Supporting Information

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

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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 PC₆₁BM (99.1%) were purchased from Xi’an Polymer Light Technology Corp. PbI₂ (99.9%) was purchased from Advanced Election Technology CO., Ltd. BCP (95%) were purchased from Aladdin. PbCl₂ (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 N₂ atmosphere, the composition of BAI:MAI:PbCl₂:PbI₂ 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.](image-url)
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°C 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°C 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)$_3$Pb$_4$(I$_{0.98}$Cl$_{0.02}$)$_{13}$ perovskite films prepared by different processes.

<table>
<thead>
<tr>
<th>Process</th>
<th>$V_{oc}$ [V]</th>
<th>$J_{sc}$ [mA/cm$^2$]</th>
<th>FF</th>
<th>PCE [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>best</strong></td>
<td>1.05</td>
<td>17.17</td>
<td>0.69</td>
<td>12.42</td>
</tr>
<tr>
<td>DMSO-assisted</td>
<td>average</td>
<td>1.05±0.02</td>
<td>15.82±1.31</td>
<td>0.68±0.04</td>
</tr>
<tr>
<td>control</td>
<td><strong>best</strong></td>
<td>1.04</td>
<td>11.81</td>
<td>0.68</td>
</tr>
<tr>
<td>average</td>
<td>1.04±0.03</td>
<td>10.18±1.66</td>
<td>0.61±0.06</td>
<td>6.40±1.07</td>
</tr>
</tbody>
</table>

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/SnO₂/2D PVK/PC₆₁BM/BCP/Ag. The trap-state density was determined from

$$N_t = \frac{2\varepsilon_0 \varepsilon_r V_{TFL}}{q L^2}$$

the following equation:

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