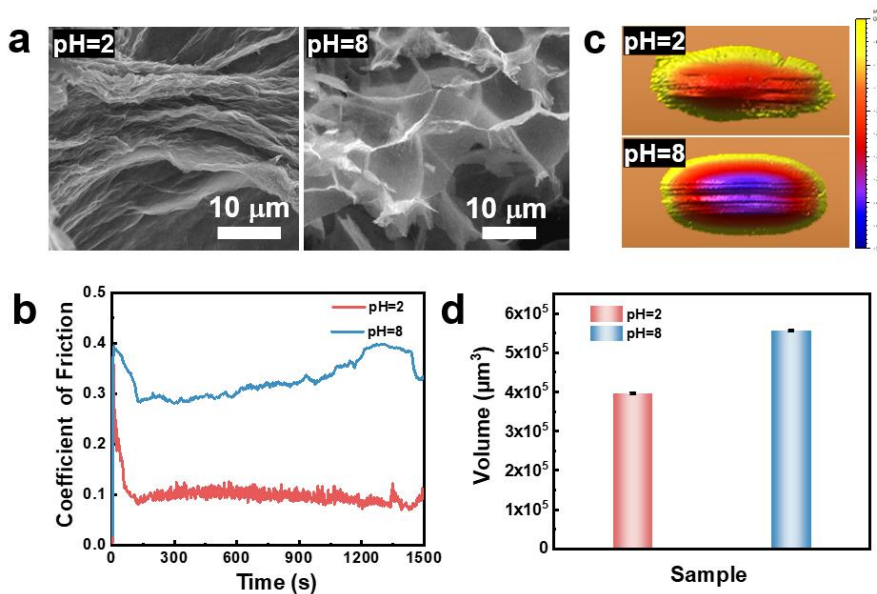


Fig. S21 **a** Internal self-assembled structure and **b** CoF of the MXene-PDDA composite gel at pH 2 and 8. **c** 3D surface topography and **d** abrasive volume of a steel substrate lubricated with the PDDA-containing gel at pH 2 and 8 under 10 N and 10 mm s⁻¹. T = 25 °C

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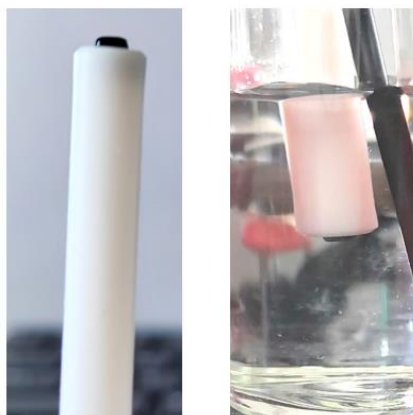


Fig. S22 Photographs of an electrode fabricated with the $\text{Ti}_3\text{C}_2\text{T}_x$ gel

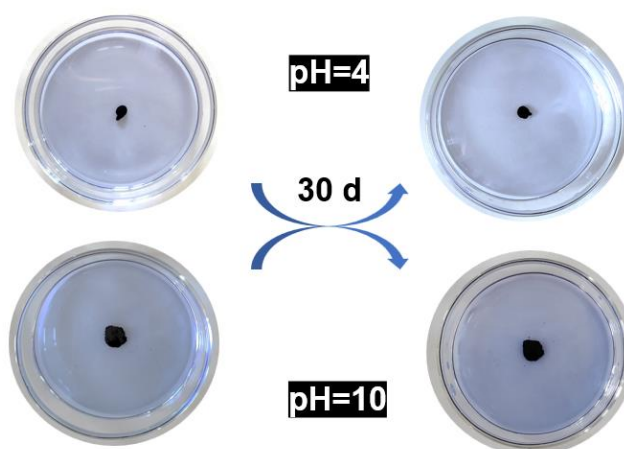


Fig. S23 Photographs of the pH 4 and 10 $\text{Ti}_3\text{C}_2\text{T}_x$ gels after immersed in water reservoirs stained with dye methylene blue for better visualization for 30 days. $T = 25\text{ }^\circ\text{C}$

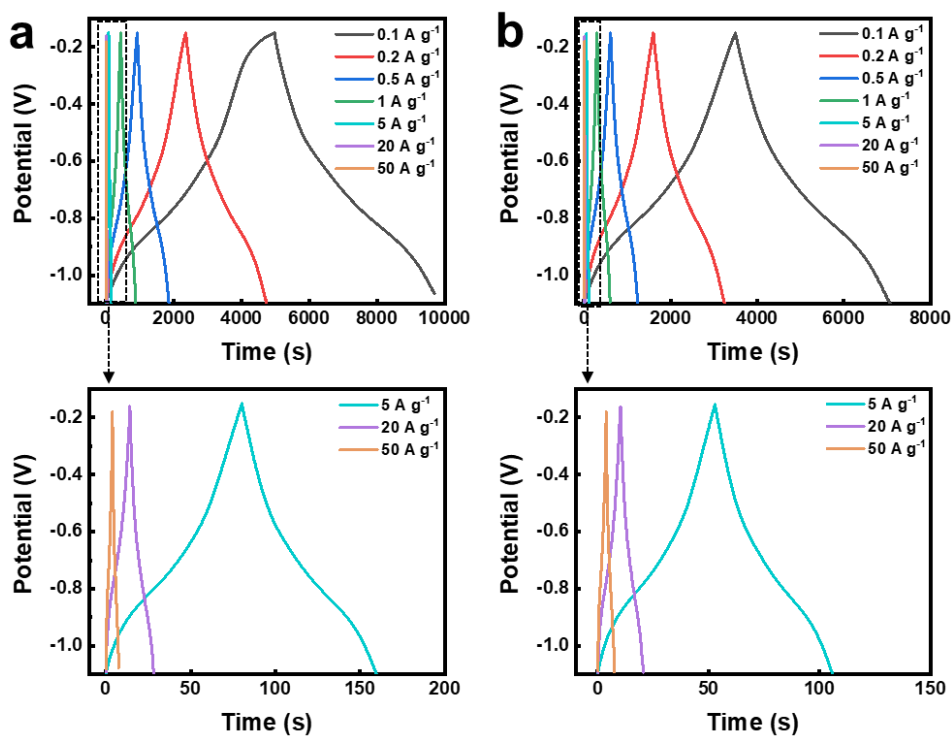


Fig. S24 GCD curves of **a** pH 4 and **b** 10 MXene gels at current densities between 0.1 and 50 A g^{-1} . $T = 25\text{ }^\circ\text{C}$

Table S2 Electrochemical performances of the MXene gel-based electrodes reported previously

| Gels | Electrolyte | Potential (V) | Specific capacitance (F g ⁻¹) | Cyclic stability | Refs. |
|--|------------------------------------|---|---|---|-----------|
| MXene/Fe ²⁺ | 3 M H ₂ SO ₄ | -1.1 to -0.15 (Hg/Hg ₂ SO ₄) | ~270 (10 mV s ⁻¹) ~255 (100 mV s ⁻¹) | 97.1% after 10,000 cycles | [S1] |
| MXene/HA | 3 M H ₂ SO ₄ | -1.1 to -0.15 (Hg/Hg ₂ SO ₄) | ~255 (10 mV s ⁻¹) ~90 (100 mV s ⁻¹) | 91.7% after 10,000 cycles | [S2] |
| MXene/H ₂ SO ₄ hydrogel film | 3 M H ₂ SO ₄ | -1.1 to -0.1 (Hg/Hg ₂ SO ₄) | ~375 (10 mV s ⁻¹) | 90% after 10,000 cycles | [S3] |
| H ₂ SO ₄ -thawed MXene | 3 M H ₂ SO ₄ | -1.2 to -0.2 (Hg/Hg ₂ SO ₄) | ~393 (5 mV s ⁻¹) | 95.5% after 10,000 cycles | [S4] |
| MXene/rGO hydrogel film | 3 M H ₂ SO ₄ | -1.1 to -0.15 (Hg/Hg ₂ SO ₄) | ~300 (10 mV s ⁻¹) ~280 (100 mV s ⁻¹) | 94.3% after 10,000 cycles | [S5] |
| MXene/GO | 3 M H ₂ SO ₄ | -0.5 to 0.3 (Ag/AgCl) | ~470 (10 mV s ⁻¹) ~380 (100 mV s ⁻¹) | ~98% after 8,000 cycles | [S6] |
| MXene/Al ³⁺ | 1 M H ₂ SO ₄ | -0.4 to 0.3 (Ag/AgCl) | ~275 (100 mV s ⁻¹) | ~90% after 5,000 cycles | [S7] |
| MXene/rGO/CNT | 3 M H ₂ SO ₄ | -0.6 to 0.25 (Ag/AgCl) | ~300 (100 mV s ⁻¹) | 97.1% after 10000 cycles | [S8] |
| Zn ²⁺ /MXene hydrogel film | 1 M H ₂ SO ₄ | -0.6 to 0.2 (Ag/AgCl) | ~390 (10 mV s ⁻¹) ~350 (100 mV s ⁻¹) | ~98% after 10,000 cycles | [S9] |
| MXene | 3 M H ₂ SO ₄ | -1.1 to -0.15 (Hg/Hg ₂ SO ₄) | pH 4: ~635 (5 mV s ⁻¹) ~604 (10 mV s ⁻¹) ~408 (100 mV s ⁻¹) pH 10: ~344 (5 mV s ⁻¹) ~322 (10 mV s ⁻¹) ~305 (100 mV s ⁻¹) | pH 4: 85.9% after 10,000 cycles pH 10: 96.7% after 10,000 cycles | This work |

Table S3 Electrical conductivity of previously reported MXene-based aerogels

| Aerogels | Electrical conductivity (S m ⁻¹) | Refs. |
|-----------------------|--|-----------|
| MXene/CNF | 1.8 | [S10] |
| MXene/rGO | 695.9 | [S11] |
| MXene/rGO/CNT | 9-92 | [S8] |
| MXene/CNF/CNT | 2400 | [S12] |
| MXene/polyimide | 4 | [S13] |
| MXene/silver nanowire | 1532 | [S14] |
| MXene/acidified CNT | 447 | [S15] |
| MXene/rGO | 36.2 | [S16] |
| MXene | 20400 (pH 4) 3800 (pH 10) | This work |

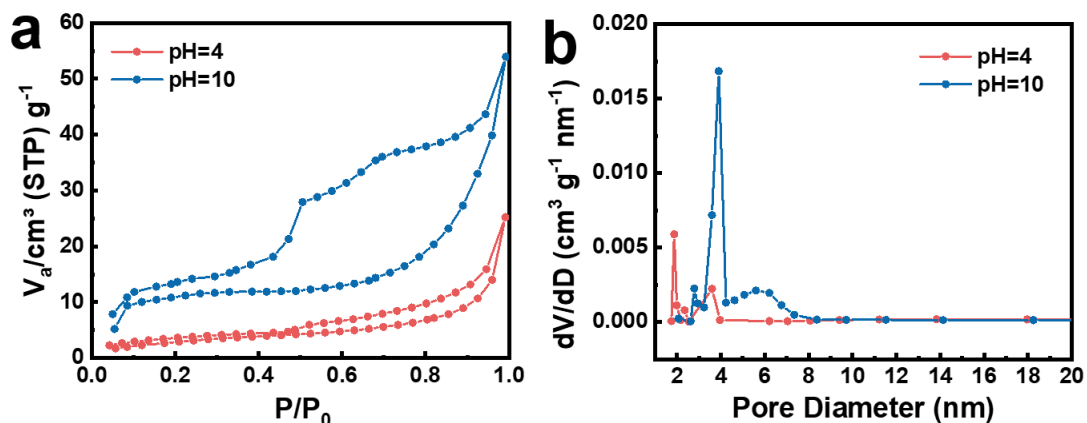


Fig. S25 **a** N_2 adsorption/desorption isotherms and **b** pore size distribution of lyophilized $Ti_3C_2T_x$ gels at $25\text{ }^\circ\text{C}$

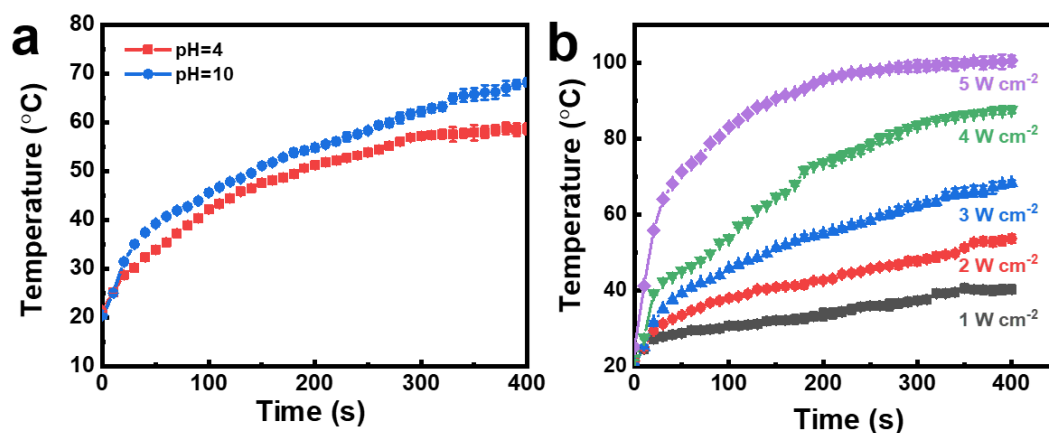


Fig. S26 **a** Temperature variations of the $Ti_3C_2T_x$ gels after irradiated by a 3 W cm^{-2} NIR light for 400 s. **b** Effect of irradiation power on the temperature increment of a pH 10 $Ti_3C_2T_x$ gel. It can be seen that a power above 3 W cm^{-2} would cause a relatively abrupt temperature elevation that may lead to severe water evaporation and changes in the gel properties



Fig. S27 Photograph of the jellyfish-shaped MXene gel coating on a PET substrate in the bending state

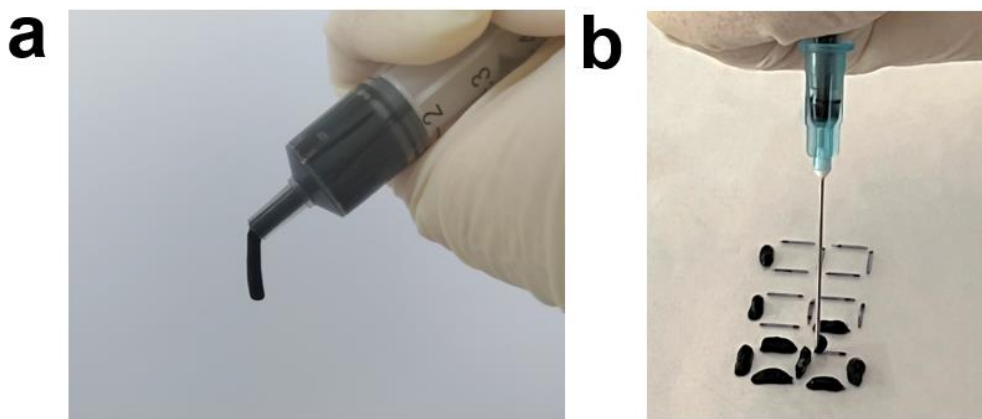


Fig. S28 Photographs of **a** extrusion of the gel through a needle syringe and **b** extrusion-printing of the MXene gel into anti-counterfeiting passwords

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