Electronic Supplementary Material (ESI) for Energy & Environmental Science.

SUPPORTING INFORMATION

Highly efficient and stable planar perovskite solar cells by solution-processed tin oxide

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Note 1:

Optimization of solution processed SnO_2 layers.

In order to improve the reproducibility of the deposition of the layers, two additional procedures were tested. A third method, involved solely the chemical bath deposition in a diluted form (CBD-d) as shown in Figure 1b. Finally, a method using spin coating and a dilute version of the chemical bath (SC-CBD-d) was used to optimize this layer. The properties of these films and their respective devices will be discussed throughout the supporting information.

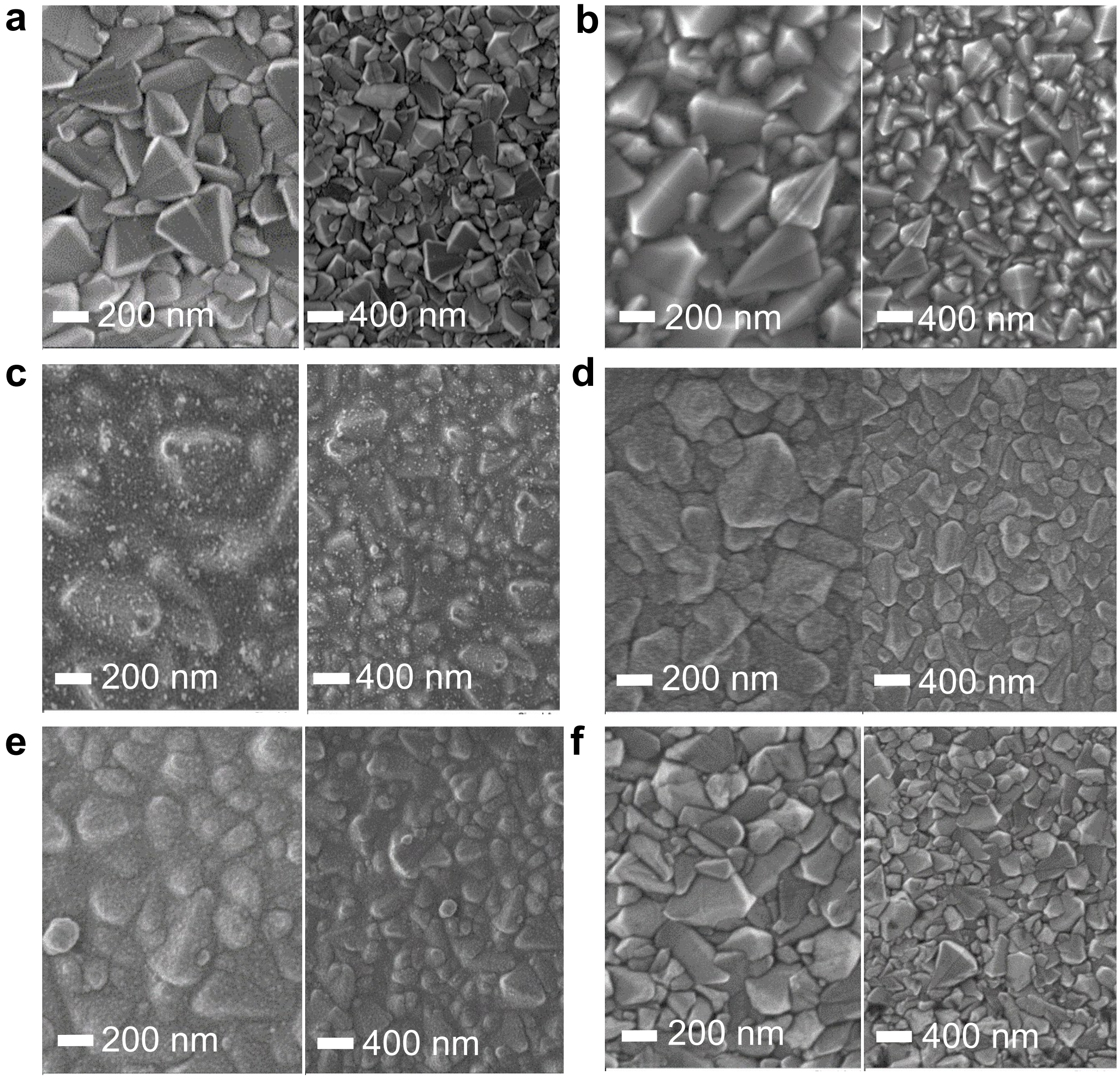

Figure S1. Top view SEM images of different ESL deposition methods. a. ALD , **b.** spin coated SnO_2 (SC), **c.** spin coated SnO_2 and chemical bath post-treatment (SC-CBD), **d.** chemical bath only (CBD), **e.** spin coated SnO_2 and dilute chemical bath post-treatment (dilute SC-CBD) deposited on FTO as ESLs, and (f) Bare FTO. All micrographs in two different magnifications.

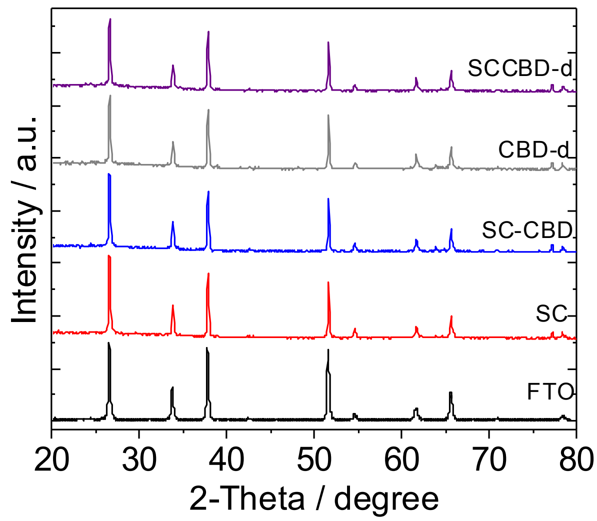

Figure S2. Structural features of the different ESLs. XRD patterns of bare FTO, spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), dilute chemical bath only (CBD-d), spin coated SnO_2 and dilute chemical bath post-treatment (SC-CBD-d), deposited on FTO as ESLs

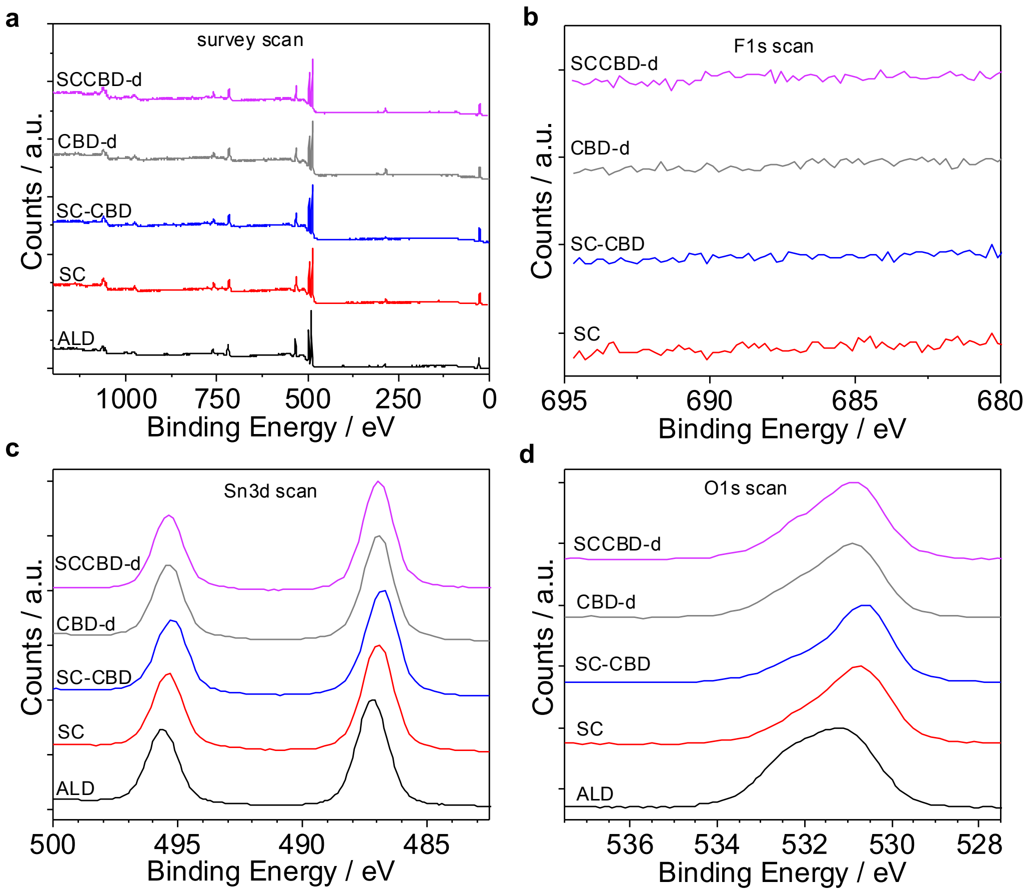

Figure S3. X-ray photoelectron spectroscopy of the different ESLs. (a) Survey of elements and their related high resolution spectra of (b) F 1s, (c) Sn 3d and (d) O1s and peaks as a function of their deposition method containing: atomic layer deposited (ALD), spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), dilute chemical bath only (CBD-d), spin coated SnO_2 and dilute chemical bath post-treatment (SC-CBD-d), deposited on FTO as ESLs.

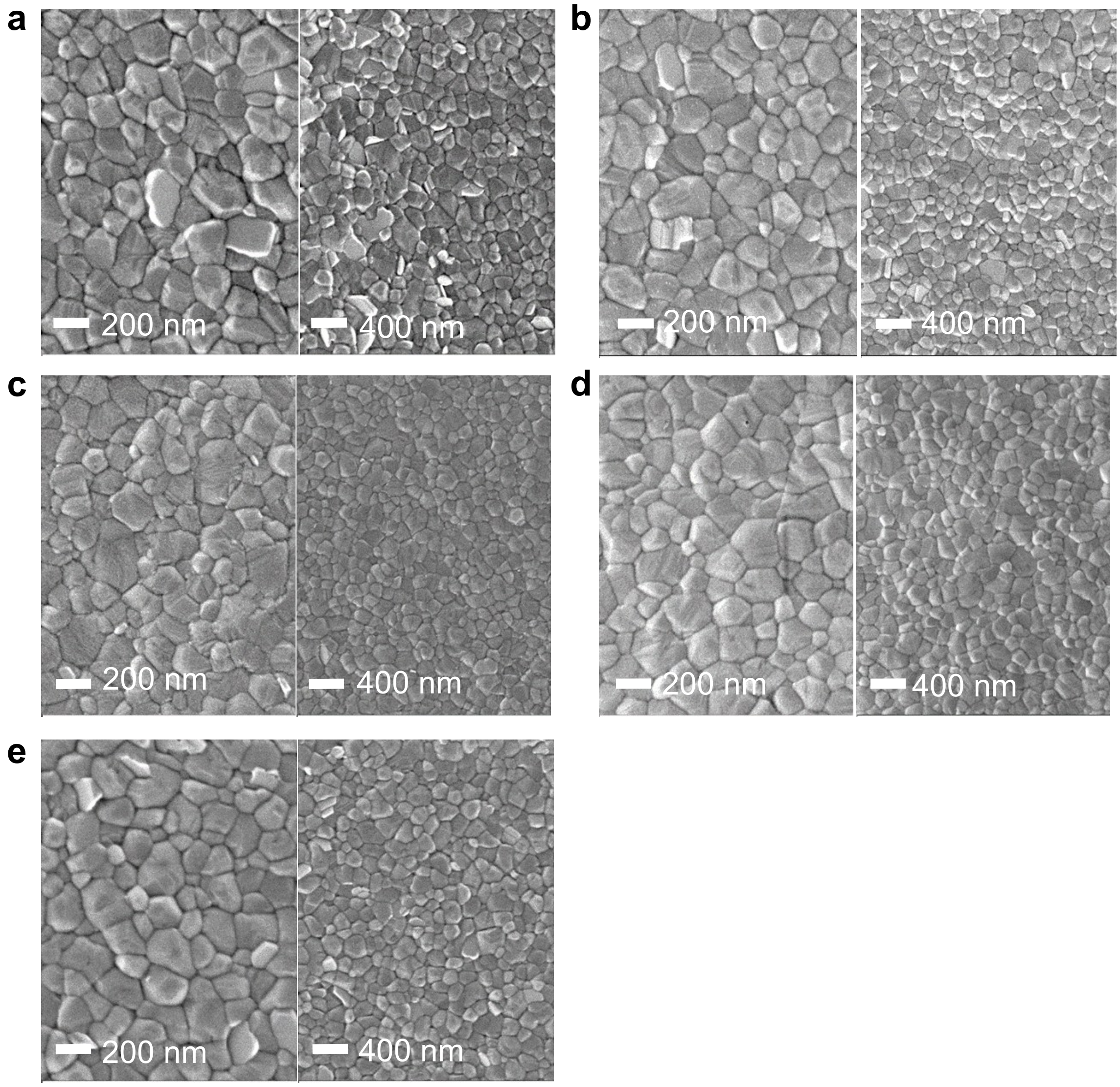

Figure S4. Morphological features of perovskite films. Top view SEM images of perovskite deposited on: **a.** ALD , **b.** spin coated SnO_2 (SC), **c.** spin coated SnO_2 and chemical bath post-treatment (SC-CBD), **d.** dilute chemical bath only (CBD-d), **e.** spin coated SnO_2 and dilute chemical bath post-treatment (dilute SC-CBD). Two different magnifications per sample are shown.

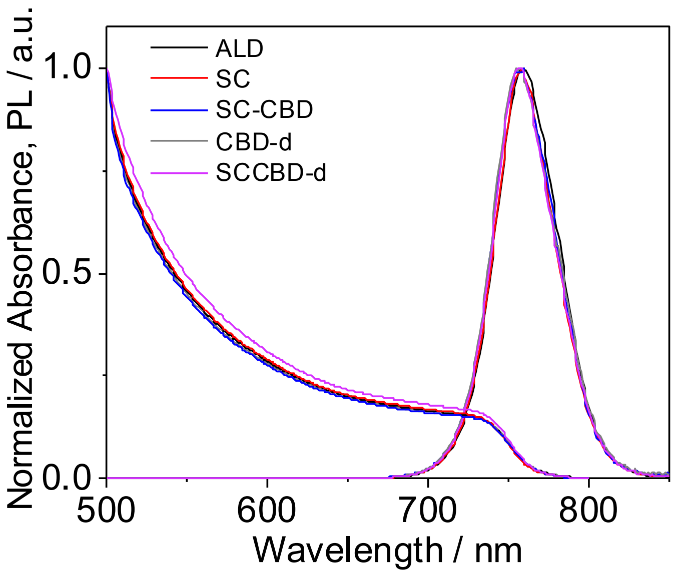

Figure S5. Optical properties of perovskite films. Normalized UV-vis absorption and normalized steady state photoluminescence of perovskite films deposited on: atomic layer deposited (ALD), spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), dilute chemical bath only (CBD-d), spin coated SnO_2 and dilute chemical bath post-treatment (SC-CBD-d), deposited on FTO as ESLs.

Photoluminescence and UV-visible spectra (Figure S5) of the mixed ion perovskite material show a narrow peak with the maxima located at 758 nm, and a sharp absorption onset at ca. 760 nm, respectively.

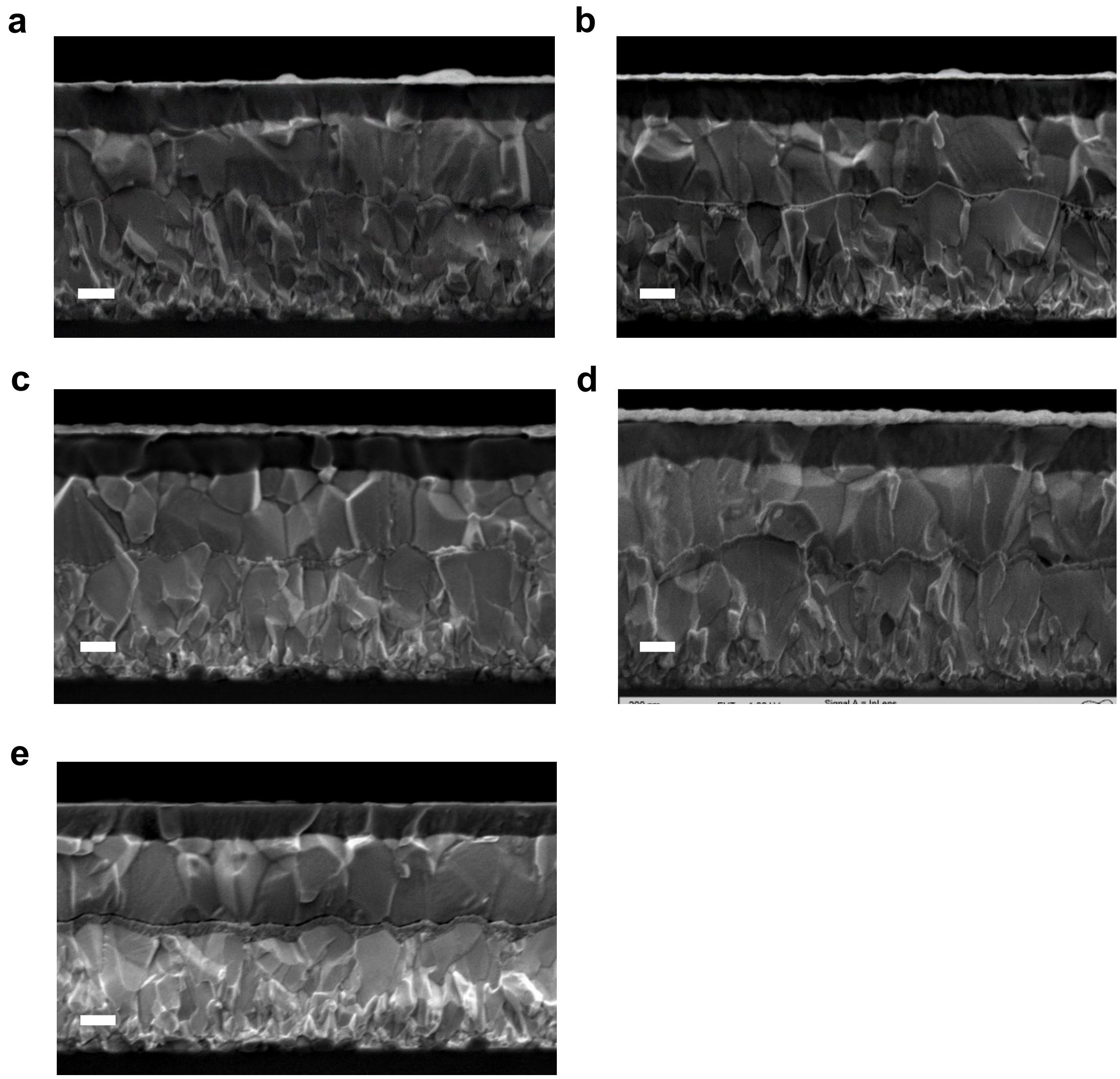

Figure S6. Cross-sectional SEM images for devices using different configurations. **a.** ALD, **b.** spin coated SnO_2 (SC), **c.** spin coated SnO_2 and chemical bath post-treatment (SC-CBD), **d.** dilute chemical bath only (CBD-d), **e.** spin coated SnO_2 and dilute chemical bath post-treatment (dilute SC-CBD). The scale bar is 200 nm.

Note 2:

Investigation of reproducibility and blocking properties of all layers:

To understand the electron blocking properties and coverage quality of the SnO_2 layers deposited with the different methods, cyclic voltammetry was performed using the aqueous solution of $Fe(CN)_6^{3-/4-}$ as the redox couple.[1] Cyclic voltammetry was performed for at least three random samples of each condition and at least three consecutive voltage scans for each sample to be sure of reproducibility. All deposition methods yielded sufficient blocking compared to bare FTO as shown in Figure S7a, with SC and SC-CBD SnO_2 films showing some source of leakage. Depositions of SnO_2 films with CBD and the dilute SC-CBD showed perfectly blocking properties. This behavior is also seen in the dark current-voltage sweeps of full devices, as shown in Figure S7b. Here, the SC and SC-CBD show more leakage than the ALD controls and the devices treated with the dilute chemical bath (CBD-d and SCCBD-d).

Because reproducibility is important in the lab and industry, we produced a large number of samples per deposition group to understand the number of shunt devices due to poor coverage of the SnO_2, as summarized in Figure S7c. The lowest number of shunts was found for devices in the ALD control and SC-CBD-d whereas relatively higher percentage of shunts was found for SC devices and those with CBD-d. These results stress the importance of using a combination of spin-coating and a dilute chemical bath for improved reproducibility. We note that the least number of shunts, while maintaining power conversion efficiencies close to 20%, were obtained with SCCBD-d, and therefore this is the optimized procedure for reproducibility.

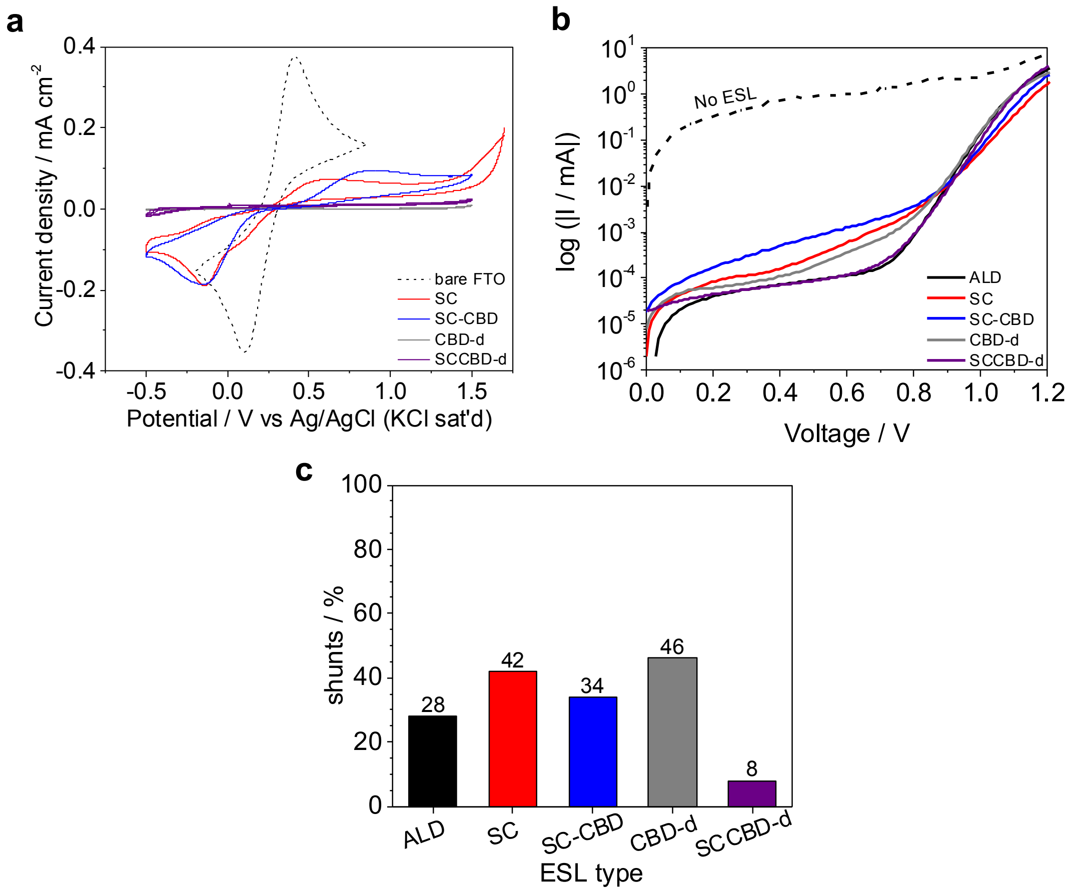

Figure S7. Electron blocking properties of SnO_2 films deposited by different methods. a. Cyclic voltammograms of different ESLs deposited on FTO electrode, measured with a scan rate of 100 mV s^{-1} in 1 mM $K_4Fe(CN)_6$ + 1 mM $K_3Fe(CN)_6$ in aqueous 0.5 M KCl, pH 7 as electrolyte solution, **b.** dark J-V characteristics and **c.** shunt percentage (Devices that showed short-circuited J-V measurements, but had no visible damage during film deposition) for devices employing bare FTO, atomic layer deposited (ALD), spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), dilute chemical bath only (CBD-d), spin coated SnO_2 and dilute chemical bath post-treatment (SC-CBD-d), deposited on FTO as ESLs.

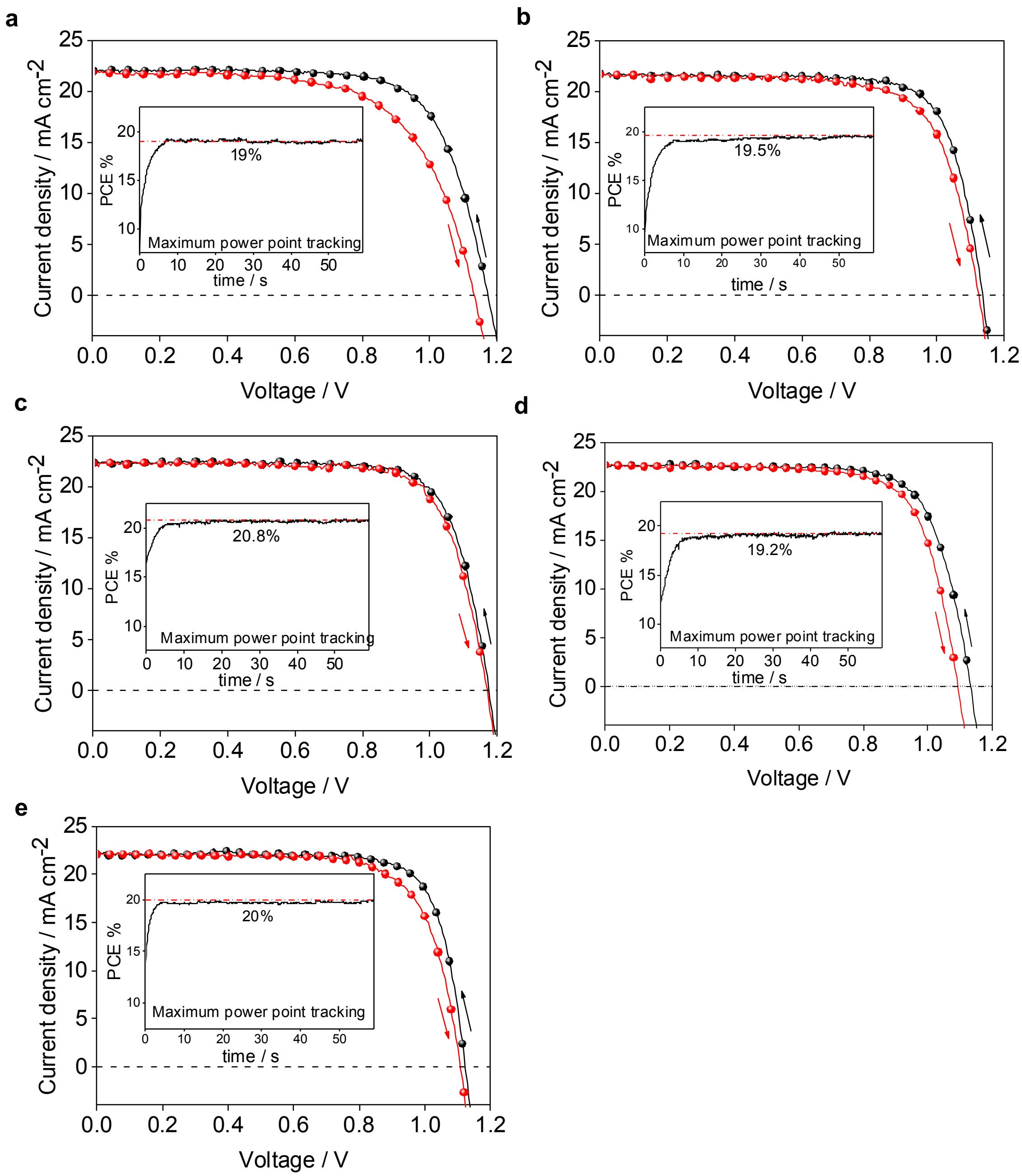

Figure S8. Current-voltage and stabilized PCE characteristics of devices. The J-V curves and maximum power point tracking (MPPT, as the inset) of the best performing planar perovskite solar cells based for **a.** ALD , **b.** spin coated SnO_2 (SC), **c.** spin coated SnO_2 and chemical bath post-treatment (SC-CBD), **d.** dilute chemical bath only (CBD-d), **e.** spin coated SnO_2 and dilute chemical bath post-treatment (dilute SC-CBD).

Table. S1 Backward scan (BW) photovoltaic parameters and maximum power point tracking of planar perovskite solar cells. Scan rate of 10 mV/s at room temperature.

ESL type	$\mathbf{V_{OC}}$ **[V]**	$\boldsymbol{J_{SC}}$ **[mA cm^{-2}]**	***FF***	***Champion PCE*** **[%]**	**Light intensity [mW cm^{-2}]**	**MPP [%]**
ALD	1.17	21.95	0.72	19.17	97.1	19.01
SC	1.14	21.72	0.76	19.21	97.2	19.5
SC-CBD	1.18	22.37	0.77	20.46	98.4	20.78
CBD-d	1.13	22.70	0.74	19.66	97	19.24
SC-CBD-d	1.12	22.12	0.77	19.94	96.3	19.96

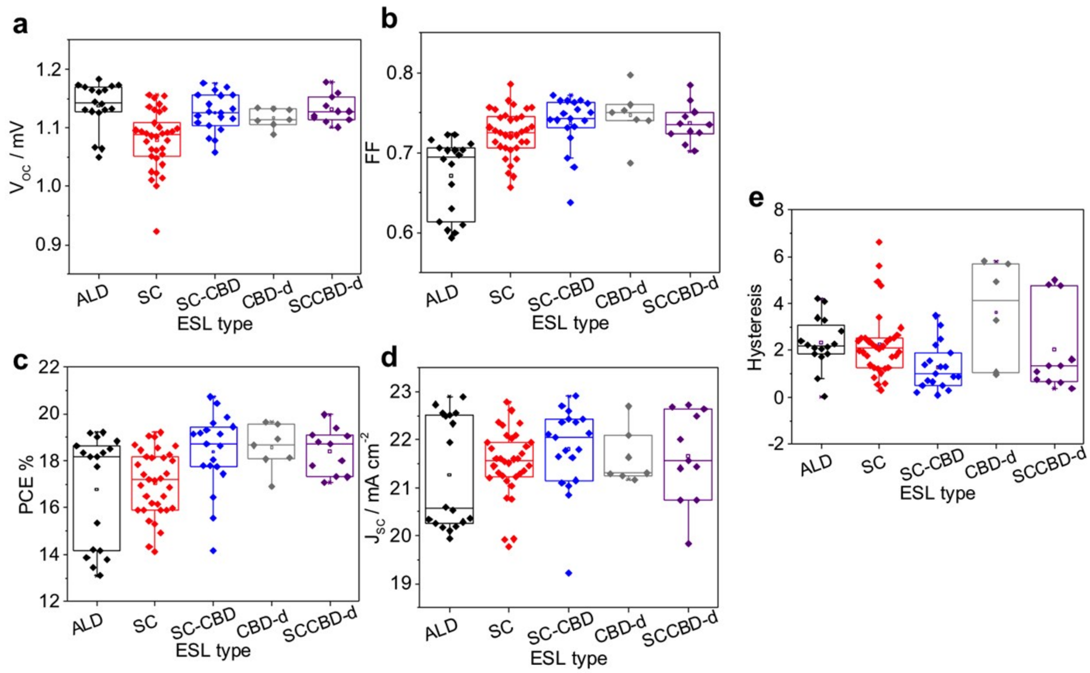

Figure S9. Performance parameters of PSCs. a. Open-circuit voltage **b.** fill factor, **c.** short-circuit current and **d.** power conversion efficiency and **e.** hysteresis (defined as the difference in efficiency between the backward and forward scan) of several planar perovskite solar cells based on atomic layer deposited (ALD), spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), dilute chemical bath only (CBD-d), spin coated SnO_2 and dilute chemical bath post-treatment (SC-CBD-d), deposited on FTO as ESLs. All J-V data were recorded with a scan rate of 10 mV/s under simulated one sun illumination (AM 1.5G) at room temperature.

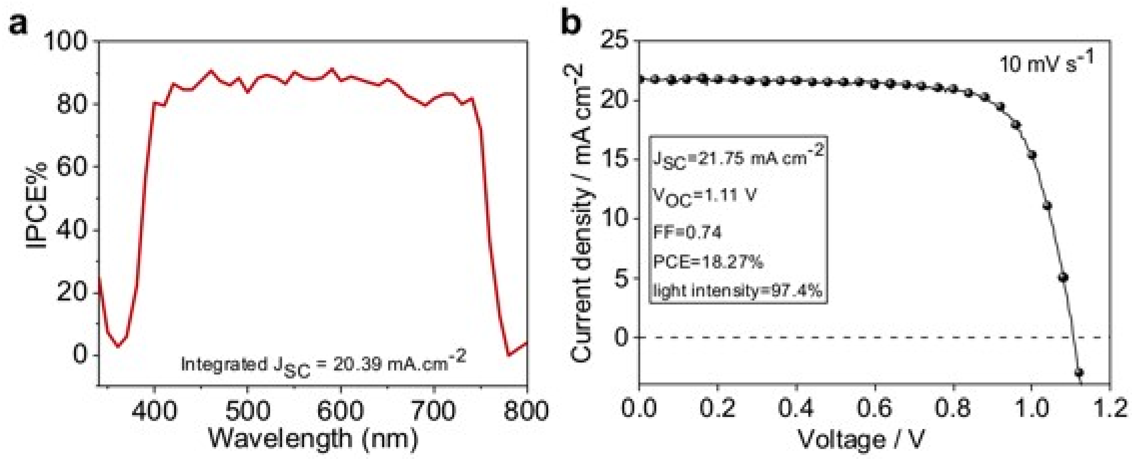

Figure S10. Incident photon-to-current efficiencies (IPCE) of solution processed planar perovskite solar cell based on spin coated SnO_2 and chemical bath post-treatment (SC-CBD).

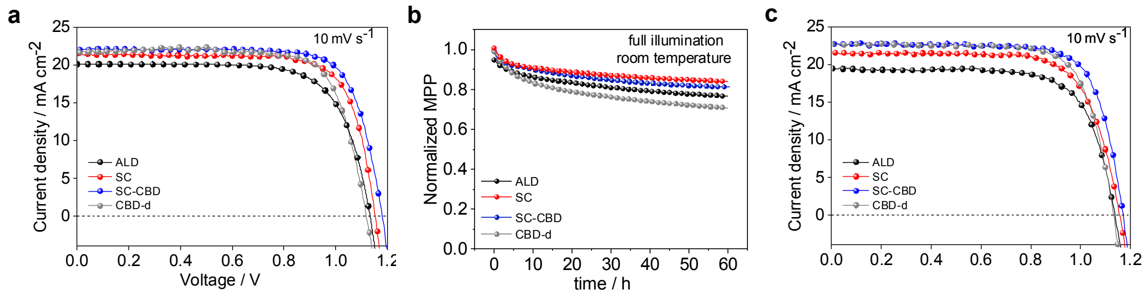

Figure S11. Current-voltage and stability characteristics of planar PSCs. a. Backward scan J-V characteristics before aging, **b.** long-term ageing MPPT under constant full sun illumination, and **c.** J-V curves after ageing of high performance planar devices based on ALD, spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), and dilute chemical bath only (CBD-d) ESLs.

Table S2. Backward photovoltaic characteristics before and after aging (under constant full sun illumination at room temperature and constant purging of nitrogen gas) of high performance planar perovskite solar cells with SnO_2-based ESLs based on atomic layer deposition (ALD), spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), dilute chemical bath only (CBD-d), deposited on FTO as ESLs. All measured at 10 mV s^{-1} scan rate under full sun illumination at room temperature.

ESL type	Measurement state	V_{OC} [V]	J_{SC} [mA cm^{-2}]	*FF*	*PCE* [%]	Light intensity [mW cm^{-2}]
ALD	Before aging	1.13	20.29	0.72	18.34	90.2
	After aging	1.14	19.43	0.72	16.35	96.8
SC	Before aging	1.15	21.38	0.75	19.17	96.6
	After aging	1.16	21.40	0.73	18.42	97.6
SC-CBD	Before aging	1.18	22.03	0.76	20.44	97
	After aging	1.17	22.59	0.75	20.73	96.1
CBD-d	Before aging	1.12	21.67	0.76	18.42	99.8
	After aging	1.13	22.70	0.74	19.66	97

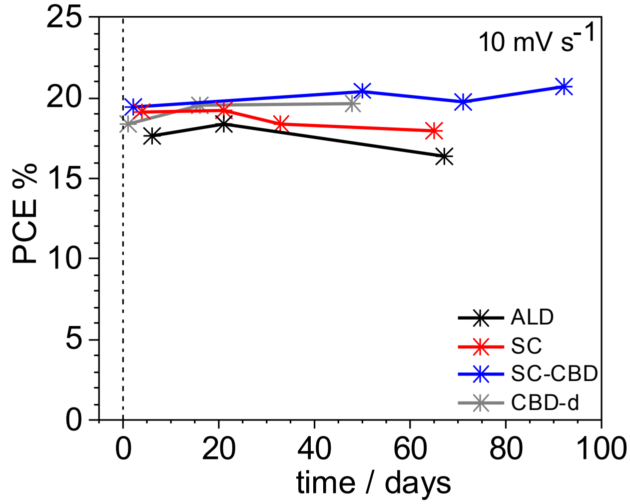

Figure S12. Dark, shelf stability of PSCs. Power conversion efficiency as a function of time for unencapsulated planar solar cells stored in dry air based on atomic layer deposition (ALD), spin coated SnO_2 (SC), spin coated SnO_2 and chemical bath post-treatment (SC-CBD), dilute chemical bath only (CBD-d), spin coated SnO_2 deposited on FTO as ESLs.

References

(1) Kavan, L.; Tétreault, N.; Moehl, T.; Grätzel, M. *The Journal of Physical Chemistry C* **2014**, *118*, 16408.