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Scalable Open-Air Deposition of Compact ETL TiO_x on Perovskite for Fullerene-Free Solar Cells

Justin P. Chen,[a,†] *Florian Hilt,*[a,c,†] *Nicholas Rolston,* [b] *Reinhold H. Dauskardt* [a*]

[a] Department of Materials Science and Engineering
Stanford University, Stanford, CA, 94305-2205, United States
E-mail: rhd@stanford.edu

[b] Department of Applied Physics
Stanford University, Stanford, CA, 94305-2205, United States

[c] Now at Institut Photovoltaïque d'Île-de-France (IPVF), 91120 Palaiseau, France

[†] Equal Contribution

Keywords: Electron Transport Layer, Stability, Titanium Oxide, Perovskite Solar Cells, Chemical Vapor Deposition

Carrier N_2 Flow Rate:	4 SLM
Precursor Concentration:	80 vol. % $Ti(OEt)_4$, 20 vol. % Toluene
Precursor Flow Rate:	2 µL/min
Evaporator Temperature:	250°C
Substrate Temperature:	100°C
Nozzle to Substrate Distance:	1 mm
Raster Line Speed:	10 cm/s
Raster Line Spacing:	0.5 mm
Number of Passes:	8

Table S1: OA-CVD deposition parameters for 70 nm thick TiO_x film.

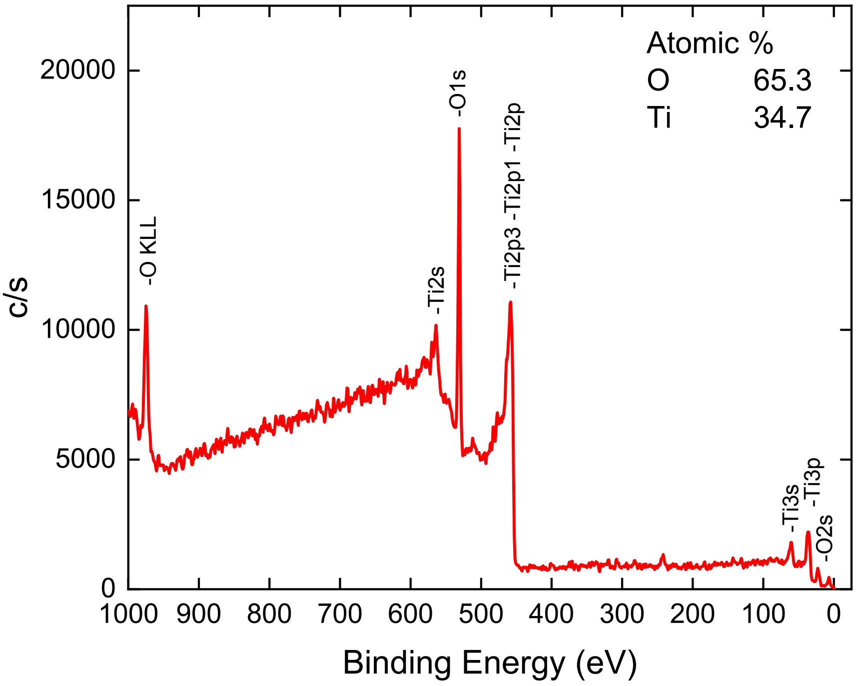

Figure S1: XPS survey spectra of TiO_x film.

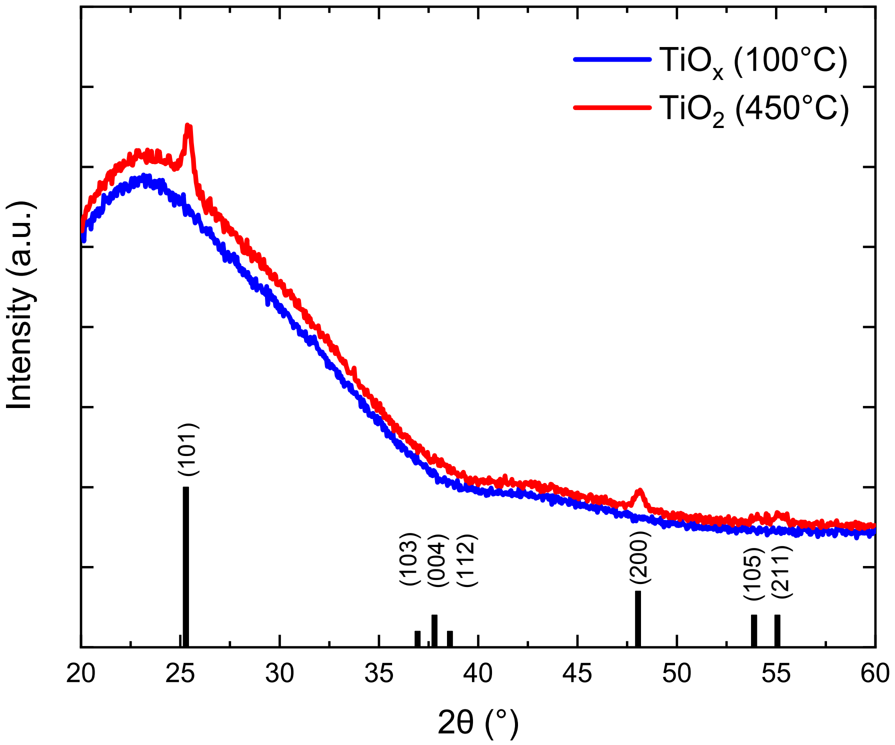

Figure S2: GIXRD of OA-CVD deposited TiO$_x$ films on glass substrate as formed (100°C) and after annealing at 450°C for 30 minutes, compared to a reference spectrum for anatase TiO$_2$ (ICCD PDF-4+ Ref. No. 00-021-1272).

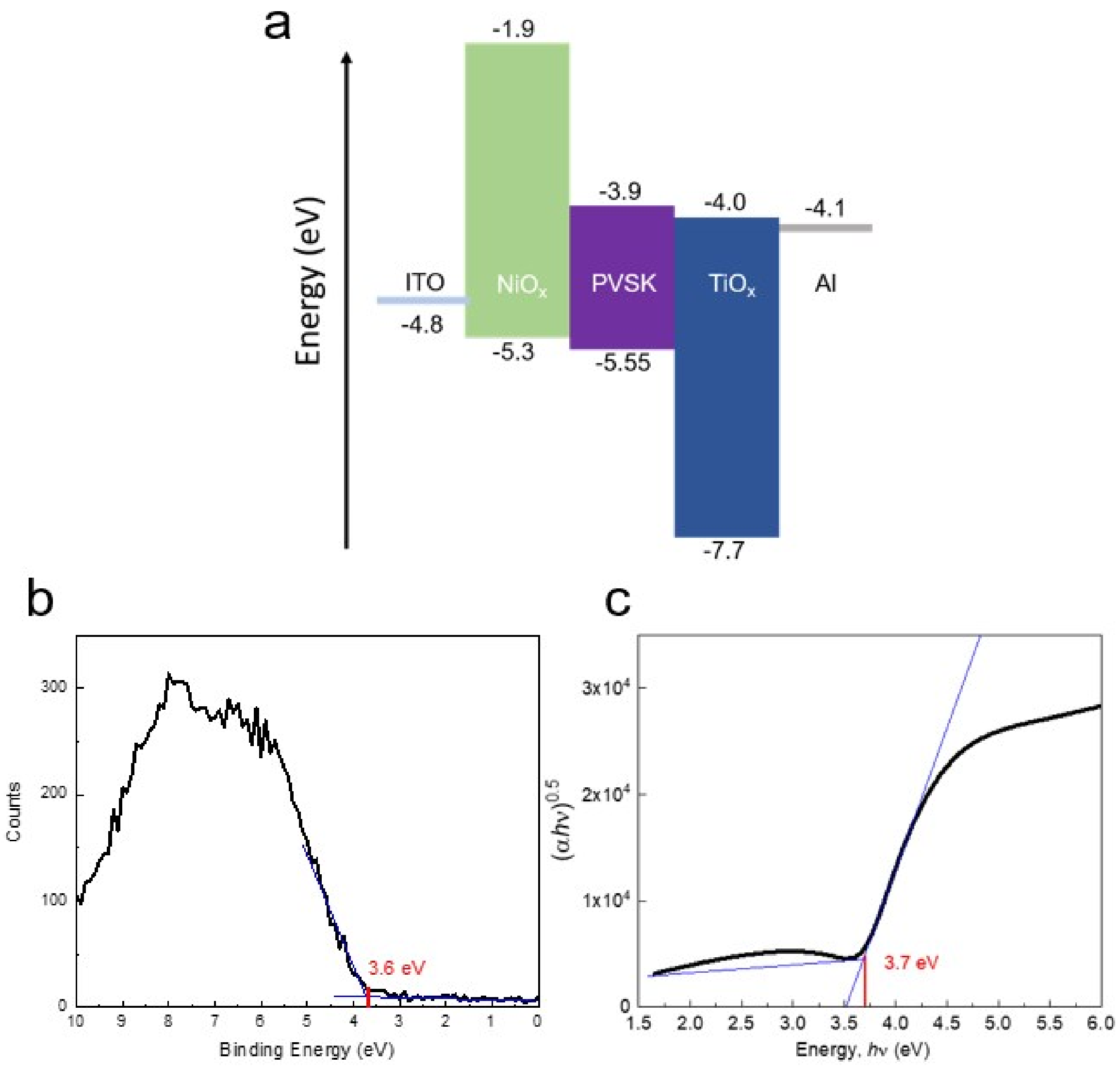

Figure S3: **(a)** Energy band diagram of complete device. **(b)** XPS measurement of TiO$_x$ valence band maximum (VBM) energy difference, E_{Fermi} - E_{VBM}. **(c)** Tauc plot indirect band gap determination of TiO$_x$.

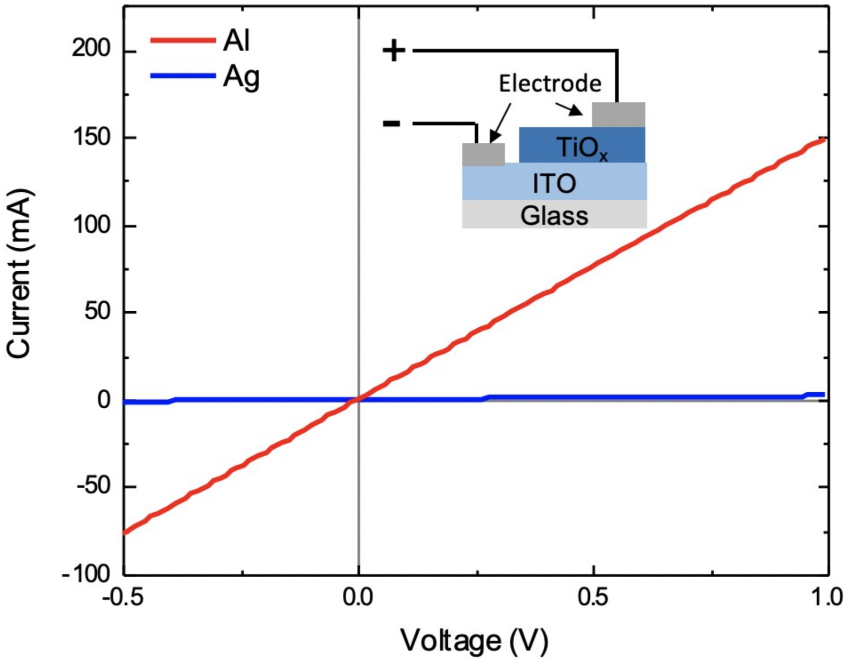

Figure S4: Current-voltage plots of glass/ITO/TiO$_x$ test structure with different metal contacts.

V_{oc} (mV)	J_{sc} (mA·cm^{-2})	Fill Factor (%)	PCE (%)
1030 ± 37 (1080)	19.7 ± 0.8 (19.8)	60 ± 1.7 (62.4)	12.2 ± 0.6 (13.3)

Table S2: Device performance summary for *N* = 15 devices of ITO/NiO$_x$/CsFA/TiO$_x$/Al architecture with optimized 70 nm TiO$_x$ thickness. Champion performance is indicated in parenthesis.

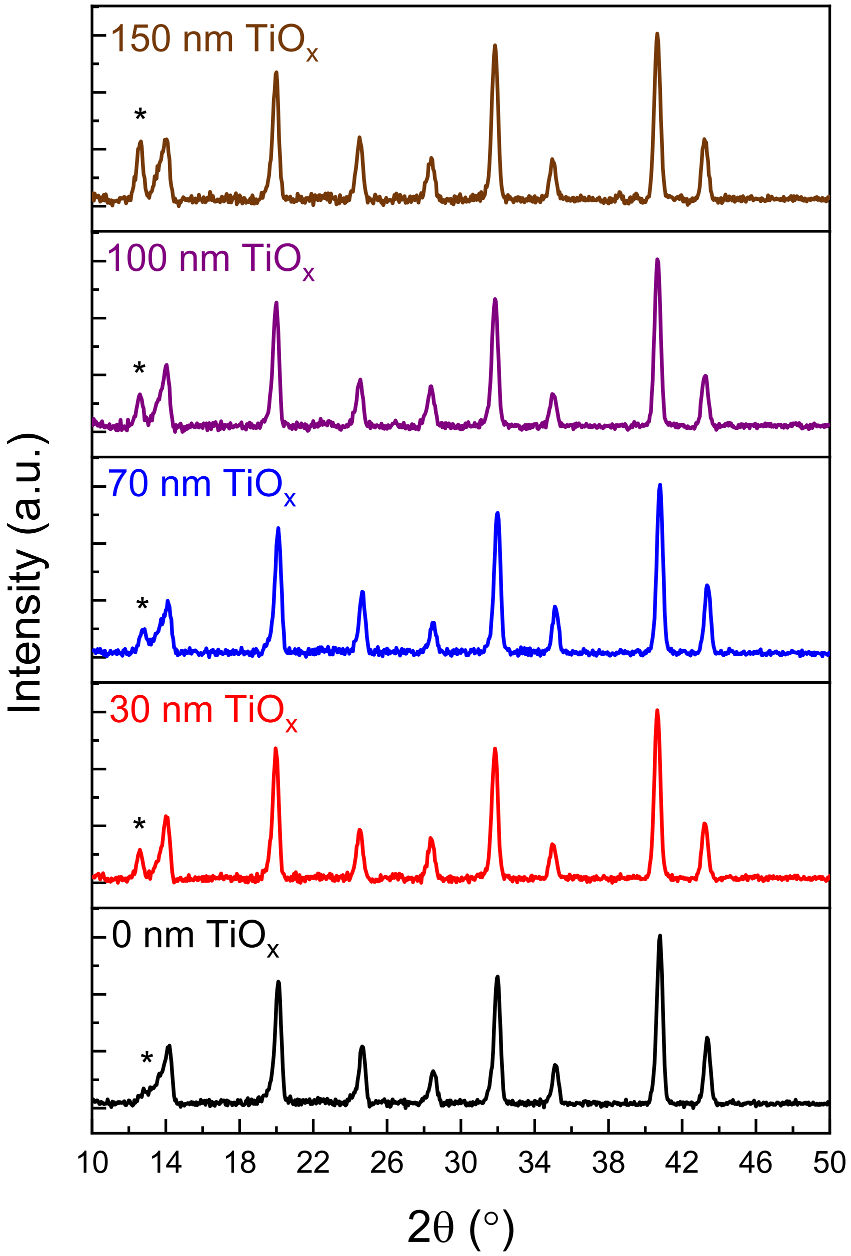

Figure S5: XRD spectra of glass/perovskite/TiO_x test structure with different TiO_x thicknesses. PbI_2 peaks are denoted with *.

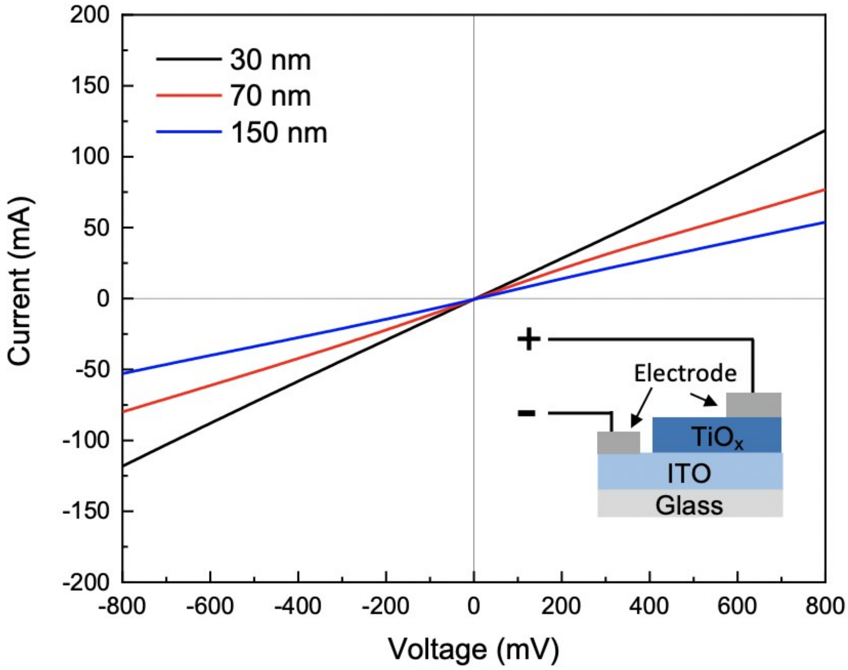

Figure S6: Current-voltage plots of glass/ITO/TiO$_x$/Al test structure with different TiO$_x$ thicknesses.

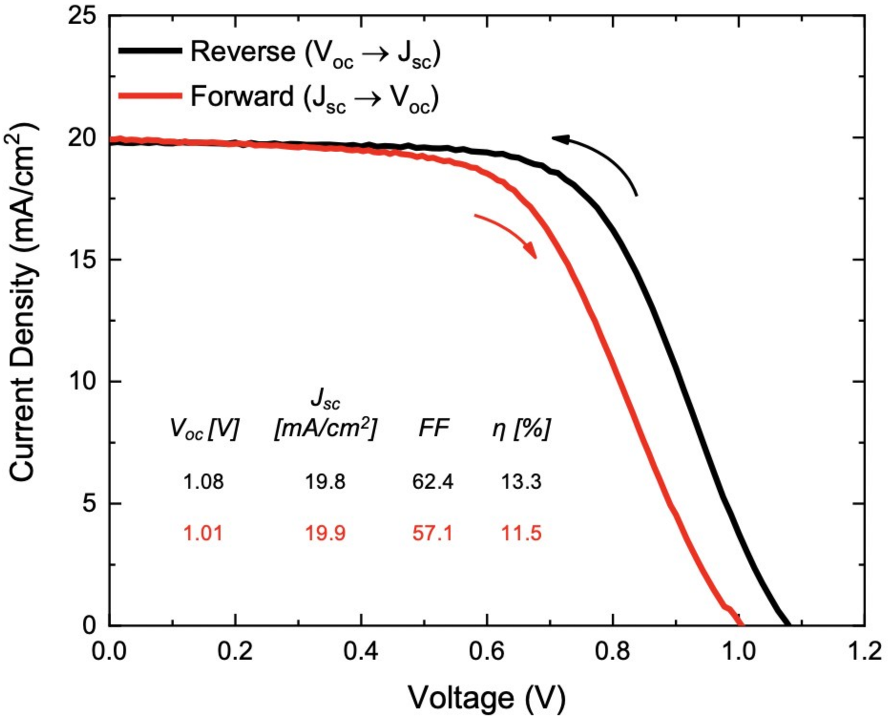

Figure S7: Forward ($J_{sc} \rightarrow V_{oc}$) and reverse ($V_{oc} \rightarrow J_{sc}$) current density vs. voltage (J-V) scans of TiO_x ETL devices on active area of 0.21cm^2.

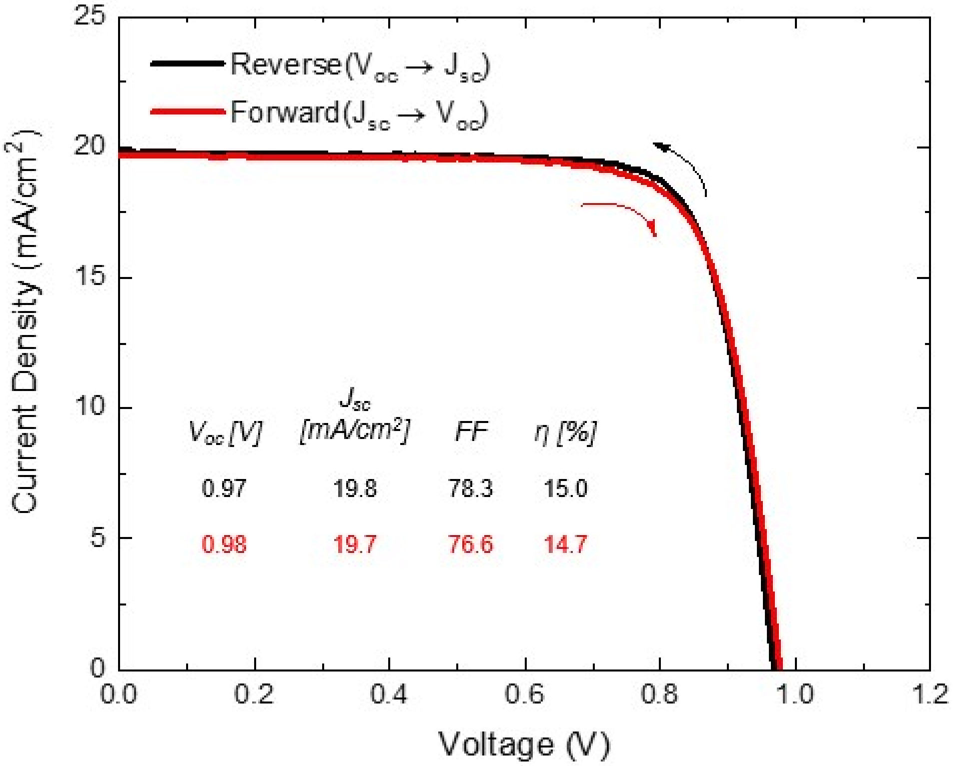

Figure S8: Forward ($J_{sc} \rightarrow V_{oc}$) and reverse ($V_{oc} \rightarrow J_{sc}$) current density vs. voltage (J-V) scans of C_{60}/BCP ETL devices on active area of 0.21cm^2.

Film	Resistivity (Ω-cm)
TiO_X (OA-CVD) 100°C	3.1×10^5
TiO_2 (OA-CVD) 450°C	8.5×10^3
TiO_2 nanoparticles[1]	5.1×10^3
SnO_2 nanoparticles[2]	2.4×10^3

Table S3: Measured resistivities of inorganic ETL films prepared by various methods.

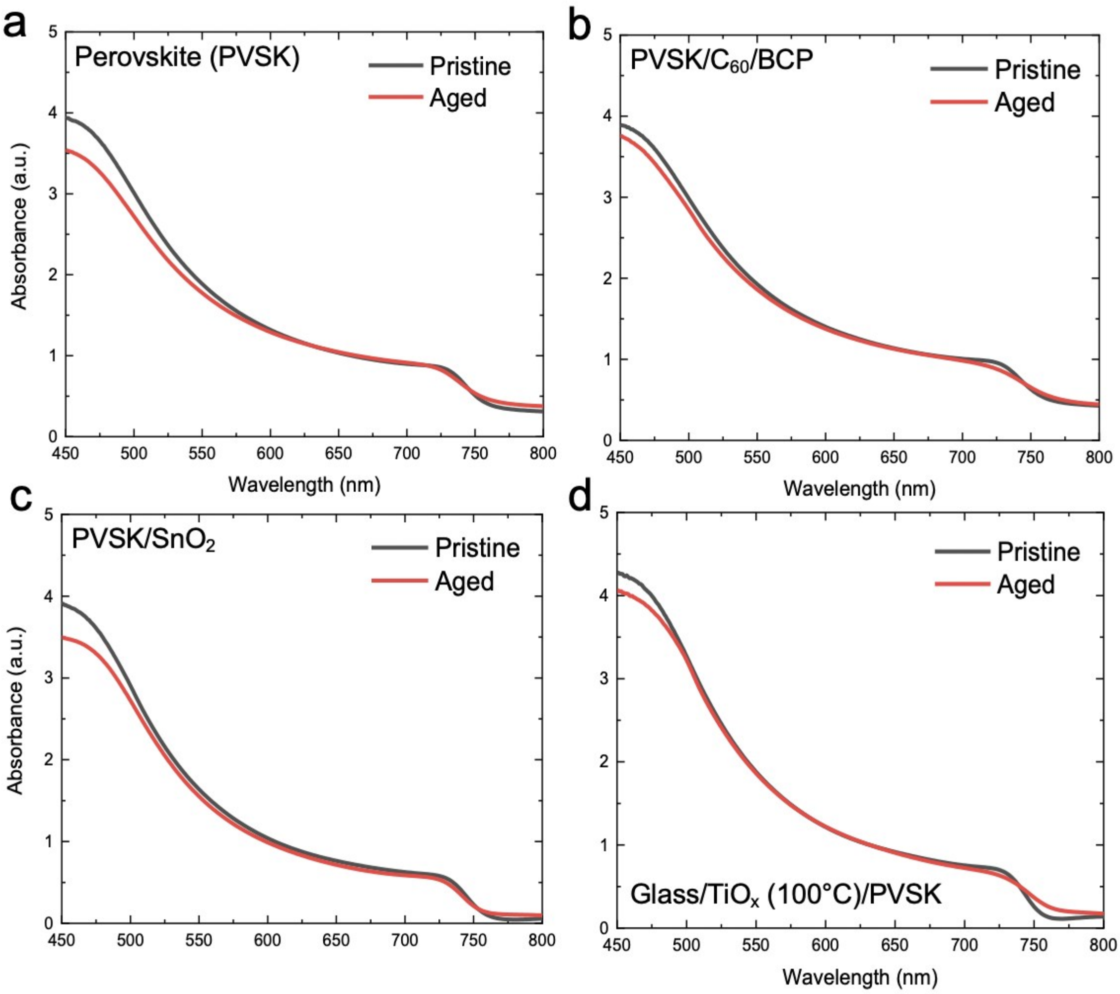

Figure S9: UV-Vis absorption spectroscopy of test structures deposited on glass before and after aging in 1-sun illumination (through glass side) for 24 hours at 40°C in air (45% RH): **(a)** bare perovskite (PVSK) **(b)** PVSK/C_{60}/BCP **(c)** PVSK/SnO_2 **(d)** TiO_x (100°C)/PVSK in NIP structure.

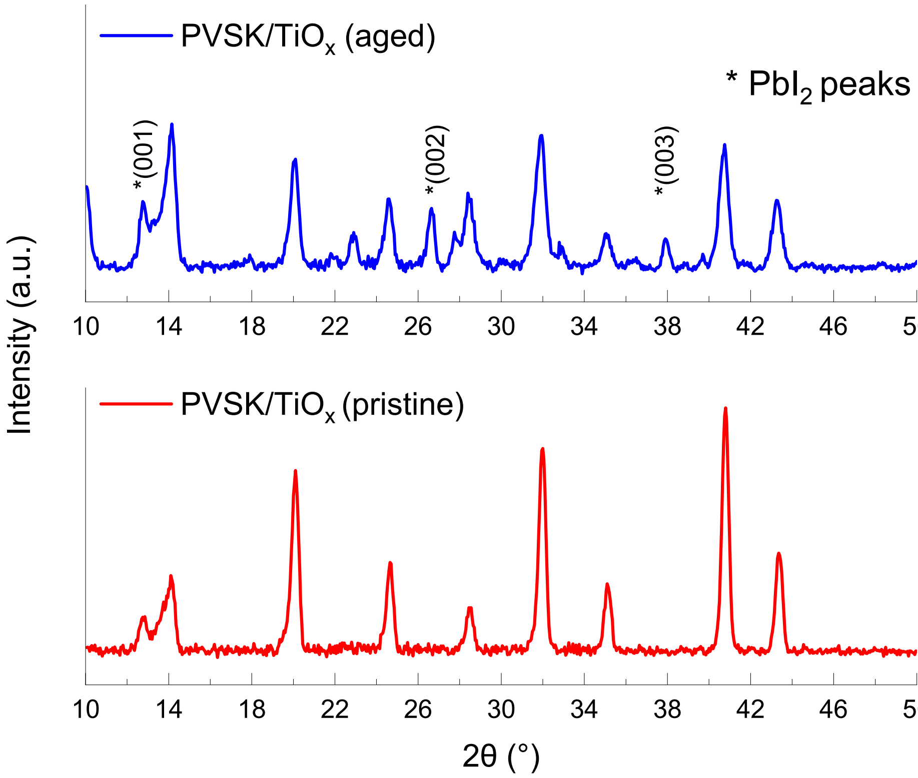

Figure S10: XRD (2θ-ω) measurements of glass/PVSK/TiO$_x$ taken before and after aging in 1-sun illumination (through glass side) for 24 hours at 40°C in air (45% RH). PbI$_2$ indexed reflections are denoted with *.

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References

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