Supplementary Information

Enhanced Crystallinity of CH₃NH₃PbI₃ by the Pre-Coordination of PbI₂-DMSO Powders for Highly Reproducible and Efficient Planar Heterojunction Perovskite Solar Cells

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Figure S1. Thermogravimetric analysis (TGA) of the pure PbI_2 and pre-coordinated PbI_2 -DMSO powders. The heating rate was 2°C/min under ambient air environment.



Figure S2. TGA analyses of PbI_2 -DMF and DPC-10 min powders before and after dissolving in DMF. The TGA data of DPC-10 min powders before dissolving were reproduced from Figure S1 for comparison.



Figure S3. Top-view SEM images of CH₃NH₃PbI₃ films prepared without DMSO (A), by the one-step blending method (B), and by the DPC method (C-E). The scale bar is 2 μ m. (F) The number of pin-holes in the perovskite films prepared by the DPC method (counting area = 400 μ m²).



Figure S4. Cross-sectional SEM images of the CH₃NH₃PbI₃ films prepared by the one-step blending method (A) and DPC method (B-D). The scale bar is 200 nm.

Structure	(A) CH ₃ NH ₃ Pbl ₃ /Glass			
Method	One-step blending		DPC	
$^{ au}$ (ns)	$ au_1$	$ au_2$	$ au_1$	$ au_2$
	2.03	13.61	2.51	17.08
A (%)	A_1	A ₂	A_1	A ₂
	54	47	48	52
Average carrier lifetime (ns)	11.93		15.34	
Structure	(B) CH ₃ NH ₃ Pbl ₃ /TiO ₂ /Glass			
Method	One-step blending		DPC	
au (ns)	$ au_1$	$ au_2$	$ au_1$	$ au_2$
	1.76	9.49	1.43	8.84
A (%)	<i>A</i> ₁	A ₂	<i>A</i> ₁	A ₂
	53	47	66	34
Average carrier lifetime (ns)	8.16		7.07	
Structure	(C) Spiro-OMeTAD/CH ₃ NH ₃ PbI ₃ /Glass			
Method	One-step blending		DPC	
au (ns)	$ au_1$	$ au_2$	$ au_1$	$ au_2$
	0.57	5.02	0.55	3.47
A (%)	<i>A</i> ₁	A ₂	<i>A</i> ₁	A ₂
	54	46	58	42
Average carrier lifetime (ns)	4.49		2.95	

Table S1. Bi-exponential decay parameters and average life time of TRPL analysis. (A)Perovskie/Glass. (B) Perovskite/TiO2/Glass. (C) Spiro-OMeTAD/Perovskite/Glass.



Figure S5. The hysteresis analysis of *J-V* curves depending on the scan direction. The voltage scan rate was 0.3V/s. (A) One-step blend mixing method. The forward scan and average PCE values were 13 and 15 %. (B) DPC method. The forward scan and average PCE values were 15.2 and 16.7 %.



Figure S6. The performance comparison of perovskite solar cells prepared by the DPC method (mechanical mixing time = 3, 6, 10 and 15 min). (A) The best J-V curves. (B) Power conversion efficiency. Five cells were analyzed for each type.



Figure S7. The performance comparison of perovskite solar cells prepared by the one-step method (stirring time = 12 h and 36 h) (A) The best J-V curves. (B) Power conversion efficiency. Five cells were analyzed for each type.