Supplementary Information

Enhanced Crystallinity of CH$_3$NH$_3$PbI$_3$ by the Pre-Coordination of PbI$_2$-DMSO Powders for Highly Reproducible and Efficient Planar Heterojunction Perovskite Solar Cells

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Figure S1. Thermogravimetric analysis (TGA) of the pure PbI₂ and pre-coordinated PbI₂-DMSO powders. The heating rate was 2°C/min under ambient air environment.
**Figure S2.** TGA analyses of PbI₂-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 CH$_3$NH$_3$PbI$_3$ films prepared without DMSO (A), by the one-step blending method (B), and by the DPC method (C-E). The scale bar is 2 μm. (F) The number of pin-holes in the perovskite films prepared by the DPC method (counting area = 400 μm$^2$).
Figure S4. Cross-sectional SEM images of the CH$_3$NH$_3$PbI$_3$ films prepared by the one-step blending method (A) and DPC method (B-D). The scale bar is 200 nm.
Table S1. Bi-exponential decay parameters and average life time of TRPL analysis. (A) Perovskie/Glass. (B) Perovskite/TiO$_2$/Glass. (C) Spiro-OMeTAD/Perovskite/Glass.

| Structure                        | (A) CH$_3$NH$_3$PbI$_3$/Glass | (B) CH$_3$NH$_3$PbI$_3$/TiO$_2$/Glass | (C) Spiro-OMeTAD/CH$_3$NH$_3$PbI$_3$/Glass |
|---------------------------------|--------------------------------|--------------------------------------|------------------------------------------|
| Method                          | One-step blending               | DPC                                  |                                          |
| $\tau$ (ns)                     | $\tau_1$ $\tau_2$ $\tau_1$ $\tau_2$ | $\tau_1$ $\tau_2$ $\tau_1$ $\tau_2$ |                                          |
|                                | 2.03 13.61 2.51 17.08          | 1.76 9.49 1.43 8.84                   | 0.57 5.02 0.55 3.47                      |
| $A$ (%)                         | $A_1$ $A_2$ $A_1$ $A_2$        | $A_1$ $A_2$ $A_1$ $A_2$              | $A_1$ $A_2$ $A_1$ $A_2$                |
|                                | 54 47 48 52                     | 53 47 66 34                            | 54 46 58 42                            |
| Average carrier lifetime (ns)   | 11.93                          | 8.16                                  | 4.49                                    |
|                                | 15.34                          | 7.07                                  | 2.95                                    |
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.