Electronic Supplementary Material (ESI) for ChemComm.

Supporting Information

Solvent-assisted Crystallization of Two-dimensional Ruddlesden-Popper Perovskite

Yingjie Su[a], Cai Xu[a], Liguo Gao[a,*], Guoying Wei[b], Tingli Ma[b,c,*]

a State Key Laboratory of Fine Chemicals, Dalian University of Technology, Dalian 116023, China.

b Department of Materials Science and Engineering, China Jiliang University, Hangzhou, 310018, P. R. China.

c Graduate School of Life Science and Systems Engineering, Kyushu Institute of Technology, Kitakyushu, Fukuoka 808-0196, Japan.

Corresponding Author

*Liguo Gao

E-mail: liguo.gao@dlut.edu.cn; ORCID: 0000-0002-5390-3663.

*Tingli Ma

E-mail: tinglima@life.kyutech.ac.jp; ORCID: 0000-0002-3310-459X.

1. Materials

15 Ω·□$^{-1}$ ITO was purchased from Yingkou OPV Tech New Energy Co. PEDOT:PSS (Al 4083), BAI (99.5%), MAI (99.5%) and $PC_{61}BM$ (99.1%) were purchased from Xi'an Polymer Light Technology Corp. PbI_2 (99.9%) was purchased from Advanced Election Technology CO,. Ltd. BCP (95%) were purchased from Aladdin. $PbCl_2$ (99.999%), dimethyl sulfoxide (DMSO, hybridoma, 99.7%), N,N-dimethylformamide (DMF, for HPLC, 99.9%), Chlorobenzene (for HPLC, 99.9%) and isopropanol (99.5%) were purchased from Sigma-Aldrich.

2. Device Fabrication

PEDOT:PSS was spin-coated on clean and UV-ozone-treated ITO at 5000 rpm for 30 s, and then annealed at 130°C for 20 min. Under N_2 atmosphere, the composition of BAI:MAI:$PbCl_2$:PbI_2 with a stoichiometric ratio of 2:3:0.08:3.92 was dissolved in DMF to prepare a perovskite precursor solution with a concentration of 0.8 M.

Control method to deposit perovskite film: the 2D perovskite films were prepared by a one-step method with a spin speed of 5000 rpm for 30 s. Solvent-assisted method to deposit perovskite film: drop 160 μL DMSO on the ITO with PEDOT:PSS, spin-coated at a low speed of 1000 rpm for 6 s, and dynamic spin-coated 60 μL the perovskite precursor solution at the 5th second, and then spin at a high speed of 5000 rpm for 10 s, and then anneal at 100°C for 15 min. PCBM (20 mg mL^{-1} in CB) and BCP (0.5 mg mL^{-1} in IPA) were spin-coated at 2000 rpm and 5000 rpm for 30 s, and then annealed at 75°C for 10 min and 5 min, respectively. Finally, 70 nm silver was deposited as a metal electrode in a vacuum environment.

3. Characterization

Keithley 2460 (Keithley, America) was used to measure the *J-V* characteristics

of cells at AM 1.5G. The Incident photon-to-electron conversion efficiency (IPCE) is measured by using a computer-controlled xenon lamp combined with a monochromator (PEC-S20, Peccell). Photoluminescence (PL) spectra is measured by C5410 (Hamamatsu) at an excitation wavelength of 495 nm. The scanning electron microscopy (SEM) images were measured on a high-resolution field emission Nano SEM 450 (FEI, USA). The crystal structure and light absorption of 2D perovskite were measured using X-ray diffraction (XRD) (XRD-7000S Shimadzu, Japan) and UV-vis spectral (Lambda950), respectively. EIS/TPC/TPV/IMPS/IMVS are all measured using electrochemical workstation (Zennium Zahner, Germany).

Figures

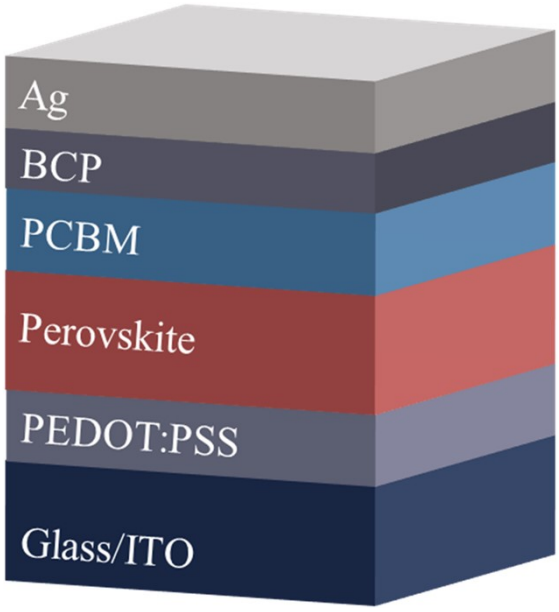

Fig. S1. Schematic diagram of devices with the structure of Glass/ITO/PEDOT:PSS/perovskite/$PC_{61}BM$/BCP/Ag.

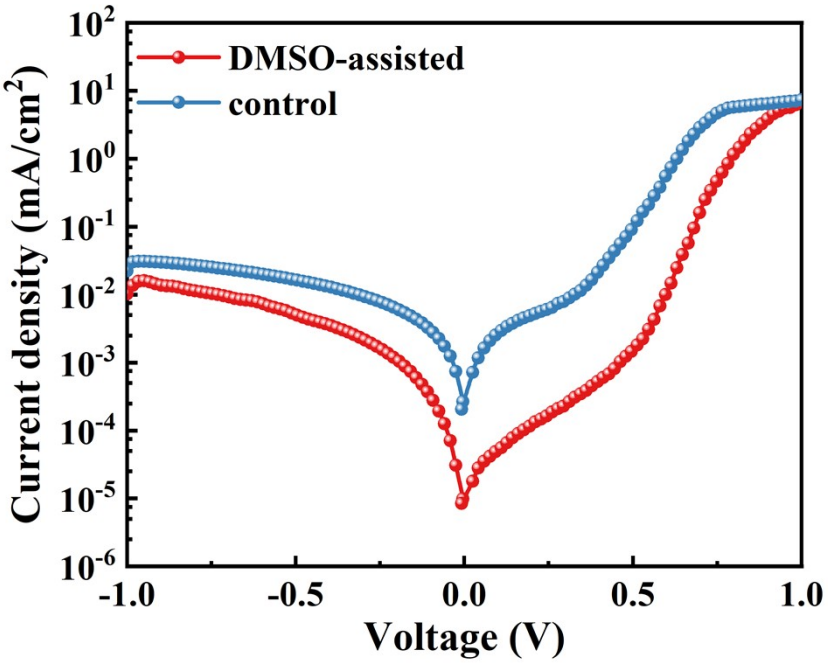

Fig. S2. Dark *J*–*V* curves.

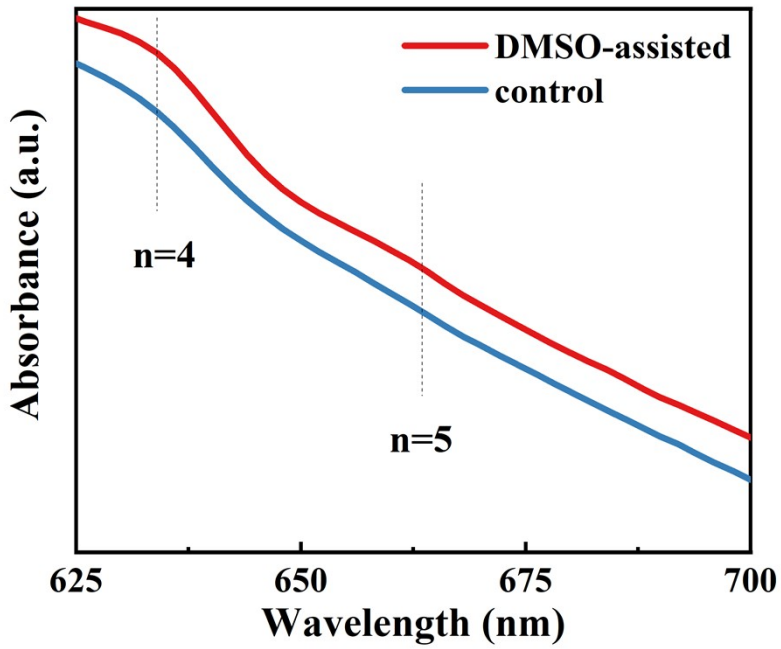

Fig. S3. UV-vis absorption characteristics of perovskite films prepared by different methods.

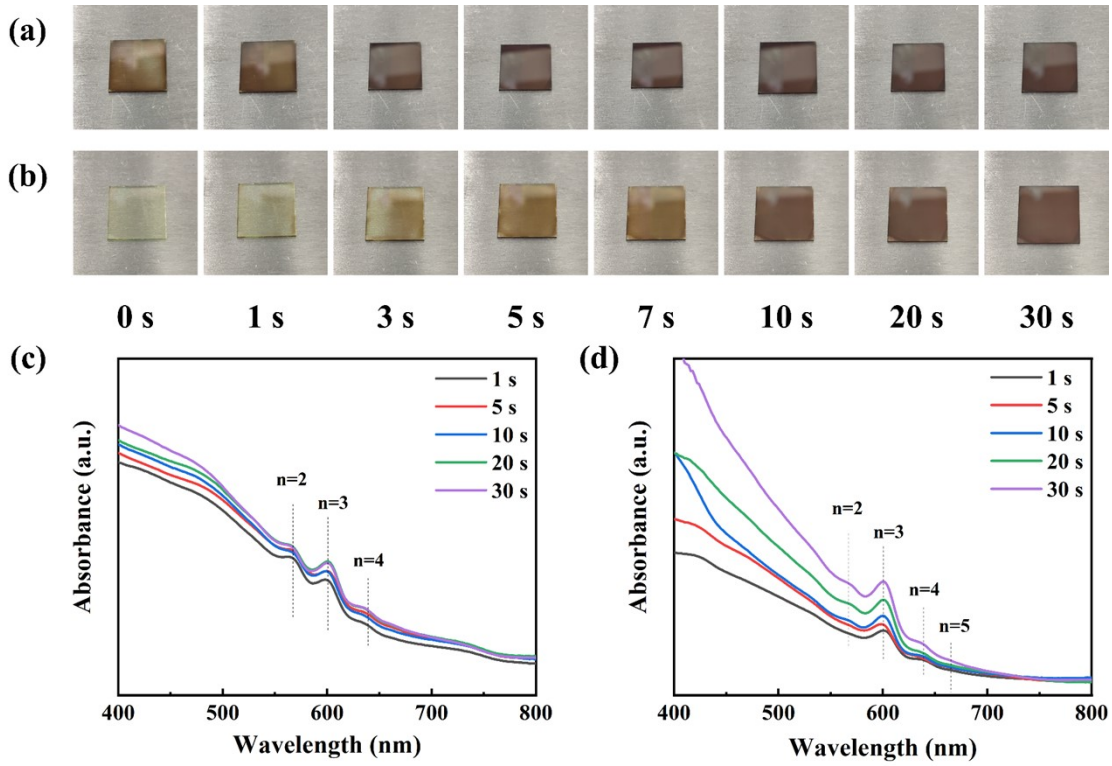

Fig. S4. Optical images for perovskite crystalline growth with different time on 100℃ hot plate, (a) control; (b) DMSO-assisted. UV-vis absorption spectra of perovskite films with time variation, (c) control, (d) DMSO-assisted.

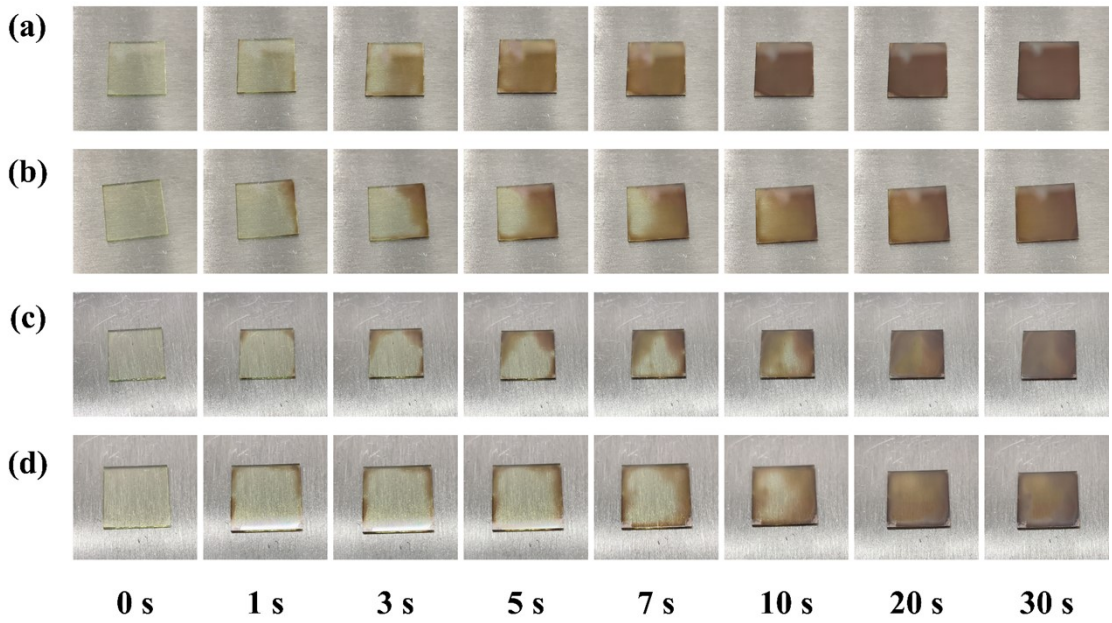

Fig. S5. Optical images for perovskite crystalline growth with different time on 100℃ hot plate, (a) DMSO-assisted; (b) control-8:2 (DMF:DMSO); (c) control-7:3 (DMF:DMSO); (d) control-5:5 (DMF:DMSO).

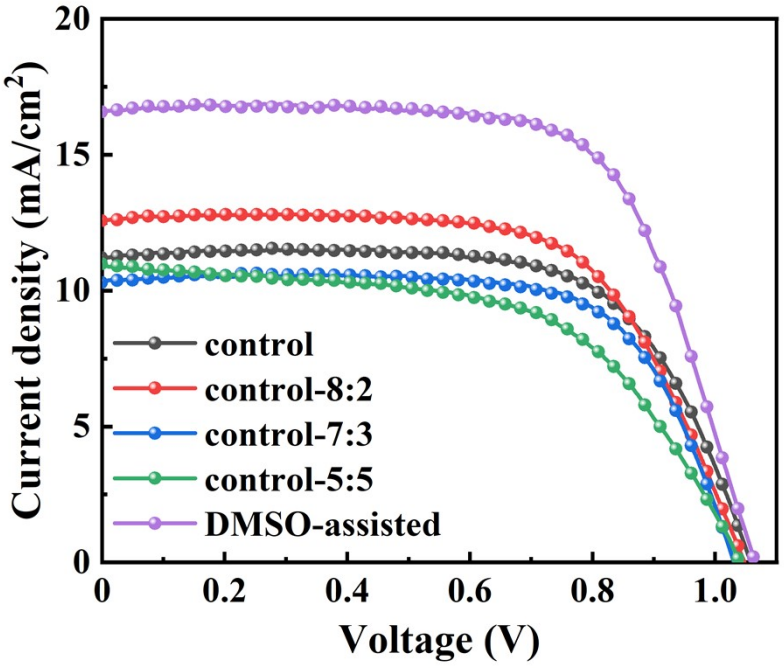

Fig. S6. *J-V* curves of devices with different preparation methods, corresponding to Figure S3.

Table S1. Photovoltaic parameters of solar cells based on $(BA)_2(MA)_3Pb_4(I_{0.98}Cl_{0.02})_{13}$ perovskite films prepared by different processes.

		V_{oc} [V]	J_{sc} [mA/cm^2]	FF	PCE [%]
DMSO-assisted	best	1.05	17.17	0.69	12.42
	average	1.05±0.02	15.82±1.31	0.68±0.04	11.31±0.67
control	best	1.04	11.81	0.68	8.37
	average	1.04±0.03	10.18±1.66	0.61±0.06	6.40±1.07

There are 30 devices taken into account to calculate the average parameters of devices.

SI 1. The density of defect states is calculated by the dark *J-V* curve of the device with the

structure of ITO/SnO_2/2D PVK/$PC_{61}BM$/BCP/ Ag. The trap-state density was determined from the following equation: $$N_t = \frac{2\epsilon_0\epsilon_r V_{TFL}}{qL^2}$$

where N_t is the trap density, V_{TFL} is the trap-filling limit voltage, and q is the elemental charge.