# A rhodium(I)-catalysed formal intramolecular C–C/C–H bond metathesis

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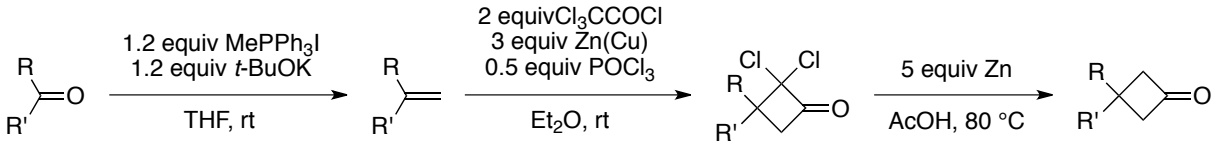
## Electronic Supplementary Information

**General.** All reactions were carried out with standard Schlenk techniques under an argon or nitrogen atmosphere. Column chromatography was carried out on Wakogel® C-200 (75–150 $\mu$m). Preparative thin-layer chromatography (TLC) was performed on Wakogel® B-5F. Proton chemical shifts were referenced to residual $CHCl_3$ signal at 7.26 ppm. Carbon chemical shifts were referenced to $CDCl_3$ at 77.0 ppm.

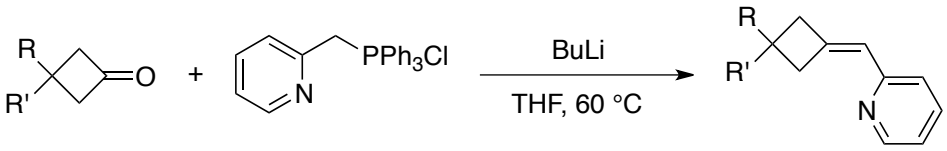
**Materials.** Cyclobutanones and (pyridylmethylene)cyclobutanes were prepared according to the literature methods. All other commercially available chemical resources were used as received without further purification.

**Preparation of Substrates.**

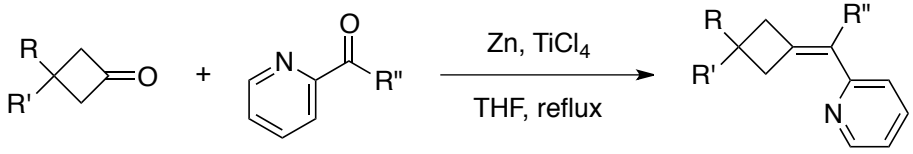
**(1) preparation of cyclobutanones**

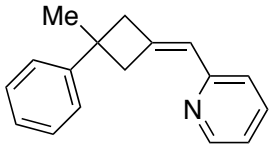



**(2) trisubstituted alkenes**

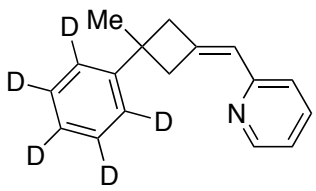



**(3) tetrasubstituted alkenes**

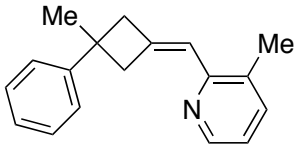


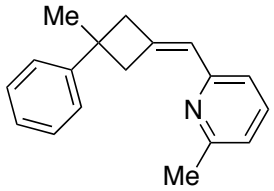
**2-Methyl-2-phenyl-4-(2-pyridylmethylene)cyclobutane (1a):** White solid, mp 42–43 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.52 (s, 3H), 2.93–2.99 (m, 1H), 3.18–3.24 (m, 1H), 3.28–3.34 (m, 1H), 3.46–3.52 (m, 1H), 6.42 (quint, $J$ = 2.2 Hz, 1H), 7.02–7.07 (m, 1H), 7.15–7.23 (m, 2H), 7.28–7.37 (m, 4H), 7.60 (dt, $J$ = 2.0, 7.5 Hz, 1H), 8.54–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 30.8, 40.3, 45.5, 46.0, 120.5, 121.7, 123.4, 125.1, 125.5, 128.2, 135.9, 144.6, 149.3, 150.3, 156.7; HRMS (ESI) calcd for $C_{17}H_{17}NNa$ $[M + Na]^+$ 258.1253, found 258.1253.



**2-Methyl-2-($^2H_5$)phenyl-4-(2-pyridylmethylene)cyclobutane (1a-$d_5$):** White solid, mp 43–45 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.53 (s, 3H), 2.93–2.99 (m, 1H), 3.18–3.24 (m, 1H), 3.28–3.34 (m, 1H), 3.46–3.52 (m, 1H), 6.42 (quint, $J$ = 2.2 Hz, 1H), 7.02–7.07 (m, 1H), 7.17 (d, $J$ = 8.0 Hz, 1H), 7.60 (dt, $J$ = 1.7, 7.6 Hz, 1H), 8.54–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.7, 40.2, 45.6, 46.1, 120.4, 121.7, 123.4, 124.7 (t, $J_{C–D}$ = 24.0 Hz), 125.0 (t, $J_{C–D}$ = 25.2 Hz), 127.7 (t, $J_{C–D}$ = 24.0 Hz), 135.8, 144.6, 149.3, 150.1, 156.7; HRMS (ESI) calcd for $C_{17}H_{13}D_5N$ $[M + H]^+$ 241.1748, found 241.1745.

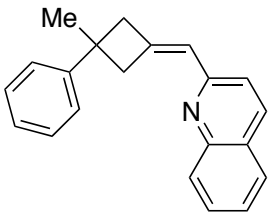



**2-Methyl-4-[(3-methyl-2-pyridyl)methylene]-2-phenylcyclobutane (1b):** Pale yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.52 (s, 3H), 2.32 (s, 3H), 2.92–2.98 (m, 1H), 3.26–3.34 (m, 2H), 3.52–3.58 (m, 1H), 6.47 (quint, $J$ = 2.2 Hz, 1H), 6.96 (dd, $J$ = 7.8, 4.8 Hz, 1H), 7.16–7.21 (m, 1H), 7.28–7.35 (m, 4H), 7.37–7.40 (m, 1H), 8.39–8.42 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 19.0, 30.8, 40.3, 45.6, 46.6, 118.6, 120.4, 125.2, 125.3, 128.1, 129.9, 137.3, 146.1, 146.6, 150.6, 155.2; HRMS (MALDI) calcd for $C_{18}H_{19}NNa$ $[M + Na]^+$ 272.1410, found 272.1397.

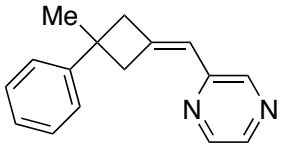



**2-Methyl-4-[(6-methyl-2-pyridyl)methylene]-2-phenylcyclobutane (1c):** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.52 (s, 3H), 2.53 (s, 3H), 2.91–2.97 (m, 1H), 3.14–3.20 (m, 1H), 3.27–3.33 (m, 1H), 3.43–3.50 (m, 1H), 6.41 (quint, $J$ = 2.2 Hz, 1H), 6.92 (d, $J$ = 7.5 Hz,

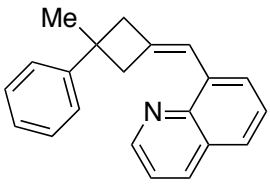
1H), 6.99 (d, $J$ = 8.0 Hz, 1H), 7.17–7.22 (m, 1H), 7.27–7.31 (m, 2H), 7.31–7.37 (m, 2H), 7.50 (t, $J$ = 7.5 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 24.3, 30.4, 40.0, 45.2, 45.8, 118.3, 119.7, 123.5, 124.8, 125.2, 127.9, 135.8, 143.7, 150.0, 155.7, 157.4; HRMS (ESI) calcd for $C_{18}H_{19}NNa$ $[M + Na]^+$ 272.1410, found 272.1414.



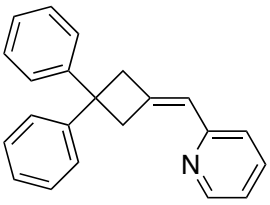

**2-Methyl-2-phenyl-4-(2-quinolylmethylene)cyclobutane (1d):** Pale yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.55 (s, 3H), 2.98–3.05 (m, 1H), 3.29–3.41 (m, 2H), 3.58–3.64 (m, 1H), 6.63 (quint, $J$ = 2.3 Hz, 1H), 7.19–7.24 (m, 1H), 7.28–7.38 (m, 5H), 7.44–7.48 (m, 1H), 7.65–7.69 (m, 1H), 7.74–7.77 (m, 1H), 8.02 (d, $J$ = 8.0 Hz, 1H), 8.07 (d, $J$ = 8.5 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.7, 40.3, 45.8, 46.2, 120.2, 124.2, 125.1, 125.59, 125.64, 126.3, 127.3, 128.2, 129.0, 129.3, 135.7, 146.8, 148.1, 150.1, 156.8; HRMS (ESI) calcd for $C_{21}H_{20}N$ $[M + H]^+$ 286.1590, found 286.1587.



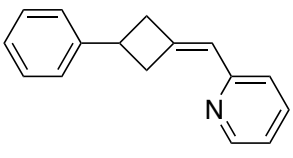

**2-Methyl-2-phenyl-4-(2-pyrazylmethylene)cyclobutane:** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.53 (s, 3H), 2.96–3.02 (m, 1H), 3.24–3.30 (m, 1H), 3.31–3.37 (m, 1H), 3.50–3.56 (m, 1H), 6.40 (quint, $J$ = 2.4 Hz, 1H), 7.19–7.23 (m, 1H), 7.27–7.31 (m, 2H), 7.32–7.38 (m, 2H), 8.29 (d, $J$ = 2.0 Hz, 1H), 8.45 (d, $J$ = 1.0 Hz, 1H), 8.48–8.50 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.7, 40.2, 45.5, 46.3, 119.6, 124.9, 125.5, 128.1, 140.7, 143.3, 143.8, 148.5, 149.7, 152.3; HRMS (ESI) calcd for $C_{16}H_{17}N_2$ $[M + H]^+$ 237.1386, found 237.1386.



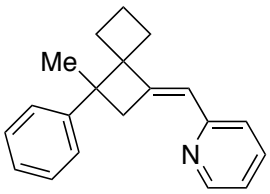

**2-Methyl-2-phenyl-4-(8-quinolylmethylene)cyclobutane:** Yellow oil; $^{1}$H NMR ($CDCl_3$, 500 MHz) δ 1.54 (s, 3H), 3.05–3.16 (m, 2H), 3.38–3.44 (m, 1H), 3.50–3.56 (m, 1H), 7.17–7.23 (m, 1H), 7.28–7.37 (m, 4H), 7.41 (dd, *J* = 8.5, 4.5 Hz, 1H), 7.52 (t, *J* = 7.7 Hz, 1H), 7.59 (quint, *J* = 2.5 Hz, 1H), 7.65–7.69 (m, 2H), 8.14 (dd, *J* = 8.3, 2.3 Hz, 1H), 8.96 (dd, *J* = 4.0, 2.0 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.7, 40.6, 45.7, 45.8, 118.2, 120.9, 125.2, 125.5, 125.8, 126.2, 126.5, 128.2, 128.4, 135.8, 136.2, 140.4, 145.5, 149.1, 150.6; HRMS (ESI) calcd for $C_{21}H_{20}N$ $[M + H]^+$ 286.1590, found 286.1592.



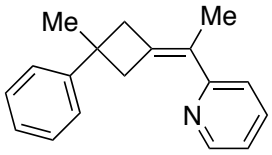

**2,2-Diphenyl-4-(2-pyridylmethylene)cyclobutane (1e):** White solid, mp 91–92 °C; $^{1}$H NMR ($CDCl_3$, 500 MHz) δ 3.68 (s, 2H), 3.92 (s, 2H), 6.39 (quint, *J* = 2.5 Hz, 1H), 7.01–7.06 (m, 1H), 7.14–7.20 (m, 3H), 7.26–7.33 (m, 8H), 7.59 (dt, *J* = 2.2, 7.6 Hz, 1H), 8.54–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 46.8, 47.6, 48.2, 120.5, 121.9, 122.5, 125.8, 126.4, 128.2, 135.9, 143.9, 148.8, 149.3, 156.5; HRMS (ESI) calcd for $C_{22}H_{19}NNa$ $[M + Na]^+$ 320.1410, found 320.1415.



**2-Phenyl-4-(2-pyridylmethylene)cyclobutane (1f):** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 3.05–3.12 (m, 1H), 3.23–3.38 (m, 2H), 3.55–3.63 (m, 1H), 3.67–3.76 (m, 1H), 6.38 (quint, *J* = 2.2 Hz, 1H), 7.02–7.07 (m, 1H), 7.17 (d, *J* = 8.0 Hz, 1H), 7.19–7.24 (m, 1H), 7.30–7.36 (m, 4H), 7.60 (dt, *J* = 2.0, 7.7 Hz, 1H), 8.54–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 36.3, 40.5, 40.8, 120.5, 121.7, 122.2, 126.1, 126.4, 128.4, 135.9, 145.3, 146.0, 149.3, 156.7; HRMS (ESI) calcd for $C_{16}H_{16}N$ $[M + H]^+$ 222.1277, found 222.1278.

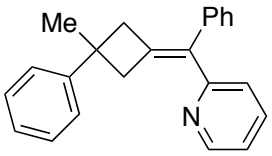



**(*E*)-1-Methyl-1-phenyl-3-(2-pyridylmethylene)spiro[3.3]heptane (1g):** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.41 (s, 3H), 1.70–1.78 (m, 1H), 1.80–2.01 (m, 3H), 2.18–2.26 (m, 1H), 2.28–2.37 (m, 1H), 2.95 (dd, *J* = 15.8, 1.8 Hz, 1H), 3.54 (dd, *J* = 15.8, 2.2 Hz, 1H), 6.75 (s, 1H), 7.06–7.12 (m, 1H), 7.20–7.25 (m, 1H), 7.26–7.33 (m, 3H), 7.34–7.39 (m, 2H), 7.62–7.68 (m, 1H), 8.56–8.60 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 15.4, 25.0, 28.4, 31.5, 41.6, 45.3, 58.3, 119.1, 120.5, 121.8, 125.7, 126.2, 128.1, 135.9, 146.0, 149.3, 156.3, 156.9; HRMS (MALDI) calcd for $C_{20}H_{21}NNa$ $[M + Na]^+$ 298.1566, found 298.1554.

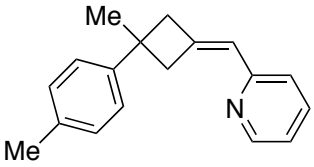



**2-Methyl-2-phenyl-4-[1-(2-pyridyl)ethylidene]cyclobutane (1h):** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.49 (s, 3H), 2.03–2.06 (m, 3H), 2.93–2.99 (m, 1H), 3.03–3.09 (m, 1H), 3.22–3.28 (m, 1H), 3.39–3.46 (m, 1H), 7.05–7.09 (m, 1H), 7.16–7.21 (m, 1H), 7.23–7.27 (m, 1H), 7.28–7.36 (m, 4H), 7.58–7.63 (m, 1H), 8.58–8.61 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6

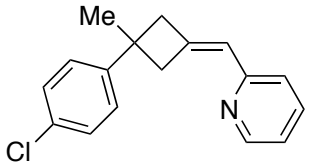
MHz) δ 15.4, 31.1, 39.0, 44.4, 46.2, 120.5, 121.4, 125.2, 125.5, 128.1, 128.2, 135.5, 137.5, 148.8, 150.6, 158.7; HRMS (ESI) calcd for $C_{18}H_{19}NNa$ $[M + Na]^+$ 272.1410, found 272.1407.



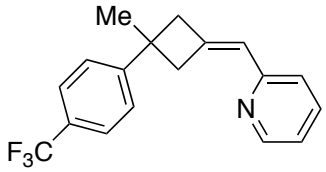

**2-Methyl-2-phenyl-4-[phenyl(2-pyridyl)methylene]cyclobutane (1i):** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.54 (s, 3H), 2.91–2.97 (m, 1H), 3.21–3.31 (m, 2H), 3.46–3.51 (m, 1H), 7.06–7.12 (m, 2H), 7.16–7.20 (m, 1H), 7.22–7.38 (m, 9H), 7.58 (dt, $J$ = 1.7, 7.8 Hz, 1H), 8.60–8.64 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 31.1, 39.4, 45.2, 46.0, 120.8, 123.1, 125.1, 125.4, 126.6, 128.1, 128.2, 129.1, 134.3, 135.7, 139.3, 140.6, 149.1, 150.4, 158.9; HRMS (ESI) calcd for $C_{23}H_{22}N$ $[M + H]^+$ 312.1747, found 312.1746.



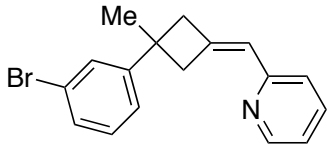

**2-Methyl-2-(4-methylphenyl)-4-(2-pyridylmethylene)cyclobutane (1j):** Pale yellow solid, mp 33–37 °C; $^1$H NMR ($CDCl_3$, 301 MHz) δ 1.51 (s, 3H), 2.34 (s, 3H), 2.89–2.98 (m, 1H), 3.13–3.23 (m, 1H), 3.24–3.33 (m, 1H), 3.42–3.51 (m, 1H), 6.42 (quint, $J$ = 2.0 Hz, 1H), 7.02–7.08 (m, 1H), 7.12–7.22 (m, 5H), 7.61 (dt, $J$ = 1.6, 7.7 Hz, 1H), 8.53–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 20.9, 30.7, 40.0, 45.6, 46.1, 120.4, 121.6, 123.4, 125.0, 128.9, 135.0, 135.8, 144.8, 147.3, 149.3, 156.7; HRMS (ESI) calcd for $C_{18}H_{19}NNa$ $[M + Na]^+$ 272.1410, found 272.1408.



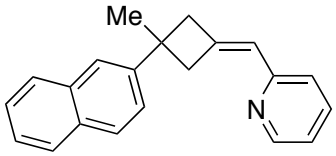

**2-(4-Chlorophenyl)-2-methyl-4-(2-pyridylmethylene)cyclobutane (1k):** White solid, mp 59–62 °C; $^{1}$H NMR ($CDCl_3$, 500 MHz) δ 1.50 (s, 3H), 2.92–2.98 (m, 1H), 3.18–3.29 (m, 2H), 3.42–3.48 (m, 1H), 6.41 (quint, $J$ = 2.3 Hz, 1H), 7.03–7.07 (m, 1H), 7.15 (d, $J$ = 8.0 Hz, 1H), 7.20–7.24 (m, 2H), 7.28–7.32 (m, 2H), 7.60 (dt, $J$ = 2.0, 7.5 Hz, 1H), 8.54–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 30.6, 40.0, 45.5, 46.1, 120.5, 121.8, 123.5, 126.6, 128.3, 131.2, 135.9, 143.9, 148.8, 149.3, 156.6; HRMS (ESI) calcd for $C_{17}H_{17}ClN$ $[M + H]^+$ 270.1044, found 270.1046.



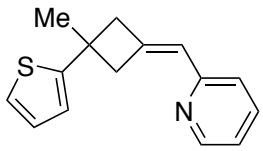

**2-Methyl-4-(2-pyridylmethylene)-2-[4-(trifluoromethyl)phenyl]cyclobutane (1l):** White solid, mp 48–50 °C; $^{1}$H NMR ($CDCl_3$, 500 MHz) δ 1.53 (s, 3H), 2.96–3.03 (m, 1H), 3.24–3.33 (m, 2H), 3.47–3.53 (m, 1H), 6.42 (quint, $J$ = 2.0 Hz, 1H), 7.04–7.08 (m, 1H), 7.15 (d, $J$ = 8.0 Hz, 1H), 7.39 (d, $J$ = 8.0 Hz, 2H), 7.59 (d, $J$ = 8.0 Hz, 2H), 7.61 (dt, $J$ = 2.0, 7.6 Hz, 1H), 8.55–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.6, 40.5, 45.4, 46.0, 120.6, 121.8, 123.6, 124.3 (q, $^{1}J_{C–F}$ = 271.0 Hz), 125.2 (q, $^{3}J_{C–F}$ = 3.5 Hz), 125.6, 127.9 (q, $^{2}J_{C–F}$ = 32.4 Hz), 135.9, 143.5, 149.4, 154.3, 156.5; HRMS (ESI) calcd for $C_{18}H_{17}F_3N$ $[M + H]^+$ 304.1308, found 304.1303.



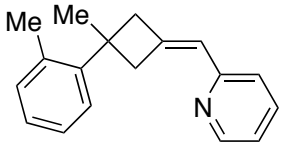

**2-(3-Bromophenyl)-2-methyl-4-(2-pyridylmethylene)cyclobutane (1m):** Yellow oil; $^{1}$H NMR ($CDCl_3$, 301 MHz) δ 1.51 (s, 3H), 2.90–3.00 (m, 1H), 3.17–3.33 (m, 2H), 3.42–3.52 (m, 1H), 6.41 (quint, *J* = 2.2 Hz, 1H), 7.02–7.09 (m, 1H), 7.15 (d, *J* = 7.8 Hz, 1H), 7.19–7.24 (m, 2H), 7.24–7.37 (m, 1H), 7.40–7.44 (m, 1H), 7.61 (dt, *J* = 1.7, 7.7 Hz, 1H), 8.53–8.58 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.6, 40.2, 45.3, 46.0, 120.5, 121.7, 122.3, 123.5, 123.8, 128.4, 128.6, 129.8, 135.8, 143.6, 149.2, 152.6, 156.4; HRMS (ESI) calcd for $C_{17}H_{17}BrN$ $[M + H]^+$ 314.0539, found 314.0534.



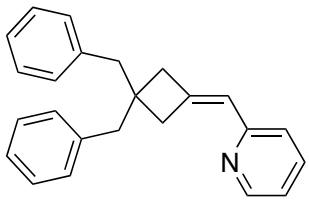

**2-Methyl-2-(2-naphthyl)-4-(2-pyridylmethylene)cyclobutane (1n):** White solid, mp 102–104 °C; $^{1}$H NMR ($CDCl_3$, 500 MHz) δ 1.61 (s, 3H), 3.01–3.07 (m, 1H), 3.27–3.33 (m, 1H), 3.39–3.45 (m, 1H), 3.58–3.64 (m, 1H), 6.45 (quint, *J* = 2.0 Hz, 1H), 7.04–7.08 (m, 1H), 7.19 (d, *J* = 7.5 Hz, 1H), 7.41–7.50 (m, 3H), 7.62 (dt, *J* = 1.5, 7.7 Hz, 1H), 7.68–7.71 (m, 1H), 7.80–7.85 (m, 3H), 8.55–8.59 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 30.6, 40.5, 45.5, 46.0, 120.5, 121.7, 122.9, 123.5, 124.2, 125.2, 126.0, 127.5, 127.6, 128.0, 131.6, 133.2, 135.8, 144.5, 147.5, 149.3, 156.6; HRMS (ESI) calcd for $C_{21}H_{20}N$ $[M + H]^+$ 286.1590, found 286.1590.



**2-Methyl-4-(2-pyridylmethylene)-2-(2-thienyl)cyclobutane (1o):** Yellow oil; $^1$H NMR ($CDCl_3$, 301 MHz) δ 1.66 (s, 3H), 2.95–3.05 (m, 1H), 3.20–3.35 (m, 2H), 3.46–3.56 (m, 1H), 6.42 (quint, *J* = 2.3 Hz, 1H), 6.89–6.97 (m, 2H), 7.02–7.08 (m, 1H), 7.11–7.18 (m, 2H), 7.60 (dt, *J* = 2.0, 7.7 Hz, 1H), 8.52–8.57 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 29.4, 37.9, 48.1, 48.7, 120.6, 121.8, 122.0, 123.0, 123.9, 126.7, 136.0, 143.7, 149.4, 154.9, 156.6; HRMS (ESI) calcd for $C_{15}H_{15}NNaS$ $[M + Na]^+$ 264.0817, found 264.0818.

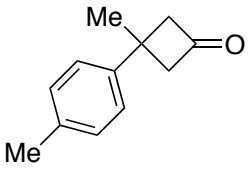



**2-Methyl-2-(2-methylphenyl)-4-(2-pyridylmethylene)cyclobutane (1p):** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.49 (s, 3H), 2.34 (s, 3H), 2.93–2.99 (m, 1H), 3.21–3.27 (m, 1H), 3.34–3.40 (m, 1H), 3.50–3.56 (m, 1H), 6.41–6.44 (m, 1H), 7.03–7.08 (m, 1H), 7.10–7.21 (m, 5H), 7.61 (dt, *J* = 1.7, 7.6 Hz, 1H), 8.55–8.58 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 19.9, 28.1, 41.2, 45.9, 46.1, 120.3, 121.6, 122.7, 125.7, 125.8, 125.9, 131.1, 134.7, 135.7, 145.2, 147.3, 149.2, 156.6; HRMS (ESI) calcd for $C_{18}H_{20}N$ $[M + H]^+$ 250.1590, found 250.1594.

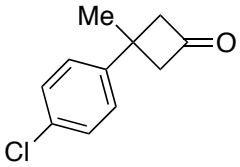



**2,2-Dibenzyl-4-(2-pyridylmethylene)cyclobutane (1q):** Yellow oil; $^1$H NMR ($CDCl_3$, 301 MHz) δ 2.78–2.82 (m, 2H), 2.86 (s, 4H), 2.99–3.03 (m, 2H), 6.29 (quint, *J* = 2.1 Hz, 1H), 7.00–7.06 (m, 1H), 7.09 (d, *J* = 8.1 Hz, 1H), 7.13–7.33 (m, 10H), 7.57 (dt, *J* = 2.0, 7.7 Hz, 1H), 8.50–8.55 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 40.1, 41.0, 41.7, 43.4, 119.9, 121.1,

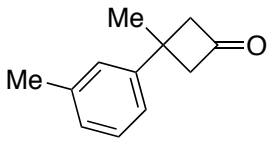
122.7, 125.6, 127.6, 129.7, 135.3, 138.4, 144.7, 148.8, 156.2; HRMS (ESI) calcd for $C_{24}H_{24}N$ $[M + H]^+$ 326.1903, found 326.1905.




**3-Methyl-3-(4-methylphenyl)cyclobutanone (6b):** Colourless oil; $^1$H NMR ($CDCl_3$, 301 MHz) δ 1.59 (s, 3H), 2.35 (s, 3H), 3.04–3.15 (m, 2H), 3.39–3.50 (m, 2H), 7.15–7.24 (m, 4H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 20.7, 30.8, 33.3, 59.0, 125.3, 129.0, 135.5, 145.1, 206.3; HRMS (ESI) calcd for $C_{12}H_{14}NaO$ $[M + Na]^+$ 197.0937, found 197.0933; IR ($\nu$/cm$^{-1}$): 1782, 1381, 818.

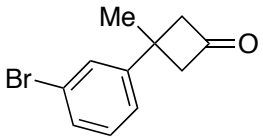



**3-(4-Chlorophenyl)-3-methylcyclobutanone (6c):** Colourless oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.60 (s, 3H), 3.08–3.15 (m, 2H), 3.39–3.47 (m, 2H), 7.22–7.27 (m, 2H), 7.32–7.36 (m, 2H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.7, 33.4, 59.0, 127.0, 128.4, 131.8, 146.6, 205.5; HRMS (ESI) calcd for $C_{11}H_{11}ClNaO$ $[M + Na]^+$ 217.0391, found 217.0396; IR ($\nu$/cm$^{-1}$): 1790, 1496, 1095, 825.

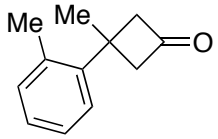



**3-Methyl-3-(3-methylphenyl)cyclobutanone (6d):** Colourless oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.60 (s, 3H), 2.38 (s, 3H), 3.06–3.13 (m, 2H), 3.43–3.50 (m, 2H), 7.05–7.13 (m,

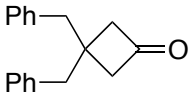
3H), 7.24–7.29 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 21.4, 31.0, 33.7, 59.1, 122.6, 126.3, 126.9, 128.4, 138.1, 148.2, 206.7; HRMS (ESI) calcd for $C_{12}H_{14}NaO$ $[M + Na]^+$ 197.0937, found 197.0935; IR ($\nu$/cm$^{-1}$): 2962, 1782, 787.




**3-(3-Bromophenyl)-3-methylcyclobutanone (6e):** White solid, mp 64–68 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.60 (s, 3H), 3.08–3.15 (m, 2H), 3.41–3.48 (m, 2H), 7.22–7.25 (m, 2H), 7.37–7.41 (m, 1H), 7.44–7.46 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.7, 33.8, 59.0, 122.5, 124.3, 128.9, 129.3, 130.1, 150.5, 205.4; HRMS (ESI) calcd for $C_{11}H_{11}BrNaO$ $[M + Na]^+$ 260.9885, found 260.9888. IR ($\nu$/cm$^{-1}$): 1774, 1072, 787, 694.



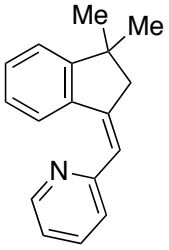

**3-Methyl-3-(2-methylphenyl)cyclobutanone (6f):** Pale yellow oil; $^1$H NMR ($CDCl_3$, 301 MHz) δ 1.55 (s, 3H), 2.34 (s, 3H), 3.05–3.15 (m, 2H), 3.45–3.56 (m, 2H), 7.15–7.22 (m, 4H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 20.1, 28.3, 34.3, 58.8, 125.9, 126.3, 126.5, 131.3, 134.8, 145.2, 206.3; HRMS (ESI) calcd for $C_{12}H_{14}NaO$ $[M + Na]^+$ 197.0937, found 197.0941; IR ($\nu$/cm$^{-1}$): 1790, 1381, 764.




**3,3-Dibenzylcyclobutanone (6h):** Yellow solid, mp 69–74 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 2.90 (s, 4H), 2.95 (s, 4H), 7.15–7.19 (m, 4H), 7.24–7.29 (m, 2H), 7.30–7.35 (m, 4H);

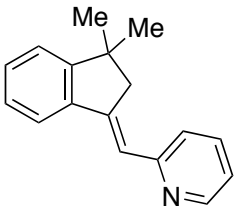
$^{13}$C NMR ($CDCl_3$, 126 MHz) δ 33.6, 44.0, 54.3, 126.6, 128.3, 130.1, 137.8, 206.3; HRMS (ESI) calcd for $C_{18}H_{18}NaO$ $[M + Na]^+$ 273.1250, found 273.1254; IR ($\nu$/cm$^{-1}$): 1774, 1119, 756, 702.

**General Procedure for the Rhodium(I)-Catalysed Skeletal Reorganisation of (Pyridylmethylene)cyclobutanes 1.** A Schlenk tube was charged with (pyridylmethylene)cyclobutane **1** (0.100 mmol), $RhCl(PPh_3)_3$ (4.0 $\mu$mol, 4 mol%). The tube was evacuated and backfilled with nitrogen. *p*-Xylene (1.0 mL) was added via syringe through the septum. After heating at 150 °C for the indicated time, the reaction mixture was cooled to room temperature and was filtered through a plug of Florisil® washing with hexane–AcOEt (1:1), and the filtrate was concentrated under reduced pressure. The residue was purified by preparative TLC on silica gel to afford the following compounds.

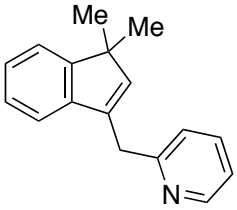



**(*Z*)-1,1-Dimethyl-3-(2-pyridylmethylene)indane (2a).** The general procedure was followed using **1a** (23.5 mg, 0.100 mmol), $RhCl(PPh_3)_3$ (3.6 mg, 3.9 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.0 mL) for 30 min. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1, twice) yielded **2a** (20.9 mg, 0.089 mmol, 89%) as a yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.34 (s, 6H), 2.81 (d, $J$ = 2.5 Hz, 2H), 6.61 (t, $J$ = 2.0 Hz, 1H), 7.03–7.08 (m, 1H), 7.12–7.16 (m, 1H), 7.22–7.25 (m, 2H), 7.42 (d, $J$ = 8.0 Hz, 1H), 7.63 (dt, $J$ = 2.0, 7.7 Hz, 1H), 7.80 (d, $J$ = 7.5 Hz, 1H), 8.64–8.67 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 29.3, 41.3, 52.0,

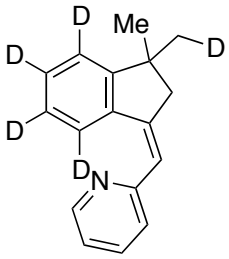
121.2, 121.6, 122.4, 123.8, 125.3, 125.9, 129.1, 136.0, 137.4, 144.8, 149.3, 156.8, 157.7; HRMS (ESI) calcd for $C_{17}H_{18}N$ $[M + H]^+$ 236.1434, found 236.1437.



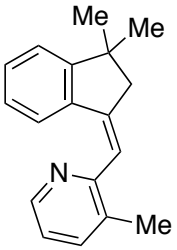

**(*E*)-1,1-Dimethyl-3-(2-pyridylmethylene)indane (3a):** Yellow oil; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.35 (s, 6H), 3.18 (d, *J* = 2.5 Hz, 2H), 7.04–7.10 (m, 2H), 7.24–7.33 (m, 3H), 7.38 (d, *J* = 8.0 Hz, 1H), 7.62–7.68 (m, 2H), 8.63–8.65 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.1, 42.8, 48.2, 118.8, 120.4, 120.6, 122.8, 123.5, 126.8, 129.1, 135.9, 140.4, 146.8, 149.3, 155.4, 157.2; HRMS (ESI) calcd for $C_{17}H_{18}N$ $[M + H]^+$ 236.1434, found 236.1435.




**1,1-Dimethyl-3-(2-pyridylmethyl)indene (4a):** Yellow oil; $^1$H NMR ($CDCl_3$, 301 MHz) δ 1.32 (s, 6H), 4.04 (d, *J* = 1.2, 2H), 6.08 (t, *J* = 1.2 Hz, 1H), 7.10–7.21 (m, 5H), 7.28–7.34 (m, 1H), 7.57 (dt, *J* = 1.9, 7.7 Hz, 1H), 8.55–8.90 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 24.6, 36.9, 48.3, 119.8, 121.0, 121.3, 122.9, 125.2, 126.3, 136.6, 137.5, 142.8, 143.9, 149.0, 154.1, 159.5; HRMS (ESI) calcd for $C_{17}H_{18}N$ $[M + H]^+$ 236.1434, found 236.1435.

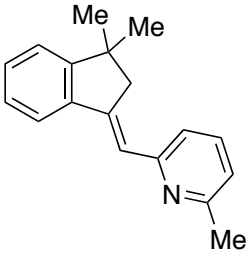



**(*Z*)-1-Methyl-1-($^2H_1$)methyl-3-(2-pyridylmethylene)-(4,5,6,7-$^2H_4$)indane (2a-$d_5$).** The general procedure was followed using **1a**-$d_5$ (33.0 mg, 0.137 mmol), $RhCl(PPh_3)_3$ (5.6 mg, 6.1 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.5 mL) for 1 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1) yielded **2a**-$d_5$ (25.2 mg, 0.105 mmol, 76%) as a yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.31–1.36 (m, 5H), 2.81 (d, $J$ = 1.5 Hz, 2H), 6.62 (t, $J$ = 2.0 Hz, 1H), 7.11–7.16 (m, 1H), 7.42 (d, $J$ = 8.0 Hz, 1H), 7.62 (dt, $J$ = 1.7, 7.6 Hz, 1H), 8.64–8.68 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 29.0 (t, $J_{C–D}$ = 19.2 Hz), 29.3, 41.2, 52.0, 121.2, 121.6, 122.0 (t, $J_{C–D}$ = 23.4 Hz), 123.8, 125.0 (t, $J_{C–D}$ = 24.6 Hz), 125.4 (t, $J_{C–D}$ = 24.6 Hz), 128.6 (t, $J_{C–D}$ = 24.0 Hz), 136.0, 137.3, 144.9, 149.3, 156.8, 157.6; HRMS (ESI) calcd for $C_{17}H_{13}D_5N$ $[M + H]^+$ 241.1748, found 241.1749.

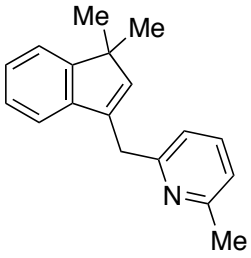



**(*Z*)-1,1-Dimethyl-3-[(3-methyl-2-pyridyl)methylene]indane (2b).** The general procedure was followed using **1b** (37.8 mg, 0.152 mmol), $RhCl(PPh_3)_3$ (5.6 mg, 6.1 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.5 mL) for 1 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1) yielded **2b** (27.8 mg, 0.111 mmol, 74%) as a pale yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.34 (s, 6H), 2.25 (s, 3H), 2.84 (d, $J$ = 2.0 Hz, 2H), 6.58–6.60 (m, 1H), 6.90–6.96 (m, 2H), 7.14 (dd, $J$ = 7.7, 4.7 Hz, 1H), 7.16–7.22 (m, 2H), 7.49–7.53 (m, 1H),

8.49–8.52 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 19.0, 29.4, 41.3, 50.7, 119.7, 121.9, 122.3, 124.4, 126.2, 128.8, 131.8, 137.6, 137.8, 144.2, 146.8, 156.1, 157.0; HRMS (MALDI) calcd for $C_{18}H_{19}NNa$ $[M + Na]^+$ 272.1410, found 272.1703.




**3c**

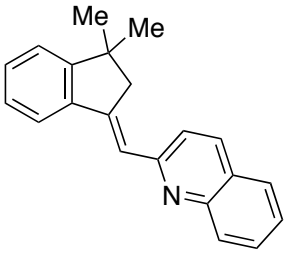



**4c**

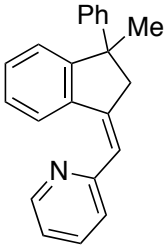
**(*E*)-1,1-Dimethyl-3-[(6-methyl-2-pyridyl)methylene]indane (3c) and 1,1-dimethyl-3-[(6-methyl-2-pyridyl)methyl]indene (4c).** The general procedure was followed using **1c** (37.3 mg, 0.150 mmol), $RhCl(PPh_3)_3$ (5.6 mg, 6.1 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.5 mL) for 24 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1, twice) yielded **3c** (23.7 mg, 0.095 mmol, 64%) as a pale yellow oil and **4c** (10.0 mg, 0.040 mmol, 27%) as a yellow oil.

**3c:** $^1$H NMR ($CDCl_3$, 301 MHz) δ 1.34 (s, 6H), 2.58 (s, 3H), 3.12 (d, $J$ = 3.0 Hz, 2H), 6.95 (d, $J$ = 7.5 Hz, 1H), 7.06 (t, $J$ = 2.6 Hz, 1H), 7.20–7.30 (m, 4H), 7.56 (t, $J$ = 7.8 Hz, 1H), 7.60–7.66 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 24.7, 30.1, 42.8, 48.2, 119.5, 120.1, 120.2, 120.6, 122.7, 126.8, 129.0, 136.2, 140.6, 146.2, 155.2, 156.5, 157.9; HRMS (ESI) calcd for $C_{18}H_{20}N$ $[M + H]^+$ 250.1590, found 250.1590.

**4c:** $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.32 (s, 6H), 2.58 (s, 3H), 4.02 (s, 2H), 6.06 (t, $J$ = 1.0 Hz, 1H), 6.95 (d, $J$ = 7.5 Hz, 1H), 6.99 (d, $J$ = 7.5 Hz, 1H), 7.16–7.20 (m, 3H), 7.29–7.33 (m, 1H), 7.45 (t, $J$ = 7.7 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 24.5, 24.6, 36.9, 48.3, 119.7, 119.8, 120.8, 121.0, 125.1, 126.2, 136.7, 137.7, 142.9, 143.8, 154.1, 157.7, 158.9; HRMS (ESI) calcd for $C_{18}H_{20}N$ $[M + H]^+$ 250.1590, found 250.1593.

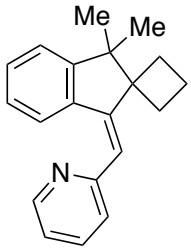



**(*E*)-1,1-Dimethyl-3-(2-quinolylmethylene)indane (3d).** The general procedure was followed using **1d** (40.0 mg, 0.140 mmol), $RhCl(PPh_3)_3$ (5.6 mg, 6.1 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.5 mL) for 35 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1, twice) yielded **3d** (14.6 mg, 0.051 mmol, 37%) as a pale yellow solid. Mp 60–65 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.38 (s, 6H), 3.34 (d, *J* = 3.0 Hz, 2H), 7.23 (t, *J* = 2.7 Hz, 1H), 7.26–7.35 (m, 3H), 7.46–7.50 (m, 1H), 7.53–7.56 (m, 1H), 7.67–7.72 (m, 2H), 7.75–7.79 (m, 1H), 8.07–8.12 (m, 2H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.1, 42.9, 48.5, 119.3, 120.8, 122.1, 122.9, 125.8, 126.3, 126.9, 127.4, 129.2, 129.4, 135.7, 140.5, 148.3, 148.8, 155.8, 157.3; HRMS (ESI) calcd for $C_{21}H_{20}N$ $[M + H]^+$ 286.1590, found 286.1590.

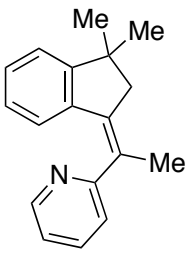



**(*Z*)-1-Methyl-1-phenyl-3-(2-pyridylmethylene)indane (2e).** The general procedure was followed using **1e** (32.7 mg, 0.110 mmol), $RhCl(PPh_3)_3$ (4.1 mg, 4.4 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.1 mL) for 2 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1, twice) yielded **2e** (26.0 mg, 0.087 mmol, 80%) as a pale yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.75 (s, 3H), 3.10 (dd, *J* = 15.8, 2.2 Hz, 1H), 3.28 (dd, *J* = 15.8, 1.8 Hz, 1H), 6.61 (t, *J* = 2.0 Hz, 1H), 7.04 (d, *J* = 7.0 Hz, 1H), 7.08–7.25 (m, 4H), 7.27–7.30 (m, 4H), 7.41 (d, *J*

= 8.0 Hz, 1H), 7.64 (dt, *J* = 2.0, 7.8 Hz, 1H), 7.94 (d, *J* = 8.0 Hz, 1H), 8.66–8.69 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 27.8, 49.0, 54.5, 121.4, 121.8, 123.9, 124.6, 125.4, 125.9, 126.3, 126.6, 128.1, 129.3, 136.0, 138.1, 144.6, 148.8, 149.3, 156.6, 156.7; HRMS (ESI) calcd for $C_{22}H_{19}NNa$ $[M + Na]^+$ 320.1410, found 320.1413.

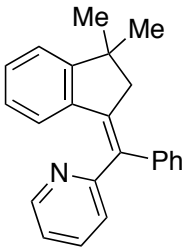



**(*Z*)-1′,1′-Dimethyl-3′-(2-pyridylmethylene)spiro[cyclobutane-1,2′-indane] (2g).** The general procedure was followed using **1g** (29.7 mg, 0.108 mmol), $RhCl(PPh_3)_3$ (10.2 mg, 11.0 μmol, 10 mol% Rh), and *p*-xylene (1.1 mL) for 22.5 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 7:1, twice) yielded **2e** (20.4 mg, 0.074 mmol, 69%) as a yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.21 (s, 6H), 1.75–1.84 (m, 1H), 1.92–2.00 (m, 2H), 2.11–2.30 (m, 3H), 6.83 (s, 1H), 6.97–7.03 (m, 1H), 7.12–7.17 (m, 1H), 7.19–7.24 (m, 2H), 7.41–7.52 (m, 2H), 7.55–7.61 (m, 1H), 8.65–8.68 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 14.5, 24.0, 27.2, 45.6, 60.2, 119.1, 121.3, 122.4, 123.8, 125.5, 125.6, 128.9, 135.9, 136.5, 149.4, 152.3, 154.8, 157.4; HRMS (MALDI) calcd for $C_{20}H_{21}NNa$ $[M + Na]^+$ 298.1566, found 298.1552.

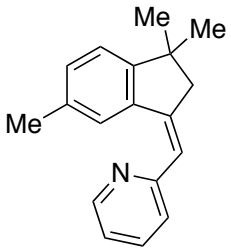



**(*Z*)-1,1-Dimethyl-3-[1-(2-pyridyl)ethylidene]indane (2h).** The general procedure was followed using **1h** (37.9 mg, 0.152 mmol), $RhCl(PPh_3)_3$ (13.9 mg, 15.0 μmol, 10 mol% Rh),

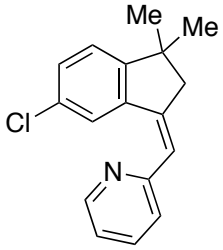
and *p*-xylene (1.5 mL) for 24 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1, twice) yielded **2h** (17.5 mg, 0.070 mmol, 46%) as a pale yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.34 (s, 6H), 2.16–2.19 (m, 3H), 2.73–2.75 (m, 2H), 6.15 (d, *J* = 8.0 Hz, 1H), 6.76–6.81 (m, 1H), 7.05–7.10 (m, 1H), 7.16 (d, *J* = 7.5 Hz, 1H), 7.21–7.25 (m, 1H), 7.27–7.31 (m, 1H), 7.65 (dt, *J* = 1.8, 7.7 Hz, 1H), 8.69–8.72 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 22.0, 30.0, 41.2, 48.0, 121.8, 122.4, 123.68, 123.69, 125.8, 127.3, 129.4, 136.6, 137.6, 138.6, 150.0, 156.0, 162.3; HRMS (ESI) calcd for $C_{18}H_{20}N$ $[M + H]^+$ 250.1590, found 250.1595.




**(*Z*)-1,1-Dimethyl-3-[phenyl(2-pyridyl)methylene]indane (2i).** The general procedure was followed using **1i** (31.3 mg, 0.101 mmol), $RhCl(PPh_3)_3$ (9.3 mg, 10.1 μmol, 10 mol% Rh), and *p*-xylene (1.0 mL) for 18 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 3:1) yielded **2i** (18.2 mg, 0.058 mmol, 58%) as a yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.30 (s, 6H), 2.85 (s, 2H), 6.15 (d, *J* = 8.0 Hz, 1H), 6.81–6.86 (m, 1H), 7.11–7.16 (m, 1H), 7.17–7.21 (m, 1H), 7.22–7.30 (m, 4H), 7.30–7.35 (m, 3H), 7.64–7.69 (m, 1H), 8.69–8.73 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 29.3, 41.9, 50.4, 122.0, 122.2, 124.6, 125.4, 125.8, 126.7, 128.0, 128.1, 129.1, 134.7, 136.5, 139.0, 140.6, 142.0, 150.2, 156.1, 161.0; HRMS (ESI) calcd for $C_{23}H_{22}N$ $[M + H]^+$ 312.1747, found 312.1744.

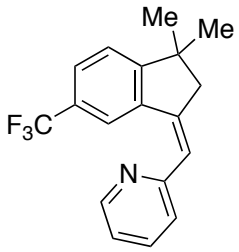



**(*Z*)-1,1,5-Trimethyl-3-(2-pyridylmethylene)indane (2j).** The general procedure was followed using **1j** (37.4 mg, 0.150 mmol), $RhCl(PPh_3)_3$ (5.4 mg, 5.8 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.5 mL) for 40 min. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1) yielded **2j** (31.4 mg, 0.126 mmol, 84%) as a yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.32 (s, 6H), 2.22 (s, 3H), 2.79 (d, *J* = 1.5 Hz, 2H), 6.59 (t, *J* = 2.0 Hz, 1H), 7.04–7.08 (m, 1H), 7.10–7.16 (m, 2H), 7.42 (d, *J* = 8.0 Hz, 1H), 7.59–7.65 (m, 2H), 8.64–8.67 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 21.4, 29.4, 40.9, 52.3, 121.2, 121.4, 122.2, 123.8, 125.8, 130.1, 135.3, 135.9, 137.5, 145.0, 149.2, 155.0, 156.9; HRMS (ESI) calcd for $C_{18}H_{20}N$ $[M + H]^+$ 250.1590, found 250.1591.

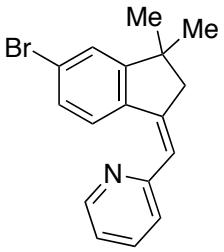



**(*Z*)-5-Chloro-1,1-dimethyl-3-(2-pyridylmethylene)indane (2k).** The general procedure was followed using **1k** (40.7 mg, 0.151 mmol), $RhCl(PPh_3)_3$ (5.4 mg, 5.8 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.5 mL) for 50 min. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1) yielded **2k** (35.4 mg, 0.131 mmol, 87%) as a yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.32 (s, 6H), 2.82 (d, *J* = 1.5 Hz, 2H), 6.62 (t, *J* = 2.0 Hz, 1H), 7.14 (d, *J* = 8.0 Hz, 1H), 7.15–7.19 (m, 1H), 7.21 (dd, *J* = 8.3, 2.2 Hz, 1H), 7.34 (d, *J* = 8.0 Hz, 1H), 7.66 (dt, *J* = 1.5, 7.7 Hz, 1H), 8.08 (d, *J* = 2.5 Hz, 1H), 8.67–8.70 (m, 1H); $^{13}$C NMR ($CDCl_3$,

75.6 MHz) δ 29.2, 40.9, 52.2, 121.6, 122.8, 123.4, 123.9, 125.9, 129.1, 131.7, 136.2, 139.1, 143.7, 149.2, 156.01, 156.04; HRMS (ESI) calcd for $C_{17}H_{17}ClN$ $[M + H]^+$ 270.1044, found 270.1044.

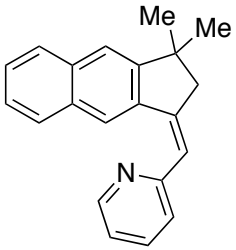



**(*Z*)-1,1-Dimethyl-3-(2-pyridylmethylene)-5-(trifluoromethyl)indane (2l).** The general procedure was followed using **1l** (30.3 mg, 0.100 mmol), $RhCl(PPh_3)_3$ (3.7 mg, 4.0 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.0 mL) for 75 min. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1) yielded **2l** (24.8 mg, 0.082 mmol, 82%) as a white solid. Mp 81–85 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.35 (s, 6H), 2.86 (d, $J$ = 2.0 Hz, 2H), 6.67 (t, $J$ = 2.0 Hz, 1H), 7.15–7.20 (m, 1H), 7.29–7.33 (m, 2H), 7.48–7.52 (m, 1H), 7.67 (dt, $J$ = 2.0, 7.8 Hz, 1H), 8.57 (s, 1H), 8.66–8.69 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 29.1, 41.4, 52.1, 121.7, 122.7, 123.2, 123.5 (q, $^3J_{C–F}$ = 4.9 Hz), 124.0, 124.4 (q, $^1J_{C–F}$ = 272.2 Hz), 125.9 (q, $^3J_{C–F}$ = 3.6 Hz), 128.4 (q, $^2J_{C–F}$ = 31.6 Hz), 136.2, 138.0, 143.6, 149.1, 155.8, 161.1; HRMS (ESI) calcd for $C_{18}H_{17}F_3N$ $[M + H]^+$ 304.1308, found 304.1308.

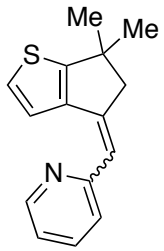



**(*Z*)-6-Bromo-1,1-dimethyl-3-(2-pyridylmethylene)indane (2m).** The general procedure was followed using **1m** (48.0 mg, 0.153 mmol), $RhCl(PPh_3)_3$ (13.9 mg, 15.0 $\mu$mol,

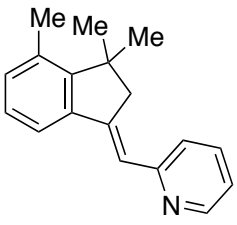
10 mol% Rh), and *p*-xylene (1.5 mL) for 9.5 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 6:1, thrice, then 8:1) yielded **2m** (21.6 mg, 0.069 mmol, 45%) as a yellow solid. Mp 64–69 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.32 (s, 6H), 2.81 (d, *J* = 2.5 Hz, 2H), 6.61 (t, J = 2.0 Hz, 1H), 7.12–7.17 (m, 1H), 7.19 (dd, *J* = 8.5, 2.0 Hz, 1H), 7.32–7.35 (m, 2H), 7.64 (dt, *J* = 1.7, 7.6 Hz, 1H), 7.87 (d, *J* = 8.5 Hz, 1H), 8.63–8.67 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 29.2, 41.5, 52.0, 121.4, 122.3, 123.1, 123.9, 125.8, 127.2, 129.2, 136.1, 136.4, 143.7, 149.2, 156.4, 159.8; HRMS (ESI) calcd for $C_{17}H_{17}BrN$ $[M + H]^+$ 314.0539, found 314.0537.



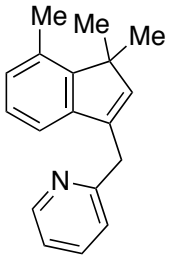

**(*Z*)-1,1-Dimethyl-3-(2-pyridylmethylene)-2,3-dihydro-1*H*-cyclopenta[*b*]naphthalene (2n).** The general procedure was followed using **1n** (42.8 mg, 0.150 mmol), $RhCl(PPh_3)_3$ (5.6 mg, 6.1 μmol, 4 mol% Rh), and *p*-xylene (1.5 mL) for 7.5 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1, twice) yielded **2n** (27.0 mg, 0.095 mmol, 63%) as a white solid. Mp 73–75 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.44 (s, 6H), 2.88 (d, *J* = 2.5 Hz, 2H), 6.73 (t, *J* = 2.0 Hz, 1H), 7.17–7.22 (m, 1H), 7.32–7.36 (m, 1H), 7.38–7.43 (m, 1H), 7.51 (d, *J* = 8.0 Hz, 1H), 7.62 (s, 1H), 7.63–7.68 (m, 2H), 7.76 (d, *J* = 7.5 Hz, 1H), 8.43 (s, 1H), 8.72–8.75 (m, 1H); $^{13}$C NMR ($CDCl_3$, 75.6 MHz) δ 29.5, 41.0, 52.7, 120.1, 121.5, 123.0, 123.9, 124.8, 125.0, 126.1, 127.3, 128.9, 132.4, 134.2, 136.0, 136.2, 144.3, 149.3, 155.2, 156.7; HRMS (ESI) calcd for $C_{21}H_{20}N$ $[M + H]^+$ 286.1590, found 286.1590.



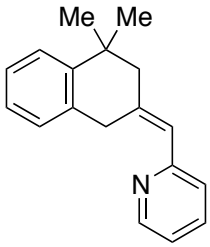

**(*Z*)- and (*E*)-6,6-Dimethyl-4-(2-pyridylmethylene)-5,6-dihydro-4*H*-cyclopenta[*b*]thiophene (2o and 3o).** The general procedure was followed using **1o** (32.8 mg, 0.136 mmol), $RhCl(PPh_3)_3$ (13.9 mg, 15.0 $\mu$mol, 11 mol% Rh), and *p*-xylene (1.5 mL) for 24 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 4:1) yielded a mixture of **2o** and **3o** (17.7 mg, 0.073 mmol, 54%; **2o**:**3o** = 54:46 determined by $^1$H NMR) as a yellow oil. Spectral data were obtained as a mixture of the two compounds. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.41 (s, 6H; major), 1.43 (s, 6H; minor), 3.15 (d, $J$ = 1.5 Hz, 2H; major), 3.48 (d, $J$ = 2.0 Hz, 2H; minor), 6.28 (t, $J$ = 2.0 Hz, 1H; major), 6.65 (t, $J$ = 2.3 Hz, 1H; minor), 7.01–7.13 (m, 2H), 7.23–7.28 (m, 1H), 7.58–7.63 (m, 2H), 8.58–8.65 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 30.5, 30.8, 40.9, 42.7, 53.0, 55.7, 116.6, 117.0, 118.6, 120.0, 120.6, 123.0, 123.4, 124.6, 126.6, 128.9, 135.8, 135.9, 141.2, 142.1, 143.3, 146.0, 149.0, 149.2, 156.8, 157.5, 160.9, 165.1; HRMS (ESI) calcd for $C_{15}H_{16}NS$ $[M + H]^+$ 2242.0998, found 242.0997.




**3p**




**4p**

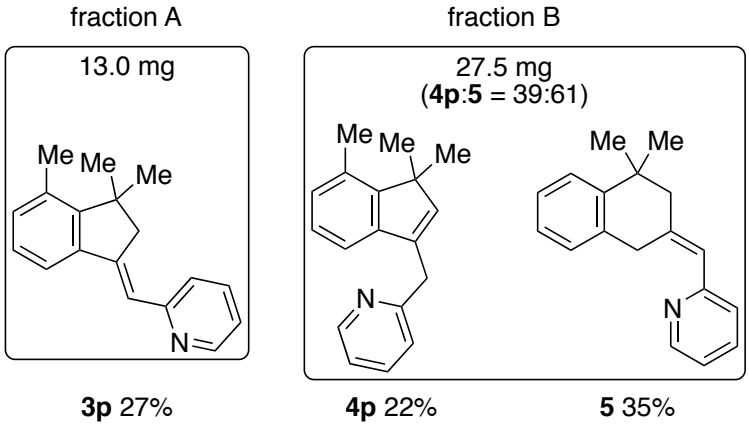



**5**

**(*E*)-1,1,7-Trimethyl-3-(2-pyridylmethylene)indane (3p), 1,1,7-trimethyl-3-(2-pyridylmethyl)indene (4p) and (*E*)-1,1-dimethyl-3-(2-pyridylmethylene)-1,2,3,4-tetrahydronaphthalene (5).** The general procedure was followed using **1p** (47.7 mg, 0.191

mmol), $RhCl(PPh_3)_3$ (7.4 mg, 8.0 $\mu$mol, 4 mol% Rh), and *p*-xylene (2.0 mL) for 18.5 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 5:1, twice) yielded two fractions, A and B.

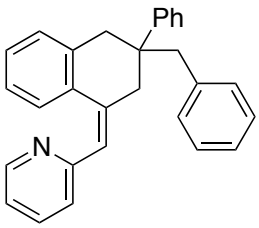
The fraction A contained **3p** (13.0 mg, 0.052 mmol, 27%), and the fraction B contained **4p** and **5** (27.5 mg, 0.110 mmol, 58%, **4p**:**5** = 39:61 determined by $^1$H NMR).




**3p:** $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.44 (s, 6H), 2.45 (s, 3H), 3.18 (d, *J* = 2.5 Hz, 2H), 7.02 (t, *J* = 2.5 Hz, 1H), 7.04–7.09 (m, 2H), 7.17 (t, *J* = 7.5 Hz, 1H), 7.36 (d, *J* = 8.0 Hz, 1H), 7.51 (d, *J* = 7.5 Hz, 1H), 7.65 (dt, *J* = 1.7, 7.7 Hz, 1H), 8.62–8.65 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 19.3, 28.2, 44.1, 50.1, 118.4, 118.6, 120.4, 123.5, 127.1, 132.0, 134.3, 136.0, 141.2, 146.9, 149.3, 151.7, 157.3; HRMS (ESI) calcd for $C_{18}H_{20}N$ $[M + H]^+$ 250.1590, found 250.1590.

**4p and 5:** Spectral data were obtained as a mixture of the two compounds. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.16 (s, 6H; **5**), 1.41 (s, 6H; **4p**), 2.13 (s, 2H; **5**), 2.47 (s, 3H; **4p**), 3.71 (s, 2H; **5**), 4.03 (s, 2H; **4p**), 6.00 (s, 1H; **4p**), 6.35 (s, 1H; **5**), 6.93 (d, *J* = 7.0 Hz), 7.00–7.05 (m), 7.07–7.20 (m), 7.23–7.28 (m), 7.55–7.67 (m), 8.54–8.59 (m); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 18.7, 22.1, 28.3, 34.0, 36.9, 42.2, 46.7, 49.3, 117.5, 121.2, 121.3, 122.8, 123.3, 123.5, 124.1, 126.1, 126.4, 126.6, 127.1, 127.6, 132.6, 133.3, 136.3, 136.4, 136.8, 137.5, 143.0, 143.5,

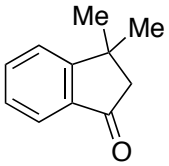
145.0, 149.1, 149.2, 150.3, 159.3, 159.6; HRMS (ESI) calcd for $C_{18}H_{20}N$ $[M + H]^+$ 250.1590, found 250.1590.



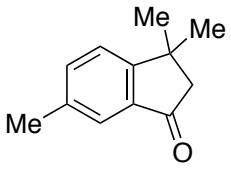

**(*Z*)-3-Benzyl-3-methyl-1-(2-pyridylmethylene)-1,2,3,4-tetrahydronaphthalene (2q).** The general procedure was followed using **1q** (30.6 mg, 0.094 mmol), $RhCl(PPh_3)_3$ (3.7 mg, 4.0 $\mu$mol, 4 mol% Rh), and *p*-xylene (1.0 mL) for 10 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1) yielded **6** (17.4 mg, 0.053 mmol, 57%) as a yellow oil. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.04 (s, 3H), 2.37 (d, *J* = 12.5 Hz, 1H), 2.46–2.52 (m, 1H), 2.59 (d, *J* = 17.0 Hz, 1H), 2.68 (d, *J* = 13.5 Hz, 1H), 2.76 (d, *J* = 13.0 Hz, 1H), 2.89 (d, *J* = 16.5 Hz, 1H), 6.51 (s, 1H), 6.81–6.87 (m, 1H), 7.05–7.33 (m, 10H), 7.44 (dt, *J* = 1.5, 7.7 Hz, 1H), 8.58–8.62 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 25.7, 35.8, 41.7, 47.4, 48.1, 121.2, 124.2, 124.6, 126.1, 126.2, 127.8, 128.2, 128.8, 129.5, 130.7, 133.2, 135.6, 137.7, 138.3, 139.4, 149.6, 157.9; HRMS (ESI) calcd for $C_{24}H_{24}N$ $[M + H]^+$ 326.1903, found 326.1903.

**General Procedure for the Rhodium(I)-Catalysed Skeletal Reorganisation of Cyclobutanones 6.** A Schlenk tube was charged with cyclobutanone **6** (0.200 mmol), $RhCl(PPh_3)_3$ (10 $\mu$mol, 5 mol% Rh), benzoic acid (0.100 mmol, 0.5 equiv), and aminopyridine **7a** (0.240 mmol, 1.2 equiv, liquid **7b** was added via syringe after *p*-xylene). The tube was evacuated and backfilled with nitrogen. *p*-Xylene (2.0 mL) was added via syringe through the septum, and the mixture was heated at 150 °C for the indicated time.

After cooling to room temperature, HCl aq. (1.0 M, 0.20 mL) was added to the reaction mixture, and the mixture was stirred at room temperature for 1.5 h. The mixture was filtered through a plug of Florisil® washing with hexane–AcOEt (1:1), and the filtrate was concentrated under reduced pressure. The residue was purified by preparative TLC on silica gel to afford the following compounds.



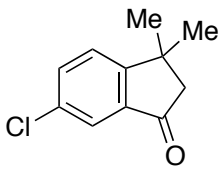

**3,3-Dimethyl-1-indanone (8a).** The general procedure was followed using **6a** (80.8 mg, 0.504 mmol), $RhCl(PPh_3)_3$ (23.1 mg, 25 $\mu$mol), benzoic acid (30.8 mg, 0.252 mmol), 2-aminopyridine (**7a**, 57.0 mg, 0.606 mmol) and *p*-xylene (5.0 mL) for 22.5 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1, twice) yielded **8a** (68.0 mg, 0.424 mmol, 84%) as a colourless oil. The reaction of **6a** (29.9 mg, 0.187 mmol) employing 2-amino-3-picoline (**7b**) afforded **8a** (23.6 mg, 0.147 mmol) in 79% yield. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.42 (s, 6H), 2.59 (s, 2H), 7.34–7.38 (m, 1H), 7.48–7.51 (m, 1H), 7.59–7.63 (m, 1H), 7.68–7.71 (m, 1H). The spectral data matched those reported in the literature.[1] HRMS (ESI) calcd for $C_{11}H_{12}NaO$ $[M + Na]^+$ 183.0780, found 183.0781; IR ($\nu$/cm$^{-1}$): 1712, 1604, 1242, 764.



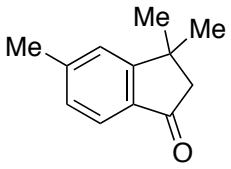

**3,3,6-Trimethyl-1-indanone (8b).** The general procedure was followed using **6b** (34.1 mg, 0.196 mmol), $RhCl(PPh_3)_3$ (9.3 mg, 10 $\mu$mol), benzoic acid (12.5 mg, 0.102 mmol), 2-

1 L.-Q. Cui, K. Liu and C. Zhang, *Org. Biomol. Chem.*, 2011, **9**, 2258.

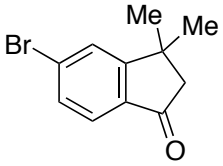
amino-3-picoline (**7b**, 26.0 mg, 0.240 mmol) and *p*-xylene (2.0 mL) for 12 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 12:1, twice) yielded **8b** (25.2 mg, 0.145 mmol, 74%) as a yellow oil. The reaction of **6b** (34.8 mg, 0.200 mmol) employing 2-aminopyridine (**7a**) afforded **8b** (22.5 mg, 0.129 mmol) in 65% yield. $^{1}$H NMR ($CDCl_3$, 500 MHz) δ 1.40 (s, 6H), 2.40 (s, 3H), 2.58 (s, 2H), 7.37–7.40 (m, 1H), 7.42–7.45 (m, 1H), 7.49–7.51 (m, 1H); $^{13}$C NMR (DMSO-$d_6$, 126 MHz) δ 20.5, 29.7, 37.9, 52.5, 122.4, 123.7, 135.0, 136.1, 137.0, 161.1, 204.9; HRMS (ESI) calcd for $C_{12}H_{14}NaO$ $[M + Na]^+$ 197.0937, found 197.0937; IR ($\nu$/cm$^{-1}$): 2962, 1712, 1288.




**6-Chloro-3,3-dimethyl-1-indanone (8c).** The general procedure was followed using **6c** (38.3 mg, 0.197 mmol), $RhCl(PPh_3)_3$ (9.3 mg, 10 $\mu$mol), benzoic acid (12.3 mg, 0.101 mmol), 2-amino-3-picoline (**7b**, 25.3 mg, 0.234 mmol) and *p*-xylene (2.0 mL) for 13 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1, twice) yielded **8c** (29.1 mg, 0.149 mmol, 76%) as a yellow solid. The reaction of **6c** (30.2 mg, 0.155 mmol) employing 2-aminopyridine (**7a**) afforded **8c** (22.0 mg, 0.113 mmol) in 73% yield. Mp 123–127 °C; $^{1}$H NMR ($CDCl_3$, 500 MHz) δ 1.41 (s, 6H), 2.60 (s, 2H), 7.43 (d, $J$ = 8.5 Hz, 1H), 7.55 (dd, $J$ = 8.0, 2.5 Hz, 1H), 7.63 (d, $J$ = 1.5 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 29.8, 38.4, 53.0, 123.1, 124.9, 133.8, 134.9, 136.8, 161.8, 204.3; HRMS (ESI) calcd for $C_{11}H_{11}ClNaO$ $[M + Na]^+$ 217.0391, found 217.0392; IR ($\nu$/cm$^{-1}$): 1705, 1466, 1242.

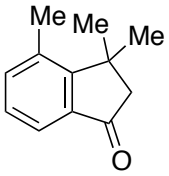



**3,3,5-Trimethyl-1-indanone (8d).** The general procedure was followed using **6d** (34.8 mg, 0.200 mmol), $RhCl(PPh_3)_3$ (9.3 mg, 10 $\mu$mol), benzoic acid (12.3 mg, 0.101 mmol), 2-amino-3-picoline (**7b**, 25.9 mg, 0.239 mmol) and *p*-xylene (2.0 mL) for 15.5 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1, thrice) yielded **8d** (26.8 mg, 0.154 mmol, 77%) as a yellow oil. The reaction of **6c** (34.7 mg, 0.199 mmol) employing 2-aminopyridine (**7a**) afforded **8c** (17.6 mg, 0.101 mmol) in 51% yield. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.41 (s, 6H), 2.46 (s, 3H), 2.57 (s, 2H), 7.17 (d, $J$ = 8.5 Hz, 1H), 7.26–7.29 (m, 1H), 7.59 (d, $J$ = 8.0 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 22.2, 29.9, 38.3, 53.1, 123.1, 123.8, 128.6, 133.0, 146.0, 164.3, 205.4; HRMS (ESI) calcd for $C_{12}H_{14}NaO$ $[M + Na]^+$ 197.0937, found 197.0938; IR ($\nu$/cm$^{-1}$): 2962, 1712, 1604.

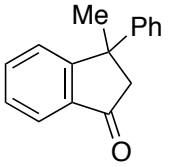



**5-Bromo-3,3-dimethyl-1-indanone (8e).** The general procedure was followed using **6e** (36.0 mg, 0.151 mmol), $RhCl(PPh_3)_3$ (6.9 mg, 7.5 $\mu$mol), benzoic acid (9.5 mg, 0.078 mmol), 2-aminopyridine (**7a**, 17.2 mg, 0.183 mmol) and *p*-xylene (1.5 mL) for 20 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 7:1) yielded **8e** (25.4 mg, 0.106 mmol, 71%) as a yellow solid. The reaction of **6e** (47.8 mg, 0.200 mmol) employing 2-amino-3-picoline (**7b**) afforded **8e** (29.3 mg, 0.123 mmol) in 61% yield. Mp 104–110 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.41 (s, 6H), 2.58 (s, 2H), 7.50 (dd, $J$ = 8.0, 1.5 Hz, 1H), 7.55 (d, $J$ = 8.0 Hz, 1H), 7.65 (d, $J$ = 1.5 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 29.8, 38.6, 52.8, 124.7, 127.1, 130.2,

131.0, 134.1, 165.3, 204.5; HRMS (ESI) calcd for $C_{11}H_{11}BrNaO$ $[M + Na]^+$ 260.9885, found 260.9885; IR ($\nu$/cm$^{-1}$): 1705, 1589, 1242, 802.

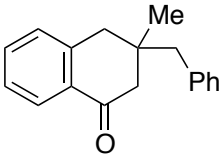



**3,3,4-Trimethyl-1-indanone (8f).** The general procedure was followed using **6f** (34.9 mg, 0.200 mmol), $RhCl(PPh_3)_3$ (9.3 mg, 10 $\mu$mol), benzoic acid (12.0 mg, 0.098 mmol), 2-amino-3-picoline (**7b**, 26.0 mg, 0.240 mmol) and *p*-xylene (2.0 mL) for 20 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1, twice) yielded **8f** (14.0 mg, 0.080 mmol, 40%) as a yellow solid. The reaction of **6f** (34.6 mg, 0.199 mmol) employing 2-aminopyridine (**7a**) afforded **8f** (9.8 mg, 0.056 mmol) in 28% yield. Mp 47–51 °C; $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.52 (s, 6H), 2.52 (s, 3H), 2.61 (s, 2H), 7.26 (t, $J$ = 7.2 Hz, 1H), 7.36 (d, $J$ = 7.5 Hz, 1H), 7.57 (d, $J$ = 7.5 Hz, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 19.6, 28.0, 39.8, 54.7, 121.4, 127.7, 135.4, 136.3, 137.7, 159.9, 206.2; HRMS (ESI) calcd for $C_{12}H_{14}NaO$ $[M + Na]^+$ 197.0937, found 197.0936; IR ($\nu$/cm$^{-1}$): 1705, 1281, 795.




**3-Methyl-3-phenyl-1-indanone (8g).** The general procedure was followed using **6g** (44.6 mg, 0.201 mmol), $RhCl(PPh_3)_3$ (9.3 mg, 10 $\mu$mol), benzoic acid (12.7 mg, 0.104 mmol), 2-amino-3-picoline (**7b**, 26.3 mg, 0.243 mmol) and *p*-xylene (2.0 mL) for 13.5 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1, twice) yielded **8g** (29.4 mg, 0.132 mmol, 66%) as a yellow oil. The reaction of **6g** (44.5 mg, 0.200 mmol) employing 2-

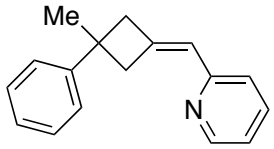
aminopyridine (**7a**) afforded **8g** (23.2 mg, 0.104 mmol) in 52% yield. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.84 (s, 3H), 2.88 (d, $J$ = 19.0 Hz, 1H), 3.00 (d, $J$ = 19.5 Hz, 1H), 7.17–7.23 (m, 3H), 7.26–7.31 (m, 3H), 7.40–7.45 (m, 1H), 7.58–7.62 (m, 1H), 7.77–7.81 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 28.3, 46.0, 55.7, 123.3, 125.6, 126.2, 126.4, 127.7, 128.5, 135.2, 135.7, 147.3, 162.8, 205.8. The spectral data matched those reported in the literature.[2] IR ($\nu$/cm$^{-1}$): 1712, 1597, 1234, 764, 702.




**3-Benzyl-3-methyl-1-tetralone (8h).** The general procedure was followed using **6h** (50.1 mg, 0.200 mmol), $RhCl(PPh_3)_3$ (9.3 mg, 10 $\mu$mol), benzoic acid (12.3 mg, 0.101 mmol), 2-amino-3-picoline (**7b**, 25.8 mg, 0.239 mmol) and *p*-xylene (2.0 mL) for 11 h. Purification by preparative TLC on silica gel (hexane:AcOEt = 10:1) yielded **8h** (38.3 mg, 0.153 mmol, 76%) as a yellow oil. The reaction of **6h** (30.2 mg, 0.121 mmol) employing 2-aminopyridine (**7a**) afforded **8h** (12.5 mg, 0.050 mmol) in 41% yield. $^1$H NMR ($CDCl_3$, 500 MHz) δ 1.03 (s, 3H), 2.50 (dd, $J$ = 16.5, 1.5 Hz, 1H), 2.60 (d, $J$ = 16.5 Hz, 1H), 2.63–2.71 (m, 2H), 2.77 (d, $J$ = 16.0 Hz, 1H), 2.95 (d, $J$ = 16.0 Hz, 1H), 7.06–7.10 (m, 2H), 7.21–7.34 (m, 5H), 7.48–7.53 (m, 1H), 8.02–8.05 (m, 1H); $^{13}$C NMR ($CDCl_3$, 126 MHz) δ 25.3, 37.5, 41.1, 46.8, 50.8, 126.4, 126.6, 126.7, 128.0, 129.4, 130.5, 131.9, 133.8, 137.3, 142.3, 198.2; HRMS (ESI) calcd for $C_{18}H_{18}NaO$ $[M + Na]^+$ 273.1250, found 273.1251; IR ($\nu$/cm$^{-1}$): 1682, 1604, 1288, 756.

2 T. Matsuda, M. Shigeno, M. Makino and M. Murakami, *Org. Lett.*, 2006, **8**, 3379.

**1a**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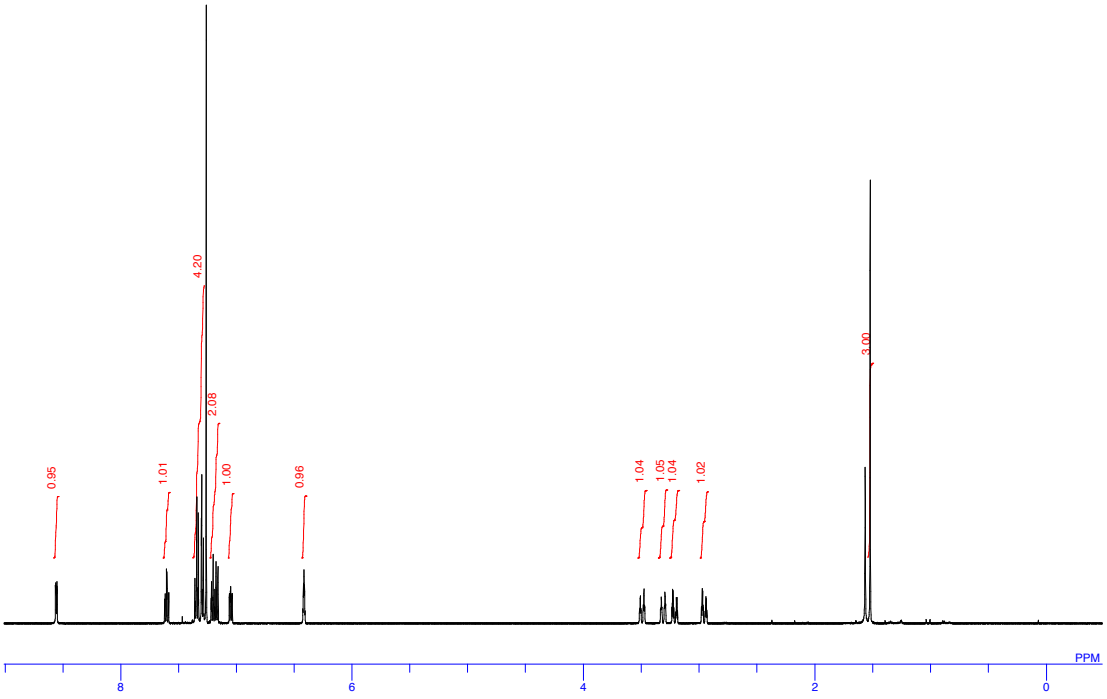
N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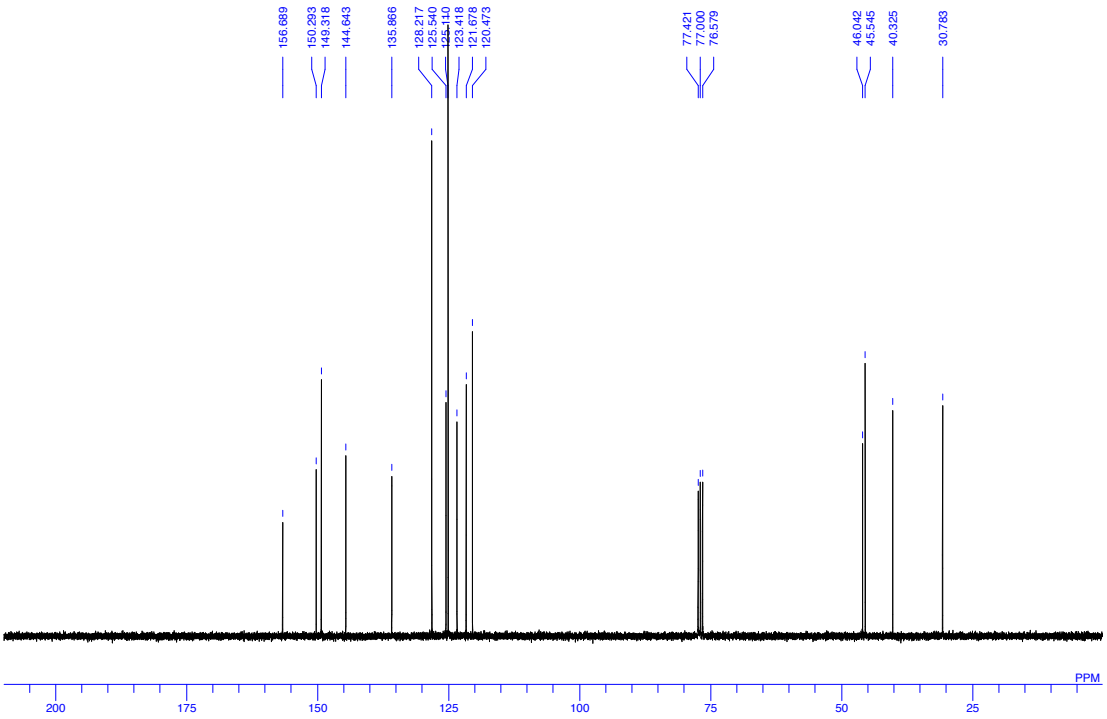
6
4
2
0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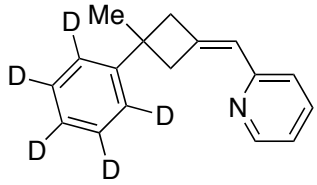
**1a**-$d_5$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Me
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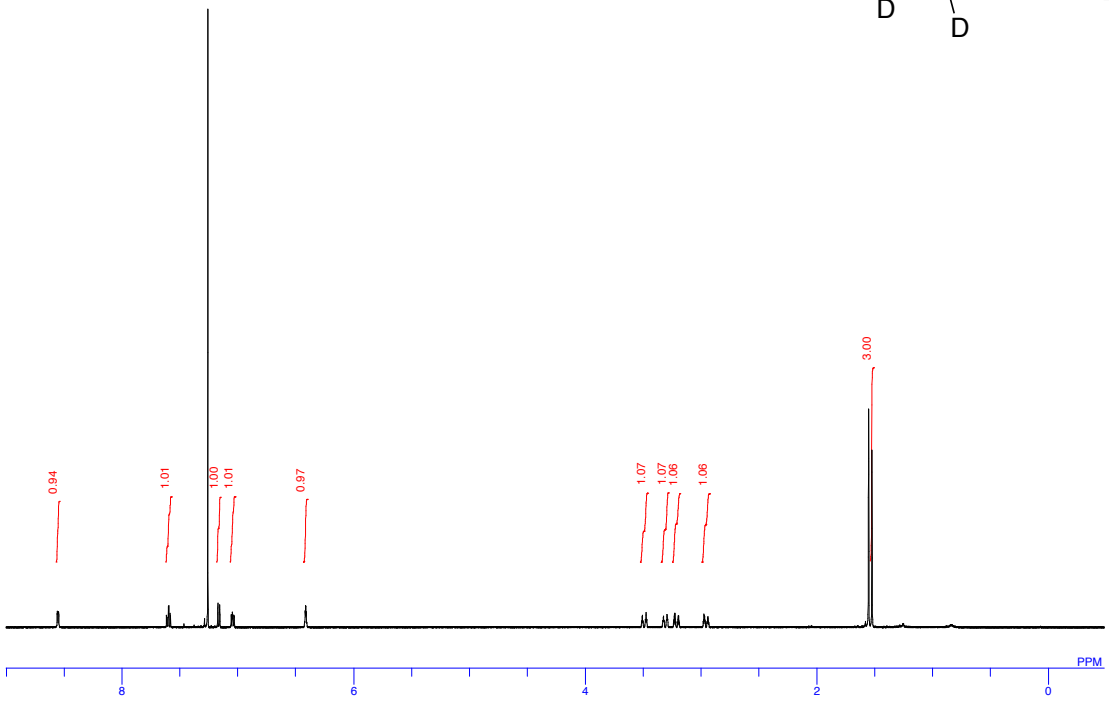
D
D
D
N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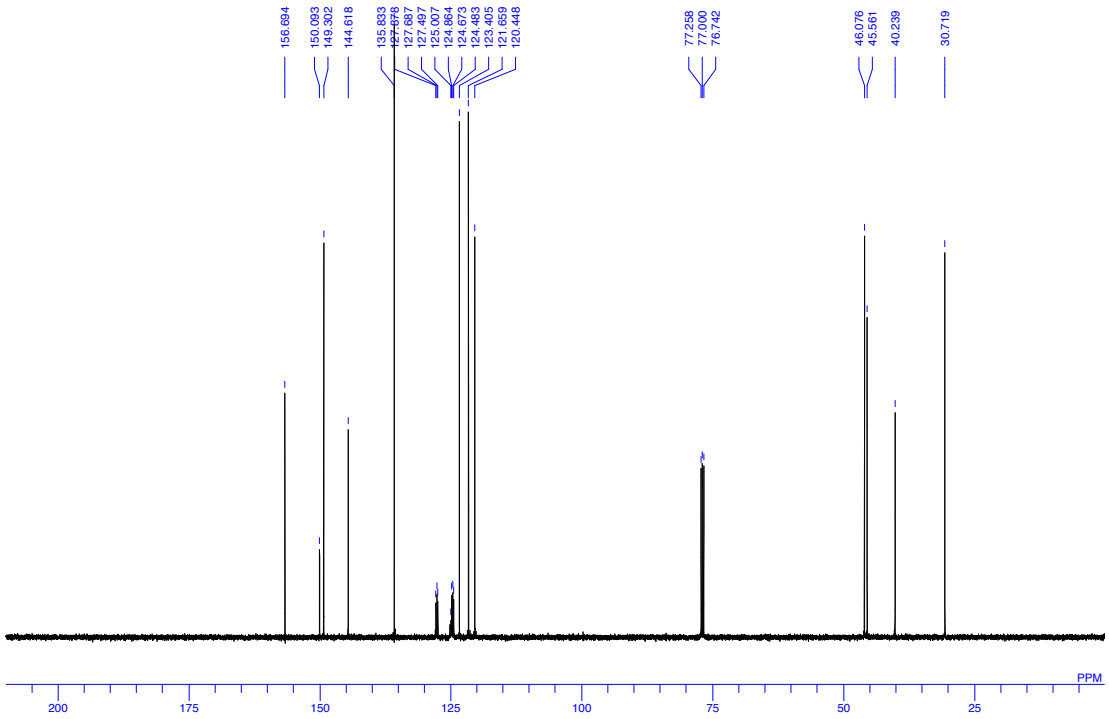
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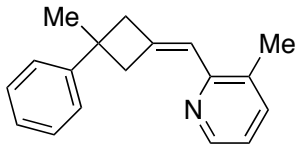
**1b**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Me
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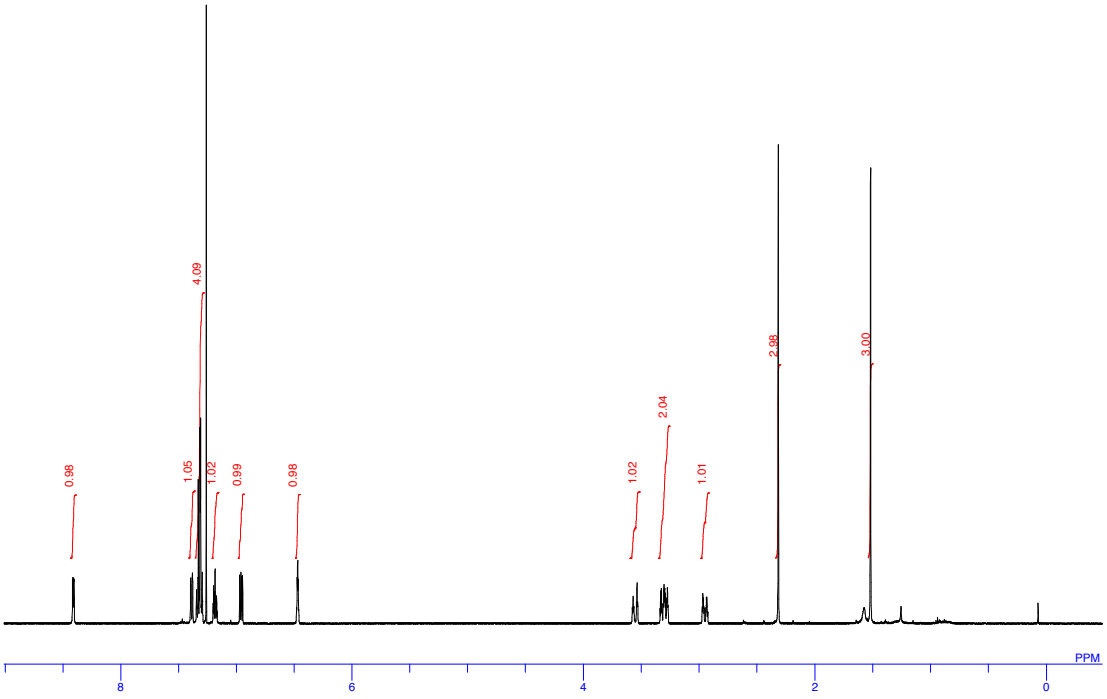
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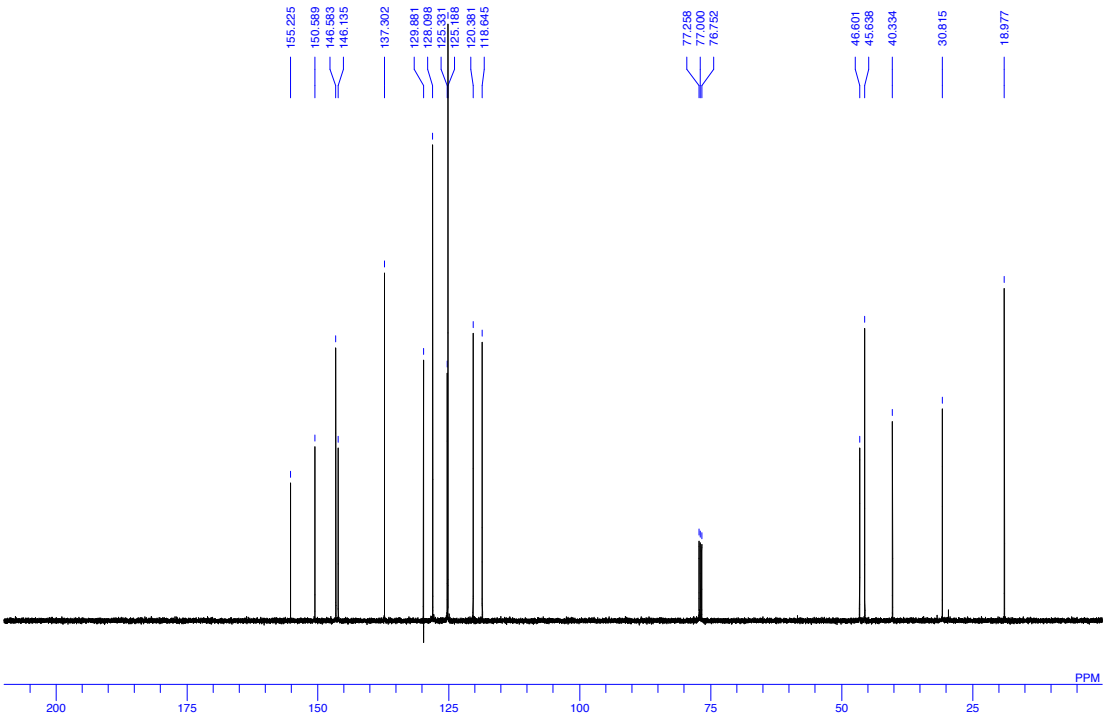
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50
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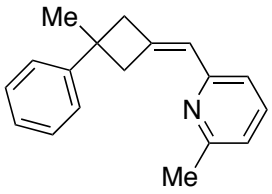
**1c**


Me
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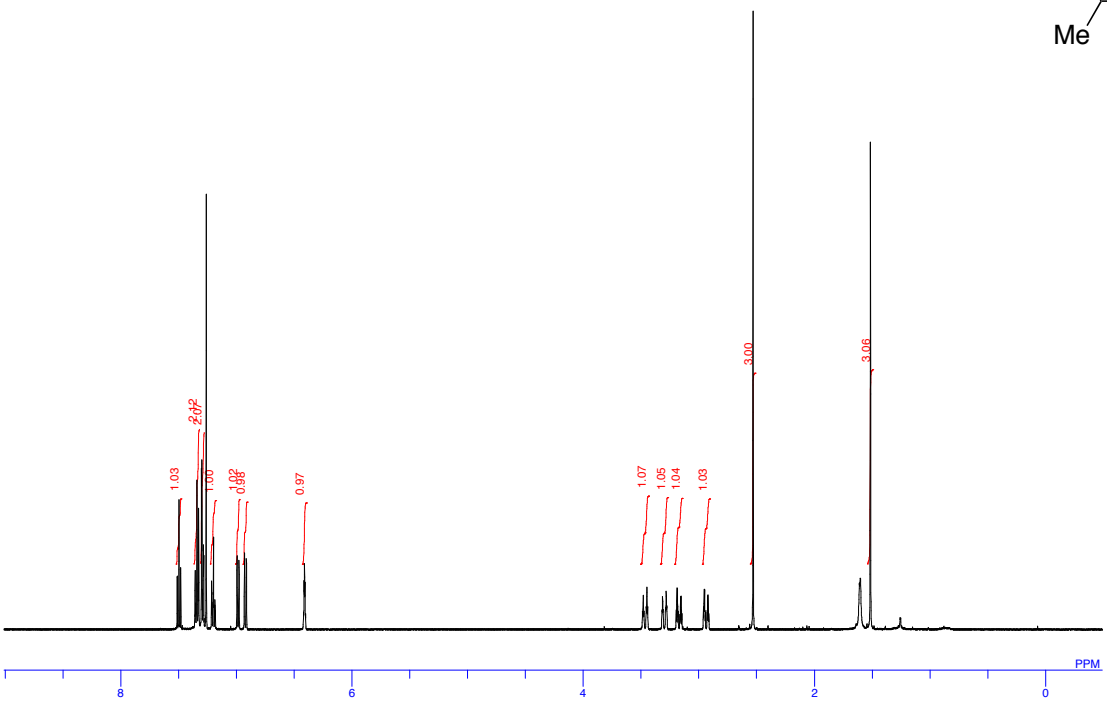
Me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2.12
2.07
1.00
1.02
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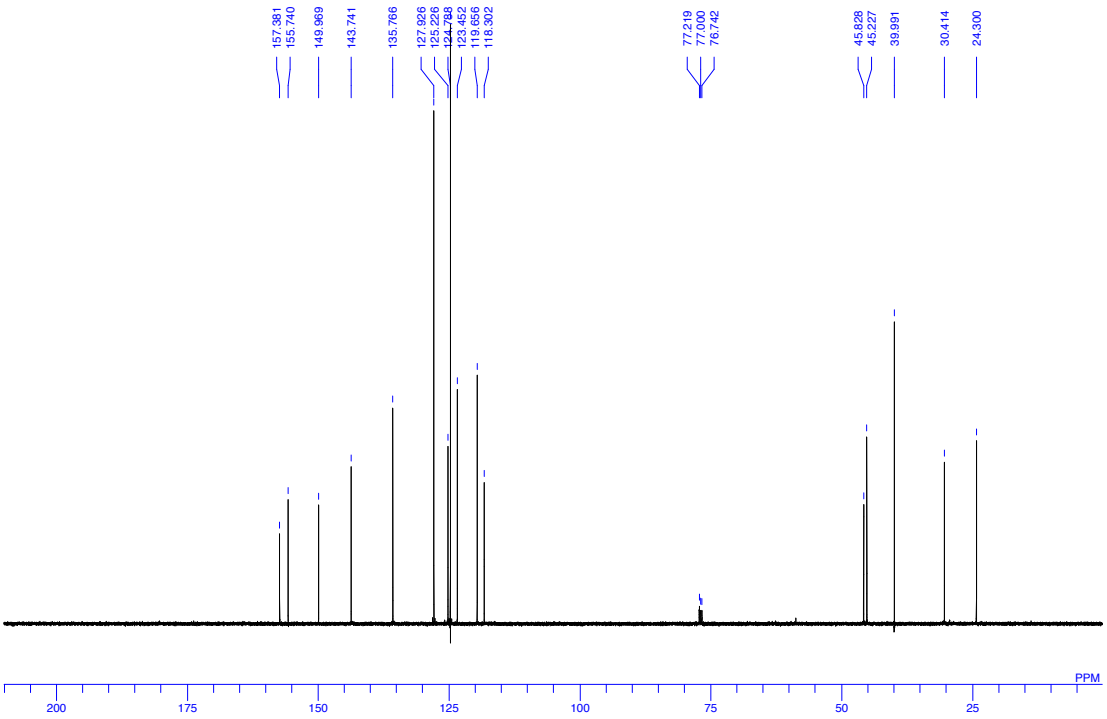
1.07
1.05
1.04
1.03
3.00
3.06
PPM
8
6
4
2
0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127.926
125.226
124.788
123.452
119.656
118.302
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77.000
76.742
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39.991
30.414
24.300
PPM
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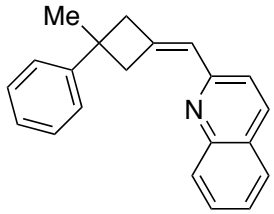
175
150
125
100
75
50
25

**1d**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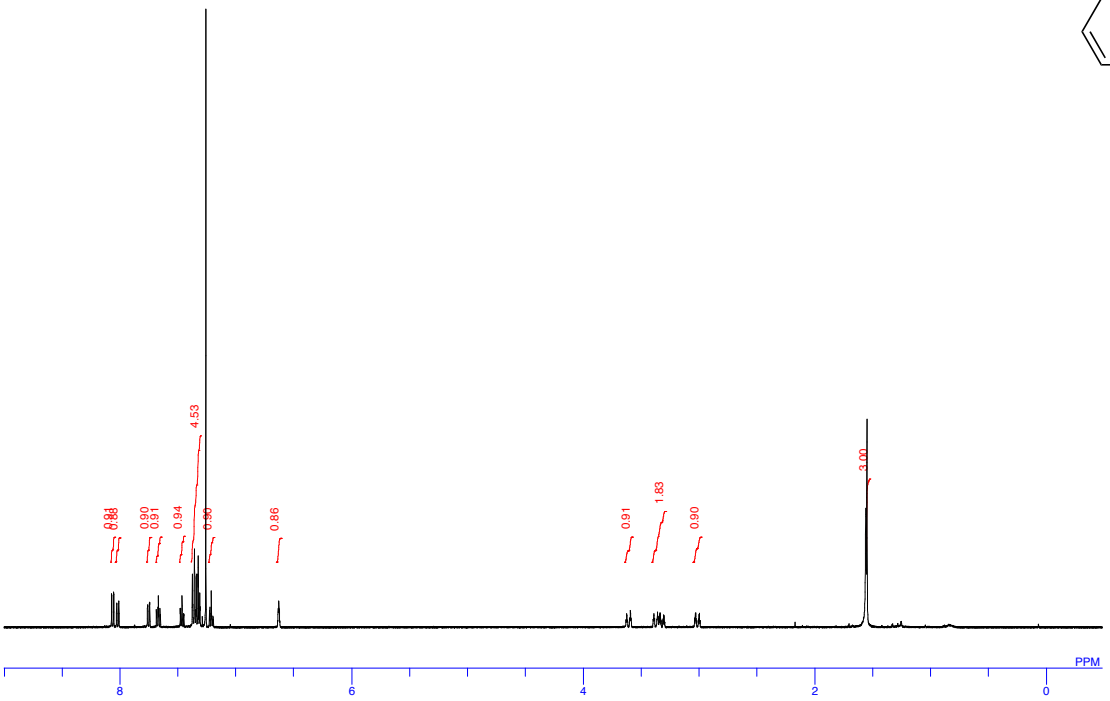
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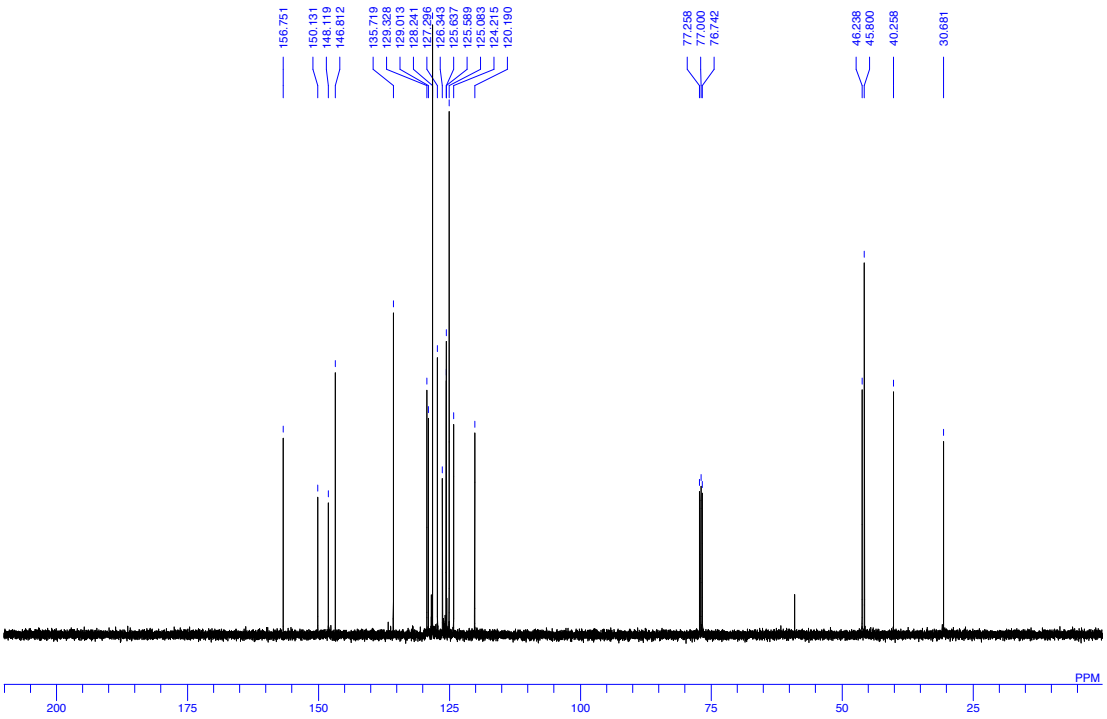
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126.343
125.637
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125.083
124.215
120.190
77.258
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40.258
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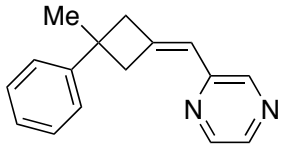
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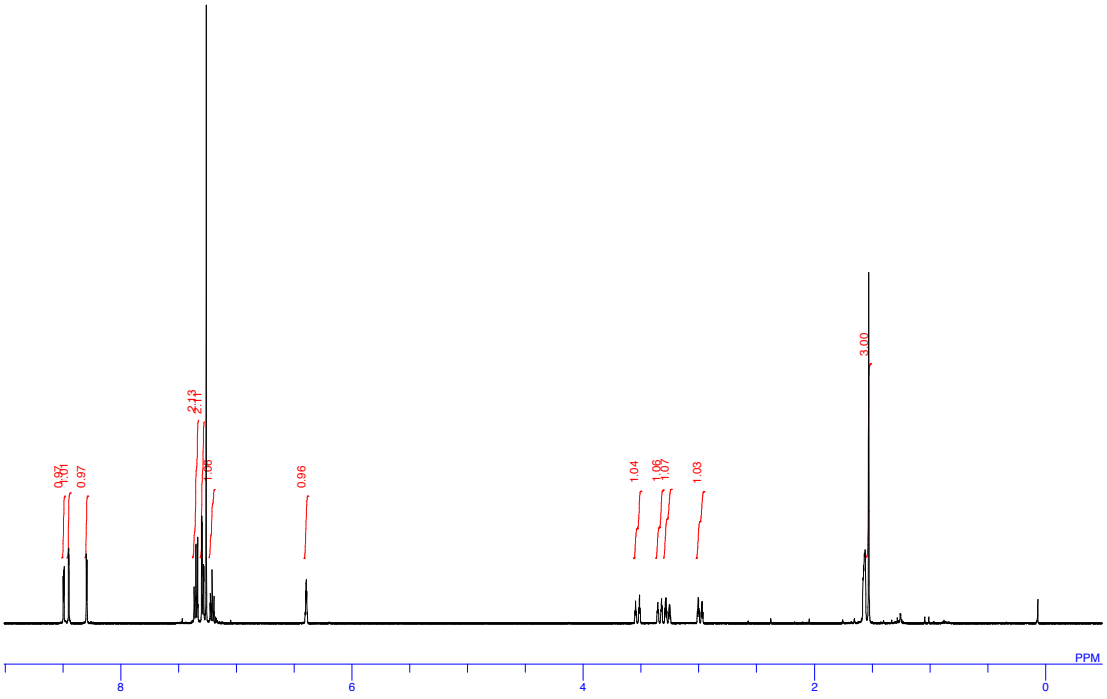
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1.04
1.06
1.07
1.03
3.00
PPM
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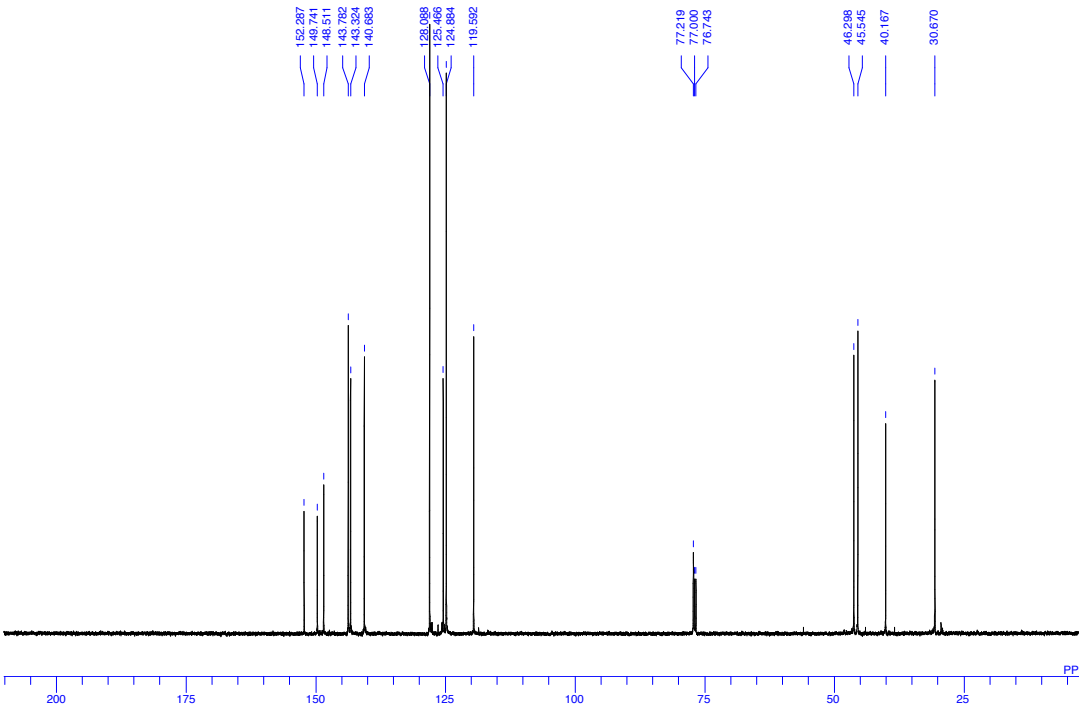
2
0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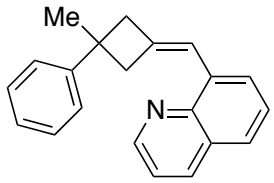
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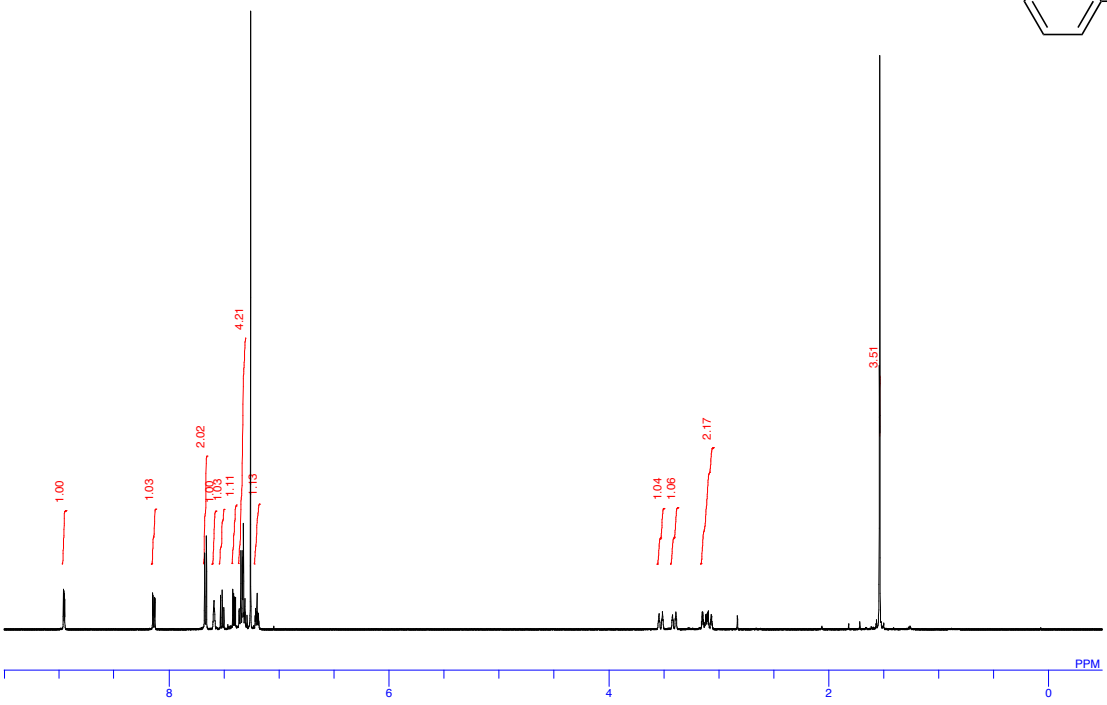
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1.04
1.06
2.17
3.51
PPM
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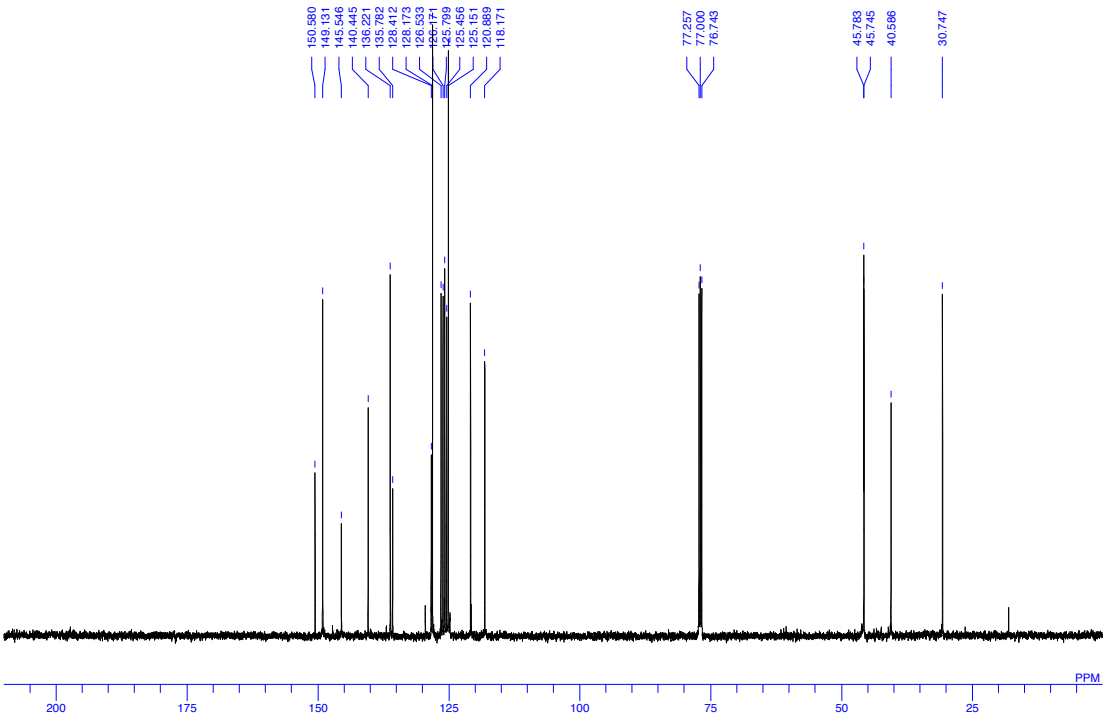
6
4
2
0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128.173
126.533
126.171
125.799
125.456
125.151
120.889
118.171
77.257
77.000
76.743
45.788
45.745
40.586
30.747
PPM
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150
125
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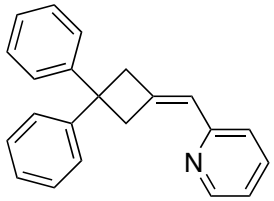
75
50
25

**1e**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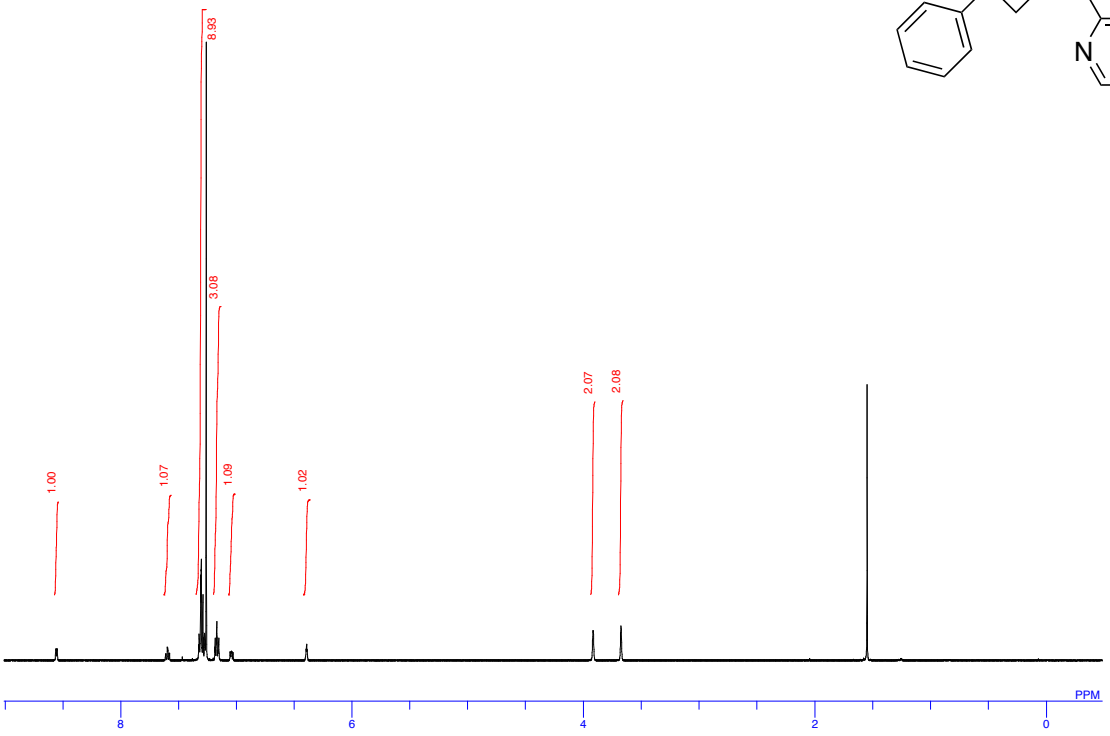
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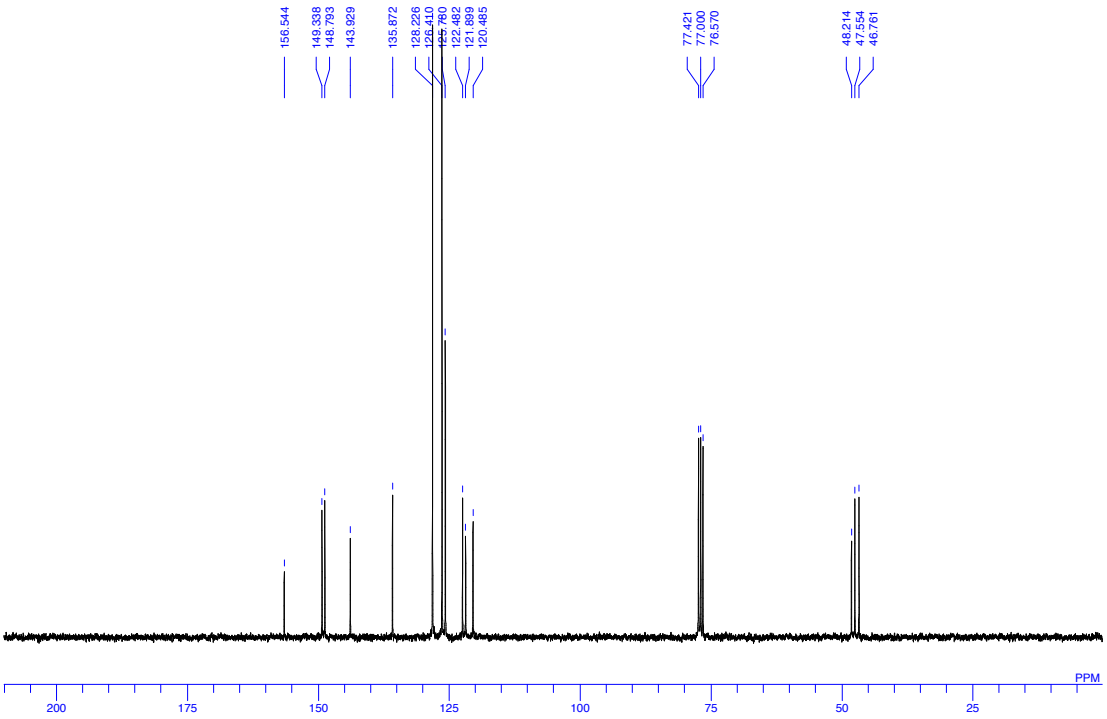
1.02
2.07
2.08
PPM
8
6
4
2
0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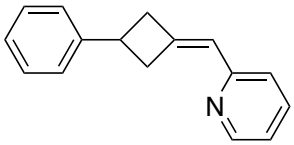
125.780
122.482
121.899
120.485
77.421
77.000
76.570
48.214
47.554
46.761
PPM
200
175
150
125
100
75
50
25

**1f**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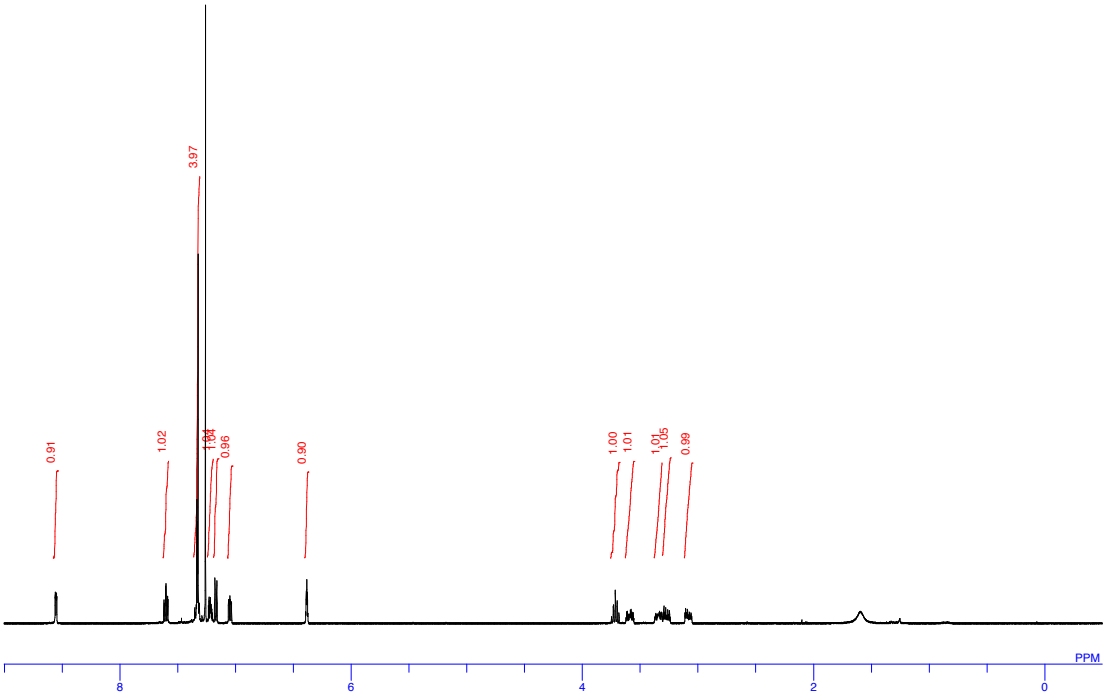
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0.90
1.00
1.01
1.01
1.05
0.99
PPM
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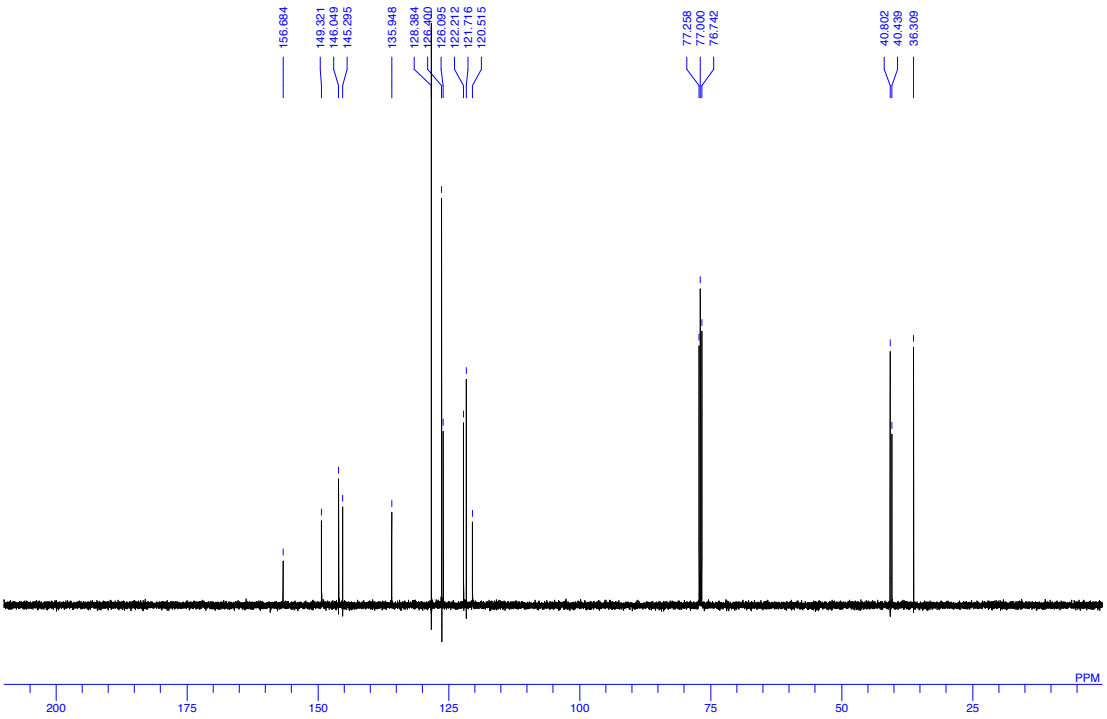
6
4
2
0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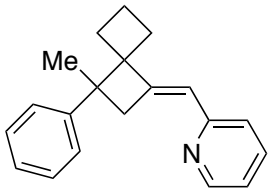
128.384
126.400
126.095
122.212
121.716
120.515
77.258
77.000
76.742
40.802
40.439
36.309
PPM
200
175
150
125
100
75
50
25

**1g**


Me
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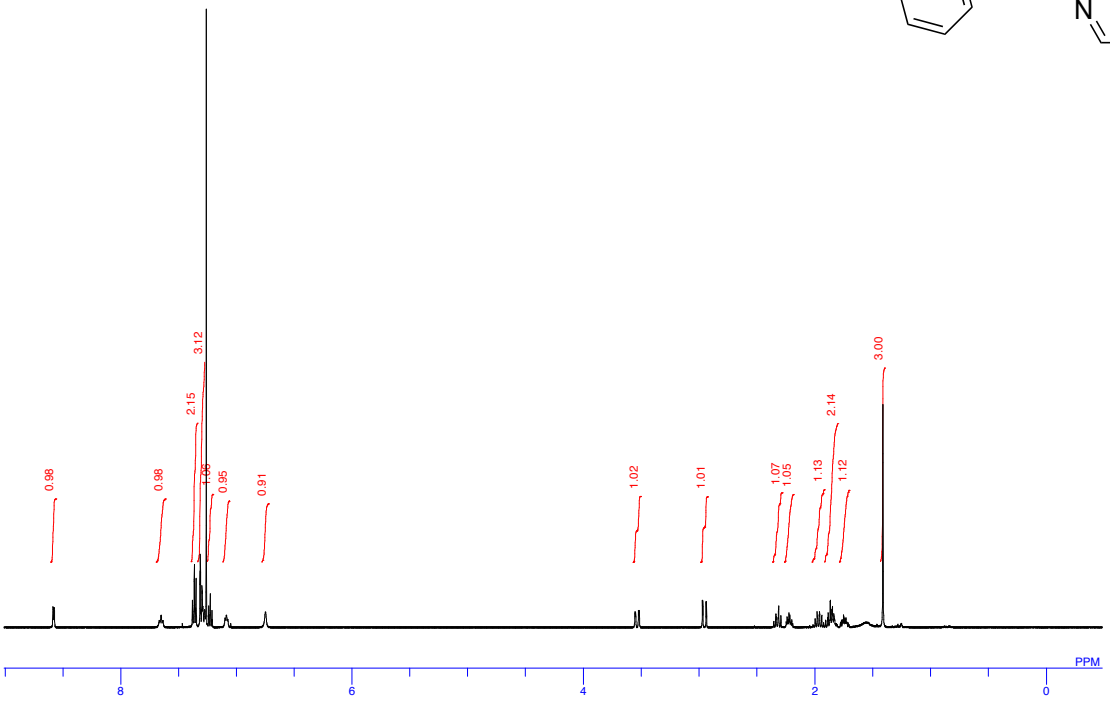
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0.98
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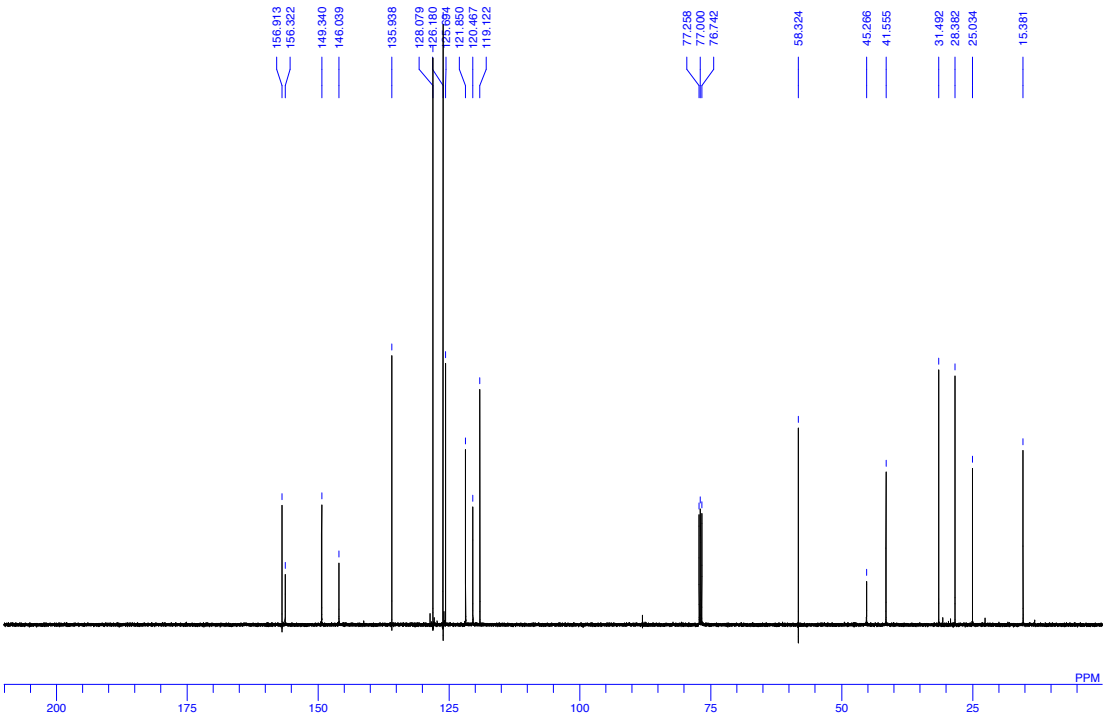
1.06
0.95
0.91
1.02
1.01
1.07
1.05
1.13
1.12
2.14
3.00
PPM
8
6
4
2
0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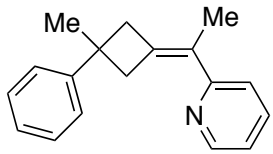
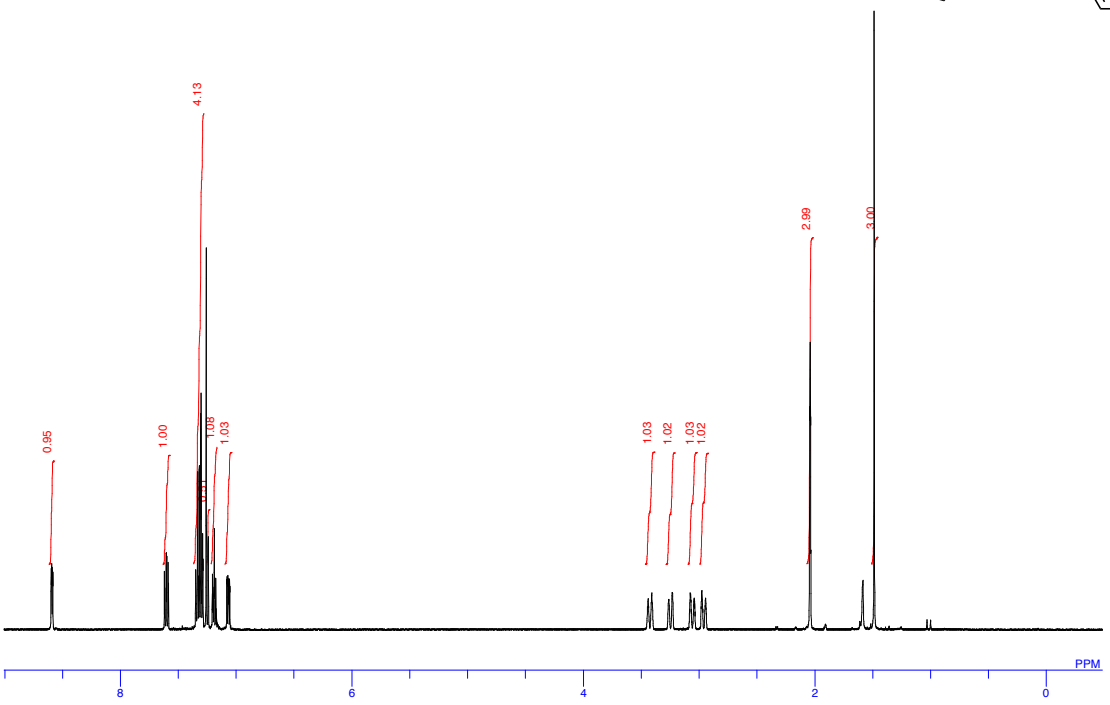
146.039
135.938
128.079
126.180
125.694
121.850
120.467
119.122
77.258
77.000
76.742
58.324
45.266
41.555
31.492
28.382
25.034
15.381
PPM
200
175
150
125
100
75
50
25

**1h**


Me
Me
N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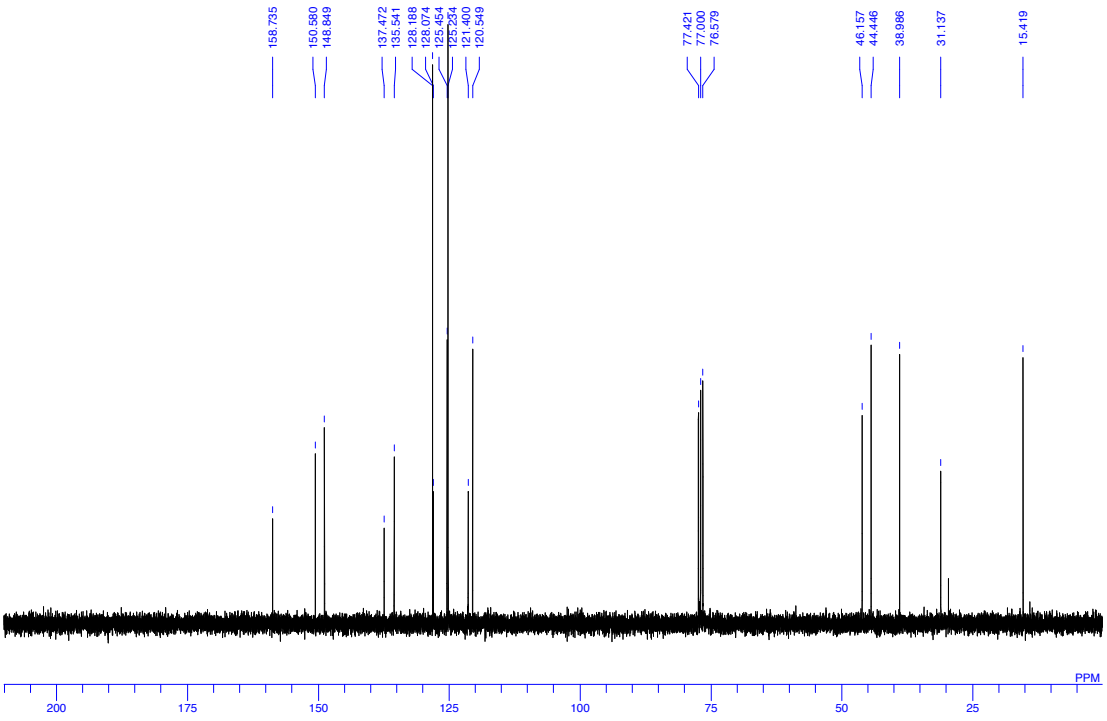
1.00
4.13
1.05
1.03
1.03
1.02
1.03
1.02
2.99
3.00
PPM
8
6
4
2
0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125.454
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121.400
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77.421
77.000
76.579
46.157
44.446
38.986
31.137
15.419
PPM
200
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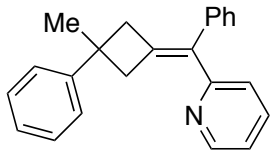
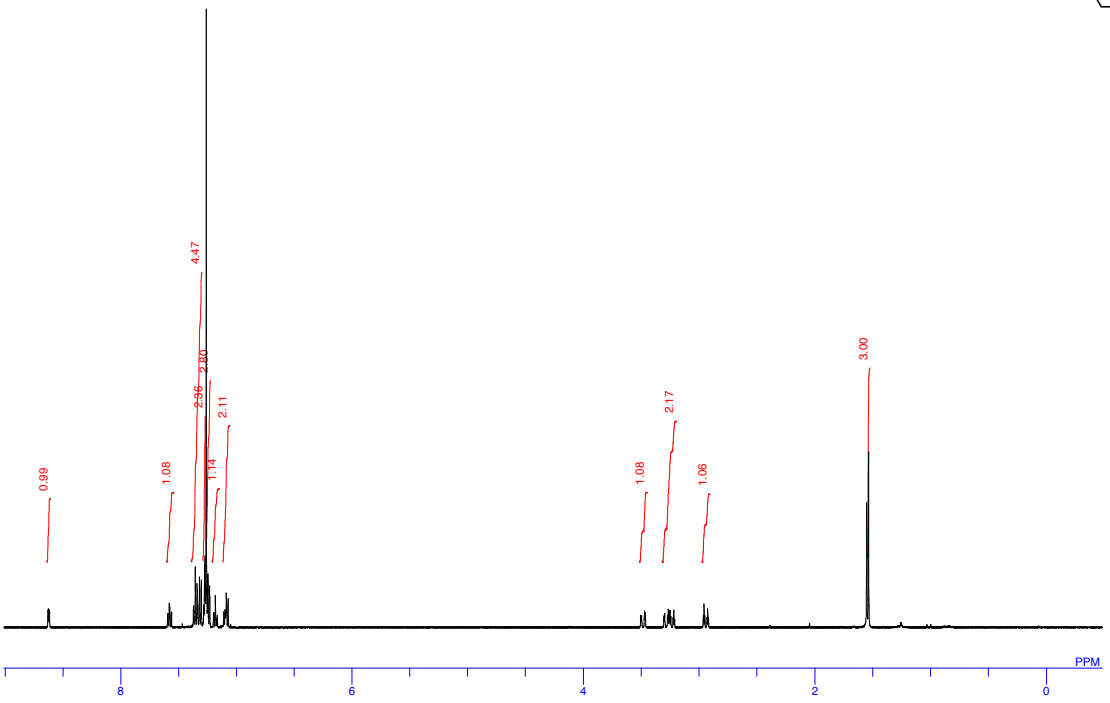
150
125
100
75
50
25

**1i**


Me
Ph
N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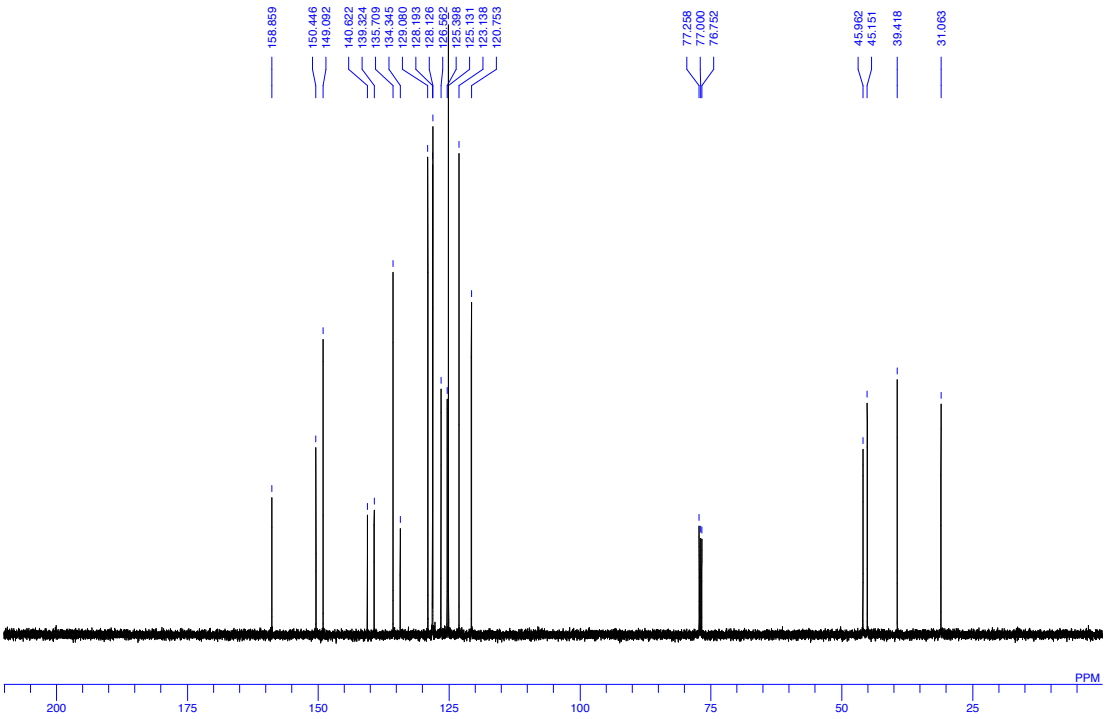
1.14
1.08
2.17
1.06
3.00
PPM
8
6
4
2
0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128.126
126.562
125.398
125.131
123.138
120.753
77.258
77.000
76.752
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45.151
39.418
31.063
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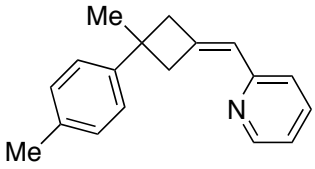
150
125
100
75
50
25

**1j**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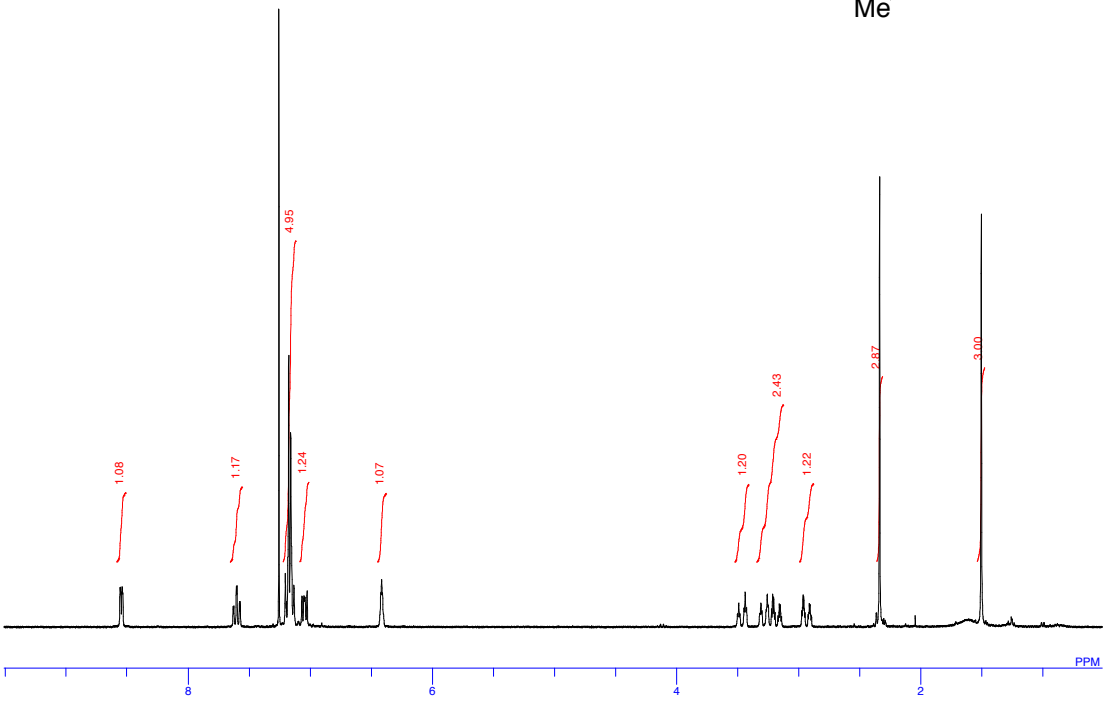
N
Me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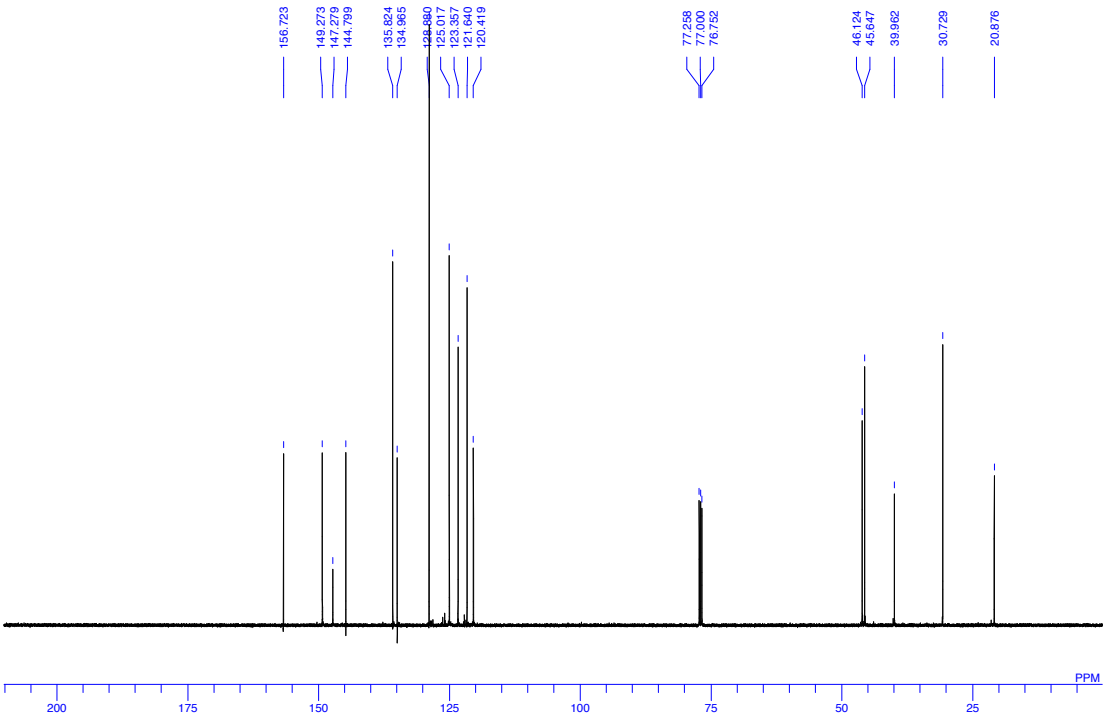
2.87
3.00
PPM
8
6
4
2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125.017
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120.419
77.258
77.000
76.752
46.124
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39.962
30.729
20.876
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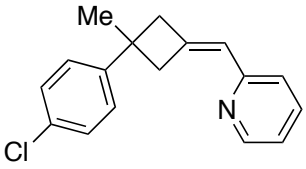
150
125
100
75
50
25

**1k**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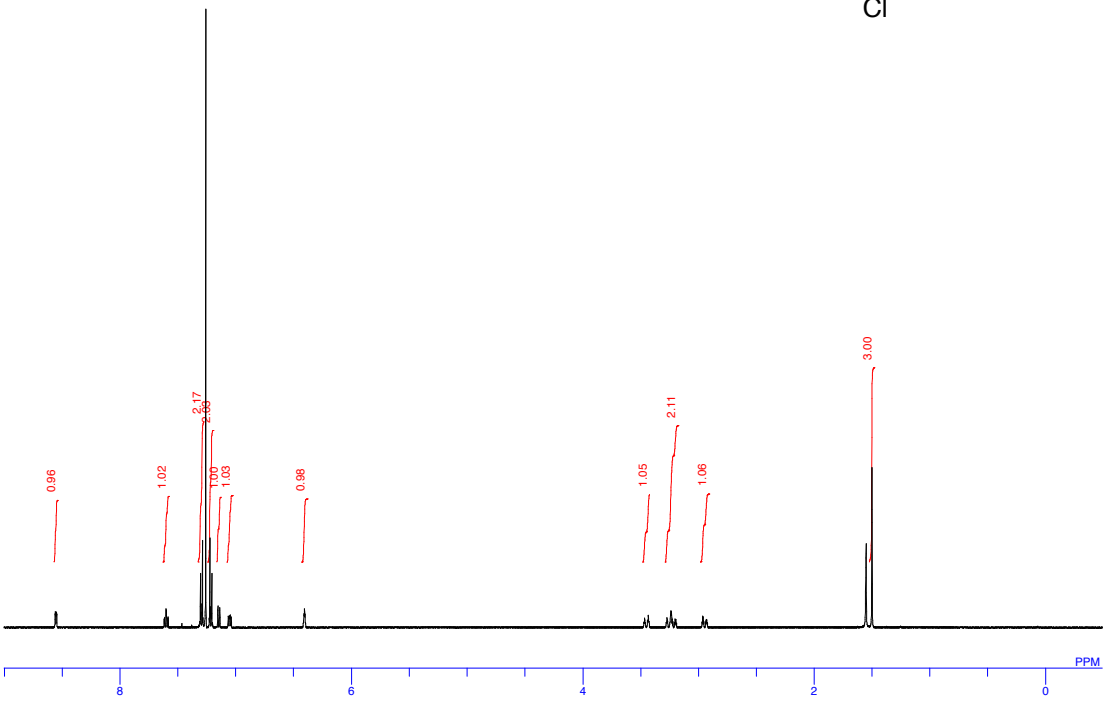
N
Cl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2.11
1.06
3.00
PPM
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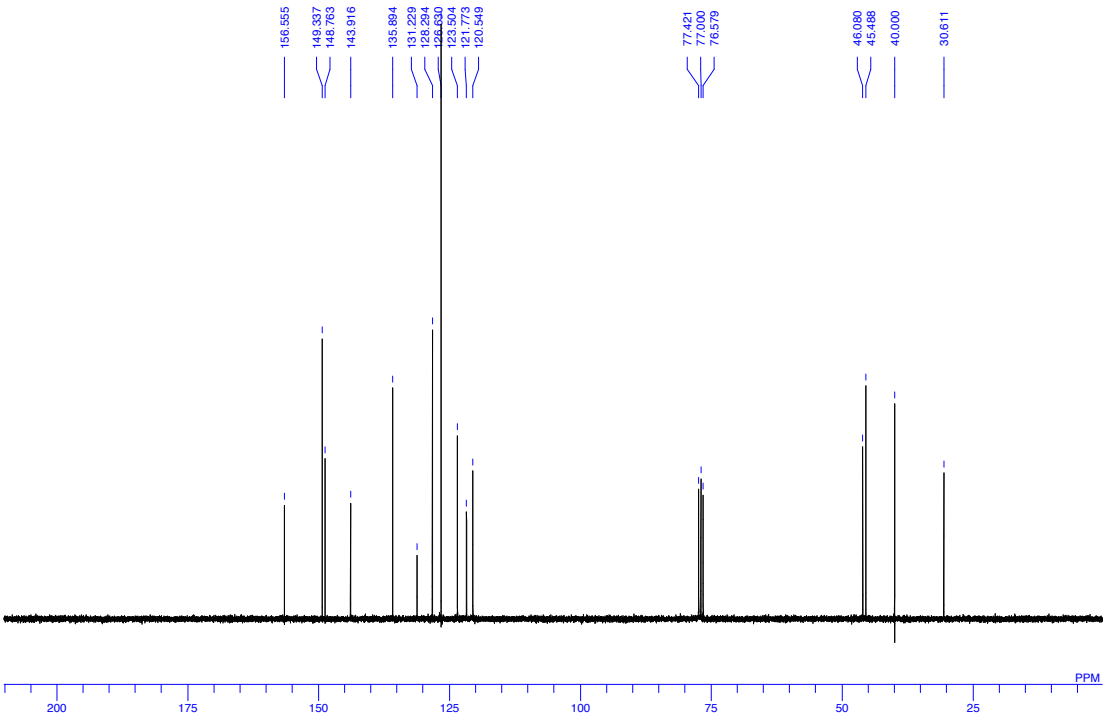
6
4
2
0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123.504
121.773
120.549
77.421
77.000
76.579
46.080
45.488
40.000
30.611
PPM
200
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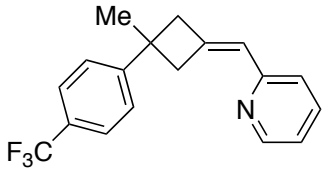
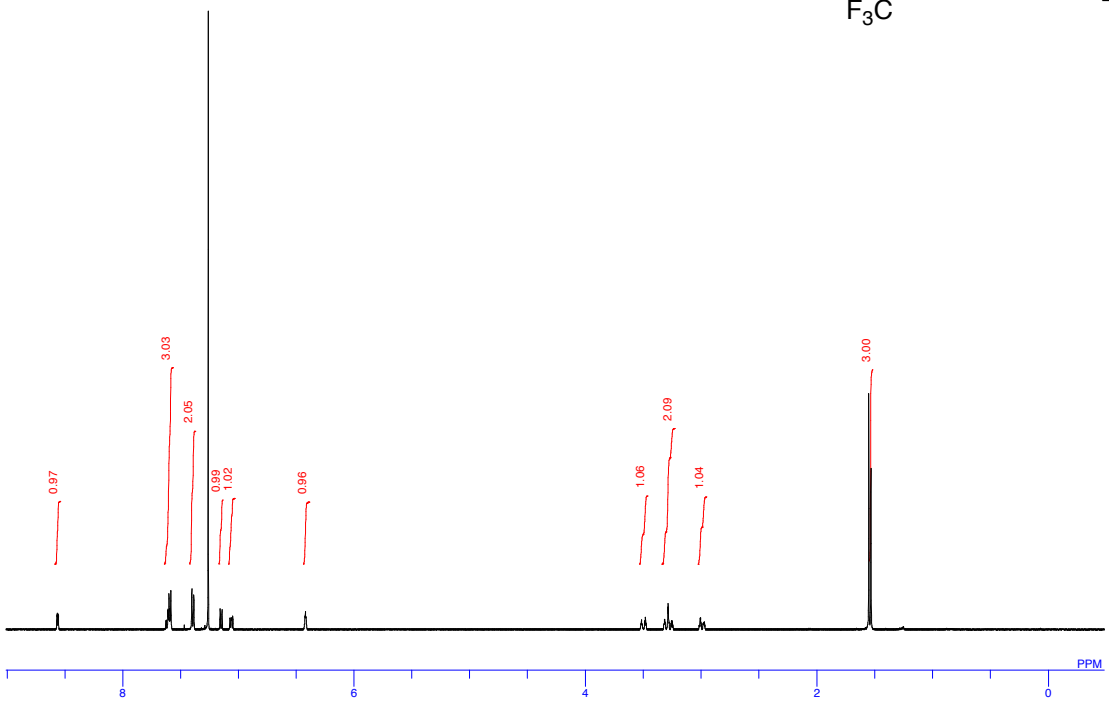
150
125
100
75
50
25

**1l**


Me
N
F3C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3.03
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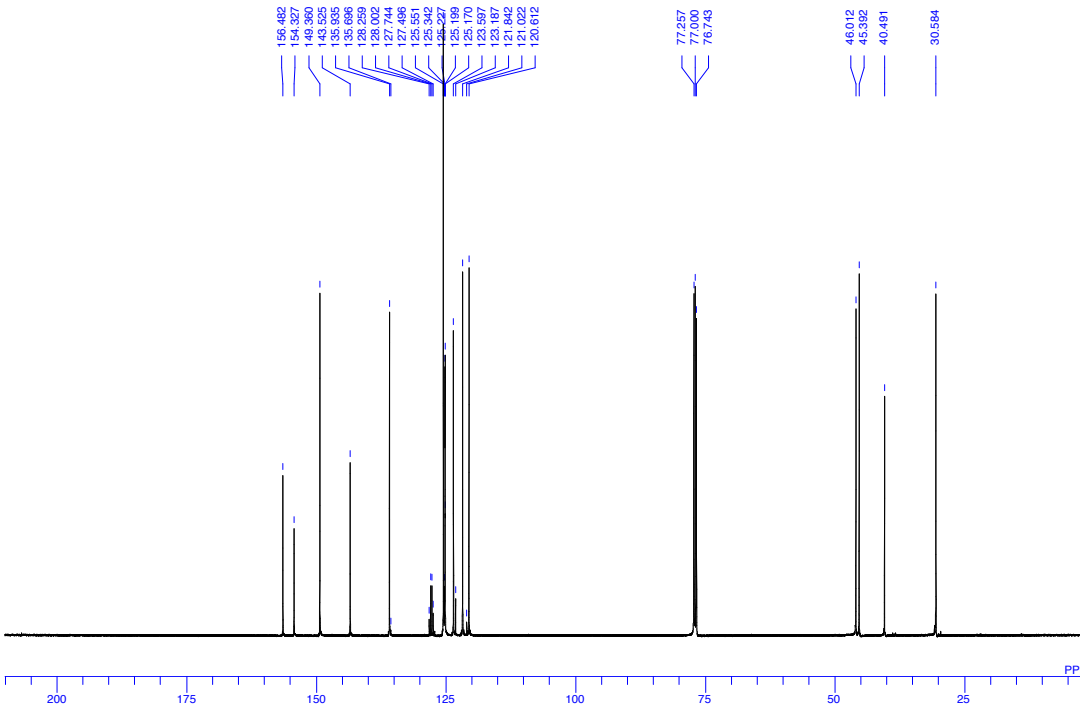
1.02
0.96
1.06
2.09
1.04
3.00
PPM
8
6
4
2
0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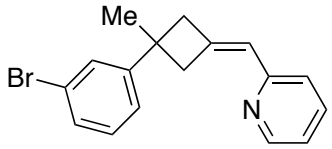
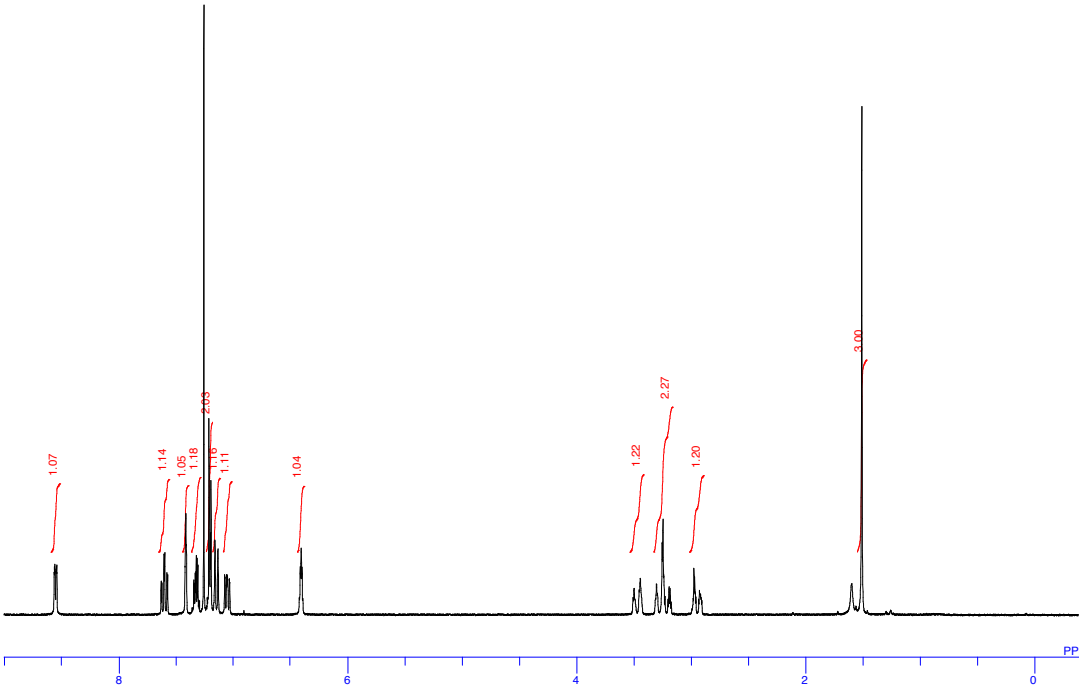
127.496
125.551
125.342
125.227
125.199
125.170
123.597
123.167
121.842
121.022
120.612
77.257
77.000
76.743
46.012
45.392
40.491
30.584
PP
200
175
150
125
100
75
50
25

**1m**


Me
Br
N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1.05
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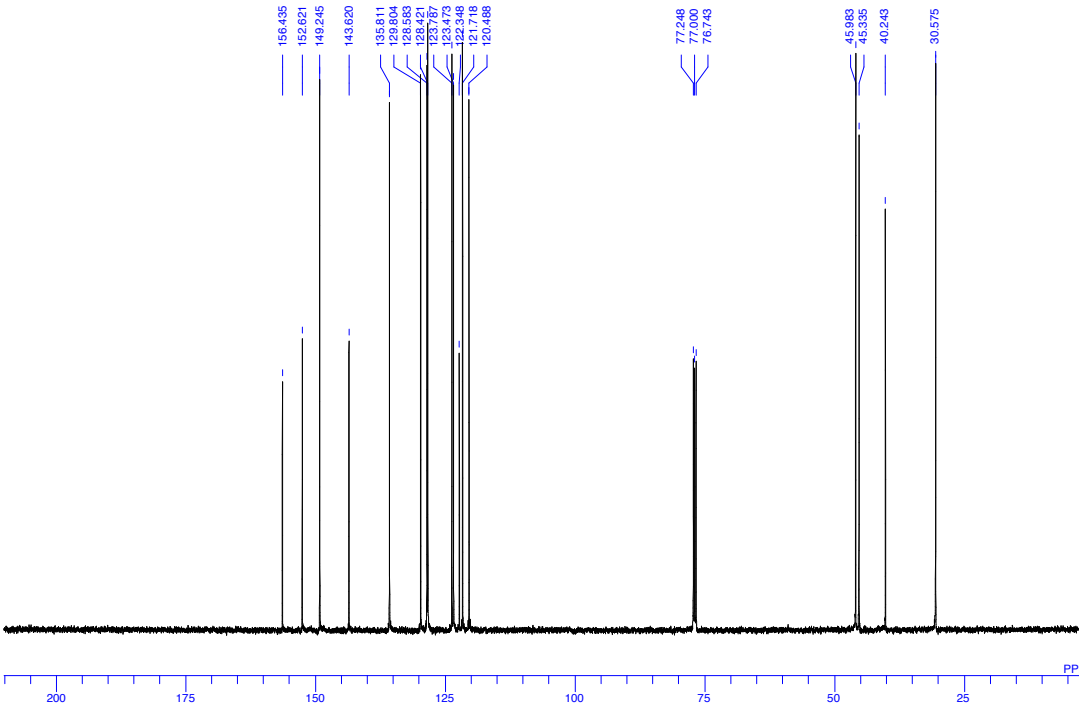
1.16
1.11
1.04
1.22
2.27
1.20
3.00
PP
8
6
4
2
0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123.473
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121.718
120.488
77.248
77.000
76.743
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45.335
40.243
30.575
PP
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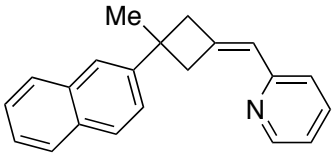
150
125
100
75
50
25

**1n**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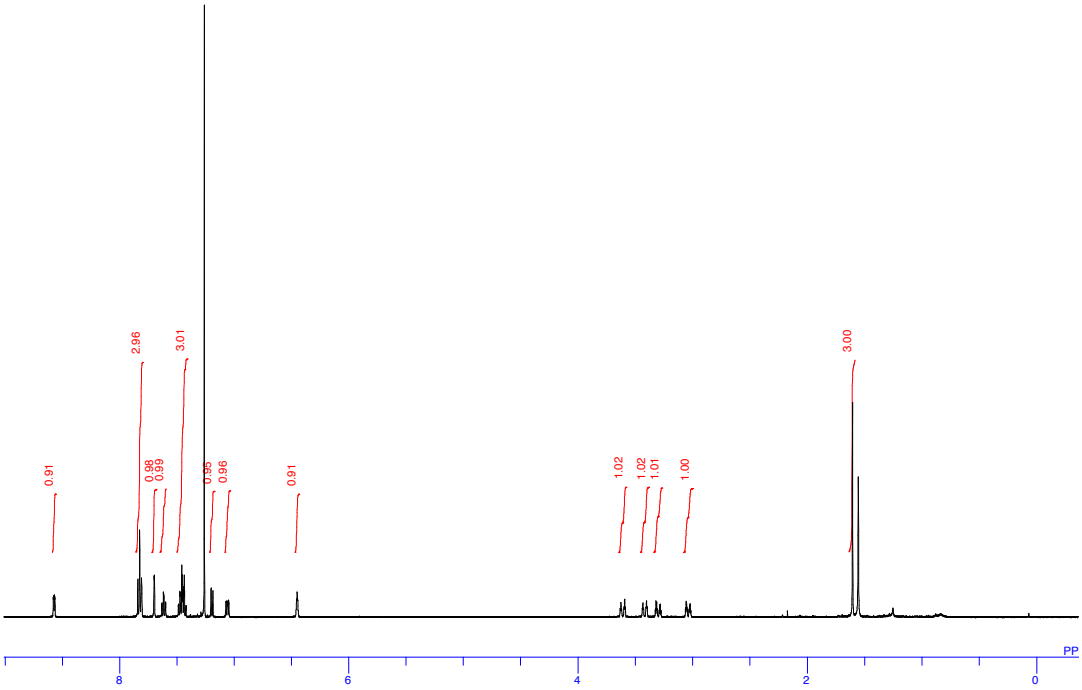
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0.91
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1.02
1.01
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3.00
PP
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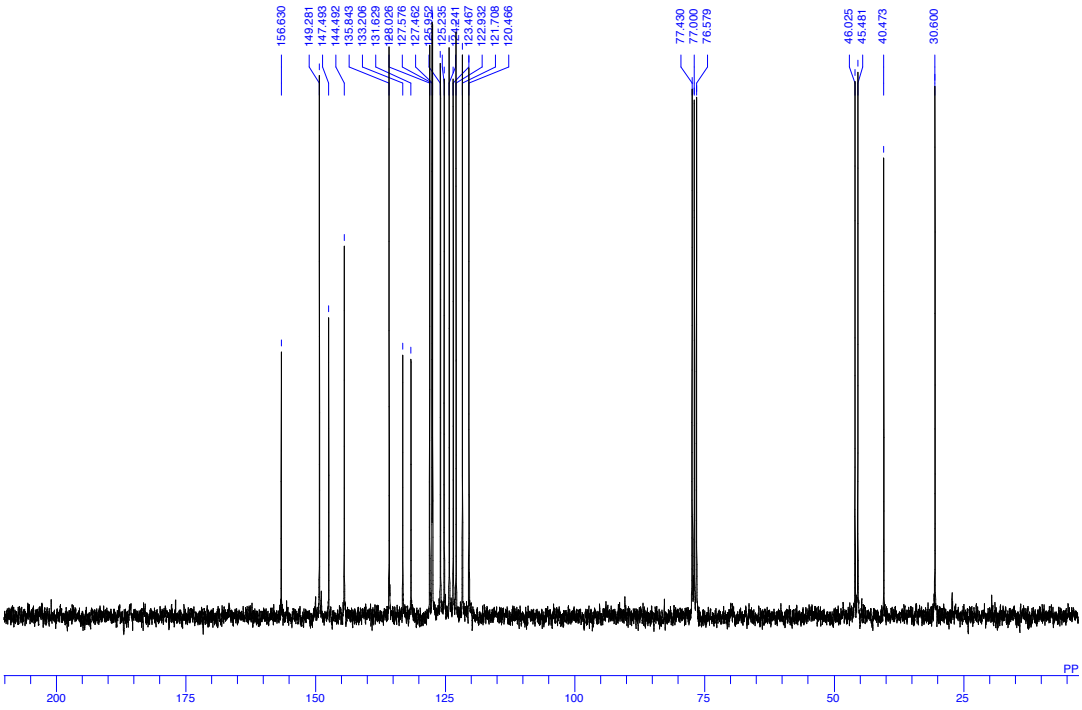
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124.241
123.467
122.932
121.708
120.466
77.430
77.000
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40.473
30.600
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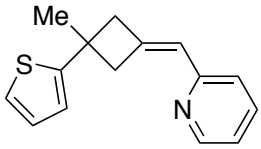
25

**1o**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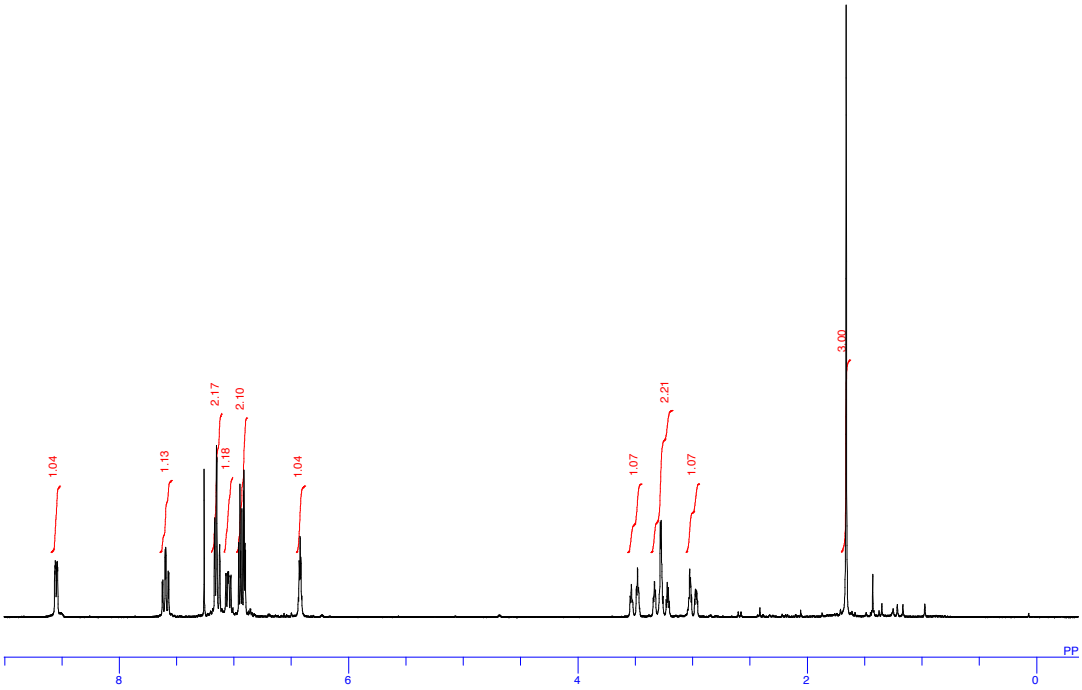
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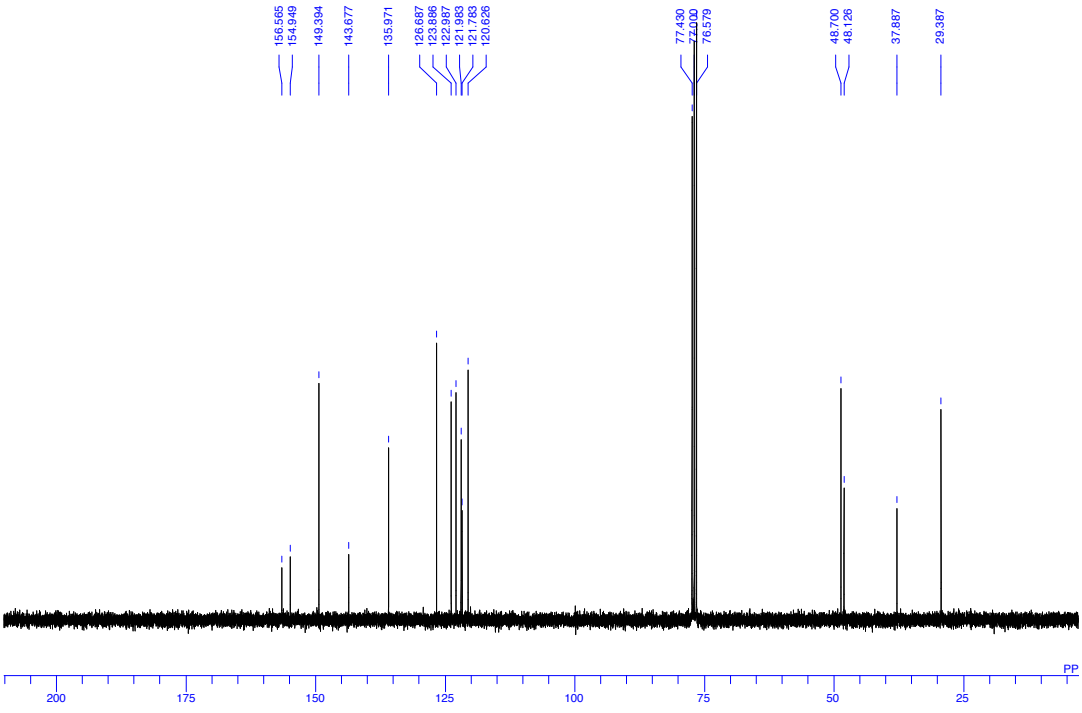
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77.000
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48.126
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29.387
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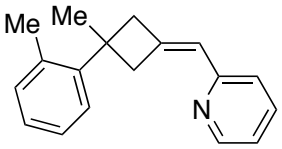
75
50
25

**1p**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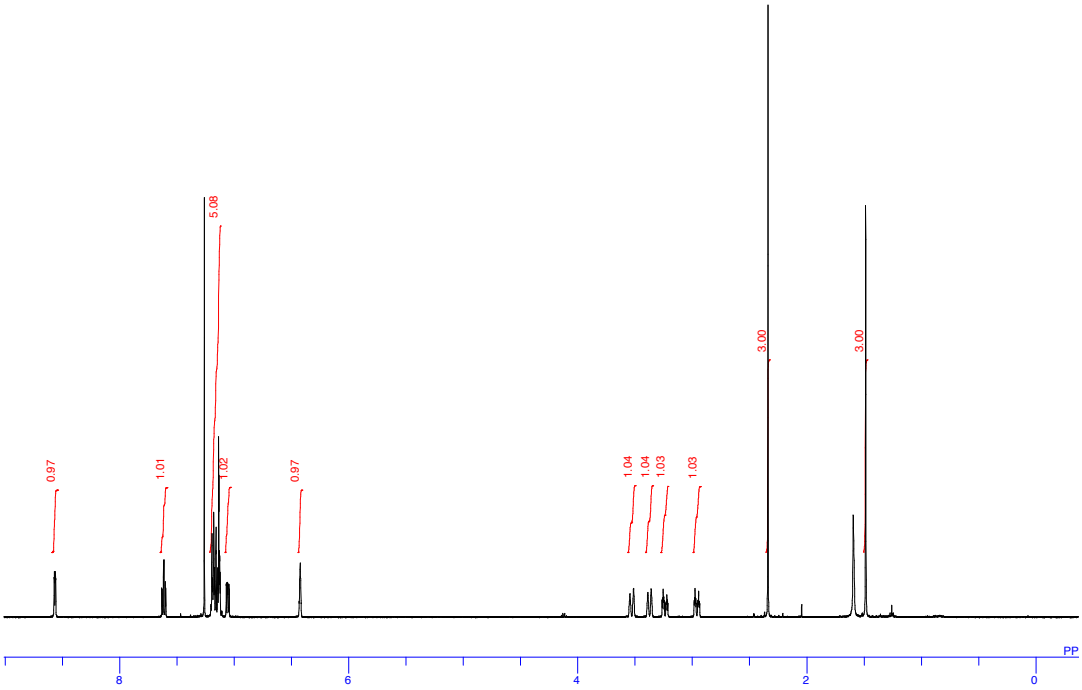
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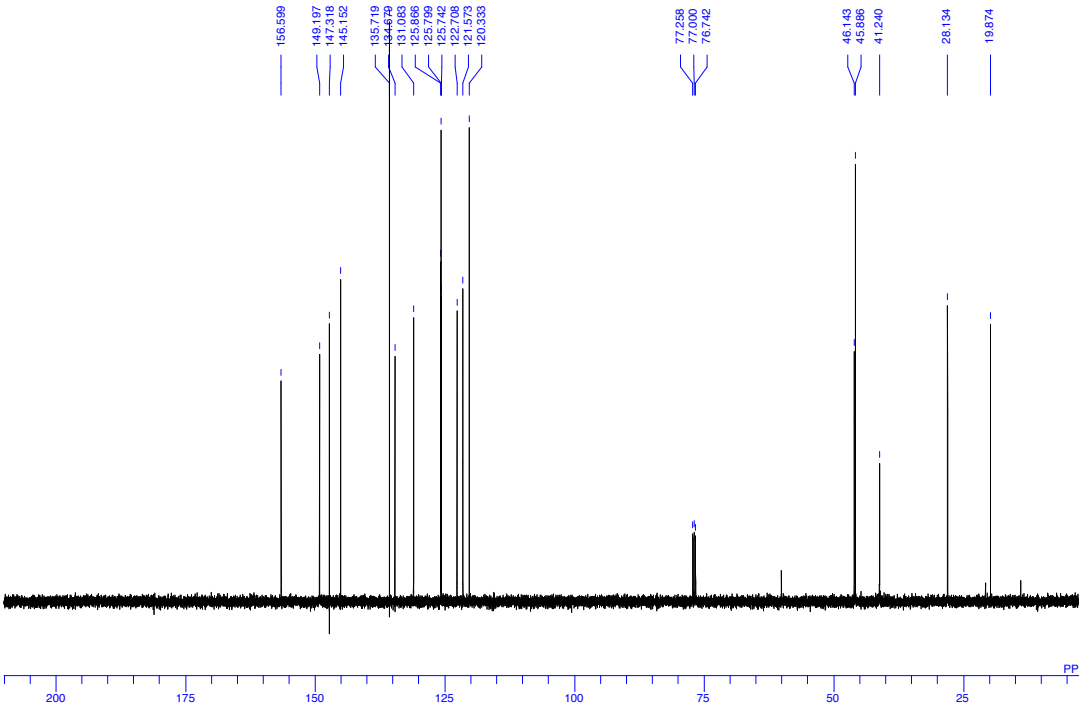
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77.000
76.742
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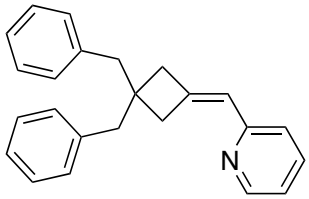
**1q**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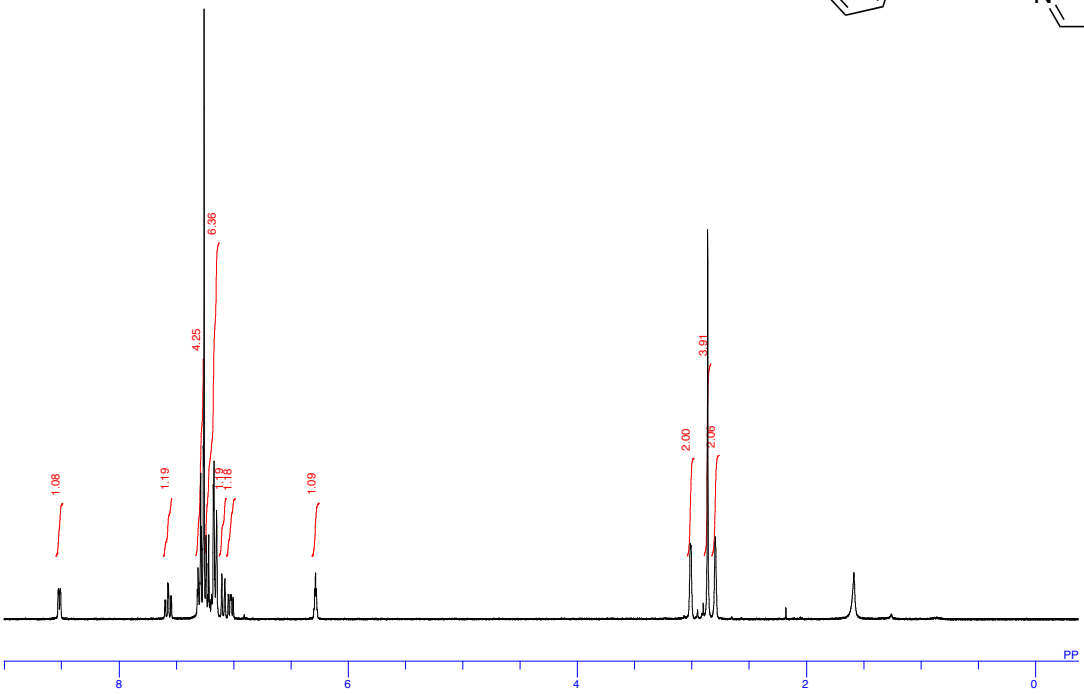
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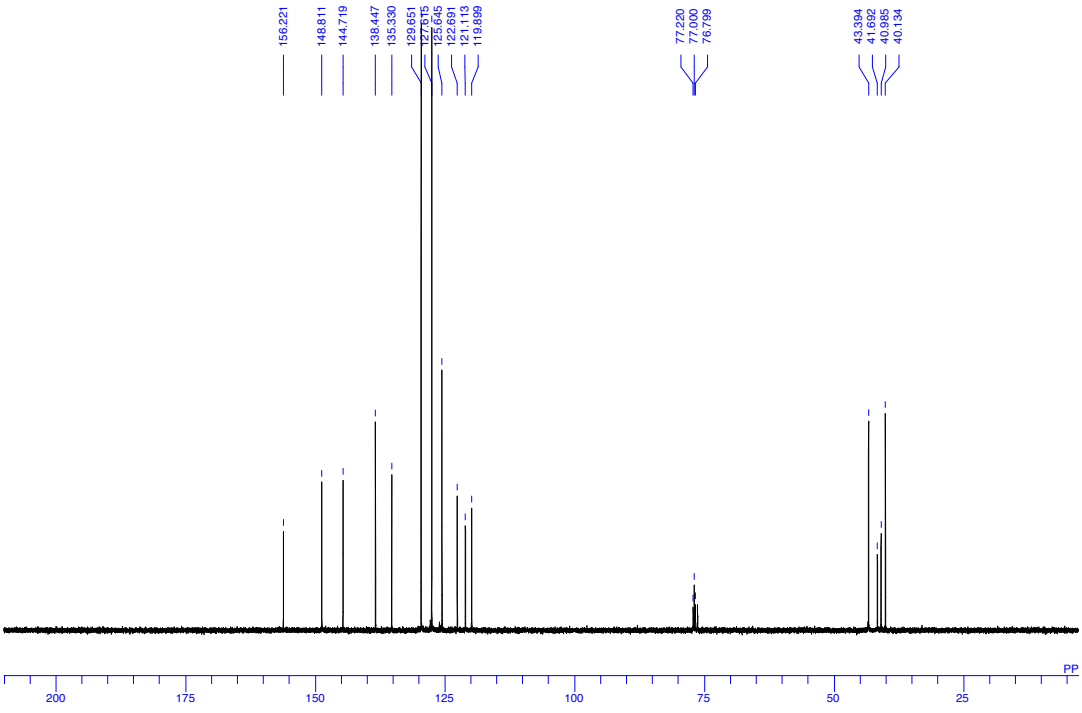
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77.220
77.000
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41.585
40.134
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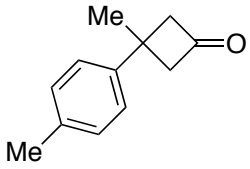
150
125
100
75
50
25

**6b**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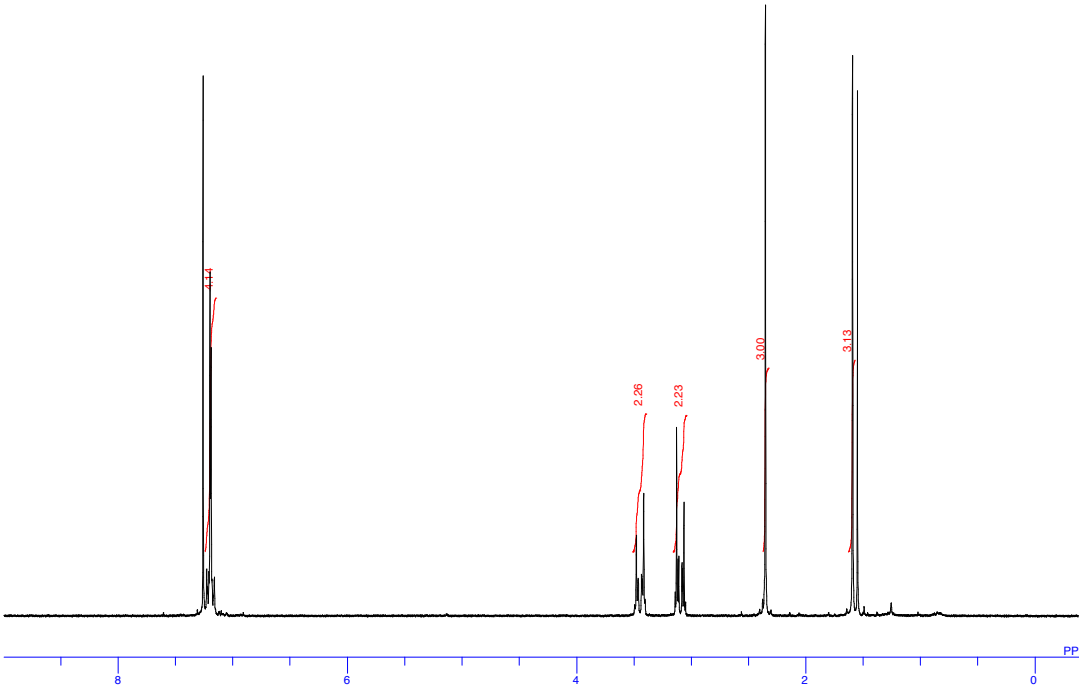
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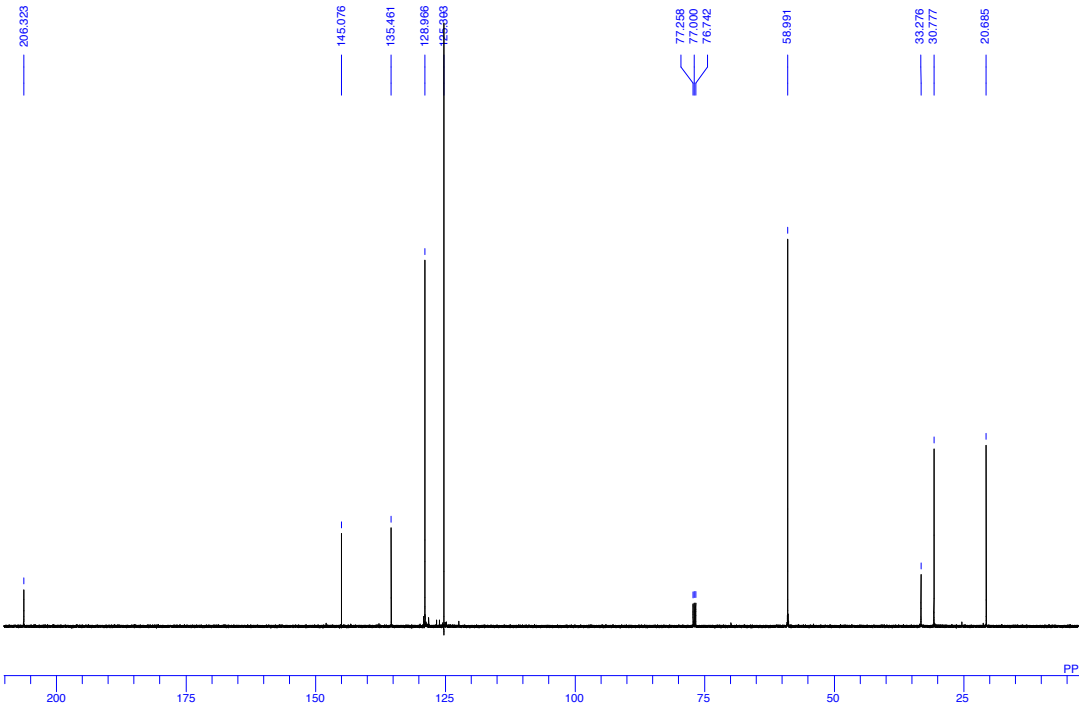
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125
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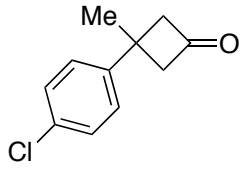
75
50
25

**6c**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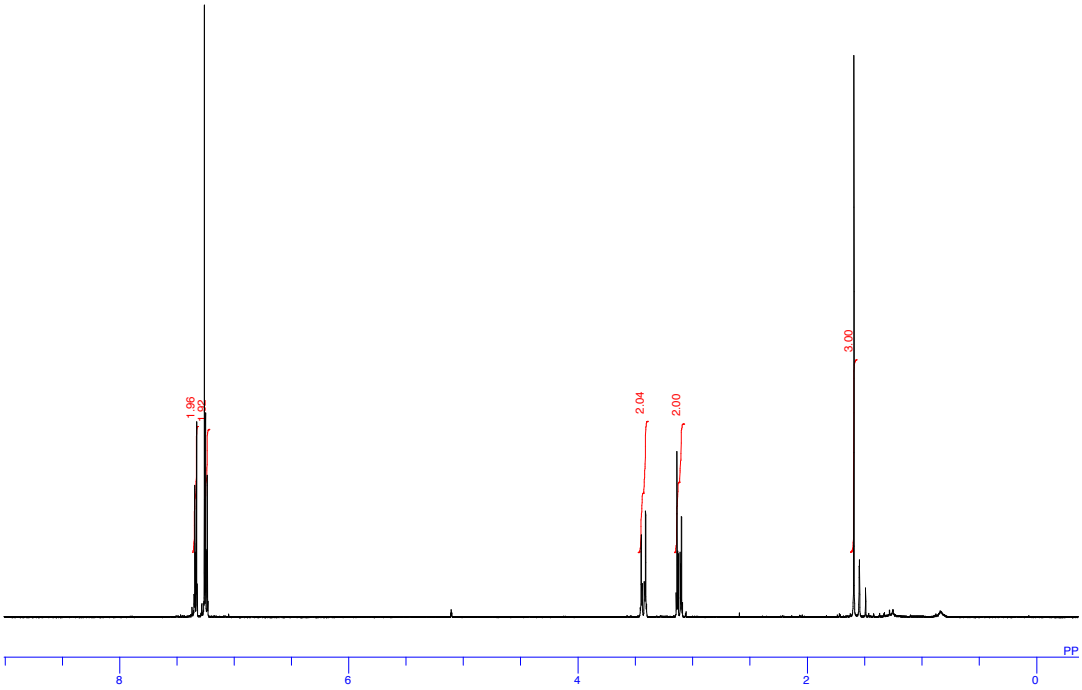
Cl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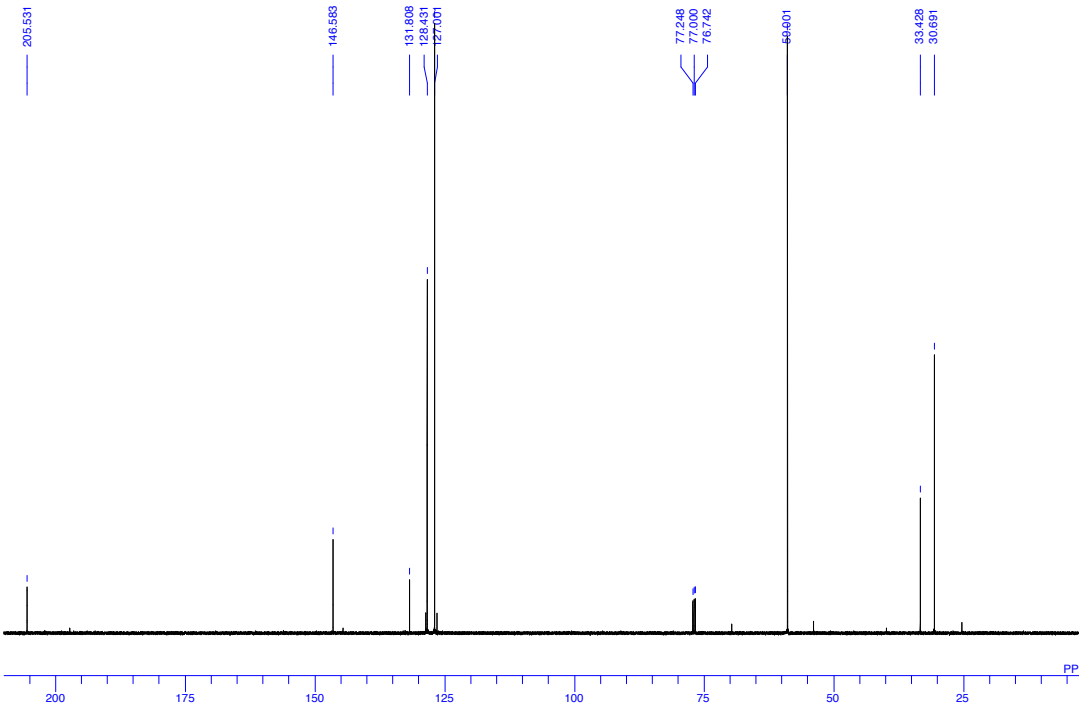
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125
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75
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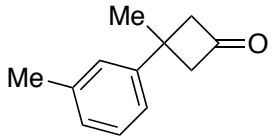
**6d**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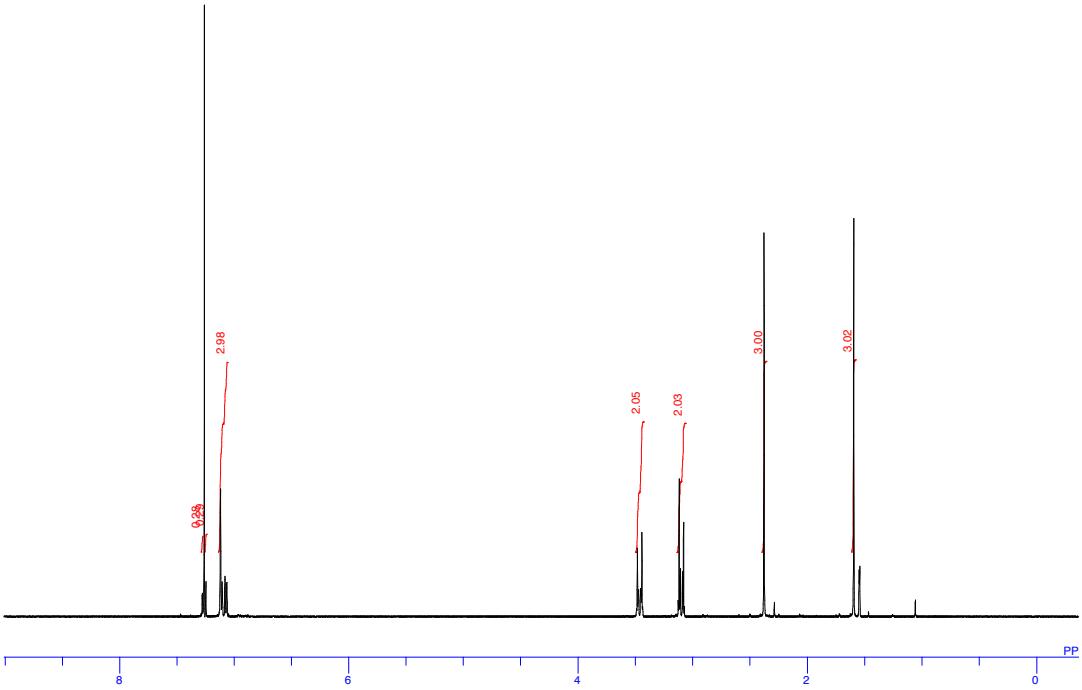
Me
O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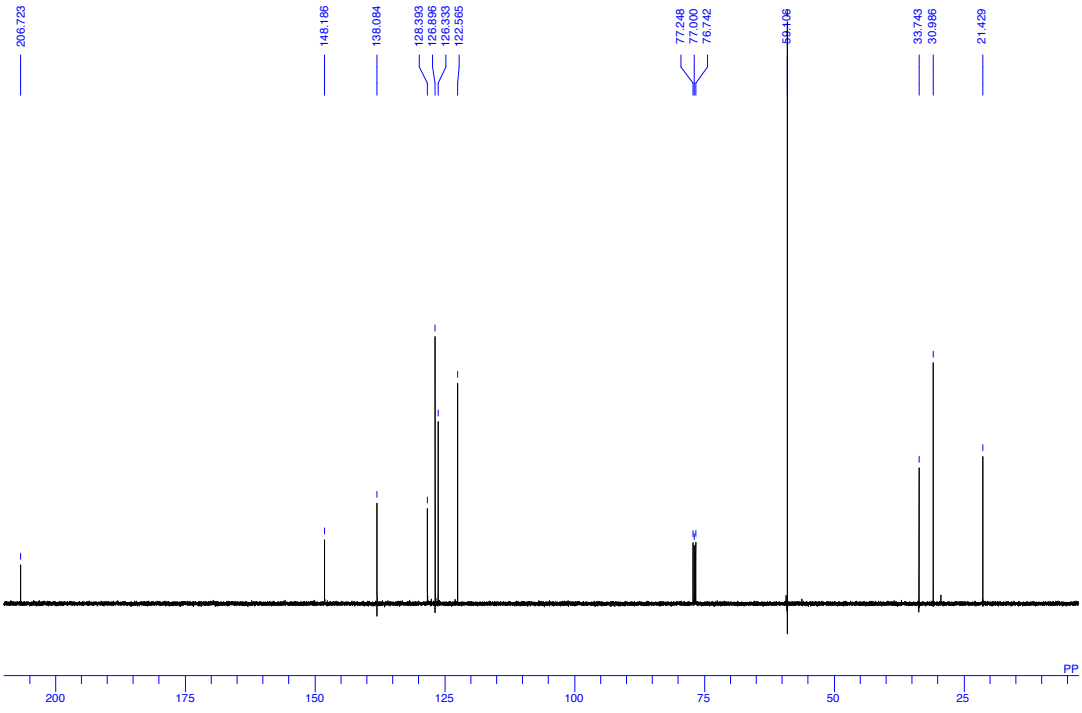
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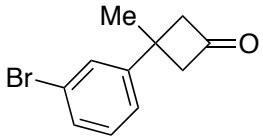
50
25

**6e**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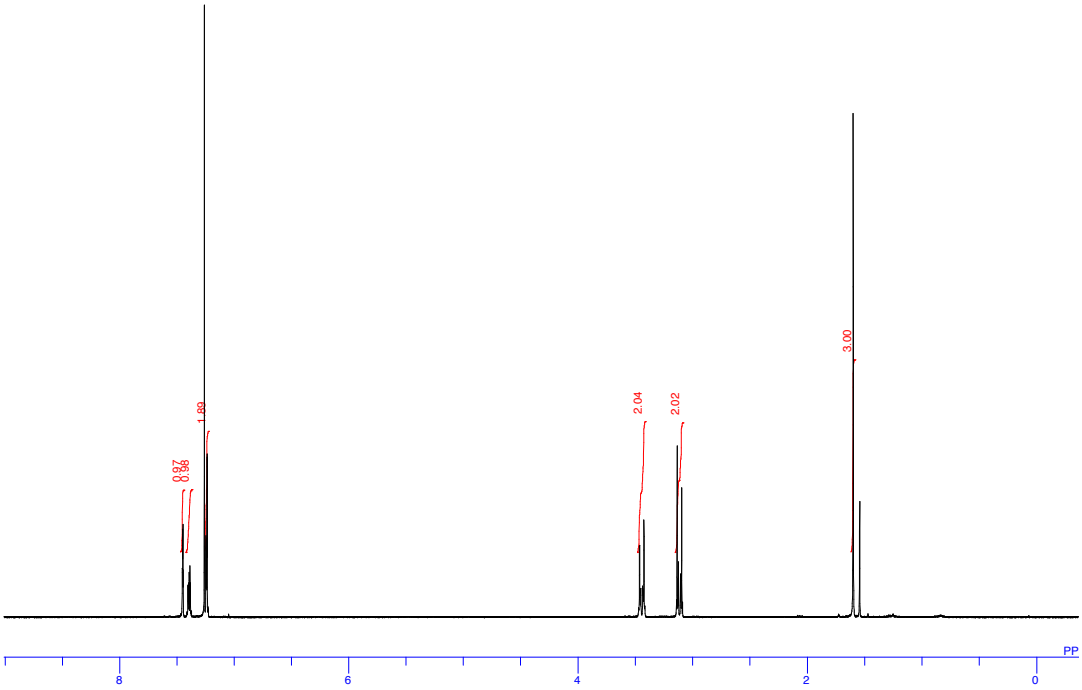
O
Br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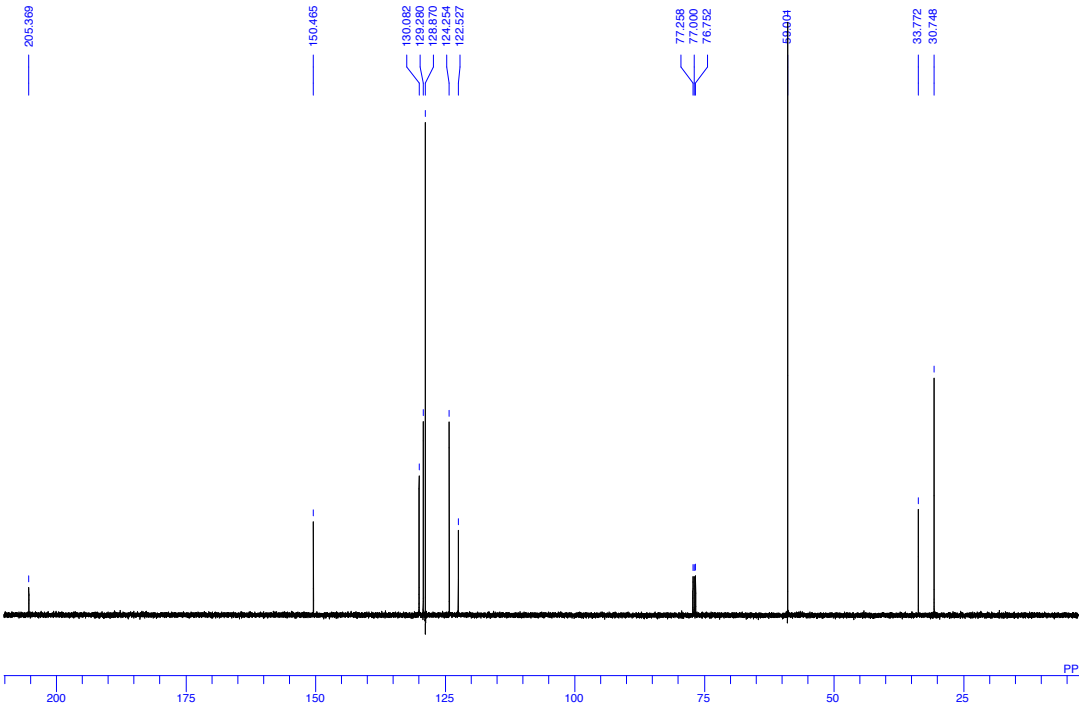
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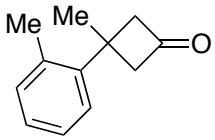
50
25

**6f**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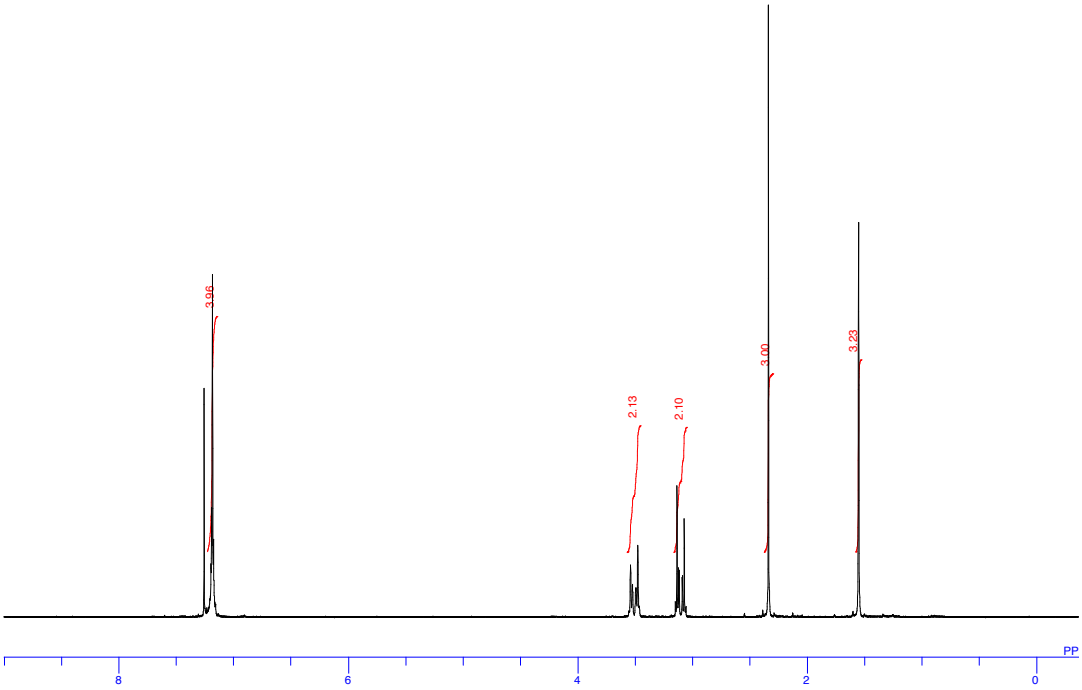
Me
O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6
4
2
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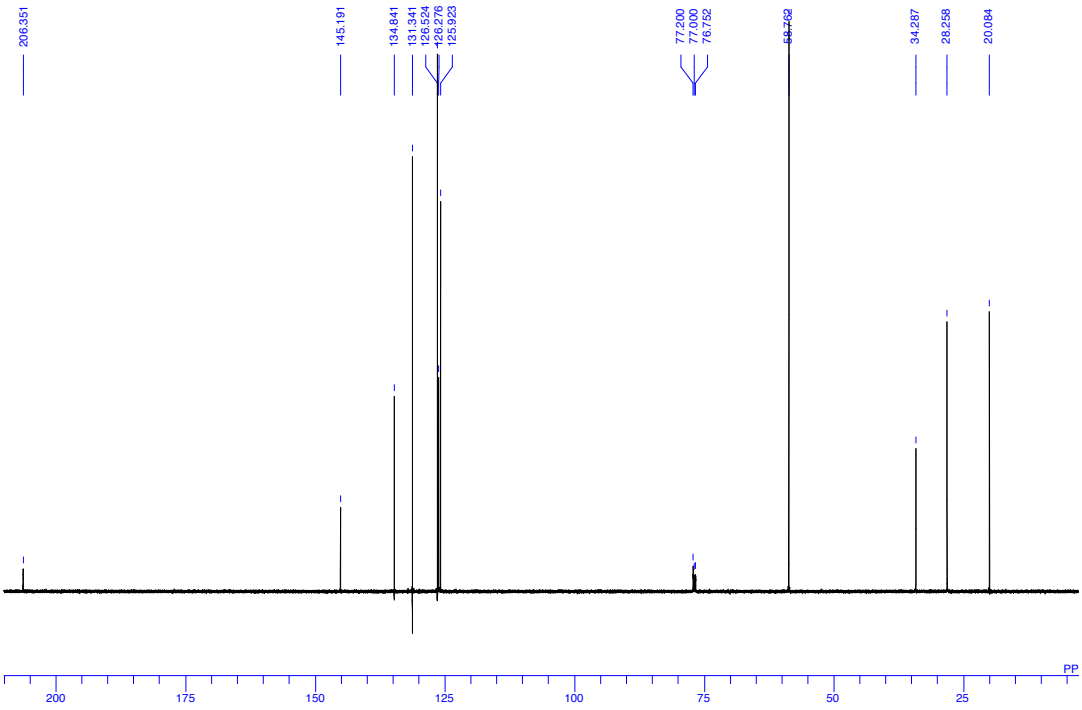
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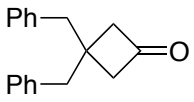
**6h**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Ph
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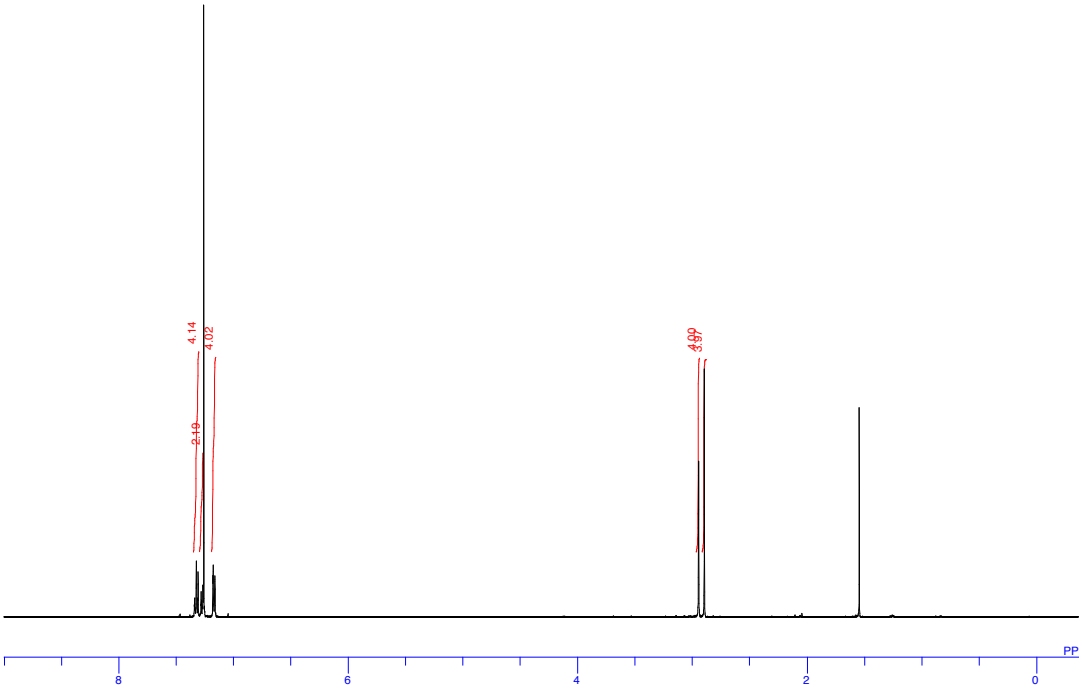
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6
4
2
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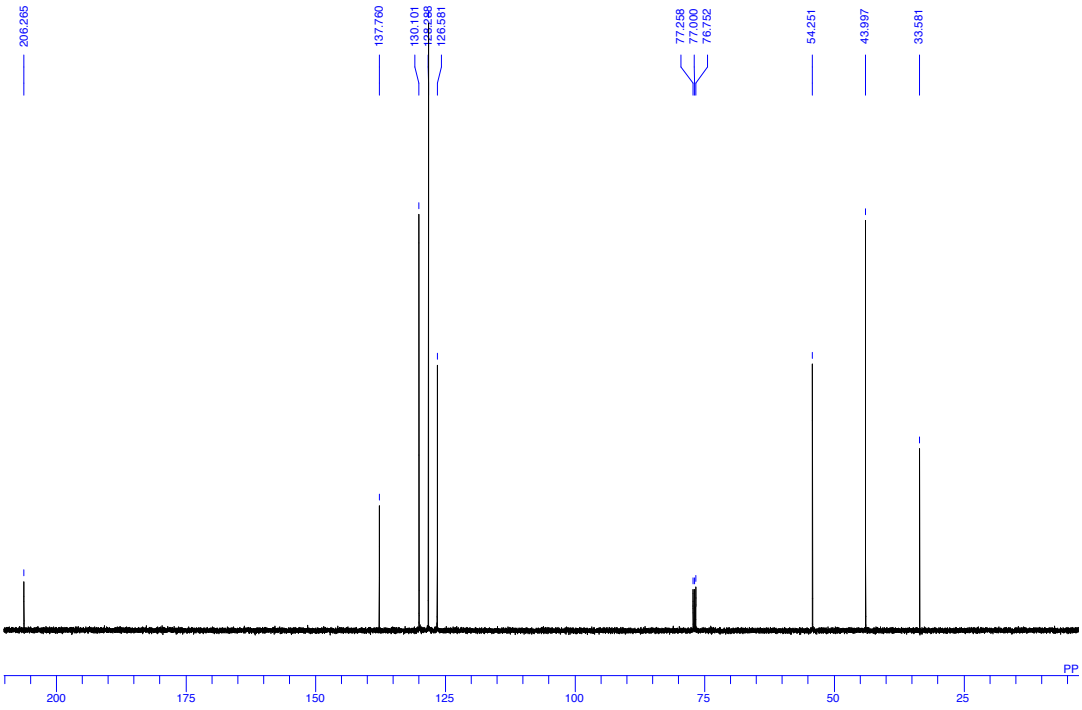
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150
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100
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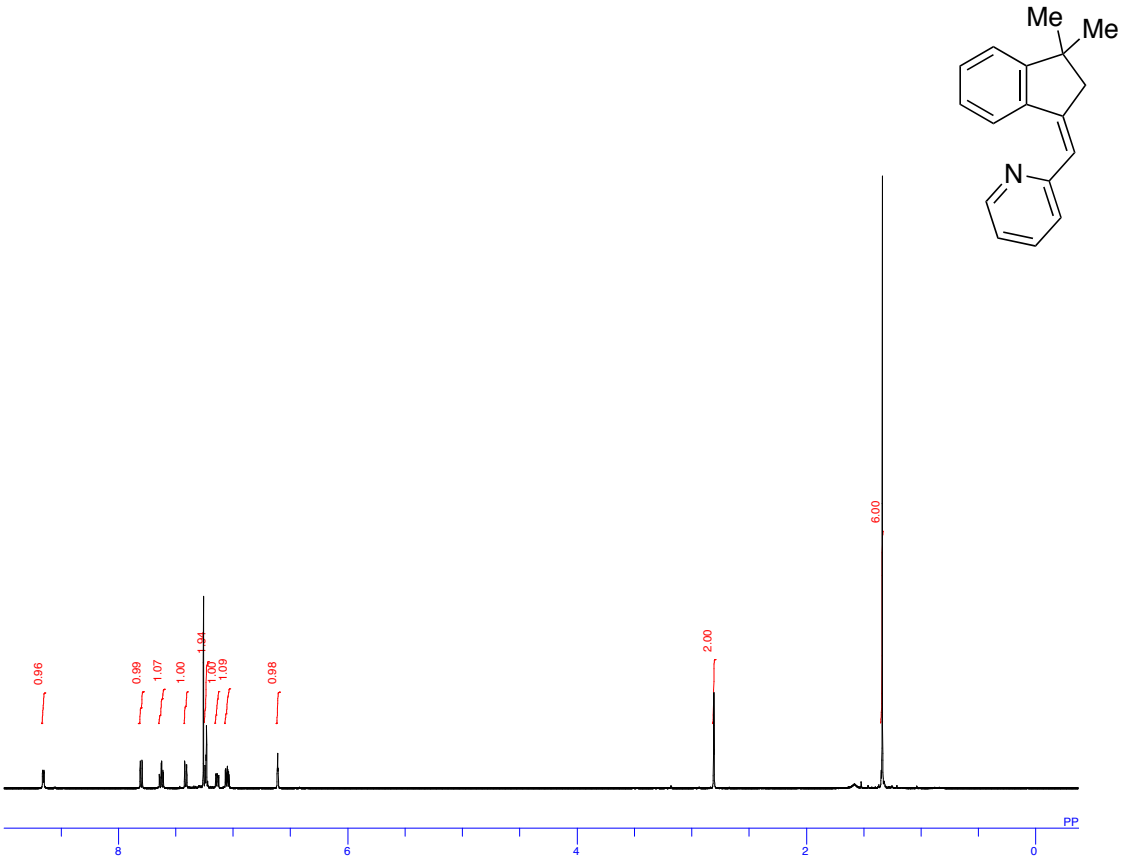
50
25

**2a**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Me
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1.09
0.98
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6.00
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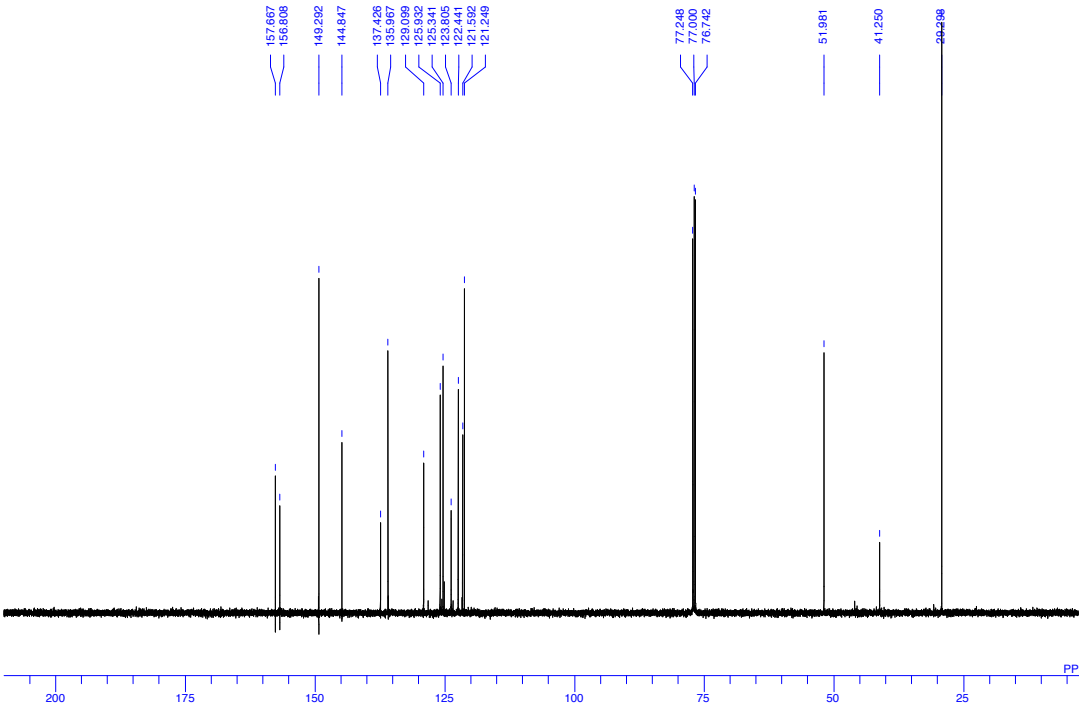
6
4
2
0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77.248
77.000
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41.250
29.398
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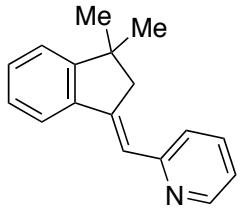
150
125
100
75
50
25

**3a**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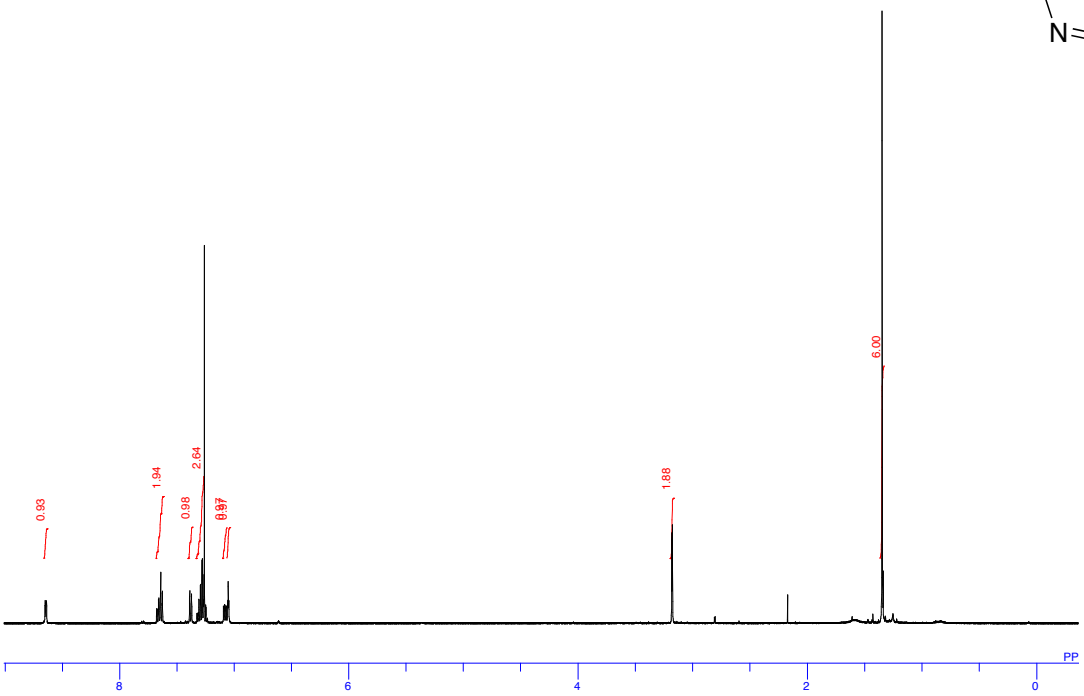
Me
N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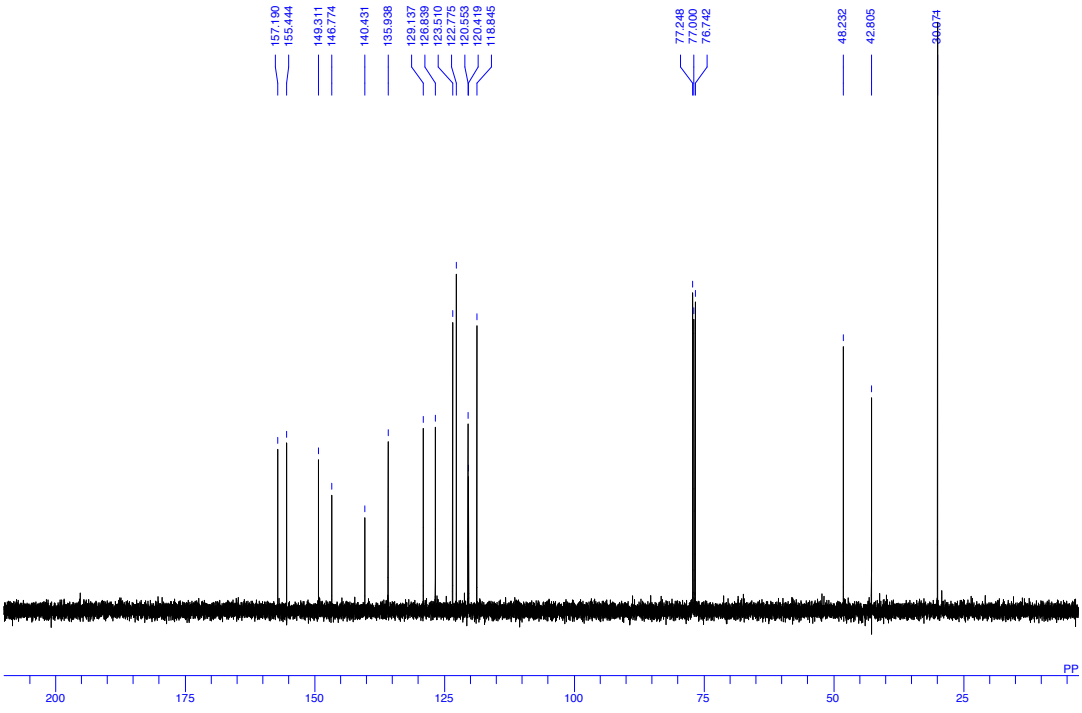
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118.845
77.248
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42.805
30.971
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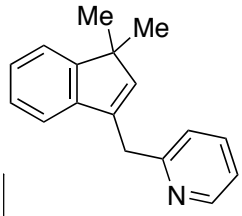
150
125
100
75
50
25

**4a**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Me
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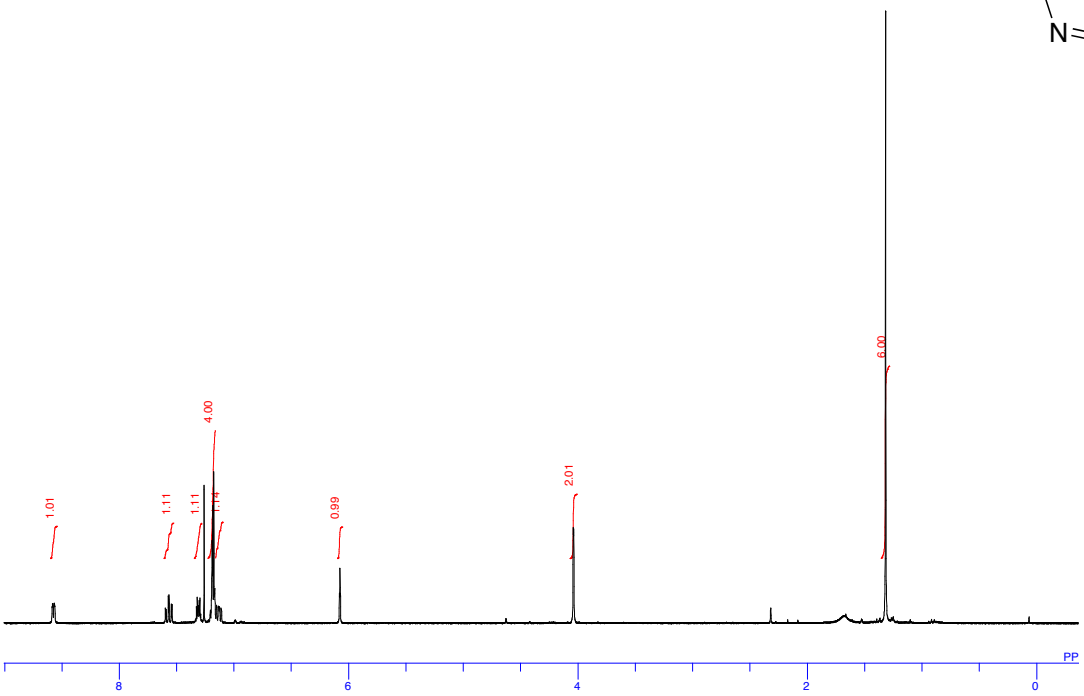
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6.00
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6
4
2
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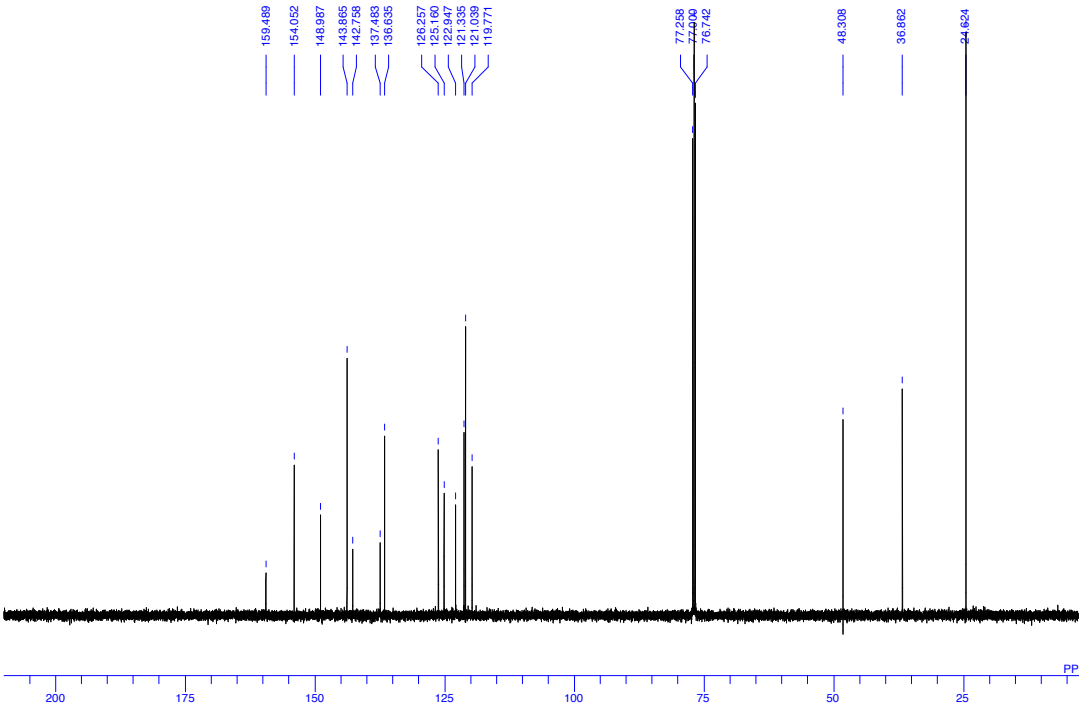
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77.258
77.000
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36.862
24.624
PP
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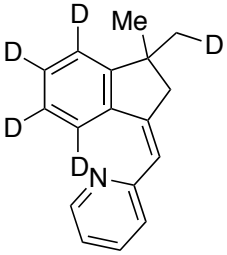
175
150
125
100
75
50
25

**2a**-$d_5$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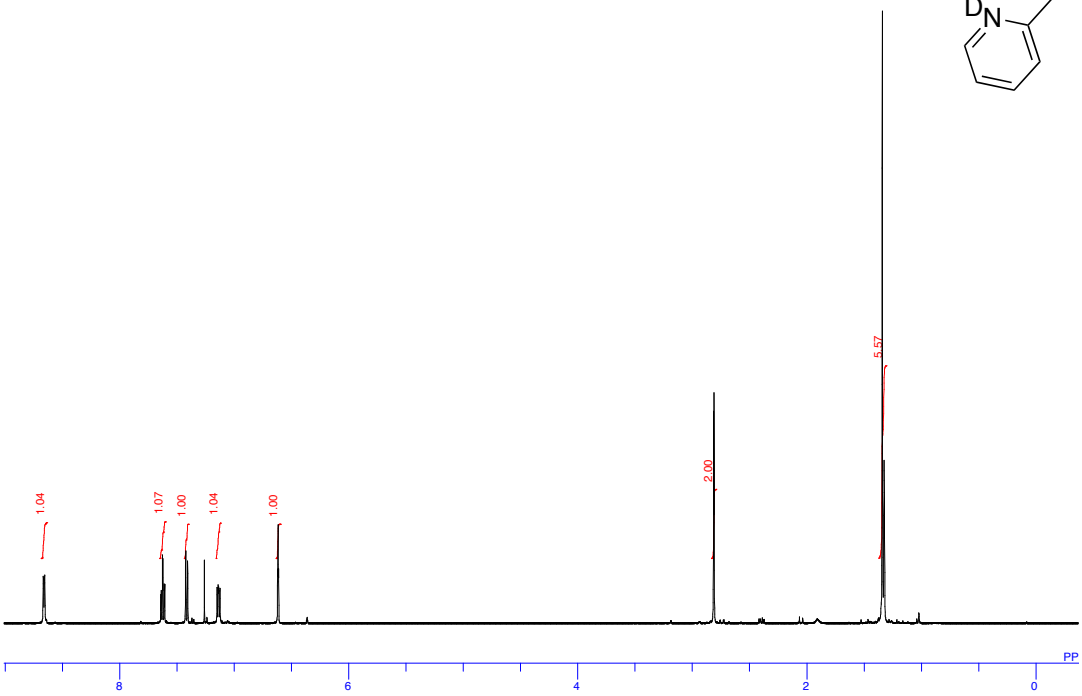
D
D
D
D
N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1.00
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2
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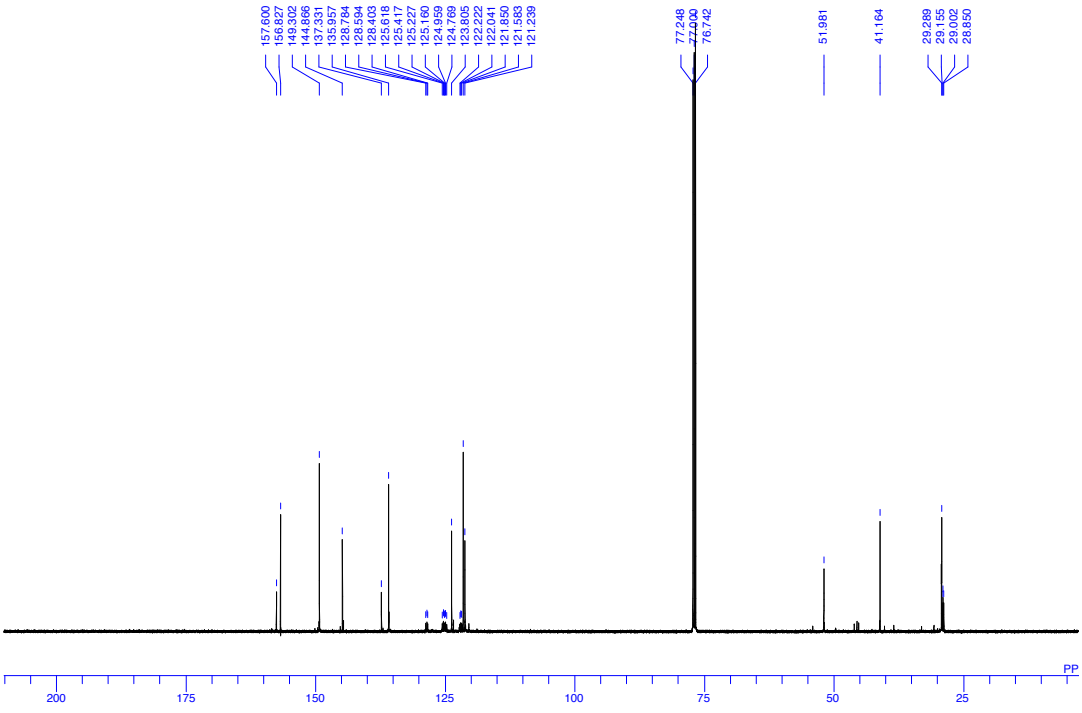
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77.000
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51.981
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150
125
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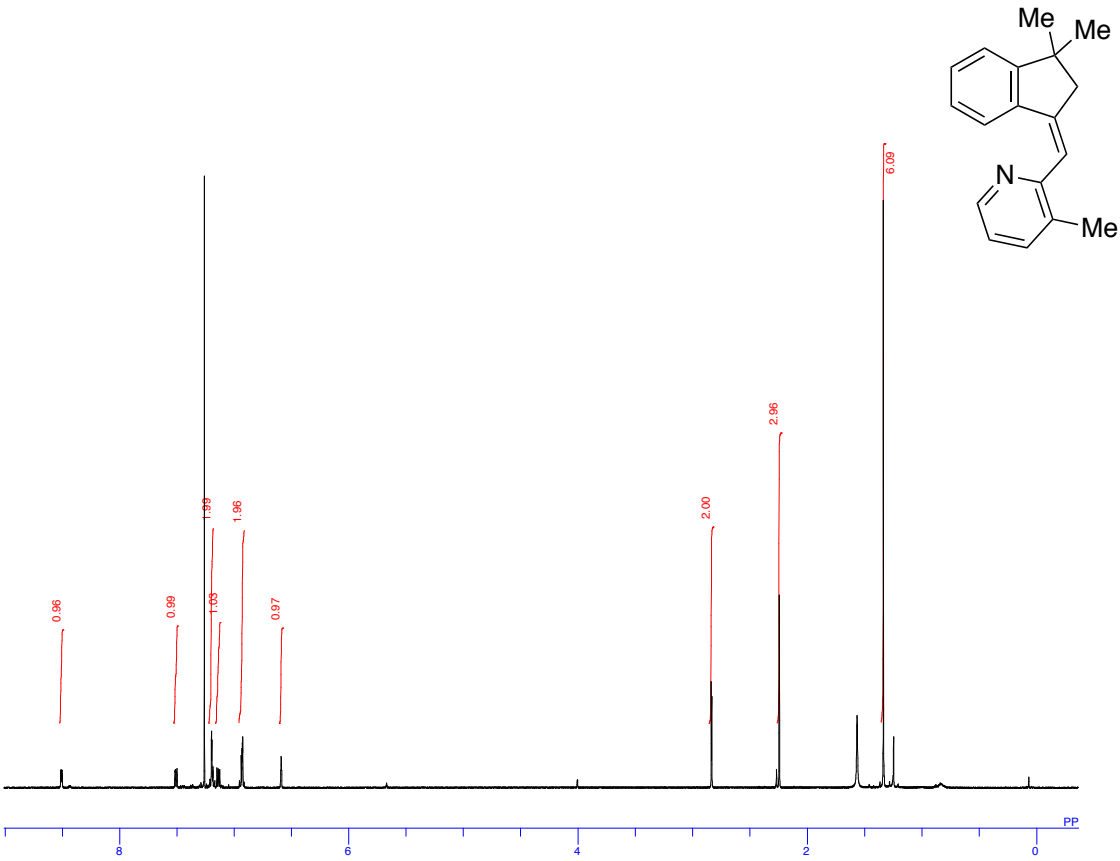
75
50
25

**2b**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2.00
1.99
1.96
1.03
0.99
0.97
0.96
PP
8
6
4
2
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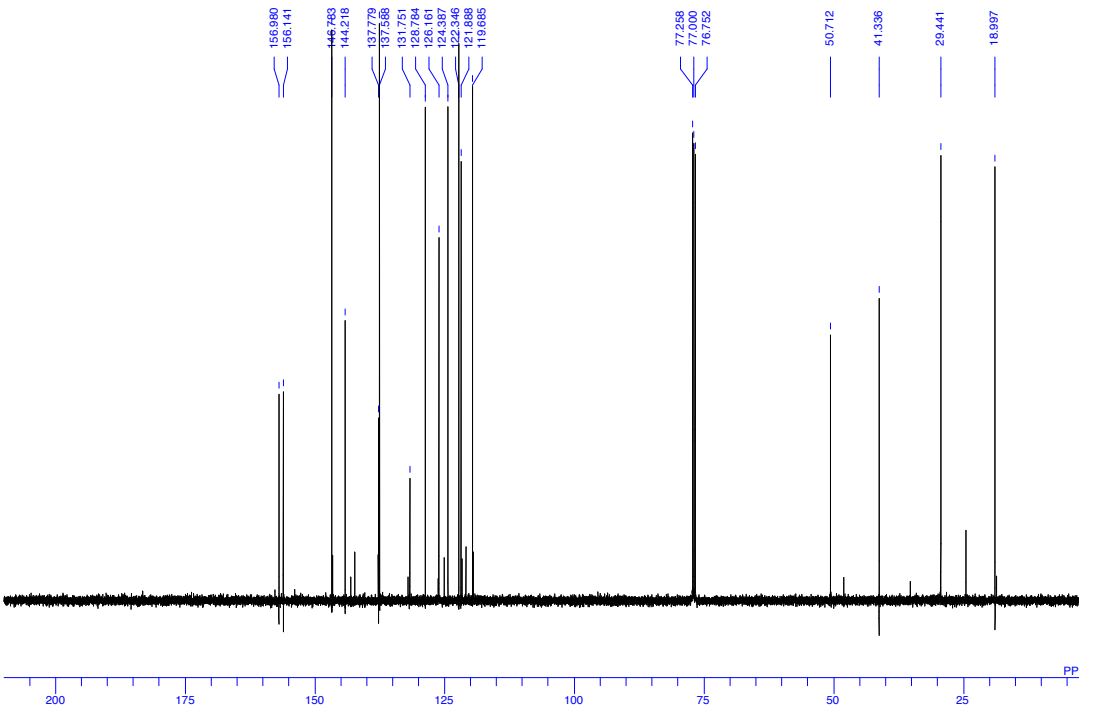
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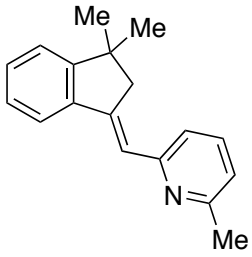
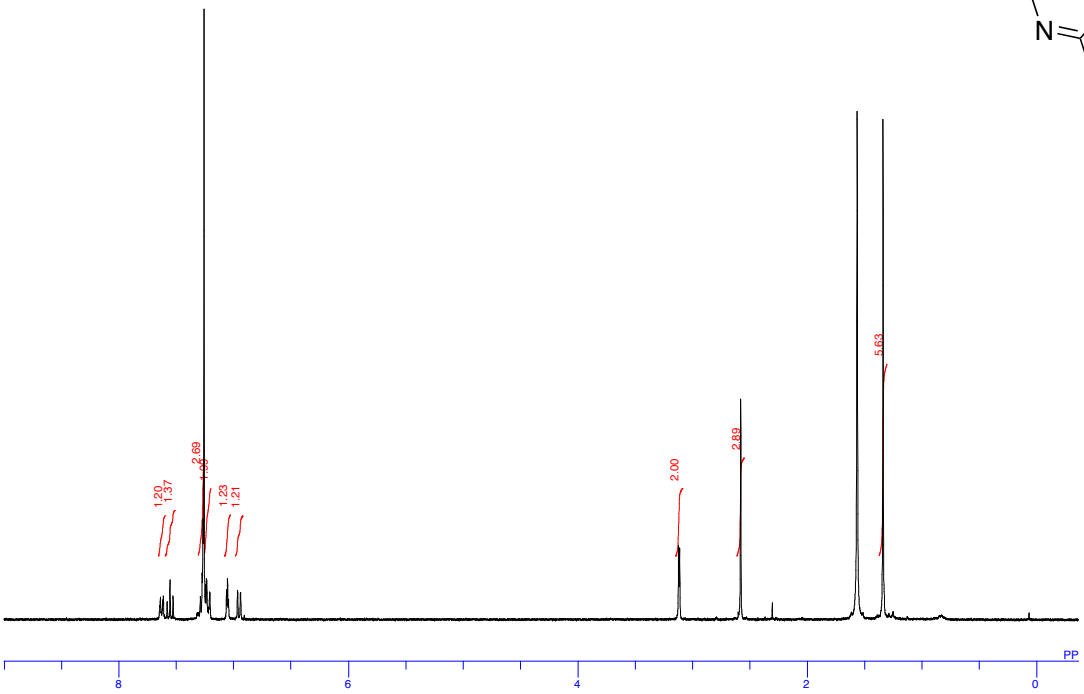
156.141
146.783
144.218
137.779
137.588
131.751
128.784
126.161
124.387
122.346
121.888
119.685
77.258
77.000
76.752
50.712
41.336
29.441
18.997
PP
200
175
150
125
100
75
50
25

**3c**


Me
Me
N
Me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1.37
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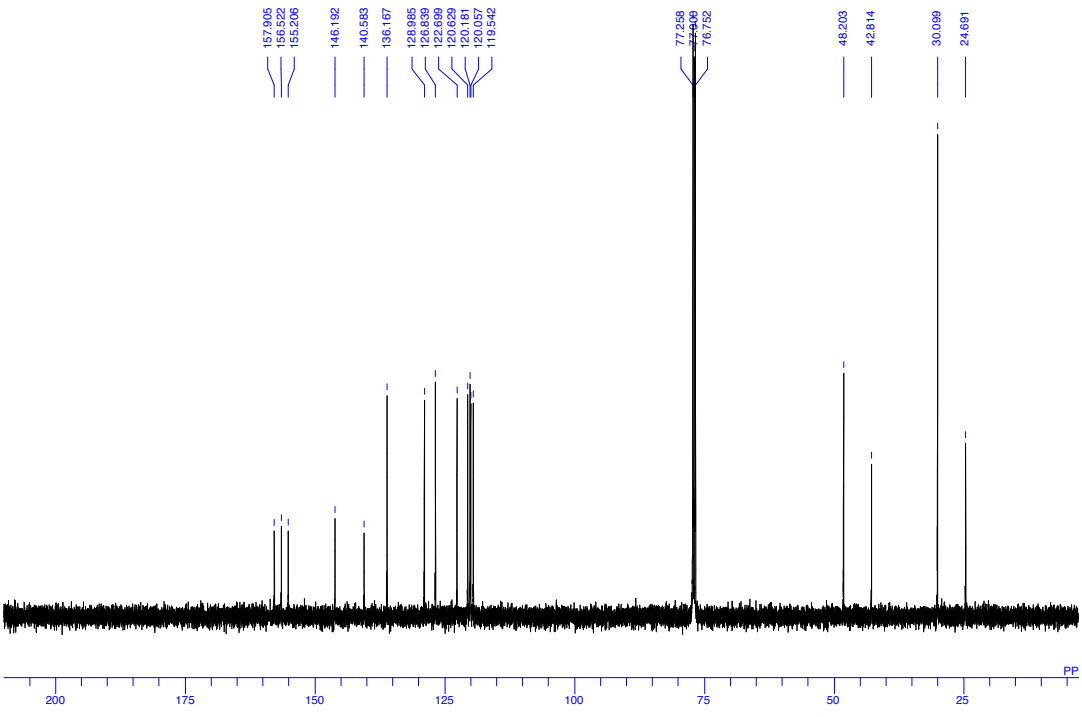
1.00
1.23
1.21
2.00
2.89
5.63
PP
8
6
4
2
0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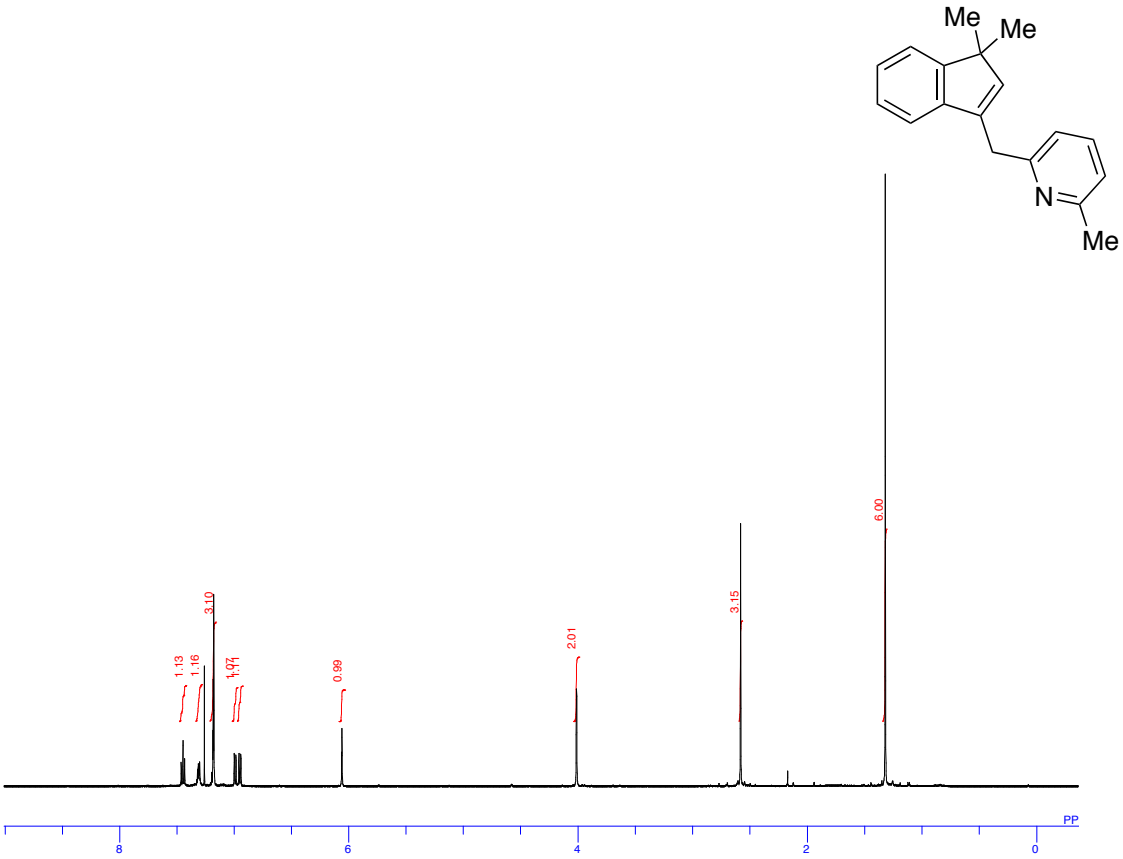
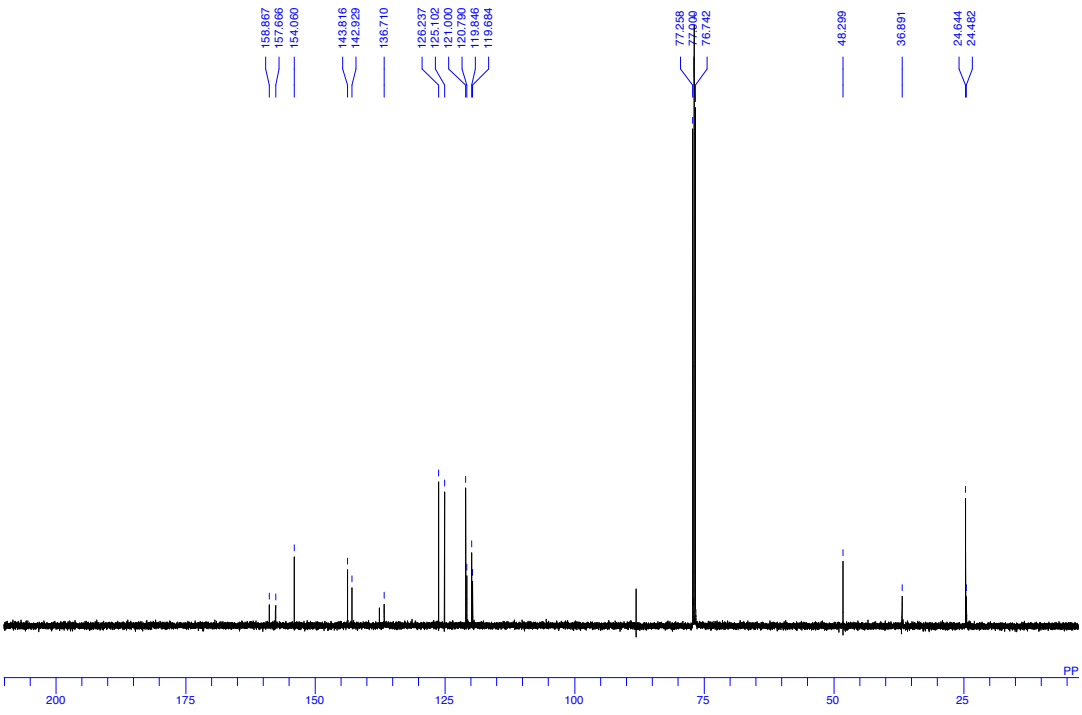
156.522
155.206
146.192
140.583
136.167
128.985
126.839
122.699
120.629
120.151
120.057
119.542
77.258
77.000
76.752
48.203
42.814
30.099
24.691
PP
200
175
150
125
100
75
50
25

**4c**


Me
Me
N
Me
1.13
1.16
3.10
1.97
1.91
0.99
2.01
3.15
6.00
PP
8
6
4
2
0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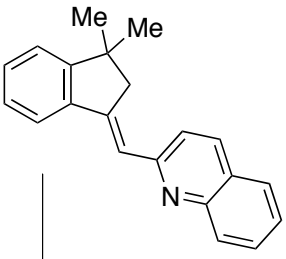
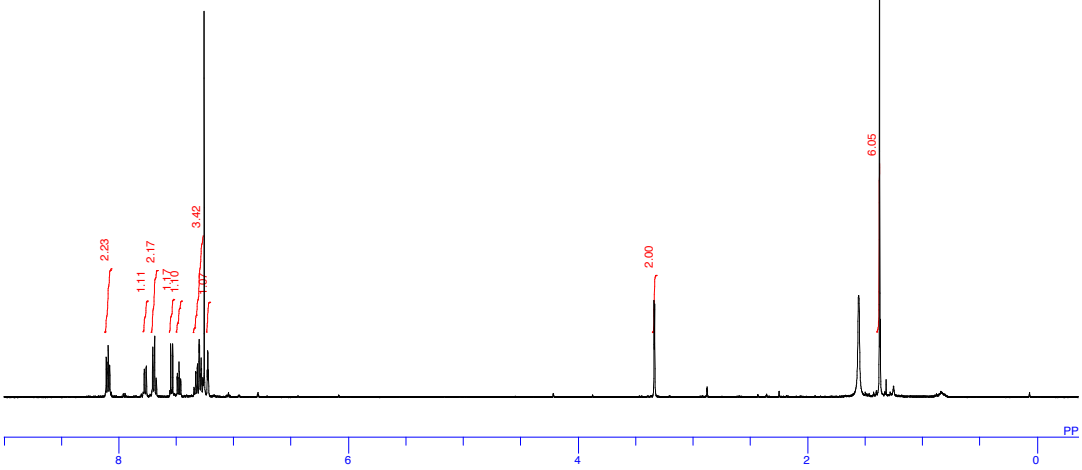
157.666
154.060
143.816
142.929
136.710
126.237
125.102
121.000
120.790
119.846
119.684
77.258
77.000
76.742
48.299
36.891
24.644
24.482
PP
200
175
150
125
100
75
50
25

**3d**


Me
Me
N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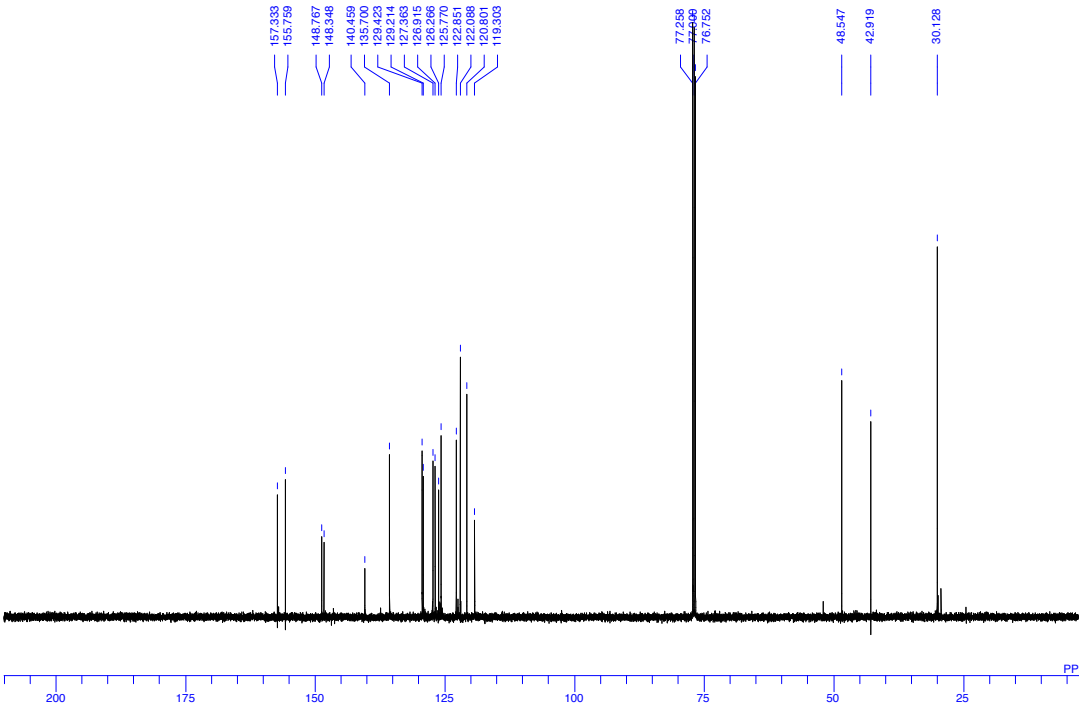
1.11
2.17
1.17
1.16
3.42
1.07
2.00
6.05
PP
8
6
4
2
0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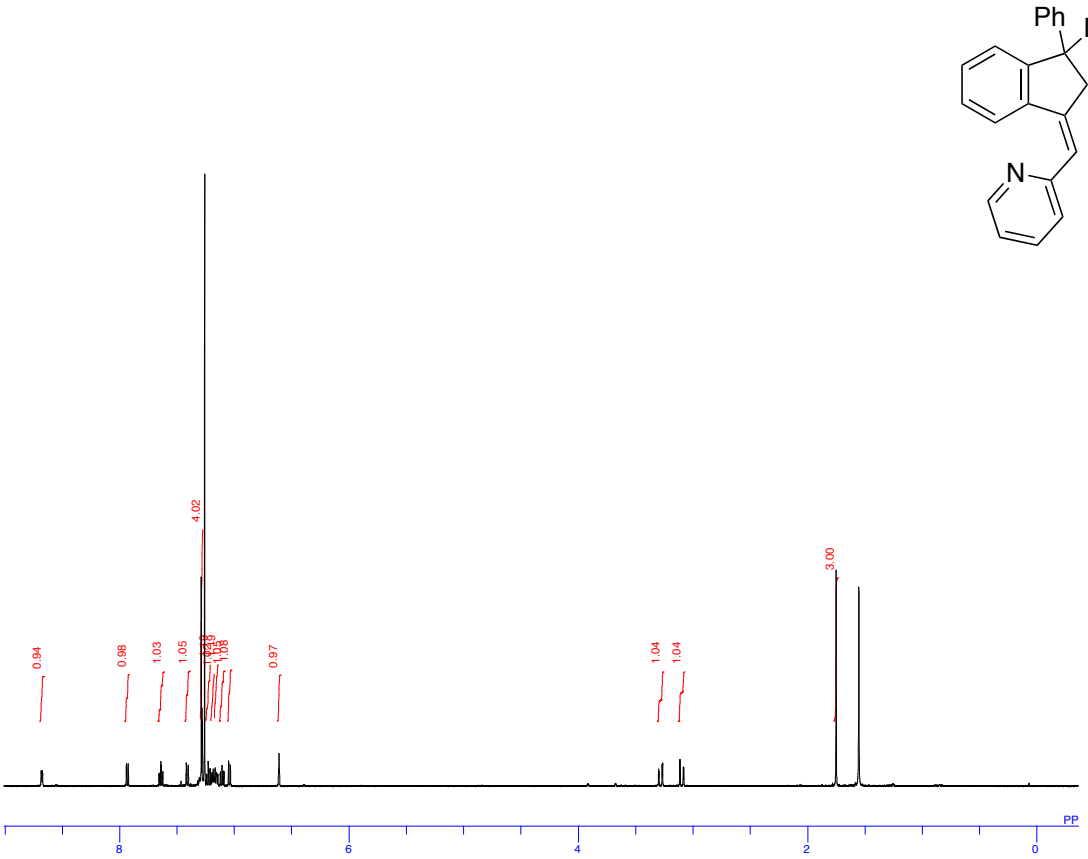
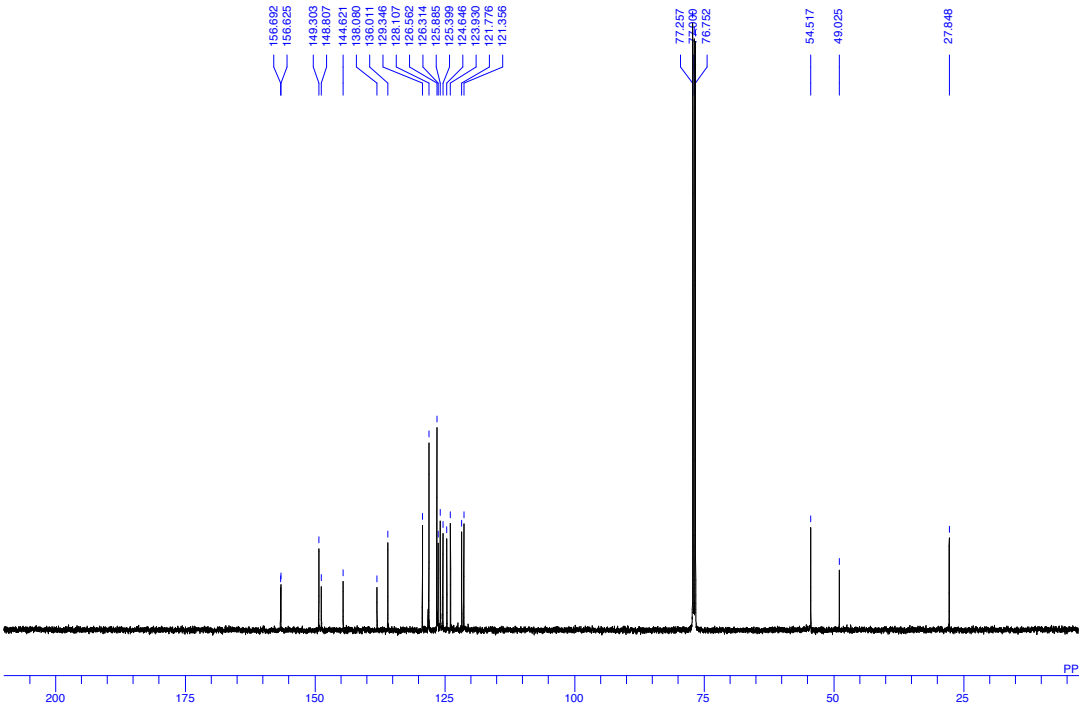
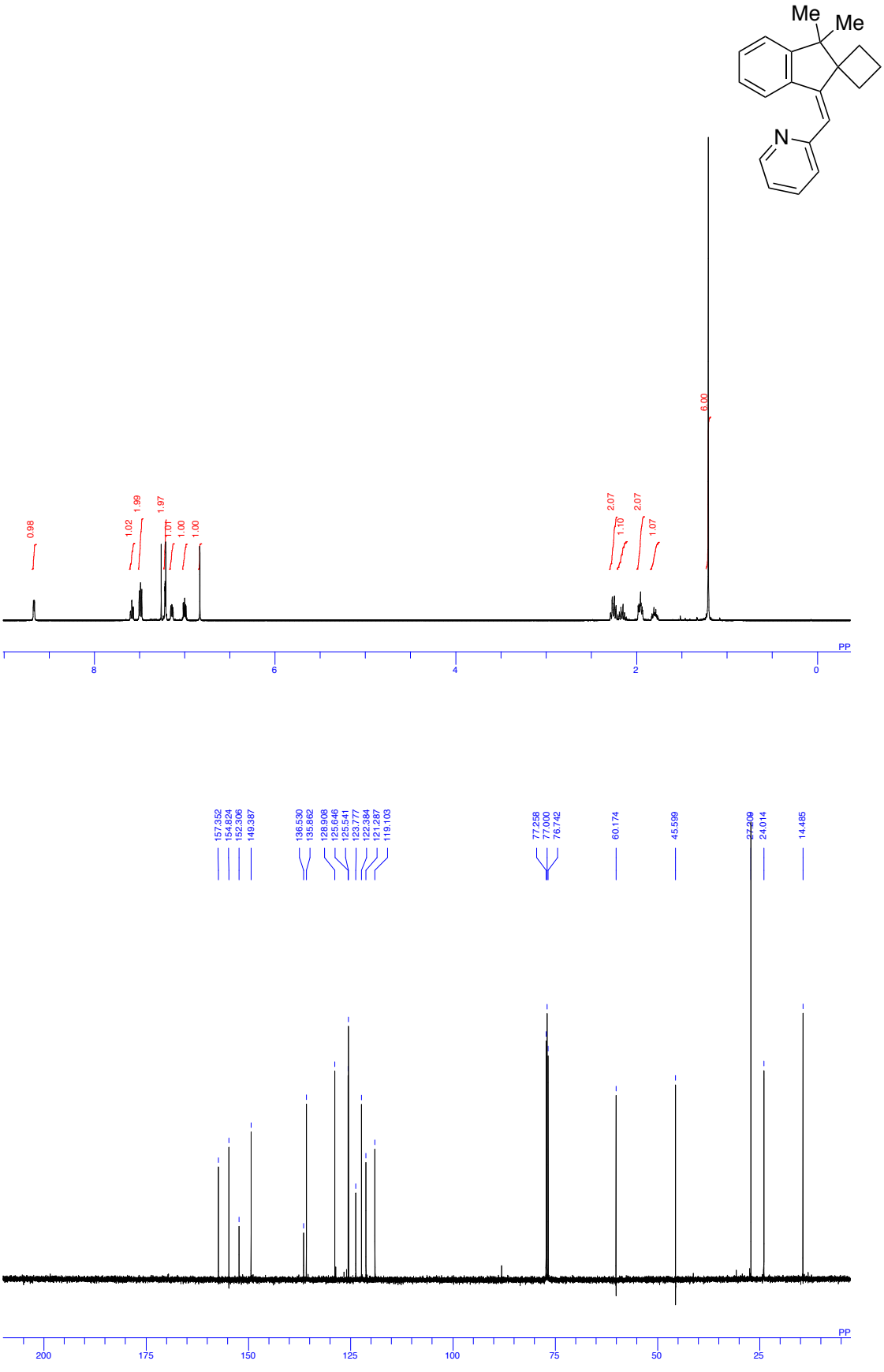
155.759
148.767
148.348
140.459
135.700
129.423
129.214
127.363
126.915
126.266
125.770
122.851
122.088
120.801
119.303
77.258
76.752
48.547
42.919
30.128
PP
200
175
150
125
100
75
50
25

**2e**

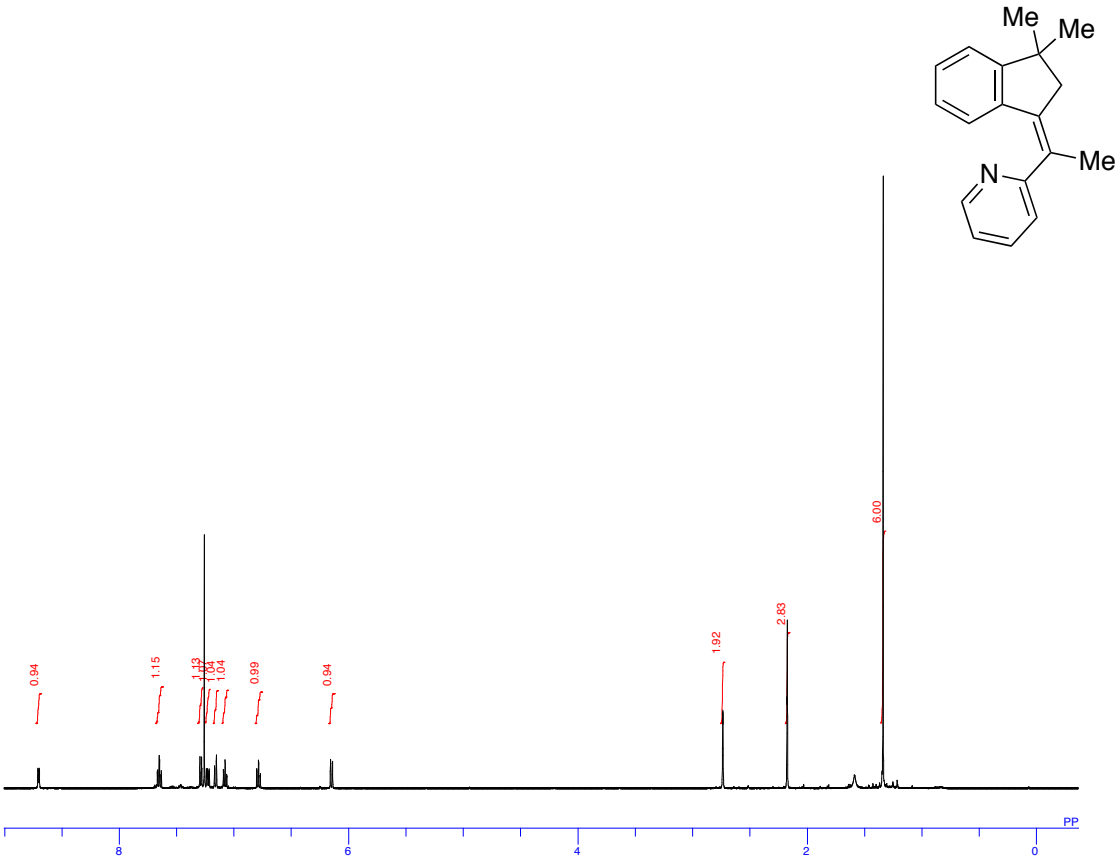

Ph
Me
N
4.02
3.00
0.94
0.98
1.03
1.05
1.08
0.97
1.04
1.04
PP
8
6
4
2
0

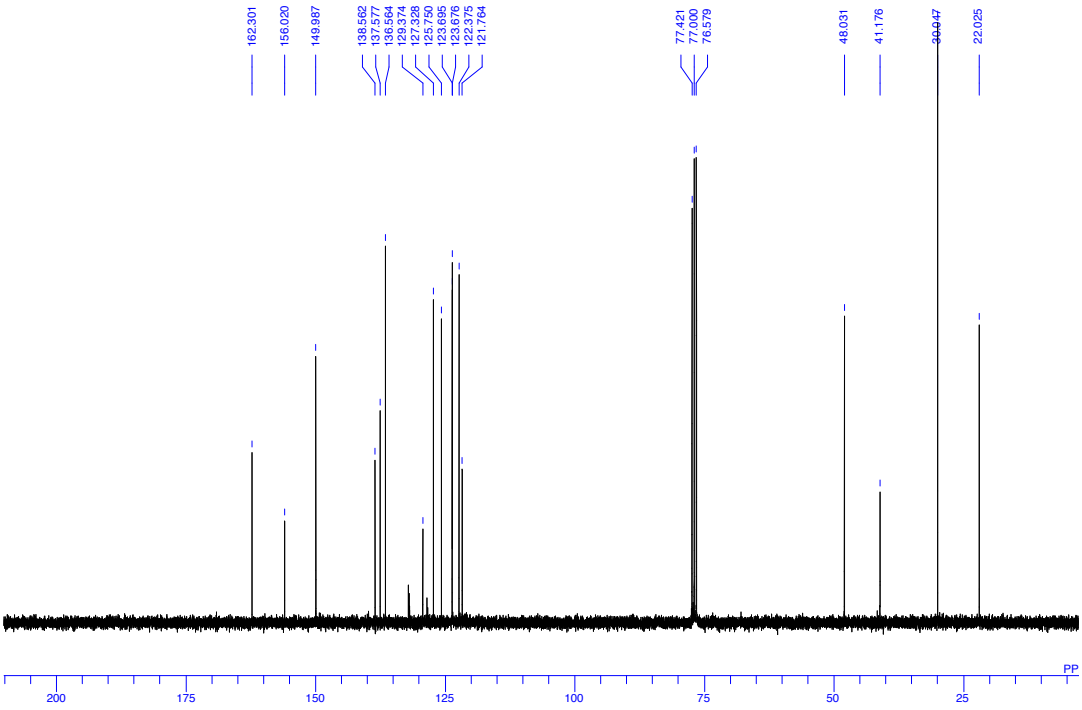

156.692
156.625
149.303
148.807
144.621
138.080
136.011
129.346
128.107
126.502
126.314
125.885
125.399
124.646
123.930
121.776
121.356
77.257
76.752
54.517
49.025
27.848
PP
200
175
150
125
100
75
50
25

**2g**

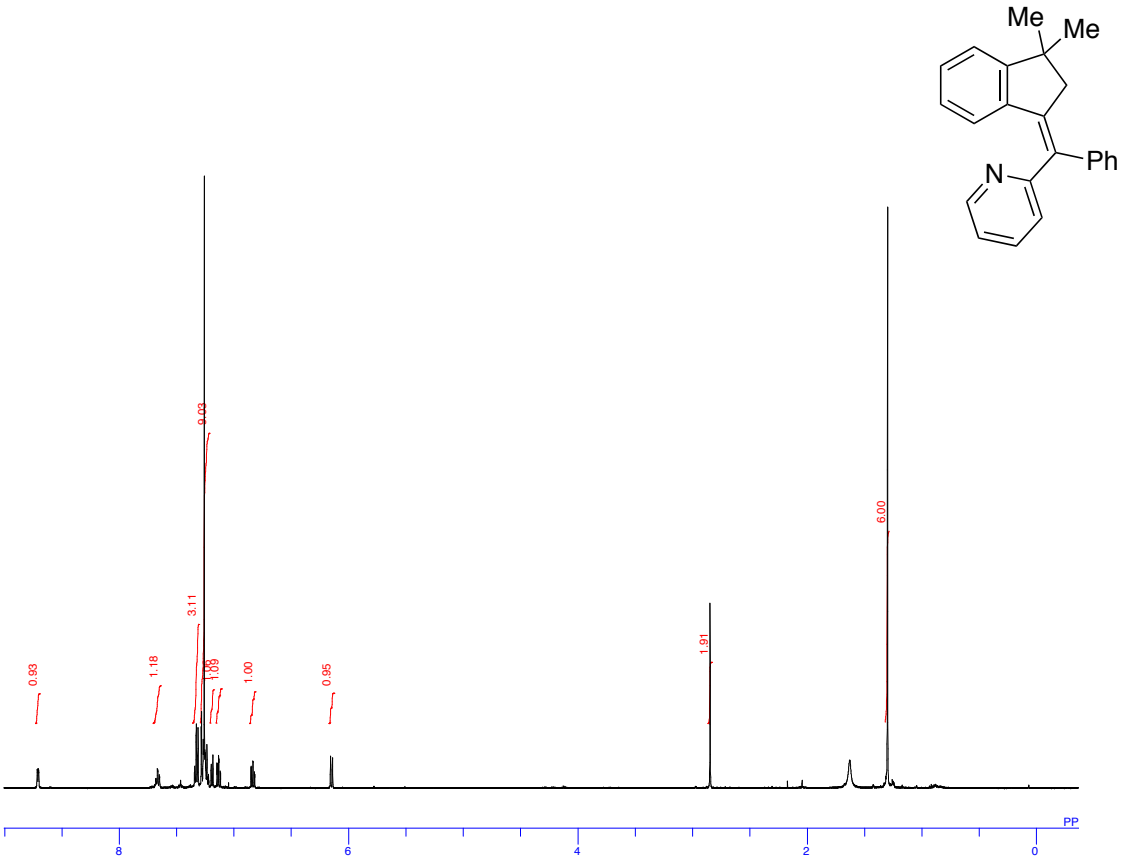

Me
Me
N
0.98
1.02
1.99
1.97
1.01
1.00
1.00
2.07
1.10
2.07
1.07
6.00
PP
8
6
4
2
0
157.352
154.824
152.306
149.387
136.530
135.862
128.908
125.646
125.541
123.777
123.384
121.287
119.103
77.258
77.000
76.742
60.174
45.599
27.209
24.014
14.485
PP
200
175
150
125
100
75
50
25

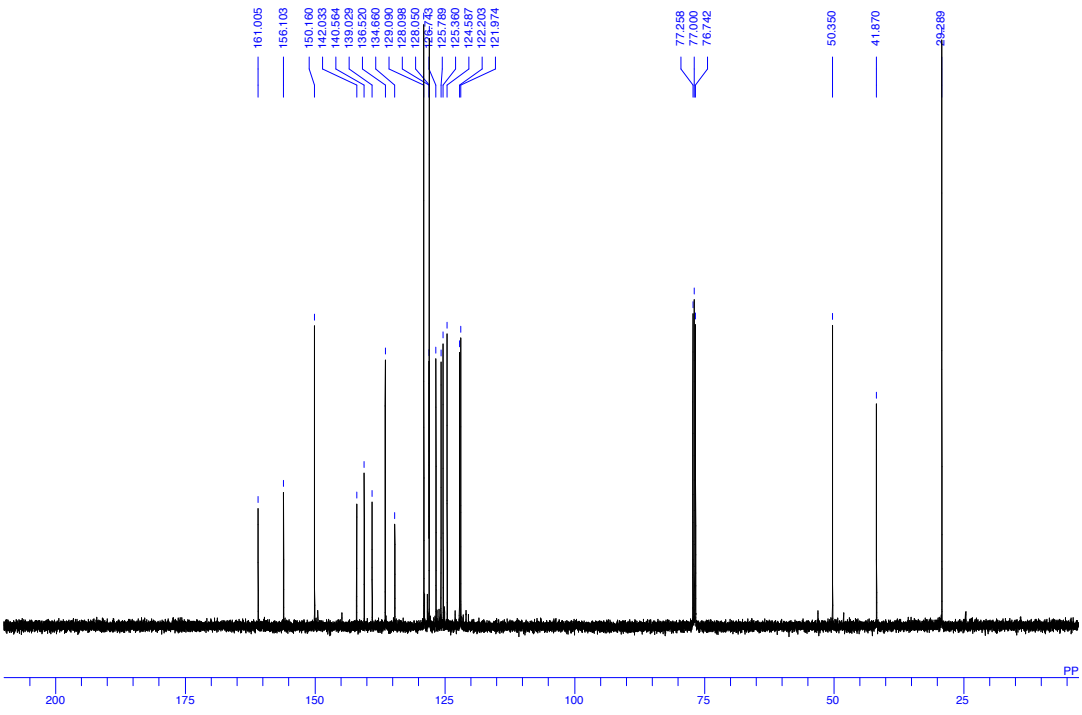
**2h**


Me
Me
Me
N
0.94
1.15
1.13
1.00
1.04
0.99
0.94
1.92
2.83
6.00
PP
8
6
4
2
0


162.301
156.020
149.987
138.562
137.577
136.564
129.374
127.328
125.750
123.695
123.676
122.375
121.764
77.421
77.000
76.579
48.031
41.176
30.947
22.025
PP
200
175
150
125
100
75
50
25

**2i**


Me
Me
Ph
N
0.93
1.18
3.11
9.03
1.08
1.09
1.00
0.95
1.91
6.00
PP
8
6
4
2
0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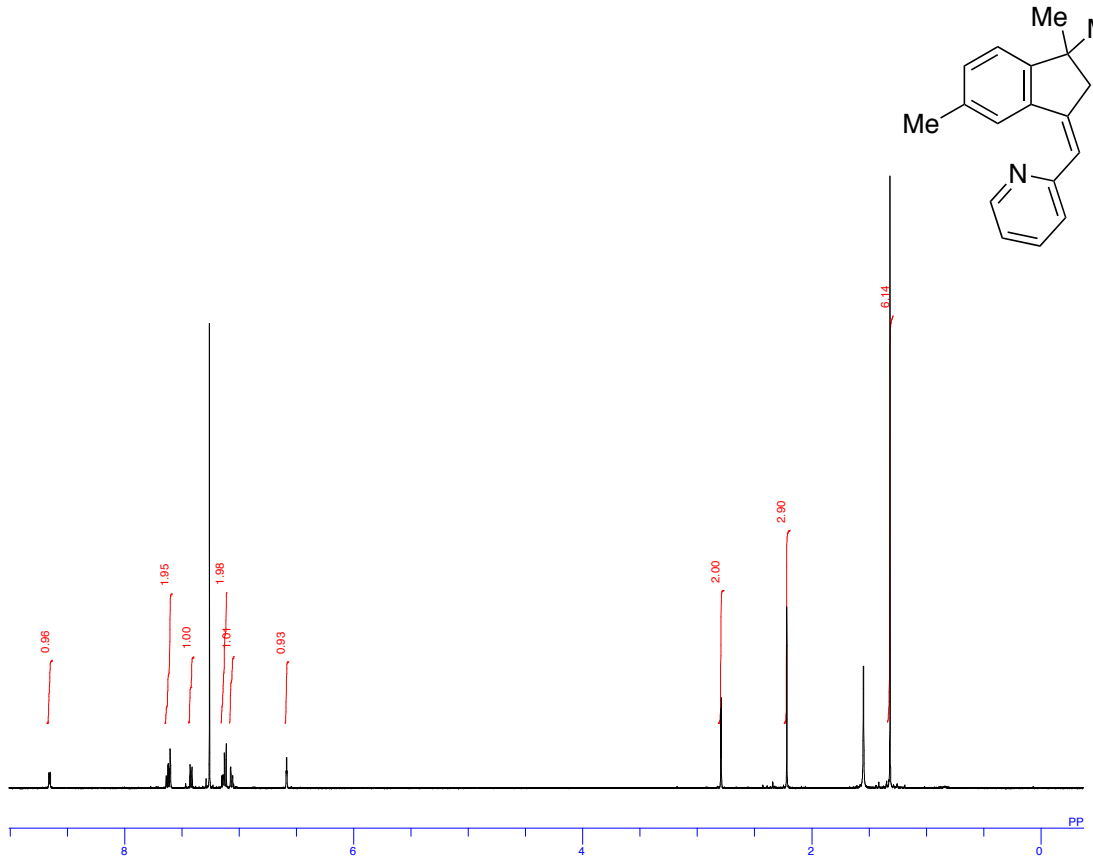
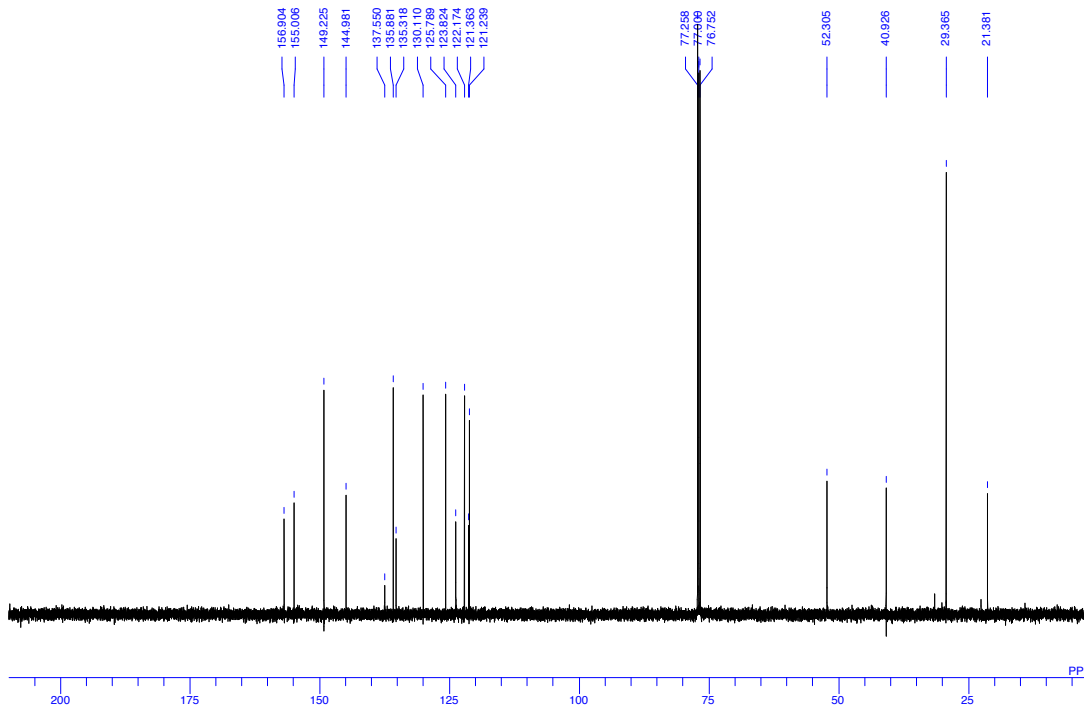
156.103
150.160
142.033
140.564
139.029
136.520
134.660
129.090
128.098
128.050
126.743
125.789
125.360
124.587
122.203
121.974
77.258
77.000
76.742
50.350
41.870
29.289
PP
200
175
150
125
100
75
50
25

**2j**


Me
Me
Me
N
0.96
1.95
1.00
1.98
1.01
0.93
2.00
2.90
6.14
PP
8
6
4
2
0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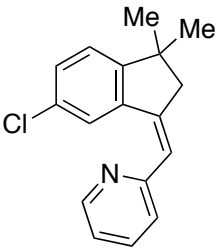
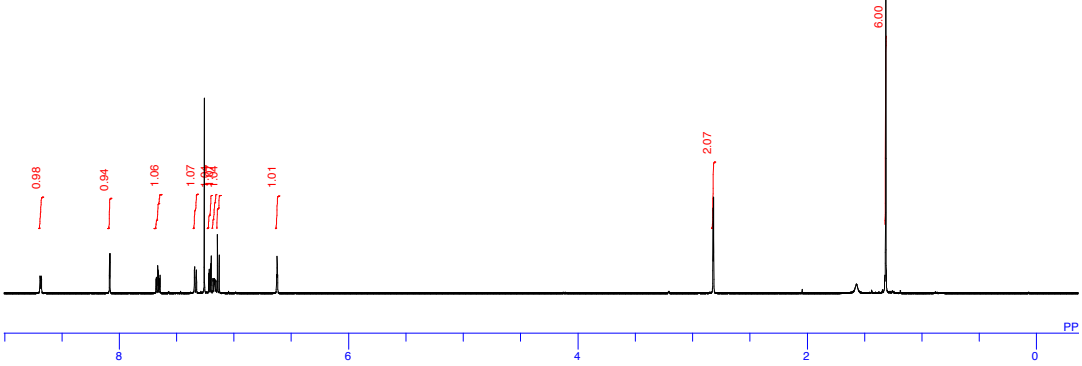
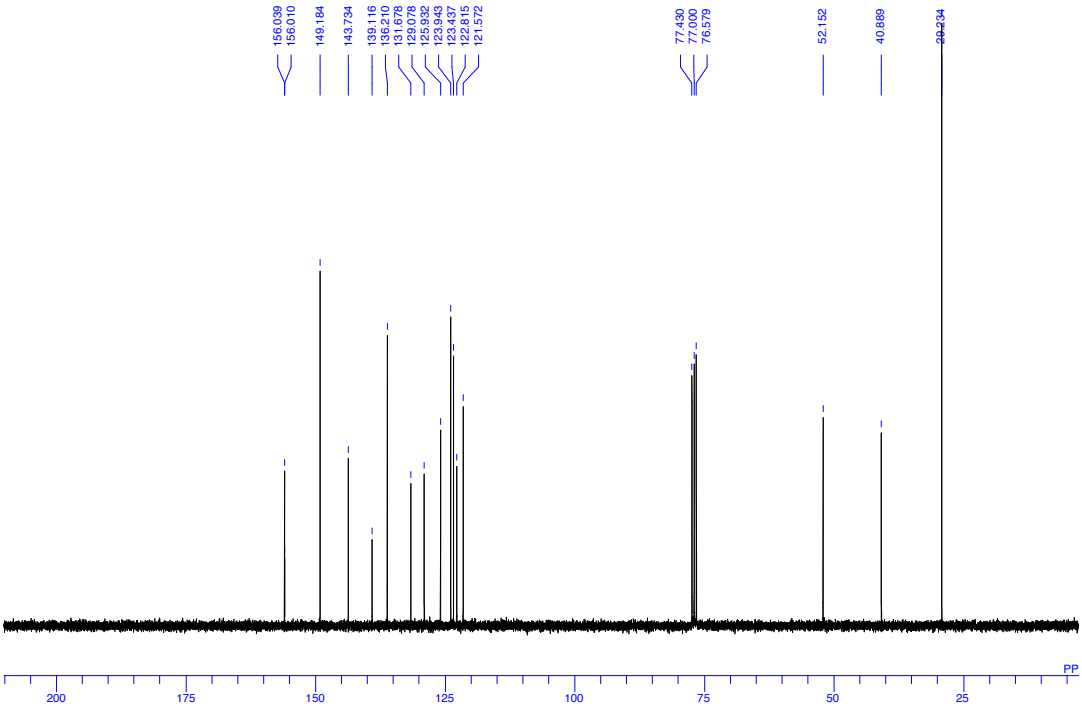
155.006
149.225
144.961
137.550
135.881
135.318
130.110
125.789
123.824
122.174
121.363
121.239
77.258
77.006
76.752
52.305
40.926
29.365
21.381
PP
200
175
150
125
100
75
50
25

**2k**


Me
Me
Cl
N


0.98
0.94
1.06
1.07
1.04
1.01
2.07
6.00
PP
8
6
4
2
0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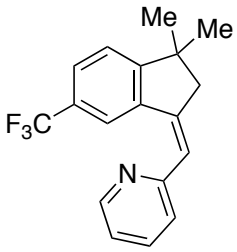
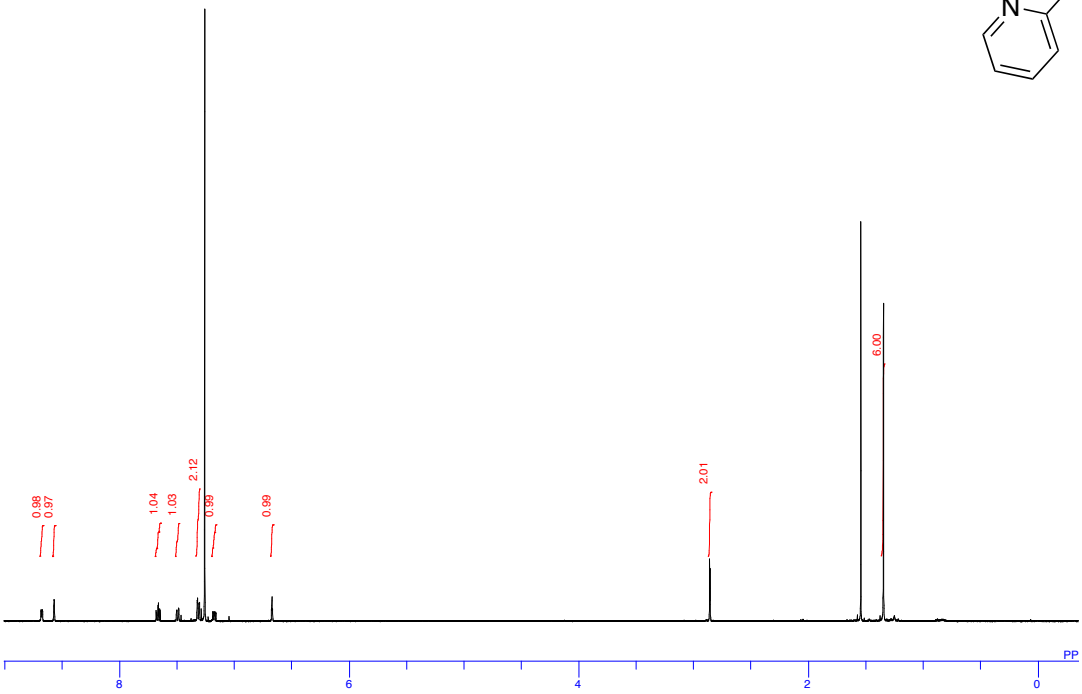
156.010
149.184
143.734
139.116
136.210
131.678
129.078
125.932
123.943
123.457
122.615
121.572
77.430
77.000
76.579
52.152
40.889
29.234
PP
200
175
150
125
100
75
50
25

**2l**


Me
Me
F3C
N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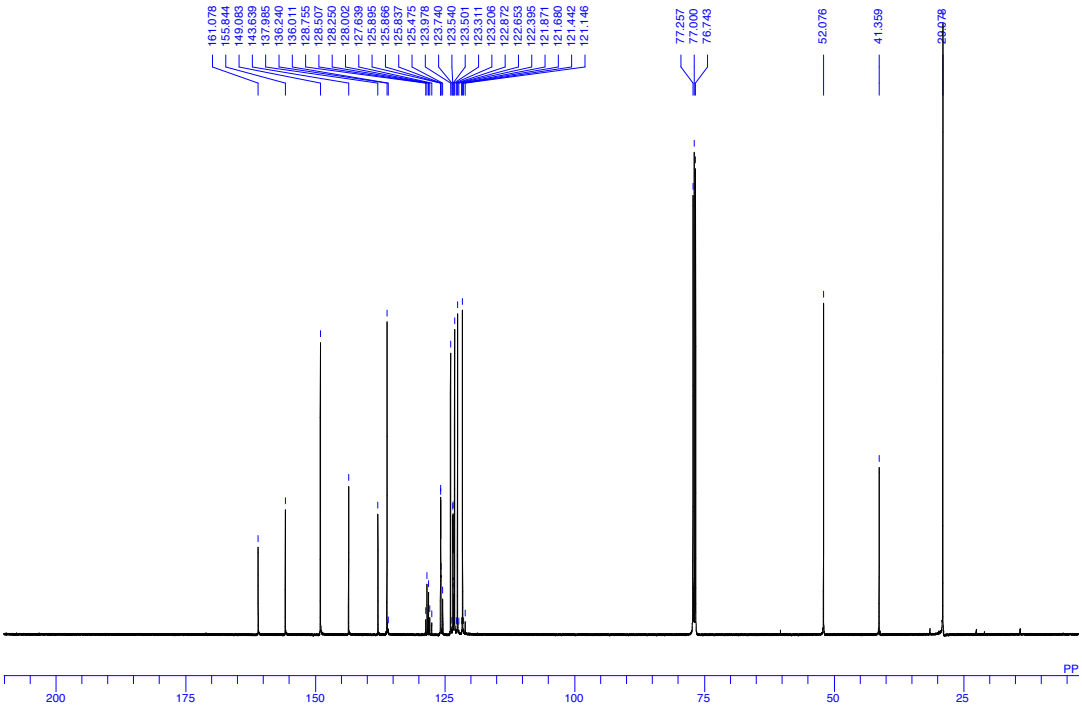
0.97
1.04
1.03
2.12
0.99
0.99
2.01
6.00
PP
8
6
4
2
0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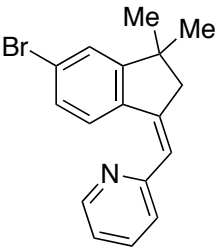
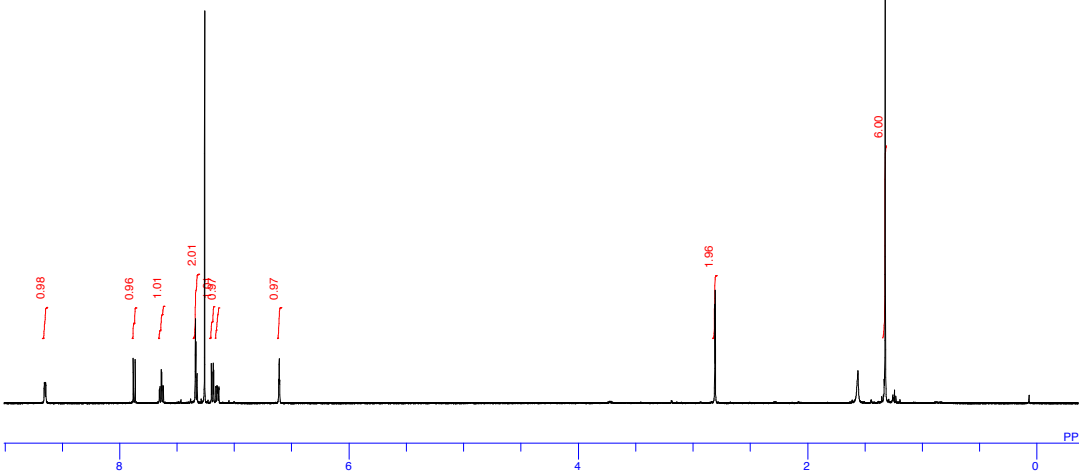
143.639
137.985
136.240
136.011
128.755
128.507
128.250
128.002
127.639
125.895
125.866
125.837
125.475
123.978
123.740
123.540
123.511
123.311
123.206
122.872
122.653
122.395
121.871
121.680
121.442
121.146
77.257
77.000
76.743
52.076
41.359
29.078
PP
200
175
150
125
100
75
50
25

**2m**


Me
Me
Br
N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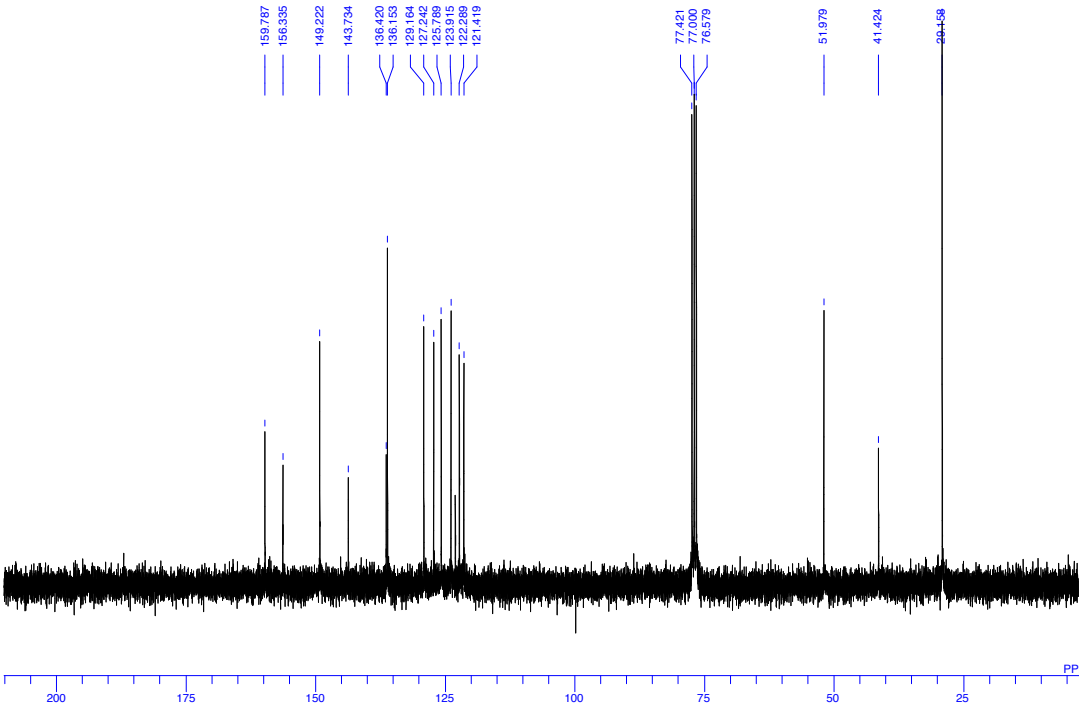
0.96
1.01
2.01
0.97
0.97
1.96
6.00
PP
8
6
4
2
0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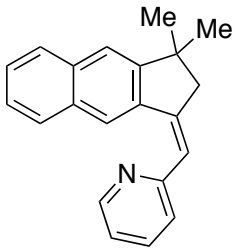
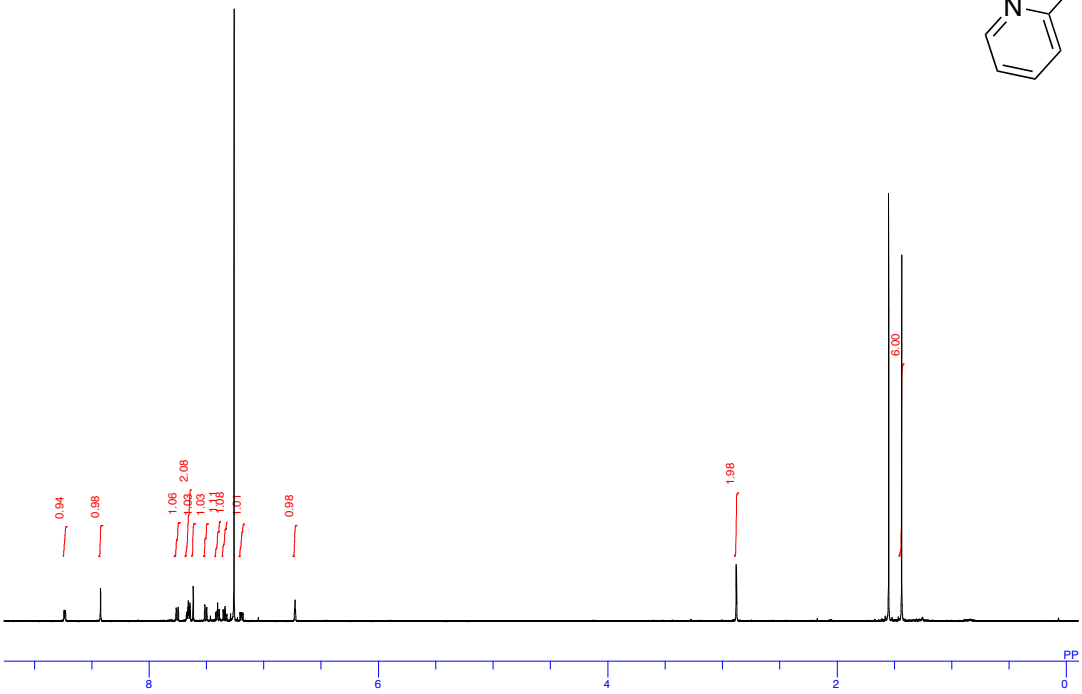
143.734
136.420
136.153
129.164
127.242
125.798
123.915
122.289
121.419
77.421
77.000
76.579
51.979
41.424
29.458
PP
200
175
150
125
100
75
50
25

**2n**


Me
Me
N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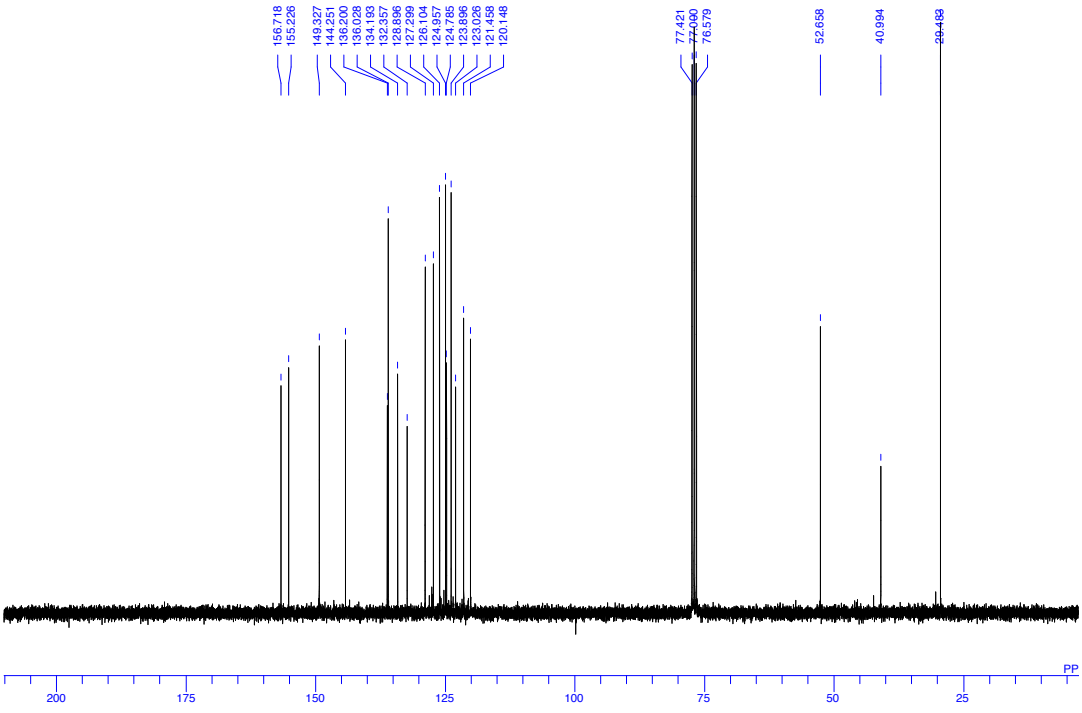
0.98
1.06
2.08
1.03
1.01
0.98
1.98
6.00
PP
8
6
4
2
0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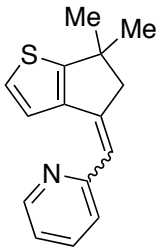
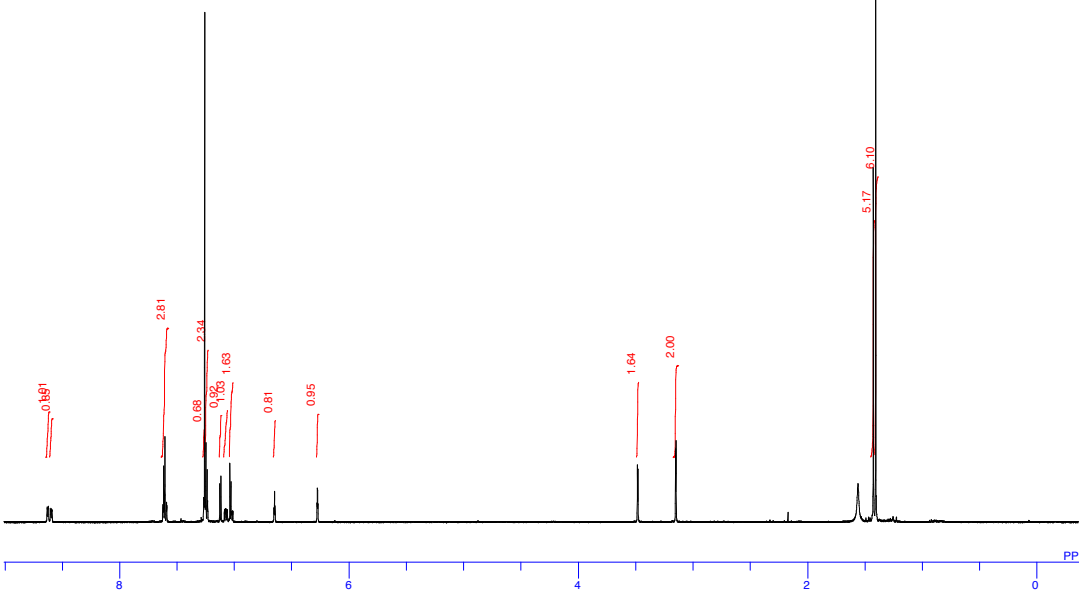
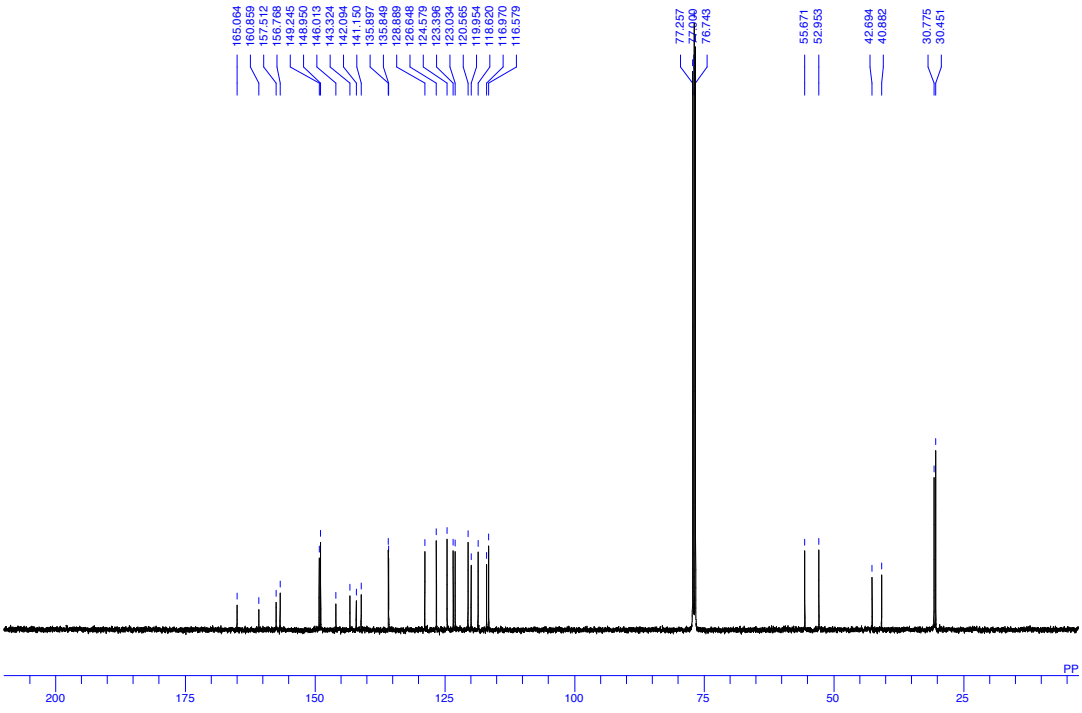
155.226
149.327
144.251
136.200
136.028
134.193
132.357
128.896
127.299
126.104
124.957
124.785
123.896
123.026
121.458
120.148
77.421
77.000
76.579
52.658
40.994
29.483
PP
200
175
150
125
100
75
50
25

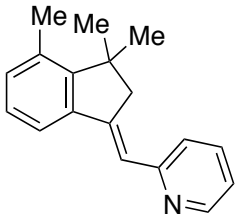
**2o** and **3o**

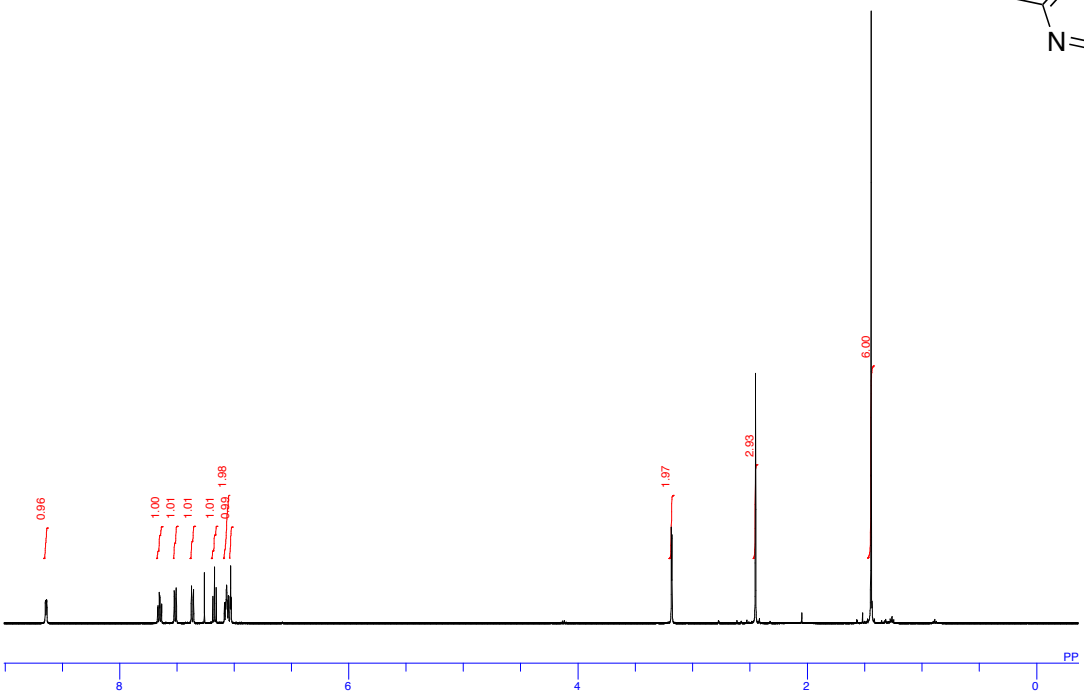

Me
Me
S
N

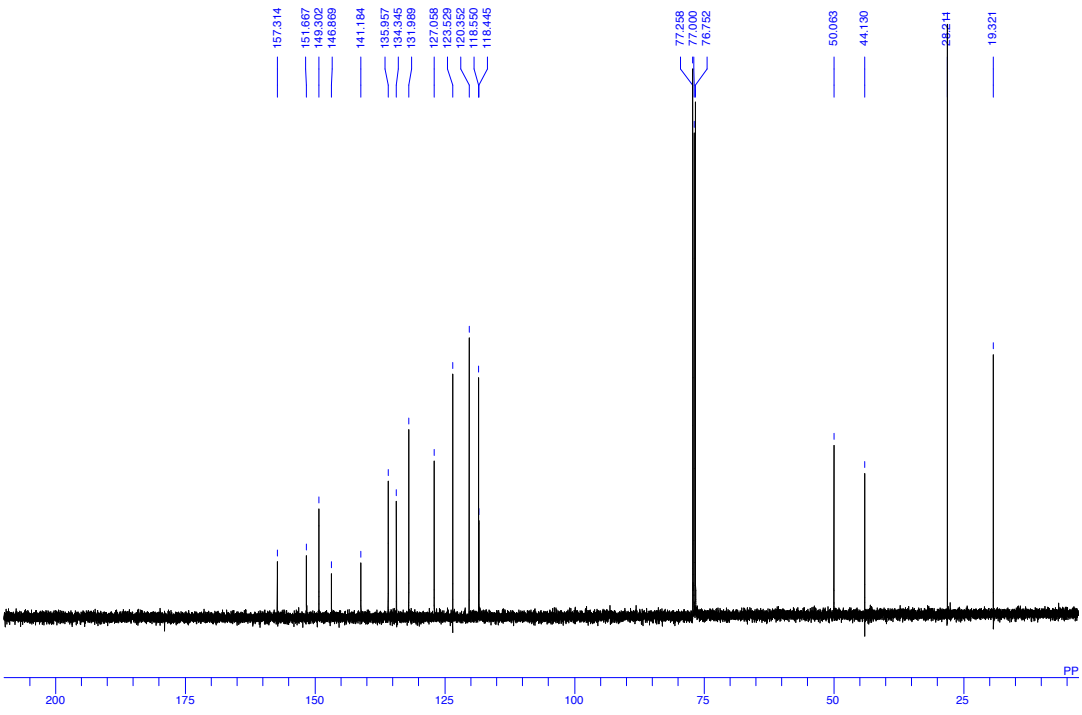

0.91
0.89
2.81
2.94
0.68
0.92
1.03
1.63
0.81
0.95
1.64
2.00
5.17
6.10
PP
8
6
4
2
0


165.064
160.859
157.512
156.768
149.245
148.950
146.013
143.324
142.094
141.150
135.897
135.849
128.889
126.648
124.578
123.396
123.034
120.565
119.954
118.620
116.970
116.579
77.257
77.000
76.743
55.671
52.953
42.694
40.882
30.775
30.451
PP
200
175
150
125
100
75
50
25

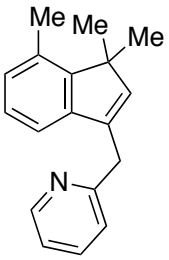
**3p**

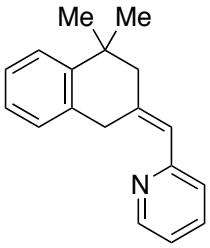

Me
Me
Me
N


0.96
1.00
1.01
1.01
1.01
1.01
0.99
1.98
1.97
2.93
6.00
PP
8
6
4
2
0


157.314
151.667
149.302
146.869
141.184
135.957
134.345
131.969
127.058
123.529
120.352
118.550
118.445
77.258
77.000
76.752
50.063
44.130
28.214
19.321
PP
200
175
150
125
100
75
50
25

**4p** and **5**


Me
Me
Me
N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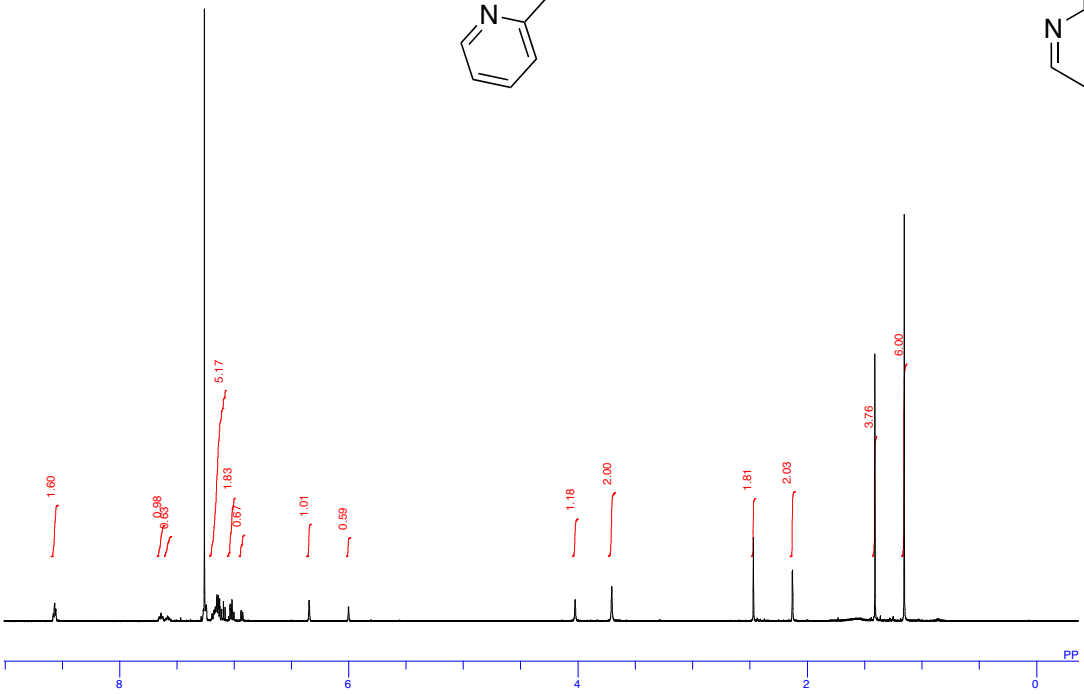
Me
N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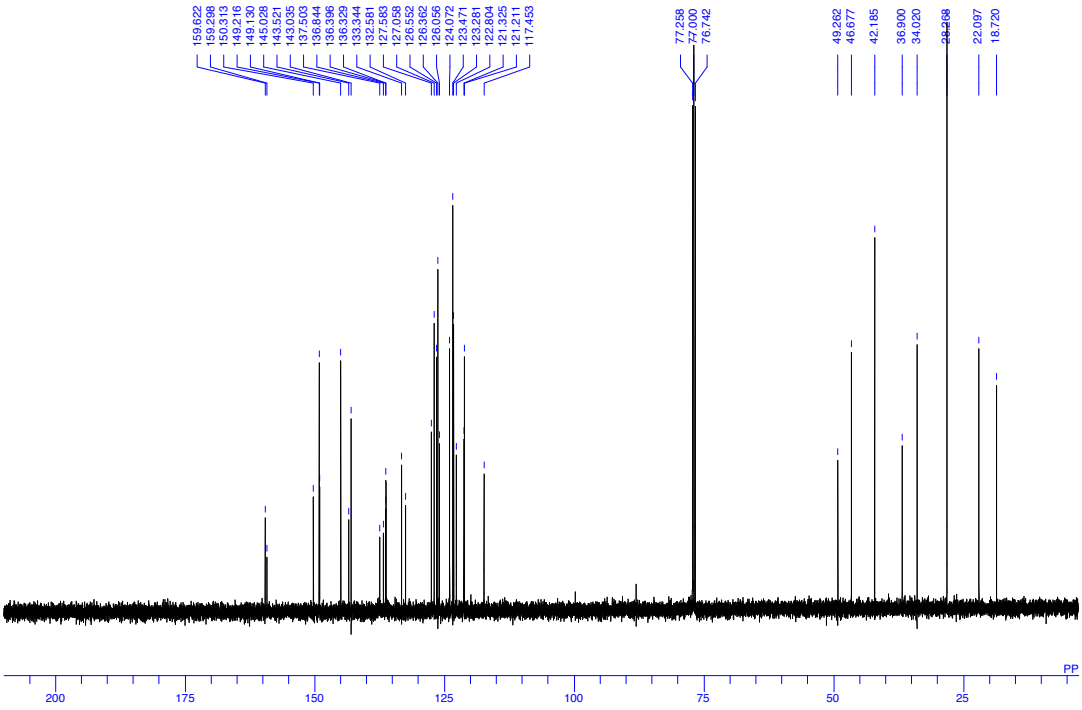
0.98
6.83
5.17
1.83
0.67
1.01
0.59
1.18
2.00
1.81
2.03
3.76
6.00
PP
8
6
4
2
0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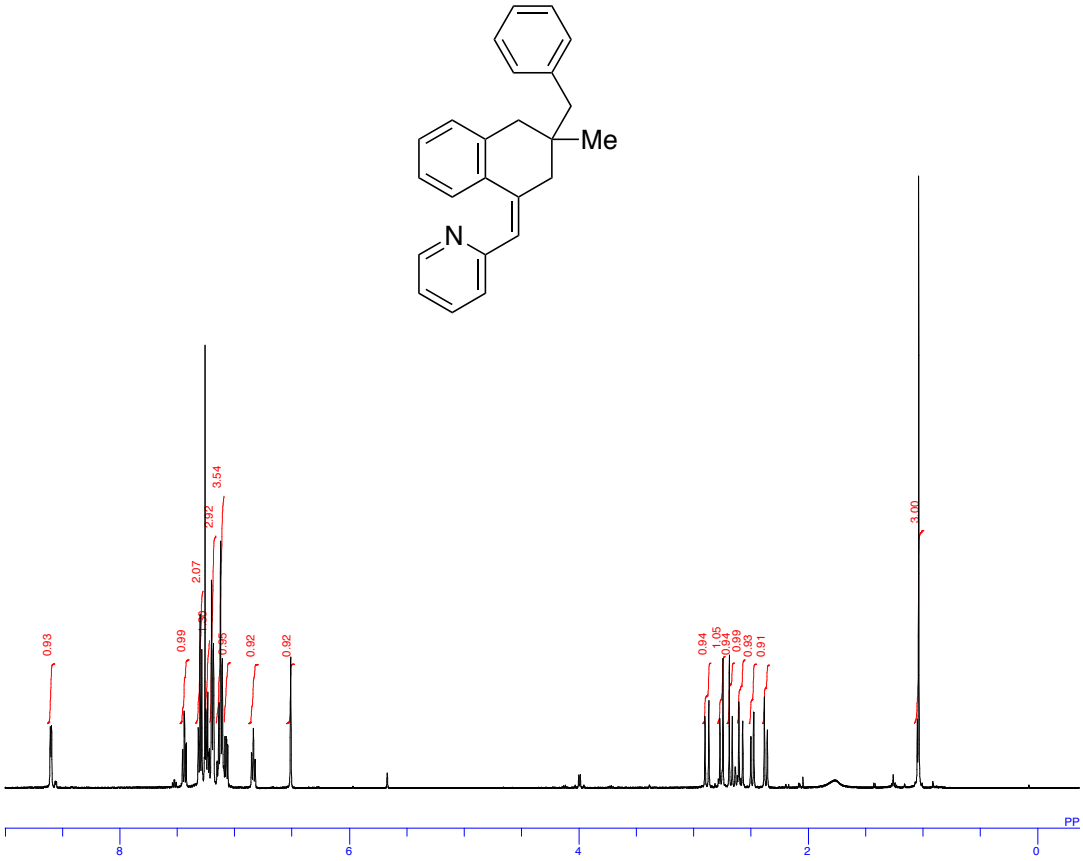
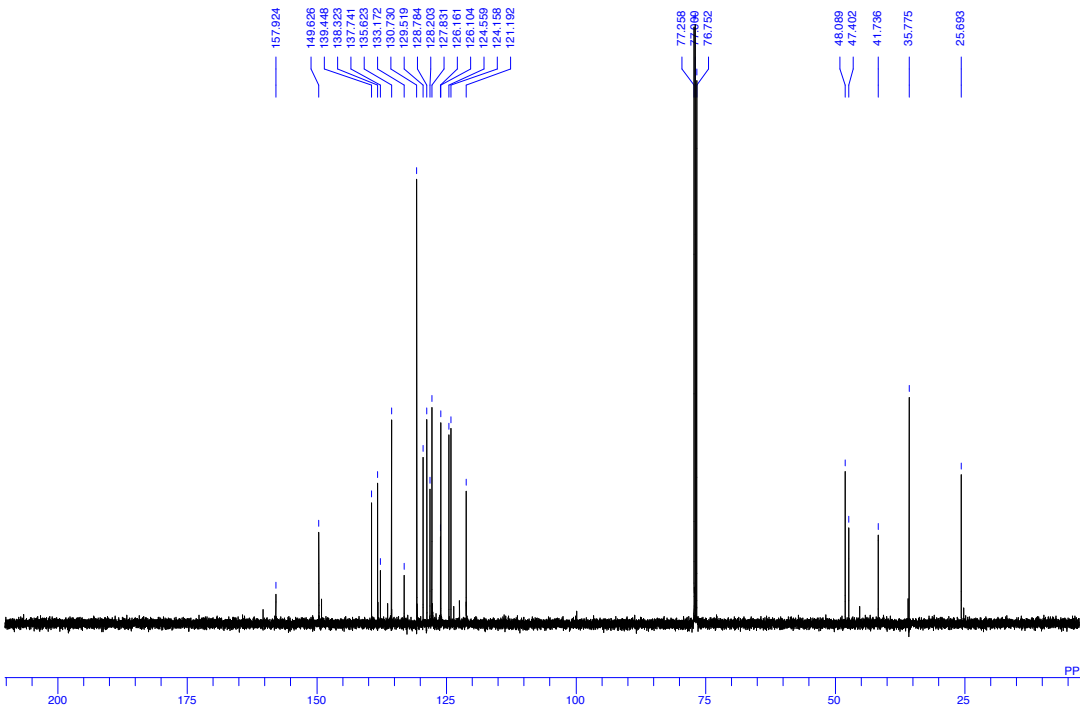
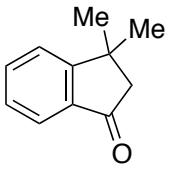
159.298
150.313
149.216
149.130
145.028
143.521
143.035
137.503
136.844
136.396
136.329
133.344
132.581
127.583
127.038
126.552
126.362
126.055
124.072
123.471
123.261
122.804
121.325
121.211
117.453
77.258
77.000
76.742
49.262
46.677
42.185
36.900
34.020
22.097
18.720
PP
200
175
150
125
100
75
50
25

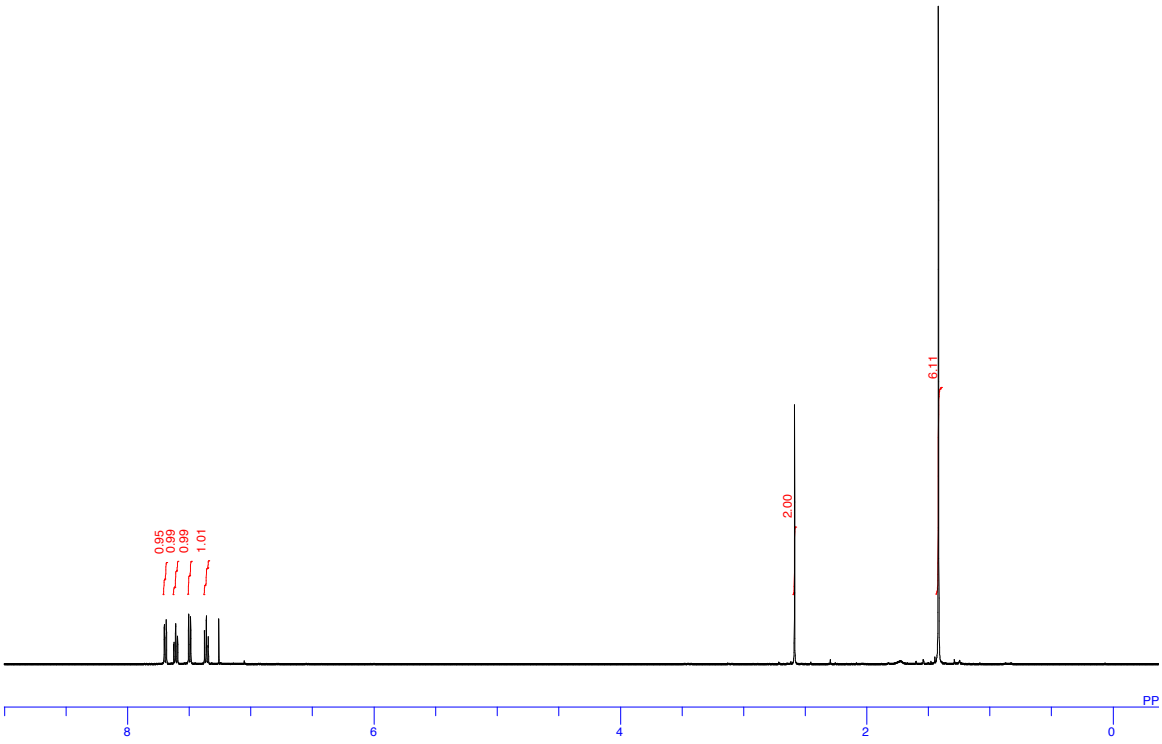
**2q**


Me
N
0.93
0.99
2.07
2.92
3.54
0.95
0.92
0.92
0.94
1.05
0.94
0.99
0.93
0.91
3.00
PP
8
6
4
2
0

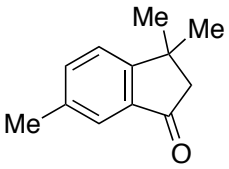

157.924
149.626
139.448
138.323
137.741
135.623
133.172
130.730
129.519
128.784
128.203
127.831
126.161
126.104
124.559
124.198
121.192
77.258
76.752
48.089
47.402
41.736
35.775
25.693
PP
200
175
150
125
100
75
50
25

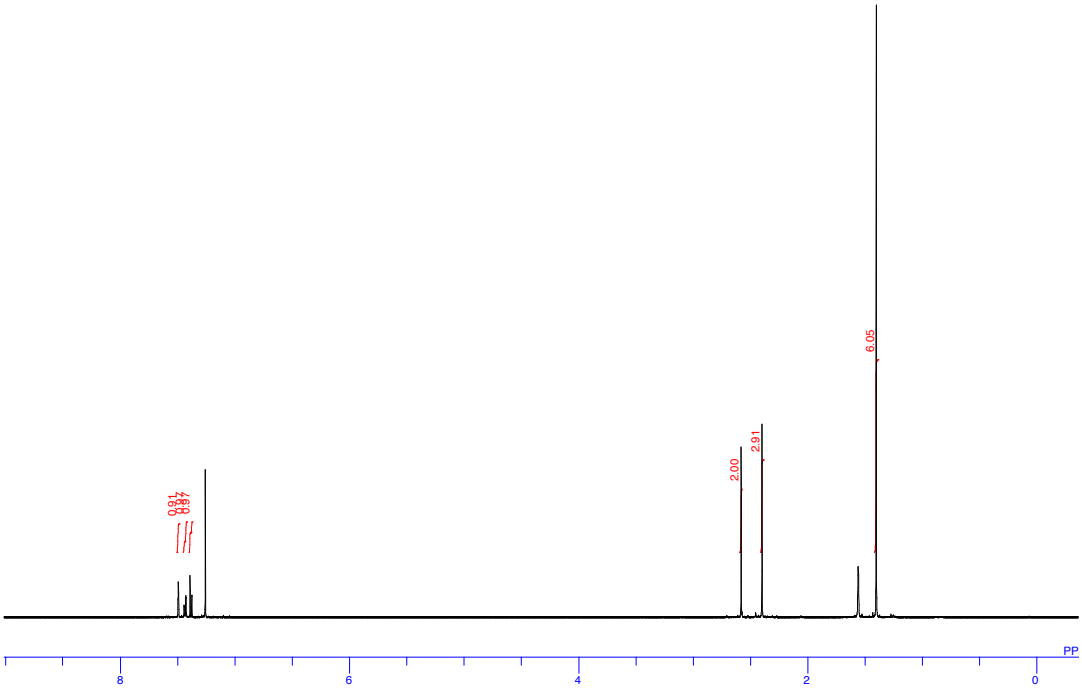
**8a**



**8b**


Me
Me
Me
O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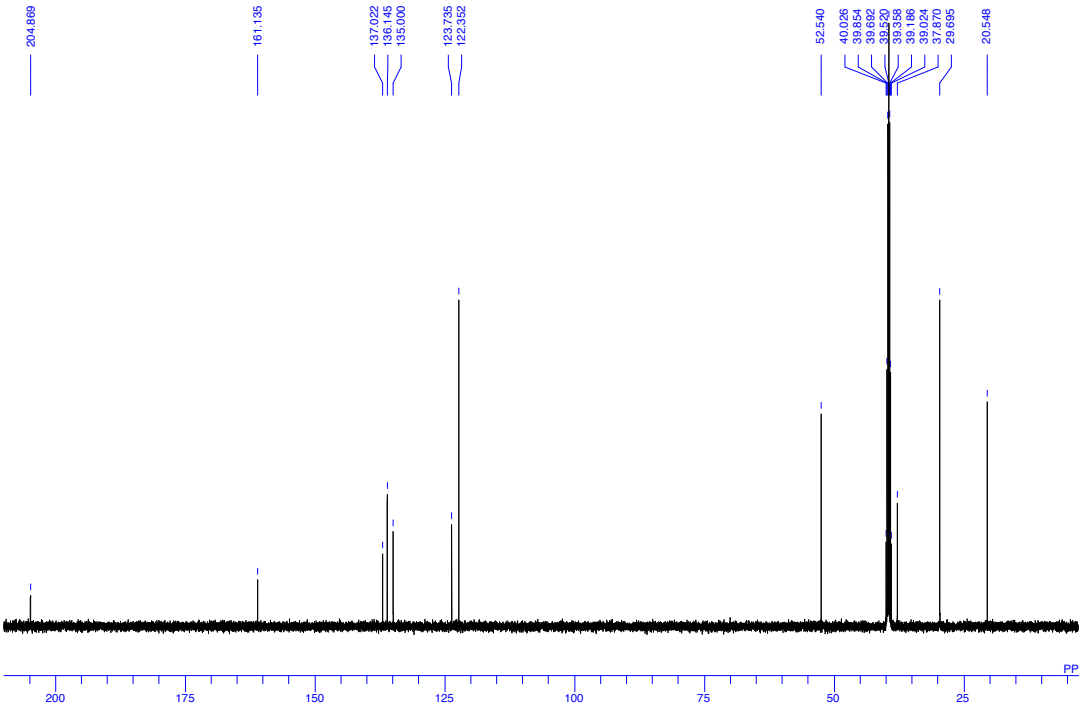
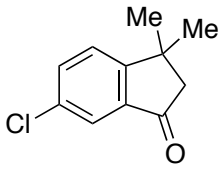
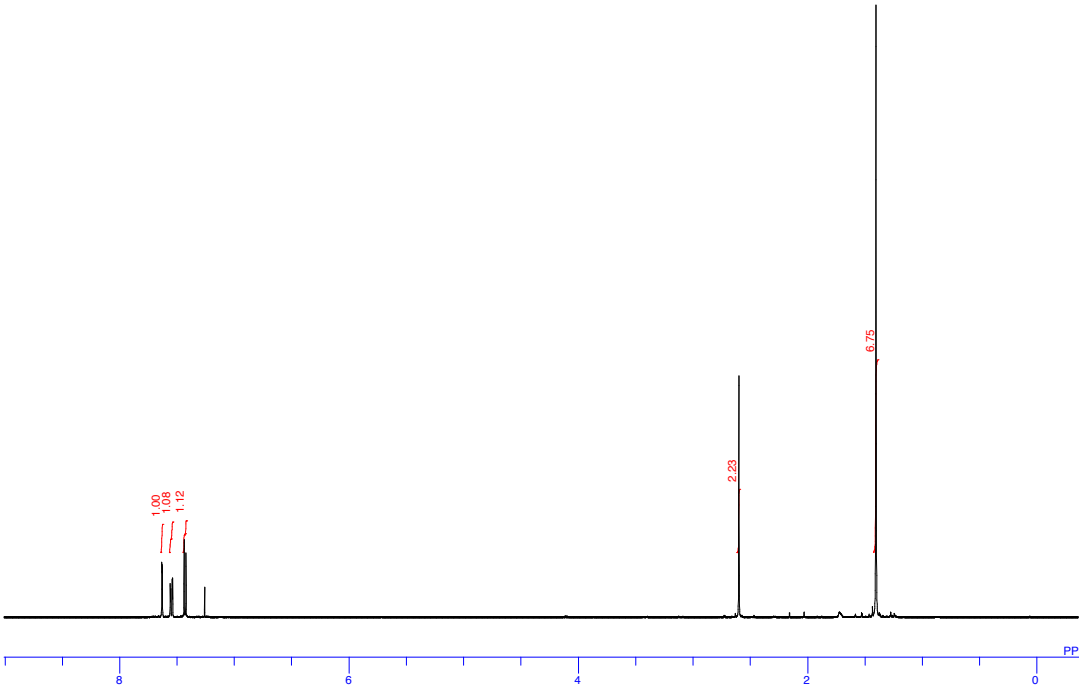
0.94
0.97
2.00
2.91
6.05
PP
8
6
4
2
0


204.869
161.135
137.022
136.145
135.000
123.735
122.352
52.540
40.026
39.854
39.682
39.530
39.358
39.186
39.024
37.870
29.695
20.548
PP
200
175
150
125
100
75
50
25

**8c**


Me
Me
Cl
O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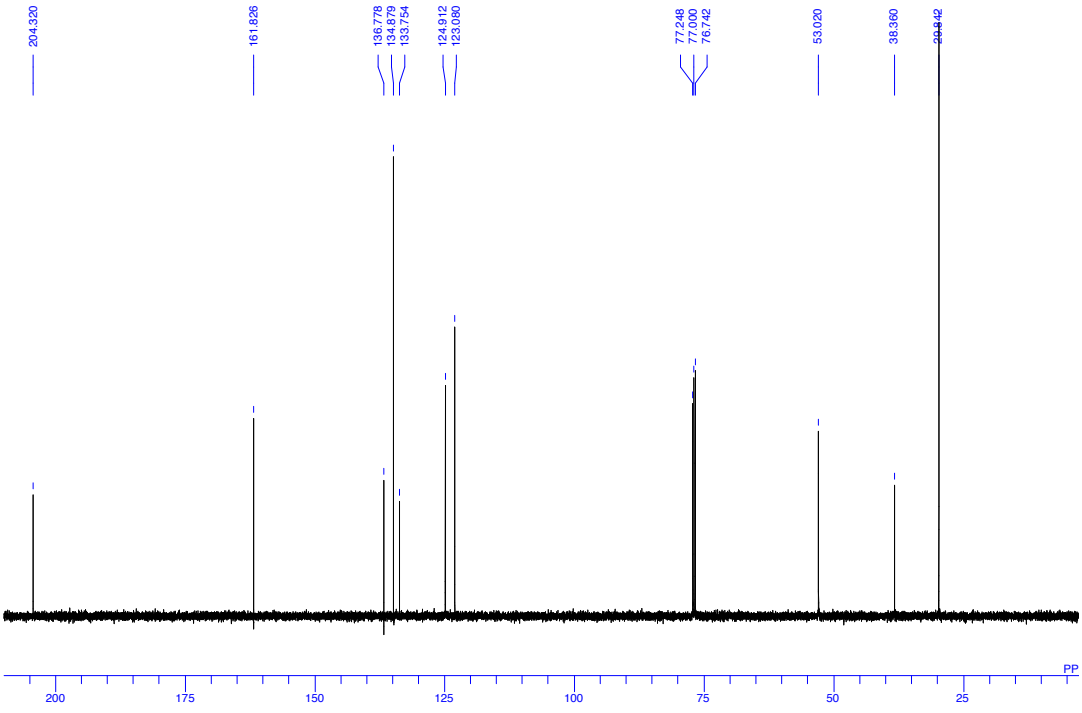
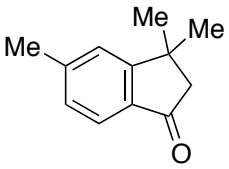
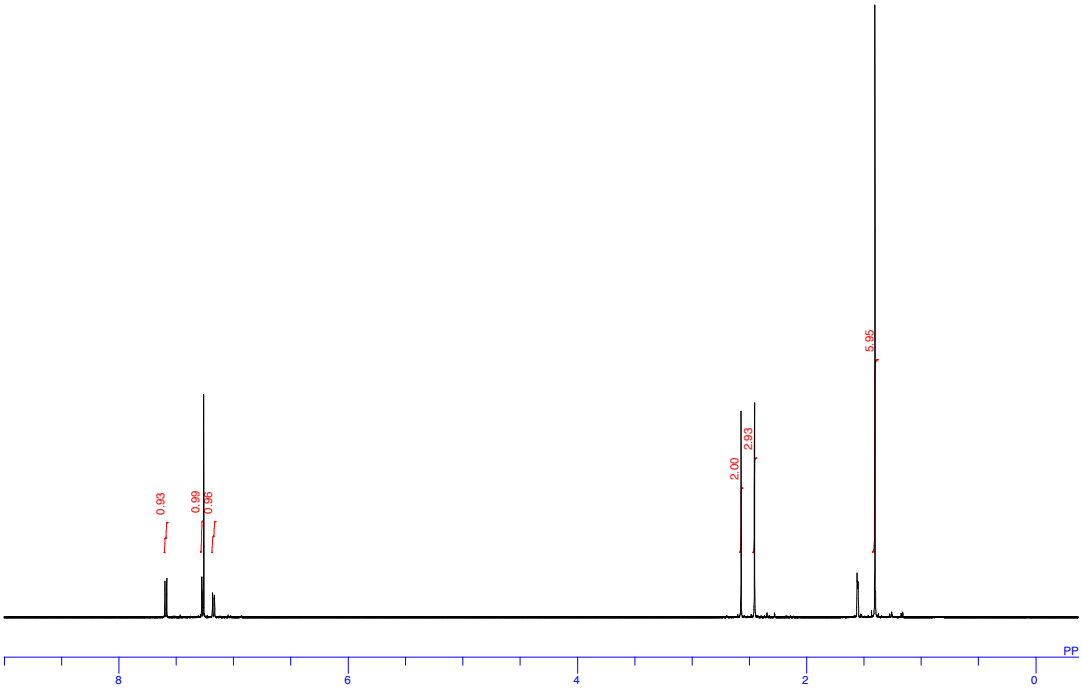
1.08
1.12
2.23
6.75
PP
8
6
4
2
0


204.320
161.826
136.778
134.879
133.754
124.912
123.080
77.248
77.000
76.742
53.020
38.360
PP
200
175
150
125
100
75
50
25

**8d**


Me
Me
Me
O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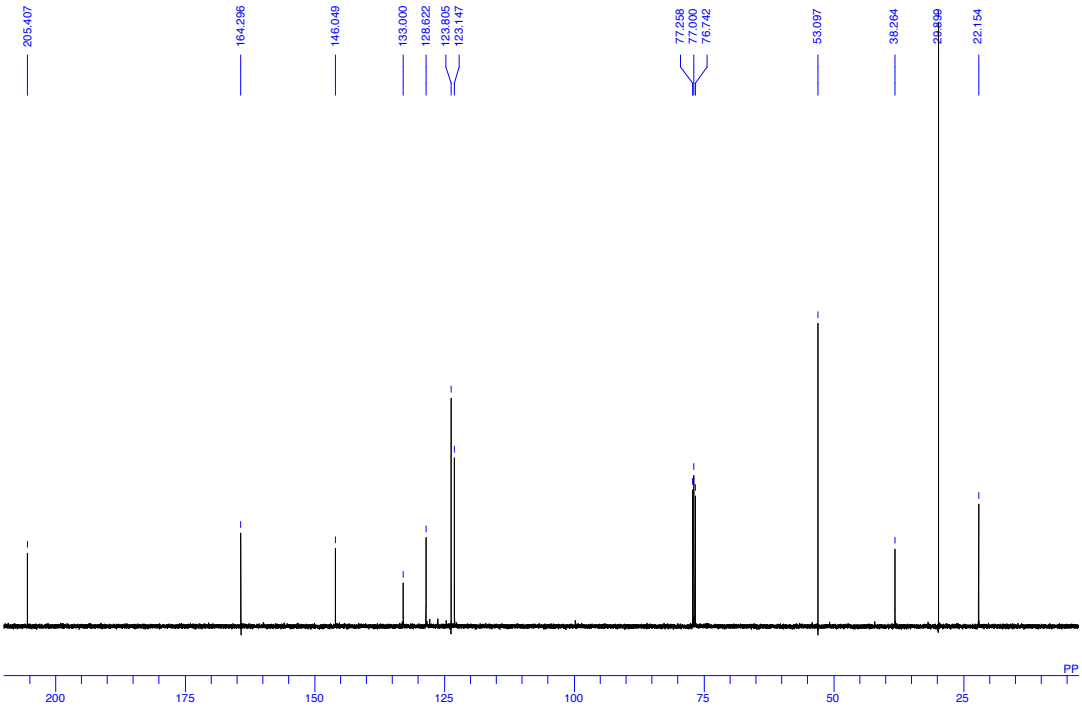
0.99
0.96
2.00
2.93
5.95
PP
8
6
4
2
0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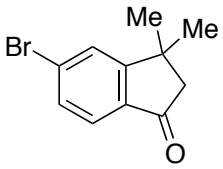
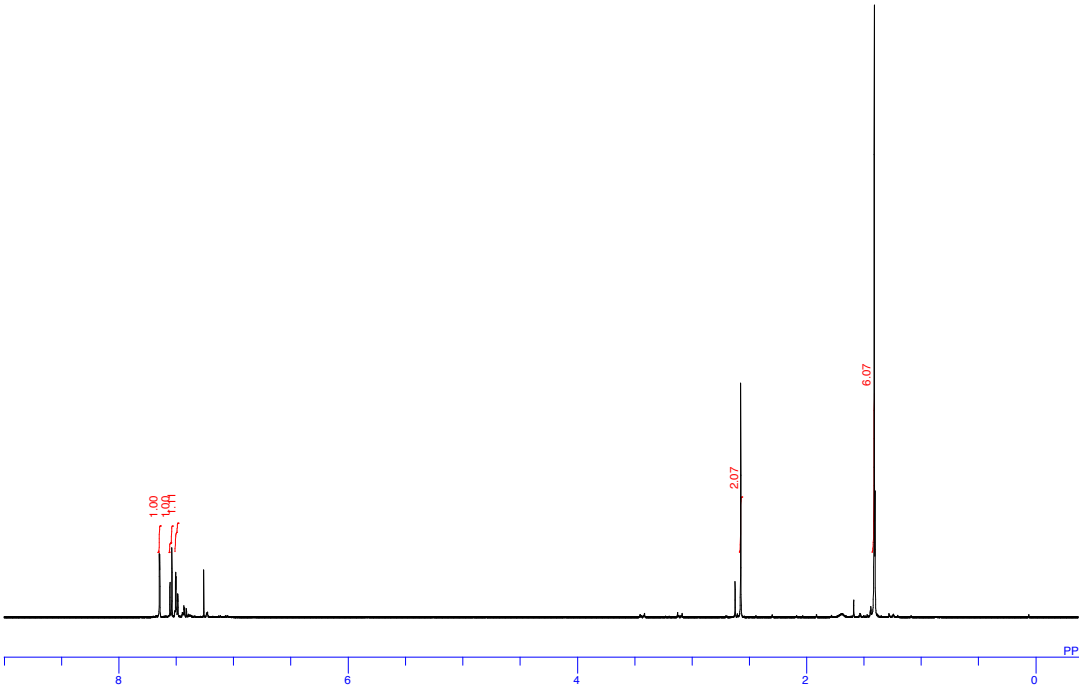
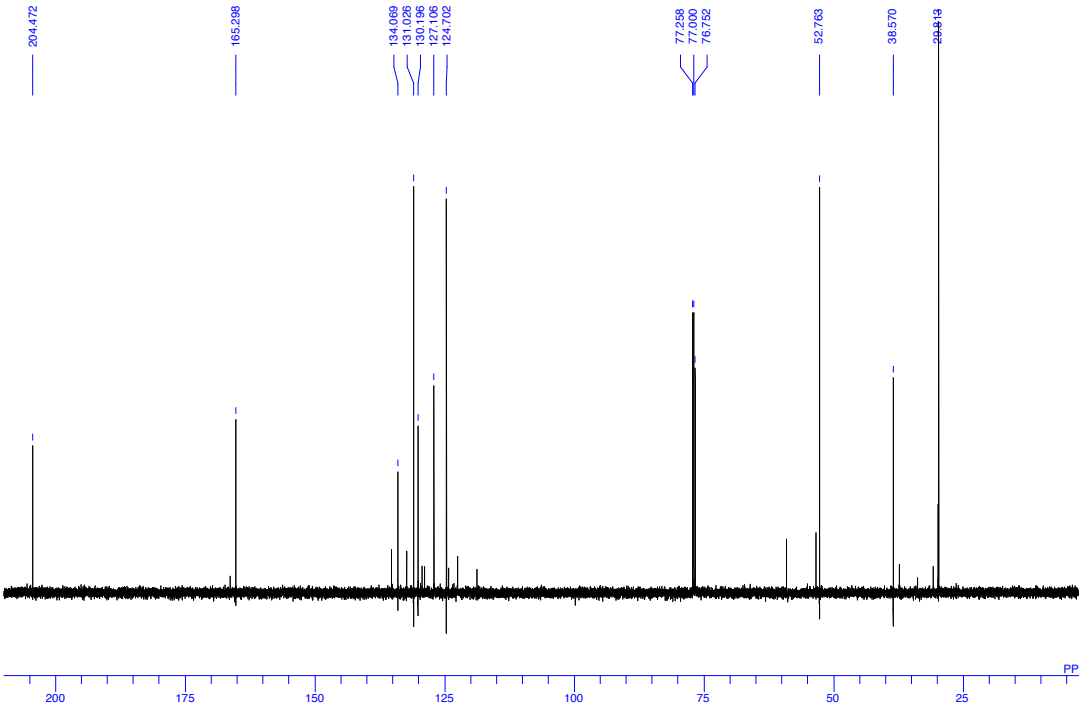
164.296
146.049
133.000
128.622
123.805
123.147
77.258
77.000
76.742
53.097
38.264
29.839
22.154
PP
200
175
150
125
100
75
50
25

**8e**


Me
Me
Br
O


1.00
1.01
2.07
6.07
PP
8
6
4
2
0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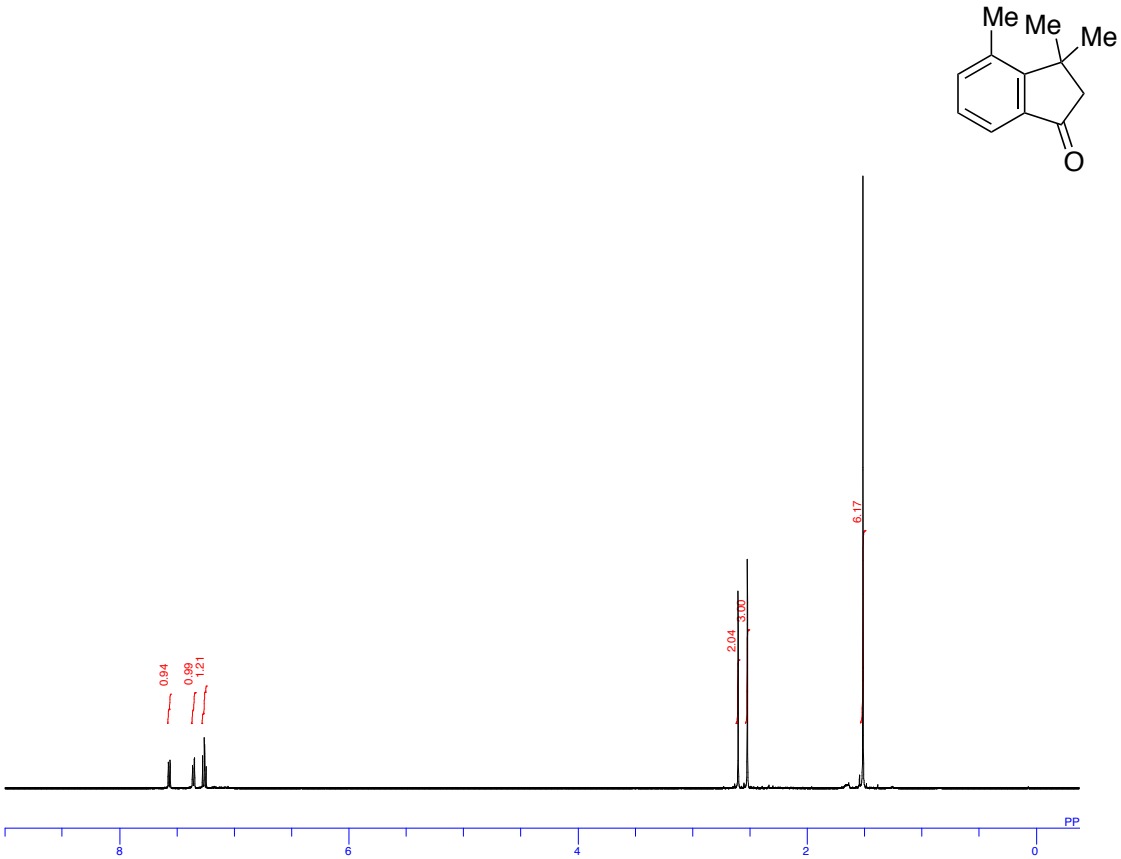
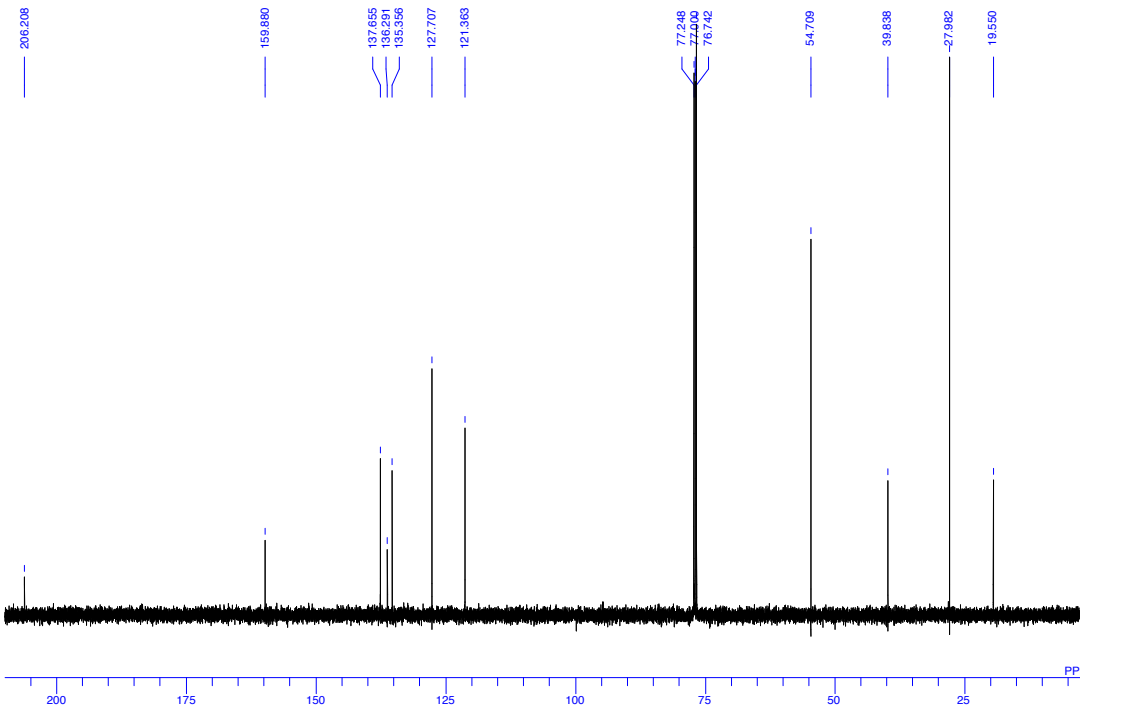
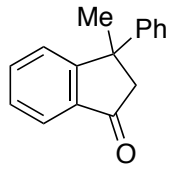
165.298
134.069
131.026
130.196
127.106
124.702
77.258
77.000
76.752
52.763
38.570
29.413
PP
200
175
150
125
100
75
50
25

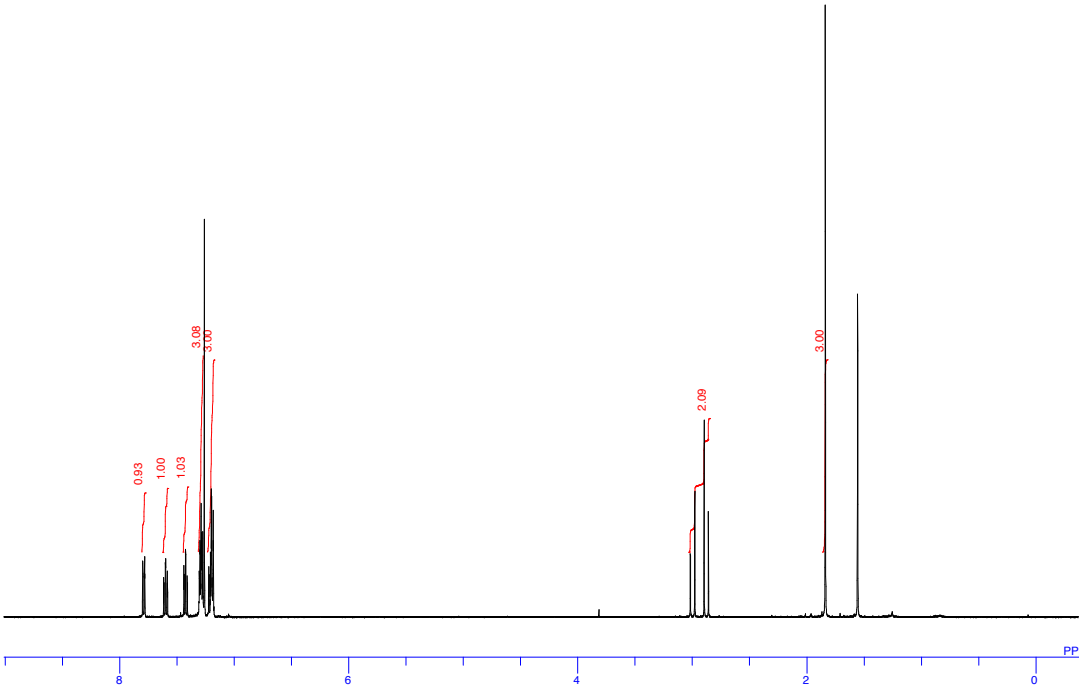
**8f**

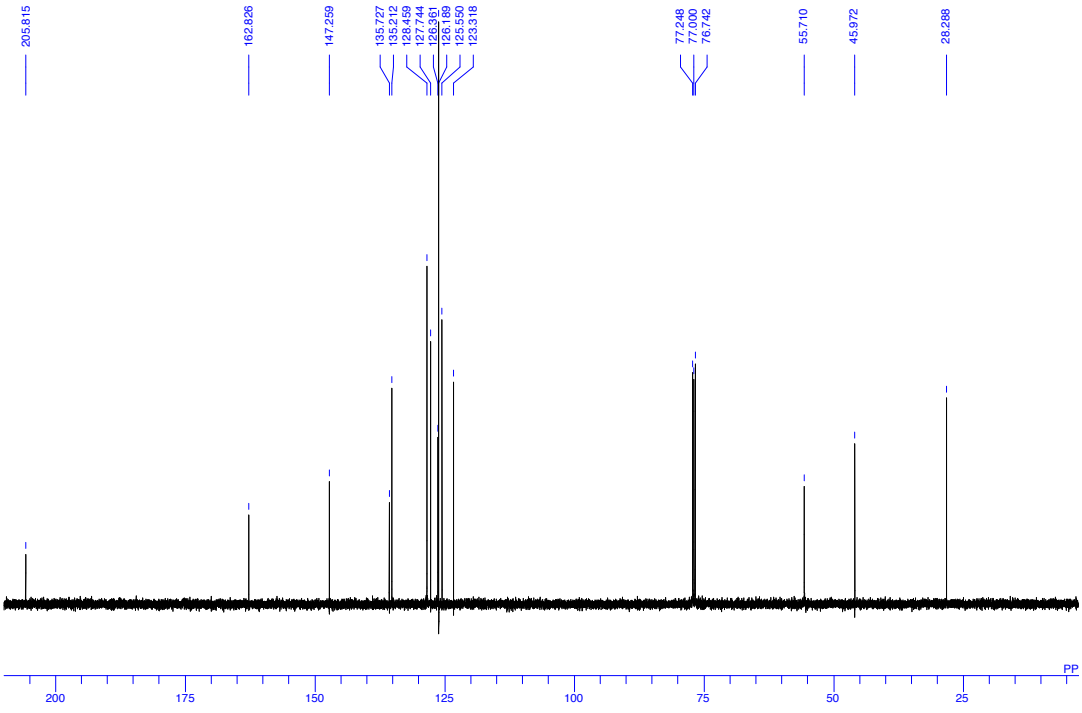

Me
Me
Me
O
0.94
0.99
1.21
2.04
3.00
6.17
PP
8
6
4
2
0


206.208
159.880
137.655
136.291
135.356
127.707
121.363
77.248
77.000
76.742
54.709
39.838
27.982
19.550
PP
200
175
150
125
100
75
50
25

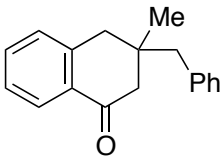
**8g**

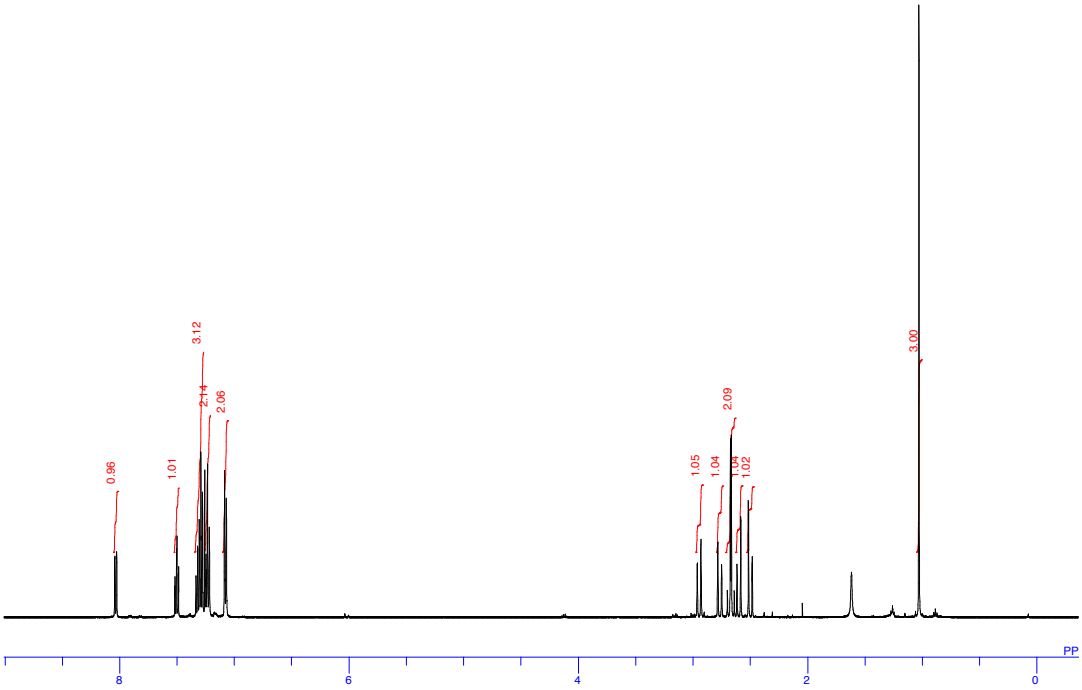

Me
Ph
O

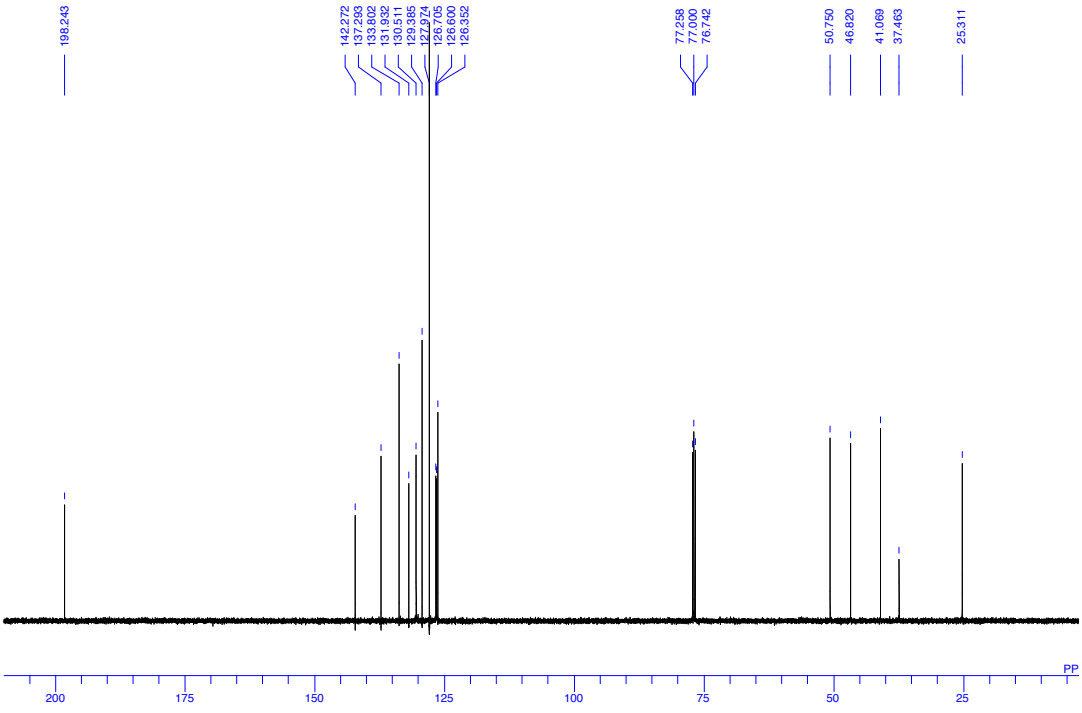

0.93
1.00
1.03
3.08
3.00
2.09
3.00
PP
8
6
4
2
0


205.815
162.826
147.259
135.727
135.212
128.459
127.744
126.361
126.189
125.550
123.318
77.248
77.000
76.742
55.710
45.972
28.288
PP
200
175
150
125
100
75
50
25

**8h**


Me
Ph
O


0.96
1.01
3.12
2.14
2.06
1.05
1.04
1.04
1.02
2.09
3.00
PP
8
6
4
2
0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142.272
137.293
133.802
131.932
130.511
129.385
127.974
126.705
126.600
126.352
77.258
77.000
76.742
50.750
46.820
41.069
37.463
25.311
PP
200
175
150
125
100
75
50
25