Supporting Information

Structurally Engineered Bifunctional Oxygen Electrocatalyst from Cobalt-Based LDH-MOF Architecture

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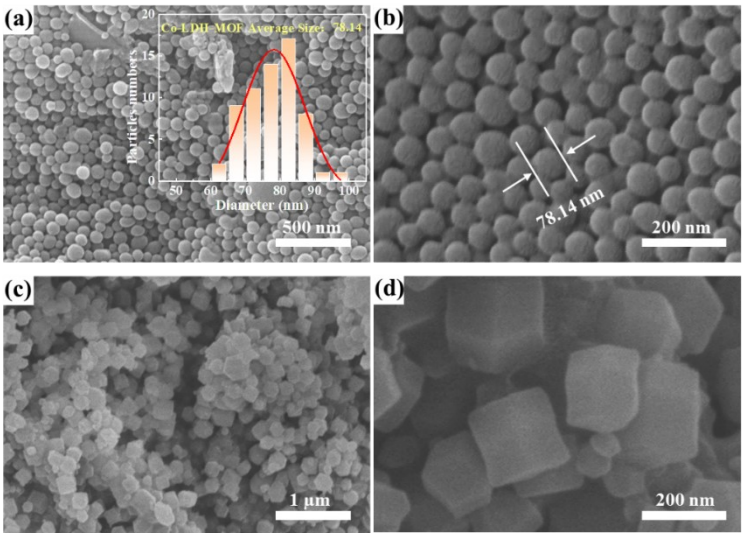

Fig. S1. Typical SEM images of Co-LDH-MOF (a, b) and Co-MOF (c, d).

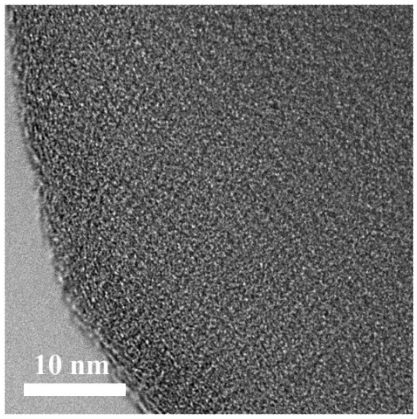

Fig. S2. Typical HRTEM image of Co-LDH-MOF.

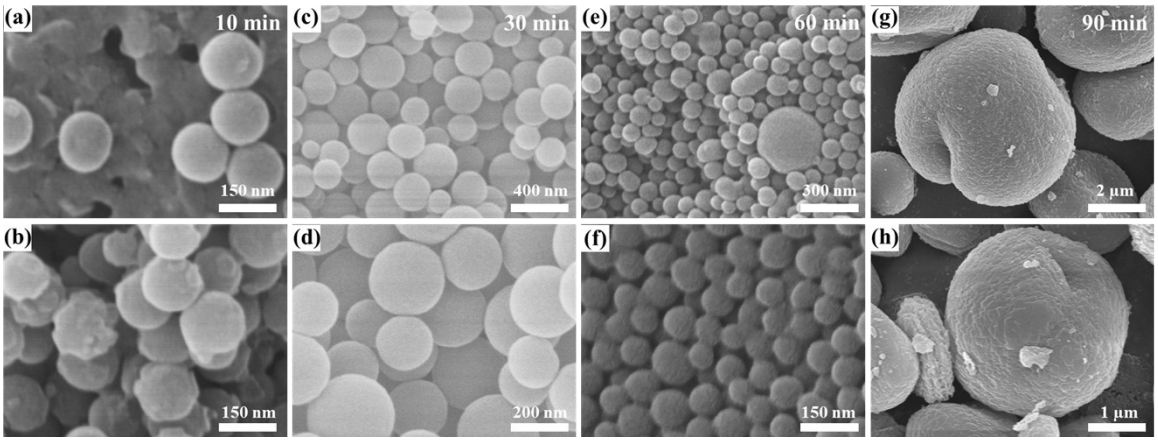

Fig. S3. Typical SEM images of Co-LDH-MOF and control samples prepared at different ultrasonication times: (a, b) 10 min, (c, d) 30 min, (e, f) 60 min (Co-LDH-MOF), and (g, h) 90 min.

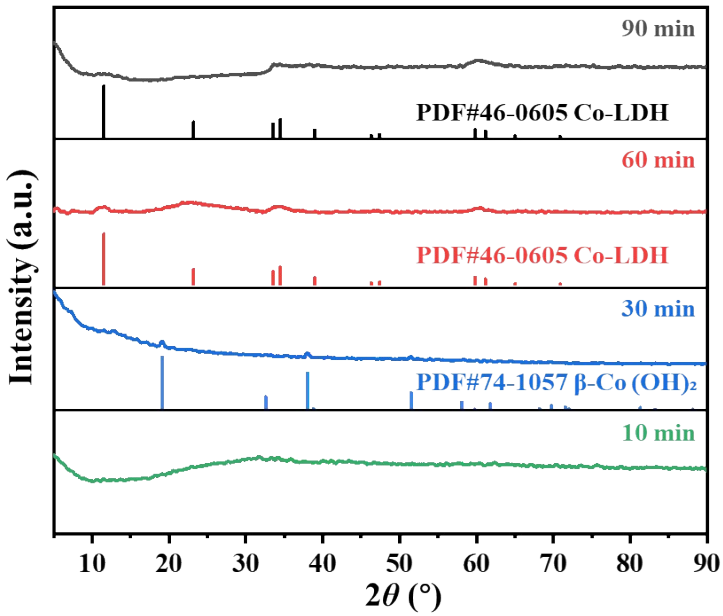

Fig. S4. XRD patterns of Co-LDH-MOF and control samples prepared at ultrasonication times of 10, 30, 60 (Co-LDH-MOF), and 90 min.

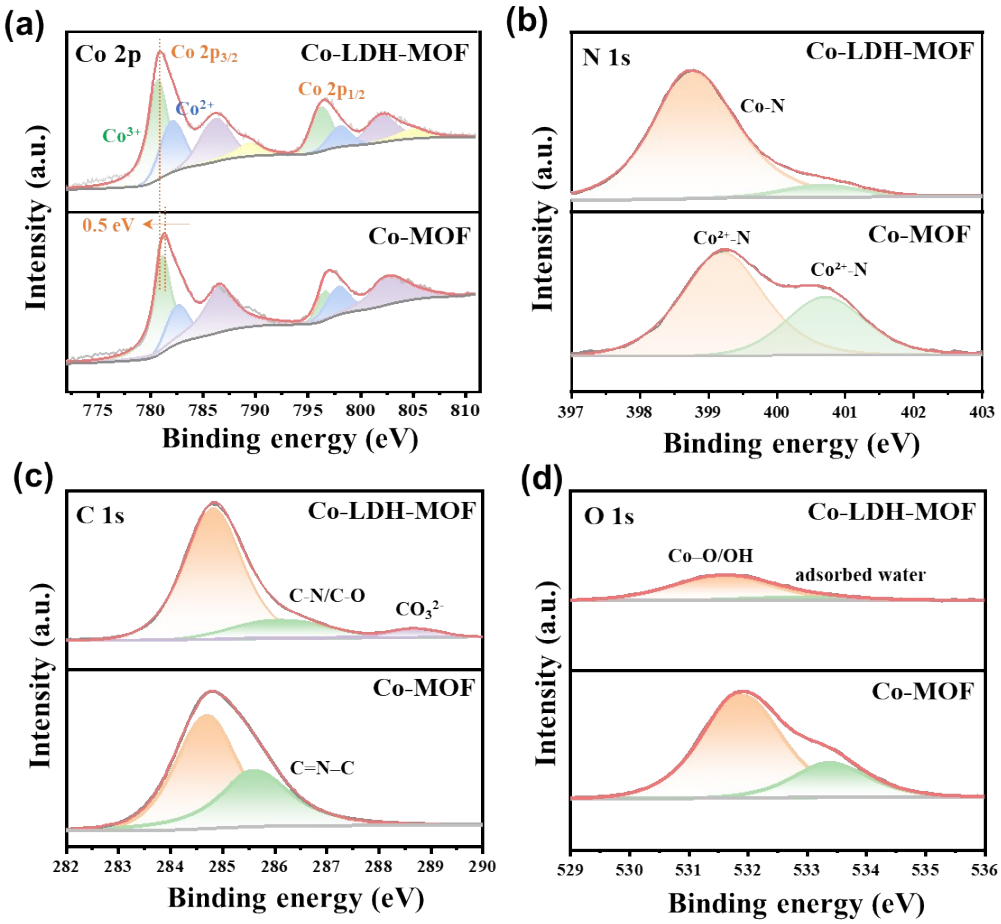

Fig. S5. Comparative high-resolution XPS spectra of Co 2p (a), N 1s (b), C 1s (c), O 2p (d) for Co-LDH-MOF and Co-MOF.

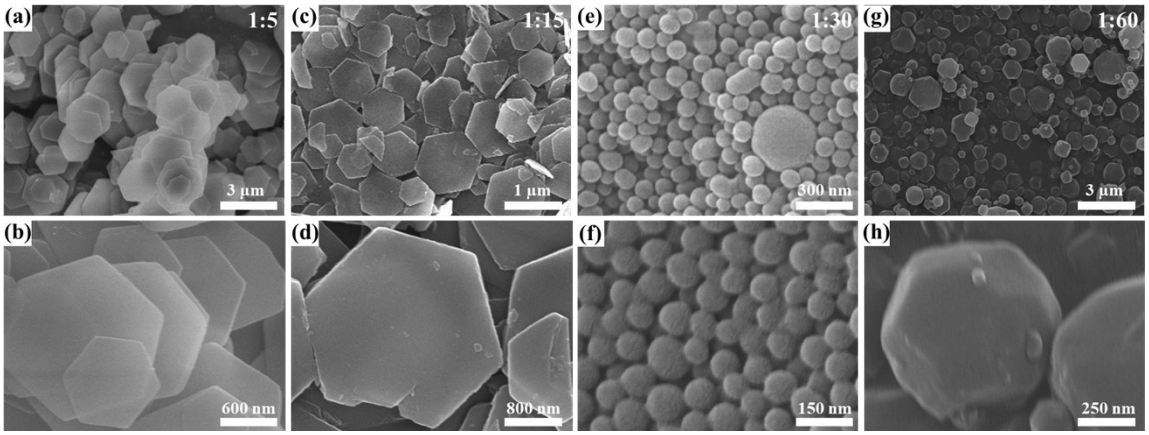

Fig. S6. Typical SEM images of Co-LDH-MOF and control samples prepared with different $Co(NO_3)_2$-to-MeIm molar ratios: (a, b) 1:5, (c, d) 1:15, (e, f) 1:30 (Co-LDH-MOF), and (g, h) 1:60.

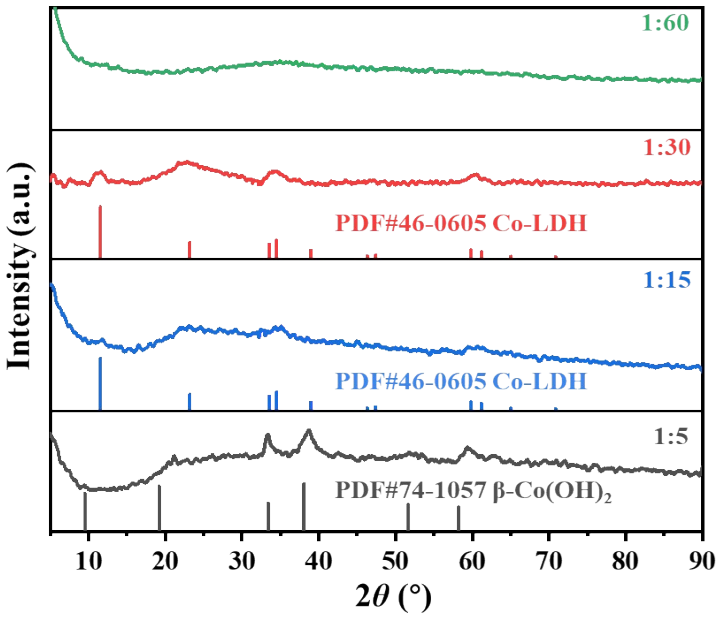

Fig. S7. XRD patterns of Co-LDH-MOF and control samples prepared at $Co(NO_3)_2$-to-MeIm molar ratios of 1:5, 1:15, 1:30 (Co-LDH-MOF), and 1:60.

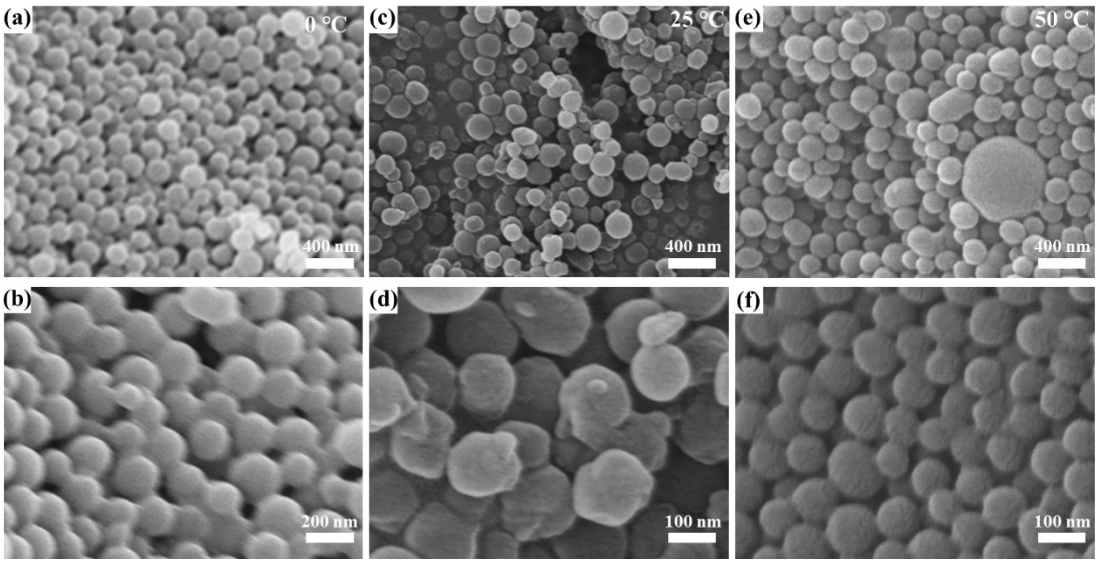

Fig. S8. Typical SEM images of Co-LDH-MOF and control samples prepared under similar conditions with different temperatures: (a, b) 0 °C, (c, d) 25 °C, and (e, f) 50 °C (Co-LDH-MOF).

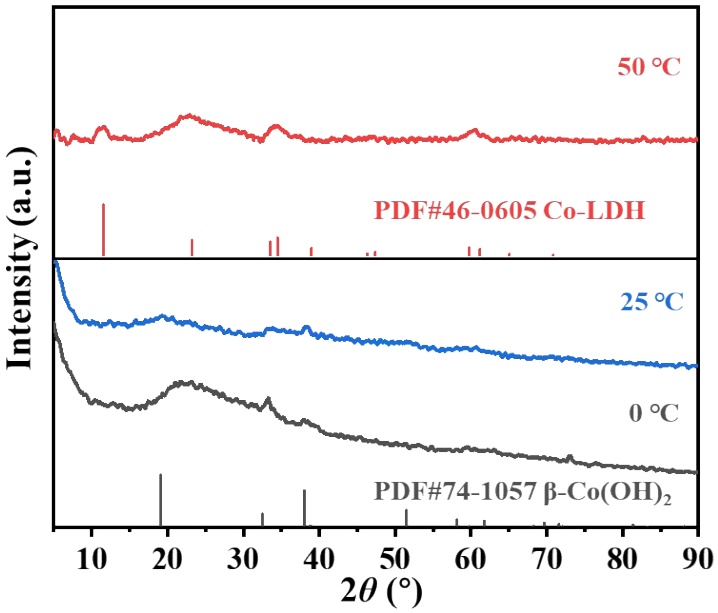

Fig. S9. XRD patterns of Co-LDH-MOF and control samples prepared at temperatures of 0, 25, and 50 °C (Co-LDH-MOF).

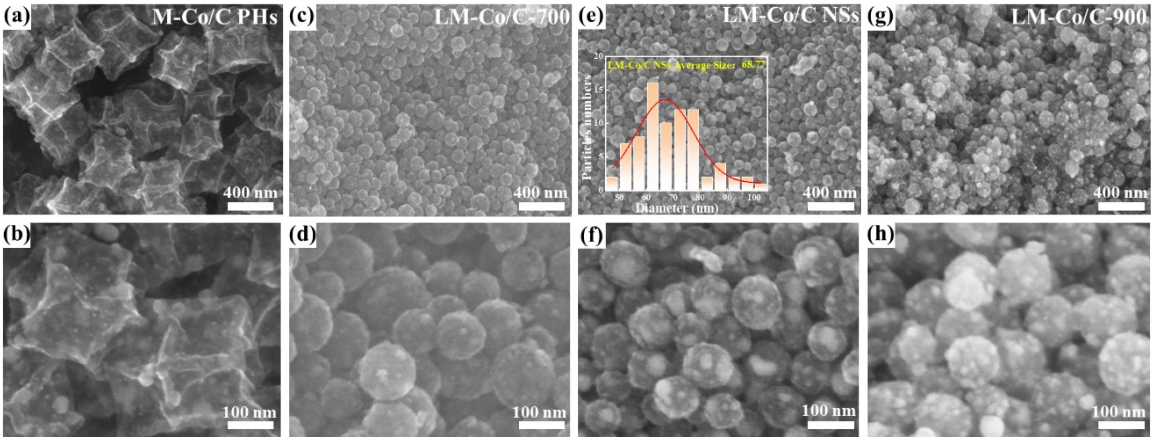

Fig. S10. Typical SEM images of M-Co/C PHs (a, b), LM-Co/C-700 (c, d), LM-Co/C-800 (the target sample) (e, f), and LM-Co/C-900 NSs (g, h).

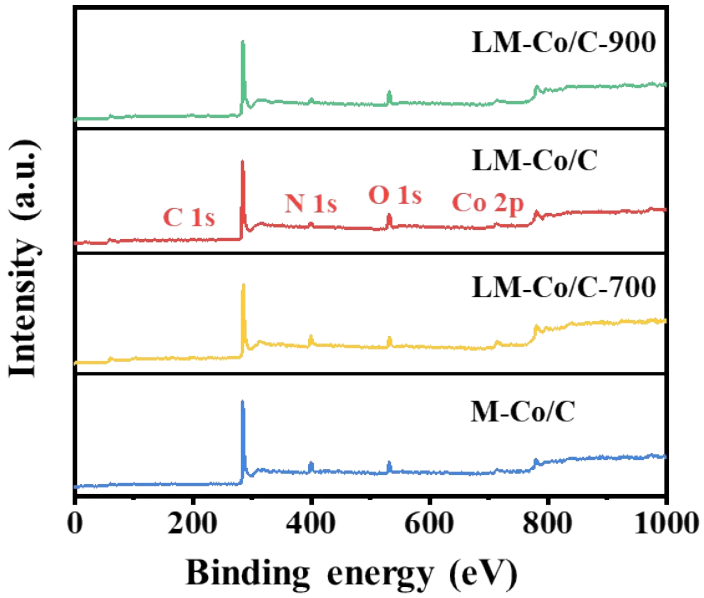

Fig. S11. XPS survey scans of LM-Co/C-T and M-Co/C PHs.

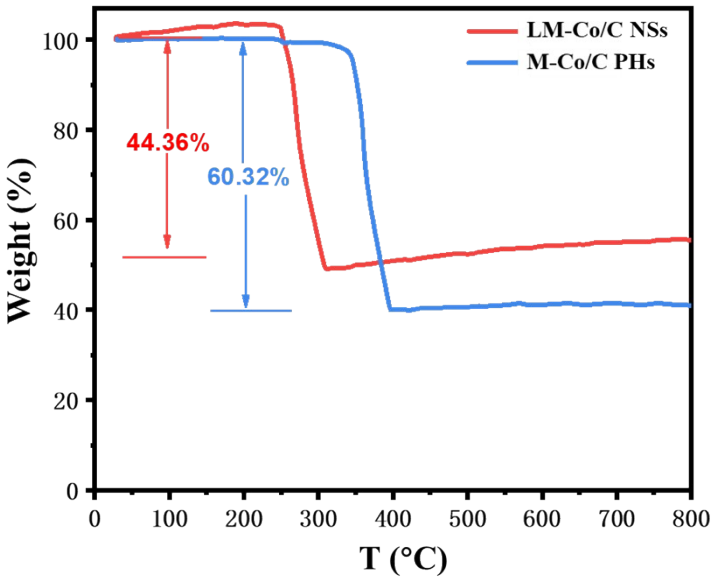

Fig. S12. TGA curves of LM-Co/C NSs and M-Co/C PHs conducted in air atmosphere.

Table S1. EDX compositional data of LM-Co/C NSs and M-Co/C PHs.

	Atomic Fraction (%)	Co	N	C	O
EDX	LM-Co/C NSs	38.79	2.46	44.63	14.12
	M-Co/C PHs	16.60	23.10	54.59	5.72

Table S2. Performance comparison of reported Co-based bifunctional oxygen electrocatalysts.

Catalysts	$E_{1/2}$ (V)	E_{j10} (V)	ΔE (V)	Power density (mW cm^{-2})	Stability	Refs.
LM-Co/C NSs	**0.855**	**1.548**	**0.693**	**167.25**	**1000 h@10 mA cm^{-2}**	**This work**
$(CoO_x@NC)_2$-NiFe	0.806	1.553	0.757	136.00	200 h@10 mA cm^{-2}	1
Co_3O_4/CoNGDY	0.854	1.540	0.689	167.60	/	2
RuO_2-Co_3O_4/NC	0.845	1.461	0.616	100.20	140 h@5 mA cm^{-2}	3
Co@CoO/BNC	0.850	1.539	0.710	115.72	300 h@2 mA cm^{-2}	4
CoO@Fe-N-C	0.880	1.540	0.710	134.00	150 cycles@5 mA cm^{-2}	5
CoO/CoN_x-C	0.844	1.614	0.770	161.00	1000 h@5 mA cm^{-2}	6
CoO_X/Co_9S_8@NC-2	0.830	1.460	0.630	114.50	427 h@2 mA cm^{-2}	7
Co/CoO@N-C-40	0.854	1.579	0.725	186.00	750 h@10 mA cm^{-2}	8
Co-LCFS-800	0.834	1.584	0.750	/	/	9
CoFe@CNT	0.850	1.600	0.750	115.00	105 h@20 mA cm^{-2}	10
ZIF-67-6	0.830	1.500	0.670	/	/	11
Co/Co_3O_4@NC	0.820	1.577	0.759	156.50	220 h@10 mA cm^{-2}	12
0.1Ni@Co_3O_4	0.800	1.579	0.780	/	24 h@5 mA cm^{-2}	13
N-GNRS/COO	0.825	1.554	0.729	138.81	135 h@10 mA cm^{-2}	14
CoO_x/Co@Co-N-C	0.880	1.576	0.700	168.40	250 h@5 mA cm^{-2}	15
CO/CoO-NG	0.819	1.600	0.784	244.28	340 h@5 mA cm^{-2}	16
CoO@Co-N-C	0.790	1.630	0.840	133.50	210 cycles@50 mA cm^{-2}	17
Co_3S_4@Co_3O_4/NSC	0.822	1.542	0.720	122.00	200 h@10 mA cm^{-2}	18

260-8						
CON/Co_3O_4HNPs@NCNWs	0.835	1.560	0.725	118.30	200 cycles@5 mA cm^{-2}	19
$NixCo_{1-x}$@Ni_xCo_{1-x}O/NCNT	0.790	1.610	0.820	157.00	30 h@5 mA cm^{-2}	20

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