Electronic Supplementary Information

Surface Nitrided MXene Sheet with Outstanding Electroconductivity and Oxidation Stability

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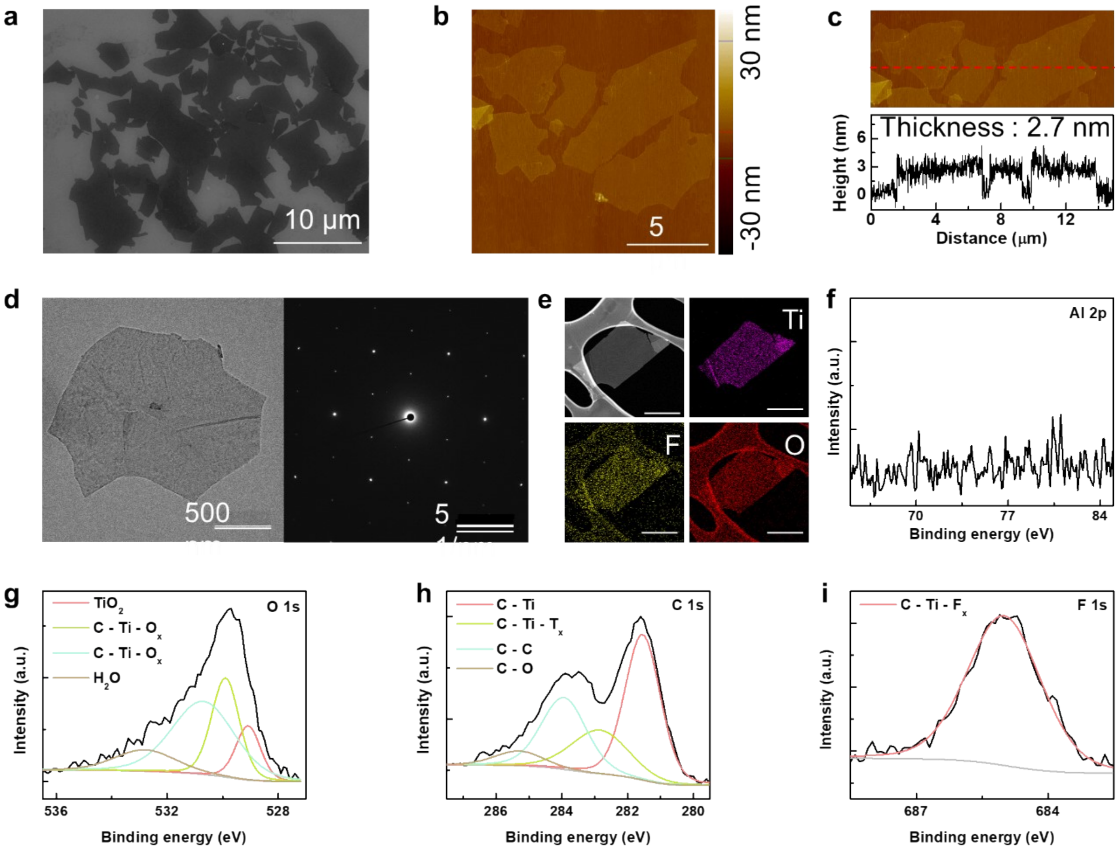

Fig. S1 Characterization of the as-synthesized MXene. **a** SEM image of MXene sheets. **b, c** AFM image and profile of an MXene monolayer. **d** TEM image, SAED pattern, and **e** EDS maps of an MXene monolayer. **f** Al 2p XPS spectrum showing the complete elimination of the Al layer in MXene. **g** O 1s, **h** C 1s, and **i** F 1s XPS spectra of as-synthesized MXene.

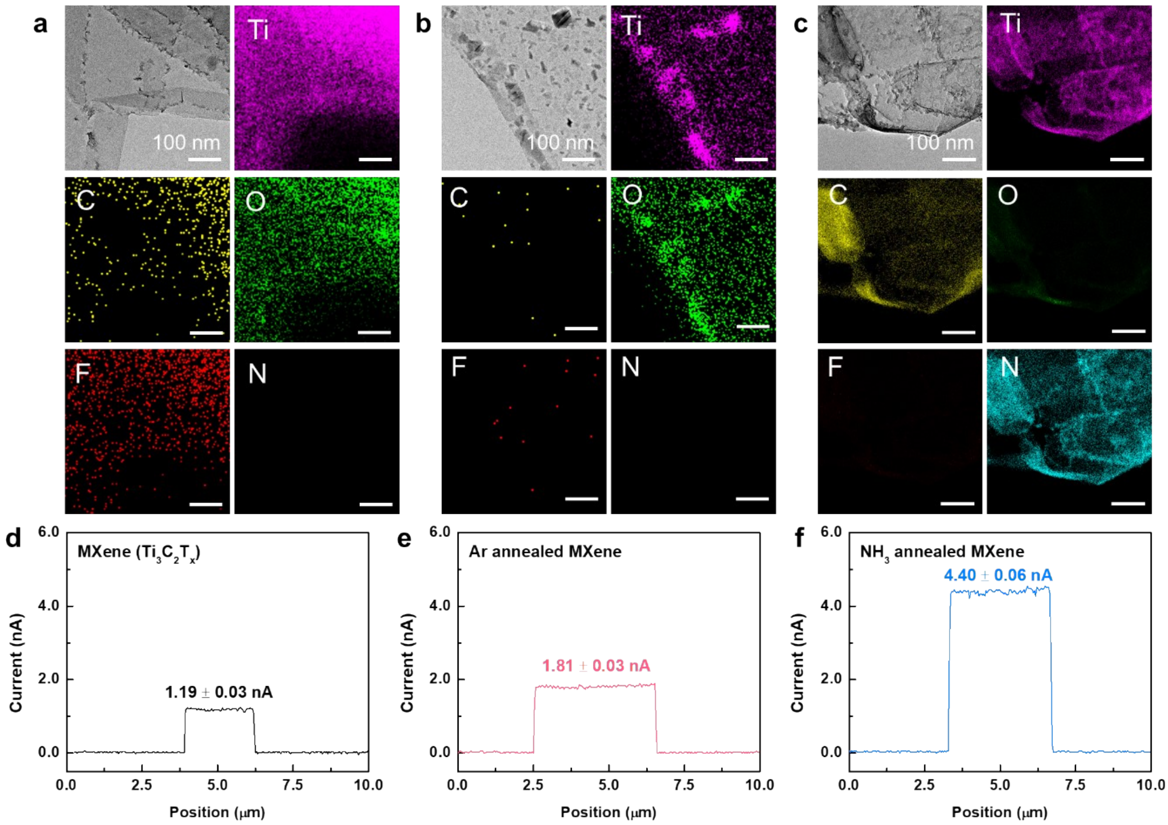

Fig. S2 TEM–EDS and line profile of current maps of **a, d** as-prepared MXene, **b, e** Ar-MXene, and **c, f** NH_3-MXene. The current maps are presented in Figure 1.

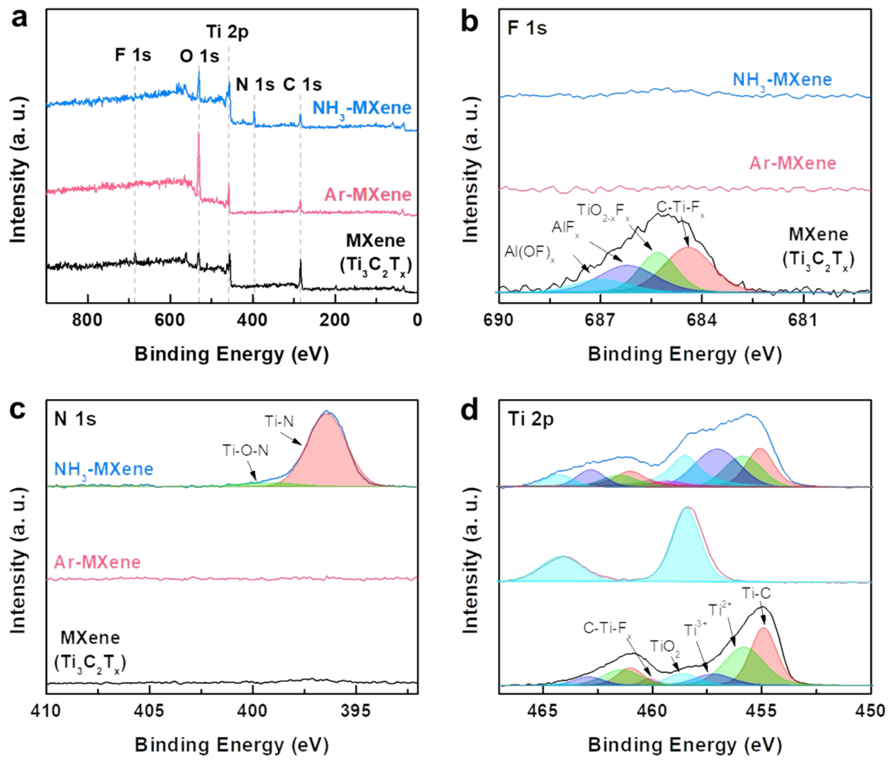

Fig. S3 Chemical state of as-prepared MXene and MXene annealed in argon (Ar-MXene) or ammonia gas (NH_3-MXene). XPS spectra of **a** survey, **b** F 1s, **c** N 1s, and **d** Ti 2p.

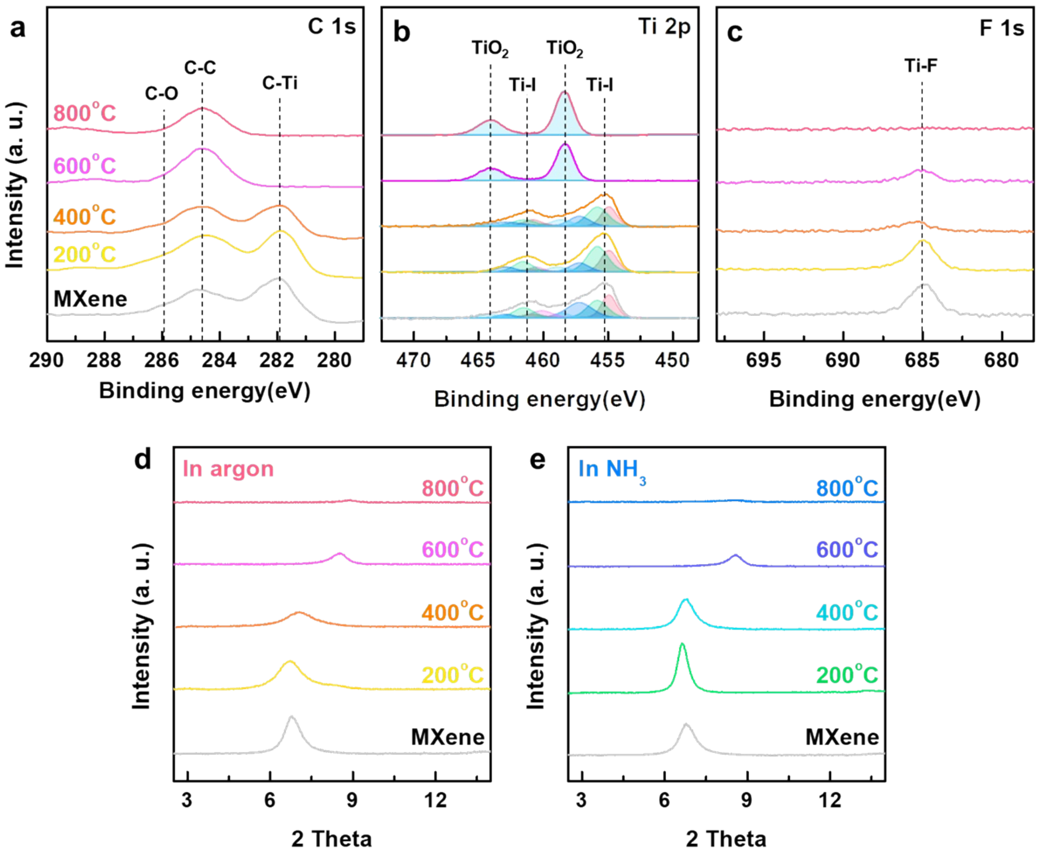

Fig. S4 XPS **a** C 1s, **b** Ti 2p, and **c** F 1s spectra of Ar-MXene. XRD patterns of MXene films prepared in **d** argon and **e** ammonia (NH_3) atmosphere.

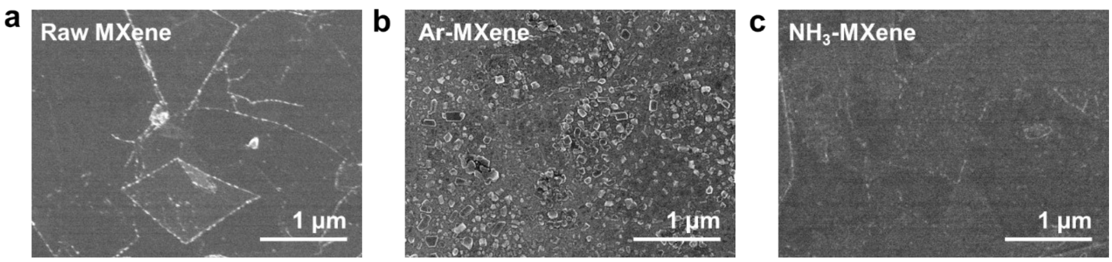

Fig. S5 SEM image of **a** raw MXene film and MXene film annealed at 600 °C in **b** argon and **c** ammonia (NH_3) atmosphere.

Table S1 Summary of the samples compared in Figure 4b.

	Electrical conductivity (S/cm)	*Note*	*Reference*
R1	2402.3	As-prepared MXene	Ling, Z.; Ren, C. E.; Zhao, M.-Q.; Yang, J.; Giammarco, J. M.; Qiu, J.; Barsoum, M. W.; Gogotsi, Y., Flexible and conductive MXene films and nanocomposites with high capacitance. Proc. Natl. Acad. Sci. 2014, 111, 16676-16681.
R2	2656	As-prepared MXene	Liu, Z.; Zhang, Y.; Zhang, H.-B.; Dai, Y.; Liu, J.; Li, X.; Yu, Z.-Z., Electrically conductive aluminum ion-reinforced MXene films for efficient electromagnetic interference shielding. J. Mater. Chem. C 2020, 8, 1673-1678.
R3	4858.81	Thermal treated MXene	Zhao, X.; Holta, D. E.; Tan, Z.; Oh, J.-H.; Echols, I. J.; Anas, M.; Cao, H.; Lutkenhaus, J. L.; Radovic, M.; Green, M. J., Annealed $Ti_3C_2T_z$ MXene Films for Oxidation-Resistant Functional Coatings. ACS Appl. Nano Mater. 2020, 3, 10578-10585.
R4	7713	As-prepared MXene	Eom, W.; Shin, H.; Ambade, R. B.; Lee, S. H.; Lee, K. H.; Kang, D. J.; Han, T. H., Large-scale wet-spinning of highly electroconductive MXene fibers. Nat. Commun. 2020, 11, 1-7.
R5	7750	As-prepared MXene	Zhang, J.; Uzun, S.; Seyedin, S.; Lynch, P. A.; Akuzum, B.; Wang, Z.; Qin, S.; Alhabeb, M.; Shuck, C. E.; Lei, W., E. C. Kumar, W. Yang, G. Dion, J. M. Razal, Y. Gogotsi, Additive-free MXene liquid crystals and fibers. ACS Cent. Sci. 2020, 6, 254-265.
R6	8259.51	As-prepared MXene	Shin, H.; Eom, W.; Lee, K. H.; Jeong, W.; Kang, D. J.; Han, T. H., Highly Electroconductive and Mechanically Strong $Ti_3C_2T_x$ MXene Fibers Using a Deformable MXene Gel. ACS nano 2021, 15, 3320-3329.
R7	8333.94	Thermally annealed MXene	Zhao, X.; Holta, D. E.; Tan, Z.; Oh, J.-H.; Echols, I. J.; Anas, M.; Cao, H.; Lutkenhaus, J. L.; Radovic, M.; Green, M. J., Annealed Ti3C2Tz MXene Films for Oxidation-Resistant Functional Coatings. ACS Appl. Nano Mater. 2020, 3, 10578-10585.
R8	8900	As-prepared MXene	Tang, X.; Murali, G.; Lee, H.; Park, S.; Lee, S.; Oh, S. M.; Lee, J.; Ko, T. Y.; Koo, C. M.; Jeong, Y. J., Engineering Aggregation-Resistant MXene Nanosheets As Highly Conductive and Stable Inks for All-Printed Electronics. Adv. Funct. Mater. 2021, 2010897.
R9	9629.03	As-prepared MXene	Shin, H.; Eom, W.; Lee, K. H.; Jeong, W.; Kang, D. J.; Han, T. H., Highly Electroconductive and Mechanically Strong $Ti_3C_2T_x$ MXene Fibers Using a Deformable MXene Gel. ACS nano 2021, 15, 3320-3329.
R10	10400	A- prepared MXene	Chen, H.; Wen, Y.; Qi, Y.; Zhao, Q.; Qu, L.; Li, C., Pristine titanium carbide MXene films with environmentally stable conductivity and superior mechanical strength. Adv. Funct. Mater. 2020, 30, 1906996.
R11	11000	Thermally annealed MXene	Lipatov, A.; Goad, A.; Loes, M. J.; Vorobeva, N. S.; Abourahma, J.; Gogotsi, Y.; Sinitskii, A., High electrical conductivity and breakdown current density of individual monolayer Ti3C2Tx MXene flakes. Matter 2021, 4, 1413-1427.
R12	12300	As-prepared MXene	Zhang, J.; Kong, N.; Uzun, S.; Levitt, A.; Seyedin, S.; Lynch, P. A.; Qin, S.; Han, M.; Yang, W.; Liu, J., Scalable Manufacturing of Free-Standing, Strong Ti3C2Tx MXene Films with Outstanding Conductivity. Adv. Mater. 2020, 32, 2001093.
R13	12484.79	Thermally annealed MXene	Zhao, X.; Holta, D. E.; Tan, Z.; Oh, J.-H.; Echols, I. J.; Anas, M.; Cao, H.; Lutkenhaus, J. L.; Radovic, M.; Green, M. J., Annealed Ti3C2Tz MXene Films for Oxidation-Resistant Functional Coatings. ACS Appl. Nano Mater. 2020, 3, 10578-10585.
R14	12503.78	As-prepared MXene	Shin, H.; Eom, W.; Lee, K. H.; Jeong, W.; Kang, D. J.; Han, T. H., Highly Electroconductive and Mechanically Strong $Ti_3C_2T_x$ MXene Fibers Using a Deformable MXene Gel. ACS nano 2021, 15, 3320-3329.
R15	15100	Thermally annealed MXene	Zhang, J.; Kong, N.; Uzun, S.; Levitt, A.; Seyedin, S.; Lynch, P. A.; Qin, S.; Han, M.; Yang, W.; Liu, J., Scalable Manufacturing of Free-Standing, Strong $Ti_3C_2T_x$ MXene Films with Outstanding Conductivity. Adv. Mater. 2020, 32, 2001093.
This work	17834	Nitrided MXene	-

Experimental section

Materials. Layered ternary carbide (Ti_3AlC_2) MAX-phase powders (particle size < 200 μm, Ukraine) were purchased from Carbon-Ukraine Ltd. The chemicals, including hydrochloric acid (HCl), lithium fluoride (LiF), and hydrogen peroxide (H_2O_2) were purchased from Sigma-Aldrich Co. (USA). Deionized water was obtained using a water-purification system (Direct Q3) purchased from Millipore (Bedford, MA, USA).

Synthesis of the $Ti_3C_2T_x$ MXene. $Ti_3C_2T_x$ MXene was obtained from the Ti_3AlC_2 precursor by modifying a previously reported method.[25] First, 2 g of LiF was dissolved in 40 mL of 9 M HCl solution in a reactor. The solution was then stirred for 30 min at 35 °C. After slowly adding 2 g of Ti_3AlC_2 (MAX phase) powder, the mixture was stirred under an argon atmosphere for 24 h. The Al layer in the MAX phase was etched to exfoliate $Ti_3C_2T_x$. Then, 40 mL of the obtained solution was divided into 20 mL conical tubes and diluted with water (20 mL). To separate the MXene dispersion from the acid, the dispersion was washed with water using a centrifuge until the pH reached 6. As the pH of the solution was almost neutral, MXene in the dispersion did not sink well during centrifugation because of an increase in the negative zeta potential of the surface. The washed solution (pH 6) was centrifuged repeatedly to purify the MXene sheets and concentrate the MXene dispersion. The obtained solution was sealed with Parafilm and stored at approximately 5 °C.

Oxidation stability test of MXene film

MXene films were prepared by Vacuum filtration method. Both ends of the MXene film were connected to the conductive metal block, the gap between the electrodes was 10 mm and the width of the MXene film was 3 mm. 200 uL of 30% H_2O_2 was dropped in the middle of MXene film and the resistance change was measured over time.

Characterization

The morphologies of the MXene sheets were characterized using SEM (S4800, Hitachi, Japan) at 15 kV and 10 μA, without Pt sputtering. The MXene samples were examined using high-resolution transmission electron microscopy (HR-TEM, JEM-2100 F, JEOL), and elemental mapping was conducted using energy-dispersive X-ray spectroscopy. The MXene samples were sonicated in cold water, and the obtained flakes were coated on a TEM grid (Holey Silicon Nitride Support Film, 2500 nm Single pores, PELCO). The chemical state of MXene was analyzed using X-ray photoelectron spectroscopy (XPS; Theta probe, Thermo Scientific, UK) with monochromatic Al Kα radiation. The XPS spectra were analyzed using Avantage software. The electrical conductivity of the MXene films was measured using a multimeter (DMM 7510 1/2, Keithley Instruments, USA) via the four-point probe method. The four electrodes were separated by 1 mm. Measurements were performed in a dry chamber.

Safety implications of using ammonia

NH_3 is a colorless gas with a strong pungent smell, as well as it can irritate or burn human skin and even kill a researcher as the reviewer pointed out. According to NIOSH (National Institute for Occupational Safety and Health), 300 ppm of ammonia gas corresponds to IDLH (Immediately dangerous to life and health) and is the concentration that causes an immediate danger to life or health. Thus, a gas scrubber, gas supply system, and vent system are required to control the gas safely. We used the gas scrubber and gas supply system (SKY-D400 and EGC-M-02, EG solution) to prevent safety accidents. Also, surplus gas was disposed of by dissolving it in water through a vent system installed in tube furnace equipment.