Electronic Supplementary Material (ESI) for Dalton Transactions.

Supporting Information

Syntheses and Exploration of the Catalytic Activities of Organotin(IV) Compounds

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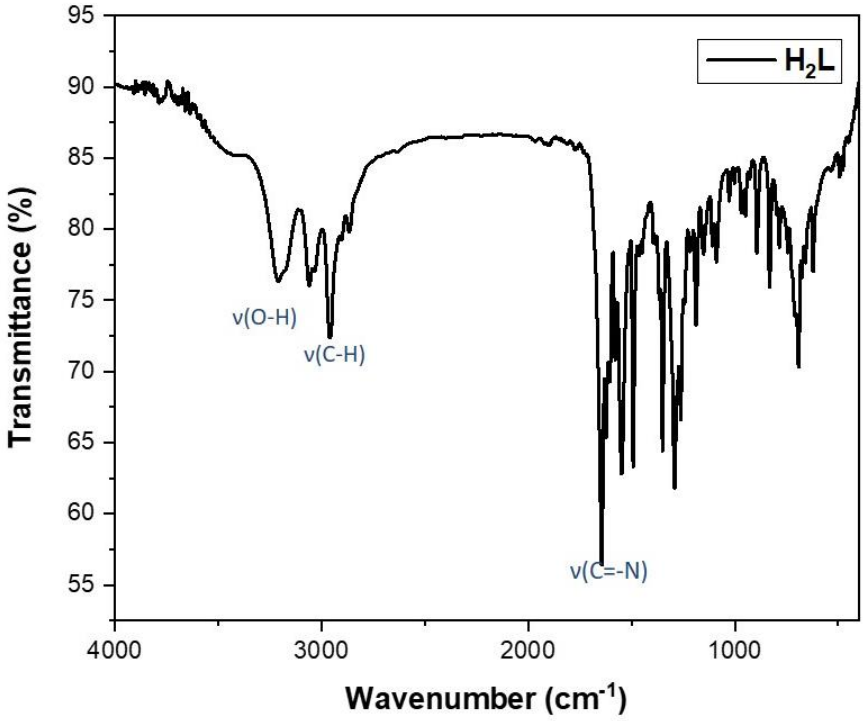


Figure S1: FT-IR spectrum of **H_2L**.

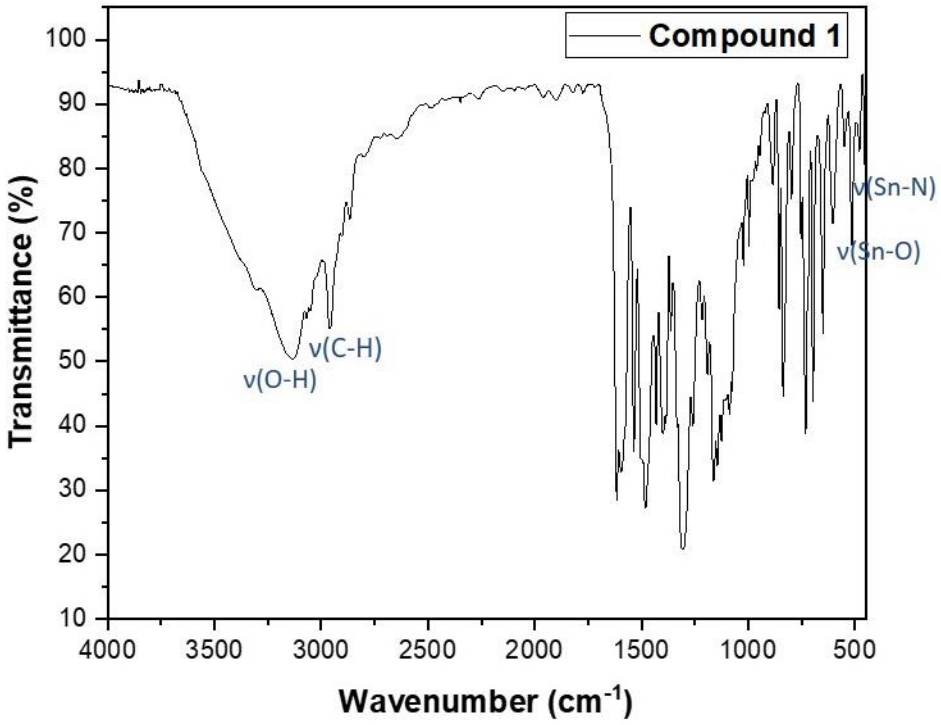


Figure S2: FT-IR spectrum of compound **1**.

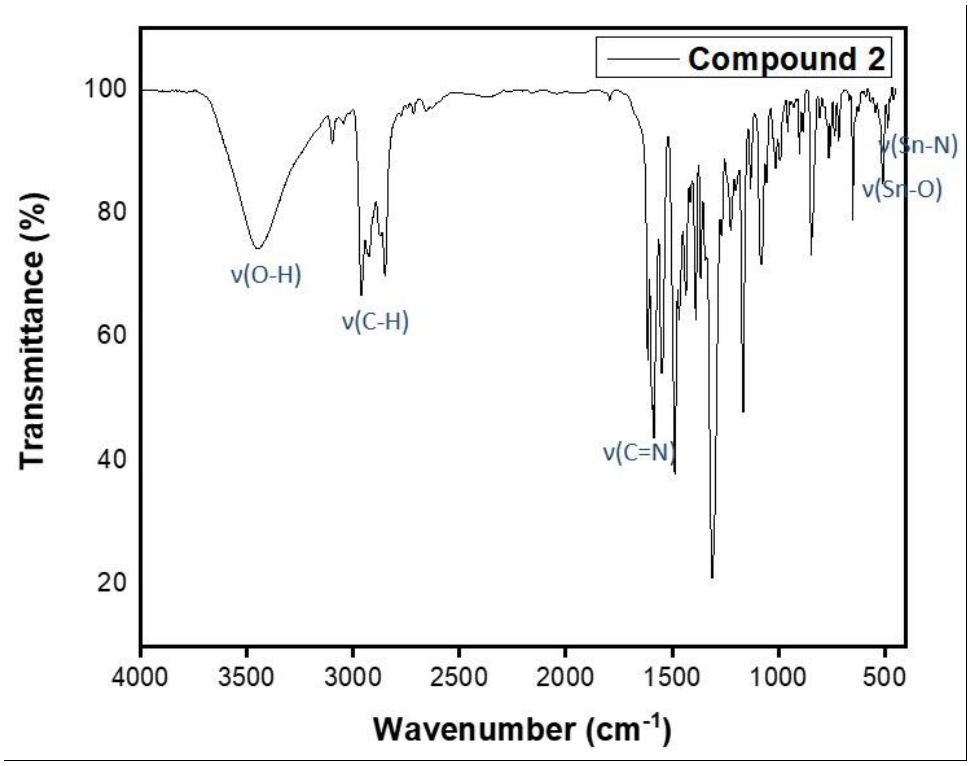


Figure S3: FT-IR spectrum of compound **2**.

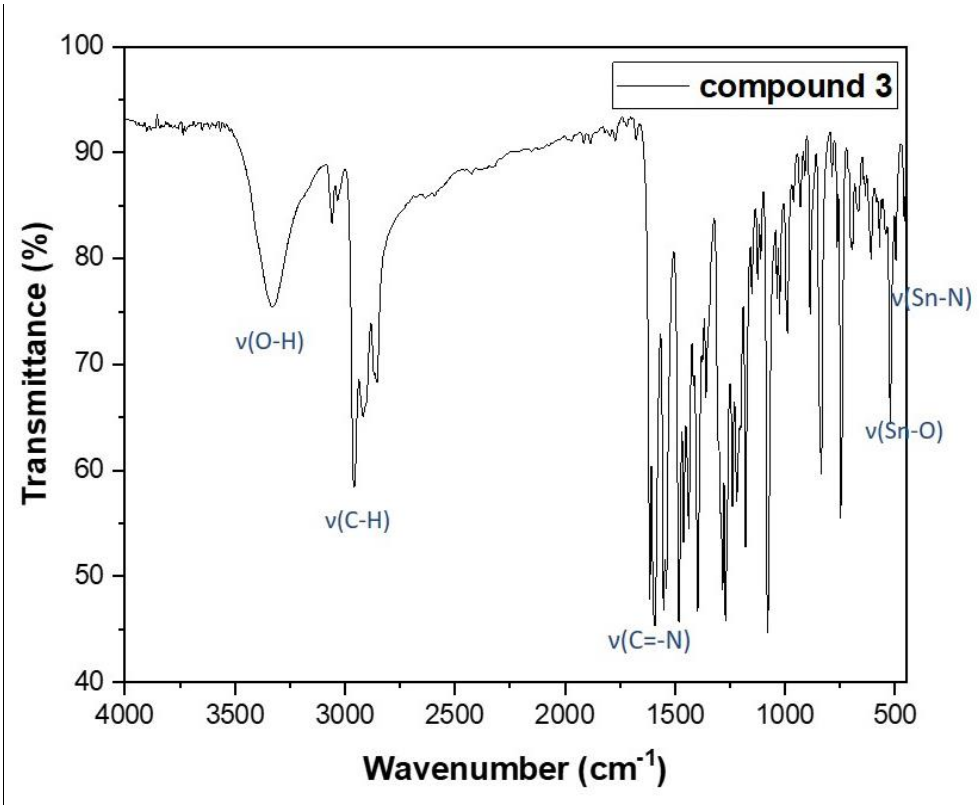


Figure S4: FT-IR spectrum of compound **3**.

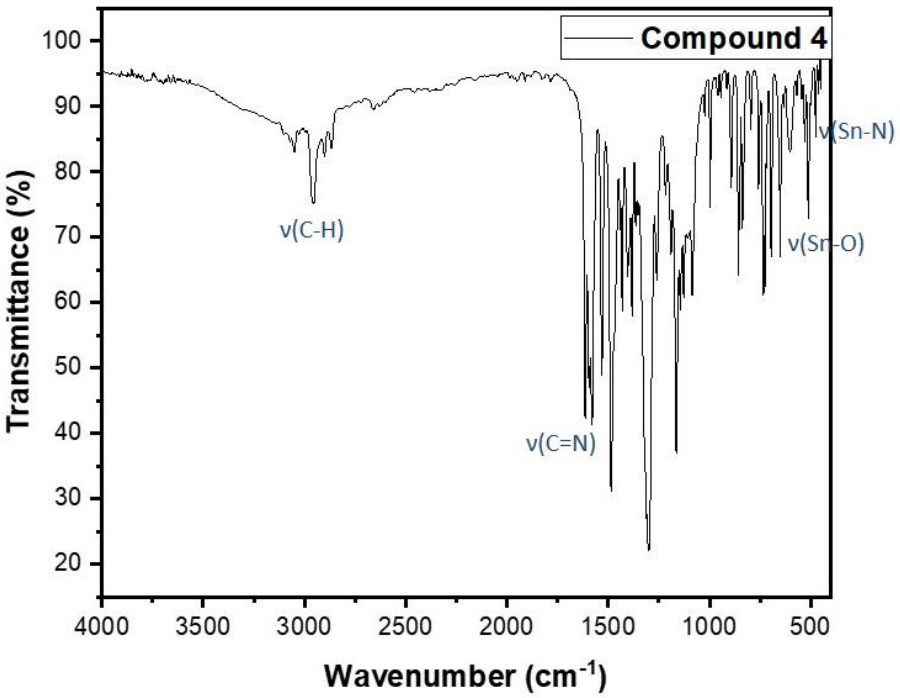


Figure S5: FT-IR spectrum of compound **4**.

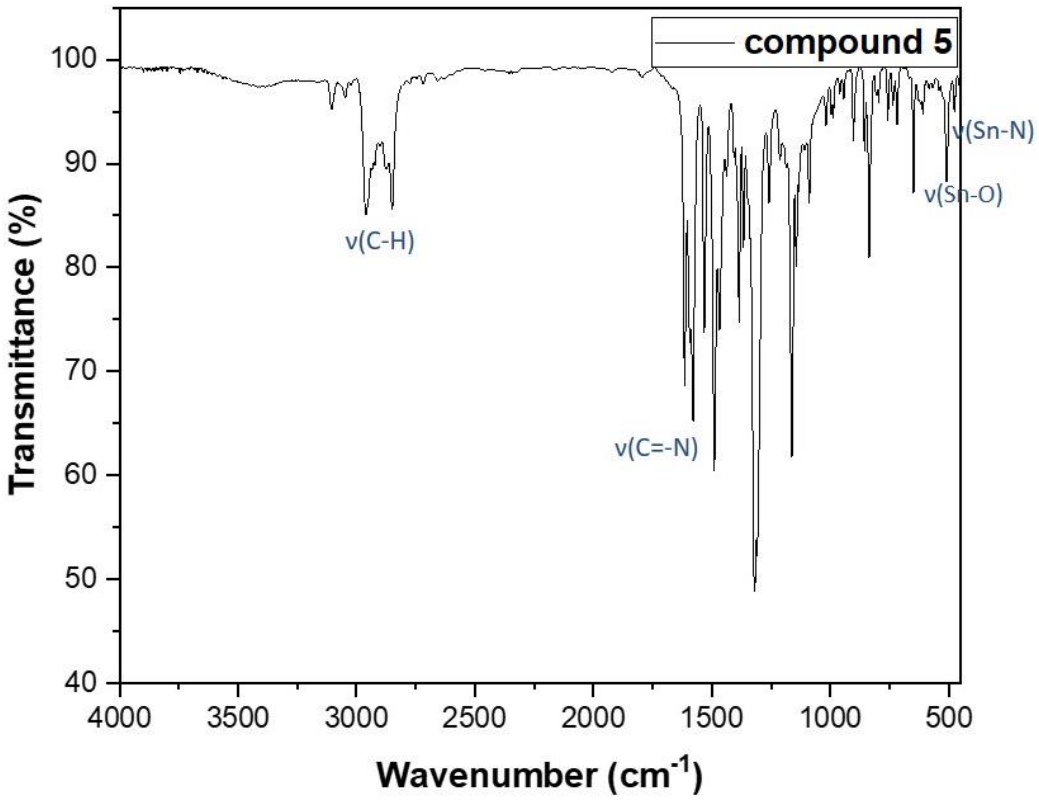


Figure S6: FT-IR spectrum of compound **5**.

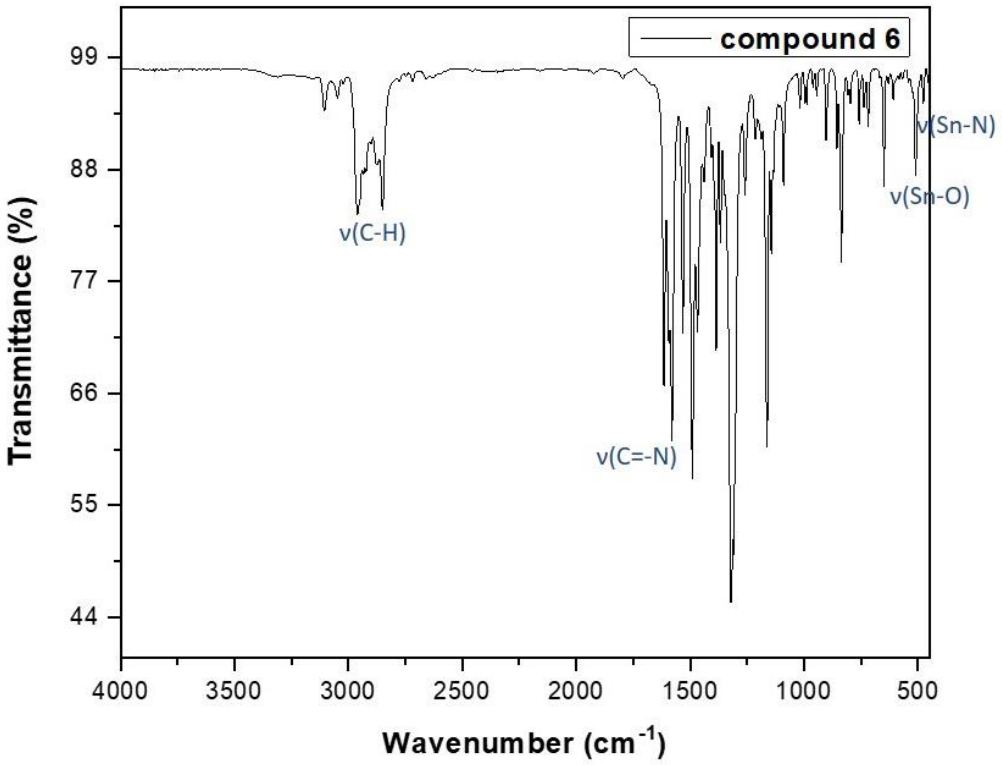


Figure S7: FT-IR spectrum of compound **6**.

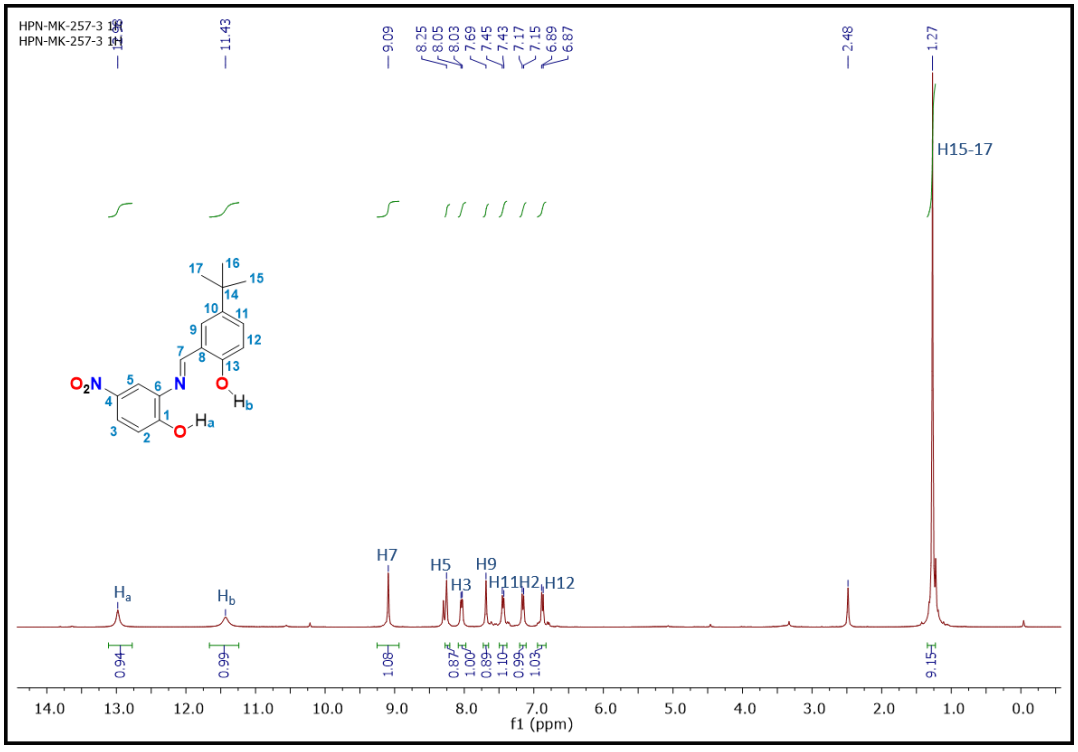


Figure S8: ^{1}H NMR (400 MHz, DMSO-d_6) spectrum of **H_2L**.

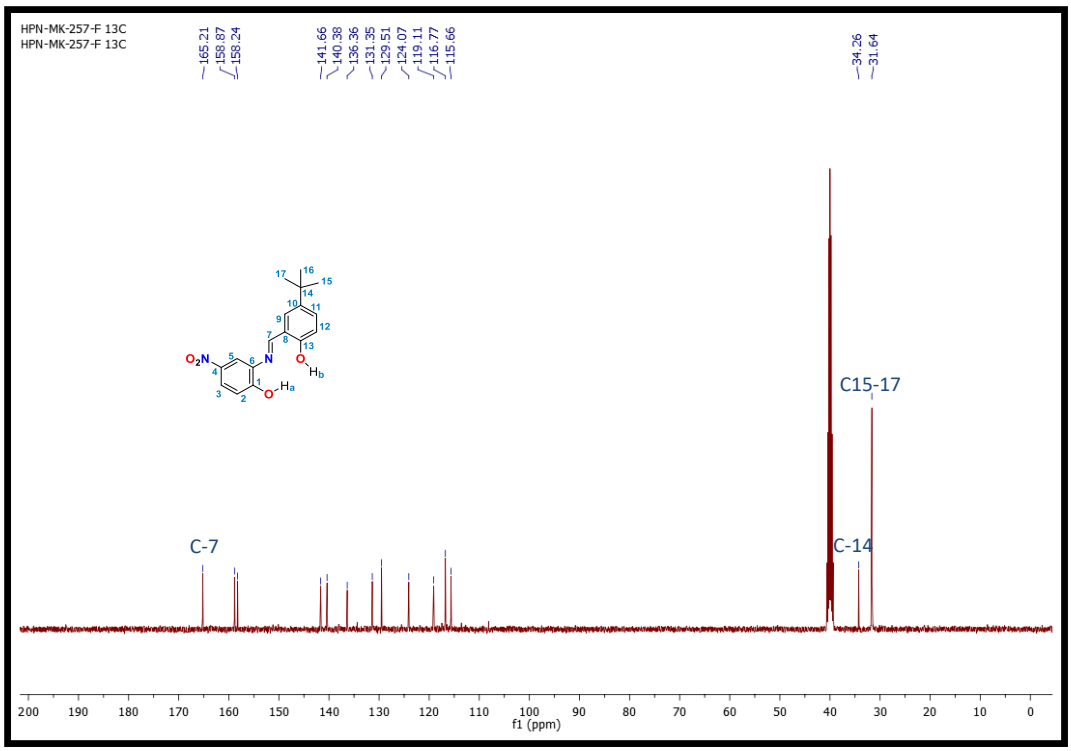


Figure S9: $^{13}C(^{1}H)$ NMR (100 MHz, DMSO-d_6) spectrum of **H_2L**.

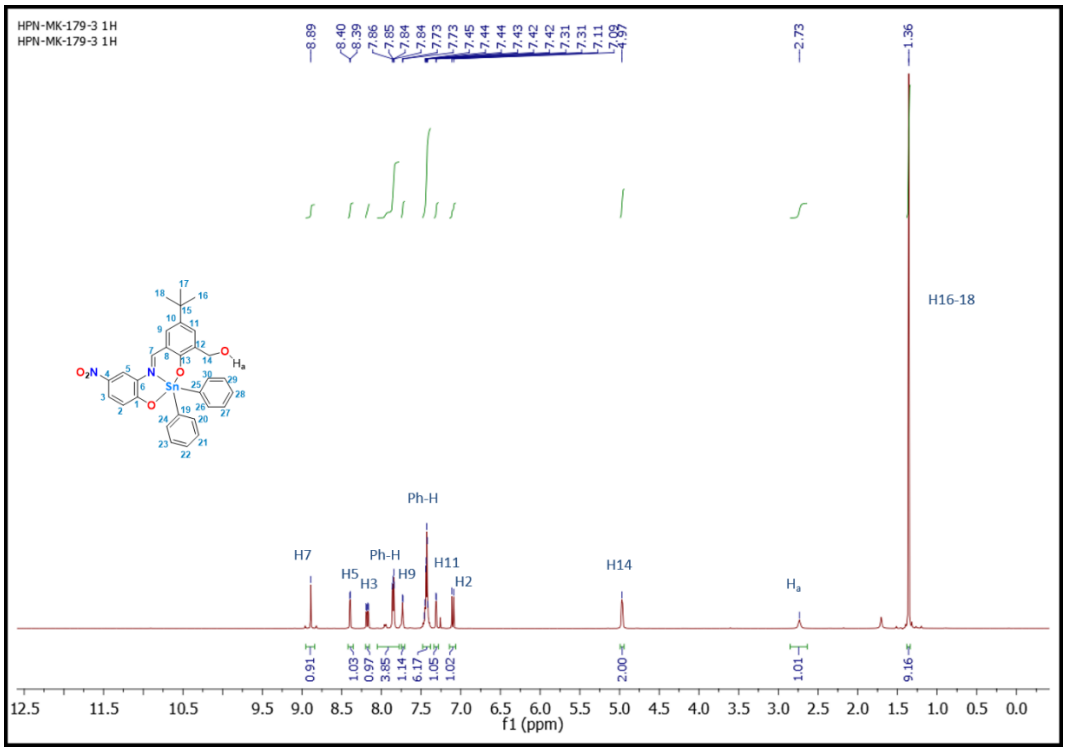


Figure S10: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of compound **1**.

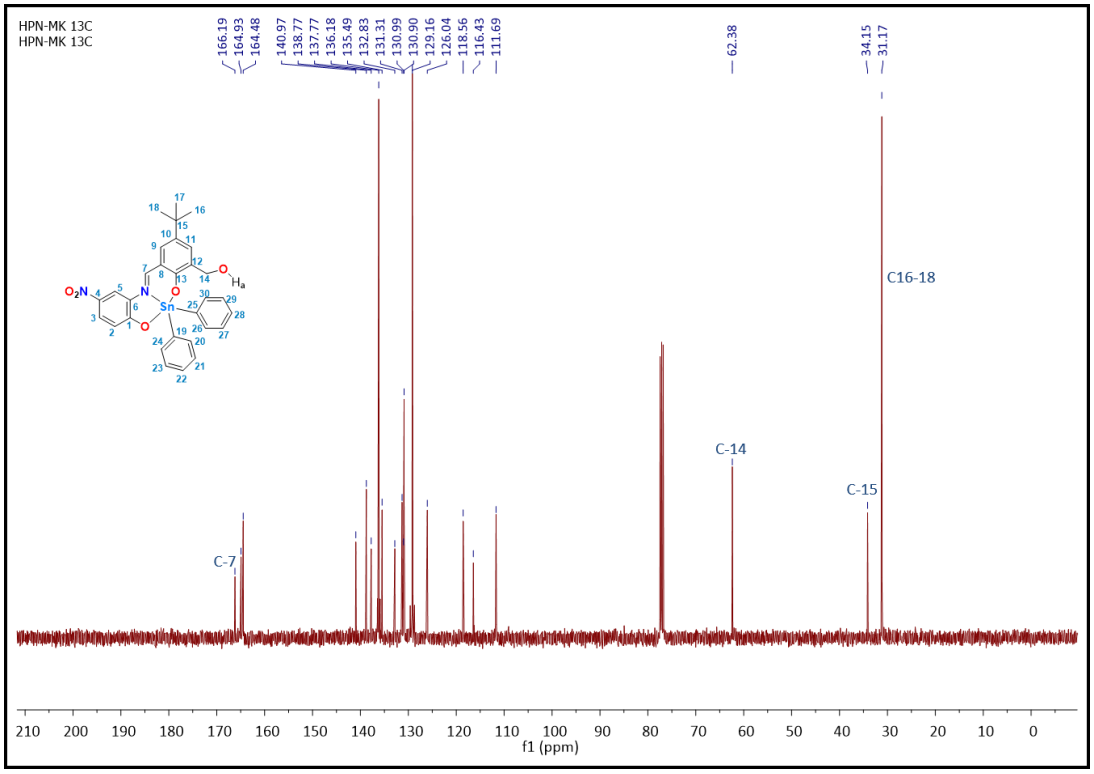


Figure S11: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of compound **1**.

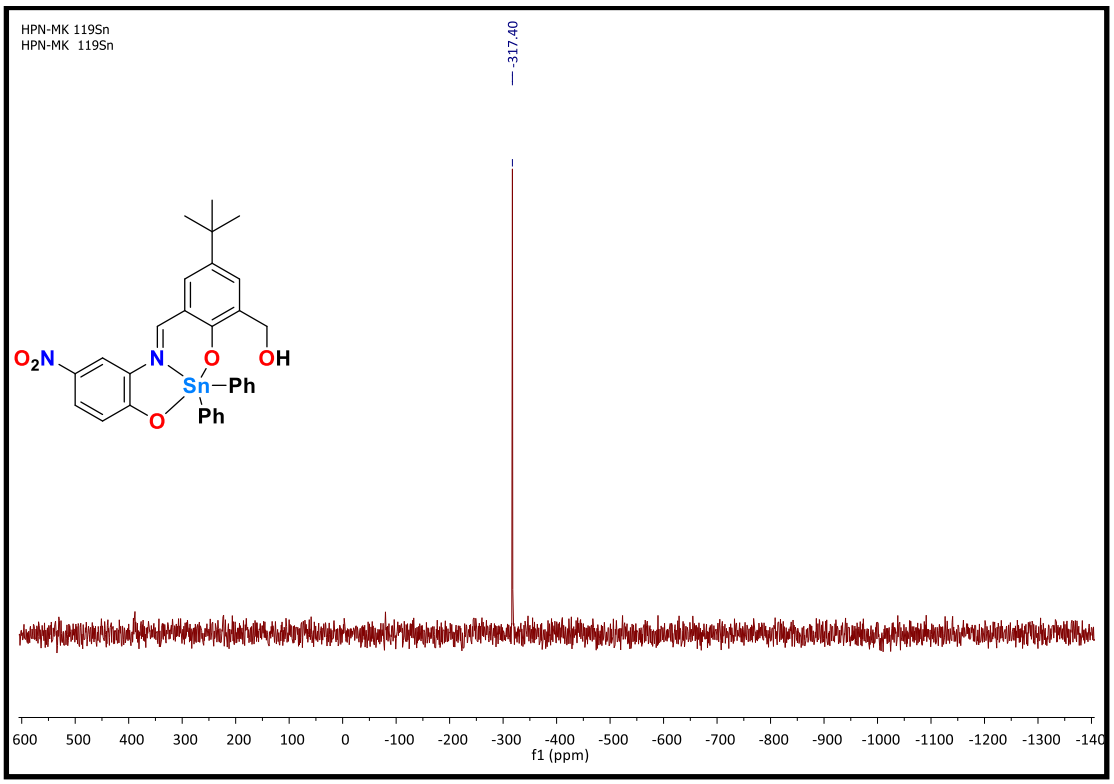


Figure S12: ^{119}Sn NMR (149 MHz, $CDCl_3$) spectrum of compound **1**.

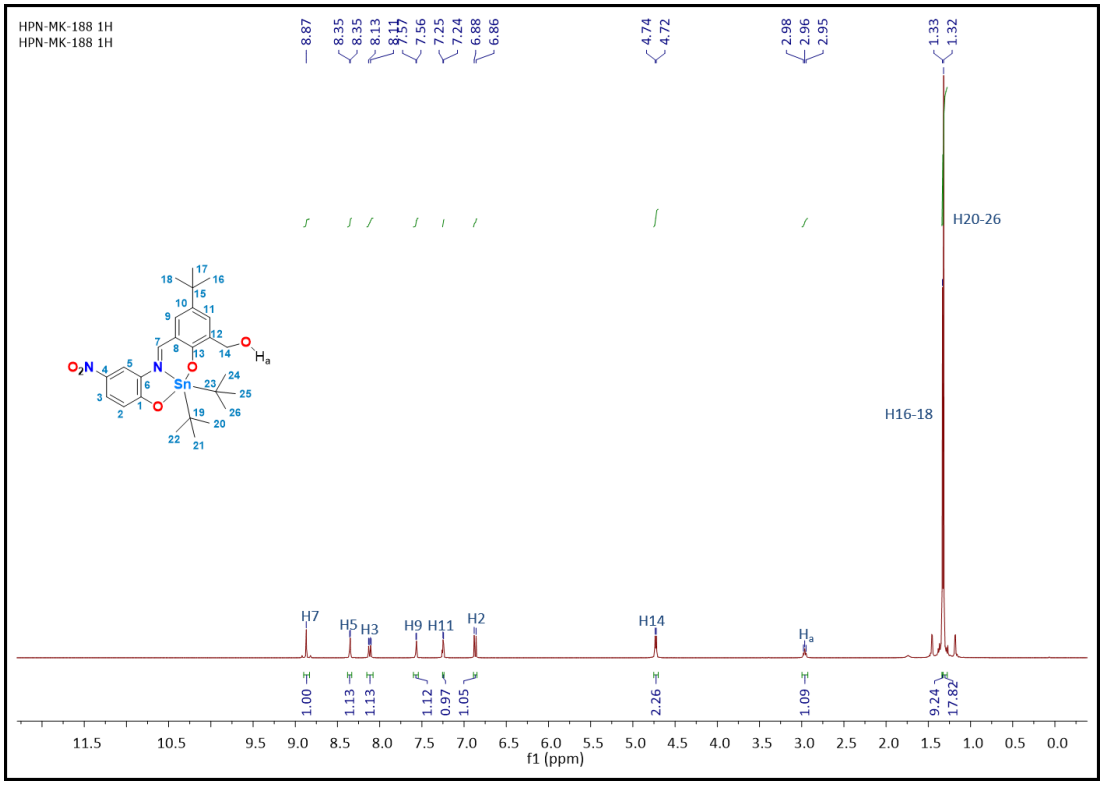


Figure S13: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of compound **2**.

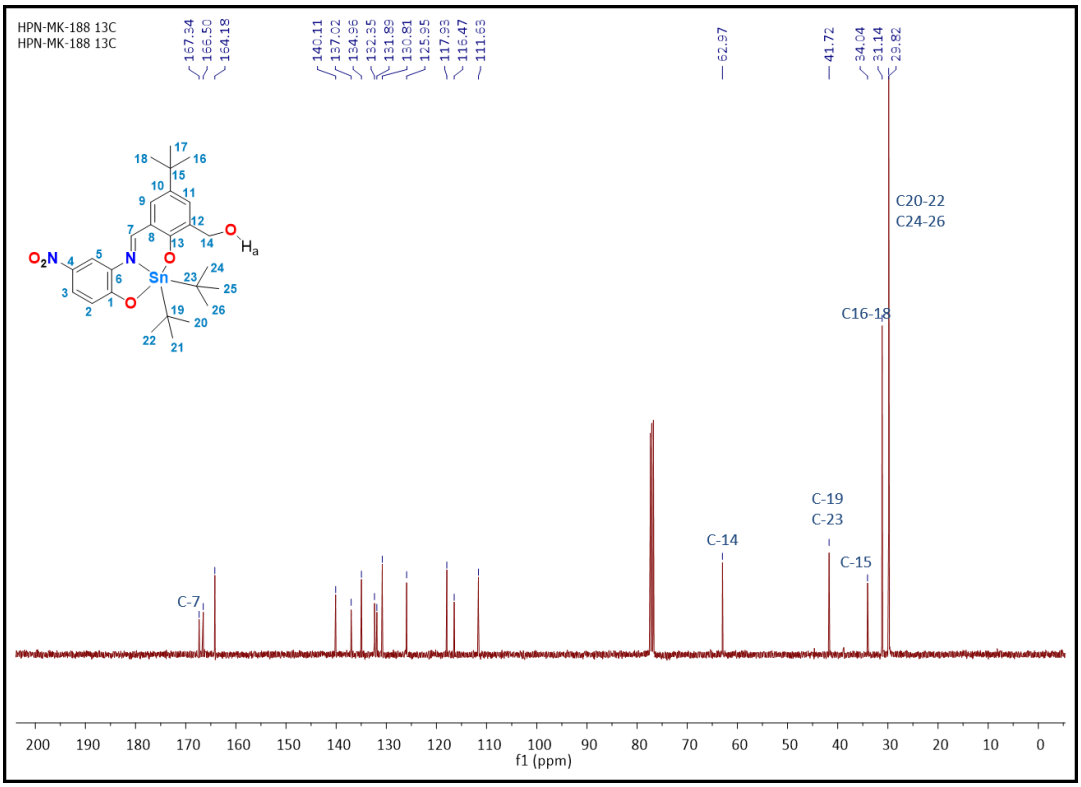


Figure S14: ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) spectrum of compound **2**.

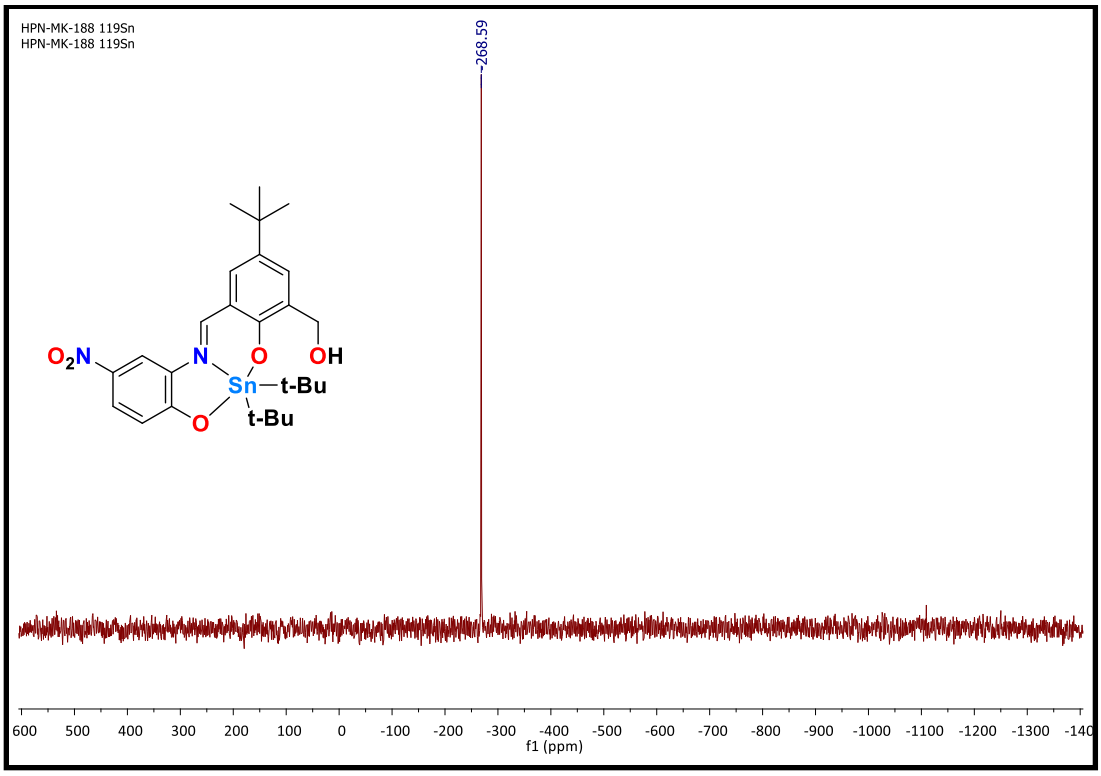


Figure S15: ^{119}Sn NMR (149 MHz, $CDCl_3$) spectrum of compound **2**.

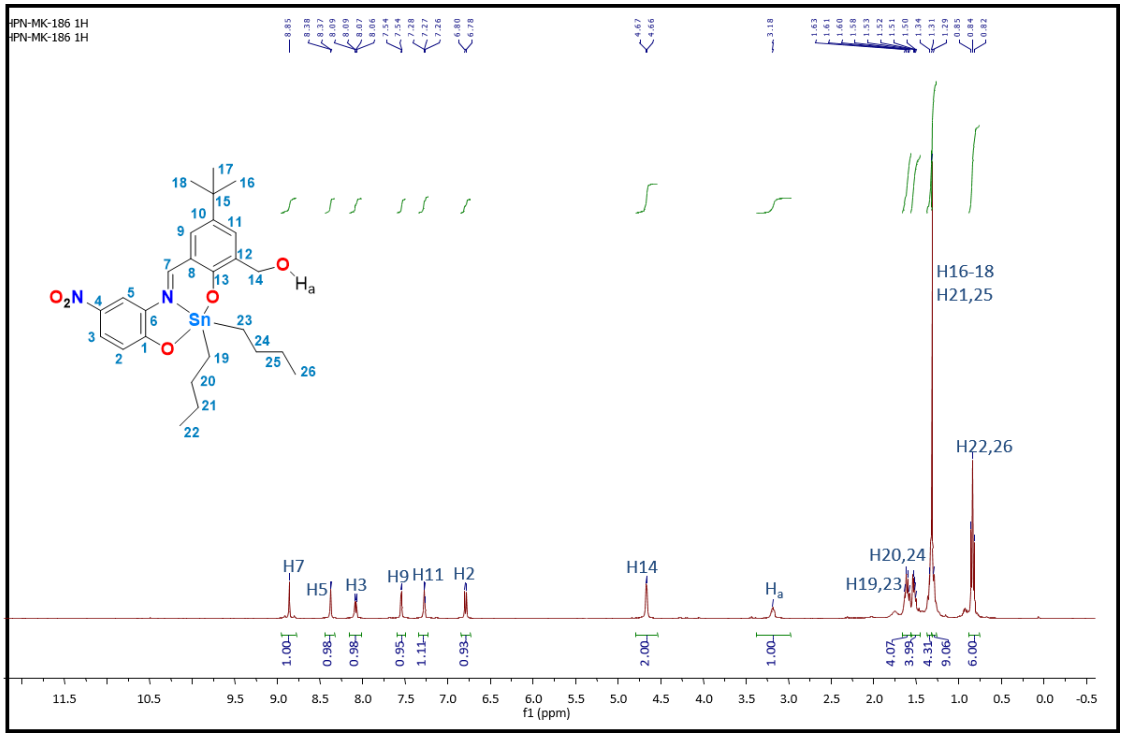


Figure S16: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of compound **3**.

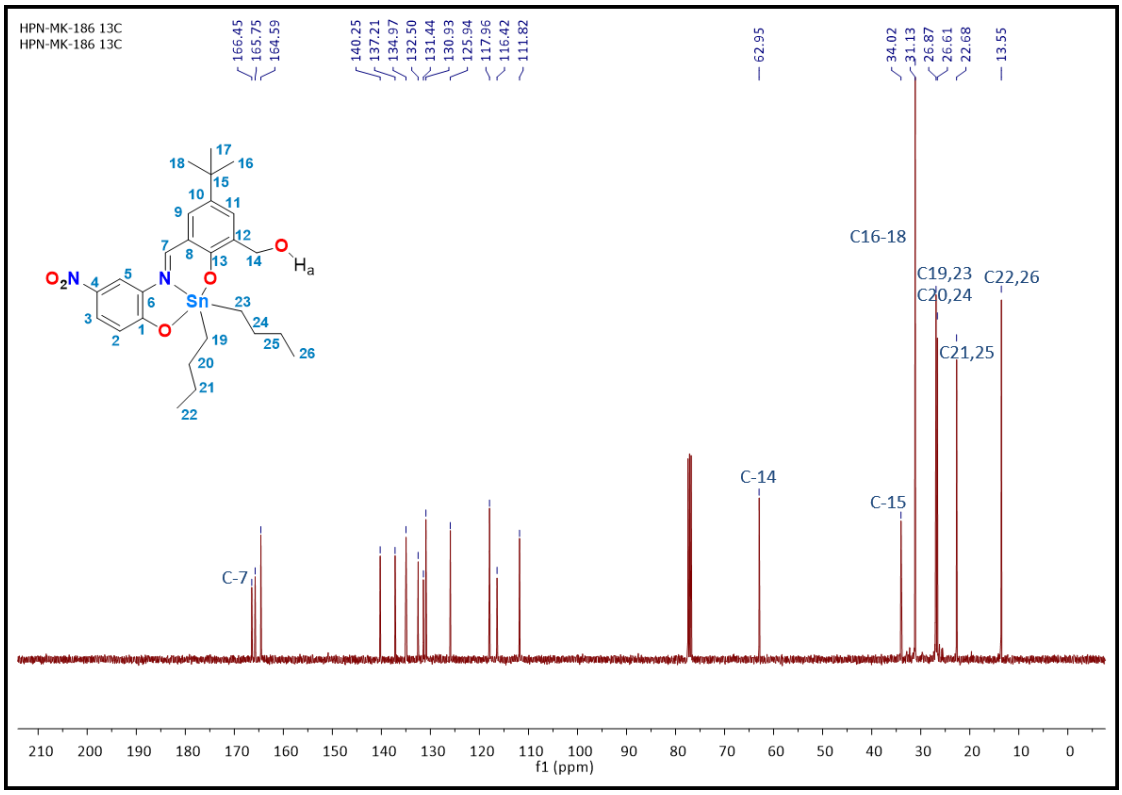


Figure S17: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of compound **3**.

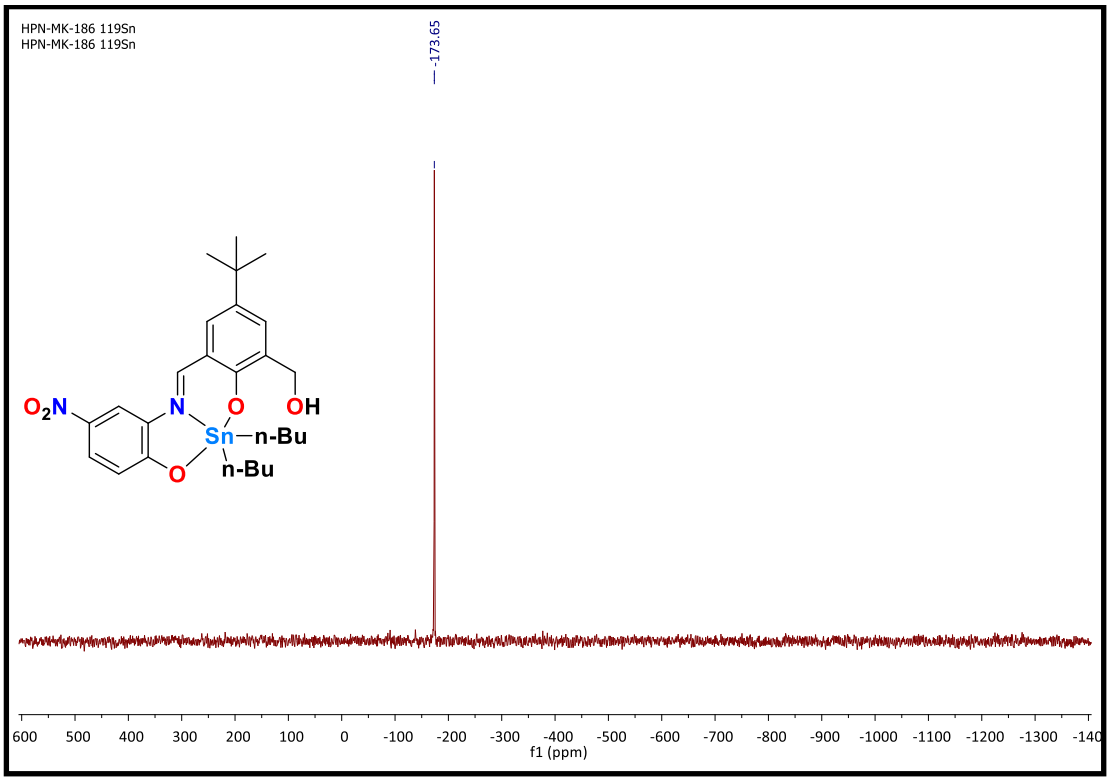


Figure S18: ^{119}Sn NMR (149 MHz, $CDCl_3$) spectrum of compound **3**.

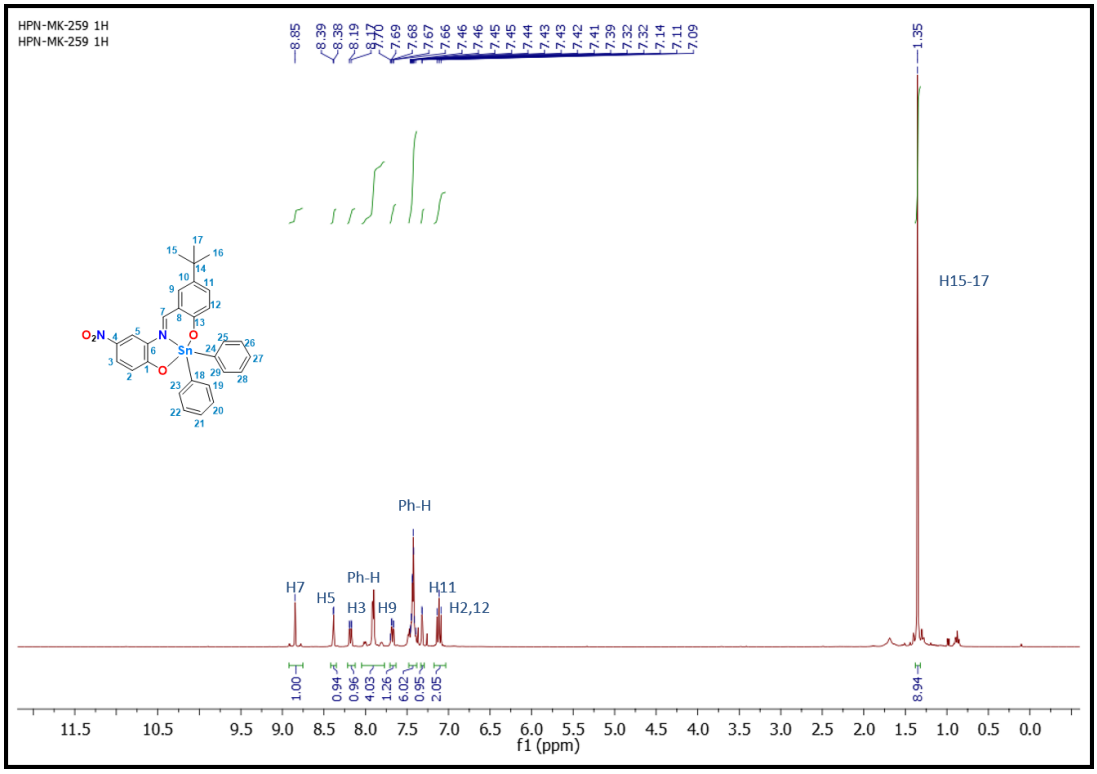


Figure S19: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of compound **4**.

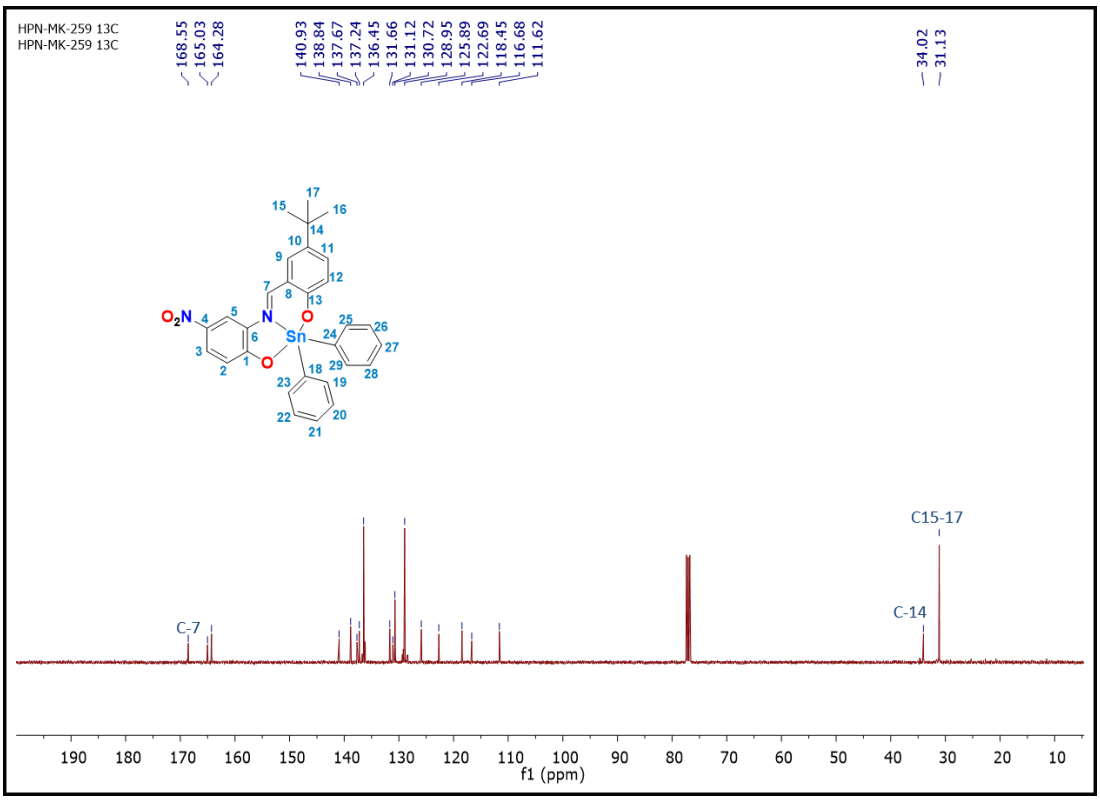


Figure S20: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of compound **4**.

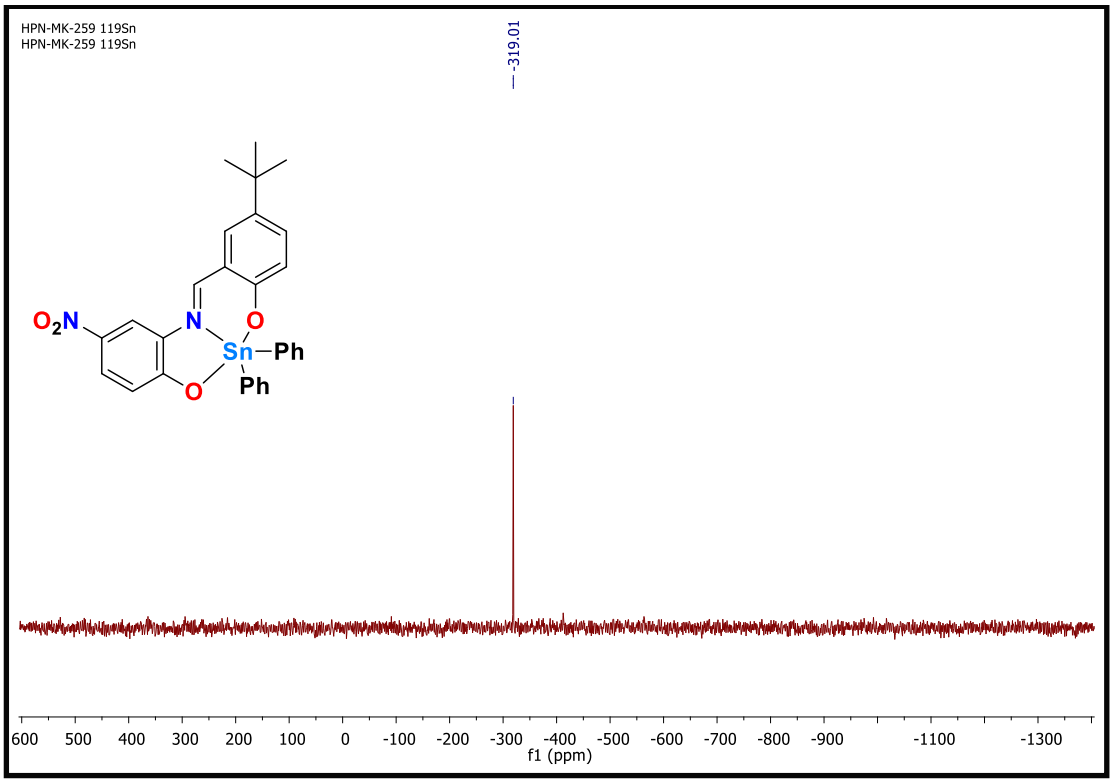


Figure S21: ^{119}Sn NMR (149 MHz, $CDCl_3$) spectrum of compound **4**.

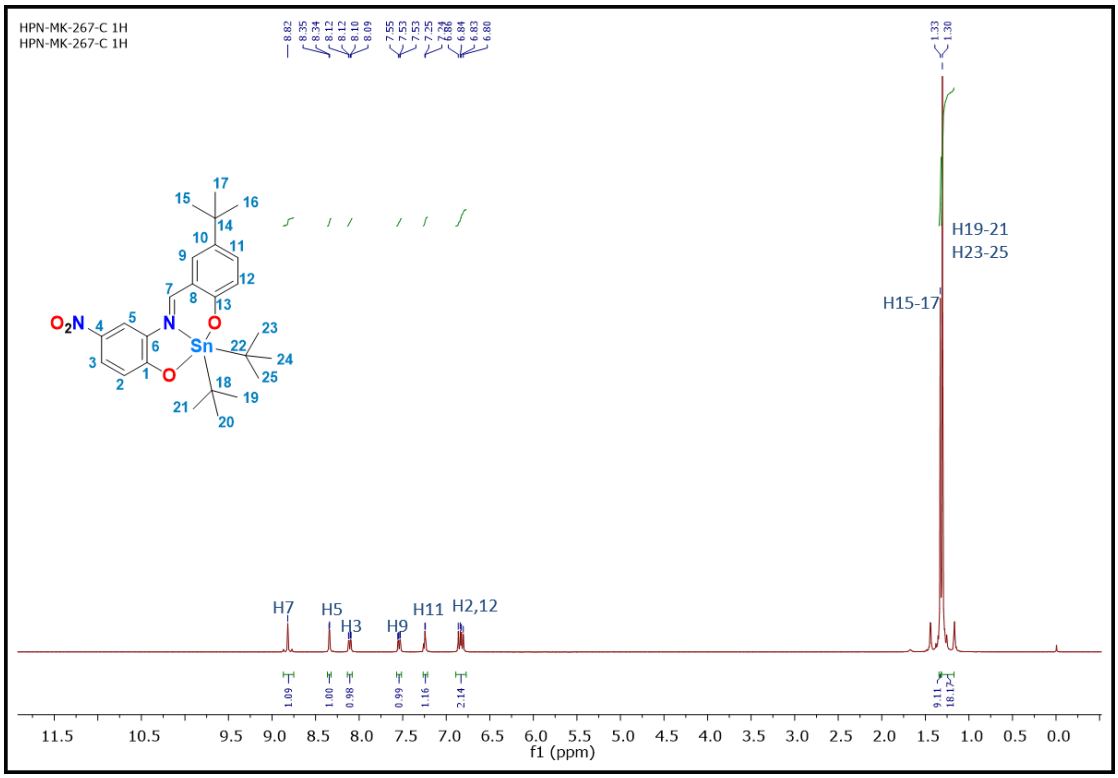


Figure S22: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of compound **5**.

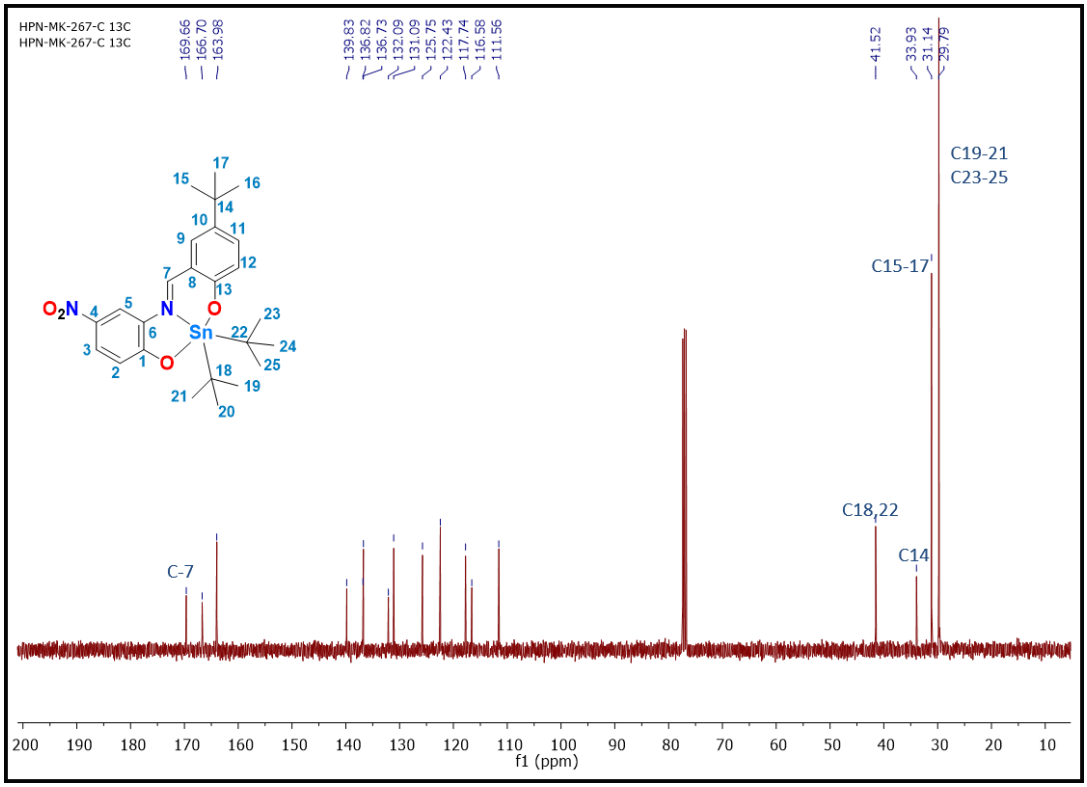


Figure S23: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of compound **5**.

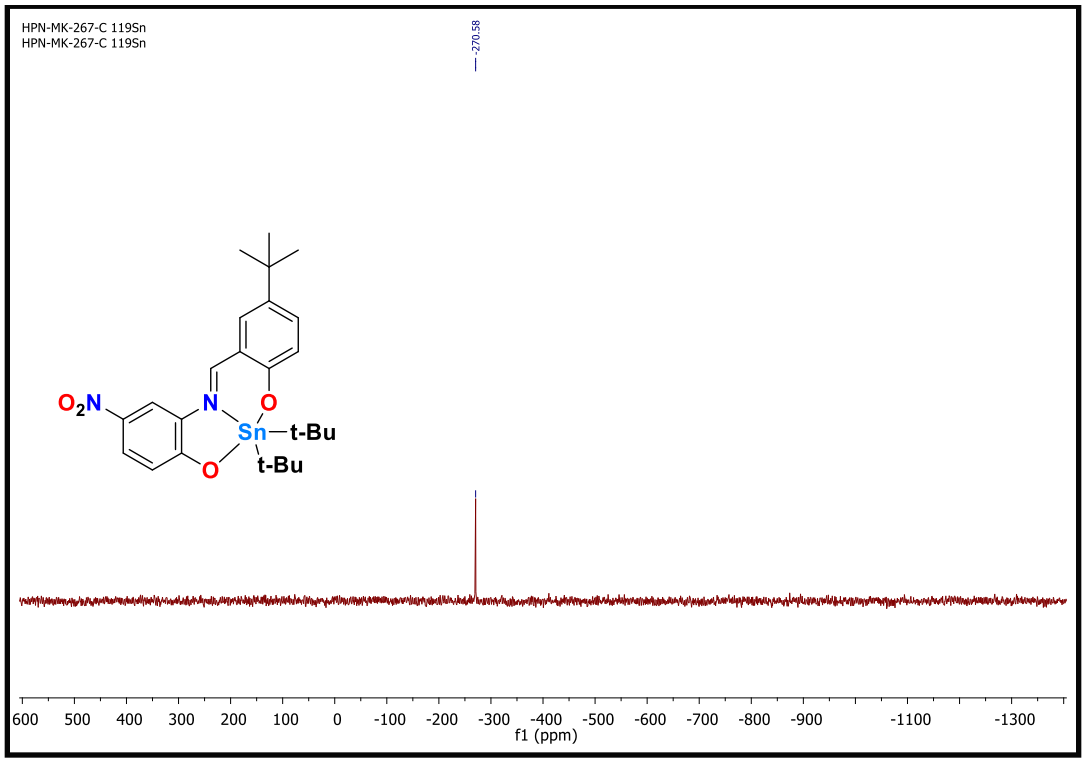


Figure S24: ^{119}Sn NMR (149 MHz, $CDCl_3$) spectrum of compound **5**.

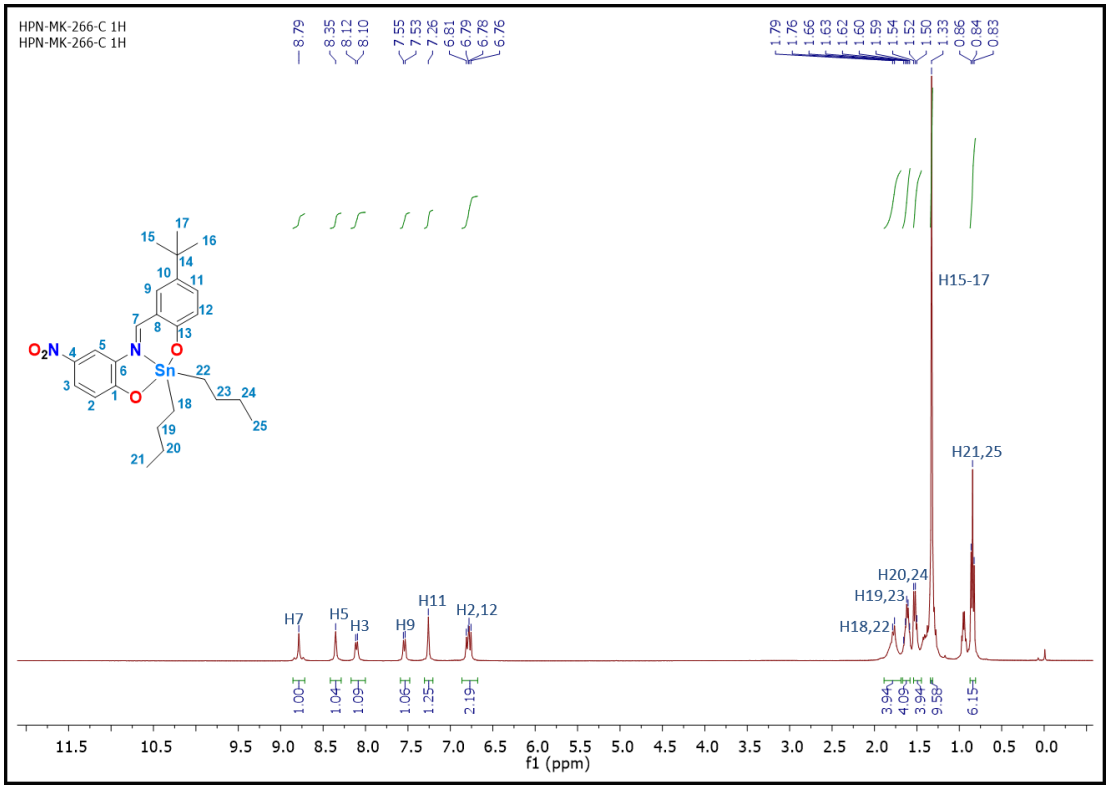


Figure S25: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of compound **6**.

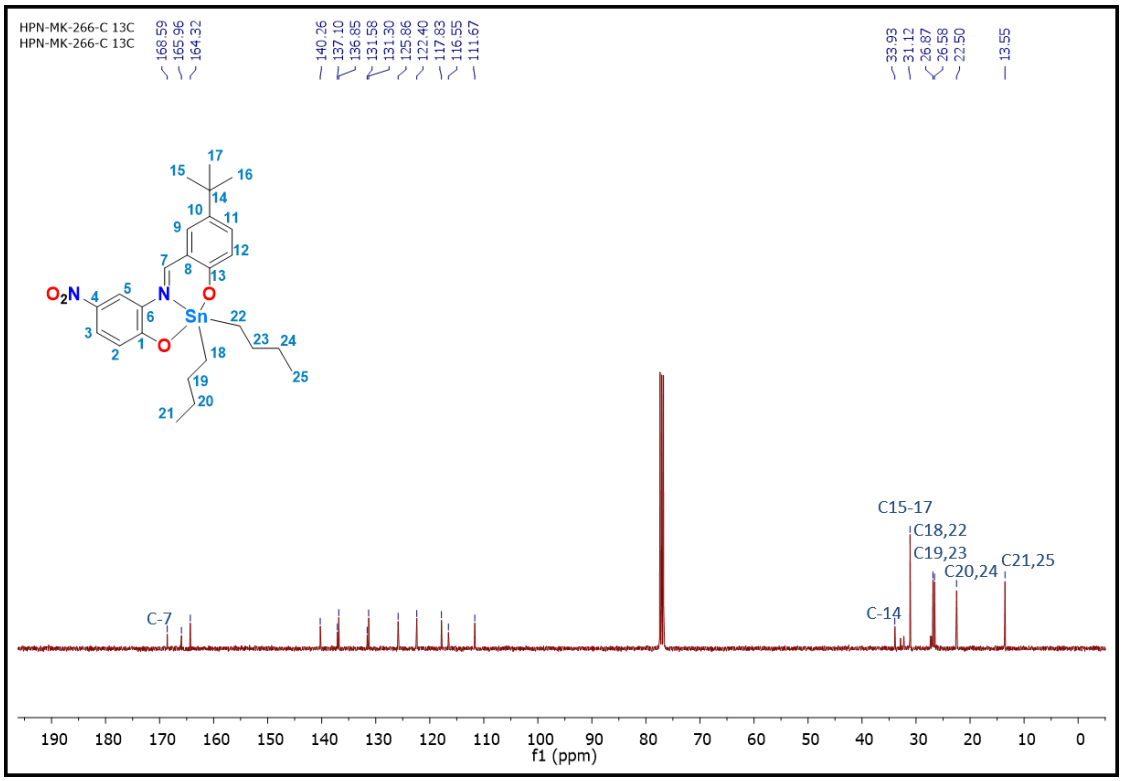


Figure S26: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of compound **6**.

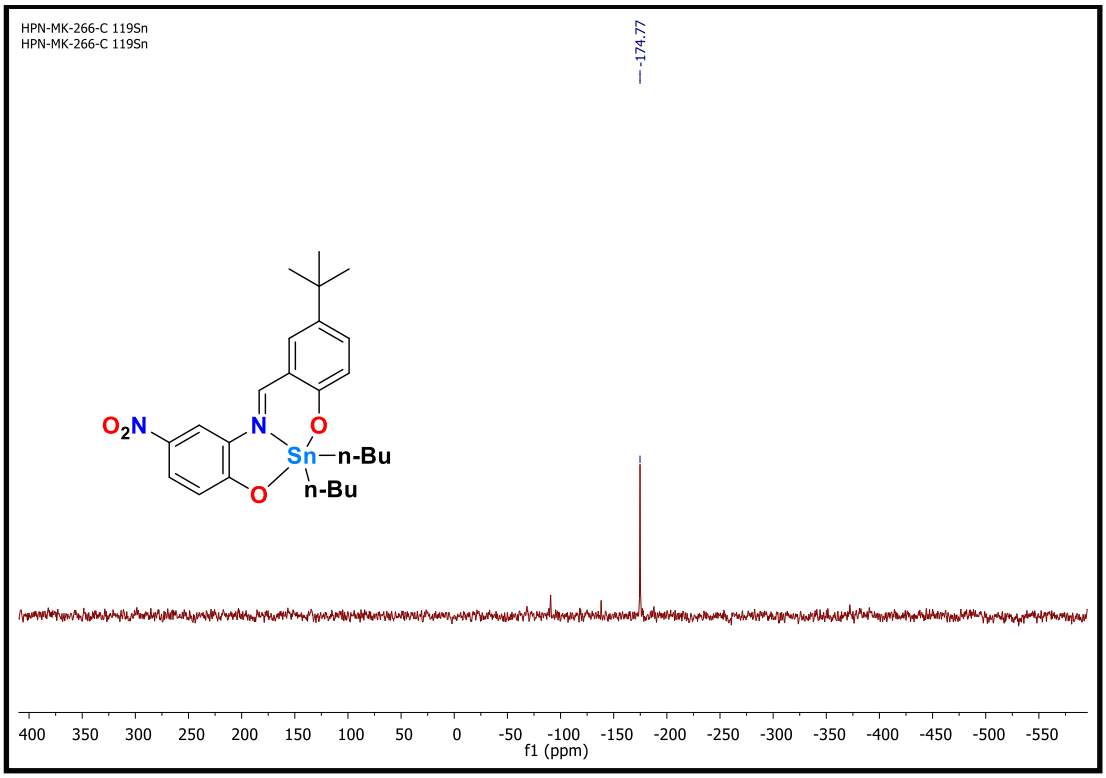


Figure S27: ^{119}Sn NMR (149 MHz, $CDCl_3$) spectrum of compound **6**.

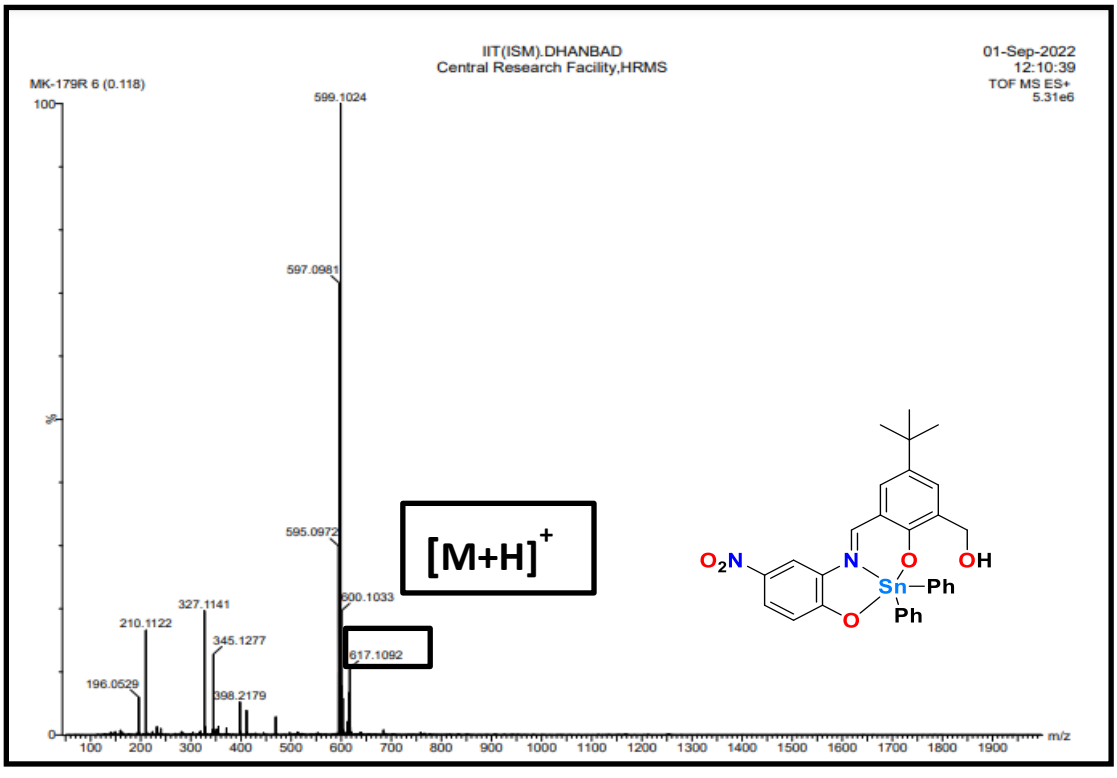


Figure S28: HRMS (ESI) spectrum of compound **1**.

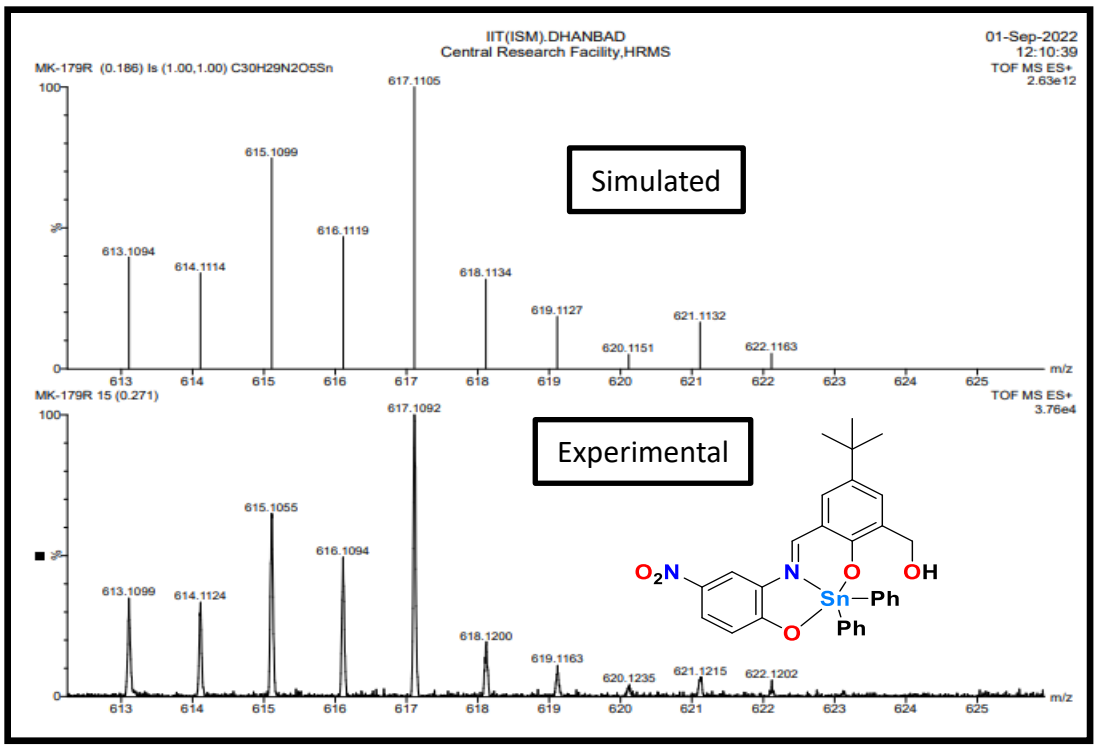


Figure S29: HRMS (ESI) spectrum of **[M+H]**$^{+}$ ion of compound **1**.

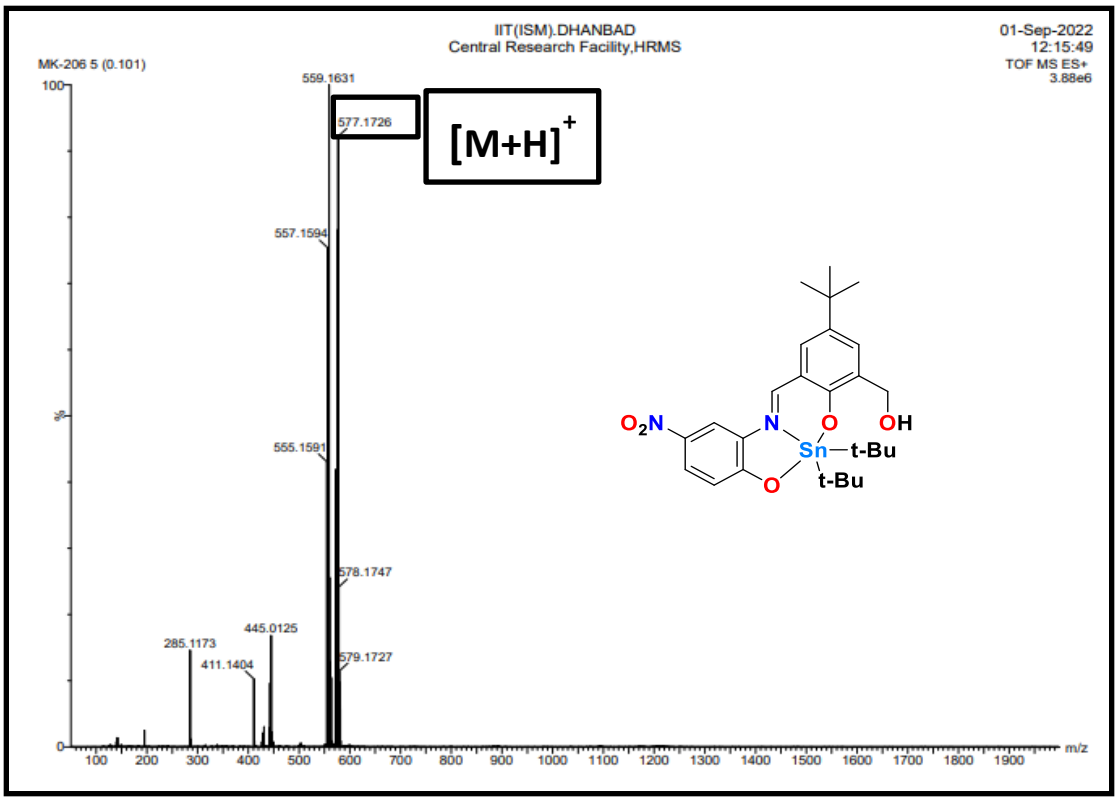


Figure S30: ESI-MS spectrum of compound **2**.

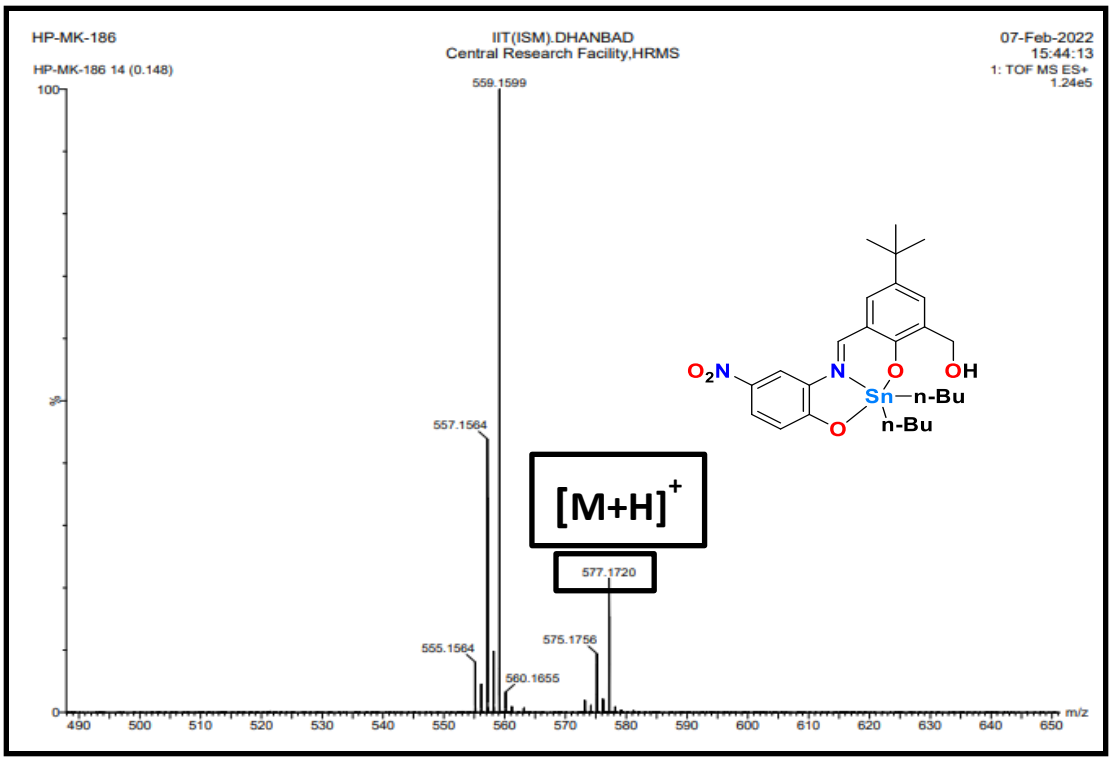


Figure S31: HRMS (ESI) spectrum of compound **3**.

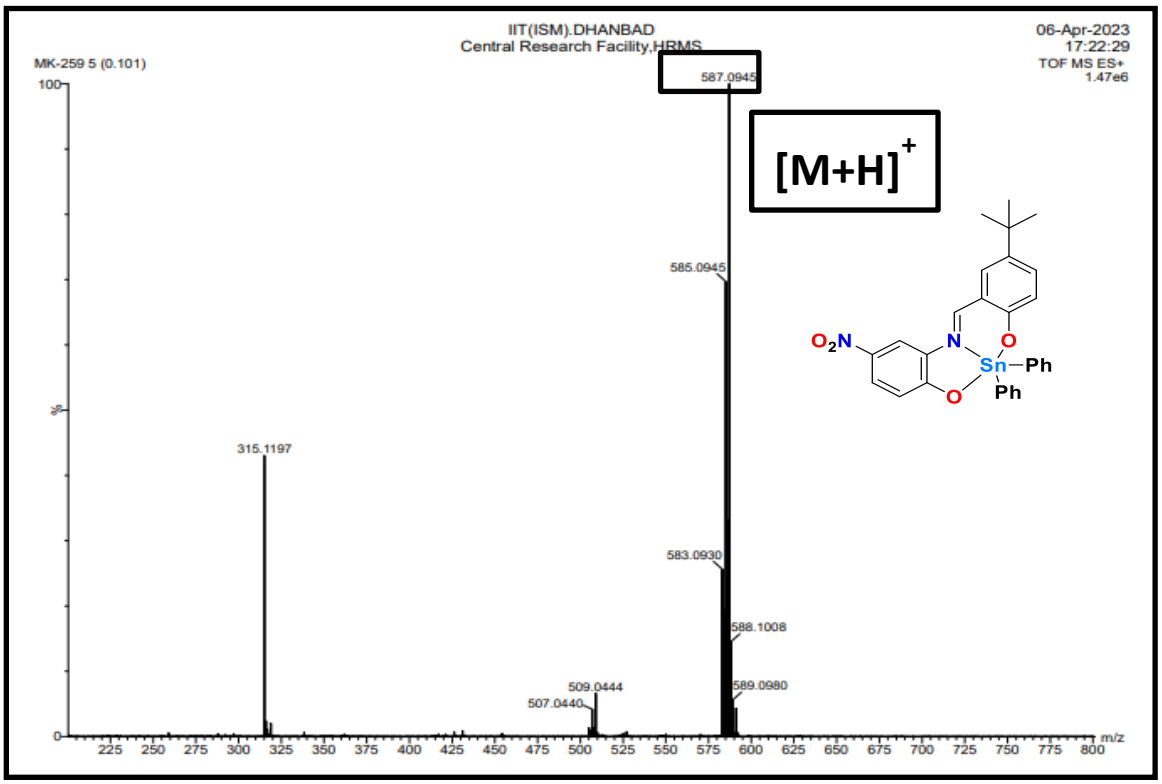


Figure S32: HRMS (ESI) spectrum of compound **4**.

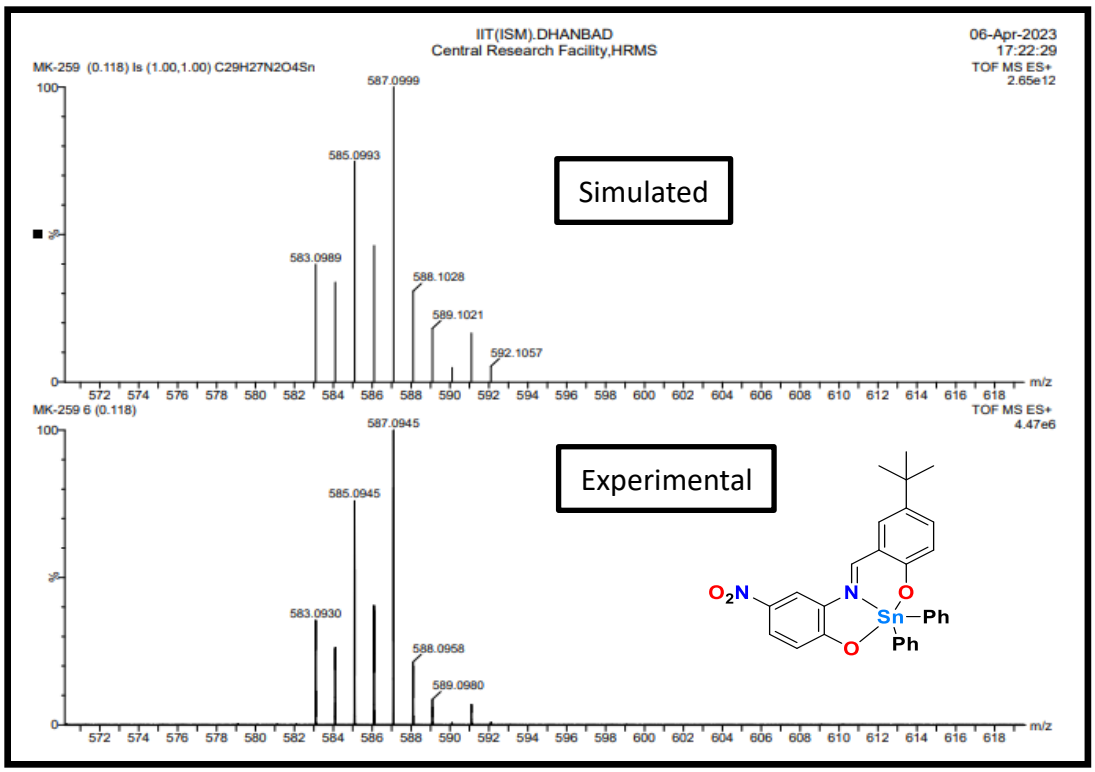


Figure S33: HRMS (ESI) spectrum of **[M+H]**$^+$ ion of compound **4**.

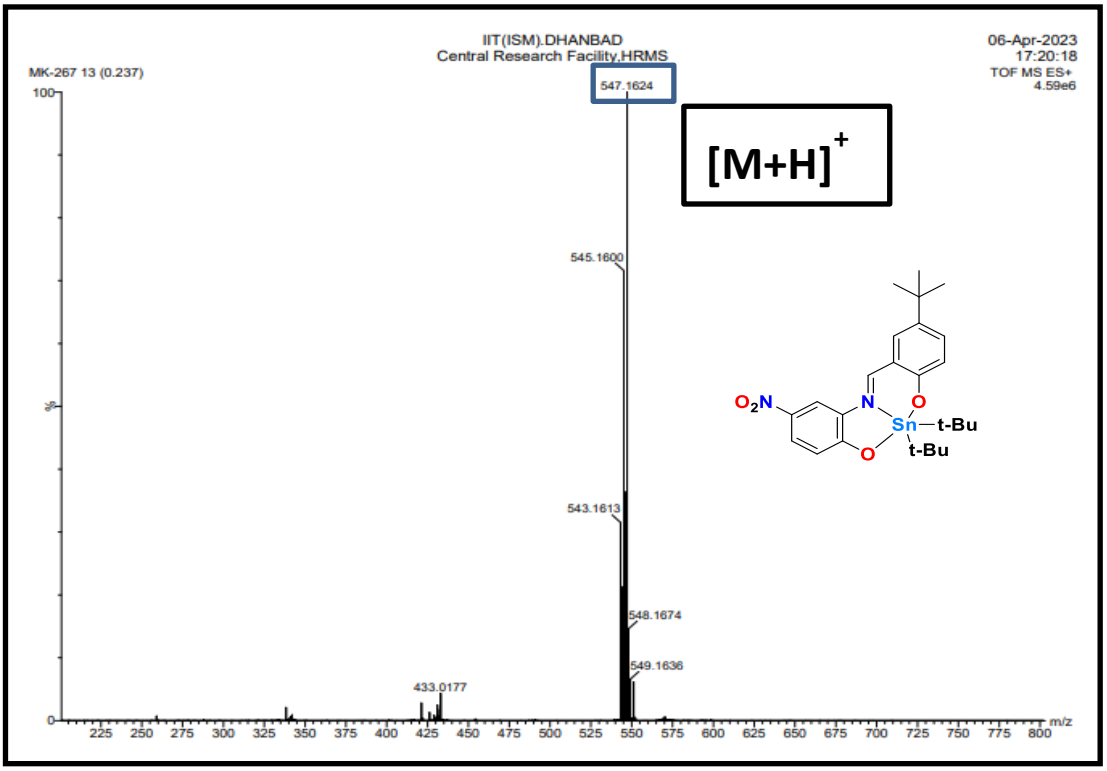


Figure S34: HRMS (ESI) spectrum of compound **5**.

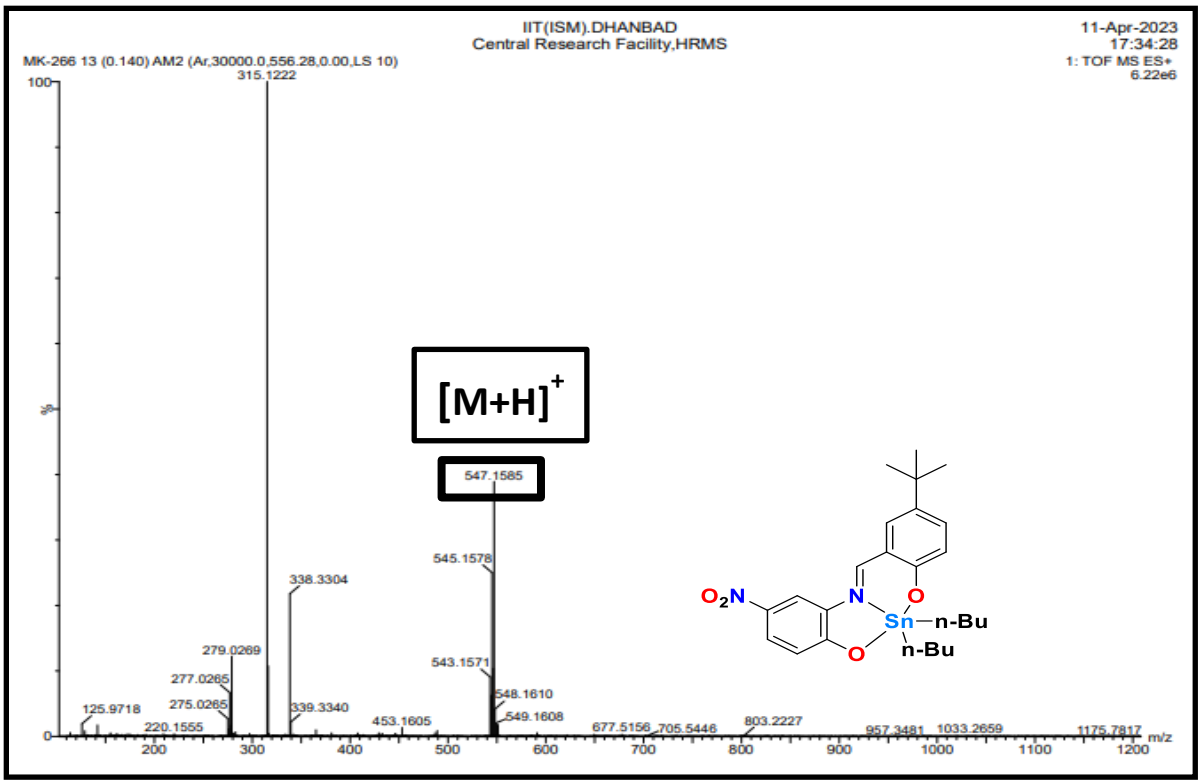


Figure S35: HRMS (ESI) spectrum of compound **6**.

X-ray crystallography:

The crystallographic data of **1-6** were collected on a Rigaku SuperNova diffractometer equipped with an Eos S2 CCD detector, using Mo-Kα radiation with graphite monochromator (λ = 0.71073 Å) at T = 293(2) K. The structure was solved with the ShelXT 2014/5 (Sheldrick, 2014) structure solution program and by using Olex2 as the graphical interface.[1,2] The model was refined with version 2018/3 of XL using Least Squares minimization.[3] Non-hydrogen atoms were anisotropically refined. H-atoms were included in the refinement of calculated positions riding on their carrier atoms. The function minimized was $[\sum w(Fo^2 - Fc^2)^2]$ ($w = 1 / [\sigma^2 (F_o^2) + (aP)^2 + bP]$), where $P = (\mathrm{Max}(F_o^2,0) + 2F_c^2) / 3$ with $\sigma^2(F_o^2)$ from counting statistics. The function $R1$ and $wR2$ were $(\sigma||F_o| - |F_c||) / \sigma|F_o|$ and $[\sigma w (F_o^2 - F_c^2)^2 / \sigma(wF_o^4)]^{1/2}$, respectively. Specific refinement details: Compound **2**: O3 atom is split over two positions O3A and O3B with 57% and 43% respectively. Regarding compound **3**, some highly disordered carbon atoms were refined isotropically. Compound **5**: three disordered carbon atoms (C19-C21) are split over two positions with 57% and 43% occupancy. Compound **6**: C22-C25 atoms are split over two positions with 59% and 41% occupancy respectively. C15-C17 are split over two position with 84% and 16% occupancy. CCDC 2270674-2270679 contain the supplementary crystallographic data for compounds **1-6**.

These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

Table S1 : Single crystal data collection and data refinement parameters for compounds **1-6.**

Compound	**1· MeOH**	**2**	**3**	**4**	**5**	**6**
Formula	$C_{31}H_{32}N_2O_6Sn$	$C_{26}H_{36}N_2O_5Sn$	$C_{52}H_{72}N_4O_{10}Sn_2$	$C_{29}H_{26}N_2O_4Sn$	$C_{25}H_{34}N_2O_4Sn$	$C_{25}H_{34}N_2O_4Sn$
Formula mass	647.320	575.26	1150.51	585.21	545.23	545.270
T/K	293(2)	293(2)	293(2)	293(2)	293(2)	293(2)
λ/Å	0.71073	0.71073	0.71073	0.71073	0.71073	0.71073
Crystal system	triclinic	triclinic	triclinic	triclinic	triclinic	triclinic
Space group	*P*-1	*P*-1	*P*-1	*P*-1	*P*-1	*P*-1
a/Å	10.7364(3)	11.8788(5)	10.3278(6)	10.4626(3)	11.6108(4)	10.3748(7)
b/Å	12.0005(3)	13.7443(3)	15.8695(8)	12.2766(4)	13.7661(4)	10.7560(6)
c/Å	12.1066(3)	18.5417(4)	17.9400(12)	12.4511(5)	17.6211(5)	13.2715(6)
α/°	90.921(2)	70.101(2)	112.274(5)	64.844(4)	79.327(2)	90.075(4)
β/°	97.576(2)	81.539(2)	93.904(5)	65.315(3)	81.161(3)	110.462(5)
γ/°	112.023(2)	76.408(3)	96.094(4)	66.761(3)	77.032(3)	111.715(6)
V/Å^3	1429.75(7)	2759.29(2)	2686.7(3)	1268.69(9)	2678.62(15)	1274.47(1)
Z	2	4	2	2	4	2
D_c/g cm^{-3}	1.504	1.382	1.422	1.532	1.352	1.421
μ/mm^{-1}	0.940	0.962	0.988	1.045	0.984	1.034
F(000)	659	1184	1184	592	1120	559
2θ Range/°	4.14 to 54.00	4.096 to 54.00	3.994-54.00	3.8 to 54.00	4.164 to 54.00	4.12 to 50
Measured reflections	23232	44097	25148	19775	23275	11420
Independent reflections/R_{int}	6211/R_{int} = 0.0620	11981[R_{int} = 0.0608	11557[R_{int} = 0.0424	5491 [R_{int} = 0.0431,	11494 [R_{int} = 0.0349	4491 [R_{int} = 0.0559
Parameters	367	642	573	328	611	375
R_1 (I >2σ(I))[a]	0.0275	0.0458	0.0585	0.0302	0.0382	0.0392
wR_2 (all data)[b]	0.0667	0.1158	0.1733	0.0749	0.0779	0.0727
Goodness-of-fit on F^2	1.052	1.056	1.039	1.047	0.964	0.887
$\Delta\rho_{max,\ min}$/e Å^{-3}	0.65/-0.54	1.41/-0.62	1.35/-1.10	0.46/-0.53	0.43/-0.55	0.88/-0.76

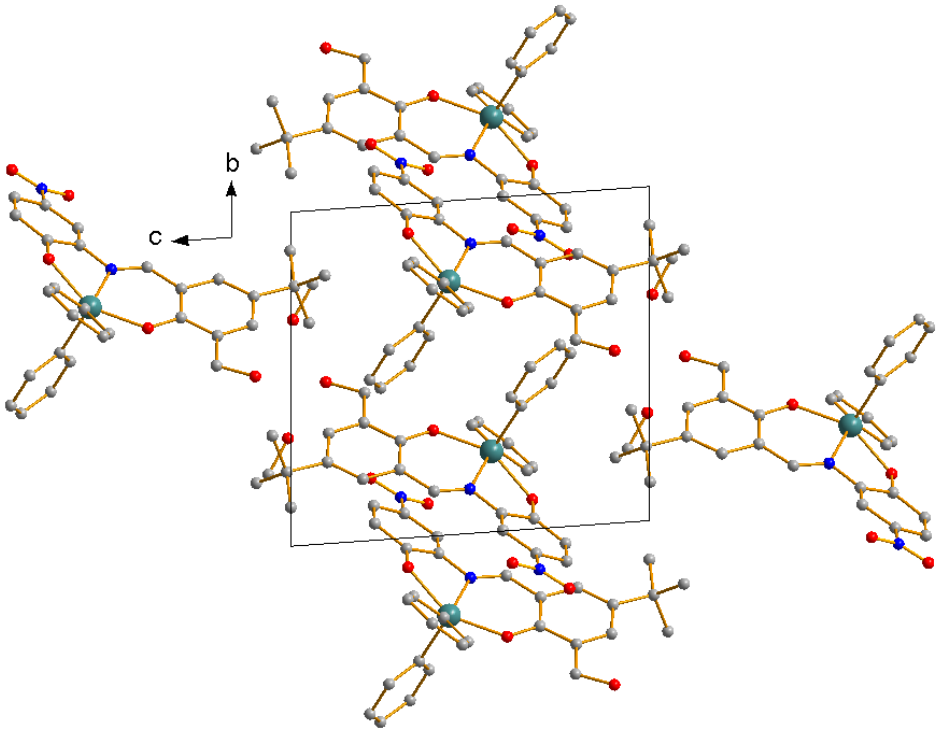


Figure S36: Packing of **1** within the unit cell viewed along crystallographic *a* axis.

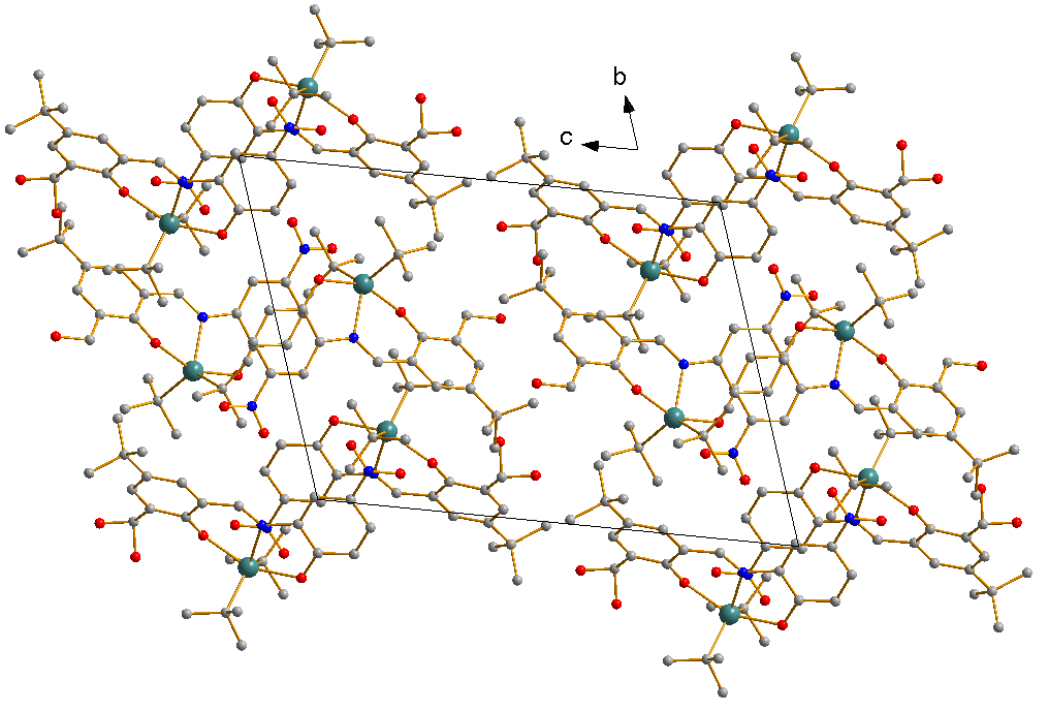


Figure S37: Packing of **2** within the unit cell viewed along crystallographic *a* axis.

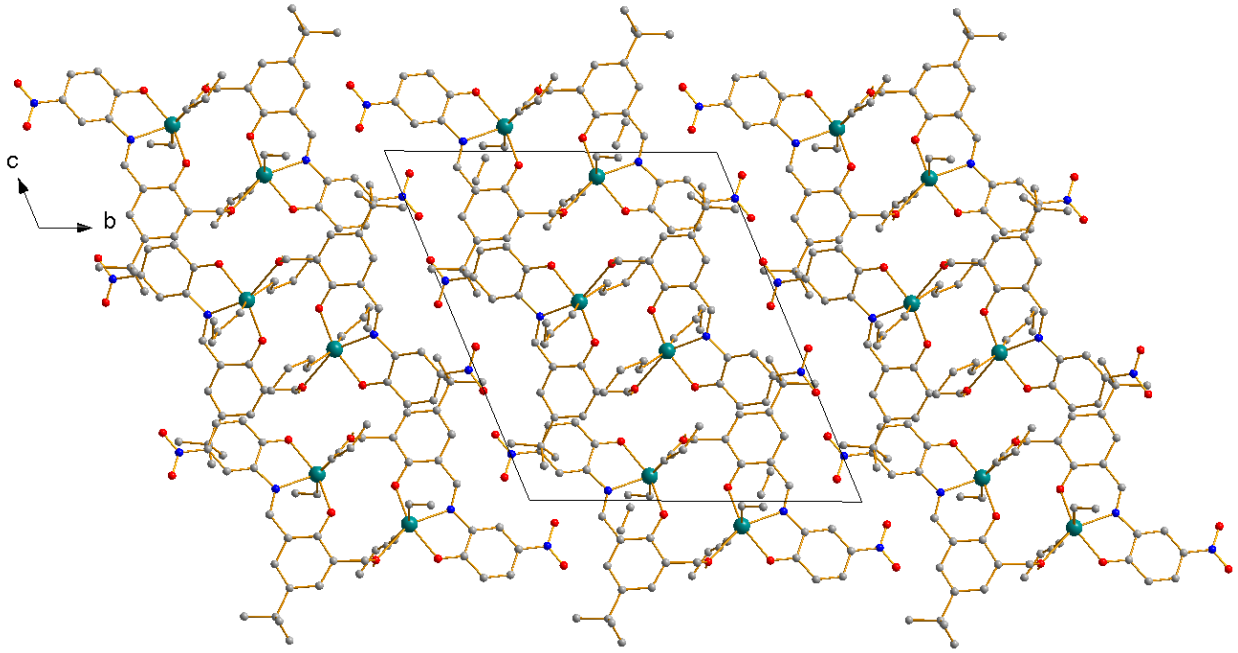


Figure S38: Packing of **3** within the unit cell viewed along crystallographic *a* axis.

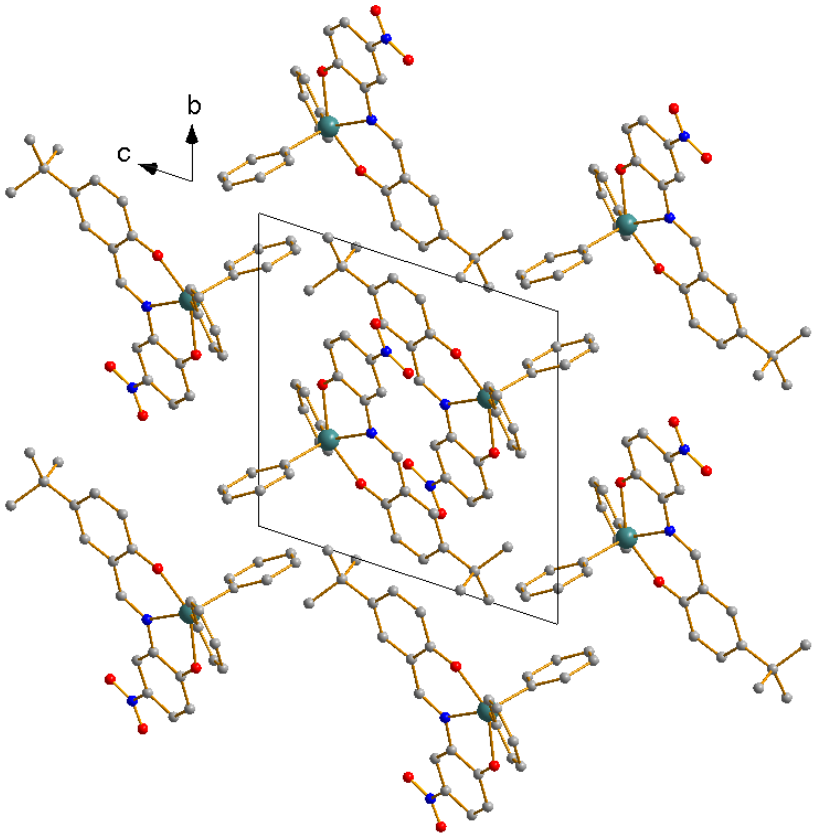


Figure S39: Packing of **4** within the unit cell viewed along crystallographic *a* axis.

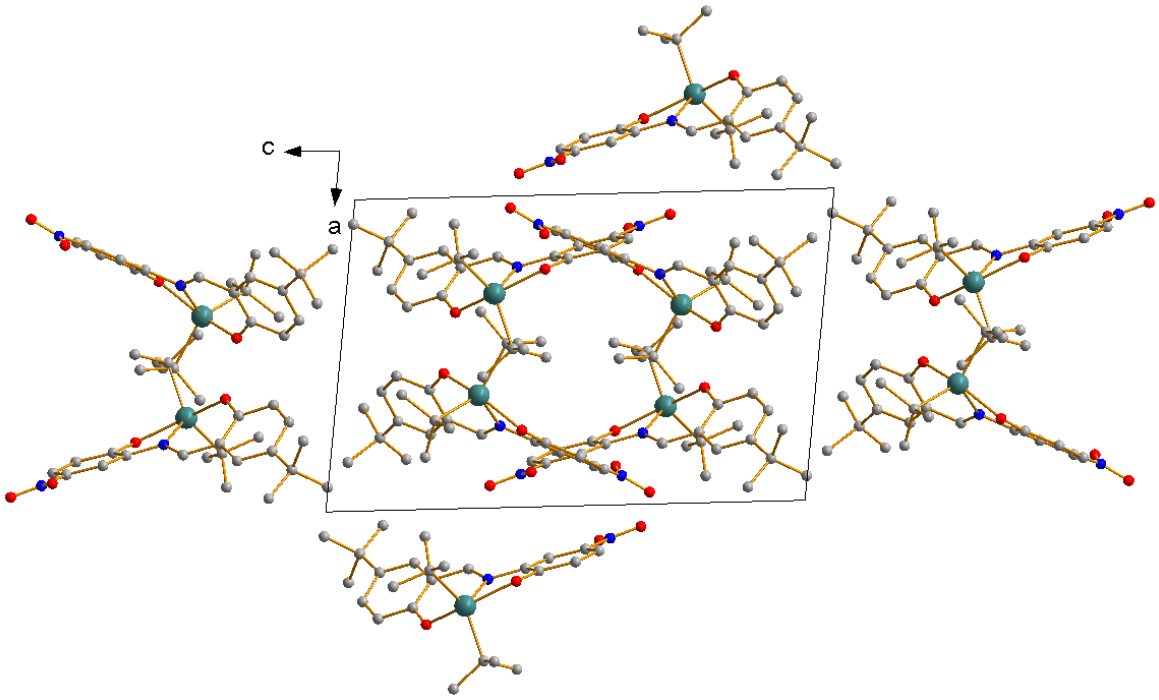


Figure S40: Packing of **5** within the unit cell viewed along crystallographic *b* axis.

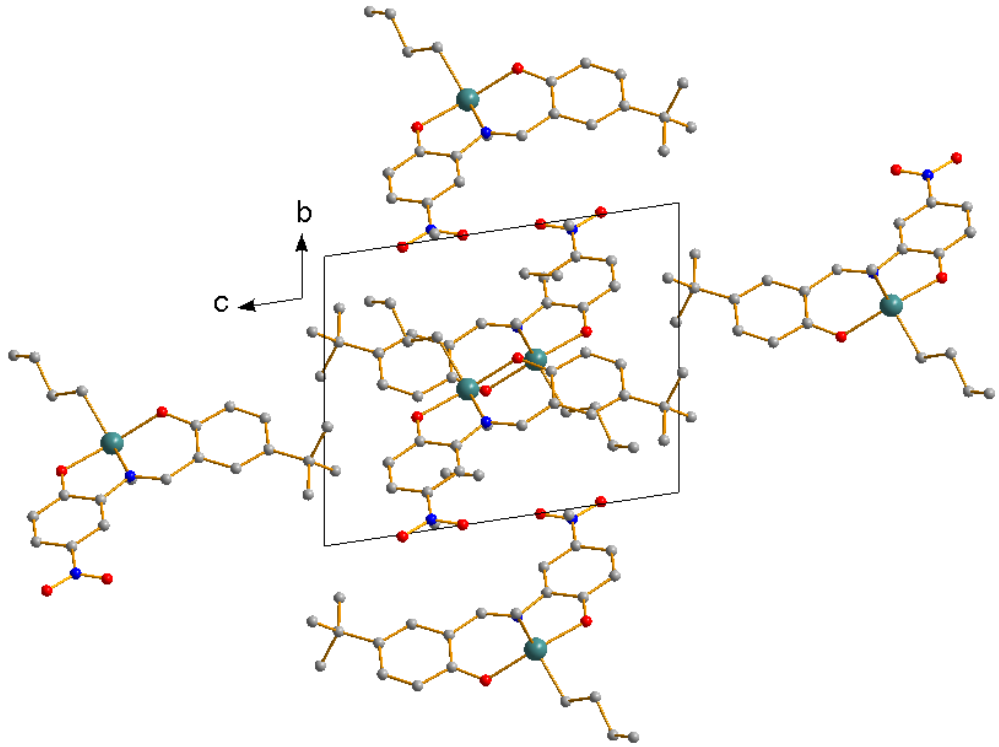
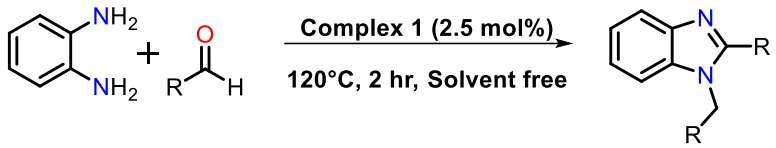
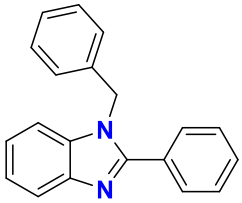
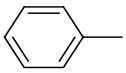
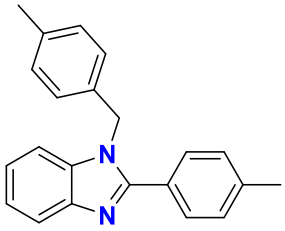
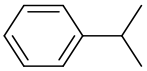
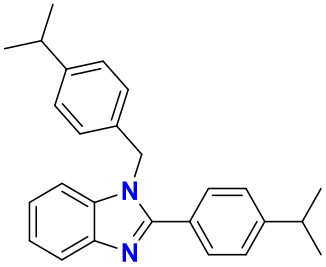
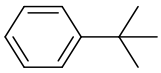
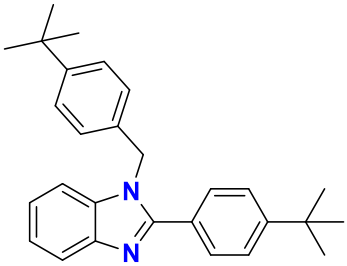
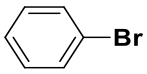
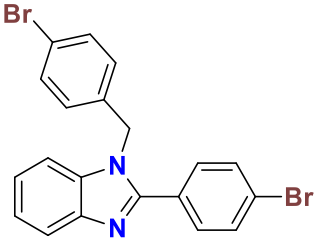
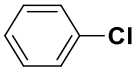
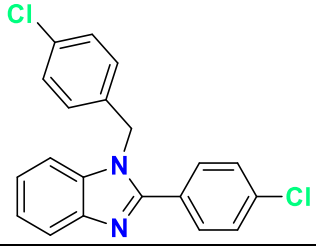


Figure S41: Packing of **6** within the unit cell viewed along crystallographic *a* axis.

Table S2: Isolated yield of 1,2- disubstituted benzimidazoles.



Entry	R	Product	Code	Yield(%)	TON	TOF (h^{-1})
1			2a	92	36.8	18.4
2			2b	82	32.8	16.4
3			2c	85	34	17
4			2d	87	34.8	17.4
5			2e	81	32.4	16.2
6			2f	79	31.6	15.8

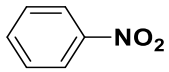
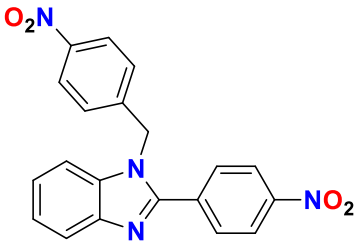
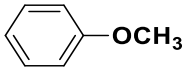
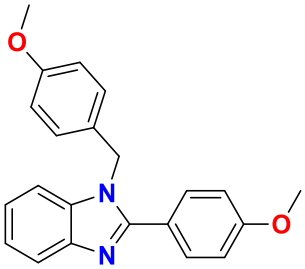
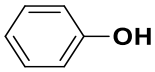
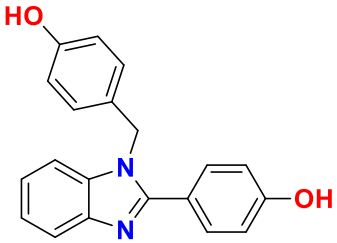
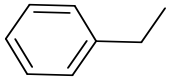
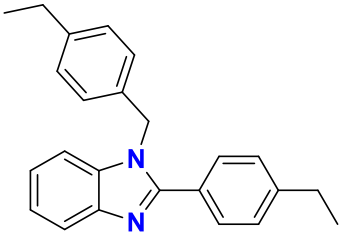
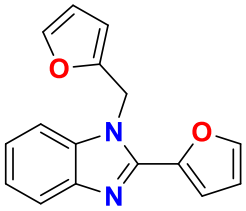
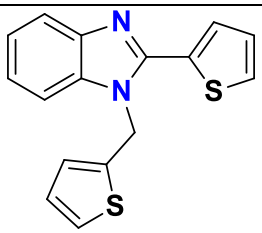
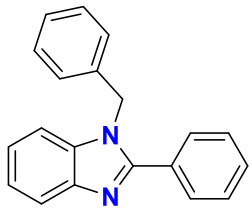
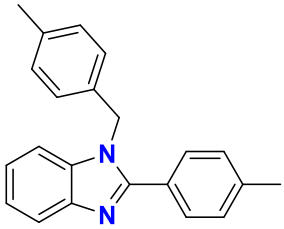
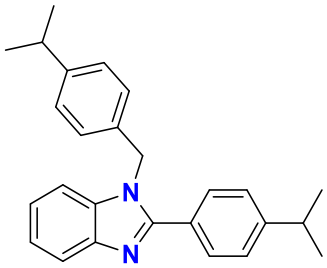
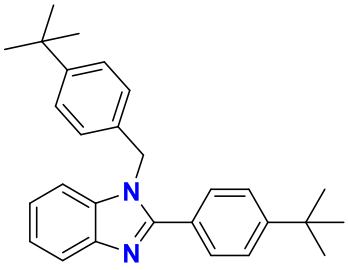
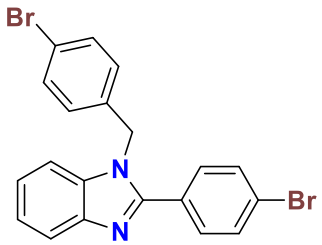
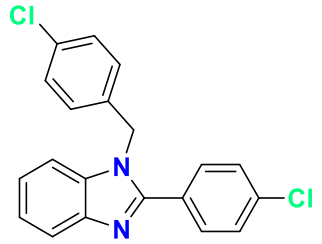
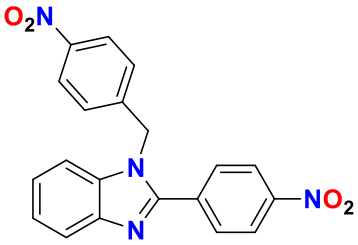
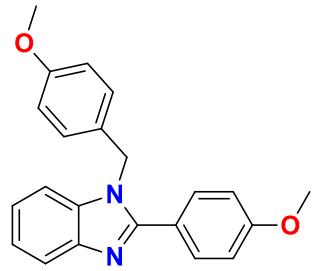
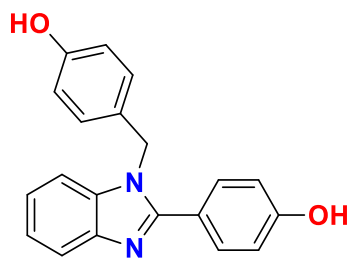
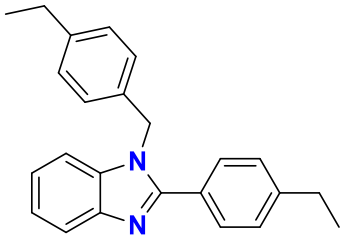
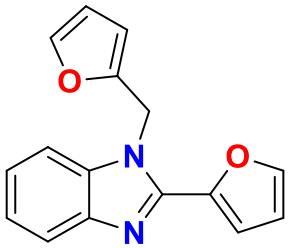
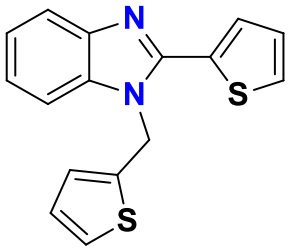
7			2g	72	28.8	14.4
8			2h	86	34.4	17.2
9			2i	82	32.8	16.4
10			2j	84	33.6	16.8
11			2k	73	29.2	14.6
12			2l	76	30.4	15.2

Table S3: Characterization of isolated 1,2-disubstituted benzimidazoles:

	**2a**[4]**:** Off white solid. R_f = 0.24 (20% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.80 (d, *J* = 8.0 Hz, 1H), 7.67–7.56 (m, 2H), 7.43 – 7.31 (m, 3H), 7.26-7.22 (m,, 4H), 7.16-7.12 (m, 2H), 7.05–6.98 (m, 2H), 5.37 (s, 2H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 154.2, 143.2, 136.4, 136.1, 130.1, 129.9, 129.3, 129.1, 128.8, 127.8, 126.0, 123.1, 122.7, 120.0, 110.5, 48.4.
	**2b**[4]: Off white solid. R_f = 0.2 (20% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.87 (d, *J* = 8.0 Hz, 1H), 7.58 (d, *J* = 8.1 Hz, 2H), 7.32-7.28 (m, 1H), 7.24 (d, *J* = 6.6 Hz, 2H), 7.23– .19 (m, 2H), 7.14 (d, *J* = 7.9 Hz, 2H), 7.00 (d, *J* = 8.0 Hz, 2H), 5.41 (s, 2H), 2.39 (s, 3H), 2.34 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 154.4, 142.9, 140.2, 137.5, 136.0, 133.4, 129.7, 129.5, 129.2, 126.8, 125.9, 123.0, 122.5, 119.7, 110.6, 48.2, 21.4, 21.1.
	**2c**[5]: White solid. R_f = 0.4 (20% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.87-7.86 (d, *J* = 8.0 Hz, 1H), 7.65 – 7.63 (m, 2H), 7.33-7.28 (m, 3H), 7.24 – 7.21 (m, 2H), 7.19-7.17 (d, *J* = 8.1 Hz, 2H), 7.04 (d, *J* = 8.2 Hz, 2H), 5.44 (s, 2H), 3.01-2.84 (m, 2H), 1.28-1.23 (m, 12H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 154.4, 150.9, 148.4, 143.2, 136.1, 133.8, 129.3, 127.5, 127.1, 126.9, 126.0, 122.8, 122.5, 119.8, 110.6, 48.2, 34.1, 33.8, 23.9, 23.8.
	**2d**[6]: Pale brown solid. R_f = 0.4 (20% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.88-7.86 (d, *J* = 8.0 Hz, 1H), 7.68-7.66 (d, *J* = 8.3 Hz, 2H), 7.48 (d, *J* = 8.3 Hz, 2H), 7.39-7.32 (m, 3H), 7.22 (d, *J* = 3.0 Hz, 2H), 7.06 (d, *J* = 8.1 Hz, 2H), 5.43 (s, 2H), 1.35 (s, 9H), 1.31 (s, 9H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 154.3, 153.6, 150.7, 143.2, 136.1, 133.4, 129.0, 127.7, 126.0, 125.8, 125.7, 122.8, 122.5, 119.8, 110.6, 48.2, 34.9, 34.6, 31.3, 31.2.
	2e[7]: Pale yellow solid. R_f = 0.25 (20% EtOAc/ Petroleum ether).^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.86 (d, *J* = 7.9 Hz, 1H), 7.59 (d, *J* = 8.4 Hz, 2H), 7.51 (d, *J* = 8.5 Hz, 2H), 7.46 (d, *J* = 8.3 Hz, 2H), 7.35 – 7.25 (m, 2H), 7.19 (d, *J* = 8.0

	Hz, 1H), 6.95 (d, *J* = 8.3 Hz, 2H), 5.37 (s, 2H).$^{13}C\{^{1}H\}$ (100 MHz, $CDCl_3$) δ = 152.9, 143.1, 135.9, 135.2, 132.4, 132.1, 130.67, 128.8, 127.6, 124.7, 123.5, 123.1, 121.9, 120.2, 110.3, 47.9.
	**2f**[8]: Pale yellow solid. R_f = 0.3 (20% EtOAc/ Petroleum ether).^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.87 (d, *J* = 8.0 Hz, 1H), 7.61-7.56 (m, 2H), 7.46-7.41 (m, 2H), 7.37-7.26 (m, 4H), 7.19 (d, *J* = 7.9 Hz, 1H), 7.01 (d, *J* = 8.4 Hz, 2H), 5.39 (s, 2H). $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ = 152.9, 143.1, 136.4, 135.9, 134.7, 133.9, 130.5, 129.4, 129.2, 128.4, 127.3, 123.51, 123.1, 120.2, 110.3, 47.8.
	**2g**[7]: Yellow solid. R_f = 0.2 (20% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 8.36-8.29 (m, 2H), 8.26-8.20 (m, 2H), 7.93 (d, *J* = 8.0 Hz, 1H), 7.87-7.81 (m, 2H), 7.44-7.31 (m, 2H), 7.27 (d, *J* = 8.5 Hz, 2H), 7.21 (d, *J* = 8.0 Hz, 1H), 5.58 (s, 2H). $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ = 151.4, 148.7, 147.9, 143.2, 142.8, 136.0, 135.8, 130.0, 126.7, 124.7, 124.6, 124.2, 123.8, 120.9, 110.2, 48.0.
	**2h**[7]: Off white solid. R_f = 0.3 (20% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.84 (d, *J* = 8.0 Hz, 1H), 7.63 (d, *J* = 8.7 Hz, 2H), 7.30-7.28 (m, 1H), 7.21 (d, *J* = 3.5 Hz, 2H), 7.03 (d, *J* = 8.6 Hz, 2H), 6.96 (d, *J* = 8.7 Hz, 2H), 6.85 (d, *J* = 8.7 Hz, 2H), 5.38 (s, 2H), 3.84 (s, *J* = 8.1 Hz, 3H), 3.78 (s, *J* = 7.0 Hz, 3H). $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ = 160.9, 159.1, 154.1, 143.1, 136.1, 130.7, 128.5, 127.2, 122.8, 122.6, 122.4, 119.7, 114.4, 114.2, 110.5, 55.4, 55.3, 47.0.
	2i[5]: White solid. R_f = 0.3 (20% EtOAc/ Petroleum ether).^{1}H NMR (400 MHz, DMSO-D_6): δ = 9.96 (s, 1H), 9.44 (s, 1H), 7.63 (dd, *J* = 7.3, 1.5 Hz, 1H), 7.55-7.47 (m, 2H), 7.42-7.37 (m, 1H), 7.21-7.14 (m, 2H), 6.89-6.85 (m, 2H), 6.81 (d, *J* = 8.6 Hz, 2H), 6.66- 6.60 (m, 2H), 5.39 (s, 2H). $^{13}C\{^{1}H\}$ NMR (100 MHz, DMSO-d_6): δ = 159.4, 157.2, 154.1, 143.2, 136.3, 131.1, 128.6, 128.0, 127.6, 122.6, 122.3, 121.2, 119.3, 116.0, 116., 111.4, 47.5.

	**2j**[6]: Brown solid. R_f = 0.2 (30% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.91 (d, *J* = 8.0 Hz, 1H), 7.66-764 (m, 2H), 7.32–7.28 (m, 3H), 7.24–7.20 (m, 2H), 7.18-7.16 (m, 2H), 7.05-7.03(m, 2H), 5.43 (s, 2H), 2.73-2.63 (m, 4H), 1.29-1.22 (m, 6H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 154.3, 146.4, 143.8, 143.0, 136.1, 133.7, 129.3, 128.5, 128.3, 127.2, 126.0, 122.9, 122.7, 119.8, 110.6, 48.3, 28.8, 28.5, 15.5, 15.4.
	**2k**[7]: White solid. R_f = 0.2 (30% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.85-7.72 (m, 1H), 7.63 (d, *J* = 1.1 Hz, 1H), 7.54-7.43 (m, 1H), 7.34-7.23 (m, 3H), 7.24-7.15 (m, 1H), 6.59 (m, 1H), 6.30-6.17 (m, 2H), 5.61 (s, 2H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 149.6, 145.4, 144.0, 143.9, 143.0, 142.7, 135.5, 123.3, 122.9, 119.8, 113.0, 112.1, 110.5, 110.0, 108.4, 41.7.
	**2l**[7]: Brown solid**.** R_f = 0.2 (30% EtOAc/ Petroleum ether). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.85-7.82 (m, 1H), 7.52 (dd, *J* = 5.1, 1.0 Hz, 1H), 7.47 (dd, *J* = 3.7, 1.0 Hz, 1H), 7.39-7.36 (m, 1H), 7.34-7.28 (m, 2H), 7.25 (dt, *J* = 5.1, 1.9 Hz, 1H), 7.14 (dd, *J* = 5.1, 3.7 Hz, 1H), 6.95 (dd, *J* = 5.1, 3.5 Hz, 1H), 6.89- 6.84 (m, 1H), 5.71 (s, 2H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ = 147.6, 143.0, 138.9, 135.9, 131.9, 129.0, 128.0, 128.0, 127.3, 125.5, 125.5, 123.4, 123.0, 120.0, 110.0, 44.1.

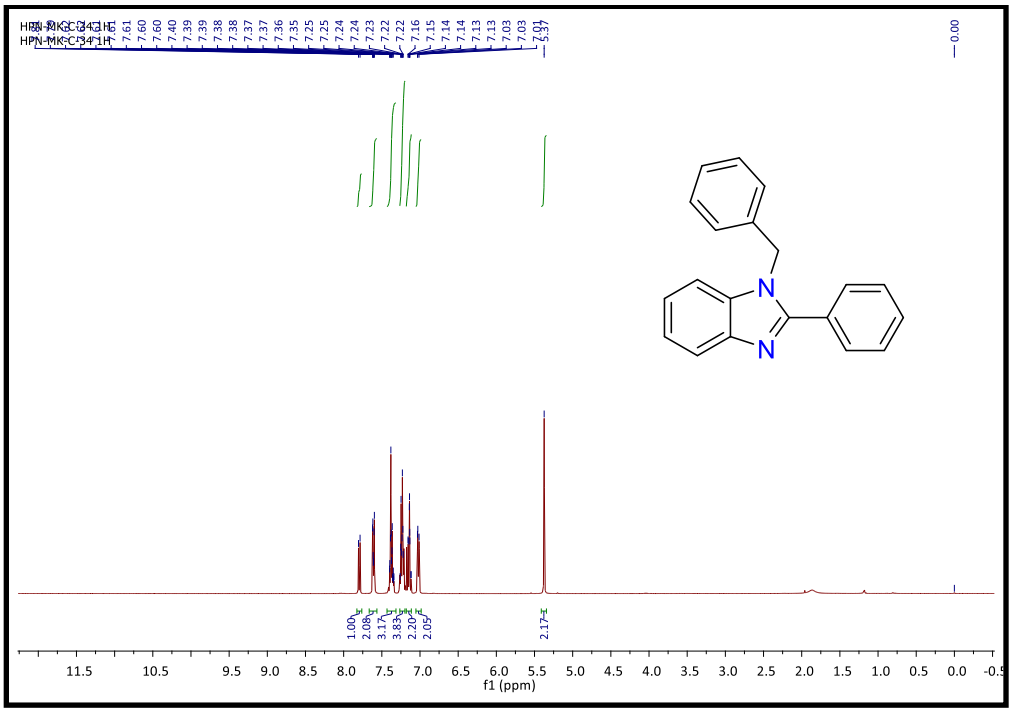


Figure S42: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2a**.

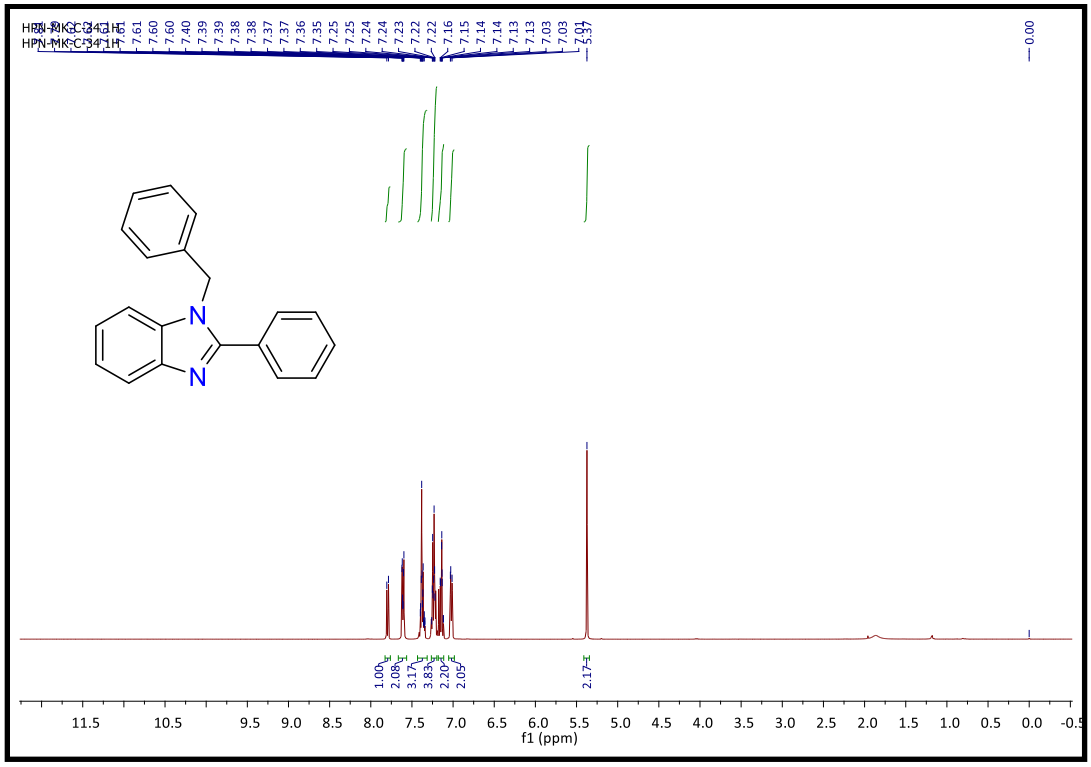


Figure S43: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2a**.

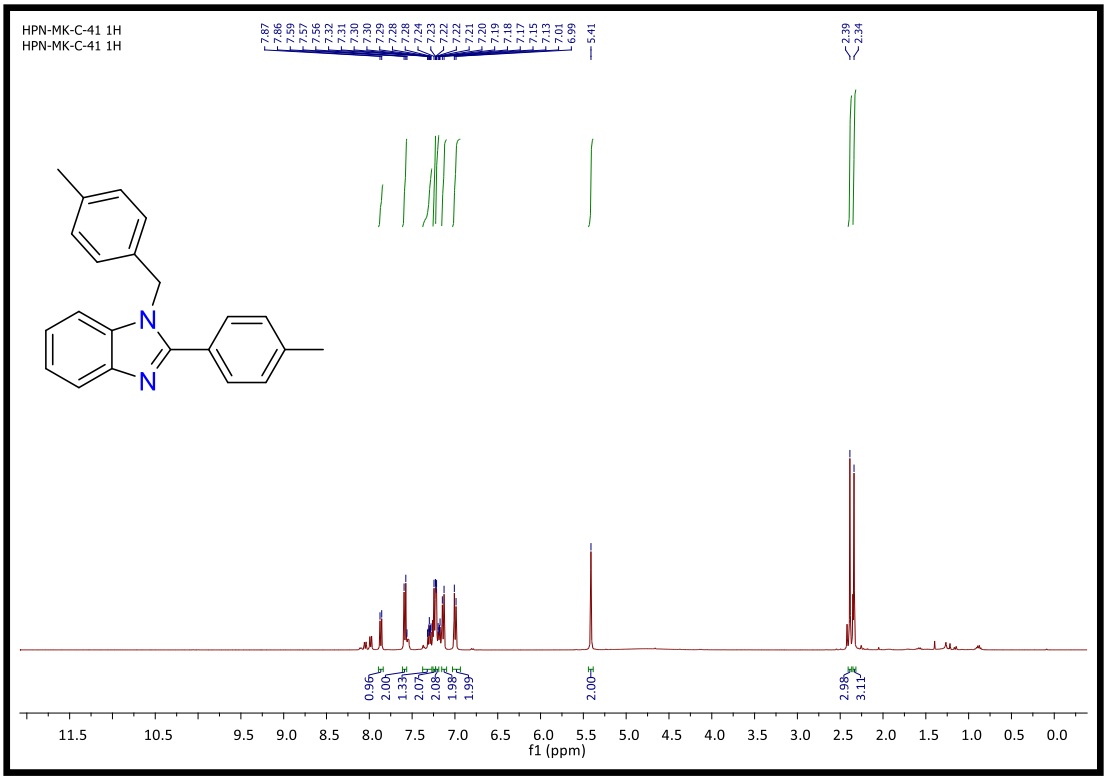


Figure 44: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2b**.

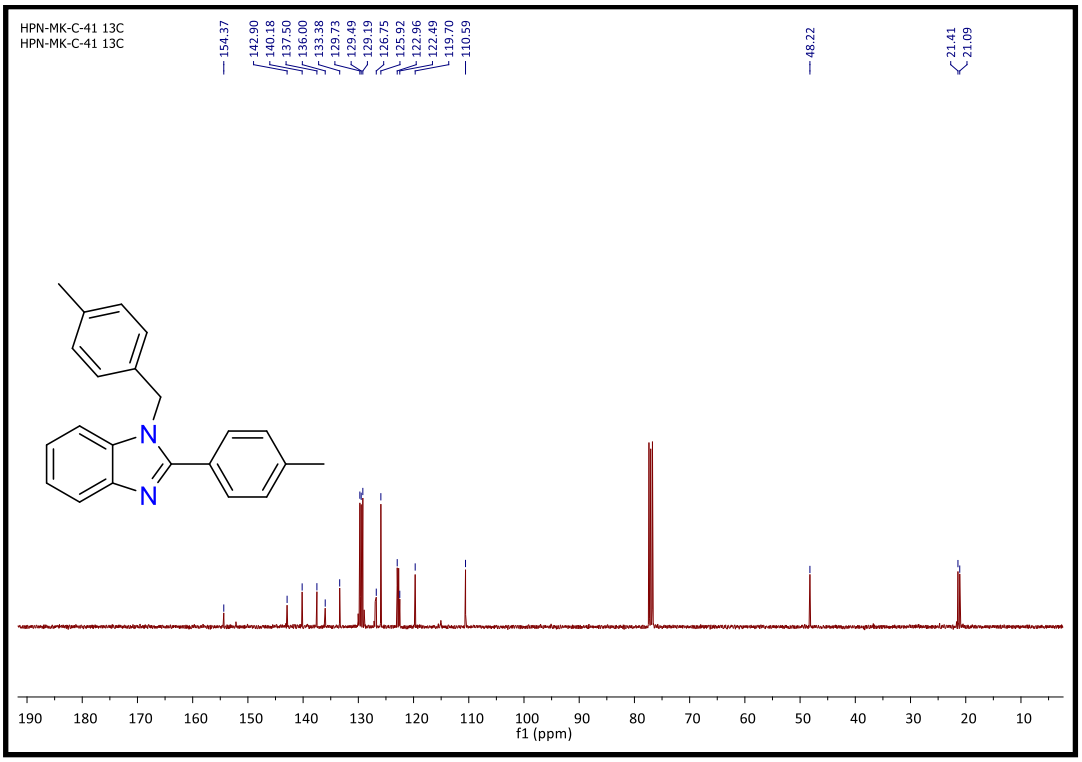


Figure 45: ^{13}C NMR (100 MHz, $CDCl_3$) spectrum of **2b**.

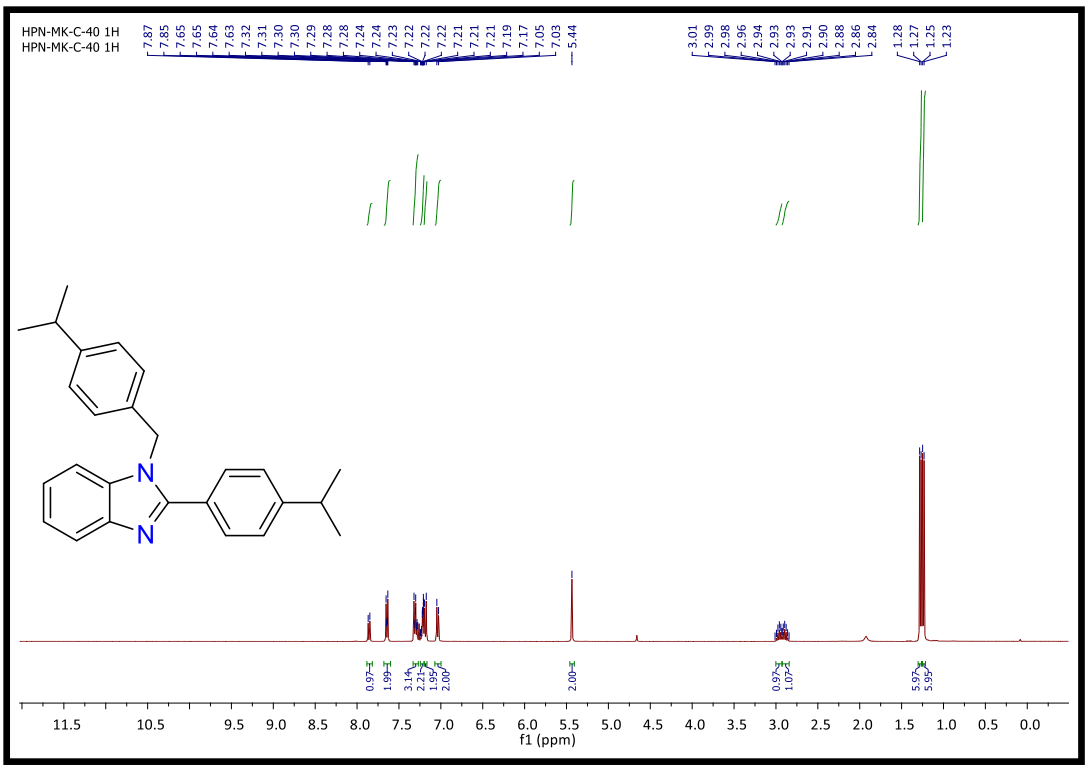


Figure S46: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2c**.

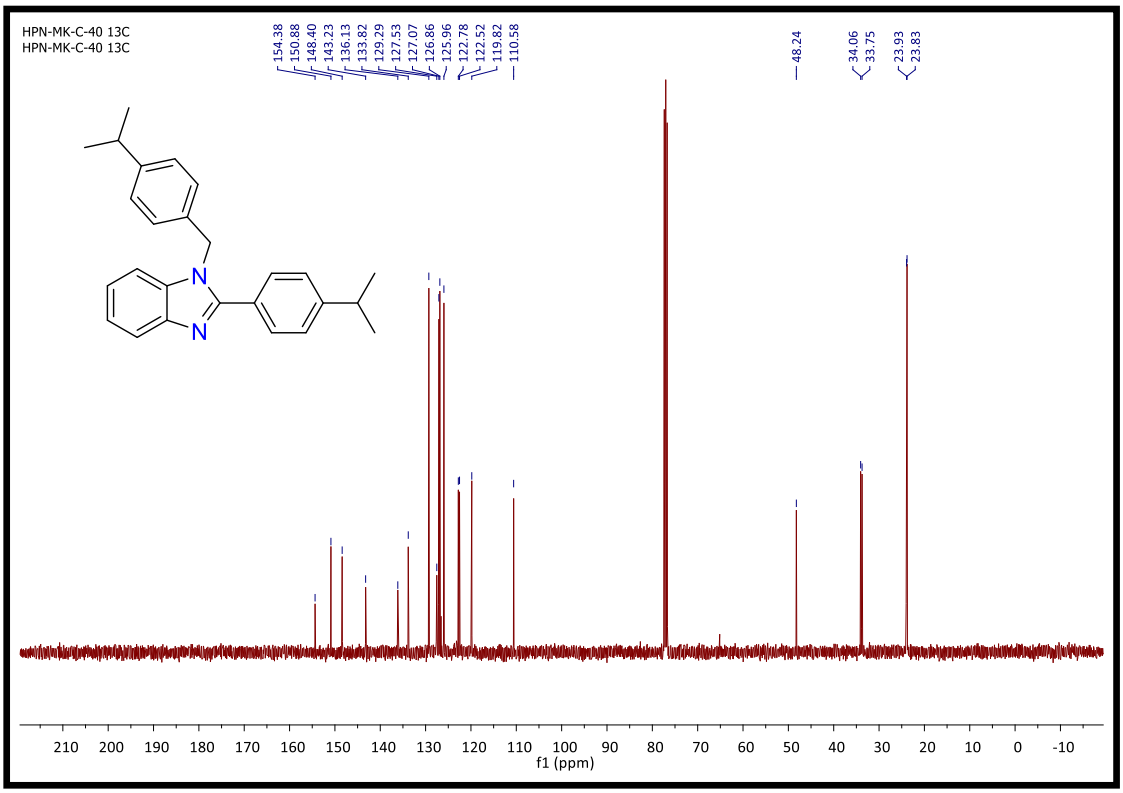


Figure S47: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2c**.

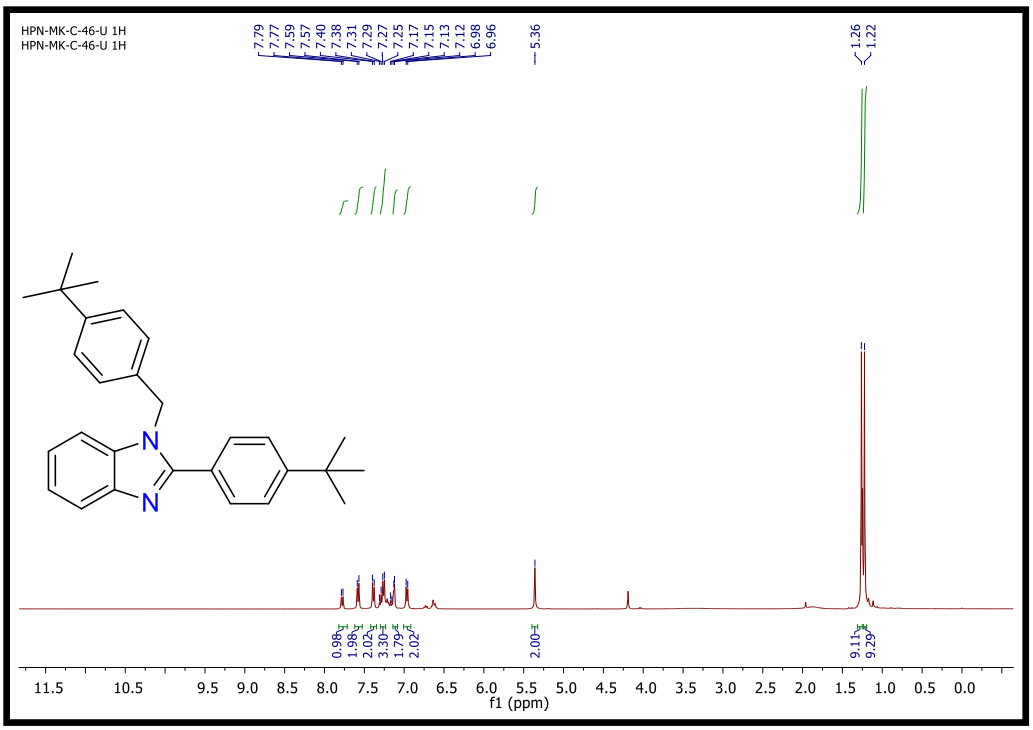


Figure S48: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2d**.

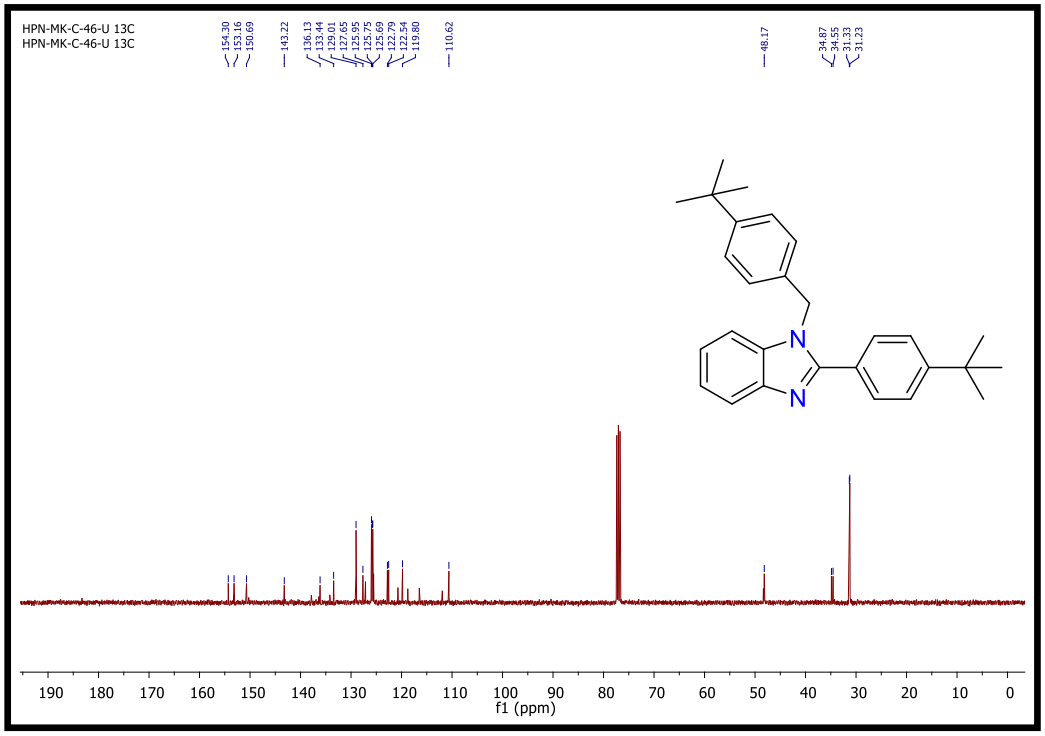


Figure S49: ^{13}C {^{1}H} NMR (100 MHz, $CDCl_3$) spectrum of **2d**.

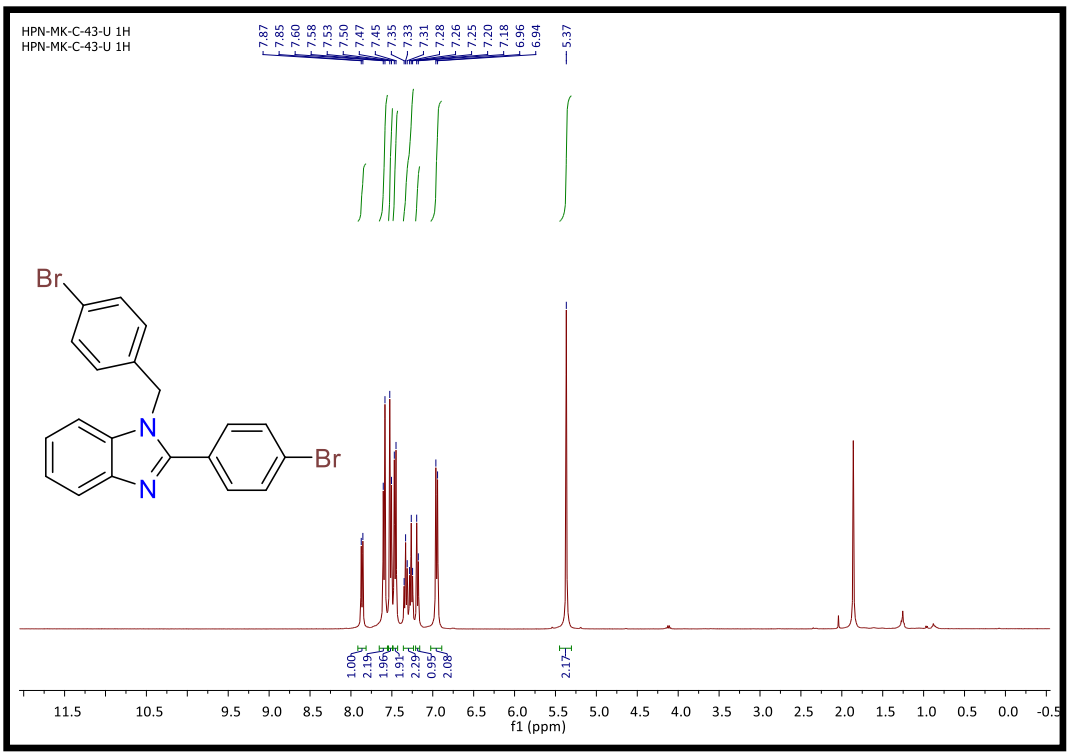


Figure S50: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2e**.

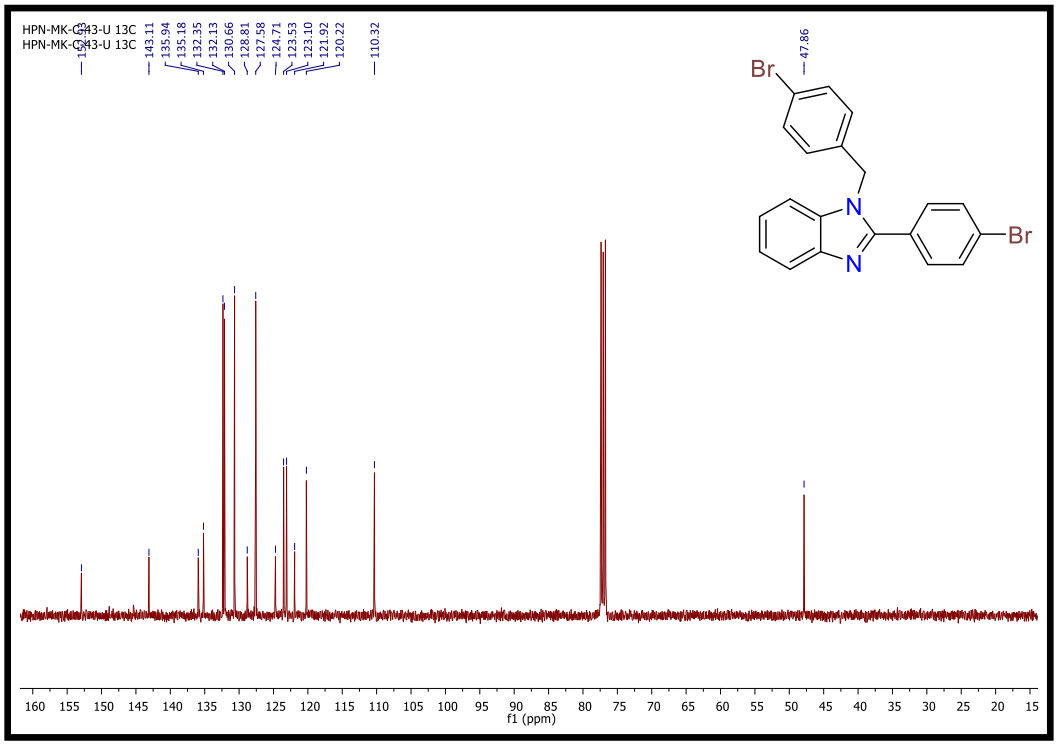


Figure 51: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2e**.

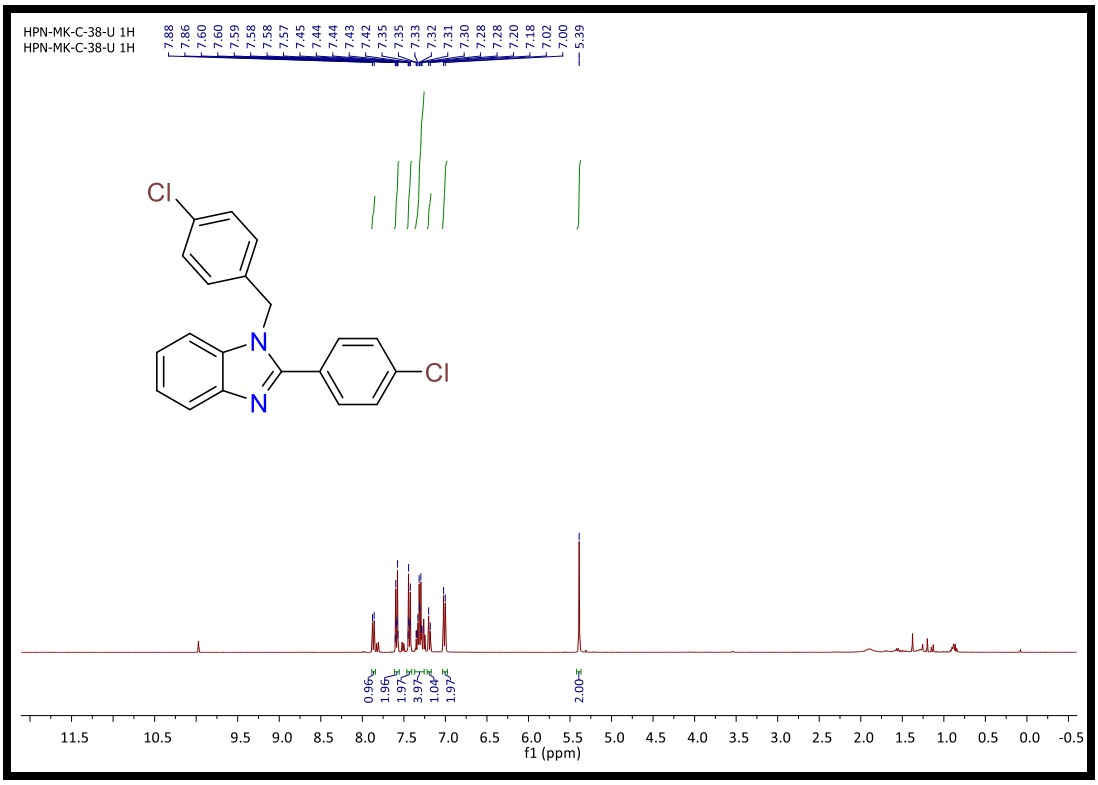


Figure S52: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2f**.

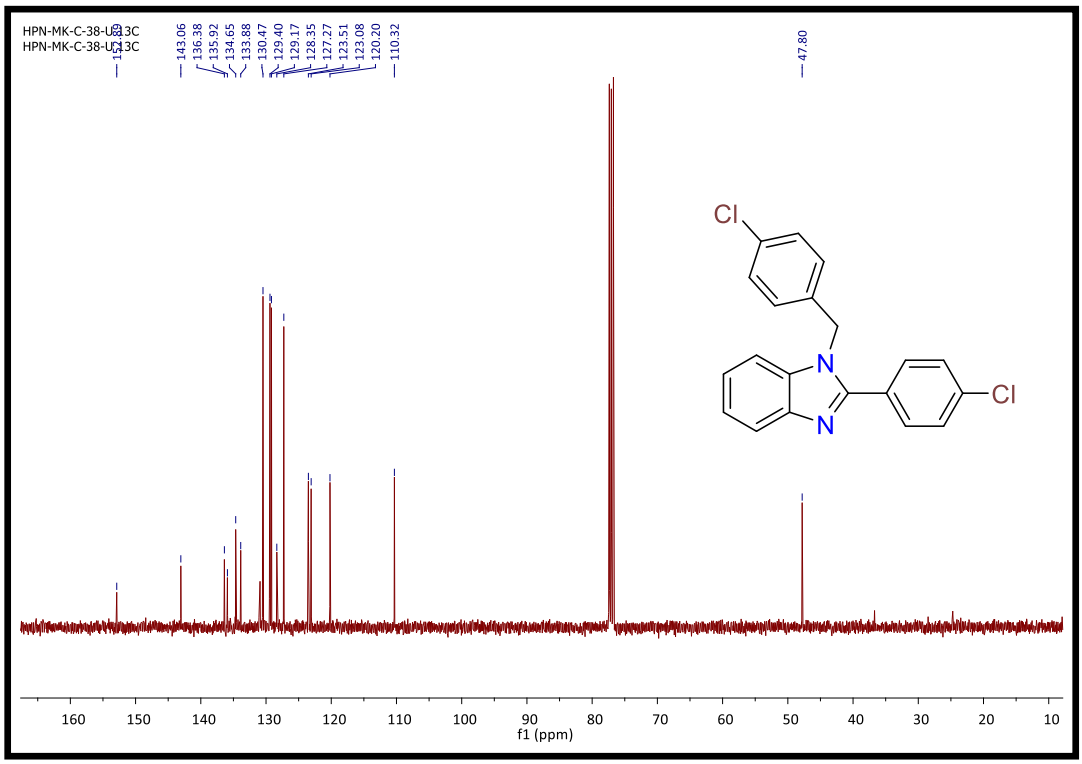


Figure S53: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2f** .

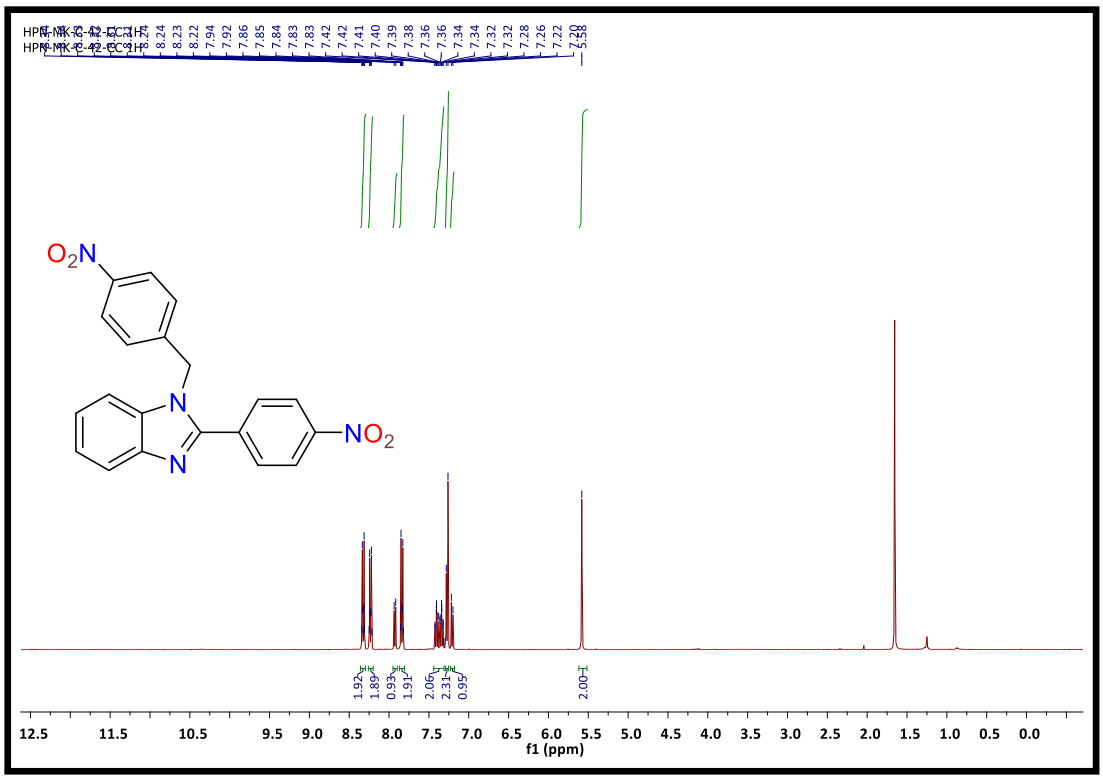


Figure S54: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2g**.

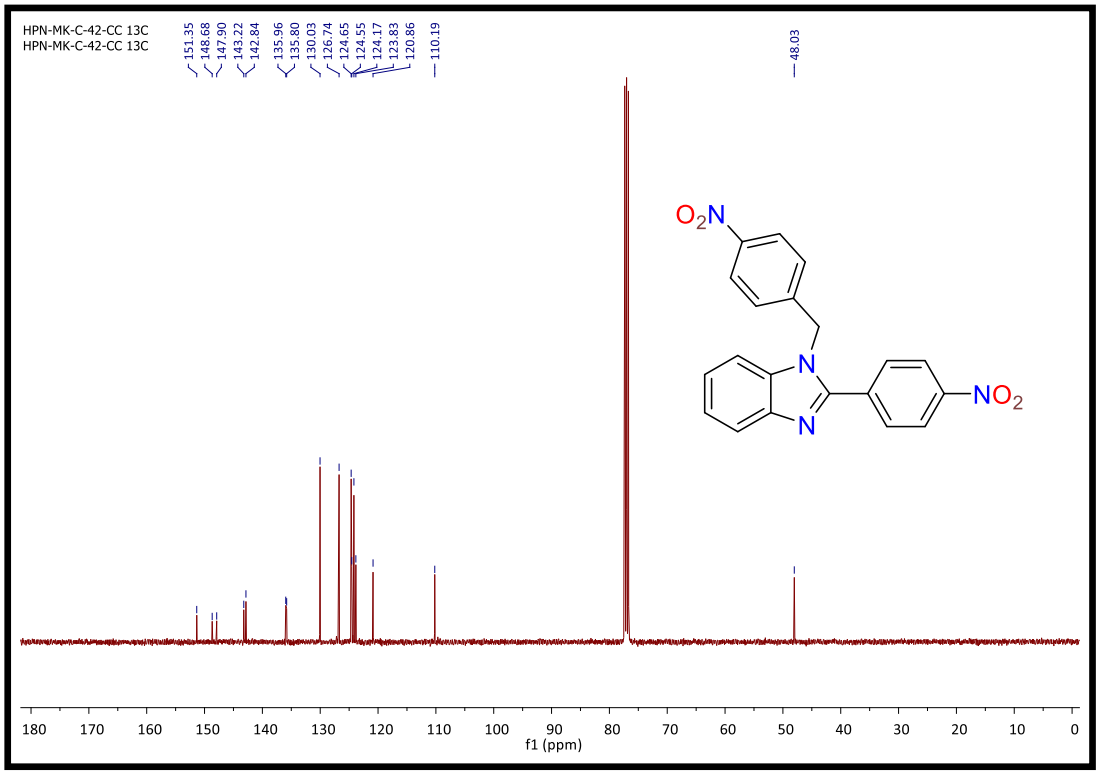


Figure S55: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2g**.

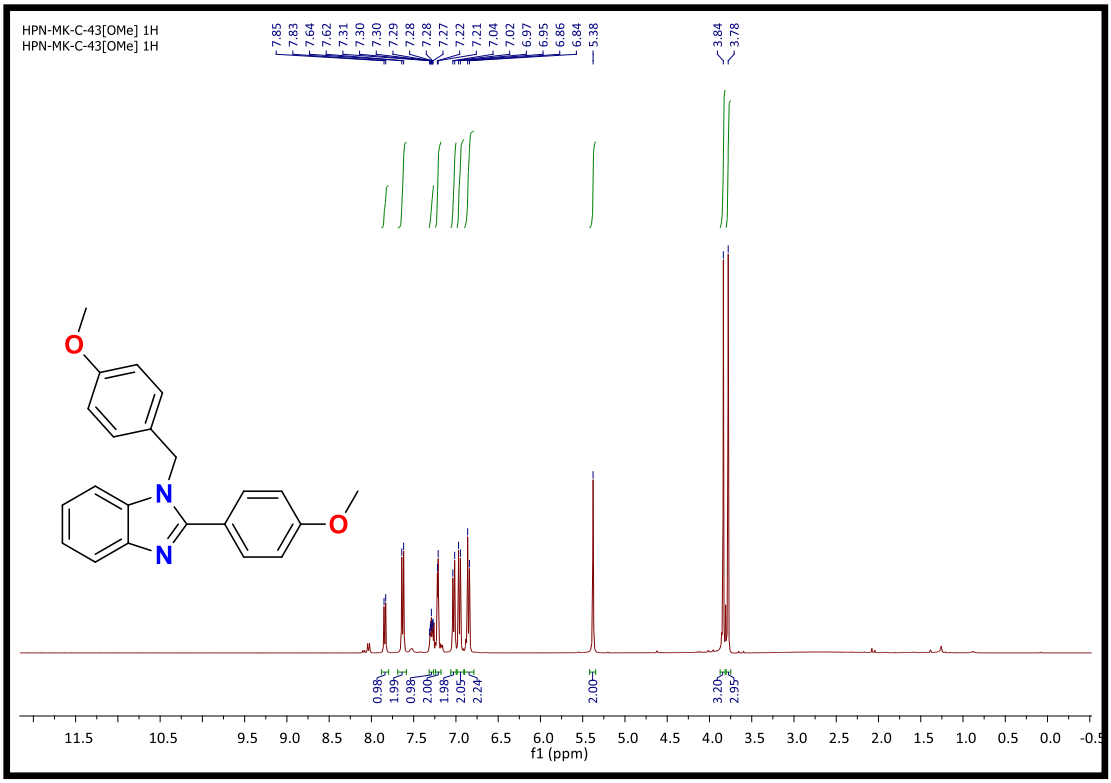


Figure 56: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2h**.

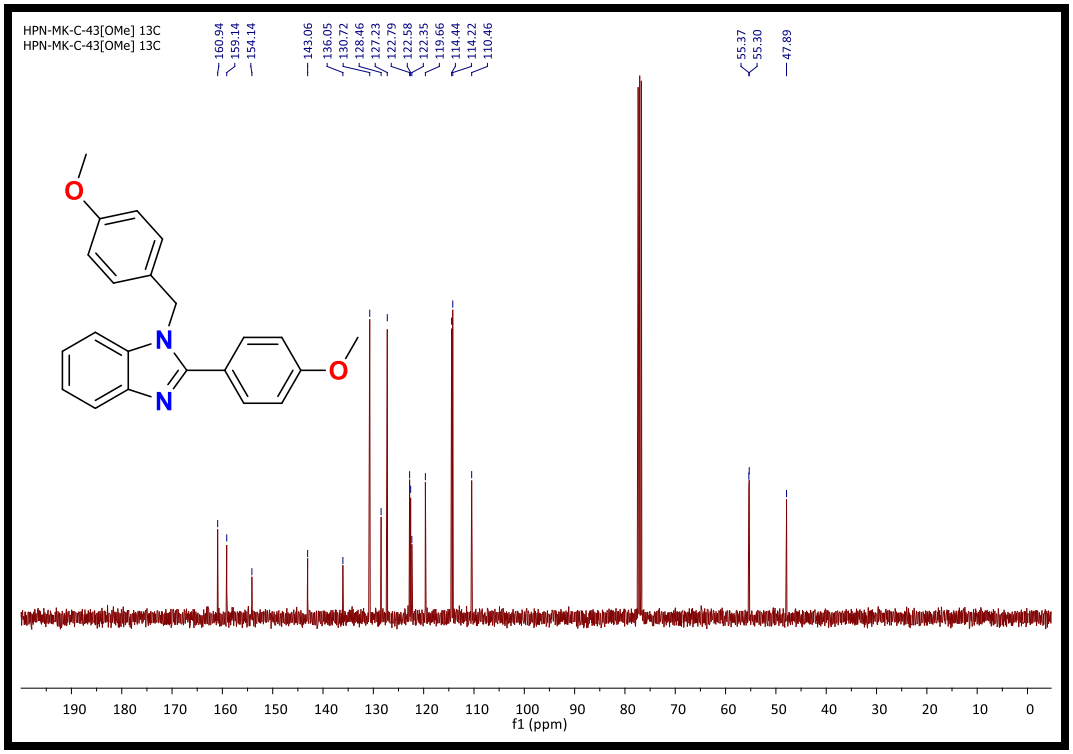


Figure 57: ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) spectrum of **2h**.

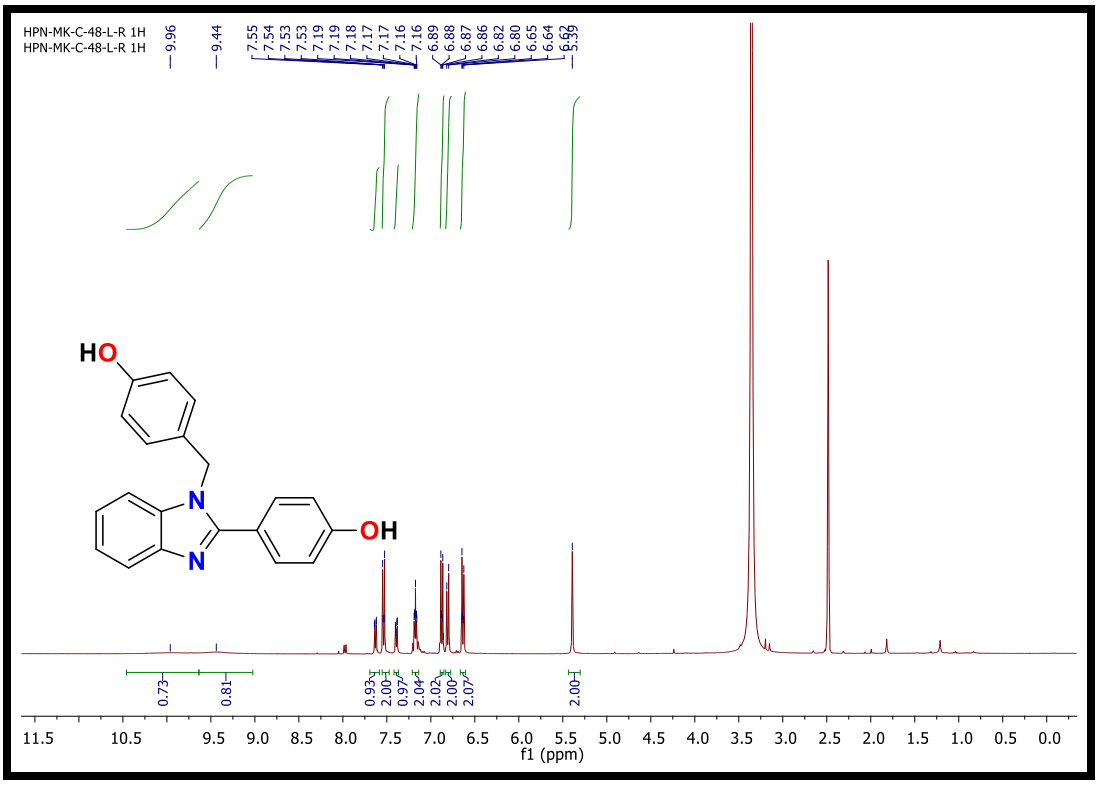


Figure 58: ^{1}H NMR (400 MHz, DMSO-d_6) spectrum of **2i**.

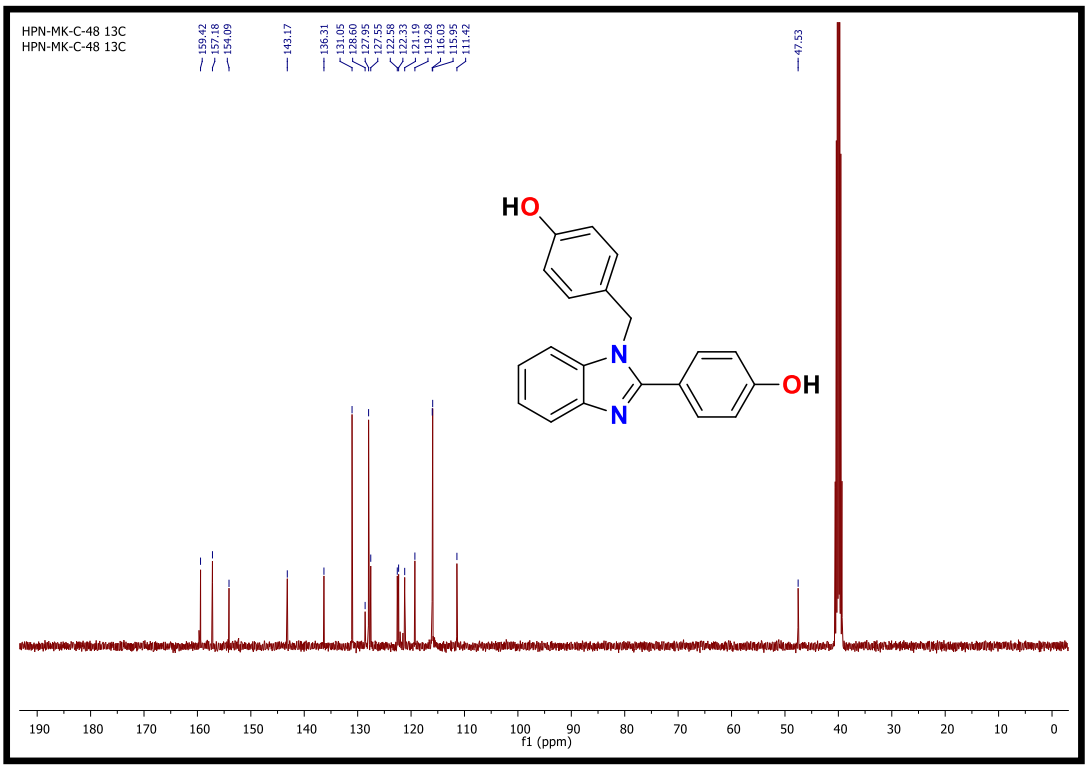


Figure 59: ^{13}C{^{1}H} NMR (100 MHz, DMSO-d_6) spectrum of **2i**.

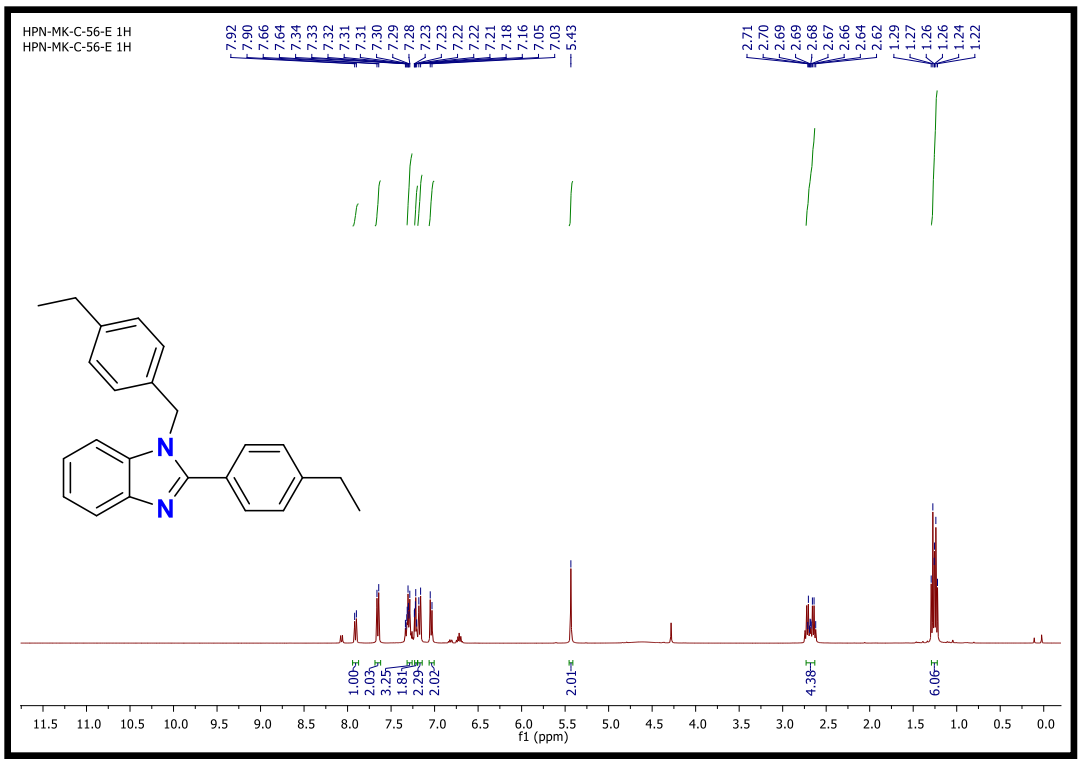


Figure S60: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2j**.

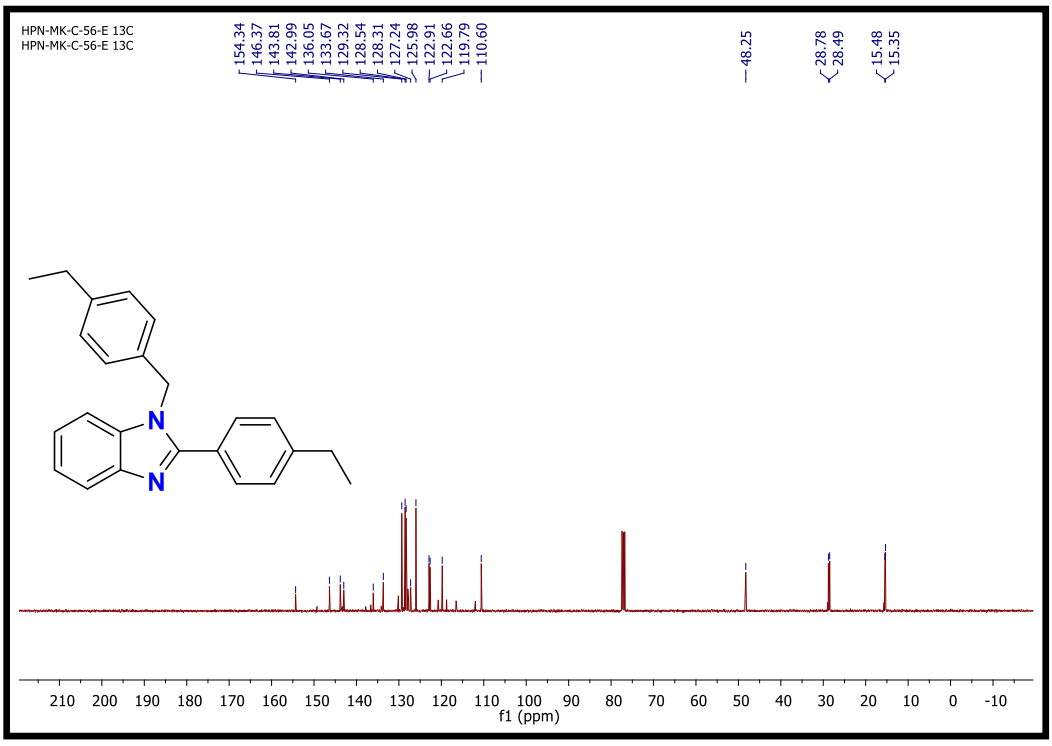


Figure S61: ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$) spectrum of **2j**.

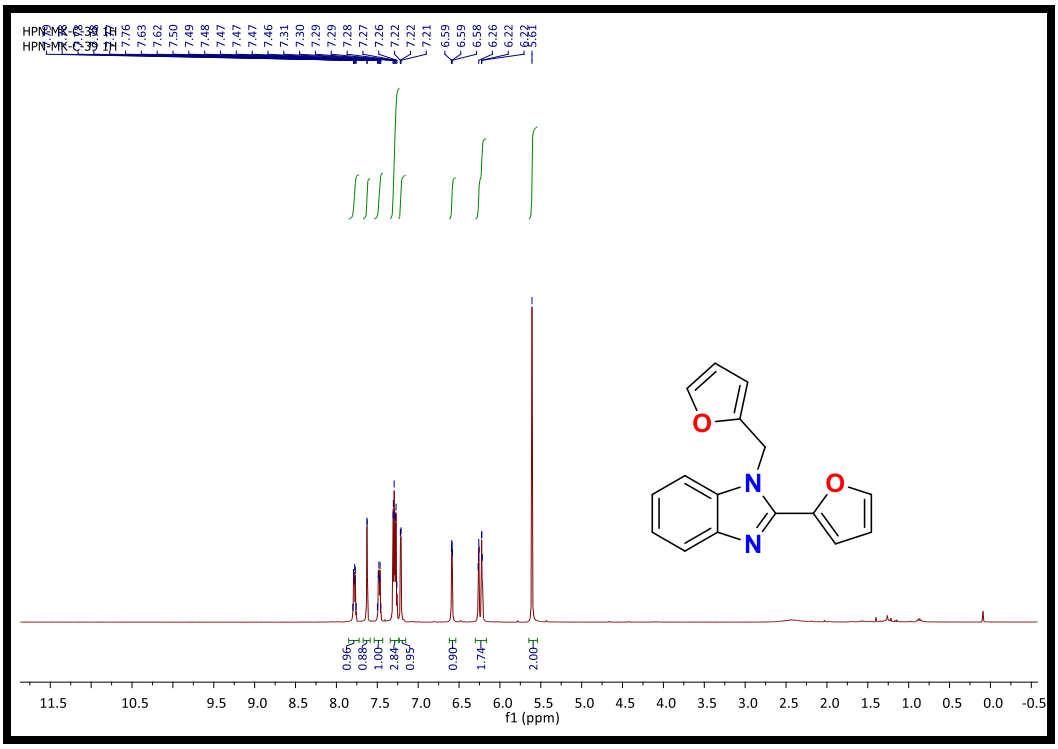


Figure S62: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2k**.

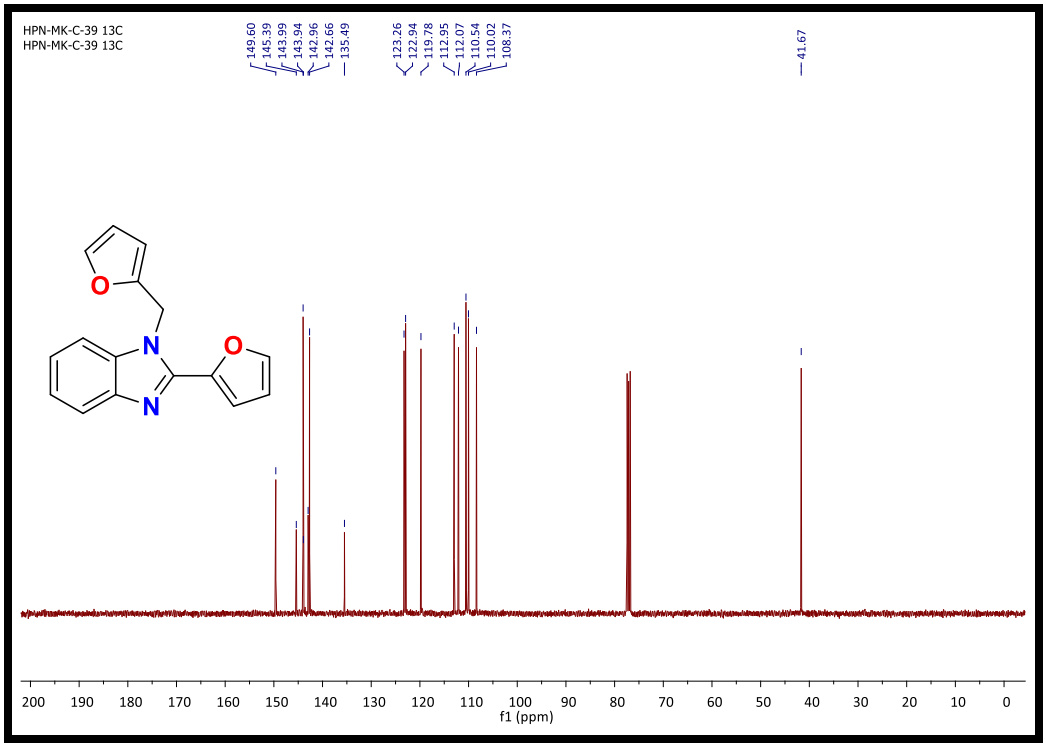


Figure S63: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2K**.

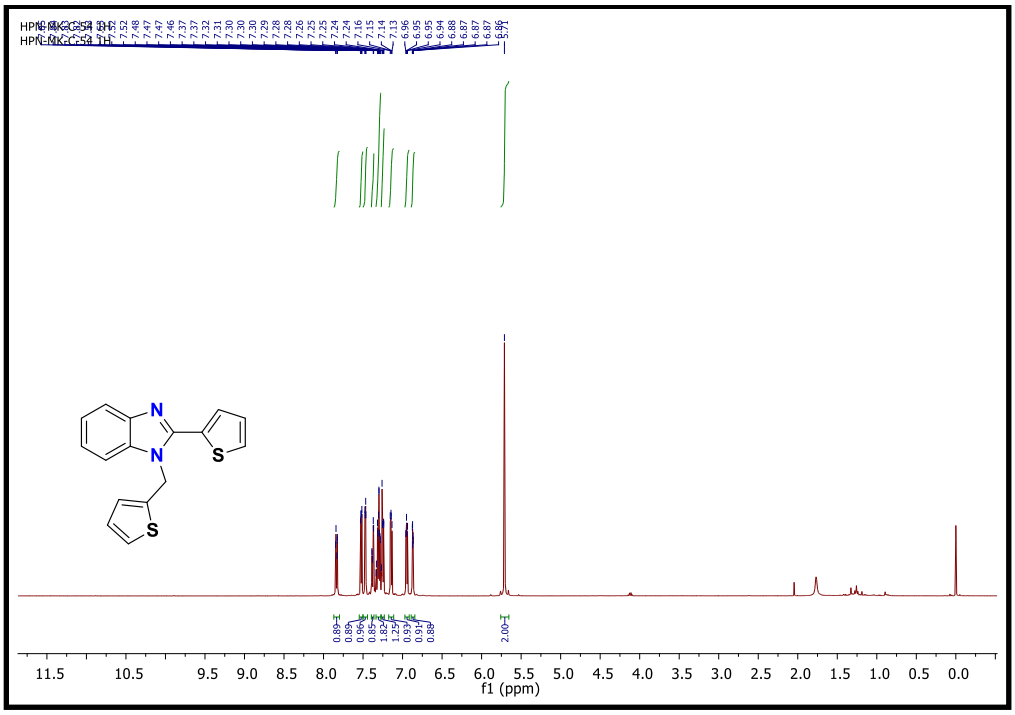


Figure S64: ^{1}H NMR (400 MHz, $CDCl_3$) spectrum of **2l**.

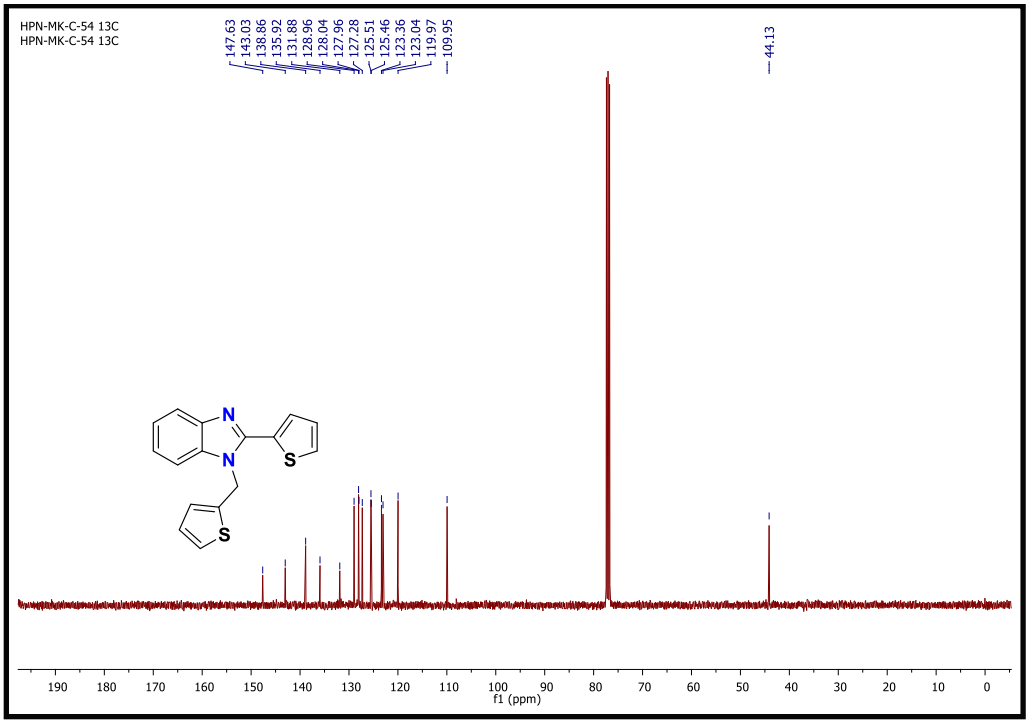


Figure S65: $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$) spectrum of **2l**.

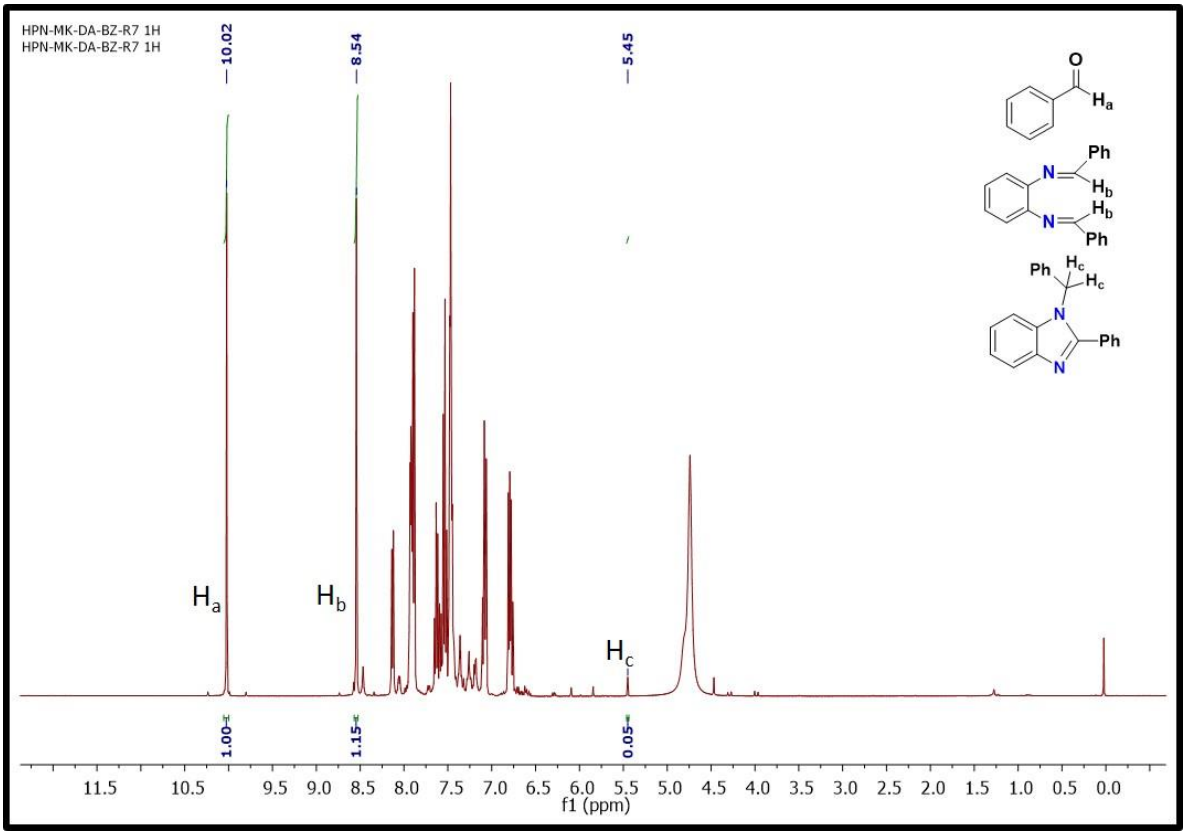


Figure S66: ^{1}H NMR spectrum of the reaction mixture obtained by reacting *o*-phenylenediamine and benzaldehyde in the absence of the catalyst at 60 °C in $CDCl_3$. The spectrum was recorded within 30 minutes of adding the reactants.

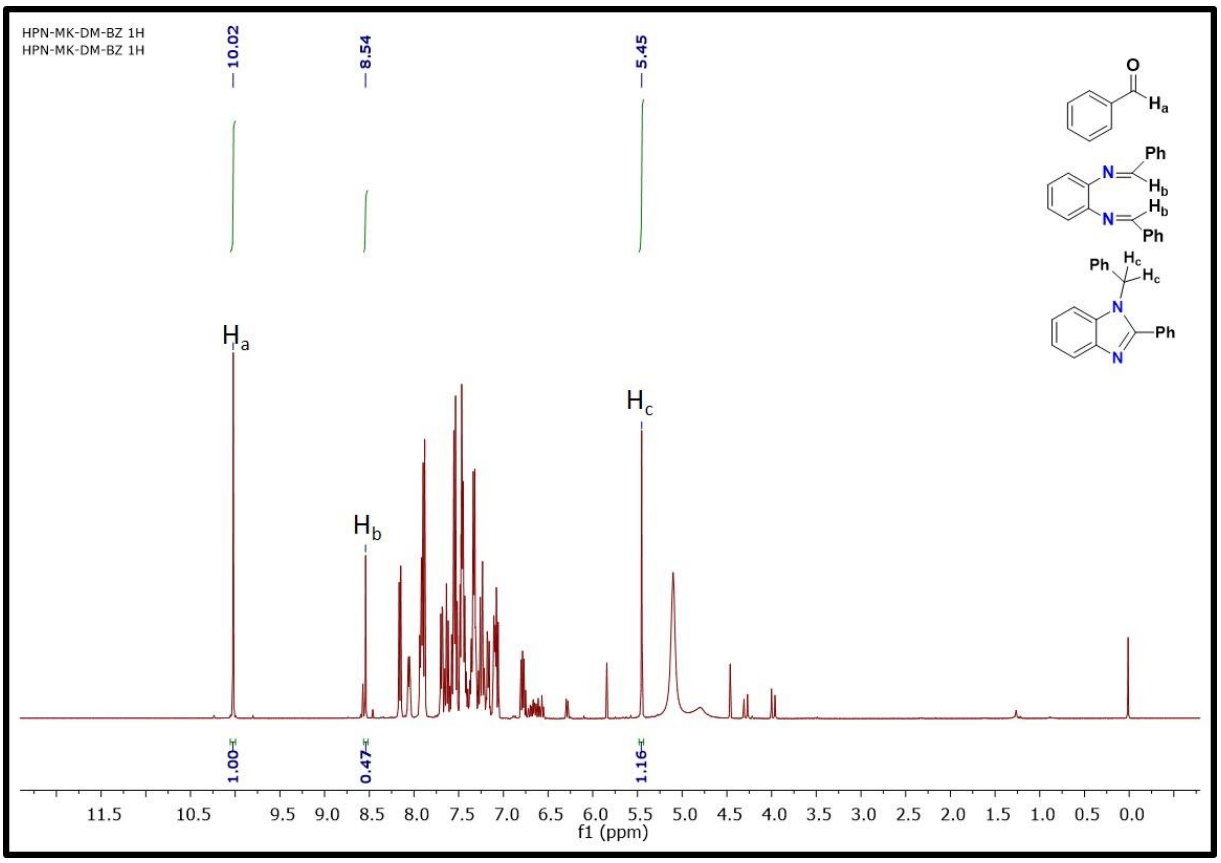


Figure S67: ^{1}H NMR spectrum of the reaction mixture obtained by reacting *o*-phenylenediamine and benzaldehyde in presence of the pro ligand **H_3L** at 60 °C in $CDCl_3$. The spectrum was recorded within 30 minutes of adding the reactants.

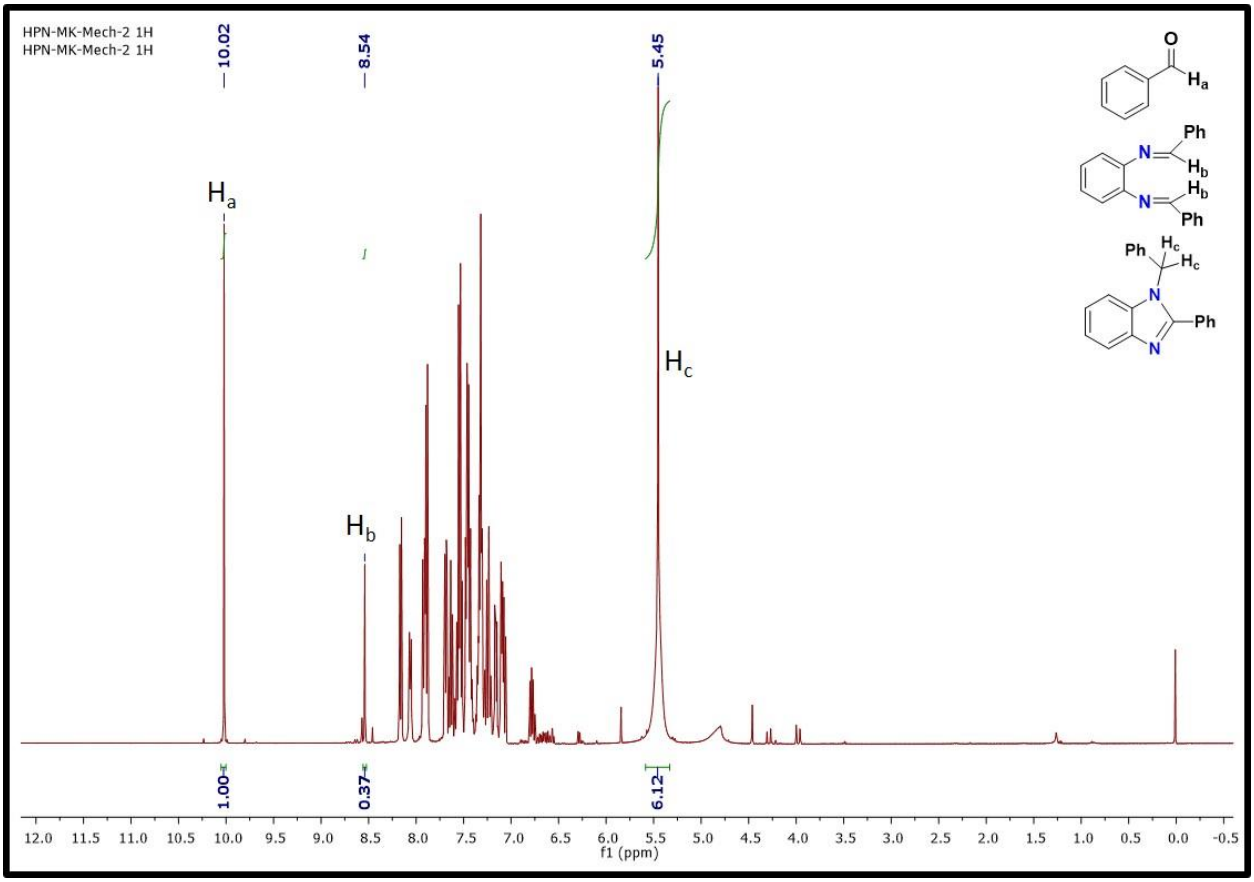


Figure S68: ^{1}H NMR spectrum of the reaction mixture obtained by reacting *o*-phenylenediamine and benzaldehyde in presence of compound **1** at 60 °C in $CDCl_3$. The spectrum was recorded within 30 minutes of adding the reactants.

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