Supporting Information

Enhanced Long-term Stability of Perovskite Solar Cells by Passivating Grain Boundary with Polydimethylsiloxane (PDMS)

Woochul Kim[a], Jong Bae Park[b], Hyeonghun Kim[a], Kihyeun Kim[c], Jiyoon Park[a], Sungjun Cho[a], Heon Lee[d], Yusin Pak[a] and Gun Young Jung*[a]*

Affiliations

[a.] *School of Materials Science and Engineering, Gwangju Institute of Science and Technology (GIST), 123 Cheomdan-gwagiro, Buk-gu, Gwangju, 61005, Republic of Korea.*

Email: rhpys@gist.ac.kr, gyjung@gist.ac.kr

[b.] *Jeonju center, Korea Basic Science Institute (KBSI), Jeonju, Jeollabuk-do, 561180, Republic of Korea.*

[c.] *Department of Physics and Photon Science, Gwangju Institute of Science and Technology (GIST), 123 Cheomdan-gwagiro, Buk-gu, Gwangju, 61005, Republic of Korea.*

[d.] *Department of Materials Science and Engineering, Korea University, Seoul 02841, Republic of Korea.*

S1. The depth profiles of the perovskite films

The depth profiles along the surface of the bare perovskite film and the PDMS passivated-perovskite film (PDMS 0.03 wt %) were measured by tapping-mode atomic force microscope (AFM).

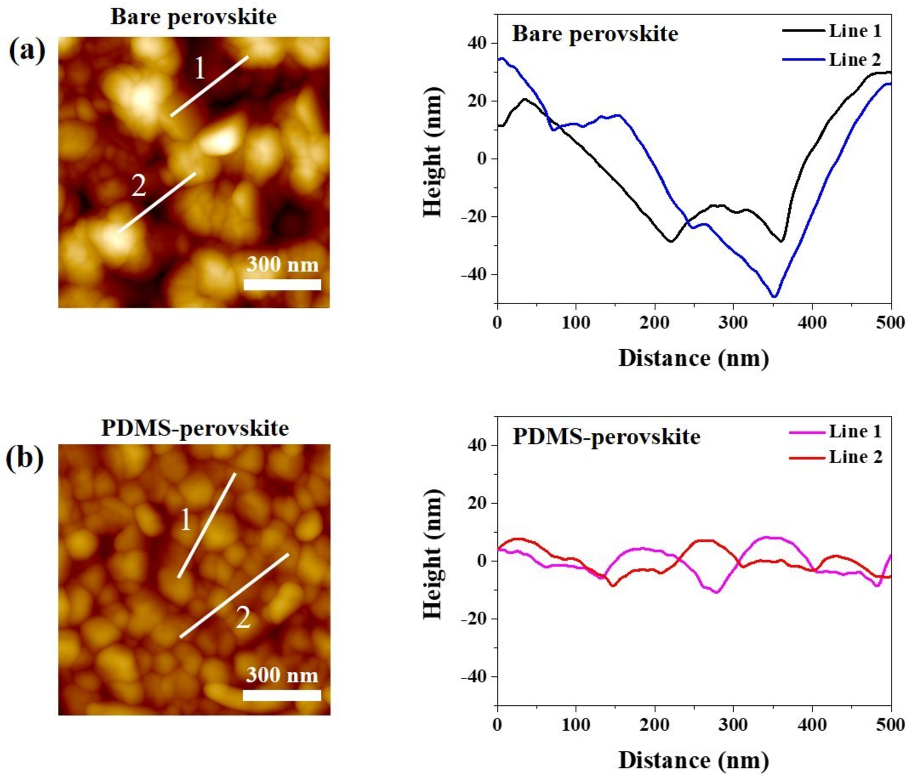

Fig. S1. Depth profiles of (a) the bare perovskite film and (b) the PDMS-perovskite film, measured along the profile line 1 and 2.

S2. The AFM images of perovskite films with PDMS concentrations

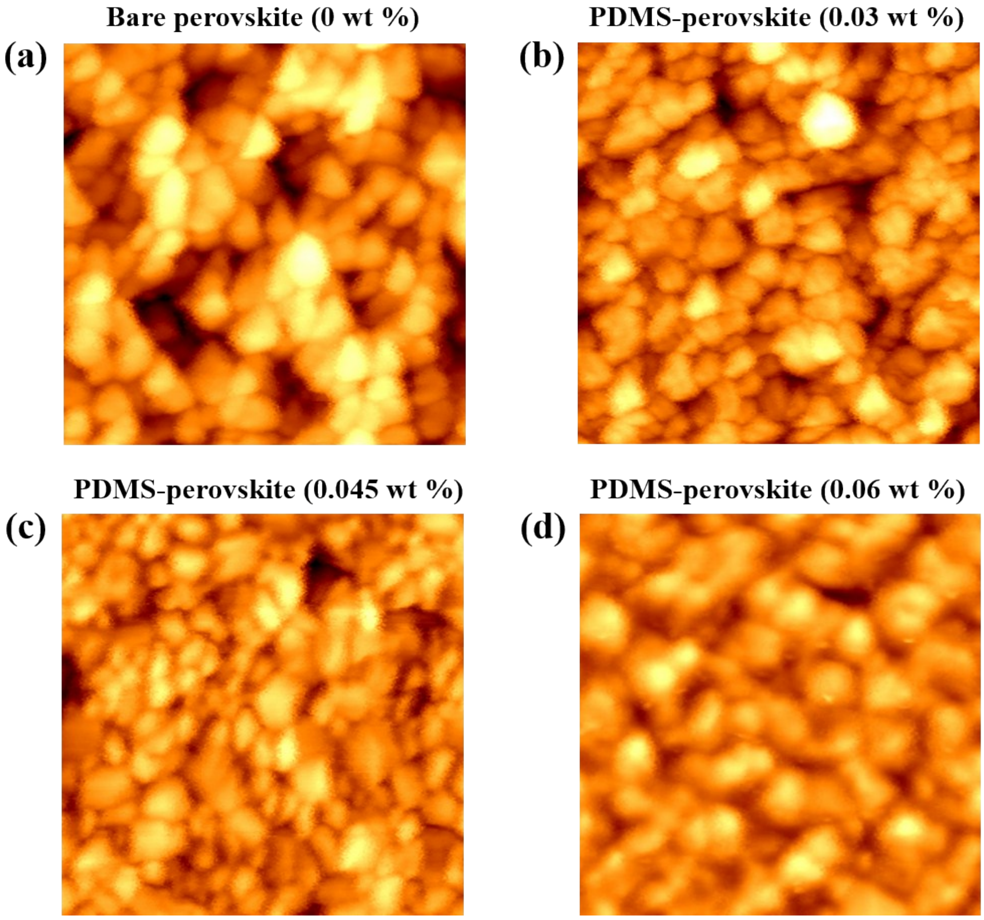

Fig. S2. The AFM images of perovskite films at PDMS concentrations of (a) 0, (b) 0.03, (c) 0.045, (d) 0.06 wt %.

S3. The XRD spectra of perovskite films with PDMS concentrations

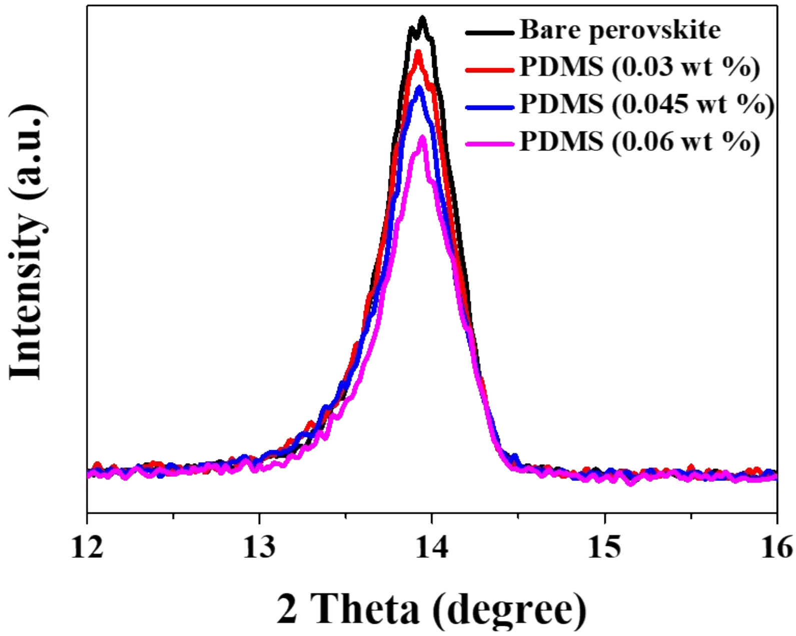

Fig. S3. The XRD spectra of perovskite films at PDMS concentrations (0, 0.03, 0.045, and 0.06 wt %)

S4. The PL spectra of bare perovskite film and PDMS-perovskite film

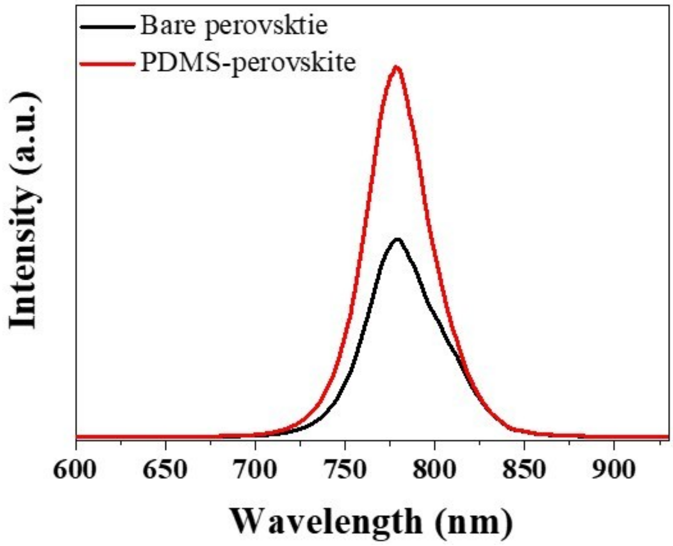

Fig. S4. The PL spectra acquired after preparing both perovskite films (bare and w/ PDMS additives) on a SiO_2 glass substrate.

S5. Equation of light intensity dependence of V_{oc} of solar cell devices

In the case of perfectly trap-free solar cell, the carrier recombination occurs only as a form of radiative recombination (Langevin recombination) and it has been shown that the open circuit voltage V_{oc} of the solar cell is given by the following equation,[1]

$$V_{oc} = \frac{E_{gap}}{q} - \frac{k_B T}{q} \ln\left[\frac{(1-P)\gamma N_C^2}{PG}\right] \tag{1}$$

where E_{gap} is the energy gap between the highest occupied molecular orbital (HOMO) and the highest occupied molecular orbital (LUMO), q is the elementary charge, k_B is the Boltzmann constant, T is the absolute temperature, P is the dissociation probability of electron hole pairs (EHPs) into the free carriers by thermal excitation, γ is the recombination constant, N_c is the density of states in the conduction band, and G is the generation rate of EHPs.

In the presence of the traps, the charge recombination includes both bimolecular and trap-assisted recombination and the recombination rate is presented as Equation 2 called Schokely-Read-Hall equation.[2,3]

$$R = \gamma np + \left[\frac{C_n C_p N_t (np - n_1 p_1)}{C_n(n+n_1) + C_p(p+p_1)}\right] \tag{2}$$

where, n and p are the electron density in the conduction band and hole density in the valence band, respectively, C_n and C_p are the capture coefficient of electrons and holes, respectively, N_t is the density of electron traps, p_1 and n_1 is the intrinsic carrier concentration in the sample.

In the equation 1, density of electron and hole has linear relationship to the light intensity (E_{gap} and N_c do not depend on the light intensity). We can modify the equation 1 and represent the V_{oc} in terms of the light intensity (I), as shown in Equation 3.

$$V_{oc} \propto \frac{k_B T}{q} \ln\left[\left(1 + \frac{C_n C_p N_t}{nC_n + pC_p}\right) I\right] \tag{3}$$

S6. Device stability with PDMS concentrations

The PSCs were stored in an illuminating laboratory space where the relative humidity is RH 70% without any protection or encapsulation for 1000 h. Three PDMS-PSCs (0.03, 0.045, and 0.06 wt %) maintained more than 85 % of their initial PCEs.

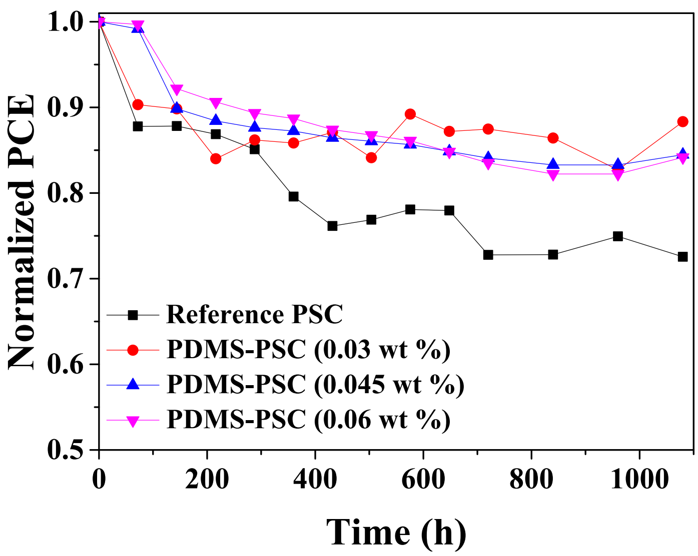

Fig. S6. Stability test of the reference PSC (w/o PDMS) and PDMS-PSCs (0.03, 0.045, and 0.06 wt %) for 1000 h, while being stored under a RH of 70 % and an illumination of yellow light.

S7. Observation of PSC devices until 180 days

The perovskite solar cells were stored in an illuminating laboratory space where the relative humidity is RH 70% without any protection or encapsulation until 180 days.

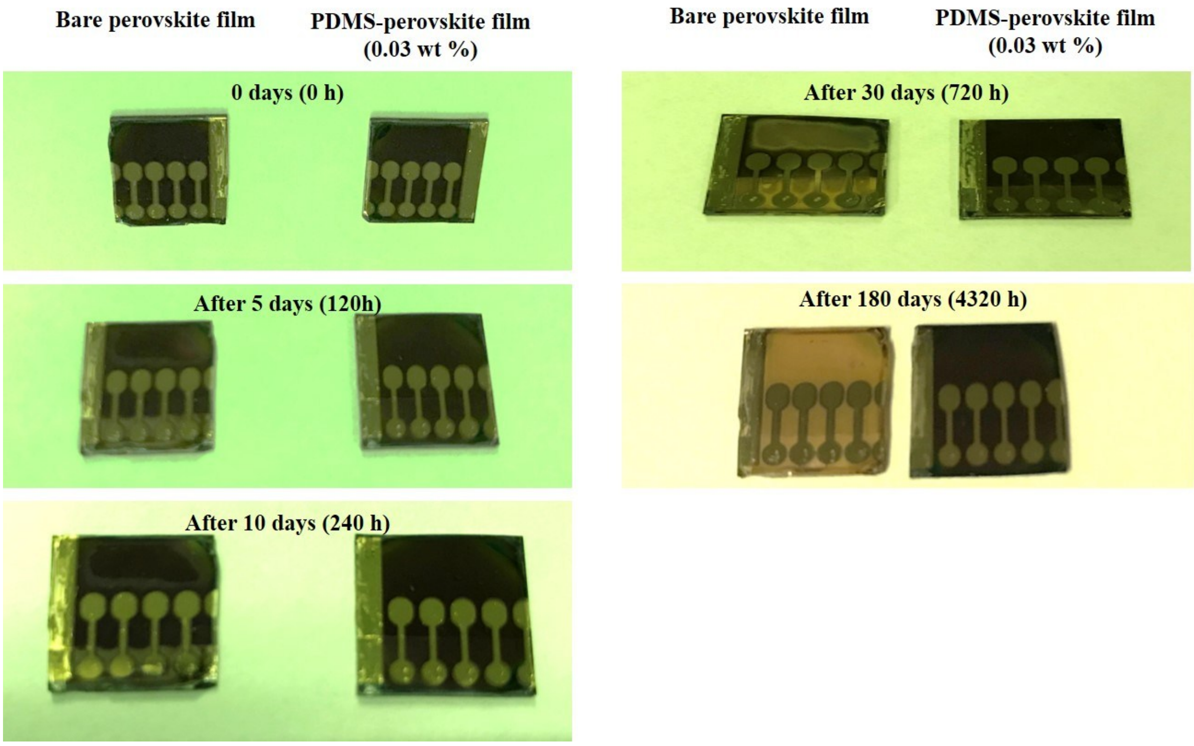

Fig. S7. Real surface photographs of both reference PSC and PDMS-PSC, taken at first day, 5 days, 10 days, 30 days, and 180 days after the fabrication.

References for supporting Information

1 L. J. A. Koster, V. D. Mihailetchi, R. Ramaker, P. W. M. Blom, *Appl. Phys. Lett.* 2005, **86**, 123509.
2 M. M. Mandoc, W. Veurman, L. J. A. Koster, B. Boer, P. W. M. Blom , *Adv. Funct. Mater.* 2007, **17**, 2167.
3 M. M. Mandoc, F. B. Kooistra, J. C. Hummelen, B. de Boer, P. W. Blom, *Appl. Phys. Lett.* 2007, **91**, 263505.