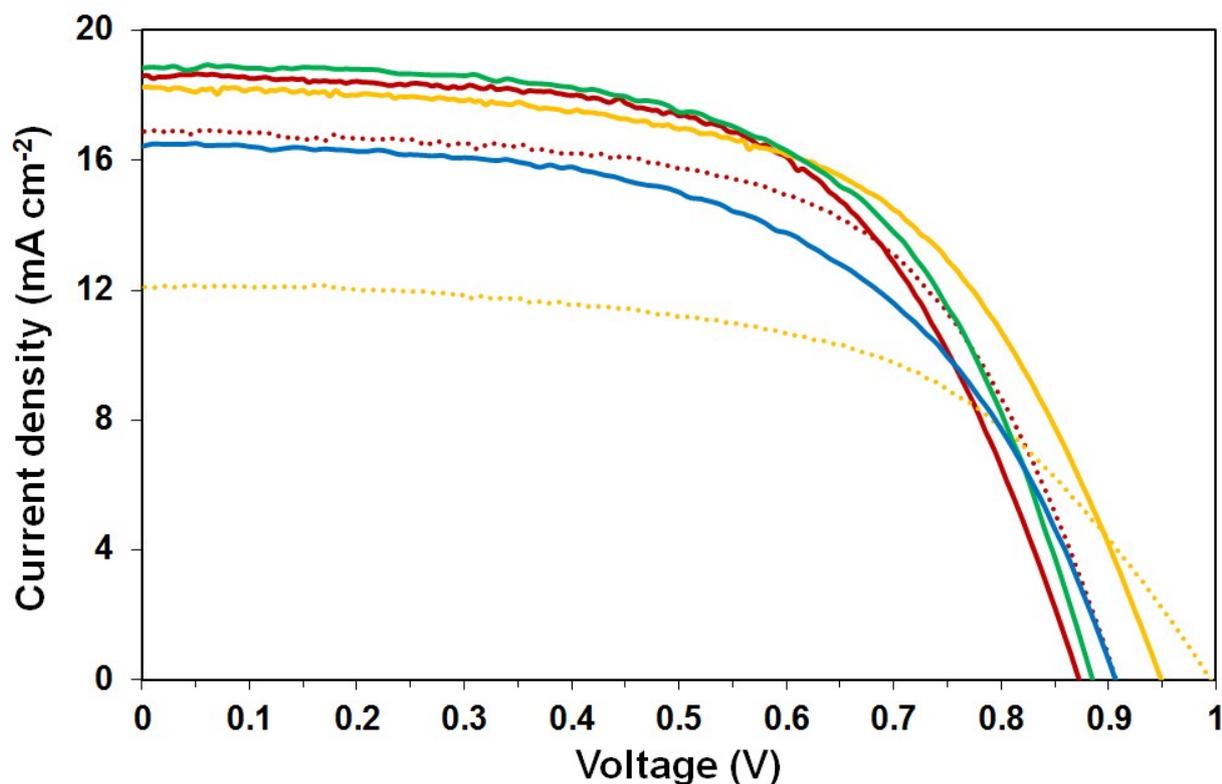


## Nanocolumnar 1-dimesnional TiO<sub>2</sub> photoanodes deposited by PVD-OAD for perovskite solar cells fabrication

F. Javier Ramos<sup>a</sup>, Manuel Oliva-Ramirez<sup>b</sup>, Mohammad Khaja Nazeeruddin<sup>c,d</sup> Michael Grätzel<sup>d</sup>, Agustín R. González-Elipe<sup>b</sup> and Shahzada Ahmad<sup>a\*</sup>



**Figure S1.** Current density-voltage performance of the PV devices employed in the study the different forming perovskite conditions over nanocolumnar 300nm thick TiO<sub>2</sub> photoanodes. [PbI<sub>2</sub>]=1M, concentration of MAI = 10mg/mL at simulated AM1.5G solar irradiation of 99.3mW·cm<sup>-2</sup> (red dotted line). [PbI<sub>2</sub>]=1M, concentration of MAI = 8mg/mL at simulated AM1.5G solar irradiation of 98.1mW cm<sup>-2</sup> (red solid line). [PbI<sub>2</sub>]=1.25M, concentration of MAI = 10mg/mL at simulated AM1.5G solar irradiation of 99.3mW·cm<sup>-2</sup> (yellow dotted line). [PbI<sub>2</sub>]=1.25M, concentration of MAI = 8mg mL<sup>-1</sup> at simulated AM1.5G solar irradiation of 99.6mW·cm<sup>-2</sup> (yellow solid line). [PbI<sub>2</sub>]=1.35M, concentration of MAI = 8mg mL<sup>-1</sup> at simulated AM1.5G solar irradiation of 99.6mW cm<sup>-2</sup> (green solid line). [PbI<sub>2</sub>]=1.45M, concentration of MAI = 8mg/mL at simulated AM1.5G solar irradiation of 98.7mW cm<sup>-2</sup> (blue solid line).

**Table S1.** Photovoltaic performance of the previous works published using 1-dimensional TiO<sub>2</sub> photoanode structures for perovskite-sensitized solar cells.

Ref.	Photoanode	Perovskite	HTM	TiO <sub>2</sub> photoanode thickness (nm)	$J_{SC}$ (mA·cm <sup>-2</sup> )	$V_{OC}$ (mV)	$FF$	$PCE$ (%)
Qiu et al. <sup>[30]</sup>	TiO <sub>2</sub> nanowires	CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	Spiro-	1500	10.67	740	0.54	4.29
		CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3-x</sub> Br <sub>x</sub>	OMeTAD	1500	10.12	820	0.59	4.87
Kim et al. <sup>[31]</sup>	TiO <sub>2</sub> nanorod (rutile)	CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	Spiro-OMeTAD	560	15.6	955	0.63	9.4
				920	12.6	929	0.62	7.3
				1580	11.2	865	0.61	5.9
Dharani et al. <sup>[32]</sup>	TiO <sub>2</sub> nanofiber	CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	Spiro-OMeTAD	413	15.88	980	0.63	9.82
				844	6.41	780	0.66	3.32
				1215	5.14	740	0.66	2.49
Jiang et al. <sup>[33]</sup>	TiO <sub>2</sub> nanowires (rutile)	CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub>	Spiro-OMeTAD	400	18.6	850	0.62	9.7
				600	20.4	780	0.68	10.8
				900	22.3	770	0.68	11.7
				1200	9.3	720	0.71	4.8
Lee et al. <sup>[34]</sup>	TiO <sub>2</sub> nanohelix	HC(NH <sub>2</sub> ) <sub>2</sub> PbI <sub>3</sub>	Spiro-OMeTAD	400	19.88	912	0.66	12.03

**Table S2.** Optimization of the perovskite recipe employed over the 1-dimensional nanocolumnar TiO<sub>2</sub> films to obtain the optimum conditions. The concentration of the PbI<sub>2</sub> and the MAI solutions is tuned and the short-circuit current ( $J_{SC}$ ), open-circuit voltage ( $V_{OC}$ ), the fill factor and the power conversion efficiency were reported at 0.1 sun and 1 sun keeping constant the thickness and evaporation angle of the photoanodes on angle ( $\alpha$ ).

Thickness (nm)	$\alpha$ (°)	[PbI <sub>2</sub> ] (M)	MAI concentration (g mL <sup>-1</sup> )	Sun	$J_{SC}$ (mA·cm <sup>-2</sup> )	$V_{OC}$ (mV)	$FF$	Efficiency (%)
300	70	1	10	0.1	1.69	785.9	0.669	9.13
				1	16.89	907.2	0.604	9.31
300	70	1	8	0.1	1.87	754.4	0.690	9.95
				1	18.59	872.7	0.597	9.88
300	70	1.25	10	0.1	1.27	901.9	0.537	6.37
				1	12.10	994.9	0.569	6.89
300	70	1.25	8	0.1	1.81	830.4	0.671	10.39
				1	18.25	949.0	0.588	10.22
300	70	1.35	8	0.1	1.88	761.2	0.630	9.31
				1	18.84	885.0	0.594	9.95
300	70	1.45	8	0.1	1.68	791.3	0.568	7.77
				1	16.43	906.3	0.558	8.42

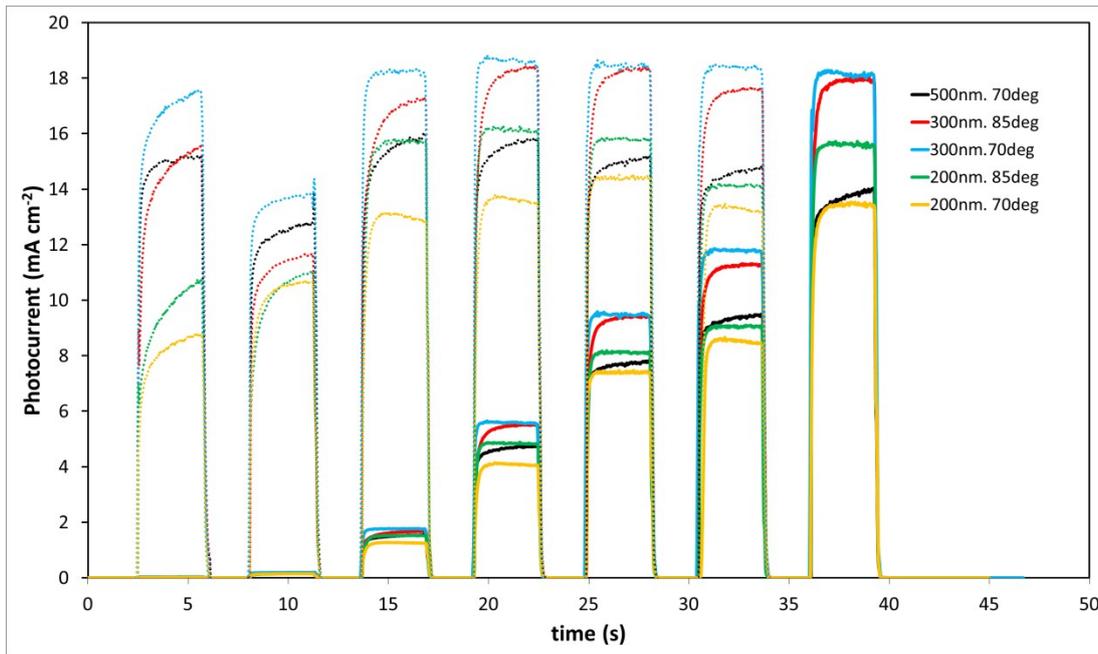
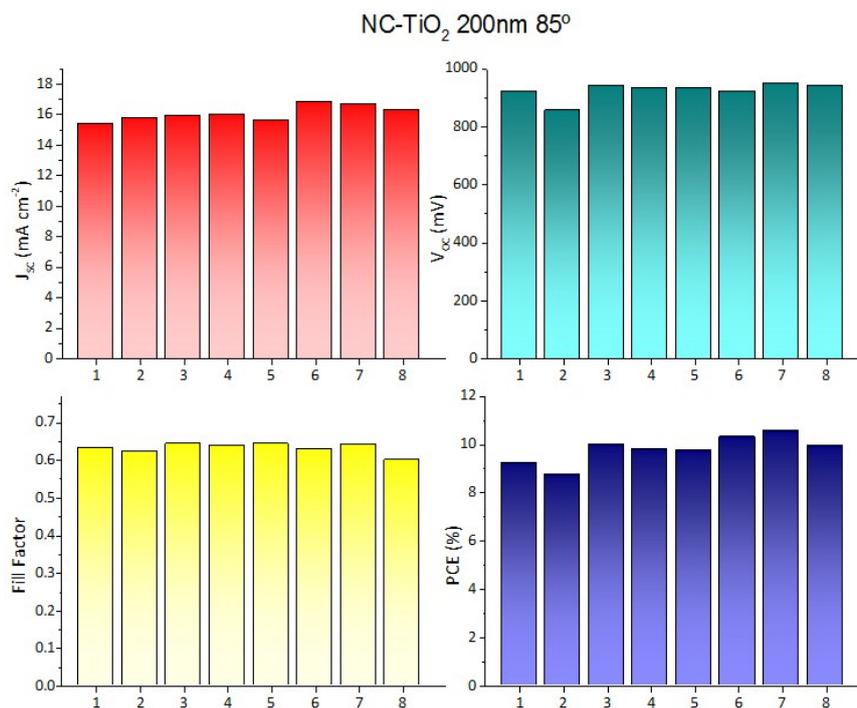


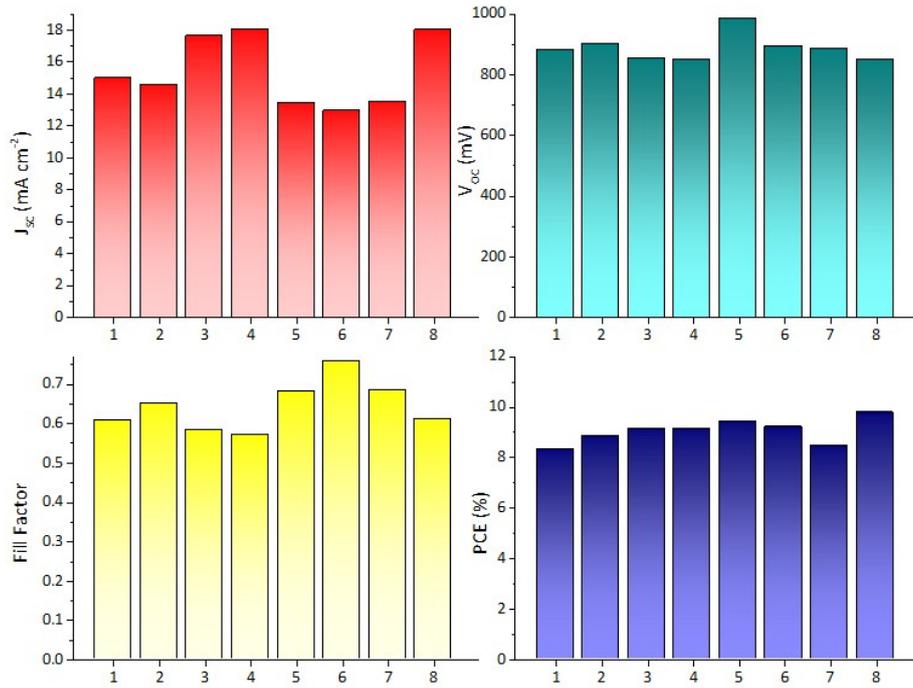
Figure S2: Transient photocurrent experiments for different thickness and porosity of nanocolumnar  $\text{TiO}_2$ . A linear behavior is observed, as the expected photocurrent at full sun (dashed lines) matches quite well with the observed photocurrent at 1 Sun. Here a relatively, slow response was observed in devices made from 500 nm thick NC- $\text{TiO}_2$  (black line) that is attributed due to the higher thickness of photoanode. Similarly for the 300nm ( $\alpha=85^\circ$ ) a marginally exponential increase was observed. Both for 300nm ( $\alpha=70^\circ$ ) and 200nm ( $\alpha=85^\circ$ ) the behavior was much more flat showing a better photo-transient response that is in agreement with the efficient devices reported elsewhere.

### Histograms of the fabricated devices with different thickness and deposition angle (porosity).

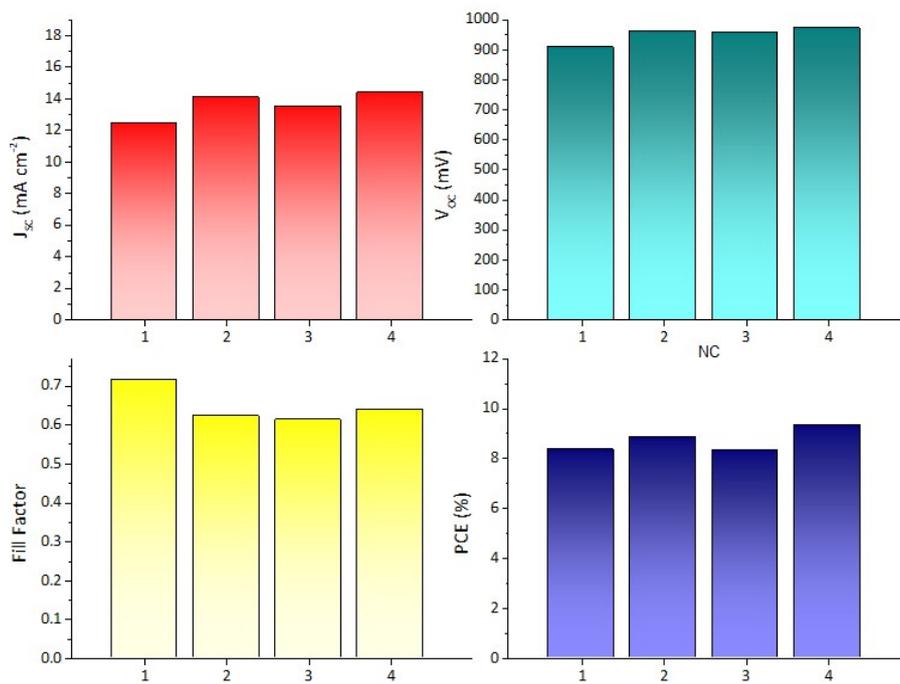
The thickness was varied from 200-500 nm while the deposition angle was switched between  $85^\circ$  and  $70^\circ$ .



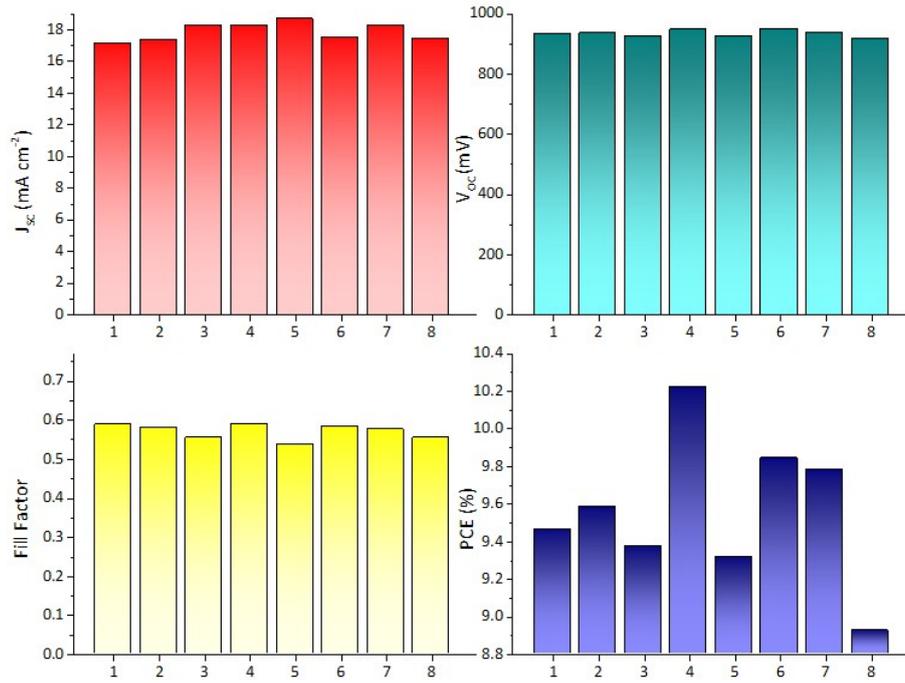
NC-TiO<sub>2</sub> 300nm 85°



NC-TiO<sub>2</sub> 200nm 70°



### NC-TiO<sub>2</sub> 300nm 70°



### NC-TiO<sub>2</sub> 500nm 70°

