Tuning Dimensionality and Linkage in Metal–Organic Frameworks for Enhanced Electrochemical Energy Storage

Altaf Husain[a †] Nissar Hussain[a, †] and Shaikh M. Mobin*[, a, b]

[a] Department of Chemistry, Indian Institute of Technology Indore, Simrol, Khandwa Road, Indore 453552, India.

[b] Centre for Advanced Electronics (CAE), Indian Institute of Technology Indore, Simrol, Khandwa Road, Indore 453552, India.

*E-mail: xray@iiti.ac.in

Equal contribution

[†] These authors contributed equally to this work

Chemicals

All organic ligands were purchased from Sigma Aldrich, and the reagents and solvents were obtained from Merck and used without further purification.

Instrumentation

A Rigaku Oxford Supernova CCD diffractometer was used to determine the single-crystal X-ray structure of the MOF. The XRD spectra were recorded on an X-ray diffractometer (Rigaku SmartLab) equipped with monochromated Cu Kα radiation (λ = 1.54 Å). Field-emission scanning electron microscopy (FESEM) and energy-dispersive X-ray (EDX) elemental mapping were performed to examine the surface morphology and elemental composition using the JEOL JSM-7610 F plus. Brunauer–Emmett–Teller (BET) surface area was conducted on an Autosorb iQ (Quantachrome Instruments, version 1.11). Thermogravimetric analysis (TGA) experiments were performed using a Mettler Toledo (TGA/DSC1) analyzer with *STAR*^{e} software and heated up to 800 °C using a 10 °C/min rate under a N_2 flow.

X-Ray Structural Studies

Single-crystal X-ray diffraction (SCXRD) data for the **3D-PL-MOF** and the **2D-L-MOF** were collected at 293 K on a Rigaku Oxford SuperNova CCD diffractometer using monochromated, graphite-filtered Mo Kα radiation (λ = 1.548 Å). Data were acquired using standard φ–ω scan techniques and processed with *CrysAlisPro* CCD and RED software. The structure was solved by direct methods using *SHELXS-97* and refined by full-matrix least-squares on F^2 with *SHELXL-97*.[1] All non-hydrogen atoms were located via direct methods and refined anisotropically. Hydrogen atoms were positioned geometrically and refined using isotropic displacement parameters set at 1.2 U_eq of the parent atoms. Non-covalent interactions, mean-plane analyses, and molecular graphics were prepared using *DIAMOND* (v3.1d).[2]

Electrochemical measurements

The as-synthesized **3D-PL-MOF** and **2D-L-MOF** freestanding electrode was precisely cut into 1 × 1 cm^2 pieces (with a mass loading of 3 mg cm^{-2}) and employed as the working electrode in a three-electrode configuration, using Ag/AgCl as the reference electrode and a graphite rod as the counter electrode. To evaluate the electrochemical performance, cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) measurements were conducted using an Autolab workstation with 1 M KOH aqueous solution as the electrolyte. Additionally, electrochemical studies were performed by drop-casting the MOF dispersion (in ethanol, achieving a mass loading of approximately 3 mg cm^{-2}) onto carbon paper, which served as the working electrode. A two-electrode device was assembled with activated carbon deposited on carbon paper as the current collector. In the solid-state supercapacitor assembly, the positive and negative electrodes were separated by cellulose paper and securely fixed to facilitate evaluation of device performance.

Electrochemical Calculations

The specific capacitance (C_s) (Fg^{-1}) of the symmetric device was measured by using the following equation:

$$Cs = \frac{I\ x\ \Delta t}{m\ x\ \Delta V} \quad \textbf{(1)}$$

where I/m is current density, and Δt and ΔV convey the discharge time and potential window of the GCD profile, respectively.

In the case of the asymmetric supercapacitor device, the energy density (E_d) and power density (P_d) were determined using the following equations:

$$E_d = \frac{Cs\ x\ \Delta V^2}{2\ x\ 3.6} \quad \textbf{(2)}$$

And

$$P_d = \frac{E_d\ x\ 3600}{\Delta t} \quad \textbf{(3)}$$

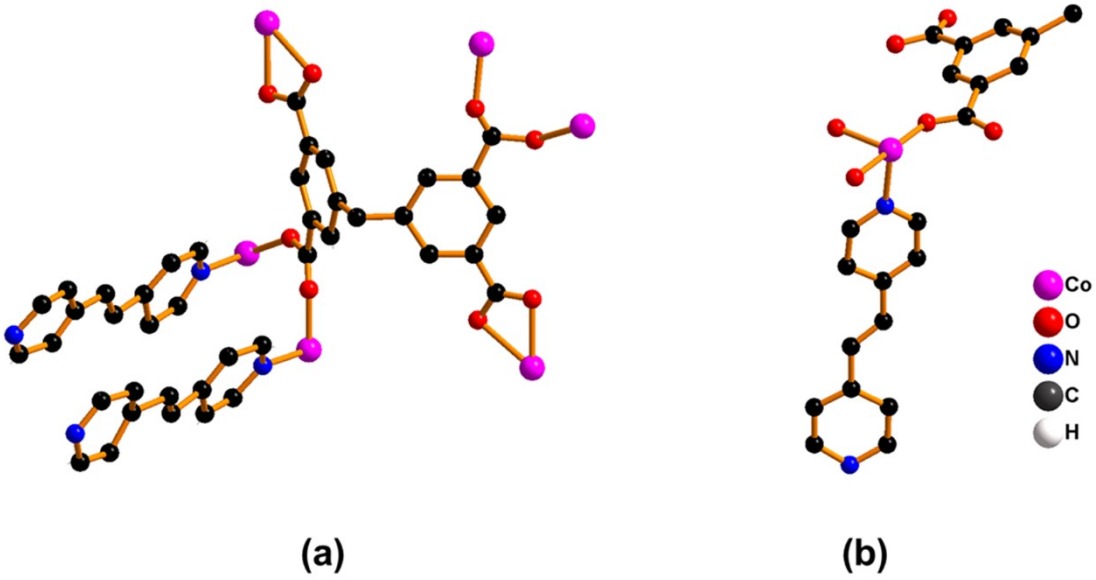

Fig. S1. (a) Asymmetric Unit (a) **3D-PL-MOF** (b)**2D-L-MOF** (Note: H atom has been omitted for clarity).

Table S1. Crystallographic parameters of **3D-PL-MOF** and **2D-L-MOF.**

Identification code	**3D-PL-MOF**	**2D-L-MOF**
Empirical formula	$C_{41}H_{24}Co_2N_4O_9$	$C_{41}H_{36}Co_2N_4O_{12}$
Formula weight	834.50	894.60
Crystal system	Orthorhombic	Monoclinic
Space group	$Pca2_1$	C2/c
a (Å)	26.3002(12)	22.7562(13)
b (Å)	13.6647(7)	13.8836(7)
c (Å)	17.4818(15	22.7193(19)
α (˚)	90	90
β (˚)	90	109.233(7)
γ (˚)	90	90
V ($Å^3$)	6282.7(7)	6777.3(8)
Z, d_{calcd} (mg m^{-3})	4, 0.882	4, 0.877
Temperature (K)	293(2)	293(2)
Wavelength (Å)	0.71073	0.71073
θ range/deg	3.171 to 29.147	3.060 to 29.903
Goodness-of-fit (GOOF)	0.951	0.974
R_1, a wR_2b [I > 2σ(I)]	R_1 = 0.0580, wR_2 = 0.1257	R_1 = 0.1102, wR_2 = 0.3267
R_1, a wR_2 b (all data)	R_1 = 0.0873, wR_2 = 0.1430	R_1 = 0.1680, wR_2 = 0.3838
Absorption correction	Semi-empirical from	Semi-empirical from

	equivalents	equivalents
Index ranges	-33<=h<=32, -17<=k<=18, -23<=l<=15	-28<=h<=28, -17<=k<=18, -30<=l<=26
Crystal size (mm^3)	0.330 x 0.260 x 0.210	0.350 x 0.320 x 0.270
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Reflections collected / unique	23896 / 10628 [R(int) = 0.1195]	23520 / 7766 [R(int) = 0.1498]
F (000)	1696	1840
CCDC no.	2471973	2471997

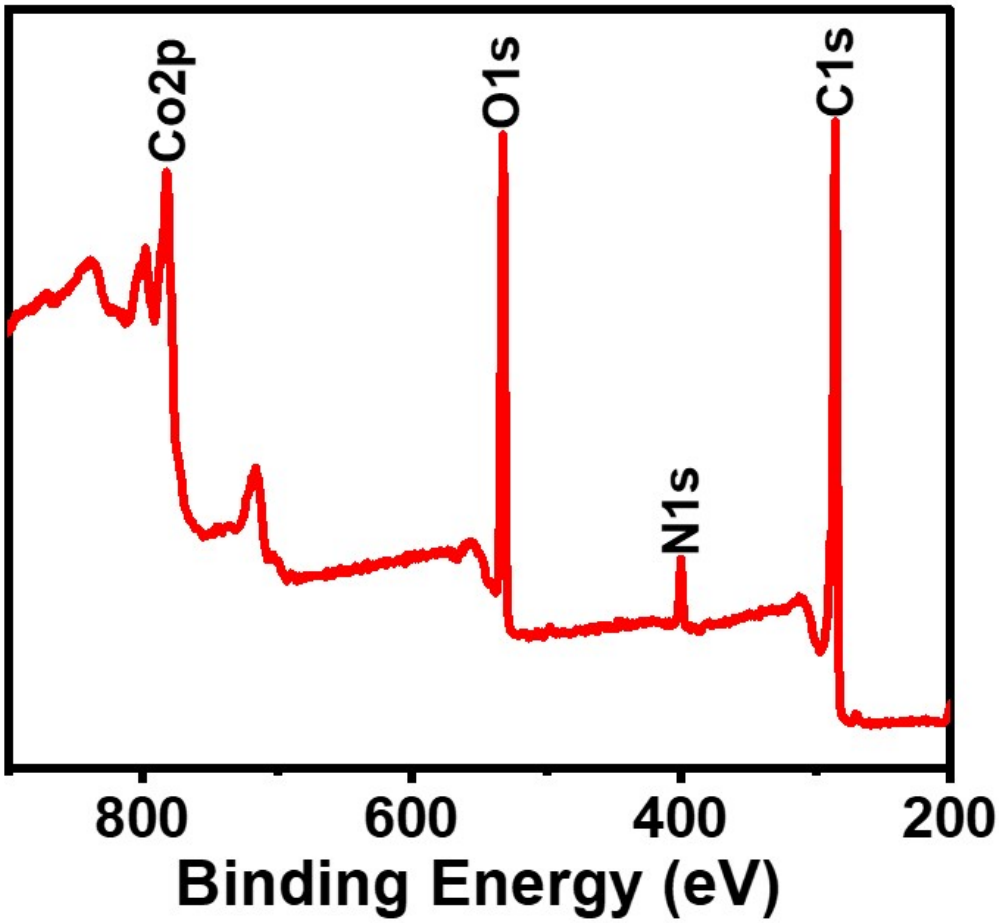

Fig. S2. XPS spectra survey scan of **3D-PL-MOF.**

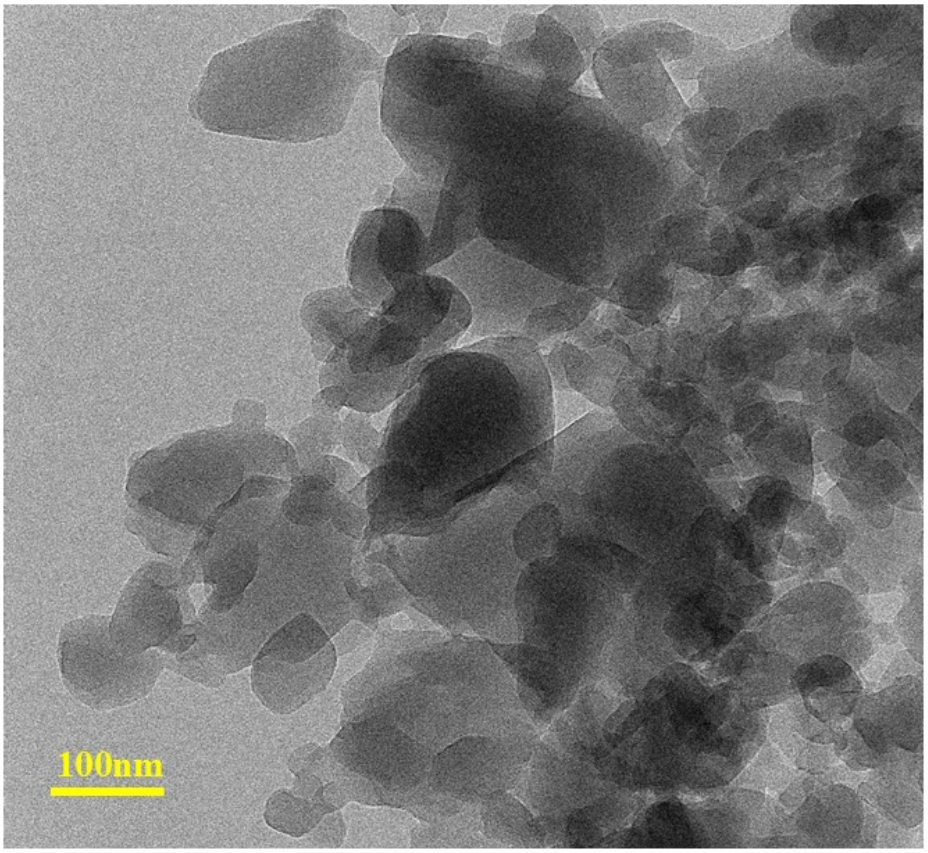

Fig. S3. TEM image of **3D-PL-MOF**.

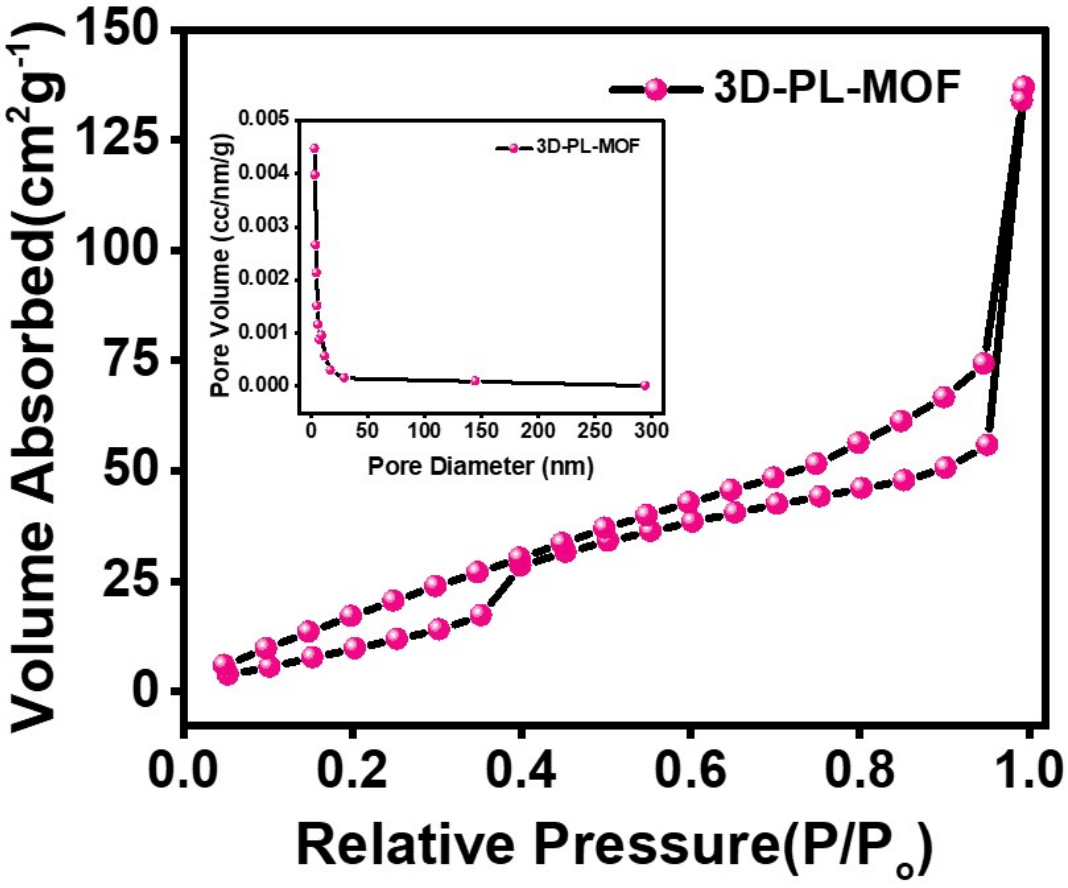

Fig. S4. Nitrogen adsorption-desorption isotherm and pore distribution (inset) of **3D-PL-MOF** at 77.3 K.

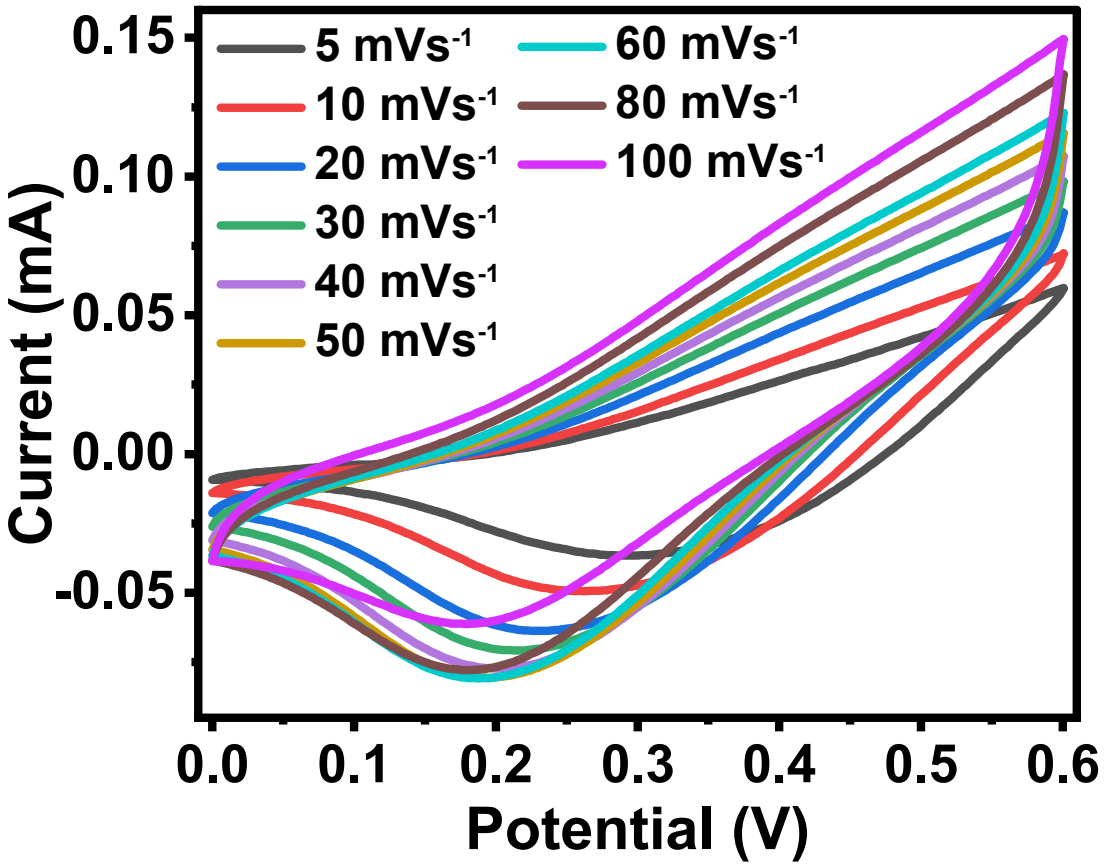

Fig. S5. CV of **2D-PL-MOF** at different scan rates in a three-electrode configuration.

Table S2. Circuit parameters obtained after fitting the EIS data to the used equivalent circuit.

Resistance Parameter	**3D-PL-MOF**	**2D-L-MOF**
Rct	6.93 Ω	106 Ω
Rs	2.82 Ω	1.52 Ω
C_{dl}	367 μF	17.4 μF
R_w	15.8mMho*s^N	2.26mMho*s^(1/2)

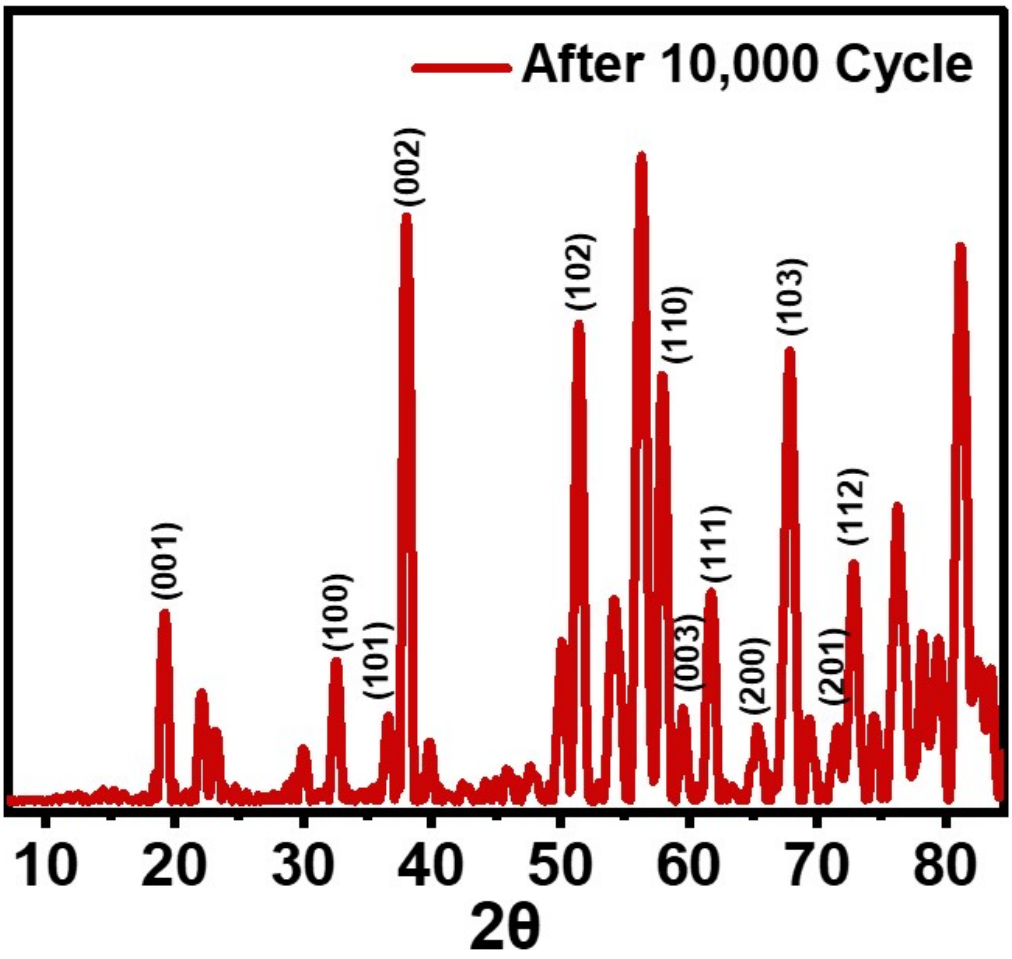

Fig. S6. XRD of **3D-PL-MOF** after 10,000 GCD cycles.

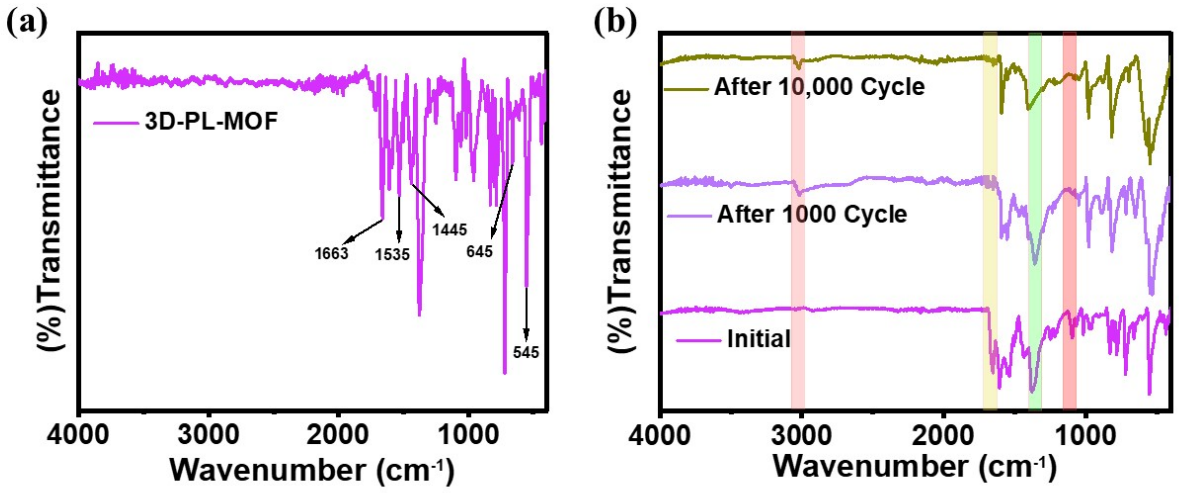

Fig. S7. (a) ATR-FTIR spectrum of the **3D-PL-MOF**. (b) ex-situ ATR-FTIR of **3D-PL-MOF** after 1000 and 10,000 cycle.

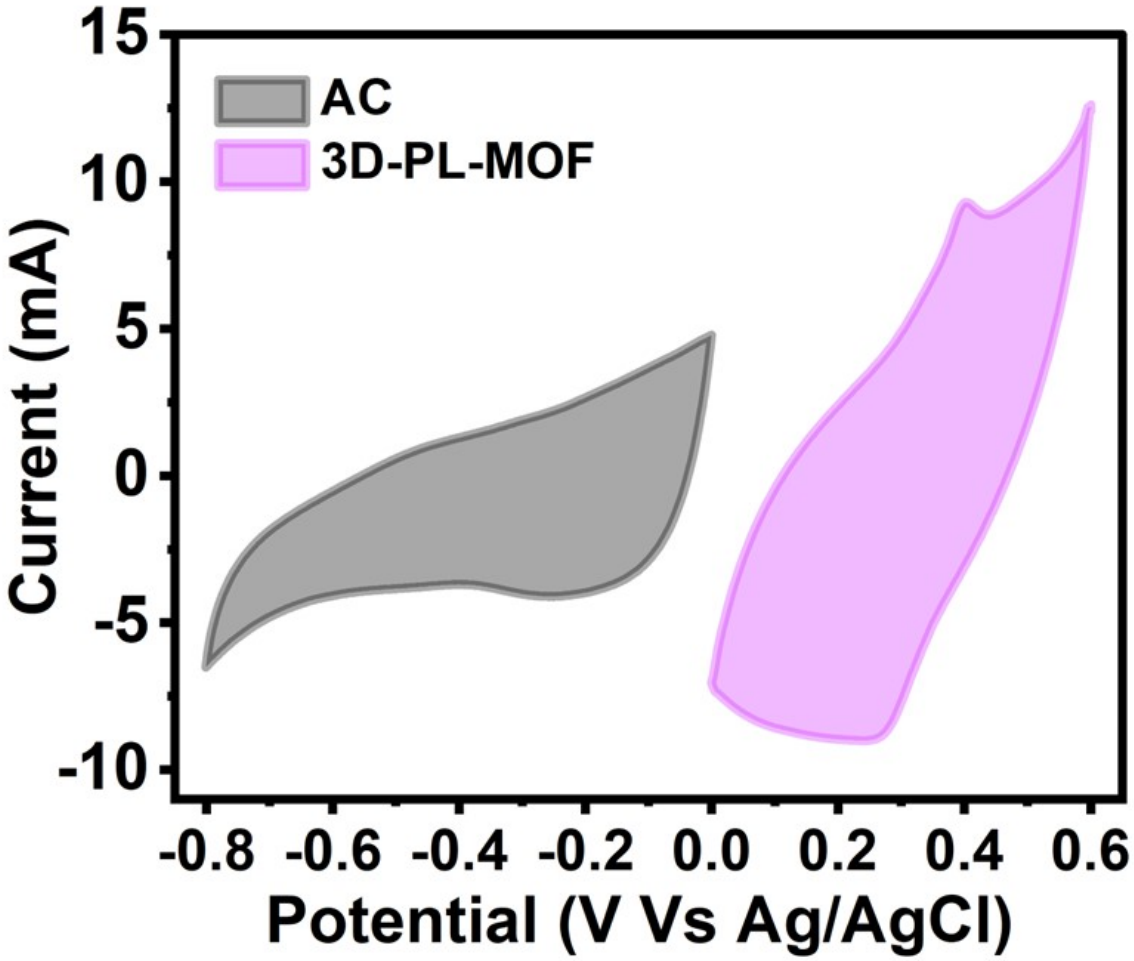

Fig. S8. CV curves of the AC and 3D-PL-MOF, at a scan rate of 50 mV s^{-1} in three-electrode configuration.

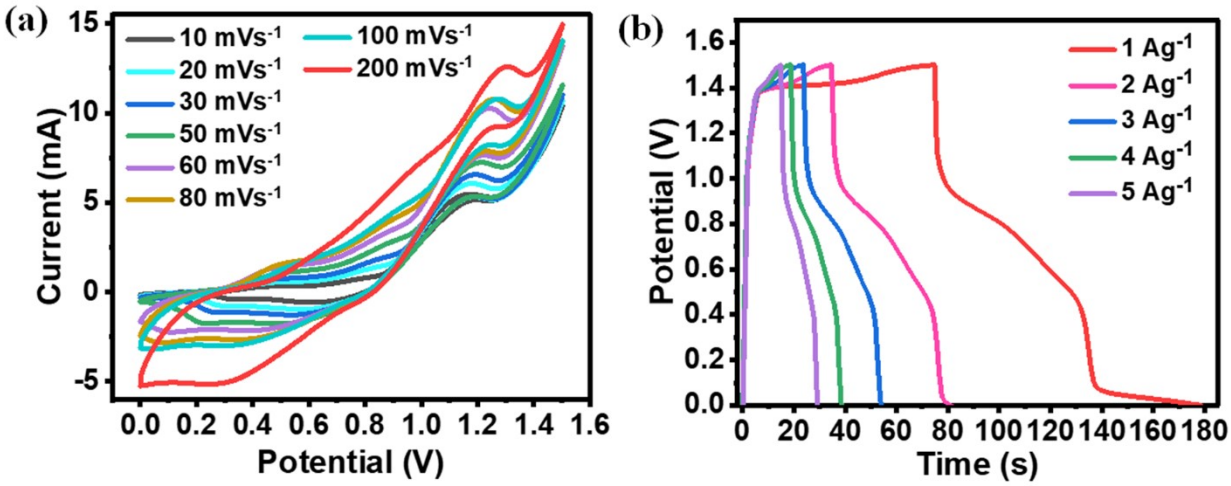

Fig. S9. Device analysis of **2D-L-MOF** (a) CV at different scan rates (b) GCD at different current densities.

Table S3. Comparison of as synthesized material with some previous reports.

Sr.No	Electrode Material	Current density (Ag^{-1} /$mAcm^{-2}$/mVs^{-1})	Specific capacitance/Capacity (Fg^{-1}/Cg^{-1} /F cm^{-2})	Electrolyte	Cyclic Stability	Reference
1.	NH_2-TMU-53	5 A g^{-1}	325 F g^{-1}	6 M KOH	92.03% after 6000 cycles	3
2	CuZn-MOF-P7	1 mA cm^{-2}	3.7 F cm^{-2}	1 M KOH	97% after 16,000 cycles	4
3	BPHCC-1	3 A g^{-1}	84 F g^{-1}	2 M KOH	64% after 500 cycles	5
4	SEU-1	1 A g^{-1}	145.5 Fg^{-1}	3 M KOH	92.7% after 3000 cycles	6
5	Na/Co-MOF (**1**	321.8 Fg^{-1}	4 A g^{-1}	0.5M Na_2SO_4	97.4% after 5000 cycles	7
6	Cu-PTA-MOF@PANI	0.5 A g^{-1}	232 C g^{-1}	1 M KOH	98.8% after 10,000 cycles	8
7	Cd-MOF	0.5 A g^{-1}	321 F g^{-1}	2 M KOH	95.2% after 1000 cycles	9
8	ZBN-M-900	1 A g^{-1}	292 F g^{-1}	1.0M H_2SO_4	98.46% after 10,000 cycles	10
9	SEU-1	1 A g^{-1}	145.5 F g^{-1}	3 M KOH	92.7 % after 6000 cycles	11
10	BBU-1	3 mV s^{-1},	134.5 F g^{-1}	2M KOH	91.3% after 1000 cycles	12
11	**3D-PL-MOF**	**1 Ag^{-1}**	**355 Fg^{-1}**	**1M KOH**	**92.85% after 10,000 cycles**	**This work**

Sr No	Electrode Material	Energy Density (W h kg^{-1})	Power Density (W kg^{-1})	Electrolyte	Potential Window	Reference
1.	Co-MOF	22.89	825	6 M KOH	1.75 V	[13]
2.	Ni-BPE	25.2	800	3 M KOH	1.60 V	[14]
3.	Ce–H_2L MOF	10.3	1425	2 M KOH	1.50 V	[15]
4.	NiFRS	28.7	400	-	1.6 V	[16]
5.	Co(II)-TMU-63	24.13	442	2 M KOH	1.7 V	[17]
6.	Co-FLF	24.88	84.5	2 M KOH	1.6 V	[18]
7	Co-MOF	25.8	1190	6 M KOH	1.6V	[19]
8.0	**3D-PL-MOF**	**31.63**	**749.87**	**1 M KOH**	**1.6 V**	**This work**

Table S4. Comparison of as synthesized material with some previous ASC report.

Table S5. Bond length of **3D-PL-MOF.**

Atom 1- Atom 2	**Length (Å)**
Co(1)-O(6)	1.999(4)
Co(1)-O(1)	2.033(6)
Co(1)-O(3)	2.139(5)
Co(1)-N(3)	2.147(5)
Co(1)-N(4)	2.151(5)
Co(1)-O(4)	2.178(6)
Co(1)-C(5)	2.486(10)
Co(2)-O(5)	1.965(5)
Co(2)-O(2)	1.996(4)
Co(2)-N(2)	2.096(5)
Co(2)-N(1)	2.141(5)
Co(2)-O(7)#1	2.250(5)
Co(2)-O(8)#1	2.259(6)
O(1)-C(1)	1.238(9)
O(2)-C(1)	1.278(8)
O(3)-C(5)	1.238(9)
O(4)-C(5)	1.275(8)
O(5)-C(13)	1.233(9)
O(6)-C(13)	1.320(8)
O(7)-C(16)	1.269(10)
O(8)-C(16)	1.249(8)
O(101)-H(10A)	0.8499
O(101)-H(10B)	0.8500
O(102)-H(10C)	0.8500
O(102)-H(10D)	0.8501
N(1)-C(18)	1.298(9)
N(1)-C(22)	1.339(10)
N(2)-C(27)	1.354(9)
N(2)-C(29)	1.376(10)
N(3)-C(34)	1.284(8)
N(3)-C(30)	1.392(9)
N(4)-C(39)	1.232(9)
N(4)-C(41)	1.349(10)
C(1)-C(2)	1.544(10)
C(2)-C(3)	1.364(11)
C(2)-C(8)	1.389(8)
C(3)-C(4)	1.428(9)
C(3)-H(3)	0.9300
C(4)-C(6)	1.371(10)

C(4)-C(5)#1	1.454(11)
C(6)-C(7)	1.430(11)
C(6)-H(6)	0.9300
C(7)-C(8)	1.400(8)
C(7)-C(9)	1.553(9)
C(8)-H(8)	0.9300
C(9)-C(10)	1.506(10)
C(9)-H(9A)	0.9700
C(9)-H(9B)	0.9700
C(10)-C(11)	1.350(9)
C(10)-C(17)	1.432(12)
C(11)-C(12)	1.397(10)
C(11)-H(11)	0.9300
C(12)-C(14)	1.298(11)
C(12)-C(13)#2	1.538(9)
C(14)-C(15)	1.347(9)
C(14)-H(14)	0.9300
C(15)-C(17)	1.384(9)
C(15)-C(16)	1.517(12)
C(17)-H(17)	0.9300
C(30)-C(31)	1.363(9)
C(30)-H(30)	0.9300
C(31)-C(32)	1.405(11)
C(31)-H(31)	0.9300
C(32)-C(33)	1.405(11)
C(32)-C(35)	1.515(9)
C(33)-C(34)	1.372(9)
C(33)-H(33)	0.93000
C(34)-H(34)	0.9300
C(35)-C(36)	1.255(11)
C(36)-C(37)	1.478(10)
C(37)-C(40)	1.291(10)
C(37)-C(38)	1.429(12)
C(38)-C(39)#3	1.381(10)
C(38)-H(38)	0.9300
C(39)-H(39)	0.9300
C(40)-C(41)#3	1.351(10)
C(40)-H(40)	0.9300
C(41)-H(41)	0.9300
C(18)-C(19)	1.403(9)
C(18)-H(18)	0.9300
C(19)-C(20)	1.398(14)
C(19)-H(19)	0.9300
C(20)-C(21)	1.423(15)
C(20)-C(23)	1.473(11)
C(21)-C(22)	1.452(12)

C(21)-H(21)	0.9300
C(22)-H(22)	0.9300
C(27)-C(26)	1.304(9)
C(27)-H(27)	0.9300
C(29)-C(28)	1.365(13)
C(29)-H(29)	0.9300
C(23)-C(24)#3	1.20(2)
C(23)-C(24A)#3	1.24(3)
C(26)-C(25)	1.202(17)
C(26)-C(25A)	1.69(2)
C(28)-C(25A)	1.10(2)
C(28)-C(25)	1.68(2)
C(24)-C(25)	1.684(16)
C(24A)-C(25A)	1.51(2)

Table S6. Bond angles of **3D-PL-MOF.**

Atom 1-Atom 2-Atom 3	**Angle (deg)**
O(6)-Co(1)-O(1)	118.4(3)
O(6)-Co(1)-O(3)	150.3(2)
O(1)-Co(1)-O(3)	91.3(2)
O(6)-Co(1)-N(3)	90.3(2)
O(1)-Co(1)-N(3)	93.6(2)
O(3)-Co(1)-N(3)	89.9(2)
O(6)-Co(1)-N(4)	88.7(2)
O(1)-Co(1)-N(4)	87.3(2)
O(3)-Co(1)-N(4)	90.8(2)
N(3)-Co(1)-N(4)	178.9(2)
O(6)-Co(1)-O(4)	90.0(2)
O(1)-Co(1)-O(4)	151.6(2)
O(3)-Co(1)-O(4)	60.48(18)
N(3)-Co(1)-O(4)	84.2(3)
N(4)-Co(1)-O(4)	95.4(3)
O(6)-Co(1)-C(5)	120.8(3)
O(1)-Co(1)-C(5)	120.7(2)
O(3)-Co(1)-C(5)	29.87(18)
N(3)-Co(1)-C(5)	83.8(3)
N(4)-Co(1)-C(5)	96.3(3)
O(4)-Co(1)-C(5)	30.8(2)
O(5)-Co(2)-O(2)	121.8(2)
O(5)-Co(2)-N(2)	93.6(2)
O(2)-Co(2)-N(2)	90.21(18)
O(5)-Co(2)-N(1)	88.5(2)
O(2)-Co(2)-N(1)	89.48(19)
N(2)-Co(2)-N(1)	177.7(3)
O(5)-Co(2)-O(7)#1	89.54(19)

O(2)-Co(2)-O(7)#1	148.7(2)
N(2)-Co(2)-O(7)#1	88.5(2)
N(1)-Co(2)-O(7)#1	90.6(2)
O(5)-Co(2)-O(8)#1	146.42(19)
O(2)-Co(2)-O(8)#1	91.4(2)
N(2)-Co(2)-O(8)#1	91.1(2)
N(1)-Co(2)-O(8)#1	86.7(3)
O(7)#1-Co(2)-O(8)#1	57.35(18)
C(1)-O(1)-Co(1)	142.1(5)
C(1)-O(2)-Co(2)	134.1(4)
C(5)-O(3)-Co(1)	90.8(5)
C(5)-O(4)-Co(1)	88.0(5)
C(13)-O(5)-Co(2)	143.6(4)
C(13)-O(6)-Co(1)	138.2(4)
C(16)-O(7)-Co(2)#4	92.0(4)
C(16)-O(8)-Co(2)#4	92.1(6)
H(10A)-O(101)-H(10B)	104.5
H(10C)-O(102)-H(10D)	104.5
C(18)-N(1)-C(22)	120.2(8)
C(18)-N(1)-Co(2)	119.0(5)
C(22)-N(1)-Co(2)	120.7(6)
C(27)-N(2)-C(29)	112.3(7)
C(27)-N(2)-Co(2)	124.4(5)
C(29)-N(2)-Co(2)	123.3(5)
C(34)-N(3)-C(30)	112.9(6)
C(34)-N(3)-Co(1)	123.2(5)
C(30)-N(3)-Co(1)	123.5(5)
C(39)-N(4)-C(41)	114.0(6)
C(39)-N(4)-Co(1)	122.9(5)
C(41)-N(4)-Co(1)	122.0(5)
O(1)-C(1)-O(2)	124.8(7)
O(1)-C(1)-C(2)	120.8(7)
O(2)-C(1)-C(2)	113.8(7)
C(3)-C(2)-C(8)	122.0(7)
C(3)-C(2)-C(1)	120.6(6)
C(8)-C(2)-C(1)	117.2(7)
C(2)-C(3)-C(4)	118.7(7)
C(2)-C(3)-H(3)	120.7
C(4)-C(3)-H(3)	120.7
C(6)-C(4)-C(3)	118.5(7)
C(6)-C(4)-C(5)#1	120.2(7)
C(3)-C(4)-C(5)#1	121.0(7)
O(3)-C(5)-O(4)	119.8(8)
O(3)-C(5)-C(4)#4	121.9(7)
O(4)-C(5)-C(4)#4	118.3(7)
O(3)-C(5)-Co(1)	59.4(4)

O(4)-C(5)-Co(1)	61.1(5)
C(4)#4-C(5)-Co(1)	173.4(5)
C(4)-C(6)-C(7)	124.0(6)
C(4)-C(6)-H(6)	118
C(7)-C(6)-H(6)	118
C(8)-C(7)-C(6)	114.8(6)
C(8)-C(7)-C(9)	123.3(7)
C(6)-C(7)-C(9)	121.9(6)
C(2)-C(8)-C(7)	121.9(7)
C(2)-C(8)-H(8)	119
C(7)-C(8)-H(8)	119
C(10)-C(9)-C(7)	109.4(6)
C(10)-C(9)-H(9A)	109.8
C(7)-C(9)-H(9A)	109.8
C(10)-C(9)-H(9B)	109.8
C(7)-C(9)-H(9B)	109.8
H(9A)-C(9)-H(9B)	108.2
C(11)-C(10)-C(17)	118.9(8)
C(11)-C(10)-C(9)	121.2(9)
C(17)-C(10)-C(9)	119.6(6)
C(10)-C(11)-C(12)	118.9(8)
C(10)-C(11)-H(11)	120.5
C(12)-C(11)-H(11)	120.5
C(14)-C(12)-C(11)	122.8(7)
C(14)-C(12)-C(13)#2	120.9(8)
C(11)-C(12)-C(13)#2	115.8(8)
O(5)-C(13)-O(6)	124.3(6)
O(5)-C(13)-C(12)#5	121.0(7)
O(6)-C(13)-C(12)#5	114.6(7)
C(12)-C(14)-C(15)	119.8(7)
C(12)-C(14)-H(14)	120.1
C(15)-C(14)-H(14)	120.1
C(14)-C(15)-C(17)	121.4(7)
C(14)-C(15)-C(16)	121.8(7)
C(17)-C(15)-C(16)	116.5(7)
O(8)-C(16)-O(7)	118.5(9)
O(8)-C(16)-C(15)	121.6(9)
O(7)-C(16)-C(15)	119.9(6)
C(15)-C(17)-C(10)	117.7(6)
C(15)-C(17)-H(17)	121.2
C(10)-C(17)-H(17)	121.2
C(31)-C(30)-N(3)	127.8(8)
C(31)-C(30)-H(30)	116.1
N(3)-C(30)-H(30)	116.1
C(30)-C(31)-C(32)	115.3(8)
C(30)-C(31)-H(31)	122.4

C(32)-C(31)-H(31)	122.4
C(33)-C(32)-C(31)	118.1(7)
C(33)-C(32)-C(35)	123.7(8)
C(31)-C(32)-C(35)	118.0(8)
C(34)-C(33)-C(32)	119.1(7)
C(34)-C(33)-H(33)	120.5
C(32)-C(33)-H(33)	120.5
N(3)-C(34)-C(33)	126.4(7)
N(3)-C(34)-H(34)	116.8
C(33)-C(34)-H(34)	116.8
C(36)-C(35)-C(32)	126.4(9)
C(35)-C(36)-C(37)	120.6(8)
C(40)-C(37)-C(38)	116.1(7)
C(40)-C(37)-C(36)	129.7(8)
C(38)-C(37)-C(36)	114.1(8)
C(39)#3-C(38)-C(37)	115.7(9)
C(39)#3-C(38)-H(38)	122.1
C(37)-C(38)-H(38)	122.1
N(4)-C(39)-C(38)#6	127.5(8)
N(4)-C(39)-H(39)	116.2
C(38)#6-C(39)-H(39)	116.2
C(37)-C(40)-C(41)#3	122.1(8)
C(37)-C(40)-H(40)	118.9
C(41)#3-C(40)-H(40)	118.9
N(4)-C(41)-C(40)#6	123.2(7)
N(4)-C(41)-H(41)	118.4
C(40)#6-C(41)-H(41)	118.4
N(1)-C(18)-C(19)	125.0(9)
N(1)-C(18)-H(18)	117.5
C(19)-C(18)-H(18)	117.5
C(20)-C(19)-C(18)	118.8(9)
C(20)-C(19)-H(19)	120.6
C(18)-C(19)-H(19)	120.6
C(19)-C(20)-C(21)	114.5(7)
C(19)-C(20)-C(23)	123.9(13)
C(21)-C(20)-C(23)	121.1(12)
C(20)-C(21)-C(22)	120.4(10)
C(20)-C(21)-H(21)	119.8
C(22)-C(21)-H(21)	119.8
N(1)-C(22)-C(21)	118.1(10)
N(1)-C(22)-H(22)	120.9
C(21)-C(22)-H(22)	120.9
C(26)-C(27)-N(2)	128.0(10)
C(26)-C(27)-H(27)	116
N(2)-C(27)-H(27)	116
C(28)-C(29)-N(2)	126.6(10)

C(28)-C(29)-H(29)	116.7
N(2)-C(29)-H(29)	116.7
C(24)#3-C(23)-C(24A)#3	71.0(16)
C(24)#3-C(23)-C(20)	120.9(17)
C(24A)#3-C(23)-C(20)	153.8(19)
C(25)-C(26)-C(27)	126.2(12)
C(27)-C(26)-C(25A)	109.9(10)
C(25A)-C(28)-C(29)	121.7(14)
C(29)-C(28)-C(25)	111.9(10)
C(23)#6-C(24)-C(25)	113.2(15)
C(26)-C(25)-C(28)	114.2(11)
C(26)-C(25)-C(24)	117.9(13)
C(28)-C(25)-C(24)	124.2(12)
C(23)#6-C(24A)-C(25A)	122(3)
C(28)-C(25A)-C(24A)	107.6(19)
C(28)-C(25A)-C(26)	119.8(12)
C(24A)-C(25A)-C(26)	118.4(15)

Table S7. Bond length of **2D-L-MOF.**

Atom 1- Atom 2	**Length (Å)**
Co(1)-O(1)	2.069(4)
Co(1)-O(3)#1	2.089(4)
Co(1)-O(6)	2.127(5)
Co(1)-O(5)	2.143(4)
Co(1)-N(2)#2	2.166(6)
Co(1)-N(1)	2.183(5)
O(1)-C(1)	1.293(7)
O(2)-C(1)	1.231(7)
O(3)-C(5)	1.296(7)
O(4)-C(5)	1.224(8)
O(5)-H(5A)	0.8501
O(5)-H(5B)	0.8499
O(6)-H(6A)	0.8499
O(6)-H(6B)	0.8501
N(1)-C(10)	1.342(8)
N(1)-C(14)	1.352(7)
N(2)-C(21)	1.391(8)
N(2)-C(19)	1.401(9)
C(1)-C(2)	1.559(9)
C(2)-C(3)	1.383(7)
C(2)-C(9)	1.411(8)
C(3)-C(4)	1.436(9)
C(3)-H(3)	0.9300
C(4)-C(6)	1.397(8)
C(4)-C(5)	1.536(8)

C(6)-C(7)	1.431(8)
C(6)-H(6)	0.9300
C(7)-C(9)	1.399(8)
C(7)-C(8)	1.557(7)
C(8)-H(8A)	0.9700
C(8)-H(8B)	0.9700
C(9)-H(9)	0.9300
C(10)-C(11)	1.413(10)
C(10)-H(10)	0.9300
C(11)-C(12)	1.399(10)
C(11)-H(11)	0.9300
C(12)-C(13)	1.330(10)
C(12)-C(15)	1.485(11)
C(13)-C(14)	1.367(9)
C(13)-H(13)	0.9300
C(14)-H(14)	0.9300
C(15)-C(16)	1.336(12)
C(15)-H(15)	0.9300
C(16)-C(17)	1.540(11)
C(16)-H(16)	0.9300
C(17)-C(18)	1.363(10)
C(17)-C(20)	1.449(11)
C(18)-C(19)	1.370(10)
C(18)-H(18)	0.9300
C(19)-H(19)	0.9300
C(20)-C(21)	1.390(10)
C(20)-H(20)	0.9300
C(21)-H(21)	0.9300

Table S8. Bond angle of **2D-L-MOF.**

Atom 1-Atom 2-Atom 3	**Angle (deg)**
O(1)-Co(1)-O(3)#1	91.37(15)
O(1)-Co(1)-O(6)	178.34(14)
O(3)#1-Co(1)-O(6)	89.93(18)
O(1)-Co(1)-O(5)	89.36(19)
O(3)#1-Co(1)-O(5)	177.90(14)
O(6)-Co(1)-O(5)	89.4(2)
O(1)-Co(1)-N(2)#2	89.72(17)
O(3)#1-Co(1)-N(2)#2	90.54(15)
O(6)-Co(1)-N(2)#2	89.24(18)
O(5)-Co(1)-N(2)#2	91.44(16)
O(1)-Co(1)-N(1)	89.86(16)
O(3)#1-Co(1)-N(1)	88.24(14)
O(6)-Co(1)-N(1)	91.20(17)
O(5)-Co(1)-N(1)	89.79(15)

N(2)#2-Co(1)-N(1)	178.70(18)
C(1)-O(1)-Co(1)	131.7(4)
C(5)-O(3)-Co(1)#1	130.0(4)
Co(1)-O(5)-H(5A)	108.9
Co(1)-O(5)-H(5B)	109.5
H(5A)-O(5)-H(5B)	104.5
Co(1)-O(6)-H(6A)	109.1
Co(1)-O(6)-H(6B)	108.7
H(6A)-O(6)-H(6B)	104.5
C(10)-N(1)-C(14)	111.6(6)
C(10)-N(1)-Co(1)	125.9(5)
C(14)-N(1)-Co(1)	122.5(4)
C(21)-N(2)-C(19)	115.9(6)
C(21)-N(2)-Co(1)#3	122.1(5)
C(19)-N(2)-Co(1)#3	121.5(5)
O(2)-C(1)-O(1)	127.1(7)
O(2)-C(1)-C(2)	118.4(6)
O(1)-C(1)-C(2)	114.4(5)
C(3)-C(2)-C(9)	120.5(6)
C(3)-C(2)-C(1)	119.7(5)
C(9)-C(2)-C(1)	119.8(5)
C(2)-C(3)-C(4)	120.0(6)
C(2)-C(3)-H(3)	120
C(4)-C(3)-H(3)	120
C(6)-C(4)-C(3)	119.5(6)
C(6)-C(4)-C(5)	120.2(7)
C(3)-C(4)-C(5)	120.3(5)
O(4)-C(5)-O(3)	126.7(6)
O(4)-C(5)-C(4)	118.8(6)
O(3)-C(5)-C(4)	114.5(6)
C(4)-C(6)-C(7)	120.1(6)
C(4)-C(6)-H(6)	119.9
C(7)-C(6)-H(6)	119.9
C(9)-C(7)-C(6)	119.2(5)
C(9)-C(7)-C(8)	121.5(5)
C(6)-C(7)-C(8)	119.3(5)
C(7)-C(8)-C(7)#4	116.2(7)
C(7)-C(8)-H(8A)	108.2
C(7)#4-C(8)-H(8A)	108.2
C(7)-C(8)-H(8B)	108.2
C(7)#4-C(8)-H(8B)	108.2
H(8A)-C(8)-H(8B)	107.4
C(7)-C(9)-C(2)	120.4(5)
C(7)-C(9)-H(9)	119.8
C(2)-C(9)-H(9)	`119.8
N(1)-C(10)-C(11)	127.0(7)

N(1)-C(10)-H(10)	116.5
C(11)-C(10)-H(10)	116.5
C(12)-C(11)-C(10)	116.3(7)
C(12)-C(11)-H(11)	121.8
C(10)-C(11)-H(11)	121.8
C(13)-C(12)-C(11)	118.0(7)
C(13)-C(12)-C(15)	120.5(7)
C(11)-C(12)-C(15)	121.5(7)
C(12)-C(13)-C(14)	121.0(7)
C(12)-C(13)-H(13)	119.5
C(14)-C(13)-H(13)	119.5
N(1)-C(14)-C(13)	125.9(7)
N(1)-C(14)-H(14)	117
C(13)-C(14)-H(14)	117
C(16)-C(15)-C(12)	125.1(9)
C(16)-C(15)-H(15)	117.4
C(12)-C(15)-H(15)	117.4
C(15)-C(16)-C(17)	125.8(9)
C(15)-C(16)-H(16)	117.1
C(17)-C(16)-H(16)	117.1
C(18)-C(17)-C(20)	119.0(7)
C(18)-C(17)-C(16)	118.2(7)
C(20)-C(17)-C(16)	122.4(7)
C(17)-C(18)-C(19)	118.3(7)
C(17)-C(18)-H(18)	120.9
C(19)-C(18)-H(18)	120.9
C(18)-C(19)-N(2)	125.5(7)
C(18)-C(19)-H(19)	117.2
N(2)-C(19)-H(19)	117.2
C(21)-C(20)-C(17)	120.1(7)
C(21)-C(20)-H(20)	119.9
C(17)-C(20)-H(20)	119.9
N(2)-C(21)-C(20)	120.9(7)
N(2)-C(21)-H(21)	119.6
C(20)-C(21)-H(21)	119.6

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