

## Supporting Information

# Interlayers Engineering for Flexible Large-Area Planar Perovskite Solar Cells

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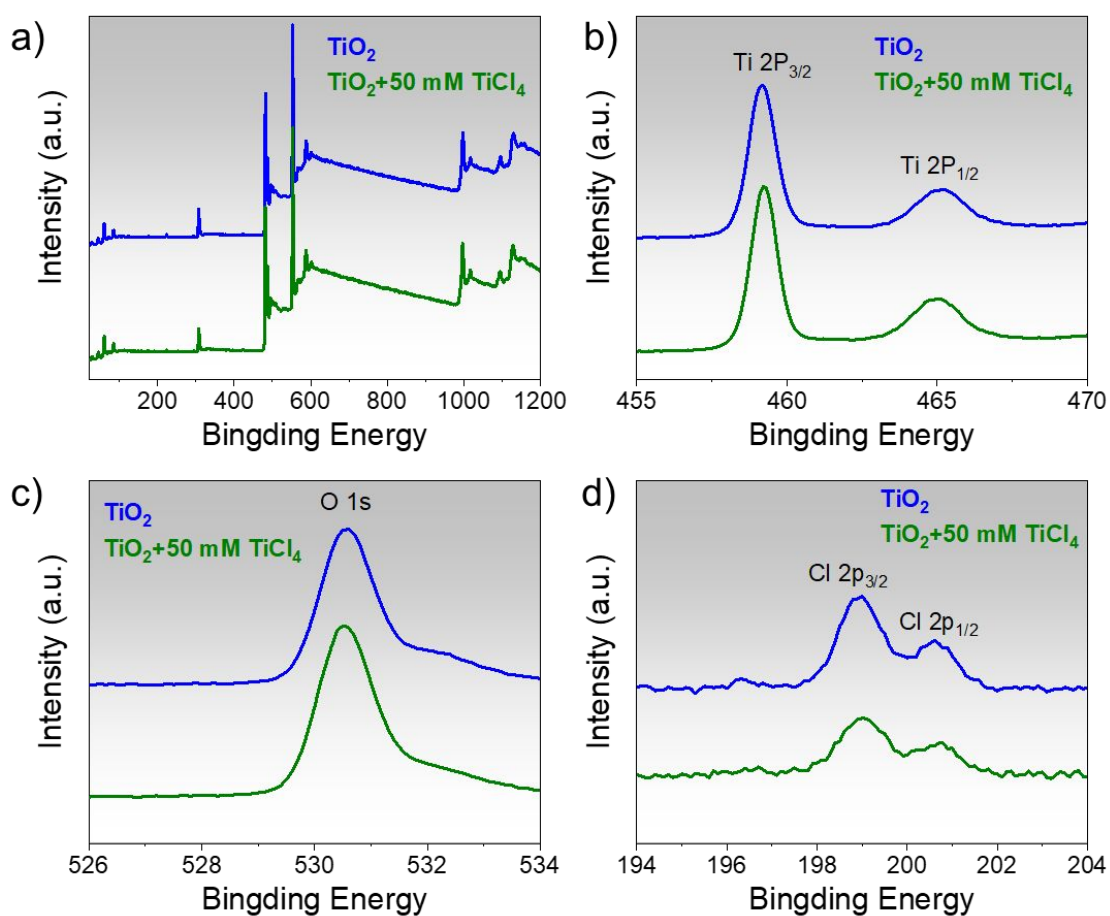
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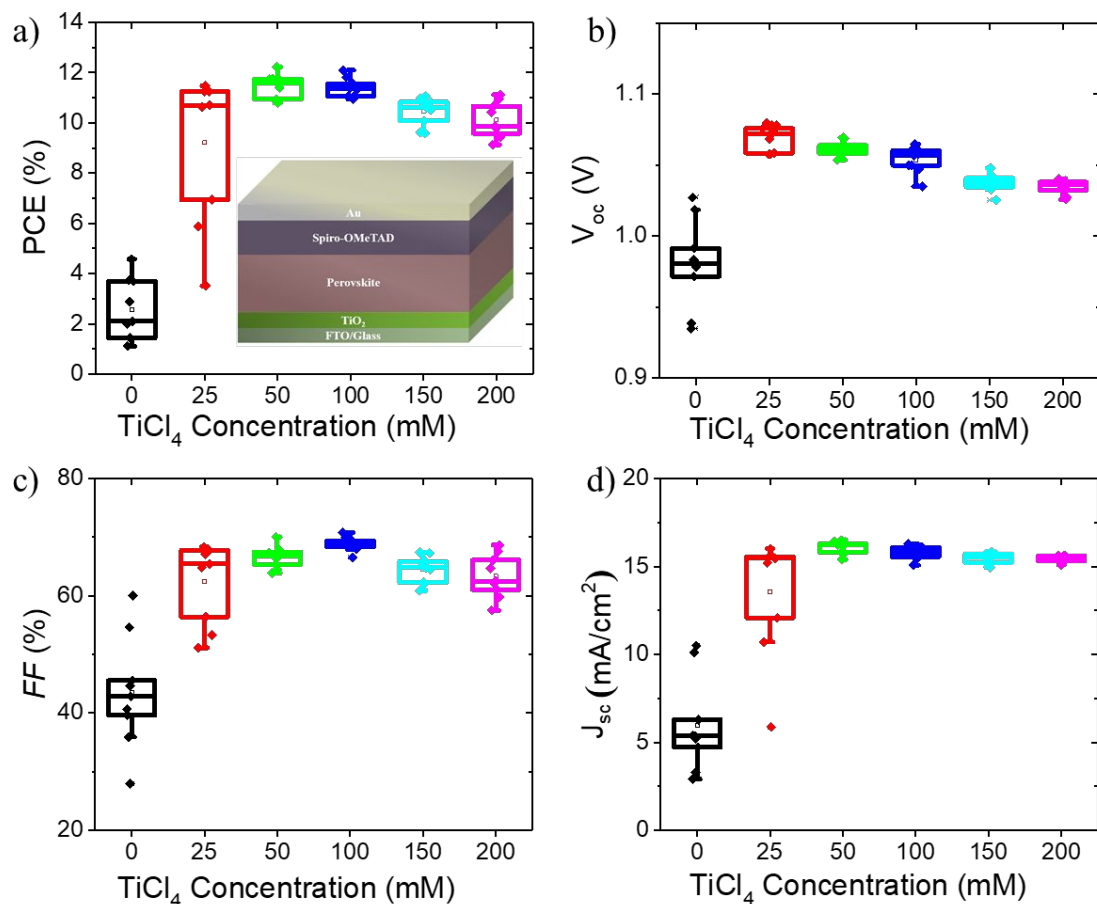
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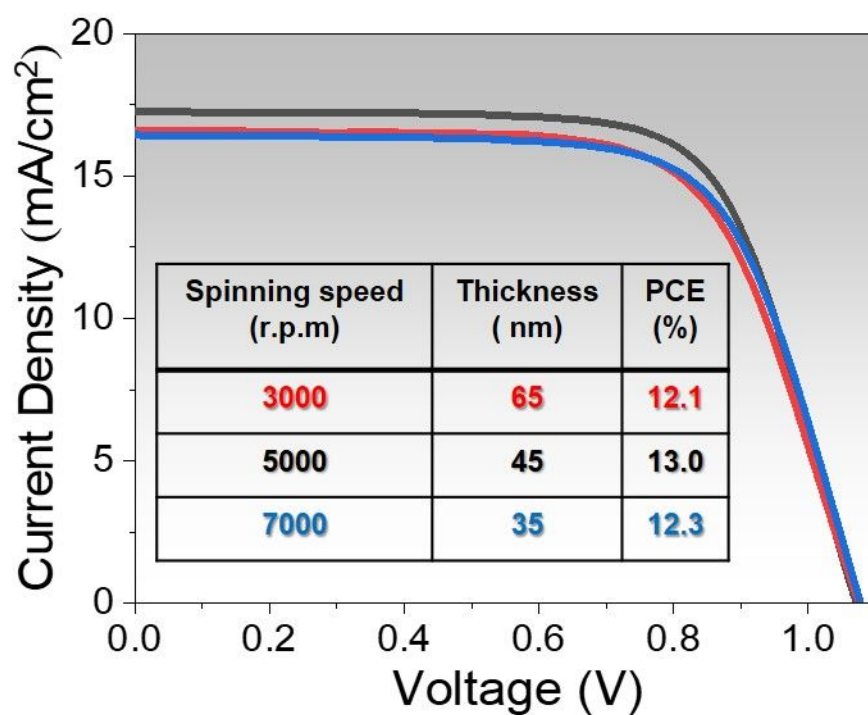
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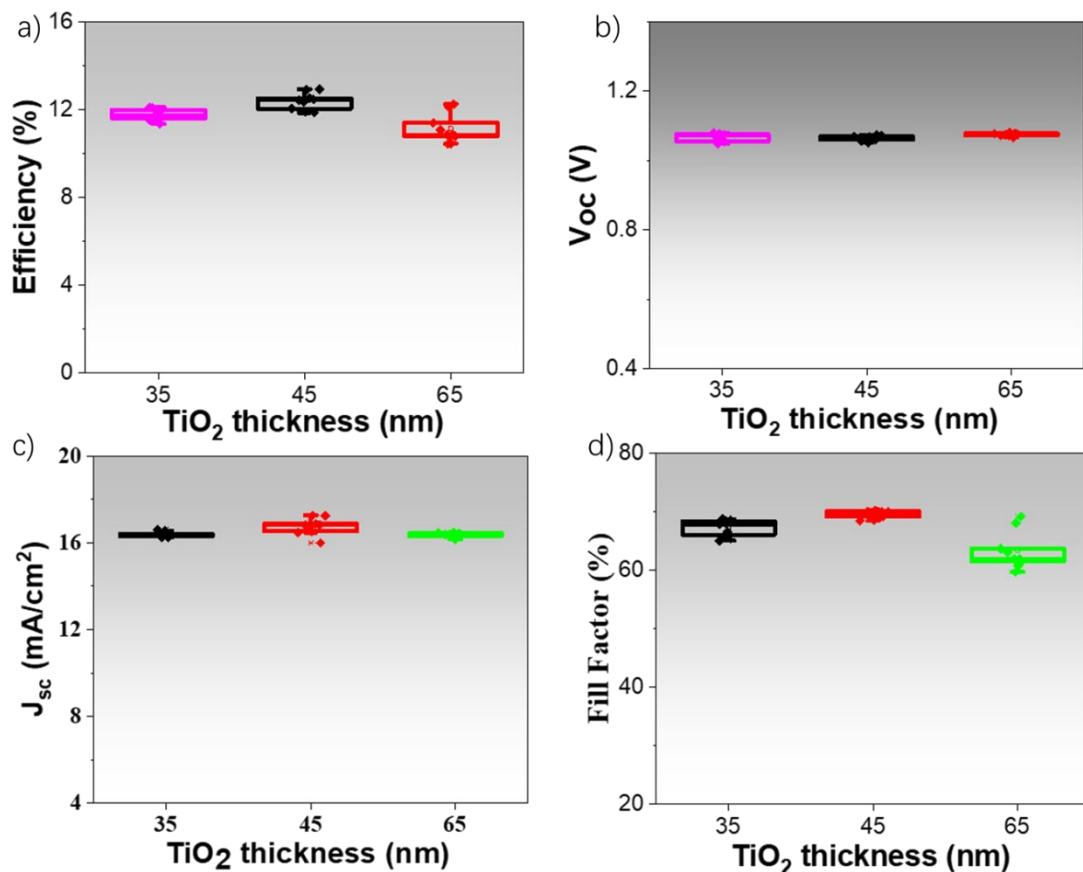
**Figure S1.** XPS spectra of  $\text{TiO}_2$  and  $\text{TiCl}_4$ -terated  $\text{TiO}_2$  films a) full spectra, b) Ti 2p, (c) O 1s, and (d) Cl 2p.



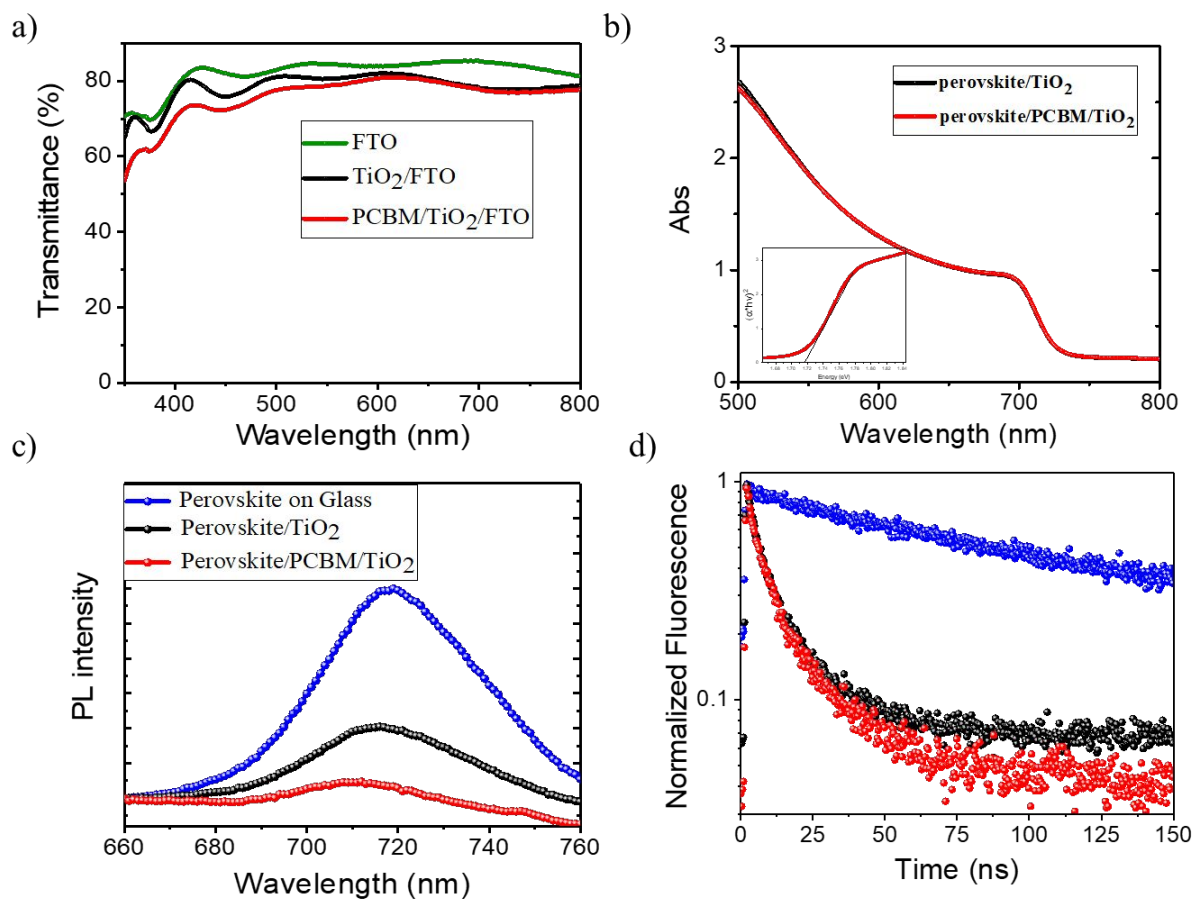
**Figure S2.** N-i-p planar 1.72 eV perovskite solar cells (PSCs) on glass with active area 0.16  $\text{cm}^2$ , the compact  $\text{TiO}_2$  layer quality was optimized to by varying  $\text{TiCl}_4$  addition and so the  $\text{TiCl}_4$  concentrations varied from 0 to 200 mM in the  $\text{TiO}_2$  dispersion. The highest PCEs (~12%) were obtained for  $\text{TiCl}_4$  concentrations 50 mM, while for higher ones the excess of  $\text{Cl}^-$  ligands leads to a reduction of film conductivity, Statistics data over 9 PSCs for each  $\text{TiCl}_4$  concentration: a) PCEs, the PSCs layered structure is reported in the inset), b)  $V_{oc}$ , c) FF and d)  $J_{sc}$  distribution.



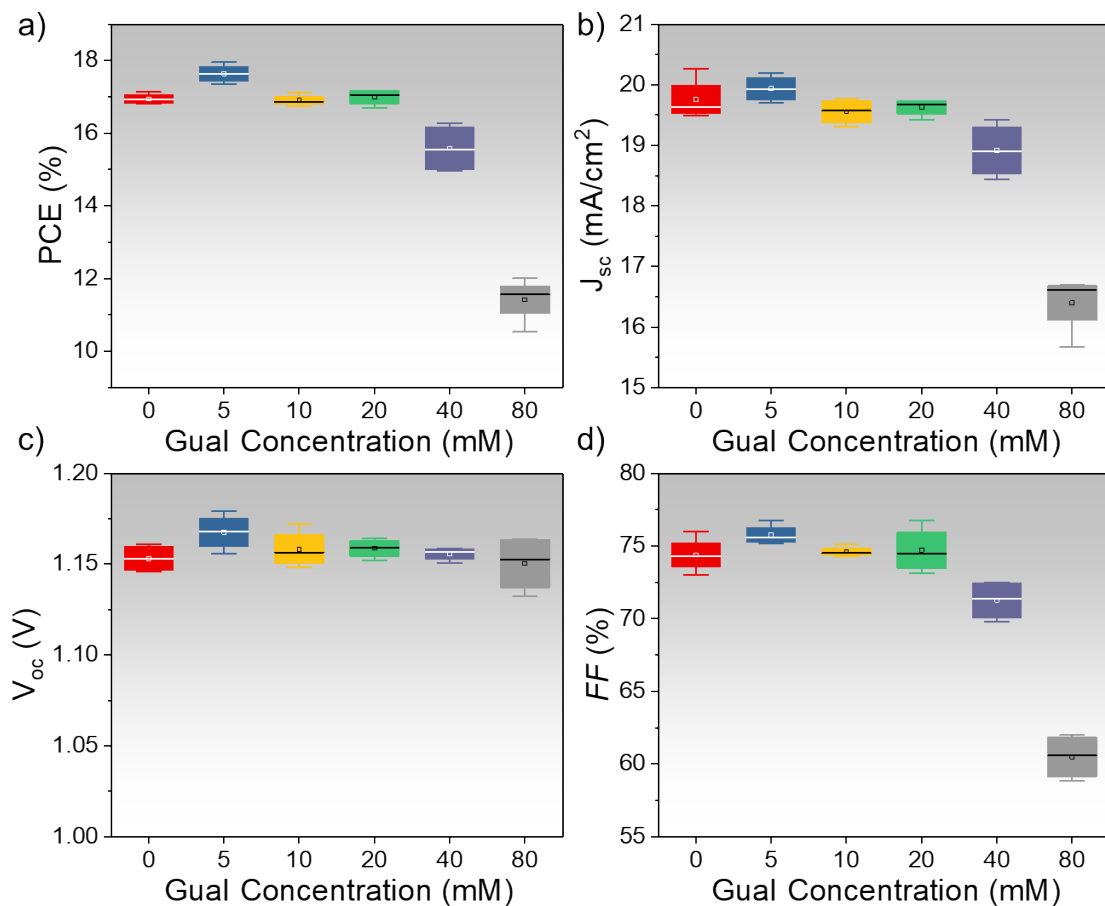
**Figure S3.** N-i-p planar 1.72 eV perovskite solar cells (PSCs) on glass with active area 0.16 cm<sup>2</sup>, the compact TiO<sub>2</sub> layer thickness was tuned by varying spinning speed of TiO<sub>2</sub> suspension with 50 mM TiCl<sub>4</sub>. J-V curves of PSCs based varying TiO<sub>2</sub> thickness, inset table show TiO<sub>2</sub> thickness and their corresponding PCE.



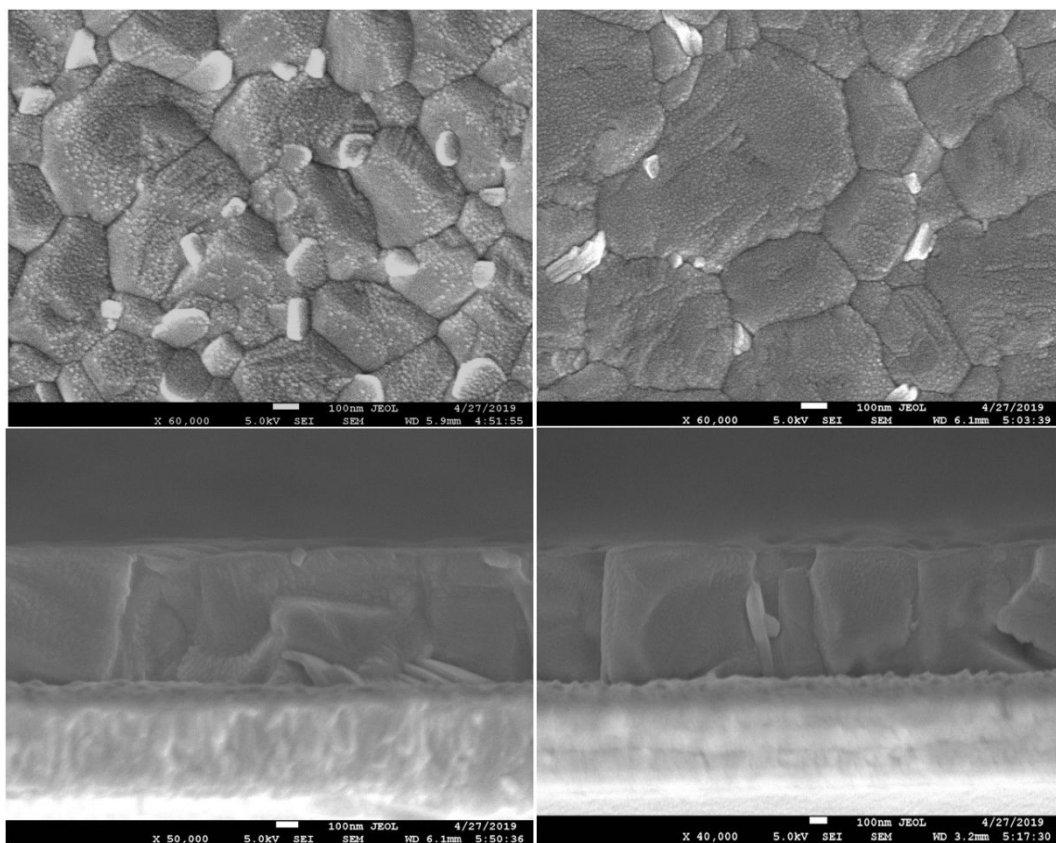
**Figure S4.** N-i-p planar ~1.72 eV perovskite solar cells (PSCs) on glass with active area 0.16 cm<sup>2</sup>, the compact  $\text{TiO}_2$  thickness was optimized to by varying spin-coating speed. The highest PCEs (~13%) were obtained for  $\text{TiO}_2$  thickness of ~45 nm, Statistics data over 9 PSCs for each  $\text{TiO}_2$  thickness: a) PCEs, the PSCs layered structure is reported in the inset), b)  $V_{oc}$ , c)  $J_{sc}$  and d)  $FF$  distribution.



**Figure S5.** Characterization of  $\text{Cs}_{0.15}\text{FA}_{0.85}\text{Pb}(\text{I}_{0.7}\text{Br}_{0.3})_3$  thin film on FTO,  $\text{TiO}_2$  and  $\text{PCBM}/\text{TiO}_2$ . a) transmittance spectra of FTO,  $\text{TiO}_2/\text{FTO}$  and  $\text{PCBM}/\text{TiO}_2/\text{FTO}$  substrates, b) absorption spectra of perovskite layer deposited on  $\text{TiO}_2/\text{FTO}$  and  $\text{PCBM}/\text{TiO}_2/\text{FTO}$ , c) steady state and d) time-resolved PL spectra of perovskite on glass,  $\text{TiO}_2/\text{FTO}$  and  $\text{PCBM}/\text{TiO}_2/\text{FTO}$  substrates.

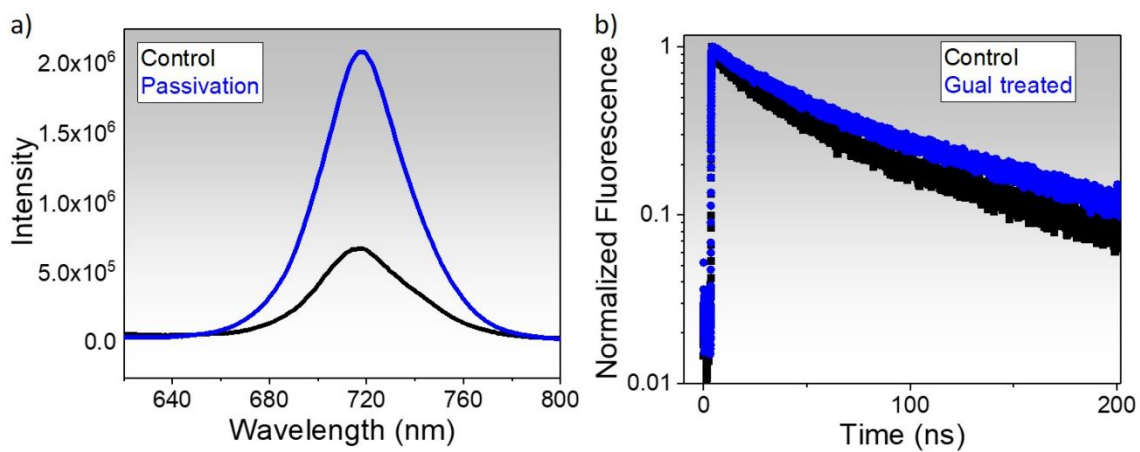


**Figure S6.** Photovoltaics performances of n-i-p planar 1.72 eV perovskite solar cells (PSCs) as function of guanidium iodide (Gual) passivation concentration (from 0 to 80 mM): a) efficiency, b)  $J_{sc}$ , c)  $V_{oc}$  and d) Fill Factor. The highest efficiencies (~18%) were obtained for Gual concentrations 5 mM.

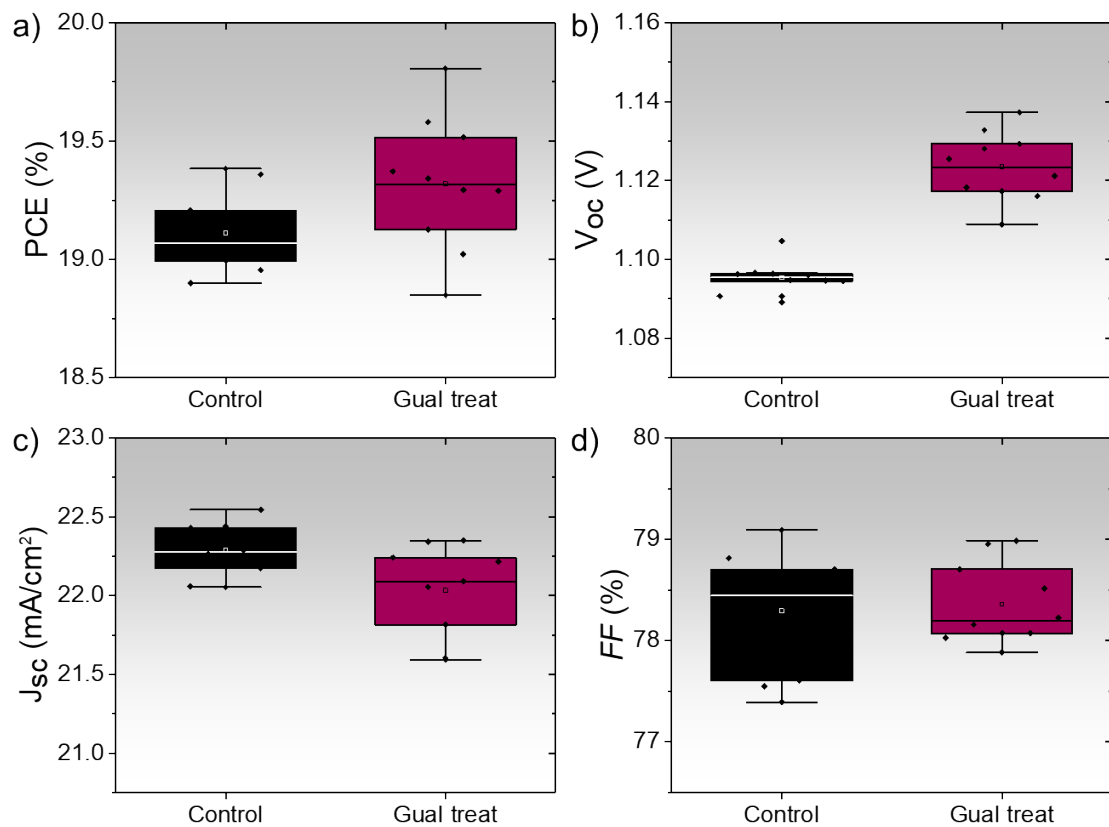


**Figure S7.** Top view and cross-sectional SEM images of 1.72 eV perovskite film before and after Gual treatment.

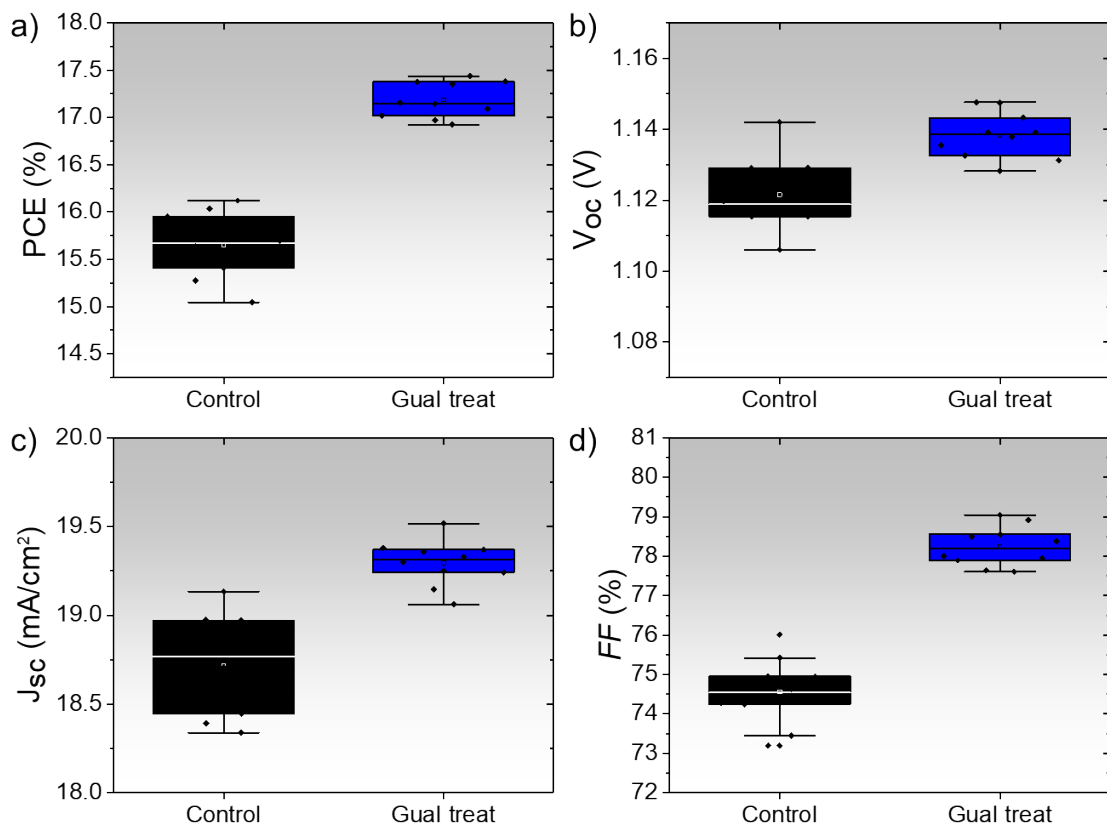




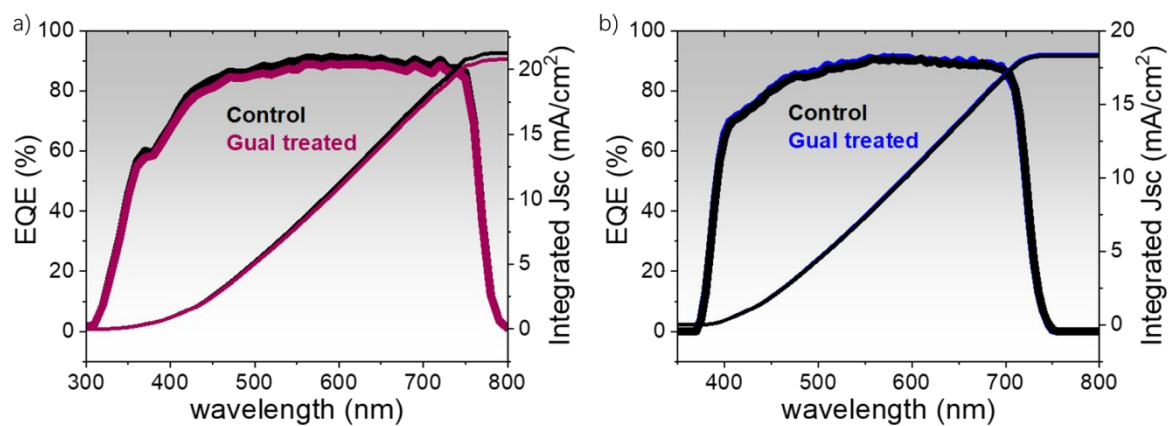
**Figure S8.** a) Steady-state spectra and b) time-resolved PL decays of untreated (control) and Gual-treated 1.72 eV perovskite on glass.



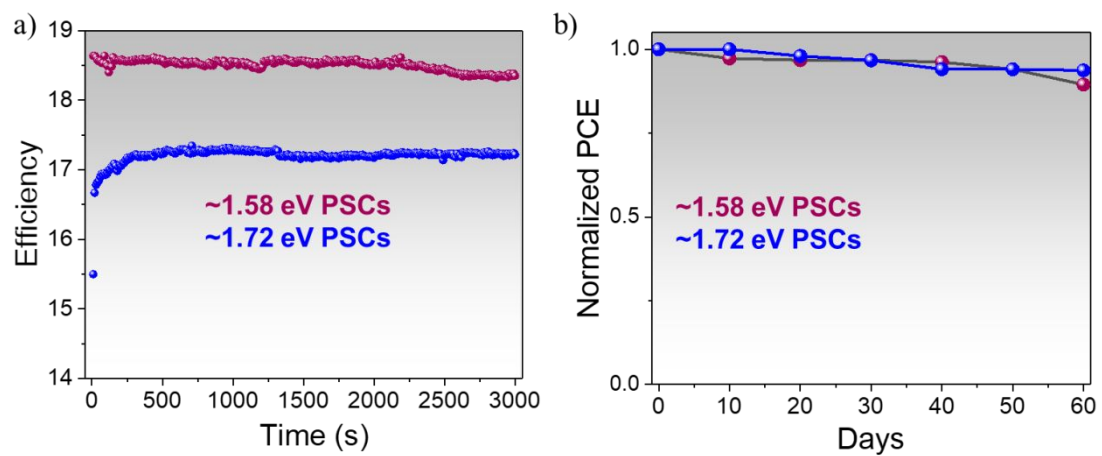
**Figure S9.** Photovoltaics performances of n-i-p planar untreated (control) and Gual-treated 1.58 eV perovskite solar cells (PSCs) on glass with active area 0.16 cm<sup>2</sup>: a) PCE, b)  $V_{oc}$  c)  $J_{sc}$  and d) Fill Factor. Statistics data over 20 PSCs for each condition.



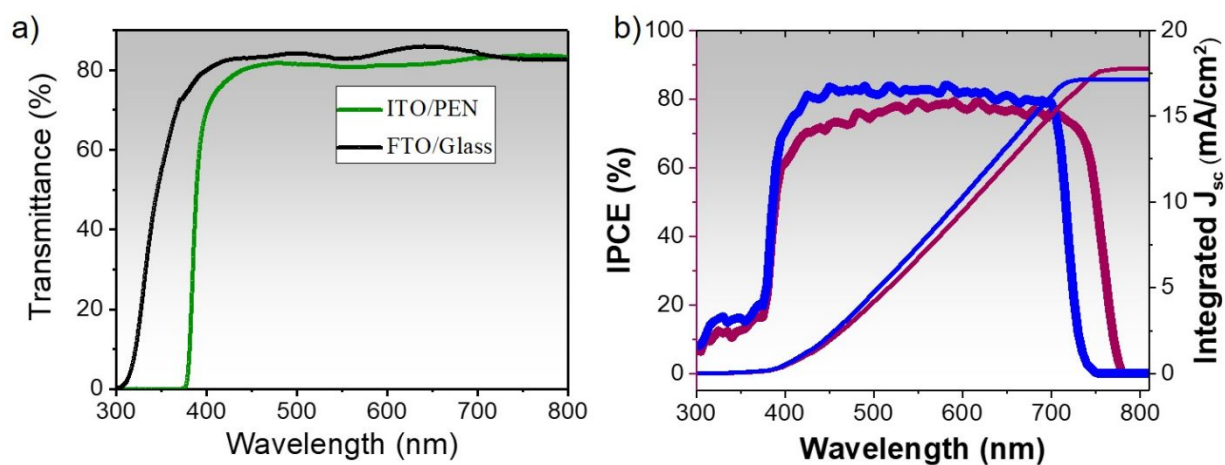
**Figure S10.** Photovoltaics performances of n-i-p planar untreated (control) and GualI-treated 1.72 eV perovskite solar cells (PSCs) on glass with active area 0.16 cm<sup>2</sup>: a) PCE, b)  $V_{oc}$  c)  $J_{sc}$  and d) Fill Factor Statistics data over 20 PSCs for each condition.



**Figure S11.** a) IPCE of untreated (control) and GuaI-treated 1.58 eV PSCs, b) IPCE of untreated (control) and GuaI-treated 1.72 eV PSCs.



**Figure S12.** a) Maximum power point track over 3000 s under 1 sun condition (AM 1.5), b) shelf stability under around 35% humidity of both 1.58 and 1.72 eV PSCs.



**Figure S13.** a) Transmittance of FTO/glass and ITO/PEN, b) IPCE of both Gual-treated 1.58 eV and 1.72 eV PSCs on PEN substrates.