

Supplementary Information

One-step Synthesis of Amorphous Nickel Iron Phosphide Hierarchical Nanostructures for Water Electrolysis with Superb Stability at High Current Density

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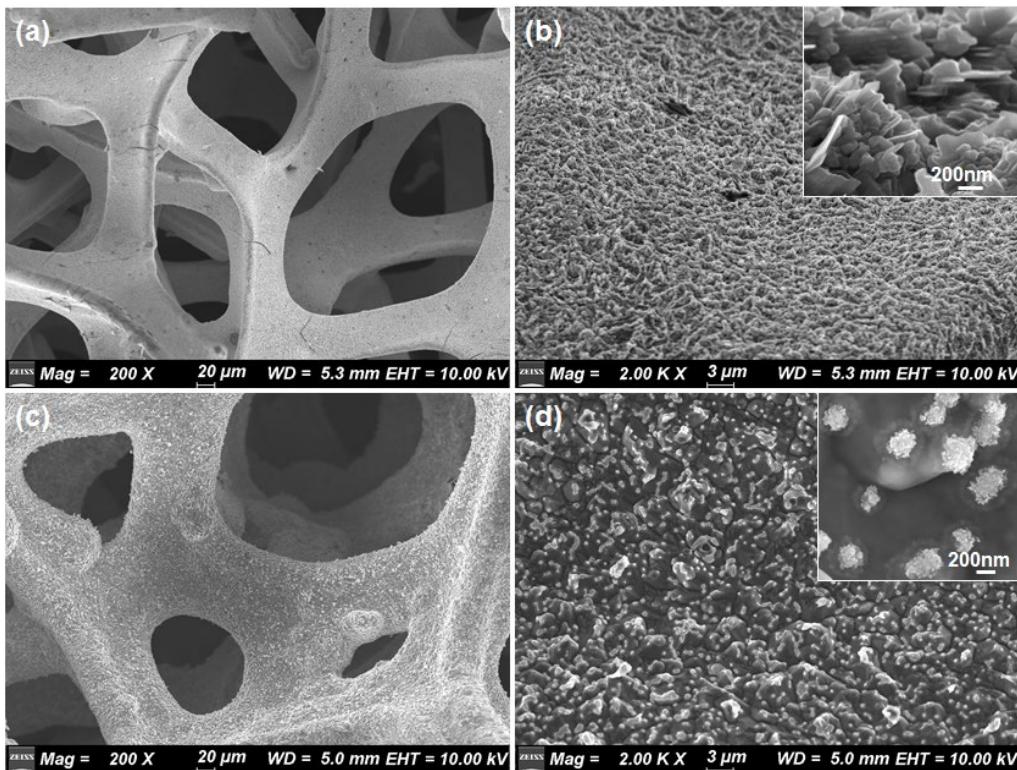


Fig. S1 SEM images of Ni₂P/NF (a,b) and Fe₂P/FF (c,d).

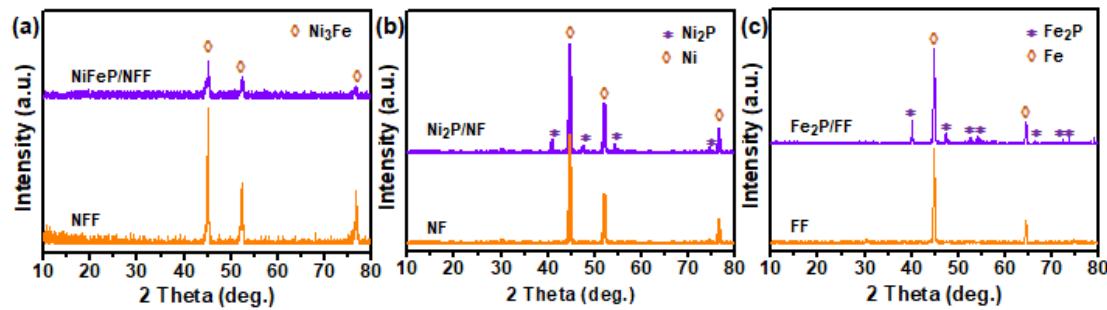


Fig. S2 XRD patterns of the as-prepared samples.

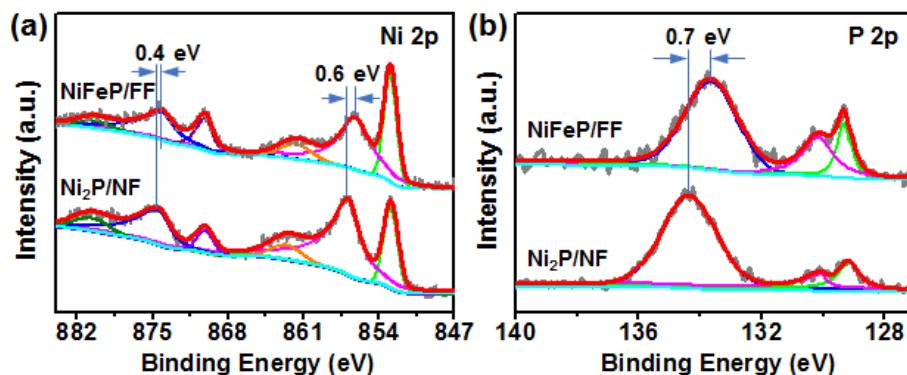


Fig. S3 Comparison of the high resolution Ni 2p (a) and P 2p (b) XPS spectra in the NiFeP/NFF and Ni₂P/NF samples.

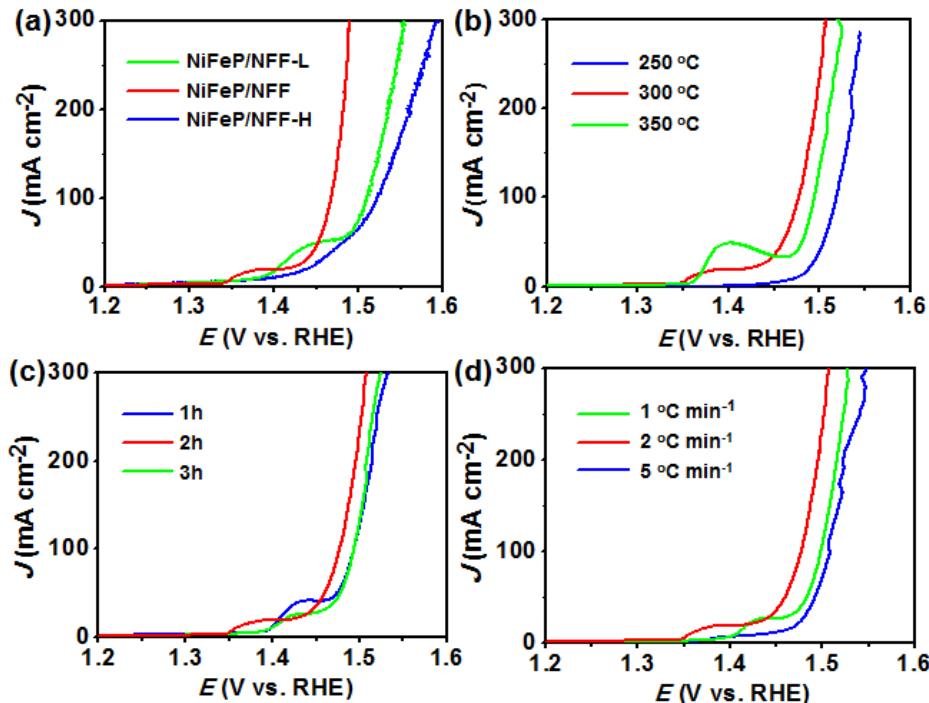


Fig. S4 Influence of the amount of NaH_2PO_2 (a), phosphidation temperature (b), phosphidation time (c) and heating rate (d) on the OER catalytic performance of the samples. NiFeP/NFF-L and NiFeP/NFF-H were obtained through the same procedure for preparation of NiFeP/NFF by adjusting the amount of NaH_2PO_2 to be 0.3 and 0.5 g, respectively.

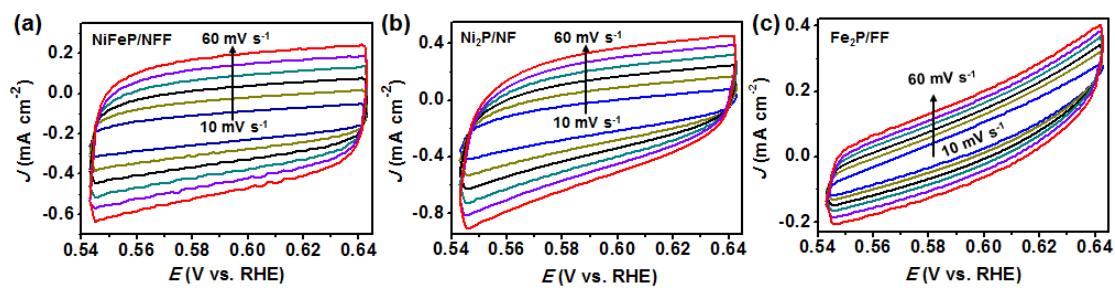


Fig. S5 CV curves of the samples at various scan rates during OER process.

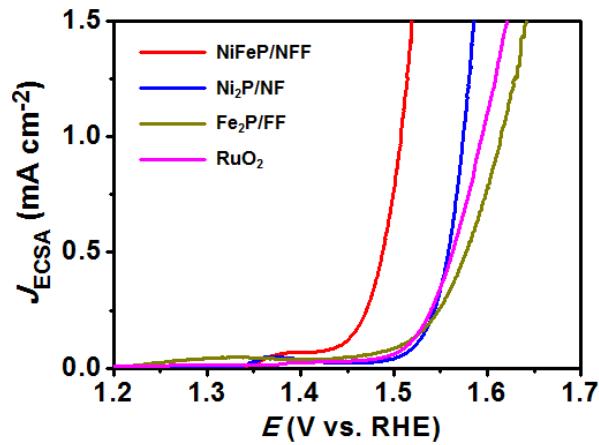


Fig. S6 ECSA-normalized LSV curves of different catalysts.

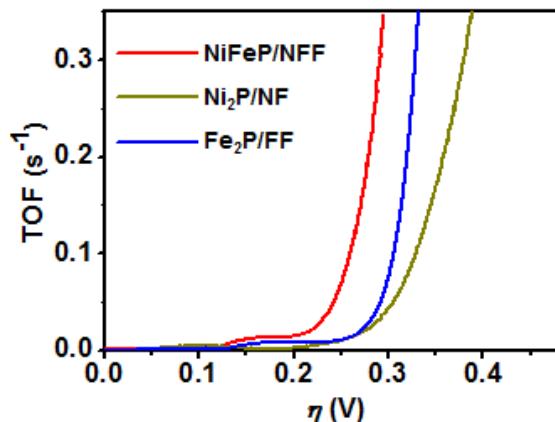


Fig. S7 TOF plots as a function of overpotential of different catalysts during OER process.

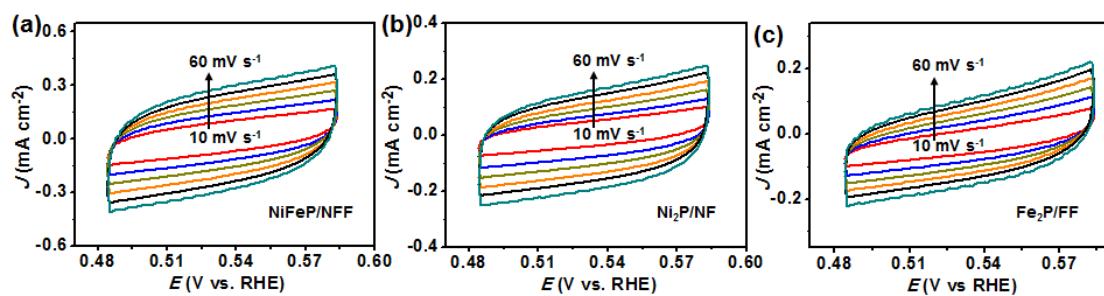


Fig. S8 CV curves of the samples at various scan rates during the HER process.

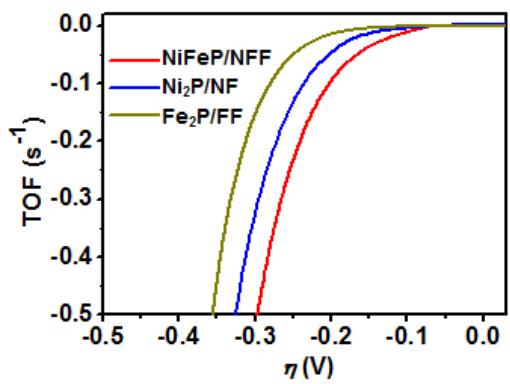


Fig. S9 TOF plots as a function of overpotential of different catalysts during HER process.

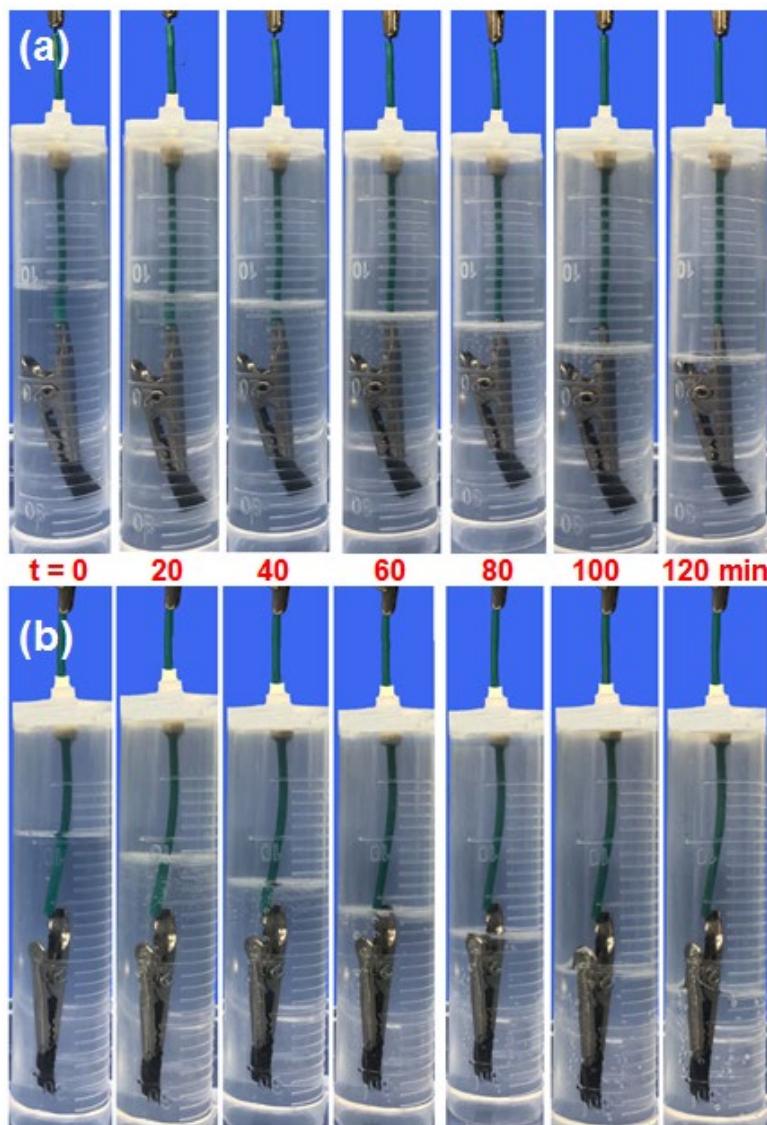


Fig. S10 Collection of oxygen (a) and hydrogen (b) evolved from water electrolysis at a current density of 30 mA cm⁻² by water drainage method.

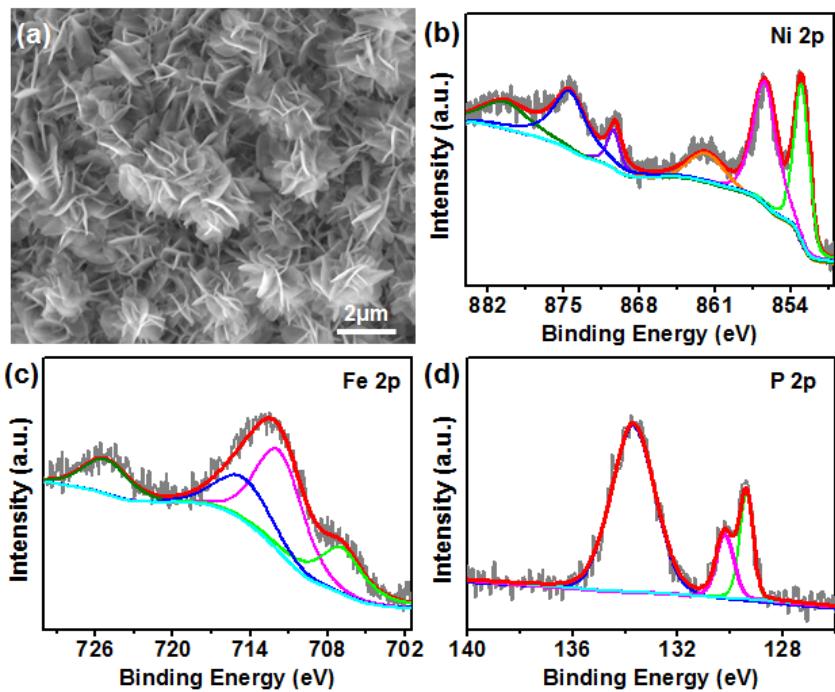


Fig. S11 SEM image (a) and high resolution XPS spectra (b-d) of the NiFeP/NFF cathode after overall water splitting test over 1000 h.

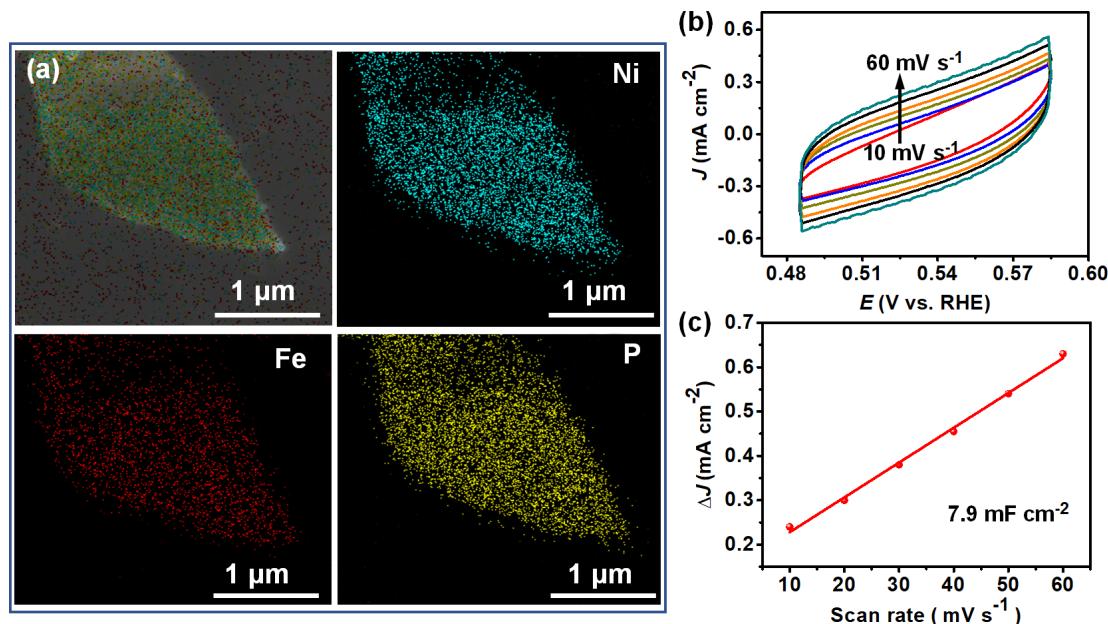


Fig. S12 EDS mapping image (a), CV curves (b) and the non-Faradaic capacitive currents against the scan rates (c) of the NiFeP/NFF cathode after overall water splitting test over 1000 h.

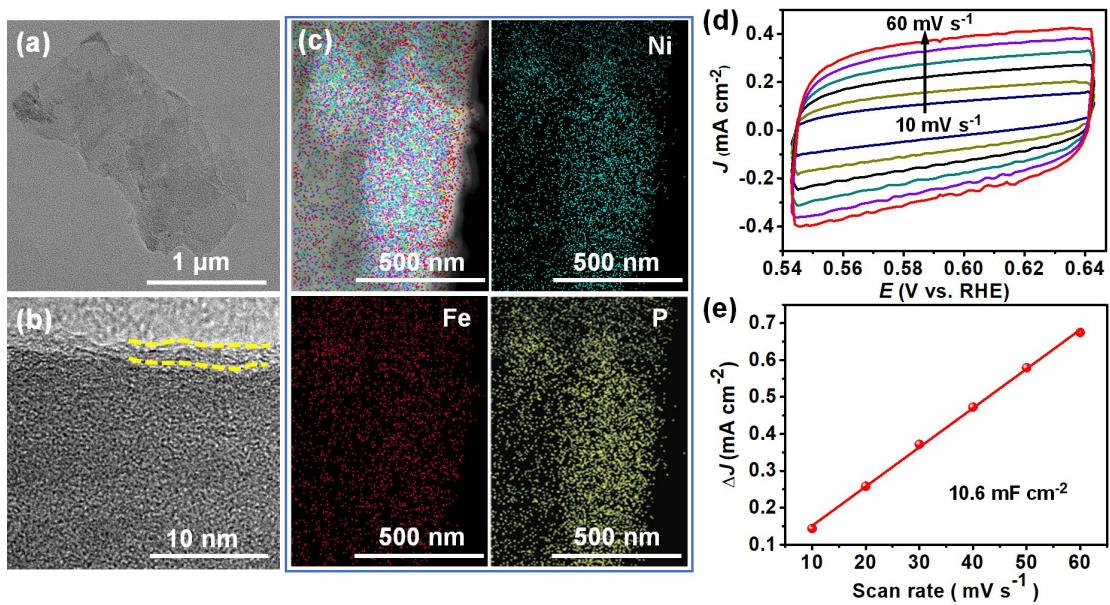


Fig. S13 TEM images (a,b), EDS mapping image (c), CV curves (d) and the non-Faradaic capacitive currents against the scan rates (e) of the NiFeP/NFF anode after overall water splitting test over 1000 h.

Table S1 Comparison of the OER performance of NiFeP/NFF with other reported non-precious electrocatalysts in 1 M KOH.

| Catalyst | Substrate | η_{20} (mV) | Tafel slope (mV dec ⁻¹) | Ref. |
|--|--------------------|---------------------|--|-----------|
| (Fe-Ni)P@CC@PC-E-15 | graphite electrode | 215 | 38 | 1 |
| Ni-Fe-P/Ni/CS(f.1) | copper sheet | 232 | 60 | 2 |
| Fe _{0.5} Ni _{1.5} P/PC | glassy carbon | 254 | 76 | 3 |
| Co/Ce-Ni ₃ S ₂ /NF | Ni foam | 286 | 71.7 | 4 |
| NiFeP@NPC | glass carbon | 398 | 78 | 5 |
| 3DOM Ni ₃ Fe-P | glassy carbon | 248 | 40 | 6 |
| NiFeP | carbon cloth | 281 | 74 | 7 |
| Ni _{0.9} Co _{0.1} P@NNCs | Cu | 236 | 54 | 8 |
| Ni ₂ P/(NiFe) ₂ P(O)NAs | Ni foam | 181 | 60 | 9 |
| NiFe LDH/Co _{1-x} S | Ni foam | 267 | 41.67 | 10 |
| Ni ₂ P-Fe ₂ P/NF | Ni foam | 230 | 58 | 11 |
| Fe ₃₅ Ni ₃₅ Co ₁₀ P ₂₀ | self-supported | 302 | 38 | 12 |
| hcp-NiFe@NC | carbon cloth | 245 | 41 | 13 |
| CoP-FeP/CC | carbon cloth | 300 | 131 | 14 |
| Mo-doped Ni ₂ P | glassy carbon | 270 | 68.5 | 15 |
| C-(Fe-Ni)P@PC/(Ni-Co)P@CC | graphite | 266 | 56 | 16 |
| FeNi-P/NF | Ni foam | 257 | 72 | 17 |
| FeP-Ni/NF | Ni foam | 241 | 60 | 18 |
| FeNiP-NP | Ni foam | 199 | 76 | 19 |
| Ni-Fe-P-B | glassy carbon | 283 | 38 | 20 |
| NiFeP/NFF | Ni-Fe foam | 189 | 37.2 | This work |

Table S2 Comparison of the HER performance of NiFeP/NFF with other reported non-precious electrocatalysts in 1 M KOH.

| Catalyst | Substrate | η_{10} (mV) | Tafel slope (mV dec ⁻¹) | Ref. |
|---|--------------------|---------------------|--|-----------|
| NiFeP | glassy carbon | 690 | 116 | 21 |
| Cr-doped FeNi-P/NCN | glassy carbon | 190 | 68.51 | 22 |
| NiFeP@C | glassy carbon | 160 | 75.8 | 23 |
| Fe _{0.5} Ni _{1.5} P/PC | glassy carbon | 200 | \ | 3 |
| FeCoNiP@NC | glassy carbon | 187 | 52.2 | 24 |
| NiFeP | glassy carbon | 182 | 69 | 25 |
| Ni-Fe-P-B | glassy carbon | 220 | 63 | 20 |
| Ni-Fe-P-300 | Ni foam | 192 | 142.2 | 26 |
| NiFeOF | self-supported | 253 | 96 | 27 |
| NiFe LDH@NiCoP/NF | Ni foam | 120 | 88.2 | 28 |
| FeNiSe-NS/EG | graphene foil | 187 | 65 | 29 |
| NiCoP/rGO | carbon fiber paper | 209 | 124.1 | 30 |
| NiFe LDH@CoP/NiP ₃ | Ni foam | 151 | 74 | 31 |
| Fe-Ni ₃ C-2% | glassy carbon | 244 | 41.3 | 32 |
| 3DOM Ni ₃ Fe-P | glassy carbon | 120 | 61 | 6 |
| CoFe@NiFe-200/NF | Ni foam | 240 | 88.88 | 33 |
| Ni ₃ S ₂ /NF | Ni foam | 189 | 89.3 | 34 |
| Ni ₂ P-Fe ₂ P/NF | Ni foam | 128 | 86 | 11 |
| Fe _{1.0} Co _{1.1} Ni _{1.4} -NC | glassy carbon | 175 | 168 | 35 |
| CoFeN _x -500 HNAs/NF | Ni foam | 200 | 66.04 | 36 |
| NiFeP/NFF | Ni-Fe foam | 155 | 67.8 | This work |

Table S3 Comparison of the overall water splitting performance of NiFeP/NFF with other reported bifunctional electrocatalysts in 1 M KOH.

| Catalyst | Substrate | Voltage at J_{10} (V) | Durability (h) @ J (mA cm $^{-2}$) | Ref. |
|---|----------------|-------------------------|---------------------------------------|-----------|
| NiSe ₂ /3DSNG/NF | Ni foam | 1.59 | 50 @ 20 | 37 |
| FeNi/PNGs | polyamide film | 1.67 | 17 @ 10 | 38 |
| Fe _{0.5} Ni _{1.5} P/PC | glassy carbon | 1.63 | 16 @ 10 | 3 |
| MnCo ₂ O ₄ @ Ni ₂ P | Ni foam | 1.63 | 30 @ 10 | 39 |
| Ni/Ni(OH) ₂ | carbon paper | 1.59 | 20 @ 10 | 40 |
| Fe _{0.29} Co _{0.71} P/NF | Ni foam | 1.59 | 10 @ 180 | 41 |
| NiFe LDH@CoP/NiP ₃ | Ni foam | 1.64 | 275 @ 100 | 31 |
| NiFeSP/NF | Ni foam | 1.58 | 20 @ 10 | 42 |
| NiFe-NCNT@MoS ₂ -12 | Ni foam | 1.6 | 12 @ 10 | 43 |
| Fe, Al-NiSe ₂ /rGO | Ni foam | 1.7 | 22 @ 10 | 44 |
| Ni ₂ P-Fe ₂ P/NF | Ni foam | 1.561 | 43 @ 500 | 11 |
| C-(Fe-Ni)P@PC/(Ni-Co)P | graphite | 1.63 | 24 @ 10 | 16 |
| np-NiFeCoP | self-supported | 1.62 | 20 @ 10 | 45 |
| CoFeN _x -500 HNAs/NF | Ni foam | 1.592 | 40 @ 10 | 36 |
| Pt/NiO/Ni/CNT-3 | carbon paper | 1.61 | 10 @ 10 | 46 |
| FeCoNiP@NC | Ni foam | 1.73 | 10 @ 10 | 24 |
| Co/CNFs(1000) | self-supported | 1.69 | 10 @ 10 | 47 |
| FeP-Ni/NF | Ni foam | 1.62 | 28 @ 20 | 18 |
| Co/ β -Mo ₂ C@N-CNTs | Ni foam | 1.64 | 24 @ 10 | 48 |
| CoP/NCNHP | carbon paper | 1.64 | 26 @ 20 | 49 |
| Ni-Fe-P-B | CFP | 1.58 | 12 @ 100 | 20 |
| np-(Ni _{0.67} Fe _{0.33}) ₄ P ₅ | self-supported | 1.62 | 20 @ 10 | 50 |
| NiCoP/rGO | CFP | 1.59 | 75 @ 10 | 30 |
| 3DOM Ni ₃ Fe-P | glassy carbon | 1.65 | 20 @ 10 | 6 |
| NiFe NTAs-NF | Ni foam | 1.62 | 20 @ 10 | 51 |
| Ni _{1.85} Fe _{0.15} P NSAs/NF | Ni foam | 1.61 | 22 @ 40 | 52 |
| FeNi/N-doped graphene | graphite | 1.701 | 10 @ 10 | 53 |
| E-Mo–NiCoP-3 | carbon cloth | 1.61 | 12 @ 50 | 54 |
| Ni _{0.7} Fe _{0.3} PS ₃ @MXene | Ni foam | 1.65 | 50 @ 10 | 55 |
| NiFeP/NFF | Ni-Fe foam | 1.58 | 1000 @ 500 | This work |

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