Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supplementary information

Deep level trapped defect analysis in $CH_3NH_3PbI_3$ perovskite solar cells by Deep Level Transient Spectroscopy

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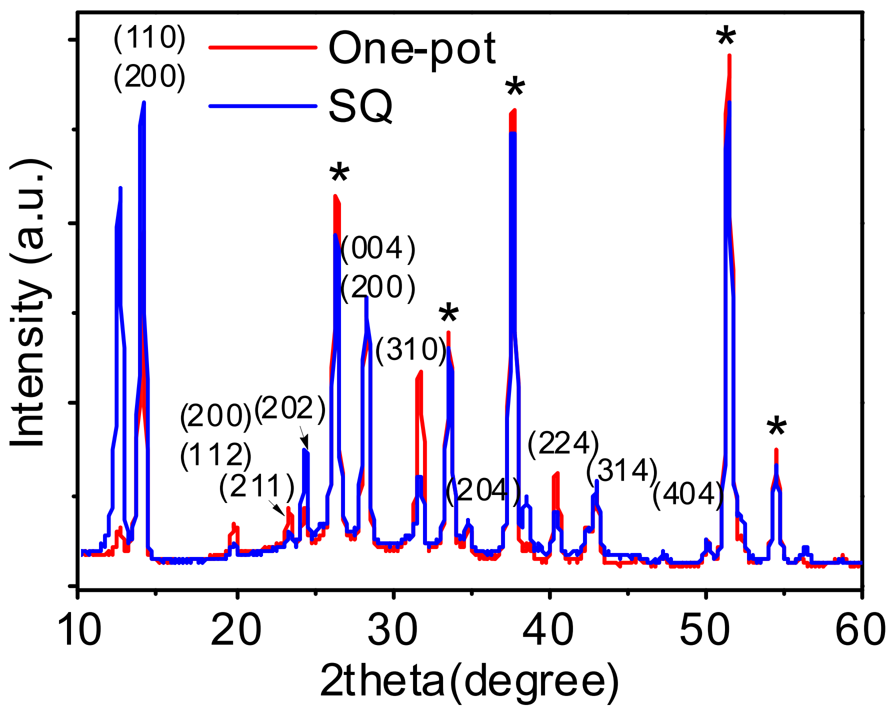


Fig. S1. X-Ray diffraction patterns of the perovskite films prepared by One-pot and SQ methods. The asterisk marks indicate that they are from FTO.

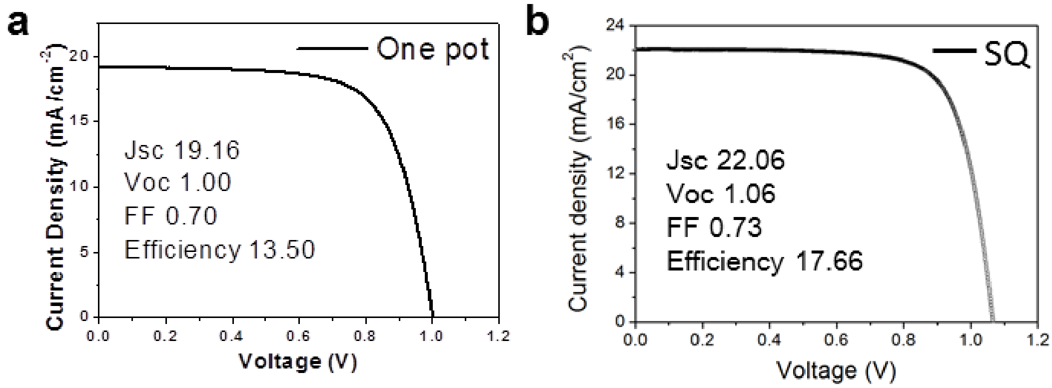


Fig. S2. The photovoltaic efficiencies of the perovskite solar cells (a) Solar cell parameters of One-pot sample and (b) SQ sample.

The DLTS measurements measurements were carried out with a PhysTech FT 1030 DLTS system at various temperatures with forward and reverse bias. Temperature scans were made between 20 and 430 K, at a heating rate of 2 K/min. The capacitances of samples were measured using a modified Boonton 72B capacitance meter with a 1 MHz capacitance meter. The pulse height, filling pulse width, and pulse period width were 0.4 V, 10 ms, and 10 ms, respectively. The applied bias voltages were 0.4eV and 0.8eV for the forward mode, and -0.8eV and -0.4eV for the reverse mode, respectively. The measurements were conducted 4 times for checking out reproducibility. The pulse frequency for the voltage was 100MHz. We characterized the cell just after fabrication (as-prepared) and repeated the measurements every 5 days.

The composition change of the perovskite layer was characterized with X-ray Photoemission Spectroscopy (XPS). The XPS depth profiles of as-prepared, aged 5 days, and aged 10 days are shown in Fig S3.

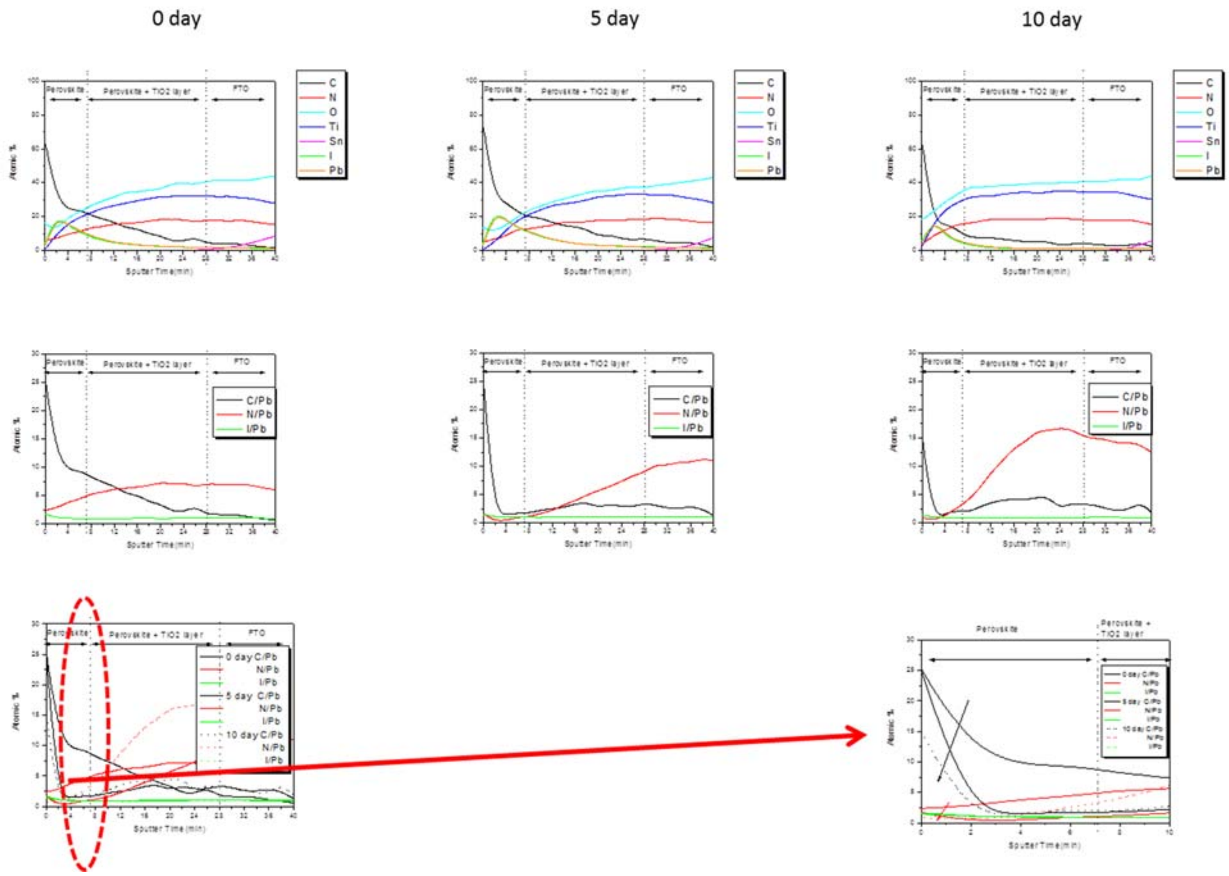
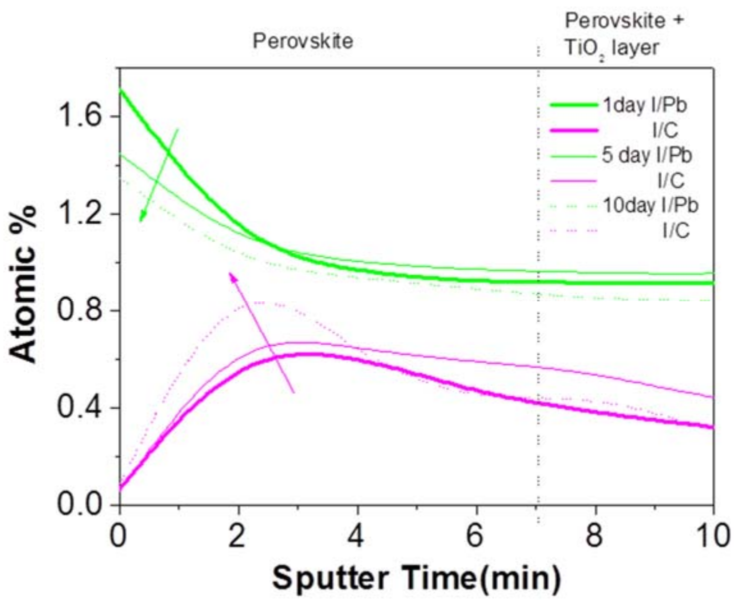
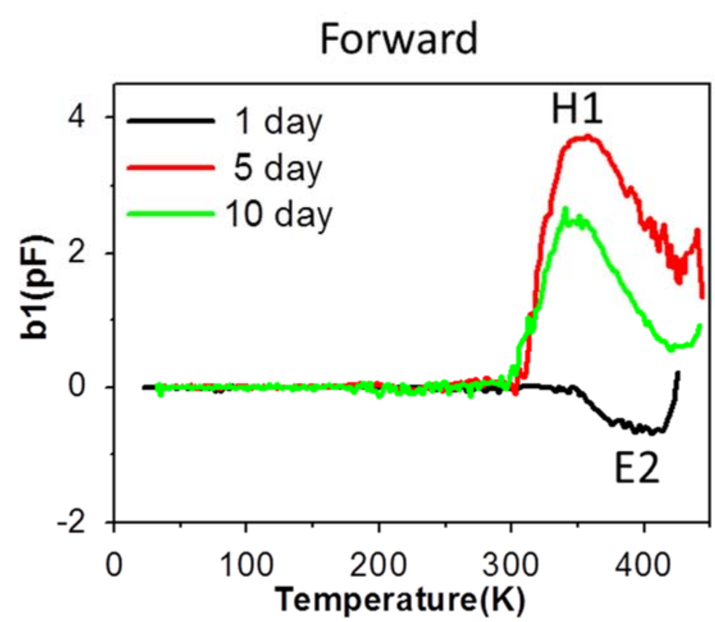


Fig. S3. The composition change of the perovskite layer was characterized with X-ray Photoemission Spectroscopy (XPS). The XPS depth profiles of as-prepared, aged 5 days, and aged 10 days.


Perovskite
Perovskite + TiO_2 layer
1day I/Pb
I/C
5 day I/Pb
I/C
10day I/Pb
I/C
Atomic %
Sputter Time(min)


Forward
1 day
5 day
10 day
H1
E2
b1(pF)
Temperature(K)

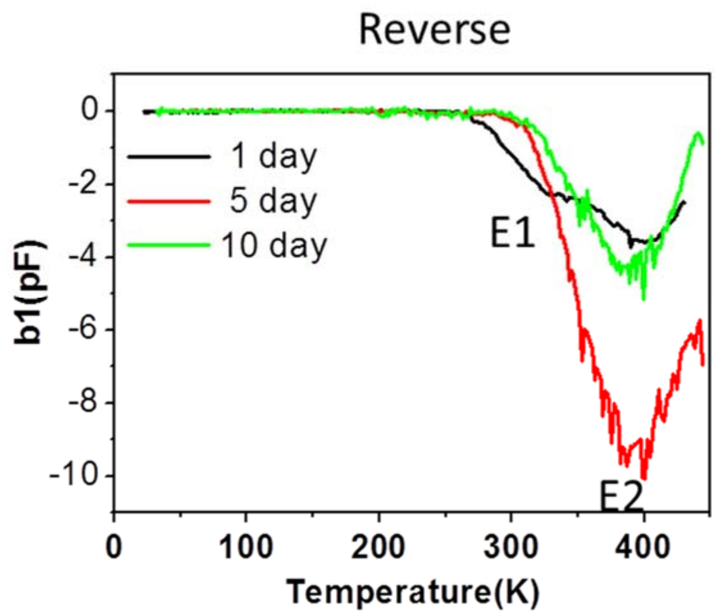


Fig. S4. The DLTS data for the aging effect

Table S1. Quantitative EDX analysis displayed in Figure 1.

Sample	C1s (at.%)	Pb4f (at.%)	I3d5 (at.%)	I/Pb (E1 defect)	I/C (E2 defect)
1 (One-pot) reverse	38.87	20.6	40.54	1.97	1.04
2 (One-pot) forward	37.54	21.7	40.76	1.88	1.09
3 (Cuboid) reverse	38.48	20.56	40.96	1.99	1.06
4 (Cuboid) forward	15.24	29.84	54.91	1.84	3.60

Table S2. Defect parameters (activation energy, density and cross section) calculated using DLTS.

		One-pot		Cuboid	
		E1	E2	E1	E2
Forward region	Et (eV)	0.62		0.62	0.76
	Nt (cm^{-3})	1.3×10^{15}		5.0×10^{14}	9.5×10^{14}
	Cross section (σ, cm^2)	2.5×10^{-15}		5.2×10^{-14}	2.5×10^{-14}
Reverse region	Et (eV)		0.75		0.75
	Nt (cm^{-3})		3.9×10^{14}		2.0×10^{15}
	Cross section (σ, cm^2)		2.7×10^{-15}		5.0×10^{-14}
Carrier concentration		1.8×10^{16}		6.7×10^{16}	

Table S3. Summary of the One-pot and SQ samples investigated in this work.

	J-V Measurement				Capacitance Measurement
Sample	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	Efficiency (%)	Carrier density (cm^{-3})
One-pot	19.16	1.00	0.70	13.50	1.8×10^{16}
Cuboid	22.06	1.06	0.73	17.66	6.7×10^{16}

Reference

1. Im, J.-H. et al., 6.5% efficient perovskite quantum-dot-sensitized solar cell. *Nanoscale* **3**, 4088-4093 (2011).

2. Heo, J. H. et al., Efficient inorganic-organic hybrid heterojunction solar cells containing perovskite compound and polymeric hole conductors. *Nature Photonics* **7**, 486-491 (2013).

3. Burschka, J. et al., Sequential deposition as a route to high-performance perovskite-sensitized solar cells. *Nature* **499**, 316-319 (2013).