Electronic Supporting Information

Perovskite-type Stabilizer for Efficient and Stable Formamidinium-based Lead Iodide Perovskite Solar Cells

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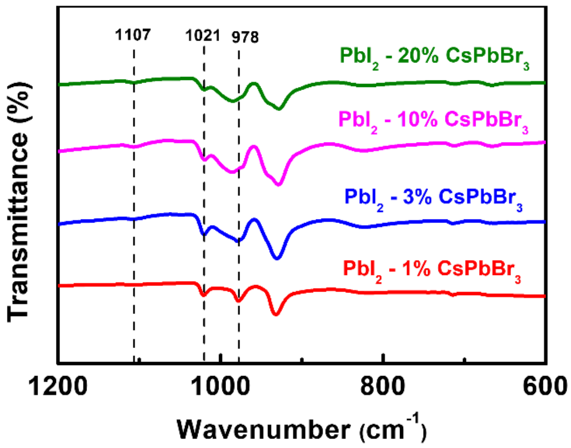

Figure S1. FTIR spectra of PbI_2 films with $CsPbBr_3$. The doping percentage of $CsPbBr_3$ is defined by the molar percentage of lead.

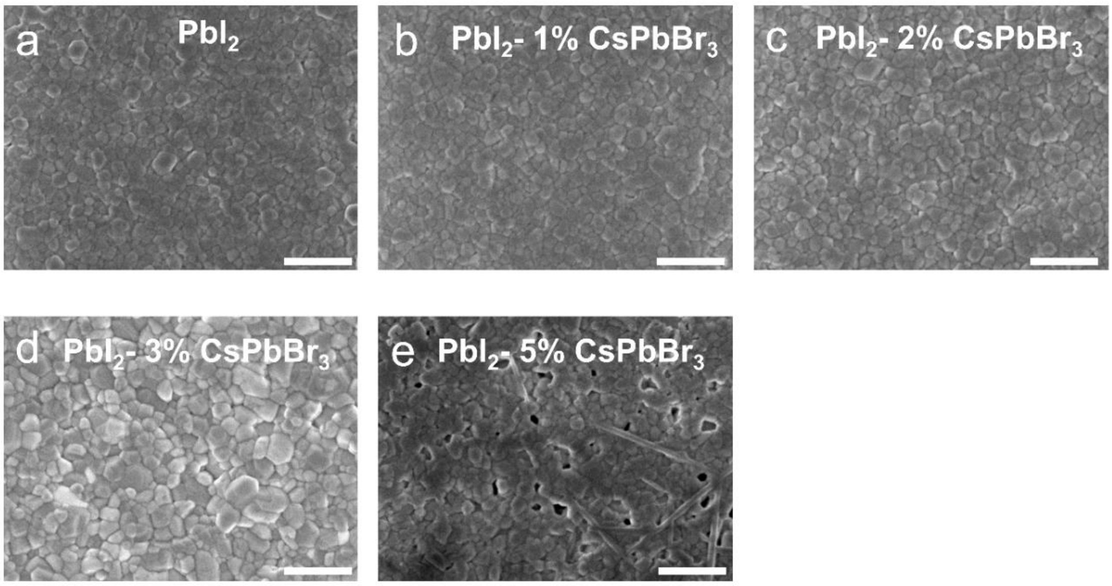

Figure S2. Top-view SEM images of PbI_2 films a) w/o, b) with 1%, c) with 2%, d) with 3%, and e) with 5% $CsPbBr_3$. Scale bar in a) b) c) d) e) represents 500 nm.

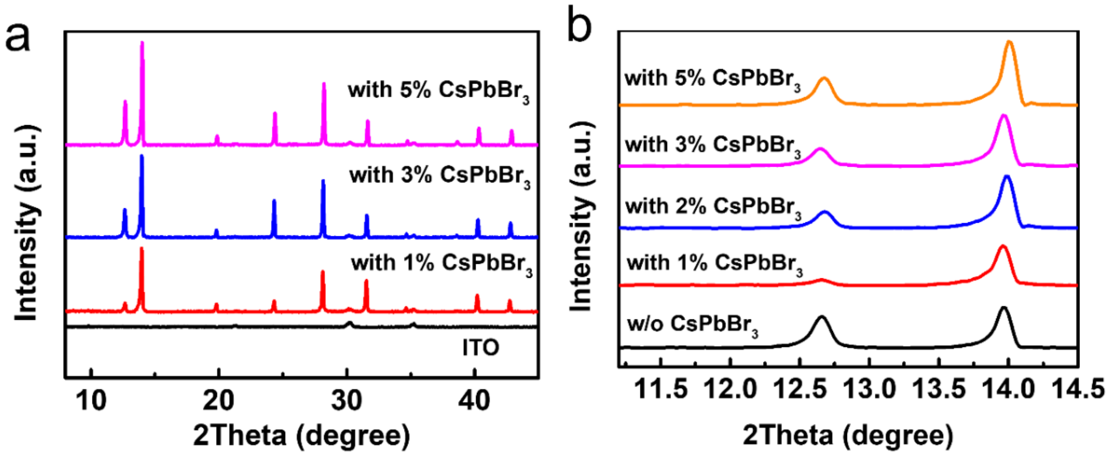

Figure S3. a) XRD patterns of $FAPbI_3$-based perovskite films stabilized with $CsPbBr_3$. b)Evolution of XRD peak intensity at around 12.7 and 14° of the perovskite films w/o and with $CsPbBr_3$. The samples are deposited on ITO.

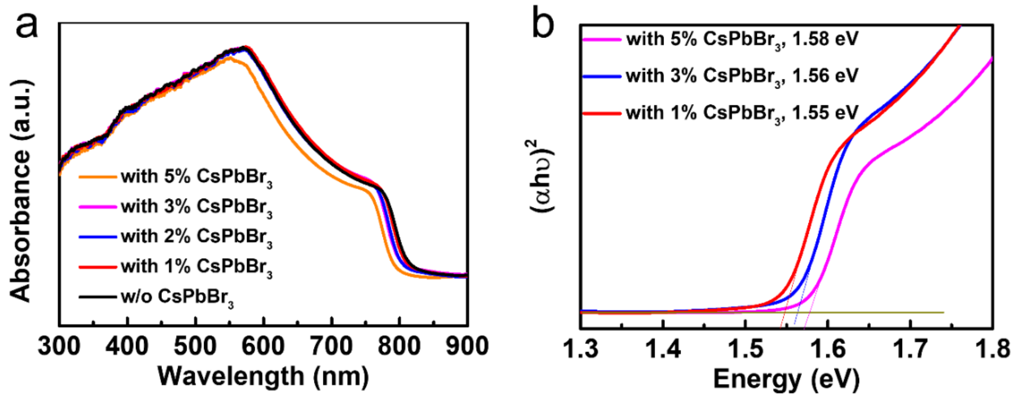

Figure S4. a) UV-vis absorption spectra of $FAPbI_3$-based perovskite films w/o and with $CsPbBr_3$. b) Tuac plot of perovskite films with $CsPbBr_3$.

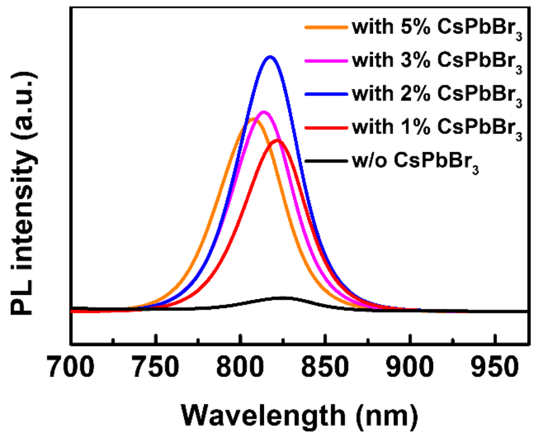

Figure S5. Steady-state PL spectra of $FAPbI_3$-based perovskite films w/o and with $CsPbBr_3$.

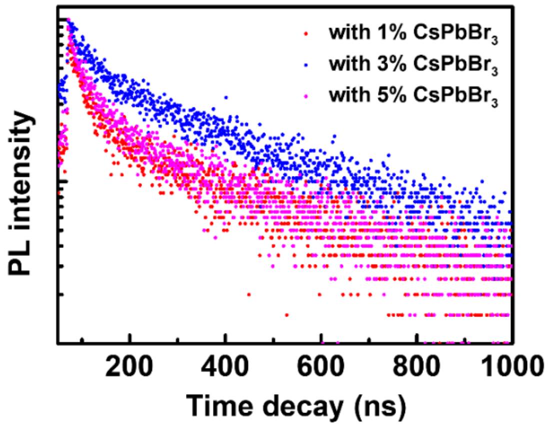

Figure S6. TRPL decay of $FAPbI_3$-based perovskite films with $CsPbBr_3$.

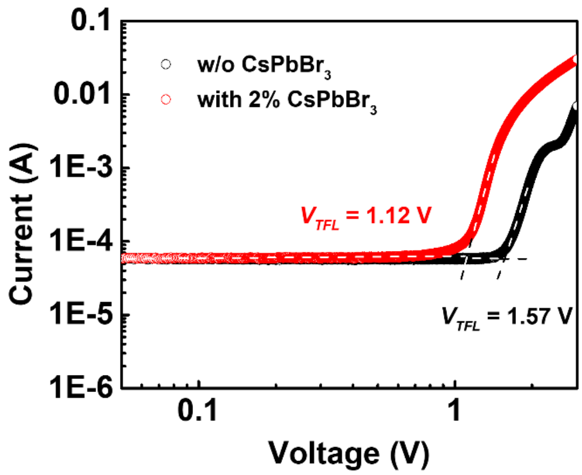

Figure S7. SCLC results of the $FAPbI_3$-based devices w/o and with $CsPbBr_3$. To calculate the trap density, the thickness of both perovskites is determined to be 680 nm by cross-sectional SEM and the employed dielectric constant ε is 46.9 (ref. 21).

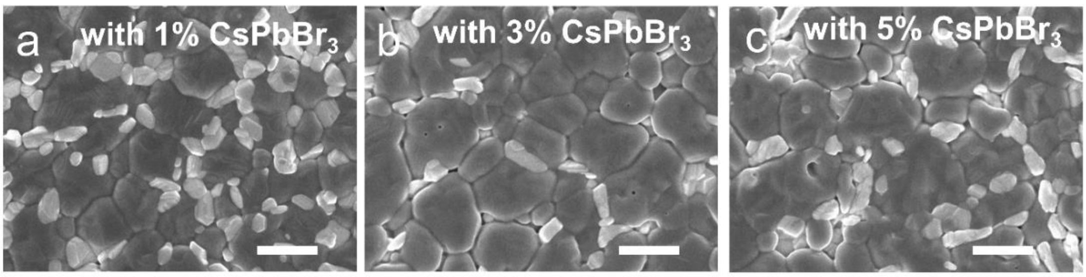

Figure S8. Top-view SEM images of $FAPbI_3$-based perovskite films a) with 1%, b) with 3%, c) with 5% $CsPbBr_3$. Scale bar in a) b) c) represents 1 μm.

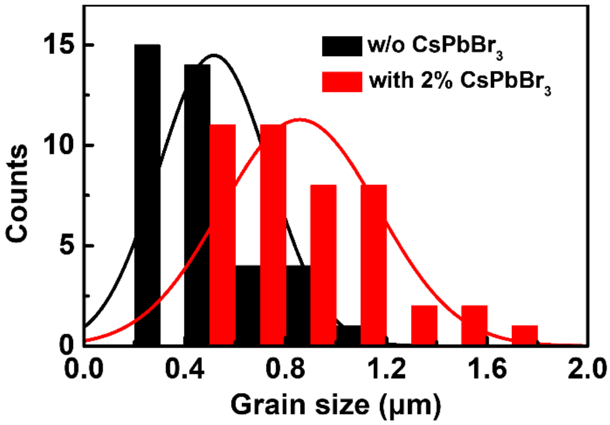

Figure S9. Grain-size distribution of the $FAPbI_3$-based perovskite films w/o and with 2% $CsPbBr_3$.

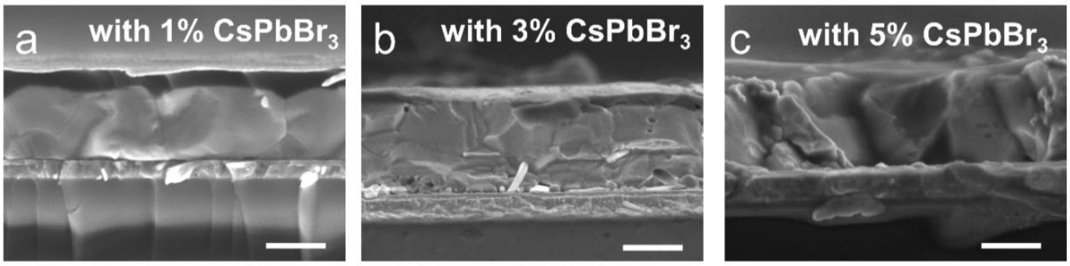

Figure S10. Cross-sectional SEM images of $FAPbI_3$-based PSCs a) with 1%, b) with 3%, c) with 5% $CsPbBr_3$. Scale bar in a) b) c) represents 500 nm.

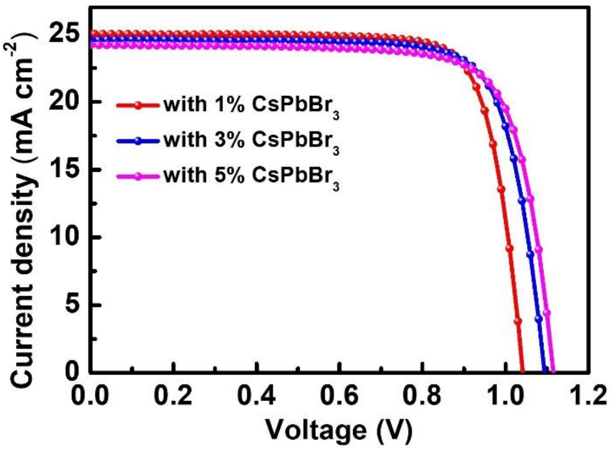

Figure S11. *J–V* curves of $FAPbI_3$-based PSCs with $CsPbBr_3$.

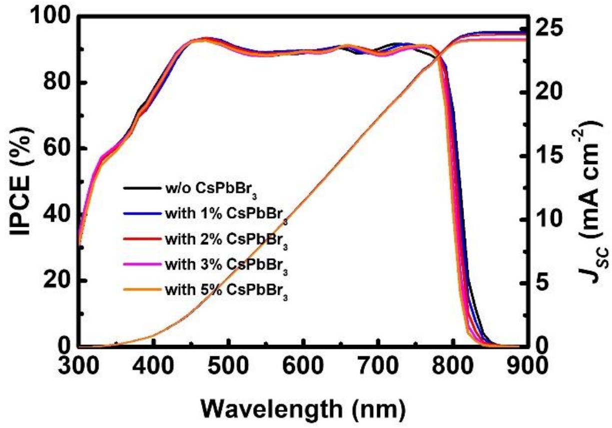

Figure S12. IPCE spectra and integrated J_{SC} of $FAPbI_3$-based PSCs w/o and with $CsPbBr_3$.

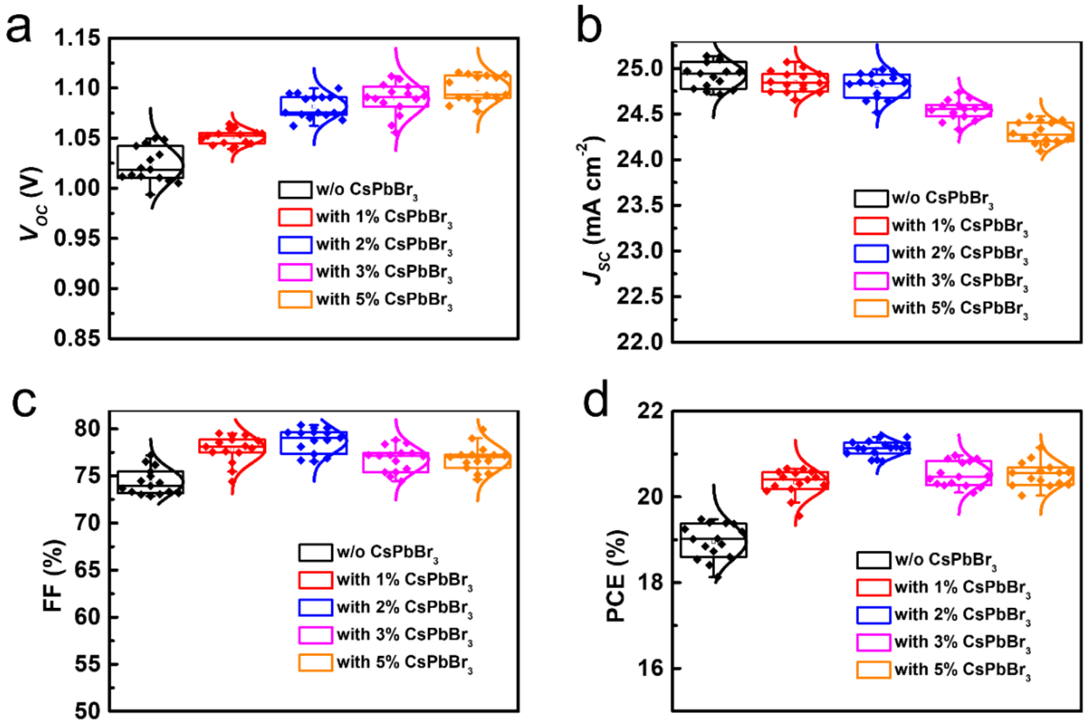

Figure S13. Statistics of photovoltaic metrics of 15 individual $FAPbI_3$-based PSCs w/o and with $CsPbBr_3$. a) V_{OC}, b) J_{SC}, c) FF, and d) PCE.

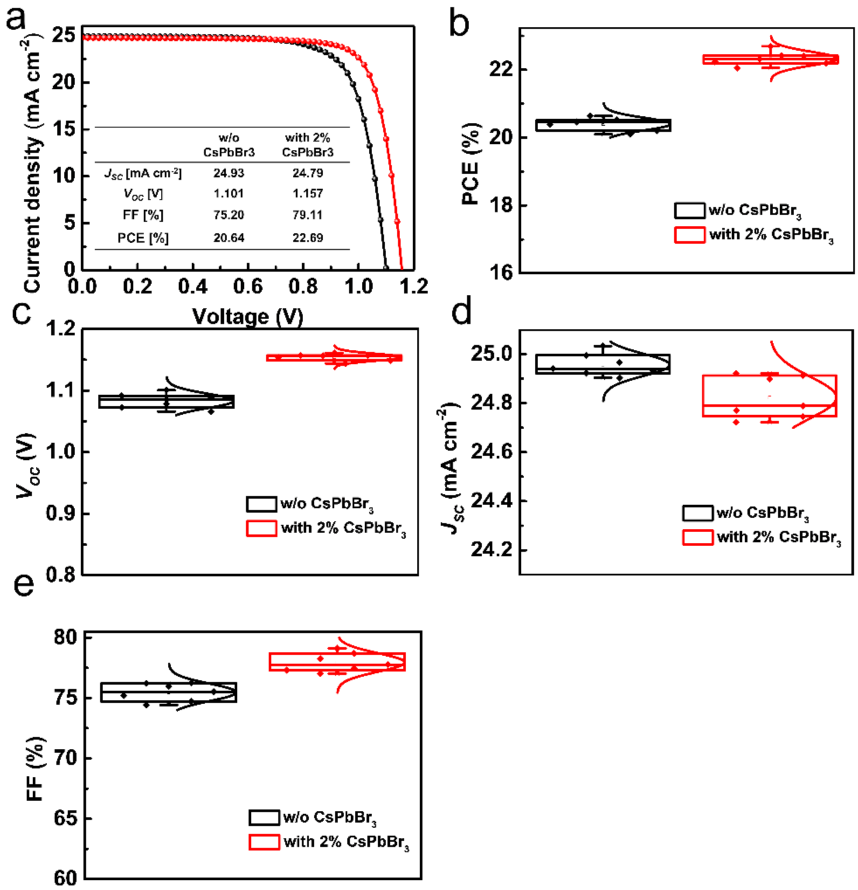

Figure S14. Photovoltaic performance of the $FAPbI_3$-based PSCs w/o and with 2% $CsPbBr_3$ optimized with surface passivation of NMAI. a) *J–V* curves. Statistics of b) PCE, c) V_{OC}, d) J_{SC}, and e) FF.

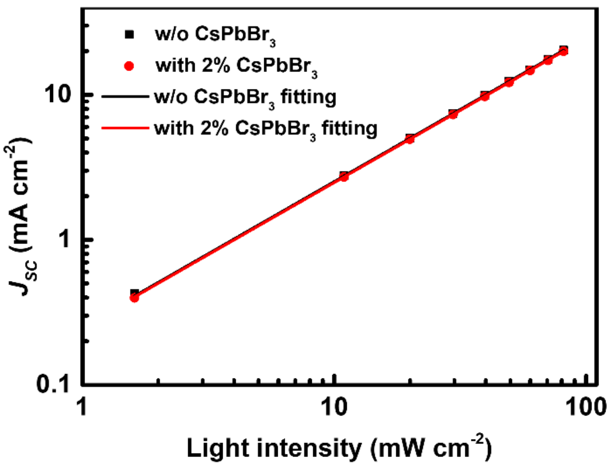

Figure S15. The dependency of J_{SC} versus illumination intensity of $FAPbI_3$-based PSCs w/o and with 2% $CsPbBr_3$.

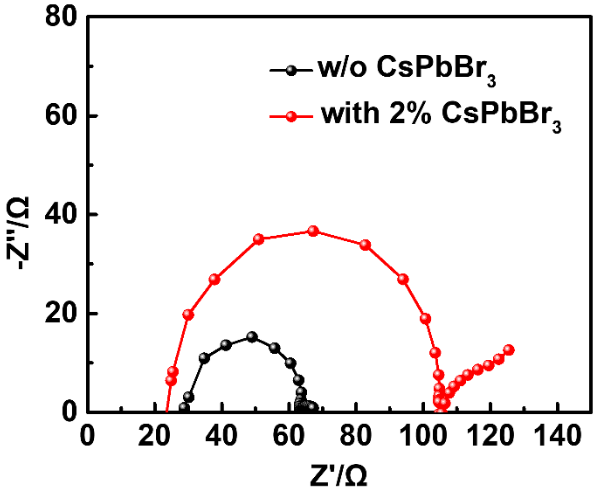

Figure S16. EIS of $FAPbI_3$-based PSCs w/o and with 2% $CsPbBr_3$. The applied bias was 1 V (close to the open circuit) and the illumination intensity was 10 mW cm^{-2} (0.1 sun). These testing conditions were employed to emphasize the recombination characteristics.

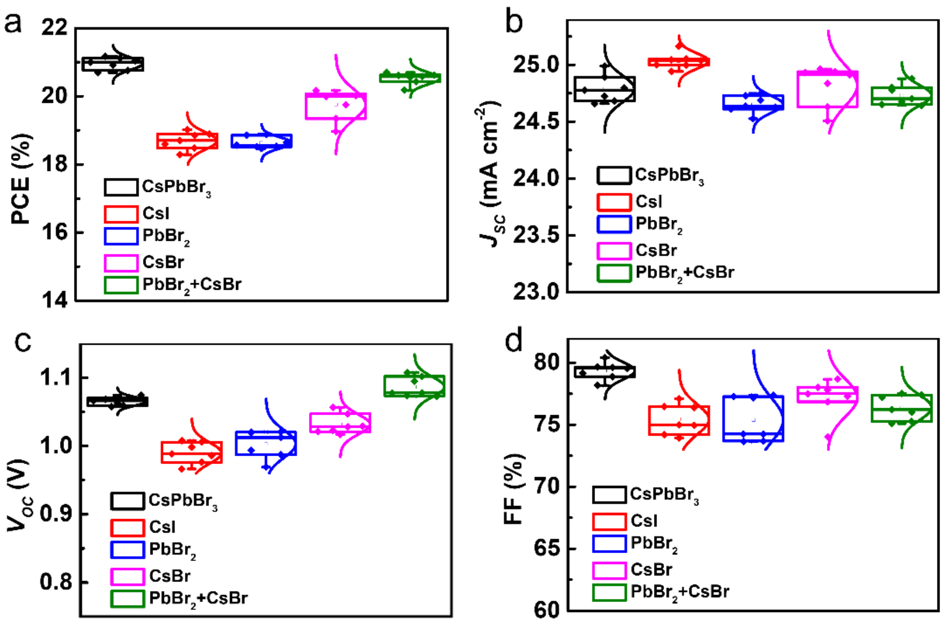

Figure S17. Photovoltaic metrics statistics of $FAPbI_3$-based PSCs stabilized with perovskite-type stabilizer ($CsPbBr_3$), precursor-type stablizers (CsI, $PbBr_2$, or CsBr) and corresponding double precursor-type stabilizers ($PbBr_2$+CsBr). The ralative content of stabilizers are fixed at 2%. a) PCE, b) J_{SC}, c) V_{OC}, and d) FF.

Table S1. Fitted parameters of TRPL results of $FAPbI_3$-based perovskite films w/o and with $CsPbBr_3$.

Devices	w/o $CsPbBr_3$	with 1% $CsPbBr_3$	with 2% $CsPbBr_3$	with 3% $CsPbBr_3$	with 5% $CsPbBr_3$
τ_1 (ns)	17.8	27.5	61.3	39.5	34
τ_2 (ns)	326.6	359.8	371.7	358	344
A_1	32.8	7.6	1.0	2.9	4.8
A_2	0.4	0.3	0.6	0.6	0.3

Table S2. Photovoltaic parameters of $FAPbI_3$-based PSCs with $CsPbBr_3$.

Devices	J_{SC} [mA cm^{-2}]	V_{OC} [V]	FF [%]	PCE [%]
with 1% $CsPbBr_3$	25.01	1.042	78.85	20.54
with 3% $CsPbBr_3$	24.58	1.095	77.43	20.83
with 5% $CsPbBr_3$	24.20	1.116	76.60	20.68

Table S3. Accuracy of J_{SC} measured by *J–V* curves evaluated by the mismatch between integrated J_{SC} values from IPCE and extracted J_{SC} from *J–V* curves.

Devices	w/o $CsPbBr_3$	with 1% $CsPbBr_3$	with 2% $CsPbBr_3$	with 3% $CsPbBr_3$	with 5% $CsPbBr_3$
Integrated *Jsc* (mA cm^{-2})	24.79	24.72	24.55	24.19	24.08
Jsc extracted from *J–V* (mA cm^{-2})	25.12	25.01	24.83	24.58	24.20
Mismatch (%)	1.3	1.2	1.1	1.6	0.5

Table S4. Summary of representative efficiency and stability of $FAPbI_3$-based PSCs stabilized with different types of additives reported in the past three years.

Stabilizer	Type	Cs-containing	Champion PCE (%)	Operational stability	Published year	Ref
MACl	Precursor	No	23.48% (Certified)	T_{90} =300 h @ 40 ℃(heat stability)	2019	[1]
MACl	Precursor	No	23.1%	T_{80} =150 h @ AM 1.5G, N_2 (light stability)	2021	[2]
MAI & MACl	Precursor	No	23.32% (Certified)	T_{80} ≈500 h @85 ℃, N_2 (heat stability)	2019	[3]
$MDACl_2$ (MDA = $^{+}H_3N\text{-}CH_2\text{-}NH_3^{+}$) & MACl	Precursor	No	23.7% (Certified)	T_{90} =600 h @ AM 1.5G in the air with encapsulation (light stability)	2019	[4]
CsI & GAI (GA = $HNC(NH_2)_2$)	Precursor	Yes	23.5%	T_{80} ≈250 h @ 0.8 sum (light stability)	2020	[5]
$Cs_{0.10}FA_{0.78}MA_{0.12}PbI_{2.55}Br_{0.45}$	Perovskite	Yes	21.7%	T_{60} =280 h @ AM 1.5G in N_2 (light stability)	2018	[6]
CsCl & $Cs_{0.10}FA_{0.78}MA_{0.12}PbI_{2.55}Br_{0.45}$	Perovskite	Yes	22.1%	T_{90} =4000 min @ AM 1.5G, N_2 (light stability)	2018	[7]
$Cs_{0.2}MA_{0.2}FA_{0.6}Pb(I_{0.22}Br_{0.78})_3$	Perovskite	Yes	21.5%	T_{80} =500 h @ AM 1.5G, N_2, (light stability)	2019	[8]
δ-$CsPbI_3$	Non-perovskite	Yes	20.45%	T_{70} =20 min @ AM 1.5G, Air (55%-60% RH) (light stability)	2019	[9]
$MAPbBr_3$ & MACl	Perovskite	No	22.51%	T_{97} =2600 h @ dark, 20% RH	2019	[10]
δ-$CsPbI_3$ & δ-$RbPbI_3$	Non-perovskite	Yes	22.30%	T_{92} =400 h @ AM 1.5G, N_2, (light stability)	2021	[11]
$MAPbBr_3$ & MACl	Perovskite	No	25.2% (Certified)	T_{80} =500 h @ AM 1.5G, 40℃ with encapsulation (light stability)	2021	[12]
$CsPbBr_3$ & MACl & MAI	Perovskite	Yes	23.34%	T_{80} =1153 h @ AM 1.5G, N_2, (light stability)	This work	

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