

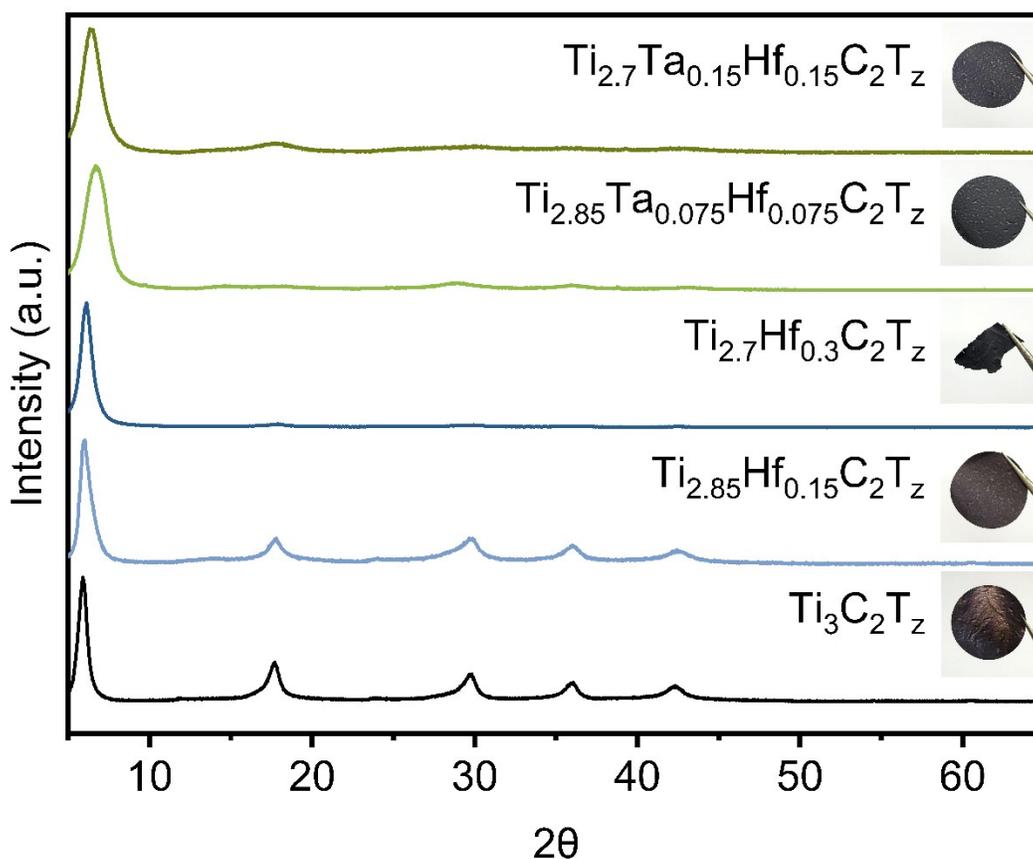
## Extending the Electrochemical Stability Window of $\text{Ti}_3\text{C}_2\text{T}_z$ MXenes via Ta–Hf Co-Substitution for Durable Proton Exchange Membrane Fuel Cell Catalyst Supports

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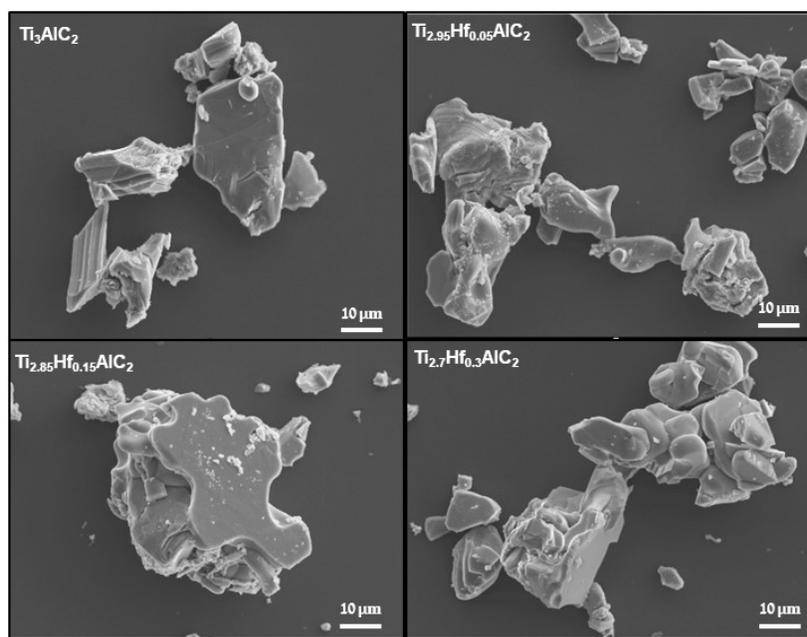
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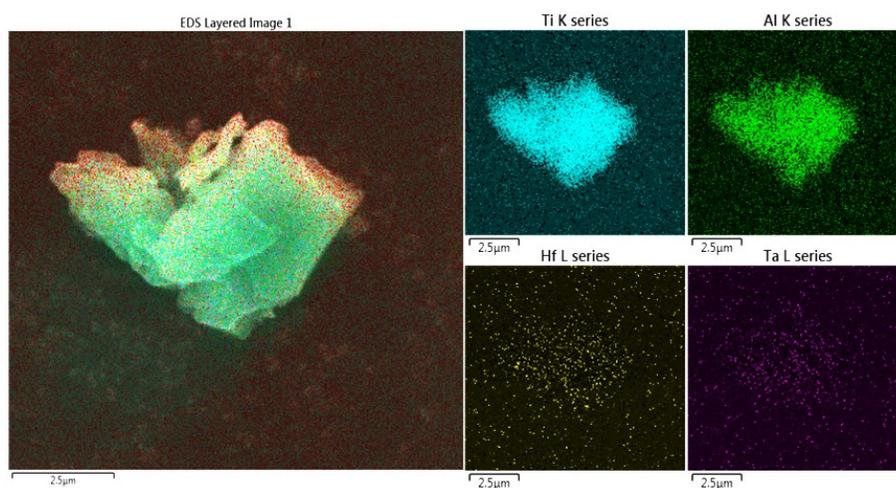
Supplementary information



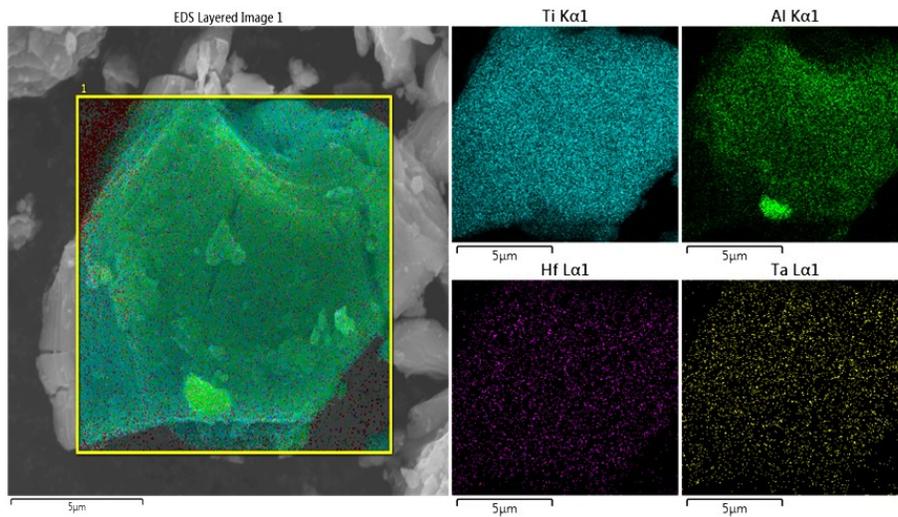
**Figure S1.** Comparative XRD pattern of the pristine  $\text{Ti}_3\text{C}_2\text{T}_z$ ,  $\text{Ti}_{2.85}\text{Hf}_{0.15}\text{C}_2\text{T}_z$ ,  $\text{Ti}_{2.7}\text{Hf}_{0.3}\text{C}_2\text{T}_z$ ,  $\text{Ti}_{2.85}\text{Ta}_{0.075}\text{Hf}_{0.075}\text{C}_2\text{T}_z$  and  $\text{Ti}_{2.7}\text{Ta}_{0.15}\text{Hf}_{0.15}\text{C}_2\text{T}_z$ . Insert shows freestanding films of as synthesized MXenes.



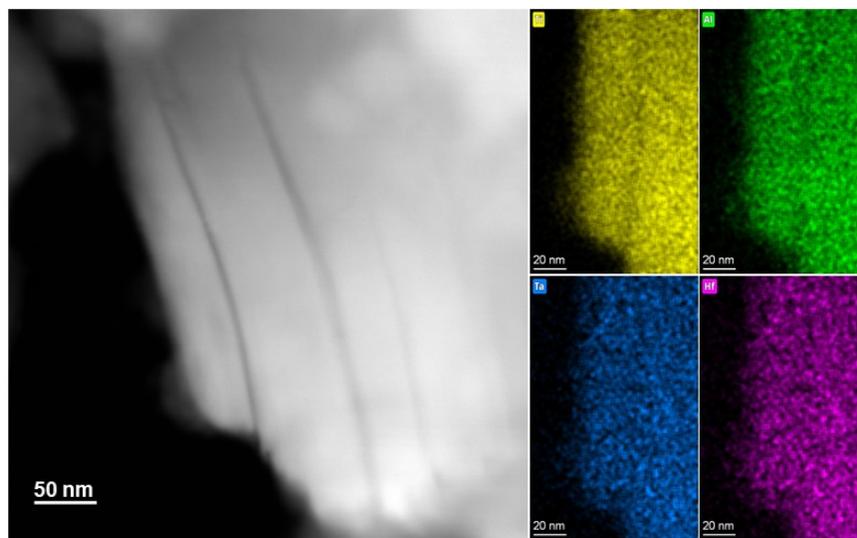
**Figure S2.** SEM images of the pristine Ti<sub>3</sub>AlC<sub>2</sub>, Ti<sub>2.95</sub>Hf<sub>0.05</sub>AlC<sub>2</sub>, Ti<sub>2.85</sub>Hf<sub>0.15</sub>AlC<sub>2</sub> and Ti<sub>2.7</sub>Hf<sub>0.3</sub>AlC<sub>2</sub>.



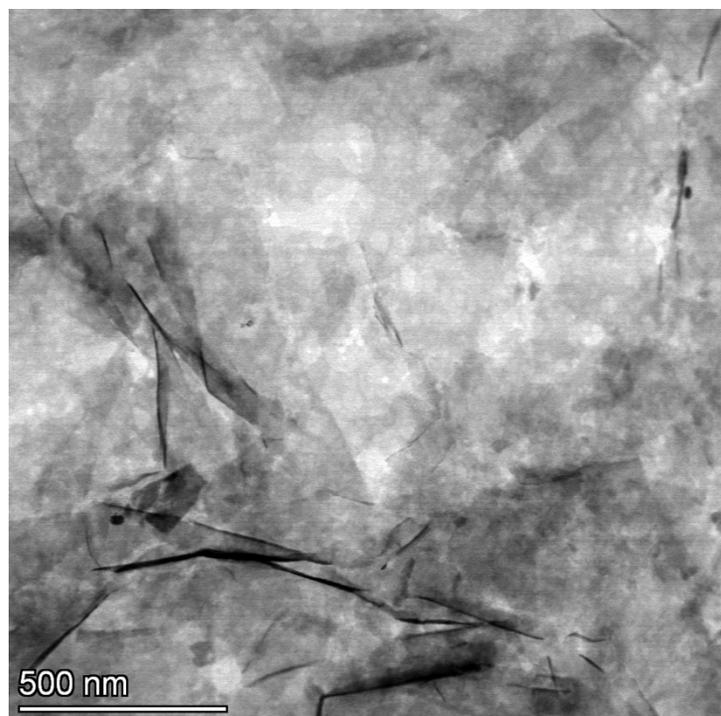
**Figure S3.** SEM-EDS elemental mapping of the Ti<sub>2.85</sub>Ta<sub>0.075</sub>Hf<sub>0.075</sub>AlC<sub>2</sub>.



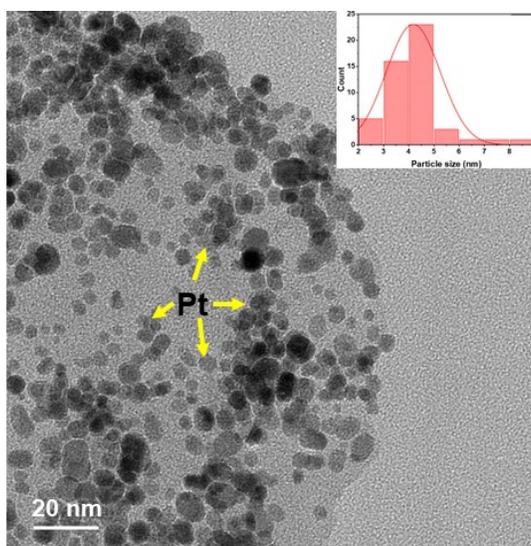
**Figure S4.** SEM-EDS elemental mapping of the  $\text{Ti}_{2.7}\text{Ta}_{0.15}\text{Hf}_{0.15}\text{AlC}_2$ .



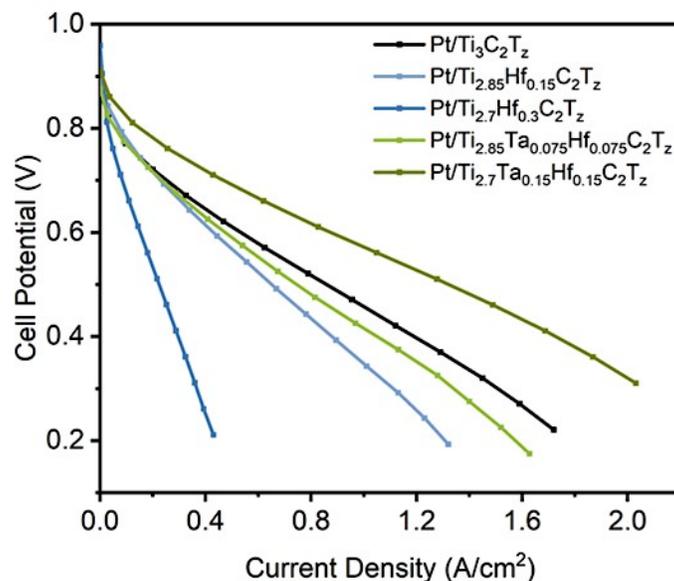
**Figure S5.** TEM-EDS elemental mapping of the  $\text{Ti}_{2.7}\text{Ta}_{0.15}\text{Hf}_{0.15}\text{AlC}_2$ .



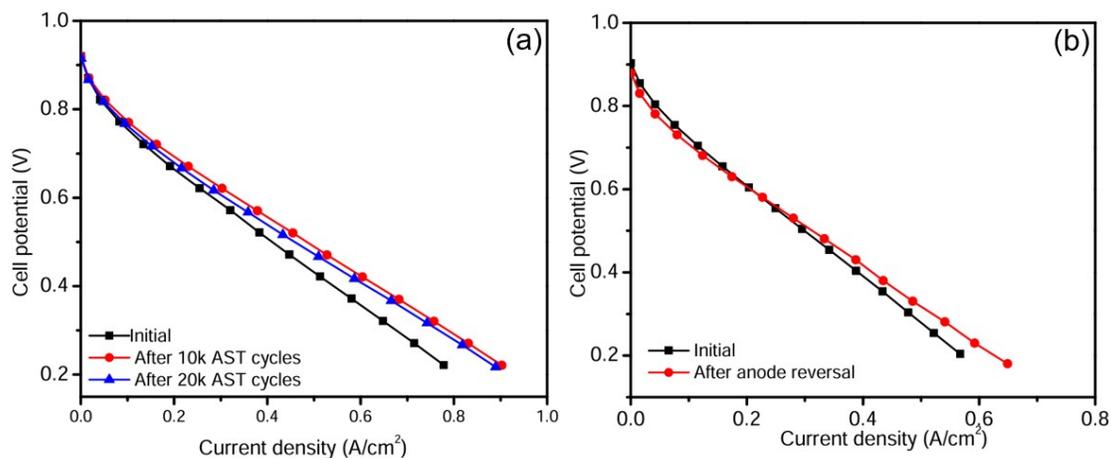
**Figure S6.** HR-TEM image of  $\text{Ti}_{2.7}\text{Ta}_{0.15}\text{Hf}_{0.15}\text{C}_2\text{T}_z$  after scanning to the 0.9 V vs. RHE at 0.167 mV/s scan rate in the 0.5 M  $\text{H}_2\text{SO}_4$  showing localized oxidation.



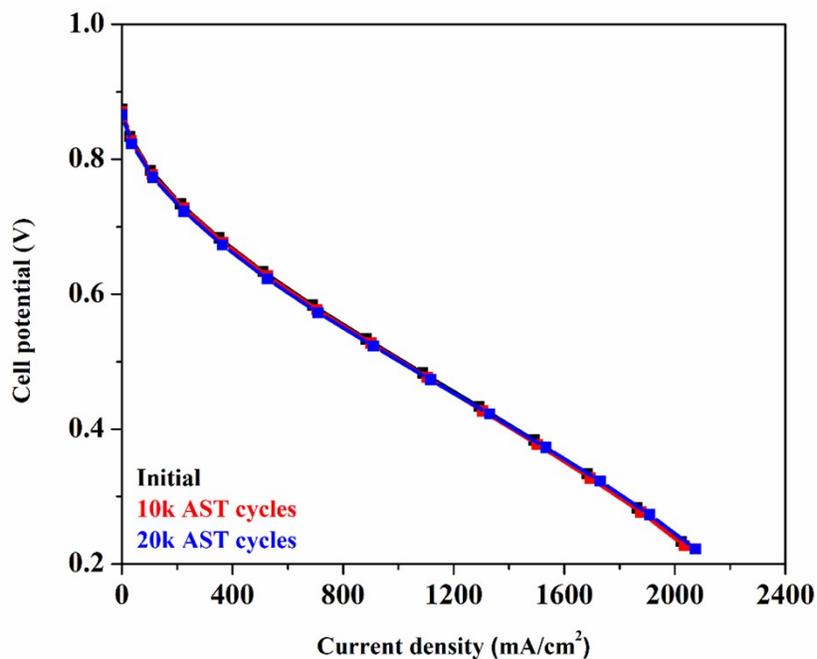
**Figure S7.** Bright field TEM images of the Pt loaded  $\text{Ti}_{2.7}\text{Ta}_{0.15}\text{Hf}_{0.15}\text{C}_2\text{T}_z$ . Inset indicate the average particle distribution of the Pt.



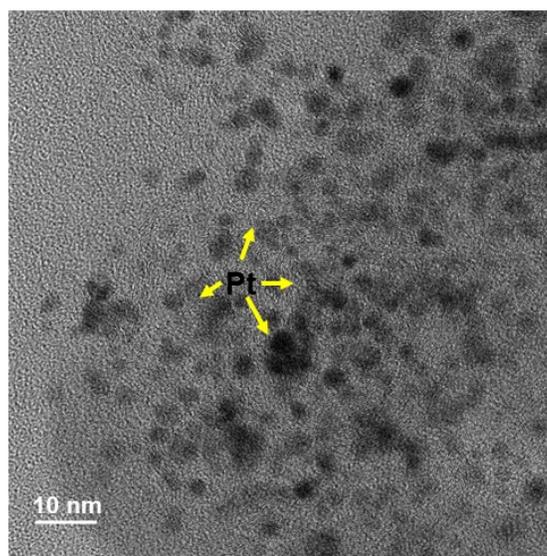
**Figure S8.** Comparative polarization curve using commercial Pt/C cathode and the Pt anode supported by  $\text{Pt}/\text{Ti}_3\text{C}_2\text{T}_z$ ,  $\text{Pt}/\text{Ti}_{2.85}\text{Hf}_{0.15}\text{C}_2\text{T}_z$ ,  $\text{Pt}/\text{Ti}_{2.7}\text{Hf}_{0.3}\text{C}_2\text{T}_z$ ,  $\text{Pt}/\text{Ti}_{2.85}\text{Ta}_{0.075}\text{Hf}_{0.075}\text{C}_2\text{T}_z$  and  $\text{Pt}/\text{Ti}_{2.7}\text{Ta}_{0.15}\text{Hf}_{0.15}\text{C}_2\text{T}_z$ .



**Figure S9.** Comparative AST polarization curves of fuel cell cycled between 0.6 to 0.9 V using state-of-the-art Pt/C cathode and Pt anode supported by (a)  $\text{Ti}_{2.7}\text{Ta}_{0.075}\text{Hf}_{0.075}\text{C}_2\text{T}_z$ . (b) Polarization curves for  $5\text{ cm}^2$  fuel cell using Pt/C cathode and Pt supported by  $\text{Ti}_{2.7}\text{Ta}_{0.075}\text{Hf}_{0.075}\text{C}_2\text{T}_z$  before and after anode reversal measurement.



**Figure S10.** AST curves for a 5 cm<sup>2</sup> PEM fuel cell cycled between 0.6-0.8 V using commercial Pt/C cathode and a Pt anode supported by (Ti<sub>0.95</sub>Ta<sub>0.05</sub>)<sub>3</sub>C<sub>2</sub>T<sub>z</sub>. b) Polarization curves for a 5 cm<sup>2</sup> PEM fuel cell with a Pt/C cathode and a Pt anode supported by (Ti<sub>0.95</sub>Ta<sub>0.05</sub>)<sub>3</sub>C<sub>2</sub>T<sub>z</sub> before and after anode reversal. Adv Funct Materials, Volume: 34, Issue: 10, First published: 03 November 2023, DOI: (10.1002/adfm.202309749).



**Figure S11.** Bright-field TEM image of Pt/Ti<sub>2.7</sub>Ta<sub>0.15</sub>Hf<sub>0.15</sub>C<sub>2</sub>T<sub>z</sub> recovered from the PEMFC anode catalyst layer after 20k AST cycles between 0.6-0.9V.