Supporting information for

Accelerating hole extraction by inserting 2D Ti_3C_2-MXene interlayer to all inorganic perovskite solar cells with long-term stability

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Experimental Section

Synthesis of MXene:

1 g raw Ti_3AlC_2 powders (purchased from Jilin 11 technology Co., Ltd.) were added slowly into 20 ml HF (40%) solution and stirred for 24 h at room temperature. Then the colloidal solution was washed with deionized water for several times until the PH was above 5. The obtained powder was dried under vacuum at 60 ^oC for 12 h. In order to delaminate the $Ti_3C_2T_x$, 0.5 g $Ti_3C_2T_x$ powders were added into 10 ml 25% tetramethylammonium hydroxide aqueous solution (TMAOH) solution stirring for 24 h. Then the mixture was washed and centrifuged by isopropanol three times. In order to prepare few-layer MXene nanosheets, 100 mg delaminated $Ti_3C_2T_x$ powder was poured into 50 ml isopropanol (≥99.7%, Sinopharm Chemical Reagent Co., Ltd.) and

sonicated for 4 h under Ar atmosphere. Finally, after centrifugation for 1 h at 3500 rpm, a light green supernatant solution was obtained (0.4 mg ml^{-1}).

Device fabrication:

The FTO substrates were first washed in deionized water (containing detergent) by sonication for 20 min. Then the substrates were cleaned by acetone, ethyl alcohol and deionized waterfor 30 min in sequence. The electron transport layer was prepared by spin-coated TiO_2 on the dried FTO substrates with a speed of 3000 rpm, 30s, and then moved to the furnace to heat. The temperature kept 500 ^oC for 30 min in the open air. The inorganic $CsPbBr_3$ film was prepared in the vacuum system with a vacuum degree of ~10^{-4} Pa. The CsBr was first deposited on the FTO/c-TiO_2 substrates with a speed of ~0.4 Å/s. The $PbBr_2$ film was coated on the top of CsBr film by vapor deposition at a speed of ~0.5 Å/s. Then the samples were annealed at 350 ^oC for 30 min in the air. The Mxene solution (2 mg ml^{-1}) was then spin-coated on the sample at a speed of 2500 rpm and treated at 100 ^oC for 5 min for two times. After repeat it for three times, the commercial carbon ink was coated on the surface of MXene layer and treated at 80^oC for several hours. The active areas of devices are 0.09 cm^2.

Characterization:

X-ray diffraction (XRD) patterns of inorganic perovskite films were identified with a Cu Kα radiation (D/MAX2500V, Japan) from 10^o to 60^o. Absorption spectra were performed by a UV-visible spectrophotometer (UV-2550). The interface component of film was conducted by the X-ray photoelectron spectra (XPS, Thermo ESCALAB 250). The ultraviolet photoemission spectroscopy (UPS) was conducted by an X-ray photoelectron spectrometer (XPS-AXIS Ultra HAS, Kratos) equipped with monochromatic Al-Kα = 1486.6 eV and nonmonochromatic He-Iα = 21.22 eV sources. The surface morphologies and cross section of inorganic perovskite films were performed by a field emission scanning electron microscopy (FE-SEM, Sigma Zeiss). Time-resolved photoluminescence (TRPL) decay tests were conducted by EI-FLS980-TCSPC (Edinburgh Instruments, UK) using 375 nm laser as the excitation light source. The space-charge limited current (SCLC) data were acquired with a semiconductor system equipped with probe station and Keithley 4200-SCS.

Electrochemical impedance spectroscopy (EIS) were carried out by using the electrochemical workstation (Zahner, Germany) with the frequency range from 0.1 Hz to 10^5 Hz under a bias voltage (10 mV) in the dark condition. J-V curves were measured on a Keithley 2636 system sourcemeter equipped with a Xenon lamp solar simulator (Zolix). The light intensity was~100 mW cm^{-2}. The spectral response was collected on the EQE measurement system (SCS100, Zolix). All the tests were carried out in the open air with RH~45%.

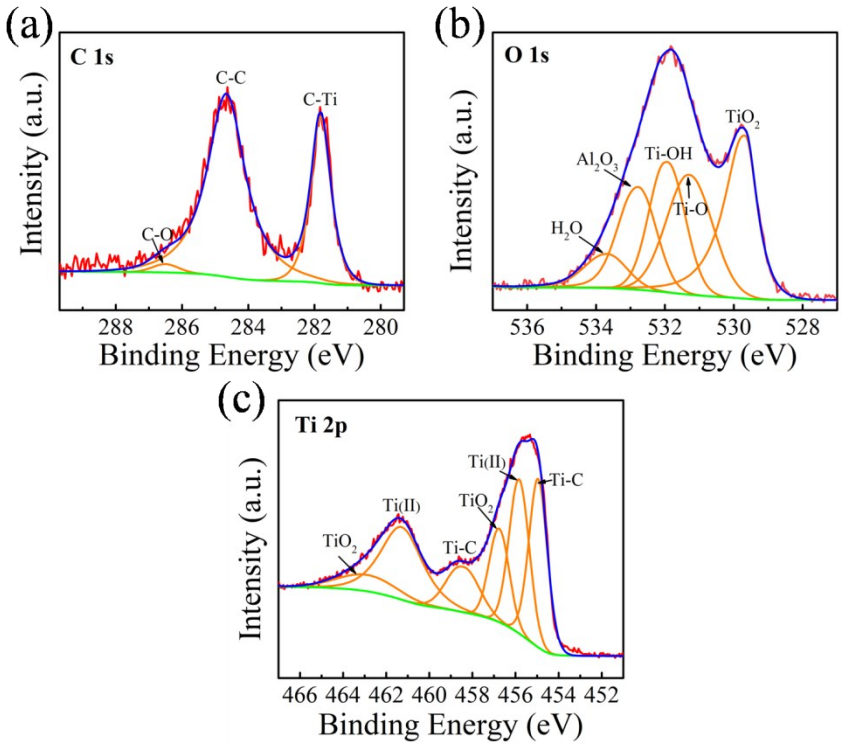

Fig. S1 (a) C 1s, (b) O 1s and (c) Ti 2p XPS spectra of Ti_3C_2-MXene.

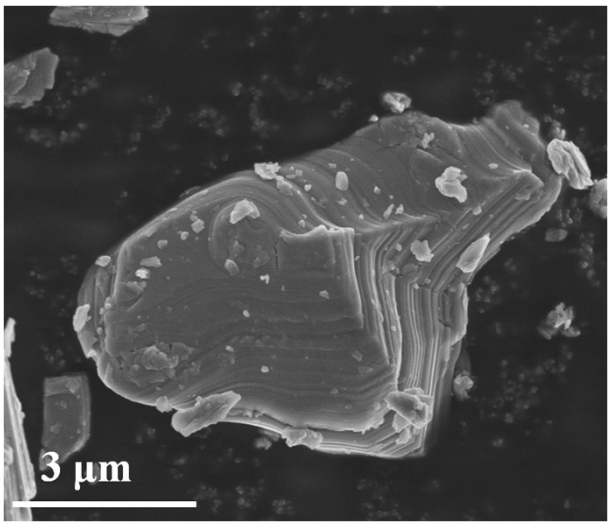

Fig. S2 Top-view SEM image of raw Ti_3AlC_2 powder.

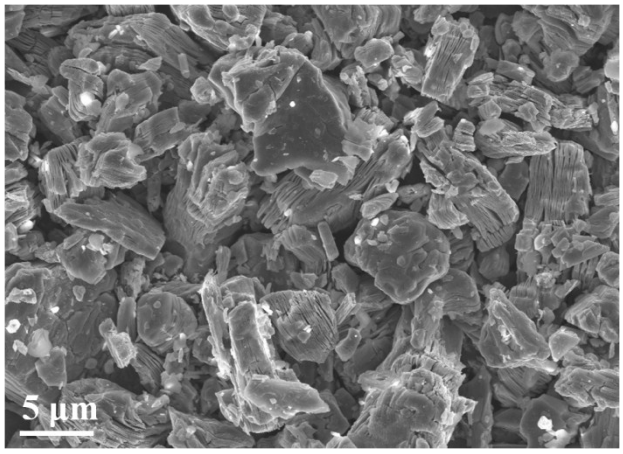

Fig. S3 Top-view SEM image of Ti_3C_2-MXene nanosheets.

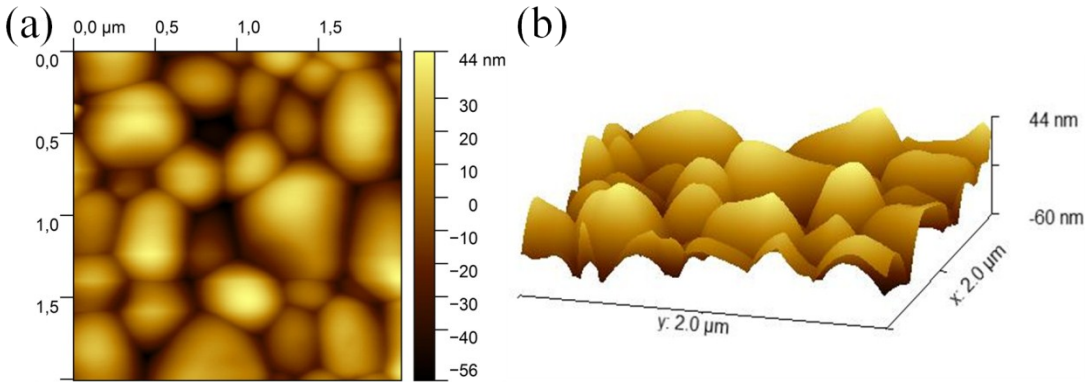

Fig. S4 (a) 2D and (b) 3D AFM image of the $CsPbBr_3$ film.

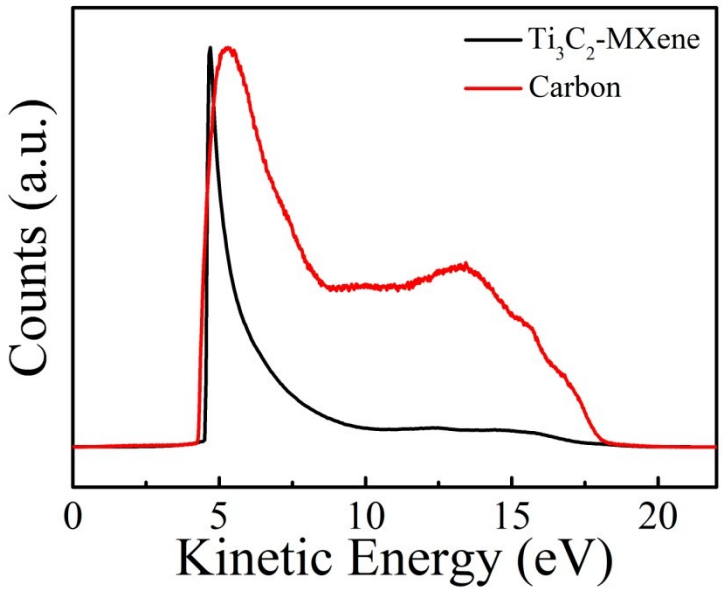

Fig. S5 The survey of UPS spectra of Ti_3C_2-MXene and carbon.

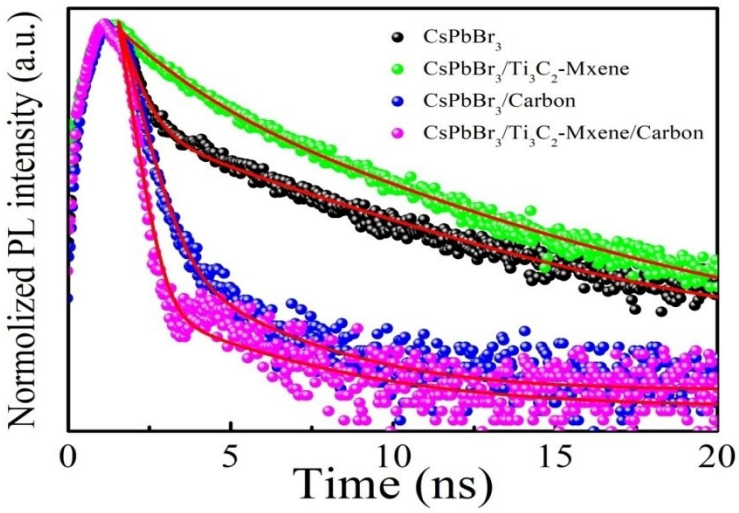

Fig. S6 TRPL curves of the $CsPbBr_3$ film with/without Ti_3C_2-MXene and carbon

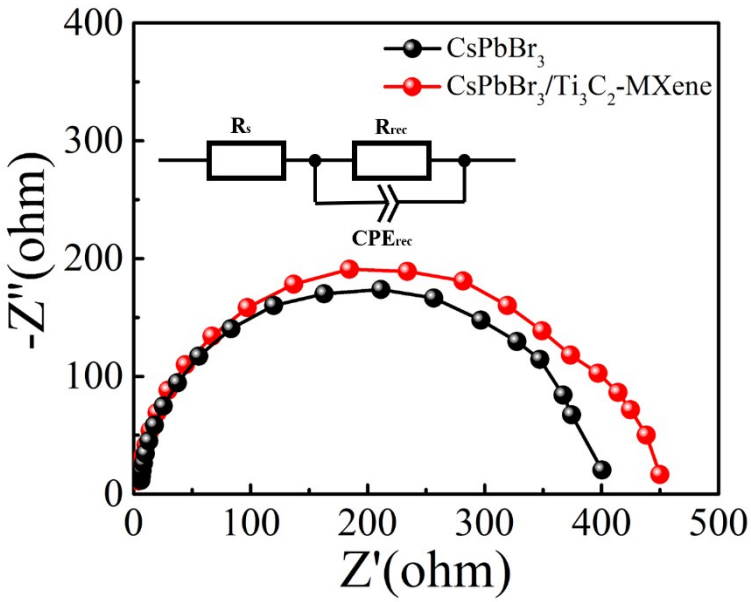

Fig. S7 Nyquist plots of inorganic PSCs with and without Ti_3C_2-MXene.

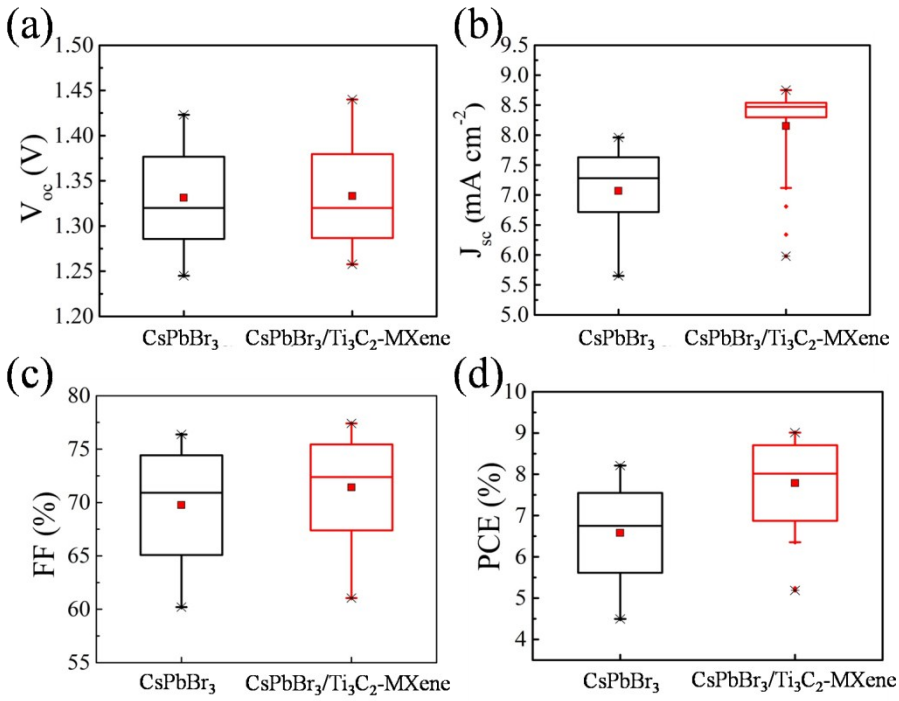

Fig. S8 Photovoltaic parameter distributions of $CsPbBr_3$ PSCs with/without MXene.

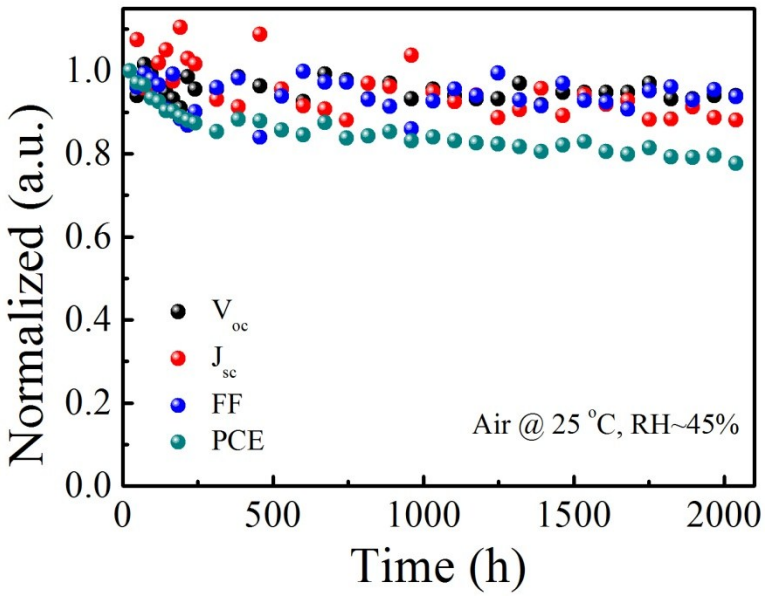

Fig. S9 Normalized V_{oc}, J_{sc}, FF and PCE stability of the $CsPbBr_3/Ti_3C_2$-MXene PSCs in the air with humidity of ~45%.

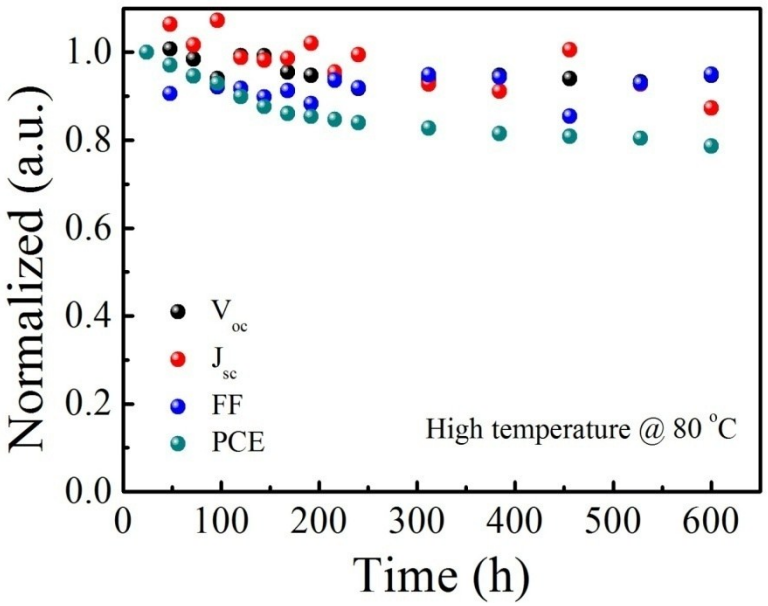

Fig. S10 Normalized V_{oc}, J_{sc}, FF and PCE stability of the $CsPbBr_3/Ti_3C_2$-MXene PSCs at the high temperature condition @ 80°C.

Table S1 The time coefficients and relative magnitude of PL decay trace under 375 nm excitation wavelength.

Sample	τ_1 (ns)	τ_2 (ns)	A_1 (%)	A_2 (%)	$\tau_{ave.}$ (ns)
$CsPbBr_3$	0.318	3.555	29.46	70.54	3.438
$CsPbBr_3/Ti_3C_2$-MXene	0.534	5.746	26.13	73.87	5.582
$CsPbBr_3$/C	0.518	3.078	85.08	14.32	2.877
$CsPbBr_3/Ti_3C_2$-MXene/C	0.333	4.103	83.08	16.92	1.921

Table S2 Photovoltaic parameters of $CsPbBr_3$ PSCs with/without MXene.

Sample	R_s (ohm)	R_{rec} (ohm)	CPE-Yo (S-sec^n/cm^2)	CPE-n (S-sec^n/cm^2)	Chsq
$CsPbBr_3$	5.226	391.5	2.126E-7	0.9653	5.74E-4
$CsPbBr_3/Ti_3C_2$-MXene	2.396	454.8	3.573E-7	0.9275	1.70E-3

Table S3 Photovoltaic parameters of $CsPbBr_3$ PSCs with/without MXene.

Device		V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
$CsPbBr_3$	Average	1.334±0.089	6.805±1.155	68.285±8.085	6.353±1.857
	Champion	1.423	7.96	76.37	8.21
$CsPbBr_3$/ Ti_3C_2-MXene	Average	1.349±0.091	7.365±1.385	69.23±8.18	7.098±1.912
	Champion	1.440	8.75	77.41	9.01

Table S4 Comparison of the performance of inorganic and hybrid PSCs using additive in previous literature and in this work.

Material	**Additive**	**PCE**	**Life time (air)**	**Life time (heat)**	**Ref.**
$CsPbBr_3$	**Ti_3C_2-MXene**	**9.01%**	**1900 h**	**600 h @80°C**	**This work**
$FA_{0.8}Cs_{0.2}PbI_{2.64}Br_{0.36}$	PEO	14.9%	192 h	192 h @85°C	Adv. Mater. 2019, 31, 1804284.
$FA_{0.85}Cs_{0.15}PbI_3$	amorphous silica	18.8%	1000 h	/	Adv. Energy Mater. 2018, 8, 1800232
$MAPbI_3$	*g*-C_3N_4	19.41%	/	/	Adv. Funct. Mater. 2018, 28, 1705875
$MAPbI_3$	TBCP	19.4%	480 h	/	Chem, 2018, 4, 1.
$(FAPbI_3)_{0.7}(CsSnI_3)_{0.3}$	SnF_2 • 3FACl	15.8%	288 h	/	Adv. Energy Mater. 2018, 8, 1800997.
$CsPbI_3$	PEAI	15.07%	300 h	/	Nat. Commun. 2018, 9, 4544.
$CsPbI_3$	PVP	10.74%	500 h	500 h @60°C	Nat. Commun. 2018, 9, 1076.
$CsPbBr_3$	$CsPb_2Br_5$	6.81%	2400 h	/	ACS Appl. Mater. Interfaces, 2018, 10, 7145

$CsPbBr_3$	$CsPb_2Br_5$	8.34%	1000 h	/	J. Mater. Chem. A 2018,6,14255.
$CsPbBr_3$	/	9.72%	3120 h (RH 90%)	960 h @80°C	Angew. Chem. Int. Ed. 2018, 57, 3787.