Electronic Supplementary Information

Integrating Photovoltaic Conversion and Lithium Ion Storage into a Flexible Fiber

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Experimental Section

Preparation of the LS part.

$LiMn_2O_4$ (LMO) and $Li_4Ti_5O_{12}$ (LTO) were synthesized by hydrothermal and solid-state methods, respectively (Figure S2).[1] The resulting LMO nanoparticles (150 mg) and an MWCNT powder (length of 10-20 μm and diameter of 10-20 nm, 15 mg) were dispersed in N,N-dimethylformamide (30 mL) to form a suspension. Two aligned MWCNT sheets (width of 2 cm) drawn out of a spinnable MWCNT array were stacked along the drawing direction and immersed into the above suspension, followed by rolling into a fiber.[2] Similarly, LTO nanoparticles (150 mg) were also dispersed in N,N-dimethylformamide (30 mL), followed by immersion of two stacked MWCNT sheets (width of 2 cm). The aligned MWCNT/LTO hybrid sheet was then scrolled into a fiber and further coated with a thin layer of graphene oxide after immersion into an aqueous graphene oxide solution (~0.5 wt%). A modified Hummer's method was used to synthesize the graphene oxide.[3] Both MWCNT/LMO and MWCNT/LTO fibers were coated with a layer of gel electrolyte and wrapped onto a rubber fiber (diameter of 500 μm) together, which served as flexible substrate for fiber electrodes (Figure S4). The wound two hybrid fibers were inserted into a heat-shrinkable tube (diameter of ~1.5 mm), followed by injection of the gel electrolyte in an argon-filled glove box and encapsulation of the two ends of the tube.

Preparation of the PC part.

Another aligned MWCNT sheet was uniformly wrapped on the surface of the above heat-shrinkable tube.[4] Many MWCNT segments were produced through the use of masks prior to the wrapping process. To prepare the photoanode, a titanium wire (diameter of 127 μm) was twisted into a spring shape, followed by washing with acetone, isopropanol and water. An anodic oxidation method was then conducted to grow perpendicularly aligned titanium dioxide nanotubes on the surface of titanium wire.[5] The resulting titanium spring was annealed at 500 °C for 60 min and treated at 70 °C in a $TiCl_4$ aqueous solution (40 mM) for 30 min, followed by annealing again at 450 °C for 30 min. The treated spring was immersed into an N719 solution (0.3 mM, mixture solvent of equal volume of dehydrated acetonitrile and tert-butanol) for 16 h.

Fabrication of the FIED.

To fabricate an FIED, the LS part was inserted into the spring-shaped photoanodes, followed by encapsulating each PC unit with a heat-shrinkable tube. The titanium wire

from one PC part and the MWCNT sheet from the neighboring PC part were connected through the use of aligned MWCNT sheet. Silver paste was further used to enhance the connection. The photoanode and counter electrode of the PC part was connected to the MWCNT/LTO and MWCNT/LMO electrodes of the LS part, respectively. A switch was incorporated between the PC and LS part to realize the photocharging and discharging processes.

Calculation on the performance.

The mass specific capacity (C_m) was calculated from the charging and discharging profiles based on the equation of $Cm = I \times \Delta t/m$, where I, Δt and m correspond to the discharging current, discharging time and mass of MWCNT/LTO electrode, respectively. The total photovoltaic conversion and energy storage efficiency (η_{total}) was calculated from the following equation.

$$\eta_{total} = \frac{E_{output}}{E_{input}} = \frac{\int UIdt}{P_{input} t_{charge} S}$$

Here E_{output} and E_{input} correspond to the output and input energy at the LS and PC part, respectively. U, I, t, P_{input}, t_{charge} and S are the voltage, discharging current, discharging time, input power density of incident light, charging time and effective area of photoanode, respectively. The effective area of the photoanode was calculated as the projected area according to our previous work.[6] The power and energy densities were calculated based on the overall mass of the cathode and anode for the energy storage part.

Supplementary Figures

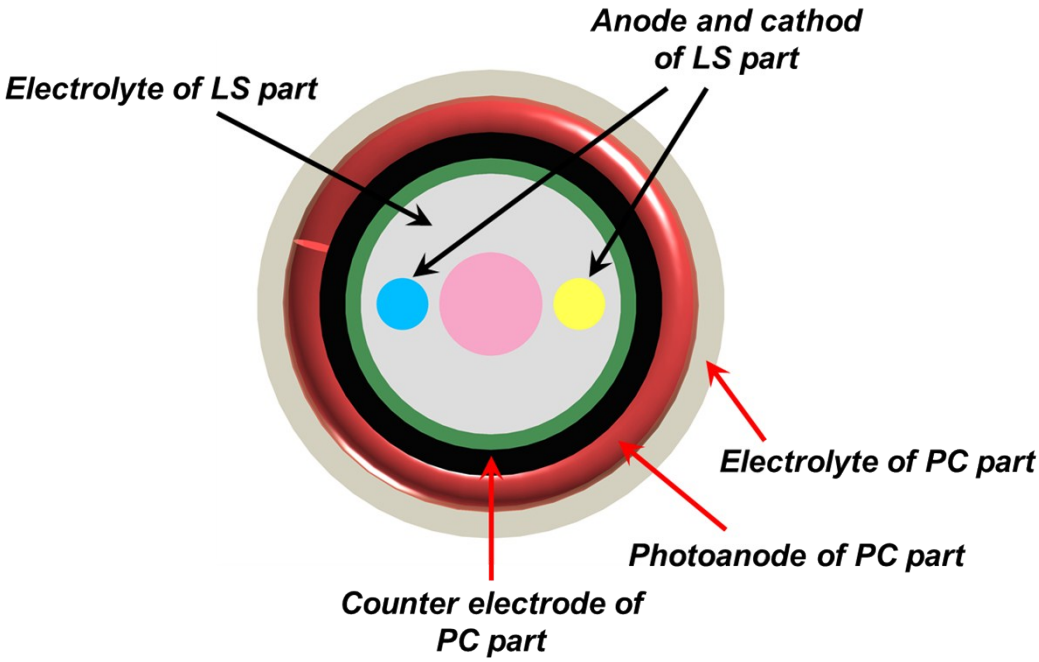

Figure S1. Schematic illustration to the cross-sectional structure of the FIED.

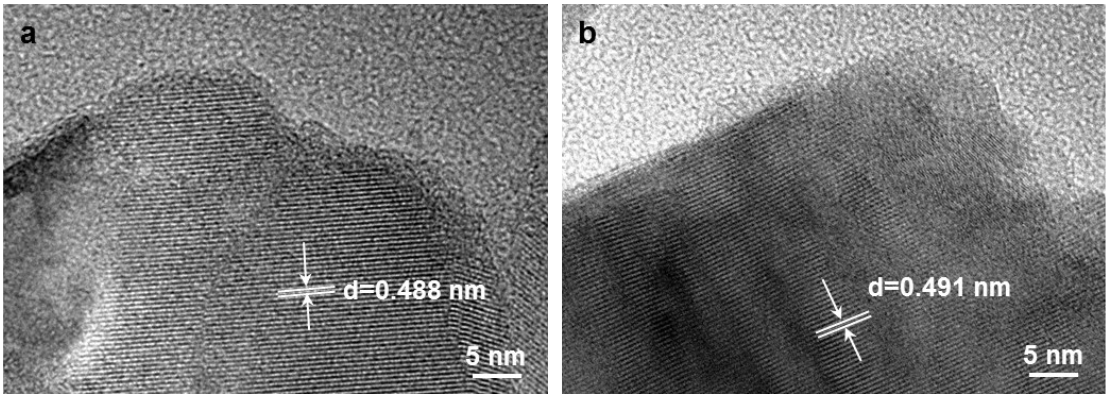

Figure S2. **a** and **b.** TEM images of $LiMn_2O_4$ (LMO) and $Li_4Ti_5O_{12}$ (LTO) nanoparticles, respectively.

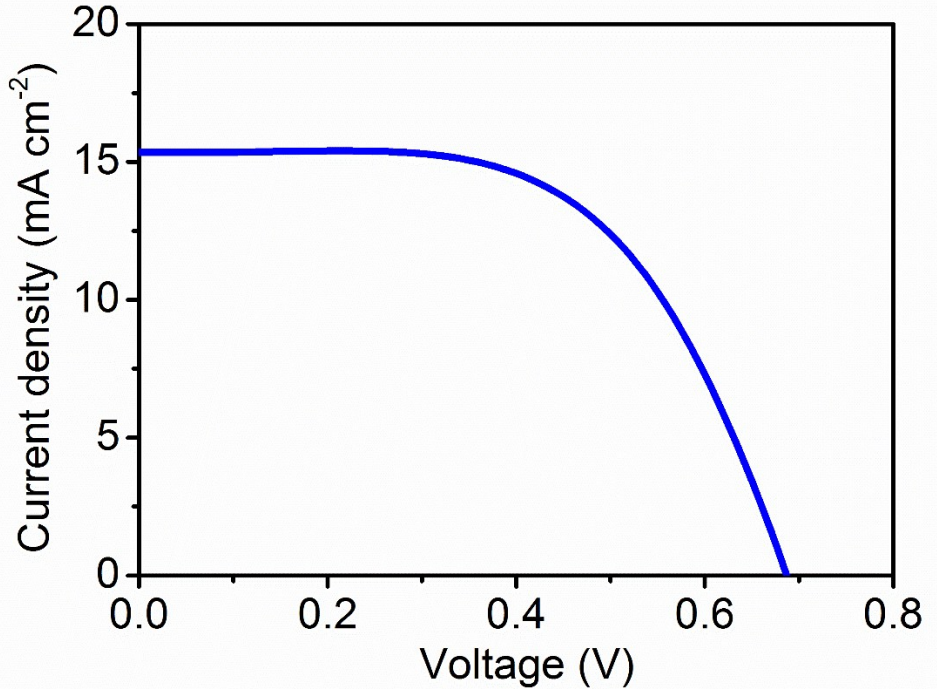

Figure S3. Current density-voltage curve of a photovoltaic conversion unit.

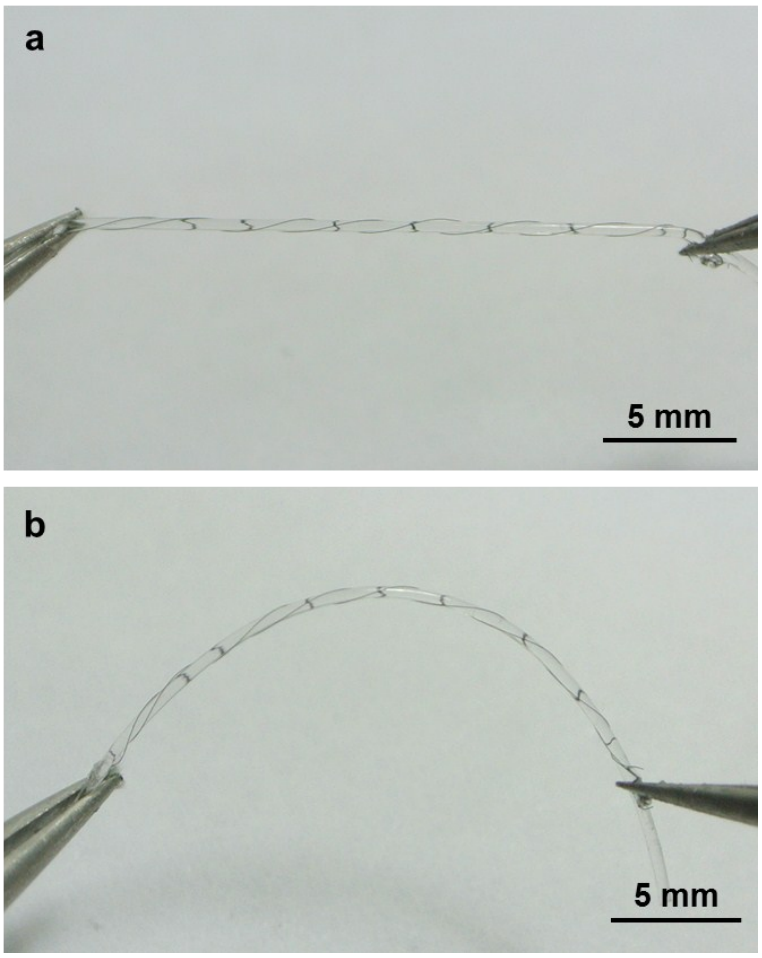

Figure S4. Photograph of multi-walled carbon nanotube (MWCNT)/LTO and MWCNT/LMO hybrid electrodes wrapping on a rubber fiber at original (top image) and bending (bottom image) states.

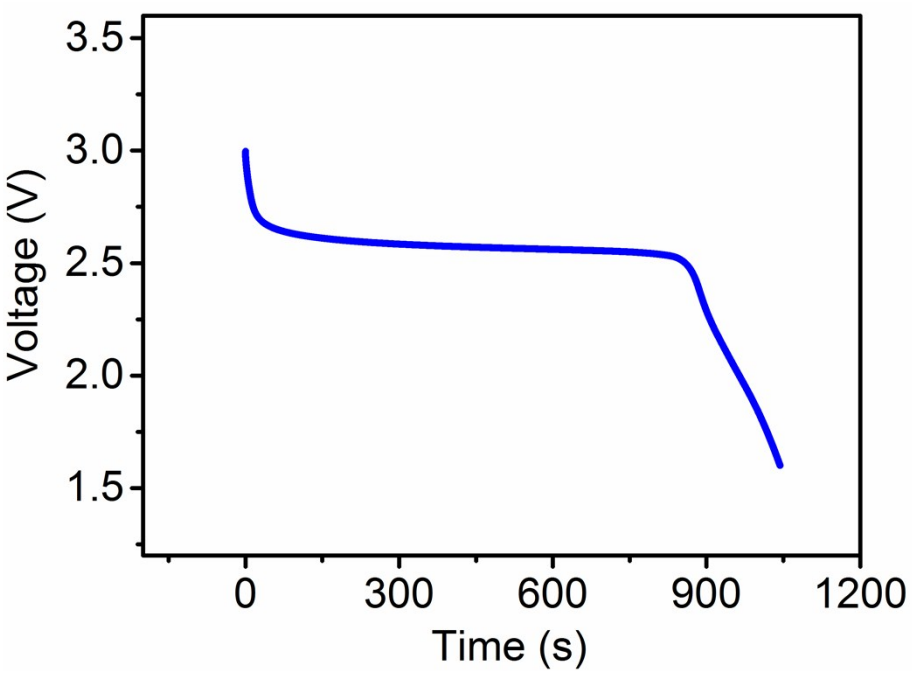

Figure S5. Discharging curve of an LS part with enhanced capacity by using more active materials at both fiber electrodes. Discharge current, 0.02 mA.

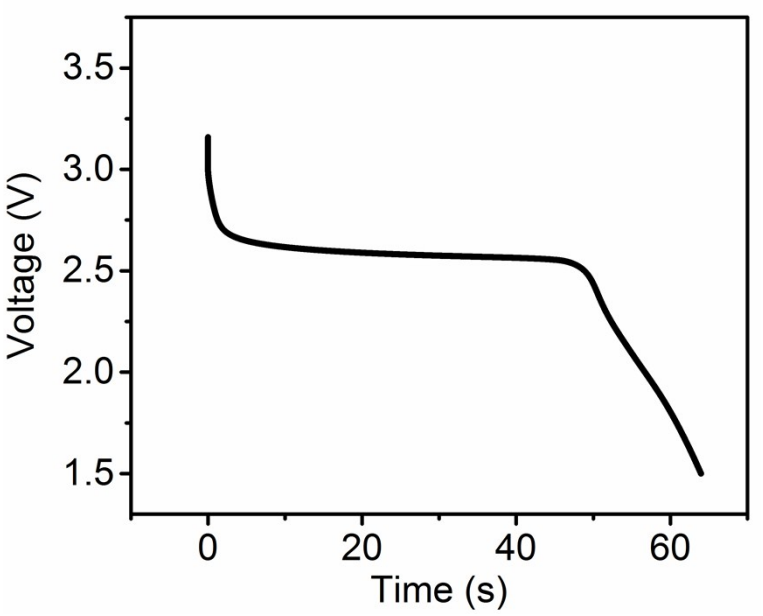

Figure S6. Discharging curve of an LS part with increased discharge plateau by adding amorphous carbon to MWCNT/LMO electrodes. Discharge current, 0.05 mA.

Reference

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4. Z. Yang, J. Deng, H. Sun, J. Ren, S. Pan, H. Peng, *Adv. Mater.* **2014**, *26*, 7038-7042.
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