

Supporting Information

Enhancing Open-Circuit Voltage in FAPbI₃ Perovskite Solar Cells via Self-Formation of Coherent Buried Interface FAPbI_xCl_{3-x}

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Experimental Section

Materials

The following materials were used in the experiments: FAI, spiro-OMeTAD were purchased from Advanced Election Technology. PbI_2 was purchased from Beijing Hawk Science & Technology Co. Ltd. PbCl_2 was purchased from Xi'an Polymer Light Technology Crop. SnO_2 colloidal solution (15%) DMF and DMSO were purchased from Alfa Aesar. Anhydrous ethanol was purchased from Yong Da Chemical. All reagents were used without further purification. Fluorinated tin oxide (FTO) conductive glass substrates (sheet resistance 14 Ω /square) were used for the PSCs.

FAPbI₃ film Fabrication

FTO glass substrates were initially cleaned using dish washing liquid, followed by treatment with deionized water and anhydrous ethanol through ultrasonic treatment for 20 minutes each in a drying box. For the experimental group, 479.44 mg of PbI_2 and 72.02 mg of PbCl_2 were dissolved in a DMSO and DMF solution. In contrast, the control group consisted solely of 599.3 mg of PbI_2 . SnO_2 is spin-coated in an air environment as an electron transport layer, and subsequently exposed to ultraviolet light for 30 minutes using an ozone meter. The electron transport layer's glass substrate was spin-coated within a glove box, where the precursor solution was applied. The PbI_2 solution was spin-coated at 3000 rpm for 30s, and the resulting spun-coated glass substrate was placed in a Petri dish containing FAI, allowing it to react in a vacuum drying oven at 170 °C to produce perovskite films.

Solar Cell Fabrications

The FTO glass substrates were cleaned with detergent, distilled water, ethanol and sonicated for 30 min. Then the SnO_2 precursor solution was spin-coated on FTO substrate at 5000 rpm for 30 s and subsequently annealed on a hot plate in ambient air at 150 °C for 30 min. The obtained SnO_2 /FTO substrates were immediately transferred into the N_2 -filled glove-box after annealing and cooling. Perovskite films are then prepared in a vacuum drying oven. Then, spiro-OMeTAD solution (25 μL), which consisted 73 mg of spiro-OMeTAD, 28 μL of 4-tert-butyl pyridine and 17.5 μL of lithium bis (trifluoromethanesulfonyl) imide (Li-TFSI) solution (520 mg of Li-

TSFI in 1 mL of acetonitrile) in 1mL of chlorobenzene, was spin-coated on the perovskite film at 3000 rpm for 20 s. Finally, Au electrode with a thickness of 60 nm was deposited by using thermal evaporation under vacuum at a constant evaporation rate of 0.6 nm/s.

Characterization

The J - V characteristic was measured with a Keithley 2400 source-meter together with a sunlight simulator (XES-300T1, SAN-EI Electric, AM 1.5G 100 mW cm⁻²), which was calibrated using a standard silicon reference cell. SEM images were taken with a SU8010 SEM (Hitachi). XRD was measured with a Bruker X-ray diffractometer with a Cu- $K\alpha$ radiation source. The UV-vis absorption spectra were measured with a UV-2450 spectrophotometer from 300 nm to 900 nm. IPCE was measured in air using a QE-R measurement system (Enli Technology). The XPS was measured with ESCALAB 250Xi, Thermo Fisher Scientific. Steady-state fluorescence and TRPL decay spectra were measured with a steady/transient state spectrophotometer (Nano LOG-TCSPC) with an excitation wavelength of 450 nm, and the films were deposited on clean glass directly. Electron-only devices (FTO/SnO₂/perovskite/Au) were fabricated to calculate the electron mobility of the devices. The dark J - V characteristics of the electron only devices were measured by a Keithley 2400 source. The mobility was extracted by fitting the J - V curves by the Mott-Gurney equation. The trap state density was determined by the trap-filled limit voltage using equation.

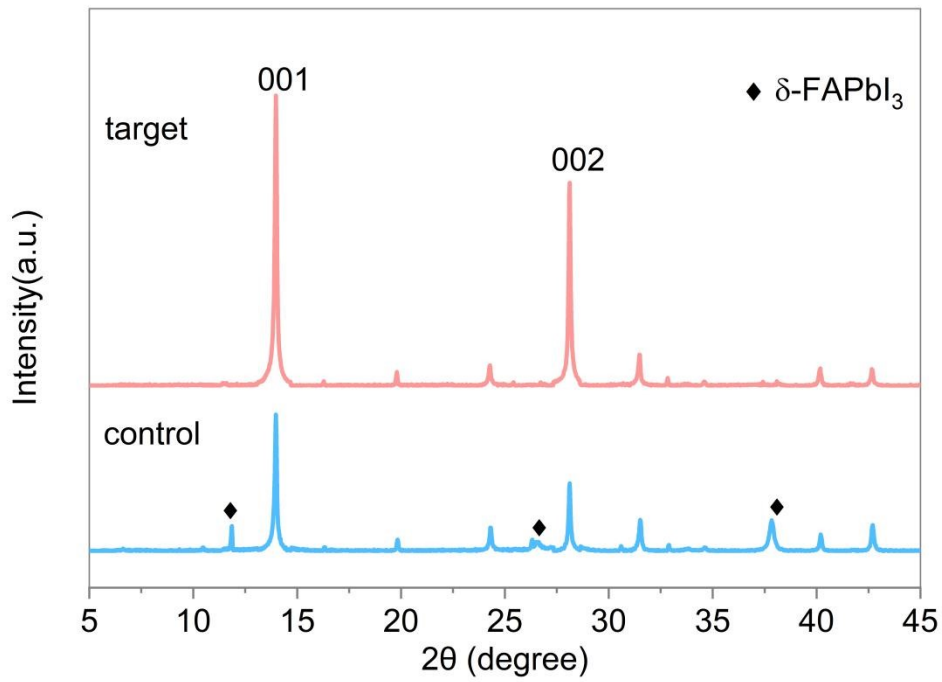


Fig. S1 The XRD patterns of the control and target perovskite films.

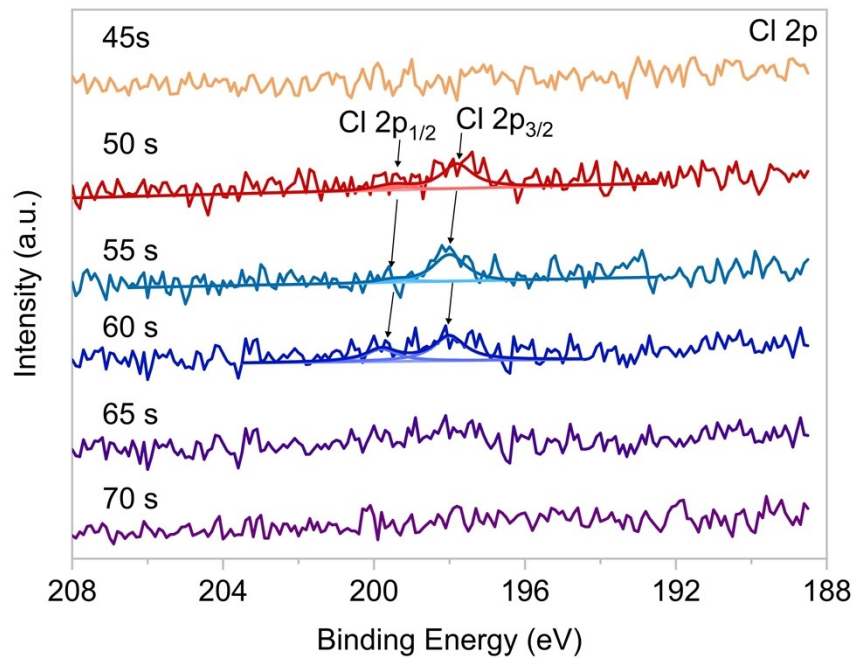


Fig. S2 XPS spectra of the Cl distribution in the bulk of the target film at different etching times.

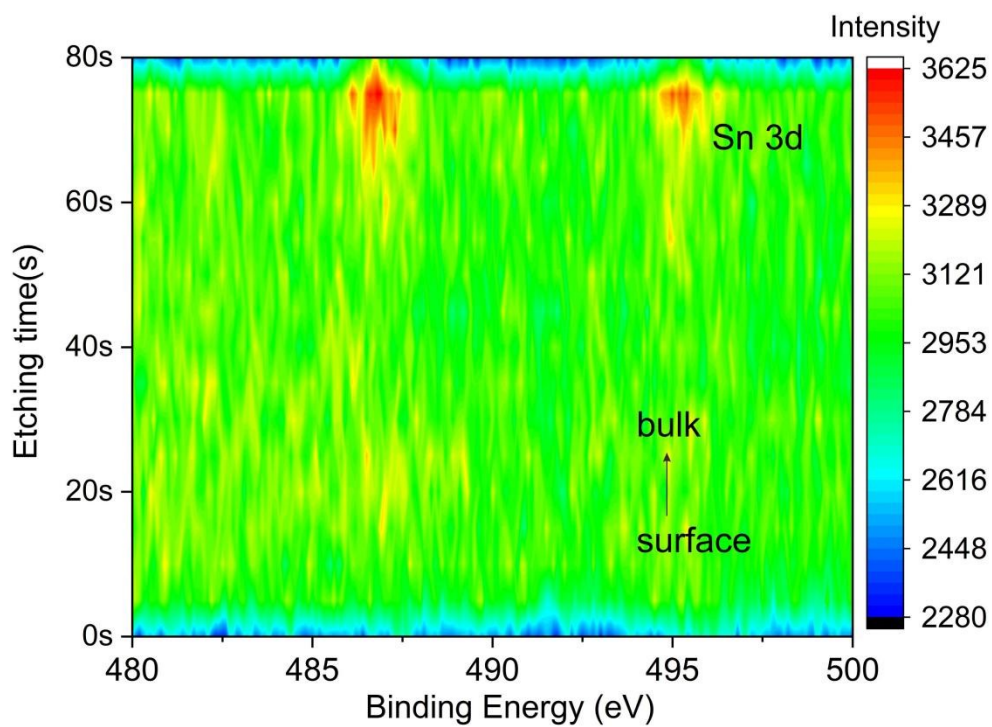


Fig. S3 The contour maps of XPS depth profile for the target perovskite film.

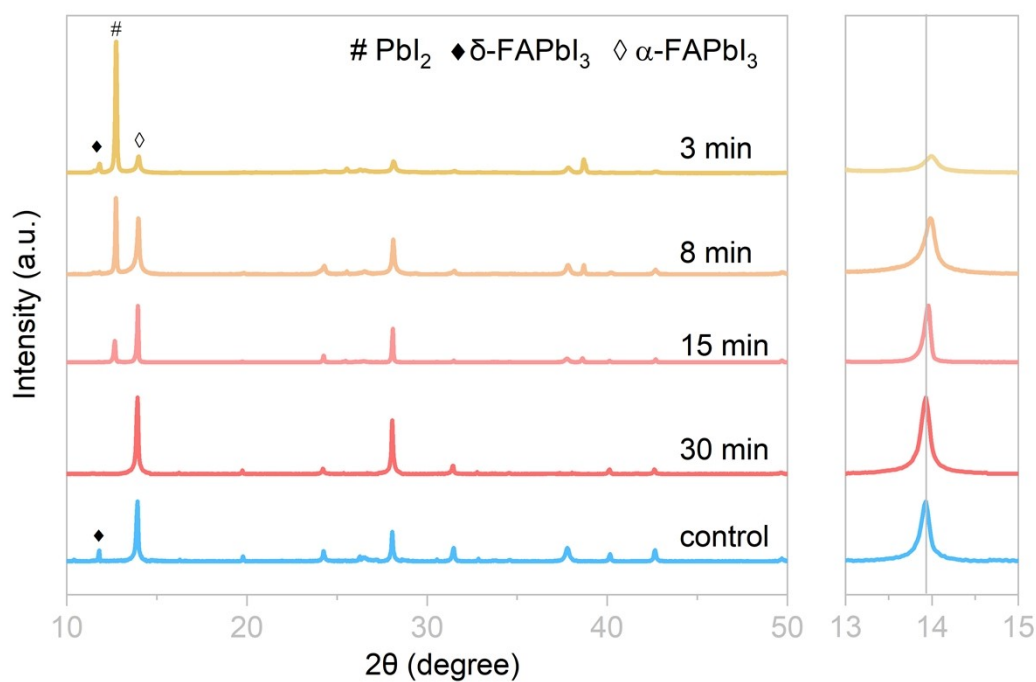


Fig. S4 XRD of the reaction at 3 min, 8 min, 15 min and 30 min perovskite films formed by $\text{PbI}_2/\text{PbCl}_2$ reacting with FAI vapor.

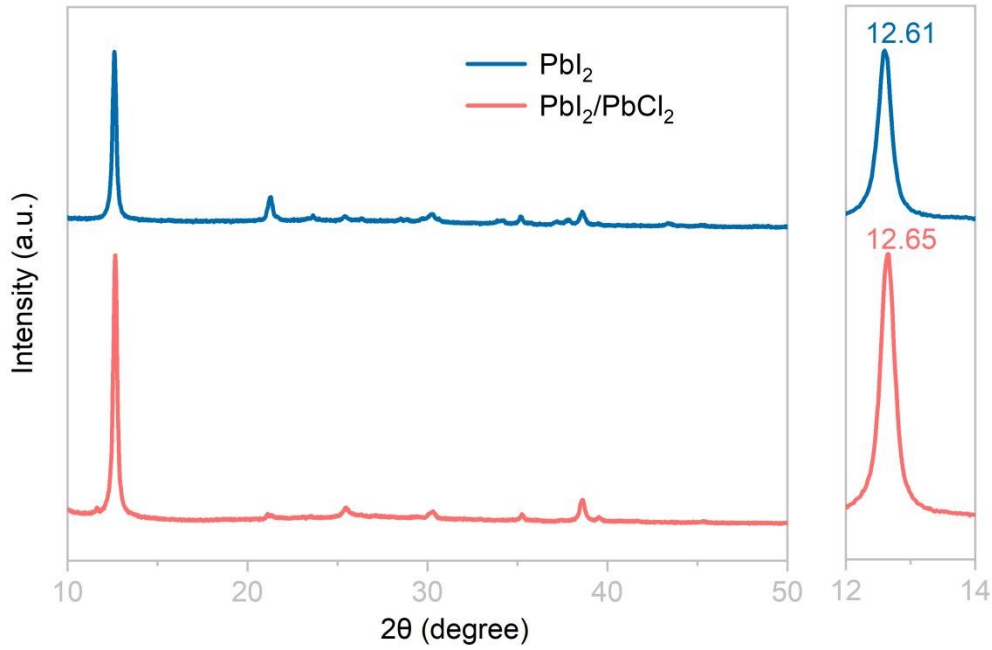


Fig. S5 XRD patterns of PbI_2 and $\text{PbI}_2/\text{PbCl}_2$.

Table S1 Time parameters derived from the fitting results of the transient TR-PL decay curves shown in Fig. 3b.

samples	τ_1		τ_2		τ_{avg}
	Value (ns)	Rel. (%)	Value (ns)	Rel. (%)	Value (ns)
control	15.83	7.2	259.50	92.8	241.96
target	28.85	4.2	363.20	95.8	349.16

Table S2 Parameters of best device performance of each case under reverse and forward scanning in Fig. 4a.

<i>samples</i>		V_{OC} (V)	J_{SC} (mA/cm ²)	FF	PCE (%)	HI	PCE _{avg}
target	reverse	1.10	24.89	79.8	22.89	0.031	22.22
	forward	1.09	24.77	77.6	22.03		
control	reverse	1.01	24.59	73.3	19.05	0.052	18.61
	forward	0.99	24.45	66.2	17.65		

$$N_{trap} = \frac{2\varepsilon_0\varepsilon_r V_{TFL}}{e}$$

Equation S1

Where the L is the thickness of the perovskite films, ε_r and ε_0 are the relative dielectric constant of corresponding perovskite film and vacuum permittivity, respectively, and e is the elementary charge.

$$\tau_n = -k_B T e^{-1} (dV_{OC}/dt)^{-1}$$

Equation S2

Where k_B is the Boltzman constant, T is the temperature and e is the elementary charge. The relationship of extracted electron lifetime τ_n of the injected carriers in the devices and V_{OC} of the PSCs based on control and target films.