Supporting Information

Ultrathin Vanadium Hydroxide Nanosheets Assembled on the Surface of Ni-Fe Layered Hydroxides as a Hierarchical Catalyst for Oxygen Evolution Reaction

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Fig. S1. (a-b) SEM images of NiFe LDHs/NF; (c-d) SEM images of NFV NSs-8h;

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and (e-f) SEM images of NiFe NFV NSs-12h.
Fig. S2. XPS survey spectra of NFV NSs-4h and NiFe LDHs/NF.
**Fig. S3.** (a) XRD patterns of NFV NSs-4h after long-term OER process; (b-c) SEM images; (d) TEM image, (e) HR-TEM image and (f) SAED patterns of NFV NSs-4h after OER.
Fig. S4. CV curves of (a) NFV NSs-4h, (b) NFV NSs-8h, (c) NFV NSs-12h, (d) NiFe LDHs/NF.
Fig. S5. CVs for NiFe-LDHs and NFV NSs-xh (4,8,12) in faradic capacitance current range at various scan rates: (a) NFV NSs-4h, (b) NFV NSs-8h, (c) NFV NSs-12h, (d) NiFe LDHs/NF.
Table S1. Comparison of the OER performance for the obtained materials in this work with other state-of-the-art OER electrocatalysts.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Electrolyte</th>
<th>$\eta_{j=100}$ (mV)</th>
<th>$\eta_{j=200}$ (mV)</th>
<th>Tafel slope [mV dec$^{-1}$]</th>
<th>Reference</th>
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<tbody>
<tr>
<td>NFV NSs-4h</td>
<td>1 M KOH</td>
<td>280</td>
<td>300</td>
<td>65</td>
<td>This work</td>
</tr>
<tr>
<td>NFV NSs-8h</td>
<td>1 M KOH</td>
<td>300</td>
<td>320</td>
<td>84</td>
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<tr>
<td>NFV NSs-12h</td>
<td>1 M KOH</td>
<td>320</td>
<td>350</td>
<td>112</td>
<td>This work</td>
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<tr>
<td>NiFe LDHs</td>
<td>1 M KOH</td>
<td>310</td>
<td>330</td>
<td>96</td>
<td>This work</td>
</tr>
<tr>
<td>CS-NiFeCu</td>
<td>1 M KOH</td>
<td>~210</td>
<td>~240</td>
<td>33</td>
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<tr>
<td>NiFe$_2$O$_4$</td>
<td>1 M KOH</td>
<td>~210</td>
<td>/</td>
<td>46.4</td>
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<tr>
<td>NiCoFe-LDH HP</td>
<td>1 M KOH</td>
<td>332</td>
<td>~340</td>
<td>56</td>
<td>3</td>
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<tr>
<td>0.1Fe-CoNiO/NF</td>
<td>1 M KOH</td>
<td>~280</td>
<td>/</td>
<td>36.8</td>
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<td>Fe$_3$O$_4$-FeSe/CoSe$_2$</td>
<td>1 M KOH</td>
<td>~350</td>
<td>~370</td>
<td>68.7</td>
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<tr>
<td>Co$<em>{2.4}$Ni$</em>{0.6}$Ge$_2$O$_3$(OH)$_4$</td>
<td>1 M KOH</td>
<td>349</td>
<td>~370</td>
<td>59.8</td>
<td>6</td>
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<tr>
<td>Cu oxide micro/nano-structures</td>
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<td>~350</td>
<td>~370</td>
<td>63</td>
<td>7</td>
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<tr>
<td>NF@NC-CoFe$_2$O$_4$/C NRAs</td>
<td>1 M KOH</td>
<td>~290</td>
<td>~310</td>
<td>45</td>
<td>8</td>
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</table>
**Experimental**

1.1 Materials and chemicals

Ammonia fluoride (NH$_4$F, 96.0%), nickel nitrate hexahydrate (Ni(NO$_3$)$_2$·6H$_2$O, 99.0%) and anhydrous ethanal (CH$_3$CH$_2$OH, 99.7%) were purchased from Sinopharm Chemical Reagent Co., Ltd. Vanadium trichloride (VCl$_3$, 99.0%) and urea (CH$_4$N$_2$O, 99%) were obtained from Aladdin. Iron chloride hexahydrate (FeCl$_3$·6H$_2$O, 99.0%) was supplied by Kelong Chemical Reagent. Nickel foam (NF) (thickness = 0.5 mm) was provided by Guangdong Candlelight New Energy Technology Co., Ltd.

1.2 Synthesis of NiFe LDHs/NF

The NiFe LDHs/NF was fabricated by a one-step hydrothermal method. Prior to the synthesis, NF (2.5 cm x 4 cm) was pretreated ultrasonically in HCl (3.0 M) for 20 minutes to remove the impurities, and then washed it by deionized water and ethanol for several times until pH ~ 7. In a typical process, Ni(NO$_3$)$_2$·6H$_2$O (1 mmol), FeCl$_3$·6H$_2$O (2 mmol), NH$_4$F(40 mmol) and CH$_4$N$_2$O were dissolved in 40 mL deionized water to form a clear solution and then transferred into a 50 mL Teflon-lined
autoclave. A piece of cleaned NF was immersed into the mixture solution. The autoclave was sealed and maintained at 120 °C for 6 h. After cooled down to room temperature, the NiFe LDHs/NF was washed with deionized water several times and dried at 60 °C for 8 hours.

1.3 Synthesis of NiFe LDH/VO(OH)$_2$-xh

VCl$_3$ (1.6 mmol) and CH$_4$N$_2$O (0.3 g) were dissolved in 40 mL deionized water to form a homogeneous solution. Then, the solution was transferred into a 50 mL Teflon-lined autoclave and the NiFe LDHs/NF was completely immersed into the solution. The autoclave was sealed and heated at 120 °C for x hours (x = 4, 8, 12). After cooled to room temperature, the NF with catalyst was washed with deionized water several times and dried at 60 °C for 8 hours. The as-obtained catalysts are denoted as NiFe LDH/VO(OH)$_2$-xh (NFV NSs-xh; x = 4, 8, 12).

1.4 Materials characterization

The morphology of the samples was observed by scanning electron microscope (SEM, S-4800, Japan) and transmission electron microscopy (TEM, G2F20, USA). The crystal structure and chemical composition of the samples was analyzed by X-ray diffraction (XRD, Smart Lab) and X-ray photoelectron spectroscopy (XPS, PHI 5000), respectively.
1.5 Electrochemical measurements

The electrochemical properties of the materials were evaluated in a three-electrode system on a CHI-660E electrochemical workstation (Chenhua instrument co., LTD., Shanghai) at room temperature. In a standard three-electrode system, the NF (0.5 cm × 0.5 cm) with different samples was directly used as working electrode, then a Hg/HgO electrode and a graphite rod were acted as the reference electrode and the counter electrode, respectively. All measured potentials were calibrated to the reversible hydrogen electrode (RHE) based on the Nernst equation: \( E_{\text{RHE}} = E_{\text{Hg/HgO}} + 0.059 \times \text{pH} + 0.098 \). The steady-state linear sweep voltammetry (LSV) curves were obtained at a scan rate of 2 mV s\(^{-1}\) in 1.0 M KOH solution. The measurements of electrochemical impedance spectroscopies (EIS) were conducted at a frequency range from 10 KHz to 100 mHz by applying an alternating current (AC) voltage with 10 mV amplitude. The chronoamperometric test (40 h) of NFV NSs-4h for OER was carried out at a current density of 100 mA cm\(^{-2}\) and cyclic voltammetry (CV) tests of 1000 cycles were conducted at a scan rate of 100 mV s\(^{-1}\) to investigate the durability of the sample. In
order to determine the electrochemical active surface area (ECSA), the double layer capacitance (C_{dl}) of the electrode can be obtained by carrying out the CV measurement and the ECSA can be calculated by C_{dl} using the formula: ECSA = C_{dl} / C_s (C_s is assuming as 0.040 mF cm^{-2}).

References


Liu, R. Ma and T. Qiu, Applied Catalysis B: Environmental, 2020, 260.

