

Supplementary information for

In-Plane Molecular Cord Bridging as a Method for Boosting

MXene Fiber Strength and Toughness

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Supplementary Note 1: Measurements

Orientation measurements

The orientation characteristics of MXene-based fibers were determined through the small- and wide-angle X-ray scattering (SAXS and WAXS) measurements using a Xeuss 3.0 system (Xenocs, France). Measurements were performed at 30 W tube power ($\lambda = 1.54$ Å) using an Eiger2R 1M detector positioned at ~60 mm sample-to-detector distance. From the 2D scattering patterns, Herman's orientation factor (f) was calculated to reflect the degree of orientation change of the fibers before and after hot-pressing using the following Equation (S1):

$$f = \frac{3\langle \cos^2 \varphi \rangle - 1}{2} \quad (\text{S1})$$

where the mean squared cosine value was calculated from the scattered intensity $I(\varphi)$ by integrating over the azimuthal angle φ according to the following Equation (S2):

$$\langle \cos^2 \varphi \rangle = \frac{\int_0^{\pi/2} I(\varphi) \sin(\varphi) \cos^2 \varphi d\varphi}{\int_0^{\pi/2} I(\varphi) \sin(\varphi) d\varphi} \quad (\text{S2})$$

where the azimuthal angle (φ) is the angle between the fiber plane and the MXene nanosheet plane. Therefore, $f = 1$ when all MXene nanosheets were parallel to the fiber plane and $f = 0$ for randomly oriented MXene nanosheets.

Rheological property measurements

The rheological properties of the MXene-based spinning dopes were measured using a DHR-2 rheometer (TA Instruments, USA) at room temperature (25.0 ± 0.3 °C). Using the following Equation (S3) to estimate the shear rate (γ , s⁻¹) during the spinning process:

$$\gamma = \frac{4Q}{\pi R^3} \quad (\text{S3})$$

where Q is the extrusion rate (5 mL·h⁻¹) and R is the inner radius of the needle. The shear rate for the 26G (220 μm) needle was calculated to be 166.2 s⁻¹.

Conductivity measurements

The electrical conductivity characterization was performed using a two-point probe configuration (Keithley 2400 SMU) with silver paste contacts to minimize contact resistance. The conductivity (σ) of the fabricated fibers was determined through Equation (S4):

$$\sigma = \frac{L}{SR} \quad (S4)$$

where L is the length of the as-prepared fibers, R is the electrical resistance, and S is the cross-section area of each fiber measured from SEM images.

Mechanical property measurements

Mechanical testing was carried out using a Shimadzu AGS-X 5N universal tester (10 N load cell) at 10 mm min⁻¹ strain rate with 10 mm gauge length. More than four samples were tested for each fiber, and the average and standard deviation were calculated as error bars. The effective cross-sectional area of the fibers was determined using ImageJ software.

Porosity calculations

The porosity (p) of the fiber was calculated from the structural parameters via Equation (S5):

$$p(\%) = \left(1 - \frac{\rho_{MF}d_{002}}{\rho_M d_M}\right) \times 100 \quad (S5)$$

where ρ_{MF} is the density of the fiber, ρ_M is the Ti₃C₂ density (4.4 g cm⁻³), d_{002} is the d -spacing (002) of the MXene based fibers, and d_M is the d -spacing of Ti₃C₂ crystals (1.37 nm). The calculation of density relies on the linear density method. Specifically, fibers of known length were weighed using a high-precision microbalance (precision: 0.01 mg), and the linear density was calculated. The density was then derived by combining the linear density with the geometric cross-sectional area obtained from SEM images.

Electrochemical performance measurement

The electrochemical properties of the MXene-based fiber electrodes were measured in a three-electrode system using a 1.0 M H₂SO₄ electrolyte, an Ag/AgCl reference electrode, and Pt counter electrode. The cyclic voltammetry (CV) and galvanostatic charge/discharge (GCD) of the MXene-based fiber electrodes were performed using a CHI660 electrochemical workstation (Chenhua Co., China). The voltage window for CV and GCD tests was set from -0.2 to 0.4 V.

The specific volumetric capacitance (C_{sp}) of the fiber electrode in a three-electrode system was calculated from the CV curves according to the Equation (S6) and (S7):

$$C_{\text{electrode}} = \frac{Q}{2V} = \frac{1}{2V\nu} \int_{V-}^{V+} i(V) dV \quad (\text{S6})$$

$$C_{sp} = C_{\text{electrode}}/V_{\text{fiber}} \quad (\text{S7})$$

where i is the current (A), V is the potential window, ν is the potential scan rate (V s⁻¹), and V_{fiber} was the volume of a single fiber electrode.

Supplementary Note 2: DFT methods

Density functional theory (DFT) calculations were conducted based on the Vienna Ab initio Simulation Package (VASP). The exchange-correlation potential was described by the Perdew-Burke-Ernzerhof (PBE) generalized gradient approach (GGA)². The electron-ion interactions were accounted by the projector augmented wave (PAW). All DFT calculations were performed with a cut-off energy of 400 eV, and the 2×3×1 Gamma centered Monkhorst-Pack grids k-points were selected to sample the Brillouin zone integration. The energy and force convergence criteria of the self-consistent iteration were set to 10⁻⁴ eV and 0.05 eV Å⁻¹, respectively. DFT-D3 method was used to describe van der Waals interactions³.

Supplementary Note 3: MD Simulation models and methods

In this work, classic molecular dynamics (MD) simulations are performed to explore the mechanical properties of MXene-based nanomaterials subjected to straining from the nanoscale level. Here, three types of MXene-based fibers are taken into investigations for comparison. The first MXene-based fiber is composed of eight MXene nanosheets that are stacked into four-layers (neat MXene fiber), while the second sample is structurally characterized by that 16 polymer chains are uniformly inserted into two interlayered spaces of four-layered MXene nanosheets (C-MXene fiber), in which the polymerization $n = 10$. For the third one, one carbon nanotube is uniquely placed into a confined space between MXene sheets for the second sample where polymer chains are surrounding (CS-MXene fiber). Moreover, 40 Mg^{2+} ions are randomly inserted into the three systems. To describe the atomic interactions in the MXene-based system, the first-principles-based ReaxFF potential⁴ is employed because it consists of bond-order-dependent covalent interaction, nonbonded standard Coulomb and Morse interaction terms, the ReaxFF forcefield can mimic dissociation and formation of covalent bonds. Periodic boundary conditions (PBCs) are imposed in the three orthogonal directions of MXene sheets to mimic large samples. Prior to conducting MD simulations, as-generated MXene-based systems are quasi-statically relaxed to a local minimum-energy configuration via conjugate gradient method within tolerances of energy and force of $1.0 \times 10^{-4} \text{ Kcal mol}^{-1}$ and $1.0 \times 10^{-4} \text{ Kcal (mol} \cdot \text{\AA})^{-1}$, respectively. Subsequently, the as-minimized MXene-based systems are equilibrated for 200,000 timesteps under NPT ensemble (constant number of particles, constant pressure, and constant temperature) at pressure of 1 bar pressure. Finally, as-relaxed MXene-based samples are uniaxially-stretched in x-direction every 1,000 timesteps under NPT ensemble, under which the pressures in other two orthogonal (y and z) directions are solely controlled by Nose-hoover barostat technique. Such settings allow the consideration of Poisson effect during the stretching process. The uniaxial straining rate is assigned to be $1.0 \times 10^{-6} \text{ fs}^{-1}$. The

velocity-Verlet integration algorithm with a timestep of 0.25 fs is employed to integrate Newton's motions in our MD calculations. All MD calculations are implemented using the Large-scale Atomic-Molecular Massively Parallel Simulator (LAMMPS) software package⁵.

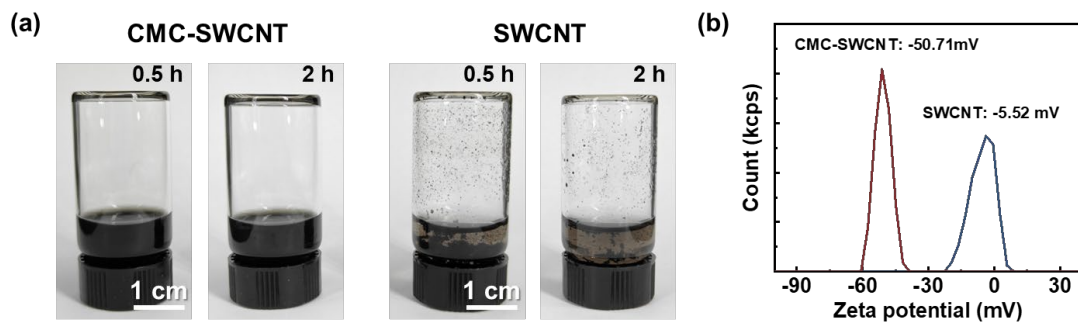
Supplementary Note 4: Discussion on orientation degree and compactness

After the sequential introduction of CMC and SWCNTs into MXene, the resulting fibers exhibit a gradually decreased porosity and an orientation degree that first decreases and then increases. This variation can be attributed to the following factors. The improvement in fiber compactness primarily stems from CMC acting as a flexible polymer binder that can fill the interlayer voids between adjacent MXene nanosheets during the wet-spinning and drying processes, thereby producing a denser structure with lower porosity⁶. Upon further introduction of SWCNTs, their one-dimensional nanostructure can insert into and bridge the microporous regions between adjacent layers, further compacting the internal packing of the fibers and thus causing a slight additional decrease in porosity⁷.

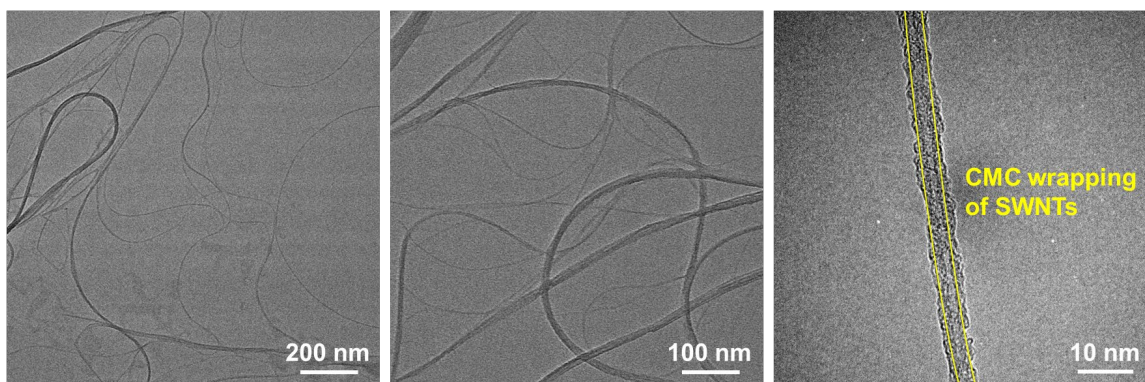
However, compactness and orientation are governed by different mechanisms. Pristine MXene nanosheets can form a highly ordered liquid-crystalline phase under shear and align efficiently along the fiber axis, whereas the introduction of CMC partially disrupts this ordered alignment because of the steric hindrance and chain entanglement of its random-coil polymer chains, leading to a reduced orientation factor. This interpretation is supported by the following evidence: a higher orientation factor was observed at lower CMC content (Supplementary Fig.10), and the C-MXene spinning dope exhibited weaker liquid-crystalline textures in the POM images (Supplementary Fig.4). In contrast, in CS-MXene fibers, SWCNTs can align along the fiber axis during spinning and act as one-dimensional anisotropic scaffolds to guide MXene flakes into a more parallel arrangement⁸. As a result, the orientational order is restored to a level comparable to that of pristine MXene fibers.

154 Therefore, the apparently higher compactness but lower orientation in C-MXene fibers is
155 not contradictory, but rather reflects the difference between CMC-induced densification
156 and flow-induced nanosheet alignment.
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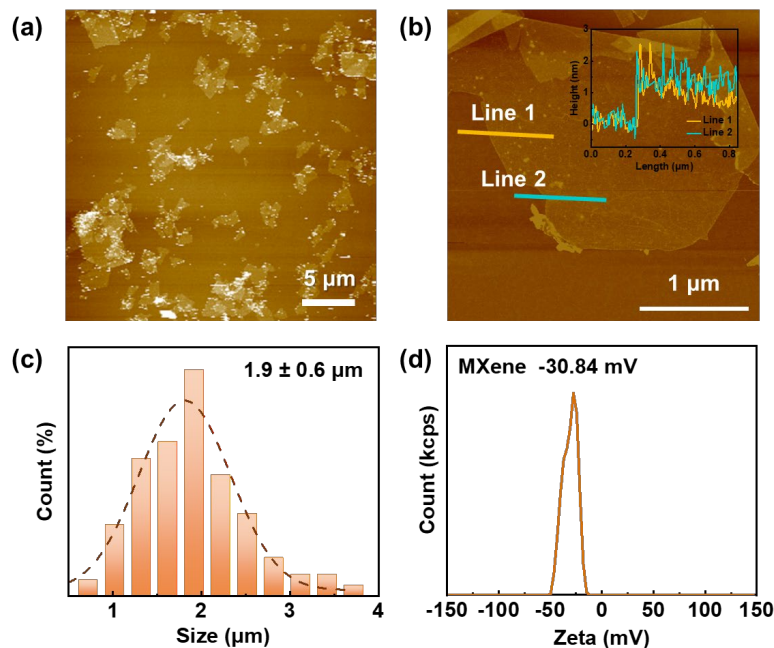
Supplementary Figures



Supplementary Fig. 1. (a) Photos of CMC-SWCNT and SWCNT aqueous solutions after standing for 0.5 h and 2 h. (b) Zeta potential of CMC-SWCNT and SWCNT aqueous solutions.

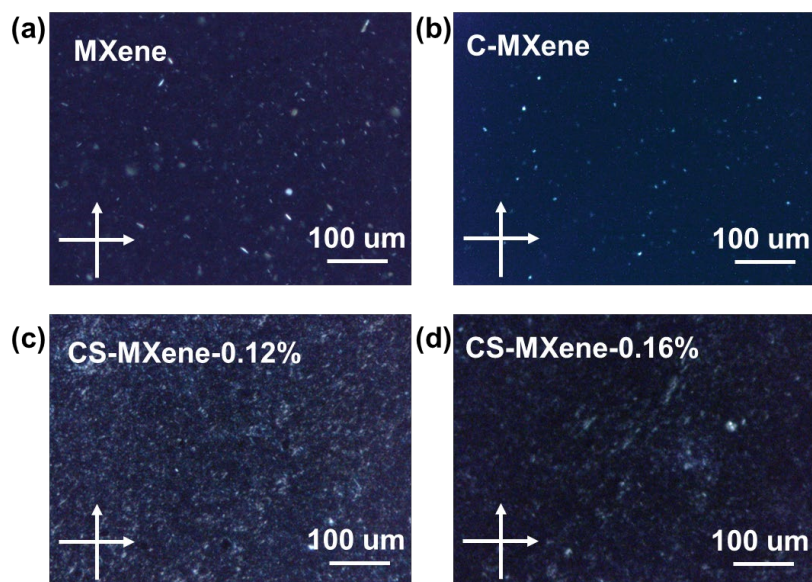


Supplementary Fig. 2. HRTEM images of CS molecular cords dispersed in aqueous solution.



Supplementary Fig. 3. Characterization of MXene. (a) and (b) AFM images of MXene nanosheets. (c) Lateral size distribution of MXene nanosheets. MXene nanosheets with a lateral size of about 1.9 μm and a thickness of about 1.8 nm were synthesized and used in this work. (d) Zeta potential of MXene dispersion.

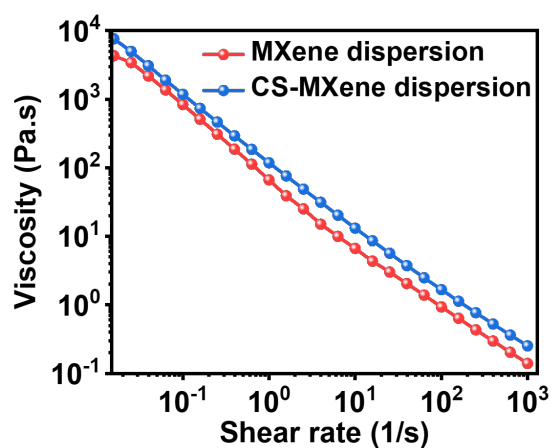
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176 **Supplementary Fig. 4.** POM images of the liquid-crystalline phase of (a) neat MXene
 177 dispersion (220 mg mL^{-1}), (b) C-MXene dispersion (220 mg mL^{-1} , weight ratio of MXene:
 178 CMC=1:0.0012), (c) CS-MXene dispersion (220 mg mL^{-1} of MXene, weight ratio of
 179 MXene: CS =1:0.0012) and (d) CS-MXene dispersion (220 mg mL^{-1} of MXene, weight
 180 ratio of MXene: CS = 1:0.0016), which exhibited optical birefringence between two
 181 crossed polarizers, indicating the formation of a liquid-crystalline phase. The formation of
 182 nematic liquid crystalline phase is attributed to the local orientation of the MXene in the
 183 dispersion.

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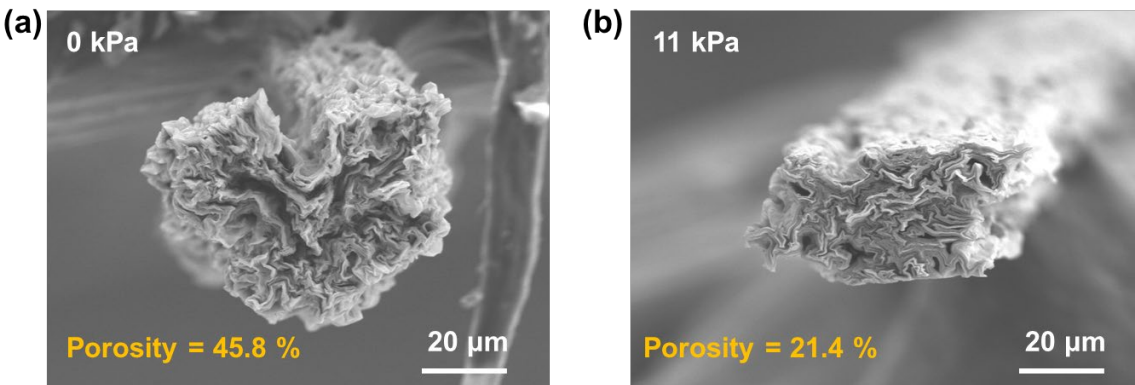


Supplementary Fig. 5. Viscosity versus shear rate for MXene (220 mg mL⁻¹) and CS-MXene (220 mg mL⁻¹ of MXene, weight ratio of MXene: CS = 1:0.0012) dispersions.



Supplementary Fig. 6. Optical images showing the apparatus for the mild fabrication of CS-MXene fibers, including wet-spinning, crosslinking, and hot-pressing processes.

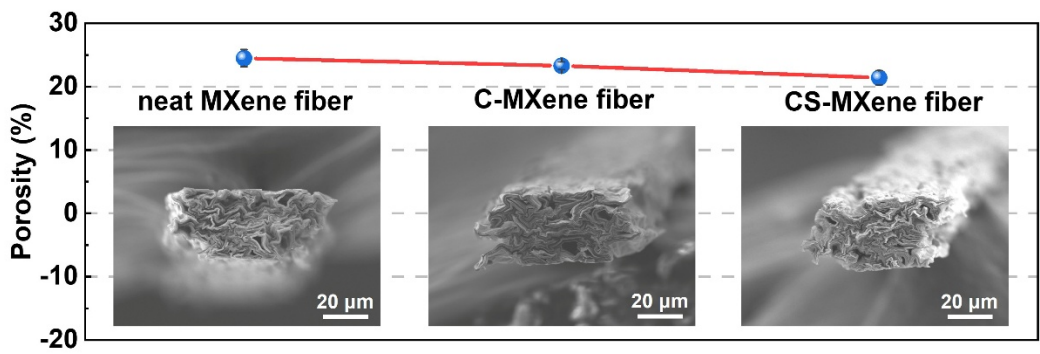
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195 **Supplementary Fig. 7.** SEM images and porosity of CS-MXene fibers before (a) and after
196 (b) hot-pressing under 11 kPa and 70°C for 5 min.

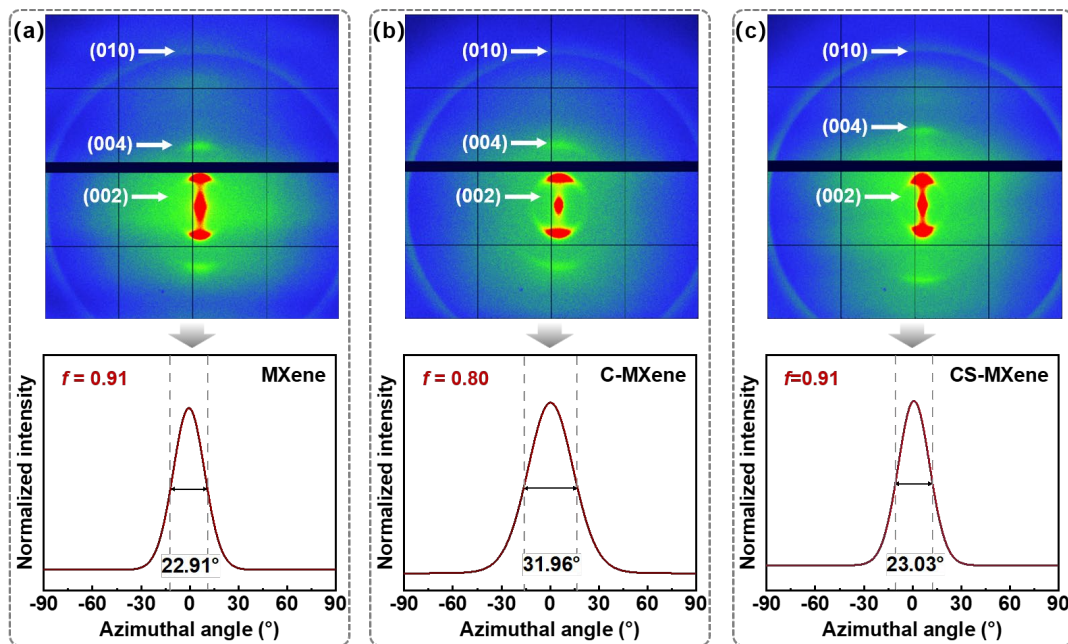
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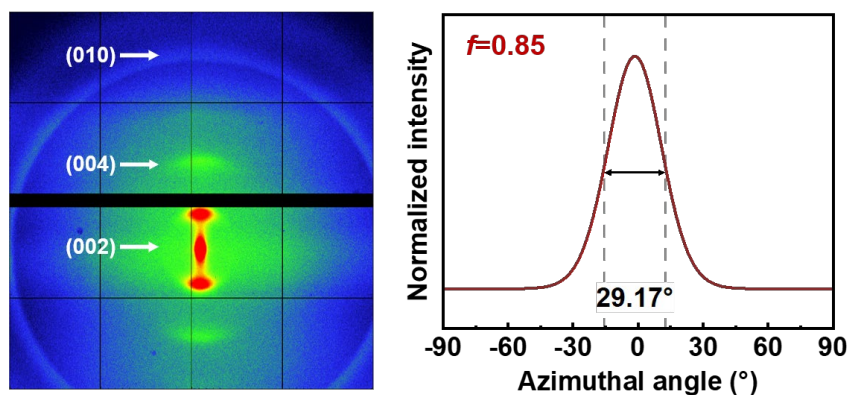
199 **Supplementary Fig. 8.** The porosity of neat MXene, C-MXene, and CS-MXene fibers.
200 Inset SEM images show the cross-section of neat MXene, C-MXene, and CS-MXene fibers.
201 The error bars represent the mean $\pm$ standard deviation ($n = 3$ independent experiments).

202



Supplementary Fig. 9. 2D WAXS patterns and azimuthal scanning profiles of (a) MXene, (b) C-MXene, and (c) CS-MXene fibers at (002) along the φ direction in the region of $\pm 90^\circ$. The Herman orientation factor (f) was calculated from the azimuthal scan along the (002) peak in the WAXS patterns. The high value of f indicates the high degree of MXene nanosheet alignment along the fiber axial direction in neat MXene, C-MXene, and CS-MXene fibers.

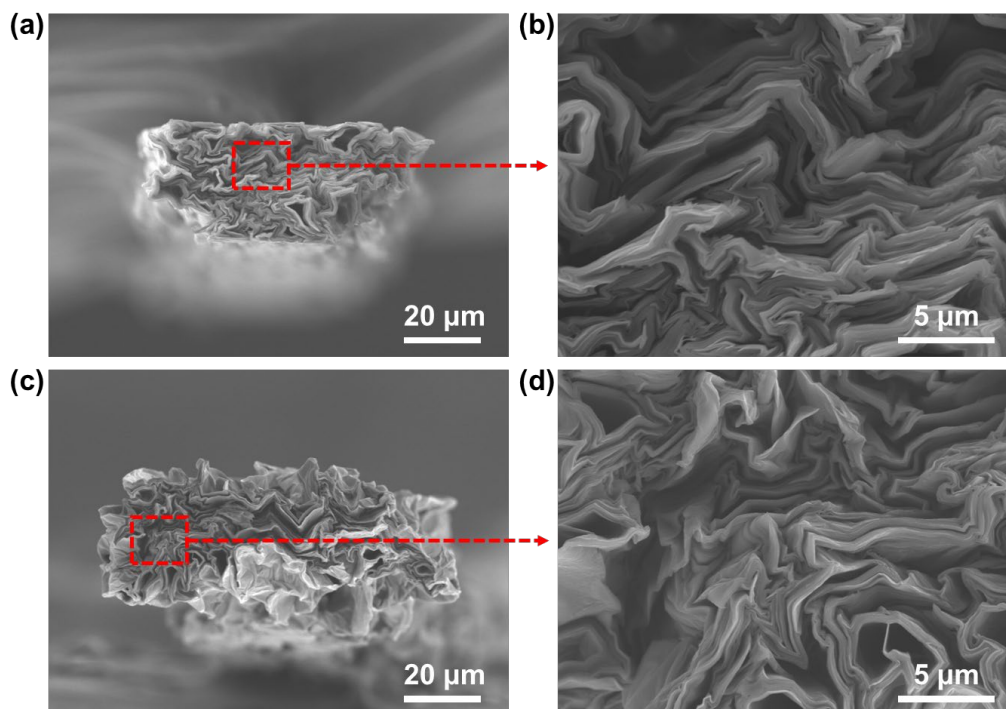
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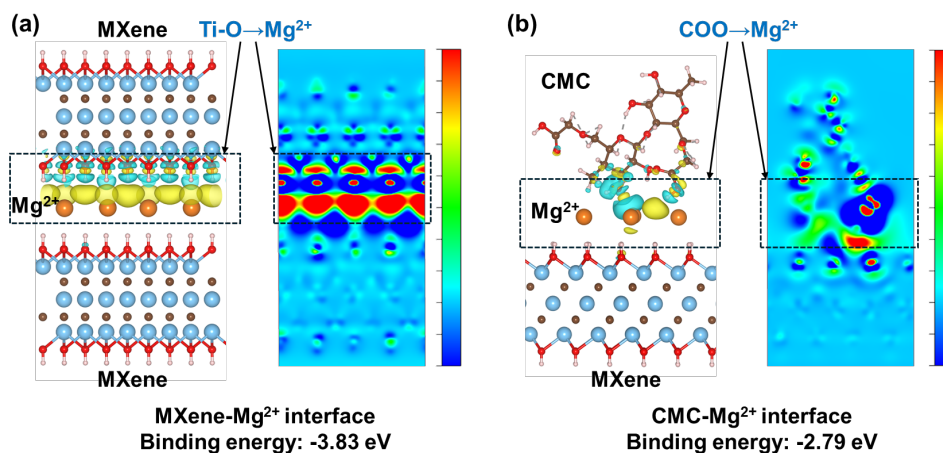
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213 **Supplementary Fig. 10.** 2D WAXS patterns and azimuthal scanning profiles of C-MXene
 214 (content 0.06% CMC) at (002) along the φ direction in the region of $\pm 90^\circ$. The Herman
 215 orientation factor (f) was calculated from the azimuthal scan along the (002) peak in the
 216 WAXS patterns. The orientation factor of C-MXene (content 0.06% CMC) is higher than
 217 C-MXene (content 0.12% CMC), confirming that CMC disrupts the orientational order of
 218 the MXene sheets.

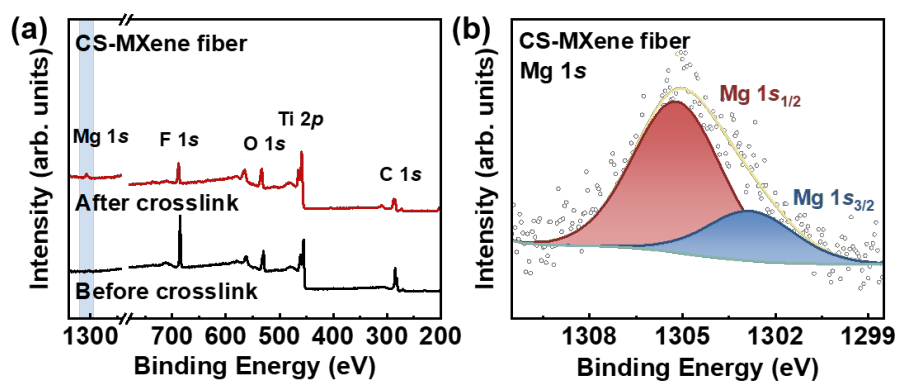
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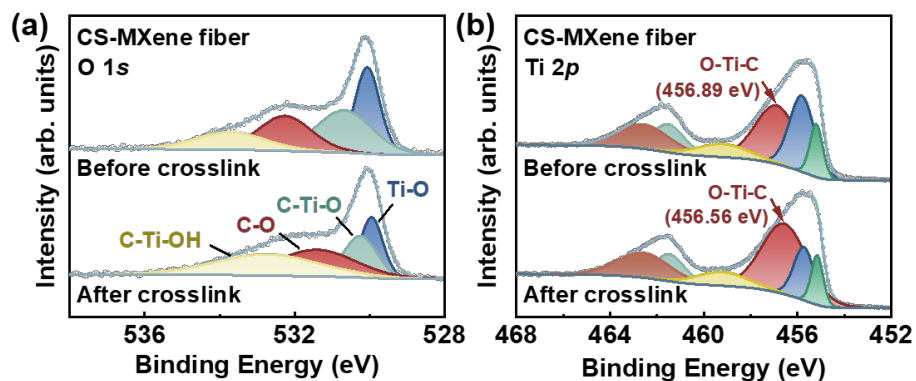
Supplementary Fig. 11. SEM images of the liquid nitrogen-fractured cross-section for neat MXene (a, b) and C-MXene fibers (c, d).



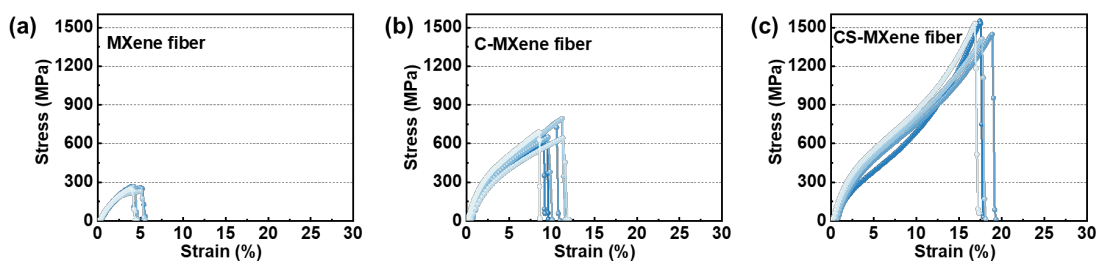
Supplementary Fig. 12. DFT calculations for coordination bonds of $\text{Ti-O} \rightarrow \text{Mg}^{2+}$ in MXene-Mg²⁺ interface (a) and $\text{COO} \rightarrow \text{Mg}^{2+}$ in CMC-Mg²⁺ interface (b).



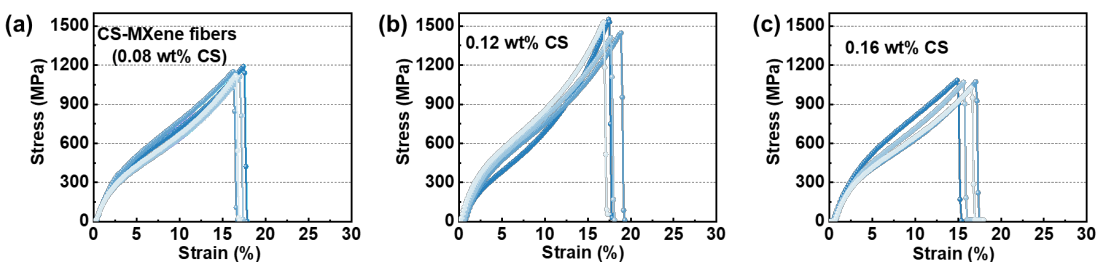
Supplementary Fig. 13. The XPS survey spectra before and after Mg^{2+} crosslinking (a) and Mg 1s spectra (b) of the CS-MXene fiber.



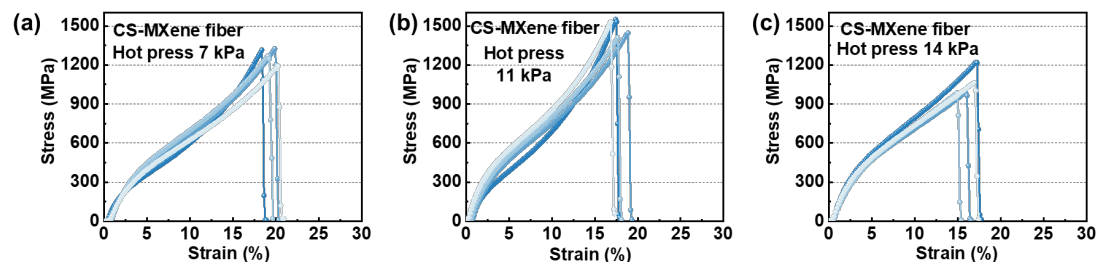
Supplementary Fig. 14. The XPS O 1s (a) and Ti 2p (b) spectra of the CS-MXene fiber before and after Mg^{2+} crosslinking.



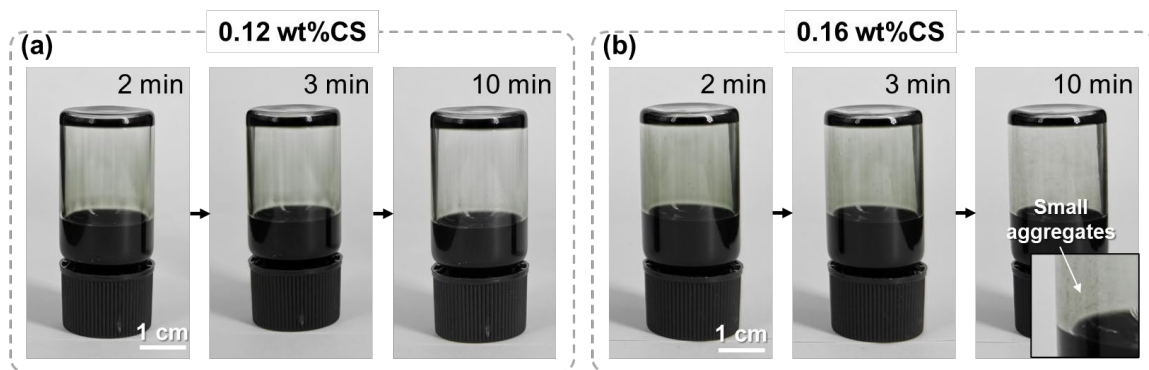
Supplementary Fig. 15. Tensile stress-strain curves of (a) neat MXene, (b) C-MXene, and (c) CS-MXene fibers.



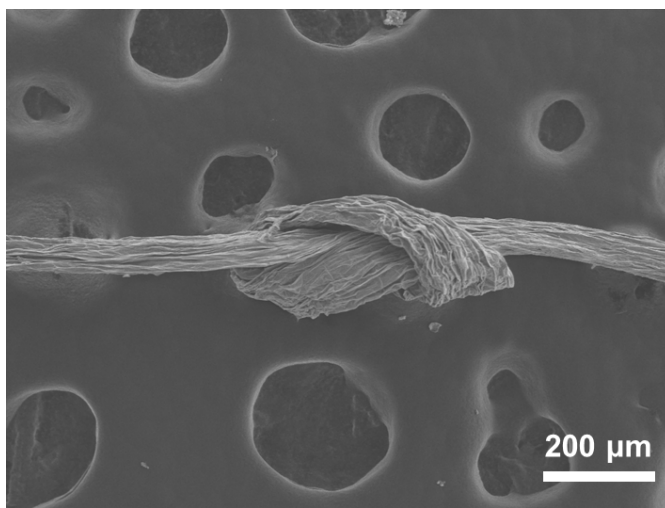
Supplementary Fig. 16. Tensile stress-strain curves of CS-MXene fibers with 0.08 wt% (a), 0.12 wt% (b), and 0.16 wt% (c) CS contents.



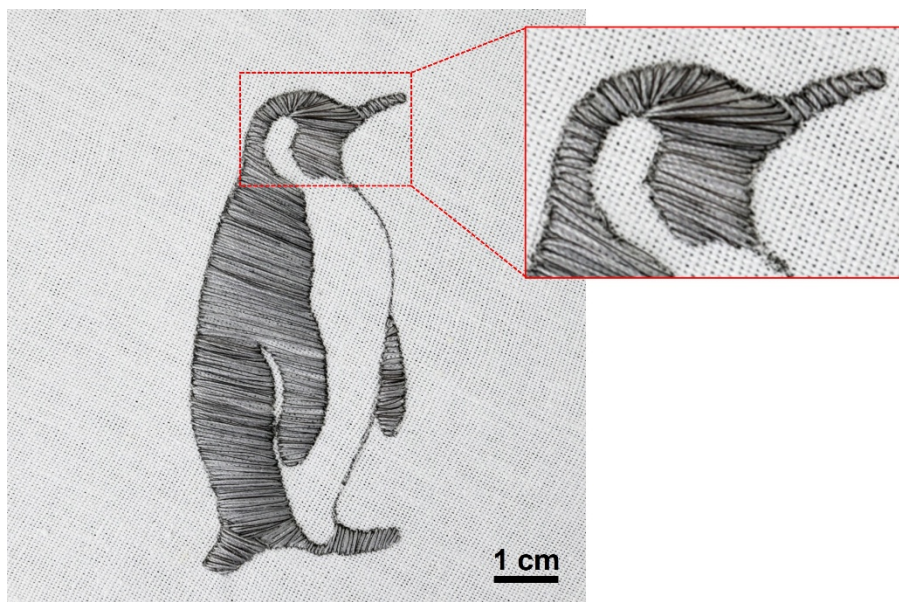
Supplementary Fig. 17. Tensile stress-strain curves of CS-MXene fibers at hot-pressing pressures of (a) 7 kPa, (b) 11 kPa, and (c) 14 kPa.



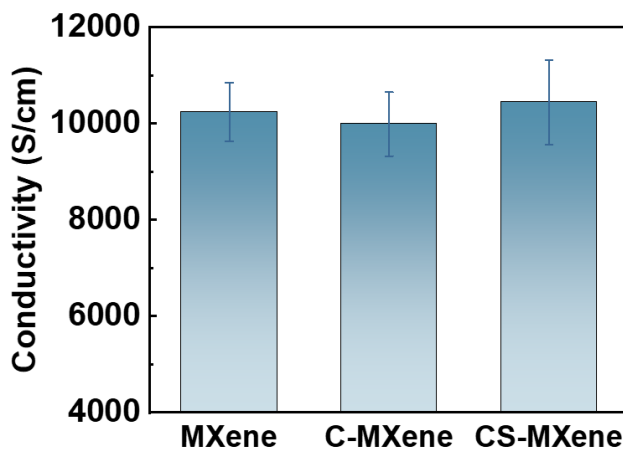
Supplementary Fig. 18. Photos of CS-MXene aqueous solutions at 0.12 wt% CS loading (a) and 0.16 wt% CS loading (b) after standing for 2-10 min.



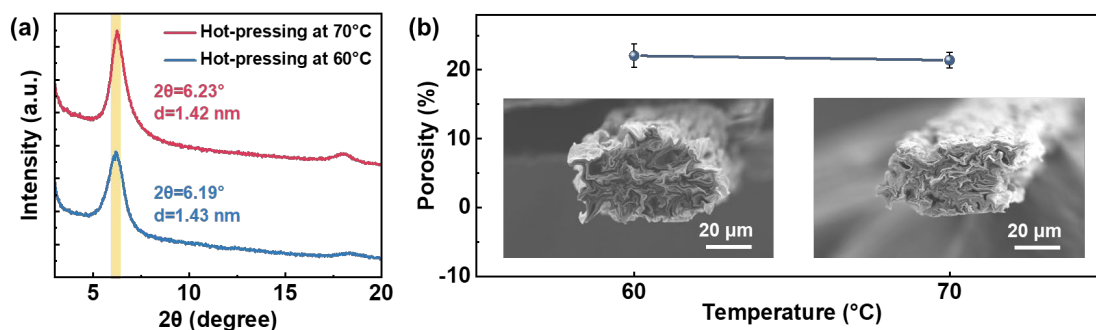
Supplementary Fig. 19. SEM image of a knotted CS-MXene fiber.



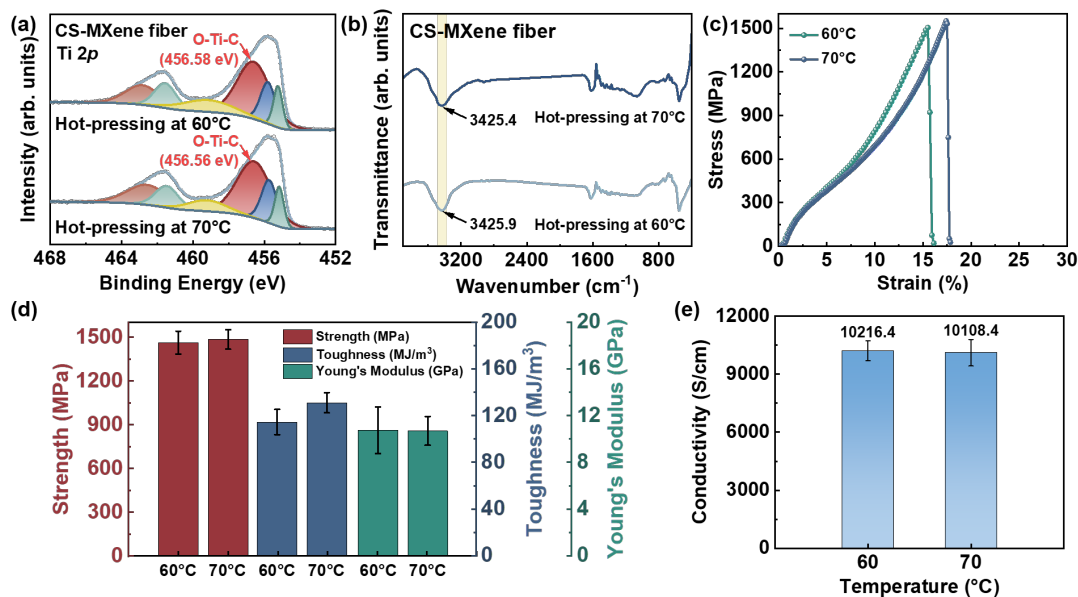
Supplementary Fig. 20. Photograph of a textile with a penguin pattern weaved with CS-MXene fibers and commercial cotton fibers.



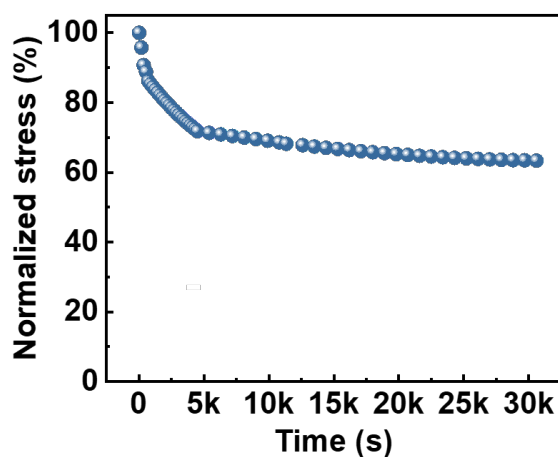
Supplementary Fig. 21. Electrical conductivity of neat MXene, C-MXene, and CS-MXene fibers. More than four samples were measured for each fiber type. The error bars represent the mean $\pm$ standard deviation ($n = 4$ independent experiments).



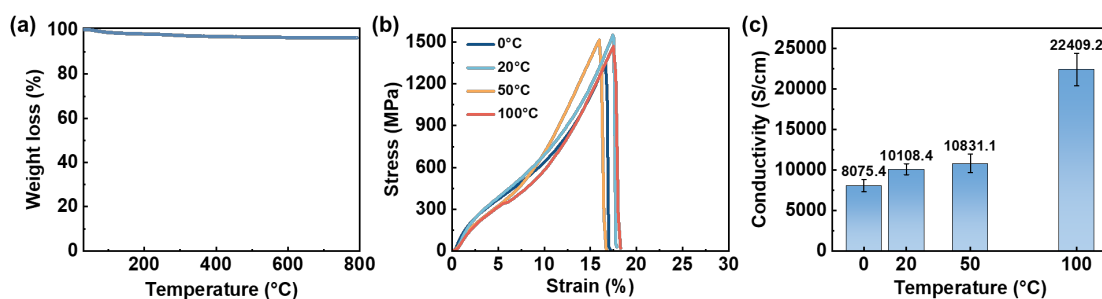
Supplementary Fig. 22. Morphology characterization of CS-MXene fibers at hot-pressing temperatures before and after the methanol boiling point. XRD patterns (a) and porosity (b) of CS-MXene fibers at hot-pressing temperatures of 60°C and 70°C. The error bars represent the mean $\pm$ standard deviation ($n = 3$ independent experiments) Inset SEM images show the cross-section of CS-MXene fibers at hot-pressing temperatures of 60°C and 70°C. Hot pressing at 70°C allows for more complete removal of solvents between fiber layers without affecting the fiber structure.



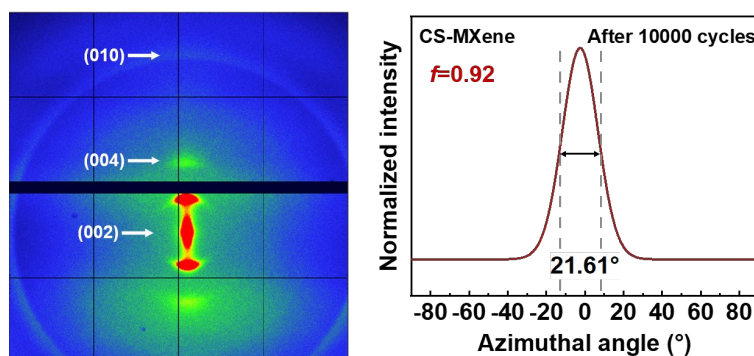
Supplementary Fig. 23. Comparisons of bonded structure, mechanical stability, and electrical stability for CS-MXene fibers at hot-pressing temperatures above and below the methanol boiling point. The XPS Ti 2p spectra (a) and FT-IR spectra (b) for the CS-MXene fiber at hot-pressing temperatures of 60°C and 70°C. Tensile stress-strain curves (c), comparison of mechanical properties (d) and conductivity (e) for the CS-MXene fiber at hot-pressing temperatures of 60°C and 70°C. The error bars represent the mean $\pm$ standard deviation ($n = 4$ independent experiments). The above results indicate that CS-MXene fibers prepared at hot-pressing temperatures around the boiling point of methanol exhibit consistent microstructures, mechanical properties, and electrical properties, demonstrating that a hot-pressing temperature of 70°C does not adversely affect the fibers. The error bars represent the mean $\pm$ standard deviation ($n = 4$ independent experiments).



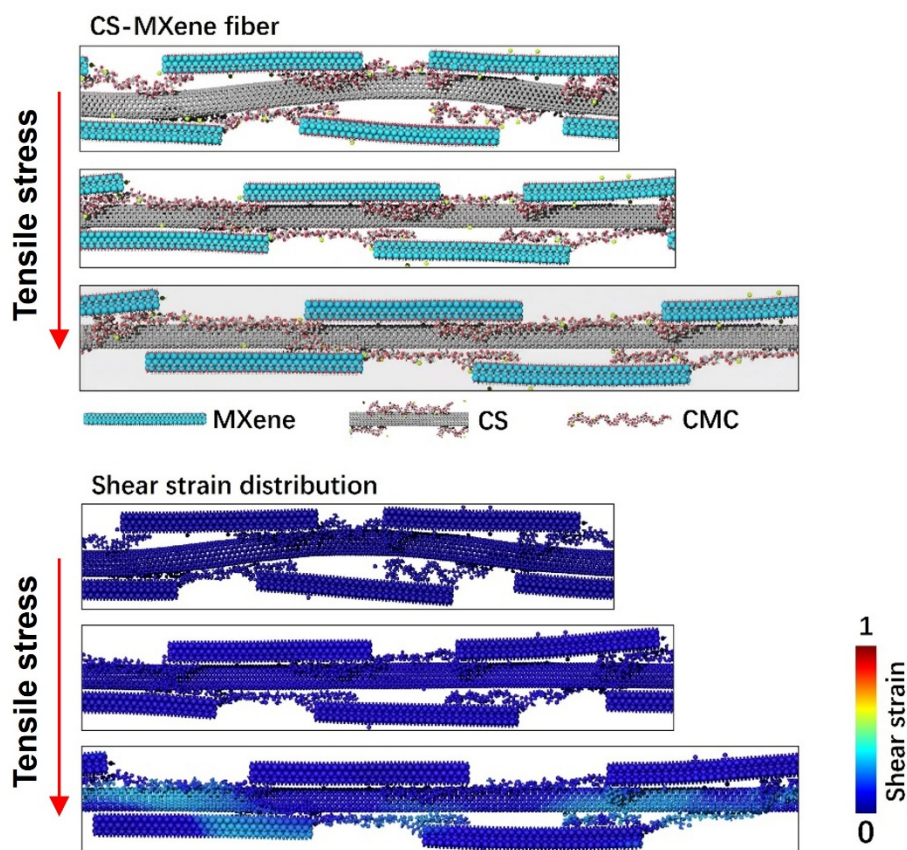
Supplementary Fig. 24. Stress relaxation curves of CS-MXene fibers under 12%-15% strain.



Supplementary Fig. 25. (a) TGA curves of CS-MXene fiber. Tensile stress-strain curves (b) and electrical conductivity (c) of CS-MXene fibers at temperature of 0 °C, 20 °C, 50 °C, and 100 °C The error bars represent the mean $\pm$ standard deviation ($n = 4$ independent experiments)..

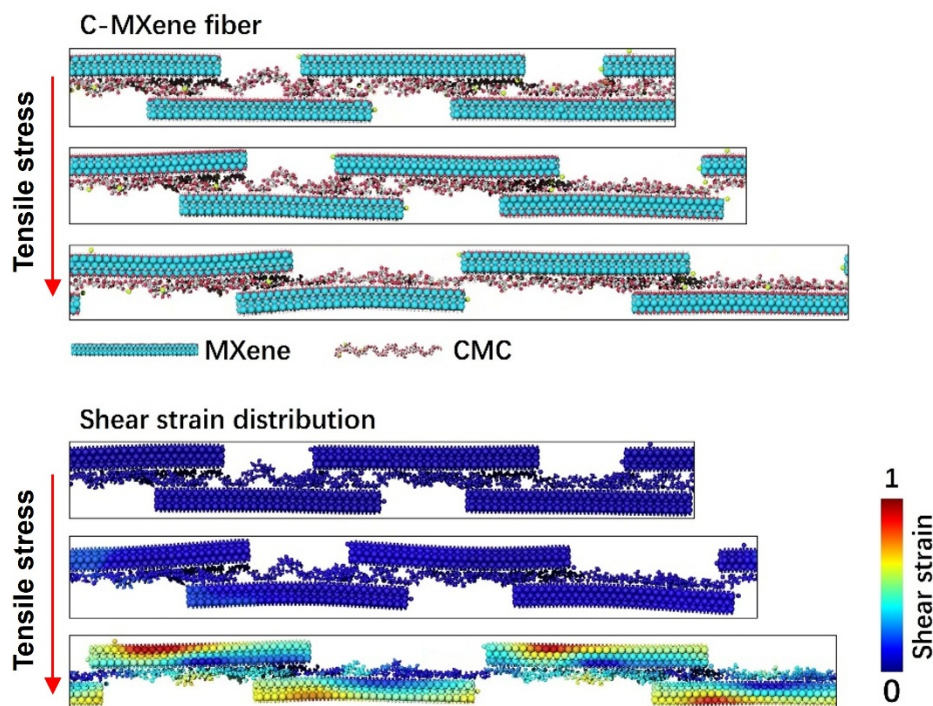


Supplementary Fig. 26. 2D WAXS patterns and azimuthal scanning profiles of CS-MXene fibers at (002) along the φ direction in the region of $\pm 90^\circ$ after 10,000 stretching cycles.



Supplementary Fig. 27. MD simulation snapshots and shear strain distribution of CS-MXene fibers. In CS-MXene fibers, CS sustained most interfacial shear deformation.

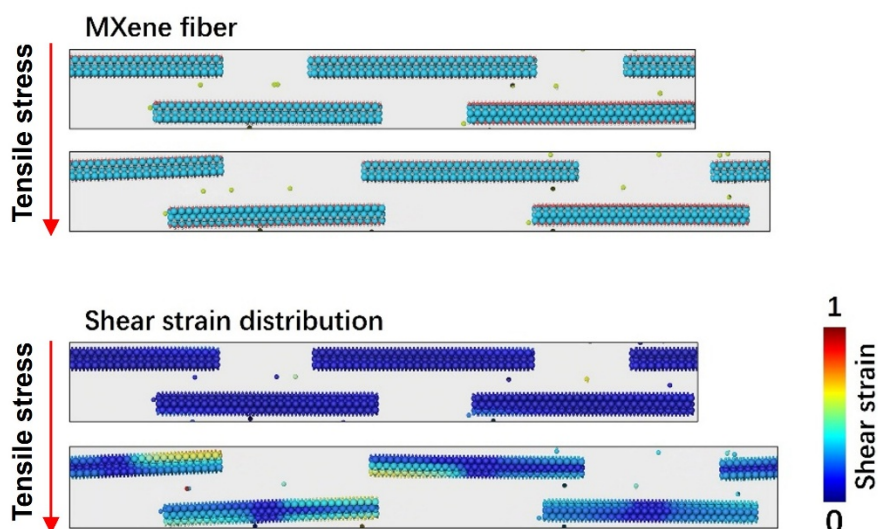
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306 **Supplementary Fig. 28.** MD simulation snapshots and shear strain distribution of C-
 307 MXene fibers. In C-MXene fibers, MXene sustained most interfacial shear deformation.

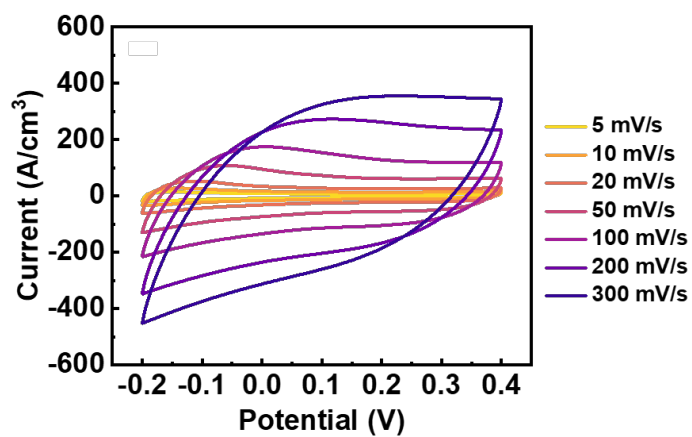
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310 **Supplementary Fig. 29.** MD simulation snapshots and shear strain distribution of neat
 311 MXene fibers. In MXene fibers, MXene sustained most interfacial shear deformation.

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314 **Supplementary Fig. 30.** CV curves of CS-MXene fiber electrodes at different scan rates.

315

316 **Supplementary Table 1.** Comparison of the mechanisms of strengthening and toughening,
 317 preparation process, tensile strength, toughness, extensibility, Young's modulus, and
 318 electrical conductivity of CS-MXene fibers with other reported MXene-based fibers.

Fiber material	Mechanisms of strengthening and toughening	Ink concentration / additive content	Coagulation bath	Post-treatment / processing conditions	Tensile strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Conductivity (S cm ⁻¹)	Ref.
MXene-Mg ²⁺	Orientation optimization and ion cross-linking	~25 mg mL ⁻¹ , -	MgSO ₄ / Ethanol/H ₂ O	Using a flat spinning nozzle	118	1.6	~1	7200	9
MXene		-		-	167.1	0.5	~0.4	11360.4	10
MXene/GA/PVA	Heat-stretching-induced stress densification and	-, 5 wt% GA	NH ₄ Cl/NH ₃ ·H ₂ O	-	565.2	6	19.2	8110	10
MXene/GA/PVA /PC	polymer protective layer	-, 5 wt% GA, 5 wt% PVA		Thermal drawing	585.5	18.0	66.7	8802.4	10
Large sized MXene	Liquid crystal-assisted alignment	-	NH ₄ Cl/NH ₄ O H	-	63.9	0.22	0.091	7713	11
Oriented MXene	Ion cross-linking	25 mg mL ⁻¹ , -	NH ₄ Cl/NH ₄ O H	Draw ratio: 3	344	0.28	/	12504	12
MXene/COC/PC	Interface interlocking, high orientation and low porosity	160 mg mL ⁻¹ , -	-	Fluidics-assisted thermal drawing	707.7	25.2	125.1	11960	13
MXene@ANF	Outer protective layer	50 mg mL ⁻¹ , -	NH ₄ Cl	Stretching ratio: 1.1	502.9	13.1	48.1	2969	14
MXene/PEDOT: PSS	Composite elastic polymer	-, 30 wt% PEDOT:PSS	98 wt% H ₂ SO ₄	-	58.1-142.6	1.1-12.3	5.2-7.5	548-1489	15

Fiber material	Mechanisms of strengthening and toughening	Ink concentration / additive content	Coagulation bath	Post-treatment / processing conditions	Tensile strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Conductivity (S cm ⁻¹)	Ref.
MXene/Graphene	Liquid crystal-assisted alignment and interlayer interactions	~20 mg mL ⁻¹ , 90 wt% MXene	Isopropanol/DI / 5 wt% CaCl ₂	HI reduced	12.9	~3.7	~0.2	290	16
MXene/GO Fiber	Liquid crystal-assisted alignment	88 wt% MXene ~10 mg mL ⁻¹	Acetic acid 0.5 wt %	Hypophosphorous acid reduced	161	~10.2	12.4	1142	17
LC MXene	Liquid crystal-assisted alignment	26.5 mg mL ⁻¹ , -	chitosan solution	-	40.5	~1.8	~0.4	7750	18
MXene/CNT	Composite 1D flexible materials	-	-	Scroll structure	38.4	~1.0	~4.7	2.7	19
MXene/RGO	Composites of 2D materials and ion cross-linking	-, 20 wt% MXene	Ethanol/H ₂ O/5 wt% CaCl ₂ /0.5 wt% PVA/0.005 wt% glutaraldehyde	HI reduced	534	1.4	448	16.6	20
MXene-CNFs@ANFs	Dual-scale orientation	-, 20 wt% CNF	5 wt% NH ₄ Cl	-	506.7	15	/	12700	21
MXene/Alginate/PVA	Densification and ionic cross-linking	-, 20 wt% MXene	4 wt% CaCl ₂	-	364	7		1006	22
MXene/Zn ²⁺	Ionic cross-linking	~100 mg mL ⁻¹ , -	Ethanol/H ₂ O/0.05 M	Zn ²⁺ crosslinking	150		1.1	3637.9	23

Fiber material	Mechanisms of strengthening and toughening	Ink concentration / additive content	Coagulation bath	Post-treatment / processing conditions	Tensile strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Conductivity (S cm ⁻¹)	Ref.
			ZnSO ₄ /0.05 M						
			CaCl ₂						
RSF/PEI/MXene	Core-shell interface engineering	-	Ethanol/H ₂ O	-	214	9	15.2	1125	24
MXene/Kevlar/borate ions	Covalent interface enhancement and composite 1D fiber	20 mg mL ⁻¹ , -	Anhydrous acetic acid	Anneal for 4 hours at 90°C	242	0.8	1	1412	25
MXene/PR	Composite sliding polymer	150 mg mL ⁻¹ , 20 wt% PR	Acetic acid, 99.5%	MgSO ₄ /methanol bath, CDI/DMSO bath	500	27	60	1100	26
A-PPM fiber	Covalent cross-linking, slip interfaces, and pore regulation	170 mg mL ⁻¹ , 20 wt% PR, 7 wt% PEDOT:PSS	Acetic acid/DMSO	MgSO ₄ /methanol bath, CDI/DMSO bath, 1 wt% AgNWs/ethanol bath	1028	33.6	131.3	~10500	27
CS-MXene fiber	Molecular cord cross-layer edge bridging	220mg mL ⁻¹ , 0.12 wt% CS	Acetic acid	MgSO ₄ /methanol bath	1480±60	17.7±0.8	131±8.7	10438±875	This work

Supplementary Table 2. Comparison of the preparation process, tensile strength, toughness, extensibility, Young's modulus, and electrical conductivity of CS-MXene fibers with most reported graphene- and CNT-based fibers.

Fiber material	Ink concentration / additive content	Coagulation bath	Post-treatment / processing conditions	Tensile strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Conductivity (S cm ⁻¹)	Ref.
rGG-Ca ²⁺	-	1 wt% CaCl ₂	Wet-drawing Ca ²⁺ crosslinking	501.5	6.7	16.8	410	28
rGO	-	NaOH	Anneal at 220°C	115.0	~3.3	1.4	2.8	29
rGO-HPG	25–50 mg mL ⁻¹ rGO: HPG=0.1:2	NaOH/methano 1	-	165.0	~1.5	1.2	4.9	30
rGO-CTAB	10 mg mL ⁻¹ , -	0.5 mg/mL CTAB	Hydroiodic acid reduced	182.0	~4.0	3.2	35.0	31
rGO-NaDC	5–9 wt% NaDC	Ethanol	HI reduced	238.0	~2.3	2.4	308	32
Large and small sized graphene	15 mg mL ⁻¹	CaCl ₂	Anneal for 1 hours at 1800°C	1080	1.45	~6.8	2210	33
Reduced/f-GO fiber	-	80 wt% acetic acid	HI reduced, Draw ratio: 1.75	884.0	1.7	9.3	1060	34
RGO-PSE/AP-Ca ²⁺	15 mg mL ⁻¹ , -	Ethanol/H ₂ O/ 5 wt % CaCl ₂	HI reduced	740.1	3.7	18.7	433.5	35

Fiber material	Ink		Post-treatment / processing conditions	Tensile strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Conductivity (S cm ⁻¹)	Ref.
	concentration / additive content	Coagulation bath						
RGO-Chitosan-Ca ²⁺	- , 4.95 wt % CS	Ethanol/H ₂ O/ 5 wt % CaCl ₂	Hydroiodic acid reduced	743.6	6	26.3	179	36
Graphene/Carbon	- , 30 wt % GO	Ethyl acetate, H ₂ O/DMF	Anneal at 3000°C	2437	0.7	~15	3500	37
rGO	-	5 wt% CaCl ₂ / 0.25 M NH ₄ OH	HI reduced	272	~0.6	/	54	38
GGO-HPG	-	5 wt% CaCl ₂	Vc/KOH reduced	555	~5.5	18	526.1	39
rGO-Al ³⁺	15 mg mL ⁻¹ , -	Ethanol/H ₂ O/0. 5 M AlCl ₃	Draw ratio: 1.3, HI reduced	164.9	~10. 3	10	171.3	40
rGO-MXene	15 mg mL ⁻¹ , 50 wt% MXene	Glacier acetic acid	HI reduced	110.7	~7.6	3.8	743.1	41
LGO-SA	20 mg mL ⁻¹ , - 25–50 mg	Ethanol/H ₂ O/ 5 wt % CaCl ₂		784.9	~2.4	6.8	35.8	42
CRG-PVA	mL ⁻¹ , 63.5 wt% PVA	Acetone	-	160	~2.8	2.9	3.5	43
Graphene/PC	7 mg mL ⁻¹ , 10 wt % PR	Ethyl acetate /methanol/2 wt% CaCl ₂	Anneal at 1000°C	1450	1.8	14.5	840	44

	Ink		Post-treatment /	Tensile	Strai			
Fiber	concentration /	Coagulation	processing	strength	n	Toughness	Conductivity	Ref.
material	additive	bath	conditions	(MPa)	(%)	(MJ m ⁻³)	(S cm ⁻¹)	
	content							
Graphene/ PAN	-, 30 wt % PAN	Methanol/ H ₂ O	Anneal at 800°C	1100	1.27	7	/	45
rGO-Ca ²⁺ - PCDO	≈10 mg mL ⁻¹ , -	Ethanol/H ₂ O/5 wt % CaCl ₂	PCDO/tetrahydrof					
			uran bath,					
			HI reduced, Film- derived wound fibers	842	3.5	15.8	292.6	46
CNT fiber	CVD		1300°C	1300	4.6	/	/	47
SWCNT fiber	-	acetone	Heat treatment at 2000°C	1740	0.9	~8.1	2600	48
SWCNT fiber	-	-	CVD	~800	/	/	1400	49
CNT/PVA Yarns	-, 5 wt % PVA	-	Dry spinning, PVA/DMSO bath	2000	~1.3 7	/	920	50
CNT fiber	-			270	~12 %	/	1657	51
Cross- linked CNT ribbons	-, 30 wt % epoxy	-	CVD at 1100– 1200 °C	~3600	~2.8	/	4887	51
CNT/PDA fiber	-, 12.5 wt % py-PDA	-	CVD	4040	6.0	~125	~5000	52

Fiber material	Ink concentration / additive content	Coagulation bath	Post-treatment / processing conditions	Tensile strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Conductivity (S cm ⁻¹)	Ref.
CNT	-	-	CVD, heat treatment at 1050°C	1004	3.4	~18.9	~772	52
CNT	-	-	CVD	1900	5.28	~61.8	/	53
Twisted MWNY	-	-	CVD, twist assembled fibers	~300	/	~11.3	5000	54
Multilayered CNT	-	-	-	~700	/	~63	5000	55
CNT/SiC-WT	-, 10 wt% PCS	-	Pyrolysis at 800°C	1698.6	~1.7	/	6350	56
Cross-linked CNT	-, 8 wt% PEI-C	-	CVD, heat treatment at 120°C	2500	2.5	/	/	57
CNT/rGO	-	5 wt% CaCl ₂ /H ₂ O/0.2 5 M NH ₄ OH	HI reduced	690	~1.1	2.6	178	38
CNT	-	-	Continuous prestraining and rolling	8000	~4.0	/	350	58
D-PBO-CNTFs	-, 0.1 wt% PBO	-	PBOs-CSA baths	8200		170	29000	59

Fiber material	Ink concentration / additive content	Coagulation bath	Post-treatment / processing conditions	Tensile strength (MPa)	Strain (%)	Toughness (MJ m ⁻³)	Conductivity (S cm ⁻¹)	Ref.
Br-CNTFs	-, 4.8~26.1 wt% Br	-	Br intercalation by vapor transport	2460	2.58	/	106800	60
rGO	-, ~5.7 wt % rGO	NaOH/methanol	HI reduced	140.0	/	3.9	250	61
rGO-AgNW	-, 50 wt% AgNW	ethyl acetate	HI reduced	300.0	~5.5	7.8	930	62
Porous rGO fiber	-	CaCl ₂ /NaOH	Anneal at 220 °C	50.0	~0.5	0.3	25.1	63
graphene/graphite	-	Ethanol/H ₂ O/ 0.5 wt % CTAB	Anneal at 600 °C	~500	9.7	/	200	64
graphene fiber	-	Acetone/ethyl acetate	HI reduced	1900	~0.6	/	1040	65
RGO fiber	-	Methanol/ethyl acetone	HI reduced	217	3.6	/	1611	66
CS-MXene fiber	220mg mL ⁻¹ , 0.12 wt% CS	Acetic acid	MgSO ₄ /methanol bath	1480±60	17.7 ±0.8	131±8.7	10438±875	This work

Supplementary Table 3. Comparison of the specific capacitance, rate capacity, tensile strength and conductivity of CS-MXene fiber electrodes with other reported MXene-based fibers.

MXene fibers	Conductivity (S cm ⁻¹)	Specific capacitance (F cm ⁻³)	Rate capacity	Tensile strength (MPa)	Electrolyte	Ref.
MXene/CD	2405	1597 (2 A cm ⁻³)	87.3% at 12 A cm ⁻³	80	2 M H ₂ SO ₄	67
MXene/ Cellulose	916	1457.0 (1.5 A cm ⁻³)	71.2%	50	3 M H ₂ SO ₄	68
MXene/ Cellulose	1355	1531 (2 A cm ⁻³)	89.2% at 15 A cm ⁻³	76	3 M H ₂ SO ₄	69
MXene/ CMC	1073	885.0 (1 A cm ⁻³)	83.6% at 10 A cm ⁻³	81	2 M H ₂ SO ₄	70
MXene/PR	1100	362 (5 mV s ⁻¹)	/	502	1 M H ₂ SO ₄	26
S-MXene (0.31 μm)	~3512	1265 (5 mV s ⁻¹)	54.8% at 1 V s ⁻¹	18	1 M H ₂ SO ₄	18
L-MXene (3.1 μm)	~7748	1114 (5 mV s ⁻¹)	60.4% at 1 V s ⁻¹	42	1 M H ₂ SO ₄	18
MXene /PEDOT:PS S	1489	614.5 (5 mV s ⁻¹)	61% at 1Vs-1	58.1	1 M H ₂ SO ₄	15
MXene/ RGO	72.3	341 (0.5 A cm ⁻³)	37% at 10 A cm ⁻³	132	1 M H ₂ SO ₄	17

MXene fibers	Conductivity (S cm ⁻¹)	Specific capacitance (F cm ⁻³)	Rate capacity	Tensile strength (MPa)	Electrolyte	Ref.
MXene/ ANF	1025	807 (3 A cm ⁻³)	47% at 15 A cm ⁻³	104	3 M H ₂ SO ₄	71
O-fat MXene	7200	1360 (5 mV s ⁻¹)	49.6% at 1 V s ⁻¹	118	1 M MgSO ₄	9
MXene/ CNT	1142	295 (5 mV s ⁻¹)	19% at 1 V s ⁻¹	61	1 M H ₂ SO ₄	72
MXene/ RGO	/	542 (0.25 A cm ⁻³)	56% at 2 A cm ⁻³	40	1 M H ₂ SO ₄	73
MXene/ L-MXene	2813	1560 (1 A cm ⁻³)	79% at 20 A cm ⁻³	123	1 M H ₂ SO ₄	74
MXene/RG O/PEDOT:P SS	/	475 F g ⁻¹ (5 mV s ⁻¹)	77% at 1 V s ⁻¹	118	3 M H ₂ SO ₄	75
MXene/ANF /PEDOT:PS S	736.8	940 (2 A cm ⁻³)	63% at 20 A cm ⁻³	128.1	1 M H ₂ SO ₄	76
Ac-MXene	3545	1570.5 (5 mV s ⁻¹)	78.2% at 1 V s ⁻¹	205.5	1 M H ₂ SO ₄	77
Cs-MXene	2215	1083.1 (5 mV s ⁻¹)	/	261.2	1 M H ₂ SO ₄	77
MXene/ CNC	2980	945 (2 mV s ⁻¹)	~40% at 0.5 V s ⁻¹	60	1 M H ₂ SO ₄	78

MXene fibers	Conductivity (S cm ⁻¹)	Specific capacitance (F cm ⁻³)	Rate capacity	Tensile strength (MPa)	Electrolyte	Ref.
MXene ribbon	2458	309 F g ⁻¹ (1 A g ⁻¹)	74.76% at 10 A g ⁻¹	40	1 M H ₂ SO ₄	79
MXene/ rGO	2900	586.4	53% at 0.1 V s ⁻¹	12.9	1 M H ₂ SO ₄	16
B-MXene/ Kevlar	1412	885 (2 A cm ⁻³)	90% at 10 A cm ⁻³	242.6	3 M H ₂ SO ₄	25
N-Doped MXene	2295	1302	83% at 10 A cm ⁻³	64	1 M H ₂ SO ₄	80
MXene/ CNT	270	92 (1 A cm ⁻³)	68% at 5 A cm ⁻³	38.4	6 M LiCl	19
PANI@ MXene/GQ Ds	/	1829 (1 A g ⁻¹)	81.02% at 10 A g ⁻¹	1.2	1 M H ₂ SO ₄	81
MXene/ CNF-C	3007	1253 (1 A cm ⁻³)	81.8% at 10 A cm ⁻³	258	2 M H ₂ SO ₄	82
RSF/PEI/ MXene	1125	829 (1 mV s ⁻¹)	89.7% at 0.2 V s ⁻¹	214.3	3 M H ₂ SO ₄	24
CS-MXene fiber	10438±875	1645 (5 mV s ⁻¹)	45% at 0.3 V s ⁻¹	1480±60	1 M H ₂ SO ₄	Thiswor k

Supplementary Table 4. Summary of the mechanical and electrical properties of the above fiber samples. The error bars represent the mean $\pm$ standard deviation. More than four samples were measured for each fiber type.

Fiber samples	Strength (MPa)	Extensibility (%)	Toughness (MJ m ⁻³)	Young's Modulus (GPa)	Conductivity (S cm ⁻¹)
CS-MXene fibers	1484.3 $\pm$ 66.6	17.7 $\pm$ 0.8	131.0 $\pm$ 8.7	10.7 $\pm$ 1.2	10438.1 $\pm$ 875.2
C-MXene fibers	685.4 $\pm$ 68.6	9.9 $\pm$ 1.1	43.0 $\pm$ 7.4	13.7 $\pm$ 1.4	9982.7 $\pm$ 662.6
Neat MXene fibers	253.3 $\pm$ 15.8	4.4 $\pm$ 0.6	7.6 $\pm$ 1.1	8.8 $\pm$ 0.3	10236.2 $\pm$ 607.2

Supplementary Table 5. Summary of the mechanical and electrical properties of the above fiber samples. The error bars represent the mean $\pm$ standard deviation. More than four samples were measured for each fiber type.

CS contents (wt%)	Strength (MPa)	Extensibility (%)	Toughness (MJ m ⁻³)	Young's Modulus (GPa)	Conductivity (S cm ⁻¹)
0.08%	1143.6 $\pm$ 35.2	16.8 $\pm$ 0.5	106.0 $\pm$ 7.7	11.6 $\pm$ 1.6	10108.4 $\pm$ 685.7
0.12%	1484.3 $\pm$ 66.6	17.7 $\pm$ 0.8	131.0 $\pm$ 8.7	10.7 $\pm$ 1.2	10438.1 $\pm$ 875.2
0.16%	1060.1 $\pm$ 24.8	16.0 $\pm$ 1.0	95.6 $\pm$ 2.4	12.6 $\pm$ 1.4	7169.6 $\pm$ 541.9

Supplementary Table 6. Summary of the mechanical and electrical properties of the above fiber samples. The error bars represent the mean $\pm$ standard deviation. More than four samples were measured for each fiber type.

Hot-pressing pressure (kPa)	Strength (MPa)	Extensibility (%)	Toughness (MJ m ⁻³)	Young's Modulus (GPa)	Conductivity (S cm ⁻¹)
7	1275.4 $\pm$ 61.3	19.4 $\pm$ 0.8	127.2 $\pm$ 10.1	10.0 $\pm$ 1.3	9458.5 $\pm$ 359.6
11	1484.3 $\pm$ 66.6	17.7 $\pm$ 0.8	131.0 $\pm$ 8.7	10.7 $\pm$ 1.2	10438.1 $\pm$ 875.2
14	1064.5 $\pm$ 109.9	16.2 $\pm$ 1.0	103.9 $\pm$ 15.3	13.3 $\pm$ 0.8	8871.6 $\pm$ 566.4

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