Electronic Supplementary Information for

*In situ* construction of pollen-petal-like heterostructured Co$_3$O$_4$-CeO$_2$

on 3D FeNi$_3$ foam as bifunctional catalyst for overall water splitting†

Maosheng Liu‡, Tao Chen‡*, Wenxiu Zhang*, Shuang Wei, Yujun Cheng*, Jingquan Liu**

*a College of Materials Science and Engineering; Institute for Graphene Applied Technology Innovation, College of Life Sciences, Qingdao University, 266071, China.
b College of Life Sciences, Qingdao University, Qingdao 266071, China.
c College of Materials Science and Engineering, Ocean University of China, Qingdao 266100, China.

*Authors for Correspondence: Dr Tao Chen and Prof. Jingquan Liu
Fax/Tel: 86-532-83780128
E-Mail: 382388316@qq.com (T. Chen), jliu@qdu.edu.cn (J. Q. Liu)
† These authors contributed equally.
1. Experimental section

1.1 Characterizations

The surface morphologies of the synthesized samples were characterized by scanning electron microscopy (SEM, ZEISS Supra 55) at an accelerating voltage of 10 kV. Transmission electron microscopy images (JEOL JEM-2100F) of the synthesized samples were collected on a JEOL JEM-2100F microscope. The compositions of the synthesized samples were specifically analyzed by high resolution transmission electron microscopy (HRTEM). Selected area electron diffraction (SAED) analyses for the sample were performed at 200 kV on a FEI-Tecnai G2 F20 transmission electron microscope. The phase composition analyses of the synthesized samples were carried out on a Bruker D8 advanced X-ray powder diffractometer with Cu Ka radiation ($\lambda = 1.54056 \text{ Å}$). The analyses of the specific elemental composition of the synthesized samples were carried out by X-ray photoelectron spectroscopy (XPS) on a Thermo ESCALAB 250Xi using a single-color Al Ka X-ray as the excitation source. The electron paramagnetic resonance (EPR) spectra were recorded on a Bruker EMX Plus spectrometer operating at X-band frequency (9.4218 GHz) equipped with 100 kHz field modulation and phase sensitive detection to obtain the first derivative signal. The specific element contents of the samples were measured on an Agilent 720ES inductively coupled plasma emission spectrometer (ICP-OES). The active catalyst was exfoliated from the substrate via a strong and long-time sonication process and then weighed. Then, nitric acid was applied to completely dissolve the sample to obtain the $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF solutions with different concentrations, which were analyzed by ICP-OES. Electrochemical measurements for the synthesized materials were carried out using a standard three-electrode system with a CHI-760D electrochemical workstation (CHI Instruments, Shanghai, China), using platinum foil as counter electrode and saturated calomel electrode as reference electrode (SCE).
1.2 Materials

All chemical reagents used in the experiments were of analytical grade. Cobaltous (II) nitrate (Co(NO$_3$)$_2$·6H$_2$O, AR), Nafion (5 wt %), ammonium fluoride (NH$_4$F, AR) and potassium hydroxide (KOH, AR) were purchased from Aladdin Ltd. (Shanghai, China). Ammonium cerium (IV) nitrate (Ce(NH$_4$)$_2$(NO$_3$)$_6$, AR) and urea (CO(NH$_2$)$_2$, AR) were brought from Shandong Laiyang Economic and Technological Development Zone Chemical Plant (Laiyang, China). Pt/C (10 wt % Pt) was purchased from Sinopharm Chemical Reagent Co, Ltd (Shanghai, China). FeNi$_3$ foam (FNF) was brought from Suzhou Taili Foam Metal Factory and cleaned by sonication sequentially in water and ethanol several times. Unless otherwise stated, all the above chemicals were used as received without further purification. Deionized water was generated from our lab-owned Flom water purification system and used to prepare all the solutions in this study.

1.3 Preparation of Co$_3$O$_4$-CeO$_2$@FNF and Co$_3$O$_4$-CeO$_2$ nanohybrids

Co$_3$O$_4$-CeO$_2$@FNF and Co$_3$O$_4$-CeO$_2$ nanohybrids were prepared by the following processes: First, Co(NO$_3$)$_2$·6H$_2$O (0.146 g, 0.5 mmol), Ce(NH$_4$)$_2$(NO$_3$)$_6$ (0.274 g, 0.5 mmol), CO(NH$_2$)$_2$ (0.12 g, 2 mmol) and NH$_4$F (0.037 g, 1 mmol) were added to 40 mL deionized water and then stirring for 25 min to form homogeneous solution. The solution and a piece of FNF (1 cm × 4 cm) were then transferred into a 50 mL Teflon-lined stainless steel autoclave. Then the sealed autoclave was transferred into an oven and maintained at 180 °C for 6 h. After that, the as-prepared Co$_3$O$_4$-CeO$_2$@FNF was taken out from the naturally cooled autoclave and then thoroughly washed with deionized water as well as ethanol several times, respectively, then dried for 6 h at 60 °C in air. Second, the generated precipitate (Co$_3$O$_4$-CeO$_2$ nanohybrids) was then centrifuged and separated by washing with a mixture of ethanol and water for several times. The product was dried in a vacuum oven at 60 °C for 12 h. Moreover, in order to further explore the effect of cobalt-cerium ion pairing on electrocatalytic
performance, the input amount of Ce(NH$_4$)$_2$(NO$_3$)$_6$ was adjusted to obtain Co$_3$O$_4$-CeO$_2$@FNF with different ratios of cobaltous to cerium (Co : Ce = 1 : 0.5, 1 : 2 and 1 : 3).

1.4 Synthesis of Co$_3$O$_4$@FNF and Co$_3$O$_4$ NPs

The synthesis of Co$_3$O$_4$@FNF was similar to that of Co$_3$O$_4$-CeO$_2$@FNF just without the addition of Ce(NH$_4$)$_2$(NO$_3$)$_6$. The generated precipitate (Co$_3$O$_4$ NPs) was centrifuged and then washed with a mixture of ethanol and water for several times to afford the expected product, which was dried in a vacuum oven at 60 °C for 12 h for the following usage.

1.5 Synthesis of CeO$_2$@FNF and CeO$_2$ NPs

The synthesis of CeO$_2$@FNF was similar to that of Co$_3$O$_4$-CeO$_2$@FNF just without the addition of Co(NO$_3$)$_2$·6H$_2$O. The generated precipitate (CeO$_2$ NPs) was centrifuged and then washed with a mixture of ethanol and water for several times to afford the expected product, which was dried in a vacuum oven at 60 °C for 12 h for the following usage.
1.6 Electrochemical measurements

When the samples were tested for HER and OER in 1.0 M KOH solutions using linear sweep voltammetry (LSV) technique, the scan rates were all controlled at 5 mV s \(^{-1}\). The potentials measured can be converted into a reversible hydrogen electrode (RHE) scale by the following formula: 
\[ E_{\text{RHE}} = E_{\text{SCE}} + E_{0\text{SCE}}^0 + 0.059 \, \text{pH} \]
where \( E_{\text{SCE}} \) is the experimentally measured potential and \( E_{0\text{SCE}}^0 = 0.242 \, \text{V} \). In addition, iR compensation was applied for all electrochemical measurements to eliminate the effect of ohmic resistance on the as-measured reaction currents obtained from the tests. All the measurements were carried out at ambient temperature.

1.7 Preparation of Pt/C and RuO\(_2\) electrodes

10 mg of Pt/C was mixed with 450 \( \mu \text{L} \) of deionized water, 450 \( \mu \text{L} \) of absolute ethanol and 20 \( \mu \text{L} \) of Nafion (5 wt%), and the resulting mixture was sonicated for 1200 minutes to obtain Pt/C ink. RuO\(_2\) ink was prepared using the same method. Then using a pipette to draw 5 \( \mu \text{L} \) ink each time, which was carefully dropped onto the FeNi\(_3\) foam (FNF) and then dried naturally at room temperature. The total ink added was 100 \( \mu \text{L} \), which was dropped in 20 times for the coating. Using this method, Pt/C electrodes and RuO\(_2\) electrodes were prepared separately. The loading of Pt/C and RuO\(_2\) on FNF are 4.37 mg cm\(^{-2}\).

1.8 Electrochemical impedance spectroscopy (EIS)

In order to perform a more comprehensive investigation for the materials, the EIS tests of the electrode materials were carried out under the operating conditions of HER. A fixed voltage is applied to each material and the test was performed in a scanning frequency range from 100 kHz to 10 MHz.
1.9 Effective electrode surface area (ECSA) calculation

In order to fully demonstrate the excellent properties of the material, the ECSA of the material was estimated. In 1.0 M KOH electrolyte solution, the material was subjected to 10 CV cycles in a voltage window from -0.13V to -0.23V vs. RHE. The test was carried out to obtain CVs of the material. The electrochemical double layer capacitor \( (C_{dl}) \) of the material under the non-Faradaic overpotential, it can be obtained by CV at different scanning rates (10, 20, 30, 40 and 50 mV s\(^{-1}\)). The difference in current density between the anodic and cathodic sweeps is linear with the scan rate and the slope is then obtained by data fitting, which is twice the \( C_{dl} \). Then the ECSA of the material can be estimated by Eqn. S1:

\[
ECSA = \frac{C_{dl}}{C_s} \quad \text{Eqn. S1}
\]

where \( C_s \) is the specific capacitance, whose value is reported to be 0.040 mF cm\(^{-2}\) in 1.0 M KOH.\(^2\)
1.10 Turnover frequency (TOF) calculations

During the CV scanning of the electrochemical test, it can be observed that during the OER process, the current difference between the oxidation peak and the reduction peak of the redox species is linear with the scan rate. The slope of the line can be calculated according to Eqn. S2:

\[
\text{Slope} = \frac{(n^2 F^2 C)}{(4 R T)} \quad \text{Eqn. S2}
\]

where \( n \) is the number of transferred electrons, \( F \) is the Faraday’s constant, \( s \) is the area of the electrode, \( C \) is the surface concentration of the active site of the electrode (mol cm\(^2\)), \( R \) is the ideal gas constant, and \( T \) is the absolute temperature.\(^3\)

Wherein \( m \) is derived by Eqn. S2, so that the TOF value can be calculated according to Eqn. S3:

\[
\text{TOF} = \frac{(j s)}{(4 F m)} \quad \text{Eqn. S3}
\]

Where \( j \) represents the current density at a certain overpotential, \( s \) is the area of the electrode, \( 4 \) represents the number of moles of electrons consumed by electrolysis of 1 mol of \( \text{O}_2 \) from water, \( F \) is the Faraday constant, and \( m \) is the number of moles of the active site of the electrode.\(^4\)

1.11 Calculation of the activity retention proportions

\( A_{ct} \) is used to represent the activity retention ratio. The activity retention ratio of the catalyst after a long-term stability test was calculated by the formula \( A_{ct} = 1 - \frac{\Delta E}{E_0} \). Where, \( E_0 \) is used to indicate the initial overpotential and \( E_1 \) to represent the terminate overpotential. The potential difference between \( E_0 \) and \( E_1 \) is \( \Delta E \) (\( \Delta E = E_1 - E_0 \)).
2. Results and Discussion

Fig. S1. Digital photographs of FNF (left) and Co$_3$O$_4$-CeO$_2$@FNF (right).
**Fig. S2.** (A-B) SEM images of FNF at different magnifications.
Fig. S3. (A-C) SEM images of Co$_3$O$_4$@FNF. (D) EDS elemental mapping images for O, Fe, Ni and Co elements of Co$_3$O$_4$@FNF. (E) EDX spectrum of Co$_3$O$_4$@FNF. (F) XRD spectrum of Co$_3$O$_4$ NPs.
**Fig. S4.** (A-C) SEM images of CeO$_2$@FNF. (D) EDS elemental mapping images for O, Fe, Ni and Ce elements of CeO$_2$@FNF. (E) EDX spectrum of CeO$_2$@FNF. (F) XRD spectrum of CeO$_2$ NPs.
Fig. S5. (A-B) TEM images of heterostructured $\text{Co}_3\text{O}_4$-$\text{CeO}_2$ nano-hybrids at different magnifications.
As shown in Fig. S5A, the overall spectrum indicated the existence of Fe, Ni, Co and O in the obtained samples. The two characteristic peaks observed in Fig. S5B at 855.4 eV and 873.1 eV should correspond to Ni$_{3/2}$ and Ni$_{1/2}$, respectively, while two satellite peaks at 861.1 eV and 879.4 eV belong to the rocking peak of Ni. Moreover, the existence of Fe element was strongly confirmed by XPS characterization as shown in Fig. S5C. Besides, there are two distinct peaks in the Fe 2p curve after fitting, which are located at 713.1 eV and 721.8 eV, respectively, indicating the presence of Fe. What’s more, the peaks appearing at 782.6 eV and 798.2 eV were assigned to Co 2p$_{3/2}$ and Co 2p$_{1/2}$, respectively (Fig. S5D). These results indicated that the Co$_3$O$_4$@FNF has been successfully prepared.
As shown in Fig. S6A, the overall spectrum indicated the existence of Fe, Ni, Ce and O in the characterized samples. The two characteristic peaks observed in Fig. S6B at 856.2 eV and 873.4 eV should correspond to Ni\textsubscript{3/2} and Ni\textsubscript{1/2}, respectively, while two satellite peaks at 861.9 eV and 881.7 eV belong to the rocking peak of Ni. Moreover, the following XPS characterization shown in Fig. S6C further confirmed the existence of Fe element. Two distinct peaks in the Fe 2p curve after fitting located at 712.3 eV and 719.2 eV, respectively, indicated the presence of Fe element. Furthermore, as shown in Fig. S6D, the peaks appearing at 879.1 eV and 882.6 eV were assigned to Ce\textsuperscript{4+} and Ce\textsuperscript{3+} 3d\textsubscript{5/2}, respectively. Moreover, those at 898.9 eV and 918.0 eV were attributed to Ce\textsuperscript{4+} 3d\textsubscript{3/2}, whereas the peaks located at 902.2 eV and 918.6 eV were assigned to Ce\textsuperscript{3+} 3d\textsubscript{3/2}. All these results indicated that the CeO\textsubscript{2}@FNF has been successfully prepared.
Fig. S8. Room temperature EPR spectra of (A) Co$_3$O$_4$, (B) CeO$_2$ and (C) Co$_3$O$_4$-CeO$_2$. 
**Fig. S9.** (A-C) SEM images of Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 0.5). (D-F) SEM images of Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 2). (G-I) SEM images of Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 3).
Fig. S10. Electrochemical HER properties of the electrocatalysts in 1.0 M KOH solution. (A) Polarization curves of Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 0.5 ; 1 : 1 ; 1 : 2 ; 1 : 3) for the HER at a scan rate of 5 mV s$^{-1}$. (B) Potentials corresponding to different current densities (10, 30, 50 mA cm$^{-2}$) and (C) Tafel plots of Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 0.5 ; 1 : 1 ; 1 : 2 ; 1 : 3).

The HER catalytic property of Co$_3$O$_4$-CeO$_2$@FNF prepared in different ratios of cobaltous and cerium (Co : Ce = 1 : 0.5, 1 : 1, 1 : 2, 1 : 3) were tested in 1.0 M KOH alkaline media. Fig. S8A indicated that Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 1) showed the best catalytic performance via contrasting the polarization curves. In order to acquire the current density of 10 mA cm$^{-2}$, Co$_3$O$_4$-CeO$_2$@FNF prepared in different ratios of cobaltous and cerium (Co : Ce = 1 : 0.5, 1 : 2, 1 : 3) required the overpotential of 122, 76 and 70 mV, respectively, which were much higher than that of Co$_3$O$_4$-CeO$_2$@FNF (53.8 mV, Fig. S8B), it was obvious that Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 1) exhibited the highest catalytic activity among the tested samples. In order to analyze the HER kinetics of the electrocatalysts, the Tafel slope values were also calculated for assessing the property of the electrocatalyst calculated by the Tafel slope equation from the corresponding LSV curves as shown in Fig. S8A. It could be observed from Fig. S8C that the Tafel slope values of Co$_3$O$_4$-CeO$_2$@FNF prepared in different ratios of cobaltous and cerium (Co : Ce = 1 : 0.5, 1 : 1, 1 : 2, 1 : 3) were 114, 67, 90 and 76 mV dec$^{-1}$, respectively.
Fig. S11. CVs for (A) Co$_3$O$_4$@FNF and (B) CeO$_2$@FNF at scan rates of 10, 20, 30, 40 and 50 mV s$^{-1}$. (C) The ECSA values of Co$_3$O$_4$-CeO$_2$@FNF, Co$_3$O$_4$@FNF and CeO$_2$@FNF.
Fig. S12. CV curves for the (A) $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF (Co : Ce = 1 : 0.5), (C) $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF (Co : Ce = 1 : 2) and (E) $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF (Co : Ce = 1 : 3) catalysts measured at the scan rates of 10, 20, 30, 40 and 50 mV s$^{-1}$. Calculated electrochemical double-layer capacitances ($C_{dl}$) values for the (B) $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF (Co : Ce = 1 : 0.5), (D) $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF (Co : Ce = 1 : 2) and (F) $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF (Co : Ce = 1 : 3).
Fig. S13. Electrochemical OER properties of the electrocatalysts in 1.0 M KOH solution. (A) Polarization curves of Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 0.5 ; 1 : 1 ; 1 : 2 ; 1 : 3) for the HER at a scan rate of 5 mV s$^{-1}$. (B) Potentials corresponding to different current densities (10, 30, 50 mA cm$^{-2}$) and (C) Tafel plots of Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 0.5 ; 1 : 1 ; 1 : 2 ; 1 : 3).

The OER catalytic property of Co$_3$O$_4$-CeO$_2$@FNF prepared in different ratios of cobaltous and cerium (Co : Ce = 1 : 0.5, 1 : 1, 1 : 2, 1 : 3) were tested in 1.0 M KOH alkaline media. Fig. S11A indicated that Co$_3$O$_4$-CeO$_2$@FNF (Co : Ce = 1 : 1) showed the best catalytic performance via contrasting the polarization curves. In order to acquire the current density of 10 mA cm$^{-2}$, Co$_3$O$_4$-CeO$_2$@FNF prepared in different ratios of cobaltous and cerium (Co : Ce = 1 : 0.5, 1 : 2, 1 : 3) required the overpotential of 248, 265 and 267 mV, respectively, which were much higher than that of Co$_3$O$_4$-CeO$_2$@FNF(226 mV, Fig. S11B), it was obvious that Co$_3$O$_4$-CeO$_2$@FNF(Co : Ce = 1 : 1) exhibited the highest catalytic activity contras among the tested samples. In order to analyze the OER kinetics of the electrocatalysts, the Tafel slope values were also calculated for assessing the property of an electrocatalyst calculated by the Tafel slope equation from the corresponding LSV curves as shown in Fig. S11A. It could be observed from Fig. S11C that the Tafel slope values of Co$_3$O$_4$-CeO$_2$@FNF prepared in different ratios of cobaltous and cerium (Co : Ce = 1 : 0.5, 1 : 1, 1 : 2, 1 : 3) were 147, 74, 132 and 86 mV dec$^{-1}$, respectively.
Fig. S14. Cyclic voltammogram curves of (A) Co$_3$O$_4$@FNF, (B) CeO$_2$@FNF, (C) Co$_3$O$_4$-CeO$_2$@FNF under different scan rates in 1.0 M KOH solution. (D) The values of TOF of Co$_3$O$_4$@FNF, CeO$_2$@FNF and Co$_3$O$_4$-CeO$_2$@FNF.
Fig. S15. The TOF values of $\text{Co}_3\text{O}_4$@FNF, $\text{CeO}_2$@FNF and $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF.
**Fig. S16.** Chronoamperomeric curve obtained with Co$_3$O$_4$-CeO$_2$@FNF electrode in 1.0 M KOH, and the potential applied on the working electrode is changed intermittently between −0.13 V and 1.43 V versus RHE every 10 min.
Fig. S17. SEM images of Co$_3$O$_4$-CeO$_2$@FNF at different magnifications after the stability test in a long-term for 70 h.
Table S1: The comparison of catalytic performances for HER in 1M KOH between Co$_3$O$_4$-CeO$_2$@FNF and other materials reported in the literature.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Overpotential (mV)</th>
<th>Tafel slope (mV dec$^{-1}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co$_3$O$_4$-CeO$_2$@FNF</td>
<td>53.8@10</td>
<td>67</td>
<td>This work</td>
</tr>
<tr>
<td>Co$_3$O$_4$ NCs</td>
<td>155@10</td>
<td>115</td>
<td>5$^5$</td>
</tr>
<tr>
<td>Co$_3$O$_4$-MTA</td>
<td>170@10</td>
<td>98</td>
<td>6$^6$</td>
</tr>
<tr>
<td>Co/CoP</td>
<td>138@10</td>
<td>64</td>
<td>7$^7$</td>
</tr>
<tr>
<td>Ni/CeO$_2$-CNT</td>
<td>91@10</td>
<td>----</td>
<td>8$^8$</td>
</tr>
<tr>
<td>Ni$_3$N-CeO$_2$@Ti</td>
<td>80@10</td>
<td>122</td>
<td>9$^9$</td>
</tr>
<tr>
<td>CoP/NiCoP@Ti</td>
<td>133@10</td>
<td>88</td>
<td>10$^{10}$</td>
</tr>
<tr>
<td>Co(OH)$_2$/PANI</td>
<td>90@10</td>
<td>91.6</td>
<td>11$^{11}$</td>
</tr>
<tr>
<td>MoO$_2$@MoN/NF</td>
<td>152@10</td>
<td>98</td>
<td>12$^{12}$</td>
</tr>
<tr>
<td>NF@</td>
<td>154@10</td>
<td>101</td>
<td>13$^{13}$</td>
</tr>
<tr>
<td>NiFe LDH/CeOx</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table S2 The comparison of catalytic performances for OER in 1M KOH between $\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF and other materials reported in the literature.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Overpotential (mV)</th>
<th>Tafel slope (mV dec$^{-1}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{Co}_3\text{O}_4$-$\text{CeO}_2$@FNF</td>
<td>236@10</td>
<td>74</td>
<td>This work</td>
</tr>
<tr>
<td>CeOx/CoOx</td>
<td>313@10</td>
<td>66</td>
<td>14$^{14}$</td>
</tr>
<tr>
<td>N-CG-CoO</td>
<td>340@10</td>
<td>71</td>
<td>15$^{15}$</td>
</tr>
<tr>
<td>Co$_3$O$_4$/NiCo$_2$O$_4$ DSNCs</td>
<td>340@10</td>
<td>88</td>
<td>16$^{16}$</td>
</tr>
<tr>
<td>Co$_3$O$_4$@C-MWCNTs</td>
<td>320@10</td>
<td>62</td>
<td>17$^{17}$</td>
</tr>
<tr>
<td>CC@Co$_3$O$_4$</td>
<td>380@10</td>
<td>75</td>
<td>18$^{18}$</td>
</tr>
<tr>
<td>Co(OH)$_2$@GC</td>
<td>380-400@10</td>
<td>90</td>
<td>19$^{19}$</td>
</tr>
<tr>
<td>NiCoFe-LDH@GC</td>
<td>340@10</td>
<td>93</td>
<td>20$^{20}$</td>
</tr>
</tbody>
</table>
Table S3 Comparison of electrocatalytic performance of Co$_3$O$_4$-CeO$_2$@FNF with various bifunctional electrocatalysts reported recently for overall water splitting.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>$J$ (mA cm$^{-2}$)</th>
<th>$\eta$ (mV)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co$_3$O$_4$-CeO$_2$@FNF</td>
<td>10</td>
<td>1.59</td>
<td>This work</td>
</tr>
<tr>
<td>Co-doped NiSe$_2$@Ti</td>
<td>10</td>
<td>1.62</td>
<td>21$^{21}$</td>
</tr>
<tr>
<td>NiFe/NiCo$_2$O$_4$@N i foam</td>
<td>10</td>
<td>1.67</td>
<td>22$^{22}$</td>
</tr>
<tr>
<td>Co-B@CoO@Ti</td>
<td>50</td>
<td>1.67</td>
<td>23$^{23}$</td>
</tr>
<tr>
<td>NiFeRu-LDH@NF</td>
<td>10</td>
<td>1.52</td>
<td>24$^{24}$</td>
</tr>
<tr>
<td>NiCoP</td>
<td>10</td>
<td>1.64</td>
<td>25$^{25}$</td>
</tr>
<tr>
<td>FeCoP</td>
<td>10</td>
<td>1.60</td>
<td>26$^{26}$</td>
</tr>
</tbody>
</table>
3. References

11. X. Wang, Y. Yang, L. Diao, Y. Tang and N. Zhao, ACS Applied Materials Interfaces, 2018, 10, 3514-35153.
