

Supplementary information

Mixed 2D-Cation Passivation towards Improved Durability of Perovskite Solar Cells and Dynamics of 2D-perovskite under Light irradiation and High temperature

Santa Mondal^{1†}, Naoto Eguchi^{1†}, Naoyuki Nishimura¹, Yoyo Hinuma¹, Kohei Yamamoto¹,

Atsushi Kogo¹, Takuro N. Murakami^{1*}, and Hiroyuki Kanda^{1*}

¹National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8565, Japan.

Table S1. Parameters of fitted result for TRPL measurement.

	τ_1 (ns)	τ_2 (ns)	A_1	A_2
BAI-OAI	9.5	99.6	0.592	0.264
BAI	6.5	87.2	0.595	0.262
OAI	6.5	80.6	0.621	0.273
Without	2.1	74.2	0.643	0.337

[†]The equation for the fitting is $y=y_0+A_1 \times \exp(-(x-x_0)/\tau_a)+A_2 \times \exp(-(x-x_0)/\tau_b)$.

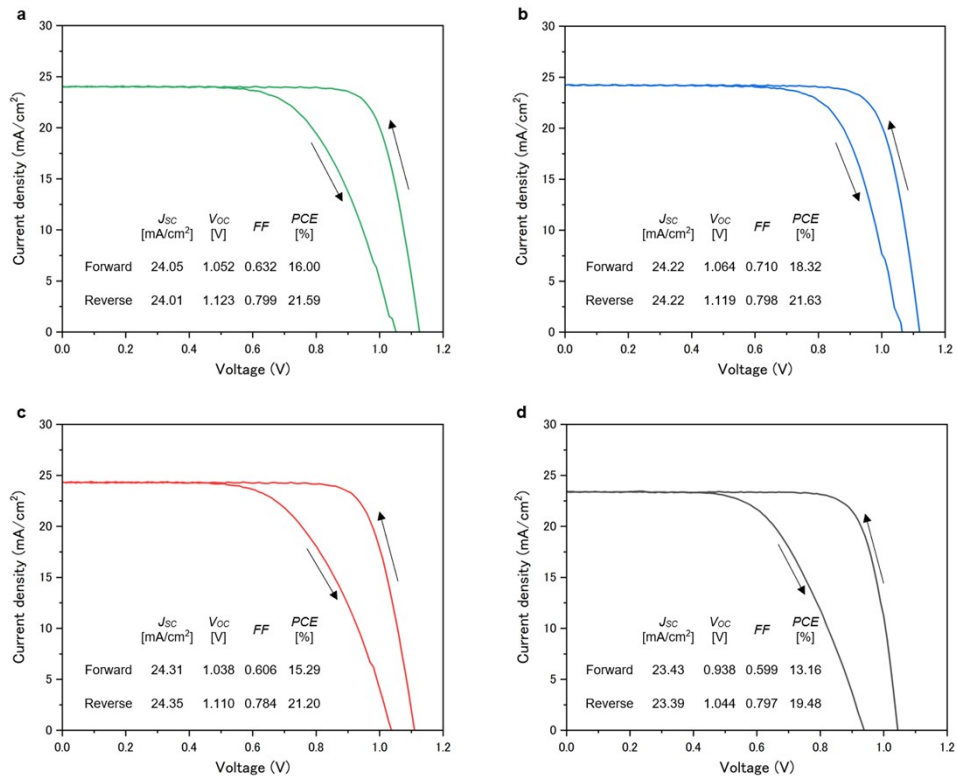


Figure S1. Hysteresis I-V curves of perovskite solar cells with (a) 2D perovskite (BAI-OAI), (b) 2D perovskite (BAI), (c) 2D perovskite (OAI), and (d) without passivation.

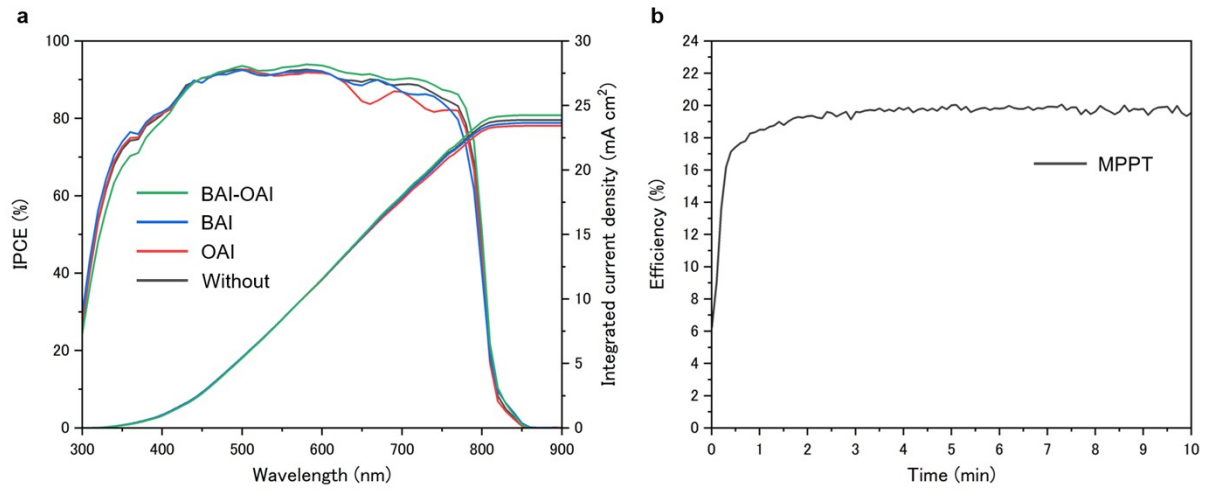


Figure S2. (a) IPCE spectra of each device. Green line, blue line, red line and black lines show 2D perovskite (BAI-OAI), 2D perovskite (BAI), 2D perovskite (OAI) and without passivation, respectively. (b) Maximum power point tracking for perovskite solar cells without passivation.

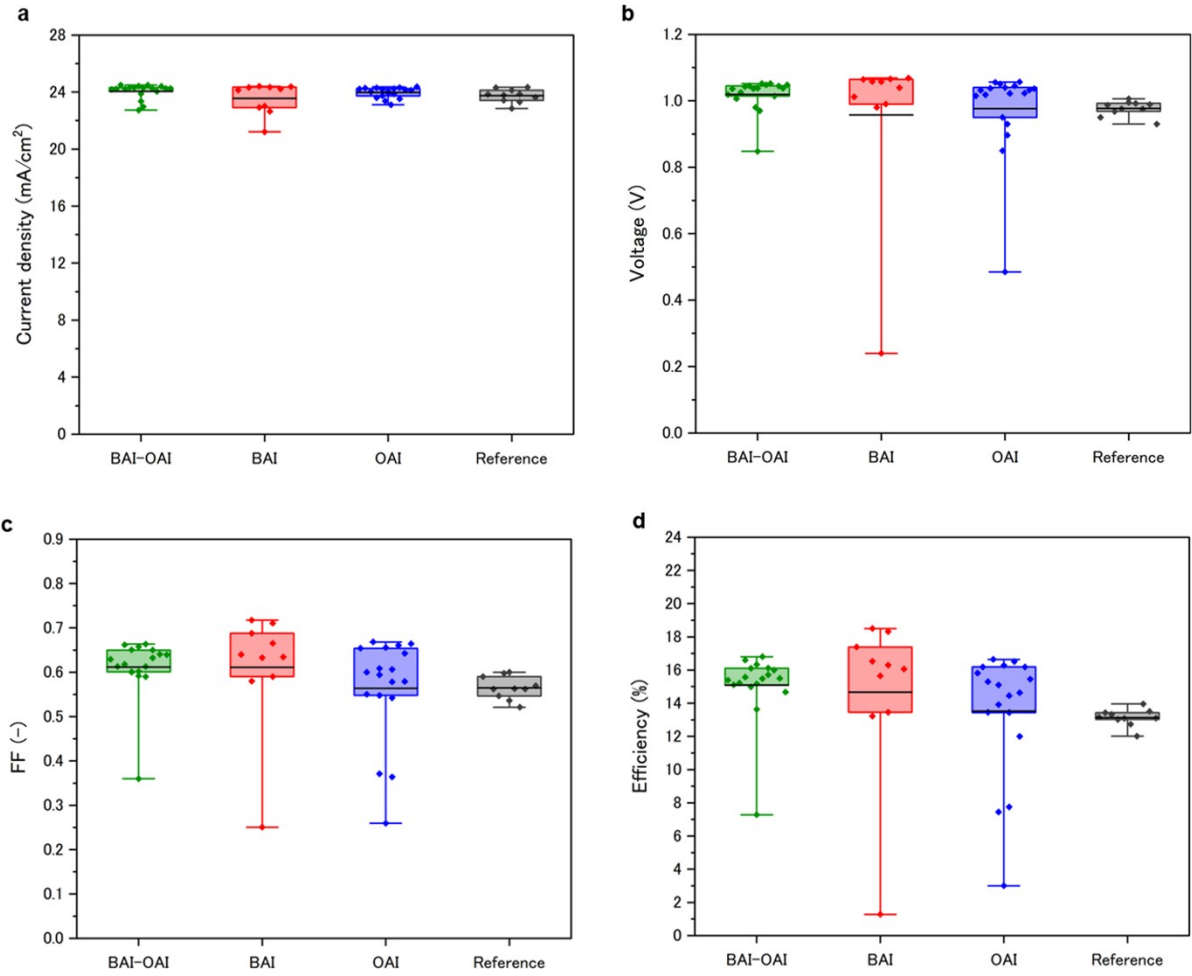


Figure S3. Boxplots of each solar cell parameters with forward scan for devices fabricated with different

material. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 50.

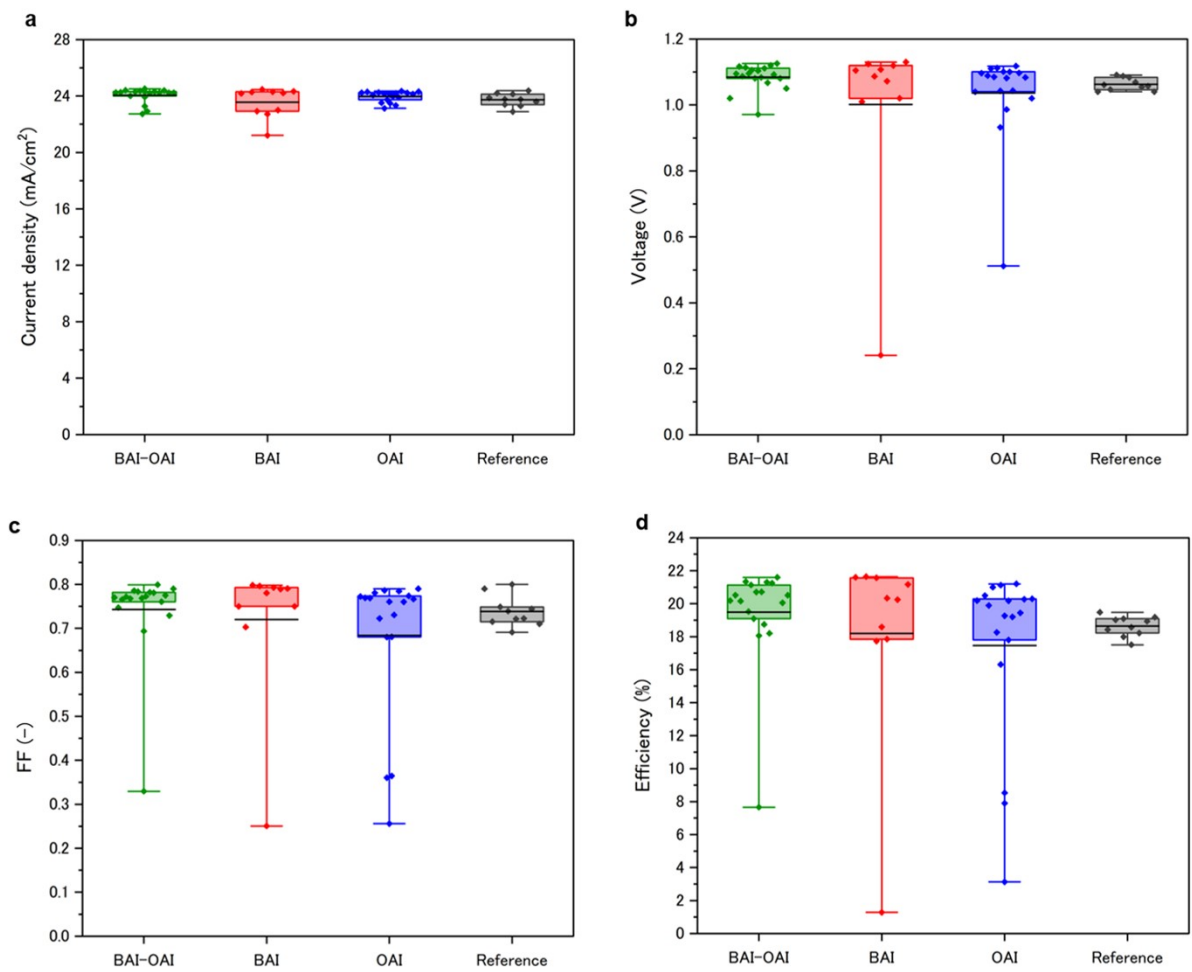


Figure S4. Boxplots of each solar cell parameter with reverse scan for devices fabricated with different material. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 50.

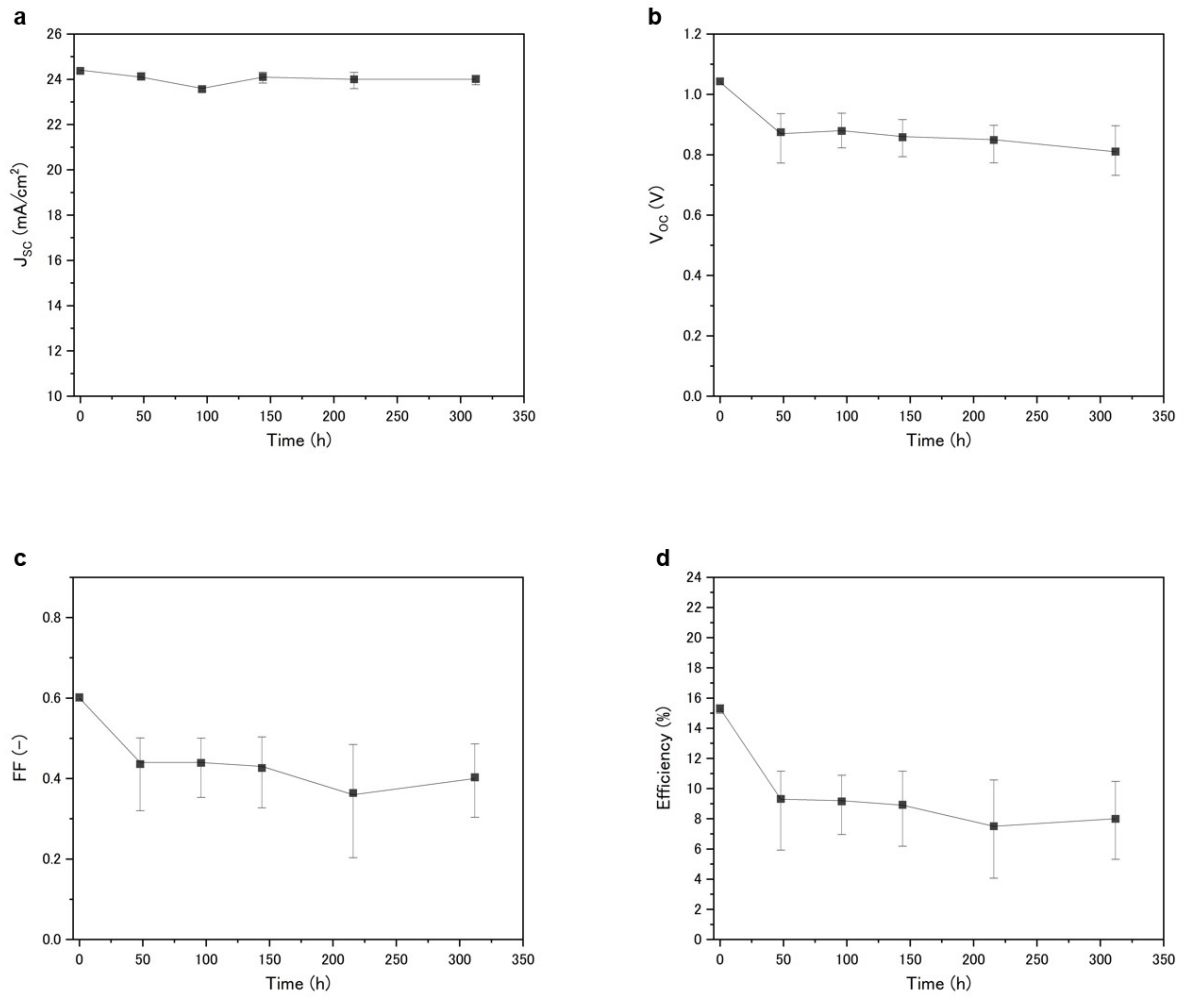


Figure S5. Stability test of devices passivated with 2D perovskite (BAI-OAI) under light irradiation for each solar cell parameter in forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

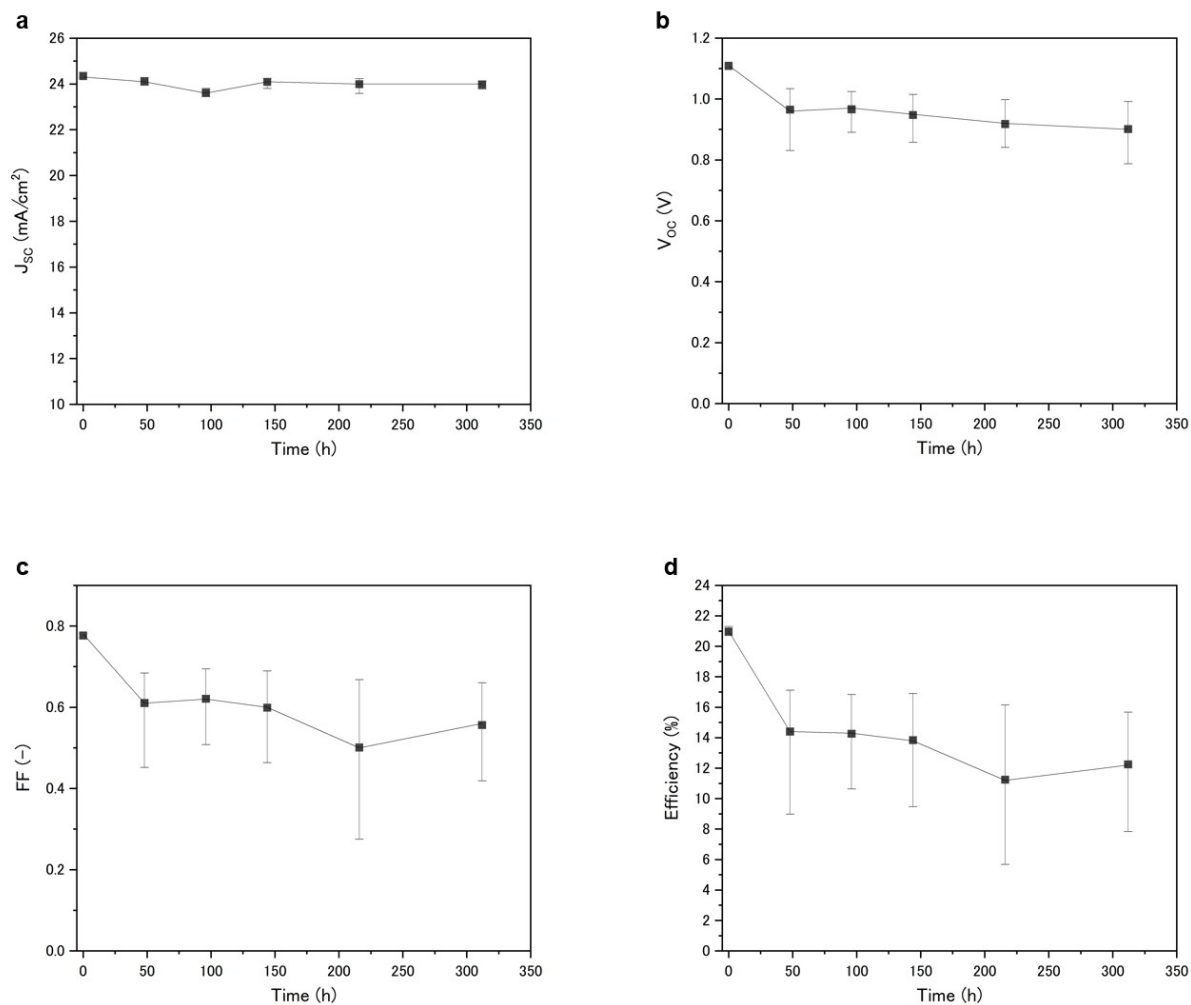


Figure S6. Stability test of devices passivated with 2D perovskite (BAI-OAI) under light irradiation for each solar cell parameter in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

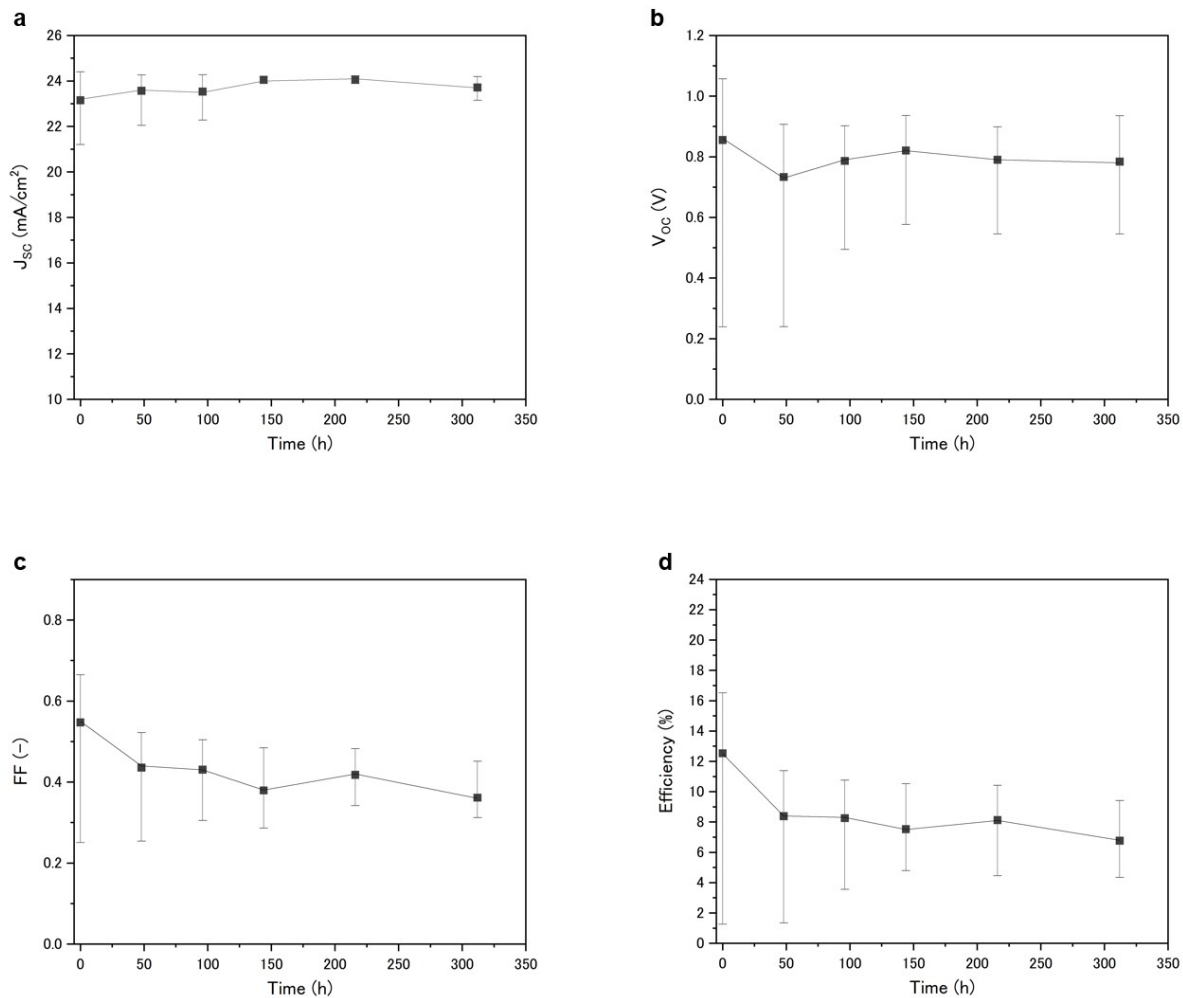


Figure S7. Stability test of devices passivated with 2D perovskite (BAI) under light irradiation for each solar cell parameter in forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

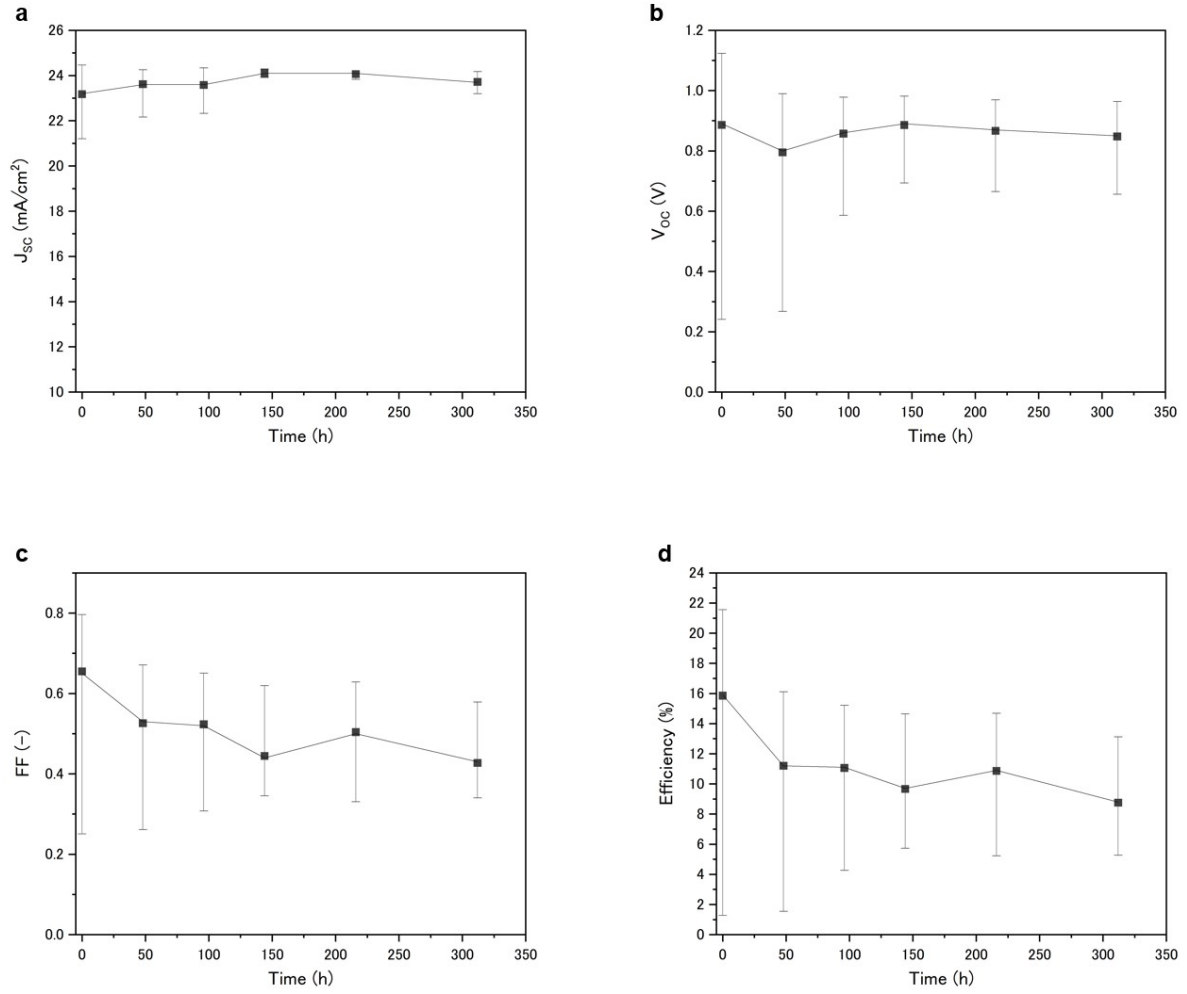


Figure S8. Stability test of devices passivated with 2D perovskite (BAI) under light irradiation for each solar cell parameter in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

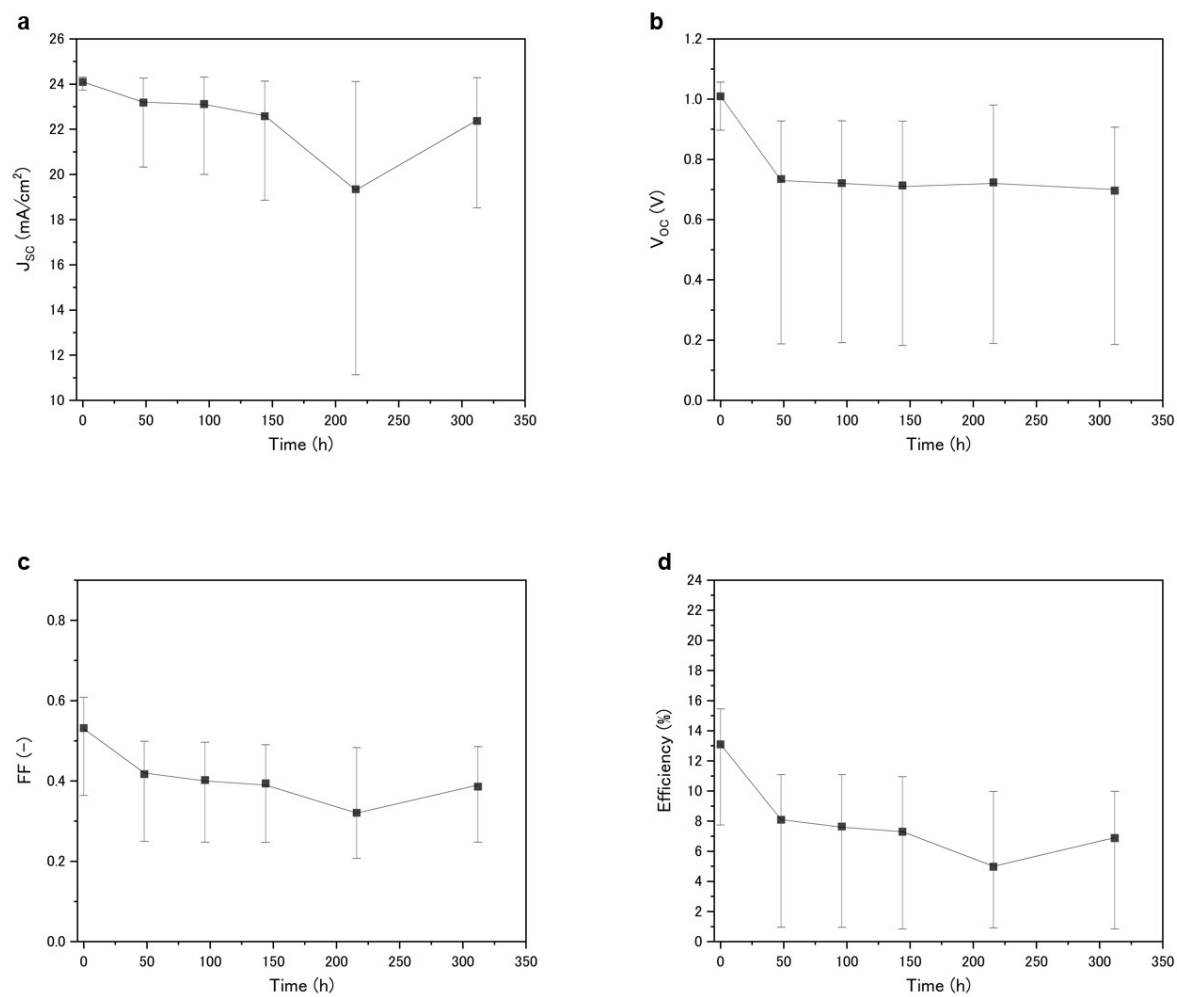


Figure S9. Stability test of devices passivated with 2D perovskite (OAI) under light irradiation for each solar cell parameter in forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

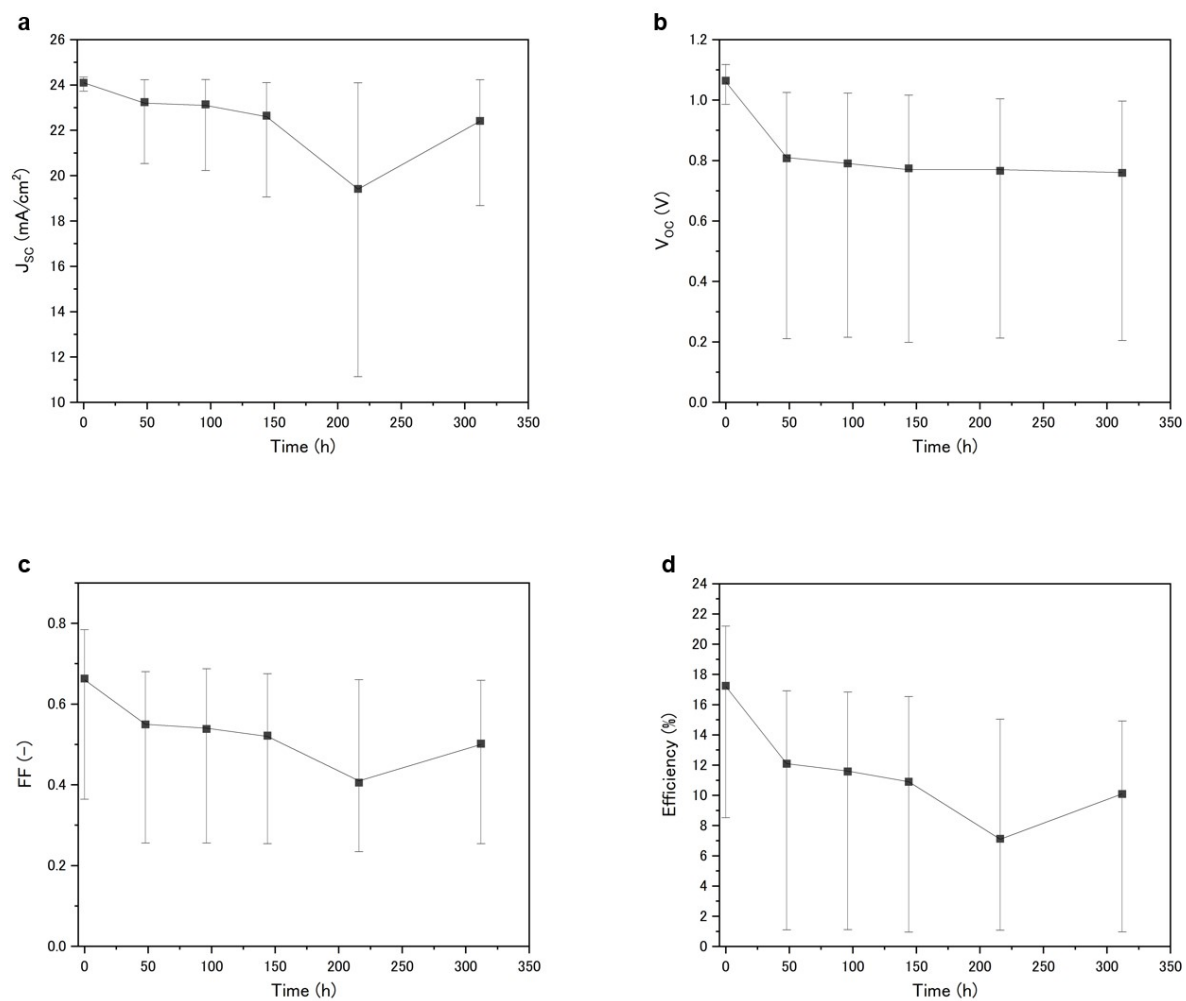


Figure S10. Stability test of devices passivated with 2D perovskite (OAI) under light irradiation for each solar cell parameter in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

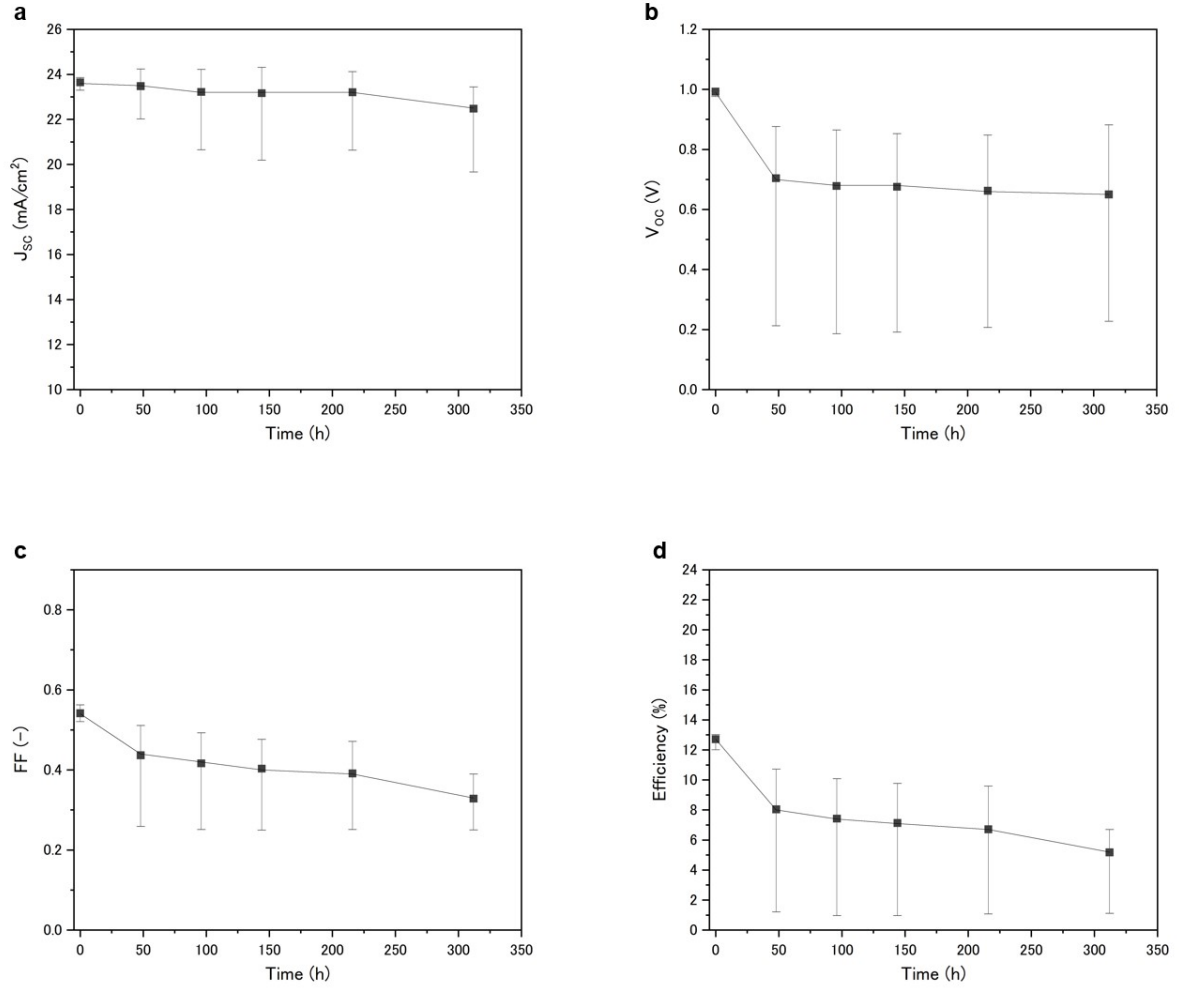


Figure S11. Stability test of devices without passivation under light irradiation for each solar cell parameter

in forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

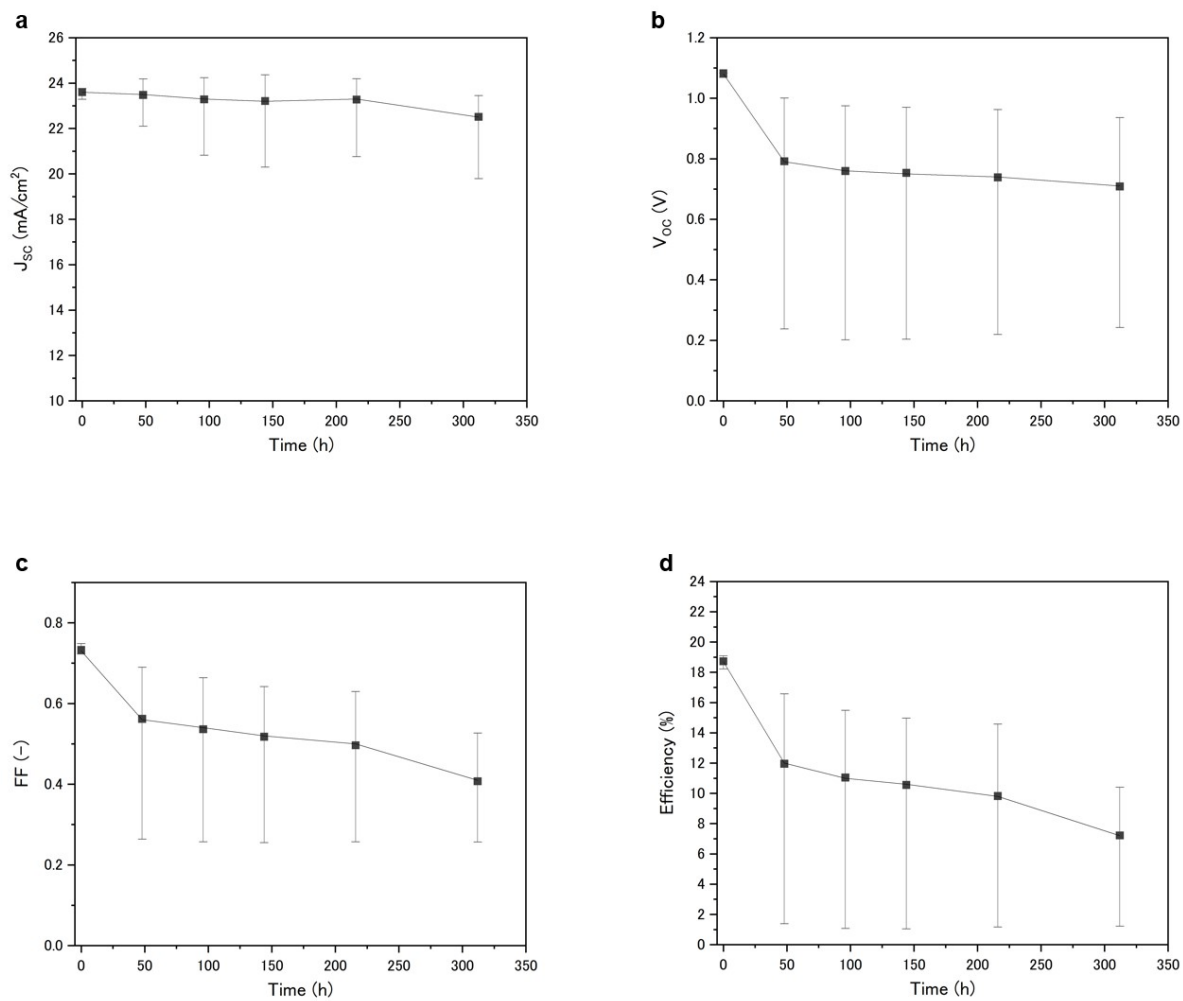


Figure S12. Stability test of devices without passivation under light irradiation for each solar cell parameter

in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

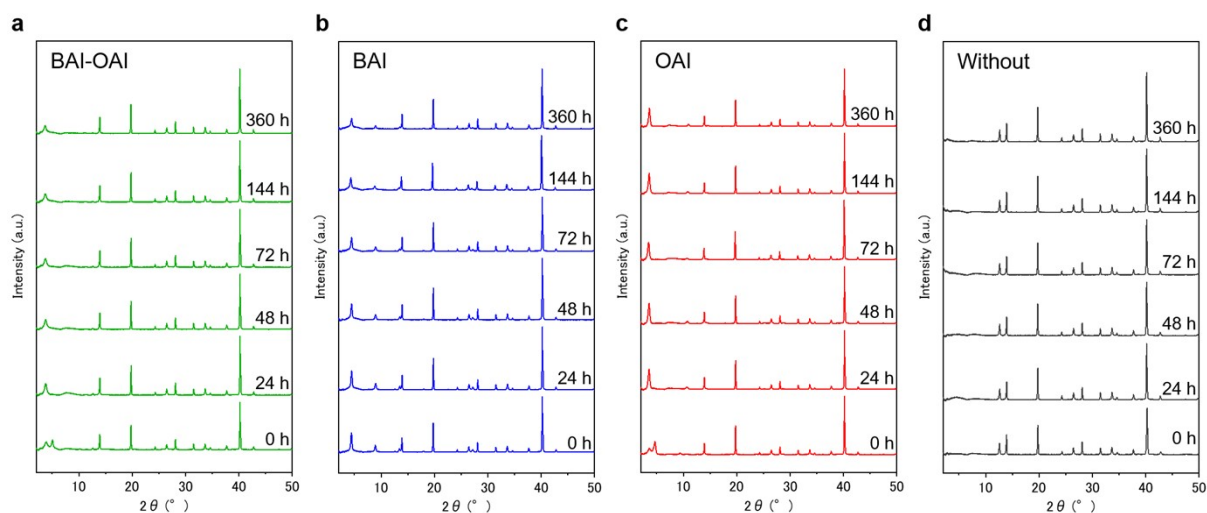


Figure S13. XRD spectra as a function of time for perovskite films with (a) 2D perovskite (BAI-OAI), (b) 2D perovskite (BAI), (c) 2D perovskite (OAI), and (d) without passivation on the perovskite films. Samples were stored at dry room dark condition.

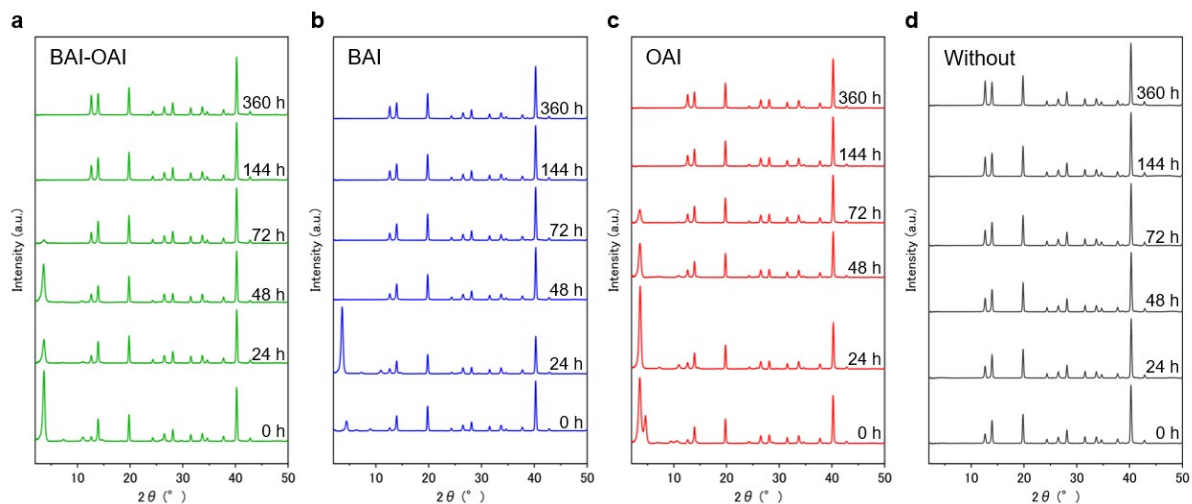


Figure S14. Full XRD spectra of perovskite layers with different passivation materials. Devices were heated under 65 °C. (a) 2D perovskite (BAI-OAI), (b) 2D perovskite (BAI), (c) 2D perovskite (OAI) and (d) without passivation.

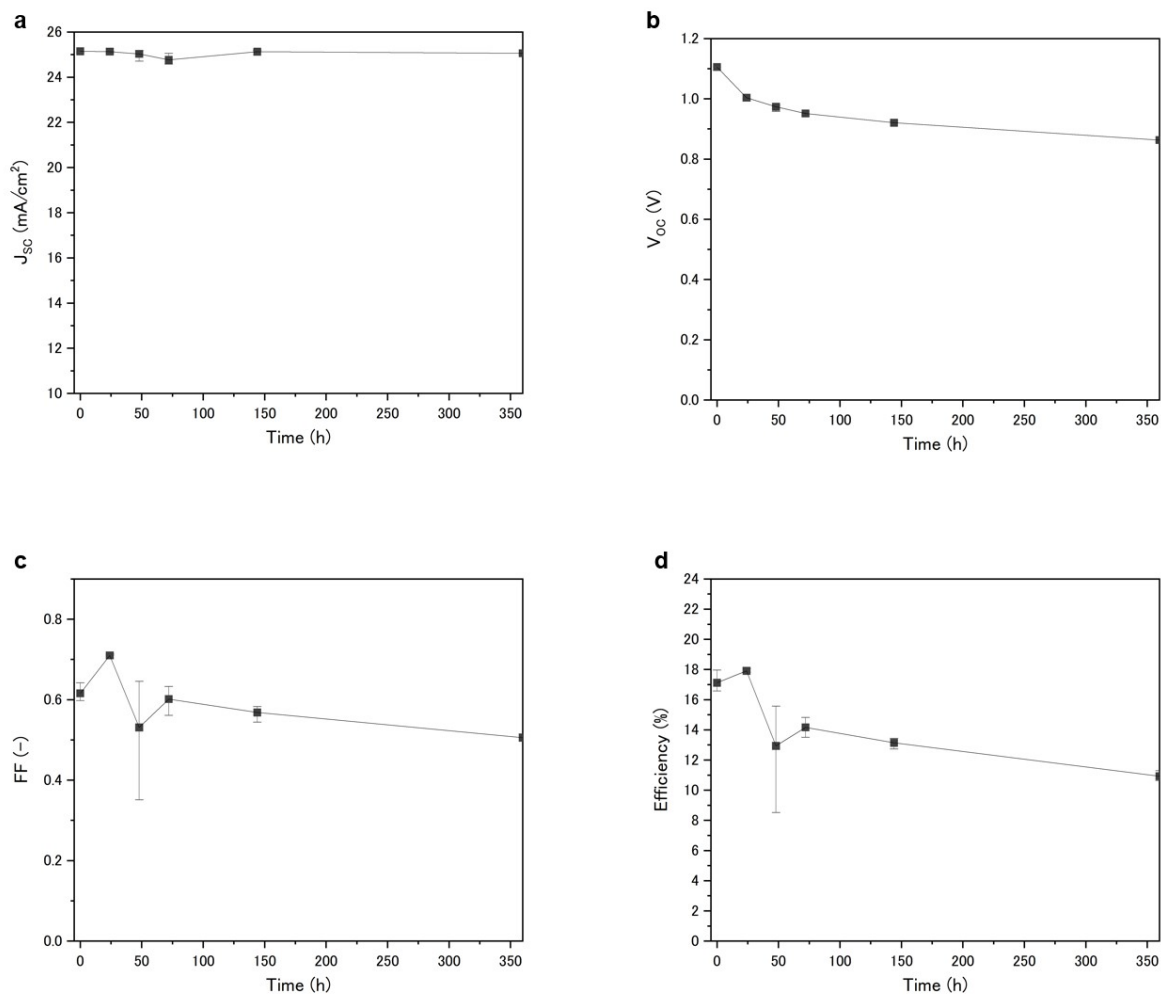


Figure S15. Stability test of devices passivated with 2D perovskite (BAI-OAI) under 65°C in dark for each solar cell parameter in forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

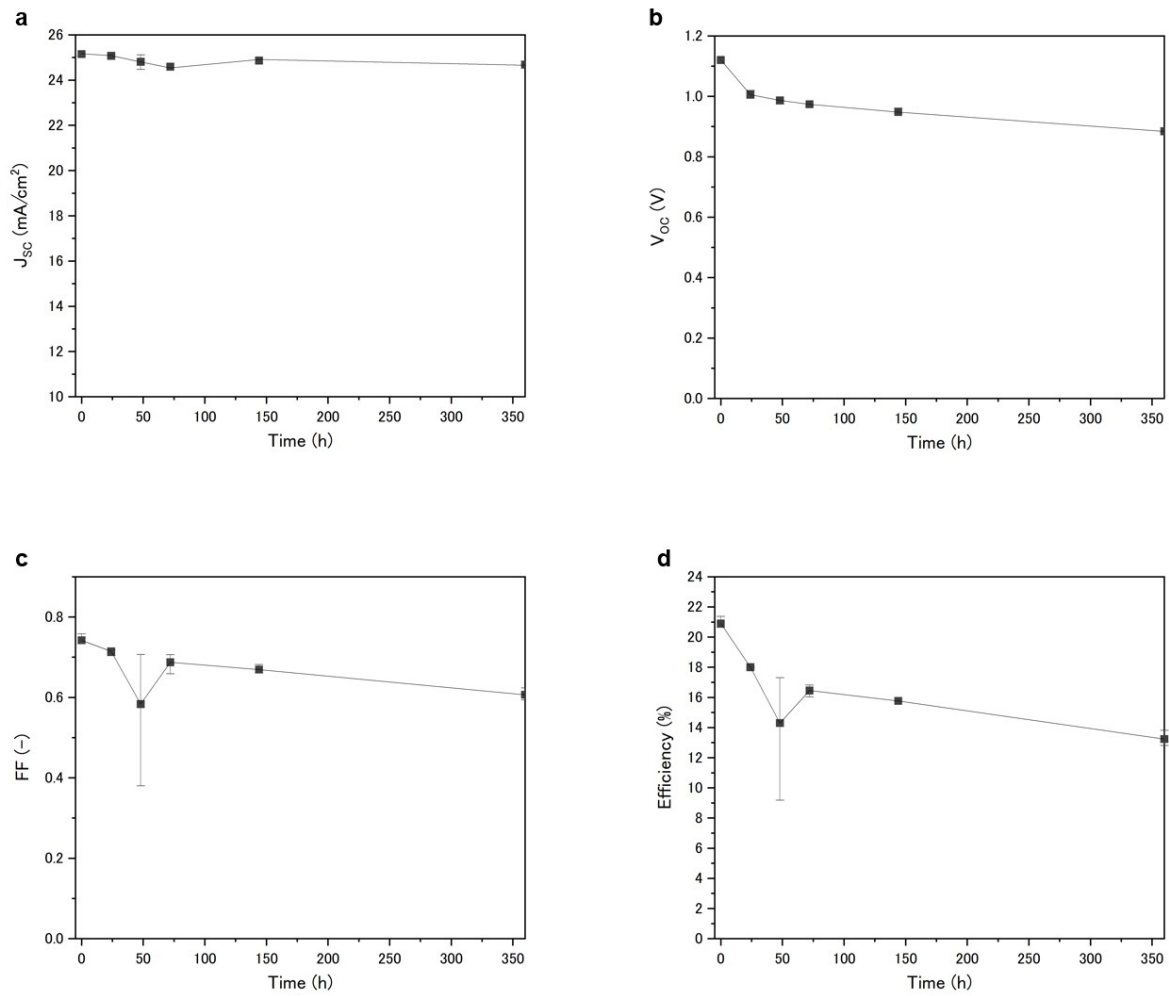


Figure S16. Stability test of devices passivated with 2D perovskite (BAI-OAI) under 65°C in dark for each solar cell parameter in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

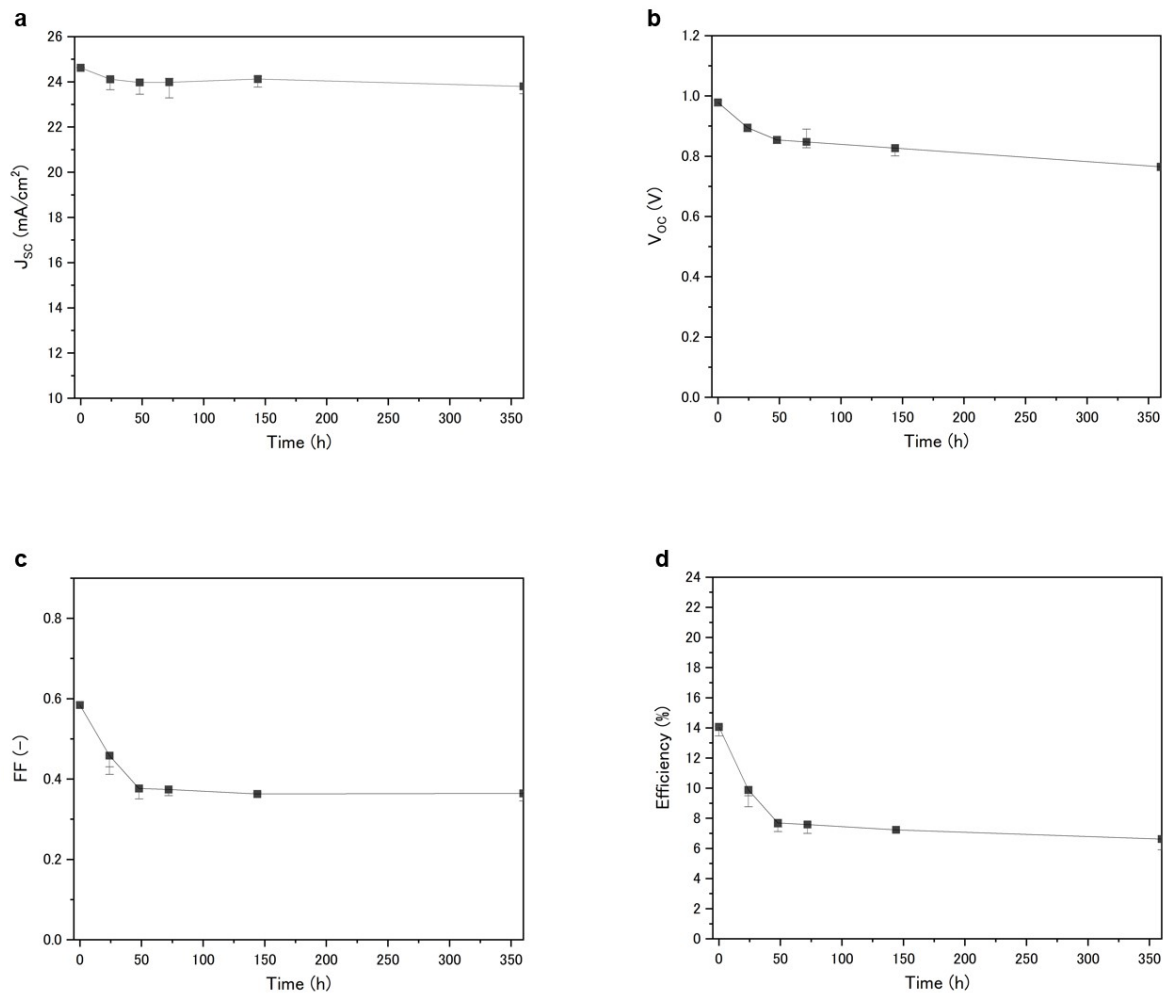


Figure S17. Stability test of devices passivated with 2D perovskite (BAI) under 65°C in dark for each solar cell parameter in forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

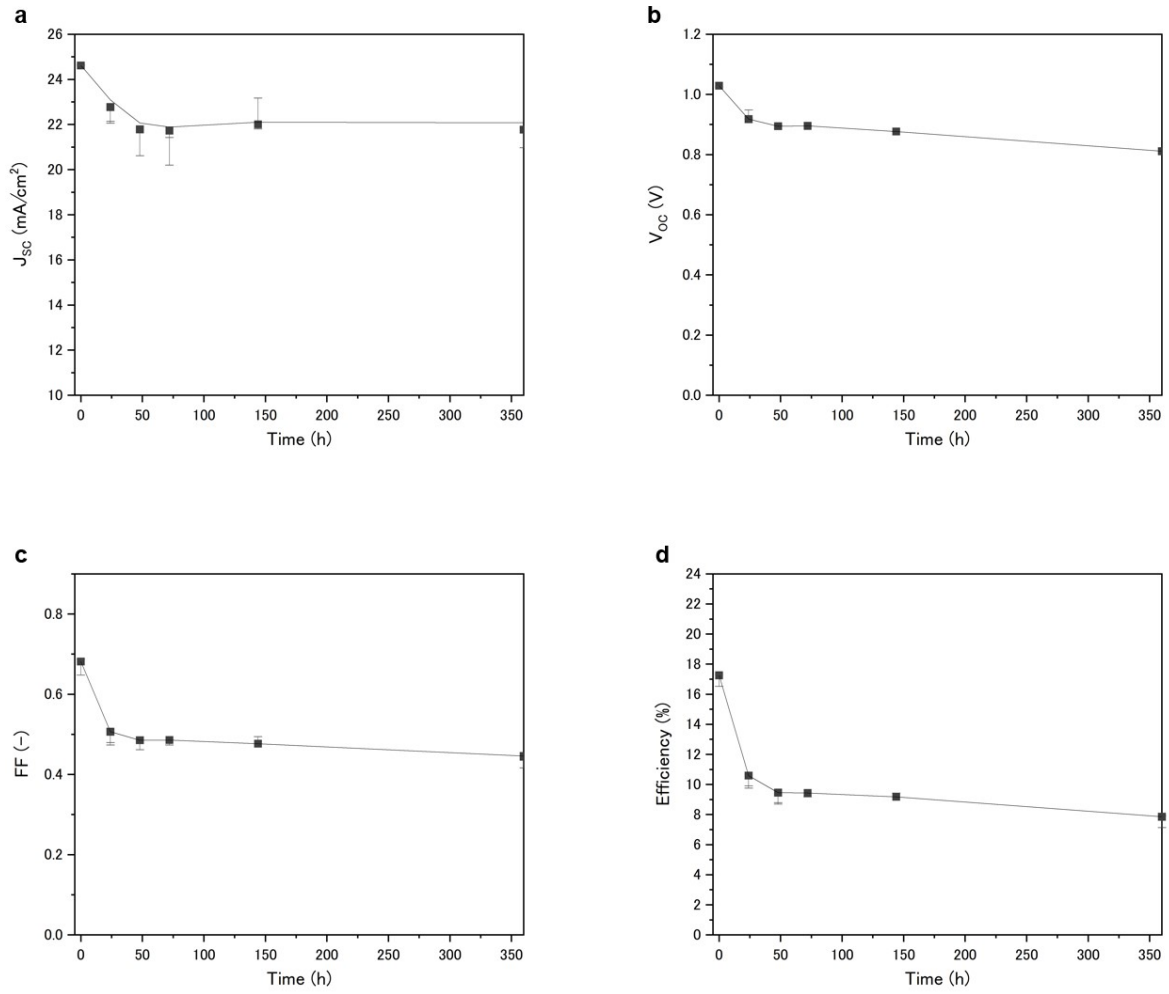


Figure S18. Stability test of devices passivated with 2D perovskite (BAI) under 65°C in dark for each solar cell parameter in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

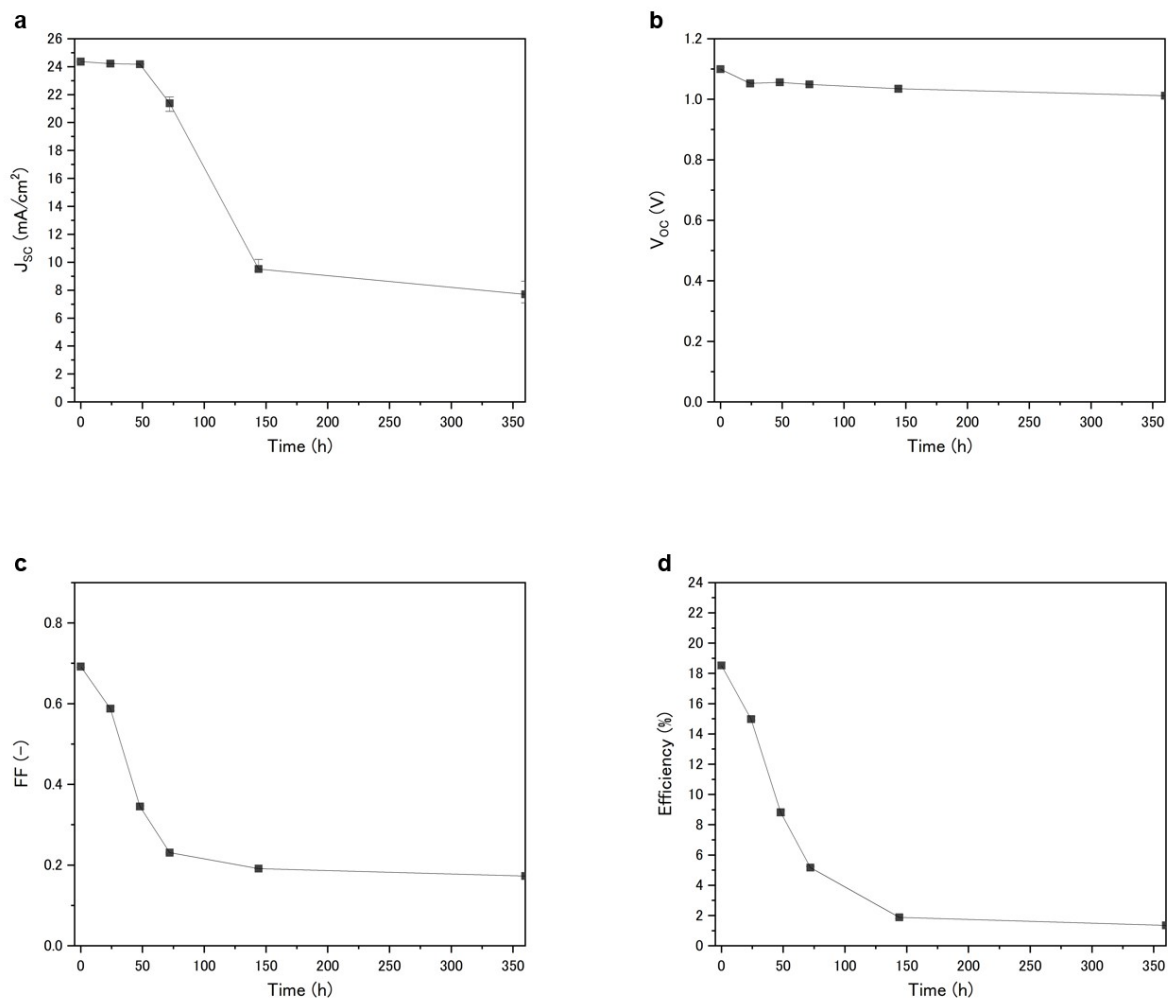


Figure S19. Stability test of devices passivated with 2D perovskite (OAI) under 65°C in dark for each solar cell parameter in forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

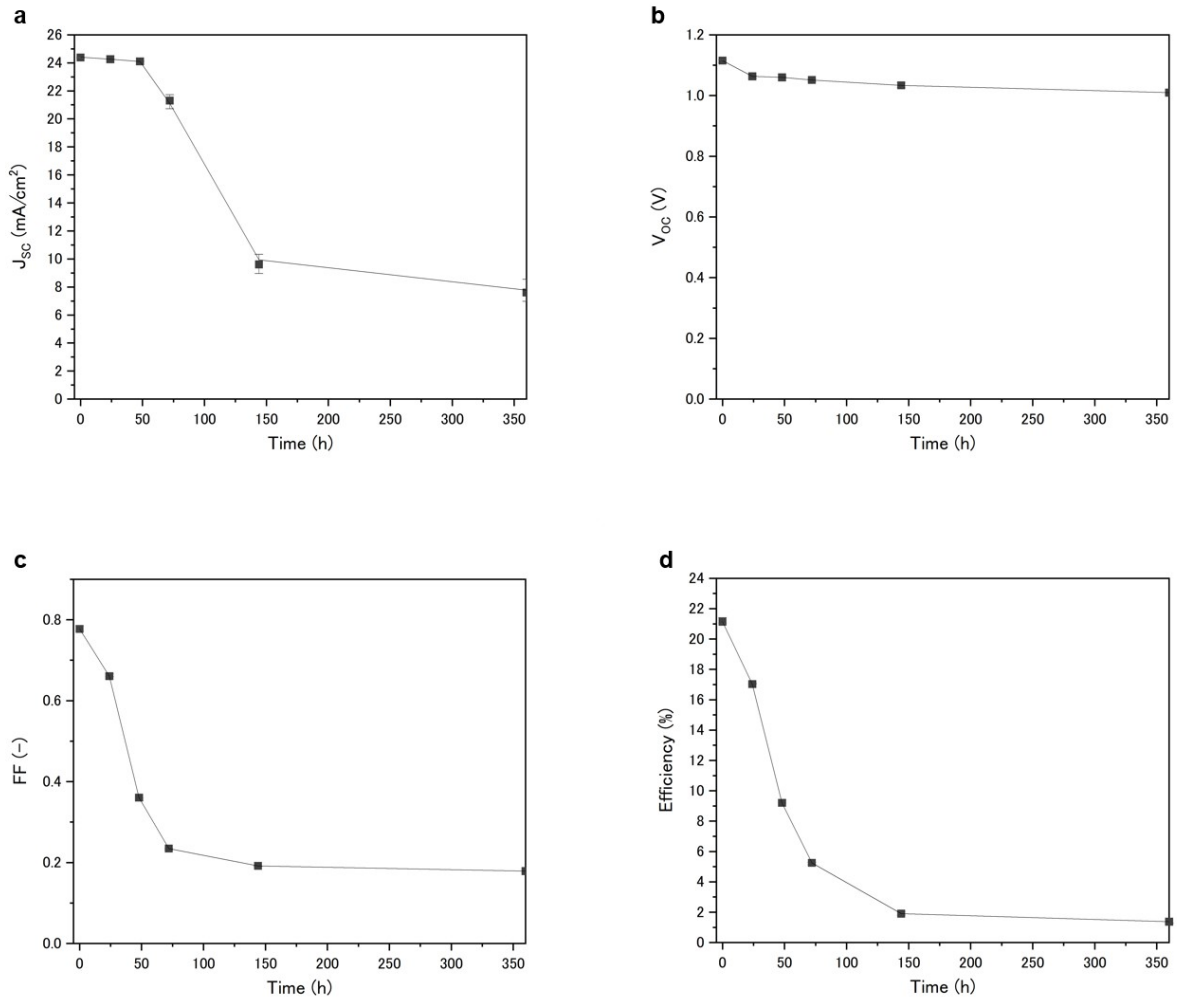


Figure S20. Stability test of devices passivated with 2D perovskite (OAI) under 65°C in dark for each solar cell parameter in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

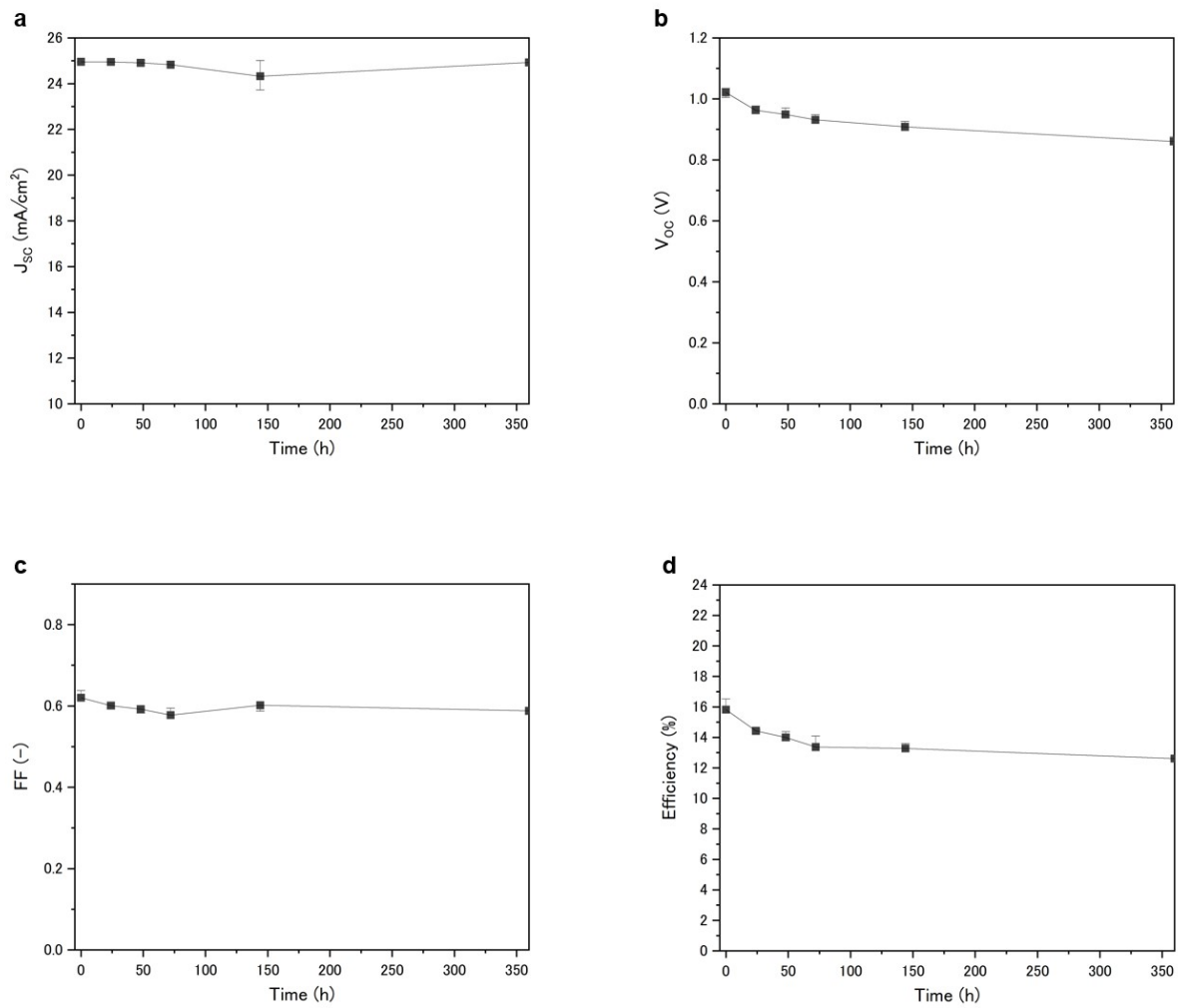


Figure S21. Stability test of devices without passivation under 65°C in dark for each solar cell parameter in

forward scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

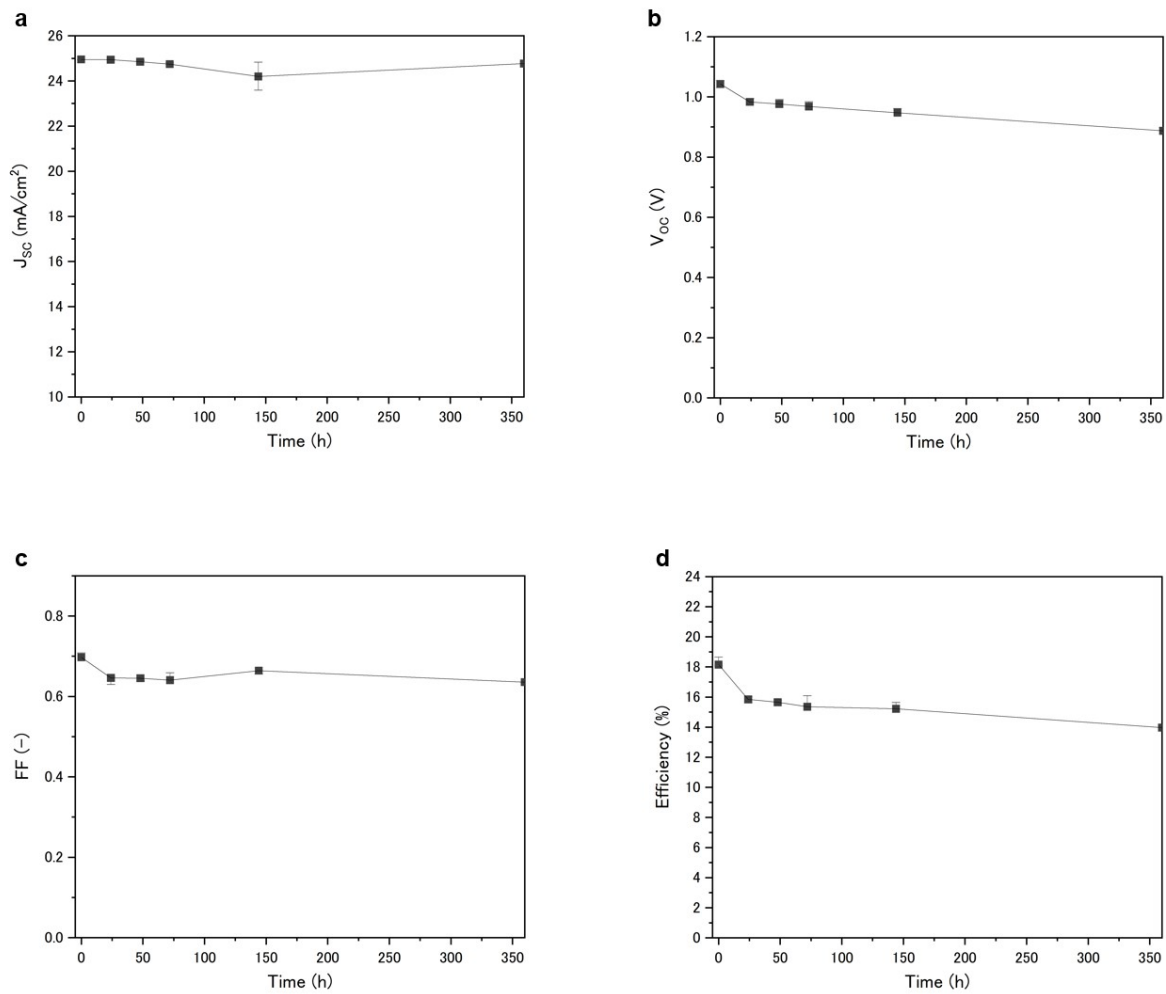


Figure S22. Stability test of devices without passivation under 65°C in dark for each solar cell parameter in reverse scan. (a) J_{sc} (b) V_{oc} (c) FF (d) efficiency. The sample number is 3.

Supplementary note 1:

Note that, in the case of the high-temperature condition, we could not observe the improvement of the device stability by the passivation (supplementary Figure S15-22). The reason can be the hole-transport layer (Spiro-OMeTAD) which is not stable under high-temperature, so we need to discuss the result carefully by considering degradation of the hole transport layer. XRD spectra for 2D peaks at room temperature under dark conditions did not show significant changes over time (supplementary Figure S13). However, in case of high-temperature condition, the 2D perovskite peaks were disappeared after 72 hours completely (supplementary Figure S14). These findings suggest that the 2D perovskite on the 3D perovskite layer is stable only when it is at room temperature and in the dark. Interestingly, the PbI_2 peak (12.6°) increased under the high-temperature condition regardless of the 2D passivation (supplementary Figure S14), which can be the reason for the decrease of the photovoltaic parameters (supplementary Figure S15-22).

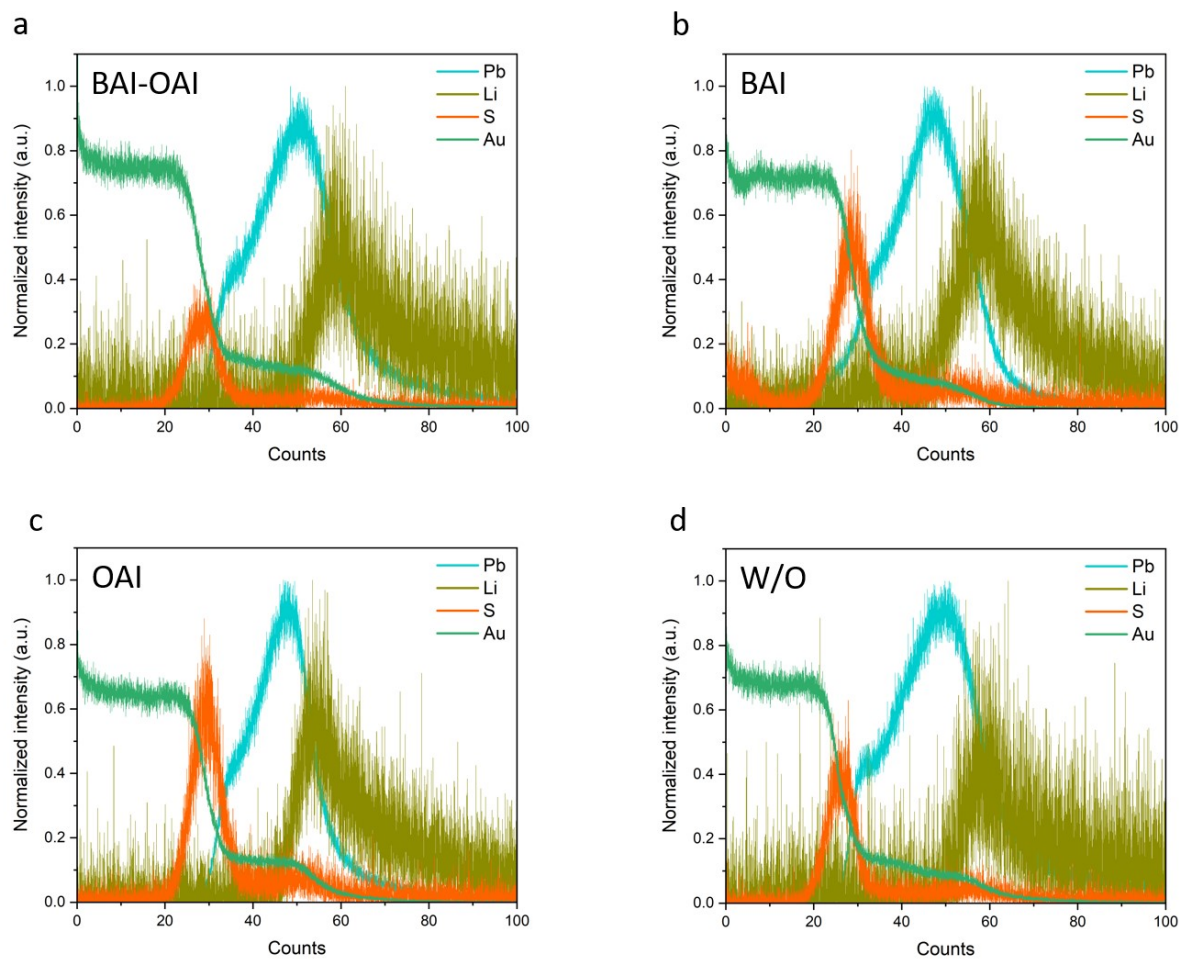


Figure S23. Depth profile for each element (Pb, Li, S, Au) with GD-OES measurement with (a) 2D perovskite (BAI-OAI), (b) 2D perovskite (OAI), (c) 2D perovskite (OAI) and (d) without passivation after 160 hours of light irradiation. Complete devices were measured.

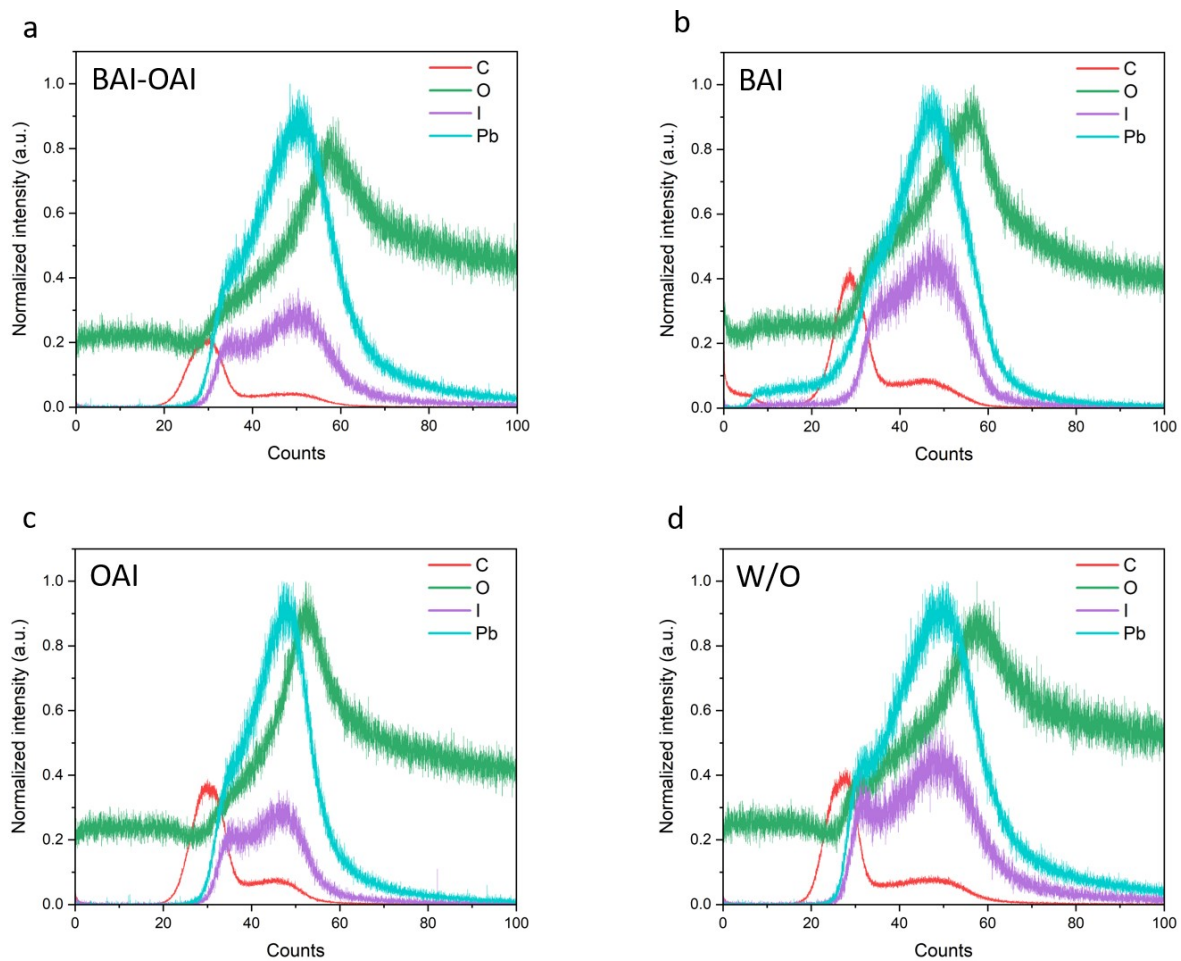


Figure S24. Depth profile for each element (C, O, I, Pb) with GD-OES measurement with (a) 2D perovskite (BAI-OAI), (b) 2D perovskite (OAI), (c) 2D perovskite (OAI) and (d) without passivation after 160 hours of light irradiation. Complete devices were measured.

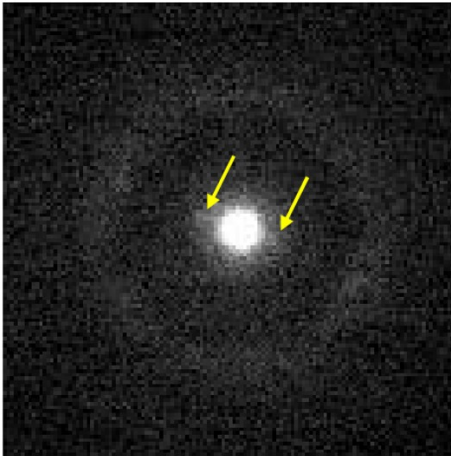
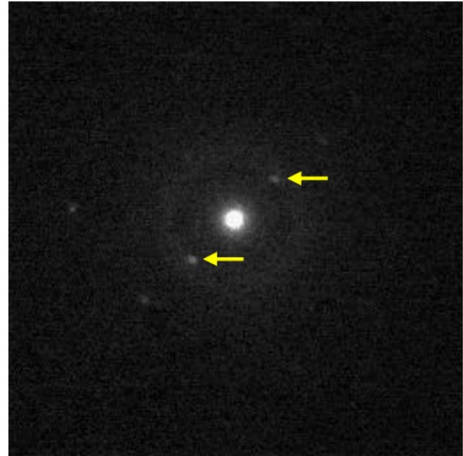
a**b**

Figure S25. TEM electron diffraction analysis of 2D perovskite layer for (a) 24.3° and (b) 8.9° .

Measurement position was perovskite surface. These diffraction peaks are consistent with XRD result of 2D perovskite (BAI).