Electronic Supplementary Material (ESI) for Inorganic Chemistry Frontiers.
This journal is © the Partner Organisations 2020

Supplementary information:

Uncover the role of Ag in layer-alternating Ni_3S_2/Ag/Ni_3S_2 as electrocatalyst with enhanced OER performance

Rui Guo[1,2,3], Yan He[1,3,*], Renchao Wang[1,3], Junhua You[4], Hongji Lin[5], Chiente Chen[5], Tingshan Chan[5], Xuanwen Liu[1,3,*], Zhiwei Hu[6,*]

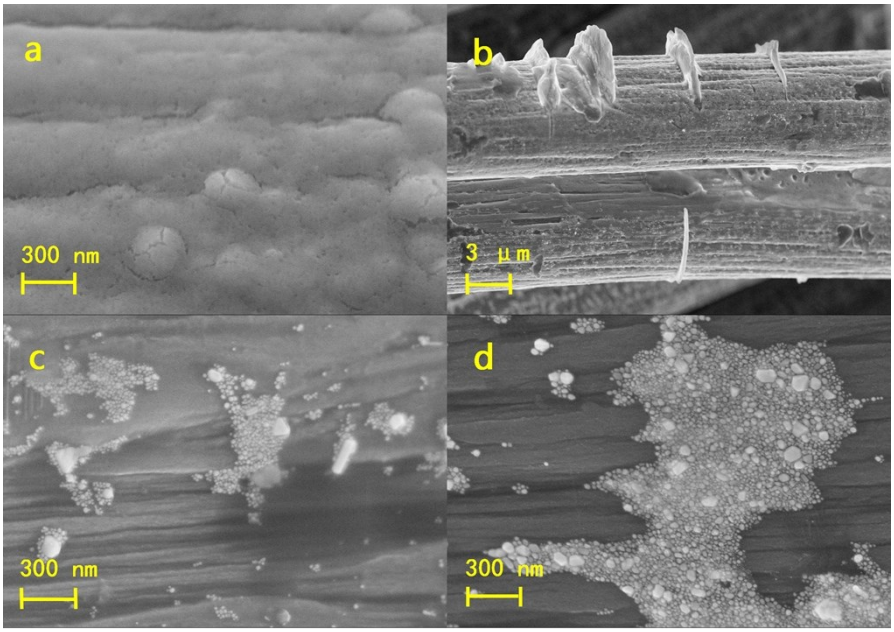

Fig. S1. SEM images of Ag samples by electrodeposition for (a) 2 s, (b) 4 s, (c) 6 s, (d) 8 s.

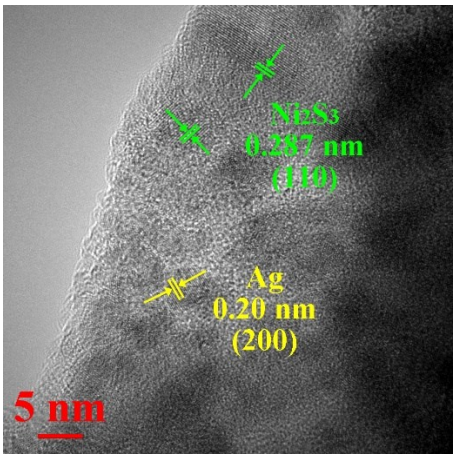

Fig. S2. The HRTEM images of N1A1.

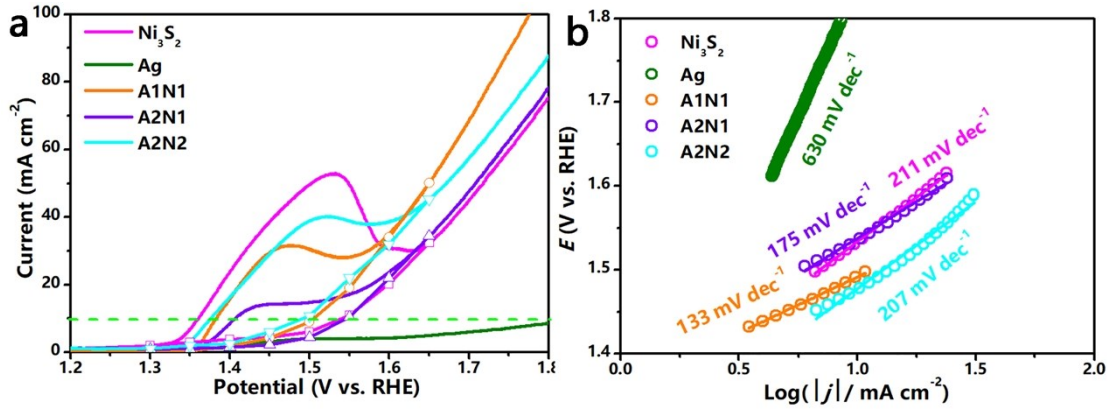

Fig. S3. (a) The OER polarization curves and (b) Tafel plots of Ni_3S_2, Ag, A1N1, A2N1, and A2N2

Table S1. Comparision of the OER performance of N2A1 with Ni_3S_2 electrocatalysts in alkaline electrolyte.

Catalysts	η_{10} (mV)	Tafel Slopel (mV dec^{-1})	Electrolyte	Ref.
Ni_3S_2 nanoporous thin films	319 @ 20 mA cm^{-2}	101.2	1.0 M KOH	S1
Ni_3S_2/VS_2	227	59.9	1.0 M NaOH	S2
Fe-doped Ni_3S_2 particles film	253 @ 100 mA cm^{-2}	65.5	1.0 M KOH	S3
Ni_3S_2 nanorods/NF	157	153	1.0 M KOH	S4
$Fe_{17.5\%}$-Ni_3S_2/NF	214	42	1.0 M KOH	S5
Ni_3S_2/NF-2	425@ 100 mA cm^{-2}	-	1.0 M KOH	S6
Co_3O_4@Ni_3S_2/NF	260 @ 20 mA cm^{-2}	121.7	1.0 M KOH	S7
CdS/Ni_3S_2/PNF	151	174	1.0 M KOH	S8
Ag/Ni_3S_2 (N2A1)	187	118	1.0 M KOH	This work

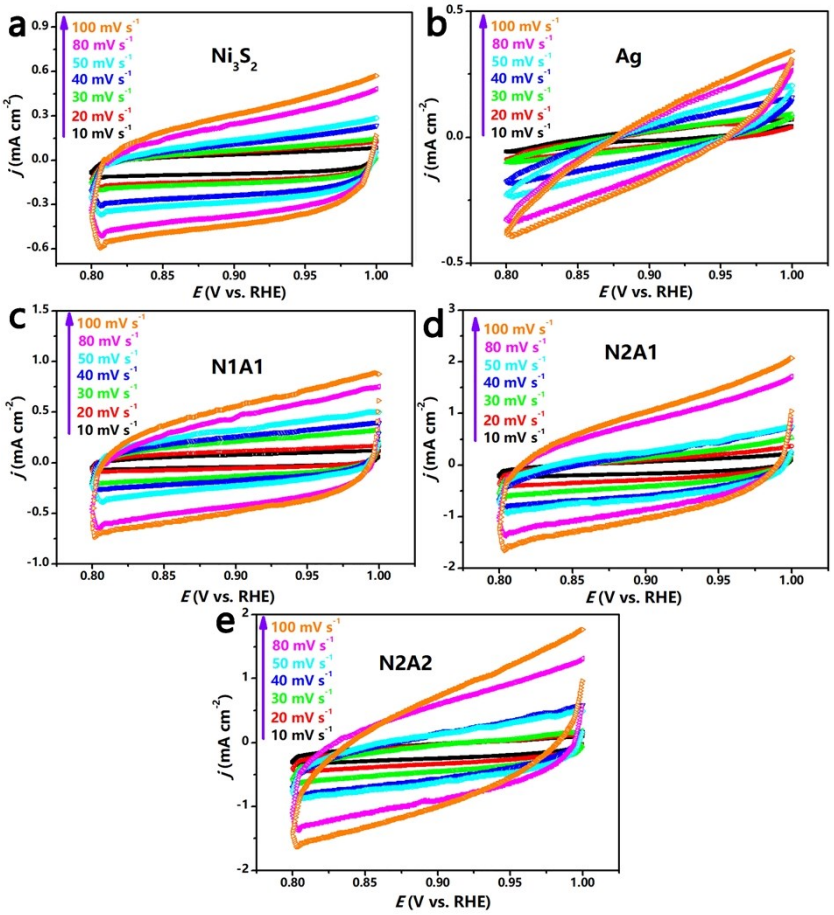

Fig. S4. Cyclic voltammogram curves of Ni_3S_2 (a), Ag (b), N1A1 (c), N2A1 (d), and N2A2 (d) measured in the range of 0.8 ~ 1.0 V vs. RHE at different scan rates: 10, 20, 30, 40, 50, 80, and 100 mV s^{-1}.

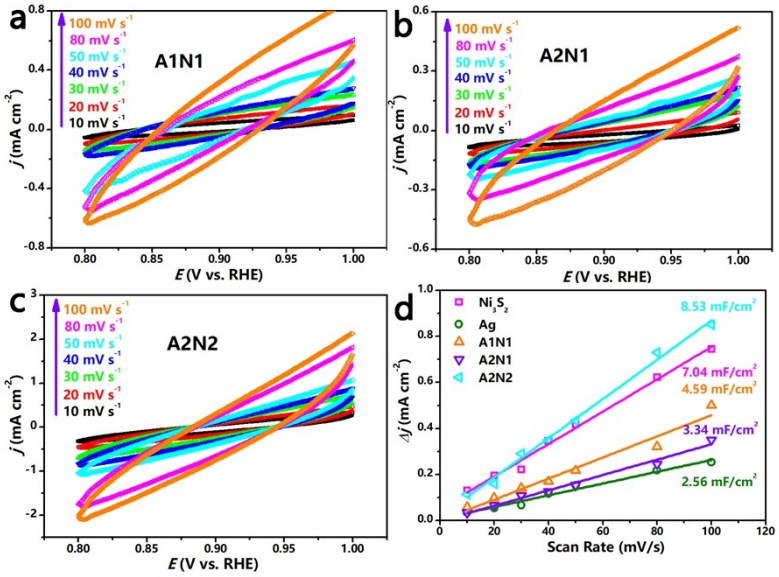

Fig. S5. Cyclic voltammogram curves of A1N1 (a), A2N1 (b), and A2N2 (c) measured in the range of 0.8 ~ 1.0 V vs. RHE at different scan rates: 10, 20, 30, 40, 50, 80, and 100 mV s^{-1}. (d)

Double layer capacitance (Cdl) of as-prepared electrocatalysts.

Table S2. Electrocatalytic performances of different samples

Sample	OER		R_{ct}/Ω	W/Ω
	η_j @ 10 mA cm^{-2}	Tafel slope (mV dec^{-1})		
Ag	-	630	6.25	53.31
Ni3S2	308	211	0.56	3.42
N1A1	289	190	0.41	2.14
N2A1	187	118	0.19	1.62
N2A2	233	154	0.50	1.61

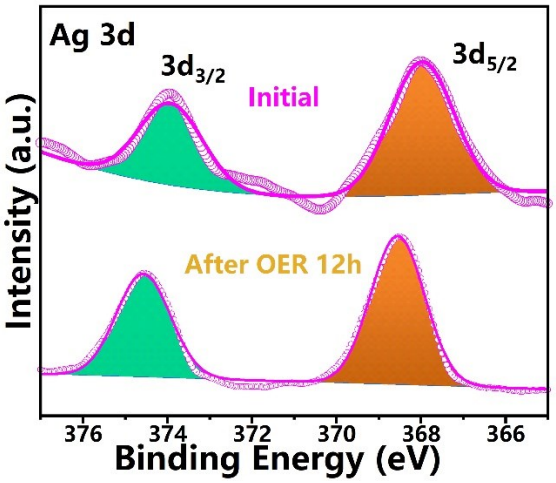

Fig. S6. High resolution XPS spectrums of Ag 3d before and after OER for 12 h.

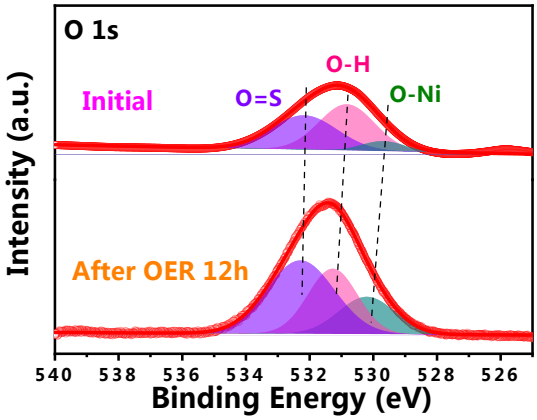

Fig. S7. High resolution XPS spectrums of O 1S of N2A1 before and after OER for 12 h.

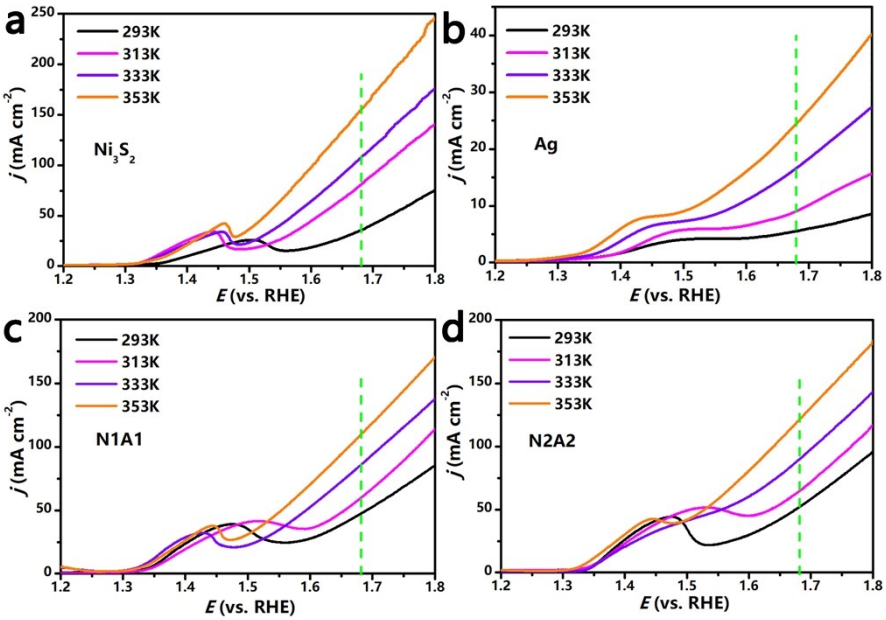

Fig. S8. LSV curves of Ni_3S_2 (a), Ag (b), N1A1 (c), and N2A2 (d) at a scan rate of 5 mV s^{-1} in 1.0 M KOH towards OER as the temperature rose from 293 to 353 K.

Reference

S1 J. Dong, F. Zhang, Y. Yang, Y. Zhang, H. He, X. Huang, X. Fan and X. Zhang, (003)-Facet-exposed Ni_3S_2 nanoporous thin films on nickel foil for efficient water splitting, *Appl. Catal. B*, 2019, **243**, 693-702.

S2 X. Zhong, J. Tang, J. Wang, M. Shao, J. Chai, S. Wang, M. Yang, Y. Yang, N. Wang, S. Wang, B. Xu and H. Pan, 3D heterostructured pure and N-Doped Ni_3S_2/VS_2 nanosheets for high efficient overall water splitting, *Electrochim. Acta*, 2018, **269**, 55-61.

S3 N. Cheng, Q. Liu, A. M. Asiri, W. Xing and X. Sun, A Fe-doped Ni_3S_2 particle film as a high-efficiency robust oxygen evolution electrode with very high current density, *J. Mater. Chem. A*, 2015, **3**, 23207-23212.

S4 W. Zhou, X. Wu, X. Cao, X. Huang, C. Tan, J. Tian, H. Liu, J. Wang and H. Zhang, Ni_3S_2 nanorods/Ni foam composite electrode with low overpotential for electrocatalytic oxygen evolution, *Energy Environ. Sci.*, 2013, **6**, 2921-2924.

S5 G. Zhang, Y. Feng, W. Lu, D. He, C. Wang, Y. Li, X. Wang and F. Cao, Enhanced Catalysis of Electrochemical Overall Water Splitting in Alkaline Media by Fe Doping in Ni_3S_2 Nanosheet Arrays, *ACS Catal.*, 2018, **8**, 5431-5441.

S6 G. Liu, Z. Sun, X. Zhang, H. Wang, G. Wang, X. Wu, H. Zhang and H. Zhao, Vapor-phase hydrothermal transformation of a nanosheet array structure $Ni(OH)_2$ into ultrathin Ni_3S_2 nanosheets on nickel foam for high-efficiency overall water splitting, *J. Mater. Chem. A*, 2018, **6**, 19201-19209.

S7 Y. Gong, Z. Xu, H. Pan, Y. Lin, Z. Yang and X. Du, Hierarchical Ni_3S_2 nanosheets coated on Co_3O_4 nanoneedle arrays on 3D nickel foam as an efficient electrocatalyst for the oxygen evolution reaction, *J. Mater. Chem. A*, 2018, **6**, 5098-5106.

S8 S. Qu, J. Huang, J. Yu, G. Chen, W. Hu, M. Yin, R. Zhang, S. Chu and C. R. Li, Ni_3S_2 Nanosheet Flowers Decorated with CdS Quantum Dots as a Highly Active Electrocatalysis Electrode for Synergistic Water Splitting, *ACS Appl. Mater. Interfaces*, 2017, **9**, 29660-29668.