Supplementary Information (SI) for Journal of Materials Chemistry C.

Efficient printable mesoscopic perovskite solar cells without post thermal-annealing processes by introduction of a volatile co-solvent

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Experimental Section

Materials

N-Cyclohexyl-2-pyrrolidone (CHP, 99%), 2-methoxyethanol (2-ME, 99%) and N-methyl-2-pyrrolidone (NMP, 99%) were purchased from Alfa. Ultrapure lead(II) iodide (PbI2, >99.999%), ITO and FTO glass, and titanium diisopropoxide bis(acetylacetonate) (75 wt % in isopropanol) were purchased from Advanced Election Technology Co., Ltd. Methylammonium iodide (MAI, ≥99.5%) and lead(II) iodide (PbI_2, >99.99%) were purchased from Xi'an Polymer Light Technology Corp. Tetrahydrofuran, (THF, 99.9%, extra dry with molecular sieves, water≤30 ppm, in resealable bottle). TiO_2, ZrO_2, and carbon paste were provided by Wonder Solar Corp.

Perovskite Solution Preparation

The perovskite solution was made up of 0.1145 g of MAI, 0.3318 g of PbI_2 in 1 mL of 2-ME/NMP (600 μL/40 μL), and the mixed solution was stirred for 4 h. And extract 300 μL pristine precursor solutions and add different vol% THF and stirred for 1 h. CHP treated precursor solution is added with the addition of 20 vol% THF in pristine precursor solution.

Device Fabrication

The structure of the device used in this work is as follows: compact TiO_2 (c-TiO_2) mesoporousTiO_2 (mp-TiO_2)/mesoporous ZrO_2 (mp-ZrO_2)/mesoporous carbon (mp-C).

Laser-etched FTO conductive glass was used as a separate electrode and ultrasonically cleaned with a detergent, deionized water, and ethanol. A 30 nm-thick layer of compact-TiO_2

(c-TiO_2) was deposited on the prepared FTO by spraying titanium diisopropoxide bis(acetylacetonate) at 450 °C for 15 min. Later, three layers of mesoporous layers, comprising a mesoporous-TiO2 layer (m-TiO_2) of approximately 500 nm, a mesoporous-ZrO_2 (m-ZrO_2) spacer layer of approximately 2 µm, and a carbon layer of approximately 10 µm, were deposited layer by layer on cooled c-TiO_2 by screen printing and sintered for 35 min at 500, 450, and 400 °C. A 4.5 µL perovskite solution was infiltrated by one-step drop-coating on the top of the carbon layer at room temperature. After waiting for the solvent to fully diffuse and penetrate the mesopores, the device was transferred to a dark environment and left for 24 h without an annealing process. All of the above-mentioned processes are conducted in the air.

Characterization

Cross-sectional and top-view images of the perovskite films were offered by scanning electron microscopy (FEI, Quanta FEG 450, USA). The crystal characteristic image and visible light absorption image of perovskite films were provided by an X-ray diffractometer (Bruker, D8 advance, Germany) and ultraviolet−visible spectrophotometer (PerkinElmer, Lambda 365, USA), respectively, In situ microscopy was used to observe the crystallization process of perovskites in situ (The MOTIC, E250223, China). The surface potential of the films was measured by KPFM with tapping mode (Bruker, Multimode 8, Germany). The current density−voltage (J−V) curves were obtained using a Keithley 2400 digital source meter.

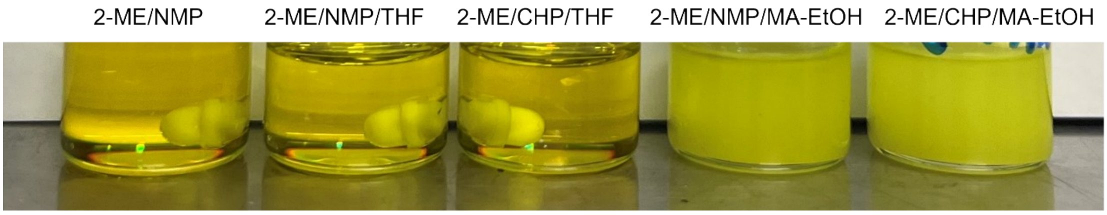

Figure S1 A photo of the precursor solution containing THF and MA-EtOH in 2-ME/NMP.

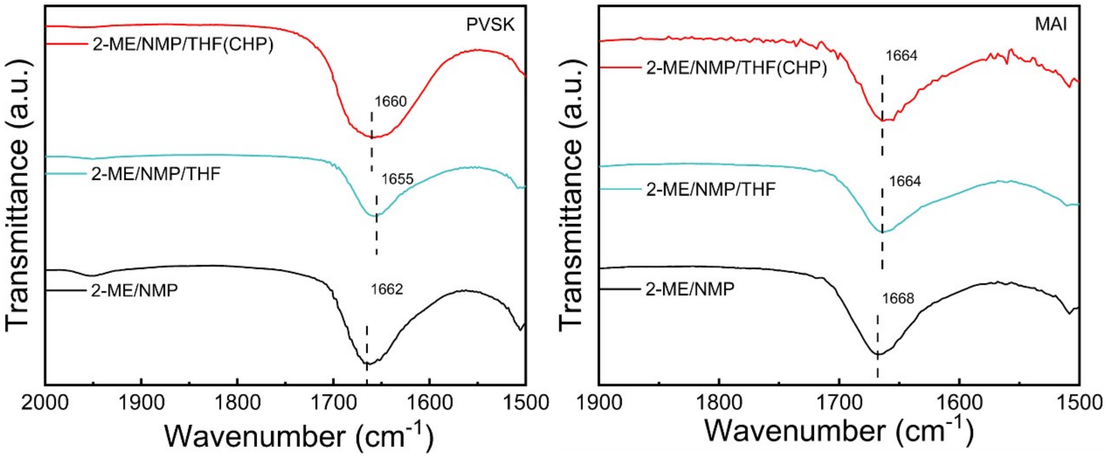

Figure S2 FTIR spectra of perovskite precursor and MAI added in different solvent compositions.

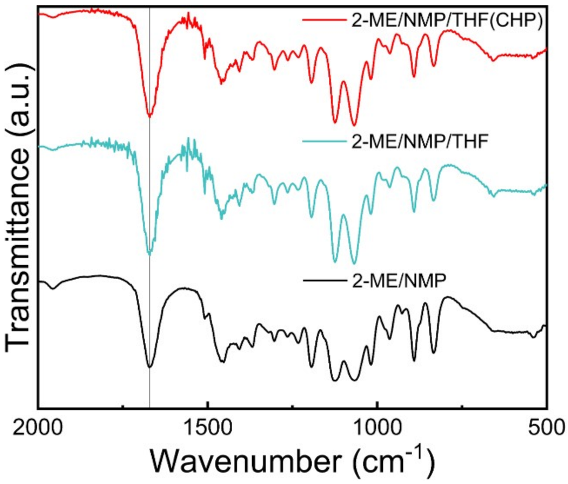

Figure S3 FTIR spectra of different solvent compositions.

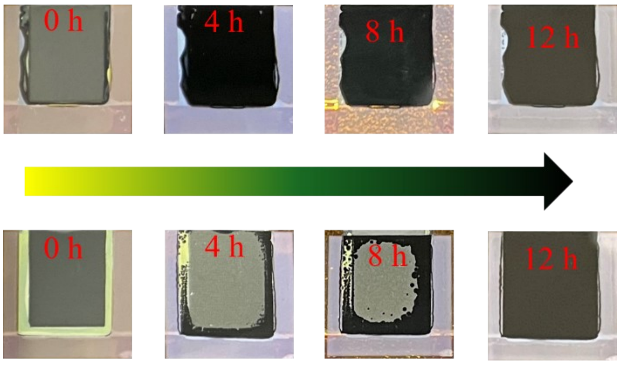

Figure S4 the photographs of devices treated by different solvent.

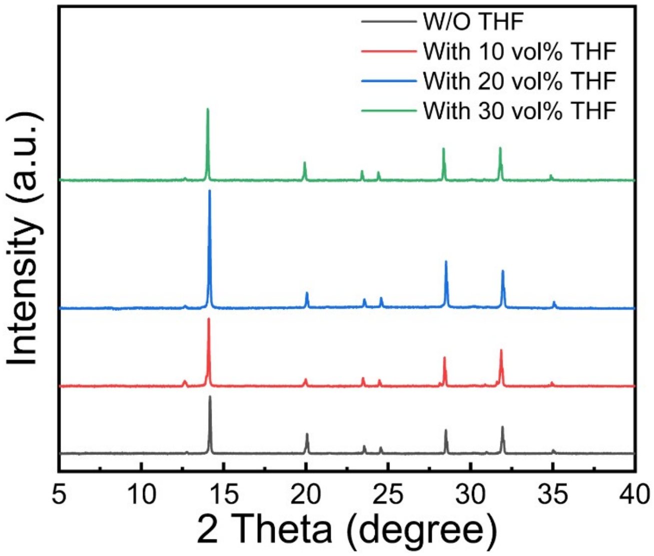

Figure S5 XRD patterns with the addition of THF at different volume fractions in 2-ME/NMP.

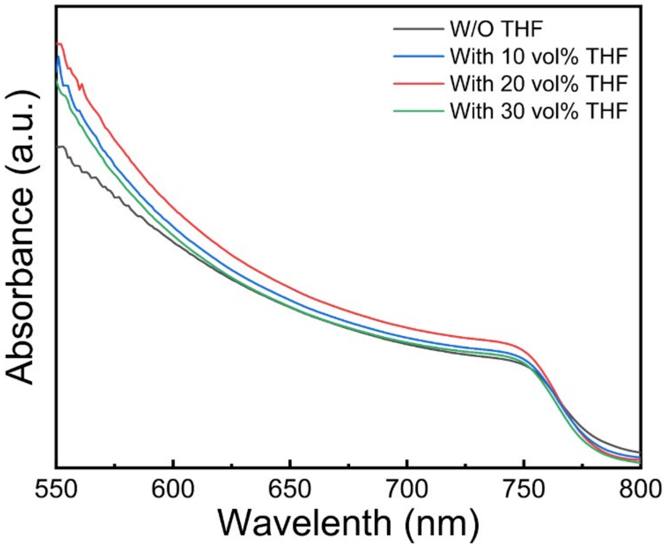

Figure S6 UV-Vis spectra of solutions with different volume fractions of THF added to 2-ME/NMP.

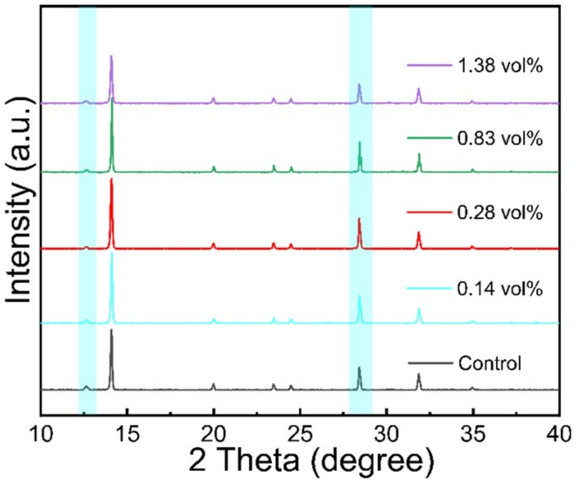

Figure S7 XRD patterns of samples with different volume fractions of CHP added to 2-ME/NMP/THF.

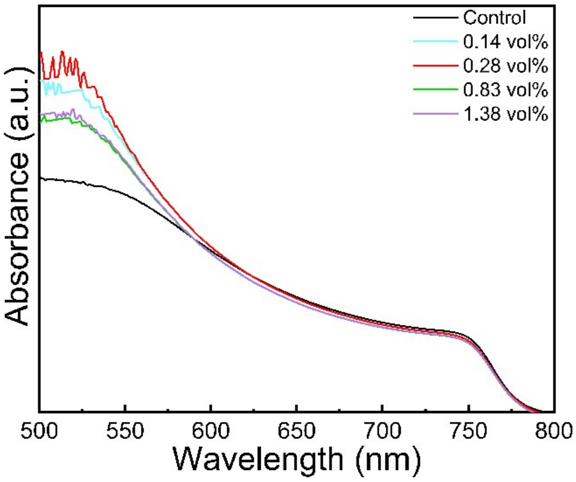

Figure S8 UV-Vis spectra of samples with different volume fractions of CHP added to 2-ME/NMP/THF.

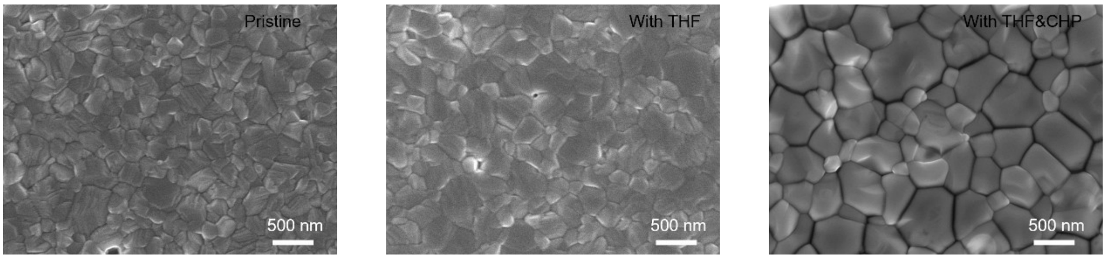

Figure S9 SEM images of Perovskite films spin-coated from different precursor solvent compositions.

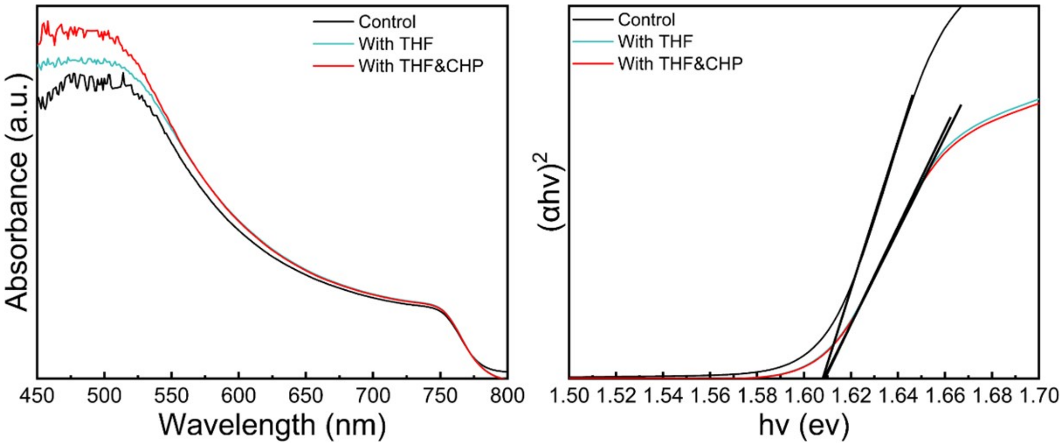

Figure S10 UV-Vis spectra and Tauc plots of thin films prepared from different precursor solution compositions.

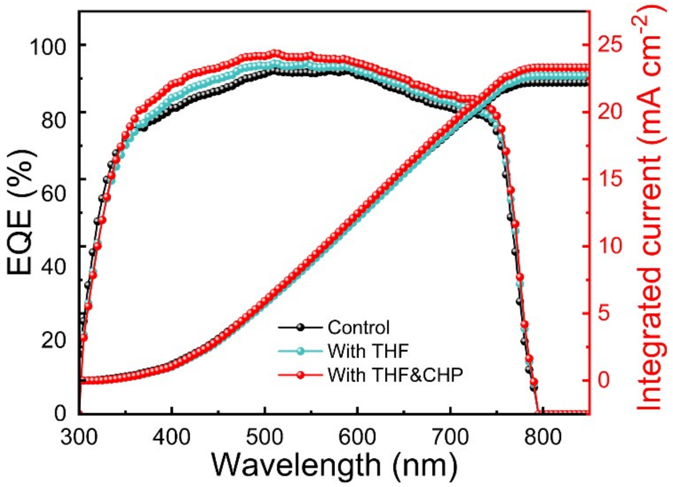

Figure S11 EQE curves of PSCs fabricated from different precursor solvent compositions.

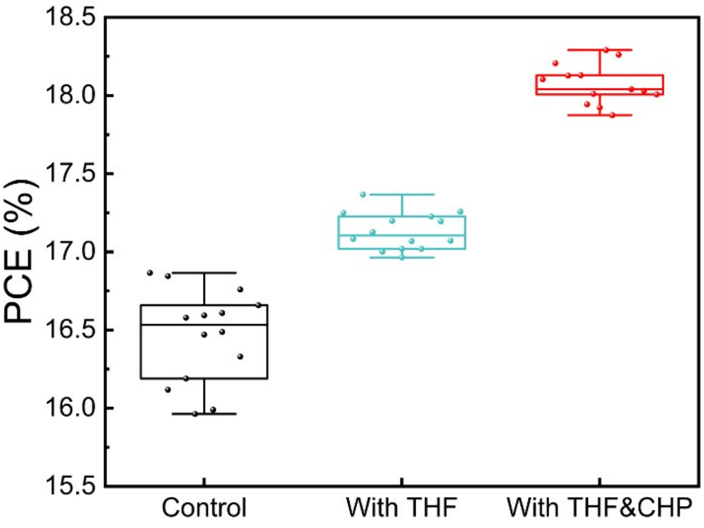

Figure S12 The statistical distribution shows the device efficiency of 10 cells.

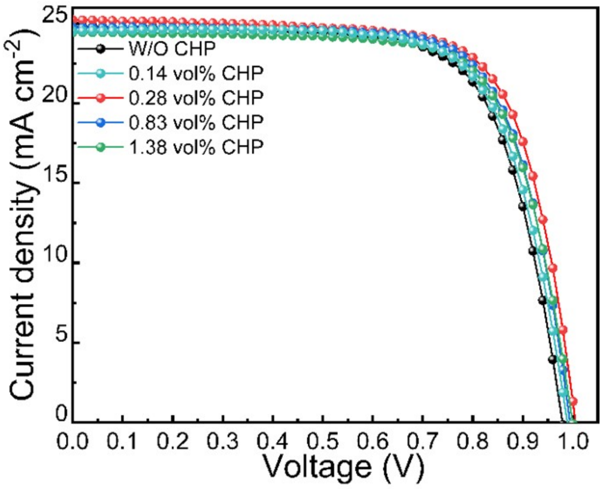

Figure S13 J-V curves of samples with different volume fractions of CHP added to 2-ME/NMP/THF.

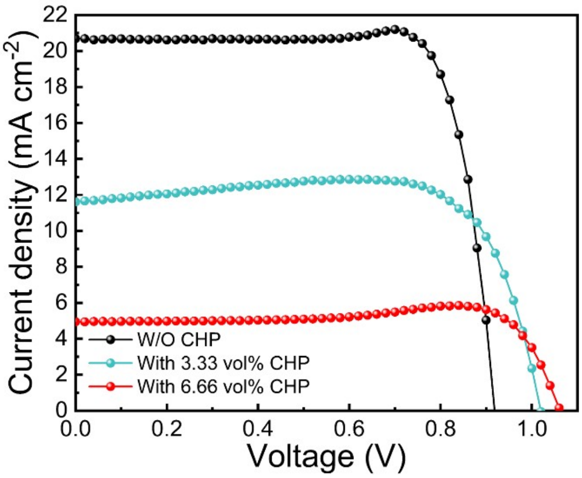

Figure S14 J-V curves with CHP replacing NMP at different volume fractions in 2-ME/NMP/THF.

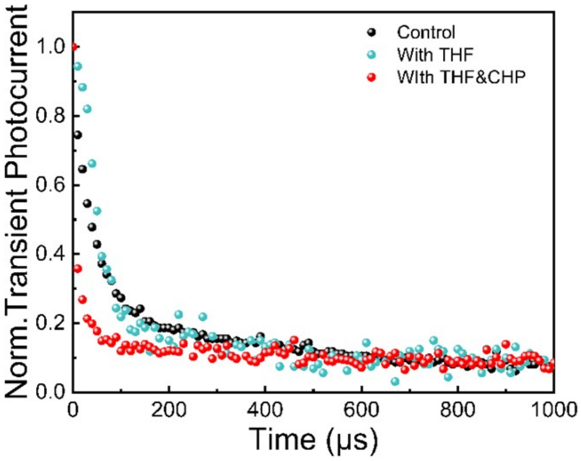

Figure S15 Transient photocurrent decay curves of PSCs fabricated from different precursor solvent compositions.

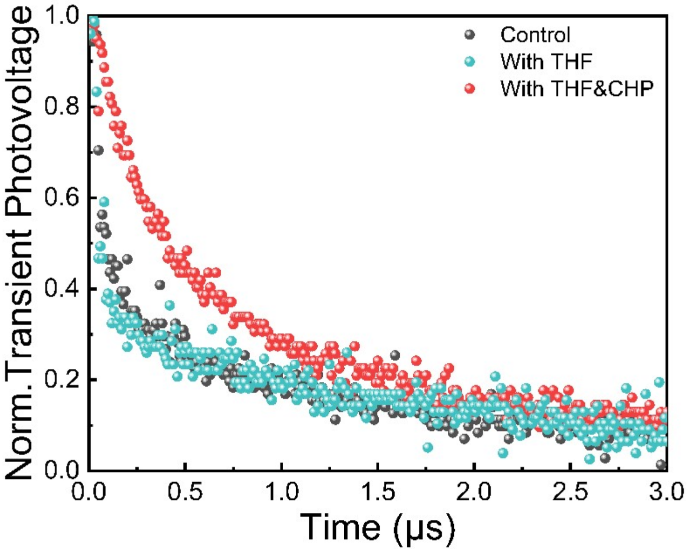

Figure S16 Transient photovoltage decay curves of PSCs fabricated from different precursor solvent compositions.

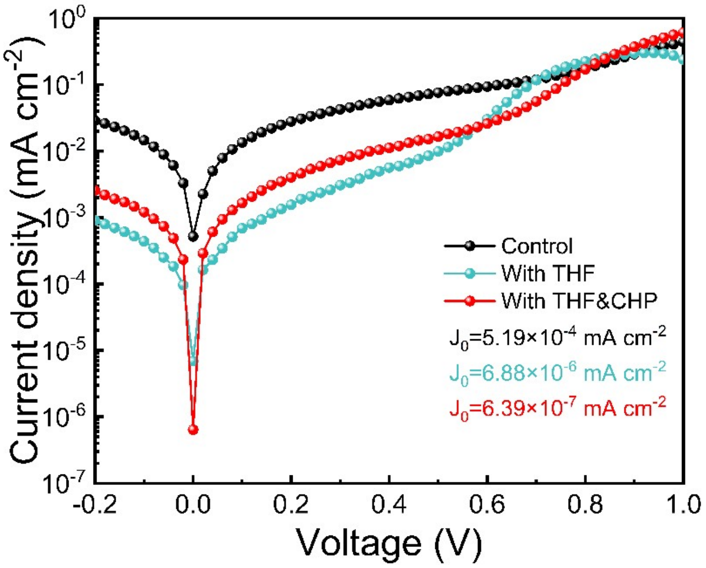

Figure S17 Dark J–V curves Dark J–V curves of PSCs fabricated from different precursor solvent compositions.

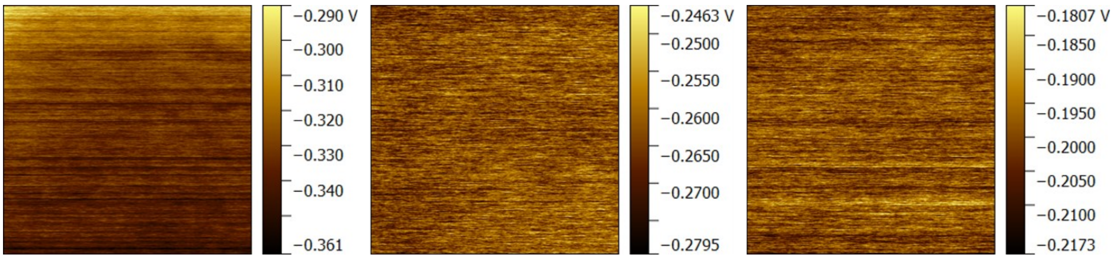

Figure S18 KPFM images of Perovskite films spin-coated from different precursor solvent compositions.

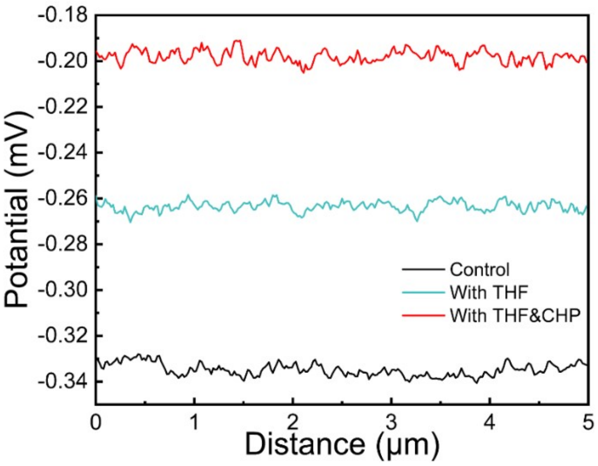

Figure S19 Potential cross-sectional profiles from kpfm image.

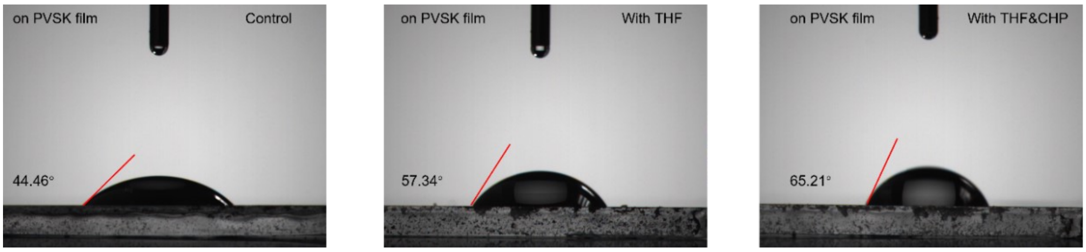

Figure 20 Water contact angle image of Perovskite films spin-coated from different precursor solvent compositions.

Table S1. Parameters of TRPL curves fitted using a bi-exponential decay method.

		J_{SC} (mA cm-2)	V_{OC} (V)	FF (%)	PCE (%)
Control	Max	24.09	0.976	71.68	16.86
	Avg.	24.36±0.61	0.950±0.032	71.45±0.31	16.50±0.43
With THF	Max	24.50	0.986	71.85	17.37
	Avg.	24.58±0.23	0.980±0.013	71.38±0.53	17.19±0.28
With THF&CHP	Max	25.21	1.005	72.171	18.29
	Avg.	25.40±0.31	1.000±0.010	71.39±0.22	18.07±0.23

Table S2. Parameters of TRPL curves fitted using a bi-exponential decay method.

	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)	τ_{avg} (ns)
Control	2.35	83.88	14.10	16.12	4.24
With THF	2.21	80.58	15.49	19.41	4.78
With THF&CHP	2.99	83.39	20.20	16.60	5.84