

Supporting information

**Heterointerface engineering of NiFe (oxy)hydroxides/CNTs
by *In situ* anchoring of sub-nano Au for efficient water
oxidation**

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Additional Figures and Tables

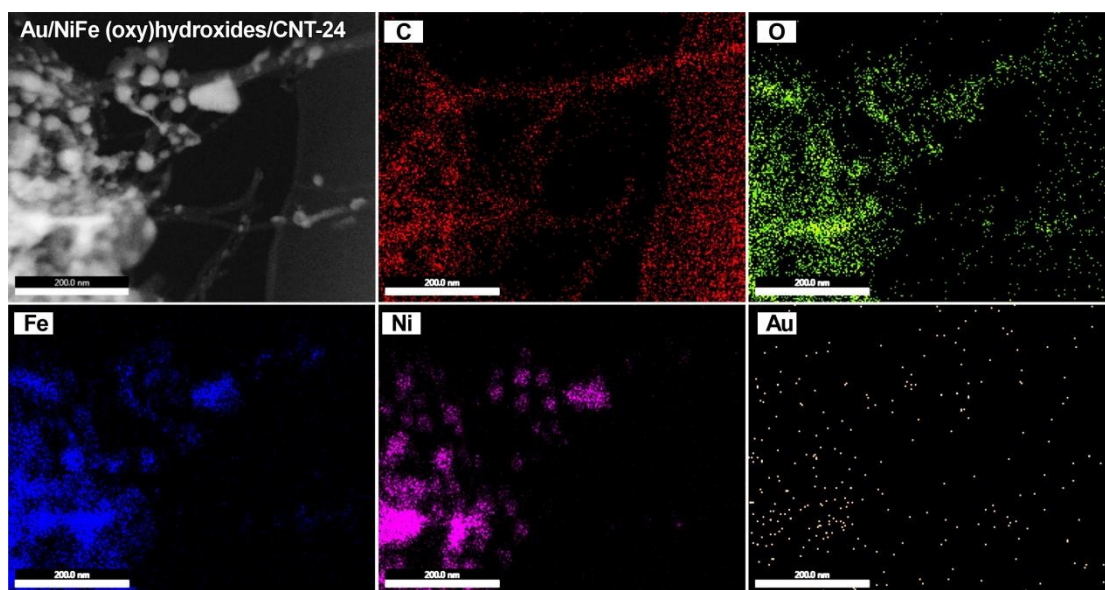


Fig. S1. High-resolution TEM image and EDS elemental mapping of Au/NiFe (oxy)hydroxides/CNTs-24.

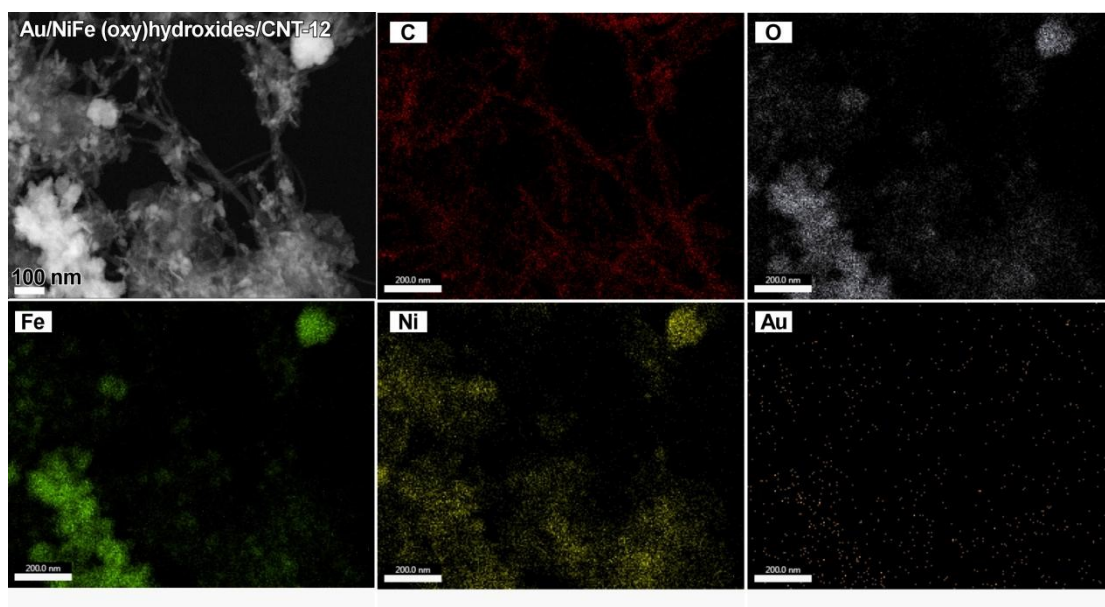


Fig. S2. High-resolution TEM image and EDS elemental mapping of Au/NiFe (oxy)hydroxides/CNTs-12.

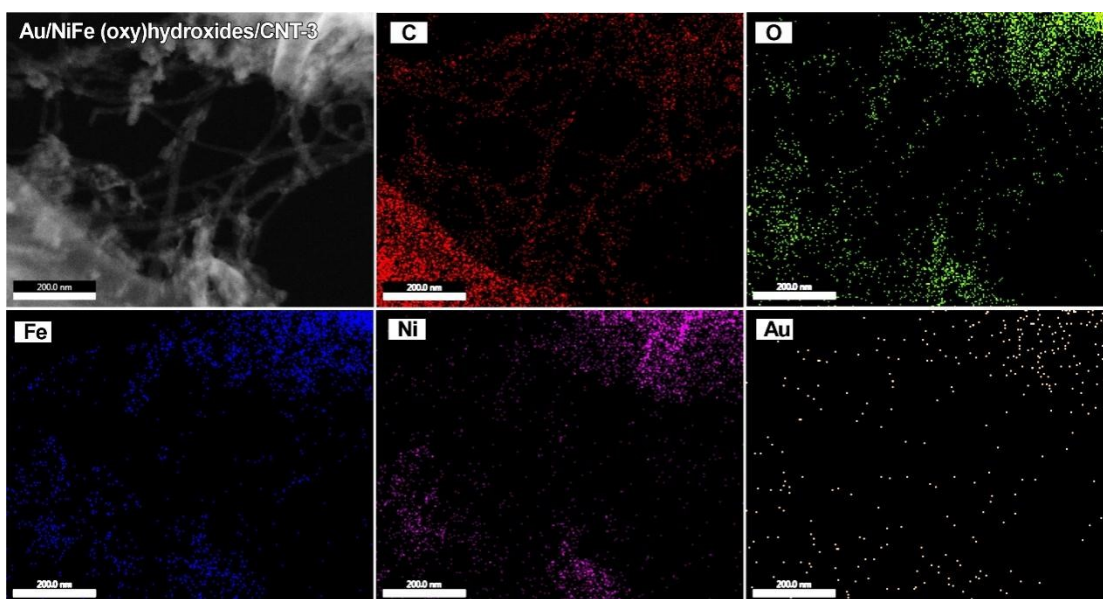


Fig. S3. High-resolution TEM image and EDS elemental mapping of Au/NiFe (oxy)hydroxides/CNTs-3.

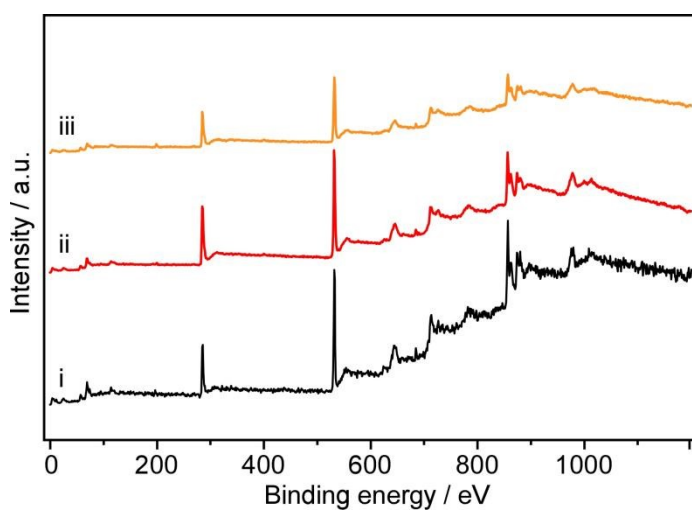


Fig. S4. XPS full spectra of (i) as-synthesized $\text{Ni}_{1/2}\text{Fe}_{1/2}(\text{OH})_2/\text{CNTs}$, (ii) Au/NiFe (oxy)hydroxides/CNTs-24 and (iii) Au/NiFe (oxy)hydroxides/CNTs-3, respectively.

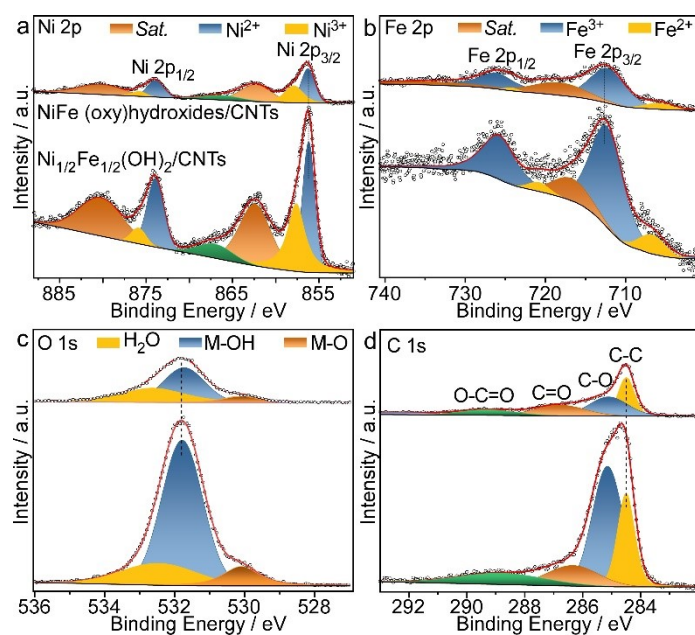


Fig. S5. (a) Ni 2p XPS spectra, (b) Fe 2p XPS spectra, (c) O1s XPS spectra and (d) C1s XPS spectra of the as-synthesized $\text{Ni}_{1/2}\text{Fe}_{1/2}(\text{OH})_2/\text{CNTs}$ (bottom) and NiFe (oxy)hydroxides/CNTs (top), respectively.

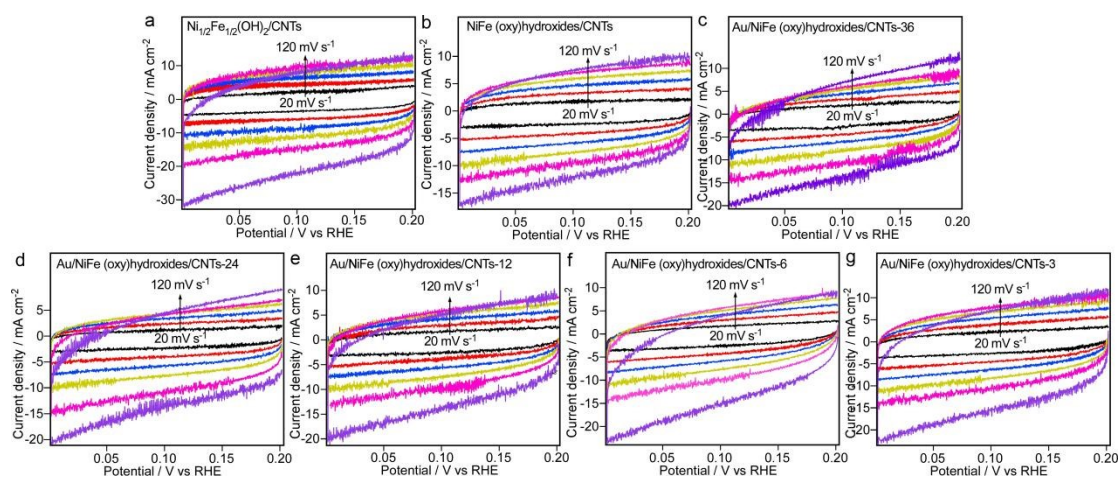


Fig. S6. Cyclic voltammograms of (a) as-synthesized $\text{Ni}_{1/2}\text{Fe}_{1/2}(\text{OH})_2/\text{CNTs}$, (b) NiFe (oxy)hydroxides/CNTs, (c) Au/NiFe (oxy)hydroxides/CNTs-36, (d) Au/NiFe (oxy)hydroxides/CNTs-24, (e) Au/NiFe (oxy)hydroxides/CNTs-12, (f) Au/NiFe (oxy)hydroxides/CNTs-6 and (g) Au/NiFe (oxy)hydroxides/CNTs-3 at different scan rate of 20, 40, 60, 80, 100, 120 mV s^{-1} , respectively.

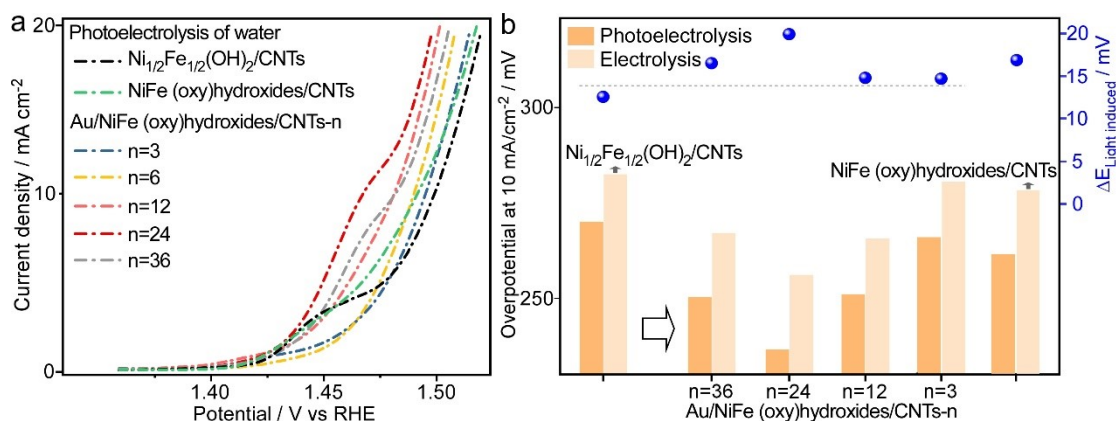


Fig. S7. (a) Photoelectrochemical OER LSV curves of the as-synthesized $\text{Ni}_{1/2}\text{Fe}_{1/2}(\text{OH})_2/\text{CNTs}$, $\text{NiFe (oxy)hydroxides}/\text{CNTs}$, and $\text{Au/NiFe (oxy)hydroxides}/\text{CNTs-n}$, respectively. (b) (left axis) Comparison of the OER overpotentials obtained from the photoelectrolysis and electrolysis of water of the as-synthesized $\text{Ni}_{1/2}\text{Fe}_{1/2}(\text{OH})_2/\text{CNTs}$, $\text{NiFe (oxy)hydroxides}/\text{CNTs}$, and $\text{Au/NiFe (oxy)hydroxides}/\text{CNTs-n}$, respectively, and (right axis) the corresponding photo-induced lower of the overpotentials.

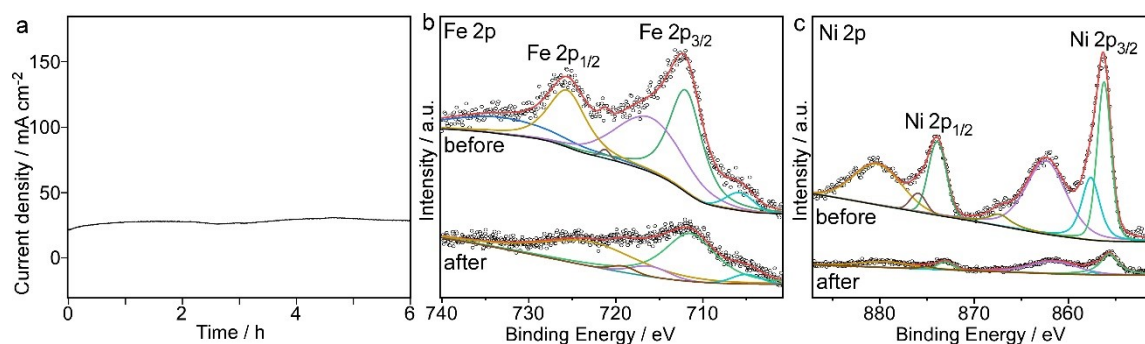


Fig. S8. (a) $J-t$ curve $\text{Au/NiFe (oxy)hydroxides}/\text{CNTs-24}$ photoanode measured in 1.0 M KOH under illumination of the simulated solar light. (b) Fe 2p and (c) Ni 2p XPS spectra of $\text{Au/NiFe (oxy)hydroxide}/\text{CNTs-24}$ before and after 1000 cycles of CV scans under the simulated solar light irradiation.

Table S1. The ratio of $\text{Ni}^{3+}/\text{Ni}^{2+}$, and $\text{Fe}^{3+}/\text{Fe}^{2+}$ in the surface of catalysts.

Catalysts	$\text{Ni}^{3+}/\text{Ni}^{2+}$	$\text{Fe}^{3+}/\text{Fe}^{2+}$
$\text{Ni}_{1/2}\text{Fe}_{1/2}(\text{OH})_2/\text{CNTs}$	0.71	5.68
$\text{Au/NiFe (oxy)hydroxides}/\text{CNTs-24}$	1.17	12.19
$\text{Au/NiFe (oxy)hydroxides}/\text{CNTs-3}$	0.36	8.08
$\text{O-Ni}_{1/2}\text{Fe}_{1/2}(\text{OH})_2/\text{CNTs}$	0.62	10.75

Table S2. Comparison of η_{10} of Au/NiFe(oxy)hydroxides/CNTs-24 with other NiFe (oxy)hydroxides-based electrocatalysts reported previously for water splitting.

Catalyst	Electrolyte	η_{10} (mV)	Ref.s
Au/NiFe(oxy)hydroxides/CNTs-24	1.0 M KOH	256.1	This work
NiFe-27%	1.0 M KOH	306	<i>Nat Commun</i> 13 , 2191 (2022)
MoNiFe-27% (oxy)hydroxide	1.0 M KOH	242	<i>Nat Commun</i> 13 , 2191 (2022)
NiCoFe-B _i	1.0 M KOH	300	<i>Nat Commun</i> 12 , 5980 (2021)
NiFe LDH	1.0 M KOH	348	<i>Nat Commun</i> 11 , 2522 (2020)
CoFe LDH	1.0 M KOH	404	<i>Nat Commun</i> 11 , 2522 (2020)
NiFe-oxyhydroxide/C	1.0 M KOH	269.6	<i>J. Mater. Chem. A</i> 10 , 10342 (2022)