

Supporting Information

CoFe layered double hydroxides with adjustable composition and structure for enhanced oxygen evolution reaction

Wan Rong,^{ab} Rui Dang,^b Yunfei Chen,^b Kang Huang,^a Jiuyang Xia,^a Bowei Zhang,^{*a} Jianfei Liu,^b Meixin Li,^c Qigao Cao ^{*b} and Junsheng Wu ^{*a}

¹Institute of Advanced Materials and Technology, University of Science and Technology Beijing, Beijing 100083, P.R. China.

²Northwest Institute For Nonferrous Metal Research, 96 Weiyang Road, Xi'an, Weiyang district, 710016, P.R. China.

³The Faculty of Printing, Packaging Engineering and Digital Media Technology, Xi'an University of Technology, Xi'an 710054, P.R. China.

Corresponding Author

¹Institute of Advanced Materials and Technology, University of Science and Technology Beijing, Beijing 100083, P.R.China.

*Email: bwzhang@ustb.edu.cn (Bowe Zhang)

*Email: wujs76@163.com (Junsheng Wu)

²Northwest Institute For Nonferrous Metal Research, 96 Weiyang Road, Weiyang district, 710016, P.R. China.

*Email: caoqigao@c-nin.com (Qigao Cao)

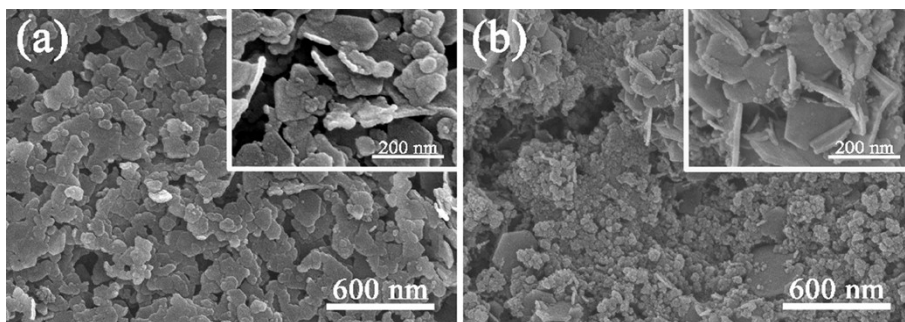


Fig. S1 SEM images of different samples: (a) Co_2Fe_1 LDH and (b) Co_1Fe_2 LDH

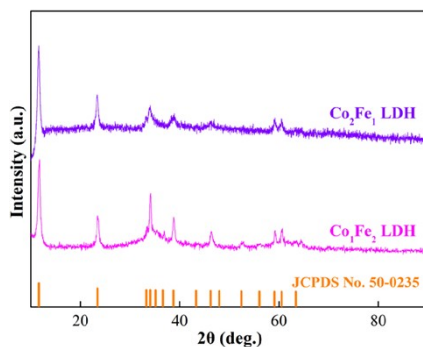


Fig. S2 XRD patterns of the Co_2Fe_1 LDH and Co_1Fe_2 LDH

Table S1. Mass fraction of Co and Fe in different LDHs defined by ICP-OES.

Sample	Co (wt. %)	Fe (wt. %)	Other elements (wt. %)
Co_2Fe_1 LDH	25.3	11.9	62.8
Co_1Fe_1 LDH	19.2	18.5	62.3
Co_1Fe_2 LDH	13.5	25.7	60.8
Co LDH	35.3	/	64.7
Fe LDH	/	41.2	58.8

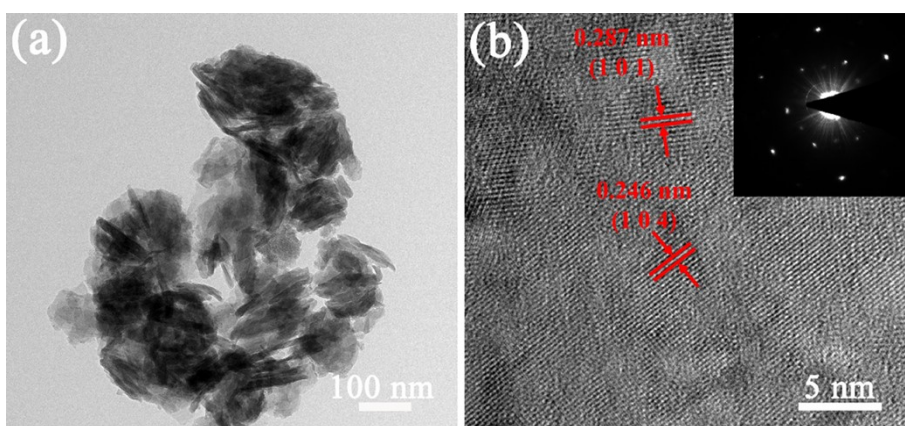


Fig. S3 (a) TEM and (b) HRTEM images of Co_2Fe_1 LDH, Inset to (b): Corresponding SAED pattern

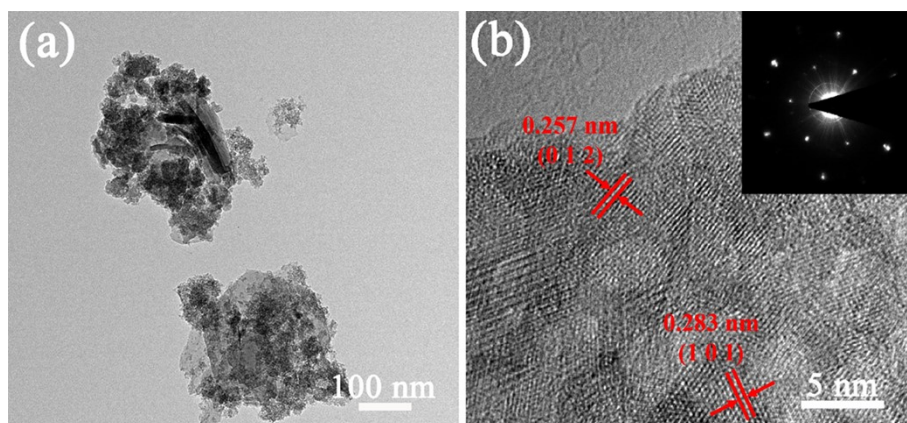


Fig. S4 (a) TEM and (b) HR-TEM images of Co_1Fe_2 LDH, Inset to (b): Corresponding SAED pattern

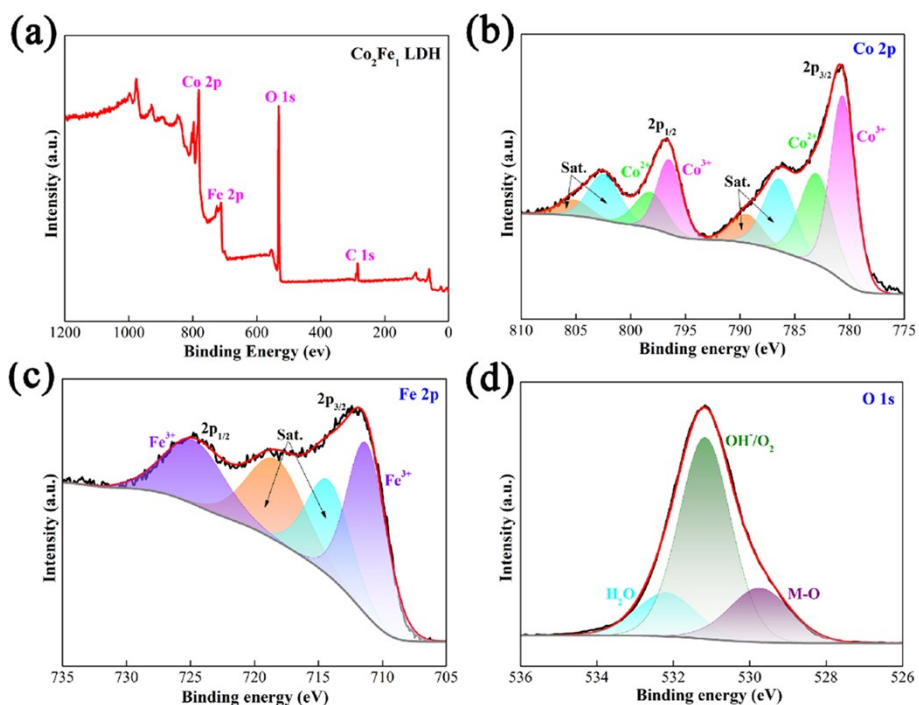


Fig. S5 Electronic structure analysis of the Co_2Fe_1 LDH: (a) XPS survey spectra, (b) Co 2p XPS spectra, (c) Fe 2p XPS spectra and (d) O 1s XPS spectra

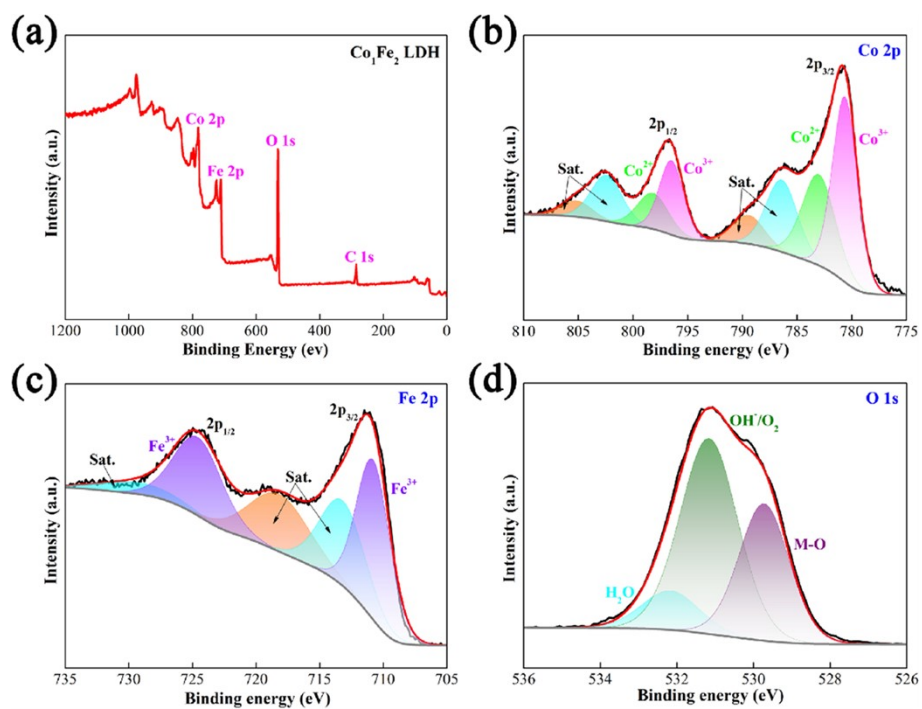


Fig. S6 Electronic structure analysis of the Co_1Fe_2 LDH: (a) XPS survey spectra, (b) Co 2p XPS spectra, (c) Fe 2p XPS spectra and (d) O 1s XPS spectra

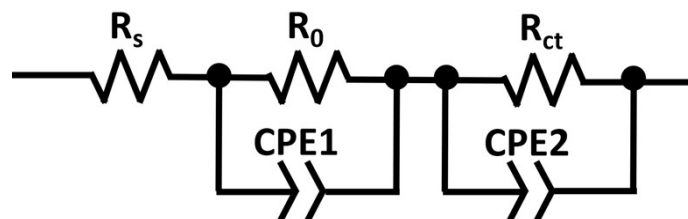


Fig. S7 Equivalent electrical circuit of electrochemical impedance spectroscopy. An equivalent electrical circuit used to model the OER process.

Table S2. The values of R_{ct} of the Co_2Fe_1 LDH, Co_1Fe_1 LDH, Co_1Fe_2 LDH, Co LDH and Fe LDH from EIS spectra.

Samples	Co_2Fe_1 LDH	Co_1Fe_1 LDH	Co_1Fe_2 LDH	Co LDH	Fe LDH	IrO_2
R_{ct} (Ω)	1.53	1.14	1.46	3.25	2.68	3.9

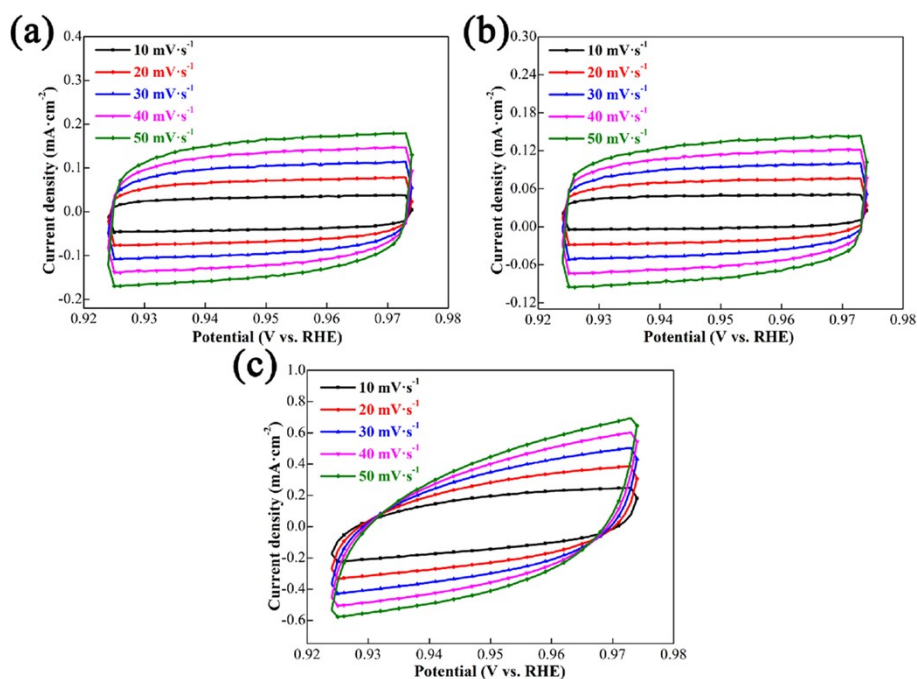


Fig. S8 The scan rate-dependent CV curves of (a) Co LDH, (b) Fe LDH and (c) Co_1Fe_1 LDH

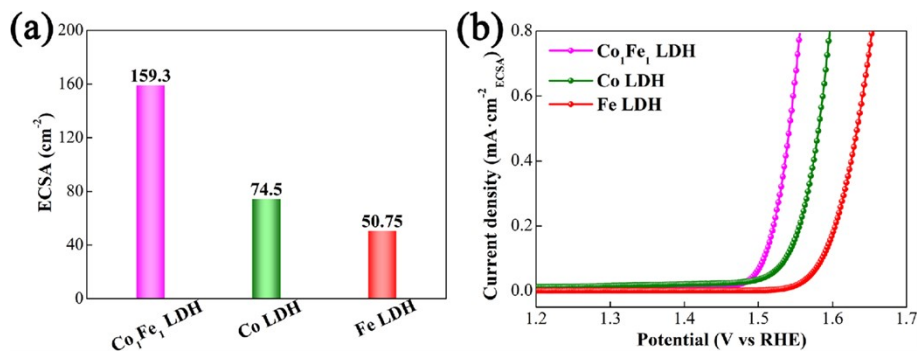


Fig. S9 (a) ECSA and (b) ECSA-normalized OER activity of the Co_1Fe_1 LDH, Co LDH and Fe LDH

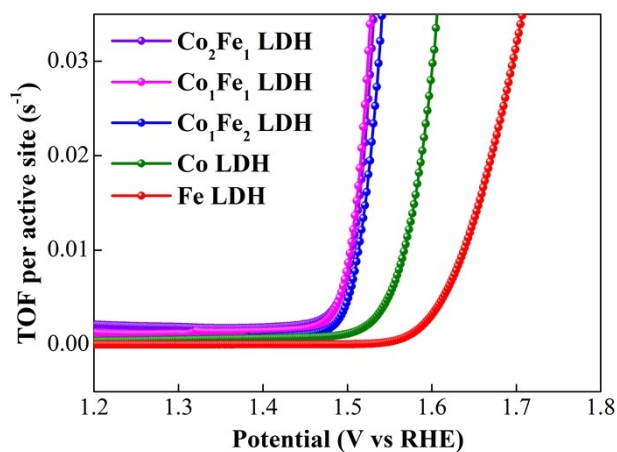


Fig. S10 TOF curves of the Co_2Fe_1 LDH, Co_1Fe_1 LDH, Co_1Fe_2 LDH, Co LDH and Fe LDH

Table S3. The values of TOFs of the Co_2Fe_1 LDH, Co_1Fe_1 LDH, Co_1Fe_2 LDH, Co LDH and Fe LDH for OER.

Samples	Co_2Fe_1 LDH	Co_1Fe_1 LDH	Co_1Fe_2 LDH	Co LDH	Fe LDH
TOFs for OER @1.53 V vs. RHE (S^{-1})	0.03295	0.03839	0.02246	0.00253	0.000096

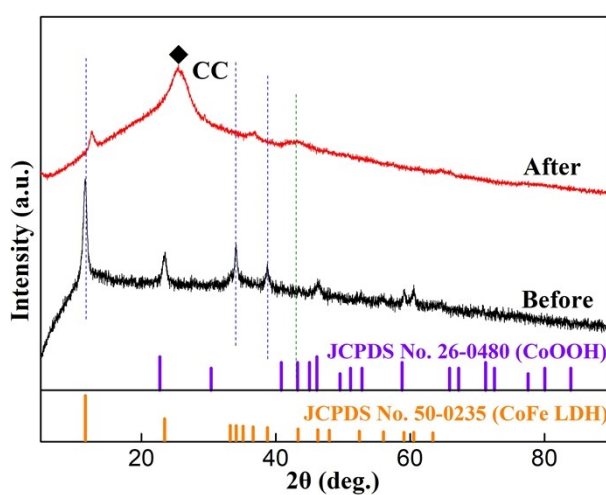


Fig. S11 XRD patterns of Co_1Fe_1 LDH before and after OER stability test

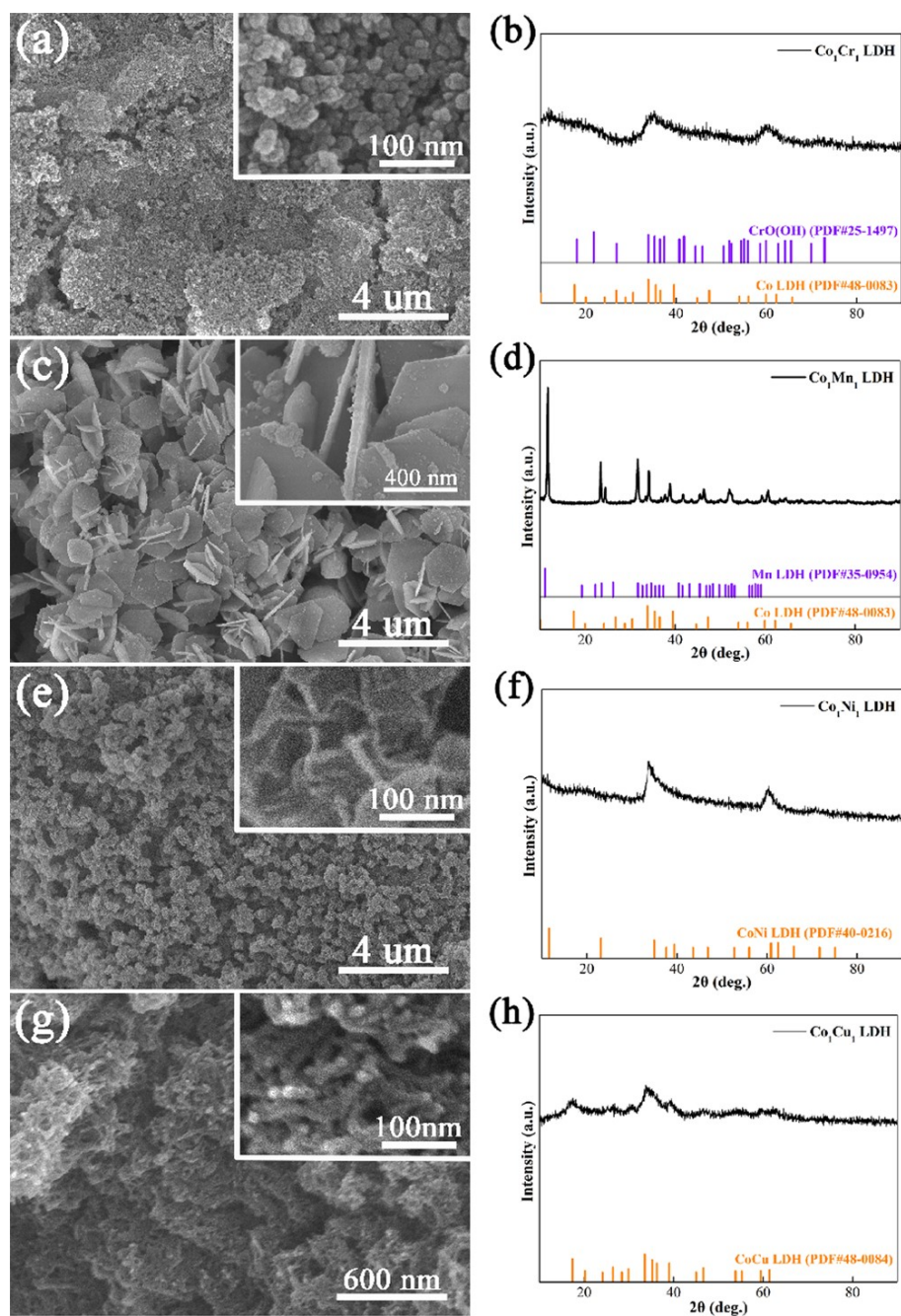


Fig. S12 SEM images of different samples: (a) $\text{Co}_1\text{Cr}_1\text{LDH}$, (c) $\text{Co}_1\text{Mn}_1\text{LDH}$, (e) $\text{Co}_1\text{Ni}_1\text{LDH}$ and (g) $\text{Co}_1\text{Cu}_1\text{LDH}$; (b,d,f,h) the corresponding PXRD patterns of them

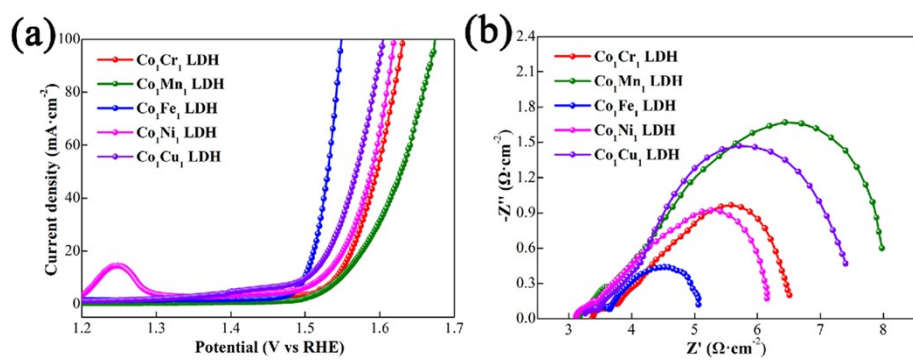


Fig. S13 The LSV and EIS curves of different $\text{Co}_1\text{M}_1\text{LDH}$ s

Table S4. Comparison of the OER activity between Co₁Fe₁ LDH with other electrocatalysts in 1.0 M KOH

Catalysts	Catalyst loading (mg cm ⁻²)	η_{10} (mV)	Tafel slope (mV dec ⁻¹)	Reference
Co₁Fe₁ LDH	1.00	270	42.7	This Work
Co-LDH FNSAs	0.18	300	110	Co-precipitation [S1]
CoFe LDH	0.71	310	72.7	Hydrothermal [S2]
Co ₁ Fe _{0.2} NPs	0.28	246	37.0	Co-precipitation [S3]
Co _{0.55} Fe _{0.45} BPO-OER	0.70	270	26.0	Hydrothermal [S4]
CoFe-LDHs	1.00	310	59.0	Co-precipitation [S5]
Co ₂ FeO ₄	0.20	293	67.0	Hydrothermal [S6]
CoFe-MOF-OH	0.21	351	48.0	Hydrothermal [S7]
Co ₄ Fe ₆ -MOF	1.00	241	30.1	Co-precipitation [S8]
CoFe LDH	0.10	404	-	Solvothermal [S9]
Co ₅ Fe ₅ O(OH)	0.20	276	52.0	Hydrothermal [S10]
Fe-Co ₃ O ₄ @C/FTO	1.10	396	68.6	MOF-derived pyrolysis [S11]
FeSe ₂	0.01	330	48.1	Solvothermal [S12]
NF-PVP/CoFe _{1.3}	2.00	234	46.4	electro-spinning [S13]
CoFe LDH	--	280	58.2	Co-precipitation [S14]

References

- [S1] T Wang, X Liu, Y Li, F Li, Z Deng and Y Chen, *Nano Res*, 2020, **13**, 79-80.
- [S2] M Li, Y Gu, Y Chang, X Gu, J Tian, X Wu and L Feng, *Chem. Eng. J.*, 2021, 130686.
- [S3] X Bai, Z Duan, B Nan, L Wang, T Tang and J Guan, *Chin. J. Catal.*, 2022, **43**, 2240–2248.
- [S4] L Reith, J Hausmann, S Mebs, I Mondal, H Dau, M Driess and P Menezes, *Adv. Energy Mater.*, 2023, 2203886-2203896.
- [S5] P Li, M Wang, X Duan¹, L Zheng, X Cheng, Y Zhang, Y Kuang, Y Li, Q Ma, Z Feng, W Liu and X Sun, *Nat. Commun.*, 2019, **10**, 1711-1721.
- [S6] A Hanan, M Lakhan, D Shu, A Hussain, M Ahmed, I Soomro, V Kumar and D Cao, *Int. J. Hydrogen Energy.*, 2023, **2**, 49-63.
- [S7] Z Zou, T Wang, X Zhao, W Jiang, H Pan, D Gao and Cailing Xu, *ACS Catal.*, 2019, **9**, 7356–7364.
- [S8] X Hou, Z Han, X Xu, D Sarker, J Zhou, M Wu, Z Liu, M Huang and H Jiang, *Chem. Eng. J.*, 2021, **418**, 129330-129339.
- [S9] F Dionigi, Z Zeng, I Sinev, T Merzdorf, S Deshpande, M Lopez, S Kunze, I Zegkinoglou, H Sarodnik, Di Fan, A Bergmann, J, Drnec, J Araujo, M Gliech, D Teschner, J Zhu, We Li, J Greeley, B Cuenya and P Strasser, *Nat. Commun.*, 2020, **11**, 2522.
- [S10] J Chen, H Li, S Chen, J Fei, C Liu, Z Yu, K Shin, Z Liu, L Song, G Henkelman, L Wei and Y Chen, *Adv. Energy Mater.*, 2021, **11**, 2003412.
- [S11] D Raja, P Cheng, C Cheng, S Chang, C Huang and S Lu, *Appl. Catal. B-environ.*, 2022, **303**, 120899.
- [S12] R Gao, H Zhang and D Yan, *Nano Energy*, 2017, **31**, 90.
- [S13] Z Guo, W Ye, X Fang, J Wan, Y Ye, Y Dong, D Cao and D Yan, *Inorg. Chem. Front.*, 2019, **6**, 687.
- [S14] R Gao and D Yan, *Nano Res*, 2018, **11**, 1883.