Supplementary Information

for

Solution-processed ITO Nanoparticles as Holeselective Electrode for Mesoscopic Lead-free Perovskite Solar Cells

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Experiments

Formulation of ITO ink: ITO nanoparticles (6 g, US Research Nanomaterials Inc.) were dispersed in the presence of acetic $acid^{[s1]}$ (1 mL) and dried ethanol (10 g) with a ball mill for 1 h. This step helps to avoid formation of micrometre-sized aggregates, a major source for the formation of cracks in eventual films, as well as greatly enhancing dispersity. Poly(ethandiol) (0.3 g, Sigma), terpineol (20 g, Alfa Aesar), ethyl cellulose solution (10 mass %, 30 g) and dried ethanol (10 g) were added to the well dispersed ITO. The same ball-mill process but with duration 6 h and rotary evaporation at ~38 °C for ~10 h were sequentially performed. Before use, it was 3-roll-milled three times to improve the paste quality and hence the printability.

Preparation of Perovskite Solution: FAI (Greatcell Solar), GAI (Greatcell Solar), SnI₂ (Alfa Aesar), SnF₂ (Aldrich), DMSO (Aldrich), DMF (Aldrich), EDAI₂ (Greatcell Solar) were used without further purification. The perovskite precursor solutions were prepared on dissolving SnI₂ (1 M), $GA_{0.2}FA_{0.8}$ (1 M), SnF₂ (0.2 M) and EDAI₂ (x M, x= 0, 0.05, 0.1, 0.15 and 0.2) in a DMF/DMSO solution with a blend ratio 50 % by volume. For the better pore-filled ITO devices, the solution containing EDAI₂ (15 %) was concentrated 1.15-fold straightforwardly on decreasing the volume of the blended solution.

Device fabrication: A compact layer of TiO₂ (thickness: ≈ 50 nm) was thermally sprayed onto etchcleaned FTO glasses, followed by annealing at 500 °C for 30 min. Using screen printing, mesoporous TiO₂ (thickness 1 µm), Al₂O₃ (thickness 1 µm) and ITO (thickness 2.1-14.3 µm) or carbon (thickness 1 µm) layers, for which multiple printings of ITO or carbon were performed for target thicknesses, were stacked in sequence on top of the TiO₂ compact layer. Intermittently, we added for ITO and carbon ambient temperature waiting for 6 min, ensuring excellent contact with the underlying Al₂O₃, with annealing at 120 °C for 6 min and 200 °C for 6 min to evaporate part of the terpineol. The TiO₂ was annealed at 500 °C for 10 min; the other samples (Al₂O₃/ITO and /carbon) were annealed at 400 °C for 30 min. Using the resulting electrodes, we fabricated ITO or carbon devices on infiltrating hot perovskite solution into the TiO₂/Al₂O₃ through capillarity and then crystallizing the perovskite. The environment of the device fabrication was strictly controlled to create high-performance tin perovskites in a glove box in which water (≤ 0.1 ppm) and dioxygen $(\leq 0.1 \text{ ppm})$ were minimal) at temperature ~22 °C. For perovskite deposition, we applied two annealing conditions, 70 °C for 3-5 min and 100 °C for 15 min. The former was required for thorough infiltration of the perovskite solution; the latter was required to form excellent perovskite crystals. During the infiltration, the electrodes were capped with glass dishes. The volume of perovskite solution was set to 2 μ L, which suffices to cover completely an active device of area 0.4 cm². Following crystallization, we applied a silver pen at both sides of the electrodes to decrease the contact resistance. For the MAPbI₃ device, a two step deposition method consisting of PbI₂ infiltration (at 70 °C for 10 min), annealing (at 70 °C for 30 min), MAI dipping (at ~25 °C for 20 min) and annealing (at 70 °C for 30 min) was applied; 1 M PbI₂ in DMSO/DMF/NMP (3:3:4, v/v/v) and 62.9 mM MAI in IPA were used; 1 layer of carbon was deposited onto 4 layers of ITO to enhance infiltration of PbI2. Meanwhile, the Cu:NiO (WF: ~5.25 eV close to the perovskite) in the hole-only device was synthesized and formulated into a screen printable ink according to literatures.^[s2,s3]

Characterization of film and devices: *J-V* characteristic curves were measured with a solar simulator (XES-40S1, SAN-EI), under standard illumination conditions (AM 1.5G, 100 mW cm⁻²), which we calibrated with a standard Si reference cell (Oriel, PN 91150V, VLSI standards), and used a reverse scan at rate 0.07 V s⁻¹ and a metal mask of area 0.09 cm². The IPCE spectra were recorded following calibration with a standard Si photodiode (S1337-1012BQ, Hamamatsu) and

with a Xe lamp (A-1010, PTi, 150 W), monochromator (PTi, 1200 g mm⁻¹ blazed at 500 nm) and source meter (Keithley 2400). The XRD patterns were acquired with a commercial diffractometer (Bruker D8-Advance, Cu K_a radiation). A field-emission SEM (Hitachi SU8010) generated highresolution images of top and cross-sectional views. The light reflectance and transmittance spectra were recorded with a UV-vis/NIR spectrophotometer integrated with a spherical accessory (V-570, Jasco). For measurements of PL, we used a home-built system based on a combination of an amplified detector, AC-modulated pump source and programmable high-resolution monochromator, with excitation at 450 nm. X-rray and ultraviolet photoelectron (UPS) spectra were recorded (Thermo K-ALPHA Surface Analysis). For the UPS the photon energy was 21.22 eV, with bias -5.0 V and Au as reference. The EIS was recorded under 0.5-sun LED light and at 1 MHz – 1 Hz with AC amplitude 10 mV. Each open-circuit voltage of a device as DC voltage was applied. The device thickness was evaluated from the cross-sectional SEM images in Figure S6. The photodetection spectra were acquired using the standard Si cell.

Simulation of Mie scattering: According to the theory, to simulate Mie scattering refractive indices (n) of a particle and a medium, absorption coefficient (k) of particle, particle diameter, and wavelength of incident light must be specified. Absorption coefficient ~0.0028 was derived for an ITO nanoparticular film (thickness 2.1 µm). We assumed the ITO nanoparticles to have a spherical shape and a particle diameter ranging between 20 and 160 nm, as observed in Figure 1a. The effective refractive index of the porous ITO was approximated with the Lorentz-Lorenz relation,^[s4] $f(n_{porous ITO}) = p \times f(n_{air}) + (1-p) \times f(n_{flat ITO})$, in which $f(n_x) = (n_x^2-1)/(n_x^2+2)$, *p* is a porosity of the porous ITO film, n_{air} and $n_{flat ITO}$ are the refractive indices of air and a flat ITO film,^[s5] respectively. The porosity was calculated to be 0.70 on subtracting a volume fraction of porous ITO at a given volume (>0.015 cm³), for which a sufficient amount (>0.03 g) of the ITO was collected; ITO

density 7.14 g/cm³ was used. The resulting $n_{\text{porous ITO}}$ was 1.207, which is less than reported values (~1.5) of a ITO film comprising nanoparticles of similar size.^[s6] Whereas the reported ITO film might be dense from being readily deposited from a commercial ITO dispersion, our ITO is porous, because of, in large part, the added ethyl cellulose that creates mesopores during decomposition under annealing. Using the obtained parameters and a computer program (MiePlot v4.6.14), we simulated the back-scattering efficiency as a function of wavelength on varying the size of the ITO nanoparticles. The results appear in Figure S2.

Dark J-V analysis: Using the diode equation,^[s7] we calculated a characteristic parameter of saturation current (J_0). Although a small J_0 is desired for a large V_{OC} , the calculated J_0 , 3.85 × 10⁻⁶ mA cm⁻², for ITO was about one ten-thousandth that (4.84 × 10⁻² mA cm⁻²) of carbon. The series resistance (R_s) was obtained near V_{OC} from the inverse slope of the dark *J-V* curve. The resulting R_s values, 60.0 Ω cm² (carbon) and 52.2 Ω cm² (ITO), indicate that the ITO has a much smaller resistance, interpreted as a smaller barrier for hole injection.

EIS analysis. The EIS results represented with Nyquist plots were fitted with an equivalent-circuit model shown in Figure S8 and Table S8. The Nyquist plots feature three distinct arcs in relation to charge transfer (high frequency), charge recombination (intermediate frequency) and dielectric relaxation (low frequency);^[s8,s9] these are restricted to the perovskite/ITO (or carbon) interface because the other interfaces of the perovskite/TiO₂ and /Al₂O₃ are the same. The high-frequency arc was scarcely detectable in the ITO representing a minimal barrier for charge injection whereas several ohms were tracked for the carbon, to support the dark *J*-*V* result. The next two arcs emerged larger for the ITO with 3.4 and 1.5 times greater resistances of charge recombination and dielectric relaxation. Although the greater resistance of charge recombination represents a small recombination as already monitored in the dark *J*-*V*, the greater resistance of the dielectric

relaxation reflects a favorable ITO interface that would minimize charge accumulation for an efficient charge separation.^[s8,s9]



Figure S1. (a) UPS and (b,c) high-resolution XPS of an ITO nanoparticular electrode for which signature properties were confirmed: in (a) work function measured to be 4.7 eV; in (b) entire ITO elements appear; in (c) three surface properties, O-metal, V_0 (oxygen near oxygen vacancies) and -OH (either In(OH)₃ or InOOH) emerge.^[s10] These surface states can be physically or chemically tailored toward altering its WF in a range of 3.3-6.1 eV to be compatible with other applications.^[s11,s12] (d) A high-resolution SEM image of ITO nanoparticles.



Figure S2. Simulated back scattering (ie. reflection) efficiency of ITO nanoparticles ranging in diameter (a) from 20 to 160 nm (10-nm increase in the plot) corresponding to the range in Figure 1b (b) from 100 to 400 nm (50-nm increase in the plot) and showing light reflection systematically enhanced with increasing diameter. In particular in (b), near-infrared light reflectance becomes pronounced for the particles ≥ 200 nm diameter.



Figure S3. Representative current density-voltage curves of carbon devices with varied amounts of EDAI₂; 15 % yielded the greatest PCE. The corresponding photovoltaic parameters are summarized in Table S2.



Figure S4. Evidence of emergence of hollow vacancies upon adding EDAI₂ of varied proportion (0-20%) into tin-based perovskites ($GA_{0.2}FA_{0.8}SnI_3$). (a) Light absorption and photoluminescence spectra showing systematic blue shifts with increasing EDAI₂ proportion. From the absorption onset, band gaps were calculated to be 1.39 (0 %), 1.46 (5 %), 1.54 (10 %), 1.61 (15 %) and 1.65 (20 %) eV. (b) XRD patterns of the perovskites; the substrate represents the ITO device without perovskites infiltrated. All perovskites show signature signals that are systematically shifted to low angles with increasing EDAI₂.



Figure S5. (a,b) High-resolution XPS of perovskite films with (a) 0 % and (b) 15 % EDAI₂, in which two Sn²⁺ and Sn⁴⁺ (defect) components were deconvoluted. The proportions of Sn²⁺ and Sn⁴⁺ were 0.612:0.388 for 0 % and 0.927:0.073 for 15 %, which implies a significant decrease of Sn⁴⁺ defects following incorporation of EDAI₂ (15 %) into the perovskite. (c,d) An UPS spectrum of the perovskite with 15% EDAI₂. In (c), the work function (4.44 eV) was determined from the spectral onset. In (d), the energy gap (0.76 eV) between the work function (the zero point) and the valence band maximum was determined from the spectral onset. Combined with the band gap (Figure S4a), the energy level of the optimal perovskite is provided in Figure 2d.



Figure S6. (a-e) Cross-sectional SEM images of ITO devices with varied layers, by which we can explicitly confirm the degree of TiO_2/Al_2O_3 filling with perovskite. Meanwhile, residual perovskites deposited into ITO is observed. Particularly in (b), layer boundaries between TiO_2 , Al_2O_3 and ITO are manifested as distinctions in brightness, which is related to conductivity.



Figure S7. (a) Representative current density-voltage curves of ITO devices with increasing concentration of perovskite solution; 1 M was optimal for carbon devices, 1.15 M for ITO devices. The corresponding photovoltaic parameters are summarized in Table S4. (b) Representative hysteresis J-V curves of a ITO device at the optimal condition.



Figure S8. Representative Nyquist plots of ITO and carbon devices; red lines represent fitted lines with an equivalent-circuit model shown in the inset. The fitted parameters are summarized in Table S9.



Figure S9. Time-dependent performance of encapsulated devices. (a) PCE evolution over periods exceeding 1000 h. The devices were stored in a glove box; for intermittent measurements of performance, they were removed and measured under ambient conditions. (b) Output current density measured under the standard one-sun conditions, at maximum power point and under ambient air.

Table S1. Film properties of ITO nanoparticular electrodes; *R*, *T* and *R*_{sh} stand for total reflectance and transmittance, and sheet resistance, respectively. *R* and *T* were obtained from Figure 1e and 1f. *R*_{sh} was obtained using 4-probe measurement and 10 electrodes of each ITO layer. In particular, the 4-8 ITO layers are highly comparable to carbon black (~680 Ω /sq.),^[S13] CNT (~2-5 k Ω /sq.),^[S14] graphene (~550 Ω /sq.),^[S15] materials utilized for efficient perovskite solar cells.

No. of ITO layer(s)	Thickness /μm	R at peaks /%	<i>T</i> at 1150 nm /%	R_{sh} / Ω /sq.
1	2.1	37	93	1600±250
2	4.2	49	84	629±26
4	8.3	59	72	242±12
6	12.2	63	63	153±10
8	16.2	66	53	118±4

EDAI ₂ proportion	V _{oc} /V	J _{SC} /mA cm ⁻²	FF /%	PCE /%
0%	0.194	16.2	32.0	1.0
5%	0.311	20.9	36.9	2.4
10%	0.366	19.1	39.7	2.8
15%	0.373	16.2	49.6	3.0
20%	0.308	15.0	48.2	2.2

Table S2. Representative *J*-*V* parameters of carbon devices with $EDAI_2$ in varied proportions. The corresponding *J*-*V* curves are presented in Figure S3. The optimal condition (15 % $EDAI_2$) was utilized for fabrication of the ITO device.

No. of ITO layer(s)	ITO thickness /μm	V _{oc} /V	J _{SC} /mA cm ⁻²	FF /%	PCE /%
1	2.1	0.479	15.1	42.4	3.1
2	4.2	0.478	15.6	50.6	3.8
4	7.8	0.474	16.7	55.6	4.4
6	11.0	0.439	15.7	50.8	3.5
8	14.3	0.365	13.2	39.8	1.9

Table S3. Representative J-V parameters of ITO devices with ITO films of varied thickness and EDAI₂ at optimal proportion (15 %). The corresponding J-V curves are presented in Figure 3a.

Concentration	V _{oc} /V	J _{SC} /mA cm ⁻²	FF /%	PCE /%
Standard (1 M)	0.483	16.1	56.3	4.4
1.06 M	0.502	16.7	56.3	4.7
Optimal (1.15 M)	0.505	17.2	58.4	5.1
1.23 M	0.488	16.4	57.9	4.6

Table S4. Representative J-V parameters of ITO devices with increasing concentration of the perovskite solution. The corresponding J-V curves are presented in Figure S7.

Device no.	Voc /V	J _{SC} /mA cm ⁻²	FF /%	PCE /%
1	0.501	13.7	55.8	3.8
2	0.499	14.1	53.9	3.8
3	0.493	14.3	57.7	4.1
4	0.448	13.5	48.8	3.0
5	0.485	14.9	55.5	4.0
6	0.504	12.7	56.9	3.6
7	0.456	15.6	56.8	4.0
8	0.474	16.7	55.6	4.4
9	0.499	16.1	58.2	4.7
10	0.484	14.6	59.4	4.2
11	0.475	16.1	54.2	4.1
12	0.462	16.6	52.3	4.0
13	0.469	16.5	52.5	4.1
14	0.454	17.1	50.2	3.9
15	0.478	16.5	54.9	4.3
16	0.510	15.7	55.9	4.5
17	0.487	15.2	56.1	4.2
18	0.510	15.6	58.6	4.7
19	0.480	17.9	57.5	4.9
20	0.479	17.5	57.3	4.8
21	0.483	16.1	56.3	4.4
22	0.444	14.1	50.8	3.2
23	0.429	17.6	52.6	4.0
24	0.458	16.1	49.3	3.6
25	0.453	17.8	49.8	4.0
26	0.447	17.2	55.7	4.3
27	0.446	17.5	54.6	4.3
28	0.480	16.8	57.6	4.6
29	0.476	17.7	55.2	4.7
30	0.453	17.8	49.8	4.0
mean±s.d.	0.474±0.022	16.0±1.5	54.7±3.0	4.1±0.4

Table S5. *J-V* parameters of 30 ITO devices with four ITO layers before optimization. These data are used to form statistical histograms presented in Figures 3c-f.

Device no.	V _{oc} /V	<i>J_{SC}</i> /mA cm ⁻²	FF /%	PCE /%
1	0.469	18.2	56.4	4.8
2	0.482	18.3	56.1	4.9
3	0.482	17.6	56.8	4.8
4	0.503	17.7	57.7	5.2
5	0.502	17.2	53.5	4.6
6	0.486	18.3	54.6	4.9
7	0.522	17.8	52.4	4.9
8	0.495	19.3	54.7	5.2
9	0.492	17.7	56.5	4.9
10	0.483	16.9	56.8	4.6
11	0.485	16.8	56.1	4.6
12	0.485	16.3	55.0	4.4
13	0.482	16.5	57.6	4.6
14	0.499	16.7	53.1	4.4
15	0.502	16.5	54.0	4.5
16	0.482	17.5	52.3	4.4
17	0.482	17.9	52.3	4.5
18	0.464	18.5	54.1	4.6
19	0.499	17.1	53.3	4.6
20	0.531	17.1	58.2	5.3
21	0.505	17.2	58.4	5.1
22	0.499	18.3	51.4	4.7
23	0.488	17.0	55.4	4.6
24	0.480	16.9	54.5	4.4
25	0.515	17.8	58.3	5.4
26	0.481	17.9	56.8	4.9
27	0.476	17.9	52.7	4.5
28	0.519	17.3	51.2	4.6
29	0.496	17.5	51.2	4.5
30	0.441	19.7	55.9	4.8
mean±s.d.	0.491±0.018	17.6±0.8	54.9±2.2	4.7±0.3

Table S6. *J-V* parameters of 30 ITO devices with four ITO layers after optimization. These data are used to form statistical histograms presented in Figures 3c-f.

E _g of perovskite /eV	Device structure	HTM	V _{oc} /V	J _{SC} /mA cm ⁻²	FF /%	PCE /%	Mask size /cm ²	Ref.
1.41	Regular	Spiro- OMeTAD	0.32	23.7	63	4.8	0.096	[S16]
1.41	Regular	Spiro- OMeTAD	0.238	24.45	36	2.1	unknown	[S17]
1.5	Regular	Spiro- OMeTAD	0.36	19.11	54.44	3.76	unknown	[S18]
1.5	Regular	Spiro- OMeTAD	0.43689	20.76	57.35	5.20	0.09	[S19]
1.5	Regular	Undoped Spiro- OMeTAD	0.42885	13.38	30.79	1.77	0.09	[819]
1.5	Regular	_	0.18239	20.27	42.09	1.56	0.09	[S19]
1.61	Regular	_	0.515	17.8	58.3	5.4	0.09	this work

Table S7. Comparison of photovoltaic parameter of ITO PSC (in this work) with HTM-based tin PSC (in the references); FA was used as a major cation. E_g stands for the band gap.

Device	V _{oc} /V	J _{SC} /mA cm ⁻²	FF /%	PCE /%
ΙΤΟ	0.515	17.8	58.3	5.4
Carbon	0.373	16.2	49.6	3.0

Table S8. J-V parameters of ITO and carbon devices after optimization. The corresponding J-V curves are presented in Figure 4a.

Publication year	Device structure	<i>IPCE at 650 nm /%</i>	Ref.
2020 (expected)	ITO	~78	This work
2020 (expected)	Carbon	~66	This work
2020	Inverted (p-i-n)	~77	[S20]
2020	Inverted	~74	[S21]
2020	Inverted	~57	[S22]
2020	Carbon	~69	[S23]
2020	Inverted	~69	[S24]
2020	Carbon	~58	[S25]
2020	Inverted	~75	[S26]
2020	Inverted	~67	[S27]
2020	Inverted	~71	[S28]
2020	Inverted	~67	[S29]
2020	Inverted	~73	[S30]
2020	Carbon	~64	[S31]
2019	Inverted	~64	[S32]
2019	Inverted	~79	[S33]
2019	Inverted	~62	[S34]
2019	Regular (n-i-p)	~75	[S18]
2019	Inverted	~78	[S35]
2019	Inverted	~65	[S36]
2019	Inverted	~60	[S37]
2019	Regular	~74	[S38]
2019	Regular	~55	[S39]
2019	Inverted	~64	[S40]
2019	Inverted	~88	[S41]

Table S9. Summary of IPCE at 650 nm of tin-based perovskite solar cells, for which the best IPCE was extracted from each literature. The numerical IPCE values were used in Figure 4c.

2019	Inverted	~77	[S42]
2019	Inverted	~87	[S43]
2019	Inverted	~69	[S44]
2018	Carbon	~69	[S45]
2018	Inverted	~75	[S46]
2018	Inverted	~79	[S47]
2018	Inverted	~70	[S48]
2018	Inverted	~68	[S49]
2018	Regular	~78	[S50]
2018	Inverted	~63	[S50]
2018	Inverted	~78	[S51]
2018	Regular	~72	[S51]
2017	Regular	~64	[S52]
2017	Inverted	~55	[\$53]
2017	Carbon	~31	[89]
2017 2017	Carbon Inverted	~31 ~60	[S9] [S54]
2017 2017 2017	Carbon Inverted Regular	~31 ~60 ~75	[S9] [S54] [S55]
2017 2017 2017 2017 2017	Carbon Inverted Regular Regular	~31 ~60 ~75 ~82	[S9] [S54] [S55] [S56]
2017 2017 2017 2017 2017 2017	Carbon Inverted Regular Regular Inverted	~31 ~60 ~75 ~82 ~54	[S9] [S54] [S55] [S56] [S57]
2017 2017 2017 2017 2017 2017 2017	Carbon Inverted Regular Regular Inverted Regular	~31 ~60 ~75 ~82 ~54 ~76	[\$9] [\$54] [\$55] [\$56] [\$57] [\$58]
2017 2017 2017 2017 2017 2017 2017 2016	Carbon Inverted Regular Regular Inverted Regular Inverted	~31 ~60 ~75 ~82 ~54 ~76 ~19	[S9] [S54] [S55] [S56] [S57] [S58] [S59]
2017 2017 2017 2017 2017 2017 2017 2016 2016	Carbon Inverted Regular Regular Inverted Regular Inverted Inverted	~31 ~60 ~75 ~82 ~54 ~54 ~76 ~19 ~69	[S9] [S54] [S55] [S56] [S57] [S58] [S59] [S60]
2017 2017 2017 2017 2017 2017 2017 2016 2016 2016	Carbon Inverted Regular Regular Inverted Regular Inverted Inverted Regular	~31 ~60 ~75 ~82 ~54 ~54 ~76 ~19 ~69 ~74	[S9] [S54] [S55] [S56] [S57] [S58] [S59] [S60] [S16]
2017 2017 2017 2017 2017 2017 2017 2016 2016 2016 2016 2016	Carbon Inverted Regular Regular Inverted Regular Inverted Inverted Regular Regular Regular	-31 -60 -75 -82 -54 -76 -19 -69 -74 -74 -47	[S9] [S54] [S55] [S56] [S57] [S58] [S59] [S60] [S16] [S61]
2017 2017 2017 2017 2017 2017 2017 2016 2016 2016 2016 2016 2016	Carbon Inverted Regular Regular Inverted Regular Inverted Inverted Regular Regular Regular Regular	$ \begin{array}{c} $	[S9] [S54] [S55] [S56] [S57] [S58] [S59] [S60] [S16] [S61] [S62]
2017 2017 2017 2017 2017 2017 2017 2016 2016 2016 2016 2016 2016 2016 2015	Carbon Inverted Regular Regular Inverted Regular Inverted Inverted Regular Regular Regular Inverted Regular	$ \begin{array}{c} -31 \\ -60 \\ -75 \\ -82 \\ -54 \\ -76 \\ -19 \\ -69 \\ -74 \\ -47 \\ -30 \\ -82 \\ \end{array} $	[S9] [S54] [S55] [S56] [S57] [S58] [S59] [S60] [S60] [S16] [S61] [S62] [S63]
2017 2017 2017 2017 2017 2017 2017 2016 2016 2016 2016 2016 2016 2016 2015	Carbon Inverted Regular Regular Inverted Regular Inverted Inverted Regular Regular Inverted Regular Regular Regular	$\begin{array}{c} -31 \\ -60 \\ -75 \\ -82 \\ -54 \\ -76 \\ -19 \\ -69 \\ -74 \\ -47 \\ -47 \\ -30 \\ -82 \\ -50 \end{array}$	[S9] [S54] [S55] [S56] [S56] [S57] [S58] [S59] [S60] [S60] [S16] [S61] [S62] [S63] [S63] [S64]

Device	R_S / Ω	R_{ch} / Ω	R_{rec} / Ω	R_{rel} / Ω
ΙΤΟ	39.0	-	64.0	17.9
Carbon	33.8	3.8	19.0	12.1

Table S10. Fitted EIS parameters of ITO and carbon devices; R_S , R_{ch} , R_{rec} and R_{rel} stand for series, charge-transfer, recombination, dielectric-relaxation resistances, respectively. The corresponding EIS plots and equivalent circuits are shown in Figure S8.

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