Electronic Supplementary Material (ESI) for Materials Advances.

One-pot synthesis and microstructure analysis of Fe doped NiS_2 for efficient oxygen evolution electrocatalysis

Haofei Zhao, † Wenrui Li † and Rongming Wang *

Beijing Advanced Innovation Center for Materials Genome Engineering, Beijing Key Laboratory for Magneto-Photoelectrical Composite and Interface Science, School of Mathematics and Physics, University of Science and Technology Beijing, Beijing 100083, China.

E-mail: rmwang@ustb.edu.cn

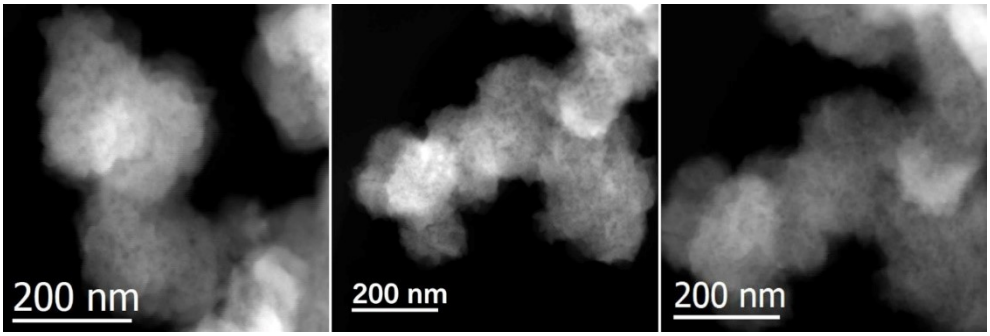

Fig. S1 Annular dark-field images of nanoparticles of as-prepared NiS_2.

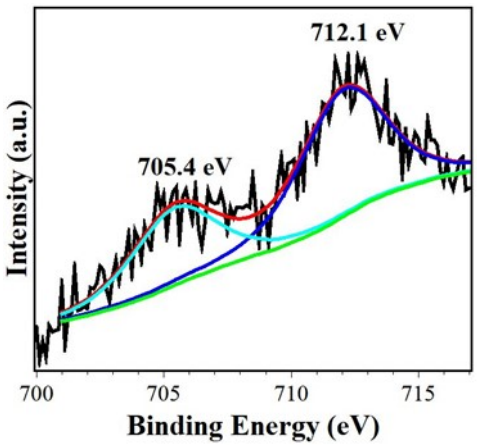

Fig. S2 XPS spectrum of NiS_2 ranging from 700 eV and 717 eV, exhibiting two Ni LMM Auger peaks. The fitted positions of the two peaks are 705.4 eV and 712.1 eV, respectively, with an area ratio of 0.6. The fitted FWHM and Lorentzian character percent are 4.0 eV and 45%, respectively.

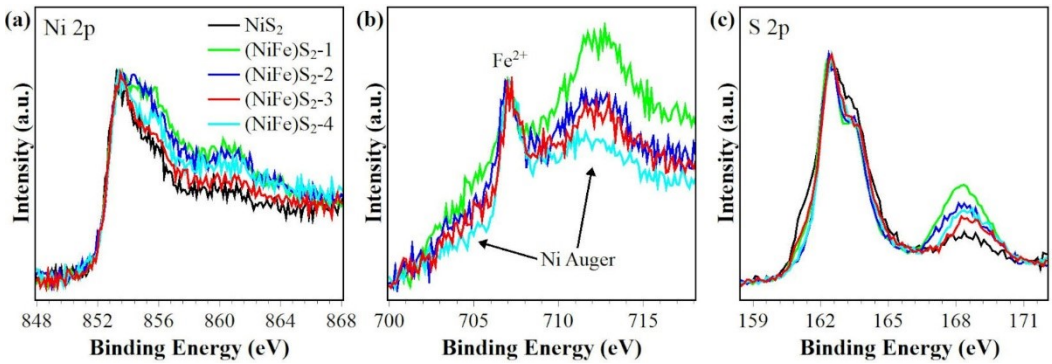

Fig. S3 (a) Ni 2p, (b) Fe 2p and (c) S 2p XPS spectra of the as-synthesized samples. The patterns are all normalized for comparation.

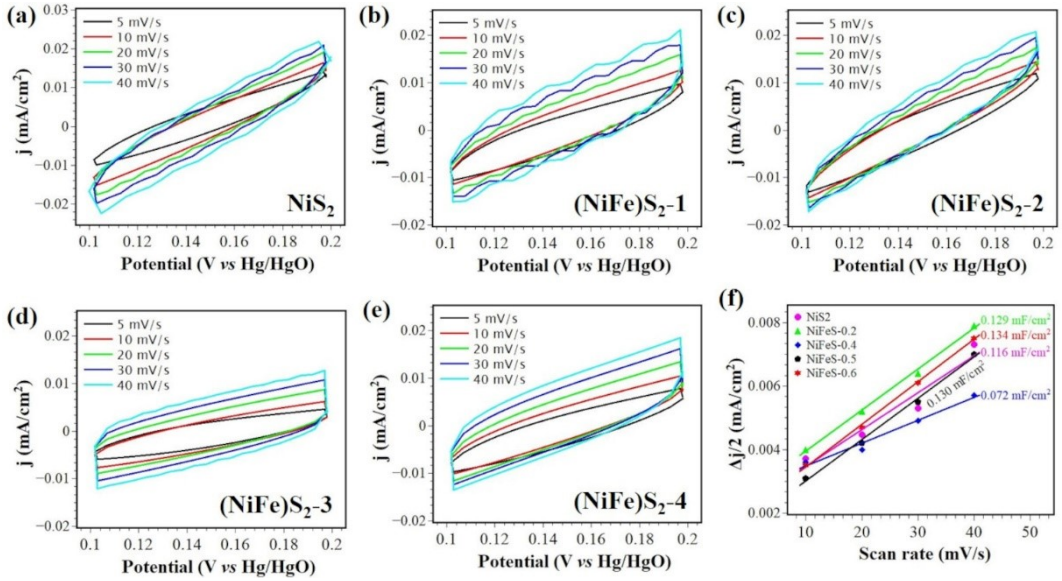

Fig. S4 (a-e) Cyclic voltammograms tested in a non-Faradic potential range of 0.1 – 0.2 V *vs* Hg/HgO and (f) Capacitive currents as a function of scan rate of NiS_2, $(NiFe)S_2$-1, $(NiFe)S_2$-2, $(NiFe)S_2$-3 and $(NiFe)S_2$-4.

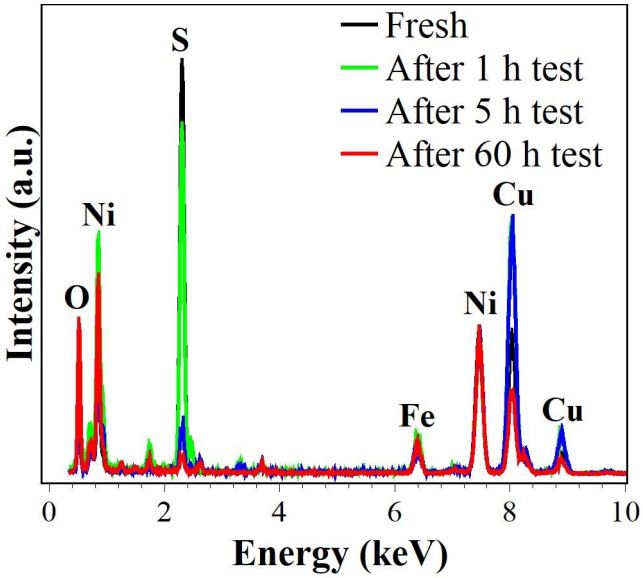

Fig. S5 Comparison of the EDX spectra of $(NiFe)S_2$-3 after being tested for different times at ~ 20 mA/cm^2.

Table S1 Summarization of fitting parameters of the XRD data using MAUD and the Fe contents measured by EDX quantification analysis.

Sample	**Lattice parameter (Å)**	**Crystallite size（nm）**	**Rwp (%)**	**Fe content (%)**
NiS_2	5.6803 (6)	34.0 (4)	4.83	0
$(NiFe)S_2$-1	5.6722 (6)	46.5 (6)	5.59	5.9
$(NiFe)S_2$-2	5.6632 (5)	37.1(3)	5.28	12.5
$(NiFe)S_2$-3	5.6483 (9)	24.7 (5)	6.13	16.3
$(NiFe)S_2$-4	5.6114 (13)	20.8 (1.1)	6.02	29.0

Table S2 Summarization of fitting parameters of the Ni 2p XPS spectra.

Samples	**Ni^{2+} 2p3/2**			**Ni^{3+} 2p3/2**			**Sat.**		
	B.E. (eV)	FWHM (eV)	Area	B.E. (eV)	FWHM (eV)	Area	B.E. (eV)	FWHM (eV)	Area
NiS_2	853.33	1.74	1.00	855.24	2.40	0.71	858.74	5.51	0.53
$(NiFe)S_2$-1	853.51	1.86	-	855.59	2.67	1.13	859.58	6.09	1.40
$(NiFe)S_2$-2	853.59	1.89	-	855.52	2.67	1.02	859.72	6.46	1.31
$(NiFe)S_2$-3	853.43	1.72	-	855.40	2.48	0.76	859.03	5.89	0.72
$(NiFe)S_2$-4	853.49	1.69	-	855.46	2.47	0.98	859.38	7.10	1.26

Table S3 Summarization of fitting parameters of the S 2p XPS spectra.

Samples	**S_2^{2-} 2p3/2**			**S^{2-}**			**S_n^{2-}**			**SO_4^{2-}**		
	B.E. (eV)	FWHM (eV)	Area	B.E. (eV)	FWHM (eV)	Area	B.E. (eV)	FWHM (eV)	Area	B.E. (eV)	FWHM (eV)	Area
NiS_2	162.35	1.03	1	161.31	1.05	0.389	164.43	1.43	0.372	168.35	2.64	0.420
$(NiFe)S_2$-1	162.42	1.03	-	161.35	1.18	0.236	164.03	0.95	0.224	168.27	2.53	0.903

$(NiFe)S_2$-2	162.42	1.11	-	161.18	1.20	0.098	164.25	1.11	0.148	168.27	2.60	0.640
$(NiFe)S_2$-3	162.45	1.15	-	161.36	1.15	0.266	164.19	1.94	0.479	168.62	2.36	0.507
$(NiFe)S_2$-4	162.45	1.10	-	161.28	1.20	0.137	164.52	1.84	0.305	168.54	2.52	0.599

Table S4 Overpotentials at 10 mA/cm^2, Tafel slopes and fitting parameters of the EIS data.

Samples	**η_{10} (mV)**	**Tafel slope (mV/dec)**	**C_{dl} (mF/cm^2)**	**R_s (Ω)**	**R_{ct1} (Ω)**	**CPE_1**	**R_{ct2} (Ω)**	**CPE_2**
NiS_2	411	89.8	0.116	1.389	1.705	0.628	3.292	0.615
$(NiFe)S_2$-1	303	59.5	0.129	1.386	0.753	0.618	1.823	0.766
$(NiFe)S_2$-2	337	77.3	0.072	1.511	0.784	0.87	2.609	0.651
$(NiFe)S_2$-3	257	41.2	0.130	1.675	0.883	0.546	1.404	0.778
$(NiFe)S_2$-4	295	59.6	0.134	1.559	0.537	0.447	0.97	0.93

Table S5 Comparison of the electrocatalytic OER activity of $(NiFe)S_2$-3 with various transition-metal based catalysts in 1 M KOH electrolyte.

Catalyst / Pre-catalyst	**η_{10} (mV)**	**Tafel slope (mV/dec)**	**Reference**
$(NiFe)S_2$-3	257	41	This work
$Fe_{0.1}Ni_{0.9}S_2$ nanosheets	260	46	1
$(NiFe)S_2$	230	42.6	2
$(NiFe)S_2$@Graphene	320	61	3
$Ni_{6/7}Fe_{1/7}$-OH-6/CNT derived from disulfides	190	24	4

Fe doped Ni_3S_2 in mesoporous carbon	350	93	5
$V_{0.1}Ni_{0.9}S_2$ nanosheets	290	45	6
$(Ni,Co)S_2$	270	58	7
$Fe_{0.2}Co_{0.8}S_2$	290	52.6	8
$Co_{0.25}Fe_{0.75}S_2$	324	50	9
Co-FeS_2/CoS_2	278	73	10
Ni doped Co_3S_4 nanowires	283	65	11
Ni_3S_2@NiV-LDH	190	57	12
NiFe–$NiFe_2O_4$ composite nanofibers	316	74	13
CoS_2/Ni foam	298	94	14
Ni_3S_2 nanosheets/Ni foam	223	60.5	15
NiCoP/C	330	96	16
Fe-O_x/Fe foam	238	82.7	17
h-NiS_x/Ni foam	180	96	18
FeOOH decorated CoP porous nanofiber	250	56.6	19
Mn-doped CoP nanosheets	288	77.2	20
Ni/FeOOH@$NiFe_2O_4$ derived from Ni_5P_4@FeP	205	42.3	21
monolayer $(NiCo)(OH)_2$ on GC electrode	208	-	22
Ultrathin NiFe LDH	210	30	23
Zn-doped $NiFeO_xH_y$	250	28.3	24

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