

Supporting information

Up-Scaleable Fabrication of SnO₂ with Multifunctional Interface for High Performance Perovskite Solar Modules

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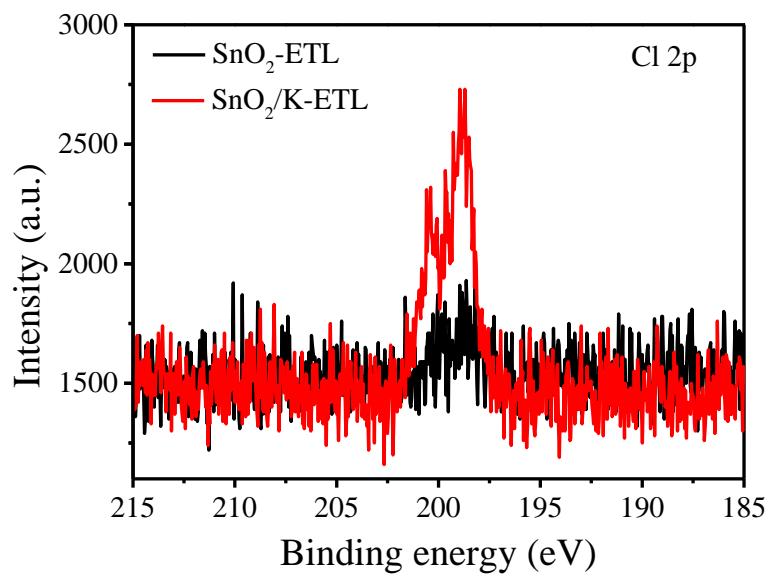


Fig. S2 XPS spectra of the Cl 2p core-level region on SnO₂-ETL and SnO₂/K-ETL films.

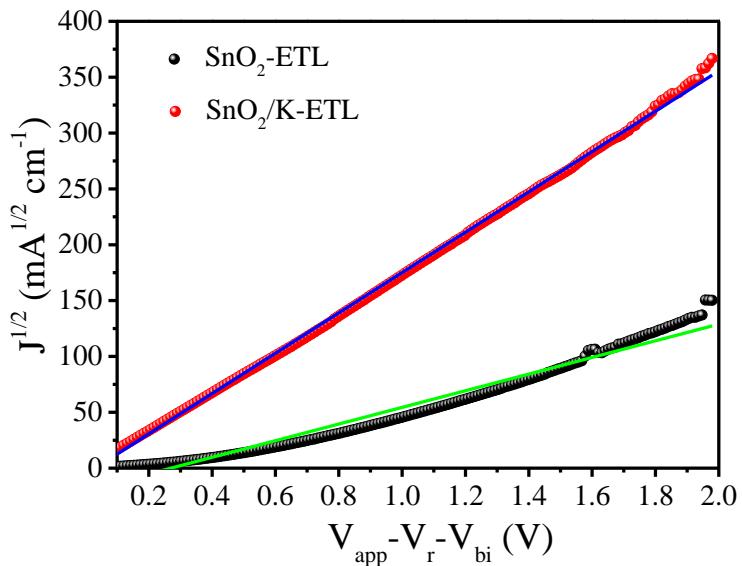


Fig. S3 Dark I–V curves of the SnO_2 films based on the FTO/ SnO_2 /Au structure.

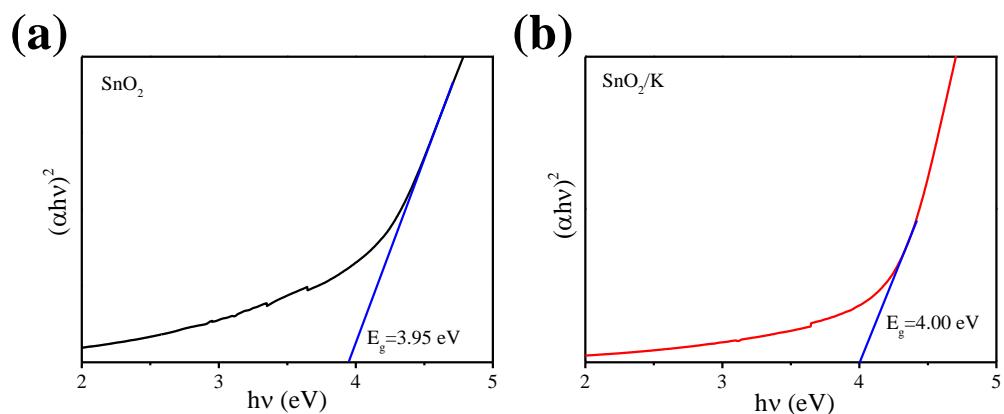


Fig. S4 The relationship of $(\alpha h\nu)^2$ vs energy ($h\nu$) for the SnO_2 -ETL film and SnO_2/K -ETL film.

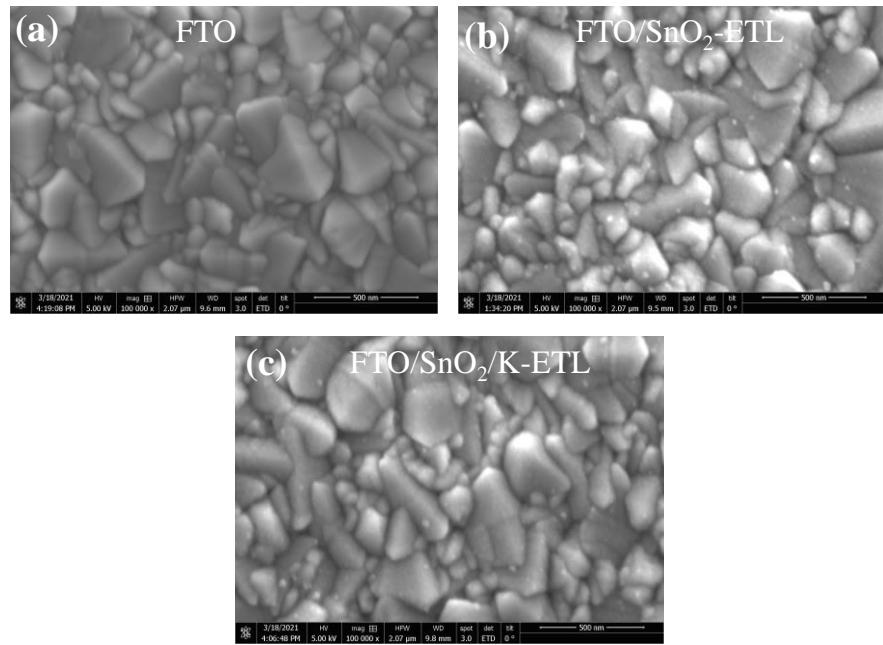


Fig. S5 SEM images of FTO, SnO₂-ETL and SnO₂/K-ETL films.

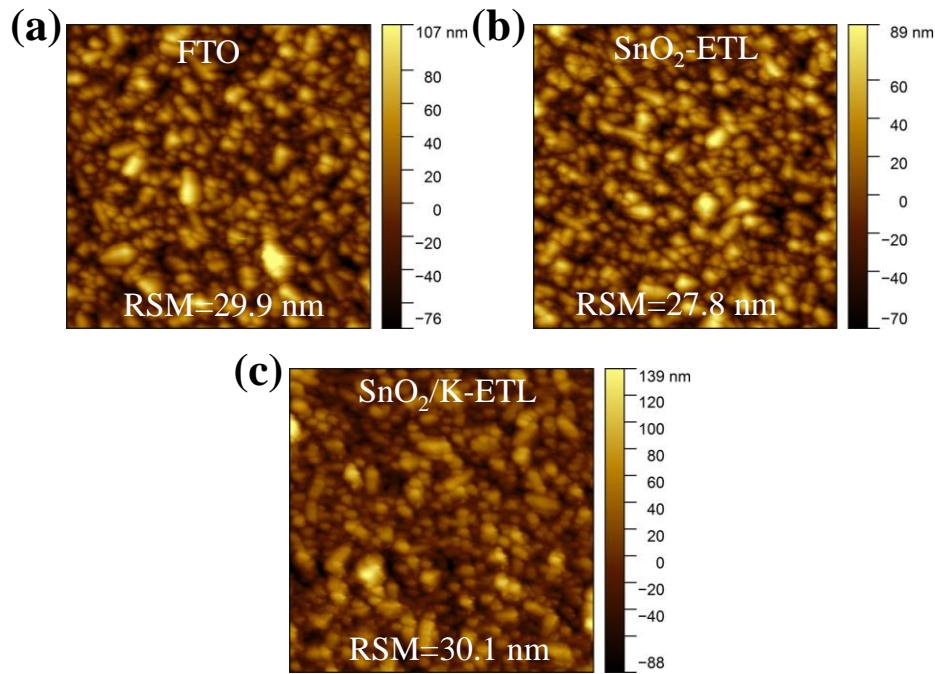


Fig. S6 Tapping-mode AFM images of (a) FTO substrate, (a) SnO₂-ETL film and (b) SnO₂/K-ETL film.

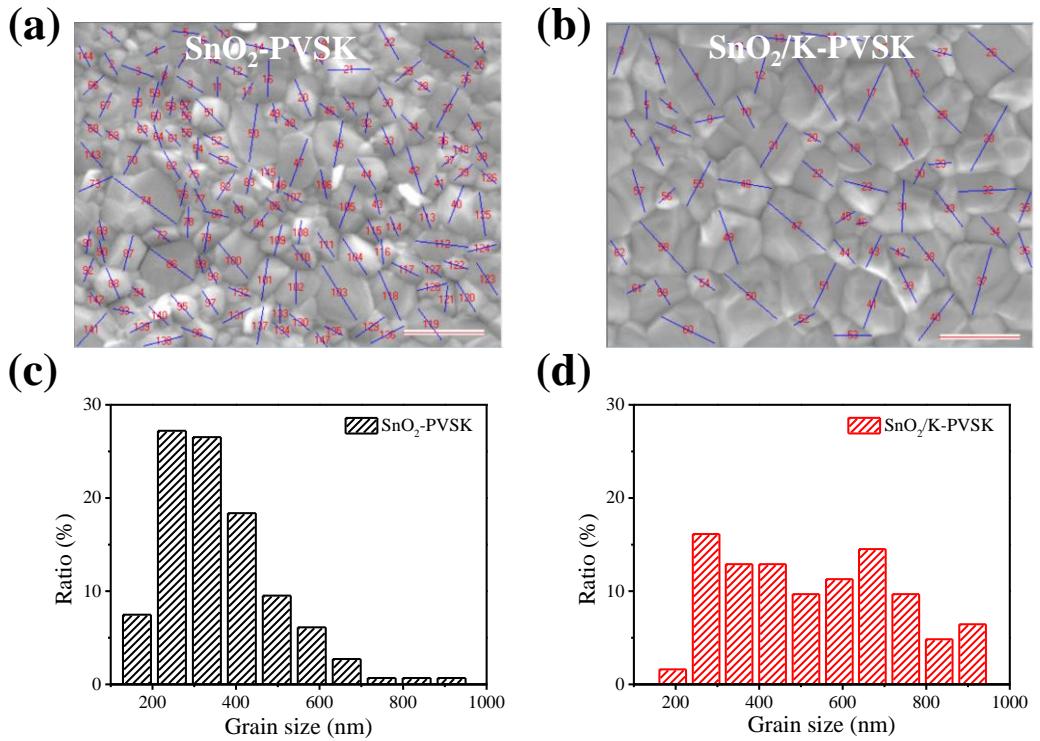


Fig. S7 Grain size distributions of the perovskite films on (a, c) the SnO_2 and (b, d) SnO_2/K substrates. Scale bars are all $1 \mu\text{m}$.

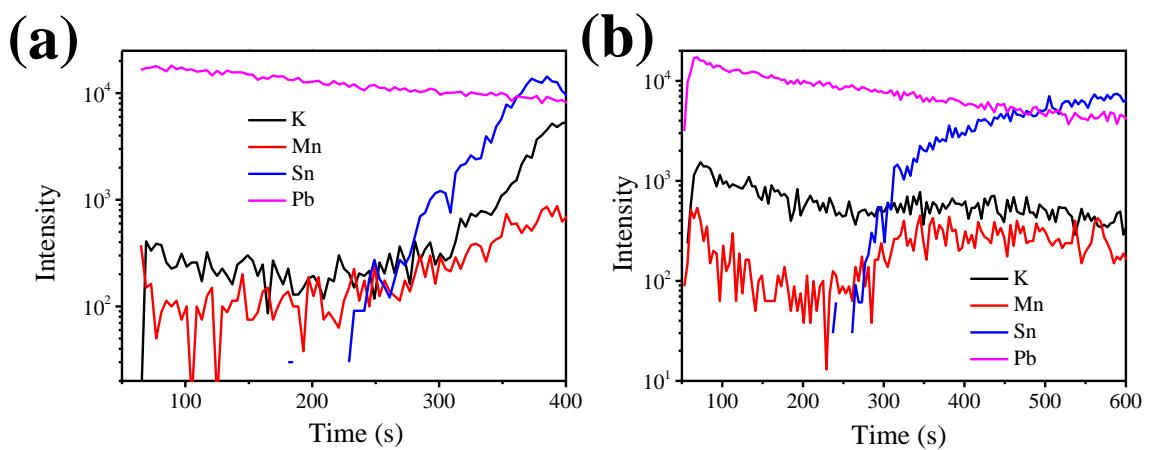


Fig. S8 The distribution of K, Mn, Sn and Pb ions determined from the secondary ion mass spectrometry (SIMS) measurement for the perovskite film (a) without and with annealing.

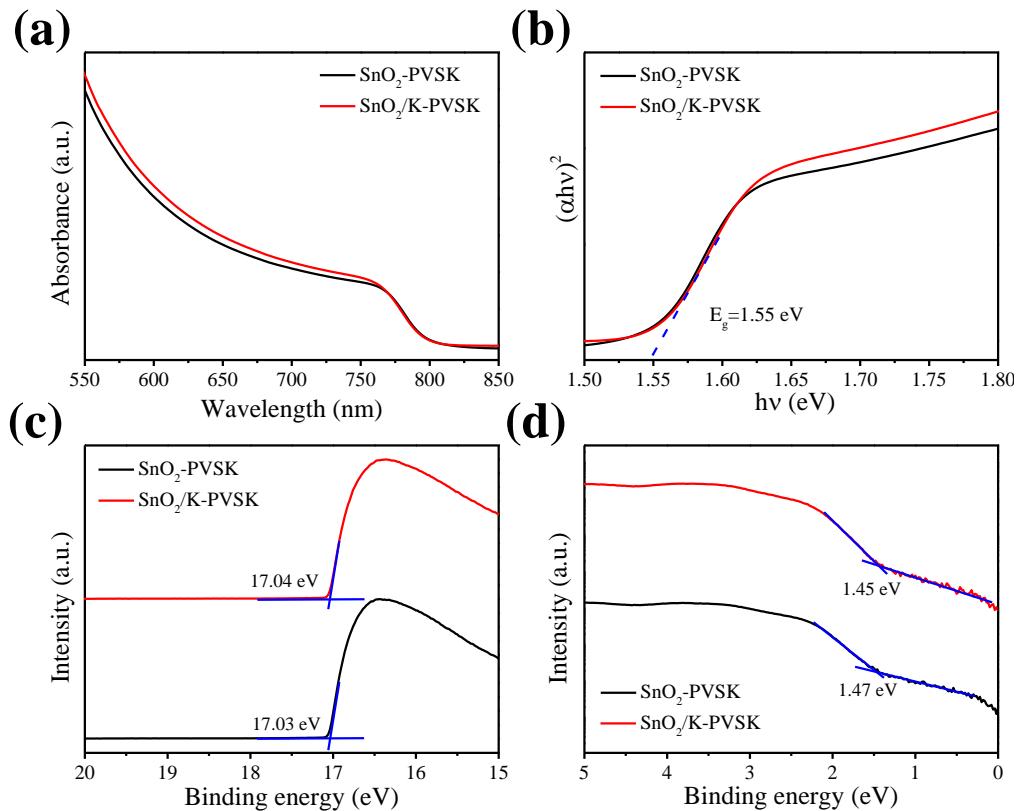


Fig. S9 (a) Absorbance of the perovskite films based on the SnO_2 and SnO_2/K substrates. (b) The corresponding optical bandgap of the perovskite films deduced by the Kubelka-Munk equations. UPS spectra of (c) the valence features, and (d) secondary electron onset of the perovskite films based on the SnO_2 and SnO_2/K substrates.

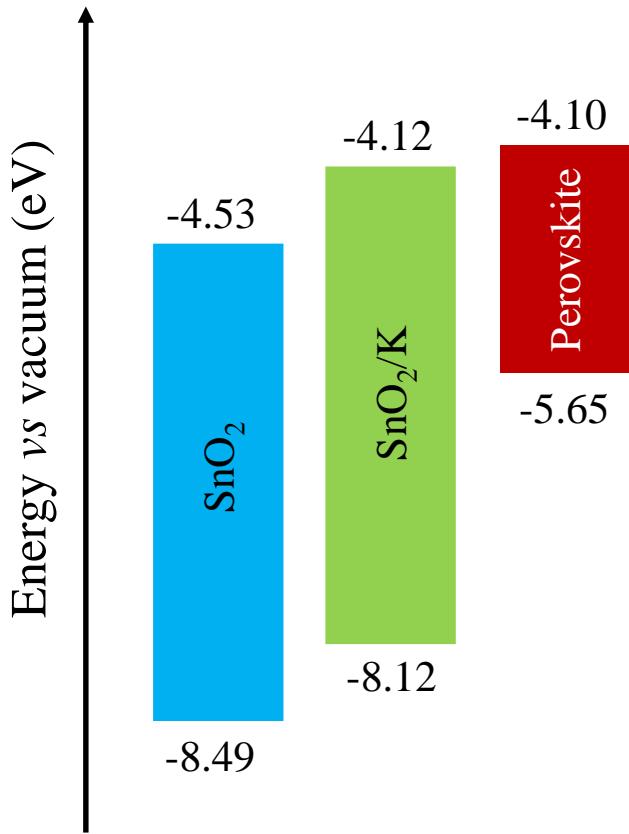


Fig. S10 The energy level diagram for SnO₂-ETL, SnO₂/K-ETL and perovskite.

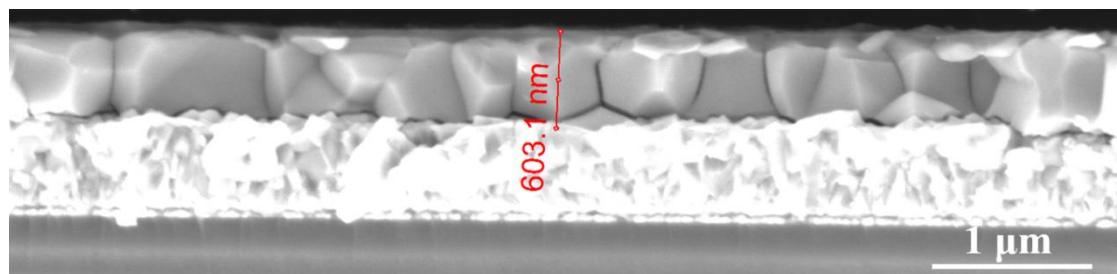


Fig. S11 Thickness of the perovskite film determined by the cross-section SEM image.

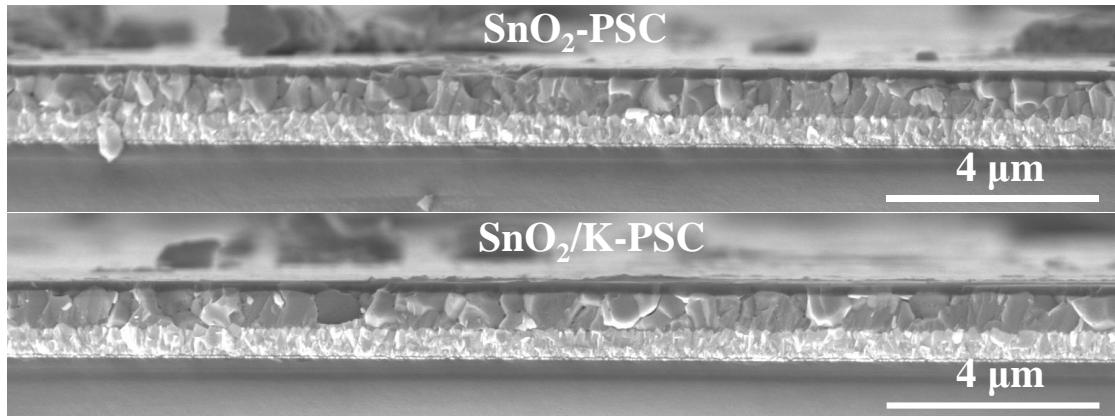


Fig. S12 Low magnification cross-section SEM images of the PSCs based on the SnO_2 with/without KMnO_4 treatment.

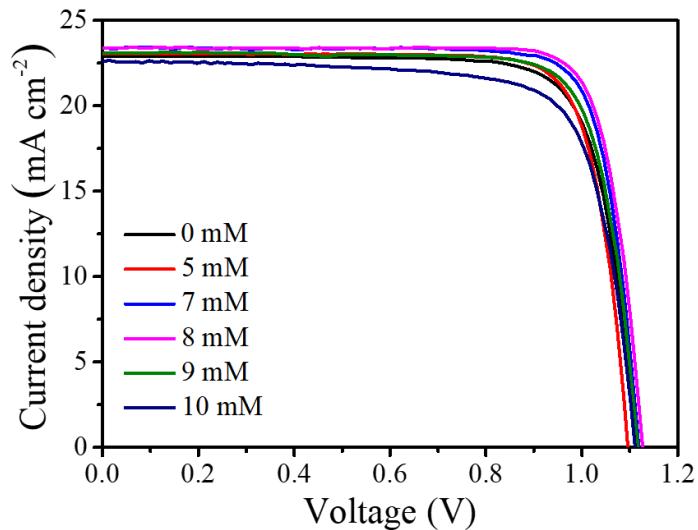


Fig. S13 J-V curves of the SnO_2 based PSCs with different amounts of KMnO_4 .

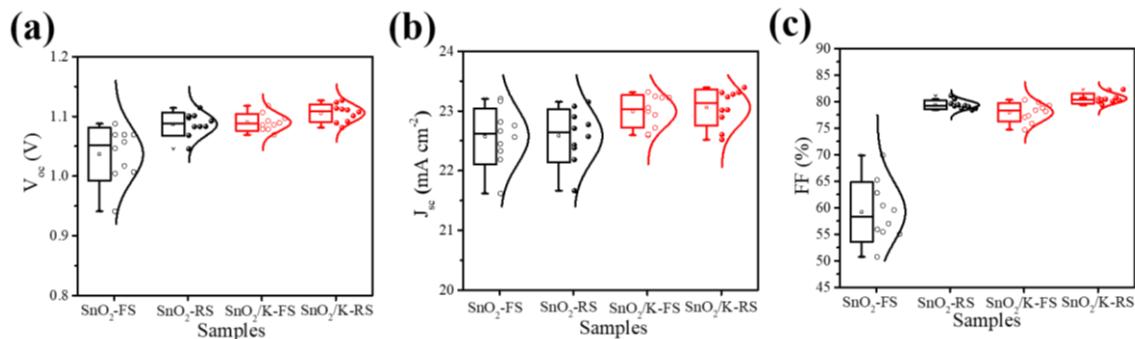


Fig. S14 Statistical distribution of SnO_2 -PSCs and SnO_2/K -PSCs based on 10 devices.
(a) V_{oc} , (b) J_{sc} and (c) FF.

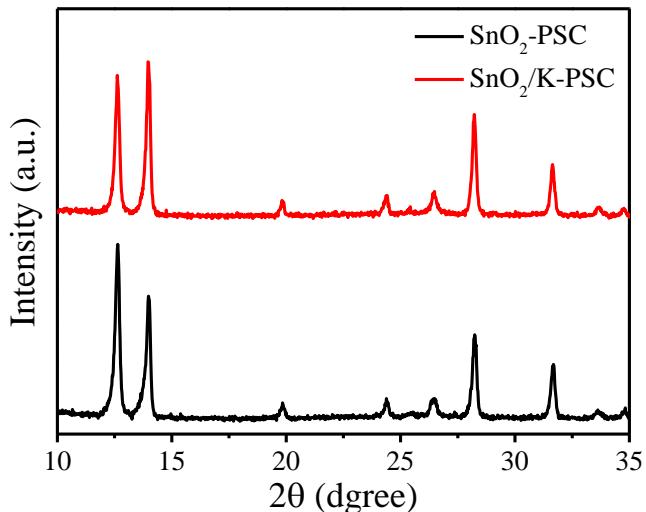


Fig. S15 XRD of SnO_2 -PSC and SnO_2/K -PSCs after storage for 5 months in ambient air in a dry room with a relative humidity of $\sim 20\%$ without encapsulation.

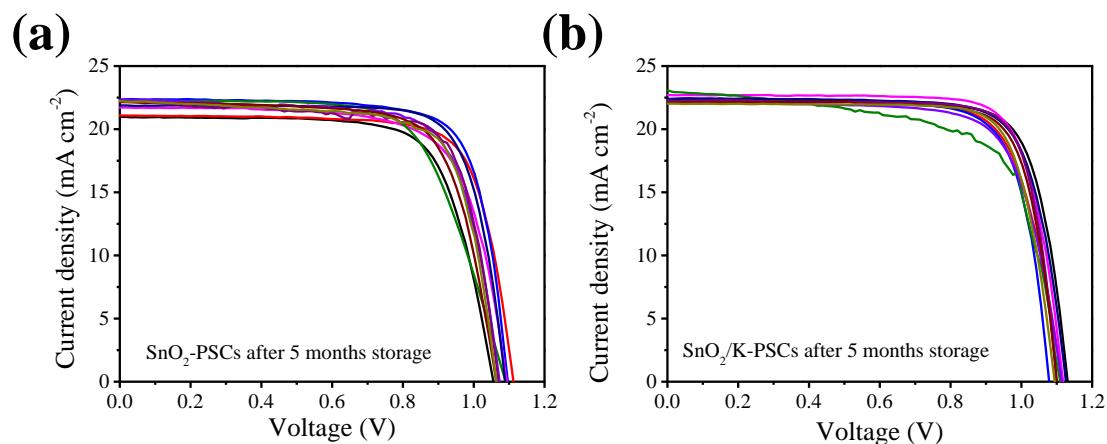


Fig. S16 J-V curves of the SnO_2 -PSCs and SnO_2/K -PSCs based on 10 devices after 5 months storage in ambient air in a dry room with a relative humidity of $\sim 20\%$ without encapsulation.

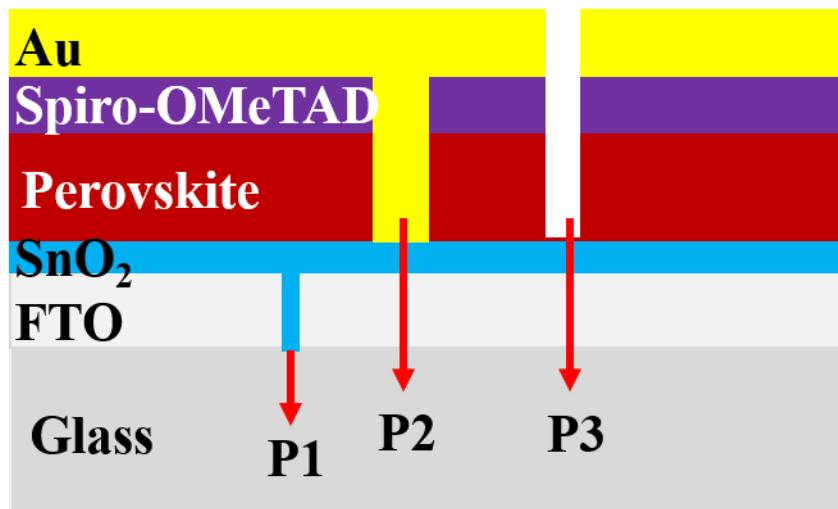


Fig. S17 Schematic drawing showing the side view of the perovskite solar module showing the interconnections including P1, P2, P3 lines.

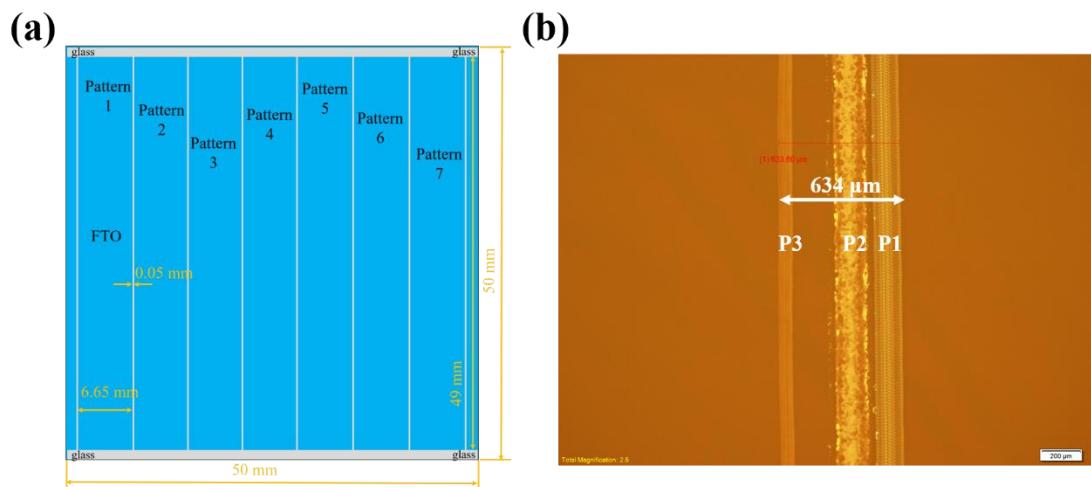


Fig. S18 (a) Top view of the $5 \times 5 \text{ cm}^2$ FTO substrate pattern. (b) Optical photograph of the sub-cell separation including P1, P2 and P3 patterns in the $5 \times 5 \text{ cm}^2$ SnO₂/K-PSM. The corresponding geometric fill factor (GFF) is determined to be approximately 0.905.

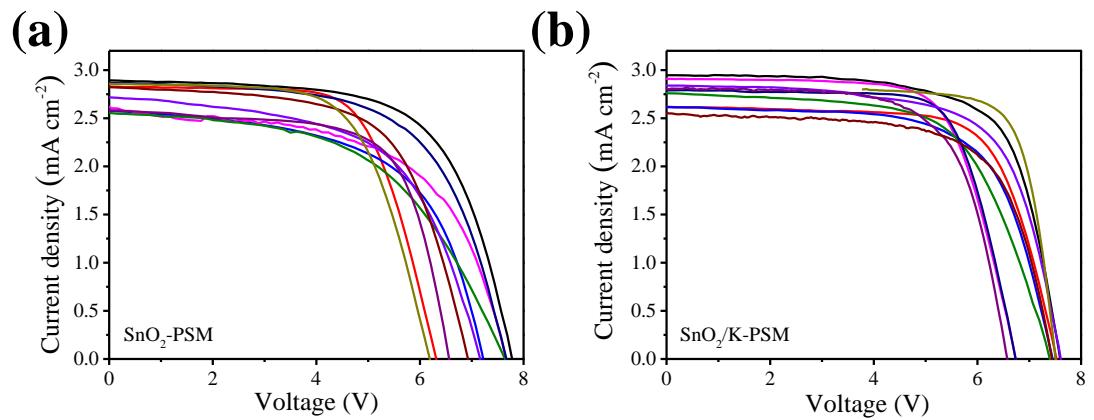


Fig. S19 J-V curves of the $5 \times 5 \text{ cm}^2$ PSMs based on 10 devices under reverse scan for (a) $\text{SnO}_2\text{-PSMs}$. (b) $\text{SnO}_2/\text{K-PSMs}$.

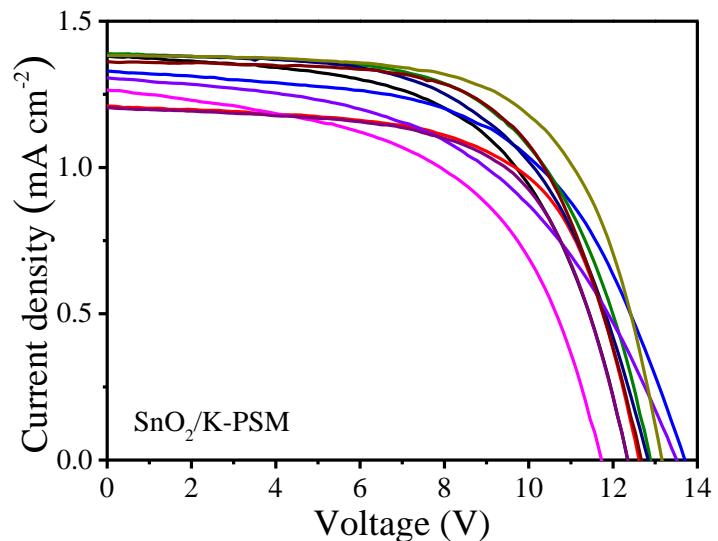


Fig. S20 J-V curves of the $10 \times 10 \text{ cm}^2$ $\text{SnO}_2/\text{K-PSMs}$ based on 10 devices under reverse scan.

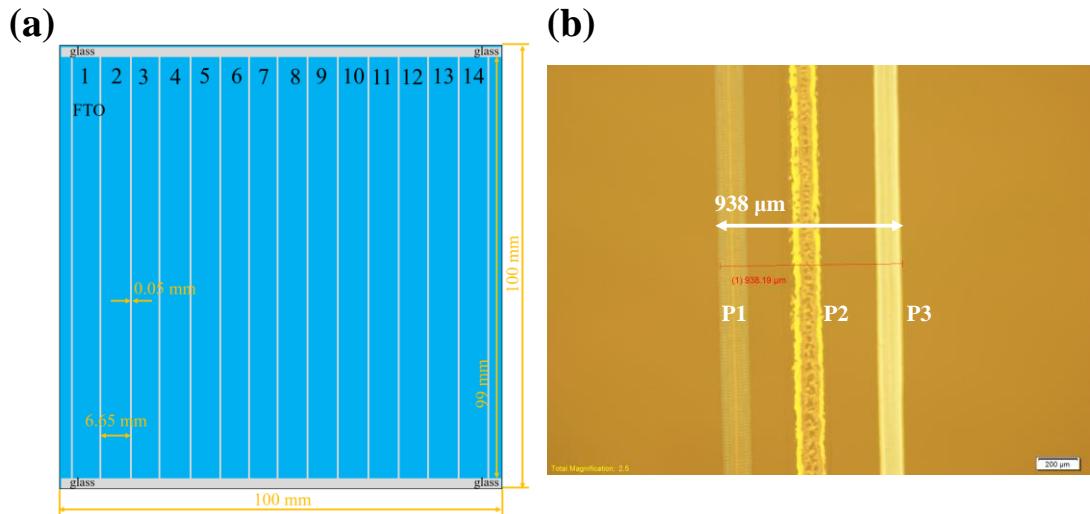


Fig. S21 (a) Top view of the $10 \times 10 \text{ cm}^2$ FTO substrate pattern. (b) Optical photograph of the sub-cell separation including P1, P2 and P3 patterns in $10 \times 10 \text{ cm}^2$ SnO_2/K -PSM. The corresponding GFF is determined to be approximately 0.860.

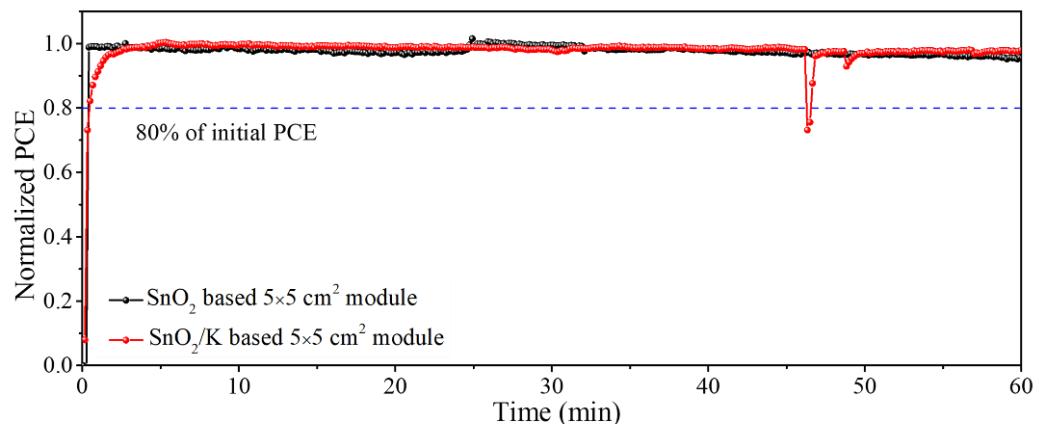


Fig. S22 Operational stability of the $5 \times 5 \text{ cm}^2$ PSMs with encapsulation under a steady applied voltage and constant illumination (AM 1.5G, 100 mW cm^{-2}).

Tables

Table S1 The comparison of the efficiency and active area of perovskite solar cells by employing chemical bath deposition.

SnO ₂ deposition	Device	Active area (cm ²)	Total area (cm ²)	PCE ^a (%)	PCE ^b (%)	Lifetime	Ref.
CBD	FTO/SnO ₂ /PVSK/Spiro/Au	0.09	/	/	21.70	/	This work
		22.4	25 (5 × 5)	15.62	17.26	1006 h/T ₈₀	
		91.8	100 (10 × 10)	11.80	13.72	/	
CBD	FTO/SnO ₂ /PVSK/Spiro/Au	0.0937	/	/	25.4	/	[1]
		0.984	/	/	23	/	
CBD	FTO/SnO ₂ /PVSK/Spiro/Au	0.16	/	/	20.56	/	[2]
		20.0	36 (6 × 6)	/	15.76	/	
CBD	FTO/SnO ₂ /PCBM/PVK/Spiro/Ag	0.0919	/	/	17.1	/	[3]
CBD	FTO/SnO ₂ /PVSK/Spiro/Au	0.16	/	/	20.7	/	[4]
CBD	FTO/SnO ₂ /PVSK/Spiro/Au	0.049	/	/	23.2	/	[5]
CBD	ITO/SnO ₂ /PVSK/Spiro/Au	0.1	/	/	14.8	/	[6]
Spin-coating+CBD	FTO/SnO ₂ /PVSK/SWNT-Spiro/Ag	0.0919	/	/	18.8	/	[7]

(Note: a is designated area of solar module; b is normalized by active area.)

Table S2 Fast and slow components for the TRPL decay.

Sample	A ₁ (%)	τ ₁ (ns)	A ₂ (%)	τ ₂ (ns)	τ _{average} (ns)
SnO ₂	0.656	143.8	0.344	9.2	139.4
SnO ₂ /K	0.623	76.3	0.382	10.3	71.3

Table S3 Photovoltaics parameters of the SnO₂ based PSCs with different amounts of KMnO₄.

Sample	Scan direction	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)	HI
0 mM	FS.	1.069	22.82	0.699	17.05	1.18
	RS.	1.114	22.90	0.787	20.09	
5 mM	FS.	1.056	23.03	0.728	17.72	1.15
	RS.	1.096	23.06	0.807	20.39	
7 mM	FS.	1.107	23.28	0.778	20.07	1.06
	RS.	1.118	23.39	0.817	21.36	
8 mM	FS.	1.118	23.22	0.792	20.58	1.05
	RS.	1.127	23.39	0.823	21.70	
9 mM	FS.	1.108	23.08	0.786	20.10	1.03
	RS.	1.117	23.10	0.799	20.62	
10 mM	FS.	1.106	22.88	0.772	19.53	0.97
	RS.	1.111	22.61	0.757	19.03	

Table S4 Statistical photovoltaic parameters of open-circuit voltage (V_{oc}), short-circuit photocurrent density (J_{sc}), fill factor (FF) and power conversion efficiency (PCE) of the SnO₂-PSCs and SnO₂/K-PSCs based on 10 devices.

Sample	Scan direction	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)	HI
SnO ₂	FS.	1.04±0.04	22.6±0.4	0.59±0.05	13.9±1.4	1.40
	RS.	1.09±0.02	22.6±0.4	0.79±0.01	19.5±0.5	
SnO ₂ /K	FS.	1.09±0.01	23.0±0.3	0.78±0.02	19.5±0.5	1.05
	RS.	1.11±0.01	23.1±0.3	0.81±0.01	20.6±0.5	

Table S5 Fitting parameters of the EIS measurement of the PSCs based on the SnO₂ and SnO₂/K substrates.

Sample	R _s (Ω)	R _{ct} (Ω)
SnO ₂	26.01	62.57
SnO ₂ /K	16.67	22.44

Table S6 Statistical photovoltaic parameters of open-circuit voltage (V_{oc}), short-circuit photocurrent density (J_{sc}), fill factor (FF) and power conversion efficiency (PCE) of the SnO₂-PSCs and SnO₂/K-PSCs based on 10 devices after 5 months storage in ambient air in a dry room with a relative humidity of ~20% without any encapsulation

Sample	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
SnO ₂	1.08±0.02	21.9±0.5	0.73±0.03	17.4±0.8
SnO ₂ /K	1.11±0.01	22.4±0.3	0.76±0.04	18.8±0.8

Table S7 Photovoltaics parameters of 10 SnO₂ based perovskite solar modules (5 × 5 cm² PSMs) under reverse scan.

Sample	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
1	7.158	2.72	0.582	11.34
2	6.560	2.59	0.677	11.48
3	6.920	2.82	0.632	12.36
4	6.186	2.86	0.647	11.44
5	7.663	2.87	0.620	13.64
6	7.635	2.56	0.530	10.35
7	7.656	2.60	0.580	11.55
8	6.311	2.83	0.677	12.09
9	7.218	2.58	0.583	10.87
10	7.770	2.89	0.649	14.58

Average	7.108 ± 0.536	2.73 ± 0.12	0.618 ± 0.043	11.97 ± 1.16
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Table S8 Photovoltaics parameters of 10 SnO₂/K based perovskite solar modules ($5 \times 5 \text{ cm}^2$ PSMS) under reverse scan.

Sample	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
1	7.520	2.62	0.706	13.91
2	7.602	2.84	0.674	14.55
3	6.738	2.91	0.698	13.68
4	7.400	2.76	0.631	12.88
5	7.437	2.55	0.674	12.76
6	6.573	2.81	0.672	12.41
7	6.735	2.79	0.725	13.63
8	7.478	2.80	0.661	13.83
9	7.450	2.62	0.664	12.93
10	7.591	2.95	0.699	15.62
Average	7.252 ± 0.363	2.76 ± 0.12	0.680 ± 0.024	13.62 ± 0.87

Table S9 Photovoltaics parameters of the champion efficiency of the SnO₂/K based perovskite solar modules ($5 \times 5 \text{ cm}^2$ PSMS).

Sample	Scan direction	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)	HI
SnO ₂	FS.	7.413	2.66	0.379	7.49	1.95
	RS.	7.770	2.89	0.649	14.58	
SnO ₂ /K	FS.	7.166	2.98	0.667	14.25	1.10
	RS.	7.591	2.95	0.699	15.62	

Table S10 The comparison of the efficiency and operational stability of perovskite solar modules with the area over 20 cm².

Device	Active area (cm ²)	Total area (cm ²)	PCE ^a (%)	PCE ^b (%)	Lifetime	Ref
FTO/SnO₂/Cs_{0.05}FA_{0.85}MA_{0.10}PbI_{2.85}Br_{0.15}/Spiro/Au	22.4	25 (5 × 5)	15.62	17.26	1006 h/T₈₀	This work
	91.8	100 (10 × 10)	11.80	13.72	/	
FTO/SnO ₂ /MAPbI ₃ /Spiro/Au	21	36 (6 × 6)	/	18.13	100 h/T ₈₀	[8]
ITO/P3HT/MAPbI ₃ /PCBM/Ag	36.6	49 (7 × 7)	/	16.06	250 h/T ₉₀	[9]
ITO/PTAA/MA _{0.7} FA _{0.3} PbI ₃ /C ₆ /BCP/Cu	/	35.8	/	18.5	/	[10]
FTO/SnO ₂ [(CsPbI ₃) _{0.05} (FAPbI ₃) _{0.85} (MAPbBr ₃) _{0.15}] _{0.95} /Spiro/Au	25	36 (6.5 × 6.5)	15.3	16.02	/	[11]
FTO/TiO ₂ /MAPbI ₃ /Spiro/Au	50.6	100 (10 × 10)	/	12.6	1630 h/T ₈₀	[12]
FTO/SnO ₂ /(K _x (Cs _{0.05} (FA _{0.85} MA _{0.15}) _{0.95} Pb(I _{0.85} Br _{0.15}) ₃)/Spiro/Au	20	36 (6 × 6)	/	15.76	/	[2]
FTO/SnO ₂ /(FAPbI ₃) _{0.95} (CsPbBr ₃) _{0.05} /Spiro/Au	~20 (19.69)	25 (5 × 5)	/	17.94	/	[13]
ITO/SnO ₂ /Cs _{0.05} FA _{0.54} MA _{0.41} Pb(I _{0.98} Br _{0.02}) ₃ /Spiro/Au	22.4	25 (5 × 5)	14.55	16.35	1625 h/T ₈₀	[14]
FTO/ZnO-ZnS/FA _{0.97} Cs _{0.03} PbI ₃ /Spiro/Au	49	100 (10 × 10)	/	13.84	/	[15]
FTO/TiO ₂ /SnO ₂ /Cs _{1-x} FA _x Pb(I _y Cl _{1-y}) ₃ /BJ-GO/TFB/Cr/Au	35.8	64 (8 × 8)	15.3	/	1000 h/T ₉₀	[16]
ITO/SnO ₂ /EDTAK(EAMA)/Cs _{0.05} FA _{0.54} MA _{0.41} Pb(I _{0.98} Br _{0.02}) ₃ /Spiro-P3HT/Au	22.4	25 (5 × 5)	16.6	18.2	2680 h/T ₈₀	[17]
FTO/SnO ₂ /C ₆₀ /Cs _{0.1} FA _{0.9} PbI _{2.9} Br _{0.1} /Spiro/Au	22.4	25 (5 × 5)	~10	/	250 h/T ₉₀ 500 h/T ₈₀	[18]
	91.8	100 (10 × 10)	9.34	10.37	/	
FTO/NiO _x [(CH(NH ₂) ₂) _{0.85} [CH ₃ NH ₃] _{0.15} Pb(I _{0.85} Br _{0.15}) ₃ /PCBM/BCP/Ag	36.1 ^{ap}	64 (8 × 8)	/	15.6	1000 h/T ₉₁ 2222 h/T ₈₀	[19]
FTO/SnO ₂ /MAPbI ₃ /Spiro/Au	22.8	25 (5 × 5)	12.03	/	280 h/T ₉₀ 515 h/T ₈₀	[20]
FTO/TiO ₂ /MAPbI ₃ /Spiro/Au	36.1 ^{ap}	64	/	15.7	500 h/T ₉₀	[21]

		(8 × 8)			1000 h/T ₈₀	
FTO/TiO ₂ /ZrO ₂ /(5- AVA) _x (MA) _{1-x} PbI ₃ /carbon	49	100 (10 × 10)	/	10.4	1000 h	[22]
FTO/c-TiO ₂ /ZrO ₂ /(5- AVA) _x (MA) _{1-x} PbI ₃ /carbon	46.7	100 (10 × 10)	/	11.2	> 10000 h	[23]
FTO/TiO ₂ /ZrO ₂ /(5- AVA) _x (MA) _{1-x} PbI ₃ /carbon	31	50 (5 × 10)	/	10.46	72 h	[24]
	70	100 (10 × 10)	/	10.75	/	

(Note: a is designated area of solar module; b is normalized by active area.)

Table S11 Photovoltaics parameters of 10 SnO₂/K based perovskite solar modules (10 × 10 cm² PSMs) under reverse scan.

Sample	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
1	12.337	1.38	0.586	9.97
2	12.615	1.21	0.634	9.68
3	13.698	1.33	0.571	10.39
4	11.713	1.27	0.539	7.99
5	12.880	1.39	0.611	10.92
6	12.828	1.38	0.588	10.45
7	13.498	1.31	0.510	8.99
8	12.346	1.20	0.636	9.45
9	12.661	1.36	0.637	10.98
10	13.158	1.38	0.648	11.80
Average	12.773±0.529	1.32±0.07	0.596±0.042	10.06±0.99

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