Supporting Information

Interfacial Engineering Enables High Efficiency with High Open-circuit Voltage above 1.23 V in 2D Perovskite Solar Cells

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Key words: 2D perovskite, high open-circuit voltage, interfacial engineering

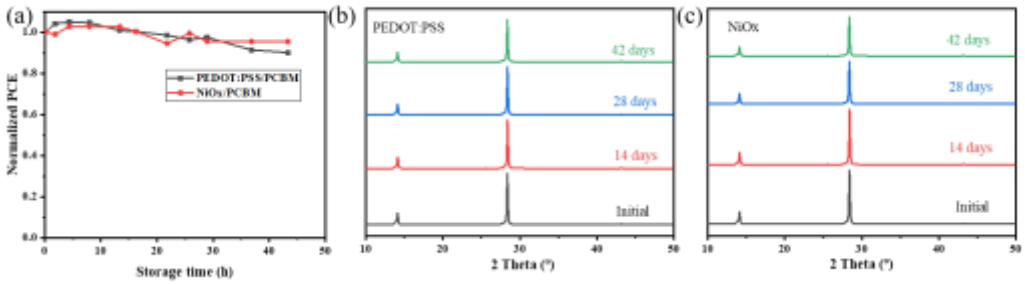

Fig. S1 a) The device storage stability in glovebox. XRD patterns for PEDOT:PSS/ perovskite film (b) and NiO_x/perovskite film (c) stored in ambient air with a humidity of 60%.

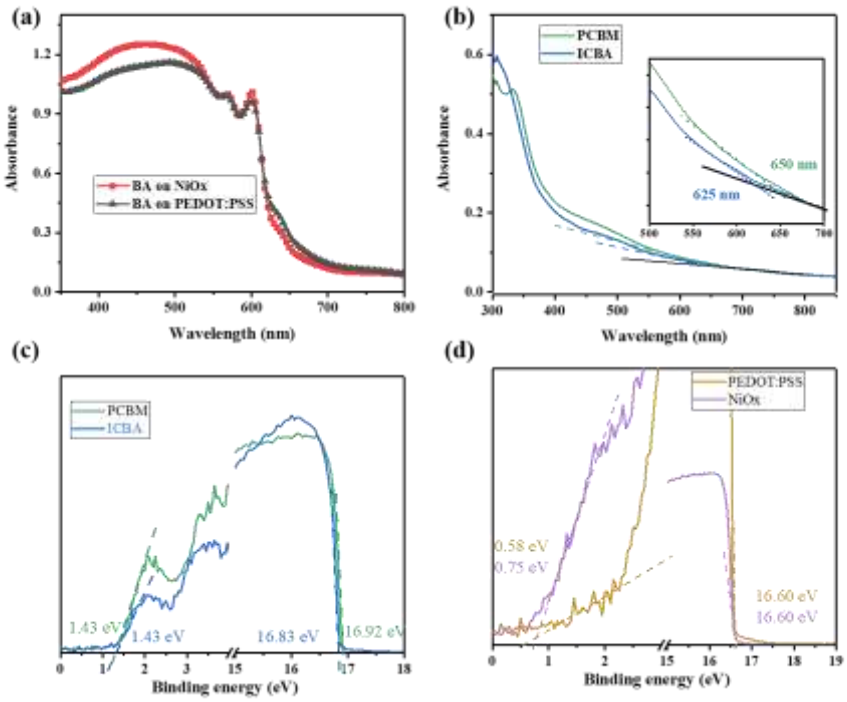

Fig. S2 The UV-Vis absorption spectra of $BA_2MA_2Pb_3I_{10}$ (a) and the bare ETLs (b), the inset inside (b) is the enlarged graph ranging from 500 nm to 700 nm. The UPS spectra for different ETLs (c) and HTLs (d).

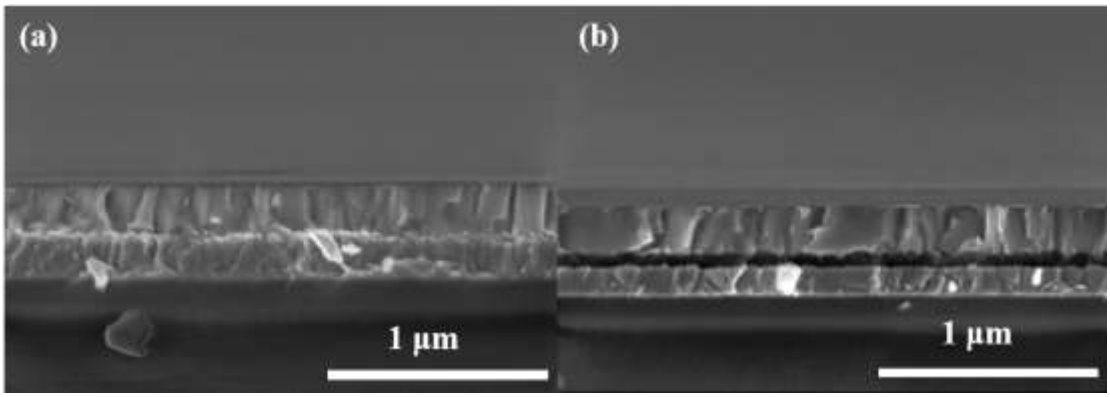

Fig. S3 The cross-sectional SEM imagies of $BA_2MA_2Pb_3I_{10}$ films deposited on NiO_x (a) and PEDOT:PSS (b).

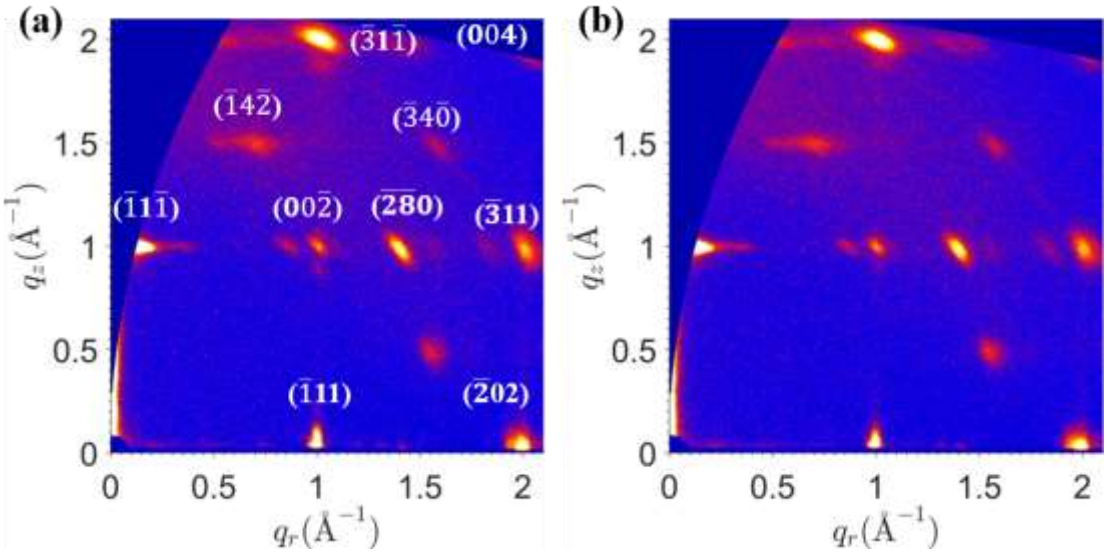

Fig. S4 GIWAXS patterns for BA2MA2Pb3I10 films deposited on PEDOT:PSS (a) and NiOx (b). The unit cell parameters are a=8.9275 Å, b=51.959 Å, c=8.8777 Å, respectively.

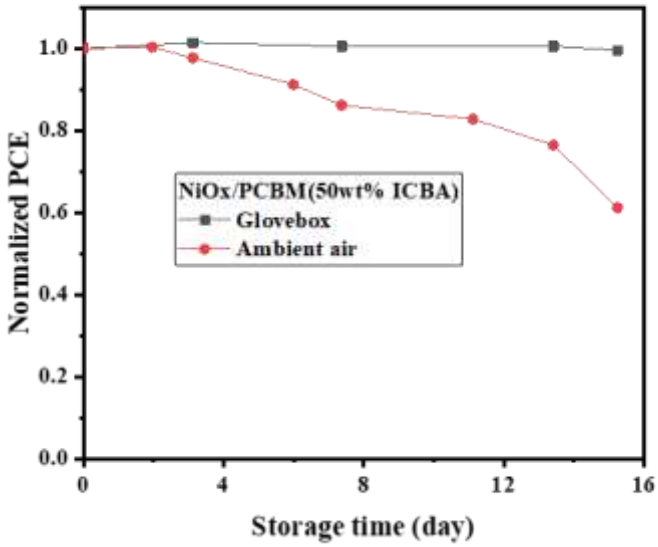

Fig. S5 The device storage stability in glovebox and ambient air (~40% humidity, 25-30 °C) for NiO_x/PCBA(50wt% ICBA).

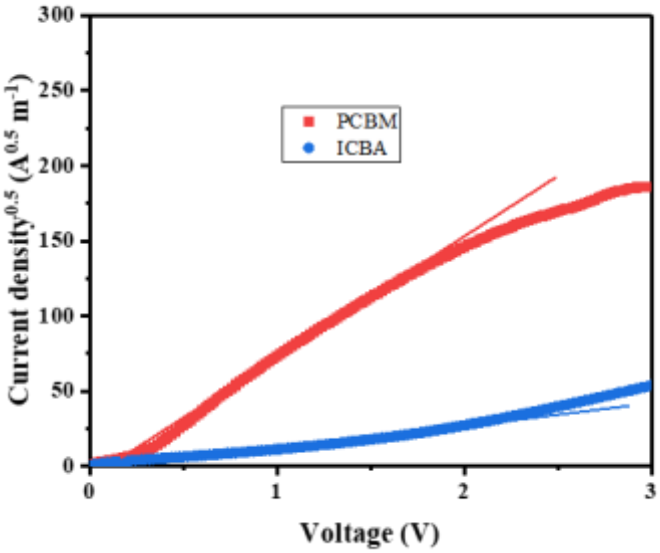

Fig. S6 *J-V* characterization of the electron-only devices with structures of ITO/ZnO/PCBM/BCP/Ag and ITO/ZnO/ICBA/BCP/Ag. The thickness of PCBM and ICBA layers are 50 nm.

Table S1 A summary of the high efficiency (>10%) 2D perovskite solar cells and their device structures.

Device structure	*Voc*	PCE	Reference
FTO/PEDOT:PSS/ $BA_2MA_3Pb_4I_{13}$ /PCBM/Al	1.01 V	12.52%	*Nature* **2016,** *536* (7616), 312-6.
ITO/PEDOT:PSS/ $PEA_2MA_4Pb_5I_{16}$ /PCBM/BCP/Ag	1.11 V	11.01%	*Adv. Energy Mater.* **2018**. 10.1002/aenm.201702498
ITO/PEDOT:PSS/$BA_2(MA_{0.2}FA_{0.8})_3Pb_4I_{13}$ /PCBM/BCP/Ag	1.00 V	12.82%	*J. Am. Chem. Soc.* **2018,** *140* (1), 459-465.
FTO/TiO_2/ $BA_2(MA_{0.95}Cs_{0.05})_3Pb_4I_{13}$ /Spiro-OMeTAD/Au	1.08 V	13.7%	*Energy Environ. Sci.* **2017,** *10* (10), 2095-2102.
ITO/PEDOT:PSS/ $BA_2MA_4Pb_5I_{16}$ /PCBM/Al	1.07 V	10.0%	*Adv. Energy Mater.* **2018,** *8* (1), 1700979-1700988.
ITO/PEDOT:PSS/ $PDA_2MA_2Pb_4I_{13}$ /C_{60}/BCP/Al	0.98 V	13.0%	*Adv. Mater.* **2018**, 10.1002/adma.201800710
FTO/TiO_2/ $BA_2MA_3Pb_4I_{13}$ /Spiro-OMeTAD/Au	1.08 V	12.17%	*Adv. Mater.* **2018**, 10.1002/adma.201707166
FTO/C_{60}/ $(iso\text{-}BA)_2MA_3Pb_4I_{13}$ /Spiro-OMeTAD/Au	1.20 V	10.63%	*Adv. Energy Mater.* ***2017****, 7, 1700162-170018*

Table S2 TRPL data of perovskite films with different mixed ETLs, fitting by the bi-exponential model.

HTL-ETL	Excited side	τ_1 [ns]	$Frac_1$	τ_2 [ns]	$Frac_2$	τ_{ave} [ns]
NiO_x-NONE	front	0.853	0.770	15.118	0.230	12.855
NiO_x- 0wt% ICBA	front	0.855	0.967	5.085	0.033	1.560
NiO_x-25wt% ICBA	front	0.836	0.953	4.636	0.047	1.648
NiO_x-50wt% ICBA	front	0.747	0.910	3.632	0.090	1.685
NiO_x-75wt% ICBA	front	0.834	0.946	5.146	0.054	1.959
NiO_x-100wt% ICBA	front	0.706	0.842	6.283	0.158	4.197