# **Electronic supplementary information**

# Ultrafine FeCo<sub>2</sub>O<sub>4</sub>/Fe<sub>2</sub>O<sub>3</sub> nanoparticles anchored on carbon nanotubes as high-performance anode materials for lithium-ion battery

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### **Experimental section**

#### **1.1 Pretreatment of CNTs**

CNTs with radial diameters ranging from 1 to 2 nm were obtained from Jiangsu Xianfeng Nanomaterials Technology Co., Ltd. In a round-bottom flask, varying amounts of CNTs were added, followed by deionized water (70 mL) and 8 M nitric acid solution (70 mL). After heating, the mixture was refluxed in a water bath at 40°C for 6 hours with constant stirring. Following the acid treatment, the CNTs were collected and rinsed with deionized water and anhydrous ethanol until the filtrate reached a neutral pH. Finally, the acid-treated CNTs were vacuum-dried at 40°C.

## 1.2 Synthesis of FeCo<sub>2</sub>O<sub>4</sub>@Fe<sub>2</sub>O<sub>3</sub>/CNTs

The synthesis procedure for FeCo<sub>2</sub>O<sub>4</sub>@Fe<sub>2</sub>O<sub>3</sub>/CNTs composite material is illustrated in Figure 1(a). First, 12 mmol of 2-methylimidazole was dissolved in methanol (20 mL) and stirred for 30 minutes (designated as solution A). Concurrently, 1 mmol of Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O and 2 mmol of Co(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O were dissolved in methanol (20 mL) and stirred for 30 minutes (designated as solution B). Solution B was gradually poured into solution A, followed by stirring for an additional 30 minutes, thus forming solution C. Various amounts of pretreated CNTs were introduced into solution C and sonicated for 15 minutes. The mixture was placed in a 100 mL reaction vessel, followed by a solvent thermal reaction at 180 °C for 24 hours. The resulting precipitate was rinsed 3 times with methanol, collected by centrifugation, and dried in a vacuum oven at 60°C. The dried sample underwent calcination at 425°C in a muffle furnace for 3 hours to produce the final FeCo<sub>2</sub>O<sub>4</sub>/Fe<sub>2</sub>O<sub>3</sub>@CNT material.

For mass ratios of FeCo<sub>2</sub>O<sub>4</sub>/Fe<sub>2</sub>O<sub>3</sub> to CNTs of 20:1, 10:1, and 5:1 (12, 24, and 48 mg of CNTs were added, respectively), resulting in three distinct FeCo<sub>2</sub>O<sub>4</sub>/Fe<sub>2</sub>O<sub>3</sub>@CNT materials, labeled as FC@CNTs-12, FC@CNTs-24, and FC@CNTs-48. The sample without CNTs was labeled as FC.

#### **1.3 Material characterization**

X-ray diffraction (XRD, Cu Kα, 40 mA, 40 kV; D8 ADVANCE) was used to study the crystal structure of the materials. The morphology and the lattice fringe information of the materials were analyzed by scanning electron microscopy (SEM, SU8010) and transmission electron microscopy (TEM, JEOL JEM-2100). X-ray photoelectron spectroscopy (XPS, ESCALAB, Thermo Scientific)

was used to determine the valence state distribution of elements in the materials. Thermogravimetric analyzer (TGA, Mettler Toledo TGA/DSC<sup>3+</sup>) was applied to investigate the weight loss of material. The samples were subjected to TGA from 50 to 900 °C at a heating rate of 10 °C min<sup>-1</sup> in an air atmosphere.

# **1.4 Electrode preparation**

The electrode was fabricated using a conventional slurry method. The required materials were weighed according to the mass ratio of CMC: acetylene black: active substance (1: 1: 8). The mass fraction of CMC was 3%. The weighed materials were placed into an agate mortar, deionized water was added dropwise using a pipette, and the mixture was ground evenly to achieve a slurry of moderate thickness. The obtained slurry was uniformly coated onto copper foil using a 200  $\mu$ m coater. The coated electrode was then placed in a drying oven at 60 °C for 12 h. Finally, the dried electrode was cut into circular electrode sheets with a diameter of 11 mm for subsequent use.

#### **1.5 Electrochemical Testing**

CR2025 coin cells were used for electrochemical measurement. The as-prepared electrode was used as the working electrode and lithium sheet (d=15.6 mm) as the counter electrode. The electrolyte was 1 M LiPF<sub>6</sub> dissolved in the mixture of EC: DEC of (volume ratio 1:1). The separator was Celgard 2400. The coin cells were assembled in a glove box filled with argon to ensure that the oxygen and humidity levels are lower than 0.1 ppm. To test the electrochemical performance, the cells were charged and discharged in the voltage range of 0.001–3 V at room temperature using the Land Battery Test System (Wuhan, China) to measure the cycle stability of the electrodes. Cyclic voltammetry was performed on an electrochemical workstation (CHI660E; Chenhua, China) with a sweep voltage range of 0.001–3 V and a sweep rate of 0.2 mV s<sup>-1</sup>. Electrochemical impedance spectroscopy (EIS) was also performed on the CHI660E electrochemical workstation with a frequency range of  $10^5 \sim 10^{-2}$  Hz.



Fig. S1. SEM images of (a, b) FC, (c, d) FC@CNTs-12 and (e, f) FC@CNTs-48



Fig. S2. Particle size distribution of (a) FC, (b) FC@CNTs@-12, and (c) FC@CNTs@-48.



Fig. S3. TGA results of FC and FC@CNTs.



Fig. S4. XPS spectra of FC and FC@CNTs-24.



Fig. S5. CV curves of (a) FC, (b) FC@CNTs-12, and (c) FC@CNTs- 48 materials at the first cycle.



**Fig. S6.** Charge-discharge curves of (a) FC, (b) FC@CNTs-12, (c) FC@CNTs-24, and (d) FC@CNTs-48 at different cycles.



Fig. S7. (a) EIS spectra of FC and FC@CNTs electrodes and the inset is an equivalent circuit; (b) Comparison histogram of Rct after EIS fitting; (c) Linear relationships between Z' and  $\omega^{-1/2}$ .



Fig. S8. CV curves of (a) FC, (b) FC@CNTs-12, (c) FC@CNTs-24, and (d) FC@CNTs-48 electrode at different scanning rate. The plots of log(i) versus log(v) of (e) FC, (f) FC@CNTs-12, (j) FC@CNTs-24, and (h) FC@CNTs-48 electrode.



Fig. S9. SEM images of the (a-c) FC and (d-f) FC@CNTs-24 after 100 cycles.



Fig. S10. A comparison of our work with previously reported bimetallic oxides.

Sampla	Content			
Sample	FeCo <sub>2</sub> O <sub>4</sub> (%)	FeCo <sub>2</sub> O <sub>4</sub> (%) Fe <sub>2</sub> O <sub>3</sub> (%)	CNTs (%)	
FC	62.6	37.4	-	
FC@CNTs-12	57.2	37.9	4.8	
FC@CNTs-24	50.1	40.8	9.1	
FC@CNTs-48	46.2	36.9	16.7	

 Table S1. Proportion of component content of FC and FC@CNTs.

Table 32. Electrochemical data of FC@CINTS.							
	First	First	100th	100th			
Sampla	discharge	Coulombic	charge	discharge			
Sample	capacity	efficiency	capacity	capacity			
	(mAh g <sup>-1</sup> )	(%)	(mAh g <sup>-1</sup> )	(mAh g <sup>_1</sup> )			
FC	859.2	76.9	164.5	179.7			
FC@CNTs-12	1045.0	75.5	476.9	498.0			
FC@CNTs-24	1469.8	78.1	597.4	609.8			
FC@CNTs-48	1264.2	81.6	511.6	523.0			

Table S2. Electrochemical data of FC@CNTs.

Sample –	Specific capacity (mAh g <sup>-1</sup> )				
	100 mA g <sup>-1</sup>	200 mA g <sup>-1</sup>	500 mA g <sup>-1</sup>	1000 mA g <sup>-1</sup>	
This work	1197	1105	1019	730	
CoFe <sub>2</sub> O <sub>4</sub> (Ref. 1)	545	488	459	394	
CoO/ZnO (Ref. 2)	-	836.7	620.2	398.64	
CoMn <sub>2</sub> O <sub>4</sub> (Ref. 3)	830	680	503	428	
Fe-Mn-O (Ref. 4)	808	722	604	521	

Table S3. Electrochemical data of different anode materials.

# References

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