Electronic Supplementary Material (ESI) for Nanoscale.

Stepwise chemical oxidation to accessing ultrathin metal (oxy)-hydroxide nanosheets for oxygen evolution reaction

Jiangquan Lv,*[a,b] Xiangfeng Guan,[a,b] Yiyin Huang,*[c] Lanxin Cai,[a] Muxin Yu,[a] Xiaoyan Li,[a] Yunlong Yu,[a] and Dagui Chen[a]

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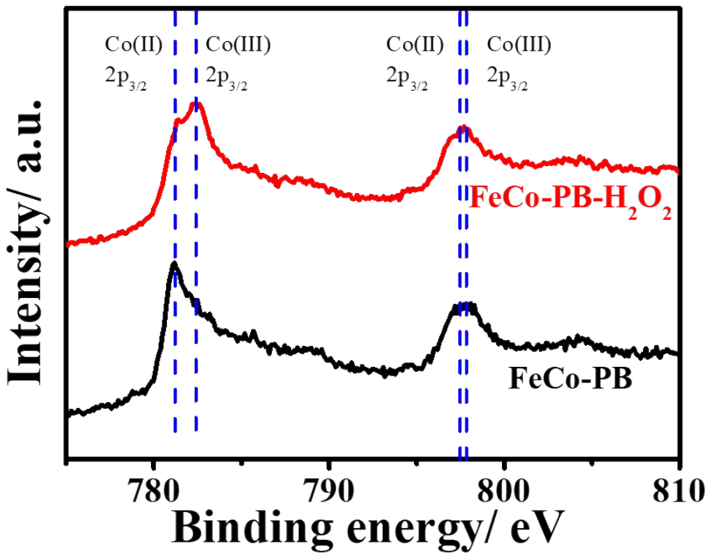

Figure S1. High-resolution Co 2p spectra of the FeCo-PB and FeCo-PB-H_2O_2.

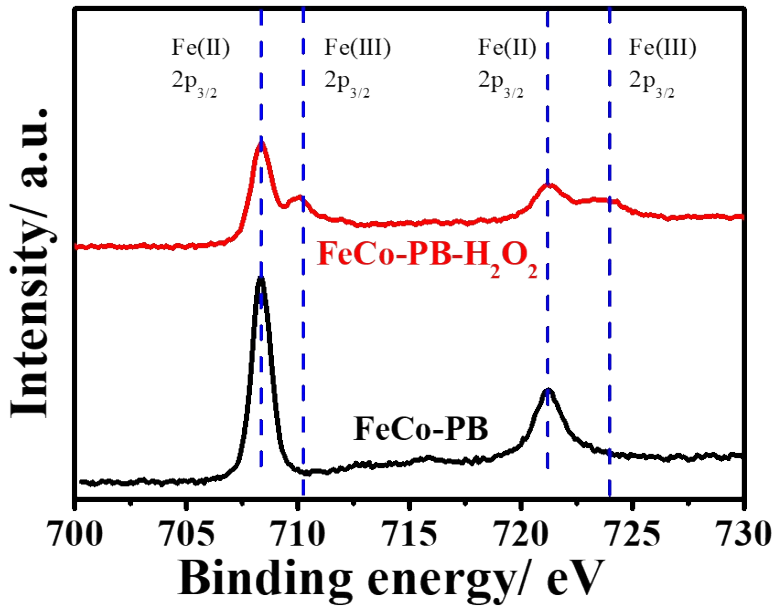

Figure S2. High-resolution Fe 2p spectra of the FeCo-PB and FeCo-PB-H_2O_2.

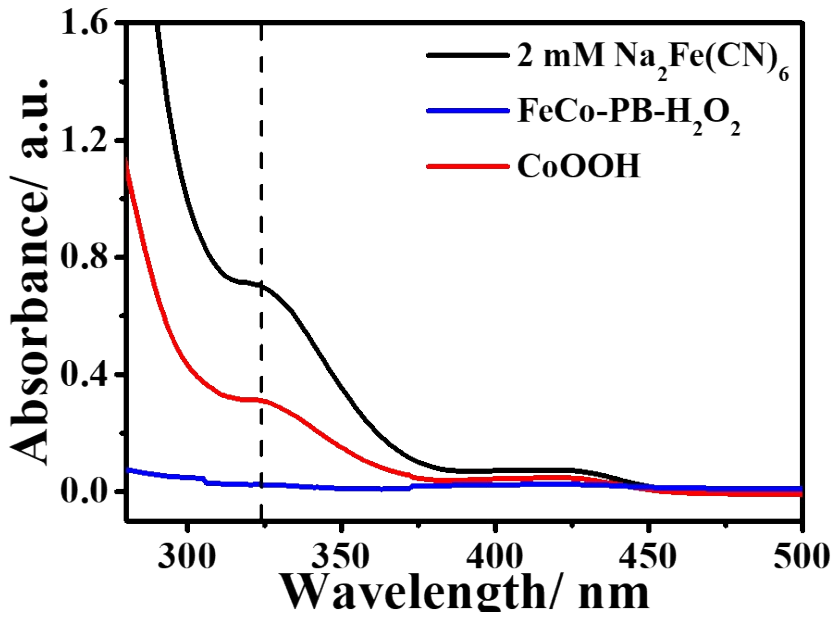

Figure S3. UV−vis absorption spectra of 2 mM $Na_2Fe(CN)_6$ and the solution after H_2O_2 oxidation (FeCo-PB-H_2O_2) and two-step oxidation (CoOOH).

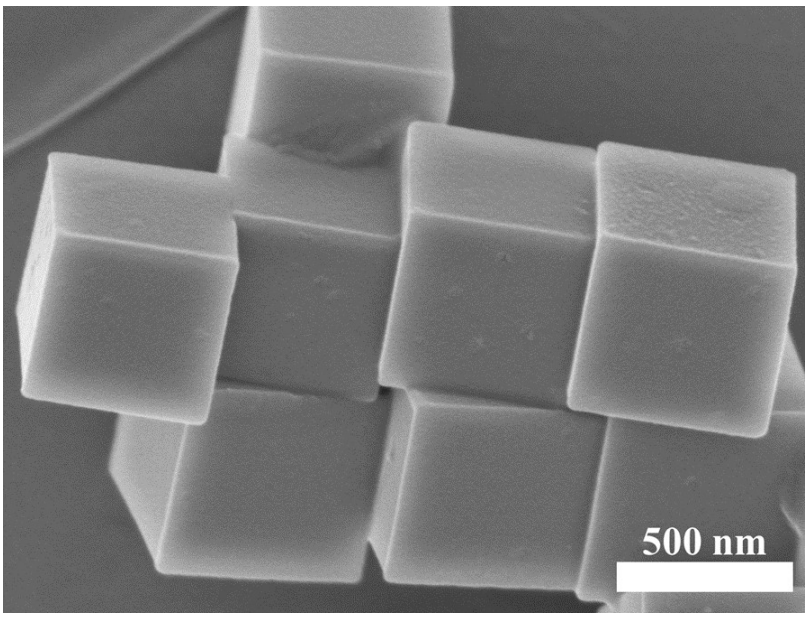

Figure S4. SEM image of FeCo-PB showed a cubic morphology with dimensions of about 550 nm.

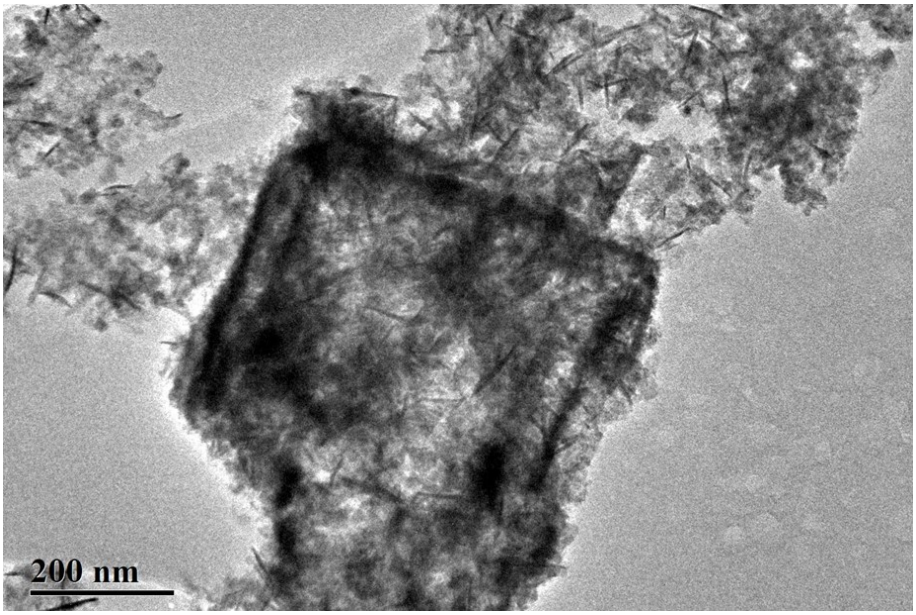

Figure S5. The TEM image of FeCo-PB-NaClO showed a deficient structure transformation.

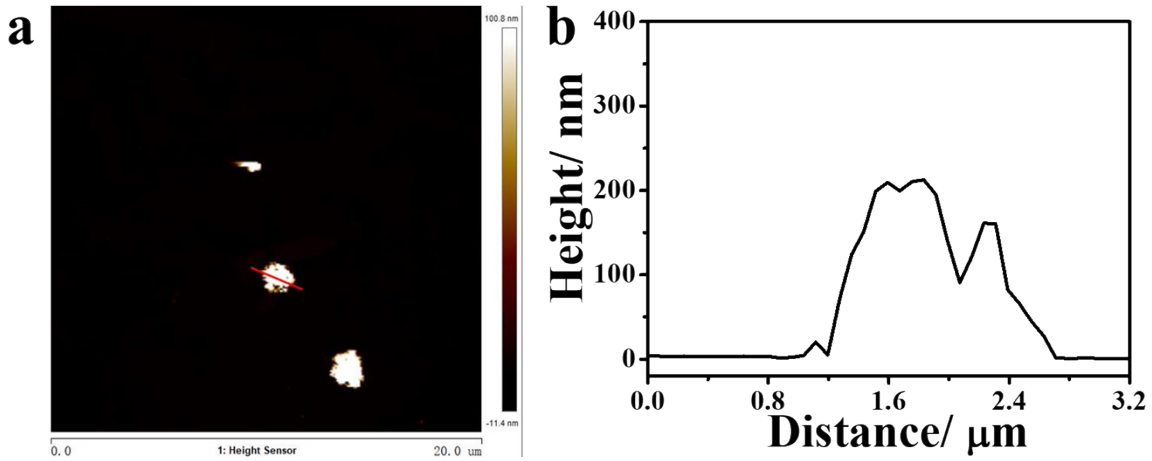

Figure S6. (a) Representative AFM image of FeCo-PB-NaClO and (b) the corresponding thickness analysis taken around the red line in (a).

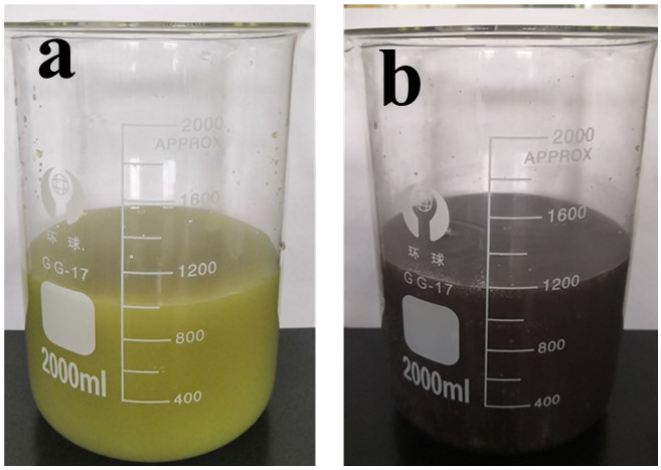

Figure S7. Digital images of (a) FeCo-PB and (b) FeCo-PB-H_2O_2.

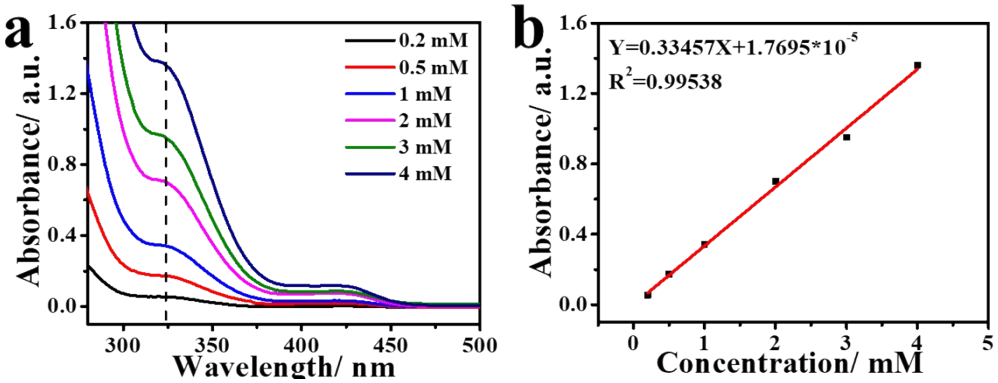

Figure S8. (a) UV−vis absorption spectra of different concentrations of $Na_2Fe(CN)_6$ (range from 0.2 mM to 4 mM); (b) Calibration curve used for calculation of $Fe(CN)_6^{2-}$ concentrations.

The absorbance of the solution after two-step oxidation was 0.31, according to the equation, the concentration was calculated to be 0.9265 mmol/L. The theoretical $Fe(CN)_6^{2-}$ concentration of 500 mg $Na_2Fe(CN)_6$ in final 1600 mL solution was 0.9864 mmol/L, so the recycling ratio of $Fe(CN)_6^{2-}$ was calculated to be 93.8%.

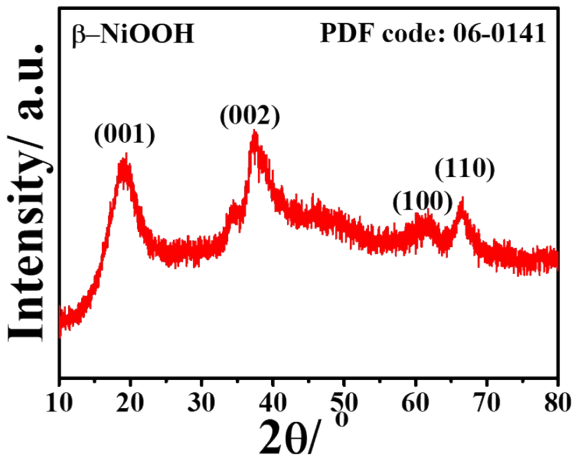

Figure S9. XRD pattern of β-NiOOH, which synthesized by two-step oxidation procedure of $Na_xNiFe(CN)_6$.

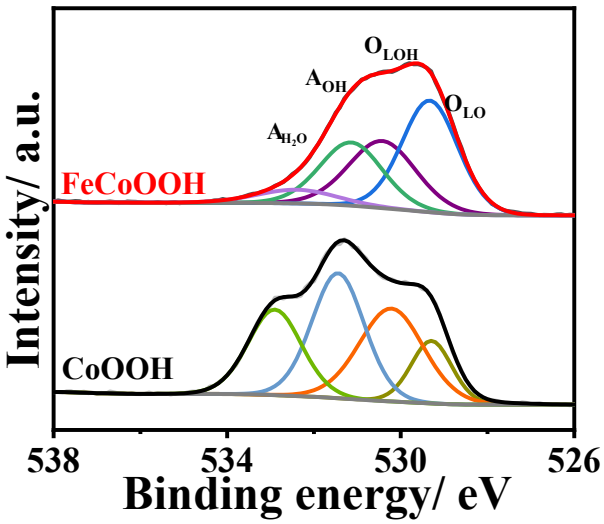

Figure S10. High-resolution O 1s spectra of the FeCoOOH nanosheets and CoOOH nanosheets;

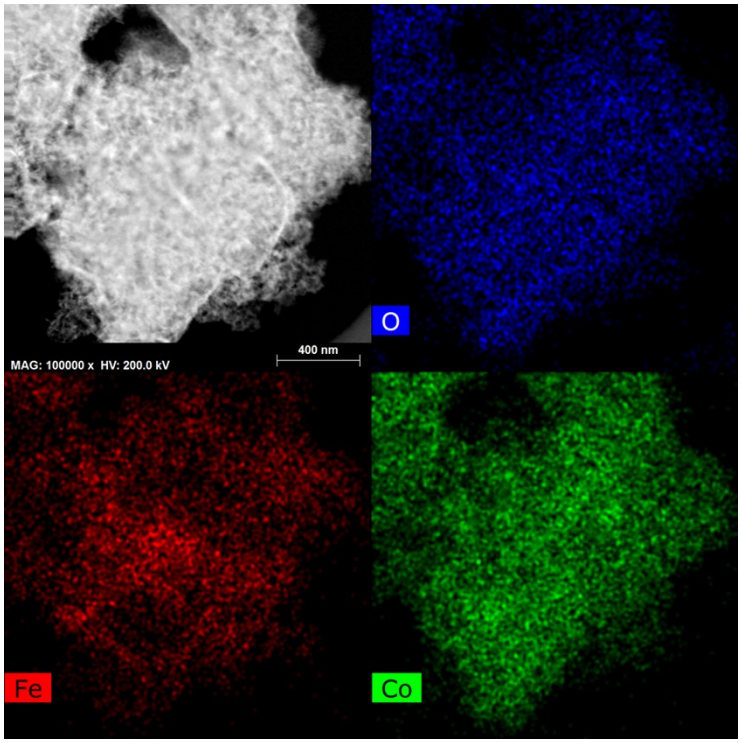

Figure S11. Element mapping of FeCoOOH nanosheets with O, Fe and Co.

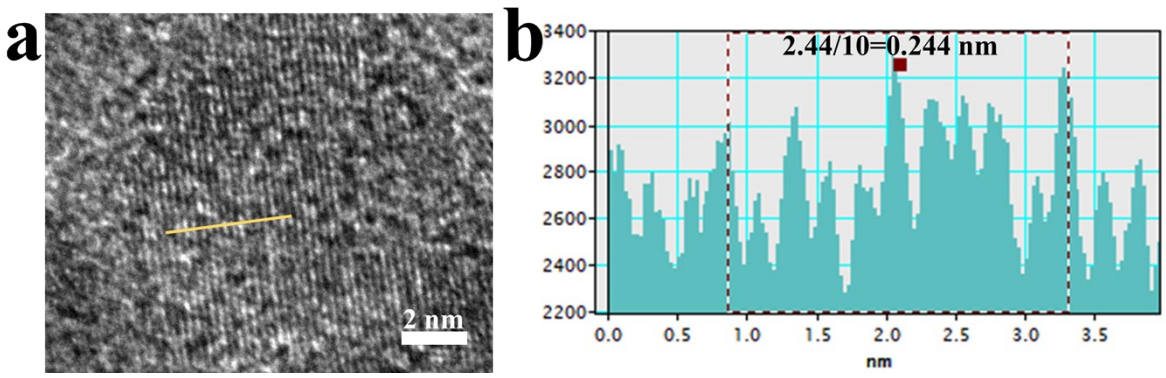

Figure S12. (a) HRTEM image of FeCoOOH nanosheets; (b) the profile of HRTEM for the calculation of lattice fringes.

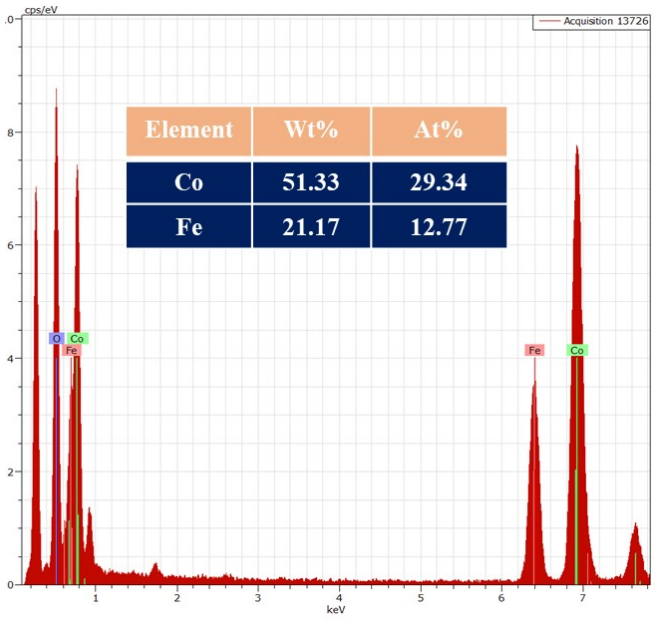

Figure S13. EDS result of FeCoOOH.

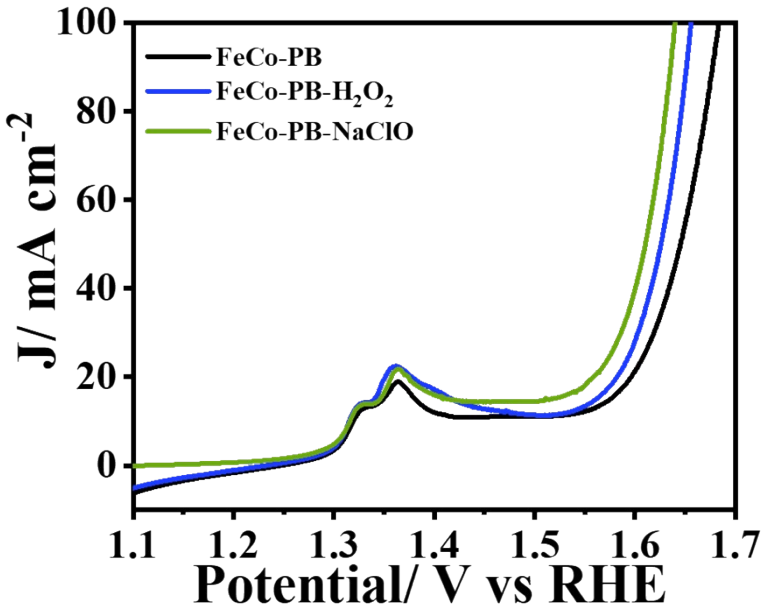

Figure S14. LSV plots of FeCo-PB, FeCo-PB-H_2O_2 and FeCo-PB-NaClO.

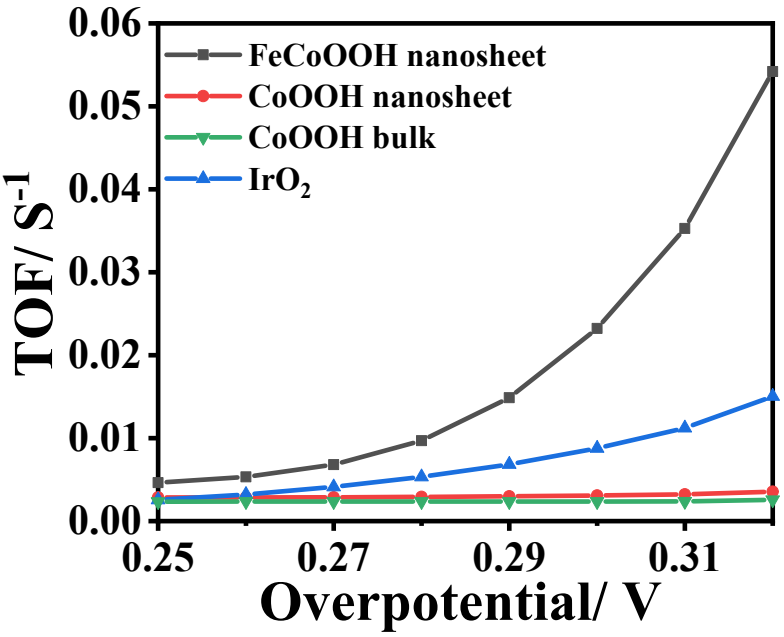

Figure S15. TOF plots of FeCoOOH nanosheet, CoOOH nanosheet, CoOOH bulk and IrO_2 at different overpotentials.

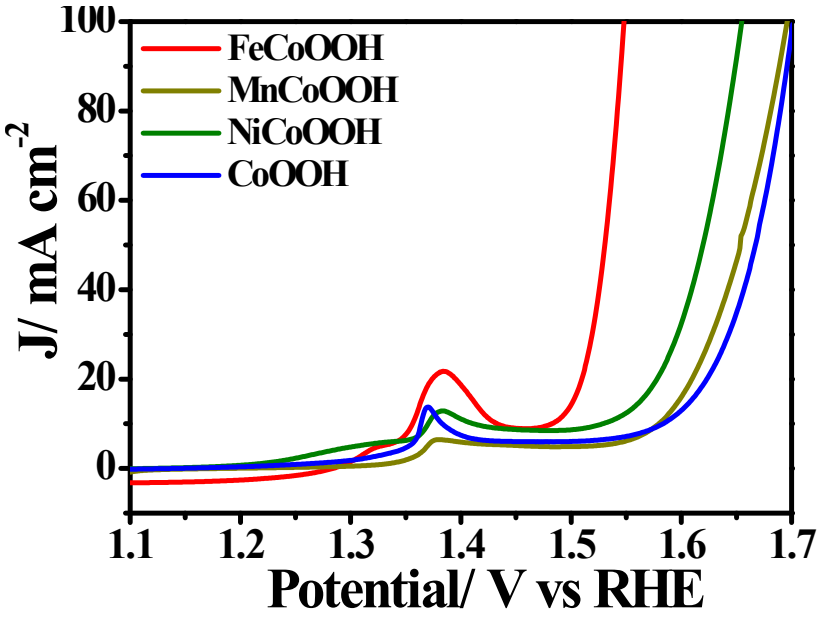

Figure S16. LSV plots of FeCoOOH, MnCoOOH, NiCoOOH and CoOOH.

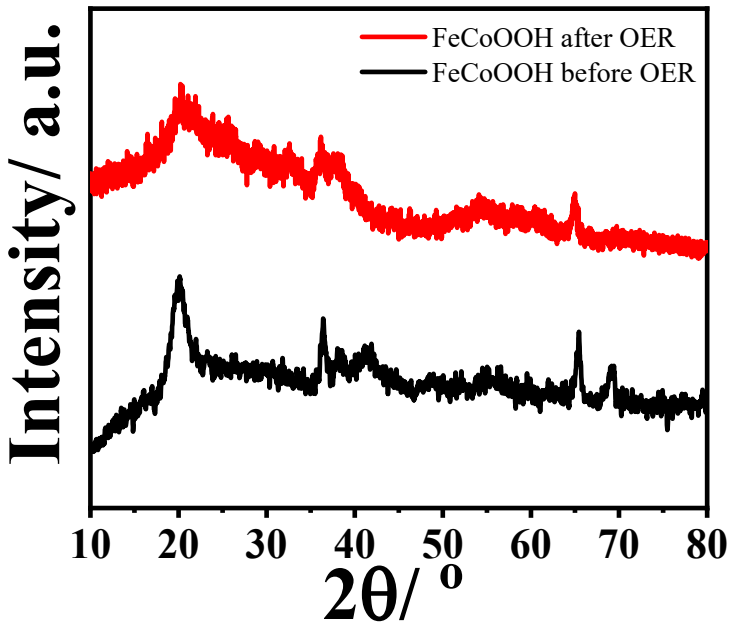

Figure S17. XRD patterns of FeCoOOH nanosheets before and after OER stability test.

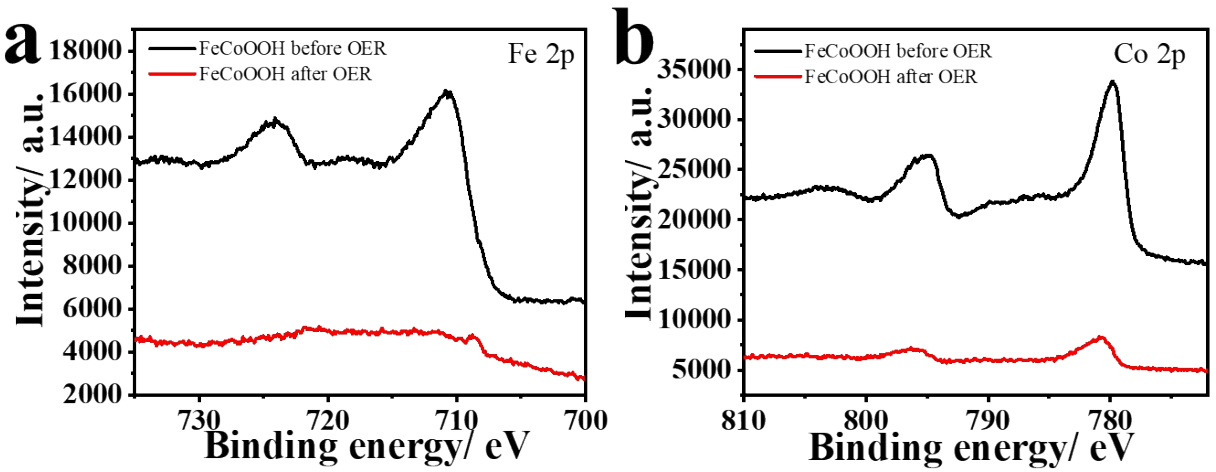

Figure S18. (a) Co 2p XPS spectra and (b) Fe 2p XPS spectra for FeCoOOH nanosheets before and after OER stability test.

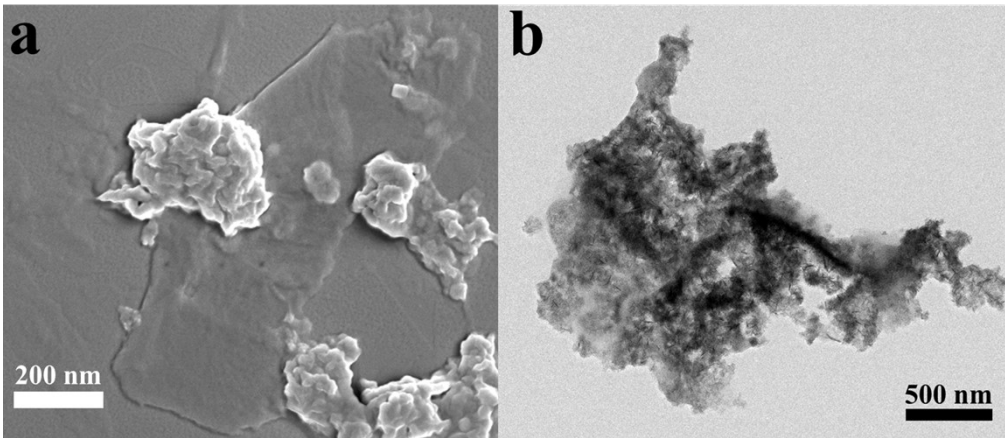

Figure S19. (a) SEM and (b) TEM images for FeCoOOH nanosheets after OER stability test.

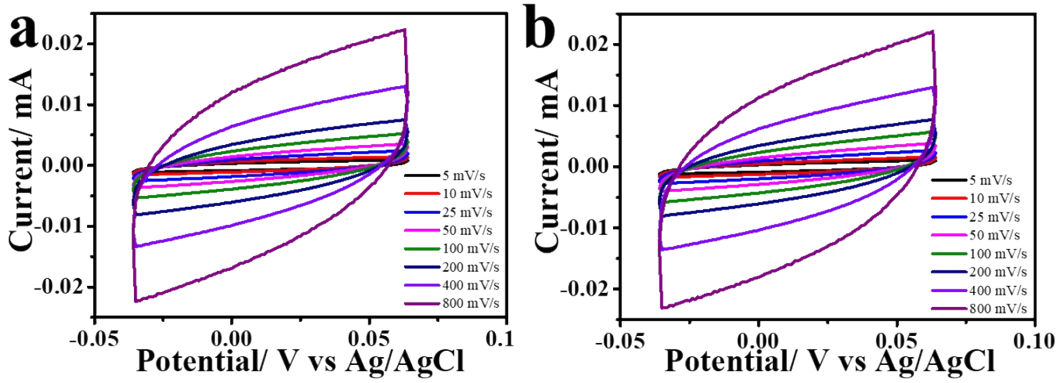

Figure S20. CV curves of (a) CoOOH and (b) FeCoOOH at scan rates ranging from 5 mV s^{-1} to 800 mV s^{-1}.

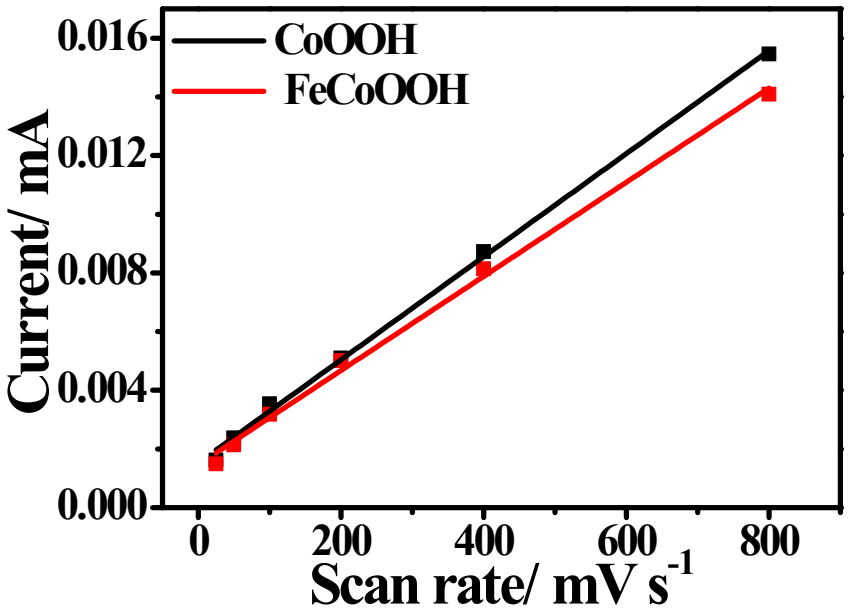

Figure S21. Calculation of C_{dl} for CoOOH and FeCoOOH by plotting the capacitive current density against the scan rate to fit a linear regression.

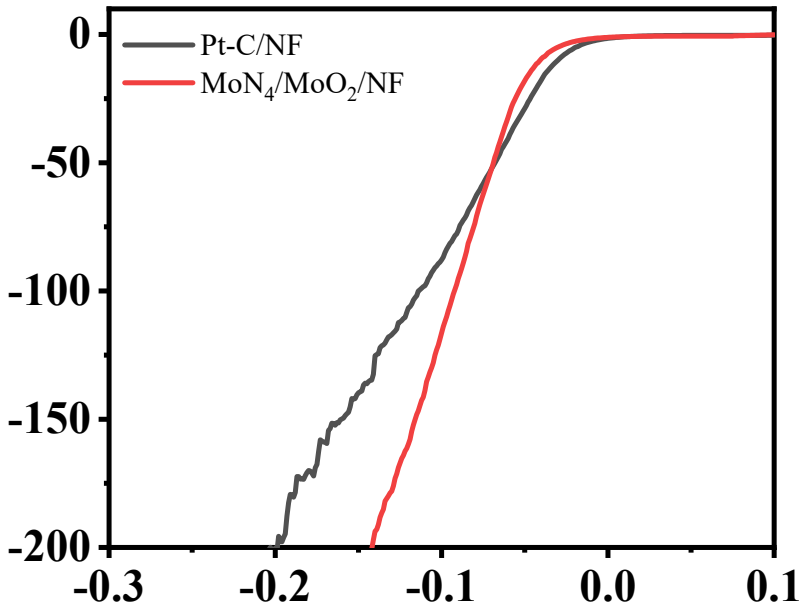

Figure S22. LSV polarization curve of $MoNi_4/MoO_2$/NF and Pt-C/NF for HER with 90% iR-compensation.

2. Supplementary Table.

Table S1. Comparison of OER activities of different cobalt-based materials.

Catalyst	Electrolyte	Overpotential at 10 mA cm^{-2} (mV)	Substrate	Reference
FeCoOOH	1 M KOH	252	Ni foam	This work
Fe/CoOOH	1 M KOH	266	Carbon cloth	S1
Co-PBA-plasma 2 h	1 M KOH	274	Ni foam	S2
FeCo-MOF-EH	1 M KOH	231	Ni foam	S3
γ-CoOOH nanosheets	1 M KOH	275	Ni foam	S4
Co_2AlO_4 nanosheets	1 M KOH	280	Carbon paper	S5
Co_3O_4-CoO nanosheets	1 M KOH	270	Carbon cloth	S6
nPBA@$Co(OH)_2$/NF	1 M KOH	258	Ni foam	S7
F-CoOOH/NF	1 M KOH	270	Ni foam	S8
$Co(OH)_2$/NCNTs-NF	1 M KOH	270	Ni foam	S9
$CoCO_3$@CoSe Nanowires	1 M KOH	255	Ni foam	S10
CoS	1 M KOH	297	Ni foam	S11

Table S2. Comparison of overall water splitting performance in recent reports.

Catalyst	Electrolyte	Potential at 10 mA cm^{-2} (V)	Substrate	Reference
FeCoOOH	1 M KOH	1.50	Ni foam	This work
NiO–Ni_3S_2/NF	1 M KOH	1.57	Ni foam	S12
2D/3D ZIF-67@CC	1 M KOH	1.545	Carbon cloth	S13
Co_3S_4/EC-MOF	1 M KOH	1.55	carbon cloth	S14
NC/CuCo/$CuCoO_x$	1 M KOH	1.53	Ni foam	S15
1T-MoS_2	1 M KOH	1.57	Glassy carbon	S16
Am $FePO_4$/NF	1 M KOH	1.54	Ni foam	S17

Supplementary References

S1. S.-H. Ye, Z.-X. Shi, J.-X. Feng, Y.-X. Tong and G.-R. Li, *Angewandte Chemie-International Edition*, 2018, **57**, 2672-2676.

S2. Y. Guo, T. Wang, J. Chen, J. Zheng, X. Li and K. Ostrikov, *Adv. Energy Mater.*, 2018, **8**, 1800085.

S3. J. Tian, F. Jiang, D. Yuan, L. Zhang, Q. Chen and M. Hong, *Angewandte Chemie-International Edition*, 2020, **59**, 13101-13108.

S4. H. Wang, E.-m. Feng, Y.-m. Liu and C.-y. Zhang, *J. Mater. Chem. A*, 2019, **7**, 7777-7783.

S5. J. Wang, Y. Shen, G. Wei, W. Xi, X. Ma, W. Zhang, P. Zhu and C. An, *Science China Materials*, 2020, **63**, 91-99.

S6. J. Qi, Y. Yan, T. Liu, X. Zhou, J. Cao and J. Feng, *Journal of Colloid and Interface Science*, 2020, **565**, 400-404.

S7. Y. Wang, J. Ma, J. Wang, S. Chen, H. Wang and J. Zhang, *Adv. Energy Mater.*, 2019, **9**, 1802939.

S8. P. Chen, T. Zhou, S. Wang, N. Zhang, Y. Tong, H. Ju, W. Chu, C. Wu and Y. Xie, *Angewandte Chemie-International Edition*, 2018, **57**, 15471-15475.

S9. P. Guo, J. Wu, X.-B. Li, J. Luo, W.-M. Lau, H. Liu, X.-L. Sun and L.-M. Liu, *Nano Energy*, 2018, **47**, 96-104.

S10. R. Que, G. Ji, D. Liu, M. Li, X. Wang and S. P. Jiang, *Energy Technology*, 2019, **7**, 1800741.

S11. P. Guo, Y.-X. Wu, W.-M. Lau, H. Liu and L.-M. Liu, *Journal of Alloys and Compounds*, 2017, **723**, 772-778.

S12. L. Peng, J. Shen, X. Zheng, R. Xiang, M. Deng, Z. Mao, Z. Feng, L. Zhang, L. Li and Z. Wei, *Journal Of Catalysis*, 2019, **369**, 345-351.

S13. Z. Chen, Y. Ha, H. Jia, X. Yan, M. Chen, M. Liu and R. Wu, *Adv. Energy Mater.*, 2019, **9**, 1803918.

S14. T. Liu, P. Li, N. Yao, T. Kong, G. Cheng, S. Chen and W. Luo, *Adv. Mater.*, 2019, **31**, 1806672.

S15. J. Hou, Y. Sun, Y. Wu, S. Cao and L. Sun, *Adv. Funct. Mater.*, 2018, **28**, 1704447.

S16. S. Wei, X. Cui, Y. Xu, B. Shang, Q. Zhang, L. Gu, X. Fan, L. Zheng, C. Hou, H. Huang, S. Wen and W. Zheng, *Acs Energy Letters*, 2019, **4**, 368-374.

S17. L. Yang, Z. Guo, J. Huang, Y. Xi, R. Gao, G. Su, W. Wang, L. Cao and B. Dong, *Adv. Mater.*, 2017, **29**, 1704574.