Supporting Information

High-performance Fiber-shaped Li-ion Battery Enabled by Surface-reinforced Self-supporting Electrode

Cong Liu,[a†] Yining Lao,[a†] Fan Yang,[a] Xiaoqi Gong,[a] Xiaotong Wang,[a] Jiang Lu,[*a] Zhiping Zeng[*b] and Dingshan Yu[*a]

a. Key Laboratory for Polymeric Composite and Functional Materials of Ministry of Education, Key Laboratory of High-Performance Polymer-based Composites of Guangdong Province, School of Chemistry, Sun Yat-sen University, Guangzhou 510006, China.
b. State Key Laboratory of Optoelectronic Materials and Technologies, Guangzhou Key Laboratory of Flexible Electronic Materials and Wearable Devices, Nanotechnology Research Center, School of Materials Science and Engineering, Sun Yat-sen University, Guangzhou, 510275 P. R. China. Email: zzhip8@mail.sysu.edu.cn.

* Corresponding author

Email addresses: gaofenzi@mail.sysu.edu.cn (Jiang Lu), zzhip8@mail.sysu.edu.cn (Zhiping Zeng), yudings@mail.sysu.edu.cn (Dingshan Yu)

† These authors contributed equally to this work.

Experimental section

Preparation of fiber electrodes

In typical, sodium alginate binder was dissolved in 10 mL distilled water to form an aqueous solution. Then，8g active material (LTO or LFP material) and 1g Super P carbon were added to the above solution. Subsequently, the mixture was diluted to 20 mL, followed by vigorously stirring for 2 h. The solution was placed into a 2.5 mL syringe and squeezed into the coagulation solution containing chitosan and acetic acid at a proper extrusion pressure to obtain the fiber-shaped electrode after a through solvent exchange. The fiber was placed in the coagulation solution for 1 h, then naturally-dried at room temperature. The obtained fiber electrodes were used for the subsequent characterization.

Materials Characterizations

Powder X-ray diffraction (PXRD, D/MAX 2200 VPC) with Cu Kα radiation (λ = 0.154 nm) were executed to investigate the crystalline phases. FESEM (ZEISS Ultra-55) was applied to examine the microstructure properties. The rheological tests were carried out using the AR 2000 rotary rheometer (TA Instruments, USA) at 25 °C.

Electrochemical tests

The separator-wrapped-cathode and anode fiber electrodes are twisted together and packaged into a heat-shrinkable tube (diameter: ~ 2.0 mm), and then electrolyte (1M $LiPF_6$ in EC: DEC: DMC=1:1:1) was injected into the tube in an Ar-filled glove box. Once the cell was well packaged, it was taken out of the glove box for battery tests under ambient lab conditions. The assembled batteries were galvanostatic discharged/charged in the voltage window of 1.0-2.5 V. Cyclic voltammograms (CV) and electrochemical impedance spectroscopy (EIS) were performed on electrochemical workstation (CHI660E). Considering the theoretical capacity of LFP and LTO electrode (~ 170 mAh g^{-1} Vs. ~ 175 mAh g^{-1}), the N/P ratio (~ 1.1) was set to achieve a capacity balance.

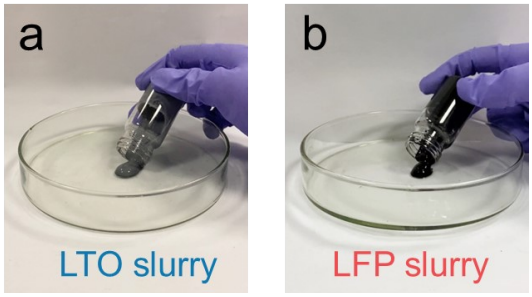

Fig. S1 Photographs of the prepared electrode slurries for fiber batteries.

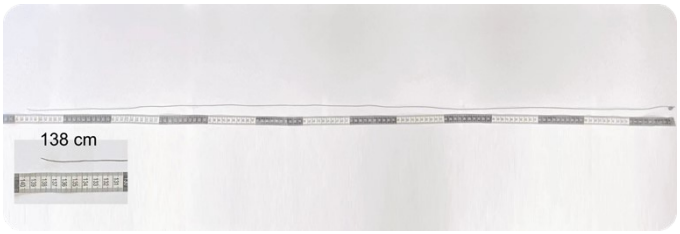

Fig. S2 Photographs of fiber-shape electrode with a length of about 138 cm.

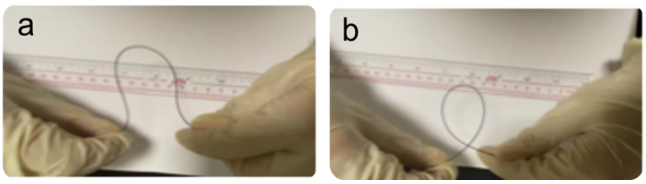

Fig. S3 Photographs of fiber-shape electrode under folding deformation.

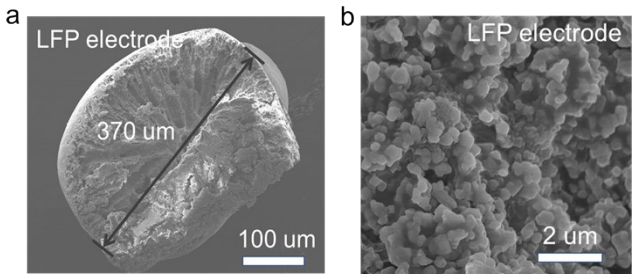

Fig. S4 (a) Cross-sectional and (b) SEM image of LFP fiber electrode.

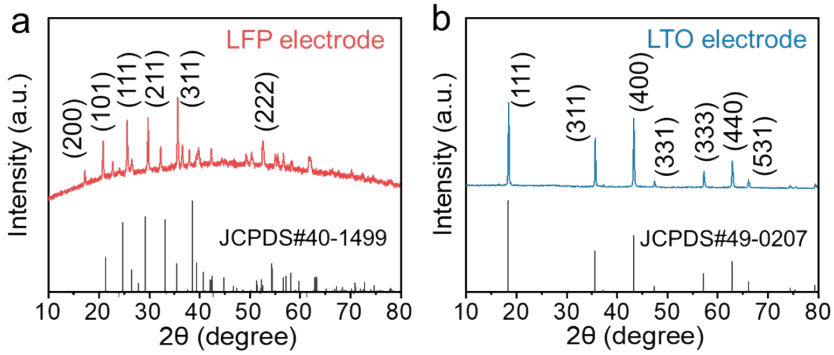

Fig. S5 XRD patterns of (a) LFP electrode and (b) LTO electrode.

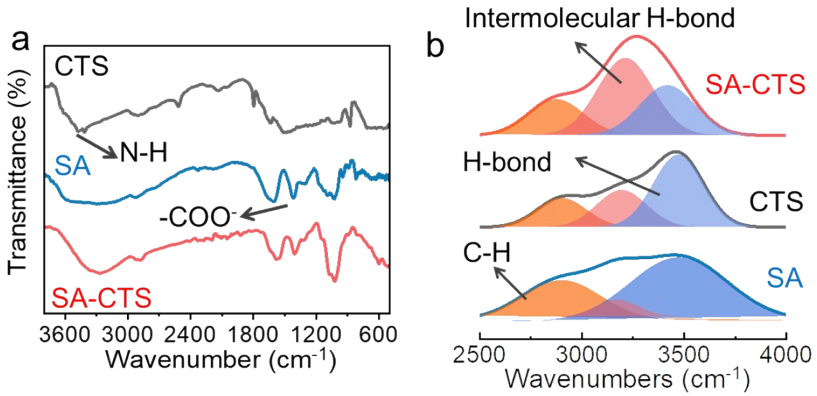

Fig. S6 FT-IR spectra of SA, CTS and SA-CTS.

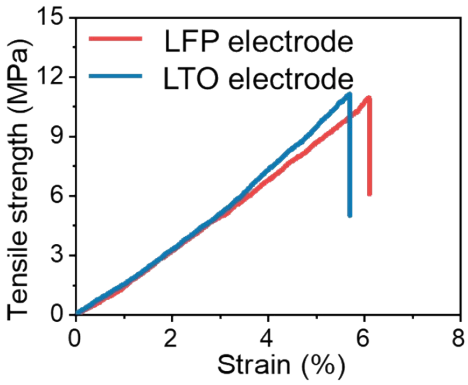

Fig. S7 Typical curves of tensile strength testing for LFP and LTO fiber electrodes.

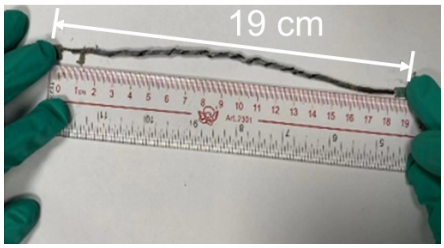

Fig. S8 The photo of as-fabricated fiber-shape full battery.

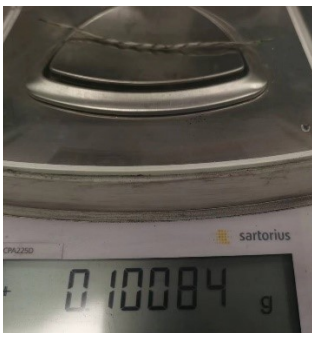

Fig. S9 The overall mass of the fiber-shape full battery.

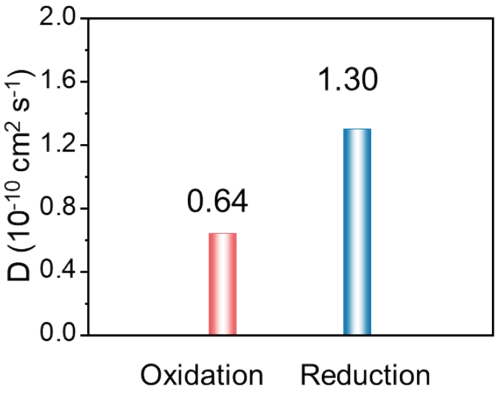

Fig. S10 Diffusion coefficient derived from CV response for fiber LFP//LTO battery.

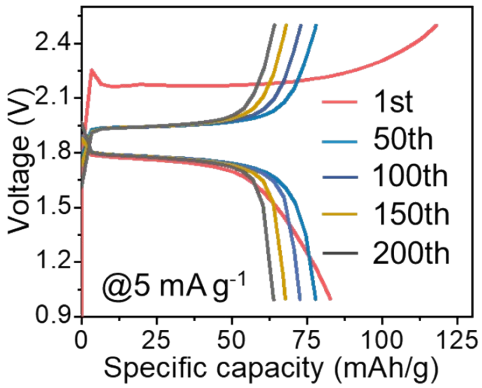

Fig. S11 Selected GCD profiles at 5 mA g^{-1} during long-term cycling.

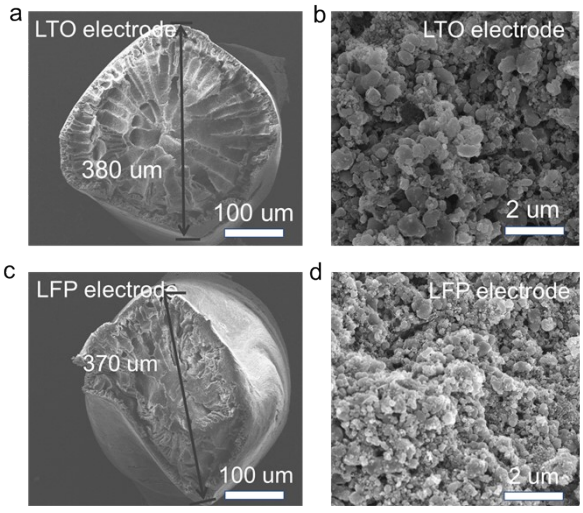

Fig. S12 Cross-sectional and high magnification SEM images of (a, b) LTO and (c, d) fiber electrode after cycling.

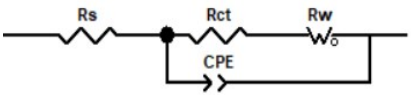

Fig. S13 The equivalent circuit of EIS results.

Fig. S14 The open-circuit voltage (Voc) of this fiber device.

Tab. S1 The comparison of linear capacity and gravimetric energy density of fiber-shaped full cells.

Cathode/Anode	Liner capcatity (mAh cm^{-1})	Gravimetric energy density (Wh kg^{-1})	Ref.
$LiFePO_4$ $Li_4Ti_5O_{12}$	0.144	50.6	This work
$LiFePO_4$ $Li_4Ti_5O_{12}$	0.14	---	Adv. Mater., 2023, 15, 2211201
$LiFePO_4$ $Li_4Ti_5O_{12}$	0.092	---	Adv. Funct. Mater., 2017, 27, 1703140
$LiMn_2O_4$-CNT $LiTi_2(PO_4)_3$-CNT	0.05	---	Nat. Nanotechnol., 2022, 17, 372-377
$LiMn_2O_4$ TiO_2/rGO	0.028	---	Nano Lett. , 2017, 17, 3432-3549
$LiMn_2O_4$-CNT $Li_4Ti_5O_{12}$-CNT	0.0036	30	J. Mater. Chem. A, 2014, 2, 11054-11059
$LiMn_2O_4$-CNT $Li_4Ti_5O_{12}$-CNT	0.0028	27	Angew. Chem. Int. Ed., 2014, 126, 7998-8003
$LiMn_2O_4$-CNT Polyimide-CNT	0.0012	48.93	J. Mater. Chem. A, 2016, 4, 9002-9008

Tab. S2 The resistances of the fiber-shaped full cell before and after 200 cycles.

Fiber-shaped LFP//LTO full cell	Internal resistance (Rs, Ω)	Charge transfer resistance (Rct, Ω)	Warburg resistance (Rs, Ω)
Before cycles	8.53	57.35	82.76
After cycles	9.25	83.93	95.86