Supporting Information

## Antisolvent Effects in Green Solvent Engineering of FAbased Quasi-2D Ruddlesden-Popper Perovskite Films for Efficient Solar Cells

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Fig. S1 The optical image solution of FA-based quasi-2D perovskite in TEP.

Solvents	Boiling Point(°C)	Viscosity	Polarity	GHS symbol
N,N-Dimethylf- ormamide (DMF)	153	1.3	6.4	
Chlorobenzene (CB)	132	0.8	2.70	
Triethyl phosphate (TEP)	219	1.6	3.30	()
Dibutyl ether (DBE)	143	0.7	0.72	<u>!</u>
Petroleum ether(PE)	60~90	0.3	0.01	1.

**Table S1** Physical properties of solvents and antisolvents.



**Fig. S2** The photograph of films prepared from TEP without dipping antisolvent, (a) the unannealed film; (b) the annealed perovskite film.



Fig. S3 The UV absorption spectra of intermediate phase and quasi-2D perovskite film prepared from DBE and PE. (a) DBE; (b) PE.



**Fig. S4** The distribution histogram of grain sizes from FA-based quasi-2D perovskite films fabricated with (a) DBE (b) PE.



Fig. S5 The steady-state PL spectra from top (film) to bottom (glass) side of prepared films.



Fig. S6 The out-plane curves derived from GIWAXS patterns.



Fig. S7 The decay kinetics of different phases curves derived from TA spectral patterns.

Antisolvent	n-	$ au_{\text{\tiny et}}$ (ps)	$ au_1$ (ps)	$ au_2$ (ps)	τ <sub>3</sub> (ps)
S	value				
DBE	n=1	/	2.15	30.24	133.0
	n=∞	1.04	195.5	836.8	3141
PE	n=1	/	0.216	3.257	72.22
	n=∞	0.90	136.4	672.4	3630.0

**Table S2** The fitting results from transient absorption spectra.



Fig. S8 The cross-sectional SEM image of FA-based quasi-2D perovskite film fabricated with antisolvent of PE.



**Fig. S9** The *J*-*V* curves obtained from reverse and forward scan of the best PSCs prepared from different antisolvents. (a) DBE; (b) PE.



**Fig. S10** Water contact angle images of the FA-based quasi-2D perovskite films fabricated with antisolvent of DBE and PE.

**Table S3** The performance parameters of different n value FA-based quasi-2D perovskite solar cells prepared from traditional toxic solvent and green solvent system.

Solvents	Perovskites	n	V <sub>oc</sub> (V)	<i>J<sub>sc</sub></i> (mA cm⁻²)	FF(%)	PCE(%)	Ref.
DMF/CB	$BA_2FA_8Pb_9I_{28}$	9	1.102	18.89	63.22	13.16	1
DMF/CB	$BA_2FA_8Pb_9I_{28}$	9	1.098	21.09	68.17	15.01	2
DMF/CB	(FPEA <sub>2</sub> FA <sub>8</sub> Pb <sub>9</sub> I <sub>28</sub> )	9	1.07	20.88	72	16.15	3
DMF/CB	$EA_2FA_8Pb_9I_{28}$	9	1.09	21.89	73.05	17.4	4
DMF/IPA	(ThMA) <sub>2</sub> FA <sub>4</sub> Pb <sub>5</sub> I <sub>16</sub>	5	1.075	23.39	75.8	19.06	5
DMF/		F	1 1 2	22.42	76 71	10 11	C
Anisole	( <i>p</i> -FPEA) <sub>2</sub> (FA) <sub>4</sub> PD <sub>5</sub> I <sub>16</sub>	Э	1.13	22.13	/6./1	19.11	0
DMF	(4F-PEA) <sub>2</sub> FA <sub>4</sub> Pb <sub>5</sub> I <sub>16</sub>	5	1.18	21.7	80.35	21.07	7
DMF	PDAFA <sub>3</sub> Pb <sub>4</sub> I <sub>13</sub>	4	1.10	17.30	72.5	13.8	8
DMF	$PDA_{0.9}PA_{0.2}FA_3Pb_4I_{13}$	4	1.09	18.9	77.7	16.0	9
DMF/IPA	$BA_2FA_3Pb_4I_{13}$	4	1.062	21.62	78.96	18.14	10
DMF	BDA(FA) <sub>3</sub> Pb <sub>4</sub> I <sub>13</sub>	4	1.15	19.5	76.4	17.2	11
DMF	$BA_2FA_2Pb_3I_{10}$	3	0.98	11.89	59	6.88	12
	$BA_2FA_3Pb_4I_{13}$	4	1.00	20.83	68.7	14.31	This
TEP/DEE							Work
		4	1.02	23.34	72.5	17.42	This
IEP/PE	ва <sub>2</sub> га <sub>3</sub> гр <sub>4</sub> I <sub>13</sub>		1.03				Work

n defines the number of inorganic octahedron  $(MX_6)^{4-}$  slabs.

Antisolvent	A <sub>1</sub>	$ au_1$ (ns)	A <sub>2</sub>	$ au_{2}$ (ns)	Average life time (ns)
S					
DBE	60%	38.1	40%	137.0	160
PE	33%	27.2	67%	220.9	78

Table S4 The fitting results of TRPL.

**Table S5** The fitting results of impedance spectroscopy spectra.

Antisolvents	R <sub>s</sub> (Ω) R <sub>rec</sub> (Ω		
DEE	53.11	7984	
PE	34.5	56676	

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