

## Supporting Information

### 2D Iron/Cobalt Metal-Organic Frameworks with an Extended Ligand for Efficient Oxygen Evolution Reaction

Wenjing Shang, Qiulin Li, Xiang Li, Ke Zhang, Binghao Wang, Yongbing Lou, Jinxi Chen\*

*School of Chemistry and Chemical Engineering, Jiangsu Engineering Laboratory of Smart Carbon-Rich Materials and Device, Southeast University, Nanjing 211189, PR China*

\*Corresponding author

\*E-mail address: chenjinxi@seu.edu.cn (J. Chen)

#### Calculation formulas

Some data needed to be calculated in the experimental process, and the calculation formulas were as follows:

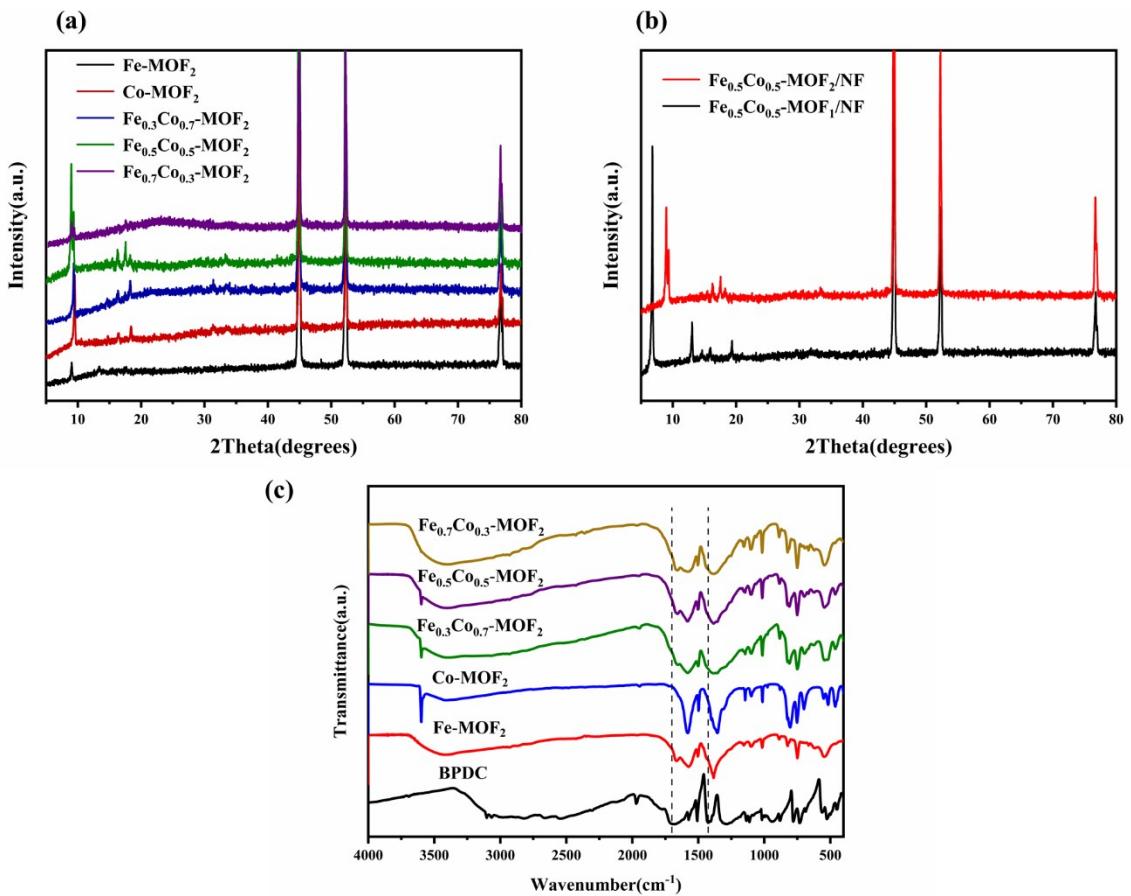
Overpotential:  $\eta = E_{\text{RHE}} - 1.23 \text{ V}$ ,

where  $E_{\text{RHE}}$  referred to reversible hydrogen electrode,  $E_{\text{RHE}} = E_{\text{Ag}/\text{AgCl}} + 0.059 * \text{pH} + 0.197 \text{ V}$

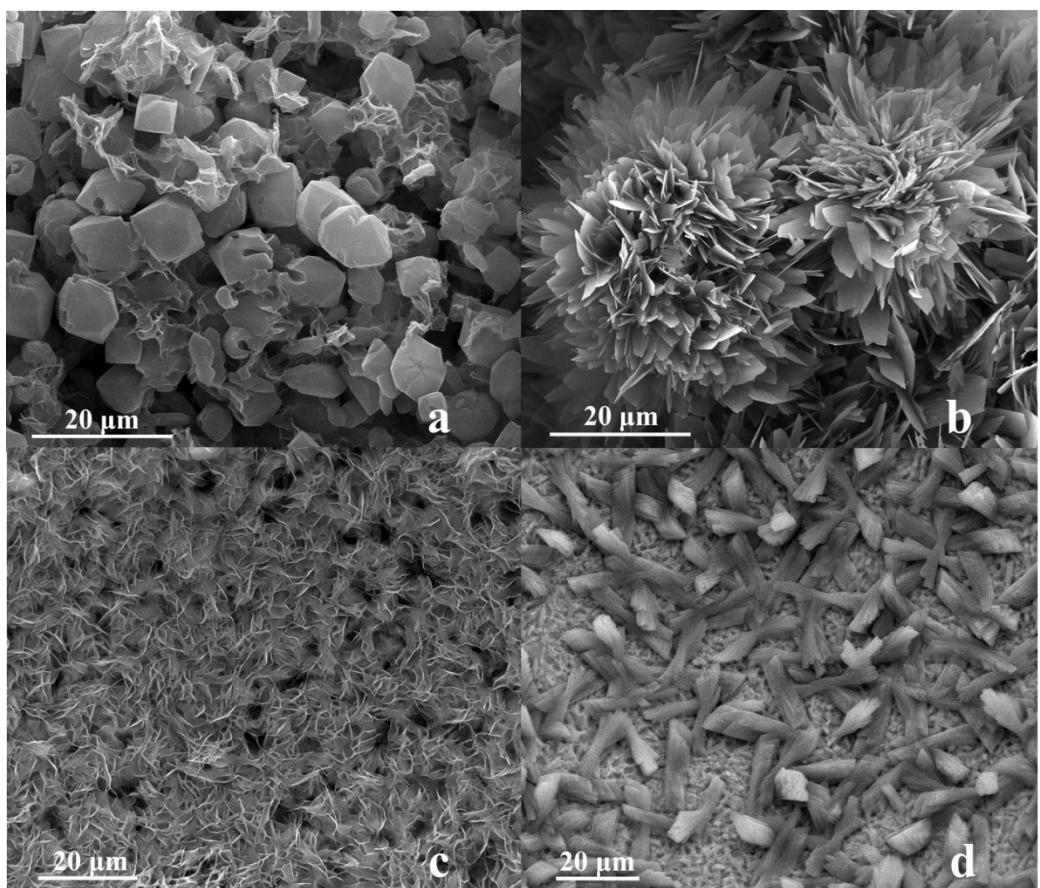
Tafel slope:  $\eta = b \log j + a$ ,

where  $b$  was the Tafel slope. It was derived from the LSV curve,  $\log j$  ( $j$  was the current density) as the abscissa and  $\eta$  as the ordinate, and the resulting slope was called the Tafel slope.

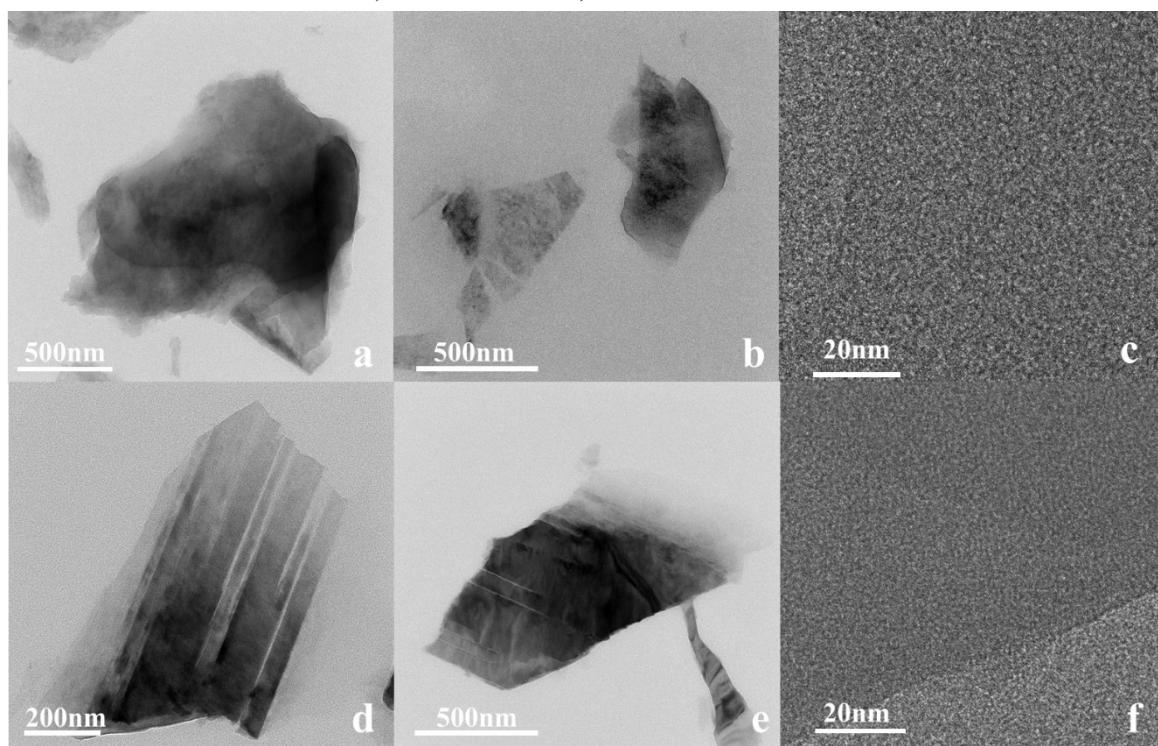
$2C_{\text{dl}}$  is estimated by plotting  $\Delta J = (J_a - J_c)$  at 0.8794 V against the scan rates.



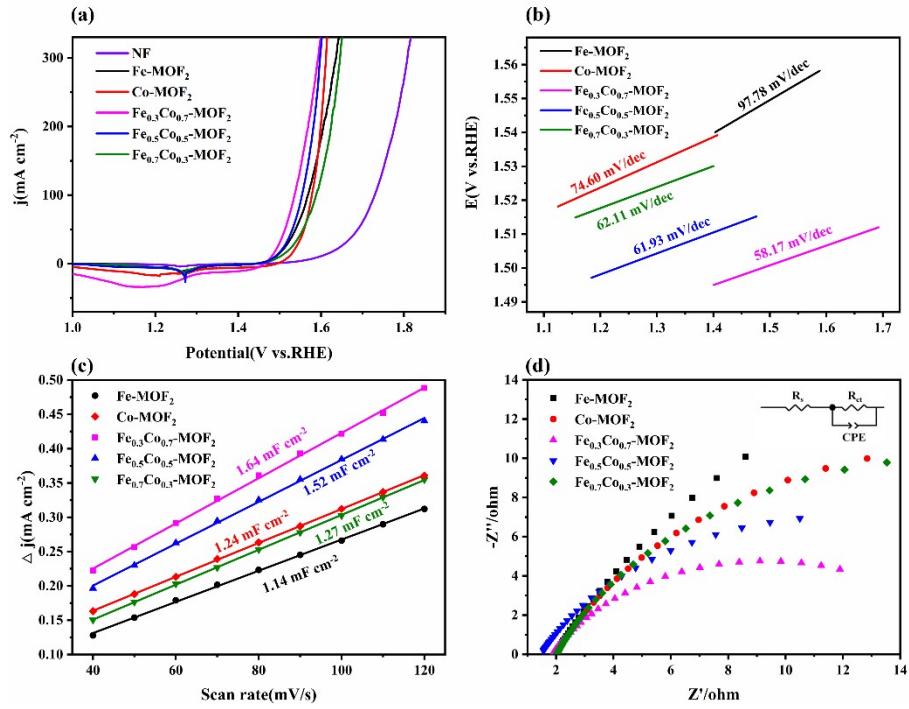
**Figure S1.** a) XRD patterns of  $\text{Fe}_x\text{Co}_{1-x}\text{-MOF}_2$ , b)  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1$  and  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_2$ , c) FT-IR spectra of  $\text{Fe}_x\text{Co}_{1-x}\text{-MOF}_2$



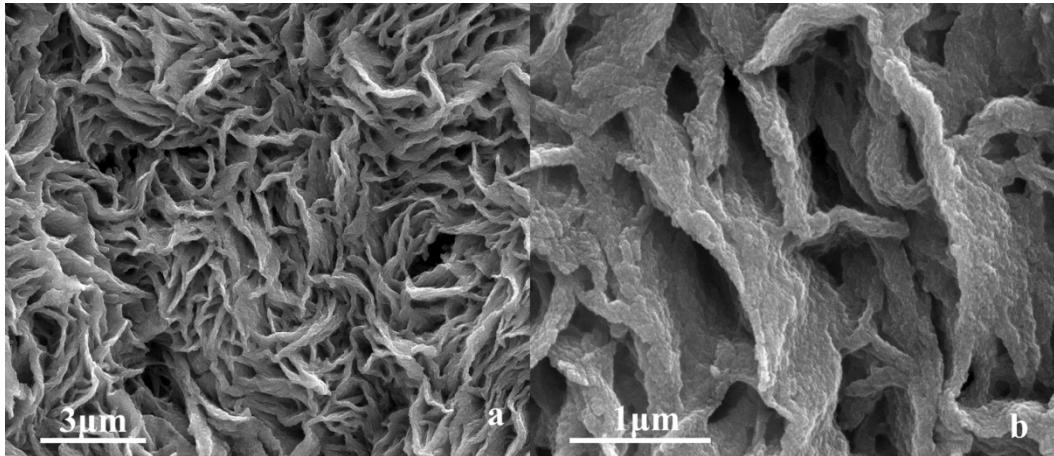
**Figure S2.** a) SEM images of Fe-MOF<sub>1</sub>/NF, b) Co-MOF<sub>1</sub>/NF,  
c) Fe-MOF<sub>2</sub>/NF, d) Co-MOF<sub>2</sub>/NF



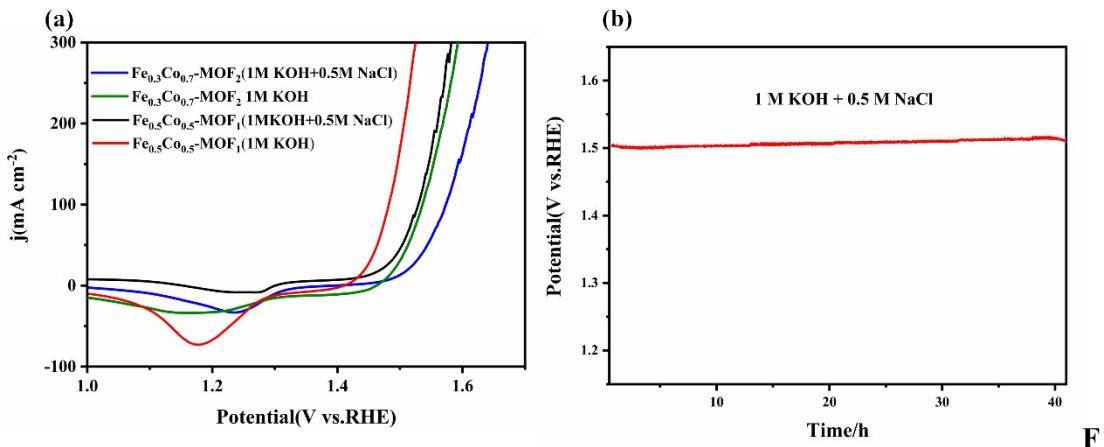
**Figure S3.** a,b) TEM and c) HRTEM image of Fe<sub>0.5</sub>Co<sub>0.5</sub>-MOF<sub>1</sub>/NF before the OER test,  
d,e) TEM and f) HRTEM image of Fe<sub>0.3</sub>Co<sub>0.7</sub>-MOF<sub>2</sub>/NF before the OER test



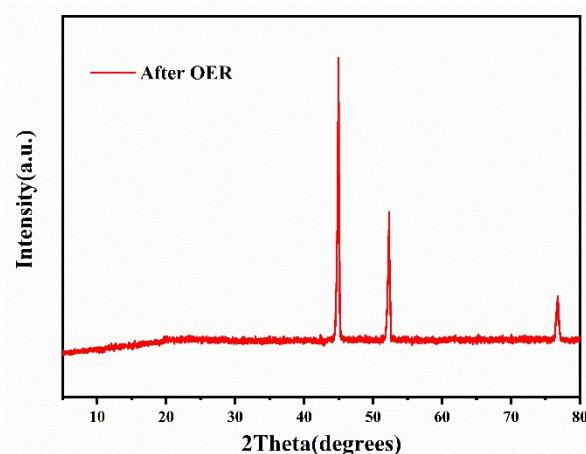
**Figure S4.** a) LSV curves b) Tafel plots c)  $C_{\text{dl}}$  curves and d) electrochemical impedance of  $\text{Fe}_x\text{Co}_{1-x}\text{-MOF}_2/\text{NF}$



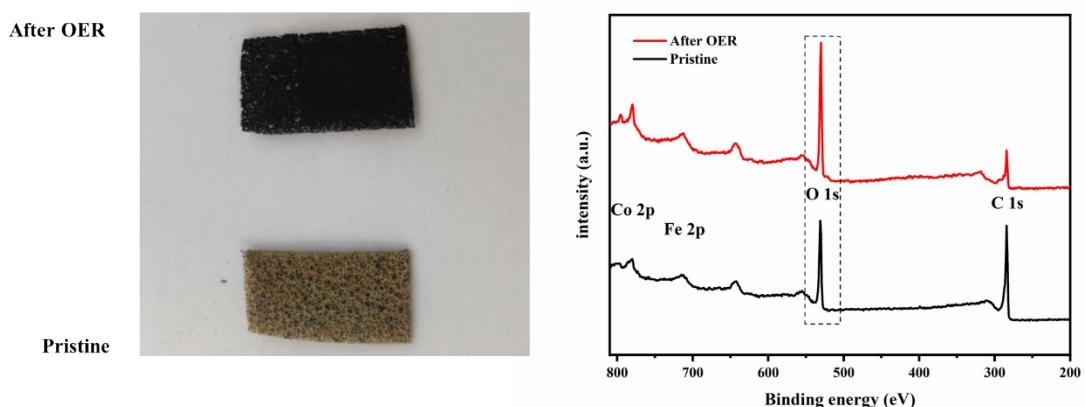
**Figure S5.** SEM  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  after the 30 h stability test in 1 M KOH



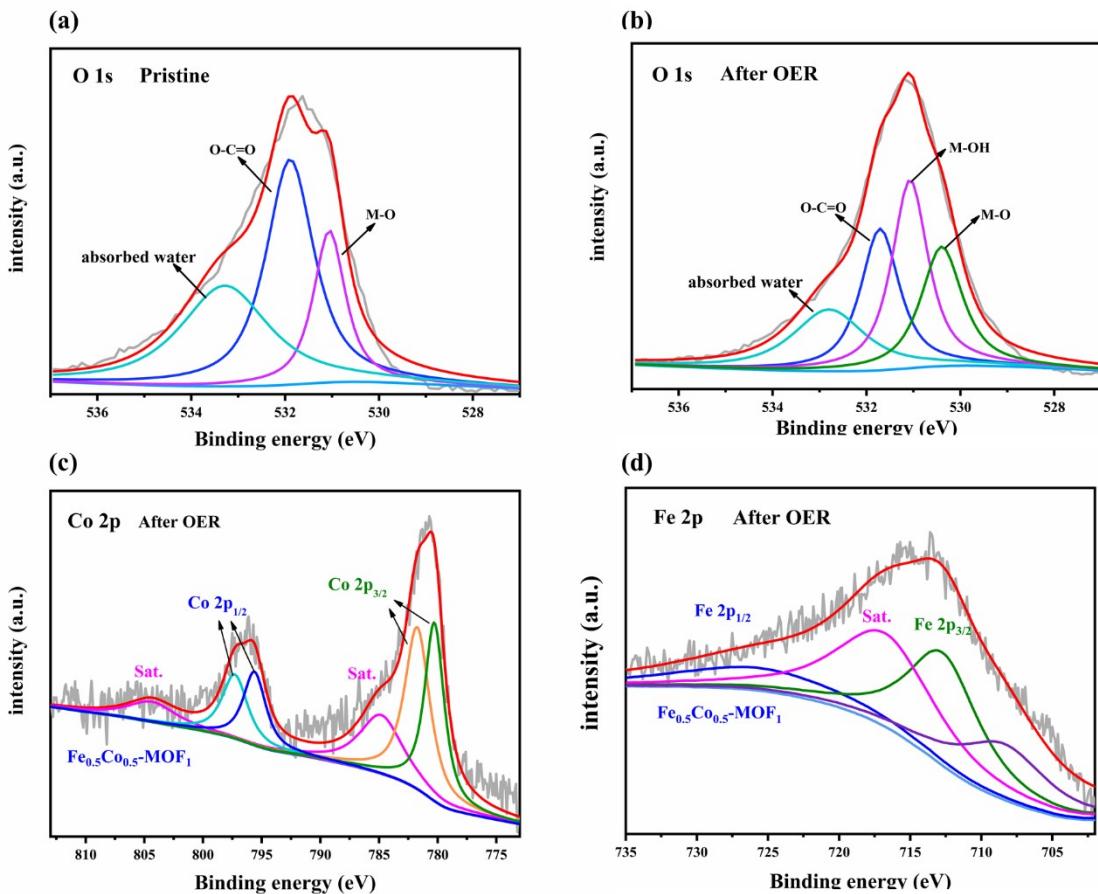
**figure S6.** a) LSV curves in 1 M KOH and 1 M KOH+0.5 M NaCl of  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  and  $\text{Fe}_{0.3}\text{Co}_{0.7}\text{-MOF}_2$ , b) Chronopotentiometric curve of  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  in 1 M KOH+0.5 M NaCl



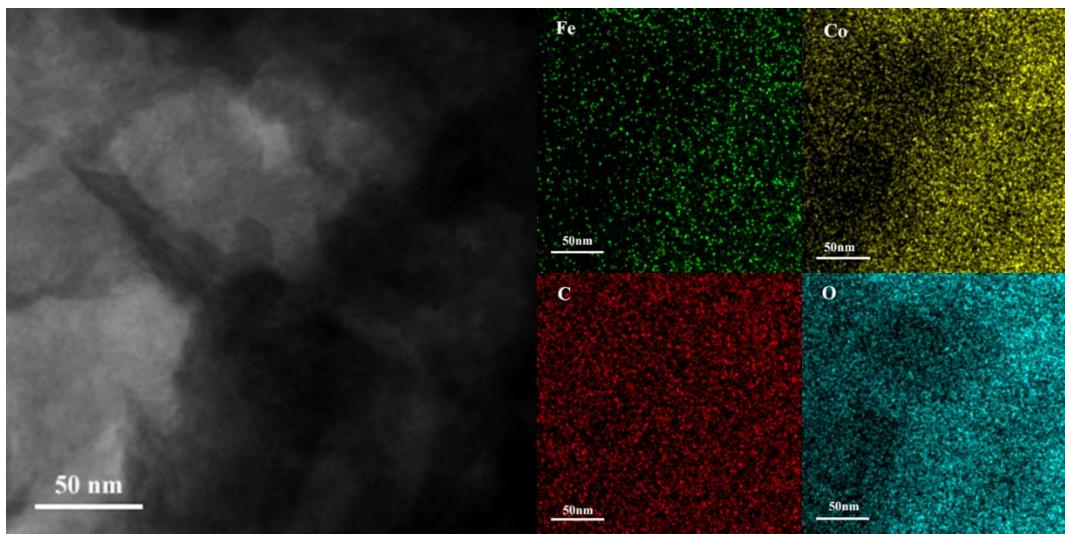
**Figure S7.** XRD patterns  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  after the 30 h stability test in 1 M KOH



**Figure S8.** a) Photos, b) survey spectrum of pristine  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  and  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  after OER test



**Figure S9.** The high-resolution XPS spectra of a) O 1s in pristine  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  b) O 1s c) Co 2p d) Fe 2p in  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  after OER test



**Figure S10.** TEM image and corresponding EDS elemental mapping images of  $\text{Fe}_{0.5}\text{Co}_{0.5}\text{-MOF}_1/\text{NF}$  after OER test

**Table S1.** The amount of metal ions and ligands used to synthesize MOF<sub>1</sub>

Catalyst	Fe(NO <sub>3</sub> ) <sub>3</sub> ·9H <sub>2</sub> O	Co(NO <sub>3</sub> ) <sub>2</sub> ·6H <sub>2</sub> O	BPDC
Fe-MOF <sub>1</sub> /NF	404.0 mg (1 mmol)	0 mg	242.2 mg (1 mmol)
Fe <sub>0.3</sub> Co <sub>0.7</sub> -MOF <sub>1</sub> /NF	121.2 mg (0.3 mmol)	203.7 mg (0.7 mmol)	242.2 mg (1 mmol)
Fe <sub>0.5</sub> Co <sub>0.5</sub> -MOF <sub>1</sub> /NF	202.0 mg (0.5 mmol)	145.5 mg (0.5 mmol)	242.2 mg (1 mmol)
Fe <sub>0.7</sub> Co <sub>0.3</sub> -MOF <sub>1</sub> /NF	282.8 mg (0.7 mmol)	87.3 mg (0.3 mmol)	242.2 mg (1 mmol)
Co-MOF <sub>1</sub> /NF	0 mg	291.0 mg (1 mmol)	242.2 mg (1 mmol)

**Table S2.** The amount of metal ions and ligands used to synthesize MOF<sub>2</sub>

Catalyst	Fe(NO <sub>3</sub> ) <sub>3</sub> ·9H <sub>2</sub> O	Co(NO <sub>3</sub> ) <sub>2</sub> ·6H <sub>2</sub> O	BDC
Fe-MOF <sub>2</sub> /NF	404.0 mg (1 mmol)	0 mg	166.1 mg (1 mmol)
Fe <sub>0.3</sub> Co <sub>0.7</sub> -MOF <sub>2</sub> /NF	121.2 mg (0.3 mmol)	203.7 mg (0.7 mmol)	166.1 mg (1 mmol)
Fe <sub>0.5</sub> Co <sub>0.5</sub> -MOF <sub>2</sub> /NF	202.0 mg (0.5 mmol)	145.5 mg (0.5 mmol)	166.1 mg (1 mmol)
Fe <sub>0.7</sub> Co <sub>0.3</sub> -MOF <sub>2</sub> /NF	282.8 mg (0.7 mmol)	87.3 mg (0.3 mmol)	166.1 mg (1 mmol)
Co-MOF <sub>2</sub> /NF	0 mg	291.0 mg (1 mmol)	166.1 mg (1 mmol)

**Table S3.** Comparisons of electrochemical performance and the mass loading of electrodes

Catalyst	Overpotential at 10 mA cm <sup>-2</sup> (mV)	Tafel slope (mV/dec)	C <sub>dl</sub> (mF cm <sup>-2</sup> )	R <sub>ct</sub> (Ω)	Mass loading (mg cm <sup>-2</sup> )
Fe-MOF <sub>1</sub> /NF	280	86.33	1.13	22.32	3.7
Fe <sub>0.3</sub> Co <sub>0.7</sub> -MOF <sub>1</sub> /NF	252	41.21	2.17	17.12	4.3
Fe <sub>0.5</sub> Co <sub>0.5</sub> -MOF <sub>1</sub> /NF	217	31.16	2.40	4.19	4.1
Fe <sub>0.7</sub> Co <sub>0.3</sub> -MOF <sub>1</sub> /NF	254	51.90	1.86	2.68	4.0
Co-MOF <sub>1</sub> /NF	270	74.38	1.24	6.62	4.7
Fe-MOF <sub>2</sub> /NF	272	97.78	1.14	60.95	4.2
Fe <sub>0.3</sub> Co <sub>0.7</sub> -MOF <sub>2</sub> /NF	249	58.17	1.64	14.91	4.4
Fe <sub>0.5</sub> Co <sub>0.5</sub> -MOF <sub>2</sub> /NF	269	61.93	1.52	21.34	4.5
Fe <sub>0.7</sub> Co <sub>0.3</sub> -MOF <sub>2</sub> /NF	258	62.11	1.27	29.04	4.2
Co-MOF <sub>2</sub> /NF	284	74.60	1.24	31.21	3.8

**Table S4.** Comparison of OER catalytic performances of various MOF-based electrocatalysts

Catalysts	Overpotential (mV@ mA/cm <sup>2</sup> )	Tafel slope (mV/dec)	Ligand	Reference
<b>Fe<sub>0.5</sub>Co<sub>0.5</sub>-MOF<sub>1</sub>/NF</b>	<b>217@10</b>	<b>31.16</b>	<b>BPDC</b>	<b>This work</b>
2D MOF-Fe/Co(1:2)	238@10	52	1,4-BDC	<sup>1</sup>
MIL-53(Co-Fe)/NF	262@100	69	TPA	<sup>2</sup>
Fe <sub>2</sub> Co-MOF	224@10	45.3	TPA	<sup>3</sup>
Ni-Fe-MOF NSs	221@10	56	1,4-BDC	<sup>4</sup>
(Fe,Co)OOH/MI	230@10	53	MI	<sup>5</sup>
BaTiO <sub>3</sub> @MOF-Fe/Co	247@10	38.4	1,4-BDC	<sup>6</sup>
CoFe-MOF-OH	265@10	44	C <sub>5</sub> H <sub>4</sub> N <sub>2</sub> O <sub>4</sub>	<sup>7</sup>
Co <sub>3</sub> Fe-MOF	280@10	38	NH <sub>2</sub> -BDC	<sup>8</sup>
Au <sub>5,30</sub> /(FCN)MOF/NP	216@10	31.7	1,4-BDC	<sup>9</sup>
Fe-Co-O/Co@				
NC-mNS/NF	257@10	41.56	MI	<sup>10</sup>
Co <sub>2</sub> Fe-MOF	280@10	44.7	H <sub>3</sub> BTC	<sup>11</sup>
CF-PBA-400	254@10	51	PBA	<sup>12</sup>
CoFeBiP	273@10	77.3	MI	<sup>13</sup>
NiFc-MOF/NF	195@10	44.1	FcDA	<sup>14</sup>
Ni <sub>2</sub> Fe <sub>1</sub> Sq-zbr-MOF	230@10	37	C <sub>4</sub> H <sub>2</sub> O <sub>4</sub>	<sup>15</sup>

TPA, 1, 4-BDC: 1, 4-bezenedicarboxylate

MI: 2-Methylimidazole

C<sub>5</sub>H<sub>4</sub>N<sub>2</sub>O<sub>4</sub>: 4, 5-Imidazoledicarboxylic acidNH<sub>2</sub>-BDC: 2-Aminoterephthalic acidH<sub>3</sub>BTC: Trimesic acid

PBA: Prussian blue analogue

FcDA: 1, 1'-Ferrocene dicarboxylate

C<sub>4</sub>H<sub>2</sub>O<sub>4</sub>: 3, 4-Dihydroxy-3-cyclobutene-1, 2-dione (squaric acid)

## References

- 1 K. Ge, S. Sun, Y. Zhao, K. Yang, S. Wang, Z. Zhang, J. Cao, Y. Yang, Y. Zhang, M. Pan and L. Zhu, *Angew. Chem., Int. Ed.*, 2021, **60**, 12097-12102.
- 2 M. Xie, Y. Ma, D. Lin, C. Xu, F. Xie and W. Zeng, *Nanoscale*, 2020, **12**, 67-71.
- 3 X. Ling, F. Du, Y. Zhang, Y. Shen, W. Gao, B. Zhou, Z. Wang, G. Li, T. Li, Q. Shen, Y. Xiong, X. Wang, Y. Zhou and Z. Zou, *J. Mater. Chem. A*, 2021, **9**, 13271-13278.
- 4 F. L. Li, P. Wang, X. Huang, D. J. Young, H. F. Wang, P. Braunstein and J. P. Lang, *Angew. Chem., Int. Ed.*, 2019, **58**, 7051-7056.

- 5 W. Huang, J. Li, X. Liao, R. Lu, C. Ling, X. Liu, J. Meng, L. Qu, M. Lin, X. Hong, X. Zhou, S. Liu, Y. Zhao, L. Zhou and L. Mai, *Adv. Mater.*, 2022, **34**, 2200270.
- 6 S. Wang, Q. Li, S. Sun, K. Ge, Y. Zhao, K. Yang, Z. Zhang, J. Cao, J. Lu, Y. Yang, Y. Zhang, M. Pan, Z. Lin and L. Zhu, *J. Mater. Chem. A*, 2022, **10**, 5350-5360.
- 7 Z. Zou, T. Wang, X. Zhao, W.-J. Jiang, H. Pan, D. Gao and C. Xu, *ACS Catal.*, 2019, **9**, 7356-7364.
- 8 W. Li, W. Fang, C. Wu, K. N. Dinh, H. Ren, L. Zhao, C. Liu and Q. Yan, *J. Mater. Chem. A*, 2020, **8**, 3658-3666.
- 9 C.-C. Cheng, P.-Y. Cheng, C.-L. Huang, D. Senthil Raja, Y.-J. Wu and S.-Y. Lu, *Appl. Catal. B: Environ.*, 2021, **286**.
- 10 T. I. Singh, G. Rajeshkhanna, U. N. Pan, T. Kshetri, H. Lin, N. H. Kim and J. H. Lee, *Small*, 2021, **17**, 2101312.
- 11 S. Xie, F. Li, S. Xu, J. Li and W. Zeng, *Chin. J. Catal.*, 2019, **40**, 1205-1211.
- 12 J. Zhou, Y. Hu, Y.-C. Chang, Z. Hu, Y.-C. Huang, Y. Fan, H.-J. Lin, C.-W. Pao, C.-L. Dong, J.-F. Lee, C.-T. Chen, J.-Q. Wang and L. Zhang, *ACS Catal.*, 2022, **12**, 3138-3148.
- 13 C. Wang, H. Shang, Y. Wang, J. Li, S. Guo, J. Guo and Y. Du, *Nanoscale*, 2021, **13**, 7279-7284.
- 14 J. Liang, X. Gao, B. Guo, Y. Ding, J. Yan, Z. Guo, E. C. M. Tse and J. Liu, *Angew. Chem., Int. Ed.*, 2021, **60**, 12770-12774.
- 15 S. Kandambeth, V. S. Kale, D. Fan, J. A. Bau, P. M. Bhatt, S. Zhou, A. Shkurenko, M. Rueping, G. Maurin, O. Shekhah and M. Eddaoudi, *Adv. Energy Mater.*, 2023, **13**, 2202964.