Supporting Information

Reentrant Spin-Glass Behavior in Cobalt (II) based Coordination Polymers

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Table S1. Crystallographic parameters for Compound **1-2**.

Parameters	Compound 1	Compound 2
Empirical formula	C_{26} H_{16} Co N_6 $O_{4.50}$	C_{28} H_{17} Co N_7 O_4
Formula weight	543.38	574.41
Crystal System	Monoclinic	Monoclinic
Space Group	C 2/c	C 2/c
a (Å)	15.43(7)	24.21(5)
b (Å)	12.84(6)	13.18(2)
c (Å)	24.90(11)	15.71(3)
α (º)	90	90
β (º)	99.43(5)	99.33(5)

γ (º)	90	90
Volume ($Å^3$)	4865(37)	4947(17)
Z	8	8
Calculated density (mg/m^3)	1.484	1.543
Absorption coefficient (mm^{-1})	0.754	0.746
θ range (^{0})	1.658 to 28.307	2.281 to 28.558
Reflections collected	28573	34680
Unique reflections	6025	6209
Number of parameters	339	352
Goodness-of-fit on F^2	0.996	1.016

Final R indices [I>2sigma(I)]	R_1 = 0.0466, wR_2 = 0.1040	R_1 = 0.0609, wR_2 = 0.1520
R indices (all data)	R_1 = 0.1022, wR_2 = 0.1362	R_1 = 0.1271, wR_2 = 0.1871

[a] $R_1 = \Sigma||F_0| - |F_c||/ \Sigma|F_0|$; $wR_2 = \{[w(F_0^2 - F_c^2)^2]/ [w(F_0^2)^2]\}^{1/2}$; $w = 1/[\sigma^2 (F_0)^2 + (aP)^2 + bP]$; $P = [max(F_0^2,0) + 2(F_c)^2]/3$, where a = 0.0620 and b = 0.00 for compound **1** and a = 0.0917 and b = 0.00 for compound **2**, respectively.

Table S2. Selected bond lengths (Å) and angles (^{0}) of Compound **1.**

Co(01)-O(2)#1	2.031(7)	O(1)-Co(01)-N(5)#3	89.0(3)
Co(01)-O(1)	2.033(6)	O(4)#2-Co(01)-N(5)#3	92.9(2)
Co(01)-O(4)#2	2.120(7)	N(6)-Co(01)-N(5)#3	177.39(8)
Co(01)-N(6)	2.165(7)	O(2)#1-Co(01)-O(3)#2	91.9(2)
Co(01)-N(5)#3	2.176(7)	O(1)-Co(01)-O(3)#2	150.98(10)
Co(01)-O(3)#2	2.229(8)	O(4)#2-Co(01)-O(3)#2	60.2(3)

Co(01)-C(26)#2	2.496(10)	N(6)-Co(01)-O(3)#2	93.1(3)
O(2)#1-Co(01)-O(1)	116.9(2)	N(5)#3-Co(01)-O(3)#2	89.1(3)
O(2)#1-Co(01)-O(4)#2	152.15(10)	O(2)#1-Co(01)-C(26)#2	121.78(14)
O(1)-Co(01)-O(4)#2	91.0(3)	O(1)-Co(01)-C(26)#2	121.3(2)
O(2)#1-Co(01)-N(6)	91.3(3)	O(4)#2-Co(01)-C(26)#2	30.38(13)
O(1)-Co(01)-N(6)	89.8(3)	N(6)-Co(01)-C(26)#2	91.3(3)
O(4)#2-Co(01)-N(6)	89.4(2)	N(5)#3-Co(01)-C(26)#2	91.3(3)
O(2)#1-Co(01)-N(5)#3	87.2(3)	O(3)#2-Co(01)-C(26)#2	29.86(17)

Table S3. Selected bond lengths (Å) and angles (^{0}) of Compound **2.**

Co(1)-O(5)#1	1.997(4)	O(6)#2-Co(1)-N(2)	87.83(18)
Co(1)-O(2)	2.004(4)	O(5)#1-Co(1)-N(6)#3	92.92(16)

Co(1)-O(6)#2	2.088(4)	O(2)-Co(1)-N(6)#3	89.35(18)
Co(1)-N(2)	2.163(4)	O(6)#2-Co(1)-N(6)#3	95.24(18)
Co(1)-N(6)#3	2.171(4)	N(2)-Co(1)-N(6)#3	176.89(12)
Co(1)-O(1)#2	2.343(5)	O(5)#1-Co(1)-O(1)#2	144.75(13)
O(5)#1-Co(1)-O(2)	121.06(16)	O(2)-Co(1)-O(1)#2	93.59(14)
O(5)#1-Co(1)-O(6)#2	87.61(19)	O(6)#2-Co(1)-O(1)#2	57.33(15)
O(2)-Co(1)-O(6)#2	150.75(14)	N(2)-Co(1)-O(1)#2	87.42(15)
O(5)#1-Co(1)-N(2)	87.65(16)	N(6)#3-Co(1)-O(1)#2	93.85(15)
O(2)-Co(1)-N(2)	87.74(18)		

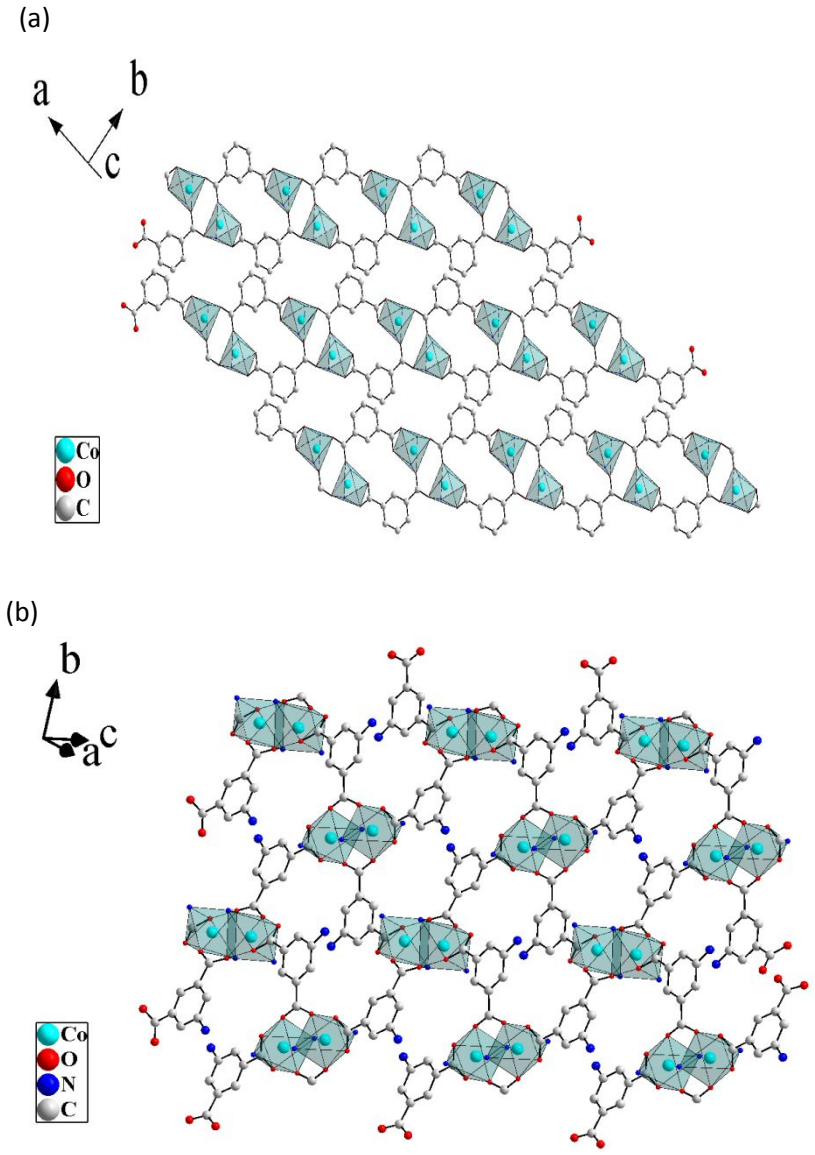

Figure S1. (a) Planar two-dimensional layered structure containing cobalt atom linked by IA ligand in **1** and (b) corrugated two-dimensional layered structure formed by AIA ligand coordinated with cobalt atom in **2**.

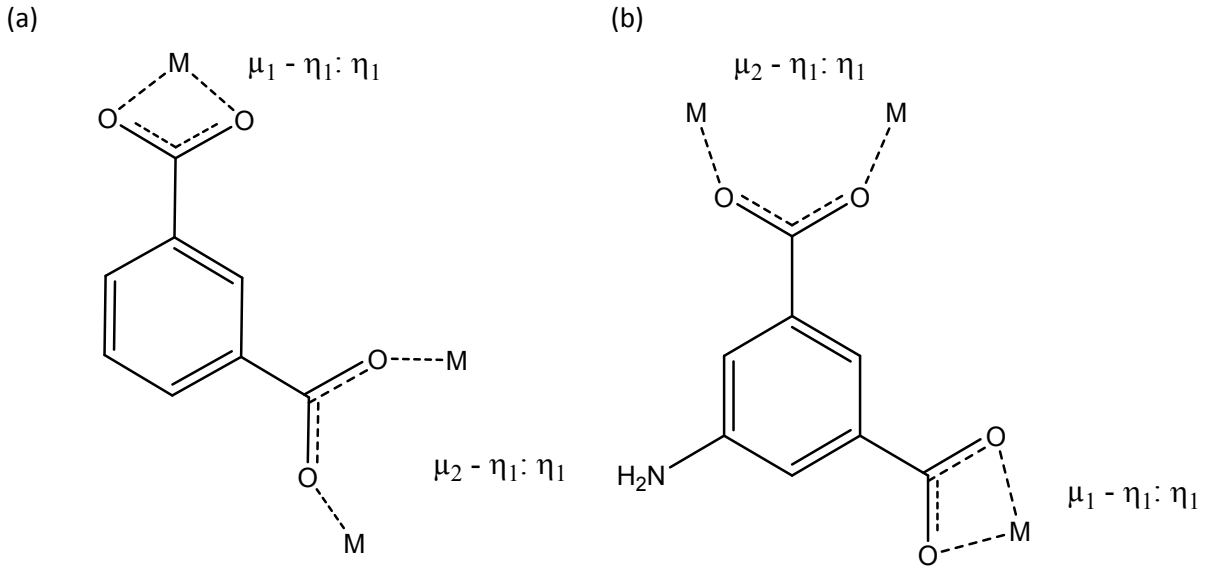

Figure S2. Different coordination modes of (a) isophthalate in **1** and (b) 5-aminoisopathalate ligand in **2**.

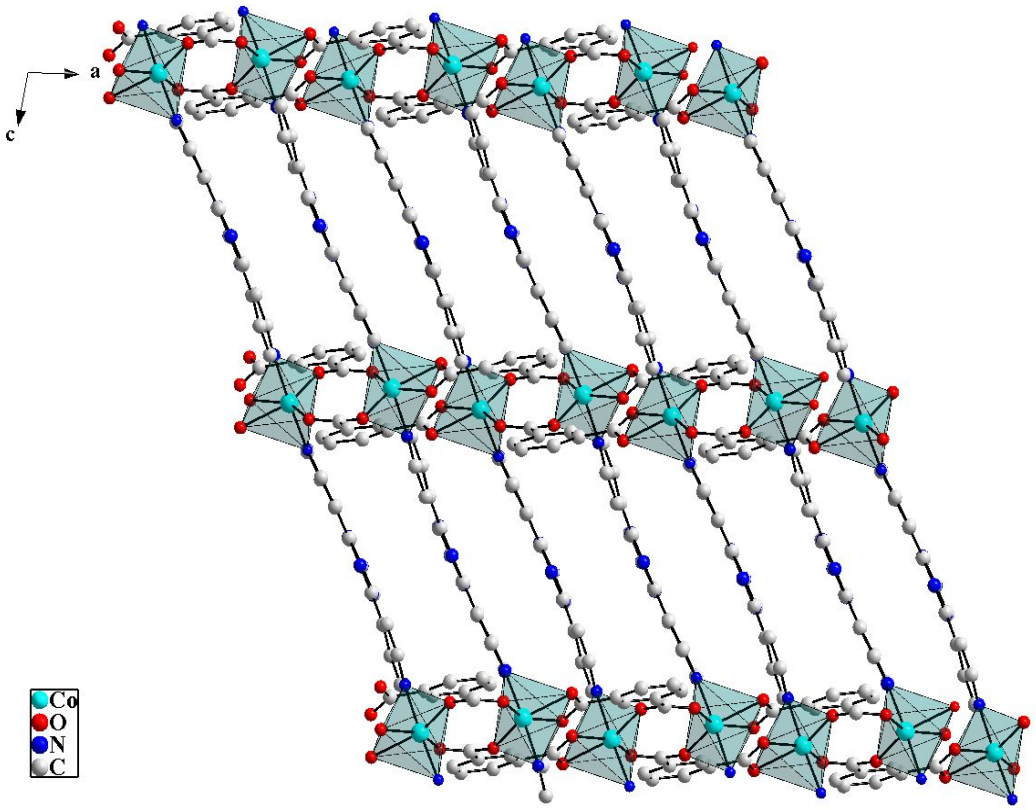

Figure S3. Three-dimensional architecture of **1**, where the two-dimensional layer containing cobalt atom and IA ligand, is pillared by TPT ligand.

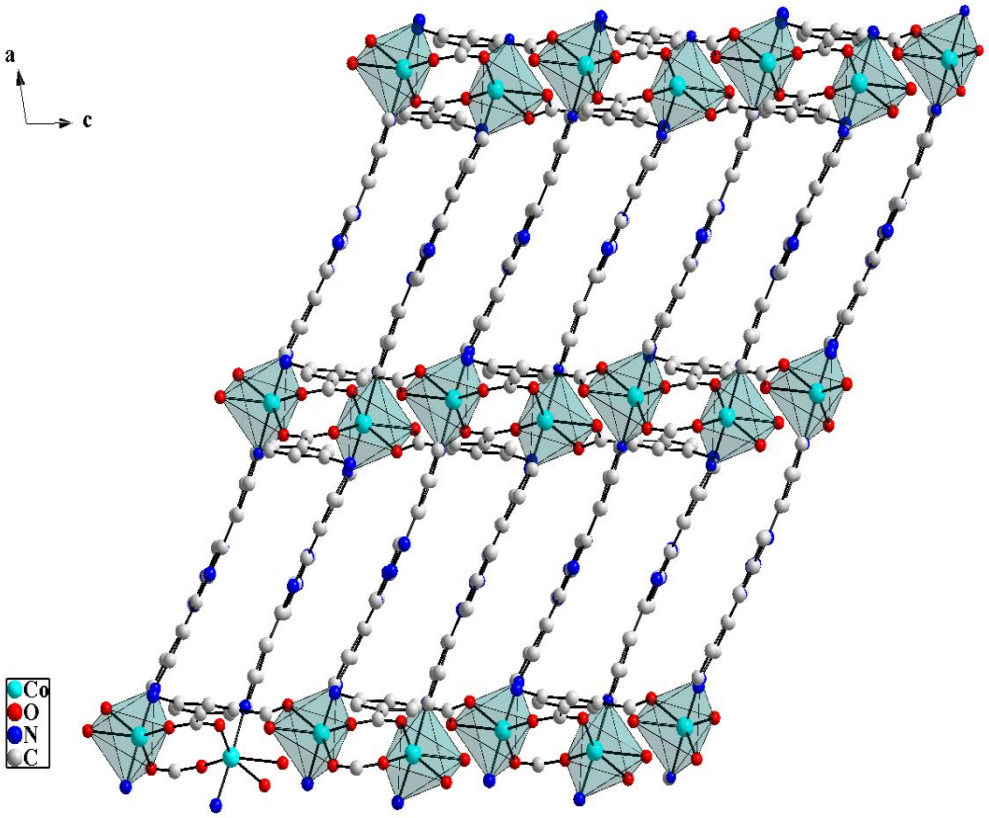

Figure S4. Three-dimensional architecture of **2**, where the two-dimensional layer containing cobalt atom and AIA ligand is, pillared by TPT ligand.

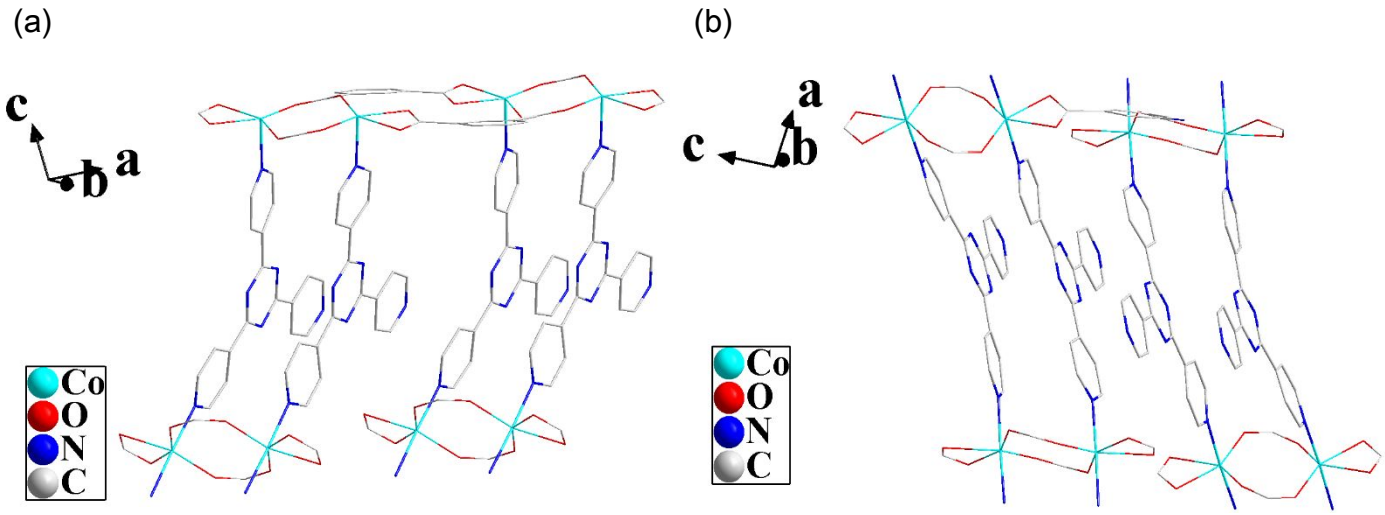

Figure S5. The co-parallel orientation of two consecutive TPT ligand connected to adjacent octamer unit (a) in **1** and antiparallel orientation (b) in **2**; and co-parallel orientation of two TPT ligands coordinated to cobalt atoms of same octamer unit in both **1** and **2**.

TOPOS OF COMPOUND 1

##########################

Topology for Co1

Atom Co1 links by bridge ligands and has

Common vertex with					R(A-A)	f
Co 1	0.6203	1.2480	0.5141	(0 0 0)	10.037A	1
Co 1	-0.3797	0.2480	0.5141	(-1-1 0)	10.037A	1
Co 1	0.6203	0.7520	1.0141	(0 1 0)	13.530A	1
Co 1	-0.3797	0.7520	0.0141	(-1 1-1)	13.530A	1
Common edge with					R(A-A)	
Co 1	0.3797	0.7520	0.4859	(0 1 1)	4.176A	2
Co 1	-0.1203	0.2520	0.4859	(0 1 1)	7.346A	2

Structural group analysis

Structural group No 1

Structure consists of 3D framework with CoO4N6C26H16

Coordination sequences

Co1: 1 2 3 4 5 6 7 8 9 10

Num 6 20 53 107 177 263 365 483 617 767

Cum 7 27 80 187 364 627 992 1475 2092 2859

Rad 9.8(3.6) 17.4(4.2) 23.3(6.8) 29.8(8.7) 37.2(9.8) 45.0(11.0) 52.9(12.2) 60.7(13.5) 68.6(14.9) 76.5(16.3)

Cmp Co6 Co20 Co53 Co107 Co177 Co263 Co365 Co483 Co617 Co767

TD10=2859

Vertex symbols for selected sublattice

Co1 Point symbol:{3^3.4^4.5^5.6^3}

Extended point symbol:[3.3.3.4.4.4.4.5.5.5.5.5.6(2).6(2).6(4)]

Point symbol for net: {3^3.4^4.5^5.6^3}

6-c net; uninodal net

TOPOS OF COMPOUND 2

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1:C28 H17 Co N7 O4

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Topology for Co1

Atom Co1 links by bridge ligands and has

Common vertex with				R(A-A)	f
Co 1	0.2666	-0.2357	0.8783 (0-1 1)	7.699A	1
Co 1	0.2666	0.7643	0.8783 (0 0 1)	7.699A	1
Co 1	0.2334	0.7357	0.1217 (0 1-1)	10.016A	1

Co 1 0.2334 0.7357 1.1217 (0 1 0) 10.016A 1

Co 1 0.7334 0.2357 1.1217 (0 0 0) 13.325A 1

Co 1 -0.2666 0.2357 0.1217 (-1 0-1) 13.325A 1

Common edge with R(A-A)

Co 1 0.2666 0.2357 0.3783 (0 0 1) 4.051A 2

Structural group analysis

Structural group No 1

Structure consists of 3D framework with CoO4N7C26H17

Coordination sequences

Co1: 1 2 3 4 5 6 7 8 9 10

Num 7 27 58 105 163 236 321 421 532 659

Cum 8 35 93 198 361 597 918 1339 1871 2530

Rad 9.4(3.3) 15.7(4.1) 22.8(5.6) 30.0(7.2) 37.4(8.7) 44.8(10.3) 52.2(12.0) 59.7(13.6) 67.2(15.3) 74.6(17.0)

Cmp Co7 Co27 Co58 Co105 Co163 Co236 Co321 Co421 Co532 Co659

TD10=2530

Vertex symbols for selected sublattice

Co1 Point symbol:{3^3.4^9.5^8.6}

Extended point symbol:[3.3.3.4.4.4.4.4.4.4.4.4.5.5.5.5(2).5(2).5(2).5(2).5(3).6(5)]

Point symbol for net: {3^3.4^9.5^8.6}

7-c net; uninodal net

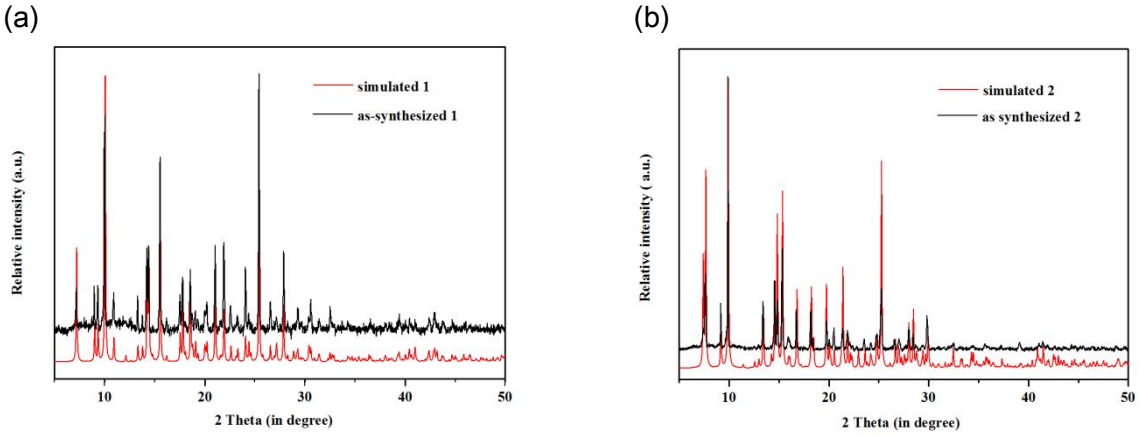
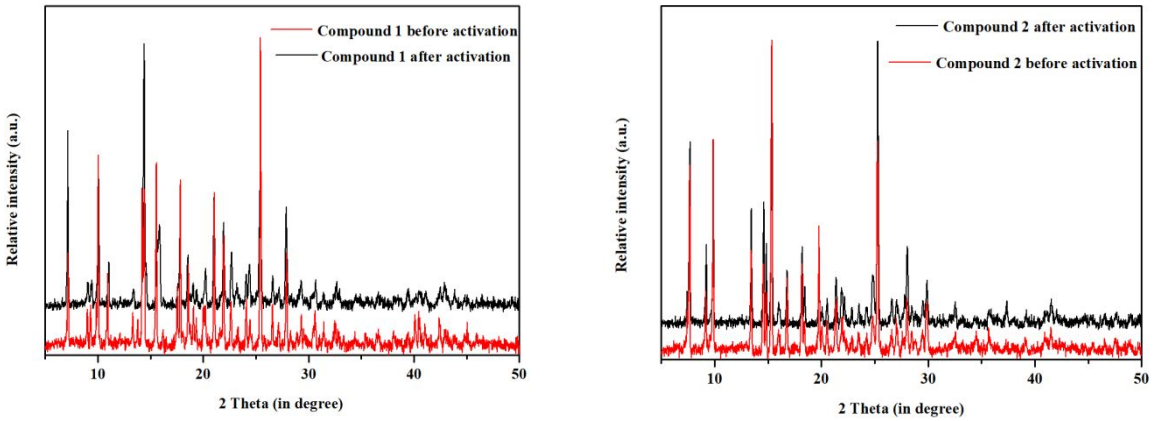


Figure S6. Simulated and experimental PXRD data of (a) **1** and (b) **2**.

Gas sorption studies

Gas sorption measurements of **1** and **2** were carried out on a Micromeritics' 3Flex 3500 BET-surface area analyzer. Ultra-high purity N_2 (99.99 %) was used in the sorption experiments. Around 100 mg of the synthesized sample was taken and activated at 423 K for 4 h under vacuum before gas sorption measurement. The PXRD data was confirmed the phase purity of **1** and **2**, before and after activation under above-mentioned condition.



(a) (b)

Figure S7. PXRD data of (a) **1** and (b) **2** before and after activation at 423 K for 4 h under vacuum.

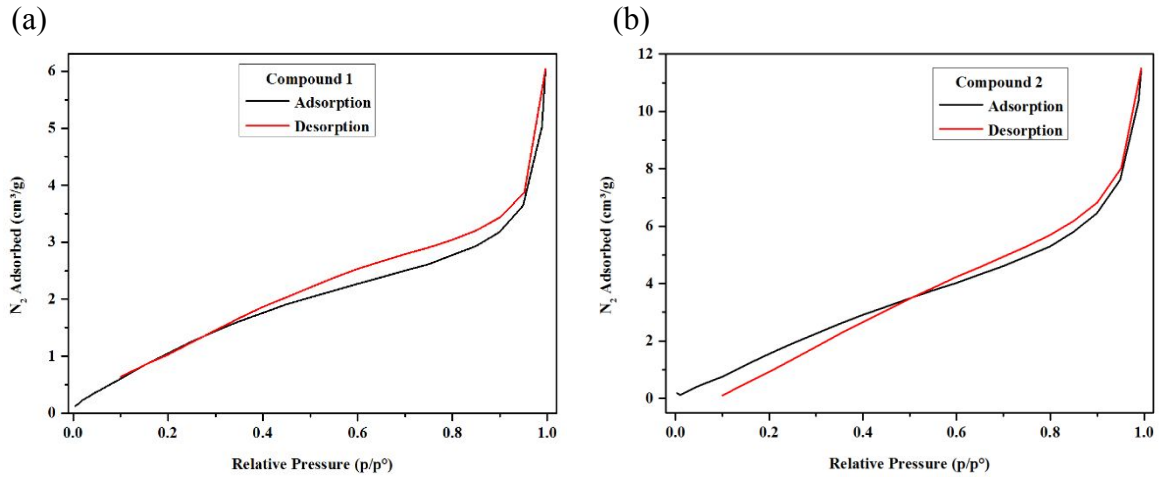


Figure S8. N_2 - adsorption isotherm for (a) **1** and (b) **2**.

Table S4. Relevant data of the N_2 adsorption isotherm.

Compound 1	
Single point surface area at p/p° = 0.149064058	3.0972 m²/g
BET surface area	5.7573 m²/g
Adsorption cumulative surface area of pores between 1.7 nm and 300 nm width	4.9466 m²/g
Single point adsorption total pore volume of pores less than 345.8368 nm width at p/p° = 0.994441056	0.008961 cm³/g
Compound 2	
Single point surface area at p/p° = 0.149040868	4.2810 m²/g
BET surface area	11.3707 m²/g

Adsorption cumulative surface area of pores between 1.7 nm and 300 nm width	7.5032 m^2/g
Single point adsorption total pore volume of pores less than 324.2608 nm width at p/p° = 0.994067918	0.017843 cm^3/g

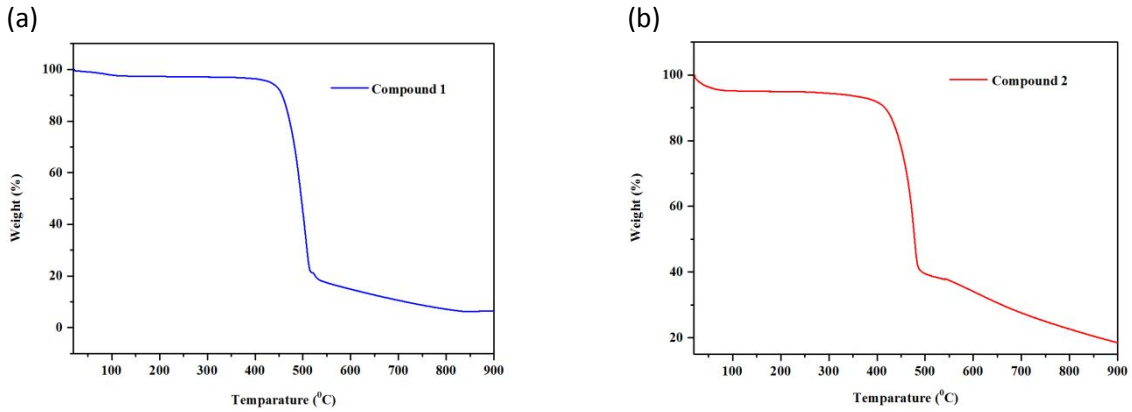


Figure S9. Thermogravimetric analysis of (a) **1** and (b) **2**.

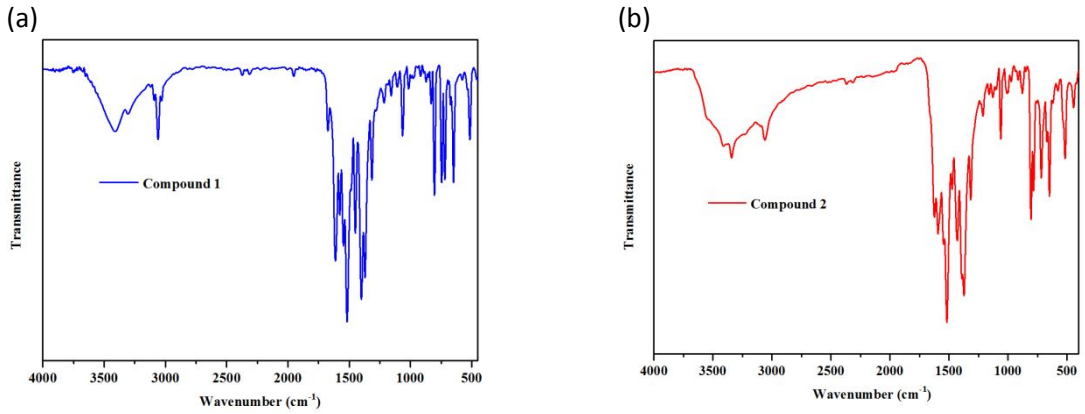


Figure S10. IR-spectra of (a) **1** and (b) **2**.

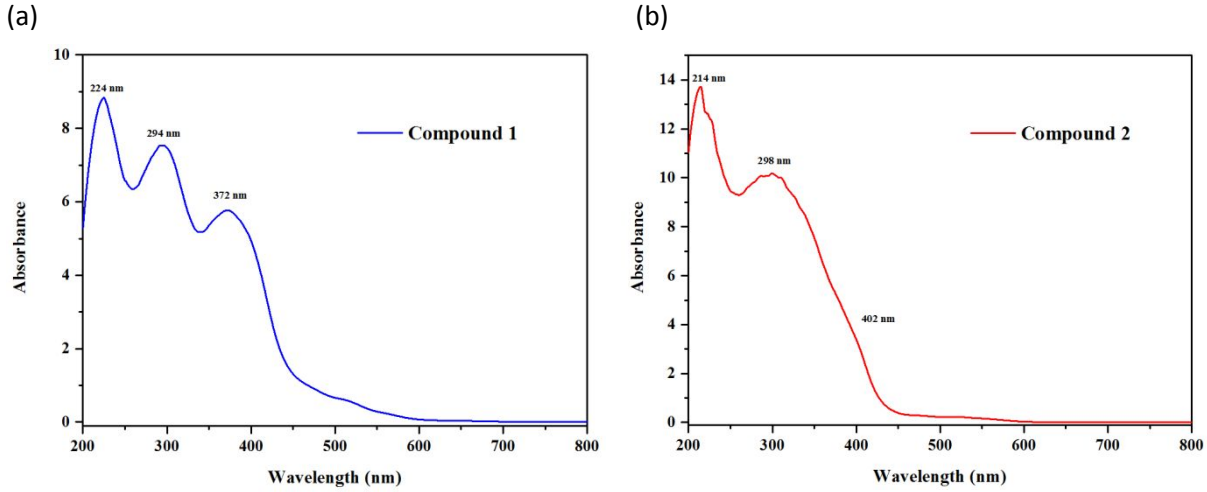


Figure S11. The UV-Vis absorption spectrum of (a) **1** and (b) **2**.

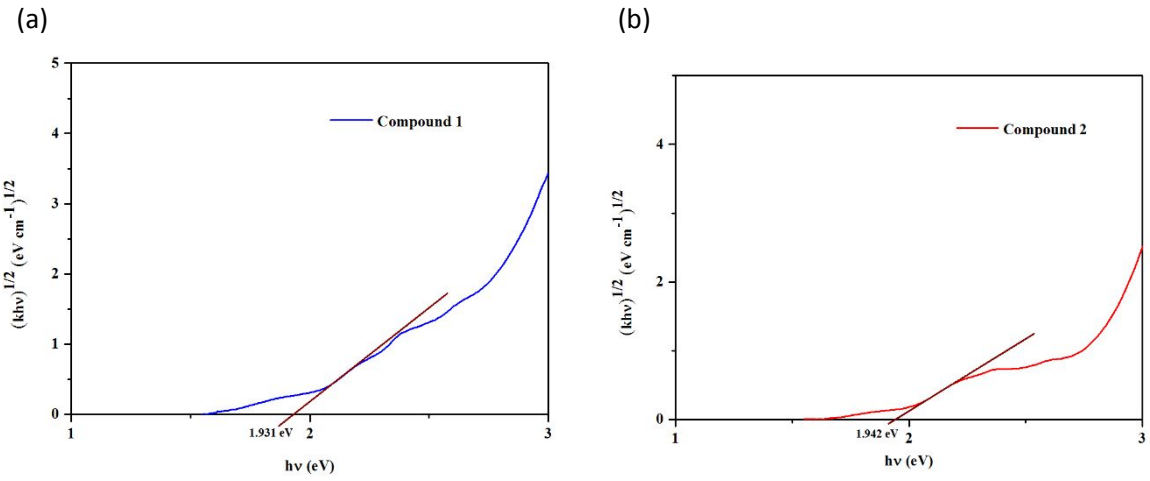
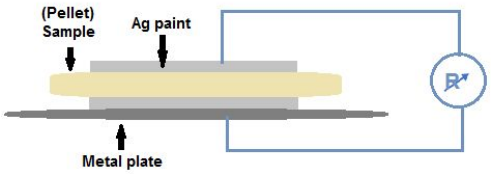


Figure S12. Tauc plot of (a) **1** and (b) **2**.

Temperature dependence of resistivity measurement

The electrical resistance of **1** and **2** with varying temperature was measured using Keithley 6517B electrometer with Linkam temperature controller. Both the compounds were gently grind using mortar-pestle and then pelletized (thickness of 1 mm and radius of 5 mm). For making contacts, both sides of the pellet were coated with the silver paint and then the pellet was placed above the metal plate. The measurements



were performed using 80 V potential and 3 ºC/min ramp. The readings were taken after being stabilized the temperature for 4 min at every temperature point during the experiment.

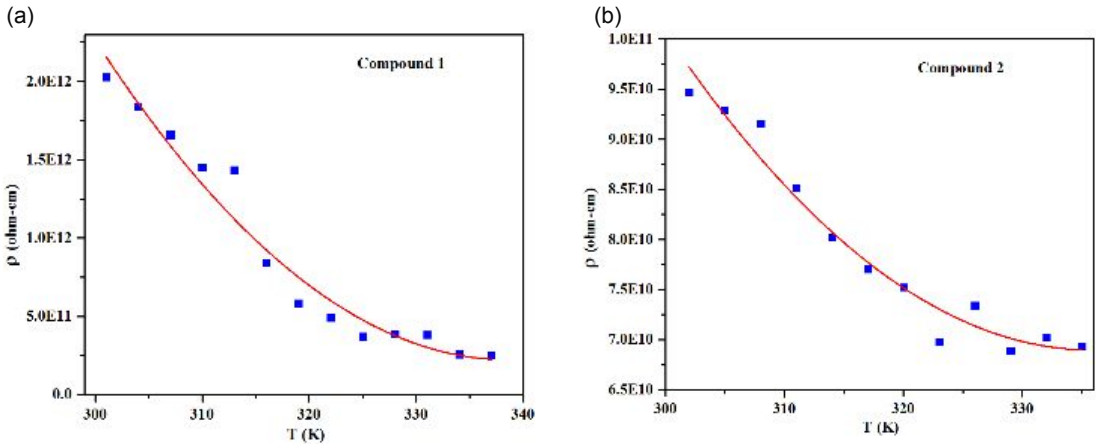


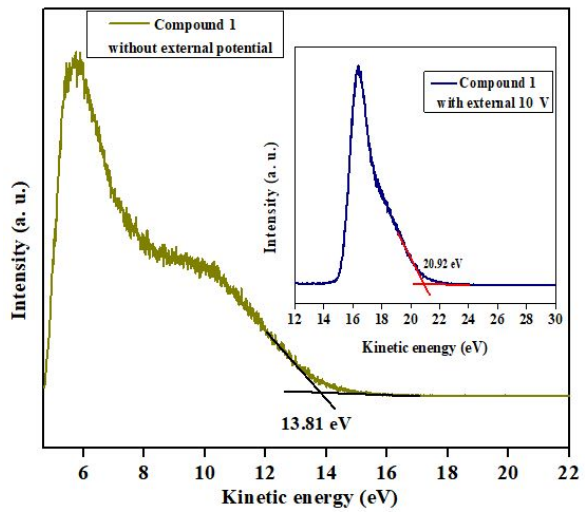
Figure S13. Temperature-dependent resistivity plots (ρ vs. T) of (a) **1** and (b) **2**.

Table S5. Relevant instrumental parameters for UPS measurement.

Parameters	Compound 1 (0 V)	Compound 1 (10 V)	Compound 2 (0 V)	Compound 2 (10 V)
Energy Mode	KE	KE	KE	KE
Start Energy (eV)	22	30	22	22
End Energy (eV)	4.72	12	5.2	5.2
Energy Step (eV)	-0.01	-0.01	-0.05	-0.025
Number Of Sweeps	10	10	1	10
Dwell Time per Sweep (sec)	0.1	0.1	0.1	0.1
Analyser Mode	CAE	CAE	CAE	CAE
Pass Energy (eV) /Retard Ratio	0.2	1	0.65	0.65
Magnification	High	High	High	High
Divide Voltages By 10	On	On	On	On
Excitation Source	He I a 21.2 eV	He I a 21.2 eV	He I a 21.2 eV	He I a 21.2 eV
Excitation Energy (eV)	21.2 eV	21.2 eV	21.2	21.2

Analyser Workfunction (eV)	4.5 eV	4.5 eV	4.5 eV	4.5 eV
Aperture	2	2	2	2
Exit Slit (nm)	5x11	5x11	5x11	5x11

(a)



(b)

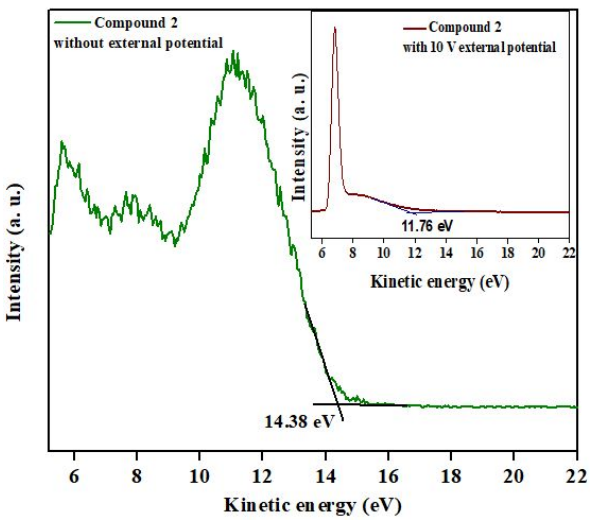


Figure S14. UPS spectrum of **1** and **2** without external potential. Inset: UPS spectrum of **1** and **2** with 10 V external potential.