Supporting Information

Title
Novel Cathode Interfacial Layer using Creatine for Enhancing Photovoltaic Properties of Perovskite Solar Cell

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Contents

Experimental Section

a. Solar cell fabrication: FTO coated glasses were partially etched with 2 M HCl solution and zinc powder. Thereafter, the substrate was washed in the order of deionized (DI) water, ethanol, acetone, and isopropyl alcohol. The substrate is exposed to UV-Ozone for 15 min before depositing electron transport layer to eliminate organic residues and increase wettability. The 0.1M $SnCl_2$ solution ($SnCl_2 \cdot 2H_2O$ in ethanol) was spin-coated with the speed of 2000 rpm, 30s and then annealed at 200 ℃ for 30 min. The various concentration of creatine solution (0.1–1.0 wt% in DI water) is prepared and deposited onto SnO_2 layer with the speed of 5000 rpm for 60s. The film is annealed for 10min at 100 ℃. Before depositing perovskite solution, UV-Ozone treatment was conducted for another 15 min. The perovskite precursor solution was made following the previous report: 1M FAI, 1.1M PbI_2, 0.2M MABr, 0.22M $PbBr_2$ in DMF : DMSO = 4 : 1 (v : v) and 5% of CsI solution (1.5 M in DMSO), and spin-coated using consecutive steps: 1000 rpm for 10s and 6000 rpm for 20 s. The 500 µL of CB was dispensed on the spinning substrate 5 s before the end of the second program. The substrates were annealed at 100 °C for 1 h. The Spiro-OMeTAD solution was prepared with a composition of: 72.3 mg Spiro-OMeTAD, 27.8 µL tBP, 17.8 µL Li-TFSI (520 mg mL^{-1} in AN), and 2 mg FK209 in 1 mL CB spin-coated at 5000 rpm for 30 s. Finally, a 100 nm thickness of metal electrode (Ag/Au) was thermally deposited with the rate of 1 Å/s. In the case of $(FAPbI_3)_{0.95}(MAPbBr_3)_{0.05}$, we fabricated devices following the reported method.[1]

b. Device characterization: The *J-V* curve and maximum power point efficiency (MPP) was obtained using Keithley 2400 SMU and an Oriel xenon lamp (450 W) with an AM 1.5 filter. The solar cell was characterized under AM 1.5 G illumination of 100 mW cm^{-2} (Oriel 1 kW solar simulator), calibrated with a KG 5 filter (NREL certified). The *J-V* curves of all devices were measured by 0.02 V s^{-1} of scan rate with reverse (1.2 V to -0.2 V) or forward (-0.2 V to 1.2 V) bias. The device active area is 0.09 cm^2. The stabilized power output was measured under the maximum power point voltage.

c. IPCE Measurement: Constant 100 W Xenon lamp source with an automated monochromator filters and 0.76 mm x 1.0 mm rectangular spot size was used for incident-photon-to-current-efficiency (IPCE) spectra. The measurements were conducted in the wavelength range from 300 to 850 nm, chopped at 4 Hz (IQE-200B model).

d. PL Measurement: The TRPL and steady-state PL measurements were conducted using prepared samples: glass/creatine/perovskite or FTO/SnO_2/creatine/perovskite. The samples were excited from the glass side under ambient conditions with excitation wavelength of 474nm. Time-resolved photoluminescence (TRPL) was measured using time correlated single photon counting (TCSPC) system (HAMAMATSU/C11367-31). For TRPL measurements, a pulsed laser source was laser diode with a wavelength of 474 nm, a repetition rate of 100 kHz, fluence of ~ 4 nJ/cm^2 and a pulse width of 70 ps. Steady state photoluminescence (PL) was measured using the high resolution monochromator and hybrid photomultiplier detector (PMA Hybrid 40, PicoQuant GmbH).

e. SEM measurement: Field emission scanning electron microscope (FE-SEM, Hitachi S 4800) was employed to obtain the optical top and cross-sectional images.

f. UPS measurement: The UPS measurements were carried out using AXIS-NOVA and Ultra DLD (KRATOS Inc.) Mono-chromatic He I (21.22 eV) for UPS base pressure 2.0 x 10^{-9} Torr without surface treatment.

g. SCLC measurement: The device structure of FTO/SnO_2/creatine/perovskite/PCBM/Ag was measured using a Keithely 2400 SMU (0 to 5 V) to evaluate the trap densities (n_t) of devices following the equation ($V_{TFL} = \frac{en_t d^2}{2\varepsilon\varepsilon_0}$). The film thickness is around 400 nm, and the dielectric constant comes from reference 33.

h. Impedance measurement: The impedance of PSCs was measured using a computer-controlled potentiostat (SP-200, BioLogic). For FTO/SnO_2/creatine/perovskite/Au devices, EIS plot was measured in the dark condition under a bias of V_{OC}.

Reference

1. E. H. Jung, N. J. Jeon, E. Y. Park, C. S. Moon, T. J. Shin, T.-Y. Yang, J. H. Noh, J. Seo, *Nature*, 2019, **567**, 511.

Supplementary Figures and Tables

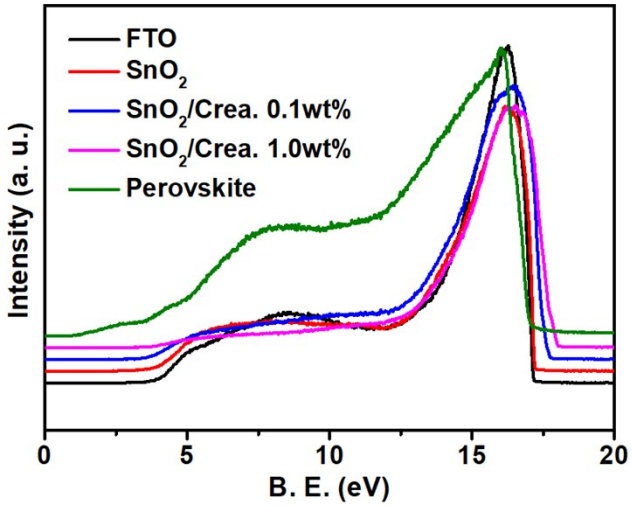

Fig. S1. Ultraviolet photoelectron spectroscopy results

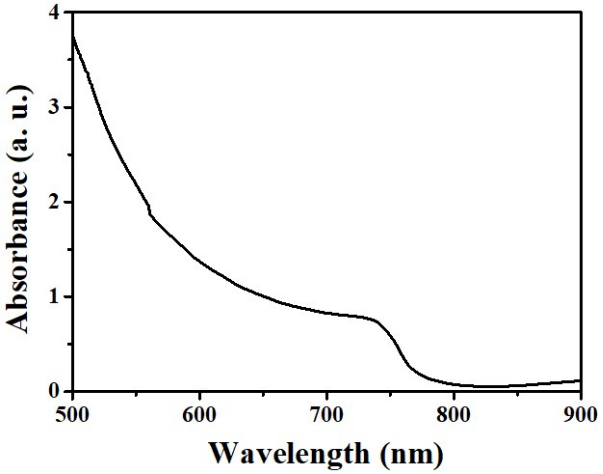

Fig. S2. Absorbance specrum of perovskite layer.

Fig. S3. Solubility tests of creatine in dimethyl formamide (DMF) and dimethyl sulfoxide (DMSO)

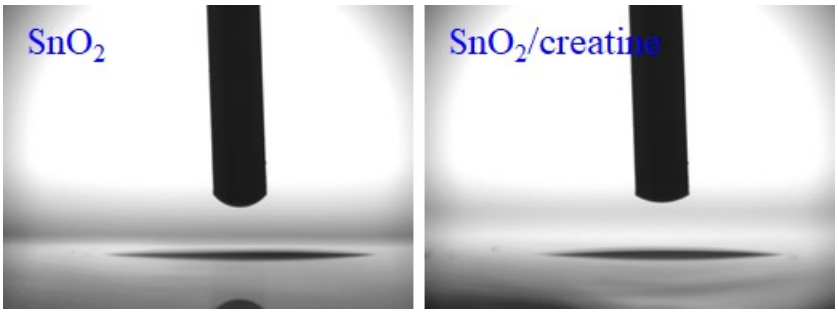

Fig. S4. Contact angles of DMF:DMSO=8:2 solution on the SnO_2 and SnO_2/creatine layers.

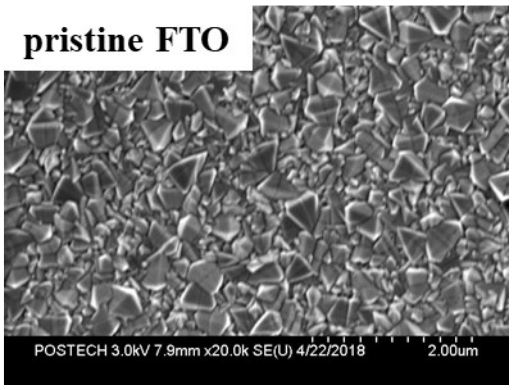

Fig. S5. Top scanning electron microscopy (SEM) image of fluorine-doped tin oxide (FTO).

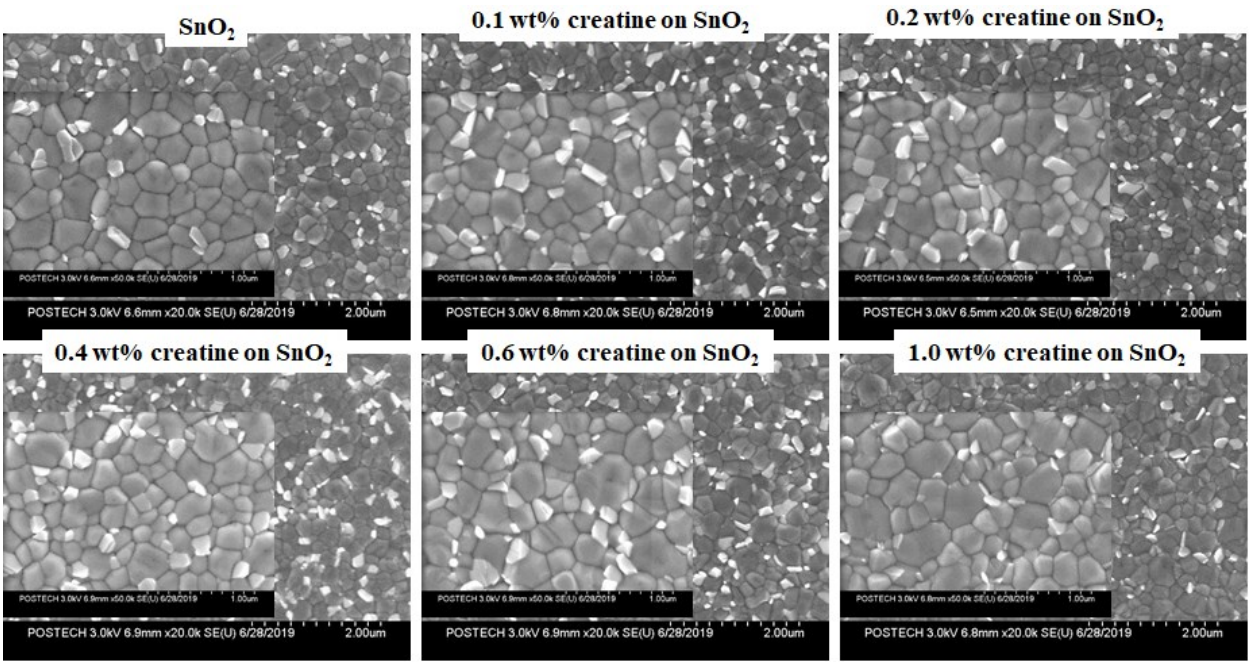

Fig. S6. Top SEM images of FTO/SnO_2/creatine 0.1–1.0 wt%/perovskite.

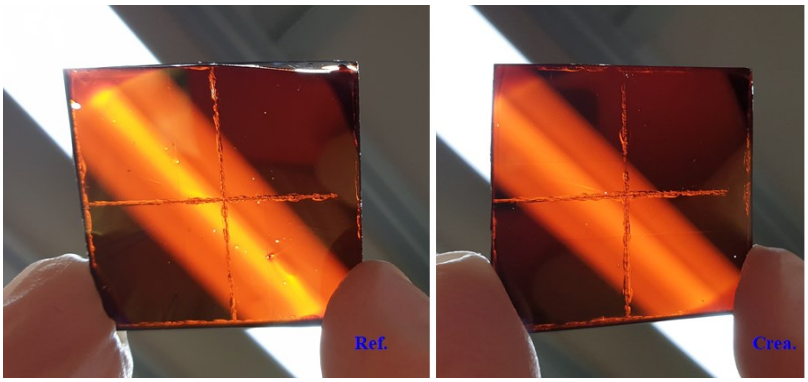

Fig. S7. Optical images of pristine and creatine-based perovskite layers.

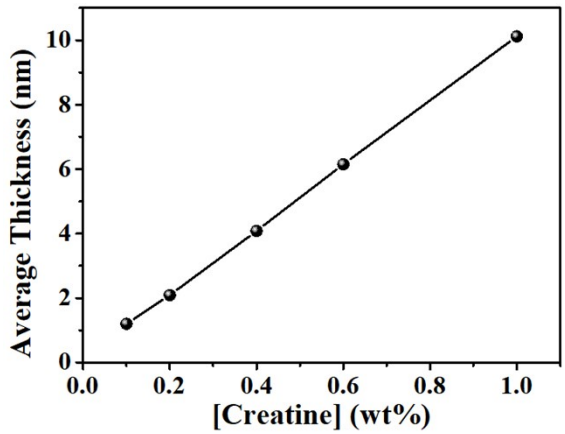

Fig. S8. Thickness change as a function of the creatine precursor concentration.

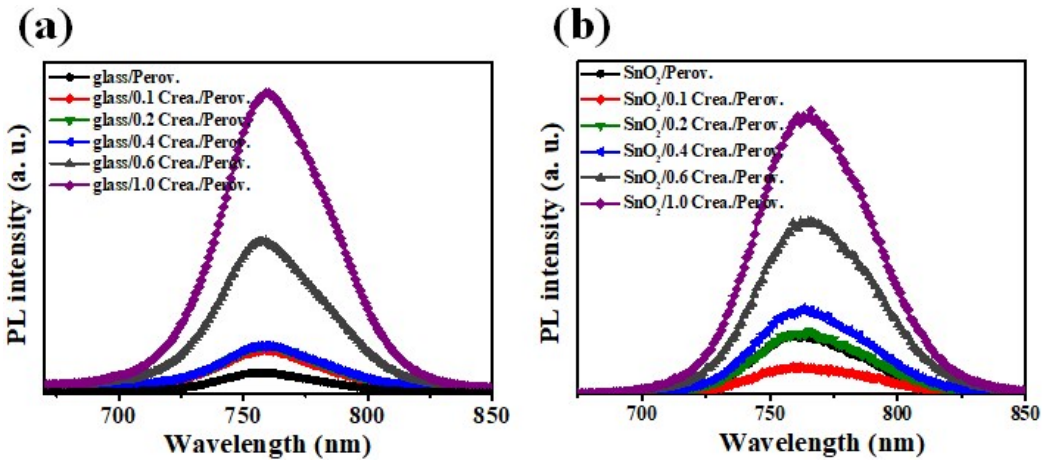

Fig. S9. Steady state photoluminescence (PL) of (a) glass/creatine/perovskite and (b) glass/SnO_2/creatine/perovskite.

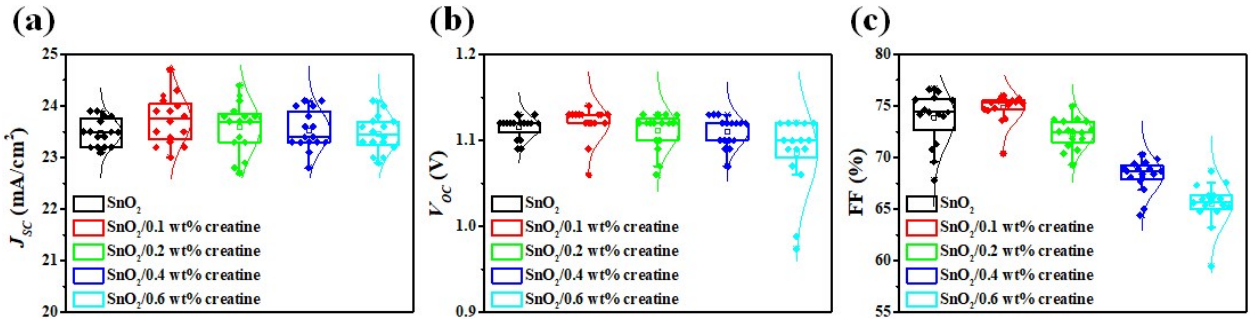

Fig. S10. Histograms of creatine devices for optimization: (a) short circuit current (J_{SC}), (b) open circuit voltage (V_{OC}), and fill factor (FF).

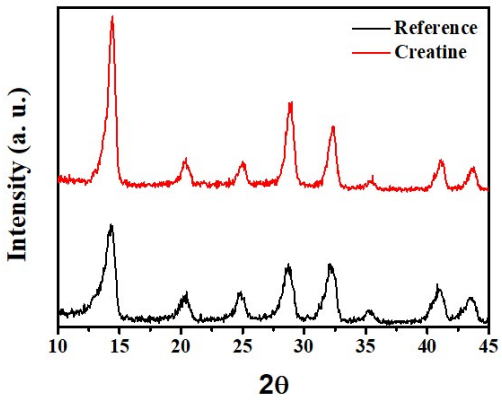

Fig. S11. X-ray diffraction results of glass/perovskite and glass/creatine/perovskite.

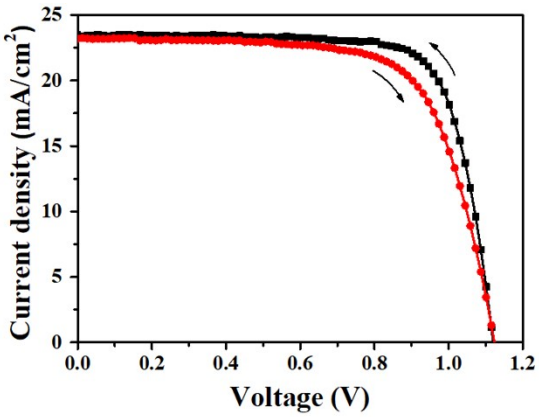

Fig. S12. Hysteresis of reference solar cell.

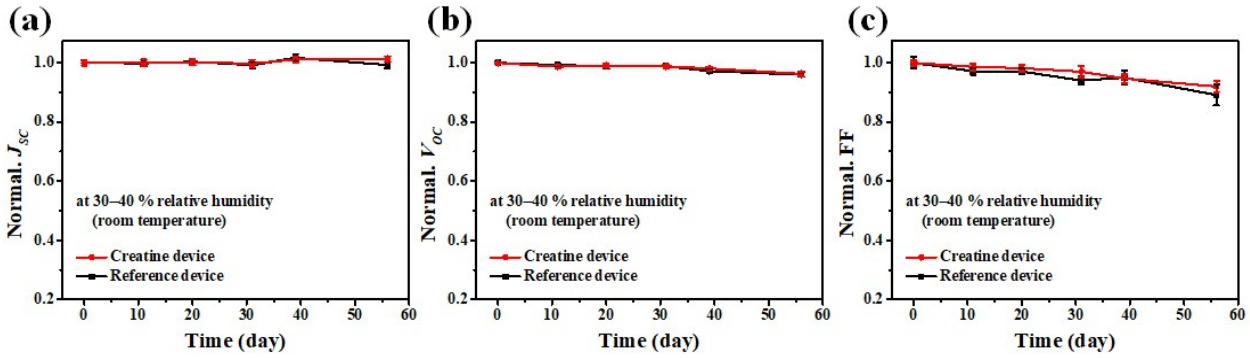

Fig. S13. Stability test at the ambient condition and its parameter changes: (a) J_{SC}, (b) V_{OC}, and (c) FF.

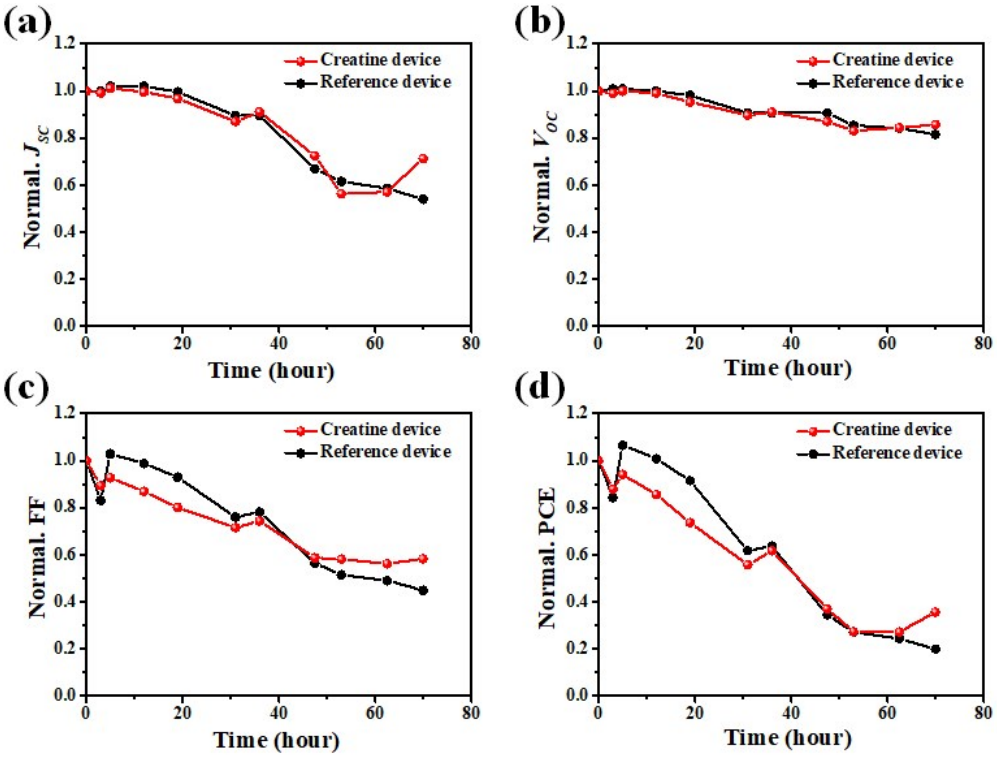

Fig. S14. Stability test at the thermal condition (85 °C) and its parameter changes: (a) J_{SC}, (b) V_{OC}, (c) FF, and (d) efficiency.

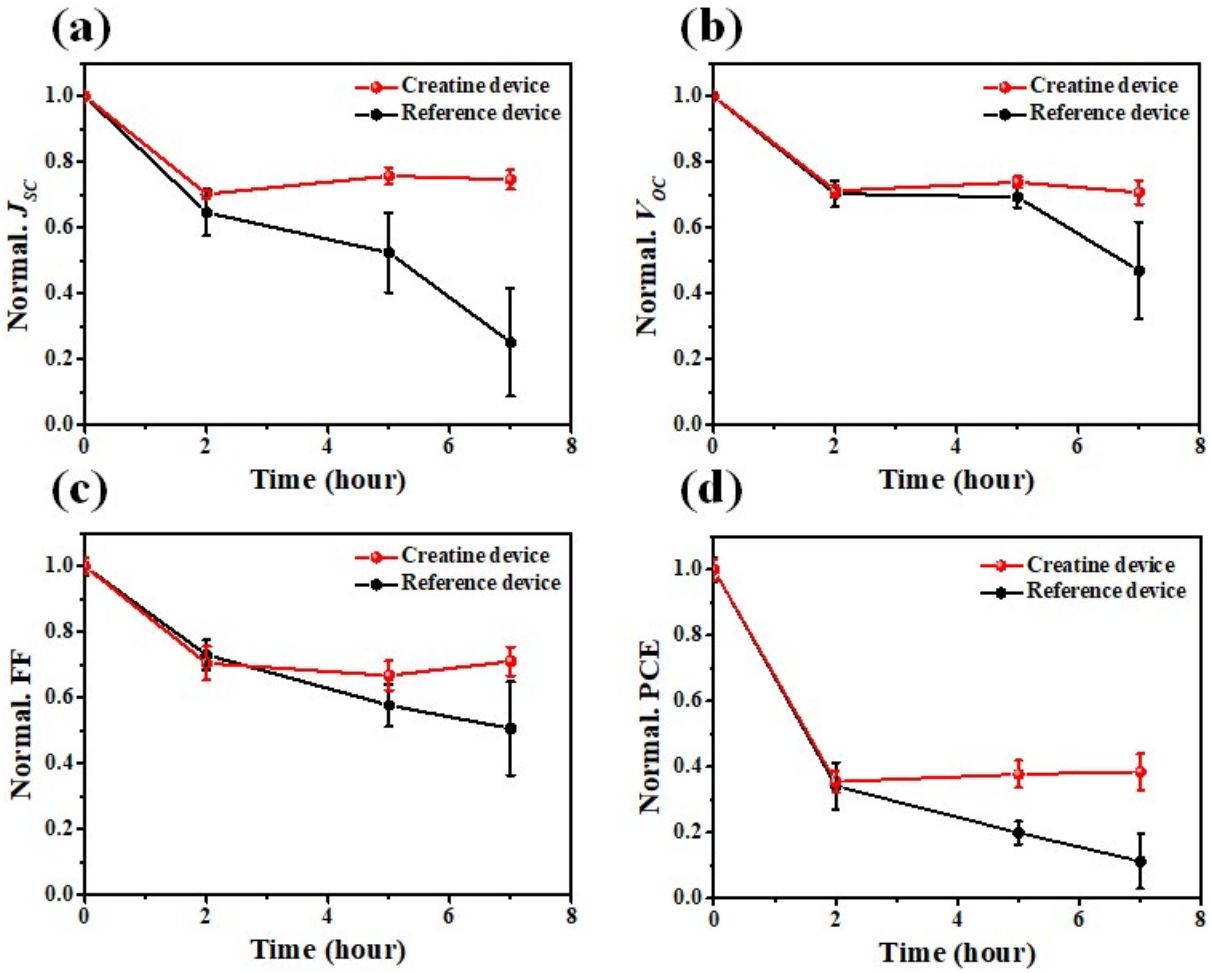

Fig. S15. Stability test at the thermal and humid condition (85 °C and 85 % of relative humidity) and its parameter changes: (a) J_{SC}, (b) V_{OC}, (c) FF, and (d) efficiency

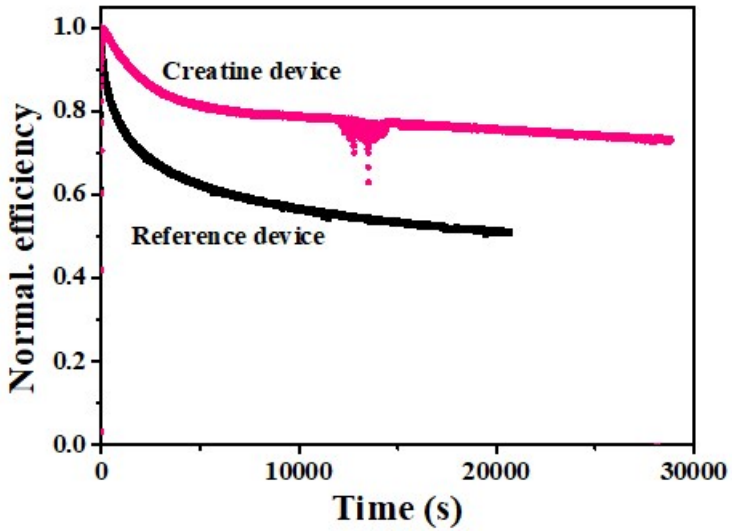

Fig. S16. Stability test at the ambient condition under 1 sun illumination (maximum power point tracking)

Table S1 Creatine thickness reliability data

Concentration (wt%)	1[a)] (nm)	2[a)] (nm)	3[a)] (nm)	4[a)] (nm)	5[a)] (nm)	Average (nm)	Standard deviation (nm)
0.1	1.181	1.200	1.200	1.204	1.210	1.199	0.0118
0.2	2.080	2.085	2.150	2.076	2.063	2.091	0.0340
0.4	4.076	4.073	4.080	4.084	4.092	4.081	0.0074
0.6	6.164	6.189	6.142	6.121	6.134	6.150	0.0268
1.0	10.09	10.11	10.14	10.14	10.14	10.12	0.0230

[a)] Samples were prepared on the silicon wafer and ellipsometer was used for measurement

Table S2 Trap filled limited (TFL) voltage of space charge limited current measurements.

Sample	V_{TFL} (V)
SnO_2	1.19
SnO_2/creatine 0.1 wt%	0.95
SnO_2/creatine 0.2 wt%	0.79
SnO_2/creatine 0.4 wt%	0.75
SnO_2/creatine 0.6 wt%	0.68
SnO_2/creatine 1.0 wt%	0.65

Table S3 Fitting parameters and average lifetime of time-resolved photoluminescence (TRPL).

	A_1	τ_1 (s)	A_2	τ_2 (s)	$\tau_{average}$[1] (s)
Glass	0.557	14.4	0.266	240	87.5
Creatine 0.1 wt%	0.430	18.2	0.370	326	160
Creatine 0.2 wt%	0.417	19.3	0.352	328	161
Creatine 0.4 wt%	0.416	19.8	0.416	322	171
Creatine 0.6 wt%	0.340	22.8	0.415	335	194
Creatine 1.0 wt%	0.348	28.0	0.388	346	196

[1] $\tau_{average} = \frac{A_1\tau_1 + A_2\tau_2}{A_1 + A_2}$

Table S4 Fitting parameters surface quencing TRPL.

	A_1	τ_1 (s)	A_2	τ_2 (s)
SnO_2	0.559	6.26	0.377	64.4
SnO_2/creatine 0.1 wt%	0.558	5.28	0.362	49.1
SnO_2/creatine 0.2 wt%	0.529	6.22	0.375	60.8
SnO_2/creatine 0.4 wt%	0.552	6.46	0.392	76.6
SnO_2/creatine 0.6 wt%	0.515	9.91	0.378	132
SnO_2/creatine 1.0 wt%	0.503	11.1	0.388	150