

<Supporting Information>

Highly efficient self-power pack system integrating supercapacitor and photovoltaics with area-saving monolithic architecture

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This PDF file includes:

- A. Comparison of adhesion between silver epoxy and silver paste
- B. Effect of silver epoxy and silver paste on the underlying film
- C. Energy level diagrams of solar devices
- D. Effect of solvent on perovskite crystal and performance
- E. Hysteresis-free perovskite solar cell
- F. Fabrication process of PVA-based solid-state supercapacitor
- G. CV measurement of supercapacitor
- H. Photo-charging and galvanostatic-discharging of SPPs with external wires
- I. Specific capacitance in a two-electrode cell

Figs. S1 to S8

Table S1 to S4

A. Comparison of adhesion between silver epoxy and silver paste

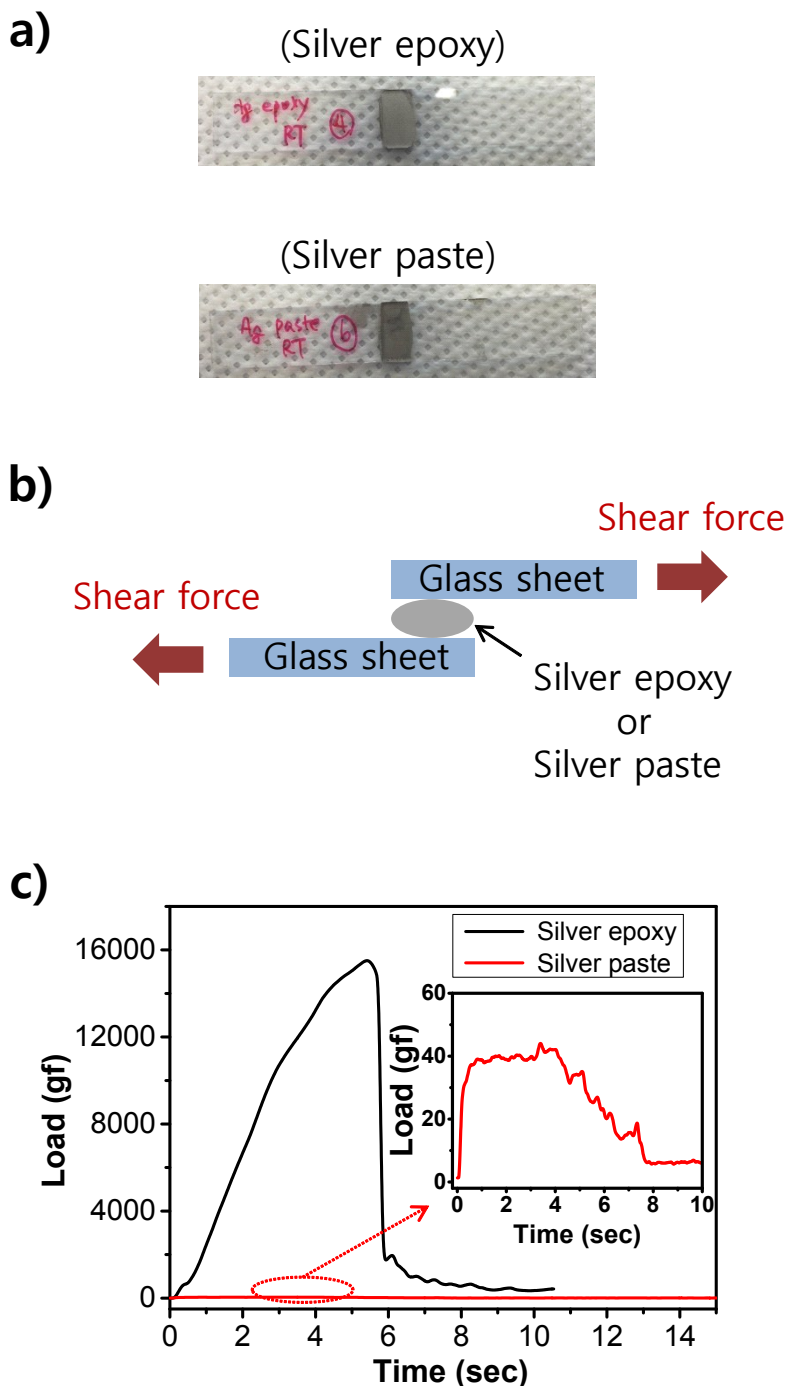


Figure S1. a) Photographs of glass sheets connected by silver epoxy (upper) and silver paste (lower) and b) schematic illustration of shear strength measurement process. c) Load applied to the samples as a function of time. From this result, it is clearly demonstrated that the adhesion of silver epoxy is ~350 times stronger than that of silver paste.

B. Effect of silver epoxy and silver paste on the underlying film

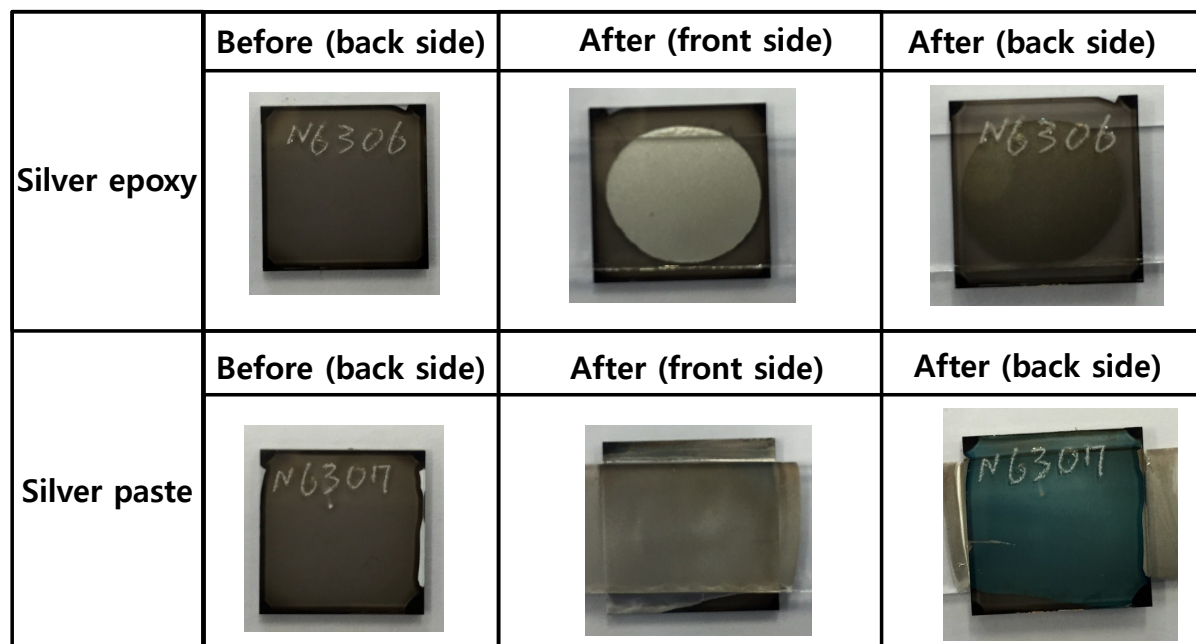


Figure S2. Photographs of PTB-Th-PC71BM films before and after deposition of silver epoxy (upper) and silver paste (lower). After depositing the silver epoxy on top of the BHJ film, it is clearly demonstrated that there is no damage to the BHJ film (upper, right). In contrast, in the case of silver paste, the colour of the BHJ film changed from dark green to bright green, indicating damage to the BHJ film (lower, right).

C. Energy level diagrams of solar devices

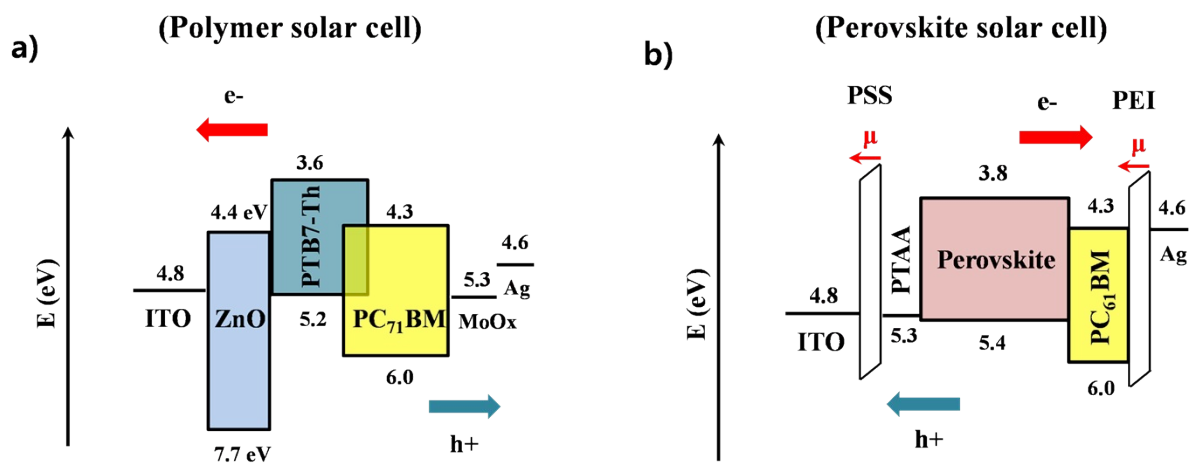


Figure S3. Energy level diagrams of a) polymer solar cell and b) perovskite solar cell.

Table S1. Device performance parameters of PeSCs and PSCs.

Type	Area	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
OPV single	4.64 mm ²	0.79	15.80	0.72	9.00
Perovskite single	4.64 mm ²	1.06	19.44	0.78	16.10
OPV single	100 mm ²	0.80	15.01	0.66	7.85
Perovskite single	100 mm ²	1.04	19.30	0.68	13.66

D. Effect of solvent on perovskite crystal and performance

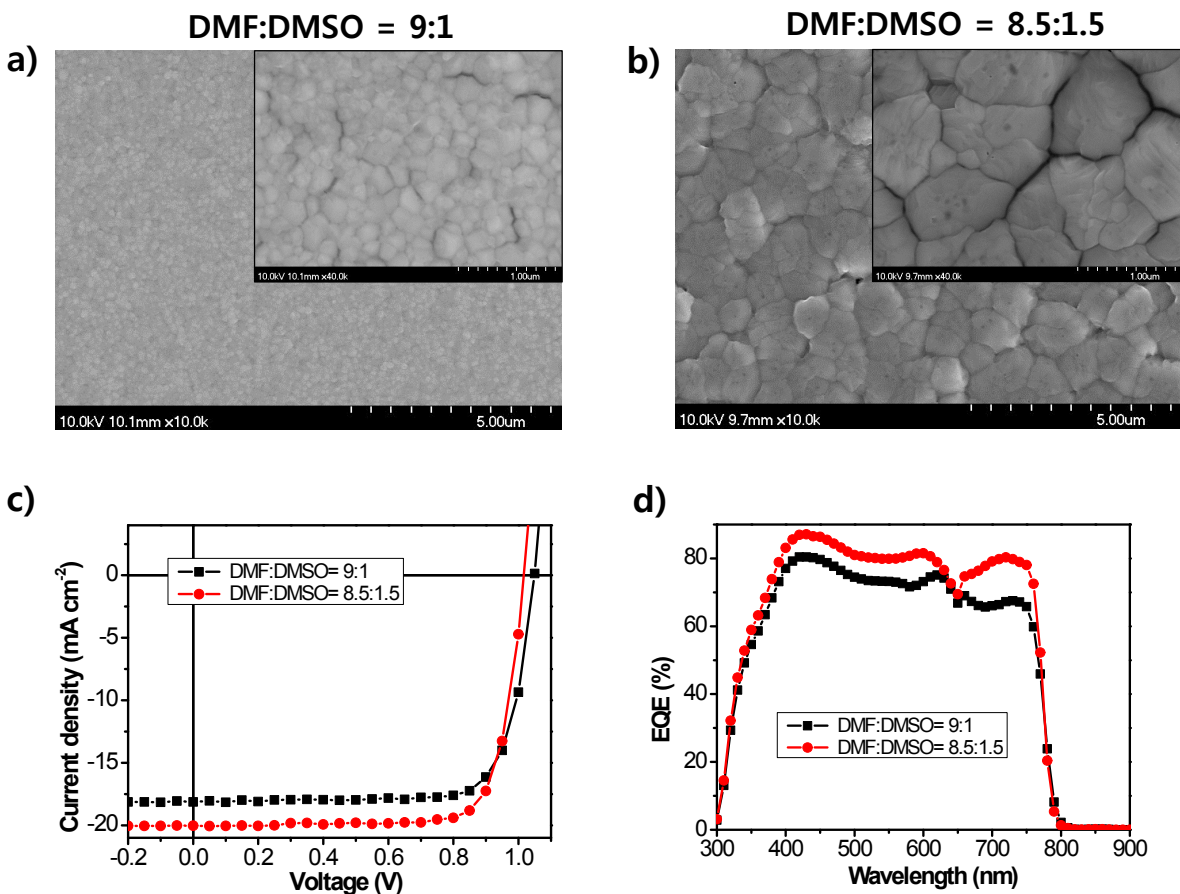


Figure S4. SEM images of (a) perovskite with DMF:DMSO=9:1 and (b) 8.5:1.5 volume ratio. (c) J-V characteristic curves and (d) EQE spectra.

Table S2. Device performance parameters of PeSCs with different solvent ratio.

DMF:DMSO	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
9:1	1.05	18.14	0.77	14.66
8.5:1.5	1.02	20.03	0.79	16.14

E. Hysteresis-free perovskite solar cells

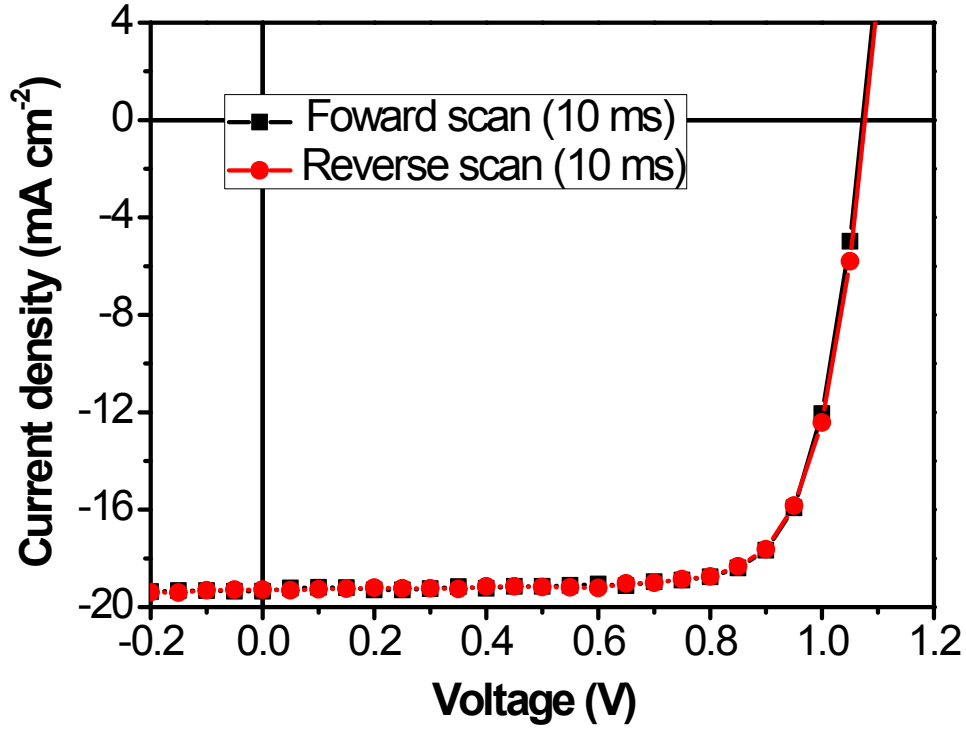


Figure S5. J-V curves of perovskite solar cells with different scan direction.

Table S3. Device performance parameters of PeSCs with different scan direction.

Type	Area	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
Forward	4.64 mm ²	1.07	19.35	0.77	15.90
Reverse	4.64 mm ²	1.08	19.29	0.76	15.86

F. Fabrication process of PVA/H₃PO₄-based solid-state supercapacitor

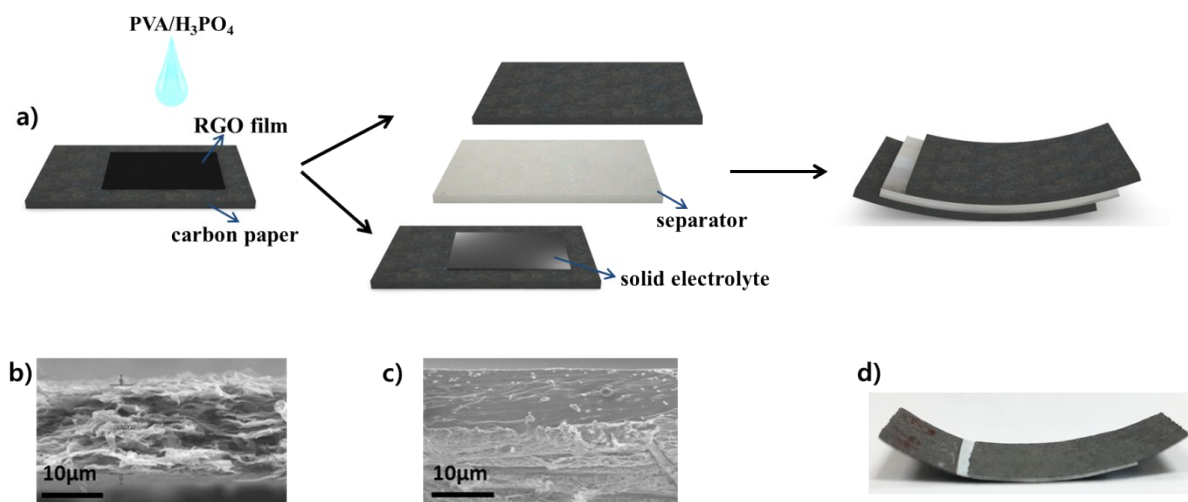


Figure S6. (a) Schematic illustration of the fabrication process of the solid-state electrochemical supercapacitor. FE-SEM images of the cross-sectional RGO electrode (b) without and (c) with PVA/H₃PO₄ solid electrolyte. (d) Photograph of the PVA/H₃PO₄-based supercapacitor.

G. CV measurement of supercapacitor

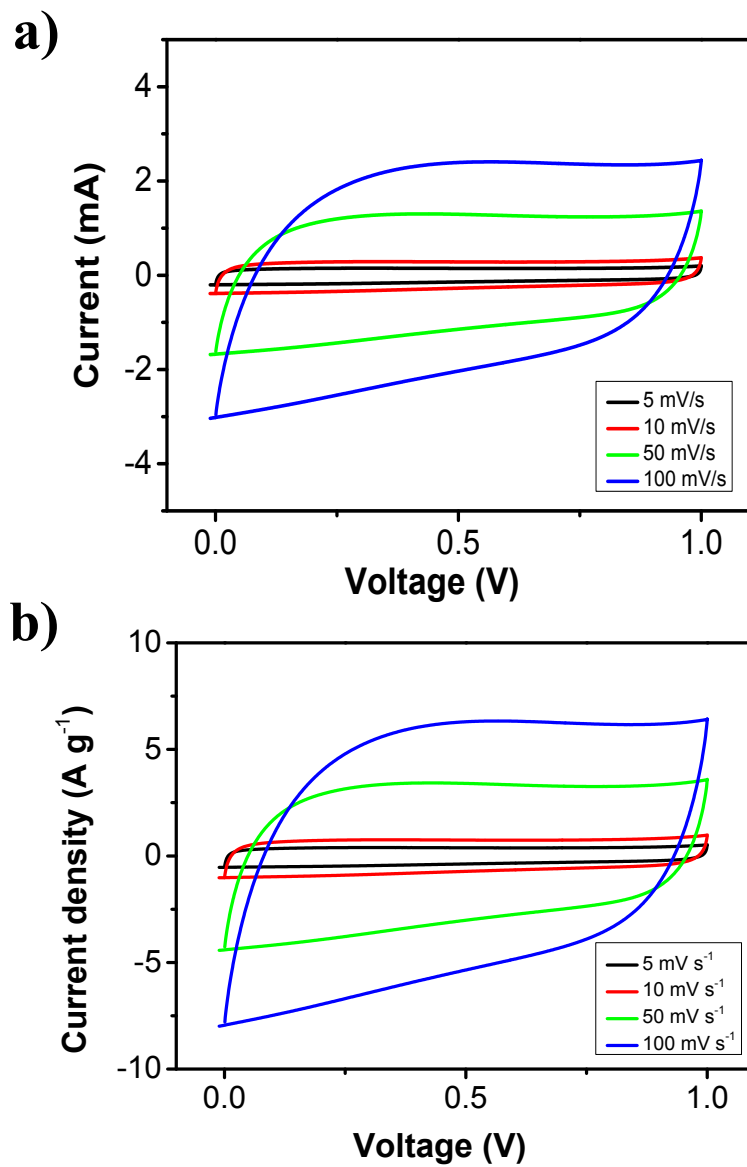


Figure S7. CV curves under various scan rates with (a) the real current and (b) the normalized current density on the y-axis.

H. Photo-charging and galvanostatic-discharging of SPPs with external wires

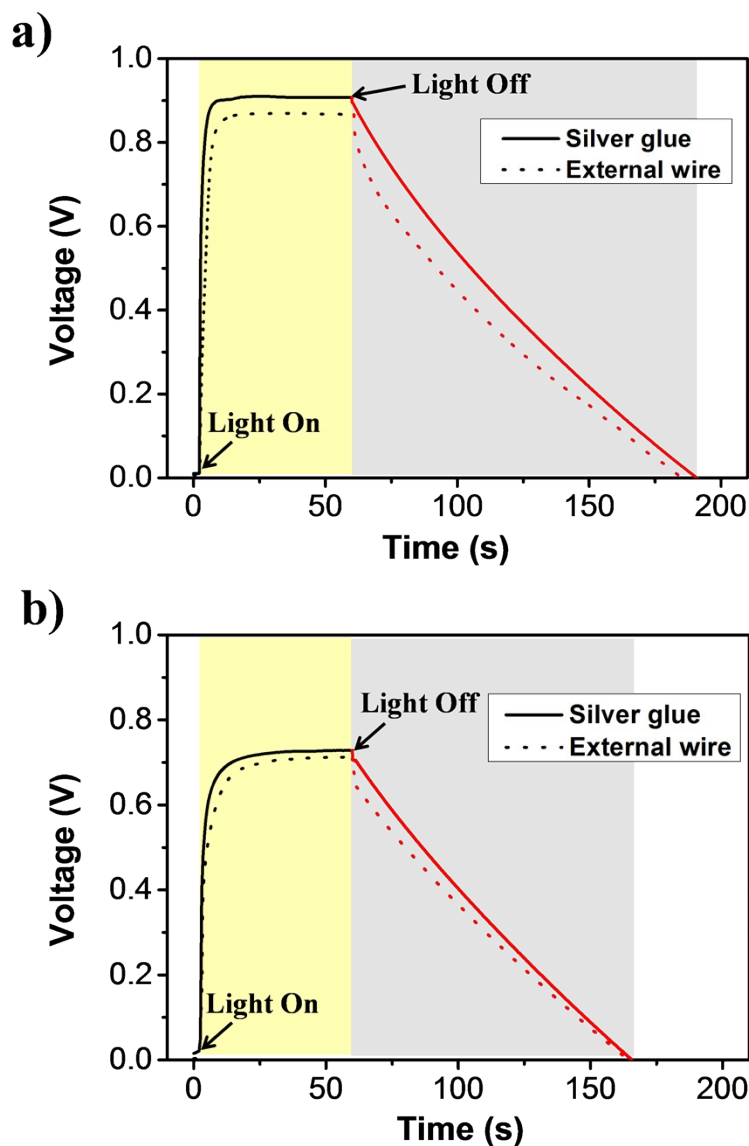
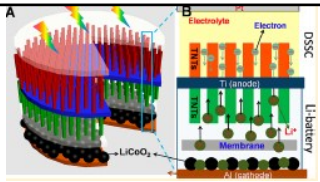
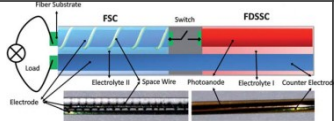
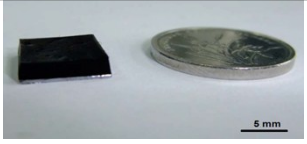
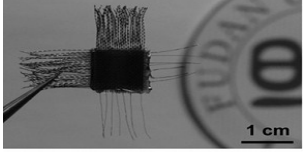
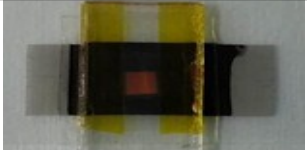
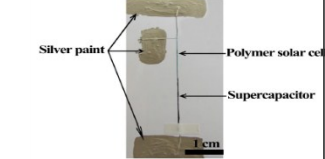


Figure S8. Photo-charging (yellow sections) and galvanostatic-discharging (grey sections) curves of (a) PeSC/EC and (b) PSC/EC devices with a silver glue (full-line) or external wire (dot-line). The galvanostatic-discharging process was carried out at a current density of 0.5 A g^{-1} .

Note that the used devices with external wire systems were not optimized. Therefore, as compared to the monolithically integrated SPPs with a silver glue, the photo-charging curves both of PeSC/EC and PSC/EC with external wire systems showed slightly decreased charging values.

Table S4. Comparison of our work with previous reports on self-power packs.

Integrated devices (PV ^{1*} /EC ^{2*})	Architecture	η_{overall} (%)	η_{storage} (%)	Efficiency of PV (%)	Capacitance	Charging time (s)	Supporting Ref.
DSSC ^{3*} /LIB ^{4*}		0.82	-	-	38.89 μAh	240	1
DSSC/SC ^{5*}		2.1	46	4.56	19 mF cm^{-2}	15	2
DSSC/SC		0.79	34	2.31	83 F g^{-1}	183	3
DSSC/SC		3.67	-	3.2	-	-	4
DSSC/SC		1.83	-	2.66	-	15	5
Polymer/SC		0.82	1.01	65.6	0.08 mF cm^{-1}	-	6

Perovskites/LIB		7.80	-	15.67		-	7
Perovskites/SC		4.3	72.1	7.80	185.2 F g ⁻¹	15	8
Perovskites/SC		10	49	13.6	572 mF cm ⁻¹	300	9
Polymer/SC		5.07	64.59	7.85 (@100 mm ²)	144 F g ⁻¹	15	Our work
Perovskites/SC		10.97	80.31	13.66 (@100 mm ²)	142 F g ⁻¹	8	

* PV¹: Photovoltaic device: EC²: Energy storage device: DSSC³: Dye-sensitized solar cells: LIB⁴: Lithium ion battery: SC⁵: Supercapacitor:

I. Specific capacitance in a two-electrode cell

The simplest electrochemical capacitor is consisted of two equivalent electrodes as a symmetric system that has the same weight, the same capacitance and where electric energy is stored through the electrical double layer (EDLC). Therefore the capacitance in symmetric system can be expressed as following: ¹⁰

$$C_{cell} = \int (I \times dt / \Delta V) = I \times \Delta t / \Delta V \quad (1)$$

where C_{cell} is the cell capacitance, I is the current intensity, t is the time and ΔV is the voltage interval of the cell.

Specific capacitance is the capacitance per unit total mass of active materials in two electrodes (m_{tot}) (equ (2)).

$$C_{spec.cell} = I \times \Delta t / m_{tot} \times \Delta V \quad (2)$$

The cell capacitance value (C_{cell}) of an EDLC is considered as single capacitors of capacitance C_{anode} and $C_{cathode}$ for the anode and cathode, respectively. ^{11, 12}

$$1/C_{cell} = 1/C_{anode} + 1/C_{cathode} \quad (3)$$

$$C_{anode} = C_{cathode} = C_{elec} \quad (4)$$

$$C_{cell} = 1/2 C_{elec} \quad (5)$$

$$C_{spec.cell} = C_{cell} / m_{tot} = (C_{ele}/2) \times (1/2 \times m_{ele}) = C_{spec.ele} / 4 \quad (6)$$

Substitution into equation (2) yields

$$C_{spec.elec} = 4 \times I \times \Delta t / m_{tot} \times \Delta V \quad (7)$$

$$C_{spec.elec} = \frac{2 \times I \times \Delta t}{m_{elec} \times \Delta V} \quad (8)$$

The specific capacitance of $C_{spec.elec}$ is obtained from equation (7) and (8) with m_{tot} and m_{elec} , respectively.¹³ Moreover, equation (8) is the most widely used function to calculate the specific capacitance of single electrode in a two-electrode cell per total mass of active materials of single electrode (m_{elec}). In summary, in the symmetric system, all the equations come from the definition of a two-electrode supercapacitor as two capacitors connected in series.

Supporting References

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