Supporting Information

MXene as hole transport interfacial layer for efficient and air-stable quasi-2D perovskite solar cells

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Hysterysis:



Fig S1: Forward and backward scan of J-V measurement (scan rate 100 mV/s to determine

$$HI = \frac{J_{rev} \left(\frac{V_{oc}}{2}\right) - J_{for} \left(\frac{V_{oc}}{2}\right)}{J_{rev} \left(\frac{V_{oc}}{2}\right)}$$
 is equal to HI =

the hysteresis index. The hysteresis index defined as

 0.24 ± 0.05 (standard deviation from 6 measurements of different cells) and within experimental error is independent on the presence or absence of MXene layer.

Recombination dynamics:

To begin a quantitative analysis of the recombination dynamics, it is necessary to obtain values for the charge carrier density n. Usually, capacitance spectroscopy (impedance)

can be a good tool to determine the charge carrier's density in PSCs under illumination¹. The corrected capacitance C_{cor} was calculated using the following formula,

$$C_{cor} = -\frac{1}{\omega} \left[\frac{Z'' - \omega L}{(Z' - R_s)^2 + (Z'' - \omega L)^2} \right]$$
(1)

where ω is the angular frequency, Z' and Z'' are the real and imaginary components of the measured impedance, $L = 5 \cdot 10^{-6}$ H is the parasitic inductance of the connected wires and R_s is the series resistance of the solar cell. The chemical capacitance C_{chem} was determined at low frequencies (30 kHz) under light for different applied biases (Figure S2). At this particular frequency, a plateau is observed, which indicates that the traps in this region have an insignificant influence on the value of the measured capacitance.



Figure S2: The chemical capacitance C_{chem} as a function of the corrected voltages ($V_{cor} = V - JR_s$) for two different PSCs.

The chemical capacitance (C_{chem}) was used to calculate the charge carrier density (n) at different corrected voltages (V_{cor}) using the following equations^{2,3},

$$n(V) = n_{sat} + \frac{1}{qAL} \int_{V_{sat}}^{V} C_{chem} dV$$

$$n_{sat} = \frac{1}{qAL} C_{sat} (V_0 - V_{sat})$$

$$(3)$$

where A is the solar cell area, L is the thickness of the perovskite layer, V_{sat} is the reverse bias where the photocurrent saturates, and n_{sat} is the charge carrier density at the saturation voltage V_{sat} . The C_{sat} is the difference in capacitance of the solar cell under light and dark at V_{sat} . Similarly, V_0 is the forward bias at which the photocurrent becomes zero. Charge carrier density *n*, determined via capacitance spectroscopy is shown in Figure S3.

The experimentally measured recombination current density (J_{rec}) was determined using the following equation 4:

$$J_{rec} = J_{ph,s} - J_{ph,} \tag{4}$$

where $J_{ph} = J_{Light} - J_{dark}$ is the photocurrent density and $J_{ph,s}$ is the saturated photocurrent density at a reverse bias.



Figure S3: Charge carrier density (n) determined via capacitance spectroscopy for two different PSCs.

Also, J_{rec} can be represented as the sum of individual recombination currents:

$$J_{rec,sum} = J_{r,rad} + J_{r,b} + J_{r,s},$$
(5)

where $J_{r,rad}$ is the radiative recombination current density, $J_{r,b}$ is the recombination current density via bulk trap states, and $J_{r,s}$ is the recombination of current density via surface trap states. Each of these terms can be represented as follows:

$$J_{r,rad} = qAk_{rad}n^2 \tag{6}$$

$$J_{r,b} = qAk_{r,b}n = qLC_pN_{t,b}n \tag{7}$$

$$J_{r,s} = qC_p \frac{N_{t,s}n}{exp\left(\frac{q(V_{bi} - V)}{kT}\right)},$$
(8)

where k_{rad} is the radiative or bimolecular recombination coefficient, $k_{r,b}$ is the bulk-trapassisted recombination coefficient, $N_{t,b}$ is the density of bulk traps, $N_{t,s}$ is the density of surface traps, and C_p is the capture coefficient¹. The fitting quality for the two cases was quantified using the adjusted R^2 value, with the lowest value of 0.9911 and determined values of the recombination coefficient k_{rad} and traps density $N_{t,b}$, and $N_{t,s}$ are shown in the manuscript (Figure 4b in the main part).

Photolumenscence (PL)



Figure S4: Stationary emission spectra for with and without passivated PSCs. The excitation wavelength was 500 nm. The sample was excited from the HTL side and the emission was collected at an angle close to 90 degrees, therefore the interface of perovskite with HTL/MXene was mainly probed.

Thickness measurement using AFM and cross-section SEM:



Figure S5: (a) Thickness profile for 2D-PVSK and MXene on 2D-PVSK; (b) Cross-section SEM image for MXene on 2D-PVSK. Both analyses depict the thickness in the range of 45-50 nm of MXene when we passivated at 4000 rpm (for 10 s) using a spin coater.

EQE with integrated current density:



Figure S6. Original EQE spectra with integrated current density.

Dry air stability:

 Table TS1: Stability after 6 months

Device	Jsc	Voc	FF	РСЕ
2D-PVSK	14.4	1.03	0.45,	6.7%
2D-PVSK/ MXene	14.9	1.03	0.45	6.9%

Transient absorption:



Figure S7. Photoinduced degradation upon pulse excitation at 475 nm.





Figure S8. Exemplary pseudo-3D colour plots of transient absorption (TA) data under 475 nm excitation. Different colours indicate different positive and negative TA amplitudes according to the scale bars. TA of reference **2D-PVSK** sample was excited at pump fluence 25 μ J/cm² from (a) ETL and (b) HTL side. TA of **2D-PVSK/MXene** sample excited at pump fluence 25 μ J/cm² from (c) ETL and (d) HTL side. TA of MXene layer excited at pump fluence 600 μ J/cm² prepared by (e) drop-cast and (f) spin-coating. The observed signal contribution of pure FTO has been subtracted in (e) and (f).



Figure S9. Example of global 3-exponential fit quality of the data presented in Figure 7a – kinetic fits to the first two most significant singular values (SV). The time scale is shown in the logarithmic scale so the time zero is shifted to 1 ps.



Figure S10. Global fit of TA for only MXene sample.

References:

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