Supporting Information

DNA-Directed Growth of $FePO_4$ Nanostructure on Carbon Nanotubes to Achieve Nearly 100% Theoretical Capacity for Lithium-Ion Batteries

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Experimental

Preparation of single-stranded DNA-wrapped double-walled carbon nanotube

To prepare single-stranded DNA, the following procedures were carried out. First, 50 mg DNA (200 base pairs) was dissolved in 45 mL of distilled water under sonication for 5 min, and then 2.5 mL of 2 M sodium hydroxide solution was added. After incubation for 5 min, the DNA solution was neutralized by adding 2.5 mL of 2 M hydrochloric acid. The freshly prepared single-stranded DNA was used for all experiments. 50 mg of double-walled carbon nanotube (DWNT, NanoLab Inc.) was suspended in above freshly prepared single-stranded DNA solution and the mixture was kept in an ice-water bath and sonicated (ca. 450 W/cm^2) for 90 min. After sonication, the mixture was centrifuged at 8,000 rpm for 30 min and insoluble material was removed, leaving DNA-dispersed DWNT solutions (DNA@DWNT). The final concentration of DNA@DWNT was adjusted to around 1.0 mg ml^{-1} by evaporating water for further use.

Fabrication of $FePO_4$-based materials

The freshly prepared DNA@DWNT was mildly sonicated for 5 min before use and then cooled to 4°C. The cooled DNA@DWNT was incubated with 10 mM Fe^{3+} aqueous solution ($FeCl_3$•$6H_2O$, pH around 2.6) for 24 hours at 4°C. The low temperature of 4°C was used in order to suppress hydrolysis of Fe^{3+} ions into the hydroxide. Growth of $FePO_4$ on DNA@DWNT ($FePO_4$-DNA@DWNT) and the assembly into network nanostructure were completed by immersing the Fe^{3+} immobilized DNA@DWNT in 10 mM PO_4^{3-} aqueous solution (NaH_2PO_4, pH 7.0) for 24 hours at 4°C. After washing and drying, the above process was repeated. The obtained product was washed several times using distilled water and dried at 80°C under vacuum overnight. Dehydration of $FePO_4$-DNA@DWNT was carried out by thermal annealing by heating with a rate of 2 °C per min to 350 ˚C, staying at 350 ˚C for 2 h, and cooling to room temperature naturally. $FePO_4$-DWNT, $FePO_4$ and their dehydrated samples were prepared using the same fabrication procedures as that of $FePO_4$-DNA@DWNT.

Material characterizations

The morphology and structure of the samples were investigated by transmission electron microscopies (TEM, JEM-2010F, Japan). The compositions and elemental mappings of the products were determined by energy dispersive X-ray spectroscopy (EDS, JSM-6700F, Japan). Thermal behaviour of samples was analyzed by simultaneous thermogravimetry and differential thermal analysis (Shimadzu DTG-60). During thermogravimetry measurements, the samples were heated from room temperature up to 800 °C at a heating rate of 10 °C min^{-1} in a dynamic atmosphere of air (35 mL min^{-1}) using α-alumina crucibles.

Lithium ion battery construction and measurements

To prepare lithium ion battery cathodes, $FePO_4$-based materials ($FePO_4$-DNA@DWNT, $FePO_4$-DWNT and $FePO_4$) were mixed with Super-P carbon and polyvinyldifluoride (Aldrich), with weight ratio of 90:5:5, in N-Methylpyrrolidone solvent to produce homogeneously mixed gel-like slurry. The slurry was punched into disks with diameters of 13 mm. The disks were coated onto aluminium foils and then punched. The samples were dried under vacuum at 80 ^oC under vacuum for 6 h to form the cathodes. The total material loading was ~2 mg cm^{-2}. Before the lithium ion battery packaging, the electrodes were dried again at 80 ^oC under vacuum for 1 h and then directly transferred to Ar-protected glove box, where the cells were assembled. Swagelok-type cells were made using as-prepared cathodes, Celgard 2250 as separator, and lithium metal foil as counter electrode. The electrolyte was 1.0 M $LiPF_6$ in 50:50 w/w mixture of ethylene carbonate and diethyl carbonate. Galvanostatic

charge/discharge measurements were carried out using a NEWARE battery testing system and data was analyzed by using BTS data analyzer. Electrochemical test was performed at charge/discharge rates from C/10 to 10C at voltage range of 2.0 to 4.0 V. Impedance measurements were carried out using Princeton Applied Research 2273 potentiostat and impedance analyzer and the response was recorded in a frequency range from 10^5 Hz to 100 mHz.

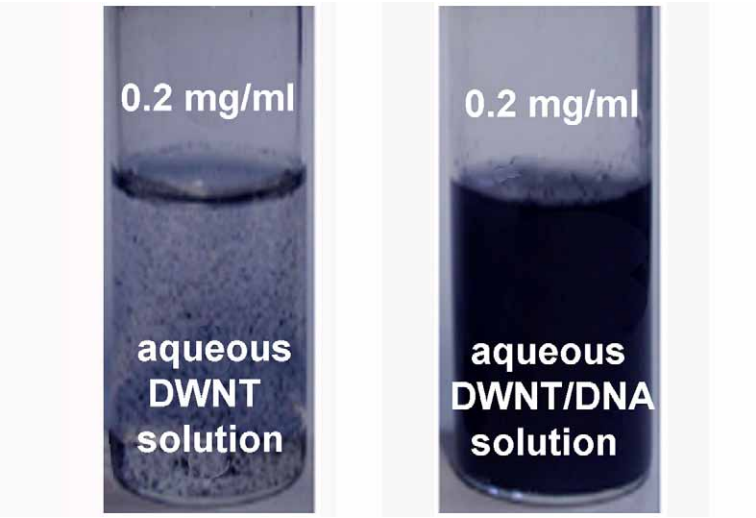

Fig. S1. DWNT (left) and DNA@DWNT (right) aqueous solution (0.2 mg/ml).

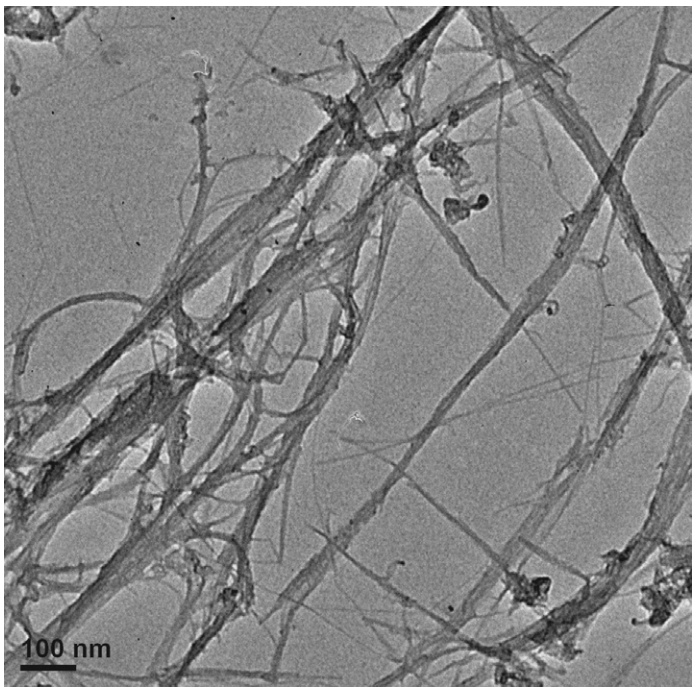

Fig. S2. TEM image of DWNT.

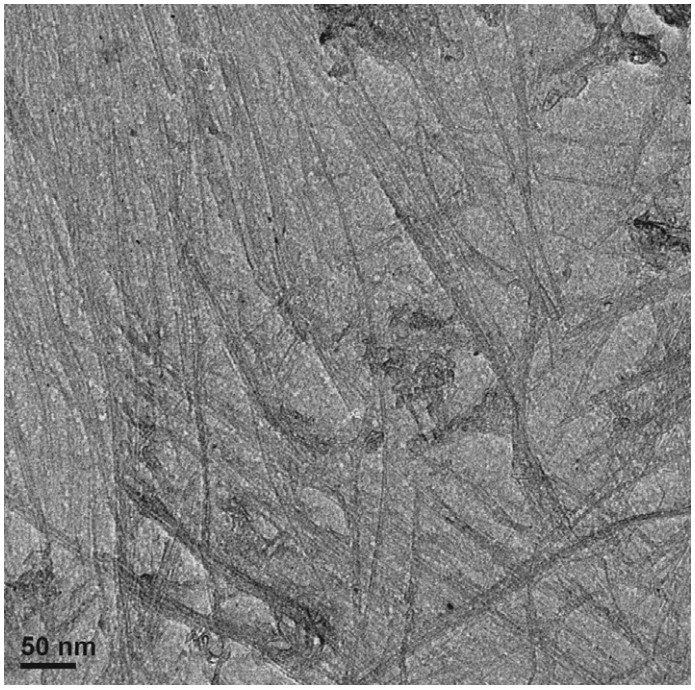

Fig. S3. TEM image of DNA@DWNT.

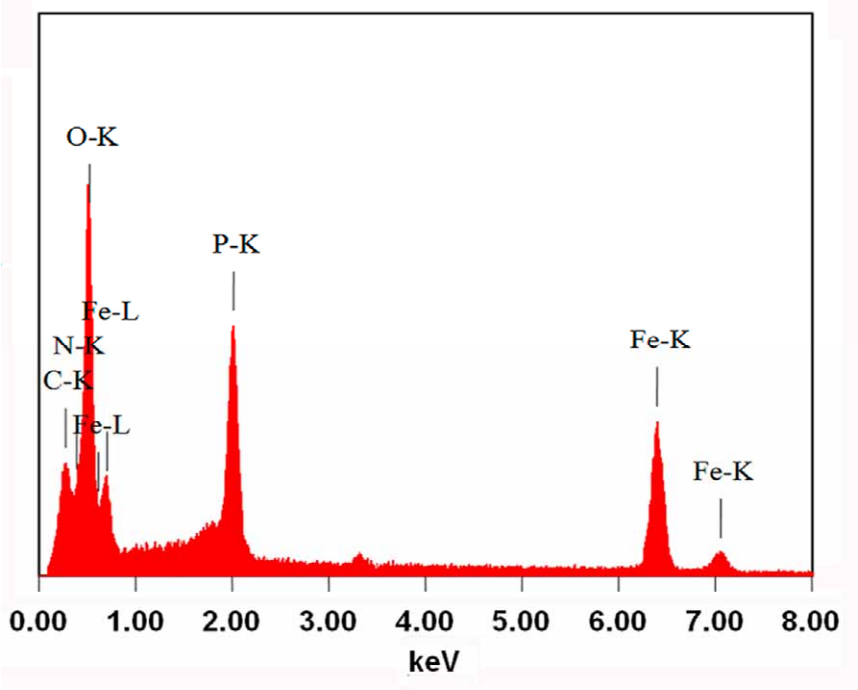

Fig. S4. Energy-dispersive X-ray spectroscopy spectra of $FePO_4$-DNA@DWNT.

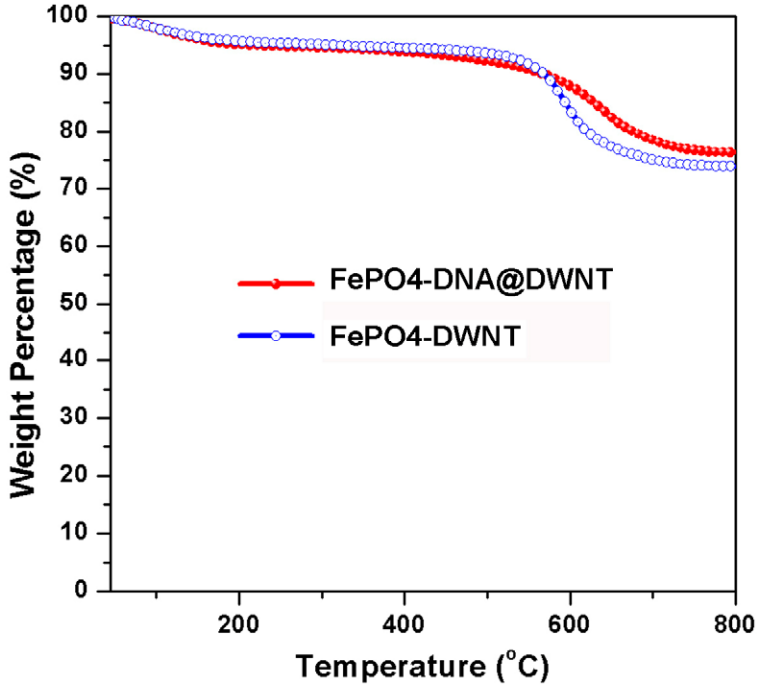

Fig. S5. Thermogravimetric analysis of $FePO_4$-DNA@DWNT and $FePO_4$-DWNT after thermal annealing.

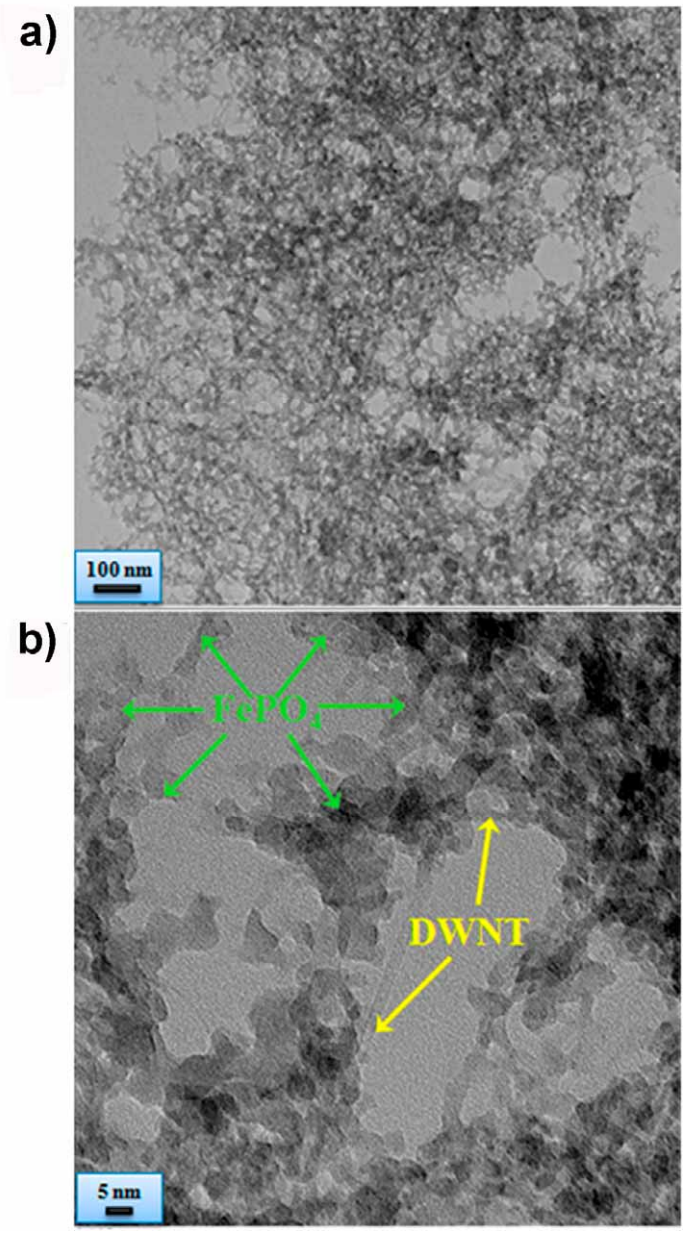

Fig. S6. TEM images with low a) and high b) magnification of $FePO_4$-DNA@DWNT after thermal annealing.