Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Mesocrystal TiO_2 films: In-situ Topotactic Transformation and Application in Dye-Sensitised Solar Cells

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Supporting Information

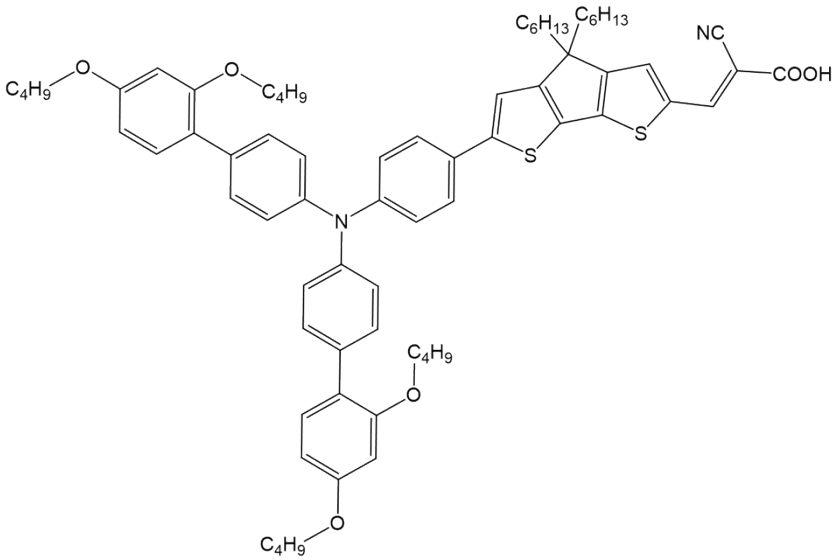

Figure S1: Molecular structure of LEG4.

Table S1: Comparison between NH_4TiOF_3 paste and commercial TiO_2 pastes.

	NH_4TiOF_3 paste	Ti-Nanoxide T/SP	Ti-Nanoxide D/SP	Ti-Nanoxide R/SP
Abbreviation	F	T	D	R
TiO_2 particle size	200~350 nm	15-20 nm	15-20 nm + diffusing particles	>100 nm
TiO_2 concentration	/	~ 18 wt.%	~ 18 wt.%	~ 18 wt.%
Ti concentration	~ 10.8 wt.%	~ 10.8 wt.%	~ 10.8 wt.%	~ 10.8 wt.%

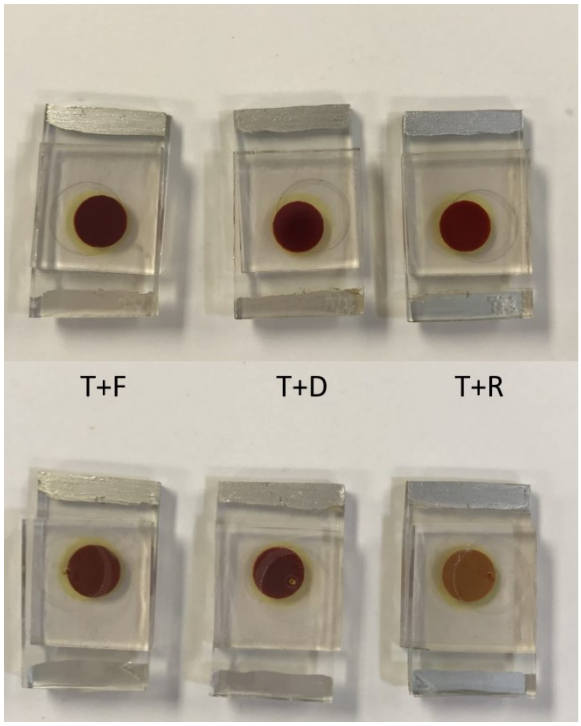

Figure S2 Digital pictures of the example DSSCs from working electrode side (top) and counter electrode side (down). T+F means the photoanode is composed with 1 layer of TiO_2 made from commercial paste Ti-Nanoxide T/SP and 1 layer of TiO_2 made from NH_4TiOF_3 paste. D: Ti-Nanoxide D/SP; R: Ti-Nanoxide R/SP.

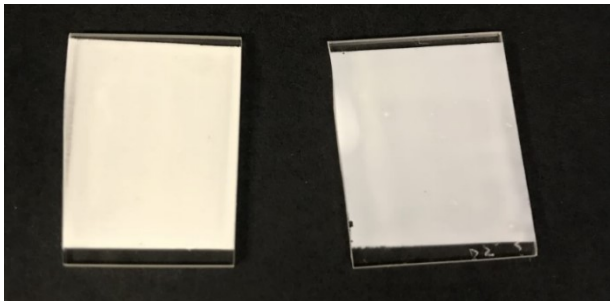

Figure S3 A digital photo of the doctor-bladed films from NH_4TiOF_3 paste (left) or commercial paste Ti-Nanoxide D/SP (right), followed by a sintering process at at 500 °C for 2 hours with a heating rate of 2 °C/min

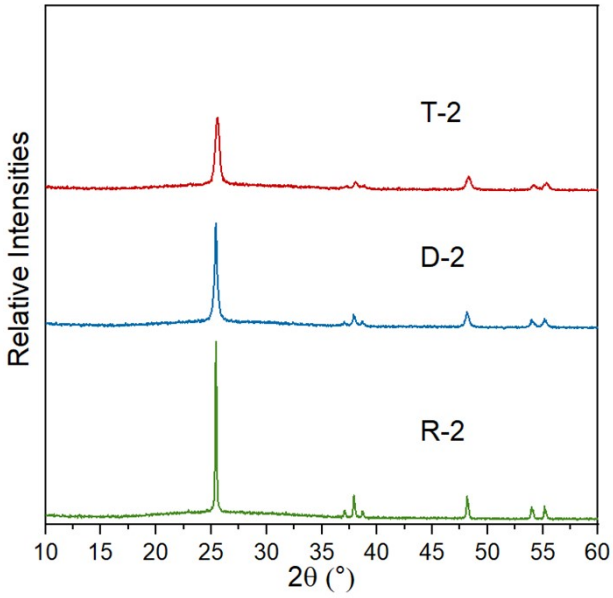

Figure S4: XRD patterns of TiO_2 films from commercial available pastes (Ti-Nanoxide T/SP, Ti-Nanoxide D/SP, Ti-Nanoxide R/SP) sintered at 500 ˚C for 2 hours with a heating rate of 2 ˚C/min.

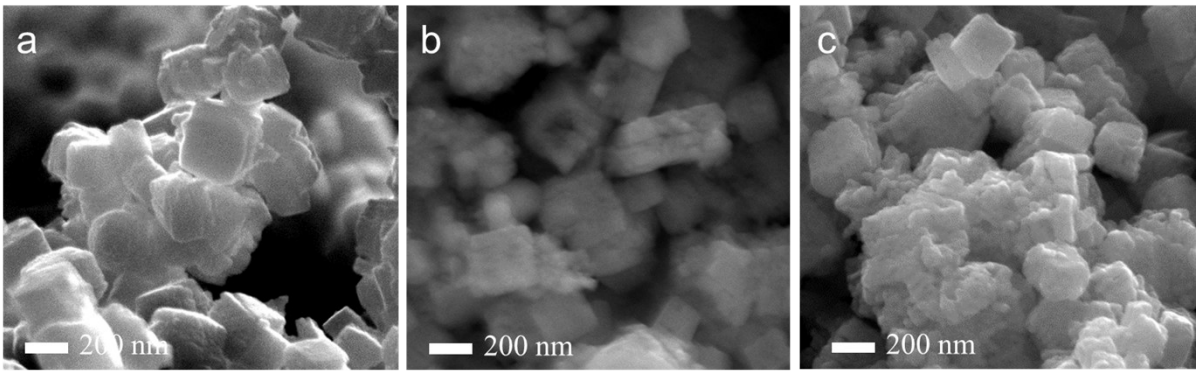

Figure S5: SEM images of $mcTiO_2$ films from NH_4TiOF_3 paste sintered at 500 ˚C for 2 hours with different heating rate, namely (a) 1 ˚C/min, (b) 5 ˚C/min, (c) 10 ˚C/min.

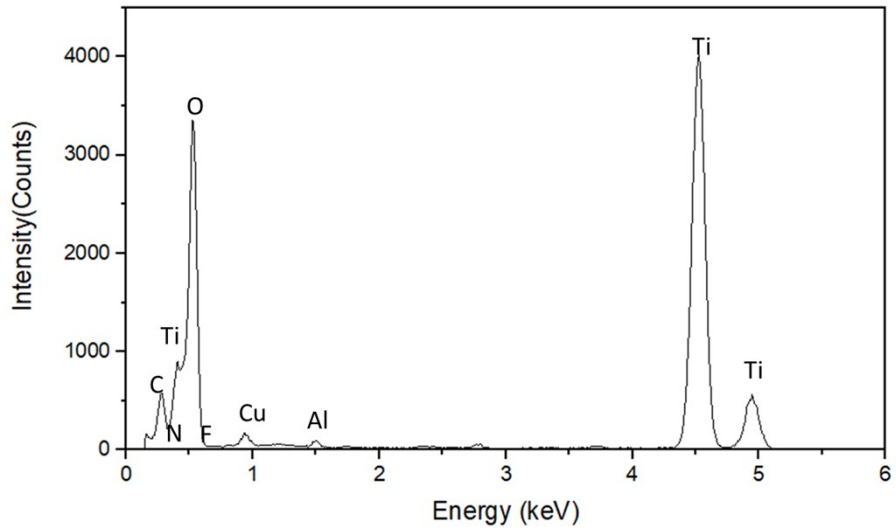

Figure S6: EDS spectrum of mcTiO_2 sintered sintered at 500 °C for 2 hours with a heating rate of 2 °C/min.

Table S2: Elemental composition of mcTiO_2 from TEM-EDS analysis.

Element	Atomic fraction (%)	Mass fraction (%)
N	1.09	0.507
O	54.5	28.9
F	0.0147	0.00928
Ti	44.4	70.6

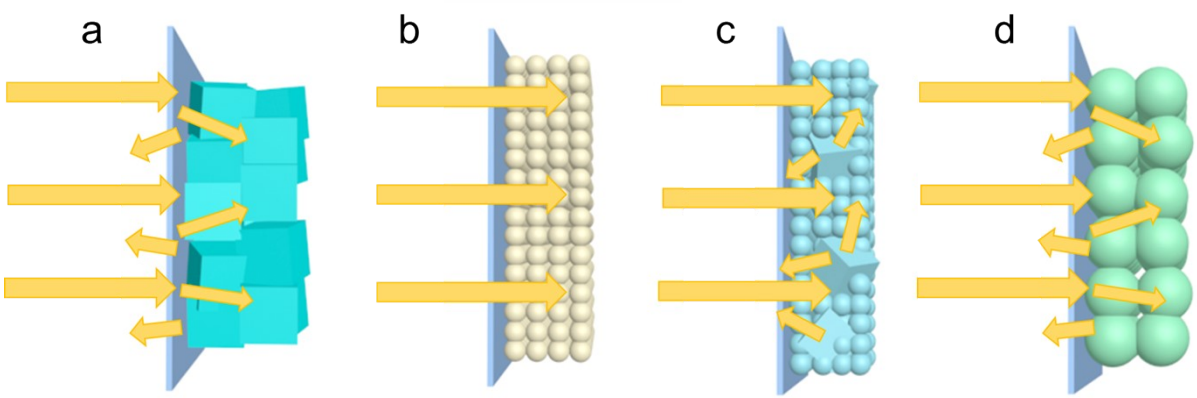

Figure S7: Schematic illustrations of the light path in DSSCs with different TiO_2 as photoanodes. (a) mesocrystal TiO_2 from NH_4TiOF_3 paste, (b) transparent TiO_2 from Ti-Nanoxide T/SP, (c) diffractive TiO_2 from Ti-Nanoxide D/SP, (d) reflecting TiO_2 from Ti-Nanoxide R/SP.

Table S3: Comparison of some EIS parameters between different single-layer photoanodes.

TiO_2	R_s (Ω)	R_{Pt} (Ω)	R_{rec} (Ω)	C_μ (μF)	α	β
	Fitted parameters from Nyquist plot under -0.7 V bias under white LED illumination				Extracted from lnC_μ v.s. bias	Extracted from lnR_{rec} v.s. bias
F	12.15	31.84	360.9	126.4	0.40	0.72
T	13.22	44.65	244.0	192.2	0.26	0.66
D	12.58	50.79	525.6	113.3	0.30	0.61
R	12	60.97	2166	40.26	0.18	0.50

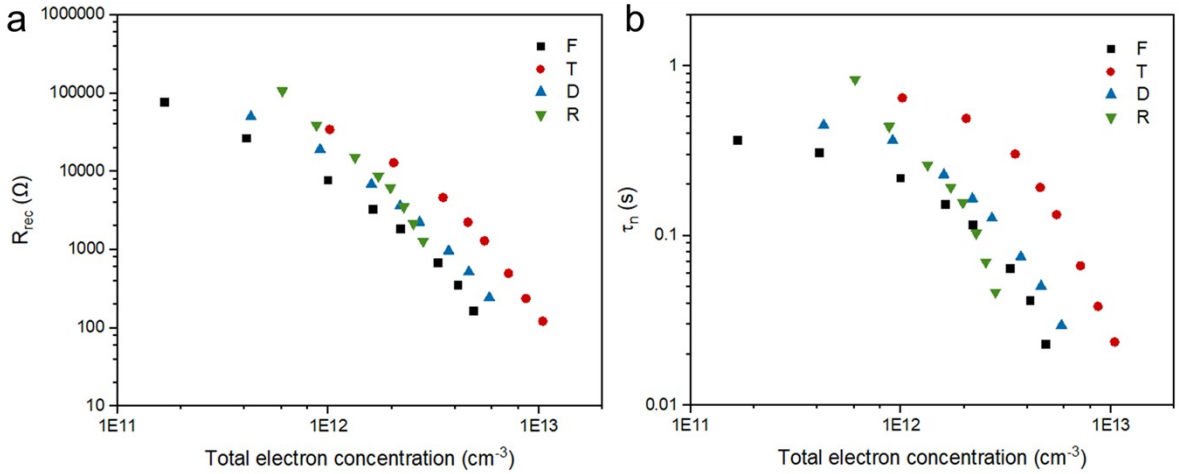

Figure S8: (a) Recombination resistance and (b) electron lifetime of single-layer photoanodes as a function of total electron concentration.

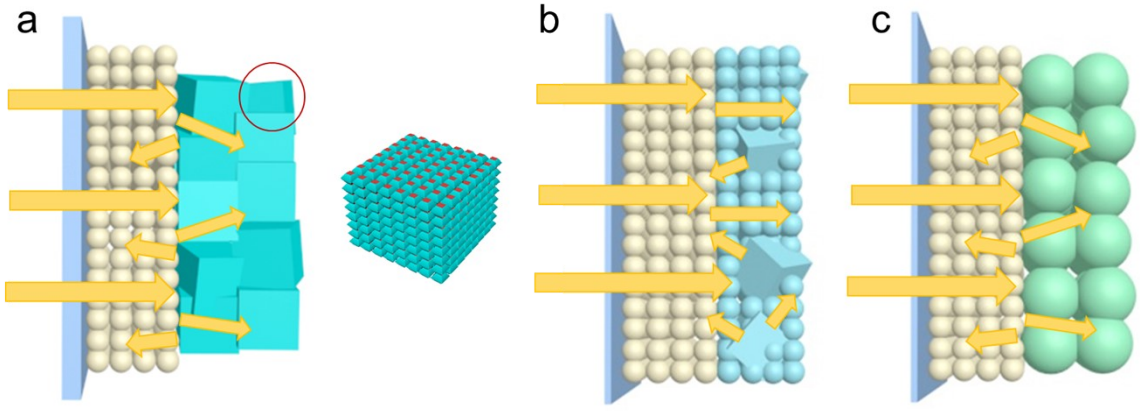

Figure S9: Schematic illustrations of the photoanodes with different scattering layer. (a) mesocrystal TiO_2, (b) diffractive TiO_2, (c) reflecting TiO_2.

Table S4: Comparison of some EIS parameters between different two-layer photoanodes.

TiO_2	α	β
T+F	0.31	0.69
T+D	0.31	0.64
T+R	0.29	0.64

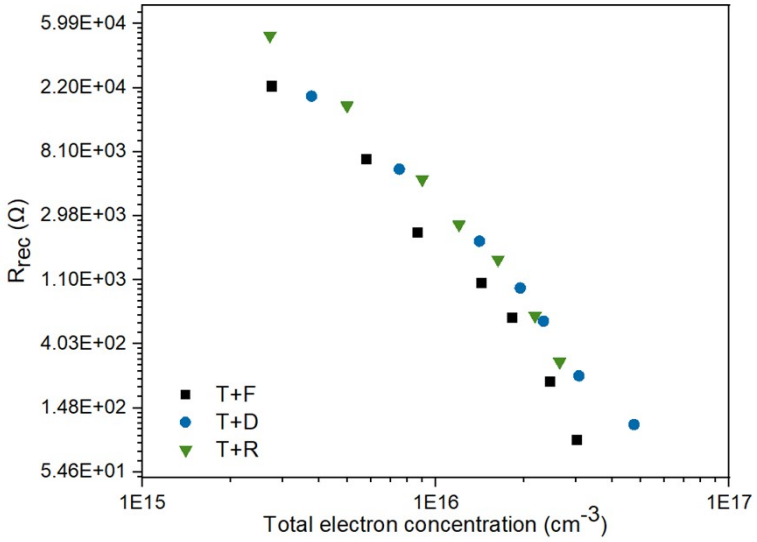

Figure S10: Recombination resistance of two-layer photoanodes as a function of total electron concentration.

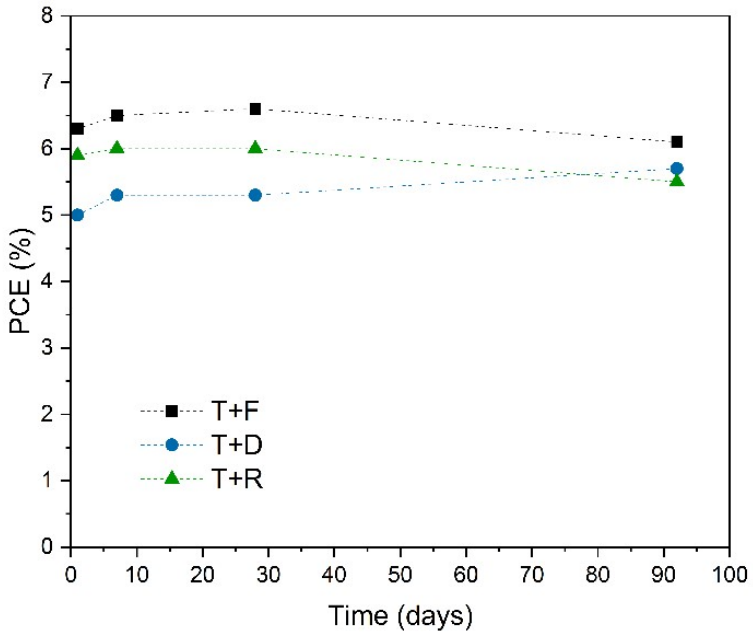

Figure S11 PCE records of DSSCs based on different photoanodes. Devices were kept in dark at the ambient condition.