

Supplementary Information

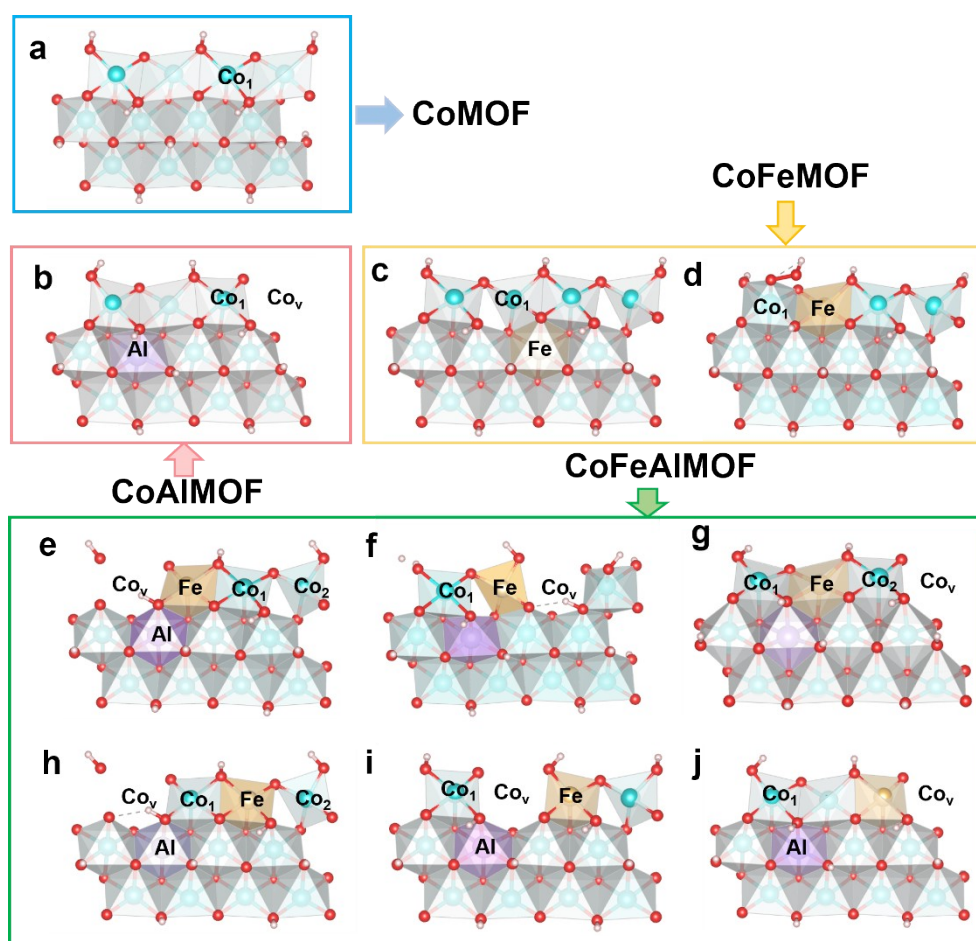


Figure S1. Potential active sites on (01-12) facets considered by Hubbard-corrected density functional theory calculations. a) Co₁ site in CoMOF. b) Co₁ site in CoAIMOF. c) Co₁ sites in CoFeMOF. d) Co₁ and Fe sites in CoFeMOF. e-j) Co₁ and Co₂ sites in CoFeAIMOF

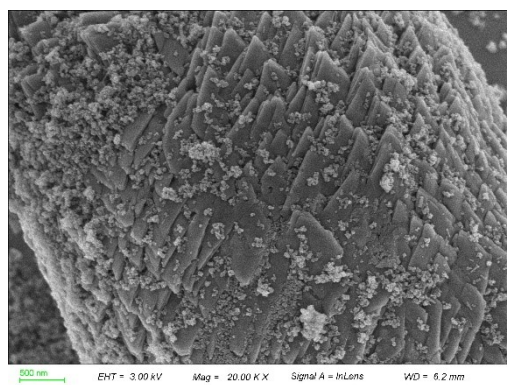


Figure S2. SEM image of CoFeAlMOF.

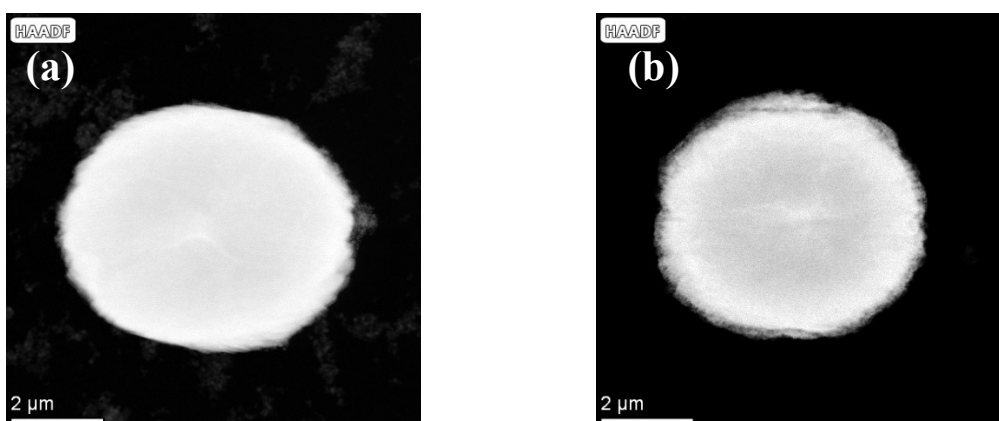


Figure S3. HAADF-STEM image of (a) CoFeAlMOF (b) CoV-CoFeAlMOF.

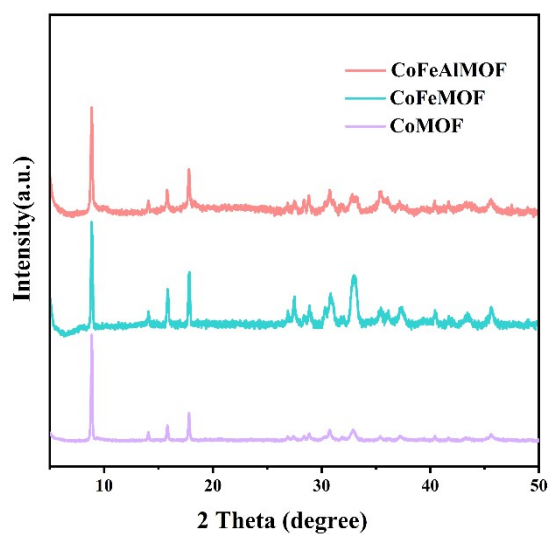


Figure S4. XRD patterns of CoFeAlMOF, CoFeMOF, CoMOF.

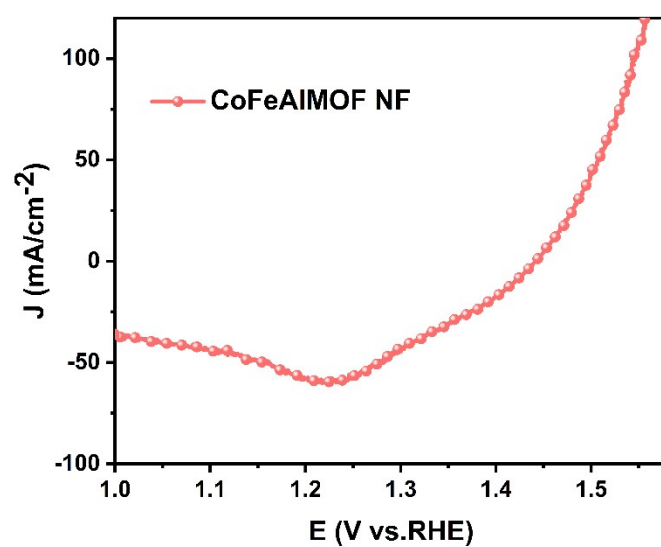


Figure S5. LSV curves with 90% iR-compensation grown on nickel foam substrates (requiring only 229 mV overpotential to achieve 10 mA cm⁻² for OER)

Supplementary Information Tables

Supplementary Table 1. Calculated energetics and overpotentials of different metal sites.

	Gibbs free energies (eV)			Gibbs free energy changes (eV)				η^{OER}
	ΔG_{OH^*}	ΔG_{O^*}	ΔG_{OOH^*}	ΔG_1	ΔG_2	ΔG_3	ΔG_4	(mV)
CoMOF-Co₁	0.62	2.36	3.92	0.62	1.74	1.56	1.00	0.51
CoAlMOF-Co₁	1.18	2.75	4.16	1.18	1.57	1.41	0.76	0.34
CoFeMOF-Co₁	0.96	2.74	3.86	0.96	1.78	1.12	1.06	0.55
CoFeMOF-Co₁	0.93	2.57	3.99	0.93	1.64	1.42	0.93	0.41
CoFeAlMOF-Co₁	0.84	2.37	3.99	0.84	1.53	1.62	0.93	0.39
CoFeAlMOF-Co₂	0.89	2.54	4.08	0.89	1.65	1.54	0.84	0.42
CoFeAlMOF-Co₁	1.23	2.68	4.03	1.23	1.45	1.35	0.89	0.22
CoFeAlMOF-Co₁	0.84	2.63	3.88	0.84	1.79	1.25	1.04	0.56
CoFeAlMOF-Co₂	1.42	2.85	4.28	1.42	1.43	1.43	0.64	0.2
CoFeAlMOF-Co₁	1.33	2.88	4.15	1.33	1.55	1.27	0.77	0.32
CoFeAlMOF-Co₂	0.96	2.63	3.85	0.96	1.67	1.22	1.07	0.44
CoFeAlMOF-Co₁	1.05	2.67	3.76	1.05	1.62	1.09	1.16	0.39
CoFeAlMOF-Co₁	0.91	2.65	4.03	0.91	1.74	1.38	0.89	0.51

Supplementary Table 2. Bader Charge of CoMOF, CoFeMOF , CoAlMOF and Co_v-CoFeAlMOF.

	d-band centre	Bader Charge
CoMOF	-1.54 eV	-1.26 e
CoFeMOF	-1.55 eV	-1.28 e
CoAlMOF	-2.10 eV	-1.30 e
CoFeAlMOF	-2.21 eV	-1.35 e

Supplementary Table 3. The atomic ratio of CoFeAlMOF, detected by ICP-OES.

Element	Co	Fe	Al
Atomic ratio (%)	48.4	25.8	25.8

Supplementary Table 4. The ratio of Al in CoFeAlMOF before and after etching.

Condition	Weight Percentage (W%) of Al
Before Etching	4.79
After Etching	2.97