Supporting Information for

Interface Engineering of Ag-Ni_3S_2 heterostructures toward Efficient Alkaline Hydrogen Evolution

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Turnover frequency (TOF) calculation: Turnover frequency is used to reflect the number of reactant molecule conversions per active site per unit time. Assuming that all Ni atoms participate in HER, according to the following formula:

$$\mathrm{TOF} = \frac{j \times A}{2 \times \mathrm{n} \times F}$$

Where j is the measured current density, A is the surface area of the electrocatalyst, F is the Faraday constant (96485.3 C mol^{-1}), and n is the number of moles of Ni on the electrode.

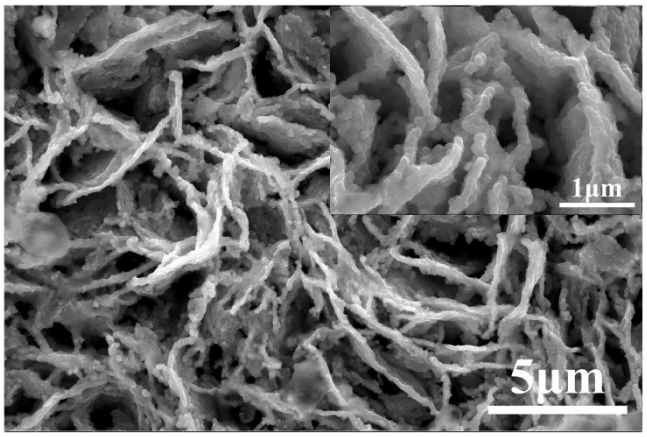

Fig. S1. SEM image of Ni_3S_2/NF.

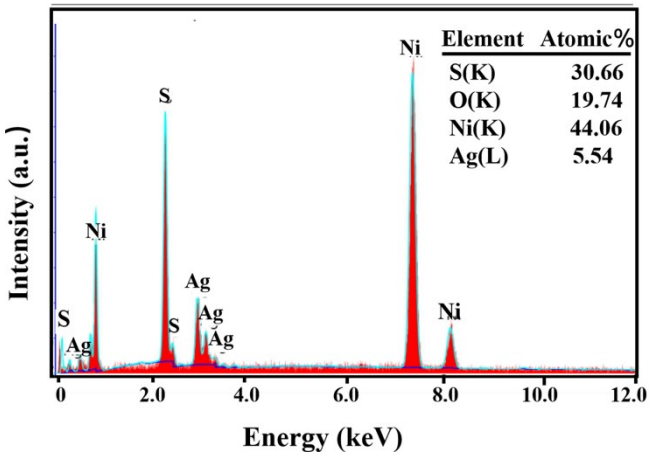

Fig. S2. EDS image of Ag-Ni_3S_2/NF-5min.

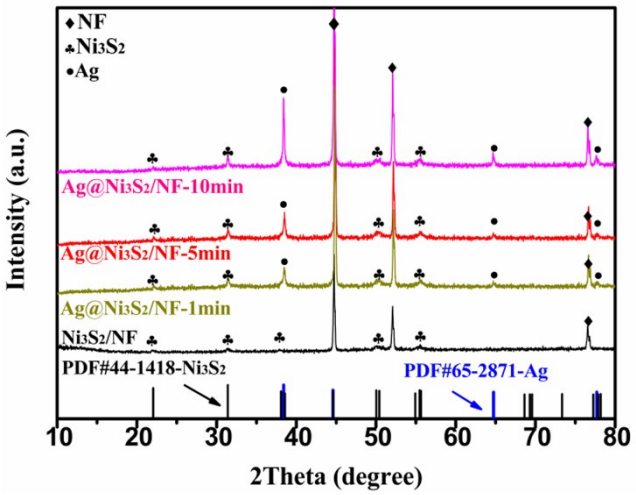

Fig. S3. XRD pattern of Ag-Ni_3S_2/NF-1min, Ag-Ni_3S_2/NF-5min Ag-Ni_3S_2/NF-10min and Ni_3S_2/NF.

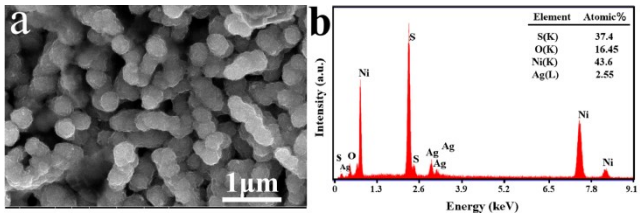

Fig. S4. (a) SEM image, (b) EDS spectrum of Ag-Ni_3S_2/NF-1min.

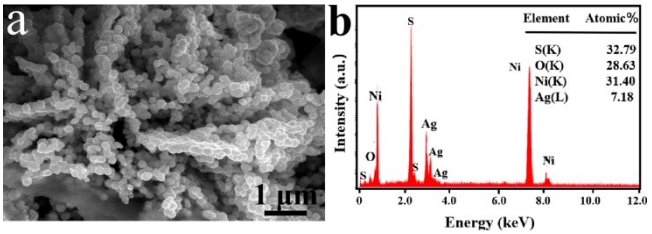

Fig. S5. (a) SEM image, (b) EDS spectrum of Ag-Ni_3S_2/NF-10min.

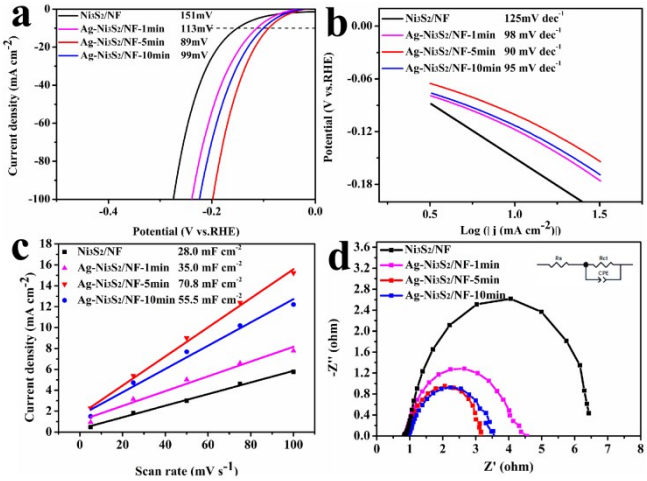

Fig. S6. (a) LSV curves, (b) Tafel slope, (c) Calculated electrochemical double-layer capacitances and (d) Nyquist plots of Ni_3S_2/NF and Ag-Ni_3S_2/NF with different Ag loading amount toward HER in 1M KOH.

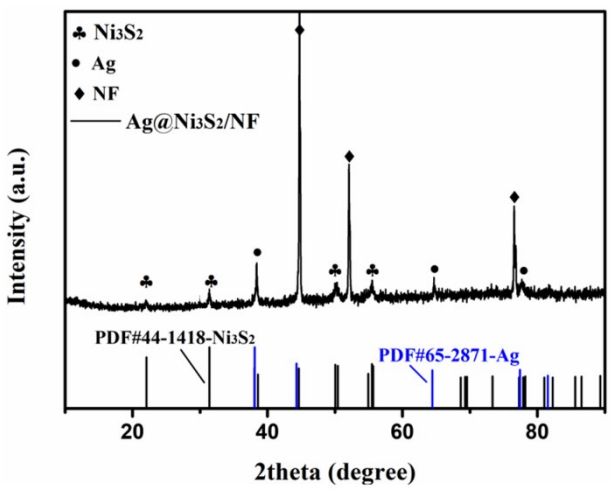

Fig. S7. XRD pattern of Ag-Ni_3S_2/NF after a stability test for 15 h.

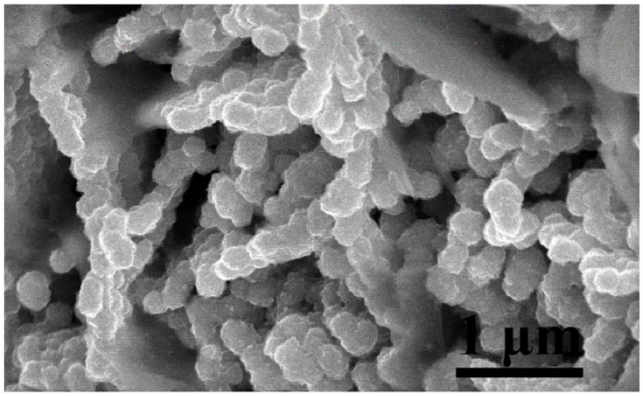

Fig. S8. SEM image of Ag-Ni_3S_2/NF after a stability test for 15 h.

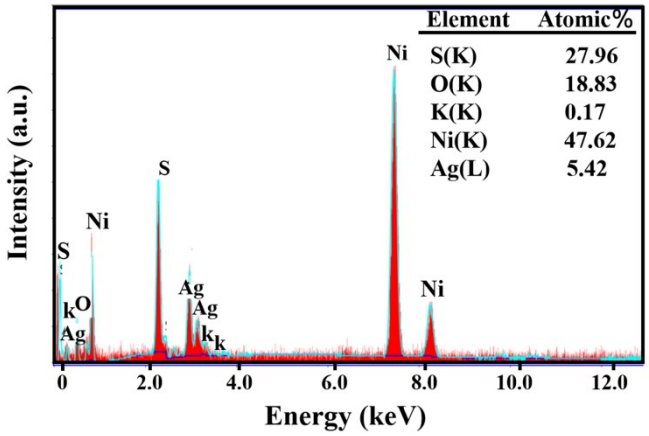

Fig. S9. EDS spectrum of Ag-Ni_3S_2/NF after a stability test for 15 h.

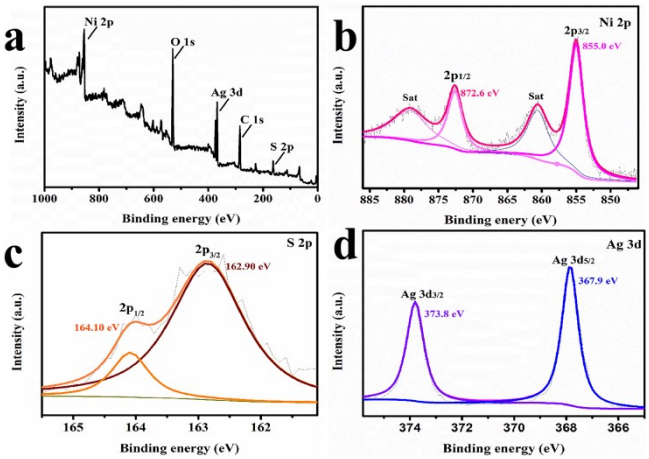

Fig. S10. (a) XPS survey, (b) Ni 2p, (c) S 2p, and (d)Ag 3d XPS spectra of Ag-Ni_3S_2/NF after a stability test for 15 h.

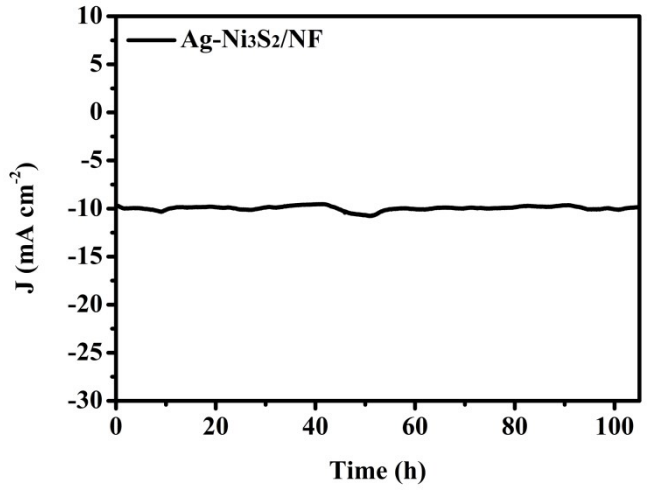

Fig.S11. long-term stability test curve of Ag-Ni_3S_2/NF under an overpotential of -103 mV (vs RHE) for 105 h.

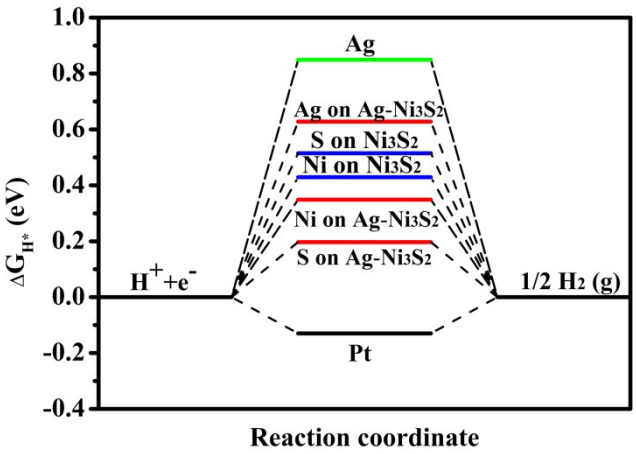

Fig. S12. Gibbs Free energy change of hydrogen on different sites of Ag-Ni_3S_2 and Ni_3S_2.

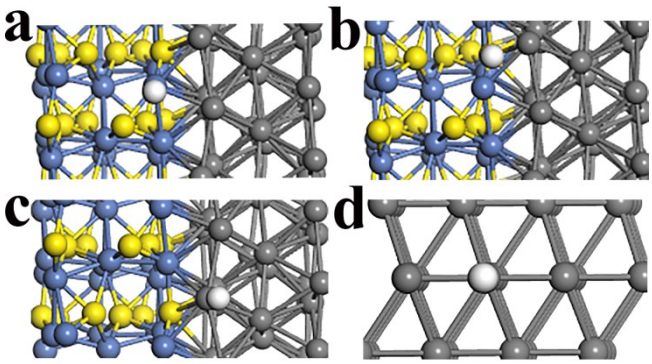

Fig. S13. DFT calculation models for ΔG_{H^*} on (a) Ni site, (b) S site, (c) Ag site of Ag-Ni_3S_2 and (d) Ag. (Hydrogen atom: white）

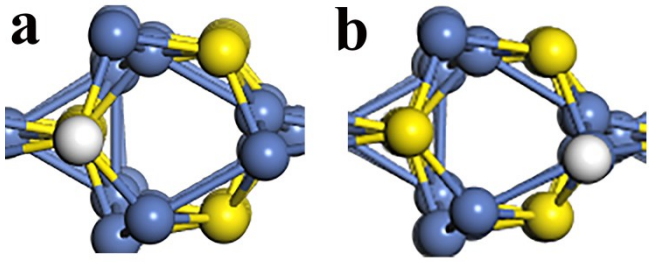

Fig. S14. DFT calculation models for ΔG_{H^*} on (a) S site and (b) Ni site of Ni_3S_2（Hydrogen atom: white）

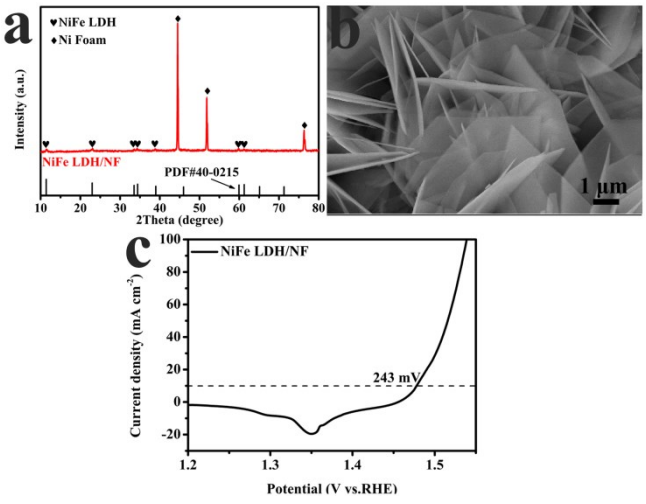

Fig. S15. (a) XRD pattern, (b) SEM image and (c) LSV curve of the NiFe LDH/NF in 1M KOH.

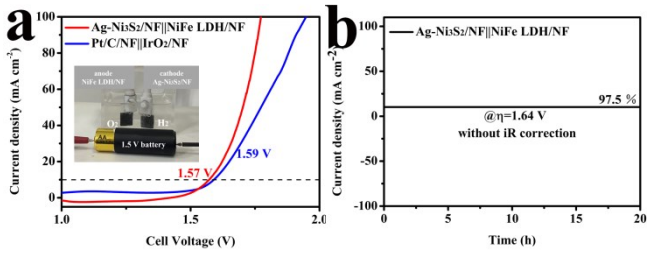

Fig. S16. (a) Overall water splitting performance LSV curves of Ag-Ni_3S_2/NF||NiFe LDH/NF, Pt/C/NF||IrO_2/NF and Ni_3S_2/NF||Ni_3S_2/NF electrode pairs in 1 M KOH. (Inset: Image of a two-electrode water splitting unit driven by a battery with a rated voltage of 1.5 V) (b) Long-term stability test of Ag-Ni_3S_2/NF||NiFe LDH/NF for water splitting at 1.64 V.

Table S1. Loading amounts of Ag-Ni_3S_2/NF-1min, Ag-Ni_3S_2/NF-5min and Ag-Ni_3S_2/NF-10min.

Sample	Ag-Ni_3S_2/NF-1min	Ag-Ni_3S_2/NF-5min	Ag-Ni_3S_2/NF-10min
Loading amount (At %)	12.0%	26.5%	30.4%

Table S2. Comparison of the HER performance of Ag-Ni_3S_2/NF with other reported catalysts in 1 M KOH.

Catalysts	Overpotential at 10 mA cm^{-2} (mV)	Tafel slope ($mV\ dec^{-1}$)	References
Ag-Ni_3S_2/NF	89	90	This work
NF@Ni_3S_2@NCNT	~94	54	[1]
NF-Ni_3S_2/MnO_2	102	69	[2]
NF/T(Ni_3S_2/MnS-O)	116	41	[3]
Ni_3S_2/MnS/NF	136	49	[3]
NiFe@Ni_3S_2	184	77	[4]
Cu NDs/Ni_3S_2NTs-CFs	128	76.2	[5]
CoMoNiS-NF	113	85	[6]
Co_9S_8-Ni_3S_2/NF	163	121	[6]
$Ni_xCo_{3-x}S_4$/Ni_3S_2/NF	136	95	[7]
NiP	234	130	[8]
Ni_3S_2-300	177	75.5	[9]
Sn-Ni_3S_2/NF	137	51	[10]
P-$(Ni,Fe)_3S_2$/NF	98	88	[11]
$Ni_{0.9}Fe_{0.1}PS_3$NS	72	73	[12]
$Fe_{17.5\%}$-Ni_3S_2/NF	47	95	[13]

Table S3. EIS and C_{dl} results of Ni_3S_2/NF, Ag-Ni_3S_2/NF-1min, Ag-Ni_3S_2/NF-5min and Ag-Ni_3S_2/NF-10min.

Sample	Ni_3S_2	Ag-Ni_3S_2/NF-1min	Ag-Ni_3S_2/NF-5min	Ag-Ni_3S_2/NF-10min
R_{ct} [Ω]	5.60	3.61	2.10	2.52
C_{dl} [mF cm^{-2}]	28.00	35.00	70.80	55.50

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