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## **ADVANCED MATERIALS**

## Supporting Information

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A Gum-Like Lithium-Ion Battery Based on a Novel Arched Structure

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## **Supporting Information**

**LMO and LTO synthesis.** LMO nanoparticles were synthesized by a hydrothermal method. LiOH (0.377 g) and MnO<sub>2</sub> (1.37 g) were first dissolved in H<sub>2</sub>O (40 mL), followed by addition of glucose (0.2 g) and H<sub>2</sub>O (40 mL). After reacting at 200 °C for 24 h, LMO nanoparticles were obtained. LTO nanoparticles were synthesized by a solid-state method. TiO<sub>2</sub> and Li<sub>2</sub>CO<sub>3</sub> with a molar ratio of 2.5 was mixed and heated at 800 °C for 24 h in a N<sub>2</sub> atmosphere. LTO nanoparticles were produced after a ball-milling treatment for 20 h.

**Gel electrolyte synthesis.** The gel electrolyte was prepared by mixing methylene chloride and acetone at a weight ratio of 40/1, followed by addition of poly (ethylene oxide) ( $M_w \sim 600,000, 0.35$  g), uccinonitrile (0.35 g) and lithium bis (trifluoromethane) sulfonimide (0.30 g).

Arched electrode preparation. Specifically for the preparation of the arched electrodes, LTO or LMO nanoparticles were first blended with CNT powders (90/10, wt/wt) and dispersed in N-methyl pyrrolidone with a concentration of 5 mg/mL. The resulting suspension was spray-deposited onto a CNT sheet that was paved on a Cu foil, followed by stacking another CNT sheet to form a sandwich structure. Afterwards, an un-curing PDMS fluid was incorporated into the sandwich structure by spin-coating. The rotating speed was 5000 rpm and the lasting time was 90 s. Then the PDMS was soaked into the sandwich structure by vacuum pumping to form a composite. The composite was transferred on a prestretched PDMS substrate. After curing, peeling off the Cu foil and releasing the substrate, an arched electrode was obtained. Also, the composite could be free-standing if not transferred onto the substrate after curing. The weight percentage of PDMS could be tested by weighing in-process products after every step. They were ranged from 40 to 50 wt% for composites, in which areal mass densities of LTO or LMO nanoparticles were changed from 1 to 10 mg cm<sup>-2</sup>. Additionally, the composites were porous to facilitate the infiltration of the electrolyte (Figure S13).

**Battery prototype.** For electrodes, composites were made with a rectangular shape with dimensions of 16.5 cm  $\times$  1.5 cm and became irregular with an area of ~6 cm<sup>2</sup> after

release (Figure S14a). The extended bare CNT sheet was connected to the wires of the outer circuit. The tree pattern demonstrated a length of 3 cm and covered the underlying composite (Figure S14b).

**Characterization.** The structures were analyzed by X-ray diffraction (Bruker, D8 ADVANCE). The morphologies were characterized by scanning electron spectroscopy (Hitachi, 4800-1) and transmission electron microscopy (JEOL, JEM-2100F).

**Electrical property test.** The sheet resistance was tested by a four-point probe meter (Guangzhou institute of semiconductors, SDY-4). The rippled conductors and arched electrodes were placed on the test platform. After the probes touched the sample and the current was adjusted, the data of sheet resistance could be directly read. As to study the resistance change under stretching, a digital multimeter (Victor, VC9807A<sup>+</sup>) was used, whose two clamps were fixed on the two ends of the sample. The gauge length between two clamps was increased/decreased when stretching/releasing. The resistance could be monitored on the stretching strain.

**Electrochemical test.** For half cells, LiClO<sub>4</sub> in a mixture solvent of ethylene carbonate and diethyl carbonate (50/50, v/v) with a concentration of 1 M was used as the electrolyte for half cells. 175 mA g<sup>-1</sup> and 148 mA g<sup>-1</sup> were used as 1C for LTO-incorporated and LMO-incorporated composites, respectively. For half cells, the specific capacity and current density were calculated from the active LTO or LMO material. For the full cells that were limited by the anode, the specific capacity and current density were calculated from the later capacity and current density were calculated from the specific capacity and current density were system to be less than 1 ppm.



Figure S1. Schematic illustration to the preparation of the arched electrodes.



Figure S2. High-resolution transmission electron microscopy image of carbon nanotubes (CNTs).



**Figure S3. a**. SEM image of a CNT sheet on PDMS substrate without prestretching. **b**. SEM image of a rippled conductor with a prestrain of 450% by a cross-sectional view.



**Figure S4.** X-ray diffraction patterns of lithium manganese oxide (LMO) and lithium titanium oxide (LTO).



**Figure S5. a.** Mixture of CNT powders and LTO nanoparticles. **b.** A blend of CNT powders and LTO nanoparticles on CNT sheet. **c.** Mixture of CNT powders and LMO nanoparticles. **d.** A blend of CNT powders and LMO nanoparticles on CNT sheet.



**Figure S6.** Schematic illustration to the movement of the arched electrodes under stretching and releasing.



**Figure S7.** Comparison of the super-stretchy arched electrodes with the other reported stretchable conductors<sup>S1-S8</sup>.

- S1: Adv. Mater. 2012, 24, 1073.
- S2: Adv. Mater. 2009, 21, 4793.
- S3: Adv. Mater. 2013, 25, 1058.
- S4: ACS Nano 2014, 8, 1039.
- S5: Sci. Rep. 2014, 4, 3612.
- S6: Nano Lett. 2012, 12, 6366.
- S7: Nat. Nanotechnol. 2011, 6, 788.
- S8: Adv. Mater. 2011, 23, 3989.



**Figure S8.** Schematic illustration to the preparation of another type of stretchable electrodes without sandwich and arched structures, i.e., CNT-LTO and CNT-LMO composites.



**Figure S9.** SEM images of a CNT-LTO composite before stretching (**a**) and after stretching for 500 times at a strain of 400% (**b**).



**Figure S10.** An optical photograph showing a gum-like battery fixed on a moving stage to test the dynamic electrochemical performance under stretching/releasing.



**Figure S11.** Dynamic test within a full charge/discharge cycle at a current density of 2 C and a stretching/releasing speed of  $0.5 \text{ mm s}^{-1}$ .



**Figure S12. a.** Conventional wavy structure showing a wavy conductive film on an elastic substrate. **b.** Novel arched structure enabling a high stretchability and a stable performance for a functional guest-anchored composite. The composite exhibits a sandwich structure in which the functional guests are between two conductive films.



Figure S13. SEM image of the surface of a CNT/LTO-CNT composite showing a porous structure.



Figure S14. a. Photograph of the composite electrode. The composite became irregular in shape after released from a rectangle of  $16.5 \text{ cm} \times 1.5 \text{ cm}$  with a prestrain of 450%. b. Photograph of a battery. The tree pattern exhibited a length of 3 cm to cover the underlying composite. The brown and silver belts connect the negative and positive poles, respectively.



**Figure S15.** SEM images of the CNT/LMO-CNT composite before stretching (**a**) and after stretching for 500 times at a strain of 400% (**b**). The prestrain is 450%.



**Figure S16.** Dependence of resistance change on stretching strain (**a**) and stretching number (**b**) for CNT/LMO-CNT composites. The prestrain is 450%.



**Figure S17.** Effect of prestretching and stretching treatments on voltage profiles (**a**) and long-life performance (**b**) for CNT/LMO-CNT composites. Rate capability of CNT/LMO-CNT composites after 500 stretching cycles at a strain of 400% (**c**).