# Supplementary Materials

## Uniquely integrated Fe-doped $Ni(OH)_2$ nanosheets for highly efficient oxygen and hydrogen evolution reactions

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## 1. Supplementary Figures

**Fig. S1** Optical images of pristine Ni foam (*left*) and the FeNiOH/NF electrode (*right*).

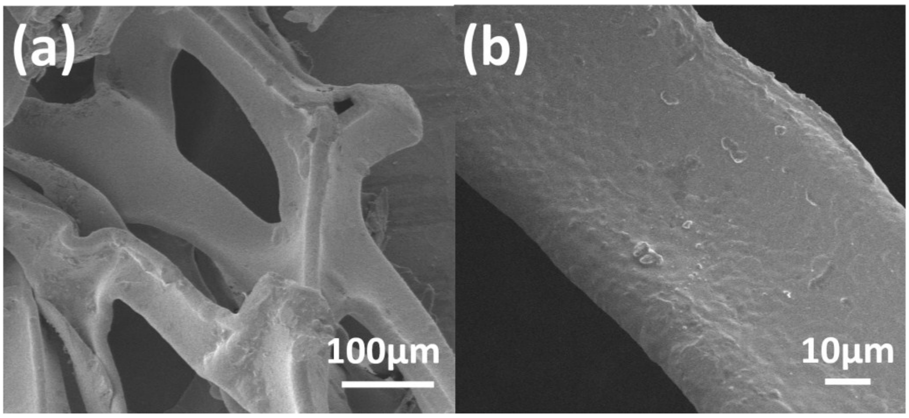


**Fig. S2** (a) and (b) SEM images of pristine Ni foam at different magnifications.

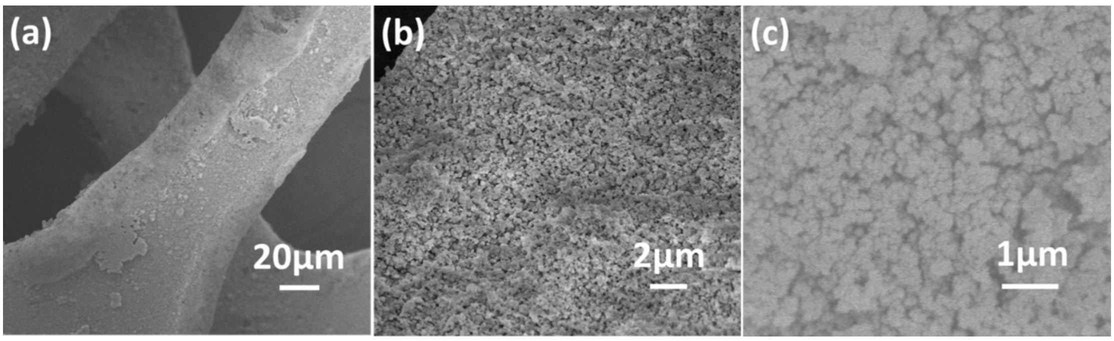


**Fig. S3** (a) and (b) SEM images of NiOH/NF at different magnifications.

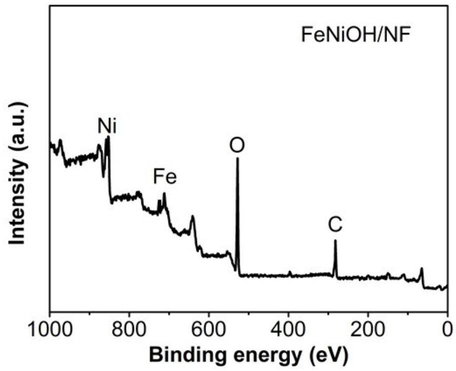


**Fig. S4** XPS survey scans of FeNiOH/NF.

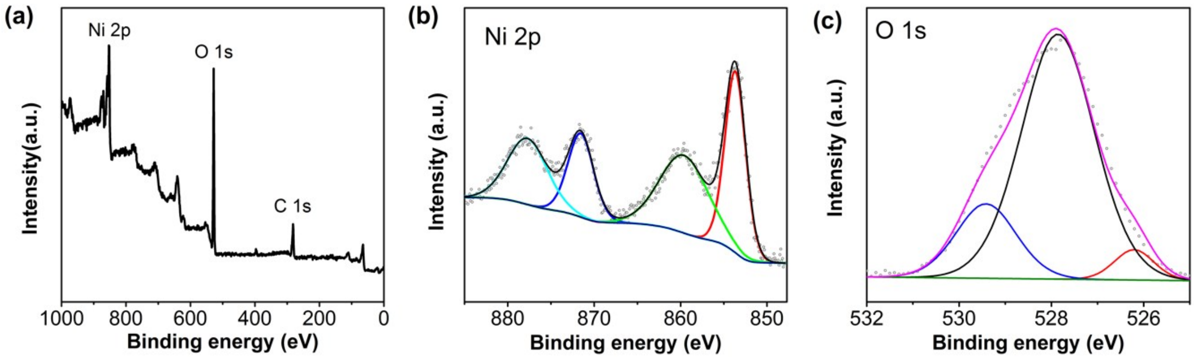


**Fig. S5** The XPS survey scans of NiOH/NF electrode (a), the corresponding Ni 2p (b) and O 1s (c) core level spectra. The high-resolution Ni 2p spectrum shows two major peaks at 853.5 and 871.6 eV, corresponding to Ni $2p_{3/2}$ and Ni $2p_{1/2}$, respectively, accompanying with other two satellite peaks at 859.8 and 877.8 eV. The high resolution spectrum of O 1s contains a strong peak at 527.9 eV which usually is attributed to oxygen-metal bond, and the other fitted peaks at 526.2 and 529.4 eV can be ascribed to oxygen in hydroxide group, and surface-adsorbed water molecule. The shift of binding energy of Ni 2p and O 1s of NiOH/NF as compared with FeNiOH/NF (Fig. 2) indicates the strong doping effect of Fe into $Ni(OH)_2$ lattice.

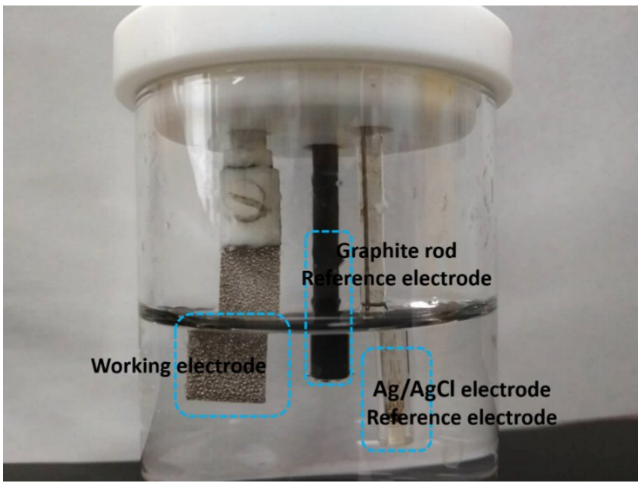


**Fig. S6** Optical image of electrocatalytic OER process about the three-electrode glass cell and the fabricated electrode was directly used as the working electrode.

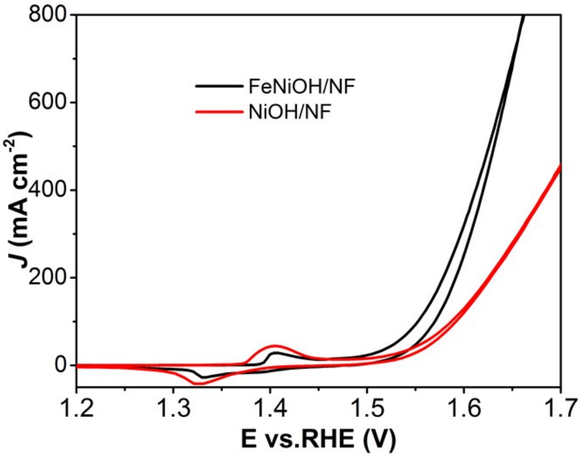


**Fig. S7** Cyclic voltammograms of FeNiOH/NF and NiOH/NF electrodes in $O_2$-saturated 1.0 M KOH.

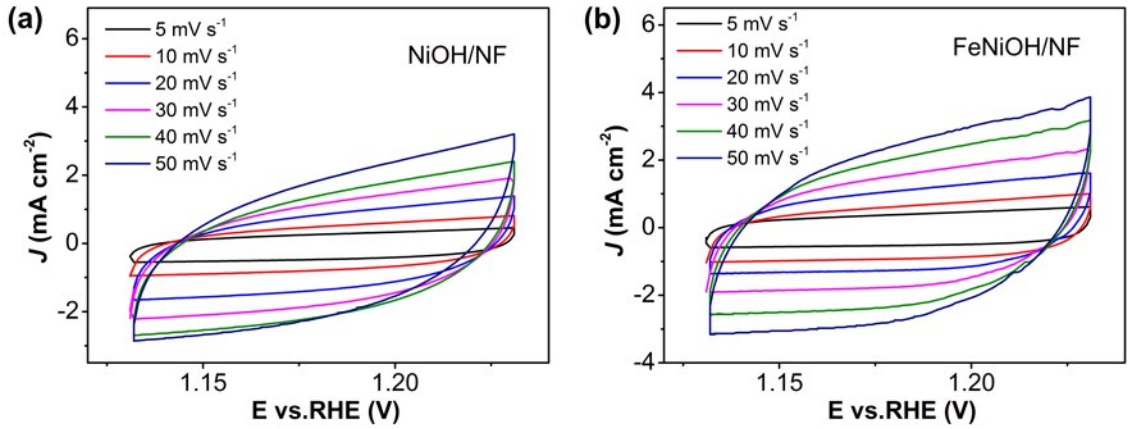


**Fig. S8** Cyclic voltammograms of (a) NiOH/NF and (b) FeNiOH/NF measured at scan rates from 5 to 50 mV $s^{-1}$.

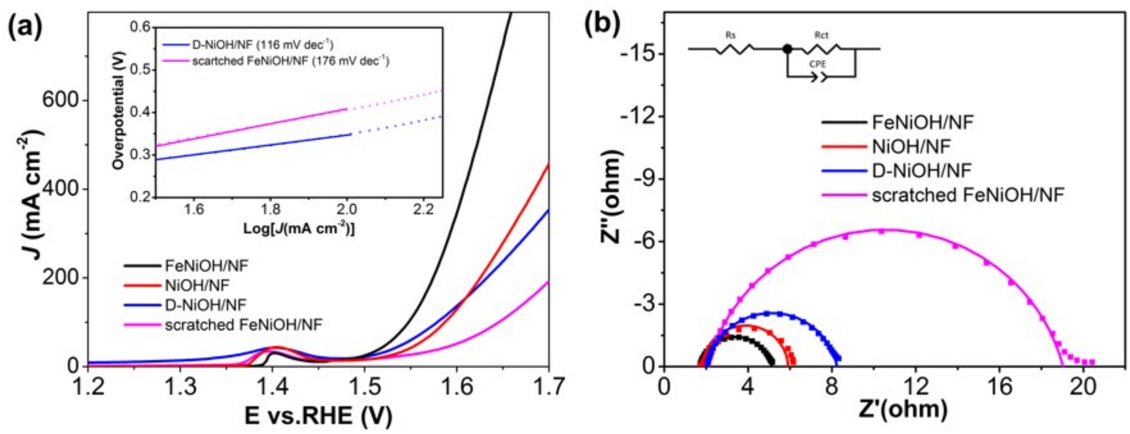


**Fig. S9** (a) Polarization curves and corresponding Tafel plots (*inset*) of FeNiOH/NF, D-NiOH/NF, and scratched FeNiOH/NF. (b) EIS of FeNiOH/NF, D-NiOH/NF, and scratched FeNiOH/NF recorded at 1.60 V. Inset in (b): the corresponding equivalent circuit diagram.

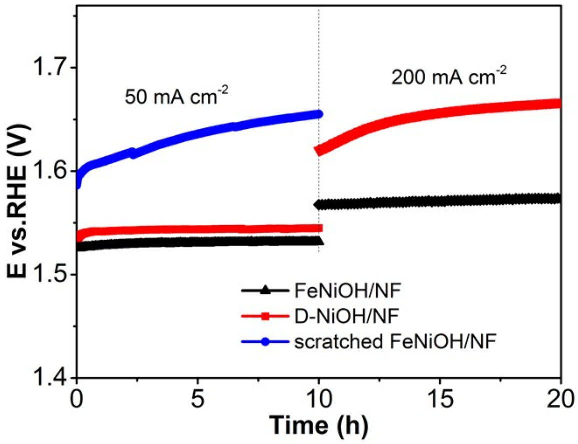


**Fig. S10** Chronopotentiometric curve of FeNiOH/NF, D-NiOH/NF, and scratched FeNiOH/NF at 50 and 200 mA cm$^{-2}$, respectively.

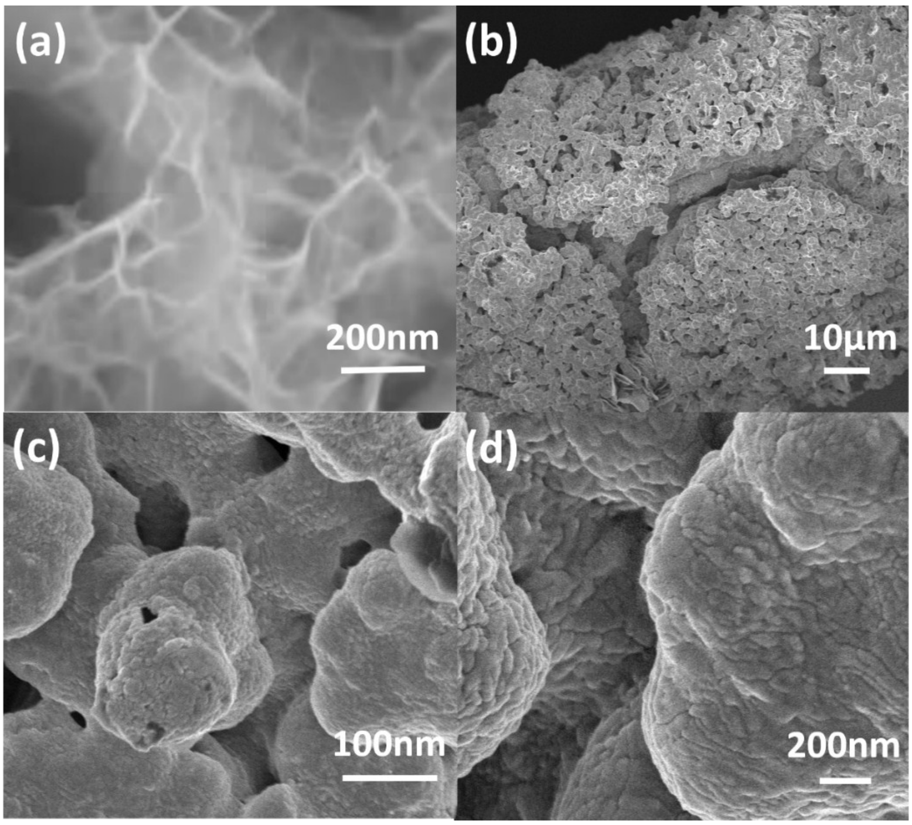


**Fig. S11** SEM images of D-NiOH/NF prepared by electrodeposition method (a) and after long-term OER stability test (b-d).

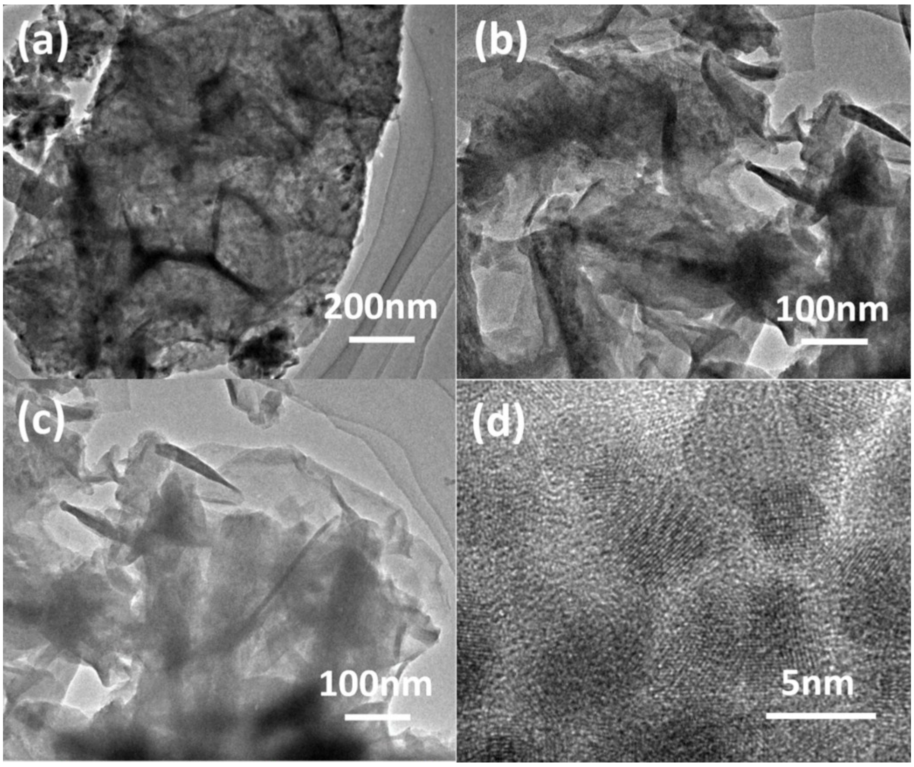


**Fig. S12** (a-c) TEM and (d) HR-TEM images of FeNiOH/NF after long–term OER stability test. It can be found that the nanosheet structure is well retained and the clear lattice fringes are observed, revealing the high structural stability of this integrated electrocatalyst after long-term OER stability test.

## 2. Supplementary Tables

**Table S1.** Comparison of OER performance of several recently reported nonprecious electrocatalytsts.

| Catalyst | Substrate | Electrolyte | $E_{J=10}$ (V) | $E_{J=20}$ (V) | Tafel slop (mV dec$^{-1}$) | Reference |
|---|---|---|---|---|---|---|
| Fe doped $Ni(OH)_2$ | Ni foam | 1.0 M KOH | N.A. | 1.50 | 99 | This work. |
| $Ni_5P_4/Ni_2P$-C | Glassy carbon | 1.0 M KOH | 1.53 | N.A. | 64 | [1] |
| Ni-doped FeOOH | FTO | 0.1 M NaOH | 1.66 | N.A. | N.A. | [2] |
| FeNi LDH/GO | Glassy carbon | 1.0 M KOH | 1.45 | N.A. | 39 | [3] |
| Au@$Co_3O_4$ | Glassy carbon | 0.1 M KOH | > 1.60 V | N.A. | 60 | [4] |
| FeOOH/$CeO_2$ nanotube | Ni foam | 1.0 M KOH | > 1.45 V | N.A. | 92.3 | [5] |
| CoP | Glassy carbon | 1.0 M KOH | 1.55 V | N.A. | 84 | [6] |
| $Co(OH)_2$ | Glassy carbon | 1.0 M KOH | 1.55 V | N.A. | 53 | [7] |
| NiFeMn layered double hydroxides | Glassy carbon | 1.0 M KOH | N.A. | 1.519 V | 47 | [8] |
| $NiCo_2O_4$ nanocages | Glassy carbon | 1.0 M KOH | 1.57 V | N.A. | 75 | [9] |
| $Co_3O_4$ nanosheet | Glassy carbon | 1.0 M KOH | 1.56 V | N.A. | 69 | [10] |
| NiO | Ni foam | 1.0 M KOH | 1.575 V | N.A. | 53 | [11] |
| NiCo-LDH nanosheets | Ni foam | 1.0 M KOH | > 1.53 V | N.A. | 113 | [12] |
| $NiCo_2O_4$ | Glassy carbon | 1.0 M KOH | 1.52 V | N.A. | 53 | [13] |
| $Co_3O_4$ nanowires | Glassy carbon | 1.0 M KOH | > 1.65 V | N.A. | 72 | [14] |

**Table S2.** Comparison of HER performance of several recently reported nonprecious electrocatalytsts.

| Catalyst | Substrate | Electrolyte | $E_{J=10}$(mV) vs. RHE | $E_{J=100}$(mV) vs. RHE | Tafel Slop (mV $dec^{-1}$) | Reference |
|---|---|---|---|---|---|---|
| Fe doped $Ni(OH)_2$ | Ni foam | 1.0 M KOH | 138 | 291 | 63.7 | This work. |
| NiS | Ni foam | 1.0 M KOH | 122 | 137 | 69 | [15] |
| $Ni_8P_3$ | Ni foam | 1.0 M KOH | 130 | N.A. | 58.5 | [16] |
| NiCoP | Ti film | 1.0 M KOH | 97 | 258 | 50 | [17] |
| $NiCo_2S_4$ | Ni foam | 1.0 M KOH | 240 | N.A. | 58.9 | [18] |
| $NiFe/NiCo_2O_4$ | Ni foam | 1.0 M KOH | 105 | N.A. | 88 | [19] |
| Co-P film | Copper film | 1.0 M KOH | 94 | 158 | 42 | [20] |
| $Ni_3S_2$ leaves | Ni foam | 1.0 M KOH | 182 | N.A. | 89 | [21] |
| NiS nanoframes | Ni foam | 1.0 M KOH | 94 | N.A. | N.A. | [22] |
| FeP | Carbon cloth | 1.0 M KOH | 218 | N.A. | 146 | [23] |
| CoP | Carbon cloth | 1.0 M KOH | 67 | N.A. | 129 | [24] |
| CoOx@CN | Glassy carbon | 1.0 M KOH | 232 | N.A. | N.A. | [25] |
| NiP/NF | Ni foam | 1.0 M KOH | 102 | 195 | 90 | [26] |
| Co-NRCNTs | Glassy carbon | 1.0 M KOH | 370 | N.A. | N.A. | [27] |
| $NiS_X$ | Ni foam | 1.0 M KOH | 60 | N.A | 99 | [28] |
| $Ni_2P$ | Ni foam | 1.0 M KOH | 98 | N.A. | 72 | [29] |
| $Ni_5P_4$ Films | Ni foil | 1.0 M KOH | 150 | N.A. | 53 | [30] |
| $Ni_3S_2$ | Ni foam | 1.0 M KOH | 292 | N.A. | N.A. | [31] |
| $CoS_2@MoS_2/RGO$ | Glassy carbon | 0.5 M $H_2SO_4$ | 98 | 175 | 37.4 | [32] |

**Table S3.** Comparison of several recently reported nonprecious OER/HER bifunctional electrocatalysts toward overall water splitting.

| Catalyst | Substrate | Electrolyte | $E_{J=10}$(V) | $E_{J=20}$(V) | Reference |
|---|---|---|---|---|---|
| Fe doped $Ni(OH)_2$ | Ni foam | 1.0 M KOH | 1.64 | 1.69 | This work. |
| NiS | Ni foam | 1.0 M KOH | 1.61 | 1.67 | [15] |
| NiCoP | Ti film | 1.0 M KOH | 1.65 | N.A. | [17] |
| $NiCo_2S_4$ | Ni foam | 1.0 M KOH | 1.63 | N.A. | [18] |
| $NiFe/NiCo_2O_4$ | Ni foam | 1.0 M KOH | 1.67 | N.A. | [19] |
| Co-P film | Copper film | 1.0 M KOH | > 1.62 | N.A. | [20] |
| $Ni_5P_4$/Ni foil | Ni foil | 1.0 M KOH | about 1.68 | N.A. | [30] |
| $Ni_3S_2$/Ni foam | Ni foam | 1.0 M KOH | about 1.70 | N.A. | [31] |
| NiSe | Ni foam | 1.0 M KOH | 1.63 | 1.75 | [33] |
| $Ni_2P$ | Ni foam | 1.0 M KOH | 1.63 | N.A. | [34] |
| NiP/Ni | Ni foam | 1.0 M KOH | 1.62 | 1.66 | [35] |
| CoO-CNF | Stainless steel | 1.0 M KOH | 1.63 | N.A. | [36] |
| Cu@NiFe LDH | Cu foam | 1.0 M KOH | 1.54 | N.A. | [37] |
| $CNTs/CoS_2$ | Carbon paper | 1.0 M KOH | 1.743 | N.A. | [38] |
| $Co_{0.85}Se$/NiFe-LDH | Exfoliated Graphene Foil | 1.0 M KOH | 1.67 | N.A. | [39] |
| NiFe LDH | Ni foam | 1.0 M KOH | 1.70 | N.A. | [40] |
| $Ni/Mo_2C$ | Ni foam | 1.0 M KOH | 1.66 | N.A. | [41] |
| NiSe | Ni foam | 1.0 M KOH | 1.63 | N.A. | [42] |
| $NiFeO_x$/CNF | Carbon nanofibres | 1.0 M KOH | 1.62 | N.A. | [36] |
| Ni@N doped graphene | Ni foam | 1.0 M KOH | 1.60 | N.A. | [43] |
| $MoNi_4$ | Ni foam | 1.0 M KOH | 1.58 | N.A. | [44] |

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