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Supporting Information

Cost-effective green antisolvent towards reproducible fabrication of high-efficiency perovskite solar cells

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Experimental Section

Materials:

The SnO_2 colloid precursor was purchased from Alfa Aesar. N, N-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), Tert-butanol (TBA), and chlorobenzene (CB) were purchased from Sigma-Aldrich. PbI_2, FAI, MACl, CsI, bis(trifluoromethane) sulfonimide lithium salt (Li-TFSI), 4-tert-butylpyridine (TBP) and Spiro-OMeTAD were purchased from Xi'an Yuri Solar Co., Ltd.

Solar cells Fabrication:

The ITO glass substrate (1.5×1.5 cm) was cleaned with deionized water, acetone and isopropanol respectively for 15 minutes. Then the clean substrate was dried by a nitrogen stream, and treated with oxygen plasma for 5 minutes. The SnO_2 colloidal precursor was diluted with ultrapure water (volume ratio 9:40) to prepare the SnO_2 precursor. The SnO_2 precursor was spin-coated on the ITO substrate at a speed of 3500 rpm for 30 s, and then

annealed at 150°C for 30 minutes in the air (relative humidity RH=40%). Then the substrate was transferred to a nitrogen glove box to prepare the perovskite film. The perovskite precursor solution was prepared by dissolving corresponding amounts of CsI (30 mg), FAI (377.3 mg), PbI_2 (1102.4 mg), and MACl (51.8 mg) in DMF: DMSO (8:2, 5:5, 2:8 and 0:10 v/v) mixed solvent (1567 µL). The perovskite film was prepared by one-step solution deposition method. 60 µL perovskite precursor solution was deposited onto ITO/SnO_2 substrates and a two-step spin coating procedure with 1000 rpm for 10 s and 5000 rpm for 30 s was adopted. During the second spin-coating step, antisolvent (TBA or CB) was dripped on the rotating substrate at 1^{th}, 5^{th}, 10^{th}, 15^{th}, 20^{th}, or 25^{th} s. After that, the sample was annealed at 120°C for 60 min in a box (relative humidity RH=20-30%). The Spiro-OMeTAD solution was prepared by dissolving 72.3 mg Spiro-OMeTAD with 28.8 µL TBP and 35 µL LiTFSI (260 mg mL^{-1} in ACN) in 1 mL chlorobenzene. Spiro-OMeTAD solution was then spin-coated on the perovskite layer with 4000 rpm for 30 s. Finally, about 120 nm Au was deposited on the spiro-OMeTAD layer.

Characterization:

FTIR spectra were measured by using VERTEX 70v (BRUKER, Germany). The wavenumber range was 400-4000 cm^{-1}, the resolution was 4 cm^{-1}, the scanning mode was a reflection mode, and the scan number was 64. XPS was measured by XPS machine (ESCALAB250XI, Thermo Fisher Scientific). The X-ray diffraction patterns were measured by using a Rigaku-2500 X-ray diffractometer with an X-ray tube (Cu Kα, λ =1.5406 Å). The UV light source is He I, and the energy of He I is 21.22 eV. The basic vacuum of the analytical chamber is $3.0x10^{-8}$ Torr, and the bias voltage is -9 V. Scanning electron microscopy (SEM) and energy dispersive spectrometer (EDS) images were observed by using a JEOL JSM-8020 field-emission scanning electron microscope (JEOL, Japan) at 5-10 kV. EQE was recorded using a Newport Oriel QE-200 (Newport 300 W xenon lamp). All J-V curves were measured using a source meter (Keithley 2420, USA) under AM 1.5 sunlight at an irradiance of 100 mW cm^{-2} provided by a solar simulator (Newport, Oriel Sol3A Class AAA, 94043A). The device area was 0.04 cm^2. Light intensity was calibrated using a monocrystalline silicon

reference cell with a KG5 window (Newport, Oriel 91150). Electrochemical impedance spectroscopy (EIS) was measured by Zennium (Zahner).

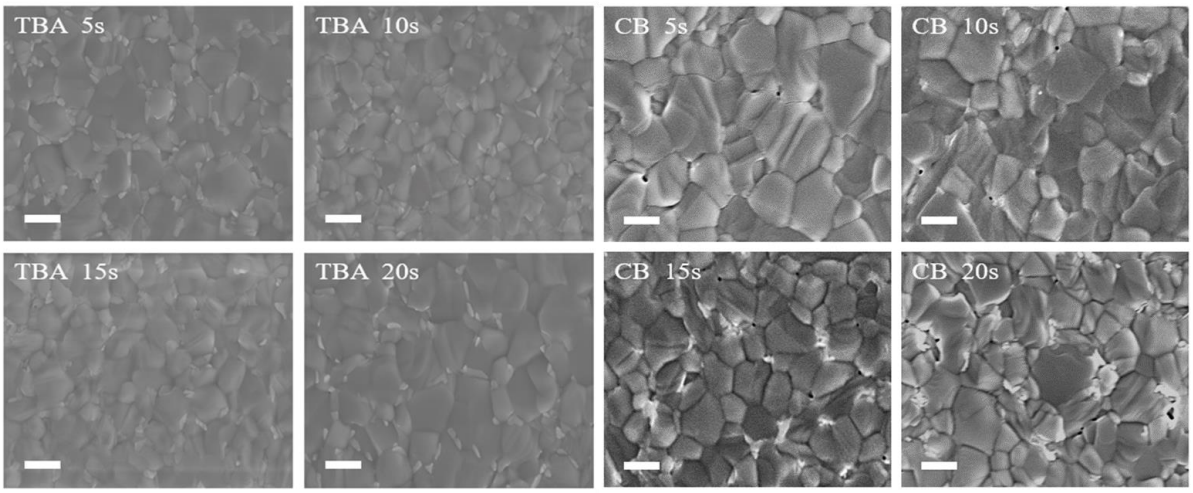


Figure S1 SEM images of perovskite films prepared with different antisolvent dripping time (DMF:DMSO ratio is 8:1, dripping volume is 300 μL), all scale bars are 1.5 μm.

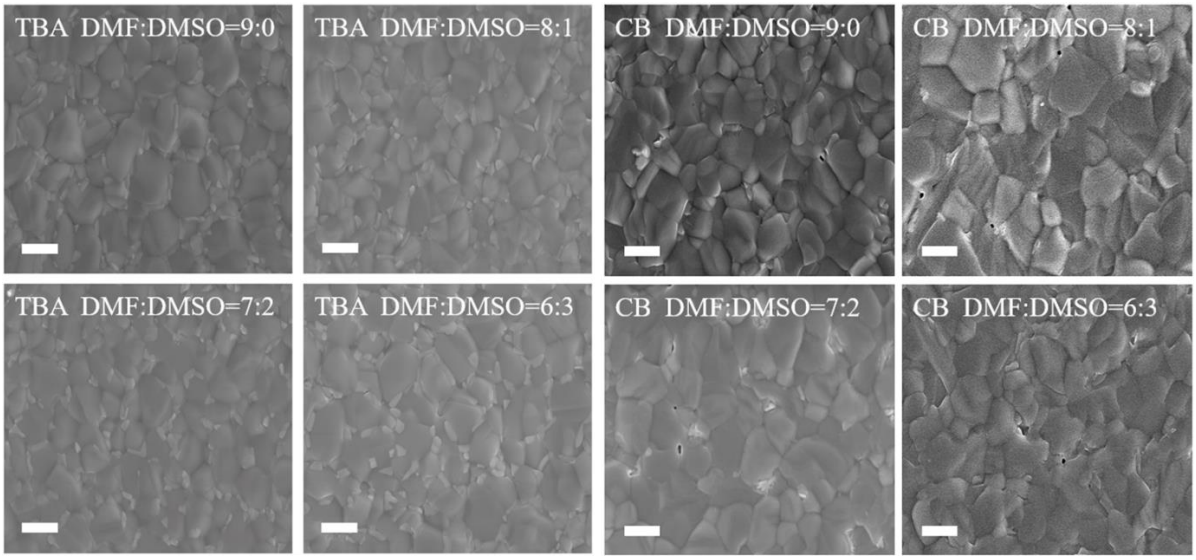


Figure S2 SEM images of perovskite films prepared with different DMF:DMSO ratio (dripping time is 10th s, dripping volume is 300 μL), all scale bars are 1.5 μm.

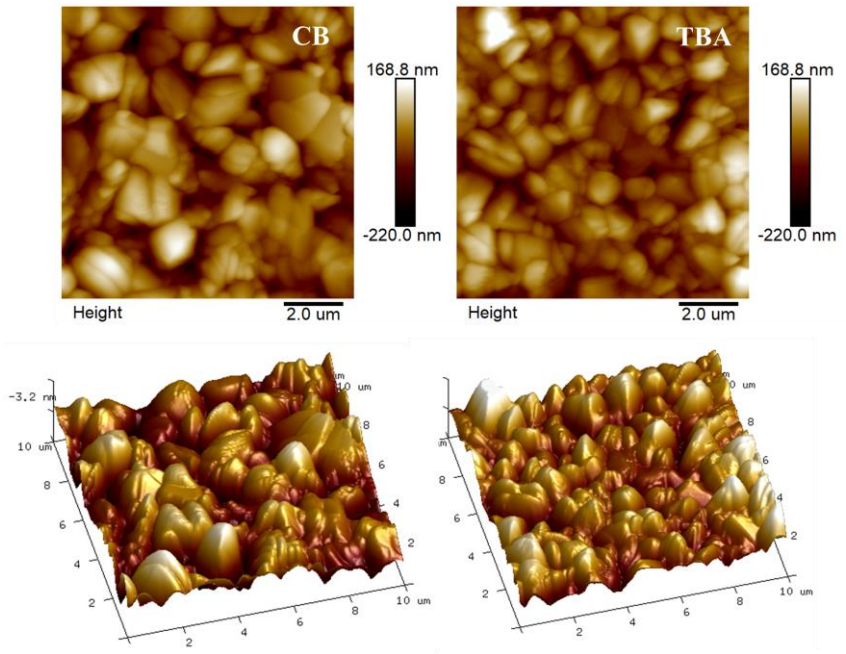


Figure S3 AFM images of perovskite films treated with CB or TBA antisolvent.

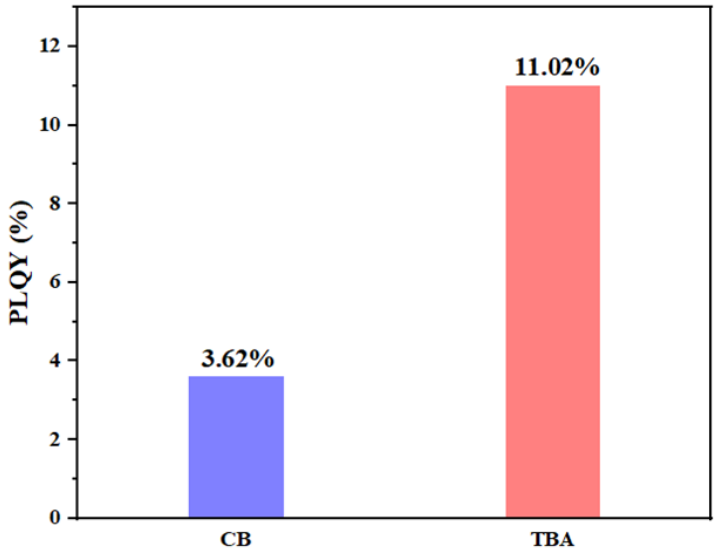


Figure S4 Absolute photoluminescence quantum yield (PLQY) measurements.

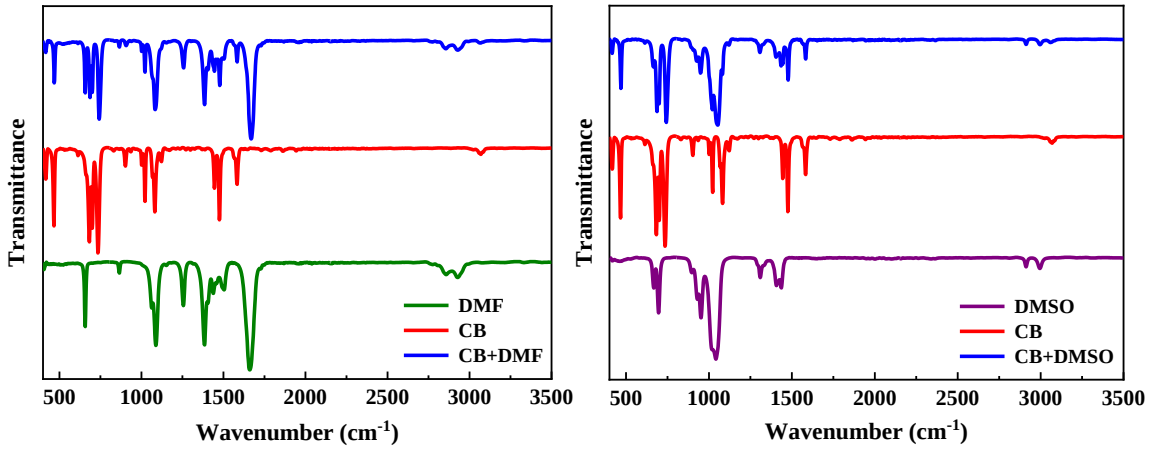


Figure S5 FTIR spectroscopy of solvent.

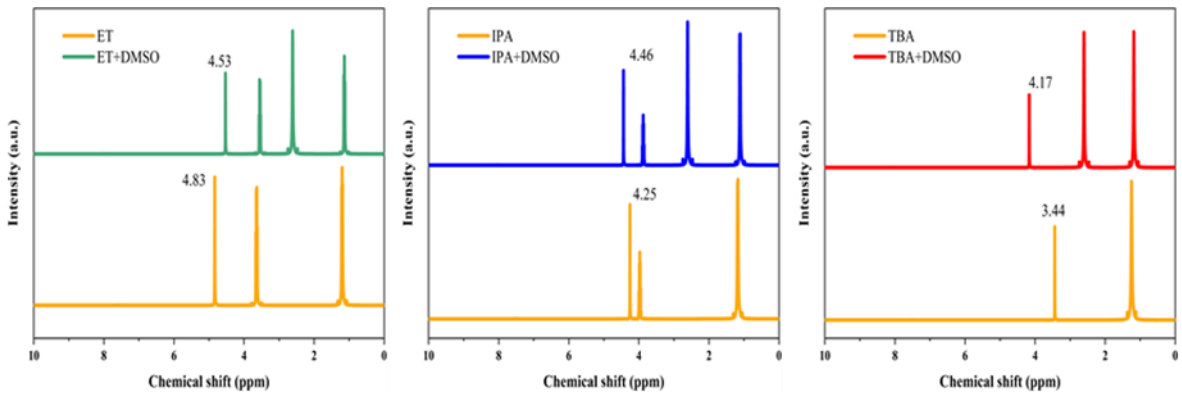
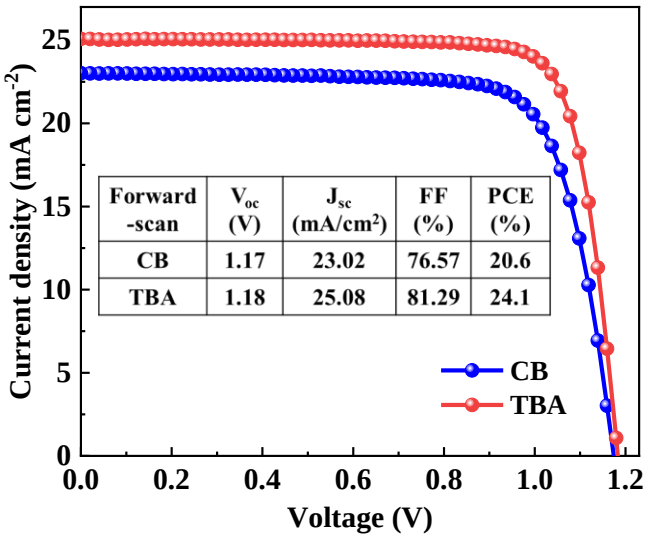


Figure S6 ^{1}H NMR spectra of solvent.



Forward -scan	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
CB	1.17	23.02	76.57	20.6
TBA	1.18	25.08	81.29	24.1

Figure S7 J-V curves of devices (Forward-scan).

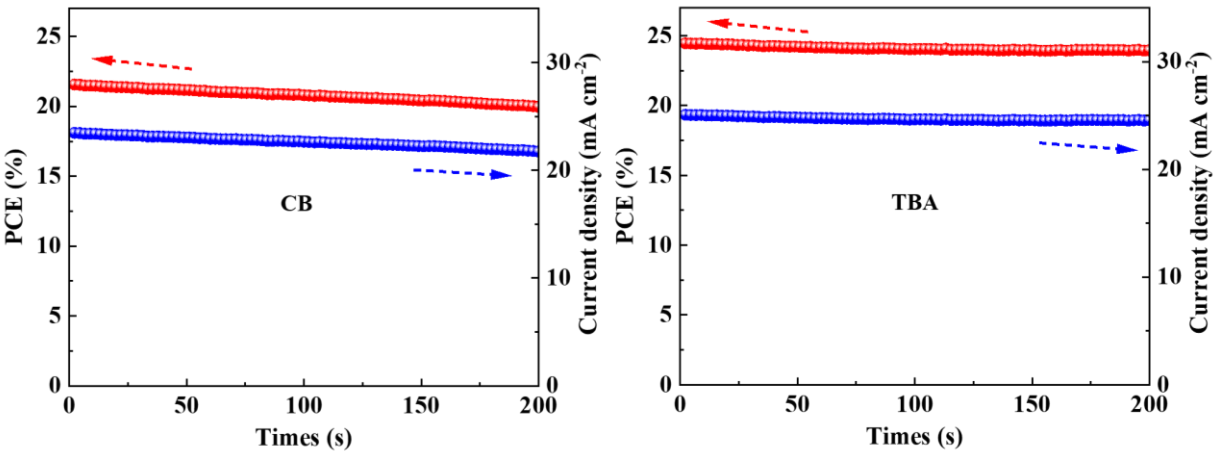


Figure S8 Steady-state output of devices at MPP under 1-sun illumination.

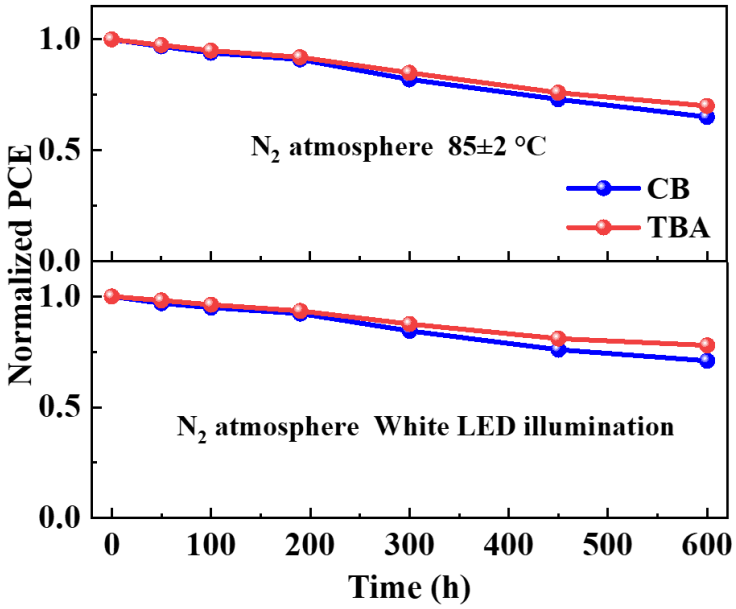


Figure S9 Thermal stability at 85 °C and light stability in simulated one sun illumination achieved by a white-light LED array.

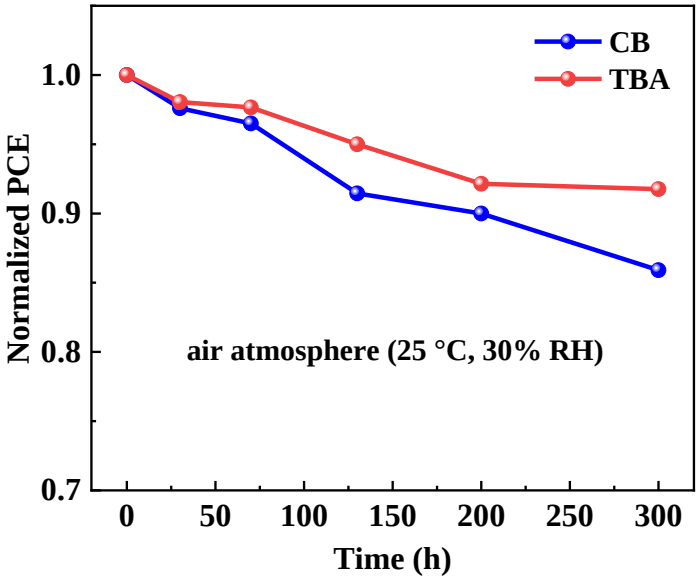


Figure S10 Long-term stability of devices in ambient air at 25 °C and 30% relative humidity without encapsulation.

The average recombination lifetime was estimated from the TRPL spectra fitted by a bi-exponential decay function, the equation is as follows:[1]

$$\tau_{ave} = \frac{\sum A_i \tau_i^2}{\sum A_i \tau_i} \quad \text{S1}$$

where τ_i is decay times and A_i is amplitudes.

Table S1 Parameters of the TRPL spectra of perovskite films prepared with CB or TBA antisolvent.

Sample	τ_1 [ns]	A_1	τ_2 [ns]	A_2	τ_{ave} [ns]
CB	223.38	16.15	1852.27	173.99	1834.24
TBA	342.55	31.63	3382.88	304.64	3351.25

The trap density (N_t) can be determined from the curves via the following relation:[2]

$$N_t = \frac{2\varepsilon_0 \varepsilon_r V_{TFL}}{eL^2} \quad \text{S2}$$

where ε_0 is the vacuum permittivity, ε_r is the relative dielectric constant of the perovskite, V_{TFL} is the trap-filled limit voltage, e is the electron charge, and L is the perovskite film thickness.

Table S2 Calculated trap density (N_t) of perovskite films prepared with CB or TBA antisolvent.

Sample	L [nm]	ε	V_{TFL} [V]	N_t [$\times 10^{15}$cm^{-3}]
CB	620	42	0.30	3.6
TBA	620	42	0.24	2.9

Table S3 EIS parameters of PSCs prepared with CB or TBA antisolvent.

Sample	R_s [Ω]	R_{tr} [kΩ]	C_{tr} [F]	R_{rec} [kΩ]	C_{rec} [F]
CB	1.48	0.777	1.083E-7	12.69	5.576E-7
TBA	0.92	0.443	9.709E-8	15.64	2.234E-7

References

1 J. Zhuang, P. Mao, Y. Luan, X. Yi, Z. Tu, Y. Zhang, Y. Yi, Y. Wei, N. Chen, T. Lin, F. Wang, C. Li, and J. Wang, *ACS Energy Lett.*, 2019, **4,** 2913.

2 L. Zhang, H. Li, J. Zhuang, Y. Luan, S. Wu, G. Niu, L. Chu, X. Cao, X. Li, and J. Wang, *J. Mater. Chem. C*, 2021, **9,** 15428-15434.