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Supporting Information

Supercritical CO_2-assisted Solid-Phase Etching Preparation of MXenes for High-efficiency Alkaline Hydrogen Evolution

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Supplementary Text

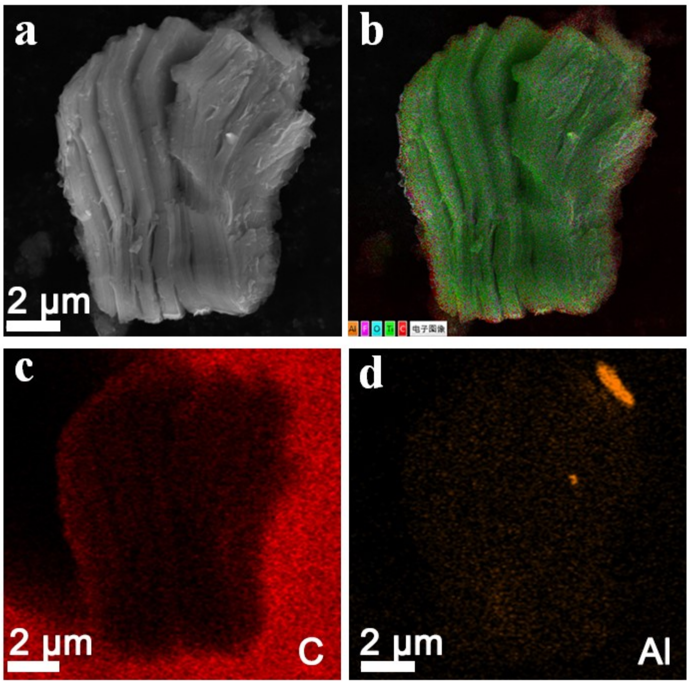

Fig. S1. (a-d) SEM image (a), EDS layered image (b) and elemental-mapping images (C and Al elements) (c-d) of the HCl-treated SC CO_2-$Ti_3C_2T_x$ multilayer MXenes.

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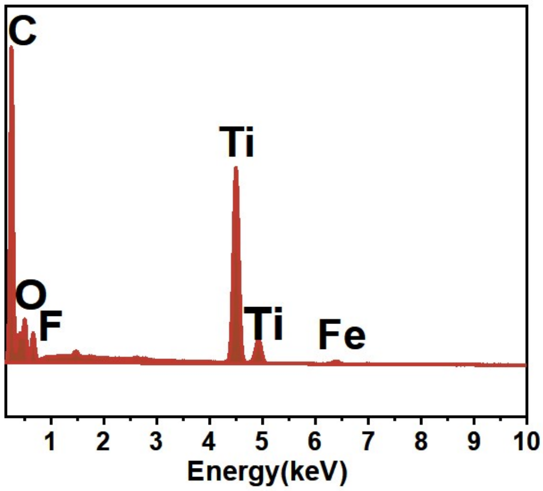

Fig. S2. EDS analysis of the HCl-treated SC CO_2-$Ti_3C_2T_x$ multilayer MXenes samples.

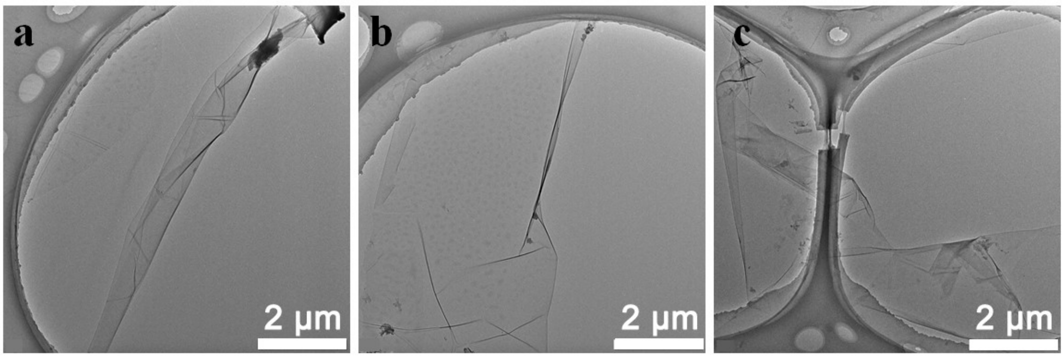

Fig. S3. TEM images of the delaminated 2D SC CO_2-$Ti_3C_2T_x$ MXenes flakes.

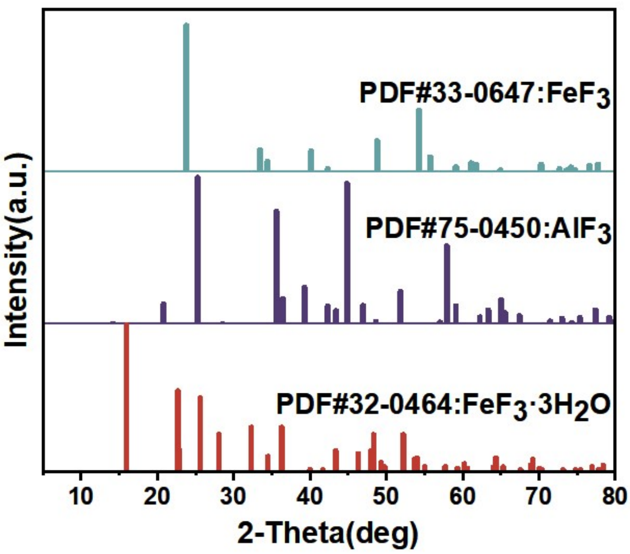

Fig. S4. the standard PDF cards of FeF_3•$3H_2O$ (red line), AlF_3 (purple line) and FeF_3 (blue line).

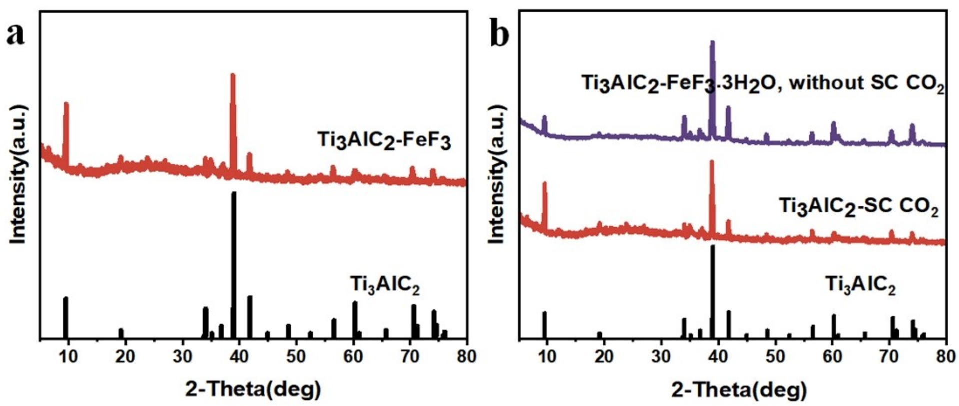

Fig. S5. (a) XRD patterns of pristine Ti_3AlC_2 MAX phase (black line) and HCl-treated Ti_3AlC_2-FeF_3 (red line). (b) XRD patterns of pristine Ti_3AlC_2 MAX phase (black line), HCl-treated Ti_3AlC_2-SC CO_2 (red line) and Ti_3AlC_2-FeF_3•$3H_2O$, without SC CO_2 (purple line).

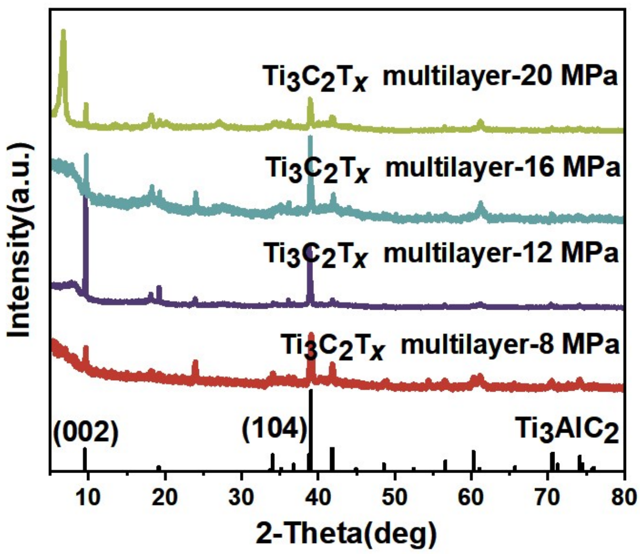

Fig. S6. XRD patterns of pristine Ti_3AlC_2 MAX phase (black line) and HCl-treated SC CO_2-$Ti_3C_2T_x$ multilayer MXenes samples after etching at the temperature of 90 °C and under 8 MPa (red line), 12 MPa (purple line), 16 MPa (blue line), 20 MPa (yellow line), respectively.

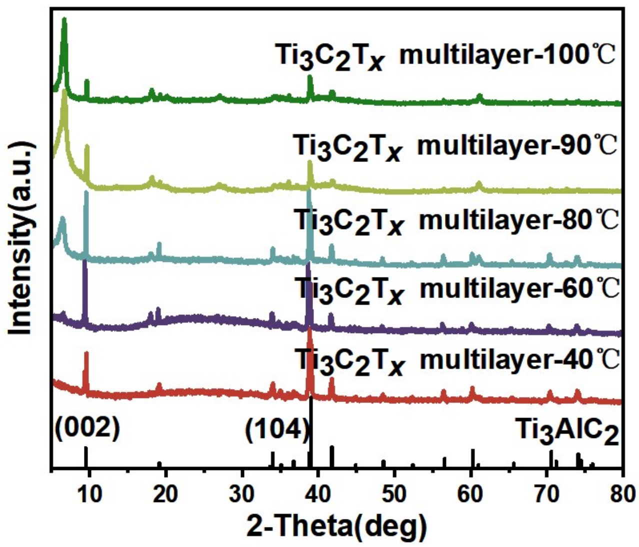

Fig. S7. XRD patterns of pristine Ti_3AlC_2 MAX phase (black line) and HCl-treated SC CO_2-$Ti_3C_2T_x$ multilayer MXenes samples etching under the pressure of 20 MPa and at 40 ℃ (red line), 60 ℃ (purple line), 80 ℃ (blue line), 90 ℃ (yellow line), 100 ℃ (green line), respectively.

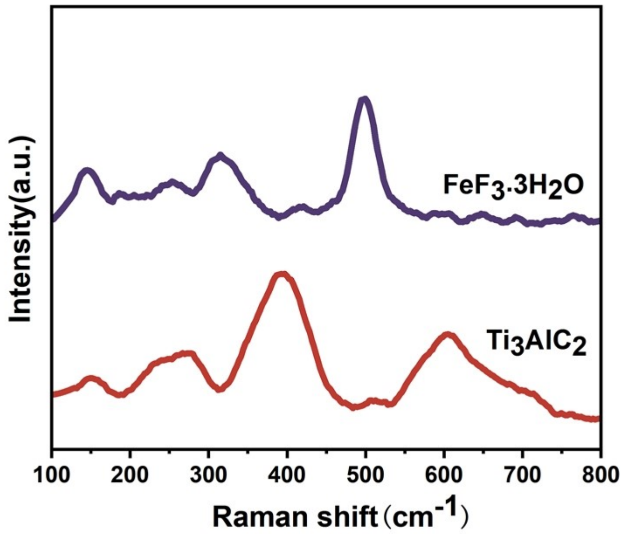

Fig. S8. Raman spectra of Ti_3AlC_2 (red line) and FeF_3•$3H_2O$ (purple line).

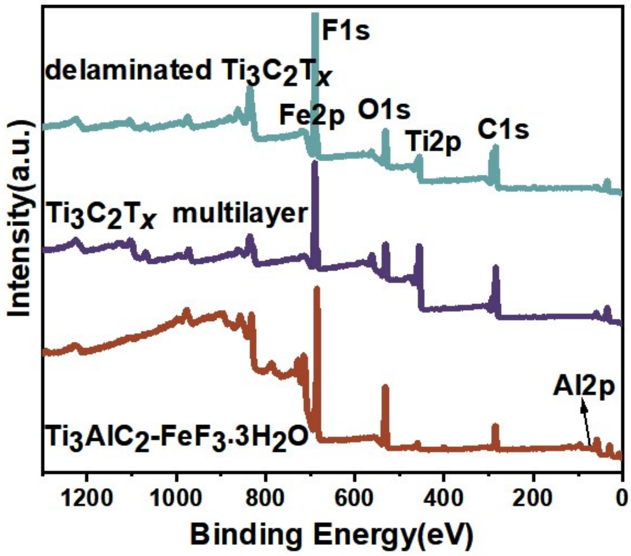

Fig. S9. The global view of the XPS spectra in the as-reacted product, HCl-treated $Ti_3C_2T_x$ MXenes and the delaminated MXenes flakes.

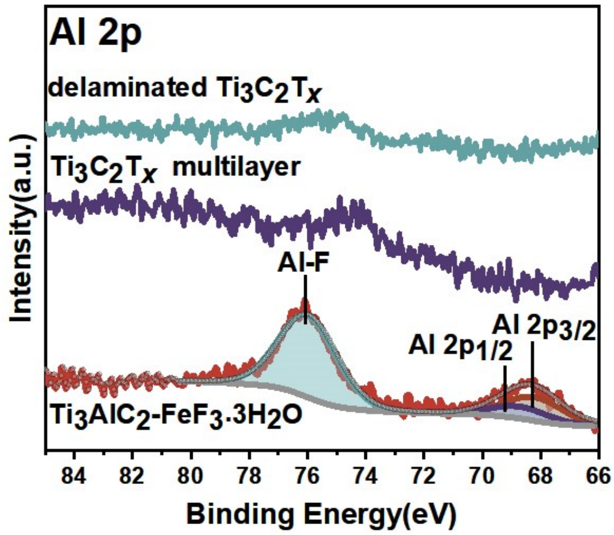

Fig. S10. High-resolution XPS spectra of Al 2p in the as-reacted product, HCl-treated $Ti_3C_2T_x$ MXenes and the delaminated MXenes flakes.

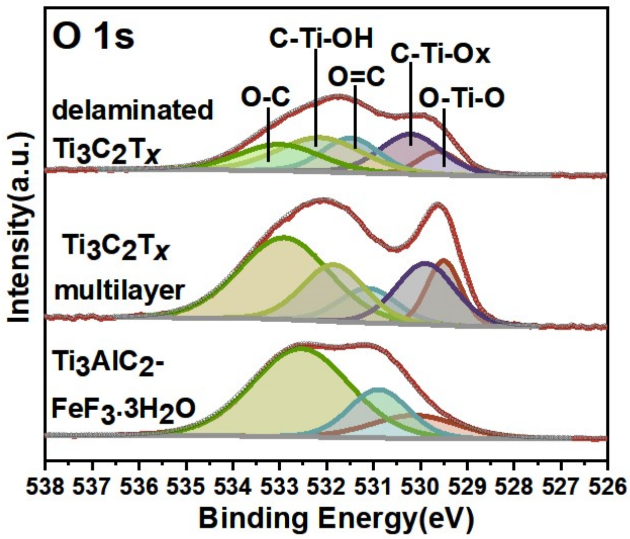

Fig. S11. High-resolution XPS spectra of O 1s in the as-reacted product, HCl-treated $Ti_3C_2T_x$ MXenes and the delaminated MXenes flakes.

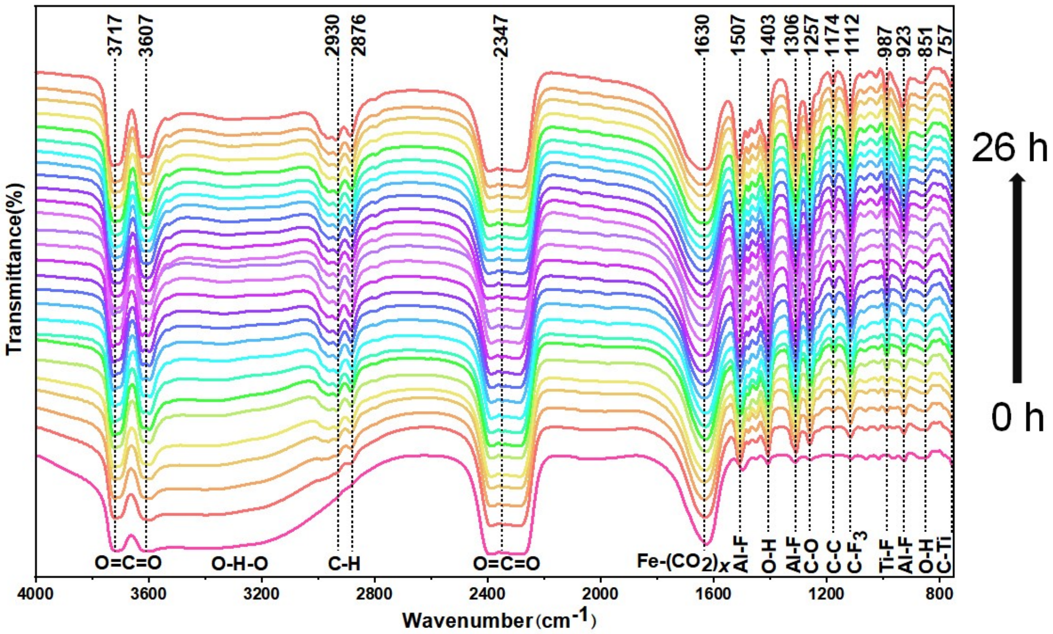

Fig. S12. Time-resolved in situ IR experimental results (20 MPa SC CO_2, 90 °C), collected in the 4000-750 cm^{-1} region.

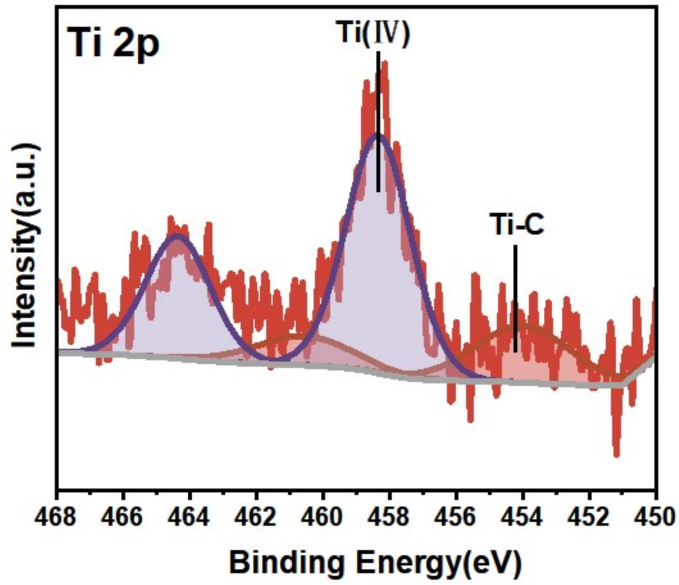

Fig. S13. High-resolution XPS spectra of Ti 2p in the SC CO_2-$Ti_3C_2T_x$-Pt.

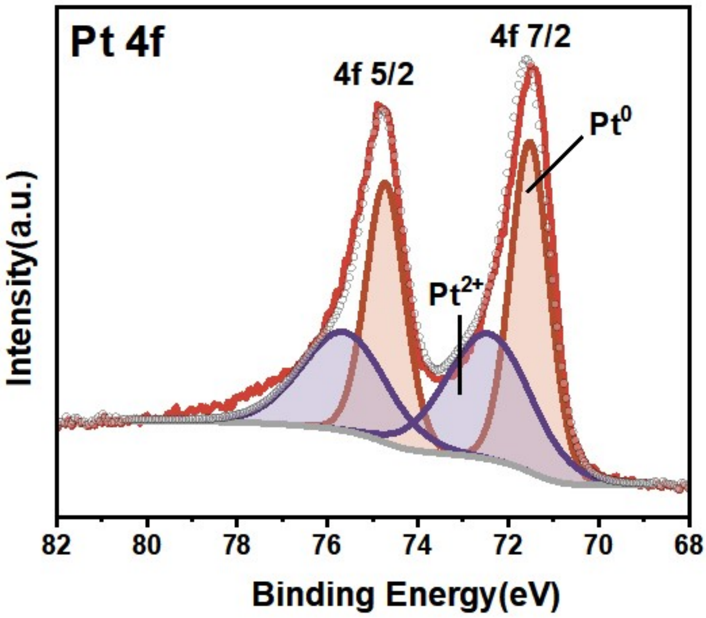

Fig. S14. High-resolution XPS spectra of Pt 4f in the SC CO_2-$Ti_3C_2T_x$-Pt.

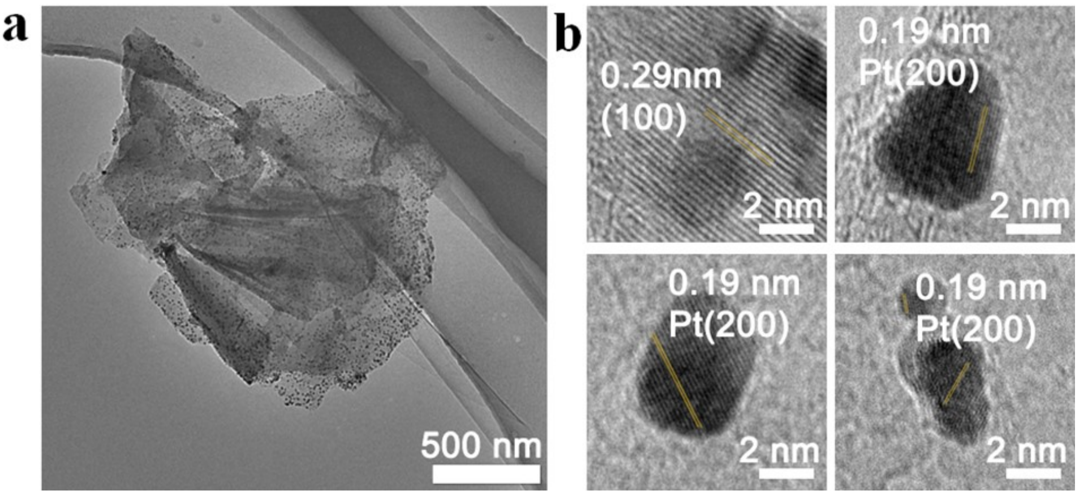

Fig. S15. TEM image (a) and high-resolution TEM images (b) of the SC CO_2-$Ti_3C_2T_x$-Pt.

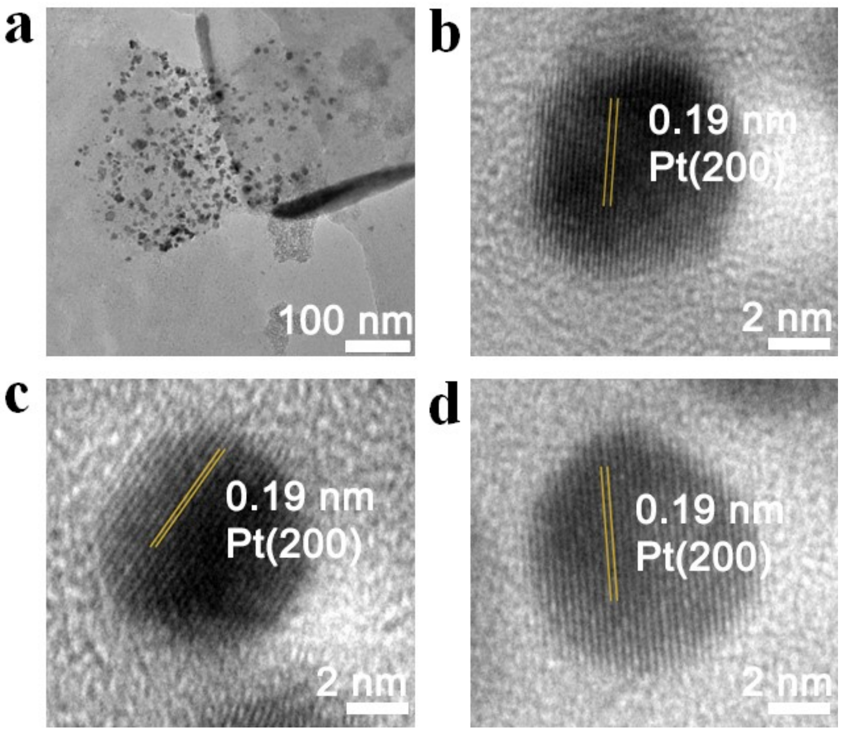

Fig. S16. TEM image (a) and high-resolution TEM images (b, c, d) of the LiF-$Ti_3C_2T_x$-Pt.

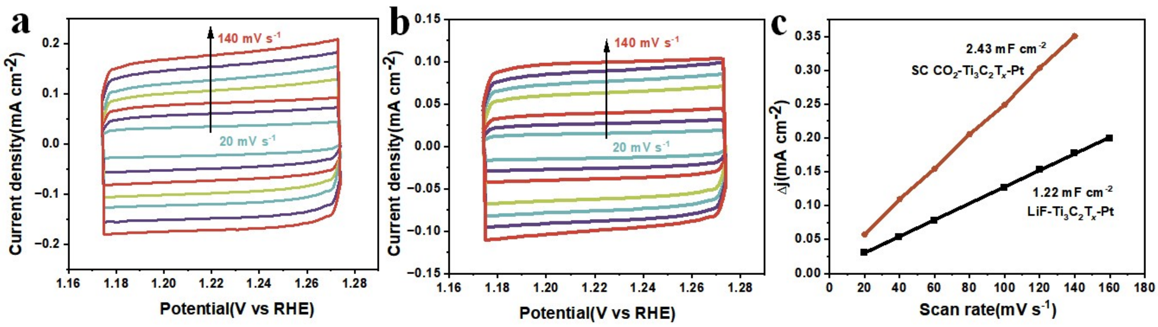

Fig. S17. Cyclic voltammograms performed in 1 M KOH solution in the potential window without the participation of faradaic processes for (a) SC CO_2-$Ti_3C_2T_x$-Pt and (b) LiF-$Ti_3C_2T_x$-Pt. (c) Plots of the differences in current density variation against scan rate, where the slope is C_{dl}.

Table. S1. Comparison of HER performances for SC CO_2-$Ti_3C_2T_x$-Pt with the reported HER catalysts in 1.0 M KOH electrolyte.

	Electrocatalysts	Overpotential at 10 mA cm^{-2} (mV)	Overpotential at 100 mA cm^{-2} (mV)	Tafel slop (mV dec^{-1})	Reference
1	**SC CO_2-$Ti_3C_2T_x$-Pt**	**29**	**117**	**67.22**	**This work**
2	Pt_{SA}-C_1N_1	46	200	36.8	Nat Commun. 2020, 11, 1029
3	Pt/np-$Co_{0.85}Se$	58	-	35	Nat. Commun. 2017, 10, 1743
4	$Ti_3C_2T_x$/Ni_3S_2/NF	72	-	45	ACS Applied Energy Materials 2019, 2 (9), 6931-6938
5	Pt NWs/SL-$Ni(OH)_2$	~70	-	-	Nat Commun. 2015, 6, 6430
6	Ni_3N/Pt	50	110	36.5	Adv. Energy Mater. 2017, 7, 1601390
7	WS_2-Pt	~40	-	65	Adv. Mater. 2017, 30, 1704779
8	Pt/MXene	34	185	29.7	Adv. Funct. Mater. 2022, 32, 2110910