Supporting Information for

Al-Doped Nickel Sulfide Nanosheet Arrays as Highly-E□cient Bifunctional Electrocatalysts for Overall Water Splitting

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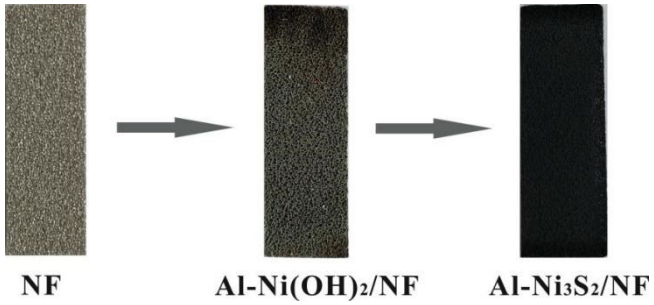

Fig. S1. Photographic images of Ni foam (NF), Al-Ni(OH)$_2$/NF and Al-Ni$_3$S$_2$/NF.

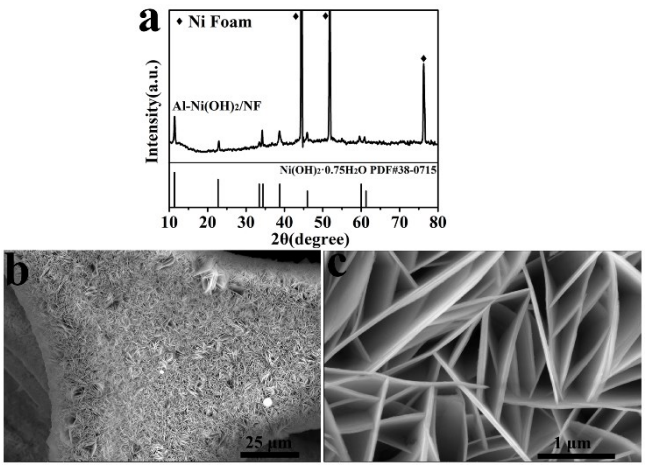

Fig. S2. (a) XRD, and (b-c) SEM images of Al-Ni(OH)$_2$/NF.

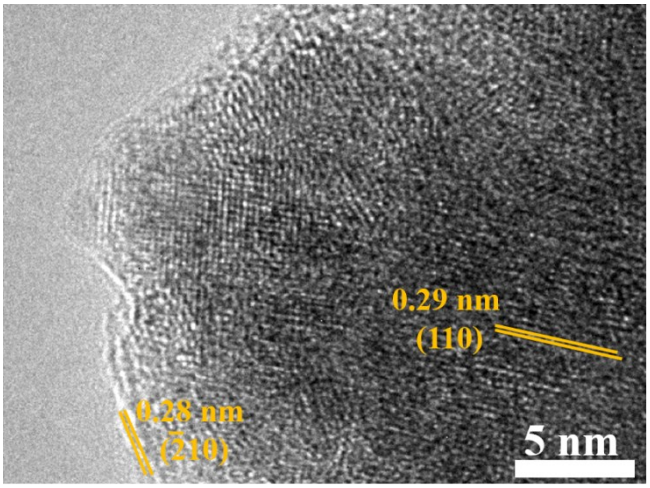

Fig. S3. HRTEM image of the edge of Al-Ni$_3$S$_2$/NF nanosheet.

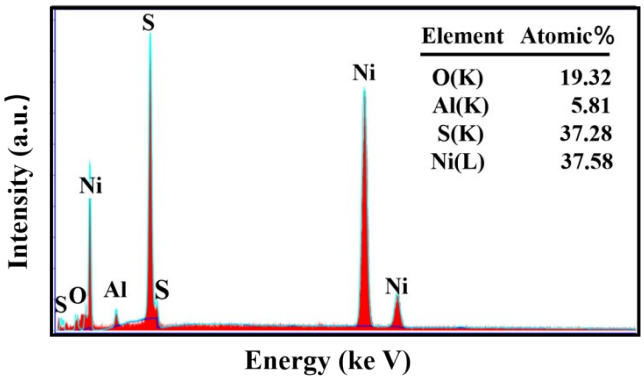

Fig. S4. EDS spectrums of Al-Ni_3S_2/NF

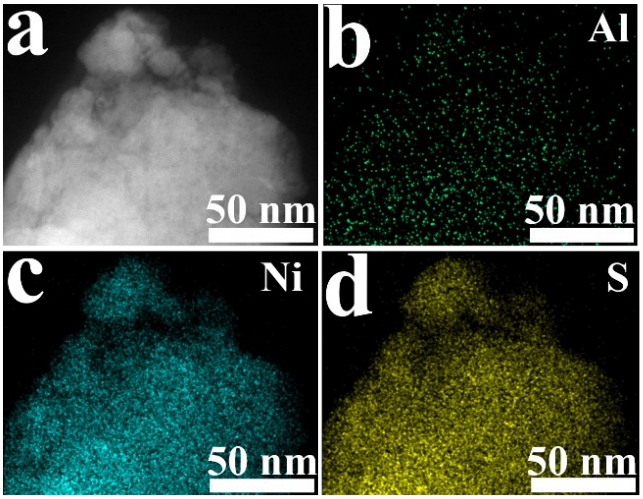

Fig. S5. (a) survey, (b) Al, (c) Ni, and (d) S elemental mapping images of Al-Ni_3S_2/NF.

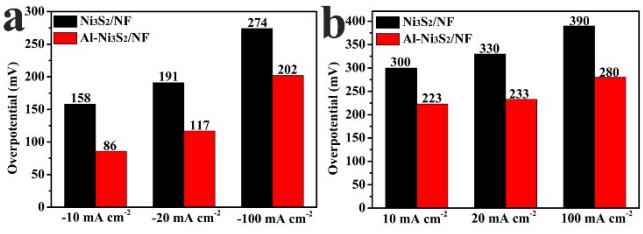

Fig. S6. The overpotential of Ni_3S_2/NF and Al-Ni_3S_2/NF at the current density of 10, 20 or 100 mA cm^{-2} for (a) HER, and (b) OER.

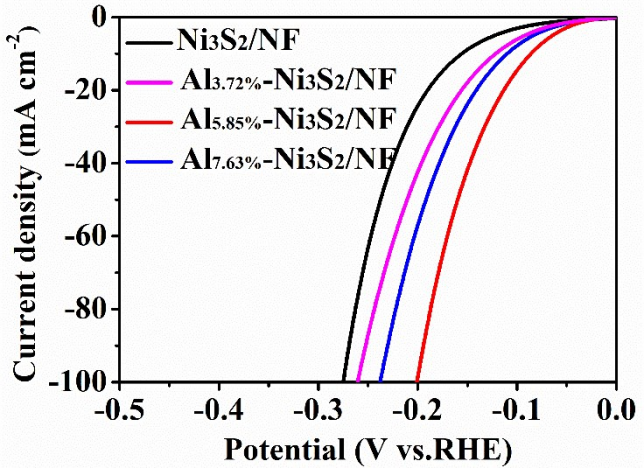

Fig. S7. The LSV curves of the HER performance of $Al_{X\%}$-Ni_3S_2/NF (x=3.72, 5.85 and 7.63).

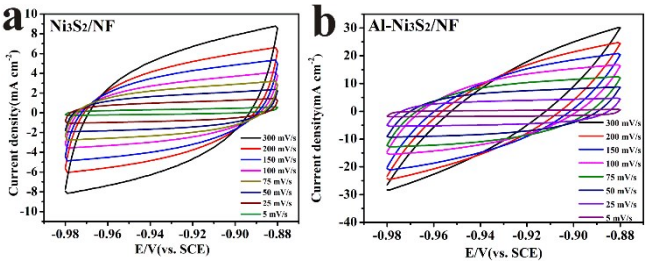

Fig. S8 Typical cyclic voltammetry curves of (a) Ni_3S_2/NF and (b) $Al_{5.85\%}$-Ni_3S_2/NF with different scan rates in 1M KOH.

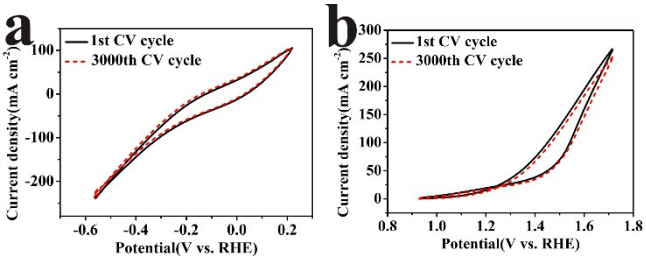

Fig. S9 The first and 3000th CV cycle curves of Ni_3S_2/NF and $Al_{5.85\%}$-Ni_3S_2/NF for (a) HER and (b) OER.

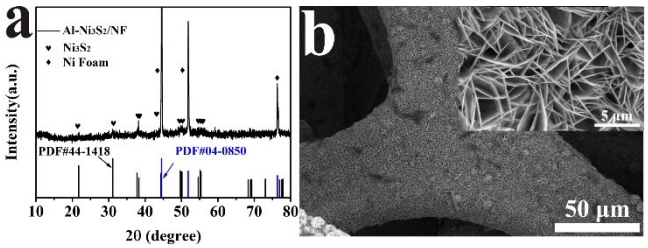

Fig. S10 (a) XRD pattern and (b) SEM of $Al_{5.85\%}$-Ni_3S_2/NF after HER.

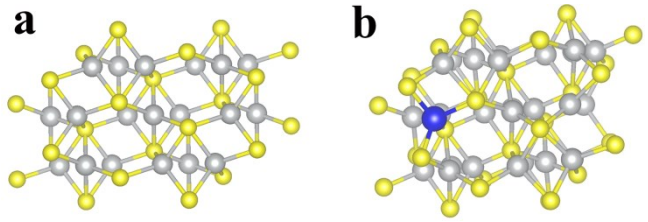

Fig. S11. Top views of the $(\bar{2}10)$ surface and of (a) Ni_3S_2/NF and (b) Al-Ni_3S_2.

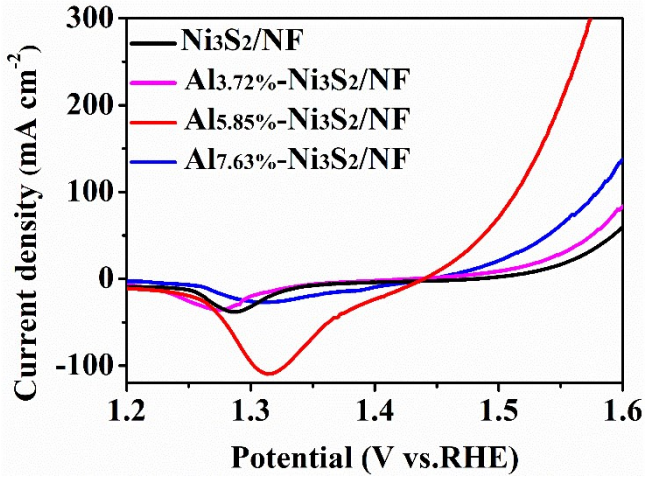

Fig. S12. The LSV curves of the OER performance of $Al_{X\%}$-Ni_3S_2/NF (x=3.72, 5.85 and 7.63).

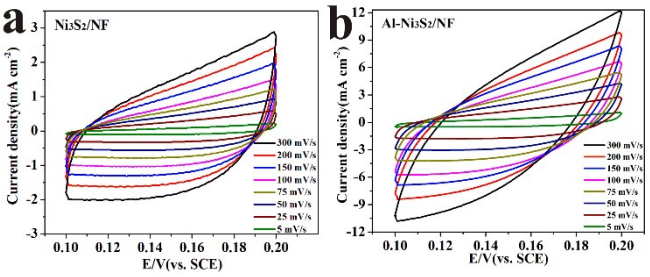

Fig. S13 Typical cyclic voltammetry curves of (a) Ni_3S_2/NF and (b) $Al_{5.85\%}$-Ni_3S_2/NF with different scan rates in 1M KOH.

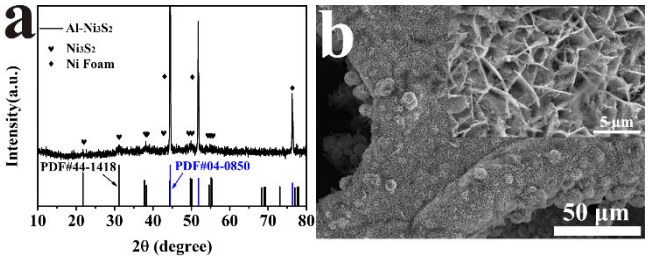

Fig. S14 (a) XRD pattern and (b) SEM of $Al_{5.85\%}$-**Ni_3S_2/NF** after OER.

Table S1. The chemical composition of Al-Ni_3S_2 and Ni_3S_2 measured by XPS.

Samples	Atomic Percentage (At %)				F Doping concentration (At %)
	Ni	Al	S	O	$\frac{n(Al)}{n(Al)+n(Ni)}$
Ni_3S_2	14.45	--	9.72	29.02	--
$Al_{5.85\%}$-Ni_3S_2	14.30	0.89	9.49	28.72	5.85

Table S2. Comparison of the HER performance of $Al_{5.85\%}$-Ni_3S_2/NF with other reported catalysts in 1 M KOH.

Catalysts	Overpotential at			Tafel slope	References
	10 mA cm^{-2}	20 mA cm^{-2}	100 mA cm^{-2}	mV dec^{-1}	
$Al_{5.85\%}$-Ni_3S_2/NF	86	117	202	75	This work
MoS_2/Co_9S_8/Ni_3S_2/Ni	113	--	--	58	[1]
Ni_3S_2/MnO_2/NF	102	--	197	69	[2]
$P_{9.03\%}$-(Ni, Fe)$_3S_2$/NF	98	135	218	88	[3]
$Fe_{17.5\%}$- Ni_3S_2/NF	47	142	232	95	[4]
Sn-Ni_3S_2/NF	137	200	320	51	[5]
V-Ni_3S_2/NF	--	203	350	112	[6]
$Ni_{1.5}Fe_{0.5}P$/CF	158	--	319	125	[7]

Table S3. Comparison of the OER performance of $Al_{5.85\%}$-Ni_3S_2/NF with other reported catalysts in 1 M KOH.

Catalysts	Overpotential at 10 mA cm^{-2}	20 mA cm^{-2}	100 mA cm^{-2}	Tafel slope mV dec^{-1}	References
$Al_{5.85\%}$-Ni_3S_2/NF	223	233	280	37	This work
Ni/NiS	--	320	390	109.03	[8]
$Ni_{1.5}Fe_{0.5}P$/CF	--	--	293	55	[7]
Fe-Ni_3S_2/FeNi	282	--	--	54	[9]
NiFe LDH@NiCoP/NF	220	--	--	48.6	[10]
$P_{9.03\%}$-$(Ni, Fe)_3S_2$/NF	196	202	242	30	[3]
MoS_2/Ni_3S_2	218	--	290	88	[11]
NC–NiCu–NiCuN	232	--	295	41	[12]

Table S4. Comparison of the Overall water splitting performance of $Al_{5.85\%}$-Ni_3S_2/NF with other well-performed electrocatalysts.

Catalysts	Overpotential at 10 mA cm^{-2}	20 mA cm^{-2}	100 mA cm^{-2}	References
$Al_{5.85\%}$-Ni_3S_2/NF	1.58 V	1.63 V	1.80 V	This work
$Fe_{11.1\%}$-Ni_3S_2/NF	1.60 V	1.66 V	--	[13]
$P_{9.03\%}$-$(Ni, Fe)_3S_2$ /NF	1.54 V	1.58 V	1.72 V	[3]
MoS_2/Ni_3S_2	1.56 V	--	--	[11]
$Ni_{1.5}Fe_{0.5}P$/CF	1.54 V	1.58 V	1.72	[7]

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