Electronic Supplementary Information

A Heterostructured FeNi Hydroxide for Effective Electrocatalytic Oxygen Evolution

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3. Reference
1. Experimental Procedures

1.1. Materials

FeCl$_3$·6H$_2$O (≥ 99.0%, Shanghai Titan Scientific Co., Ltd.), NiCl$_2$·6H$_2$O (≥ 99.0%, Shanghai Titan Scientific Co., Ltd.), CoCl$_2$·6H$_2$O (≥ 99.0%, Shanghai Titan Scientific Co., Ltd.), gelatin (C$_{102}$H$_{151}$N$_{31}$O$_{39}$, G7041-500G, Sigma-Aldrich), urea (CO(NH$_2$)$_2$, 99.0%, Shanghai Titan Scientific Co., Ltd.) were used as received without further purification. Deionized (DI) water was used in all experiments.

1.2. Synthesis

1.2.1 Synthesis of FeNi/Ni HS

A solution was prepared by dissolving FeCl$_3$·6H$_2$O (2.16 g, 8 mmol) and NiCl$_2$·6H$_2$O (2.85 g, 12 mmol) in 40 mL of DI water with sonication, to which an aqueous solution of gelatin (8 mL) was prepared by dissolving 1 g of gelatin in 9 mL of DI water at 85 ℃) was slowly added over 15 min with sonication, followed by the addition of 8 g of urea. The resulting mixture was sonicated for another 20 min till a clear solution was obtained. The reaction continued at 100 ℃ for 36 h. The reaction mixture was then cooled to room temperature and then centrifuged at 8000 rpm for 5 min. The precipitate was collected and washed with absolute ethanol and DI water three times and dried at 60 ℃ in a vacuum oven.

1.2.2. Synthesis of FeOOH

The synthetic procedure of FeOOH was similar to that of FeNi/Ni HS, except that with the use of FeCl$_3$·6H$_2$O (2.70 g, 10 mmol), DI water (20 mL), and urea (3 g).

1.2.3. Synthesis of Ni$_2$(CO$_3$)(OH)$_2$

The synthetic procedure of Ni$_2$(CO$_3$)(OH)$_2$ was similar to that of FeNi/Ni HS, except that with the use of NiCl$_2$·6H$_2$O (2.38 g, 10 mmol), DI water (20 mL), and urea (3 g).

1.2.4. Synthesis of the Physical Mixture of FeOOH and Ni$_2$(CO$_3$)(OH)$_2$
The physical mixture was prepared by mixing FeOOH (0.71 g, 8 mmol) and Ni$_2$(CO$_3$)(OH)$_2$ (1.27 g, 6 mmol).

1.2.5. Synthesis of Ni-doped FeOOH

Ni-doped FeOOH was obtained by reacting according to the synthesis conditions of FeNi/Ni HS for 3h.

1.2.6. Synthesis of Co(CO$_3$)$_{0.5}$(OH)

The synthetic procedure of Co(CO$_3$)$_{0.5}$(OH) was similar to that of FeNi/Ni HS, except that with the use of CoCl$_2$·6H$_2$O (2.38 g, 10 mmol), DI water (20 mL), and urea (3 g).

1.2.7. Synthesis of the Physical Mixture of FeOOH and Co(CO$_3$)$_{0.5}$(OH)

The physical mixture was prepared by mixing FeOOH (0.89 g, 10 mmol) and Co(CO$_3$)$_{0.5}$(OH) (1.06 g, 10 mmol).

1.2.8. Synthesis of Co-doped FeOOH

Co-doped FeOOH was obtained by reacting according to the synthesis conditions of FeNi/Ni HS for 3h.

1.2.9. Synthesis of FeCo/Co HS

The synthetic procedure of FeCo/Co HS was similar to that of FeNi/Ni HS, except that with a mixture of FeCl$_3$·6H$_2$O (2.70 g, 10 mmol) and CoCl$_2$·6H$_2$O (2.38 g, 10 mmol).

1.3. Characterization

The field emission scanning electron microscopy (SEM) images were taken on a Merlin scanning electron microscopy with an accelerating voltage of 5 kV. Transmission electron microscope (TEM) images were acquired on a Hitachi HT-7700 (Japan) transmission electron microscope with an accelerating voltage of 100 kV. Atomic-resolution high-angle annular dark field-scanning transmission electron microscopy (HAADF-STEM) images, energy-dispersive X-
ray spectroscopy (EDX), and electron energy loss spectroscopy (EELS) characterizations were performed with a Titan Themis G2 operating at 300 kV.

X-ray diffraction (XRD) patterns of the dried samples were measured in a Rigaku D/Max-RB (Japan) X-ray diffractometer with Cu-Kα radiation (λ=1.5406 Å). X-ray photoelectron spectroscopy (XPS) was recorded on a PHI 5000 Versa Probe III with Al Kα (hv=1486.6 eV) as the excitation source. With reference to C 1s to 284.6 eV, the binding energy obtained in XPS spectra analysis was corrected. Raman spectra of powder samples were recorded on a Renishaw Invia Raman Microscope with a laser excitation wavelength of 532 nm. In-situ Raman tests for catalysts were conducted in a homemade electrochemical cell under controlled potentials. The concentration of catalysts was determined by inductively coupled plasma mass spectroscopy (ICP-MS) (Agilent 7700x).

1.4. Electro Catalytic Research

1.4.1. Electro Catalytic Measurements

electrochemical measurements of OER performance were performed using the CHI 760E Electrochemical Workstation (rotating disk electrode (RDE), electrode area: 0.07 cm²) with a standard three-electrode system, the RDE as the working electrode, carbon rode as the counter electrode, and a saturated Ag/AgCl electrode (in saturated KCl solution) as the reference electrode. The potentials values used in this study were calculated according to the following equation: \( E_{(vs. \text{RHE})} = E_{\text{Ag/AgCl}} + 0.197 \ V + 0.0591 \ \text{pH} \). Overpotentials (\( \eta \)) were calculated based on the formula \( \eta = E_{(vs. \ \text{RHE})} - 1.23 \ V \). Current density (\( j \)) was calculated by dividing the current by the electrode area. The working electrode was prepared according to the following methods. Typically, 4 mg of as-prepared catalyst and 1 mg of activated carbon powder were dispersed in 480 μL deionized (DI) water, 480 μL ethanol, and 40 μL Nafion mixture solution. Then the mixture solution was sonicated for 2 h to form a homogeneous ink. After that, 5 μL of the dispersion was loaded onto the RDE. All the loading mass of the
catalysts on the RDE was about 0.028 mg cm$^{-2}$. Electrocatalytic performances were carried out in 1.0 M KOH. The working electrode was performed with a rotation rate of 1600 rpm. The electrochemical impedance spectroscopy (EIS) measurements were carried out in the same configuration at 1.32 V (vs. RHE) from 0.1 Hz to 100 kHz.

The electrical double layer capacitance ($C_{dl}$) of the samples was measured from double-layer charging curves using cyclic voltammograms (CVs) in a potential range of 1.02-1.12 V in 1.0 M KOH at the scan rate of 10, 20, 30, 40, 50, 60, 70, and 80 mV s$^{-1}$.

1.4.2. Calculation of ECSA-normalized Current Density

The electrochemical surface area (ECSA) is estimated from $C_{dl}$ of the catalyst. The ECSA-normalized current density ($j_{ECSA}$) is calculated as follows:

$$\text{ECSA} = \frac{C_{dl}}{C_s}$$

$$j_{ECSA} = \frac{j}{\text{ECSA}}$$

where $C_s$ is the specific capacitance (0.04 mF cm$^{-2}$ in this work). While $j$ is the current (mA).$^{1,2}$

1.4.3. Calculation of Mass Activity and TOF

The mass activity (A/g) values were calculated from the catalyst loading $m$ (0.28 mg cm$^{-2}$) and the measured current density $j$ (mA cm$^{-2}$) at $\eta = 268$ mV.

$$\text{Mass activity} = \frac{j}{m}$$

The TOF values were calculated by assuming that every metal atom is involved in the catalysis (lower TOF limits were calculated):

$$\text{TOF} = \frac{jS}{4Fn}$$
Where \( j \) (mA cm\(^{-2}\)) is the measured current density at \( \eta = 268 \) mV, \( S \) (0.07 cm\(^2\)) is the surface area of the glassy carbon electrode, the constant 4 means 4 electrons/mol of \( O_2 \), \( F \) is Faraday’s constant (96485.30 C/mol), and \( n \) is the mole of coated metal atom on the electrode calculated from \( m \), the molecular weight of the coated catalysts.

### 1.4.4. Measurement of the Faradic Efficiency

The Faradic efficiency for OER was determined by a rotating ring-disk electrode (RRDE) consisting of a glassy carbon disk electrode and a Pt ring electrode. A constant current of 200 \( \mu \)A was applied to the disk electrode, and the ring electrode was held constant at 0.40 V vs RHE to reduce \( O_2 \) generated at the disk. The Faradic efficiency \( (f) \) is calculated using the following equation:

\[
    f = \frac{I_r}{N I_d}
\]

Where \( I_d \) is the disk current and \( I_r \) is the ring current. \( N \) is the current collection efficiency of the RRDE (0.2) determined using the same configuration with an \( IrO_2 \) thin-film electrode.\(^{3,4}\)
2. Supporting Figures and Tables

Fig. S1 Crystal structures of (a) FeOOH and (b) Ni$_2$(CO$_3$)(OH)$_2$.

Fig. S2 TEM images of FeNi/Ni HS.
**Fig. S3** Cs-corrected HRTEM characterization of the “torso” region: (a) the HAADF-STEM. The red rectangular box is the EDX-mapping collection area, and the yellow circles indicate where the EELS data were collected. (b) the high-resolution EDX-mapping, (c) the spot-scanning EELS. The Fe and O signals are clearly shown in EELS spectrum because FeOOH as substrate is present in a large quantity.

**Fig. S4** The basic molecular unit of gelatin.
Fig. S5 Growth characteristics of FeNi/Ni HS over a course of 36 h. (a) 3 h, (b) 6 h, (c) 12 h, (d) 18 h, (e) 21 h, (f) 27 h, (g) 33 h, (h) 36 h.
Fig. S6 Other conditions-dependent growth characteristics of FeNi/Ni HS. (a-d) After reaction for 36 h, with different dosages of gelatin (urea=8 g, NiCl$_2$·6H$_2$O/FeCl$_3$·6H$_2$O=3/2): (a) gelatin=0 mL, (b) gelatin=4 mL, (c) gelatin=8 mL, (d) gelatin=16 mL. (e-h) After reaction for 36 h, with different dosages of urea (gelatin=8 mL, NiCl$_2$·6H$_2$O/FeCl$_3$·6H$_2$O=3/2): (e) urea=0 g, (f) urea=4 g, (g) urea=8 g, (h) urea=16 g. (i-l) After reaction for 36 h, with different precursor mole ratios (urea=8 g, gelatin=8 mL): (i) NiCl$_2$·6H$_2$O/FeCl$_3$·6H$_2$O=1/2, (j) NiCl$_2$·6H$_2$O/FeCl$_3$·6H$_2$O=1/1, (k) NiCl$_2$·6H$_2$O/FeCl$_3$·6H$_2$O=3/2, (l) NiCl$_2$·6H$_2$O/FeCl$_3$·6H$_2$O=2/1.
Fig. S7 Morphological and structural characterizations of the compared samples. TEM images of (a) FeOOH, (b) Ni₂(CO₃)(OH)₂, and (c) Ni-FeOOH; XRD patterns of (d) FeOOH, (e) Ni₂(CO₃)(OH)₂, and (f) Ni-FeOOH.
Fig. S8 TEM images of different FeNi/Ni HS more ratios (a) Ni:Fe=1:1, (b) Ni:Fe=3:2, (c) Ni:Fe=2:1. Comparative electrochemical studies: (d) Linear sweep voltammetry (LSV), (e) Tafel plots (log $j$ - $\eta$), (f) Nyquist plots.

Fig. S9 $C_{dl}$ calculations of catalysts.
Fig. S10 Typical cyclic voltammetry curves of (a) FeOOH, (b) Ni$_2$(CO$_3$)(OH)$_2$, (c) FeOOH&Ni$_2$(CO$_3$)(OH)$_2$, (d) Ni-FeOOH and (e) FeNi/Ni HS in 1.0 M KOH with different scan rates (10-80 mV/s).
Fig. S11 Ring current measurements: (a) FeNi/Ni HS, (b) FeOOH, (c) \( \text{Ni}_2(\text{CO}_3)(\text{OH})_2 \), (d) FeOOH&\( \text{Ni}_2(\text{CO}_3)(\text{OH})_2 \), and (e) Ni-doped FeOOH, all being on an RRDE (1600 rpm) in N\(_2\)-saturated 1.0 M KOH solution (ring potential at 0.40 V vs. RHE).

Fig. S12 Morphological and structural characterizations of FeNi/Ni HS recovered after the 40-h test. (a) TEM image. (b) XRD patterns.
**Fig. S13** XPS spectra of (a) Fe 2\(p\), (b) Ni 2\(p\) for FeNi/Ni HS before and after the OER tests.

**Fig. S14** Schematics of the electronic interplay between Ni and Fe via oxo bridge in FeNi/Ni HS.
**Fig. S15** The characterizations of Ni vacancies in the “wing” Region C. (a) HAADF-STEM image. (b) Electron transfer of Ni vacancies.

**Fig. S16** *In-situ* Raman spectra of FeOOH at different voltages.
Fig. S17 Characterization and properties of FeCo/Co HS. (a) HAADF-STEM image and EDX elemental maps. (b) XRD patterns. (c) XPS spectrum of Co 2p. (d) Polarization curves. (e) Nyquist plots. (f) Overpotential at 10 mA cm\(^{-2}\) and current density at 1.65 V vs. RHE derived from the LSV curves.

The HAADF-STEM image (Fig. S17a) exhibited a saber-like structure, in which the hilt is about 400 nm length and 100 nm width, and the sword body is about 600 nm length and 20 nm width. EDX-mapping showed Co element throughout the structure, while Fe only in the hilt. XRD (Fig. S17b) confirmed the heterostructure consists of FeOOH (JCPDS: 34-1266) and Co(CO\(_3\))\(_{0.5}\)(OH) (JCPDS: 48-0083). The Co 2p XPS spectra (Fig. S17c) of FeCo/Co HS is fitted into two pairs of peaks, corresponding to Co\(^{2+}\) (781.2 eV, 797.5 eV) and Co\(^{3+}\) (780.0 eV, 796.3 eV),\(^5,6\) compare to Co(CO\(_3\))\(_{0.5}\)(OH), the binding energies of FeCo/Co HS exhibit positive shifts (0.3 eV), demonstrating in higher valence Co\(^{3+}\) in the heterostructures. LSV (Fig. S17d)
showed that the onset overpotential of FeCo/Co HS is 270 mV, which is the smallest in comparison groups. EIS (Fig. S17e) demonstrates that $R_{ct}$ of FeCo/Co HS is also the smallest indicating superior charge transfer kinetics. Fig. S17f showed FeCo/Co HS exhibits the smallest overpotential and the largest current density, which again demonstrates its excellent catalytic activity.

**Table S1.** ICP analysis of FeNi/Ni HS with different metal ratios.

| Catalysts                  | Ni (wt.%) | Fe (wt.%) | Ni/Fe    |
|----------------------------|-----------|-----------|----------|
| FeNi/Ni HS(Ni:Fe=1/1)      | 20.21     | 22.83     | 1.00/1.13|
| FeNi/Ni HS(Ni:Fe=3/2)      | 22.48     | 15.16     | 2.97/2.00|
| FeNi/Ni HS(Ni:Fe=2/1)      | 23.74     | 12.18     | 1.95/1.00|

**Table S2.** Changes in metal content of FeNi/Ni HS over a course of 36 hours.

| Time | Ni (wt.%) | Fe (wt.%) |
|------|-----------|-----------|
| 3 h  | 19.84     | 80.16     |
| 6 h  | 38.97     | 61.03     |
| 18 h | 50.97     | 49.03     |
| 36 h | 61.14     | 38.96     |
Table S3. Comparisons of the electrocatalytic performance with reported catalysts.

| Electro catalyst | Medium        | Oset potential (V vs. RHE) | Overpotential \(^a\) (mV) | Stability \(^b\) (h) | Reference                                      |
|------------------|---------------|----------------------------|---------------------------|----------------------|-----------------------------------------------|
| FeNi/Ni HS       | 1.0 M KOH     | 1.45                       | 268                       | 40                   | This work                                     |
| Fe-CoOOH/G\(^7\) | 1.0 M KOH     | 1.51                       | 330                       | 5                    | *Adv. Energy Mater.* 2017, 7, 1602148.       |
| CoNi(OH)x\(^8\)  | 1.0 M KOH     | 1.48                       | 280                       | 24                   | *Adv. Energy Mater.* 2016, 6, 150166.        |
| α-Co\(_4\)Fe(OH)_x \(^9\) | 1.0 M KOH | 1.48                       | 295                       | 3                    | *J. Mater. Chem. A*, 2017, 5, 1078-1084.    |
| NiCo LDH\(^10\)  | 1.0 M KOH     | 1.53                       | 367                       | 6                    | *Nano Lett.* 2015, 15, 1421-1427.           |
| NiCo\(_{2.7}\)(OH)x\(^11\) | 1.0 M KOH | 1.48                       | 350                       | 10                   | *Adv. Energy Mater.* 2015, 5, 1401880.      |
| Fe-CoOOH/G\(^12\) | 1.0 M KOH     | 1.50                       | 330                       | 6                    | *Adv. Energy Mater.* 2017, 7, 1602148.      |
| FeOOH(Se)/IF\(^13\) | 1.0 M KOH | 1.49                       | 287                       | 14                   | *J. Am. Chem. Soc.* 2019, 141, 7005-7013.   |
| Co(OH)_2@NCNTs@N\(_F\)\(^14\) | 1.0 M KOH | 1.40                       | 270                       | 200                  | *Nano Energy* 2018, 47, 96-104.             |
| Fe-CoNi LDH\(^15\) | 1.0 M KOH     | 1.45                       | 260                       | 12                   | *Appl. Surf. Sci.* 2021, 565, 150506.       |
| NiCo LDH\(^16\)  | 1.0 M KOH     | 1.40                       | 264                       | 25                   | *Nano Res.* 2022, 15, 4986-4995.           |
| NiCo-LDH/ZnCo\(_2\)O\(_4\)\(^17\) | 1.0 M KOH | 1.47                       | 260                       | /                    | *J. Colloid Interface Sci.* 2021, 604, 832-843. |
| Fe\(_{0.33}\)Co\(_{0.67}\)OOH | 1.0 M KOH     | 1.45                       | 266                       | 24                   | *Angew. Chem. Int. Ed.* 2018, 57, 2672-2676. |
| ZNDP-1\(^19\)    | 1.0 M KOH     | 1.30                       | 170                       | /                    | *Appl. Surf. Sci.* 2021, 562, 150253.       |
| NiFeV nanofibers\(^20\) | 1.0 M KOH | 1.41                       | 263                       | /                    | *Angew. Chem.Int. Ed.*                      |
| Material          | KOH Concentration | Overpotential (V) | Stability (mA cm$^{-2}$) | Journal Details |
|-------------------|-------------------|-------------------|--------------------------|----------------|
| Nb-NiFe-LDH$^{21}$ | 1.0 M KOH         | 1.34              | 242                      | Chem. Eng. J. 2022, 427, 131643. |
| FeBi@FeNi LDH$^{22}$ | 1.0 M KOH       | 1.47              | 246                      | J. Colloid Interface Sci. 2022, 610, 173-181. |

$^a$ Overpotential at a current density of 10 mA cm$^{-2}$; $^b$ Stability at a current density of 10 mA cm$^{-2}$. 
3. Reference

1. X. H. Sun, Q. Shao, Y. C. Pi, J. Guo, X. Q. Huang, *J. Mater. Chem. A*, 2017, **5**, 7769.

2. D. Voiry, M. Chhowalla, Y. Gogotsi, N. A. Kotov, Y. Li, R. M. Penner, R. E. Schaak, P. S. Weiss, *ACS Nano* 2018, **12**, 9635-9638.

3. H. L. Fei, J. C. Dong, Y. X. Feng, C. S. Allen, C. Z. Wan, B. Volosskiy, M. F. Li, Z. P. Zhao, Y. L. Wang, H. T. Sun, P. F. An, W. X. Chen, Z. Y. Guo, C. Lee, D. L. Chen, I. Shakir, M. J. Liu, T. D. Hu, Y. D. Li, A. I. Kirkland, X. F. Duan, Y. Huang, *Nat. Catal.* 2018, **1**, 63.

4. T. Y. Ma, S. Dai, M. Jaroniec, S. Z. Qiao, *J. Am. Chem. Soc.* 2014, **136**, 13925.

5. L. Hui, Y. Xue, D. Jia, Yu, H.; Zhang, C.; Li, Y. *Adv. Energy Mater.* 2018, **8**, 1800175.

6. P. W. Menezes, S. Yao, R. Beltran-Suito, J. N. Hausmann, P. V. Menezes, M. Driess, *Angew. Chem. Int. Ed.* 2021, **60**, 4640-4647.

7. X. T. Han, C. Yu, S. Zhou, C. T. Zhao, H. W. Huang, J. Yang, Z. B. Liu, J. J. Zhao, J. S. Qiu, *Adv. Energy Mater.* 2017, **7**, 1602148.

8. S. W. Li, Y. C. Wang, S. J. Peng, L. J. Zhang, A. M. Al-Enizi, H. Zhang, X. H. Sun, G. F. Zheng, *Adv. Energy Mater.* 2016, **6**, 150166.

9. H. Y. Jin, S. J. Mao, G. P. Zhan, F. Xu, X. B. Bao, Y. Wang, *J. Mater. Chem. A*, 2017, **5**, 1078-1084.

10. H. F. Liang, F. Meng, M. C. Acevedo, L. S. Li, A. Forticaux, L. C. Xiu, Z. C. Wang, S. Jin, *Nano Lett.* 2015, **15**, 1421-1427.

11. J. W. Nai, H. J. Yin, T. T. You, L. R. Zheng, J. Zhang, P. X. Wang, Z. Jin, Y. Tian, J. Z. Liu, Z. Y. Tang, L. Guo, *Adv. Energy Mater.* 2015, **5**, 1401880.

12. X. Han, C. Yu, S. Zhou, C. Zhao, H. Huang, J. Yang, Z. Liu, J. Zhao, J. Qiu, *Adv. Energy Mater.* 2017, **7**, 1602148.

13. S. Niu, W. J. Jiang, Z. X. Wei, T. Tang, J. M. Ma, J. S. Hu, L. J. Wan, *J. Am. Chem. Soc.* 2019, **141**, 7005-7013.

14. P. Guo, J. Wu, X. B. Li, J. Luo, W. M. Lau, H. Liu, X. L. Sun, L. M. Liu, *Nano Energy* 2018, **47**, 96-104.

15. Y. L. Shi, J. Q. Li, B. Y. Zhang, S. Y. Lv, T. Wang, X. Liu, *Appl. Surf. Sci.* 2021, **565**, 150506.
16. J. H. Li, L. L. Wang, H. J. He, Y. Q. Chen, Z. R. Gao, N. Ma, B. Wang, L. L. Zheng, R. L. Li, Y. J. Wei, J. Q. Xu, Y. Xu, B. W. Cheng, Z. Yin, D. Ma, *Nano Res.* 2022, **15**, 4986-4995.

17. M. Shamloofard, S. Shahrokhian, M. K. Amini, *J. Colloid Interface Sci.* 2021, **604**, 832-843.

18. S. H. Ye, Z. X. Shi, J. X. Feng, Y. X. Tong, G. R. Li, *Angew. Chem. Int. Ed.* 2018, **57**, 2672-2676.

19. J. Jana, K. C. Bhamu, Y. T. Ngo, S. G. Kang, J. S. Chuang, *Appl. Surf. Sci.* 2021, **562**, 150253.

20. B. Zhang, Z. H. Wu, W. J. Shao, Y. Gao, W. W. Wang, T. Ma, L. Ma, S. Li, C. Cheng, C. S. Zhao, *Angew. Chem. Int. Ed.* 2022, **61**, e2021153.

21. Y. N. Zhou, F. L. Wang, S. Y. Dou, Z. N. Shi, B. Dong, *Chem. Eng. J.* 2022, **427**, 131643.

22. F. G. Wang, B. Liu, H. Y. Wang, Z. Y. Lin, Y. W. Dong, N. Yu, R. N. Luan, Y. M. Chai, B. Dong, *J. Colloid Interface Sci.* 2022, **610**, 173-181.