

Supporting Information

Soft-cover deposition of scaling-up uniform perovskite thin films for high cost-performance solar cells

Fei Ye^{†a}, Han Chen^{†a}, Fengxian Xie^{†b}, Wentao Tang^a, Maoshu Yin^a, Jinjin He^a,
Enbing Bi^a, Yanbo Wang^a, Xudong Yang^{*a}, Liyuan Han^{*b}

Affiliations:

^a State Key Laboratory of Metal Matrix Composites, Shanghai Jiao Tong University,
800 Dong Chuan RD. Minhang District, Shanghai 200240, China. Email:
yang.xudong@sjtu.edu.cn

^b Photovoltaic Materials Unit, National Institute for Materials Science, Tsukuba,
Ibaraki 305-0047, Japan. Email: han.liyuan@nims.go.jp

[†] These authors contributed equally to this work

Calculations of material usage.

For spin-coating, according to the normal protocol, 1.45 M perovskite precursor was used and about 60 μl precursor was added on a substrate of $2 \times 2 \text{ cm}^2$. If the final perovskite film is about 600 nm, then the total mass of the final perovskite can be expressed by Equation 1

$$m_1 = \rho Sh \quad \text{Equation 1}$$

Where, m_1 is the mass of final perovskite film; ρ is perovskite density, 4.3 g/cm^3 ; S is the substrate area, 4 cm^2 ; and h is the film thickness, 350 nm. The calculated mass of the final perovskite film is about 0.60 mg.

While the mass of perovskite in the original added precursor can be expressed in Equation 2

$$m_0 = CVM \quad \text{Equation 2}$$

Where m_0 is the mass of perovskite in the original added precursor, C is the precursor concentration, 1.45 M; V is the added volume, 60 μl ; and M is the molar mass of perovskite, 620 g/mol. The calculated mass of perovskite in the original added precursor is about 53.94 mg. The material utilization ratio is the ratio of the final perovskite film mass to the perovskite mass in the added volume of perovskite precursor. Thus the material utilization ratio of the perovskite for spin-coating is about 1%.

For SCD, 35 μl 0.5 M precursor was added on $6 \times 8.5 \text{ cm}^2$ substrate to obtain 600nm final perovskite film, the film mass m_1 is about 10.85mg, and the perovskite mass in precursor m_0 is 13.16mg. The utilization ratio for SCD is about 82%.

Programmed mechanical hand.

The programmed mechanical hand was a robot arm with 6 steering engine, which can move at x , y , and z direction and rotate in an angle range of 270° . At the end of the robot arm we installed a clamp, which can bite one end of the PI film. The robot arm is fixed on a screw guide, when the screw guide works, the robot arm moving accordingly. The robot motion, the screw guide action and the clamp bite are all controlled by a computer through a servo controller, STM32F103 microcontroller, which connect all of the 6 steering engines, the clamp and the screw guide with computer, and through the programming of the motion of every steering engine of the

robot hand, the clamp and the screw guide, we can get the expected action of the robot hand. The moving speed of robot arm was controlled by the screw guide. The height of one spiral of the screw guide is 4 mm, and it requires 3200 pulses to move 4 mm. By changing the pulses sent out by the computer every second we can control the speed of the guide screw. For example, if we give 40000 pulses in a period of 1 second, the moving distance is $40000 \times 4 / 3200$ mm, it's about 50mm. Thus the peeling speed is about 50mm/s. For a simpler set-up, we also refitted the Interconnect dispenser, WS 1100A (Worldwide) by installing the clamp at the nozzle position. The dispenser's arm can be controlled by its separate controller. Thus, it's only the clamp that needed to be controlled on computer, which simplified the programming process.

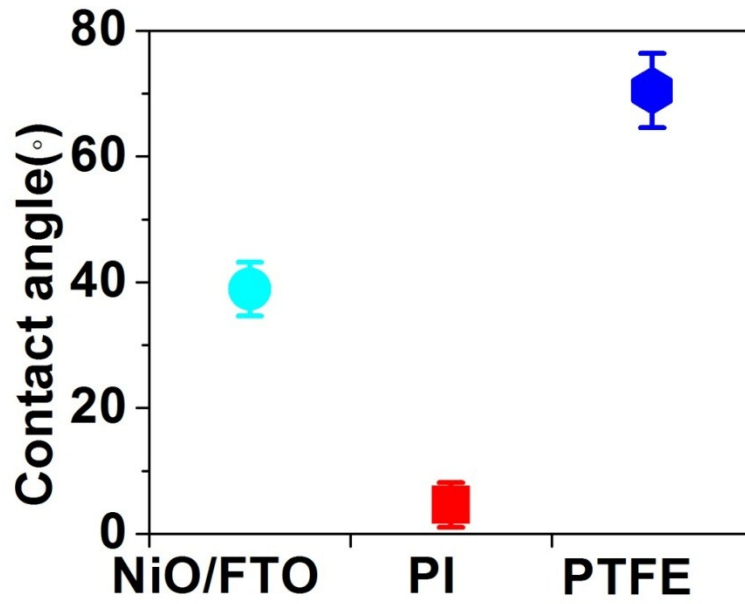


Fig. S1 The contact angles for 2 μl perovskite precursor droplets on NiO/FTO substrate, polyimide film (PI), Polytetrafluoroethylene (PTFE). The contact angle on PI film is the smallest among the representative films, and much smaller than that on NiO/FTO substrate, which brings the precursor greatly spreading on the substrate.

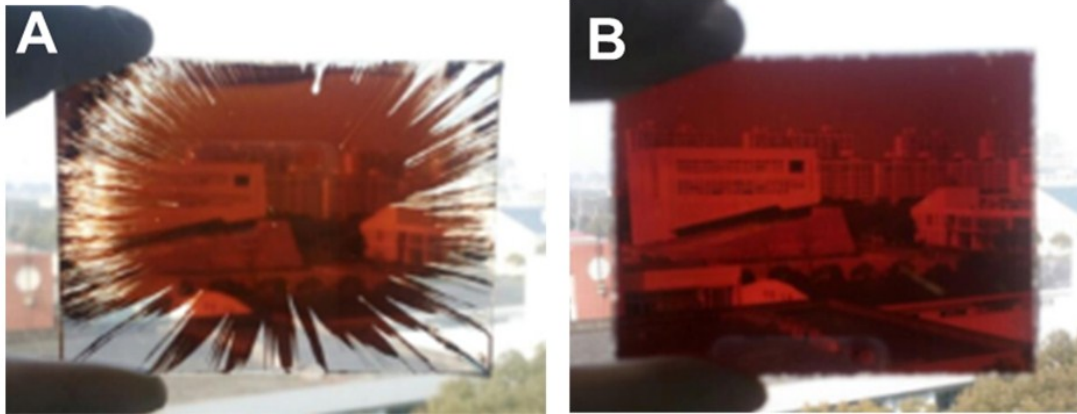


Fig. S2 The appearance of perovskite film by spin-coating on a 51 cm² substrate, using the same precursor amount of 35 μ l as that in SCD. The spin-coated perovskite film was prepared with a 1200 rpm speed for 12 s plus a 5000 rpm speed for 40 s, and 1 ml toluene was dropped at the 22 s rotation. After spin-coating, the perovskite film was heated for 10min at 100°C, and the whole preparation process was conducted in N₂-filled glove box.

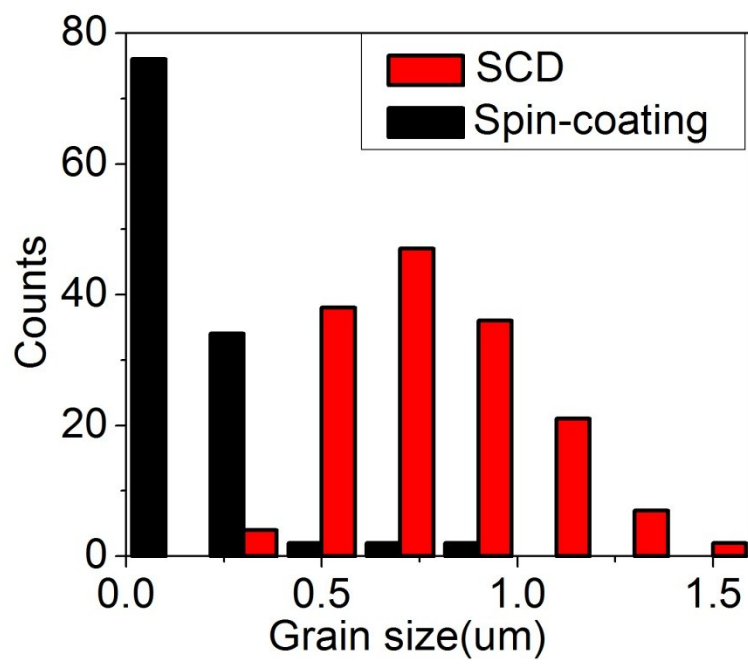


Fig. S3 The distributions of perovskite grain size for SCD and spin-coating based films.

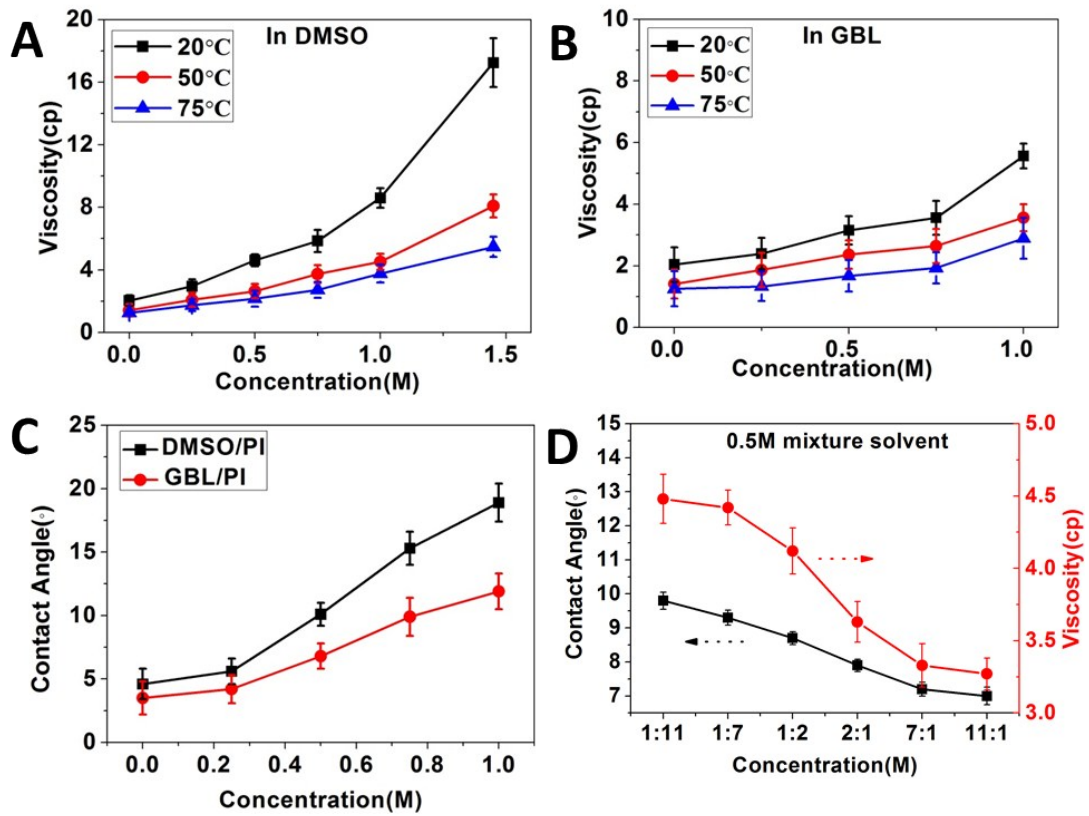


Fig. S4 The viscosities of perovskite precursors in DMSO and GBL changes according to the concentration at the temperature of 20 °C, 50 °C, 70 °C (A and B), and the contact angles of perovskite precursor in DMSO and GBL with different concentrations at room temperature and atmospheric pressure (C), in which DMSO/PI represents the precursor in DMSO dropping on PI film, and GBL/PI represents the precursor in GBL dropping on PI film. And the contact angles and viscosity of 0.5M precursor with GBL/DMSO in the molar ratios of 1:11, 1:7, 1:2, 2:1, 7:1 and 11:1 (D). The larger viscosity benefits the convection suppression in SCD, while the precursors with larger viscosity have poor wettability on PI film, since the contact angle increases when the precursor concentration increases, and the addition of some GBL in DMSO balance the viscosity increase and wettability deterioration.

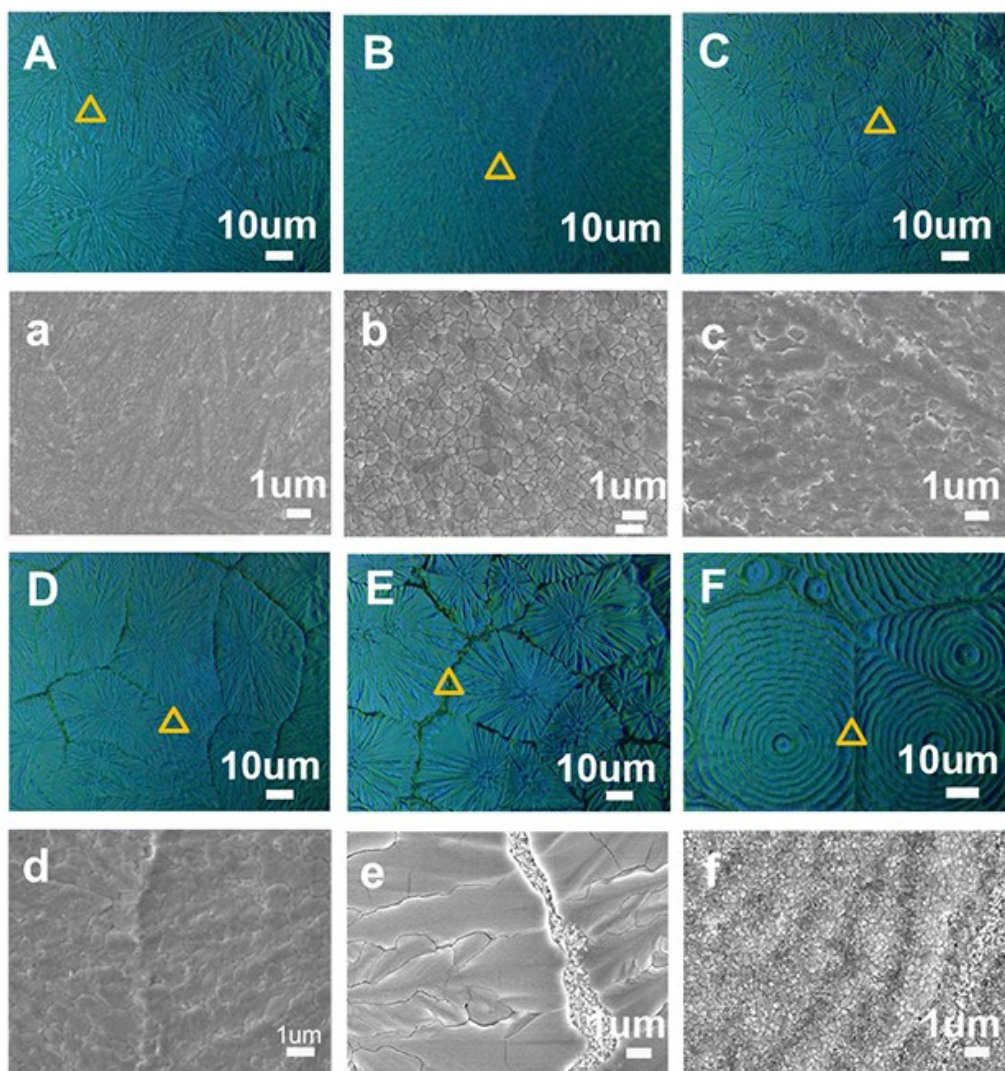


Fig. S5 Top-view optical and SEM images of perovskite films formed by SCD with mixed solvent of GBL and DMSO with the molar ratio of 1:11 (A), (a); 1:7 (B), (b); 1:2 (C), (c); 2:1 (D), (d); 7:1 (E), (e); 11:1 (F), (f). The yellow triangles in optical images indicate the locations of SEM images.

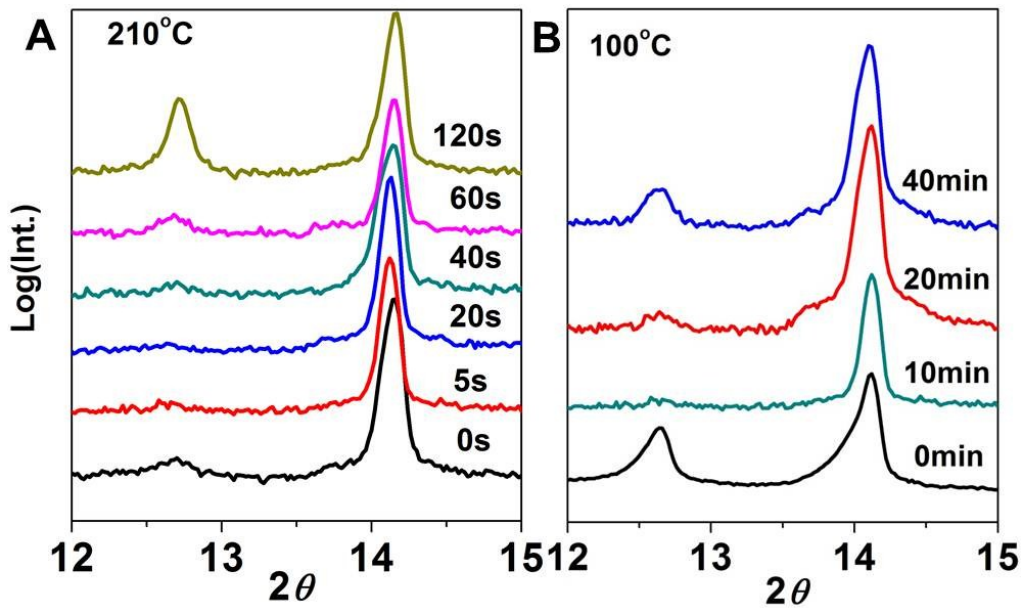


Fig. S6 The X-ray diffraction patterns of 2θ between 12~15 with heat extensions from 0~120 s at 210 °C for perovskite film (A), and the X-ray diffraction patterns of unthoroughly transformed perovskite film prepared at low heating bed temperature (130 °C) for post treatment at 100 °C in the glove box (B). The perovskite prepared at high temperature of 210 °C can convert thoroughly to perovskite after a 20s heat extension. Low temperature (130 °C) prepared perovskite needs a further post heat treatment in glove box, engendering more production time, complicated process and expensive equipment.

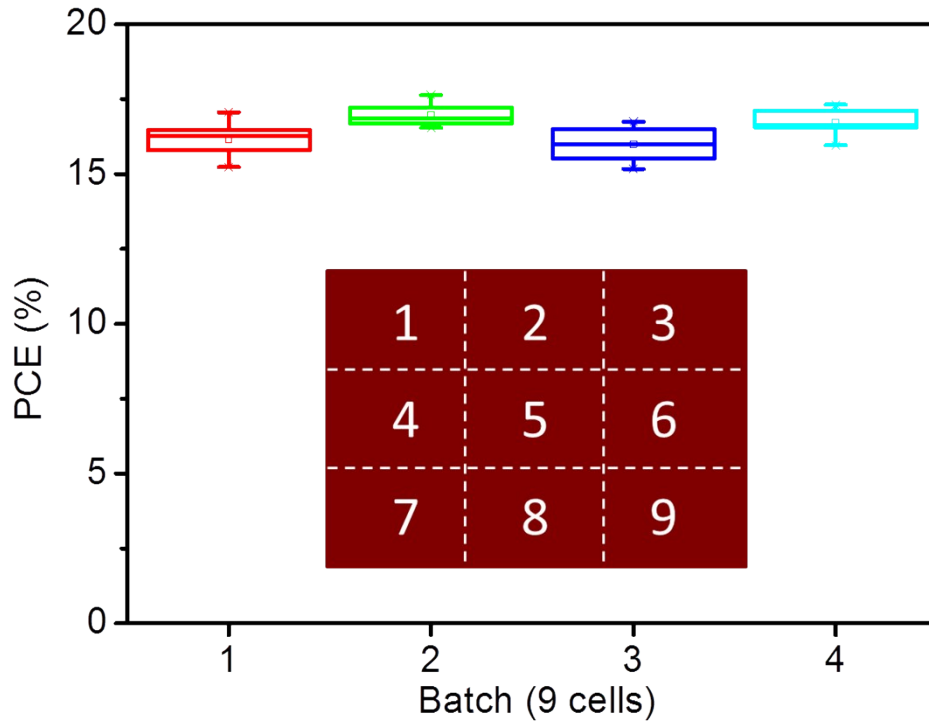


Fig. S7 The statistical analysis of PCEs of 4 batch of devices (9 cells per batch). PSCs from each batch were fabricated on the 9 sub-areas divided along the white dash lines on SCD perovskite films (6 cm×8.5 cm).

Table 1. The viscosities of the common used solvents at room temperature and atmospheric pressure. DMSO and GBL have the highest viscosity among all the listed solvents.

Solvent	Viscosity(cp)
DMSO	1.996
GBL	1.7
DMCA	0.92
DMF	0.802