Encapsulation of Nanoscale Metal Oxides into Ultra-thin Ni Matrix for Superior Li-Ion Batteries: A Versatile Strategy

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1. Experimental Details:

1.1 Preparation of Ni(OH)₂ film: 0.3 g Ni(NO₃)₂·6H₂O and 0.7 g hexamethylenetetramine (HMT) were dissolved in 50 mL distilled water. After 30 min ultrasonication treatment, the transparent suspension was then transferred into a sealed container (80 mL) and held at 95 °C for 6 h. The pure Ni(OH)₂ films can be harvested after wash for several times.

1.2 Preparation of Fe₃O₄@Ni hybrids: Single-crystalline Fe₃O₄ hexagonal nanoplates were synthesized by referring to the previous work. Single-crystalline Fe₃O₄ hexagonal nanoplates were synthesized by referring to the previous work. In brief, a green mixture of FeSO₄·7H₂O (0.4 g), NaOH (0.53 g), ethylene glycol (45 mL) and distilled water (15 mL) was treated by ultrasonication for 30 min, transferred into a Teflon-lined stainless steel autoclave and held at 95 °C for 20 h. The as-formed Fe₃O₄ precipitation was then washed, dried and collected as raw materials for later fabrication. In typical synthesis of Fe₃O₄@Ni hybrids, 0.15 g Fe₃O₄ nanoplates, 0.7 g HMT, 0.3 g Ni(NO₃)₂·6H₂O and 50 mL distilled water were mixed and dissolved by ultrasonication for 15 min. The obtained suspension was then transferred into a sealed container (80 mL) and held at 95 °C for 6 h. Later, the Fe₃O₄@Ni(OH)₂ intermediate was collected and washed by distilled water several times. The reduction procedure was performed in a horizontal, quartz tube-furnace system. 0.3 g Fe₃O₄@Ni(OH)₂ (pre-dispersed into 10 ml distilled water, dropped onto a ceramic wafer and dried at 60 °C) were placed into the centre of a quartz tube (tube diameter: 2 inch). 1.5 mL of ethylene glycol (EG) loaded in an alumina boat was put at the upstream zone of quartz tube. Before heating, the tube reactor was sealed and flushed by Ar gas (200 sccm) for 10 min. The furnace was then heated to 610±10K at a heating rate of ~10 K min⁻¹ under a constant Ar flow of 80 sccm, kept for 15 min and allowed to cool down to room temperature naturally.
1.3 Preparation of C/CoFeO₃@Ni NWAs: The C/CoFeO₃ NWAs were applied directly as raw materials and synthesized according to our previous work. In brief, one piece of Fe-Co-Ni alloy substrate with C/CoFeO₃ NWAs (area: 2.5×3.5 cm²) grown on was immersed into a 80 mL container wherein there was a 50 mL homogeneous aqueous solution containing 0.15 g Ni(NO₃)₂·6H₂O and 0.3 g HMT. The container was then sealed and held at 95 °C for 6 h. The obtained C/CoFeO₃ NWAs were fetched out, rinsed and dried at 60 °C. The next reduction treatment was exactly the same with that of Fe₃O₄@Ni hybrids except that the C/CoFeO₃@Ni(OH)₂ NWAs replaced the above Fe₃O₄@Ni(OH)₂.

2. Figures

Fig. S1 (A-C) SEM and (D) TEM observations on sheet-like ultra-thin Ni(OH)₂ nanofilms. (E) XRD pattern of as-formed Ni(OH)₂ products. Inset in Fig. S1(D) shows their SAED spectrum.
**Fig. S2** XRD patterns showing the evolution process of MnO@Ni core-shell hybrid nanowires.

**Fig. S3** (A-B) TEM and (C) HRTEM images of the MnO$_2$@Ni(OH)$_2$ intermediate.
Fig. S4 (a) EDS spectrum of MnO@Ni core-shell hybrid NWs; (b) EDS spectrum and SEM image (inset) of Ni products evolved from pure Ni(OH)$_2$ thin films.
Fig. S5 (A) XRD patterns monitoring the evolution process of Fe₃O₄@Ni hybrid. SEM observations toward samples within different evolution stages: (B-C) the pristine Fe₃O₄ nanoplates, (D-E) the intermediate Fe₃O₄@Ni(OH)₂ and (F) the final products of Fe₃O₄@Ni hybrid.

Fig. S6 XRD pattern of CoO@Ni core-shell hybrid NWs.
**Fig. S7** SEM observations monitoring the overall evolution of C/CoFeO₅@Ni NWAs: (A-B) the pristine C/CoFeO₅ NWAs, (C-D) the intermediate C/CoFeO₅@Ni(OH)₂ NWAs and (E-F) the final products of C/CoFeO₅@Ni NWAs.

**Fig. S8** (A-B) SEM and (C-D) TEM images of MnO@Ni hybrids after 250 charge-discharge cycles.
**Fig. S10** (A) Cyclic comparison of pure CoFeO₃ NWAs, C/CoFeO₃ NWAs and C/CoFeO₃@Ni NWAs at a constant current density of ~350 mA/g; (B) Charge/discharge profiles of C/CoFeO₃@Ni NWAs in a voltage window of 0.005-3V at ~350 mA/g; (C) Charge/discharge profiles of C/CoFeO₃@Ni NWAs performed under varied current densities; (D) Comparison of rate capabilities on bare CoFeO₃ NWAs, C/CoFeO₃ NWAs and C/CoFeO₃@Ni NWAs.