Supporting Information

Dual Tuning of Nickel Sulfide Nanoflake Array Electrocatalyst through Nitrogen Doping and Carbon Coating for Efficient and Stable Water Splitting

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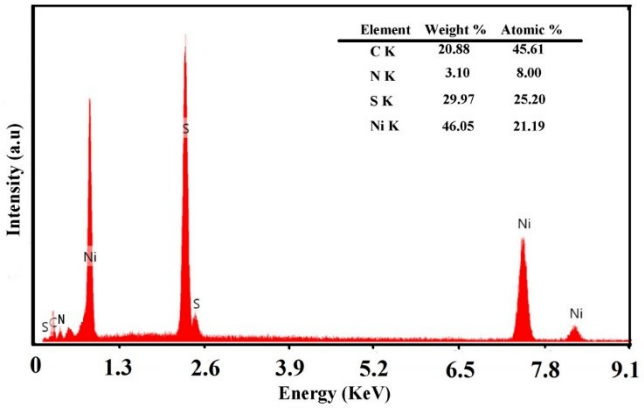

Figure S1. The EDS of N-Ni_3S_2@C/NF

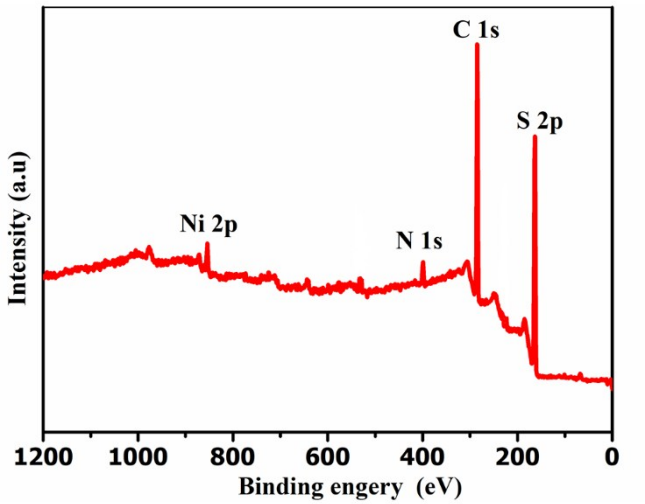

Figure S2. Survey XPS spectra of N-Ni_3S_2@C/NF

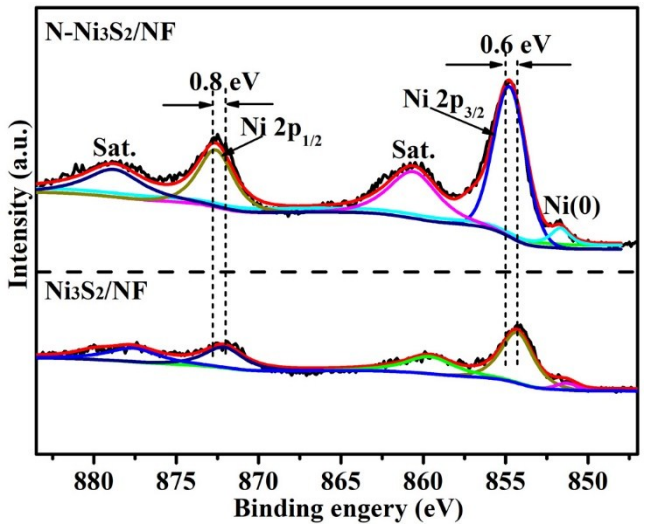

Figure S3. The XPS spectra of Ni 2p for Ni_3S_2/NF and N-Ni_3S_2/NF

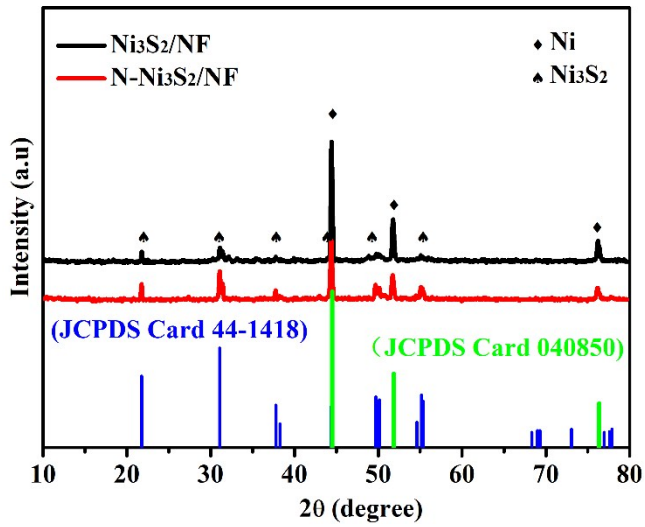

Figure S4. The XRD pattern of Ni_3S_2/NF and N-Ni_3S_2/NF

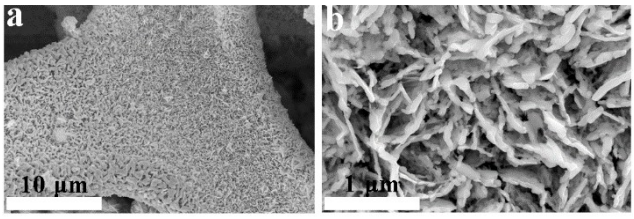

Figure S5. The SEM imagine of Ni_3S_2/NF with different magnifications

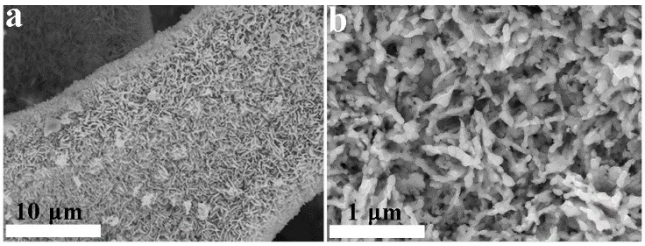

Figure S6. The SEM imagine of N-Ni_3S_2/NF with different magnifications

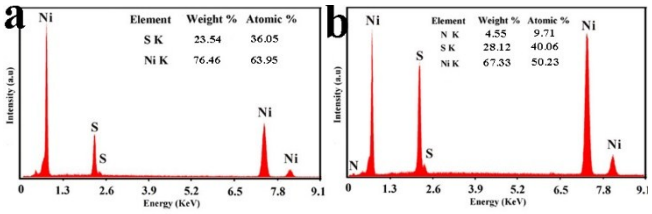

Figure S7. a)The EDS of Ni_3S_2/NF and b) N-Ni_3S_2/NF

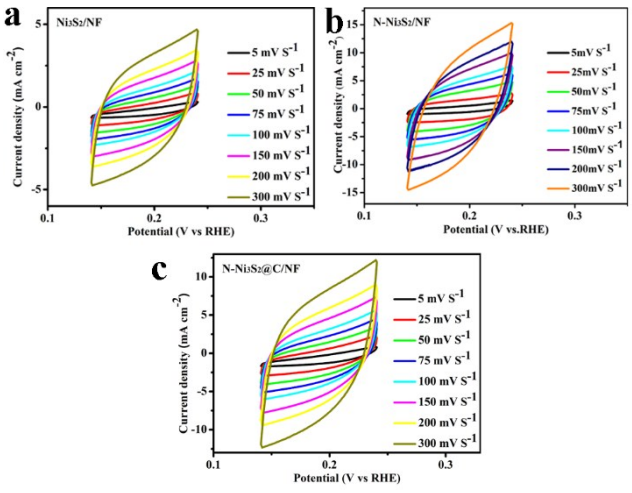

Figure S8. a)Typical cyclic voltammetry curves of N-Ni_3S_2@C/NF, b) N-Ni_3S_2 /NF and c)Ni_3S_2/NF in 1M KOH with different scan rates

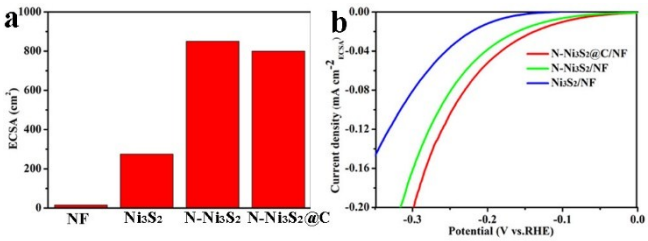

Figure S9. a) The ESCA of N-Ni_3S_2@C/NF, N-Ni_3S_2 /NF , Ni_3S_2/NF and NF.b)Polarization curves from normalized to the electrochemical active surface area (ECSA) for HER.

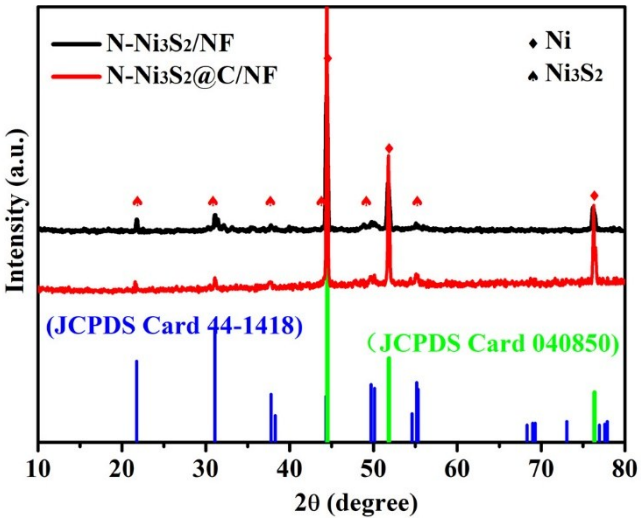

Figure S10. The XRD of N-Ni_3S_2/NF and N-Ni_3S_2@C/NF after HER

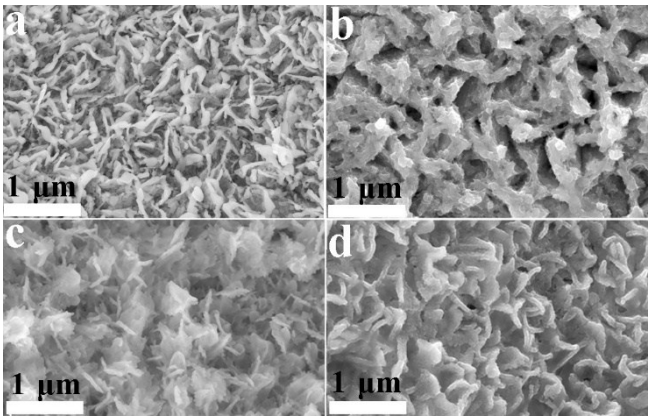

Figure S11. SEM images of a-b)N-Ni_3S_2/NF and c-d) N-Ni_3S_2@C/NF before and after HER in 1 M KOH

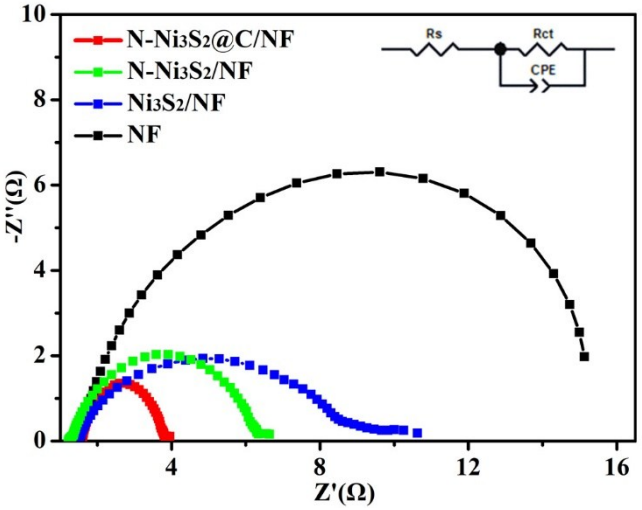

Figure S12. Electrochemical impedance spectroscopy (EIS) Nyquist plots at 1.53 V vs.RHE for all the synthesized materials.

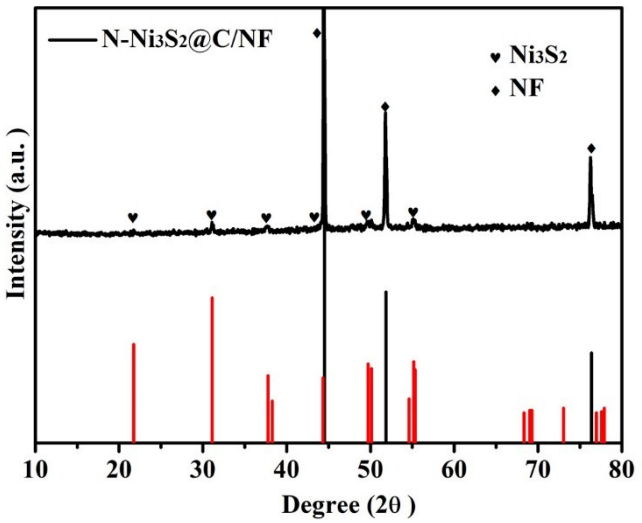

Figure S13. The XRD of N-Ni_3S_2@C/NF after OER

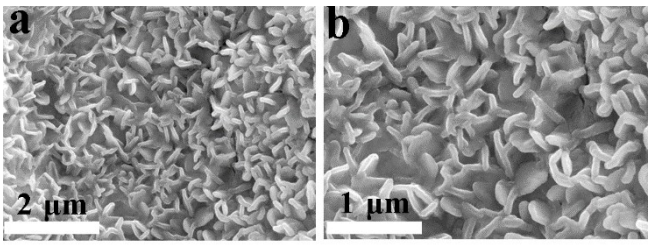

Figure S14. a-b)The SEM imagine of N-Ni_3S_2@C/NF after OER in 1 M KOH with different magnifications

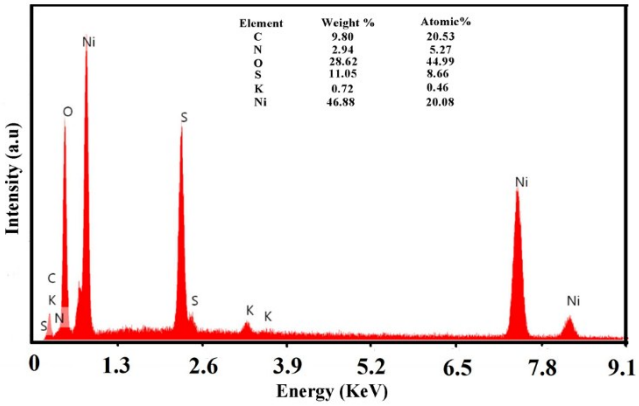

Figure S15. The EDS of N-Ni_3S_2@C/NF after OER

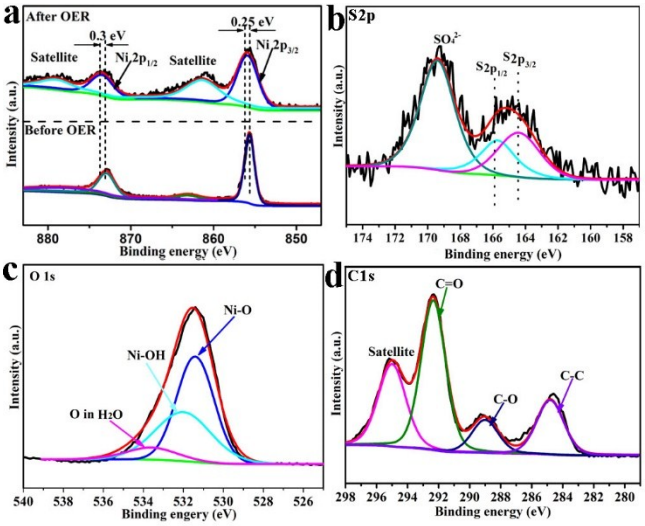

Figure S16. The XPS spectra of a) Ni 2p, b) S 2p, c) O 1s and d) C 1s for N-Ni_3S_2@C/NF after OER

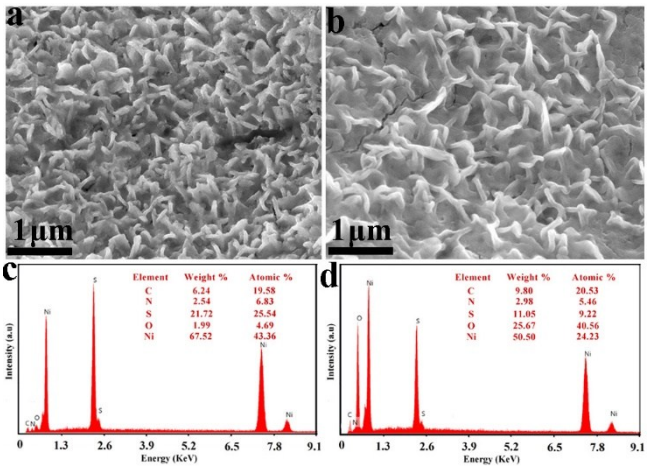

Figure S17. SEM images of N-Ni_3S_2@C/NF after overall water splitting. a) cathode b) anode and EDS results of N-Ni_3S_2@C/NF after overall water splitting. c) cathode d) anode

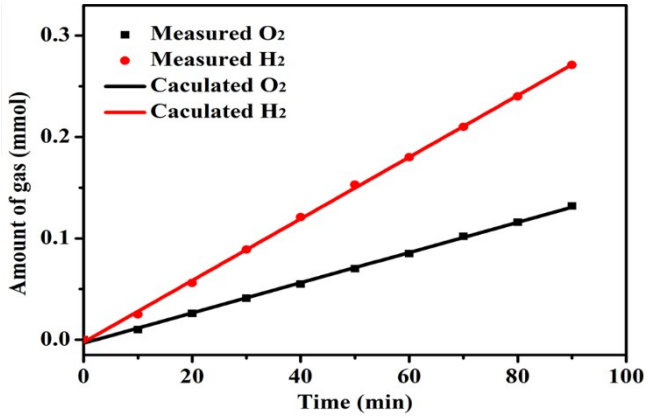

Figure S18. The amount of gas theoretically calculated and experimentally measured versus time for overall water splitting.

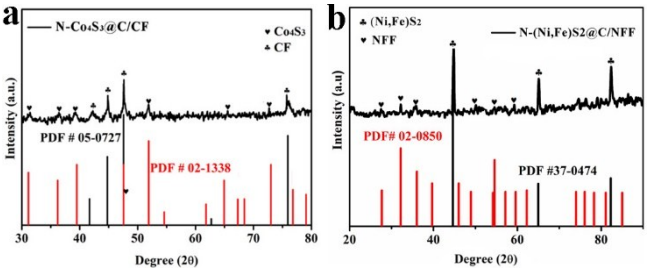

Figure S19. The XRD of a) N-Co_4S_3@C/CF and b) N-(Ni,Fe)S_2@C/NFF

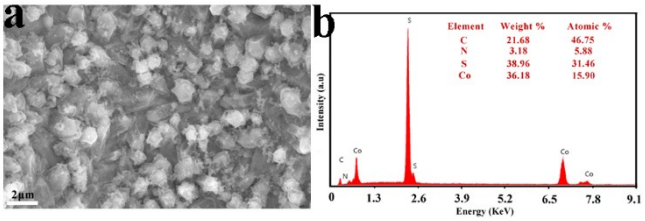

Figure S20. a) The SEM image and b) EDS of N-Co_4S_3@C/CF

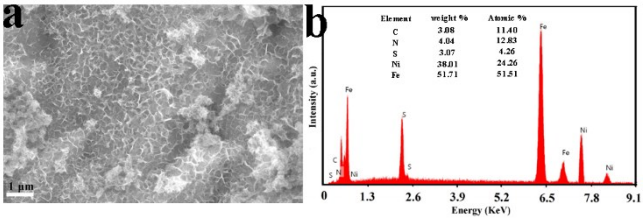

Figure S21. a) The SEM image and b) EDS of N-(Ni,Fe)S_2@C/NFF

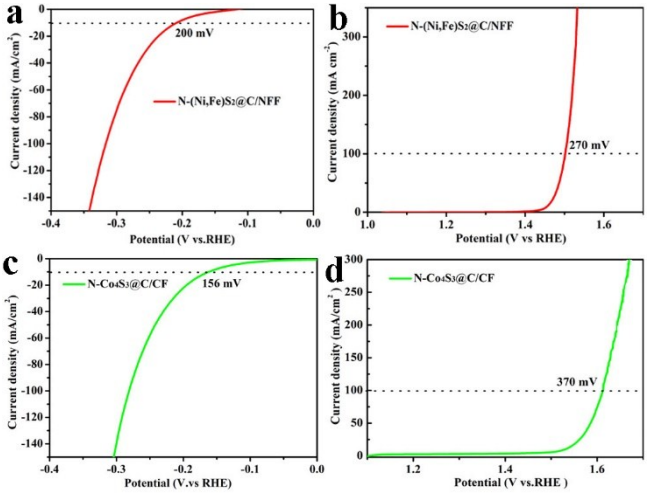

Figure S22. a) *iR*-corrected linear sweep voltammetry curves of N-(Ni,Fe)S_2@C/NFF for HER and b)OER in 1 M KOH.c) *iR*-corrected linear sweep voltammetry curves of N-Co_4S_3@C/CF for HER and d)OER in1 M KOH.

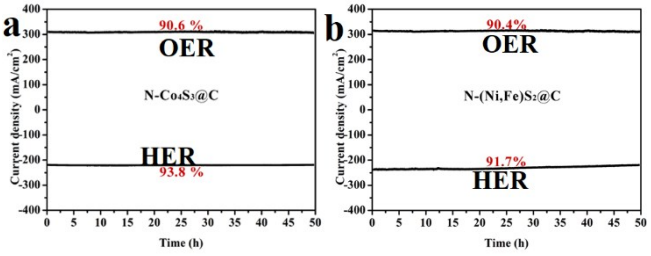

Figure S23. a) The i-t curves of N-Co_4S_3@C, b) N-(Ni,Fe)S_2@C for HER and OER in 1 M KOH .

Table S1 Comparison of HER performance of N-Ni_3S_2@C/NF product with various well-developed electrocatalysts in the literature.

Catalyst	Electrolyte	J ($mA\ cm^{-2}$)	η (mV)	Tafel ($mV\ dec^{-1}$)	Ref.
N-Ni_3S_2@C/NF	1 M KOH	10	113	90	This work
N-Ni_3S_2/NF	1 M KOH	10	110	-	[1]
Ni_3S_2 nanorod	1 M KOH	10	200	107	[2]
Ni_3S_2 nanosheet	1 M KOH	10	223	-	[3]
NiS microsphere	1 M KOH	20	158	83	[4]
Fe_2Ni_2N	1 M KOH	10	110	-	[5]
$NiCo_2S_4$ nanowire	1 M KOH	10	210	59	[6]
Ni_3S_2 needle array	1 M KOH	10	117	130	[7]
NiFe/$NiCo_2O_4$/NF	1 M KOH	10	105	88	[8]
NiFe@NC	1 M KOH	10	200	-	[9]
CoP	1 M KOH	10	209	51	[10]
Co_3Se_4/CF	1 M KOH	10	179	-	[11]

Table S2 Comparison of OER performance of N-Ni_3S_2@C/NF product with various well-developed electrocatalysts in the literature

Catalyst	Electrolyte	J ($mA\ cm^{-2}$)	η (mV)	Tafel ($mV\ dec^{-1}$)	Ref.
N-Ni_3S_2@C/NF	1M KOH	100	310	75	This work
N-Ni_3S_2/NF	1M KOH	170	350	70	[1]
Ni_3S_2 nanorod	1M KOH	100	370	140	[12]
Ni_3S_2 nanosheet	1M KOH	100	350	-	[3]
NiS microsphere	1M KOH	50	335	120	[4]
NiFe-LDH	1M KOH	100	300	47	[13]
NiCoFe LDH	1M KOH	30	233	53	[14]
NiCo LDH	1M KOH	100	340	57	[15]
FeCoW	1M KOH	10	191	-	[16]

Table S3 Comparison of water splitting cell voltage of N-Ni_3S_2@C/NF product with various well-developed electrocatalysts in the literature

Catalyst	Electrolyte	J ($mA\ cm^{-2}$)	Overall voltage (V)	Durability (h)	Ref.
N-Ni_3S_2@C/NF	1 M KOH	10	1.57	140	This work
N-Ni_3S_2/NF	1 M KOH	10	1.48	8	[1]
NiS microsphere	1 M KOH	10	1.64	35	[4]
NiSe nanowire	1 M KOH	10	1.63	20	[17]
Fe_2Ni_2N nanoarrays	1 M KOH	10	1.65	10	[5]
Ni@Co-Ni-P	1 M KOH	100	1.61	528	[18]
$NiCo_2S_4$ nanowire	1 M KOH	10	1.63	50	[6]
MoS_2/Ni_3S_2 Heterostructures	1 M KOH	10	1.56	10	[19]
CoS-$Co(OH)_2$	1 M KOH	10	1.58	28	[20]
Ni/Ni_8P_3	1 M KOH	10	1.61	10	[21]
NiCoP	1 M KOH	10	1.58	12	[22]

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