Supporting Information

Synergy of transmittance fluctuation factor and absorption selectivity for efficient semitransparent perovskite/organic tandem solar cells with high color-fidelity

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Device	Active	TEE	AVT	$T_{\rm max}$	CPI	CIE	PCE	LUE	Pof
type	layer	IFFVIS	(%)	(%)	UKI	(x, y)	(%)	(%)	I\C1
SJ-PSCs	Island-FAPbI ₃	10.32	28.10	34.61	-	(0.349,0.358)	5.20	1.46	[25]
	CH ₃ NH ₃ PbI ₃₋ _x (SCN) _x	4.67	50.00	53.72	-	(0.325,0.342)	2.40	1.20	[26]
	$\frac{Cs_{0.05}FA_{0.83}MA_{0.}}{{}_{12}PbBr_{0.33}I_{0.27}}$	-	32.50	-	-	(0.366,0.344)	10.53	3.42	[27]
	$\begin{array}{c}(FA_{0.65}MA_{0.2}Cs_{0.}\\ \\ {}_{15})Pb(I_{0.8}Br_{0.2})_3\end{array}$	13.90	21.74	78.50	-	(0.403,0.359)	9.30	2.02	[28]
SJ-OSCs	PTB7:PCBM	5.96	39.98	65.18	-	(0.311,0.336)	3.35	1.34	[29]
	PffBT4T- 2OD:PC ₇₁ BM	8.35	16.30	48.77	90.7	(0.314,0.356)	6.60	1.08	[17]
	PCDTBT/PC ₇₁ B M/ITIC	4.19	39.20	43.40	97.0	(0.312,0.344)	4.02	1.58	[30]
	J71:Y6:PC71BM	5.27	36.00	53.57	90.0	(0.329,0.343)	7.00	2.52	[31]
	PM6-Ir1:BTP- eC9:PC ₇₁ BM	3.17	20.44	29.06	96.5	(0.333,0.341)	14.09	2.88	[32]
	PCE-10:BTCIC	13.26	49.00	54.21	89.0	(0.330,0.360)	8.30	3.04	[33]
	PCE-10:BTCIC	11.17	43.30	48.96	86.0	(0.380,0.390)	8.10	3.64	[34]
P/O- TSCs	CsPbI ₂ Br/PTB7- Th:IEICO-4F	2.99	26.28	30.60	96.4	(0.344,0.349)	13.56	3.56	This work

Table S1. A comprehensive investigation on performances of state-of-the-art SJ-PSCs, SJ-OSCs and our semitransparent P/O-TSCs.

Table S2. The careful investigation on performances of state-of-the-art ST-TSCs and oursemitransparent P/O-TSCs.

Front-cell	$E_{\rm g}$, Thickness	Rear-cell	$E_{\rm g}$, Thickness	PCE (%)	AVT (%)	CRI	CIE (x,y)	LUE (%)	Ref.
FAPbBr _{2.43} Cl 0.57	2.36 eV, 350 nm	PTB7-Th:6TI C-4F	1.27 eV, 100 nm	10.7	52.91	68.0	(0.45,0.42)	5.66	[39]
PIDT-phanQ:P C ₆₁ BM	1.59 eV, 70 nm	PIDT-phanQ: PC ₇₁ BM	1.55 eV, 85 nm	6.7	40	97.2	(0.33,0.34)	2.68	[40]
PSEHTT:IC ₆₀ BA	- 80 nm	PBDTT-DPP: PC ₇₁ BM	- 100 nm	4.9	24	96.2	(0.34,0.35)	1.18	[41]
PBDT[2F]T: PC ₇₁ BM	1.00 eV, 100 nm	PCE10:PC ₆₁ B M	1.00 eV, 70 nm	5.3	49.20	-	(0.39,0.32)	2.61	[42]
P3TEA:FTT B-PDI4	1.88 eV,	PTB7-Th:IEI CS-4F	1.00 eV,	10.5	20	-	(0.29,0.36)	2.10	[43]
PBDTT-FDP P-C12:PC ₆₁ BM	1.30 eV,	PBDTT- SeDPP:PC ₇₁ B M	1.30 eV,	7.3	30	-	-	2.19	[44]
CsPbI ₂ Br	1.92 eV, 200 nm	PTB7-Th:IEI CO-4F	1.18 eV, 90 nm	13.56	26.28	96.4	(0.34,0.35)	3.56	This work

Experimental Section

Materials:

Indium Tin oxide (ITO) glass was purchased from CSG Holding Co., Ltd. (Shenzhen, China). The thickness of ITO film is 110 nm and the square resistance is $12 \Omega \text{ sq}^{-1}$. CsI, PbI₂ and PbBr₂ are purchased from Xi'an Paulette Optoelectronics Co., Ltd. PTB7-Th, IEICO-4F, PFN-Br and poly3-hexylthiophene (P3HT) were purchased from Solarmer Material Inc. (Beijing, China). Tin oxide (IV) (15% in H₂O colloid dispersing solution), dimethyl sulfone (DMSO, 99.7%+), ultra-dry methanol, isopropyl alcohol (IPA), chloroform (CF), ultra-dry chlorobenzene (CB), 1-chloralin (CN) and molybdenum trioxide (MoO₃) were purchased from Alfa Aesar (Shanghai, China). All of the above materials are commercial products and can be used directly without further processing.

Device fabrication:

Solution preparation: Tin oxide (15% H₂O colloidal dispersion) was diluted with distilled water (dilution ratio 1:5), and stirred for 30 min to fully disperse SnO₂ precursor solution. Then, CsI (1.2 M), PbI₂ (0.6 M) and PbBr₂ (0.6 M) were dissolved in DMSO, and heated and stirred at 60 °C for 2 h to obtain the CsPbI₂Br precursor solution. PTB7-Th (8, 9 and 10 mg/mL) was dissolved in ultra-dry chlorobenzene and stirred for 6 h to obtain PTB7-Th solution. P3HT (10 mg/mL) was dissolved in ultra-dry chlorobenzene and stirred for 5 h to obtain P3HT solution. After that, PFN-Br (0.5 mg/mL) was dissolved in methanol and stirred for 6 h to obtain PFN-Br solution. PTB7-Th:IEICO-4F (1:2 w/w, 24 mg/mL) was dissolved in chloroform and stirred for 3 h to obtain polymer active layer solution, and 1-CN was added 15 min before use, the volume ratio of chloroform:1-CN was 99.5:0.5.

Device fabrication: First, the ITO glass substrate was washed twice in the ultrasonic bath containing detergent, tap water, ultra-pure water and isopropyl alcohol, respectively, for 15min each time. The cleaned ITO substrate was blown dry with nitrogen gun and dried in oven (150 °C) for 5 min, and then treated with ultraviolet ozone (UVO) for 15 min. The SnO₂ precursor solution was spin-coated on ITO substrate at 2500 rpm, and then annealed at 150 °C for 30 min to obtain the SnO₂ electron transport layer (ETL). The substrate was transferred to a nitrogen-atmosphere glove box, and the CsPbI₂Br precursor solution was spin-coated on SnO₂ layer in a multi-step spin coating mode (500 rpm 5s+3000 rpm 30s), and heat treated (42 °C 1.2 min+160 °C 10 min). Polycrystalline CsPbI₂Br films with a thickness of about 350 nm were obtained. Then, the PTB7-Th solution was spin-coated on the perovskite layer at 3850 rpm for 30 s and annealed at 100 °C for 10 min. For SJ-PSCs, MoO₃ (10 nm)/Ag (100 nm) was sequentially deposited on it in a high vacuum thermal evaporator under 1×10^{-5} Pa. For tandem devices,

MoO₃ (8 nm)/Ag (1 nm) was deposited by thermal evaporation, and then PFN-Br was spincoated at 3000 rpm for 30 s. After that, PTB7-Th:IEICO-4F solution was spin-coated on PFN-Br at 3000 rpm for 60 s to obtain organic active layer (about 200 nm). For semitransparent tandem devices, the CsPbI₂Br precursor solution was spin-coated on SnO₂ with a multi-step spin coating mode (500 rpm 5s+5500 rpm 30s), and the CsPbI₂Br film (thickness of 200 nm) was obtained after heat treatment (42 °C 1.0 min+160 °C 10 min). Next, the PTB7-Th solution was spin-coated on perovskite layer at 4500 rpm for 30 s and annealed at 100 °C for 10 min. Then MoO₃(6 nm)/Ag (1 nm) was evaporated on the PTB7-Th layer, and PFN-Br solution was spin-coated on Ag at 3000 rpm for 30 s. Then, PTB7-Th:IEICO-4F solution was spin-coated on PFN-Br at 5000 rpm for 60 s, resulting in an organic active layer with a thickness of about 90 nm. Finally, MoO₃(10 nm)/Ag (100 nm) or MoO₃(10 nm)/Ag(15 nm) were subsequently evaporated by vacuum thermal evaporation for opaque and semitransparent devices under the pressure of 1×10⁻⁵ Pa. The device area is about 4 mm².

Material and device characterization and measurement

SEM characterization: The planar-view of ITO/SnO₂/CsPbI₂Br and ITO/SnO₂/CsPbI₂Br/PTB7-Th and the cross-sectional-view of TSCs were scanned by HITACHI Model SU8600 field emission scanning electron microscope (FE-SEM).

Transmittance and Absorption characterization: The absorption and transmittance spectra of the UV-Vis spectrum were measured using the UV-Vis NIR3600 spectrometer in Shimadzu, Japan.

AFM characterization: The surface morphology of the films was tested by Agilent 5500 atomic force microscope (AFM).

Characterization of photovoltaic performance of solar cells:

The current density voltage (*J-V*) characteristic curve was developed using a solar simulator (SAN-EI LTD., Class AAA grade) with a computer-controlled Keithley 2400 source measuring device under one-sun AM 1.5G (100 mW cm⁻², 25 °C) standard test conditions in a nitrogen-filled glove box. EQEs under ambient atmosphere was measured by QE-R system of Enli Technology at room temperature. In addition, the EQE spectra of front and rear sub-cell were characterized by adding an additional biased monochromatic light source (the emission peaks were L750 nm and S550 nm, respectively). The light intensity at each wavelength was calibrated using standard monocrystalline silicon solar cells purchased from the National Renewable Energy Laboratory (NREL). In addition, all the PV parameters obtained in this work were averaged over 12 individual devices.

Parameters	description	Units	CsPbI ₂ Br	MAPbI ₃	PM6:Y6	PTB7-Th:IEICO-4F
χ	affinity	eV	3.97	3.95	4.10	4.20
$E_{ m g}$	energy band gap	eV	1.92	1.55	1.36	1.18
$\mu_{ m n}$	electron mobility	cm ² (V•s) ⁻¹	16	22	2.6×10-4	2.5×10 ⁻⁴
$\mu_{ m p}$	hole mobility	cm ² (V•s) ⁻¹	16	22	2.6×10 ⁻⁴	2.5×10 ⁻⁴
$N_{ m c}$	effective DOS in LUMO	cm ⁻³	1.1×10 ¹⁸	1.5×10 ¹⁸	2.5×10 ²¹	7.5×10 ¹⁹
$N_{ m v}$	effective DOS in HOMO	cm ⁻³	1.1×10 ¹⁸	8.0×10 ¹⁹	2.5×10 ²¹	7.5×10 ¹⁹
а	exciton binding distance	nm	_	_	1.321	1.312
E_{b}	binding energy	eV	_	_	0.14	0.16
N_{TC}	total DOS in exponential LUMO	cm ⁻³ eV ⁻¹	_	_	2.0×10 ²¹	2×10^{21}
N_{TV}	total DOS in exponential LUMO	cm ⁻³ eV ⁻¹	_	_	2.0×10 ²¹	2×10^{21}
\mathcal{E}_{r}	relative permittivity of active layer	_	_	_	3.8	3.5

Table S3. Basic device parameters of SJ-PSCs, SJ-OSCs and TSCs utilized in the simulation.

Parameters	Description	Units	Value		
$\mathcal{E}_{ m vac}$	vacuum permittivity	F/m	8.854×10 ⁻¹²		
\mathcal{E}_{r}	relative permittivity of	-	3.5		
	active layer		(for PTB7-Th:IEICO-4F)		
k_B	Boltzmann constant	J/K	1.38×10 ⁻²³		
h	Planck's constant	$J \cdot s$	6.626×10 ⁻³⁴		
С	speed of light	m/s	2.998×10 ⁸		
q	electric charge	С	1.602×10 ⁻¹⁹		
Т	temperature	Κ	300		
t _{ITO}	thickness of ITO	nm	110		
t _{SnO2}	thickness of SnO ₂	nm	20		
ta arras	thickness of CsPhI-Br	nm	350 (for Opaque)		
<i>v</i> CsPbI2Br	$Unexhess \ Or \ CSr \ Or_2Dr$	11111	200 (for Semitransparent)		
to mo - mi	thickness of PTR7-Th	nm	42 (for Opaque)		
ℓPTB7-Th		11111	30 (for Semitransparent)		
$t_{\rm MoO3}$ of ICL	MoO ₃ thickness of ICL	nm	6		
$t_{\rm Ag}$ of ICL	Ag thickness of ICL	nm	1		
<i>t</i> _{PFN-Br}	thickness of PFN-Br	nm	10		
t _{PTB7-Th:IEICO-}	thickness of PTB7-		200 (for Opaque)		
4F	Th:IEICO-4F	nm	90 (for Semitransparent)		

Table S4. The relevant simulation parameters of optical transfer matrix simulation.



Figure S1. (a) Solar flux (AM 1.5G) and normalized absorption spectra of CsPbI₂Br, MAPbI₃, PM6, PTB7-Th, Y6 and IEICO-4F. (b) Theoretical and experimental EQE curves of singlejunction MAPbI₃-based PSCs. (c) Theoretical EQE curves of front and rear sub-cells based on MAPbI₃/PTB7-Th:IEICO-4F TSCs. (d) Theoretical summed EQE as well as derived shortcircuit current density of MAPbI₃/PTB7-Th:IEICO-4F-based TSCs.



Figure S2. (a) The hole mobility diagram of PTB7-Th and P3HT, (b) the energy levels of materials involved in CsPbI₂Br-based SJ-PSCs, (c) experimental J-V curves and (d) experimental EQE of SJ-OSCs under the illumination of AM 1.5G with light intensity of 100 mW cm⁻².

Table S5. Photovoltaic parameters for PTB7-Th:IEICO-4F-based SJ-OSCs under the illumination of AM 1.5G with the light intensity of 100 mW cm⁻².

Device type	V _{oc} (V)	$J_{ m sc}$ (mA cm ⁻²)	FF (%)	PCE _{avg} (%)	J _{inte} (mA cm ⁻²)
Opaque SJ-OSCs	0.72	23.52	64.22	10.87	23.24
Semitransparent SJ-OSCs	0.71	17.60	63.16	7.89	17.28



Figure S3. Comparison of photovoltaic performance between opaque and semitransparent wide-bandgap CsPbI₂Br-based PSCs (aperture area, 0.04 cm^2) processed in the AM 1.5G, 100 mW cm⁻² (12 devices for each type). The center indicates the mean value, while the box lines show the standard deviation.



Figure S4. Comparison of photovoltaic performance between opaque and semitransparent narrow-bandgap PTB7-Th:IEICO-4F-based OSCs (aperture area, 0.04 cm²) processed in the AM 1.5G, 100 mW cm⁻² (12 devices for each type). The center indicates the mean value, while the box lines show the standard deviation.



Figure S5. Comparison of photovoltaic performance between opaque and semitransparent $CsPbI_2Br/PTB7$ -Th:IEICO-4F-based TSCs (aperture area, 0.04 cm²) processed in the AM 1.5G, 100 mW cm⁻² (12 devices for each type). The center indicates the mean value, while the box lines show the standard deviation.