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

Supporting Information

Aging-Induced Light-Soaking Effects and Open-Circuit Voltage Hysteretic Behavior of Inverted Perovskite Solar Cells Incorporating a Hole Transport Metal Halide Layer via Morphology-Dependent Inflow of Iodide Ions

Jungyun Hong, Hyebin Kim and Inchan Hwang*

Department of Electronic Materials Engineering, Kwangwoon University, Seoul 01897,

Republic of Korea

*Corresponding author. E-mail address: ihwang@kw.ac.kr

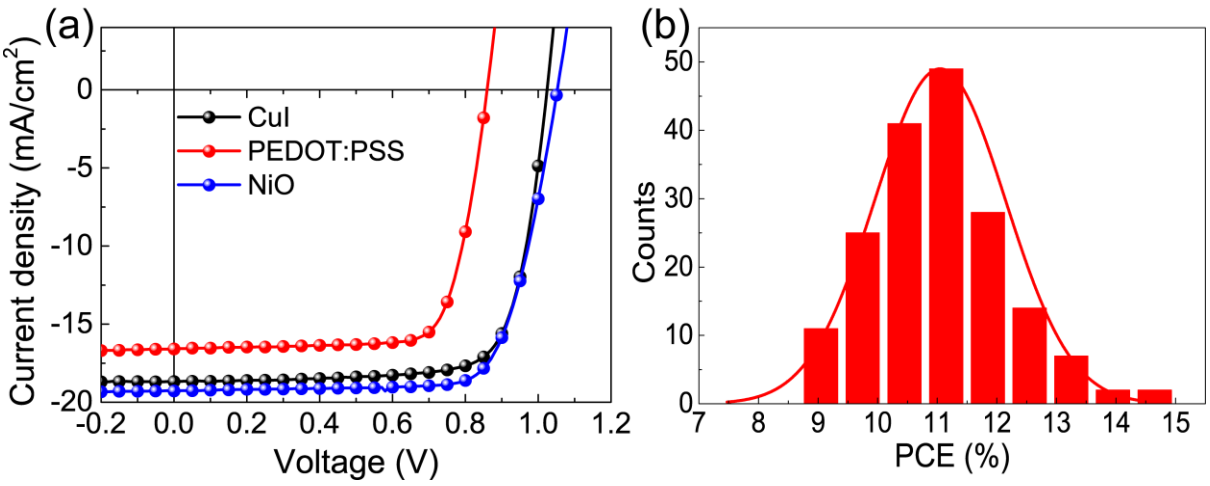

Figure S1. (a) *J-V* characteristics of the devices structured with ITO/HTL/$MAPbI_3$/PCBM/ZnO NP/Ag where HTL is CuI (black), PEDOT:PSS (red) and NiO (blue). (b) Histogram of the PCE for 180 devices with a CuI HTL.

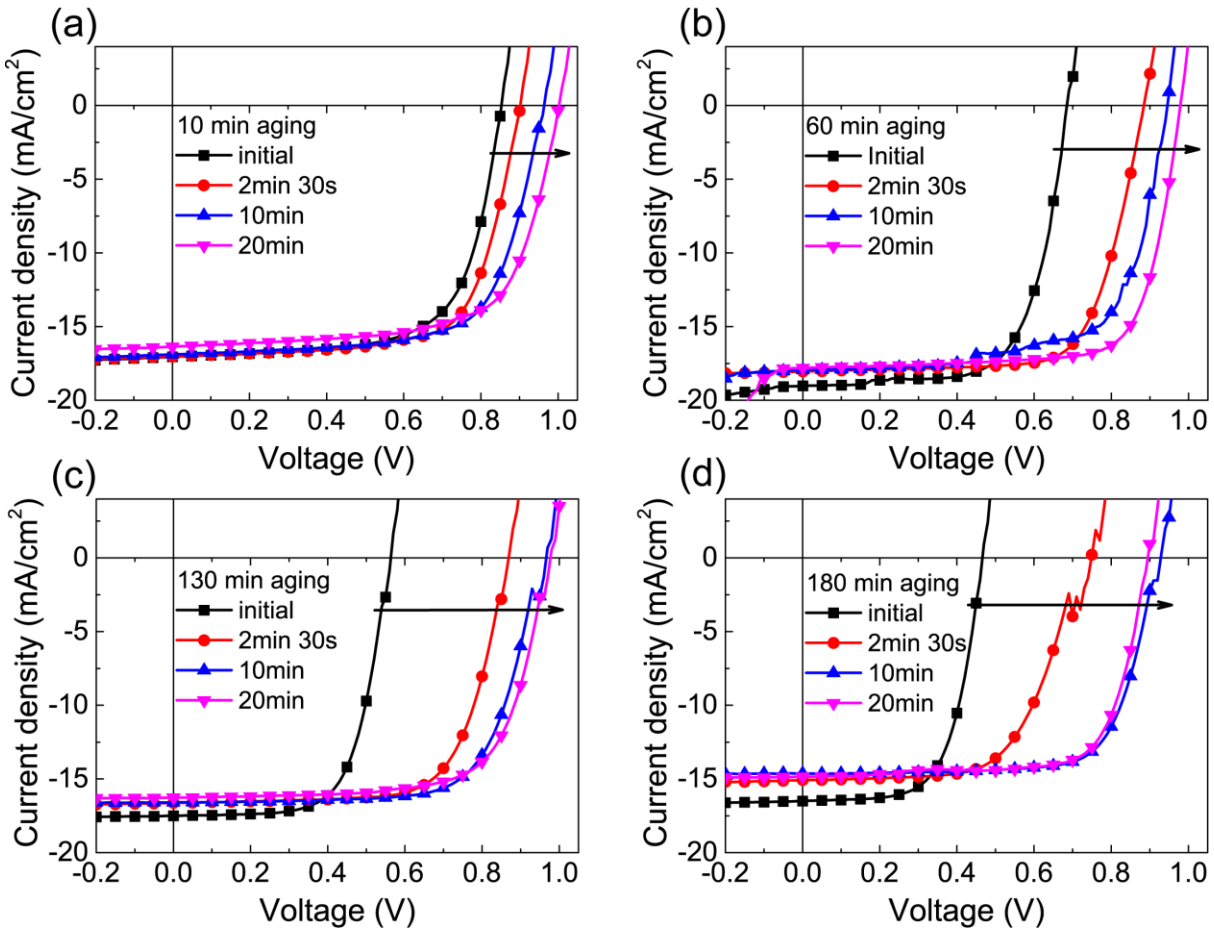

Figure S2. *J-V* characteristics of $MAPbI_3$ perovskite solar cells with CuI as a hole transport layer. The *J-V* scans started in the forward direction after kept in the dark for different times (10, 60, 130 and 180 min aging) in the nitrogen-filled glove box after device completion. For each aging time, light illumination is continuously given to the devices to identify light-soaking effects. The initial *J-V* characteristics were recorded after the devices had been in the dark for 2 min before illumination.

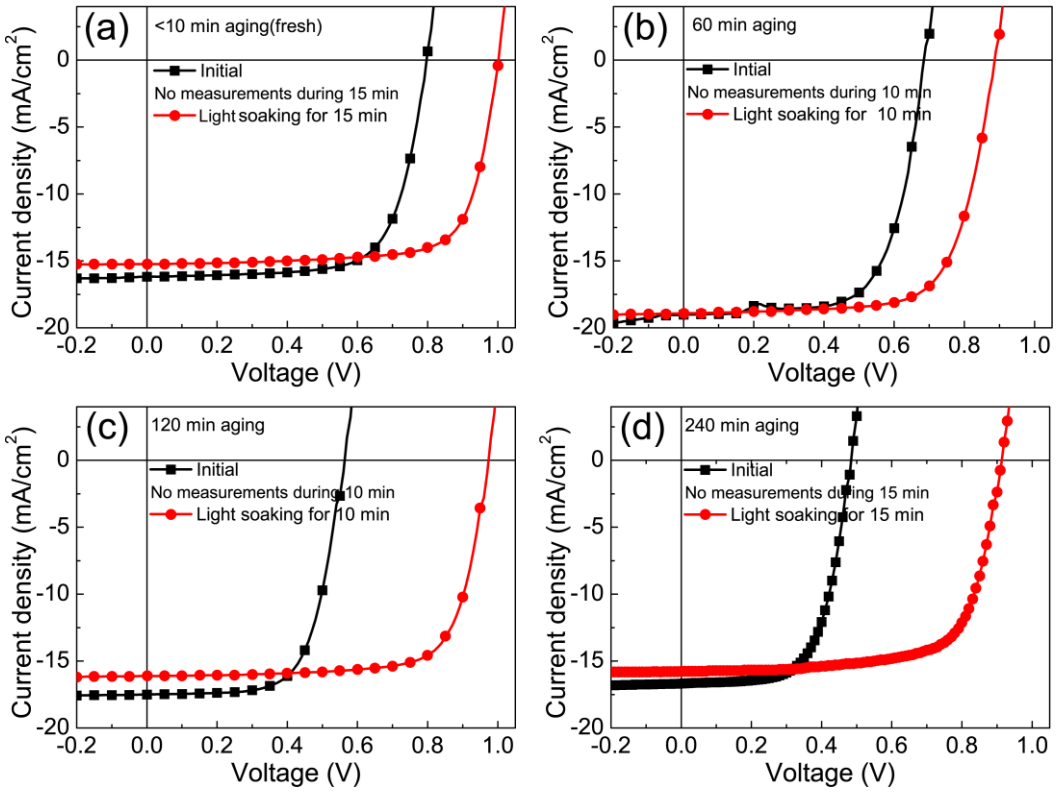

Figure S3. *J-V* characteristics recorded in the forward direction at different aging times, (a) less than 10 min, (b) 60 min aging, (c) 120 min aging and (d) 240 min aging for $MAPbI_3$ perovskite solar cells with CuI as a hole transport layer. The measurement procedure is the same as Figure S2, except that the *J-V* scans were carried out only for initial (without light soaking) and after 10 or 15 min light soaking, in order to demonstrate that the continuous *J-V* scans do not much influence the photovoltaic performance of light-soaked devices.

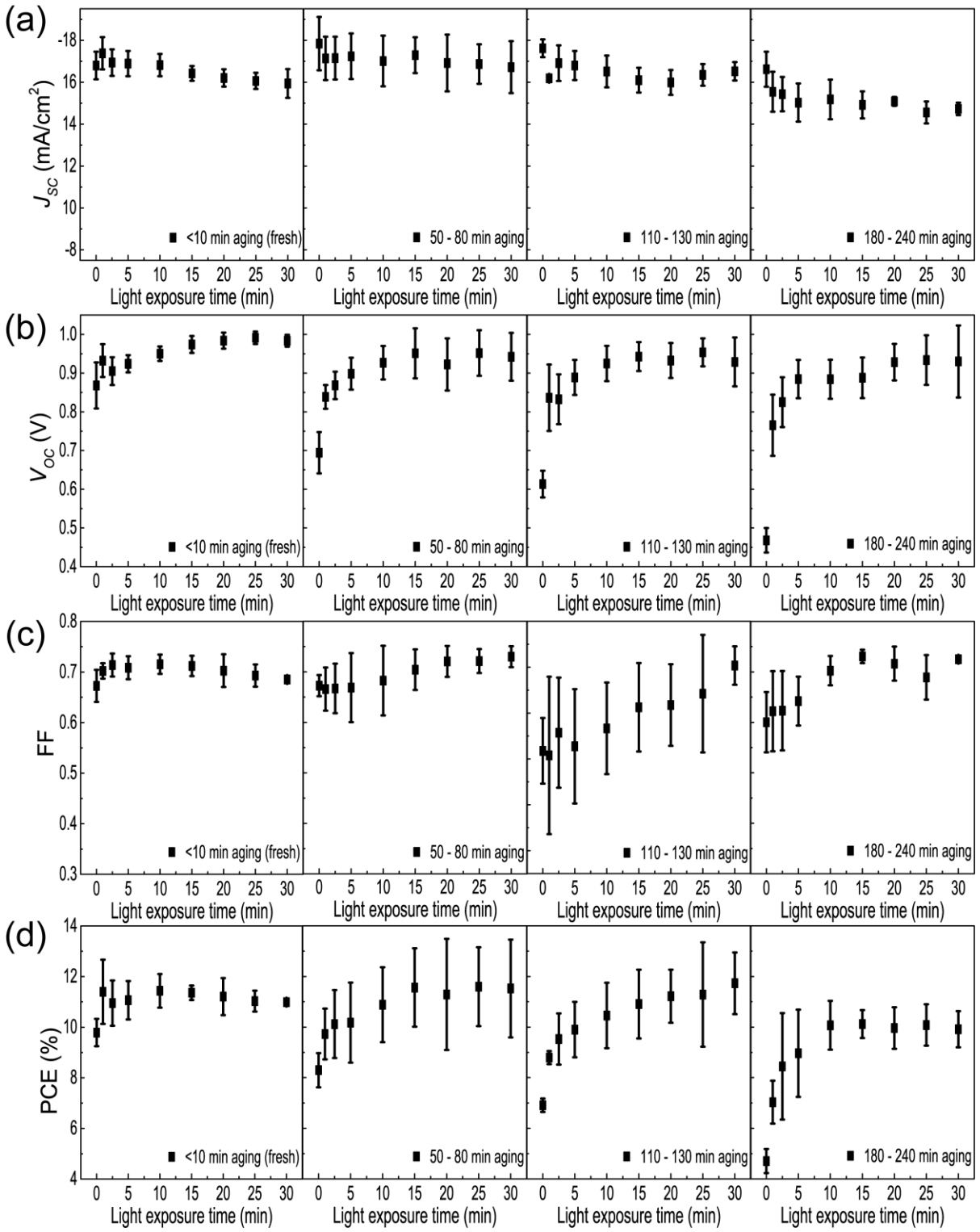

Figure S4. Statistical photovoltaic parameters such as (a) J_{sc}, (b) V_{oc}, (c) FF and (d) PCE as a function of light soaking time for a range of aging times; less than 10 min, 50-80 min, 110-130 min and 180-240 min.

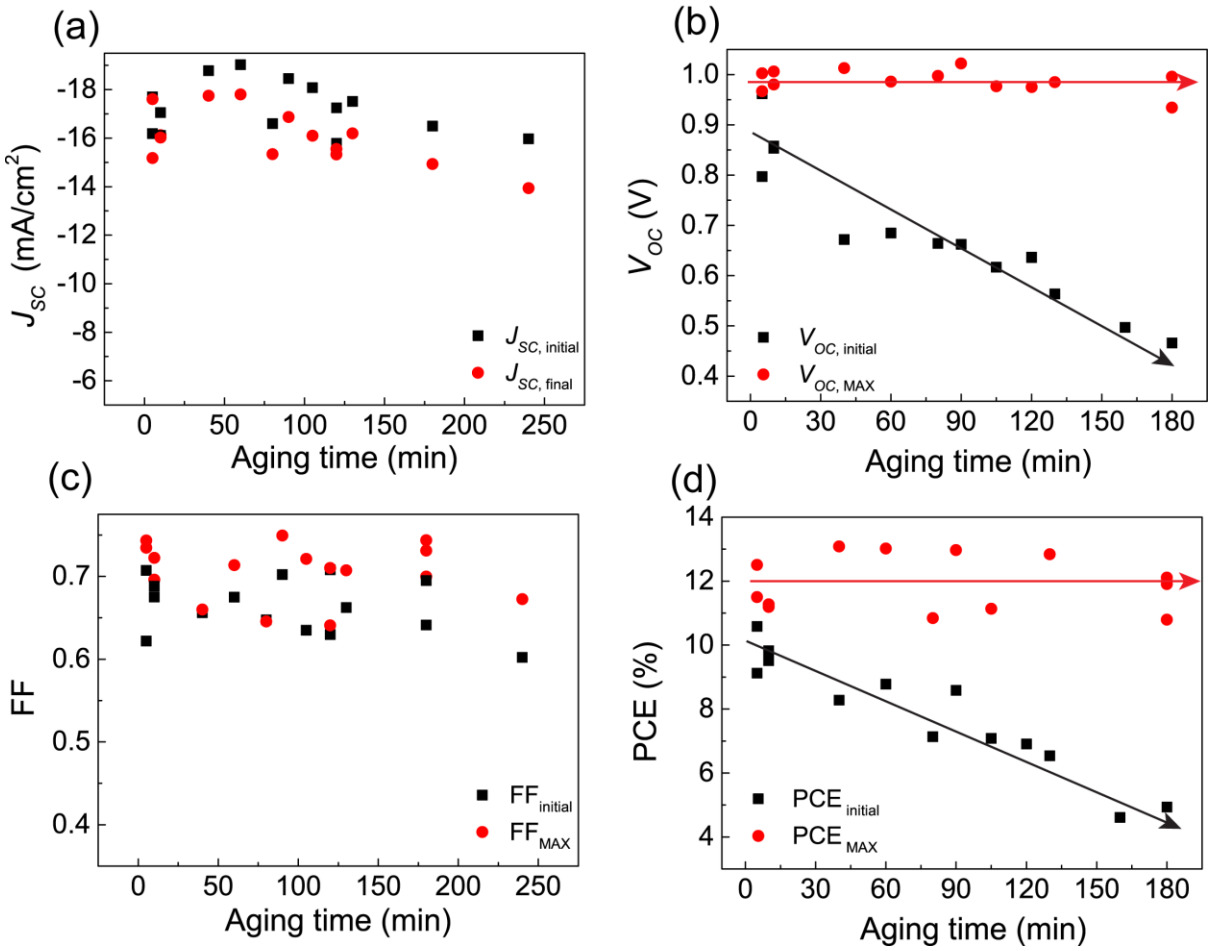

Figure S5. Initial and light-soaked (max or final) photovoltaic parameters such as (a) J_{sc}, (b) V_{oc}, (c) FF and (d) PCE from forward-scanned *J-V* characteristics as a function of aging time. The photovoltaic parameters after full light-soaking are constant with aging time. The dramatic changes with aging time can be found for initial open-circuit voltage ($V_{oc,\ initial}$). The initial open-circuit voltage notably decreases with aging time, and so does $PCE_{initial}$.

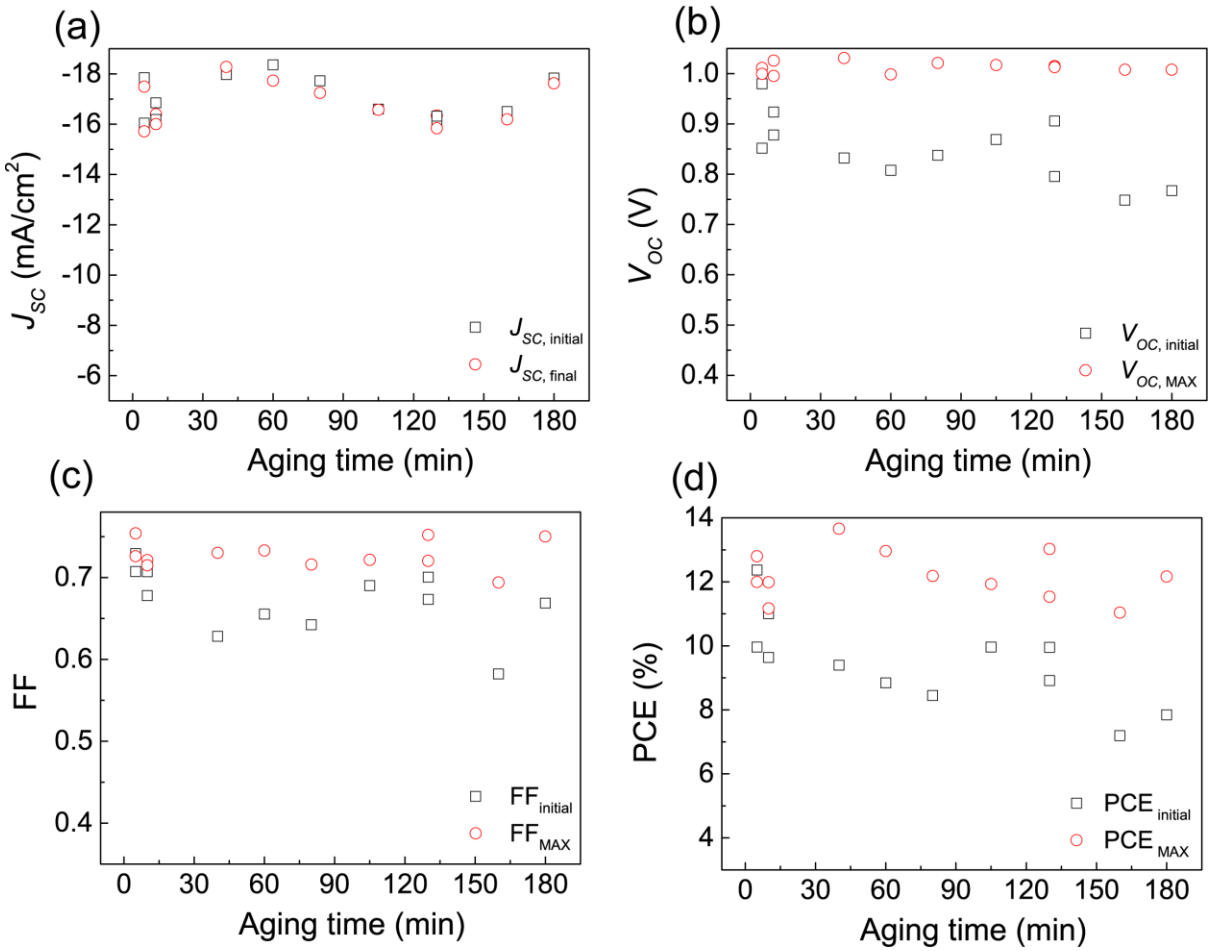

Figure S6. Initial and light-soaked (max or final) photovoltaic parameters such as (a) J_{sc}, (b) V_{oc}, (c) FF and (d) PCE determined from reverse-scanned *J-V* characteristics as a function of aging time. The changes are similar to the forward scan cases, but the degree is small.

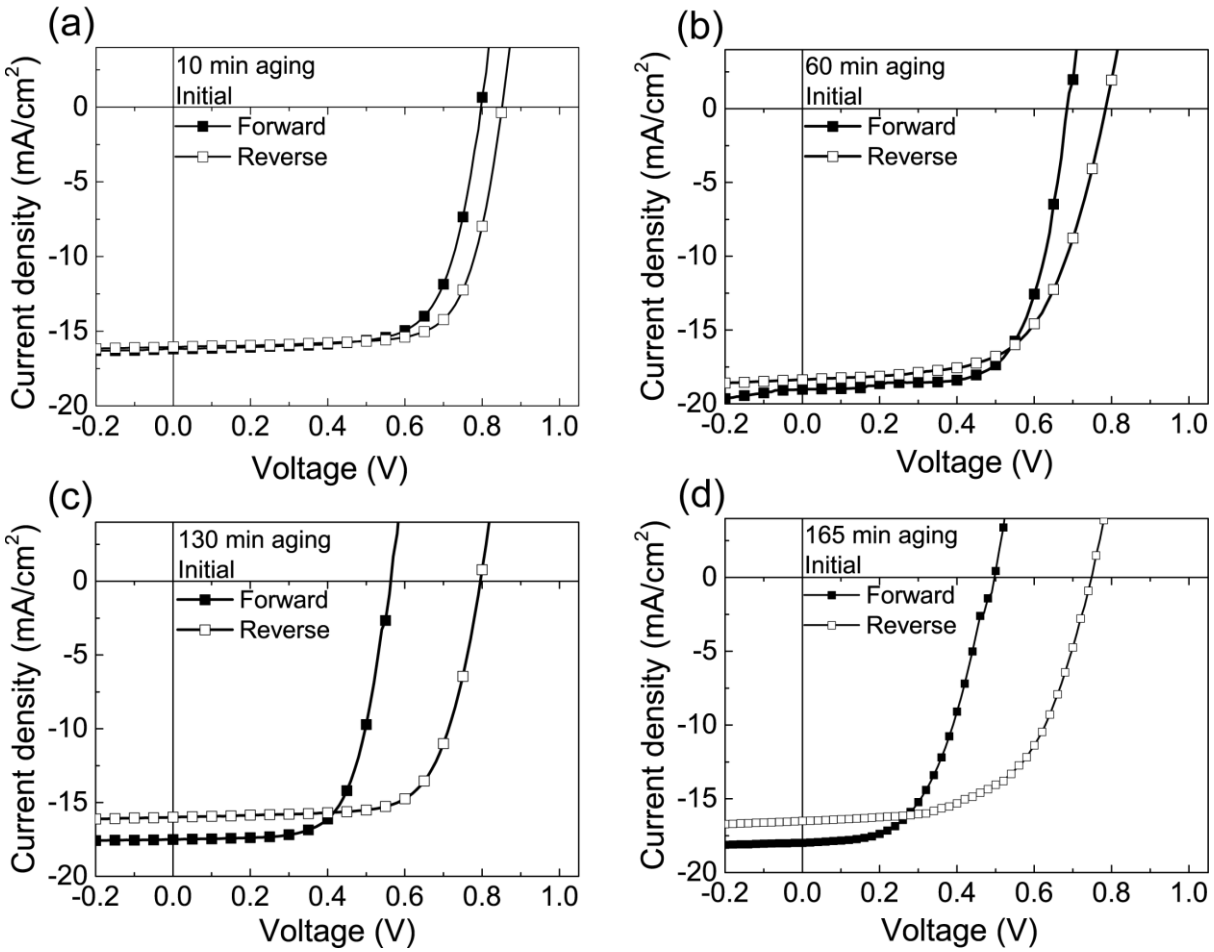

Figure S7. *J-V* characteristics in the forward (solid symbols) and reverse directions (open symbols) without light soaking, recorded at an aging time of (a) 10 min, (b) 60 min, (c) 130 min and (d) 165 min.

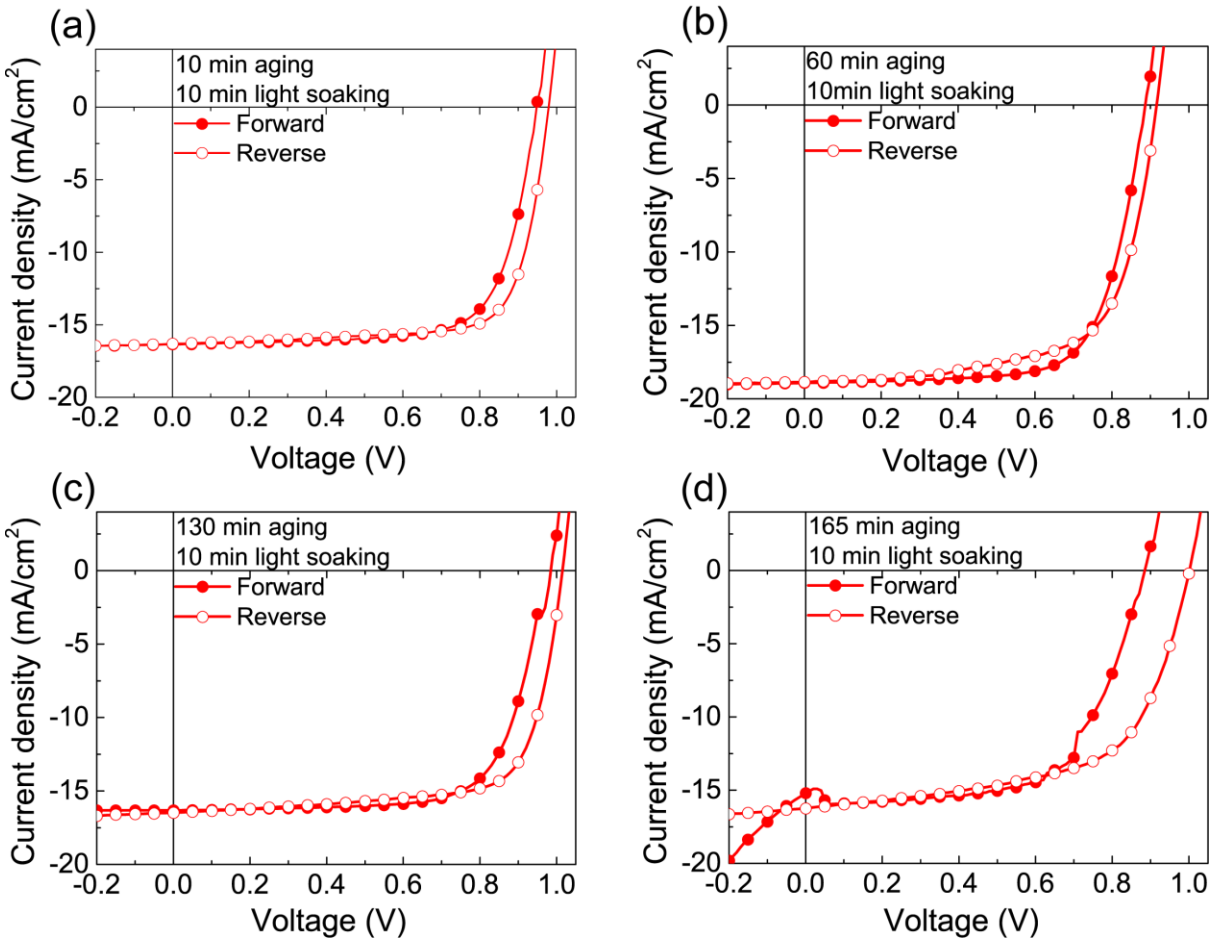

Figure S8. *J-V* characteristics in the forward (solid symbols) and reverse directions (open symbols) after 10 min light soaking, recorded at an aging time of (a) 10 min, (b) 60 min, (c) 130 min and (d) 165 min.

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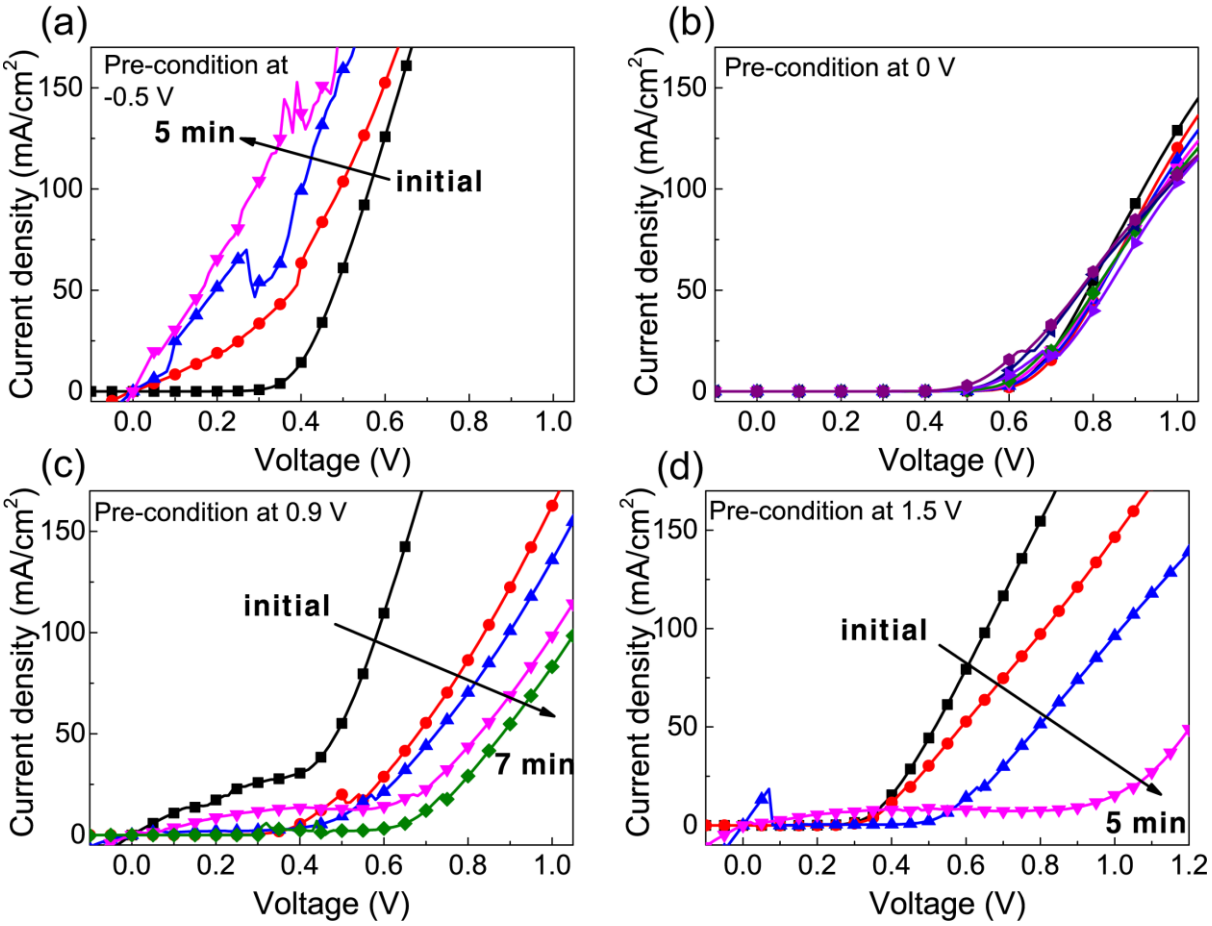

Figure S9. Dark *J-V* characteristics recorded in the forward direction after preconditioning at an external voltage of (a) -0.5 V (negative bias), (b) 0 V (short circuit condition), (c) 0.9 V (near the open-circuit voltage), and (d) 1.5 V kept in the dark for different times.

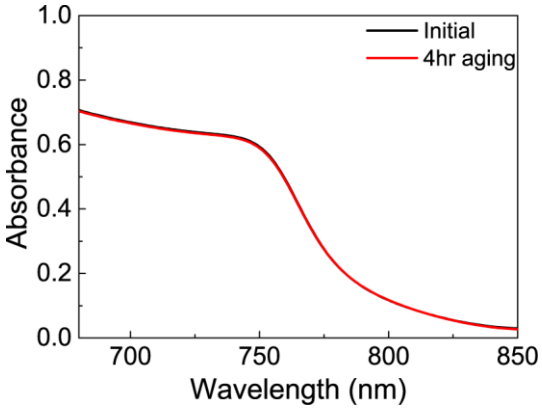

Figure S10. Absorption spectra of $MAPbI_3$ prepared on CuI with and without aging. The absorption spectra are identical.

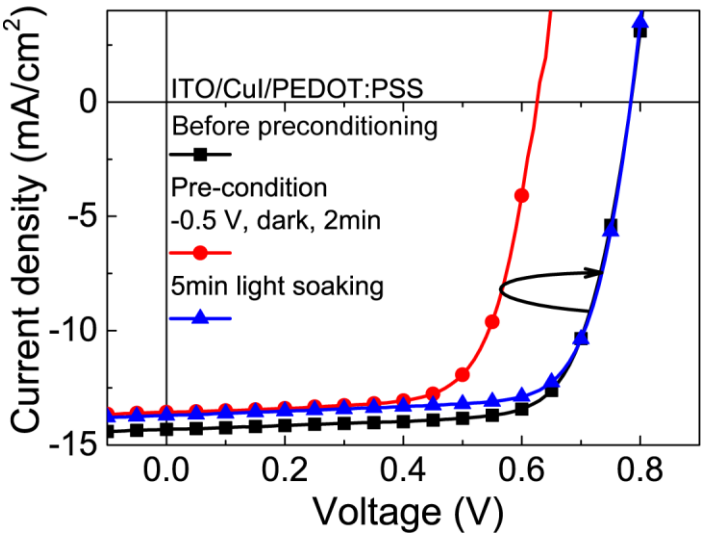

Figure S11. *J-V* characteristics recorded in the forward direction for the perovskite solar cells structured with ITO/CuI/PEDOT:PSS/$MAPbI_3$/PCBM/ZnO NP/Ag after 4 hours aging. The insertion of the PEDOT:PSS layer between CuI and $MAPbI_3$ aims to block the inflow of iodide ions from CuI to $MAPbI_3$. After preconditioning at -0.5 V, the open-circuit voltage is reduced, but just by 0.15 V, which is far small compared to the case of ITO/CuI/$MAPbI_3$/PCBM/ZnO/Ag. This supports the additional inflow of iodide ions from CuI. After 5 min light soaking following the preconditioning, the open-circuit voltage was restored.

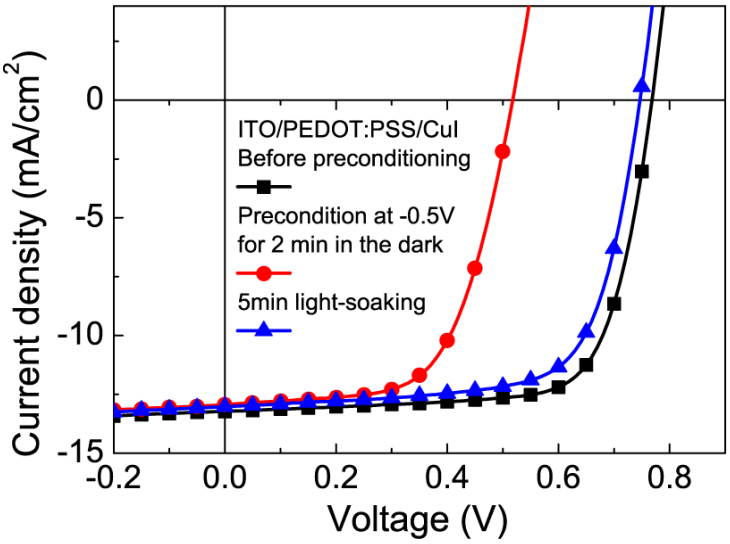

Figure S12. *J-V* characteristics recorded in the forward direction for the perovskite solar cells structured with ITO/PEDOT:PSS/CuI/$MAPbI_3$/PCBM/ZnO NP/Ag after 4 hours aging.

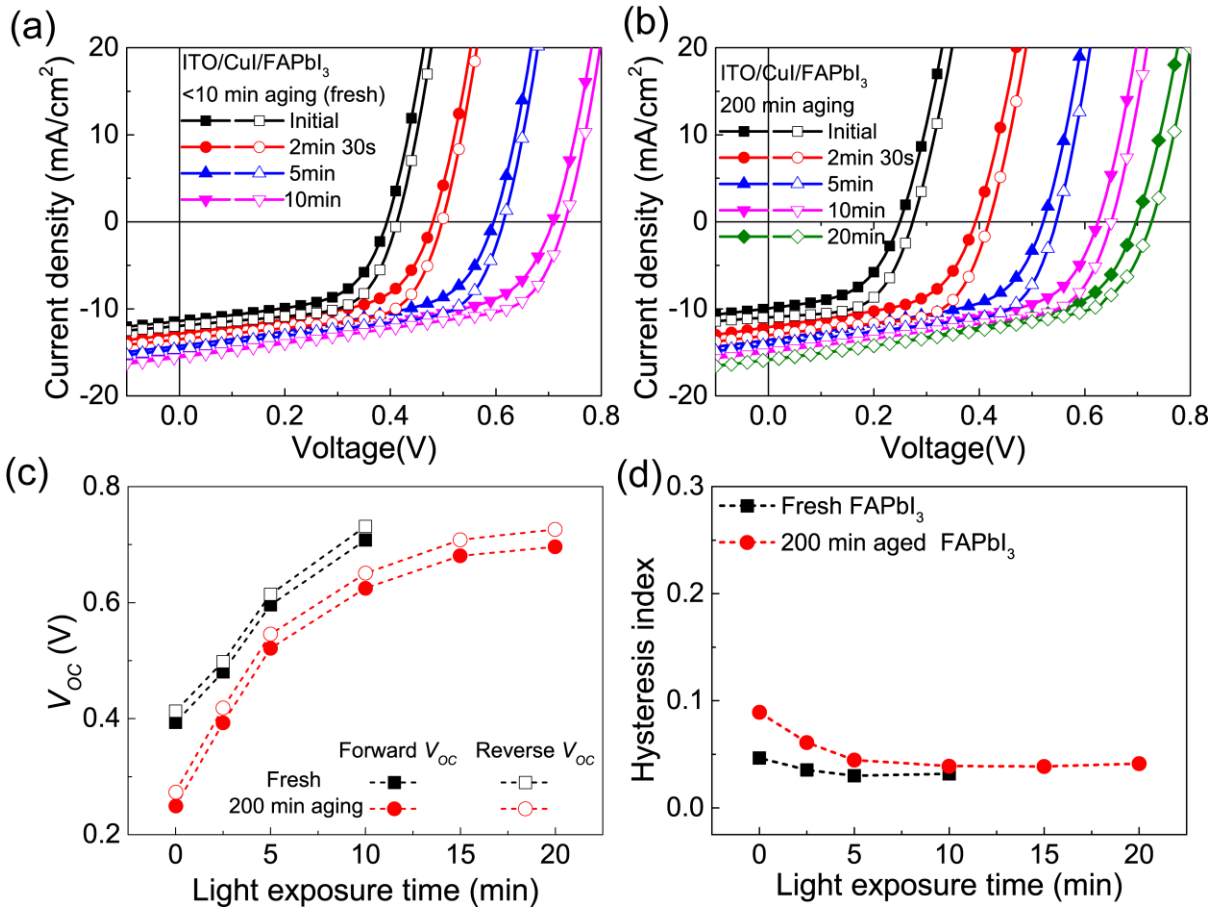

Figure S13. *J-V* characteristics of the $FAPbI_3$ devices (a) with less than 10 min aging and (b) with 200 min aging. The solid and open symbol represents the forward and reverse *J-V* scans, respectively. (c) Open-circuit voltage and (d) hysteresis index as a function of light-soaking time for the fresh and aged devices. Please note that the devices have to be further improved for further work.