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 TiO₂ (c-TiO₂)/mesoporous TiO₂(mp-TiO₂)/perovskite;(MAPbI₃)_{0.85}(FAPbI₃)_{0.15}/spiro-OMeTAD/Au. (d) A cross-sectional FE-SEM image of PSC.



0.60 0.55 0.50

1 5

10

Time (min)

20 Ref

15

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.

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5

10 15 Time (min) Ref



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	$0.99(1.01 \pm 0.05)$	$17.66(17.44 \pm 0.91)$	$0.67 \ (0.63 \pm 0.03)$	$11.68 (11.05 \pm 0.44)$
5 min	$1.05 (1.07 \pm 0.01)$	20.91 (20.31 ± 0.83)	$0.71 \ (0.66 \pm 0.03)$	$15.56~(14.35\pm0.71)$
10 min	$1.06 (1.06 \pm 0.02)$	24.33 (21.94 ± 0.97)	$0.68~(0.67\pm0.02)$	$17.37~(15.49\pm0.95)$
15 min	$1.03 (1.02 \pm 0.01)$	21.72 (21.16 ± 0.64)	$0.66~(0.64\pm0.02)$	$14.65 (13.72 \pm 0.52)$
20 min	$1.04~(0.99\pm0.05)$	$19.49~(19.50\pm0.31)$	$0.68 \ (0.63 \pm 0.04)$	$13.76(12.27 \pm 1.17)$
Reference	$1.04 (1.04 \pm 0.03)$	$23.20(20.26 \pm 1.58)$	$0.67 \ (0.65 \pm 0.03)$	$16.02 (13.74 \pm 1.31)$