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

Supplementary Information

Notorious Role of Spiro-OMeTAD in Performance Deterioration of Perovskite Solar Cells at High Temperature and Reuse of the Perovskite Films to Avoid Pb-waste

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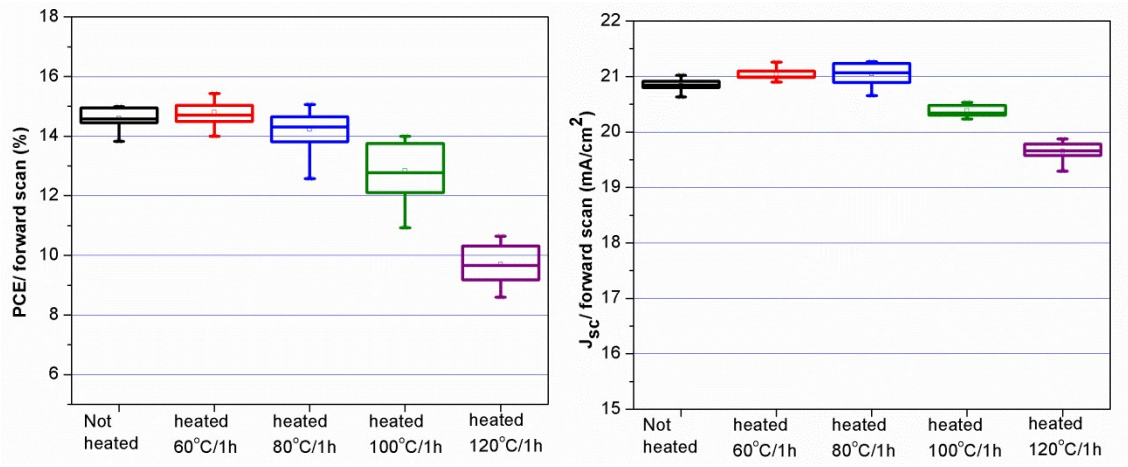
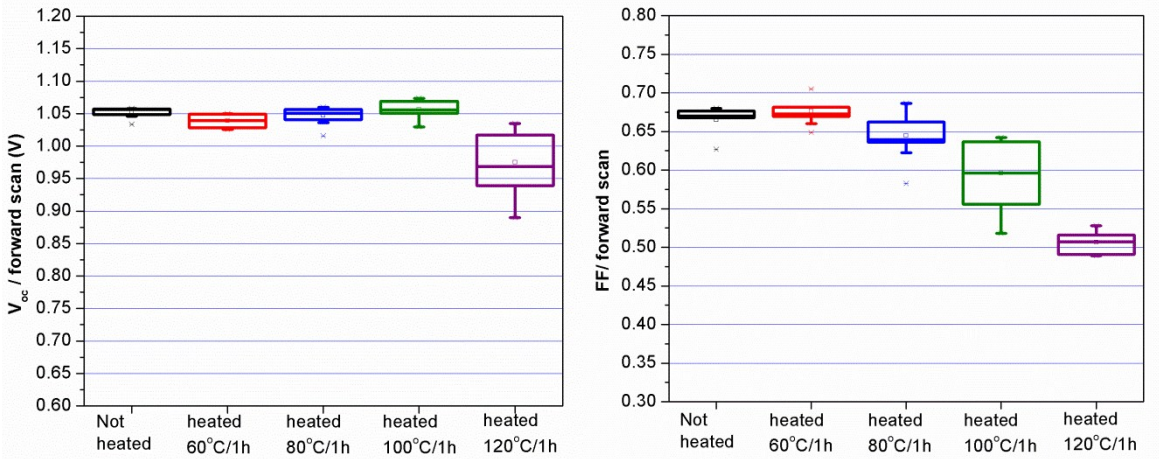
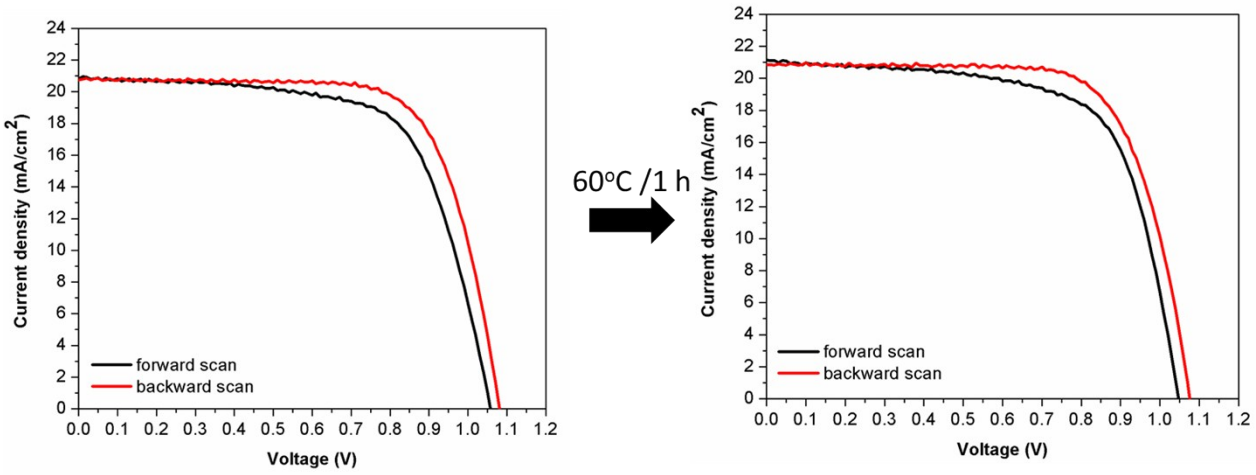


(a) **(b)**

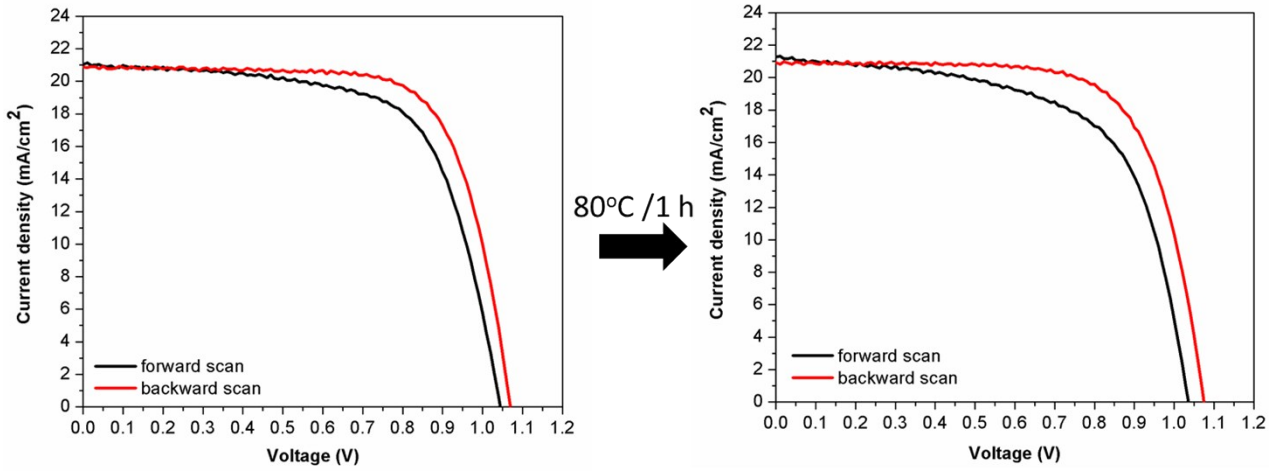


(c) **(d)**

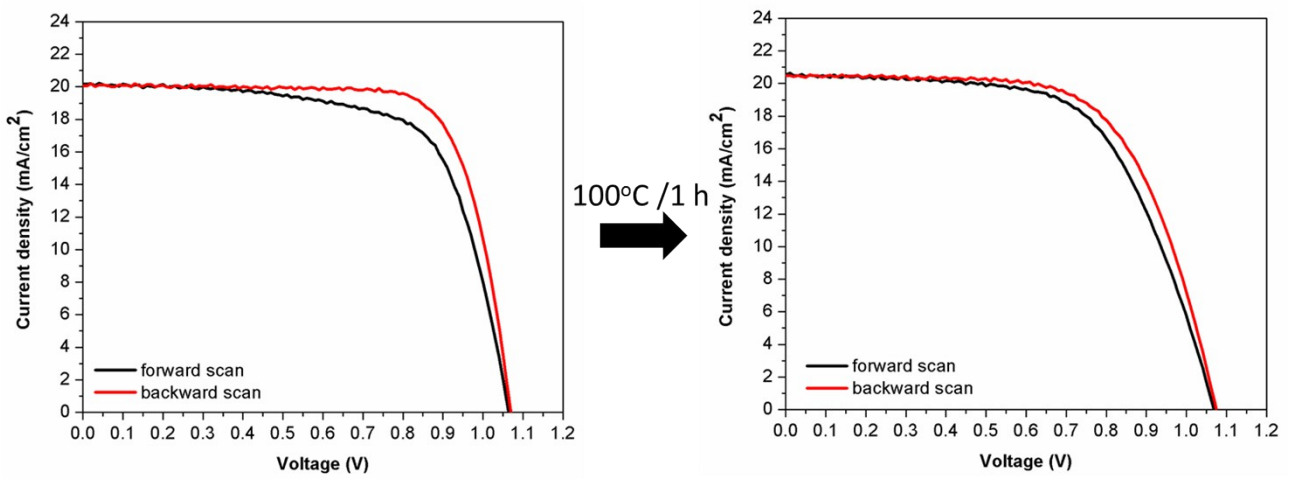
Figure S1 (a) PCE, (b) J_{sc}, (c) V_{oc}, and (d) FF of planar $MAPbI_3$ perovskite solar cells (measured on forward scan) before heating and after heating at different temperatures (60, 80, 100 and 120 ^{o}C).



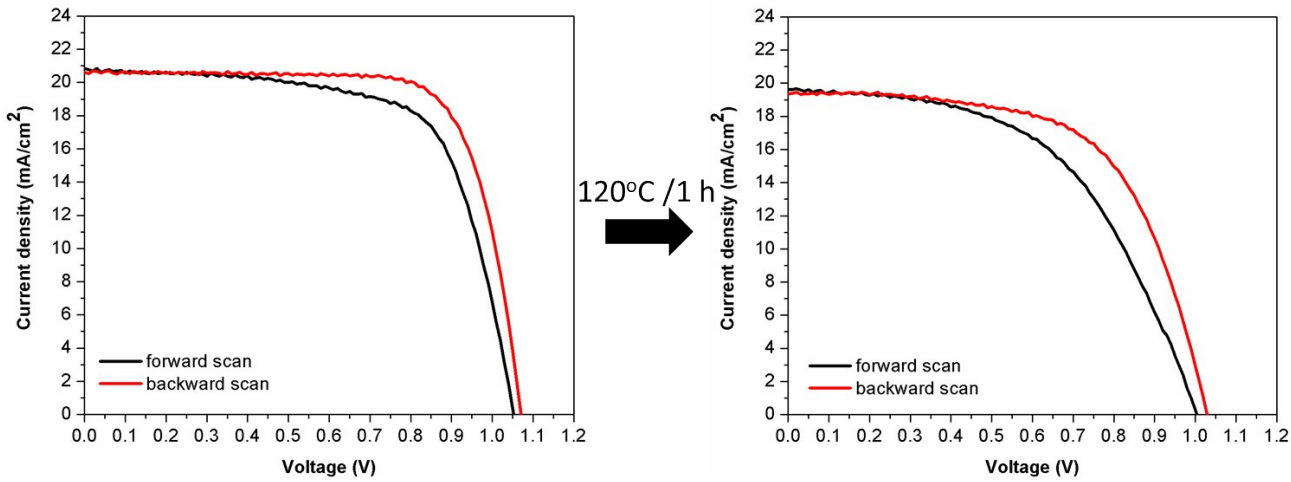
(a)



(b)

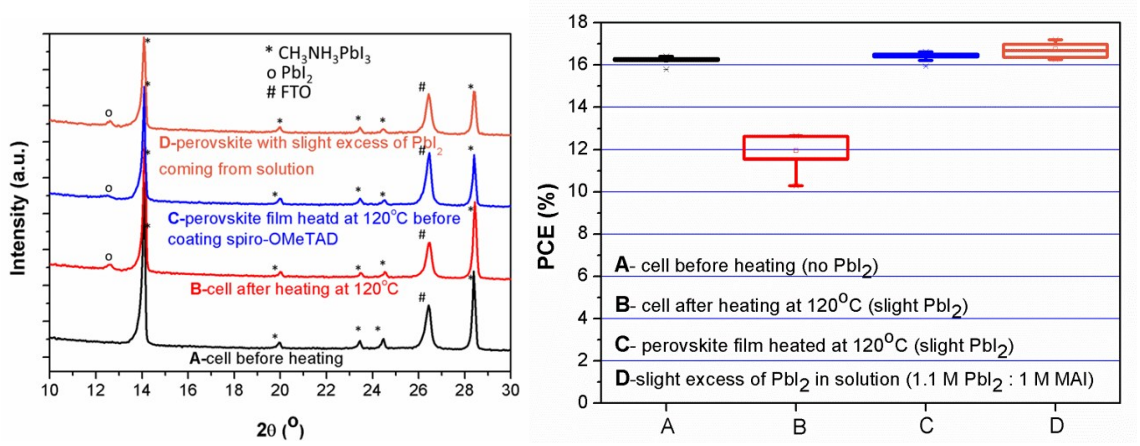


(c)



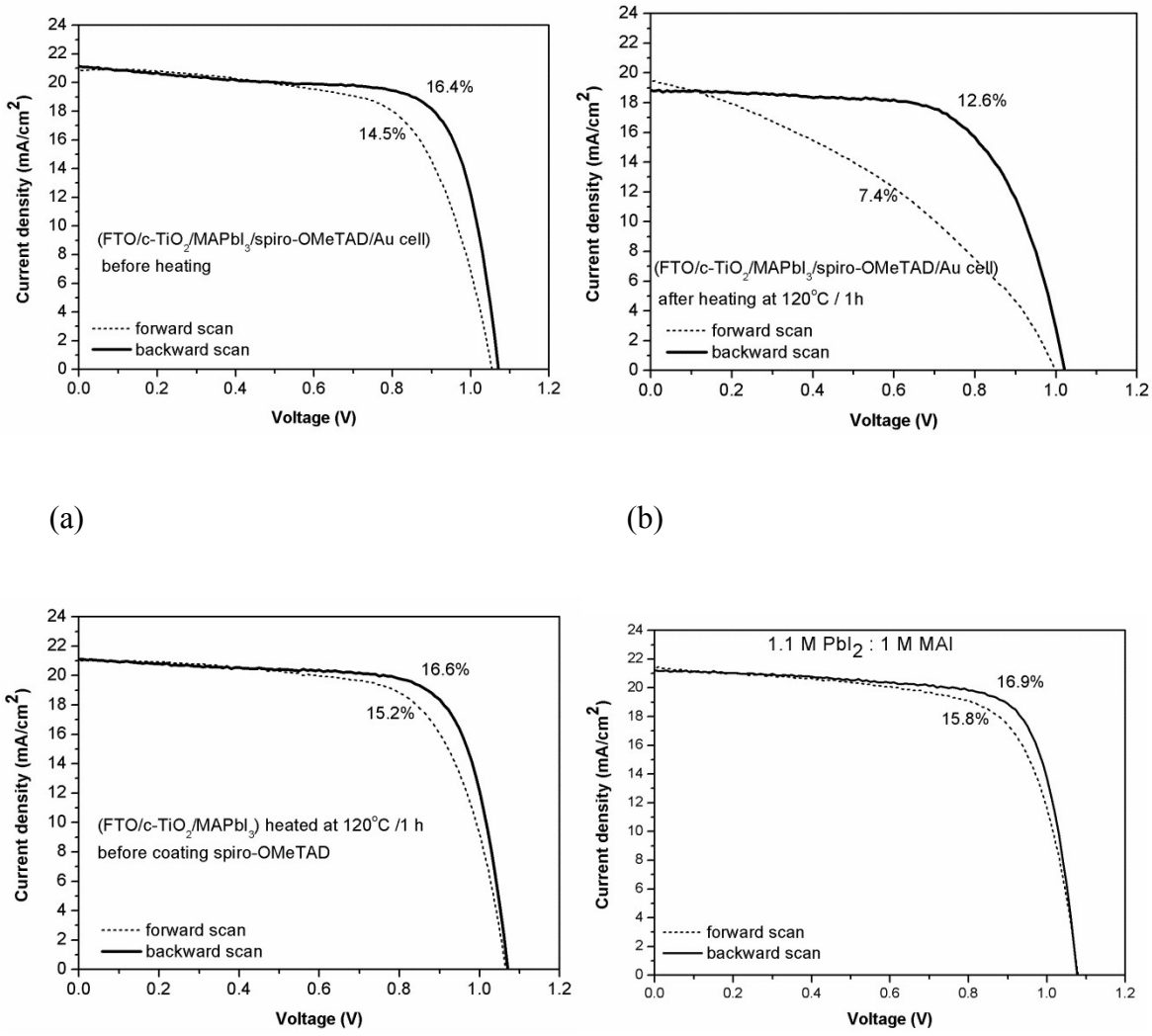
(d)

Figure S2 J-V curves of planar $MAPbI_3$ perovskite solar cells before and after heating at (a) 60°C, (b) 80°C, (c) 100°C and 120°C for 1 h.



(a) (b)

Figure S3 (a) XRD patterns and (b) PCE (backward scan) of $MAPbI_3$ solar cells before heating (black), after heating at 120 °C for 1 h (red), with a perovskite film heated at 120 °C for 1 h before coating spiro-OMeTAD (blue), with remnant PbI_2 coming from a precursor solution with slight excess of PbI_2 (i.e. 1.1 M PbI_2: 1 M MAI) (orange). Representative J-V curves of the four cells are given in Figure S4.



(a) (b)

(c) (d)

Figure S4 J-V curves of planar $MAPbI_3$ perovskite solar cells (a) before (no PbI_2) and (b) after (slight PbI_2) heating at 120°C for 1 h, (c) with $MAPbI_3$ film heated at 120°C for 1 h (slight PbI_2) before coating spiro-OMeTAD, (d) with slight excess of PbI_2 coming from solution (1.1 M PbI_2 : 1 M MAI)

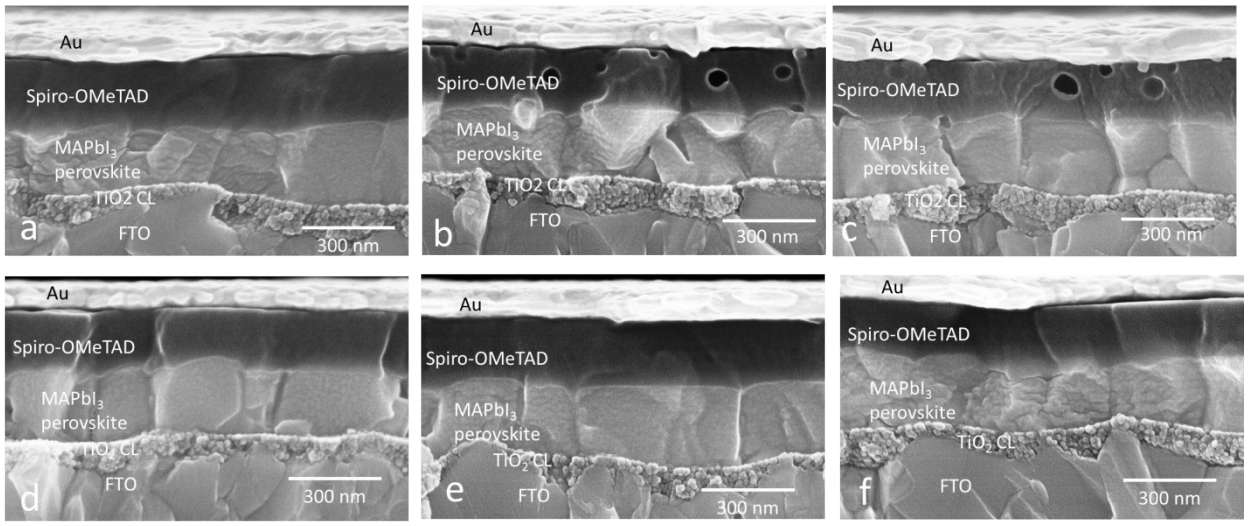


Figure S5 Cross-sectional SEM micrographs of (a, b, c) planar $MAPbI_3$ solar cells heated (a-0 ^oC, b-100 ^oC and c-120 ^oC for 1 h) with Au electrode (FTO/c-TiO_2/$MAPbI_3$/Spiro-OMeTAD/Au) and (d, e. f) devices heated (d-0 ^oC, e-100 ^oC and f-120 ^oC for 1 h) before deposition of Au electrode (FTO/c-TiO_2/$MAPbI_3$/Spiro-OMeTAD).

Selecting a method of recycle

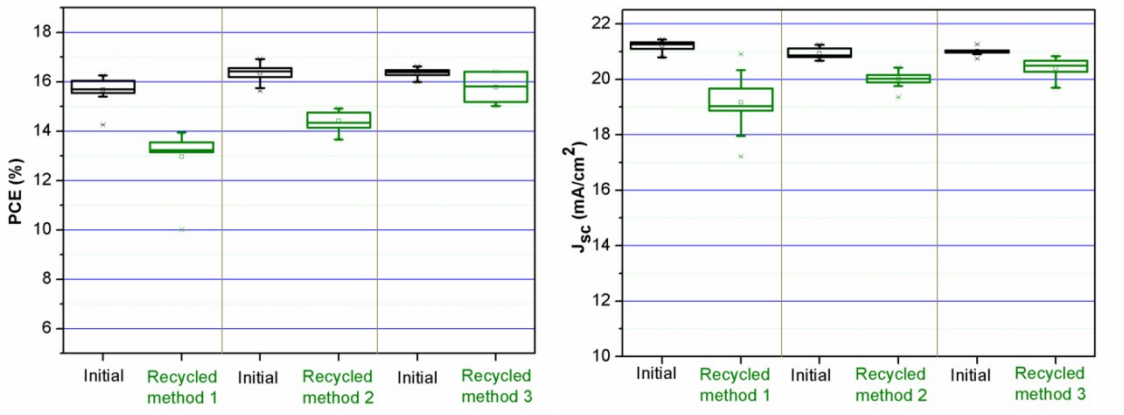
In order to establish a easy and short method of recycle, a number of methods were tried for washing off the spiro-OMeTAD layer and the methods essentially differed slightly in the durations for which the devices were exposed to chlorobenzene used to dissolve the spiro-OMeTAD. In first two methods, the devices were dipped in chlorobenzene for 10 and 5 minutes to dissolve the spiro-OMeTAD layer. In these methods, the Au film was removed

automatically after dissolution of spiro-OMeTAD. In the third method, the Au film was pulled off the device surface by an adhesive tape (3M Scotch) and then the spiro-OMeTAD layer, instead of dipping in chlorobenzene, was spin-washed (dripping chlorobenzene while spinning the substrate). In comparison to first two methods, the third method, where the devices were not dipped in chlorobenzene, showed slightly better recovery in the device performance (Figure S). Moreover, since the third method was easier and quicker, it was used for all further study undertaken here.

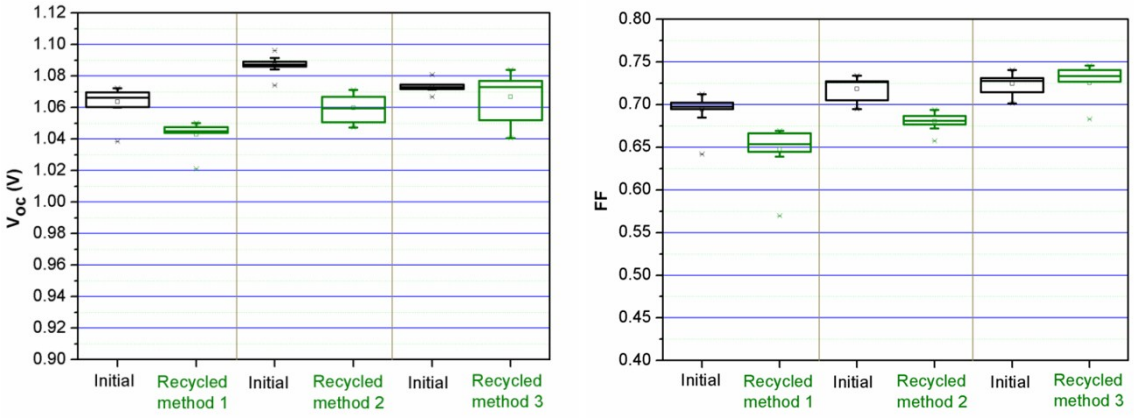
<u>Method 1</u>: Dip-washing (Dipping in CB for <u>10 min</u>) x 3 times → spin-washing [(0 rpm x 60s (dripping 500 µl CB)→4000 rpm x 30s (dripping 1000 µlx 3 times CB)]x 3 times

<u>Method 2</u>: Dip-washing (Dipping in CB for <u>5 min</u>) x 3 times → spin-washing [(0 rpm x 60s (dripping 500 µl CB)→4000 rpm x 30s (dripping 1000 µlx 3 times CB)]x 3 times

<u>Method 3</u>: Spin-washing [(0 rpm x 60s (dripping 500 µl CB)→4000 rpm x 30s (dripping 1000 µlx 3 times CB)]x 2 times

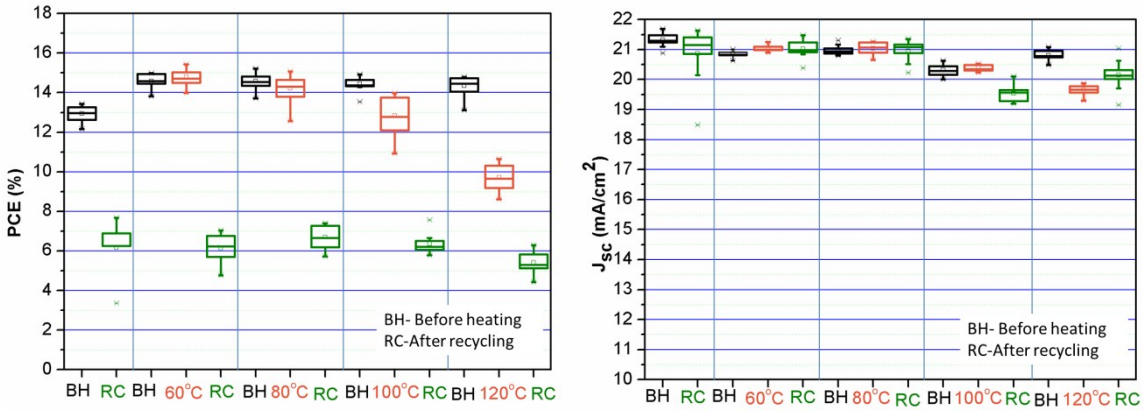


(a) (b)

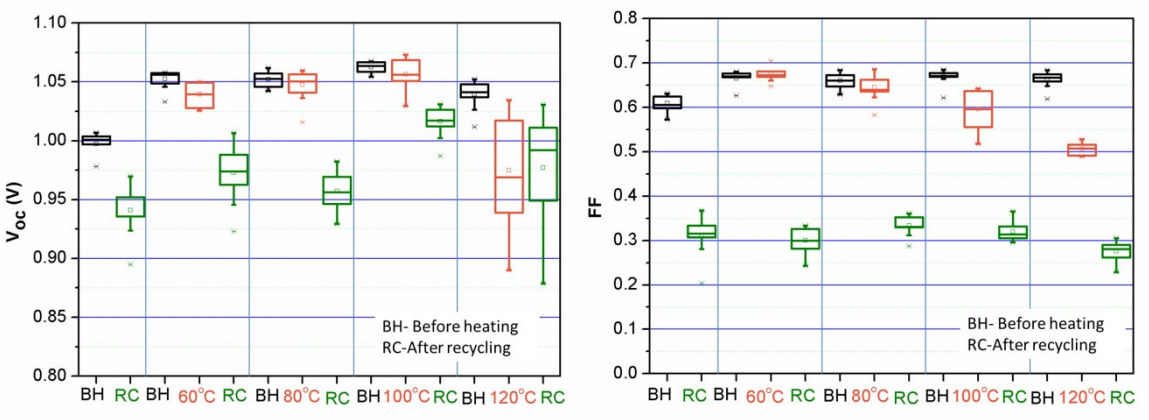


(c) (d)

Figure S6 Backward scan (a) PCE, (b) J_{sc}, (c) V_{oc}, and (d) FF of planar perovskite solar cells after recycled by method 1, method 2 and method 3.

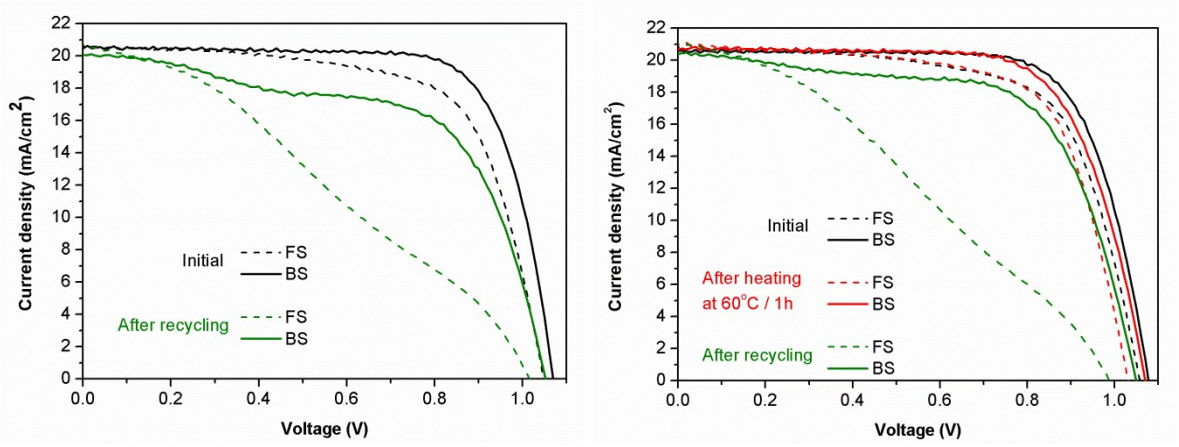


(a) **(b)**



(c) **(d)**

Figure S7 Forward scan (a) PCE, (b) J_{sc}, (c) V_{oc}, and (d) FF of planar $MAPbI_3$ perovskite solar cells before heating (BH), after heating at different temperatures (60, 80, 100 and 120°C) and after recycling (RC) by replacing the aged/degraded spiro-OMeTAD layer with fresh one.



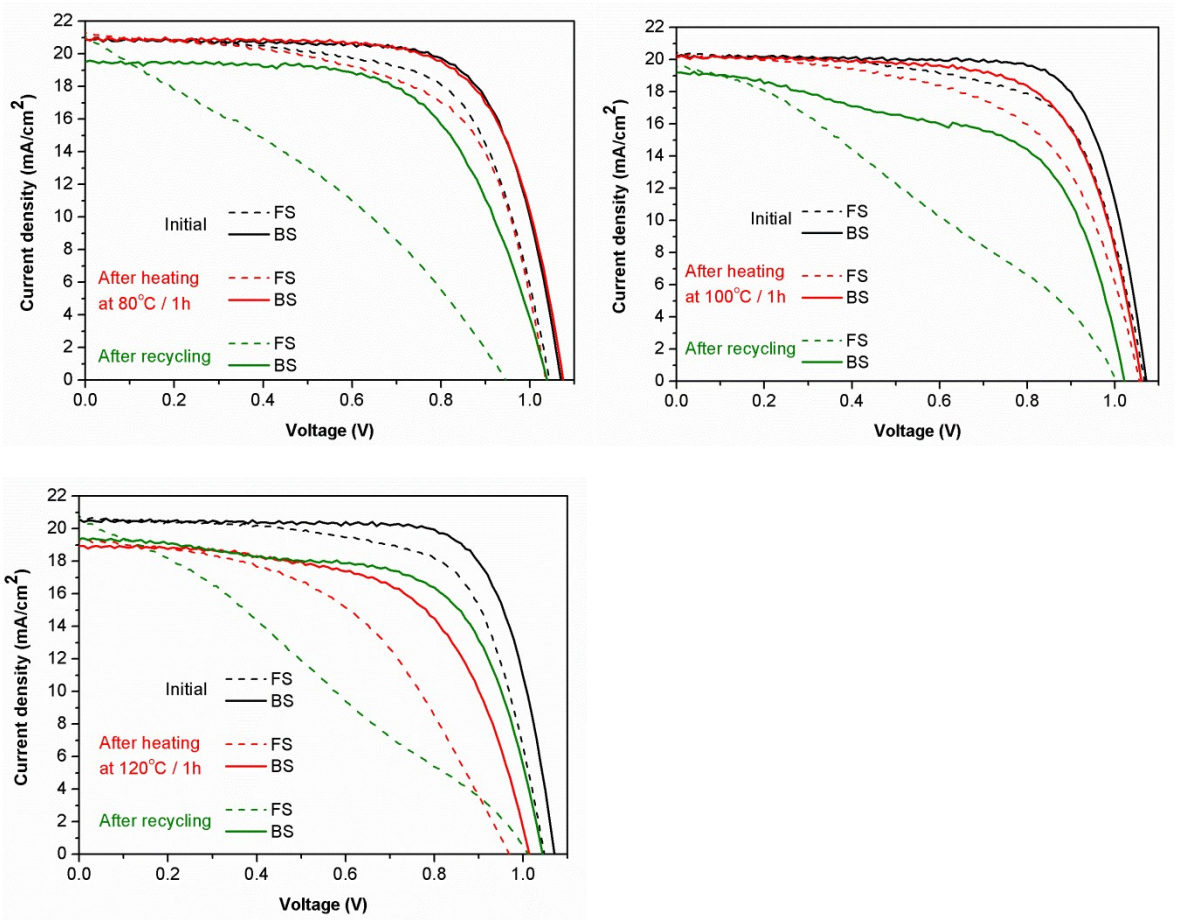
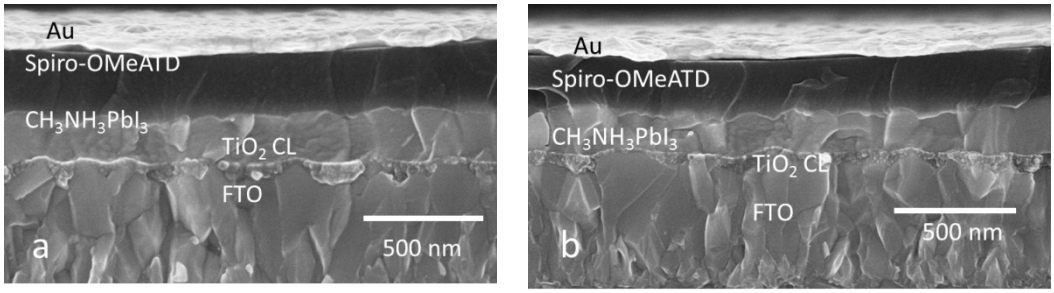


Figure S8 Forward and backward J-V curves of planar perovskite solar cells before heating at different temperatures and after recycling



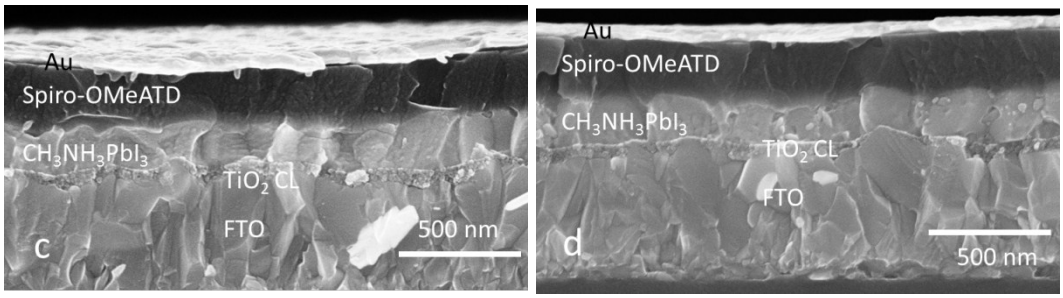


Figure S9 Cross-sectional SEM micrographs of planar $MAPbI_3$ perovskite solar cells recycled (with a fresh spiro-OMeTAD layer) after (a) without heating and heating at (b) 80, (c) 100 and (d) 120°C for 1 hour.

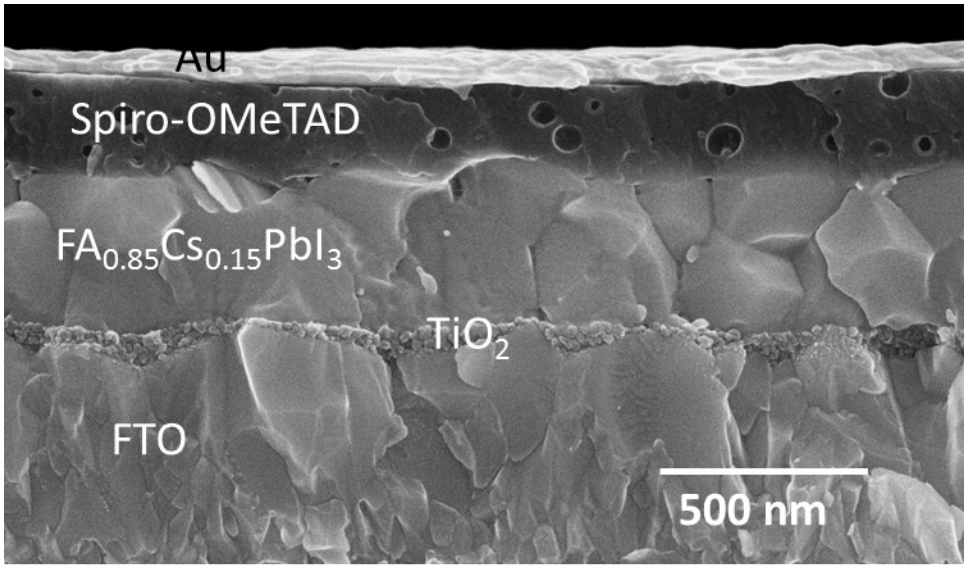


Figure S10 Cross-sectional SEM micrographs of planar $FA_{0.85}Cs_{0.15}PbI_3$ perovskite solar cells after heating at 100°C for 1 hour.

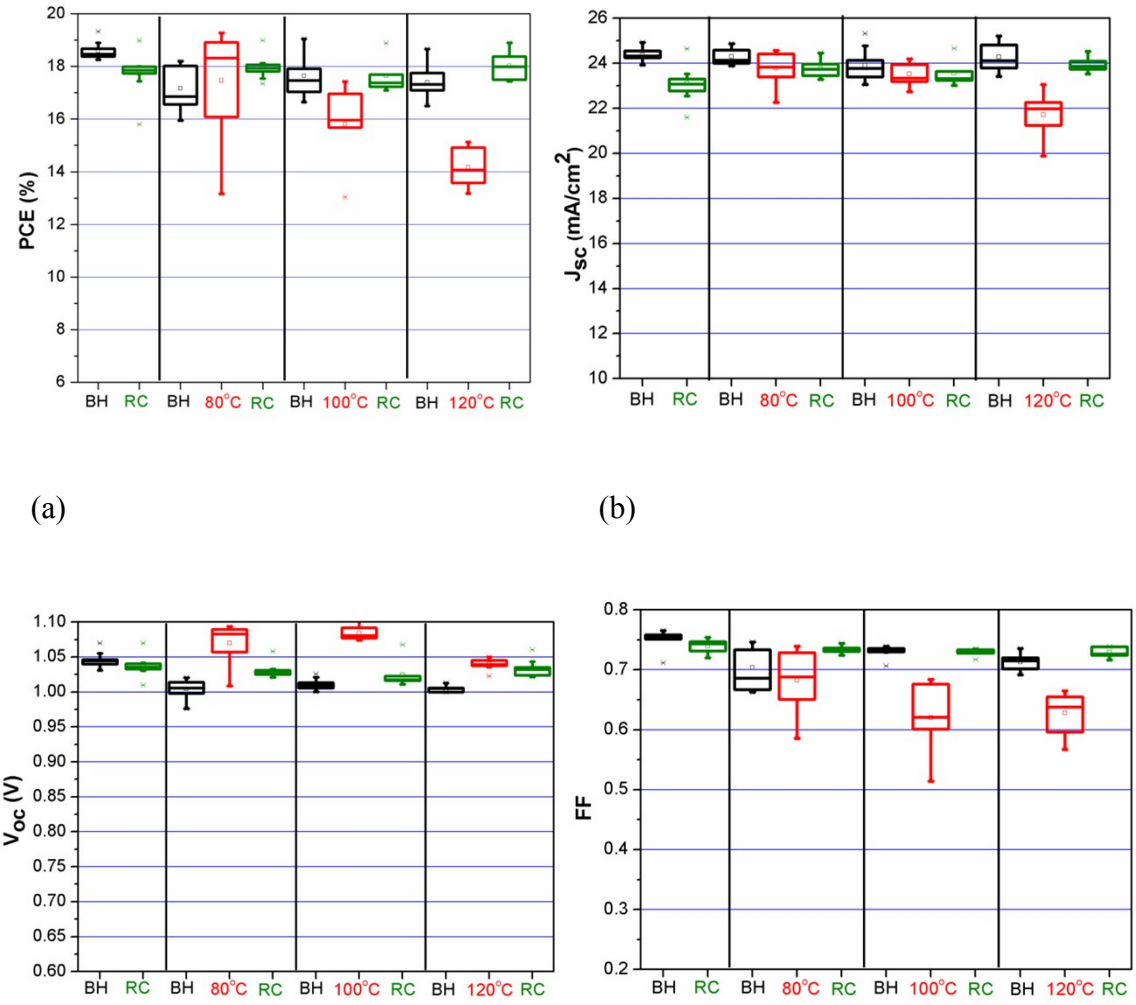


Figure S11 (a) PCE, (b) J_{sc}, (c) V_{oc} and (d) FF of $FA_{0.85}Cs_{0.15}PbI_3$ perovskite solar cells measured on backward scan before heating (BH), after heating at different temperatures (80, 100 and 120 ^oC) and after recycling (RC) by replacing the aged/degraded spiro-OMeTAD layer with fresh one. Corresponding forward scan PCE, J_{sc}, V_{oc} and FF are given in Figure S12.

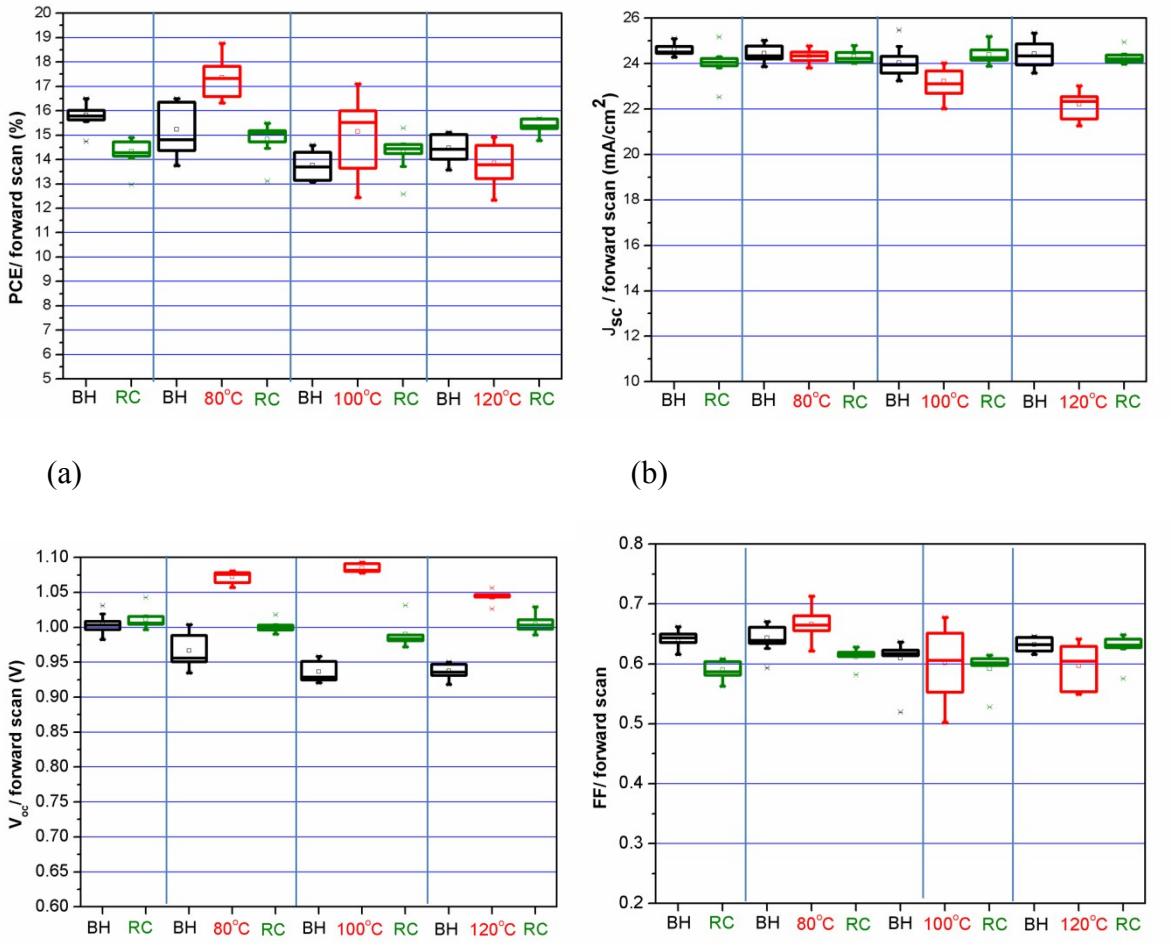


Figure S12 (a) PCE, (b) Jsc, (c) Voc and (d) FF of $FACsPbI_3$ perovskite solar cells measured on forward scan before heating (BH), after heating at different temperatures (80, 100 and 120^{o}C) and after recycling (RC) by replacing the aged/degraded spiro-OMeTAD layer with fresh one.

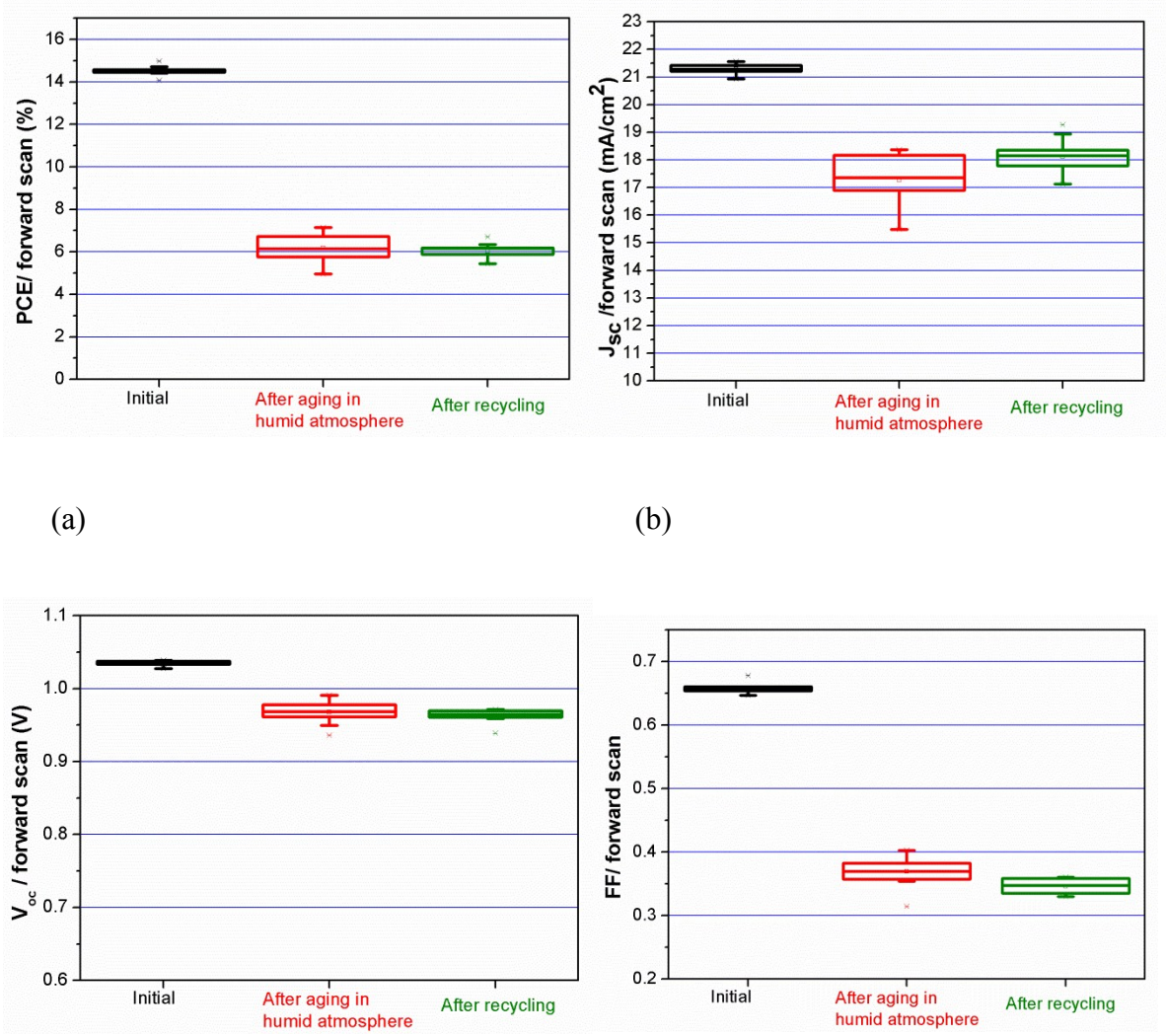
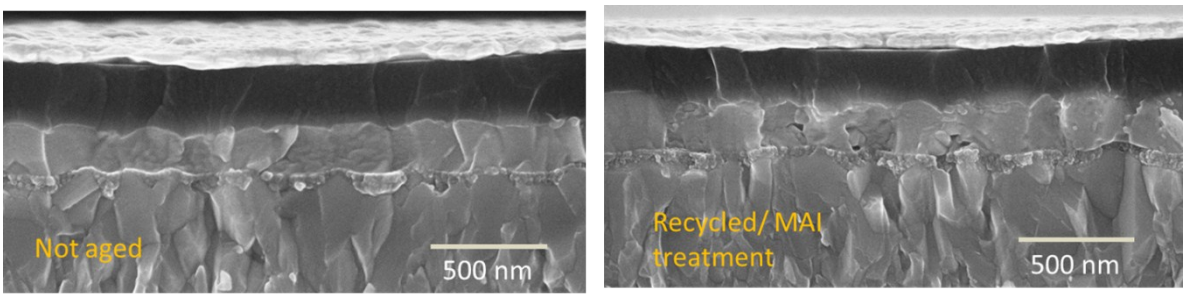


Figure S13 Forward scan (a) PCE, (b) J_{sc}, (c) V_{oc} and (d) FF of planar $MAPbI_3$ perovskite solar cells before and after aging in humid atmosphere and after recycling the degraded cells.



(a) (b)

Figure S14 Cross-sectional SEM micrographs of planar $MAPbI_3$ perovskite solar cells (a) before aging in humid atmosphere and (b) after recycling the degraded cell.