Table S1. Crystallographic Table for compounds **4** – **21** and **H_2L3** and **H_2L19**.

	4	**5**	**6**	**7**	**8**
Empirical formula	$C_{36}H_{44}CoN_5O_{12}$	$C_{45}H_{52}Br_2CoN_3O_9$	$C_{120.5}H_{115}Cl_5Dy_2N_4Ni_2O_{20}$	$C_{122}H_{146}Cl_2Dy_2N_{14}Ni_2O_{30}$	$C_{112}H_{114}Br_6Dy_2N_4Ni_2O_{26}$
Formula weight	797.69	997.64	2558.83	2801.84	2848.01
Temperature/K	100	173	173	173	173
Crystal system	monoclinic	monoclinic	monoclinic	triclinic	monoclinic
Space group	$P2_1/c$	$P2_1/n$	C2/c	P-1	C2/c
a/Å	18.8624(5)	13.0590(8)	22.1776(15)	12.5403(7)	20.4620(7)
b/Å	17.0300(4)	22.7258(11)	18.8940(11)	14.7915(10)	20.8069(7)
c/Å	25.6461(18)	15.4472(13)	27.8333(11)	18.880(2)	27.3553(7)
α/°	90	90	90	100.291(8)	90
β/°	108.353(8)	109.616(8)	100.127(4)	106.198(7)	98.444(3)
γ/°	90	90	90	105.143(5)	90
Volume/Å^3	7819.2(7)	4318.3(5)	11481.1(11)	3124.5(5)	11520.3(7)
Z	8	4	4	1	4
ρ_{calc}g/cm^3	1.355	1.535	1.48	1.489	1.592
μ/mm^{-1}	0.505	2.307	1.795	1.602	3.754
F(000)	3344	2048	5196	1438	5472
Crystal size/mm^3	0.24 × 0.13 × 0.02	0.12 × 0.1 × 0.06	0.1 × 0.05 × 0.02	0.38 × 0.32 × 0.24	0.12 × 0.08 × 0.03
Radiation	MoKα (λ = 0.71075)	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)
2Θ range for data collection/°	6.104 to 54.962	4.88 to 52.742	5.564 to 58.632	4.956 to 58.074	5.06 to 58.266
Index ranges	-24 ≤ h ≤ 24, -19 ≤ k ≤ 22, -26 ≤ l ≤ 33	-14 ≤ h ≤ 16, -27 ≤ k ≤ 26, -14 ≤ l ≤ 19	? ≤ h ≤ ?, ? ≤ k ≤ ?, ? ≤ l ≤ ?	-15 ≤ h ≤ 10, -9 ≤ k ≤ 18, -24 ≤ l ≤ 6	-27 ≤ h ≤ 14, -14 ≤ k ≤ 26, -29 ≤ l ≤ 37
Reflections collected	61781	13404	?	9418	22076
Independent reflections	17458 [R_{int} = 0.0518, R_{sigma} = 0.0621]	8410 [R_{int} = 0.0365, R_{sigma} = 0.0800]	12896 [R_{int} = ?, R_{sigma} = 0.1362]	7781 [R_{int} = 0.0478, R_{sigma} = 0.0683]	12759 [R_{int} = 0.0281, R_{sigma} = 0.0604]
Data/restraints/parameters	17458/93/955	8410/0/543	12896/264/535	7781/23/779	12759/7/652
Goodness-of-fit on F^2	1.054	1.041	1.041	1.083	1.018
Final R indexes [I>=2σ (I)]	R_1 = 0.0855, wR_2 = 0.2181	R_1 = 0.0590, wR_2 = 0.1182	R_1 = 0.1138, wR_2 = 0.2803	R_1 = 0.0437, wR_2 = 0.1070	R_1 = 0.0501, wR_2 = 0.1310
Final R indexes [all data]	R_1 = 0.1216, wR_2 = 0.2379	R_1 = 0.1023, wR_2 = 0.1402	R_1 = 0.1828, wR_2 = 0.3192	R_1 = 0.0524, wR_2 = 0.1165	R_1 = 0.0746, wR_2 = 0.1431
Largest diff. peak/hole / e Å^{-3}	1.40/-1.25	0.77/-0.81	2.48/-2.56	1.03/-0.62	2.77/-3.54

	9	**10**	**11**	**12**	**13**
Empirical formula	$C_{122}H_{130}Dy_2N_4Ni_2O_{24}$	$C_{102}H_{110}Cl_2Dy_2N_{10}Ni_2O_{26}$	$C_{64}H_{72}Cl_2Dy_2N_4Ni_2O_{26}$	$C_{82}H_{100}Dy_2F_6N_{12}Ni_2O_{26}S_2$	$C_{174}H_{170}Br_8Cl_4Dy_4N_{18}Ni_4O_{52}$
Formula weight	2478.64	2405.31	1826.57	2290.27	5011.19
Temperature/K	100	173	173	173	100
Crystal system	trigonal	monoclinic	monoclinic	monoclinic	triclinic
Space group	R-3	$P2_1/n$	$P2_1/n$	$P2_1/c$	P-1
a/Å	38.159(3)	15.8855(6)	12.1724(12)	16.6632(4)	13.4199(5)
b/Å	38.159(3)	17.2105(11)	14.1352(15)	18.8674(3)	19.4201(7)
c/Å	18.7654(13)	20.5903(11)	19.123(2)	15.7017(3)	20.7033(15)
α/°	90	90	90	90	88.220(6)
β/°	90	96.747(4)	99.425(11)	109.993(3)	85.020(6)
γ/°	120	90	90	90	84.859(6)
Volume/Å^3	23664(4)	5590.3(5)	3245.9(6)	4638.96(19)	5352.1(5)
Z	9	2	2	2	1
ρ_{calc}g/cm^3	1.507	1.429	1.869	1.64	1.555
μ/mm^{-1}	1.832	1.774	3.019	2.13	3.34
F(000)	10962	2444	1828	2316	2480
Crystal size/mm^3	0.15 × 0.12 × 0.06	0.36 × 0.28 × 0.2	0.12 × 0.1 × 0.04	0.26 × 0.2 × 0.06	0.12 × 0.08 × 0.04
Radiation	MoKα (λ = 0.71075)	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)	MoKα (λ = 0.71073)	MoKα (λ = 0.71075)
2Θ range for data collection/°	4.992 to 54.966	5.072 to 58.376	5.184 to 58.058	6.762 to 52.744	6.118 to 54.97
Index ranges	$-38 \le h \le 49$, $-49 \le k \le 39$, $-24 \le l \le 22$	$-21 \le h \le 19$, $0 \le k \le 23$, $0 \le l \le 26$	$-15 \le h \le 13$, $-10 \le k \le 18$, $-13 \le l \le 25$	$-20 \le h \le 20$, $-23 \le k \le 23$, $-18 \le l \le 19$	$-17 \le h \le 16$, $-25 \le k \le 25$, $-26 \le l \le 26$
Reflections collected	56666	13138	12310	17646	72585
Independent reflections	11972 [R_{int} = 0.0483, R_{sigma} = 0.0370]	13129 [R_{int} = 0.0000, R_{sigma} = 0.0495]	7192 [R_{int} = 0.1031, R_{sigma} = 0.2047]	9384 [R_{int} = 0.0267, R_{sigma} = 0.0449]	24304 [R_{int} = 0.0441, R_{sigma} = 0.0524]
Data/restraints/parameters	11972/73/638	13129/446/664	7192/5/462	9384/0/599	24304/1/1203
Goodness-of-fit on F^2	0.992	1.075	0.872	1.039	1.112
Final R indexes [I>=2σ (I)]	R_1 = 0.0408, wR_2 = 0.1095	R_1 = 0.0464, wR_2 = 0.1115	R_1 = 0.0650, wR_2 = 0.0947	R_1 = 0.0382, wR_2 = 0.0885	R_1 = 0.0480, wR_2 = 0.1253
Final R indexes [all data]	R_1 = 0.0553, wR_2 = 0.1157	R_1 = 0.0708, wR_2 = 0.1220	R_1 = 0.1319, wR_2 = 0.1300	R_1 = 0.0501, wR_2 = 0.0976	R_1 = 0.0603, wR_2 = 0.1312
Largest diff. peak/hole / e Å^{-3}	1.46/-0.90	1.24/-1.24	1.35/-1.82	1.03/-1.20	2.65/-3.19

	14	**15**	**16**	**17**	**18**
Empirical formula	$C_{164}H_{184}N_8Ni_8O_{44}$	$C_{92}H_{112}Cl_2Dy_2N_4Ni_2O_{30}$	$C_{106}H_{122}Cl_2Dy_2N_{10}Ni_2O_{28}$	$C_{62}H_{58}Dy_2N_4Ni_2O_{18}$	$C_{88}H_{76}Co_2Dy_2N_{14}O_{34}$
Formula weight	3432.72	2267.18	2497.46	1589.54	2316.48
Temperature/K	100	173	173	173	100
Crystal system	triclinic	monoclinic	monoclinic	monoclinic	triclinic
Space group	P-1	$P2_1/n$	$P2_1/c$	$P2_1/c$	P-1
a/Å	18.0377(13)	12.19560(19)	15.0768(5)	10.4776(4)	13.1490(9)
b/Å	18.3422(13)	19.3817(6)	20.6499(5)	21.3408(8)	13.6200(10)
c/Å	29.900(2)	20.2019(7)	17.8256(8)	14.3681(5)	15.4701(11)
α/°	71.347(2)	90	90	90	68.869(5)
β/°	71.670(2)	98.464(2)	103.010(4)	106.977(4)	65.650(4)
γ/°	67.774(2)	90	90	90	63.614(4)
Volume/Å^3	8462.6(11)	4723.1(2)	5407.3(3)	3072.7(2)	2207.1(3)
Z	2	2	2	2	1
ρ_{calc}g/cm^3	1.262	1.594	1.534	1.718	1.743
μ/mm^{-1}	0.941	2.095	1.838	14.105	2.141
F(000)	3348	2308	2548	1580	1160
Crystal size/mm^3	0.22 × 0.08 × 0.03	0.28 × 0.14 × 0.1	0.28 × 0.24 × 0.12	0.08 × 0.06 × 0.03	0.03 × 0.03 × 0.01
Radiation	MoKα (λ = 0.71075)	Mo Kα (λ = 0.71073)	Mo Kα (λ = 0.71073)	CuKα (λ = 1.54184)	MoKα (λ = 0.71075)
2Θ range for data collection/°	4.49 to 55.226	5.32 to 58.32	5.08 to 58.5	9.75 to 140.882	4.986 to 55.256
Index ranges	$-23 \le h \le 23$, $-23 \le k \le 23$, $-38 \le l \le 38$	$-14 \le h \le 16$, $-26 \le k \le 22$, $-26 \le l \le 25$	$-19 \le h \le 19$, $-26 \le k \le 27$, $-23 \le l \le 16$	? ≤ h ≤ ?, ? ≤ k ≤ ?, ? ≤ l ≤ ?	$-17 \le h \le 17$, $-17 \le k \le 17$, $-20 \le l \le 20$
Reflections collected	106354	29019	27684	?	49522
Independent reflections	38045 [R_{int} = 0.0854, R_{sigma} = 0.0739]	11050 [R_{int} = 0.0320, R_{sigma} = 0.0412]	12547 [R_{int} = 0.0313, R_{sigma} = 0.0458]	5641 [R_{int} = ?, R_{sigma} = 0.0927]	10202 [R_{int} = 0.1033, R_{sigma} = 0.0580]
Data/restraints/parameters	38045/206/1922	11050/9/613	12547/0/686	5641/3/386	10202/15/621
Goodness-of-fit on F^2	1.049	1.053	1.065	0.99	1.078
Final R indexes [I>=2σ (I)]	R_1 = 0.0643, wR_2 = 0.1750	R_1 = 0.0322, wR_2 = 0.0687	R_1 = 0.0362, wR_2 = 0.0816	R_1 = 0.0666, wR_2 = 0.1672	R_1 = 0.0745, wR_2 = 0.1889
Final R indexes [all data]	R_1 = 0.0845, wR_2 = 0.1896	R_1 = 0.0451, wR_2 = 0.0755	R_1 = 0.0491, wR_2 = 0.0908	R_1 = 0.0899, wR_2 = 0.1837	R_1 = 0.0859, wR_2 = 0.1971
Largest diff. peak/hole / e Å^{-3}	0.91/-1.15	1.18/-0.54	1.05/-0.94	3.24/-1.36	4.51/-2.63

	19	**20**	**21**	**H2L3**	**H2L19**
Empirical formula	$C_{94}H_{90}Co_2Dy_2N_{14}O_{32}$	$C_{81}H_{69}Dy_3N_{24}O_{60}$	$C_{181}H_{171}Dy_4N_{27}Ni_4O_{66}$	$C_{14}H_{11}ClN_2O_5$	$C_{21}H_{20}BrNO_4$
Formula weight	2370.65	2826.1	4665.28	322.7	430.29
Temperature/K	100	100	173	173	173
Crystal system	triclinic	triclinic	triclinic	triclinic	triclinic
Space group	P-1	P-1	P-1	P-1	P-1
a/Å	12.7821(9)	16.5657(12)	14.1586(6)	6.9697(4)	7.5662(4)
b/Å	13.5743(10)	17.6842(12)	17.2590(7)	12.6645(7)	8.5456(4)
c/Å	14.1583(10)	18.4698(13)	22.3936(8)	15.0866(8)	14.8304(8)
α/°	106.940(3)	81.572(2)	98.527(3)	76.153(5)	84.747(4)
β/°	91.065(2)	80.148(2)	98.268(3)	89.966(5)	84.775(4)
γ/°	97.754(2)	83.020(2)	111.403(4)	88.423(5)	82.090(4)
Volume/Å^3	2324.2(3)	5247.5(6)	4920.7(4)	1292.45(13)	942.80(8)
Z	1	2	1	4	2
ρ_{calc}g/cm^3	1.694	1.789	1.574	1.658	1.516
μ/mm^{-1}	2.034	2.233	1.966	2.9	2.207
F(000)	1194	2802	2350	664	440
Crystal size/mm^3	0.14 × 0.12 × 0.04	0.13 × 0.1 × 0.05	0.2 × 0.18 × 0.14	0.12 × 0.08 × 0.01	0.28 × 0.14 × 0.04
Radiation	MoKα (λ = 0.71075)	MoKα (λ = 0.71075)	MoKα (λ = 0.71073)	CuKα (λ = 1.54184)	Mo Kα (λ = 0.71073)
2Θ range for data collection/°	4.98 to 55.026	4.68 to 55.01	6.714 to 58.828	10.444 to 142.68	5.36 to 58.26
Index ranges	-16 ≤ h ≤ 16, -17 ≤ k ≤ 16, -18 ≤ l ≤ 16	-21 ≤ h ≤ 21, -22 ≤ k ≤ 22, -22 ≤ l ≤ 23	-18 ≤ h ≤ 19, -23 ≤ k ≤ 21, -30 ≤ l ≤ 25	-5 ≤ h ≤ 8, -15 ≤ k ≤ 15, -18 ≤ l ≤ 18	-10 ≤ h ≤ 10, -11 ≤ k ≤ 11, -19 ≤ l ≤ 19
Reflections collected	37052	76415	33792	7064	21895
Independent reflections	10595 [R_{int} = 0.0498, R_{sigma} = 0.0413]	23648 [R_{int} = 0.0367, R_{sigma} = 0.0304]	21989 [R_{int} = 0.0315, R_{sigma} = 0.0630]	4806 [R_{int} = 0.0243, R_{sigma} = 0.0398]	4630 [R_{int} = 0.0562, R_{sigma} = 0.0474]
Data/restraints/parameters	10595/112/653	23648/0/1489	21989/19/1284	4806/2/417	4630/2/253
Goodness-of-fit on F^2	1.047	1.05	1.053	1.054	1.049
Final R indexes [I>=2σ (I)]	R_1 = 0.0427, wR_2 = 0.1152	R_1 = 0.0314, wR_2 = 0.0764	R_1 = 0.0425, wR_2 = 0.0968	R_1 = 0.0493, wR_2 = 0.1324	R_1 = 0.0373, wR_2 = 0.0719
Final R indexes [all data]	R_1 = 0.0469, wR_2 = 0.1197	R_1 = 0.0360, wR_2 = 0.0797	R_1 = 0.0618, wR_2 = 0.1096	R_1 = 0.0593, wR_2 = 0.1451	R_1 = 0.0506, wR_2 = 0.0768
Largest diff. peak/hole / e Å^{-3}	2.80/-1.53	2.10/-1.47	1.32/-0.93	0.66/-0.33	0.40/-0.50

Synthesis of compound 4 – 21

Compound $(Et_3NH)[Co^{III}(\mathbf{L6})_2]\cdot 3MeOH$ [**4**·3MeOH]**.** $\mathbf{H_2L6}$ (0.2 mmol, 56 mg) was suspended in a methanolic solution (20 mL), to which $Co(ClO_4)_2.6H_2O$ (0.1 mmol), $Dy(OTf)_3$ (0.1 mmol, 61 mg) and Et_3N (0.5 mmol, 69 μL) was added. The resultant solution was stirred for 1h and on completion filtered. The filtrate was left for slow evaporation and after 3 days dark red crystals formed.

Compound $(Et_3NH)[Co^{III}(\mathbf{L20})_2]\cdot 2MeOH$ [**5**·2MeOH]**.** $\mathbf{H_2L20}$ (0.2 mmol, 74 mg) was suspended in a methanolic solution (20 mL), to which $Co(ClO_4)_2.6H_2O$ (0.1 mmol), $Dy(OTf)_3$ (0.1mmol, 61mg) and Et_3N (0.5 mmol, 69 μL) was added. The resultant solution was stirred for 1h and on completion filtered. The filtrate was left for slow evaporation and after 6 days dark red crystals formed.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L14})_4(allyl\text{-}o\text{-}vanillin)_2(EtOH)_2]\cdot 3CH_2Cl_2$ [**6**·$3CH_2Cl_2$]**.** $\mathbf{H_2L14}$ (0.2 mmol, 72 mg) was suspended in a EtOH (10 mL) and Et_3N (0.5 mmol, 69 μL) was immediately added. The solution was left to stir and $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg) and $Dy(Of)_3$ (0.1 mmol, 61 mg were added after 5 minutes. The resultant solution was refluxed for 1h, before CH_2Cl_2 (5 mL) was added and left for slow evaporation. After 3 days brown, block like crystals had formed.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L14})_4(DMF)_6]$ $2(ClO_4)$ 4DMF [**7** 4DMF]**.** $\mathbf{H_2L14}$ (0.2 mmol, 72 mg), $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf)_3$ (0.1 mmol, 61 mg) and Et_3N (0.5 mmol, 69 μL) were added to DMF (10 mL) and left to stir for 1h. The solution was filtered and filtrate underwent vapour diffusion with Et_2O. After 7 days large brown crystals were formed. Elemental analysis for $C_{122}H_{146}Cl_2Dy_2N_{14}Ni_2O_{30}$: calcd. C 51.00 , H 4.52, N 5.61; found C51.32, H 4.05, N 5.83.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L19})_4(bromo\text{-}o\text{-}vanillin)_2(EtOH)_2]$ 6EtOH [**8**·6EtOH]**.** $\mathbf{H_2L19}$ (0.2 mmol, 80mg) was suspended in a EtOH (15 mL) and Et_3N (0.5 mmol, 69 μL) was immediately added. The solution was left to stir and $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg) and $Dy(OTf)_3$ (0.1 mmol, 61 mg) were added after 5 minutes. The resultant solution was stirred for 1h, before THF (5 mL) was added and left for slow evaporation. After 5 days brown, block like crystals had formed.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L15})_4(\text{allyl-o-vanillin})_2(EtOH)_2]$ 2THF 2EtOH [**9** 2THF 2EtOH]**. H_2L15** (0.2 mmol, 66 mg), $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf)_3$ (0.1 mmol, 61 mg) and Et_3N **(**0.5 mmol, 69 µL) were added to a mixture a 1:1 of CH_2Cl_2 and THF (20 mL). The reaction mixture was stirred for 1h and on completion filtered. The filtrate was left for 6 days after which time small red block crystals had formed.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L15})_4(DMF)_2]$ 2(ClO_4) [**10**]**. H_2L15** (0.2 mmol, 66 mg), $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf)_3$ (0.1 mmol, 61 mg) and Et_3N (0.5 mmol, 69 µL) were added to DMF (10 mL) and left to stir for 1h. The solution was filtered and filtrate underwent vapour diffusion with Et_2O. After 9 days large brown cuboid crystals were formed. Elemental analysis for $C_{102}H_{110}Cl_2Dy_2N_{10}Ni_2O_{26}$: calcd. C 50.87 , H 4.77, N 5.82; found C 50.92, H 5.25, N 6.15.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L1})_4(EtOH)_4(H_2O)_2](ClO_4)_2$ (**11**)**. H_2L1** (0.6 mmol, 144 mg), $Ni(ClO_4)_2.6H_2O$ (0.3 mmol, 111 mg), $Dy(OTf)_3$ (0.3 mmol, 183 mg) and Et_3N (1.5 mmol, 205.5 µL) were added to EtOH (20 mL) and left to stir for 1h. The solution was filtered and left for slow evaporation. After 5 days brown needle like crystals had formed. Elemental analysis for $C_{64}H_{72}Cl_2Dy_2N_4Ni_2O_{26}$: calcd. C 48.22 , H 4.67, N 6.44; found C 46.94, H 4.64, N 6.08.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L1})_4(DMF)_6](OTf_3)_2\cdot 2DMF$ (**12**·2DMF). **H_2L1** (0.6 mmol, 144 mg), $Ni(ClO_4)_2.6H_2O$(0.3 mmol, 111 mg), $Dy(OTf)_3$ (0.3 mmol, 183 mg) and Et_3N (1.5 mmol, 205.5 µL) were added to DMF (10 mL) and left to stir for 1h. The solution was filtered and underwent vapour diffusion with Et_2O. After 3 days brown block like crystals had formed.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L20})_4(DMF)_6]$ $[Ni^{II}_2Dy^{III}_2(\mathbf{L20})_4(DMF)_4(H_2O)_2]$ 4(ClO_4) 5DMF (**13**·5DMF). **H_2L20** (0.2 mmol, 74 mg), $Ni(ClO_4)_2.6H_2O$(0.1 mmol, 37 mg), $Dy(OTf)_3$ (0.1 mmol, 61 mg) and an excess of Et_3N were added to DMF(10 mL) and left to sir for 1h. The solution was filtered and underwent vapour diffusion with Et_2O. After 2 weeks, light brown crystals were formed. Elemental analysis for $C_{174}H_{170}Br_8Cl_4Dy_4N_{18}Ni_4O_{52}$: calcd. C 52.23 , H 5.37, N 6.99; found C 43.31, H 3.85, N 5.56.

Compound $[Ni^{II}_4(\mathbf{L5})_4(MeOH)_4]$ ·6MeOH (**14** · 6MeOH)**.** $\mathbf{H_2L5}$ (0.1 mmol, 58 mg), $Ni(ClO4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf_3)_3$ (0.1 mmol, 61 mg) and Et_3N (0.5 mmol) were suspended in methanol(20 mL) forming a light yellow solution. The solution was stirred for 1h before being filtered and the filtrate left for slow evaporation. After 2 days light green crystals were collected.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L5})_4(EtOH)_6]$ $2(ClO_4)$ · 4EtOH (**15** · 4EtOH). $\mathbf{H_2L5}$ (0.2 mmol, 58 mg), $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf)_3$ (0.1 mmol, 61 mg) and Et_3N (0.5 mmol, 69 μL) were added to EtOH (20 mL) and left to stir for 1h. The solution was filtered and left for slow evaporation. After 13 days brown cuboid crystals were formed.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L4})_4(DMF)_6]$ $2(ClO_4)$ ·$2Et_2O$ (**16** ·$2Et_2O$)**.** $\mathbf{H_2L4}$ (0.2 mmol, 64 mg), $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf)_3$ (0.1 mmol, 61 mg) and Et_3N (0.5 mmol, 69 μL) were added to DMF (10 mL) and left to stir for 1h. The solution was filtered and the filtrate underwent vapour diffusion with Et_2O. After 4 days brown cuboid crystals were formed. Elemental analysis for $C_{106}H_{122}Cl_2Dy_2N_{10}Ni_2O_{28}$: calcd. C 48.72 , H 4.77, N 5.77; found C 47.93, H 4.66, N 5.52.

Compound $[Ni^{II}_2Dy^{III}_2(\mathbf{L1})_4(OAc)_2(MeOH)_2]$ ·2MeOH (**17** · 2MeOH)**.** $\mathbf{H_2L1}$ (0.1 mmol, 25 mg), $Dy(OTf)_3$ (122 mg, 0.2 mmol) and Et_3N (27.6 μL) were added to MeOH (20 mL) and left to stir for 20minutes. $Ni(OAc)_2.4H_2O$ (0.1 mmol, 25 mg) was added at the end of this time and left to stir for a further 20 minutes. The solution was filtered and left for slow evaporation. Small rectangular black crystals formed after 3 days.

Compound $[Co^{II}_2Dy^{III}_2(\mathbf{L12})_4(\mathbf{M1})_2(DMF)_2]$ (**18**)**.** $\mathbf{H_2L12}$ (0.25 mmol, 65 mg) and 3,5-dinirtobenzoic acid (0.5 mmol,) were suspended in DMF (10 mL) to form a cloudy brown solution. Upon addition of $Co(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf_3)_3$ (0.1 mmol, 61 mg) and Et_3N (0.9 mmol, 121 μL) the solution became a clear porter red. The solution was stirred for 1h and upon completion filtered. The filtrate underwent vapour diffusion with Et_2O and after 2 weeks large dark red crystals were collected.

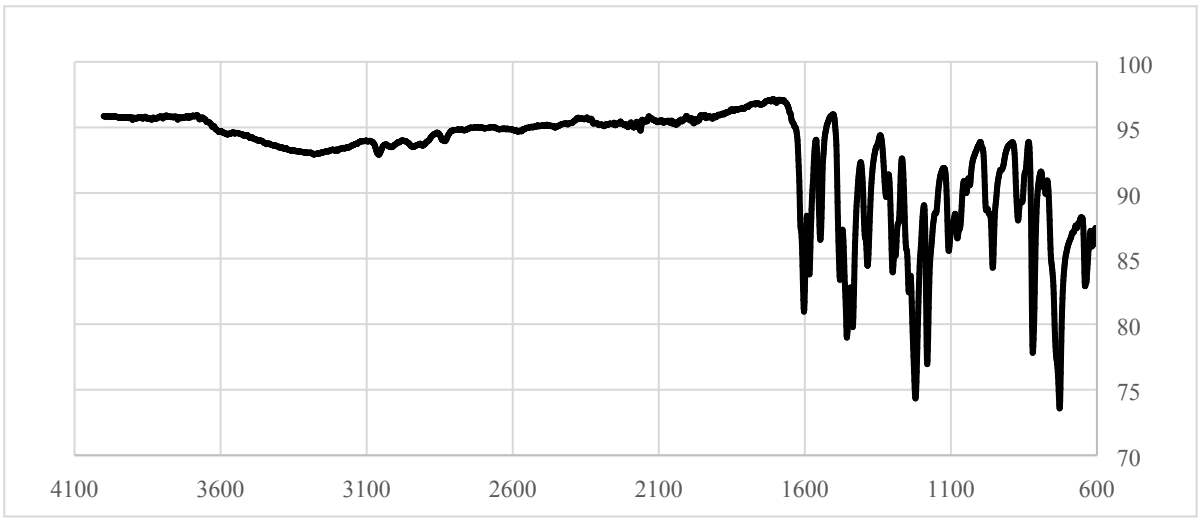
Compound $[Co^{II}_2Dy^{III}_2(\mathbf{L12})_4(\mathbf{M2})_2(DMF)_2] \cdot 2DMF$ (**19**·2DMF)**.** $\mathbf{H_2L12}$ (0.25 mmol, 85 mg) and 3-nirtobenzoic acid (0.5 mmol) were suspended in DMF (10 mL) to form a cloudy brown solution. Upon addition of Co $(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf_3)_3$ (0.1 mmol, 61 mg) and Et_3N (0.9 mmol, 121 μL) the solution became a clear porter red. The solution was stirred for 1h and upon completion filtered. The filtrate underwent vapour diffusion with Et_2O and after 2 weeks large dark red crystals were collected.

Compound $[Dy^{III}(\mathbf{M1})_3(DMF)_2]$ (**20**)**.** H_2L2 (0.25 mmol, 85 mg) and 3,5-dinirtobenzoic acid (0.5 mmol) were suspended in DMF (10 mL) to form a cloudy brown solution. Upon addition of Co $(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf_3)_3$ (0.1 mmol, 61 mg) and Et_3N (0.9 mmol, 121 μL) the solution became a clear porter red. The solution was stirred for 1h and upon completion filtered. The filtrate underwent vapour diffusion with Et_2O and after 3 days clear needle-like crystals were collected.

Compound $[Ni^{II}_2Dy^{III}_2(OH)(\mathbf{L5})_3(\mathbf{M1})_3(DMF)_2] \cdot 1.5DMF \cdot Et_2O$ (**21**·1.5DMF·Et_2O)**.** $\mathbf{H_2L_5}$ (0.2 mmol, 58 mg), $Ni(ClO_4)_2.6H_2O$ (0.1 mmol, 37 mg), $Dy(OTf)_3$ (0.1 mmol, 61 mg), 3,5 -dintrobenzoic acid (0.5 mmol, 70 mg) and Et_3N (0.5 mmol, 69 μL) were added to DMF(10 mL) and left to stir for 1h. The solution was filtered and underwent vapour diffusion with Et_2O. After 8 days black small cube crystals were formed.

FT-IR of Complexes

4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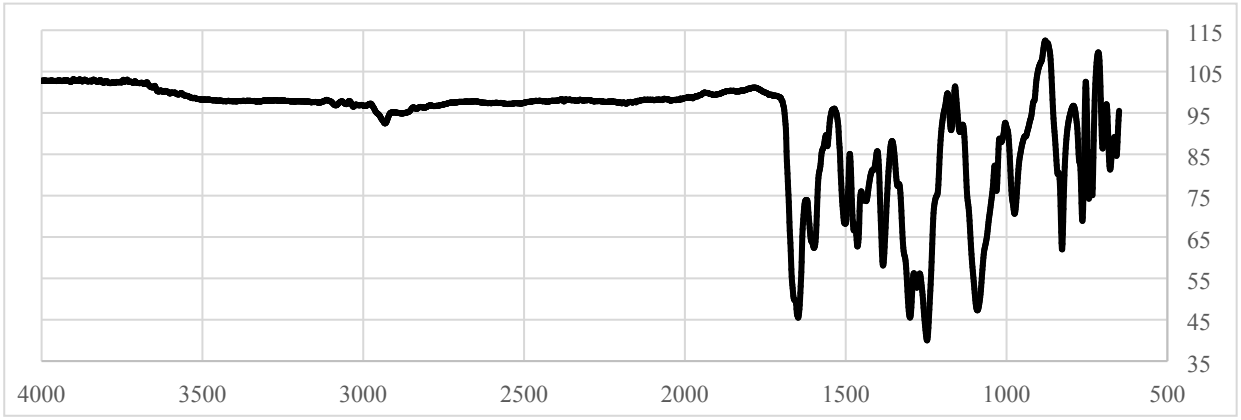
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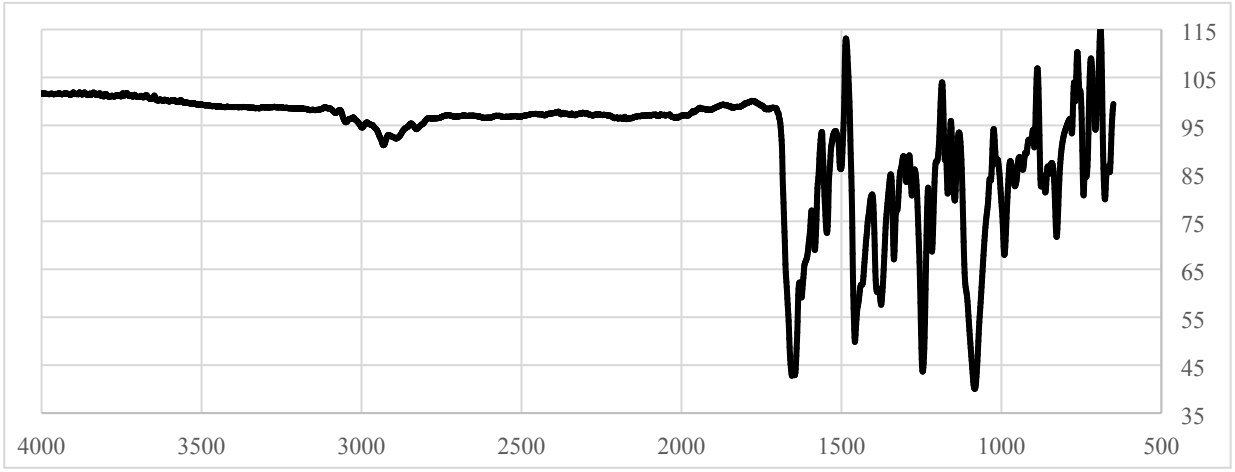
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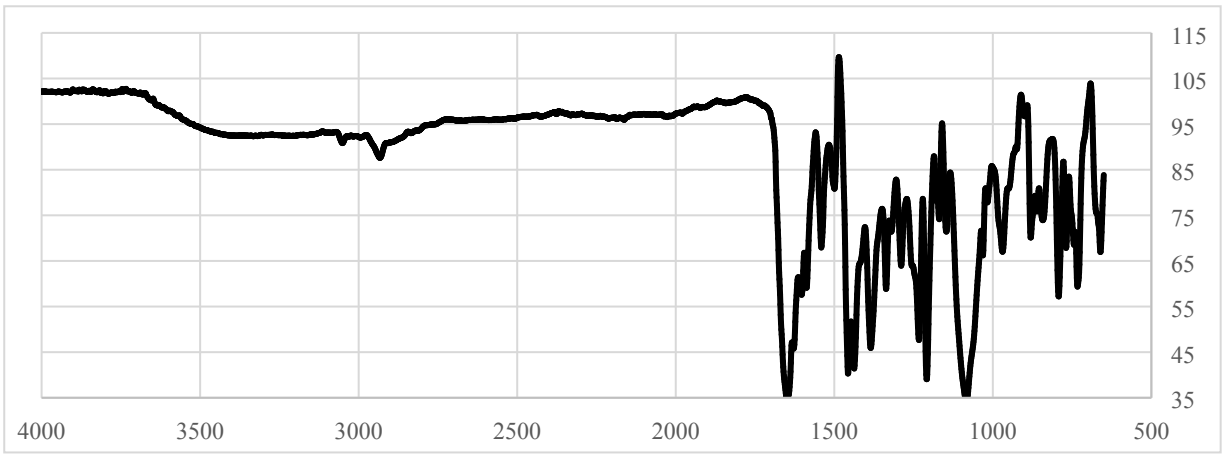
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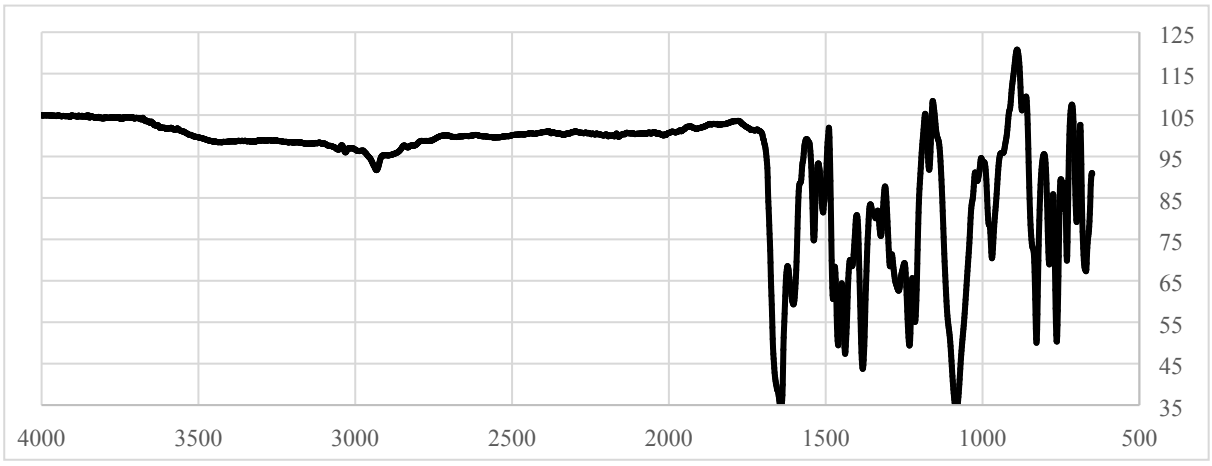
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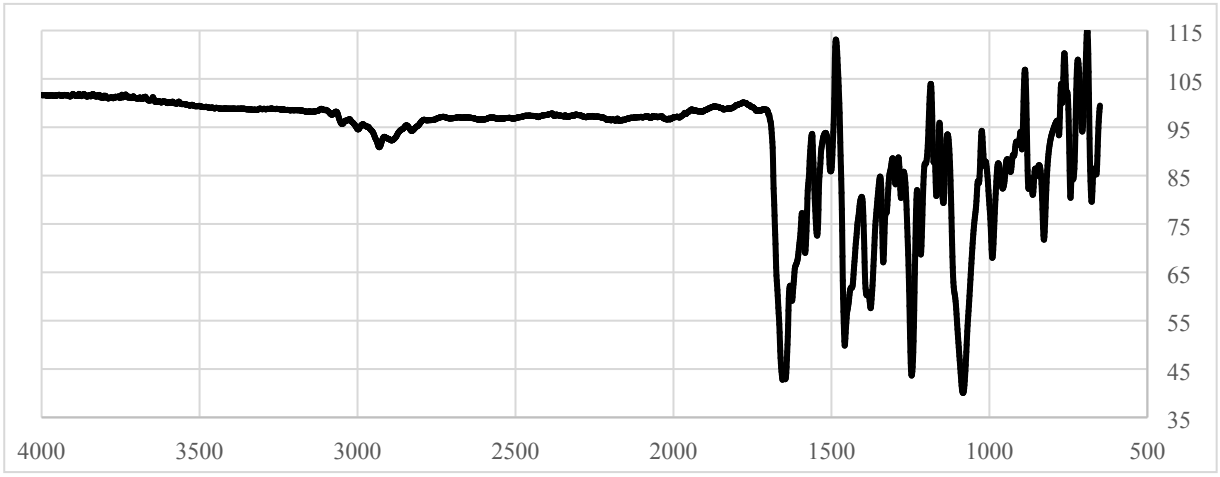
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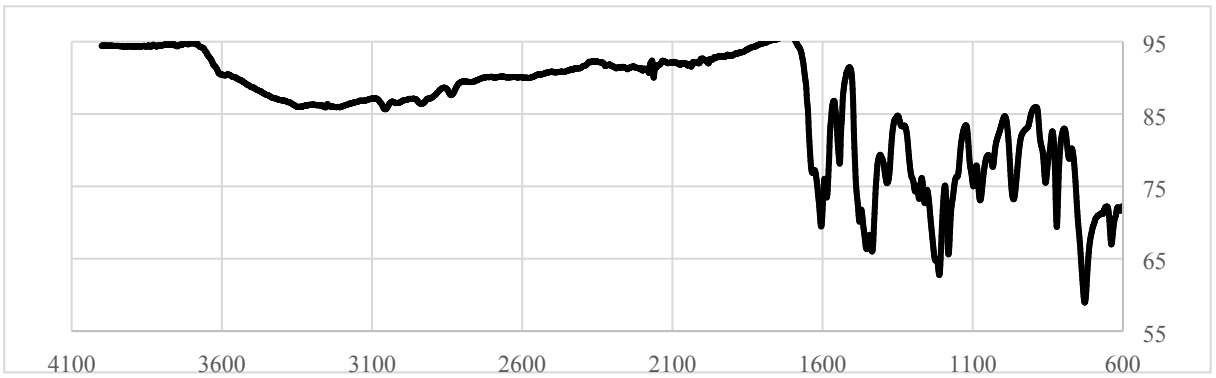
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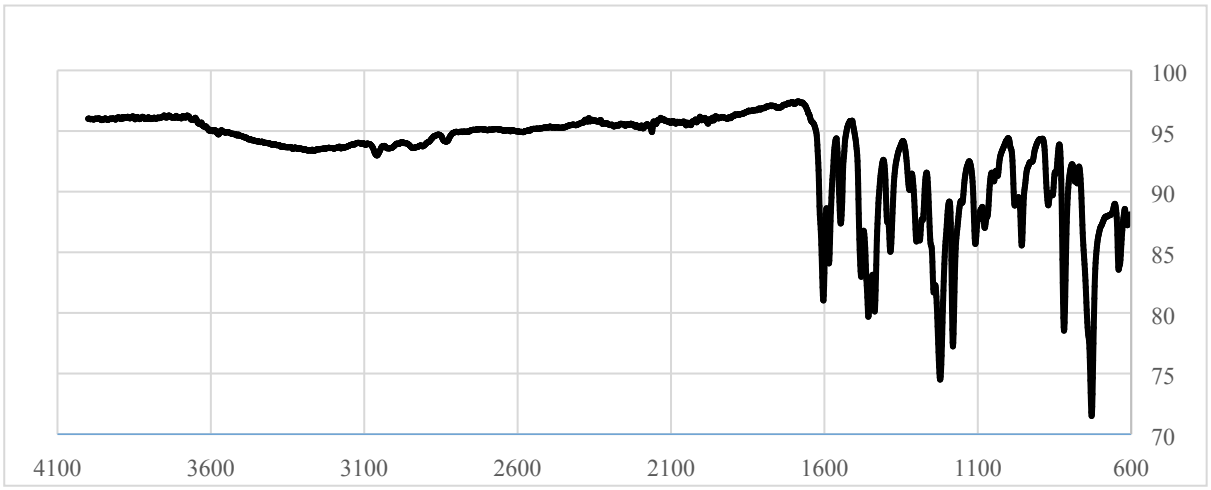
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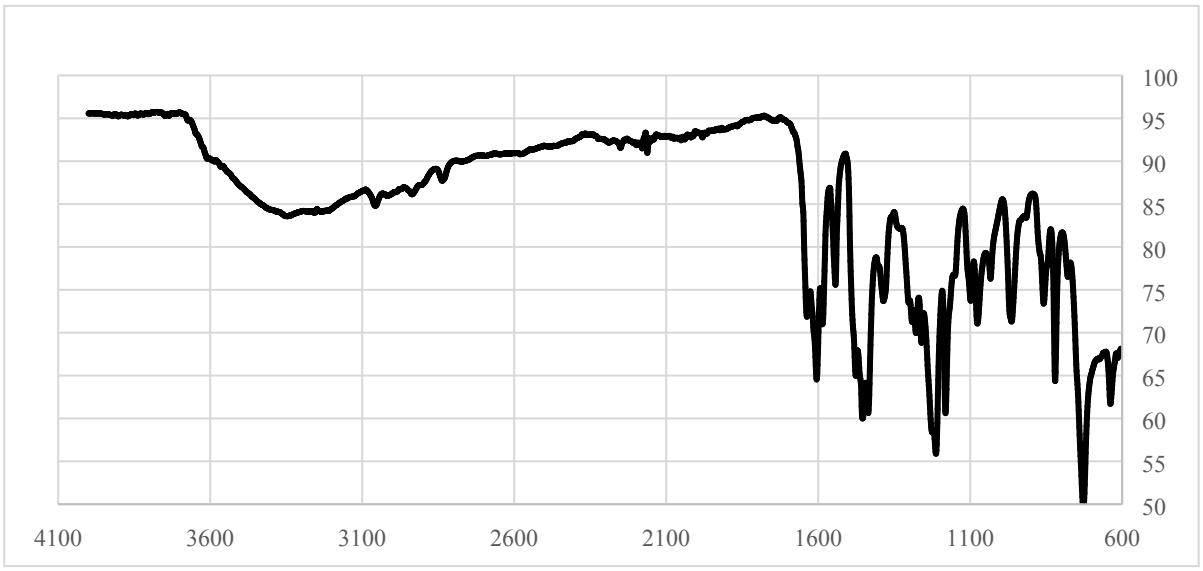
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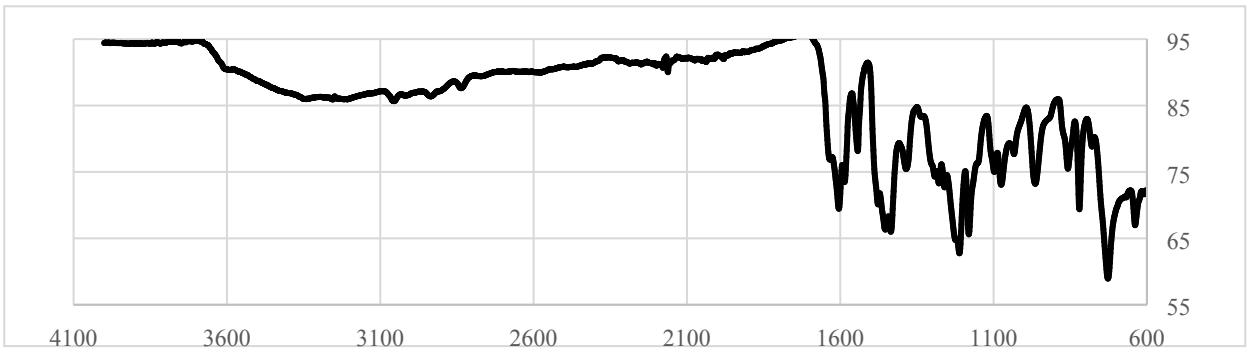
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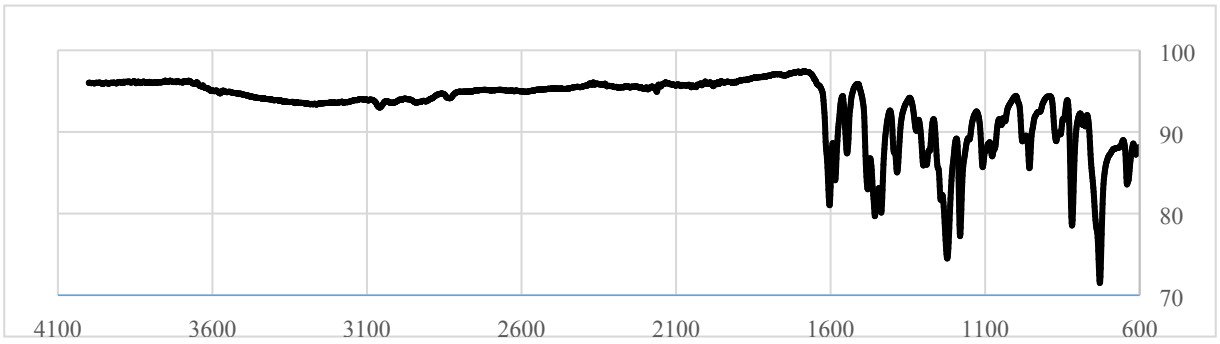
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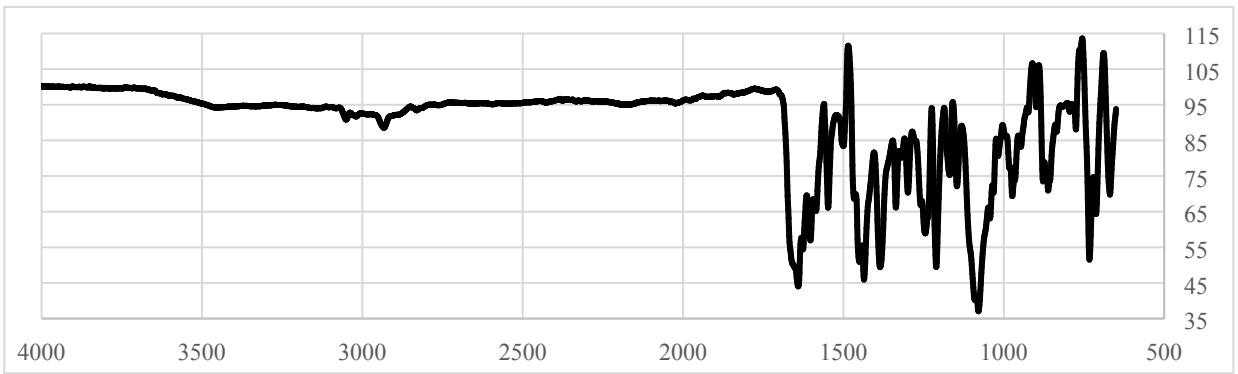
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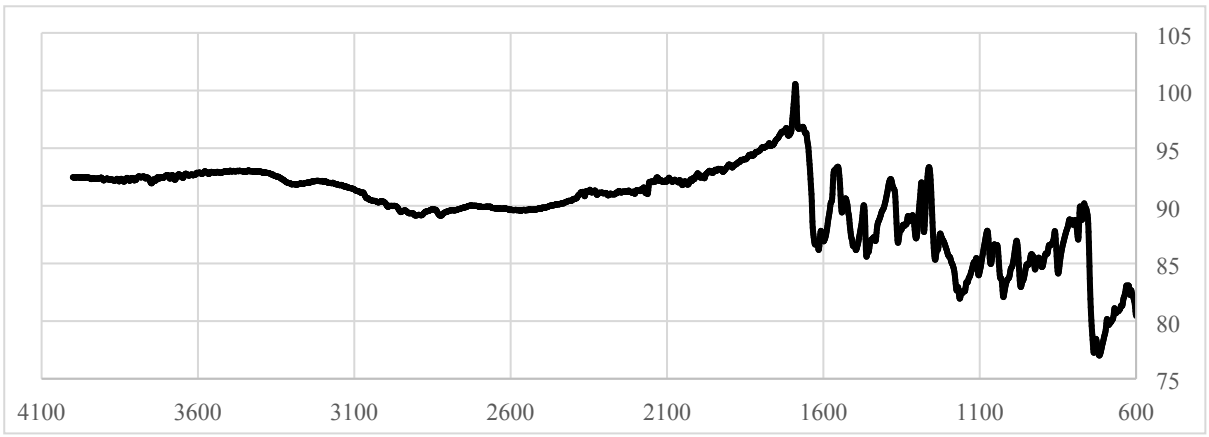
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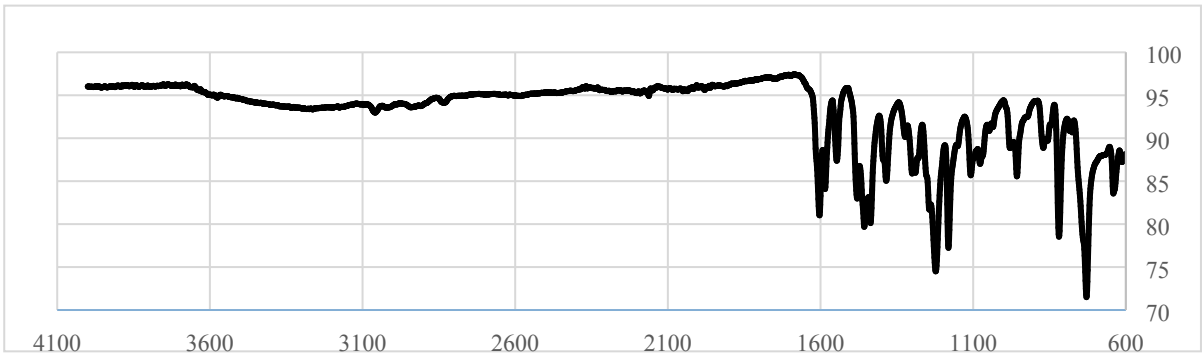
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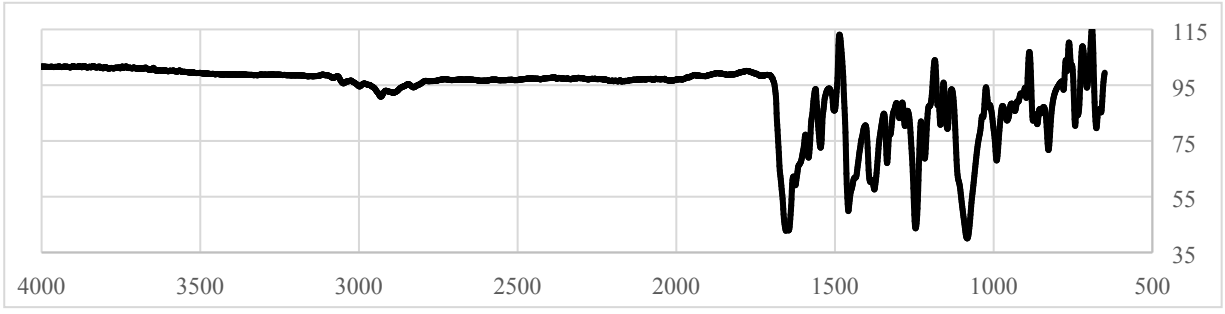
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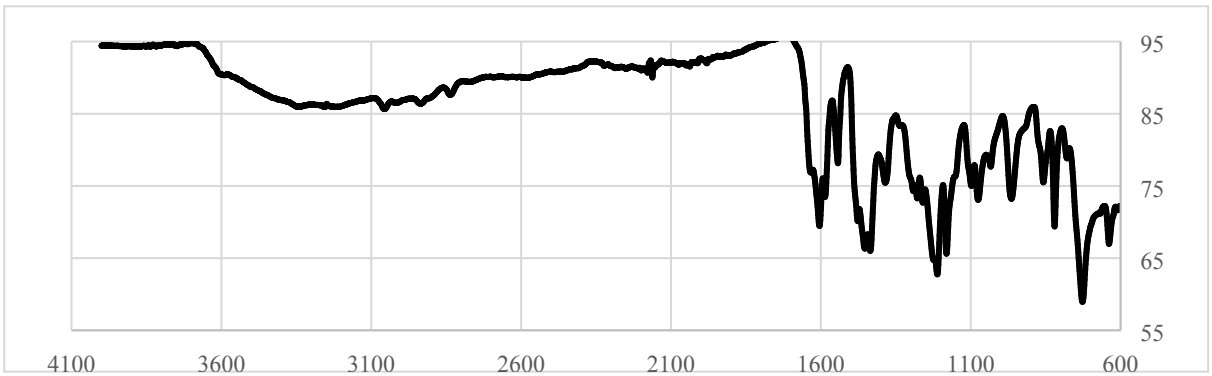

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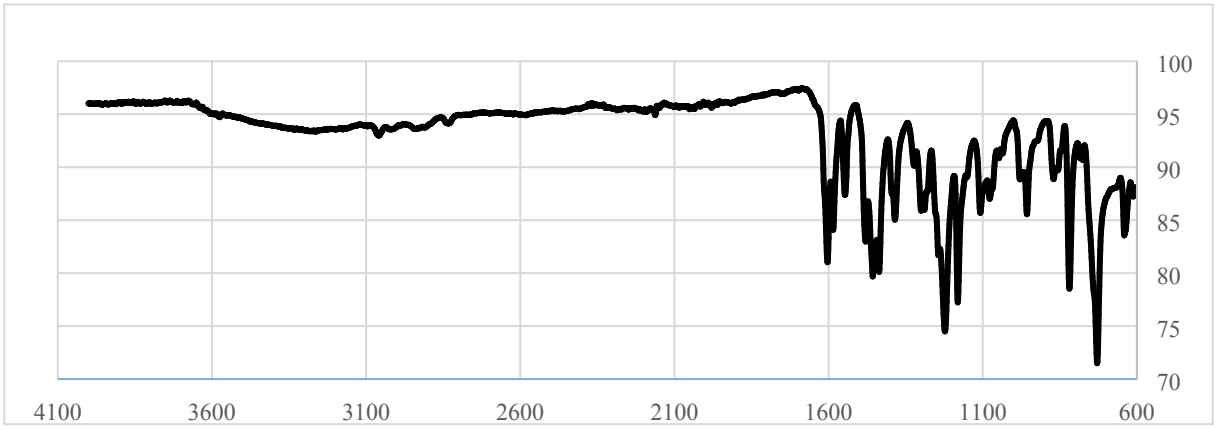
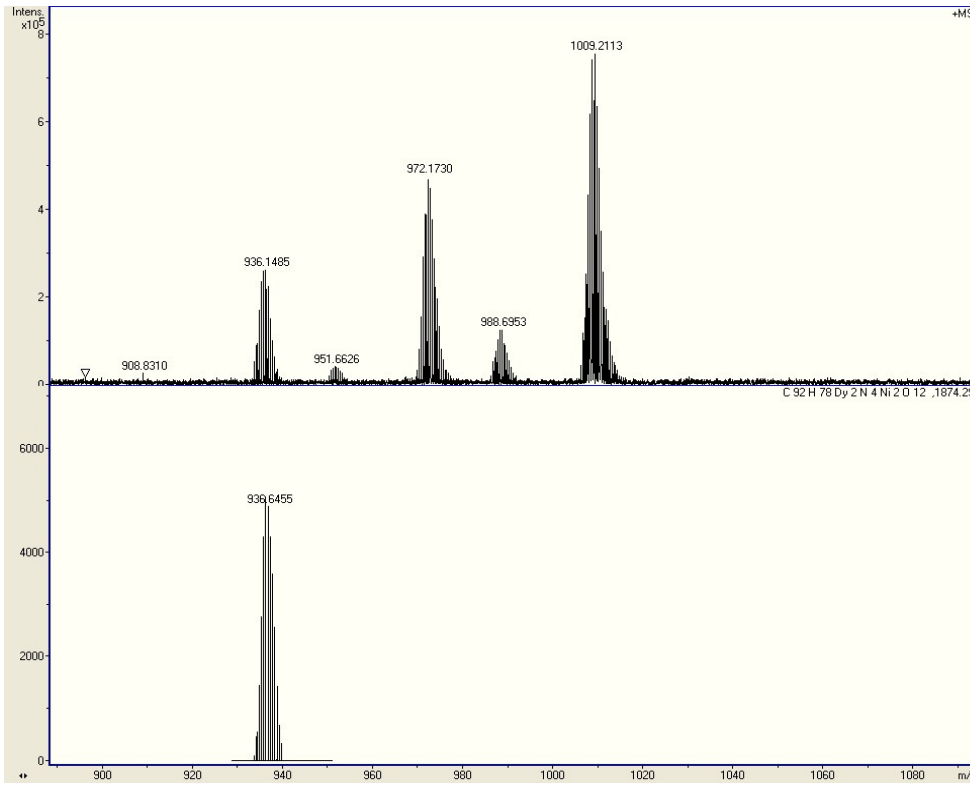


ESI-MS of Compounds 7,10 and 16

Compound 7



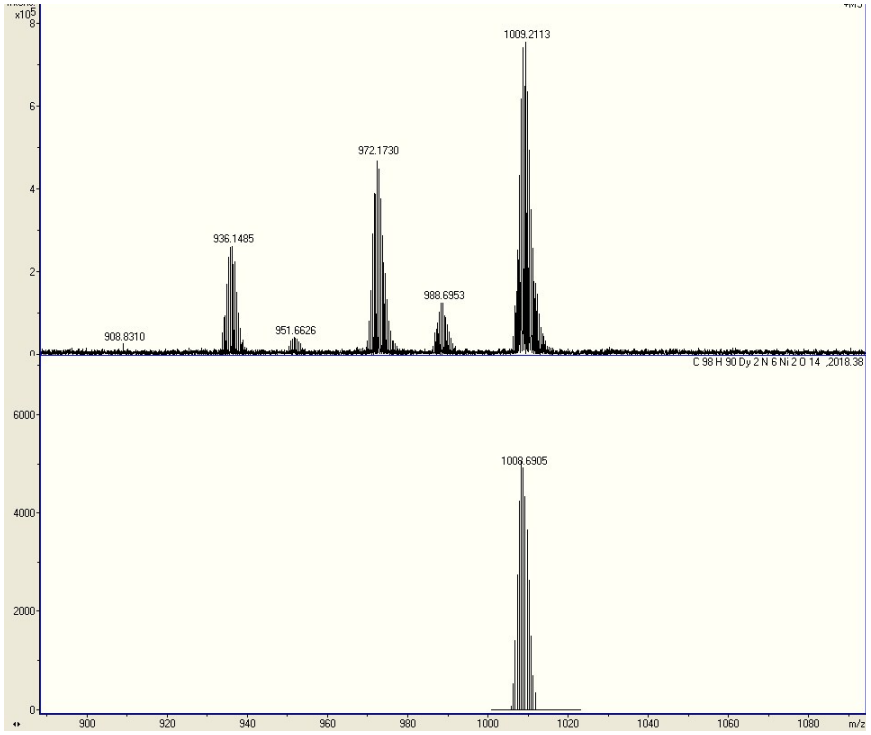
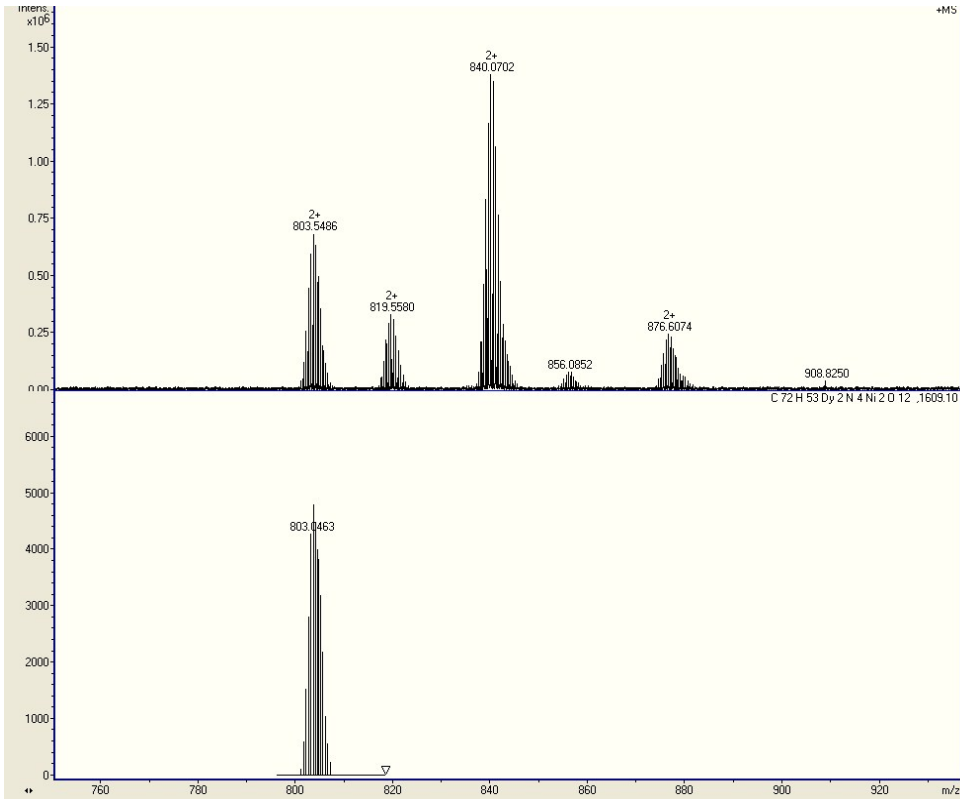


Fig S1 ESI-MS of Compound **7**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2+
819.5580
2+
876.6074
856.0852
908.8250
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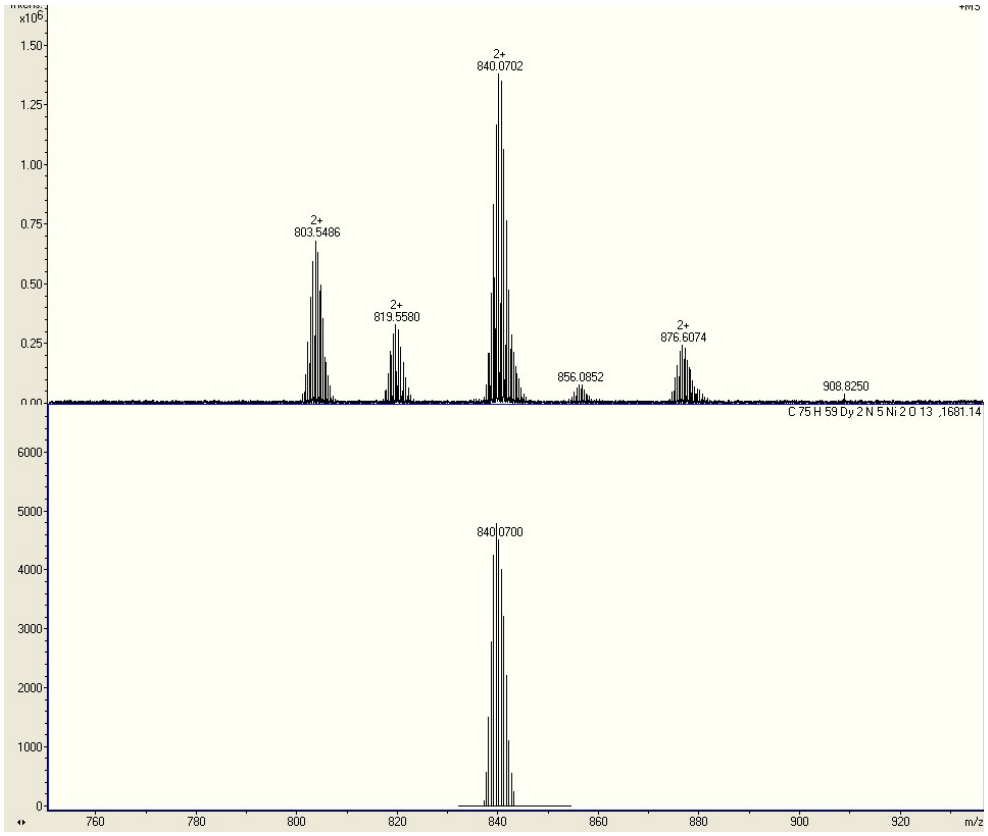
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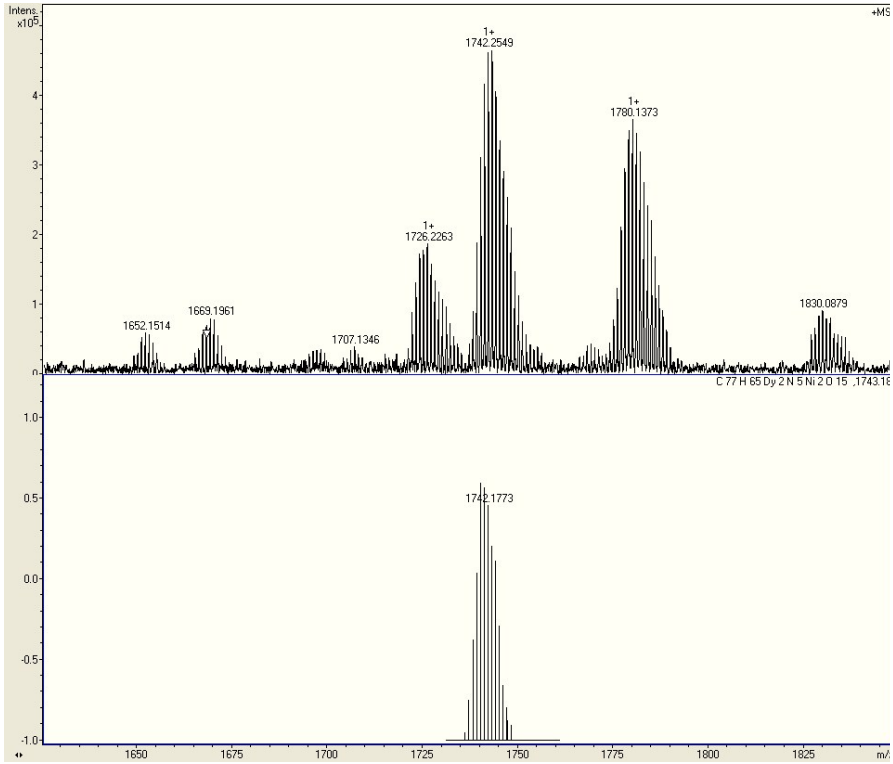
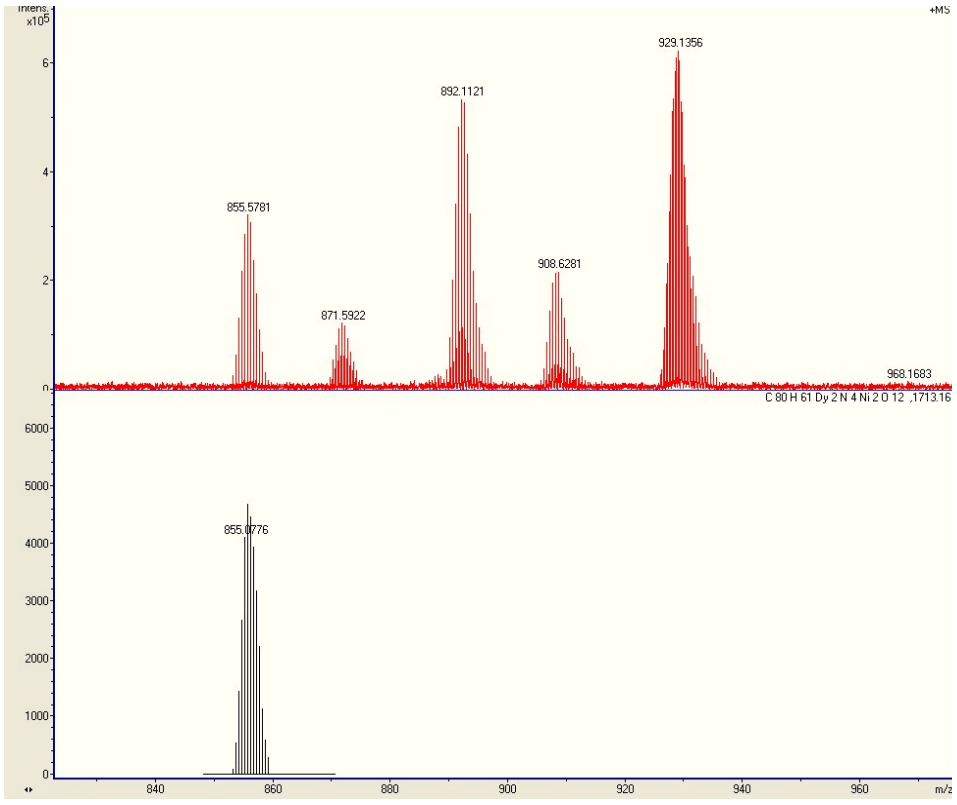
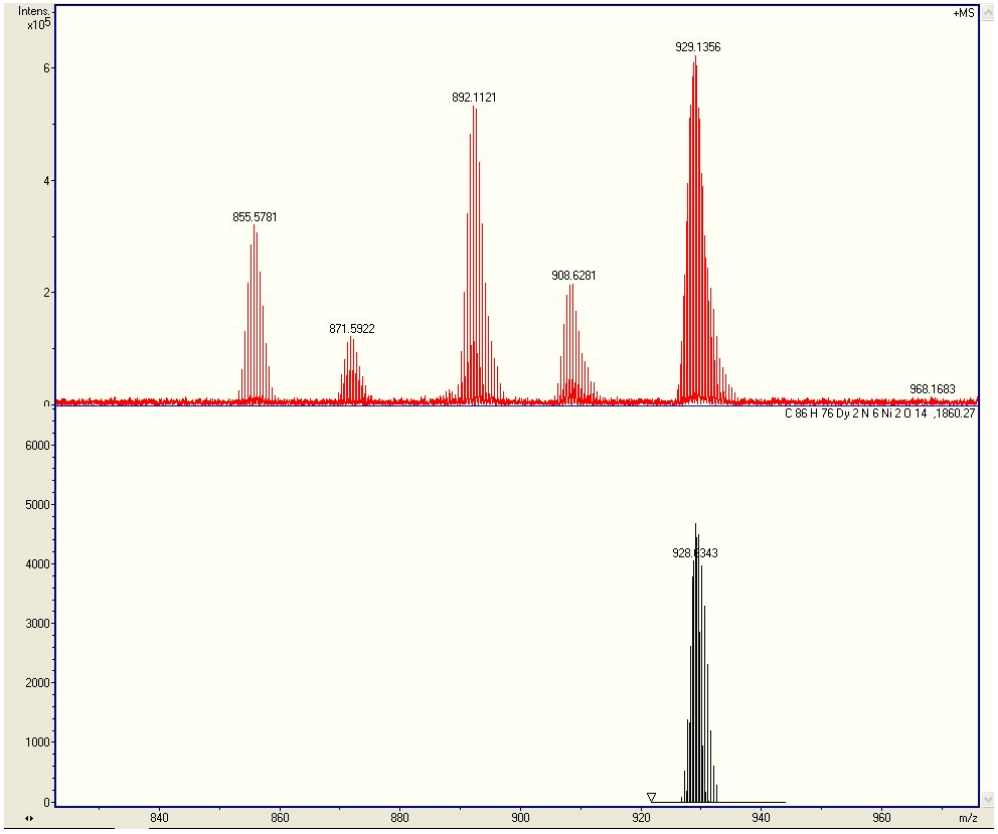


Fig S2 ESI-MS of Compound **10**





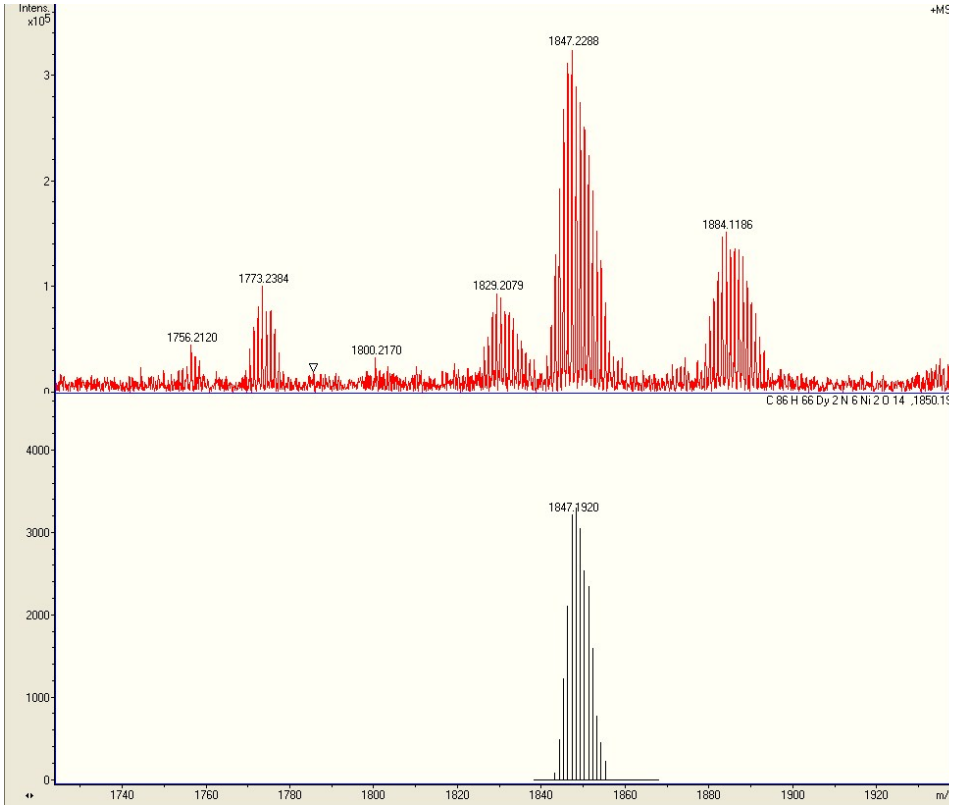


Fig S3 ESI-MS of Compound **16**

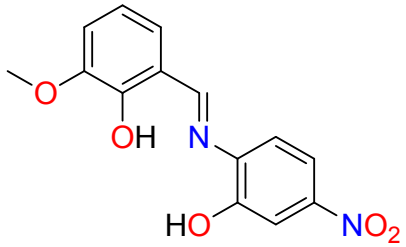
Protocols for Reflux Synthesis and Microwave Synthesis of Ligands

General Synthetic protocol for Ligands via Reflux (H_2L1-H_2L20)

5-R-3-methoxy salicyldahyde (0.0125mol) and 2-amino-4/5-R-phenol (0.0125mol) were dissolved in MeOH (5mL). The resultant solution was refluxed from 1-2 hours, during which time a bright coloured precipitate would form. The reaction mixture would be filtered and the precipitate washed with cold methanol (15mL) and Et_2O (10mL). The solid would be dried overnight under vacuo.

E)-2-((2-hydroxy-3-methoxybenzylidene)amino)-5-nitrophenol (H_2L2)

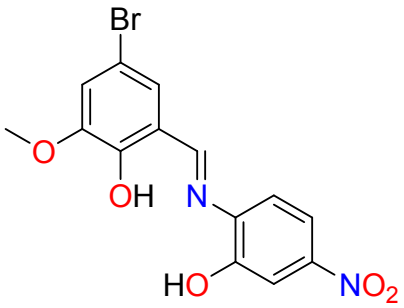
O-Vanillin (0.025mol, 3.35g) and 2-amino-5-nitrophenol (0.025mol, 3.30g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time a dark brown to red solid separated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 66%.



^{1}H NMR (500 MHz,) δ 13.18 (s, 1H), 10.72 (s, 1H), 9.01 (s, 1H), 7.77 (dd, J = 8.4, 1.7 Hz, 2H), 7.57 – 7.51 (m, 1H), 7.26 (dd, J = 7.9, 1.5 Hz, 1H), 7.15 (dd, J = 8.0, 1.5 Hz, 1H), 6.90 (t, J = 7.9 Hz, 1H), 3.82 (s, 3H).

(E)-4-bromo-2-(((2-hydroxy-4-nitrophenyl)imino)methyl)-6-methoxyphenol (H_2L17)

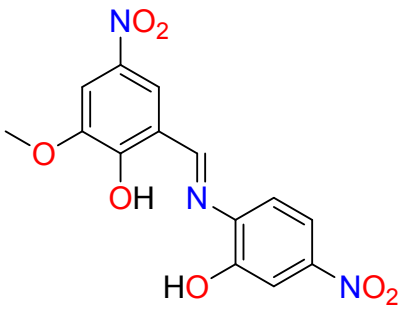
5-bromo-3-methoxy salilyldahyde (0.0125mol, 2.333g) and 2-amino-5-nitrophenol (0.0125mol, 1.926g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time the orange solution had formed a red precipitate. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 79%.



^{1}H NMR (500 MHz,) δ 13.16 (s, 1H), 10.76 (s, 1H), 8.98 (s, 1H), 7.81 – 7.73 (m, 2H), 7.55 – 7.47 (m, 2H), 7.25 (d, *J* = 2.3 Hz, 1H), 3.85 (s, 3H);^{13}C NMR (126 MHz, dmso) δ 190.35, 150.67, 150.30, 145.99, 142.89, 121.73, 120.18, 118.73, 111.63, 110.89, 109.12, 57.07, 40.53, 40.45, 40.36, 40.29, 40.19, 40.12, 40.03, 39.95, 39.86, 39.69, 39.53.

(E)-2-(((2-hydroxy-4-nitrophenyl)imino)methyl)-6-methoxy-4-nitrophenol (H_2L7)

2-hydroxy-3-methoxy-5-nitro benzaldehyde (0.025mol, 3.853g) and 2-amino-5-nitrophenol (0.025mol, 4.923g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time the orange solution had formed a deep orange precipitate. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 95.69%.

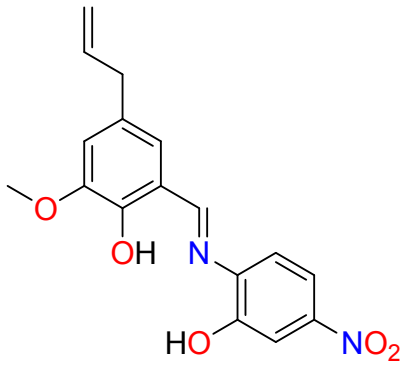


^{1}H NMR (500 MHz,) δ 13.79 (s, 1H), 9.71 (s, 1H), 8.90 (s, 1H), 7.35 (d, *J* = 7.8 Hz, 1H), 7.11 (t, *J* = 7.8 Hz, 1H), 7.01 – 6.83 (m, 4H), 6.04 – 5.92 (m, 1H), 5.14 – 5.03 (m, 2H), 3.79 (d, *J* = 1.3 Hz, 3H); ^{13}C NMR (126 MHz, dmso) δ 189.82, 149.55, 145.99, 142.88, 122.04, 118.73, 116.25, 111.63,

110.78, 109.12, 57.28, 40.61, 40.52, 40.44, 40.35, 40.27, 40.18, 40.10, 40.02, 39.94, 39.85, 39.68, 39.51.

(E)-4-allyl-2-(((2-hydroxy-4-nitrophenyl)imino)methyl)-6-methoxyphenol (H_2L12)

5-allyl-2 hydroxy-3-methoxy benzaldehyde (0.0125mol, 2.290g) and 2-amino-5-nitrophenol (0.0125mol, 1.926g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time the dark brown solution had formed a dark red precipitate. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 72%.

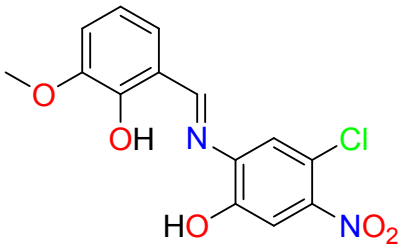


^{1}H NMR (500 MHz,) δ 12.94 (s, 1H), 10.69 (s, 1H), 8.97 (s, 1H), 7.79 – 7.73 (m, 2H), 7.56 – 7.50 (m, 1H), 7.07 (d, *J* = 1.9 Hz, 1H), 6.98 (d, *J* = 2.1 Hz, 1H), 5.98 (ddt, *J* = 16.7, 9.7, 6.6 Hz, 1H), 5.15 – 5.04 (m, 2H), 3.81 (s, 3H), 3.37 – 3.31 (m, 2H).

(E)-4-chloro-2-((2-hydroxy-3-methoxybenzylidene)amino)-5-nitrophenol (H_2L3)

O-Vanillin (0.01mol, 1.541g) and 2-amino-4-chloro-5-nitro phenol (0.01mol, 1.541g) were suspended in MeOH(5mL). The suspension was refluxed for 1h, during which time the dark brown

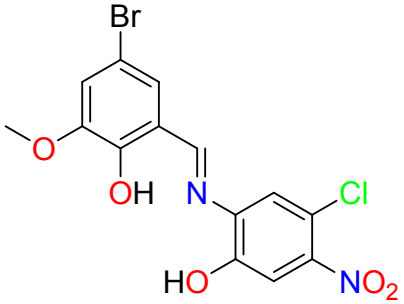
solution had formed a grey precipitate. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 76%.



^{1}H NMR (500 MHz,) δ 12.95 (s, 1H), 10.89 (s, 1H), 9.02 (s, 1H), 7.72 (s, 1H), 7.59 (s, 1H), 7.25 (dd, *J* = 7.9, 1.5 Hz, 1H), 7.16 (dd, *J* = 8.0, 1.5 Hz, 1H), 6.92 (t, *J* = 7.9 Hz, 1H), 3.82 (s, 3H), 2.49 (s, 3H); ^{13}C NMR (126 MHz, dmso) δ 192.46, 142.07, 122.97, 120.62, 119.65, 119.11, 118.10, 113.68, 111.69, 109.99, 56.57, 56.42, 40.62, 40.53, 40.45, 40.36, 40.29, 40.20, 40.12, 40.03, 39.96, 39.86, 39.69, 39.53.

(E)-2-((5-bromo-2-hydroxy-3-methoxybenzylidene)amino)-4-chloro-5-nitrophenol (H_2L18)

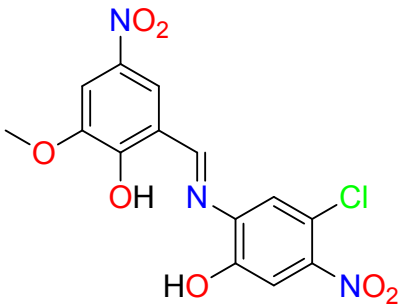
5-bromo-3-methoxy saliyladehyde (0.0072mol, 1.650g) and 2-amino-4-chloro-5-nitro phenol were dissolved in MeOH (4mL). The suspension as refluxed for 1H, during which time orange-red solid precipitated. After cooling to room temperature, the solid was filtered off and washed with Et_2O and dried in vacuo. Yield 35%.



^{1}H NMR (500 MHz,) δ 12.85 – 12.80 (m, 2H), 10.91 (s, 2H), 8.97 (d, *J* = 1.4 Hz, 2H), 7.69 (d, *J* = 1.6 Hz, 2H), 7.59 (s, 2H), 7.48 (d, *J* = 2.2 Hz, 2H), 7.27 (d, *J* = 2.2 Hz, 2H), 3.85 (s, 6H), 3.29 (s, 2H), 3.19 – 3.14 (m, 1H); ^{13}C NMR (126 MHz, dmso) δ 190.35, 164.19, 150.67, 150.30, 145.41, 142.07, 125.41, 124.07, 122.71, 121.73, 120.71, 120.17, 118.70, 113.68, 113.63, 111.69, 110.89, 57.06, 56.80, 40.62, 40.53, 40.45, 40.36, 40.28, 40.19, 40.12, 40.03, 39.94, 39.86, 39.69, 39.52.

(E)-4-chloro-2-((2-hydroxy-3-methoxy-5-nitrobenzylidene)amino)-5-nitrophenol (H_2L8)

2-hydroxy-3-methoxy-5-nitro benzaldehyde (0.025mol, 3.853g) and 2-amino-4-chloro-5-nitro phenol (0.025mol, 4.71g) were dissolved in MeOH (4mL). The suspension was refluxed for 1H, during which time dark brow solid precipitated. After cooling to room temperature, the solid was filtered off and washed with Et_2O and dried in vacuo. Yield 80%.

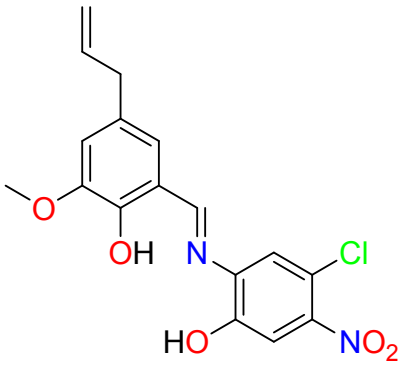


^{1}H NMR (500 MHz,) δ 12.70 (s, 1H), 10.86 (s, 1H), 8.97 (s, 1H), 7.71 (s, 1H), 7.59 (s, 1H), 7.06 (d, *J* = 2.0 Hz, 1H), 6.99 (d, *J* = 2.0 Hz, 1H), 5.98 (ddt, *J* = 16.7, 9.8, 6.6 Hz, 1H), 5.15 – 5.04 (m, 2H), 3.81 (s, 3H), 3.34 (d, *J* = 6.6 Hz, 2H).

(E)-2-((5-allyl-2-hydroxy-3-methoxybenzylidene)amino)-4-chloro-5-nitrophenol (H_2L13)

2-hydroxy-3-methoxy-5-allyl-benzaldehyde (0.025mol,4.81g) and 2-amino-4-chloro-5-nitro phenol (0.025mol, 4.71g) were dissolved in MeOH (4mL). The suspension was refluxed for 1H, during

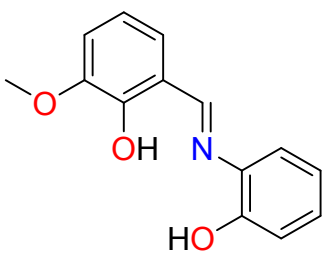
which time a cream brown solid precipitated. After cooling to room temperature, the solid was filtered off and washed with Et_2O and dried in vacuo. Yield 70%.



^{1}H NMR (500 MHz,) δ 12.70 (s, 1H), 10.86 (s, 1H), 8.97 (s, 1H), 7.71 (s, 1H), 7.59 (s, 1H), 7.06 (d, *J* = 2.0 Hz, 1H), 6.99 (d, *J* = 2.0 Hz, 1H), 5.98 (ddt, *J* = 16.7, 9.8, 6.6 Hz, 1H), 5.15 – 5.04 (m, 2H), 3.81 (s, 3H), 3.34 (d, *J* = 6.6 Hz, 2H); ^{13}C NMR (126 MHz, dmso) δ 192.25, 149.64, 148.79, 145.41, 142.08, 137.90, 133.24, 131.18, 122.66, 120.71, 119.51, 118.56, 116.44, 113.68, 111.69, 56.56, 40.61, 40.52, 40.44, 40.35, 40.27, 40.19, 40.11, 40.02, 39.94, 39.85, 39.68, 39.52, 39.19.

(E)-2-(((2-hydroxyphenyl)imino)methyl)-6-methoxyphenol (H_2L1)

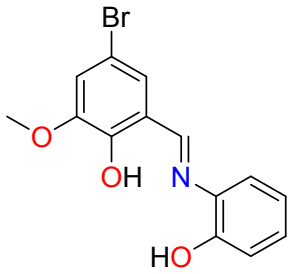
O-Vanillin (0.025mol, 3.35g) and 2-amino-phenol (0.025mol, 2.73g) were dissolved in MeOH (5mL) .The suspension was refluxed for 1h, during which time a bright orange solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 95%.



^{1}H NMR (500 MHz, DMSO-d_6) δ 9.75 – 9.71 (m, 1H), 8.95 (s, 1H), 7.36 (dd, J = 8.0, 1.6 Hz, 1H), 7.21 – 6.99 (m, 3H), 6.96 (dd, J = 8.1, 1.4 Hz, 1H), 6.91 – 6.81 (m, 2H), 3.80 (s, 3H); ^{13}C NMR (126 MHz, dmso) δ 162.01, 152.27, 151.42, 148.63, 134.95, 128.46, 124.26, 120.05, 119.96, 119.70, 118.37, 116.97, 115.71, 56.34, 40.62, 40.53, 40.45, 40.36, 40.28, 40.19, 40.11, 40.03, 39.95, 39.86, 39.69, 39.53.

(E)-4-bromo-2-(((2-hydroxyphenyl)imino)methyl)-6-methoxyphenol (H_2L16)

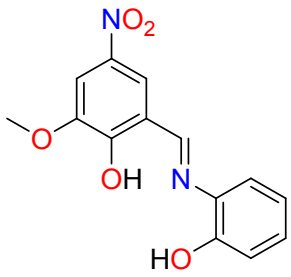
5-bromo-3-methoxy salilcyldahyde (0.025mol, 4.666g) and 2-amino-phenol (0.025mol, 2.73g) were dissolved in MeOH (5mL). The suspension was refluxed for 1H, during which time a red solid precipitated. After cooling to room temperature, the solid was filtered off and washed with Et_2O and dried in vacuo. Yield 84%.



^{1}H NMR (500 MHz, DMSO-d_6) δ 9.85 (s, 1H), 8.94 (d, J = 1.7 Hz, 1H), 7.42 – 7.34 (m, 2H), 7.18 – 7.10 (m, 2H), 6.96 (dd, J = 8.0, 1.5 Hz, 1H), 6.88 (t, J = 7.6 Hz, 1H), 3.82 (s, 3H), 2.49 (s, 1H); ^{13}C NMR (126 MHz, dmso) δ 160.11, 152.85, 151.42, 150.09, 133.98, 128.85, 125.58, 120.42, 120.10, 119.68, 117.71, 117.01, 108.62, 56.64, 40.62, 40.53, 40.45, 40.36, 40.28, 40.20, 40.11, 40.03, 39.95, 39.86, 39.70, 39.53.

(E)-2-(((2-hydroxyphenyl)imino)methyl)-6-methoxy-4-nitrophenol (H_2L6)

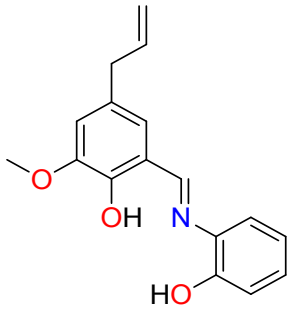
2-hydroxy-3-methoxy-5-nitro benzaldehyde (0.025mol, 3.853g) and 2-amino-phenol (0.025mol, 2.73g) were dissolved in MeOH (5mL) .The suspension was refluxed for 1h, during which time a clay brown solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 93%.



^{1}H NMR (500 MHz, DMSO-d_6) δ 15.63 (d, J = 10.3 Hz, 1H), 10.68 (s, 1H), 9.33 (d, J = 9.1 Hz, 1H), 8.25 (d, J = 2.6 Hz, 1H), 7.70 (d, J = 8.1 Hz, 1H), 7.47 (d, J = 2.7 Hz, 1H), 7.22 (t, J = 7.4 Hz, 1H), 7.11 – 6.86 (m, 2H), 3.83 (s, 3H). ^{13}C NMR (126 MHz, dmso) δ 171.05, 158.90, 151.81, 149.73, 134.62, 129.63, 127.09, 125.03, 120.44, 118.44, 116.92, 113.36, 106.48, 56.17, 40.61, 40.52, 40.44, 40.35, 40.28, 40.19, 40.11, 40.02, 39.94, 39.85, 39.69, 39.52.

(E)-4-allyl-2-(((2-hydroxyphenyl)imino)methyl)-6-methoxyphenol (H_2L11)

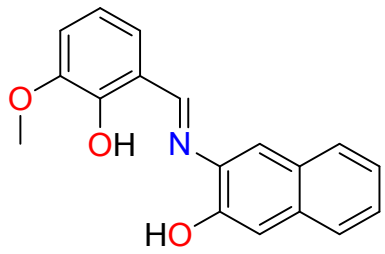
2-hydroxy-3-methoxy-5-allyl-benzaldehyde (0.025mol,4.81g) and 2-aminophenol (0.025mol, 2.73g) were dissolved in MeOH (5mL). The suspension was refluxed for 1h, during which time a bright red solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 70%.



^{1}H NMR (500 MHz,) δ 13.80 (s, 1H), 9.71 (s, 1H), 8.90 (s, 1H), 7.38 – 7.32 (m, 1H), 7.14 – 7.07 (m, 1H), 7.01 – 6.83 (m, 4H), 6.04 – 5.92 (m, 1H), 5.14 – 5.03 (m, 2H), 3.79 (d, *J* = 1.2 Hz, 3H), 3.35 – 3.28 (m, 3H). ^{13}C NMR (126 MHz, dmso) δ 192.25, 149.64, 148.79, 145.41, 142.08, 137.90, 133.24, 131.18, 122.66, 120.71, 119.51, 118.56, 116.44, 113.68, 111.69, 56.56, 40.61, 40.52, 40.44, 40.35, 40.27, 40.19, 40.11, 40.02, 39.94, 39.85, 39.68, 39.52, 39.19.

(E)-3-((2-hydroxy-3-methoxybenzylidene)amino)naphthalen-2-ol (H_2L5)

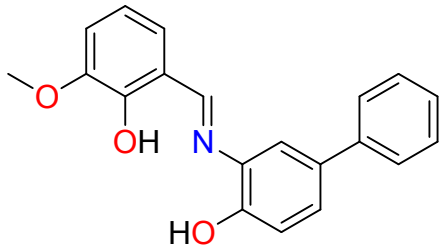
O-Vanillin (0.025mol, 3.35g) and 3-amino-2-napthol (0.025mol, 3.98g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time a bright orange solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 88%.



^{1}H NMR (500 MHz, DMSO-d_6) δ 13.78 (s, 0H), 10.12 (s, 0H), 9.07 (s, 1H), 7.85 – 7.77 (m, 2H), 7.69 (d, *J* = 8.2 Hz, 1H), 7.41 – 7.34 (m, 1H), 7.33 – 7.22 (m, 3H), 7.12 (dd, *J* = 8.0, 1.3 Hz, 1H), 6.89 (t, *J* = 7.9 Hz, 1H), 3.83 (s, 3H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 163.49 , 152.35 , 150.36 , 128.07 , 126.38 , 126.17 , 124.37 , 123.85 , 118.60 , 117.52 , 116.06 , 110.42 , 56.38 , 40.56 , 40.31 (d, *J* = 20.9 Hz), 40.06 .

(E)-3-((2-hydroxy-3-methoxybenzylidene)amino)-[1,1'-biphenyl]-4-ol (H_2L4)

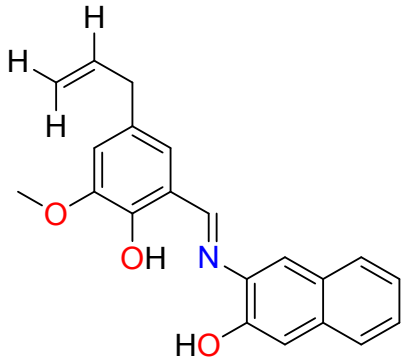
O-Vanillin (0.025mol, 3.35g) and 2-amino-5-phenylphenol (0.025mol,4.63g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time a dark orange solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 91%.



^{1}H NMR (500 MHz, DMSO-d_6) δ 9.92 (s, 1H), 9.10 (d, J = 2.0 Hz, 1H), 7.72 – 7.65 (m, 3H), 7.50 – 7.27 (m, 4H), 7.21 (dt, J = 7.8, 1.4 Hz, 1H), 7.12 – 7.02 (m, 2H), 6.87 (tt, J = 7.8, 1.3 Hz, 1H), 3.84 – 3.80 (m, 3H), 3.17 (s, 1H), -1.96 (s, 0H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 162.56 , 152.41 , 151.14 , 140.22 , 135.32 , 129.22 , 127.17 , 126.64 (d, J = 9.2 Hz), 124.37 , 119.77 , 118.39 , 118.05 , 117.39 , 115.83 , 56.37 , 40.55 , 40.38 , 40.21 , 40.05 .

(E)-3-((5-allyl-2-hydroxy-3-methoxybenzylidene)amino)naphthalen-2-ol (H_2L15)

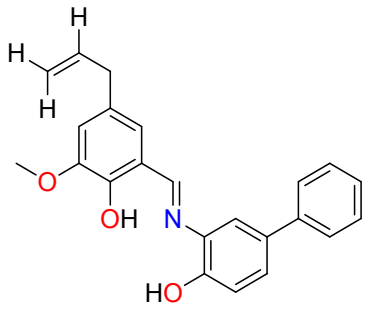
2-hydroxy-3-methoxy-5-allyl-benzaldehyde (0.025mol,4,81g) and 3-amino-2-napthol (0.025mol,3.98g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time a dark red solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 61%.



^{1}H NMR (500 MHz, Chloroform-*d*) δ 10.96 (s, 0H), 9.89 (d, *J* = 0.9 Hz, 0H), 8.80 (s, 1H), 7.77 (d, *J* = 8.2 Hz, 1H), 7.72 (d, *J* = 8.3 Hz, 1H), 7.55 (s, 1H), 7.47 – 7.30 (m, 4H), 7.23 (s, 0H), 7.02 – 6.92 (m, 2H), 6.88 (d, *J* = 1.6 Hz, 1H), 6.06 – 5.92 (m, 2H), 5.18 – 5.08 (m, 3H), 3.93 (d, *J* = 7.8 Hz, 4H), 3.39 (dd, *J* = 7.0, 5.0 Hz, 3H).

(E)-3-((5-allyl-2-hydroxy-3-methoxybenzylidene)amino)-[1,1'-biphenyl]-4-ol (H_2L14)

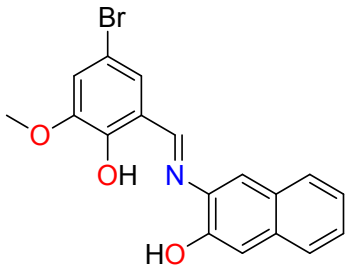
2-hydroxy-3-methoxy-5-allyl-benzaldehyde (0.025mol,4,81g) and 2-amino-5-phenylphenol (0.025mol,4.63g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time a dark orange solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 69%.



^{1}H NMR (500 MHz, Chloroform-*d*) δ 10.96 (s, 0H), 8.76 (s, 1H), 7.61 – 7.51 (m, 2H), 7.49 – 7.41 (m, 2H), 7.40 – 7.30 (m, 2H), 7.27 (s, 1H), 7.12 (d, *J* = 8.3 Hz, 1H), 7.02 – 6.94 (m, 1H), 6.89 (dd, *J* = 21.8, 1.9 Hz, 2H), 5.99 (ddt, *J* = 17.4, 9.2, 6.7 Hz, 1H), 5.13 (ddq, *J* = 12.9, 4.1, 1.9 Hz, 2H), 3.93 (d, *J* = 6.5 Hz, 3H), 3.39 (d, *J* = 6.6 Hz, 2H).

(E)-3-((5-bromo-2-hydroxy-3-methoxybenzylidene)amino)naphthalen-2-ol (H_2L20)

5-bromo-3-methoxy salilcyldahyde (0.025mol, 4.666g) and 3-amino-2-napthol (0.025mol,3.98g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during which time a dark red solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 79%.

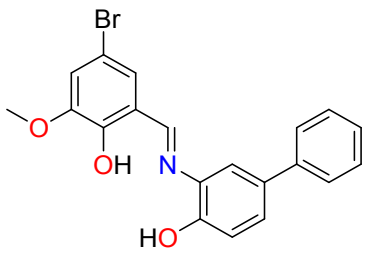


^{1}H NMR (500 MHz, DMSO-d_6) δ 13.90 (s, 1H), 10.14 (s, 1H), 9.06 (s, 1H), 7.86 – 7.77 (m, 2H), 7.70 (d, *J* = 8.2 Hz, 1H), 7.50 – 7.45 (m, 1H), 7.39 (t, *J* = 7.5 Hz, 1H), 7.34 – 7.26 (m, 2H), 7.21 (dd, *J* = 2.2, 1.1 Hz, 1H), 3.85 (s, 3H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 161.63 , 128.11 , 126.55 , 126.23 , 125.63 , 123.98 , 118.06 , 117.46 , 56.71 , 40.55 , 40.30 (d, *J* = 21.0 Hz).

(E)-3-((5-bromo-2-hydroxy-3-methoxybenzylidene)amino)-[1,1'-biphenyl]-4-ol (H_2L19)

5-bromo-3-methoxy salilcyldahyde (0.025mol, 4.666g) and 2-amino-5-phenylphenol (0.025mol,4.63g) were dissolved in MeOH (5mL) . The suspension was refluxed for 1h, during

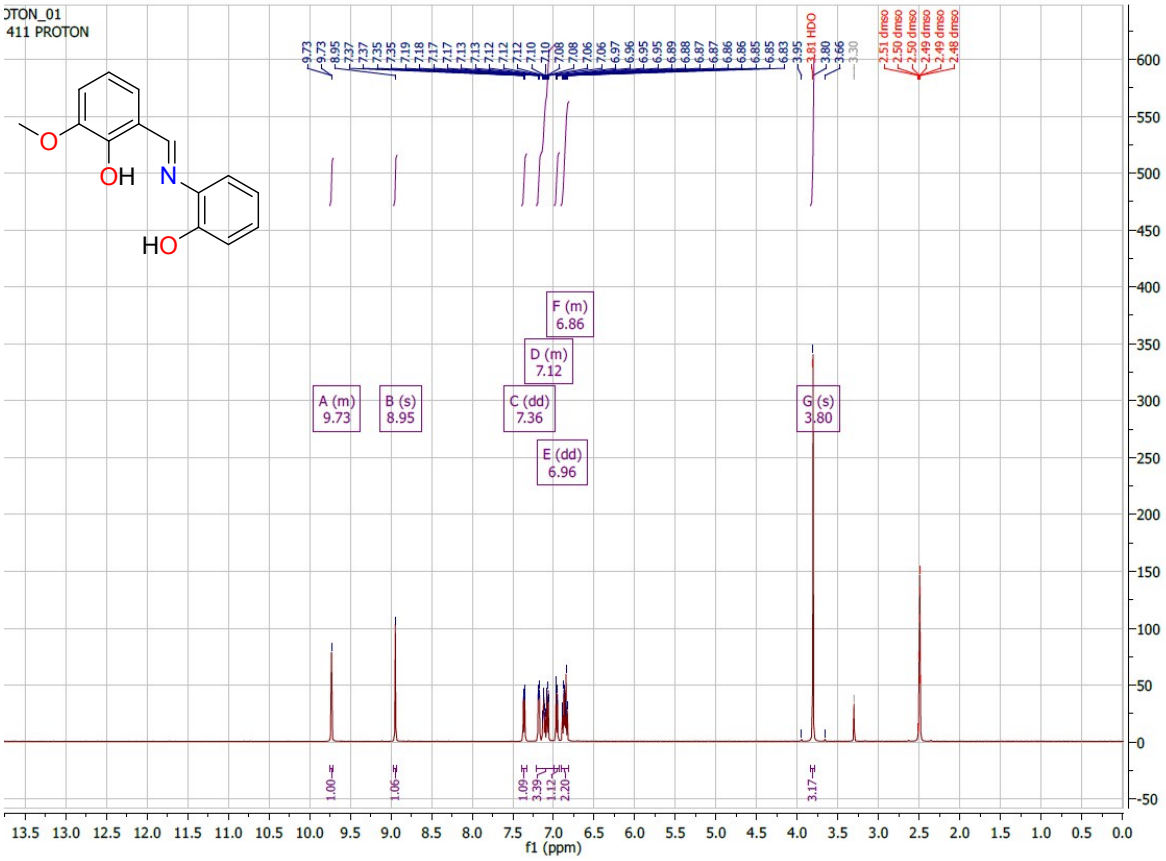
which time a dark orange solid precipitated. After cooling to room temperature the solid was filtered off and washed with cold MeOH and Et_2O. The solid was dried in vacuo. Yield 83%.

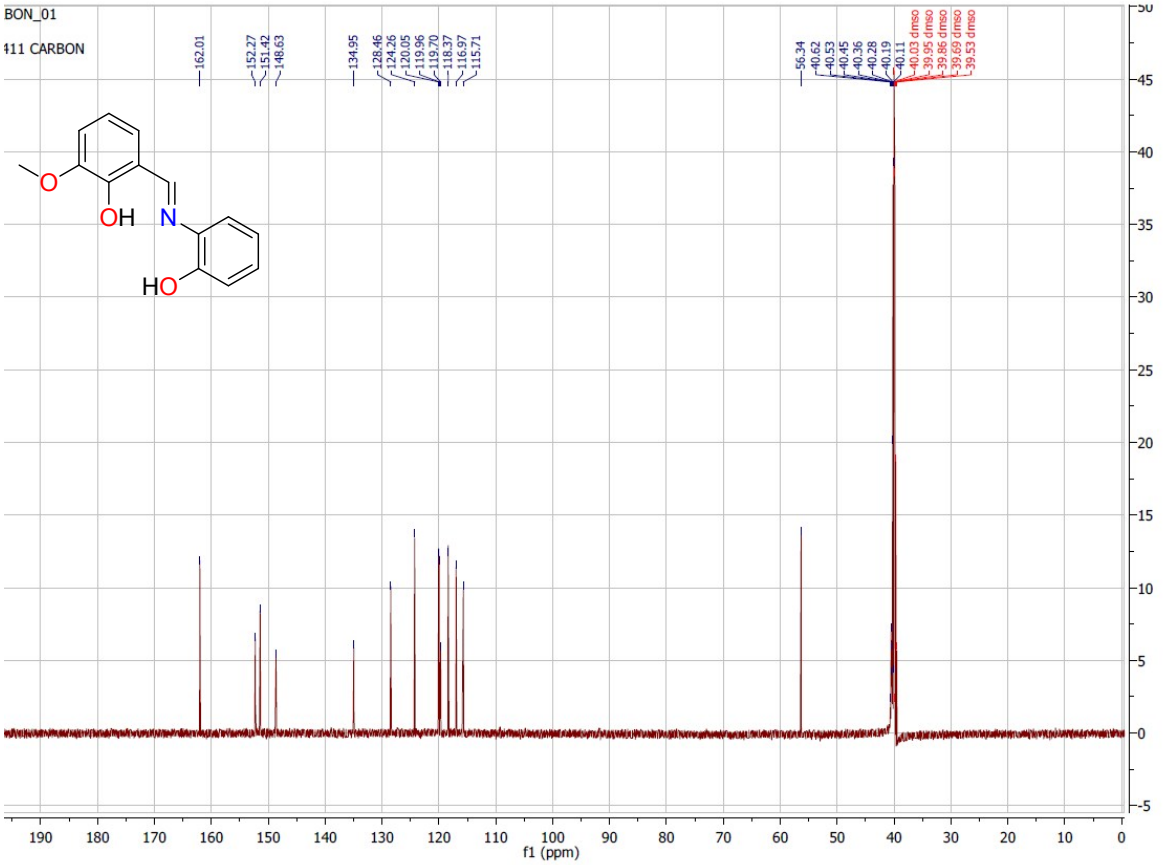


^{1}H NMR (500 MHz, DMSO-d_6) δ 10.03 (s, 1H), 9.10 (s, 1H), 7.74 – 7.65 (m, 3H), 7.50 – 7.39 (m, 4H), 7.35 – 7.27 (m, 1H), 7.18 (d, J = 2.3 Hz, 1H), 7.06 (d, J = 8.4 Hz, 1H), 4.07 (p, J = 5.1, 4.3 Hz, 1H), 3.84 (s, 3H), 3.20 – 3.15 (m, 2H); ^{13}C NMR (126 MHz, DMSO-d_6) δ 160.71 , 152.98 , 151.11 , 150.14 , 140.11 , 134.39 , 132.31 , 129.24 , 127.24 , 126.97 , 126.65 , 125.73 , 120.46 , 117.82 , 117.45 , 56.67 , 40.46 (d, J = 21.1 Hz), 40.21 .

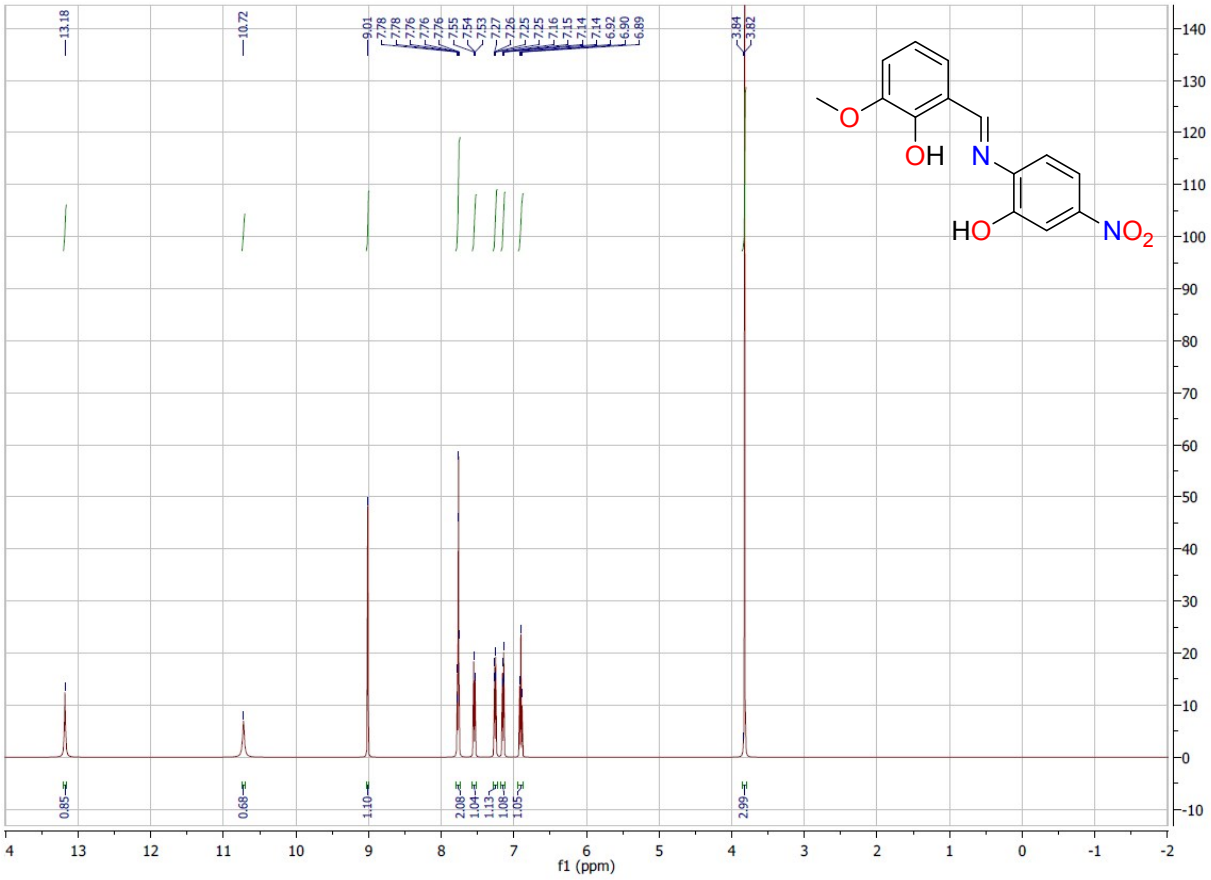
^{1}H and ^{13}C NMR of Ligands

H_2L1

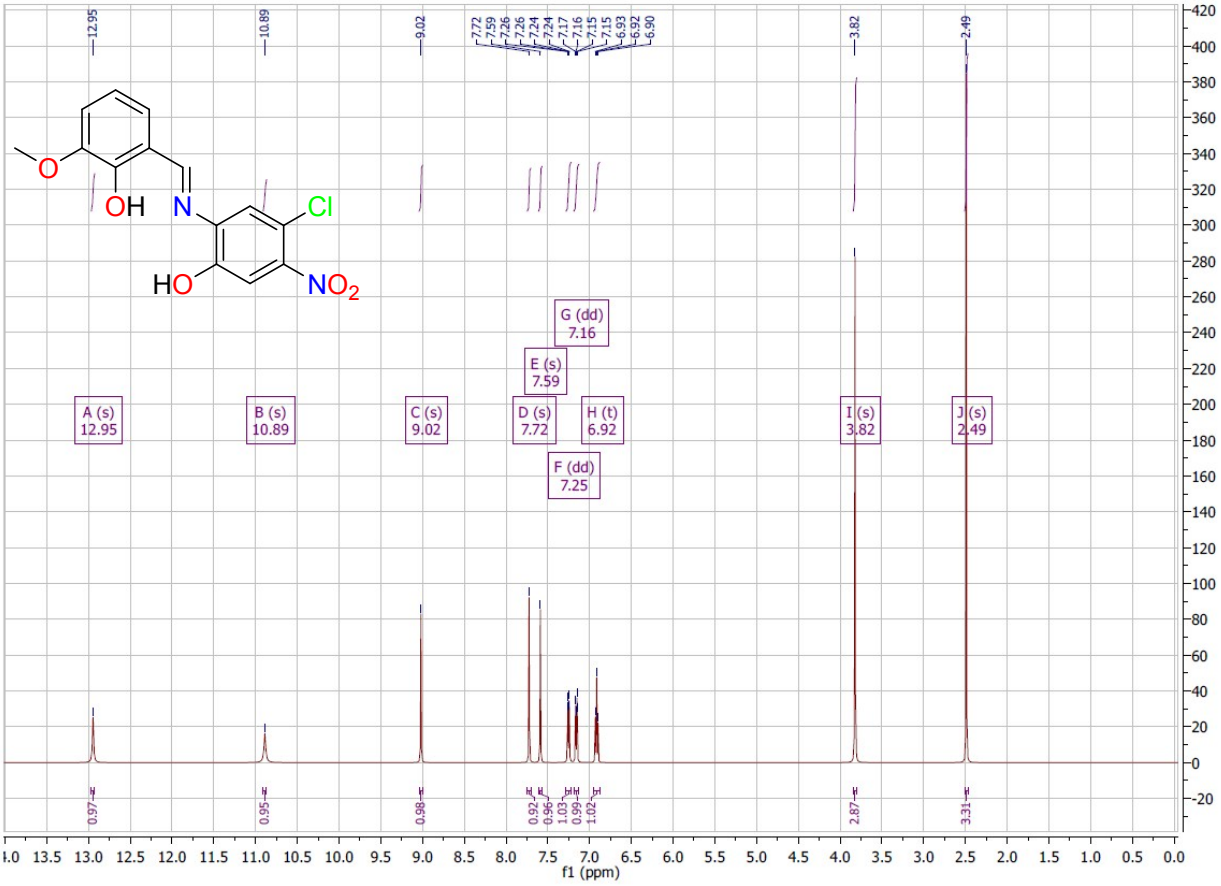

OTON_01
411 PROTON
F (m)
6.86
D (m)
7.12
A (m)
9.73
B (s)
8.95
C (dd)
7.36
G (s)
3.80
E (dd)
6.96
f1 (ppm)

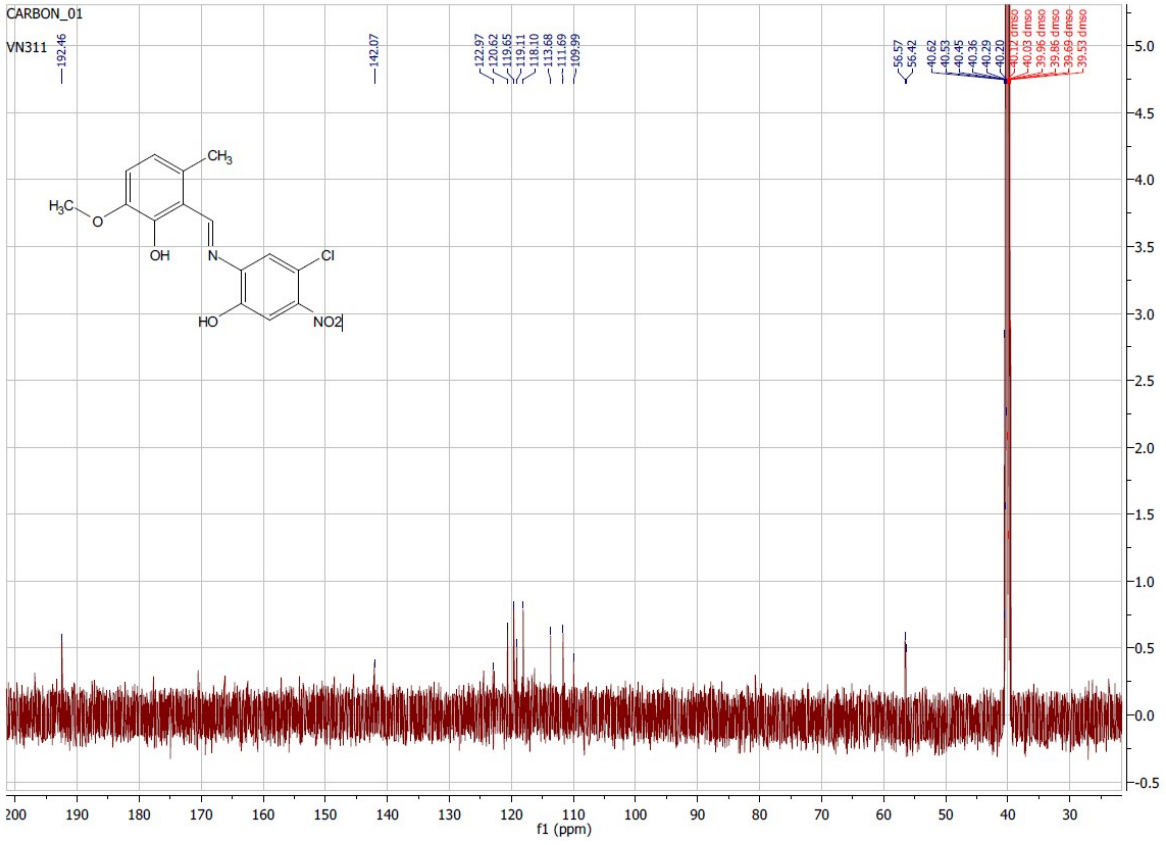

BON_01
411 CARBON
f1 (ppm)

H_2L2

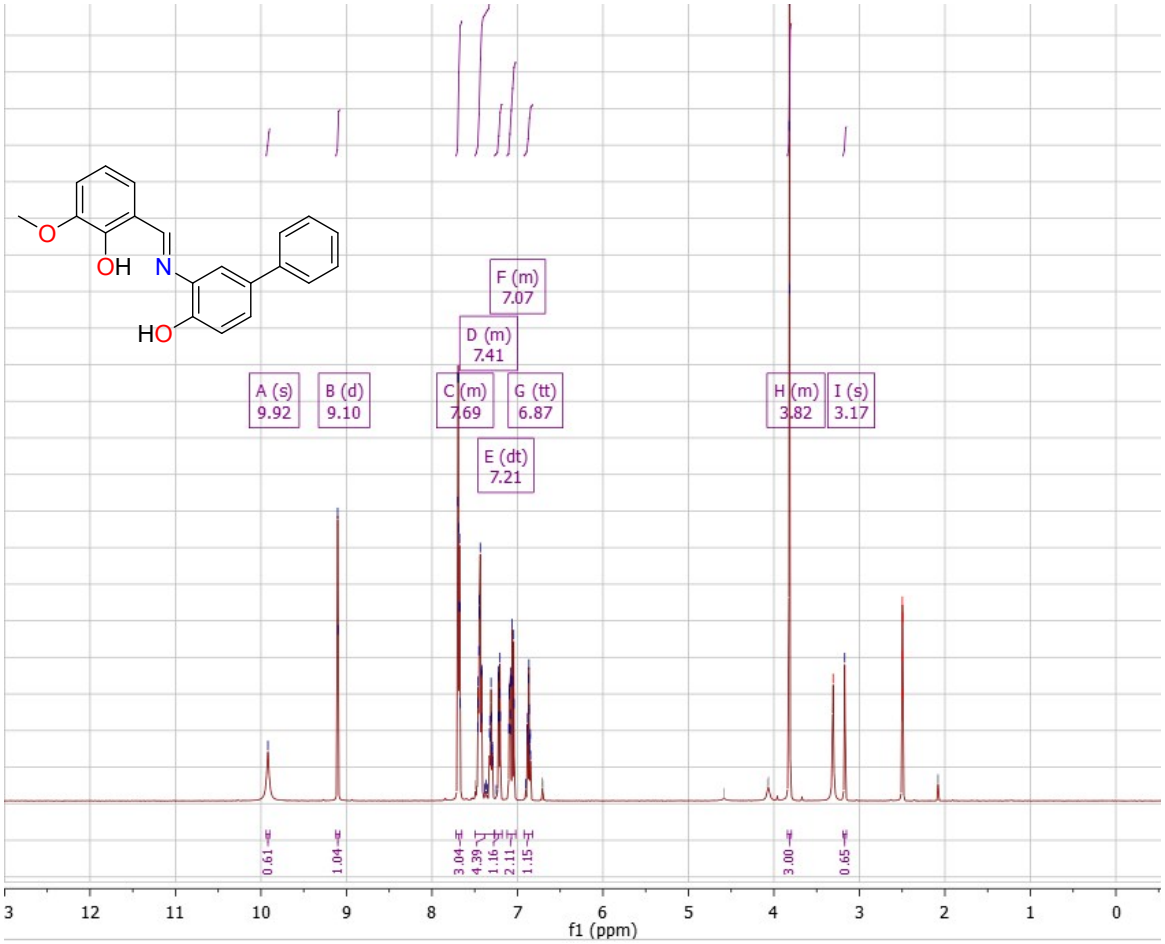

13.18
10.72
9.01
7.78
7.78
7.78
7.76
7.76
7.76
7.76
7.55
7.54
7.53
7.27
7.26
7.25
7.25
7.25
7.16
7.15
7.14
7.14
6.92
6.90
6.89
3.84
3.82
O
OH
N
HO
NO2
0.85
0.68
1.10
2.08
1.04
1.13
1.08
1.05
2.99
f1 (ppm)

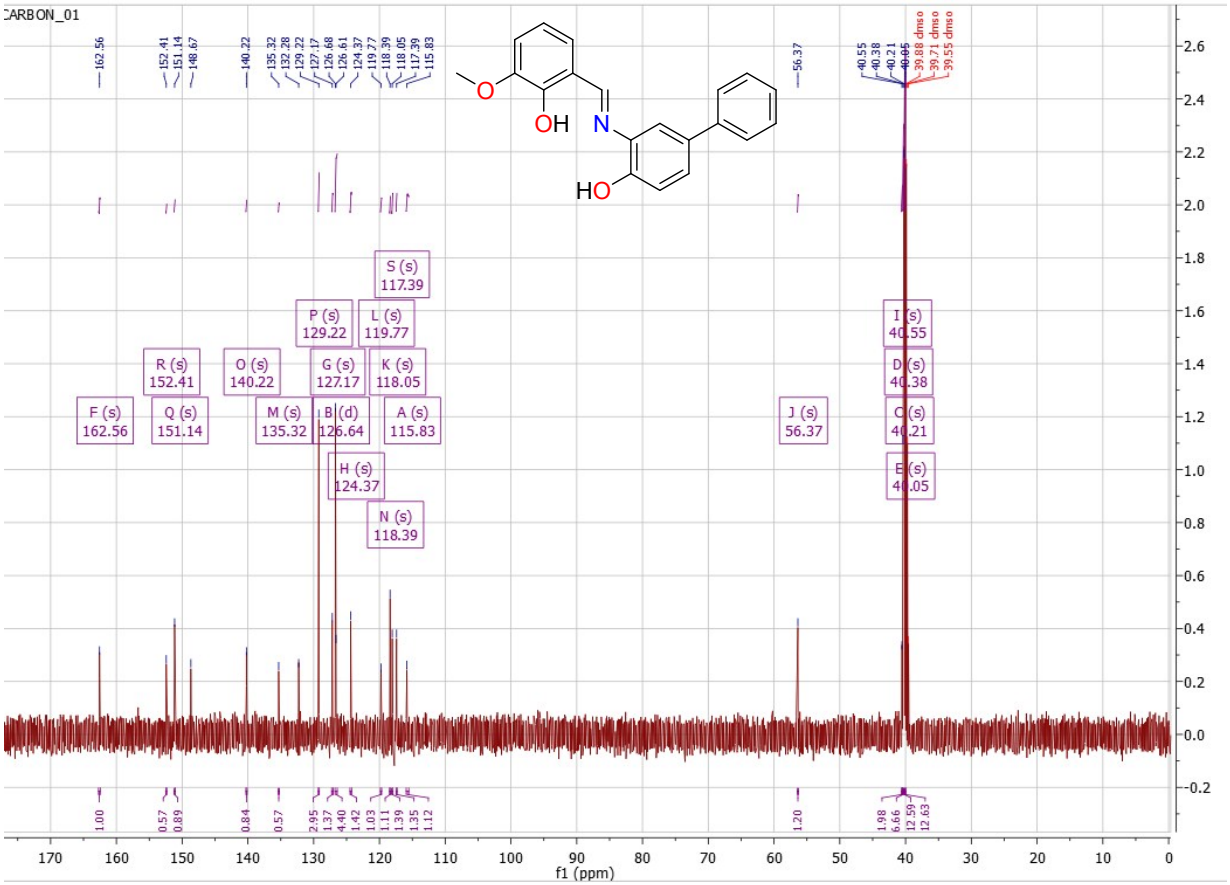
H_2L3


12.95
10.89
9.02
7.72
7.59
7.26
7.24
7.17
7.16
7.15
6.93
6.92
6.90
3.82
2.49
O
OH
N
Cl
HO
NO2
G (dd)
7.16
E (s)
7.59
A (s)
12.95
B (s)
10.89
C (s)
9.02
D (s)
7.72
H (t)
6.92
I (s)
3.82
J (s)
2.49
F (dd)
7.25
0.97
0.95
0.98
0.92
0.96
1.03
0.99
1.02
2.87
3.31
f1 (ppm)

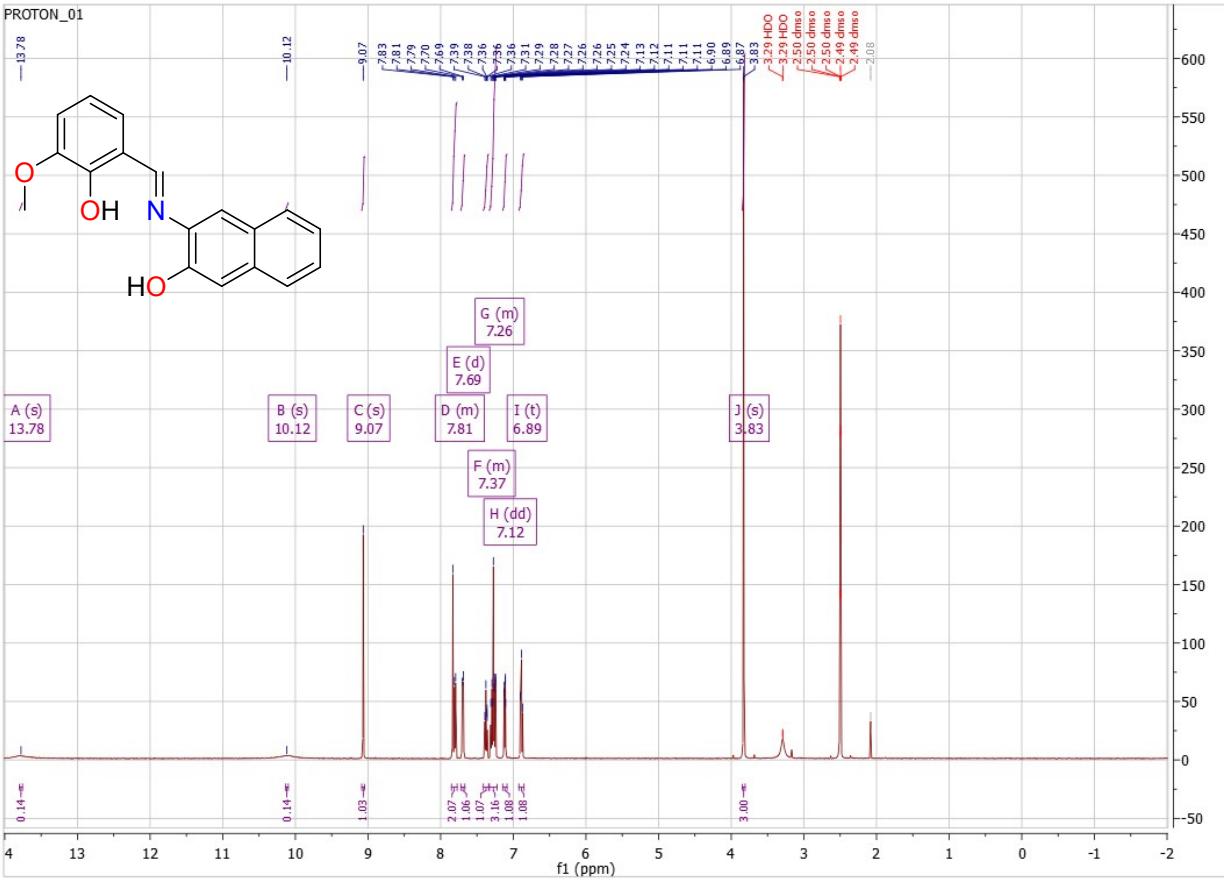

CARBON_01
VN311
192.46
142.07
122.97
120.62
119.65
119.11
118.10
113.68
111.69
109.99
56.57
56.42
40.62
40.53
40.45
40.36
40.29
40.20
40.12 dmso
40.03 dmso
39.96 dmso
39.86 dmso
39.69 dmso
39.53 dmso
CH3
H3C
O
OH
N
Cl
HO
NO2
f1 (ppm)

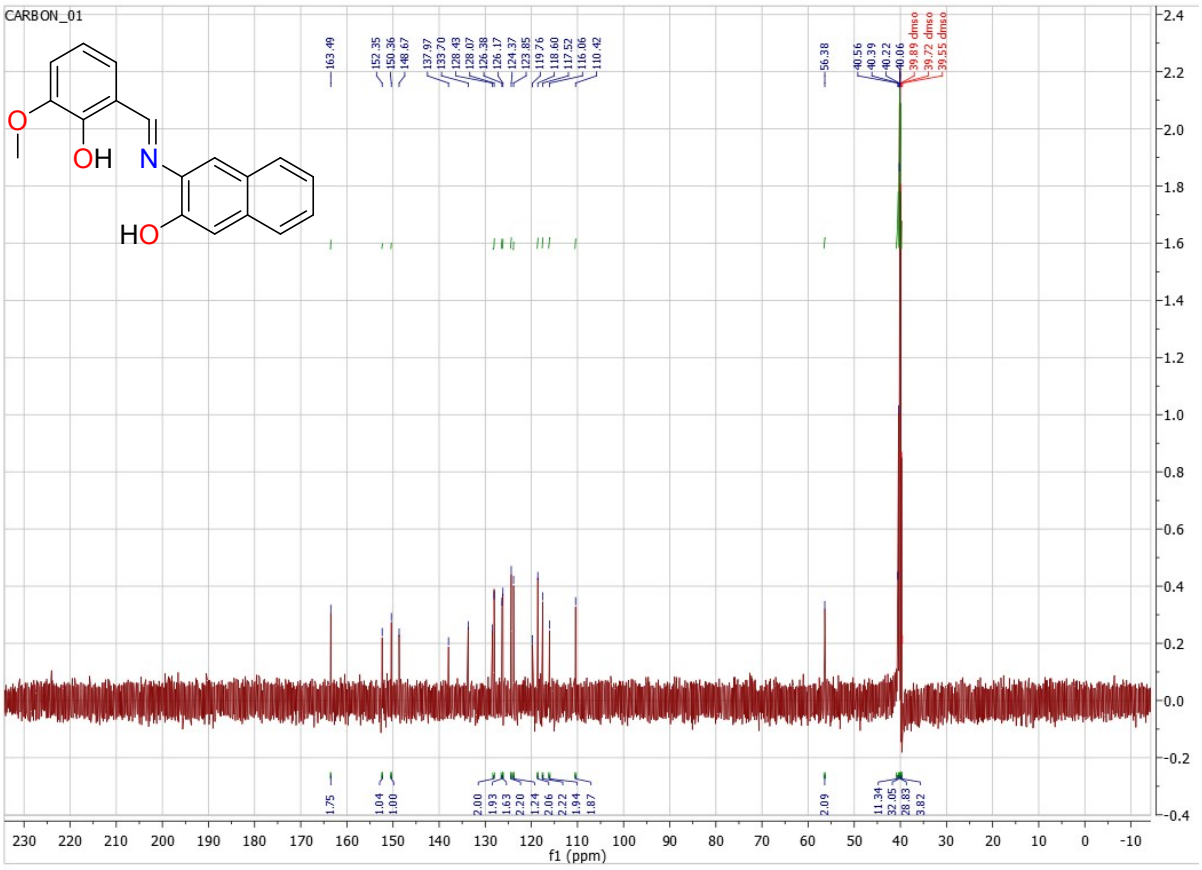
H_2L4


O
OH
N
HO
A (s) 9.92
B (d) 9.10
C (m) 7.69
D (m) 7.41
E (dt) 7.21
F (m) 7.07
G (tt) 6.87
H (m) 3.82
I (s) 3.17
f1 (ppm)

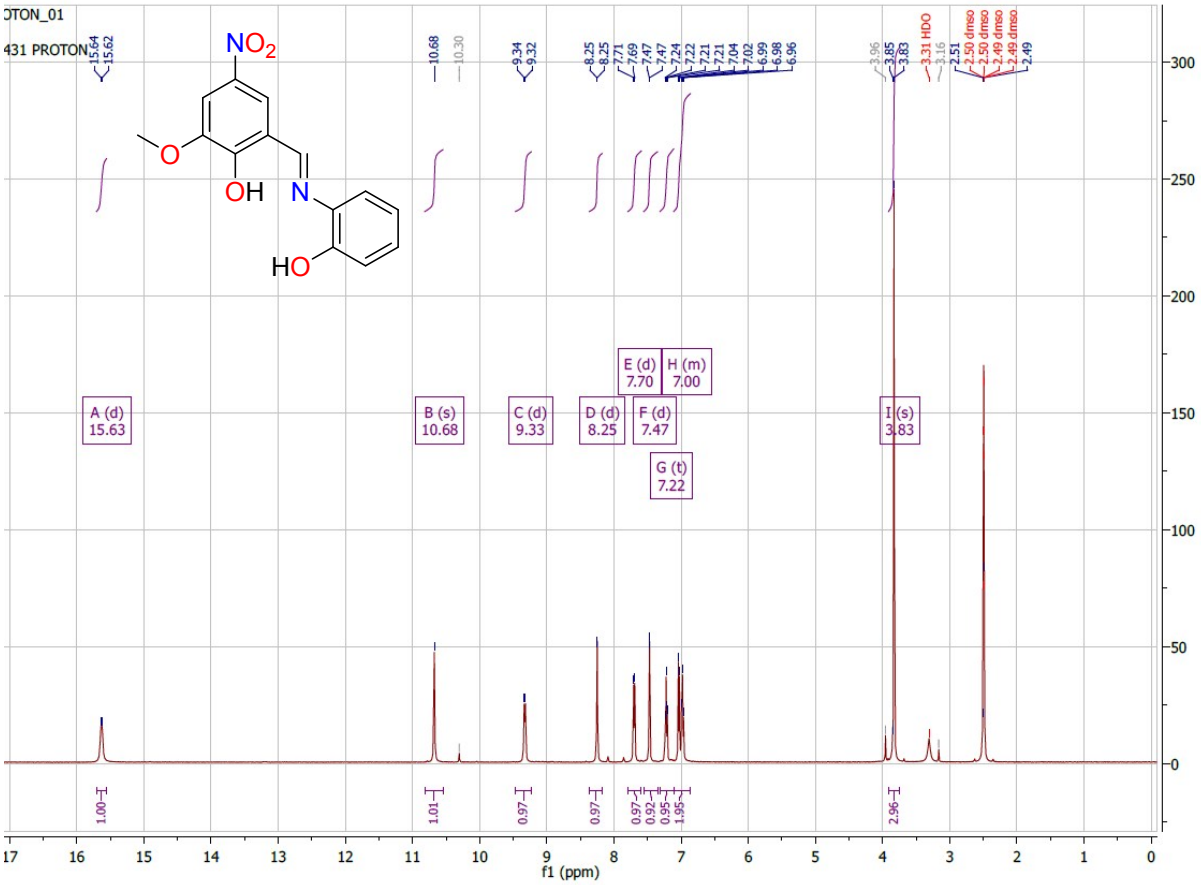

CARBON_01
O
OH
N
HO
S (s) 117.39
P (s) 129.22
L (s) 119.77
R (s) 152.41
O (s) 140.22
G (s) 127.17
K (s) 118.05
F (s) 162.56
Q (s) 151.14
M (s) 135.32
B (d) 126.64
A (s) 115.83
H (s) 124.37
N (s) 118.39
J (s) 56.37
I (s) 40.55
D (s) 40.38
C (s) 40.21
E (s) 40.05
f1 (ppm)

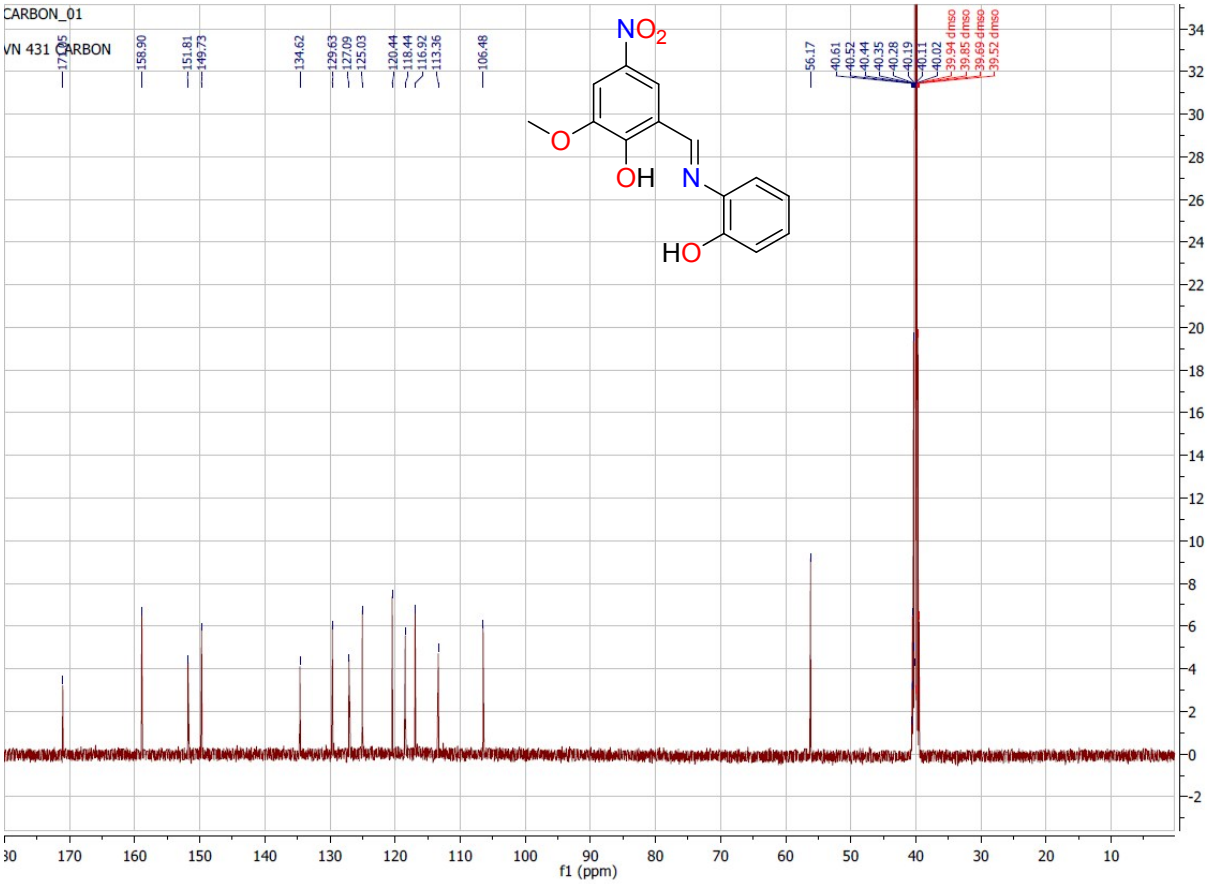
H_2L5


PROTON_01
A (s) 13.78
B (s) 10.12
C (s) 9.07
D (m) 7.81
E (d) 7.69
F (m) 7.37
G (m) 7.26
H (dd) 7.12
I (t) 6.89
J (s) 3.83
f1 (ppm)

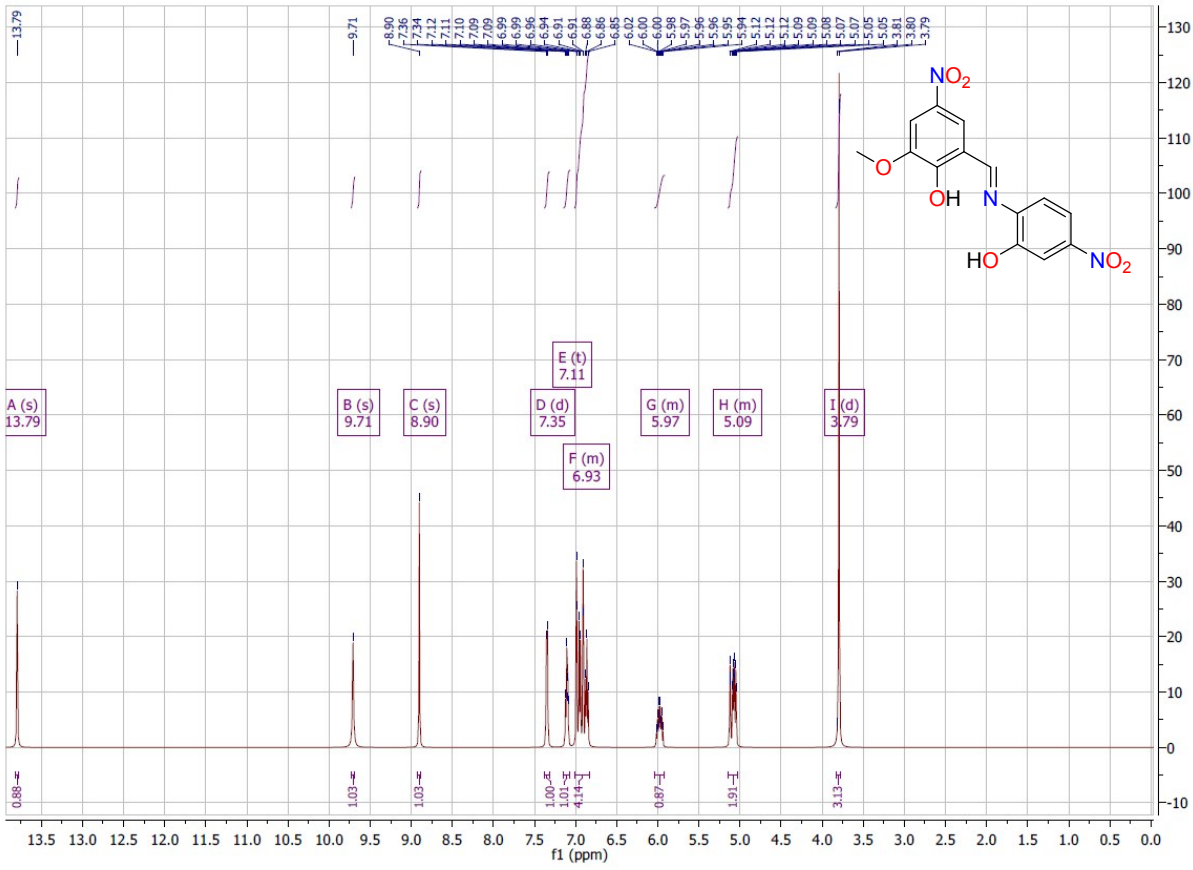

CARBON_01
f1 (ppm)

H_2L6

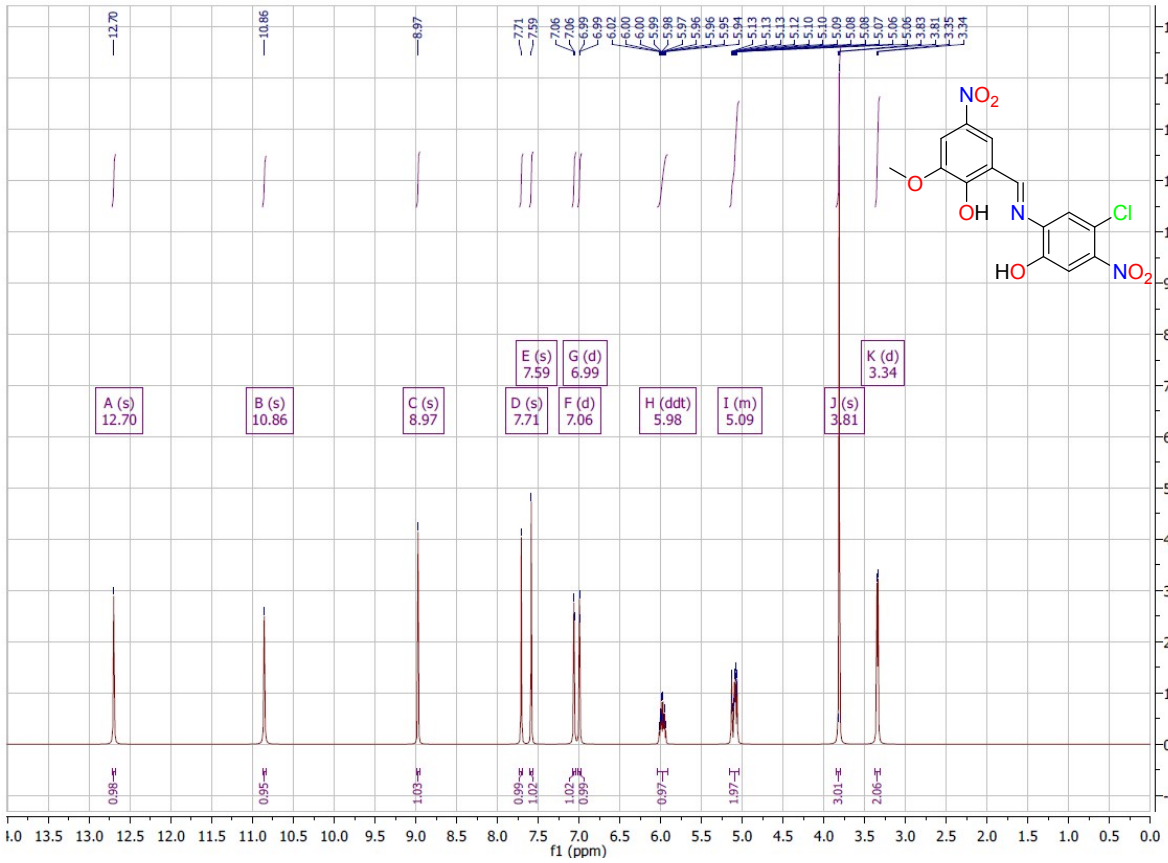

NO2
O
OH
N
HO
A (d) 15.63
B (s) 10.68
C (d) 9.33
D (d) 8.25
E (d) 7.70
F (d) 7.47
G (t) 7.22
H (m) 7.00
I (s) 3.83
f1 (ppm)


CARBON_01
VN 431 CARBON
NO2
O
OH
N
HO
f1 (ppm)

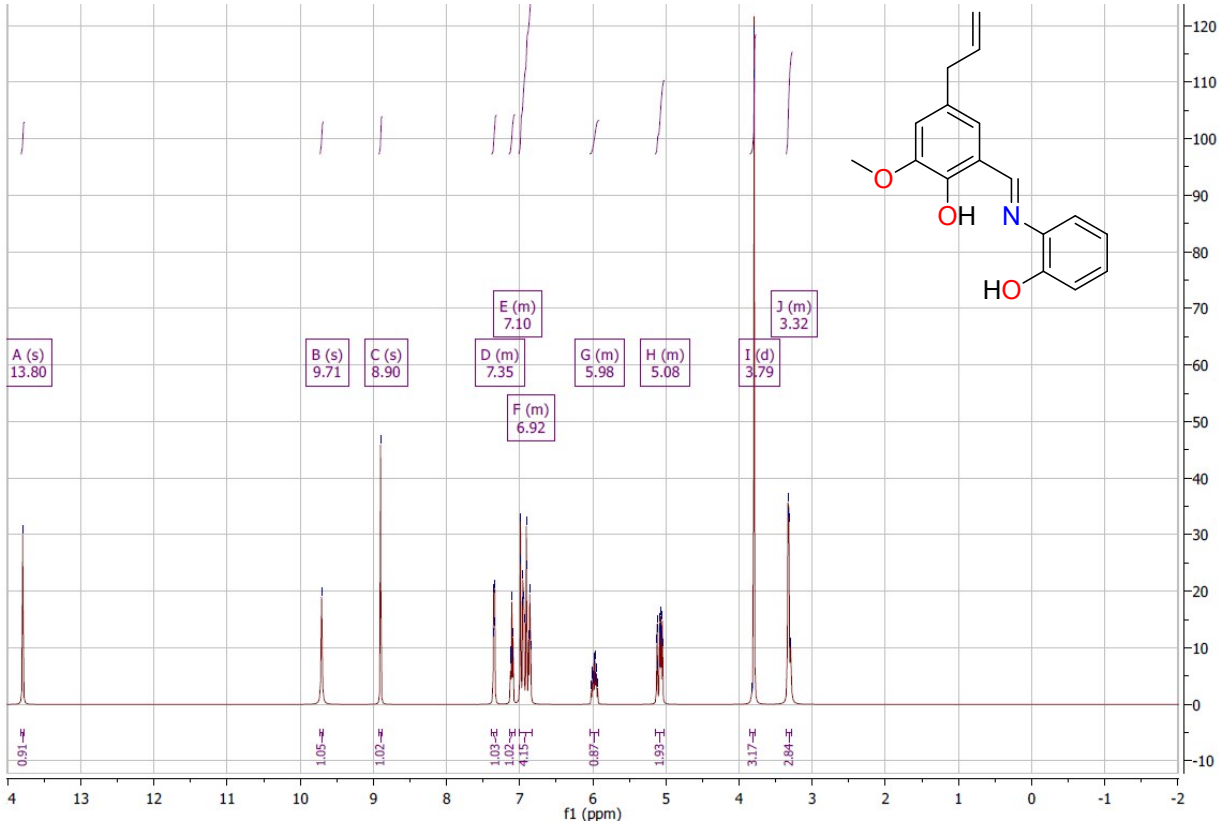
H_2L7


NO2
O
OH
N
HO
NO2
A (s) 13.79
B (s) 9.71
C (s) 8.90
D (d) 7.35
E (t) 7.11
F (m) 6.93
G (m) 5.97
H (m) 5.09
I (d) 3.79
f1 (ppm)

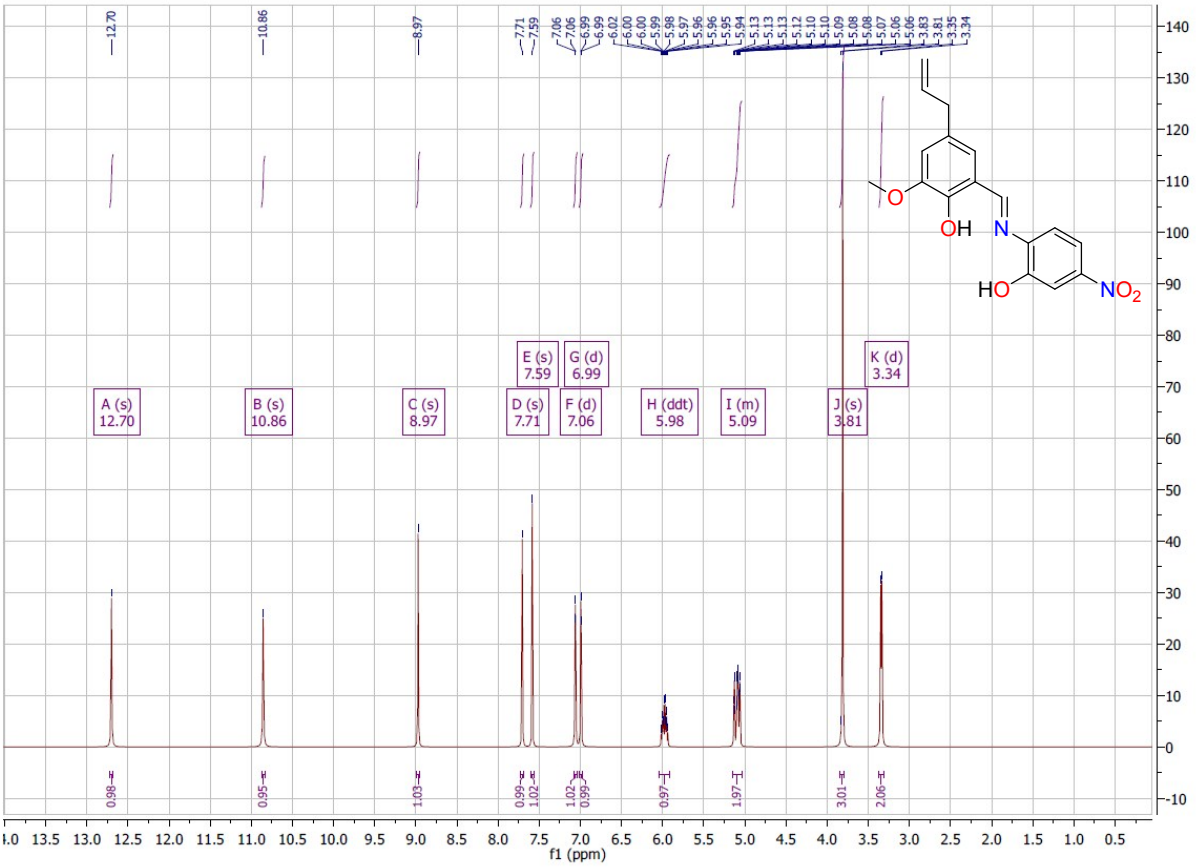
H_2L8


A (s) 12.70
B (s) 10.86
C (s) 8.97
D (s) 7.71
E (s) 7.59
F (d) 7.06
G (d) 6.99
H (ddt) 5.98
I (m) 5.09
J (s) 3.81
K (d) 3.34
f1 (ppm)

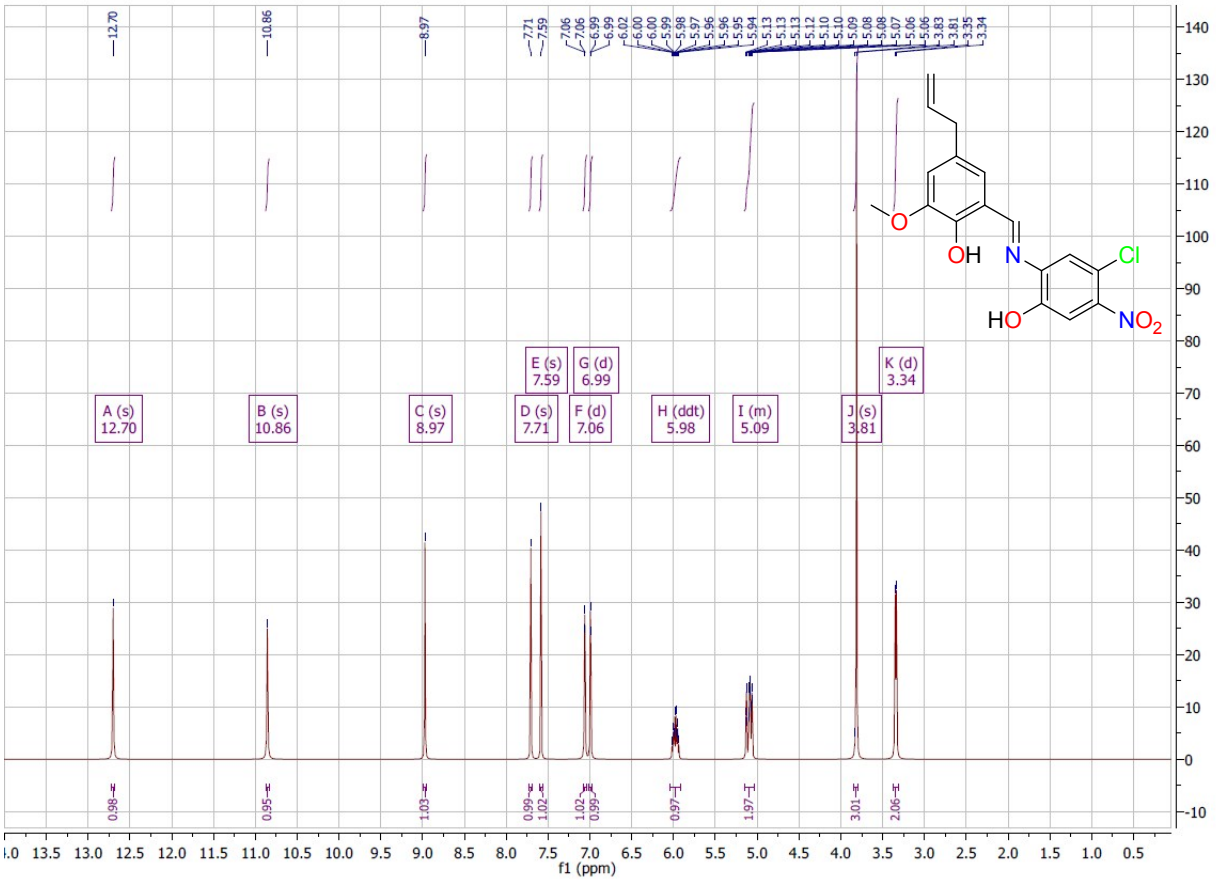
H_2L_{11}

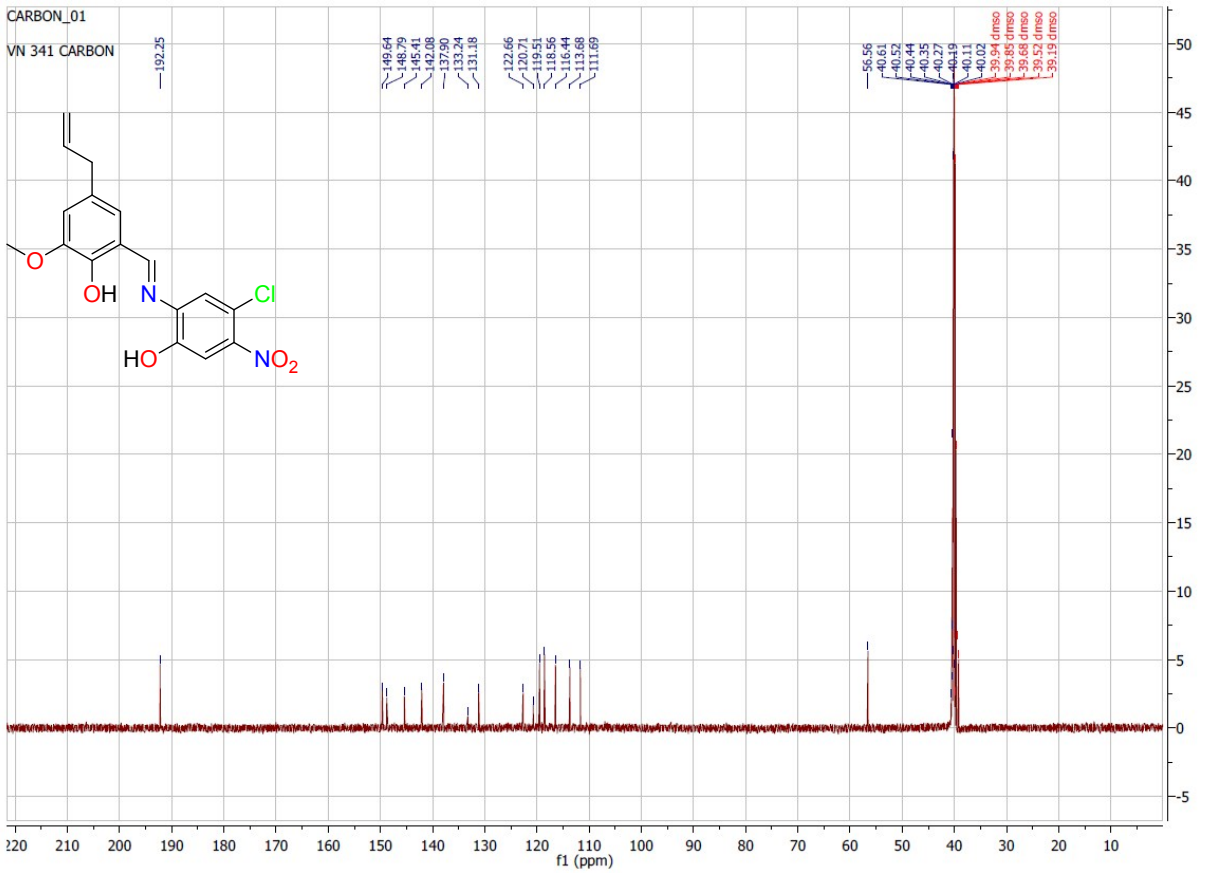

A (s) 13.80
B (s) 9.71
C (s) 8.90
D (m) 7.35
E (m) 7.10
F (m) 6.92
G (m) 5.98
H (m) 5.08
I (d) 3.79
J (m) 3.32
f1 (ppm)

H_2L12

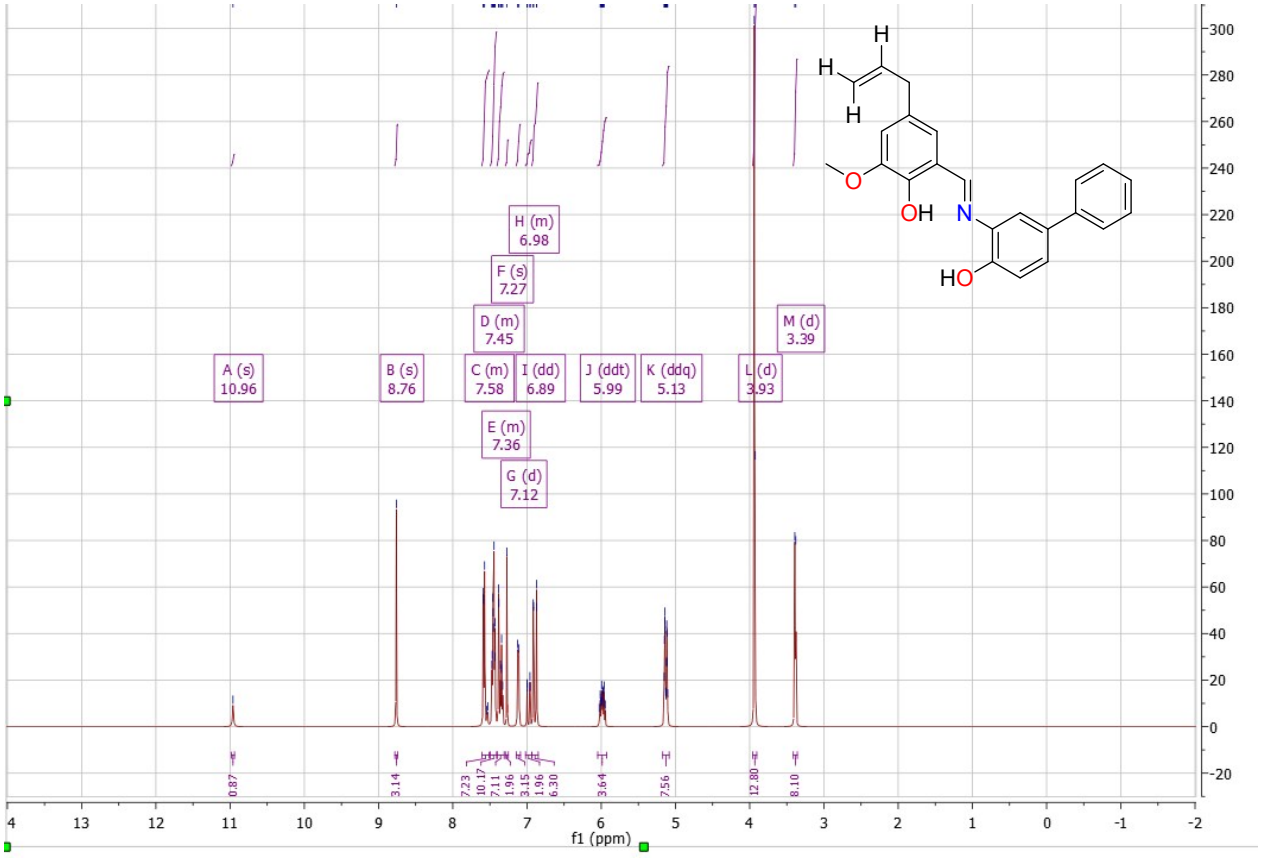

A (s) 12.70
B (s) 10.86
C (s) 8.97
D (s) 7.71
E (s) 7.59
F (d) 7.06
G (d) 6.99
H (ddt) 5.98
I (m) 5.09
J (s) 3.81
K (d) 3.34
O
OH
N
HO
NO2
f1 (ppm)

H_2L13

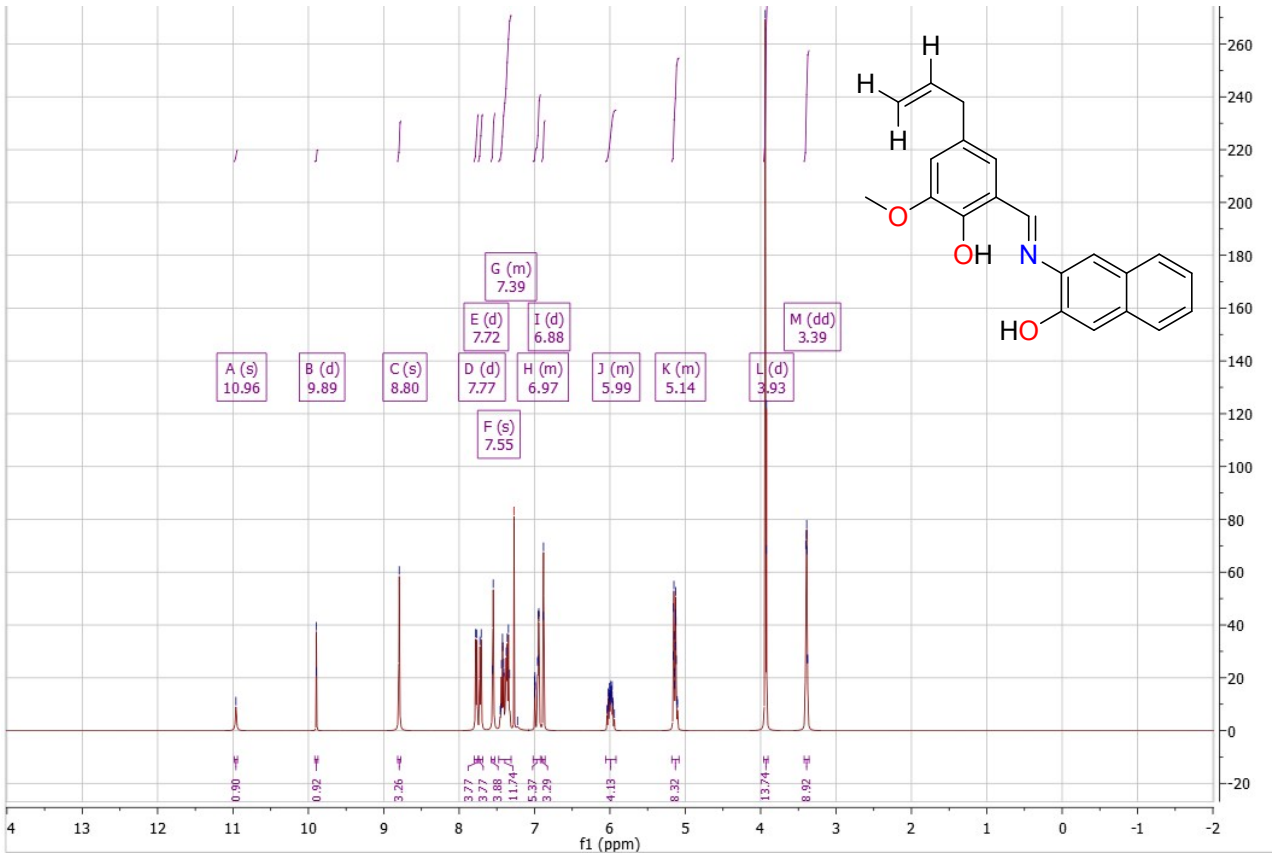

A (s) 12.70
B (s) 10.86
C (s) 8.97
D (s) 7.71
E (s) 7.59
F (d) 7.06
G (d) 6.99
H (ddt) 5.98
I (m) 5.09
J (s) 3.81
K (d) 3.34
f1 (ppm)


CARBON_01
VN 341 CARBON
f1 (ppm)

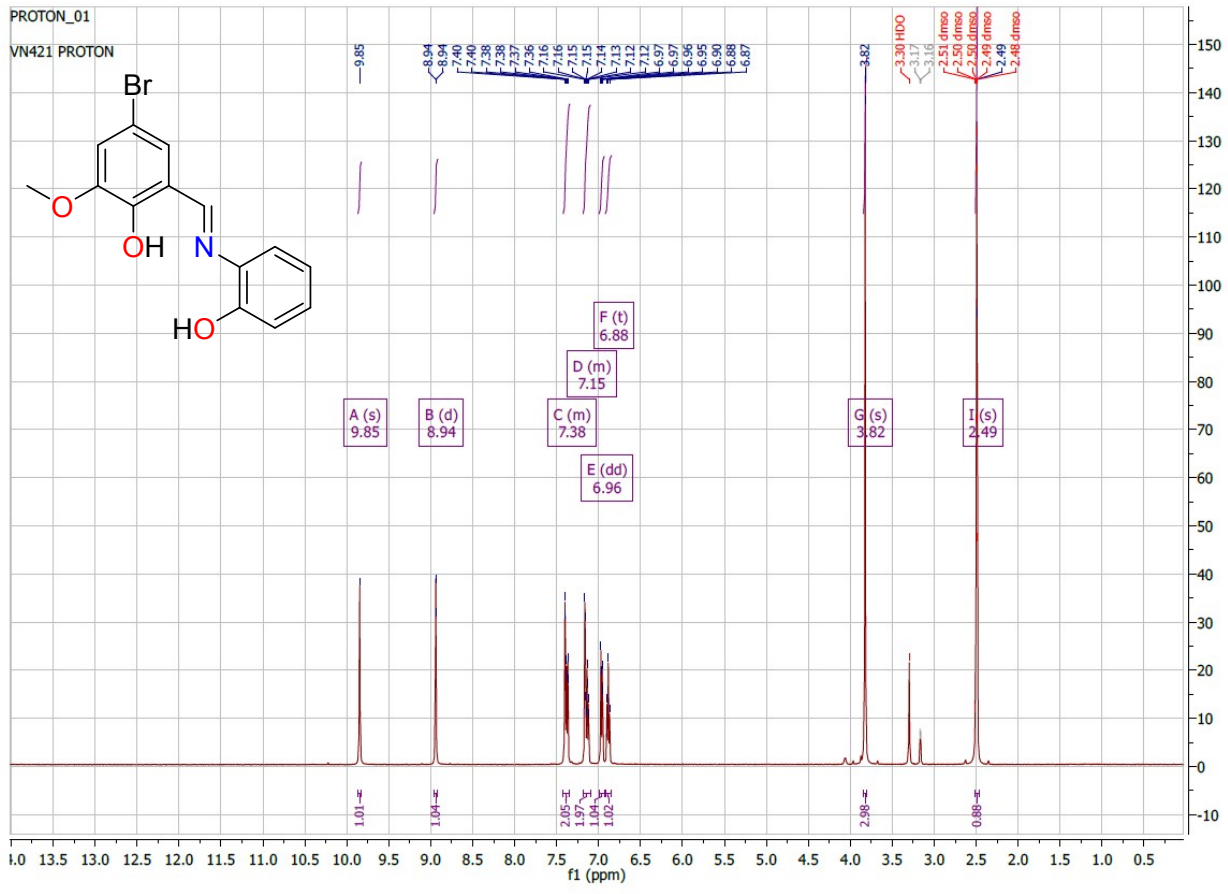
H_2L14


H (m)
6.98
F (s)
7.27
D (m)
7.45
M (d)
3.39
A (s)
10.96
B (s)
8.76
C (m)
7.58
I (dd)
6.89
J (ddt)
5.99
K (ddq)
5.13
L (d)
3.93
E (m)
7.36
G (d)
7.12
f1 (ppm)

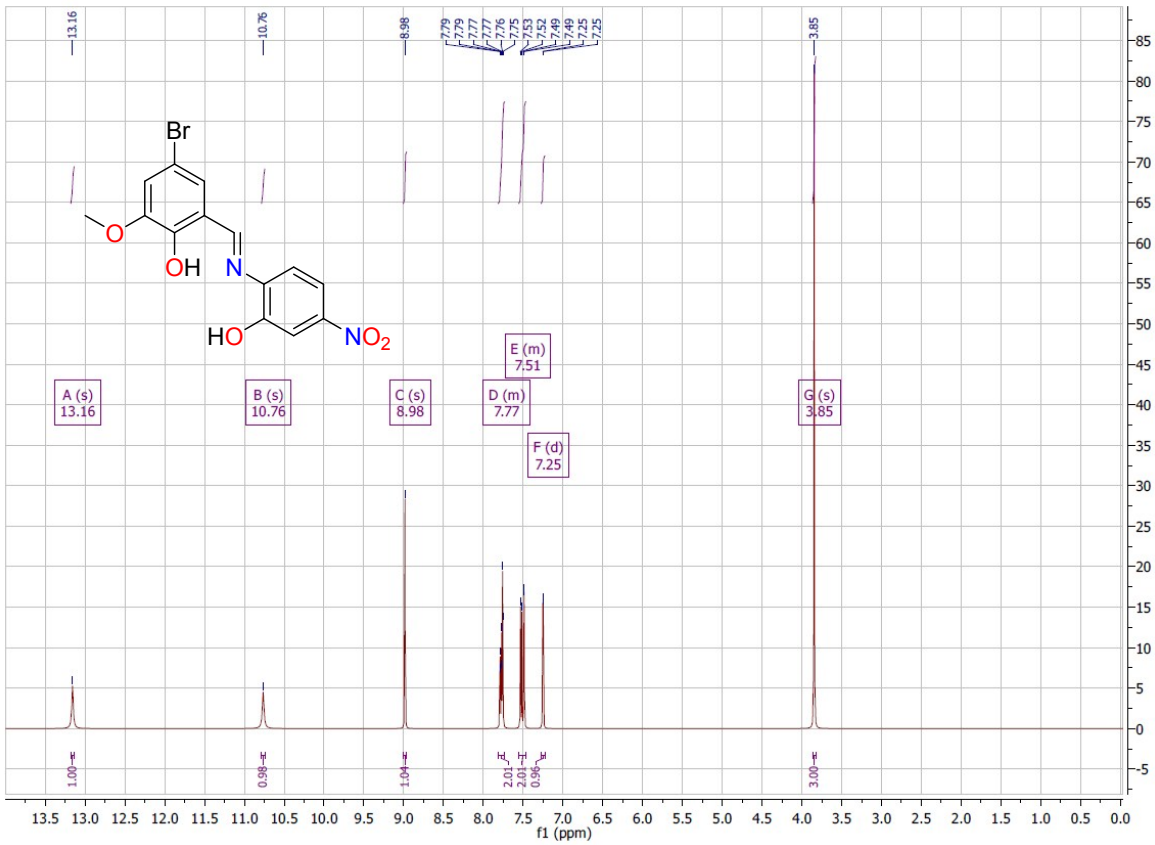
H_2L15

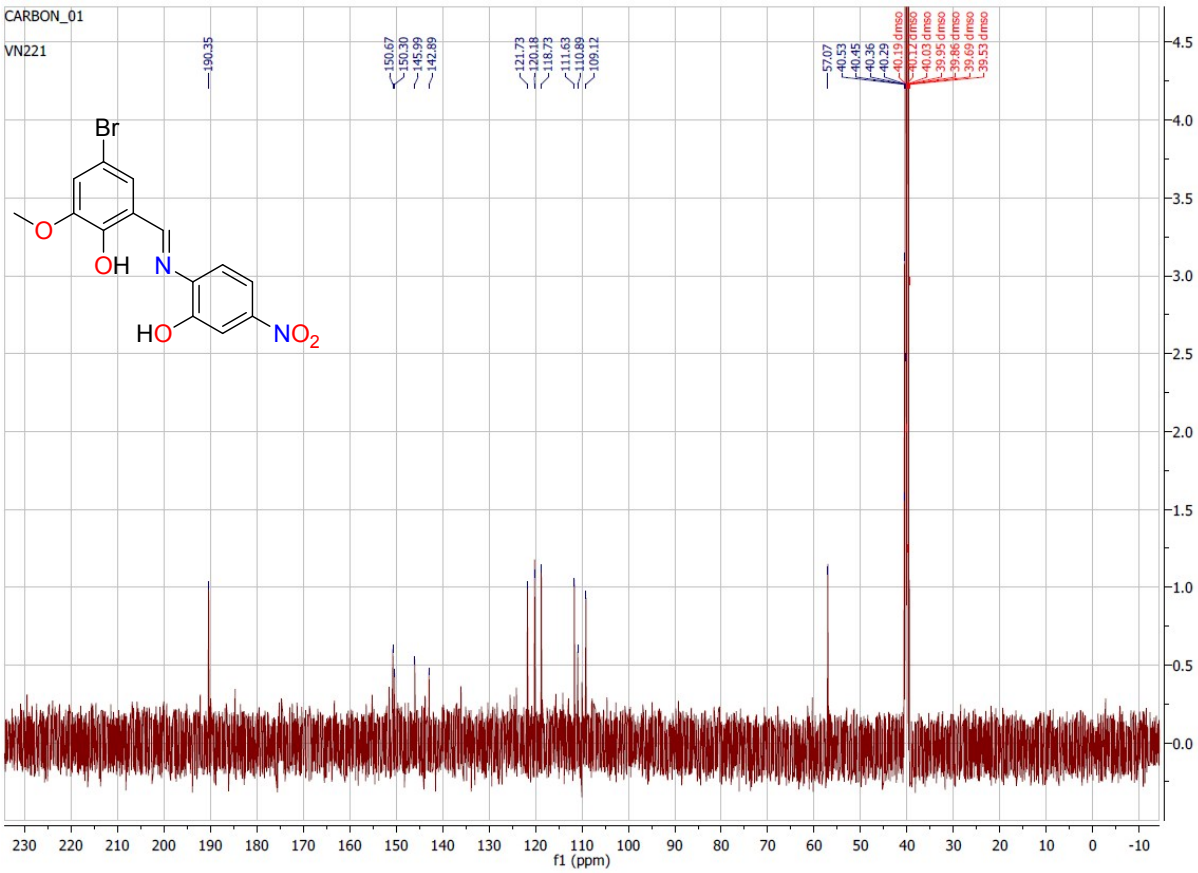

A (s) 10.96
B (d) 9.89
C (s) 8.80
D (d) 7.77
E (d) 7.72
F (s) 7.55
G (m) 7.39
H (m) 6.97
I (d) 6.88
J (m) 5.99
K (m) 5.14
L (d) 3.93
M (dd) 3.39
f1 (ppm)

H_2L16

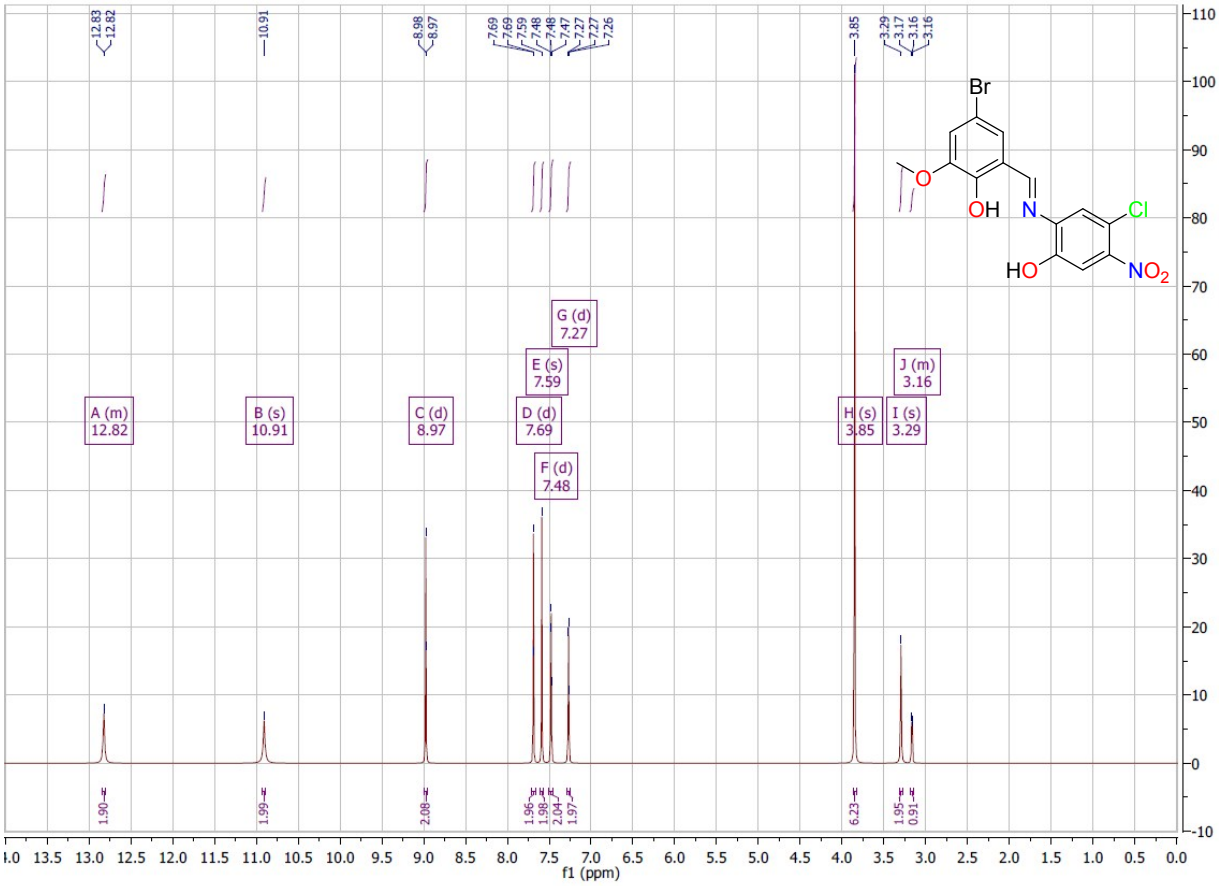

PROTON_01
VN421 PROTON
A (s) 9.85
B (d) 8.94
C (m) 7.38
D (m) 7.15
E (dd) 6.96
F (t) 6.88
G (s) 3.82
I (s) 2.49
f1 (ppm)

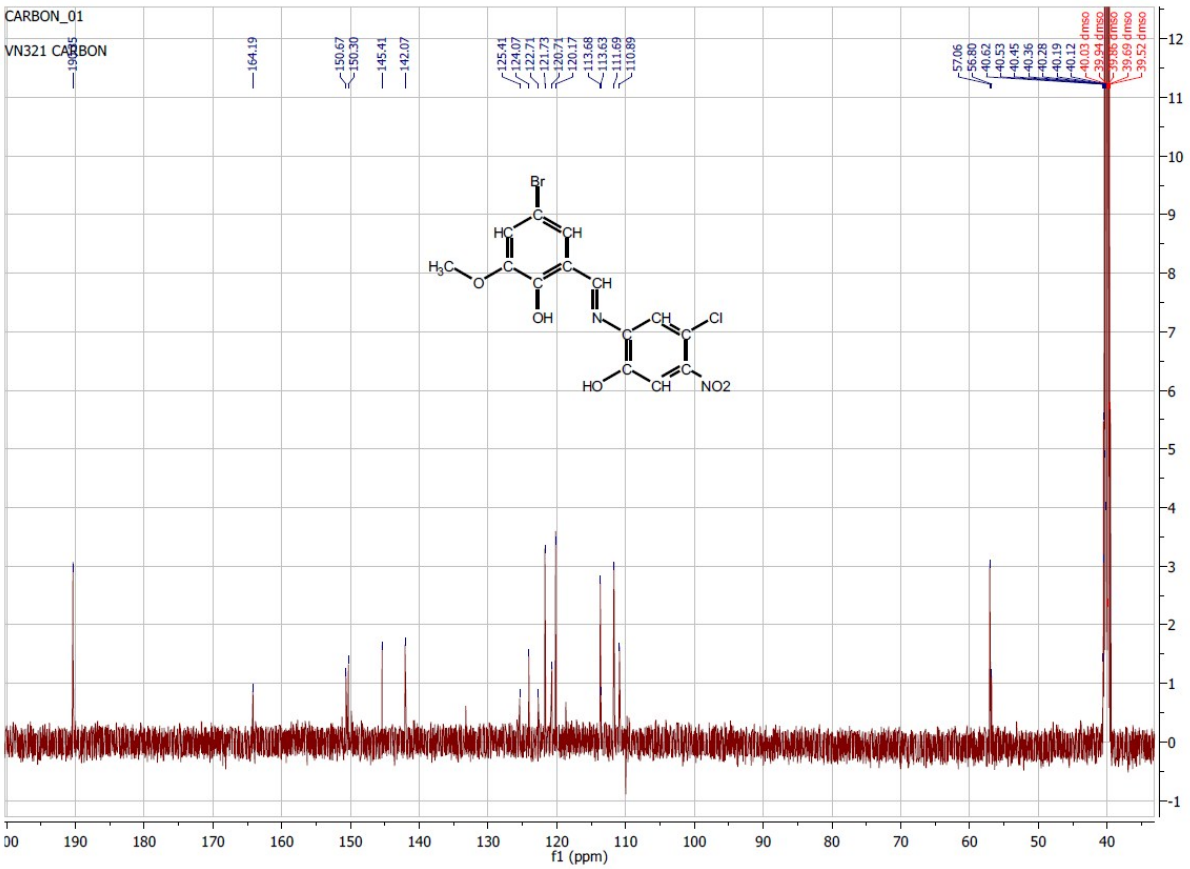
H_2L17


13.16
10.76
8.98
7.79
7.79
7.77
7.77
7.76
7.75
7.53
7.52
7.49
7.49
7.25
7.25
3.85
Br
O
OH
N
HO
NO2
A (s)
13.16
B (s)
10.76
C (s)
8.98
D (m)
7.77
E (m)
7.51
F (d)
7.25
G (s)
3.85
1.00
0.98
1.04
2.01
2.01
0.96
3.00
f1 (ppm)

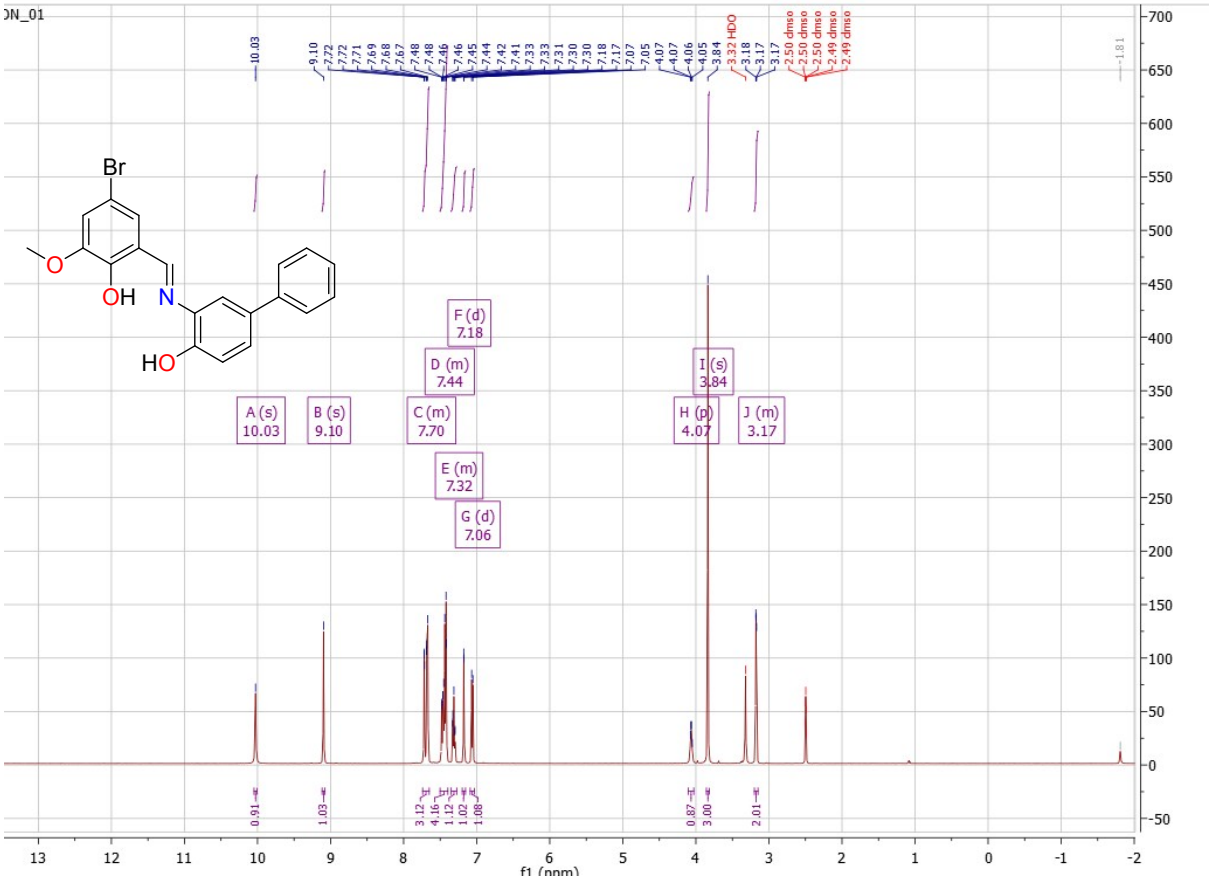

CARBON_01
VN221
190.35
150.67
150.30
145.99
142.89
121.73
120.18
118.73
111.63
110.89
109.12
57.07
40.53
40.45
40.36
40.29
40.19 dmso
40.12 dmso
40.03 dmso
39.95 dmso
39.86 dmso
39.69 dmso
39.53 dmso
Br
O
OH
N
HO
NO2
f1 (ppm)

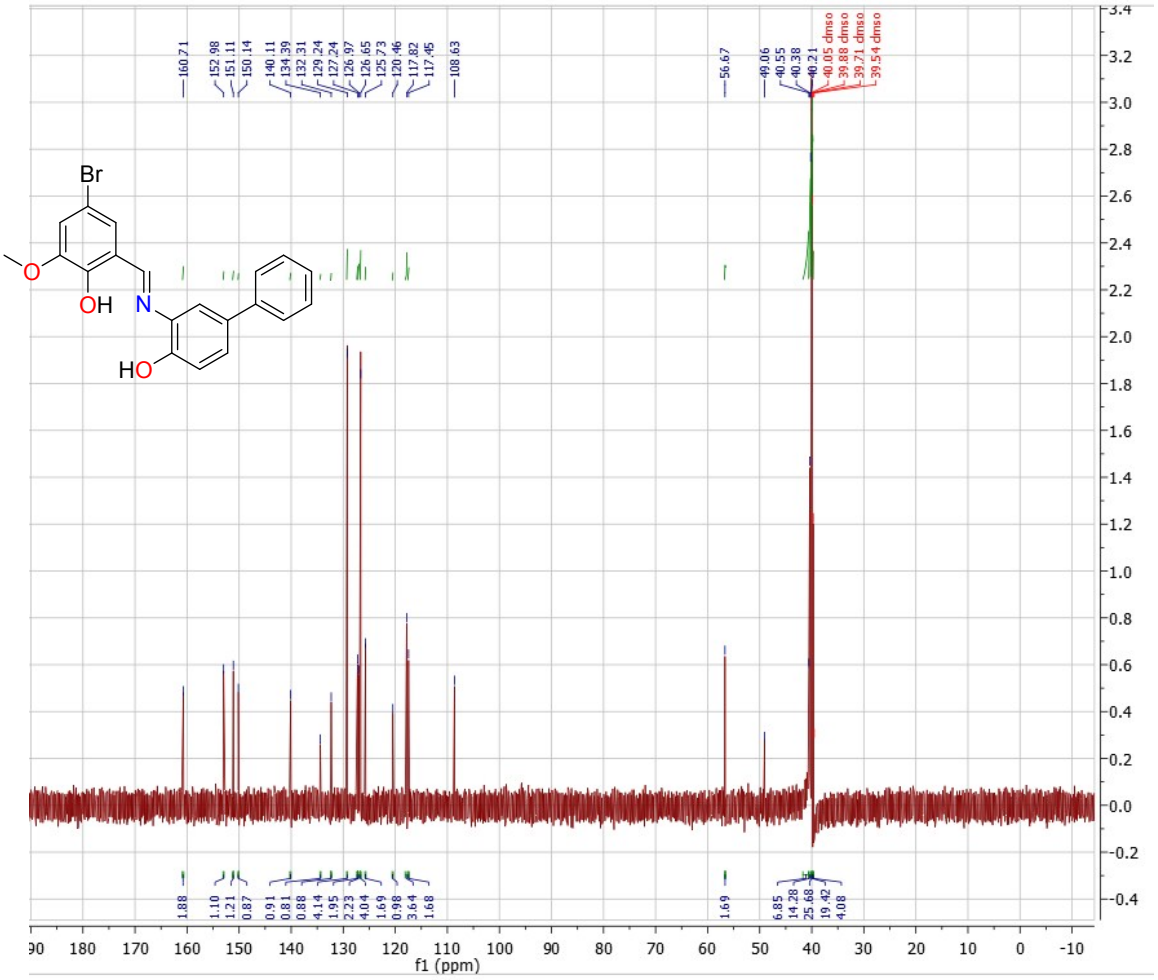
H_2L18


A (m) 12.82
B (s) 10.91
C (d) 8.97
D (d) 7.69
E (s) 7.59
F (d) 7.48
G (d) 7.27
H (s) 3.85
I (s) 3.29
J (m) 3.16
Br
O
OH
N
Cl
HO
NO2
f1 (ppm)

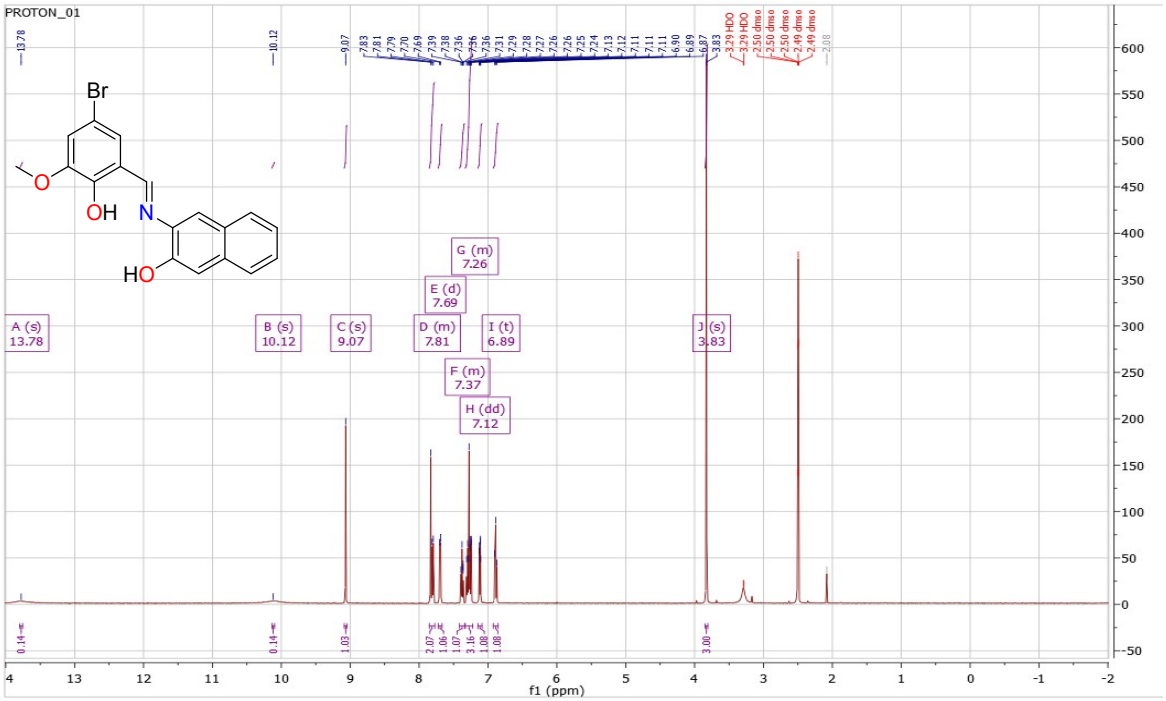

CARBON_01
VN321 CARBON
164.19
150.67
150.30
145.41
142.07
125.41
124.07
122.71
121.71
120.71
120.17
113.68
113.63
111.69
110.89
57.06
56.80
40.62
40.53
40.45
40.36
40.28
40.19
40.12
40.03 dmso
39.94 dmso
39.86 dmso
39.69 dmso
39.52 dmso
Br
HC
CH
H3C
O
C
OH
CH
N
CH
Cl
HO
CH
NO2
f1 (ppm)

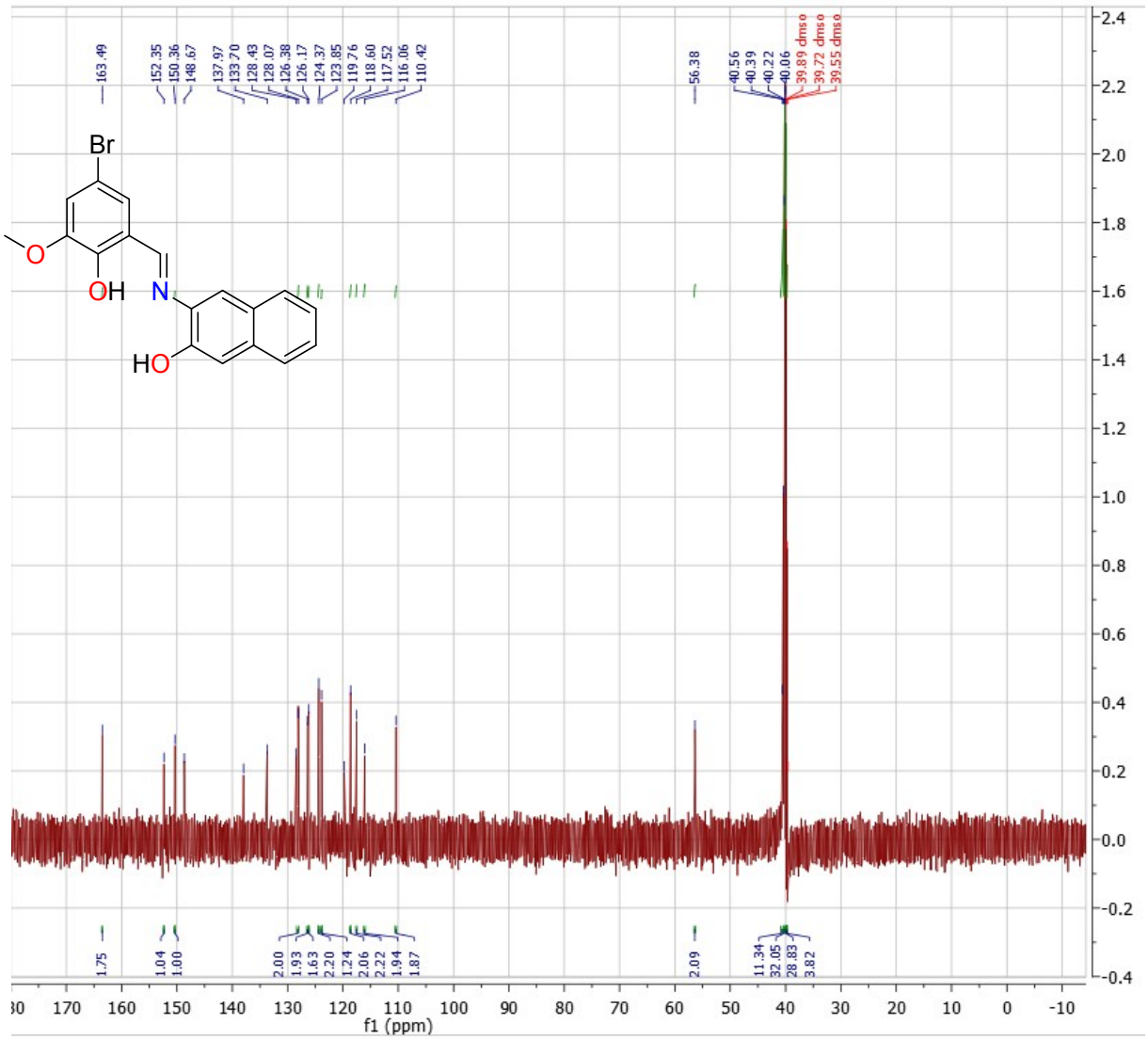
H_2L19


ON_01
10.03
9.10
7.72
7.71
7.69
7.68
7.67
7.48
7.46
7.45
7.44
7.42
7.41
7.33
7.31
7.30
7.18
7.17
7.07
7.05
4.07
4.06
4.05
3.84
3.32 HDO
3.18
3.17
2.50 dmso
2.49 dmso
Br
O
OH
N
HO
A (s) 10.03
B (s) 9.10
C (m) 7.70
D (m) 7.44
E (m) 7.32
F (d) 7.18
G (d) 7.06
H (p) 4.07
I (s) 3.84
J (m) 3.17
f1 (ppm)


Br
O
OH
N
HO
f1 (ppm)

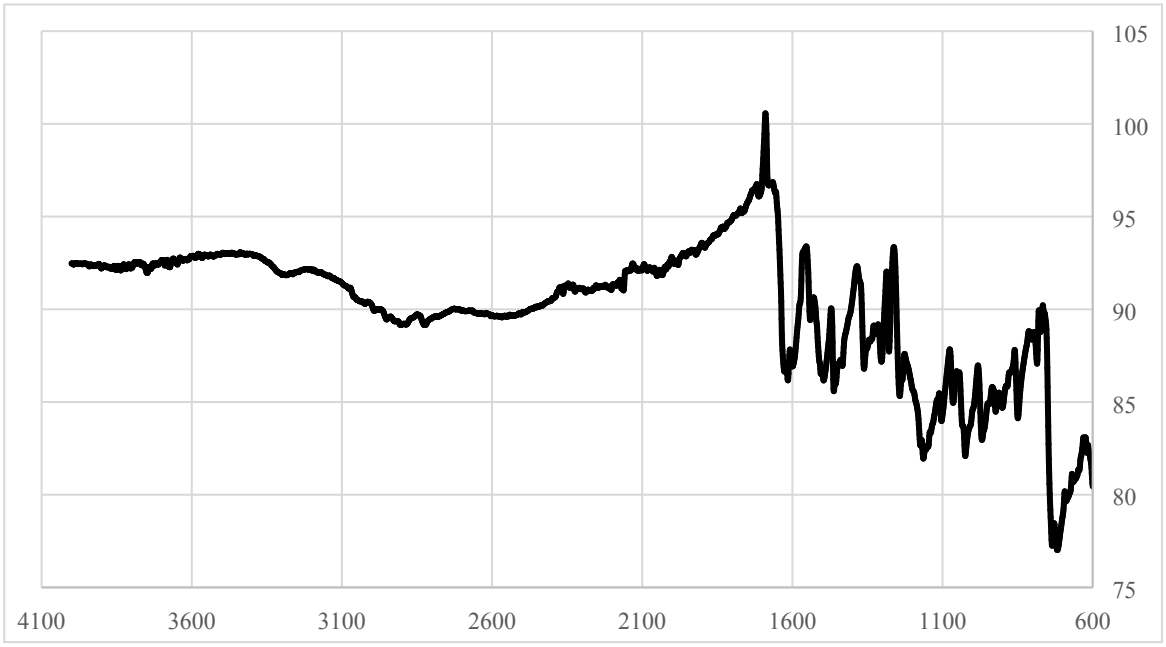
H_2L20


PROTON_01
Br
O
OH
N
HO
A (s) 13.78
B (s) 10.12
C (s) 9.07
D (m) 7.81
E (d) 7.69
F (m) 7.37
G (m) 7.26
H (dd) 7.12
I (t) 6.89
J (s) 3.83
f1 (ppm)

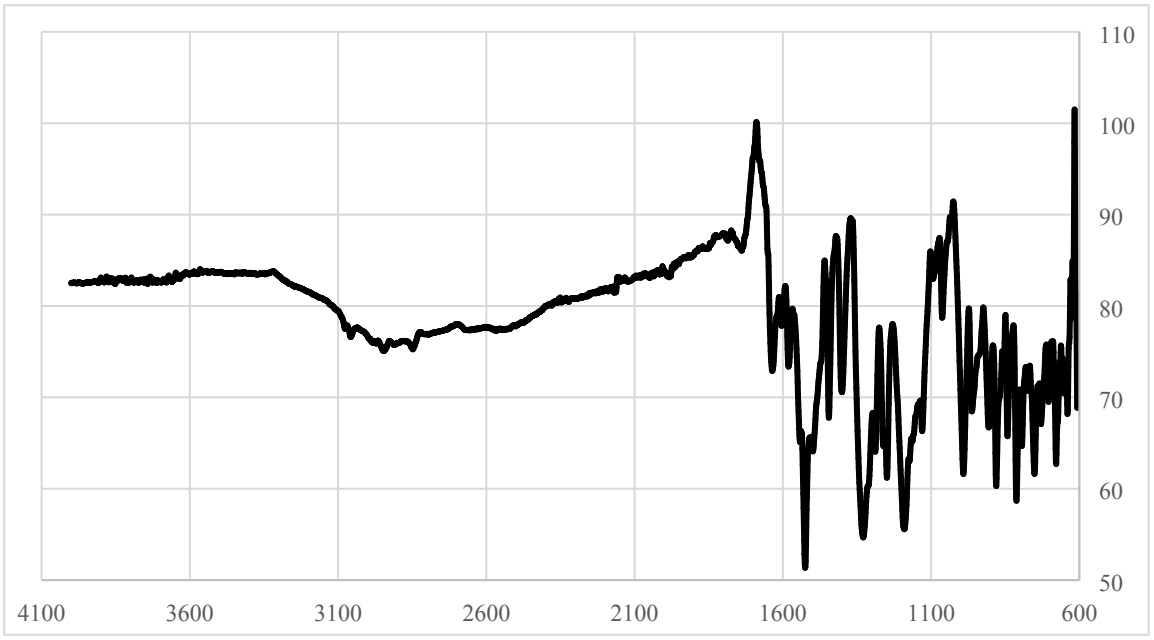

163.49
152.35
150.36
148.67
137.97
133.70
128.43
128.07
126.38
126.17
124.37
123.85
119.76
118.60
117.52
116.06
110.42
56.38
40.56
40.39
40.22
40.06
39.89 dmso
39.72 dmso
39.55 dmso
Br
O
OH
N
HO
1.75
1.04
1.00
2.00
1.93
1.63
2.20
1.24
2.06
2.22
1.94
1.87
2.09
11.34
32.05
28.83
3.82
f1 (ppm)

FT-IR of Ligands H_2L1-H_2L20

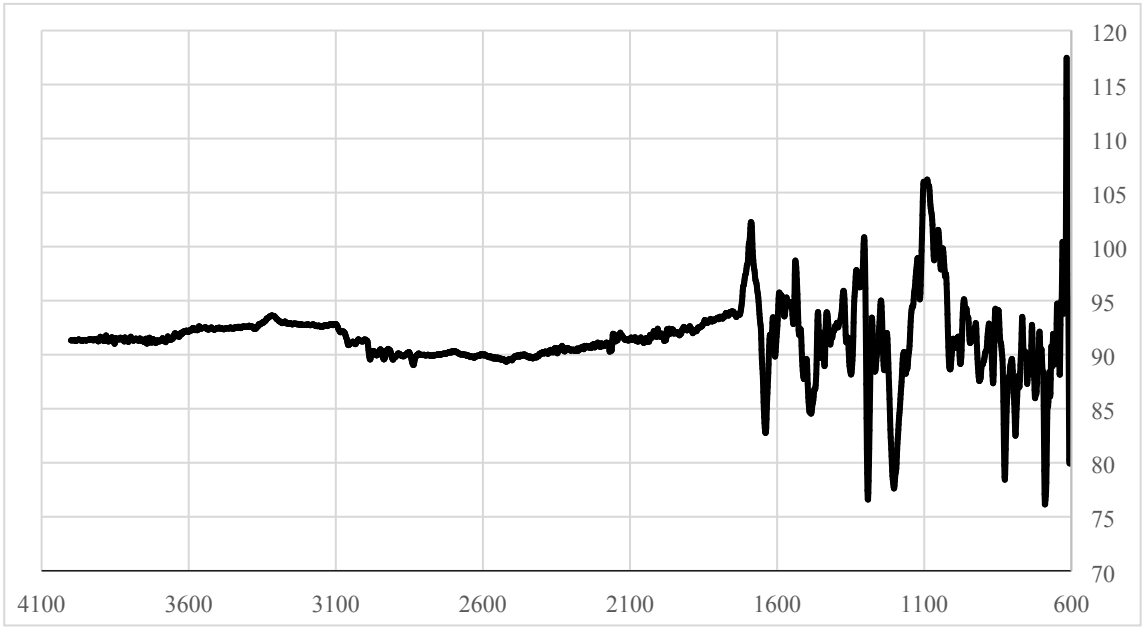
H_2L1



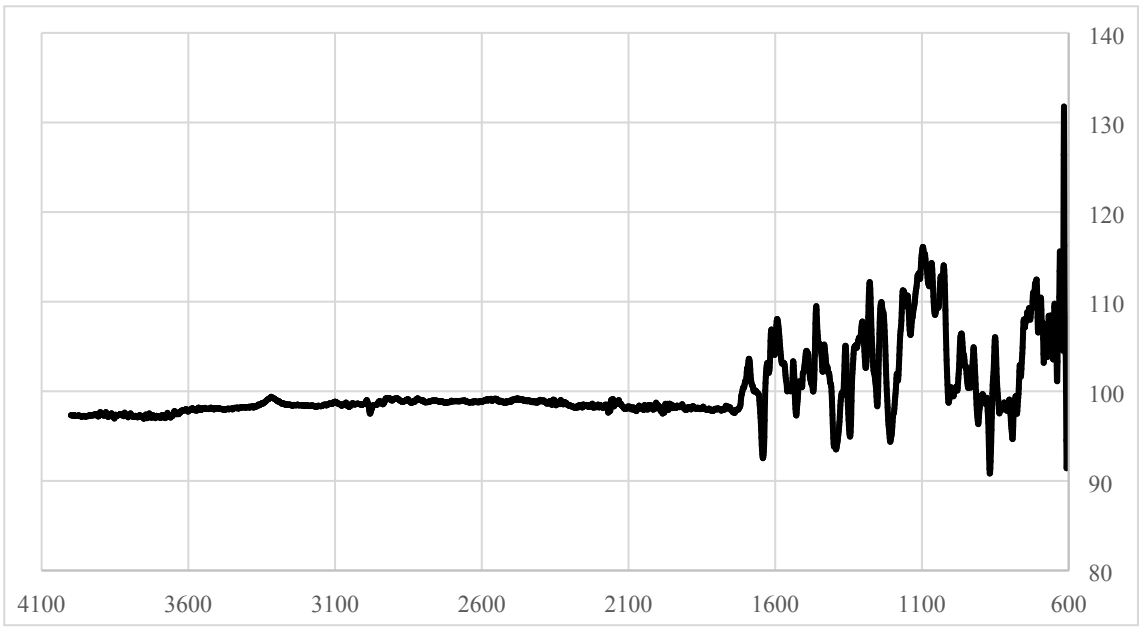
H_2L3



H_2L4


120
115
110
105
100
95
90
85
80
75
70
4100
3600
3100
2600
2100
1600
1100
600

H_2L5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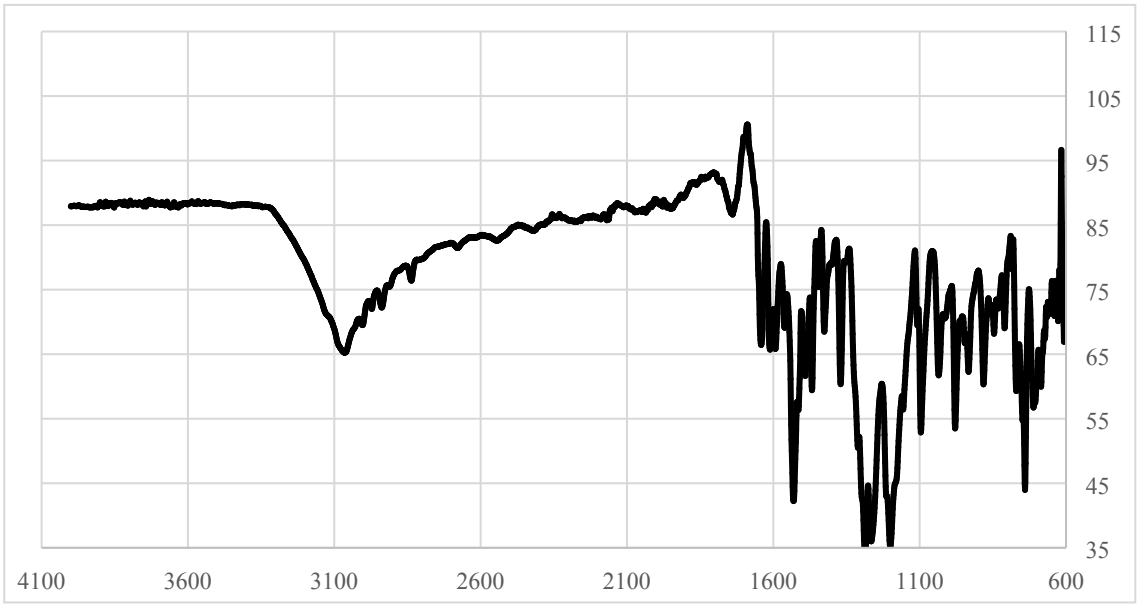

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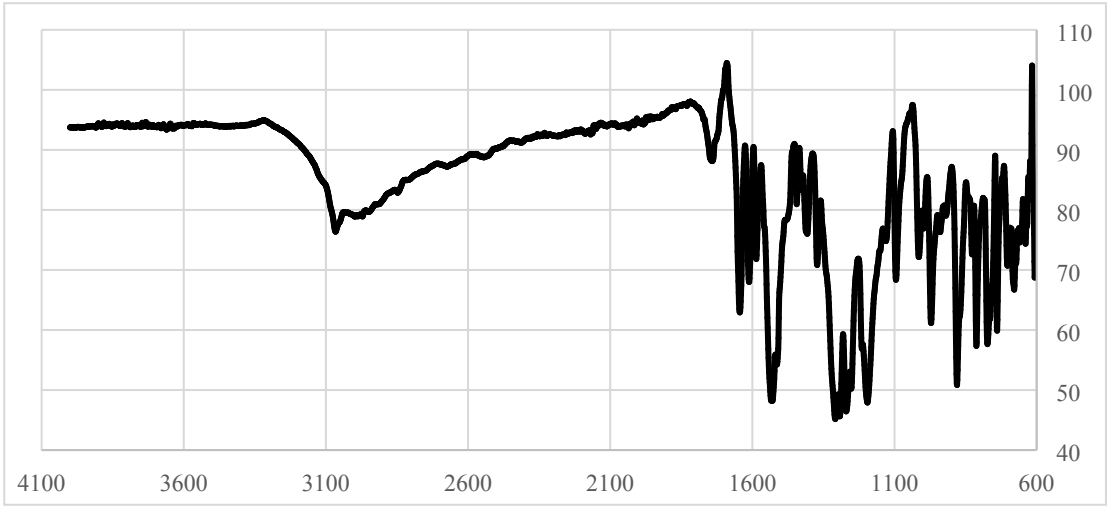

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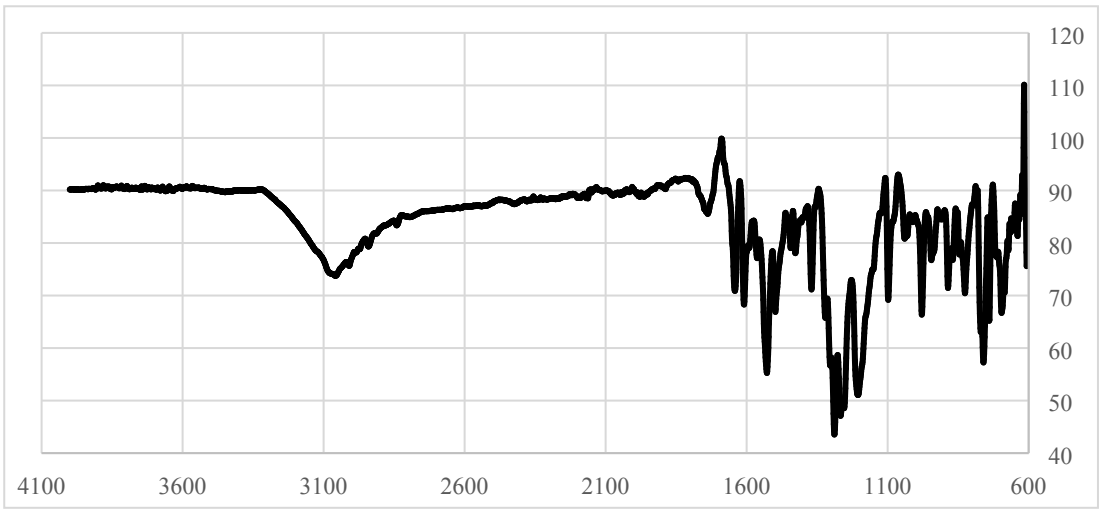
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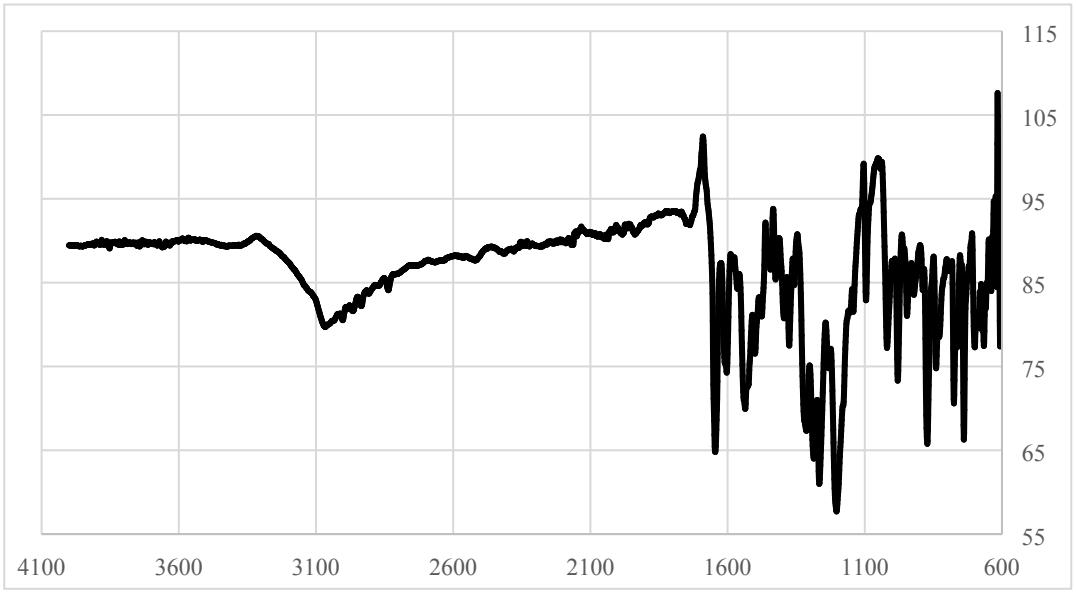
H_2L10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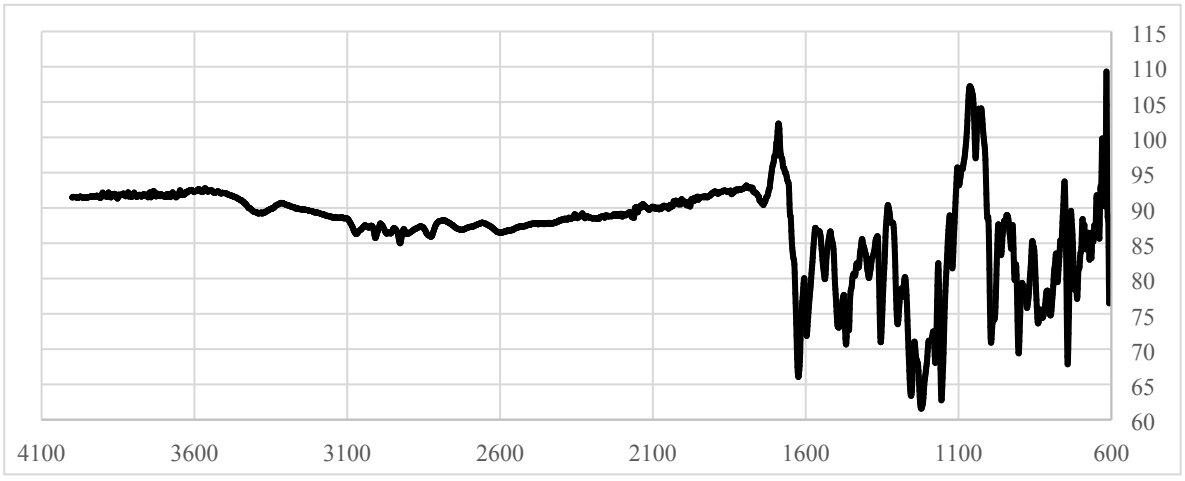
H_2L11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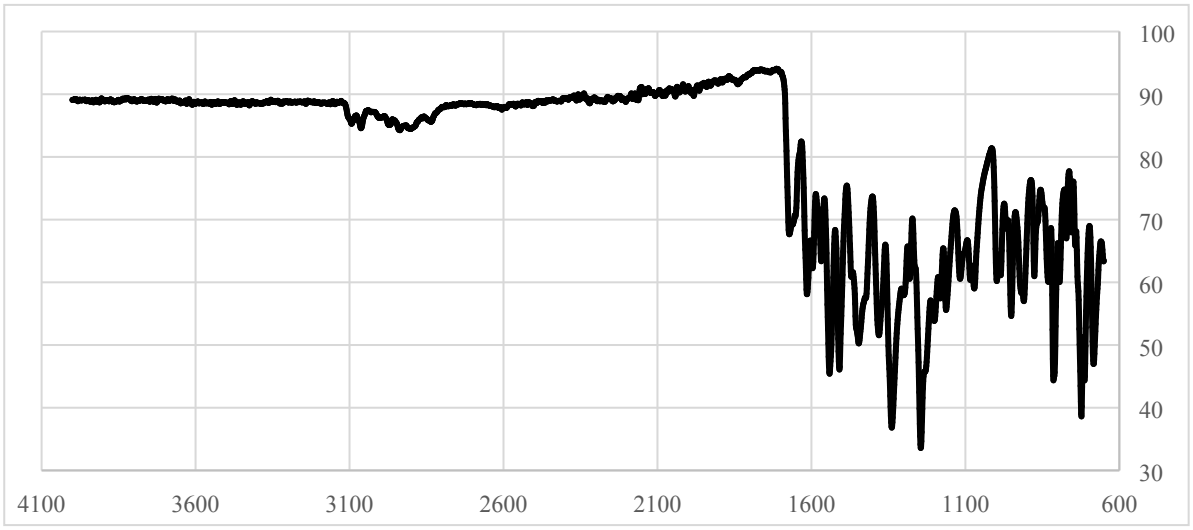
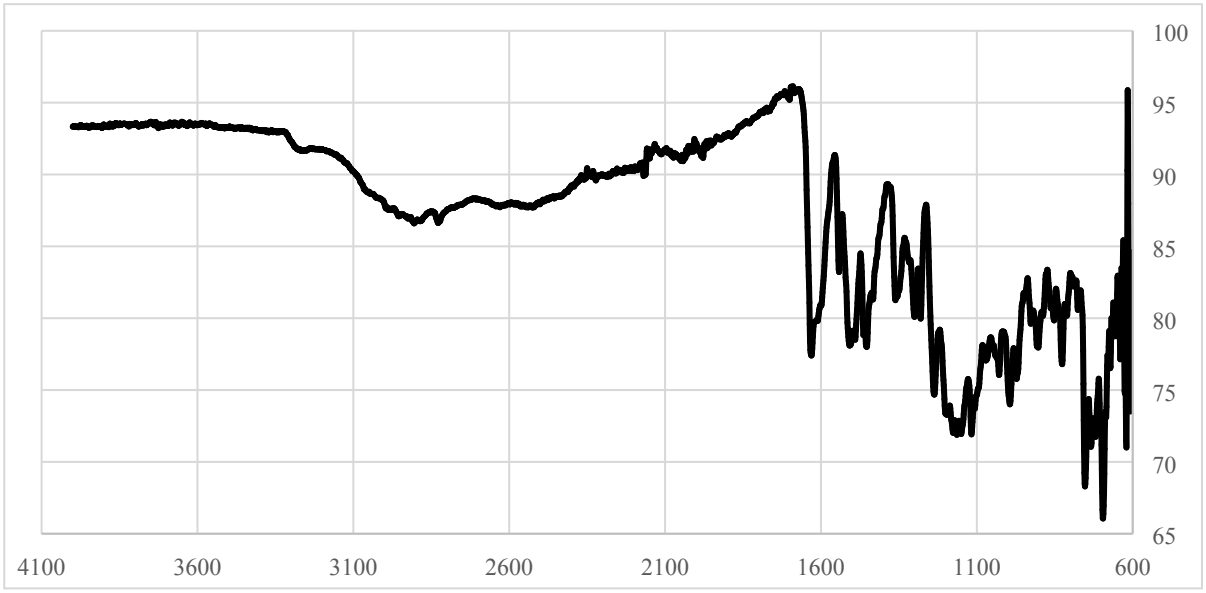
H_2L13



H_2L14



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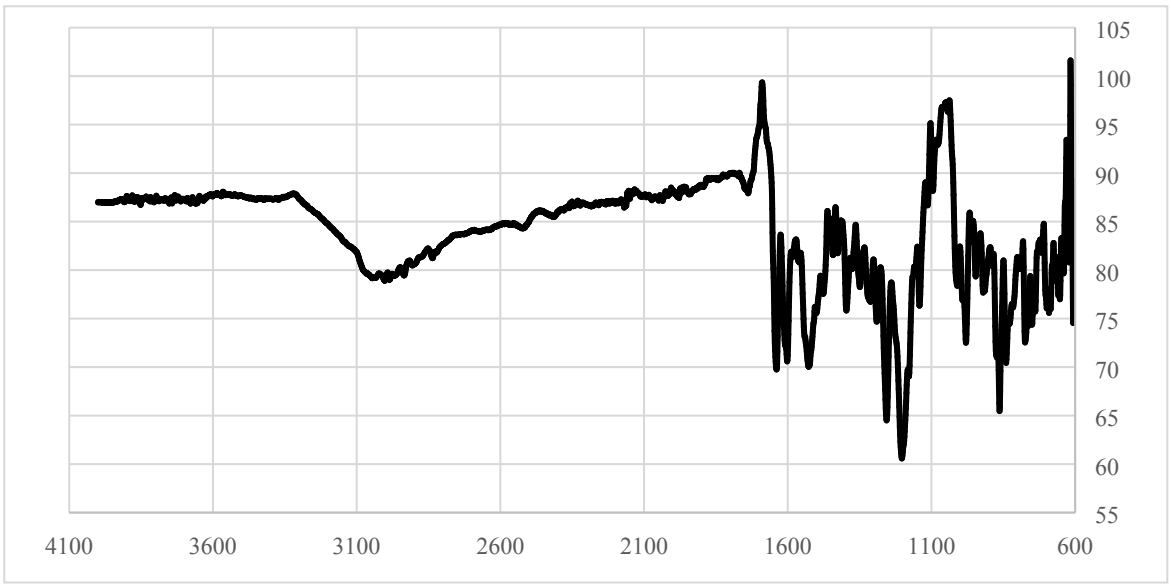
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