

## Supporting Information

### **Low-temperature suitability of flexible photo-rechargeable devices integrated with hydrogel-based lithium-ion battery and perovskite solar cell**

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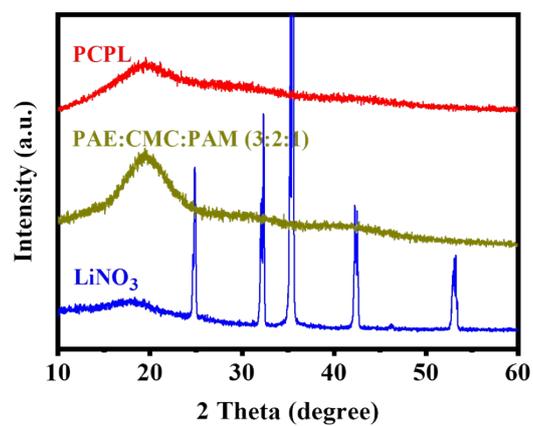
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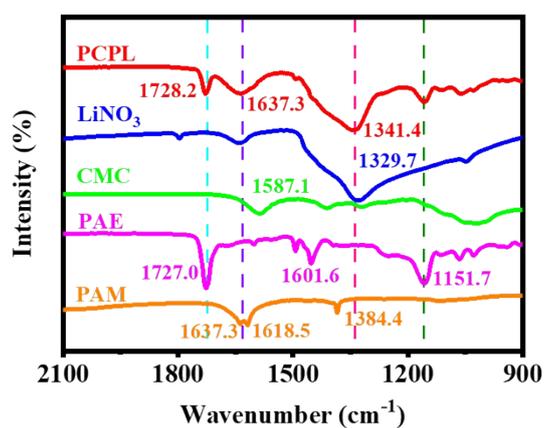
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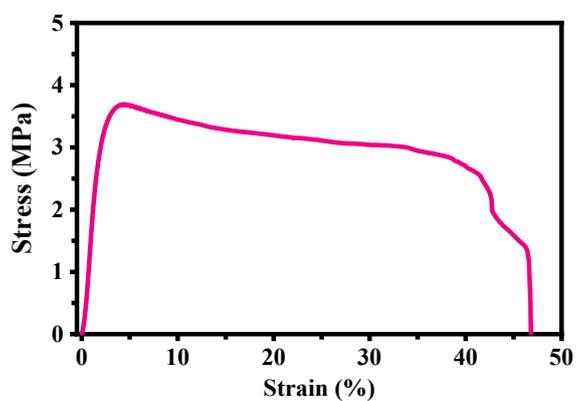
**Fig. S1** XRD spectra of PCPL film, PAE:CMC:PAM (3:2:1) film, and LiNO<sub>3</sub> powders.



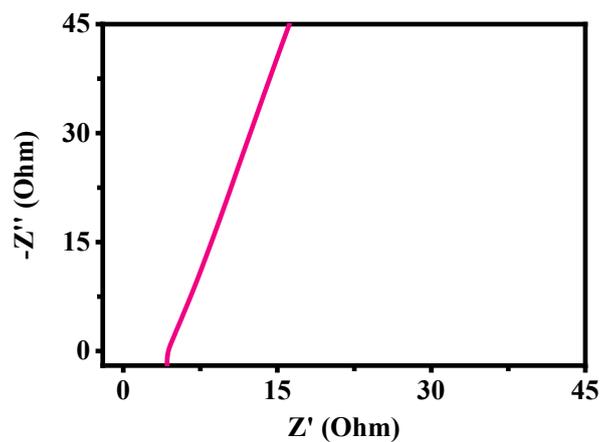
**Fig. S2** FTIR of PCPL film, LiNO<sub>3</sub> powders, CMC powders, PAE film, and PAM powders.



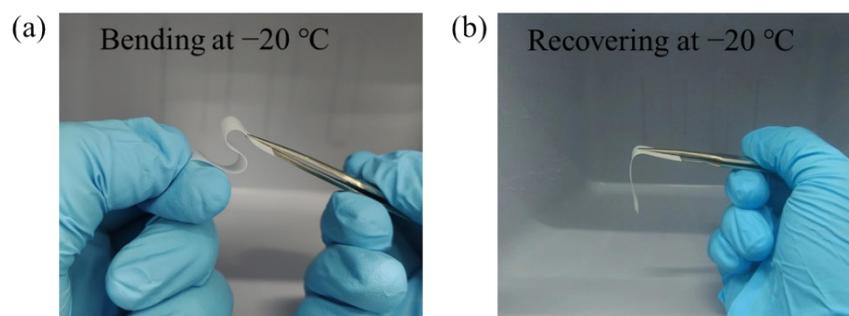
**Fig. S3** Stress-strain curve of PCPL film.



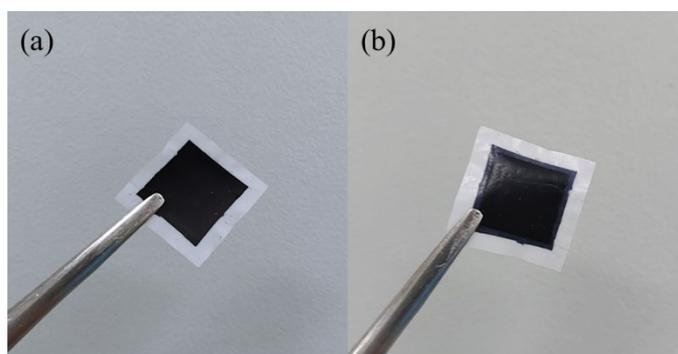
**Fig. S4** EIS curve of PCPL film and the calculated ionic conductivity was obtained at  $-20\text{ }^{\circ}\text{C}$ .



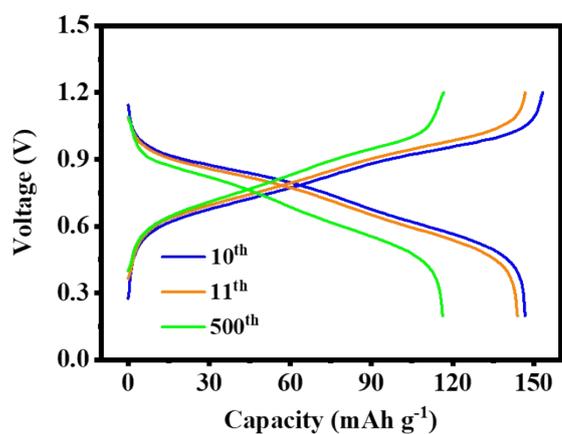
**Fig. S5** The bending property of the hydrogel-based electrolyte at  $-20\text{ }^{\circ}\text{C}$ .



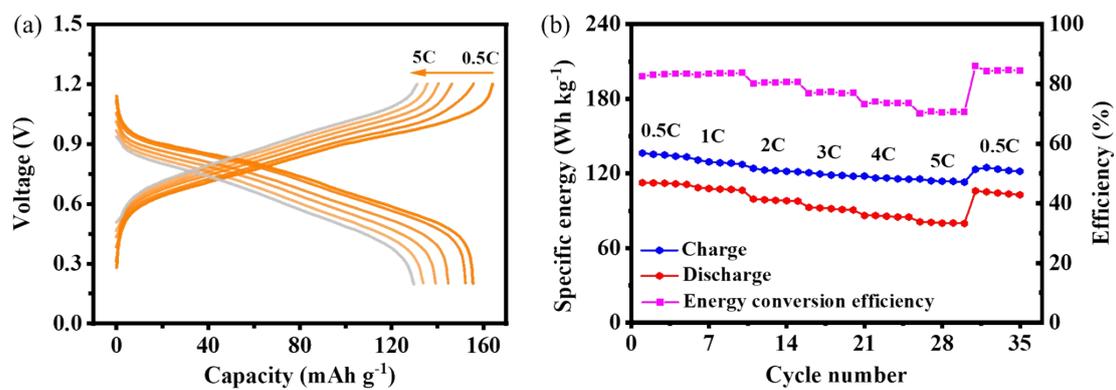
**Fig. S6** Photography of HE-LIB (a) top view is anode side; (b) top view is cathode side.



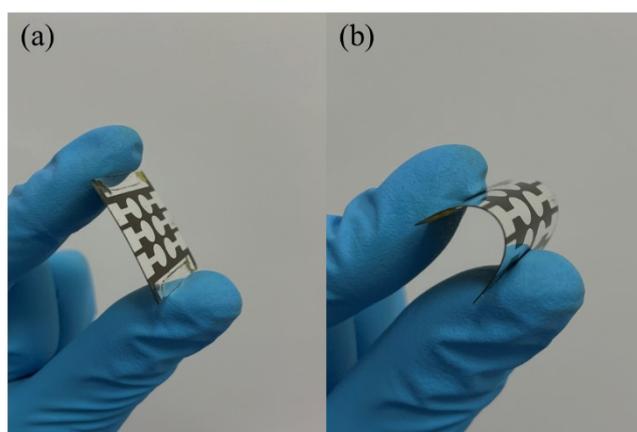
**Fig. S7** Corresponding voltage–capacity profiles at selected cycles in Fig. 2d.



**Fig. S8** (a) Corresponding voltage–capacity profiles at current densities ranging from 0.5C to 5C in Fig. 2e; (b) Corresponding energy density and ESE (as calculated by the weight of LiFePO<sub>4</sub>) at various C-rates.



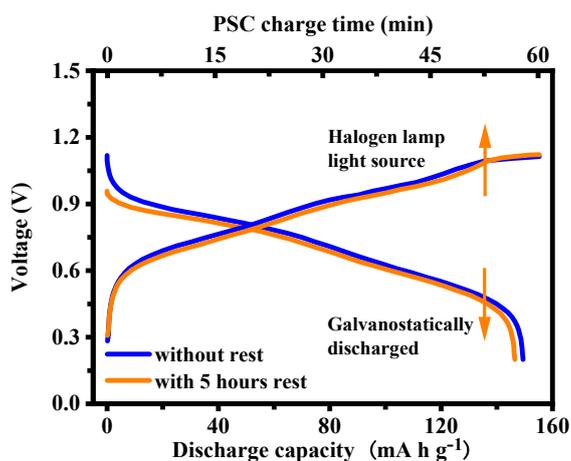
**Fig. S9** Photography of *f*-PSC with area 0.12 cm<sup>2</sup> (a) flat state; (b) bend state.



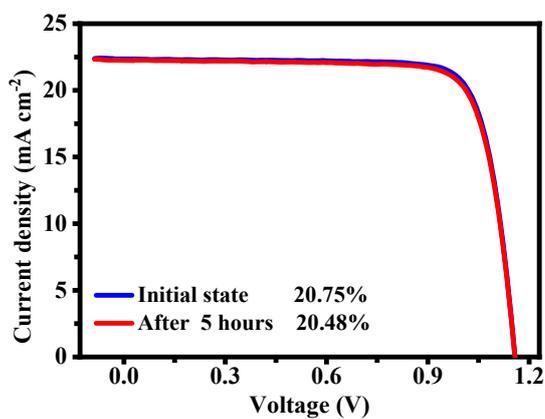
**Table S1** Comparison of  $\eta_{\text{OEE}}$  with a recently reported photo-rechargeable device.

|   | <b>OEE</b> | <b>PCE</b> | <b>Energy storage device</b>      | <b>Photovoltaic cells</b>       | <b>Ref.</b> |
|---|------------|------------|-----------------------------------|---------------------------------|-------------|
| 1 | 5.28%      | 16.6%      | Zn-MnO <sub>2</sub> micro-battery | Flexible perovskite solar cells | [1]         |
| 2 | 11.2%      | 25.0%      | All-solid-state Li-S battery      | Perovskite solar cells          | [2]         |
| 3 | 12.04%     | 18.5%      | Aluminum-ion battery              | Perovskite solar cells          | [3]         |
| 4 | 5.14%      | 15.9%      | Non-aqueous Li-S battery          | Perovskite solar cells          | [4]         |
| 5 | 9.3%       | 16.8%      | Aqueous Li/Na-ion battery         | Perovskite solar cells          | [5]         |
| 6 | 6.4%       | 14.85%     | Rechargeable zinc battery         | Perovskite solar cells          | [6]         |
| 7 | 12.88%     | 21.48%     | Quasi-solid-state Li-ion battery  | Flexible perovskite solar cells | This work   |

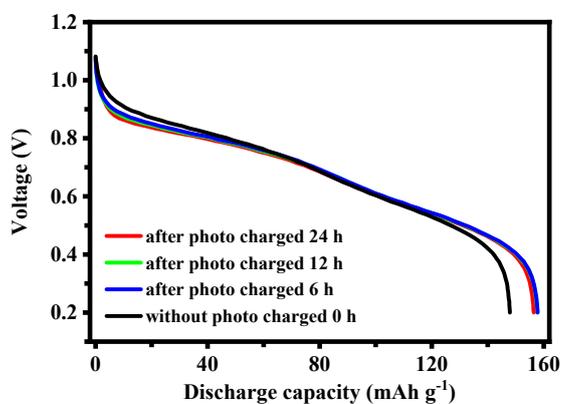
**Fig. S10** The corresponding voltage–capacity profiles of HE-LIB in Fig. 4b.



**Fig. S11** Effect of the photo-rechargeable device without an antireverse charging circuit.



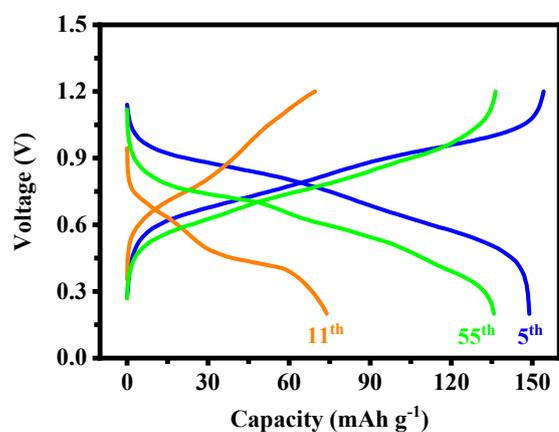
**Fig. S12** The corresponding galvanostatically discharge profile of the HE-LIB unit in photo-rechargeable device under light condition at 0, 6, 12, and 24 hours.



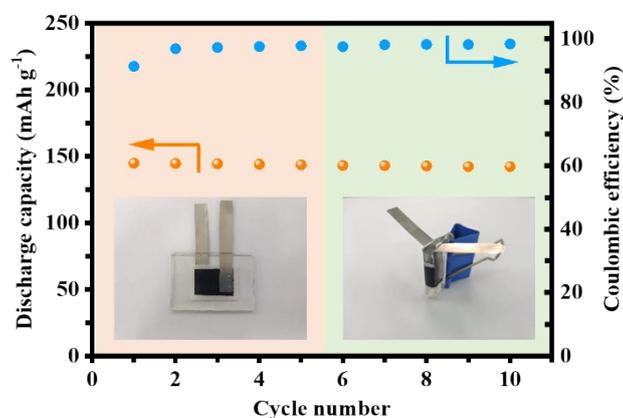
**Table S2** The corresponding photovoltaic parameters of encapsulated *f*-PSC in Fig. 4c.

|               | PCE (%) | $V_{OC}$ (%) | $J_{SC}$ ( $\text{mA cm}^{-2}$ ) | FF(%) |
|---------------|---------|--------------|----------------------------------|-------|
| Without rest  | 20.75   | 1.15         | 22.35                            | 80.92 |
| With 5 h rest | 20.48   | 1.15         | 22.27                            | 80.15 |

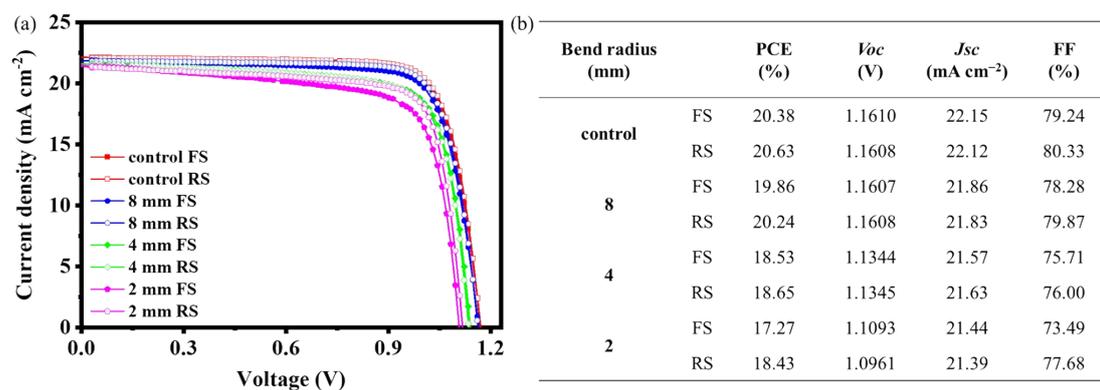
**Fig. S13** Corresponding voltage–capacity profiles at 5<sup>th</sup> (25 °C), 11<sup>th</sup> (–20 °C), and 55<sup>th</sup> (–20 °C) in Fig. 4f.



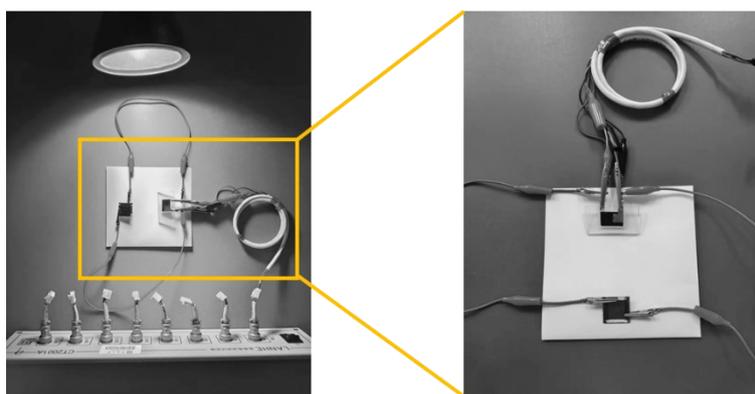
**Fig. S14** Cyclic stability of flexible HE-LIB at initial state and bending state (with a curvature radius of  $\sim 1$  mm).



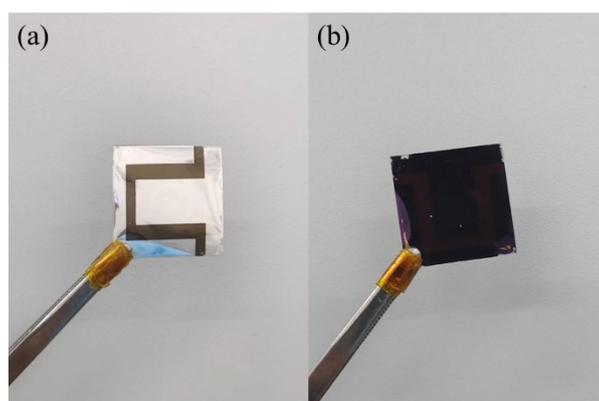
**Fig. S15**  $J$ - $V$  curves and photovoltaic parameters of  $f$ -PSC under different bending states.



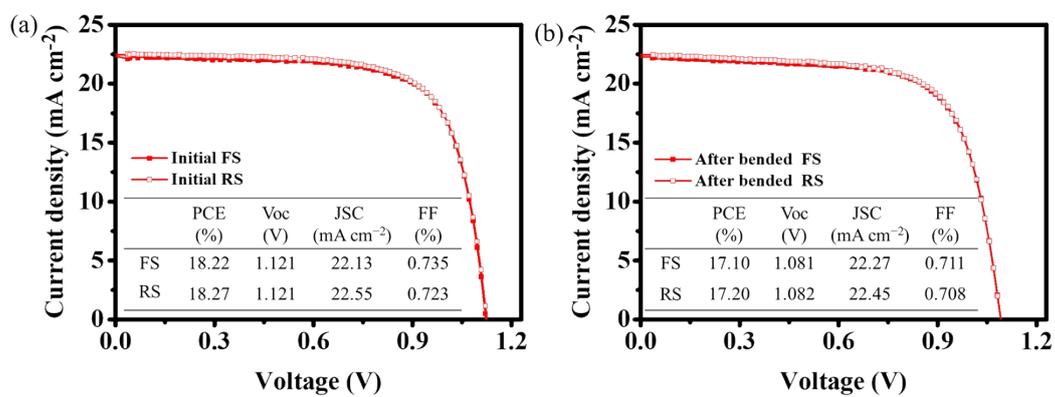
**Fig. S16** Photography of photo-rechargeable device integrated by flexible HE-LIB unit and  $f$ -PSC unit.



**Fig. S17** Photography of  $f$ -PSC with an area of 1.00 cm<sup>2</sup>.



**Fig. S18**  $J$ - $V$  curves and photovoltaic parameters of  $f$ -PSC with an area of  $1.00 \text{ cm}^2$  under (a) initial state and (b) after 10 bending cycles state (bend radius is 4 mm).



## Reference:

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- [3] Y. Hu, Y. Bai, B. Luo, S. Wang, H. Hu, P. Chen, M. Lyu, J. Shapter, A. Rowan, L. Wang, A portable and efficient solar-rechargeable battery with ultrafast photo-charge/discharge rate, *Advanced Energy Materials*, 9 (2019) 1900872.
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