

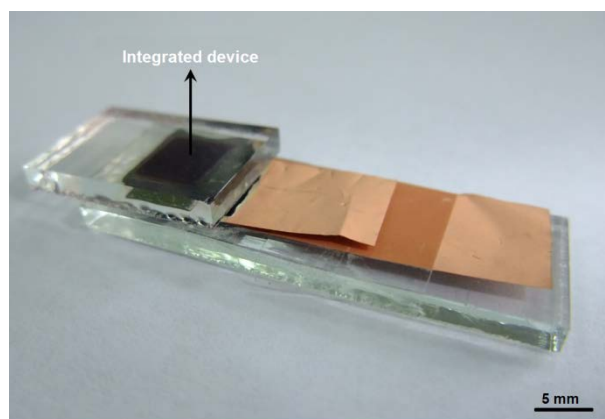
## Supporting information

### Experimental section

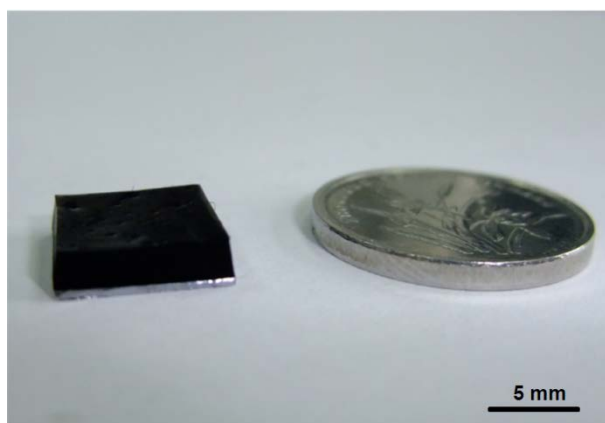
Materials: Fluorine-doped tin oxide conductive glass was obtained from Nippon Sheet Glass Co., Japan. Poly (ethylene naphthalate) was obtained from Peccell Co., Japan. Titanium dioxide slurry (DHS-TPP3 and DHS-TPP200, hydrothermal titanium dioxide dispersed in terpinol) and powder (DHS-SN1760-500) were obtained from Dalian Heptachroma Solartech Co., Ltd.

Calculation of the specific capacitance: The discharge capacitance of each cell was calculated according to the following equation:  $C=2(I*\Delta t)/(m*\Delta V)$ , where  $C$  is the specific capacitance, while  $I$ ,  $\Delta t$ ,  $m$ , and  $\Delta V$  are discharge current, discharge time, electrode weight, and voltage variation in the time range, respectively.<sup>S3</sup>

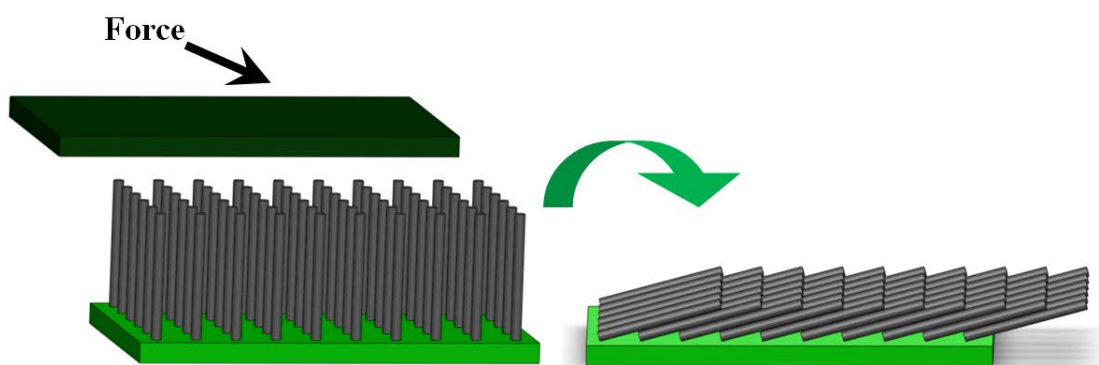
Calculation of the entire energy conversion and storage efficiency of the integrated device: The entire energy conversion and storage efficiency is calculated by the equation of  $\eta_{entire} = \eta\Delta VJS_1t_1/(P_{in}S_2\eta t_2)$ , where  $\eta_{entire}$  is the entire energy conversion and storage efficiency,  $\eta$  is the photoelectric conversion efficiency,  $\Delta V$  and  $t_1$  are the voltage variation and discharging time during galvanostatic discharge, respectively.  $J$  is the constant galvanostatic discharge current density of  $1.4 \text{ mA cm}^{-2}$ ,  $S_1$  is the effective area of energy storage part,  $P_{in}$  is the incident light power density ( $100\text{mW cm}^{-2}$ ),  $S_2$  is the effective area of photoelectric conversion part and  $t_2$  is the illumination time during photocharging.



**Fig. S1.** Photograph of an integrated device with aligned MWCNT films as electrodes.



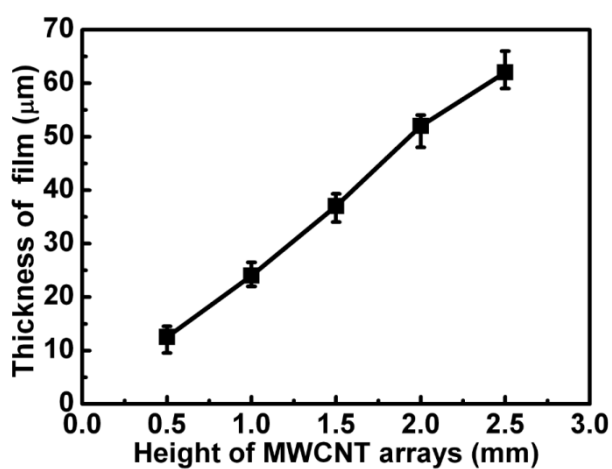
**Fig. S2.** Photograph of a MWCNT array compared with a coin with thickness of about 2 mm.



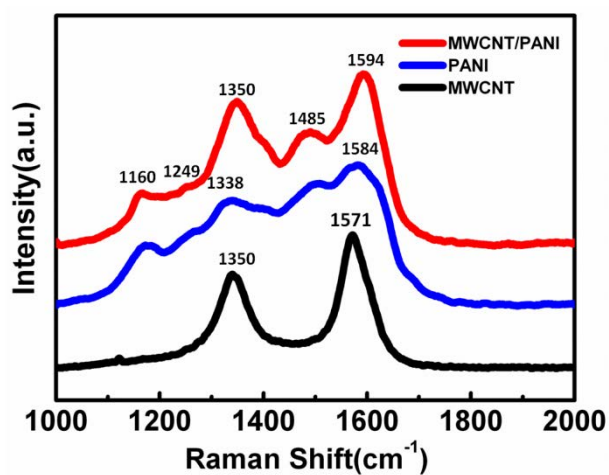
**Fig. S3.** Schematic illustration for the preparation of aligned MWCNT film.



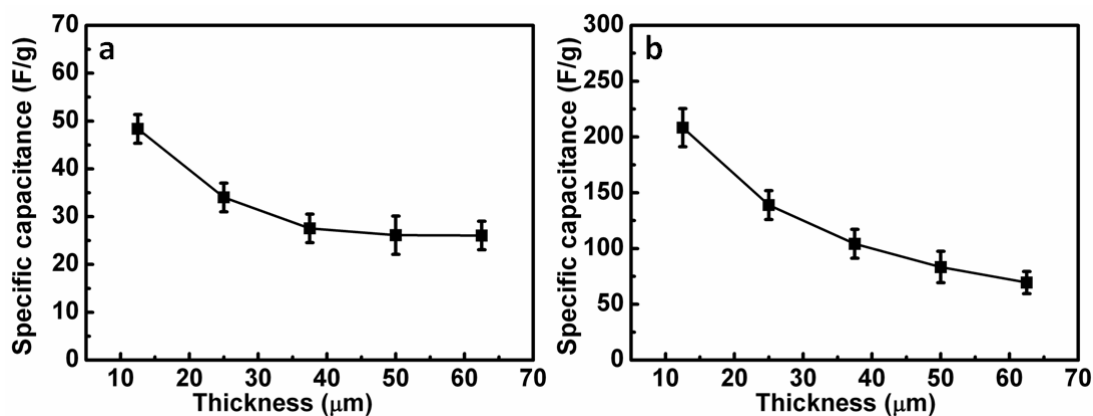
**Fig. S4.** Photograph of a free-standing and aligned MWCNT film.



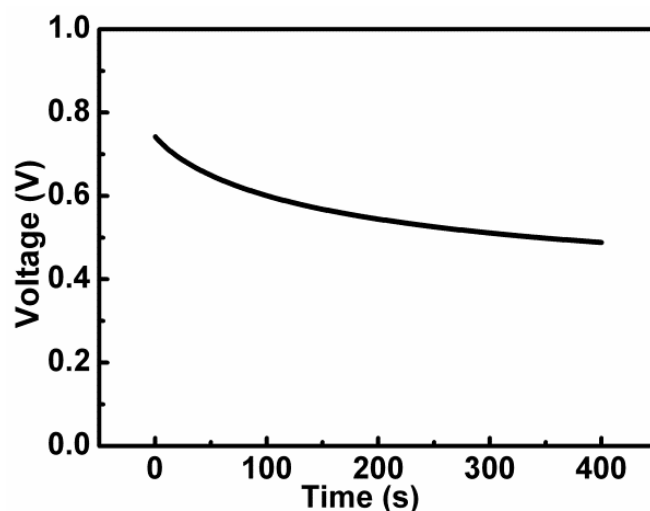
**Fig. S5.** Dependence of the thickness of MWCNT films on the height of MWCNT arrays.



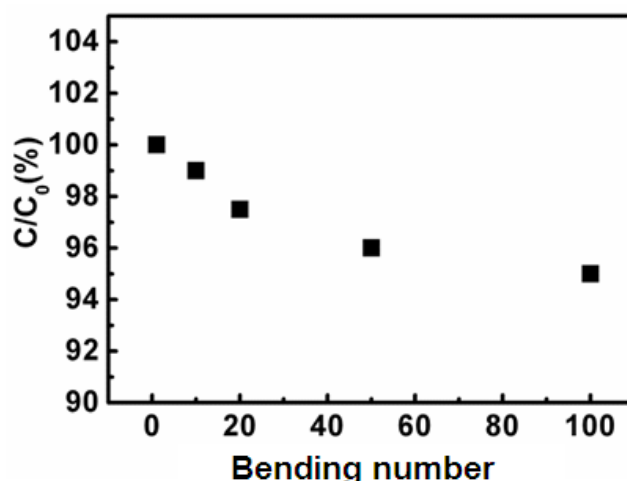
**Fig. S6.** Raman spectra of MWCNTs, PANI, and MWCNT/PANI composite.



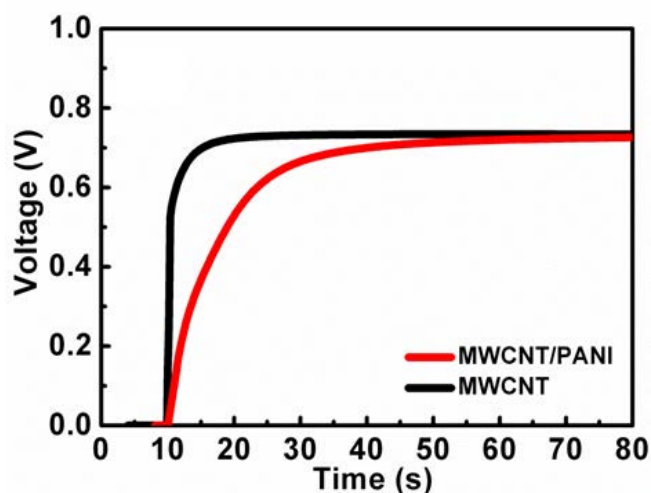
**Fig. S7.** Dependence of the specific capacitance on the thickness of MWCNT films. **a.** The specific capacitance of supercapacitor based on bare MWCNT films as electrodes with the increasing thickness from 10 to 50 μm. **b.** The specific capacitance of supercapacitor based on MWCNT/PANI composite films as electrodes with the increasing thickness from 10 to 50 μm. The same weight of PANI was used in all composite films.



**Fig. S8.** Self discharge of the integrated device based on a MWCNT/PANI composite film.



**Fig. S9.** Specific capacitances for the integrated device based on the MWCNT/PANI composite film during bending. Here  $C_0$  and  $C$  correspond to specific capacitances before and after bending, respectively.



**Fig. S10.** The dynamic voltage of the photo-supercapacitor with bare MWCNT and MWCNT/PANI composite films as electrodes in PVA- $H_3PO_4$  electrolyte under AM1.5 illumination during the photocharging process.

### References for the Supporting Information

- S1. F. Li, J. J. Shi and X. Qin, *Chinese Sci. Bull.* **2009**, *54*, 3900-3905.
- S2. S. Ito, S. M. Zakeeruddin, R. Humphry-Baker, P. Liska, R. Charvet, P. Comte, M. K. Nazeeruddin, P. Péchy, M. Takata, H. Miura, S. Uchida and M. Grätzel, *Adv. Mater.* **2006**, *18*, 1202-1205.
- S3. C. Yu, C. Masarapu, J. Rong, B. Wei and H. Jiang, *Adv. Mater.* **2009**, *21*, 4793-4797.