Supporting Information

Hexachlorotriphosphazene-assisted buried interface passivation for stable and efficient wide-bandgap perovskite solar cells

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Experimental

Materials

Tin-doped indium oxide (ITO) glass (15 Ω sq⁻¹) was purchased from South China Xiangcheng Technology Co., Ltd. PTAA, Cesium iodide (CsI), Formamidine iodide (FAI), Methylammonium Iodide (MAI), Methylamine chloride (MACl), Lead (II) iodide (PbI₂), Lead bromide (PbBr₂), Lead chloride (PbCl₂), fullerene (C60), bathocuproine (BCP), silver (Ag) were purchased from Xi'an Polymer Light Technology Corp. Dimethyl sulfoxide (DMSO), N, N-dimethylformamide (DMF), chlorobenzene (CB) and Ethyl acetate (EA) were purchased from Alfa Aesar. Hexachlorotriphosphazene (HCCP) was purchased from Sigma-aldrich. No further purification was carried out unless otherwise stated.

Device Fabrication

The pre-patterned ITO glass substrates were sequentially cleaned using acetone and ethanol. Before use, the as-cleaned substrates were further cleaned with UV ozone treatment. PTAA solution (2 mg/ml in chlorobenzene) was spin-coated on an ITO substrate at 4000 rpm for 30 s, followed by annealing at 100°C for 10 min. HCCP solution (0.5 mg/ml in methanol) was spin-coated on PTAA film at 4000 rpm for 30 s. Wide-bandgap perovskite solutions were prepared by dissolving FAI, CsI, PbI₂, PbBr₂, MACl, PbCl₂ molar ratios of which were adjusted to form stoichiometric 1.4M $[Cs_{0.22}FA_{0.78}Pb(I_{0.85}Br_{0.15})_3]_{0.97}(MAPbCl_3)_{0.03}$, in DMF and DMSO mixed solvent system (DMF: DMSO= 4:1 volume ratio). In the anti-solvent quenching method, the perovskite solution is spin-cast at 5000 rpm for 50 s. Ethyl acetate (300 uL) is dropped on the film at 25~30 s from the start of the spin. Post-annealing is done at 100 °C for 30mins. Subsequent layers (C60 (30 nm), bathocuproine (BCP) (7 nm), Ag (100 nm) electrode) following the perovskite absorber were deposited using a thermal evaporator.

Characterization and Measurement

UV-vis spectroscopy was performed on a UV-vis-NIR 3600 spectrometer (Shimadzu, Japan). ATR-FTIR spectra were obtained on Nicolet 8700 (Thermo Electron

Corporation). XPS measurements were performed on the THERMO VG ESCALAB 250. PL spectra (excitation at 488 nm, front-side excitation) were obtained using a fluorescence spectrophotometer (FLS980, Edinburgh Instruments). The XRD patterns were measured using an X-ray powder diffractometer (XRD-6000, SHIMADZU) with an angle range of $2\theta = 5^{\circ}$ to 60° . The contact angles were measured by a drop shape analyzer (DSA30S, Krüss). AFM spectra were obtained on a Bruker DMFASTSCAN2-SYS. The surface morphology of the samples was conducted by Scanning electron microscopy (SEM) of Hitachi HITACHI S-470. The 1H-NMR analyses were performed on a Bruker AVANCE spectrometer (1H frequency = 400 MHz). Current density-voltage (*J-V*) characteristics were performed inside a nitrogen-filled glove-box under 100 mW cm⁻² simulated AM 1.5 G irradiation with a solar simulator (SS-F5-3A, EnliTech), calibrated by standard silicon cell (SRC-2020, EnliTech). The EQE with the measurement scope of 300-900 nm was characterized by the QE-R systems (EnliTech) under an ambient atmosphere at room temperature.



Figure S1. ³¹P Nuclear magnetic resonance (NMR) of HCCP solution without (left) and with (right) PbI_2 .



Figure S2. Fourier transform infrared (FTIR) spectra of HCCP.



Figure S3. Fourier transform infrared (FTIR) spectra of perovskite film without and with HCCP.



Figure S4. X-ray photoelectron spectroscopy (XPS) spectra of perovskite film.



Figure S5. AFM images of perovskite film (a) without and (b) with HCCP.



Figure S6. (1 1 0) crystal phase characteristic diffraction peak.



Figure S7. J_{sc} and the relevant fitting curves under different illuminated light intensities for the devices with and without HCCP.



Figure S8. J-V curve based on different HCCP concentrations

Sample	w/o HCCP	with HCCP
A ₁ (%)	248.62	147.31
τ_1 (ns)	4.83	6.85
A ₂ (%)	40.82	19.34
τ_2 (ns)	57.67	226.23
$\tau_{ave} \left(ns \right)$	39.70	185.50

Table S1. Fitted results of TRPL curves of the perovskite films with or without modifiers deposited on glass.

Table S2. Detail photovoltaic parameters of best performing WBG PerSCs based on without and with HCCP

	$J_{\rm sc}$ (mA/cm ²)	$V_{ m oc}$ (V)	FF (%)	PCE (%)
w/o HCCP	20.62	1.15	78.35	18.41
with HCCP	21.31	1.21	83.30	21.47

Bandgap (eV)	$J_{\rm sc}~({\rm mA/cm^2})$	$V_{\rm oc}$ (V)	FF (%)	PCE (%)	Ref.
1.65	21.2	1.23	83.80	21.90	1
1.66	20.10	1.19	82.00	20.10	2
1.66	20.80	1.23	82.30	21.05	3
1.67	22.40	1.21	82.30	22.30	4
1.67	21.20	1.22	80.00	20.70	5
1.67	21.10	1.20	80.80	20.42	6
1.67	21.10	1.20	80.89	20.39	7
1.67	21.30	1.21	83.30	21.47	This work
1.68	20.90	1.19	81.80	20.31	8
1.68	20.70	1.22	82.00	20.80	9
1.68	21.65	1.20	81.50	21.10	10

Table S3. Summarized photovoltaic parameters of advanced WBG PerSC reported.

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