

Supplementary Information

A novel ball milling technique for room temperature processing of TiO₂ nanoparticles employed as electron transport layer in perovskite solar cells and modules

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Table S1: PV performance of PSCs on different types of TiO₂ ETLs at the reverse and forward scans, measured under AM 1.5G illumination (100 mW/cm²).

Device Structure	Scan direction	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (-)	PCE (%)
S-TiO ₂	Reverse	1.02	20.00	0.60	12.32
	Forward	0.91	19.85	0.56	10.20
S-TiO ₂ /m-TiO ₂	Reverse	1.10	21.48	0.77	18.21
	Forward	1.01	21.49	0.63	13.92
G-TiO ₂	Reverse	1.07	21.72	0.75	17.43
	Forward	1.05	21.63	0.75	17.03

Table S2: The device performance of PSCs (small-area: ≤ 0.1 cm²) based on low-temperature TiO₂ ETL prepared with various deposition methods.

No.	Device Structure (only ETL)	ETL deposition method	ETL processing temperature (°C)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (-)	PCE (%)	Year/ Ref.
1	FTO/TiO ₂ FTO/TiO ₂ + graphene	hydrothermal	150	1.00	17.7	0.61	10.0	2013/
				1.04	21.9	0.73	15.6	[1]
2	ITO/ TiO ₂ ITO/PEIE/ Y:TiO ₂	non-hydrolytic sol-gel	150	1.06	19.9	0.65	13.8	2014/
				1.13	22.7	0.75	19.3	[2]
3	FTO/TiO _x FTO/lt-TiO ₂	non-hydrolytic sol-gel	150	0.96	15.4	0.56	8.3	2014/
				1.05	20.0	0.72	15.3	[3]
4	ITO/TiO ₂ ITO/TiCl- TiO ₂	non-hydrolytic sol-gel	150	1.02	21.0	0.72	15.6	2016/
				1.09	19.7	0.75	16.4	[4]
5	ITO/TiO ₂ ITO/CQDs /TiO ₂	non-hydrolytic sol-gel	150	1.03	19.1	0.77	15.1	2017/
				1.13	21.3	0.78	18.8	[5]
6	ITO/TiO ₂ ITO/TiO ₂ -Cl	non-hydrolytic sol-gel	150	1.12	21.6	0.76	18.5	2017/
				1.18	22.3	0.80	21.4	[6]
7	ITO/G-TiO ₂	ball milling	<150	1.07	21.7	0.75	17.4	[This work]

Table S3: The efficiency of the perovskite solar submodules with various area based on TiO₂ ETL prepared at various temperature.⁷

No.	Device Structure (only ETL)	ETL processing temperature (°C)	Aperture area (cm ²)	Active area (cm ²)	Designated area (cm ²)	PCE (%)	Ref.
1	C-TiO ₂	130	4.00	—	—	13.6	[8]
2	C-TiO ₂ /M-TiO ₂	450	—	49.0	—	10.4	[9]
3	C-TiO ₂	450	—	11.09	—	13.3	[10]
4	C-TiO ₂ /M-TiO ₂	500	—	31.0	—	10.46	[11]
			—	70.0	—	10.74	
5	C-TiO ₂ /Graphene + M-TiO ₂	450	—	50.6	—	12.6	[12]
6	C-TiO ₂ /M-TiO ₂	500	—	8.40	—	6.22	[13]
7	C-TiO ₂ /M-TiO ₂	480	—	8.80	—	9.50	[14]
8	C-TiO ₂ /TiO ₂ NRs	450	—	8.80	—	8.07	[15]
9	C-TiO ₂ /M-TiO ₂	500	36.1	—	—	15.8	[16]
			17.6	—	—	13.9	
			36.1	—	—	12.1(certified)	
10	C-TiO ₂ /M-TiO ₂	480	—	10.1	—	10.3	[17]
			—	100	—	4.30	
11	C-TiO ₂	450	10.36	—	—	15.14	[18]
12	C-TiO ₂	500	—	2.02	—	13.53	[19]
13	G-TiO ₂	<150	—	43.2	25.2	14.19	This work

Note. C-TiO₂: Compact TiO₂ layer; M-TiO₂: Mesoporous TiO₂ layer; G-TiO₂: Ground TiO₂ layer

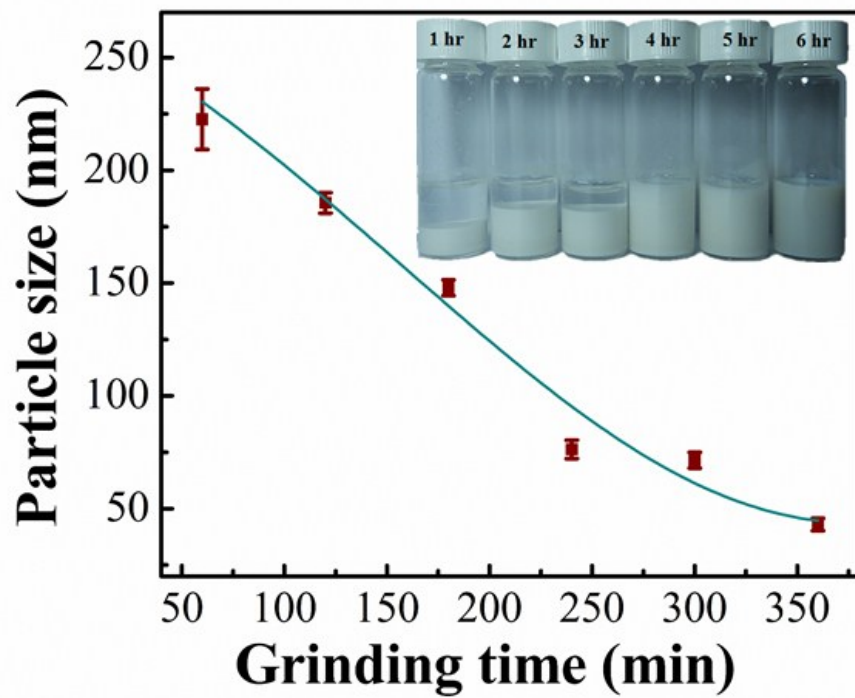


Fig. S1: Average particle size of TiO_2 powder plotted with respect to the grinding time (insert: the pictures of the ground TiO_2 solutions obtained different grinding times).

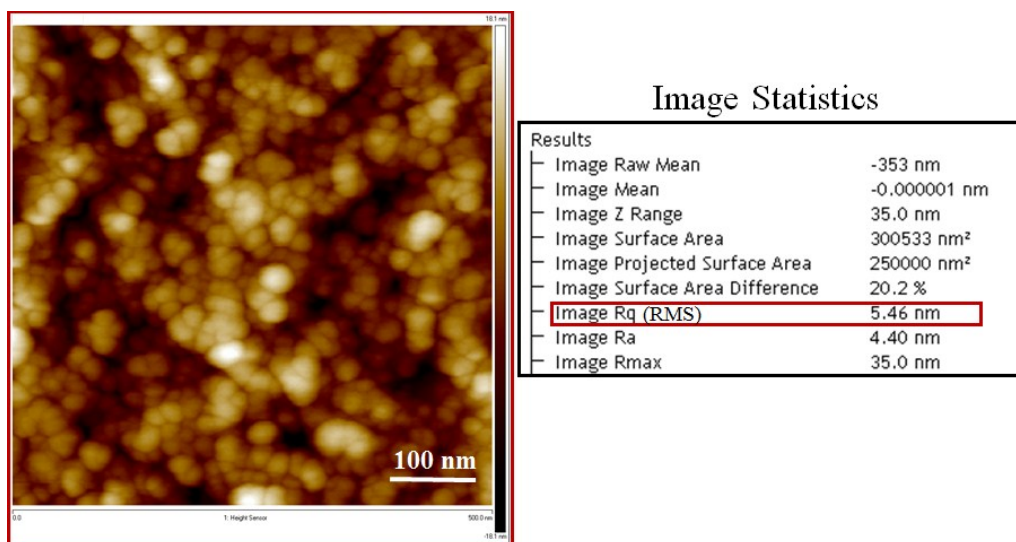


Fig. S2: Atomic force microscopy image of G-TiO₂ film spin-coated on ITO.

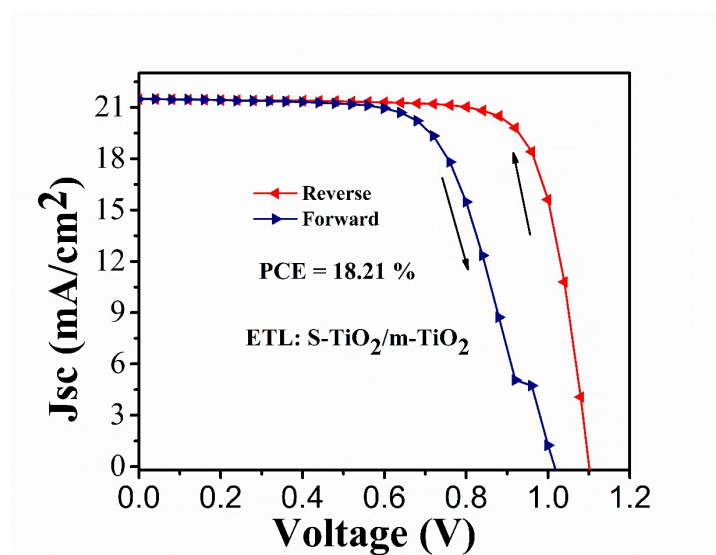


Fig. S3: *J-V* curve of PSCs with the device structure FTO/S-TiO₂/m-TiO₂/CH₃NH₃PbI₃/spiro-MeOTAD/MoO₃/Ag measured at reverse and forward scans.

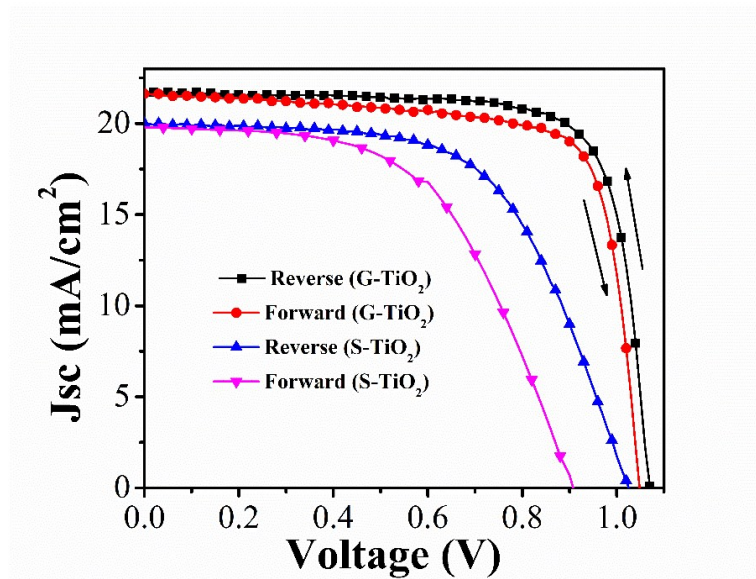


Fig. S4: J - V curves of PSCs with the G-TiO₂ and S-TiO₂ as an electron transporting layers measured at reverse and forward scans.

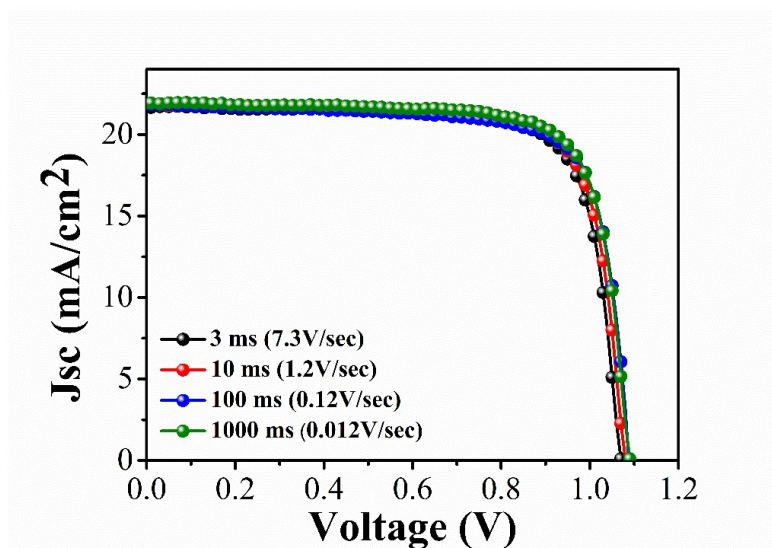


Fig. S5: J - V curves of of the G-TiO₂ based PSCs measured with different delay times.

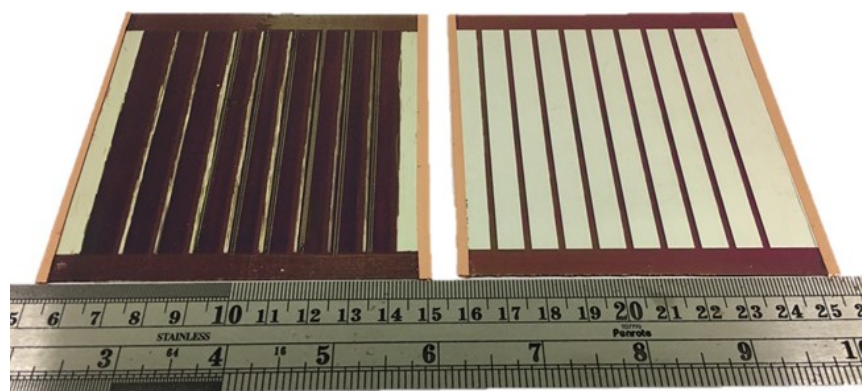


Fig. S6: Photograph of large-area (25.2 cm²) solar submodules incorporating G-TiO₂ as the ETL.

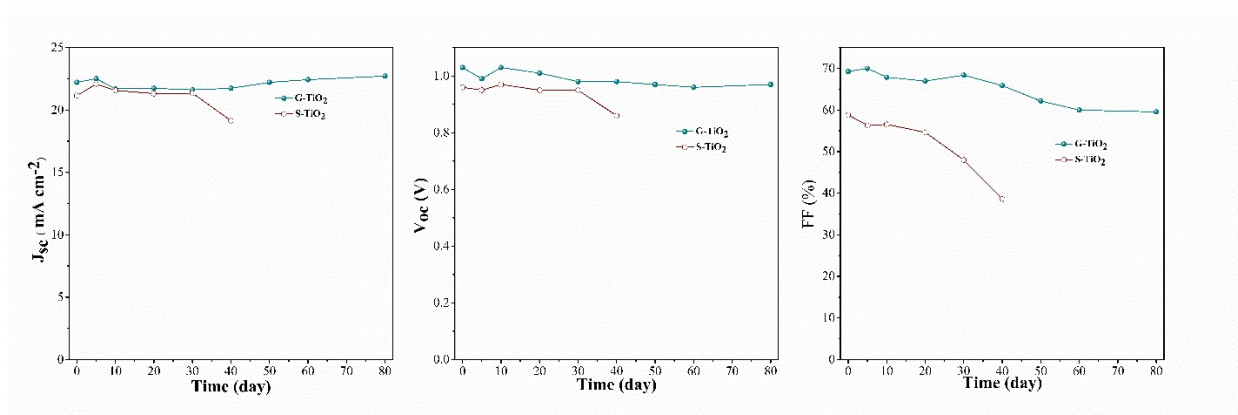


Fig. S7: Long-term stability tests of the best cell [ITO (or FTO)/G-TiO₂ (or S-TiO₂) /CH₃NH₃PbI₃/spiro-MeOTAD/MoO₃/Ag] stored in a glove box under N₂.

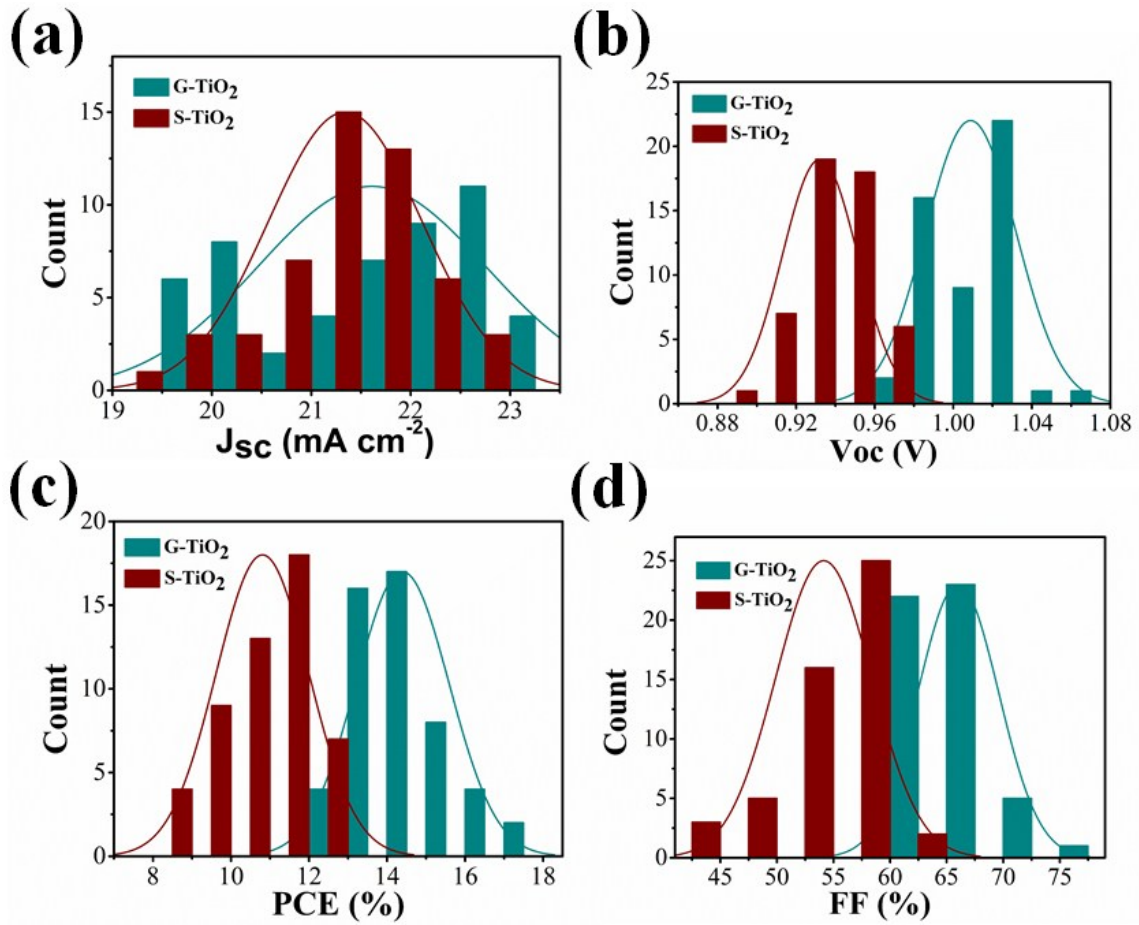


Fig. S8: Histograms of the performance (current-density, open-circuit voltage, power conversion efficiency, and fill factor) of 51 G-TiO₂ and S-TiO₂ devices, measured under 1 Sun light illumination (100 mW cm⁻²).

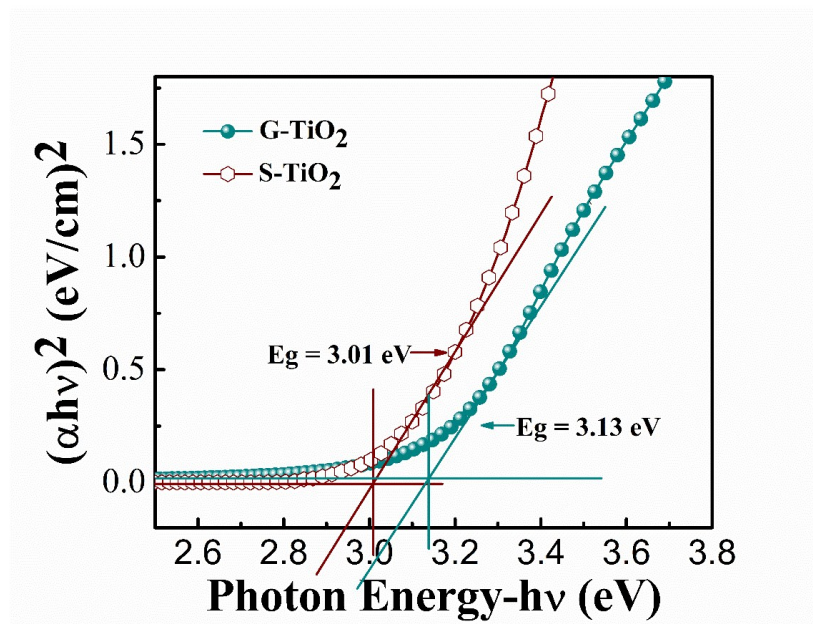


Fig. S9: Tauc plots of G-TiO₂ (dark cyan) and S-TiO₂ (burgundy).

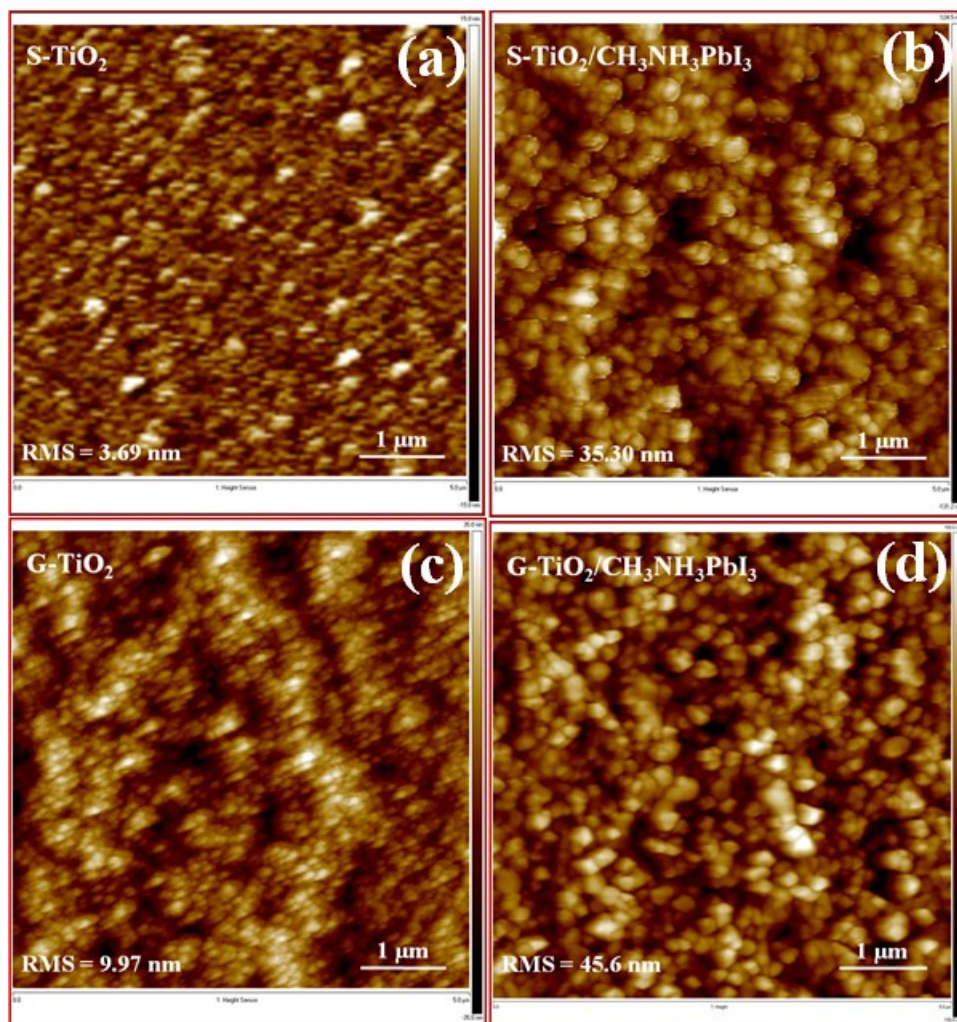


Fig. S10: AFM images of (a) glass/FTO/S-TiO₂, (b) glass/FTO/S-TiO₂/CH₃NH₃PbI₃, (c) glass/ITO/G-TiO₂ and (d) glass/ITO/G-TiO₂/CH₃NH₃PbI₃.

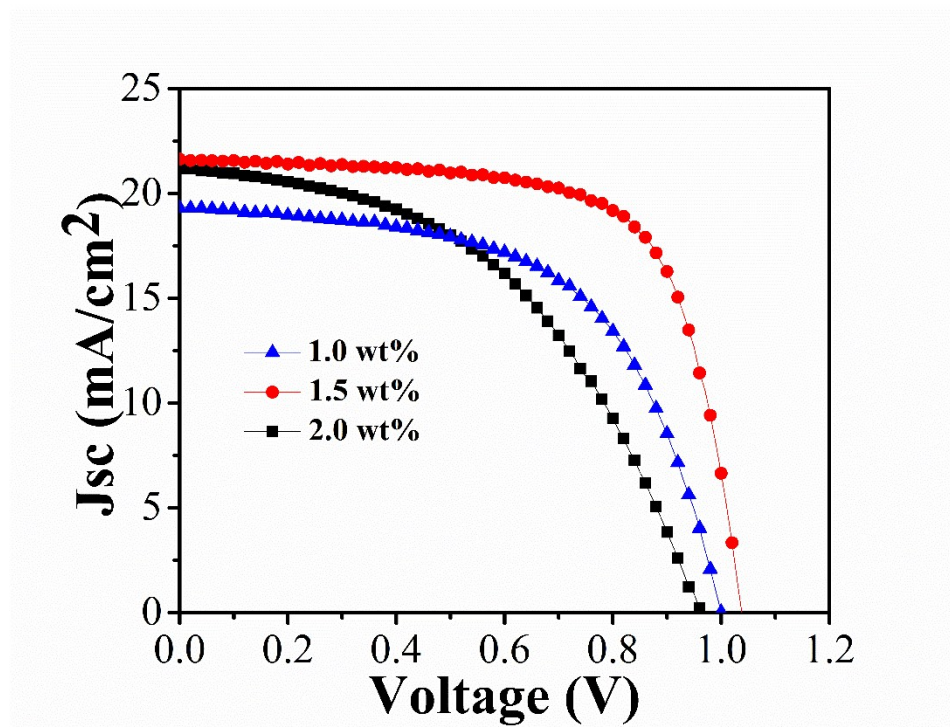


Fig. S11: J - V curves of PSCs based on G-TiO₂ ETL with various thicknesses prepared from different concentrations of G-TiO₂ suspension.

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