Electronic Supplementary Information

Room-Temperature Crystallization of Hybrid-Perovskite Thin Films *via* Solvent-Solvent Extraction for High-Performance Solar Cells

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Supplementary Figures:



Fig. S1. Photographs of the SSE process in DEE bath: (A) as-spin-coated MAPbI₃ precursor thin film, (B) during SSE (2 s), and (C) after SSE (2 min).



Fig. S2. Sequence of photographs after dropping a spin-coated MAPbI₃ precursor thin film (patterned) in DEE bath showing rapid MAPbI₃ perovskite thin film formation.



Fig. S3. Sequence of photographs after adding drops of $MAPbI_3$ precursor solution (in NMP) in DEE bath showing sluggish $MAPbI_3$ perovskite precipitate formation.



Fig. S4. Cross-sectional SEM images of MAPbI₃ perovskite films of various thicknesses deposited on TiO₂blocking-layer coated FTO-glass substrates using the SSE process at room temperature: (A) ~50 nm, (B) ~80 nm, (C) ~250 nm, and (D) ~410 nm.



Fig. S5. Cross-sectional bright-field TEM image of a \sim 20-nm MAPbI₃ perovskite film deposited on TiO₂blocking-layer coated FTO-glass substrate.



Fig. S6. Bright field TEM images of a typical MAPbI₃ perovskite-based solar cell, where the MAPBI₃ thin film (~700 nm thickness) is deposited using the SSE process at room temperature: (A) overall view, (B) detailed view of region 'B' showing hole-transporting material (HTM) and MAPbI₃, (C) detailed view of region 'C' showing MAPbI₃ grains, and (D) detailed view of region 'D' showing MAPbI₃, TiO₂ blocking layer and FTO. The thickness of the MAPbI₃ layer in this solar cell is unusually thick, and Au was used as the top contact instead of the Ag used in the other cells.



Fig. S7. Elemental EDS mapping of the solar cell in Fig. S6 showing the distribution of elements Sn, Ti, Pb, I, and Au.



Fig. S8. Optical properties of MAPbI₃ perovskite films of various thicknesses deposited on TiO_2 -blockinglayer coated FTO-glass substrates using the SSE process at room temperature: (A) transmission, (B) reflection, and (C) absorption. Inset in (B) are photographs of the films.



Fig. S9. *J-V* characteristics of a PSC based on 250-nm SSE MAPbI₃ perovskite thin film, under simulated one-sun AM 1.5G (100 mW cm⁻²) illumination, in forward (normal) and reverse scan.



Fig. S10. Mean, minimum and maximum PCE values of PSCs tested under simulated one-sun AM 1.5G (100 mW cm⁻²) illumination (0.16 cm² typical active area) as a function of SSE MAPbI₃ perovskite film thickness. About 10-20 solar cells were tested for each thickness.



Fig. S11. (A) Typical Nyquist plots of the impedance responses for a solar cell made from SSE MAPbI₃ film of ~250 nm thickness, with three different bias voltages. The impedance spectra are dominated by a large semicircle at low frequencies. (B) Plots of recombination resistance as a function of voltage for solar cells with different thickness SSE MAPbI₃ perovskite films.



Fig. S12. (A) XRD pattern and (B) SEM micrograph of top surface of 250-nm SSE MAPbI₃ perovskite thin film that has been heat-treated at 100 °C for 15 min in air.



Fig. S13. Mean, minimum and maximum PCE values of PSCs tested under simulated one-sun AM 1.5G (100 mW cm⁻²) illumination (0.16 cm² typical active area) as a function of annealing heat-treatment (100 °C, 15 min, in air) of SSE MAPbI₃ perovskite films (~250 nm thickness). About 10-20 solar cells each were tested.