Supporting Information

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SI Materials and Methods

Synthesis of Multilayer $Ti_3C_2T_x$ Powder. The processing details of MAX phase, Ti_3AlC_2, used in this study can be found elsewhere (7). Stacked or multilayer $Ti_3C_2T_x$ (Ml-$Ti_3C_2T_x$) powder was prepared by selective etching the Al layer from Ti_3AlC_2. Specifically, Ti_3AlC_2 powder, with a particle size less than 38 μm, was immersed, at room temperature (RT), in a 50 wt% aqueous HF solution (Fisher Scientific) with a ratio of 1 g of Ti_3AlC_2 per 10 mL of HF solution for 18 h while magnetically stirring the solution. The obtained suspension was diluted with deionized water and centrifuged to obtain the Ml-$Ti_3C_2T_x$ powders, which were then washed until the pH of the supernatant was above 6. The produced powder was dried at RT over 24 h.

Delamination of $Ti_3C_2T_x$. To obtain few- and/or single-layer flakes, the Ml-$Ti_3C_2T_x$ powders were delaminated. Dimethyl sulfoxide (DMSO) was used to enhance the delamination process (13). In this case, the Ml-$Ti_3C_2T_x$ powders were magnetically stirred in DMSO, at a ratio of 1 g of Ml-$Ti_3C_2T_x$ per 12 mL, respectively, for 18 h at RT. Then 30 mL of deionized water was added to the suspension, and the DMSO intercalated Ml-$Ti_3C_2T_x$ was separated by centrifugation at 3,500 rpm for 5 min. The obtained powder was dispersed in deaerated water with a weight ratio of Ml-$Ti_3C_2T_x$:water of 1:300. The suspension was sonicated under flowing Ar for 5 h, and then centrifuged at 3,500 rpm for 1 h to obtain the supernatant containing $Ti_3C_2T_x$ flakes, henceforth referred to as the MXene colloidal solution.

Characterization. A scanning electron microscope (SEM) (Zeiss Supra 50VP) was used to study the morphology of the produced flakes and films. Elemental analysis was conducted using an energy-dispersive X-ray spectrometer (Oxford EDS, with INCA software). A TEM (JEOL JEM-2100F) operating at 200 kV was used to obtain images of the $Ti_3C_2T_x$ MXene flakes and $Ti_3C_2T_x$/PVA films. The $Ti_3C_2T_x$ flakes for TEM were prepared by dropping the colloidal solution of MXene on a lacey carbon-coated copper grid. The $Ti_3C_2T_x$/PVA cross-sections were produced by first embedding the films in epoxy resin and then cutting them using a glass microtome. The produced chips were placed on a lacey carbon-coated copper grid.

The conductivity was measured at RT via a four-probe technique (Jandel Engineering Limited). The distance between probes was 1.0 mm. The range of voltage used was from 0 to 4 V. The contact angle was measured at RT using the sessile drop technique. A water drop with a volume of 10 μL was placed on the surface of a $Ti_3C_2T_x$ film and allowed to stabilize for 45 s before a picture was taken. The XRD patterns were recorded by a powder diffractometer (Rigaku SmartLab) with Cu K_α radiation at a step size of 0.02° and a collection time of 0.5 s per step. The ζ-potential was measured using a particle size analyzer (Zetasizer NanoZS; Malvern). The $Ti_3C_2T_x$ solution tested had a concentration of 0.3 mg/mL. The centrifugation was conducted on a HERMLE centrifuge (Z400, South of Germany) with 12 rotors at 3,500 rpm (2,547 × g).

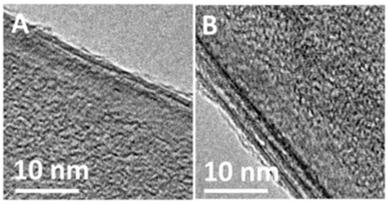


Fig. S1. High-resolution TEM images of (*A*) two- and (*B*) three-layer $Ti_3C_2T_x$ flakes.

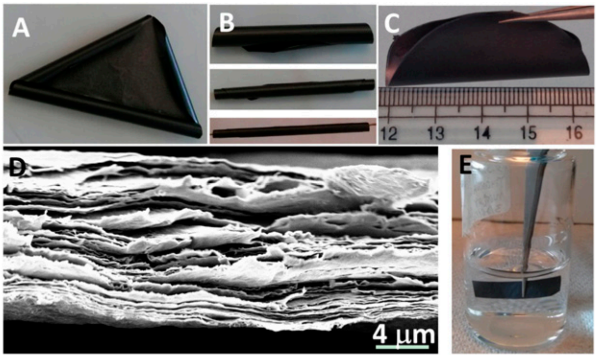


Fig. S2. (*A*) Digital image of a free-standing $Ti_3C_2T_x$ film with diameter of ∼40 mm. (*B*) Rolled films on to 10-, 5-, and 1-mm-diameter (from *Top* to *Bottom*) glass rods and copper wire. The films weight was ∼6 mg. (*C*) A 13-μm-thick $Ti_3C_2T_x$ film having a mass of 50.1 mg still shows impressive flexibility. (*D*) Cross-sectional SEM image of a 50-mg film. (*E*) Digital image of $Ti_3C_2T_x$ film after storage in deionized water for 1 month (see associated digital Movie S2).

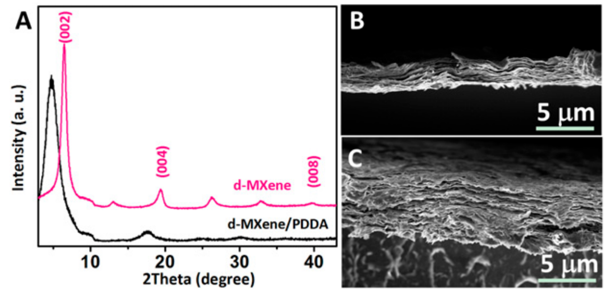


Fig. S3. (*A*) XRD patterns of $Ti_3C_2T_x$/PDDA and $Ti_3C_2T_x$ films. (*B* and *C*) Fractured, cross-sectional SEM micrographs of 2.2-μm-thick (*B*) and 5.3-μm-thick (*C*) $Ti_3C_2T_x$/PDDA films.

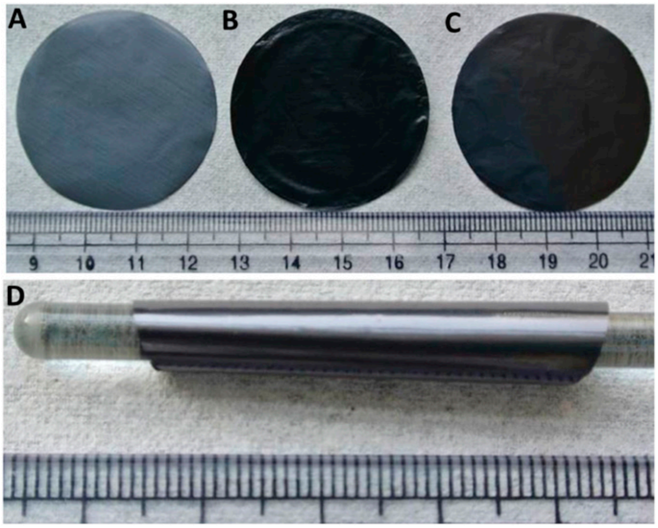


Fig. S4. Digital images of flat (*A*) $Ti_3C_2T_x$, (*B*) $Ti_3C_2T_x$/PVA, and (*C*) $Ti_3C_2T_x$/PDDA films, and (*D*) rolled shiny $Ti_3C_2T_x$ film on a glass rod (diameter of 10 mm).

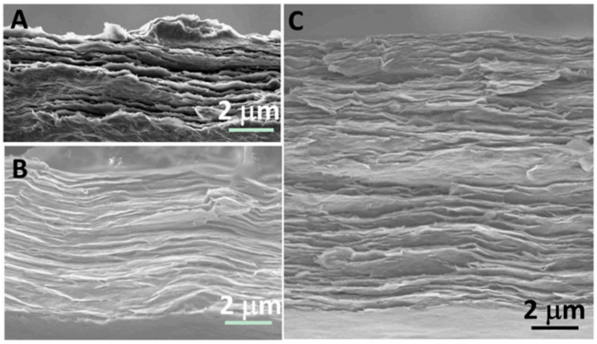


Fig. S5. Cross-sectional SEM images of (*A*) 90 wt% $Ti_3C_2T_x$/PVA, (*B*) 60 wt% $Ti_3C_2T_x$/PVA, and (*C*) 40 wt% $Ti_3C_2T_x$/PVA films.

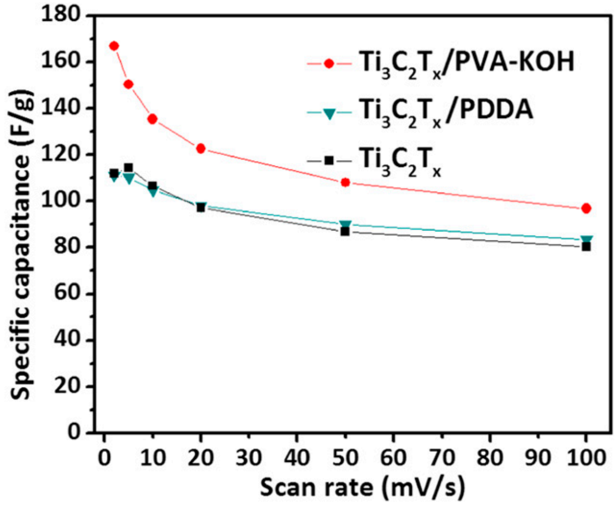
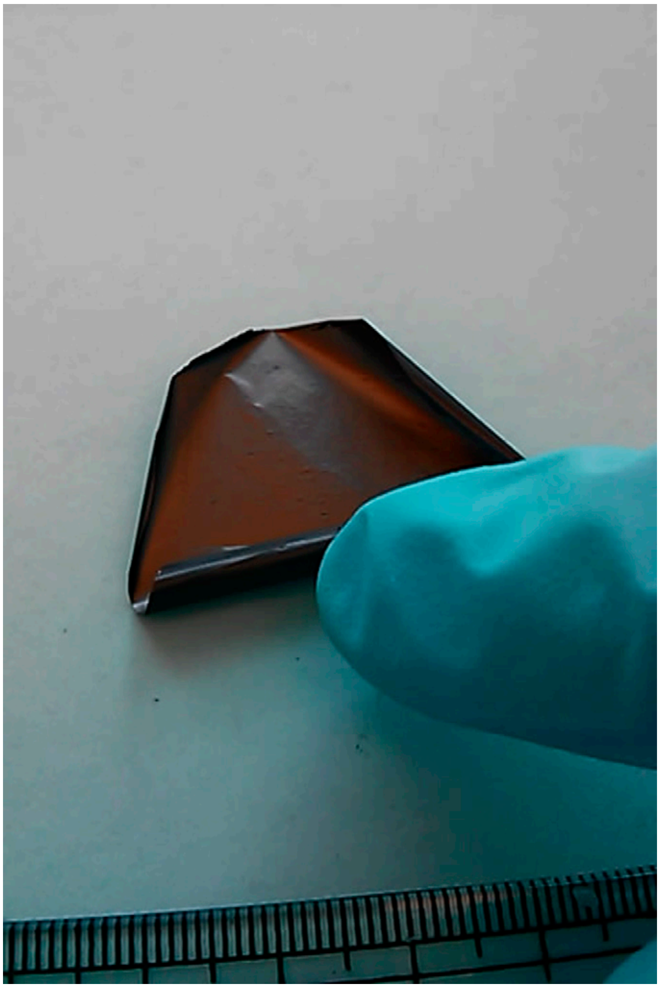


Fig. S6. Gravimetric capacitances of $Ti_3C_2T_x$, $Ti_3C_2T_x$/PDDA, and $Ti_3C_2T_x$/PVA-KOH films at different scan rates.



Movie S1. Highly flexible d-MXene film.

Movie S1

Movie S2. Flexible free-standing d-MXene film remains intact when subjected to violent shaking under water.

Movie S2