

## **Supporting Information**

### **In Situ Growth of S-Incorporated CoNiFe(oxy)hydroxides Nanoarrays as Efficient Multifunctional Electrocatalysts**

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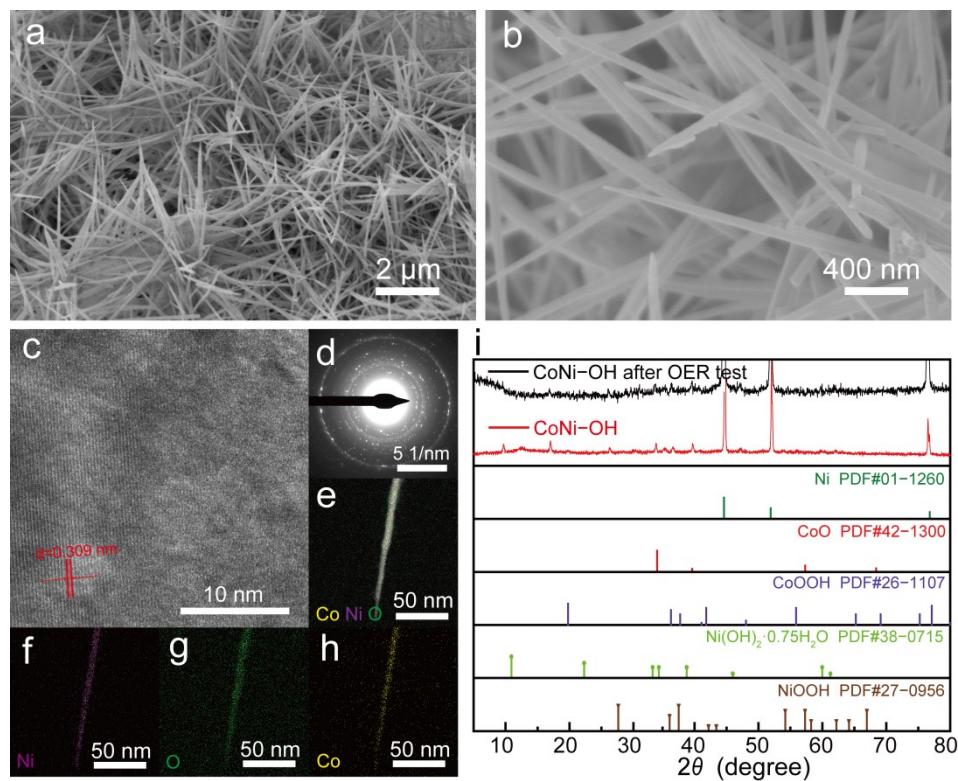


Fig. S1. (a,b) SEM images at different magnifications, (c) HRTEM image,(d) SAED patterns, (e–h) elemental mappings, (i) XRD patterns of the of CoNi–OH nanostructures.

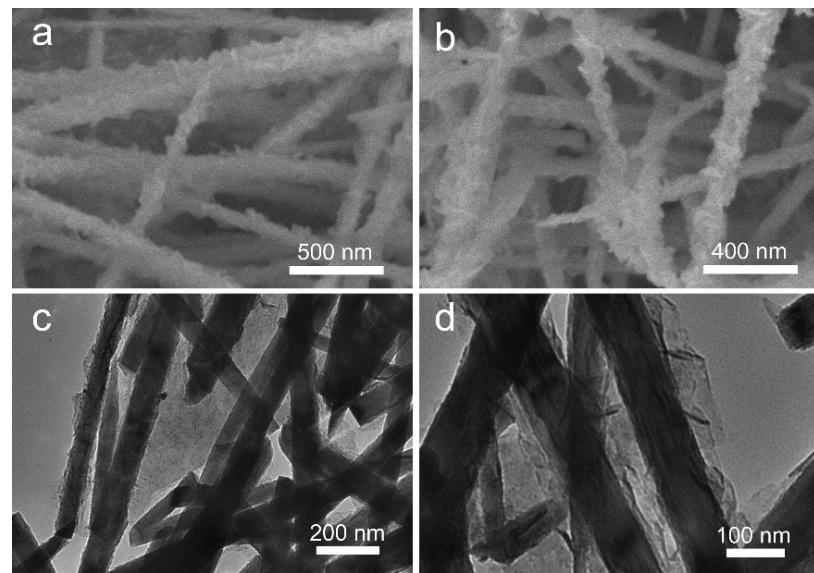


Fig. S2 (a–b) SEM and (c–d) TEM images of CoNiFeS–OH at different magnifications.

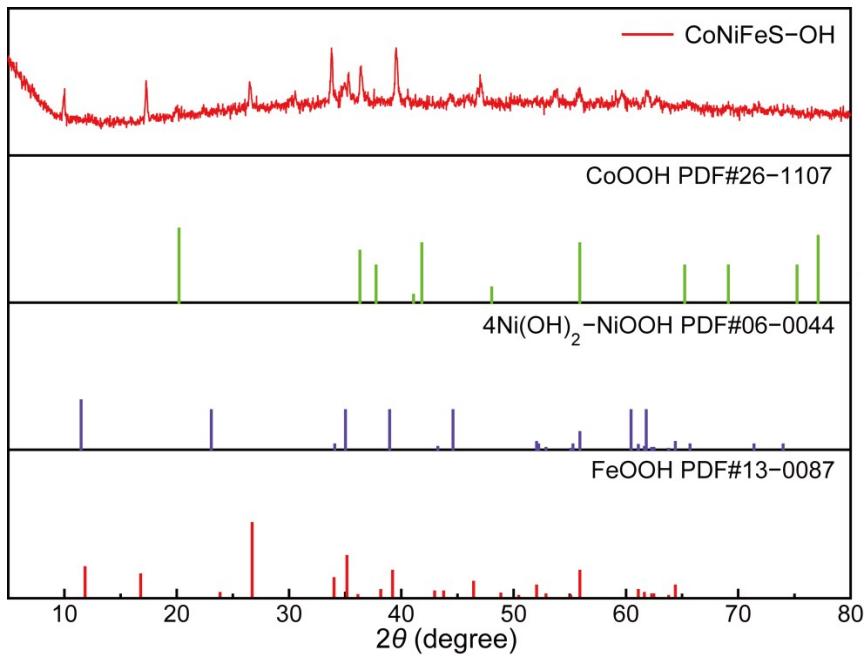


Fig. S3 XRD patterns of CoNiFeS–OH.

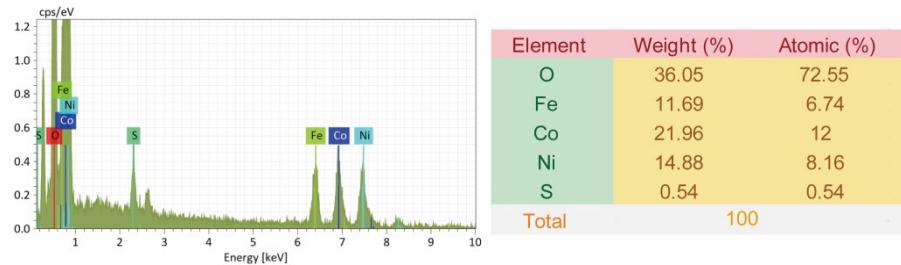


Fig. S4 EDS spectra and the corresponding elemental compositions of CoNiFeS–OH.

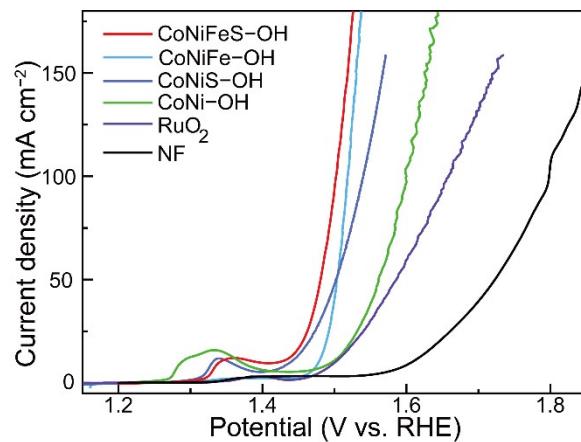


Fig. S5 LSV curves for the OER of CoNiFeS–OH, CoNi–OH, CoNiFe–OH, CoNiS–OH, RuO<sub>2</sub>, and NF in 1.0 M KOH at a scan rate of 0.5 mV s<sup>-1</sup>.

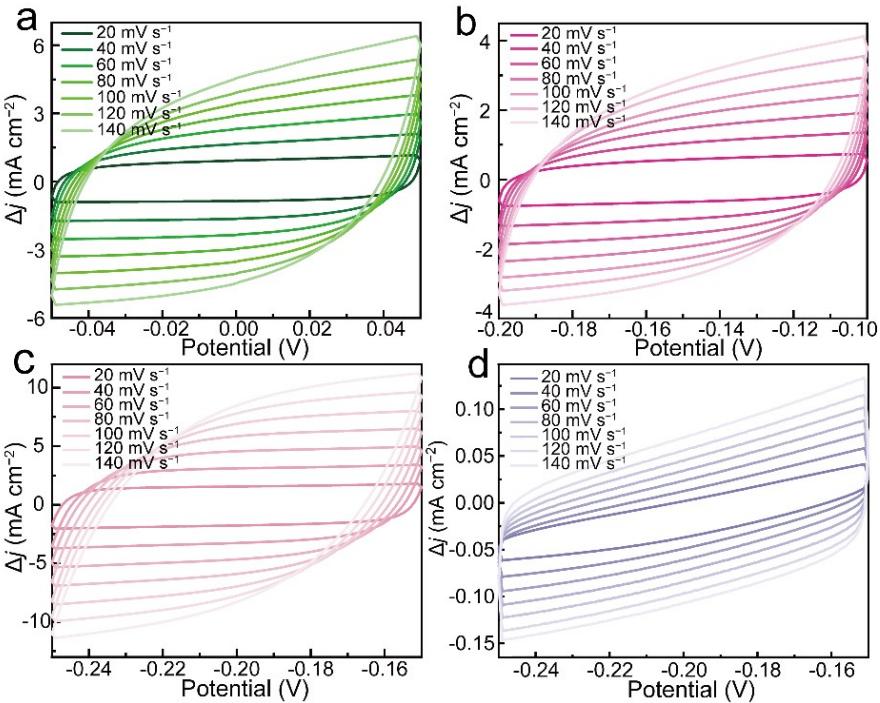


Fig. S6 CV curves for OER measured in KOH (1.0 M) at various scan rates of 20, 40, 60, 80, 100, 120 and 140  $\text{mV s}^{-1}$ : (a) CoNiFeS–OH, (b) CoNi–OH, (c) RuO<sub>2</sub>, and (d) NF, respectively.

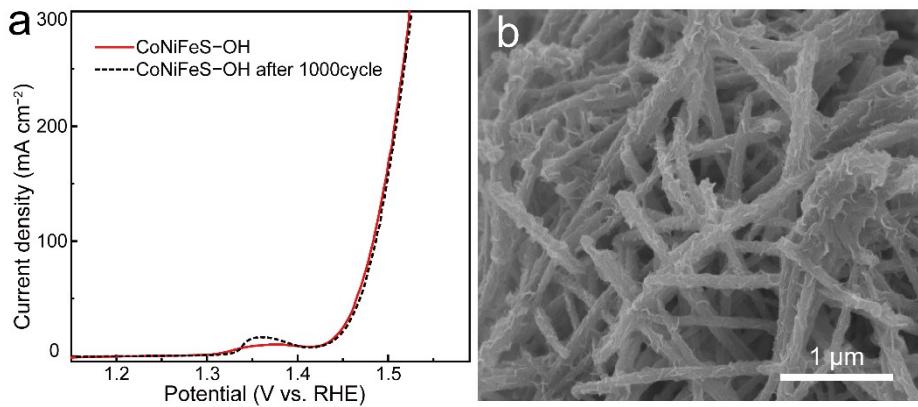


Fig. S7 (a) The polarization curve LSV for OER before and after 1000 CV test of CoNiFeS–OH. (b) SEM of CoNiFeS–OH after 1000 CV in 1M KOH (1.0 M).

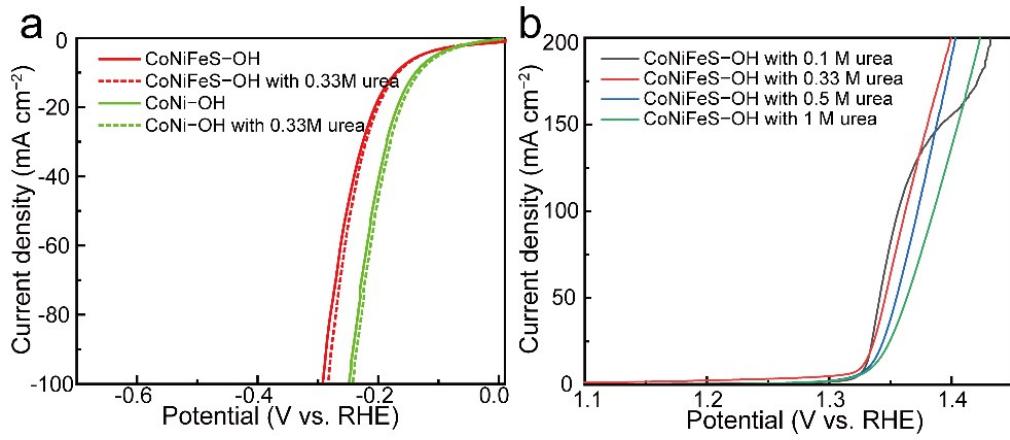


Fig. S8 (a) LSV curves for the HER of CoNiFeS–OH and CoNi–OH in KOH (1.0 M) and in KOH (1.0 M) with urea (0.33 M) at a scan rate of  $5 \text{ mV s}^{-1}$ . (b) LSV curves for the UOR of CoNiFeS–OH in different urea concentrations at a scan rate of  $1 \text{ mV s}^{-1}$ .

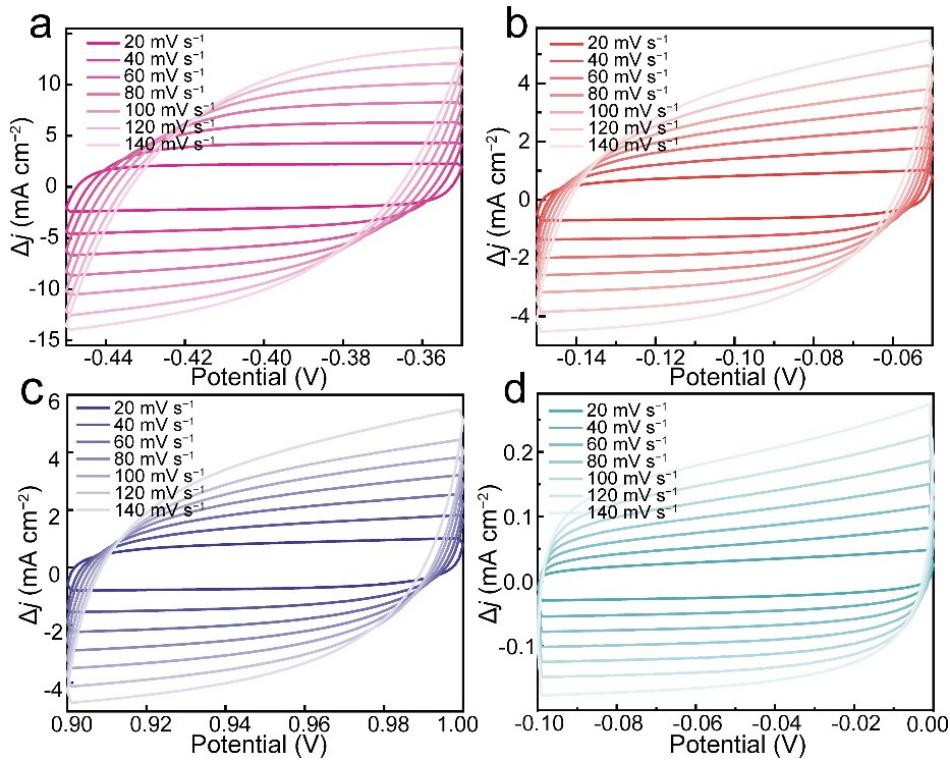


Fig. S9 CV curves for UOR in KOH (1.0 M) with urea (0.33 M) at scan rate of 20, 40, 60, 80, 100, 120 and 140  $\text{mV s}^{-1}$ : (a) CoNiFeS–OH, (b) CoNi–OH, (c) RuO<sub>2</sub>, (d) NF.

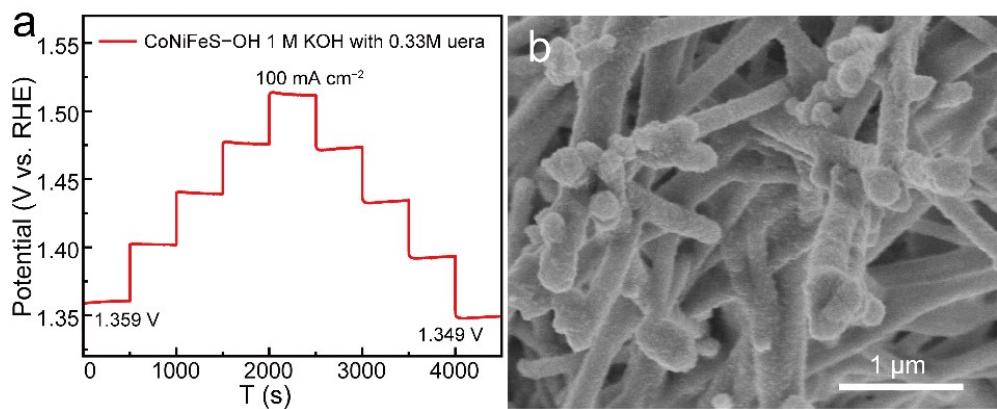


Fig. S10 (a) Multi-step chronopotentiometric curve of CoNiFeS–OH from 20 to 100 mA cm<sup>-2</sup> in KOH (1.0 M) with urea (0.33 M). (b) SEM of CoNiFeS–OH after 1000 CV in KOH (1.0 M) with urea (0.33 M).

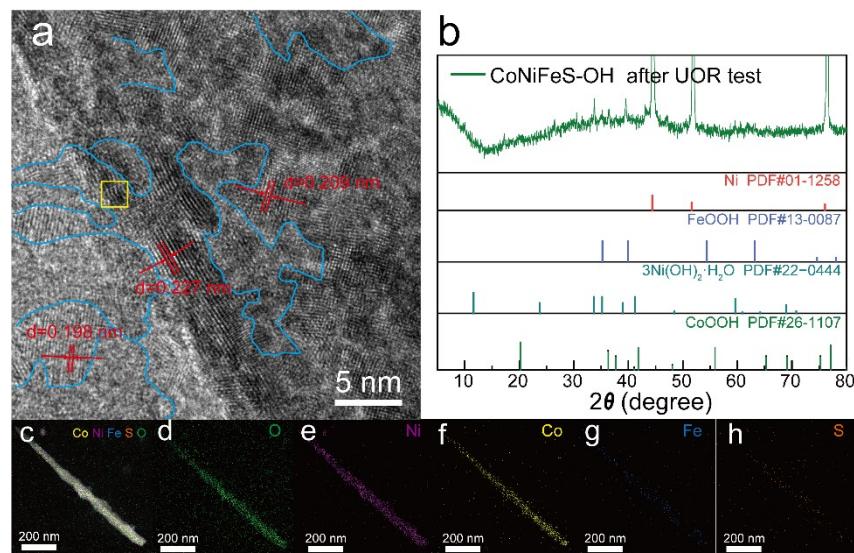


Fig. S11 Characterizations of CoNiFeS–OH catalysts after UOR stability test. (a,b) HRTEM image and XRD patterns, (c–h) elemental mapping of immersed, O, Ni, Co, Fe, and S, respectively.

Table S1 Overpotential of CoNiFeS–OH, CoNi–OH, RuO<sub>2</sub>, and NF toward OER at  $\eta_{10}$ ,  $\eta_{20}$ ,  $\eta_{50}$ , and  $\eta_{100}$  in KOH (1.0 M).

	10 mA cm <sup>-2</sup> (mV)	20 mA cm <sup>-2</sup> (mV)	50 mA cm <sup>-2</sup> (mV)	100 mA cm <sup>-2</sup> (mV)
CoNiFeS–OH	192	226	251	272
CoNi–OH	260	290	330	370
RuO <sub>2</sub>	271	298	355	428
NF	377	415	493	568

Table S2 Comparison for OER properties of CoNiFeS–OH with other non-noble metal based electrocatalysts in KOH (1.0 M).

Catalysts	Substrate	Electrolyte Conc. (KOH)	Current density (mA cm <sup>-2</sup> )	Overpotenti al (mV)	Ref.
Fe–CoNi–OH	Nickel foam	1M	10	210	1
NiFeCr LDH	Nickel foam	1M	10	225	2
Fe–MOFs@Ni-MOFs@NiFe–LDH	Nickel foam	1M	10	275	3
Fe–Co– $\alpha$ MOF	Nickel foam	1M	10	249	4
CoFeV LDH	Nickel foam	1M	10	242	5
Fe–Ni(OH) <sub>2</sub>	Nickel foam	1M	10	235	6
NiCo <sub>2</sub> S <sub>4</sub> /FeOOH	Nickel foam	1M	10	~200	7
NiCo–LDH@FeOOH	Carbon papers	1M	10	224	8
NiFePS	Nickel foam	1M	10	242	9
FeCoNiPB/ (FeCoNi) <sub>3</sub> O <sub>4-x</sub>	copper foil	1M	10	229	10
CoNiFeS–OH	Nickel foam	1M	10	192	This work

Table S3 Potential of CoNiFeS–OH, CoNi–OH, RuO<sub>2</sub>, and NF toward UOR at  $\eta_{10}$ ,  $\eta_{20}$ ,  $\eta_{50}$ , and  $\eta_{100}$  in KOH (1.0 M) with urea (0.33 M).

	10 mA cm <sup>-2</sup> (V)	20 mA cm <sup>-2</sup> (V)	50 mA cm <sup>-2</sup> (V)	100 mA cm <sup>-2</sup> (V)
CoNiFeS–OH	1.329	1.338	1.354	1.373
CoNi–OH	1.316	1.333	1.366	1.425
RuO <sub>2</sub>	1.388	1.423	1.549	1.645
NF	1.360	1.390	/	/

Table S4 Comparison for UOR properties of CoNiFeS–OH with other non-noble metal based electrocatalysts in KOH (1.0 M) with urea.

Catalysts	substrate	Urea concentration (M)	Current density (mA cm <sup>-2</sup> )	Voltage (V vs RHE)	Ref.
MoS <sub>2</sub> /Ni <sub>3</sub> S <sub>2</sub>	Nickel foam	0.33	20	1.45	11
NiMo alloy	Nickel foam	0.1	10	1.48	12
NiCo <sub>2</sub> S <sub>4</sub>	Carbon cloth	0.33	10	1.45	13
NiMoS	Carbon cloth	0.5	10	1.59	14
NiCo <sub>2</sub> S <sub>4</sub>	Carbon cloth	0.33	10	1.45	15
NiCo LDH	Nickel foam	0.33	10	1.353	16
FeNi <sub>3</sub> –MoO <sub>2</sub>	Nickel foam	0.5	10	1.29	17
Ni@C–250	CP	0.5	10	1.35	18
V–Ni <sub>3</sub> N/NF	Nickel foam	0.5	10	1.361	19
NiS/MoS <sub>2</sub>	Carbon cloth	0.4	100	1.43	20
CoNiFeS–OH	Nickel foam	0.33	10	1.329	This work

Table S5 Comparison for urea electrolysis efficiency of CoNiFeS-OH//CoNiFeS-OH with other non-noble metal based electrocatalysts in KOH (1.0 M) with urea.

Catalysts	Urea concentration (M)	$j$ (mA cm $^{-2}$ )	Cell voltage (V)	Ref.
NiFeCo LDH/NF	0.33	10	1.49	21
NiCo <sub>2</sub> S <sub>4</sub> NS/CC	0.5	10	1.49	13
Ni <sub>3</sub> S <sub>2</sub> -NiS/NF	0.5	50	1.54	22
NC-FNCP	0.5	10	1.52	23
Ni-Co <sub>9</sub> S <sub>8</sub> /CC	0.33	10	1.52	24
NiMoS/CC	0.5	10	1.59	14
NiCo <sub>2</sub> S <sub>4</sub> /CC	0.33	10	1.45	15
P-NiCoZn LDH/NF	0.5	10	1.479	25
NF/PPy700Ni <sub>3</sub> S <sub>2</sub> -8-Ar	0.33	10	1.47	26
(Ni <sub>0.25</sub> Fe <sub>0.75</sub> ) <sub>3</sub> S <sub>2</sub> /NF	0.33	10	1.49	27
CoNiFeS-OH/NF	0.33	10	1.46	This work

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