Supporting information

Environmentally stable MXene ink for direct writing flexible electronics

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1 Experimental detail

1.1 Synthesis of self-delaminated $Ti_3C_2T_x$

All chemicals were used as received without further purification. Layered ternary carbide Ti_3AlC_2 (MAX phase) powder were commercially procured (Carbon-Ukraine ltd. particle size < 40 μm). $Ti_3C_2T_x$ (MXene) was synthesized following minimal intensive layer delamination method by selective etching of aluminium from Ti_3AlC_2. The etching solution was prepared by adding 1.6 g lithium fluoride (LiF, 99 %, Sigma-Aldrich Pty Ltd) to 20 ml 9 M hydrochloric acid (HCl), followed by stirring for 5 min. 1 g of Ti_3AlC_2 powder was slowly added to the etchant at 35 °C and stirred for 24 h. The mixture was washed using centrifugation method on Centrifuge-5810R (Eppendorf AG, Germany). The acidic suspension was washed with deionized (DI) water until pH ~ 6 via centrifugation at 3500 rpm and decanting the supernatant after each cycle. When pH ≥ 6, stable dark green supernatant of self-delaminated $Ti_3C_2T_x$ was observed by hand shaking or revolving mixing for 10 minutes. Afterwards, the dark dispersion contained single layer MXene, multilayer MXene and unreacted MAX phase was centrifuged at 1500 rpm (30 mins) to separate single layer $Ti_3C_2T_x$ from multi-layer MXene and unreacted MAX phase. The supernatant was collected and then concentrated at 8000 rpm for 20 mins, which has a concentration of 42 mg mL^{-1}.

1.2 The environmental stability of MXene inks

The self-delaminated $Ti_3C_2T_x$ dispersion was added into mixtures of water and organic solvents, such as DMSO, EG, 1,2-propanediol, and 1,3-propanediol, and then stored in air and room temperature to observe the color changes of dispersions. For example, 0.2 mL of MXene aqueous dispersion (5 mg mL^{-1}) was diluted with 0.8 mL EG and 1 mL DI water to achieve 2 mL of 0.5 mg mL^{-1}MXene ink. All samples have the same volume of 2 mL and concentration of 0.5 mg mL^{-1}. Then, the color change was recorded in Day 1, Day 14, Day 20 and Day 35.

1.3 Synthesis of MXene ink in EG and water

The MXene ink in composite solvents of water and EG was synthesized by diluting the as-synthesized MXene (40 mg mL^{-1}) and made the final concertation to 10 mg mL^{-1} in the mixed solvent. The MXene inks were infilled into a rollerball pen (Deli CO., LTD, China) with ball diameter of 0.5 mm, and all the patterns were written on the Reflex Ultra A4 paper.

1.4 Characterizations

A field emission scanning electron microscope (SEM) (Zeiss SUPRA 55-VP) was used to study the film morphology and the cross-section of the MXene films. MXene film samples were broken in liquid nitrogen. The X-ray diffraction (XRD) patterns of the MAX phase and various MXene films were obtained using a powder diffractometer (PANalytical X'Pert Powder) equipped with a Cu Kα radiation (40 kV, 30 mA) with an X-ray wavelength (λ) of 1.54 Å at a 2θ scan step of 0.013°. Atomic force microscopy (AFM) images were obtained under Bruker's proprietary ScanAsyst atomic force microscopy scan mode (Bruker MultiMode 8-HR) to measure the flake thickness. AFM samples were prepared by drop casting MXene solution on silicon wafers. The rheological properties of MXene dispersions were measured using a rheometer (TA Instruments AR-G2) with a cone-shaped geometry (angle: 2°, diameter: 40 mm). The dynamic light scattering (DLS) analysis of the MXene flake size was recorded using Zetasizer Nano ZS (Malvern Instruments, USA). Viscosity change as a function of shear rate was measured at shear rates between 0.01 to 1,000 s^{-1} using logarithmic steps. The viscosity of the dispersion at 0.01 s^{-1} were taken as the zero-shear rate viscosity, as measuring viscosity at lower shear rates (< 0.01 s^{-1}) can be challenging for dilute samples. The electrical resistances of MXene traces on paper were measured using a source meter unit (Keysight B2901A) by the aid of a custom-built four-point probe set-up. The capacitive sensing performance was measured from a capacitance (C_f) is formed between two electrodes when finger is touching the sensor pattern. The capacitance change was recorded using a digital multimeter (Keysight

34461A). All data were collected at room temperature with the humidity of .60%. The infrared imaging photographs were recorded using TiS20+MAX infrared imaging camera (FLUKE, China)

2 Supporting results

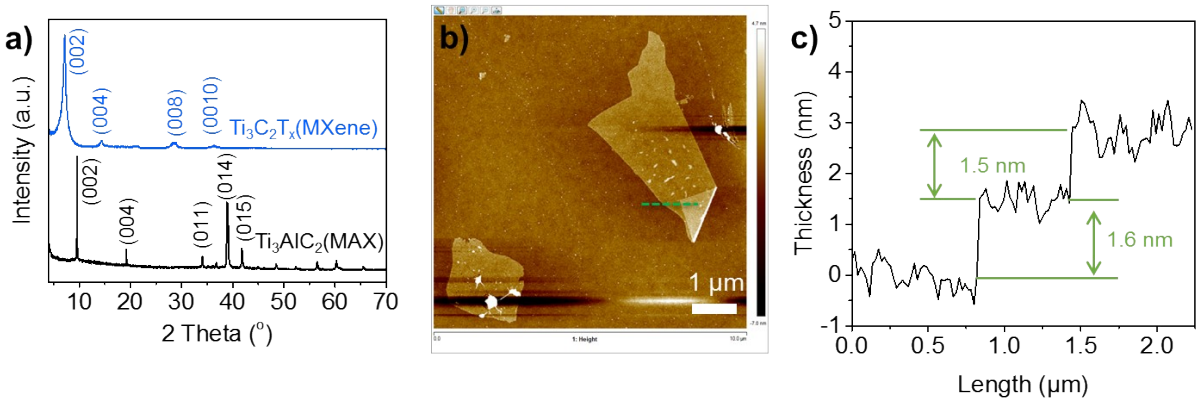

Figure S1. (a) The XRD patterns of Ti_3AlC_2 powder and vacuum filtrated $Ti_3C_2T_x$ film. (b) The AFM image of $Ti_3C_2T_x$ flakes using silicon wafer as substrate. (c) The thickness profile of MXene flakes that measured from the green dash line in AFM image.

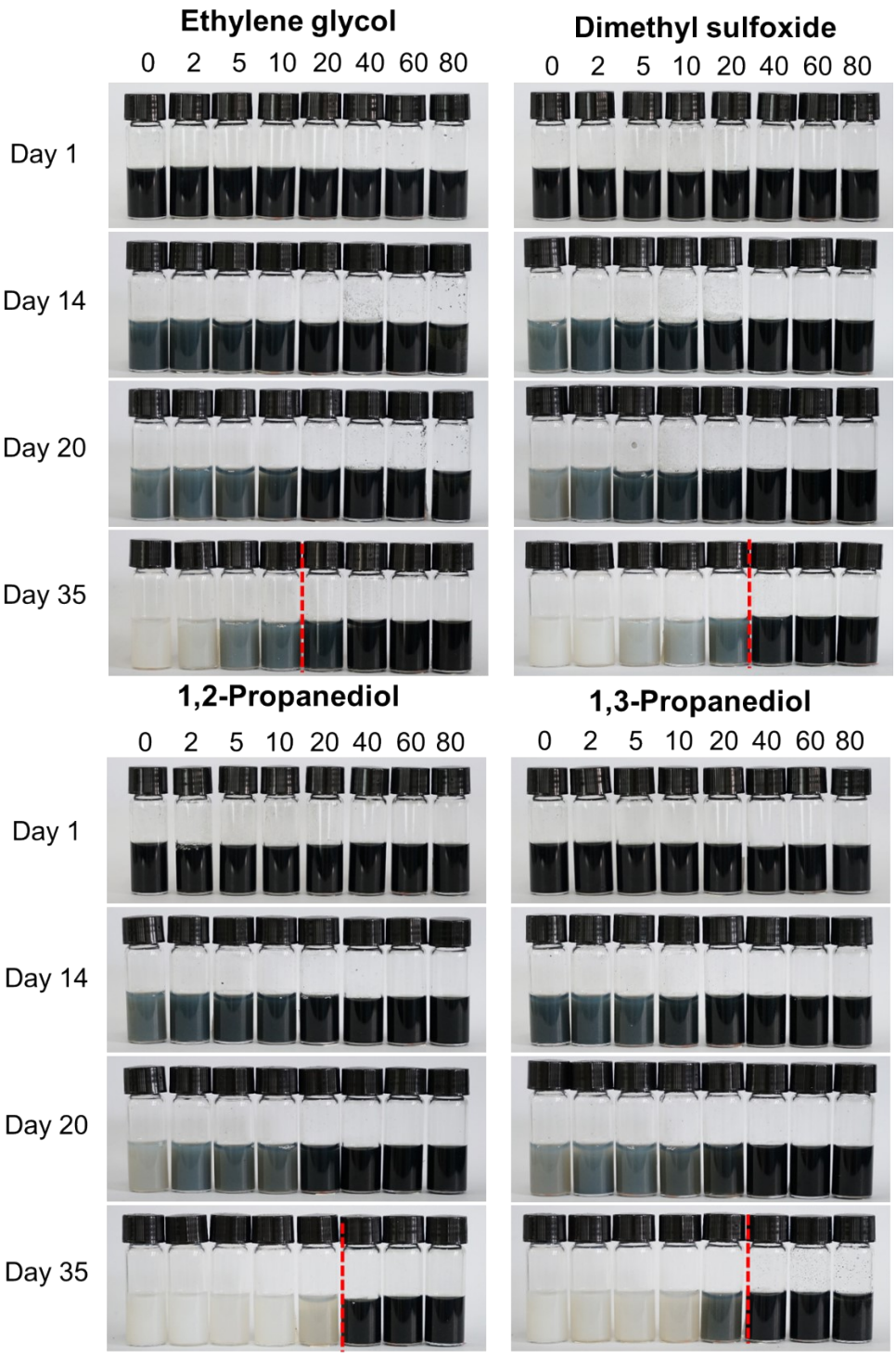

Figure S2. The color change of MXene inks those dispersed in water with different ratio of ethylene glycol, dimethyl sulfoxide, 1,2-propanediol and 1,3-propanediol.

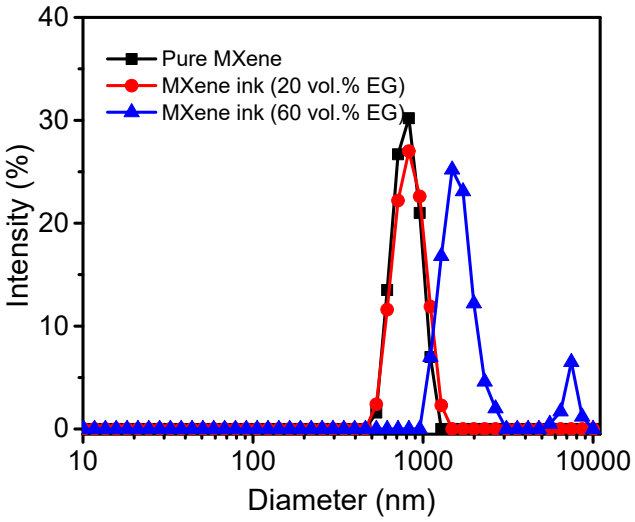

Figure S3. Dynamic light scattering (DLS) intensity distributions of MXene water dispersion, MXene in 20 vol.% and 60 vol.% of EG, respectively.

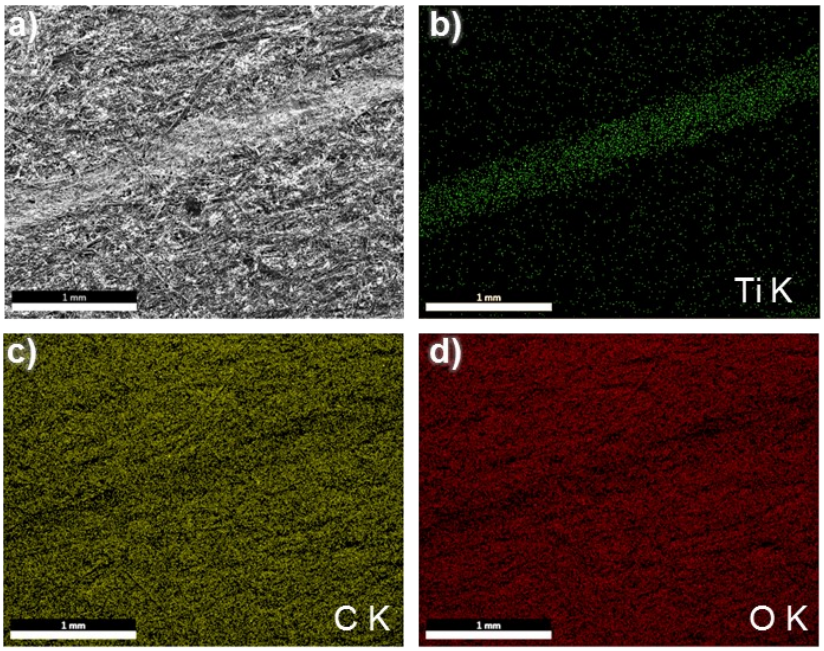

Figure S4. (a) SEM image of MXene trace that draw on paper. (b-d) Energy dispersive x-ray mapping of the selected area in SEM image with titanium, carbon and oxygen elements, respectively.