

## Supporting Information

### Chlorinated-Ti<sub>3</sub>C<sub>2</sub>T<sub>F</sub> as Dual Functional Buried-Interface on SnO<sub>2</sub> Electron Transporting Layer for 25.09% High Performance *n-i-p* Perovskite Solar Cells

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## Methods

### Materials

ITO-coated glasses for small-scale device and large-scale device (<15 Ω sq<sup>-1</sup>) were both bought from Advanced Election Technology Co. SnO<sub>2</sub> colloid precursor (15% in H<sub>2</sub>O colloidal dispersion) was purchased from Alfa Aesar. Multilayer MXene was purchased from Jilin 11 Technology Co., Ltd. PbI<sub>2</sub> (99.99%), formamidinium iodide (FAI), methylammonium chloride (MACl), methylammonium bromide (MABr), methylammonium iodide (MAI), Spiro-OMeTAD and bis(trifluoromethane)sulfonamide lithium salt (Li-TFSI) were purchased from Xi'an Polymer Light Technology Corp. *N,N*-Dimethylformamide (DMF, anhydrous, 99.8%), dimethyl sulfoxide (DMSO, anhydrous, 99.8%), isopropanol (IPA, anhydrous, 99.8%) and chloroform (CF) were acquired from J&K Technology. Chlorobenzene (CB) and acetonitrile were acquired from Sigma-Aldrich.

### Preparation of Ti<sub>3</sub>C<sub>2</sub>T<sub>F, Cl</sub> chloroform solution

First, dispersed multilayer MXene in an aqueous solution, sonicated the solution at 300 W for 0.5, 1.0, 1.5, 2.0 h below 20 °C, and centrifuged the solution at 6000 rpm for 10 minutes to separate the unflaked MXene to obtain the single-layer MXene nanosheet. Then, the MXene nanosheet was reacted with concentrated HCl (9 mol/ml) and stirred for 20, 40, 60 and 80 min at room temperature, to substitute excess terminal groups -OH, -O and -F. Subsequently, washed the treated Ti<sub>3</sub>C<sub>2</sub>T<sub>F, Cl</sub> nanosheet with deionized water several times until the pH value ≈ 6.5, and dried in a vacuum environment. Finally, the dispersion solution can be obtained by dispersing Ti<sub>3</sub>C<sub>2</sub>T<sub>F, Cl</sub> powder in chloroform.

### Perovskite lift-off process

First, PTAA layer (30 mg/ml, toluene) solution was spin coated at 4000 rpm for 25 s and annealed at 150 °C for 15 min. Then the SnO<sub>2</sub> was spin-coated at 3000 rpm for 30 s and annealed at 150 °C for 30 min. After spin coating the perovskite film, Ag electrode (~200 nm) was deposited to ensure that the perovskite film is not easy to break or fold during the stripping process. The obtained multilayer structure sample was immersed in toluene solvent at room temperature for 90 minutes. After the PTAA layer is dissolved by toluene, the perovskite/Ag film naturally separates from the substrate and floats on toluene, and again cleaned the buried interface of the perovskite with toluene to remove the remaining PTAA.

## Device fabrication

The Pero-SCs were fabricated with a configuration of ITO/SnO<sub>2</sub> or T-SnO<sub>2</sub>/perovskite/Spiro-OMeTAD/MoO<sub>3</sub>/Ag. First, ITO was cleaned with deionized water, acetone, ethanol and isopropyl alcohol with sequential ultrasonication, and each solvent was cleaned for twice. Then, treated the ITO with ultraviolet ozone for 15 min. Prepared the SnO<sub>2</sub> nanoparticles solution with the ratio SnO<sub>2</sub>: H<sub>2</sub>O (1:5, v/v), and sonicated for 30 min. Then spin-coated the SnO<sub>2</sub> nanoparticle at 3000 rpm for 30 s and annealed at 150 °C for 30 min. For T-SnO<sub>2</sub>, Ti<sub>3</sub>C<sub>2</sub>T<sub>F, Cl</sub> dissolved in chloroform was spin-coated onto the SnO<sub>2</sub> at 4000 rpm for 30 s. Perovskite films were fabricated by a two-step deposition. First, dissolved PbI<sub>2</sub> (1.5 mol/L) in DMF/DMSO (9:1, v/v) and stirred on a 70 °C heat plate for 10 h. Under a N<sub>2</sub> atmosphere, spin-coated the PbI<sub>2</sub> solution onto the SnO<sub>2</sub> films at 2000 rpm for 20 s and then annealed at 65 °C for 1 min. Then, a mixture of FAI: MAI: MACl (90 mg: 6.39 mg: 9 mg) dissolved in 1 mL IPA was dripped onto the PbI<sub>2</sub> films for 30 s, annealed the film at 150 °C for 15 min. Subsequently, the hole-transport material Spiro-OMeTAD was deposited on the upper perovskite layer at 3500 rpm for 30 s. Note that Spiro-OMeTAD was dissolved in 1 mL

chlorobenzene (72.3 mg) mixed with 17.5 mL Li-TFSI (520 mg mL<sup>-1</sup> in acetonitrile) and 28.8 mL 4-tert-butyl-pyridine (tBP). Finally, MoO<sub>3</sub> (8 nm) and Ag (100 nm) were evaporated on top of Spiro-OMeTAD.

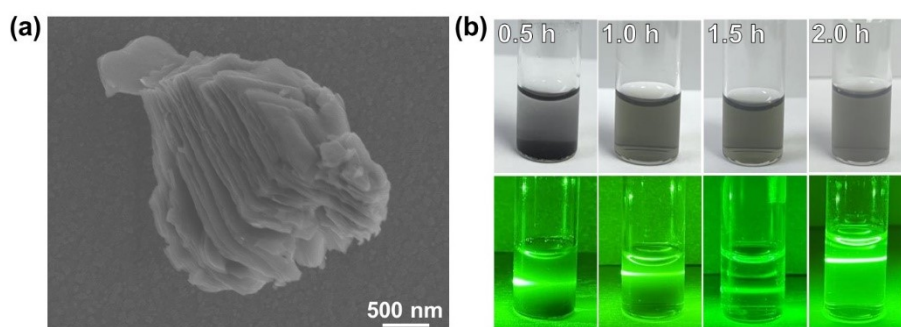
## Characterization

### Film Characterization Measurements

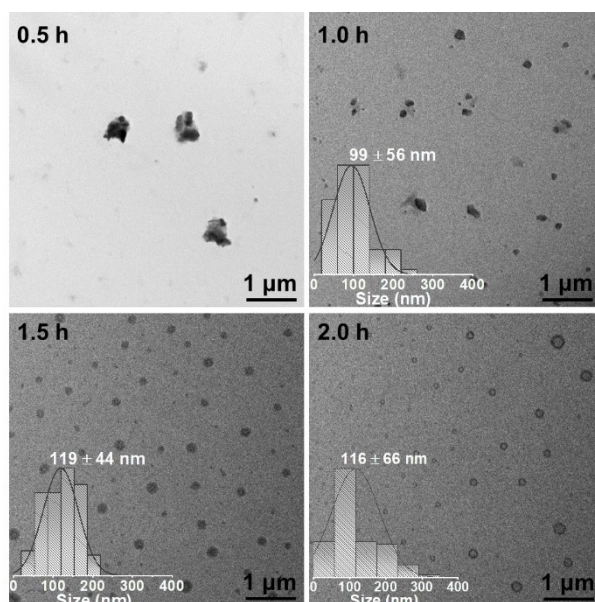
KPFM was measured with peakforce mode using PFQNE-AI conducting tips. C-AFM using TUNA mode was carried out with a bias voltage of 0.5 V. UPS measurement was collected using custom-built ultrahigh vacuum apparatus. SEM and EDS images were acquired on Hitachi SU8010 SEM, and the distribution of perovskite grain size was analyzed using a Nano measurer 1.2 software. XRD data were measured on D2 PHASER diffractometer with Cu K $\alpha$  radiation (Bruker, Germany). The TEM images were collected using JEOL JEM 2100F operated at 200 kV. GIXRD spectra were recorded by xenocs xeuss 2.0 (France). XPS measurement was conducted on a VG Scientific ESCALab 220 XL electron spectrometer with 300 W Al K $\alpha$  radiation. Fs-TA was measured with broadband capabilities and 120 fs resolution, and an ultrafast systems helios femtosecond transient absorption spectrometer equipped with UV-visible and near-infrared detectors was used to measure the perovskites samples. TRPL spectroscopy and steady-state photoluminescence PL were measured on an FLS1000 (Edinburgh Instrument, UK), the excitation wavelength was 470 nm. PL mapping was measured on a JMATE-S confocal laser scanning microscope (South Port Co., Ltd., Taiwan).

### Device Characterization Measurements

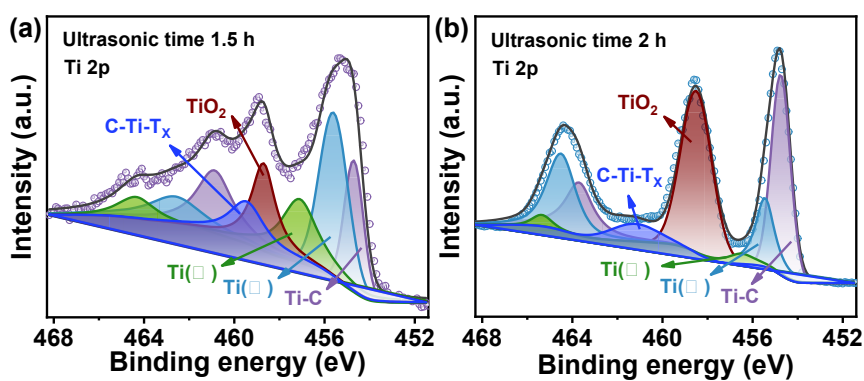
The *J-V* characteristics of Pero-SCs were measured under a simulated AM 1.5G spectrum. The external quantum efficiencies (EQEs) were measured by an Enli Technology (Taiwan) EQE measurement system. ACIS measurements were all measured in the dark at room temperature with a biased  $V_{OC}$  of the cells. The instrument is an IM6 electrochemical workstation (Zahner Zennium, Germany). The TPV and TPC spectra were measured on TPVC-1G (South Port Co., Ltd.).



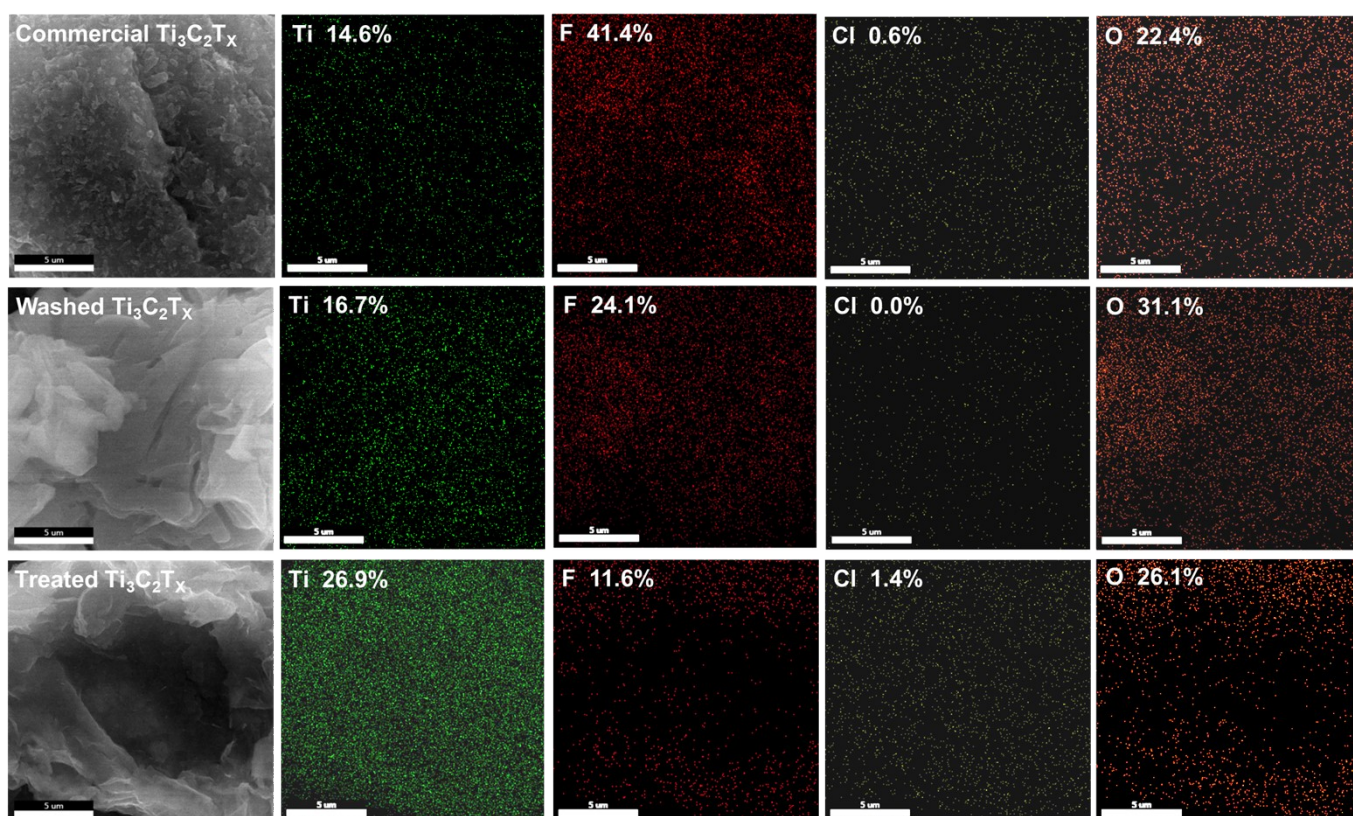
**Figure S1.** (a) SEM image of commercial MXene; (b) Tyndall phenomenon under different ultrasonic duration.



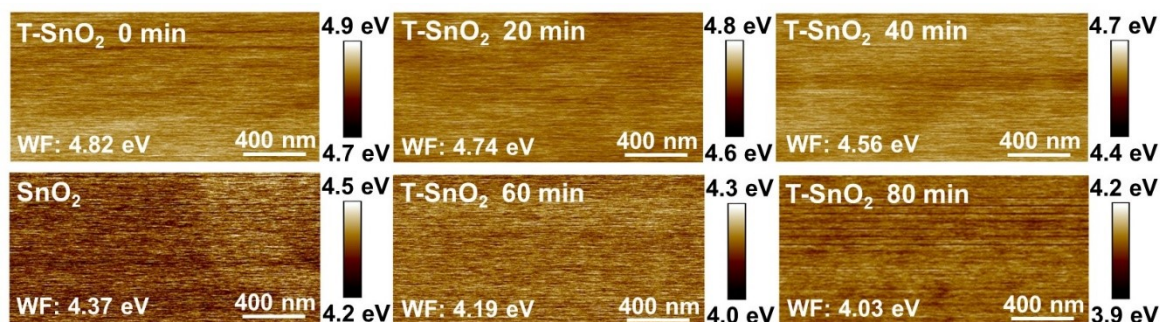
**Figure S2.** Relationship between ultrasonic time and the size of MXenes through TEM images.



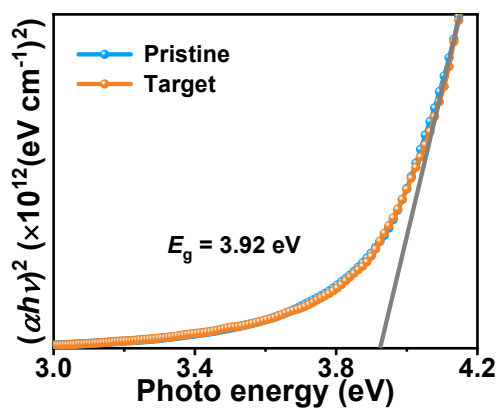
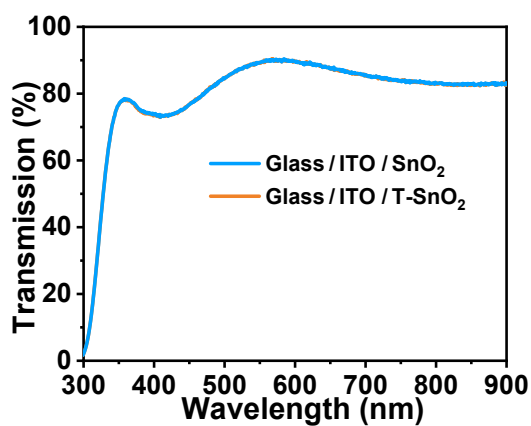
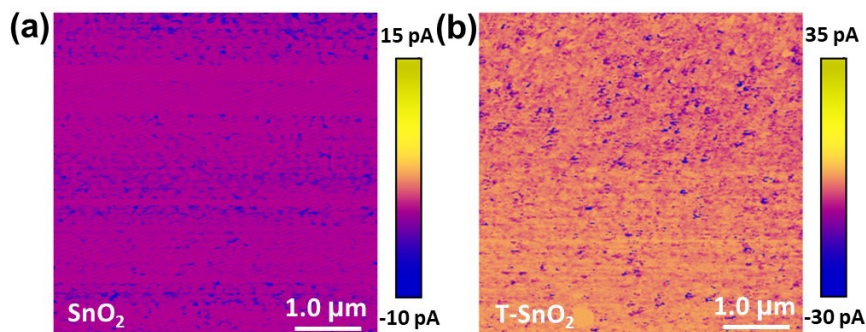
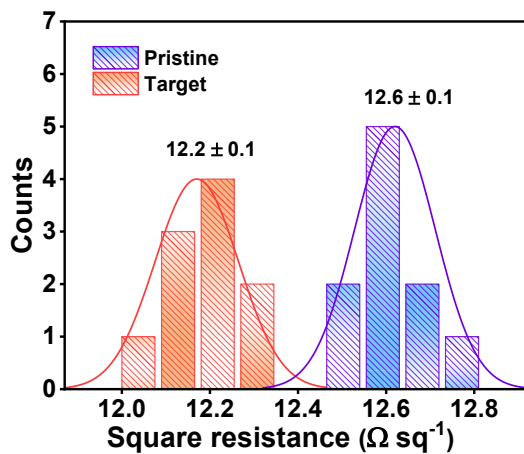
**Figure S3.** XPS spectra of  $\text{Ti}_3\text{C}_2\text{T}_x,\text{Cl}$  after (a) 1.5 hours and (b) 2 hours of ultrasonication.



**Figure S4.** EDS images of (a) commercial MXene; (b) MXene washed by deionized water; (c)  $\text{Ti}_3\text{C}_2\text{T}_x,\text{Cl}$  treated by concentrated hydrochloric acid.



**Figure S5.** Relationship between acid treatment time and the WF of  $\text{Ti}_3\text{C}_2\text{T}_x,\text{Cl}$  through KPFM images.

Figure S6. Tauc plots of SnO<sub>2</sub> and T-SnO<sub>2</sub>.Figure S7. Ultraviolet absorption spectra of SnO<sub>2</sub> and T-SnO<sub>2</sub>.Figure S8. C-AFM images of (a) SnO<sub>2</sub> and (b) T-SnO<sub>2</sub>.Figure S9. Film surface resistance of ITO/SnO<sub>2</sub> and ITO/T-SnO<sub>2</sub>.

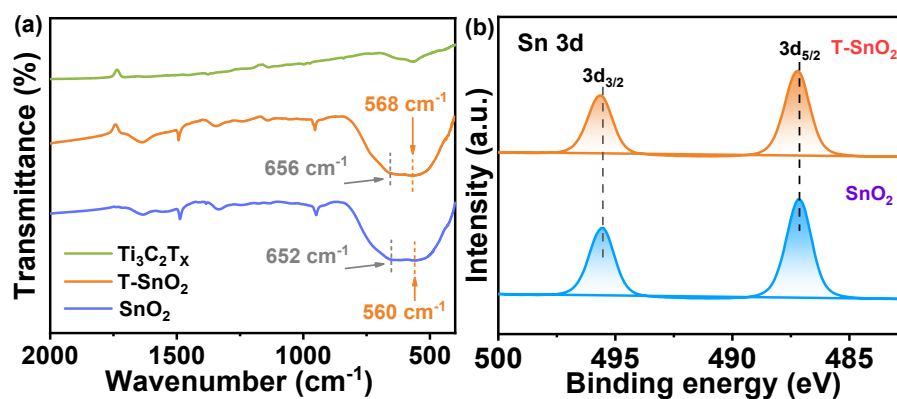


Figure S10. (a) FTIR spectra for SnO<sub>2</sub> and T-SnO<sub>2</sub>; (b) XPS spectra of Sn 3d.

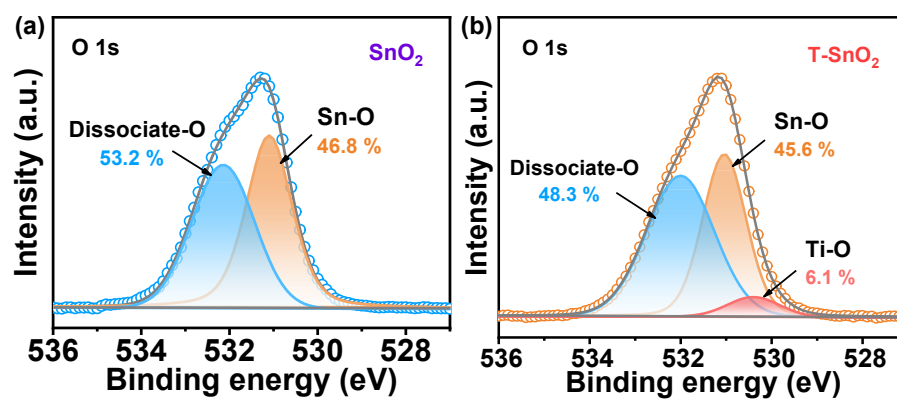


Figure S11. XPS spectra of O 1s of (a) SnO<sub>2</sub> and (b) T-SnO<sub>2</sub>.

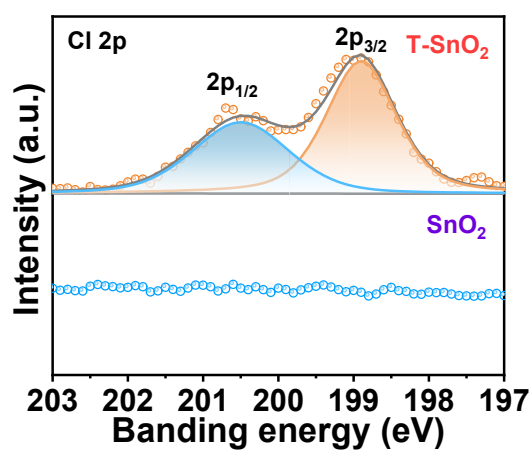


Figure S12. XPS spectra of Cl 1s.

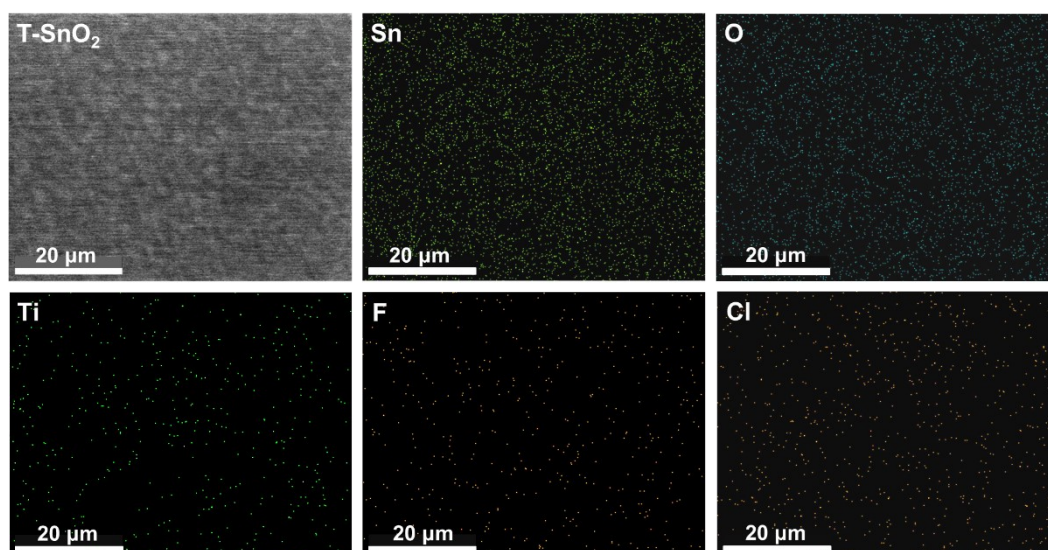


Figure S13. EDS images of T-SnO<sub>2</sub> after flushed by DMF/DMSO (v/v, 9/1).

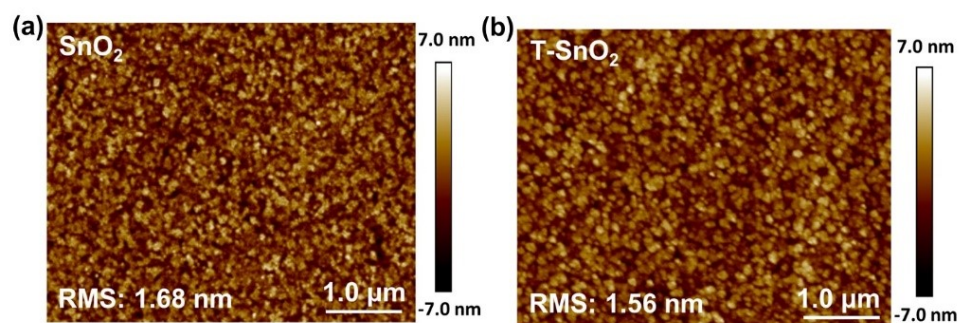


Figure S14. AFM images of (a) SnO<sub>2</sub> and (b) T-SnO<sub>2</sub>.

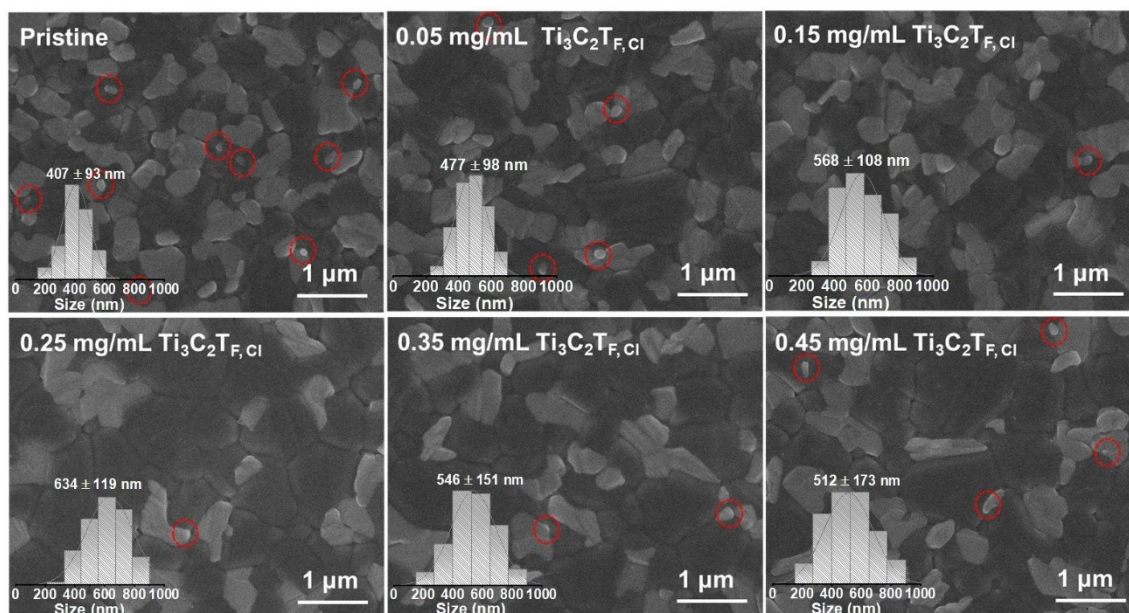


Figure S15. Top-view SEM images of perovskite film after modification with the different concentration of Ti<sub>3</sub>C<sub>2</sub>T<sub>F,Cl</sub>.

Table S1. The relationship between thickness and concentration of MXene nanosheets.

Concentration (mg/ml)	0.05	0.15	0.25	0.35	0.45
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Thickness (nm)	$2.6 \pm 0.2$	$3.6 \pm 0.2$	$5.4 \pm 0.3$	$7.5 \pm 0.4$	$9.6 \pm 0.3$
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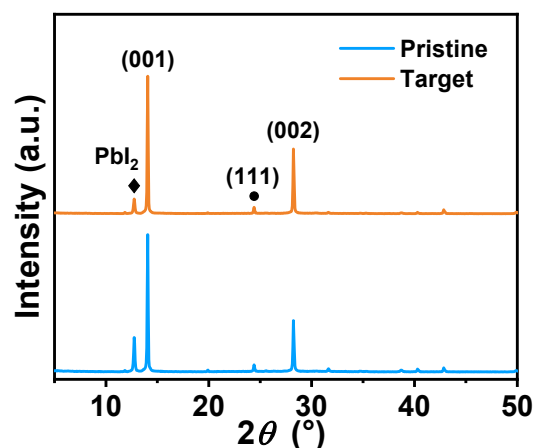


Figure S16. XRD spectra for SnO<sub>2</sub>/perovskite and T-SnO<sub>2</sub>/perovskite.

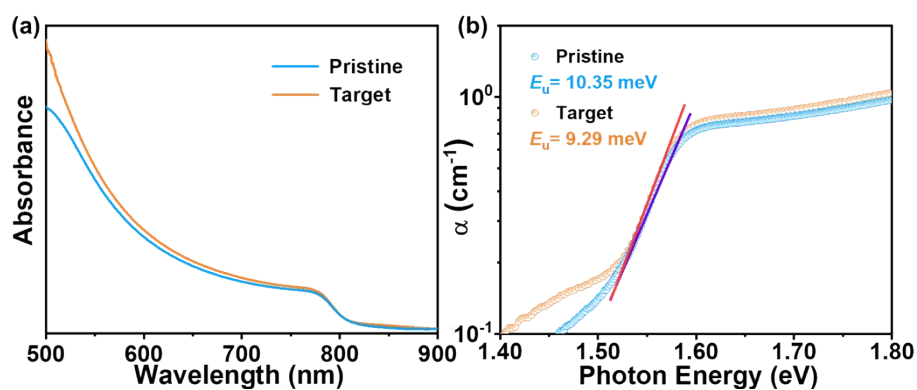


Figure S17. (a) UV-vis absorption spectra and (b) Urbach edge absorption coefficient of SnO<sub>2</sub>/perovskite and T-SnO<sub>2</sub>/perovskite.

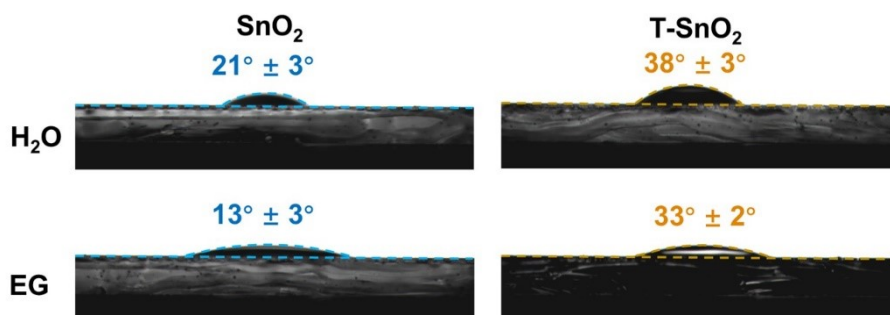


Figure S18. Water and ethylene glycol droplet contact angle measurements on SnO<sub>2</sub> and T-SnO<sub>2</sub>.

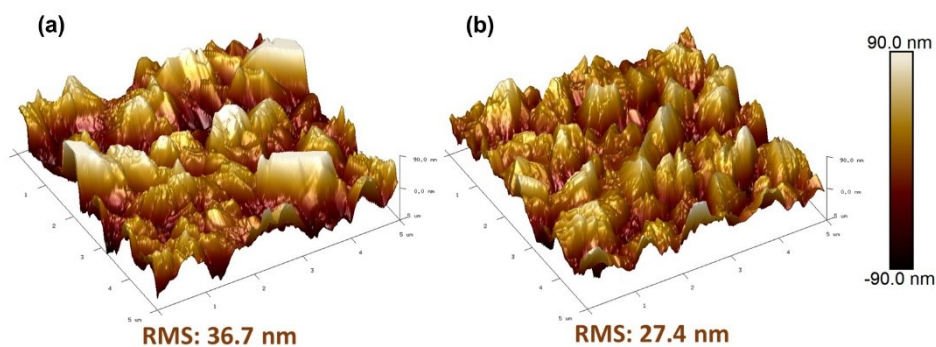
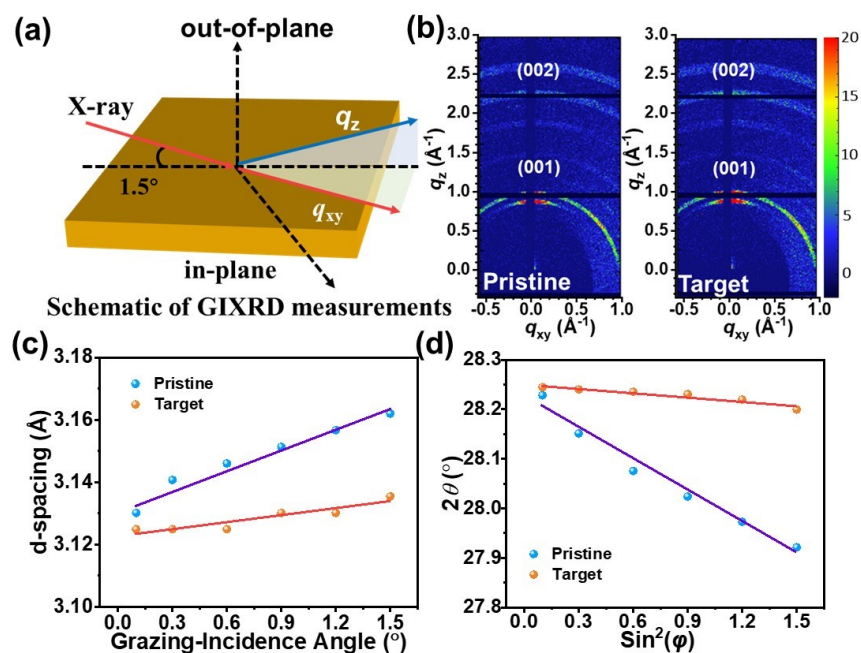
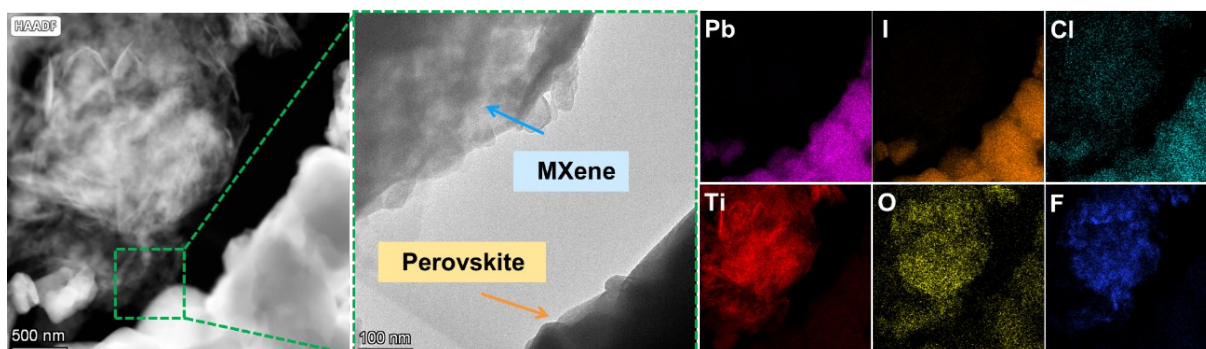


Figure S19. AFM images of (a) perovskite without Ti<sub>3</sub>C<sub>2</sub>T<sub>F,Cl</sub> and (b) perovskite with Ti<sub>3</sub>C<sub>2</sub>T<sub>F,Cl</sub>.



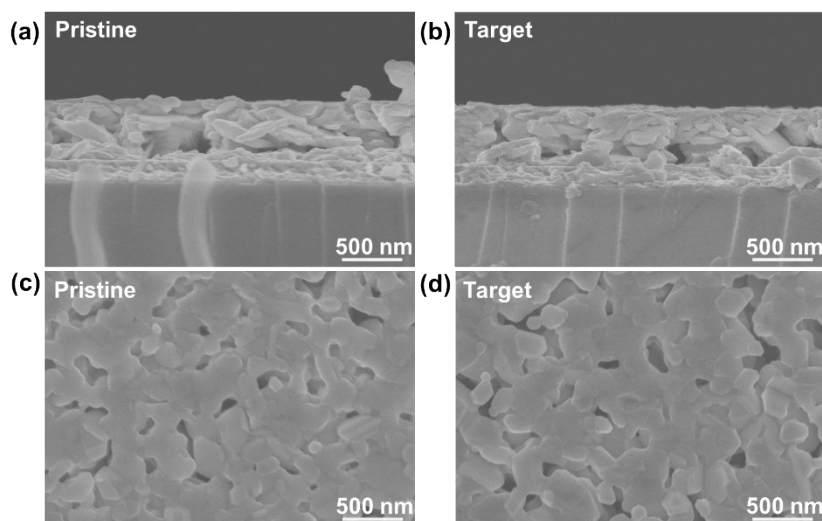
**Figure S20.** (a) Schematic of 2D GIXRD measurements; (b) 2D GIXRD images; (c) D-spacing values of the (002) plane of the perovskite films depending on the grazing incidence angle; (d) D-spacing values of the (002) plane of the perovskite films depending on the grazing incidence angle.



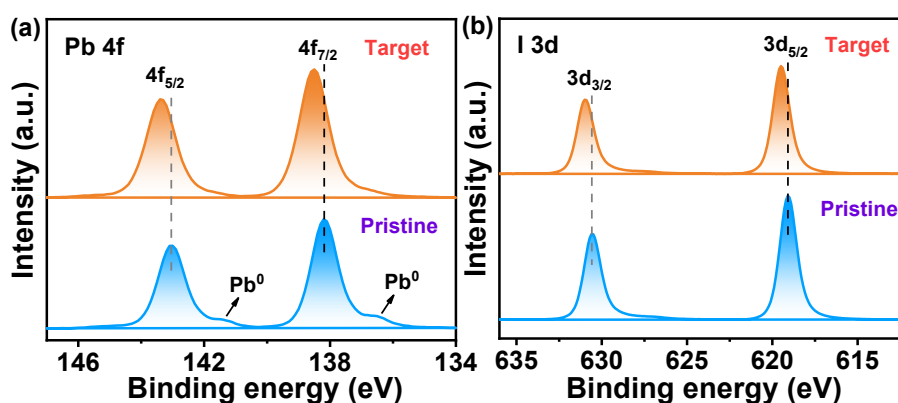
**Figure S21.** HAADF images of elements distributed on the copper wire mesh.

**Note S1.** To study whether  $\text{Ti}_3\text{C}_2\text{T}_{\text{F,Cl}}$  can affect the film quality of the  $\text{PbI}_2$  film and subsequently affect the growth of the perovskite, the top-view and cross-sectional morphologies of the  $\text{PbI}_2$  films were obtained via SEM (Figure S22). Neither the top-view and sectional SEM images showed observable difference between the pristine and target  $\text{PbI}_2$  films. Therefore, it is believed that the difference in the quality of perovskite films is due to the epitaxial growth of perovskite and the change in surface energy.



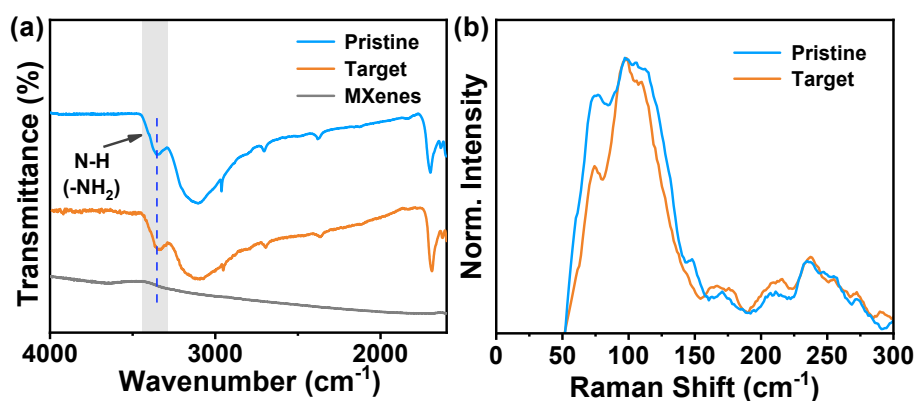


**Figure S22.** SEM images of the plane and section of the (a-c) pristine  $\text{PbI}_2$  and (b-d) target  $\text{PbI}_2$ .



**Figure S23.** XPS spectra (corrected by the peak of C 1s 284.8 eV) of (a) Pb 4f and (b) I 3d.

**Note S2.** To further prove the interaction between  $\text{Ti}_3\text{C}_2\text{T}_x\text{F}_y\text{Cl}_z$  and FAI or MAI, Raman spectroscopy was carried out (Figure S23). The  $\text{BX}_6$  octahedron shared by the perovskite ( $\text{ABX}_3$ ) is an unstable inorganic cage since the A-site cations (FA, MA) are only loosely bound to the inorganic cage by electrostatic forces. Therefore, A-site cations are able to move freely within the cage void (through translation, rotation and motion), and the vibration of the inorganic cage will show a wider frequency distribution.<sup>1</sup> Figure R17 shows the corresponding spectral region of the perovskite inorganic cage with wavenumbers less than  $300\text{ cm}^{-1}$ .<sup>2</sup> The Raman peak of the pristine sample is unevenly widened due to moving A-site cations. In contrast, because the organic cations are locked by  $\text{Ti}_3\text{C}_2\text{T}_x\text{F}_y\text{Cl}_z$  through N-H $\cdots$ F hydrogen bonds and are arranged orderly in the cage void, the Raman peak linewidth is significantly reduced; that is, the dynamic disorder is weakened.<sup>1</sup>



**Figure S24.** (a) FTIR spectra of  $\text{SnO}_2/\text{perovskite}$  (pristine) and  $\text{T-SnO}_2/\text{perovskite}$  (target); (b) Raman spectra of perovskite in the spectral range of the inorganic cage (the spectra were normalized to their maximum intensity).

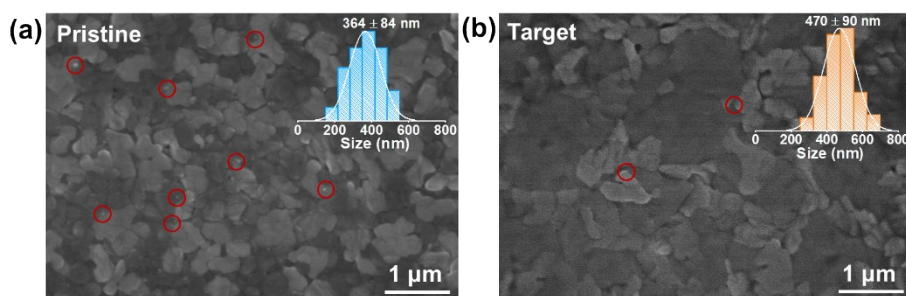


Figure S25. SEM images of the buried interface of the (a) pristine and (b) target perovskite.

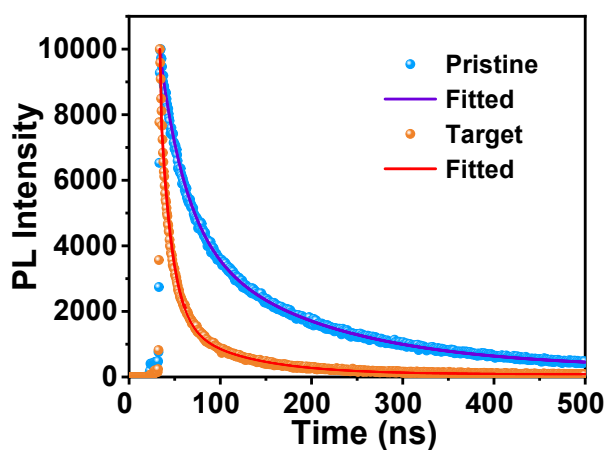


Figure S26. Time-resolved PL spectra.

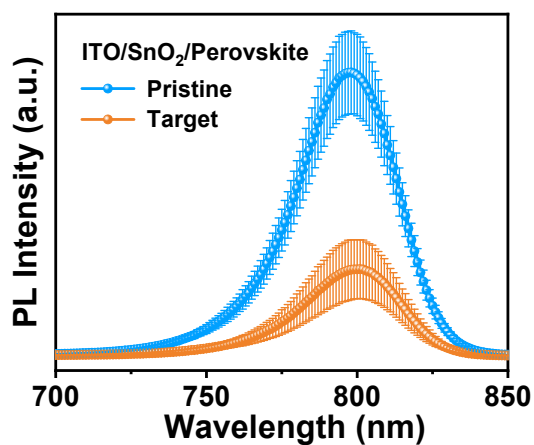


Figure S27. Steady-state PL spectra obtained by multiple curves of structure containing transport layer.

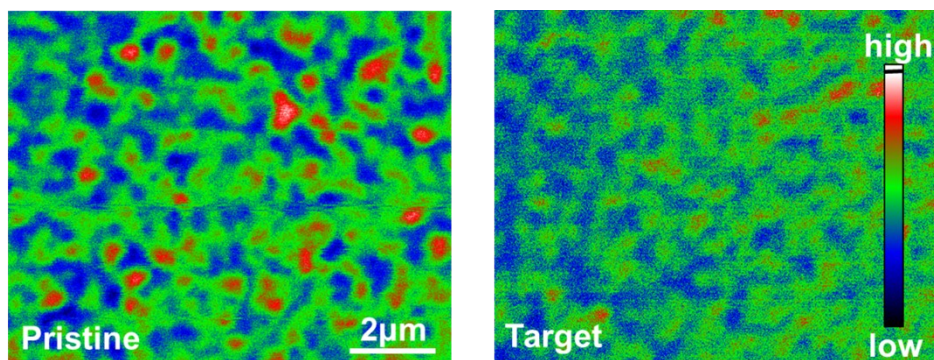


Figure S28. CLSM of the (a) pristine and (b) target.

**Table S2.** The fitting results of TAS.

Sample	$A_1$	$\tau_1$ (ps)	$A_2$	$\tau_2$ (ps)	$A_3$	$\tau_3$ (ns)
SnO <sub>2</sub> /perovskite	-0.960	3.35	-0.029	3770	-0.011	inf
T-SnO <sub>2</sub> /perovskite	-0.996	1.37	-0.002	1442	-0.002	inf

**Table S3.** The fitting results of transient photoluminescence curves.

Sample	$f_1$ (%)	$\tau_1$ (ns)	$f_2$ (%)	$\tau_2$ (ns)
SnO <sub>2</sub> /perovskite	15.68	2.19	84.32	38.91
T-SnO <sub>2</sub> /perovskite	27.72	1.98	72.28	27.63

The TRPL decay can be fitted by a biexponential equation:  $I(t) = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2}$ , where  $\tau_1$  and  $\tau_2$  are the fast and slow decay time components, respectively, and  $A_1$  and  $A_2$  are the relevant decay amplitudes.

**Table S4.** Photovoltaic parameters derived from pristine Pero-SCs, based on FA<sub>0.92</sub>MA<sub>0.08</sub>PbI<sub>3</sub>.

Number	$V_{OC}$ (V)	$J_{SC}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
1	1.09	24.07	77.55	20.42
2	1.11	23.97	77.00	20.50
3	1.11	24.36	75.82	20.59
4	1.10	23.99	77.83	20.62
5	1.10	24.53	76.67	20.73
6	1.09	24.31	78.58	20.78
7	1.10	24.33	77.67	20.81
8	1.09	24.54	78.13	20.90
9	1.11	24.48	77.04	20.95
10	1.10	24.07	79.18	21.02
11	1.09	24.54	78.60	21.12
12	1.10	24.30	78.89	21.18
13	1.11	24.01	79.46	21.28
14	1.11	24.27	78.87	21.30
15	1.10	24.45	79.18	21.38
16	1.12	24.14	79.17	21.44
17	1.12	24.29	79.01	21.53
18	1.12	24.48	78.39	21.59
19	1.11	24.54	79.51	21.71
20	1.12	24.47	78.91	21.77
21	1.12	24.48	79.66	21.89
22	1.12	24.50	79.71	21.96
23	1.11	25.24	78.48	22.01
24	1.13	25.36	76.37	22.03
25	1.12	25.01	78.99	22.18
26	1.12	25.11	78.86	22.20
27	1.12	25.20	78.83	22.24

28	1.13	24.75	80.02	22.42
29	1.12	25.01	79.9	22.42
30	1.12	25.23	79.24	22.45

**Table S5.** Photovoltaic parameters derived from target Pero-SCs, based on  $\text{FA}_{0.92}\text{MA}_{0.08}\text{PbI}_3$ .

Number	$V_{\text{OC}}$ (V)	$J_{\text{SC}}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
1	1.17	24.47	82.75	23.63
2	1.17	24.56	82.42	23.71
3	1.18	24.88	80.79	23.80
4	1.16	24.58	83.26	23.87
5	1.17	24.84	82.10	23.91
6	1.17	25.37	80.76	23.95
7	1.17	25.02	81.80	24.02
8	1.16	25.22	82.34	24.07
9	1.16	25.25	82.29	24.09
10	1.16	24.74	83.85	24.10
11	1.16	25.32	82.19	24.15
12	1.17	25.22	81.82	24.17
13	1.17	25.12	82.14	24.20
14	1.17	25.11	82.54	24.24
15	1.17	25.26	81.73	24.25
16	1.16	25.18	83.25	24.33
17	1.16	25.10	83.47	24.35
18	1.18	25.47	81.03	24.36
19	1.16	25.10	83.42	24.42
20	1.18	25.39	81.83	24.43
21	1.18	25.37	81.63	24.45
22	1.17	25.38	82.55	24.52
23	1.17	25.00	83.79	24.59
24	1.17	25.47	82.65	24.67
25	1.17	25.91	81.70	24.73
26	1.17	25.35	83.69	24.88
27	1.17	25.50	83.57	24.95
28	1.17	25.71	83.30	25.03
29	1.17	25.95	82.88	25.07
30	1.18	25.81	82.74	25.09

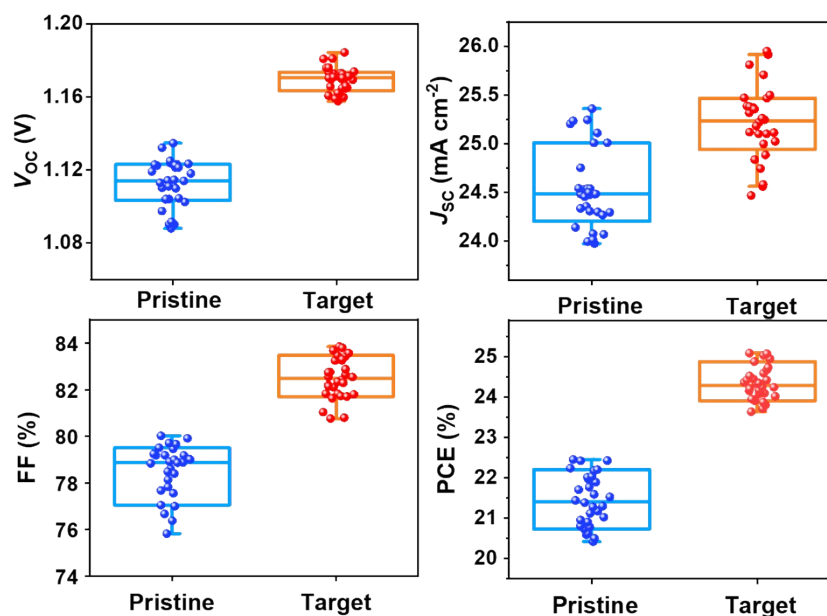


Figure S29. Device performance distribution from 30 cells.

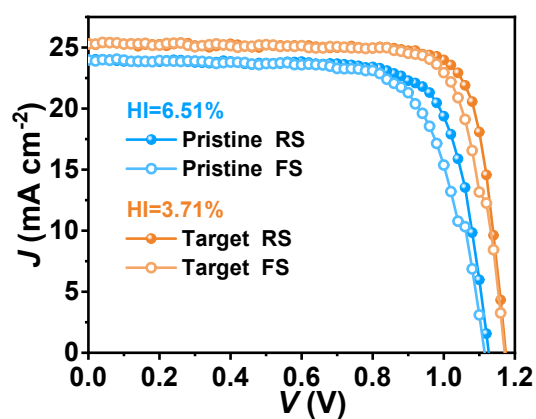


Figure S30. Hysteresis investigation of the Pero-SCs.

Table S6. Hysteresis investigation of the Pero-SCs.

Devices		$V_{oc}$ (V)	$J_{sc}$ ( $\text{mA cm}^{-2}$ )	FF (%)	PCE (%)	$h$ (%)
Pristine	RS	1.13	23.80	76.23	20.43	6.51
	FS	1.11	23.92	71.64	19.10	
Target	RS	1.17	25.33	80.57	23.96	3.71
	FS	1.17	25.22	78.13	23.07	

The semiempirical hysteresis factor ( $h$ ) is obtained from equation:  $h = (PCE_{RS} - PCE_{FS}) / PCE_{RS}$ , the "FS" is forward scanning and the "RS" is reverse scanning.

Table S7. Parameters fitted from the Nyquist plots for Pero-SCs based on  $\text{SnO}_2$  and T- $\text{SnO}_2$ .

ETL	$R_s$ ( $\Omega \text{ cm}^{-2}$ )	$R_{tr}$ ( $\Omega \text{ cm}^{-2}$ )
$\text{SnO}_2$	26.9	101.4
T- $\text{SnO}_2$	27.7	47.1

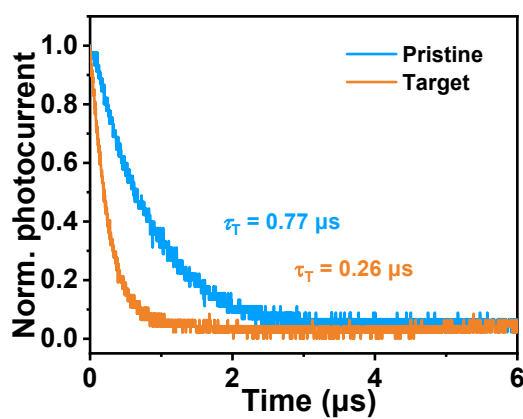


Figure S31. The transient photocurrent measurement in the dark.

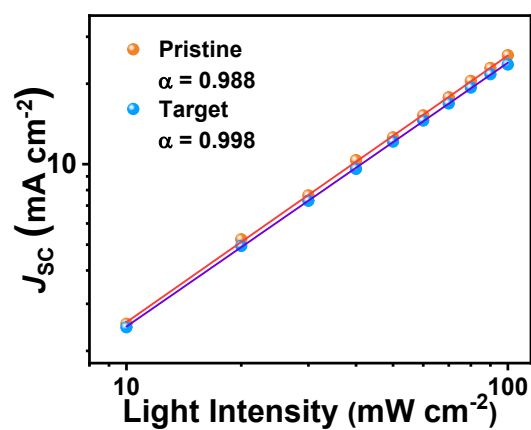


Figure S32.  $J_{sc}$  versus light intensity of the Pero-SCs.

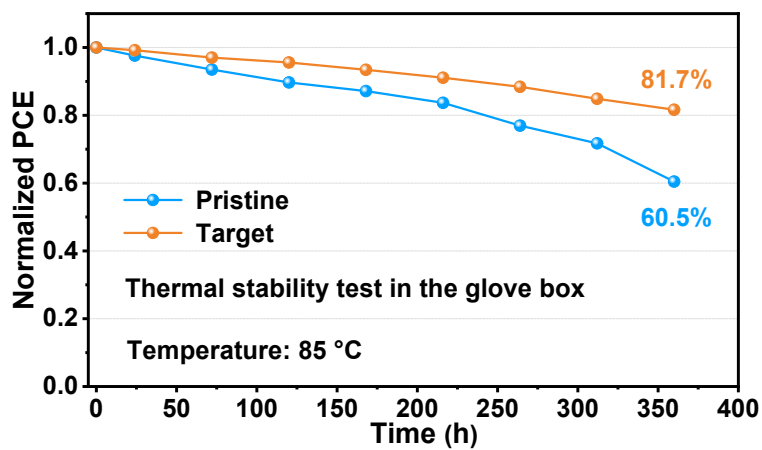
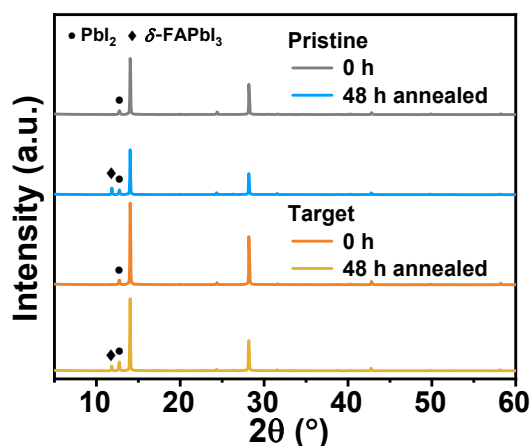


Figure S33. The thermal stability (85 °C) of Pero-SCs in N<sub>2</sub>.



**Figure S34.** XRD Spectra of the thermal stability (85 °C, in N<sub>2</sub>) of perovskite film.

**Table S8.** The comparison of FF and PCE for the previous studies based on the MXenes modification of *n-i-p* Pero-SCs.

Materials/Methods	Device structure	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Year <sup>Ref</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /ETL	ITO/SnO <sub>2</sub> /MAPbI <sub>3</sub> /Spiro-OMeTAD/Ag	1.08	22.63	70.0	17.17	2019 <sup>3</sup>
HO-Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /ETL	ITO/SnO <sub>2</sub> /MAPbI <sub>3</sub> /Spiro-OMeTAD/Ag	1.07	23.11	74.0	18.29	2021 <sup>4</sup>
O-Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /ETL	FTO/SnO <sub>2</sub> /MAPbI <sub>3</sub> /Spiro-OMeTAD/Ag	1.07	23.88	78.4	20.09	2021 <sup>5</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> -SWCNTs/ ETL interface	ITO/SnO <sub>2</sub> /FAMAPbI <sub>3</sub> /Spiro-OMeTAD/Au	1.07	25.09	80.0	21.42	2021 <sup>6</sup>
Nb <sub>2</sub> CT <sub>x</sub> /ETL additive	ITO/SnO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/MoO <sub>3</sub> /Au	1.14	25.29	79.5	22.86	2021 <sup>7</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> MQDs/ ETL additive	ITO/SnO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/MoO <sub>3</sub> /Au	1.17	24.96	79.8	23.34	2021 <sup>8</sup>
T-Nb <sub>2</sub> CT <sub>x</sub> /ETL and perovskite additive	ITO/SnO <sub>2</sub> /FA <sub>0.85</sub> Cs <sub>0.15</sub> PbI <sub>3</sub> /Spiro-OMeTAD/Ag	1.12	25.07	77.4	21.79	2022 <sup>9</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> -FOTS/ ETL additive	ITO/SnO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/MoO <sub>3</sub> /Ag	1.09	25.22	81.1	22.36	2022 <sup>10</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /ETL additive	ITO/SnO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/Au	1.13	25.07	81.1	23.07	2022 <sup>11</sup>
Nb <sub>2</sub> CT <sub>x</sub> /Dual interfacial	FTO/SnO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/Ag	1.25	23.70	79.9	23.68	2022 <sup>12</sup>
Ti <sub>3</sub> C <sub>2</sub> F <sub>x</sub> /anti-solvent	FTO/TiO <sub>2</sub> /FAPbI <sub>3</sub> /Spiro-OMeTAD/Au	1.13	25.84	82.6	24.17	2022 <sup>13</sup>
Nb <sub>2</sub> O <sub>5</sub> -Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> / ET bilayer	FTO/TiO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/Au	1.07	24.56	73.9	19.46	2023 <sup>14</sup>
V <sub>2</sub> CT <sub>x</sub> /anti-solvent	ITO/SnO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/Ag	1.18	25.03	79.8	23.47	2023 <sup>15</sup>
Au@Nb <sub>2</sub> CT <sub>x</sub> / ETL interface	FTO/SnO <sub>2</sub> /CsFAMAPbI <sub>3</sub> /Spiro-OMeTAD/Au	1.22	24.28	80.6	23.78	2023 <sup>16</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> /ETL additive	ITO/TiO <sub>2</sub> /FAPbI <sub>3</sub> /Spiro-OMeTAD/MoO <sub>3</sub> /Au	1.18	25.58	81.7	24.63	2023 <sup>17</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>F,Cl</sub> / ETL interface	ITO/SnO <sub>2</sub> /FA <sub>0.92</sub> MA <sub>0.08</sub> PbI <sub>3</sub> /Spiro-OMeTAD/MoO <sub>3</sub> /Ag	1.18	24.46	83.3	24.05	This work
	ITO/SnO <sub>2</sub> /FA <sub>0.85</sub> MA <sub>0.15</sub> PbI <sub>2.55</sub> Br <sub>0.45</sub> /Spiro-OMeTAD/MoO <sub>3</sub> /Ag	1.18	25.81	82.7	25.09	

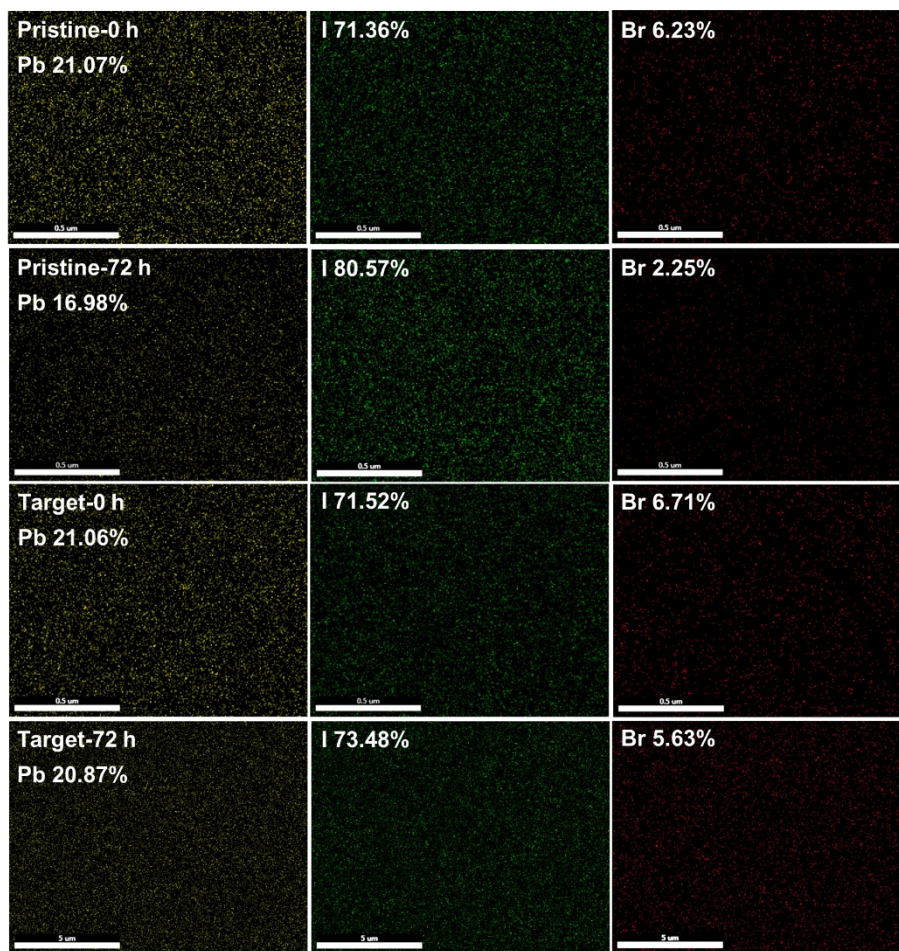


Figure S35. EDS of the mixed halide (I/Br) perovskite before and after heated at 85 °C for 72 h.

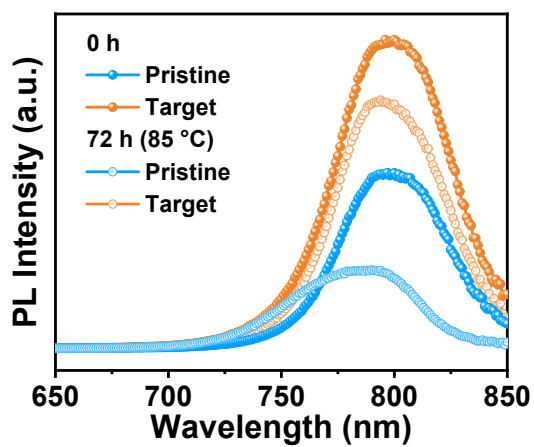
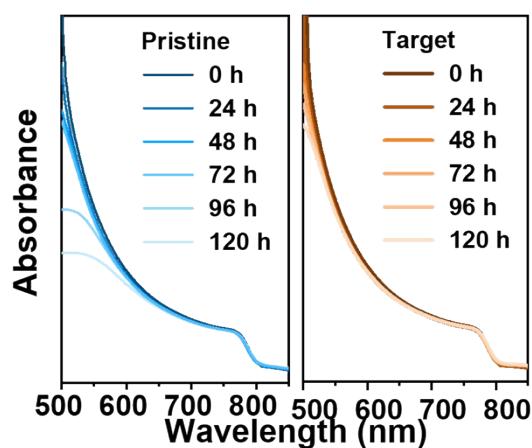


Figure S36. Steady-state PL spectra of the mixed halide (I/Br) perovskite film before and after heated at 85 °C for 72 h.





**Figure S37.** UV-vis spectra of the mixed halide (I/Br) perovskite film before and after heated at 85 °C for 120 h.

**Table S9.** Photovoltaic parameters of the 1.1 cm<sup>2</sup> devices.

Devices	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
Pristine	1.11	25.22	65.45	18.28
Target	1.18	25.48	68.34	20.62

**Table S10.** Photovoltaic parameters of the Pero-SCs based on FA<sub>0.85</sub>MA<sub>0.15</sub>PbI<sub>2.55</sub>Br<sub>0.45</sub>.

Devices		$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
Pristine	Best	1.14	24.41	80.56	22.38
	Average	1.12 ± 0.02	23.99 ± 0.41	78.92 ± 0.96	21.43 ± 0.46
Target	Best	1.18	24.46	83.28	24.05
	Average	1.16 ± 0.01	24.17 ± 0.35	83.04 ± 0.89	23.30 ± 0.41

**Table S11.** Photovoltaic parameters derived from pristine Pero-SCs, based on FA<sub>0.85</sub>MA<sub>0.15</sub>PbI<sub>2.55</sub>Br<sub>0.45</sub>.

Number	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
1	1.10	23.61	80.17	20.90
2	1.14	23.32	78.89	20.96
3	1.10	23.91	79.70	20.99
4	1.09	24.48	78.55	21.04
5	1.10	24.05	79.47	21.05
6	1.15	23.23	79.07	21.06
7	1.15	23.45	78.34	21.06
8	1.13	24.21	76.96	21.08
9	1.13	24.46	76.74	21.14
10	1.13	23.80	78.62	21.19
11	1.11	24.17	79.00	21.20
12	1.12	24.09	78.56	21.26
13	1.15	23.34	79.35	21.27
14	1.14	24.24	77.3	21.30
15	1.13	23.49	80.12	21.33
16	1.14	23.83	78.63	21.38

17	1.13	24.19	78.32	21.40
18	1.13	24.29	79.07	21.66
19	1.15	24.03	78.56	21.78
20	1.12	24.30	79.90	21.82
21	1.15	23.69	80.00	21.86
22	1.15	24.56	78.44	22.18
23	1.15	24.29	79.58	22.25
24	1.15	24.46	79.22	22.30
25	1.14	24.41	80.56	22.38

**Table S12.** Photovoltaic parameters derived from target Pero-SCs, based on  $\text{FA}_{0.85}\text{MA}_{0.15}\text{PbI}_{2.55}\text{Br}_{0.45}$ .

Number	$V_{\text{OC}}$ (V)	$J_{\text{SC}}$ ( $\text{mA cm}^{-2}$ )	FF (%)	PCE (%)
1	1.14	24.40	81.32	22.65
2	1.15	24.25	81.39	22.72
3	1.15	24.55	80.88	22.77
4	1.16	23.54	83.44	22.84
5	1.17	23.43	83.37	22.89
6	1.16	23.86	82.80	22.92
7	1.15	24.22	82.43	22.97
8	1.17	23.76	82.86	23.02
9	1.16	23.86	83.15	23.06
10	1.14	24.06	84.16	23.10
11	1.14	24.03	84.52	23.15
12	1.16	24.03	83.20	23.19
13	1.16	24.01	83.31	23.20
14	1.16	23.94	84.13	23.29
15	1.17	23.76	83.94	23.33
16	1.16	24.04	83.79	23.41
17	1.16	24.35	83.32	23.48
18	1.15	24.55	83.51	23.55
19	1.16	24.52	83.38	23.64
20	1.18	24.54	82.0	23.72
21	1.16	24.59	83.15	23.77
22	1.17	24.32	83.32	23.79
23	1.17	24.63	82.96	23.92
24	1.19	24.49	82.3	23.95
25	1.18	24.46	83.28	24.05

**Table S13.** Processing methods of MXenes terminals in Pero-SCs.

Materials	Methods	$V_{\text{OC}}$ (V)	$J_{\text{SC}}$ ( $\text{mA cm}^{-2}$ )	FF (%)	PCE (%)	Year <sup>Ref</sup>
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Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> (T=O, F)	UV-ozone treatments Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	1.08	22.63	70.0	17.17	2019 <sup>3</sup>
Ti <sub>3</sub> C <sub>2</sub> Cl <sub>x</sub>	Lewis acidic CdCl <sub>2</sub> melts etching	1.70	7.87	82.7	11.08	2021 <sup>18</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> (T=O, F, OH)	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> aqueous solution at 50 °C under stirring in air	1.07	23.11	74.0	18.29	2021 <sup>4</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> (T=O)	Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> sintered at 500 °C for 30 min in air	1.07	23.88	78.4	20.09	2021 <sup>5</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> -MQDs (T=S, N)	Add Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub> and NH <sub>3</sub> ·H <sub>2</sub> O to Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> in autoclave at 150 °C for 12 h	1.17	24.96	79.8	23.34	2021 <sup>8</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> (T=AC)	Added Pb(CH <sub>3</sub> COO) <sub>2</sub> to Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	1.29	14.96	80.3	15.48	2022 <sup>11</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> -MQDs (T=F)	Add Ti <sub>3</sub> AlC <sub>2</sub> to HF stir at room temperature for 3 days, and sonicate Ti <sub>3</sub> C <sub>2</sub> F <sub>x</sub> for 30 min at power of 600 W	1.22	20.59	81.6	20.44	2022 <sup>19</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> (T=F/Cl/Br/I)	Add Ti <sub>3</sub> AlC <sub>2</sub> to HF acid or molten salts	1.13	25.84	82.6	24.17	2022 <sup>13</sup>
Ti <sub>3</sub> C <sub>2</sub> Cl <sub>x</sub>	Etch the Al layer in the Ti <sub>3</sub> AlC <sub>2</sub> by ZnCl <sub>2</sub>	1.29	11.56	69.9	10.43	2023 <sup>20</sup>
Ti <sub>3</sub> C <sub>2</sub> T <sub>F, Cl</sub>	Add Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> to HCl (9 mol/ml) and stirred for 20, 40, 60 and 80 min at room temperature in air	1.18	25.81	82.7	25.09	This work

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