Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Improved Stability and Efficiency of Perovskite/Organic Tandem Solar Cells with an All-inorganic Perovskite Layer

Xin Wu,[a] Yizhe Liu,[a] Feng Qi,[a] Francis Lin,[a] Huiting Fu,[b] Kui Jiang,[b] Shengfan Wu,[a] Leyu Bi,[a] Deng Wang,[b]

Fang Xu,[c] Alex. K.-Y. Jen[a,b] and Zonglong Zhu*[a]

a. Department of Chemistry, City University of Hong Kong, Kowloon 999077, Hong Kong. Email: zonglzhu@cityu.edu.hk

b. Department of Materials Science and Engineering, City University of Hong Kong, Kowloon 999077, Hong Kong.

c. Center for Advanced Material Diagnostic Technology, College of Engineering Physics, Shenzhen Technology University, Shenzhen 518118, People's Republic of China.

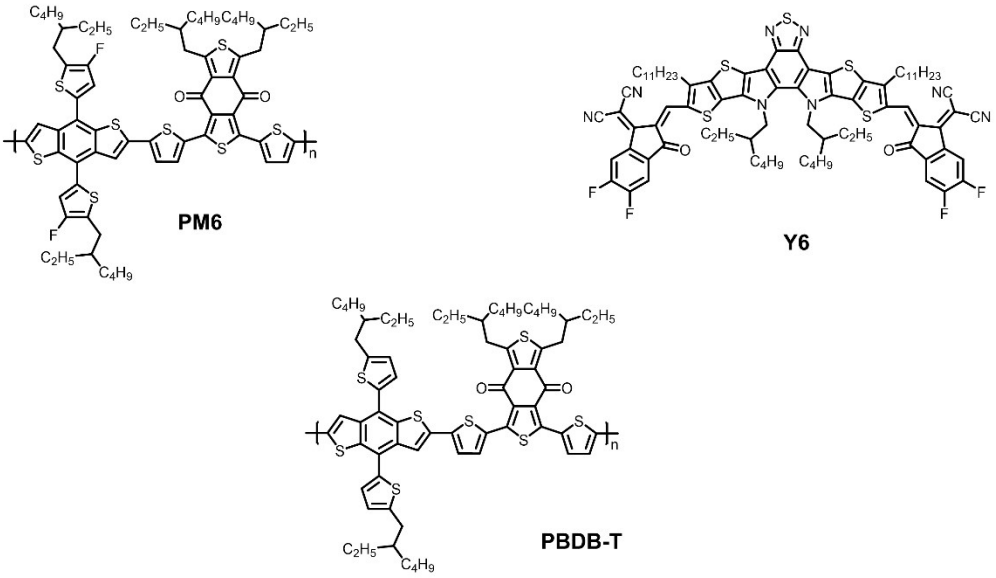

Figure S1. Chemical structures of PM6, Y6 and PBDB-T.

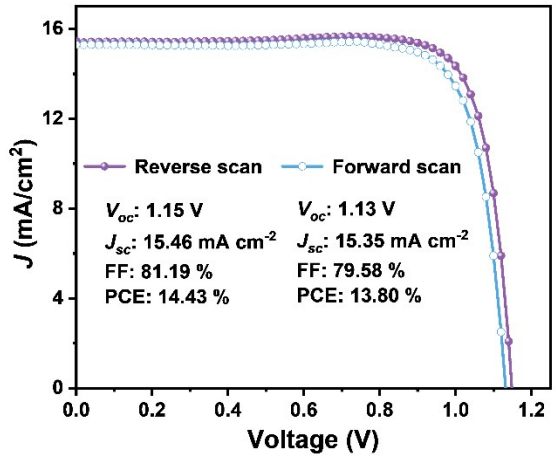

Figure S2. The J-V curves of champion device with PMACl passivation measured at reverse and forward scans under AM 1.5G illumination.

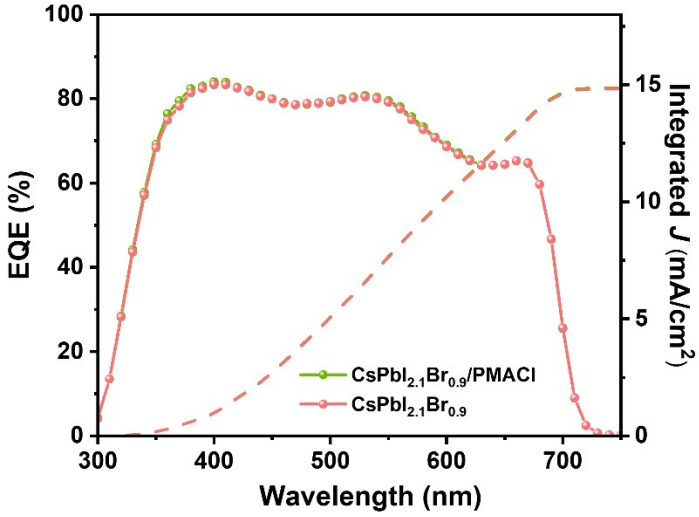

Figure S3. EQE spectra of $CsPbI_{2.1}Br_{0.9}$ and $CsPbI_{2.1}Br_{0.9}$/PMACl-based devices.

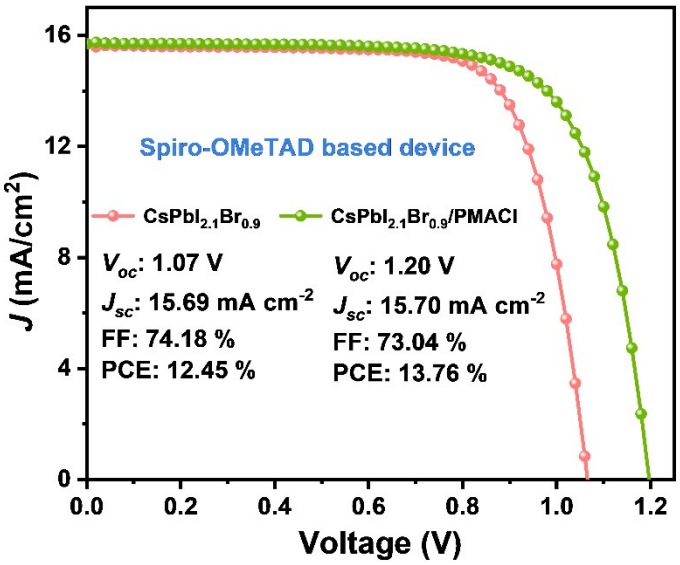

Figure S4. *J-V* curves of devices with structure of ITO/SnO_2/$CsPbI_{2.1}Br_{0.9}$/Spiro-OMeTAD/MoO_3/Ag.

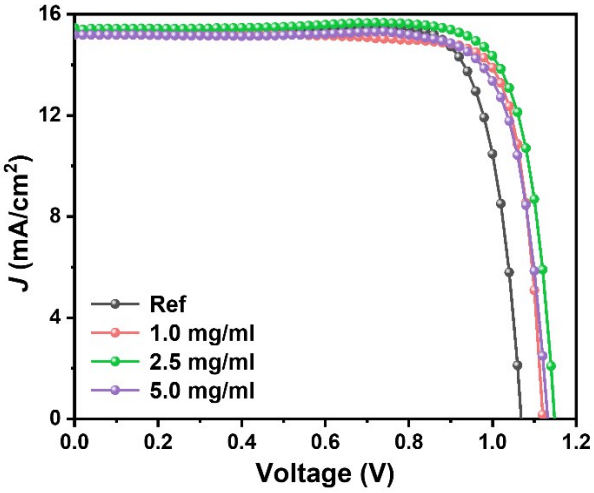

Figure S5. *J-V* curves of $CsPbI_{2.1}Br_{0.9}$ -based devices treated with PMACl solution of different concentration.

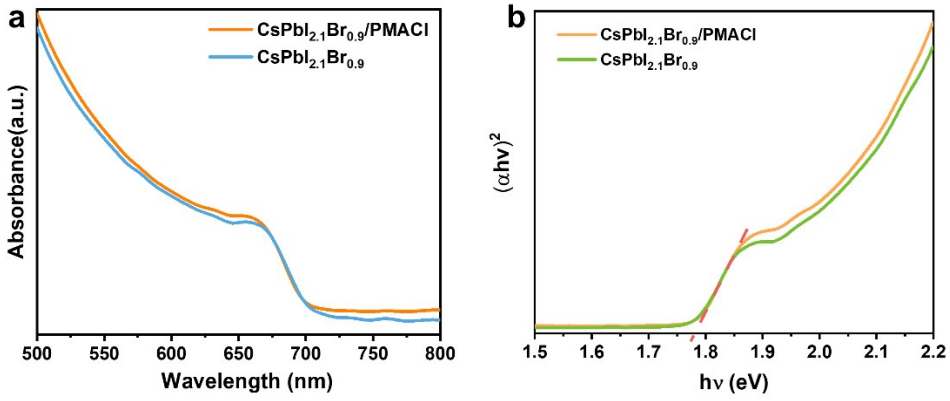

Figure S6. (a) The UV–vis absorption profiles of $CsPbI_{2.1}Br_{0.9}$ and $CsPbI_{2.1}Br_{0.9}$/PMACl films; (b) Tauc plots of $CsPbI_{2.1}Br_{0.9}$ and $CsPbI_{2.1}Br_{0.9}$/PMACl films.

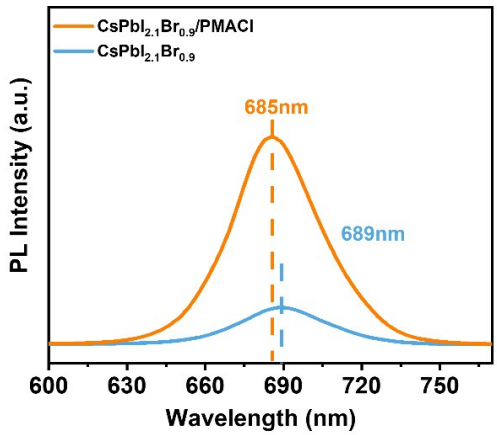

Figure S7. PL spectra of $CsPbI_{2.1}Br_{0.9}$ and $CsPbI_{2.1}Br_{0.9}$/PMACl films on ITO/SnO_2 substrate.

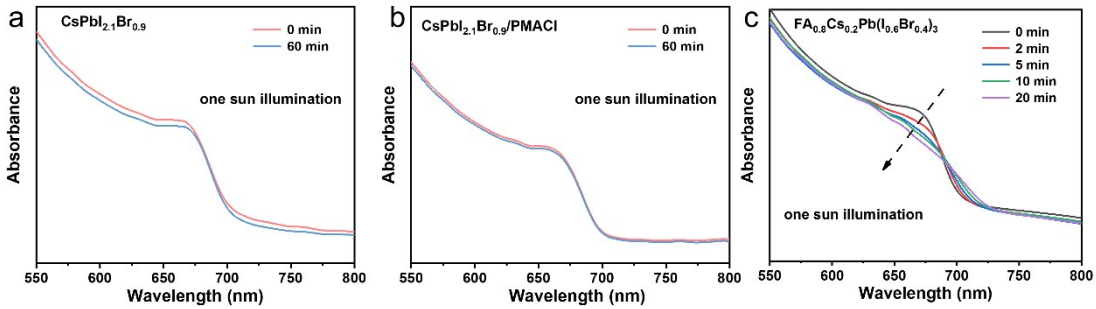

Figure S8. The UV–vis absorption profiles of (a) $CsPbI_{2.1}Br_{0.9}$, (b) $CsPbI_{2.1}Br_{0.9}$/PMACl, and (c) $FA_{0.8}Cs_{0.2}Pb(I_{0.6}Br_{0.4})_3$ films after one sun illumination for different time.

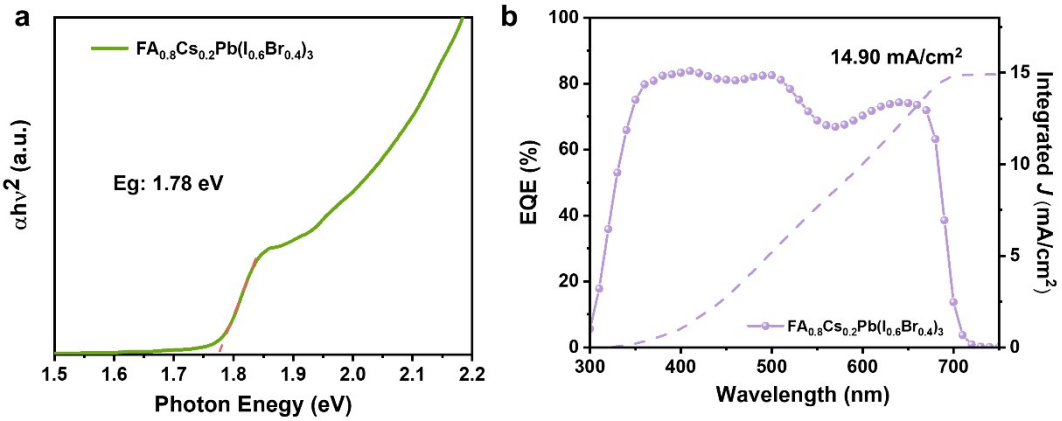

Figure S9. (a) Tauc plot of $FA_{0.8}Cs_{0.2}Pb(I_{0.6}Br_{0.4})_3$ film; (b) EQE spectrum of $FA_{0.8}Cs_{0.2}Pb(I_{0.6}Br_{0.4})_3$-based single-junction device.

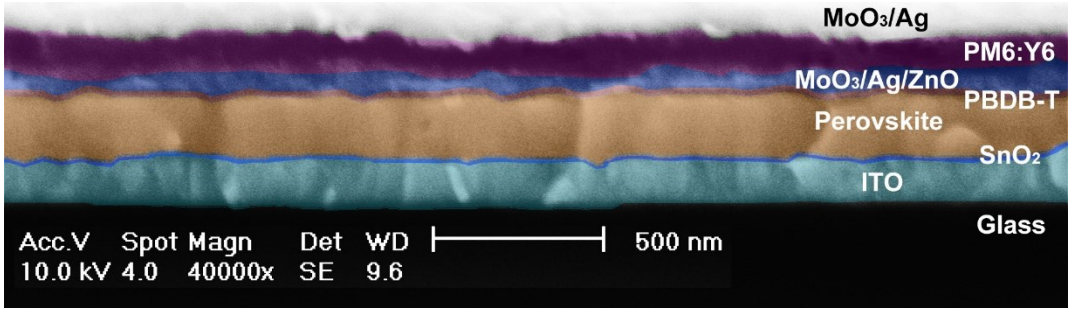

Figure S10. Cross-section SEM image of fabricated TSC.

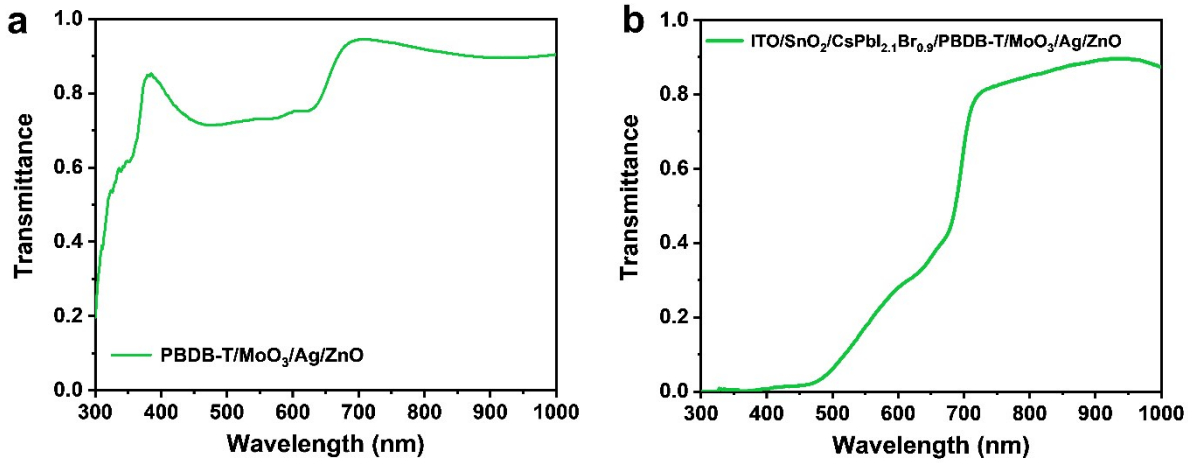

Figure S11. transmittance spectra of (a) PBDB-T/MoO_3/Ag/ZnO, and (b) ITO/SnO_2/$CsPbI_{2.1}Br_{0.9}$/PBDB-T/MoO_3/Ag/ZnO.

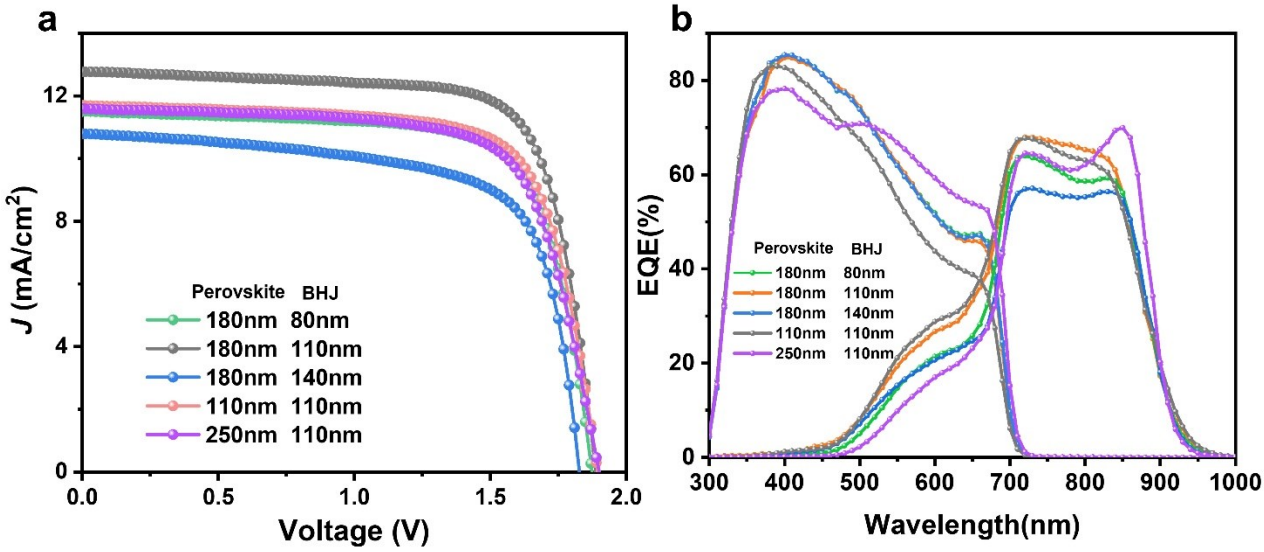

Figure S12. The *J-V* curves and EQE spectra of tandem devices with perovskite and BHJ layers with different thickness.

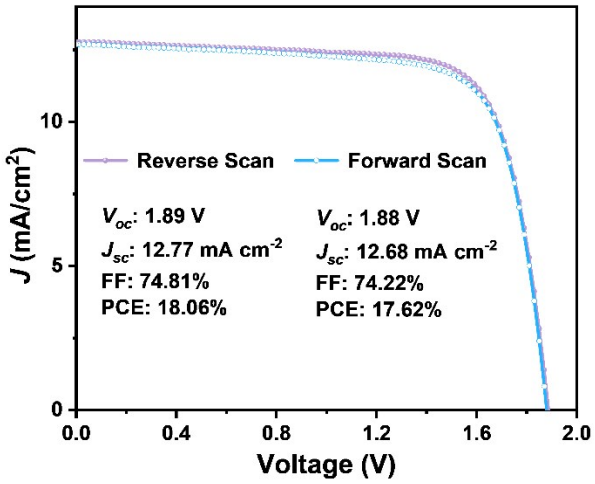

Figure S13. The J-V curves of champion tandem device measured at reverse and forward scans under AM 1.5G illumination.

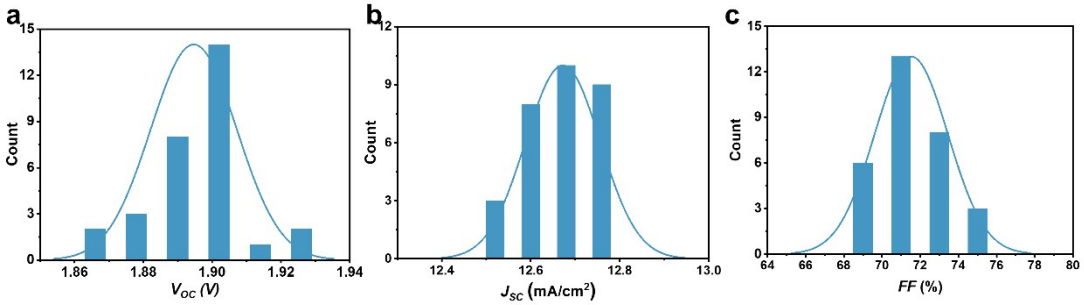

Figure S14. The histograms of (a) V_{OC}, (b) J_{SC} and (c) *FF* of 2T tandem cells extracted from 30 devices.

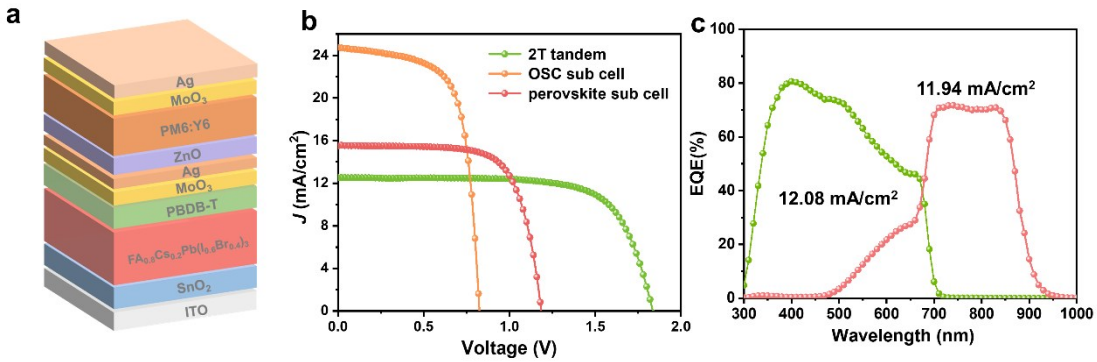

Figure S15. (a) Schematic structure of the tandem device based on $FA_{0.8}Cs_{0.2}Pb(I_{0.6}Br_{0.4})_3$ perovskite; (b) The *J*–*V* curves of perovskite top cell, organic bottom cell, and 2T tandem solar cell under AM 1.5G illumination; (c) EQE spectra of perovskite top cell and organic bottom cell, operating in the 2T tandem solar cell.

Table S1. The photovoltaics parameters of the all-inorganic perovskite solar cells with different concentration of PMACl for passivation under simulated AM 1.5G illumination.

	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
REF	1.07	15.40	80.52	13.27
1.0 mg/ml	1.12	15.42	80.98	13.99
2.5 mg/ml	1.15	15.46	81.19	14.43
5.0 mg/ml	1.13	15.18	79.58	13.65

Table S2. The fitted parameters of time-resolved PL data of films on $CsPbI_{2.1}Br_{0.9}$ and $CsPbI_{2.1}Br_{0.9}$/PMACl on ITO/SnO_2 substrates.

	τ_1 (ns)	A_1	% of τ_1	τ_2 (ns)	A_2	% of τ_2	τ_{avg}
$CsPbI_{2.1}Br_{0.9}$	4.57	464.92	18.64	8.56	1081.90	81.36	7.81
$CsPbI_{2.1}Br_{0.9}$/PMACl	5.10	191.78	8.89	17.21	582.89	91.12	16.13

Table S3. Summary of device peformance of tandem solar cells with different thickness of active layers.

				J_{int} (mA/cm^2)		active layer thickness (nm)	
Voc(V)	Jsc(mA/cm^2)	FF(%)	PCE(%)	Top cell	Bottom cell	$CsPbI_{2.1}Br_{0.9}$	BHJ
1.87	11.50	72.98	15.69	12.43	10.83	180	80
1.89	12.77	74.81	18.06	12.32	12.12	180	110
1.83	10.80	68.73	13.58	12.39	10.24	180	140
1.89	11.70	72.72	16.08	11.06	12.20	110	110
1.90	11.59	71.04	15.64	12.85	10.86	250	110

Table S4. Energy loss parameters for sub cells and tandem cell.

System	E_{trans}	E_{abs} + E_{IQE}	$E_{thermal}$		E_{Voc}		E_{FF}		$E_{conv.}$
	[mW/cm^2]	[mW/cm^2]	[mW/cm^2]	[eV][a)]	[mW/cm^2]	[eV][b)]	[mW/cm^2]	[eV][c)]	[mW/cm^2]
PVSC	53.34	12.31	8.47	0.57	9.22	0.62	3.13	0.21	13.53
OSC	30.44	21.89	15.93	0.68	12.23	0.52	6.21	0.26	13.30
Tandem	30.44	18.71	12.46	0.51	14.16	0.58	6.10	0.25	18.13

a) Averaged thermalization loss per electron; b) Averaged V_{OC} loss per electron; c) Averaged FF loss per electron.

Table S5. The photovoltaic parameters of $FA_{0.8}Cs_{0.2}Pb(I_{0.6}Br_{0.4})_3$-based perovskite top cell, OSC bottom cell, and 2T tandem solar cell under simulated AM 1.5G illumination.

	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)
PVSK	1.19	15.50	71.20	13.13
OSC	0.83	24.75	68.18	14.01
Tandem	1.84	12.53	71.72	16.53