Low-Temperature Plasma-Enhanced Atomic Layer Deposition of Tin Oxide Electron Selective Layers for Highly Efficient Planar Perovskite Solar Cells

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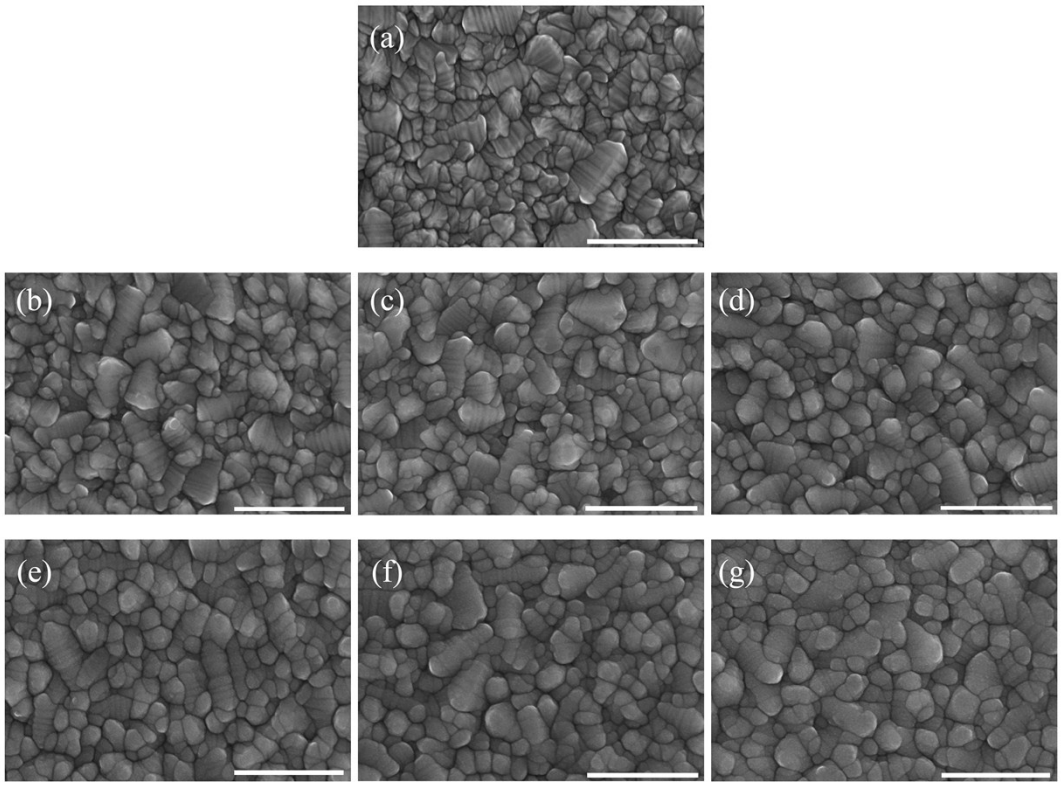

Fig. S1 SEM images of PEALD grown SnO_2 ESLs with different reaction cycles on FTO coated glass substrates: a-g: 0, 40, 70, 100, 130, 160, and 190 cycles. The processing temperature was 100 ^{o}C. The scale bar is 500 nm for all images.

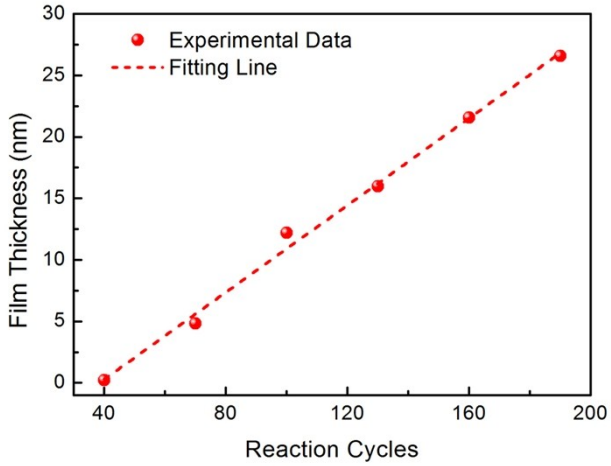

Fig. S2 Film thickness of SnO_2 ESLs deposited by PEALD at 100 °C with different reaction cycles as determined by spectroscopic ellipsometry.

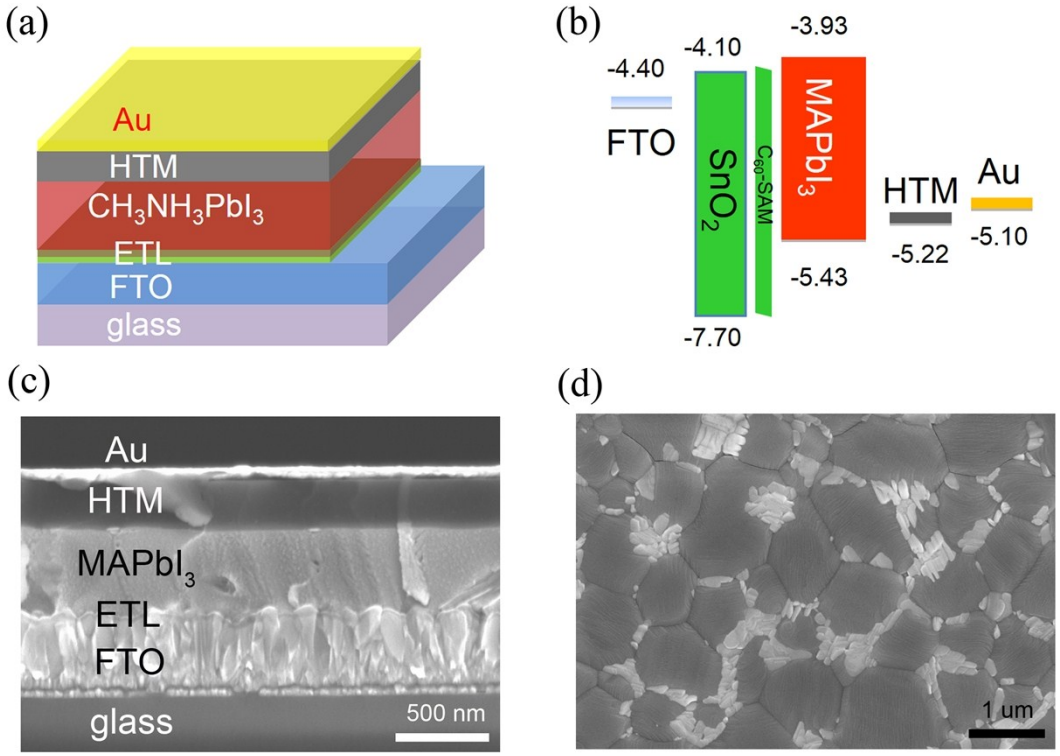

Fig. S3 (a) Device structure and (b) schematic energy band diagram of our perovskite solar cells (PVSCs). (c) Cross-sectional SEM image of our entire PVSC. (d) Top-view SEM image of perovskite film deposited on SnO_2/C_{60}-SAM coated FTO substrate.

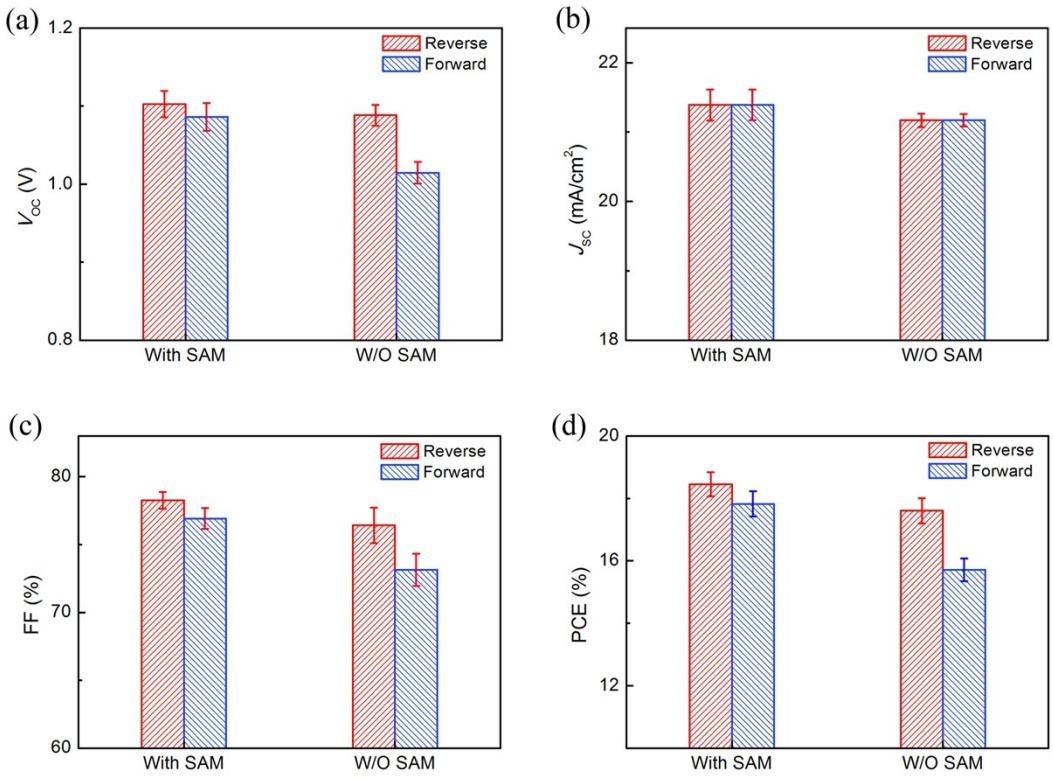

Fig. S4 Statistical device performance results of 20 cells with and without C_{60}-SAM interlayers. The SnO_2 ESLs were deposited by PEALD at 100 °C with 130 reaction cycles. Error bars represent the standard deviation calculated from 20 devices prepared at the same conditions.

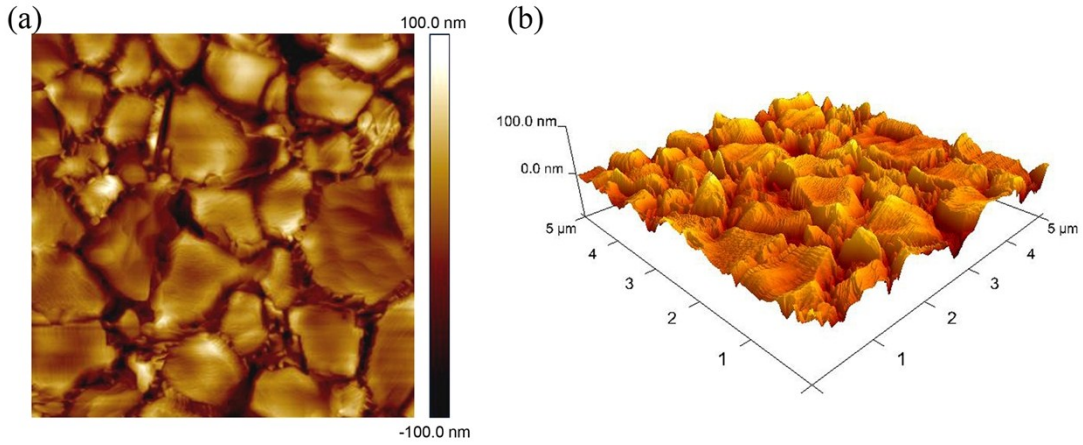

Fig. S5 AFM images (5µm×5µm) of perovskite film deposited on C_{60}-SAM/SnO_2/FTO/Glass substrate. The average surface roughness value was found to be R_a = 11.5 nm.

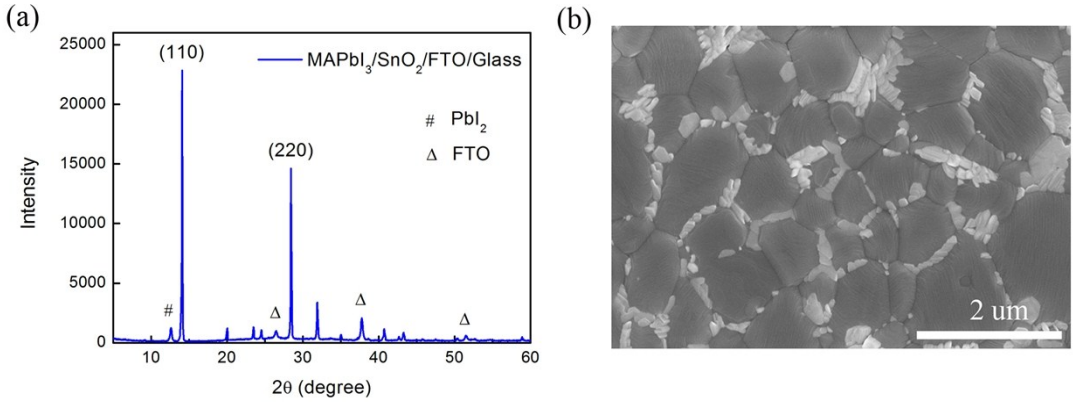

Fig. S6 XRD pattern (left) and SEM (right) of a perovskite film deposited on C_{60}-SAM/SnO_2/FTO/Glass substrate. The bright particles at the grain boundaries are PbI_2 as confirmed in the XRD pattern with a PbI_2 peak (#) at ~12.80^o.

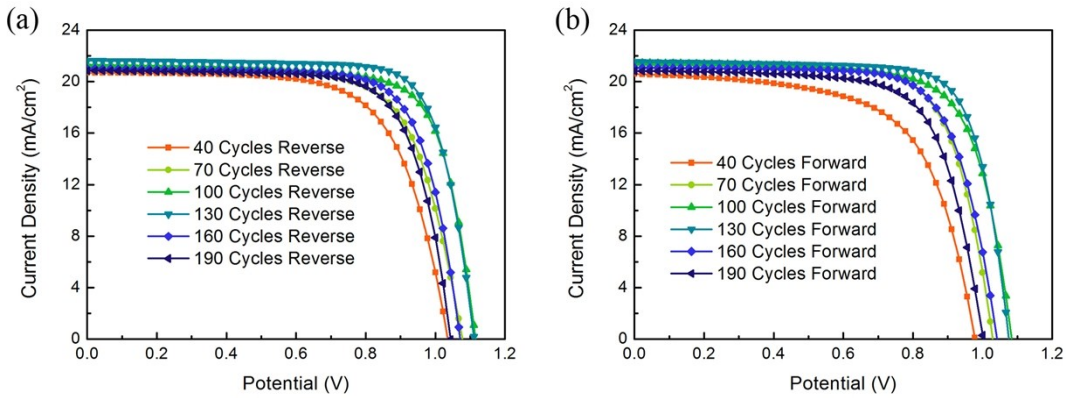

Fig. S7 *J–V* curves of representative perovskite cells using PEALD SnO_2 ESLs deposited with various reaction cycles measured under reverse and forward voltage scans. The number of reaction cycles for the PEALD SnO_2 was 130.

Table S1 Average data for 54 of PVSCs based on PEALD SnO_2 ESL on FTO/glass substrates with various reaction cycles. The processing temperature was 100 °C for all samples. Error values represent the standard deviation (s.d.) calculated from 54 devices prepared at the same conditions.

Reaction cycles	Scan direction	V_{OC} (V)	J_{SC} (mA/cm^2)	FF (%)	PCE (%)
40	Reverse	1.08±0.02	19.96±0.35	67.80±1.99	14.61±0.90
	Forward	1.02±0.04	19.95±0.38	62.57±2.23	12.76±1.04
70	Reverse	1.09±0.01	20.62±0.08	72.77±1.08	16.30±0.45
	Forward	1.05±0.02	20.62±0.08	68.74±1.34	14.88±0.61
100	Reverse	1.09±0.01	21.30±0.10	75.03±1.05	17.43±0.27
	Forward	1.06±0.01	21.31±0.12	70.29±1.28	15.83±0.36
130	Reverse	1.08±0.01	21.43±0.08	76.53±1.27	17.73±0.25
	Forward	1.05±0.01	21.43±0.07	72.62±2.08	16.30±0.66
160	Reverse	1.07±0.01	21.32±0.11	73.63±1.03	16.87±0.24
	Forward	1.04±0.01	21.32±0.12	69.37±0.56	15.41±0.39
190	Reverse	1.05±0.01	20.97±0.12	72.52±0.53	15.96±0.14
	Forward	1.01±0.03	20.97±0.12	67.59±1.38	14.28±0.12

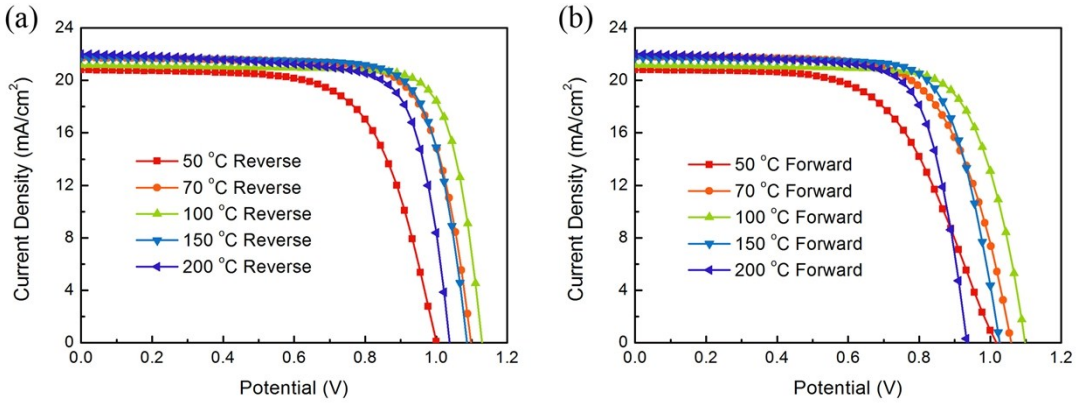

Fig. S8 *J–V* curves of representative perovskite cells using PEALD SnO_2 ESLs deposited at different processing temperatures measured under reverse and forward voltage scans. The number of reaction cycles for the PEALD was 130.

Table S2 Average photovoltaic performance of 40 PVSCs fabricated on SnO_2 ESL at different deposition temperature. The number of reaction cycles was kept at 130 for all samples. Error values represent the standard deviation calculated from 45 devices prepared at the same conditions.

Reaction temperature (°C)	Scan direction	V_{OC} (V)	J_{SC} (mA/cm^2)	FF (%)	PCE (%)
50	Reverse	1.01±0.02	20.82±0.13	71.96±1.63	15.15±0.31
	Forward	1.00±0.02	20.79±0.14	64.45±2.16	13.42±0.54
70	Reverse	1.07±0.04	21.18±0.29	73.86±1.43	16.71±1.10
	Forward	1.03±0.04	21.18±0.29	70.37±1.42	15.32±0.63
100	Reverse	1.11±0.02	21.08±0.42	77.62±1.34	18.21±0.59
	Forward	1.10±0.01	21.08±0.42	73.18±1.77	16.98±0.49
150	Reverse	1.08±0.02	21.41±0.33	76.72±0.88	17.64±0.53
	Forward	1.02±0.02	21.41±0.33	74.63±0.90	16.25±0.54
200	Reverse	1.03±0.02	21.66±0.27	73.23±1.24	16.28±0.55
	Forward	0.93±0.01	21.66±0.27	72.69±1.26	14.70±0.45

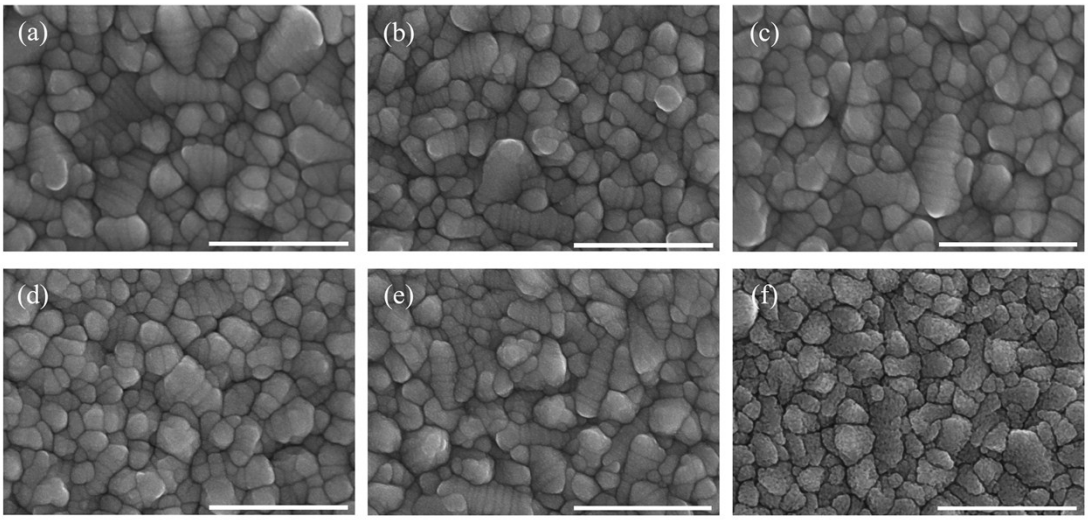

Fig. S9 The SEM images of PEALD SnO_2 coated FTO/glass substrates with different deposition temperature: (a) 50 ^oC, (b) 70 ^oC, (c) 100 ^oC, (d) 150 ^oC, (e) 200 ^oC and (f) 400 ^oC. The number of PEALD reaction cycles is 130 for all samples. The scale bar is 500 nm for all images. We can see that under higher temperature, SnO_2 will crystallize and resulting in pinholes.

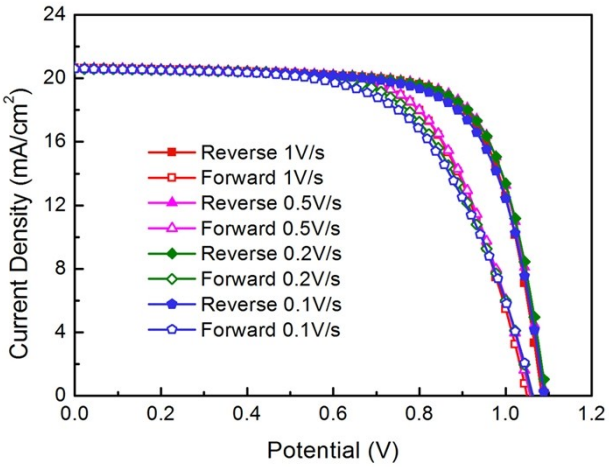

Fig. S10 *J-V* curves of one flexible PVSC measured under reverse and forward voltage scans with various scan rates.

Table S3 *J-V* parameters of the flexible PVSC measured under reverse and forward voltage scans with various scan rates.

Scan direction	Scan speed (V/s)	V_{OC} (V)	J_{SC} (mA/cm^2)	FF (%)	PCE (%)
Reverse	1	1.09	20.65	73.32	16.50
Forward	1	1.05	20.63	66.57	14.42
Reverse	0.5	1.09	20.63	73.64	16.56
Forward	0.5	1.06	20.62	66.13	14.45
Reverse	0.2	1.09	20.63	73.12	16.44
Forward	0.2	1.06	20.61	63.52	13.88
Reverse	0.1	1.09	20.62	71.23	16.01
Forward	0.1	1.06	20.61	62.27	13.60

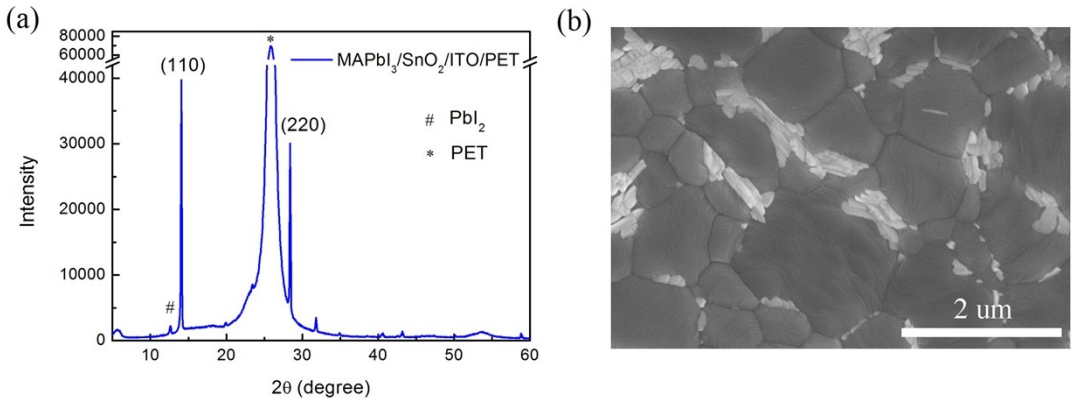

Fig. S11 (a) XRD pattern and (b) SEM of a perovskite film deposited on C_{60}-SAM/SnO_2/ITO/PET substrate. The relatively bright particles are PbI_2.

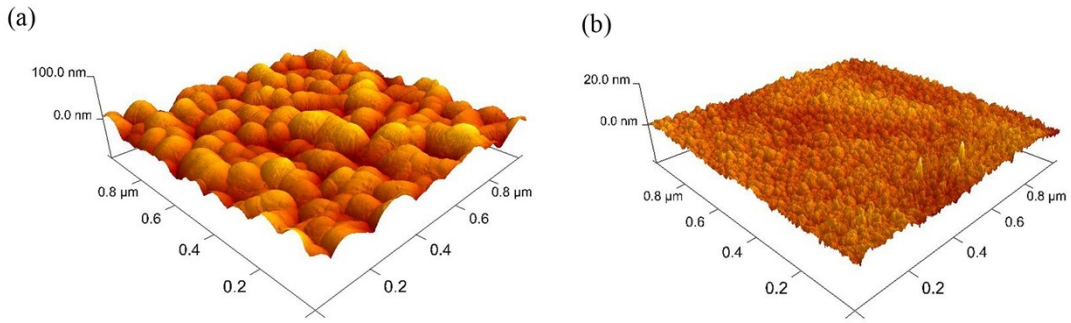

Fig. S12 AFM images (1µm×1µm) of (a) FTO-coated glass substrate and (b) ITO-coated PET substrate.

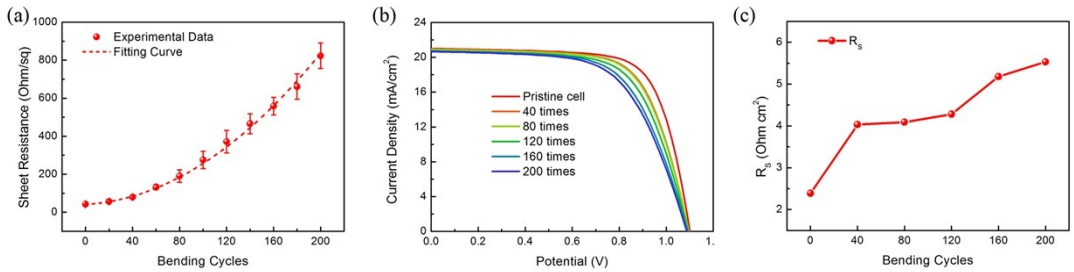

Fig. S13 (a) Sheet resistance of ITO/PET flexible substrate versus bending cycles. (b) *J-V* curves of a flexible PVSC measured under reverse voltage scans after various bending cycles. (c) The measured series resistance of the flexible PVSC after bending cycles.

Table S4 Average photovoltaic performance of 30 flexible PVSCs fabricated on SnO_2 ESL coated ITO/PET substrate with different scan directions. The deposition temperature of SnO_2 ESL is 100 ^oC with 130 reaction cycles. Error values represent the standard deviation calculated from 30 devices prepared at the same conditions.

Scan direction	V_{OC} (V)	J_{SC} (mA/cm^2)	FF (%)	PCE (%)
Reverse	1.07±0.04	20.68±0.22	70.33±3.18	15.57±1.14
Forward	1.03±0.05	20.73±0.28	61.70±3.42	13.18±1.00