Supplementary Information for

2D Material-Integrated Hydrogels as Multifunctional Protective Skins for Soft Robots

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Supplementary References

Experimental Section

Materials.

Lithium fluoride (LiF, Sigma-Aldrich, BioUltra, ≥99.0%), hydrochloric acid (HCl, Sigma-Aldrich, ACS reagent, 37%), Ti₃AlC₂ MAX powders (Tongrun Info Technology Co. Ltd, China), sodium montmorillonite (MMT, BYK Additives Incorporation; Cloisite Na⁺), graphene oxide (GO) aqueous dispersion (Angstron Materials Incorporation, 5 mg mL⁻¹), and dichloromethane (DCM, J.T. Baker, 99.9%) were used as received without further purification. Magnesium chloride hexahydrate $(MgCl_2 \cdot 6H_2O),$ dimethyl sulfoxide (DMSO), tetrahydrofuran (THF), chloroform, hexane, gelatin (from cold water fish skin), glutaraldehyde solution (25% in H₂O), dopamine hydrochloride, and tris(hydroxymethyl)aminomethane were purchased from Sigma-Aldrich and used as received. The silicone elastomers (EcoflexTM-0020, EcoflexTM-0030, EcoflexTM-0035) were purchased from Smooth-On. Polydimethylsiloxane (PDMS, SYLGARD[™] 184 Silicone Elastomer Kit) was purchased from Dow Inc. Latex actuators (Aihua Balloons, Hebei, P. R. China) were used after cleaning with ethanol. Deionized (DI) water (18.2 M Ω) was obtained from a Milli-Q water purification system (Millipore Corp., Bedford, MA, USA) and used as the water source throughout the work.

Preparation of Ti₃C₂T_x MXene nanosheets.

 $Ti_3C_2T_x$ MXene nanosheets were prepared according to our previous work.¹ 1.0 g of LiF was added to 6.0 M HCl solution (20 mL) under vigorous stirring. After the dissolution of LiF, 1.0 g of Ti_3AlC_2 MAX powder was slowly added into the HF-containing solution. The mixture was kept at 35 °C for 24 hours. Afterwards, the solid residue was washed with deionized (DI) water several times until the pH value increased to ca. 7.0. Subsequently, the washed residue was added into 100 mL of deionized water, ultrasonicated for 1 hour under N₂ atmosphere, and centrifuged at 3,000 r.p.m. for 30 minutes. The supernatant was collected as the final suspension of $Ti_3C_2T_x$ MXene nanosheets with the concentration of ca. 5 mg mL⁻¹.

Preparation of MMT nanosheets.

MMT nanosheet dispersion was synthesized by mixing as-received MMT powders in DI water (10 mg mL⁻¹), followed by the ultrasonication (Sonica 5200) for 2 hours and continuous stirring for 12 hours. The dispersion was then centrifuged at 4,000 rpm for 60 minutes, and the

supernatant was collected. The concentration of obtained MMT dispersion is about 7 mg mL⁻¹ (solid weight was measured after solvent evaporation at 90 °C), which was diluted to 5 mg mL⁻¹ for further usage.

Fabrication of soft pneumatic grippers.

The molds of soft pneumatic gripper were designed using a computer-aided design software (Alibre, Inc.) and created by a 3D printer with acrylonitrile butadiene styrene plastic. An air channel was designed on the mold, and the robot's body was formed by curing silicone elastomers in the 3D printed molds. Two silicone elastomers were used: a highly extensible, elastomeric one (EcoflexTM-0020) and a relatively inextensible, compliant one (PDMS). The mold (with the design of air channel) was first filled with EcoflexTM-0020, and the elastomer was allowed to cure for 5 hours at room temperature. Then, the cured robotic body was peeled off from the mold and sealed with a flat PDMS layer with the adhesion of EcoflexTM-0035.

Preparation of gelatin hydrogel and MXene-blended hydrogel (M-H) layers.

Generally, the precursor solution of hydrogel layer consisted of gelatin (12.0 wt.%), glutaraldehyde (0.40 wt.%), MgCl₂ (7.6 wt.%, i.e., 1.0 M), and DI water. Gelatin and MgCl₂ powders were first dissolved into DI water and stirred overnight. After the introduction of glutaraldehyde, the hydrogel precursor solution started to crosslink and finally became a freestanding gelatin hydrogel after 6 minutes.

On the other hand, the precursor solution of MXene-blended gelatin hydrogel (M-H) was composed of gelatin (12.0 wt.%), glutaraldehyde (0.40 wt.%), MgCl₂ (7.6 wt.%, i.e., 1.0 M), MXene nanosheets at various loadings (0.25, 0.50 and 0.75 wt.%), and DI water. Gelatin powders were first dissolved into the MXene dispersion and stirred overnight. Then, the solution of glutaraldehyde and MgCl₂ was added into the MXene-gelatin mixture, and the conductive M-H layer was obtained after *in situ* crosslinking for 7 minutes.

Characterization of temperature regulation performance.

We first prepared bare elastomer, gelatin hydrogel, GO/H, MMT/H, and MXene/H samples with identical dimensions. These samples were then put in an oven at 50 °C for 60 minutes, and their mass changes were monitored every 5 minutes. We further examined their

temperature regulation performance on a hotplate at 200 °C for 10 minutes, and their surface temperature was monitored every 30 seconds using a thermocouple. The temperature regulation performance at different stretching stages was examined by stretching the 2DM/H skins under certain uniaxial strains (50% and 100%) and then fixing them onto a 100 °C hotplate for 10 minutes. Their surface temperatures were monitored every 30 seconds using a thermocouple.

Fabrication of soft pneumatic grippers coated with 2DM/H and GO/M-H skins.

First, the soft pneumatic gripper was actuated to its full inflation volume and then immersed in the dilute solution of dopamine (5 mg mL⁻¹) and tris(hydroxymethyl)aminomethane (10 mM) at pH 8.5 for 24 hours. Afterwards, the surface of elastomeric body was grafted with polydopamine (PDA) (with thickness of ca. 50 nm). Then, the soft gripper was taken out and rinsed with DI water followed by coating with the precursor solution of hydrogel layer or M-H layer. The thickness of hydrogel (and M-H) layer was controlled to be ca. 100 μ m through brush coating, dip coating, or drop casting. The hydrogel-coated or (M-H-coated) soft gripper was then dipped into GO, MMT, or MXene dispersion, and the thickness of 2DM nanolayer was controlled by adjusting the concentration of 2DM dispersion and the coating time. For this work, we fixed the concentrations of GO, MMT, and MXene dispersions at 5.0 mg mL⁻¹, respectively, and the dip coating time was fixed at 1 minute to achieve the GO, MMT, and MXene nanolayers with thicknesses of ca. 4, 5, and 6 μ m, respectively. The above fabrication recipes were utilized throughout the soft robots demonstrated in this work.

Fabrication of a dual-bellows walker robot coated with MMT/H skin.

In brief, two bellows origami were first assembled using cellulose papers, and these cellulose origami were enclosed with glue and further stabilized with a thin layer of PDMS. Afterwards, plastic covers and pneumatic channels were connected to two bellows tubes to complete the fabrication of a dual-bellows walker robot. The connection points between pneumatic channels and walker robot were further sealed with PDMS to ensure no air leaks. Two friction feet were then 3D printed and installed at the head and tail of dual-bellows walker robot, allowing the robot to crawl as commanded.

The dual-bellows walker robot was then grafted with PDA to achieve sufficient surface hydrophilicity. Then, the hydrogel layer (with thickness of ca. $100 \ \mu m$) was deposited onto the

robot through brush coating. Afterwards, upon dipping the hydrogel coated robot into the dispersion of MMT nanosheets (5 mg mL⁻¹) for 1 minute, the MMT nanolayer (with thickness of 5 µm) was conformally coated onto the dual-bellows walker robot.

Fabrication of a soft batoid-like robot coated with GO/H skin.

Soft batoid-like robot was fabricated according to our previous work² with the fabrication details regarding electronic circuit design provided. To integrate GO/H robotic skin, the soft batoid-like robot was grafted with PDA to achieve sufficient surface hydrophilicity. Then, the hydrogel layer (with thickness of ca. 100 μ m) was deposited onto the robot through brush coating. Afterwards, upon dipping the hydrogel coated robot into the dispersion of GO nanosheets (3 mg mL⁻¹) for 1 minute, the GO nanolayer (with thickness of 3 μ m) was conformally coated onto the soft batoid-like robot.

Biofilm formation tests.

Both bacteria, *S. aureus* and *E. coli*, were cultured in LB medium overnight at 37 °C, and the bacterial solutions were then diluted at the ratio of 1:100. Afterwards, the samples of gelatin hydrogel and MXene/H skin (with diameter of 15.6 mm and height of 0.6 mm) were placed in a 24-well plate, and 1 mL of bacterial solution was added into each well followed by the incubation at 37 °C without shaking for 20 hours. Afterwards, the samples were rinsed with DI water for three times to remove unattached bacterial cells. Next, 1 mL of 0.1% crystal violet aqueous solution was added into each well followed by the incubation at room temperature for 15 minutes, after which the samples were rinsed with DI water for three times. 1 mL of 30% acetic acid was then added into each well to solubilize the crystal violet followed by the incubation at room temperature for 15 minutes. The optical density (OD) of crystal violet solution was then measured at 550 nm to quantify the degrees of biofilm formation. The statistical analyses were conducted in triplicate, and the data were reported as mean \pm standard deviation (SD).

Uniaxial tensile tests.

The samples of elastomer (EcoflexTM-0020), hydrogel-coated elastomer, 2DM/H-protected elastomer, freestanding gelatin hydrogel and M-H layers (0.5 wt.% Maxine loading) were

prepared with the identical dimensions of 5 (*L*) × 2 (*W*) × 0.25 (*T*) cm³. These samples were tested by a tensile tester (Instron 5543, Instron, USA) equipped with a 500 N load cell. The stress–strain properties were acquired at a strain rate of 25% s⁻¹ until fracture or till 500% strain, and all the tests were repeated at least three times.

Fire retardancy tests.

Fire retardancies of bare soft gripper, hydrogel-coated soft gripper, MMT/H-protected soft gripper, bare elastomer strips, hydrogel-coated elastomer strips, and MMT/H-protected elastomer strips were evaluated by the horizontal combustibility test method modified from the standard test method (ASMT D6413).³ Fire retardancy was evaluated by exposing the samples to the flame of an ethanol burner for 60 seconds (the flame temperature is reported to be 600–850 °C).⁴ The test processes and results were recorded via a digital camera.

Characterization.

Surface morphologies and cross-section information of 2DM/H skins were obtained by using a scanning electron microscope (SEM, FEI Quanta 600), a benchtop SEM (JCM-7000 NeoScopeTM Benchtop), and a field emission SEM (JEOL-JSM-6610LV) operated at 15.0 kV. As-exfoliated MXene, MMT, and as-received GO nanosheets were characterized by using a high-resolution transmission electron microscopy (HRTEM, JEOL 2010F). X-ray diffraction (XRD) patterns were recorded by an X-ray diffractometer (Bruker, D8 Advance X-ray Powder Diffractometer, Cu K α (λ = 0.154 nm) radiation) with a scan rate of 2° min⁻¹. Uniaxial tensile tests and delamination tests were conducted by using a Universal Testing System (Instron 5543, Instron, Canton, MA) at room temperature with a 500 N load cell. X-ray photoelectron spectroscopy (XPS) was measured by Kratos AXIS Ultra^{DLD} by using a microfocused (100 μm, 25 W) Al X-ray beam with a photoelectron take off angle of 90°. DCM concentrations within DCM/acetone mixtures were analyzed using gas chromatograph (Agilent 5977B GC/MSD system). *I–V* profiles and electrical resistances of MXene/H skins and M-L layers were measured using an electrochemical workstation, (AutoLab, Metrohm Singapore Pte. Ltd.). EMI shielding performance of stretchable MXene/H skin in the X-band (8.2–12.0 GHz) was characterized by using Agilent/HP 8510C Vector Network Analyzer. Optical density at 600 nm (OD₆₀₀) of bacterial cells and fluorescence intensity of dyes as well as OD₅₅₀ of crystal violet were measured by a microplate reader (Biotek H4FM).



Fig. S1. Hydrophilic surface modification of soft elastomeric bodies by polydopamine (PDA) grafting. (a) Schematic illustration of PDA grafting step on a bare soft gripper. (b) Fourier-transform infrared (FTIR) spectra of Ecoflex samples before and after PDA grafting. Characteristic peaks were observed in the PDA-grafted Ecoflex sample at 1,647 cm⁻¹ for O–H vibrations as well as at 1,530 cm⁻¹ for N–H bending. The broad peak around 3,300 cm⁻¹ indicates the hydrogen bondings between the PDA and Ecoflex surface. (c) Water contact angle measurement of Ecoflex samples before and after PDA grafting. Contact angle decreases from 110° to 79° after the PDA grafting step, demonstrating increased surface hydrophilicity.



Fig. S2. Crosslinking processes of gelatin hydrogel and MXene-blended hydrogel (M-H). (a) Photographs of gelatin precursor solution after introduction of crosslinking agent (i.e., glutaraldehyde) for 0, 4, 4.5, 5.5, and >6 minutes. Scalable deposition processes (e.g., dip coating, brush coating, and drop casting) were carried out within the period of 4.0-5.5 minutes. (b) Photographs of MXene-blended gelatin precursor solution after introduction of crosslinking agent for 0, 4, 5, 5.5, and >7 minutes. Scalable deposition processes were carried out within the period of 4.0-5.5 minutes.



Fig. S3. Scalable deposition techniques for conformal hydrogel layer. (a) Dip coating and (b) drop casting were both applicable to deposit viscous gelatin paste onto a PDA grafted soft gripper (under inflation).



Fig. S4. TEM images of 2DM nanosheets. Representative TEM images of (a) GO, (b) MXene, and (c) MMT nanosheets used in this work.



Fig. S5. Characterization of Mg^{2+} -intercalated 2DM nanolayers. (a) XPS spectra of GO/H and MXene/H skins through cation (Mg^{2+}) induced self-assembly. (b) For the cases of 1-minute dip coating in 5 mg mL⁻¹ of 2DM dispersions, 6.05 at.% and 5.01 at.% of Mg^{2+} were characterized in the self-assembled GO and MMT nanolayers. It is worth noting that the at.% of Mg^{2+} in MMT nanolayers was not able to be estimated, as MMT nanosheets contained octahedral cations (Al, Mg, Fe).



Fig. S6. Elemental characterization of 2DM/H robotic skins. SEM images, EDX mapping, and sum spectra of (a) gelatin, (b) GO/H, (c) MMT/H, and (d) MXene/H skins. Characteristic elements of C, S, and Ti were observed in GO/H, MMT/H, and MXene/H skins, respectively. Element of Mg was observed in all the samples, proving the successful Mg^{2+} intercalation into 2DM nanolayers. Scale bar: 20 µm.



Fig. S7. Mechanical delamination test results of MXene/hydrogel and MMT/hydrogel interfaces.



Fig. S8. Detailed fabrication processes of all 2DM/H soft grippers. Brush coating was used for deposition of MXene nanolayer onto a gelatin-coated gripper.



Fig. S9. Determination of characteristic wavelength of isotropic crumples under increasing areal strains. The characteristic wavelength was defined as the size of an individual crumple. With the areal strains increasing from 0% to 600% and 1,200%, the crumple sizes were extracted and varied from ~15 × 15, 56 × 56, to 77 × 77 μ m, respectively, demonstrating the unfolding process of isotropic crumples. Scale bar: 50 μ m.



Fig. S10. Fatigue tests on 2DM/H robotic skins. SEM images of GO/H, MXene/H, and MMT/H robotic skins before and after 500 cycles of pneumatic actuations. No topographical changes or emerging fractures/cracks were observed at their relaxed states, demonstrating the structural stability of all 2DM/H skins. Scale bar: 50 µm.



Fig. S11. Control experiments for installation of 2DM/H robotic skins. Control experiments showed that it is necessary to conduct two-step interfacial engineering process to achieve conformal 2DM/H skin on a bare soft gripper. (a) Without PDA grafting and intermediate hydrogel layer, 2DM nanolayer was not able to be deposited onto a bare gripper. (b) Without PDA grafting, conformal gelatin layer was not able to be coated onto a bare gripper. (c) Without intermediate hydrogel layer, 2DM nanolayer was not able to be deposited onto a bare gripper. (c) Without gripper. (d) Without MgCl₂, 2DM nanolayer was not able to be deposited onto a hydrogel-coated gripper.



Fig. S12. (a–d) Photos of three hydrogel-elastomer bilayers with the hydrogel thicknesses of 50, 100, and 200 μ m upon bending. Detachment of 200- μ m-thick hydrogel layer from the elastomer substrate was observed. (e) Heat regulation performance of three hydrogel-elastomer bilayers with the hydrogel thicknesses of 50, 100, and 200 μ m on a hotplate at 100 °C.



Fig. S13. XRD analyses of as-filtered GO, MMT, and MXene membranes.



Fig. S14. XRD analyses of GO, MMT, and MXene membranes transferred onto hydrogels.



Fig. S15. Coordinated grasping missions in water. (a) Bare and (b) hydrogel-coated soft grippers executed coordinated grasping missions in water to collect a blue cap.



Fig. S16. Grasping behaviors of various soft grippers upon DCM and THF exposure. Bare gripper was not able to conduct coordinated grasping behaviors in DCM or THF bath due to significant swelling of robotic fingers. The hydrogel coating could not prevent this deteriorated

In DCM bath

grasping behavior in the DCM bath. In contrast, GO/H gripper was able to repeatedly collect a blue cap in a DCM bath over 10 times.



Fig. S17. Solvent contact assay for stretchable GO/H robotic skin. Barrier performance of GO/H skin covering five chemical compounds (dichloromethane (DCM), chloroform, hexane, tetrahydrofuran (THF), and dimethyl sulfoxide (DMSO)) at two stretching states (600% and 1,200%). The pneumatic actuators without protection of GO/H skins burst within 8 seconds after contact with organic liquid baths. The pneumatic actuators with protection of GO/H skins survived under direct liquid contact over 100 seconds (duration of the test) under extreme strains.



Fig. S18. High-resolution characterization of barrier performance. Gas chromatography (GC) results of continuous DCM permeation across various barriers into an acetone reservoir. (a) Bare elastomer. (b) Hydrogel-coated elastomer. (c, d) GO/H-protected elastomer under areal strains of 600% and 1,200%, respectively.



Before grasping missions



After grasping missions

Fig. S19. Zoomed-in photos of the GO/H gripper before and after working in DCM for long time. The GO/H skin remained intact after multiple times of DCM exposure. Scale bar, 1 cm.



Fig. S20. Heat regulation performances of robotic skins protected elastomers upon direct exposures to flame. (a) Optical photos and (b) schematic illustration of the setup for surface temperature measurement. (c) Temperature profiles of the elastomer surface upon exposures to flame under the protection of different robotic skins.



Fig. S21. Effects of hydrogel hydration degree on the (a) heat regulation and (b) fire retardant performances of MMT/H layer.



Fig. S22. *I*–*V* curves of MXene/H skins. With the MXene thicknesses of 4, 6, and 13 μ m, the electrical conductivities of MXene/H skins were extracted to be 1.20×10^3 , 3.97×10^3 , and 1.03×10^4 S m⁻¹, respectively.



Fig. S23. Biofilm assay quantifications. Relative biofilm formation of *S. aureus* and *E. coli* on gelatin hydrogel and MXene/H skin.



Fig. S24. Effect of hydration degree on the sensing performance of M-H layer. (a) The water content of an M-H layer (0.50 wt.% MXene) decreased with time at room temperature at the relative humidity of 58.0%. (b) Gauge factors (GF) of M-H layers with different water contents.



Fig. S25. Fabrication process of a GO/M-H gripper with high-level function integration.



Fig. S26. Configuration of GO/M-H robotic skin. (a) Top-down SEM image and (b) cross-section EDX mapping (C and Ti) of GO/M-H skin.



Fig. S27. Tensile tests of elastomeric body and hydrogel layer. Young's modulus of freestanding gelatin hydrogel was extracted (to be 0.0075 MPa) as the representative of other 2DM/H (and 2DM/M-H) skins.



Fig. S28. Actuation processes of GO/M-H protected and bare grippers under a 12-second actuation cycle, where the body deformations were monitored and quantified using the arc dimensions of a specific area.



Fig. S29. Assembly of a dual-bellows walker robot.



Fig. S30. GO/H-protected batoid-like robot under continuous rinsing of DCM liquid.



Fig. S31. Swimming traces of bare and GO/H-protected batoid-like robots before and after liquid DCM rinsing for 10 minutes (in Y-Z plane).

Note S1. Reaction details of gelatin crosslinking.

After a small amount of glutaraldehyde (0.40 wt.%) was added into the precursor solution of gelatin hydrogel at a near-neutral pH value, the highly reactive aldehyde groups of glutaraldehyde instantaneously reacted with the free amine groups of gelatin. The crosslinking reaction that first took place was the formation of imine derivatives (Schiff base bonds) between gelatin and glutaraldehyde, as shown in **Fig. S32**. As more and more amino acids on the polypeptide chains of gelatin started to crosslink, the viscosity of gelatin paste started to increase. The viscous paste was then deposited through various scalable coating techniques, including brush coating, dip coating, and drop casting.



Fig. S32. Reaction details of gelatin crosslinking.

Note S2. Delamination test details.

The delamination tests were conducted to determine the adhesion energies of elastomer/hydrogel interfaces before and after PDA treatment as well as to compare the adhesion energies of GO/hydrogel interfaces prepared by cation-induced self-assembly and direct GO thin film transfer. Mechanical delamination tests were conducted by using a Universal Testing System (Instron 5543, Instron, Canton, MA). As shown in Fig. S33a, to measure the adhesion energy of GO/hydrogel interface, a certain area of the GO nanolayer was glued to another substrate (the square area circled with a white dashed line). During the tension of GO/gelatin device, this certain area of GO was peeled off from the hydrogel, and the delamination force was recorded. The adhesion stress was calculated by the loading force divided by the delaminated area, and the binding energy between GO and hydrogel was obtained by integrating the area of the corresponding curves in Fig. 1e (left). To measure the adhesion energy of elastomer/hydrogel interface, one side of the elastomer square was first glued to the glass slide (the square area circled with a green dashed line), as shown in Fig. S33b. The other side was then drop coated with gelatin precursor and was subsequently attached to another glass slide during the crosslinking process. During the tension of the elastomer/gelatin device, this certain area of gelatin was peeled off from the elastomer, and the delamination force was recorded. The adhesion stress was calculated by the loading force divided by the delaminated area, and the binding energy between elastomer and hydrogel was obtained by integrating the area of the corresponding curves in Fig. 1e (right). The adhesion stress dropped abruptly to 0 at a certain displacement indicates that the certain area of GO nanolayer was completely detached from the hydrogel surface. Similar tests were also conducted for the MXene/hydrogel and MMT/hydrogel interfaces to measure their adhesion energies. All the measurements were repeated at least three times to extract the average values.



GO/Hydrogel interface

а

b Elastomer/Hydrogel interface

Fig. S33. Mechanical delamination tests. Schematic illustrations and actual setups of the mechanical delamination tests for (a) GO/hydrogel and (b) elastomer/hydrogel interfaces.

Note S3. Organic solvent diffusion tests.

Fig. S34 shows a diffusion cell used to quantify the DCM permeation rate across a barrier sample. To characterize the DCM permeation across the GO/H skin, the side openings of Chambers A and B were both sealed with the GO/H skins under 1,200% areal strain. Afterwards, Chambers A and B sealed with stretched GO/H skins were face-to-face aligned and clamped together tightly. During DCM permeation tests, Chamber A was filled with DCM, and Chamber B was filled with acetone, and the top openings were sealed with parafilm. 0.5 mL of liquid was taken from Chamber B after 0.5, 1.0, 2.0, 3.0, and 12.0 hours, and the concentration of permeated DCM was characterized by gas chromatography (GC). A DCM-acetone mixture with 25 vol.% of DCM was measured as the reference.



Fig. S34. Diffusion cell setup for characterization of DCM permeation across GO/H skins. (a) Side opening of Chamber A covering with one GO/H skin under 1,200% areal strain. (b) Side opening of Chamber B covering with one GO/H skin under 1,200% areal strain. (c) Diffusion cell setup with face-to-face connection of Chambers A and B. During DCM permeation tests,

Chamber A was filled with DCM, and Chamber B was filled with acetone. 0.5 mL of liquid was taken from Chamber B after 0.5, 1.0, 2.0, 3.0, and 12.0 hours, and the concentration of permeated DCM was characterized by GC.

Note S4. Characterization of EMI SE.

As the incident EM radiation encounters a shielding barrier, the EM waves are divided into three parts, including reflection (R), absorption (A), and transmission (T), as illustrated in **Equation S1**,

$$A + R + T = 1 \tag{1}$$

The EMI SE is measured in decibels (dB) and defined as the logarithmic ratio of the incident power (P_I) over transmitted power (P_T), as shown in **Equation S2**,

$$EMISE(dB) = 10\log\frac{P_I}{P_T}$$
(2)

The total EMI SE (SE_T) is the sum of contributions from reflection (SE_R), absorption (SE_A), and multiple reflections (SE_{MR}). For a multilayered EMI shielding material with high EMI SE, the contribution of SE_{MR} is normally merged into the absorption part (SE_A), since the rereflected waves are absorbed or dissipated as heat in the shielding materials. Therefore, the SE_T can be simplified as **Equation S3**,

$$SE_T = SE_A + SE_R \tag{3}$$

By using the vector network analyzer (VNA), four parameters ($|S_{11}|$, $|S_{12}|$, $|S_{21}|$, $|S_{22}|$) are obtained in the EMI shielding tests and can be used to calculate reflection (*R*) and transmission (*T*) in **Equation S4** and **S5**,

$$R = |S_{11}|^2 = |S_{22}|^2 \tag{4}$$

$$T = |S_{12}|^2 = |S_{21}|^2 \tag{5}$$

 SE_R and SE_A can be derived in Equation S6 and S7,

$$SE_{R} = 10 \log \left(\frac{1}{1 - |S_{11}|^{2}} \right)$$
(6)
$$SE_{A} = 10 \log \left(\frac{1 - |S_{11}|^{2}}{|S_{21}|^{2}} \right)$$
(7)

EMI shielding efficiency is also used to express the EM shielding abilities of a material and can be obtained by using **Equation S8**,

$$EMI \ shielding \ efficiency \ (\%) = \frac{1}{10^{\frac{I \ SE_T \ I}{10}}} \times 100\%$$
(8)



Fig. S35. Setup of EMI shielding measurement. (a) Contribution of EM absorption (SE_A) and EM reflection (SE_R) to total EMI SE (SE_T) of MXene/H skins with different thicknesses of MXene nanolayers at the frequency of 8.9 GHz. (b) Vector network analyzer. (c) MXene/H skin was stabilized by double-sided tapes on the Cu calibration kit.

Supplementary References

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