Supporting Information

Surface Modification of Halide Perovskite using EDTA-complexed

SnO₂ as Electron Transport Layer in High Performance Solar Cells

Nuno Marques, Santanu Jana, Manuel J. Mendes, Hugo Águas, Rodrigo Martins, Shrabani Panigrahi^{*}

CENIMAT|i3N, Department of Materials Science, School of Science and Technology, NOVA University Lisbon and CEMOP/UNINOVA, Campus de Caparica, 2829-516 Caparica, Portugal.

Corresponding Authors

Shrabani Panigrahi - <u>s.panigrahi@campus.fct.unl.pt</u>



Fig. S1: Calculated J_{SC} values from the EQE spectra for PSCs on (a) SnO_2 (7.5 %) and (b) E- SnO_2 (7.5 %) ETLs, respectively.



Fig. S2: The graph plots show the normalized PCE values for the PSCs on SnO_2 (7.5 %) and E-SnO₂ (7.5 %) ETLs, respectively.



Fig. S3: *J-V* curves of the highest-performing PSCs based on (a) SnO_2 and (b) E-SnO₂ ETLs, measured at reverse and forward scans, respectively. The inset table shows the performance parameters of the devices, including V_{OC}, J_{SC}, FF, and PCE, are summarized.



Fig. S4: Schematic representation of shallow trap levels created along the conduction band of perovskite (CB) due to excess PbI₂ in it.



Fig. S5: (a) and (b) AFM topography images for SnO_2 and $E-SnO_2$ ETLs. (c) and (d) surface profiles for the corresponding images of SnO_2 and $E-SnO_2$ ETLs. Surface height distribution corresponding to the images for (e) SnO_2 and (f) $E-SnO_2$ ETLs which show the slight decrease in broadening of the distribution for the $E-SnO_2$ ETLs.



Fig. S6: The electronic energy levels of both the tip and sample surface are examined under two conditions: (a) while they are electrically disconnected, and (b) and (c) after they are electrically connected.

As you can see in Figure S5 a, the vacuum levels of the tip and the sample surface are lined up when they are not electrically connected, but the Fermi energy levels are not. The Fermi levels will line up through electron flow after the electrical contact if the tip and sample surface are close enough for electron tunnelling. At this point, the system will reach a state of balance (Figure S5 b). The vacuum energy levels are no longer in the same place, though, and a V_{CPD} has formed between the tip and the sample. The tip and the surface of the sample are now charged. This force can be turned off. A voltage (V_{DC}) is applied that is the same value as the V_{CPD} but goes in the reverse direction. This will remove the surface charge in the contact area (Figure S5 c). The external bias V_{DC} = V_{CPD} is the same as the change in work function between the tip and the sample. The V_{CPD} is found by using the following equation:

$$V_{CPD} = \frac{\phi_{Tip} - \phi_{Sample}}{q}$$

Table S1: The values of Hall mobility, carrier concentration and sheet resistance for SnO_2 and E-SnO₂ ETLs.

Samples	Hall mobility (cm³V ⁻¹ s ⁻¹)	Carrier concentration (cm ⁻³)	Sheet resistance (Ohm/sq))
SnO₂	36.8	9.09 x 10 ¹¹	6.21 x10 ⁹
E-SnO ₂	140	9.45 x 10 ¹²	1.58 x 10 ⁹