

Hierarchically structured multi-shell nanotube arrays by self-assembly for efficient water oxidation

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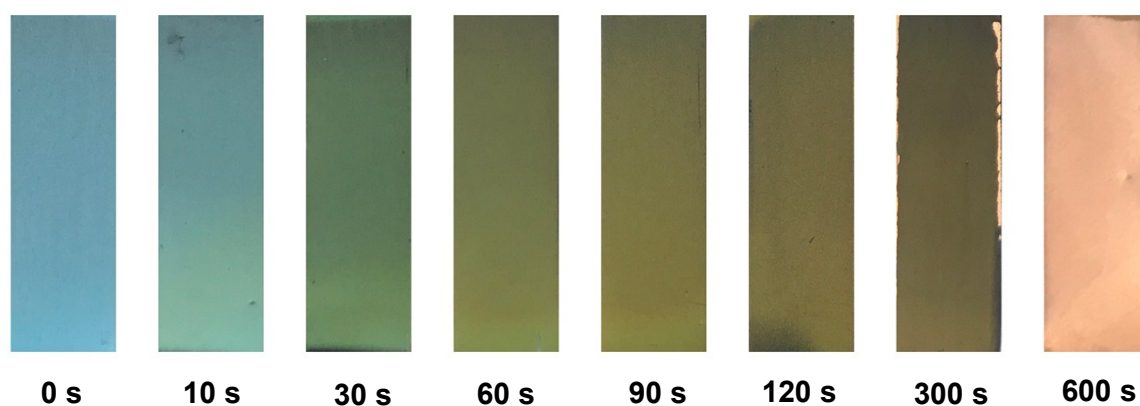


Figure S1. Camera pictures of $\text{Cu}(\text{OH})_2$ NWs after immersion in 10 mM Fe^{3+} solution for different times. ($\text{Fe}:\text{Cu}(\text{OH})_2$ NTs with different contents of iron). The electrode color changes from blue (0 s) over green (30 s) to dark yellow (90 s); beyond that, the nanowires peel off from the copper foil surface (600 s).

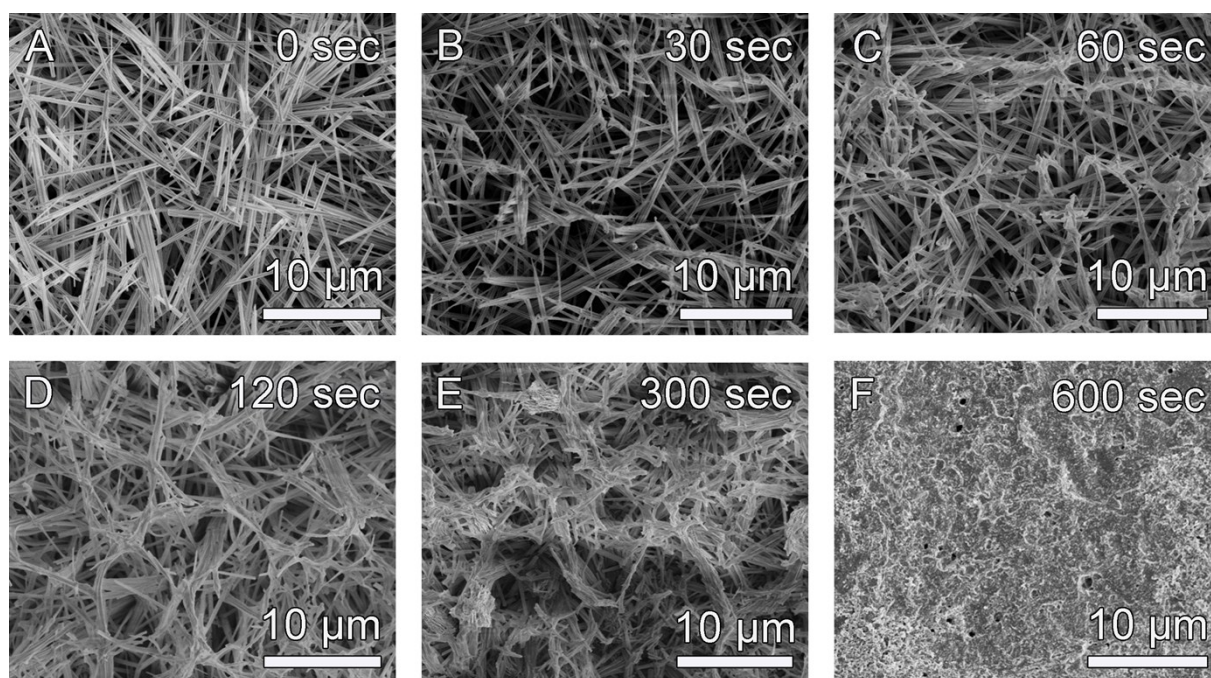


Figure S2. SEM images of Cu(OH)₂ NWs after immersion in 10 mM Fe³⁺ solution for different times. (Fe:Cu(OH)₂ NTs with different contents of iron).

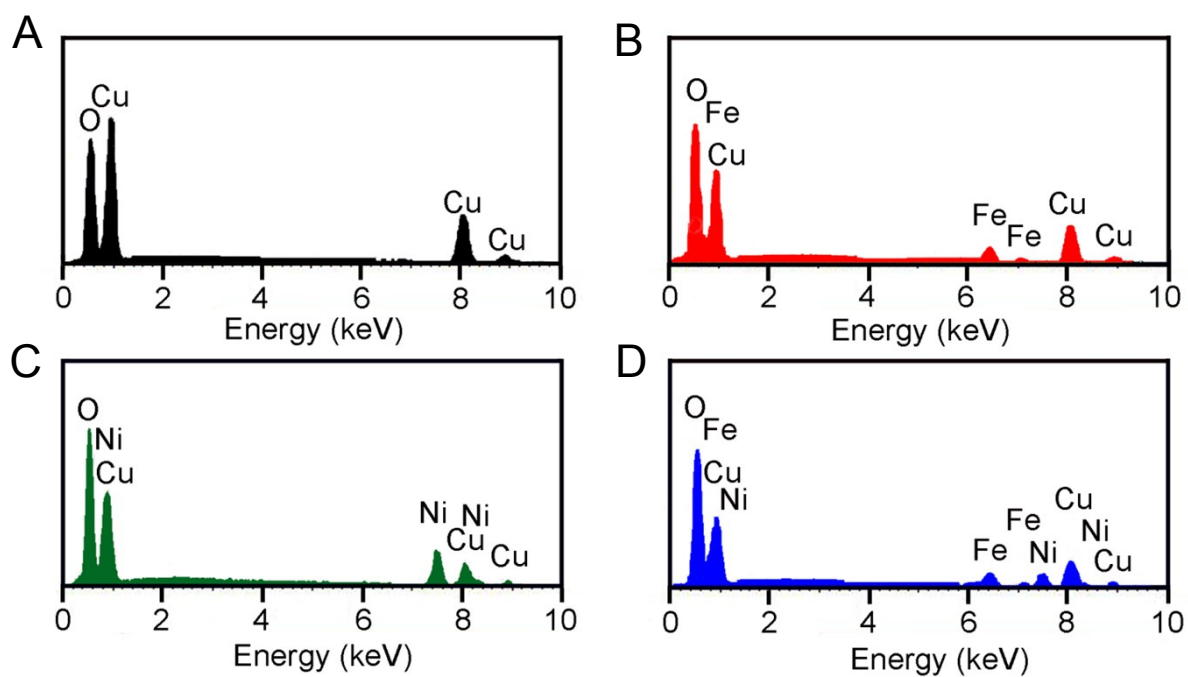


Figure S3. EDX of Cu(OH)₂ NWs (A), Fe:Cu(OH)₂ NTs (B), Ni:Cu(OH)₂ NWs (C) and Ni:Fe:Cu(OH)₂ NTs (D).

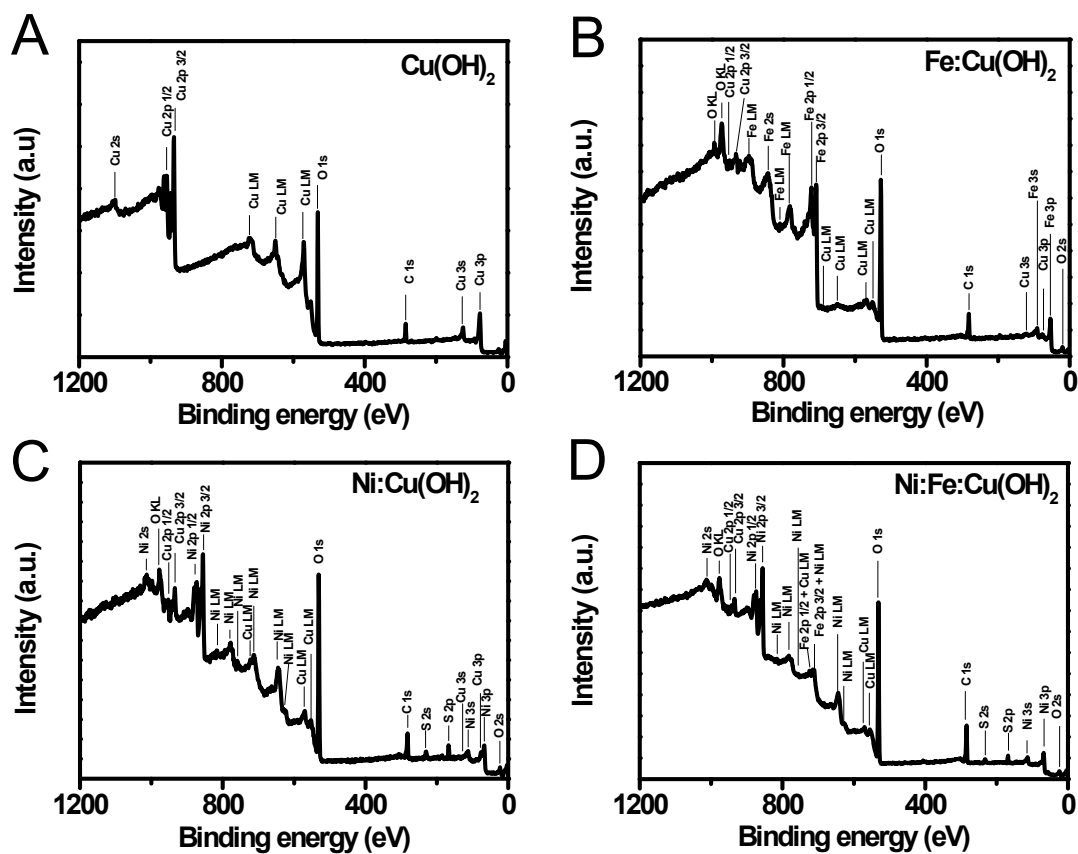


Figure S4. Survey XPS of Cu(OH)_2 NWs (A), Fe:Cu(OH)_2 NTs (B), Ni:Cu(OH)_2 NWs (C) and Ni:Fe:Cu(OH)_2 NTs.

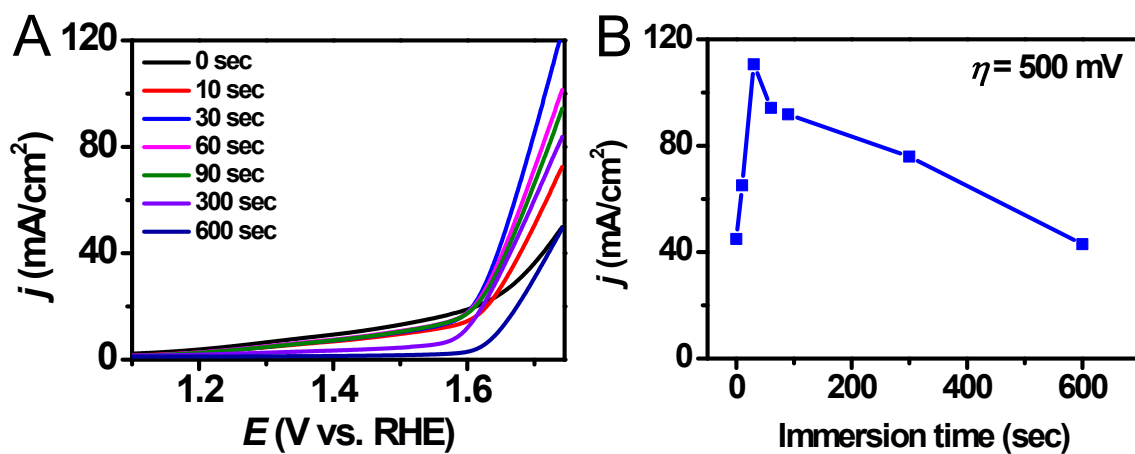


Figure S5. Optimization of Fe:Cu(OH)₂ NTs towards the immersion time of Cu(OH)₂ NWs into Fe³⁺ solution. LSV curves (A) and summarized current density vs. the immersion time at an overpotential of 500 mV (B). All electrochemical measurements were carried out in 1 M KOH at a scan rate of 20 mV/s with iR compensation.

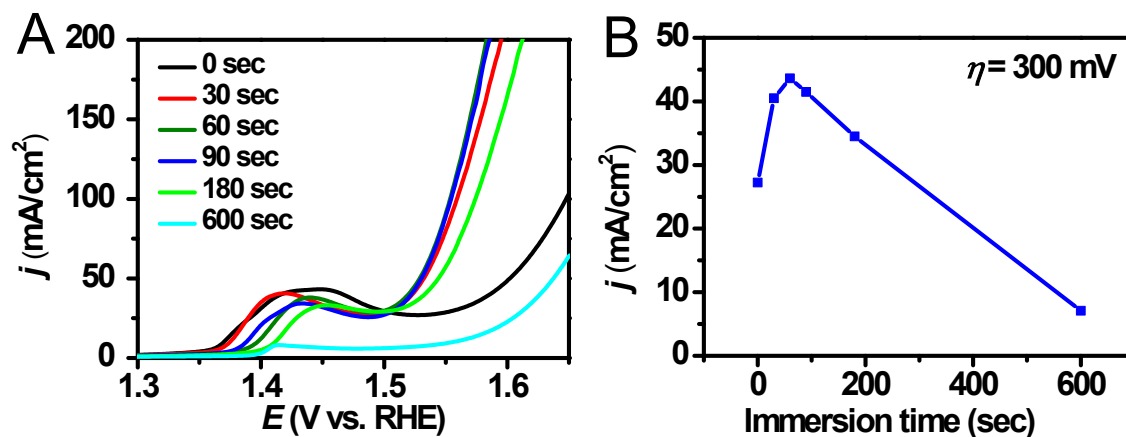


Figure S6. Optimization of Ni:Fe:Cu(OH)₂ NTs towards the immersion time of Cu(OH)₂ NWs into Fe³⁺ solution. All samples were put into the nickel chemical bath for 20 min afterwards. LSV curves (A) and summarized current density vs. the immersion time at an overpotential of 300 mV (B). All electrochemical measurements were carried out in 1 M KOH at a scan rate of 20 mV/s with iR compensation.

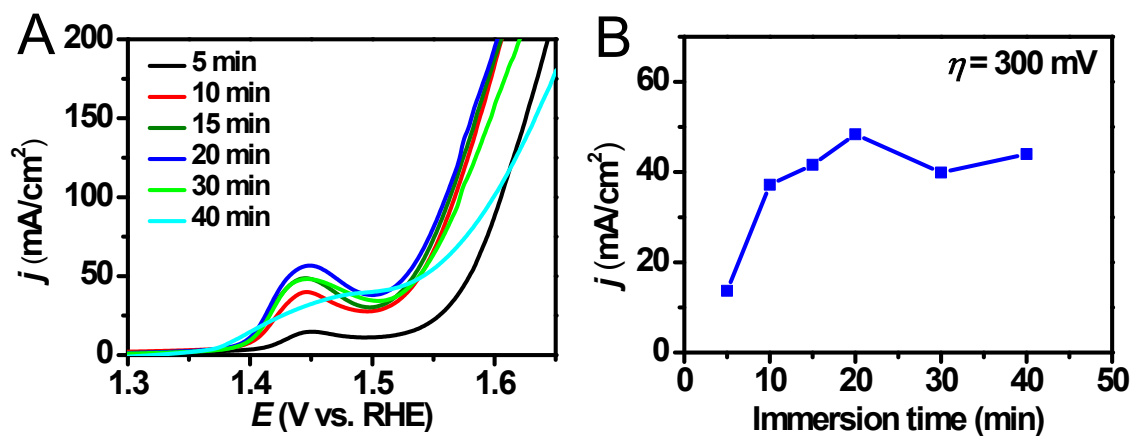


Figure S7. Optimization of Ni:Fe:Cu(OH)₂ NTs towards the immersion time of Fe:Cu(OH)₂ into the nickel chemical bath. LSV curves (A) and current density vs. immersion time at an overpotential of 300 mV (B). All electrochemical measurements were carried out in 1 M KOH at a scan rate of 20 mV/s with iR compensation.

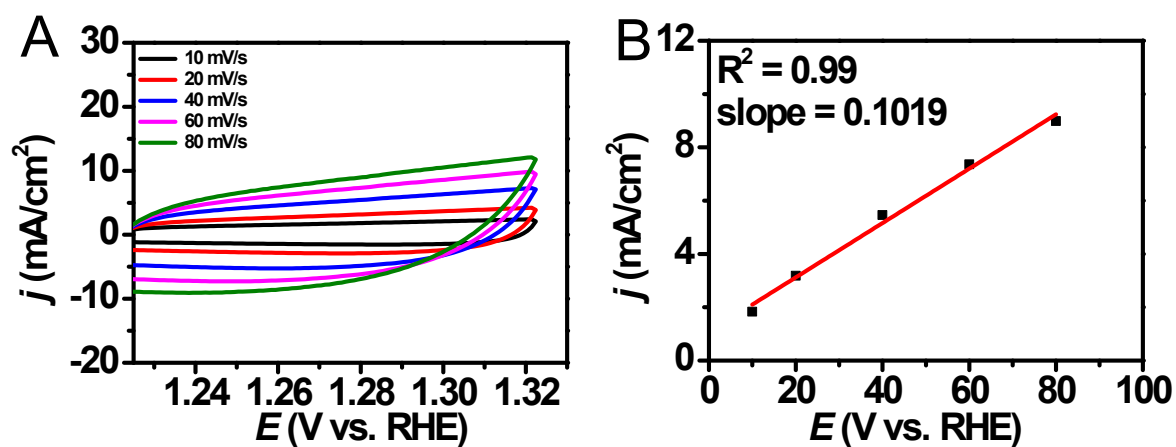


Figure S8. Double layer charging capacitance measurements for determination of the electrochemically active surface area of Fe:Cu(OH)₂ NTs in 1 M KOH.

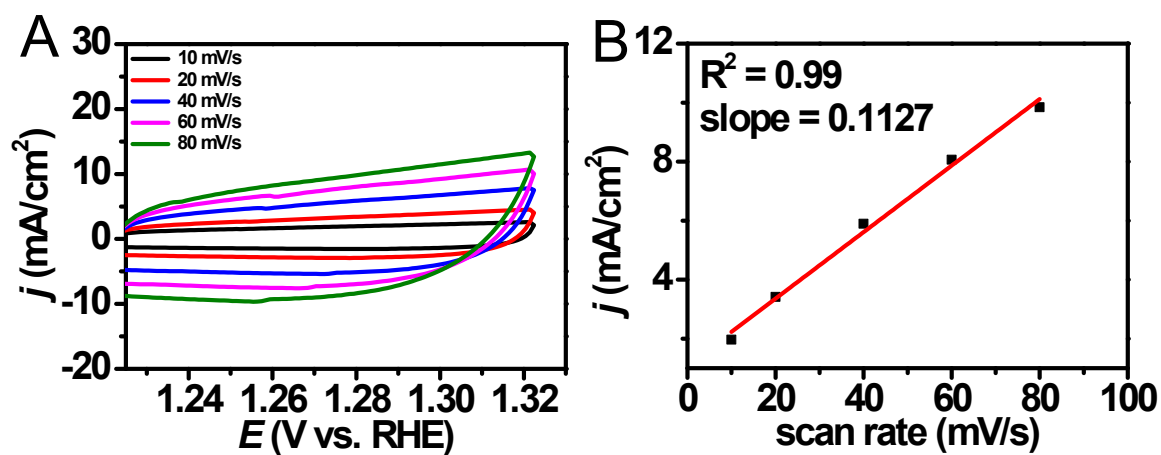


Figure S9. Double layer charging capacitance measurements for determination of the electrochemically active surface area of a Ni:Fe:Cu(OH)₂ NTs in 1 M KOH.

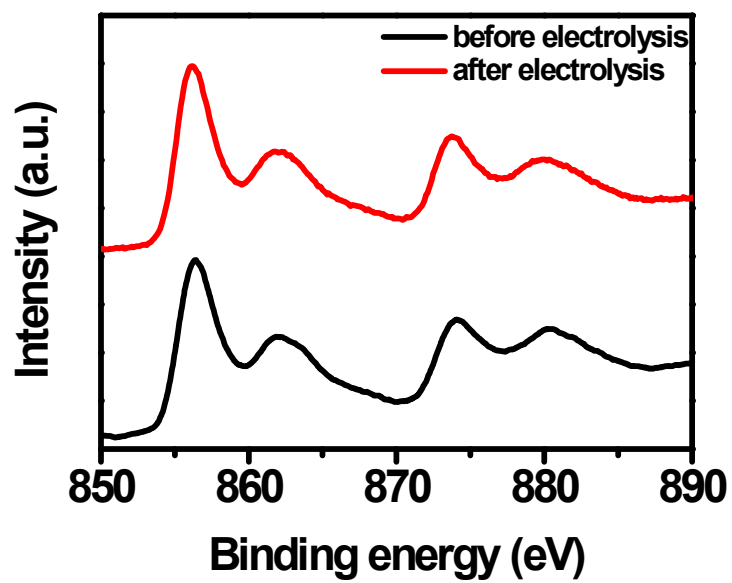


Figure S10. XPS of the Ni 2p peaks of Ni:Fe:Cu(OH)₂ NTs before (black) and after (red) 40 h electrolysis.

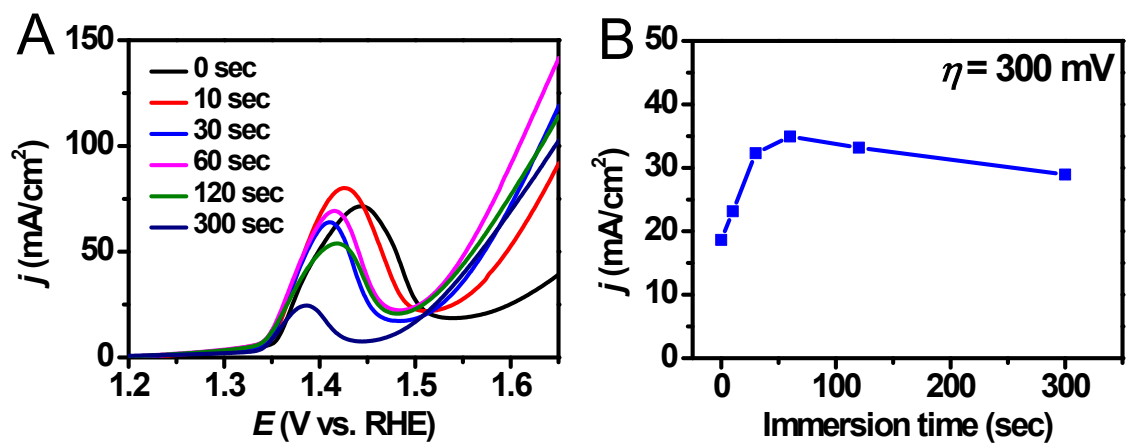


Figure S11. Optimization of Fe-doping of Ni:Cu(OH)₂. The Ni:Cu(OH)₂ NWs electrode was immersed into a solution of 10 mM Fe³⁺ for different times. LSV curves (A) and summarized current densities vs. immersion time at an overpotential of 300 mV (B). All electrochemical measurements were carried out in 1 M KOH at a scan rate of 20 mV/s with iR compensation.

Table S1. Elemental compositions as taken from the XPS peak fit.

Electrode	Oxygen	Copper	Iron	Nickel
Cu(OH)₂ NWs	68.6	31.4	-	-
Fe:Cu(OH)₂ NTs	63.7	1.9	34.4	-
Ni:Cu(OH)₂ NWs	70.0	3.4	-	26.6
Ni:Fe:Cu(OH)₂ NTs	70.8	2.1	4.5	22.6
Ni:Fe:Cu(OH)₂ NTs after electrolysis	73.2	1.4	4.8	20.6
Fe:Ni:Cu(OH)₂ NWs	72.7	1.8	4.2	21.3

Table S2. Results of ICP-OES measurements of Ni:Fe:Cu(OH)₂ NTs for Cu, Fe and Ni.

Element	Atomic ratio (%)
Copper	73.50
Iron	5.46
Nickel	21.04

Table S3. Comparison of Fe:Cu(OH)₂ NTs with Cu(OH)₂ NWs and other iron-based OER electrocatalysts in alkaline solutions.

Electrocatalyst	Substrate	Electrolyte	Current density (mA/cm ²)	η (mV)	Tafel slope	Refs
Fe:Cu(OH)₂ NTs	Cu foil	1 M KOH	10	380	40	This work
Cu(OH)₂ NWs	Cu foil	0.1 M NaOH	10	430	86	[1]
Fe(OH)₃:Cu(OH)₂	Cu foam	1 M KOH	10	365	42	[2]
FeO_x	FTO	0.1 M KOH	10	400	-	[3]
Steel S235*	Self-standing	0.1 M KOH	2	347	59	[4]
FeO_xH_y	Au/Ti	1 M KOH	1	350	43	[5]
Fe₂O₃	Carbon cloth	1 M KOH	10	420	-	[6]

*consisting of Fe₂O₃, FeO(OH), MnO(OH), and Mn₂O

Table S4. Comparison of Ni:Fe:Cu(OH)₂ NT with other NiFeO_x-based OER electrocatalysts in alkaline solutions.

Electrocatalyst	Substrate	Electrolyte	Current density (mA/cm ²)	η (mV)	Tafel slope	Refs
Ni:Fe:Cu(OH)₂ NTs	Cu foil	1 M KOH	100	320	32	This work
Fe-doped NiO	Au/QCM*	0.5 M KOH	10	297	37	[7]
Fe-doped NiOOH	Au	0.1 M KOH	10	~360	-	[8]
NiFe-LDH	Pyrolytic	1 M KOH	10	280	47	[9]
NiFe-LDH	Glassy carbon	1 M KOH	10	347	40	[10]
NiFe-hydroxide	Planar Au	1 M KOH	120	300	33	[11]
NiFe_{0.52}O_x	Glassy carbon	1 M KOH	10	330	97	[12]
NiFe-LDH	Glassy carbon	1 M KOH	10	320	31	[13]
NiFe_{0.4}O_x	FTO	0.1 M KOH	5	370	26	[14]

*Quartz Crystal Microbalance

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