Supplementary Information

Interfacial Engineering and Optical Coupling for Multicolored Inverted Semitransparent Organic Photovoltaics with Record Efficiency Over 12%

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X-ray photoelectron spectroscopy (XPS) was investigated to characterize the chemical composition and electronic states of SnO₂, as shown in Figure S1. The SnO₂ films were prepared by SnO₂ colloid precursor annealing at 150 °C for 30 min in ambient air. The binding energy (BE) is corrected for specimen charge by referencing the C 1s peak at 284.80 eV (Fig. S1e). The XPS spectrum survey ranged from 0 to 1350 eV is given in Figure S1a, and the peaks from Sn 4d, Sn 3d5/2, Sn 3d3/2, Sn 3p3/2, Sn 2p1/2, O 1s, O Auger, Sn Auger confirms the presence of presence of O and Sn. The XPS spectrum of the core level Sn 3d in Figure S1b has a strong spin-orbit doublet, the peaks located at 486.46 eV and 494.87 eV correspond to Sn 3d_{5/2} and Sn $3d_{3/2}$, respectively. It is apparent that the Sn 3d peaks revealed just one symmetrical component without any shoulder, which indicates only a Sn⁴⁺ chemical state in SnO₂ film. ¹ The asymmetric O 1s X-ray photoelectron spectrum can be fitted out two peaks, as shown in Figure S1c, the main binding energy (530.60 eV) is attributed to the O^{2-} state in SnO₂ while the higher binding energy (531.80 eV) correspond to the chemisorbed oxygen atoms or hydroxyl groups, indicating the composition of SnO₂, which shows reasonably good agreement with previous studies. ²⁻⁴ The binding energy peaks at 292.67 and 295.46 eV in Figure S1d can be assigned to K 3p_{3/2}, K 2p_{1/2}, which confirm the presence of potassium ions with the purpose of interface passivation.⁵



Figure S1 XPS spectra of a) survey, b) Sn 3d, (c) O 1s, d) K 2p and e) C 1s peaks for SnO₂ film coated on the ITO substrate.

Figure S2a-c displays the UPS survey scan of SnO_2 layer to verify its energy level. The work function (WF) of SnO_2 layer is 4.89 eV obtained from the difference between the incident light energy (He I, 21.22 eV) and the energy of the cutoff (16.33 eV), ⁶ and the valence band (VB) maximum is 7.54 eV calculated from the WF and the onset (4.89+2.65=7.54 eV), predicting an outstanding hole blocking ability ^[7]. The band gap is 3.35 eV by the onset of the

absorption spectra in **Figure S2a**. Therefore, the conduction band (CB) is 4.12 eV (7.54-3.42=4.12 eV).



Figure S2. UPS spectra of SnO₂ film.



Figure S3. (a) Refractive index n and (b) extinction coefficient k of layers involved in the devices.

Due to the variety of colors, a set of standard colors need to be selected so that they can fully represent commonly used colors. In the CIE color system, 8 typical colors with multiple tones, medium brightness values and chroma (TSC01-08) are adopted to get CRI value via comparing the spectra of different light sources with the spectrum of the specified color sample. ^[8] The specific calculation process is as follows. Firstly, the color displacement ΔE_i in the ultraviolet visible color system can be obtained by measuring color coordinate differences of samples under the illumination of test source and the reference light source. Then the special color rendering index R_i of samples is calculated according to the equation of R_i =100-4.6 ΔE_i . Lastly, the average CRI is obtained by taking an arithmetic mean of R_i measured by eight standard swatches. The chromaticity by test source after von Kris chromatic adaption can be seen in **Table S1 and S2**, which were measured by the transmitted light of PBDB-T-2F:ITIC-4F- and PBDB-T-2F:Y6-based ST-OSCs under AM1.5G illumination.

Table S1. Color rendering indices (CRIs) of the TCS01-08 illuminated by the transmitted light of the PBDB-T-2F:Y6-based ST-OSCs under AM1.5G illumination (average CRI=97.6).

	u′k,i	v′k,i	Y k,i	₩ [_] k,i	U k,i	V [⊥] k,i	ΔEi		
TSC01	0.2109	0.2861	28.16	59.06	24.52	10.66	0.48	R_1	97.8
TSC02	0.1922	0.3184	27.62	58.57	10.12	35.16	0.41	R_2	98.1
TSC03	0.1675	0.3426	29.59	60.33	-8.96	55.22	0.17	R_3	99.2
TSC04	0.1407	0.3100	30.69	61.27	-30.41	30.07	0.34	R_4	98.4
TSC05	0.1540	0.2643	32.50	62.78	-20.37	-6.49	0.42	R_5	98.0
TSC06	0.1648	0.2293	31.97	62.35	-11.43	-34.78	0.66	R ₆	96.9
TSC07	0.1918	0.2276	29.81	60.52	10.14	-35.15	0.56	R_7	97.4
TSC08	0.2075	0.2473	30.78	61.35	22.83	-22.74	1.07	R_8	95.1

Table S2. Color rendering indices (CRIs) of the TCS01-08 illuminated by the transmitted light of the PBDB-T-2F:ITIC-4F-based ST-OSCs under AM1.5G illumination (average CRI=97.5).

	u'k,i	v′k,i	Y k,i	₩ [_] k,i	U k,i	V⁻k,i	∆Ei		
TSC01	0.2163	0.2956	28.40	59.28	26.07	10.18	0.65	R_1	97.0
TSC02	0.1973	0.3249	27.85	58.78	11.40	32.46	0.58	R_2	97.3
TSC03	0.1721	0.3467	29.79	60.50	-8.12	50.53	0.53	R_3	97.6
TSC04	0.1439	0.3168	30.59	61.19	-30.66	27.33	0.43	R_4	98.0
TSC05	0.1558	0.2744	32.26	62.59	-21.63	-6.15	0.62	R_5	97.2
TSC06	0.1660	0.2408	31.58	62.02	-13.22	-33.56	0.88	R_6	95.9
TSC07	0.1947	0.2398	29.66	60.38	9.64	-33.44	0.39	R_7	98.2
TSC08	0.2117	0.2556	30.77	61.34	23.33	-21.41	0.33	R_8	98.5

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