Supplementary Information (SI) for ChemComm.

Supporting Information

Wearable and Stretchable Electronics Enabled by MXene-PVA Films via Layer-by-Layer Assembly with Integrated UV, Thermal, and Mechanical Sensing

Ji Lin,[a] Chia-Wei Chang,[a] Chun-Ting Chang,[a] Kesavan Manibalan,[a] Tsung-Hung Tsai,[a] Yu-Chun Lin,[a] Yen-Shen Hsu,[a] Ming-Hsuan Chang[a] and Jiun-Tai Chen[a,b]*

[a]Department of Applied Chemistry, National Yang Ming Chiao Tung University, 300093 Hsinchu, Taiwan

[b]Center for Emergent Functional Matter Science, National Yang Ming Chiao Tung University, 300093 Hsinchu, Taiwan

*To whom correspondence should be addressed. E-mail: jtchen@nycu.edu.tw.

EXPERIMENTAL SECTION

Materials

The MAX phase (Ti_3AlC_2) was supplied by Sigma-Aldrich. Lithium fluoride (98.5%) was purchased from Nova Materials. Polyvinyl alcohol (PVA, Mw = 130000 g/mol) was supplied by Sigma-Aldrich.

Fabrication of the $Ti_3C_2T_x$ MXene Single Layer Suspensions

$Ti_3C_2T_x$ MXene was synthesized using the minimally intensive layer delamination (MILD) method. Initially, 1.6 g of lithium fluoride (LiF) was dissolved in 20 mL of 9 M hydrochloric acid (HCl) in a polypropylene (PP) beaker and stirred for 1 h at room temperature. Subsequently, 1 g of MAX phase (Ti_3AlC_2) was slowly added to the solution and the mixture was stirred for 24 h at 35 °C to facilitate the selective etching of the aluminum (Al) layer. After the etching process, the reaction mixture was divided into two centrifuge tubes containing 20 mL of deionized (DI) water. The mixture was then washed via repeated centrifugation until the supernatant reached a pH value between 6 and 7. After purification, the obtained MXene sediment was redispersed in 28.5 mL of DI water with 1.5 mL of dimethyl sulfoxide (DMSO) to perform exfoliation. The suspension was subjected to ultrasonication in a bath sonicator for 1 h to achieve layer separation. Finally, the dispersion was centrifuged at 5000 rpm for 15 min and the supernatant containing the delaminated $Ti_3C_2T_x$ MXene nanosheets was collected.

Fabrication of the MX-PVA Films

MX-PVA films were prepared using a layer-by-layer assembly process. First, a 15 wt % polyvinyl alcohol (PVA) aqueous solution was spin-coated onto a 2.5 × 2.5 cm glass substrate. The spin-coating process was performed in two sequential steps: 300 rpm for 30 s followed by 1000 rpm for 30 s. The coated substrate was then dried on a hot plate at 50 °C to ensure complete evaporation of the solvent. Once the

PVA layer was thoroughly dried, a $Ti_3C_2T_x$ MXene suspension was drop-casted onto the surface and then dried on a hot plate at 35 °C. After the MXene layer was completely dried, the substrate was removed from the hot plate and cooled to room temperature under ambient conditions. This process was repeated to achieve a four-layer structure consisting of alternating PVA and MXene layers (glass substrate–PVA–MXene–PVA–MXene). Finally, an additional PVA layer was applied using the same procedure to complete the fabrication of the MX-PVA film.

Electrical Property Measurements

An LCR meter (GW Instek, LCR-6300) was applied to evaluate the electrical properties of the MX-PVA films. The responsive electrical behaviors of the MX-PVA films were assessed in the C_p-R_p mode at 1 V and 1 kHz frequency.

Structure Analysis and Characterization

Mechanical properties were evaluated using a Shimadzu EZ tensile testing machine, operated at a strain rate of 5 mm min^{-1} for stretching and relaxation tests. X-ray fluorescence (XRF) experiments were performed at TPS 23A, the X-ray Nanoprobe Beamline, at the National Synchrotron Radiation Research Center (NSRRC) in Hsinchu, Taiwan. The elemental distributions of the MX-PVA films were examined with an energy-dispersive X-ray spectrometer (EDS, Oxford EDS 7585). High-resolution X-ray photoelectron spectroscopy (XPS) was conducted using a PHI Quantera II spectrometer (ULVAC-PHI) equipped with a scanning X-ray microprobe (Al anode), a 180° hemispherical analyzer, and a 128-channel detector. XPS spectra were processed and interpreted using CasaXPS software (Version 2.3.26). Ultraviolet-visible (UV-vis) spectra were acquired with a Hitachi U4100 spectrometer to analyze the

MXene suspension samples, scanning wavelengths from 350 to 1000 nm at a rate of 300 nm min^{-1}. The plots were processed using OriginPro 2025 (10.2.0.196)

SUPPORTING DATA

Preparation Process and Characteristics of MXene

Single-layer $Ti_3C_2T_x$ MXene is prepared using the minimally intensive layer delamination (MILD) method. The detailed experimental procedure is depicted in Figure S1a. The final product, a stable dispersion of single-layer $Ti_3C_2T_x$ MXene in deionized (DI) water, is illustrated in Figure S1b. The layered structure of the multilayer MXene is confirmed via scanning electron microscopy (SEM), as presented in Figure S1c, where the layers are indicated by two red arrows. Transmission electron microscopy (TEM) image (Figure S1d) further confirms the successful exfoliation into single-layer MXene, with the observed sheet dimensions consistent with dynamic light scattering (DLS) measurements (Figure S2). Additionally, X-ray diffraction (XRD) analysis (Figure S1e) demonstrates a shift in the (002) peak from 9.59 (MAX phase) to 5.96° (single-layer MXene), corresponding to an increase in interlayer spacing from 4.62 to 7.42 Å, confirming successful delamination. The UV-vis absorption of the MXene single-layer suspension in different concentrations is also tested (Figure S3).

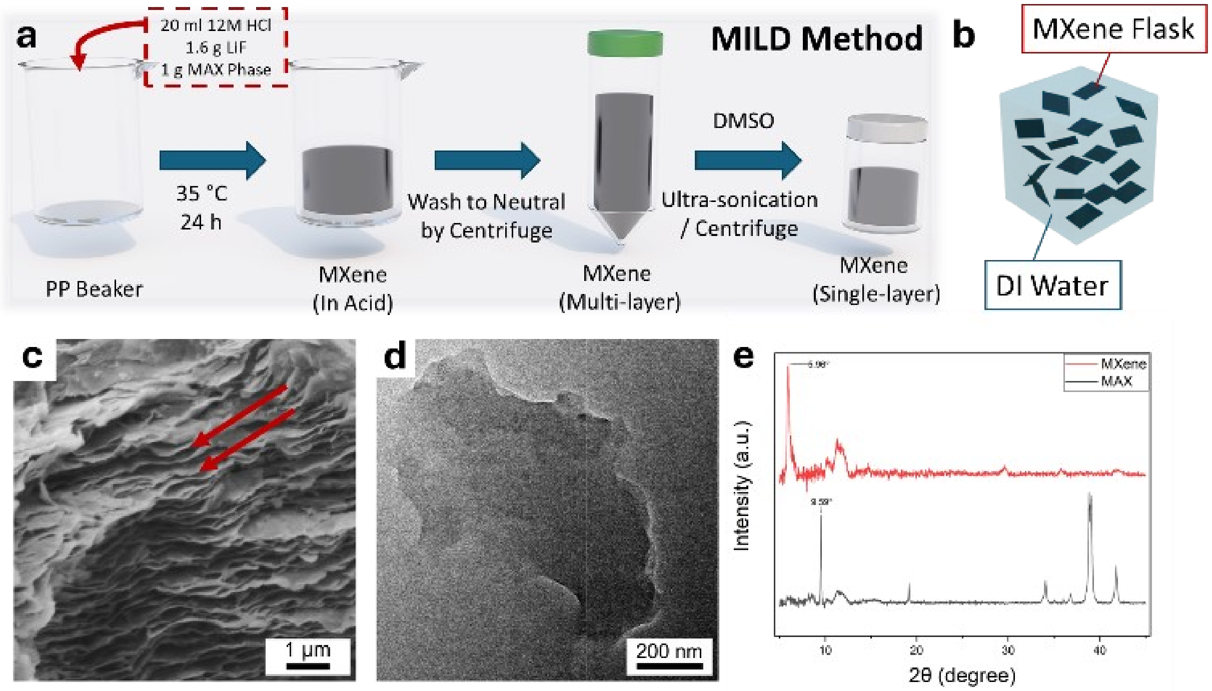

Fig. S1 (a) Schematic illustration of the MILD process to fabricate MXene. (b) Schematic illustration of MXene single layers suspended in DI water. (c) SEM image of the multi-layered MXene. (d) TEM image of the MXene single-layer nanosheet. (e) XRD spectra of the MXene and the MAX phase.

DLS Volume Distribution

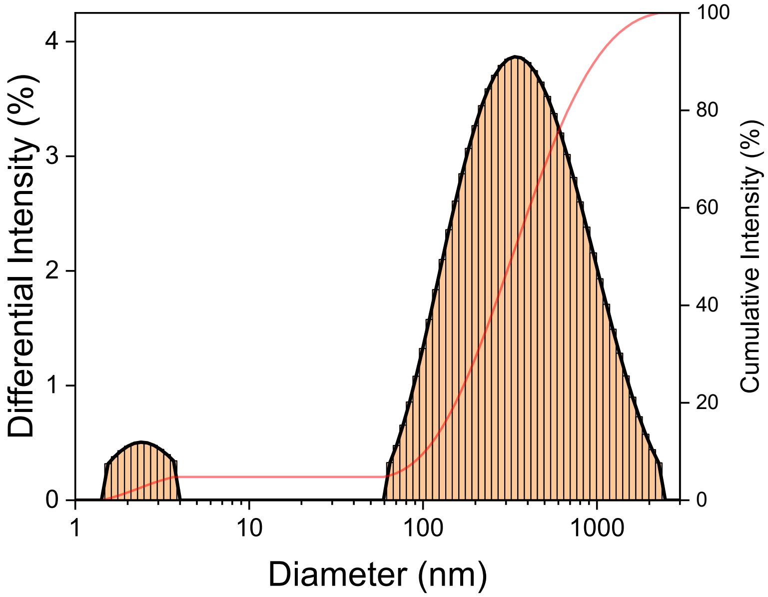

Fig. S2 DLS volume distribution of the MXene single-layer nanosheet suspension in water.

UV-vis Absorption

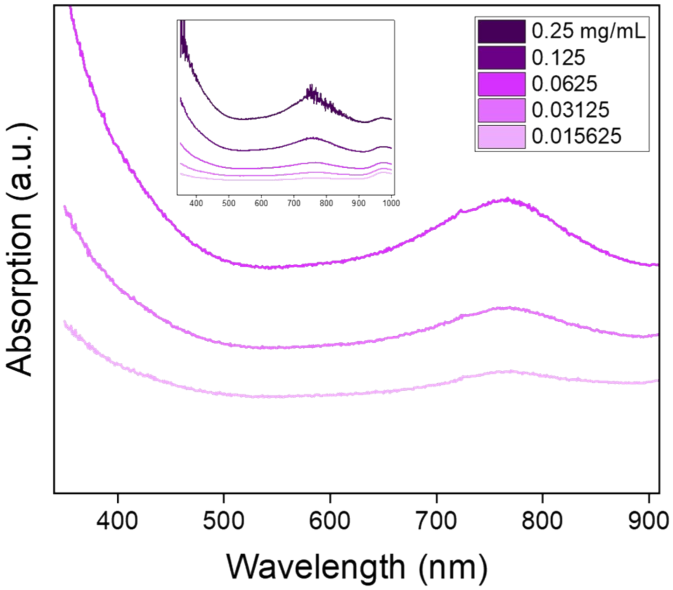

Fig. S3 UV-vis absorption spectra of the MXene single-layer suspension in 0.25, 0.125, 0.0625, 0.03125, and 0.015625 mg/mL.

Surface Uniformity

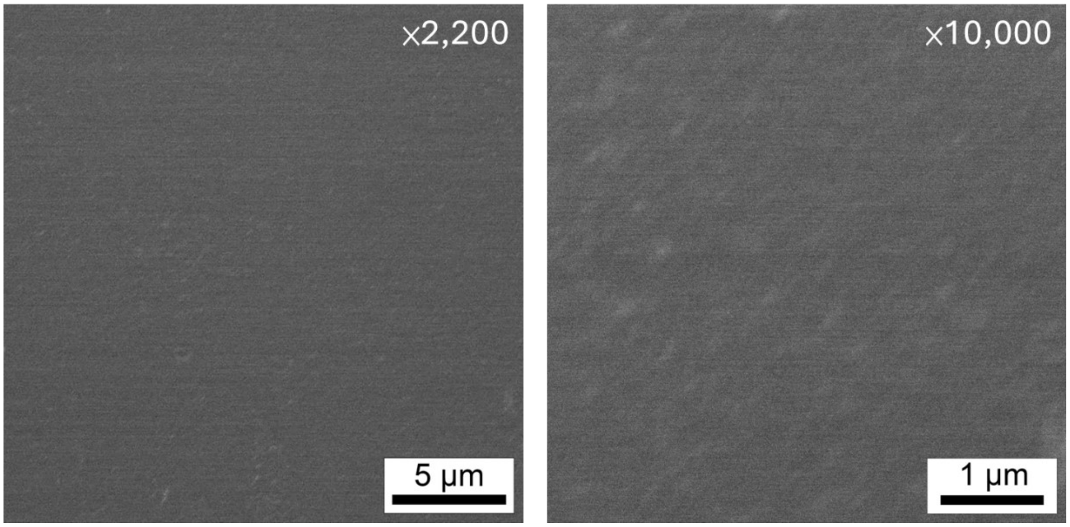

Fig. S4 SEM imaging of the MX-PVA film's surface in 2000 and 10000 times magnification.

Oxidation Test

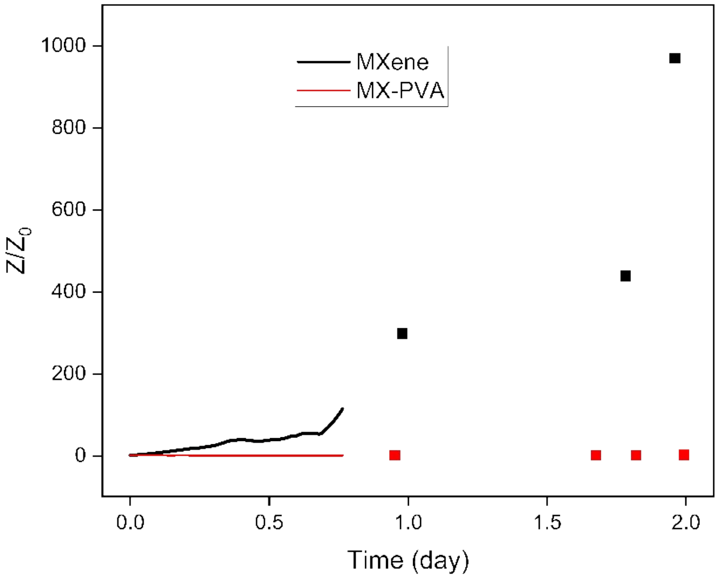

Fig. S5 Impedance change of MXene and MX-PVA film after preparation.

Humidity test

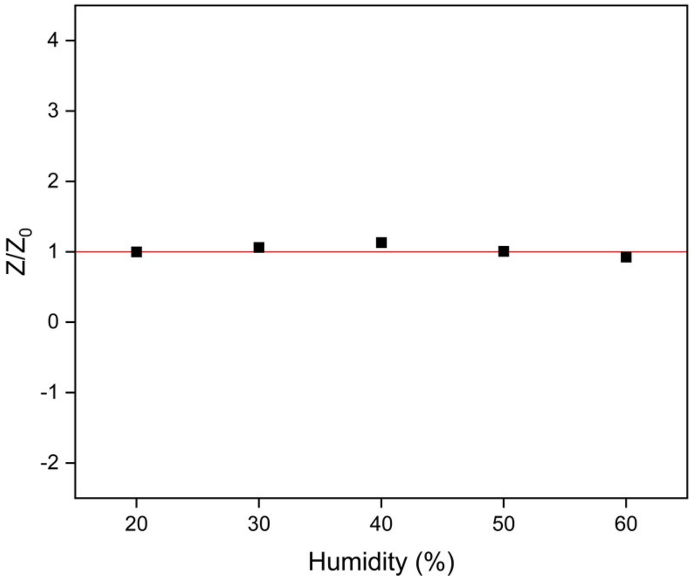

Fig. S6 Impedance of MX-PVA film under different humidity conditions.

XPS Element Ratio

Atomic%	MXene	MX-PVA
C 1s	37.85	65.00
N 1s	0.61	0.00
O 1s	30.89	34.29
F 1s	18.50	0.71
Ti 2p	12.15	0.00
Total	100	100

Table S1 Table of element ratios of MXene and MX-PVA films.

100 Cycles Durability Test

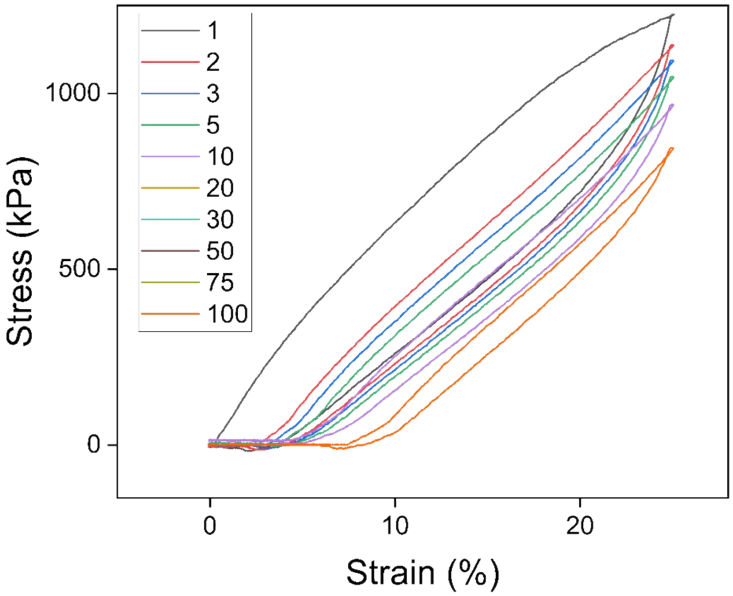

Fig. S7 Stress-strain curves of an MX-PVA film under 100 cycles.

Press Sensing Test

The pressure-sensing capability of the MX-PVA film is evaluated using a controlled press test, as illustrated in Figure S3a. MX-PVA film is placed between two glass substrates, and varying masses are applied to the upper glass substrate to simulate different levels of downward force. The impedance response of the MX-PVA film is measured using an LCR meter with copper tape electrodes attached at both ends. In Figure S3b, the applied weights ranged from 10 to 1000 g, corresponding to forces up to approximately 10 N. The results indicate a strong linear correlation between impedance and applied force for forces below 5 N. However, when the applied force exceeds 5 N, the impedance exhibits minimal change, suggesting a saturation effect in the material's pressure response. Furthermore, the repeatability of MX-PVA film's pressure-sensing behavior is assessed through cyclic tests. A 100 g weight is repeatedly placed and removed from the upper glass substrate in 30 s intervals to simulate a consistent pressing motion. As shown in Figure S3c, each cycle demonstrates a consistent impedance decrease upon force application, followed by a well-defined recovery after force release. These findings highlight the potential of the MX-PVA film for applications in pressure-sensitive devices with reliable and repeatable performance.

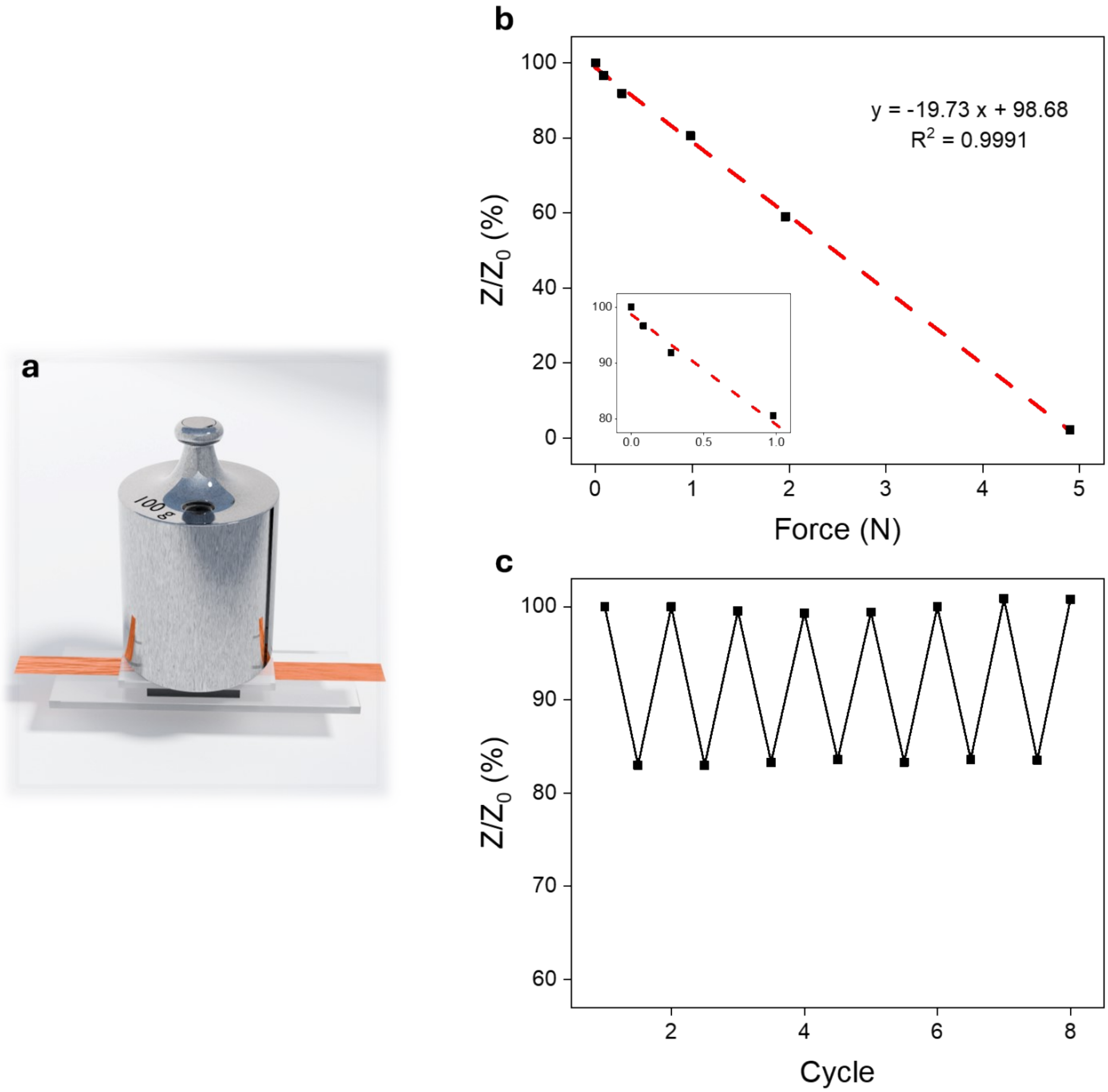

Fig. S8 (a) Schematic illustration of measuring the pressure-responsive properties of the MX-PVA films. (b) Impedances of an MX-PVA film under various pressures. (c) Impedance changes of an MX-PVA film under press-release cycles (1 N versus no pressure).