Electronic Supplementary Information

Improvement of Perovskite Crystallinity by Omnidirectional Heat Transfer via Radiative Thermal Annealing

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Fig. S1. (a) Schematic presentation of the RTA steps. (b) Fabrication process of PSCs via RTA. (c) Illustration of a PSC device; fluorine-doped tin oxide (FTO)/compact TiO$_2$ (c-TiO$_2$)/mesoporous TiO$_2$(mp-TiO$_2$)/perovskite;(MAPbI$_3$)$_{0.85}$(FAPbI$_3$)$_{0.15}$/spiro-OMeTAD/Au. (d) A cross-sectional FE-SEM image of PSC.
**Fig. S2.** Structural and photovoltaic (PV) characterization of RTA-perovskite films and RTA-PSCs at different annealing times; (a) Top-view FE-SEM images and (b) XRD patterns. (c) Statistical distribution of PV parameters vs. annealing times. PV parameters of reference PSCs (HPA-PSCs) are also included for comparison.
**Fig. S3.** Temperature-dependence of PL spectra of perovskite films prepared by (a) hot-plate annealing and (b) RTA. The temperature was varied from 60 K to 200 K. The arrows denote the shift of PL emission peak.

Each PL spectrum was normalized to clearly show the evolution of PL emission peak. Unlike the traditional semiconductors such as silicon, perovskites tend to show a blue-shift in the PL emission. The integrated PL intensity to extract the exciton binding energy is estimated by integrating the area of PL emission peak.
Table S1. Best photovoltaic parameters of $V_{oc}$, $J_{sc}$, FF and PCE obtained from 20 PSCs annealed via RTA at different annealing times. The values in parentheses are the averaged photovoltaic parameter values and their standard deviation.

|        | $V_{oc}$ (V)     | $J_{sc}$ (mA cm$^{-2}$) | FF     | PCE (%)         |
|--------|------------------|-------------------------|--------|-----------------|
| 1 min  | 0.99 (1.01 ± 0.05) | 17.66 (17.44 ± 0.91)   | 0.67 (0.63 ± 0.03) | 11.68 (11.05 ± 0.44) |
| 5 min  | 1.05 (1.07 ± 0.01) | 20.91 (20.31 ± 0.83)   | 0.71 (0.66 ± 0.03) | 15.56 (14.35 ± 0.71) |
| 10 min | 1.06 (1.06 ± 0.02) | 24.33 (21.94 ± 0.97)   | 0.68 (0.67 ± 0.02) | 17.37 (15.49 ± 0.95) |
| 15 min | 1.03 (1.02 ± 0.01) | 21.72 (21.16 ± 0.64)   | 0.66 (0.64 ± 0.02) | 14.65 (13.72 ± 0.52) |
| 20 min | 1.04 (0.99 ± 0.05) | 19.49 (19.50 ± 0.31)   | 0.68 (0.63 ± 0.04) | 13.76 (12.27 ± 1.17) |
| Reference | 1.04 (1.04 ± 0.03) | 23.20 (20.26 ± 1.58)   | 0.67 (0.65 ± 0.03) | 16.02 (13.74 ± 1.31) |