Supporting Information

Small Molecule Driven Directional Movement Enabling Pin-hole Free Perovskite Film via Fast Solution Engineering

Li-Li Gao, Ke-Jie Zhang, Lin Chen, Ni Chen, Cheng-Xin Li, Chang-Jiu Li and Guan-Jun Yang*

State Key Laboratory for Mechanical Behavior of Materials, School of Materials Science and Engineering, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, PR China

Monte Carlo simulation and parameters:

We set the current temperature as 20℃, and the pressure as 1.013×10^5 Pa.

1, Evaporated gas DMF molecule enters the boundary layer at the incident angle of 0-180° in the hemisphere, the incident distance is $\lambda \pm \delta\lambda$, ($\lambda$ is molecular free path, δ is the random coefficient), the incident velocity is $v_{DMF} \pm \delta v_{DMF}$ (v_{DMF} is mean velocity of DMF molecule, δ is the random coefficient).

2, GM moves towards DMF, GM is randomly located at spherical surface with radius $r = v_{GM} \times t_{DMF}$, ($v_{GM}$ is the velocity of GM, t_{DMF} is the time of DMF moving from liquid surface to the collision point), the incident angle is θ, shown in Fig. S1b, the incident velocity is $v_{GM} \pm \delta v_{GM}$ (v_{GM} is velocity of GM, δ is the random coefficient).

3, DMF collides with GS at the angle of θ, as shown in Fig. S1c, completing the first collision.

4, The DMF, which completes the first collision, as the beginning of the nest collision, which repeats the step from 1 to 3.

5, The collision continues until DMF crosses the boundary layer. If it does not cross the boundary layer, it returns back to the liquid surface. Now we record the collision times.

6, We set that there are 5×10^5 DMF, which evaporated from liquid surface. They all repeat the step from 1 to 6. We have the statistics of their number, which cross the boundary layer.

7, We count the number of molecules crossing boundary layer, and the collision times before their crossing boundary layer.

8, GM can be set as Nitrogen and Helium.

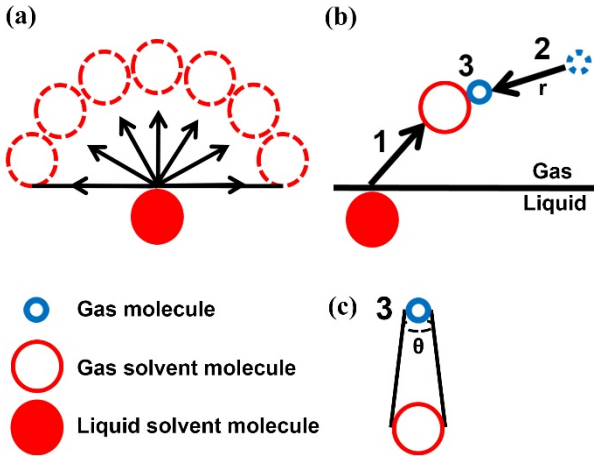

Fig. S1 Collision model schematic diagram, (a) solvent molecule evaporates from liquid surface, (b) collision route of SM and GM, (c) collision angle of SM and GM.

Parameters calculation:

1, Calculation of molecule velocity and molecular free path:

Molecule velocity, v, formula is as follows,

$$v = 1.6\sqrt{\frac{RT}{M}}$$

where R is ideal gas constant, T is the temperature, M is relative molecular mass.

Molecular free path, λ, formula is as follows,

$$\lambda = \frac{kT}{\sqrt{2}\pi d^2 P}$$

Where k is Boltzmann constant, d is molecular diameter, T and P are the temperature and pressure respectively.

Table S1 λ and v are calculated based on above formulas (T=20 °C, P=1.013 ×10^5 Pa).

Matter	Relative molecular mass	Molecular diameter (nm)	Molecular free path λ (nm)	Molecular velocity v (m/s)
Nitrogen (N_2)	28	0.36	70	472
Helium (He)	4	0.26	133	1248
DMF (C_3H_7NO)	73	0.52	33	292

2, Calculation of nitrogen and Helium boundary layer thickness:

Boundary layer thickness, δ, formulas is as follows,

$$\delta = \frac{L}{\sqrt{\mathrm{Re}}},$$

$$\mathrm{Re} = \frac{\rho v L}{\eta},$$

where Re is Reynolds number, L is characteristic length, ρ is gas density, v is gas velocity, η is gas viscosity, (T=20 ℃, P=1.013 ×10^5 Pa, L=0.025 m).

Table S2 δ is calculated based on above formulas. Gas velocity, v, is calculated based on the structure of MAK at Q=475 LPM for Nitrogen, 1250 LPM for Helium at standard state (both all 300 LPM at working condition).

Gas	Gas density ρ (Kg/m^3)	Gas viscosity η (×10^{-5} Pa▪s)	Gas velocity v (m/s)	Boundary layer thickness δ (μm)
Nitrogen (N_2)	1.165	1.81	62.09	70
Helium (He)	0.166	1.98	164.29	120

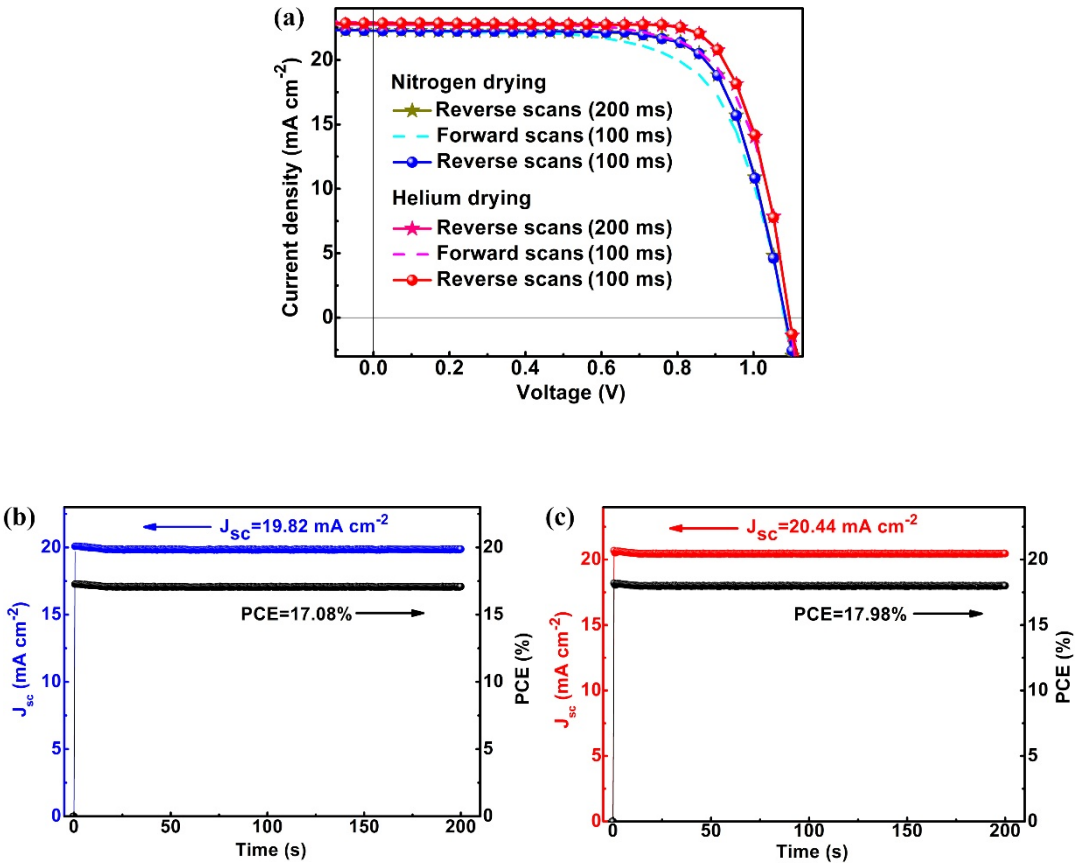

Fig. S2 (a) J–V curves of the devices with perovskite film drying by Nitrogen and Helium scanning at different direction and delay times; (b) and (c) are steady-state current density and power conversion efficiency of devices with perovskite film drying by Nitrogen and Helium, respectively.

Table S3 Solar cell performance parameters.

Devices	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF	PCE (%)
Helium	1.10±0.01	20.99±1.60	0.73±0.02	16.78±1.34
Nitrogen	1.08±0.01	19.97±1.80	0.71±0.02	15.41±1.48

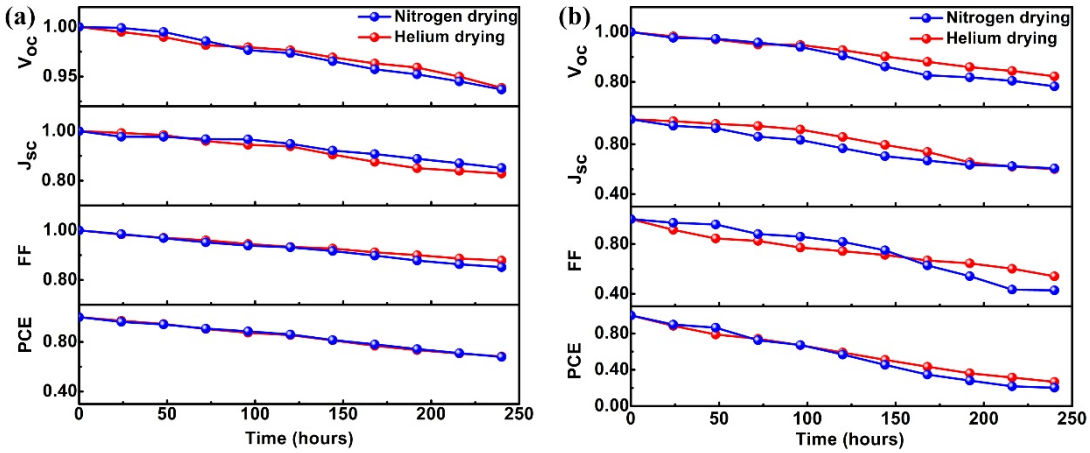

Fig. S3 The stability of corresponding perovskite solar cells without any encapsulation in ambient environment of 20 °C with different relative humidity, 50% (a) and 70% (b), respectively.

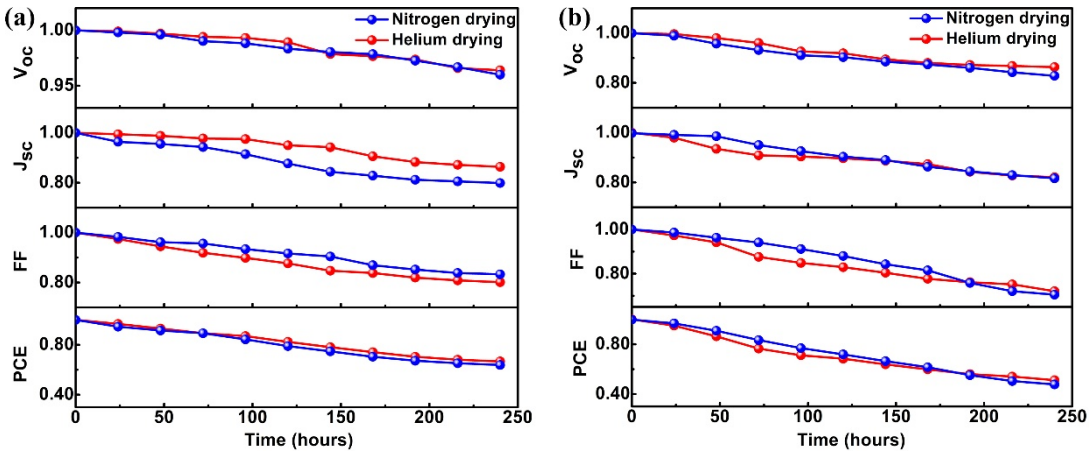

Fig. S4 The stability of corresponding perovskite solar cells without any encapsulation in ambient environment of 50% humidity with different temperature, 50 °C (a) and 80 °C (b), respectively.