

Supplementary Materials

Carbon-Based Perovskite Solar Cell Enables Graded Utilization of Full Solar Spectrum Beyond the Photovoltaic S-Q Limit via Hybrid Photovoltaics–Photothermal Conversion

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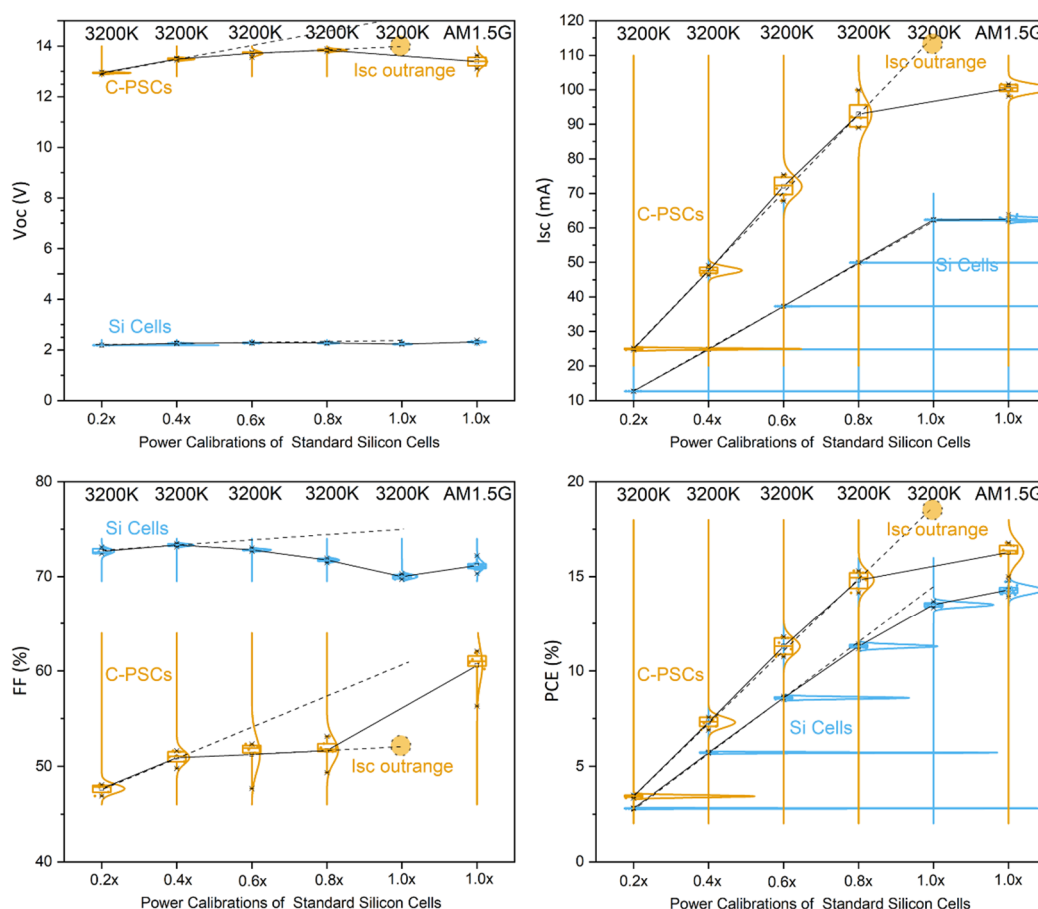


Figure S1. Performance statistics of C-PSCs and silicon cells under 3200 K light source with varying illumination intensities and under standard AM1.5G light source.



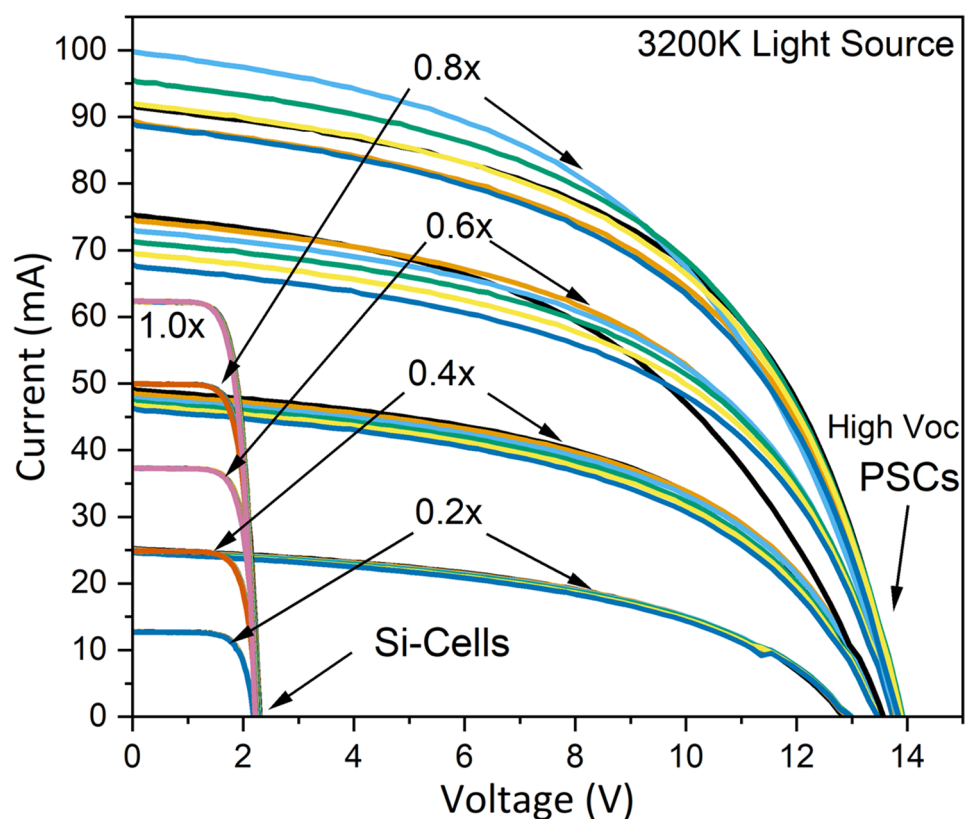


Figure S2. Current-voltage scan curves of C-PSCs and silicon cells under 3200 K light source with varying illumination intensities and under standard AM1.5G light source.

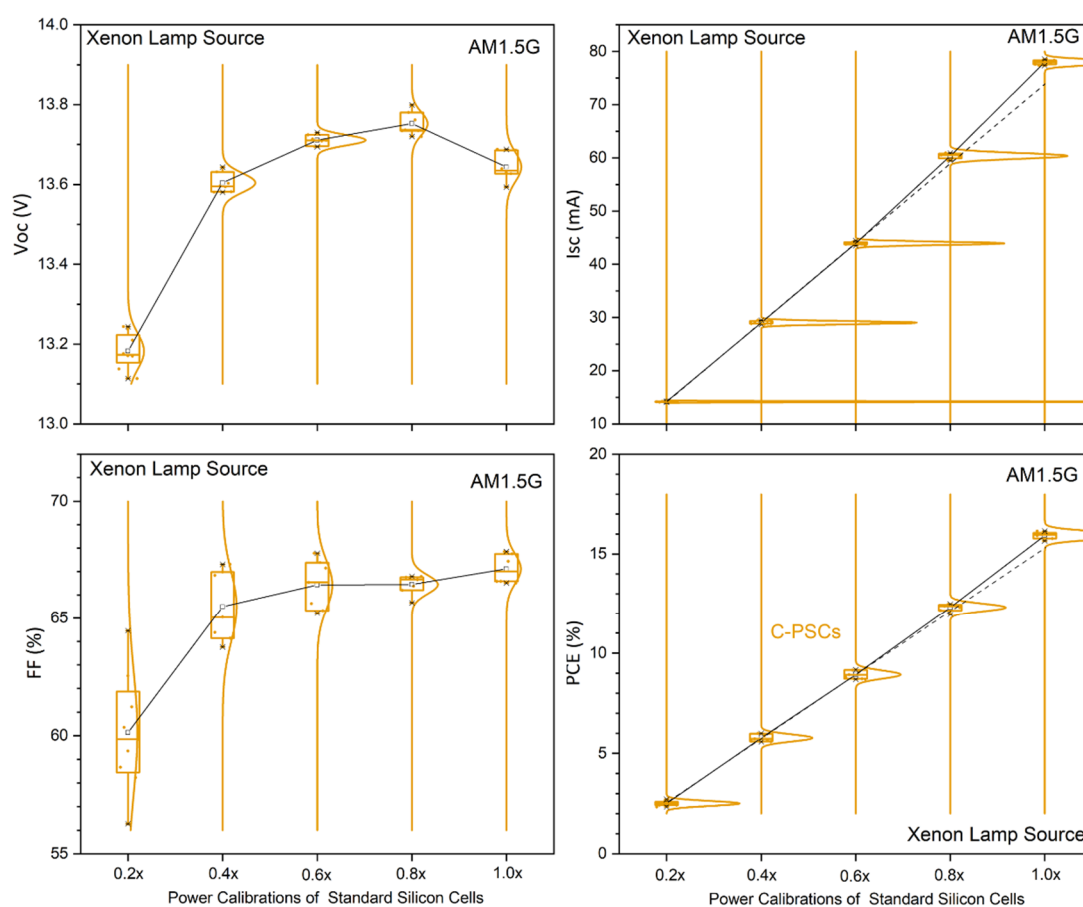


Figure S3. Performance statistics of C-PSCs under Xenon lamp source with AM1.5G filter with varying illumination intensities.

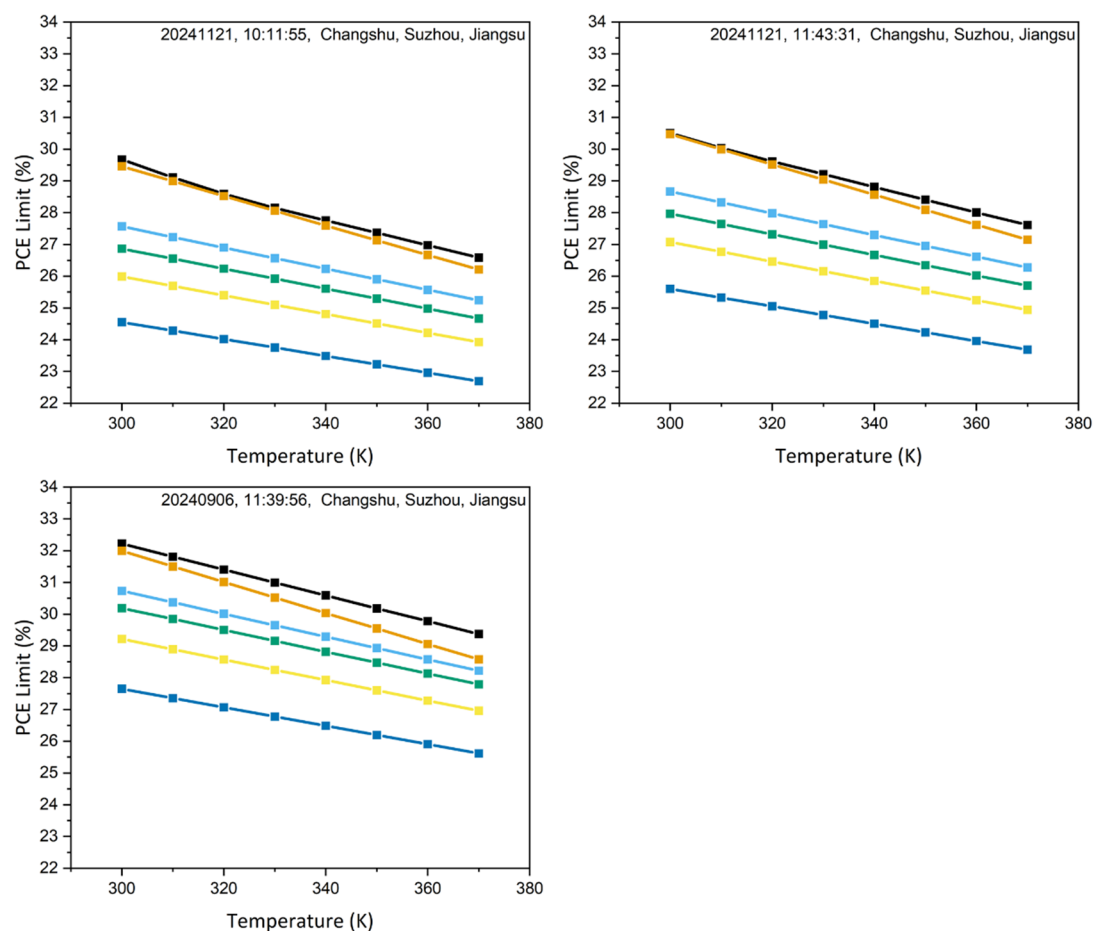


Figure S4. The line plots of PCE limits and temperature for different bandgap materials using the actual outdoor sunlight spectrum (detailed performance data are listed in the Supplementary Materials 2).

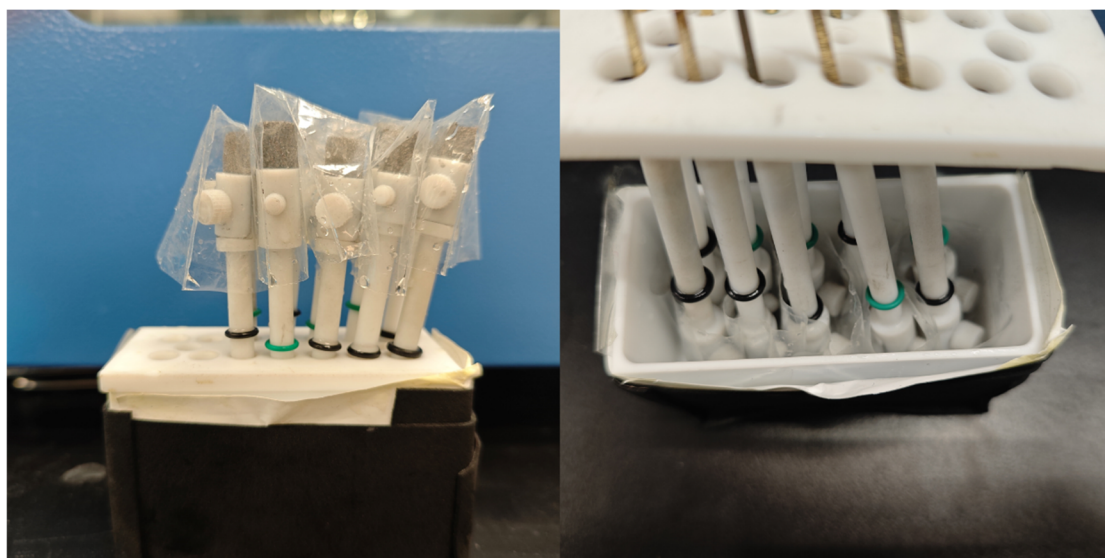


Figure S5. Schematic diagram of separating electrolytic cells using barrier bags.

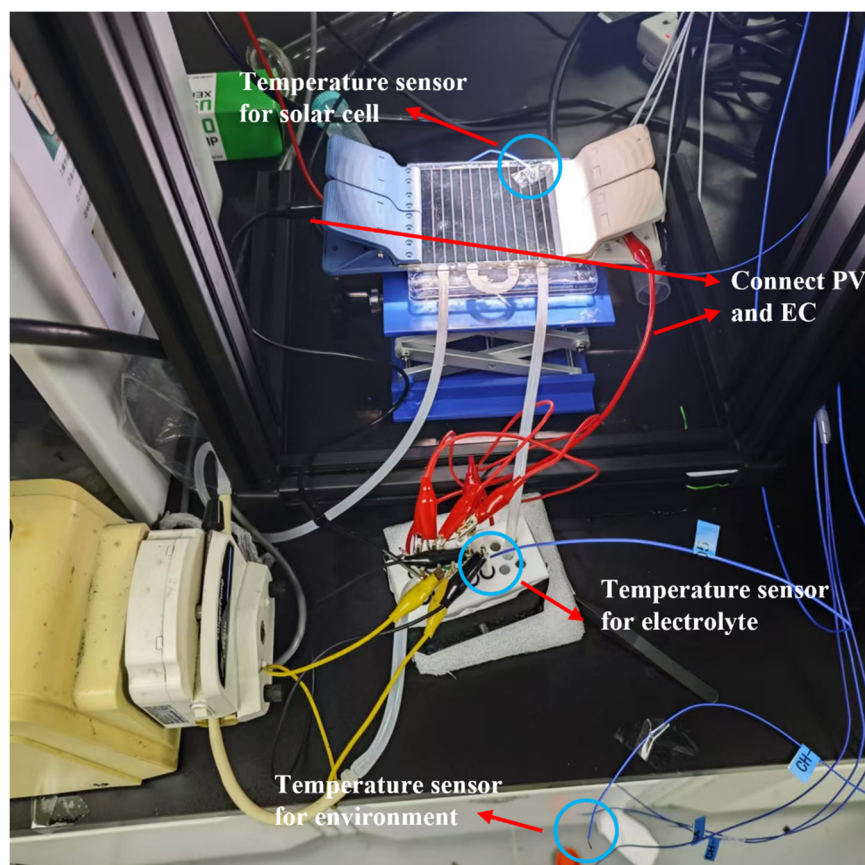


Figure S6. Graded solar energy utilization system for thermoelectric coupling water splitting demonstration.

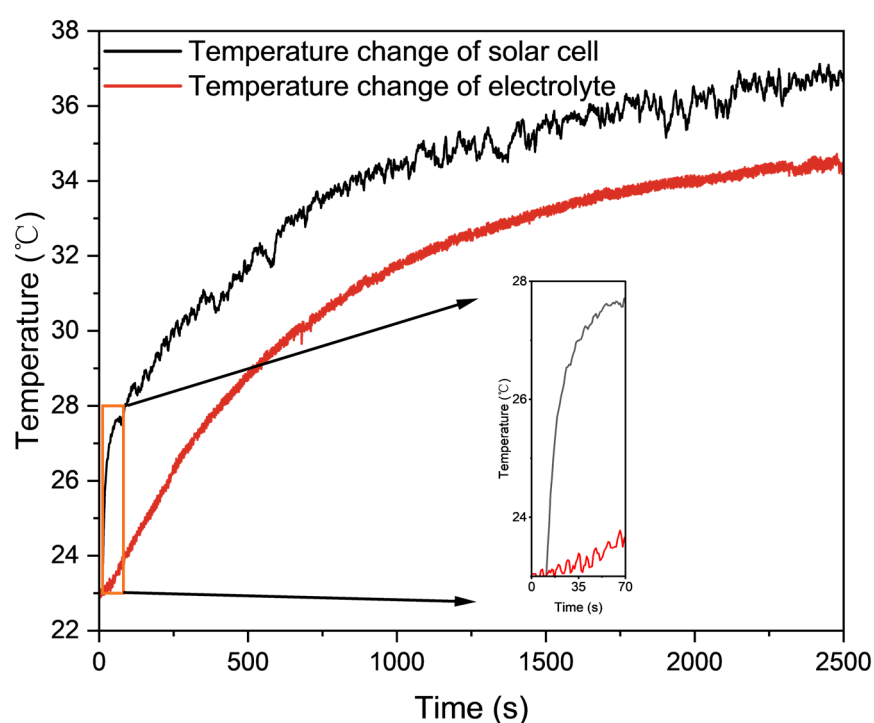


Figure S7. Variations of electrolyte temperature with time for solar cell and electrolyzer in the beginning 90 s.

In this system, 50 mL of 1 M KOH solution was employed both as an electrolyte and as a heat transfer medium. At the onset of operation, the temperature differential between the solar cell, the electrolyte, and the ambient environment was minimal. According to Newton's law of cooling, the rate of heat loss is proportional to the temperature difference [26]. For simplicity in calculating the rate of heat absorption, the initial heat loss during illumination was neglected; consequently, all heat absorbed by the system was assumed to contribute directly to the heating of the solar cell and electrolyte. During the first 70 s of operation, the temperature of the solar cell

increased linearly from 22.5 °C to 27.5 °C, while that of the electrolyte rose from 22.5 °C to 23.2 °C. Given that more than 99% of the mass in the solar cell consists of glass, its specific heat capacity (840 J/kg·K) was used as a proxy for that of the solar cell [27]. The heat absorbed by the solar cell during this period is calculated as:

$$Q_{\text{solar}} = C_{\text{glass}}m\Delta T = 840 \text{ J/(kg}\cdot\text{K)} \times 0.027 \text{ kg} \times 5 \text{ K} = 113.4 \text{ J},$$

and the heat absorbed by the electrolyte is:

$$Q_{\text{water}} = C_{\text{water}}m\Delta T = 4200 \text{ J/(kg}\cdot\text{K)} \times 0.05 \text{ kg} \times 0.7 \text{ K} = 147 \text{ J}.$$

Thus, the total energy absorbed by the system during this initial phase is 260.4 J, corresponding to a heat absorption power of 3.72 W. The light intensity used in the system was defined as AM1.5 (1000 W/m²), and the active area of the solar cell—laser-etched to 50 cm²—received a total incident power output of 5 W. As such, the heat collection efficiency of the system is calculated as 3.72/5 = 74.4%. Since the input solar power remains constant throughout, it was assumed that the rate of heat absorption by the system is also constant. As illustrated in Figure 4b, after approximately 2500 s of operation, the temperatures of both the solar cell and electrolyte stabilize. Over this time period, the total energy absorbed by the system reached 9300 J, with the electrolyte absorbing 2100 J ($Q_{\text{water}} = C_{\text{water}}m\Delta T = 4200 \times 0.05 \times 10 = 2100 \text{ J}$).

The rationale for omitting earlier heat loss calculations is based on the understanding that system heat losses are heavily dependent on fabrication and operational conditions. Nevertheless, the exceptional thermal performance of carbon-based perovskite solar cells relative to silicon has been demonstrated through their rapid temperature rise in response to solar irradiation. The ultra-high heat conversion capability of these materials was confirmed by surface temperature profiles of both the cell and electrolyte. Theoretically, carbon electrodes are capable of absorbing a wide range of solar spectral bands. This is supported by Figure 1e,f, where nearly all AM1.5G irradiation is absorbed by carbon-based structures. Furthermore, as shown in Figure 2c,d, the superior solar energy absorption and heat conversion efficiency of carbon-based solar cells over silicon has been conclusively demonstrated.

In summary, through effective thermal management, a solar hydrogen system can achieve a remarkable heat collection efficiency of 74.4%.

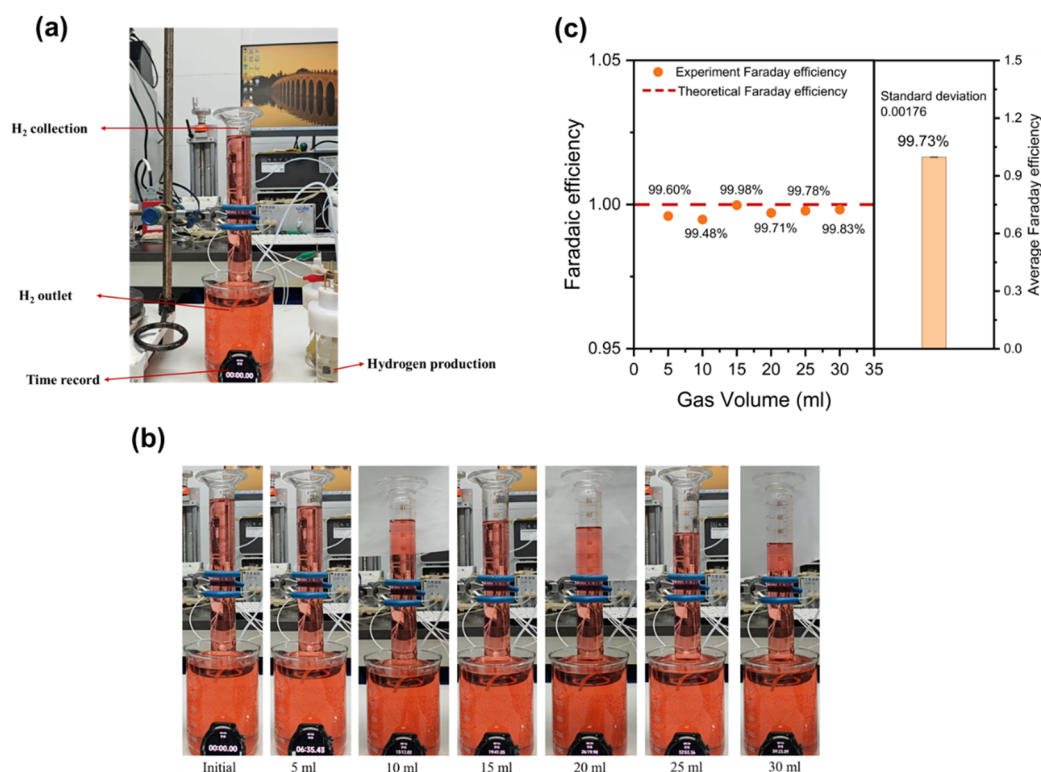


Figure S8. Faraday efficiency test (a) Faraday efficiency test set (b) Recording of volume of hydrogen production and corresponding time (c) Result of Faraday efficiency measurement six times (left), average faraday efficiency with error bar (right).

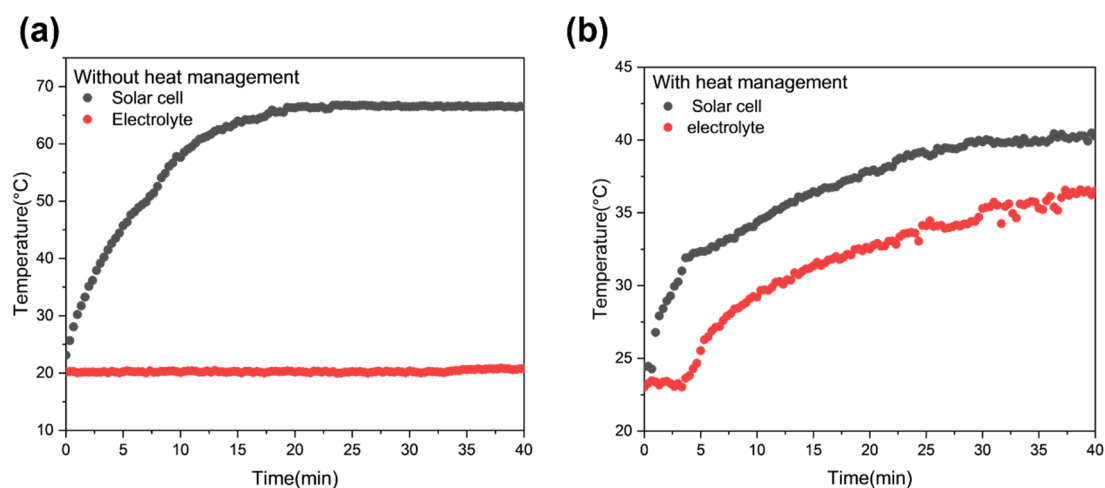


Figure S9. Temperature change of Solar cell and electrolyte (a) without heat management system and (b) with heat management system.

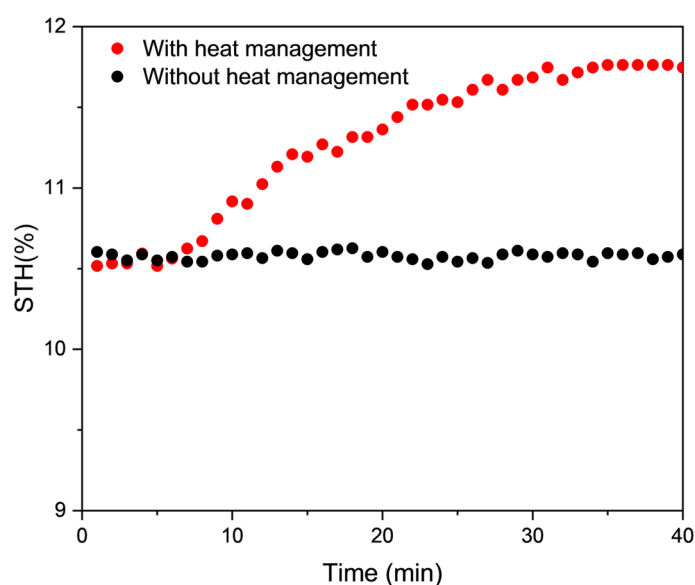


Figure S10. The solar cell module is cut into two series sub-cells to measure the change in STH of a single electrolytic cell over time.

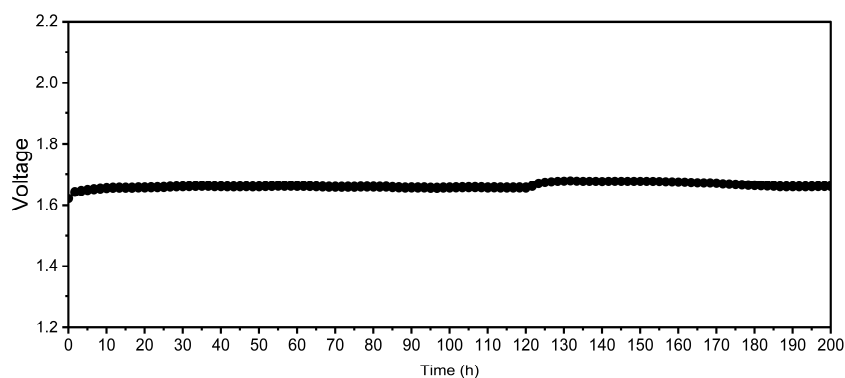


Figure S11. Stability of Pt/C||RuO₂ at 100 mA cm⁻² current density in two electrode system.

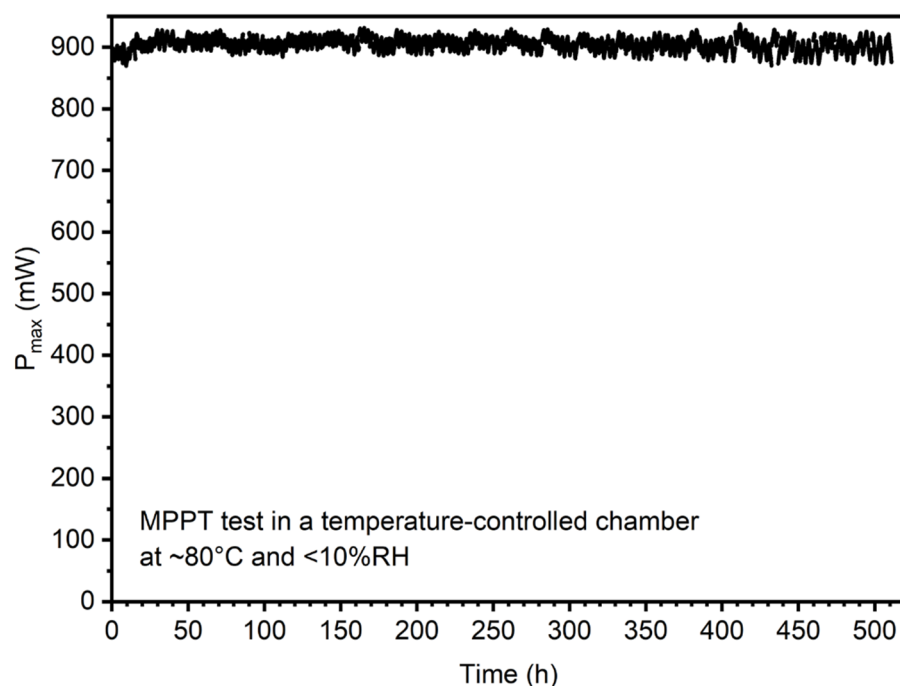


Figure S12. Maximum Power Point Tracking (MPPT) test of unencapsulated carbon electrode-based perovskite modules.

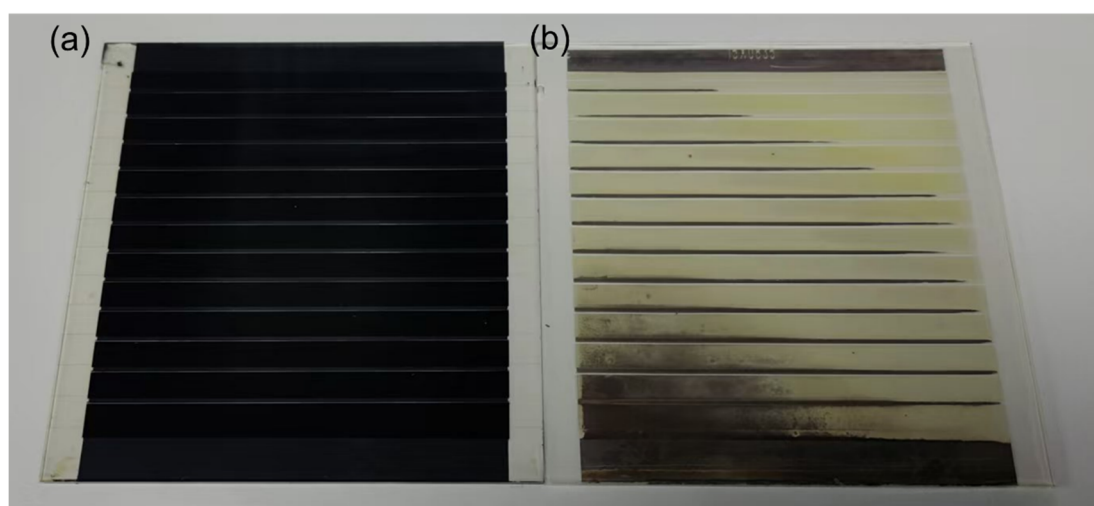


Figure S13. Photographs of (a) carbon electrode-based perovskite modules and (b) metal-electrode perovskite modules after being stored at room temperature (20–28 °C, 30–60% relative humidity) for five days.

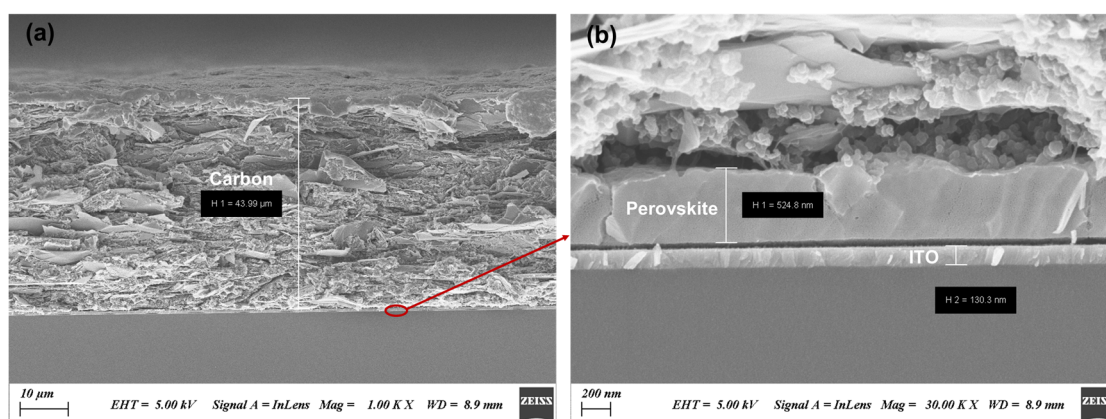


Figure S14. Scanning electron microscopy (SEM) cross-sectional images of the C-PSC. (a) Overall device stack, where H1 indicates the thickness of the carbon electrode. (b) Magnified view of the underlying layers, where H1 and H2 correspond to the thicknesses of the perovskite layer and the ITO layer, respectively.

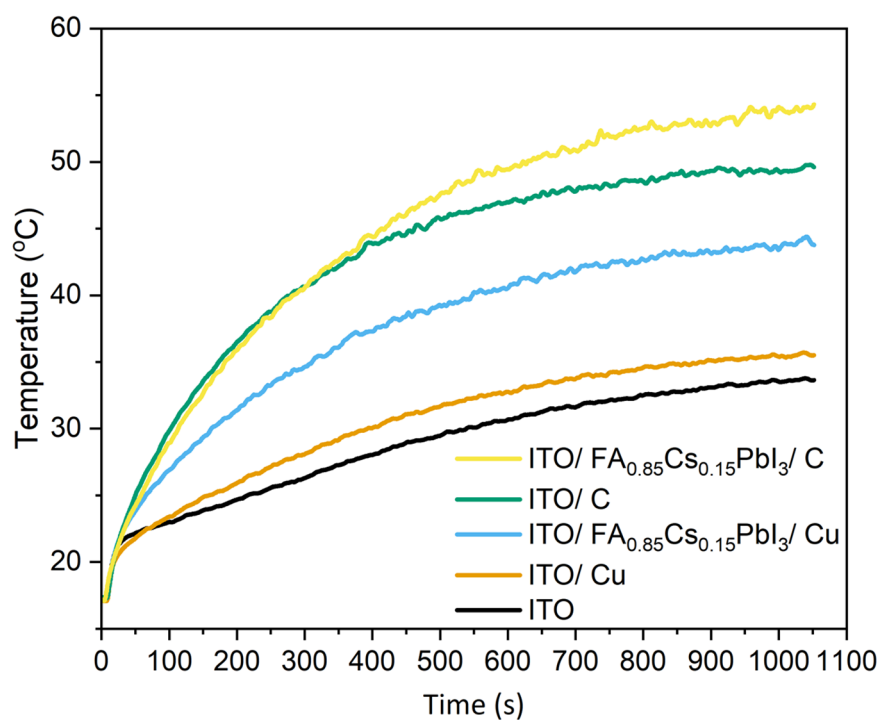


Figure S15. Temperature rise behavior of distinct film layer structures under AM1.5G illumination.

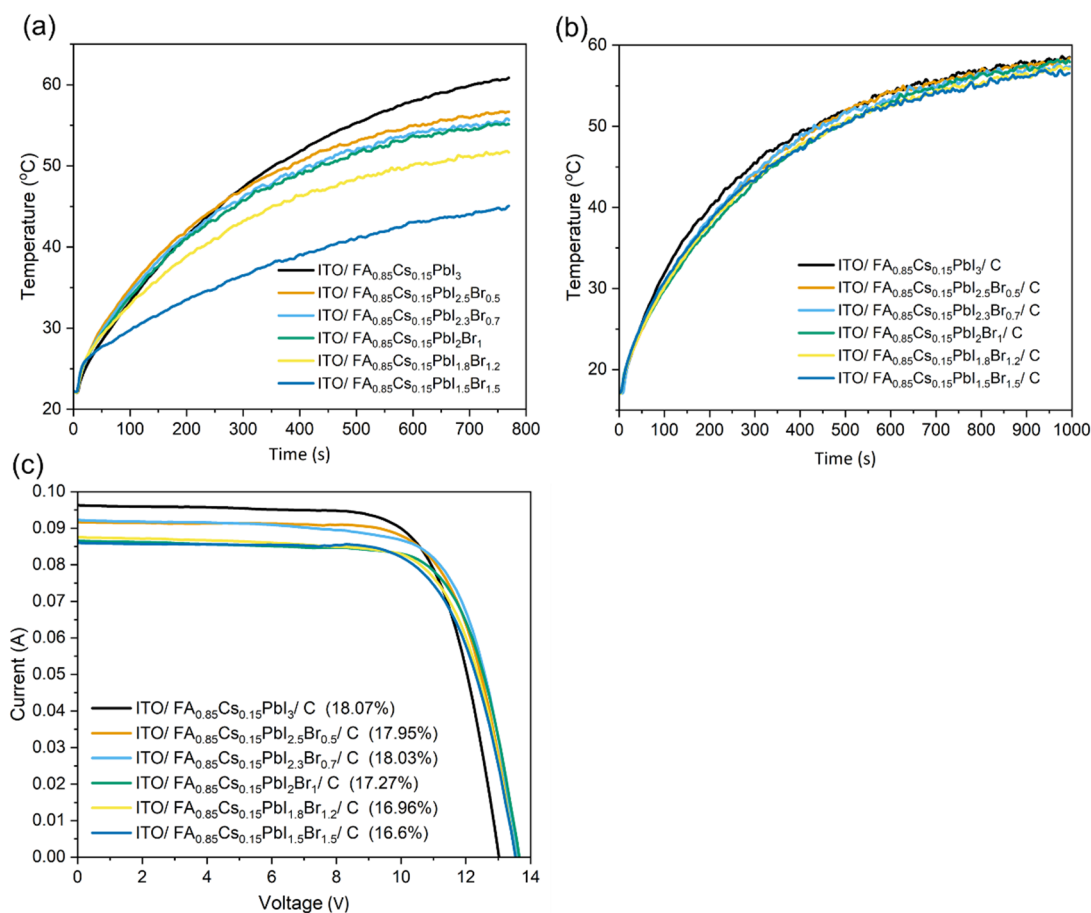


Figure S16. Temperature rise profiles of (a) individual perovskite layers (at ambient temperature of 22 °C) and (b) fully integrated C-PSCs (at ambient temperature of 17 °C), along with the corresponding current-voltage (I–V) characteristics of (c) fully integrated C-PSCs.

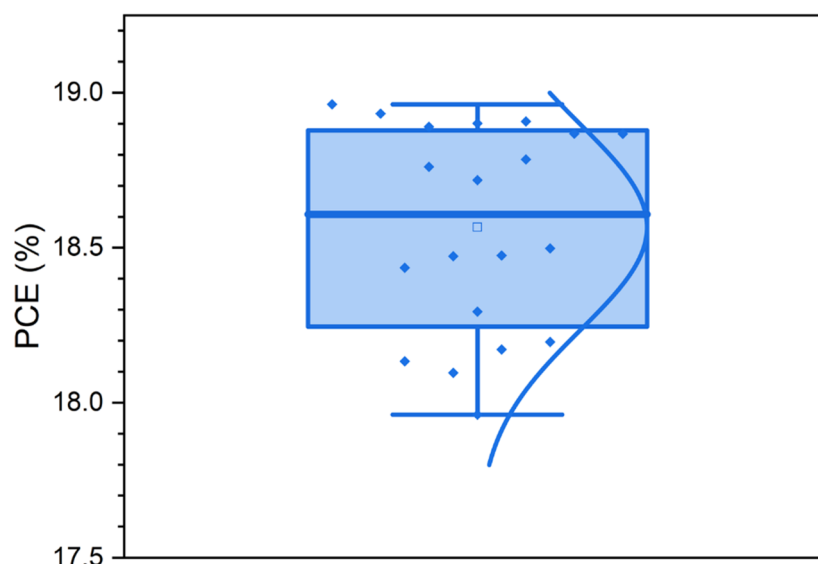


Figure S17. Power conversion efficiency (PCE) statistics of the 20 C-PSCs modules.

Table S1. Comparison between our results and state-of-the-art solar water splitting systems that have been reported recently.

PV-WS system	Active area	PCE	STH	STH/PCE	Reference
Si SC	4.9 cm ²	12.3%	7.5%	61.0%	<i>Nature Mater.</i> 2017 , <i>16</i> , 646–651 [28]
Si SC	4.73 cm ²	24.3%	16.9%	69.54%	<i>Adv. Sci.</i> 2019 , <i>6</i> , 1900465 [29]
PSC	0.236 cm ²	19.4%	13.8%	71.1%	<i>ACS Sustain. Chem. Eng.</i> 2021 , <i>9</i> , 14070–14078 [30]
PSC	7.68 cm ²	23.45%	9.8%	41.89%	<i>Nature Energy</i> 2024 , <i>9</i> , 272–284 [31]
PSC	0.07 cm ²	15.54%	10.64%	68.46%	<i>Carbon Energy</i> 2023 , <i>5</i> , e232 [32]
PSC	0.172 cm ²	18.23%	10.81%	59.29%	<i>Adv. Sci.</i> 2022 , <i>9</i> , 2104938 [33]
PSC	0.2 cm ²	20.34%	13.75%	67.6%	<i>J Colloid Interface Sci.</i> 2025 , <i>677</i> , 599–609 [34]
PSC	0.4 cm ²	12.90%	6.15%	47.67%	<i>Nano Energy</i> 2019 , <i>61</i> , 559–566 [35]
PSC	0.09 cm ² × 2	13%	6.7%	51.5%	<i>ACS Nano</i> 2020 , <i>14</i> , 5426–5434 [36]
PSC	0.5 cm ² × 12	18.7%	8.54%	45.66%	<i>Adv. Funct. Mater.</i> 2021 , 2008245 [37]
PSC	0.25 cm ² × 2	19.8%	12.67%	63.99%	<i>ChemsusChem</i> 2022 , 202102471 [38]
PSC	0.06 cm ² × 2	18.33%	11.40%	62.19%	<i>Small</i> 2023 , <i>19</i> , 2300174 [39]
PSC/Si	0.5 cm ²	27.42%	20.61%	75.1%	<i>ACS Nano</i> 2023 , <i>17</i> , 4539–4550 [40]
2-junction PSC	1 cm ²	26.2%	17%	64.8%	<i>ACS Energy Lett.</i> 2023 , <i>8</i> , 2611–2619 [41]
2-junction PSC	0.44 cm ²	29%	20.8%	71.72%	<i>Nat Commun.</i> 2023 , <i>14</i> , 3797 [42]
2-junction PSC	1 cm ²	26%	17.8%	64.46%	<i>Nat Commun.</i> 2025 , <i>16</i> , 174 [43]
3-junction PSC ¹	0.316 cm ²	46%	32%	69.5%	<i>Nat Commun.</i> 2016 , <i>7</i> , 13237. [44]
3-junction SC	0.5 cm ²	20.3%	13.3%	65.52%	<i>Adv. Mater. Technol.</i> 2020 , <i>5</i> , 2000592 [45]
3-junction SC	0.25 cm ² × 3	30%	20.7%	69%	<i>Adv. Energy Mater.</i> 2023 , <i>13</i> , 2301793 [46]
3-junction SC	1 cm ²	16.85%	10.9%	64.69%	<i>Adv. Energy. Mater.</i> 2017 , <i>7</i> , 1602791 [47]
PSC	50 cm ²	18.3%	12.98%	71.0%	This work

¹ 42-suns concentrator system.