Supporting Information
for

Electrochemically Initiated Intermolecular C-N Formation /Cyclization of Ketones with 2-Aminopyridines: an Efficient Method for the Synthesis of Imidazo[1,2-a]pyridines

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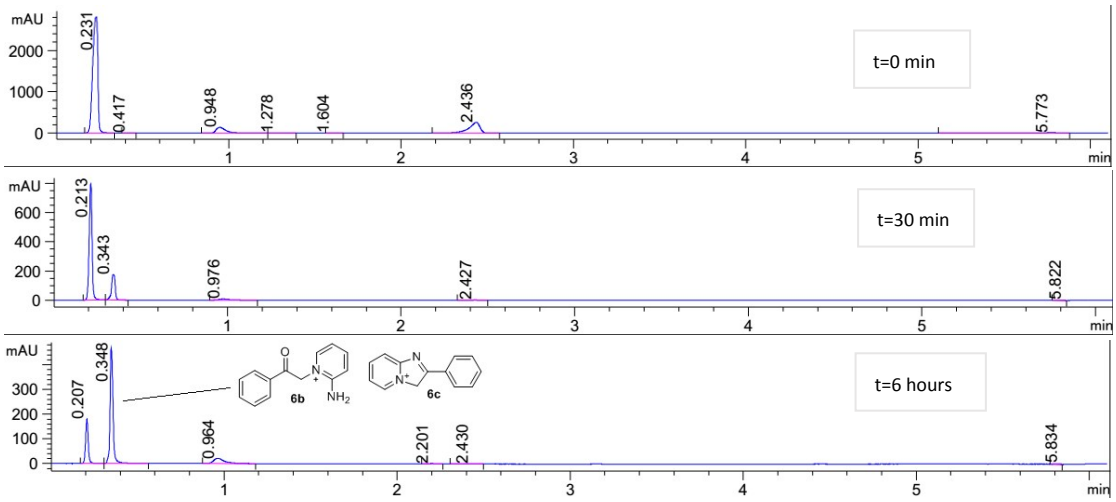
1. General

Reagents and solvents were purchased and used as received unless otherwise noted. All reactions were carried out in a sealed Schlenk tubes and monitored by TLC. Flash column chromatography was performed with silica gel (300–400 mesh). NMR spectra were recorded on Bruker AV-400 and Bruker AV-500 instruments. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker Avance-III 400 instrument (400 MHz for ^{1}H and 100 MHz for ^{13}C NMR spectroscopy). Data were reported as chemical shifts in ppm relative to TMS (0.00 ppm) for ^{1}H and $CDCl_3$ (77.16 ppm) or d_6-DMSO (39.52 ppm) for ^{13}C. Mass spectra were recorded on a Thermo Scientific LCQ Fleet spectrometer with ESI mode. Liquid chromatograph-mass spectrometer analysis was carried out on an Agilent 6120 Quadrupole LC/MS instrument.

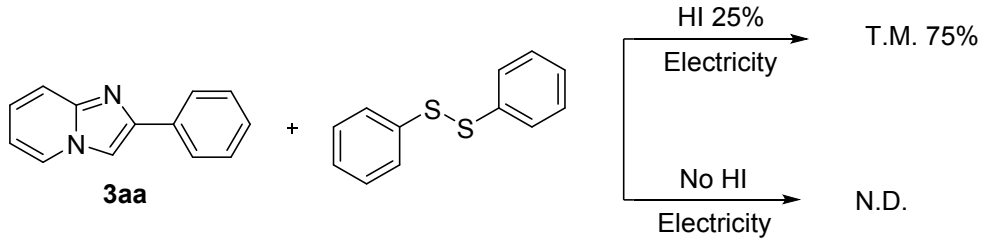
2. General Procedure for the Electrolysis

Ketone (0.8 mmol, 1 equiv), 2-aminopyridine (1.2 mmol, 1.5 equiv), hydriodic acid (25 mol %), tetrabutylammonium tetrafluoroborate (0.1M) and ethanol (3 mL) were placed in a 15 mL tube equipped with a carbon felt anode (1.5 cm^2) and a platinum cathode (1.5 cm^2) (Figure S1). The electrolysis was carried out at 50 ^{o}C under a constant cell potential of 2.0 V for 24 hours. Then saturated sodium thiosulfate solution (30 mL) and ethyl acetate (30 ml) were added. The phases were separated and the aqueous phase was extracted with ethyl acetate (2 x 30 mL). The combined organic solution was dried over anhydrous Na_2SO_4 and concentrated under reduced pressure. The residue was chromatographed through silica gel eluting with ethyl acetate/hexanes to give the product. The gram-scale reaction was conducted using the same electrodes but in a 50 mL two-necked round-bottom flask.

3. Monitoring the reaction using HPLC-MASS



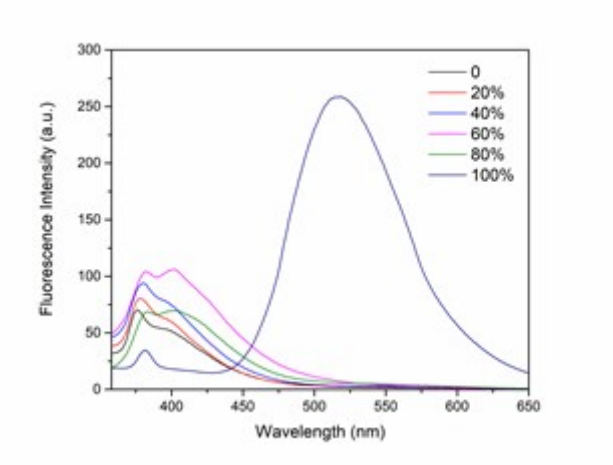
4. The control experiment of three component reaction



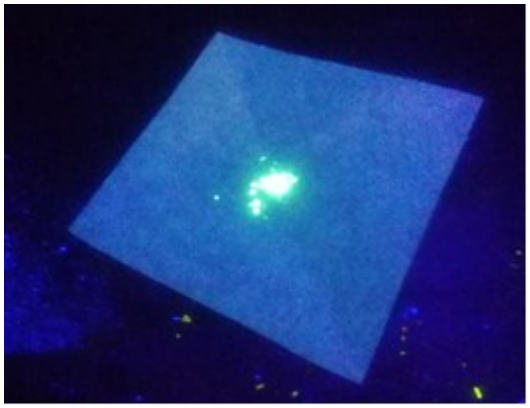
The reaction proceeded under standard condition.

5. The AIE properties and solid-state fluorescence of 5b

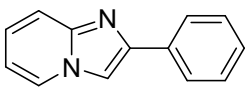
With the increasing of water, the emission of **5b** becomes stronger, showing **5b** has AIE properties.



Solid-state fluorescence of **5b**



Characterization Data for the products



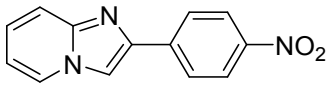
(**3aa**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.03 (d, J = 6.7 Hz, 1H), 7.96–7.92 (m, 2H), 7.79 (s, 1H), 7.60 (d, J = 9.1 Hz, 1H), 7.42 (t, J = 7.6 Hz, 2H), 7.35-7.29 (m, 1H), 7.14–7.09 (m, 1H), 6.71 (t, J = 6.7 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 145.9, 145.8, 133.9, 128.8, 128.1, 126.2, 125.7, 124.8, 117.7, 112.5,

108.2

HRMS m/z (ESI) : calcd. for $[C_{13}H_{10}N_2+H]^+$:195.0917 Found : 195.0922

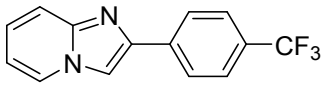


(**3ba**)

^{1}H NMR (400 MHz, dmso-d_6) δ 8.64 (s, 1H), 8.59-8.54 (m, 1H), 8.33-8.27 (m, 2H), 8.25-8.20 (m, 2H), 7.62 (d, J = 9.1 Hz, 1H), 7.34-7.28 (m, 1H), 6.95 (td, J = 6.8, 1.0 Hz, 1H)

^{13}C NMR (101 MHz, dmso-d_6) δ 146.5, 145.3, 142.0, 140.5, 127.3, 126.3, 126.0, 124.2, 117.0, 112.9, 111.7

HRMS m/z (ESI) : calcd. for $[C_{13}H_9N_3O_2+H]^+$:240.0768 Found : 240.0769

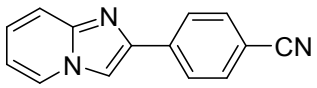


(**3ca**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.10 (dt, J = 6.8, 1.0 Hz, 1H), 8.04 (d, J = 8.1 Hz, 2H), 7.89 (s, 1H), 7.69–7.60 (m, 3H), 7.22-7.16 (m, 1H), 6.79 (td, J = 6.8, 1.0 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 145.9, 144.3, 137.3, 129.8(q, J = 32), 126.2, 125.84, 125.81, 125.77, 125.73, 125.4, 123.0, 117.8, 113.0, 109.1

HRMS m/z (ESI) : calcd. for $[C_{14}H_9F_3N_2+H]^+$:263.0791 Found : 263.0795

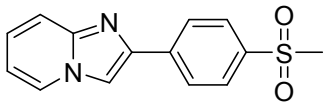


(**3da**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.11 (d, J = 6.8 Hz, 1H), 8.4-7.99 (m, 2H), 7.91 (s, 1H), 7.70-7.64 (m, 2H), 7.61 (d, J = 9.1 Hz, 1H), 7.23– 7.17 (m, 1H), 6.80 (t, J = 6.7 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 146.0, 143.7, 138.3, 132.6, 126.4, 125.9, 125.6, 119.1, 117.9, 113.1, 111.1, 109.6

HRMS m/z (ESI) : calcd. for $[C_{14}H_9N_3+H]^+$:220.0869 Found : 220.0877

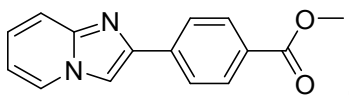


(**3ea**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.20-8.10 (m, 3H), 8.03-7.94 (m, 3H), 7.63 (d, J = 8.6 Hz, 1H), 7.26–7.16 (m, 1H), 6.88-6.78 (m, 1H), 3.09 (s, 3H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 146.0, 143.5, 139.3, 139.2, 127.9, 126.6, 126.0, 125.6, 117.8, 113.1, 109.8, 44.7

HRMS m/z (ESI) : calcd. for $[C_{14}H_{12}N_2O_2S+H]^+$:273.0692 Found : 273.0690

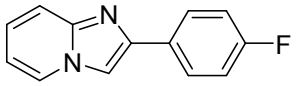


(**3fa**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12-8.07 (m, 3H), 8.02–7.99 (m, 2H), 7.91 (s, 1H), 7.63 (d, J = 9.1 Hz, 1H), 7.22-7.13 (m, 1H), 6.78 (td, J = 6.8, 0.8 Hz, 1H), 3.92 (s, 3H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 167.1, 146.0, 144.6, 138.3, 130.2, 129.4, 125.9, 125.8, 125.3, 117.8, 112.9, 109.4, 52.2

HRMS m/z (ESI) : calcd. for $[C_{15}H_{12}N_2O_2+H]^+$:253.0972 Found : 253.0971

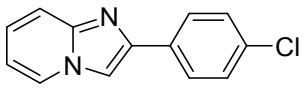


(**3ga**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.01 (d, J = 6.8 Hz, 1H), 7.91–7.84 (m, 2H), 7.71 (s, 1H), 7.57 (d, J = 9.0 Hz, 1H), 7.14–7.04 (m, 3H), 6.70 (td, J = 6.8, 0.9 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 163.9, 161.4, 145.3 (J = 82 Hz), 130.0 (J = 3.2 Hz), 127.7 (J = 8.1 Hz), 125.6, 124.8, 117.39, 115.6 (J = 22 Hz), 112.48, 107.85

HRMS m/z (ESI) : calcd. for $[C_{13}H_9FN_2+H]^+$:213.0823 Found : 213.0835

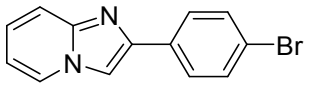


(**3ha**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.04 (d, J = 6.7 Hz, 1H), 7.84 (d, J = 8 Hz, 2H), 7.77 (s, 1H), 7.58 (d, J = 9.0 Hz, 1H), 7.36 (d, J = 8.5 Hz, 2H), 7.18–7.10 (m, 1H), 6.73 (t, J = 6.5 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 145.8, 144.7, 133.7, 132.4, 129.0, 127.3, 125.7, 125.0, 117.6, 112.7, 108.3

HRMS m/z (ESI) : calcd. for $[C_{13}H_9ClN_2+H]^+$:229.0527 Found : 229.0518

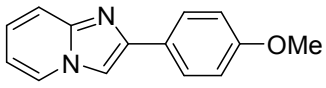


(**3ia**)

^{1}H NMR (400 MHz, dmso-d_6) δ 8.52 (d, J = 6.8 Hz, 1H), 8.43 (s, 1H), 7.94-7.90 (m, 2H), 7.64-7.60 (m, 2H), 7.57 (d, J = 9.0 Hz, 1H), 7.28–7.23 (m, 1H), 6.90 (td, J = 6.7, 0.8 Hz, 1H)

^{13}C NMR (101 MHz, dmso-d_6) δ 144.9, 143.2, 133.2, 131.7, 127.6, 127.0, 125.2, 120.7, 116.7, 112.5, 109.5

HRMS m/z (ESI) : calcd. for $[C_{13}H_9FN_2+H]^+$:273.0022 Found : 273.0018

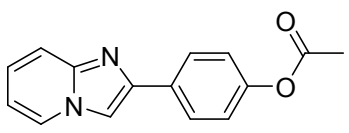


(**3ja**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07 (d, J = 6.7 Hz, 1H), 7.91–7.84 (m, 2H), 7.75 (s, 1H), 7.60 (d, J = 9.1 Hz, 1H), 7.17–7.10 (m, 1H), 7.00-6.92 (m, 2H), 6.77-6.71 (m, 1H), 3.84 (s, 2H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 159.6, 145.6, 145.5, 127.3, 126.5, 125.5, 124.5, 117.2, 114.1, 112.2, 107.3, 55.3

HRMS m/z (ESI) : calcd. for $[C_{14}H_{12}N_2O+H]^+$:225.1022 Found : 225.1015

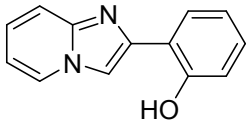


(**3ka**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08 (d, J = 6.8 Hz, 1H), 7.98–7.91 (m, 2H), 7.80 (s, 1H), 7.61 (d, J = 9.1 Hz, 1H), 7.18–7.09 (m, 3H), 6.75 (td, J = 6.8, 0.9 Hz, 1H), 2.31 (s, 3H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 169.6, 150.6, 145.7, 145.0, 131.6, 127.2, 125.7, 125.0, 121.9, 117.5, 112.6, 108.2, 21.3

HRMS m/z (ESI) : calcd. for $[C_{15}H_{12}N_2O_2+H]^+$:253.0972 Found : 253.0975

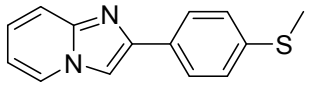


(**3la**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.09 (d, J = 6.8 Hz, 1H), 7.79 (s, 1H), 7.58–7.51 (m, 2H), 7.26–7.17 (m, 2H), 7.08-7.01 (m, 1H), 6.91–6.85 (m, 1H), 6.81 (t, J = 6.7 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 157.4, 145.2, 143.4, 129.7, 125.8, 125.5, 125.3, 119.1, 117.7, 116.7, 116.3, 113.2, 106.8

HRMS m/z (ESI) : calcd. for $[C_{13}H_{10}N_2O+H]^+$:211.0866 Found : 211.0874

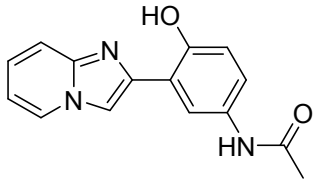


(**3ma**)

^{1}H NMR (400 MHz, CDCl3) δ 8.04 (t, J = 4.1 Hz, 1H), 7.86-7.80 (m, 2H), 7.77-7.74 (m, 1H), 7.69-7.64 (m, 1H), 7.32-7.26 (m, 2H), 7.18-7.12 (m, 1H), 6.77-6.71 (m, 1H), 2.50 (s, 3H)

^{13}C NMR (101 MHz, CDCl3) δ 174.6, 145.6, 145.1, 138.4, 130.3, 126.6, 126.5, 125.6, 125.0, 117.3, 112.7, 108.0, 15.7

HRMS m/z (ESI) : calcd. for $[C_{14}H_{12}N_2S+H]^+$:241.0794 Found : 241.0769

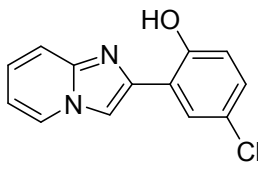


(**3na**)

^{1}H NMR (400 MHz, dmso-d_6) δ 11.50 (s, 1H), 9.82 (s, 1H), 8.64 (d, J = 6.7 Hz, 1H), 8.40 (s, 1H), 8.15 (s, 1H), 7.64 (d, J = 9.0 Hz, 1H), 7.37-7.29 (m, 2H), 6.97 (t, J = 6.5 Hz, 1H), 6.86 (d, J = 8.7 Hz, 1H), 2.02 (s, 3H)

^{13}C NMR (101 MHz, dmso-d_6) δ 167.7, 151.8, 143.1, 142.5, 131.3, 127.0, 125.7, 120.8, 117.7, 117.3, 116.5, 116.0, 112.8, 109.8, 23.9

HRMS m/z (ESI) : calcd. for $[C_{15}H_{13}N_3O_2+H]^+$: 268.1081 Found : 268.1084

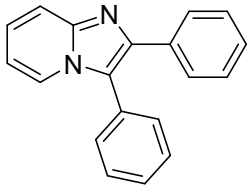


(**3oa**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 12.76 (s, 1H), 8.15 (d, J = 6.7 Hz, 1H), 7.84 (s, 1H), 7.59 (d, J = 9.0 Hz, 1H),7.53(s, 1H), 7.29-7.22 (m, 1H), 7.18-7.13 (m, 1H), 6.94-6.99 (m, 1H), 6.88 (t, J = 6.8 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 156.0, 144.1, 143.6, 129.4, 125.7, 125.6, 125.3, 123.7, 119.2, 117.5, 117.0, 113.6, 107.2

HRMS m/z (ESI) : calcd. for $[C_{13}H_9ClN_2O+H]^+$: 245.0476 Found : 245.0476

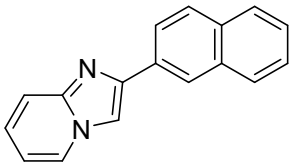


(**3pa**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97–7.93 (m, 1H), 7.71–7.64 (m, 3H), 7.55–7.51 (m, 1H), 7.51–7.41 (m, 4H), 7.31–7.23 (m, 3H), 7.22-7.16 (m, 1H), 6.72 (td, J = 6.8, 1.1 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 144.9, 142.5, 134.2, 130.8, 129.9, 129.6, 128.9, 128.4, 128.2, 127.6, 124.8, 123.4, 121.2, 117.6, 112.4

HRMS m/z (ESI) : calcd. for $[C_{19}H_{14}N_2+H]^+$:258.1151 Found : 258.1178

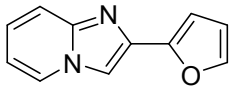


(**3qa**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.50 (s, 1H), 7.98–7.93 (m, 2H), 7.91 (d, J = 7.4 Hz, 1H), 7.87-7.80 (m, 3H), 7.64 (d, J = 9.1 Hz, 1H), 7.51–7.42 (m, 2H), 7.15–7.08 (m, 1H), 6.67 (t, J = 6.6 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 145.8, 145.6, 133.7, 133.2, 131.1, 128.4, 128.3, 127.7, 126.3, 125.98, 125.6, 124.8, 124.7, 124.2, 117.4, 112.4, 108.6

HRMS m/z (ESI) : calcd. for $[C_{17}H_{12}N_2+H]^+$:245.1073 Found : 245.1076

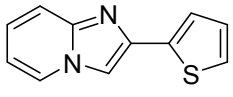


(**3ra**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.05 (d, J = 6.7 Hz, 1H), 7.75 (s, 1H), 7.56 (d, J = 9.1 Hz, 1H), 7.44 (d, J = 0.8 Hz, 1H), 7.16-7.10 (m, 1H), 6.87 (d, J = 3.2 Hz, 1H), 6.73 (t, J = 6.7 Hz, 1H), 6.50-6.46 (m, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 149.6, 145.6, 142.1, 137.9, 125.8, 125.1, 117.3, 112.6, 111.7, 108.0, 106.8

HRMS m/z (ESI) : calcd. for $[C_{17}H_{12}N_2+H]^+$:185.0709 Found : 185.0720

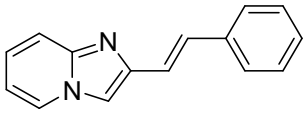


(**3sa**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.01 (d, J = 6.8 Hz, 1H), 7.71 (s, 1H), 7.57 (d, J = 9.1 Hz, 1H), 7.44 (d, J = 3.5 Hz, 1H), 7.30-7.27 (m, 1H), 7.16–7.09 (m, 1H), 7.09-7.03 (m, 1H), 6.72 (t, J = 6.7 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 145.5, 140.9, 137.6, 127.8, 125.5, 125.0, 123.7, 117.3, 112.6, 107.5

HRMS m/z (ESI) : calcd. for $[C_{11}H_8N_2S+H]^+$:201.0481 Found :201.0485

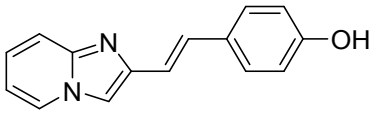


(**3ta**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.01-7.95 (m, 1H), 7.58-7.51 (m, 5H), 7.38-7.32 (m, 2H), 7.28–7.23 (m, 1H), 7.16–7.09 (m, 2H), 6.72-6.65 (m, 1H)

^{13}C NMR (101 MHz, CDCl3) δ 145.7, 144.1, 137.2, 130.4, 128.7, 127.7, 126.6, 125.5, 125.1, 120.0, 117.1, 112.2, 110.7

HRMS m/z (ESI) : calcd. for $[C_{15}H_{12}N_2+H]^+$: 221.1078 Found : 221.1065

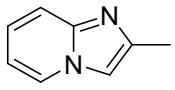


(**3ua**)

^{1}H NMR (400 MHz, $CD_3OD/CDCl_3$) δ 8.31 (d, J = 6.8 Hz, 1H), 7.81 (s, 1H), 7.47 (d, J = 9.1 Hz, 1H), 7.40 (d, J = 8.6 Hz, 2H), 7.33 (d, J = 16.3 Hz, 1H), 7.29-7.24 (m, 1H), 6.97 (d, J = 16.2 Hz, 1H), 6.85 (t, J = 6.8 Hz, 1H), 6.81-6.77 (m, 2H), 3.35 (s, 1H)

^{13}C NMR (101 MHz, $CD_3OD/CDCl_3$) δ 158.5, 146.6, 145.1, 131.7, 129.9, 128.9, 127.4, 126.9, 117.6, 116.6, 116.5, 113.6, 111.6

HRMS m/z (ESI) : calcd. for $[C_{15}H_{12}N_2+H]^+$: 237.1023 Found : 237.1007

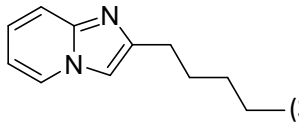


(**3va**)

^{1}H NMR (400 MHz, CDCl3) δ 8.01 (d, J = 6.7 Hz, 1H), 7.48 (d, J = 9.9 Hz, 1H), 7.31 (s, 1H), 7.11-7.05 (m, 1H), 6.70 (t, J = 6.7 Hz, 1H), 2.44 (s, 3H)

^{13}C NMR (101 MHz, CDCl3) δ 143.5, 125.3, 124.0, 116.9, 111.9, 109.5, 14.5

HRMS m/z (ESI) : calcd. for $[C_8H_8N_2+H]^+$: 133.0765 Found : 133.0758

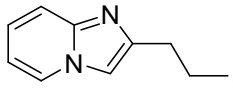


(**3wa**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.05-8.00 (m, 1H), 7.55-7.50 (m, 1H), 7.31 (s, 1H), 7.10-7.06 (m, 1H), 6.72-6.66 (m, 1H), 2.77 (t, J = 16 Hz, 2H), 1.82–1.72 (m, 2H), 1.44-1.30 (m, 4H), 0.90 (t, J = 7.0 Hz, 3H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 148.1, 144.9, 125.2, 123.9, 116.8, 111.7, 108.9, 31.7, 29.1, 28.9, 22.5, 14.1

HRMS m/z (ESI) : calcd. for $[C_{12}H_{16}N_2+H]^+$: 189.1386 Found : 189.1384

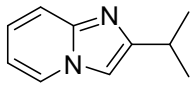


(**3xa**)

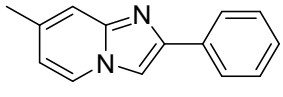
^{1}H NMR (400 MHz, $CDCl_3$) δ 8.00–7.95 (m, 1H), 7.47 (d, J = 9.0 Hz, 1H), 7.27 (s, 1H), 7.07-7.01 (m, 1H), 6.65 (t, J = 6.8 Hz, 1H), 2.71 (t, J = 7.6 Hz, 2H), 1.79-1.69 (m, 2H), 0.95 (t, J = 7.4 Hz, 3H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 147.9, 145.0, 125.3, 124.0, 116.9, 111.8, 109.0, 31.0, 22.6, 14.1

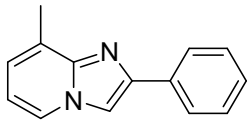
HRMS m/z (ESI) : calcd. for $[C_{10}H_{12}N_2+H]^+$: 161.1073 Found : 161.1068


(**3ya**)

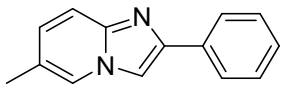
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98 (dt, J = 6.8, 1.2 Hz, 1H), 7.50 (dd, J = 9.1, 0.7 Hz, 1H), 7.27 (s, 1H), 7.07-7.02 (m, 1H), 6.65 (td, J = 6.8, 1.1 Hz, 1H), 3.13-3.01 (m, 1H), 1.32(d, J = 4 Hz, 6H)
^{13}C NMR (101 MHz, $CDCl_3$) δ 154.1, 145.0, 125.4, 124.0, 117.0, 111.7, 107.4, 28.4, 22.5
HRMS m/z (ESI) : calcd. for $[C_{10}H_{12}N_2+H]^+$: 161.1073 Found : 161.1076


(**3ab**)

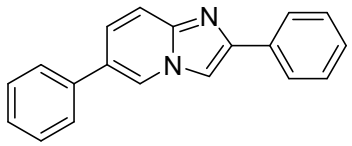
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98–7.91 (m, 3H), 7.75 (s, 1H), 7.46-7.38 (m, 2H), 7.37 (s, 1H), 7.34-7.29 (m, 1H), 6.58 (dd, J = 6.9, 1.4 Hz, 1H), 2.38 (s, 3H)
^{13}C NMR (101 MHz, $CDCl_3$) δ 146.3, 145.6, 135.6, 134.1, 128.8, 127.9, 126.1, 124.9, 116.0, 115.1, 107.6, 21.5
HRMS m/z (ESI) : calcd. for $[C_{14}H_{12}N_2+H]^+$:209.1073 Found :209.1079


(**3ac**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.97 (d, J = 7.2 Hz, 2H), 7.81 (d, J = 6.7 Hz, 1H), 7.69 (s, 1H), 7.47-7.39 (m, 2H), 7.35-7.28 (m, 1H), 6.90-6.85 (m, 1H), 6.55 (t, J = 6.8 Hz, 1H), 2.65 (s, 3H)
^{13}C NMR (101 MHz, $CDCl_3$) δ 146.0, 145.0, 134.0, 128.6, 127.6, 127.2, 126.0, 123.3, 123.2, 112.15, 108.6, 17.1
HRMS m/z (ESI) : calcd. for $[C_{14}H_{12}N_2+H]^+$:209.1073 Found :209.1068


(**3ad**)

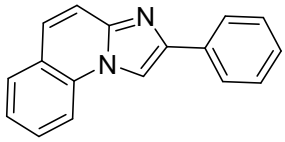
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.90 (d, J = 7.5 Hz, 2H), 7.74 (s, 1H), 7.64 (s, 1H), 7.48 (d, J = 9.2 Hz, 1H), 7.40 (t, J = 7.6 Hz, 2H), 7.32-7.26 (m, 1H), 6.94 (dd, J = 9.2, 1.5 Hz, 1H), 2.21 (s, 3H)
^{13}C NMR (101 MHz, $CDCl_3$) δ 145.3, 144.7, 133.9, 128.7, 127.8, 127.7, 125.9, 123.3, 122.0, 116.7, 107.9, 18.1
HRMS m/z (ESI) : calcd. for $[C_{14}H_{12}N_2+H]^+$: 209.1073 Found : 209.1063


(**3ae**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.24 (s, 1H), 7.97 (d, J = 8.2 Hz, 2H), 7.86 (s, 1H), 7.68 (d, J = 9.3 Hz, 1H), 7.58-7.52 (m, 2H), 7.50-7.37 (m, 6H), 7.36-7.31 (m, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 146.3, 145.1, 137.3, 133.8, 129.2, 128.8, 128.1, 128.0, 126.9, 126.1, 125.5, 122.9, 117.4, 108.6

HRMS m/z (ESI) : calcd. for $[C_{19}H_{14}N_2+H]^+$: 271.1230 Found : 271.1225

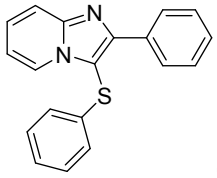


(**3af**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.24 (s, 1H), 8.00 (d, J = 8.2 Hz, 2H), 7.84 (d, J = 8.3 Hz, 1H), 7.73 (d, J = 7.9 Hz, 1H), 7.60–7.53 (m, 2H), 7.48-7.37 (m, 4H), 7.36-7.30 (m, 1H)

13C NMR (101 MHz, $CDCl_3$) δ 144.8, 144.1, 133.8, 132.5, 129.2, 128.8, 127.8, 126.4, 125.8, 124.7, 123.3, 117.0, 115.1, 106.8

HRMS m/z (ESI) : calcd. for $[C_{10}H_{12}N_2+H]^+$: 245.1073 Found :245.1058

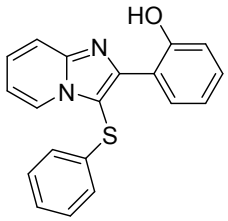


(**5a**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.27 (dt, J = 6.8, 1.1 Hz, 1H), 8.23–8.19 (m, 2H), 7.74 (dt, J = 9.0, 1.0 Hz, 1H), 7.46–7.42 (m, 2H), 7.40-7.35(m, 1H), 7.35–7.30 (m, 1H), 7.23–7.18 (m, 2H), 7.15–7.11 (m, 1H), 7.02–6.99 (m, 2H), 6.86 (td, J = 6.8, 1.1 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 151.5, 147.2, 135.3, 133.4, 129.5, 128.7, 128.52, 128.45, 126.8, 126.1, 125.6, 124.6, 117.8, 113.2, 106.3

HRMS m/z (ESI) : calcd. for $[C_{19}H_{14}N_2S+H]^+$:303.0950 Found :303.0966

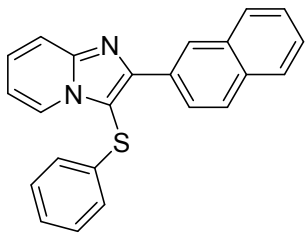


(**5b**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 13.11 (s, 1H), 8.60 (d, J = 7.9 Hz, 1H), 8.32 (d, J = 6.8 Hz, 1H), 7.63 (d, J = 9.0 Hz, 1H), 7.37-6.98 (m, 9H), 6.92-6.81 (dt, J = 15.1, 7.1 Hz, 2H)

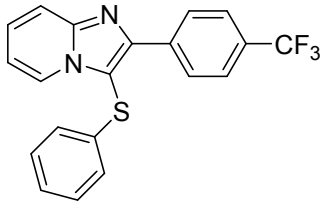
13C NMR (101 MHz, $CDCl_3$) δ 158.5, 149.5, 144.7, 134.4, 130.7, 129.6, 127.8, 127.5, 126.4, 125.9, 124.3, 118.9, 117.8, 116.7, 116.2, 113.9, 105.4

HRMS m/z (ESI) : calcd. for $[C_{19}H_{14}N_2S+H]^+$:303.0950 Found :303.0966



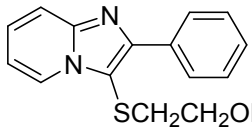
(**5c**)

1H NMR (400 MHz, $CDCl_3$) δ 8.73 (s, 1H), 8.39 (dd, J = 8.6, 1.7 Hz, 1H), 8.26 (dt, J = 6.8, 1.0 Hz, 1H), 7.90-7.86 (m, 2H), 7.84-7.78 (m, 1H), 7.76-7.72 (m, 1H), 7.46-7.42 (m, 2H), 7.31-7.26 (m, 1H), 7.20-7.15 (m, 2H), 7.12-7.07 (m, 1H), 7.04-7.00 (m, 2H), 6.80 (td, J = 6.8, 1.0 Hz, 1H)
13C NMR (101 MHz, $CDCl_3$) δ 151.3, 147.2, 135.2, 133.4, 130.9, 129.5, 128.7, 128.0, 127.9, 127.7, 126.8, 126.4, 126.2, 126.1, 126.0, 125.8, 124.5, 117.7, 113.2, 106.8
HRMS m/z (ESI) : calcd. for $[C_{23}H_{16}N_2S+H]^+$:353.1107 Found :353.1087



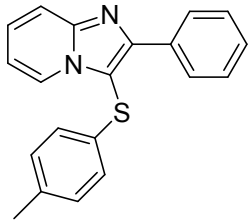
(**5d**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.36 (d, J = 8.2 Hz, 2H), 8.26 (d, J = 6.8 Hz, 1H), 7.72 (d, J = 9.0 Hz, 1H), 7.67 (d, J = 8.2 Hz, 2H), 7.35-7.30 (m, 1H), 7.23-7.16 (m, 2H), 7.15-7.09 (m, 1H), 6.97 (d, J = 7.4 Hz, 2H), 6.85 (t, J = 6.8 Hz, 1H)
^{13}C NMR (101 MHz, $CDCl_3$) δ 149.7, 147.2, 136.9, 134.7, 130.2 (J = 32 Hz), 129.6, 128.5, 127.1, 126.4, 125.6, 125.4 (J = 4 Hz), 124.6, 122.9, 117.9, 113.5, 107.3
HRMS m/z (ESI) : calcd. for $[C_{20}H_{13}F_3N_2S+H]^+$:371.0825 Found :371.0820



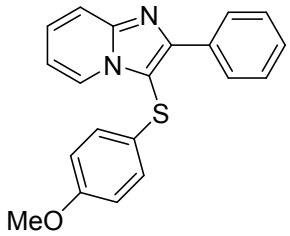
(**5e**)

^{1}H NMR (400 MHz, dmso-d_6) δ 8.69 (d, J = 8 Hz, 1H), 8.34-8.29 (m, 2H), 7.68 (dd, J = 9.0, 0.8 Hz, 1H), 7.49 (t, J = 7.7 Hz, 2H), 7.45-7.37 (m, 2H), 7.10 (t, J = 6.8 Hz, 1H), 4.91 (t, J = 5.4 Hz, 1H), 3.44-3.40 (m, 2H), 2.81 (t, J = 6.5 Hz, 2H)
^{13}C NMR (101 MHz, dmso-d_6) δ 148.1, 145.8, 133.8, 128.4, 128.2, 127.9, 126.7, 125.1, 117.0, 113.2, 109.4, 60.0, 38.1
HRMS m/z (ESI) : calcd. for $[C_{15}H_{14}N_2OS+H]^+$:271.0900 Found :271.0886



(**5f**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.27 (d, J = 6.8 Hz, 1H), 8.25-8.20 (m, 2H), 7.73 (d, J = 9.0 Hz, 1H), 7.47-7.42 (m, 2H), 7.40-7.35 (m, 1H), 7.34-7.30 (m, 1H), 7.02 (d, J = 8.2 Hz, 2H), 6.94-6.89 (m, 2H), 6.85 (t, J = 6.8 Hz, 1H), 2.26 (s, 3H)
^{13}C NMR (101 MHz, $CDCl_3$) δ 151.3, 147.1, 136.1, 133.5, 131.6, 130.3, 128.6, 128.5, 128.4, 126.7, 125.9, 124.6, 117.7, 113.1, 106.9, 21.00
HRMS m/z (ESI) : calcd. for $[C_{20}H_{16}N_2S+H]^+$:317.1107 Found :317.1097

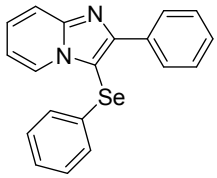


(**5g**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.29-8.23 (m, 3H), 7.68 (d, J = 9.0 Hz, 1H), 7.47-7.41 (m, 2H), 7.38-7.33 (m, 1H), 7.29-7.23 (m, 1H), 6.97 (d, J = 8.7 Hz, 2H), 6.83-6.77 (m, 1H), 6.74-6.69 (m, 2H), 3.66 (s, 3H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 158.5, 150.8, 146.8, 133.5, 128.5, 128.4, 128.4, 127.9, 126.5, 125.4, 124.4, 117.6, 115.1, 113.0, 107.7, 55.3

HRMS m/z (ESI) : calcd. for $[C_{20}H_{16}N_2OS+H]^+$:333.1056 Found :333.1051

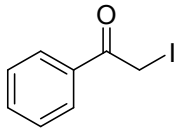


(**5h**)

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.35 (d, J = 6.9 Hz, 1H), 8.15(d, J = 8 Hz, 2H), 7.72 (d, J = 9.0 Hz, 1H), 7.47-7.41 (m, 2H), 7.40–7.35 (m, 1H), 7.33-7.29 (m, 1H), 7.20–7.15 (m, 3H), 7.12–7.08 (m, 2H), 6.85 (t, J = 6.8 Hz, 1H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 152.0, 147.9, 133.9, 131.0, 129.8, 129.0, 128.9, 128.6, 128.4, 128.3, 126.8, 126.6, 125.8, 117.6, 113.1

HRMS m/z (ESI) : calcd. for $[C_{19}H_{14}N_2Se+H]^+$:351.0395 Found :351.0380



(**6a**)

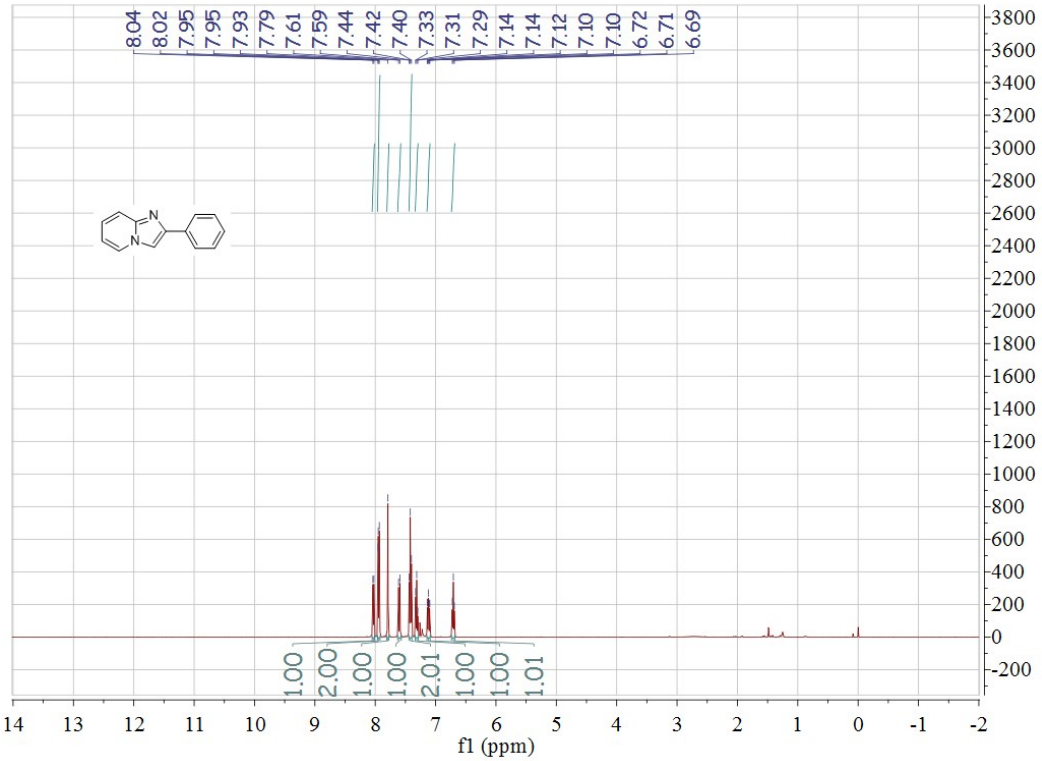
^{1}H NMR (400 MHz, $CDCl_3$) δ 7.99 (d, J = 8.0 Hz, 2H), 7.63-7.57 (m, 1H), 7.52-7.45 (m, 2H), 4.36 (s, 2H)

^{13}C NMR (101 MHz, $CDCl_3$) δ 193.0, 134.0, 133.5, 129.2, 129.0, 2.0

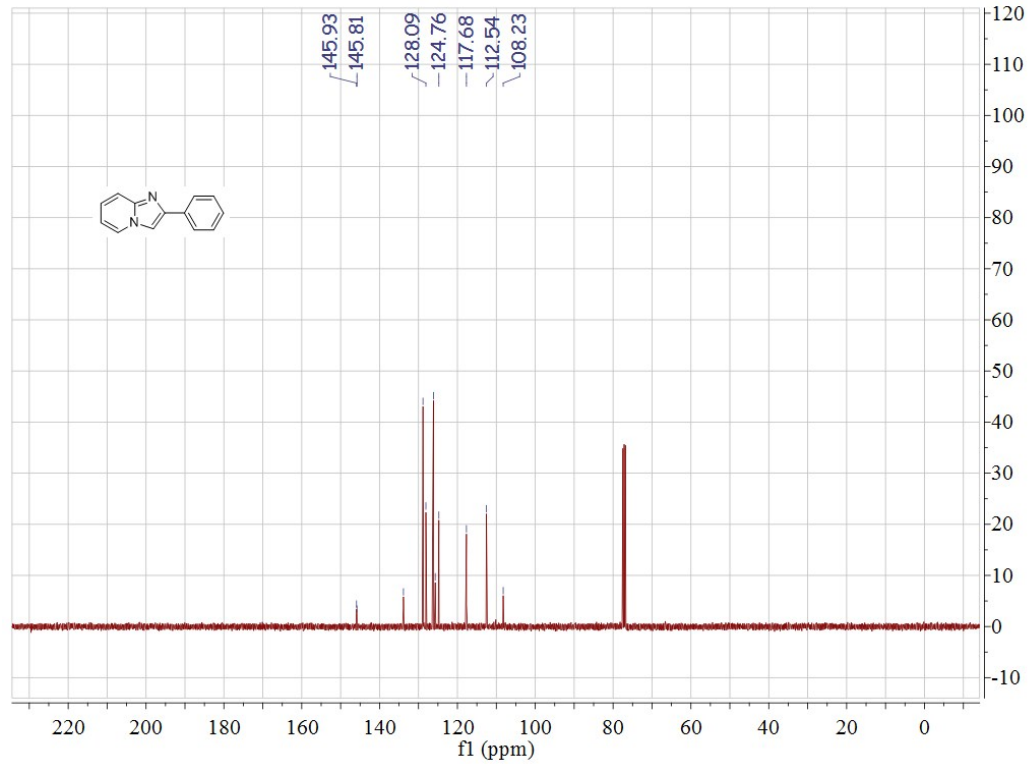
HRMS m/z (EI) : calcd. for $[C_8H_7IO]$:245.9542 Found :245.95

^{1}H and ^{13}C spectra of products

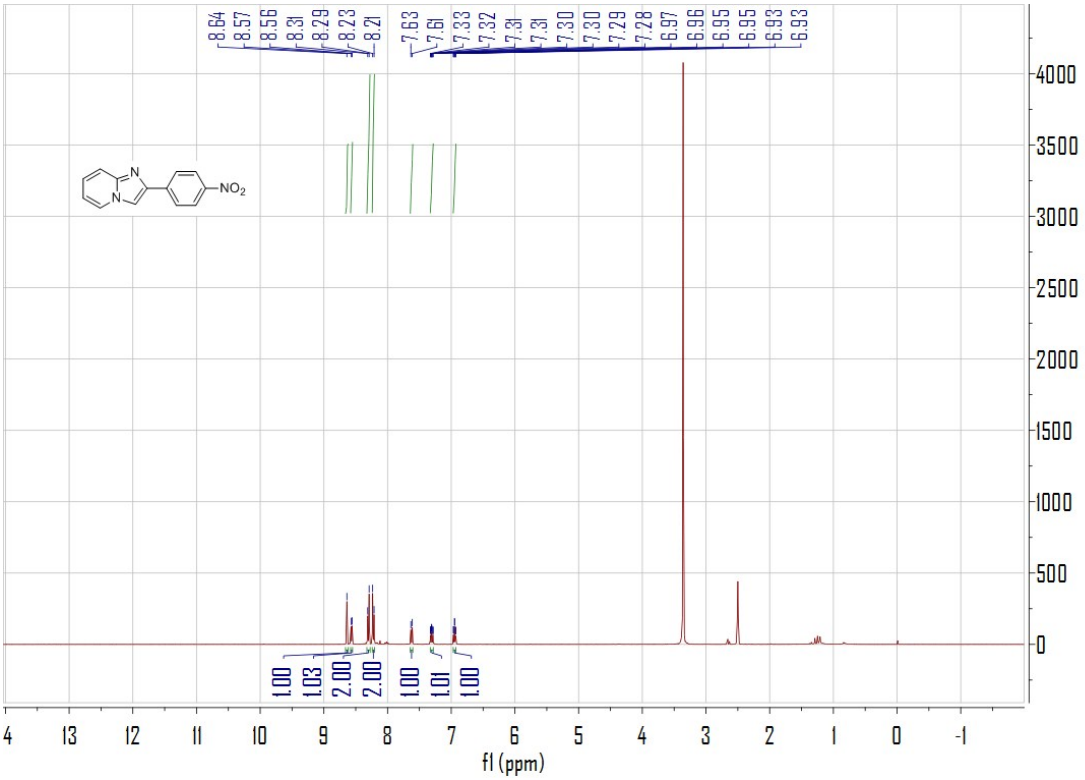
^{1}H NMR of compound **3aa**



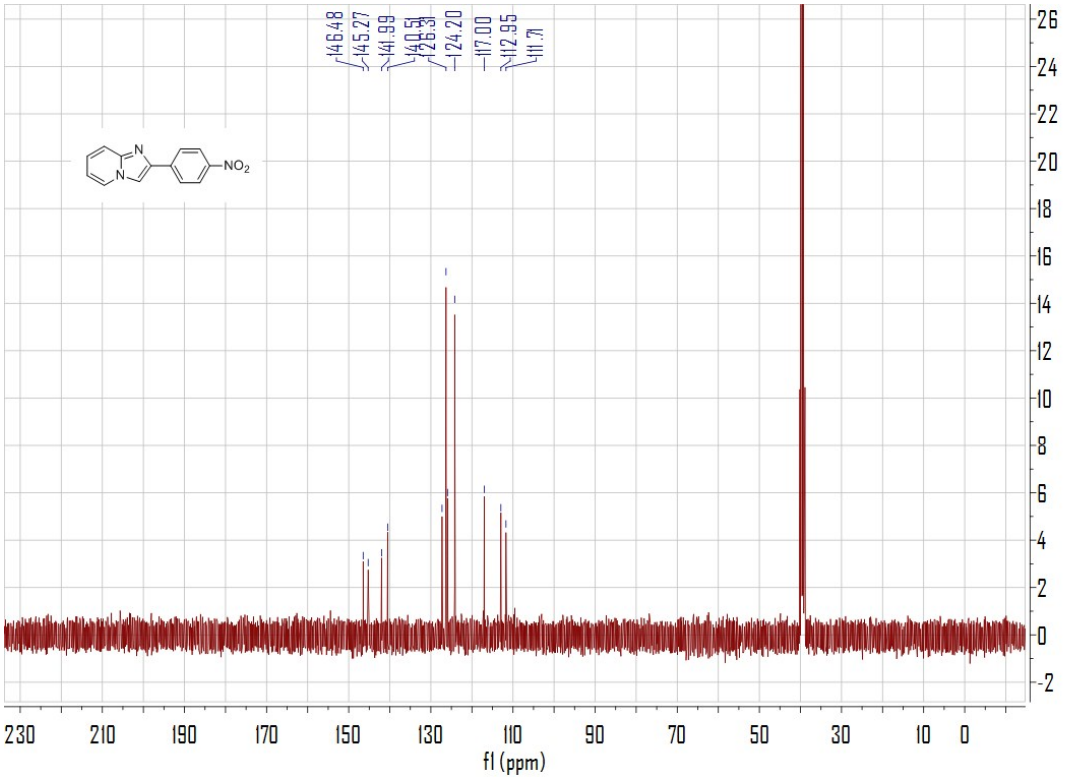
^{13}C NMR of compound **3aa**



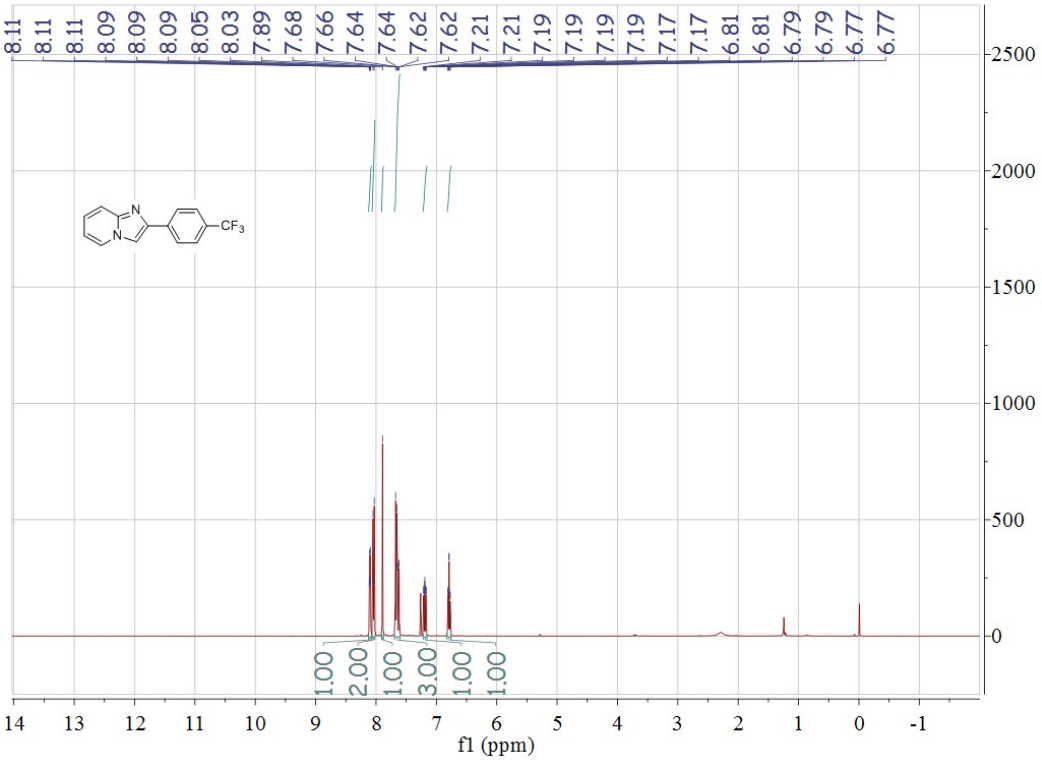
^{1}H NMR of compound **3ba**



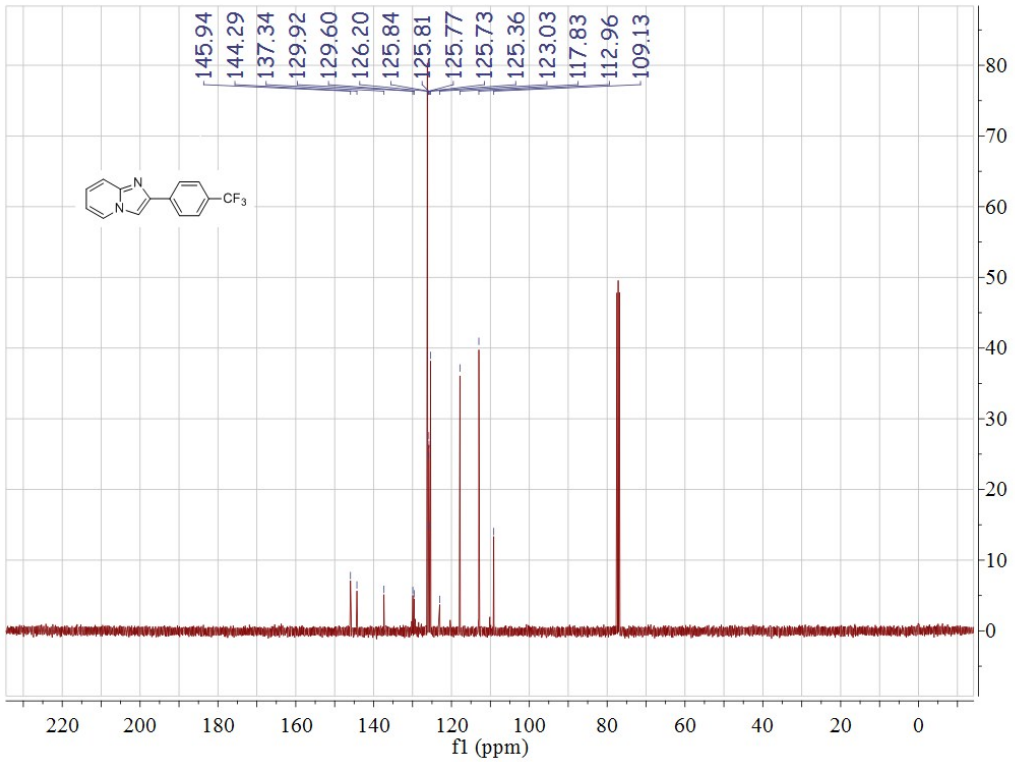
^{13}C NMR of compound **3ba**



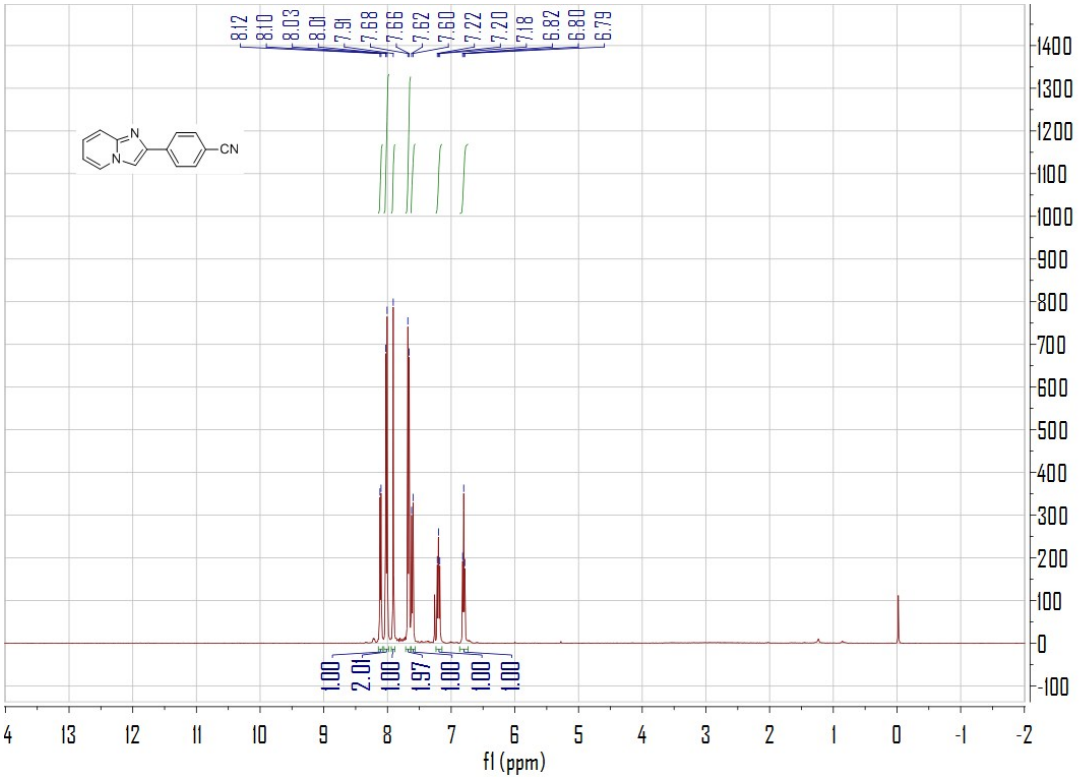
^{1}H NMR of compound **3ca**



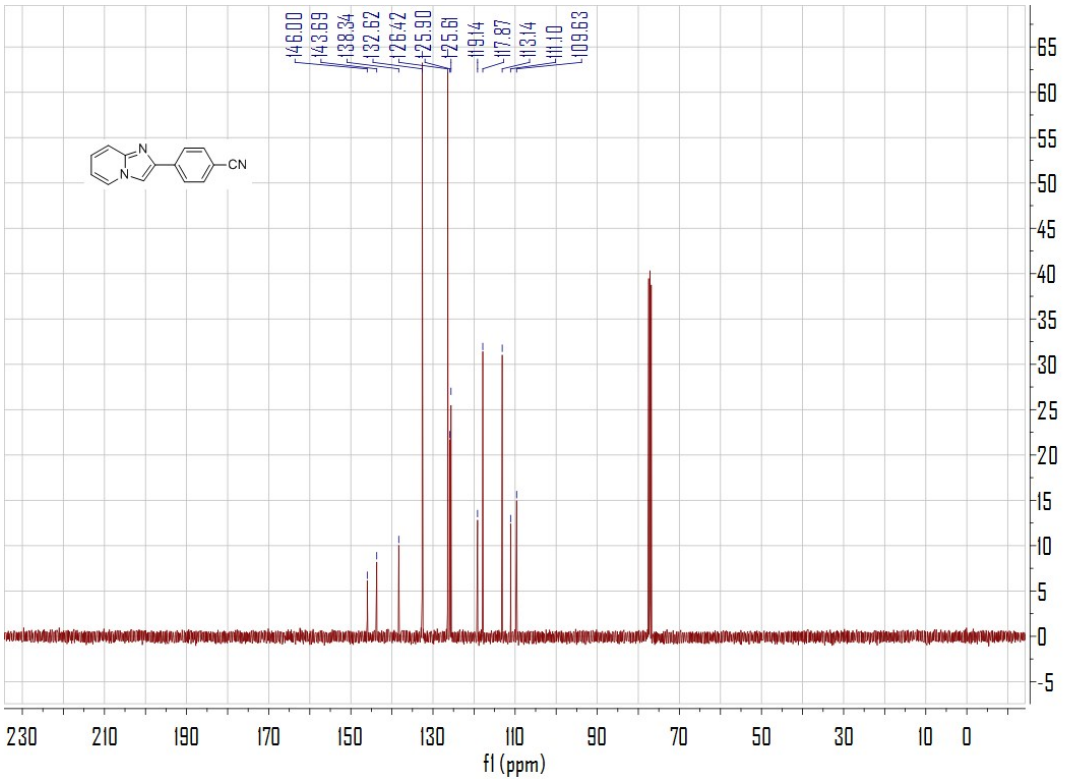
^{13}C NMR of compound **3ca**



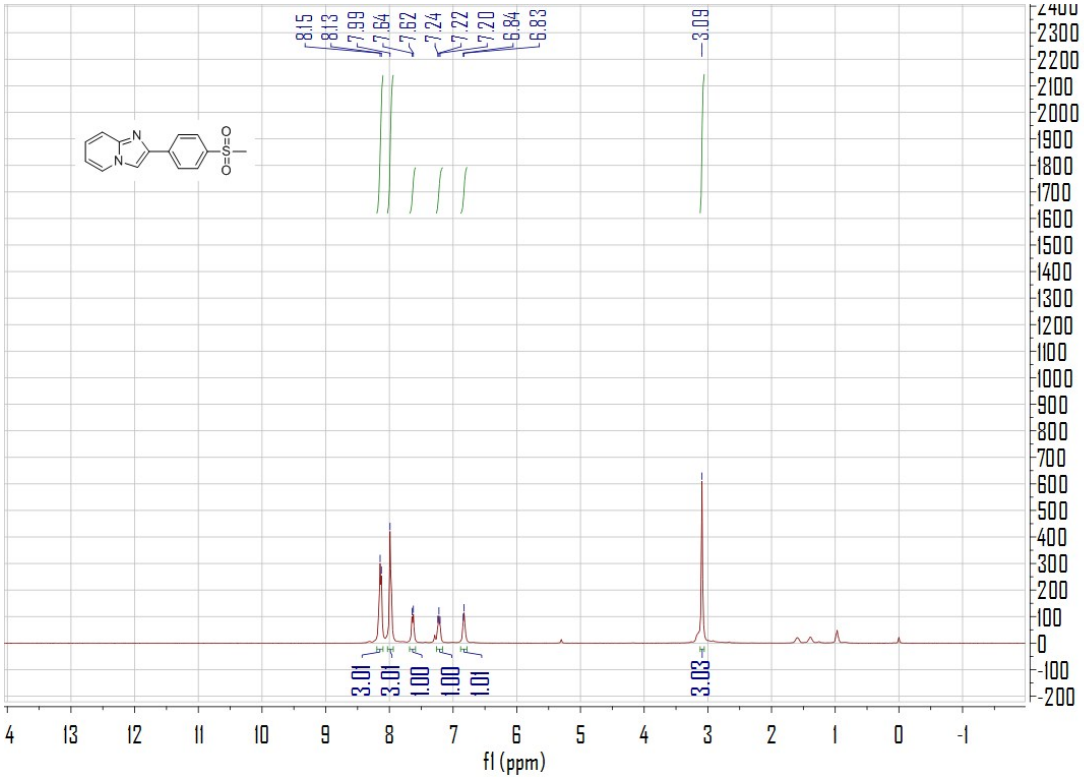
^{1}H NMR of compound **3da**



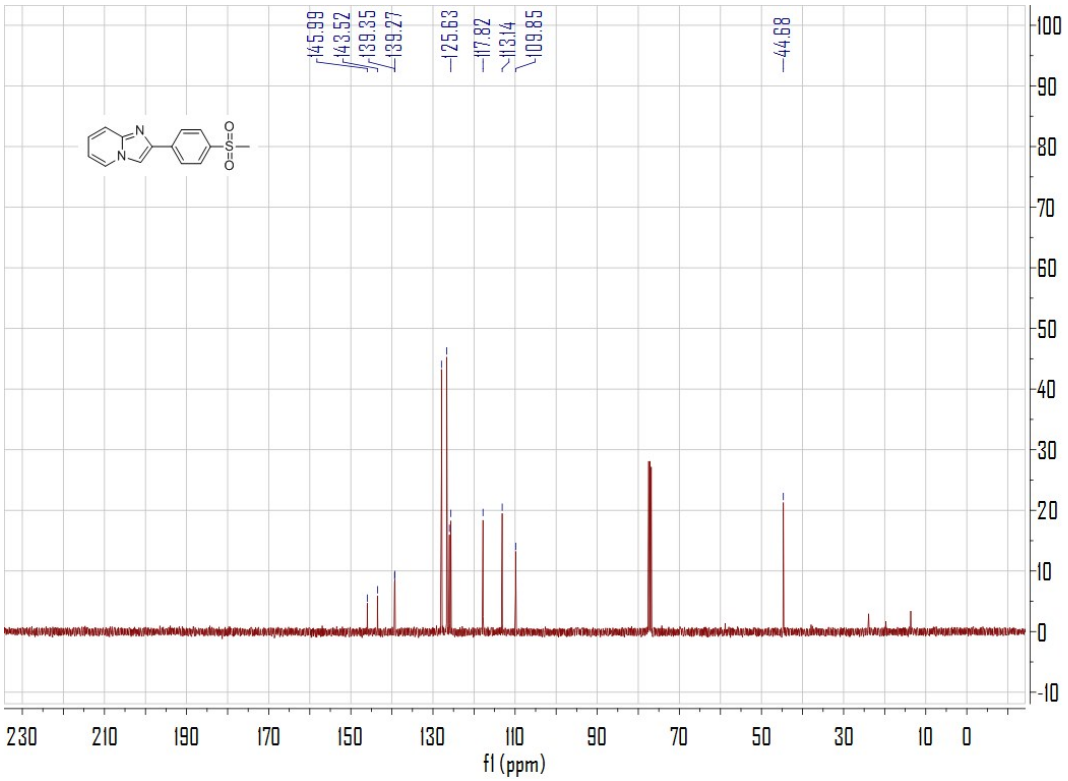
^{13}C NMR of compound **3da**



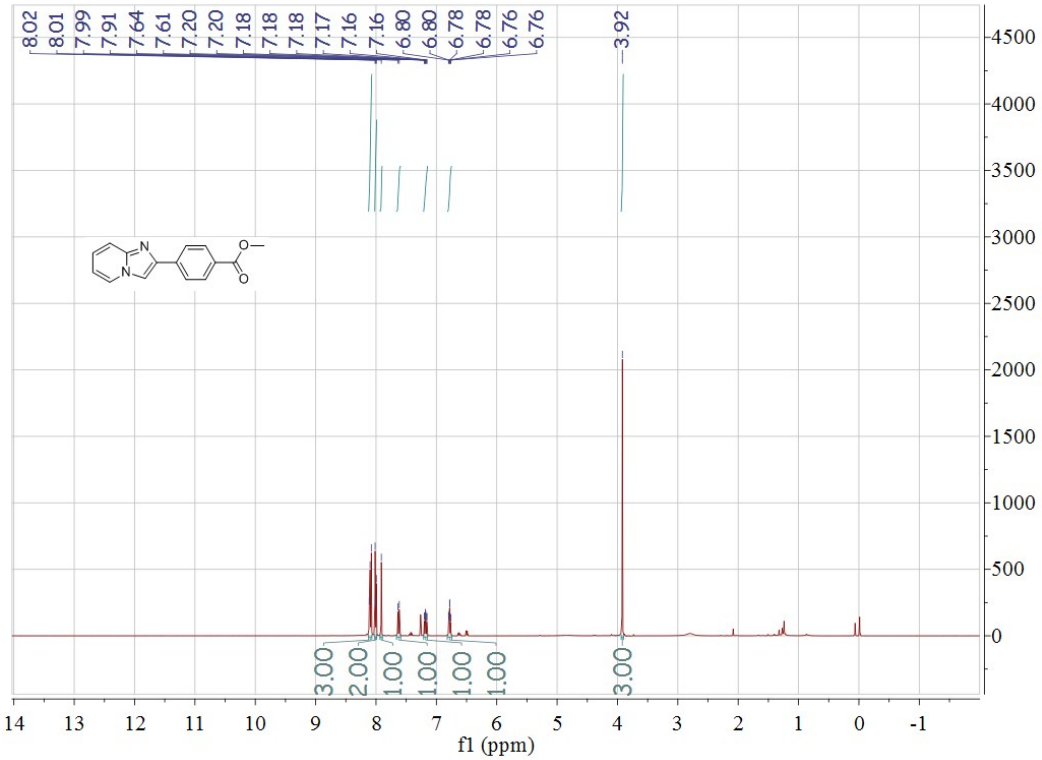
^{1}H NMR of compound **3ea**



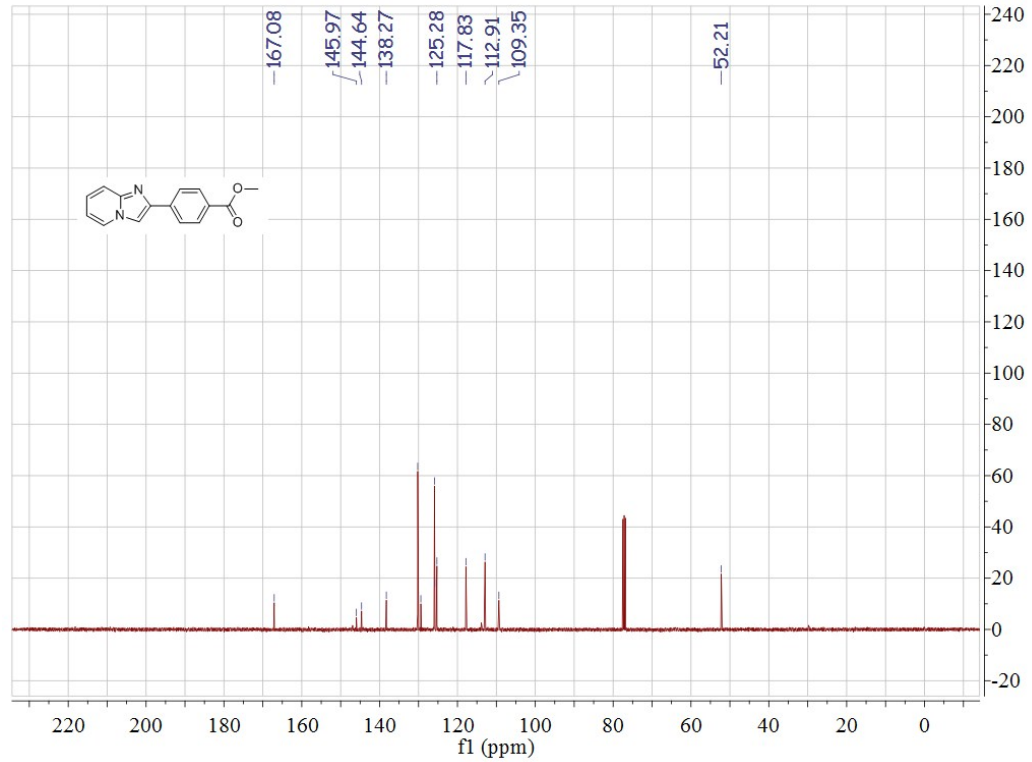
^{13}C NMR of compound **3ea**



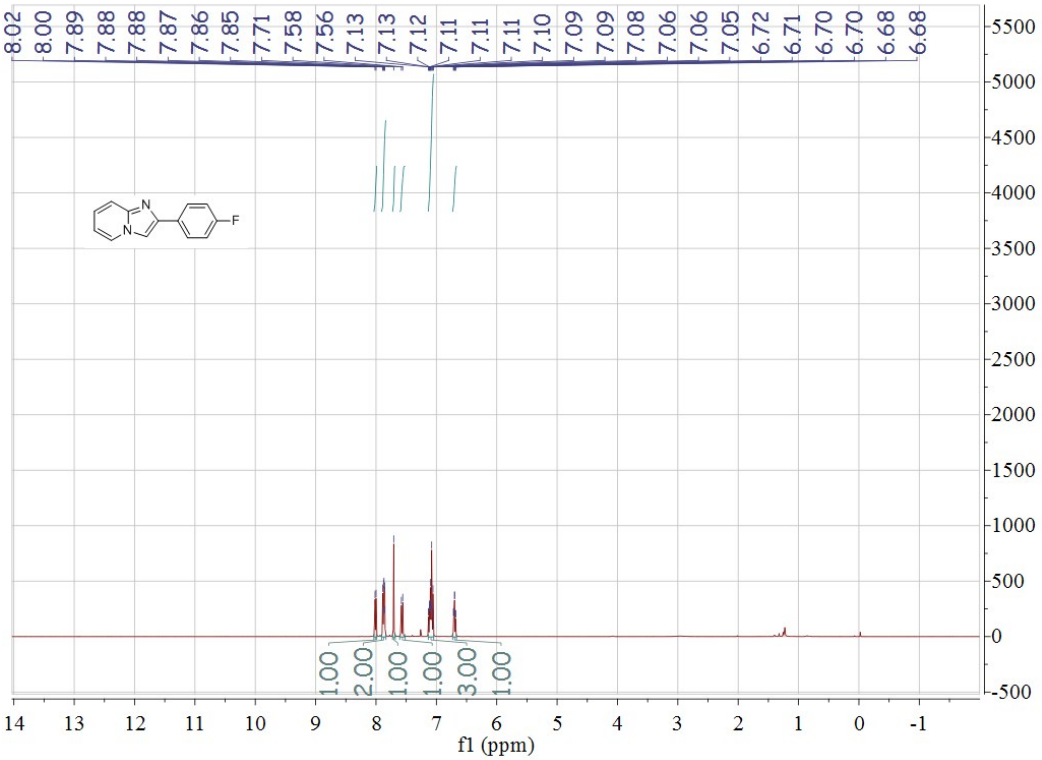
^{1}H NMR of compound **3fa**



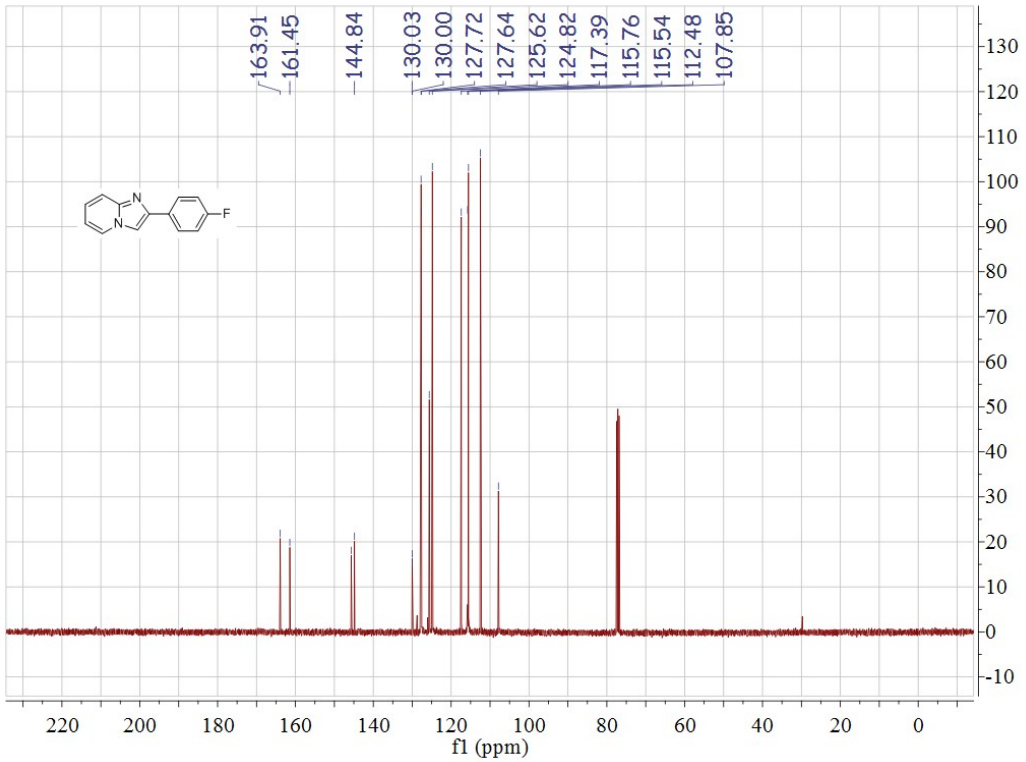
^{13}C NMR of compound **3fa**



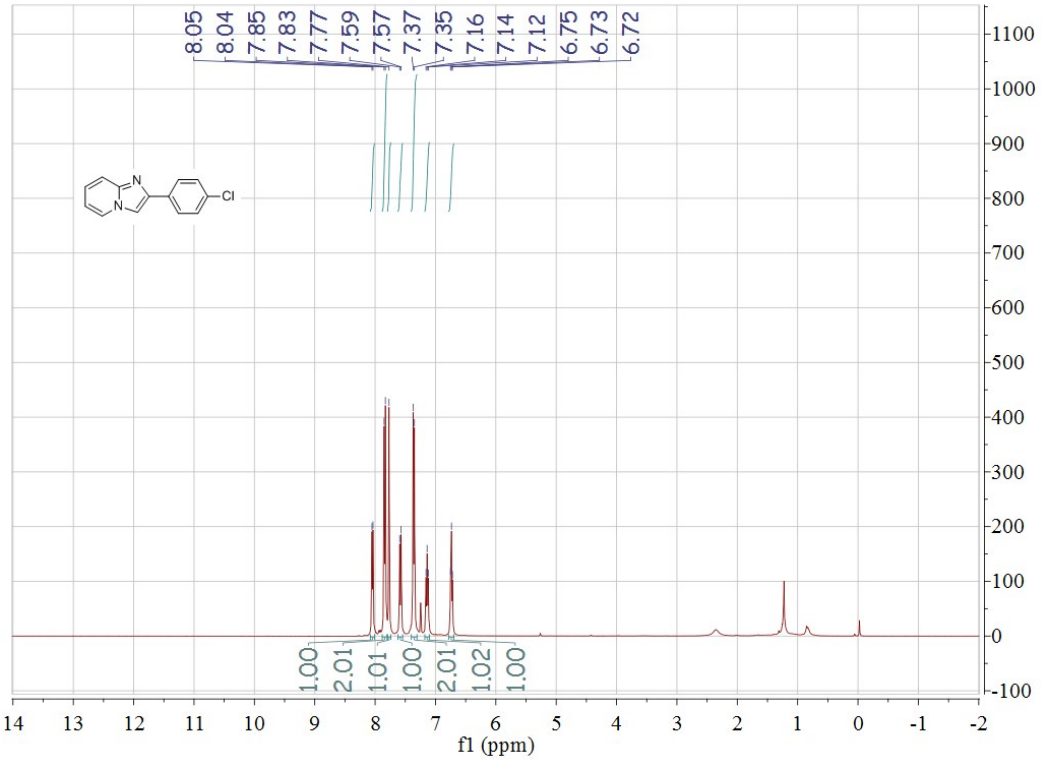
^{1}H NMR of compound **3ga**



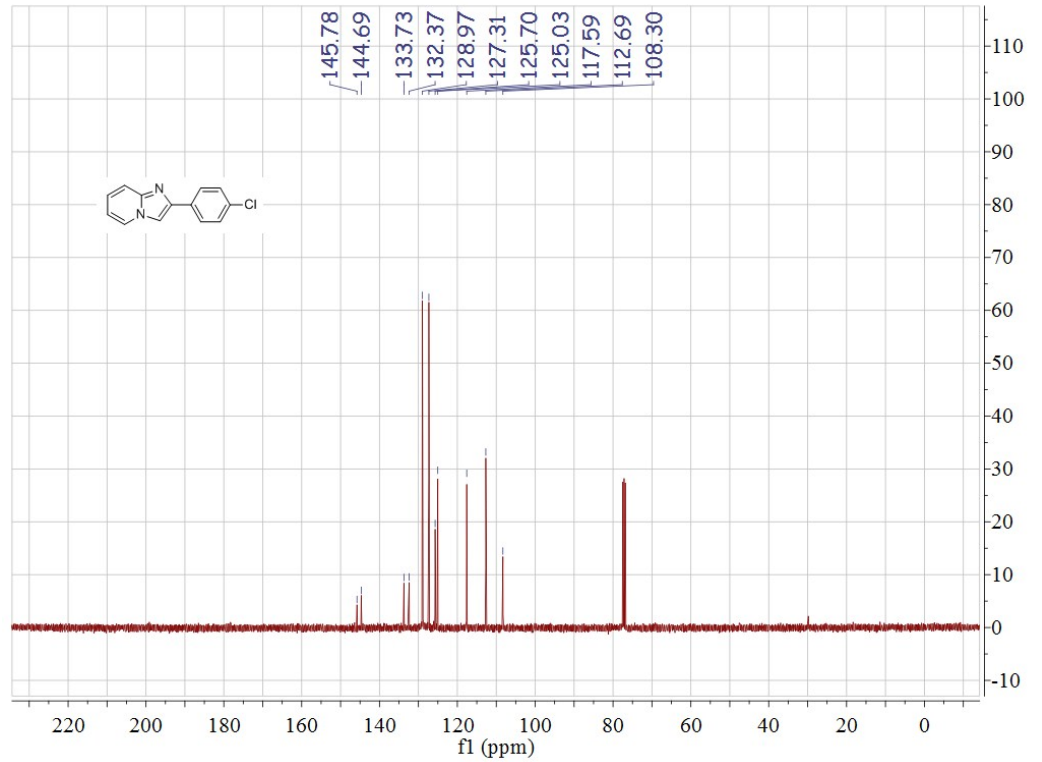
^{13}C NMR of compound **3ga**



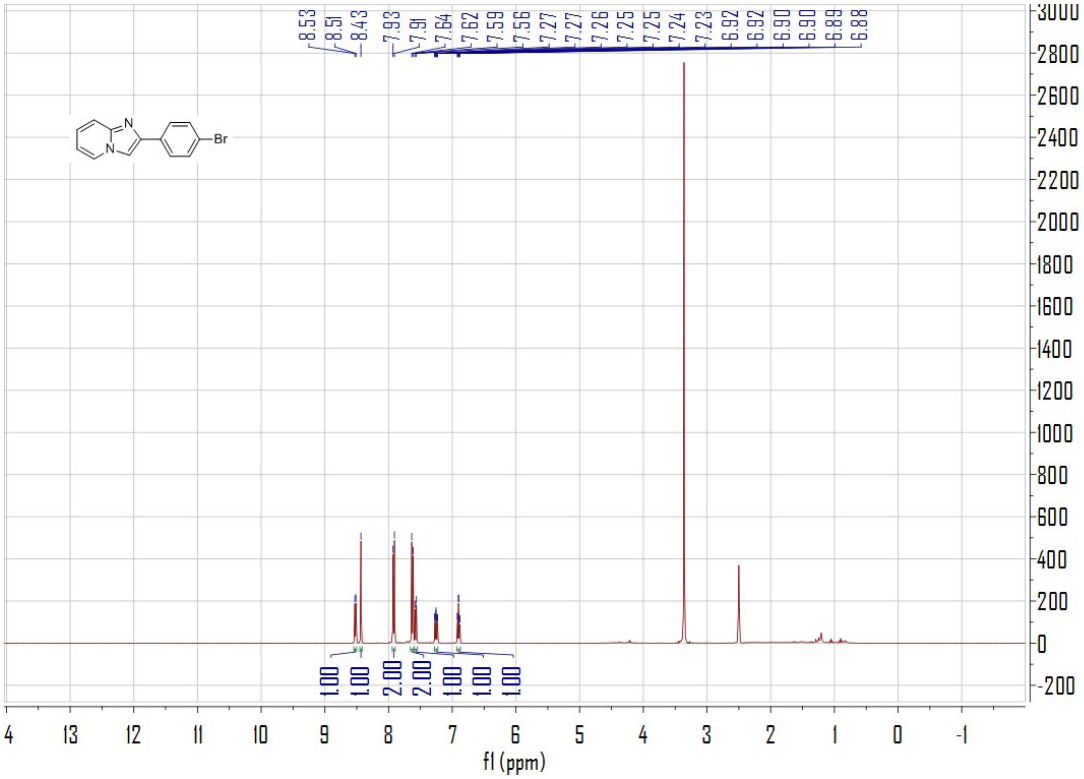
^{1}H NMR of compound **3ha**



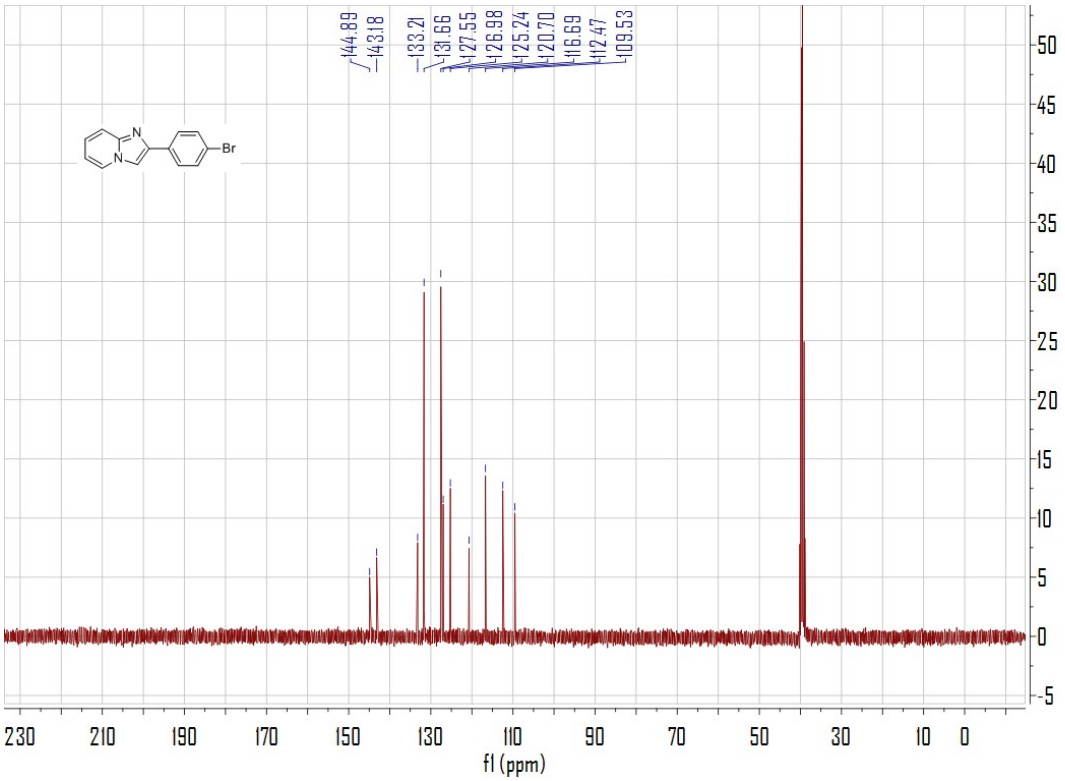
^{13}C NMR of compound **3ha**



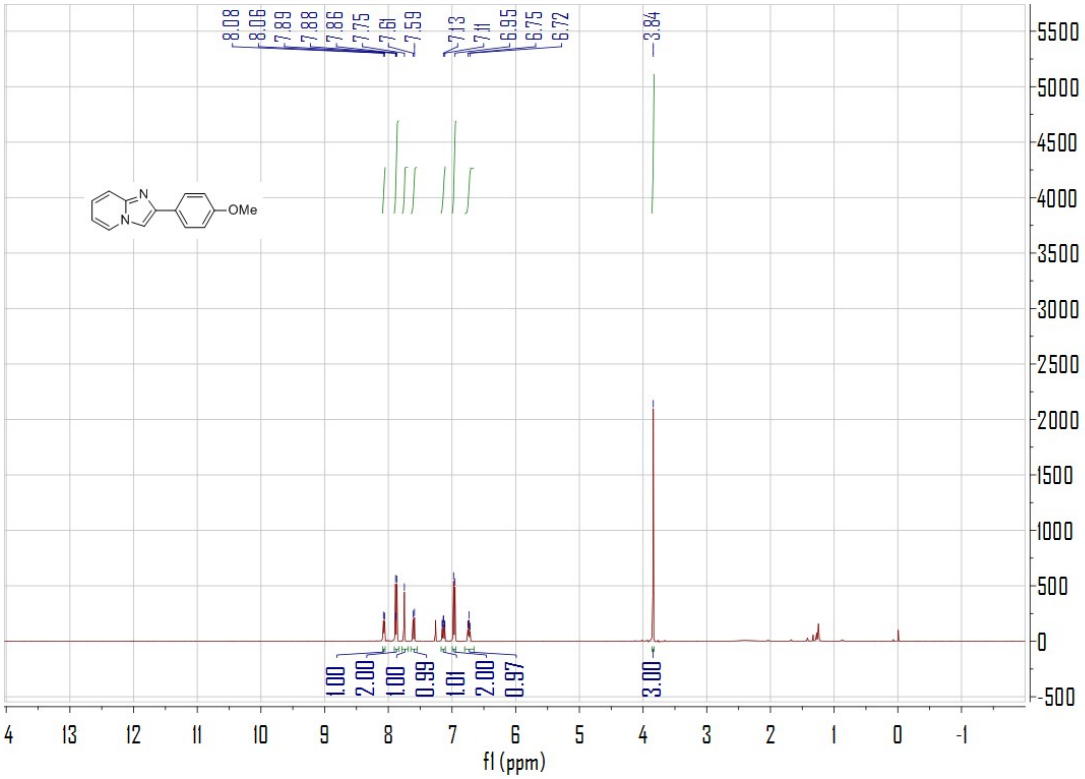
^{1}H NMR of compound **3ia**



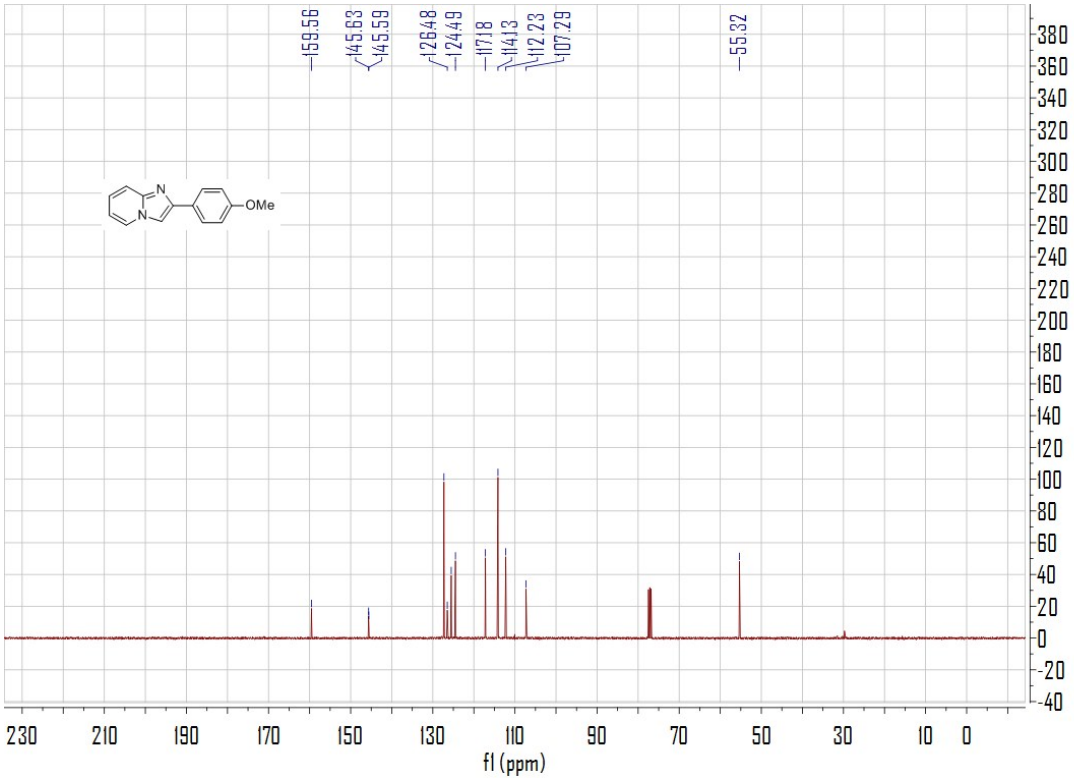
^{13}C NMR of compound **3ia**



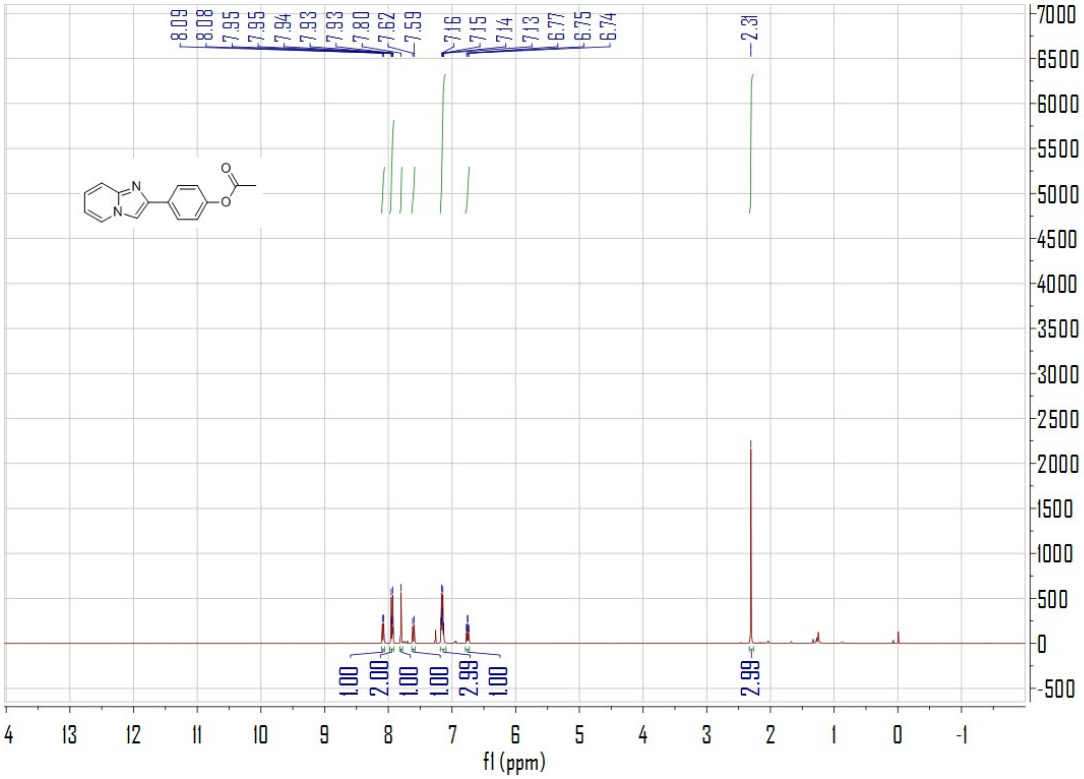
^{1}H NMR of compound **3ja**



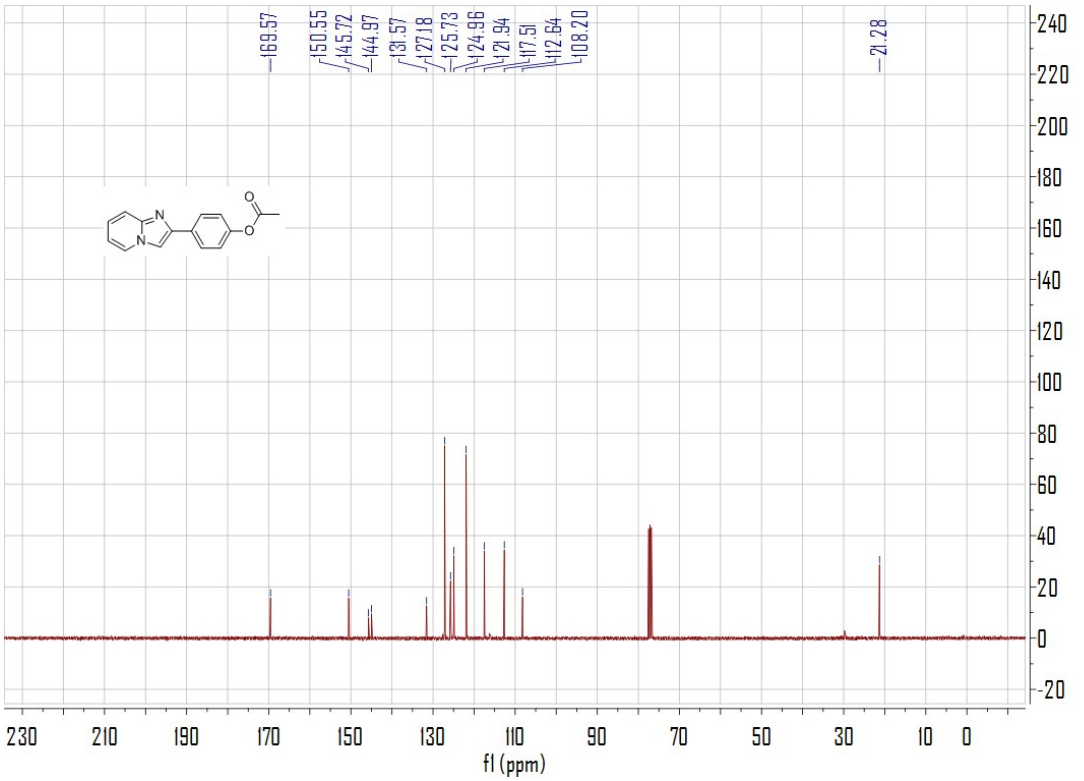
^{13}C NMR of compound **3ja**



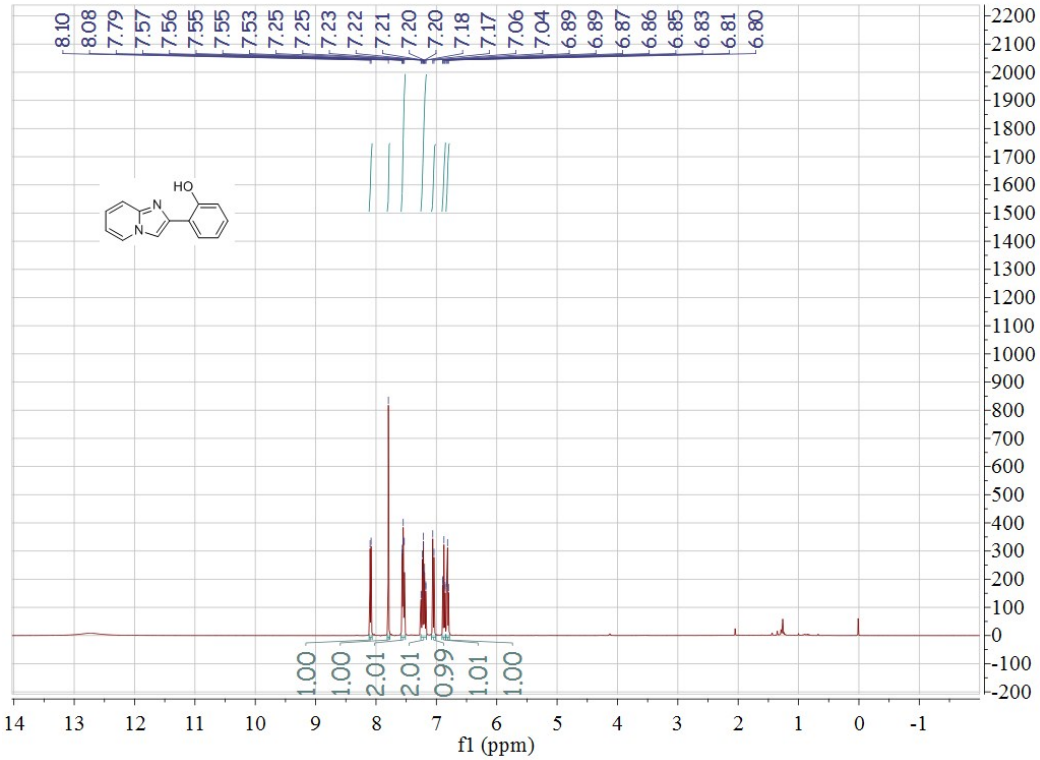
^{1}H NMR of compound **3ka**



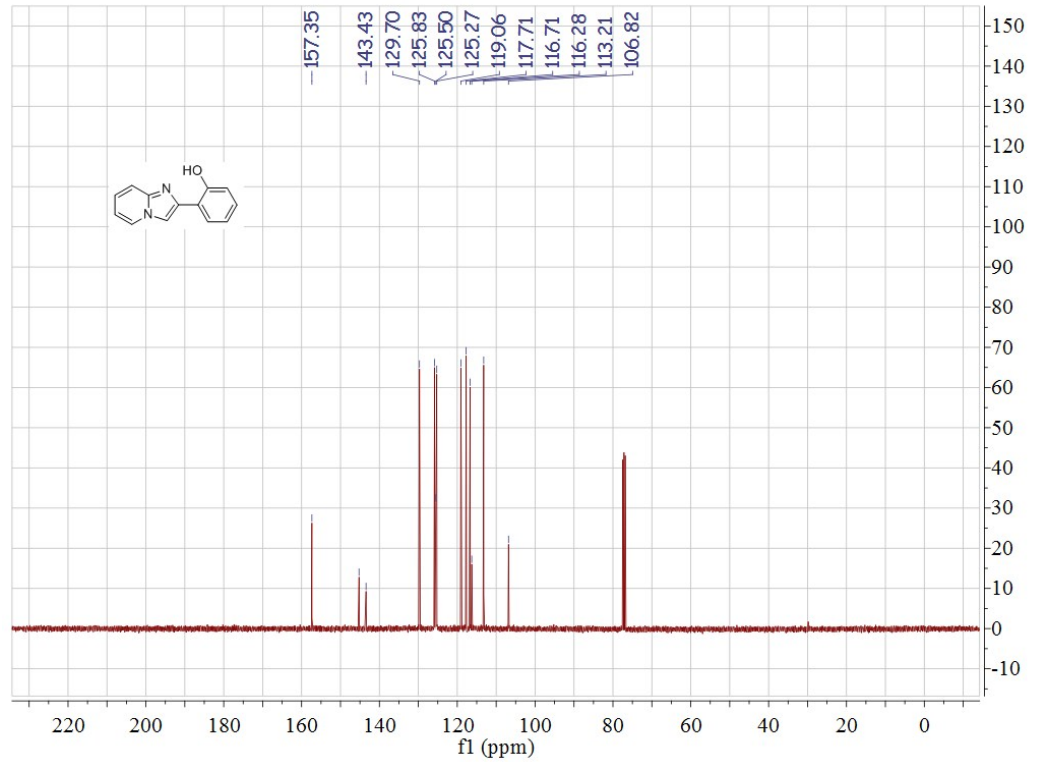
^{13}C NMR of compound **3ka**



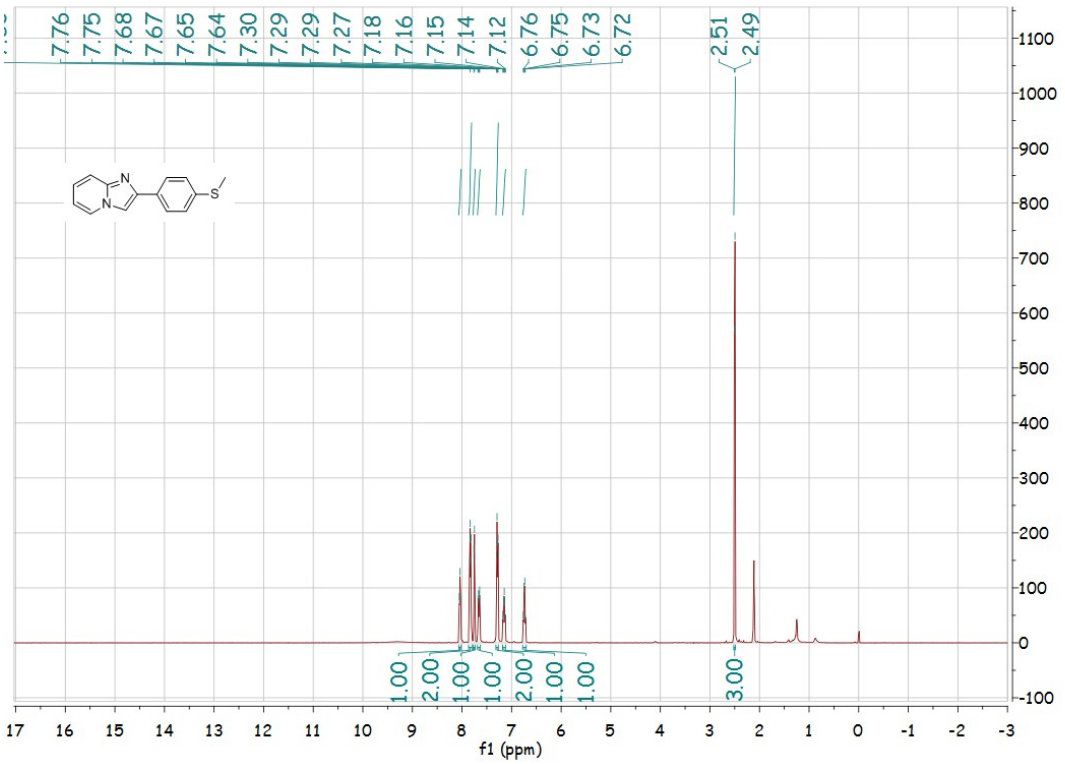
^{1}H NMR of compound **3la**



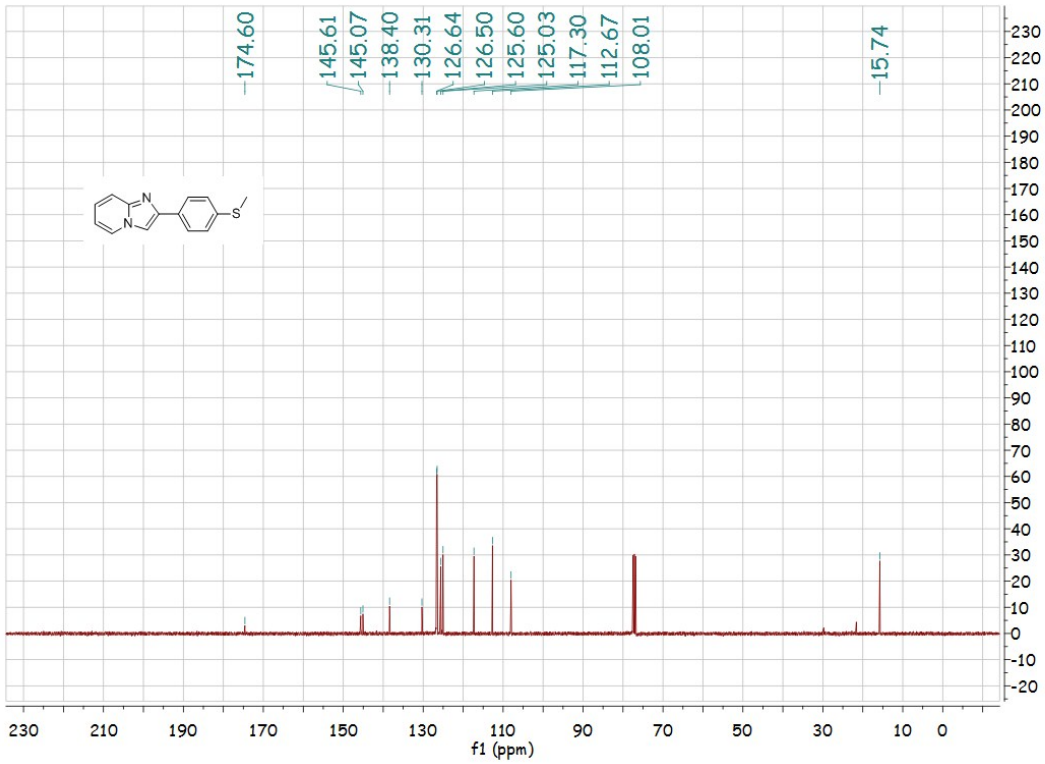
^{13}C NMR of compound **3la**



^{1}H NMR of compound **3ma**



^{13}C NMR of compound **3ma**



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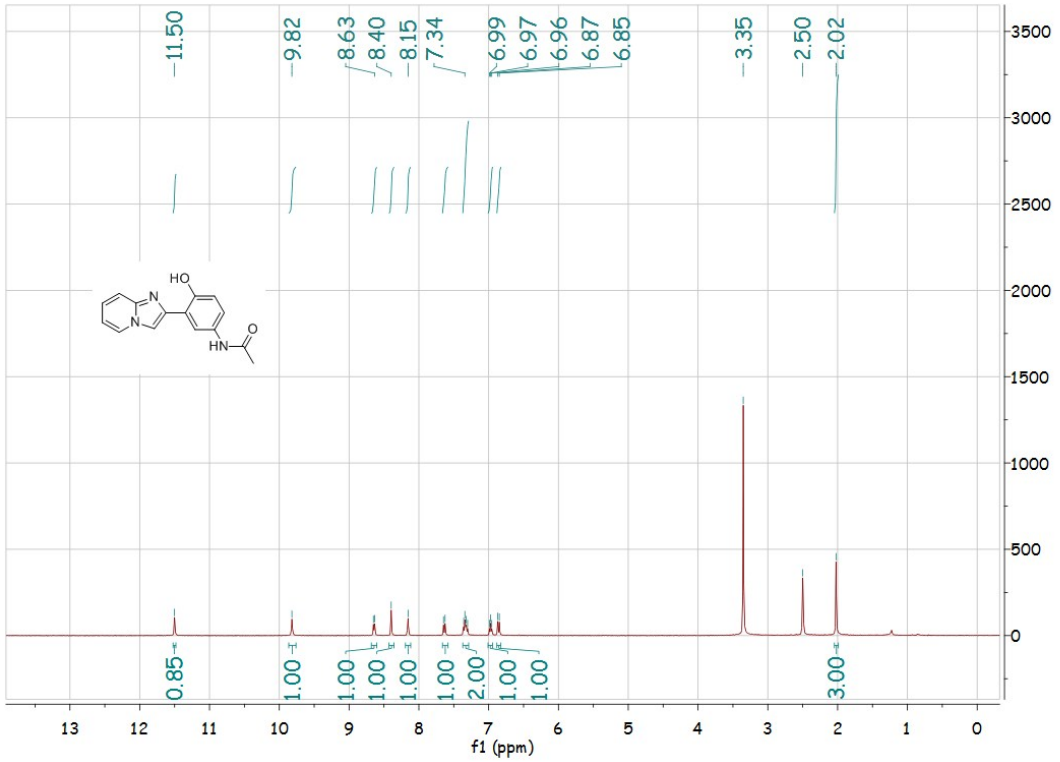
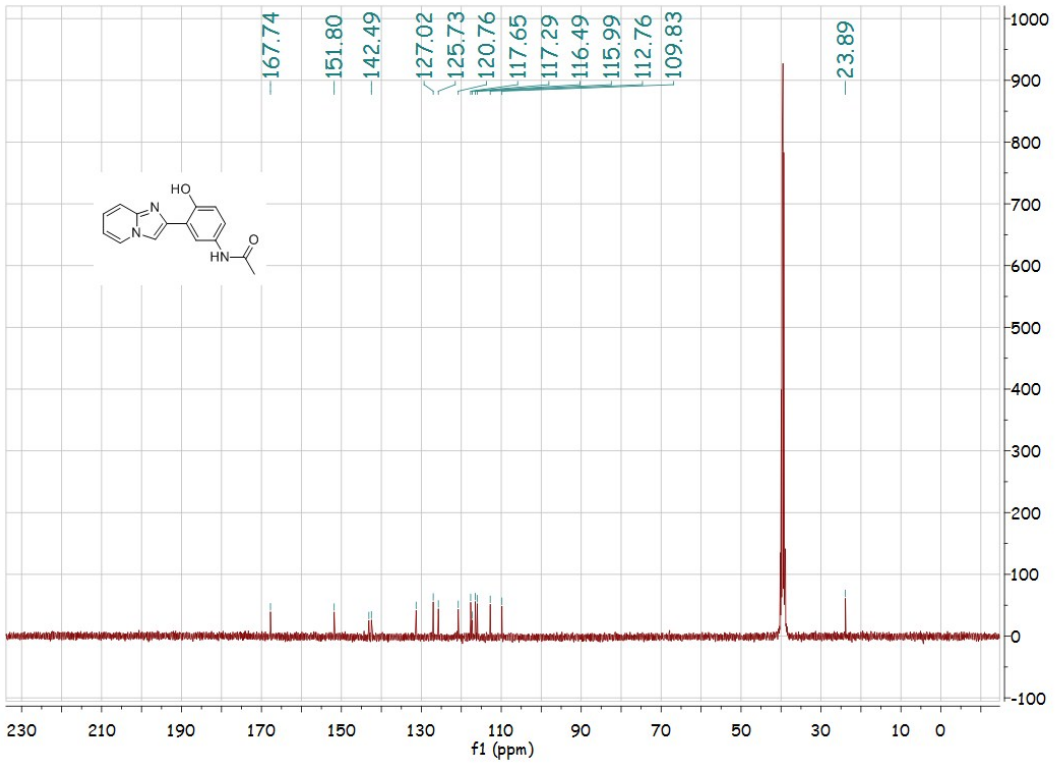
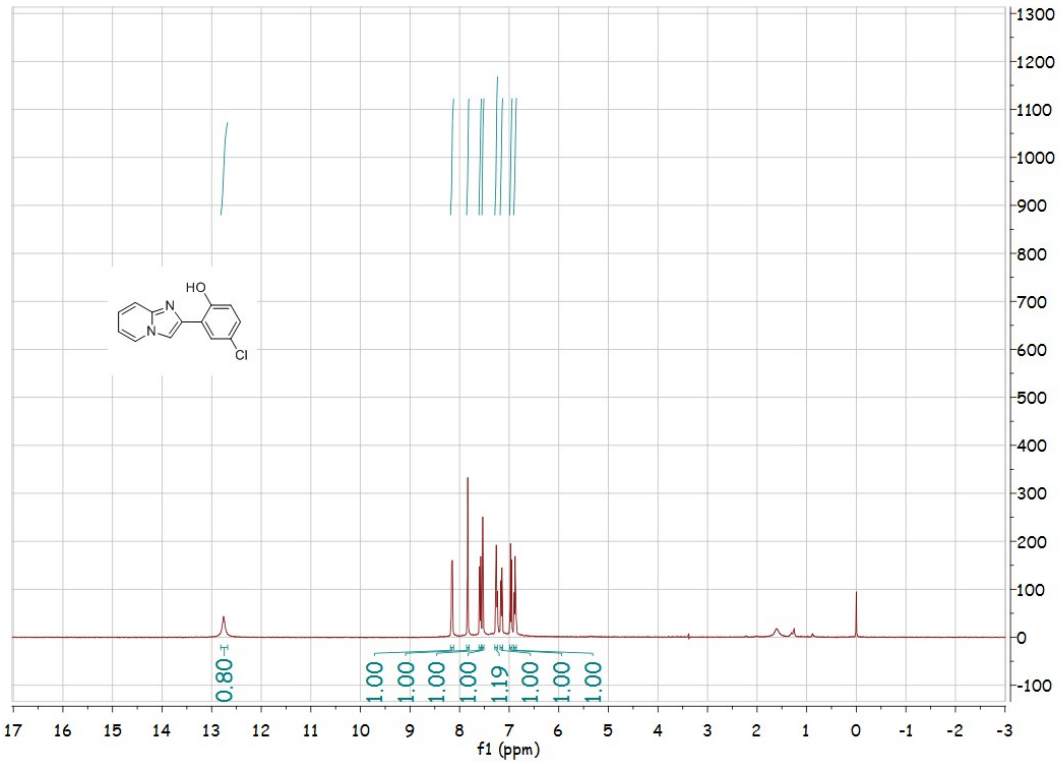


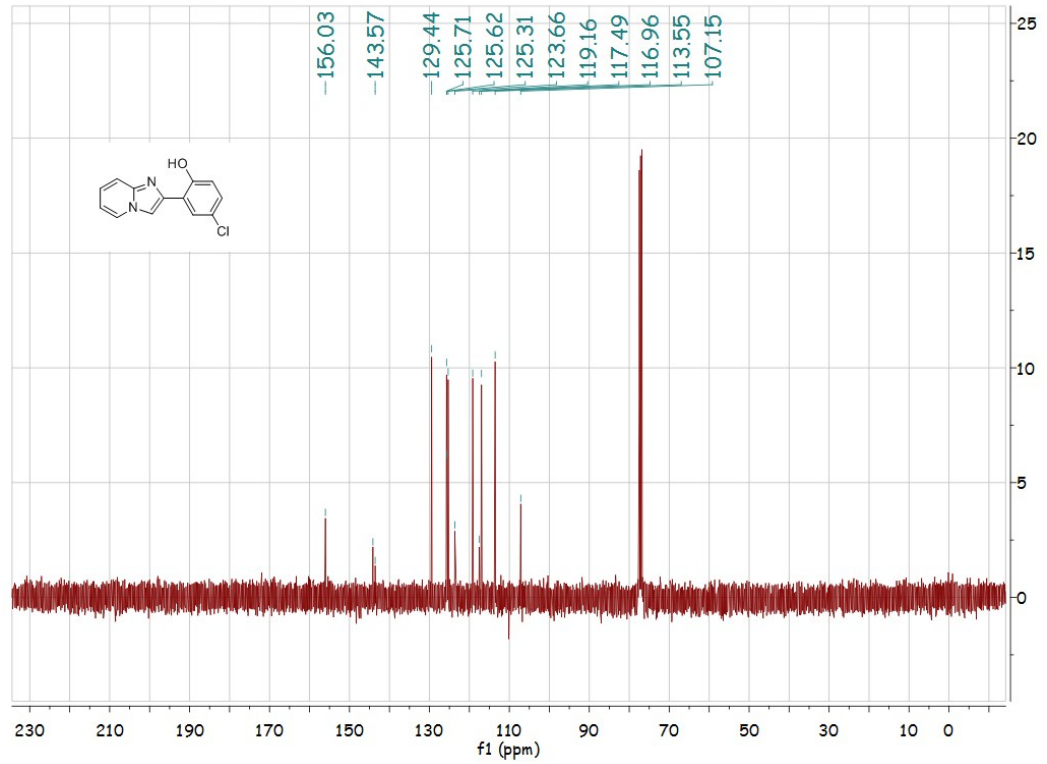
^{13}C NMR of compound **3na**



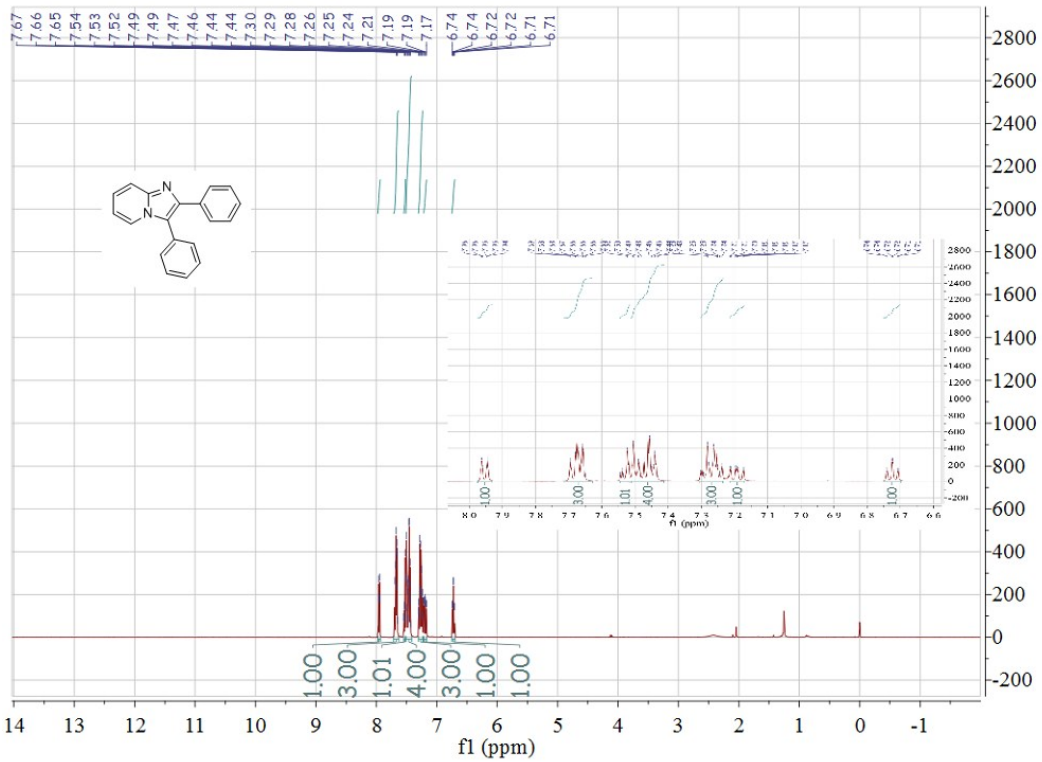
^{1}H NMR of compound **3oa**



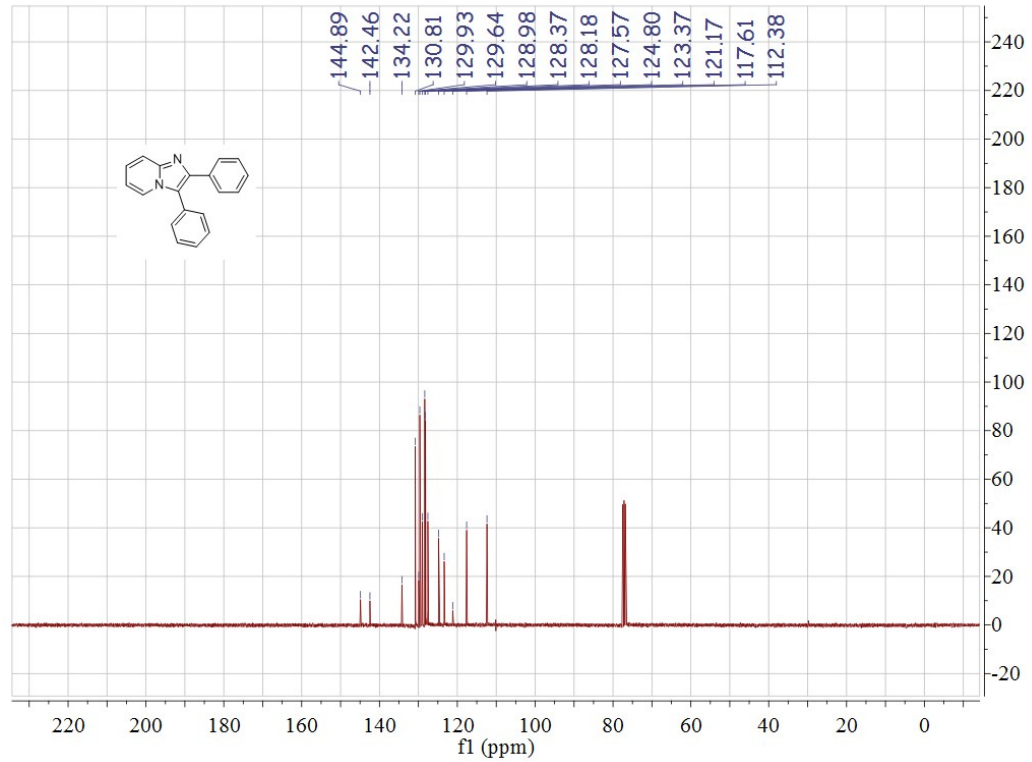
^{13}C NMR of compound **3oa**



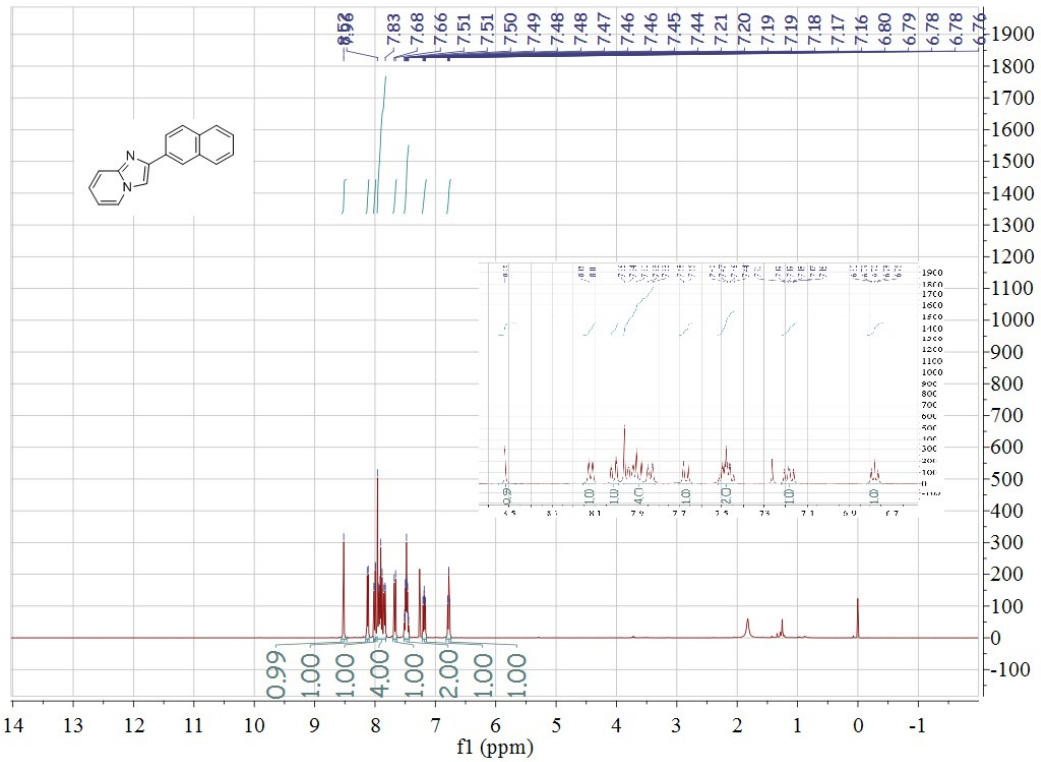
^{1}H NMR of compound **3pa**



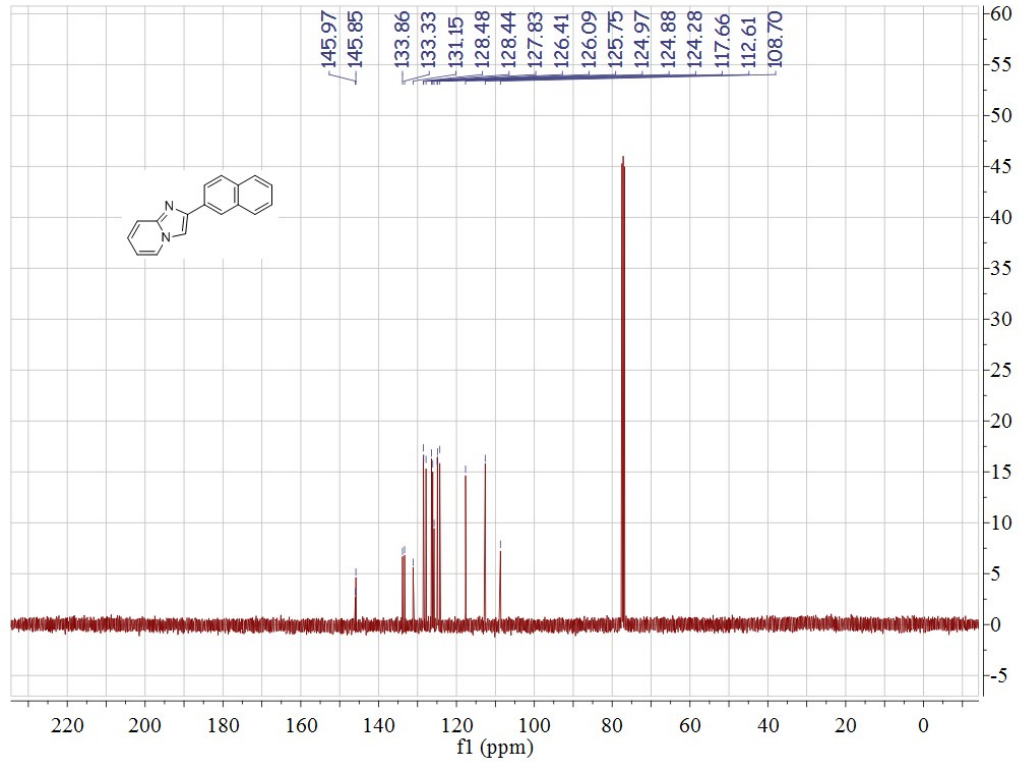
^{13}C NMR of compound **3pa**



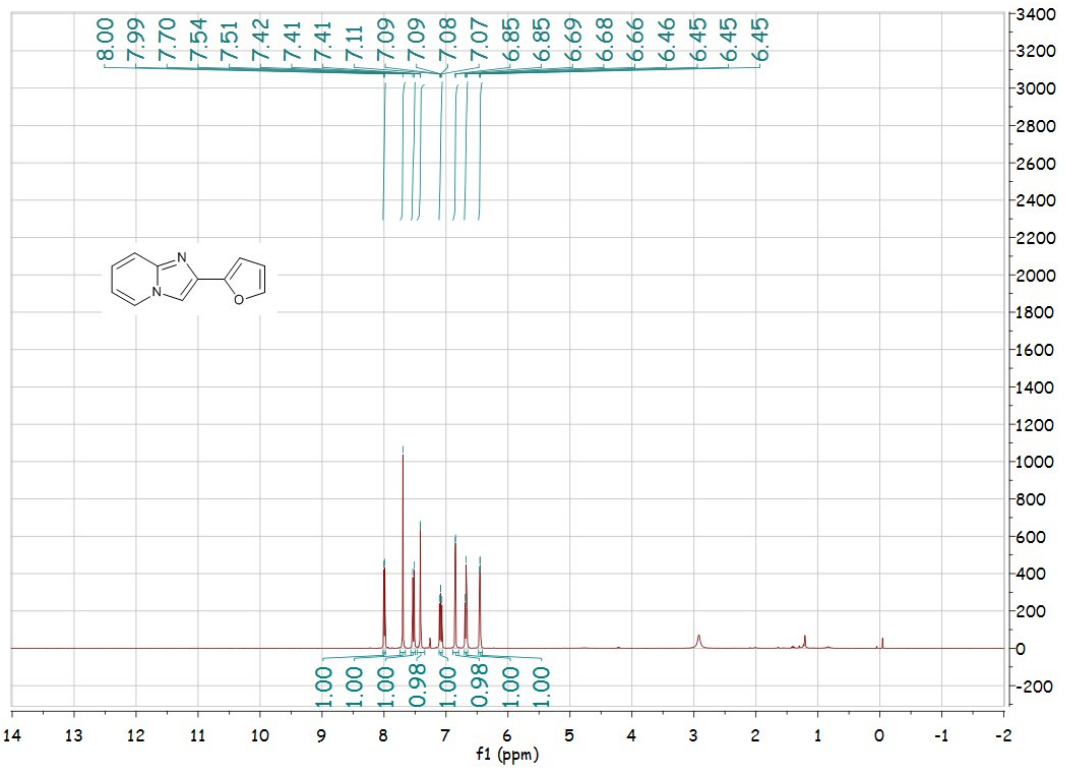
^{1}H NMR of compound **3qa**



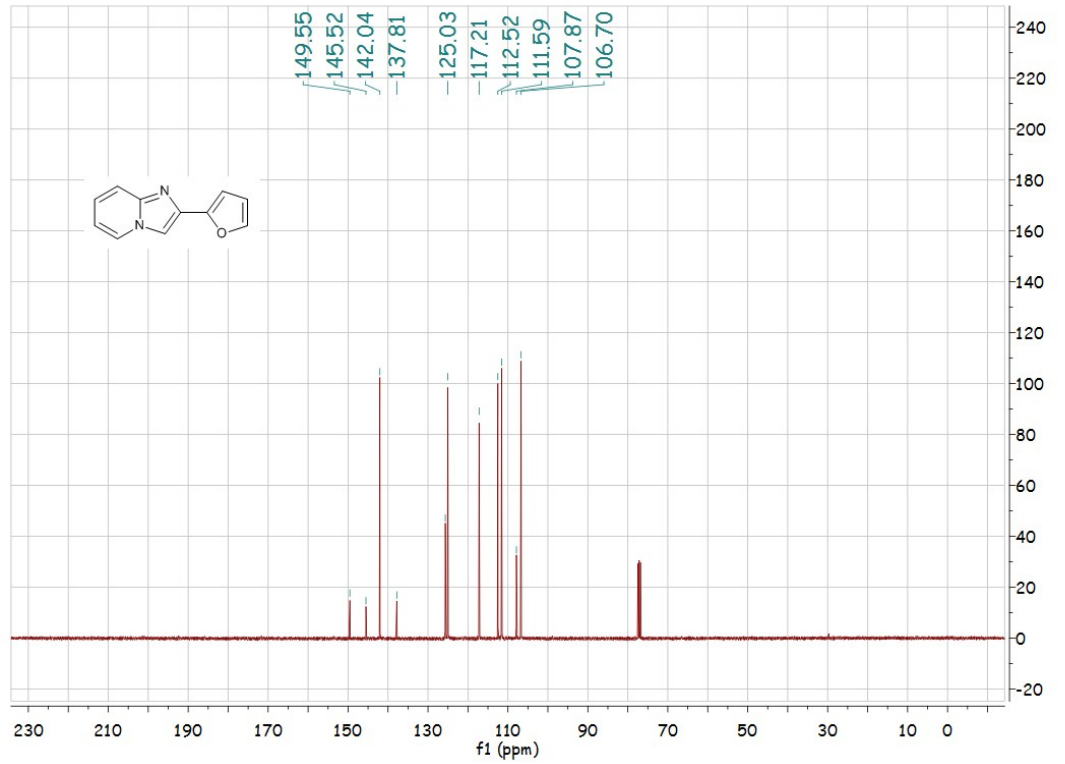
^{13}C NMR of compound **3qa**



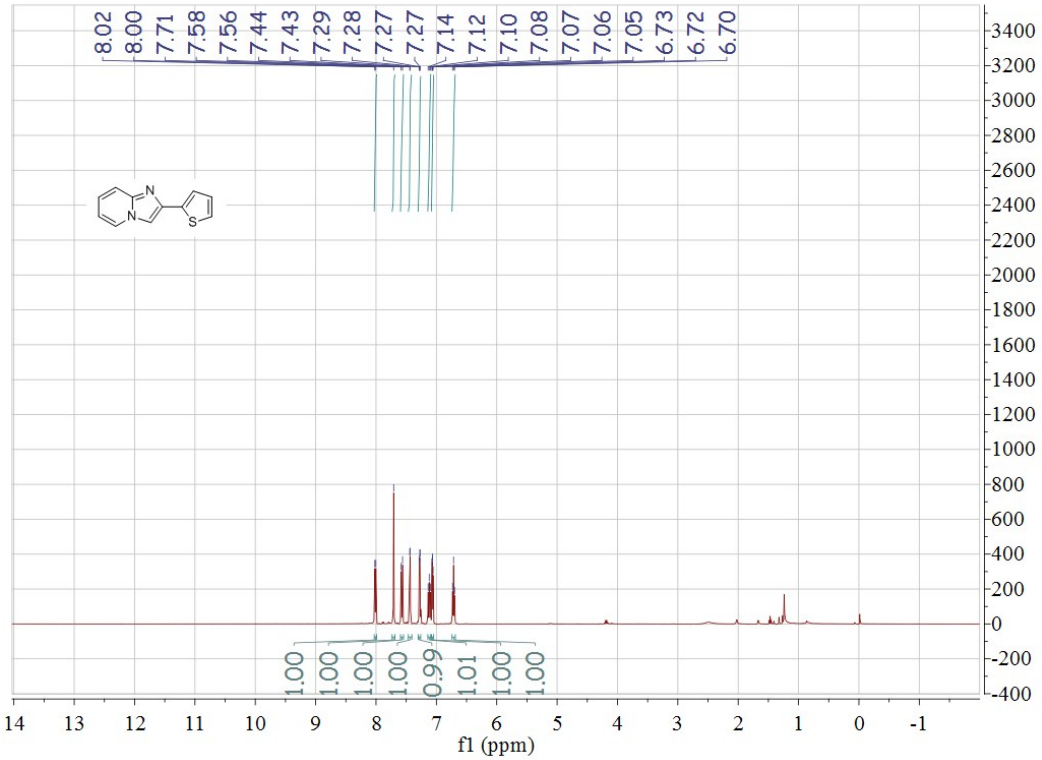
^{1}H NMR of compound **3ra**



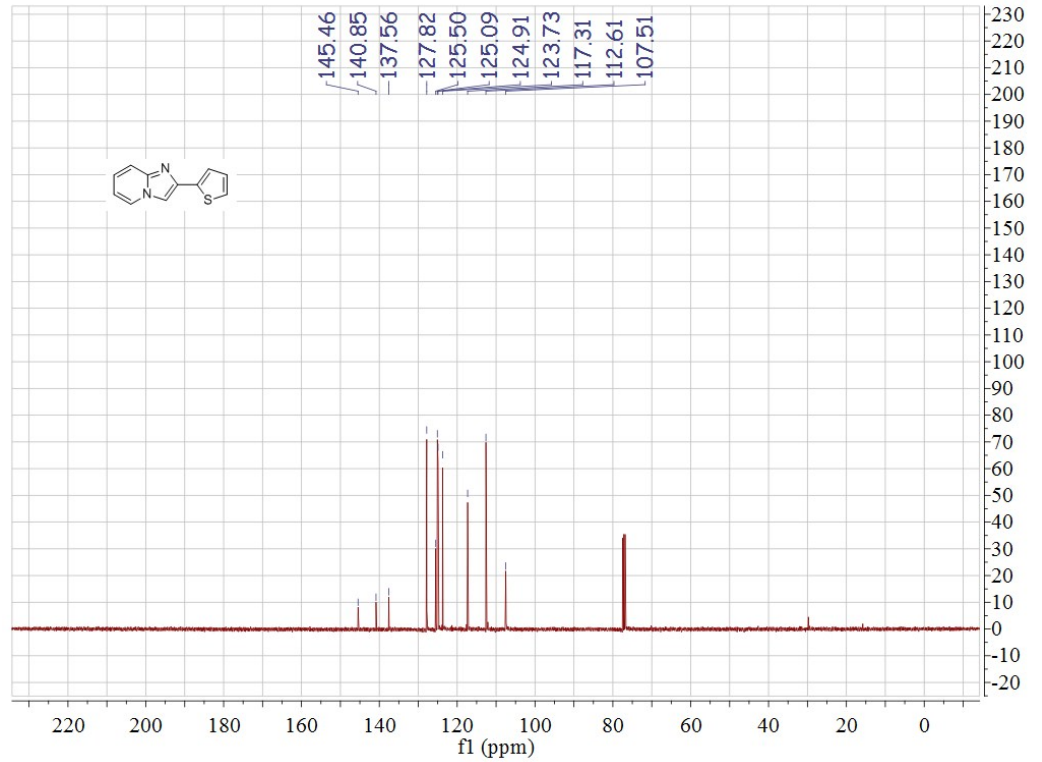
^{13}C NMR of compound **3ra**



^{1}H NMR of compound **3sa**



^{13}C NMR of compound **3sa**



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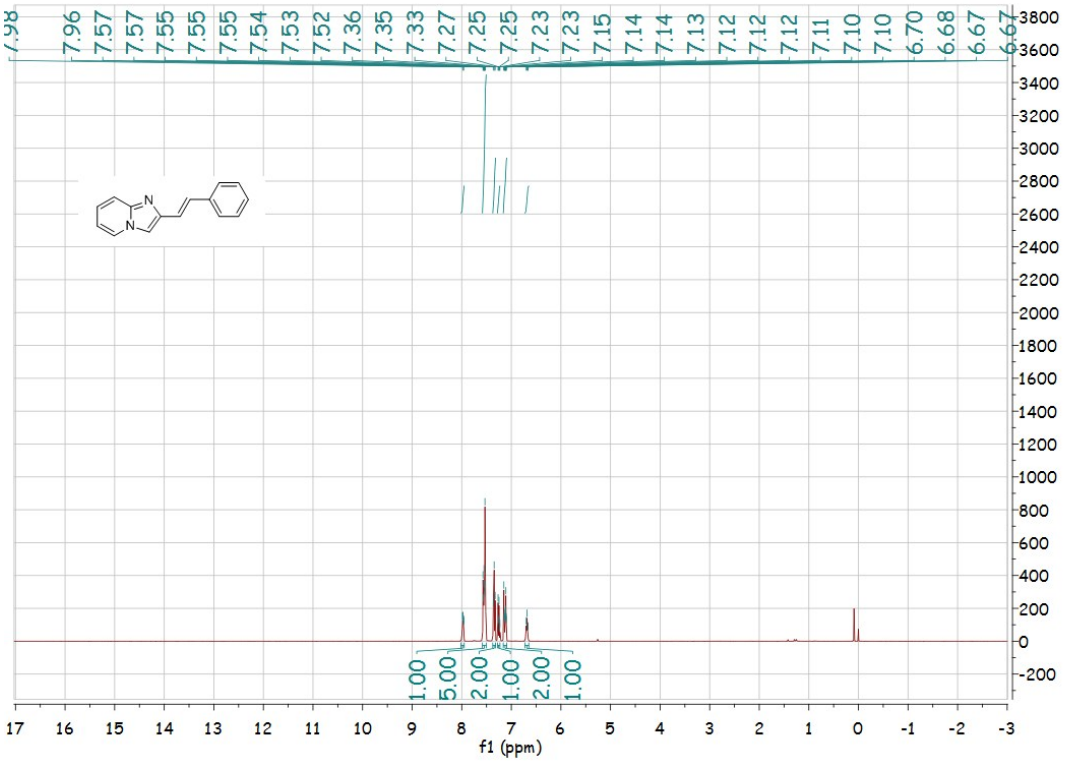
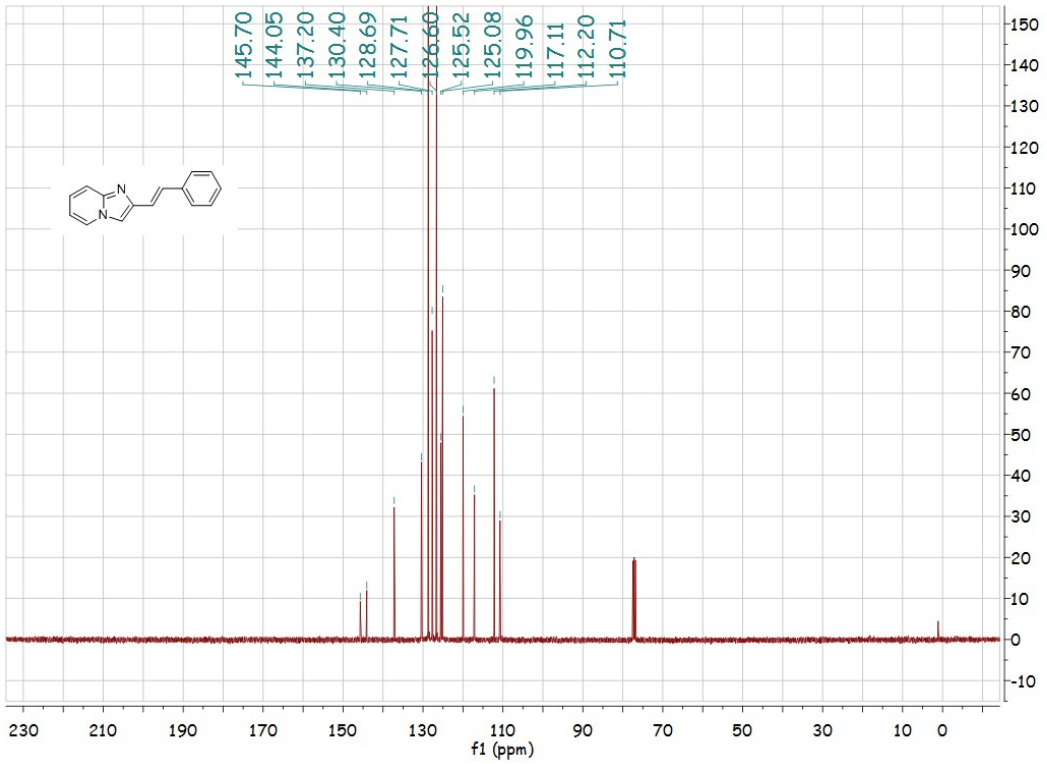


^{13}C NMR of compound **3ta**



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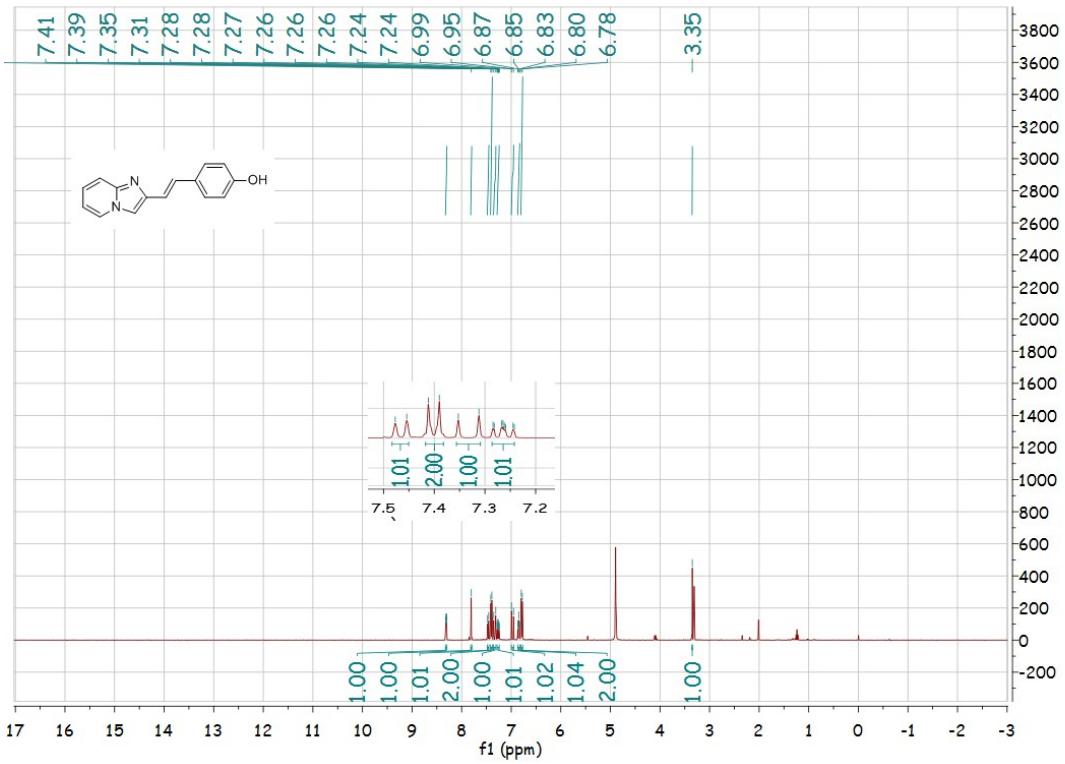

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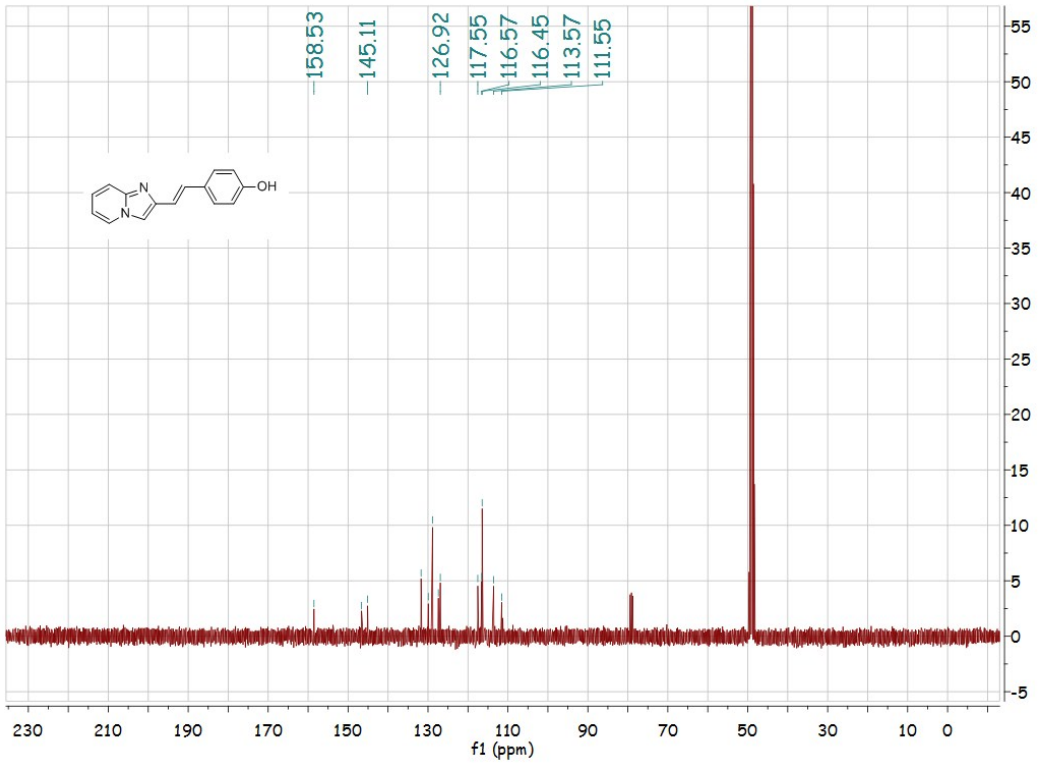
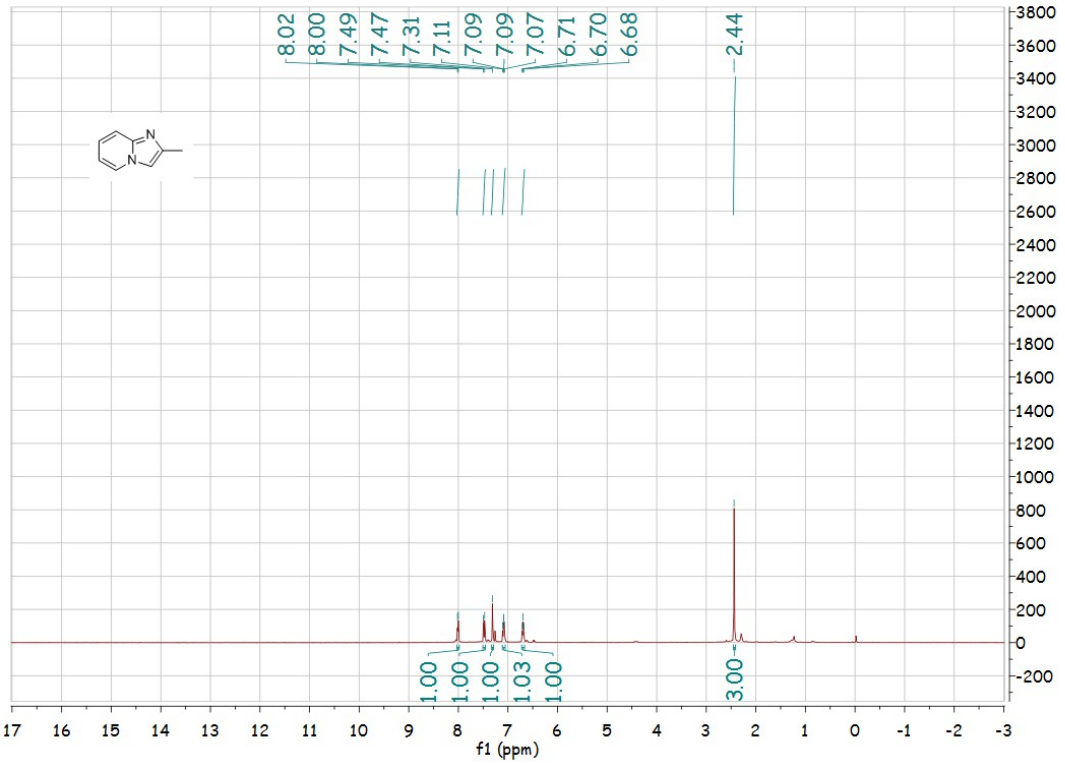
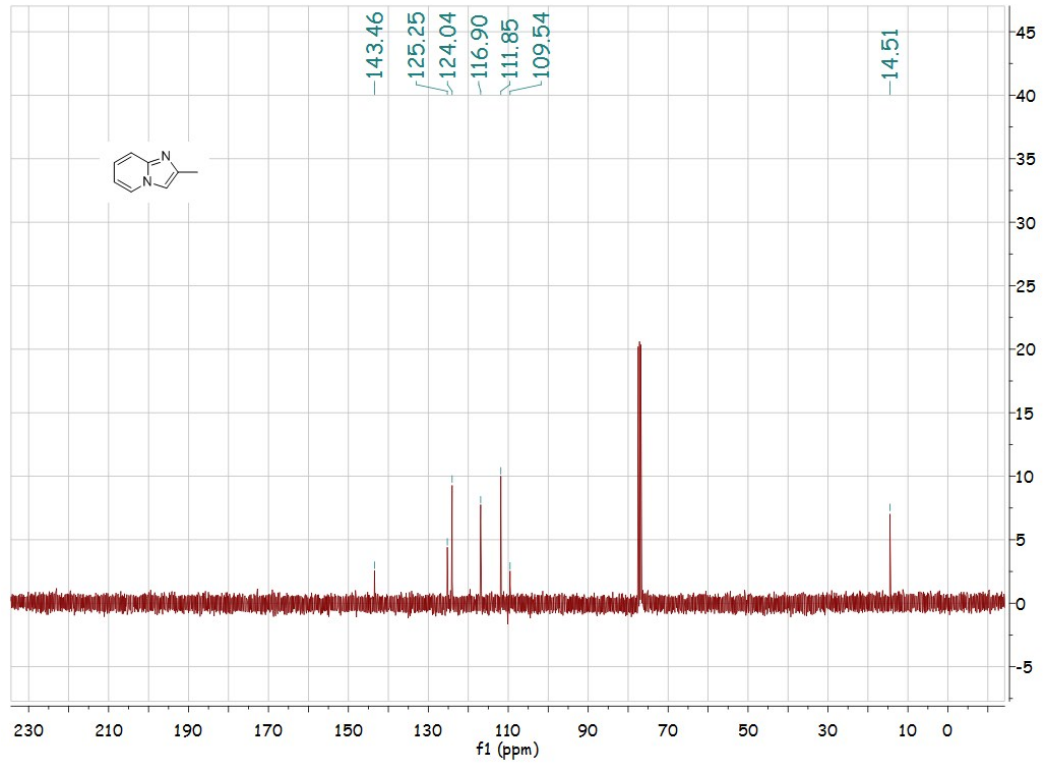


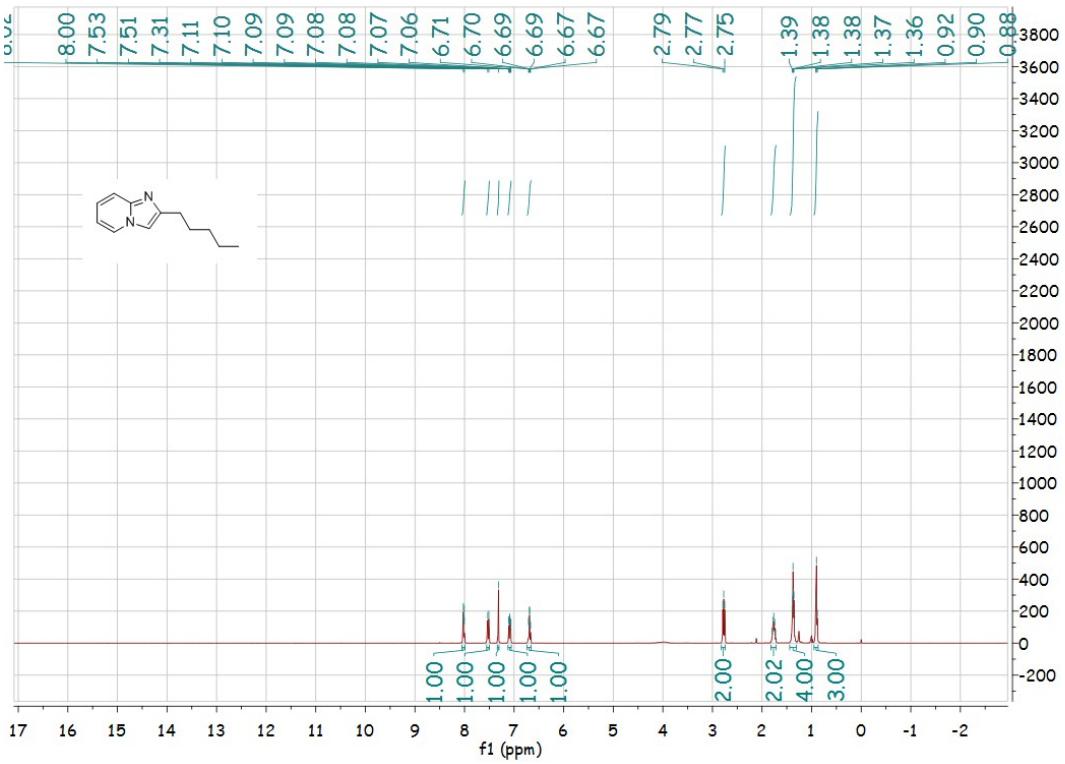
^{1}H NMR of compound **3va**



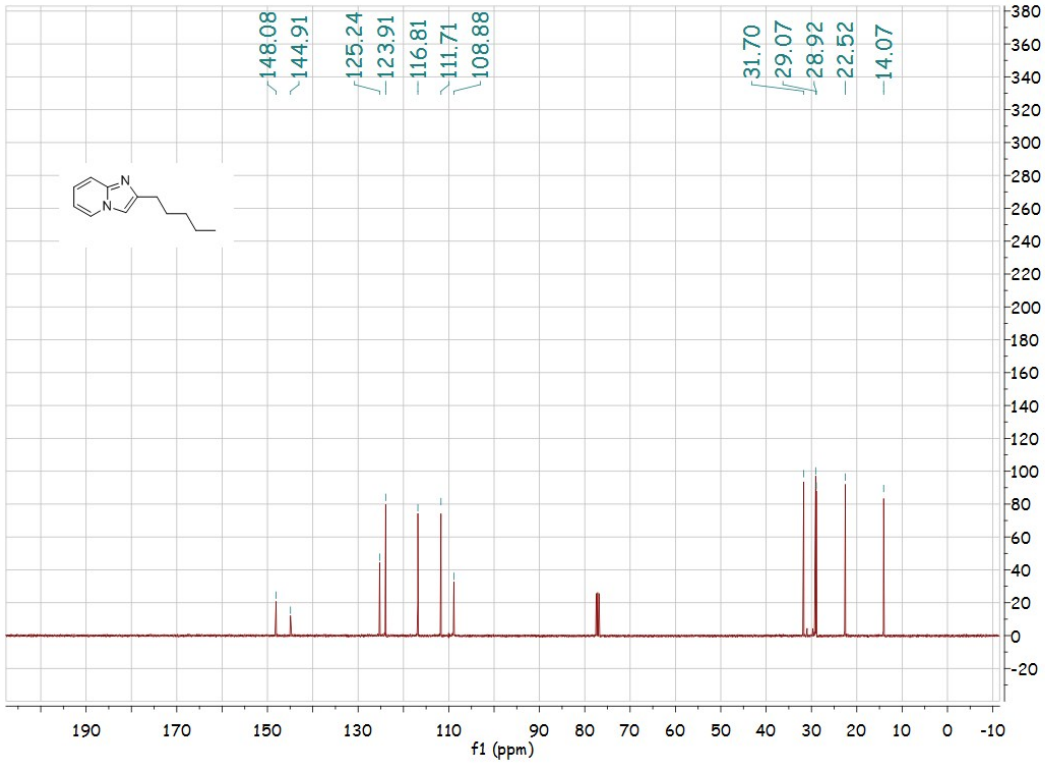
^{13}C NMR of compound **3va**



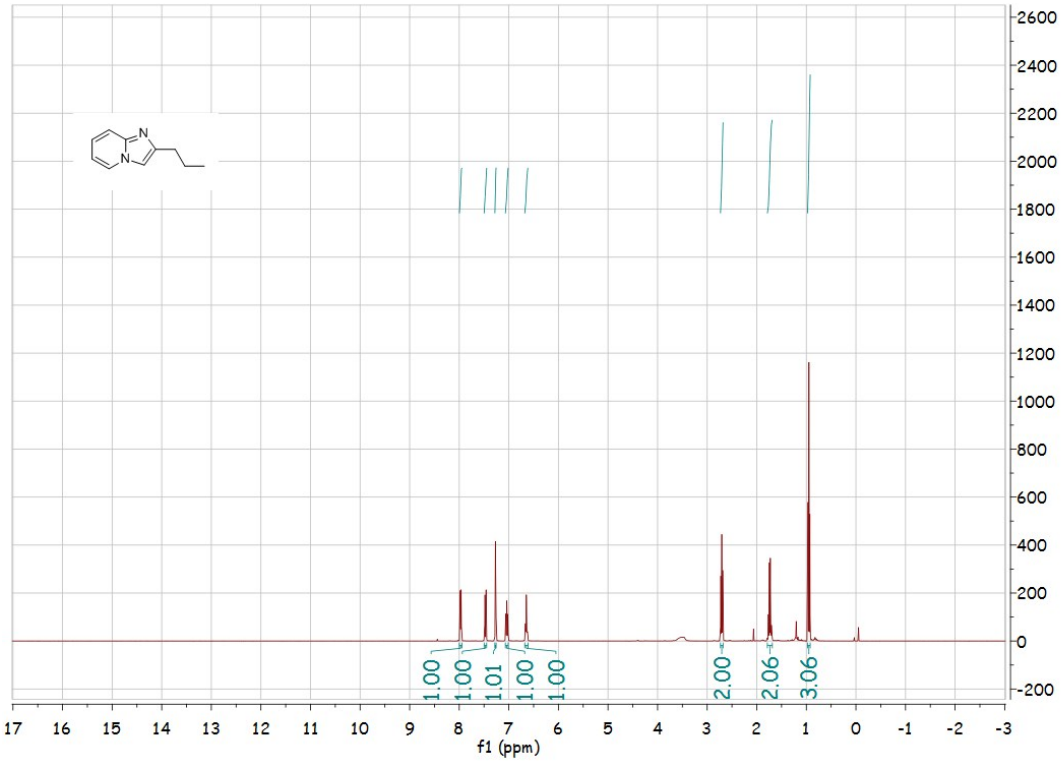
^{1}H NMR of compound **3wa**



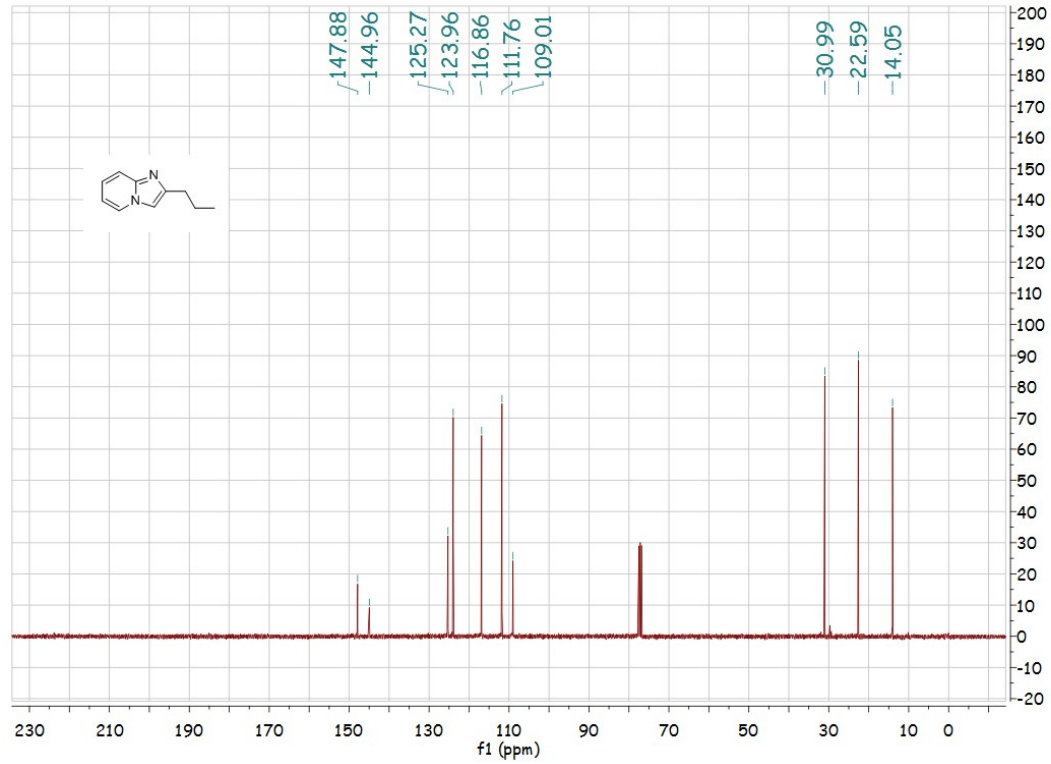
^{13}C NMR of compound **3wa**



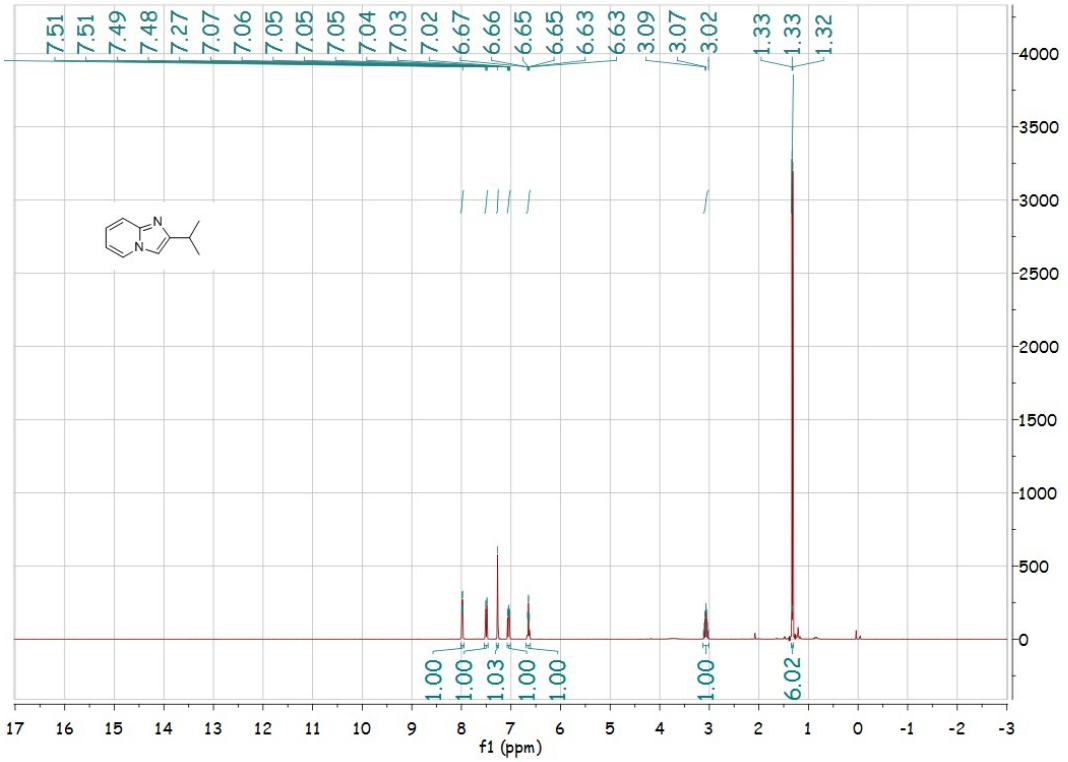
^{1}H NMR of compound **3xa**



^{13}C NMR of compound **3xa**



^{1}H NMR of compound **3ya**



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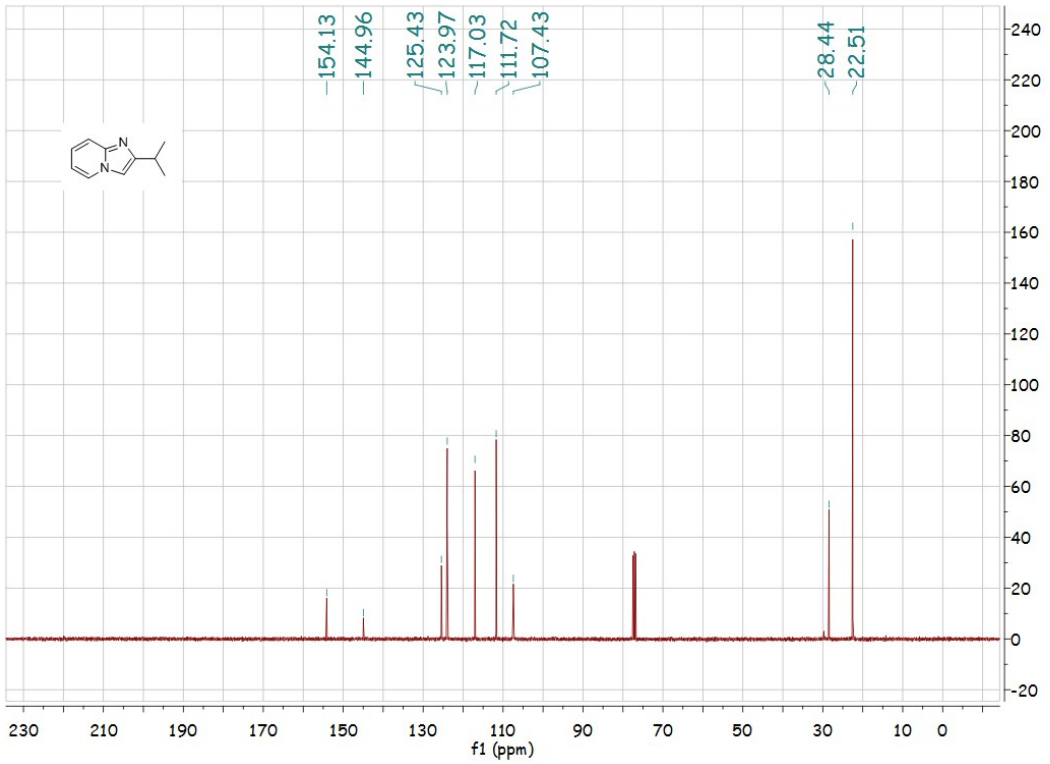
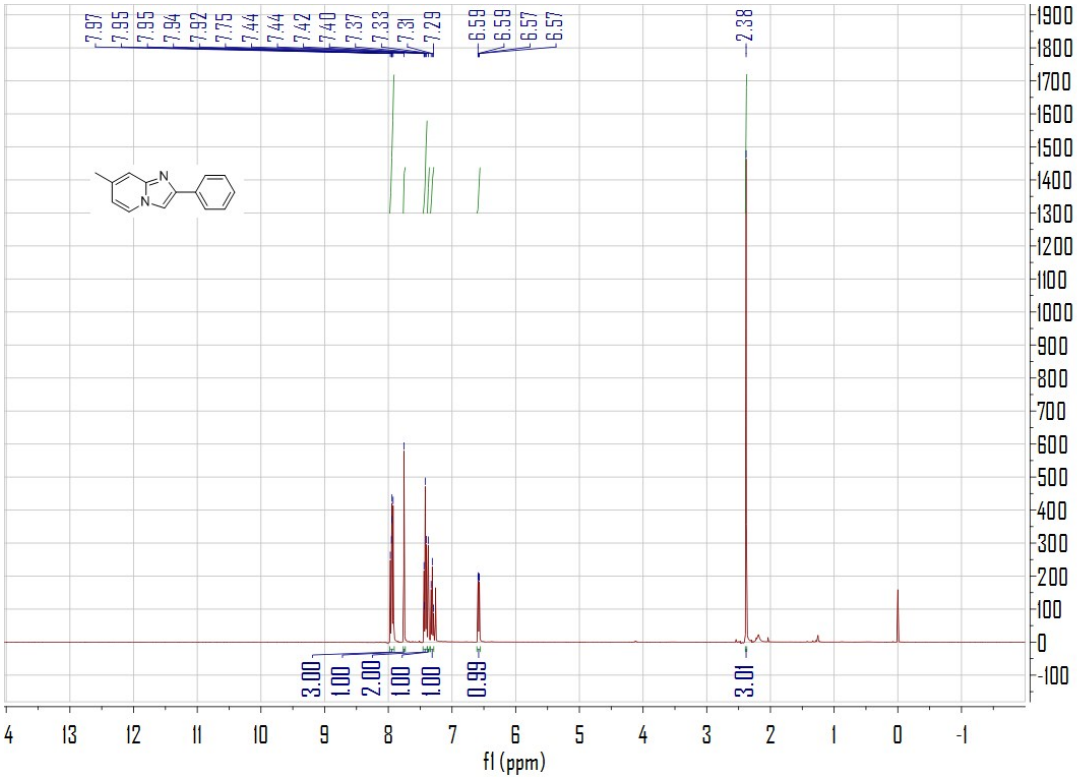
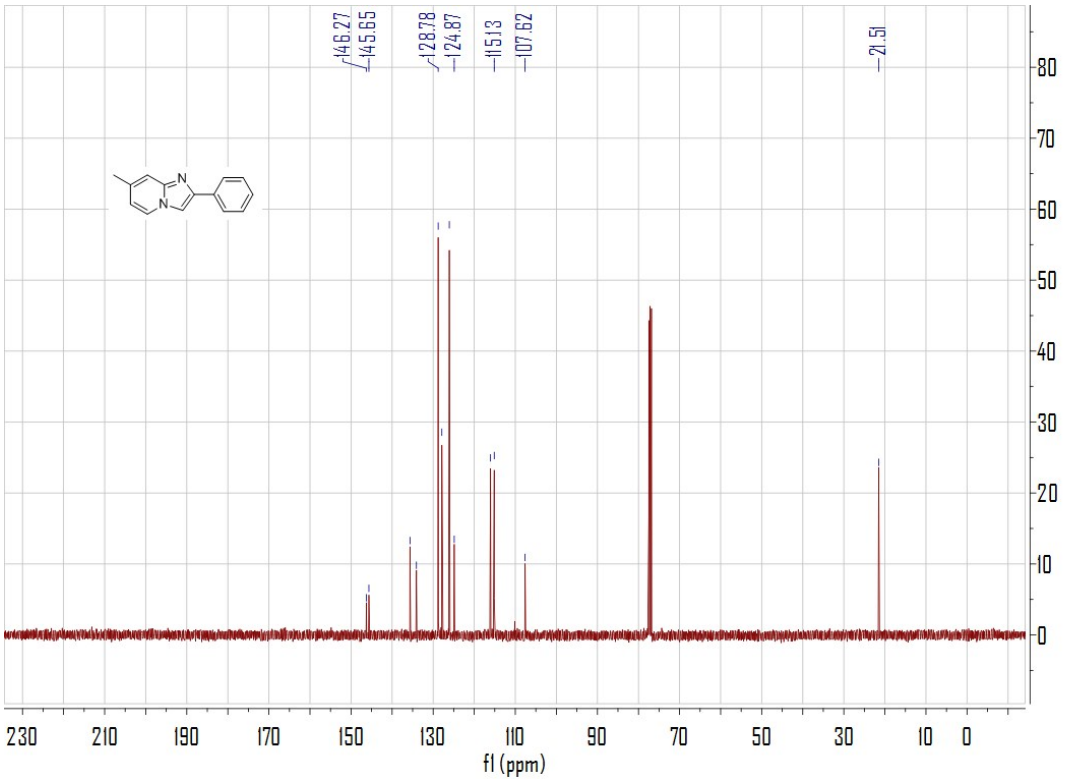


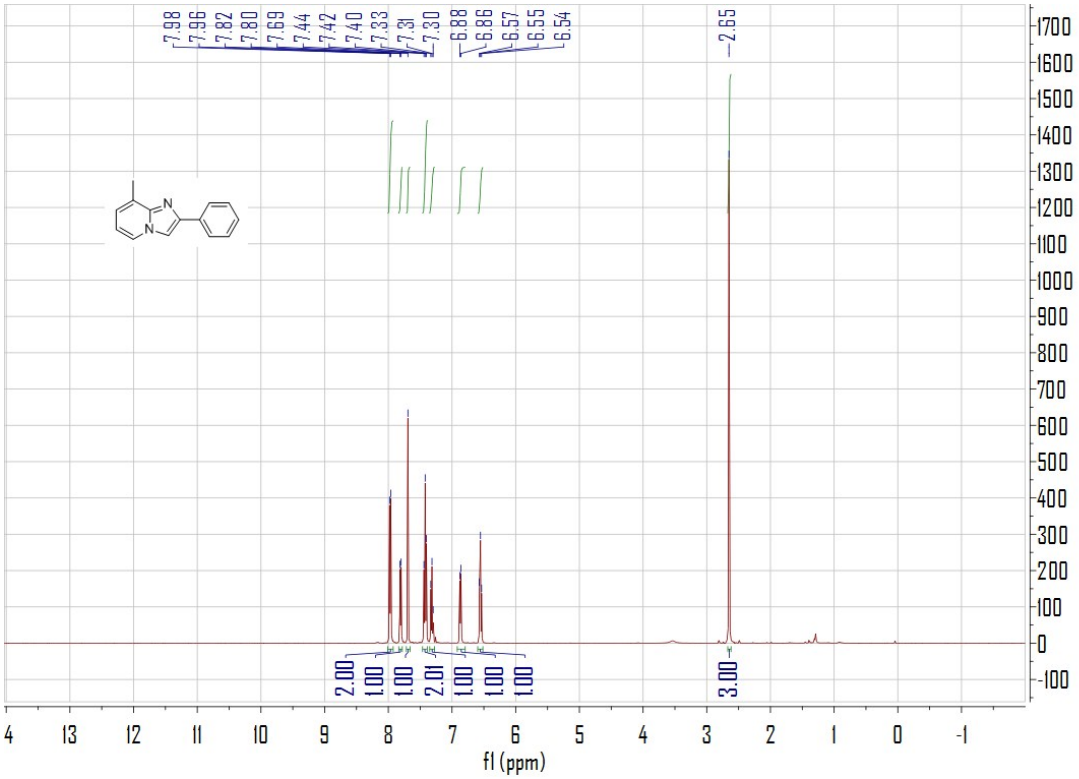
^{1}H NMR of compound **3ab**



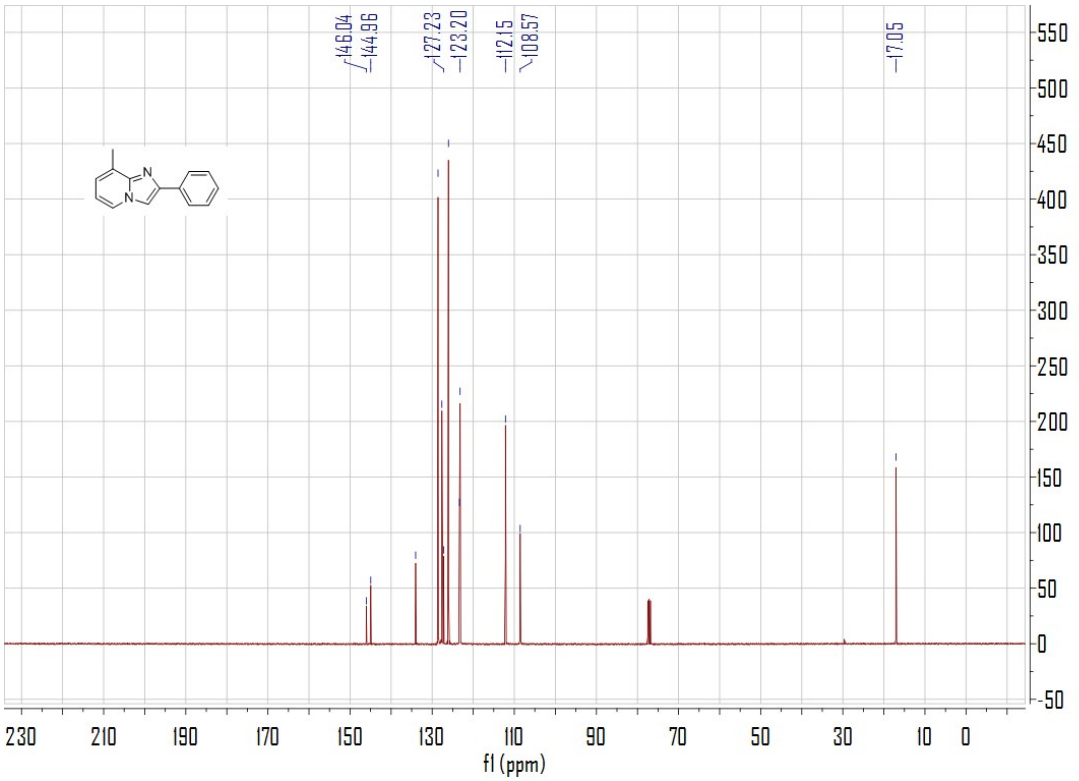
^{13}C NMR of compound **3ab**



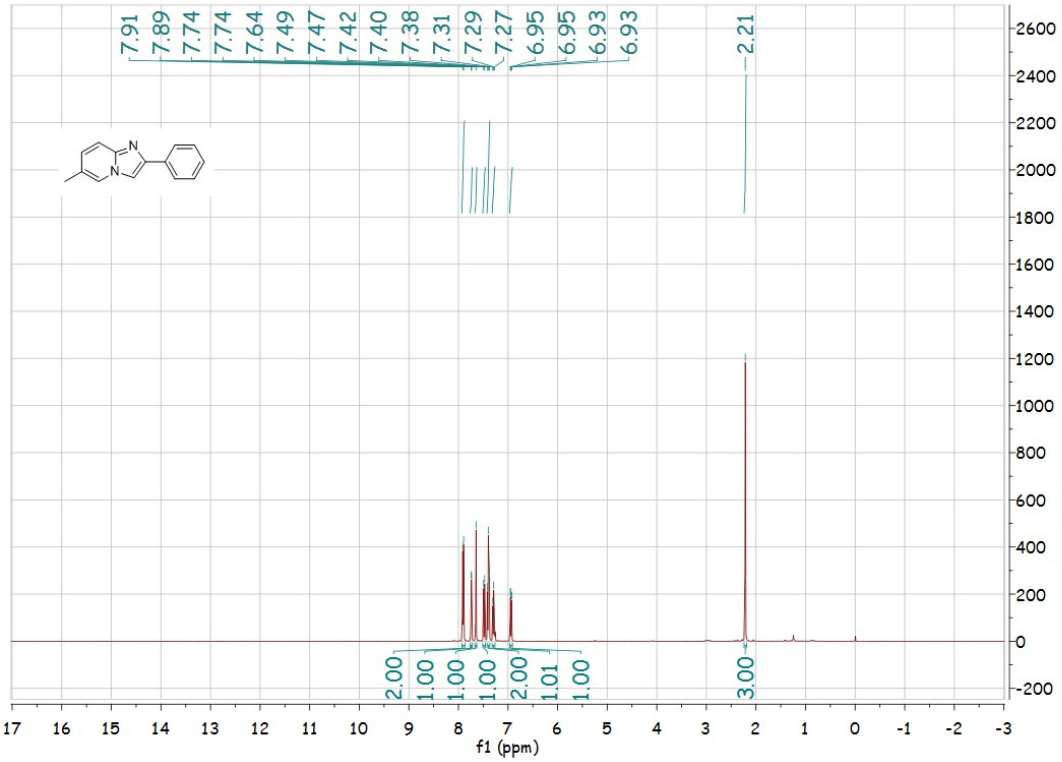
^{1}H NMR of compound **3ac**



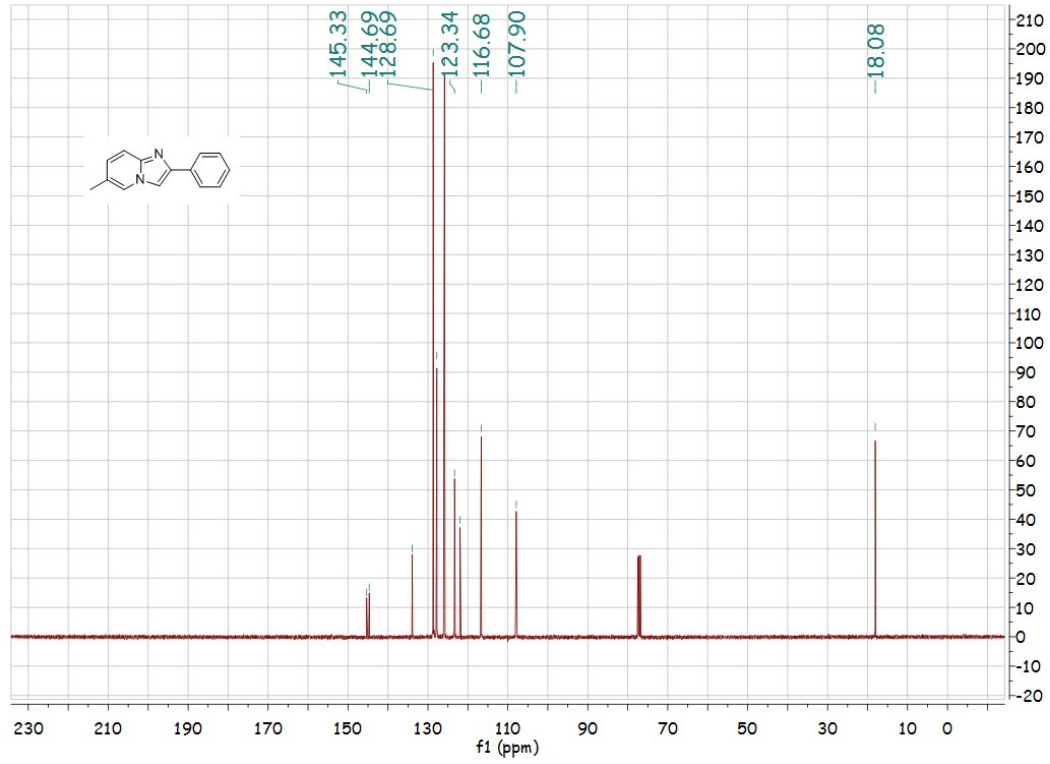
^{13}C NMR of compound **3ac**



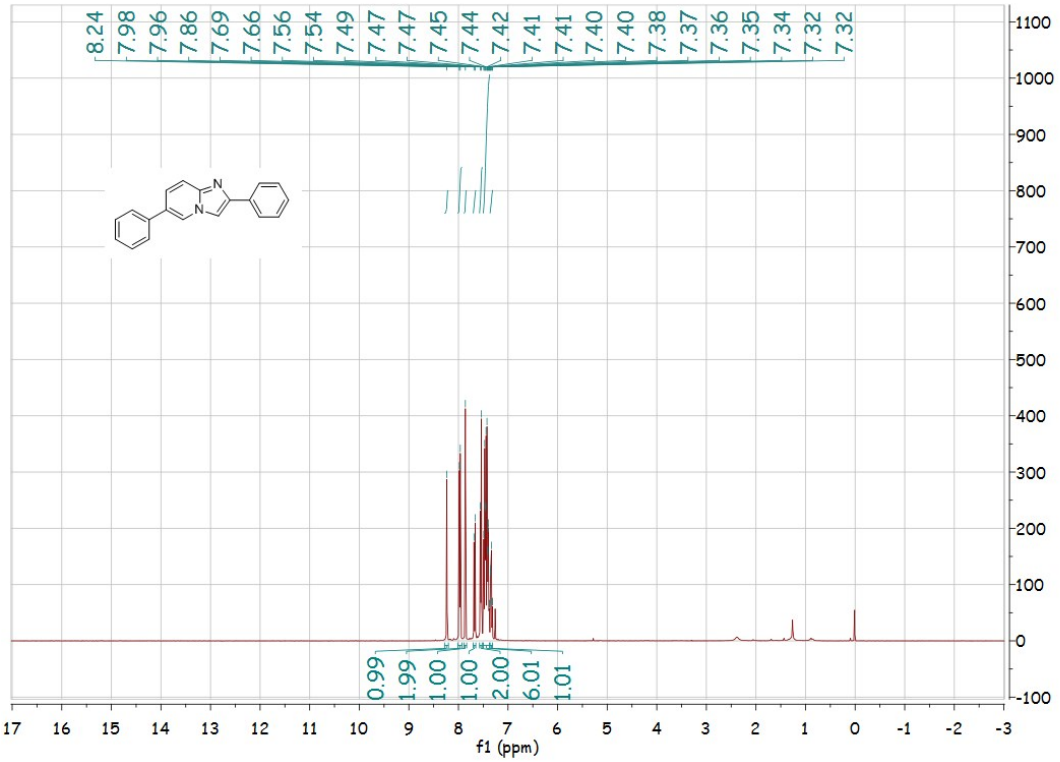
^{1}H NMR of compound **3ad**



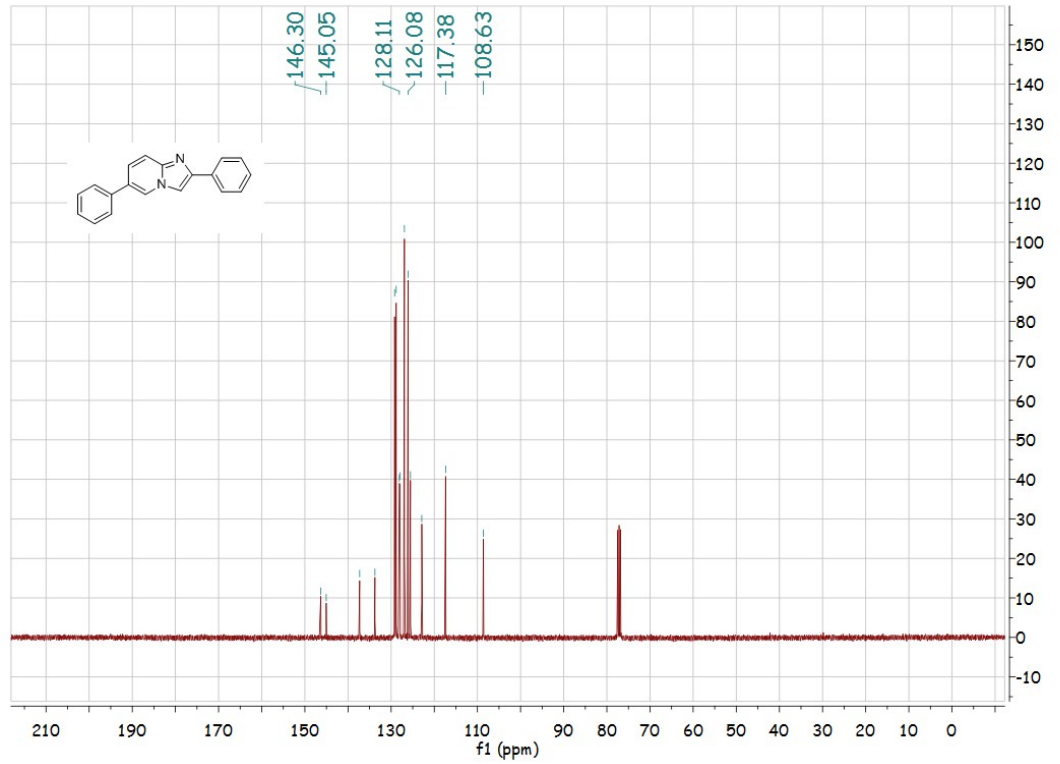
^{13}C NMR of compound **3ad**



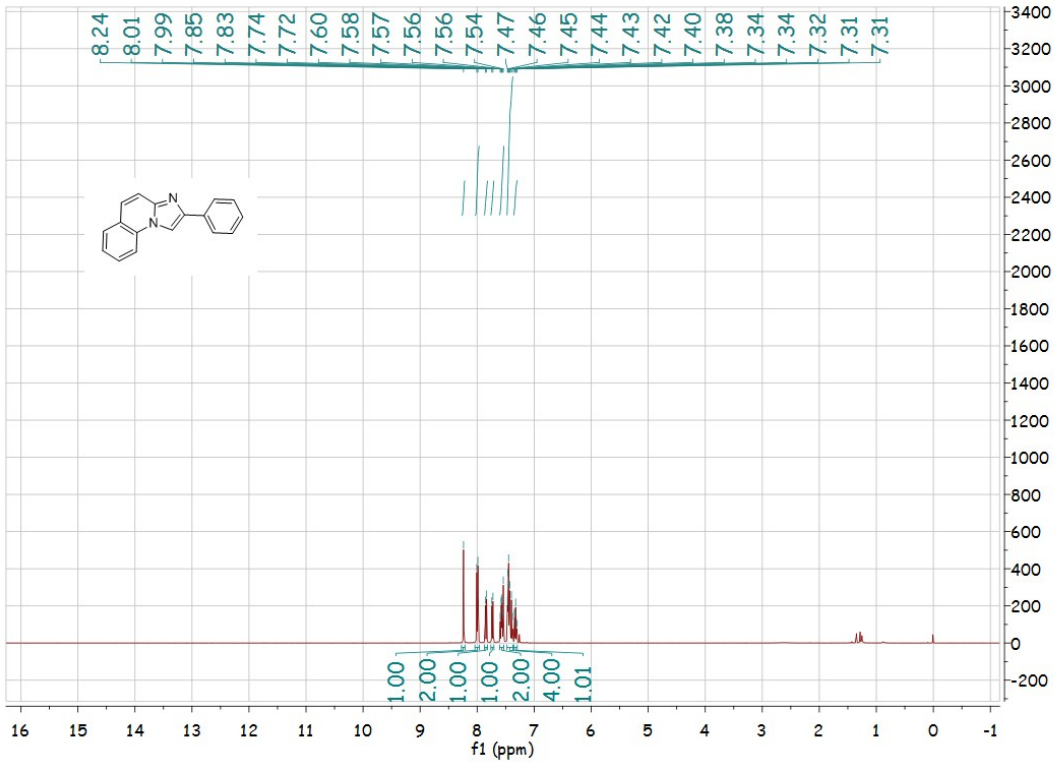
^{1}H NMR of compound **3ae**



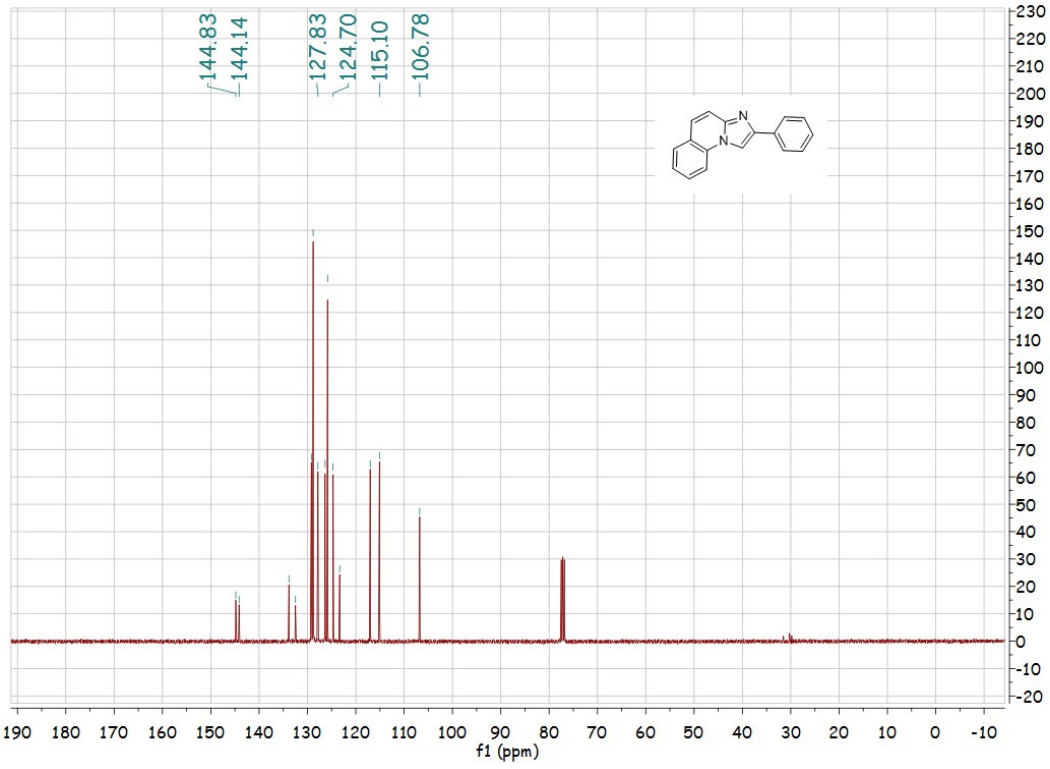
^{13}C NMR of compound **3ae**



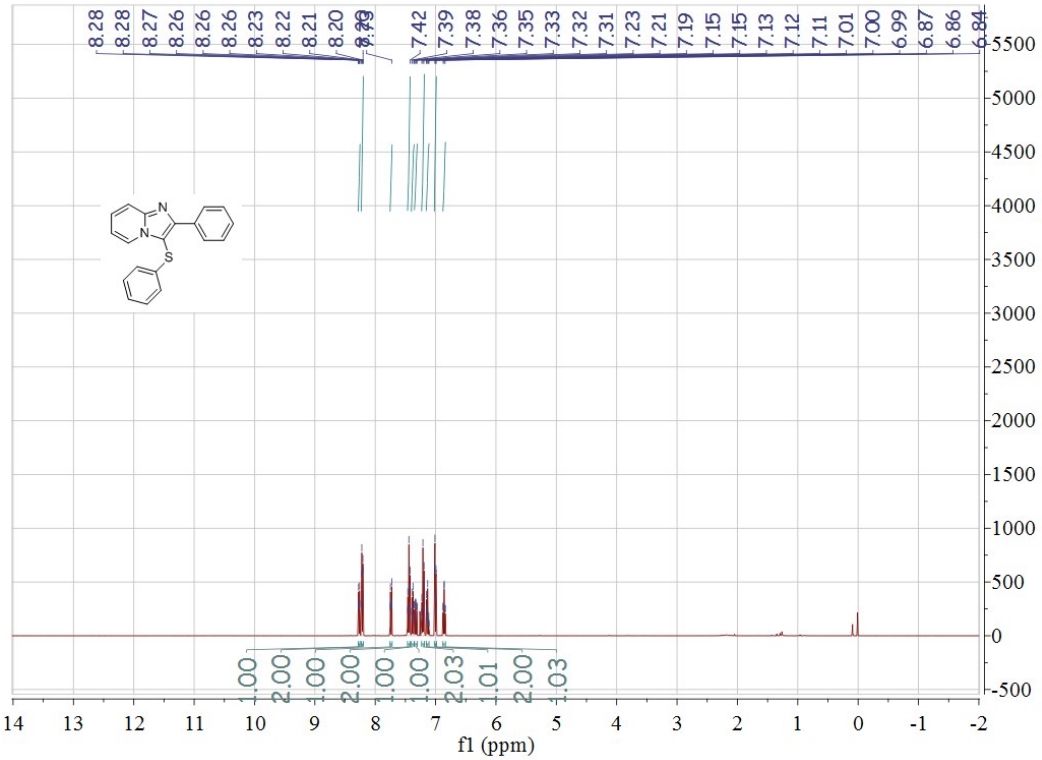
^{1}H NMR of compound **3af**



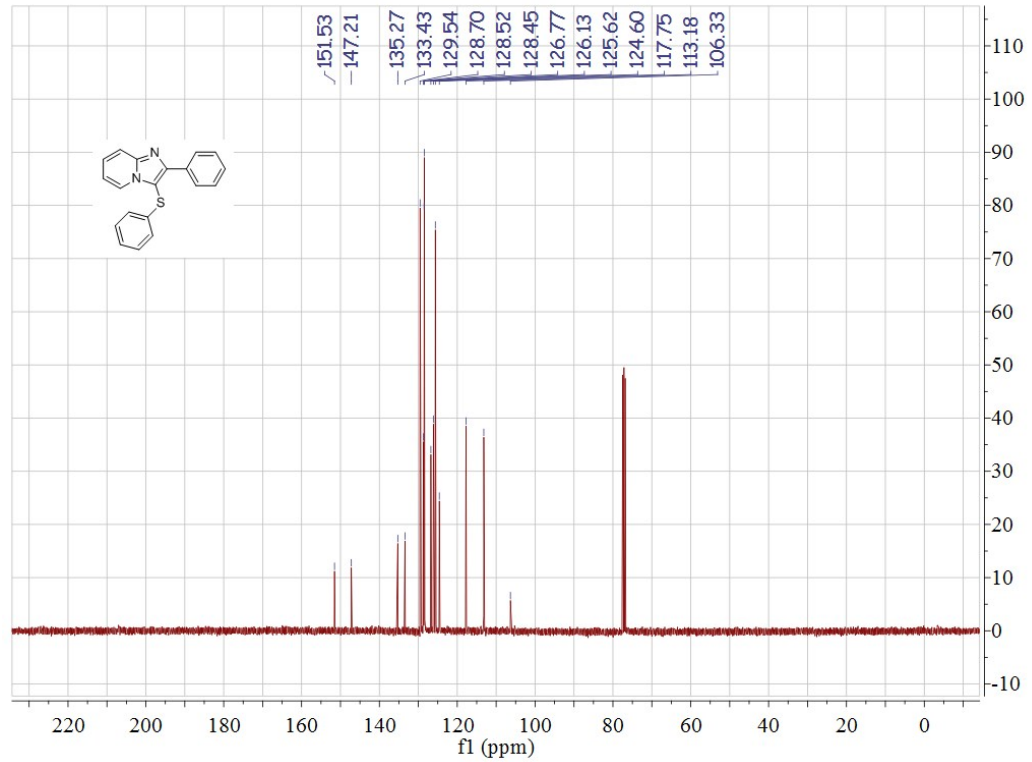
^{13}C NMR of compound **3af**



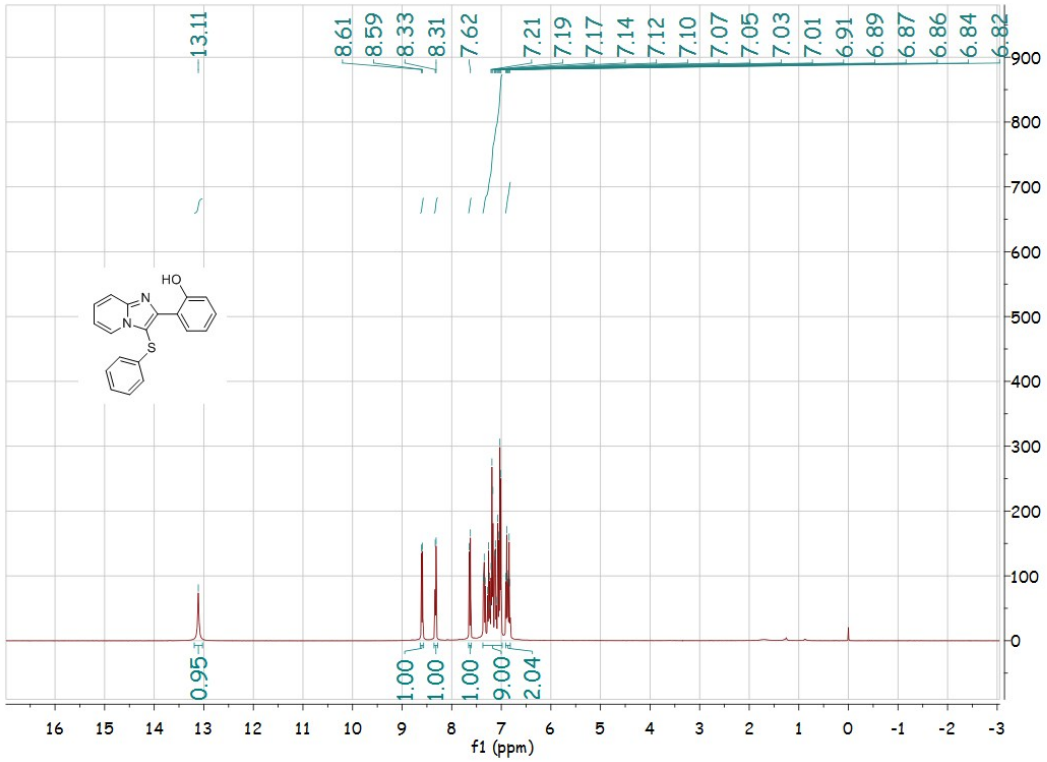
^{1}H NMR of compound **5a**



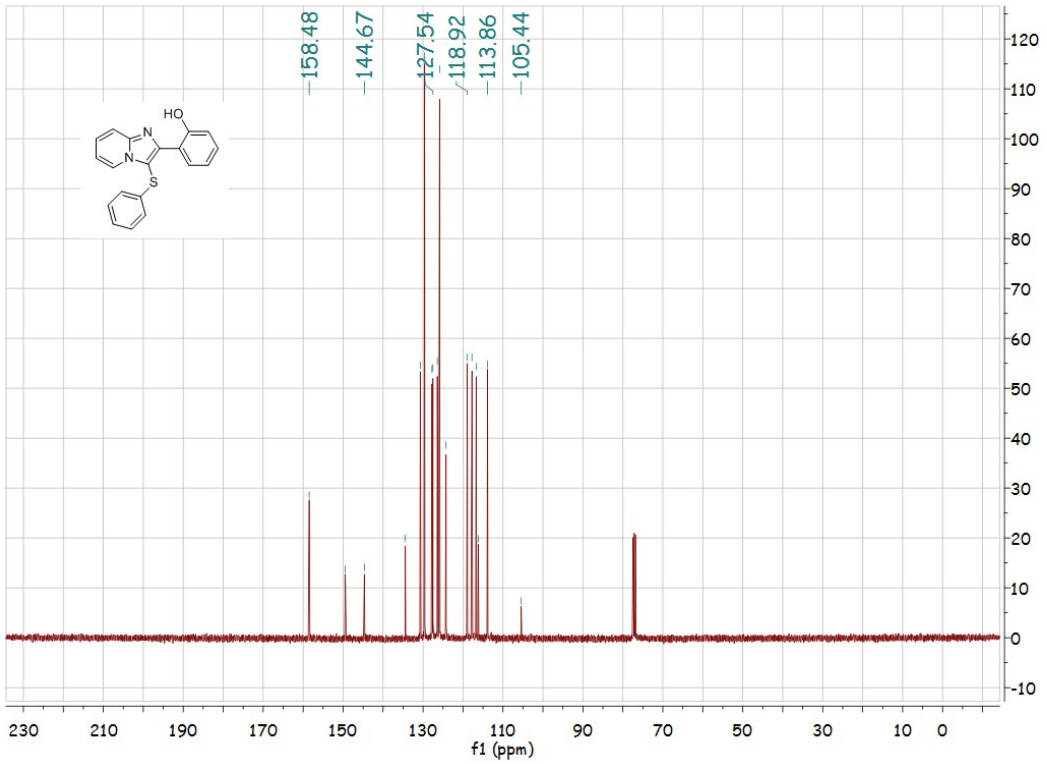
^{13}C NMR of compound **5a**



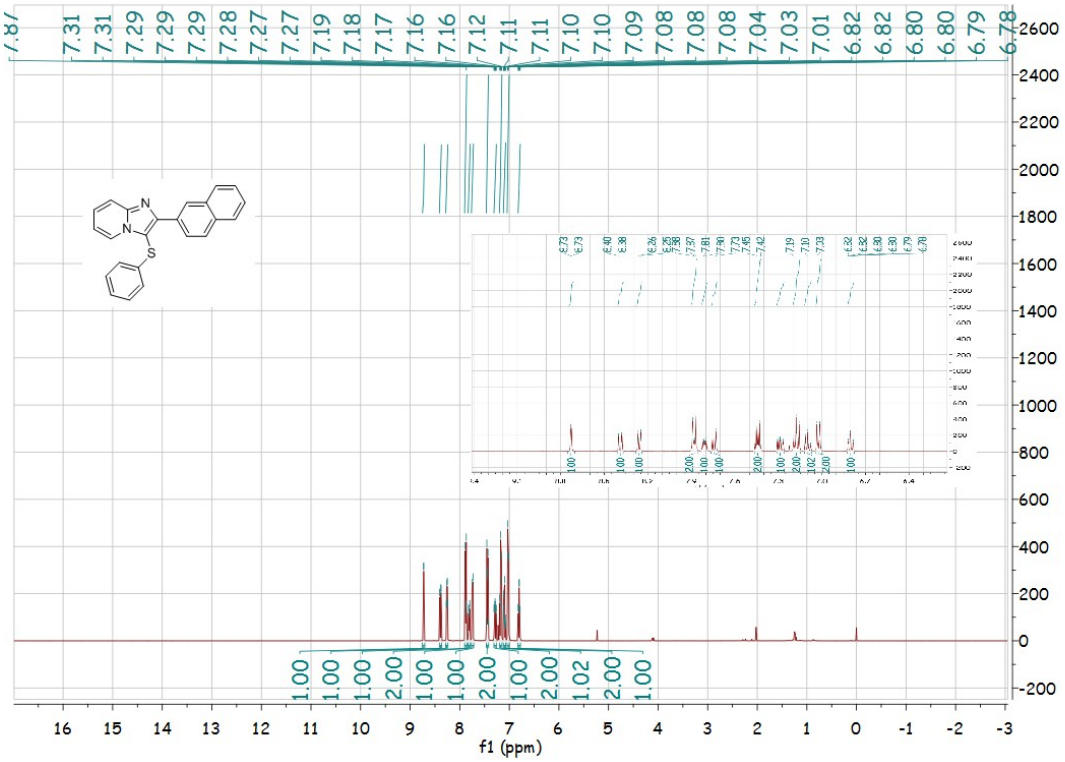
^{1}H NMR of compound **5b**



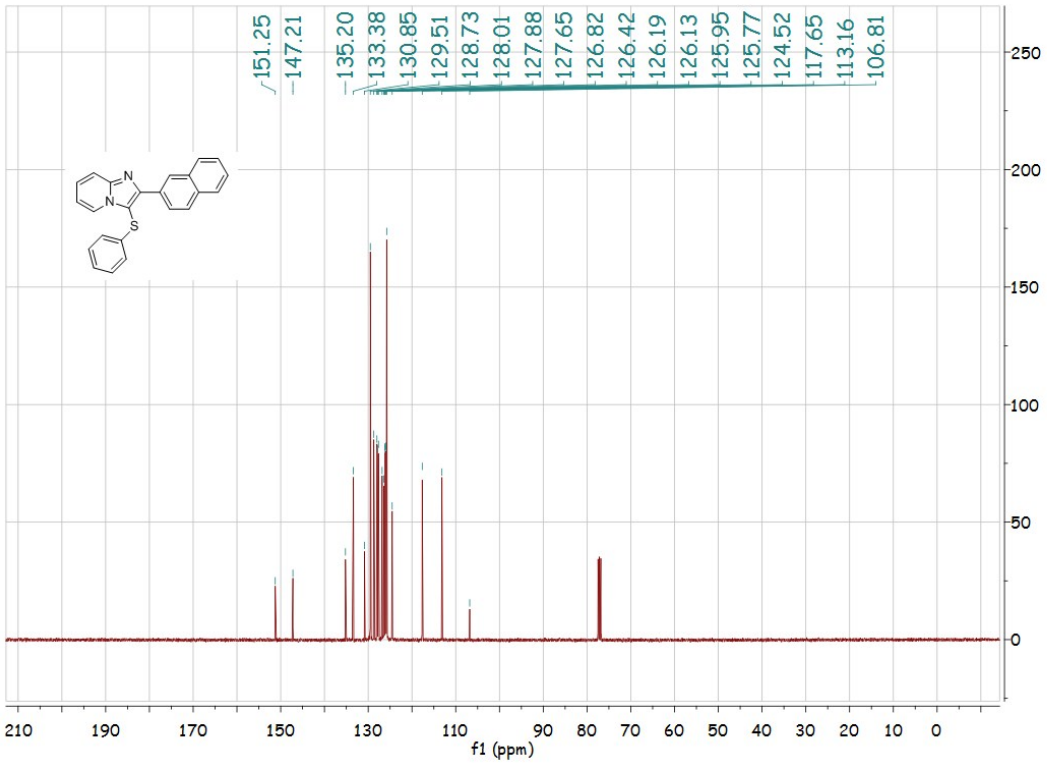
^{13}C NMR of compound **5b**



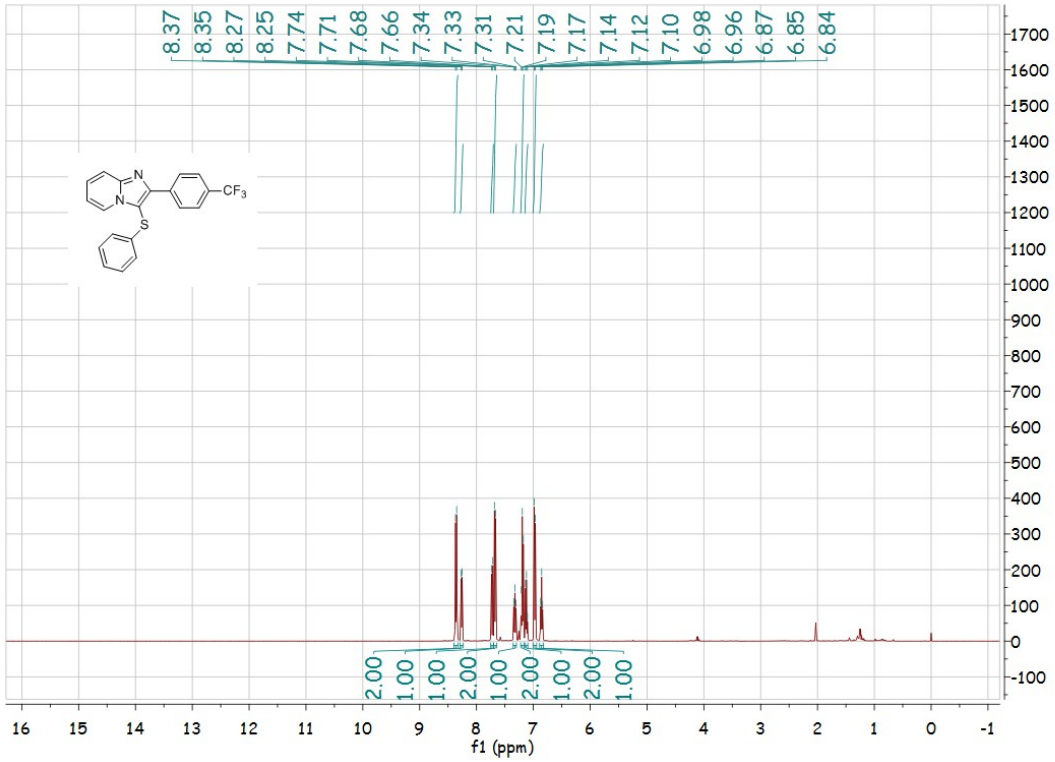
^{1}H NMR of compound **5c**



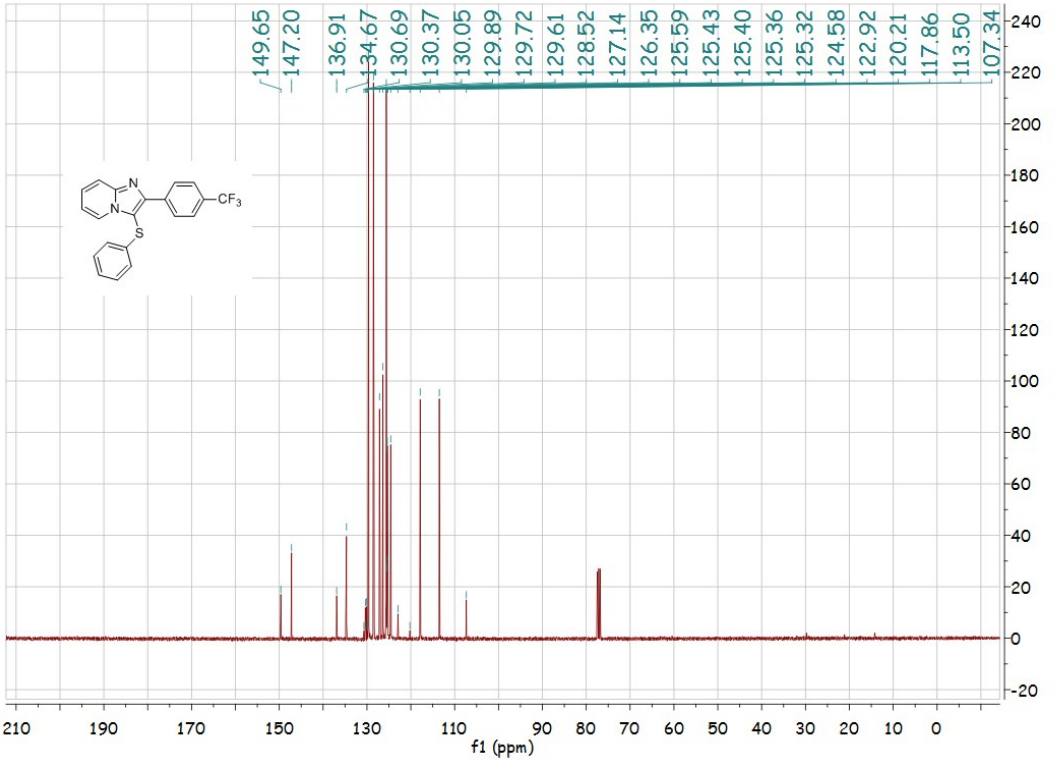
^{13}C NMR of compound **5c**



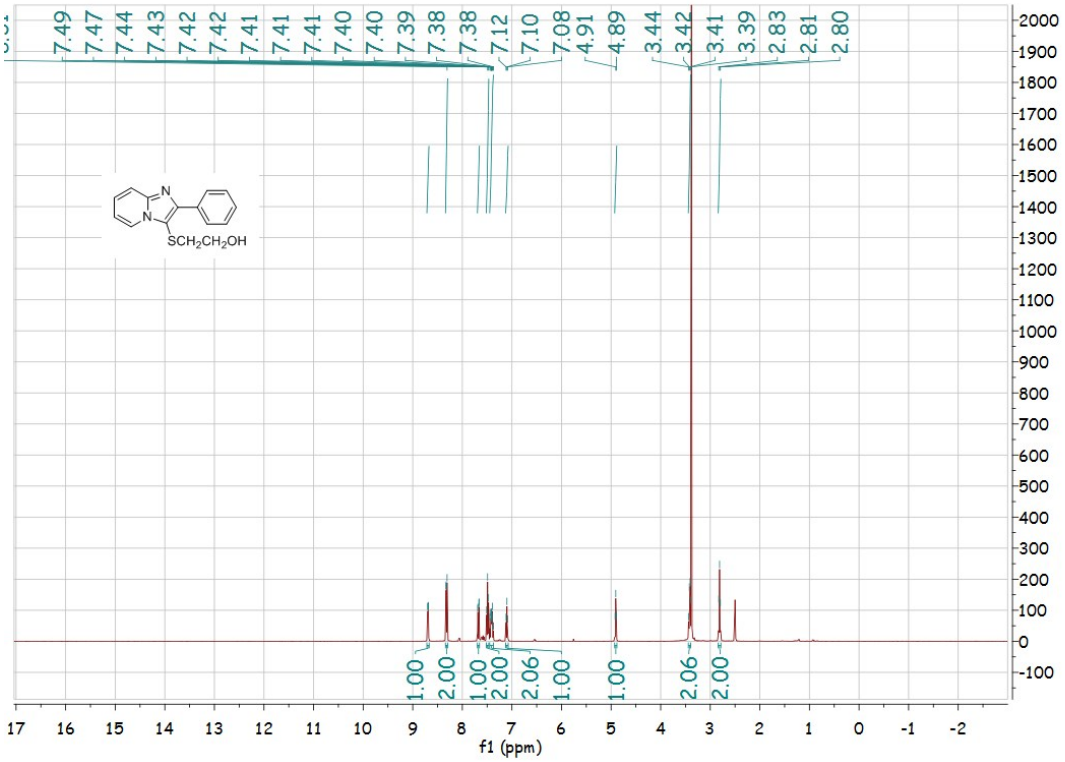
^{1}H NMR of compound **5d**



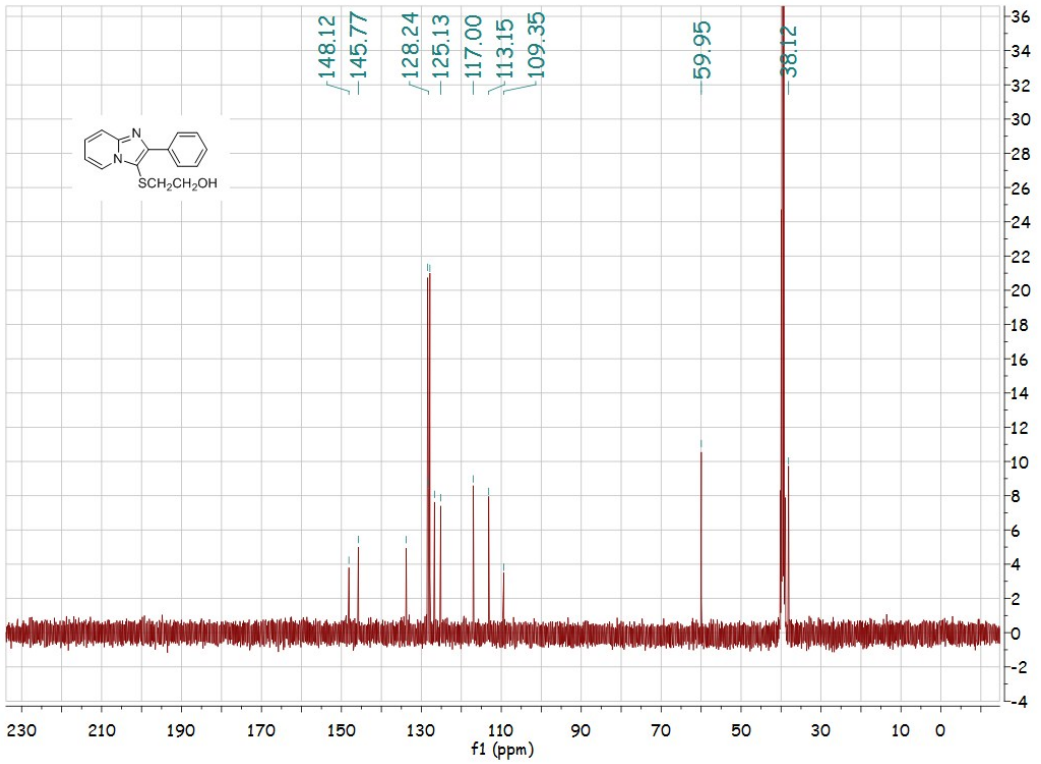
^{13}C NMR of compound **5d**



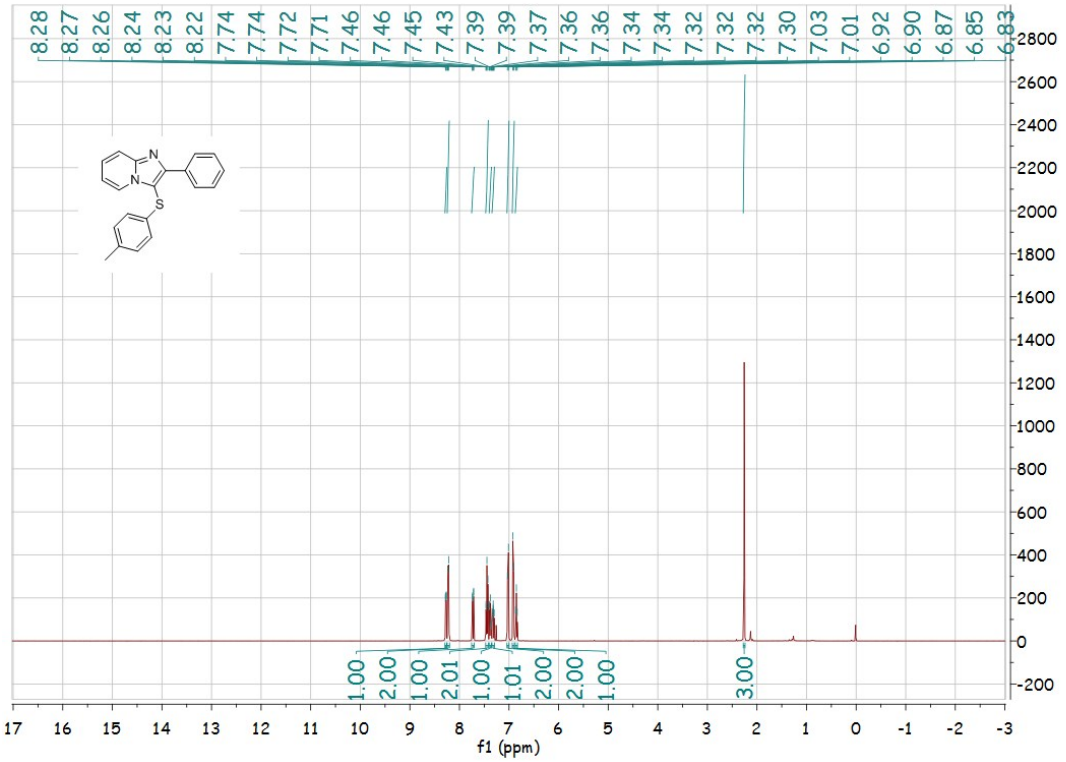
^{1}H NMR of compound **5e**



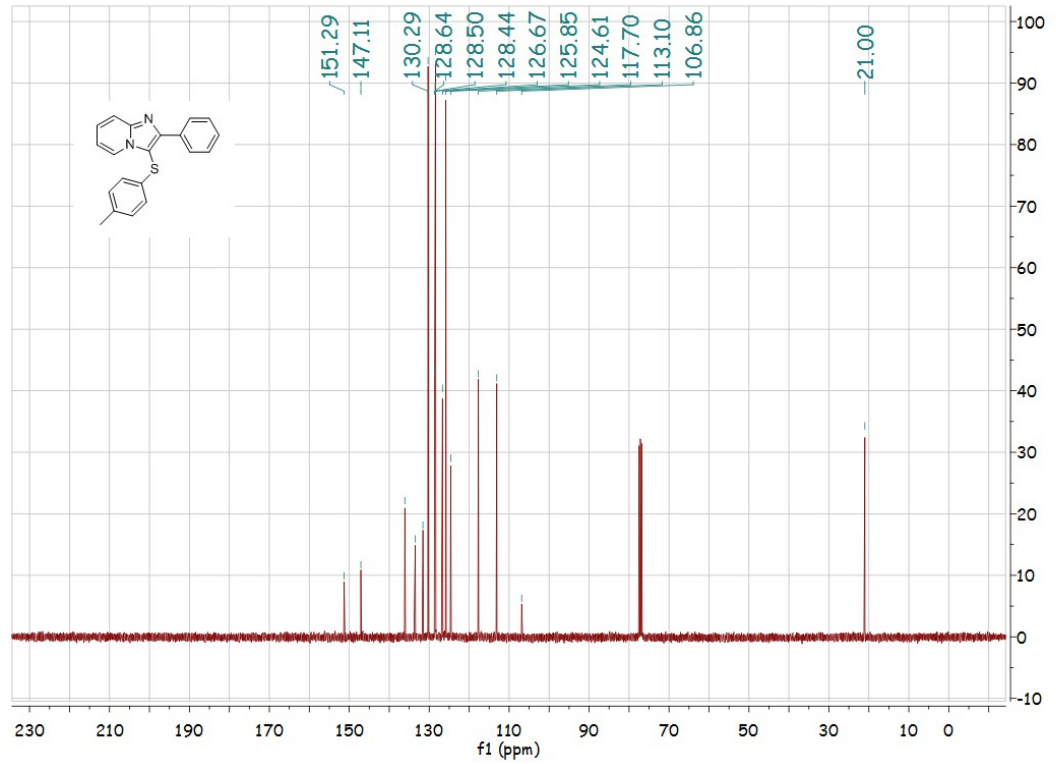
^{13}C NMR of compound **5e**



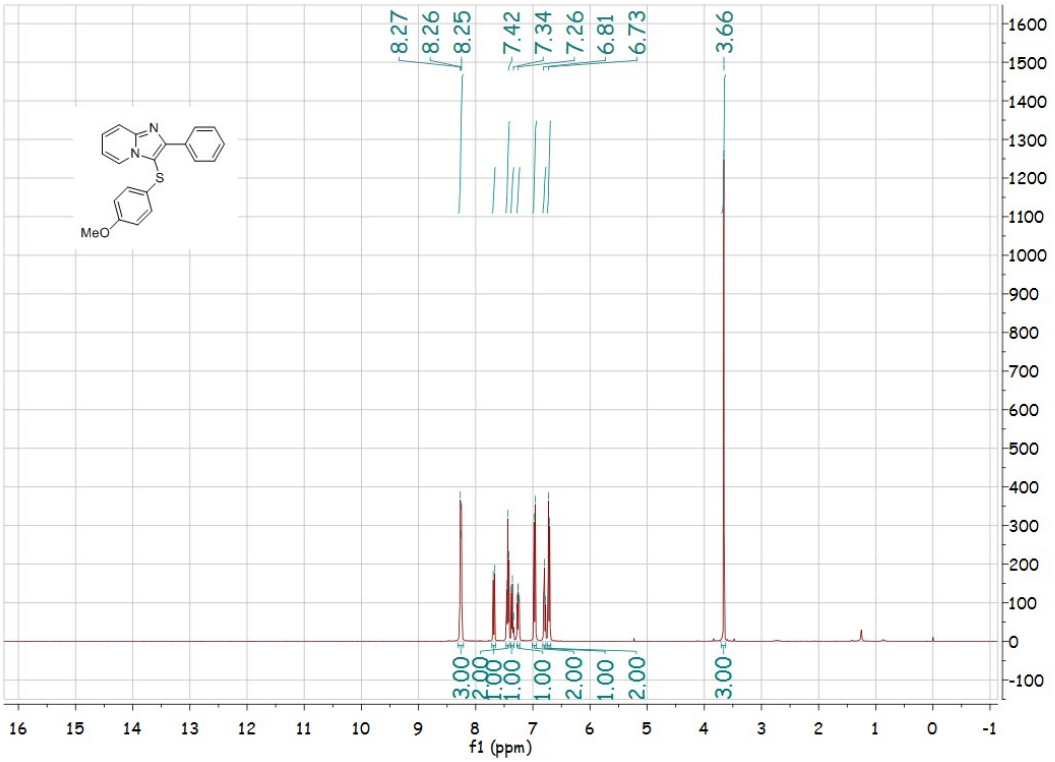
^{1}H NMR of compound **5f**



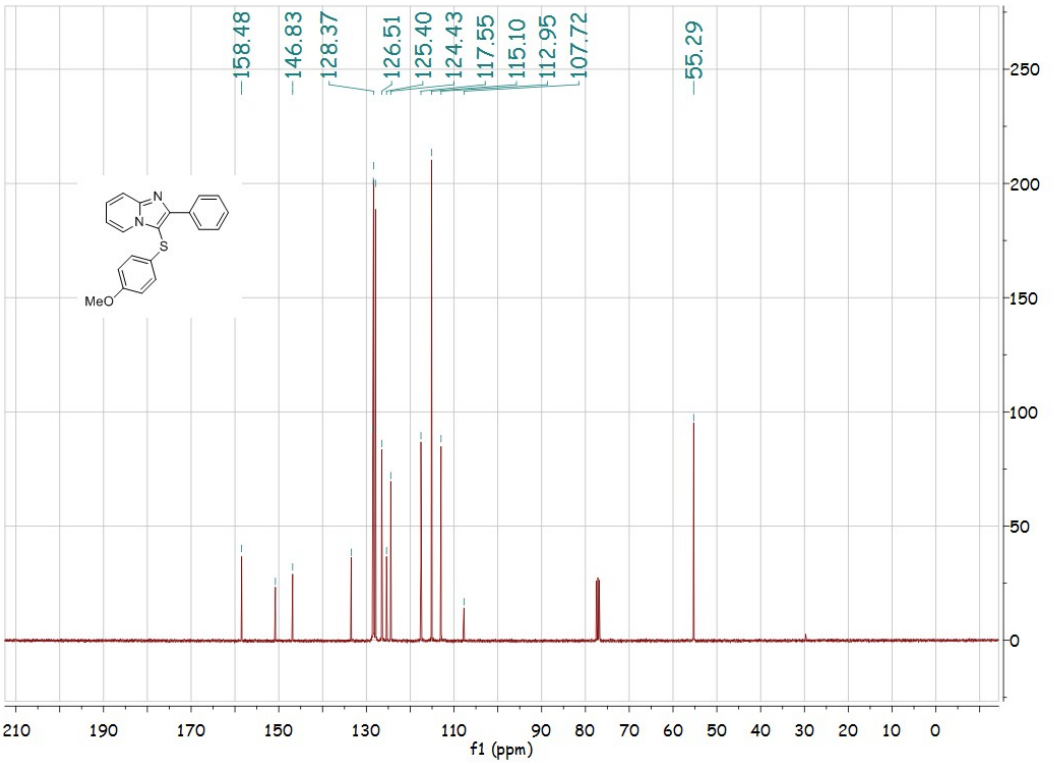
^{13}C NMR of compound **5f**



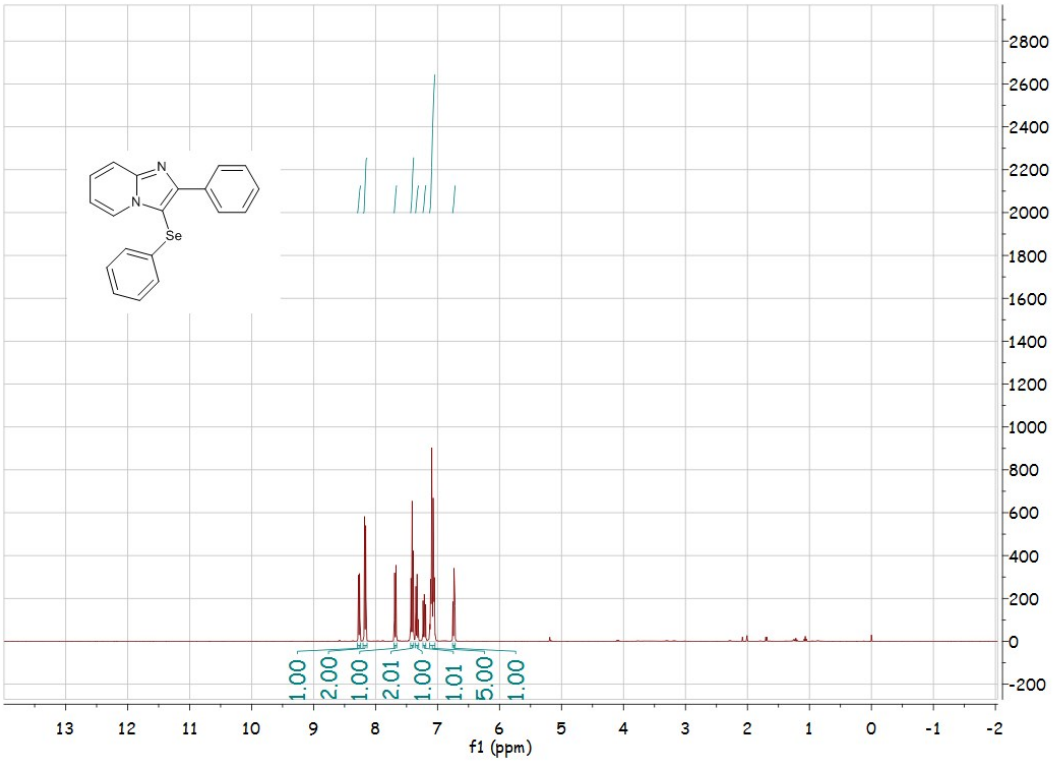
^{1}H NMR of compound **5g**



^{13}C NMR of compound **5g**



^{1}H NMR of compound **5h**



^{13}C NMR of compound **5h**