Supporting Information

In-Grown Structure of NiFe Mixed Metal Oxides and CNT Hybrid Catalyst for Oxygen Evolution Reaction

Yongmei Li\textsuperscript{a}, Haiyong He\textsuperscript{a}, Wei Fu\textsuperscript{a}, Chenzhong Mu\textsuperscript{a}, Xiu-Zhi Tang\textsuperscript{b}, Zheng Liu\textsuperscript{a}, Dongzhi Chi\textsuperscript{c}, Xiao Hu\textsuperscript{a}\textsuperscript{*}

\textsuperscript{a}School of Materials Science and Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798.

\textsuperscript{b}School of Mechanical and Aerospace Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798.

\textsuperscript{c}Institute of Materials Research and Engineering (IMRE), 3 Research Link, Singapore 117602

*Corresponding author: e-mail: asxhu@ntu.edu.sg; Fax: (65) 6790 9081; Tel: (65) 6790 4610.
Experimental Section

Materials

The pristine multi-walled carbon nanotubes (MWNTs) with diameter of 20 nm (purchased from Shenzhen Nanotech Por Co. Ltd) were firstly refluxed in nitric acid (65 wt%) at 140 °C for 6 hours before use. Reagent grade nickel nitrate hexahydrate (Ni(NO$_3$)$_2$·6H$_2$O), iron (III) nitrate nonahydrate (Fe(NO$_3$)$_3$·9H$_2$O), sodium hydroxide (NaOH) and sodium carbonate (Na$_2$CO$_3$) were purchased from Sigma Aldrich and used without further purification.

NiFe LDH synthesis

For NiFe LDH synthesis, precursors i.e., 0.015 M Ni(NO$_3$)$_2$·6H$_2$O and 0.005 M Fe(NO$_3$)$_3$·9H$_2$O, were co-precipitated in an aqueous solution containing 0.01 M Na$_2$CO$_3$. The reaction solution pH was maintained at 10. After all the precursors were added, the reaction solution was sealed and stirred for 24 hours at 80 °C. The as-prepared solid products were then purified with DI water through centrifugation until pH = 7, and finally freeze dried. This sample is denoted as NiFe LDH.

CNT-LDH synthesis

For CNT/NiFe LDH synthesis, 0.1 mg/mL oxidized CNT solution was ultrasonically treated for 6 hours to achieved a good dispersion. After that, 10 mL mixture of 0.015 M Ni(NO$_3$)$_2$·6H$_2$O and 0.005 M Fe(NO$_3$)$_3$·9H$_2$O solution were loaded into CNT solution. The solution was then moved into oil bath of 80 °C and allowed for stirring for 30 min. After that, the pH was tuned to pH 10 by titrating mixture of 1 M NaOH and 0.01 M Na$_2$CO$_3$ solution. The reaction solution was then sealed and stirred for 24 hours at 80 °C. The as-prepared solid products were washed with
DI water through centrifugation until pH = 7, and finally freeze dried. The sample was denoted as CNT-LDH.

**CNT-MMO synthesis**

The CNT-LDH catalyst was annealed in a tube furnace with Ar gas flow. Different temperature setting was used, including 500 °C for 2 hours (denoted as CNT-MMO), 500 °C for 0.5 hours (denoted as CNT-MMO-0.5 hr), and 500 °C for 3 hours (denoted as CNT-MMO-3 hr). NiFe LDH was calcined at 500 °C for 2 hours for comparison study and denoted as NiFe MMO. CNT was also calcined at 500 °C for 2 hours and denoted as Cal CNT. The sample of NiFe MMO + Cal CNT is the physical mixture of NiFe MMO and Cal CNT.

**Characterization**

Morphology of synthesized products were observed using transmission electron microscopy (TEM) (JEOL 2100F), and scanning electron microscopy (SEM) (FESEM 7600F). The scanning transmission electron microscopy (STEM) is done using JEOL 2100F. Powder X-ray diffraction (XRD) was performed by XRD Bruker D8 Powder with Cu-Kα radiation source (λ = 1.54 Å) using measurement step-size of 0.02 ° and scan-rate of 2 °/min. Thermo-gravimetric analysis (TGA) was carried out using TA Q500, which provided information on weight loss during constant-rate heating test (heating rate = 10 °C/min) under atmosphere of N₂ gas. X-Ray Photoelectron Spectroscopy (XPS) were done by VG ESCALAB 220I-XL instrument equipped with a monochromatic Al Kα X-ray source (1486.7 eV photons). The binding energy was calibrated according to C (1s) neutral carbon peak at 285.0 eV.

**Ink preparation for electrochemical measurements**
The preparation method of the working electrode is as following. In brief, 4 mg catalyst was dispersed in a solution consisting of 768 µL water, 200 µL ethanol and 32 µL nafion solution (5 wt%, Sigma Aldrich). The mixture was then ultrasonicated for about 0.5 hr to obtain a homogeneous ink. After that, 5 µL of the dispersion was transfer onto the glassy carbon disk (3 mm in diameter), based on the catalyst loading of about 0.28 mg/cm$^2$. Finally, the as prepared catalyst film was dried at room temperature.

**Electrochemical measurements**

The electrochemical studies were carried out in a standard three electrode system controlled by an autolab electrochemistry workstation. Catalyst powders cast on the glass carbon electrode was used as the working electrode, and rotating at 1600 rpm to get rid of generated oxygen. Pt foil and Ag/AgCl (4 M KCl) were used as the counter electrode and the reference electrode, separately. The reference was calibrated and converted to reversible hydrogen electrode (RHE), $E_{(RHE)} = E_{(Ag/AgCl)} + 0.059pH$. Linear sweep voltammetry was carried out at 5 mV/s for the polarization curve and tafel plot measurement. The hybrid catalyst was cycled ~50 times by cyclic voltammetry (CV) until a stable CV curve was developed before measuring the polarization curves. All polarization curves were corrected with iR compensation. Chronopotentiometry was carried out under a constant current density of 10 mA/cm$^2$. 
Figure S1. Low magnification TEM images of (A) CNT-LDH; and (B) CNT-MMO.
Figure S2. SEM images of (A) CNT-LDH; and (B) CNT-MMO.
Figure S3. Elemental mapping of (A) Ni. (B) Fe. (C) O for catalyst CNT-MMO.
Figure S4. XRD spectra of (i) CNT-MMO-0.5 hr and (ii) CNT-MMO-3 hr.
Figure S5. TEM image of (A) CNT-MMO-0.5 hr; and (B) CNT-MMO-3 hr, the CNT TEM fringes are cut off into different sections, clearly marked by the yellow line, and inorganic nanoparticles as indicated by the red circles are embedded between the broken CNT sections.
Figure S6. Low magnification TEM images of (A) CNT-MMO-0.5 hr; and (B) CNT-MMO-3 hr.
Figure S7. XPS spectrum of (A) C 1s for CNT-LDH, the deconvolution reveals the binding energy for the functional groups of the oxidized CNT; (B) Ni 2p3/2 for CNT-LDH, shows the oxidation state of Ni is 2+; (C) Fe 2p3/2 for CNT-LDH, shows the oxidation state of Fe is 3+; and (D) C 1s of CNT-MMO, the deconvolution reveals 283.6 eV corresponding to Ni$_3$C bonding energy; (E) Ni 2p3/2 of CNT-MMO, the deconvolution reveals NiO and Ni$^0$ binding energy; (F) Fe 2p3/2 for CNT-MMO, the binding energy at 709.9 eV is corresponding to FeO, and 711.9 eV binding energy is corresponding to FeOOH.
Figure S8. (A) and (B) TEM images of CNT-MMO, which show the observation of Ni$_3$C and Ni d-spacing by calculating the lattice fringes.
Figure S9. TEM image of CNT-MMO catalyst, showing the existence of FeOOH nanoparticle, and the size is about 3-5 nm.
Figure S10. The polarization curves for CNT-LDH and CNT-MMO hybrid catalyst under 1M and 0.1M KOH solution without iR correction.
Table S1. Comparison of electrochemical performance of the previous reported relevant works.

<table>
<thead>
<tr>
<th>Catalysts</th>
<th>Onset potential (V vs. RHE)</th>
<th>Tafel slope (mV/dec)</th>
<th>Linear sweep voltammetry for Tafel plot (mV/s)</th>
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