

Increasing the wettability and reducing excess PbI_2 using diamine hydrobromides with different lengths at the buried interface of the 3D perovskite film

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Materials

Materials used in experiments as received without further purification, include PbI_2 , PbBr_2 , CsBr (99.99%, Xi'an Polymer Light Technology), FABr , MABr (99.5%, Xi'an Polymer Light Technology), PTAA ($M_w \sim 2.5$, Xi'an Polymer Light Technology), Chlorobenzene (CB, 99.9%, Innochem), 1,2-Dichlorobenzene (ODCB, 99%, Innochem), DMSO (Dimethyl sulfoxide, 99.9%, Innochem), DMF (N, N-Dimethylformamide, 99.9%, Innochem), PC_{61}BM , BCP (99%, Xi'an Polymer Light Technology), EDADBr, BDADBr, HDADBr, ODADBr (99.5%, Xi'an Polymer Light Technology).

Preparation of precursor solutions

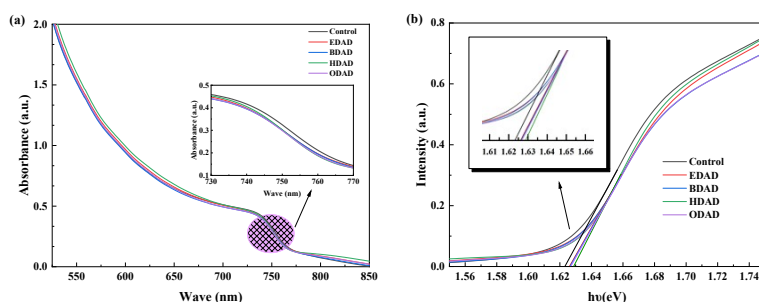
Perovskite precursor solution of $\text{Cs}_{0.05}(\text{FA}_{0.85}\text{MA}_{0.15})_{0.95}\text{Pb}(\text{I}_{0.85}\text{Br}_{0.15})_3$ was prepared by dissolving CsI (0.05 mmol), FAI (0.8075 mmol), MABr (0.1425 mmol), PbI_2 (0.88 mmol), and PbBr_2 (0.1425 mmol) in N-dimethylformamide (DMF) and dimethyl sulfoxide (DMSO) mixed solvent (volume ratio of 4:1) with molar concentration of 1.5 M.

Fabrication of p-i-n PSCs

ITO-coated glass (resistance $\leq 10 \Omega \text{ cm}^2$) was continuously cleaned in detergent, deionized water, ethanol, and deionized water ultrasonic bath for 30 minutes, and then ITO glass was treated with plasma for 90 s. HTL is prepared by dissolving PTAA (14 mg/ml) in CB and spinning onto a clean ITO for 30 s at 4000 rpm in a nitrogen-filled glove box, then annealing at 100°C for 10 minutes. For the bottom modification of the 2D interface modification layer, EDADBr, BDADBr, HDADBr, and ODADBr solutions of different concentrations (10, 15, and 20 mg mL^{-1}) were coated on PTAA substrate for 30 s at a rotational speed of 6000 rpm. It was then annealed at 100°C for 10 minutes. The perovskite precursor solution was deposited with the perovskite active layer for 100s at a rate of 7000 rpm by a spin coating method. After 20 s of spin coating, 150 μL of chlorobenzene was dropped on the active material as an antisolvent and annealed at 100°C for 30 min at the end of spin coating. After deposition of the perovskite active layer, PC_{61}BM (30 mg mL^{-1} in chlorobenzene) was spin-coated at 1200 rpm for 45 s, and then quickly transferred to a vacuum evaporation chamber to steam BCP samples with a thickness of 9 nm under vacuum condition of 4×10^{-4} Pa. It is then used to further deposit Ag (110 nm) in a vacuum chamber to complete the device fabrication process. The device's effective region is defined as using a metal mask of 0.04 cm^2 .

Characterization

The J - V characteristic curves of perovskite Solar cells were measured under the irradiation of AM 1.5G (Zolix, SS150 Solar Simulator) sunlight simulator. The degradation measurement uses an aging test system (Lancheng Technology, MPP Tracking-4B) including 4-channel synchronous scanning for testing. Space charge-limited current, voltage-capacitance, and dark current tests are performed by an electrochemical workstation (ZAHNER ENNIUM pro) under no light conditions. X-ray diffraction (XRD) patterns were tested using Bruker D8 advance (230 V, 50 Hz, 6.5 kVA). The scanning electron microscope (SEM) images were tested using Hitachi S4800. Ultraviolet-visible absorption was tested by PerkinElmer Lambda 35. XPS spectra were obtained by Thermo Fischer ESCALAB 250Xi; here, Al $K\alpha$ ray ($\nu=1253.6$ eV) was used as the excitation source, and signal accumulation was performed for 5-10 cycles. The UPS spectra were obtained by the same instrument, while the excitation source is He I ultraviolet light and the excitation source energy is 21.22 eV. Fourier transform infrared spectroscopy (FTIR) was tested with Nicolet 6700. Steady-state photoluminescence (PL) and transient photoluminescence (TRPL) were tested using FLS1000 (Edinburgh Instruments, UK).



S1. (a) UV-vis absorption spectra of films modified with different diamine hydrobromides and (b) the corresponding Tauc plots.

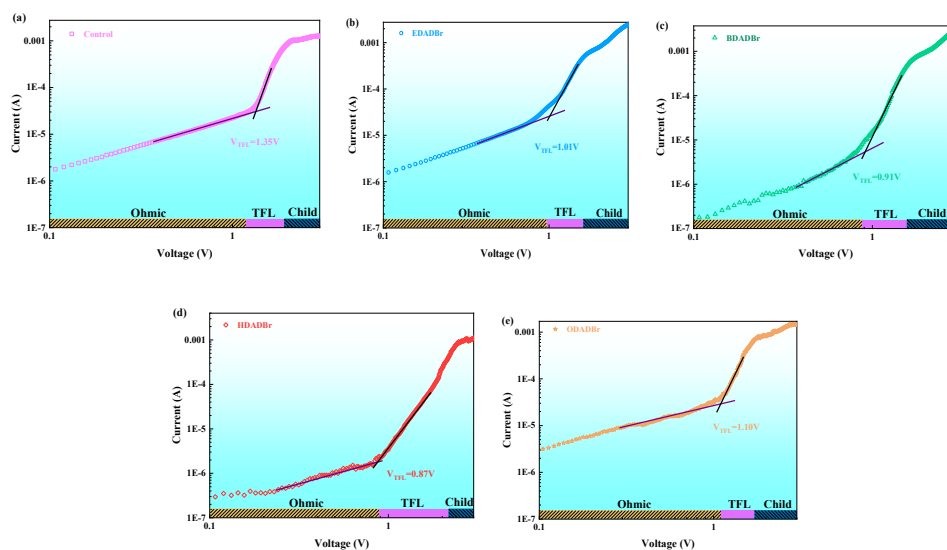


Figure S2. I - V curves of hole-only devices based on control and modified perovskite thin films with different diamine hydrobromides: (a) control, (b) EDADBr, (c) BDADBr, (d) HDADBr, and (e) ODADBr.

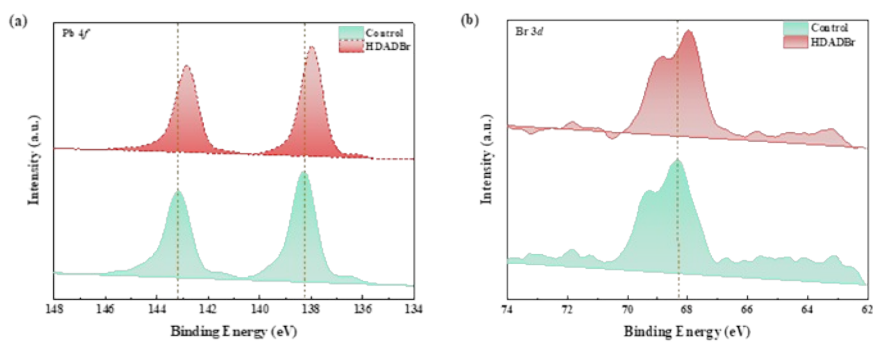


Figure S3. (a) Pb 4f and (b) Br 3d XPS spectra of control and modified perovskite thin films with HDADBr

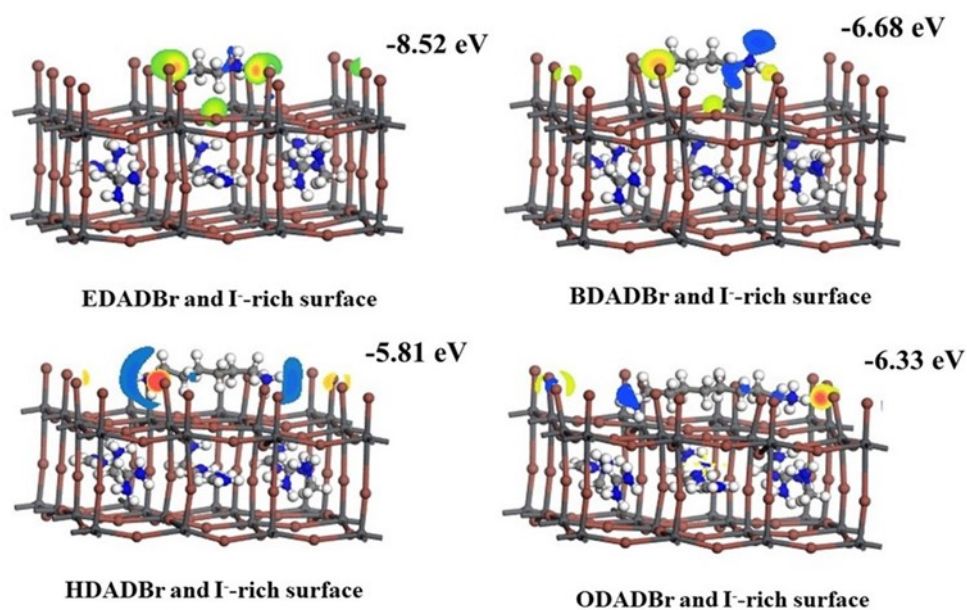


Figure S4. Interaction between different diamine hydrobromides and the perovskite surface from DFT calculations.

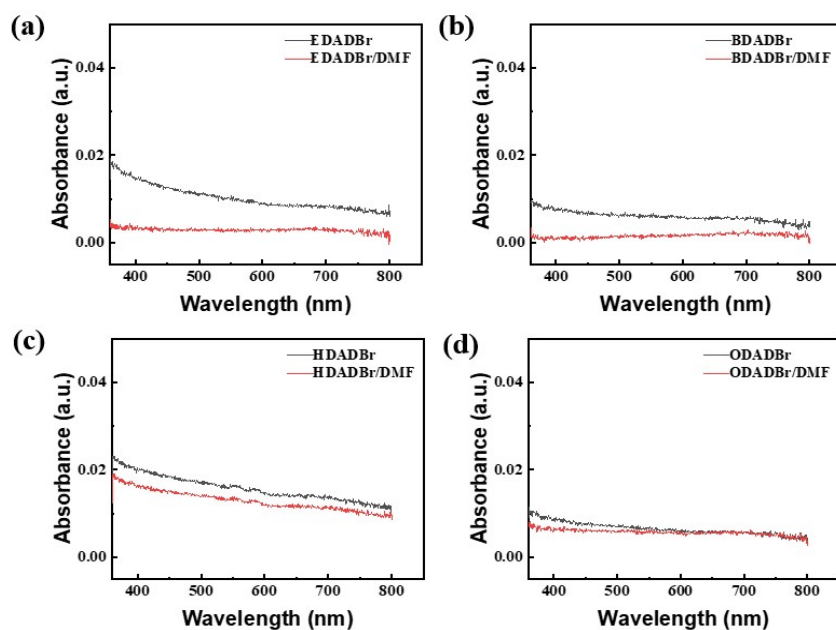


Fig. S5. UV-vis absorption spectra of films before and after spin-coating DMF: (a) EDADBr, (b) BDADBr, (c) HDADBr, and (d) ODADBr

Table S1. Device performance parameters of the PSCs with different concentrations of EDADBr.

	V_{oc} (V)	J_{sc} (mA/cm ²)	PCE (%)	FF (%)
Control	1.15	21.06	18.41	76.03
10 mg/ml EDADBr	1.15	21.17	18.67	76.71
15 mg/ml EDADBr	1.15	21.23	19.11	78.36
20 mg/ml EDADBr	1.15	20.73	18.74	78.66

Table S2. Device performance parameters of the PSCs with different concentrations of BDADBr.

	V_{oc} (V)	J_{sc} (mA/cm ²)	PCE (%)	FF (%)
Control	1.15	21.06	18.41	76.03
10 mg/ml BDADBr	1.15	21.05	19.45	80.34
15 mg/ml BDADBr	1.15	21.70	19.88	79.71
20 mg/ml BDADBr	1.16	21.23	19.71	80.02

Table S3. Device performance parameters of the PSCs with different concentrations of HDADBr.

	V_{oc} (V)	J_{sc} (mA/cm ²)	PCE (%)	FF (%)
Control	1.15	21.06	18.41	76.03
10 mg/ml HDADBr	1.16	21.32	19.67	79.57
15 mg/ml HDADBr	1.15	21.82	20.22	80.89
20 mg/ml HDADBr	1.14	22.01	19.98	79.61

Table S4. Device performance parameters of the PSCs with different concentrations of ODADBr.

	V_{oc} (V)	J_{sc} (mA/cm ²)	PCE (%)	FF (%)
Control	1.15	21.06	18.41	76.03
10 mg/ml	1.15	20.91	18.42	76.66

ODADBr				
15 mg/ml	1.15	21.04	18.93	78.31
ODADBr				
20 mg/ml	1.15	21.36	18.40	74.96
ODADBr				

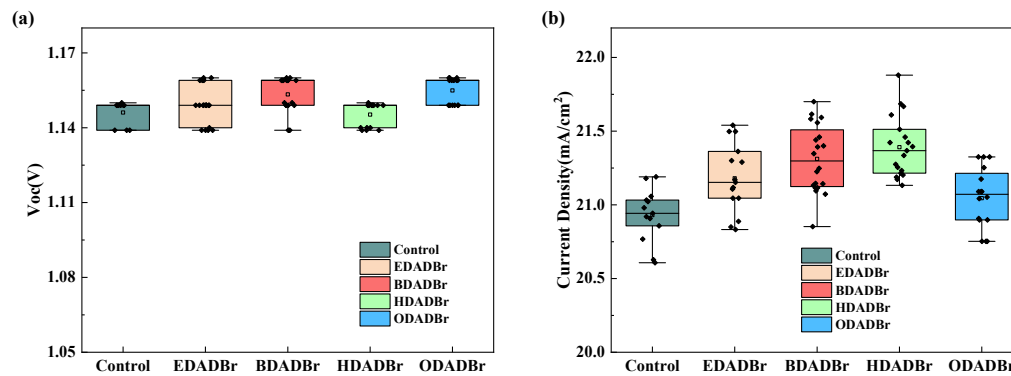


Figure S6. (a) V_{OC} and (b) J_{SC} distribution of the devices modified with different diamine hydrobromides.