Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Degradation of Encapsulated Perovskite Solar Cells Driven by Deep Trap States and Interfacial Deterioration

Dhruba B. Khadka†‡*, Yasuhiro Shirai†**, Masatoshi Yanagida† and Kenjiro Miyano†

†Global Research Center for Environment and Energy based on Nanomaterials Science (GREEN), National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan.

‡ International Center for Young Scientists (ICYS), National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan.

Corresponding Author

*E-mail:

*KHADKA.B.Dhruba@nims.go.jp

**SHIRAI.Yasuhiro@nims.go.jp

Table and Figures

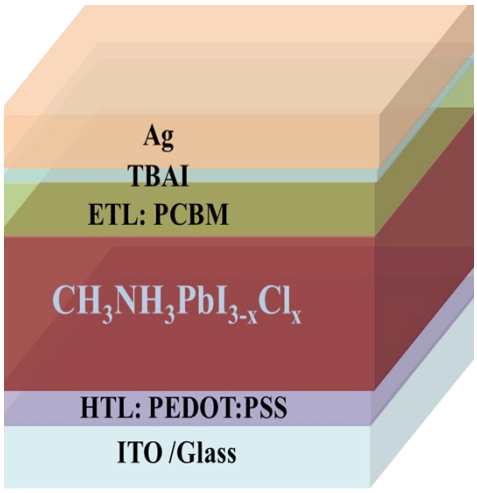

Figure S1. Schematic of device structure of the planar perovskite solar cell (PSC).

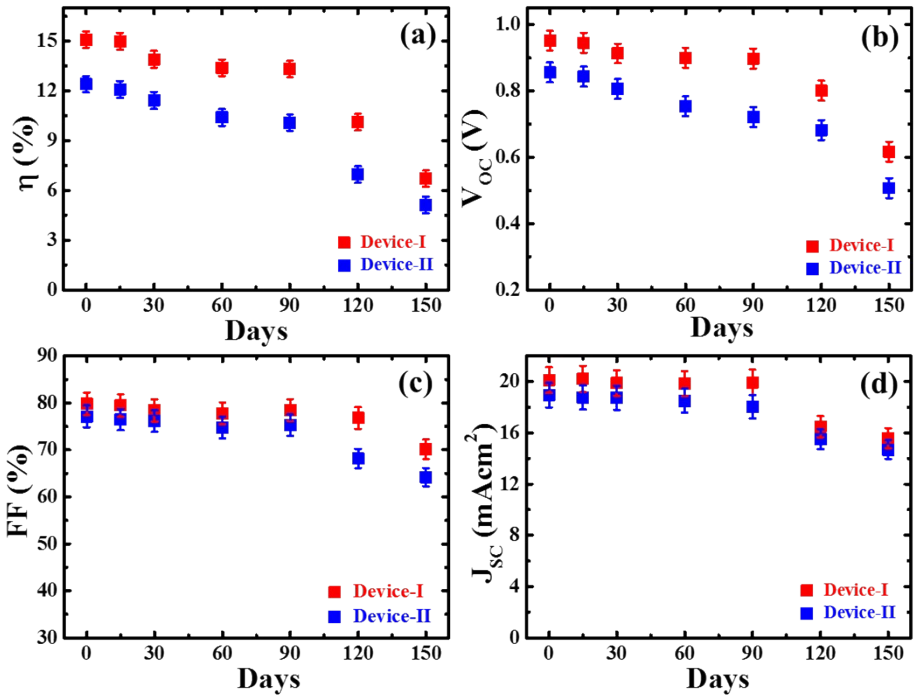

Figure S2. The device parameters (J_{SC}, V_{OC}, FF and η) of encapsulated devices stored under ambient conditions for 5 months.

Table S1. The summary of extracted parameters from analysis of J-V curve form fresh and most aged devices.

Electrical properties/ aging days	Device-I				Device-II			
	Fresh	90	120	150	Fresh	90	120	150
Jsc ($mAcm^{-2}$)	20.12	19.93	16.48	15.56	18.78	18.02	14.61	15.75
Voc (V)	0.95	0.90	0.79	0.62	0.86	0.72	0.68	0.51
FF	0.80	0.78	0.77	0.70	0.77	75.24	0.69	0.64
Rs ($\Omega \cdot cm^2$)	4.31	4.46	4.58	4.71	4.33	4.7	6.77	5.30
Rsh ($\Omega \cdot cm^2$)	$1.80x10^4$	$1.06 x10^4$	$5.89 x10^3$	$7.25 x10^2$	$3.58 x10^3$	$5.61 x10^3$	$2.59 x10^3$	$4.59 x10^2$
η(%)	15.12	13.33	10.12	6.71	12.41	10.10	6.96	5.13
A	1.12	1.39	1.51	1.67	1.41	1.53	1.60	1.96
J_0 ($mAcm^{-2}$)	$9.03 x10^{-12}$	$5.67 x10^{-10}$	$3.52 x10^{-8}$	$8.46 x10^{-6}$	$1.48 x10^{-9}$	$1.92 x10^{-7}$	$9.95 x10^{-7}$	$3.24 x10^{-4}$

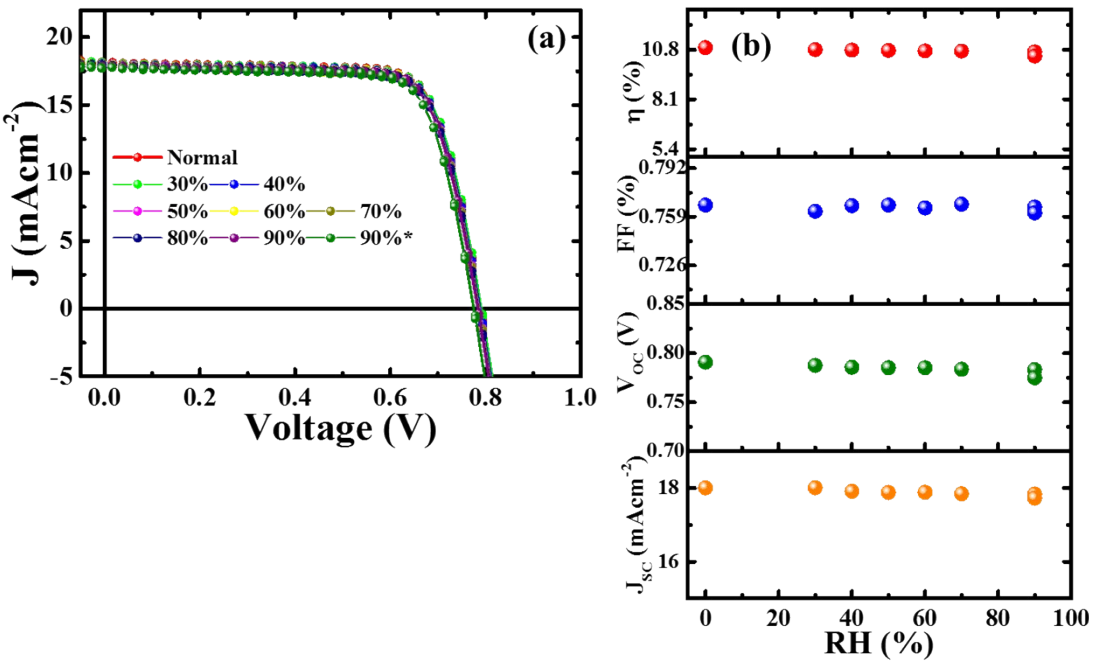

Figure S3. J-V characteristics of perovskite device measured under standard condition (one sun illumination, AM 1.5G, 100 mWcm^{-2}, 25°C) by varying relative humidity (RH) (30-90 %). Here 90%* stands for keeping the device under 90% RH ambient for 36 hours.

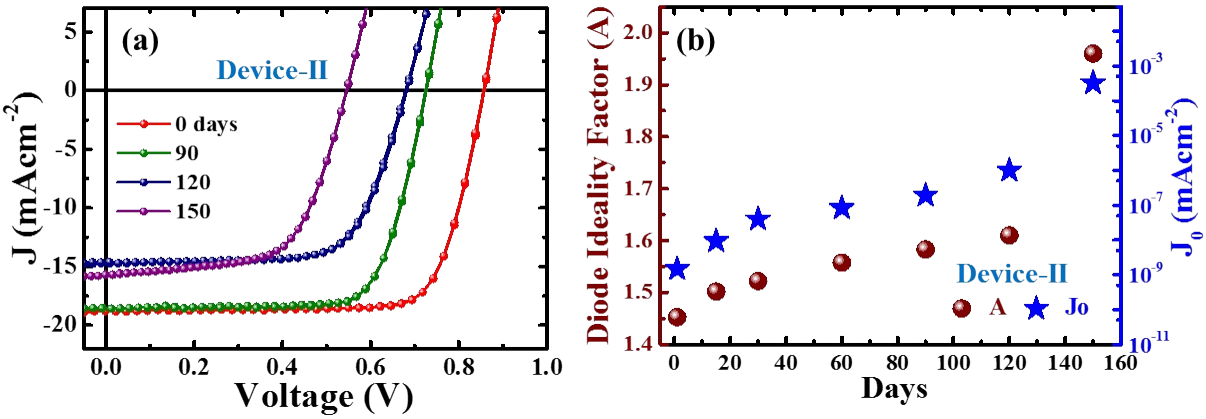

Fig. S4. J-V characteristics of the perovskite solar cells (device-II) after different aging time interval (a). Diode ideality factor (A) and reverse saturation current density (J_0) (b) of the perovskite devices extracted from analysis of respective J-V curves.

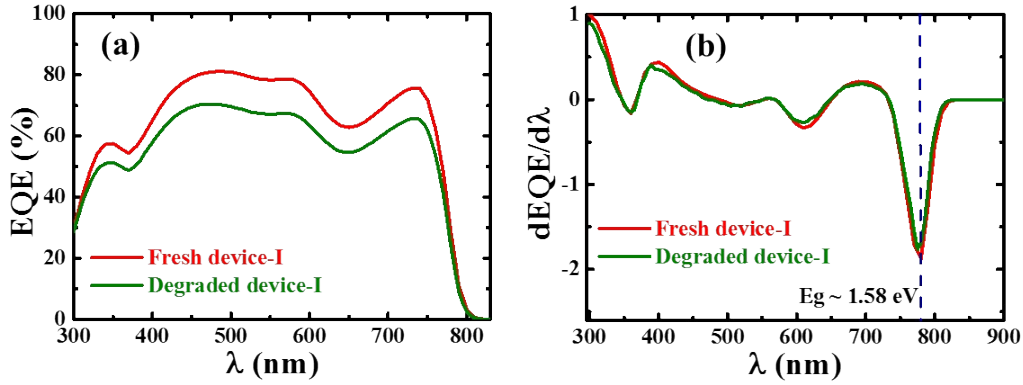

Figure S5. EQE response of fresh and aged perovskite devices of respective types and plots for estimation of band gap (E_g) of respective devices.

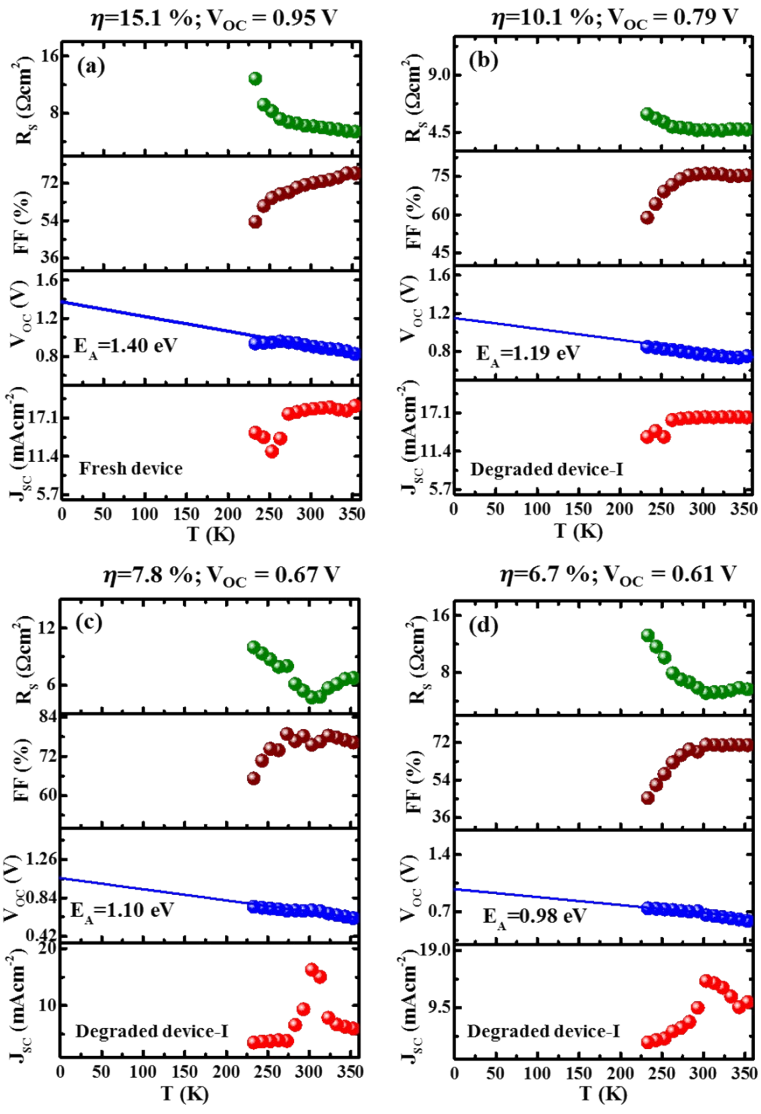

Figure S6. The device parameters of fresh (a) and degraded PSCs (b, c, d) measured in temperature range of 353- 233 K.

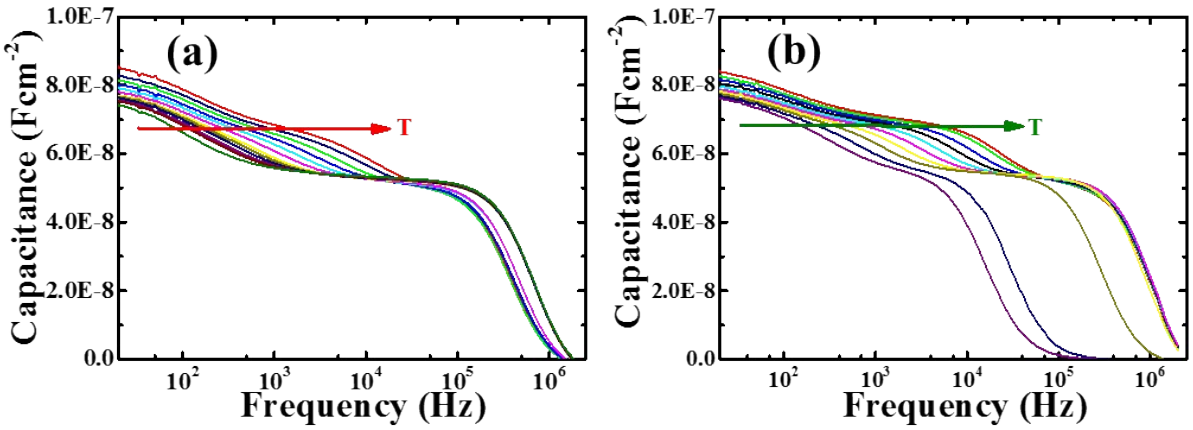

Figure S7. Capacitance- frequency (C-f-T) spectra of fresh (a) and degraded (b) devices.

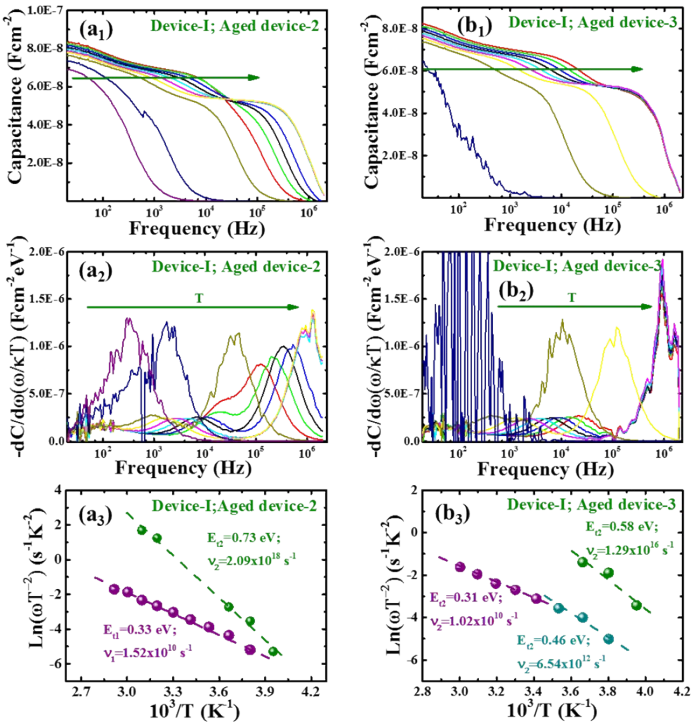

Figure S8. The plots of C-f-T spectra (a_1, b_1) and differentiation of C-f-T spectra showing the resonance frequency (a_2, b_2) and Arrhenius plots (a_3, b_3) of degraded devices.

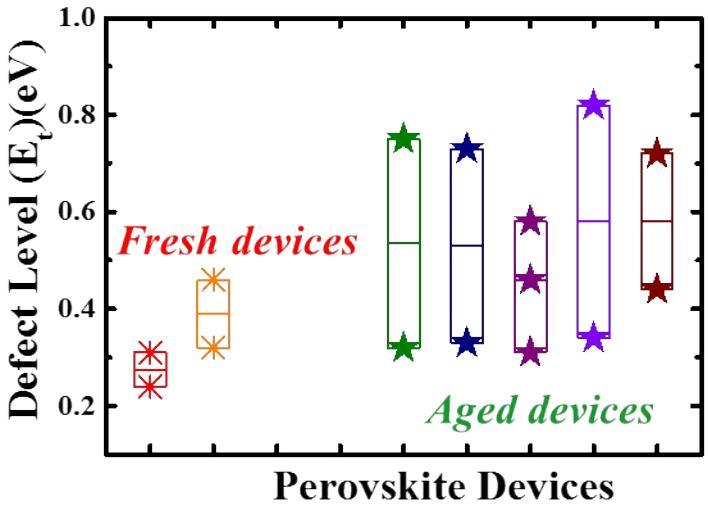

Figure S9. The plots of defect states extracted from analysis of C-f-T spectra of fresh and aged (degraded) devices.

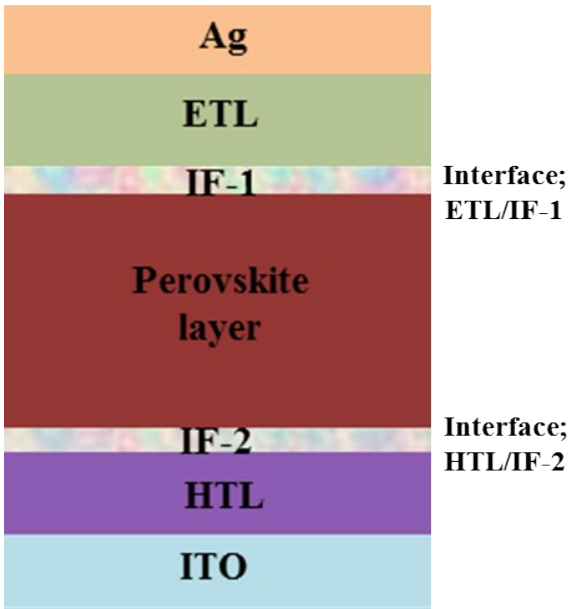

Figure S10. Schematic diagram of perovskite device layer defined for SCAPS simulation.

Table S2. Summary of simulation parameters of perovskite solar cell devices for SCAPS simulation adopted from earlier reports [1-3]. The device layers were defined considering our earlier reports and similar kind of solar cell. (HTL: hole transport layer, ETL: electron transport layer, and IF: interface layer.)

Material /Properties	HTL	IF-2	Perovskite	IF-1	ETL
x(μm)	0.035	0.015	0.350	0.010	0.060
E_g (eV)	3.2	1.55	1.55	1.55	3.2
χ(eV)	2.45	3.9	3.9	3.9	4.0
ε_r	3	22	22	22	3
Nc (cm^{-3})	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}
Nv (cm^{-3})	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}
v_n (cms^{-1})	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}
v_h (cms^{-1})	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}	1×10^{7}
μ_n/μ_h ($cm^2V^{-1}s^{-1}$)	$5\times10^{-4}/5\times10^{-4}$	2/2	4/4	2/2	$5\times10^{-4}/5\times10^{-4}$
N_d(cm^{-3})	-	1×10^{14}	1×10^{14}	1×10^{14}	5×10^{18}
N_a(cm^{-3})	1×10^{19}	1×10^{14}	1×10^{14}	1×10^{14}	-
N_t (cm^{-3})	1×10^{16}	1×10^{14}-1×10^{19}	1×10^{14} - 8×10^{15}	1×10^{14}-1×10^{19}	1×10^{16}
CC of e/h (cm^2)	$5\times10^{-14}/5\times10^{-14}$	$5\times10^{-14}/5\times10^{-14}$	$2\times10^{-14}/2\times10^{-14}$	$5\times10^{-14}/5\times10^{-14}$	$5\times10^{-14}/5\times10^{-14}$

E_t (eV)/distribution	0.5/ Gau	0.4/ Gau	(0.1-0.7)/ Gau	0.4/ Gau	0.5/ Gau
Interface; CC (cm^2)/E_t(eV)/Nt (cm^{-3})	HTM/IF-2; 1×10^{-14}/0.8/1×10^{18}			ETM/IF-1 1×10^{-14}/0.8/1×10^{18}	

Table S3. Summary of simulation data and device parameters close to experimental trend of device parameters under degradation.

	Simulation conditions							Simulated device parameter			
	E_{t1} (eV)	N_{t1} (cm^{-3})	E_{t2} (eV)	N_{t2} (cm^{-3})	IF (N_t) (cm^{-3})	E_{t3} (eV)	N_{t3} (cm^{-3})	V_{OC} (V)	J_{sc} ($mAcm^{-2}$)	FF (%)	η (%)
1	0.35	8.E14	0.25	2E15	1E18			0.96	20.59	77.09	15.33
2	0.65	2E15	0.3	3E15	5E18			0.93	19.74	72.89	13.4
3	0.9	3E15	0.6	5E15	5.E19			0.86	17.83	62.83	9.73
4	0.9	8E15	0.6	1E16	1E20	0.4	5E15	0.81	15.87	53.54	6.96

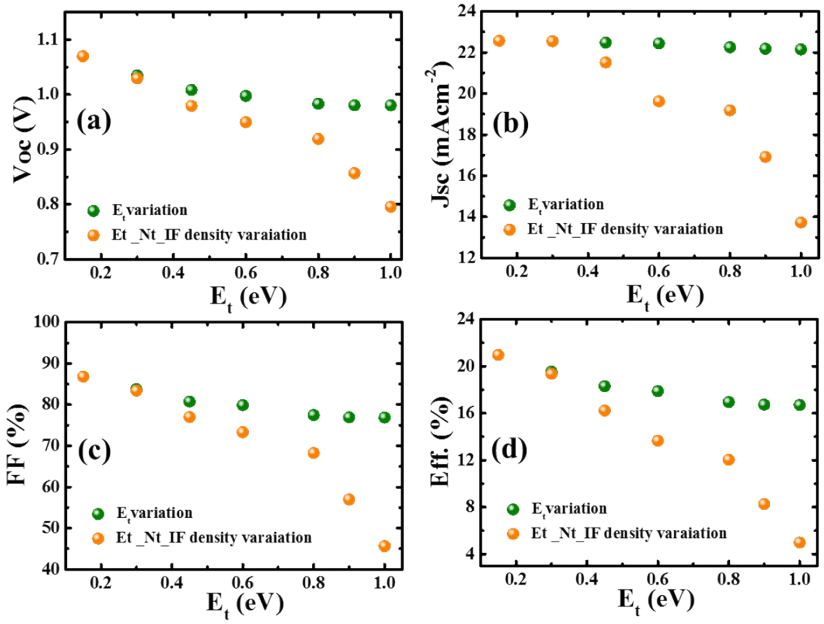

Figure S11. The plots of simulated device parameters with (1) variation of defect level (E_t) only and (2) variation of defect level (E_t), defect density (N_t) and interface quality (IF) simultaneously. As we consider only deep defect level, the impact on device parameters is not so dominant while later case revealed strong dominancy. These results implicate that the degraded device parameters must have collectively suffered from deterioration of bulk defect density, defect level and interface layer quality.

References

[1] Khadka, D. B.; Shirai, Y.; Yanagida, M.; Masuda, T.; Miyano, K. Enhancement in Efficiency and Optoelectronic Quality of Perovskite thin films Following MACl Ambient Annealing, *Sustainable Energy and Fuels,* **2017**, Accepted

[2] Khadka, D. B.; Shirai, Y.; Yanagida, M.; Miyano, K. Exploring the Effect of Interfacial Transport Layer on Device Performance of Planar Perovskite Solar Cells by Optoelectronic Approach, *under revision.*

[3] T. Minemoto, M. Murata, Device Modeling of Perovskite Solar Cells Based on Structural Similarity with Thin Film Inorganic Semiconductor Solar Cells, *J. Appl. Phys.* **2014**, 116, 054505.