## **Supporting Information**

## Highly-Integrated and Interconnected CNT Hybrid Nanofibers Decorated with α-Iron Oxide as Freestanding Anodes for Flexible Lithium Polymer Batteries

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<sup>b</sup>Department of Solar & Energy Engineering, Cheongju University, Cheongju, Chungbuk, 28503, Republic of Korea

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Jung Sang Cho Tel.: (+82) 10-2649-5966; Fax: (+82) 43-262-2380. E-mail: jscho@cbnu.ac.kr Jae-Kwang Kim Tel.: (+82) 43-229-8557; Fax: (+82) 43-229-7322. E-mail: jaekwang@cju.ac.kr In Fig. S1a, the FT-IR spectrum of PAN before acid-treatment showed a unique peak at 2243 cm<sup>-1</sup> which is attributed to the nitrile (-C=N) group of PAN. However, after treatment in Fig. S1b, PAN was hydrolyzed to form a sulfonated polyacrylamide (-SO<sub>3</sub>H, O=C-NH<sub>2</sub>) group. The sulfonated polyacrylamide group of sulfonated PAN is determined by the amide group (NH<sub>2</sub>-C=O) and sulfonic acid group (SO<sub>3</sub>H) characterized by five peaks of N-H, C-N, C=O, O=S=O, and C-S. Therefore, 588 cm<sup>-1</sup> (N-H), 1214 cm<sup>-1</sup> (C-N), 1580 cm<sup>-1</sup> (C=O), 1197 cm<sup>-1</sup> (O=S=O), and 1040 cm<sup>-1</sup> (C-S) were definitely confirmed from Fig. S1b. In the case of CNTs, peaks located at 1640 cm<sup>-1</sup> (C=O) and 3400 cm<sup>-1</sup> (O-H) corresponding to the carboxylic acid group (-COOH) attached onto surfaces of the acid-treated CNTs were newly observed after acid-treatment, as shown in Fig. S1d. Therefore, the attachments of both carboxylic group on CNTs and sulfonated polyacrylamide group on PAN after acid-treatment were directly proven by FT-IR results.



**Fig. S1**. FT-IR spectra of PAN and CNTs before and after acid-treatment: (a) PNA before acid-treatment, (b) PNA after acid-treatment, (c) CNTs before acid-treatment, and (d) CNTs after acid-treatment.



Fig. S2. TGA curve of (a) the as-spun  $Fe(acac)_3/PAN/CNTs$  composite nanofibers obtained after electrospinning process and (b) HI-CNT/Fe<sub>2</sub>O<sub>3</sub> fiber .



Fig. S3. (a,b)  $N_2$  adsorption-desorption isotherms and (c,d) BJH adsorption pore size distributions: (a,c) as-spun Fe(acac)<sub>3</sub>/PAN/CNTs composite nanofibers and (b,d) HI-CNT/Fe<sub>2</sub>O<sub>3</sub> nanofibers.



**Fig. S4**. Pore size distribution of HI-CNT/Fe<sub>2</sub>O<sub>3</sub> nanofibers calculated from the N<sub>2</sub>-DFT model using desorption isotherms.



**Fig. S5**. CV curve of the CNTs prepared by acid- and heat- treatments at a scan rate of 0.1 mV s<sup>-1</sup> in the 0.001–3.0 V range.



**Fig. S6**. (a) FE-SEM image, (b) EDS spectrum, (c) charge-discharge profile, and (d) cycle performance of HI-CNT/Fe<sub>2</sub>O<sub>3</sub> nanofibers etched by HCl solution.



**Fig. S7** Electrochemical impedance spectra (EIS) of free-standing HI-CNT/Fe<sub>2</sub>O<sub>3</sub> nanofibers electrode cell with cycling.



**Fig. S8**. Electrochemical properties of LiFePO<sub>4</sub> fiber cathode in half-cell: (a) charge/discharge profile and (b) cycling performance at a current density of 200 mA  $g^{-1}$ .