Supporting Information

Improve Efficiency and Stability of Pb-Sn Binary Perovskite Solar Cells by Cs Substitution

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Figure S1. $MASnI_3$ (a) and $CsSnI_3$ (b) perovskite films after toluene washing process before annealing.



Figure S2. Refined XRD spectra of MA_{1-y}Cs_yPb_{0.5}Sn_{0.5}I₃.



Figure S3. *J*-*V* curve of the champion device based on $MA_{0.9}Cs_{0.1}Pb_{0.5}Sn_{0.5}I_3$ perovskite measured under AM 1.5 illumination under reverse scan.



Figure S4. EQE of MA_{1-x}Cs_xPb_{0.5}Sn0.5I₃ (x=0, 0.1, 0.2, 0.3) PVSCs.



Figure S5. Tauc-plot analysis of MA_{0.9}Cs_{0.1}Pb_{1-x}Sn_xI₃ (x=0, 0.25, 0.5, 0.75, 1) perovskite films.



Figure S6. XPS analysis of MA_{0.9}Cs_{0.1}Pb_{1-x}Sn_xI₃ (x=0, 0.25, 0.5, 0.75, 1) films.



Figure S7. XRD spectra of MA_{0.9}Cs_{0.1}Pb_{1-x}Sn_xI₃ (x=0, 0.25, 0.5, 0.75, 1) films.



Figure S8. (a) UPS and (b) the corresponding energy diagram of $MA_{0.9}Cs_{0.1}Pb_{1-x}Sn_xI_3$ (x=0, 0.25, 0.5, 0.75, 1) films.



Figure S9. (a) XRD and (b) UV-Vis spectra of $FA_{0.8}Cs_{0.2}Pb_{1-x}Sn_xI_3$ (x=0.25, 0.5, 0.75, 1) films.



Figure S10. EQE of FA_{0.8}Cs_{0.2}Pb_{1-x}Sn_xI₃ (x=0.25, 0.5, 0.75, 1) PVSCs.

Crystal Lattice	a (Å)	с (Å)	Volume (Å ³)
MAPb _{0.5} Sn _{0.5} I ₃	8.85	12.53	981.4
$MA_{0.9}Cs_{0.1}Pb_{0.5}Sn_{0.5}I_3$	8.84	12.54	979.7
$MA_{0.8}Cs_{0.2}Pb_{0.5}Sn_{0.5}I_3$	8.83	12.49	973.3
$MA_{0.7}Cs_{0.3}Pb_{0.5}Sn_{0.5}I_3$	8.81	12.46	968.6

Table S1. Lattice parameters and volume of MA_{1-x}Cs_xPb_{0.5}Sn_{0.5}I₃ (x=0, 0.1, 0.2, 0.3)

Table S2. Performance of $MA_{1-x}Cs_xPb_{0.5}Sn_{0.5}I_3$ (x=0, 0.1, 0.2, 0.3) PVSCs measured under AM 1.5 illumination under reverse scan.

Composition	<i>V_{oc}</i> (V)	J _{SC} (mA/cm ²)	FF (%)	PCE (%)
MAPb _{0.5} Sn _{0.5} I ₃	0.62	18.65	55.0%	6.36
$MA_{0.9}Cs_{0.1}Pb_{0.5}Sn_{0.5}I_3$	0.70	23.32	61.7%	10.07
$MA_{0.8}Cs_{0.2}Pb_{0.5}Sn_{0.5}I_3$	0.60	20.58	60.7%	7.50
$MA_{0.7}Cs_{0.3}Pb_{0.5}Sn_{0.5}I_3$	0.39	14.96	50.4%	2.94

Table S3. Performance of $FAPb_{1-x}Sn_xI_3$ and $FA_{0.8}Cs_{0.2}Pb_{1-x}Sn_xI_3$ (x=0.25, 0.5, 0.75, 1) PVSCs measured under AM 1.5 illumination with reverse scan.

Composition	V _{oc} (V)	J_{SC} (mA/cm ²)	FF (%)	PCE (%)
FAPb _{0.75} Sn _{0.25} I ₃	0.75	19.48	70.3%	10.27
FAPb _{0.5} Sn _{0.5} I ₃	0.60	21.52	65.4%	8.44
FAPb _{0.25} Sn _{0.75} I ₃	0.46	20.43	56.1%	5.27
FASnI ₃	0.04	11.73	23.4%	0.11
FA _{0.8} Cs _{0.2} Pb _{0.75} Sn _{0.25} I 3	0.84	22.73	75.7%	14.46
FA _{0.8} Cs _{0.2} Pb _{0.5} Sn _{0.5} I ₃	0.70	24.30	68.3%	11.63
FA _{0.8} Cs _{0.2} Pb _{0.25} Sn _{0.75} I 3	0.56	24.44	57.2%	7.83
FA _{0.8} Cs _{0.2} SnI ₃	0.24	16.05	35.8%	1.38