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Supplementary Information

A photocapacitor based on organometal halide perovskite and PANI/CNT composites integrated using CNT bridge

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1 Details of experiment

1.1 Preparation of PMMA/SACNT-PANI@acSACNT/PVA (PSP) electrode material

30 mg of SACNT was heated at 500 °C for 30 min in air to get active-SACNT (acSACNT). PANI@acSACNT paper (Figure 1f) was synthesized using a previously described in-situ polymerization method [1]. In brief, 30 mL of acSACNT/ethanol solution (1 mg mL^{-1}) was ultrasonically mixed, and was filtered to get acSACNT paper. Then, the paper was soaked in 40 mL of aniline/HCl aqueous solution (1 M HCl, 0.05 M aniline) at 0 °C for 10 min. Subsequently, 40 ml of 0.05 M $(NH_4)_2S_2O_8$ aqueous solution was added into the above solution slowly. After that, the mixture was kept at 0 °C for 24 h. This step involves the so-called in-situ chemical reaction and generated PANI covered the surface of acSACNT in the paper well. The PANI@acSACNT paper was cleaned with deionized water, ethanol and acetone, dried at 80 °C for 24 h. 15 mL of SACNT/ethanol solution (1 mg mL^{-1}) was filtered through the above PANI@acSACNT paper, dried at 80 °C for 24 h to get SACNT-PANI@acSACNT paper. Conveniently, the side of the paper covered by SACNT layer was marked as S-side, and the other side was marked as P-side. 100 µL of PVA/H_2SO_4 aqueous solution (10 mg mL^{-1} PVA, 0.1 M H_2SO_4) was coated onto the P-side, dried at room temperature for 2 h. Later, the paper was soaked in PMMA/toluene solution (10 mg mL^{-1}) for 30 min, dried at room temperature for 5h. After that, the PMMA on the P-side was removed by acetone.

1.2 Device fabrication

FTO glass was ultrasonically cleaned in detergent, deionized water, acetone and isopropanol (IPA) in sequence, dried with compressed nitrogen. After that, a compact titanium dioxide (TiO_2) block layer was deposited by spin-coating 90 μL of acidic titanium isopropoxide/ethanol solution (70 μL of titanium isopropoxide, 7 μL of 2 M HCl, 1 mL of ethanol) onto the FTO substrate at a speed of 2000 rpm for 50 s. The substrate was dried at 150 ^{o}C for 30 min and then annealed at 500 ^{o}C for 30 min. A PbI_2 film was deposited by spin-coating 90 μL of PbI_2/ N,N-Dimethylformamide solution (500 mg mL^{-1}) at a speed of 2000 rpm for 50 s, and then a MWCNT film was deposited by drop-casting 120 μL of MWCNT/chlorobenzene solution (10 mg mL^{-1}). To get the perovskite of $MAPbI_3$, 120 μL of CH_3NH_3I/IPA solution (10 mg mL^{-1}) was dropped onto the above substrate for 3 min, dried by spinning at 2000 rpm for 50 s and heated at 100 ^{o}C for 30 min. After the substrate was cooled to room temperature, a PMMA film was deposited onto the substrate by spin-coating 0.5 mL of PMMA/toluene solution (100 mg mL^{-1}) at a speed of 3000 rpm for 50 s, dried at room temperature. The PSC part was prepared.

After fabricating the PSC part, a PSP electrode was placed onto the PSC substrate with the S-side of the PSP close to the PMMA film of the PSC part. Then, 100 μL of chlorobenzene was dropped onto the P-side of the PSP to dissolve and reshape the PMMA film between the PSC part and the PSP electrode. When the substrate was dry, with PVA electrolyte (100 mg mL^{-1} PVA, 1 M H_2SO_4), another PSP electrode was fabricated onto the substrate and formed the SC part. Finally, a PMMA film was coated onto the substrate to complete the photocapacitor. Moreover, a schematic showing the construction process of the photocapacitor was included in Figure S1.

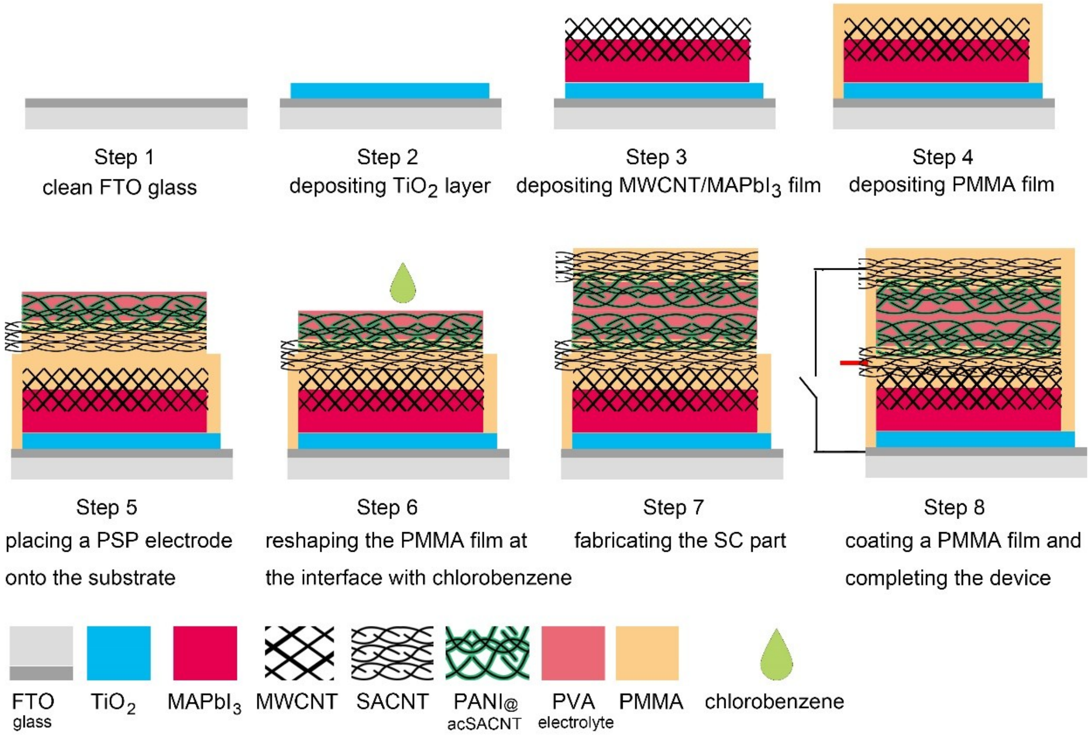

Figure S1 Schematic of the photocapacitor construction process

2 PSP electrode material and the photocapacitor

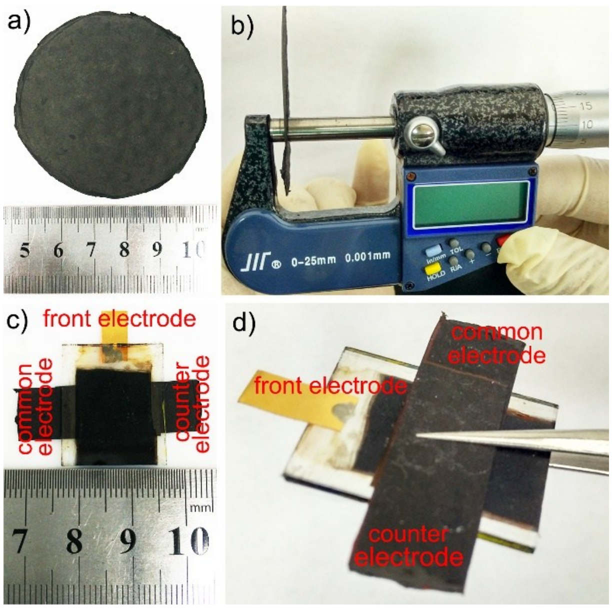

Figure S2 Photos of PSP electrode material and the photocapacitor

3 Schematic circuit and SEM of the common electrode inside the photocapacitor

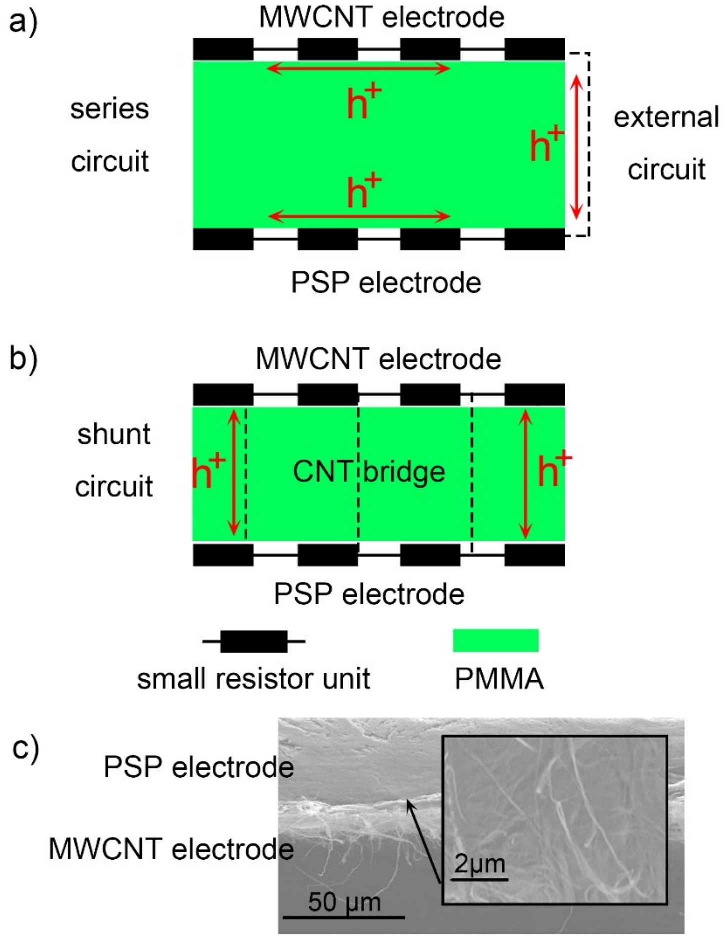

Figure S3 Schematic circuit and cross-section SEM of the common electrode

4 Equivalent circuit of solar cells

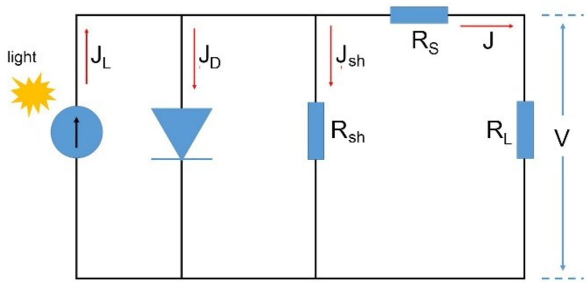

Figure S4 Equivalent circuit of solar cells

5 Stability of the PSC part

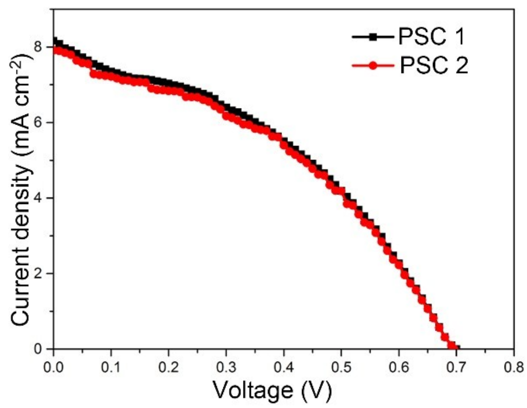

Figure S5 J-V curves of PSC 1 and PSC 2, one week after fabrication

6 Self discharge of the photocapacitor in dark

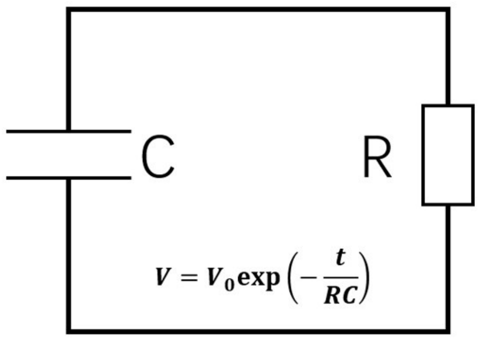

Figure S6 Equivalent RC circuit and voltage equation during self-discharge of the photocapacitor in dark

7 Equations

✧ **Equivalent equation of solar cell.** J_o is the reverse saturation current. q is the elementary charge. n is the density of charge carriers. k is the Boltzman constant. T is the absolute temperature. R_S and R_{sh} are equivalent serial resistance and shunt resistance inside solar cells respectively. R_L is external load, and J and V are corresponding current and voltage with R_L.

$$J = J_L - J_D - J_{sh} = J_L - J_O\left(e^{\frac{q(V+JR_S)}{nkT}} - 1\right) - \frac{JR_S + V}{R_{sh}} \quad \text{(S1)}$$

✧ **Charging (or discharging) process of the SC part.** V_T is the ending voltage (or the starting voltage). JR is the JR drop. ΔV is the corresponding voltage difference. Q is the capacity. A is the area of the SC part. Q_A is the capacity density. E is the energy stored (or energy consumed), and E_A is the energy density. C is the charging capacitance (or discharging capacitance), and C_A is the specific area capacitance, while C_M is the specific mass capacitance and M is the mass of two PSP electrodes of the SC part. Q_{discha} is the quantity of electric charge during discharging, Q_{cha} is the quantity during charging, and η_{coul} is the Coulombic efficiency.

$$\Delta V = V_T - JR \quad \text{(S2)}$$

$$dQ = Jdt \quad \text{(S3)}$$

$$Q = \int Jdt \quad \text{(S4)}$$

$$Q_A = \frac{Q}{A} \quad \text{(S5)}$$

$$dE = VJdt \quad \text{(S6)}$$

$$E = \int VJdt \quad \text{(S7)}$$

$$E_A = \frac{E}{A} \quad \text{(S8)}$$

$$C = \frac{Q}{\Delta V} \quad \text{(S9)}$$

$$C_A = \frac{C}{A} \quad \text{(S10)}$$

$$C_M = \frac{C}{M} \quad \text{(S11)}$$

$$\eta_{coul} = \frac{Q_{discha}}{Q_{cha}} \quad \text{(S12)}$$

✧ **Fill factor (FF) and power conversion efficiency (PCE) of the PSC part.** P_{in} is input power density during photo charging. In this study, P_{in} is 100 mW cm^{-2} from AM 1.5G solar light simulator. PCE is the power conversion efficiency. P_M is the largest output power density (V_M and J_M are the corresponding voltage and current density respectively).

$$FF = \frac{P_M}{V_{OC}J_{SC}} = \frac{V_M J_M}{V_{OC}J_{SC}} \quad \text{(S13)}$$

$$PCE = \frac{V_{OC}J_{SC}FF}{P_{in}} = \frac{P_M}{P_{in}} \quad \text{(S14)}$$

✧ **Energy storage efficiency of the photocapacitor during photo charging-galvanostatic discharging.** E_{discha} is the energy released during galvanostatic discharging. E_{phcha} is the energy stored during photo charging. Δt is the time for photo charging. $\eta_{overall}$ is the whole energy conversion and storage efficiency of the photocapacitor during photo charging-galvanostatic discharging. $\eta_{storage}$ is the energy storage efficiency during galvanostatic discharging, and η_{phcha} is the energy conversion efficiency during photo charging.

$$\eta_{overall} = \frac{E_{discha}}{P_{in}\Delta t} \quad \text{(S15)}$$

$$\eta_{storage} = \frac{E_{discha}}{E_{phcha}} \quad \text{(S16)}$$

$$\eta_{phcha} = \frac{\eta_{overall}}{\eta_{storage}} \quad \text{(S17)}$$

✧ **Degradation of $CH_3NH_3PbI_3$ ($MAPbI_3$)**

$$CH_3NH_3PbI_3(s) \overset{H_2O}{\Leftrightarrow} CH_3NH_3I(aq) + PbI_2(s) \quad \text{(S18)}$$

$$CH_3NH_3I(aq) \Leftrightarrow CH_3NH_2(aq) + HI(aq) \quad \text{(S19)}$$

$$4HI(aq) + O_2 \Leftrightarrow 2I_2(s) + 2H_2O \quad \text{(S20)}$$

$$2HI(aq) \overset{h\upsilon}{\Leftrightarrow} H_2\uparrow + I_2(s) \quad \text{(S21)}$$

8 Comparison with earlier reported photocapacitors

Table S1 Performances of earlier reported photocapacitors and photocapacitor in this work

No. [Ref.]	Photovoltaic part	PCE (%)	Capacitor part	Coulombic efficiency η_{coul} (%)	Energy storage efficiency $\eta_{overall}$ (%)	Specific Capacitance (mF cm^{-2}) / (F g^{-1})
1 [2]	DSSC	--	AC	80	--	690/--
2 [3]	DSSC	--	AC	42	--	650/--
3 [4]	DSSC	4.37	PEDOT	--	--	520/--
4 [5]	DSSC	--	PEDOT &CNT	59	--	--/95
5 [6]	Organic SC	3.39	CNT	--	--	184/--
6 [7]	DSSC	0.50	Plypyrrole	--	20	--/39.3
7 [8]	DSSC	2.31	MWCNT	--	34	--/83
8 [9]	DSSC	--	PEDOT& CNT	7.8	--	610/--
9 [10]	DSSC	4.70	MWCNT	--	--	520/--
10 [11]	DSSC	3.70	PVDF&Z nO	--	--	--/2.7
11 [12]	DSSC	0.80	Metal oxide	88	--	--/407
12 [13]	Titanium	3.17	Titanium	--	52	1.29/--

	oxide SC		oxide			
13 [14]	DSSC	4.90	AC&Nickel oxide	54	--	--/32
14 [15]	DSSC	3.38	PVDF	--	--	--/2.1
15 [16]	Quantum dot SC	3.45	MWCNT	--	--	--/150
16 [17]	DSSC	5.70	Polymer	--	--	--/1.03
17 [18]	Crystalline silicon SC	15.50	Graphene oxide	62	--	$0.2*10^{-3}$/--
18 [19]	CdS-sensitized SC	0.26	CNT	--	--	--/95.25
19 [20]	Perovskite SC	6.37	PEDOT&carbon	--	74	12/--
20 [21]	Perovskite SC	12.54	Metal oxide	--	--	43/--
21 [22]	Perovskite SC	7.79	Carbon&MnO_2	--	76	61.01/--
This work	Perovskite SC	2.55	PANI&CNT	96	70.9	422/103.4

★DSSC, dye-sensitized solar cell ★AC, activated carbon ★PANI, polyaniline

★PEDOT, poly(3,4-ethylenedioxythiophene) ★PVDF, polyvinylidene fluoride

★CNT, carbon nanotube★MWCNT, multi-walled carbon nanotube

9 More details about performances of photocapacitors

Table S2 Performance data of ten photocapacitors in our study under AM 1.5G simulated sunlight of 100 mW cm^{-2}.

V_{OC} (V)	J_{SC} (mA cm^{-2})	FF	PCE (%)	Energy storage efficiency (%)
0.710	9.226	0.389	2.548	70.7
0.705	9.275	0.386	2.524	70.9
0.705	9.058	0.392	2.503	70.6
0.711	9.368	0.375	2.498	70.6
0.703	9.057	0.391	2.490	70.4
0.703	9.053	0.390	2.482	70.8
0.712	9.377	0.371	2.477	70.9
0.690	9.039	0.394	2.457	70.4
0.687	9.048	0.394	2.449	70.5
0.695	9.051	0.382	2.403	70.3

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