

Electronic Supplementary Information for:

## Electrochemical Performance of MXenes as K-ion Battery Anodes

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### Powders washing after HF etching:

After the reaction time elapsed, the powders were repeatedly washed with de-ionized (DI) water and centrifuged until the pH of the decanted liquid reached values higher than 4. Subsequently, the settled powders were filtered using a vacuum-assisted filtration device and allowed to dry at RT on the filtration device for at least 18 h.

### Electrodes preparation:

The slurry was mixed for 10 minutes in a *Thinky* mixer and tape-cast onto copper foil via a doctor-blade. The films were dried overnight in a vacuum oven at 80°C, and then 12 mm diameter electrodes were punched out with active material density of ~1.75 mg/cm<sup>2</sup>. Stainless steel coin-type 2032 half-cells with potassium metal as the counter electrode were constructed in a high-purity glove box (99.998% Argon) with O<sub>2</sub> and H<sub>2</sub>O concentrations < 1 ppm for electrochemical testing. Whatman 934-AH glass microfiber was used as separator. The electrolyte was 0.8 M KPF<sub>6</sub> (Sigma-Aldrich 98%) in a 1:1 (by volume) of ethylene carbonate (EC, Sigma-Aldrich anhydrous 99%) and diethyl carbonate (DEC, Sigma-Aldrich anhydrous > 99%). All potentials mentioned in this study are vs. K/K<sup>+</sup> unless otherwise specified.

### Characterization:

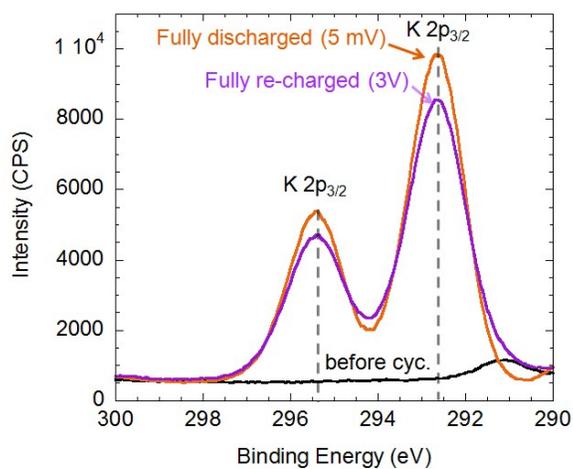
The structure of the resultant MXene was confirmed using a Cu K-alpha X-ray diffractometer (Scintag X1, Scintag, Cupertino, CA, USA). The gas adsorption-desorption isotherms were obtained using a Quantachrome Autosorb-1 with nitrogen adsorbate. Nitrogen sorption analysis at 77 K was used for calculating the specific surface area (SSA) using the Brunauer–Emmet–Teller (BET) equation. The sample was outgassed under vacuum at 200 °C for 48 h before the measurements. X-ray photoelectron spectroscopy (XPS) was performed utilizing a Kratos AXIS Ultra DLD Imaging X-ray Photoelectron Spectrometer connected to an inert glovebox, to enable direct transfer of prepared samples for measurement. XPS measurements were performed on conditioned Ti<sub>3</sub>CNT<sub>z</sub> electrodes (pristine, 5 mV fully discharged, 3 V fully charged), after opened cells were washed with dimethyl carbonate and dried.

Galvanostatic cycling was performed with an Arbin cycler at RT with a voltage range of 0.005 – 3.0 V and with current densities specified (10 mA/g – 500 mA/g). A Gamry Reference-600 electrochemical workstation was used to perform electrochemical impedance spectroscopy (EIS) in the frequency range of 0.01 Hz to 10<sup>6</sup> Hz. All the electrochemical measurements were carried out at room temperature.

## X-ray photoelectron spectroscopy

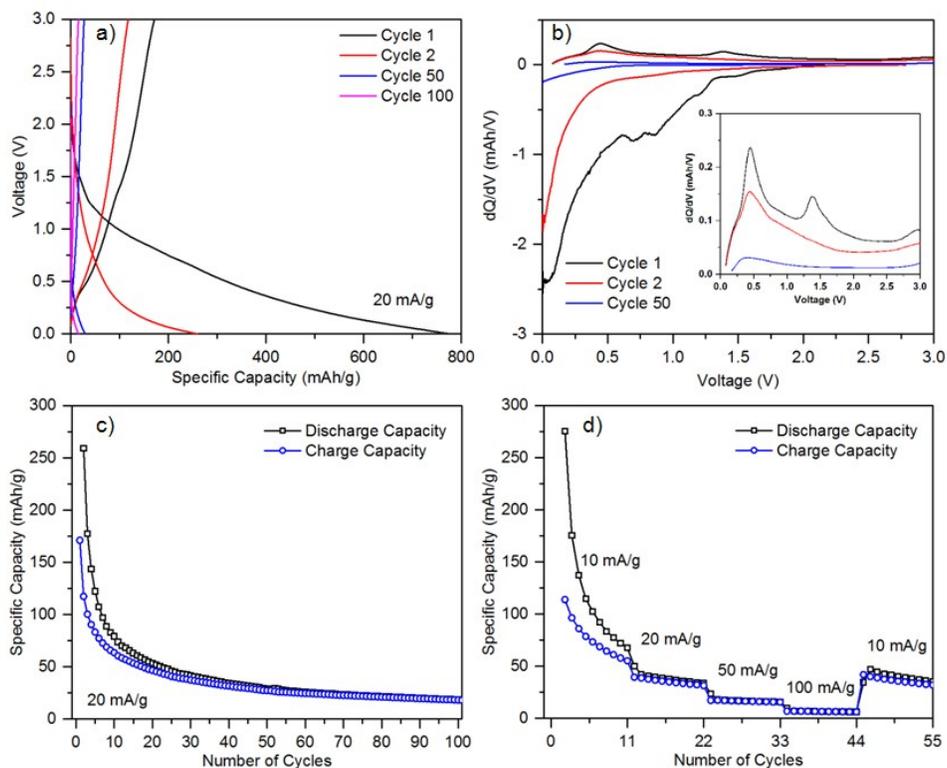
**Table S1** Atomic percentage estimated from X-ray photoelectron spectroscopy for all the elements on the surface of the  $\text{Ti}_3\text{CNT}_z$  electrodes before electrochemical cycling, after discharging to 5 mV, and recharging back to 3 V vs.  $\text{K}/\text{K}^+$ .

Sample	Atomic %								
	C 1s	Cl 2p	F 1s	K 2p	N 1s	Na 1s	O 1s	P 2p	Ti 2p
before cycling	62.5	0.0	15.0	0.0	3.6	0.2	10.9	0.0	7.7
5 mV	23.2	0.6	17.4	24.4	0.0	0.2	31.8	2.3	0.0
3.0 V	18.4	0.7	21.8	23.9	0.5	1.5	29.6	1.8	1.7

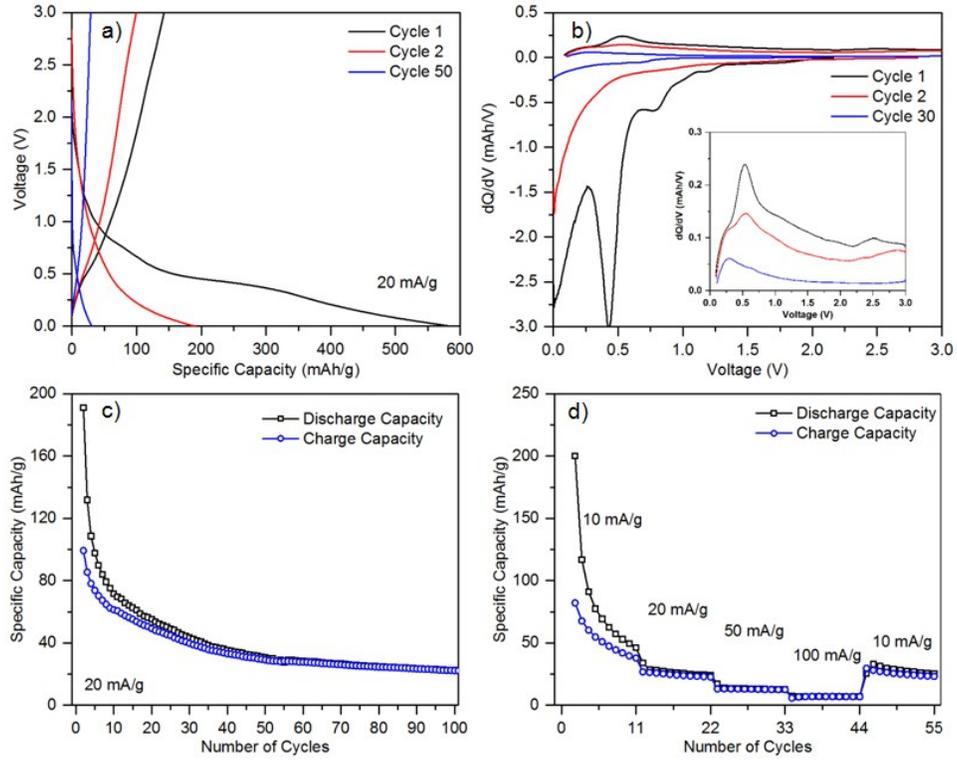


**Fig. S1** K 2p X-ray photoelectron spectra for  $\text{Ti}_3\text{CNT}_z$  electrodes before cycling (black), after fully discharging down to 5 mV (orange), and after recharging back to 3V (purple).

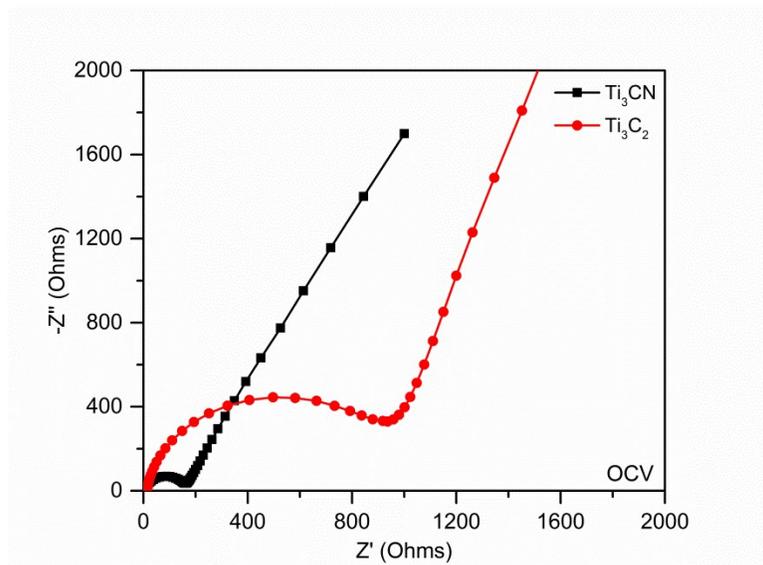
## Electrochemical Performance of $\text{Ti}_3\text{C}_2\text{T}_z$ and $\text{Nb}_2\text{CT}_z$ :



**Fig. S2** The electrochemical performance of  $\text{Ti}_3\text{C}_2\text{T}_z$  electrode. (a) Galvanostatic voltage profile for cycles number 1, 2, 50 and 100 at 20 mA/g between 5.0 mV and 3.0 V vs.  $\text{K}/\text{K}^+$ . (b) The corresponding  $dQ/dV$  for the results presented in 1a. The inset is a higher magnification for the potassiation part of the plot in b. (c) Capacity vs. cycle number at 20 mA/g. (d) Capacity vs. cycle number at rates of 10-100 mA/g.

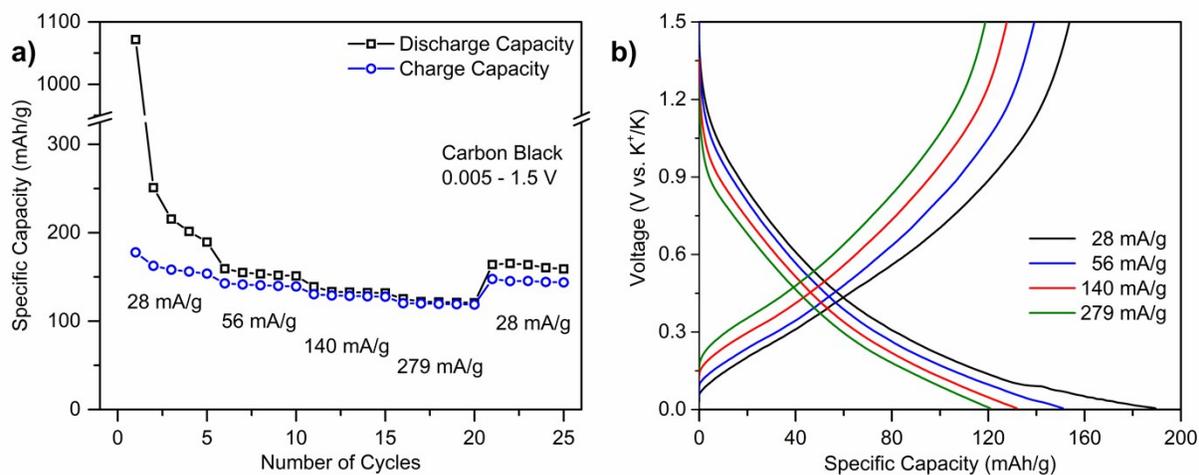


**Fig. S3** The electrochemical performance of  $\text{Nb}_2\text{CT}_z$  electrode. (a) Galvanostatic voltage profile for cycles number 1, 2, 50 and 100 at 20 mA/g between 5.0 mV and 3.0 V vs.  $\text{K}/\text{K}^+$ . (b) The corresponding  $dQ/dV$  for the results presented in 1a. The inset is a higher magnification for the potassiation part of the plot in b. (c) Capacity vs. cycle number at 20 mA/g. (d) Capacity vs. cycle number at rates of 10-100 mA/g.



**Fig. S4** Electrochemical impedance spectroscopy (EIS) Nyquist plot of  $\text{Ti}_3\text{C}_2\text{T}_z$  and  $\text{Ti}_3\text{CNT}_z$  at open circuit voltage

To elucidate the contribution of carbon black for K-ion storage, we made an electrode from carbon black and PVDF (9:1 wt. ratio). The electrochemical results are shown in Fig S5, where a reversible capacity of 150 mAh/g at 28 mA/g is achieved. The MXene electrodes have 15% wt. of carbon black, thus a small portion of the capacity are contributed by the carbon black. However, the peaks observed in the differential capacity plot (Fig 1b) are not seen in the carbon black voltage profile meaning that an intercalation storage mechanism of K-ions in the MXene anodes is occurring.



**ig S5** The electrochemical performance of a carbon black electrode (90:10 carbon black to PVDF wt. ratio) in a KIB half cell. (a) Galvanostatic cycling at various current densities between 5 mV and 1.5 V. (b) Galvanostatic voltage profiles at various current densities.