Supplementary Information for

Ultra-flexible perovskite solar cells with crumpling durability: Toward a wearable power source

Gunhee Lee\textsuperscript{a,b,†}, Min-cheol Kim\textsuperscript{a,c,†}, Yong Whan Choi\textsuperscript{a,d}, Namyoung Ahn\textsuperscript{a}, Jihun Jang\textsuperscript{a}, Jungjin Yoon\textsuperscript{a}, Sang Moon Kim\textsuperscript{e}, Jong-Gu Lee\textsuperscript{f}, Daeshik Kang\textsuperscript{g}, Hyun Suk Jung\textsuperscript{h,*} and Mansoo Choi\textsuperscript{a,b,*}

\textsuperscript{a}Global Frontier Center for Multiscale Energy Systems, Seoul National University, Seoul, Republic of Korea.

\textsuperscript{b}Department of Mechanical Engineering, Seoul National University, Seoul, Republic of Korea.

\textsuperscript{c}Department of NanoEngineering, University of California, San Diego, 9500 Gilman Drive, LaJolla, California 92093, United States.

\textsuperscript{d}Division of Mechanical Convergence Engineering, Silla University, Busan, Republic of Korea.

\textsuperscript{e}Department of Mechanical Engineering, Incheon National University, Incheon, 406-772, Republic of Korea.

\textsuperscript{f}School of Mechanical and Aerospace Engineering/IAMD, Seoul National University, Gwanak 599, Gwanak-ro, Gwanak-gu, Seoul 151-742, Republic of Korea.

\textsuperscript{g}Department of Mechanical Engineering, Ajou University, San 5, Woncheon-dong, Yeongtong-gu, Suwon 443-749, Republic of Korea.

\textsuperscript{h}School of Advanced Materials Science & Engineering, Sungkyunkwan University, Suwon, Gyeonggi-do, Republic of Korea.

† Electronic supplementary information (ESI) available. See DOI: XX.XXXX/XX
‡ These authors contributed equally to this work.
*To whom correspondence should be addressed. E-mail: hsjung1@skku.edu; mchoi@snu.ac.kr
This Supplementary Information includes:

Fig. S1 to S12
Table S1 to S3
Captions for Movies S1
References
Fig. S1. Schematic illustration and cross-sectional SEM images of device structure of the flexible perovskite solar cell.
**Fig. S2.** $J-V$ curves of the perovskite solar cells before and after cyclic bending tests (R = 0.5 mm, 1,000 cycles) with different substrate thicknesses.
Fig. S3. SEM Images of PEDOT:PSS surface after cyclic bending test for 30 μm substrate thickness at different bending radius ($R = 1$ and 0.5 mm)
Fig. S4. FEM simulation results of applied stress on the perovskite film with different substrate thicknesses at the fixed bending radius of (a) 0.5 mm and (b) 1 mm.
Fig. S5. Histograms of PCE of 45 devices using 2.5 μm substrate
Fig. S6. (a) Steady-state photocurrent density (black) and power conversion efficiency (blue) measured at the maximum power voltage of 0.81 V for 200 sec. (b) EQE spectra (blue) and the integrated $J_{SC}$ (red) of perovskite solar cell.
Fig. S7. Normalized (a) $V_{OC}$, (b) $J_{SC}$ and (c) FF as a function of bending cycles depending on bending radius ($R = 2$, 1, and 0.5 mm) at the 2.5 μm thick substrate.
Fig. S8. Histograms of PCE of 20 devices of large area (1.2 cm²) flexible perovskite solar cells
Fig. S9. Normalized PCEs devices as a function of (a) bending, (b) crumpling and (c) folding cycles depending on with and without protecting layer. (1.2 cm² active area device)
Neutral plane (NP) defines the position where the strains are zero in the film. Its position is the utmost important to determine the best location for the critical parts of the device. The determination for neutral plane starts from the fact that the total amount of force across the cross section must be 0, which can be denoted as below using stress ($\sigma_x$).

$$\int \sigma_x \, dA = 0 \quad (1)$$

From Hooke’s law, stress ($\sigma_x$) can be described from strain ($\varepsilon_x$) and elastic modulus ($E$), therefore equation (1) can be rewritten as below. ($y$ is the distance from neutral plan, $R$ is radius of curvature as depicted in Fig. S10a)

$$\sigma_x = E \varepsilon_x = \frac{Ey}{R} \quad (2)$$

$$\int Ey \, dA = 0 \quad (3)$$

For a homogeneous monolayer beam, the distance of neutral plane from the top ($h$) should be at the center as a result of equation (3).

The total amount of force across the entire film of a neutral plane should be zero for multilayer stacks such as perovskite solar cells. If there are $n$ layers having different elastic modulus and thickness, equation (3) should be as below.

$$\sum_{i=1}^{n} \int E_i y_i \, dA_i = 0 \quad (4)$$

As $y_i$s are the distances from the neutral axis to the centroid of each layer. Therefore $y_i$ can be expressed with the distance of neutral axis from the top ($h$) and thickness of each materials ($t_i$) as can be seen in Fig. S10b. Note, $y_i$ will be negative if the centroid of the material area is below the neutral axis. As a result, $h$ can be calculated as below.$^2$
$$h = \sum_{i=1}^{n} E_i t_i \left( \sum_{j=1}^{i} t_j - \frac{t_i}{2} \right)$$

$$\sum_{i=1}^{n} E_i t_i$$

(5)

**Fig. S11.** Schematic images of detailed fabrication process of parylene film on flexible perovskite solar cell. First, the parylene powder was vaporized when heated up to 150 °C, and was formed into the dimer gas. Second, as the dimer gas passed through the furnace heated to 690°C, it was changed into monomer gas. And finally, the monomer gas was deposited on the surface of the flexible perovskite solar cells in the deposition chamber (25 ~ 40 °C and 5 x 10^-2 torr) to form parylene film.
Fig. S12. Schematic images of detailed fabrication process of metal mesh grid on PET substrate. (Photolithography was performed with the MA150e Aligner and Au deposition with the EI-5 E-beam evaporator.)
<table>
<thead>
<tr>
<th></th>
<th>$V_{OC}$ (V)</th>
<th>$J_{SC}$ (mA/cm$^2$)</th>
<th>FF</th>
<th>PCE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Small area</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(0.0756 cm$^2$)</td>
<td>Average</td>
<td>0.95 ± 0.02</td>
<td>21.23 ± 0.66</td>
<td>79.47 ± 1.93</td>
</tr>
<tr>
<td></td>
<td>Best</td>
<td>0.96</td>
<td>22.45</td>
<td>79.39</td>
</tr>
<tr>
<td><strong>Large area</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(1.2 cm$^2$)</td>
<td>Average</td>
<td>0.90 ± 0.03</td>
<td>20.16 ± 1.00</td>
<td>67.68 ± 3.97</td>
</tr>
<tr>
<td></td>
<td>Best</td>
<td>0.87</td>
<td>20.76</td>
<td>75.45</td>
</tr>
</tbody>
</table>

**Table S1.** Photovoltaic parameters of flexible perovskite solar cells corresponding to the active area.
<table>
<thead>
<tr>
<th></th>
<th>Average (Ω/□)</th>
<th>Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>PEDOT:PSS</td>
<td>234.3</td>
<td>4.64</td>
</tr>
<tr>
<td>Gold Mesh/PEDOT:PSS</td>
<td>18.0</td>
<td>1.08</td>
</tr>
</tbody>
</table>

**Table S2.** Sheet resistances of PEDOT:PSS/PET and PEDOT:PSS/gold mesh/PET substrate.
<table>
<thead>
<tr>
<th></th>
<th>Parylene</th>
<th>Cu</th>
<th>BCP</th>
<th>C₆₀</th>
<th>Perovskite</th>
<th>PEDOT:PSS</th>
<th>PET</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elastic modulus (Gpa)</td>
<td>4.7</td>
<td>128</td>
<td>12</td>
<td>12</td>
<td>18.5</td>
<td>3</td>
<td>2.5</td>
</tr>
<tr>
<td>Thickness (nm)</td>
<td>2300</td>
<td>70</td>
<td>10</td>
<td>20</td>
<td>500</td>
<td>120</td>
<td>2500</td>
</tr>
</tbody>
</table>

**Table S3.** Elastic modulus and thicknesses of the materials constituting the flexible perovskite solar cell.³⁻¹²
**Movie S1.** Movie of crumpling durability test.
References


