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Electronic Supplementary Information

DMAI-drived all-inorganic antimony-based perovskite-inspired solar cells with record open-circuit voltage

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Experimental Procedures

Device Fabrication:Pre-etched fluorine doped tin oxide (FTO) substrates were firstly washed with soap water, deionized water, ethanol and dried by high-purity nitrogen gas. Then, 100nm Nb_2O_5 compact layer films were sputtered by a high vacuum sputter system (Beijing Technol Science, JCP-450) and annealed in an oven at 500°C for 30 min. The detail of sputter process can be found in related references. To form the RbCl/Nb_2O_5, 1mg/mL RbCl (Sigma-Aldrich, 99.95%) aqueous solution was spin-coated on Nb_2O_5 surface and then annealed at 500°C for 30 min.The precursor solution was prepared by dissolving 0.75 M CsI (Sigma-Aldrich, 99.9%), 0.25 M SbI_3 (Sigma-Aldrich, 98%) and 0.3125 M $SbCl_3$ (Sigma-Aldrich, 99.95%) in DMF (Sigma-Aldrich, 99.8%) solution. The DMAI incorporated $Cs_3Sb_2Cl_xI_{9-x}$ films were fabricated by adding the certain amount DMAI (Sigma-Aldrich) in precursor solution.Then, the precursor solution was spin-coated onto Nb_2O_5 coated substrates at 3000 rpm for 30 s and post-annealed under SbI_3 vapor environment at 250°C for 10 min. For the hole transport layer (HTL) layer, 6 mg/mL P3HT (Sigma-Aldrich) solution were spin-coated on the top of $Cs_3Sb_2Cl_xI_{9-x}$ film at 3000 rpm and annealed for 5 min at 100°C. After the films cooled to room temperature, commercial carbon paste was screen-printed on the top of HTL layer and dried on a hot plate at 120°C for 15 min.

Characterization: Grazing Incidence X-ray diffraction (GIXRD) measurements were carried out by a X-ray Powder diffractometer (Bruker D8 Advance). X-ray photoelectron spectroscopy (XPS) measurements were conducted by a Thermo SCIENTIFIC ESCALAB 250X with a He-discharge UV source (21.2 eV) for ultraviolet photoelectron spectroscopy (UPS) measurements. Time-of-flight secondary-ion massspectrometry (ToF-SIMS) depth profiles were measured with a Bruker ultraflextreme MALDI-TOF. Optical absorbance spectra of the films were recorded by a Cary5000 UV-VIS-NIR spectrophotometer. Scanning Electron Microscopy (SEM) measurements were characterized by a HITACHI S-4800 SEM. The photoluminescence (PL) and transient-photoluminescence (TRPL) spectra were measured by a HORIBA spectrophotometer with a excitation source of 532 nm. The current density–voltage (J-V) characteristics of PISCs were conducted by a Keithley 2400 digital source meter with a solar simulator (Newport Oriel Sol3A) calibrated to AM 1.5, 100 mW/cm^2 using a standard silicon photodiode (Newport Oriel 91150V) with an masked active area of 0.09 cm^2. The incident photo-to-current conversion efficiency (IPCE) was performed by QTEST 1000AD Station using a calibrated reference Si-cell. The electrochemical impedance spectroscopy (EIS) was recorded by a Bio-Logic VMP3 electrochemical workstation under dark.

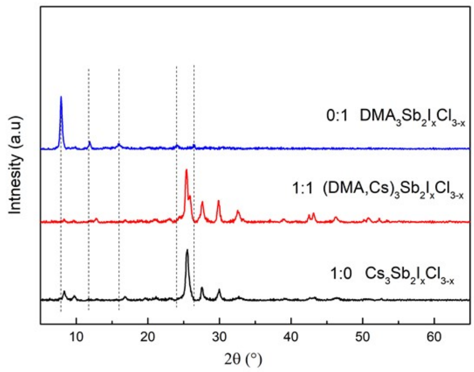

Figure S1.XRD patterns of $A_3Sb_2Cl_xI_{9-x}$ films (A=DMA/Cs).

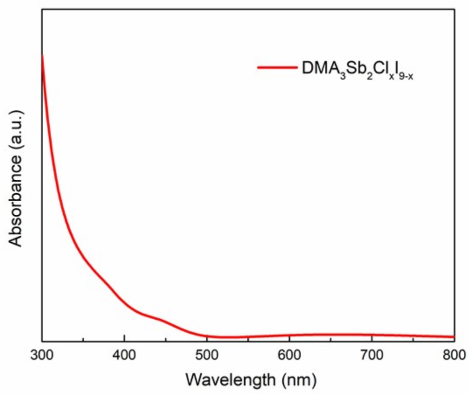

Figure S2.Optical absorbance spectra of of $DMA_3Sb_2Cl_xI_{9-x}$ film.

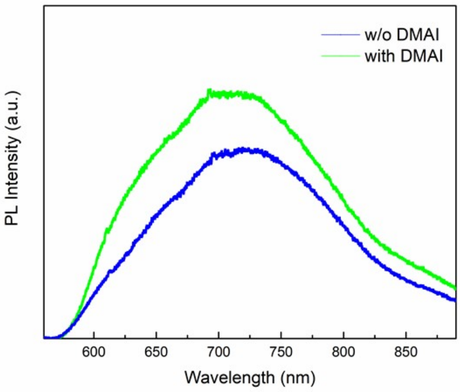

Figure S3.PL spectra for $Cs_3Sb_2Cl_xI_{9-x}$ films with/without DMAI additive.

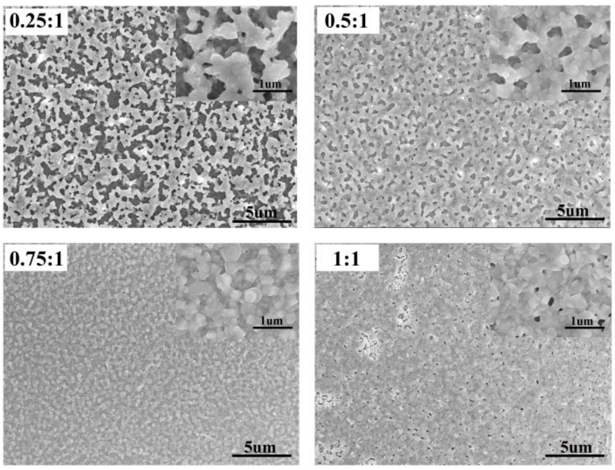

Figure S4.SEM picture of of $Cs_3Sb_2Cl_xI_{9-x}$ films prepared with different CsI/DMAI ratio.

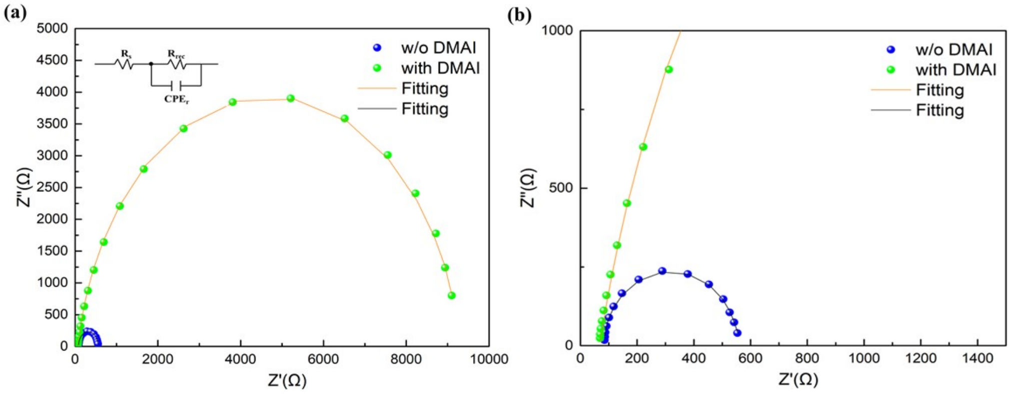

Figure S5. (a) Enlarged and (b) overall Nyquist plots of $Cs_3Sb_2Cl_xI_{9-x}$ PISCs with/without DMAI additive.

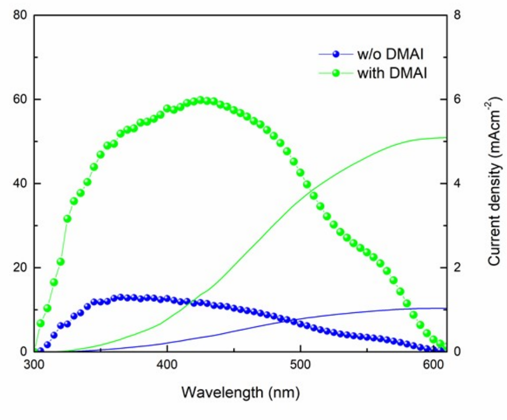

Figure S6. IPCE curves for $Cs_3Sb_2Cl_xI_{9-x}$ PISCs with/without DMAI additive.

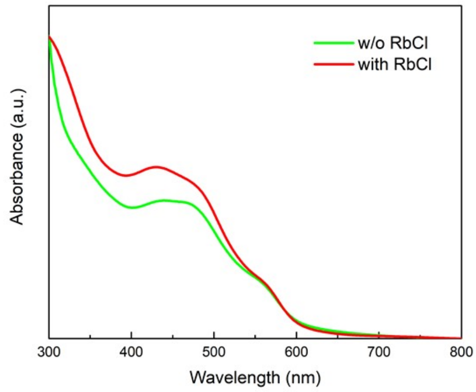

Figure S7. Optical absorbance for $Cs_3Sb_2Cl_xI_{9-x}$ film on Nb_2O_5 and $RbCl/Nb_2O_5$ substrates.

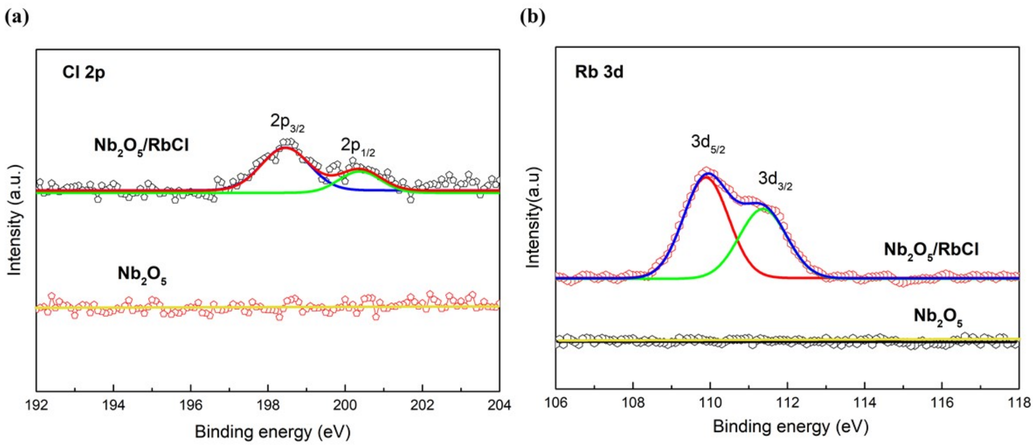

Figure S8. (a) XPS spectra for Cl 2p core level.(b) XPS spectra for Rb 3d core level.

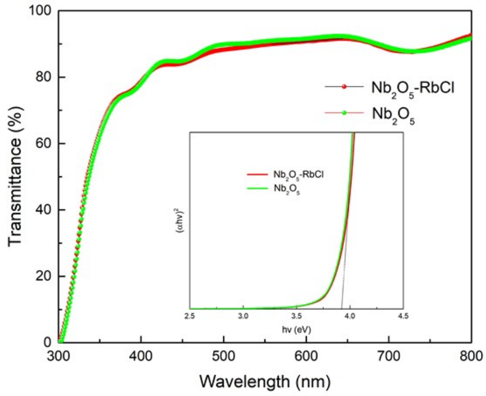

Figure S9. Optical transmittance for $RbCl/Nb_2O_5$ and Nb_2O_5 film.

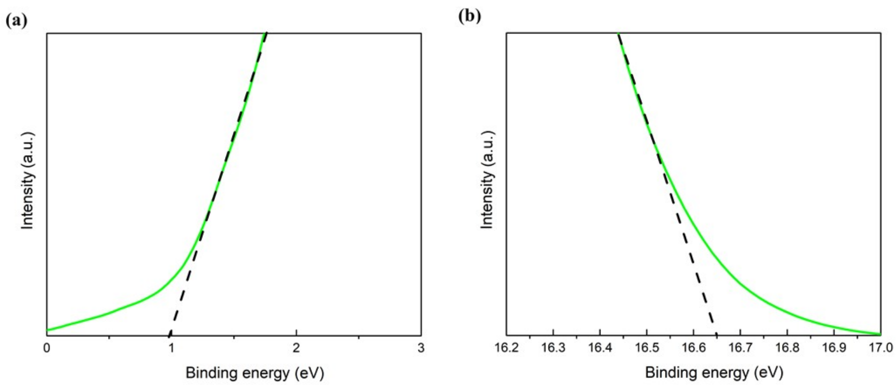

Figure S10. Fermi edges and secondary electron cutoff edges of D-$Cs_3Sb_2Cl_xI_{9-x}$ film with RbCl interface modification.

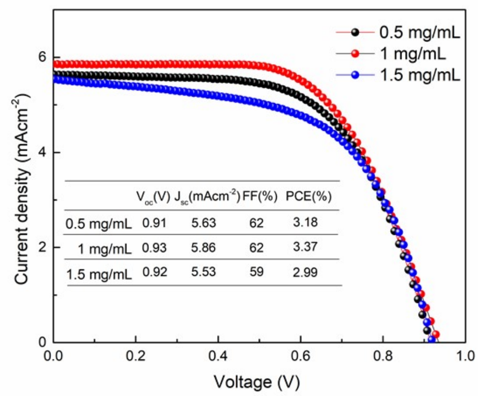

Figure S11. J-V curves for PISCs with different RbCl interface modification concentration.

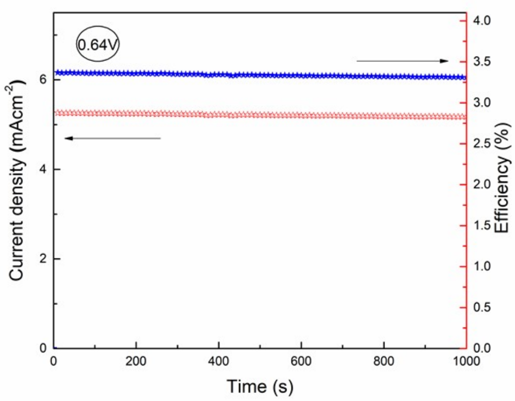

Figure S12. Stable output curve of J_{sc} and PCE for PISC with RbCl interface modification at maximum power point.

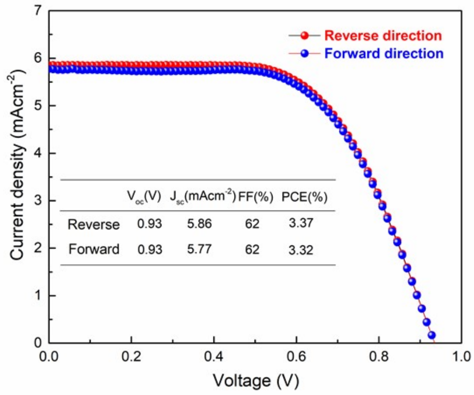

Figure S13. J-V curves for PISC with RbCl interface modification under different scanning directions.

Table S1. TRPL parameters for $Cs_3Sb_2Cl_xI_{9-x}$ films with/without DMAI additive.

Sample	A_1	τ_1 (ns)	A_2	τ_2 (ns)	τ_{avg} (ns)
w/o DMAI	0.14	1.57	0.86	13.54	3.24
with DMAI	0.68	2.03	0.32	27.15	10.06

Table S2. EIS parameters of $Cs_3Sb_2Cl_xI_{9-x}$ PISCs with/without DMAI additive.

Sample	R_s (ohm)	R_{rec} (ohm)
w/o DMAI	83	477
with DMAI	69	9246

Table S3. Temperature-related J-V performance parameters of PISCs with DMAI additive.

Sample	V_{oc} (V)	J_{sc} (mAcm^{-2})	FF (%)	PCE (%)
100°C	0.79 (0.76 $\pm$ 0.02)[a]	0.85 (0.82 $\pm$ 0.21)	56 (53 $\pm$ 1.04)	0.38 (0.34 $\pm$ 0.11)
150°C	0.81 (0.8 $\pm$ 0.01)	2.62 (2.27 $\pm$ 0.32)	54 (52 $\pm$ 1.41)	1.15 (0.96 $\pm$ 0.21)
200°C	0.84 (0.82 $\pm$ 0.03)	4.56 (4.26 $\pm$ 0.19)	56 (55 $\pm$ 1.55)	2.15 (1.93 $\pm$ 0.15)
250°C	0.89 (0.87 $\pm$ 0.01)	5.21 (4.95 $\pm$ 0.23)	59 (58 $\pm$ 1.52)	2.74 (2.46 $\pm$ 0.12)

[a] Statistic performance from 16 individual devices

Table S4. J-V performance parameters of PISCs with/without RbCl interface modification.

Sample	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
w/o RbCl	0.89 (0.87 $\pm$ 0.01)[a]	5.21 (4.95 $\pm$ 0.23)	59 (58 $\pm$ 1.52)	2.74 (2.46 $\pm$ 0.12)
with RbCl	0.93 (0.91 $\pm$ 0.01)	5.86 (5.75 $\pm$ 0.18)	62 (60 $\pm$ 1.14)	3.37 (3.15 $\pm$ 0.12)

[a] Statistic performance from 16 individual devices