Supporting Information

Efficient ambient-air-stable HTM-free carbon-based perovskite solar cells with hybrid 2D-3D lead halide photoabsorber

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Fig. S1 (a) Top view SEM image and (b) XRD pattern of fresh EA₂PbI₄ film.

Fig. S2 Photovoltaic characterization of (EA)₂(MA)ₙ₋₁PbₙI₃n+₁ films. (a) Absorption spectra of the (EA)₂(MA)ₙ₋₁PbₙI₃n+₁ films, (b) Band gap energy (E٪) of (EA)₂(MA)ₙ₋₁PbₙI₃n+₁ films corresponding to UV-Vis absorption spectra, and (c) Normalized steady-state PL spectra of (EA)₂(MA)ₙ₋₁PbₙI₃n+₁ films illuminated from the front side of the films on silica glass.
Fig. S3 Steady-state PL spectra of \((\text{EA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}\) films illuminated from the front and back sides of the films on silica glass. (a) \(n = 6\), (b) \(n = 10\), (c) \(n = 20\), and (d) \(n = \infty\).

Fig. S4 (a, b) Cross-sectional SEM images of ITO/C_{60}/perovskite/C device.
Fig. S5 $J$-$V$ curves of the champion C-PSCs with different 2D-3D hybrid perovskites ($n = 6$) measured with reverse scans.

Fig. S6 Electrochemical properties. (a) Nyquist plots of the C-PSCs with $(\text{EA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ measured at 0.8 V in dark condition measured with frequency ranging from $10^6$ to 1 Hz, (b) the equivalent circuit model for C-PSCs in EIS, and (c) variation of $R_{\text{rec}}$ under different bias voltages from 0 to 1.0 V.
Fig. S7 Steady-state PL spectra of $(\text{EA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ films illuminated from the front side of the films on silica glass.

Fig. S8 Top view SEM images of $(\text{EA})_2(\text{MA})_{n-1}\text{Pb}_n\text{I}_{3n+1}$ films: (a) $n = \infty$, (b) $n = 20$, (c) $n = 10$, and (d) $n = 6$. Pin-holes are labeled in red circles.
Fig. S9 Zoomed-in XRD patterns of (EA)$_2$(MA)$_{n-1}$Pb$_n$I$_{3n+1}$ films on ITO substrate in the region of 7° – 15°.

Table S1 Photovoltaic parameters of the best-performing C-PSCs with different 2D-3D hybrid perovskites (n = 6) measured with reverse scans.

<table>
<thead>
<tr>
<th>Samples</th>
<th>$V_{OC}$ (V)</th>
<th>$J_{SC}$ (mA cm$^{-2}$)</th>
<th>FF (%)</th>
<th>PCE (%)</th>
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<tbody>
<tr>
<td>(EA)$_2$(MA)$_6$Pb$<em>6$I$</em>{19}$</td>
<td>1.018</td>
<td>21.14</td>
<td>55.21</td>
<td>11.88</td>
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<td>(BA)$_2$(MA)$_5$Pb$<em>6$I$</em>{19}$</td>
<td>1.025</td>
<td>19.26</td>
<td>53.38</td>
<td>10.54</td>
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<tr>
<td>(PEA)$_2$(MA)$_5$Pb$<em>6$I$</em>{19}$</td>
<td>1.031</td>
<td>18.21</td>
<td>50.40</td>
<td>9.46</td>
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