# **Electronic Supplementary Information**

Rare-earth element doped NiFe-MOFs as efficient and robust

## bifunctional electrocatalysts for both alkaline freshwater and

## seawater splitting

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#### **Experimental Section**

### Materials

All chemicals were purchased from commercial sources and directly used without further purification. The nickel foam (NF) substrates  $(2.0 \text{ cm} \times 3.0 \text{ cm})$  were carefully cleaned with acetone, deionized water and absolute ethanol by ultrasonic treatment.

#### Synthesis of NiFe-MOF

Firstly, 1.0 mmol of FeCl<sub>2</sub>·4H<sub>2</sub>O (98%) and 1.0 mmol of terephthalic acid (99%) were dissolved in a mixed solvent of 10.5 mL of N,N-dimethylformamide (DMF), 0.75 mL of absolute ethanol and 0.75 mL of deionized water. Subsequently, the obtained solution and a piece of NF were transferred into 25 mL of Teflon-lined autoclave and heated at 125 °C for 12 h. After cooling, the self-supporting NiFe-MOF electrode was repeatedly washed with deionized water and then dried at 60 °C in a vacuum oven.

#### Synthesis of CeNiFe-MOF

Firstly, 1.0 mmol of FeCl<sub>2</sub>·4H<sub>2</sub>O (98%), 1.0 mmol of terephthalic acid (99%) and 0.05 mmol of Ce(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O (99.95%) were dissolved in a mixed solvent of 10.5 mL of DMF, 0.75 mL of absolute ethanol and 0.75 mL of deionized water. Subsequently, the obtained solution and a piece of NF were transferred into 25 mL of Teflon-lined autoclave and heated at 125 °C for 12 h. After cooling, the self-supporting CeNiFe-MOF electrode was repeatedly washed with deionized water and then dried at 60 °C in a vacuum oven. Other CeNiFe-MOF samples with different Ce content were also prepared in the same way by adding 0.01 and 0.1 mmol of Ce(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O, respectively.

## Synthesis of YNiFe-MOF and LaNiFe-MOF

The self-supporting YNiFe-MOF and LaNiFe-MOF electrodes were prepared in the same way by using  $Y(NO_3)_3 \cdot 6H_2O$  (99.9%) and La(NO<sub>3</sub>)<sub>3</sub>  $\cdot 6H_2O$  (99.99%) as dopant sources, respectively.

#### Materials characterization

The morphology and microstructure of samples were observed by field-emission scanning electron microscopy (FESEM; Hitachi, SU8010) and transmission electron microscopy (TEM; JEOL, JEM-1400Plus). Elemental mapping images were obtained from spherical aberration correction electron microscopy (JEOL, JEM-ARM200F). X-ray diffraction (XRD) patterns were collected on a Rigaku Smartlab diffractometer with Cu-K $\alpha$  radiation ( $\lambda = 1.5406$  Å). X-ray photoelectron spectroscopy (XPS) measurements were conducted on a Thermo Fisher Scientific K-Alpha instrument. The metal contents of samples were determined by inductively coupled plasma mass spectrometry (ICP-MS; iCAP RQ).

#### **Electrochemical measurements**

The electrochemical measurements were performed on a CHI 760E workstation by using the three-electrode system, where the self-supporting MOFs were directly used as the working electrode, saturated Ag/AgCl electrode and graphite rod were used as the reference and counter electrodes, respectively. The Pt/C (20% Pt) and IrO2 benchmarks on NF were also used for comparison. Firstly, 5 mg of commercial catalysts were dispersed in a mixed solvent of 490 µL of deionized water, 490 µL of absolute ethanol and 20 µL of Nafion by ultrasonication. Then, 50 µL of the ink was dropped onto NF with the loading area of 0.5 cm<sup>2</sup>. Linear sweep voltammetry (LSV) measurements were conducted at a scan rate of 5 mV s<sup>-1</sup> with 95% iR compensation. Electrochemical impedance spectroscopy (EIS) measurements were carried out in the frequency range of  $0.1-10^5$  Hz with an amplitude of 5 mV. Cyclic voltammetry (CV) measurements were performed at different scan rates (50, 100, 150 and 200 mV s<sup>-1</sup>) to evaluate the double-layer capacitance (C<sub>dl</sub>). Electrochemically active surface area (ECSA) was calculated by the following equation:  $ECSA = C_{dl}/C_s$ ,  $C_s = 0.06 \text{ mF cm}^{-2}$ . The measured potentials were calibrated to reversible hydrogen electrode (RHE) according to the equation:  $E_{RHE} = E_{Ag/AgCl} + 0.197 \text{ V} + 0.059 \times \text{pH}.$ 

#### **Computational details**

All density functional theory (DFT) calculations were conducted in the Vienna ab initio simulation package (VASP) code<sup>1-3</sup>. The ionic cores were described by the projector-augmented wave (PAW) pseudo-potential<sup>4, 5</sup>. The cut-off energy for plane wave expansion was set to 400 eV. The electronic self-consistent-loop criterion was set to  $10^{-8}$  eV. During geometry optimization, the structures were relaxed to forces on all atoms smaller than 0.02 eV/Å. Gaussian smearing method was used with 0.05 eV width. To avoid the interaction among slabs, the vacuum spacing between periodically repeated slabs was set to 15 Å. The Brillouin zone integration was carried out with the  $4\times8\times1$  Monkhorst-Pack k-point sampling for all structures. DFT + U method was applied to better describe the systems. U = 4.3 and 0, U = 3.8 and 0, as well as U = 5.5 and 0.5 were chosen for Fe, Ni and Ce in OER and HER calculations, respectively.

During optimization of H\*, OH\*, O\* and OOH\* adsorption, the substrate atoms were fixed to allow a time-efficient relaxation of the adsorbed species. The adsorption energies ( $E_{ads}$ ) can be obtained by the equation:  $E_{ads} = E_{ad/sub} - E_{ad} - E_{sub}$  where  $E_{ad/sub}$ ,  $E_{ad}$  and  $E_{sub}$  are the total energies of the optimized adsorbate/substrate system, the adsorbate in the structure and the clean substrate, respectively.

Gibbs free energy was calculated as follow:  $\Delta G = \Delta E_{DFT} + \Delta ZPE - T\Delta S - eU$ where  $\Delta E_{DFT}$  is the calculated reaction energy by DFT method,  $\Delta ZPE$  is the change of zero-point energy and  $\Delta S$  is the change of entropy.



Fig. S1 SEM image of NiFe-MOF.



Fig. S2 High-resolution Ce 3d spectrum of CeNiFe-MOF.



Fig. S3 The content of  $Fe^{2+}$  and  $Fe^{3+}$  obtained from XPS analysis.



Fig. S4 XRD patterns of Ce<sub>0.01</sub>NiFe-MOF and Ce<sub>0.1</sub>NiFe-MOF.



Fig. S5 (a) HER and (b) OER polarization curves of Ce<sub>x</sub>NiFe-MOF in 1.0 M KOH.



**Fig. S6** Overpotential comparison of CeNiFe-MOF, NiFe-MOF and commercial Pt/C in different electrolytes.



**Fig. S7** Overpotential comparison of CeNiFe-MOF, NiFe-MOF and commercial IrO<sub>2</sub> at different current densities in 1.0 M KOH.



**Fig. S8** CV curves of (a) NF, (b) NiFe-MOF and (c) CeNiFe-MOF at different scan rates in 1.0 M KOH. (d) The corresponding double-layer capacitance.



**Fig. S9** (a) The ECSA-normalized HER polarization curves in 1.0 M KOH. (b) The corresponding specific activities at an overpotential of 100 mV. (c) The ECSA-normalized OER polarization curves in 1.0 M KOH. (d) The corresponding specific activities at 1.43 V vs. RHE.



**Fig. S10** Nyquist plots measured at the overpotential of (a) 150 mV for HER and (b) 200 mV for OER in 1.0 M KOH.



**Fig. S11** (a) Ni 2p and (b) Fe 2p spectra of CeNiFe-MOF after water electrolysis test in 1.0 M KOH. (c) The content of  $Fe^{2+}$  and  $Fe^{3+}$  obtained from XPS analysis.



Fig. S12 Overpotential comparison of CeNiFe-MOF, NiFe-MOF and commercial  $IrO_2$  at different current densities in 1.0 M KOH + 0.5 M NaCl.



Fig. S13 Tafel slopes for (a) HER and (b) OER in 1.0 M KOH + 0.5 M NaCl.



**Fig. S14** CV curves of (a) NF, (b) NiFe-MOF and (c) CeNiFe-MOF at different scan rates in 1.0 M KOH + 0.5 M NaCl. (d) The corresponding double-layer capacitance.



**Fig. S15** The ECSA-normalized (a) HER and (b) OER polarization curves in 1.0 M KOH + 0.5 M NaCl.



**Fig. S16** Nyquist plots measured at the overpotential of (a) 150 mV for HER and (b) 200 mV for OER in 1.0 M KOH + 0.5 M NaCl.



**Fig. S17** Overpotential comparison of CeNiFe-MOF, NiFe-MOF and commercial IrO<sub>2</sub> at different current densities in 1.0 M KOH + seawater.



Fig. S18 Tafel slopes for (a) HER and (b) OER in 1.0 M KOH + seawater.



**Fig. S19** CV curves of (a) NF, (b) NiFe-MOF and (c) CeNiFe-MOF at different scan rates in 1.0 M KOH + seawater. (d) The corresponding double-layer capacitance.



**Fig. S20** The ECSA-normalized (a) HER and (b) OER polarization curves in 1.0 M KOH + seawater.



**Fig. S21** Nyquist plots measured at the overpotential of (a) 150 mV for HER and (b) 200 mV for OER in 1.0 M KOH + seawater.



**Fig. S22** Comparison of HER performance of as-prepared CeNiFe-MOF with other MOF-based electrocatalysts in 1.0 M KOH.



**Fig. S23** Comparison of OER performance of as-prepared CeNiFe-MOF with other MOF-based electrocatalysts in 1.0 M KOH.



**Fig. S24** Comparison of alkaline freshwater splitting performance of as-prepared CeNiFe-MOF with other MOF-based electrocatalysts.



**Fig. S25** Comparison of HER performance of as-prepared CeNiFe-MOF with other advanced electrocatalysts in 1.0 M KOH + seawater.



**Fig. S26** Comparison of OER performance of as-prepared CeNiFe-MOF with other advanced electrocatalysts in 1.0 M KOH + seawater.



**Fig. S27** Comparison of alkaline seawater splitting performance of as-prepared CeNiFe-MOF with other advanced electrocatalysts.



Fig. S28 The optimized structures of adsorbed H\* on (a) NiFe-MOF/Ni and (b) CeNiFe-MOF/Ni.



Fig. S29 The optimized structures of adsorbed  $H^*$  on (a) NiFe-MOF/Fe and (b) CeNiFe-MOF/Fe.



Fig. S30 The optimized structures of adsorbed H\* on CeNiFe-MOF/Ce.



**Fig. S31** The optimized structures of adsorbed intermediates on (a–c) NiFe-MOF/Ni and (d–f) CeNiFe-MOF/Ni.



**Fig. S32** The optimized structures of adsorbed intermediates on (a–c) NiFe-MOF/Fe and (d–f) CeNiFe-MOF/Fe.



Fig. S33 The optimized structures of adsorbed intermediates on CeNiFe-MOF/Ce.



**Fig. S34** The free energy diagram for OER process on NiFe-MOF/Ni, NiFe-MOF/Fe and CeNiFe-MOF/Fe.



# **Reaction coordination**

Fig. S35 The free energy diagram for HER process on CeNiFe-MOF/Ce.



Fig. S36 XRD patterns of YNiFe-MOF and LaNiFe-MOF.



**Fig. S37** Electrochemical performance of the samples in 1.0 M KOH. (a) HER polarization curves and (b) the corresponding Tafel slopes. (c) OER polarization curves and (d) the corresponding Tafel slopes.



**Fig. S38** Electrochemical performance of the samples in 1.0 M KOH + 0.5 M NaCl. (a) HER polarization curves and (b) the corresponding Tafel slopes. (c) OER polarization curves and (d) the corresponding Tafel slopes.



**Fig. S39** Electrochemical performance of the samples in 1.0 M KOH + seawater. (a) HER polarization curves and (b) the corresponding Tafel slopes. (c) OER polarization curves and (d) the corresponding Tafel slopes.



**Fig. S40** CV curves of YNiFe-MOF and LaNiFe-MOF in (a, b) 1.0 M KOH, (d, e) 1.0 M KOH + 0.5 M NaCl and (g, h) 1.0 M KOH + seawater. (c, f, i) The corresponding double-layer capacitance.



**Fig. S41** (a) The ECSA-normalized HER polarization curves in 1.0 M KOH. (b) The corresponding specific activities at an overpotential of 100 mV. (c) The ECSA-normalized OER polarization curves in 1.0 M KOH. (d) The corresponding specific activities at 1.43 V vs. RHE.



**Fig. S42** (a) The ECSA-normalized HER polarization curves in 1.0 M KOH + 0.5 M NaCl. (b) The corresponding specific activities at an overpotential of 150 mV. (c) The ECSA-normalized OER polarization curves in 1.0 M KOH + 0.5 M NaCl. (d) The corresponding specific activities at 1.48 V vs. RHE.



**Fig. S43** (a) The ECSA-normalized HER polarization curves in 1.0 M KOH + seawater. (b) The corresponding specific activities at an overpotential of 150 mV. (c) The ECSA-normalized OER polarization curves in 1.0 M KOH + seawater. (d) The corresponding specific activities at 1.48 V vs. RHE.



Fig. S44 Nyquist plots for (a, c, e) HER and (b, d, f) OER measured in different electrolytes.

| Electrocatalysts               | Substrate | $\eta_{10}$ | Tafel slope     | Ref. |
|--------------------------------|-----------|-------------|-----------------|------|
|                                |           | (mV)        | $(mV dec^{-1})$ |      |
| CeNiFe-MOF                     |           | 113         | 59.4            | This |
| YNiFe-MOF                      | NF        | 122         | 62.9            | work |
| LaNiFe-MOF                     |           | 119         | 65.7            |      |
| NiFe-MOF-5                     | NF        | 163         | 139             | 6    |
| NiFe(dobpdc)                   | NF        | 113         | 69              | 7    |
| Fe-Co-Ni MOF                   | NF        | 116         | 56              | 8    |
| NiYCe-MOF/NF                   | NF        | 136         | 74              | 9    |
| Fe(OH) <sub>x</sub> @Cu-MOF    | CP        | 112         | 76              | 10   |
| S-NiBDC                        | NF        | 113         | 75              | 11   |
| LIA-Ni-BDC                     | NF        | 146         | 116             | 12   |
| CdFe-BDC                       | NF        | 148         | 108.71          | 13   |
| BP@MOF                         | GCE       | 180         | 109             | 14   |
| Fe <sub>2</sub> V-MOF          | NF        | 198         | 182             | 15   |
| Co-MOF/NF                      | NF        | 115         | 78              | 16   |
| VFe-MOF@NF                     | NF        | 147         | 208.25          | 17   |
| CoFe-MOF@Pa                    | NF        | 255         | /               | 18   |
| Fe@Co-MOF-3                    | NF        | 151         | 86.54           | 19   |
| Co/Cu-MOF(3)                   | GCE       | 391         | 94              | 20   |
| CoNi-MOFs-DBD                  | NF        | 203         | 152.2           | 21   |
| MXene@Ce-MOF                   | /         | 220         | 149.9           | 22   |
| NiFe-MOF/NiSe <sub>x</sub> /NF | NF        | 142         | 94.7            | 23   |
| Ni@CoO@Co-MOFC                 | NF        | 138         | 59              | 24   |
| CuO@NH2-UiO-66/NF              | NF        | 166         | 87              | 25   |
| FeCoMnNi-MOF-74/NF             | NF        | 108         | 72.89           | 26   |

**Table S1.** Comparison of HER activity of MOF-based electrocatalysts in 1.0 M KOH.

NF: nickel foam; CP: carbon paper; GCE: glassy carbon electrode

| Electrocatalysts                               | Substrate | $\eta_{100}$      | $\eta_{500}$ | Tafel slope     | Ref. |
|--|-----------|-------------------|--------------|-----------------|------|
|  |           | (mV)              | (mV)         | $(mV dec^{-1})$ |      |
| CeNiFe-MOF                                     |           | 198               | 224          | 43.5            | This |
| YNiFe-MOF                                      | NF        | 209               | 233          | 48.0            | work |
| LaNiFe-MOF                                     |           | 203               | 223          | 46.2            |      |
| CdFe-BDC                                       | NF        | 290               | /            | 44.57           | 13   |
| Co-MOF/NF                                      | NF        | 311               | /            | 84              | 16   |
| 6%LS-CMMOFs/NF                                 | NF        | 220               | 300          | 54              | 27   |
| Fe-NiHF  | GCE       | 340               | /            | 44              | 28   |
| Ni-BDC/NM88B(Fe)                               | NF        | 232               | /            | 37.6            | 29   |
| NiFc-MOF/NF                                    | NF        | 241               | /            | 44.1            | 30   |
| NiFc'Fc/NF                                     | NF        | 213               | 240          | 45              | 31   |
| NiFe-MOF                                       | CFC       | /                 | 297          | 49.1            | 32   |
| NiFe-MOF/G                                     | GCE       | 326               | /            | 49              | 33   |
| NiFe-MOF@NiS/NF                                | NF        | 290               | 346          | 45.1            | 34   |
| NiFe MOF/NF                                    | NF        | 256               | /            | 40              | 35   |
| NiFe-MOF/FF                                    | FF        | 240               | /            | 73.4            | 36   |
| NiV-MOF NAs                                    | NF        | 290               | /            | 76.2            | 37   |
| FeNi-MOFs/NF                                   | NF        | 266               | /            | 52.4            | 38   |
| NiFe-LDH/MOF                                   | NF        | 275               | /            | 61              | 39   |
| ZnFe-BDC-0.75                                  | NF        | 292               | /            | 90.72           | 40   |
| Ir@NiFe-MOF/NF                                 | NF        | 251               | 298          | 38.5            | 41   |
| Dy <sub>0.05</sub> Fe-MOF/NF                   | NF        | 258               | 318          | 82              | 42   |
| FeNi-LDH/MOF/CC                                | CC        | 263               | /            | 50              | 43   |
| FeMOF/NiMOF/NF                                 | NF        | 359               | /            | 125.3           | 44   |
| NiFe(DMBD)-MOF/NF                              | NF        | 280               | /            | 54.3            | 45   |
| NiFe-MOF-BF <sub>4</sub> <sup>-</sup> -0.3 NSs | GCE       | 278               | /            | 41              | 46   |
| Fe-doped-(Ni-MOFs)/FeOOH                       | NF        | 278               | /            | 50              | 47   |
| Ce-NiBDC/OG                                    | GF        | 337               | /            | 46              | 48   |
| Ce-m-Ni(OH)2@Ni-MOF                            | NF        | 272               | /            | 43.2            | 49   |
| Cu-Fe-NH2 MOF/NF                               | NF        | 270               | 330          | 60.8            | 50   |
| Co/Mn@CNDs-MOF                                 | NF        | 320               | /            | 140             | 51   |
| Fe-MOF-U                                       | NF        | 278               | 322          | 53.3            | 52   |
| MIL-53(Fe)-2OH                                 | NF        | 266               | 314          | 45.4            | 53   |
| AOGTM-PG[R]                                    | NF        | 300               | 335          | 44.3            | 54   |
| CoNiRu-NT                                      | NF        | 335               | /            | 67              | 55   |
| Ni NDC-Co/CP                                   | CP        | 308               | /            | 49.1            | 56   |
| HMIL-88@PPy-TA                                 | NF        | 238               | 303          | 45.8            | 57   |
| CoNi-LDH/Fe MOF/NF                             | NF        | 235               | 277          | 50.7            | 58   |
| MIL-(IrNiFe)@NF                                | NF        | $230(\eta_{50})$  | 300          | 60              | 59   |
| FeMn-MOF/NF(1:1)                               | NF        | $290(\eta_{50})$  | /            | 87.02           | 60   |
| 2D MOF-Fe/Co(1:2)                              | GCE       | $285(\eta_{50})$  | /            | 52              | 61   |
| NiFe-BTC/CCHH/NF                               | NF        | $270(\eta_{50})$  | /            | /               | 62   |
| BN-CoFe-MOF                                    | NF        | $314(\eta_{50})$  | /            | 105.6           | 63   |
| Ni <sub>0.6</sub> Co <sub>1.8</sub> -MOF       | NF        | $380(\eta_{200})$ | /            | 46              | 64   |
| Coo 8Nio 2Fc-MOF                               | GE        | $252(n_{200})$    | /            | 39              | 65   |

**Table S2.** Comparison of OER activity of MOF-based electrocatalysts in 1.0 M KOH.

CFC: carbon fiber cloth; FF: Fe foam; CC: carbon cloth; GF: graphite foil; GE: graphite electrode

| Electrolyzers                  | Substrate | Cell voltage                | Operation time | Ref. |
|--------------------------------|-----------|-----------------------------|----------------|------|
|                                |           | $(V@10 \text{ mA cm}^{-2})$ | (h)            |      |
| CeNiFe-MOF                     | NF        | 1.56                        | 200            | This |
|                                |           |                             |                | work |
| NiFe-MOF-5                     | NF        | 1.57                        | 24             | 6    |
| NiFe(dobpdc)/NF                | NF        | 1.59                        | 30             | 7    |
| Fe-Co-Ni MOF                   | NF        | 1.60                        | 150            | 8    |
| LIA-Ni-BDC                     | NF        | 1.59                        | 50             | 12   |
| LIA-MIL-101(Fe)                |           |                             |                |      |
| CdFe-BDC                       | NF        | 1.63                        | 12             | 13   |
| BP@MOF                         | GCE       | 1.63                        | 10             | 14   |
| Fe <sub>2</sub> V-MOF  Pt/C    | NF        | 1.60                        | 12             | 15   |
| Co-MOF/NF                      | NF        | 1.548                       | 24             | 16   |
| VFe-MOF@NF                     | NF        | 1.61                        | 1              | 17   |
| CoFe-MOF@Pa                    | NF        | 1.66                        | 36             | 18   |
| NiFe-MOF/NiSe <sub>x</sub> /NF | NF        | 1.59                        | 40             | 23   |
| Ni@CoO@Co-MOFC                 | NF        | 1.61                        | 24             | 24   |
| FeCoMnNi-MOF-74/NF             | NF        | 1.62                        | 30             | 26   |
| ZnFe-BDC-0.75  Pt/C            | NF        | 1.64                        | /              | 40   |
| FeMn-MOF/NF(1:1)               | NF        | 1.7@50                      | 12             | 60   |
| NiFe-BTC/CCHH/NF               | NF        | 1.55                        | 12             | 62   |
| P-CCHH/NF                      |           |                             |                |      |
| dye@MOF                        | NF        | 1.98@35                     | 4              | 66   |
| NFF-MOF                        | NFF       | 1.57                        | 24             | 67   |
| NiFe-MOF array                 | NF        | 1.55                        | 100            | 68   |
| NiFe-MS/MOF@NF                 | NF        | 1.61                        | 27             | 69   |
| Ru@CoNi-MOF                    | NF        | 1.58@20                     | 20             | 70   |
| FeDy@MOF-Ni/CC  Pt/C           | CC        | 1.57                        | 80             | 71   |

 Table S3. Performance comparison of MOF-based electrolyzers in 1.0 M KOH.

NFF: NiFe foam

| aikanne seawater electrolyte.                                       |           |                  |                 |      |
|---|-----------|------------------|-----------------|------|
| Electrocatalysts  | Substrate | $\eta_{10}$      | Tafel slope     | Ref. |
|   |           | (mV)             | $(mV dec^{-1})$ |      |
| CeNiFe-MOF  |           | 136              | 63.2            | This |
| YNiFe-MOF   | NF        | 155              | 73.4            | work |
| LaNiFe-MOF  |           | 154              | 68.3            |      |
| Ni-SA/NC  | NF        | 139              | 123             | 72   |
| NiFe LDH/FeOOH  | INF       | 181.8            | /               | 73   |
| Ni-Co@Fe-Co PBA   | GCE       | 183              | 60              | 74   |
| Ti@NiB  | Ti plate  | 149              | 118             | 75   |
| NRAHM-NiO   | NF        | 178              | 115             | 76   |
| CoPx  | NF        | 117              | 71.1            | 77   |
| (Co,Fe)PO <sub>4</sub>  | IF        | 137              | /               | 78   |
| Mo-CoP <sub>x</sub> /NF   | NF        | 158              | /               | 79   |
| Co <sub>x</sub> P <sub>y</sub> /Ni <sub>x</sub> P <sub>y</sub> -NPC | GCE       | 203              | 135             | 80   |
| Co-N,P-HCS  | NF        | 164              | 109             | 81   |
| CoSe <sub>2</sub> -NCF  | CC        | 134              | 67              | 82   |
| CoSe/MoSe <sub>2</sub>  | GCE       | 189              | /               | 83   |
| 1D-Cu@Co-CoO/Rh   | CF        | 137.7            | 124.8           | 84   |
| 2D meso-Mo <sub>2</sub> C/Mo <sub>2</sub> N                         | GCE       | 197              | 67.6            | 85   |
| Co@NCNT/CoMoyOx   | CFP       | 125              | /               | 86   |
| Oct_Cu <sub>2</sub> O-NF  | NF        | $237(\eta_{20})$ | 160             | 87   |
| FMCO/NF   | NF        | $250(\eta_{50})$ | /               | 88   |

**Table S4.** Comparison of HER performance of the advanced electrocatalysts in alkaline seawater electrolyte.

INF: iron-nickel foam; IF: iron foam; CF: copper foam; CFP: carbon fiber paper

| amaime seawater electroffte.  |           |              |              |                 |      |
|---|-----------|--------------|--------------|-----------------|------|
| Electrocatalysts  | Substrate | $\eta_{100}$ | $\eta_{500}$ | Tafel slope     | Ref. |
|   |           | (mV)         | (mV)         | $(mV dec^{-1})$ |      |
| CeNiFe-MOF  |           | 224          | 277          | 58.2            | This |
| YNiFe-MOF   | NF        | 243          | 273          | 46.6            | work |
| LaNiFe-MOF  |           | 242          | 288          | 57.8            |      |
| Ni-BDC/NM88B(Fe)  | NF        | 299          | /            | 66.8            | 29   |
| NiFe-MOF@NiS/NF   | NF        | 296          | 355          | 86.2            | 34   |
| NiFe-LDH/MOF  | NF        | 307          | /            | 61              | 39   |
| ZnFe-BDC-0.75   | NF        | 308          | /            | /               | 40   |
| HMIL-88@PPy-TA  | NF        | 259          | 345          | 53.7            | 57   |
| Ni <sub>0.6</sub> Co <sub>1.8</sub> -MOF                                  | NF        | 360          | /            | 53              | 64   |
| NiFe LDH/FeOOH  | INF       | 286.2        | /            | /               | 73   |
| NRAHM-NiO   | NF        | 340          | 680          | 82              | 76   |
| CoP <sub>x</sub> @FeOOH   | NF        | 283          | 337          | 50.3            | 77   |
| Co-N,P-HCS  | NF        | 490          | /            | 121.5           | 81   |
| NiIr-LDH  | NF        | 315          | 361          | 78.8            | 89   |
| NF/NiFe LDH   | NF        | 247          | 296          | /               | 90   |
| NiFe-LDH-6-4/CC   | CC        | 301          | /            | /               | 91   |
| NiFe LDH-CeW@NFF  | NFF       | /            | 330          | 81.2            | 92   |
| Ni <sub>2</sub> Fe-LDH/FeNi <sub>2</sub> S <sub>4</sub> /NF               | NF        | 271          | /            | /               | 93   |
| S-NiMoO4@NiFe-LDH   | NF        | 315          | /            | /               | 94   |
| MnCo2O4@NiFe-LDH/NF   | NF        | 245          | 578          | /               | 95   |
| B-Co <sub>2</sub> Fe LDH  | NF        | 310          | 376          | 63.8            | 96   |
| S-(Ni,Fe)OOH  | NF        | 300          | 398          | /               | 97   |
| FeOOH <sub>0.60</sub> /Ni(HCO <sub>3</sub> ) <sub>2</sub>                 | CFP       | 324          | /            | /               | 98   |
| FeOOH@2.5Ni(OH) <sub>2</sub> /NF  | NF        | 325          | /            | /               | 99   |
| Fe-NiSOH  | NF        | 263          | 311          | /               | 100  |
| NiPS/NF   | NF        | 344          | 392          | /               | 101  |
| NiFeS/NF  | NF        | 226          | 300          | /               | 102  |
| NiCoS/NF  | NF        | 360          | 440          | /               | 103  |
| Cr-Co <sub>x</sub> P  | NF        | 334          | 392          | /               | 104  |
| MnCo/NiSe   | NF        | /            | 419.4        | 81.2            | 105  |
| Mo-Ni <sub>3</sub> S <sub>2</sub> /NF                                     | NF        | 291          | /            | /               | 106  |
| Ni <sub>2</sub> P-Fe <sub>2</sub> P/NF                                    | NF        | 305          | /            | /               | 107  |
| Ni <sub>3</sub> FeN@C/NF  | NF        | 314          | 394          | /               | 108  |
| Ni <sub>3</sub> S <sub>2</sub> /Fe-NiP <sub>x</sub> /NF                   | NF        | 290          | 336          | 61.3            | 109  |
| Ni(OH) <sub>2</sub> -TCNQ/GP  | GP        | 382          | /            | 75              | 110  |
| NiCoHPi@Ni <sub>3</sub> N/NF  | NF        | 396          | 474          | 108.6           | 111  |
| Mn-doped Ni <sub>2</sub> P/Fe <sub>2</sub> P                              | NF        | 270          | 325          | /               | 112  |
| B-MnFe <sub>2</sub> O <sub>4</sub> @MFOC                                  | NF        | 405          | /            | /               | 113  |
| MoS <sub>2</sub> -(FeNi) <sub>9</sub> S <sub>8</sub> /NFF                 | NFF       | 256          | /            | /               | 114  |
| Fe <sub>2</sub> P/Ni <sub>1.5</sub> Co <sub>1.5</sub> N/Ni <sub>2</sub> P | NF        | 255          | 307          | /               | 115  |

**Table S5.** Comparison of OER performance of the advanced electrocatalysts in alkaline seawater electrolyte.

GP: graphite paper

| Electrolyzers  | Substrate | Cell voltage                | Operation time | Ref. |
|--|-----------|-----------------------------|----------------|------|
|  |           | $(V@10 \text{ mA cm}^{-2})$ | (h)            |      |
| CeNiFe-MOF   | NF        | 1.59                        | 515            | This |
|  |           |                             |                | work |
| CdFe-BDC   | NF        | 1.68                        | /              | 13   |
| Ru@CoNi-MOF  | NF        | 1.67@20                     | 18             | 70   |
| Ni-Co@Fe-CoPBA   | GCE       | 1.6@30                      | 100            | 74   |
| NiCo@A-NiCo-PBA-AA   |           |                             |                |      |
| Ti@NiB   | Ti plate  | 1.75@20                     | 40             | 75   |
| NiFeOOH//(Co,Fe)PO4  | ĪF        | 1.625                       | 50             | 78   |
| Mo-CoP <sub>X</sub> /NF  | NF        | 1.61                        | 100            | 79   |
| Co-N,P-HCS   | NF        | 2.0@50                      | 500            | 81   |
| CoSe/MoSe <sub>2</sub> /NF                                       | NF        | 1.77                        | 38             | 83   |
| 1D-Cu@Co-CoO/Rh  | CF        | 1.70                        | 12             | 84   |
| Oct_Cu <sub>2</sub> O-NF   | NF        | 1.71                        | /              | 87   |
| FMCO/NF  | NF        | 1.59                        | 180            | 88   |
| MnCo <sub>2</sub> O <sub>4</sub> @NiFe-LDH                       | NF        | 1.56                        | 100            | 95   |
| FeOOH <sub>0.60</sub> /Ni(HCO <sub>3</sub> ) <sub>2</sub>   Pt/C | CFP       | 1.56                        | 12             | 98   |
| FeOOH@2.5Ni(OH)2/NF  | NF        | 1.63@20                     | 72             | 99   |
| Pt/C/NF  |           |                             |                |      |
| Mn-doped Ni <sub>2</sub> P/Fe <sub>2</sub> P                     | NF        | 1.64                        | 120            | 112  |
| NiMo films   | NF        | 1.62                        | 18             | 116  |
| CoF-3  CoF-2   | CFP       | 1.76                        | $\approx 20$   | 117  |
| PtOx-NiOn/NF   | NF        | 1.58                        | /              | 118  |
| FCNP@CQDs/CP   | CP        | 1.61                        | 50             | 119  |
| Nisa-Nipi/MoS2 NSs   | CC        | 1.66                        | 15             | 120  |

**Table S6.** Performance comparison of the advanced alkaline seawater electrolyzers.

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