Electronic Supporting information

Cs$_{0.13}$FA$_{0.87}$PbI$_3$ perovskite solar cells for concentrator photovoltaic applications

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Fabrication of perovskite devices.

Pre-patterned ITO substrates (8 Ω sq⁻¹, Xin Yan Technology) were cleaned by sonicating in deionized water, followed by acetone and isopropyl alcohol before being dried and treated for 10 minutes in an oxygen plasma. A SnO₂ electron transport layer was deposited by spin coating a 0.05M solution of SnCl₄.2H₂O (Sigma Aldrich) in isopropanol at 3000 rpm for 30 seconds, after which films were annealed at 180°C for 60 minutes to produce a 40 nm thick layer. Upon cooling, samples underwent the deposition of a secondary electron transport layer in the form of a 2mg ml⁻¹ PC₆₀BM (Sigma Aldrich) solution in chlorobenzene spin-coated at 3000 rpm for 20 seconds before being annealed overnight at 80°C in a glovebox.

To form the Cs₀.₁₅FA₀.₈₅PbI₃ perovskite, a precursor solution was prepared by dissolving 0.188 M Cesium iodide (Sigma Aldrich), 1.06 M formamidinium iodide (Greatcell Solar) and 1.25 M Lead iodide (TCI Chemicals) in a 4:1 by volume solution of DMF:DMSO (both Sigma Aldrich). Cs₀.₁₃FA₀.₈₇PbI₃ films were deposited on substrates by spin-coating 100 μl of precursor solution at 7000rpm for 20 seconds, half way through deposition, 80 μl chlorobenzene was dropped onto the spinning substrate. After spin-coating, samples were immediately transferred to a hotplate and annealed at 110°C for 15 minutes.

To form the MAPbI₃ perovskite, a precursor solution was prepared by dissolving 1.25M methylammonium iodide (Greatcell Solar) and 1.25M lead iodide (TCI Chemicals) in a 4:1 by volume solution of DMF:DMSO (both Sigma Aldrich). This precursor was deposited by spin-coating100 μl of precursor solution at 4000rpm for 30 seconds, half way through deposition, 150 μl chlorobenzene was dropped onto the spinning substrate. After spin-coating, samples were immediately transferred to a hotplate and annealed at 100°C for 15 minutes.

For a hole transporting layer, 85 mg ml⁻¹ Spiro-MeOTAD (99%, Sigma Aldrich) was dissolved in chlorobenzene with the addition of 30 μl ml⁻¹ 4-tert-butylpyridine (Sigma Aldrich) and 20 μl ml⁻¹ Li-TFSI solution (520 mg ml⁻¹ in acetonitrile, Sigma Aldrich). This solution was spin-coated onto the perovskite layer at 2500 rpm for 30 seconds to produce a 150 nm thick layer. Device fabrication was completed by depositing metal electrodes through a shadow mask at 10⁻⁷ mTorr (Angstrom Engineering): first by evaporating 10 nm MoO₃, followed by 100 nm of Ag. The active area of all devices was 3.2 mm² as defined by the overlap between metal and ITO electrodes.

Characterization of perovskite devices.

For AM1.5 1 sun measurements, solar cells were illuminated by a WaveLabs Sinus (>AAA rated) solar simulator calibrated against a NIST traceable, KG5 filtered Si reference cell. A Keithley 2400 source meter applied and recorded respective bias voltages and photocurrents. Devices’ J-V curves were scanned in both directions between -0.2 V and 1.2 V at 50 mV s⁻¹ with no preconditioning.

External quantum efficiency measurements were performed in AC mode (100 Hz chopping frequency) on a Newport IQE-200, calibrated against a Si reference diode. Step size was set to 10 nm with a 5 second integration time between steps.
High light intensity measurements were performed in the glovebox by illuminating the devices with a 450 nm laser-diode. By driving the laser intensity with a waveform generator (Agilent 33500B) and measuring the light intensity with a highly linear photodiode, we were allowed to reproducibly adjust the light intensity with an error below 0.5% over a range of 0.2 to 13 suns.

In transient photovoltage (TPV) measurements, a 405 nm laser-diode was settled for keeping the solar cells in approximately $V_{oc}$ condition. Driving the laser intensity with a waveform generator Agilent 33500B and measuring the light intensity with a highly linear photodiode allowed to reproducibly adjust the light intensity with an error below 0.5% over a range of 0.2 to 13 suns. A small perturbation was induced with a second 405 nm laser diode driven by a function generator from Agilent. The intensity of the short (50 ns) laser pulse was adjusted to keep the voltage perturbation below 10 mV, typically at 5 mV. After the pulse, the voltage decays back to its steady state value in a single exponential decay. The characteristic decay time was determined from a linear fit to a logarithmic plot of the voltage transient and returned the small perturbation charge carrier lifetime. In transient photocurrent (TPC) measurements a 405 nm laser diode driven by the function generator was used to generate 100 $\mu$s light pulses. The current response was amplified with a transimpedance amplifier and recorded with a digital oscilloscope.

To measure device stability at 1 sun, solar cells were placed inside a multiplexed flow cell chamber with a constantly flowing nitrogen purge. These chambers were illuminated using a white LED light source adjusted to yield the same solar cell photocurrent output as under AM1.5G conditions. Devices were held at open-circuit conditions between measurements, which were performed every 30 minutes. Device temperature was measured to be 40°C throughout the experiment. For stability at high light intensities, a 405 nm laser diode was used to illuminate individual cells in a nitrogen glovebox, with devices held at open-circuit between periodic measurements.
Figure S1. Current-Voltage characteristics of Cs$_{0.15}$FA$_{0.85}$PbI$_3$ and MAPbI$_3$ perovskite devices under forward and reverse J-V sweeps, demonstrating very little hysteresis.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$V_{OC}$ (V)</th>
<th>$J_{SC}$ (mA cm$^{-2}$)</th>
<th>Fill Factor</th>
<th>PCE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MAPbI$_3$</td>
<td>1.06</td>
<td>21.5</td>
<td>0.78</td>
<td>17.8</td>
</tr>
<tr>
<td>Cs$<em>{0.15}$FA$</em>{0.85}$PbI$_3$</td>
<td>1.04</td>
<td>23.5</td>
<td>0.74</td>
<td>18.0</td>
</tr>
</tbody>
</table>

Table S1. J-V parameters from devices in main text Figure 1, measured from $V_{OC}$ to $J_{SC}$ at 1 sun.
Figure S2. Statistics from J-V curves from both MAPbI$_3$ and Cs$_{0.15}$FA$_{0.85}$PbI$_3$ perovskite solar cells.

Figure S3. $G_{\text{mp}}$ values extracted from Figure 3 in the main text for MAPbI$_3$ and Cs$_{0.15}$FA$_{0.85}$PbI$_3$ under 1 sun and 13 suns illumination.
Figure S4. Light absorption characteristics of MAPbI$_3$ and Cs$_{0.15}$FA$_{0.85}$PbI$_3$ films pre and post light soaking at 13 suns for 6 hours under nitrogen.

Figure S5. XRD patterns for perovskite films on glass for both MAPbI$_3$ and Cs$_{0.15}$FA$_{0.85}$PbI$_3$ pre and post light soaking at 13 suns for 6 hours under nitrogen.