#### **Supporting Information**

### Rapid Microwave-Annealing Process of Hybrid Perovskites to Eliminate Miscellaneous Phase for High Performance Photovoltaics

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**Figure S1** Steady-state PL spectra of MAP and HAP perovskite films with different annealing periods on quartz substrates.



Figure S2 Time-resolved PL spectra of MAP-30s and HAP-30m perovskite films on quartz substrates.



**Figure S3** Cross-sectional SEM images of MAP and HAP devices with ITO/perovskite/Au structures for SCLC measurements.



**Figure S4** The distribution curves of  $V_{oc}$ ,  $J_{sc}$ , fill factor (*FF*) and PCEs of 20 independent MAP and HAP PSCs.

**Table S1** Parameters of the time-resolved photoluminescence (TRPL) spectroscopy of perovskite

 films on quartz substrates processed by HAP and MAP with different periods, respectively.

	$\tau_1$ [ns]	A <sub>1</sub> [%]	$\tau_2$ [ns]	A <sub>2</sub> [%]	τ <sub>3</sub> [ns]	A <sub>3</sub> [%]	$\tau_{ave}[ns]$
HAP-30m	16.98	6.16	127.12	25.91	402.69	67.93	307.53
MAP-10s	29.04	4.90	260.97	23.14	1169.84	71.97	903.74

MAP-20s	100.00	3.30	476.48	39.23	1558.19	57.47	1085.71
MAP-30s	139.62	5.39	667.21	40.84	2034.43	53.78	1374.13
MAP-40s	144.23	4.76	600.91	40.22	1871.77	55.02	1278.39
MAP-50s	85.35	4.84	506.44	38.47	1652.23	56.58	1133.87
MAP-60s	34.20	8.28	367.48	39.01	1455.67	53.71	913.46

The PL decay time and amplitudes are modeled using a biexponential expression:

$$f(t) = \sum_{i} A_{i} \exp\left(-\frac{t}{\tau_{i}}\right) + K$$

where  $A_i$  is the decay amplitude,  $\tau_i$  is the decay time and K is a constant for the base-line offset. The average PL decay times ( $\tau_{ave}$ ) are further estimated using the  $\tau_i$  and  $A_i$  values from the fitted curve data **Table S1** using:

$$\tau_{ave} = \frac{\sum A_i \tau_i^2}{\sum A_i \tau_i}$$

**Table S2** Photovoltaic performance of devices fabricated from different processing periods and different microwave output power efficiencies (microwave oven: DAEWOO, KOR-4A6BR, fixed power source type, output efficiencies set on control panel between 20% and MAX by manufacturer's programing on interval spare times). PV parameters were measured from best performing devices under the standard AM 1.5G illumination. <sup>a)</sup> The output power of MAP is 500 W and temperature of HAP is 100°C. <sup>b)</sup> The MAP period is 30 seconds. <sup>c)</sup> The values in the parentheses are the average PCEs from over 20 devices.

Period <sup>a)</sup>	V <sub>oc</sub> [V]	J <sub>sc</sub> [mA/cm <sup>2</sup> ]	FF [%]	PCE [%] <sup>c)</sup>
MAP-10s	1.13 (1.12±0.02)	20.00 (19.59±1.56)	73.38 (70.25±3.95)	16.59 (15.15±1.44)
MAP-20s	1.14 (1.14±0.01)	22.84 (22.06±1.28)	76.23 (73.56±3.57)	19.92 (19.36±0.65)

MAP-30s	1.16 (1.15±0.03)	23.81 (23.06±1.20)	78.52 (76.54±2.74)	21.59 (20.44±1.15)
MAP-40s	1.14 (1.13±0.02)	23.67 (22.65±1.64)	77.83 (75.34±3.18)	20.87 (19.87±1.08)
MAP-50s	1.13 (1.12±0.01)	22.84 (21.86±1.39)	78.48 (75.85±3.26)	20.35 (19.58±0.99)
MAP-60s	1.13 (1.12±0.02)	22.47 (21.52±1.55)	77.96 (74.98±3.52)	19.82 (19.22±0.75)
HAP-30m	1.05 (1.03±0.03)	22.43 (21.23±1.68)	77.53 (74.58±3.19)	18.33 (17.24±1.98)
Output powe efficiency <sup>b)</sup>	r V <sub>oc</sub> [V]	J <sub>sc</sub> [mA/cm2]	FF [%]	PCE [%] <sup>c)</sup>
Output powe efficiency <sup>b)</sup> 20%	r V <sub>oc</sub> [V] 1.15 (1.14±0.02)	J <sub>sc</sub> [mA/cm2] 18.30 (17.59±1.46)	<i>FF</i> [%] 72.12 (70.13±3.22)	PCE [%] <sup>c)</sup> 15.17 (14.25±1.13)
Output powe efficiency <sup>b)</sup> 20% 40%	r V <sub>oc</sub> [V] 1.15 (1.14±0.02) 1.17 (1.15±0.02)	J <sub>sc</sub> [mA/cm2] 18.30 (17.59±1.46) 18.09 (17.66±1.85)	<i>FF</i> [%] 72.12 (70.13±3.22) 75.26 (73.28±3.29)	PCE [%] <sup>c)</sup> 15.17 (14.25±1.13) 15.92 (15.53±0.57)
Output powe efficiency <sup>b)</sup> 20% 40% 60%	r V <sub>oc</sub> [V] 1.15 (1.14±0.02) 1.17 (1.15±0.02) 1.17 (1.15±0.03)	J <sub>sc</sub> [mA/cm2] 18.30 (17.59±1.46) 18.09 (17.66±1.85) 20.55 (20.05±1.87)	<i>FF</i> [%] 72.12 (70.13±3.22) 75.26 (73.28±3.29) 75.17 (74.56±3.74)	PCE [%] <sup>c)</sup> 15.17 (14.25±1.13) 15.92 (15.53±0.57) 18.07 (17.69±0.75)
Output powe efficiency <sup>b)</sup> 20% 40% 60% 80%	r V <sub>oc</sub> [V] 1.15 (1.14±0.02) 1.17 (1.15±0.02) 1.17 (1.15±0.03) 1.17 (1.15±0.02)	J <sub>sc</sub> [mA/cm2] 18.30 (17.59±1.46) 18.09 (17.66±1.85) 20.55 (20.05±1.87) 22.69 (21.98±1.56)	<i>FF</i> [%] 72.12 (70.13±3.22) 75.26 (73.28±3.29) 75.17 (74.56±3.74) 76.54 (75.66±2.88)	PCE [%] <sup>c)</sup> 15.17 (14.25±1.13) 15.92 (15.53±0.57) 18.07 (17.69±0.75) 20.31 (19.45±0.98)

**Table S3** Fitted values of different electronic parameters from dark Nyquist plots of HAP and MAPdevices.

	R <sub>s</sub> (Ω·cm²)	$R_{\rm C}(\Omega \cdot {\rm cm}^2)$	$R_{\rm REC}$ ( $\Omega \cdot {\rm cm}^2$ )	C <sub>c</sub> (nF/cm <sup>2</sup> )	$C_{\mu}$ (nF/cm <sup>2</sup> )
Device	(Deviations)	(Deviations)	(Deviations)	(Deviations)	(Deviations)
HAP	3.58 (7.67%)	24.32 (5.81%)	291.40 (1.14%)	236.66 (3.63%)	44.00 (1.08%)
MAP	1.83 (9.37%)	15.72 (1.71%)	515.20 (1.93%)	208.48 (3.07%)	40.91 (2.02%)



Figure S5 Photograph of the 10mm ×10mm large-scale perovskite films treated by MAP.



**Figure S6** Steady-state PL spectra of MAP perovskite films measured form twelve different spots of the large area perovskite films on glass substrates.



**Figure S7.** a) The microwave oven in the glovebox; b) The inside temperature of the glovebox controlled by air conditioner in an enclosed space.