Supplementary Information

Plasma-engineered Oxygen Vacancies on NiFe Sulfide for Highly Efficient and Durable Oxygen Evolution Reaction

Qiulin Xu, Yongqiang Ni, Jinyue Pan, Yongting Chen and Qin Zhang**

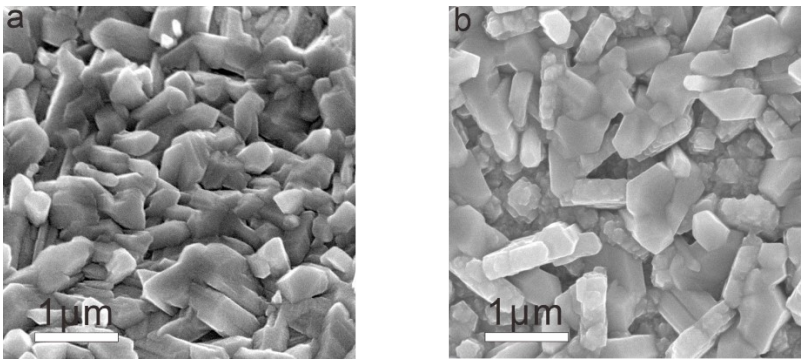

Figure S1. SEM image of (a) p-Fe-Ni_3S_2/NF-2; (b) p-Fe-Ni_3S_2/NF-8.

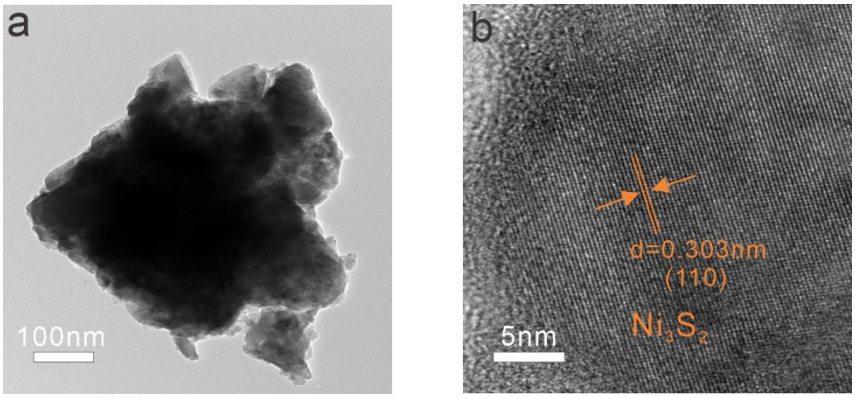

Figure S2. (a) TEM image of Fe-Ni_3S_2/NF. (b) HR-TEM image of Fe-Ni_3S_2/NF.

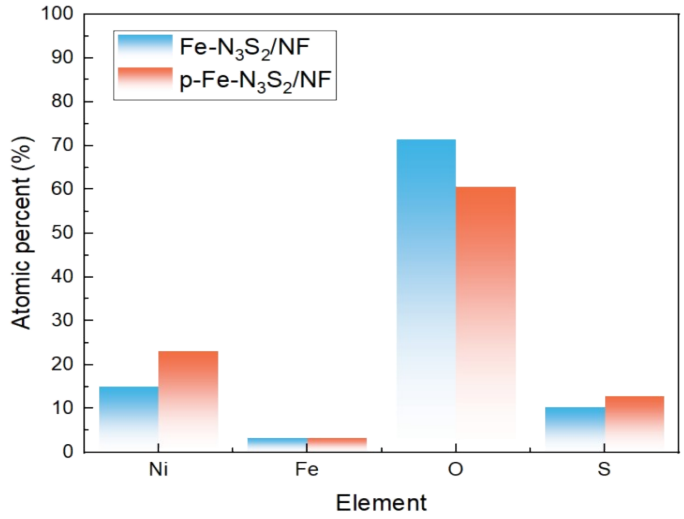

Figure S3. The atomic percent of the Fe-Ni_3S_2/NF and p-Fe-Ni_3S_2/NF samples

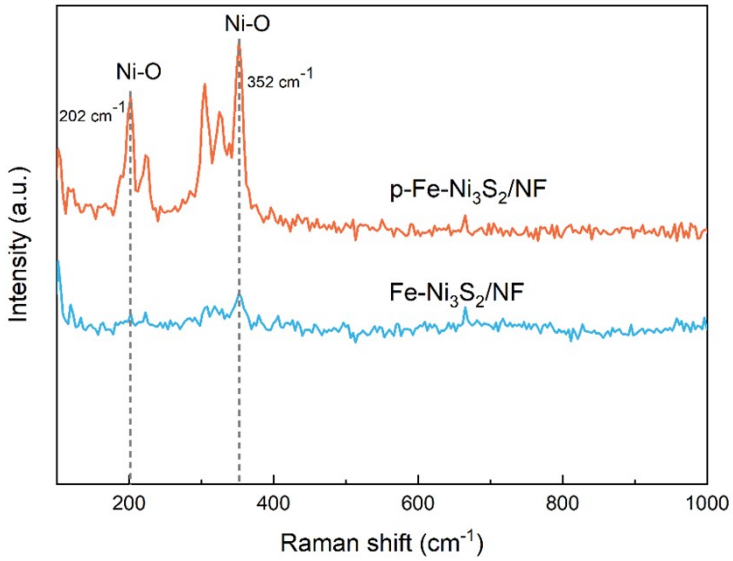

Figure S4. The Raman spectra of the Fe-Ni_3S_2/NF and p-Fe-Ni_3S_2/NF samples

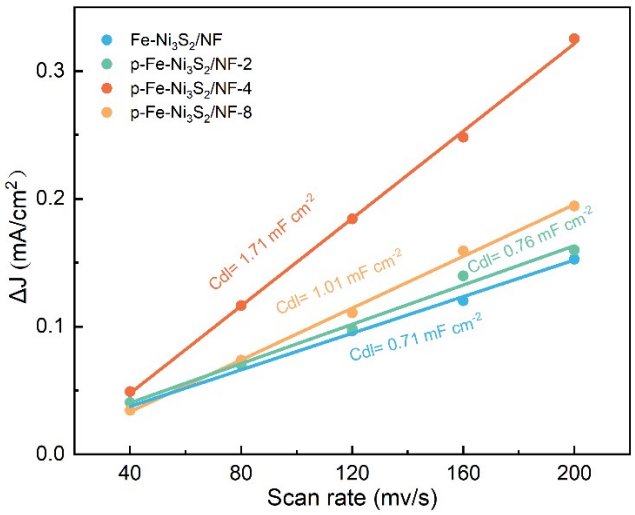

Figure S5. Derived double layer capacitance Cdl of the electrocatalysts at varied plasma exposure times

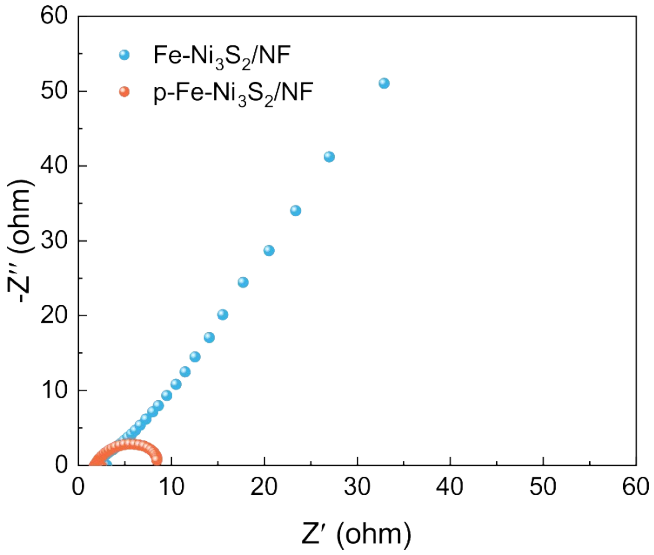

Figure S6. Electrochemical impedance diagrams of the Fe-Ni_3S_2/NF and p-Fe-Ni_3S_2/NF samples

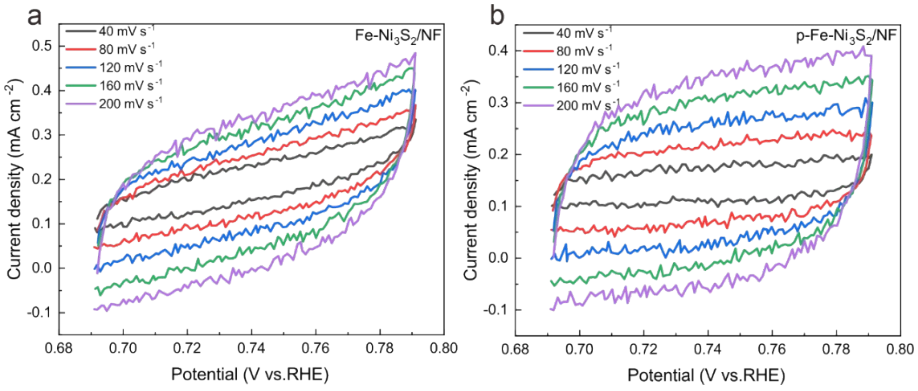

Figure S7. (a)Cyclic voltammetry curves of Fe-Ni_3S_2/NF at different scan rates. (b) Cyclic voltammetry curves of p-Fe-Ni_3S_2/NF at different scan rates

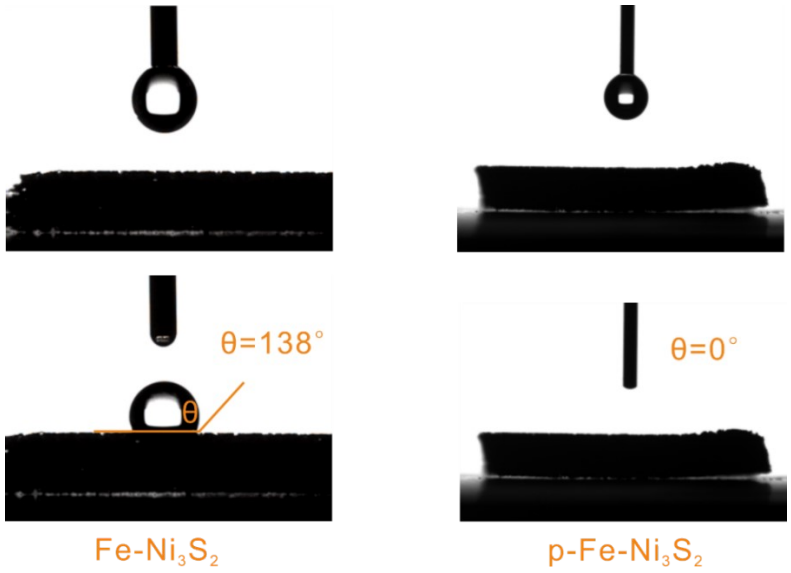

Figure S8. Contact-angle measurements of the Fe-Ni_3S_2/NF and p-Fe-Ni_3S_2/NF samples

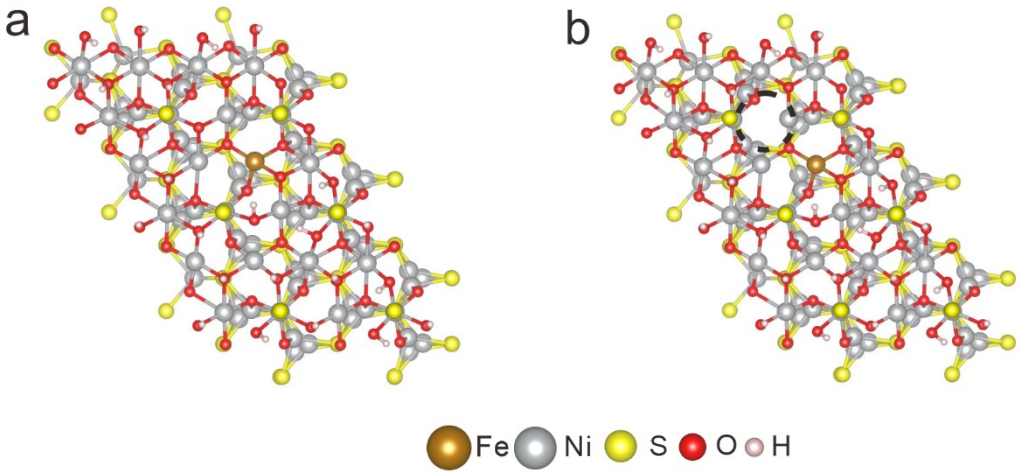

Figure S9. Density-functional theory computational models of (a) Fe-Ni_3S_2/NF. (b) p-Fe-Ni_3S_2/NF.

Table S1. The EDS elemental content of samples with different plasma treatment times

	Ni (At%)	Fe (At%)	S (At%)	O (At%)
Fe–Ni_3S_2/NF	62.68	0.60	36.16	0.55
p–Fe–Ni_3S_2/NF–2	46.37	9.84	38.77	5.02
p–Fe–Ni_3S_2/NF–4	34.51	10.63	19.87	34.99
p–Fe–Ni_3S_2/NF–8	51.11	19.31	17.63	11.94

Table S2. Comparison of the OER activity of p-Fe-Ni_3S_2 with that of recently reported electrocatalysts at 100 mA cm^{-2} in 1 M KOH.

Catalyst	**η_{100}(mV)**	**Tafel plot (mV dec^{-1})**	**References**
p-Fe-Ni_3S_2	**284**	**47.7**	**This work**
Co/Fe-SNC800	310	47.9	1
Ni_3S_2-rGO/NF	303	23.0	2
Ni_3S_2@NiFeOOH-	306	68.9	3
Co-Mo/FeS	321	30.7	4
FeCo-Ni_3S_4	279	60.3	5
NiCo-based hybrids	331	82.1	6
CeO_2-Ni_3S_2/NF	364	60.0	7
Mo-NiS_x/NF	400	76.2	8
FeOOH/Ni_3S_2	308	156.9	9
Ni_2P-CoCH/CFP	320	36.0	10

Table S3. Gibbs free energy of Fe–Ni_3S_2/NF and p–Fe–Ni_3S_2/NF catalyst for all OER elementary steps

	ΔG_1 (eV)	ΔG_2 (eV)	ΔG_3 (eV)	ΔG_4 (eV)
Fe–Ni_3S_2/NF	0.36	0.69	2.29	1.56
p–Fe–Ni_3S_2/NF	0.27	0.81	2.21	1.63

References

1. C. Chen, M. Sun, F. Zhang, H. Li, M. Sun, P. Fang, T. Song, W. Chen, J. Dong, B. Rosen, P. Chen, B. Huang and Y. Li, *Energy & Environmental Science*, 2023, **16**, 1685-1696.
2. Z. Zhao, J. Liao, R. Shi, X. Zhao, Z. Wang, J. Li, Q. Li, D. Yang, M. Nie and Y. Liu, *International Journal of Hydrogen Energy*, 2023, **48**, 27441-27449.
3. J. Luo, Y. Zhang, L. Tang, D. Lu, T. Zhang, H. Zhao, W. Jiang and C. Liu, *International Journal of Hydrogen Energy*, 2025, **114**, 394-402.
4. R.-N. Luan, Q.-X. Lv, Y.-Y. Li, J.-Y. Xie, W.-J. Li, H.-J. Liu, R.-Q. Lv, Y.-M. Chai and B. Dong, *International Journal of Hydrogen Energy*, 2023, **48**, 25730-25740.
5. X. Mao, Y. Liu, Z. Chen, Y. Fan and P. Shen, *Chemical Engineering Journal*, 2022, **427**, 130742-130751.
6. M. Chen, D. Liu, J. Feng, P. Zhou, L. Qiao, W. Feng, Y. Chen, K. Wei Ng, S. Wang, W. Fai Ip and H. Pan, *Chemical Engineering Journal*, 2022, **443**, 136432-136429.
7. Z. Huang, X. Liao, W. Zhang, J. Hu and Q. Gao, *ACS Catalysis*, 2022, **12**, 13951-13960.
8. S. Ghosh, B. Dasgupta, S. Kalra, M. L. P. Ashton, R. Yang, C. J. Kueppers, S. Gok, E. G. Alonso, J. Schmidt, K. Laun, I. Zebger, C. Walter, M. Driess and P. W. Menezes, *Small*, 2023, **19**, 2206679.
9. Y. Liu, M. Ding, Y. Tian, G. Zhao, J. Huang and X. Xu, *Journal of Colloid and Interface Science*, 2023, **639**, 24-32.
10. S. Zhang, C. Tan, R. Yan, X. Zou, F. L. Hu, Y. Mi, C. Yan and S. Zhao, *Angewandte Chemie International Edition*, 2023, **62**, e202302795.