

Supporting Information

Inducing Swift Nucleation and Morphology Control for Efficient Planar Perovskite Solar Cells by Hot-Air Quenching

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Experimental detail

a. *Chemicals and materials.* Zinc powder, titanium isopropoxide, aluminum oxide particle solution in isopropanol, lead (II) chloride, 4-tert-butylpyridine (tBP), bis(trifluoromethane)sulfonimide lithium salt (Li-TFSI), 2-methoxyethanol, hydroiodic acid, ethanol, isopropanol (IPA), chlorobenzene and dimethylformamide (DMF) were purchased from Sigma-Aldrich. Fluorine-doped SnO₂ (FTO) glass was purchased from Solaronix SA. Methylammonium iodide (CH₃NH₃I) are purchased from Dysol.

b. *Perovskite synthesis.* CH₃NH₃I_{3-x}Cl_x perovskite was synthesized following a reported method¹. 36 wt% of CH₃NH₃I and lead (II) chloride (PbCl₂) were dissolved in anhydrous N,N-dimethylformamide (DMF) at a 3:1 ratio of CH₃NH₃I to PbCl₂.

c. *Device fabrication.* Devices were fabricated on fluorine-doped SnO₂ (FTO) conducting glass (Pilkington TEC 7) 7 Ω/□. Some parts of FTO were treated with 2 M HCl solution containing zinc powder to make insulator regions. The resulting FTO was washed using detergent, acetone, ethanol and IPA and oxygen plasma was done to remove the organic residues. TiO₂ compact layer was prepared by following a reported method.² A solution of 0.5 mM titanium isopropoxide (TTIP) in ethanol and 40 mM HCl solution in ethanol were slowly mixed together. The mixed solution was drop-wised on the substrate and spin-coated at 2000 rpm for 1 min. The TiO₂-coated substrates were heated at 500 °C for 30 min. The perovskite precursor was deposited on the TiO₂ layer and spin coated at 2000 rpm for 20s. During spin coating, heat air flow was applied to the substrate. (Heat air flows are generated by using heatgun. BOSCH GHG 630 DCE) Perovskite layer was crystalized by heating at 100 °C for 2.5 hr in dried condition (<25 % humidity). 2,2',7,7'-tetrakis(N,N'-di-p-methoxyphenylamine)-9,90-spirobifluorene (Spiro-MeOTAD) (Merck KGaA) in chlorobenzene (180 mg/1 ml) solution with Li-TFSI and tBP was spin-coated on the perovskite layer. Finally, 100 nm silver electrode was vacuum-deposited on the spiro-OMeTAD overlayer.

d. *Characterization.* Axioplan microscope (ZEISS) was used for measuring an optical microscope images. Field emission scanning electron microscope (FE-SEM, Hitachi S 4800) was employed for observing the cross and top image of perovskite layer. Time-resolved PL decay measurements were obtained using a FluoTime 300, PicoQuant GmbH. The samples were photoexcited using a 507 nm laser head (LDH-P-C-510, PicoQuant GmbH) pulsed at 1 MHz for glass substrate samples and 10 MHz for FTO substrate samples, with a pulse duration of 117 ps and fluence of 30 nJ/cm²/pulse. The PL was collected from a high resolution monochromator and hybrid photomultiplier detector (PMA Hybrid 40, PicoQuant GmbH). Atomic force microscope (AFM) (Digital Instruments Multimode Nanoscope III) in conductive mode. Photovoltaic performance and SCLC measurement of devices was measured under air AM 1.5G illumination of 100 mW/cm² (Oriel 1 kW solar simulator), which was calibrated with a KG5 filter-covered silicon photovoltaic solar cell traceable to the national renewable energy laboratory (NREL). A mask was used to define the device illumination area of 0.09 cm² to minimize photocurrent generation from the edge of the electrodes. Heat air flow from heatgun considered as free jet and velocity of heat air flow converted to m/s value using following equation.^{S1}

$$\frac{U_m}{U_0} = C \frac{\sqrt{A}}{x}$$

U_m is mean center velocity at distance x, U₀ is velocity at orifice, C is constant, x is distance from orifice and A is cross section area of heatgun. Heatgun gives 50 liters/sec of air flow after it sprayed from orifice (X= 15 cm and A=4.93 cm²) Thus we get 18.34 of U₀C value. We applied heat air flow at x=30 cm. Following the equation, 50 liters/sec air flow changed as 1.35 m/s.

S1. H. Liu, S. H. Winoto and D. A. Shah, Ann. Biomed. Eng., 1997, **25**, 939.

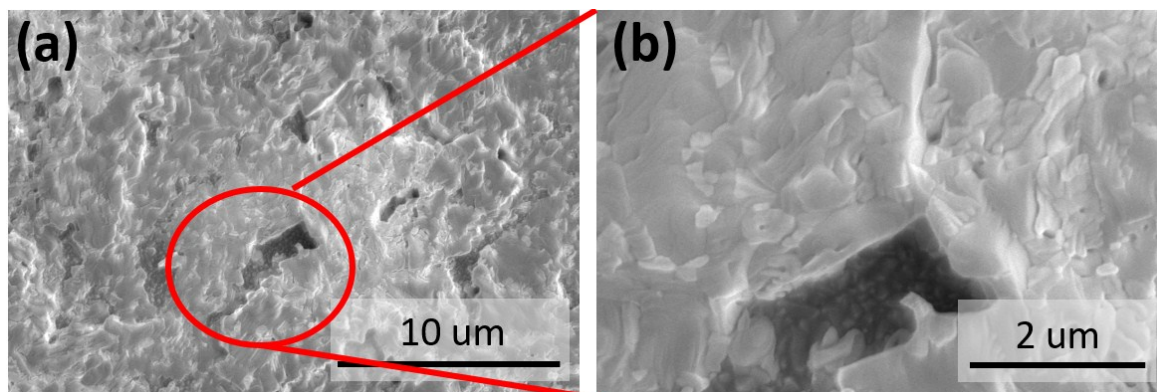


Fig. S1. SEM images of perovskite layer only with room temperature air flow.

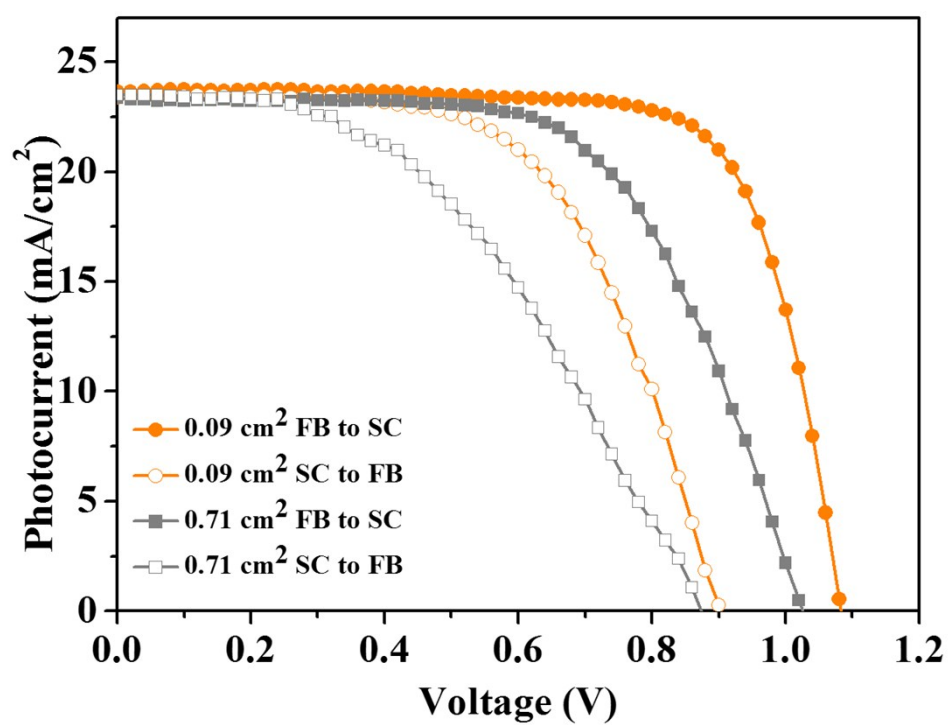


Fig. S2. Hysteresis of small and large area devices. (scan rate: 0.15 V/s)

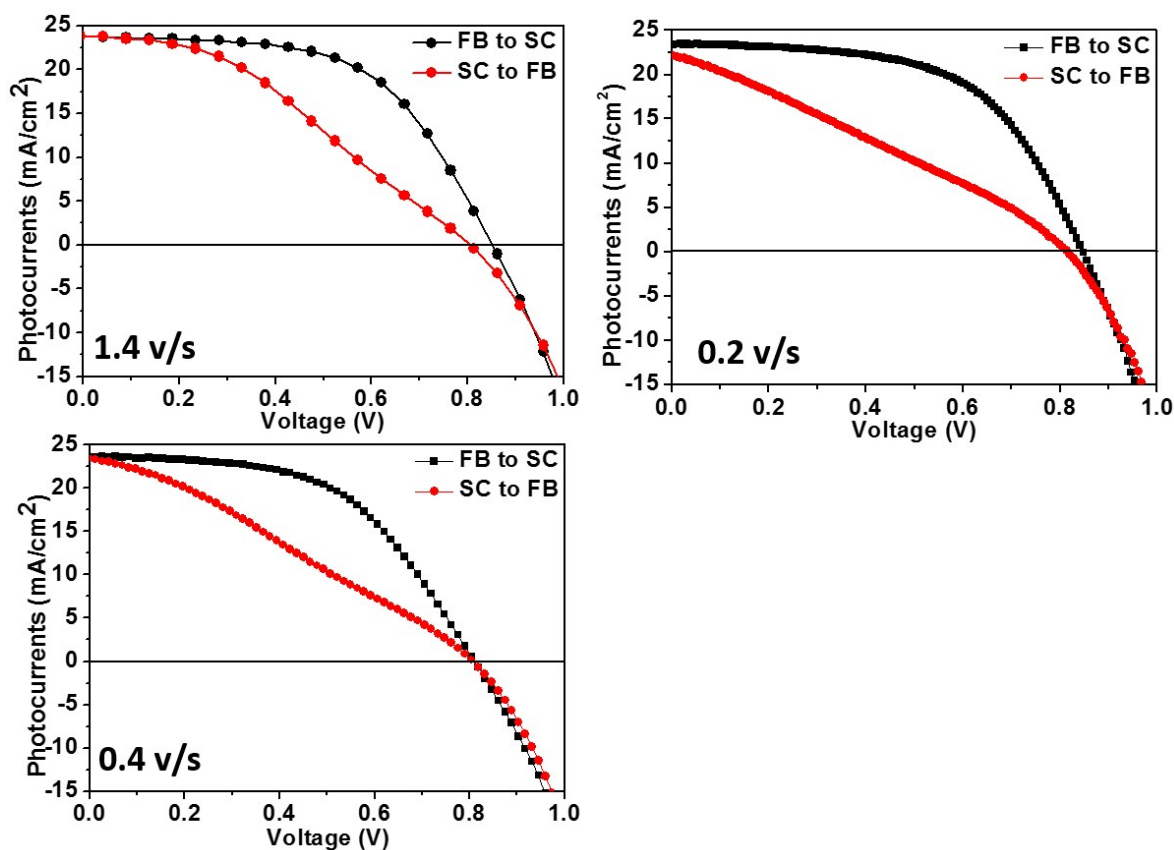


Fig. S3 Hysteresis measurement according to the scan rate.

Table S1. Summary of the J-V best efficiencies (η_{JV}) and the maximum powerpoint (MPP) efficiencies (η_{MPP}) in many literatures among the planer perovskite solar cells employing a spin-coated TiO₂ electron transporting layer and their $\eta_{JV}/\eta_{MPP} * 100$ (%) values.

<i>ref</i>	<i>Efficiency from JV measurement (%)</i>	<i>Efficiency from MPP measurement (%)</i>	$\eta_{JV}/\eta_{MPP} * 100$ (%)
11. <i>J. Phys. Chem. Lett.</i> , 2015, 6 , 2399.	13.6	6.1	44.9
38. <i>Nat. Commun.</i> , 2014, 6 , 7081.	14.2	7.0	49.3
39. <i>Phys. Status Solidi A</i> , 2015, 12 , 2708.	14.2	8.6	60.6
40. <i>Mater. Horiz.</i> , 2015, 2 , 315.	14.2	9.1	64.2
41. <i>ACS Nano</i> , 2014, 8 , 9815.	12.1	8.3	68.6
42. <i>Nano Lett.</i> , 2014, 14 , 5561.	15.3	10.6	69.3
8. <i>ACS Nano</i> , 2016, 10 , 6029.	12.5	8.9	71.2
43. <i>Energy Environ. Sci.</i> , 2015, 8 , 916.	12.3	10.0	81.3
44. <i>Nano Energy</i> , 2016, 28 , 269.	11.0	9.0	81.8
45. <i>Science</i> , 2016, 351 , 151.	15.1	12.5	82.8
46. <i>ACS Photonics</i> , 2015, 2 , 680.	13.0	11.0	84.3
15. <i>Nat. Commun.</i> , 2015, 6 , 6142.	15.2	13.4	88.1
47. <i>Nano Energy</i> , 2015, 13 , 249.	13.6	12.5	91.9
48. <i>Energy Environ. Sci.</i> , 2016, 9 , 484.	13.6	12.6	92.6