## Supporting Information

# Single-Molecule Sensing Using Nanopores in Two-Dimensional Transition Metal Carbide (MXene) Membranes

Mehrnaz Mojtabavi [1,#], Armin VahidMohammadi [2,#], Wentao Liang [3], Majid Beidaghi [2,*], Meni Wanunu [4,5,*]

[1] Department of Bioengineering, Northeastern University, Boston, MA 02115, USA.

[2] Department of Materials Engineering, Auburn University, Auburn, AL 36849, USA.

[3] Kostas Advanced Nano-Characterization Facility, Northeastern University, Burlington Campus, 141 South Bedford Street, Burlington, MA 01803

[4] Department of Physics, Northeastern University, Boston, MA 02115, USA.

[5] Department of Chemistry and Chemical Biology, Northeastern University, Boston, MA 02115, USA

[#] These authors contributed equally to the work.

* Corresponding authors: mzb0088@auburn.edu; wanunu@neu.edu

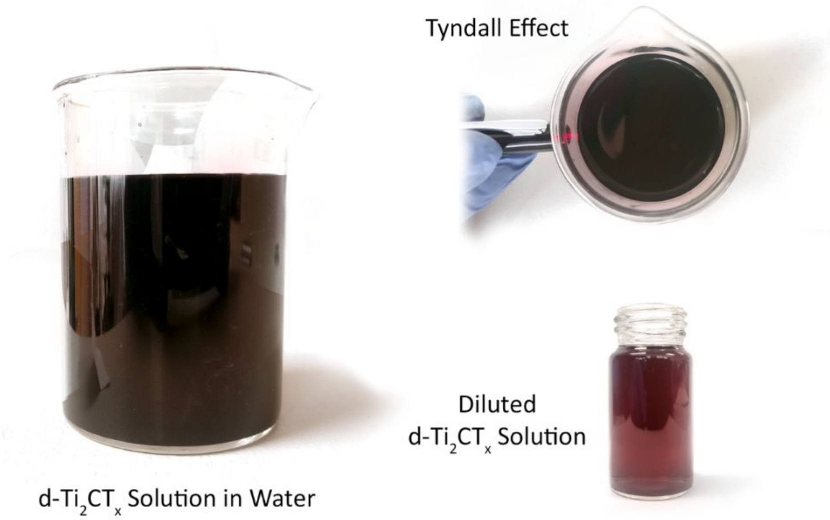



**Figure S1. Water dispersion of single/few layers delaminated $Ti_2CT_x$ (d-$Ti_2CT_x$) flakes.**

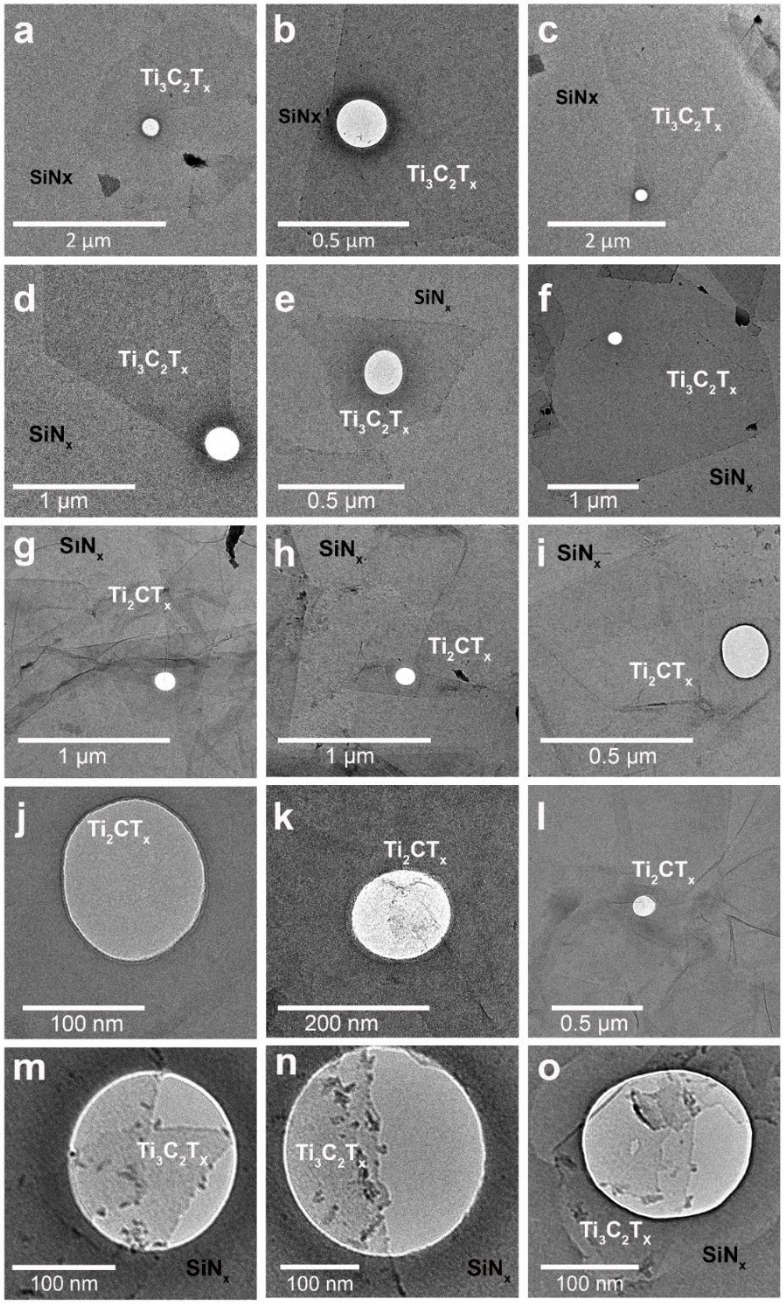



**Figure S2. Examples of transferred $Ti_3C_2T_x$ (a-f) and $Ti_2CT_x$ (g-l) flakes on $SiN_x$ membrane with pre-fabricated holes using liquid-liquid interface method**. (m-o) Unsuccessful transferred $Ti_3C_2T_x$ flakes on $SiN_x$ membrane with pre-fabricated holes using same method. Thickness variation of flakes assembly on $SiN_x$ membrane contributes to contrast in the images. Darker areas indicate thicker assembly of flakes.

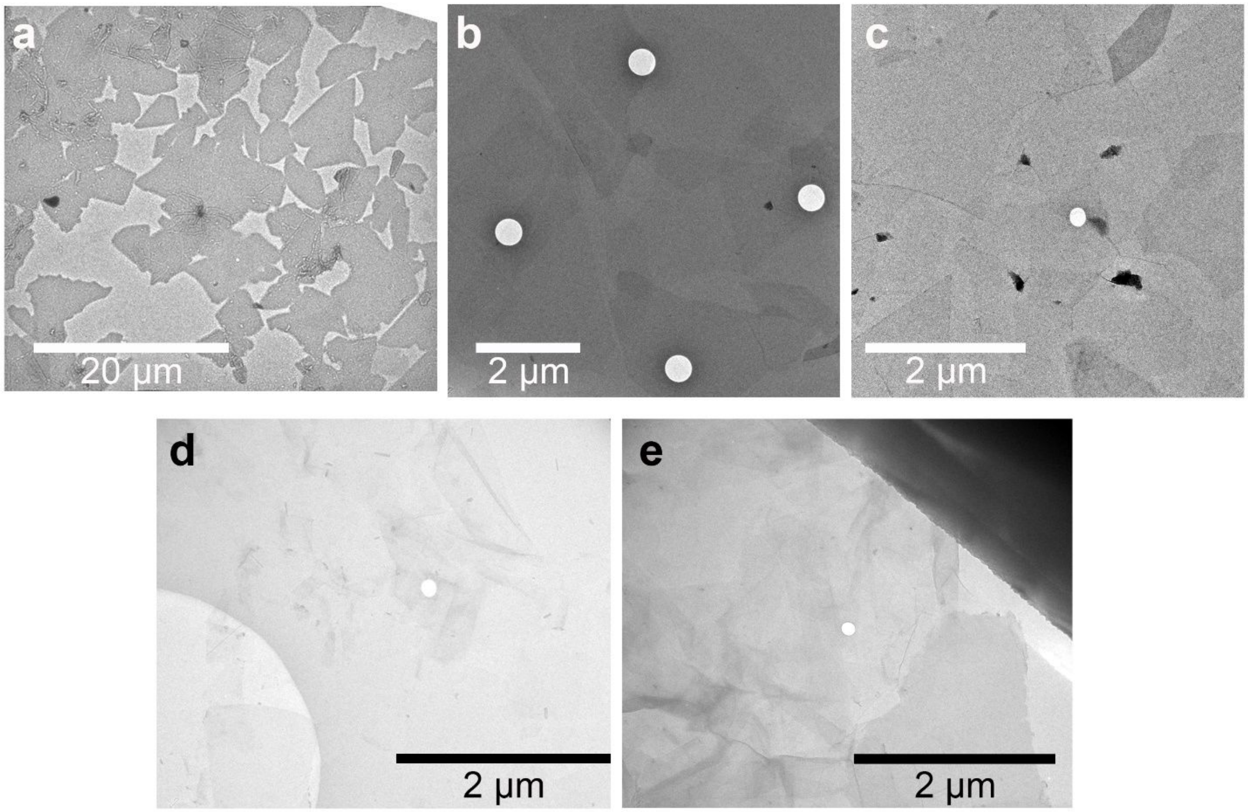



**Figure S3**. **Examples of $SiN_x$ membrane coverage by MXene flakes after transfer of flakes from water dispersion by liquid-liquid interface method** (white circles show pre-fabricated holes). (a-c) Transferred $Ti_3C_2T_x$ flakes. (d-e) Transferred $Ti_2CT_x$ flakes. The different contrasts in the images are due to different TEM magnifications used to obtain the images.

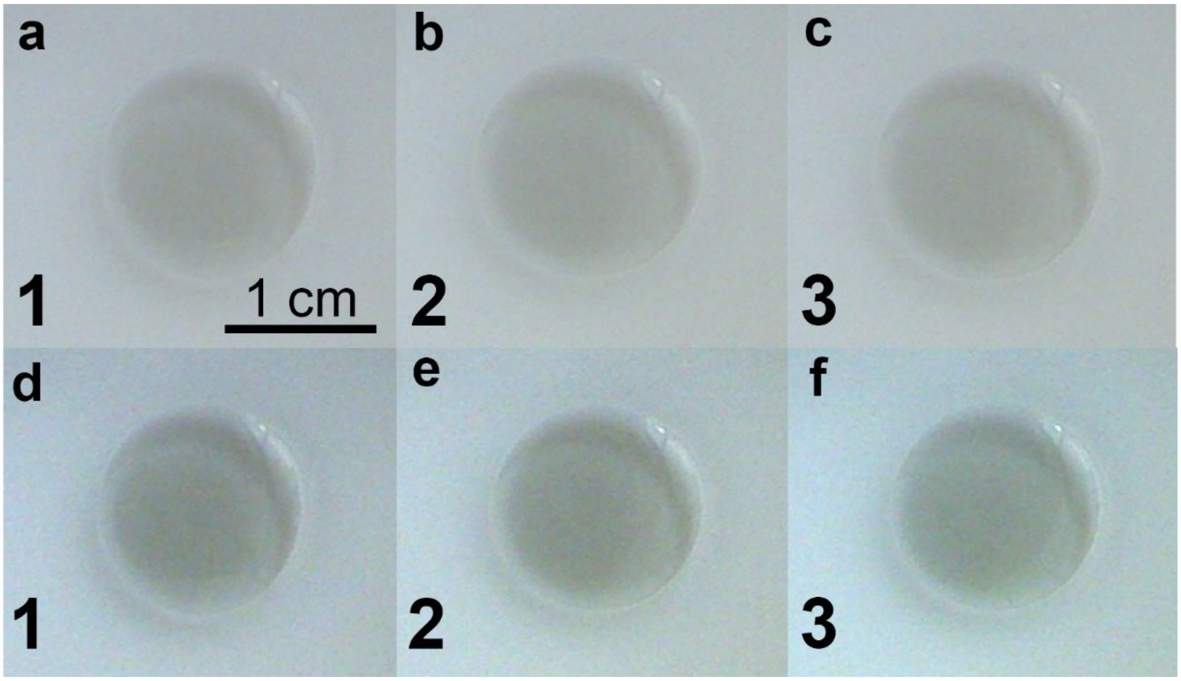



**Figure S4**. **Snapshots of flake self-assembly at liquid-liquid interface using a 5-fold diluted $Ti_3C_2T_x$ dispersion (~0.02 mg/ml).** (a-c) original images. (d-e) same images with higher contrast for visualization purposes.

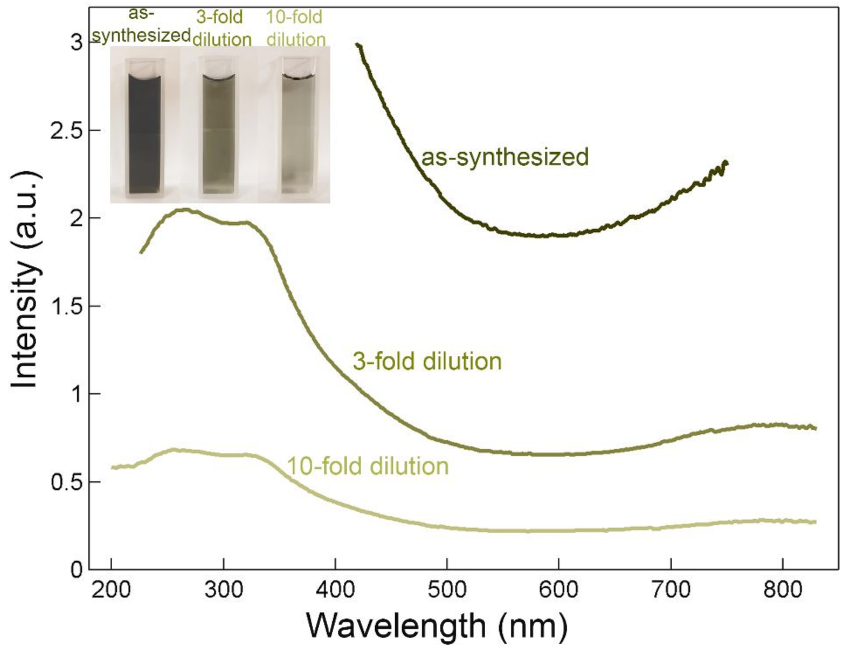



**Figure S5. UV-VIS absorbance spectra of an as-synthesized $Ti_3C_2T_x$ dispersion, a 3-fold, and a 10-fold diluted dispersions**. The inset shows photographs of the dispersions. Due to high concentration of as-synthesized dispersion, absorbance measurement was not possible in some wavelengths.

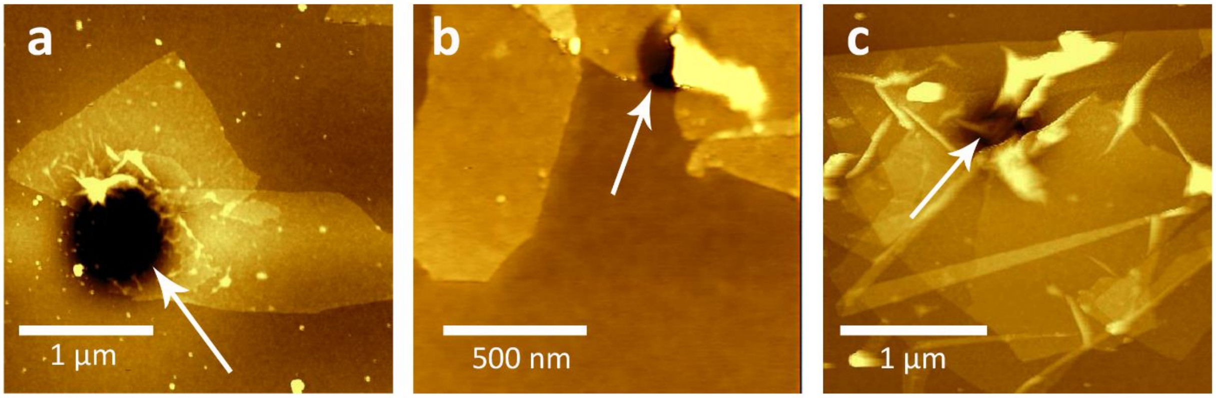



**Figure S6. $Ti_3C_2T_x$ flakes drop-casted on $SiN_x$ membranes with pre-fabricated holes.** Arrows show flakes being pulled and bent into the hole during the drying process.

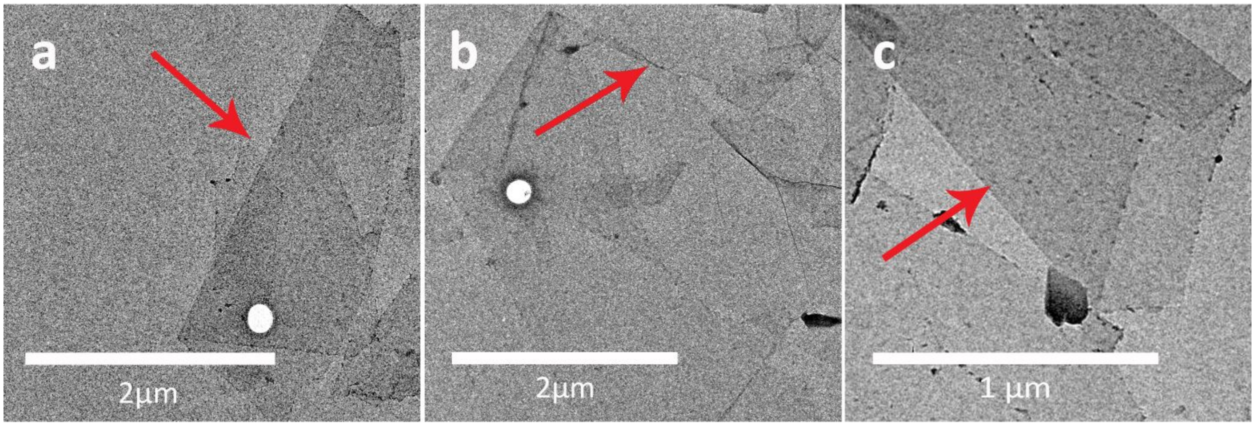



**Figure S7. $Ti_3C_2T_x$ flakes transferred on $SiN_x$ membrane with arrows showing the folded area of the flakes.** In (a) and (b) white circles are pre-fabricated holes made by FIB.

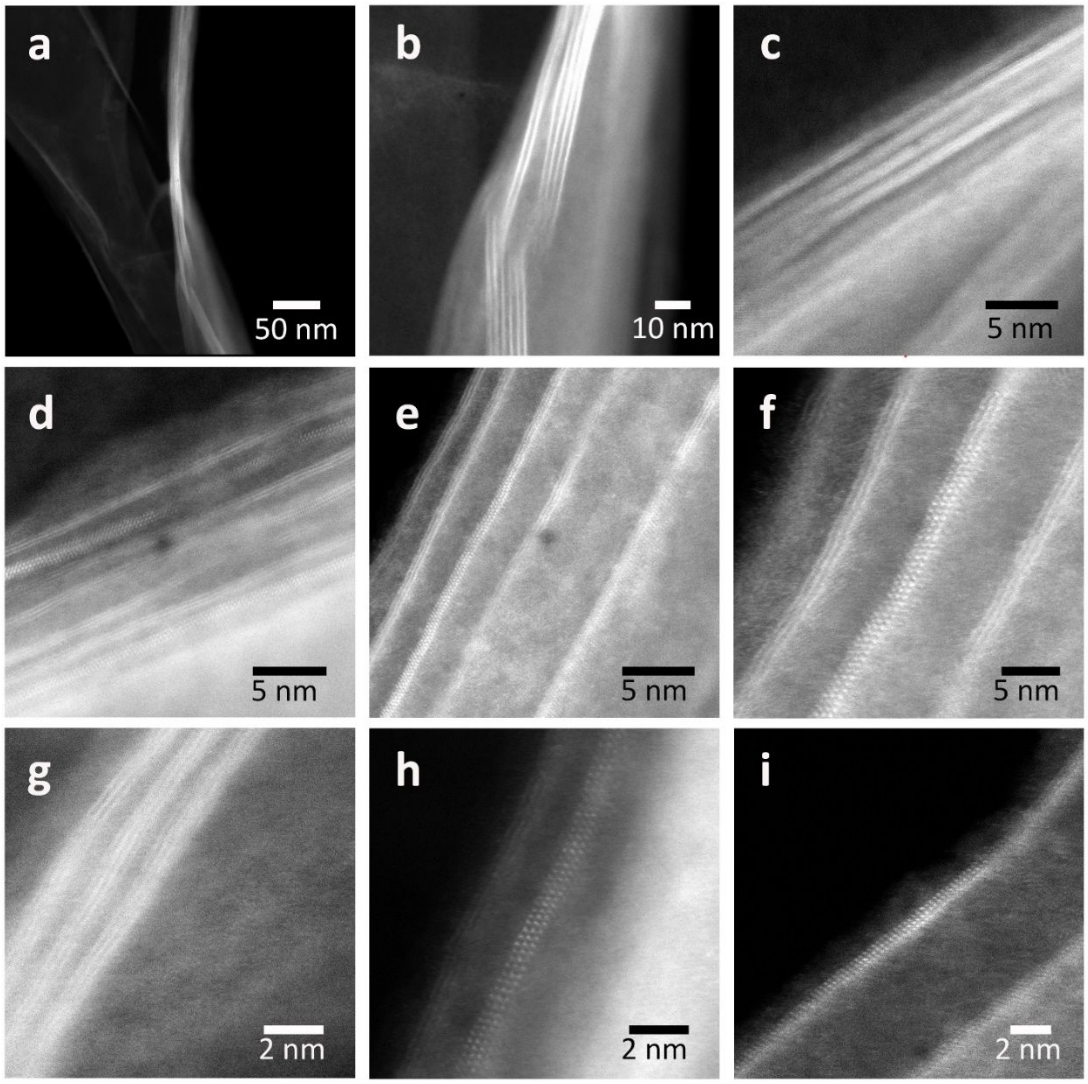



**Figure S8. HAADF STEM images show sectional-view of $Ti_3C_2T_x$ flakes transferred onto a holey carbon filmed TEM grid using the liquid-liquid interface method.** Number of layers in each case is (b) 6 layers. (c) 10 layers. (d) 8 layers. (e) 5 layers. (f) 4 layers. (g) 3 layers. (h) 2 layers. (i) 2 layers.

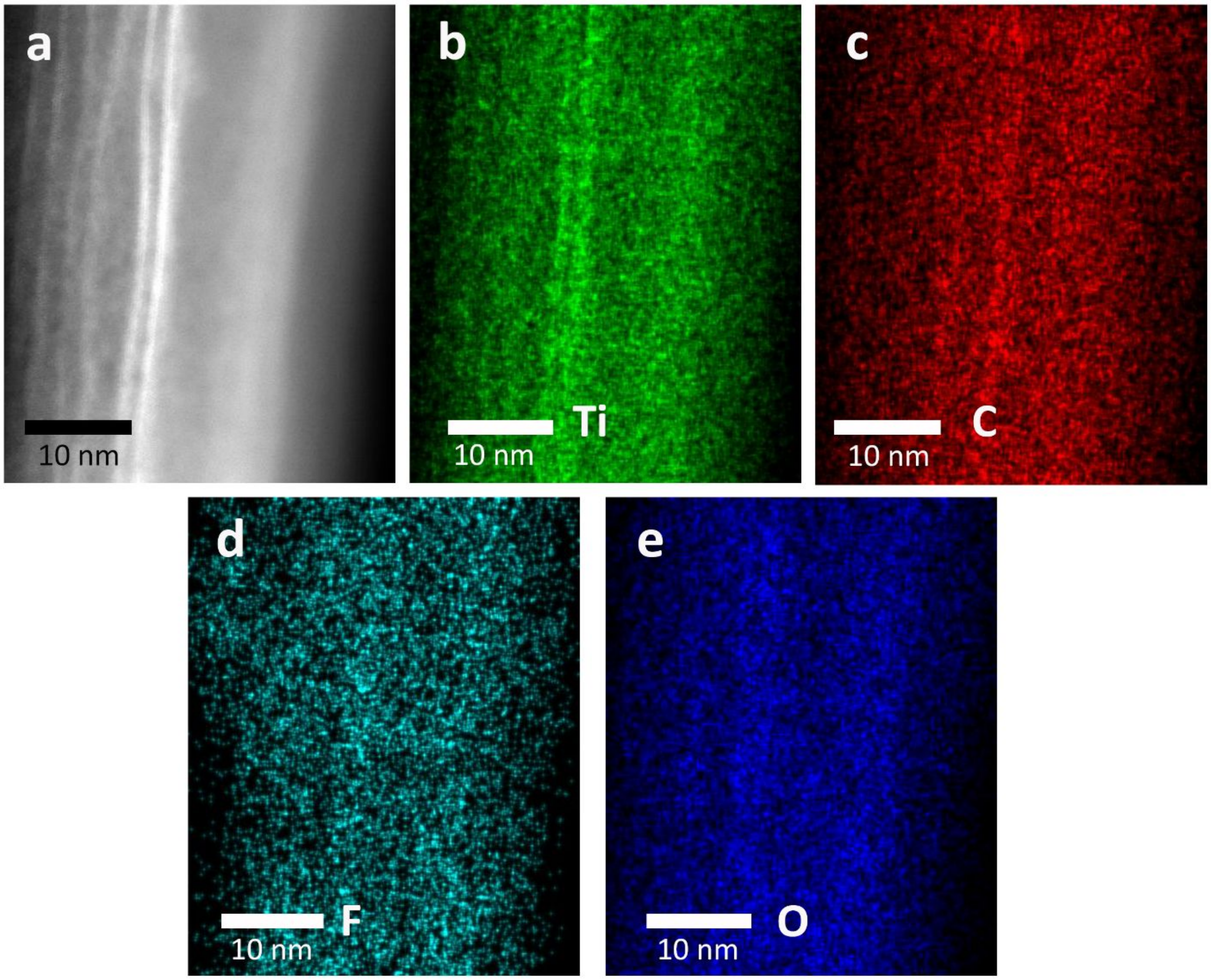
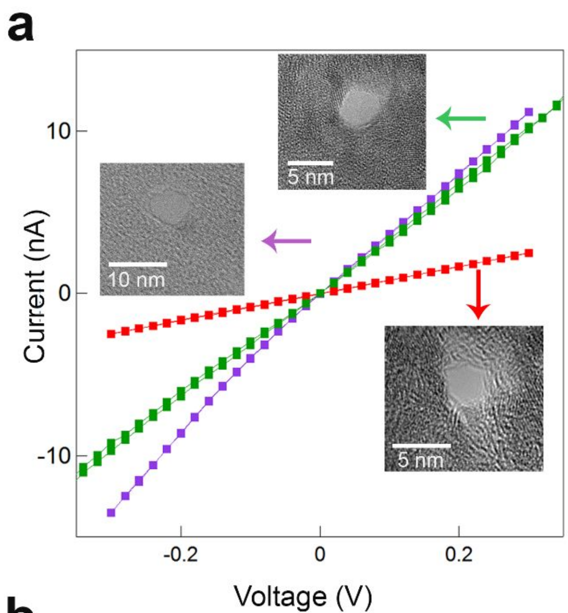



**Figure S9. EDS mapping of folded area of $Ti_3C_2T_x$ flakes.** (a) HAADF STEM image of a sectional view of a multilayer flake. (b) Green shows the density of Ti atoms. (c) Red shows the density of C atoms. (d) Aqua shows the density of F atoms. (e) Blue shows the density of O atoms.



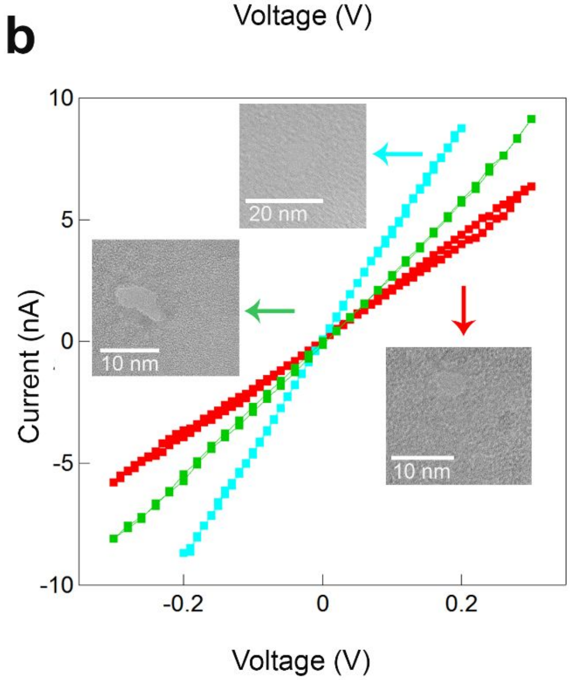




**Figure S10. Example current-voltage curves.** (a) Three $Ti_3C_2T_x$ pores. Buffer used: 0.4 M KCl, 10 mM Tris pH 7.5 (red), 1 M KCl, 10 mM Tris pH 7.5 (green), 0.6 M $NaCH_3COO$ pH 5.5 (purple). (b) Three $Ti_2CT_x$ pores. Buffer used: 0.4 M KCl, 10 mM Tris pH 7.5**.** TEM images of the corresponding pores are shown next to each I-V curve.

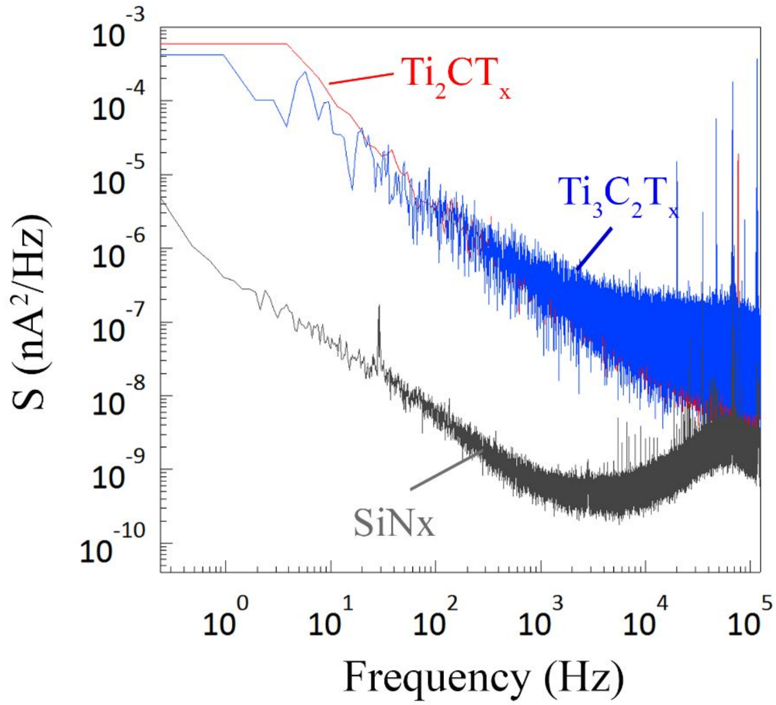



**Figure S11. Power spectral densities obtained from 1.5- second -long current traces at 200 mV for a $Ti_3C_2T_x$ (blue), $Ti_2CT_x$ (red), and a $SiN_x$ (gray) pore** (time-domain traces are shown in Figure 3d). Buffer: 400 mM KCl, 10 mM Tris pH 7.5.

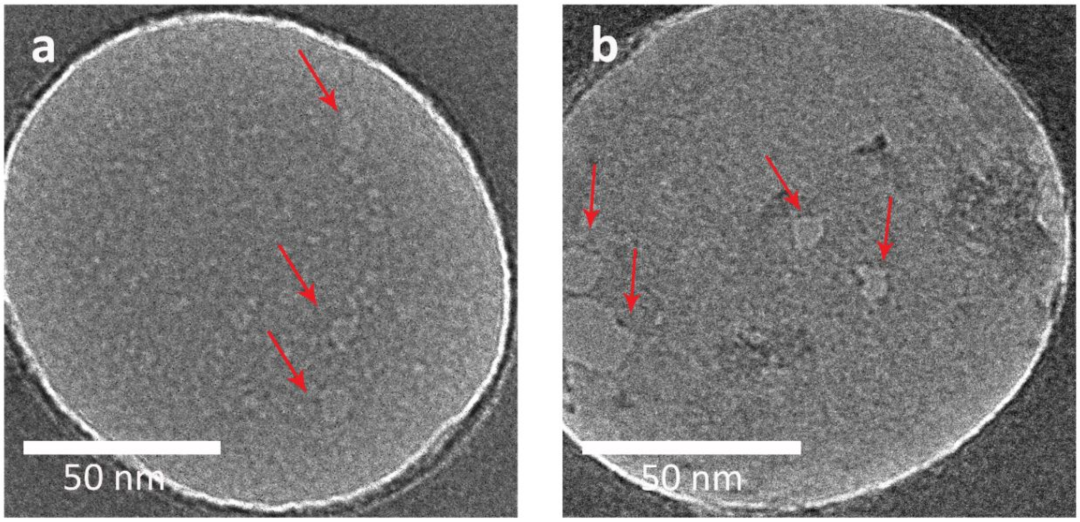



**Figure S12. Freestanding MXene flakes on a 100 nm pre-fabricated hole**. Arrows show small pores emerged as a result of electron beam exposure during nanopore drilling (beam current density: 0.5-1 $nA.nm^{-2}$) or imaging (beam current density: 0.02-0.15 $pA.nm^{-2}$)

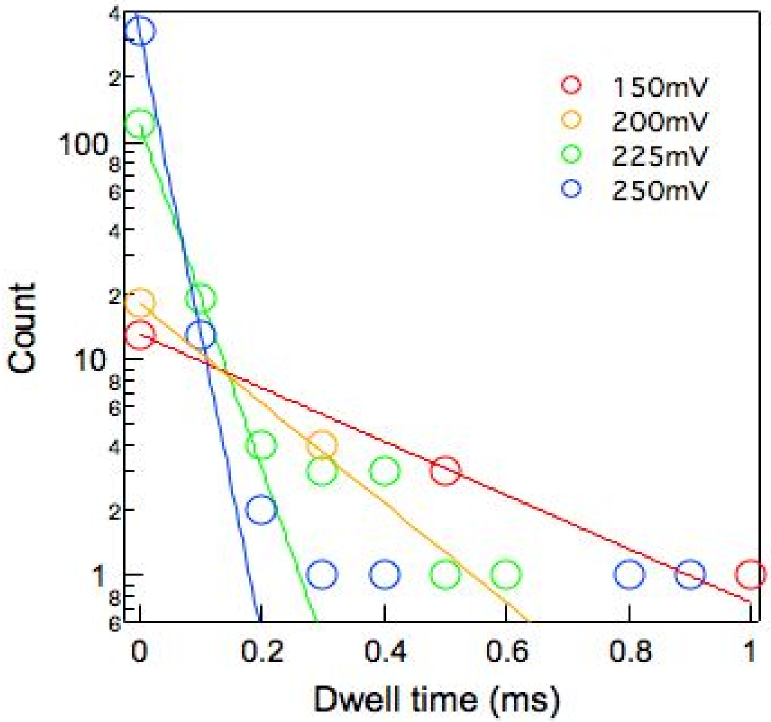



**Figure S13. Dwell time histogram at four different voltages for 4 kbp dsDNA translocation through a 7.2 nm $Ti_2CT_x$ pore (Figure 4).** Lines show the corresponding exponential fit to the data. Buffer: 400 mM KCl, 10 mM Tris pH 7.5.

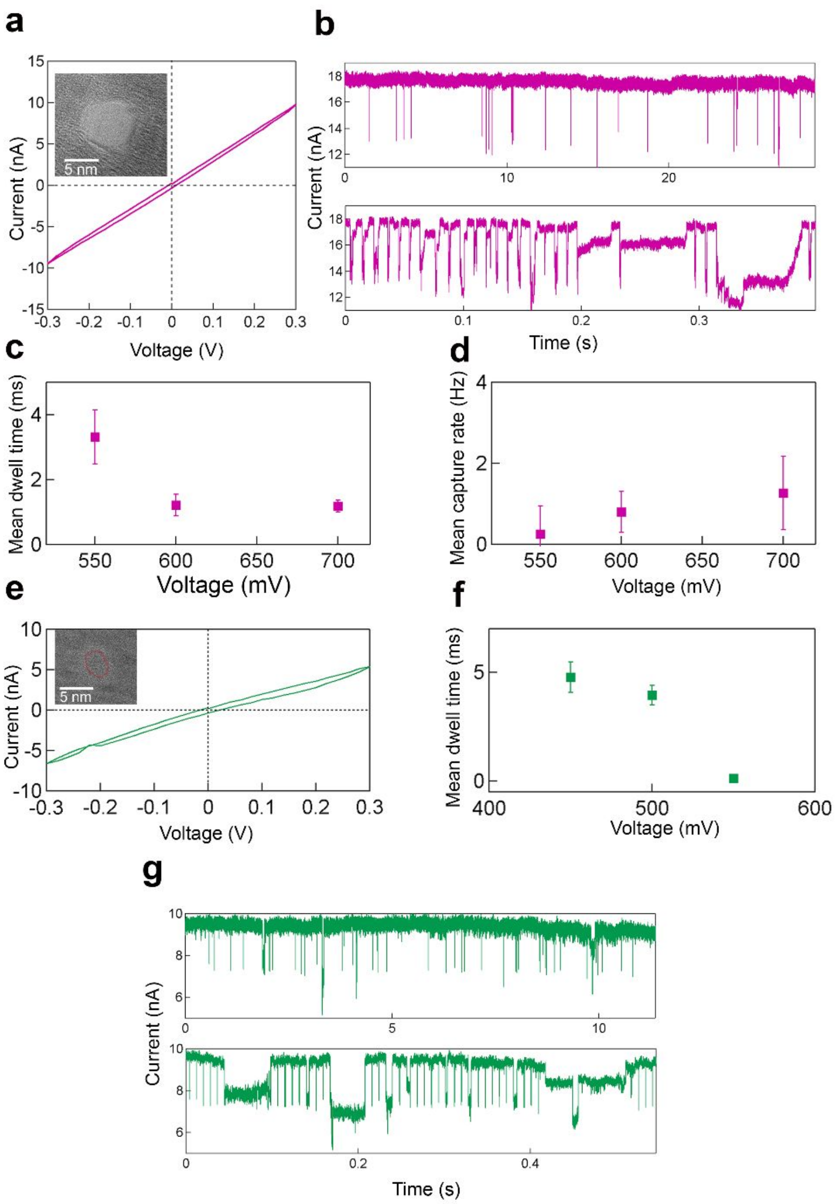



**Figure S14. DNA translocation through $Ti_2CT_x$ nanopore using 2 M LiCl, 20 mM Tris pH 7.5 buffer** (a)Current-voltage curve of 6.4 nm diameter (by TEM) pore. (b) Top panel: current vs. time traces at 700mV after 4 nM 6 kbp dsDNA added. Bottom panel: concatenated traces at 700 mV. (c) Mean DNA translocation dwell time as a function of voltage. (d) Mean DNA capture rate as a function of voltage. (e) Current-voltage curve of 4.5 nm pore (by TEM).. (f) Mean DNA translocation dwell time as a function of voltage after adding 50 nM 2 kbps dsDNA. (g) Top panel: current vs. time traces at 550 mV. Bottom panel: concatenated traces at 550 mV. All traces shown were acquired at a 250 kHz sampling rate and low-pass filtered to 100 kHz.

**Table 1. Comparison between 3 different $Ti_2CT_x$ pores used for DNA translocation**

| Pore Number | Pore Diameter | Buffer | Pore Thickness (nm) | Biomolecule Detection | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | dsDNA Length (kbp) | DNA Con. | Voltage (mV) | Translocation Dwell Time (ms) | Capture Rate (($s^{-1}nM^{-1}$)) | SNR (@10kHz) | Total Number of Events |
| 1 (manuscript) | 7.2 | 0.4 M KCl 20 mM Tris pH 7.5 | 16 | 4 | 3 | 250 | 0.031 | 1.23 | ~10 | 535 |
| 2 (SI) | 6.4 | 2 M LiCl 20 mM Tris pH 7.5 | 9 | 6 | 4 | 700 | 1.18 | 0.31 | >10 | 112 |
| 3 (SI) | 4.5 | 2 M LiCl 20 mM Tris pH 7.5 | 8 | 2 | 50 | 550 | 0.128 | 0.09 | >10 | 266 |