Electronic Supplementary Information

Enhancing Efficiency and Ambient Stability of Perovskite Solar Cells via Multifunctional Trap Passivation Molecule†

Maimur Hossain,[a] *Rahul Narasimhan Arunagirinathan,*[b] *Rabindranath Garai,*[a] *Ritesh Kant Gupta,*[b] *Parameswar Krishnan Iyer*[a,b*]

[a]Department of Chemistry, Indian Institute of Technology Guwahati, Guwahati-781039, Assam, India

[b]Centre for Nanotechnology, Indian Institute of Technology Guwahati, Guwahati-781039, Assam, India

Corresponding Author: E-mail: pki@iitg.ac.in; Fax: +91 361 258 2349

Materials

FTO glass substrates (13 Ω sq^{-1}), PbI_2 (99.8%), all anhydrous solvent e.g. DMF, DMSO, isopropanol (IPA) Toluene, Chlorobenzene, were purchased from Sigma-Aldrich. Methylammonium iodide (MAI) was obtained from Dyesol. Nickel nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$) and 5-fluoropyrimidine-2,4(1H,3H)-dione was obtained from TCI. All other chemicals were utilized as received.

NiO_x Film Preparation

NiO_x solution was prepared by dissolving Nickel nitrate hexahydrate and Ethylenediamine (in 1:1 molar ratio) in Ethylene Glycol (1 ml). Then the NiO_x layer was coated as hole transporting layer (HTL) on the cleaned FTO. The cleaning process of FTOs was started with detergent and followed by deionized (DI) water, acetone, and IPA for 15 min for each solvent, then dried and treated with UV-ozone for half an hour. NiO_x precursor solution was spin coated onto the FTO substrates at 3500 rpm for 45 sec. Afterwards, the substrates were annealed at 300 °C for 60 min under ambient condition.

Device Fabrication

The $MAPbI_3$ precursor solution was prepared by mixing 209 mg of MAI, 581 mg of PbI_2 in a solvent mixture of γ-Butyrolactone and DMSO (7:3, v/v) in a glovebox. The solution was heated for 5-6 hours and filtered with the 0.45 μm filter before spin coating. For the passivated device varied concentrations (1.5 mg/ml to 4.5 mg/ml) of FPD were added to the precursor solution. The filtered solution was spin coated on the NiO_x coated FTO in a two-step spin coating process i.e. 750 rpm for 20 sec and 4000 rpm for 60 sec. In the second step, 150 μl anhydrous Toluene was dripped dropwise after 20 sec as anti-solvent and after that the substrates were annealed at 80° C for 10 min on a hotplate. Then, for both passivated and pristine devices, 12 mg/ml $PC_{61}BM$ solution was coated at 1200 rpm as electron transporting layer (ETL) and again annealed at 80° C for 5 min on a hotplate. After that a thin layer of Rhodamine 101 inner salt was spin coated at 4000 rpm from a solution of 0.5 mg/ml in IPA. Lastly, silver was thermally deposited by using a shadow mask to obtain the devices with active area of 0.12 cm^2.[1]

Device Characterization

The XRD patterns of the perovskite films were studied using a Rigaku Micromax-007HF diffractometer equipped with Cu Kα1 irradiation (λ = 1.54184 Å). The perovskite films were analyzed by UV-vis absorption spectroscopy (Perkin Elmer Lamda-35) and FTIR spectroscopy (LabRam HR) in ATR mode. The film morphology of the samples was investigated by scanning electron microscopy (FESEM, JEOL JSM-7610F). The current density−voltage (*J*−*V*) characteristic curves were recorded using a Keithley 2400 source meter under inert atmosphere by illuminating the device with a solar simulator (AM 1.5G, 100 mW cm^{-2}, Oriel Sol 3A solar simulator, Newport). The incident external quantum efficiency (EQE) was obtained by using an Oriel IQE-200 instrument under ambient condition. Electrochemical measurements were analyzed using a CH Instruments 760D.

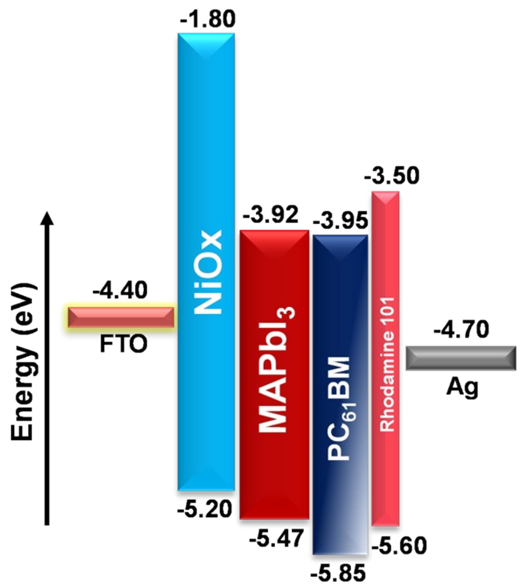

Fig. S1 Energy band diagram of constituent layers in PVSC.

Table S1. Device parameters for hysteresis study for pristine and FDP modified device.

Device	J_{SC}, mA/cm^2	V_{OC}, V	FF, %	PCE, %	HI, %
Pristine_FS	21.32	1.006	70.4	15.10	9.07
Pristine_RS	21.03	1.001	65.2	13.73	
FPD_FS	23.97	1.086	77.7	20.22	1.38
FPD_RS	24.01	1.083	76.7	19.94	

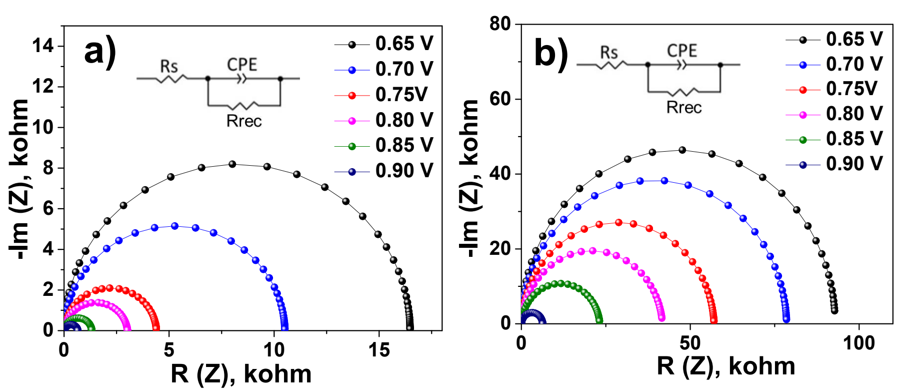

Fig. S2 Nyquist plots obtained from impedance spectroscopy conducted at varied bias (0.65V - 0.90 V) for a) pristine b) FPD passivated PVSCs.

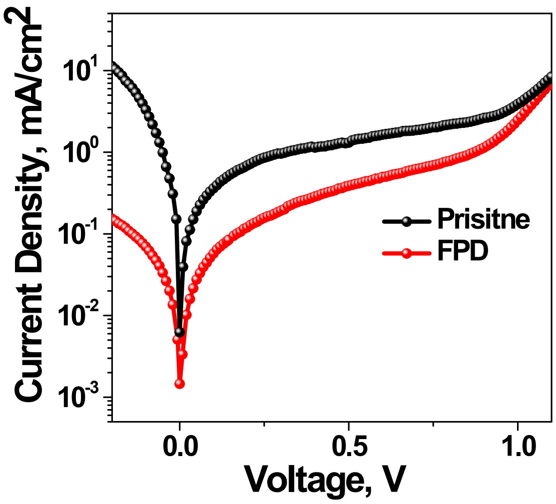

Fig. S3 Dark J–V characteristics of pristine and FPD modified PVSCs**.**

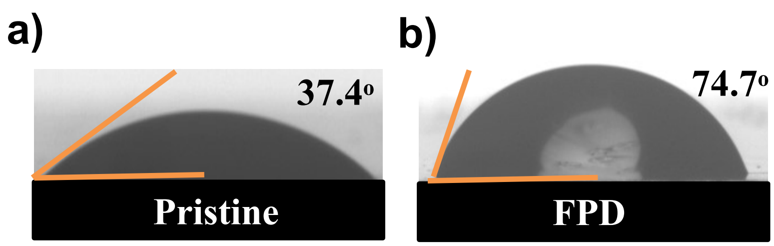

Fig. S4 Contact angle of a) pristine and b) FPD passivated films.

Table S2. Comparative list of similar research articles highlighting the PVSC performance and stability.

No.	**Article Source**	**Architecture**	**Passivation Additives**	**Pristine PCE, %**	**Champion Cell, %**	**Ambient Stability (PCE Retention)**
1	*J. Mater. Chem. A,* ***2021****, 9, 4138–4149*	FTO/SnO_2/ $FAPbI_3$/Spiro-OMeTAD/MoO_3/Au	Thiophene based additives	16.91	20.61	90% after, 60 days
2	*Adv. Energy Mater.* ***2019****, 9, 1900198*	FTO/NiOx/ $MAPbI_3$/$PC_{61}BM$/BCP/Ag	Fluorinated Perylene Diimide (F-PDI)	15.37	18.28	NA
3	*Adv. Sustain. Syst.* ***2020****, 2000078*	FTO/NiOx/ $MAPbI_3$/$PC_{61}BM$/Rhodamine 101/Ag	Chelidamic acid (CA)	13.60	19.06	80% after, 1000 h
4	*ACS Appl. Energy Mater.* ***2020****, 3, 2432−2439*	FTO/PEDOT:PSS/ $MAPbI_3$/$PC_{61}BM$/Rhodamine 101/Ag	Oxalic Acid	14.06	17.12	NA

5	*ACS Appl. Energy Mater.* ***2021**, 4, 1731−1742*	FTO/PEDOT:PSS/ $FA_{0.8}MA_{0.15}Cs_{0.05}Pb_{0.5}Sn_{0.5}I_3/C_{60}$/BCP/Ag	2-phenylethylazanium iodide	14.61	17.33	85% after, 33 h
6	*Adv. Funct. Mater.* ***2020**, 30, 2002861.*	$FTO/SnO_2/FA_{0.8}MA_{0.14}Cs_{0.05}PbI_{2.55}Br_{0.45}$/Spiro-OMeTAD /Au	indacenodithieno[3,2-b] thiophene (IDTT)	18.8	21.2	80% after, 2000 h
7	*ACS Sustain. Chem. Eng.* ***2020**, 8, 8848−8856*	ITO/PTAA/$MAPbI_3$/ PCBM/BPhen/Al	Perylene Diimide based small molecule	17.3	20.3	75% after, 50 h
8	*Sol. RRL* ***2020**, 4, 1900529*	ITO/PTAA/$MAPBI_3$/ PCBM/Al	Isatin-Cl	18.13	20.18	90% after, 350 h
9	*J. Phys. Chem. Lett.* ***2020**, 11, 6772−6778*	FTO/PTAA/$MAPBI_3$ / $PC_{61}BM$/Al	Polyvinylcarbazole	17.4	18.7	NA
10	*J. Energy Chem. 59,* ***2021** 755–762756*	FTO/SnO_2/ $MAPBI_3$/Spiro-OMeTAD /Au	indacenodithieno[3,2-b] thiophene (IDTT)	18.32	20.18	95% after, 1200 h
11	*ACS Appl. Mater. Interfaces* ***2021**, 13, 21194−21206*	FTO/TiO_2/ $MAPbI_3$ NPs/CsFAMA /Spiro-OMETAD	1-hexyl-3-methylimidazolium	17.33	19.44	80% after, 6000 h
12	*J. Mater. Chem. A,* ***2021**, 9, 5857–5865*	ITO/PTAA/$MAPbI_3$/ $PC_{61}BM$/BCP/Ag	Amino acids	17.51	20.49	94.9% after, 30 days
13	*Adv. Funct. Mater.* ***2021**, 31, 2010603*	FTO/TIO_2/ZrO_2/$MAPBI_3$/Carbon	N,1-Fluoro formamidinium Iodide	14.23	17.01	NA
14	*Adv. Funct. Mater.* ***2020**, 30, 2020778*	FTO/PEDOT:PSS/ $MAPBI_3$/PC61BM/Rhodamine 101/Ag	Phenylhydrazinium iodide	15.33	18.18	83% after, 20 days
15	***This article***	**FTO/NiOx/ $MAPBI_3$/$PC_{61}BM$/Rhodamine 101/Ag**	**5-fluoropyrimidine-2,4(1H,3H)-dione(FPD)**	**15.10**	**20.22**	**89% after, 1000 h**

References

S1. M. A. Afroz, N. Ghimire, K. M. Reza, B. Bahrami, R. S. Bobba, A. Gurung, A. H. Chowdhury, P. K. Iyer and Q. Qiao, *ACS Appl. Energy Mater.*, 2020, **3**, 2432-2439.