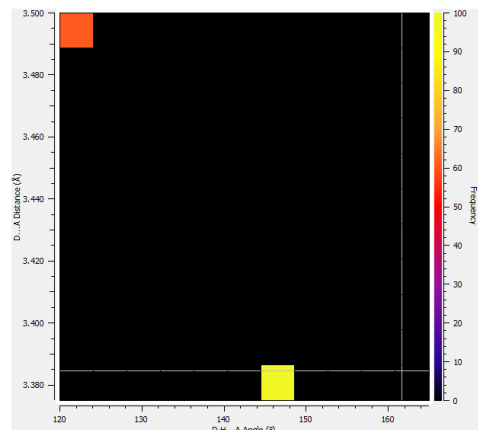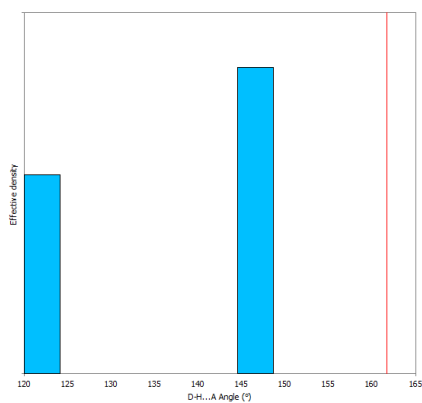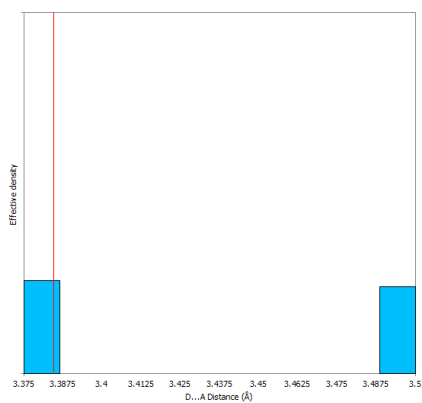# Supporting Information

SI-1: Distance and Angle Comparison of hydrogen bonds within **1** (CSD-Materials/Mercury; CSD v5.44, Apr 2023)

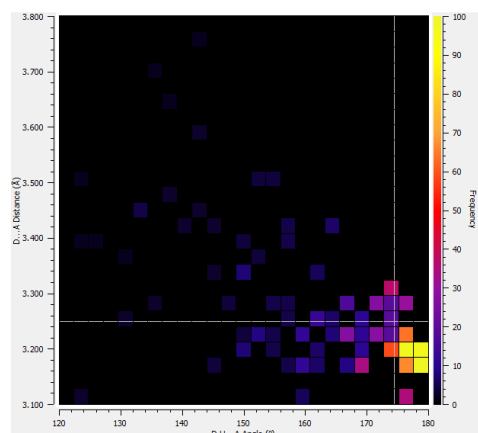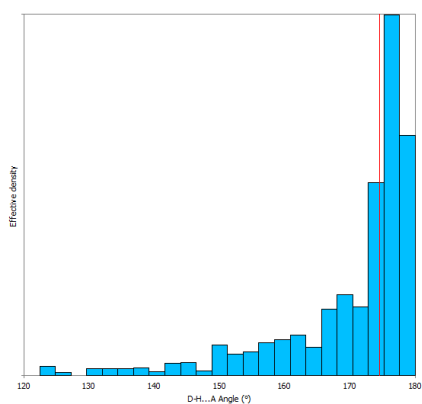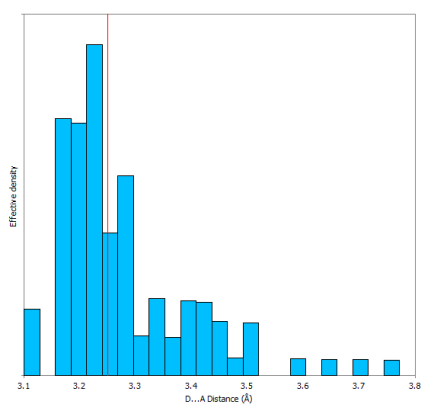| Donor	Acceptor	Distance D-A	Distance classification	Distance threshold	Distance hits	Distance mean	Distance std. dev.	Distance min.	Distance max.	Angle D-H...A	Angle classification	Angle threshold	Angle hits	Angle mean	Angle std. dev.	Angle min.	Angle max.
N2 (amine)	S1 [1] (CdoubleS)	**3.38**	Not Unusual	(3.38, 3.49)	2	3.43	0.08	3.38	3.49	**161.69**	Not Unusual	123.81	2	135.46	18.31	122.52	148.41
O2 (al_hydroxy)	S1 [2] (CdoubleS)	**3.25**	Not Unusual	(3.19, 3.51)	87	3.29	0.13	3.11	3.77	**174.54**	Not Unusual	132.65	87	166.51	15.35	122.51	179.39

[1] 1-x,1-y,1-z; [2] -1+x,+y,+z

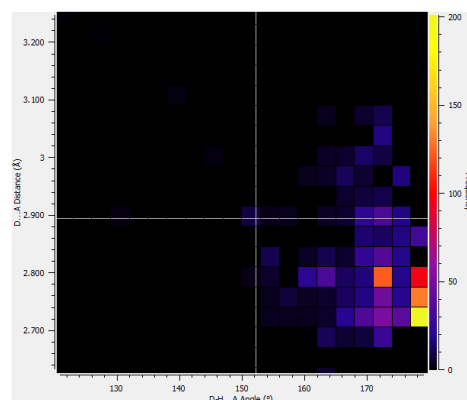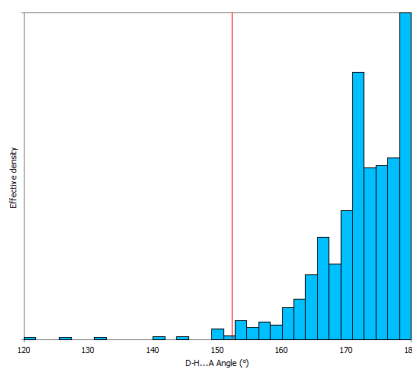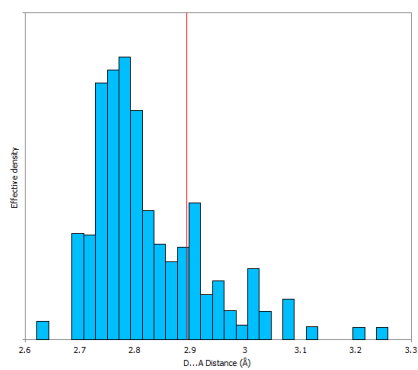**N2-H2···O3:** | Distance | Angle | Frequency | to similar examples on the CSD

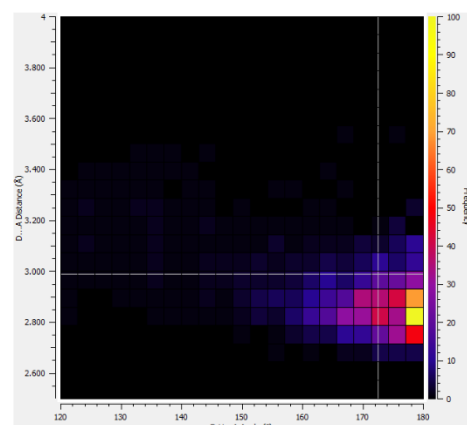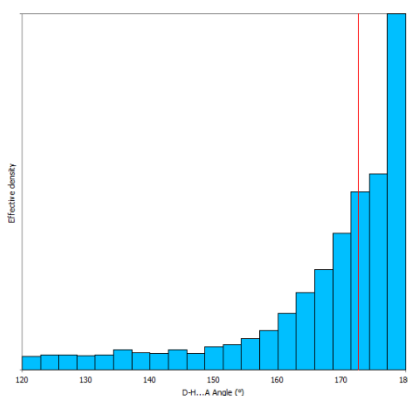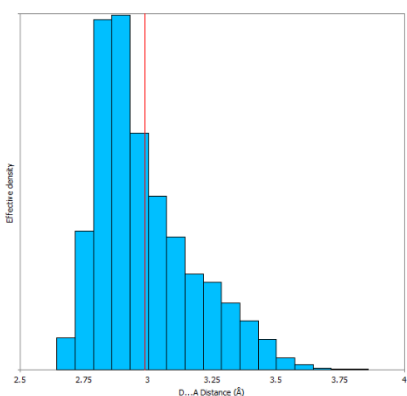**O2-H2a···O3:** | Distance | Angle | Frequency | to similar examples on the CSD

Donor	Acceptor	Distance D-A	Distance classification	Distance threshold	Distance hits	Distance mean	Distance std. dev.	Distance min.	Distance max.	Angle D-H...A	Angle classification	Angle threshold	Angle hits	Angle mean	Angle std. dev.	Angle min.	Angle max.
N2 (amine_5_1)	O3 (water)	**2.89**	Not Unusual	(2.71, 3.02)	132	2.82	0.11	2.63	3.25	**152.24**	Not Unusual	151.36	132	170.29	10.1	120.37	179.21
O3 (water)	O2 (T2OH0)	**2.99**	Not Unusual	(2.77, 3.37)	2092	2.99	0.19	2.64	3.79	**172.58**	Not Unusual	134.48	2092	168.41	13.85	120.03	179.96
N4 (amine_5_1)	S1 (CdoubleS)	**3.41**	Not Unusual	(3.38, 3.49)	2	3.43	0.08	3.38	3.49	**151.7**	Not Unusual	123.81	2	135.46	18.31	122.52	148.41
O3 (water)	S1 (CdoubleS)	**3.8**	Unusual	(3.15, 3.57)	224	3.33	0.11	2.93	3.71	**163.57**	Not Unusual	139.8	224	169.01	12.23	122.5	179.61

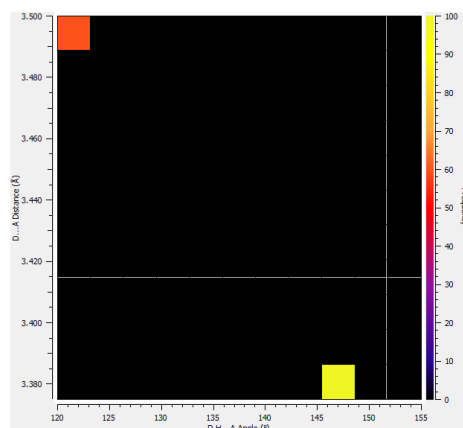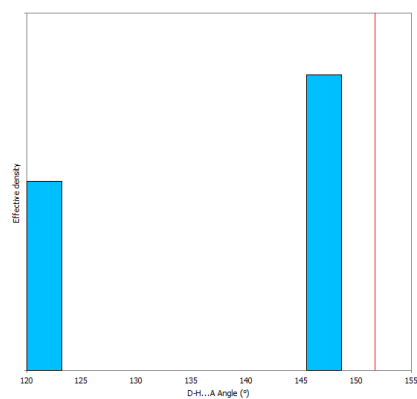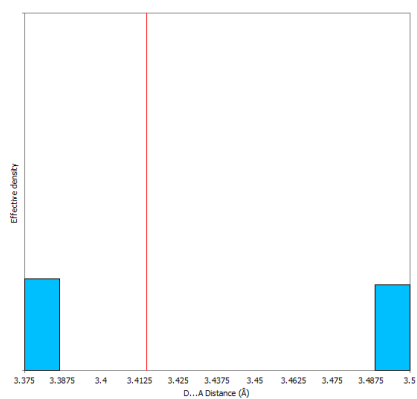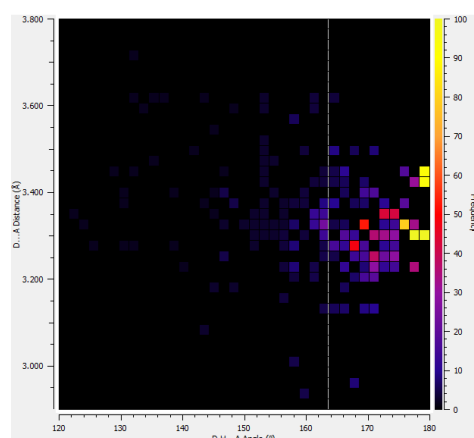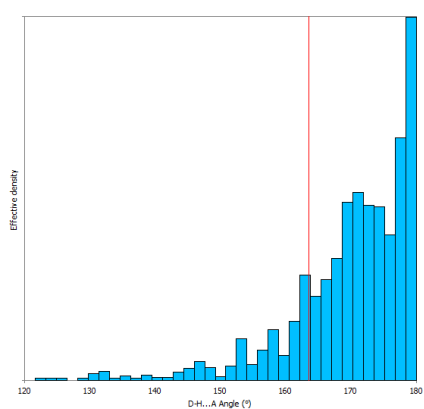**N2-H2···O3:** | Distance | Angle | Frequency | to similar examples on the CSD

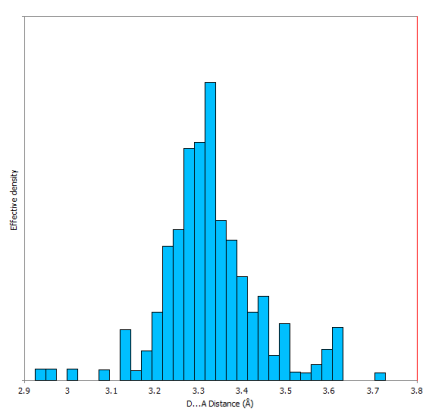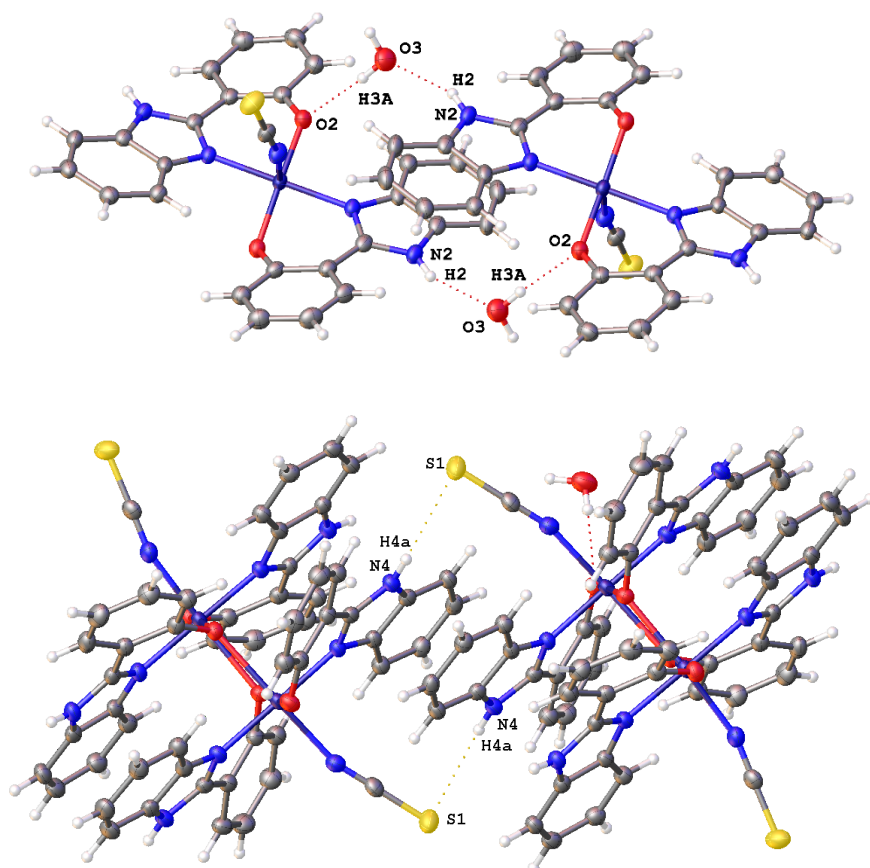**O3-H3a···O2:** | Distance | Angle | Frequency | to similar examples on the CSD

N4-H4a···S1: | Distance | Angle | Frequency | to similar examples on the CSD

**O3-H3b···S1:** | Distance | Angle | Frequency | to similar examples on the CSD

Views of Complex 2 highlighting hydrogen bonding (thermal ellipsoids drawn at 50% probability)

^1H and ^{13}C NMR acquired in DMSO-d_6 or CDCl$_3$ on BRUKER AVANCE 500 spectrometer, using TMS as an internal standard. UV-vis spectra recorded using a SCINCO S-3100 spectrophotometer with a photodiode (PDA) detector. FT-IR spectra obtained upon a PerkinElmer Spectrum Two instrument (DTGS). Molar conductivity measurements performed using an EC-214 conductivity meter at at room temperature and using a cell containing a millimolar solution of dimethylformamide.
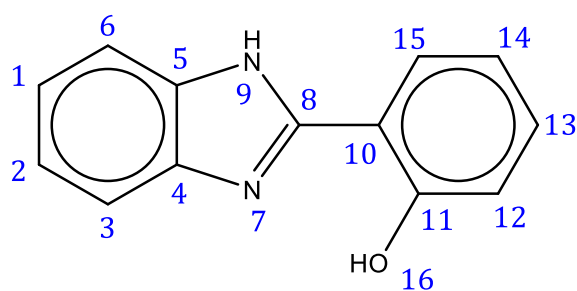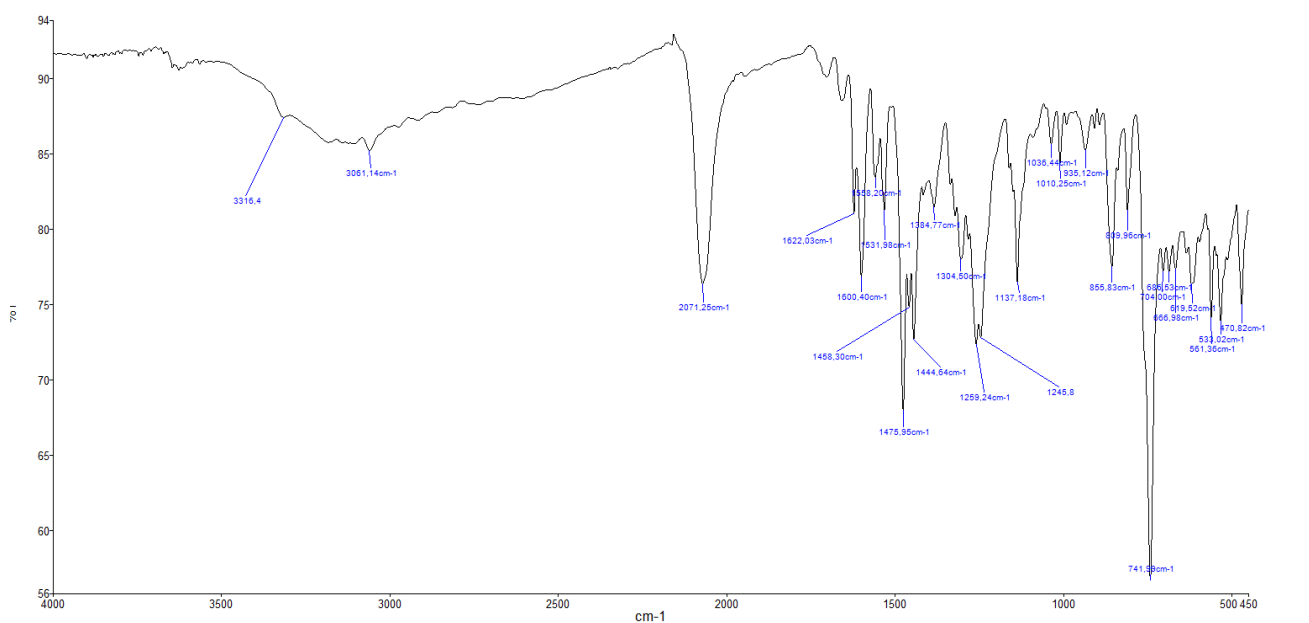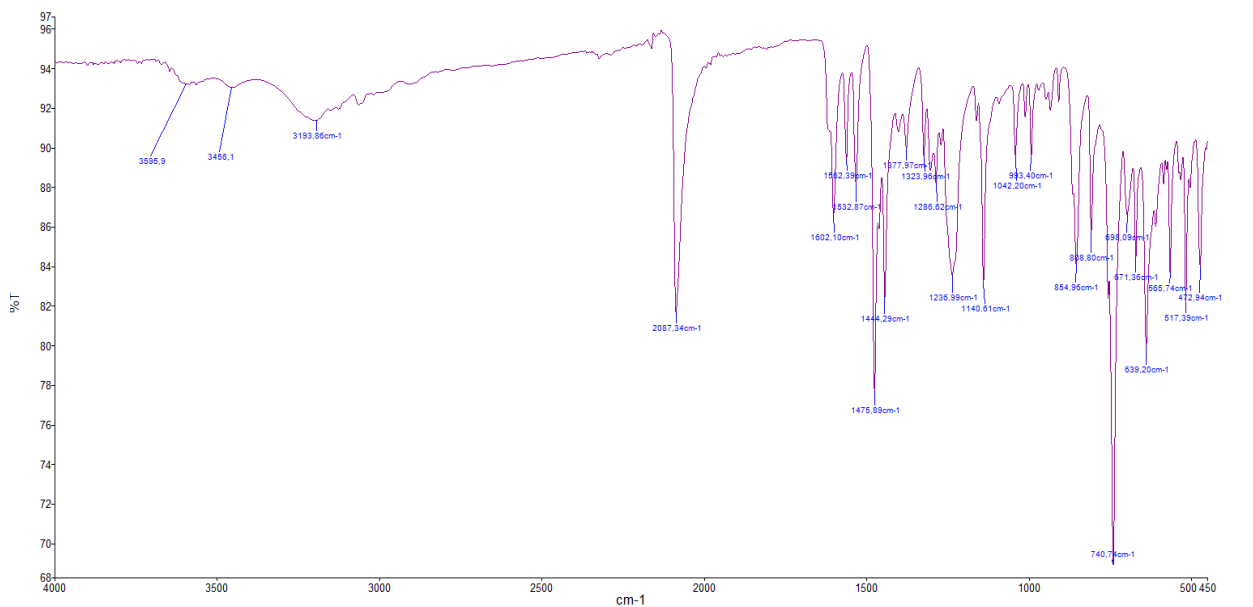

2hpbi ligand

Infrared spectroscopy: 2hpbi ligand, (2-(1H-benzimidazol-2-yl)phenol ($C_{13}H_{10}N_2O$)
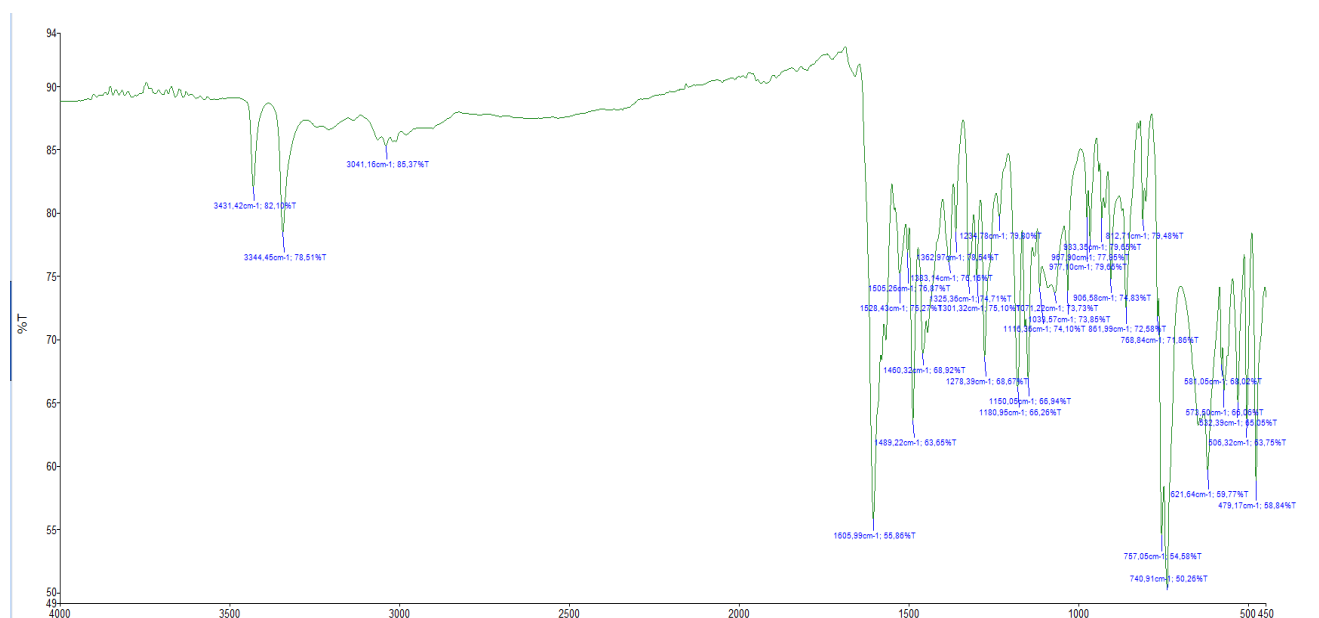
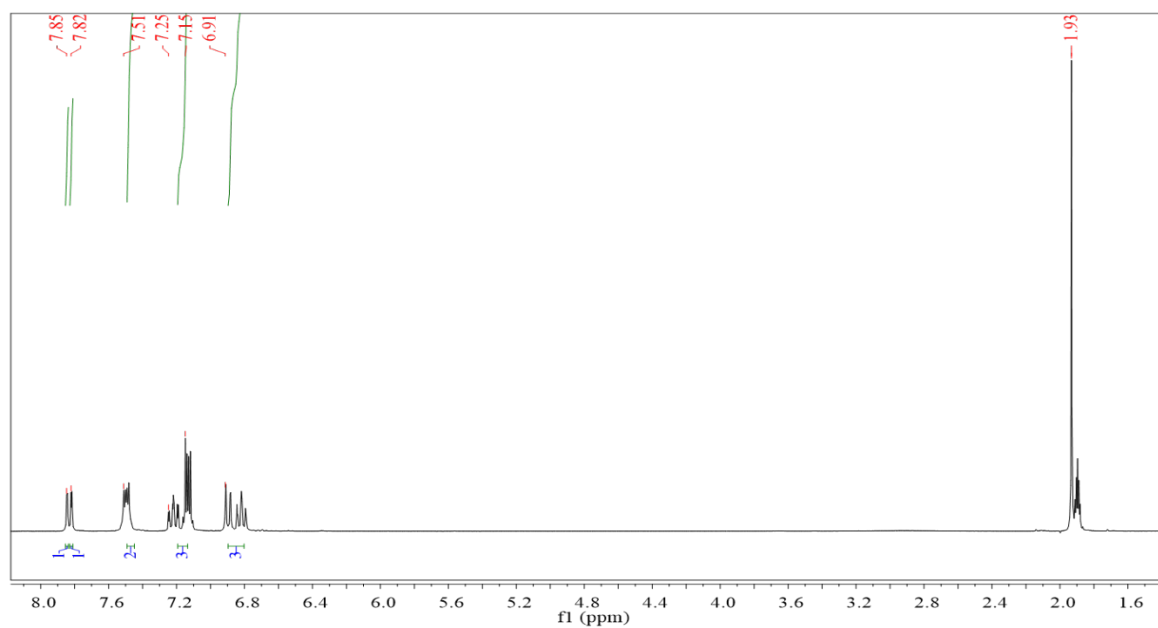**Infrared spectroscopy:** Complex **1**, $[Co_2(C_{13}H_9N_2O)_2(SCN)_2(CH_3CH_2OH)_2]$

Infrared spectroscopy: Complex **2** $[Mn_2(C_{13}H_9N_2O)_4(SCN)_2].2(H_2O)$

¹H NMR: 2hpbi ligand, (2-(1H-benzimidazol-2-yl)phenol (C₁₃H₁₀N₂O)

¹³C NMR: 2hpbi ligand, (2-(1H-benzimidazol-2-yl)phenol (C₁₃H₁₀N₂O)
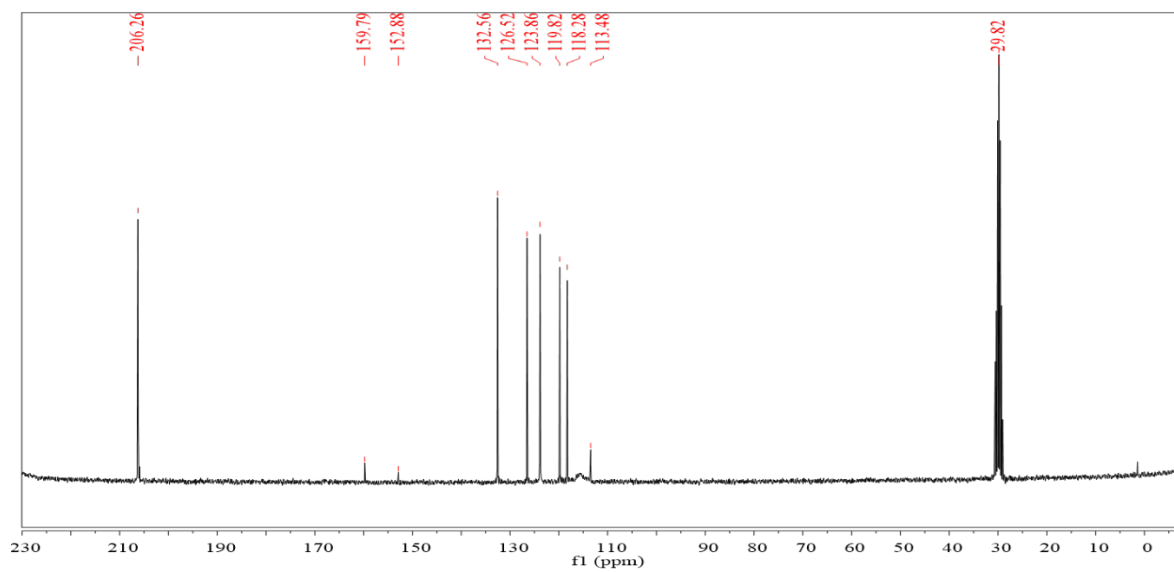
¹³C NMR (DEPT 135): 2hpbi ligand, (2-(1H-benzimidazol-2-yl)phenol ($C_{13}H_{10}N_2O$)
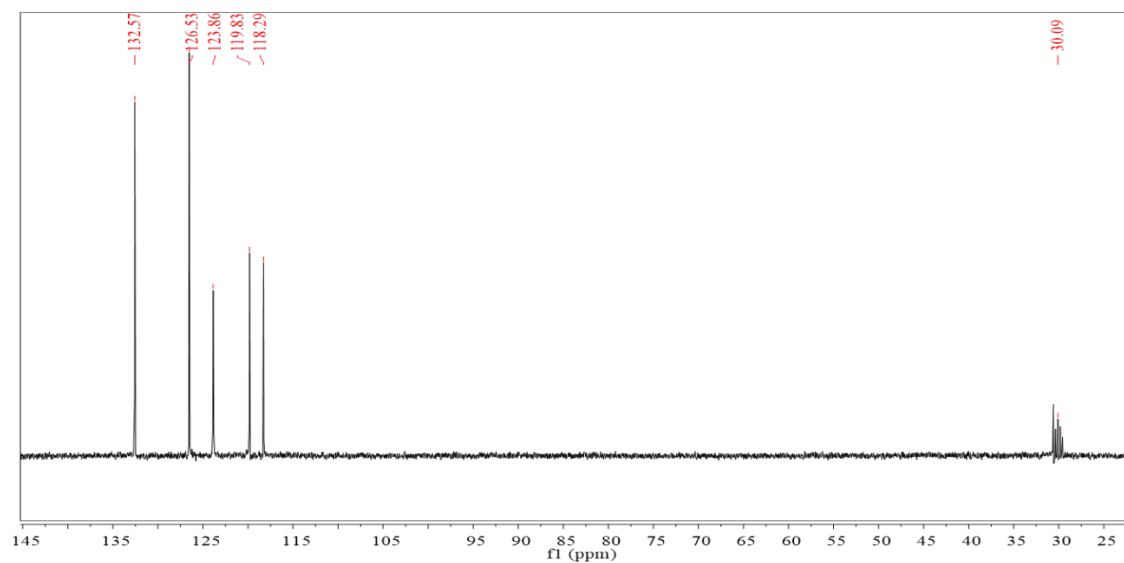

Conductivity study: Complexes **1** and **2**

	Cold Conductivity (μS/cm)	Conductivity after 15 days (μS/cm)	Electrolytes
Complex 1	19,26	17,21	Neutral
Complex 2	18,44	16,95	Neutral

UV-Vis Spectroscopy: Complex **1**, [$Co_2(C_{13}H_9N_2O)_2(SCN)_2(CH_3CH_2OH)_2$]
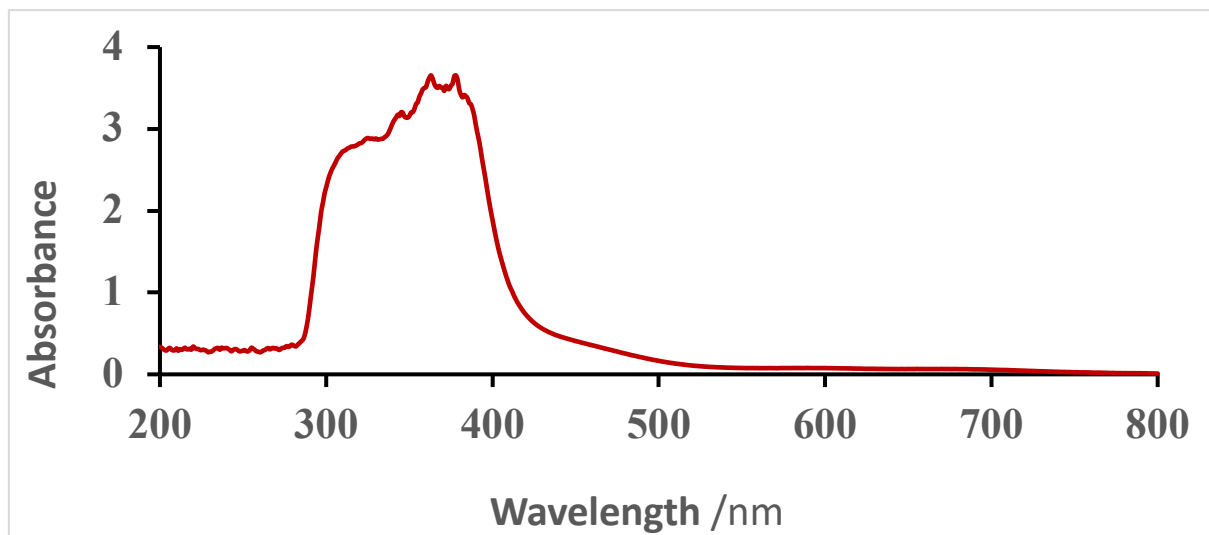
UV-Vis Spectroscopy: Complex **2** [$Mn_2(C_{13}H_9N_2O)_4(SCN)_2$].$2(H_2O)$
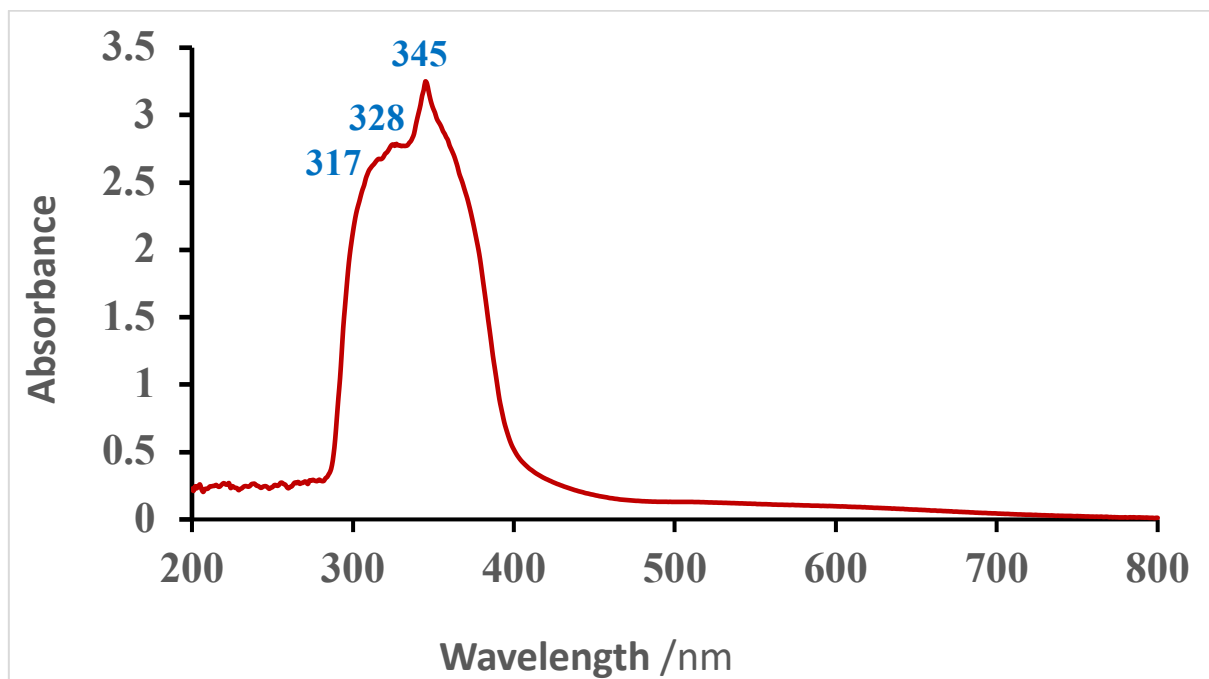
SI-4: Complex **1** coordination polyhedron geometry index

The geometry around the penta-coordinated cobalt (II) ion is deduced from the trigonality or Addison index. This is defined by the relation $\tau = (\beta - \alpha)/60$ which is applicable to five-co-ordinate structures as an index of the degree of trigonality. β and α are the greatest angles of connection around Co(II). If $\tau = 0$ the geometry is a perfect square pyramid while a value of $\tau = 1$ refers to a trigonal bipyramidal geometry. The greatest angles around Co(II) are $\beta=169.020°$ for N1-Co1-O1^i and $\alpha=119.680°$ for O2-Co-O1. The value thus found is $\tau = 0.82$ confirms a slightly deformed trigonal-bipyramidal geometry for complex.