Support information

Enhanced morphology and stability of high-performance Perovskite Solar Cells with ultra-smooth surface and high fill factor via crystal growth engineering

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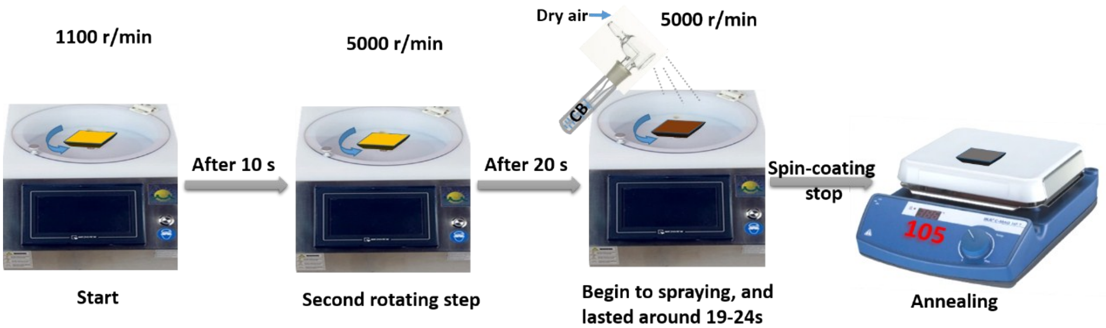


Fig. S1 the process of spraying anti-solvent process and time control.

Table S1 Operating parameters of anti-solvent spray process

No.	Carrier gas flow(L/h)	Rotating speed(r/min)	Spraying time(s)	Result *
1	30	3000	10	uncovered
2	30	4000	20	uncovered
3	60	4000	10	rough
4	60	4500	20	rough
5	60	6000	30	uncovered

6	90	4000	15	Little smooth
7	90	4500	20	smooth
8	90	5000	30	rough
9	120	4500	20	uncovered

*Significantly rough surface or un-covered film can be observed clearly.

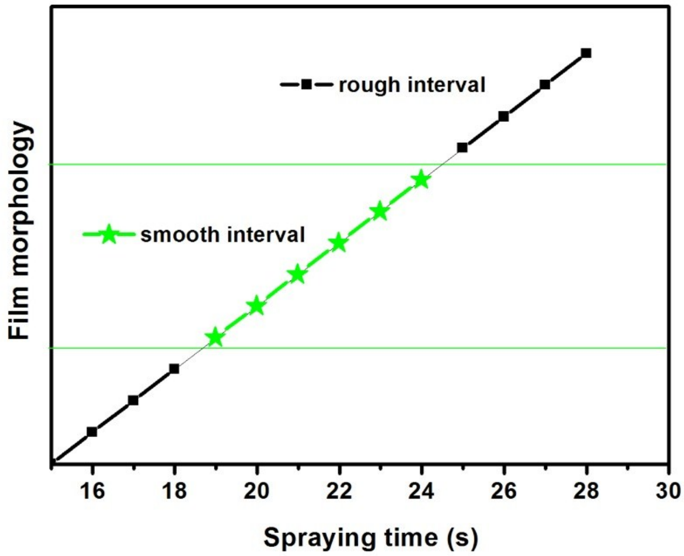


Fig. S2 The relationship between spraying time and film morphology at carrier gas flow of 90 L/h, and rotating speed of 4500 r/min.

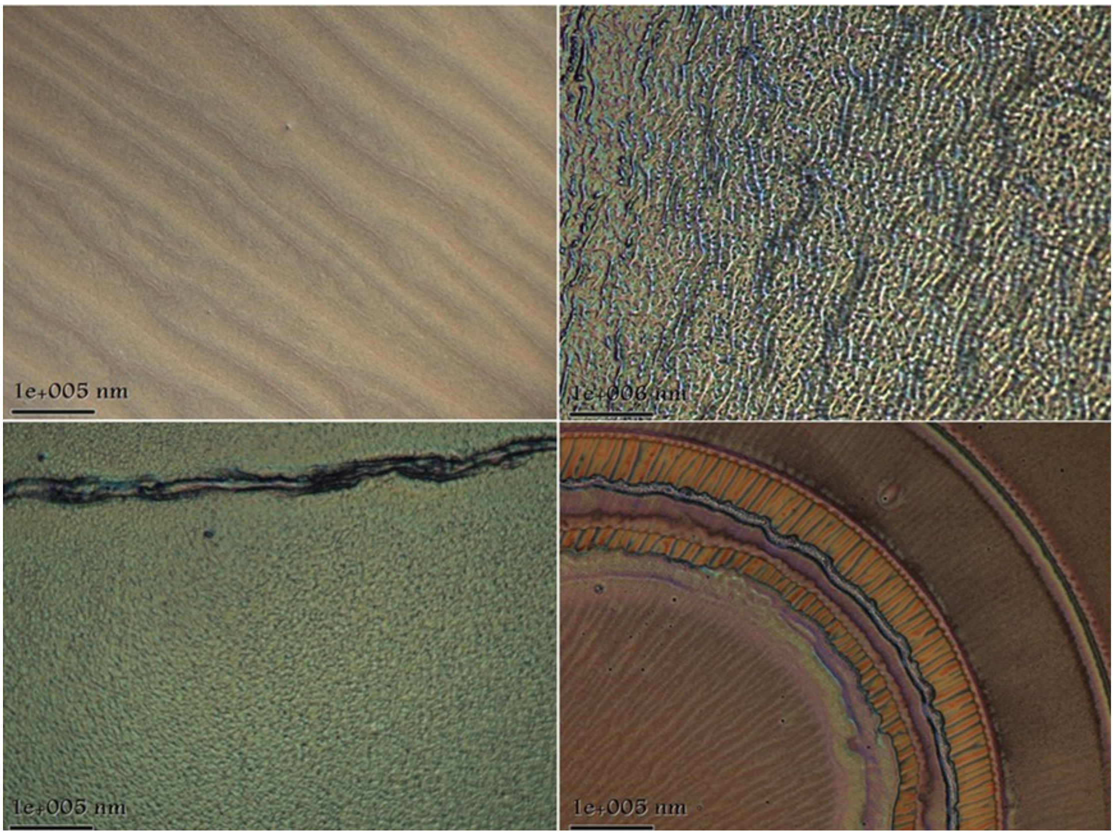


Fig. S3 Polarizing microscope images of wave like film, blur film, cracking film and film with circle around defects due to centrifugal force and anti-solvent drop impact force originated from inappropriate operating conditions. Despite a flat film can be usually got by skilled technical personnel, but sometimes defective films were also inevitable.

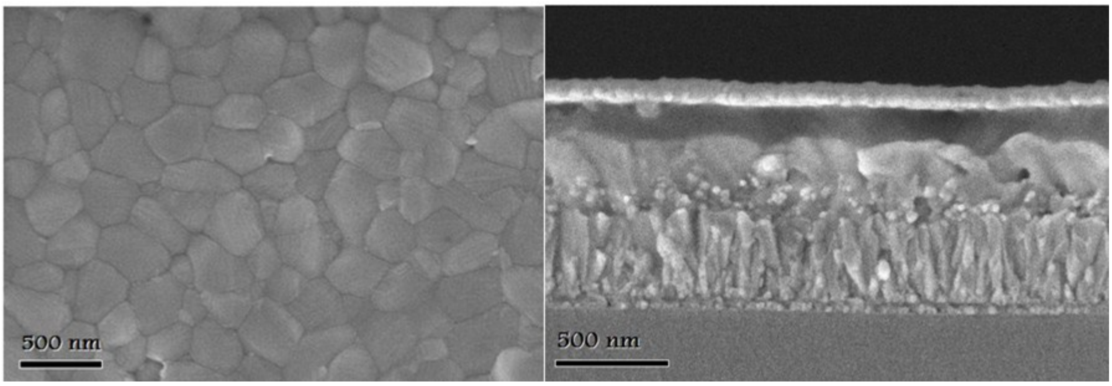


Fig. S4 surface and sectional morphology of perovskite film prepared by conventional dripping method.

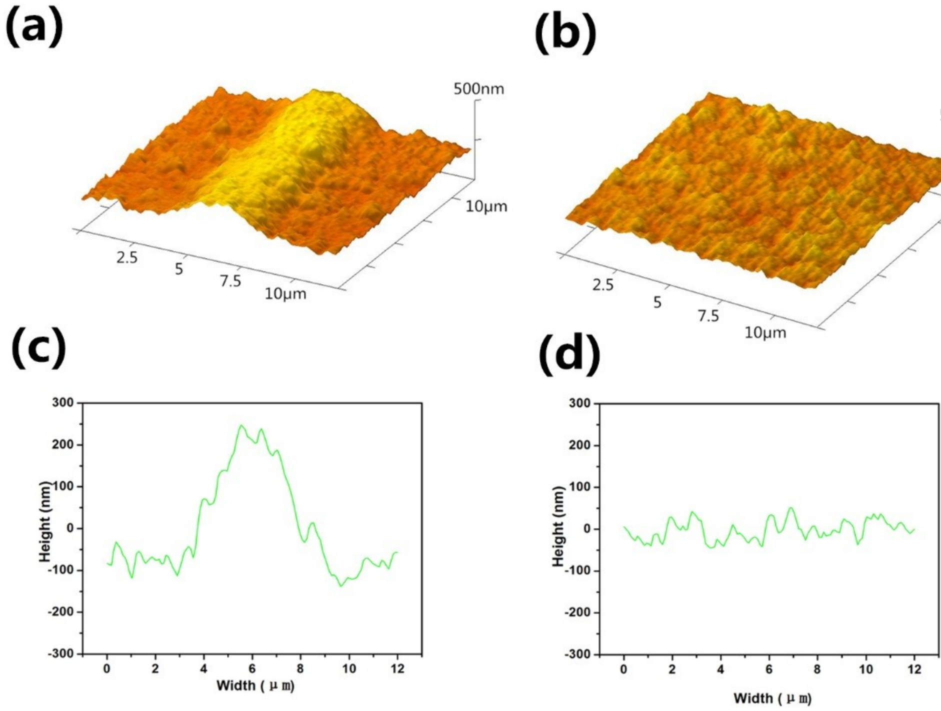
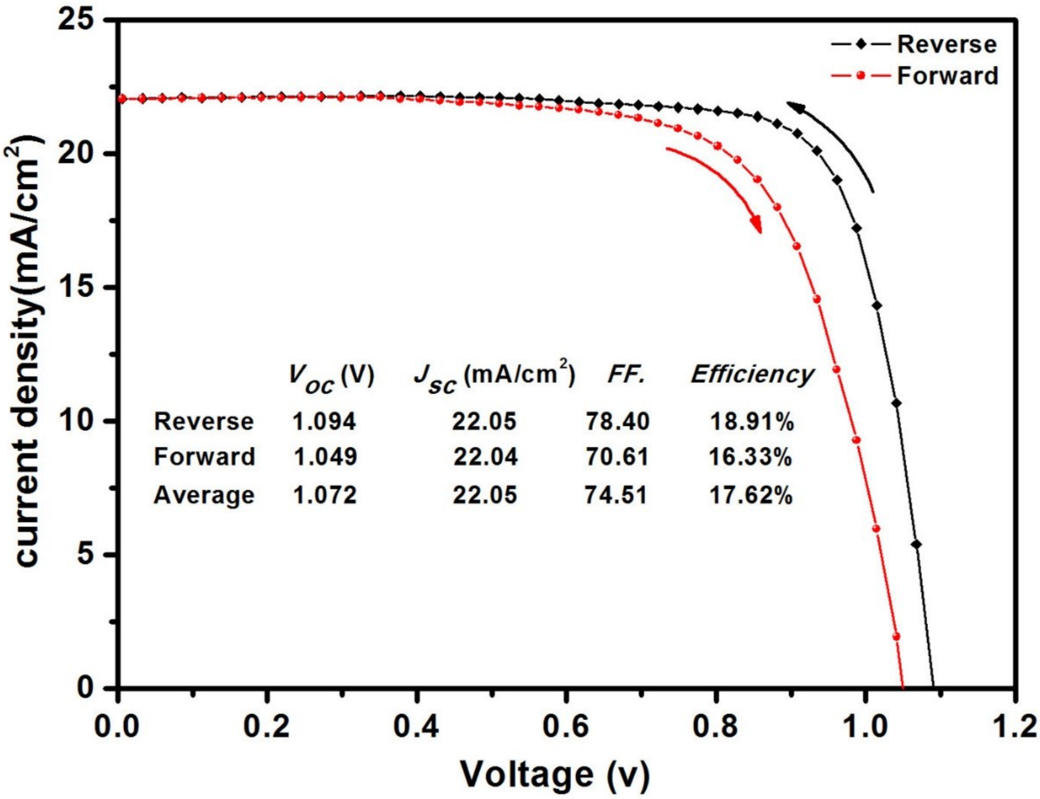


Fig.S5 (a-b) Three-dimensional graph of perovskite films with waves and smooth surface measured by atomic force microscopy (AFM). (c-d) the corresponding AFM height profile of wave films and smooth film.

(a)



Reverse
Forward
current density(mA/cm^2)
Voltage (v)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
0
5
10
15
20
25
V_{oc} (V) J_{sc} (mA/cm^2) FF. Efficiency
Reverse 1.094 22.05 78.40 18.91%
Forward 1.049 22.04 70.61 16.33%
Average 1.072 22.05 74.51 17.62%

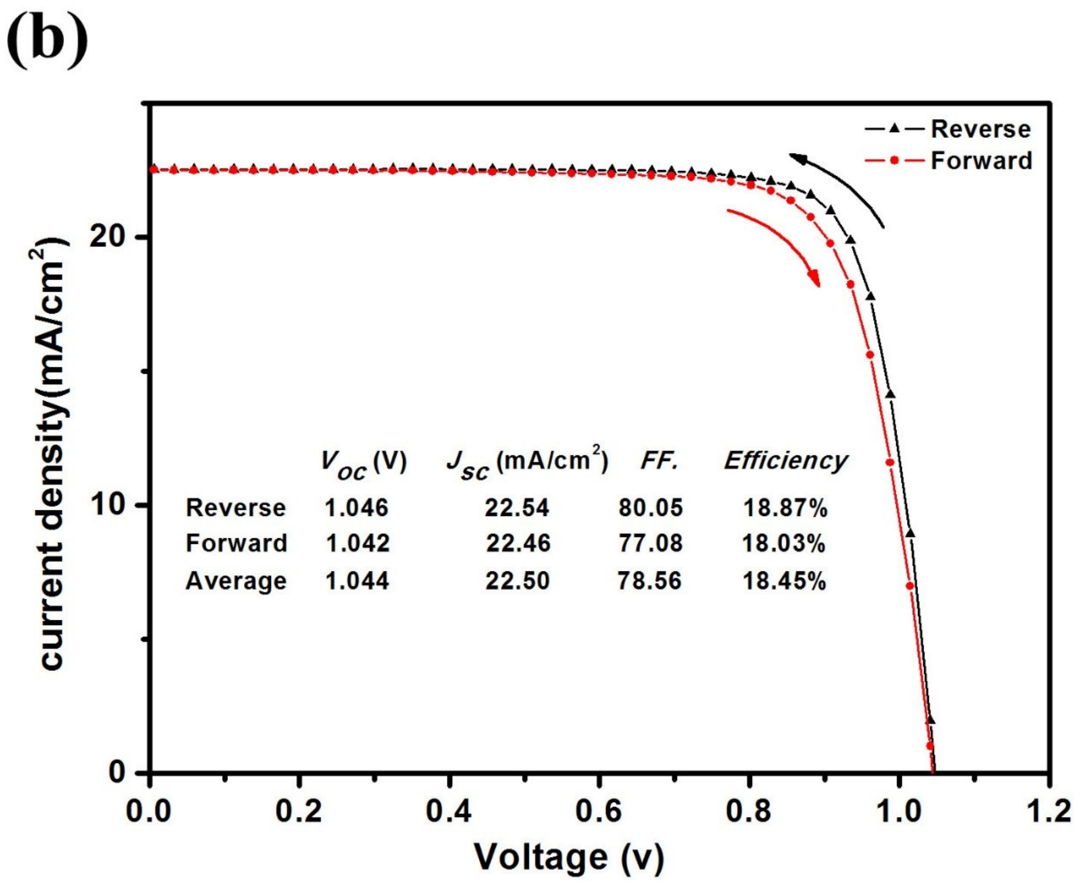


Fig. S6 (a), (b) J-V curves of forward and reverse bias sweep for the solar cell prepared by conventional and SAP method measured at 100 mW/cm^2 illumination AM 1.5G which is calibrated by certified Si-reference cell by NREL.

Table S2 TAS values of different type of film morphology

Type of film morphology	Decay time (τ)
Spraying anti-solvent process method (SAP)	**175.81 ns**
Conventional dripping anti-solvent method	**131.65 ns**
Film with ring defects prepared by dripping	**118.58 ns**