Electronic Supplementary Information

for

Control of Preferred Orientation with Slow Crystallization for Carbon-based Mesoscopic Perovskite Solar Cells Attaining Efficiency 15 %

Cheng-Min Tsai, Guan-Wei Wu, Sudhakar Narra, Hung-Ming Chang,

Nayantara Mohanta, Hui-Ping Wu, Chien-Lung Wang and Eric Wei-Guang Diau*

¹Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, Hsinchu 30010, Taiwan; Email: diau @mail.nctu.edu.tw

SC temperature	$J_{\rm SC}$ /mA cm ⁻²	V _{OC} /mV	FF	η /%
10°C	1.09	693	0.596	0.5
20°C	21.27	880	0.754	14.1
30°C	11.33	819	0.497	4.6

Table S1. Photovoltaic parameters of SC devices for corresponding current-voltagecurves shown in Fig. S3

Table S2. Photovoltaic parameters of SC devices for corresponding current-voltage curves shown in Fig. S6

SC humidity	$J_{ m SC}$	V _{OC}	FF	η
SC number	$/mA \text{ cm}^{-2}$	/mV		/%
30%	19.20	879	0.680	11.5
50%	22.58	891	0.720	14.5
70%	0.06	387	0.545	0.01

Table S3. Photovoltaic parameters of SC devices with corresponding current-voltage curves shown in Fig. 2a

SC period	$J_{ m SC}$	V _{OC}	FF	η
/h	$/mA \text{ cm}^{-2}$	/mV		/%
0	1.27	750	0.511	0.5
24	3.57	798	0.676	1.9
72	13.77	840	0.745	8.6
96	20.43	841	0.734	12.6
120	22.43	893	0.747	15.0

Table	S4 .	Photovoltaic	parameters	of	devices	obtained	from	current-voltage	curves
shown	in F	ig. 6a							

Deposition method	Scan	$J_{ m SC}$	V _{OC}	FF	η
	direction	$/mA \text{ cm}^{-2}$	/mV		/%
	OC to SC	12.893	823	0.622	6.6
IA (DMF)	SC to OC	12.549	811	0.616	6.3
acquantial	OC to SC	20.189	861	0.668	11.6
sequential	SC to OC	19.740	858	0.644	10.5
$SC(\mathbf{N}\mathbf{M}\mathbf{D})$	OC to SC	22.426	893	0.747	15.0
SC (INMP)	SC to OC	22.396	893	0.731	14.6



Fig. S1 Photographs of SC devices for crystal growth from 0-120 h under conditions of fixed humidity 50 % with temperatures 10, 20 and 30 $^{\circ}$ C.



Fig. S2 SEM side-view images of SC devices at growth duration 120 h stored at fixed humidity 50 % with temperature/°C (a)10, (b) 20 and (c) 30.



Fig. S3 Current-voltage characteristics of SC devices at growth duration 120 h stored at fixed humidity 50 % with temperature/°C 10, 20 and 30 (active area 0.09 cm²) under AM 1.5 G one-sun irradiation; the corresponding photovoltaic parameters are shown in Tables S1.



Fig. S4 Photographs of SC devices for crystal growth from 0-120 h under conditions fixed temperature 20 °C and relative humidity 30, 50 and 70 %



Fig. S5 SEM side-view images of SC devices at growth duration 120 h stored at fixed temperature 50 °C and relative humidity (a) 30, (b) 50 and (c) 70 %.



Fig. S6 Current-voltage characteristics for SC devices at growth duration 120 h stored at temperature 20 °C and relative humidiy 30, 50 and 70 % (active area 0.09 cm²) under AM 1.5 G one-sun irradiation; the corresponding photovoltaic parameters are shown in Tables S2.



Fig. S7 Simulation results via the TOPAS program for XRD patterns of SC devices prepared from treatments: (a) SC (NMP), (b) SC (NMP) + TA and (c) TA (NMP). Blue and red traces are the experimental and calculated data, respectively; grey traces represent the differences between calculation and experiment; blue shadow patterns represent the data that were excluded from the simulation.



Fig. S8 Simulation results via TOPAS program for XRD patterns of SC devices prepared from precursor solvents (a) NMP, (b) GBL, (c) DMF and (d) DMSO. Blue and red traces are the experimental and calculated data, respectively; grey traces represent the differences between calculation and experiment; the blue shadow patterns represent the data that were excluded from the simulation.



Fig. S9 Photovoltaic performance of the SC(NMP) devices without encapsulation as a function of time showing the enduring stability in the dark at 25 °C (black squares) and 70 °C (red circles) as well as under one-sun irradiation at 25 °C (blue triangles).