

Supporting Information

Enhancing the stability of perovskite solar cells and modules by two-dimensional (PM)₂PbI₂Cl₂

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1. Experimental

Materials: Lead(II) iodide (PbI₂, Purity >99.99%) was purchased from Xi'an Yilante; Formamidinium Iodide (FAI, Purity >99.99%) was purchased from Greatcell Solar Materials; Methylamine hydrochloride (MACl, Purity >99.99%) and (Al₂O₃, Particle size <50nm) were purchased from Sigma-Aldrich; Poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA, Purity >99.5%), 2-Phenylethanamine hydroiodide (PEAI, Purity >99%) and Fullerene-C60 (C60, Purity >99%) were purchased from Xi'an Baolait Company; N,N-Dimethylformamide (DMF, Purity >99.8%), Dimethyl sulfoxide (DMSO, Purity >99.8%), Isopropyl alcohol (IPA, Purity >99.5%) and Chlorobenzene (CB, Purity >99%) were purchased from Maclean; N-Cyclohexyl-2-pyrrolidone (CHP, Purity >99%) and pentamidine hydrochloride (PMCl, Purity >99%) were purchased from Aladdin. All Chemicals were used as received without further purification.

Solution preparation: The 3D perovskite solution was prepared by mixing PbI₂ and FAI in a 1:1 ratio in the mixed solvent of DMF, DMSO and CHP (8:1:1, v:v:v), and

10%M MA₂Cl as the additive. The 2D perovskite solution was prepared by dissolving PM₂Cl and PbI₂ in a 1:2 ratio in a mixed solvent of DMF and DMSO (4:1). The final 2D/3D precursor solution was obtained by mixing the 2D solution and 3D solution in a volume ratio of 1:4.

Device Fabrication: The ITO-coated substrates (2.5×2.5cm²) were ultrasonicated twice in water and detergent for 12 min each. The substrates underwent UV-ozone treatment for 10min. PTAA (1.5 mg/mL in chlorobenzene) was spin-coated onto the ITO at 4000 rpm for 30s. The layer was then annealed at 100°C for 10min. The PTAA substrate was spin-coated with 180μL of Al₂O₃ (in a volume ratio of 1:80 with IPA) and annealed at 100°C for 10min to form the hole transport layer (HTL). Next, 150μL of the perovskite precursor was spin-coated at 4000 rpm with an acceleration rate of 800 rpm/s for 40s, and 300μL of EA was added as an anti-solvent at 30 s. The sample was then annealed at 105°C for 30min. Afterwards, spin coat 100μL of PEAI (0.5mg/ml in IPA) passivation solution at 4000 rpm for 30 seconds, followed by annealing at 105°C for 10min. Next, 30nm of C₆₀ was evapo-rated, followed by the deposition of 20nm of SnO₂ using atomic layer deposition (Super ALD Corp. E300). Finally, 150nm of Ag electrode was evaporated sequentially to complete the device. Perform other tests after heat treatment at 100°C for 10 minutes.

Characterization: The current-voltage (J-V) curves of PSCs were characterized using a solar simulator with a spectrum of 1.5G. The light intensity was calibrated to 1000 W/m² using a standard silicon solar cell, and the illumination area was controlled using a black mask with an aperture area of 0.09 cm². The crystal structure of the perovskite film was characterized using a Bruker D8 Advance instrument. The time-resolved photoluminescence (TRPL) lifetime of the perovskite film was measured using a fluorescence spectrometer (FLS980) from Edinburgh Instruments. The external quantum efficiency (EQE) of the PSCs was measured using a spectral response system (Enlitech QE-R). The capacitance-voltage (C-V) curves of the PSCs were measured using an electrochemical workstation(ZAHNER GIMPS, Germany).

2. Figures and Tables

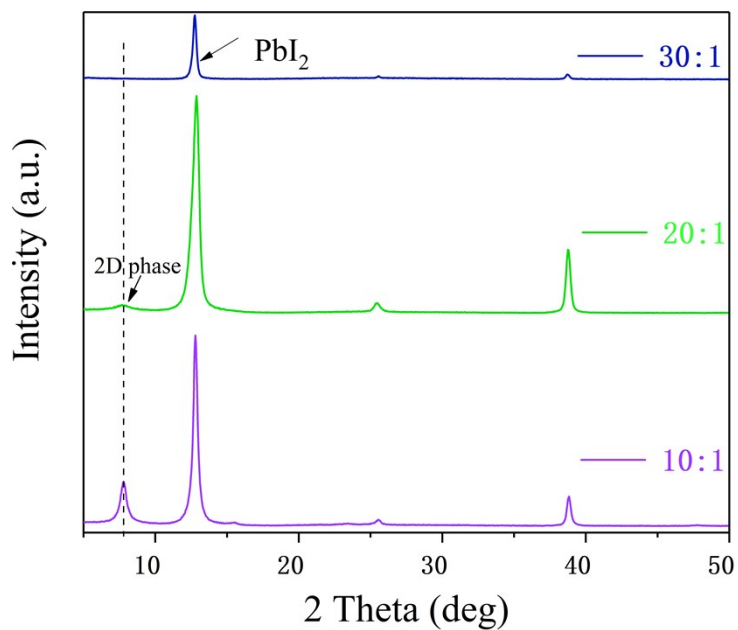


Fig. S1. XRD spectra of films prepared with different proportions of PbI₂ and PMCl.

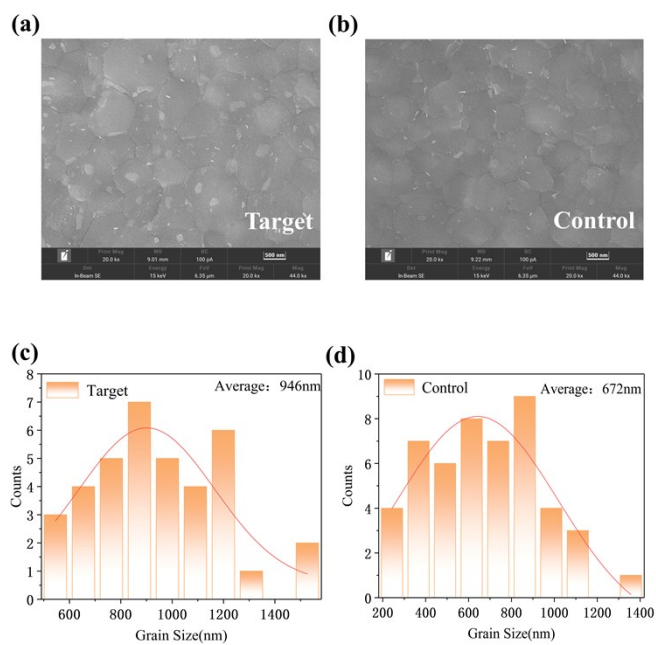


Fig. S2. a-d) SEM images and grain size distribution of perovskite films.

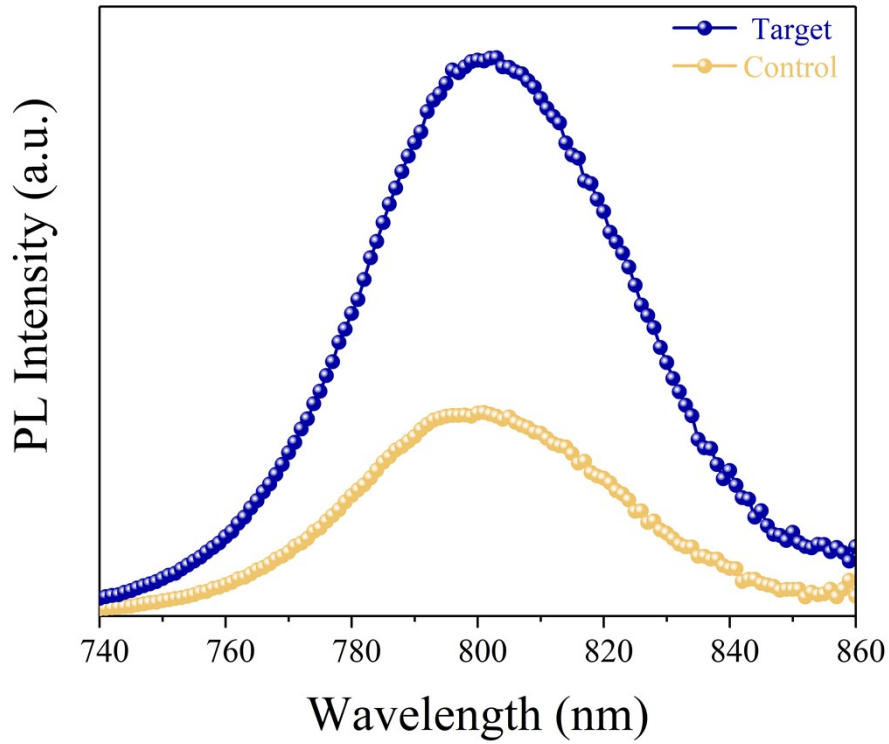


Fig. S3. Steady-state PL spectra of films incorporating bare FAPbI₃ (Control) and FAPbI₃ with 2D perovskite (Target).

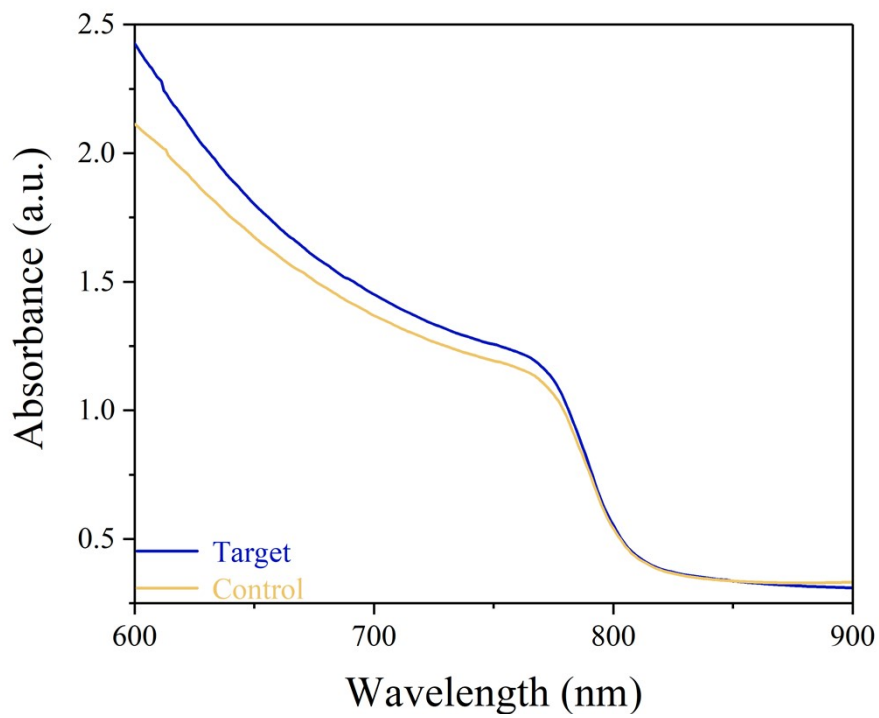


Fig. S4. The absorption spectra of PSCs incorporating bare FAPbI₃ (Control) and FAPbI₃ with 2D perovskite (Target).

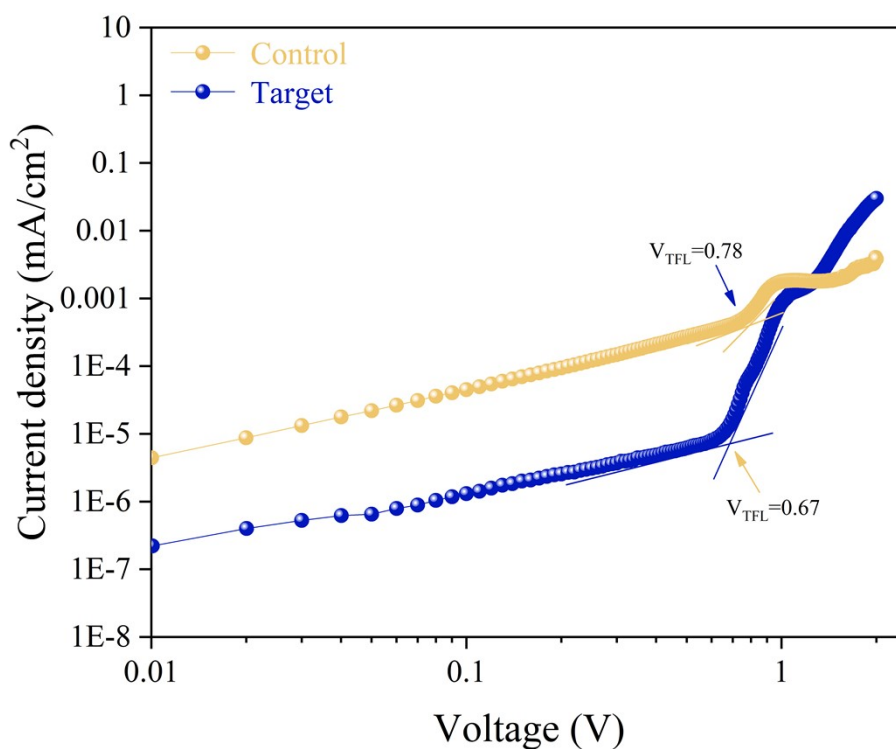


Fig. S5. J-V curves of PSCs incorporating bare FAPbI₃ (Control) and FAPbI₃ with 2D perovskite (Target) in the dark.

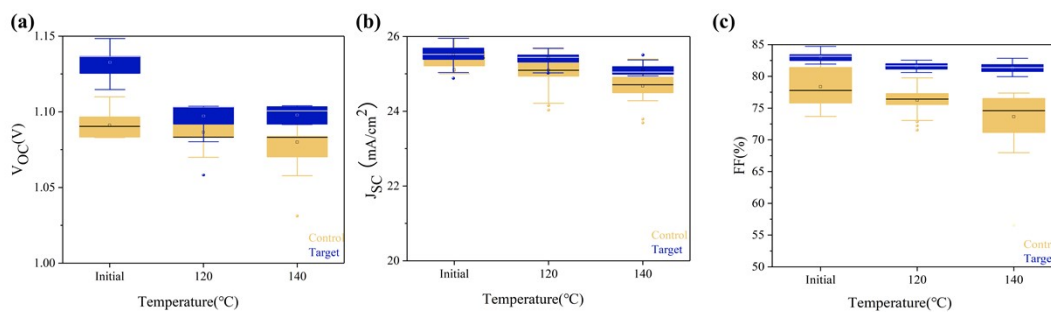


Fig. S6. statistics of V_{OC} (a), J_{SC} (b) and FF (c) of PSCs incorporating bare FAPbI₃ (Control) and FAPbI₃ with 2D perovskite (Target).

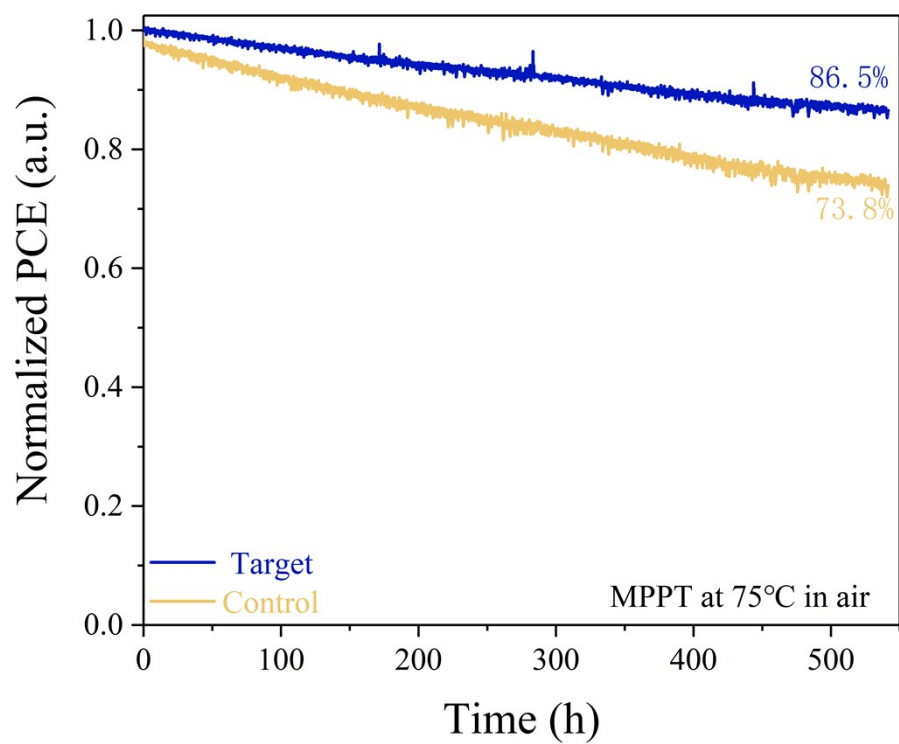


Fig. S7. Comparison of the stability of corresponding packaged module under continuous one-sun illumination at 75°C.

Table S1. Average efficiency of modules incorporating bare FAPbI₃ and FAPbI₃ with 2D perovskite (16 modules each).

	V_{OC} (V)	J_{SC} (mA/cm ²)	FF (%)	PCE (%)	σ (standard deviations)
Target	7.956	3.562	0.782	22.457	0.183
Control	7.914	3.540	0.733	20.526	0.516