Supporting Information

One Pot Tandem Dual C=C and C=O Bond Reductions in β-Alkylation of Secondary Alcohols with Primary Alcohols by Ruthenium Complexes of Amido and Picolyl Functionalized N-Heterocyclic Carbenes

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Synthesis of 1-mesityl imidazole[1]

To a mixture of mesitylamine (4.46 g, 33.0 mmol), 38 % aq. glyoxal (10.1 g, 66.0 mmol) and 35 % aq. formaldehyde (5.66 g, 66.0 mmol) in MeOH (*ca.* 60 mL) $CH_3CO_2NH_4$ (5.08 g, 66.0 mmol) was added. The reaction mixture was refluxed for 12 hours during which the initially formed yellow solid becomes black colored solution. The reaction mixture was then cooled to room temperature and the volatiles were evaporated under reduced pressure. The residue thus obtained was dissolved in ethyl acetate and washed with 10 % aq. NaOH solution followed by brine solution. The combined organic layers were dried over anhydrous Na_2SO_4, filtered, and concentrated. The crude mass so obtained was finally purified by silica gel column chromatography using ethyl acetate/petroleum ether (1:4 *v*/*v*) mixed medium to give the product as a light brown solid (3.46 g, 56 %). ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ ppm, 7.43 (s, 1H, NC*H*N), 7.23 (s, 1H, NC*H*CHN), 6.97 (s, 2H, 2,4,6-$(CH_3)_3C_6H_2$), 6.89 (s, 1H, NCHC*H*N), 2.34 (s, 3H, 2,4,6-$(CH_3)_3C_6H_2$), 1.98 (s, 6H, 2,4,6-$(CH_3)_3C_6H_2$). ^{13}C{^{1}H} NMR ($CDCl_3$, 125 MHz, 25 °C): δ ppm, 139.0 (N*C*HN), 137.6 (*ipso*-2,4,6-$(CH_3)_3C_6H_2$), 135.5 (2,4,6-$(CH_3)_3C_6H_2$), 133.5 (2,4,6-$(CH_3)_3C_6H_2$), 129.6 (N*C*HCHN), 129.1 (2,4,6-$(CH_3)_3C_6H_2$), 120.2 (NCH*C*HN), 21.1 (2,4,6-$(CH_3)_3C_6H_2$), 17.4 (2,4,6-$(CH_3)_3C_6H_2$).

Synthesis of 2-chloro-*N*-(2,6-Me$_2$-phenyl)acetamide[2]

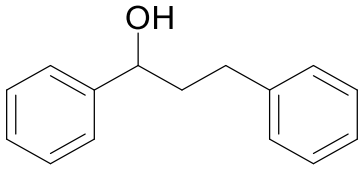
To a solution of 2,6-dimethylaniline (2.00 g, 16.5 mmol) in $CHCl_3$ (*ca.* 25 mL), 2-chloroacetyl chloride (3.73 g, 33.0 mmol) was added drop wise at 0 °C and the reaction mixture was stirred at room temperature for 8 hours. After the completion of reaction, the reaction mixture was washed with saturated $NaHCO_3$ solution (*ca.* 3 × 50 mL). The organic layer was dried over anhydrous Na_2SO_4, filtered, and concentrated. The residue so obtained was vacuum dried to give the product as a white solid (2.75 g, 84 %). ^{1}H NMR ($CDCl_3$, 500 MHz, 25 °C): δ ppm,

7.86 (br, 1H, N*H*), 7.16-7.13 (m, 1H, 2,6-$(CH_3)_2C_6H_3$), 7.11-7.09 (m, 2H, 2,6-$(CH_3)_2C_6H_3$), 4.25 (s, 2H, CH_2), 2.24 (s, 6H, 2,6-$(CH_3)_2C_6H_3$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ ppm, 164.5 (*C*O), 135.5 (*o*-2,6-$(CH_3)_2C_6H_3$), 132.8 (*ipso*-2,6-$(CH_3)_2C_6H_3$), 128.5 (*m*-2,6-$(CH_3)_2C_6H_3$), 128.1 (*p*-2,6-$(CH_3)_2C_6H_3$), 42.9 (CH_2), 18.5 (2,6-$(CH_3)_2C_6H_3$).

Synthesis of 1-(2,6-Me$_2$-phenyl)imidazole[3]

To a mixture of 2,6-dimethylaniline (4.00 g, 33.0 mmol), 38 % aq. glyoxal (10.1 g, 66.0 mmol) and 35 % aq. formaldehyde (5.66 g, 66.0 mmol) in MeOH (*ca.* 60 mL) $CH_3CO_2NH_4$ (5.08 g, 66.0 mmol) was added. The reaction mixture was refluxed for 12 hours during which the initially formed yellow solid becomes black colored solution. The reaction mixture was then cooled to room temperature and the volatiles were evaporated under reduced pressure. The residue thus obtained was dissolved in ethyl acetate and washed with 10 % aq. NaOH solution followed by brine solution. The combined organic layers were dried over anhydrous Na_2SO_4, filtered, and concentrated. The crude mass so obtained was finally purified by silica gel column chromatography using ethyl acetate/petroleum ether (1:4 *v*/*v*) mixed medium to give the product as a light brown solid (1.96 g, 34 %). ^1H NMR ($CDCl_3$, 500 MHz, 25 °C): δ ppm, 7.46 (s, 1H, NC*H*N), 7.27-7.24 (m, 1H, 2,6-$(CH_3)_2C_6H_3$), 7.25 (t, 1H, $^3J_{HH}$ = 1 Hz, NC*H*CHN), 7.17-7.15 (m, 2H, 2,6-$(CH_3)_2C_6H_3$), 6.92 (t, 1H, $^3J_{HH}$ = 1 Hz, NCHC*H*N), 2.04 (s, 6H, 2,6-$(CH_3)_2C_6H_3$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ ppm, 137.4 (N*C*HN), 136.1 (*ipso*-2,6-$(CH_3)_2C_6H_3$), 135.9 (2,6-$(CH_3)_2C_6H_3$), 129.8 (2,6-$(CH_3)_2C_6H_3$), 129.1 (N*C*HCHN), 128.5 (2,6-$(CH_3)_2C_6H_3$), 120.0 (NCH*C*HN), 17.6 (2,6-$(CH_3)_2C_6H_3$).

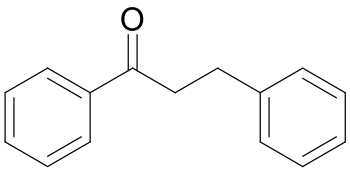
1,3-Diphenylpropan-1-ol (3)[4]



(**3**)

Colorless liquid [0.153 g, 72 % isolated yield (**1c**); 0.144 g, 68 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.37−7.36 (m, 4H, $C_6\underline{H}_5CH(OH)CH_2CH_2C_6\underline{H}_5$), 7.32−7.28 (m, 3H, $C_6\underline{H}_5CH(OH)CH_2CH_2C_6\underline{H}_5$), 7.22−7.19 (m, 3H, $C_6\underline{H}_5CH(OH)CH_2CH_2C_6\underline{H}_5$), 4.71−4.68 (m, 1H, $C_6H_5C\underline{H}(OH)CH_2CH_2C_6H_5$), 2.81−2.65 (m, 2H, $C_6H_5CH(OH)C\underline{H}_2CH_2C_6H_5$), 2.20−2.00 (m, 2H, $C_6H_5CH(OH)CH_2C\underline{H}_2C_6H_5$), 1.88 (b, 1H, $C_6H_5CH(O\underline{H})CH_2CH_2C_6H_5$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 100 MHz, 25 °C): δ 144.7 (*ipso*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_6H_5$), 141.9 ($C_6H_5CH(OH)CH_2CH_2$-*ipso*-$\underline{C}_6H_5$), 128.7 (*o*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_6H_5$), 128.6 ($C_6H_5CH(OH)CH_2CH_2$-*o*-$\underline{C}_6H_5$), 128.5 (*m*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_6H_5$), 127.8 (*p*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_6H_5$), 126.1 ($C_6H_5CH(OH)CH_2CH_2$-*m*-$\underline{C}_6H_5$), 126.0 ($C_6H_5CH(OH)CH_2CH_2$-*p*-$\underline{C}_6H_5$), 74.0 ($C_6H_5\underline{C}H(OH)CH_2CH_2C_6H_5$), 40.6 ($C_6H_5CH(OH)\underline{C}H_2CH_2C_6H_5$), 32.2 ($C_6H_5CH(OH)CH_2\underline{C}H_2C_6H_5$). GCMS (ESI): $[M]^+$ *m*/*z* = 212. Anal. Calcd. for $C_{15}H_{16}O$: C, 84.87; H, 7.60; Found: C, 84.17; H, 7.43.

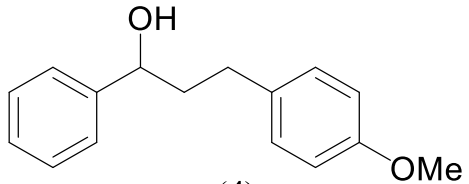
1,3-Diphenylpropan-1-one (3')[4]



(**3'**)

Colorless liquid [0.023 g, 11 % isolated yield (**1c**); 0.027 g, 13 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 500 MHz, 25 °C): δ 8.01 (dd, 2H, $^{3}J_{H\text{-}H}$ = 8 Hz, $^{4}J_{H\text{-}H}$ = 1 Hz, $C_6\underline{H}_5COCH_2CH_2C_6H_5$), 7.60 (dt, 1H, $^{3}J_{H\text{-}H}$ = 8 Hz, $^{4}J_{H\text{-}H}$ = 1 Hz, $C_6\underline{H}_5COCH_2CH_2C_6H_5$), 7.50 (t, 2H, $^{3}J_{H\text{-}H}$ = 8 Hz, $C_6\underline{H}_5COCH_2CH_2C_6H_5$), 7.37-7.34 (m, 2H, $C_6H_5COCH_2CH_2C_6\underline{H}_5$), 7.32-7.31 (m, 2H, $C_6H_5COCH_2CH_2C_6\underline{H}_5$), 7.26 (t, 1H, $^{3}J_{H\text{-}H}$ = 7 Hz, $C_6H_5COCH_2CH_2C_6\underline{H}_5$), 3.35 (t, 2H, $^{3}J_{H\text{-}H}$ = 8 Hz, $C_6H_5COC\underline{H}_2CH_2C_6H_5$), 3.13 (t, 2H, $^{3}J_{H\text{-}H}$ = 8 Hz, $C_6H_5COCH_2C\underline{H}_2C_6H_5$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ 199.3 ($C_6H_5\underline{C}OCH_2CH_2C_6H_5$), 141.4 (*ipso*-$\underline{C}_6H_5COCH_2CH_2C_6H_5$), 137.0 ($C_6H_5COCH_2CH_2$-*ipso*-$\underline{C}_6H_5$), 133.2 (*p*-$\underline{C}_6H_5COCH_2CH_2C_6H_5$), 128.7 (*o*-$\underline{C}_6H_5COCH_2CH_2C_6H_5$), 128.6 (*m*-$\underline{C}_6H_5COCH_2CH_2C_6H_5$), 128.5 ($C_6H_5COCH_2CH_2$-*m*-$\underline{C}_6H_5$), 128.1 ($C_6H_5COCH_2CH_2$-*o*-$\underline{C}_6H_5$), 126.2 ($C_6H_5COCH_2CH_2$-*p*-$\underline{C}_6H_5$), 40.5 ($C_6H_5CO\underline{C}H_2CH_2C_6H_5$), 30.2 ($C_6H_5COCH_2\underline{C}H_2C_6H_5$). GCMS (ESI): $[M]^+$ *m*/*z* = 210.

3-(4-Methoxyphenyl)-1-phenylpropan-1-ol (4)[4]

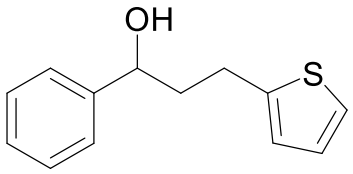


(**4**)

Colorless liquid [0.215 g, 89 % isolated yield (**1c**); 0.208 g, 86 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.36−7.35 (m, 4H, $C_6\underline{H}_5CH(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 7.31−7.28 (m, 1H, $C_6\underline{H}_5CH(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 7.12 (d, 2H, $^{3}J_{H\text{-}H}$ = 9 Hz, $C_6H_5CH(OH)CH_2CH_2$-4-OCH_3-$C_6\underline{H}_4$), 6.84 (d, 2H, $^{3}J_{H\text{-}H}$ = 8 Hz, $C_6H_5CH(OH)CH_2CH_2$-4-OCH_3-$C_6\underline{H}_4$), 4.69−4.66 (m, 1H, $C_6H_5C\underline{H}(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 3.79 (s, 3H, $C_6H_5CH(OH)CH_2CH_2$-4-$OC\underline{H}_3$-C_6H_4), 2.74−2.58 (m, 2H, $C_6H_5CH(OH)C\underline{H}_2CH_2$-4-$OCH_3$-$C_6H_4$), 2.16−1.97 (m, 2H, $C_6H_5CH(OH)CH_2C\underline{H}_2$-4-$OCH_3$-$C_6H_4$), 1.96 (b, 1H,

$C_6H_5CH(O\underline{H})CH_2CH_2$-4-$OCH_3$-$C_6H_4$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 100 MHz, 25 °C): δ 157.9 ($C_6H_5CH(OH)CH_2CH_2$-4-OCH_3-*p*-$\underline{C}_6H_4$), 144.8 (*ipso*-$\underline{C}_6H_5CH(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 134.0 ($C_6H_5CH(OH)CH_2CH_2$-4-$OCH_3$-*ipso*-$\underline{C}_6H_4$), 129.5 ($C_6H_5CH(OH)CH_2CH_2$-4-$OCH_3$-*o*-$\underline{C}_6H_4$), 128.7 (*m*-$\underline{C}_6H_5CH(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 127.8 (*p*-$\underline{C}_6H_5CH(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 126.1 (*o*-$\underline{C}_6H_5CH(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 114.0 ($C_6H_5CH(OH)CH_2CH_2$-4-$OCH_3$-*o*-$\underline{C}_6H_4$), 74.0 ($C_6H_5\underline{C}H(OH)CH_2CH_2$-4-$OCH_3$-$C_6H_4$), 55.4 ($C_6H_5CH(OH)CH_2CH_2$-4-O$\underline{C}H_3$-$C_6H_4$), 40.8 ($C_6H_5CH(OH)\underline{C}H_2CH_2$-4-$OCH_3$-$C_6H_4$), 31.3 ($C_6H_5CH(OH)CH_2\underline{C}H_2$-4-$OCH_3$-$C_6H_4$). GCMS (ESI): $[M]^+$ *m*/*z* = 242. Anal. Calcd. for $C_{16}H_{18}O_2$: C, 79.31; H, 7.49; Found: C, 78.97; H, 7.02.

1-Phenyl-3-(thiophen-2-yl)-propan-1-ol (5)[4]

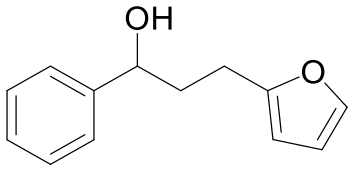


(**5**)

Colorless liquid [0.161 g, 74 % isolated yield (**1c**); 0.137 g, 63 % isolated yield (**2c**)] ^1H NMR ($CDCl_3$, 500 MHz, 25 °C): δ 7.40−7.39 (m, 4H, $C_6\underline{H}_5CH(OH)CH_2CH_2C_4H_3S$), 7.34−7.31 (m, 1H, $C_6\underline{H}_5CH(OH)CH_2CH_2C_4H_3S$), 7.15 (dd, 1H, $^3J_{H\text{-}H}$ = 5 Hz, $^4J_{H\text{-}H}$ = 1 Hz, $C_6H_5CH(OH)CH_2CH_2C_4\underline{H}_3S$), 6.95 (dd, 1H, $^3J_{H\text{-}H}$ = 5 Hz, $^4J_{H\text{-}H}$ = 2 Hz, $C_6H_5CH(OH)CH_2CH_2C_4\underline{H}_3S$), 6.84-6.83 (m, 1H, $C_6H_5CH(OH)CH_2CH_2C_4\underline{H}_3S$), 4.78−4.76 (m, 1H, $C_6H_5C\underline{H}(OH)CH_2CH_2C_4H_3S$), 2.98−2.93 (m, 2H, $C_6H_5CH(OH)C\underline{H}_2CH_2C_4H_3S$), 2.26−2.18 (m, 1H, $C_6H_5CH(OH)CH_2C\underline{H}_2C_4H_3S$), 2.15−2.08 (m, 1H, $C_6H_5CH(OH)CH_2C\underline{H}_2C_4H_3S$), 1.66 (b, 1H, $C_6H_5CH(O\underline{H})CH_2CH_2C_4H_3S$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ 144.8 (*ipso*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_4H_3S$), 144.5 ($C_6H_5CH(OH)CH_2CH_2$-*ipso*-$\underline{C}_4H_3S$), 128.8 (*m*-

$\underline{C}_6H_5CH(OH)CH_2CH_2C_4H_3S$), 127.9 ($C_6H_5CH(OH)CH_2CH_2\underline{C}_4H_3S$), 126.9 ($C_6H_5CH(OH)CH_2CH_2\underline{C}_4H_3S$), 126.1 (*o*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_4H_3S$), 124.5 (*p*-$C_6H_5CH(OH)CH_2CH_2C_4H_3S$), 123.3 ($C_6H_5CH(OH)CH_2CH_2\underline{C}_4H_3S$), 73.7 ($C_6H_5\underline{C}H(OH)CH_2CH_2C_4H_3S$), 40.9 ($C_6H_5CH(OH)\underline{C}H_2CH_2C_4H_3S$), 26.4 ($C_6H_5CH(OH)CH_2\underline{C}H_2C_4H_3S$). GCMS (ESI): $[M]^+$ m/z = 218. Anal. Calcd. for $C_{13}H_{14}OS$: C, 71.52; H, 6.46; Found: C, 69.81; H, 8.02.

3-(Furan-2-yl)-1-phenylpropan-1-ol (6)[4]

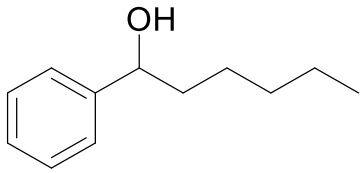


(**6**)

Colorless liquid [0.138 g, 68 % isolated yield (**1c**); 0.141 g, 70 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 500 MHz, 25 °C): δ 7.30−7.29 (m, 4H, $C_6\underline{H}_5CH(OH)CH_2CH_2C_4H_3O$), 7.24−7.23 (m, 1H, $C_6\underline{H}_5CH(OH)CH_2CH_2C_4H_3O$), 7.23−7.21 (m, 1H, $C_6H_5CH(OH)CH_2CH_2C_4\underline{H}_3O$), 6.21 (dd, 1H, $^3J_{H\text{-}H}$ = 4 Hz, $^4J_{H\text{-}H}$ = 2 Hz, $C_6H_5CH(OH)CH_2CH_2C_4\underline{H}_3O$), 5.94−5.93 (m, 1H, $C_6H_5CH(OH)CH_2CH_2C_4\underline{H}_3O$), 4.66−4.64 (m, 1H, $C_6H_5C\underline{H}(OH)CH_2CH_2C_4H_3O$), 2.70−2.63 (m, 2H, $C_6H_5CH(OH)C\underline{H}_2CH_2C_4H_3O$), 2.10−1.96 (m, 2H, $C_6H_5CH(OH)CH_2C\underline{H}_2C_4H_3O$), 1.82 (b, 1H, $C_6H_5CH(O\underline{H})CH_2CH_2C_4H_3O$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ 155.7 ($C_6H_5CH(OH)CH_2CH_2$-*ipso*-$\underline{C}_4H_3O$), 144.5 (*ipso*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_4H_3O$), 141.1 ($C_6H_5CH(OH)CH_2CH_2\underline{C}_4H_3O$), 128.7 (*m*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_4H_3O$), 127.9 (*p*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_4H_3O$), 126.1 (*o*-$\underline{C}_6H_5CH(OH)CH_2CH_2C_4H_3O$), 110.3 ($C_6H_5CH(OH)CH_2CH_2\underline{C}_4H_3O$), 105.2 ($C_6H_5CH(OH)CH_2CH_2\underline{C}_4H_3O$), 73.9 ($C_6H_5\underline{C}H(OH)CH_2CH_2C_4H_3O$), 37.3 ($C_6H_5CH(OH)\underline{C}H_2CH_2C_4H_3O$), 24.6

($C_6H_5CH(OH)CH_2\underline{C}H_2C_4H_3O$). GCMS (ESI): $[M]^+$ *m*/*z* = 202. Anal. Calcd. for $C_{13}H_{14}O_2$: C, 77.20; H, 6.98; Found: C, 77.71; H, 7.35.

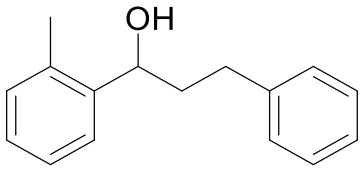
1-Phenylhexan-1-ol (7)[4]



(**7**)

Colorless liquid [0.126 g, 71 % isolated yield (**1c**); 0.123 g, 69 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.27−7.26 (m, 4H, $C_6\underline{H}_5CH(OH)CH_2CH_2CH_2CH_2CH_3$), 7.21−7.18 (m, 1H, $C_6\underline{H}_5CH(OH)CH_2CH_2CH_2CH_2CH_3$), 4.59−4.55 (m, 1H, $C_6H_5C\underline{H}(OH)CH_2CH_2CH_2CH_2CH_3$), 1.85 (b, 1H, $C_6H_5CH(O\underline{H})CH_2CH_2CH_2CH_2CH_3$), 1.76−1.57 (m, 2H, $C_6H_5CH(OH)C\underline{H}_2CH_2CH_2CH_2CH_3$), 1.35−1.18 (m, 6H, $C_6H_5CH(OH)CH_2C\underline{H}_2C\underline{H}_2C\underline{H}_2CH_3$) 0.81-0.78 (m, 3H, $C_6H_5CH(OH)CH_2CH_2CH_2CH_2C\underline{H}_3$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 100 MHz, 25 °C): δ 145.1 (*ipso*-$\underline{C}_6H_5CH(OH)CH_2CH_2CH_2CH_2CH_3$), 128.6 (*m*-$\underline{C}_6H_5CH(OH)CH_2CH_2CH_2CH_2CH_3$), 127.6 (*p*-$\underline{C}_6H_5CH(OH)CH_2CH_2CH_2CH_2CH_3$), 126.1 (*o*-$\underline{C}_6H_5CH(OH)CH_2CH_2CH_2CH_2CH_3$), 74.9 ($C_6H_5\underline{C}H(OH)CH_2CH_2CH_2CH_2CH_3$), 39.2 ($C_6H_5CH(OH)\underline{C}H_2CH_2CH_2CH_2CH_3$), 31.9 ($C_6H_5CH(OH)CH_2\underline{C}H_2CH_2CH_2CH_3$), 25.7 ($C_6H_5CH(OH)CH_2CH_2\underline{C}H_2CH_2CH_3$), 22.7 ($C_6H_5CH(OH)CH_2CH_2CH_2\underline{C}H_2CH_3$), 14.2 ($C_6H_5CH(OH)CH_2CH_2CH_2CH_2\underline{C}H_3$). GCMS (ESI): $[M]^+$ *m*/*z* = 178. Anal. Calcd. for $C_{12}H_{18}O$: C, 80.85; H, 10.18; Found: C, 80.05; H, 10.07.

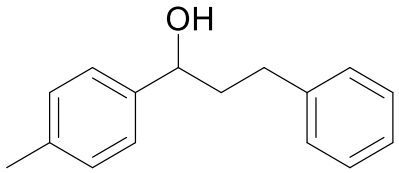
3-Phenyl-1-(*o*-tolyl)propan-1-ol (8)[4]



(**8**)

Colorless liquid [0.164 g, 73 % isolated yield (**1c**); 0.176 g, 78 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 500 MHz, 25 °C): δ 7.53 (dd, 1H, $^{3}J_{HH}$ = 8 Hz, $^{4}J_{HH}$ = 1 Hz, 2-CH_3-$C_6\underline{H}_4CH(OH)CH_2CH_2C_6H_5$), 7.33−7.30 (m, 2H, 2-$CH_3$-$C_6H_4CH(OH)CH_2CH_2C_6\underline{H}_5$), 7.28−7.14 (m, 6H, 2-$CH_3$-$C_6\underline{H}_4CH(OH)CH_2CH_2C_6\underline{H}_5$), 4.97−4.95 (m, 1H, 2-$CH_3$-$C_6H_4C\underline{H}(OH)CH_2CH_2C_6H_5$), 2.91−2.85 (m, 1H, 2-$CH_3$-$C_6H_4CH(OH)C\underline{H}_2CH_2C_6H_5$), 2.80−2.74 (m, 1H, 2-$CH_3$-$C_6H_4CH(OH)C\underline{H}_2CH_2C_6H_5$), 2.27 (s, 3H, 2-$C\underline{H}_3$-$C_6H_4CH(OH)CH_2CH_2C_6H_5$), 2.14−1.99 (m, 2H, 2-$CH_3$-$C_6H_4CH(OH)CH_2C\underline{H}_2C_6H_5$), 1.68 (b, 1H, 2-$CH_3$-$C_6H_4CH(O\underline{H})CH_2CH_2C_6H_5$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ 142.9 (2-CH_3-$C_6H_4CH(OH)CH_2CH_2$-*ipso*-$\underline{C}_6H_5$), 142.0 (2-CH_3-*ipso*-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 134.6 (2-$CH_3$-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 130.6 (2-$CH_3$-$C_6H_4CH(OH)CH_2CH_2$-*p*-$\underline{C}_6H_5$), 128.6 (2-$CH_3$-$C_6H_4CH(OH)CH_2CH_2$-*m*-$\underline{C}_6H_5$), 128.5 (2-$CH_3$-$C_6H_4CH(OH)CH_2CH_2$-*o*-$\underline{C}_6H_5$), 127.4 (2-$CH_3$-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 126.5 (2-$CH_3$-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 126.1 (2-$CH_3$-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 125.3 (2-$CH_3$-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 70.1 (2-$CH_3$-$C_6H_4\underline{C}H(OH)CH_2CH_2C_6H_5$), 39.6 (2-$CH_3$-$C_6H_4CH(OH)\underline{C}H_2CH_2C_6H_5$), 32.5 (2-$CH_3$-$C_6H_4CH(OH)CH_2\underline{C}H_2C_6H_5$), 19.1 (2-$\underline{C}H_3$-$C_6H_4CH(OH)CH_2CH_2C_6H_5$). GCMS (ESI): $[M]^+$ *m*/*z* = 226. Anal. Calcd. for $C_{16}H_{18}O$: C, 84.91; H, 8.02; Found: C, 84.72; H, 8.21.

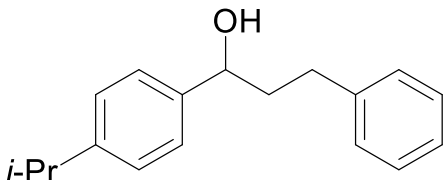
3-Phenyl-1-(*p*-tolyl)propan-1-ol (9)[4]



(**9**)

Colorless liquid [0.168 g, 74 % isolated yield (**1c**); 0.163 g, 72 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 500 MHz, 25 °C): δ 7.32-7.27 (m, 4H, 4-CH_3-$C_6\underline{H}_4$CH(OH)$CH_2CH_2C_6H_5$), 7.23-7.19 (m, 5H, 4-CH_3-C_6H_4CH(OH)$CH_2CH_2C_6\underline{H}_5$), 4.70−4.67 (m, 1H, 4-$CH_3$-$C_6H_4C\underline{H}(OH)CH_2CH_2C_6H_5$), 2.80−2.66 (m, 2H, 4-$CH_3$-$C_6H_4$CH(OH)$C\underline{H}_2CH_2C_6H_5$), 2.38 (s, 3H, 4-$C\underline{H}_3$-$C_6H_4$CH(OH)$CH_2CH_2C_6H_5$), 2.20−2.13 (m, 1H, 4-$CH_3$-$C_6H_4$CH(OH)$CH_2C\underline{H}_2C_6H_5$), 2.09−2.01 (m, 1H, 4-$CH_3$-$C_6H_4$CH(OH)$CH_2C\underline{H}_2C_6H_5$), 1.63 (b, 1H, 4-$CH_3$-$C_6H_4$CH(O$\underline{H}$)$CH_2CH_2C_6H_5$). ^{13}C{^{1}H} NMR ($CDCl_3$, 100 MHz, 25 °C): δ 142.0 (4-CH_3-C_6H_4CH(OH)CH_2CH_2-*ipso*-$\underline{C}_6H_5$), 141.8 (4-CH_3-*ipso*-$\underline{C}_6H_4$CH(OH)$CH_2CH_2C_6H_5$), 137.5 (4-CH_3-$\underline{C}_6H_4$CH(OH)$CH_2CH_2C_6H_5$), 129.4 (4-CH_3-$\underline{C}_6H_4$CH(OH)$CH_2CH_2C_6H_5$), 128.6 (4-CH_3-C_6H_4CH(OH)CH_2CH_2-*m*-$\underline{C}_6H_5$), 128.5 (4-CH_3-C_6H_4CH(OH)CH_2CH_2-*o*-$\underline{C}_6H_5$), 126.1 (4-CH_3-$\underline{C}_6H_4$CH(OH)$CH_2CH_2C_6H_5$), 126.0 (4-CH_3-C_6H_4CH(OH)CH_2CH_2-*p*-$\underline{C}_6H_5$), 73.9 (4-CH_3-$C_6H_4\underline{C}$H(OH)$CH_2CH_2C_6H_5$), 40.6 (4-CH_3-C_6H_4CH(OH)$\underline{C}H_2CH_2C_6H_5$), 32.3 (4-$CH_3$-$C_6H_4$CH(OH)$CH_2\underline{C}H_2C_6H_5$), 21.3 (4-$\underline{C}H_3$-$C_6H_4$CH(OH)$CH_2CH_2C_6H_5$). GCMS (ESI): $[M]^+$ *m*/*z* = 226. Anal. Calcd. for $C_{16}H_{18}O$: C, 84.91; H, 8.02; Found: C, 84.83; H, 7.83.

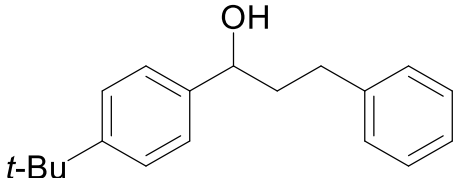
1-(4-*iso*-Propylphenyl)-3-phenylpropan-1-ol (10)[4]



(**10**)

Colorless liquid [0.188 g, 74 % isolated yield (**1c**); 0.178 g, 70 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.30-7.26 (m, 4H, 4-($CH(CH_3)_2$)-$C_6\underline{H}_4CH(OH)CH_2CH_2C_6H_5$), 7.23-7.19 (m, 5H, 4-($CH(CH_3)_2$)-$C_6H_4CH(OH)CH_2CH_2C_6\underline{H}_5$), 4.68−4.65 (m, 1H, 4-($CH(CH_3)_2$)-$C_6H_4C\underline{H}(OH)CH_2CH_2C_6H_5$), 2.92 (sept, 1H, $^{3}J_{HH}$ = 7 Hz, 4-($C\underline{H}(CH_3)_2$)-$C_6H_4CH(OH)CH_2CH_2C_6H_5$), 2.81−2.64 (m, 2H, 4-($CH(CH_3)_2$)-$C_6H_4CH(OH)C\underline{H}_2CH_2C_6H_5$), 2.18−1.99 (m, 2H, 4-($CH(CH_3)_2$)-$C_6H_4CH(OH)CH_2C\underline{H}_2C_6H_5$), 1.74 (b, 1H, 4-($CH(CH_3)_2$)-$C_6H_4CH(O\underline{H})CH_2CH_2C_6H_5$), 1.26 (d, 6H, $^{3}J_{HH}$ = 7 Hz, 4-($CH(C\underline{H}_3)_2$)-$C_6H_4CH(OH)CH_2CH_2C_6H_5$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 100 MHz, 25 °C): δ 148.6 (4-($CH(CH_3)_2$)-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 142.1 (4-($CH(CH_3)_2$)-$C_6H_4CH(OH)CH_2CH_2$-*ipso*-$\underline{C}_6H_5$), 142.0 (4-($CH(CH_3)_2$)-*ipso*-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 128.6 (4-($CH(CH_3)_2$)-$C_6H_4CH(OH)CH_2CH_2$-*m*-$\underline{C}_6H_5$), 128.5 (4-($CH(CH_3)_2$)-$C_6H_4CH(OH)CH_2CH_2$-*o*-$\underline{C}_6H_5$), 126.7 (4-($CH(CH_3)_2$)-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 126.1 (4-($CH(CH_3)_2$)-$\underline{C}_6H_4CH(OH)CH_2CH_2C_6H_5$), 126.0 (4-($CH(CH_3)_2$)-$C_6H_4CH(OH)CH_2CH_2$-*p*-$\underline{C}_6H_5$), 73.9 (4-($CH(CH_3)_2$)-$C_6H_4\underline{C}H(OH)CH_2CH_2C_6H_5$), 40.5 (4-($CH(CH_3)_2$)-$C_6H_4CH(OH)\underline{C}H_2CH_2C_6H_5$), 34.0 (4-($\underline{C}H(CH_3)_2$)-$C_6H_4CH(OH)CH_2CH_2C_6H_5$), 32.3 (4-($CH(CH_3)_2$)-$C_6H_4CH(OH)CH_2\underline{C}H_2C_6H_5$), 24.2 (4-($CH(\underline{C}H_3)_2$)-$C_6H_4CH(OH)CH_2CH_2C_6H_5$). GCMS (ESI): $[M]^+$ *m*/*z* = 254. Anal. Calcd. for $C_{18}H_{22}O$: C, 84.99; H, 8.72; Found: C, 85.07; H, 8.15.

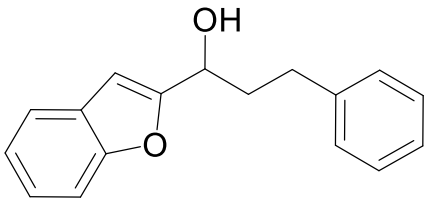
1-(4-(*tert*-Butyl)phenyl)-3-phenylpropan-1-ol (11)[4]



(**11**)

Colorless liquid [0.192 g, 72 % isolated yield (**1c**); 0.202 g, 75 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.40 (d, 2H, $^{3}J_{HH}$ = 8 Hz, 4-(C(CH$_3$)$_3$)-C$_6$*H*$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$), 7.34-7.26 (m, 4H, 4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$*H*$_5$), 7.23-7.18 (m, 3H, 4-(C(CH$_3$)$_3$)-C$_6$*H*$_4$CH(OH)CH$_2$CH$_2$C$_6$*H*$_5$), 4.69−4.66 (m, 1H, 4-(C(CH$_3$)$_3$)-C$_6$H$_4$C*H*(OH)CH$_2$CH$_2$C$_6$H$_5$), 2.81−2.66 (m, 2H, 4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)C*H*$_2$CH$_2$C$_6$H$_5$), 2.21−2.00 (m, 2H, 4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$C*H*$_2$C$_6$H$_5$), 1.88 (b, 1H, 4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(O*H*)CH$_2$CH$_2$C$_6$H$_5$), 1.35 (s, 9H, 4-(C(C*H*$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$). ^{13}C{^{1}H} NMR ($CDCl_3$, 100 MHz, 25 °C): δ 150.7 (4-(C(CH$_3$)$_3$)-*C*$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$), 142.0 (4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$-*ipso*-*C*$_6$H$_5$), 141.7 (4-(C(CH$_3$)$_3$)-*ipso*-*C*$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$), 128.6 (4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$-*m*-*C*$_6$H$_5$), 128.5 (4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$-*o*-*C*$_6$H$_5$), 126.0 (4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$-*p*-*C*$_6$H$_5$), 125.8 (4-(C(CH$_3$)$_3$)-*C*$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$), 125.6 (4-(C(CH$_3$)$_3$)-*C*$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$), 73.8 (4-(C(CH$_3$)$_3$)-C$_6$H$_4$*C*H(OH)CH$_2$CH$_2$C$_6$H$_5$), 40.4 (4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)*C*H$_2$CH$_2$C$_6$H$_5$), 34.7 (4-(*C*(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$), 32.3 (4-(C(CH$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$*C*H$_2$C$_6$H$_5$), 31.5 (4-(C(*C*H$_3$)$_3$)-C$_6$H$_4$CH(OH)CH$_2$CH$_2$C$_6$H$_5$). GCMS (ESI): $[M]^{+}$ *m*/*z* = 268. Anal. Calcd. for $C_{19}H_{24}O$: C, 85.03; H, 9.01; Found: C, 84.33; H, 8.30.

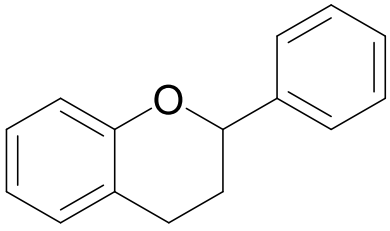
1-(Benzofuran-2-yl)-3-phenylpropan-1-ol (12)[4]



(**12**)

Colorless liquid [0.171 g, 68 % isolated yield (**1c**); 0.161 g, 64 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.55 (dd, 1H, $^{3}J_{HH}$ = 8 Hz, $^{4}J_{HH}$ = 1 Hz, $C_8\underline{H}_5OCH(OH)CH_2CH_2C_6H_5$), 7.47 (d, 1H, $^{3}J_{HH}$ = 8 Hz, $C_8\underline{H}_5OCH(OH)CH_2CH_2C_6H_5$), 7.32-7.19 (m, 7H, $C_8\underline{H}_5OCH(OH)CH_2CH_2C_6\underline{H}_5$), 6.64 (s, 1H, $C_8\underline{H}_5OCH(OH)CH_2CH_2C_6H_5$), 4.83 (t, 1H, $^{3}J_{HH}$ = 7 Hz, $C_8H_5OC\underline{H}(OH)CH_2CH_2C_6H_5$), 2.83−2.77 (m, 2H, $C_8H_5OCH(OH)C\underline{H}_2CH_2C_6H_5$), 2.31−2.25 (m, 2H, $C_8H_5OCH(OH)CH_2C\underline{H}_2C_6H_5$), 1.65 (d, 1H, $^{3}J_{HH}$ = 7 Hz, $C_8H_5OCH(O\underline{H})CH_2CH_2C_6H_5$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ 159.3 (*ipso*-$\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 155.0 ($\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 141.5 ($C_8H_5OCH(OH)CH_2CH_2$-*ipso*-$\underline{C}_6H_5$), 128.7 ($C_8H_5OCH(OH)CH_2CH_2$-*m*-$\underline{C}_6H_5$), 128.6 ($C_8H_5OCH(OH)CH_2CH_2$-*o*-$\underline{C}_6H_5$), 128.3 ($\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 126.2 ($C_8H_5OCH(OH)CH_2CH_2$-*p*-$\underline{C}_6H_5$), 124.4 ($\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 123.0 ($\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 121.2 ($\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 111.4 ($\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 102.9 ($\underline{C}_8H_5OCH(OH)CH_2CH_2C_6H_5$), 67.7 ($C_8H_5O\underline{C}H(OH)CH_2CH_2C_6H_5$), 37.2 ($C_8H_5OCH(OH)\underline{C}H_2CH_2C_6H_5$), 31.8 ($C_8H_5OCH(OH)CH_2\underline{C}H_2C_6H_5$). GCMS (ESI): $[M]^+$ *m*/*z* = 252. Anal. Calcd. for $C_{17}H_{16}O_2$: C, 80.93; H, 6.39; Found: C, 81.13; H, 5.87.

Synthesis of 2-Phenylchroman[5]

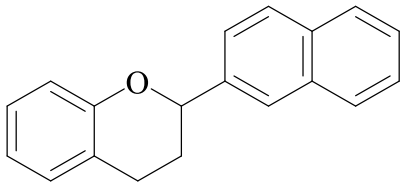


(**13**)

Colorless dense liquid [0.0346 g, 17 % isolated yield (**1c**); 0.0197 g, 10 % isolated yield (**2c**)]. ^{1}H NMR ($CDCl_3$, 500 MHz, 25 °C): δ 7.47−7.45 (m, 2H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_5$), 7.42 (t, 2H,

$^{3}J_{H\text{-}H}$ = 8 Hz, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_5$), 7.35 (t, 1H, $^{3}J_{H\text{-}H}$ = 8 Hz, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_5$), 7.17−7.11 (m, 2H, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_5$), 6.95-6.89 (m, 2H, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_5$), 5.10 (dd, 1H, $^{3}J_{H\text{-}H}$ = 8 Hz, $^{1}J_{H\text{-}H}$ = 2 Hz, $C_6H_4CH_2CH_2C\underline{H}(O)C_6H_5$), 3.07−3.00 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_5$), 2.86−2.81 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_5$), 2.27-2.22 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_5$), 2.17-2.08 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_5$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ ppm, 155.1, 141.8, 129.5, 128.5, 127.8, 127.3, 125.9, 121.8, 120.3, 116.9, 77.8, 29.9, 25.0. GCMS (ESI): $[M]^+$ *m*/*z* = 210. Anal. Calcd. for $C_{15}H_{14}O•1/4CH_2Cl_2$: C, 79.12.11; H, 6.31; Found: C, 79.34; H, 5.58 %.

2-(Naphthalen-2-yl)chroman (14)[5]

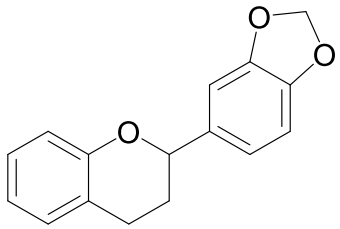


(**14**)

Yellow Solid [0.0618 g, 36 % isolated yield (**1c**); 0.0473 g, 28 % isolated yield (**2c**)]. ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.90−7.85 (m, 4H, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_3C_4\underline{H}_4$), δ 7.56−7.48 (m, 3H, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_3C_4\underline{H}_4$), δ 7.18−7.12 (m, 2H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_3C_4H_4$), δ 6.98−6.96 (m, 1H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_3C_4H_4$), 6.90 (t, 1H, $^{3}J_{H\text{-}H}$ = 8 Hz, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_3C_4H_4$), 5.25 (dd, 1H, $^{3}J_{H\text{-}H}$ = 8 Hz, $^{1}J_{H\text{-}H}$ = 2 Hz, $C_6H_4CH_2CH_2C\underline{H}(O)C_6H_3C_4H_4$), 3.09−3.01 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_3C_4H_4$), 2.87−2.81 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_3C_4H_4$), 2.34-2.27 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_3C_4H_4$), 2.14-2.14 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_3C_4H_4$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ ppm, 155.1, 139.1, 133.3, 133.1, 129.6, 128.3, 128.1, 127.7, 127.4, 126.2, 125.9, 124.9, 124.0,

121.9, 120.4, 116.9, 77.9, 29.9, 25.2. GCMS (ESI): $[M]^+$ *m*/*z* = 260. Anal. Calcd. for $C_{19}H_{16}O.1/4CH_2Cl_2$: C, 82.12; H, 5.91; Found: C, 81.68; H, 5.66 %.

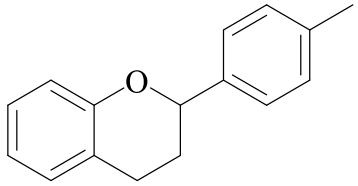
2-(Benzo[1,3]dioxol-5-yl)chroman (15)[5]



(**15**)

Colorless dense liquid [0.0301 g, 18 % isolated yield (**1c**); 0.0246 g, 15 % isolated yield (**2c**)]. ^{1}H NMR ($CDCl_3$, 500 MHz, 25 °C): δ 7.15−7.12 (m, 2H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6\underline{H}_3OCH_2O$), δ 6.97 (s, 1H, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_3OCH_2O$), δ 6.93−6.89 (m, 3H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6\underline{H}_3OCH_2O$), δ 6.85−6.84 (m, 1H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_3OCH_2O$), 5.99 (s, 2H, $C_6H_4CH_2CH_2CH(O)C_6H_3OC\underline{H}_2O$), 4.99 (dd, 1H, $^3J_{H\text{-}H}$ = 8 Hz, $^1J_{H\text{-}H}$ = 2 Hz, $C_6H_4CH_2CH_2C\underline{H}(O)C_6H_3OCH_2O$), 3.05−2.98 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_3OCH_2O$), 2.86−2.81 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_3OCH_2O$), 2.22-2.17 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_3OCH_2O$), 2.14-2.05 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_3OCH_2O$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 125 MHz, 25 °C): δ ppm, 155.1, 147.8, 147.2, 135.6. 129.5, 127.4, 121.7, 120.3, 119.6, 116.9, 108.2, 106.7, 101.1, 77.7, 29.9, 25.2. GCMS (ESI): $[M]^+$ *m*/*z* = 254. Anal. Calcd. for $C_{16}H_{14}O_3$: C, 75.58; H, 5.55; Found: C, 74.42; H, 4.29 %.

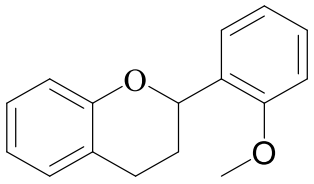
2-(4-Tolyl)chroman (16)[5]



(**16**)

Colorless solid [0.0175 g, 12 % isolated yield (**1c**); 0.0153 g, 11 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.32−7.31 (m, 2H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_4CH_3$), 7.20 (d, 2H, $^{3}J_{H-H}$ = 8 Hz, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_4CH_3$), 7.14−7.06 (m, 2H, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_4CH_3$), 6.91-6.84 (m, 2H, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_4CH_3$), 5.04 (m, 1H, $C_6H_4CH_2CH_2C\underline{H}(O)C_6H_4CH_3$), 3.05−2.97 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_4CH_3$), 2.84−2.78 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_4CH_3$), 2.40 (s, 3H, $C_6H_4CH_2CH_2CH(O)C_6H_4C\underline{H}_3$), 2.24-2.19 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_4CH_3$), 2.16-2.06 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_4CH_3$). $^{13}C\{^{1}H\}$ NMR ($CDCl_3$, 400 MHz, 25 °C): δ ppm, 155.2, 138.8, 137.2, 129.4, 129.1, 127.3, 125.9, 121.6, 120.2, 96.2, 77.5, 30.0, 25.2, 21.2. GCMS (ESI): $[M]^+$ *m*/*z* = 224. Anal. Calcd. for $C_{16}H_{16}O$: C, 85.68; H, 7.19; Found: C, 85.41; H, 6.35 %.

2-(2-Methoxyphenyl)chroman (17)[5]



(**17**)

Colorless solid [0.0214 g, 14 % isolated yield (**1c**); 0.0197 g, 13 % isolated yield (**2c**)] ^{1}H NMR ($CDCl_3$, 400 MHz, 25 °C): δ 7.53 (d, 1H, $^{3}J_{H-H}$ = 8 Hz, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_4OCH_3$), 7.31 (t, 1H, $^{3}J_{H-H}$ = 8 Hz, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_4OCH_3$), 7.17-7.11 (m, 2H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6\underline{H}_4OCH_3$), 7.03 (t, 1H, $^{3}J_{H-H}$ = 8 Hz, $C_6H_4CH_2CH_2CH(O)C_6\underline{H}_4OCH_3$),

6.96-6.88 (m, 3H, $C_6\underline{H}_4CH_2CH_2CH(O)C_6H_4OCH_3$), 5.48 (d, 1H, $^3J_{H\text{-}H}$ = 10 Hz, $C_6H_4CH_2CH_2C\underline{H}(O)C_6H_4OCH_3$), 3.87 (s, 3H, $C_6H_4CH_2CH_2CH(O)C_6H_4OC\underline{H}_3$), 3.07−2.99 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_4OCH_3$), 2.82−2.76 (m, 1H, $C_6H_4C\underline{H}_2CH_2CH(O)C_6H_4OCH_3$), 2.30-2.25 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_4OCH_3$), 2.03-1.93 (m, 1H, $C_6H_4CH_2C\underline{H}_2CH(O)C_6H_4OCH_3$). $^{13}C\{^1H\}$ NMR ($CDCl_3$, 100 MHz, 25 °C): δ ppm, 155.9, 155.5, 130.3, 129.6, 128.5, 127.2, 126.5, 122.3, 120.8, 120.1, 116.9, 110.4, 72.4, 55.4, 28.6, 25.3. GCMS (ESI): $[M]^+$ *m*/*z* = 240. Anal. Calcd. for $C_{16}H_{14}O_2$: C, 79.97; H, 6.71; Found: C, 79.21; H, 6.47 %.

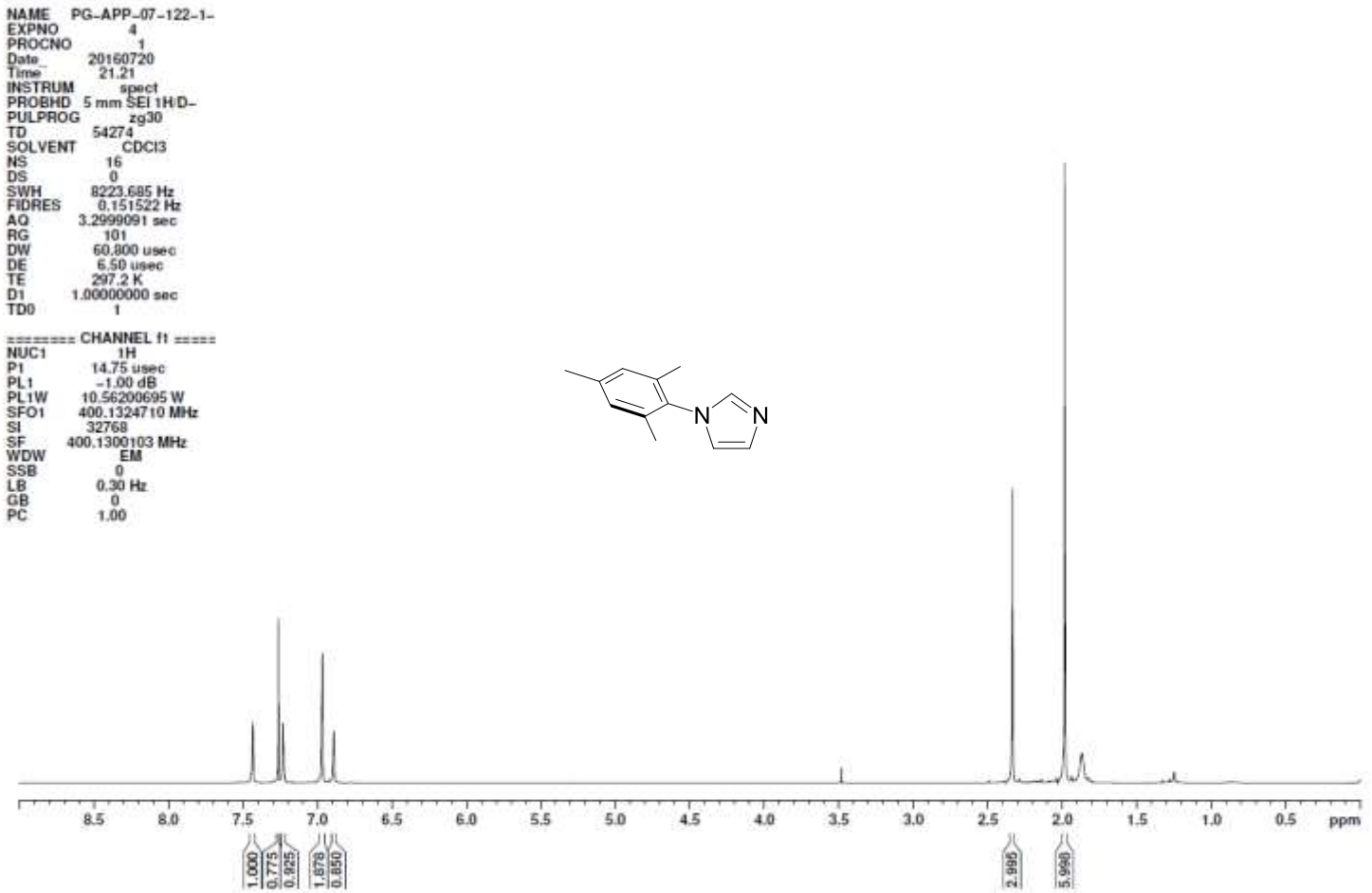


Figure S1. ^{1}H NMR spectrum of 1-Mesityl imidazole in $CDCl_3$.

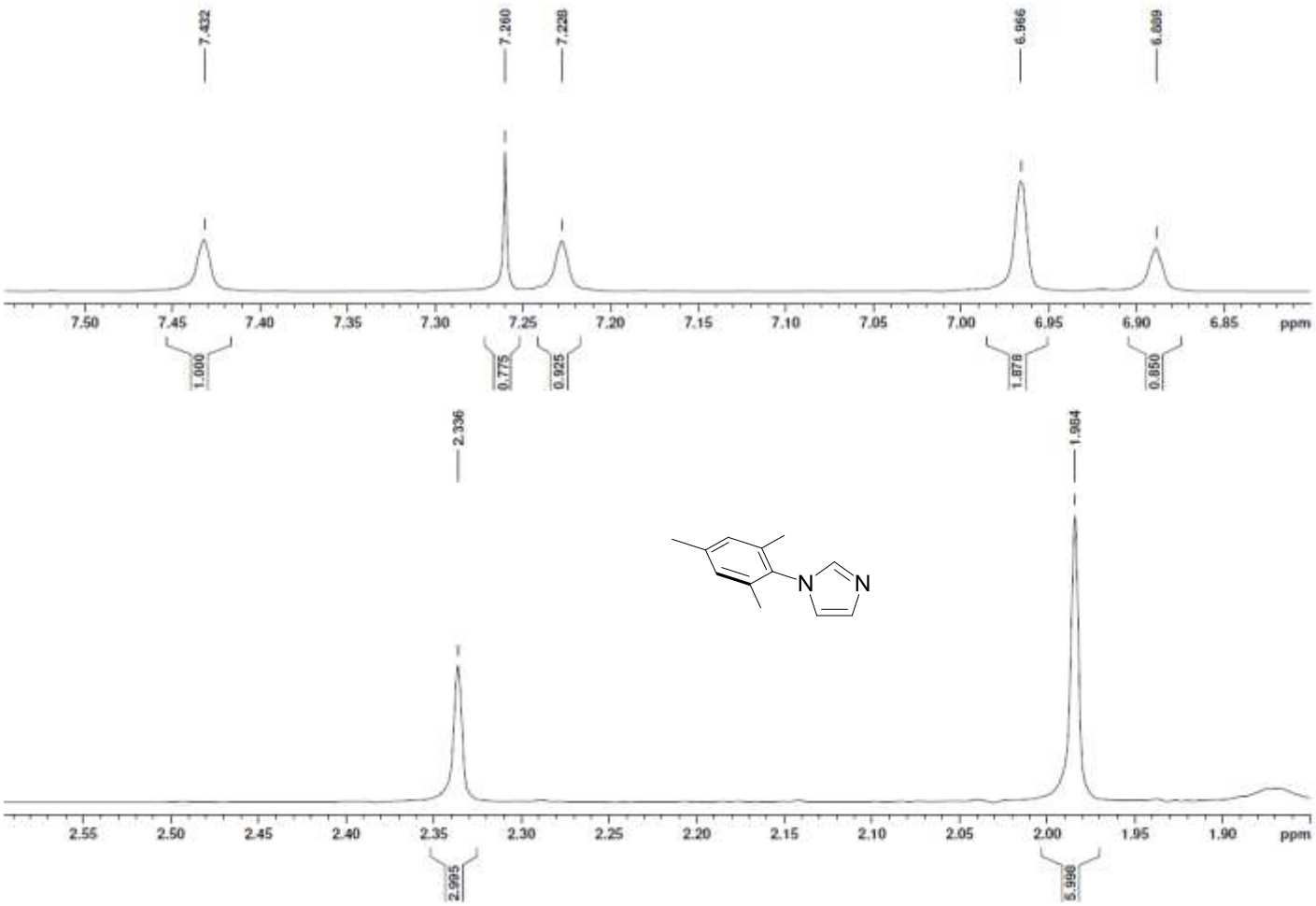


Figure S2. Expanded ^{1}H NMR spectrum of 1-Mesityl imidazole in $CDCl_3$.

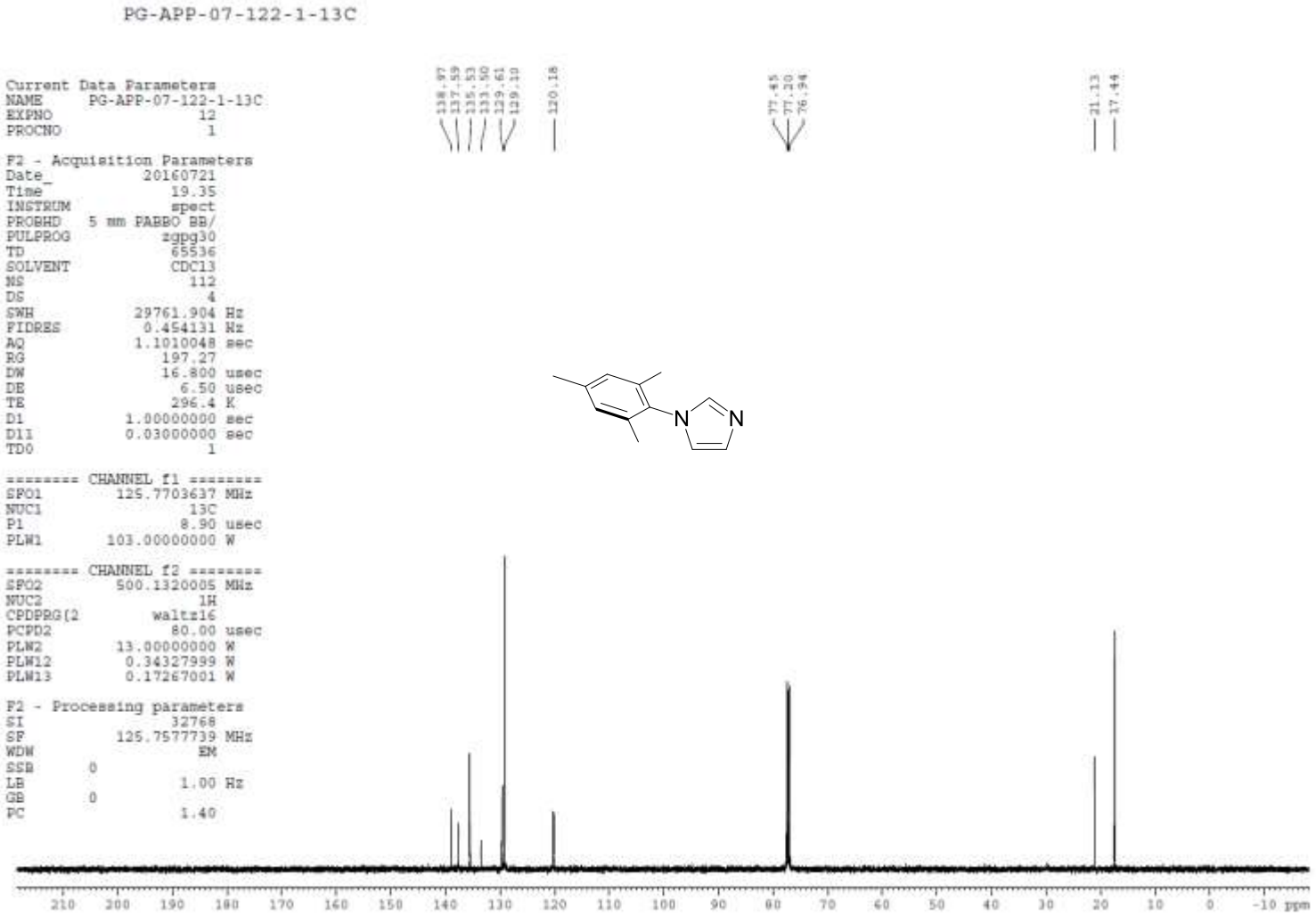


Figure S3. $^{13}C\{^{1}H\}$ NMR spectrum of 1-Mesityl imidazole in $CDCl_3$.

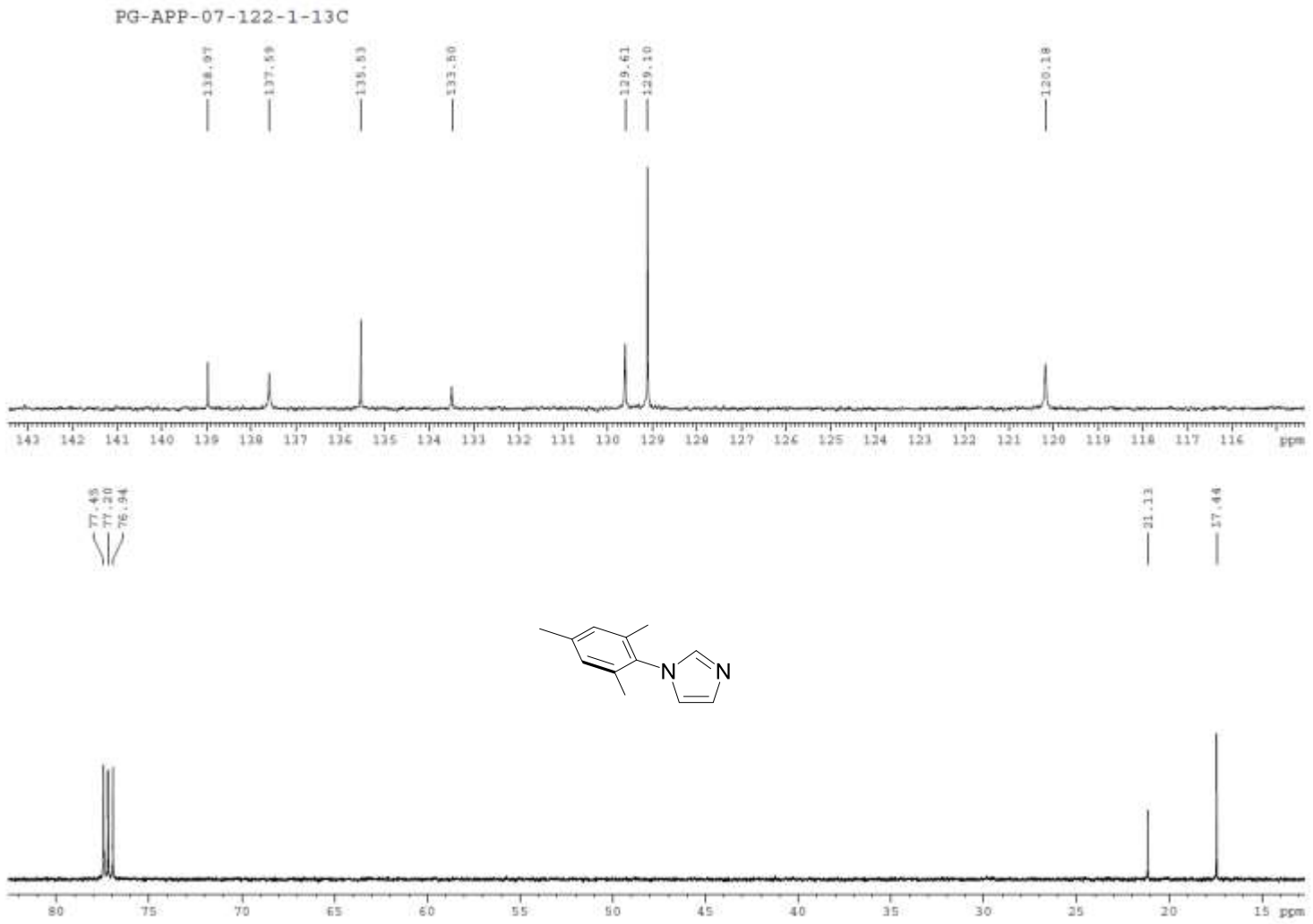


Figure S4. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of 1-Mesityl imidazole in $CDCl_3$.

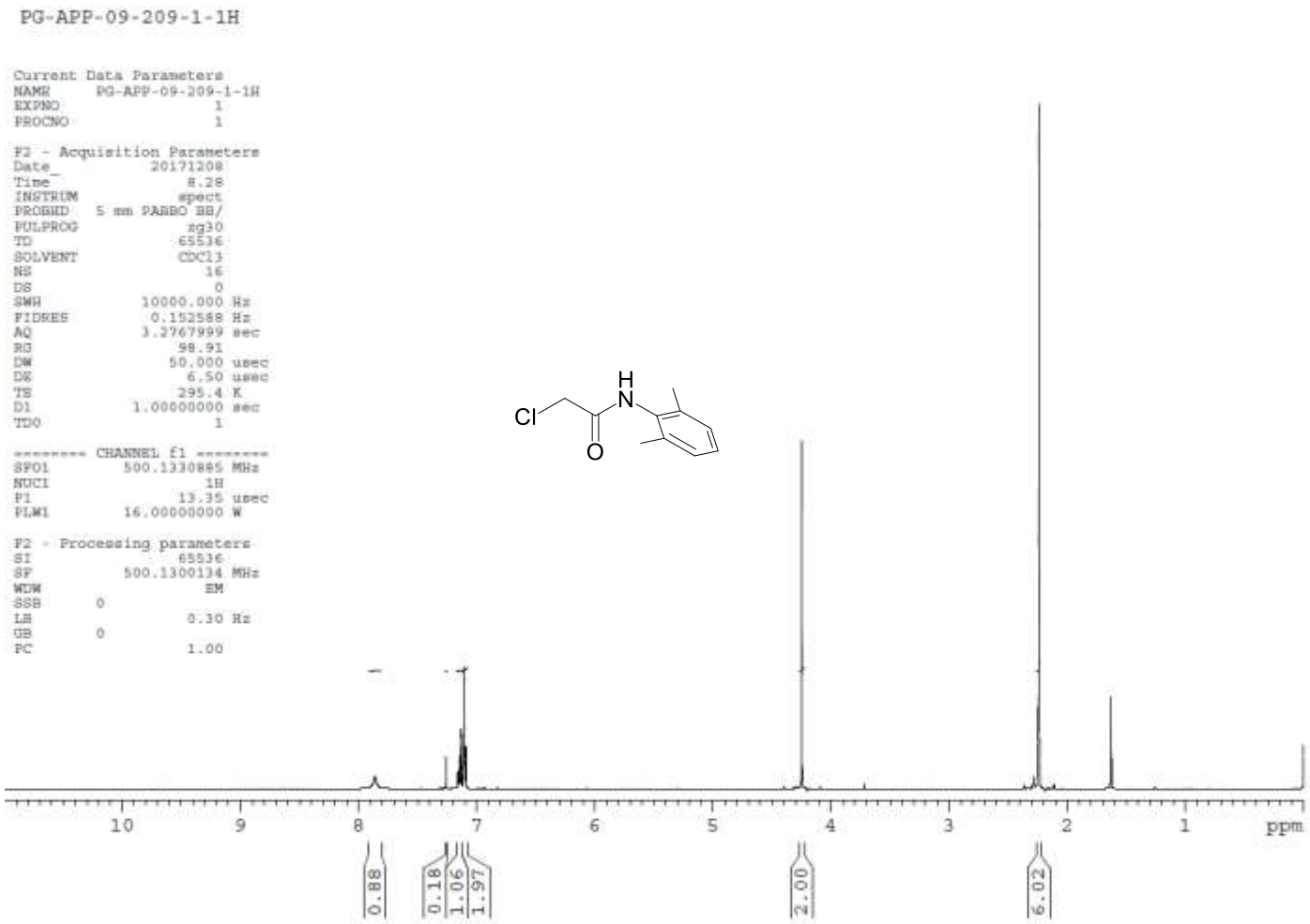


Figure S5. ^{1}H NMR spectrum of 2-chloro-*N*-(2,6-Me_2-phenyl)acetamide in $CDCl_3$.

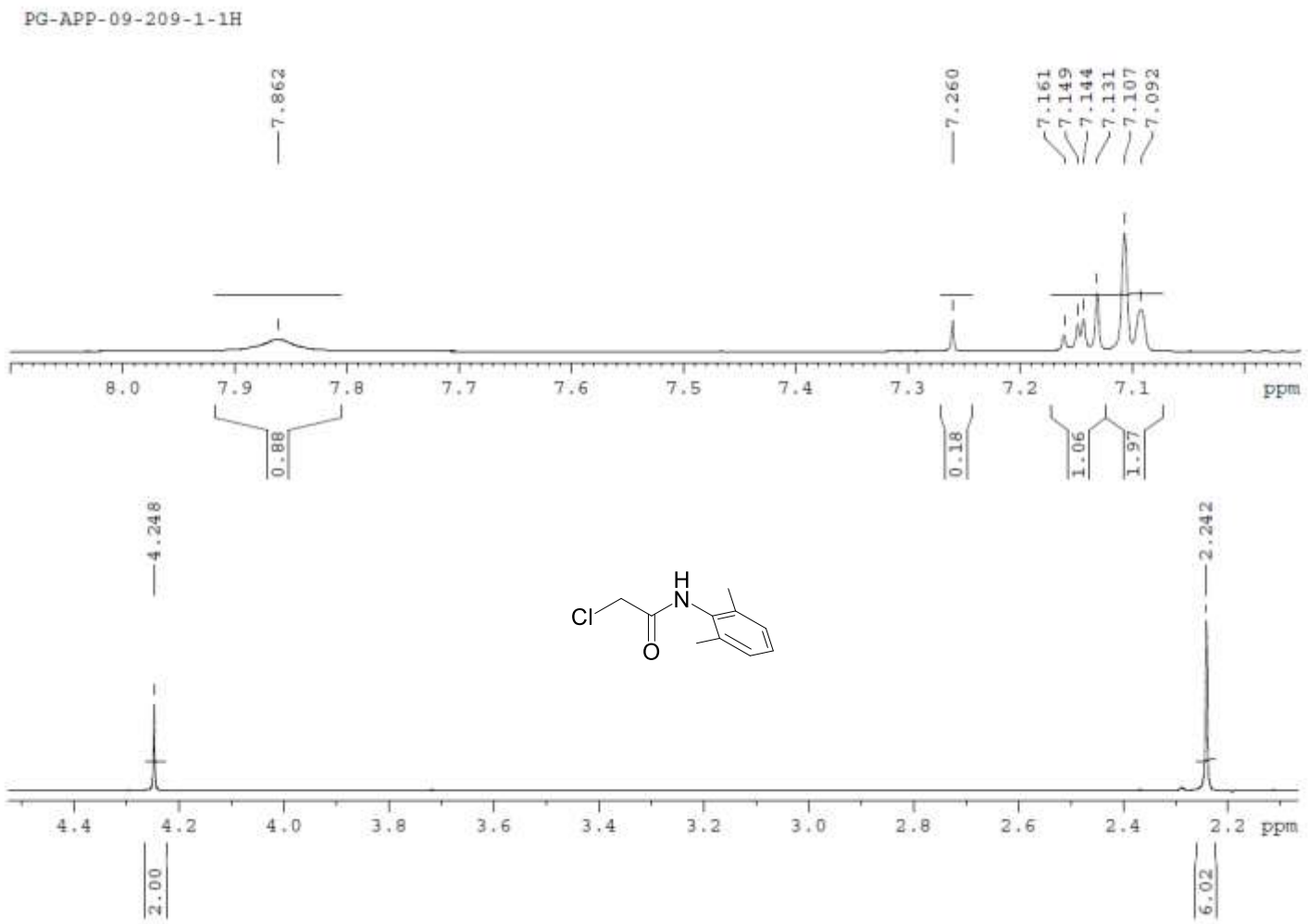


Figure S6. Expanded ^{1}H NMR spectrum of 2-chloro-*N*-(2,6-Me$_2$-phenyl)acetamide in CDCl$_3$.

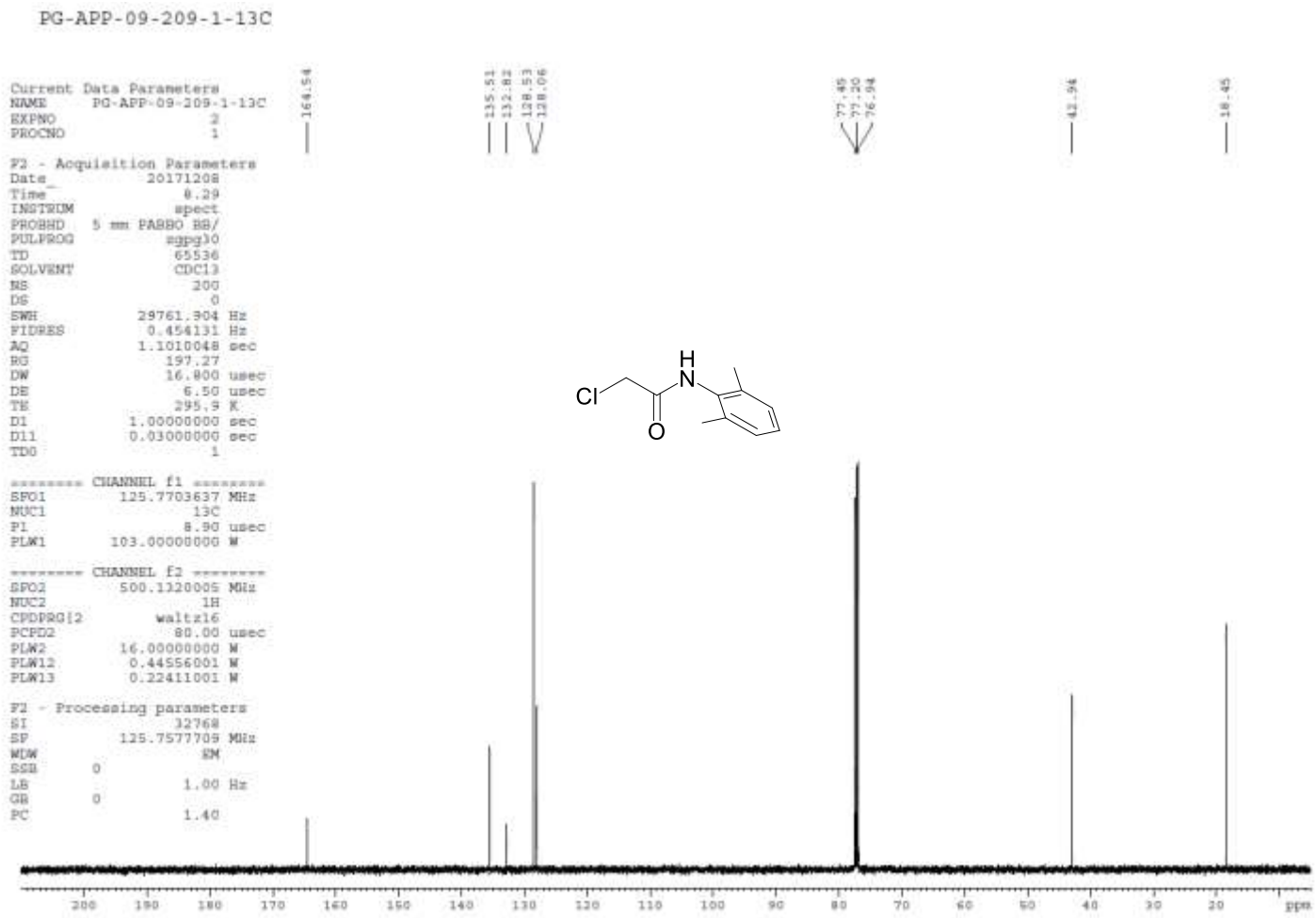


Figure S7. $^{13}C\{^{1}H\}$ NMR spectrum of 2-chloro-*N*-(2,6-Me_2-phenyl)acetamide in $CDCl_3$.

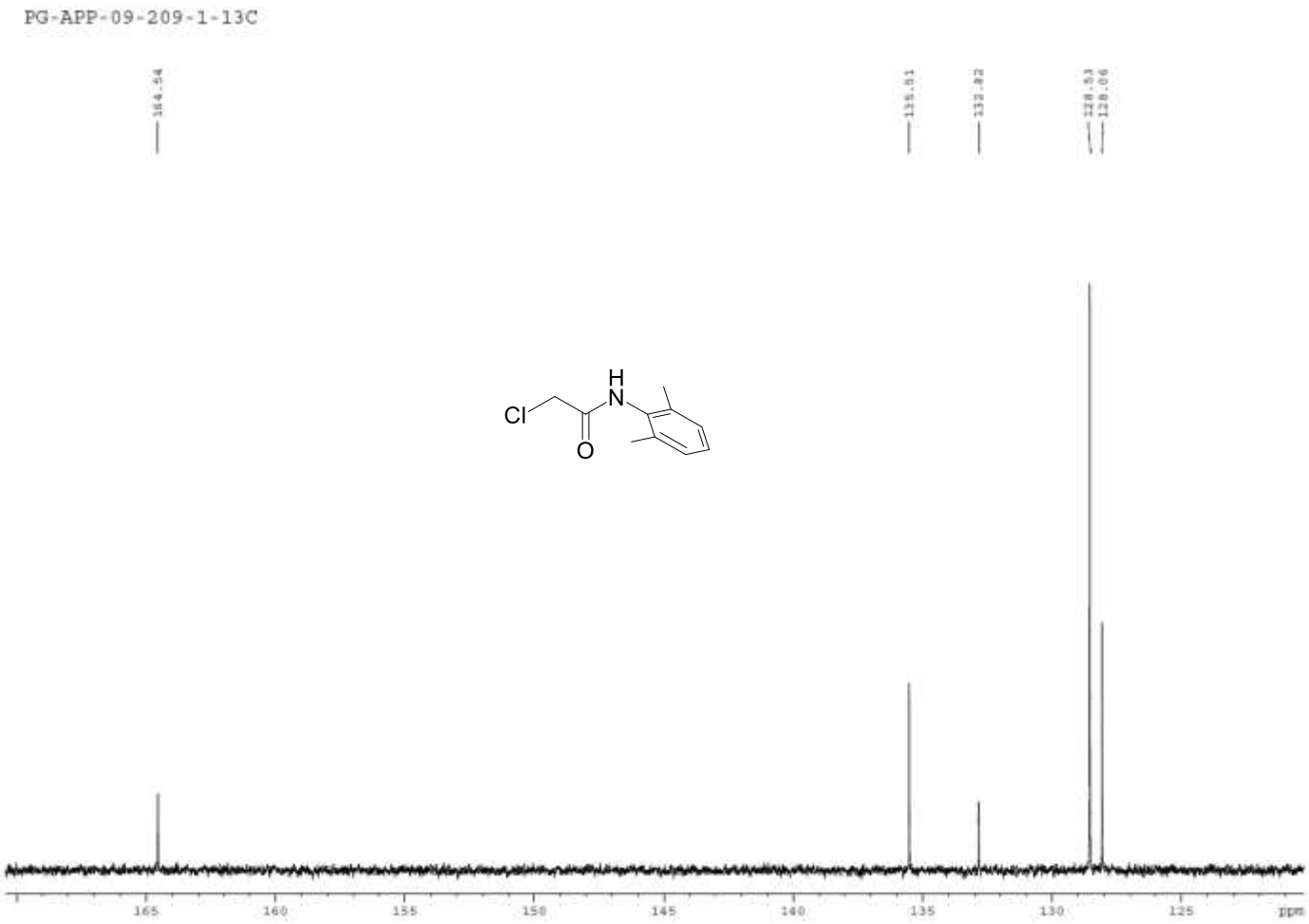
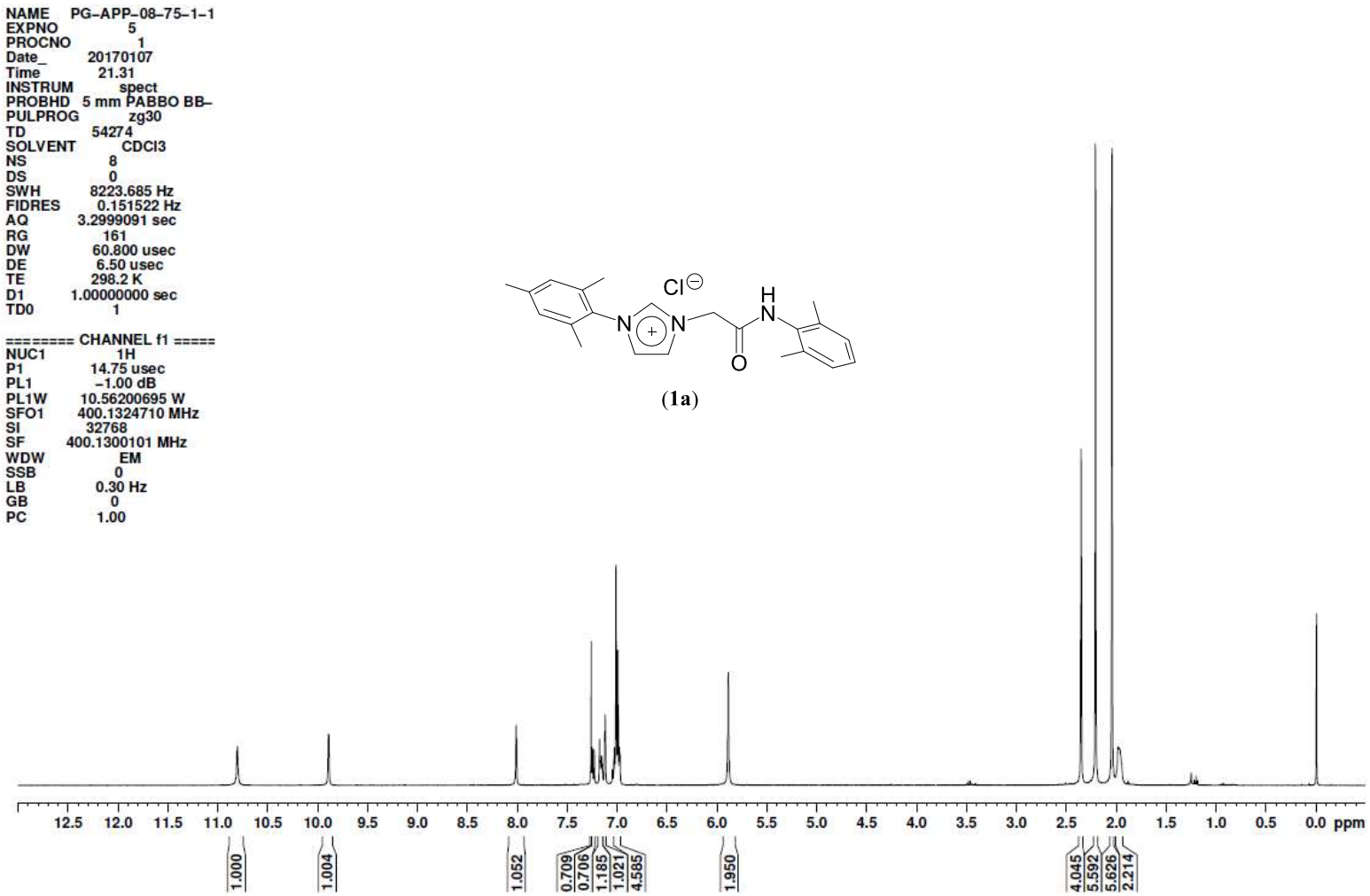


Figure S8. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of 2-chloro-*N*-(2,6-Me_2-phenyl)acetamide in $CDCl_3$.



(**1a**)

Figure S9. ^{1}H NMR spectrum of **1a** in $CDCl_3$.

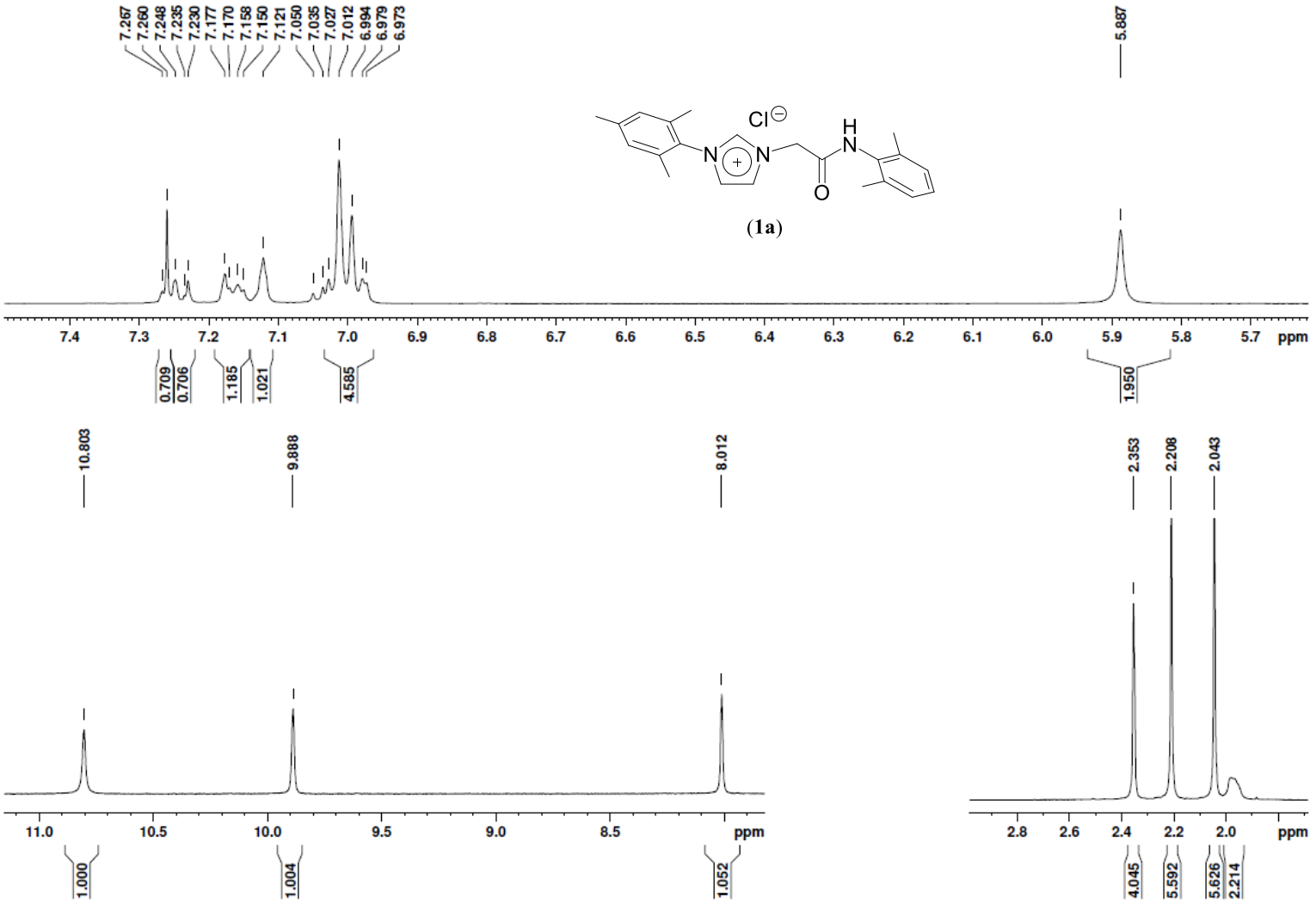


Figure S10. Expanded ^{1}H NMR spectrum of **1a** in $CDCl_3$.

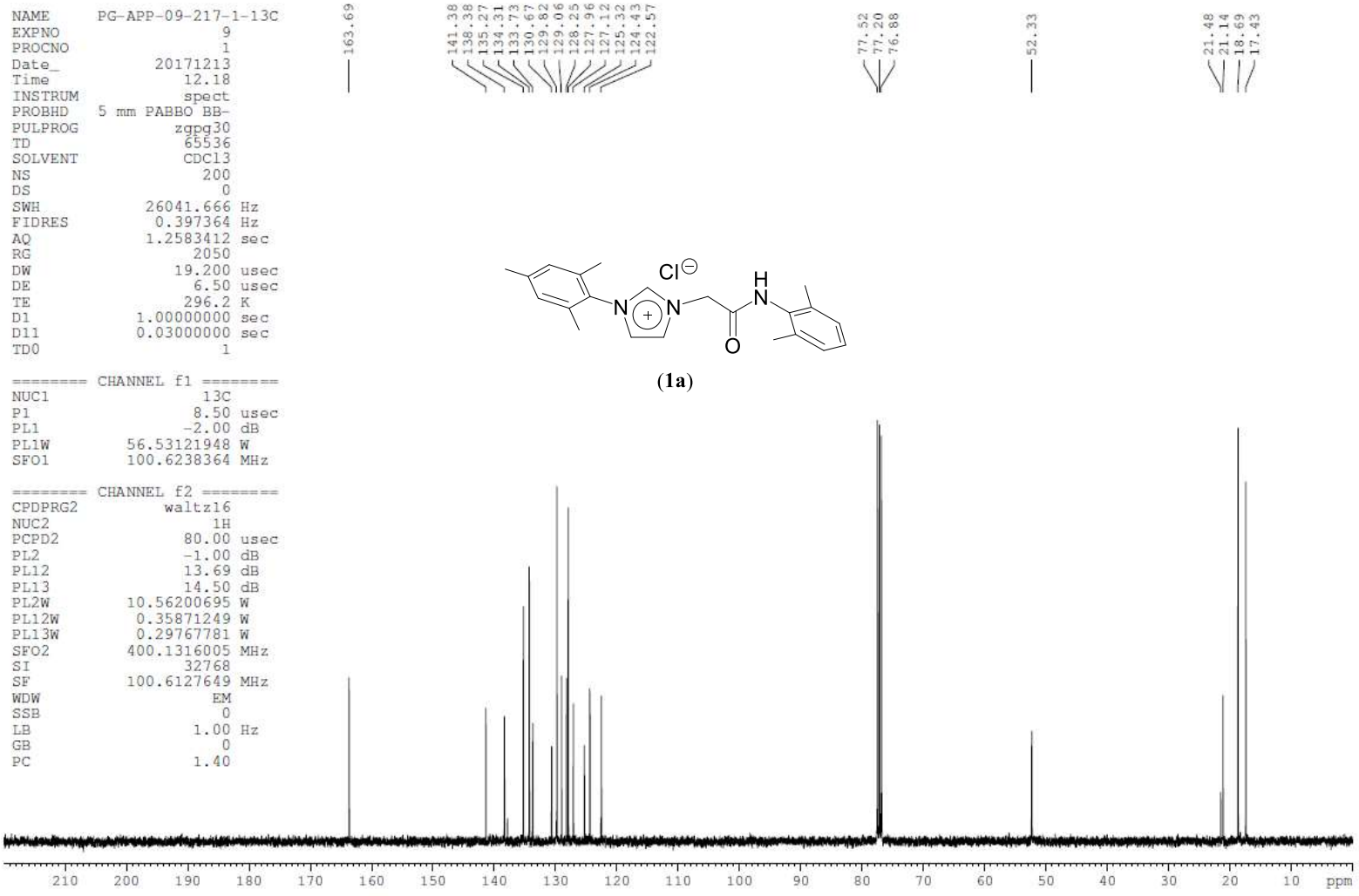
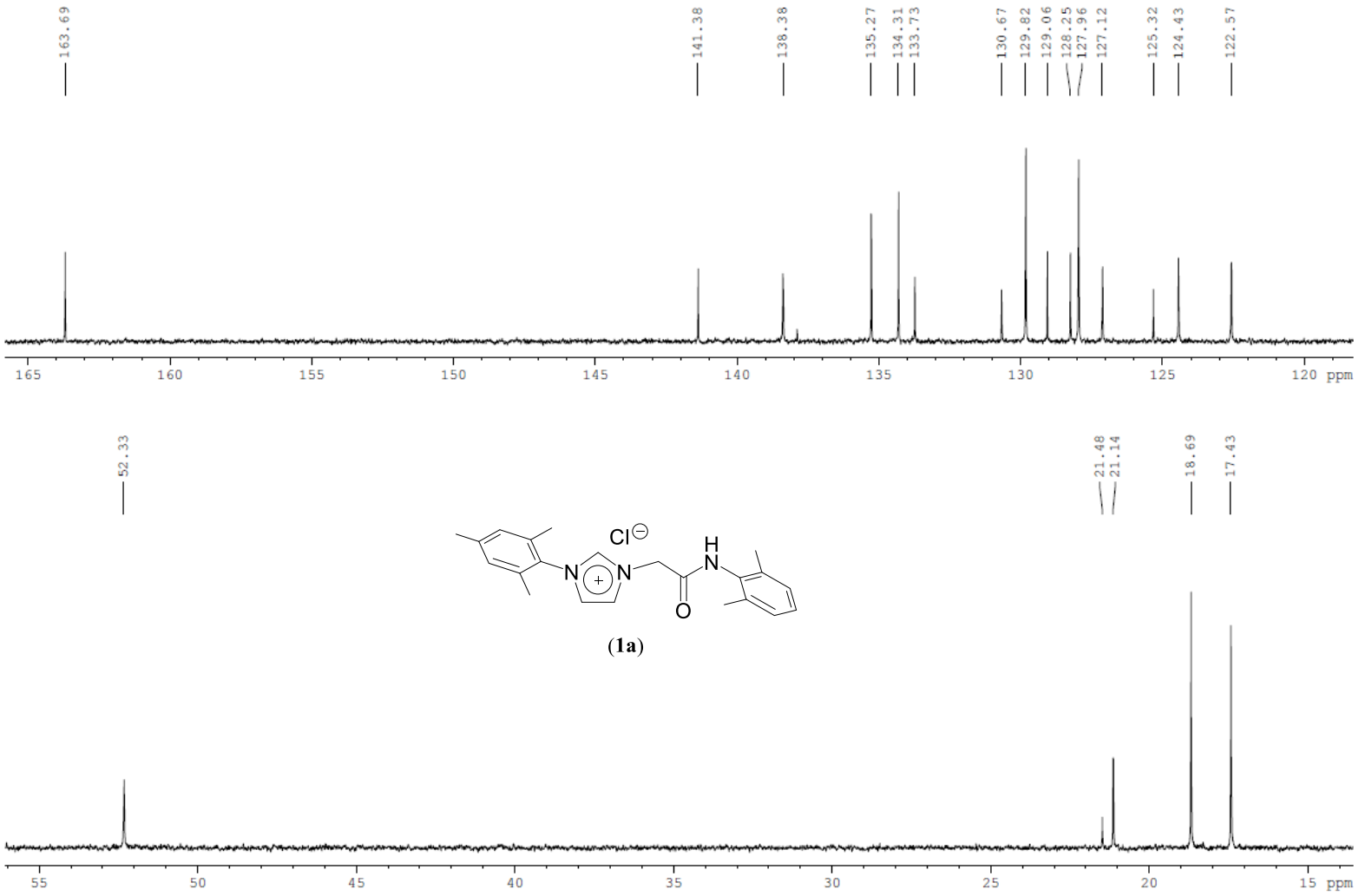


Figure S11. $^{13}C\{^{1}H\}$ NMR spectrum of **1a** in $CDCl_3$.



(**1a**)

Figure S12. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of **1a** in $CDCl_3$.

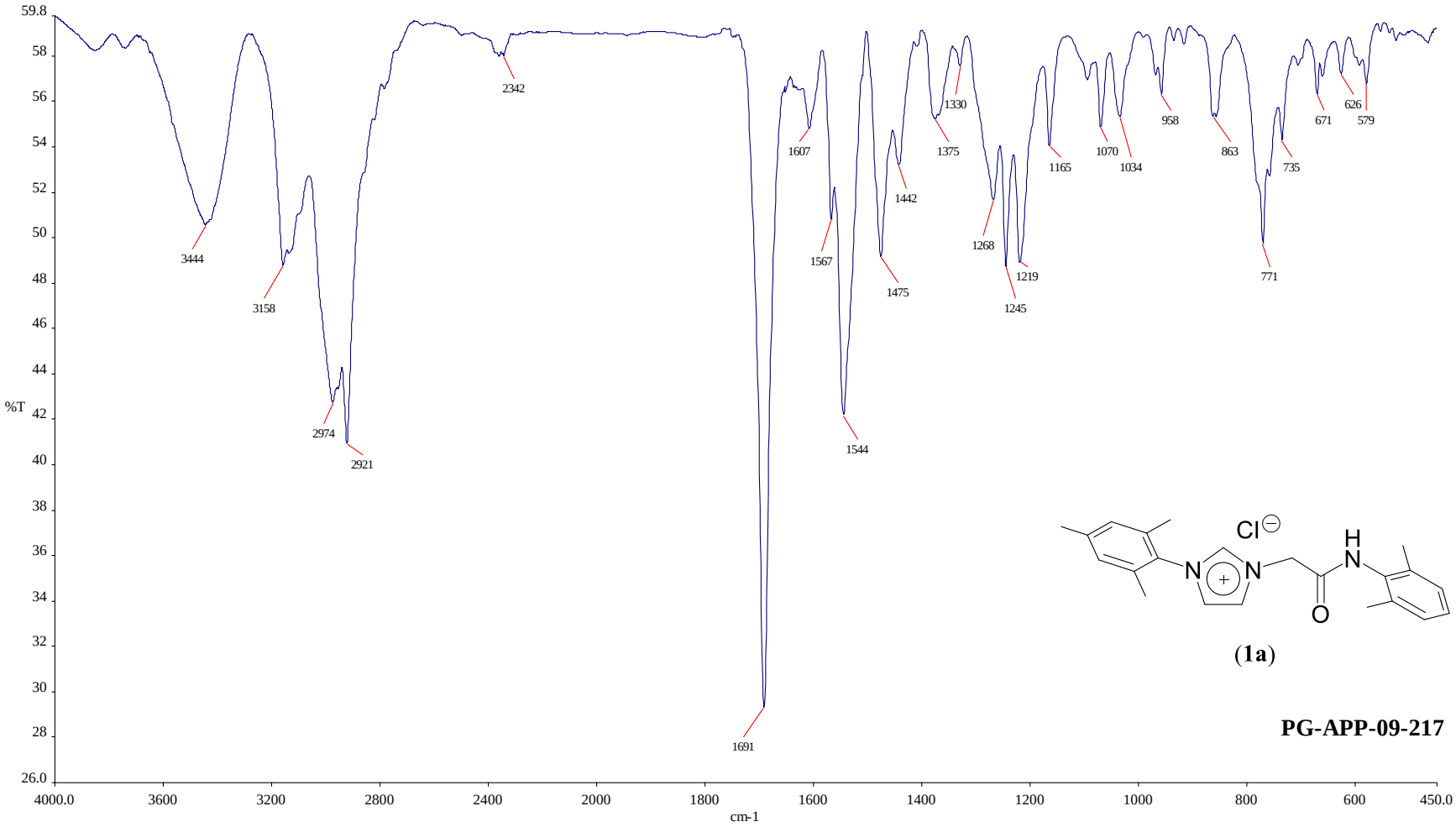
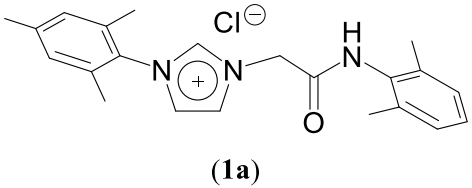


Figure S13. IR spectrum of **1a** in KBr.



(**1a**)

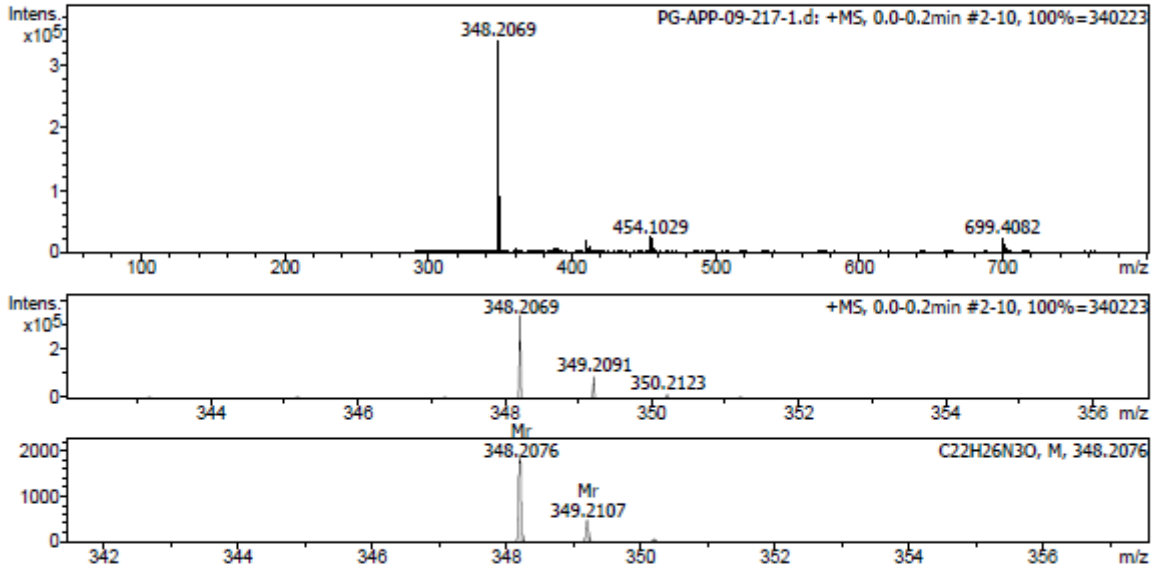
DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

Analysis Name	D:\Data\Dec 2017\PG-APP-09-217-1.d	Acquisition Date	12/22/2017 8:53:52 AM
Method	Tune_pos_NAICSI-1500A.m	Operator	PG APP IN
Sample Name	PG-APP-09-217-1	Instrument	maXis impact 282001.00081
Comment	C22H26N3OCl		

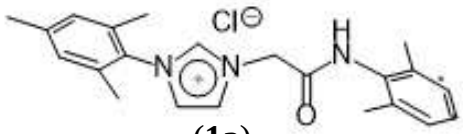
Acquisition Parameter

Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Active	Set Capillary	3700 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	800 m/z	Set Collision Cell RF	1800.0 Vpp	Set Divert Valve	Source



Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e⁻ Conf	N-Rule
348.2069	1	C22H26N3O	348.2070	0.5	6.0	1	100.00	11.5	even	ok

Figure S14. High Resolution Mass Spectrometry (HRMS) data of **1a**.



(1a)

Eager 300 Report

Page: 1 Sample: PG-APP-09-217-2 (PG-APP-09-217-2)

Method Name : PGAPP10012018
Method File : D:\CHNS2018\PGAPP10012018.mth
Chromatogram : PG-APP-09-217-2
Operator ID : Prakash
Analysed : 02/19/2018 18:09
Sample ID : PG-APP-09-217-2 (# 30)
Analysis Type : UnkNown (Area)

Company Name : C.E. Instruments
Printed : 1/26/2019 03:08
Instrument N. : Instrument #1
Sample weight : 1.257

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	18439	FU		0.0000
Nitrogen	10.8331	41	151800	FU	16.614450	.107234E+07
Carbon	68.3967	63	2522079	FU	1.00000	.262772E+07
Hydrogen	6.7151	181	600547	FU	4.199636	.660576E+07
Totals	85.9449		3292865			

Figure S15. Elemental analysis data of **1a**.

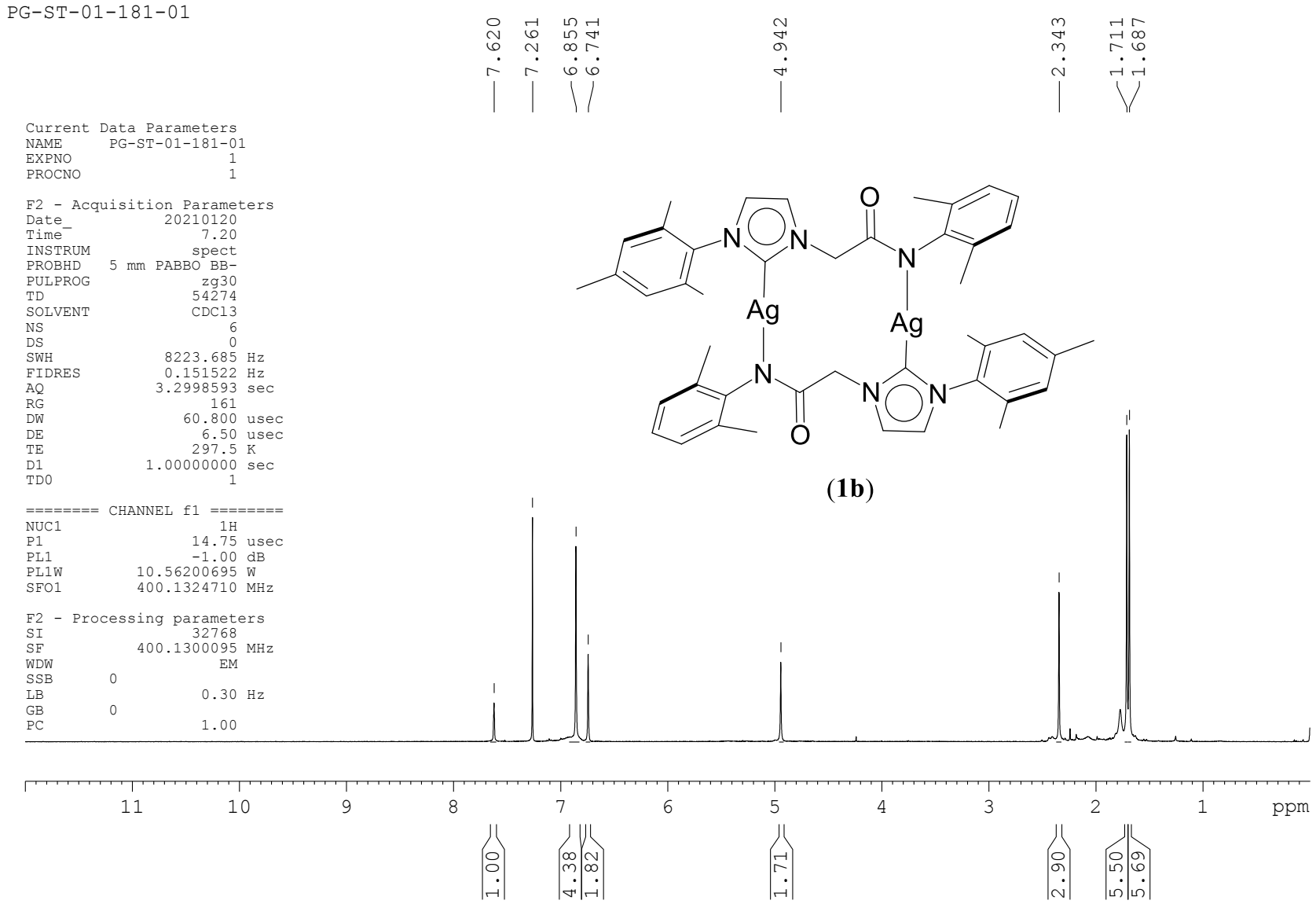


Figure S16. ^{1}H NMR spectrum of **1b** in $CDCl_3$.

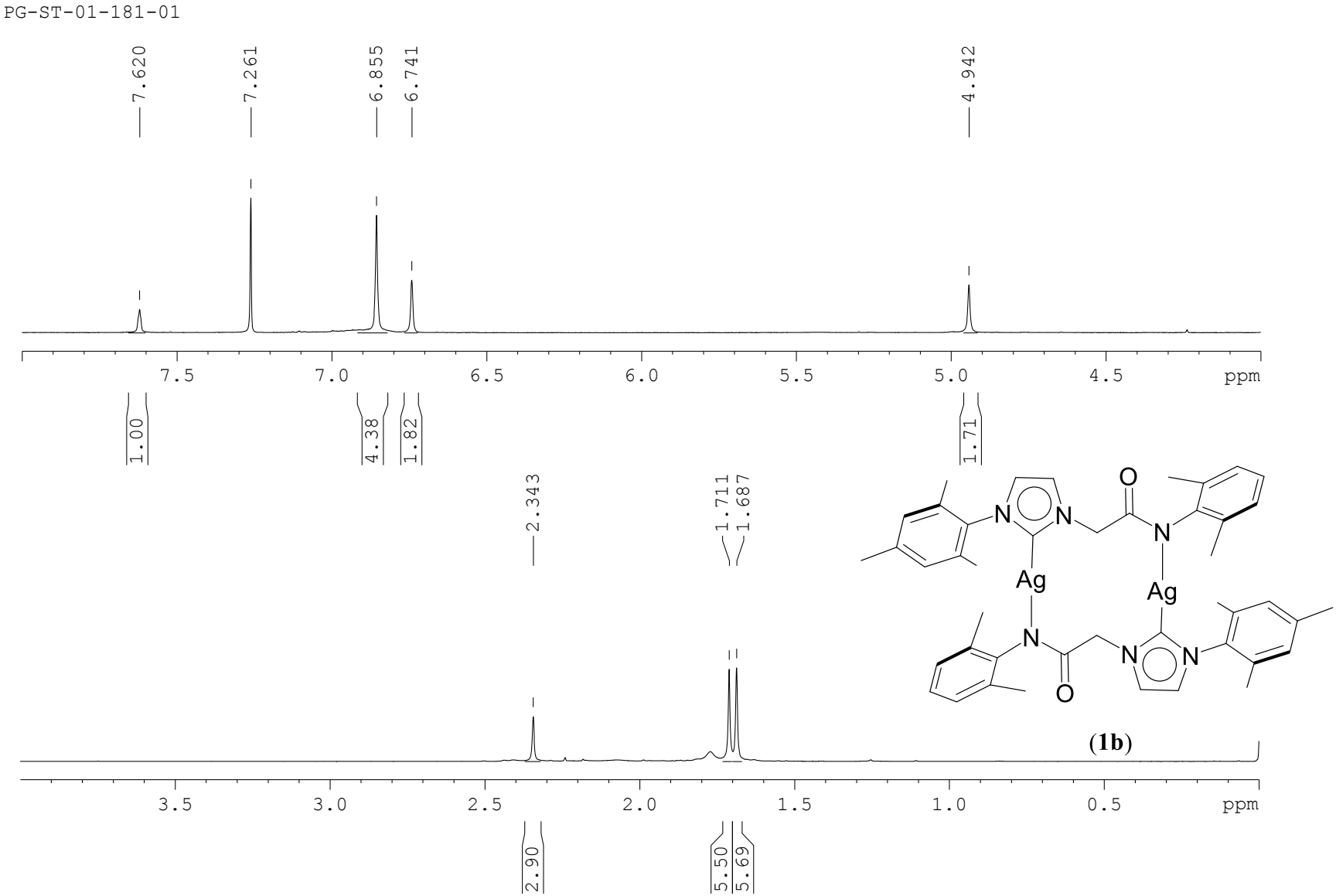


Figure S17. Expanded ^{1}H NMR spectrum of **1b** in CDCl3.

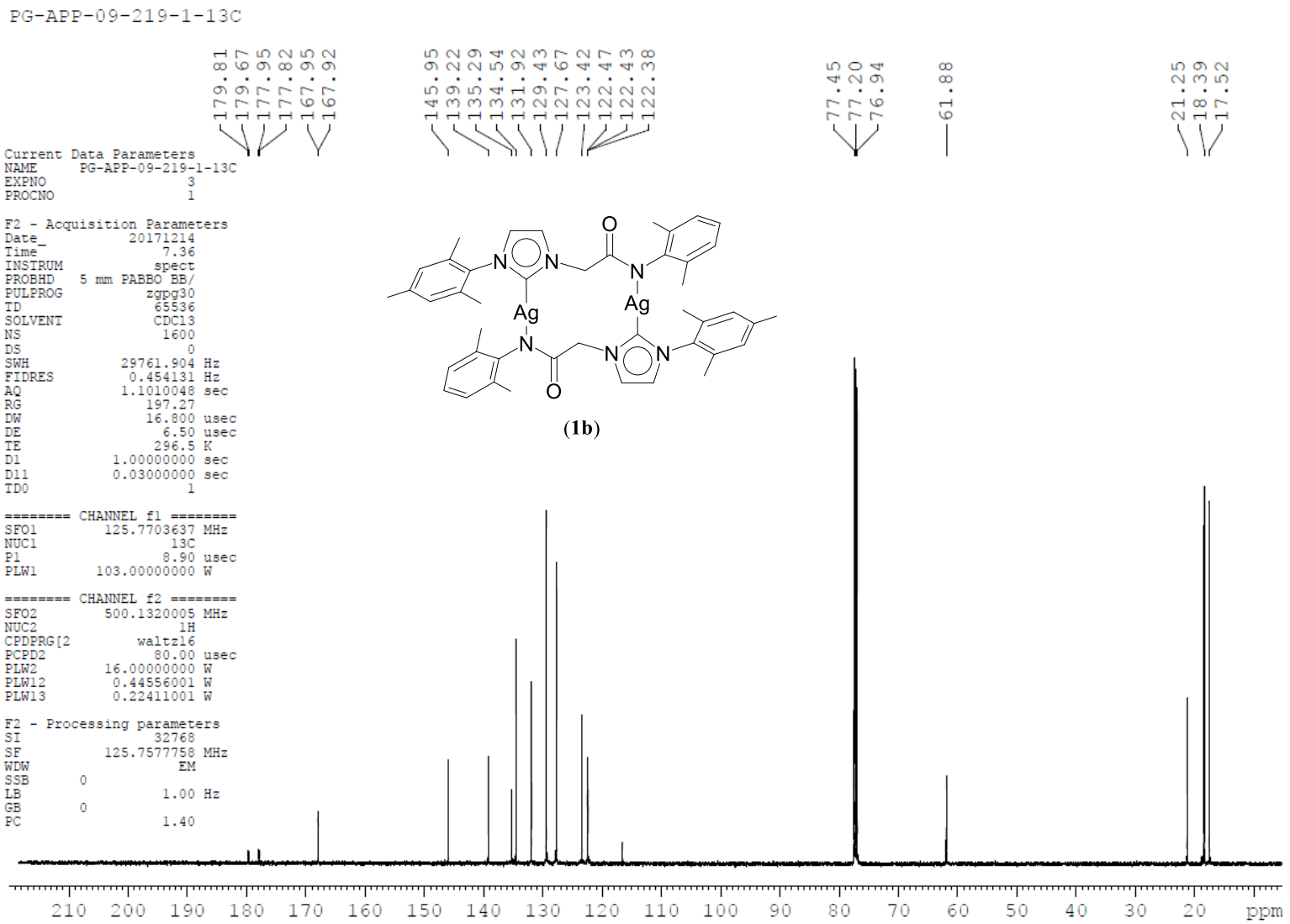


Figure S18. $^{13}C\{^{1}H\}$ NMR spectrum of **1b** in $CDCl_3$.

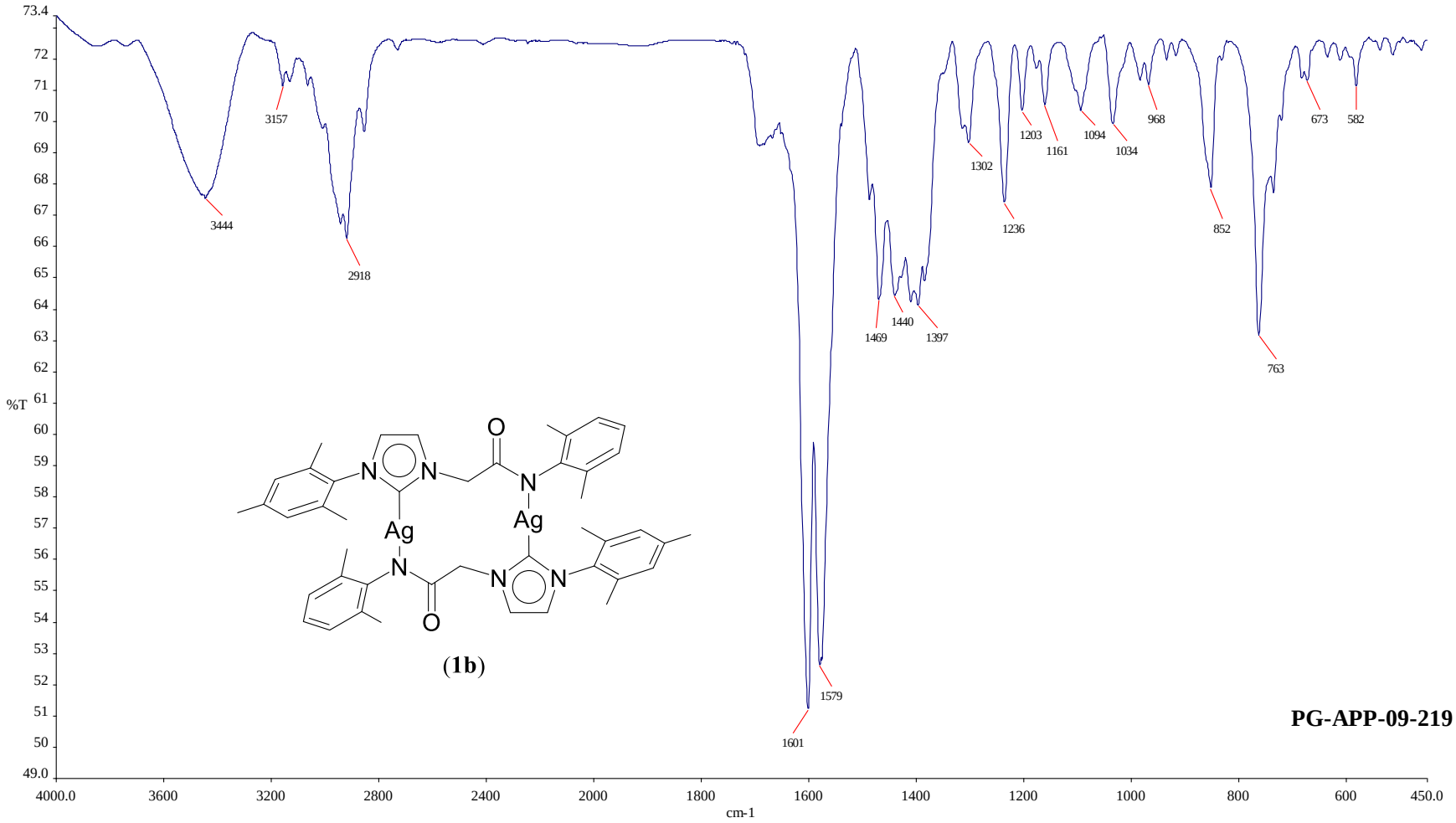


Figure S19. IR spectrum of **1b** in KBr.

SP18022016
varioMICRO CHNS
serial number: 15154051

Graphic report

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	N [%]	C [%]	H [%]	Date Time	Info
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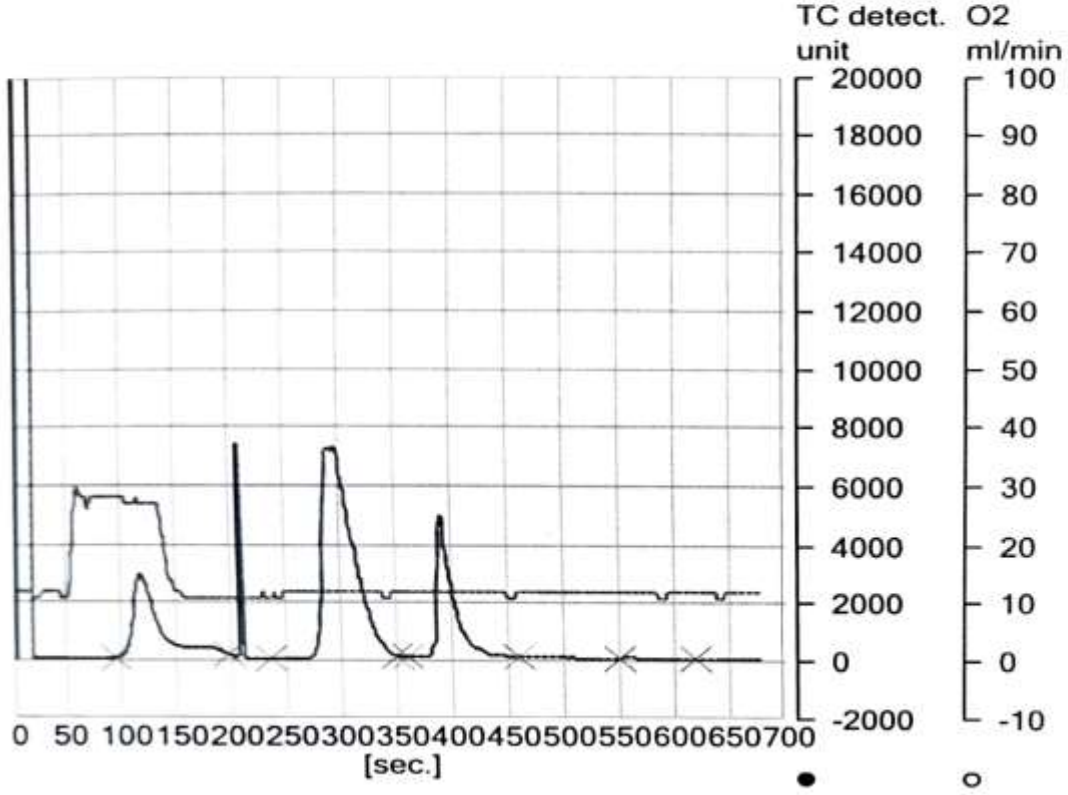
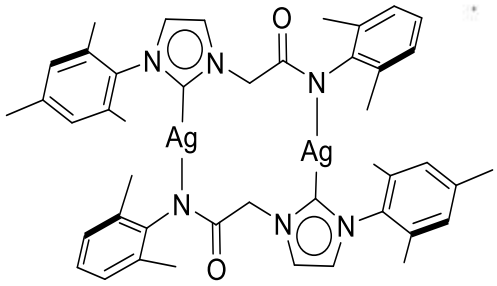




(**1b**)

Figure S20. Elemental analysis data of **1b**.

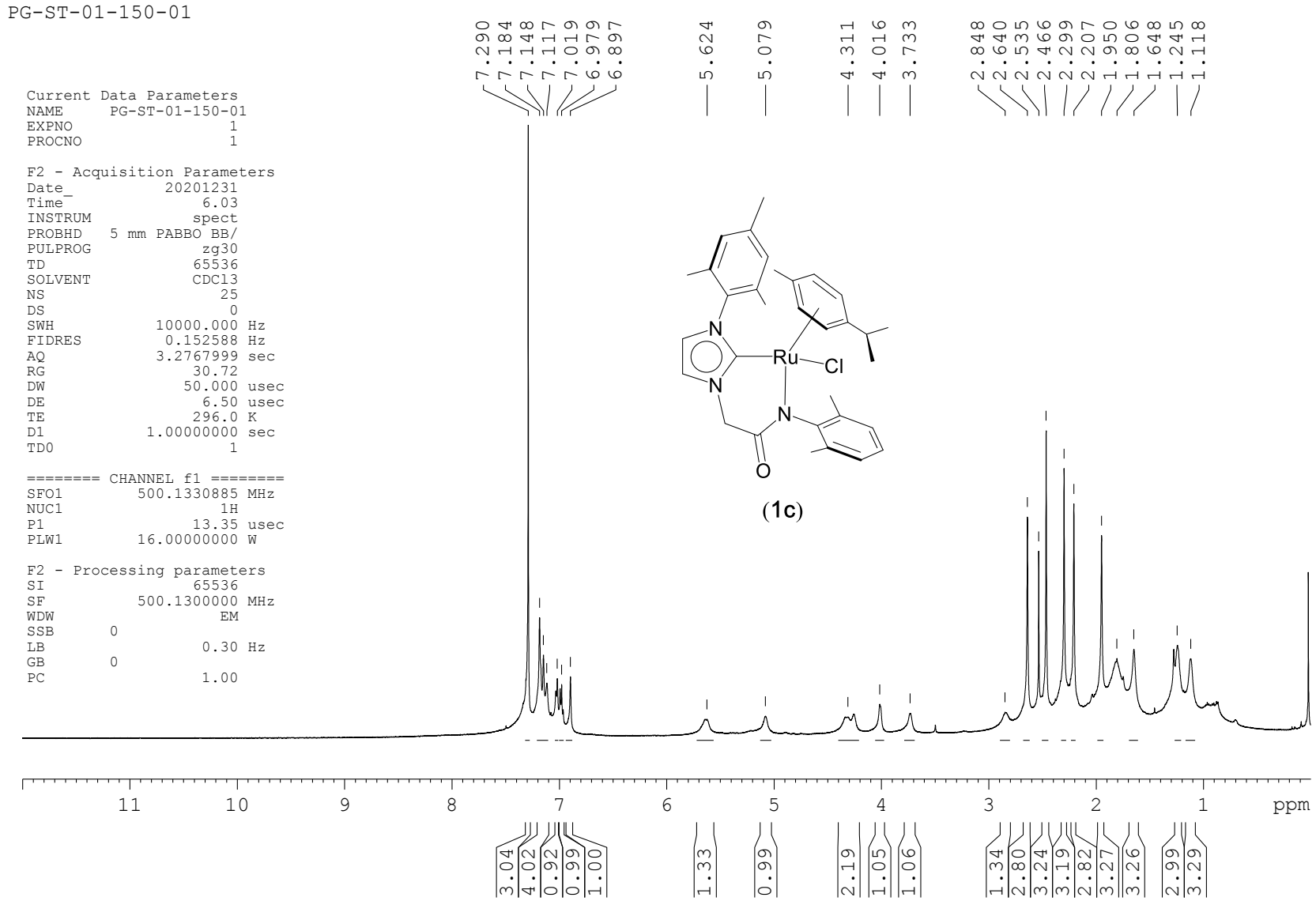


Figure S21. ^{1}H NMR spectrum of **1c** in $CDCl_3$.

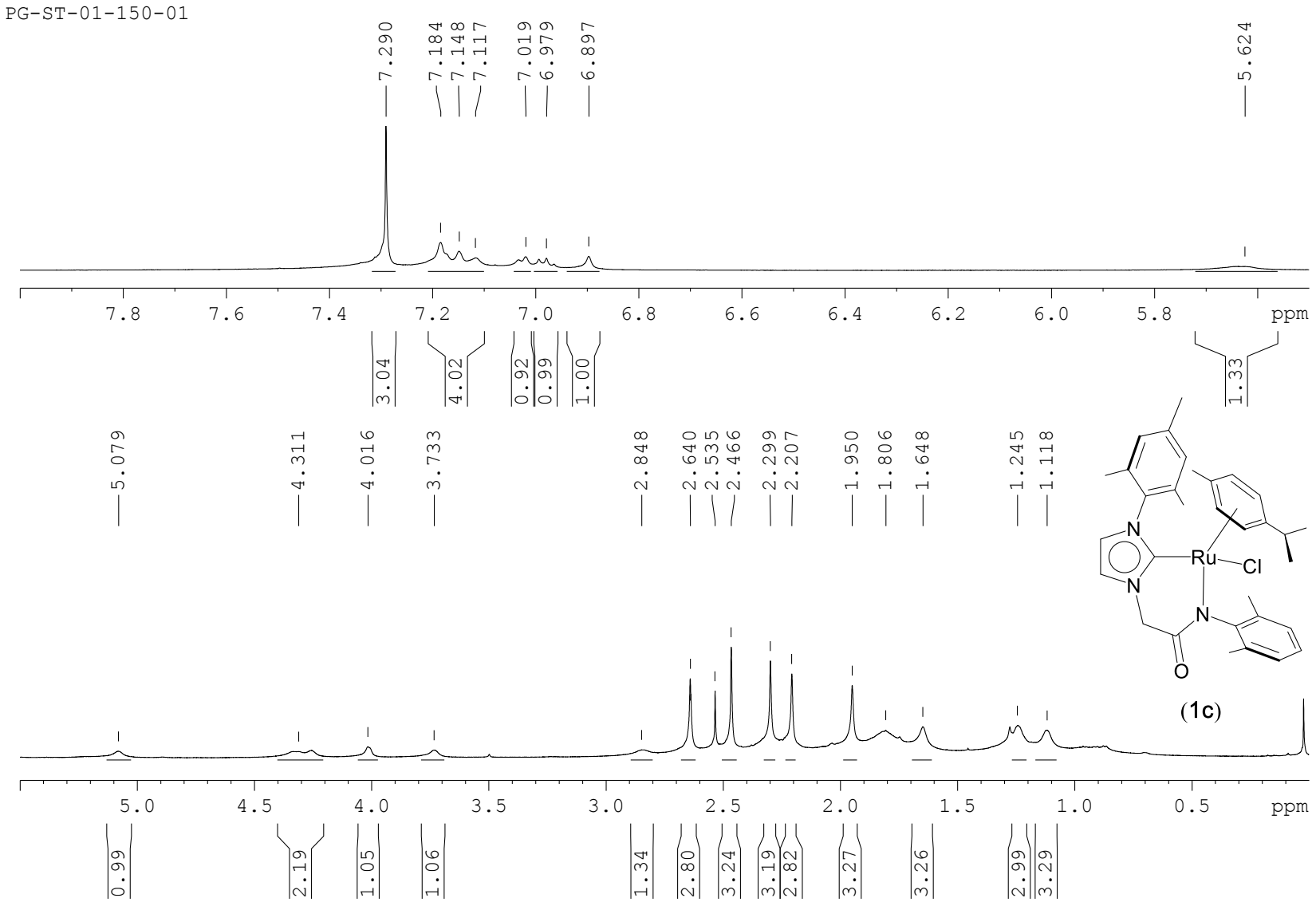


Figure S22. Expanded ^{1}H NMR spectrum of **1c** in $CDCl_3$.

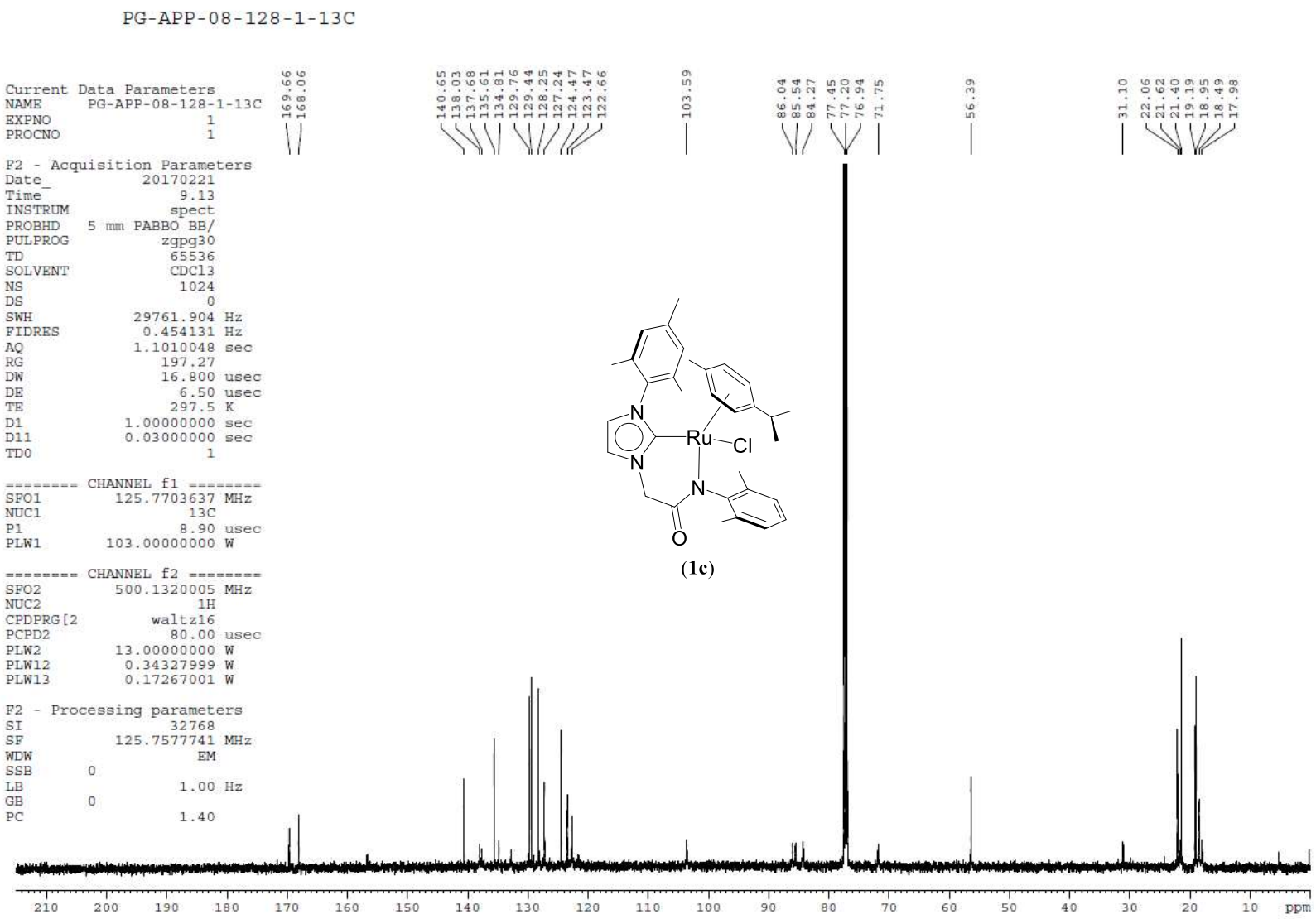
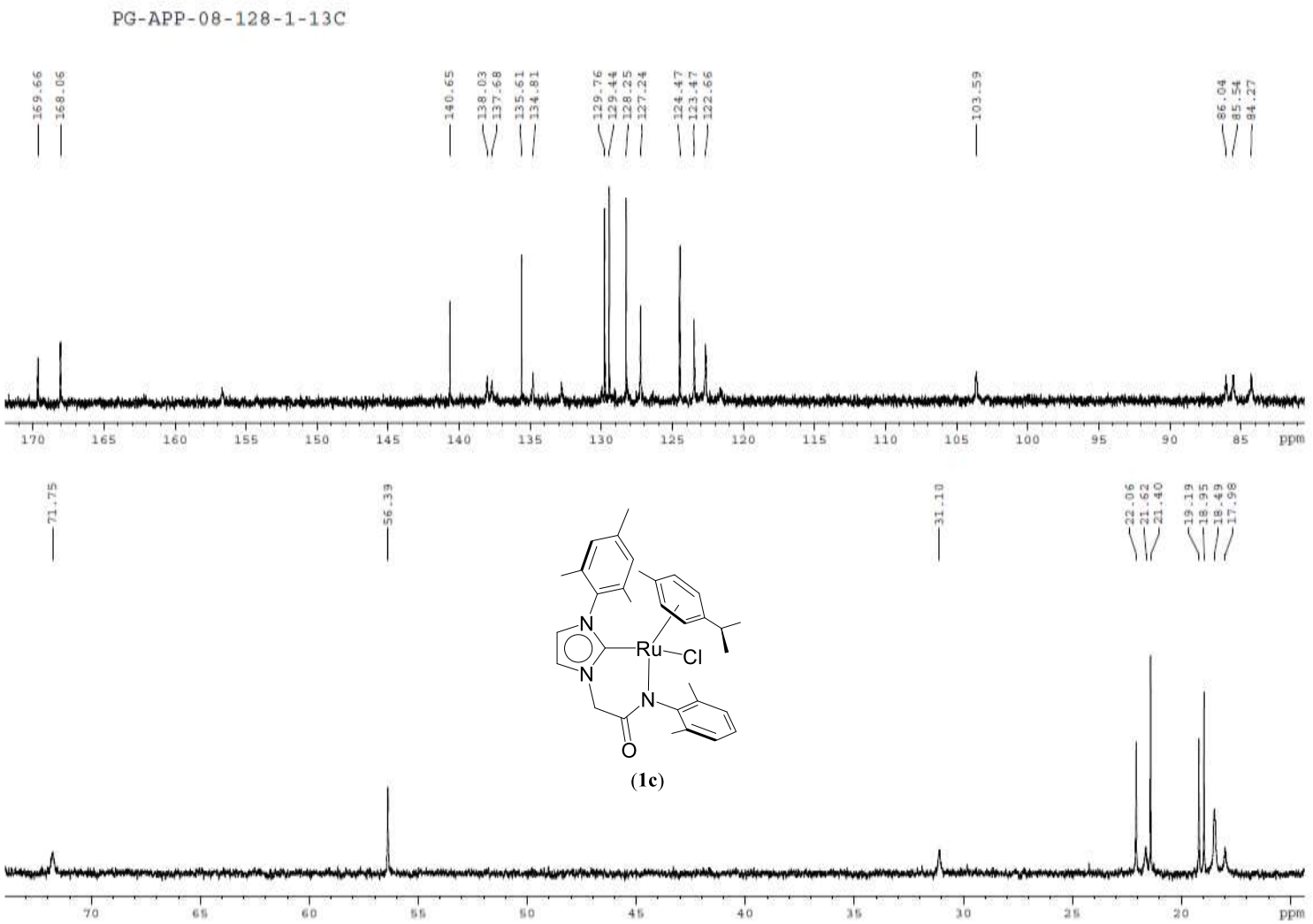


Figure S23. $^{13}C\{^{1}H\}$ NMR spectrum of **1c** in $CDCl_3$.



(**1c**)

Figure S24. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of **1c** in $CDCl_3$.

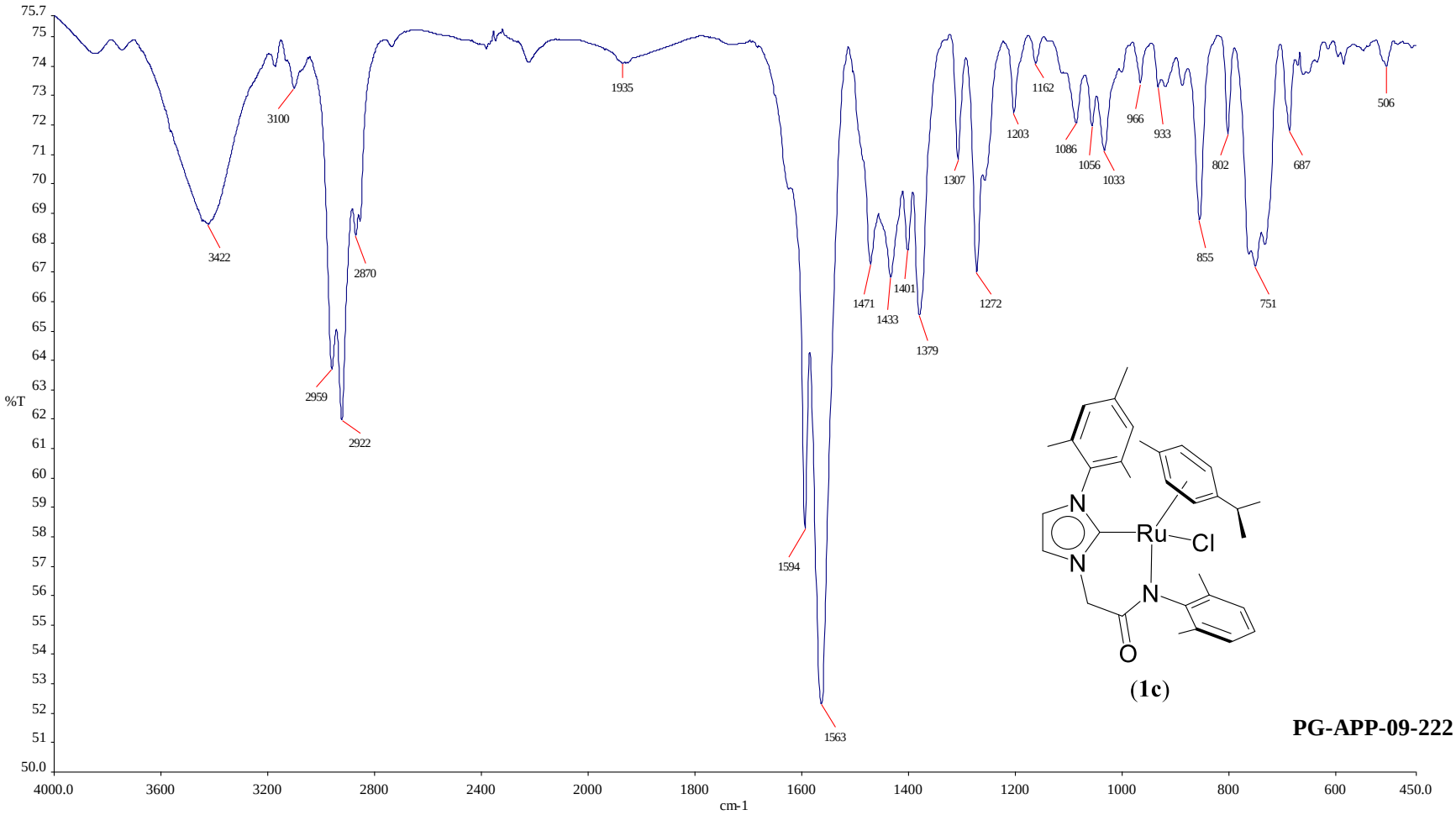
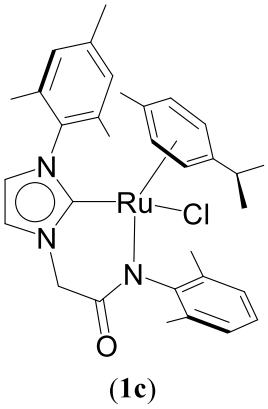


Figure S25. IR spectrum of **1c** in KBr.



(**1c**)

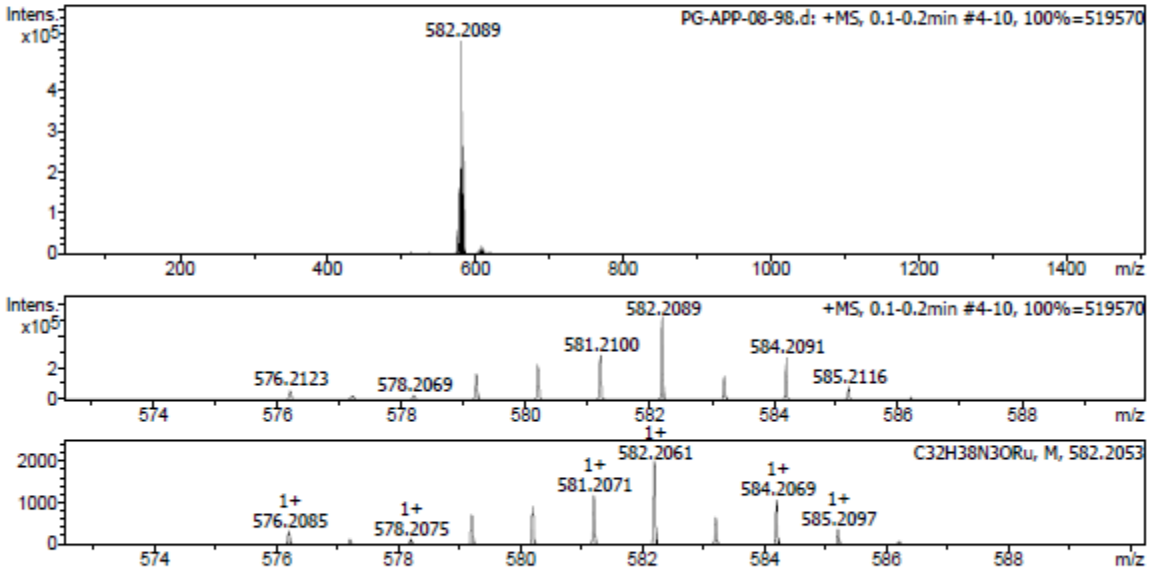
DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

Analysis Name	D:\Data\FEB-2017\PG-APP-08-98.d	Acquisition Date	2/23/2017 3:59:06 PM
Method	Tune_pos_NAF-1500.m	Operator	PG APP IN
Sample Name	PG-APP-08-98	Instrument	maXis impact 282001.00081
Comment	C32H38N3ORuCl		

Acquisition Parameter

Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.5 Bar
Focus	Active	Set Capillary	3700 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	2100.0 Vpp	Set Divert Valve	Source



Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e⁻ Conf	N-Rule
582.2089	1	C32H38N3ORu	582.2062	4.6	30.4	1	100.00	16.0	odd	-

Figure S26. High Resolution Mass Spectrometry (HRMS) data of **1c**.

Document: SP-29-01-2021 (varioMICRO) from: 30-01-2021 03:34:43

SP18022016
varioMICRO CHNS
serial number: 15154051

Graphic report

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	S Area	N [%]	C [%]	H [%]	S [%]	Date	Time
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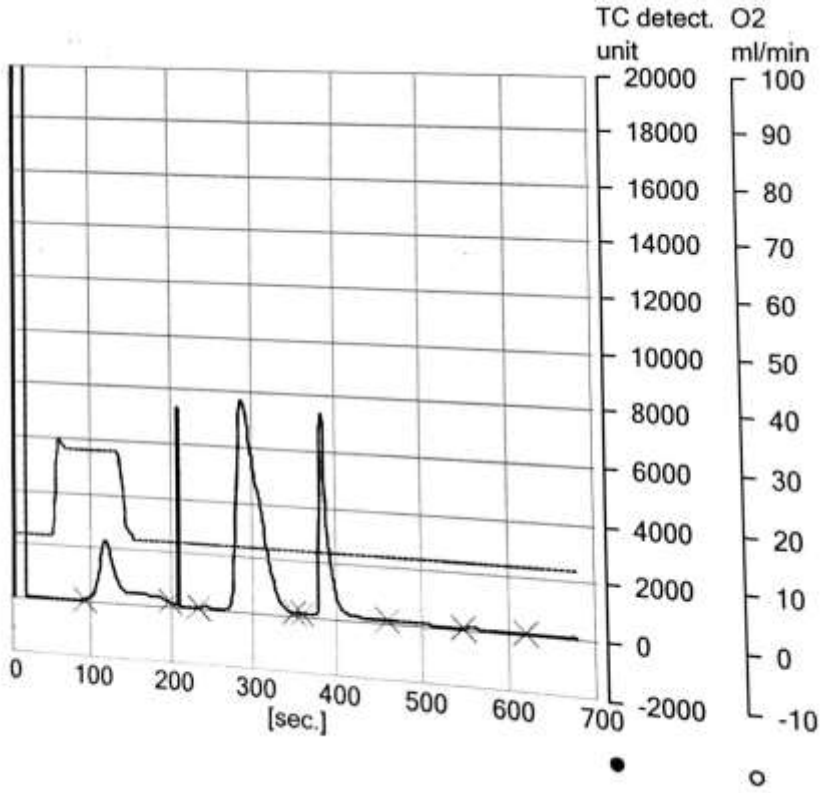
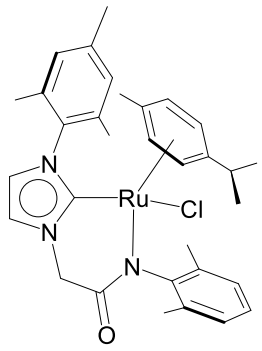




(1c)

Figure S27. Elemental analysis data of **1c**.

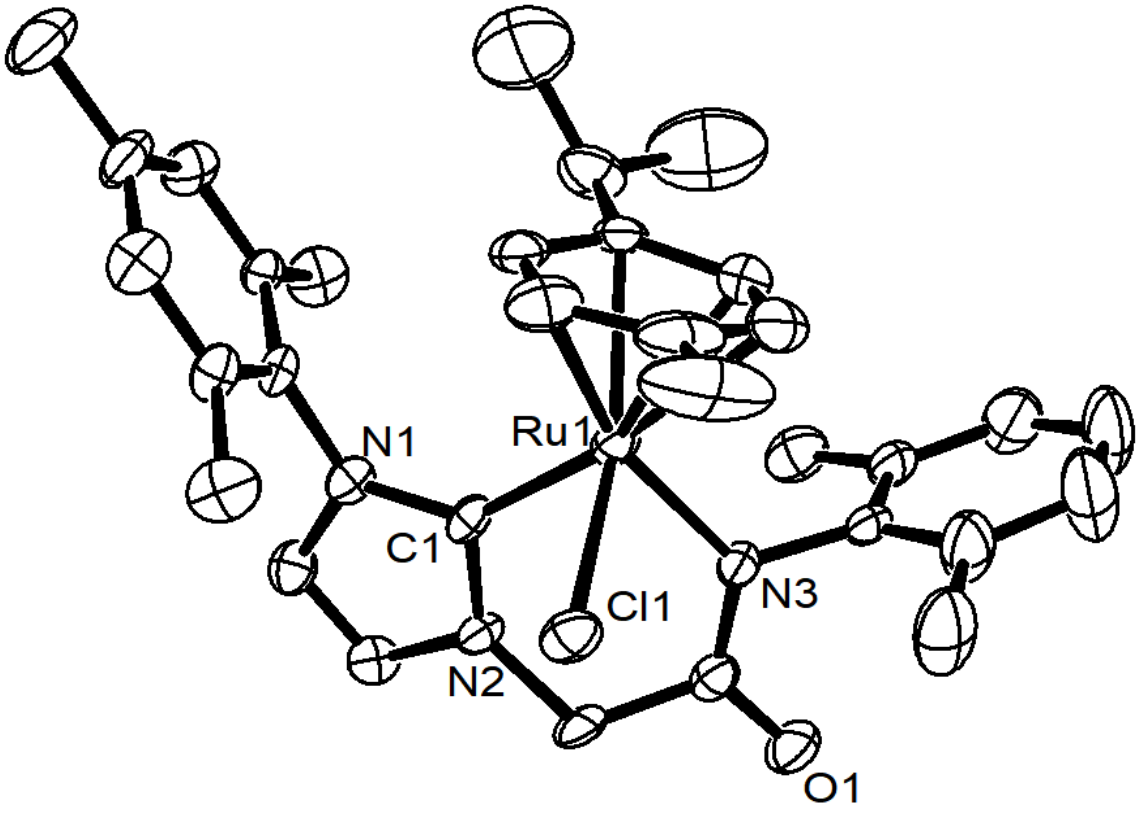


Figure S28. ORTEP of **1c** with thermal ellipsoids shown at the 50 % probability level. Hydrogen atoms were omitted for clarity. Selected bond lengths (Å) and angles (°): Ru(1)−C(1) 2.087(5), Ru(1)−Cl(1) 2.4299(14), Ru(1)−N(3) 2.153(4), C(1)−N(1) 1.367(6), C(1)−N(2) 1.366(6), C(14)−N(3) 1.330(7), C(1)−Ru(1)−Cl(1) 84.22(15), Cl(1)−Ru(1)−N(3) 87.64(12), N(3)−Ru(1)−C(1) 84.94(19), N(1)−C(1)−Ru(1) 134.6(4), N(2)−C(1)−Ru(1) 122.3(4), N(1)−C(1)−N(2) 103.0(4).

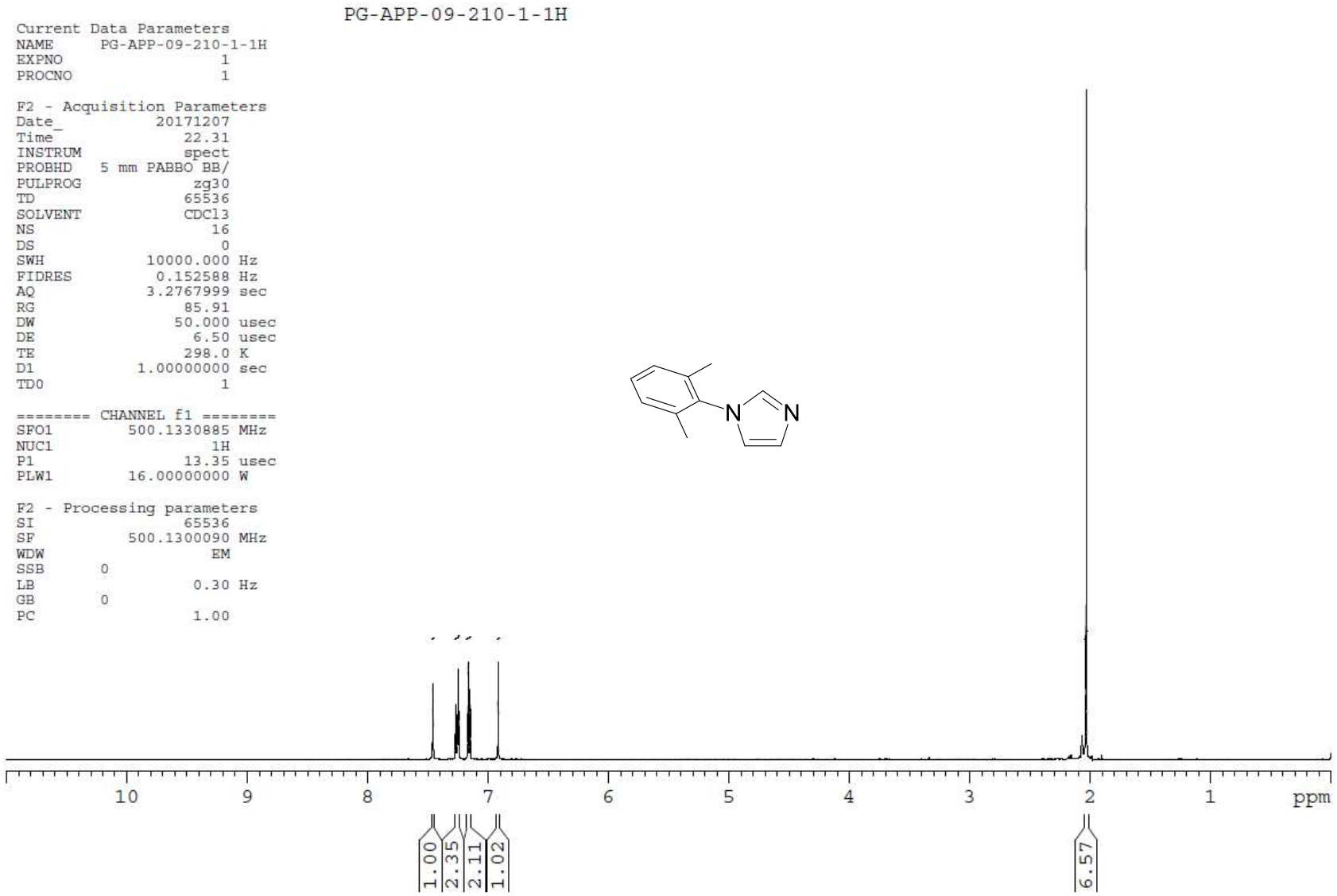


Figure S29. ^{1}H NMR spectrum of 1-(2,6-Me$_2$-phenyl)imidazole in $CDCl_3$.

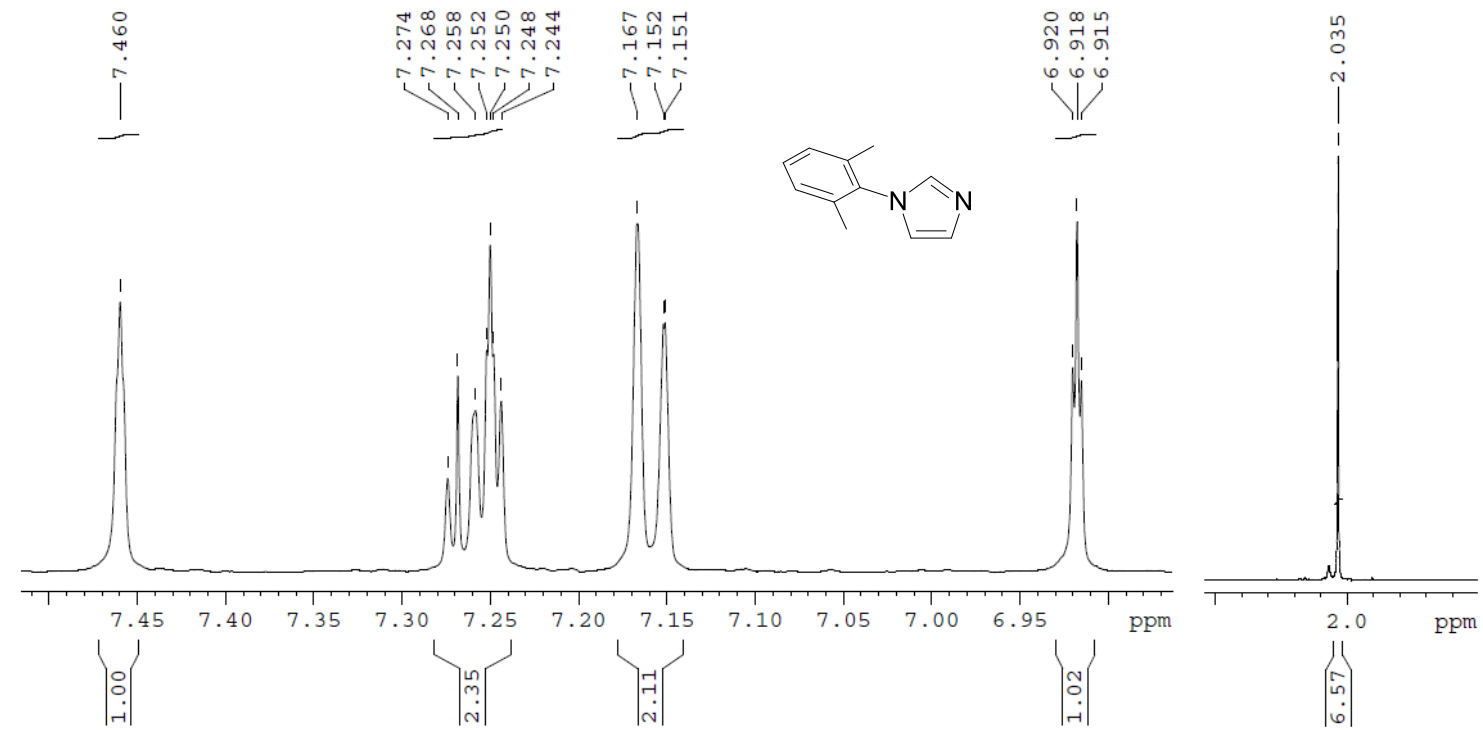


Figure S30. Expanded ^{1}H NMR spectrum of 1-(2,6-Me_2-phenyl)imidazole in $CDCl_3$.

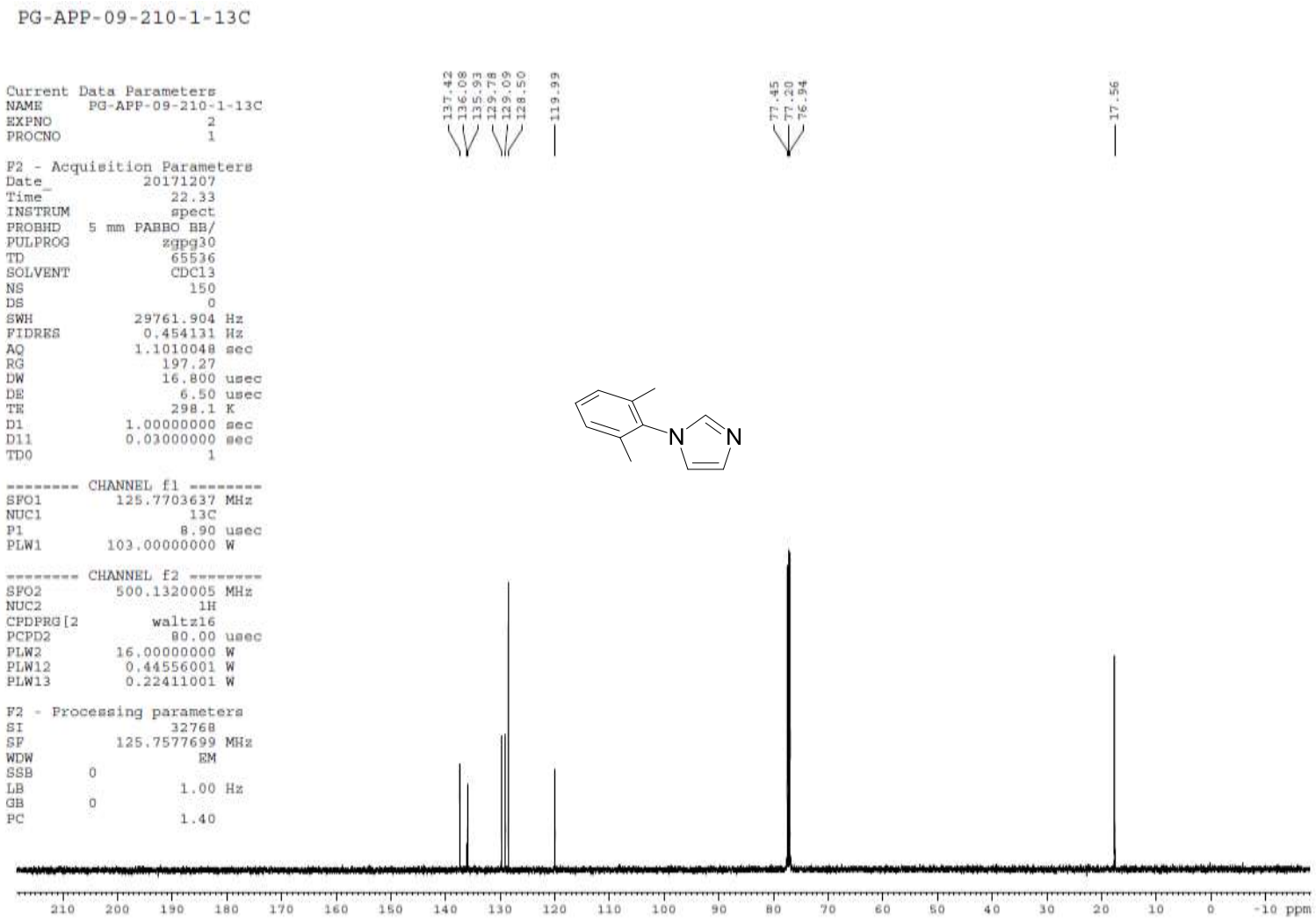


Figure S31. $^{13}C\{^{1}H\}$ NMR spectrum of 1-(2,6-Me_2-phenyl)imidazole in $CDCl_3$.

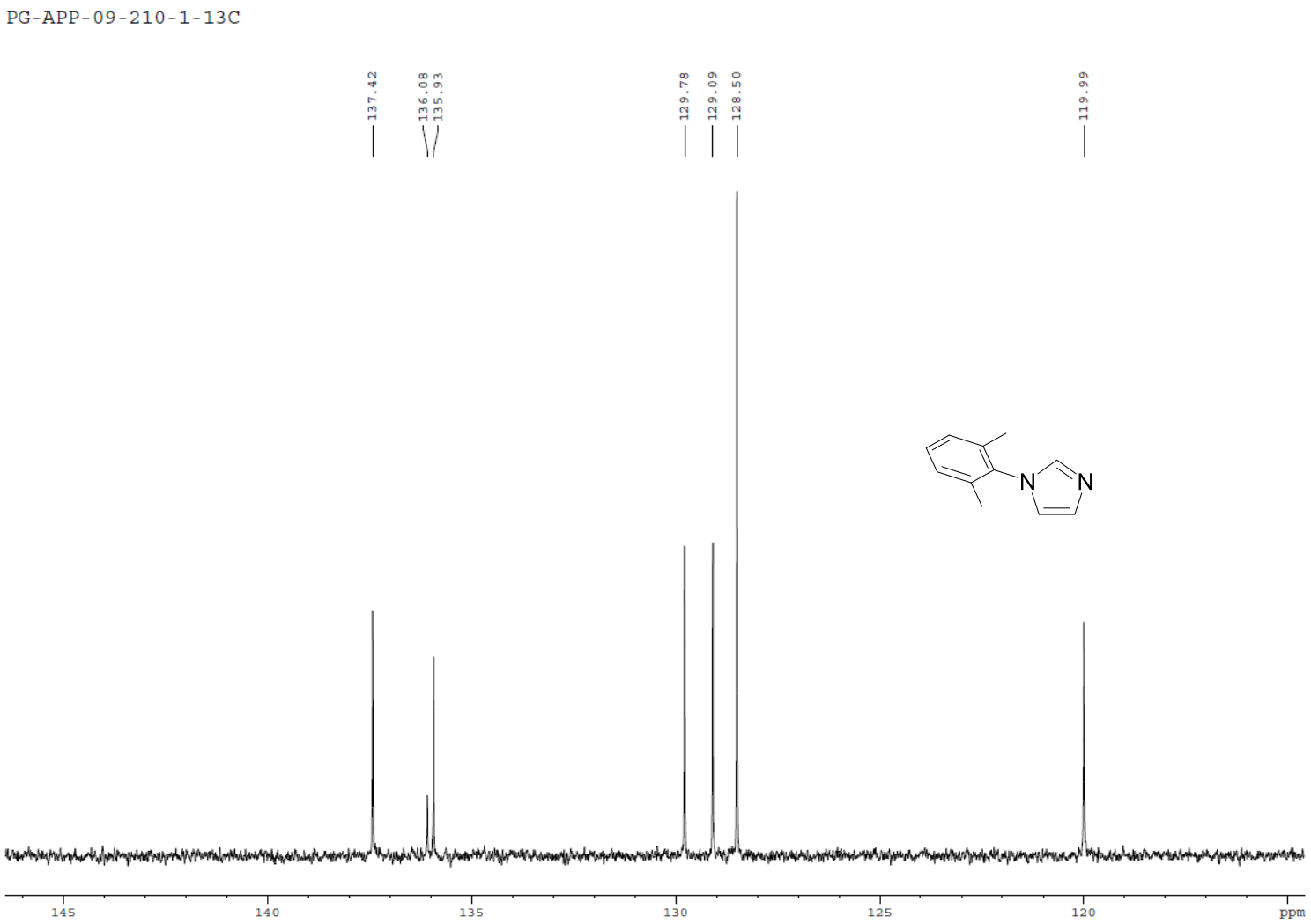


Figure S32. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of 1-(2,6-Me_2-phenyl)imidazole in $CDCl_3$.

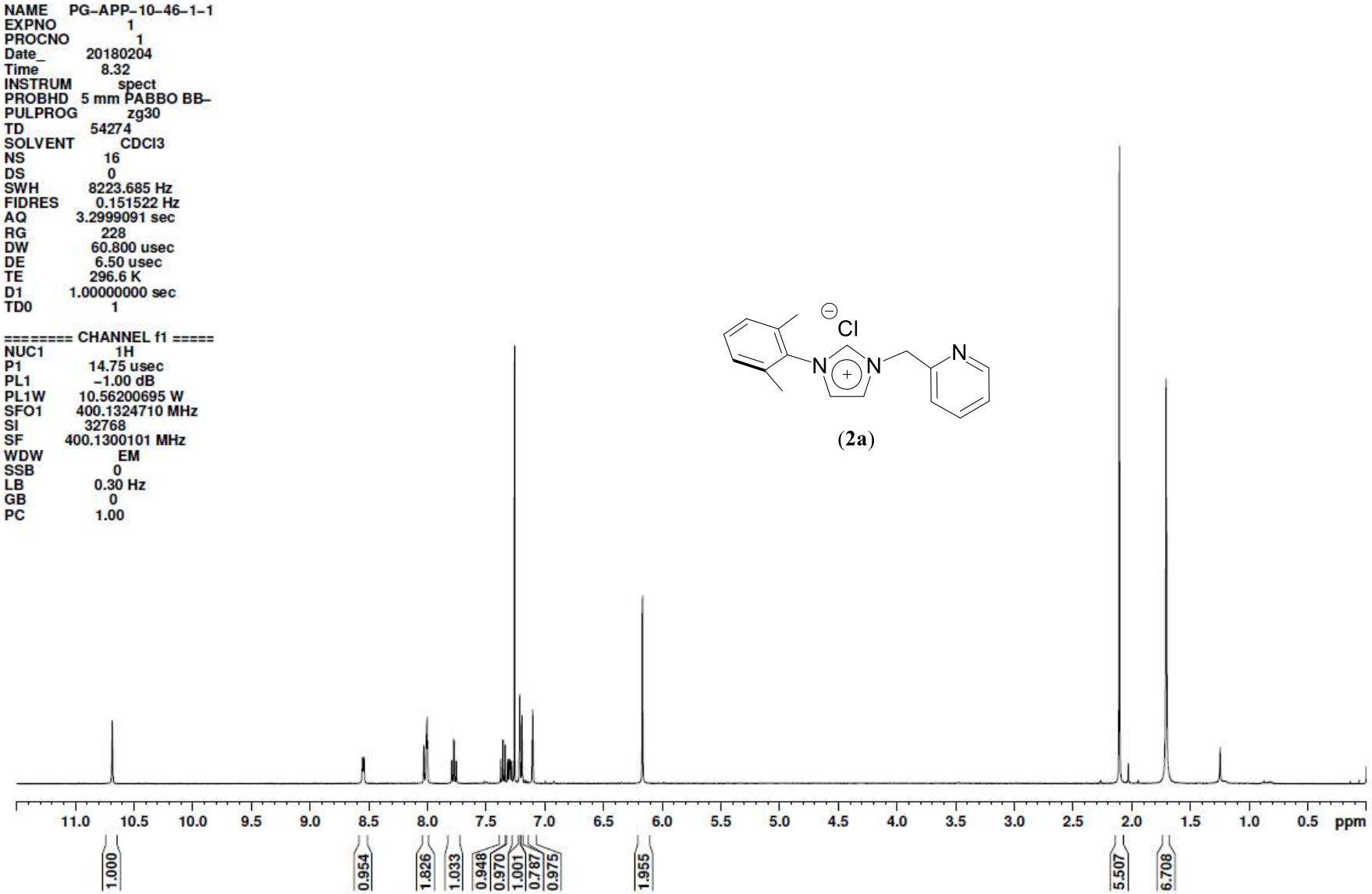


Figure S33. ^{1}H NMR spectrum of **2a** in $CDCl_3$.

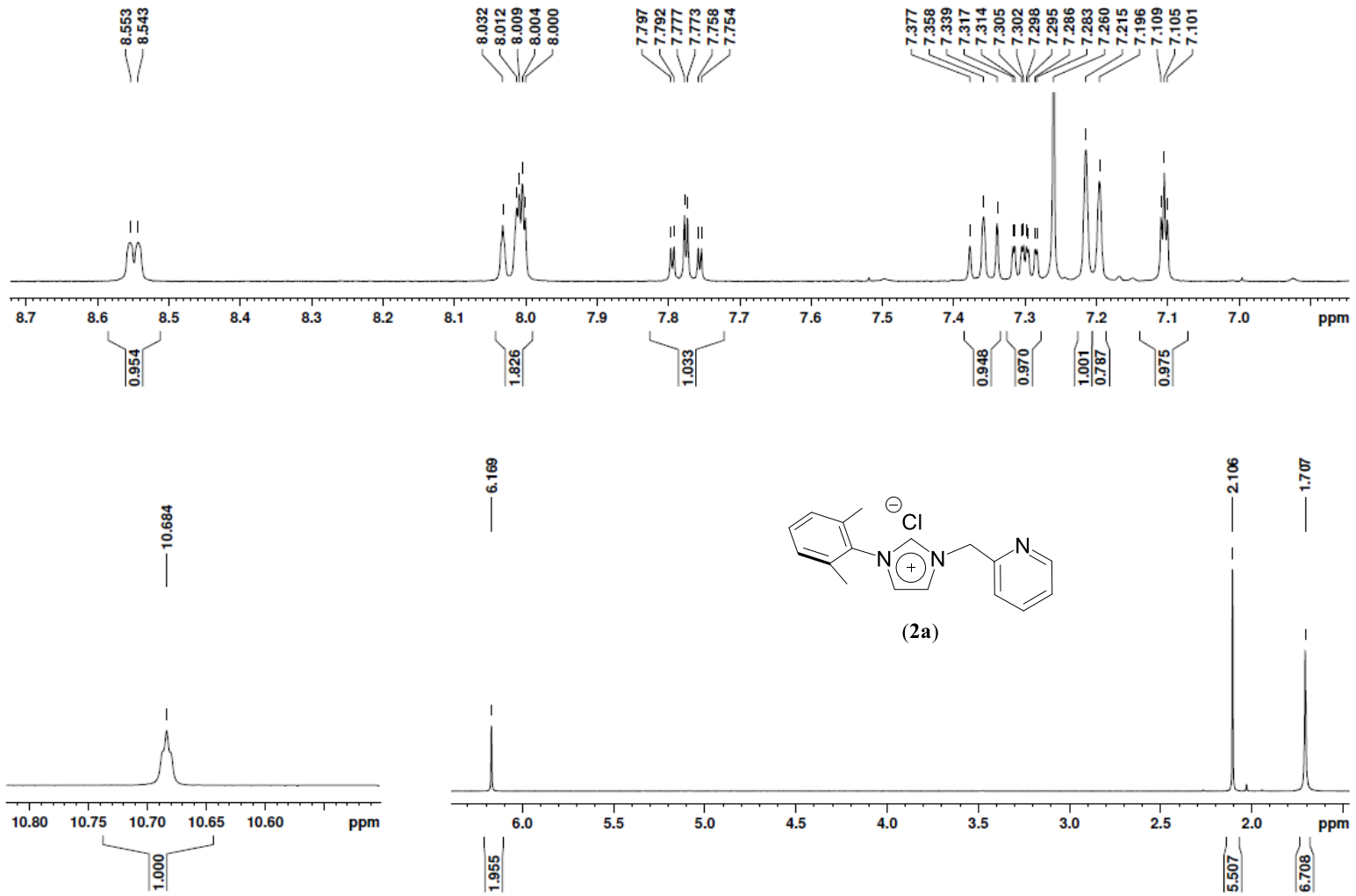


Figure S34. Expanded ^{1}H NMR spectrum of **2a** in $CDCl_3$.

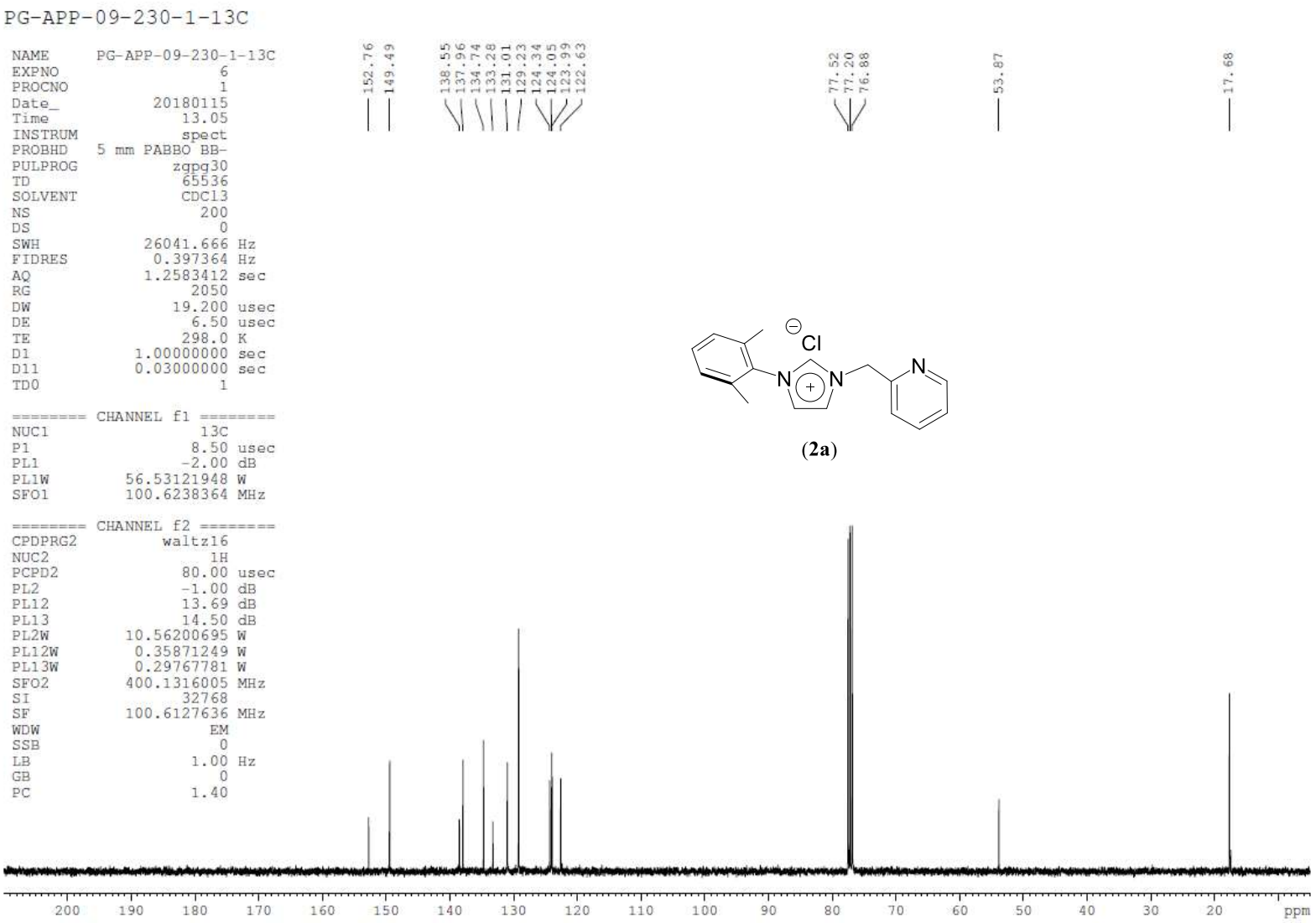


Figure S35. $^{13}C\{^{1}H\}$ NMR spectrum of **2a** in $CDCl_3$.

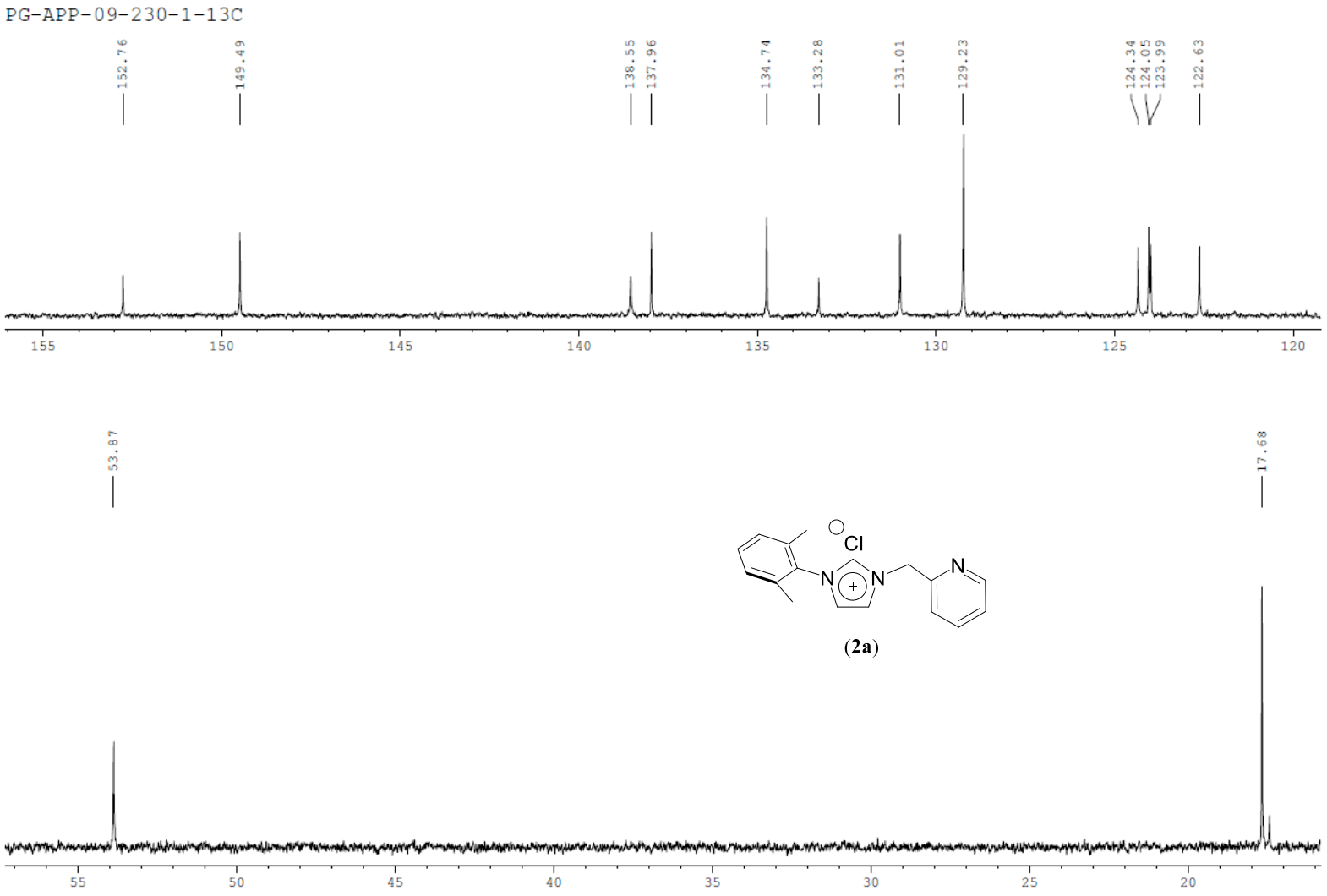


Figure S36. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of **2a** in $CDCl_3$.

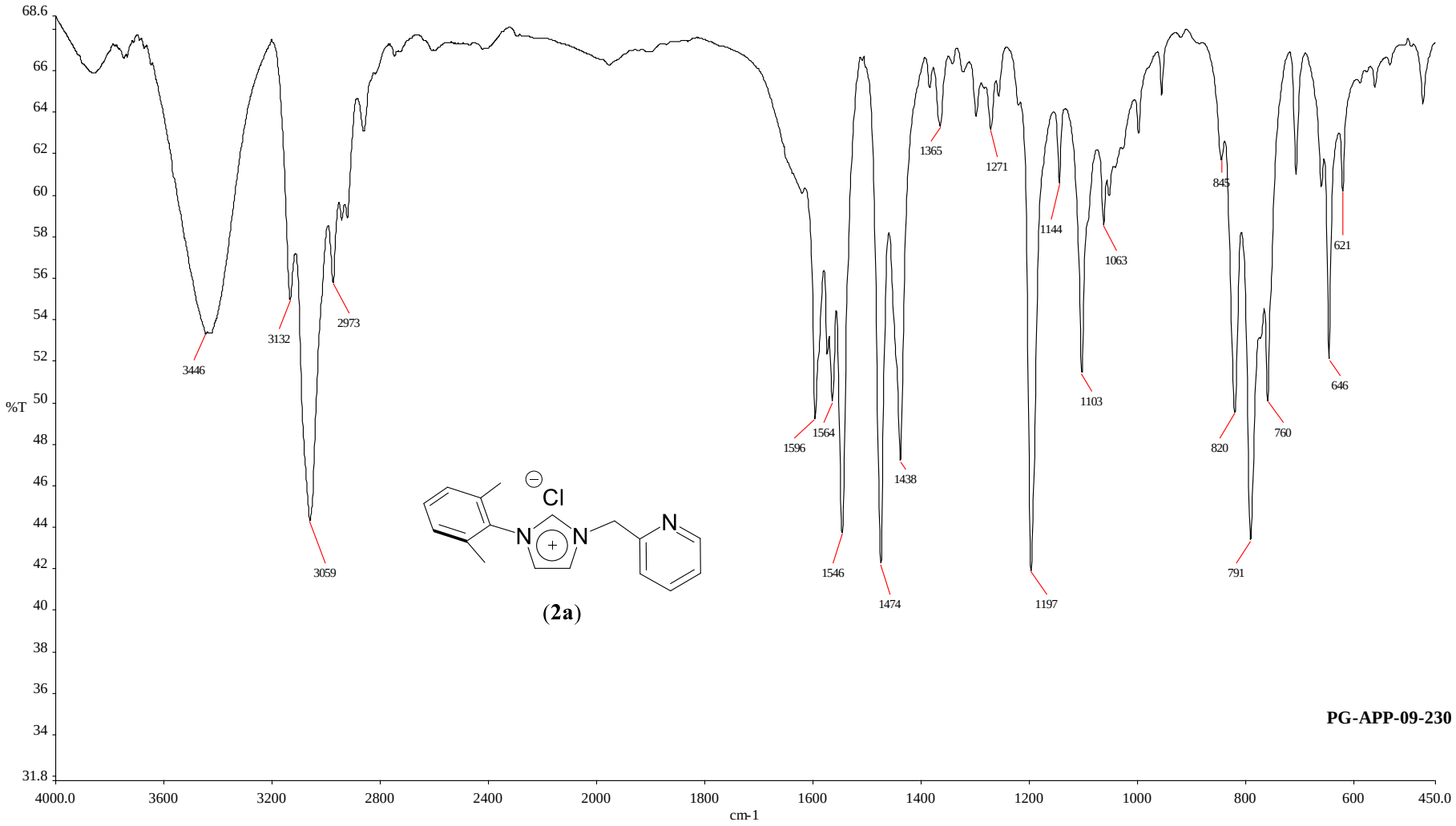
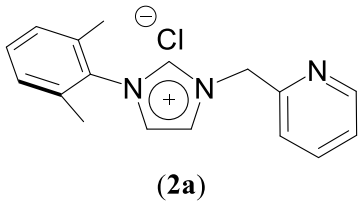


Figure S37. IR spectrum of **2a** in KBr.



(**2a**)

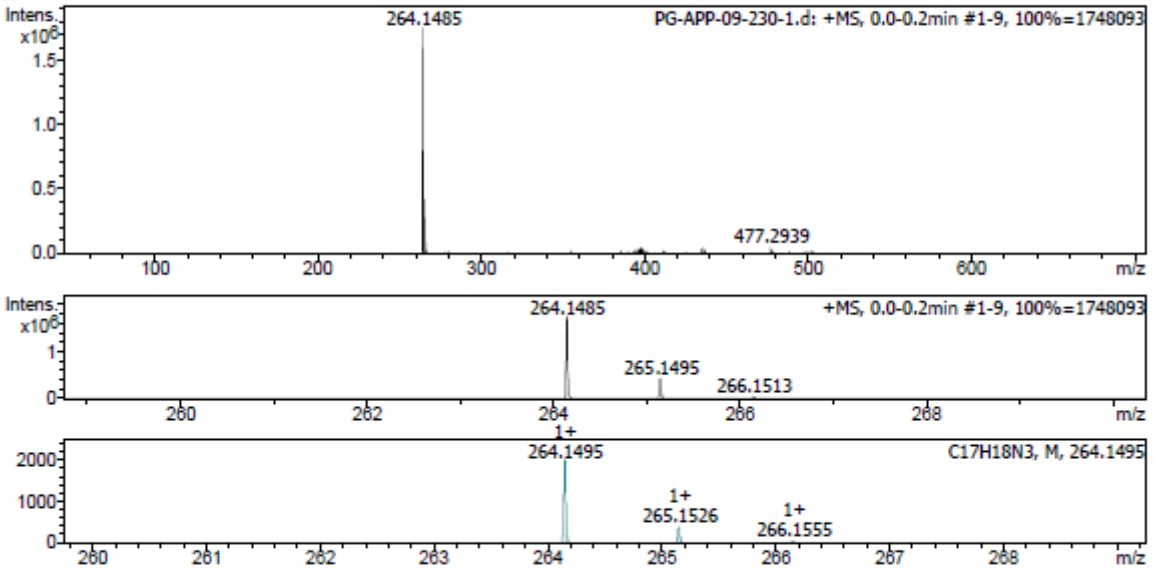
DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

Analysis Name	D:\Data\FEB-2018\PG-APP-09-230-1.d	Acquisition Date	2/1/2018 1:01:52 PM
Method	Tune_pos_NAICSI-1000A.m	Operator	PG APP IN
Sample Name	PG-APP-09-230-1	Instrument	maXis impact 282001.00081
Comment	C17H18N3Cl		

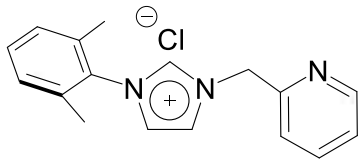
Acquisition Parameter

Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.4 Bar
Focus	Active	Set Capillary	3700 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	500 m/z	Set Collision Cell RF	1500.0 Vpp	Set Divert Valve	Source



Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e⁻ Conf	N-Rule
264.1485	1	C17H18N3	264.1495	3.8	24.5	1	100.00	10.5	even	ok

Figure S38. High Resolution Mass Spectrometry (HRMS) data of **2a**.



(**2a**)

Eager 300 Report

Page: 1 Sample: PG-APP-09-230-2 (PG-APP-09-230-2)

Method Name : PGAPP29012018R
Method File : D:\CHNS2018\PGAPP29012018R.mth
Chromatogram : PG-APP-09-230-2
Operator ID : Prakash Company Name : C.E. Instruments
Analysed : 01/29/2018 14:46 Printed : 2/22/2018 18:23
Sample ID : PG-APP-09-230-2 (# 16) Instrument N. : Instrument #1
Analysis Type : UnkNown (Area) Sample weight : .987

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	21525	FU		0.0000
2	0.0000	6	115783	FU		0.0000
Nitrogen	13.7310	41	138189	RS	12.522860	.101966E+07
Carbon	67.6290	64	1730515	RS	1.000000	.259254E+07
Hydrogen	5.8837	181	370805	RS	4.666914	.638521E+07
Totals	87.2437		2376817			

Figure S39. Elemental analysis data of **2a**.

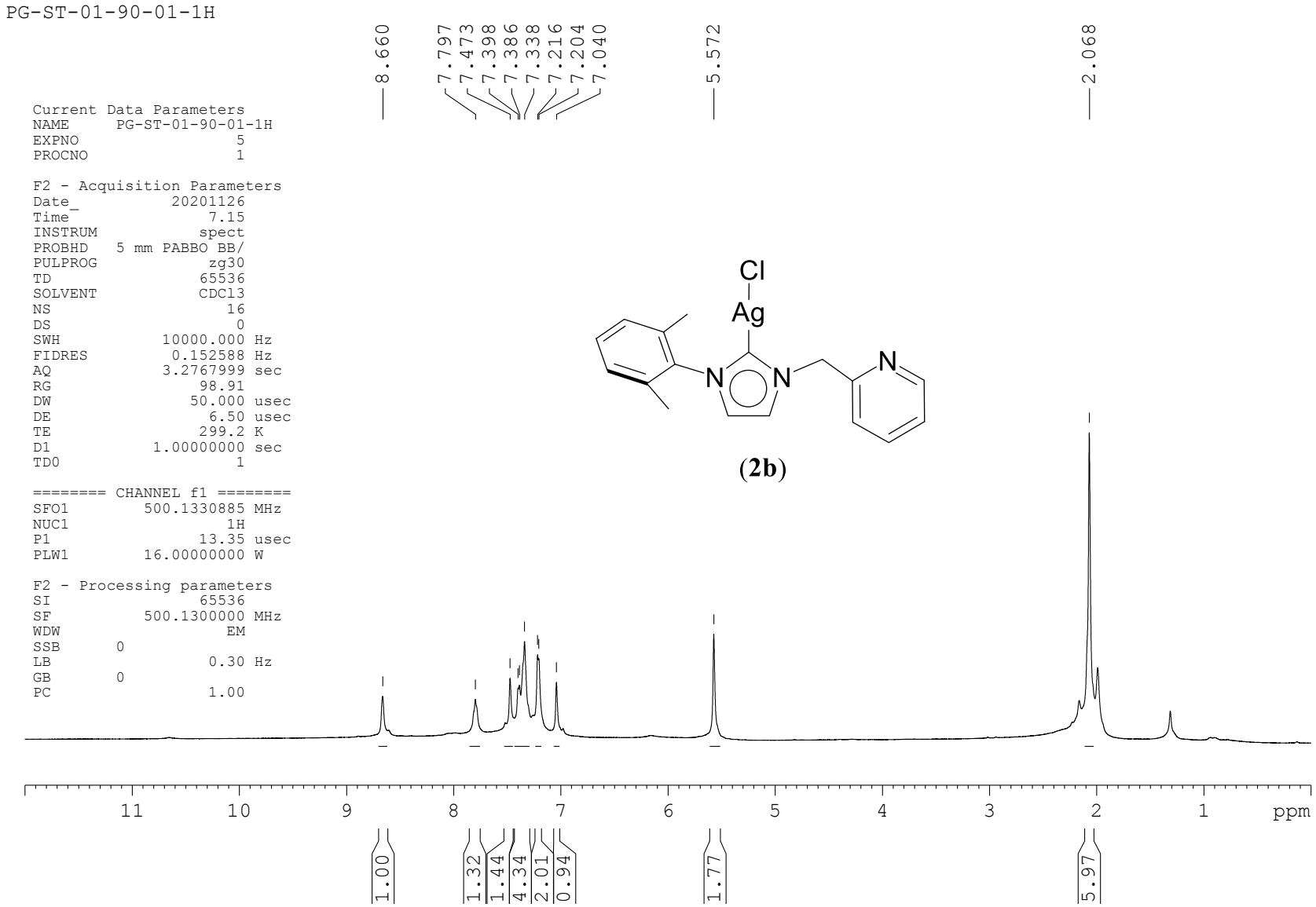


Figure S40. ^{1}H NMR spectrum of **2b** in $CDCl_3$.

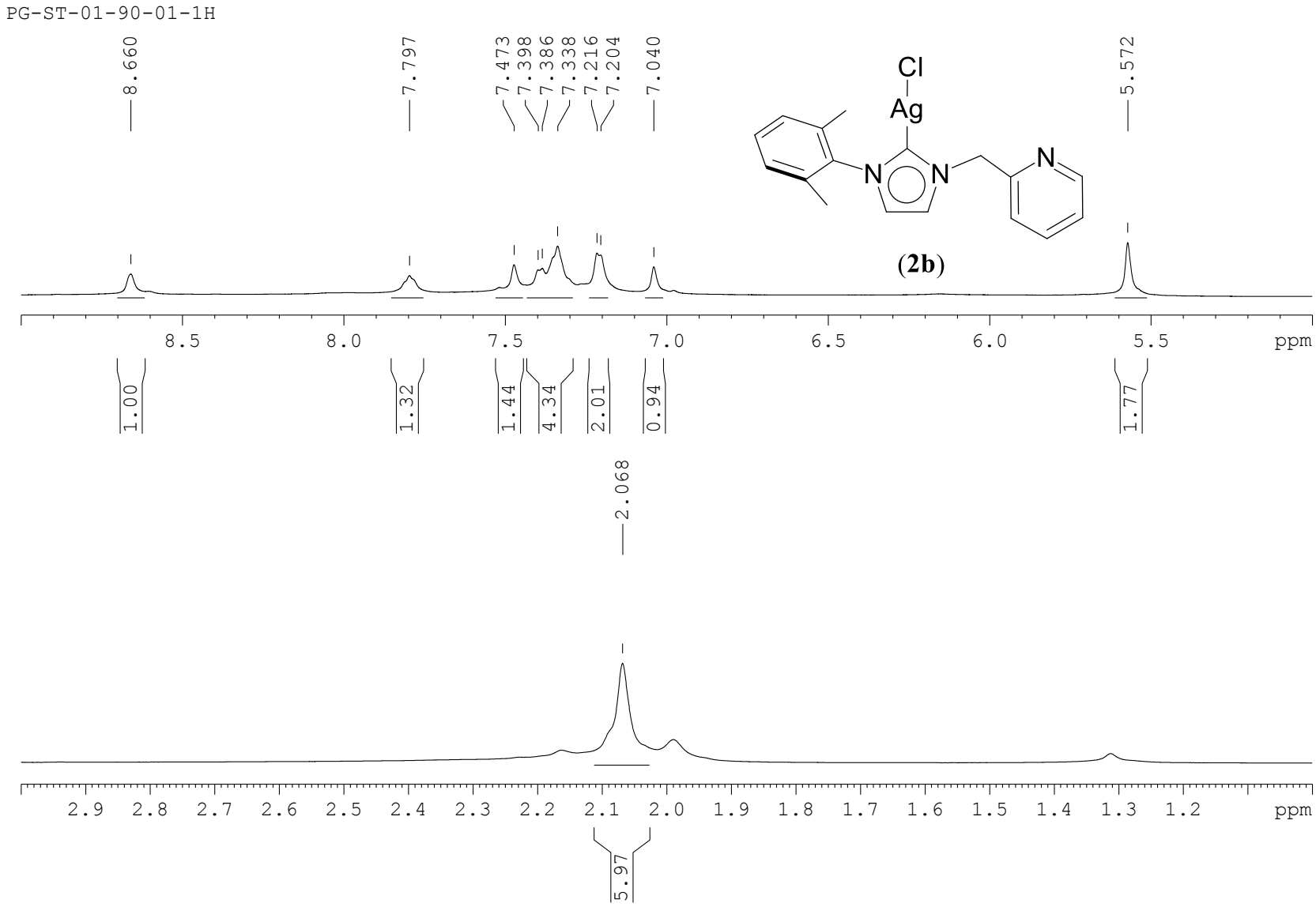


Figure S41. Expanded ^{1}H NMR spectrum of **2b** in $CDCl_3$.

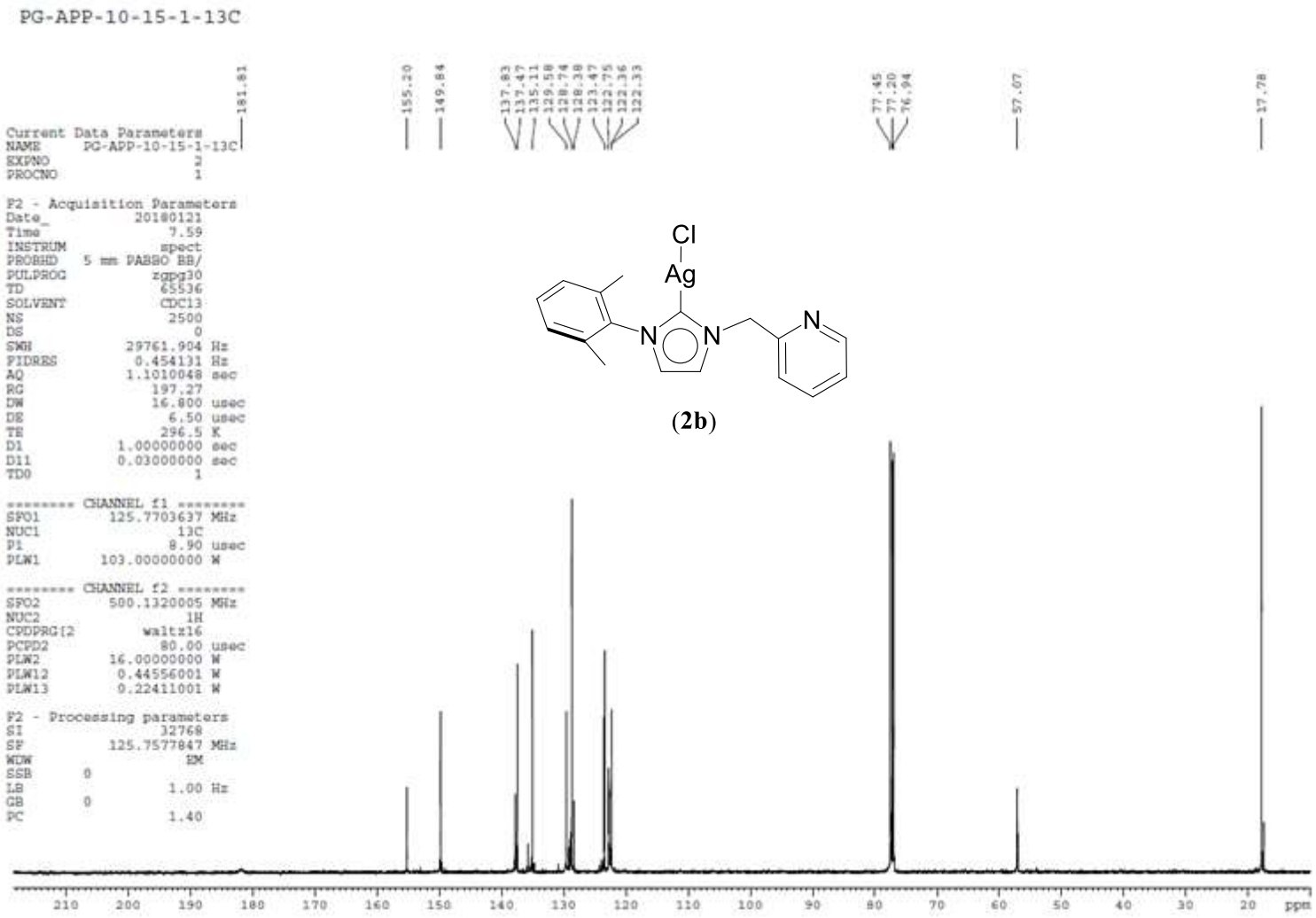


Figure S42. $^{13}C\{^{1}H\}$ NMR spectrum of **2b** in $CDCl_3$.

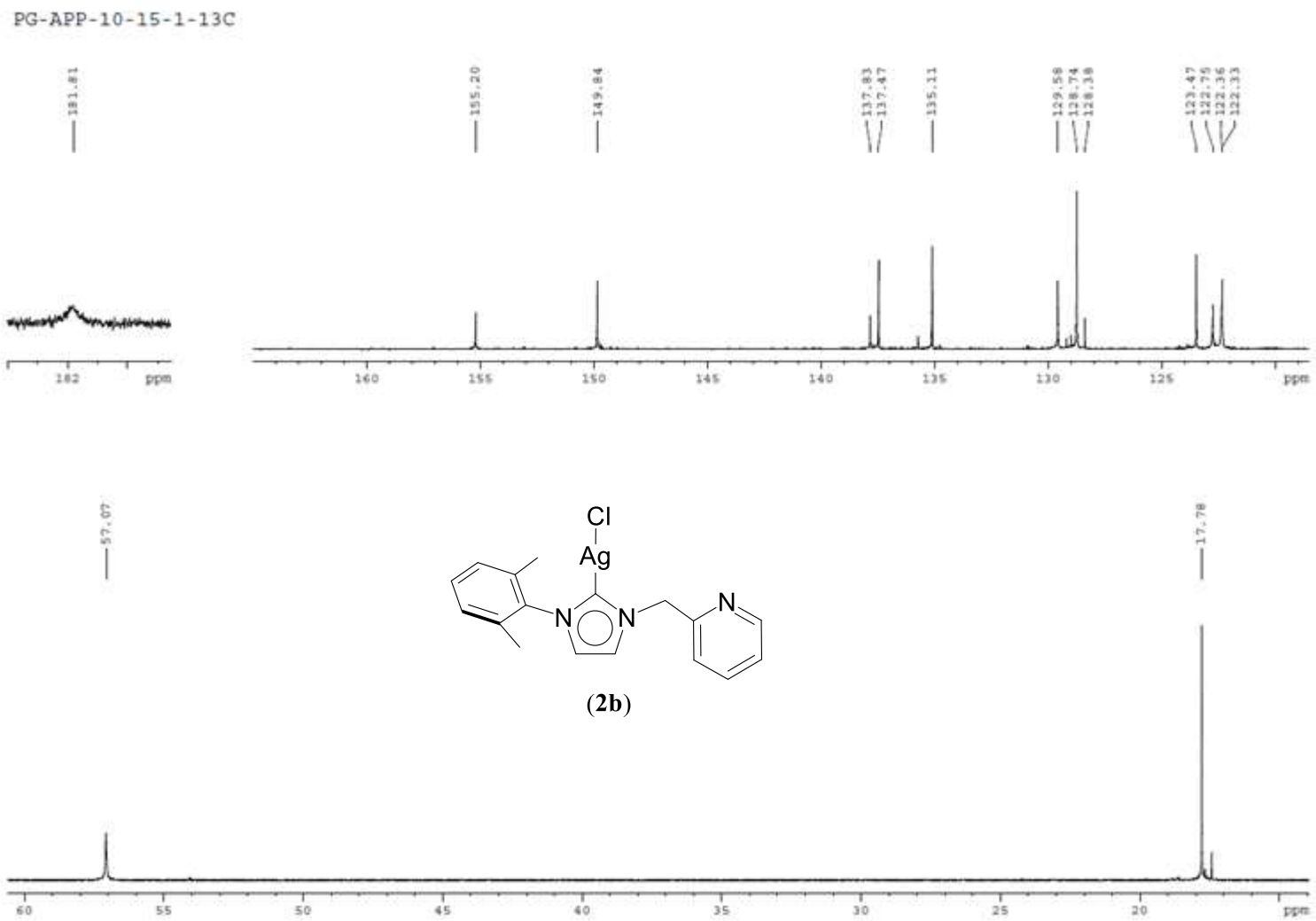


Figure S43. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of **2b** in $CDCl_3$.

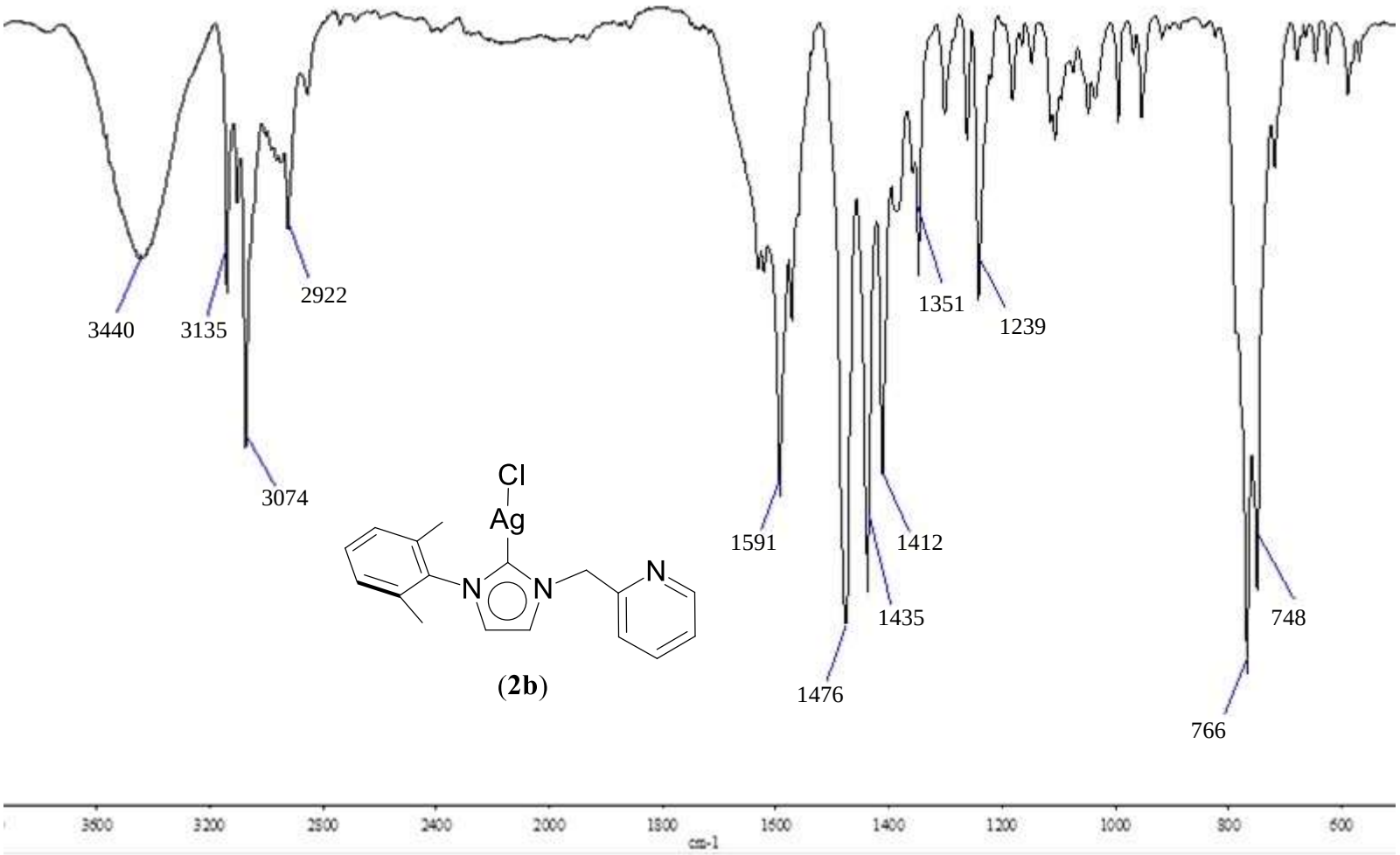
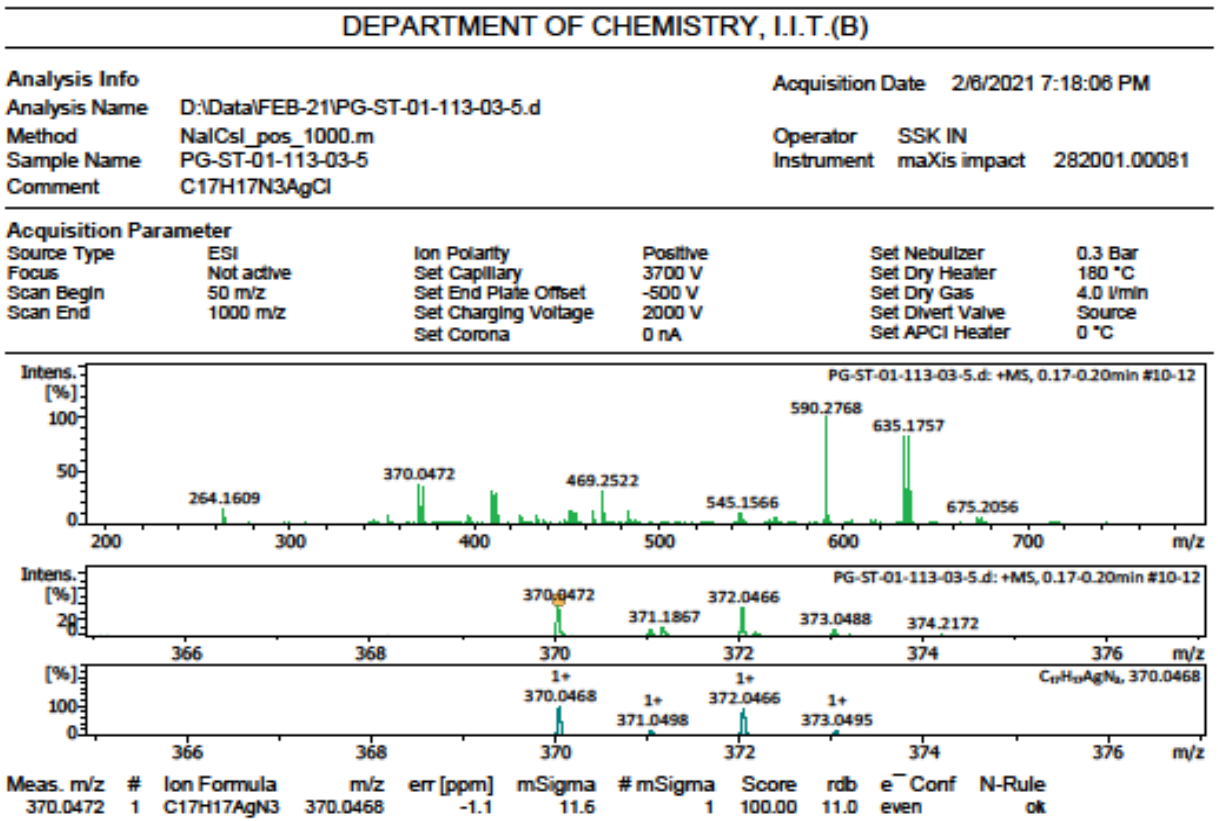
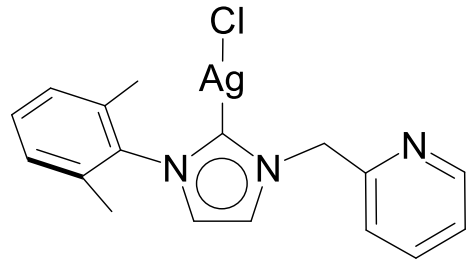


Figure S44. IR spectrum of **2b** in KBr.





(**2b**)

Figure S45. High Resolution Mass Spectrometry (HRMS) data of **2b**.

SP18022016
varioMICRO CHNS
serial number: 15154051

Graphic report

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	N [%]	C [%]	H [%]	Date Time	Info
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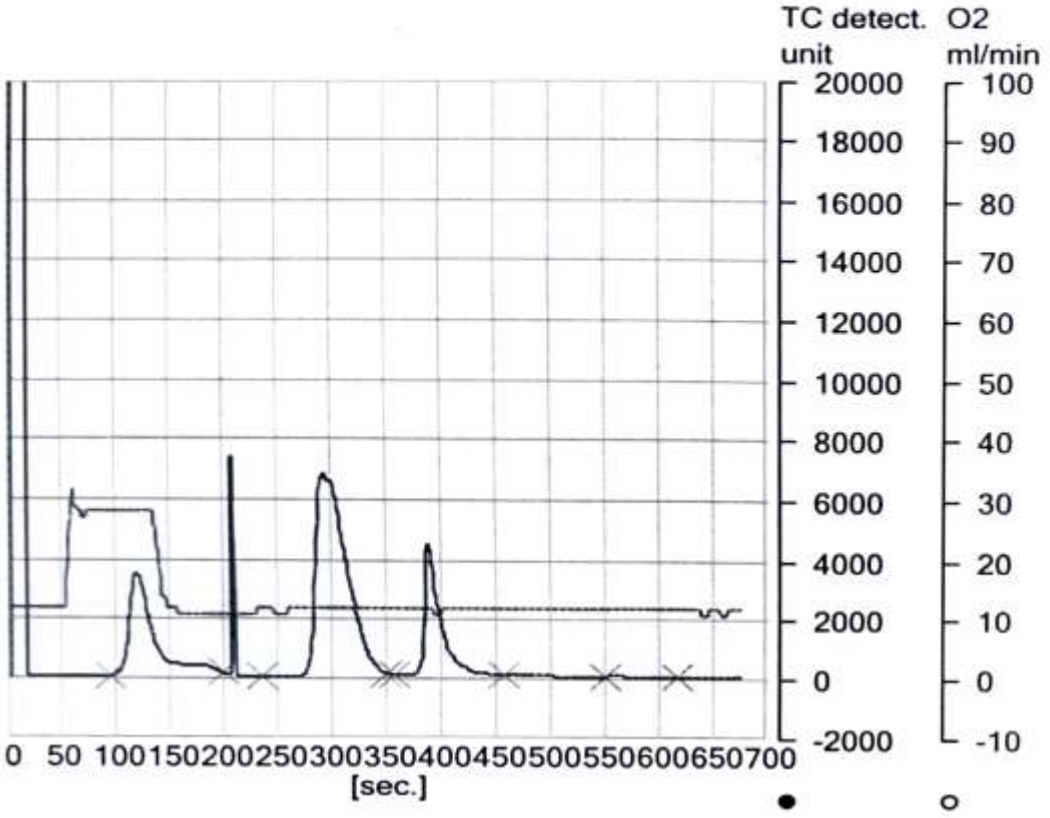
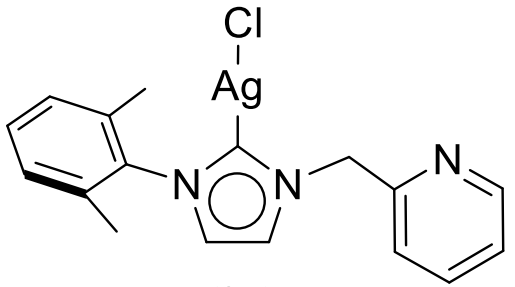




(2b)

Figure S46. Elemental analysis data of **2b**.

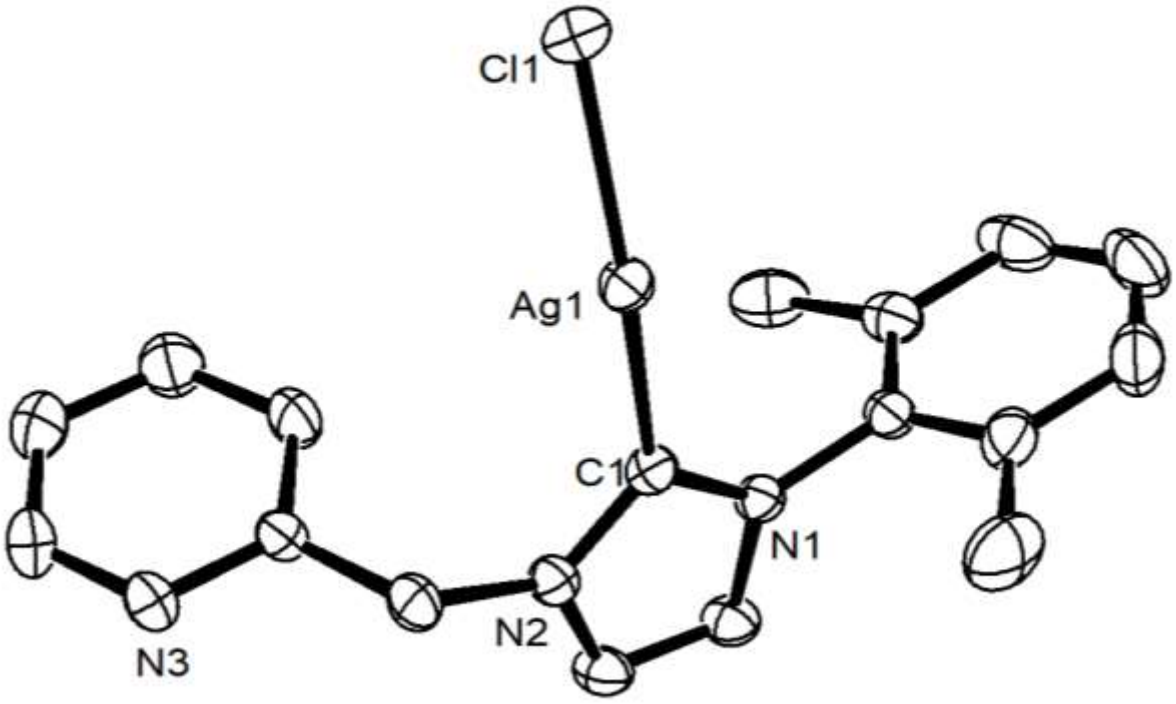


Figure S47. ORTEP of **2b** with thermal ellipsoids shown at the 50 % probability level. Hydrogen atoms were omitted for clarity. Selected bond lengths (Å) and angles (°): Ag(1)−C(1) 2.0813(17), Ag(1)−Cl(1) 2.3684(5), C(1)−N(1) 1.352(2), C(1)−N(2) 1.357(2), C(1)−Ag(1)−Cl(1) 168.09(5), N(1)−C(1)−N(2) 104.05(15), N(1)−C(1)−Ag(1) 129.13(13), N(2)−C(1)−Ag(1) 126.74(13).

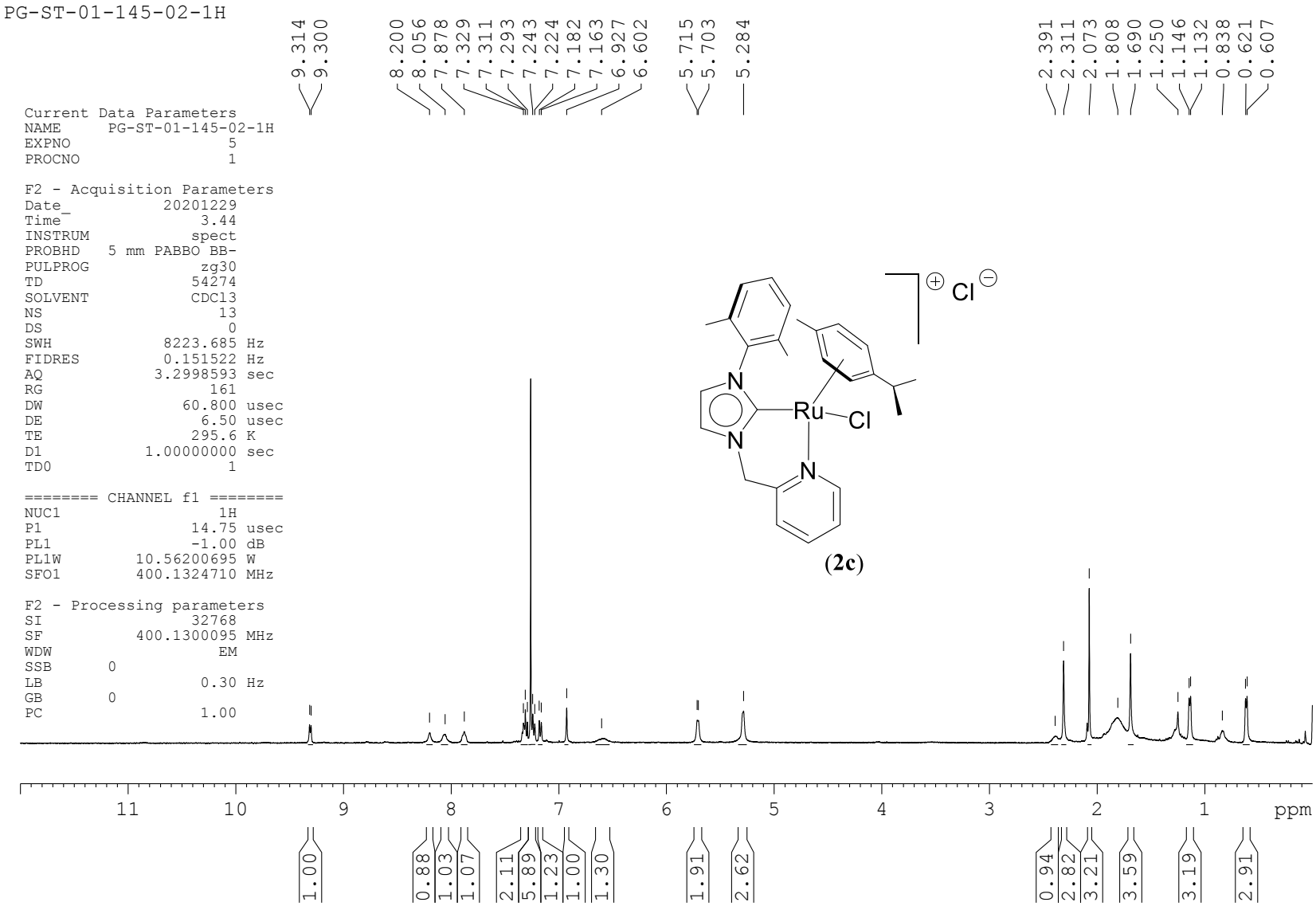


Figure S48. ^{1}H NMR spectrum of **2c** in $CDCl_3$.

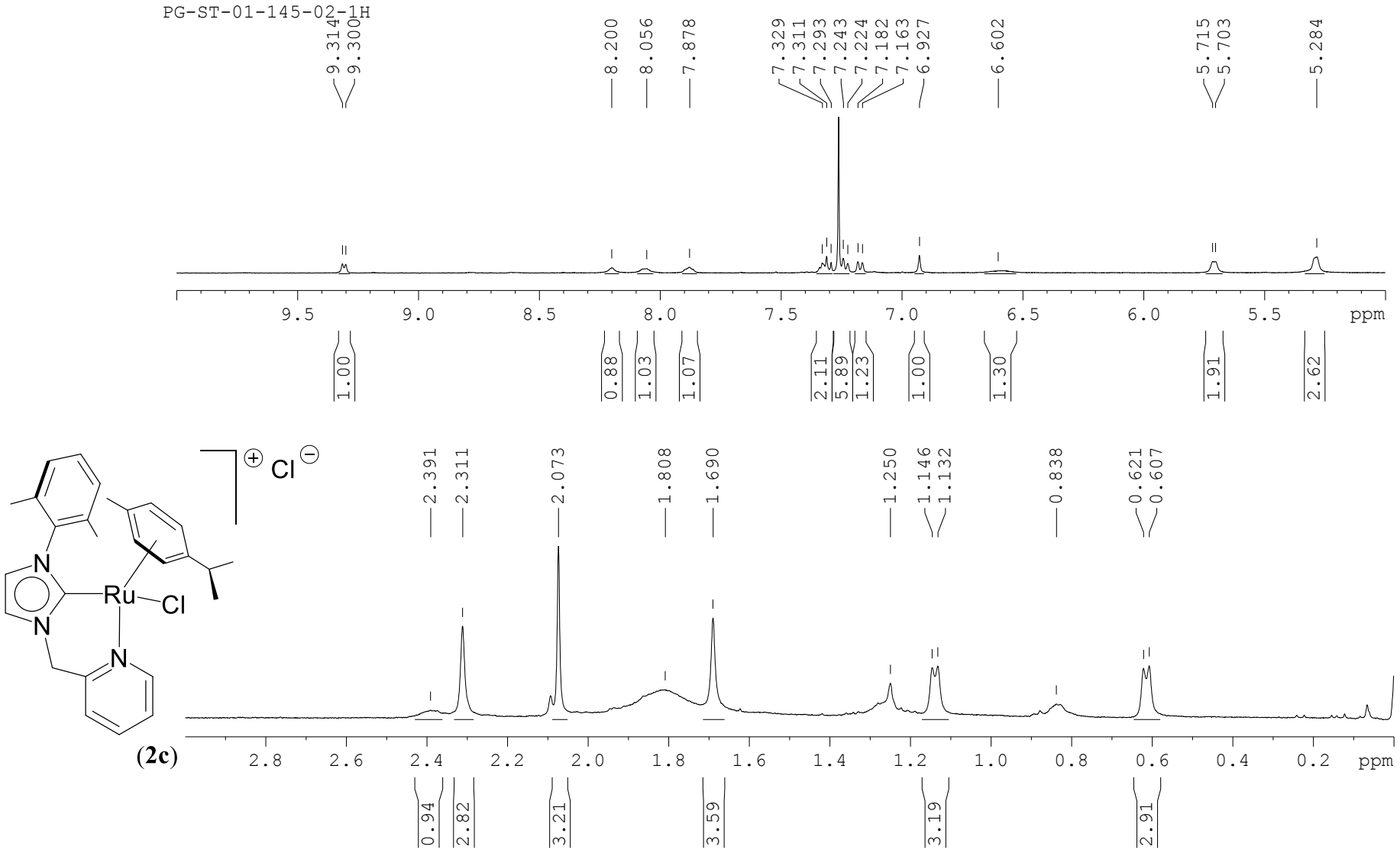


Figure S49. Expanded ^{1}H NMR spectrum of **2c** in $CDCl_3$.

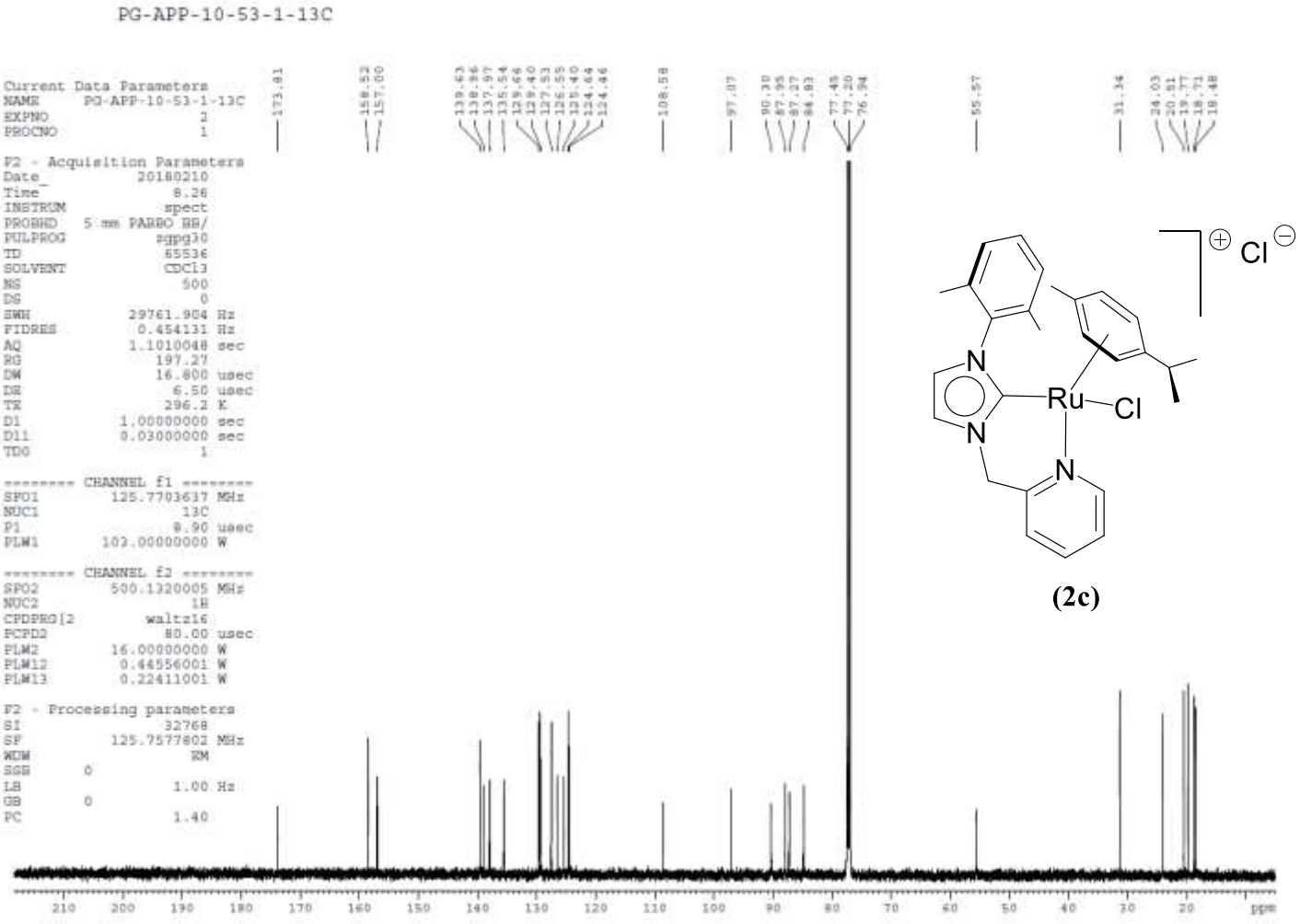


Figure S50. $^{13}C\{^{1}H\}$ NMR spectrum of **2c** in $CDCl_3$.

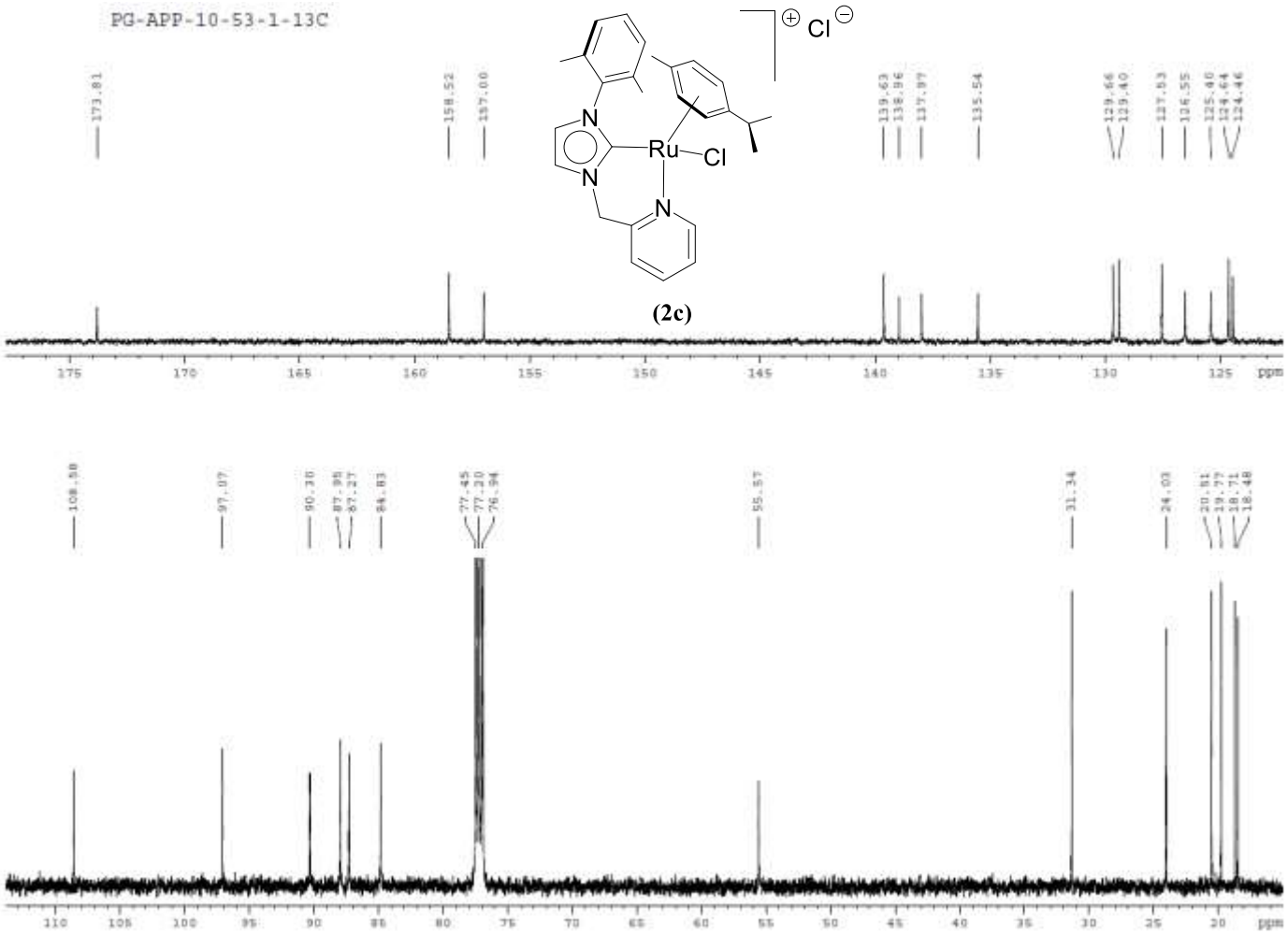


Figure S51. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of **2c** in $CDCl_3$.

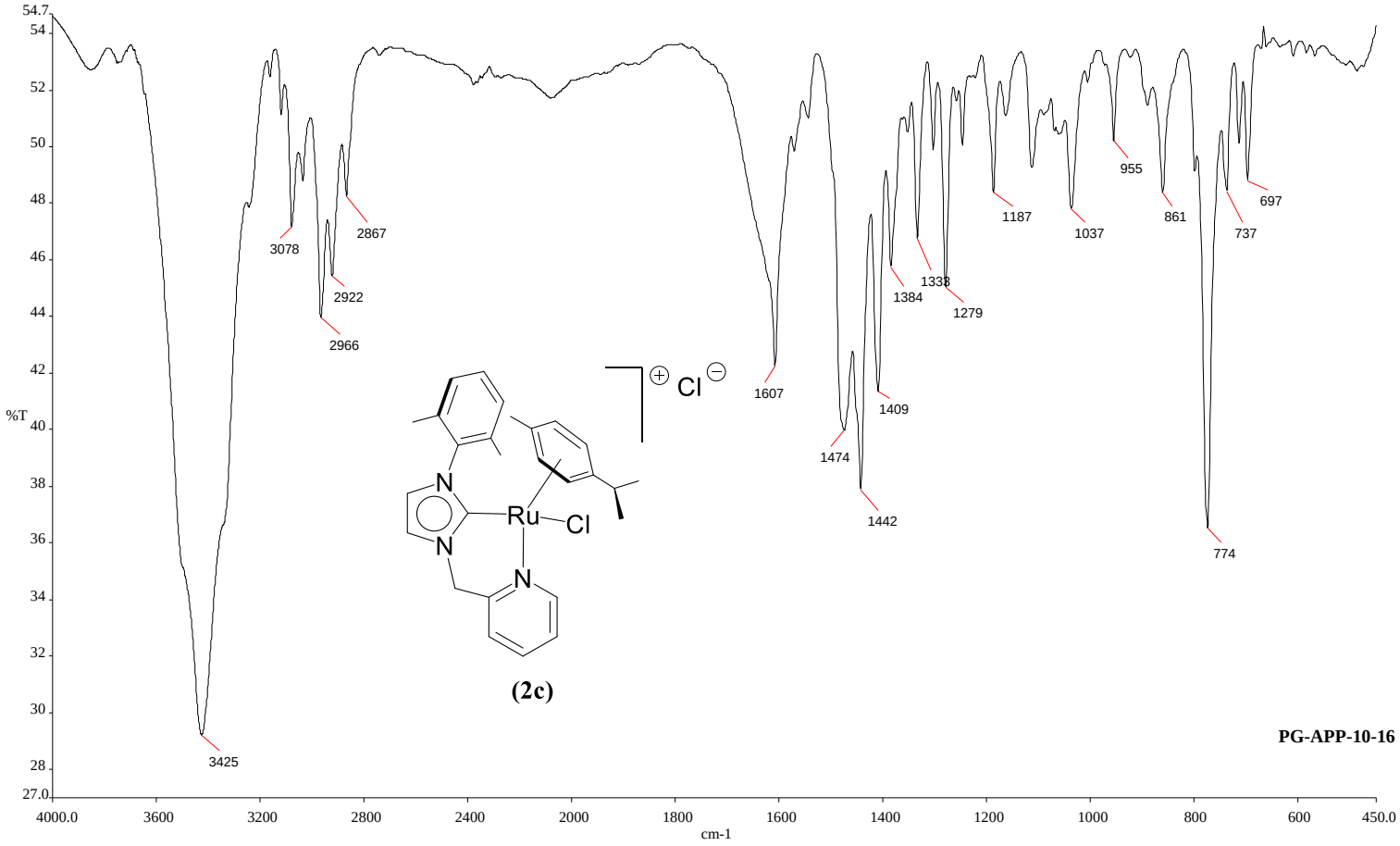
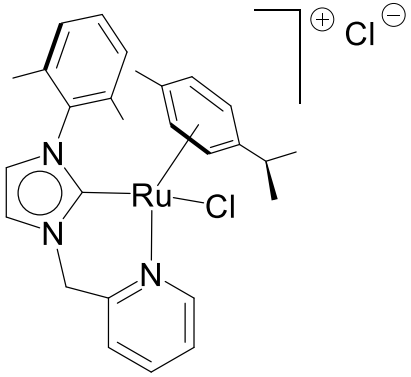


Figure S52. IR spectrum of **2c** in KBr.



(2c)

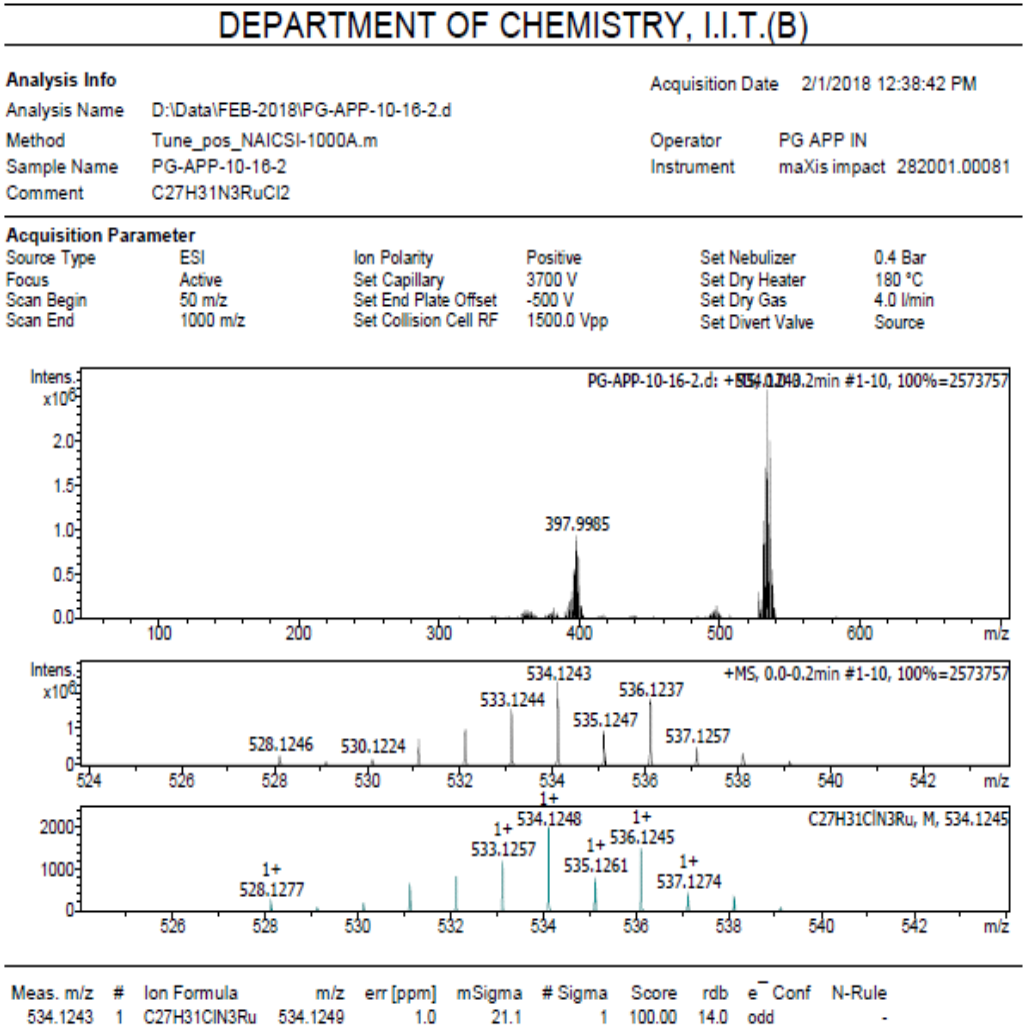


Figure S53. High Resolution Mass Spectrometry (HRMS) data of **2c**.

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	N [%]	C [%]	H [%]	Date Time	Info
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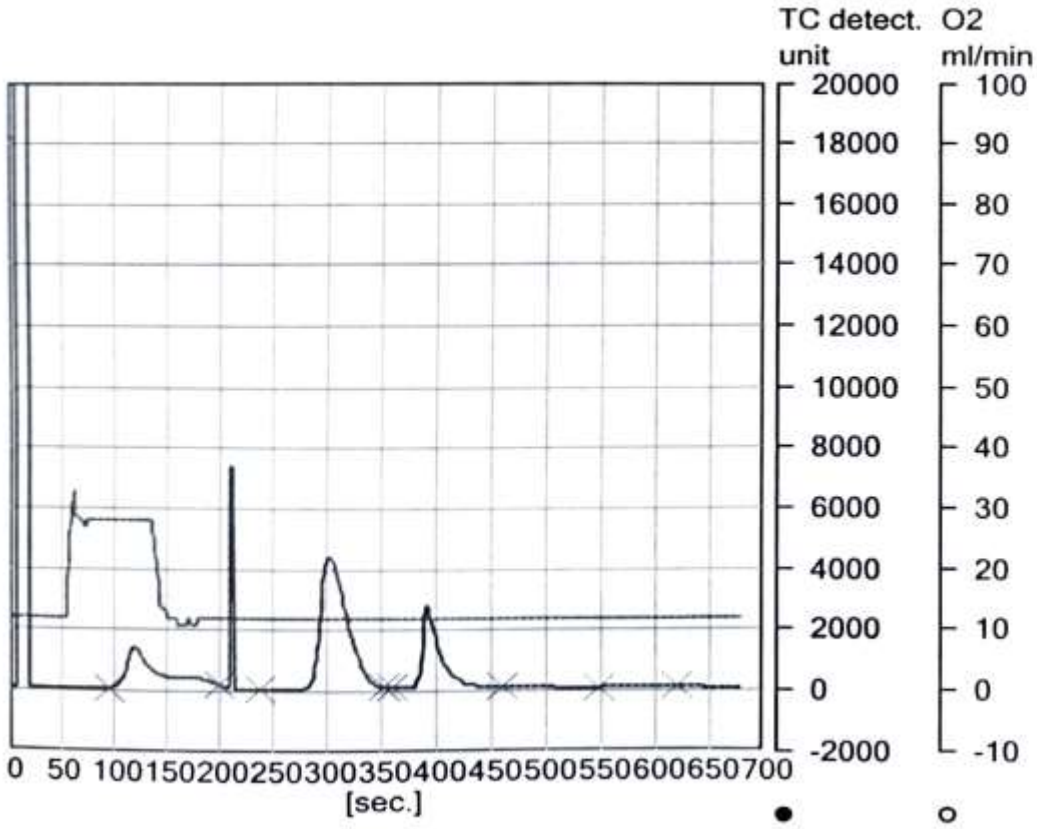
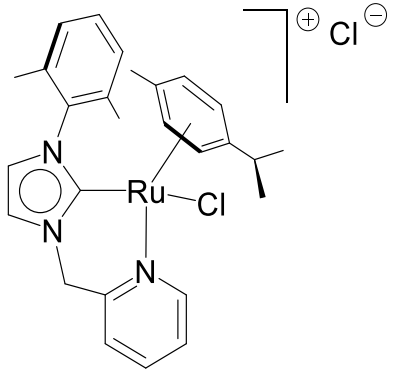




(**2c**)

Figure S54. Elemental analysis data of **2c**.

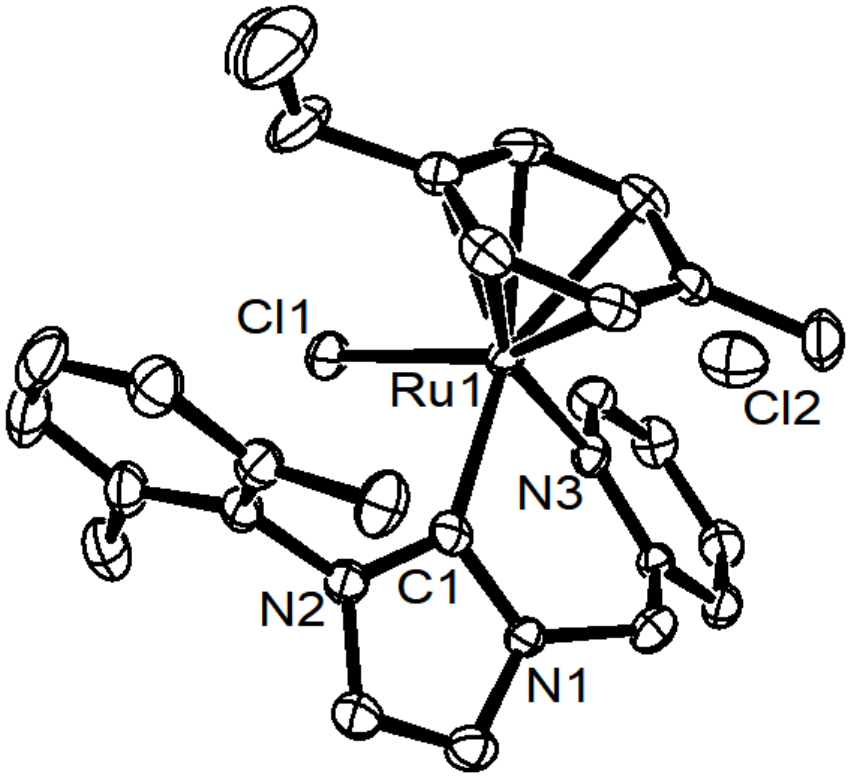
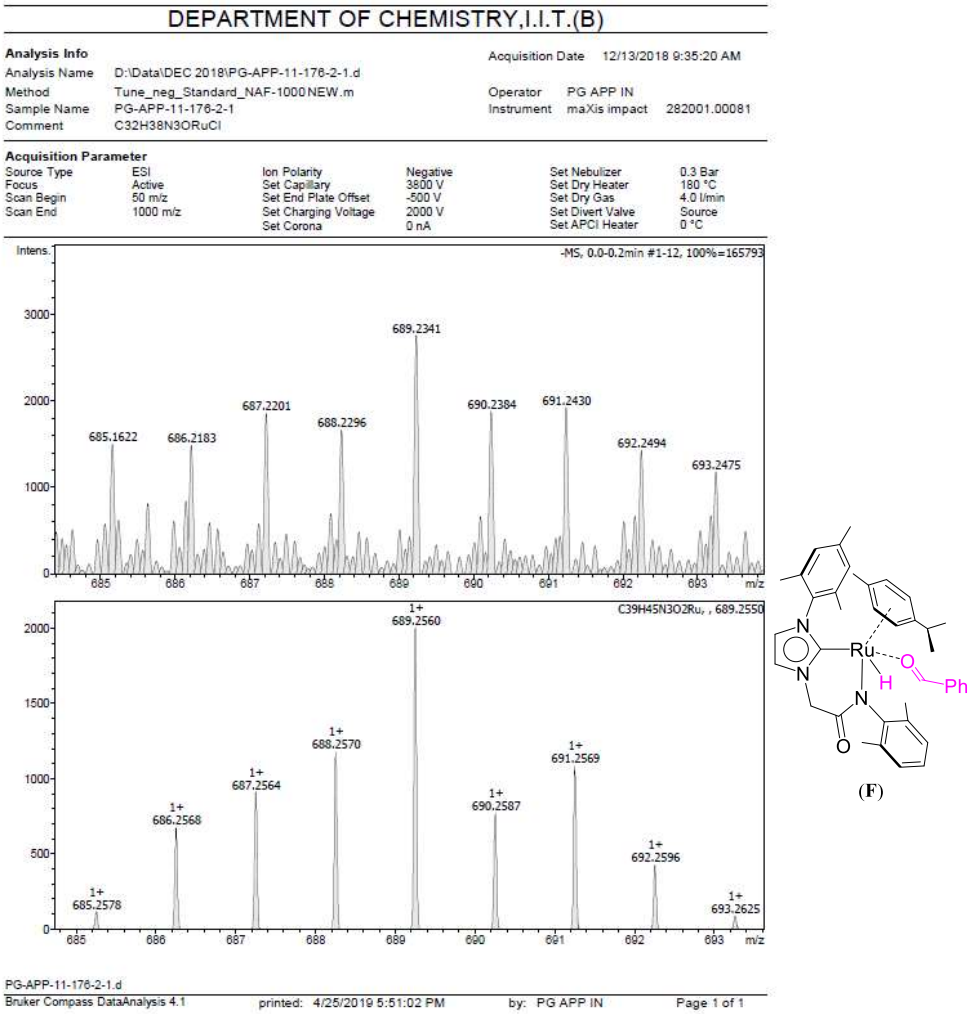


Figure S55. ORTEP of **2c** with thermal ellipsoids shown at the 50 % probability level. Hydrogen atoms and co-crystallized water molecules were omitted for clarity. Selected bond lengths (Å) and angles (°): Ru(1)−C(1) 2.052(4), Ru(1)−N(3) 2.110(3), Ru(1)−Cl(1) 2.3936(9), C(1)−N(1) 1.359(5), C(1)−N(2) 1.358(5), C(1)−Ru(1)−Cl(1) 89.28(10), C(1)−Ru(1)−N(3) 85.17(13), N(3)−Ru(1)−Cl(1) 84.85(8), N(2)−C(1)−Ru(1) 134.7(3), N(1)−C(1)−Ru(1) 121.1(3), N(1)−C(1)−N(2) 104.2(3).



(**F**)

Figure S56. ESI-MS data of the benzaldehyde bound Ru–H specie (**F**) detected in the reaction mixture of 1:1:1 ratio of benzyl alcohol:1-phenylethan-1-ol:NaO*i*Pr 0.1 mmol, 1 mol % of (**1c**), 2.0 mL of toluene at 110 °C for 1 hour.

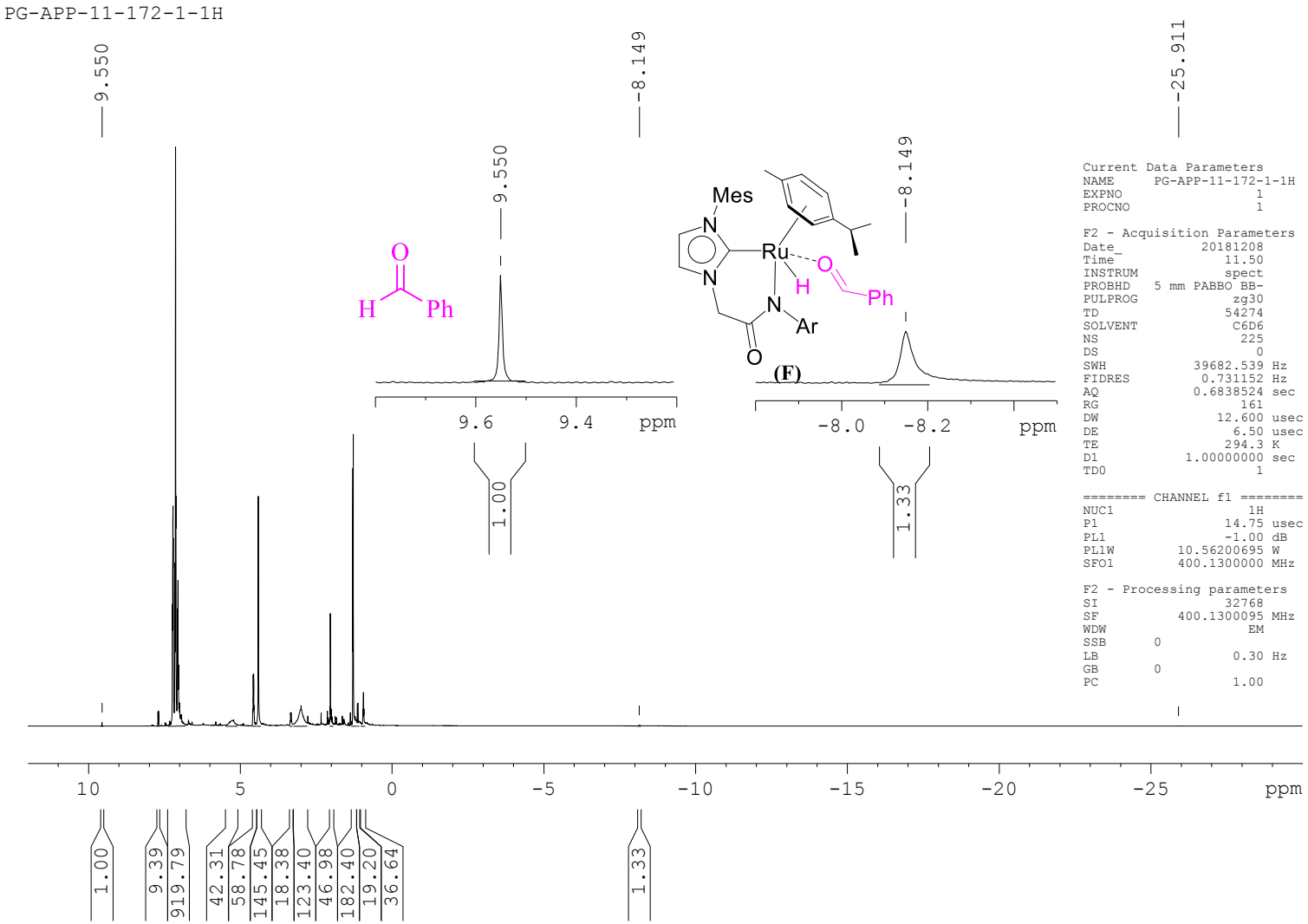


Figure S57. ^{1}H NMR spectrum of the ruthenium hydride specie (**F**) (–8.15 ppm) detected in the reaction mixture of 1:1:1 ratio of benzyl alcohol:1-phenylethanol:KOH 0.1 mmol, 1 mol % of (**1c**), 0.5 mL of C_6D_6 at 90 °C for 30 mins.

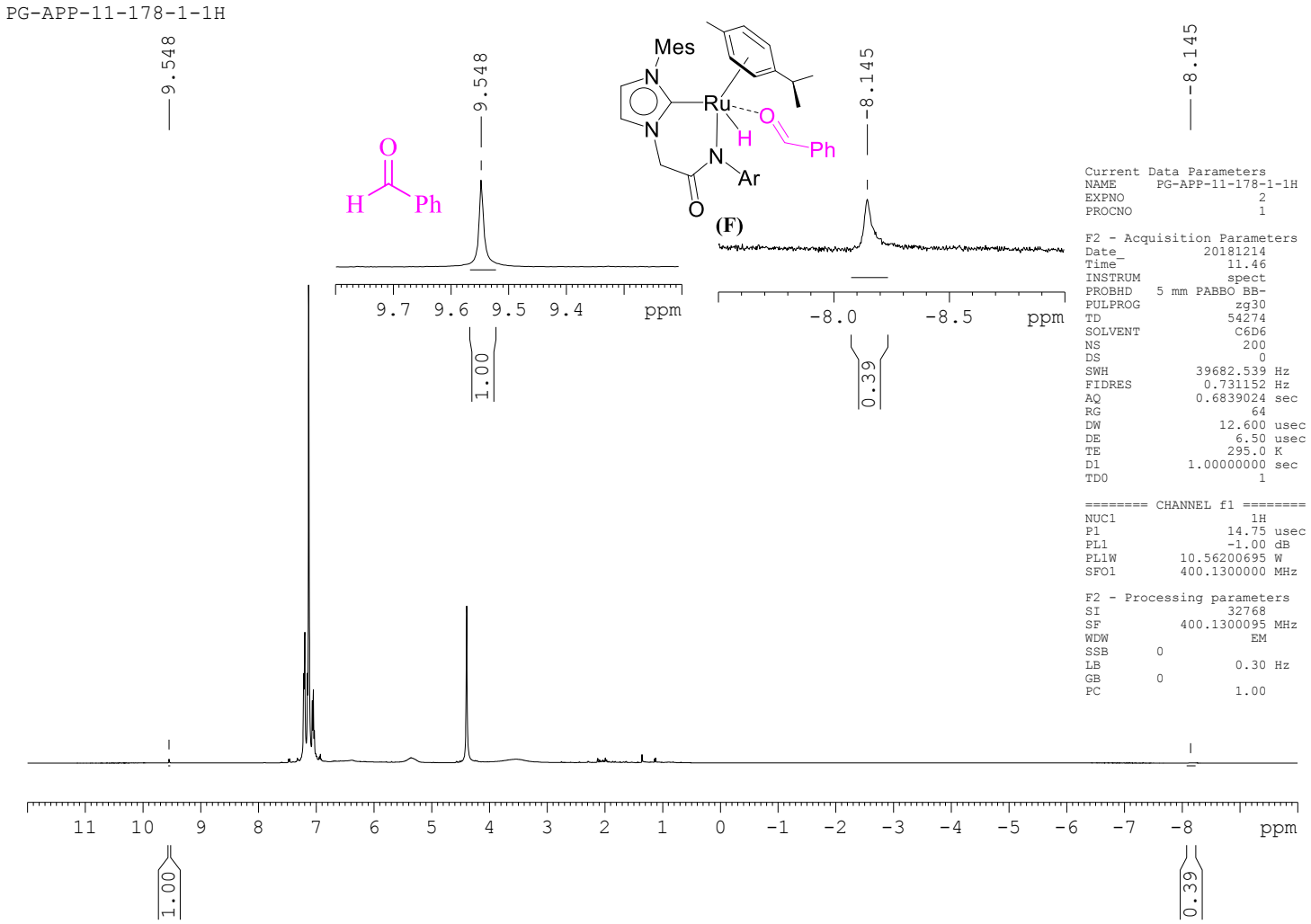


Figure S58. ^{1}H NMR spectrum of the ruthenium hydride specie (**F**) (–8.15 ppm) detected in the reaction mixture of 1:1 ratio of benzyl alcohol:KOH 0.1 mmol, 1 mol % of (**1c**), 0.5 mL of C_6D_6 at 90 °C for 30 mins.

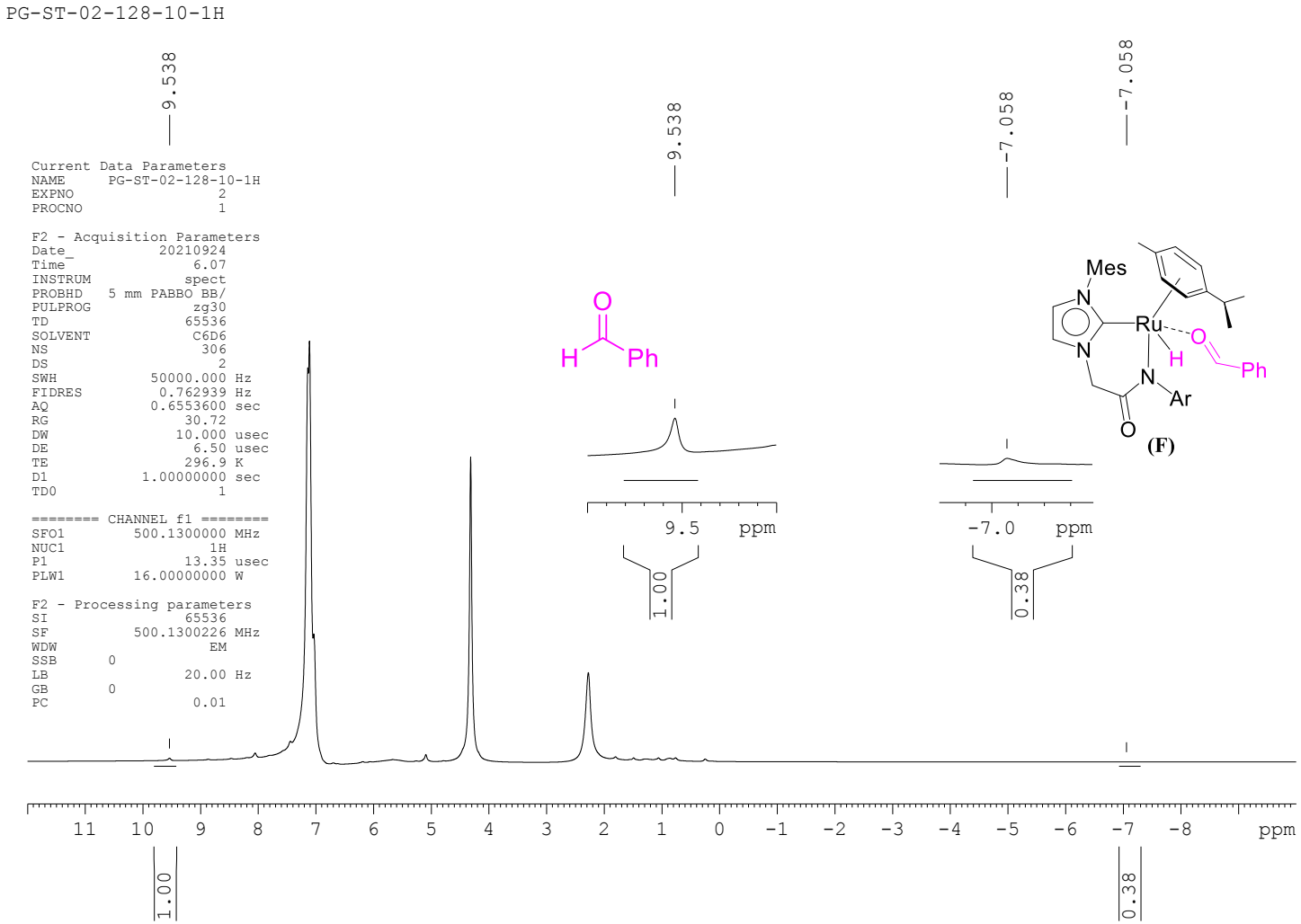


Figure S59. ^{1}H NMR spectrum of the ruthenium hydride specie (**F**) (–7.06 ppm) detected in the reaction mixture of 1:1 ratio of benzyl alcohol:NaO-*i*-Pr 0.1 mmol, 1 mol % of (**1c**), 0.5 mL of C_6D_6 at 90 °C for 30 mins.

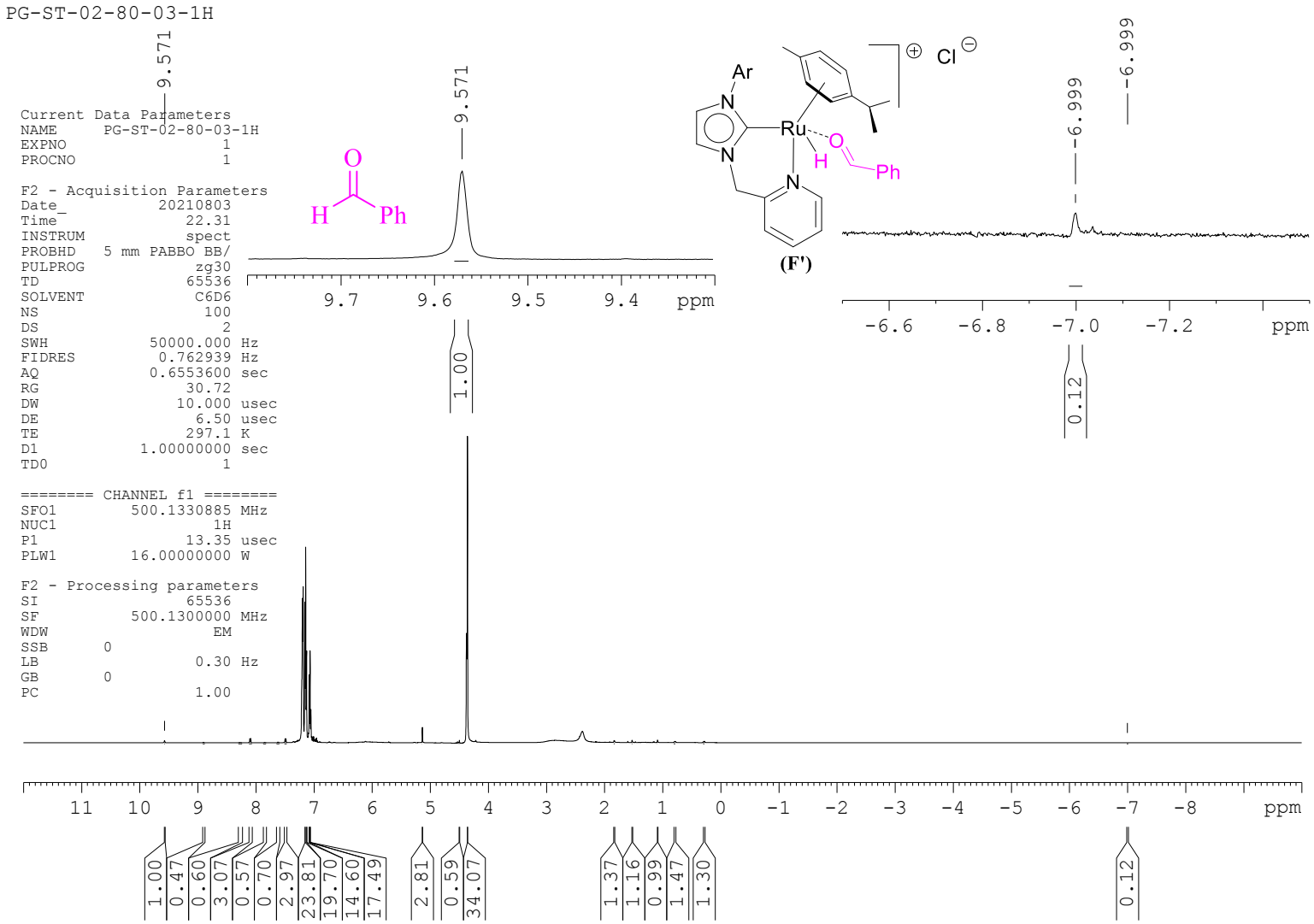


Figure S60. ^{1}H NMR spectrum of the ruthenium hydride specie (**F'**) (–6.99 ppm) detected in the reaction mixture of 1:1 ratio of benzyl alcohol: KOH 0.1 mmol, 1 mol % of (**2c**), 0.5 mL of C_6D_6 at 90 °C for 30 mins.

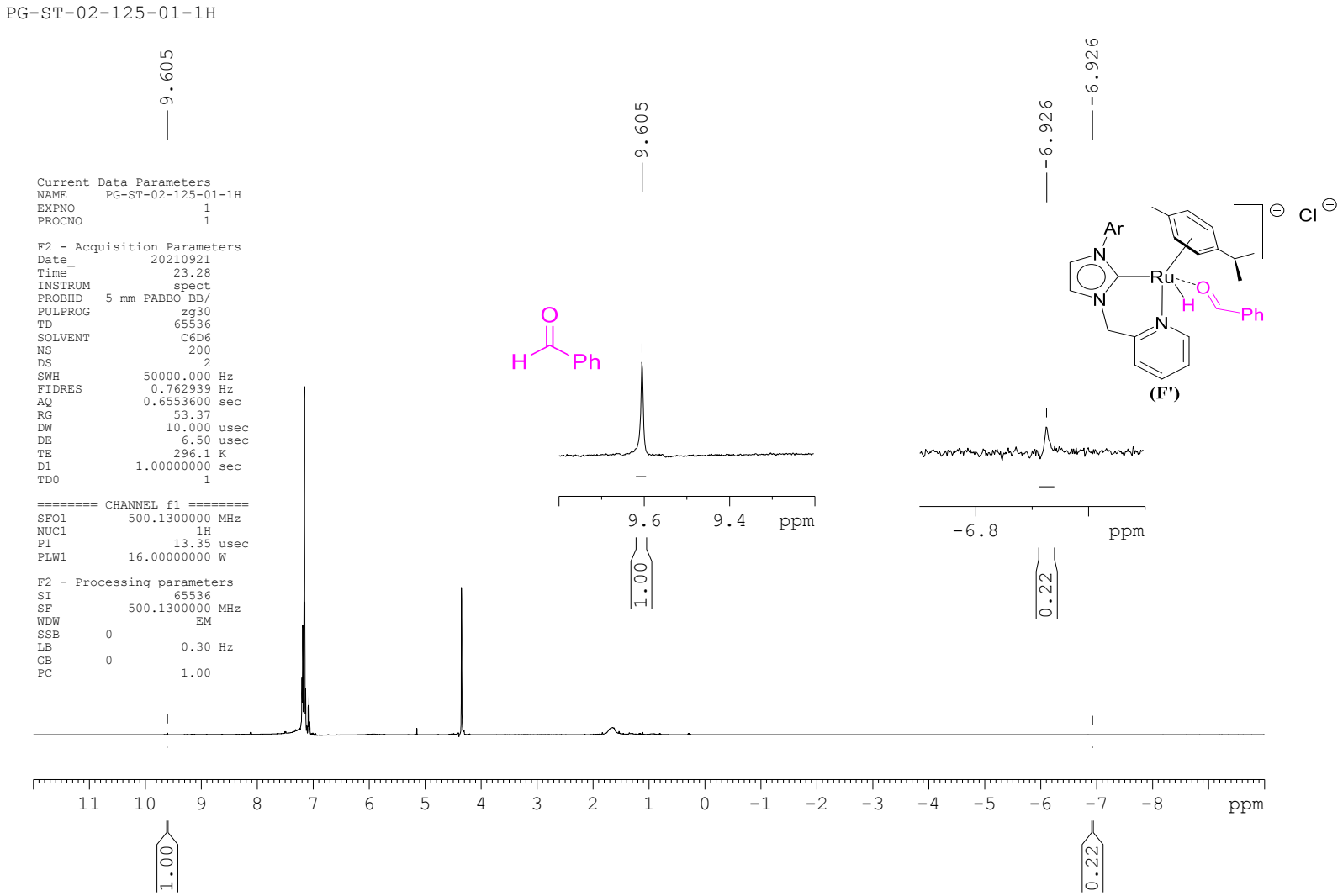


Figure S61. ^{1}H NMR spectrum of the ruthenium hydride specie (**F'**) (–6.93 ppm) detected in the reaction mixture of 1:1 ratio of benzyl alcohol:NaO-*i*-Pr 0.1 mmol, 1 mol % of (**2c**), 0.5 mL of C_6D_6 at 90 °C for 30 mins.

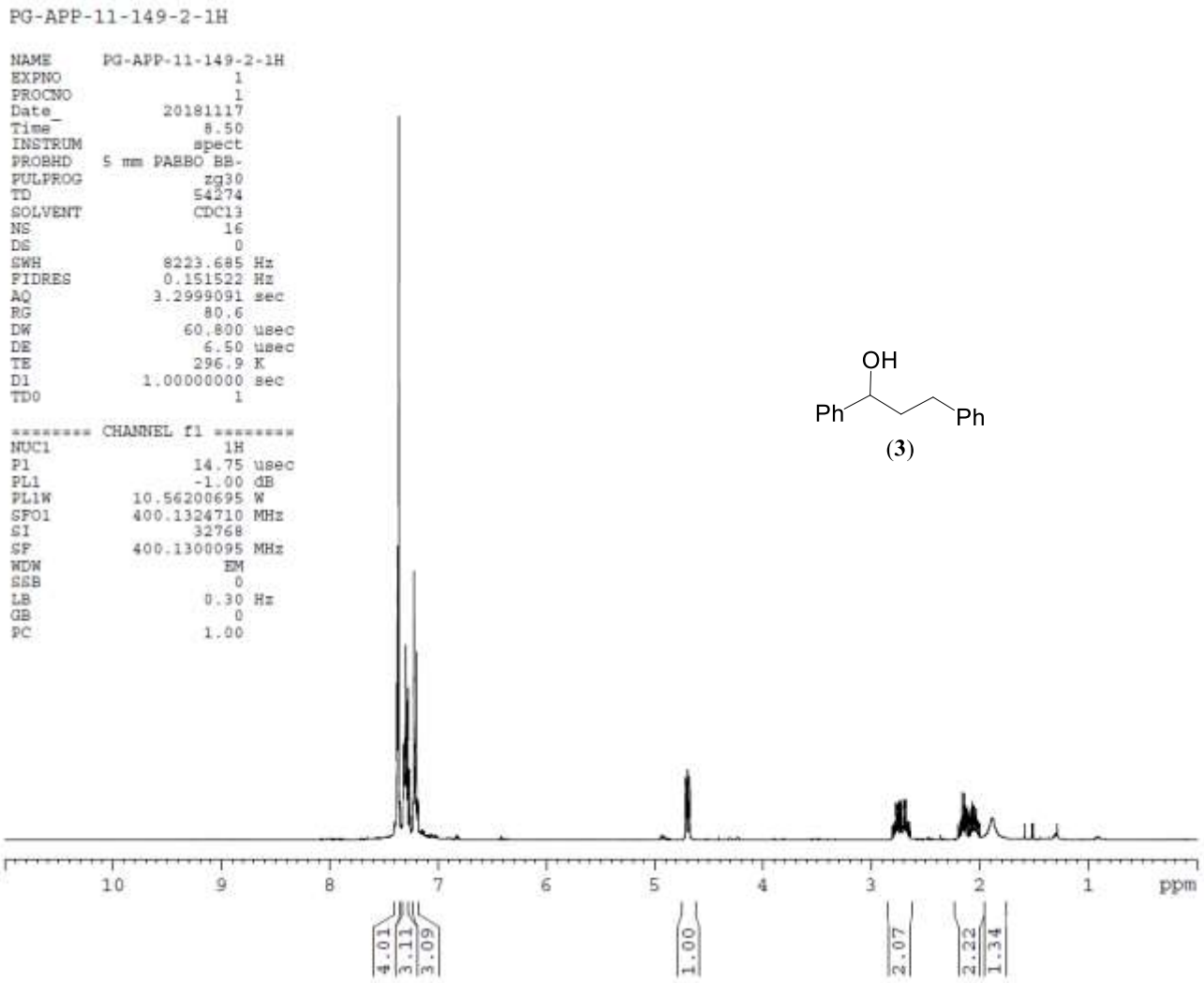


Figure S62. ^{1}H NMR spectrum of (**3**) in $CDCl_3$.

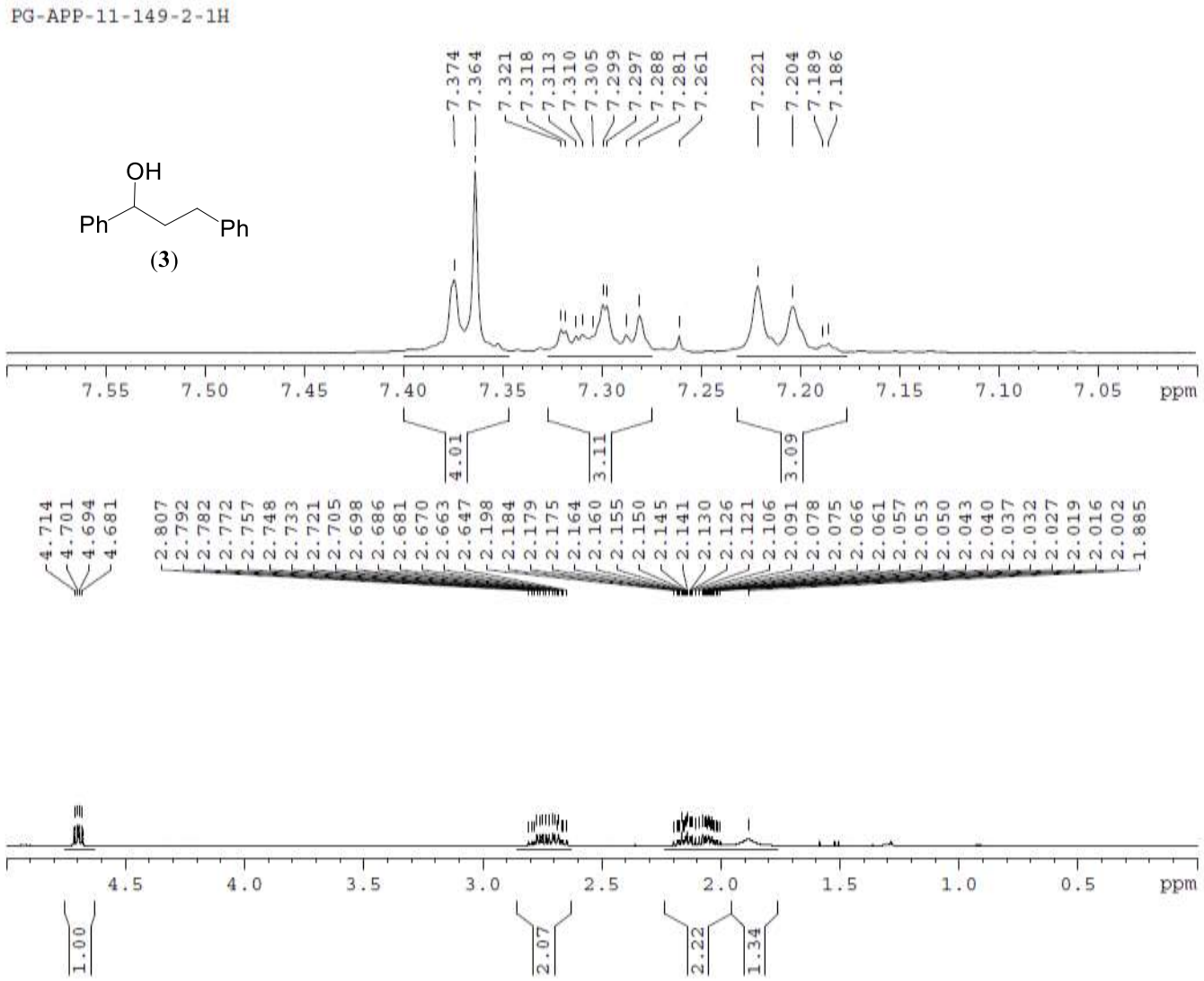


Figure S63. Expanded ^{1}H NMR spectrum of (**3**) in $CDCl_3$.

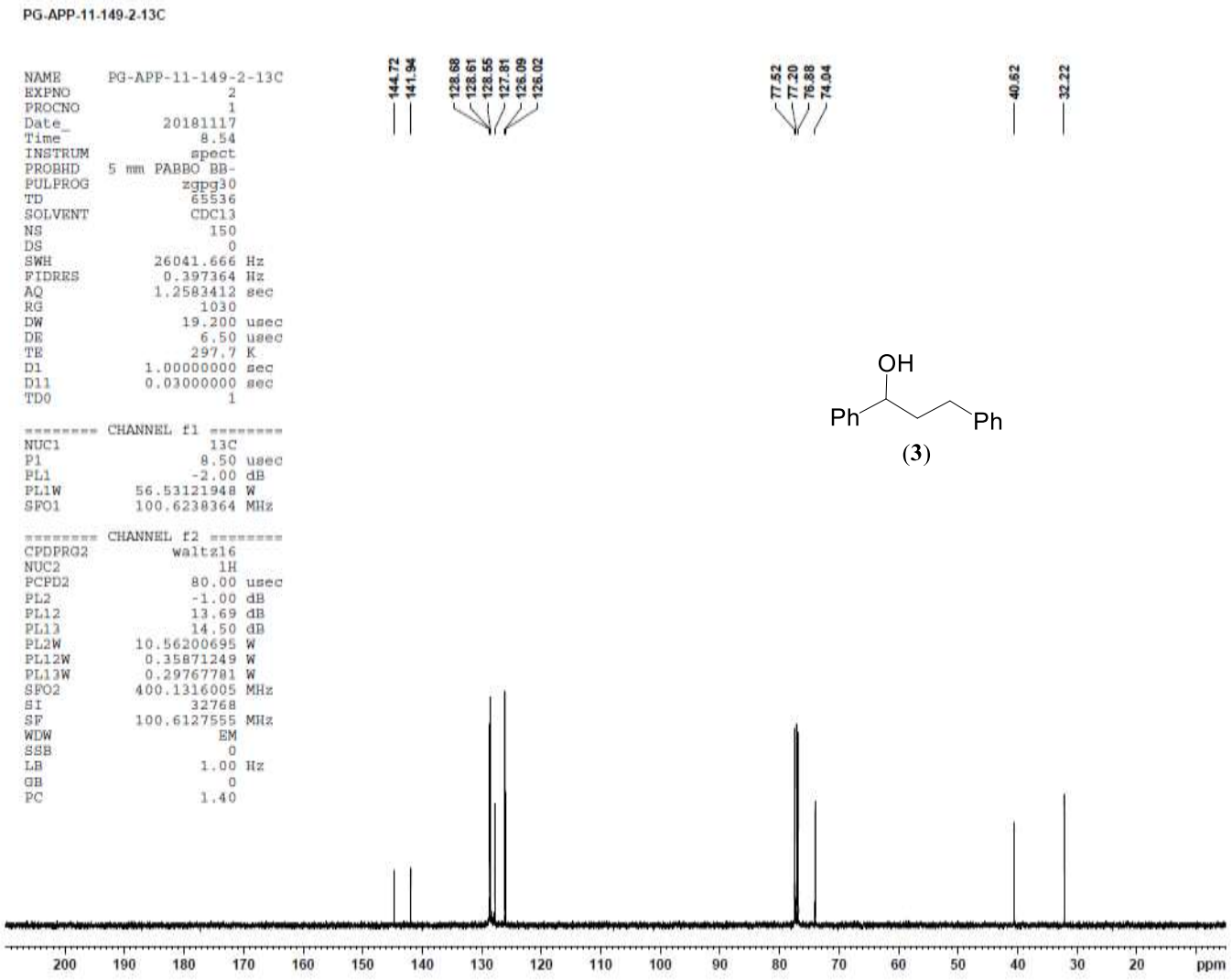


Figure S64. $^{13}C\{^{1}H\}$ NMR spectrum of (**3**) in $CDCl_3$.

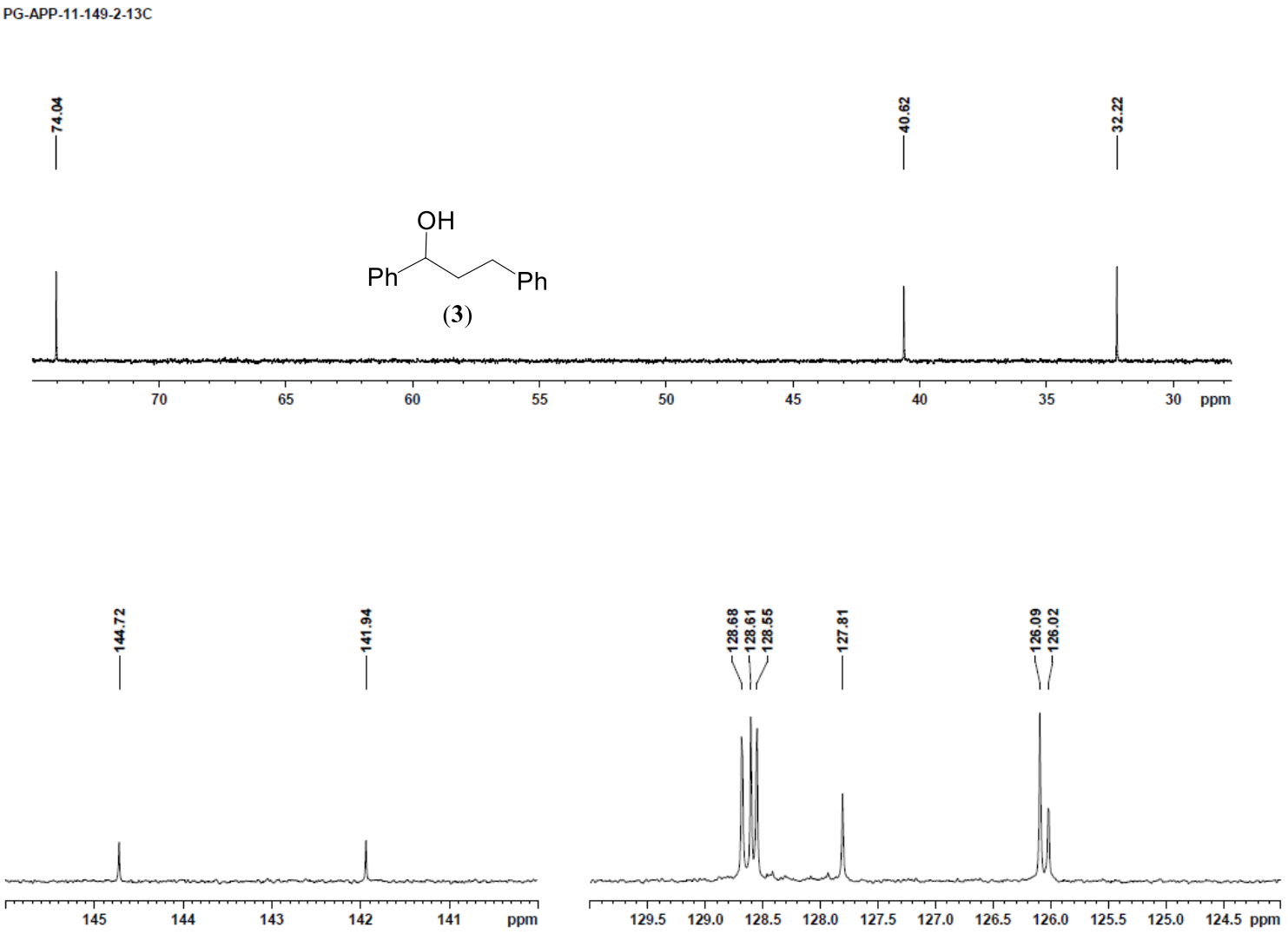


Figure S65. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**3**) in $CDCl_3$.

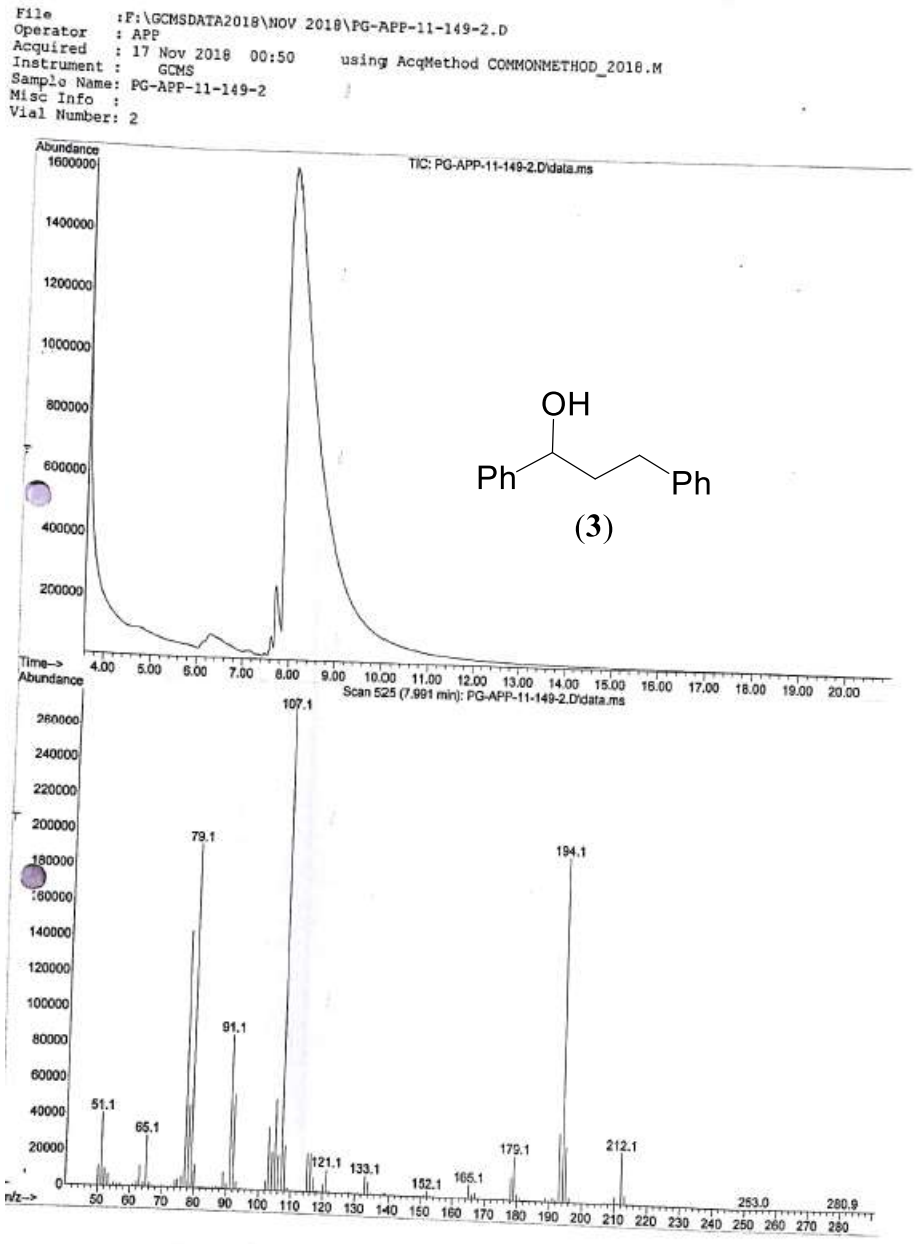
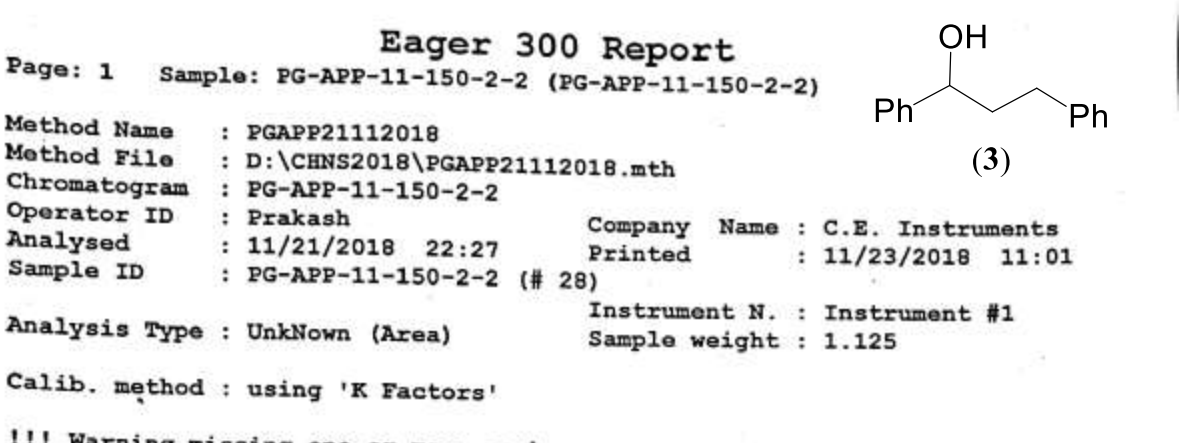


Figure S66. GCMS trace in EtOAc of (**3**) showing the M^+ peak at *m/z* 212.



Eager 300 Report

Page: 1 Sample: PG-APP-11-150-2-2 (PG-APP-11-150-2-2)

Method Name : PGAPP21112018
Method File : D:\CHNS2018\PGAPP21112018.mth
Chromatogram : PG-APP-11-150-2-2
Operator ID : Prakash
Analysed : 11/21/2018 22:27
Sample ID : PG-APP-11-150-2-2 (# 28)

Company Name : C.E. Instruments
Printed : 11/23/2018 11:01
Instrument N. : Instrument #1
Sample weight : 1.125

Analysis Type : UnkNown (Area)

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
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Carbon	84.1693	62	2509913	RS	1.000000	.265065E+07
Hydrogen	7.4285	184	532988	RS	4.709136	.637770E+07
Totals	91.5978		3180772			

OH
Ph
Ph
(3)

Figure S67. Elemental analysis data of (**3**).

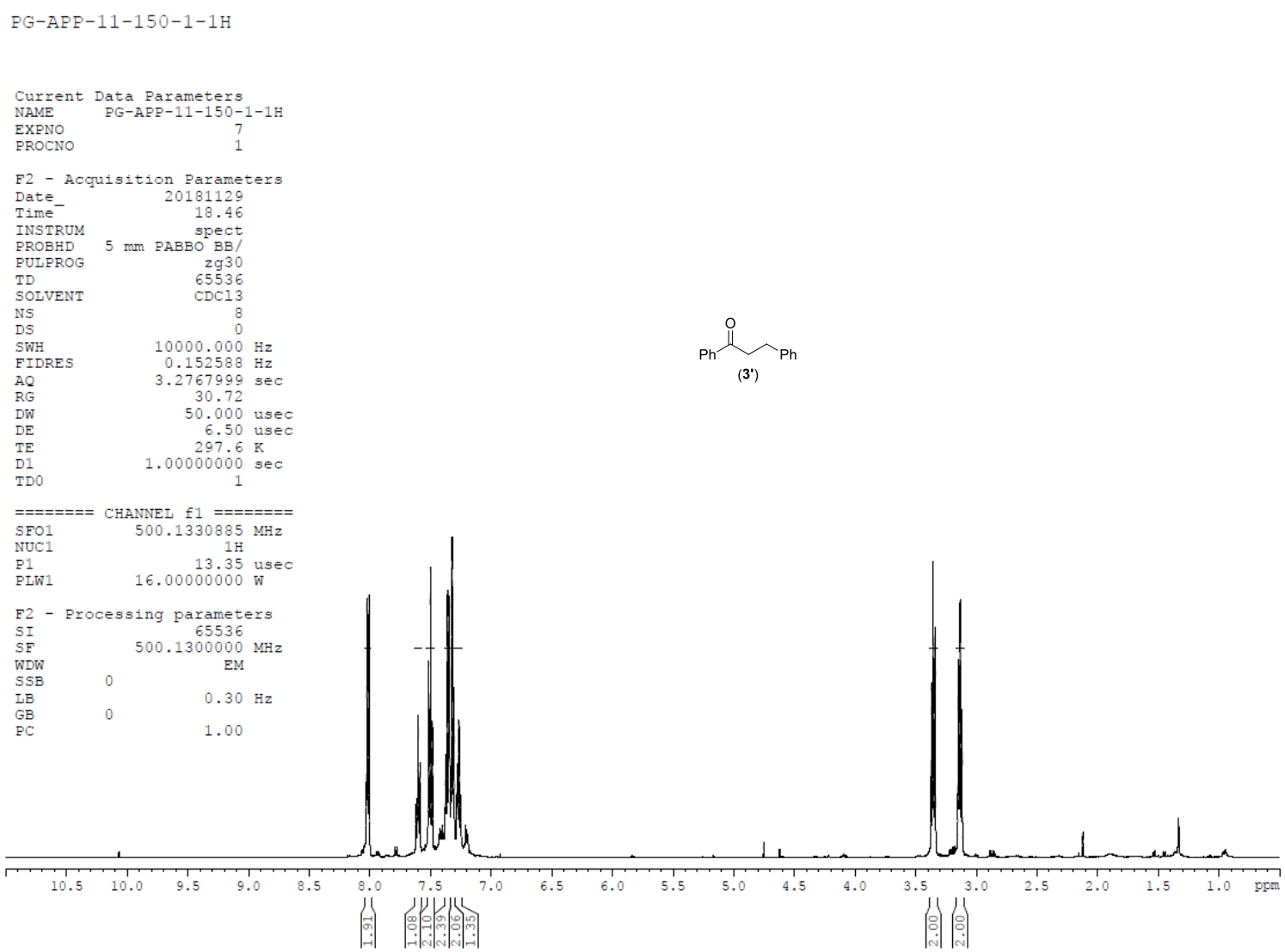

Figure S68. ^{1}H NMR spectrum of (**3'**) in $CDCl_3$.

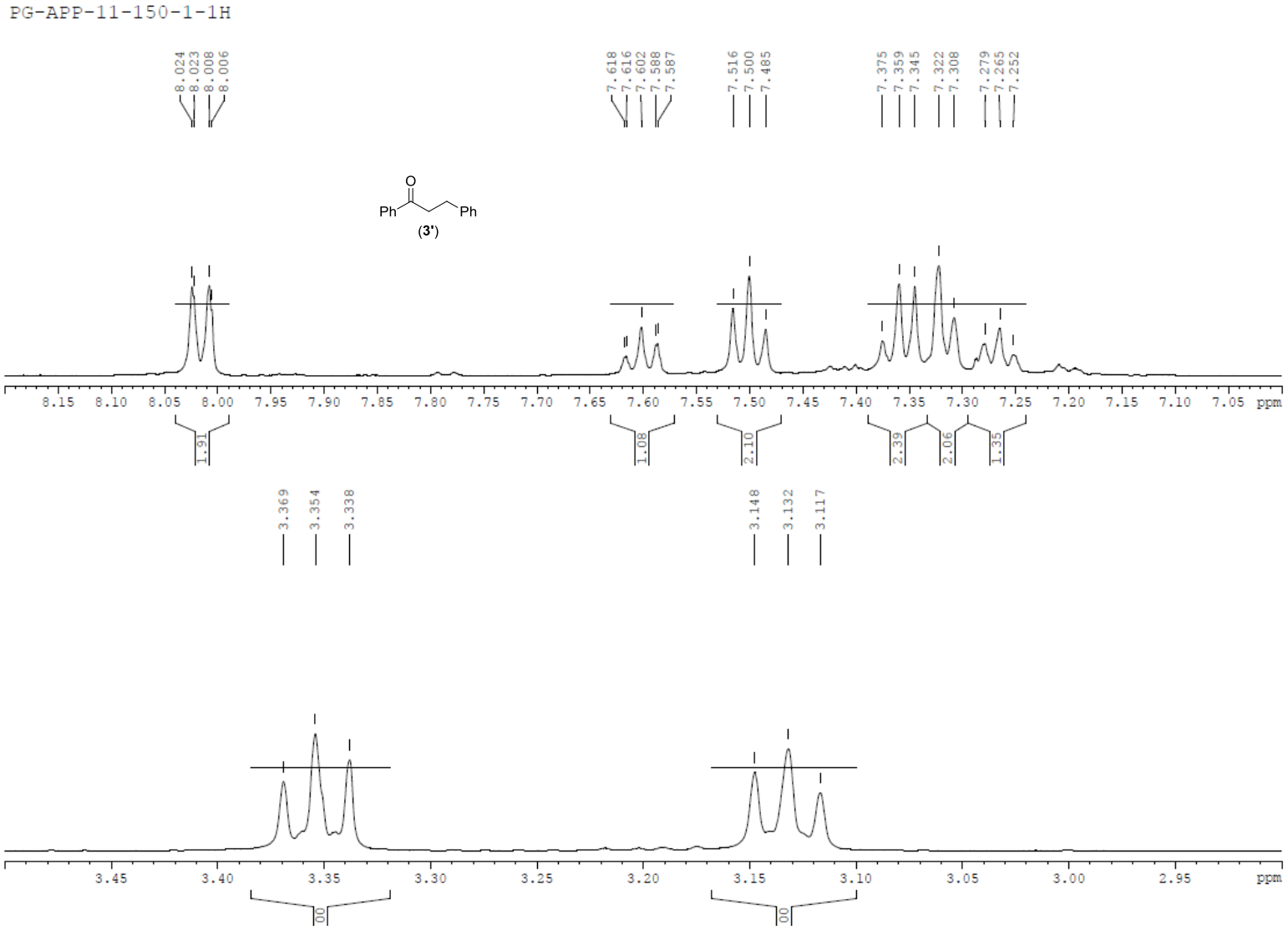

Figure S69. Expanded ^{1}H NMR spectrum of (**3'**) in $CDCl_3$.

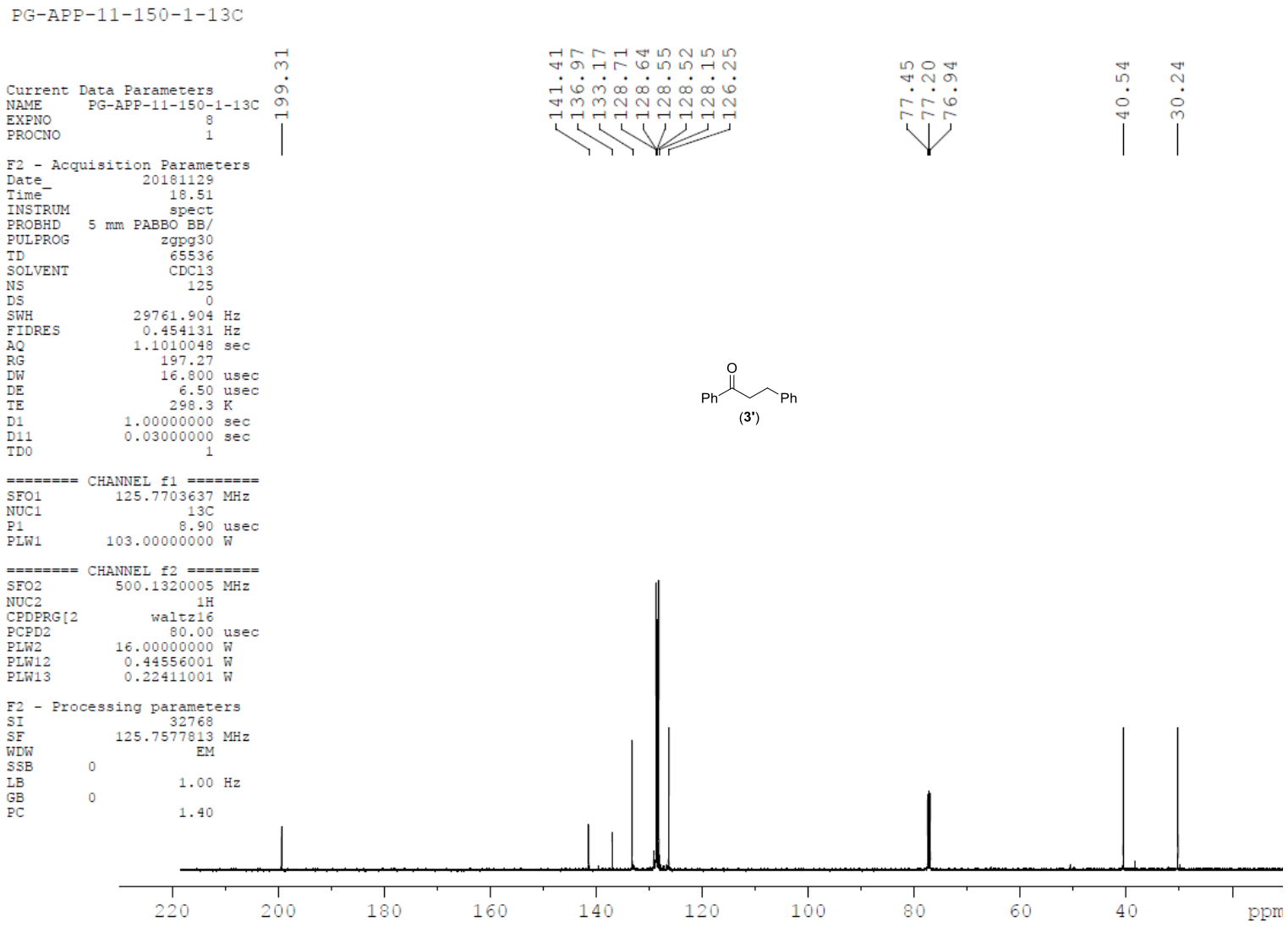

Figure S70. $^{13}C\{^{1}H\}$ NMR spectrum of (**3'**) in $CDCl_3$.

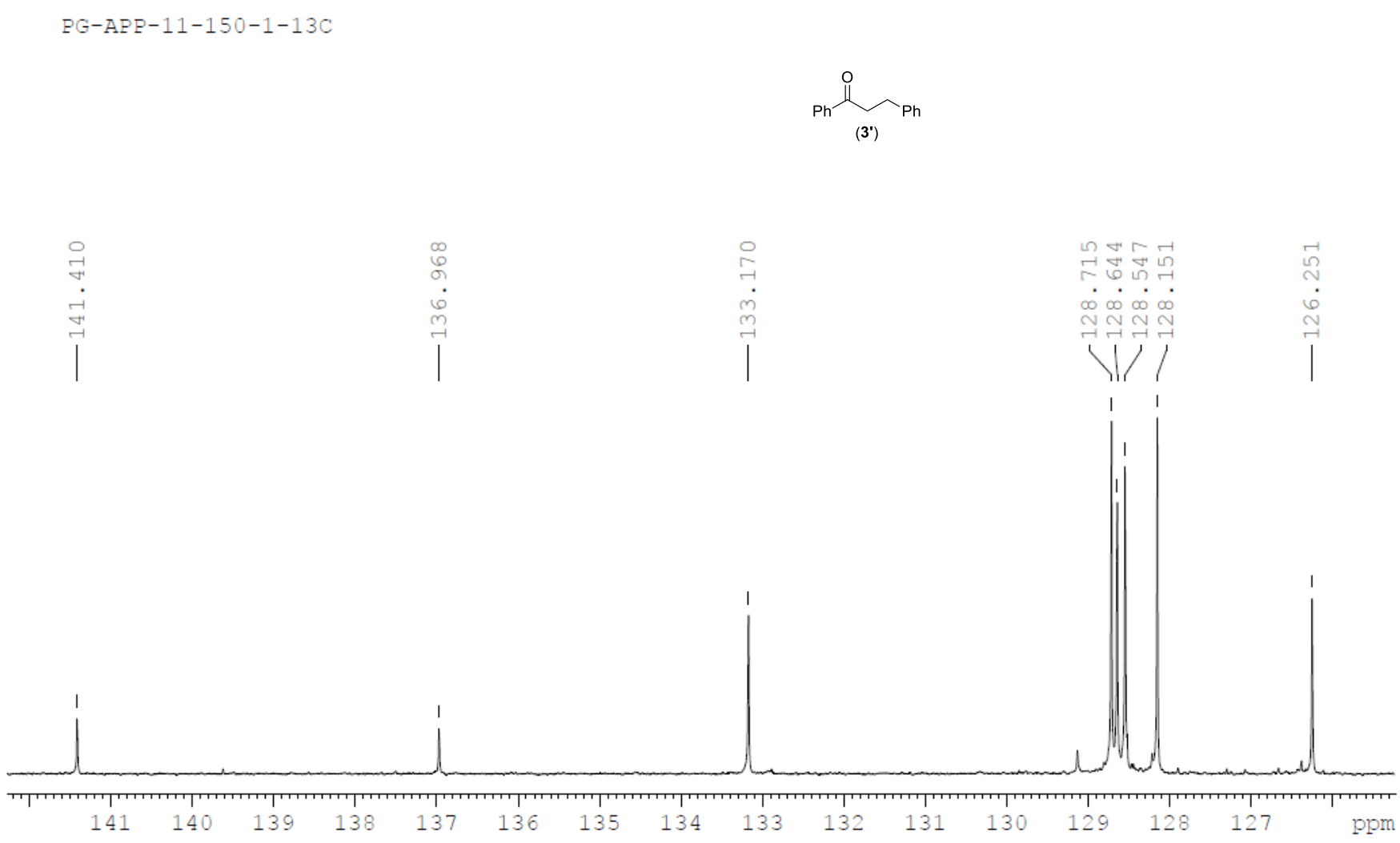

Figure S71. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**3'**) in $CDCl_3$.

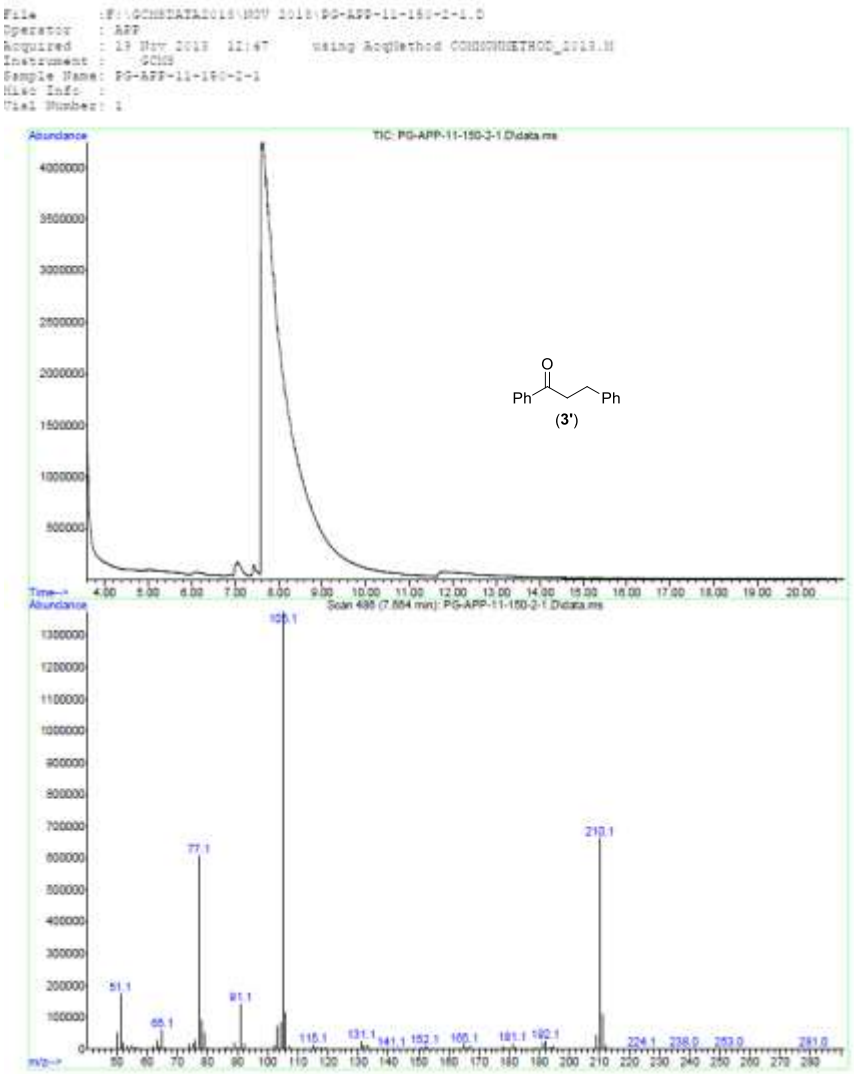

Figure S72. GCMS trace in EtOAc of (**3'**) showing the M^+ peak at *m/z* 210.

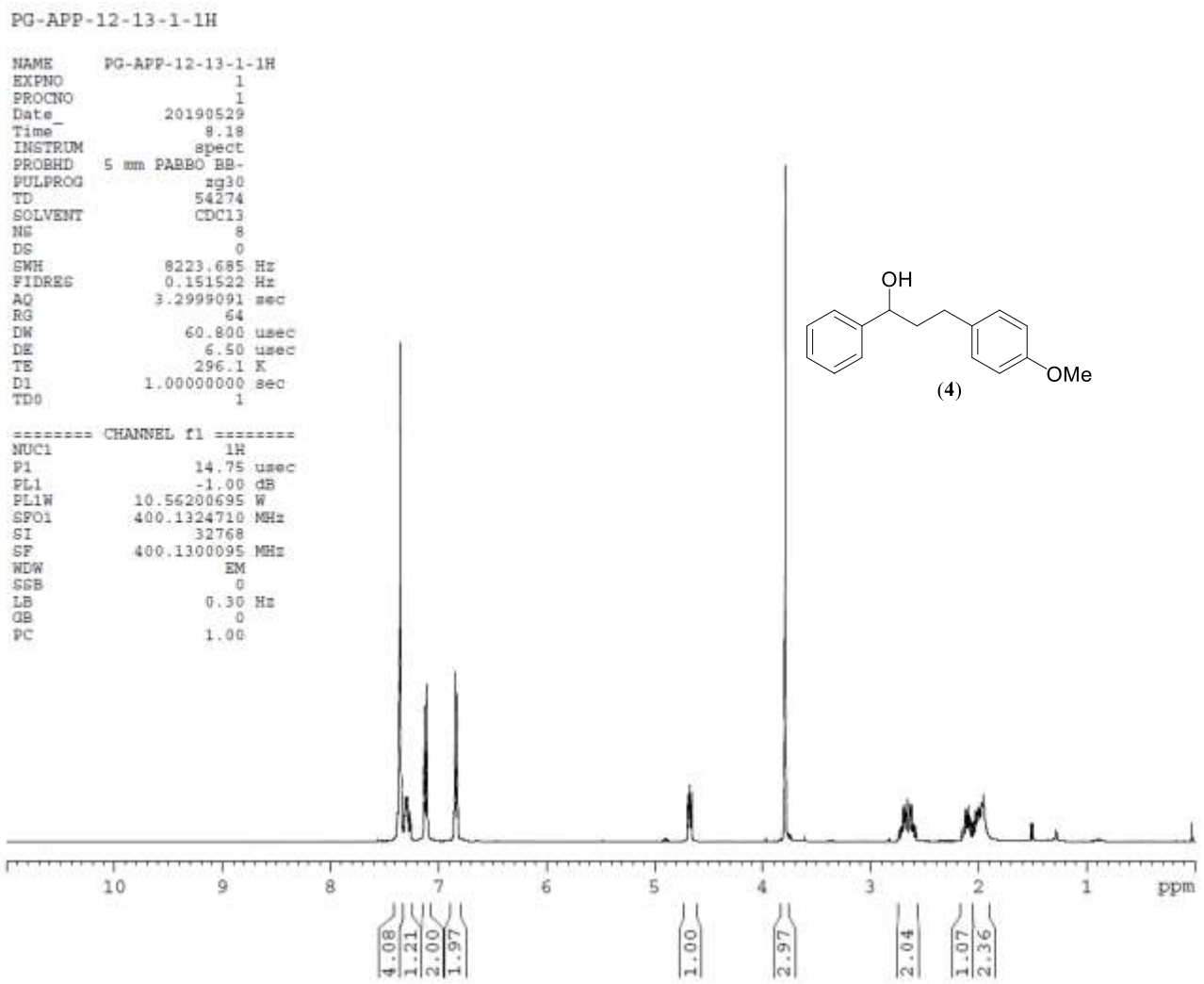

Figure S73. ^{1}H NMR spectrum of (**4**) in $CDCl_3$.

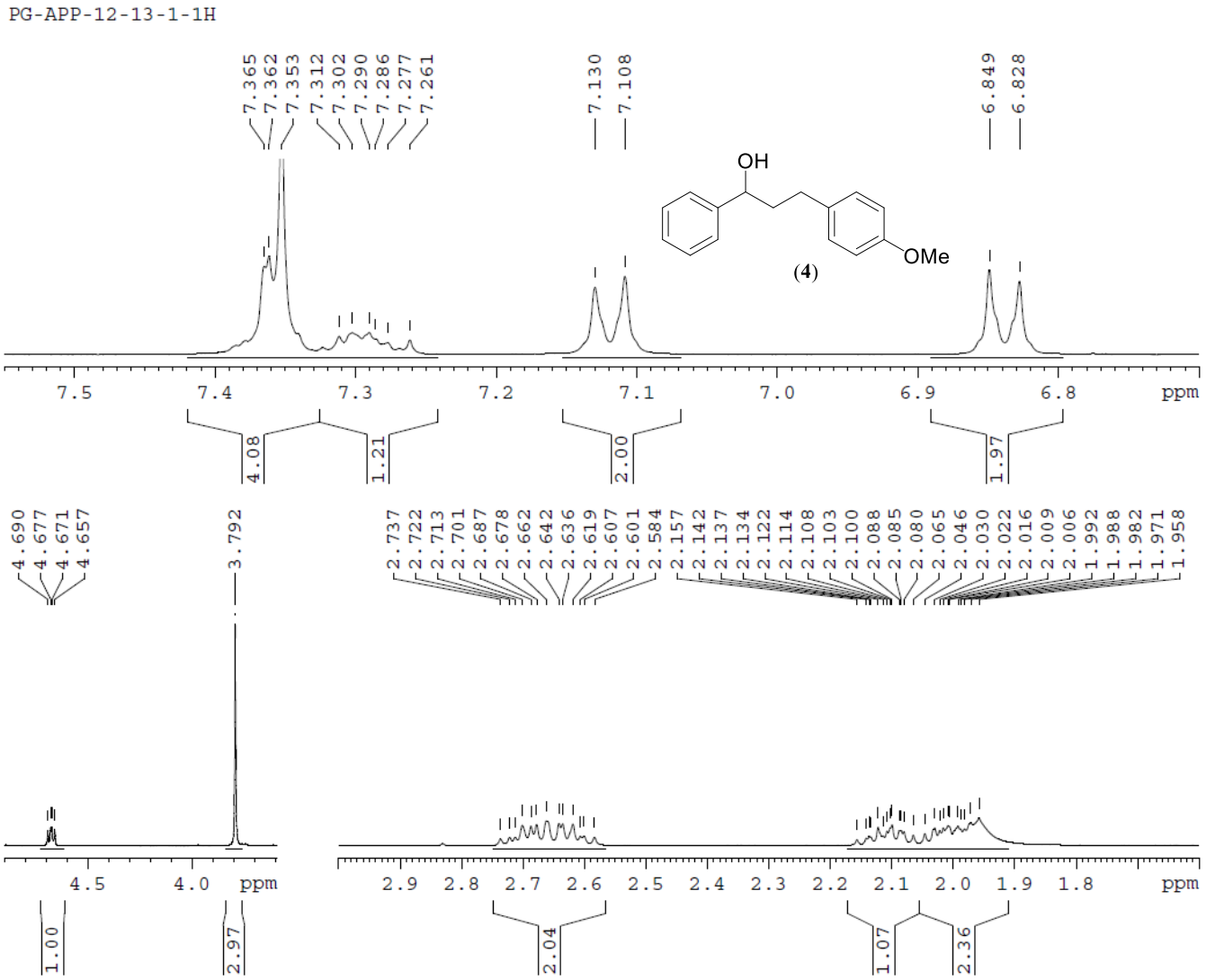

Figure S74. Expanded ^{1}H NMR spectrum of (**4**) in $CDCl_3$.

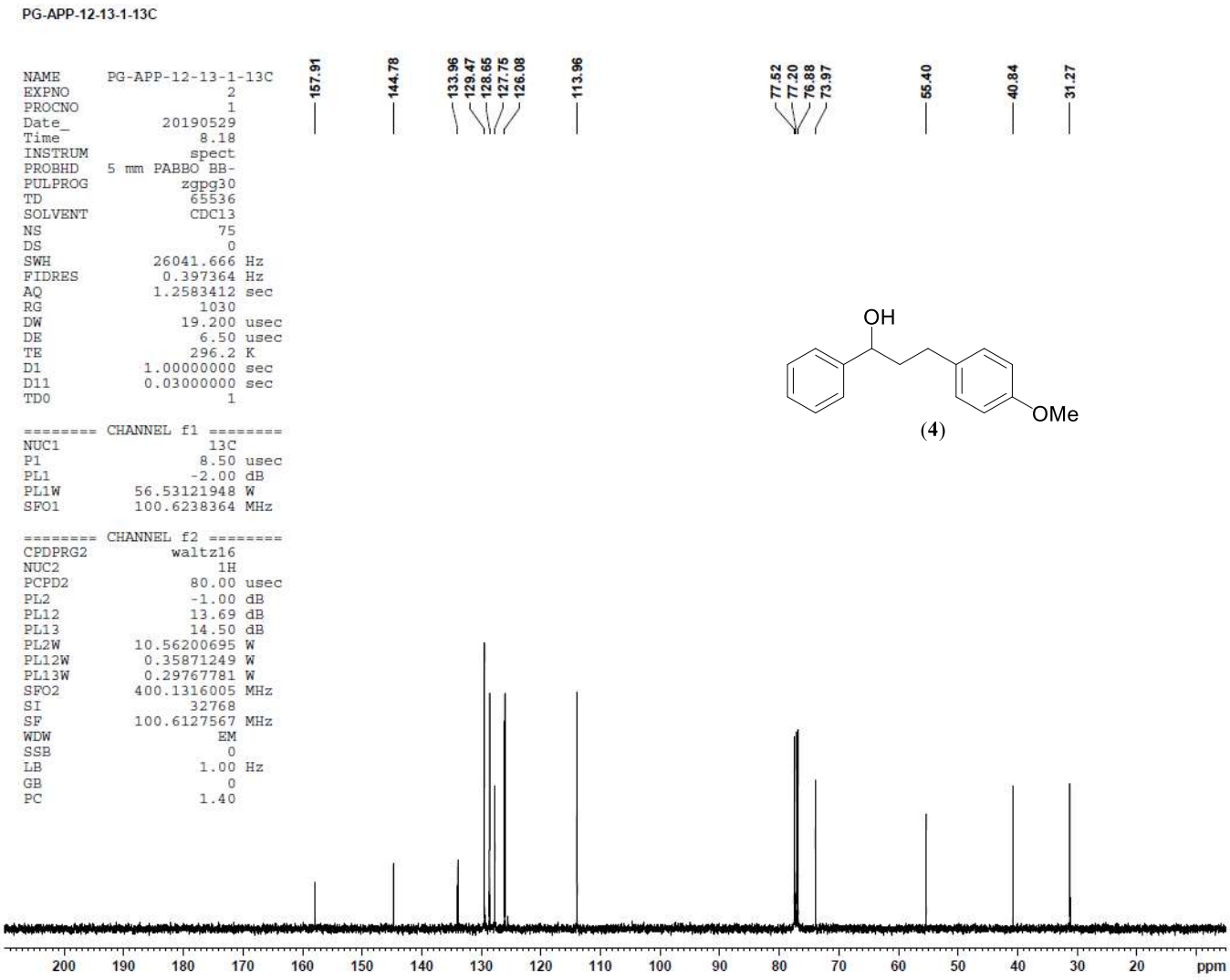

Figure S75. $^{13}C\{^{1}H\}$ NMR spectrum of (**4**) in $CDCl_3$.

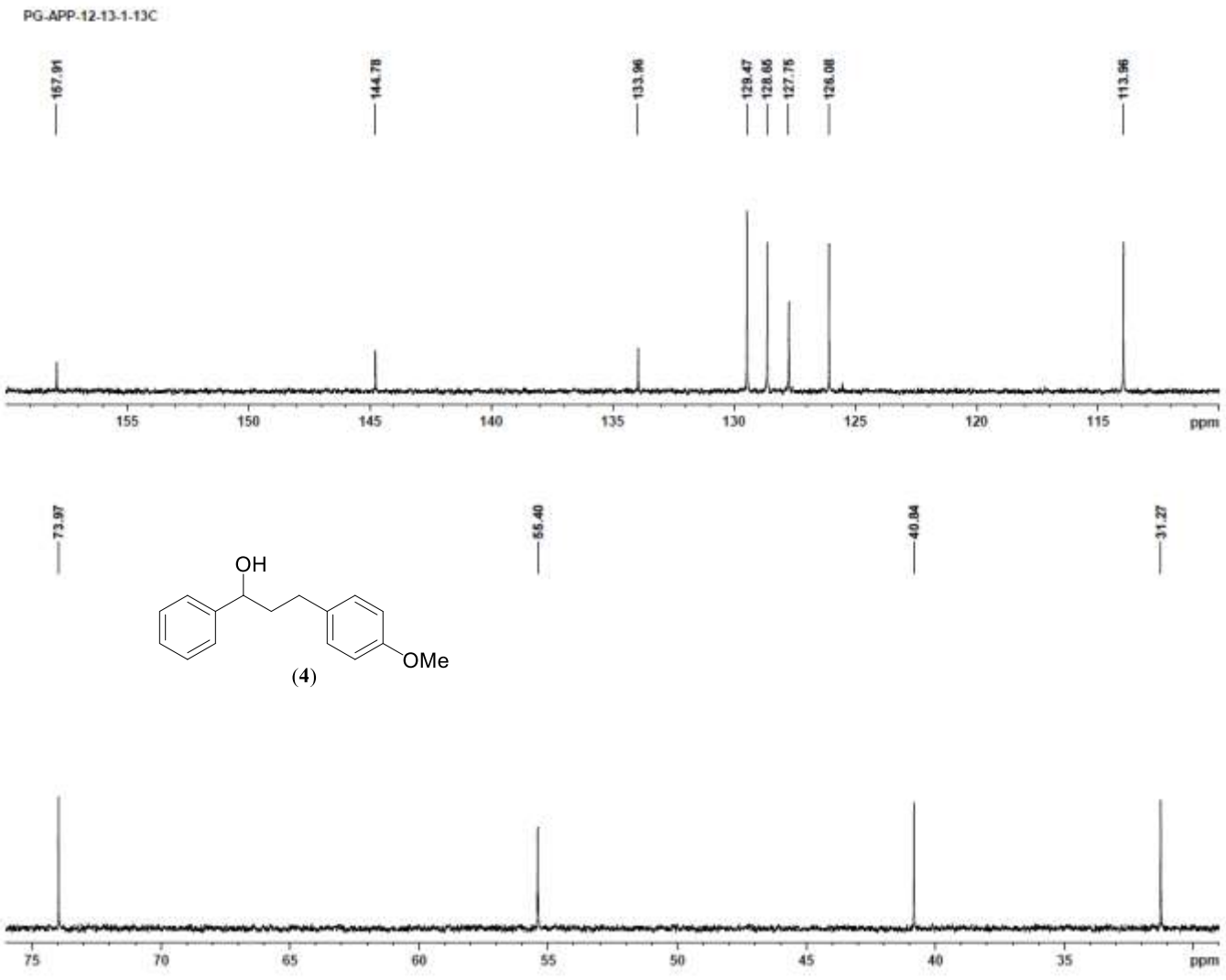

Figure S76. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**4**) in $CDCl_3$.

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Operator   : PG
Acquired   : 28 May 2019  17:55     using AcqMethod COMMONMETHOD-2018.M
Instrument :     GCMS
Sample Name: PG-APP-12-13-4
Misc Info  :
Vial Number: 3
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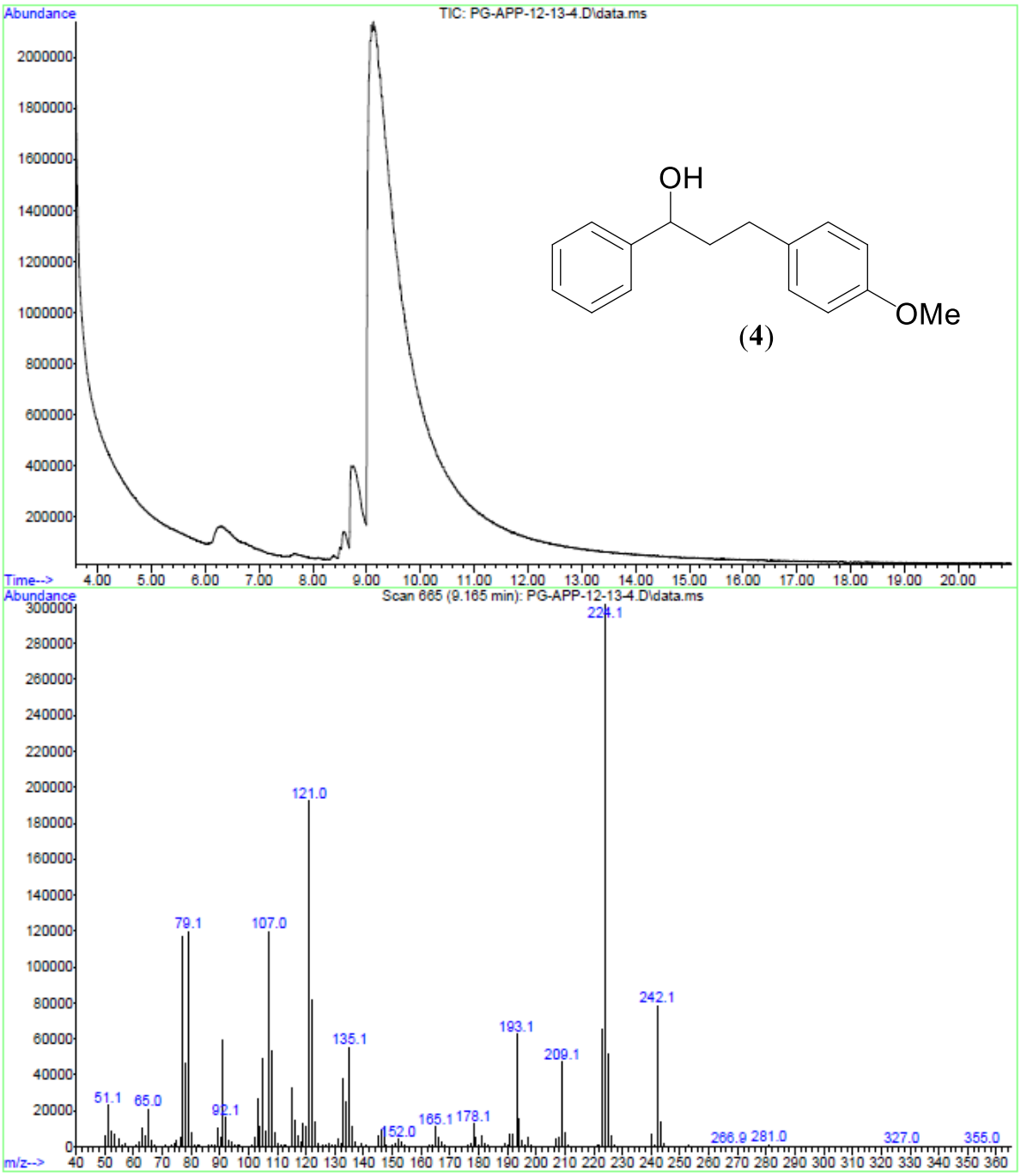
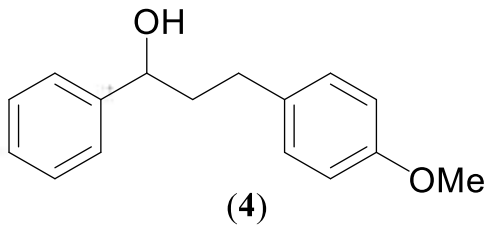


Figure S77. GCMS trace in EtOAc of (**4**) showing the M^+ peak at *m/z* 242.



(**4**)

Eager 300 Report

Page: 1 Sample: PG-APP-12-13-4 (PG-APP-12-13-4)

Method Name : PGAPP300519
Method File : D:\CHNS2019\PGAPP300519.mth
Chromatogram : PG-APP-12-13-4
Operator ID : Prakash
Company Name : C.E. Instruments
Analysed : 05/30/2019 14:30
Printed : 5/30/2019 22:59
Sample ID : PG-APP-12-13-4 (# 19)
Instrument N. : Instrument #1
Analysis Type : UnkNown (Area)
Sample weight : .789

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	6	103472	RS		0.0000
Carbon	78.9676	64	1664418	RS	1.000000	.267139E+07
Hydrogen	7.0210	184	378670	RS	4.395432	.683575E+07
Totals	85.9886		2146560			

Figure S78. Elemental analysis data of (**4**).

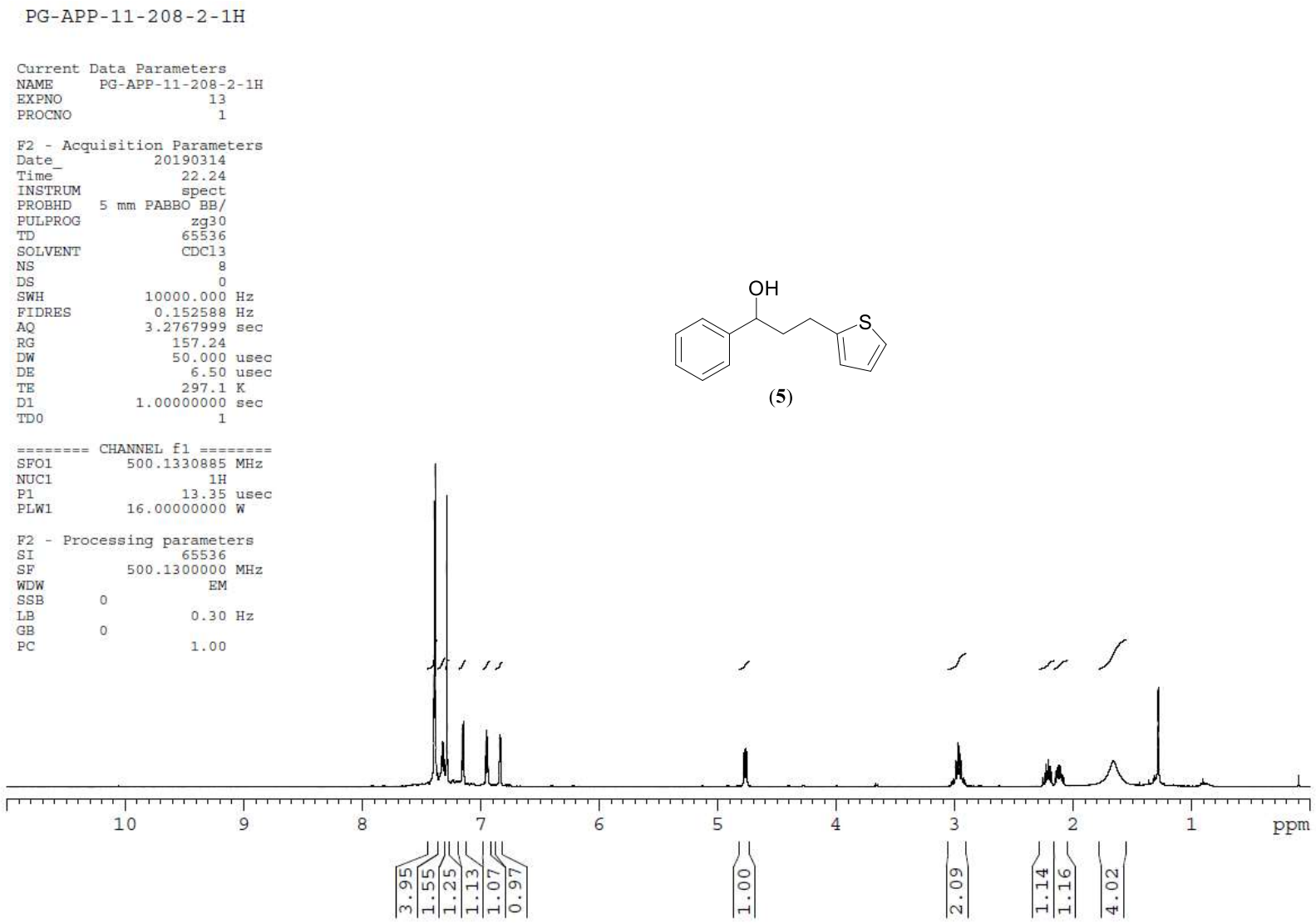


Figure S79. ^{1}H NMR spectrum of (**5**) in $CDCl_3$.

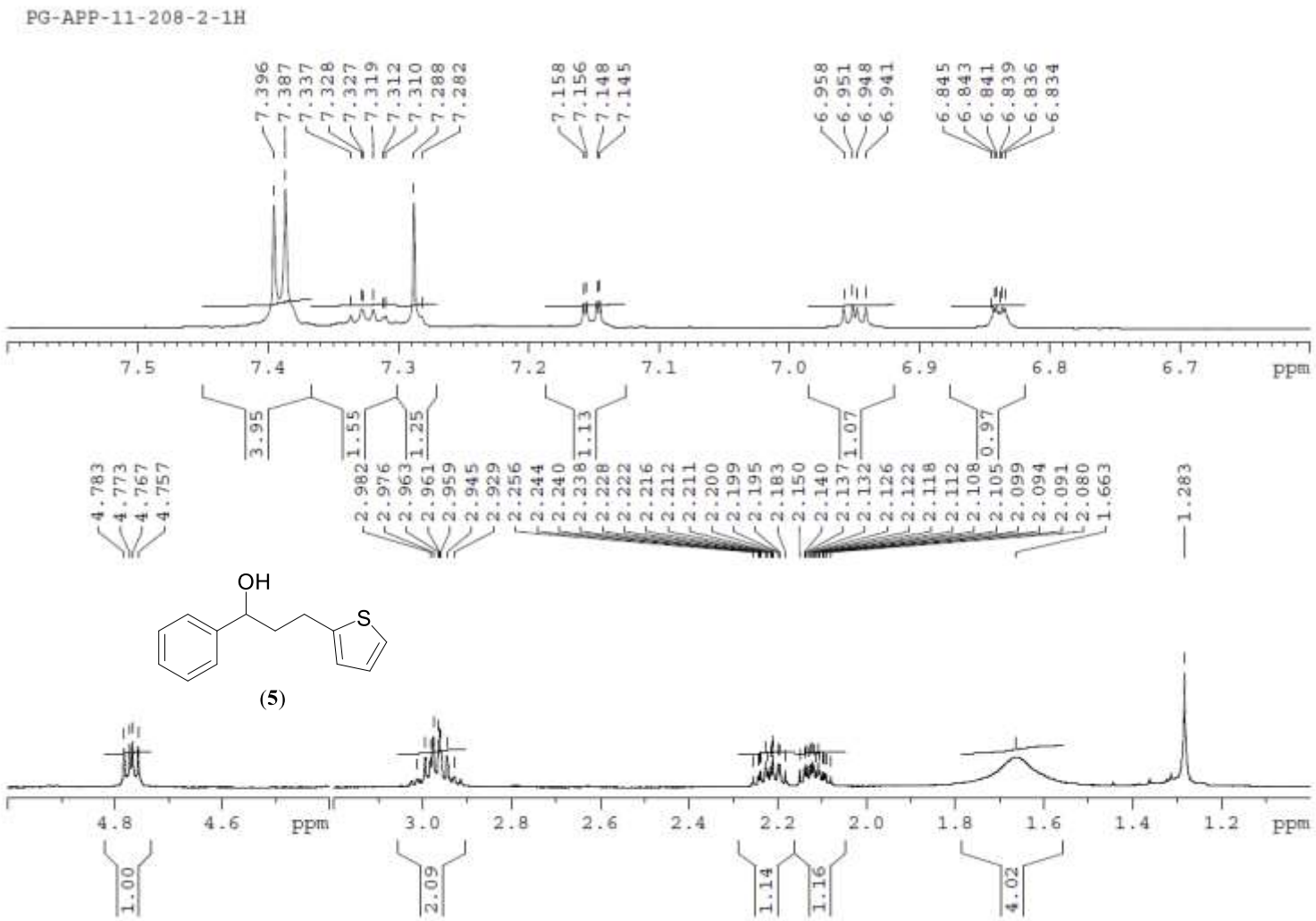


Figure S80. Expanded ^{1}H NMR spectrum of (**5**) in $CDCl_3$.

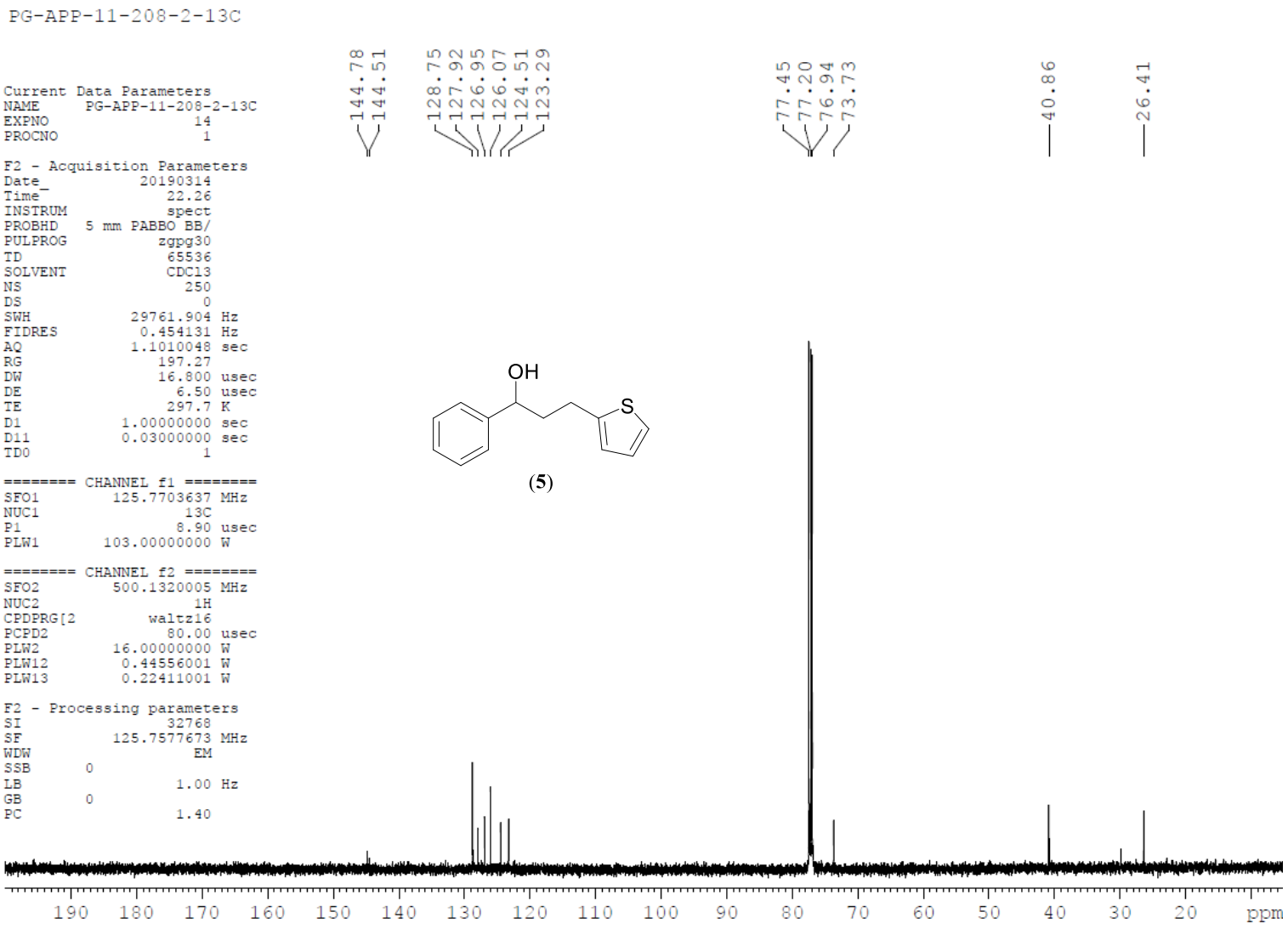


Figure S81. $^{13}C\{^{1}H\}$ NMR spectrum of (**5**) in $CDCl_3$.

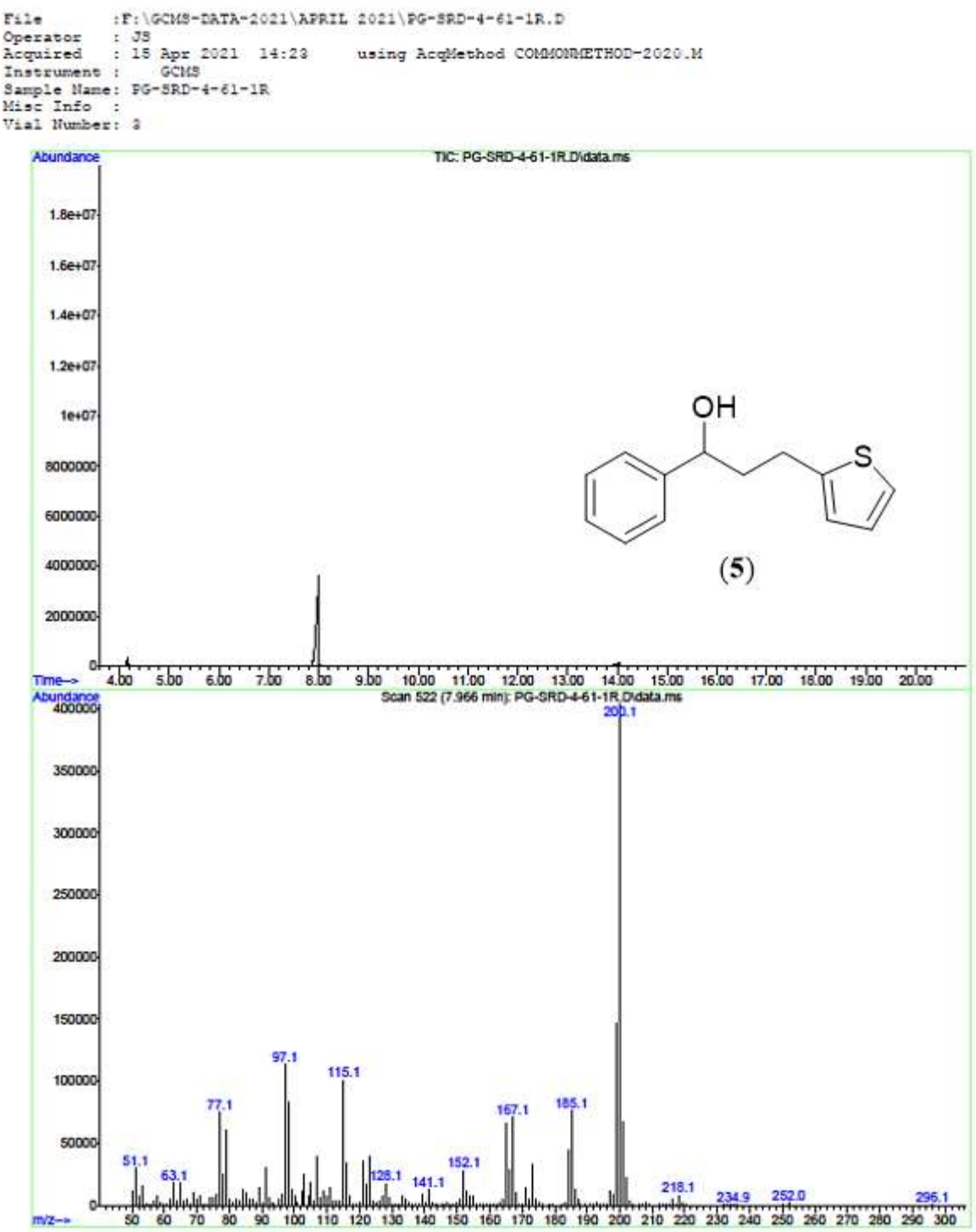


Figure S82. GCMS trace in EtOAc of (**5**) showing the M^+ peak at *m/z* 218.

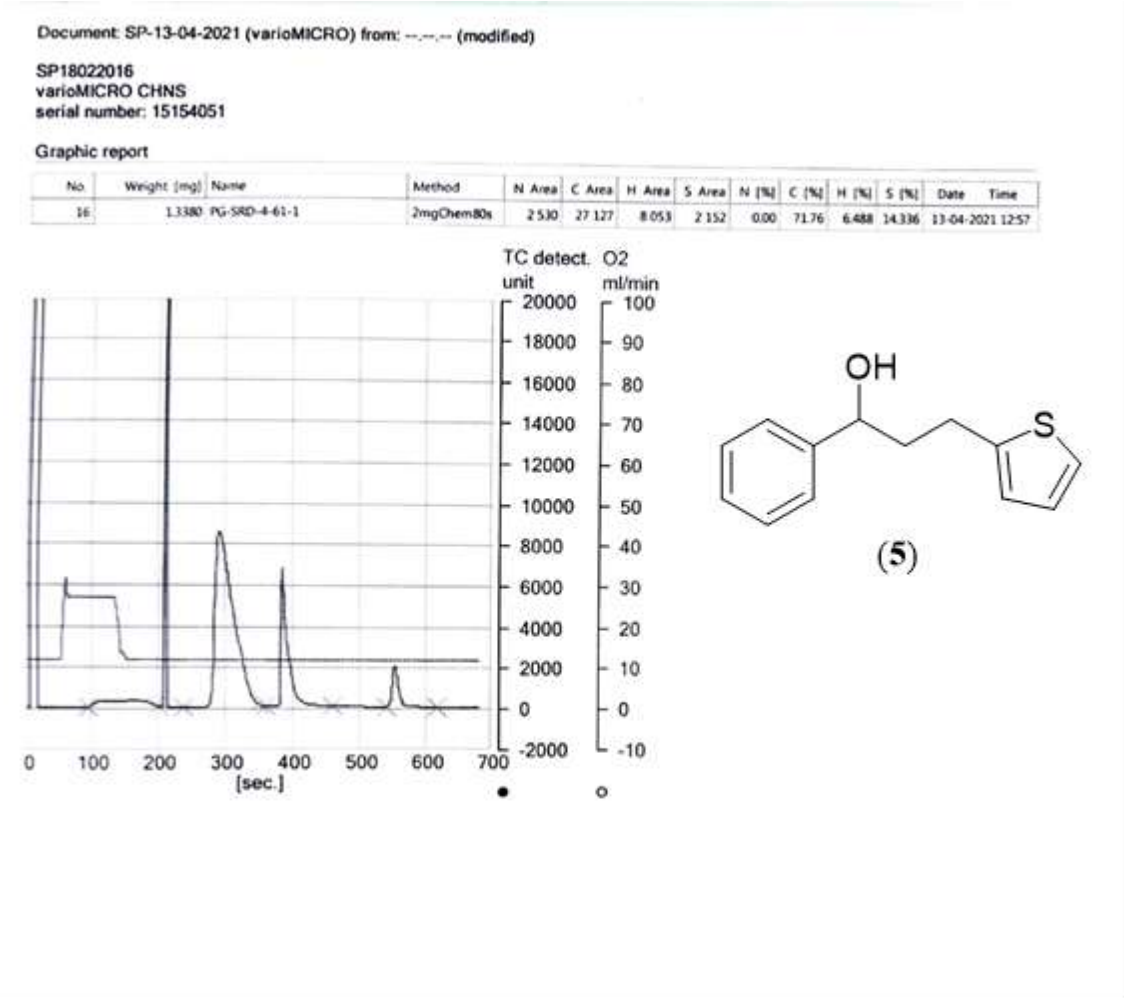


Figure S83. Elemental analysis data of (**5**).

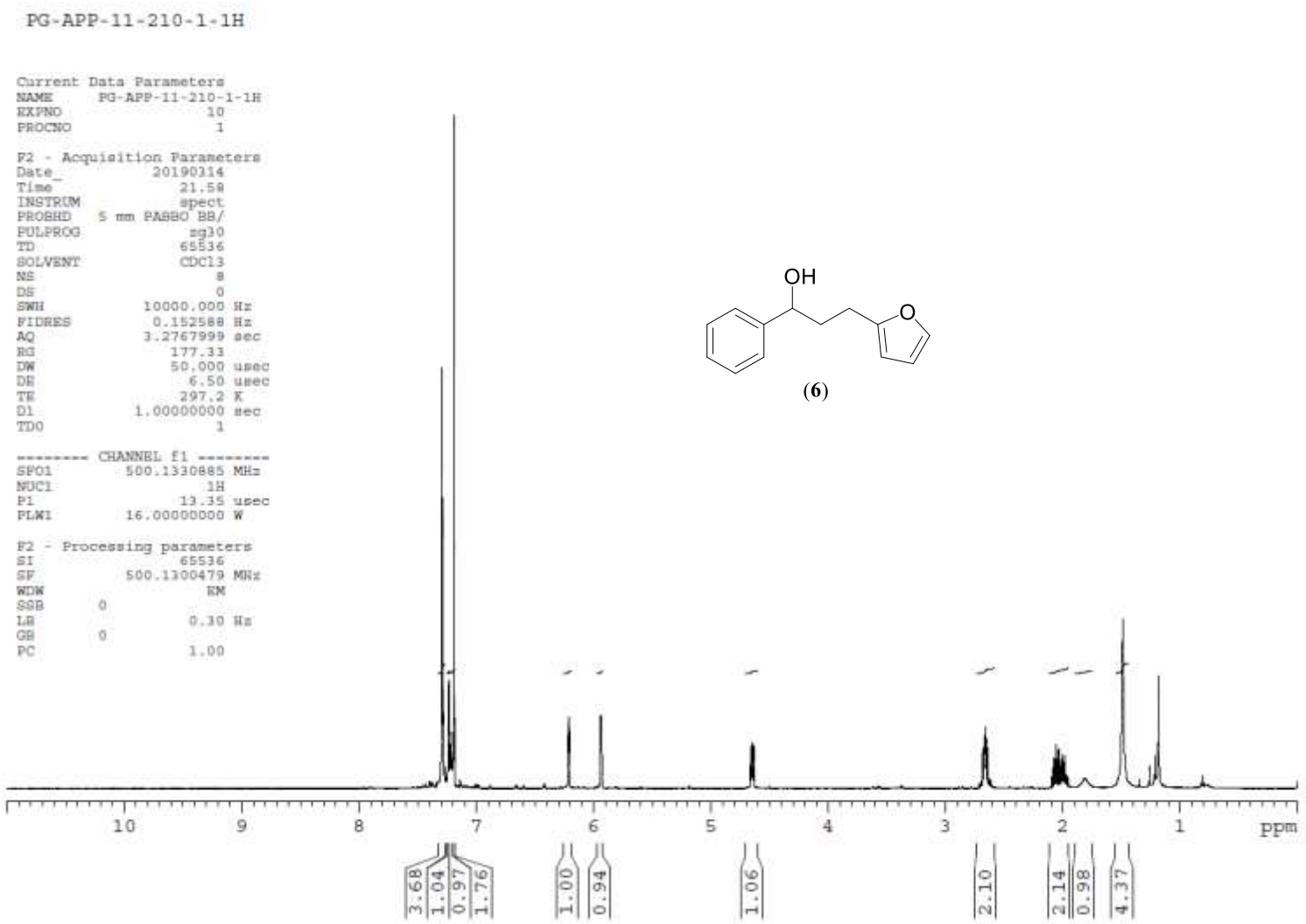


Figure S84. ^{1}H NMR spectrum of (**6**) in $CDCl_3$.

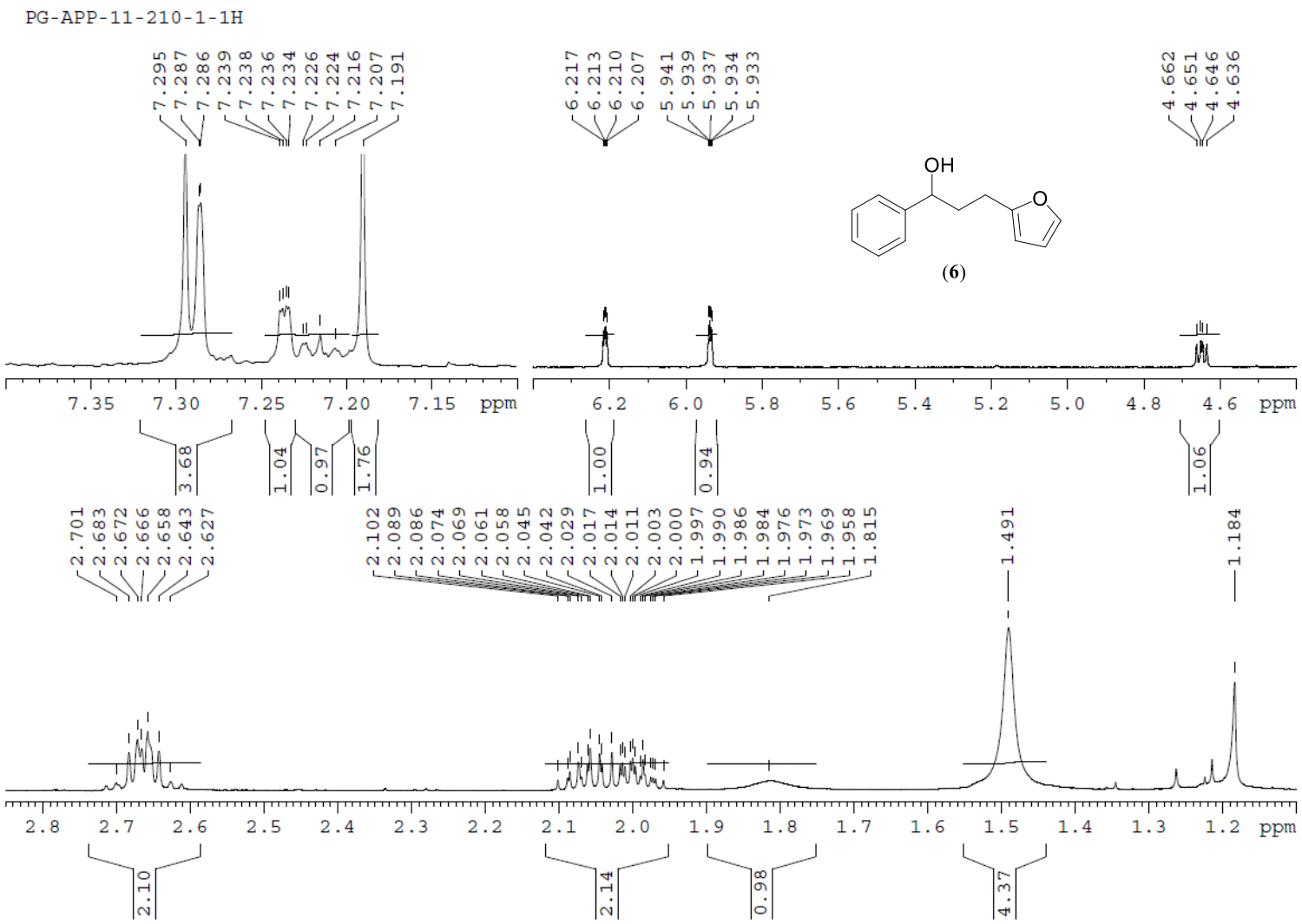


Figure S85. Expanded ^{1}H NMR spectrum of (**6**) in $CDCl_3$.

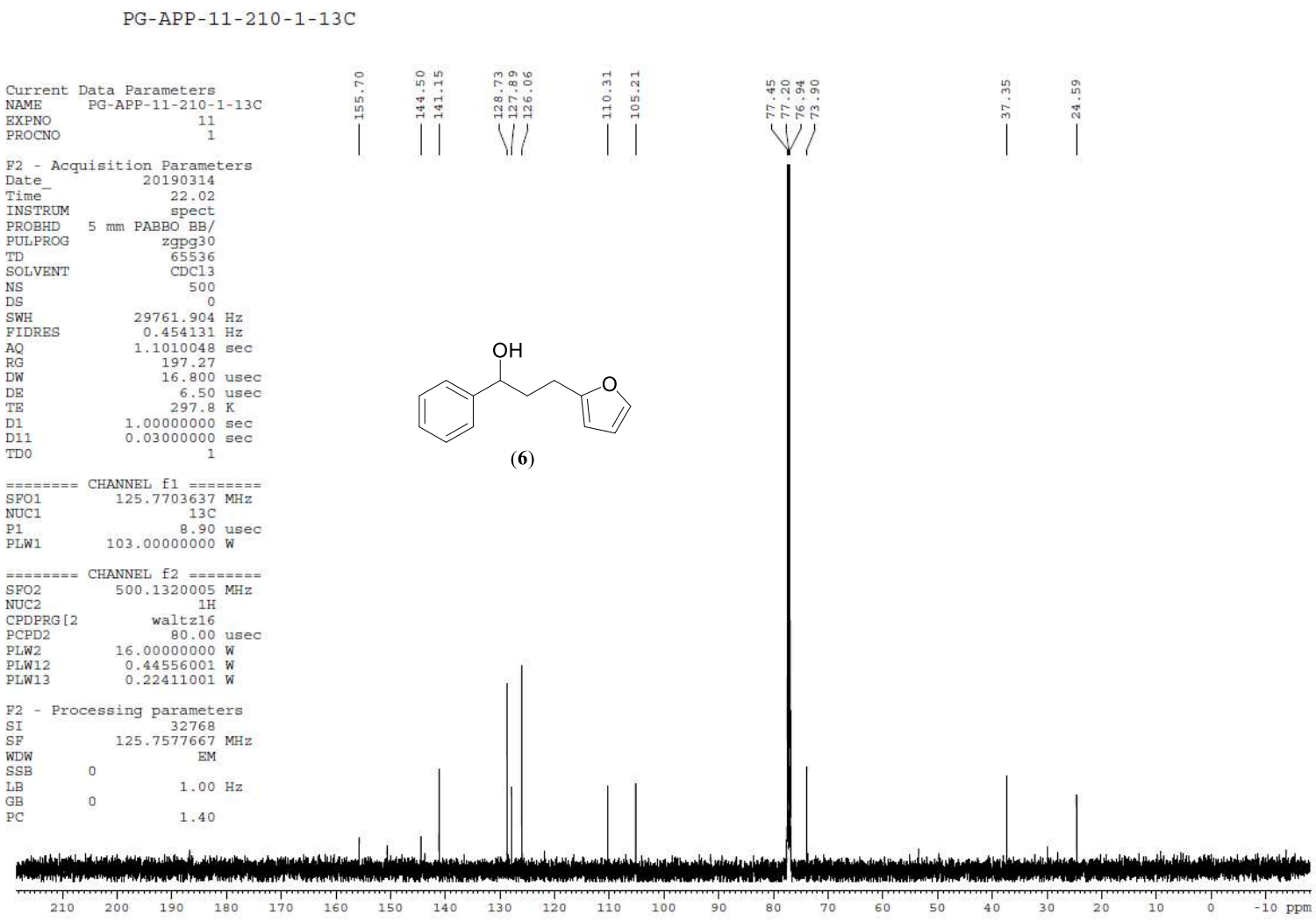


Figure S86. $^{13}C\{^{1}H\}$ NMR spectrum of (**6**) in $CDCl_3$.

```
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Operator   : APP
Acquired   : 13 Mar 2019  23:00     using AcqMethod COMMONMETHOD_2018.M
Instrument :     GCMS
Sample Name: PG-APP-11-210-28
Misc Info  :
Vial Number: 1
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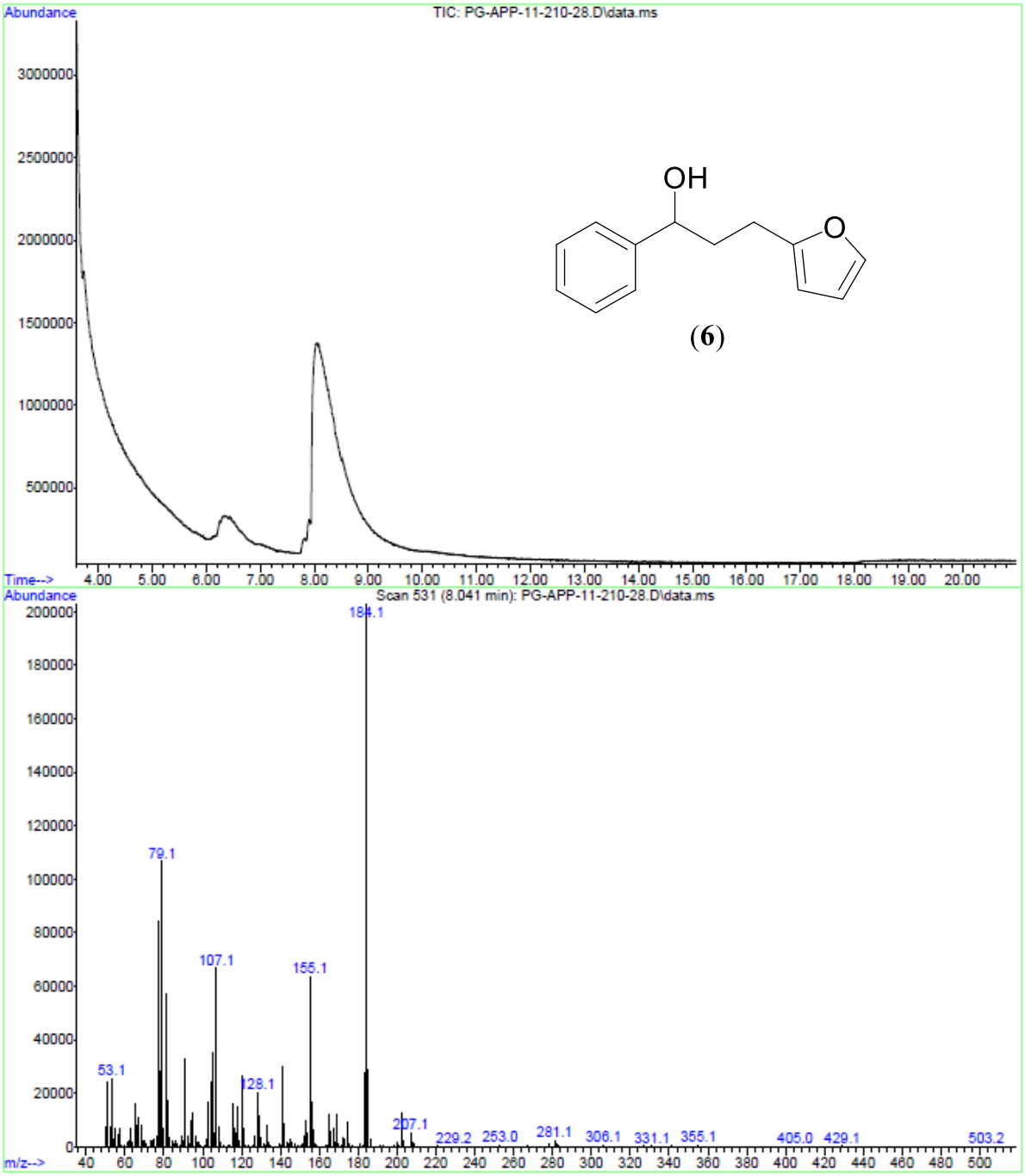
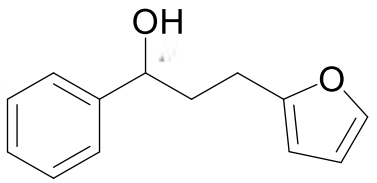


Figure S87. GCMS trace in EtOAc of (**6**) showing the M^+ peak at *m/z* 202.



(**6**)

Eager 300 Report

Page: 1 Sample: PG-APP-11-210-5 (PG-APP-11-210-5)

Method Name : PGAPP300519
Method File : D:\CHNS2019\PGAPP300519.mth
Chromatogram : PG-APP-11-210-5
Operator ID : Prakash
Analysed : 05/30/2019 17:32
Sample ID : PG-APP-11-210-5 (# 35)
Analysis Type : UnkNown (Area)

Company Name : C.E. Instruments
Printed : 5/30/2019 23:00
Instrument N. : Instrument #1
Sample weight : 1.462

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	21547	FU		0.0000
2	0.0000	6	123910	FU		0.0000
Carbon	77.7111	63	3035056	RS	1.000000	.267139E+07
Hydrogen	7.3492	187	734473	RS	4.132291	.683575E+07
Totals	85.0603		3914986			

Figure S88. Elemental analysis data of (**6**).

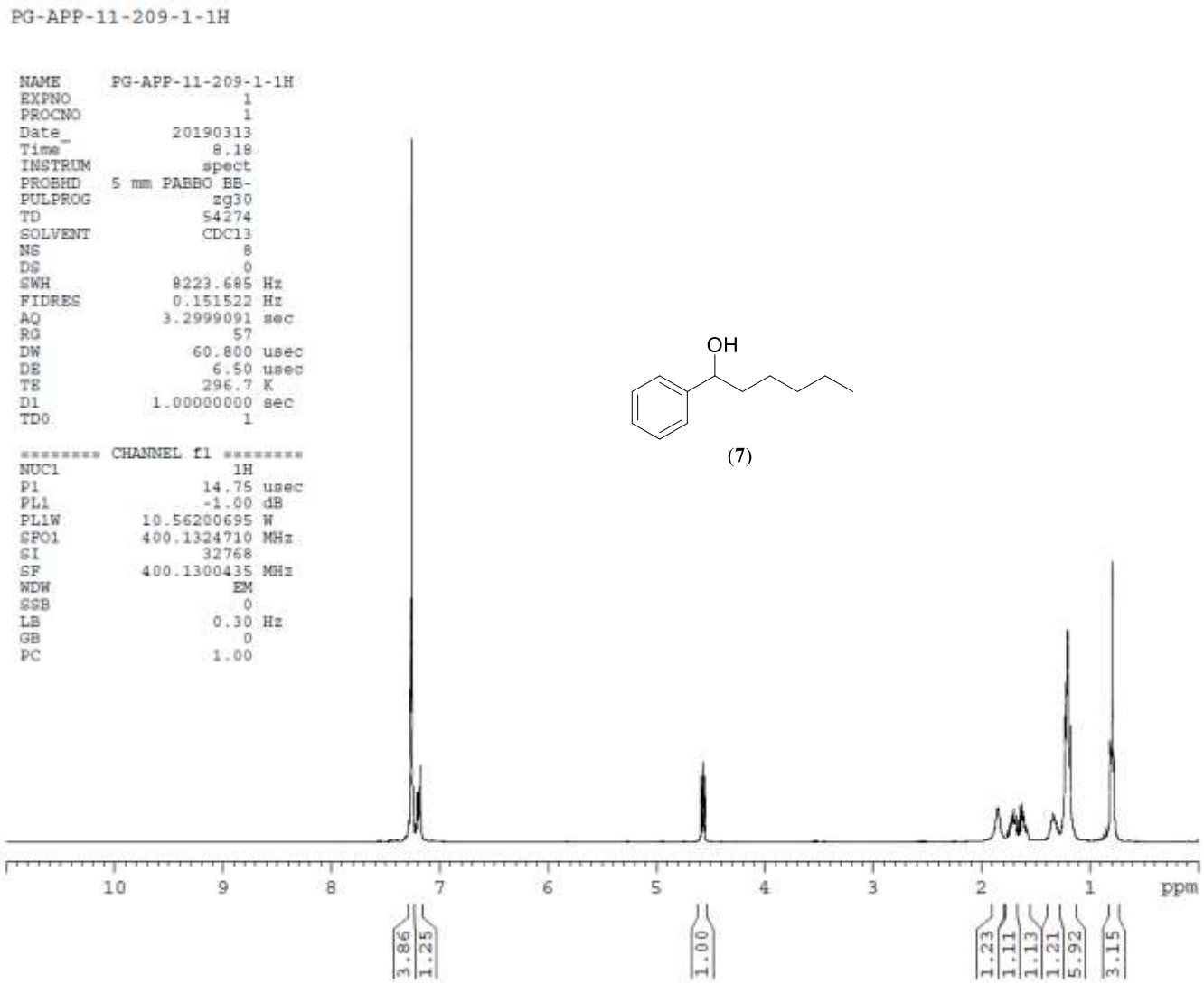


Figure S89. ^{1}H NMR spectrum of (**7**) in $CDCl_3$.

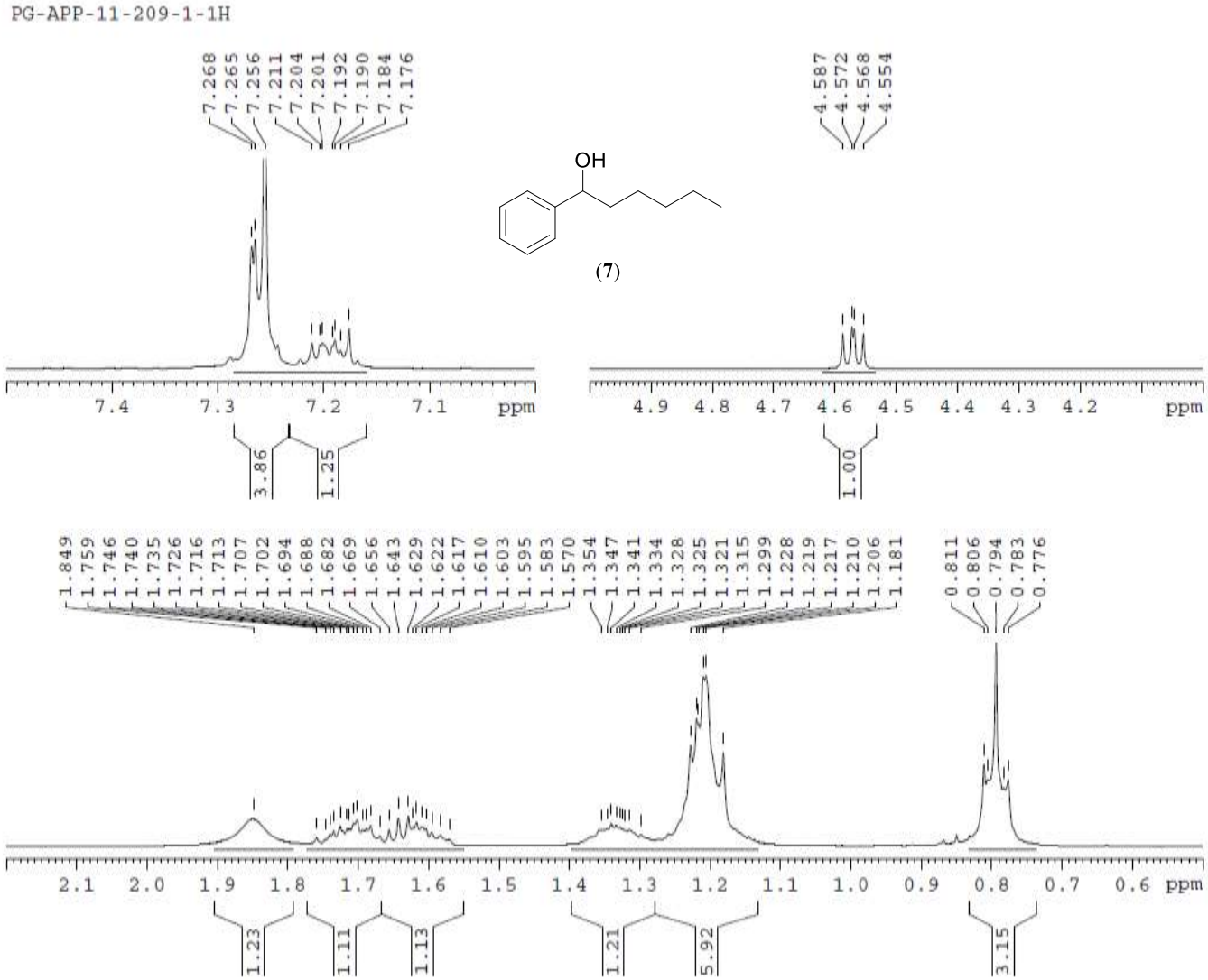


Figure S90. Expanded ^{1}H NMR spectrum of (**7**) in $CDCl_3$.

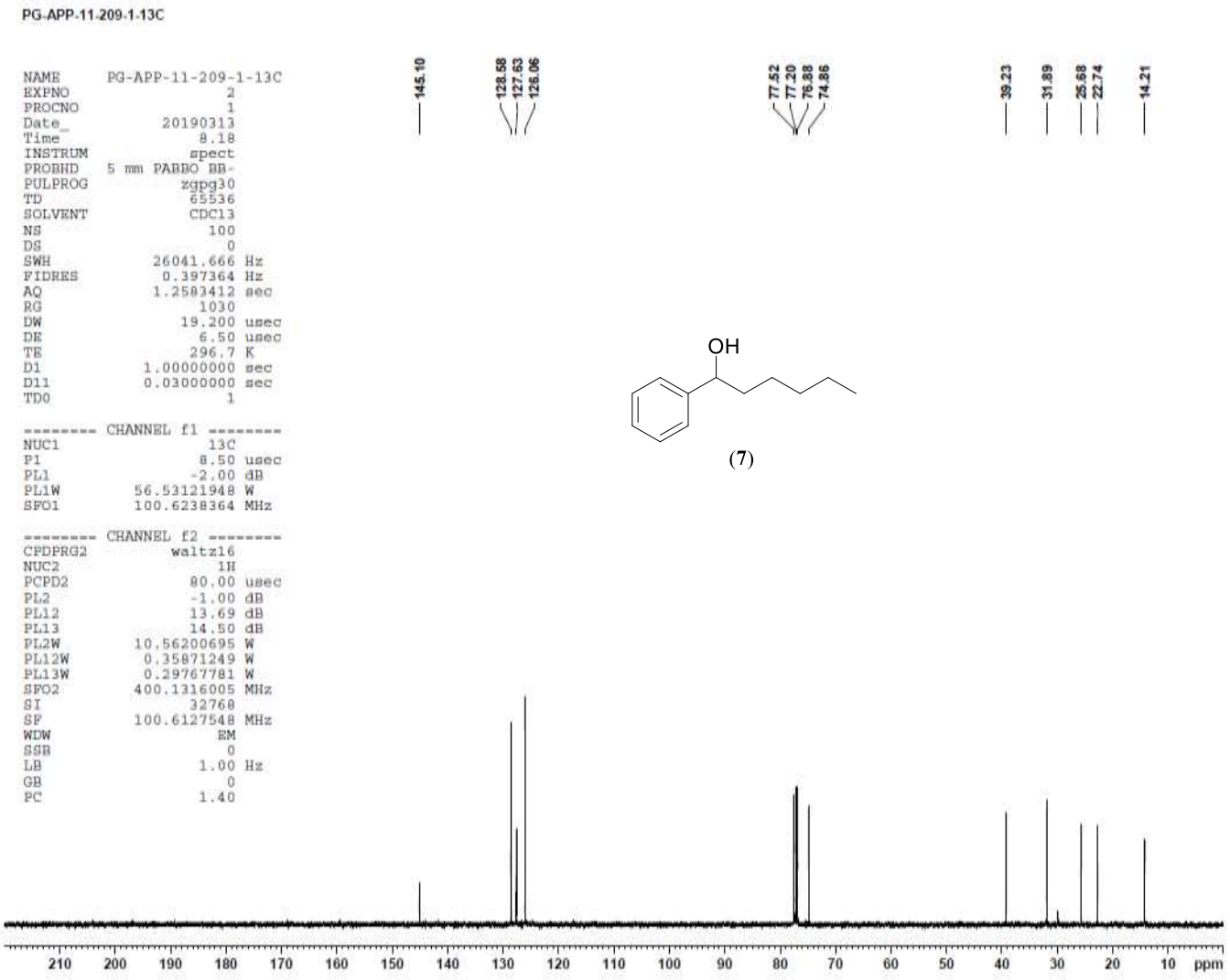


Figure S91. $^{13}C\{^{1}H\}$ NMR spectrum of (**7**) in $CDCl_3$.

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File       :F:\GCMSDATA2019\MAR 2019\PG-APP-11-209-2.D
Operator   : pg
Acquired   : 11 Mar 2019  21:53     using AcqMethod COMMONMETHOD_2018.M
Instrument :    GCMS
Sample Name: PG-APP-11-209-2
Misc Info  :
Vial Number: 1
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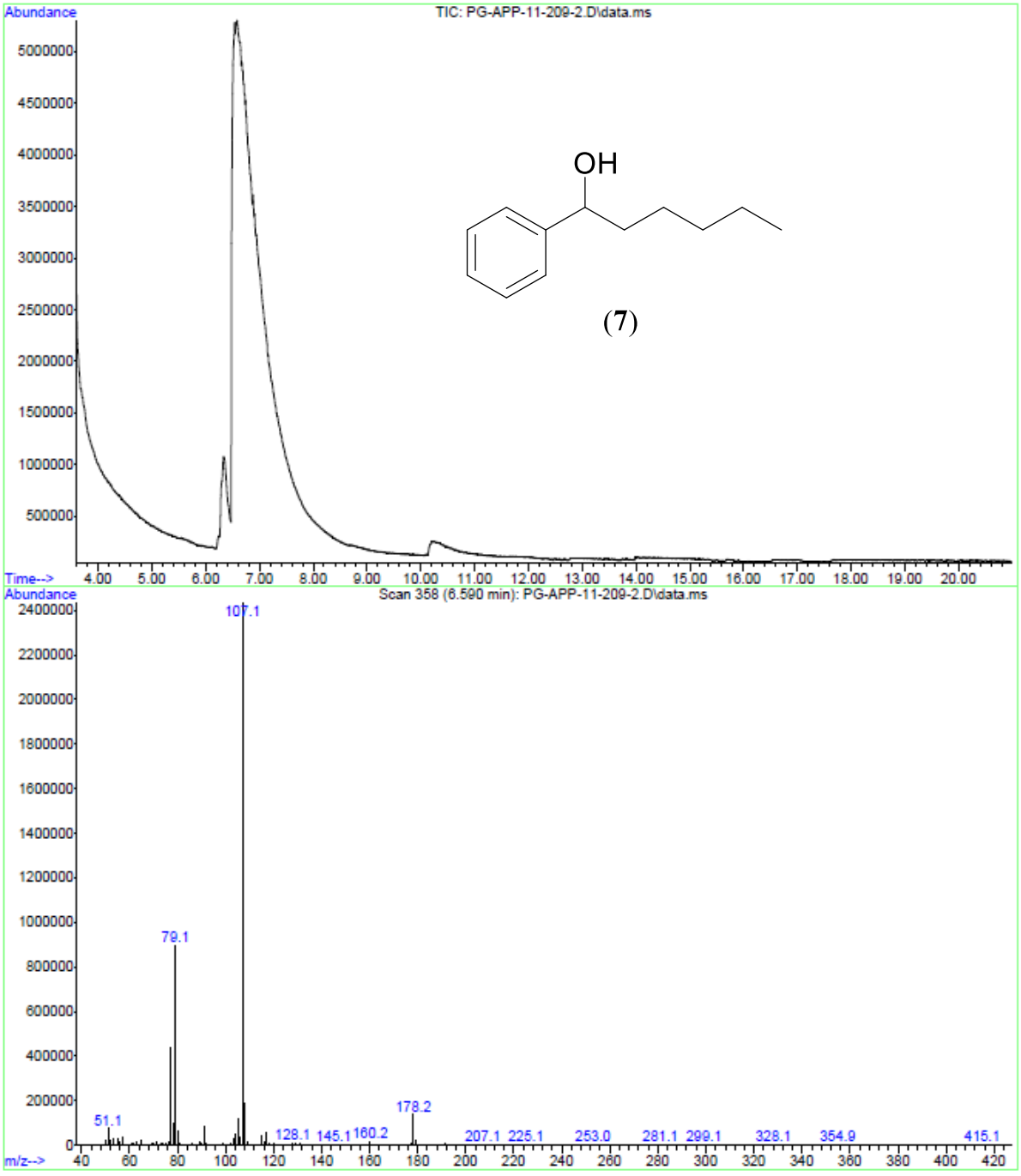
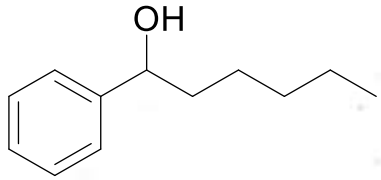


Figure S92. GCMS trace in EtOAc of (**7**) showing the M^+ peak at *m/z* 178.



(7)

Eager 300 Report

Page: 1 Sample: PG-APP-11-209-2 (PG-APP-11-209-2)

Method Name	: PGAPP060619		
Method File	: D:\CHNS2019\PGAPP060619.mth		
Chromatogram	: PG-APP-11-209-2		
Operator ID	: Prakash	Company Name	: C.E. Instruments
Analysed	: 06/06/2019 15:18	Printed	: 6/7/2019 08:27
Sample ID	: PG-APP-11-209-2 (# 16)	Instrument N.	: Instrument #1
Analysis Type	: UnkNown (Area)	Sample weight	: .979

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	141541	RS		0.0000
Nitrogen	6.7411	48	193184	FU	10.811430	.103544E+07
Carbon	80.0470	64	2088598	FU	1.000000	.265769E+07
Hydrogen	10.0655	184	646869	RS	3.228780	.656444E+07
Totals	96.8536		3070192			

Figure S93. Elemental analysis data of (**7**).

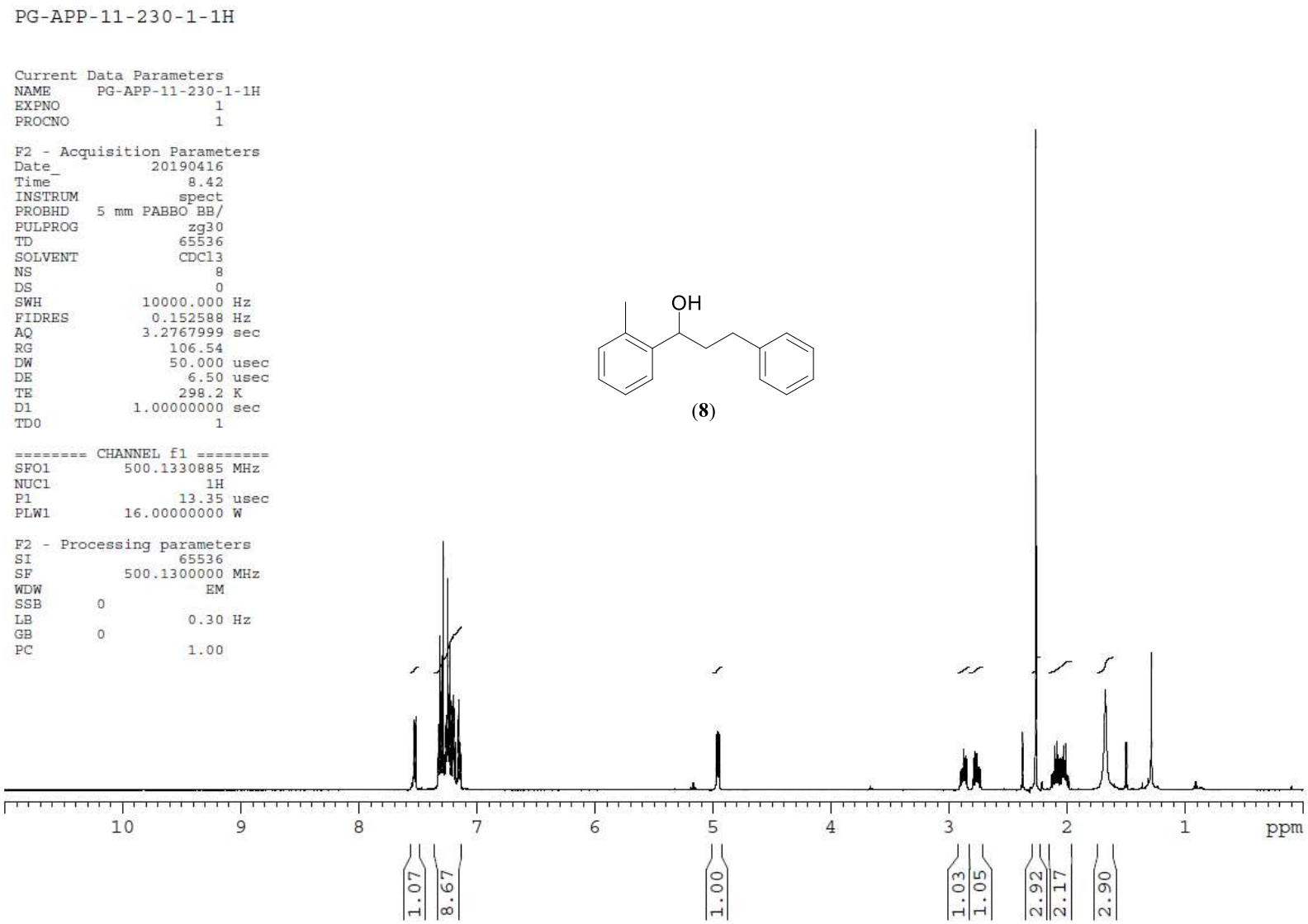


Figure S94. ^{1}H NMR spectrum of (**8**) in $CDCl_3$.

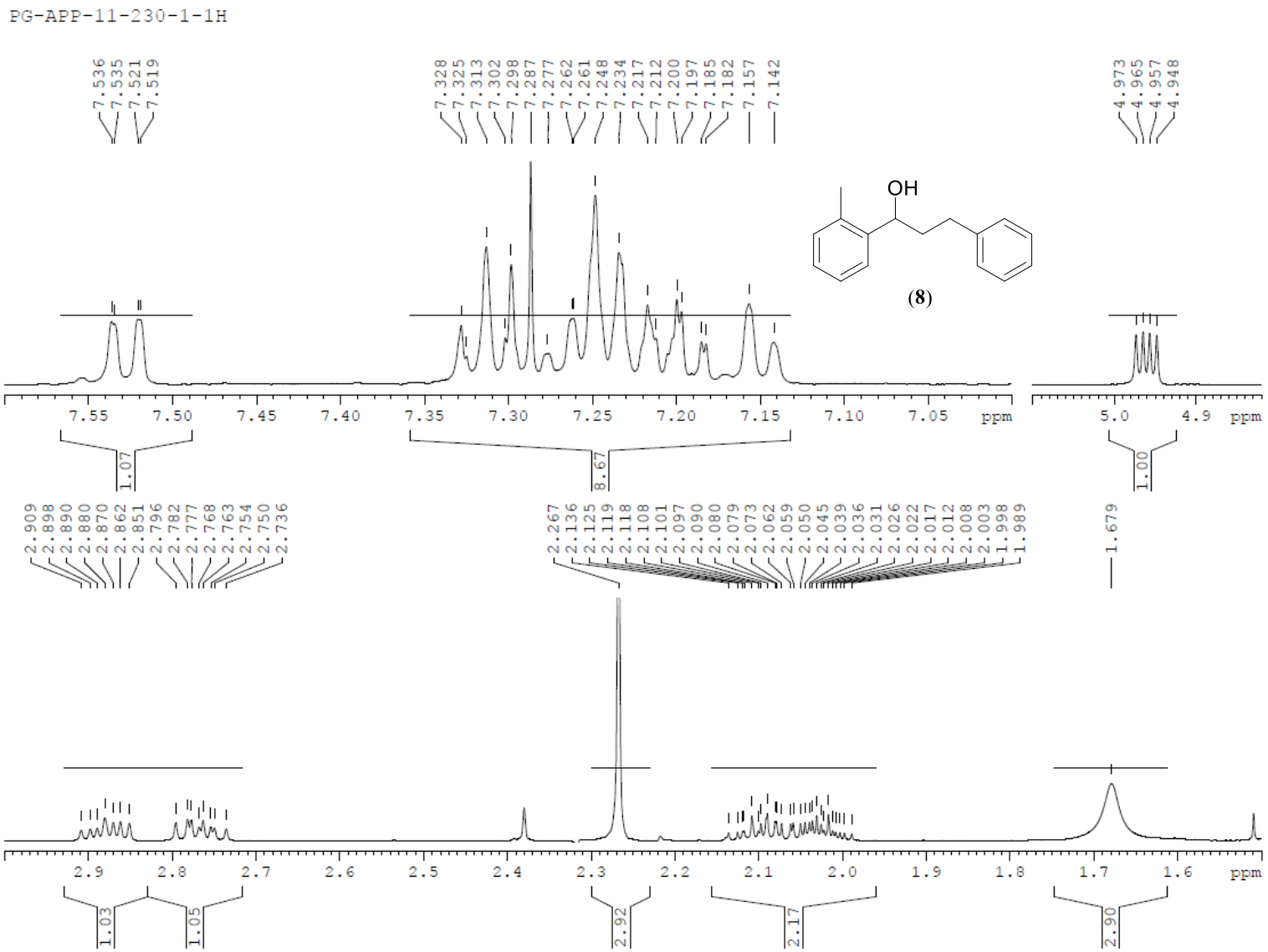


Figure S95. Expanded ^{1}H NMR spectrum of (**8**) in $CDCl_3$.

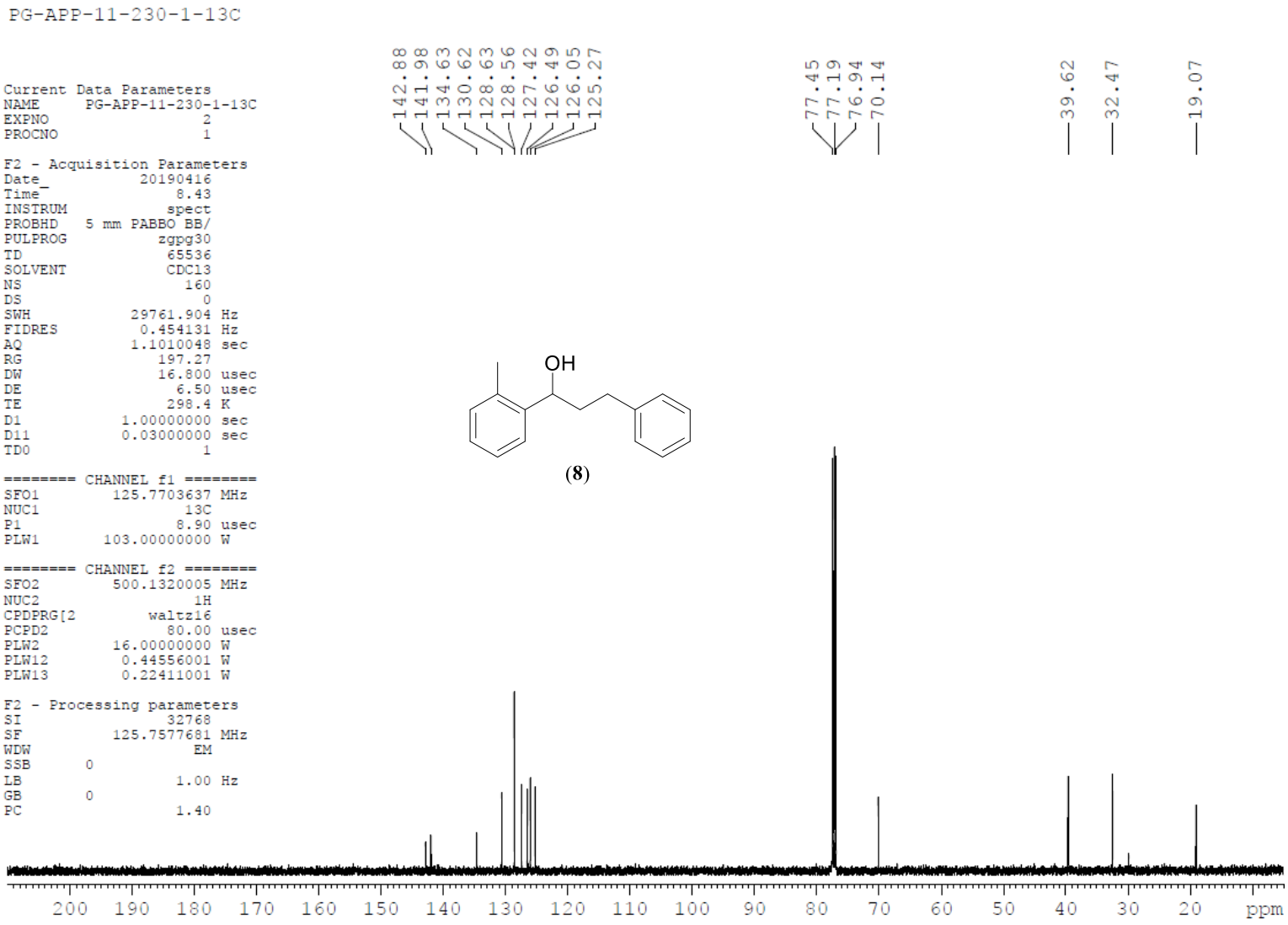


Figure S96. $^{13}C\{^{1}H\}$ NMR spectrum of (**8**) in $CDCl_3$.

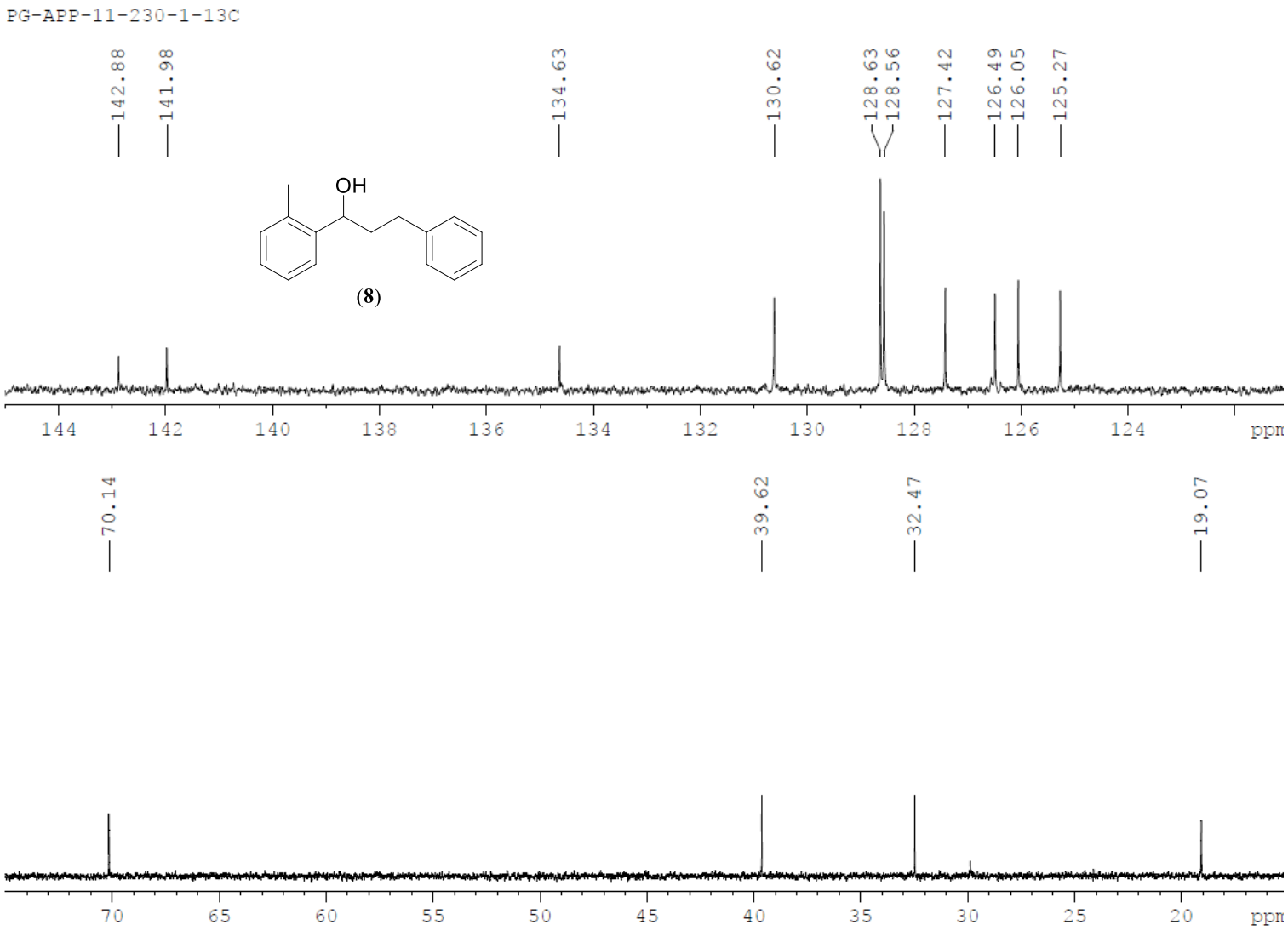


Figure S97. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**8**) in $CDCl_3$.

```
File       :F:\GCMSDATA2019\APR2019\PG-APP-11-230-29.D
Operator   : APP
Acquired   : 15 Apr 2019  20:34     using AcqMethod COMMONMETHOD_2016.M
Instrument :    GCMS
Sample Name: PG-APP-11-230-29
Misc Info  :
Vial Number: 5
```

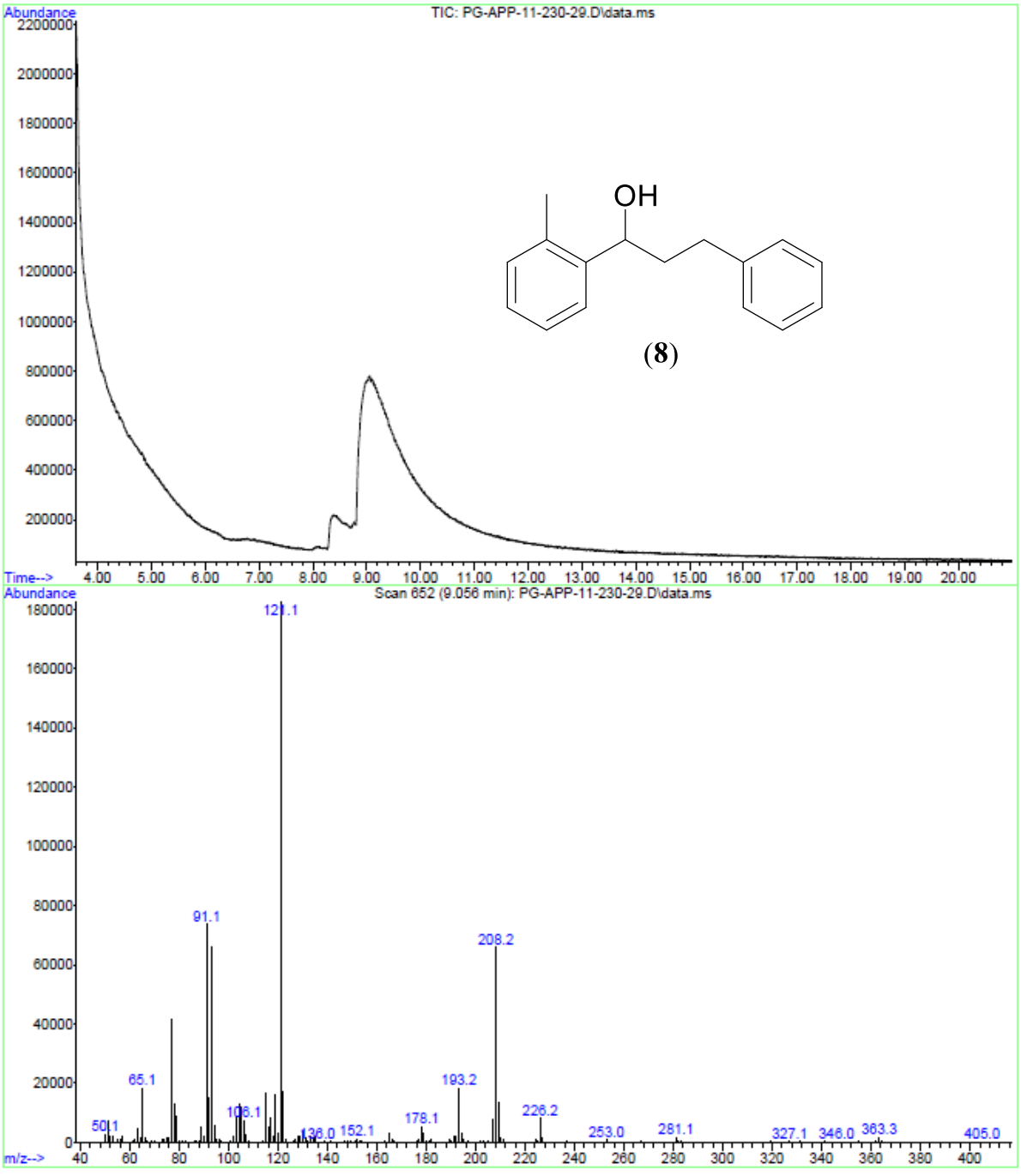
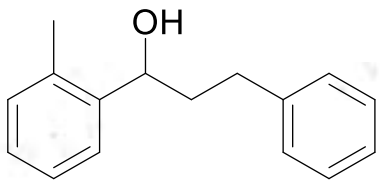


Figure S98. GCMS trace in EtOAc of (**8**) showing the M^+ peak at *m/z* 226.



(**8**)

Eager 300 Report

Page: 1 Sample: PG-APP-11-230-1 (PG-APP-11-230-1)

Method Name	: PGAPP240519		
Method File	: D:\CHNS2019\PGAPP240519.mth		
Chromatogram	: PG-APP-11-230-1		
Operator ID	: Prakash	Company Name	: C.E. Instruments
Analysed	: 05/24/2019 18:48	Printed	: 5/25/2019 00:21
Sample ID	: PG-APP-11-230-1 (# 34)	Instrument N.	: Instrument #1
Analysis Type	: UnkNown (Area)	Sample weight	: 1.244

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	135010	RS		0.0000
Carbon	84.7228	62	2809745	RS	1.000000	.266592E+07
Hydrogen	8.2142	187	610397	RS	4.603144	.597347E+07
Totals	92.9370		3555152			

Figure S99. Elemental analysis data of (**8**).

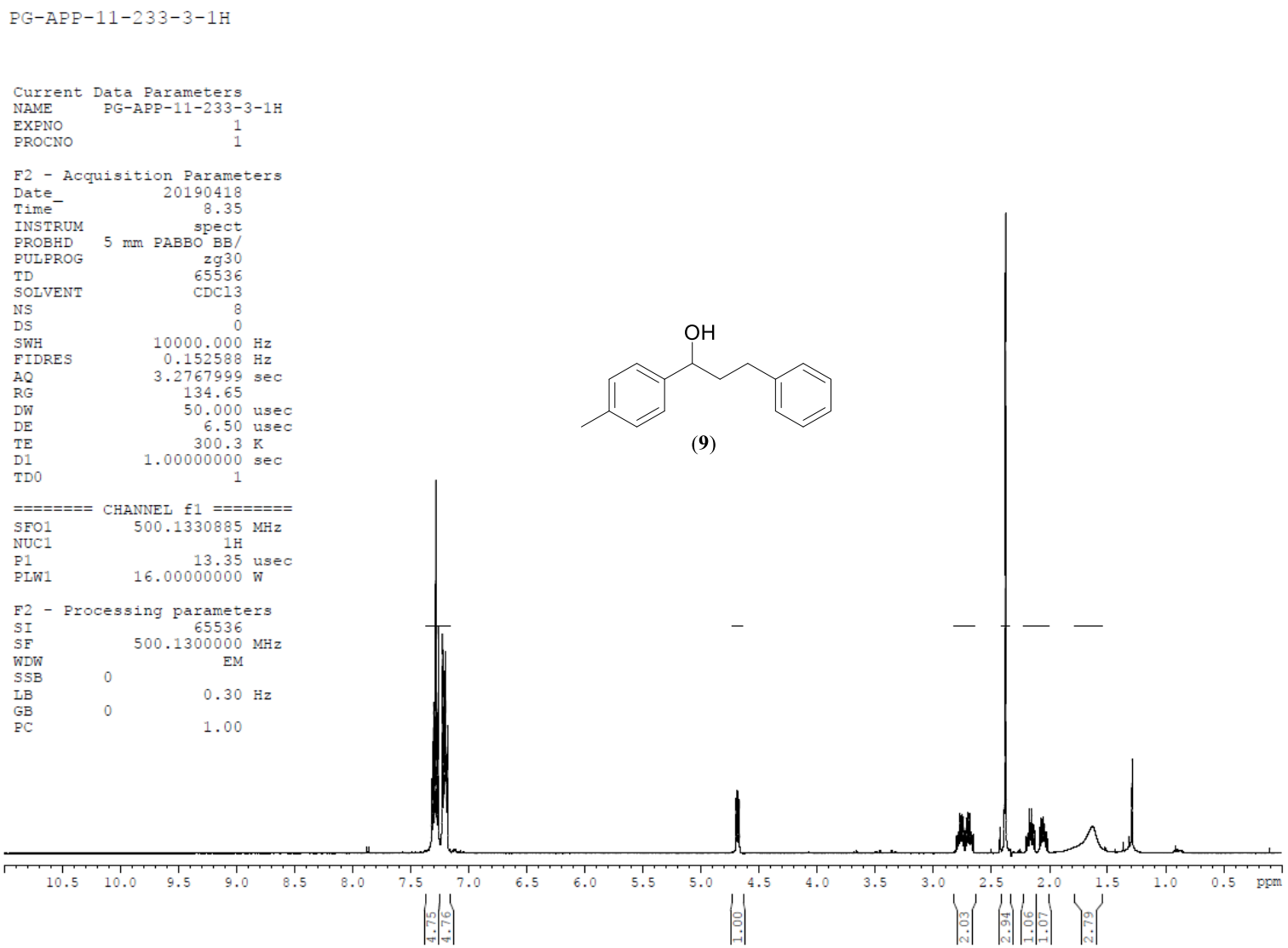


Figure S100. ^{1}H NMR spectrum of (**9**) in $CDCl_3$.

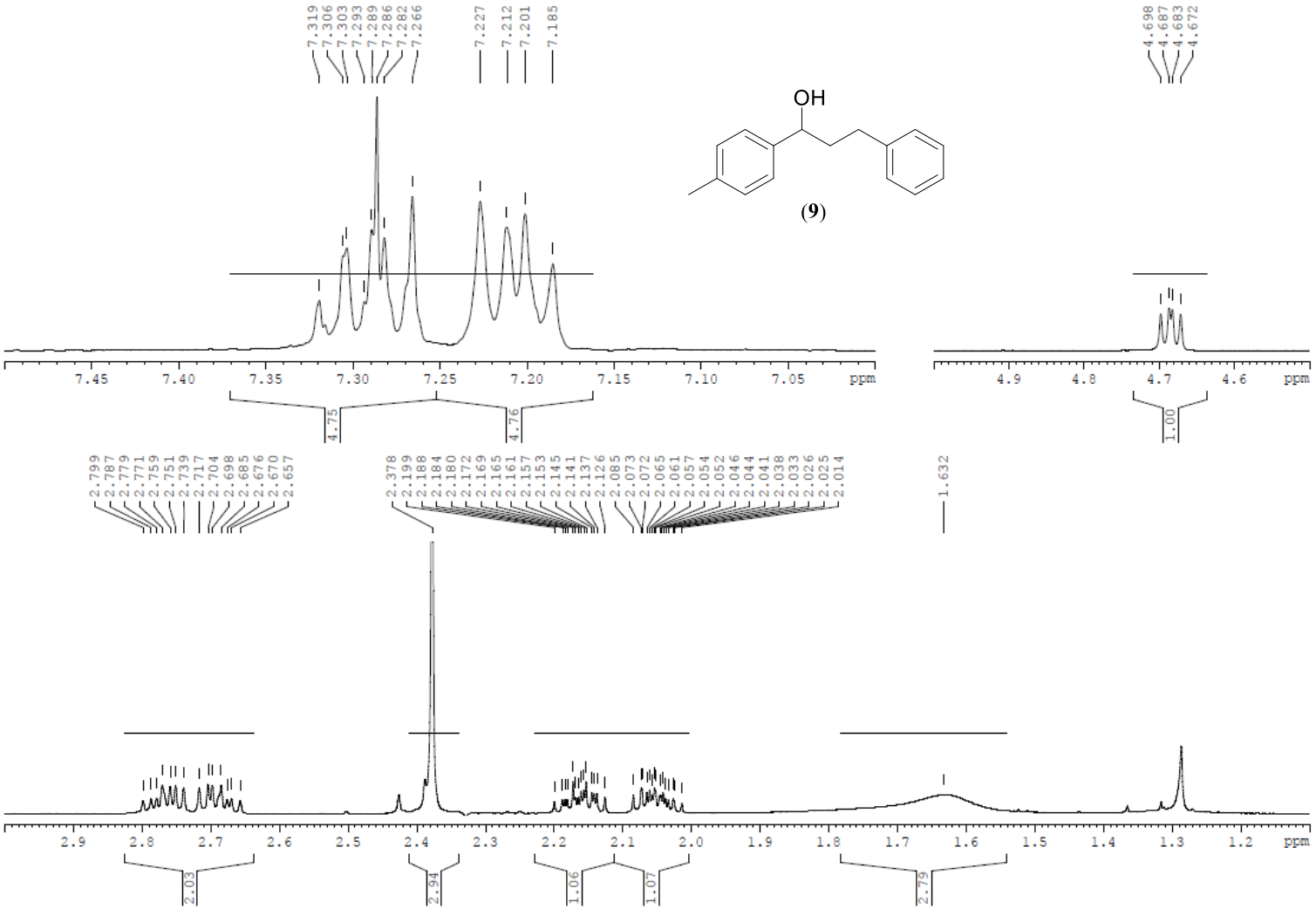


Figure S101. Expanded ^{1}H NMR spectrum of (**9**) in $CDCl_3$.

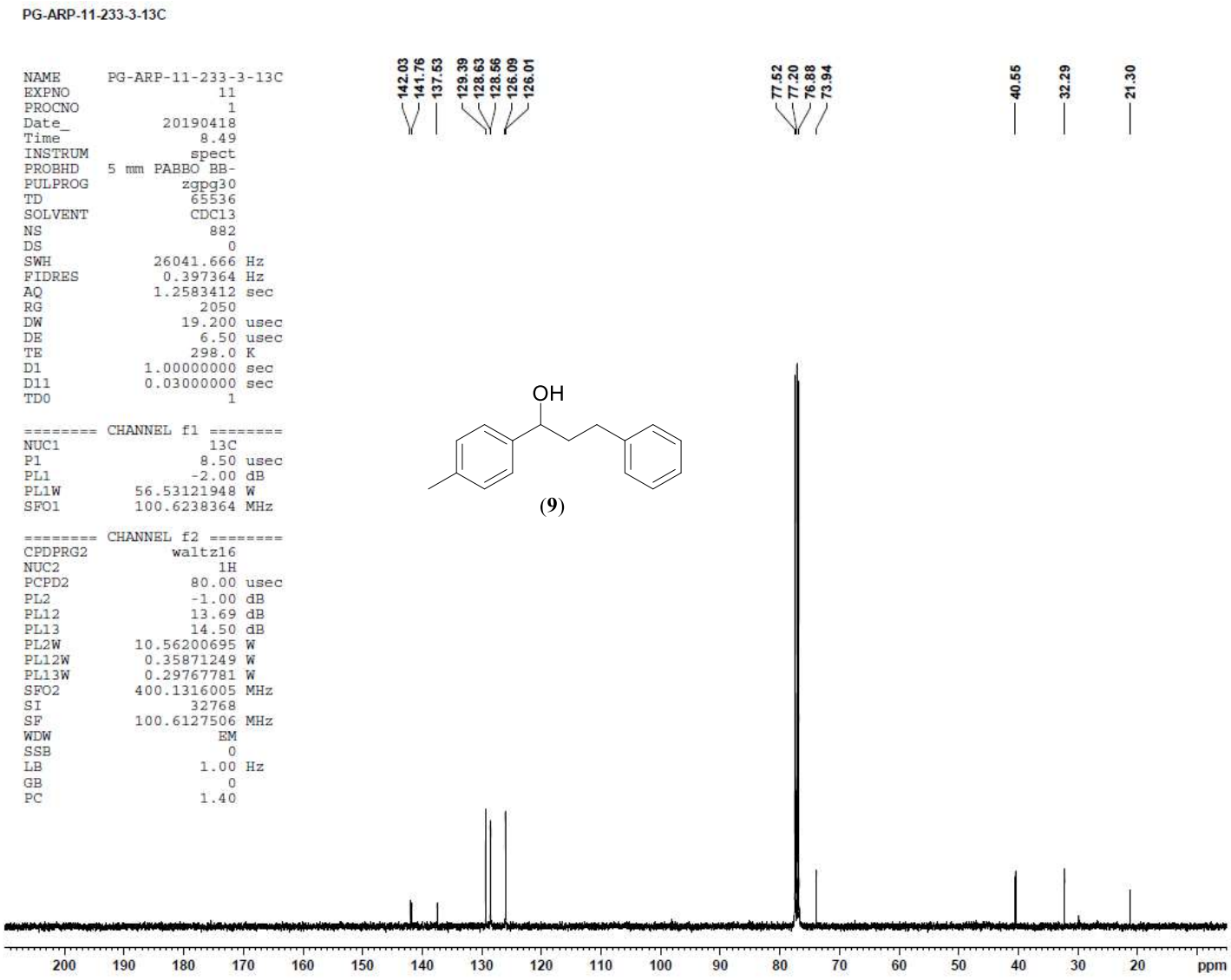


Figure S102. $^{13}C\{^{1}H\}$ NMR spectrum of (**9**) in $CDCl_3$.

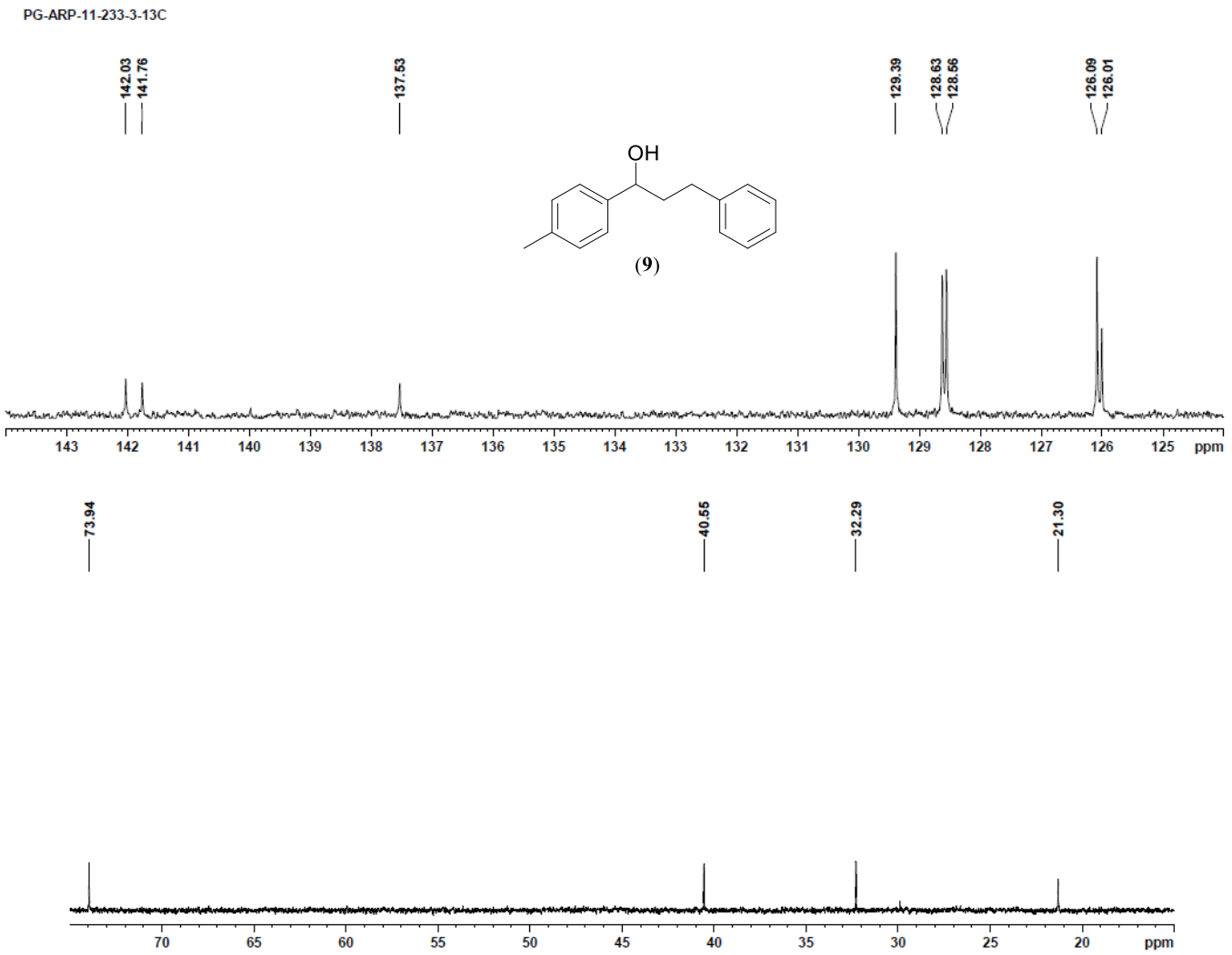


Figure S103. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**9**) in $CDCl_3$.

```
File       :F:\GCMSDATA2019\APR2019\PG-APP-11-235-3.D
Operator   : APP
Acquired   : 17 Apr 2019  16:43     using AcqMethod COMMONMETHOD_2018.M
Instrument :    GCMS
Sample Name: PG-APP-11-235-3
Misc Info  :
Vial Number: 1
```

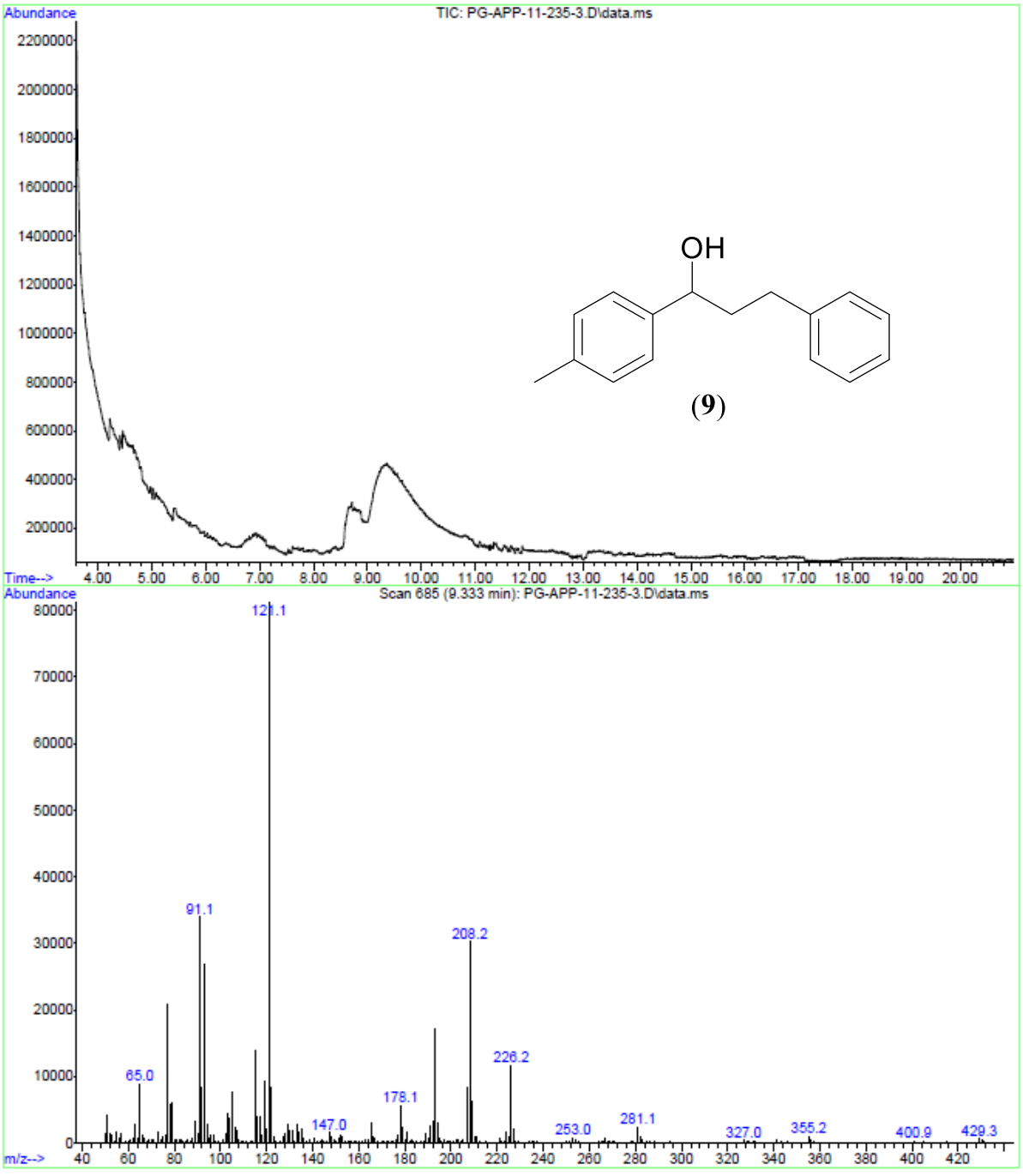
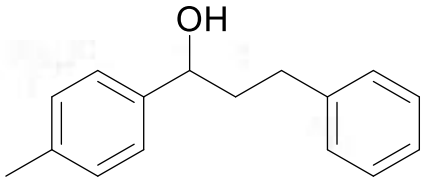


Figure S104. GCMS trace in EtOAc of (**9**) showing the M^+ peak at *m/z* 226.



(**9**)

Eager 300 Report

Page: 1 Sample: PG-APP-11-233-1 (PG-APP-11-233-1)

Method Name : PGAPP240519
Method File : D:\CHNS2019\PGAPP240519.mth
Chromatogram : PG-APP-11-233-1
Operator ID : Prakash Company Name : C.E. Instruments
Analysed : 05/24/2019 19:18 Printed : 5/25/2019 00:22
Sample ID : PG-APP-11-233-1 (# 37) Instrument N. : Instrument #1
Analysis Type : UnkNown (Area) Sample weight : .876

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	134852	RS		0.0000
Carbon	84.8281	63	1981027	RS	1.000000	.266592E+07
Hydrogen	7.8253	184	409480	RS	4.837908	.597347E+07
Totals	92.6534		2525359			

Figure S105. Elemental analysis data of (**9**).

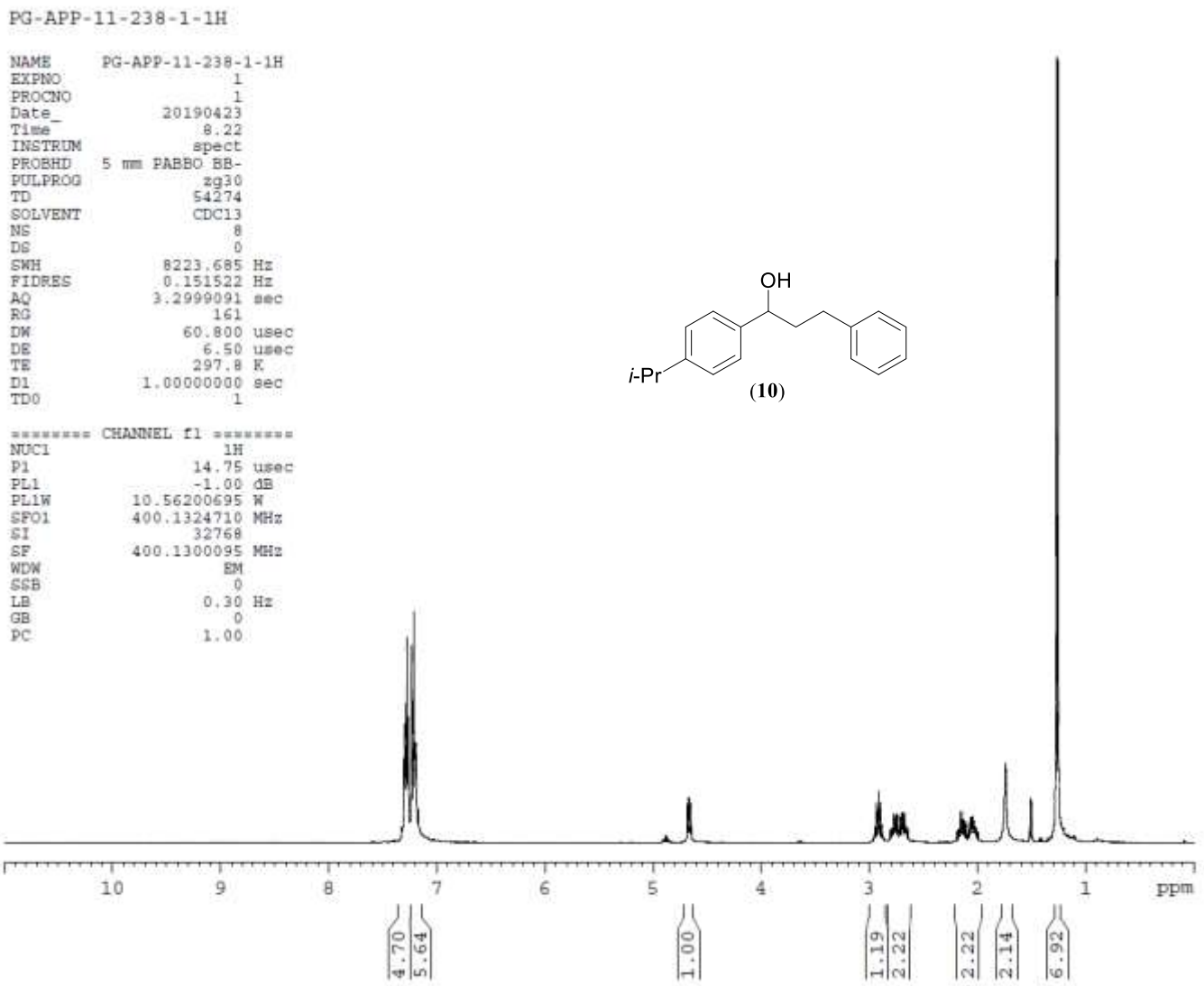


Figure S106. ^{1}H NMR spectrum of (**10**) in $CDCl_3$.

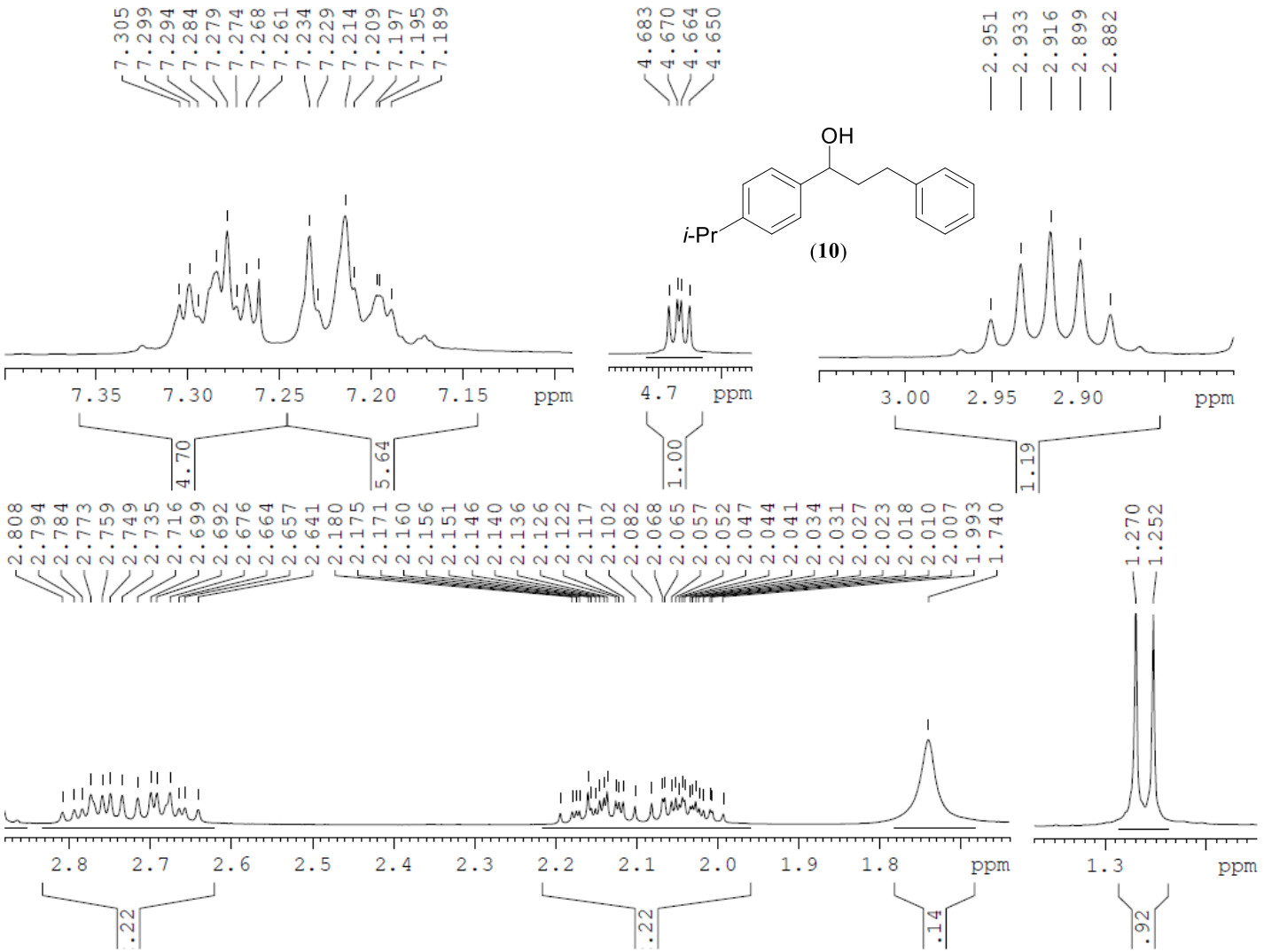


Figure S107. Expanded ^{1}H NMR spectrum of (**10**) in $CDCl_3$.

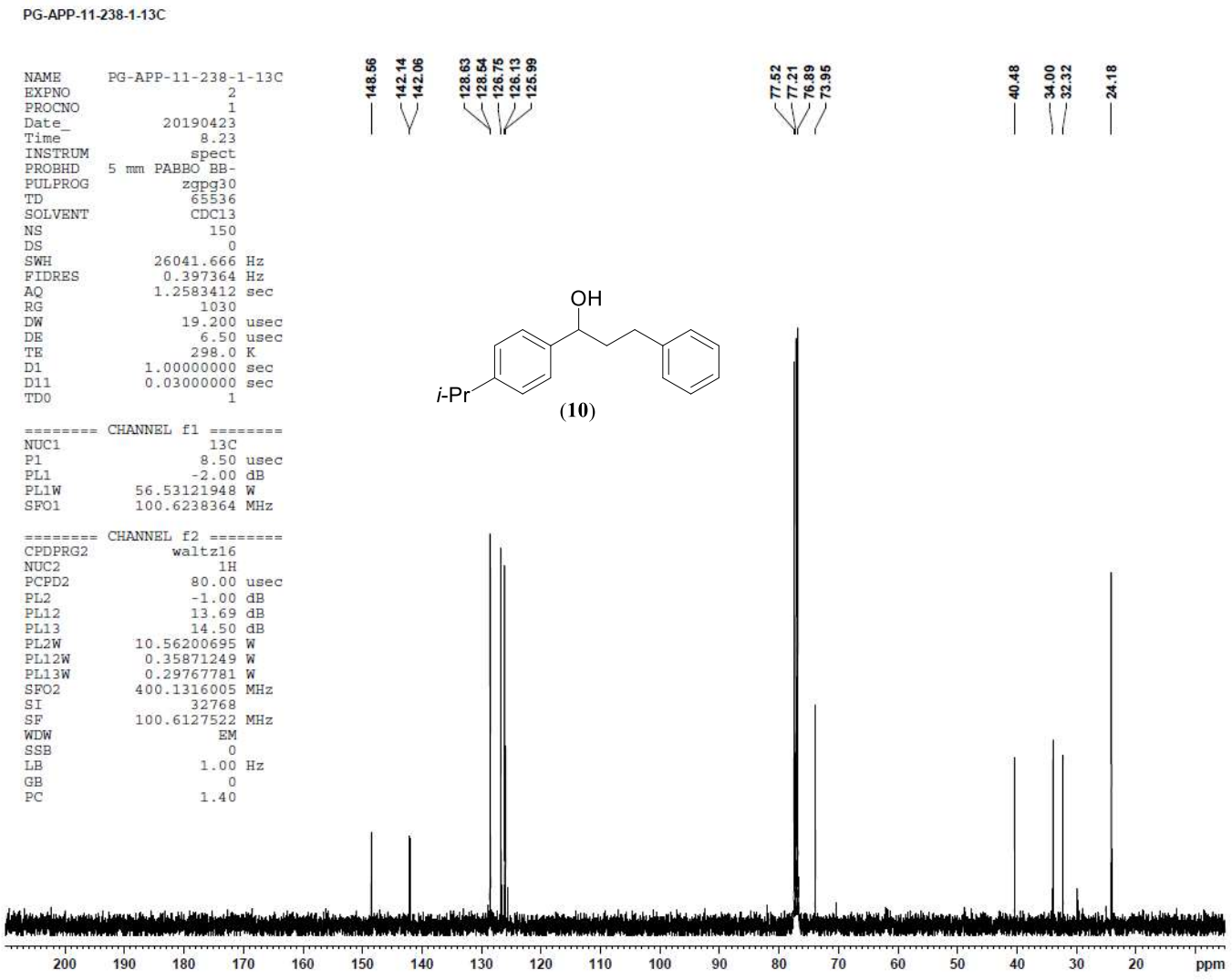


Figure S108. $^{13}C\{^{1}H\}$ NMR spectrum of (**10**) in $CDCl_3$.

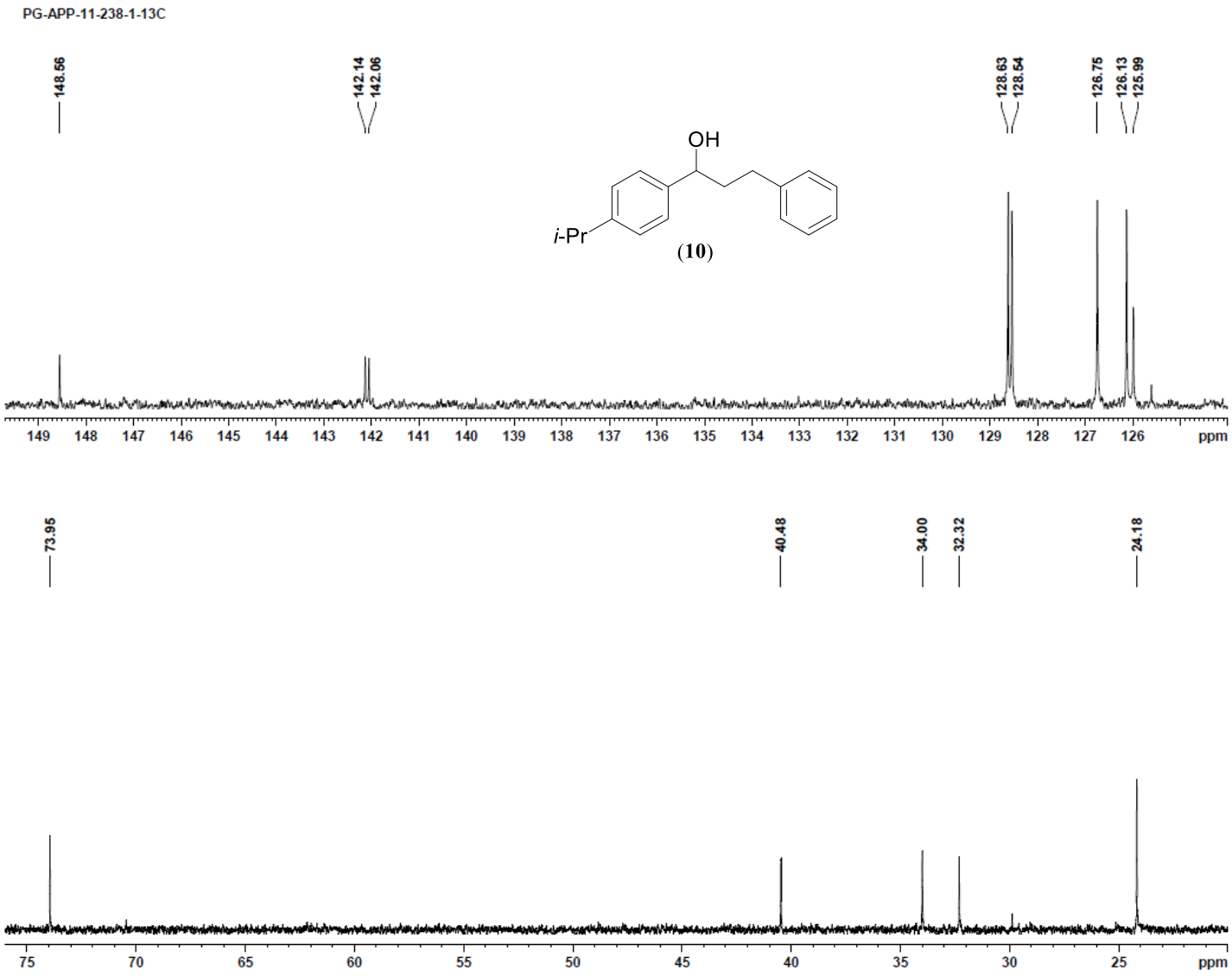


Figure S109. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**10**) in $CDCl_3$.

```
File       :F:\GCMSDATA2019\APR2019\PG-APP-11-238-4.D
Operator   : APP
Acquired   : 22 Apr 2019  20:09     using AcqMethod COMMONMETHOD_2018.M
Instrument :      GCMS
Sample Name: PG-APP-11-238-4
Misc Info  :
Vial Number: 2
```

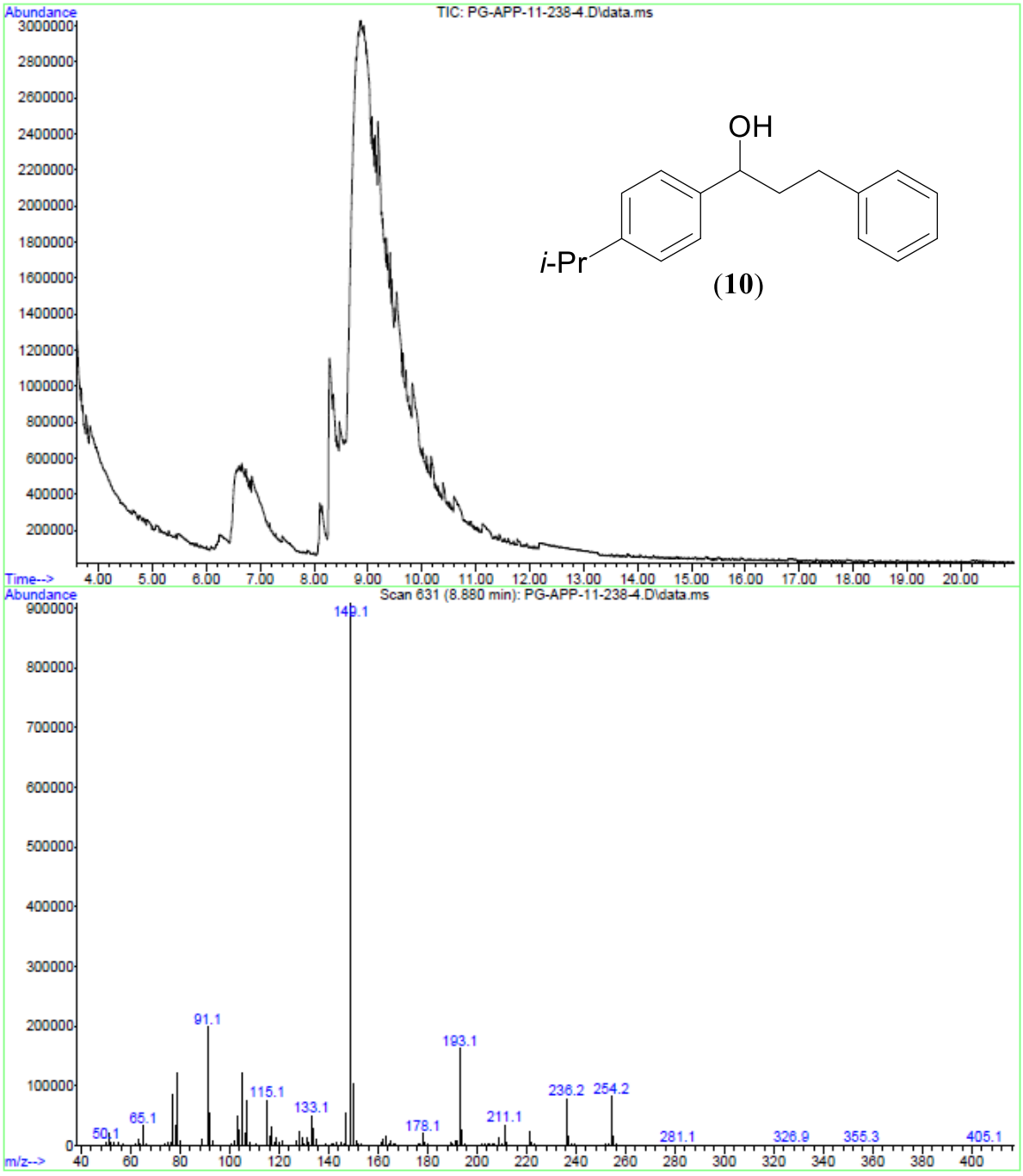
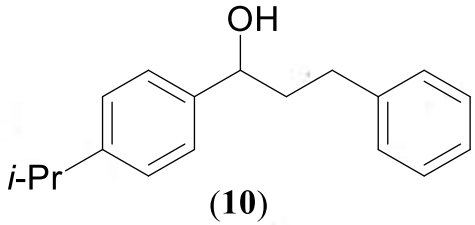


Figure S110. GCMS trace in EtOAc of (**10**) showing the M^+ peak at *m/z* 254.



(**10**)

Eager 300 Report

Page: 1 Sample: PG-APP-11-238-1 (PG-APP-11-238-1)

Method Name : PGAPP240519
Method File : D:\CHNS2019\PGAPP240519.mth
Chromatogram : PG-APP-11-238-1
Operator ID : Prakash
Analysed : 05/24/2019 19:08
Sample ID : PG-APP-11-238-1 (# 36)
Analysis Type : UnkNown (Area)

Company Name : C.E. Instruments
Printed : 5/25/2019 00:22
Instrument N. : Instrument #1
Sample weight : 1.008

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	134675	RS		0.0000
Carbon	85.0741	63	2286149	RS	1.000000	.266592E+07
Hydrogen	8.1537	184	490957	RS	4.656515	.597347E+07
Totals	93.2279		2911781			

Figure S111. Elemental analysis data of (**10**).

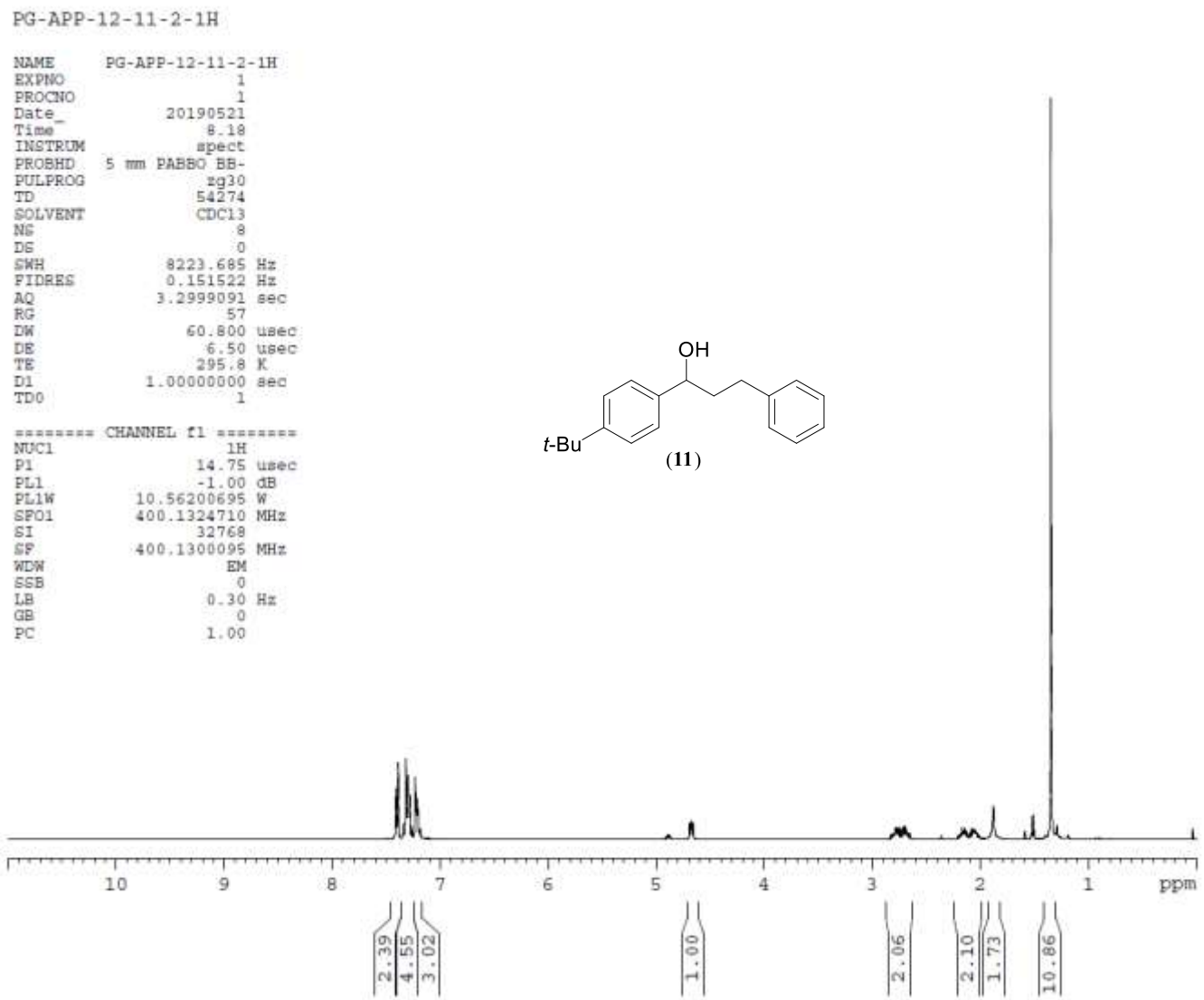


Figure S112. ^{1}H NMR spectrum of (**11**) in $CDCl_3$.

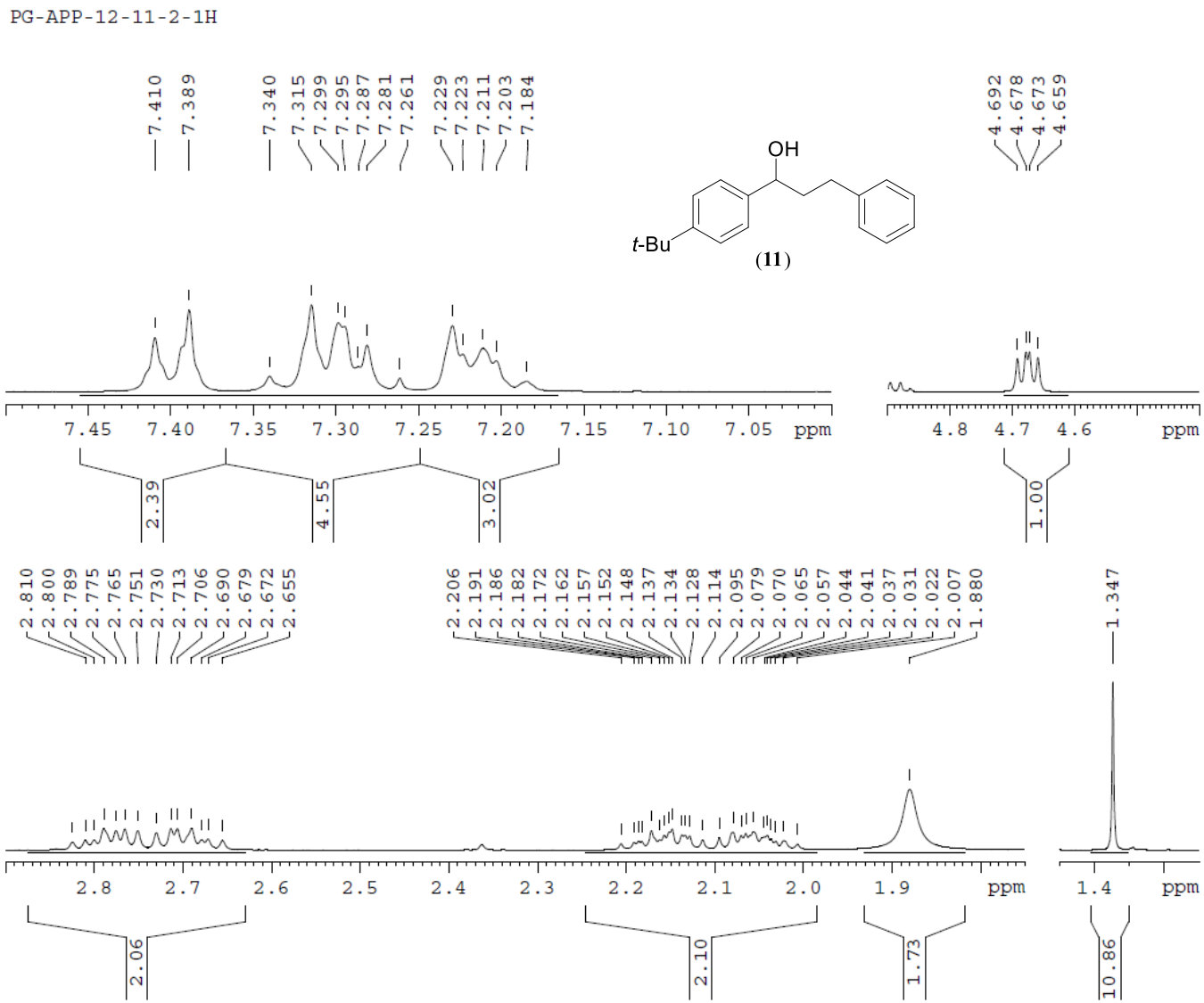


Figure S113. Expanded ^{1}H NMR spectrum of (**11**) in $CDCl_3$.

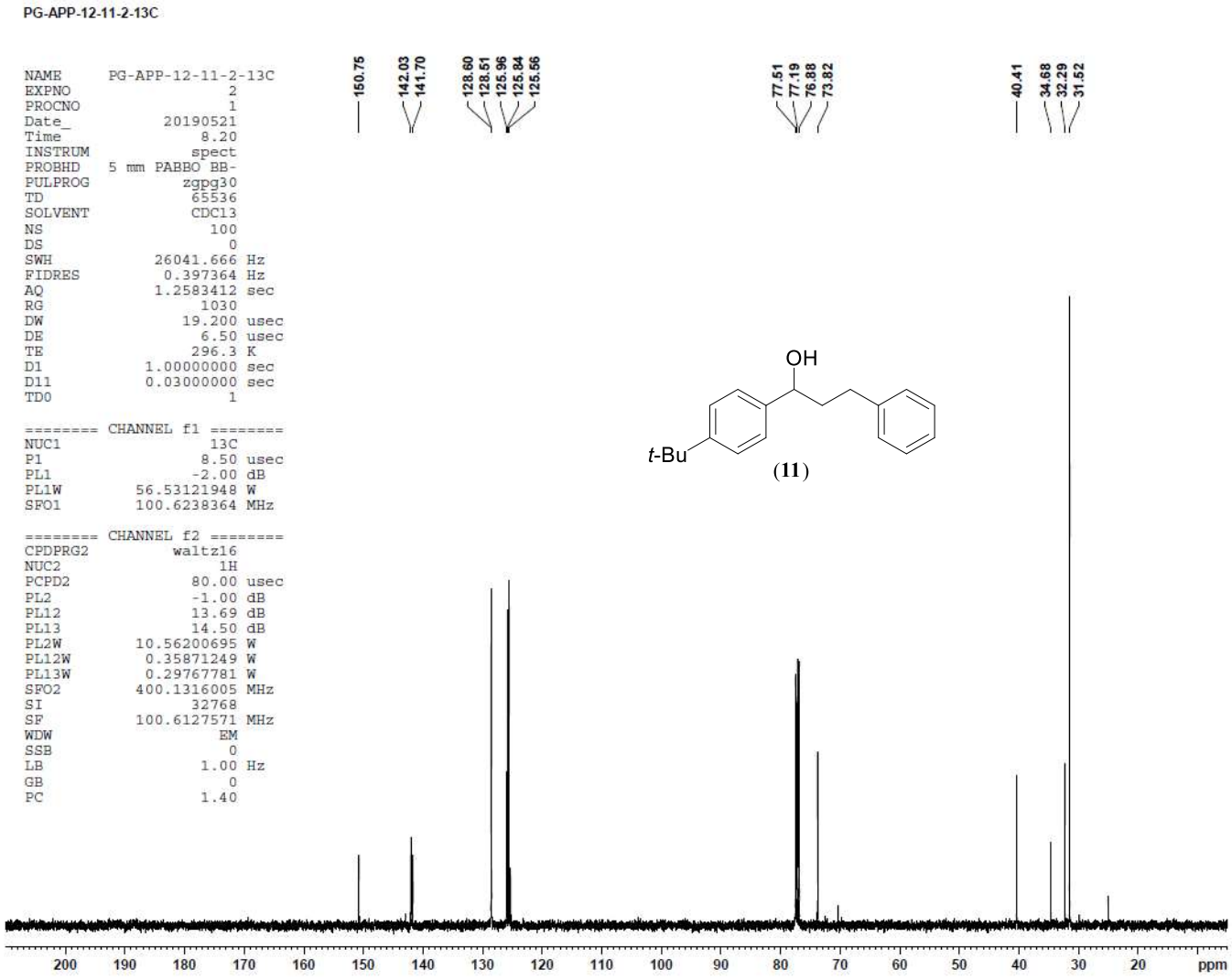


Figure S114. $^{13}C\{^{1}H\}$ NMR spectrum of (**11**) in $CDCl_3$.

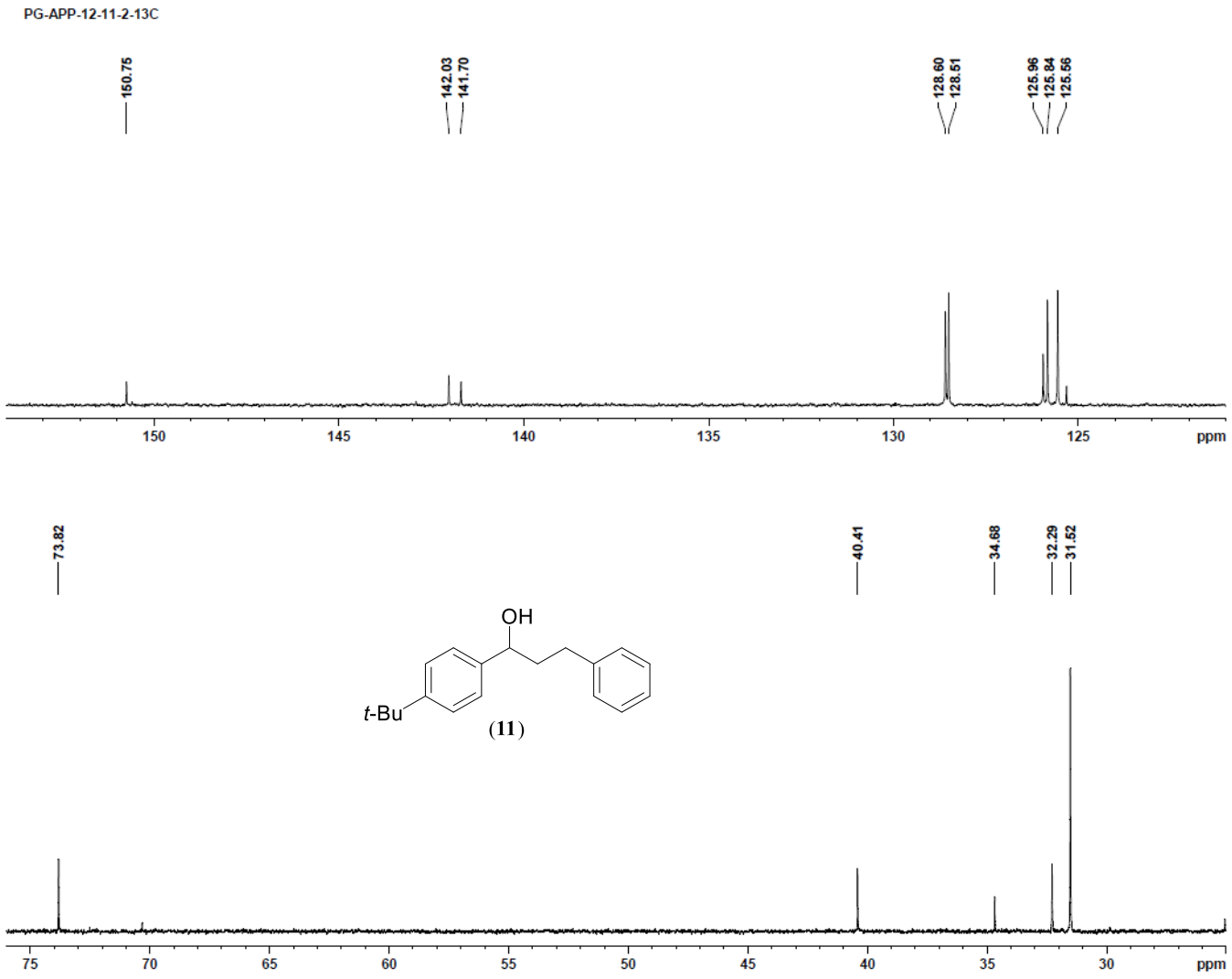


Figure S115. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**11**) in $CDCl_3$.

```
File       :F:\GCMSDATA2019\May 2019\PG-APP-12-11-31.D
Operator   : APP
Acquired   : 20 May 2019  22:57     using AcqMethod COMMONMETHOD-2018.M
Instrument :    GCMS
Sample Name: PG-APP-12-11-31
Misc Info  :
Vial Number: 1
```

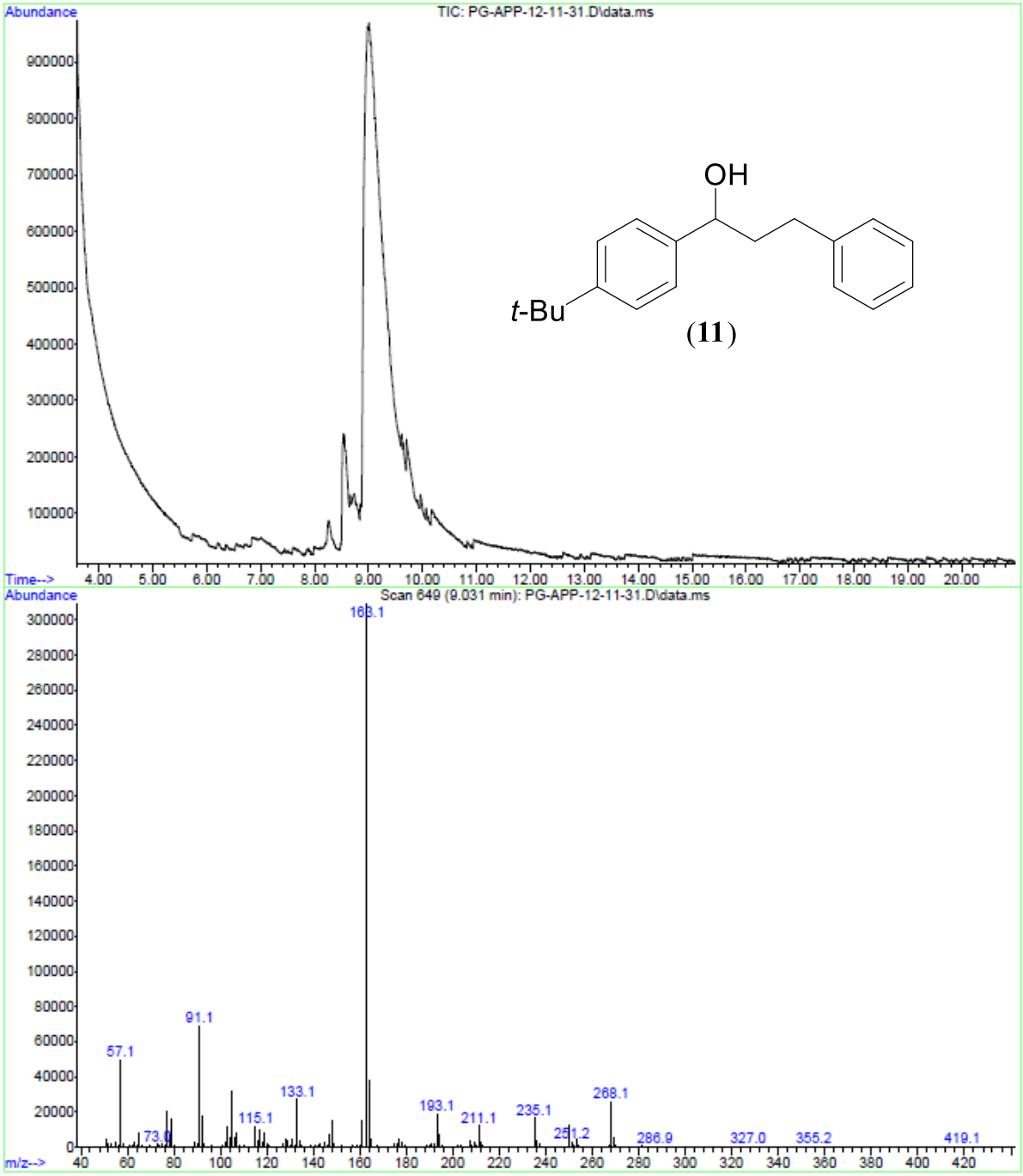
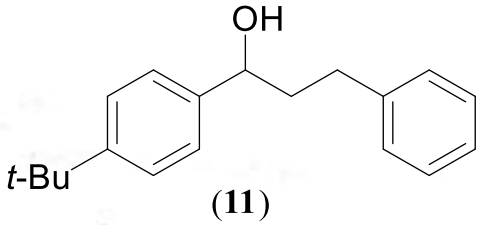


Figure S116. GCMS trace in EtOAc of (**11**) showing the M^+ peak at *m/z* 268.



(**11**)

Eager 300 Report

Page: 1 Sample: PG-APP-12-11-5 (PG-APP-12-11-5)

Method Name : PGAPP240519
Method File : D:\CHNS2019\PGAPP240519.mth
Chromatogram : PG-APP-12-11-5
Operator ID : Prakash
Analysed : 05/24/2019 20:37
Sample ID : PG-APP-12-11-5 (# 44)
Analysis Type : UnkNown (Area)

Company Name : C.E. Instruments
Printed : 5/25/2019 00:22
Instrument N. : Instrument #1
Sample weight : .759

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
						0.0000
1	0.0000	6	60133	RS		
Carbon	84.3312	63	1706383	RS	1.000000	.266592E+07
Hydrogen	8.2963	184	376143	RS	4.536527	.597347E+07
Totals	92.6275		2142659			

Figure S117. Elemental analysis data of (**11**).

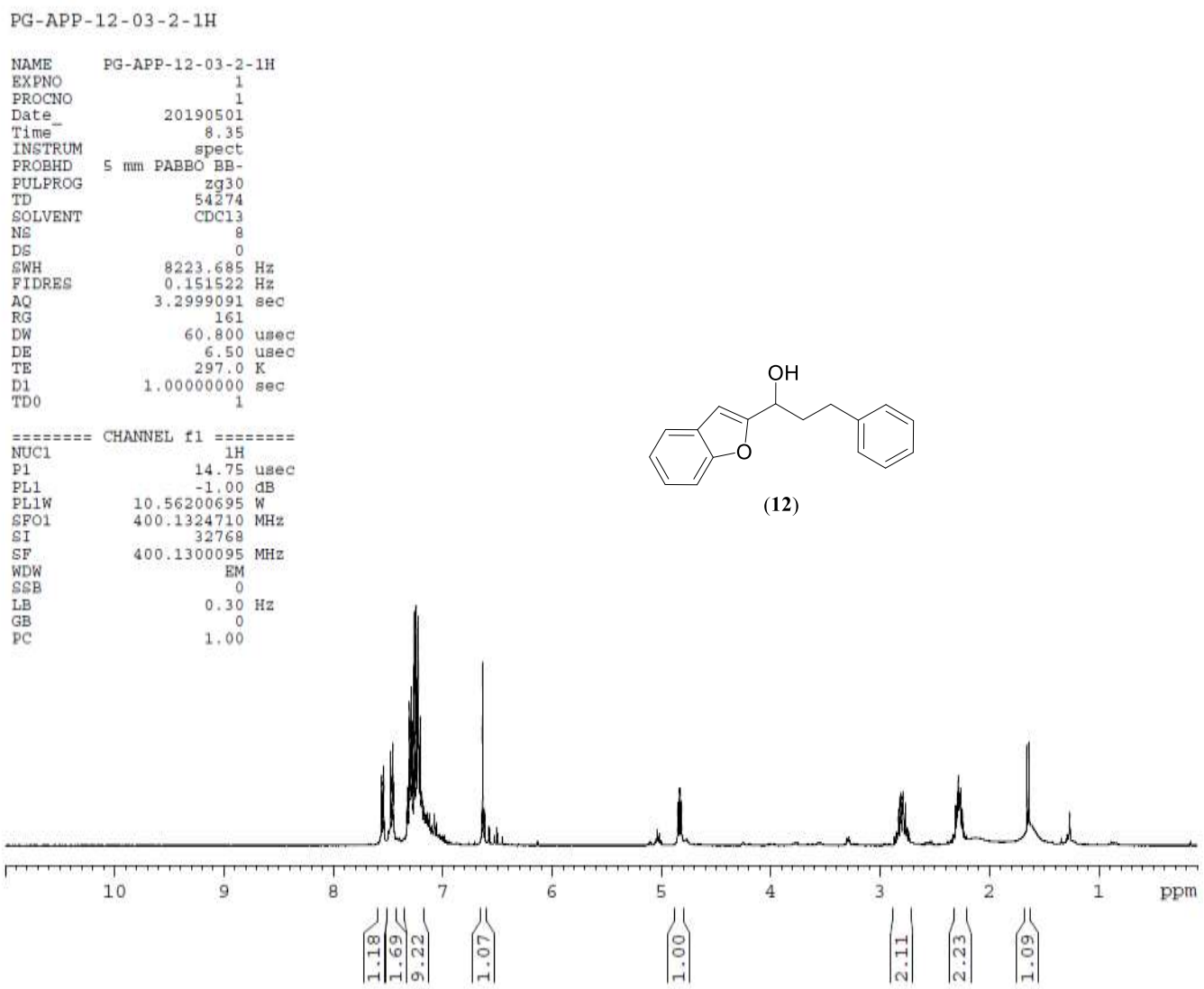


Figure S118. ^{1}H NMR spectrum of (**12**) in $CDCl_3$.

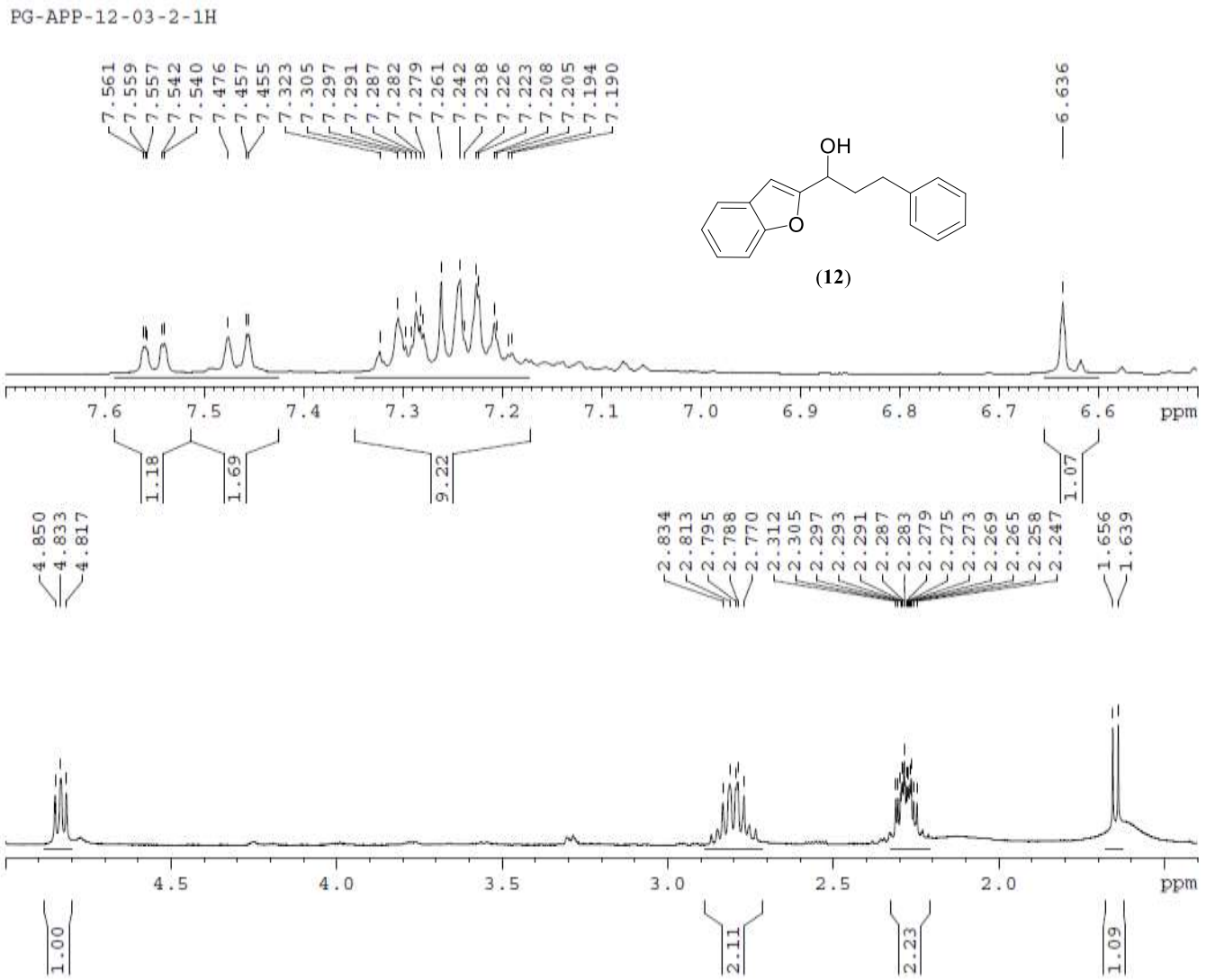


Figure S119. Expanded ^{1}H NMR spectrum of (**12**) in $CDCl_3$.

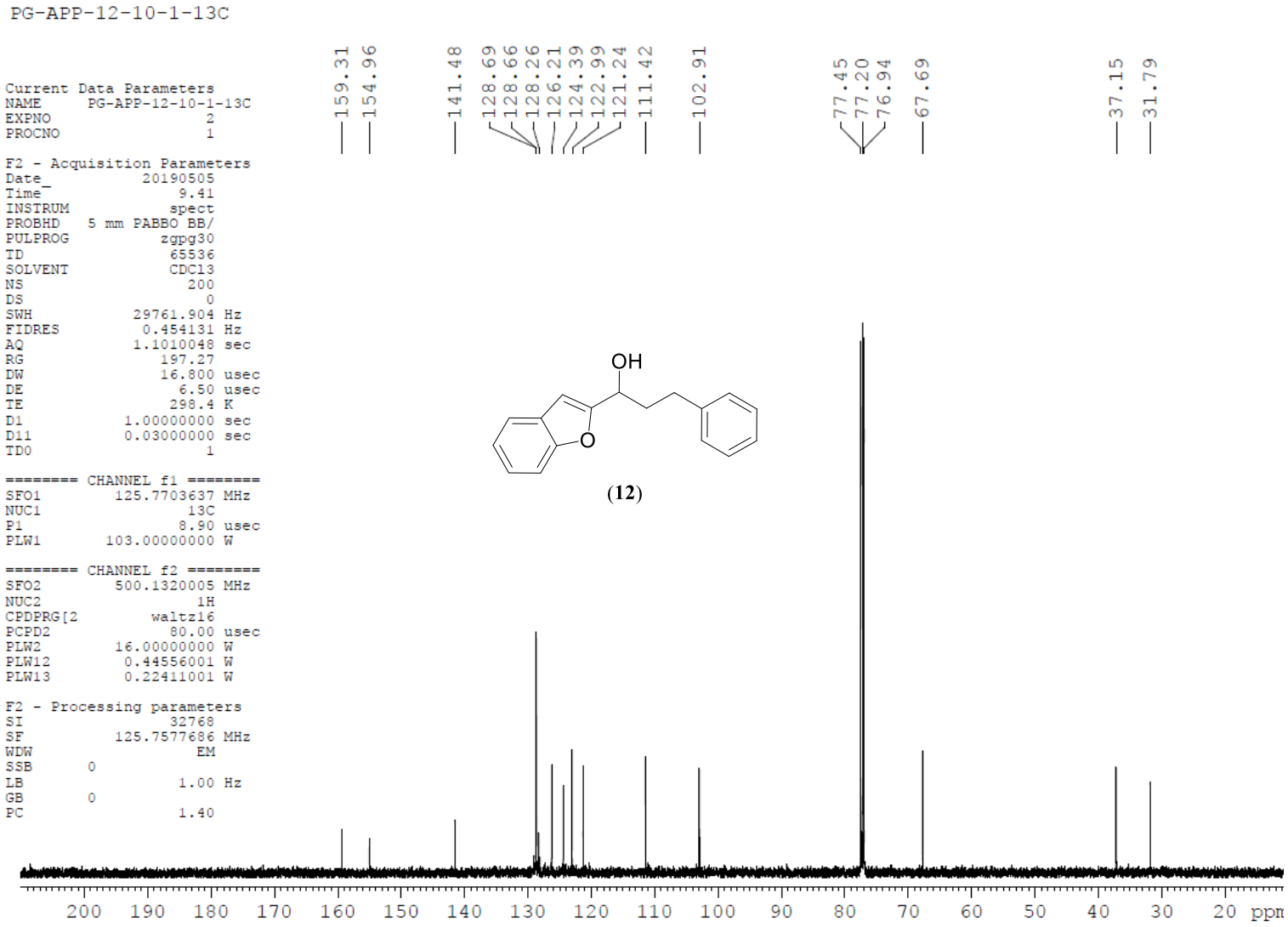


Figure S120. $^{13}C\{^{1}H\}$ NMR spectrum of (**12**) in $CDCl_3$.

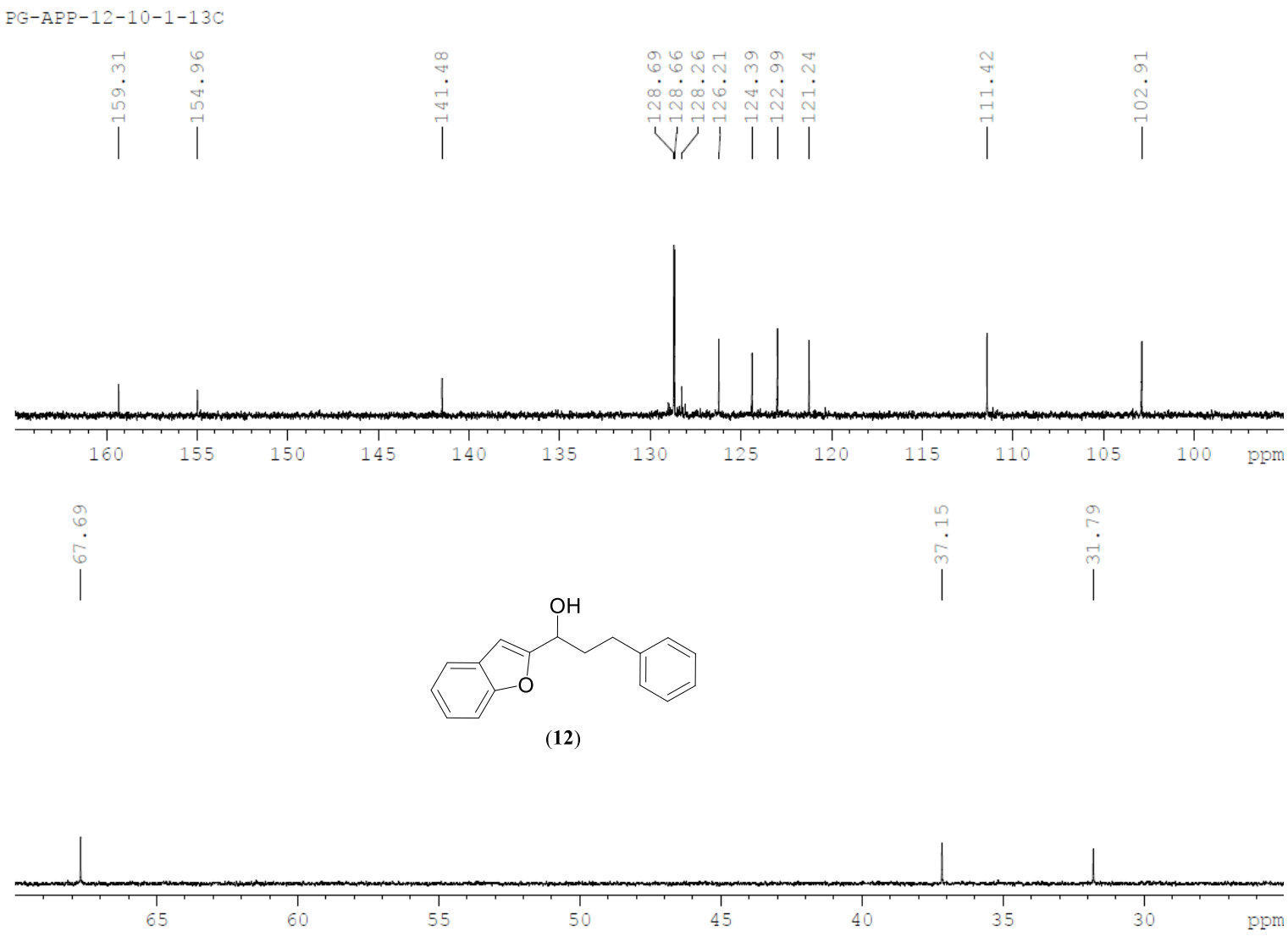


Figure S121. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**12**) in $CDCl_3$.

```
File       :F:\GCMSDATA2019\May 2019\PG-APP-12-10-2.D
Operator   : APP
Acquired   :  6 May 2019  11:26     using AcqMethod COMMONMETHOD_2018.M
Instrument :    GCMS
Sample Name: PG-APP-12-10-2
Misc Info  :
Vial Number: 3
```

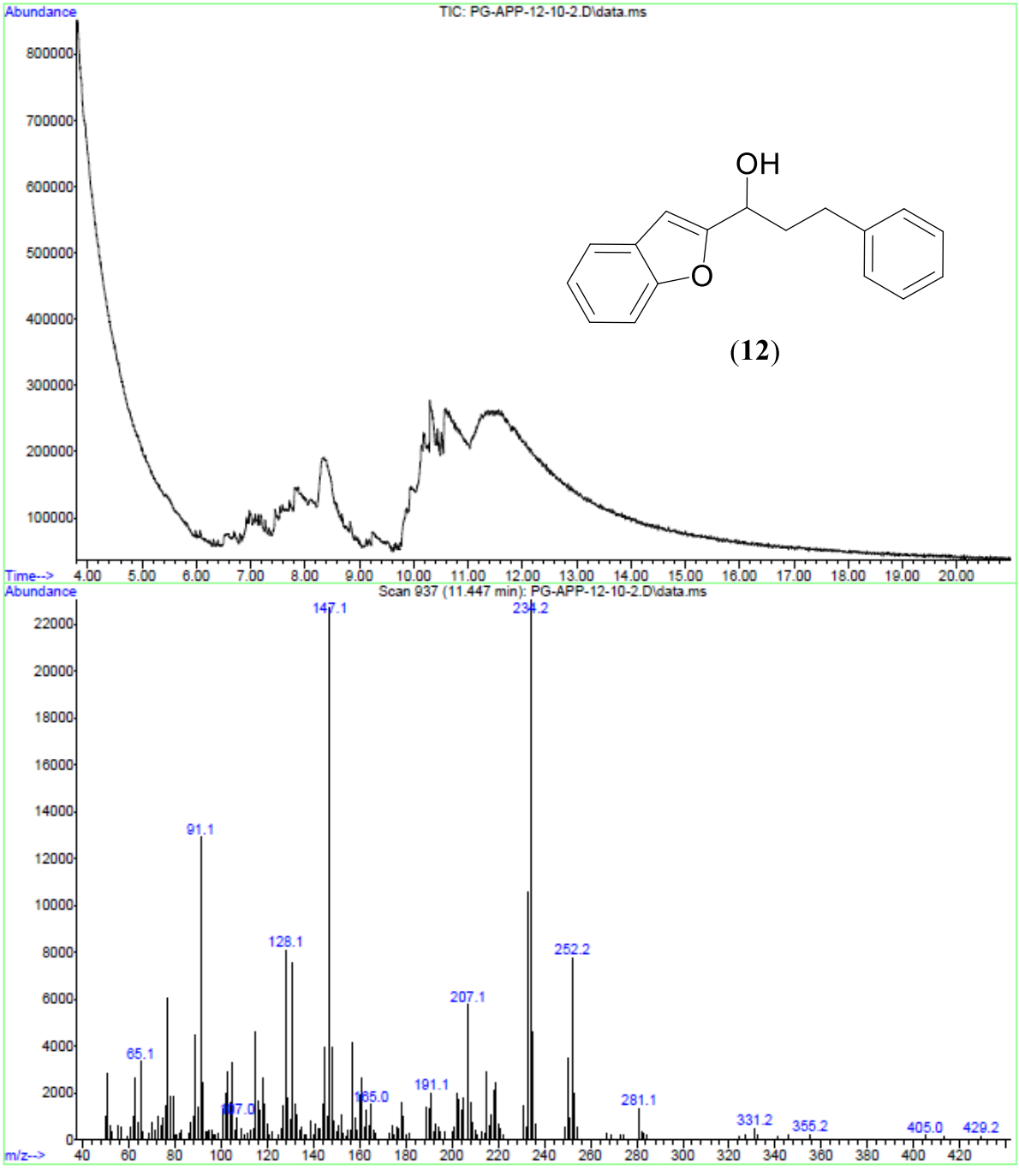
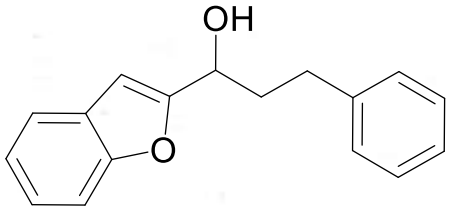


Figure S122. GCMS trace in EtOAc of (**12**) showing the M^+ peak at *m/z* 252.



(**12**)

Eager 300 Report

Page: 1 Sample: PG-APP-12-03-1 (PG-APP-12-03-1)

Method Name : PGAPP300519
Method File : D:\CHNS2019\PGAPP300519.mth
Chromatogram : PG-APP-12-03-1
Operator ID : Prakash
Analysed : 05/30/2019 17:22
Sample ID : PG-APP-12-03-1 (# 34)
Analysis Type : UnkNown (Area)

Company Name : C.E. Instruments
Printed : 5/30/2019 23:00
Instrument N. : Instrument #1
Sample weight : 1.108

Calib. method : using 'K Factors'

!!! Warning missing one or more peaks.

Element Name	%	Ret.Time	Area	BC	Area ratio	K factor
1	0.0000	2	30781	FU		0.0000
2	0.0000	6	114524	FU		0.0000
Carbon	81.1336	63	2401468	RS	1.000000	.267139E+07
Hydrogen	5.8671	184	444371	RS	5.404196	.683575E+07
Totals	87.0006		2991144			

Figure S123. Elemental analysis data of (**12**).

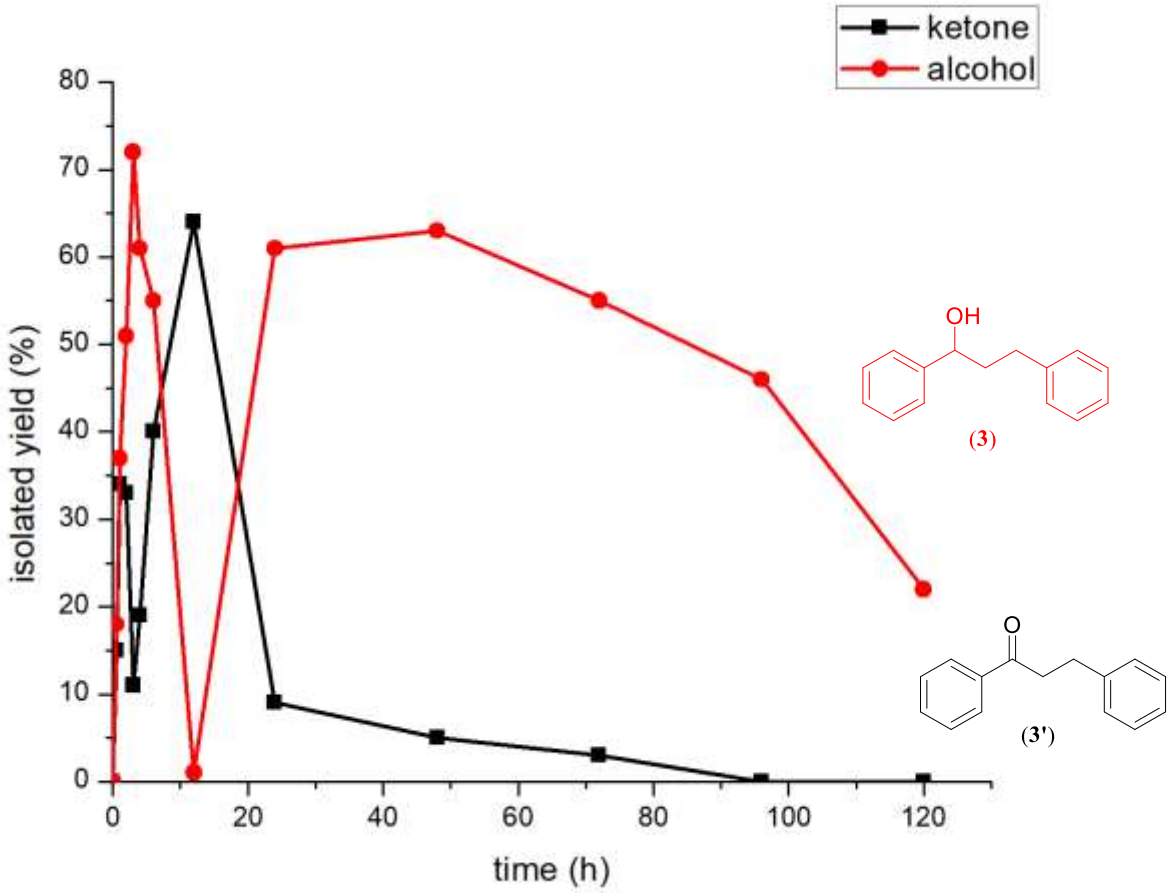


Figure S124. An overlay of the formation of (**3**) and (**3'**) as a function of time in the reaction of 1-phenylethanol and benzyl alcohol as catalyzed by the Ru−NHC complex (**1c**).

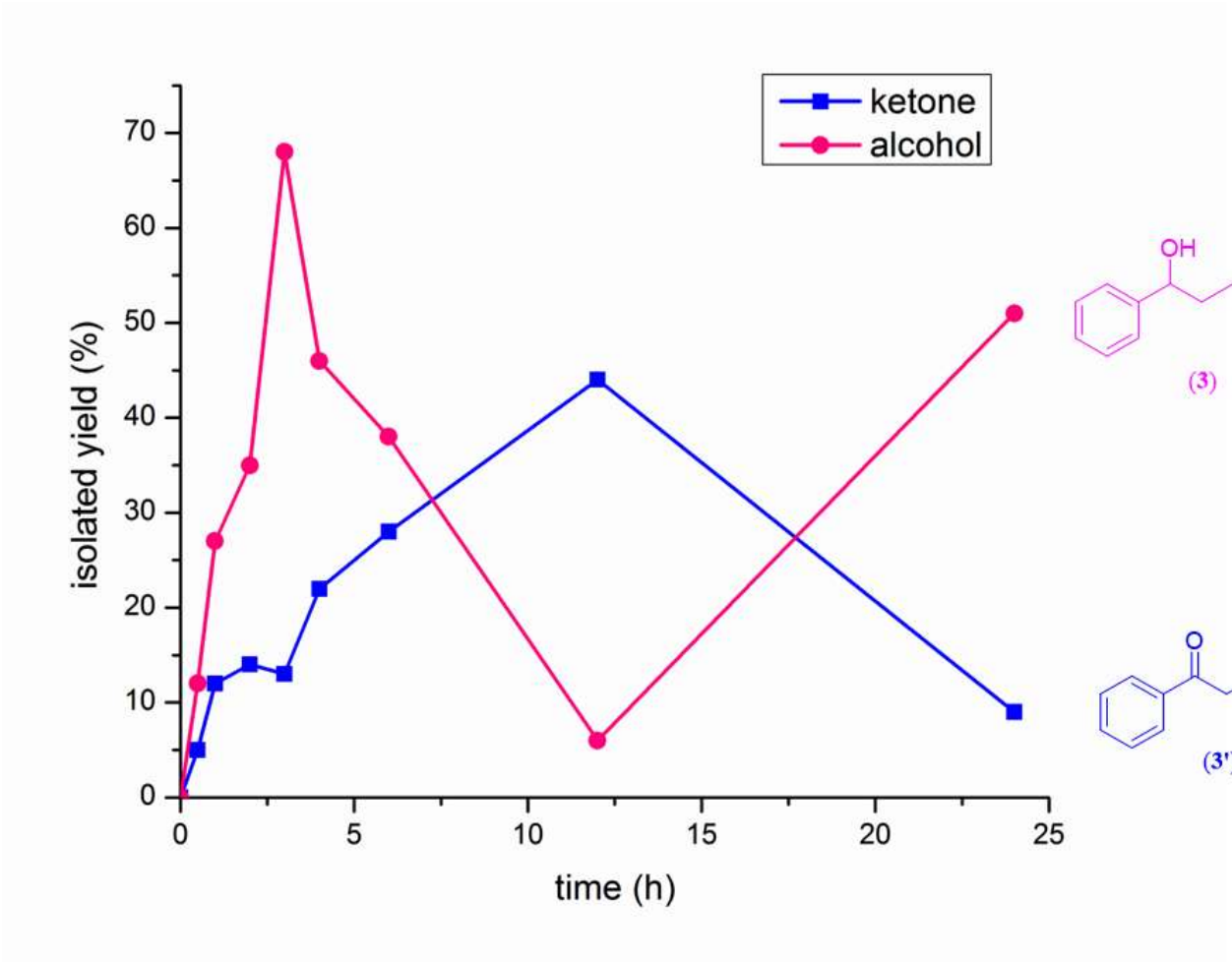


Figure S125. An overlay of the formation of (**3**) and (**3'**) as a function of time in the reaction of 1-phenylethanol and benzyl alcohol as catalyzed by the Ru−NHC complex (**2c**).

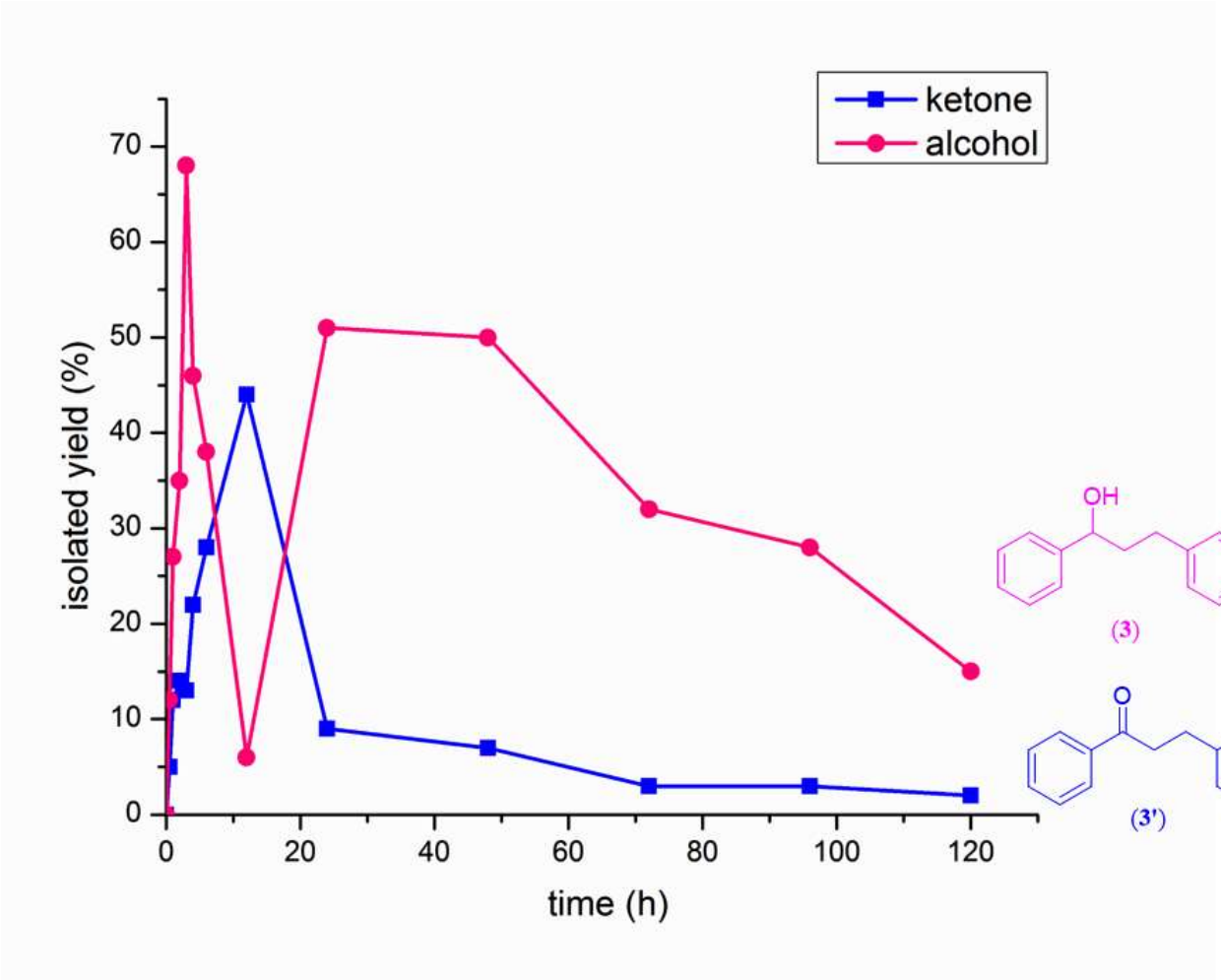


Figure S126. An overlay of the formation of (**3**) and (**3'**) as a function of time in the reaction of 1-phenylethanol and benzyl alcohol as catalyzed by the Ru−NHC complex (**2c**).

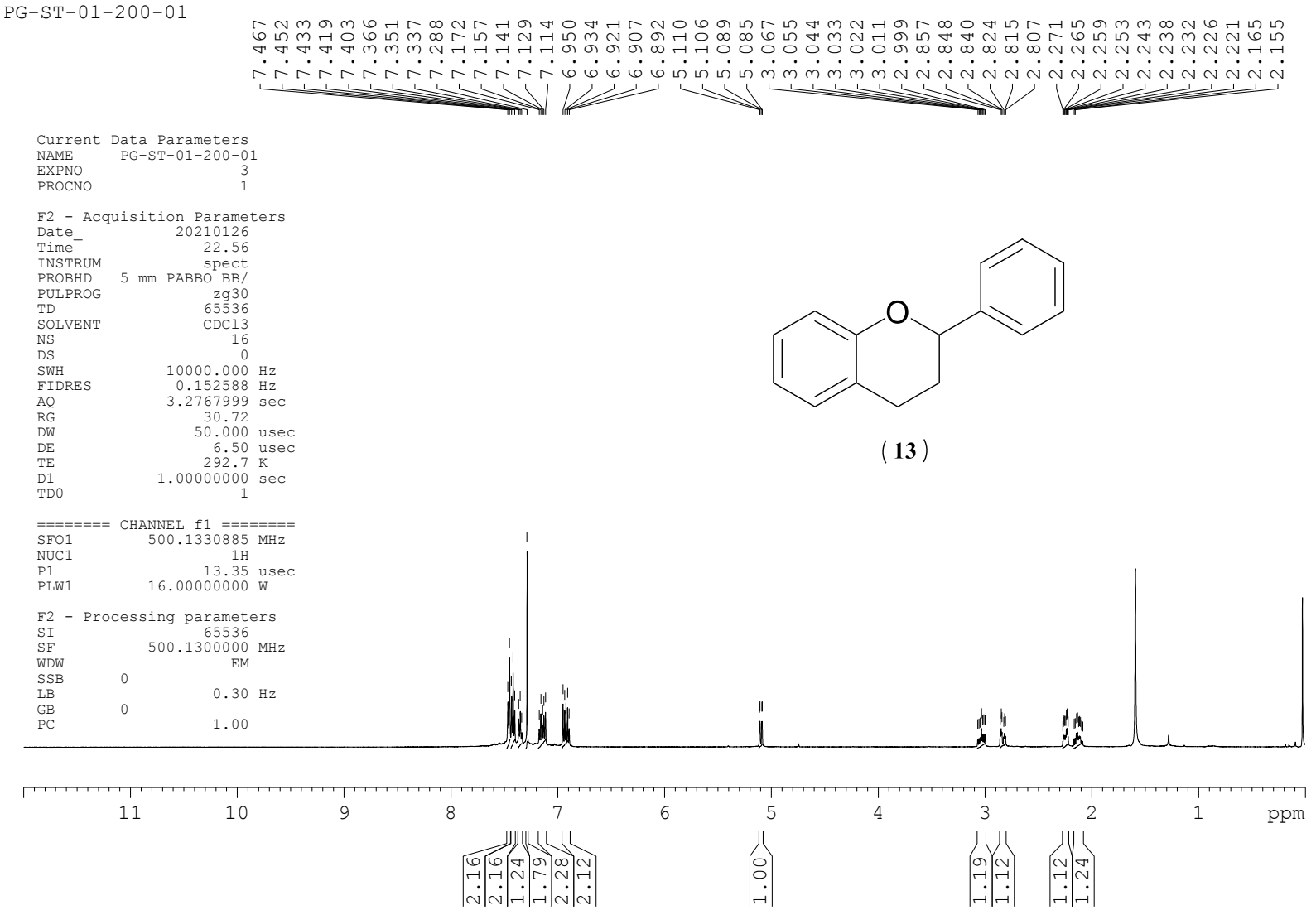


Figure S127. ^{1}H NMR spectrum of (**13**) in $CDCl_3$.

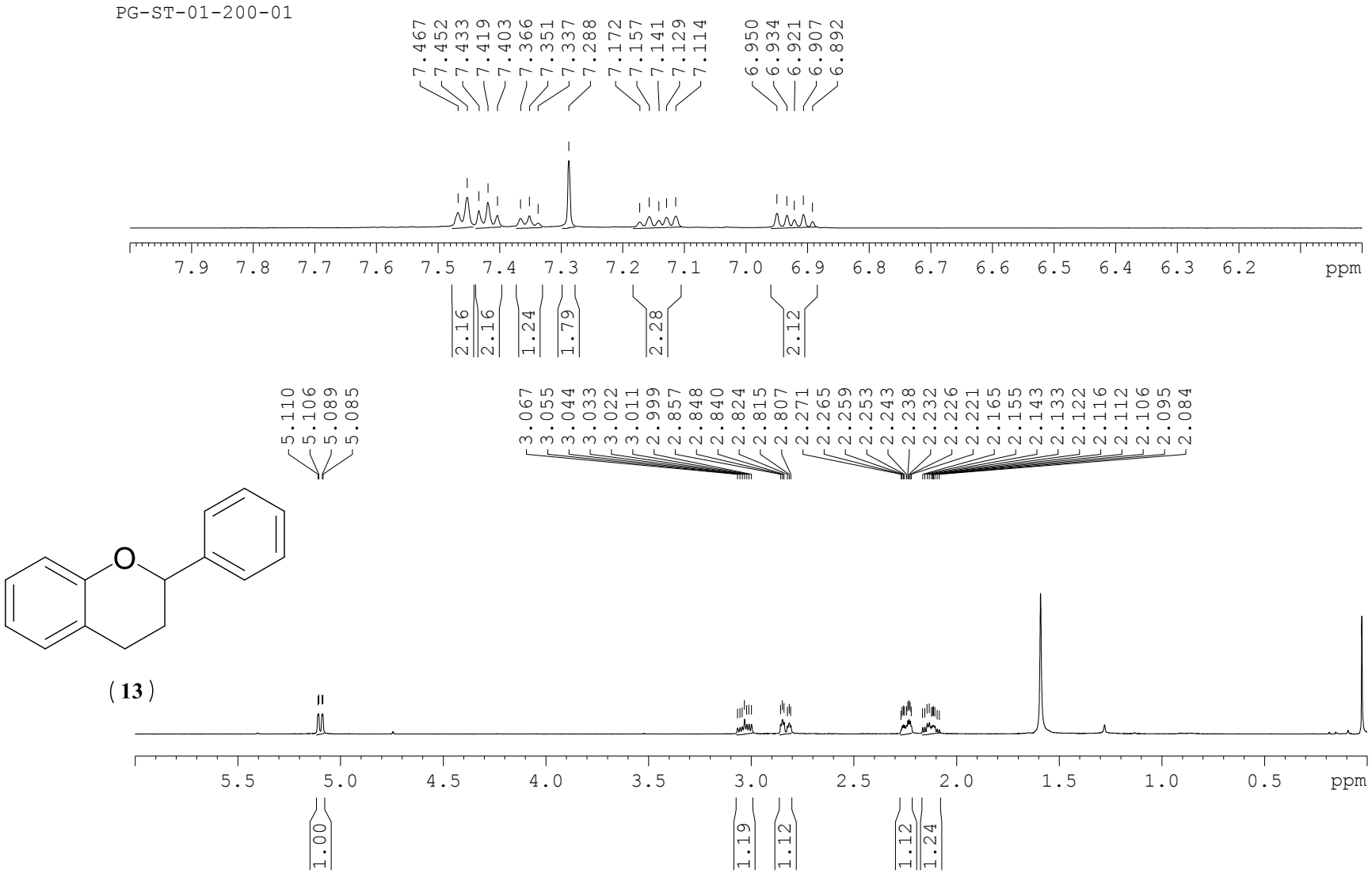
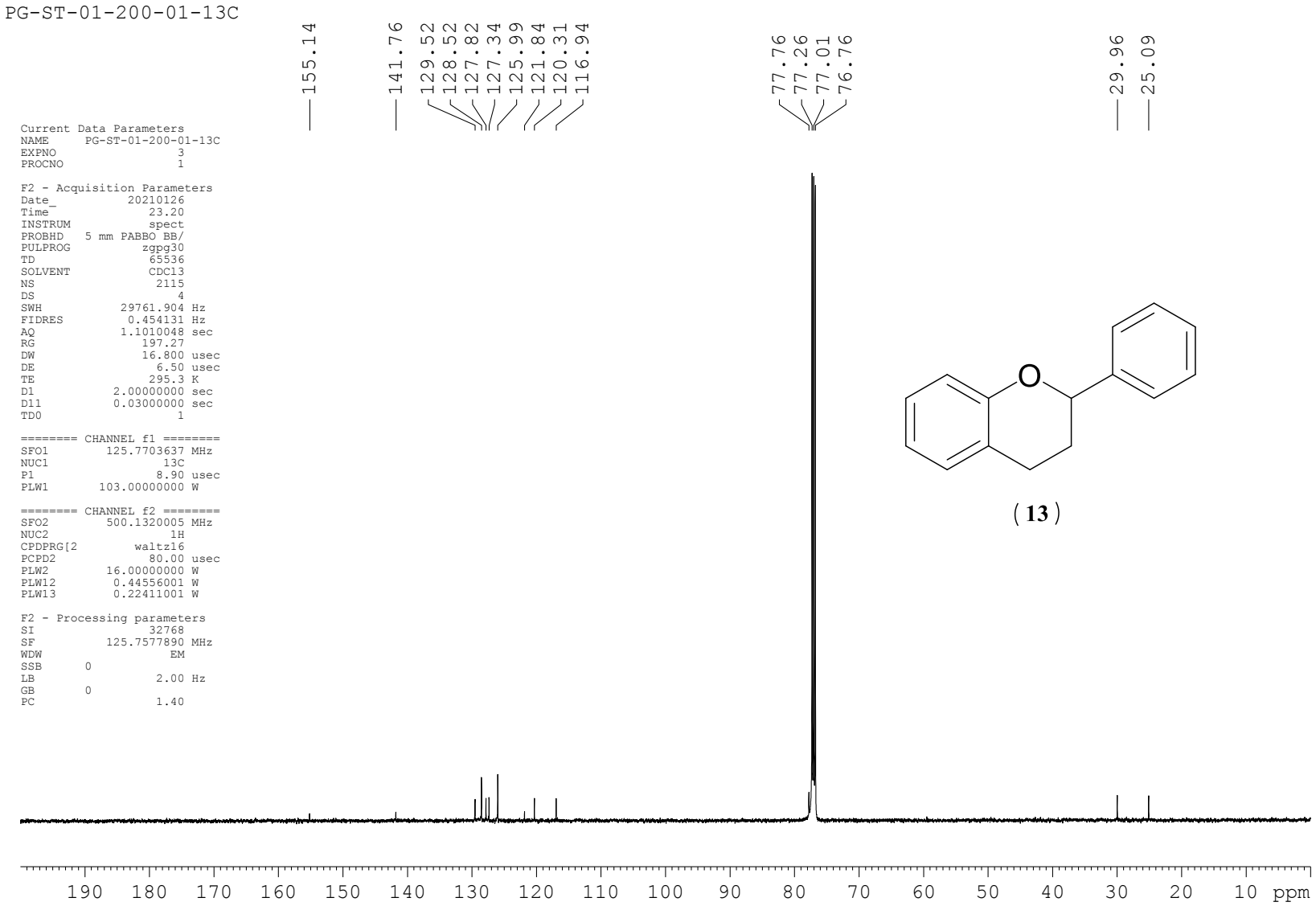
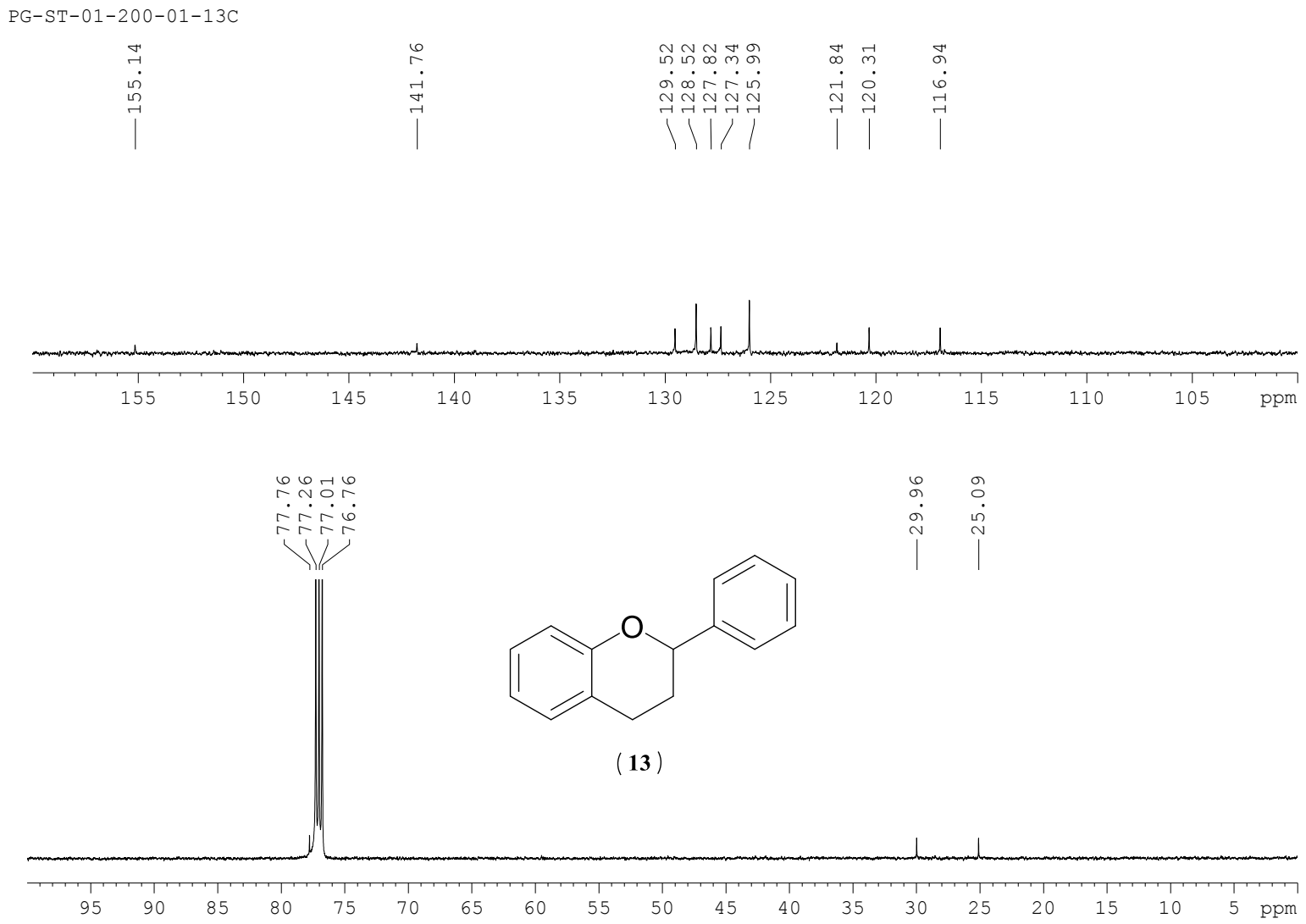


Figure S128. Expanded ^{1}H NMR spectrum of (**13**) in $CDCl_3$.



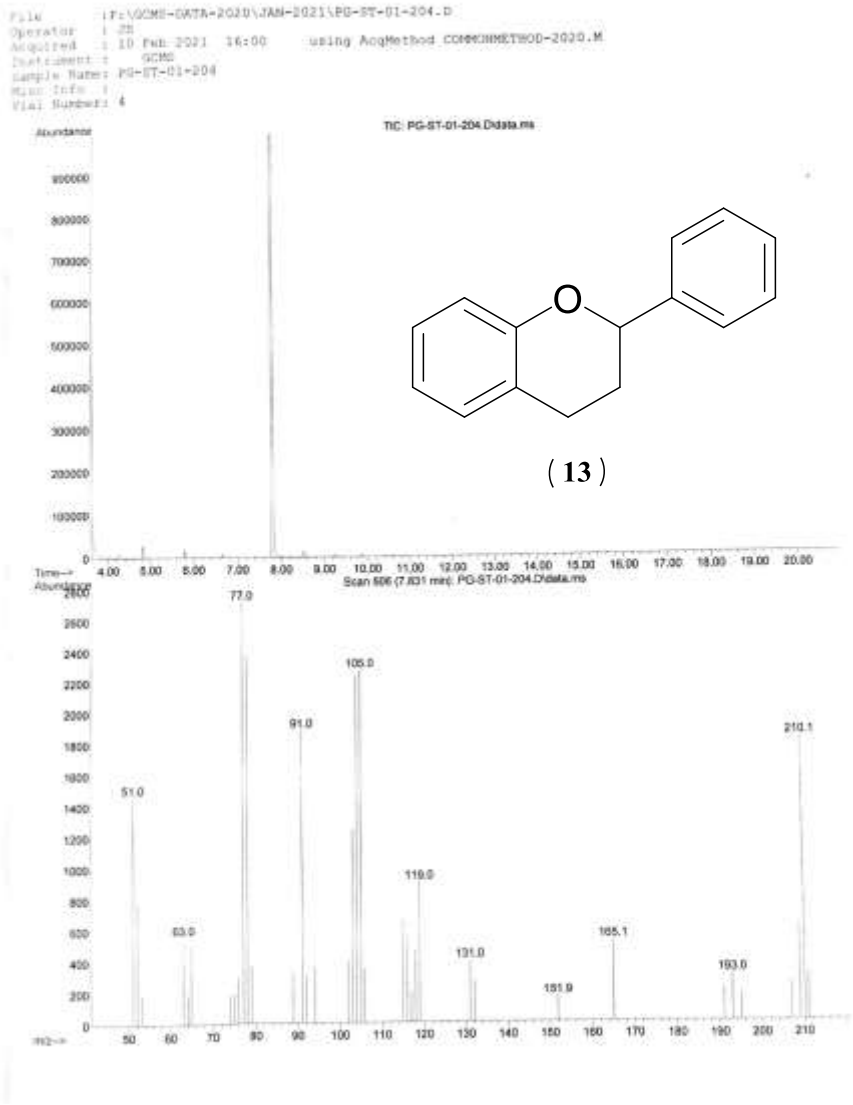
(**13**)

Figure S129. $^{13}C\{^{1}H\}$ NMR spectrum of (**13**) in $CDCl_3$.



(**13**)

Figure S130. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**13**) in $CDCl_3$.



(**13**)

Figure S131. GCMS trace in EtOAc of (**13**) showing the M^+ peak at *m/z* 210.

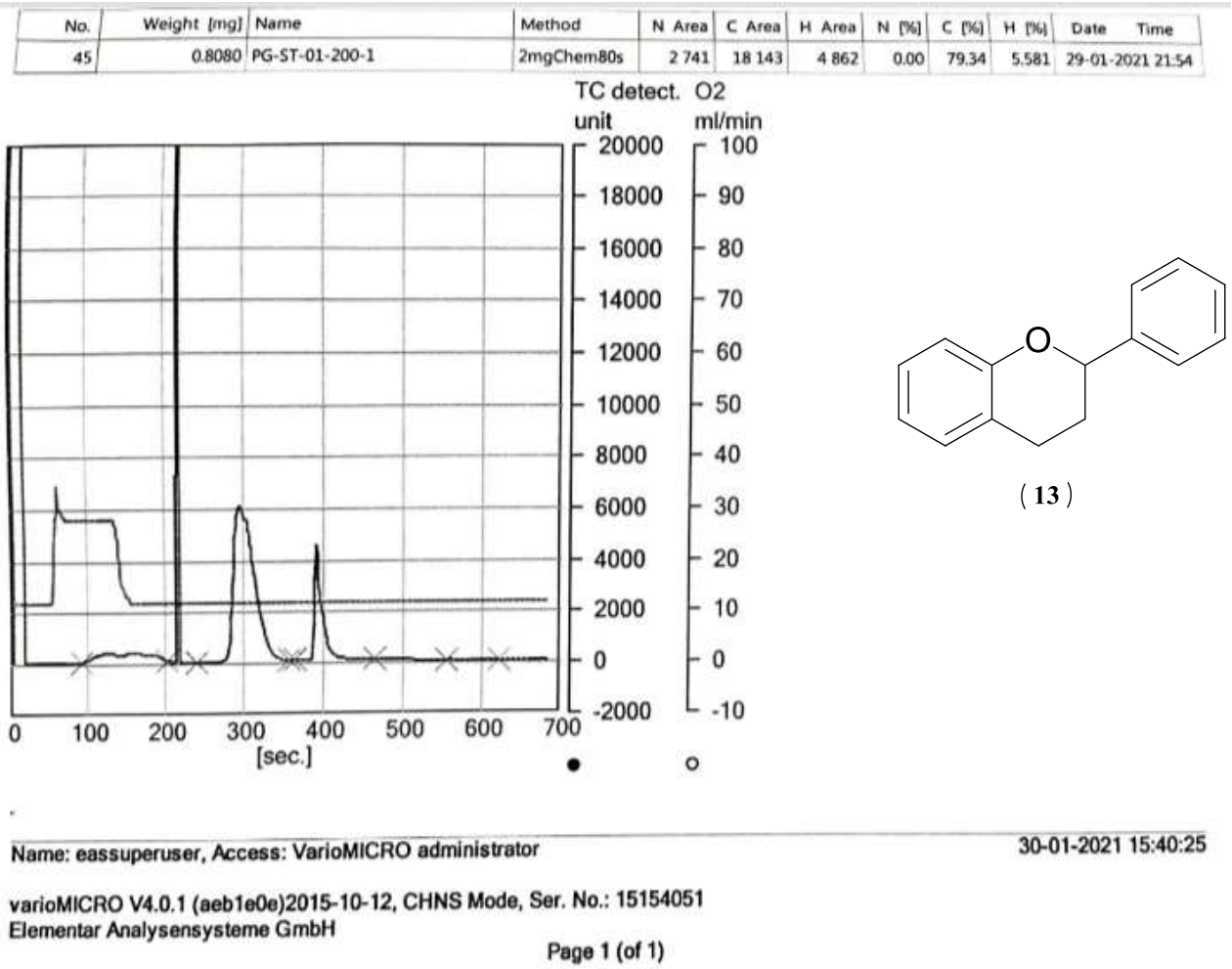


Figure S132. Elemental analysis data of (**13**).

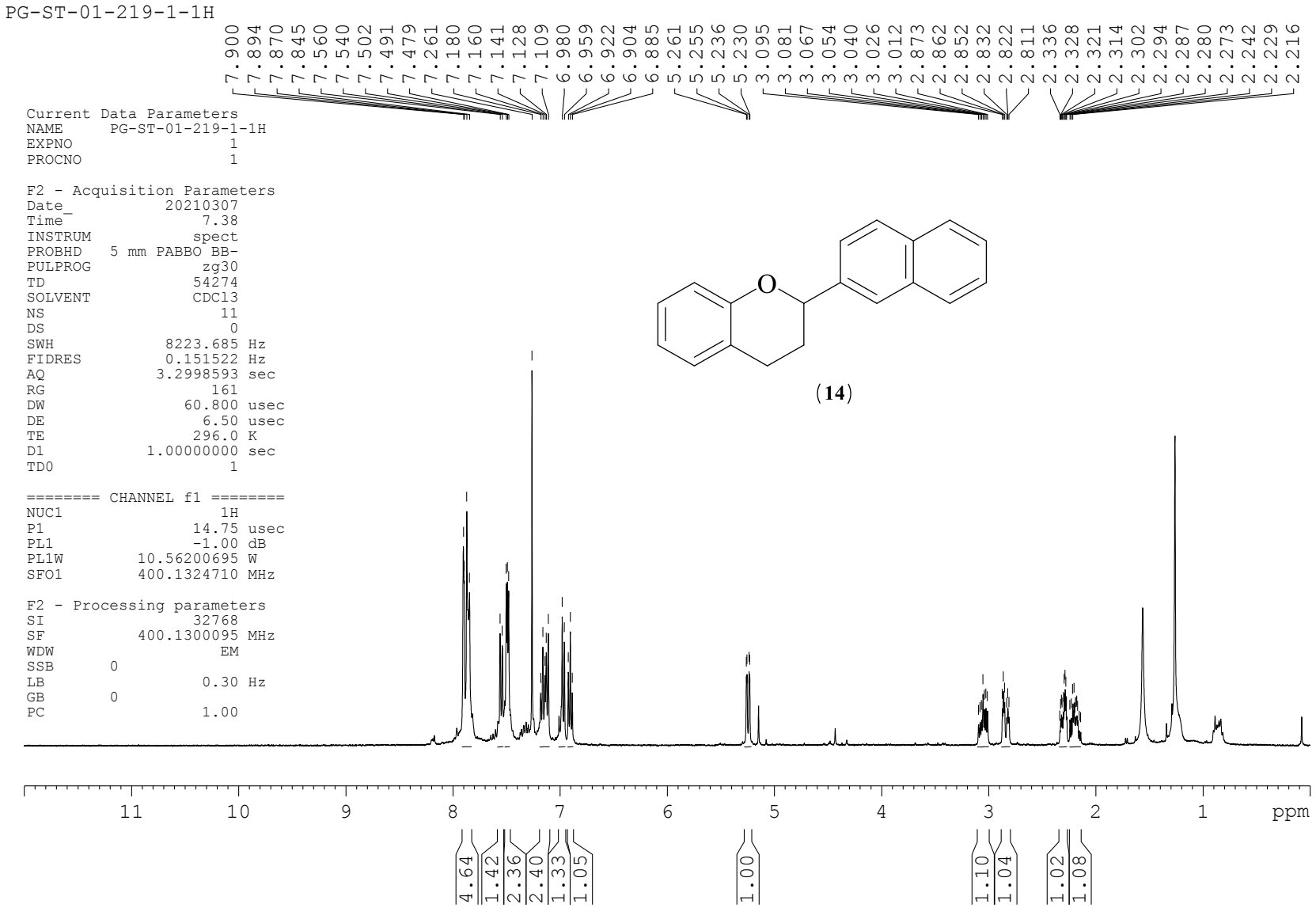


Figure S133. ^{1}H NMR spectrum of (**14**) in $CDCl_3$.

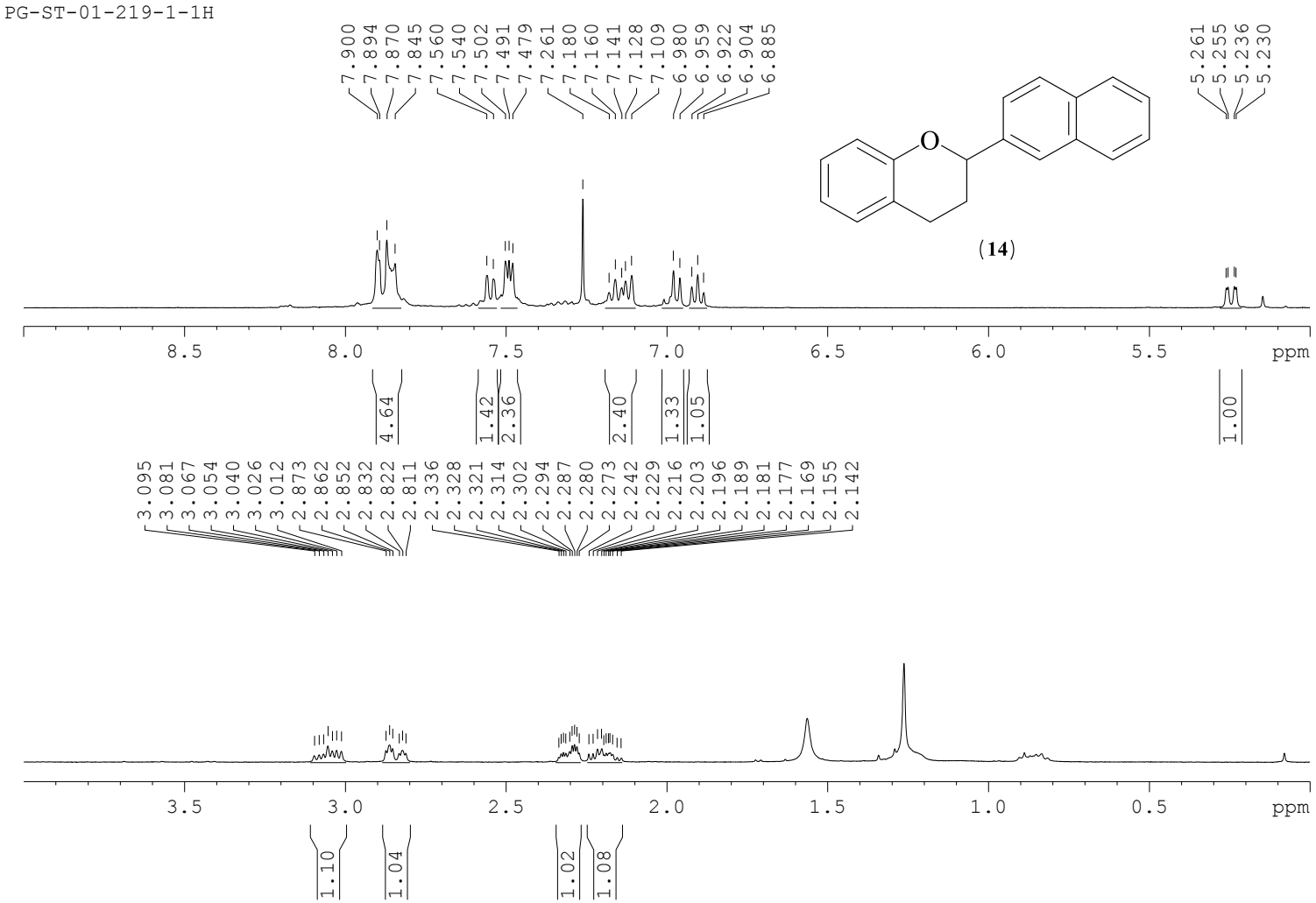


Figure S134. Expanded ^{1}H NMR spectrum of (**14**) in $CDCl_3$.

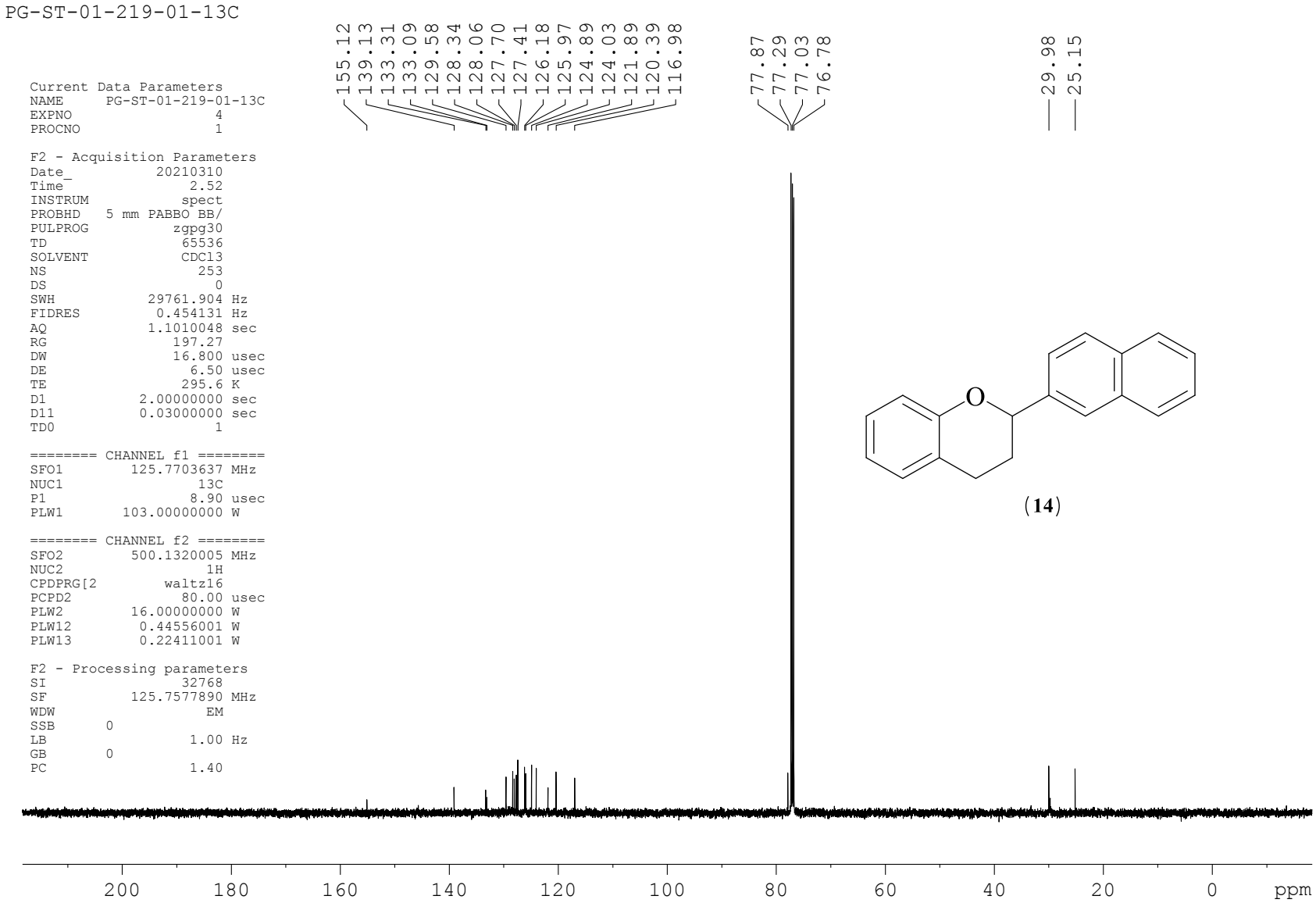


Figure S135. $^{13}C\{^{1}H\}$ NMR spectrum of (**14**) in $CDCl_3$.

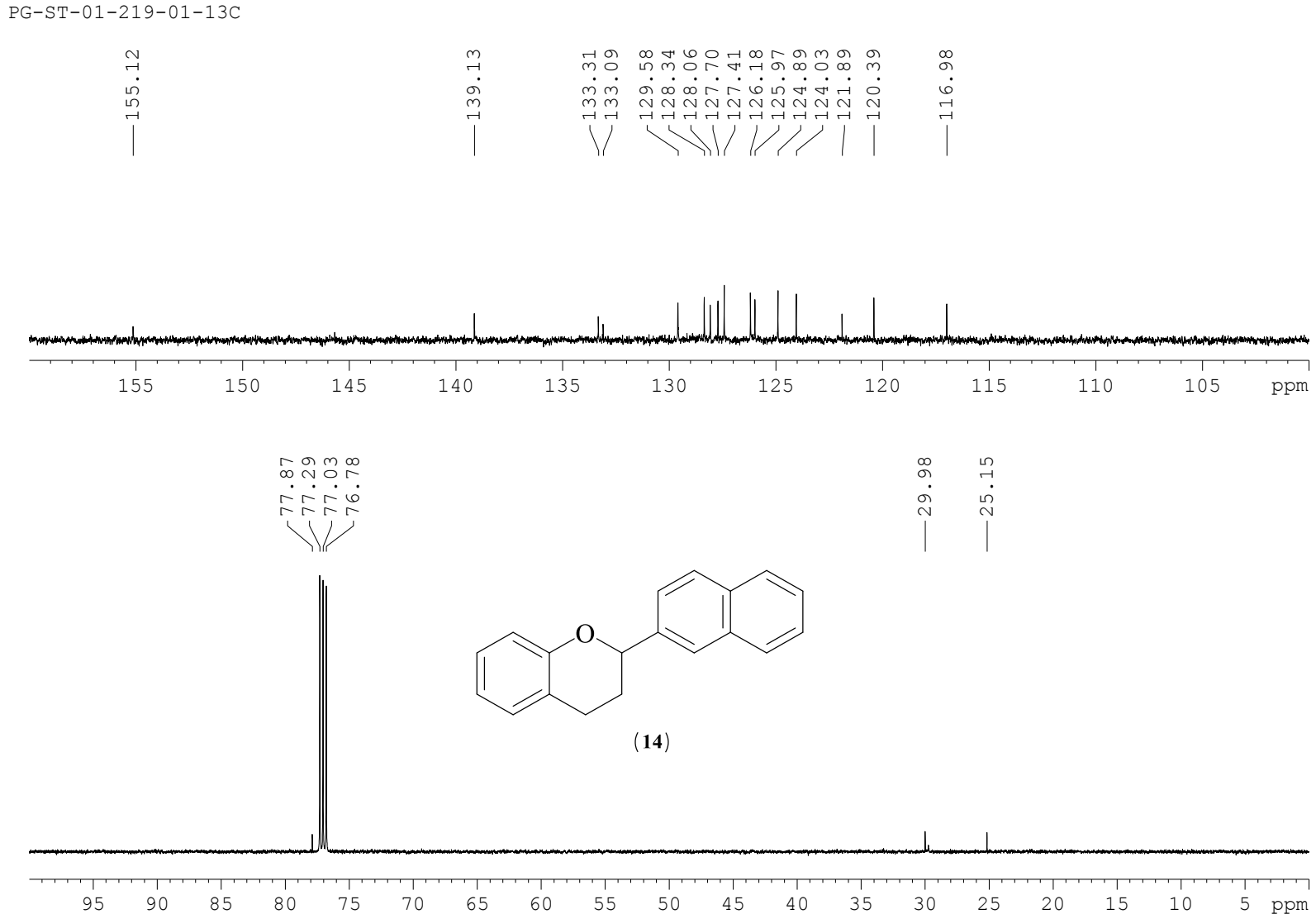


Figure S136. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**14**) in $CDCl_3$.

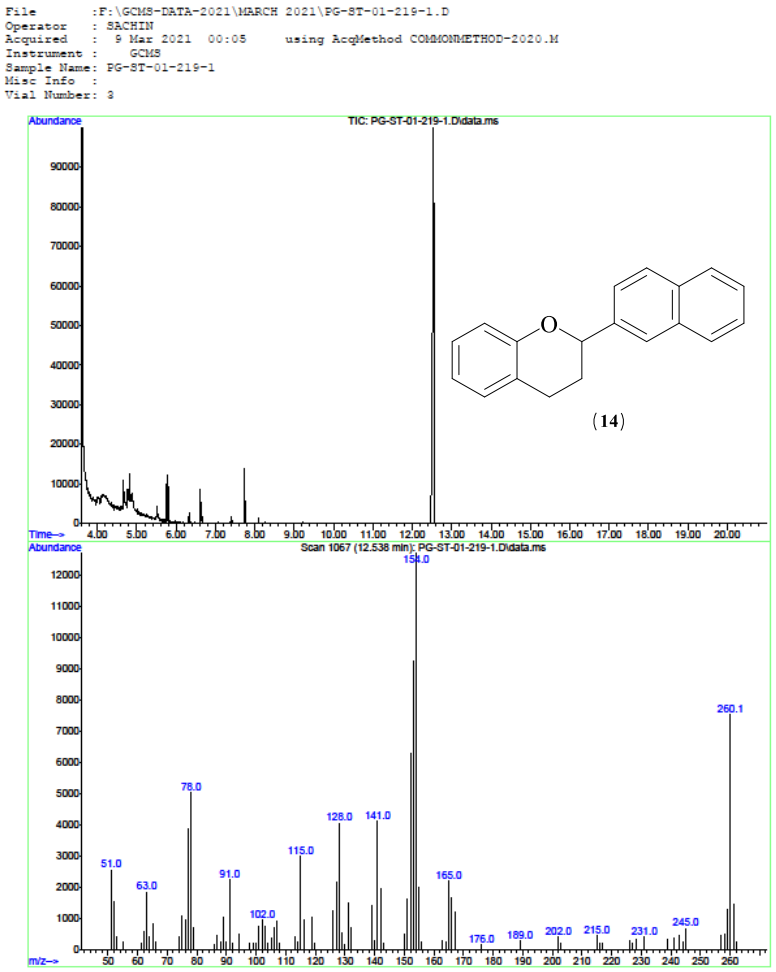


Figure S137. GCMS trace in EtOAc of (**14**) showing the M^+ peak at *m/z* 260.

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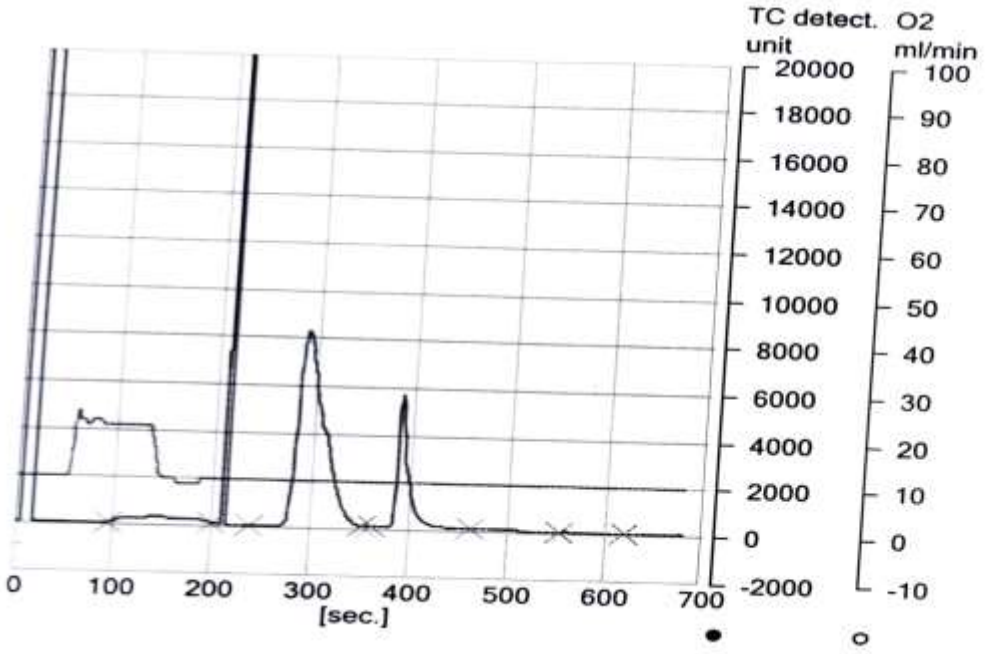
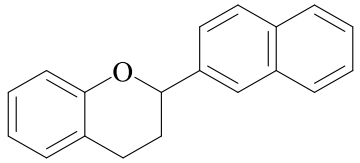
serial number: 15154051

Graphic report

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	N [%]	C [%]	H [%]	Date Time
23	1.0800	PG-ST-01-222-02-1	2mgChem80s	1 971	25 238	6 949	0.00	81.68	5.662	06-04-2021 17:45





(**14**)

Figure S138. Elemental analysis data (**14**).

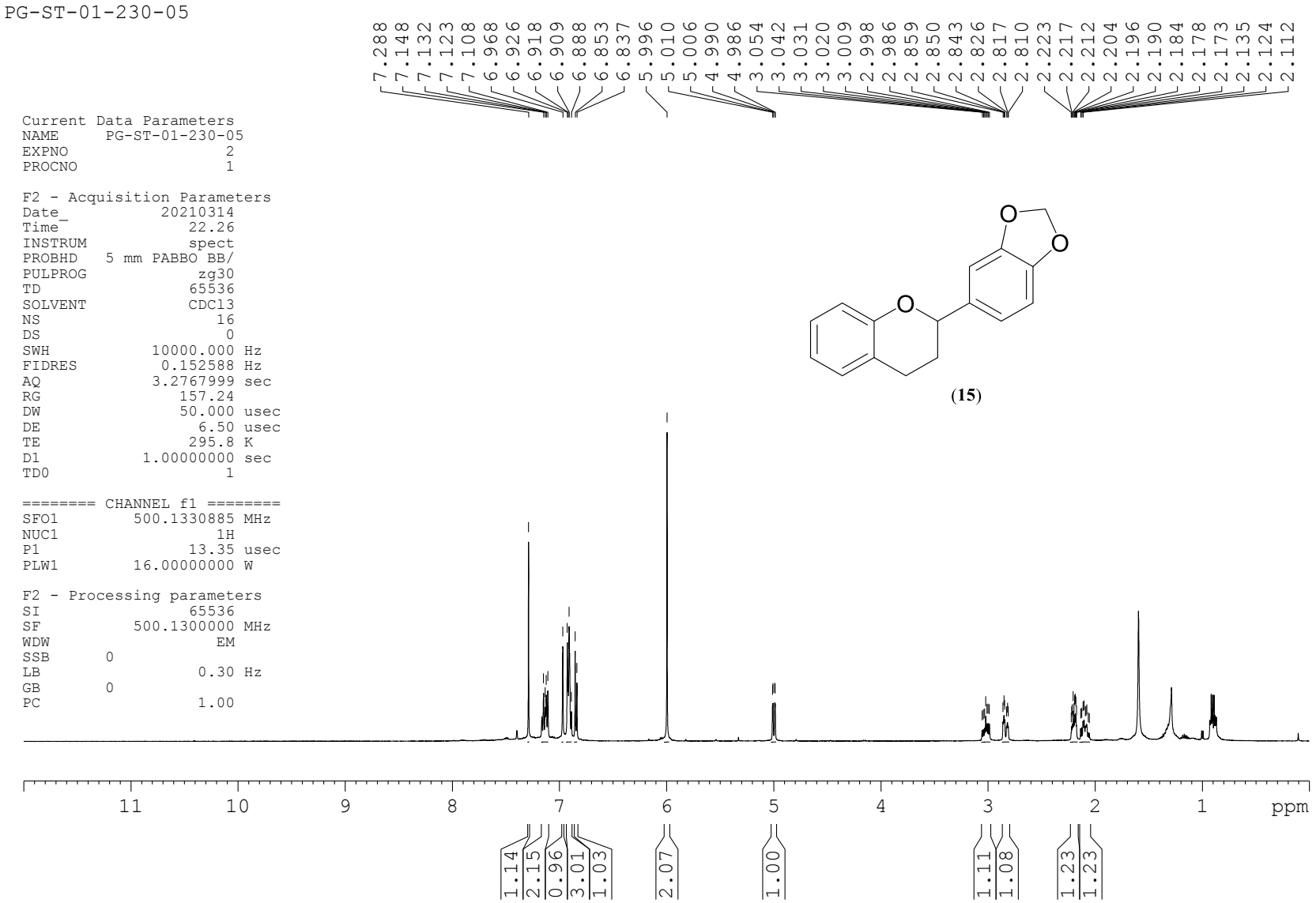


Figure S139. ^{1}H NMR spectrum of (**15**) in $CDCl_3$.

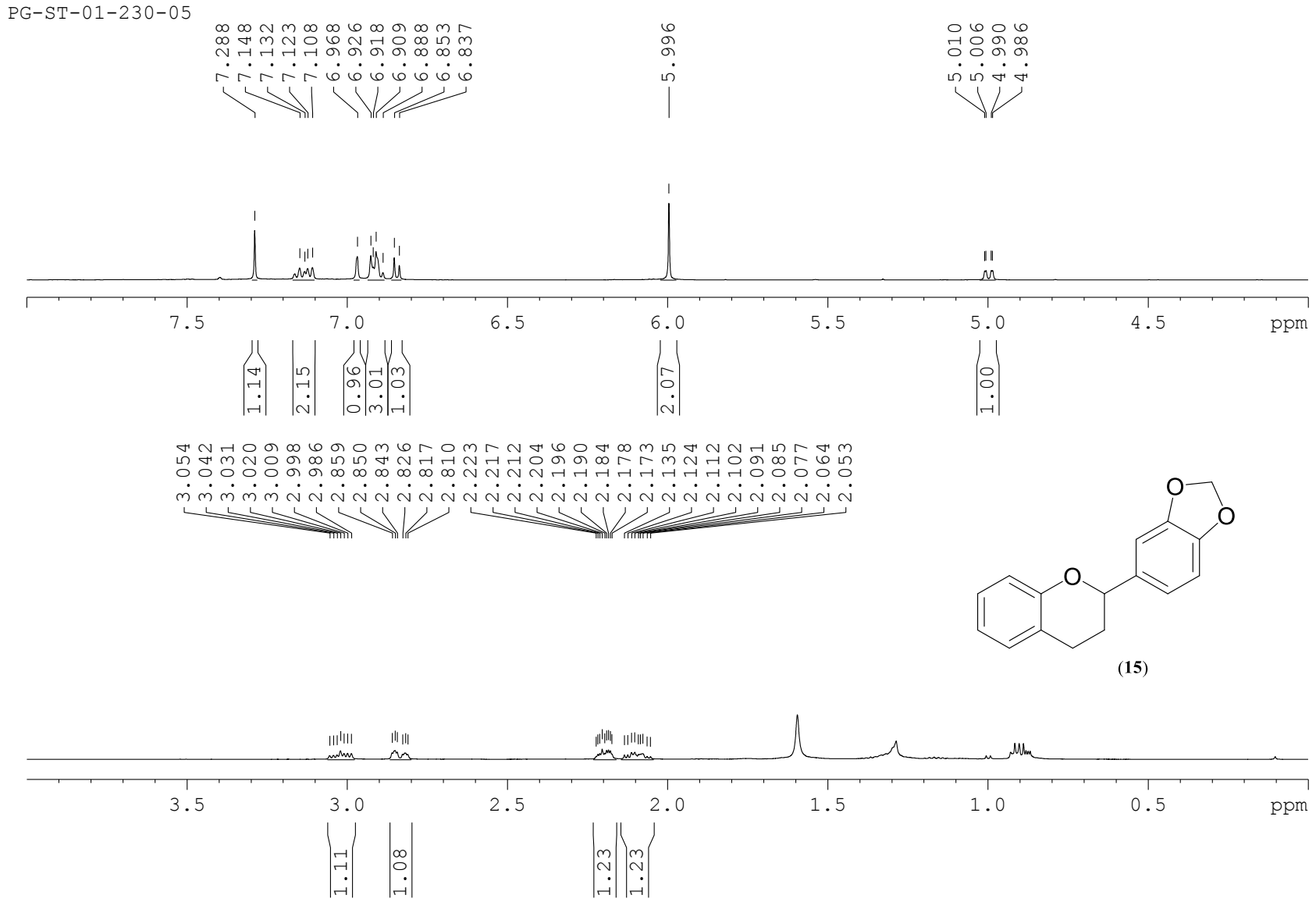


Figure S140. Expanded ^{1}H NMR spectrum of (**15**) in $CDCl_3$.

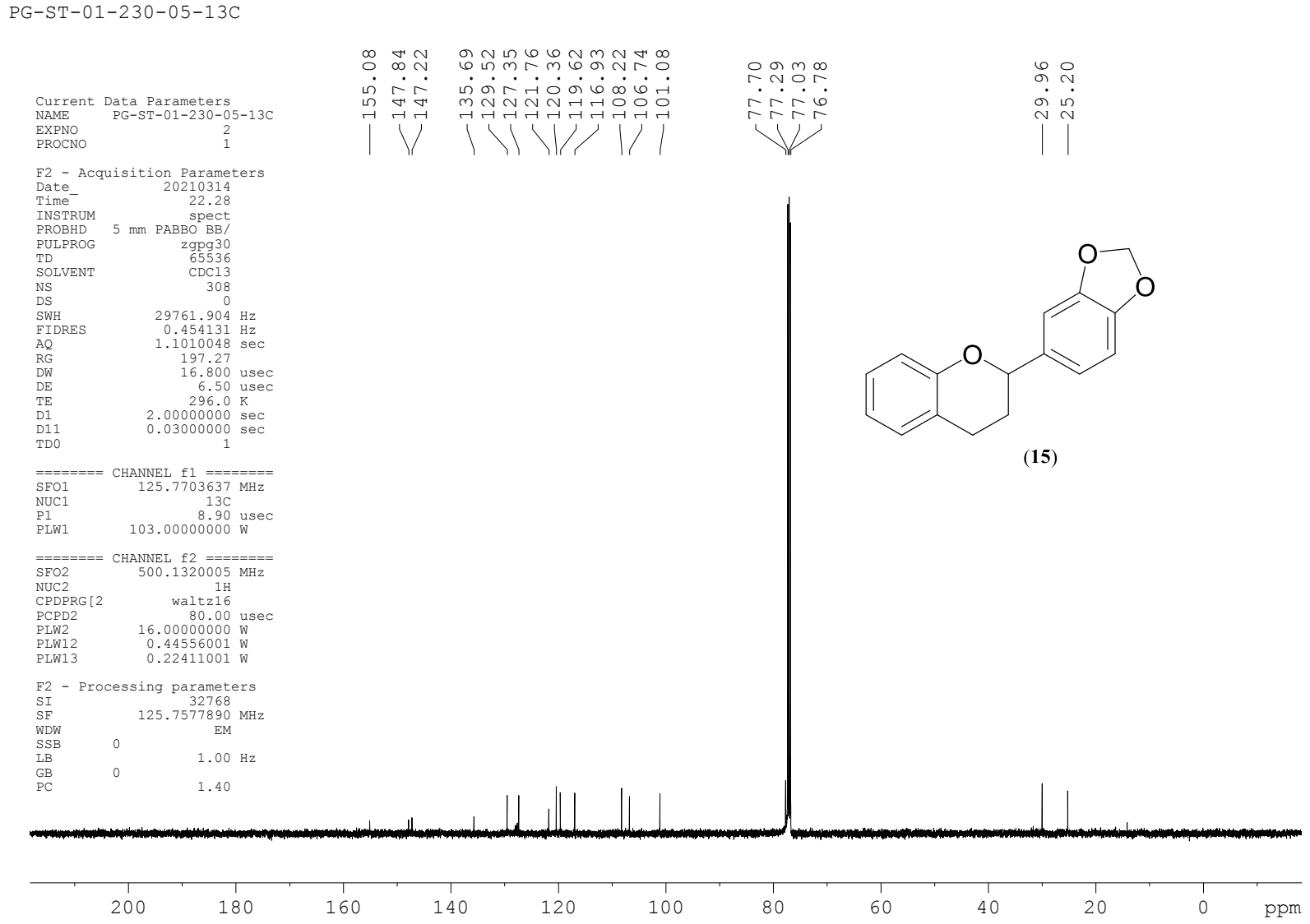


Figure S141. $^{13}C\{^{1}H\}$ NMR spectrum of (**15**) in $CDCl_3$.

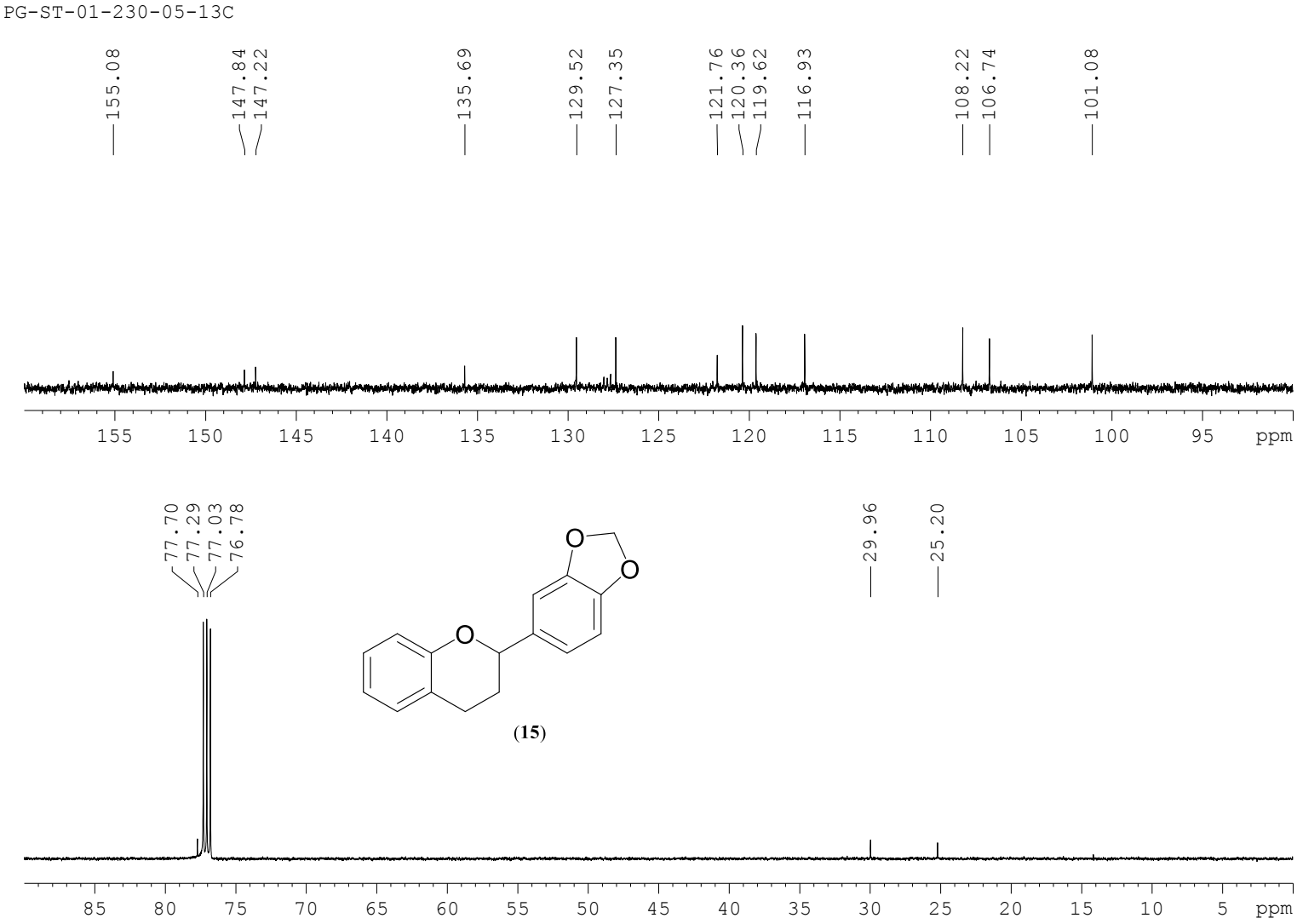


Figure S142. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**15**) in $CDCl_3$.

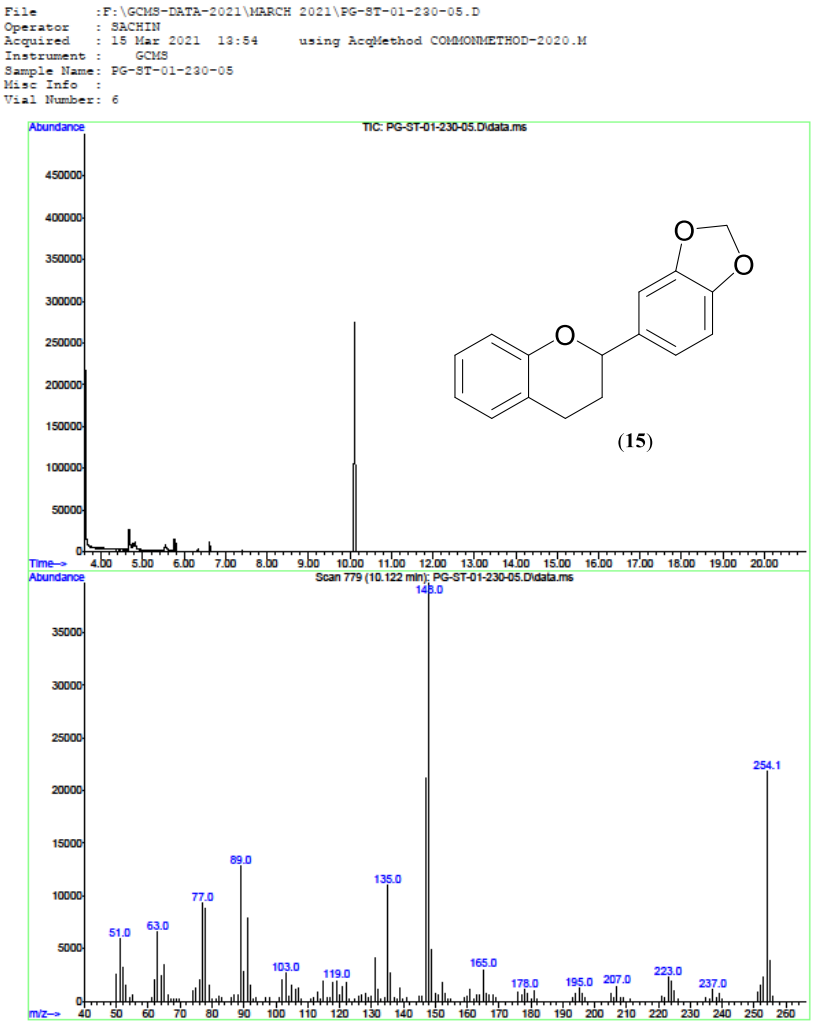
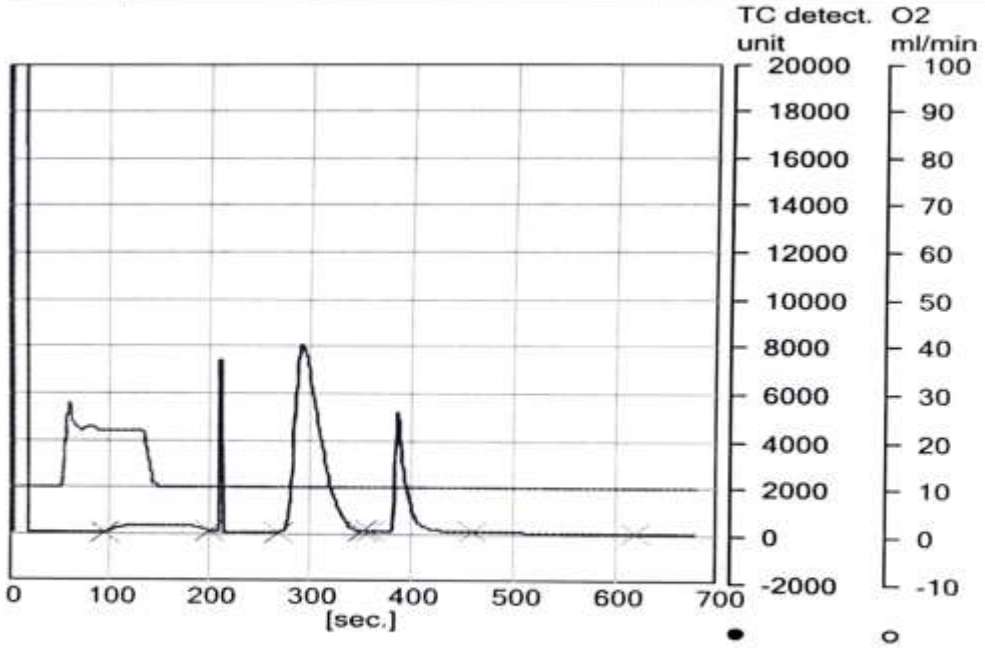
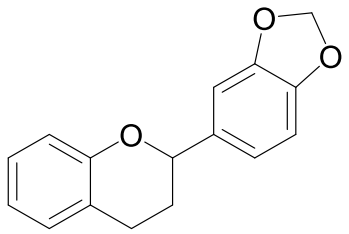


Figure S143. GCMS trace in EtOAc of (**15**) showing the M^+ peak at *m/z* 254.

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	N [%]	C [%]	H [%]	Date Time
36	1.0540	PG-ST-01-230-05-2	2mgChem80s	2 174	22 272	6 036	0.00	74.42	4.292	01-04-2021 18:35





(**15**)

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Figure S144. Elemental analysis data (**15**).

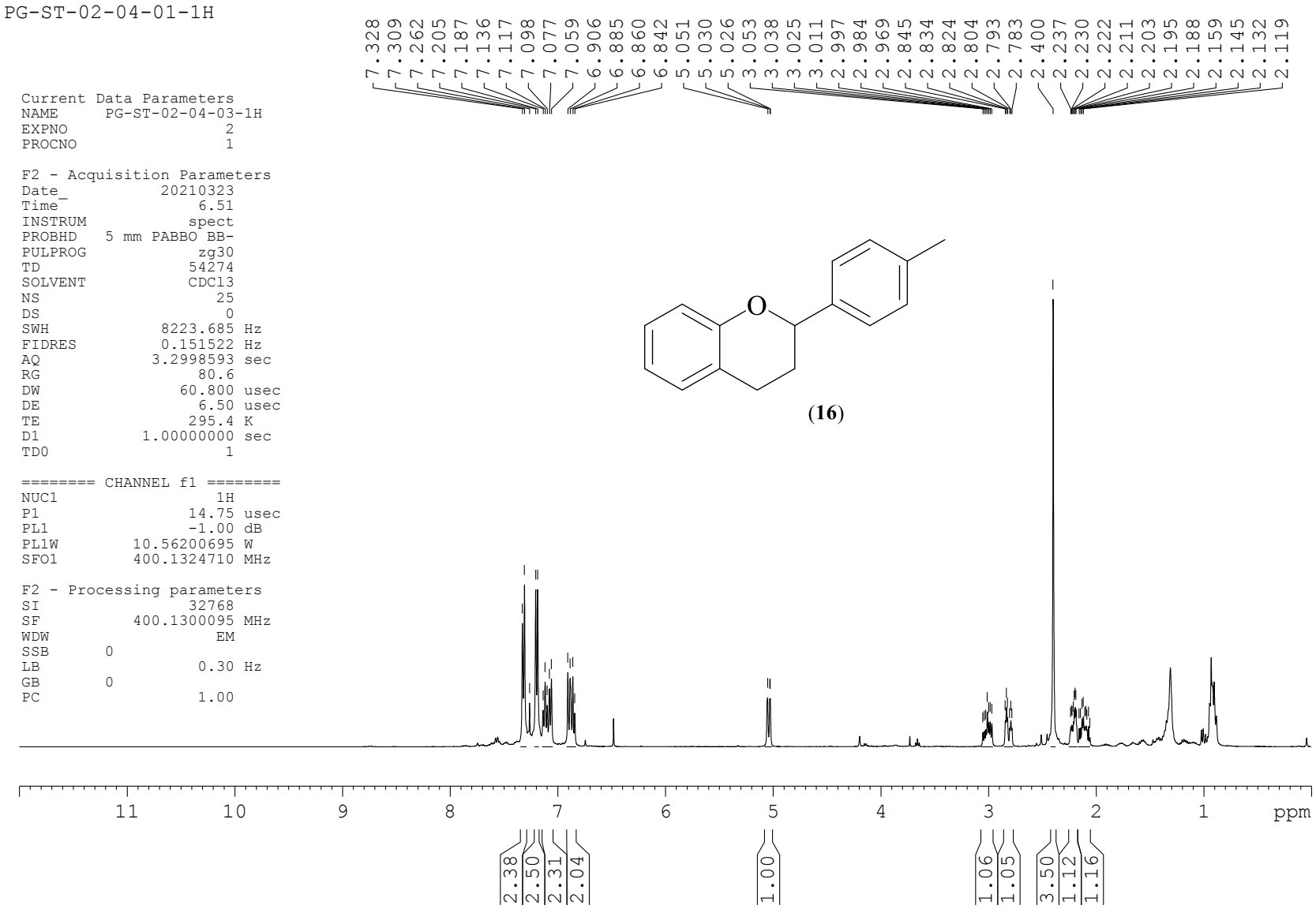


Figure S145. ^{1}H NMR spectrum of (**16**) in $CDCl_3$.

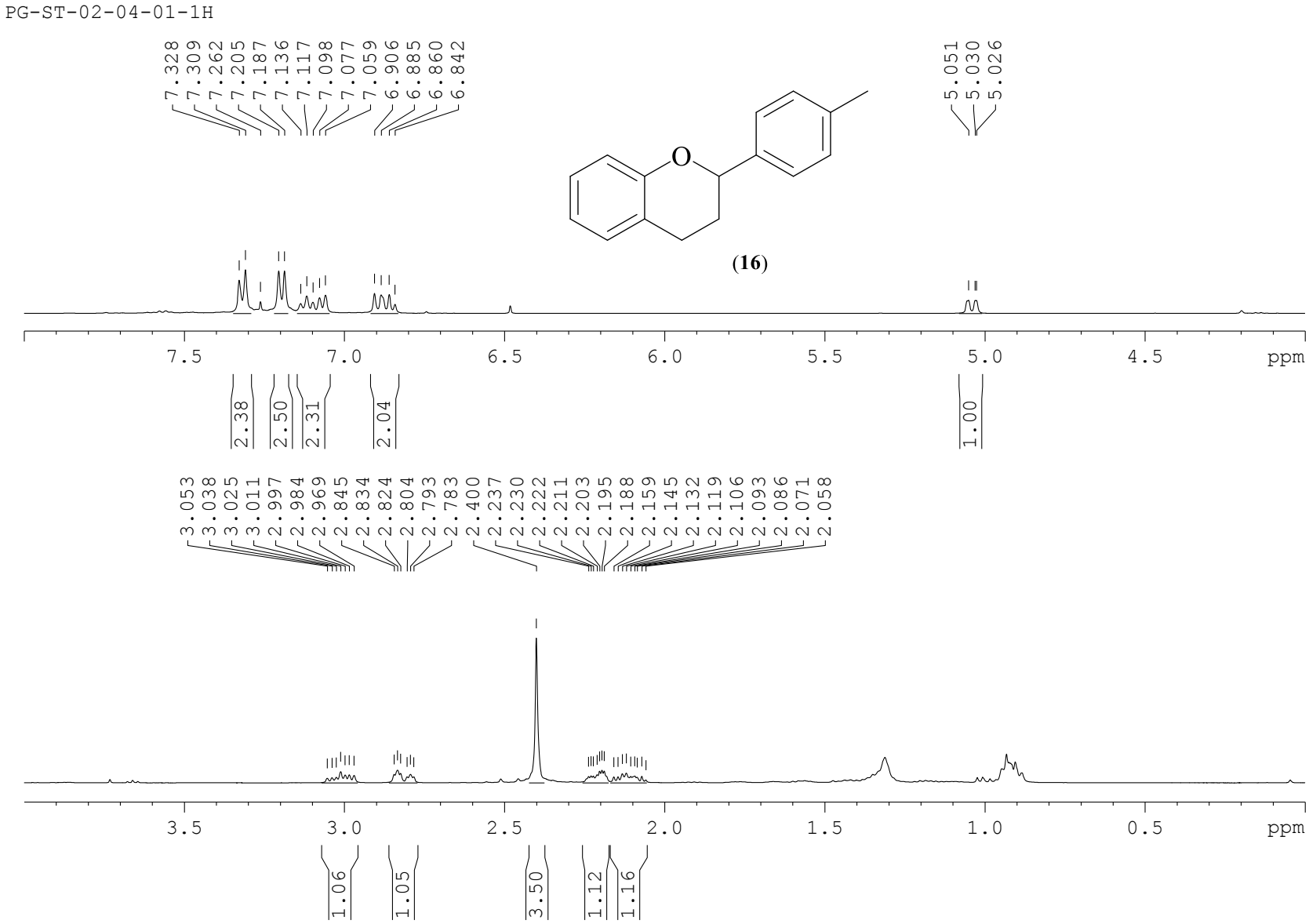


Figure S146. Expanded ^{1}H NMR spectrum of (**16**) in $CDCl_3$.

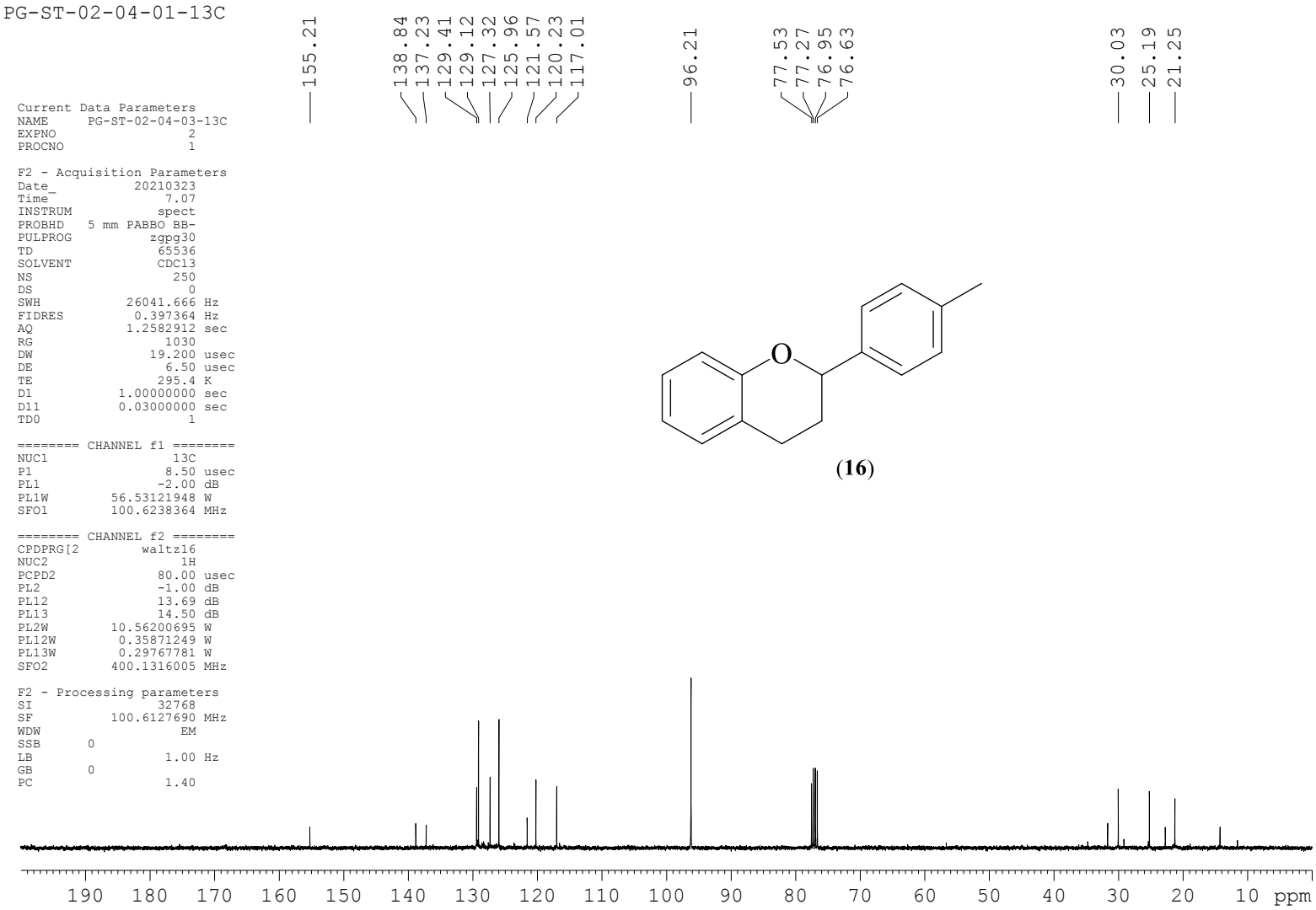


Figure S147. $^{13}C\{^{1}H\}$ NMR spectrum of (**16**) in $CDCl_3$.

(**16**)

Figure S148. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**16**) in $CDCl_3$.

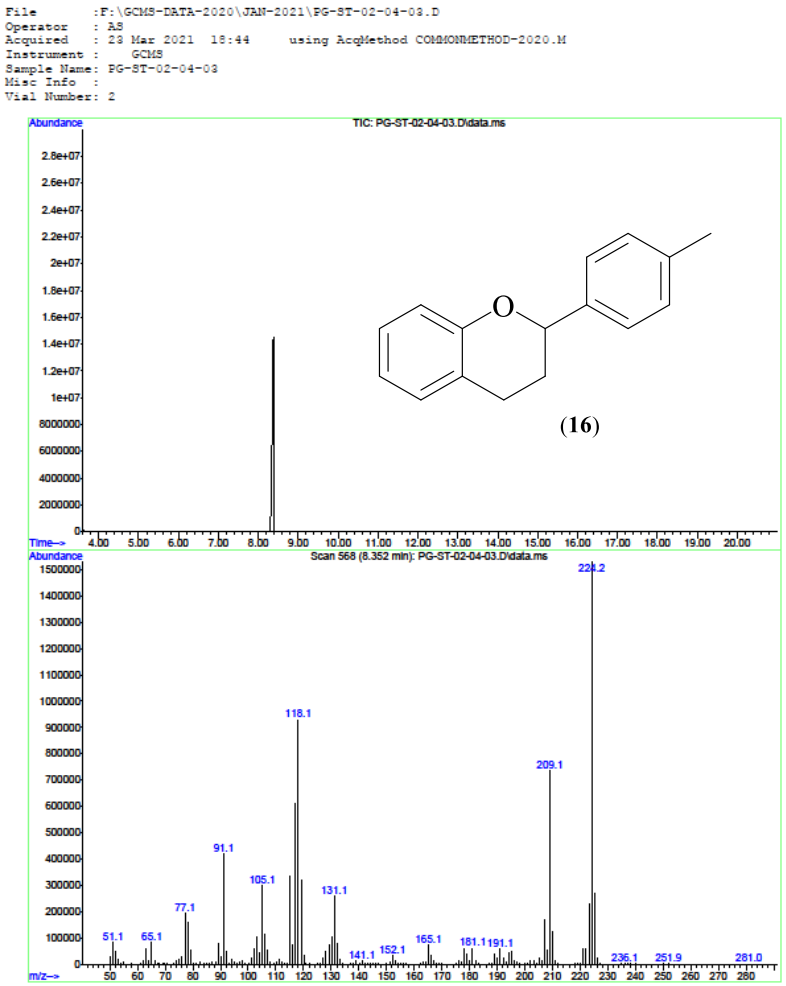
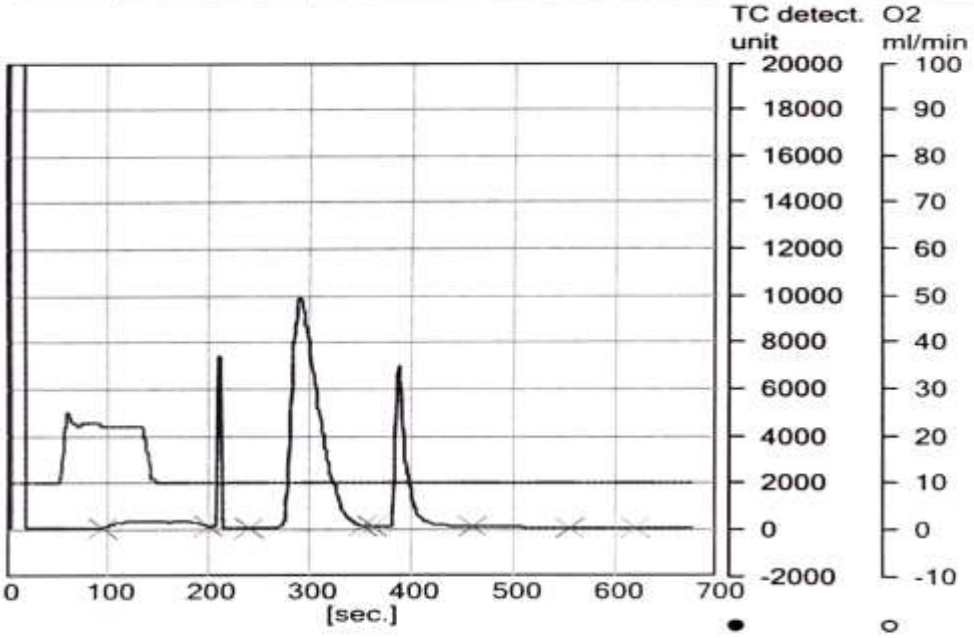
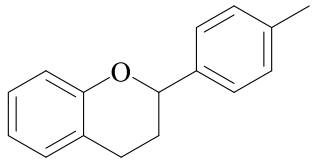


Figure S149. GCMS trace in EtOAc of (**16**) showing the M^+ peak at *m/z* 224.

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	N [%]	C [%]	H [%]	Date Time
38	1.2100	PG-ST-02-04-01-2	2mgChem80s	2 150	29 310	8 439	0.00	85.41	6.353	01-04-2021 18:58





(**16**)

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Figure S150. Elemental analysis data (**16**).

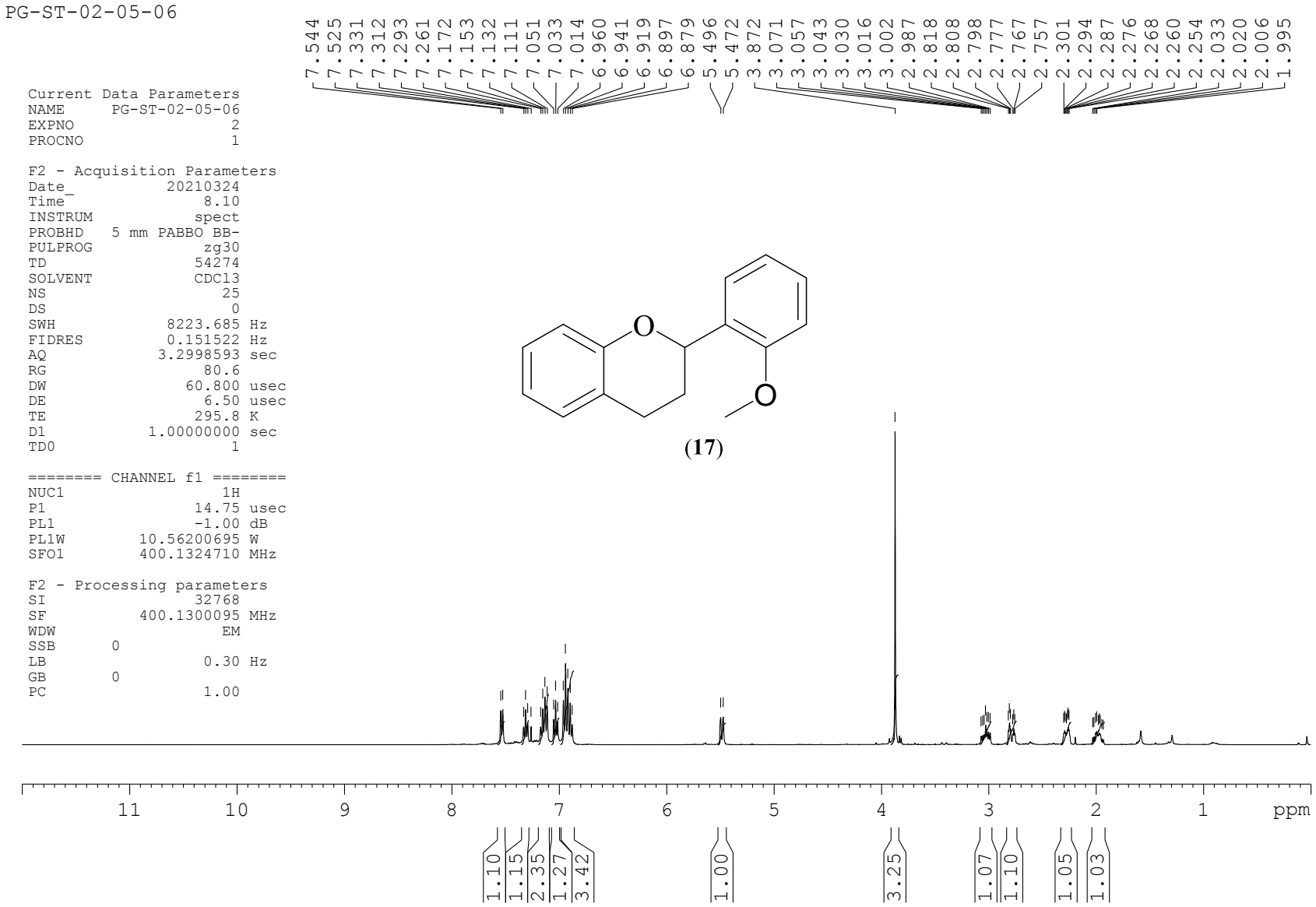


Figure S151. ^{1}H NMR spectrum of (**17**) in $CDCl_3$.

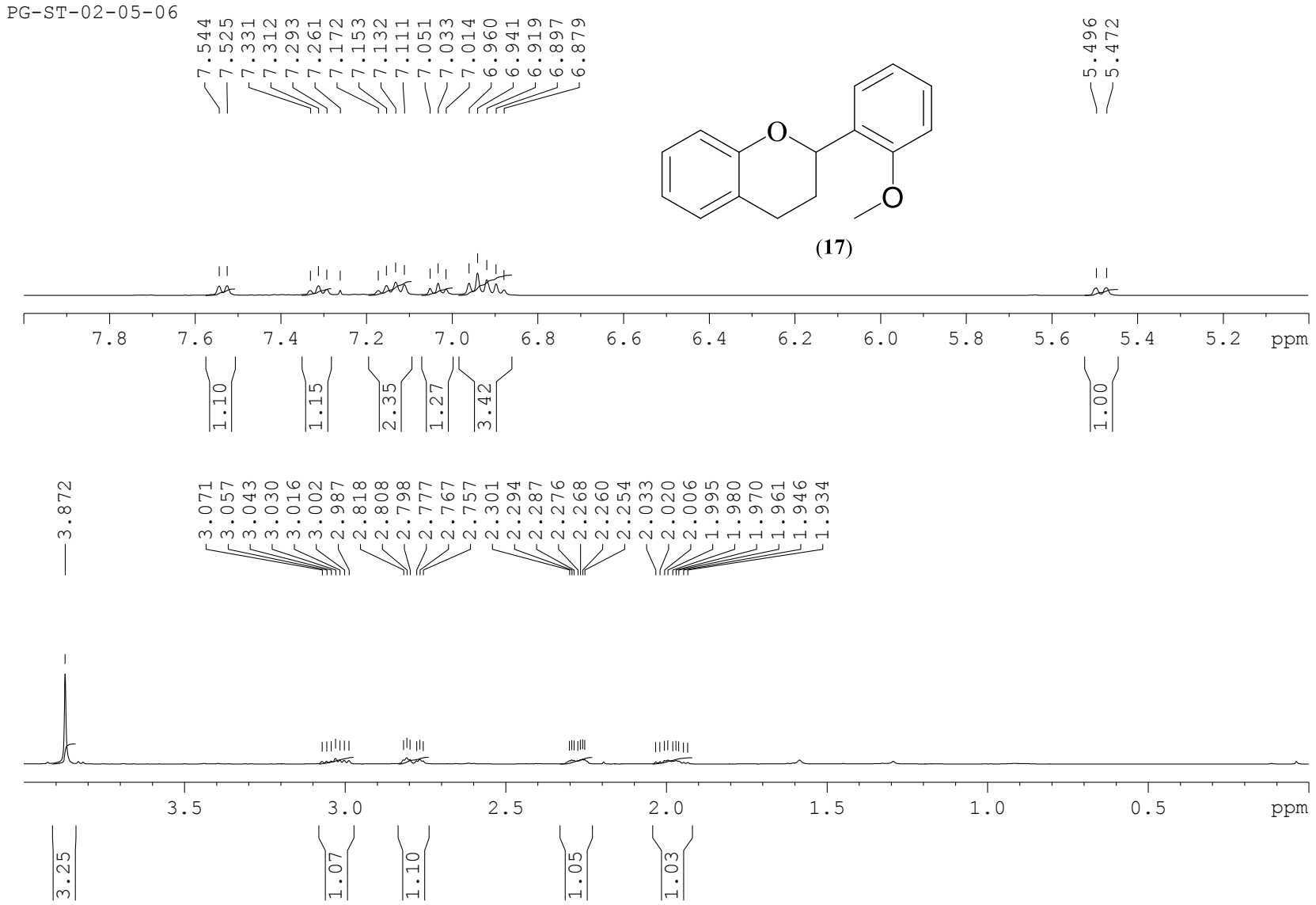
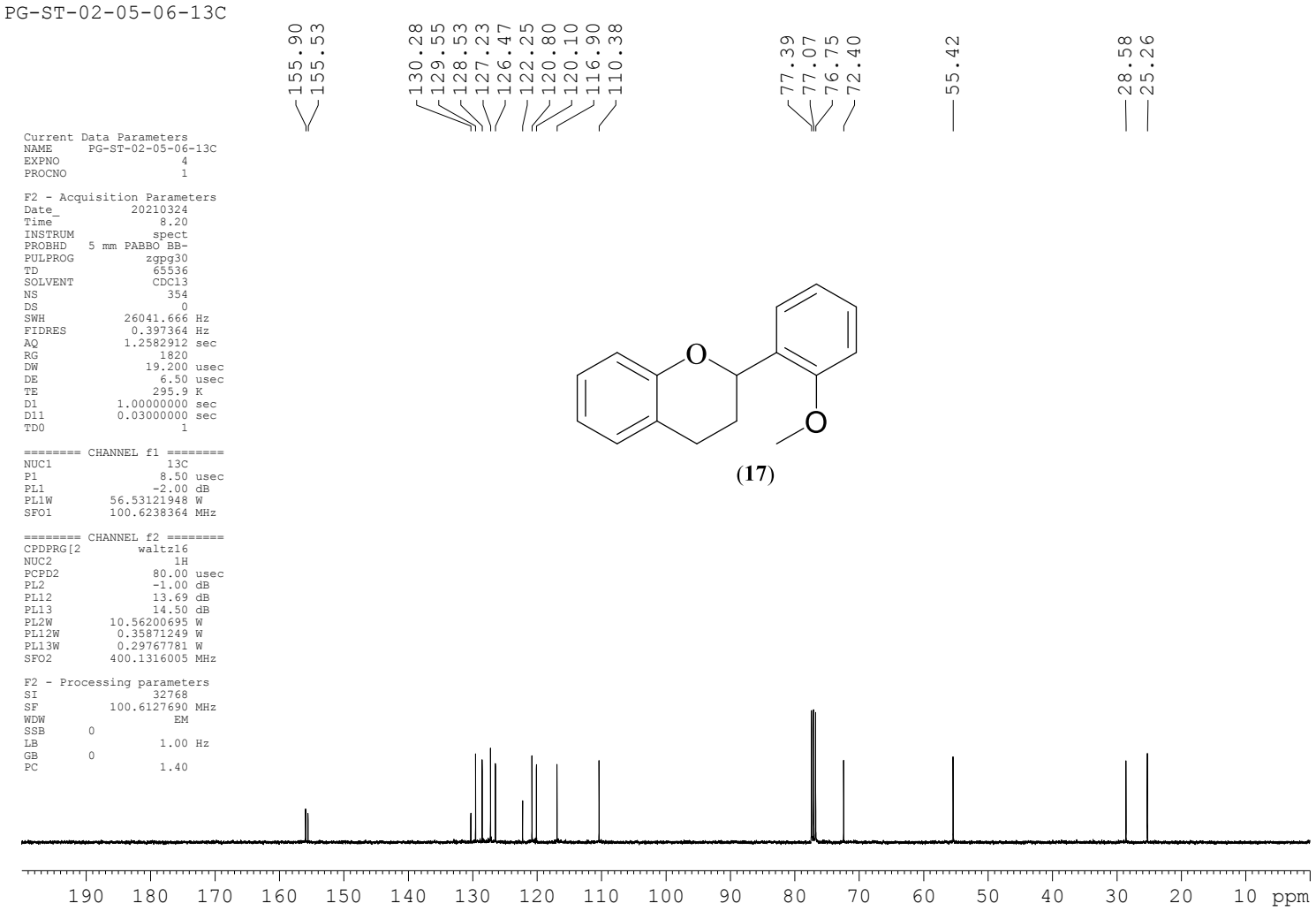


Figure S152. Expanded ^{1}H NMR spectrum of (**17**) in $CDCl_3$.



(**17**)

Figure S153. $^{13}C\{^{1}H\}$ NMR spectrum of (**17**) in $CDCl_3$.

Figure S154. Expanded $^{13}C\{^{1}H\}$ NMR spectrum of (**17**) in $CDCl_3$.

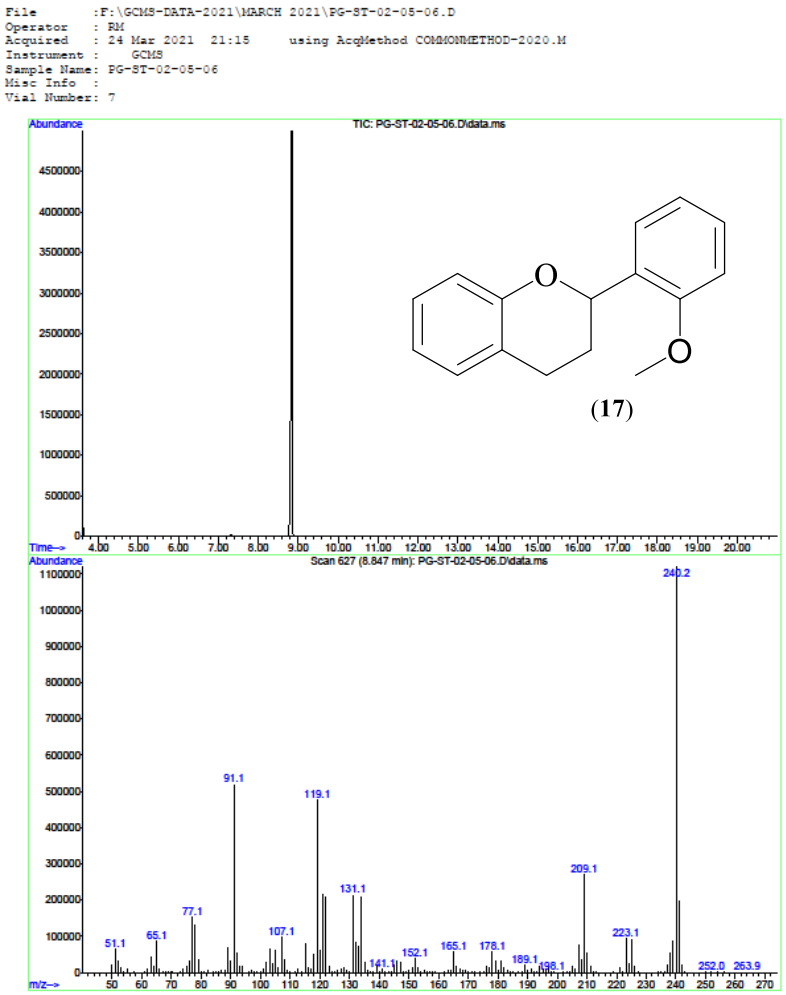
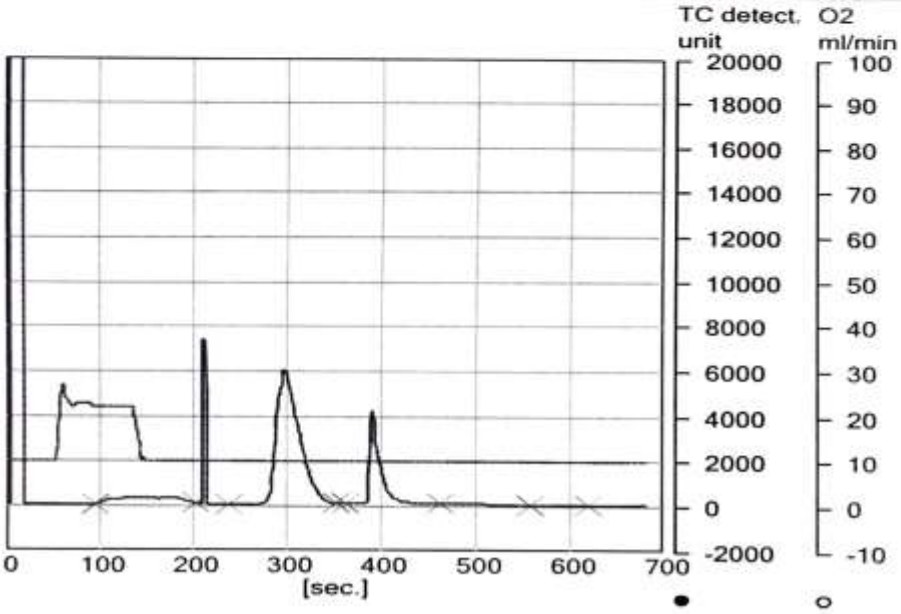
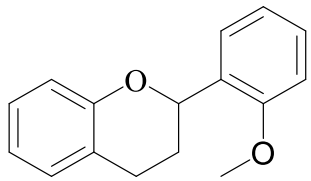


Figure S155. GCMS trace in EtOAc of (**17**) showing the M^+ peak at *m/z* 240.

No.	Weight [mg]	Name	Method	N Area	C Area	H Area	N [%]	C [%]	H [%]	Date	Time
40	0.7340	PG-ST-02-05-06-2	2mgChem80s	2 155	16 456	5 132	0.00	79.31	6.470	01-04-2021	19:21





(**17**)

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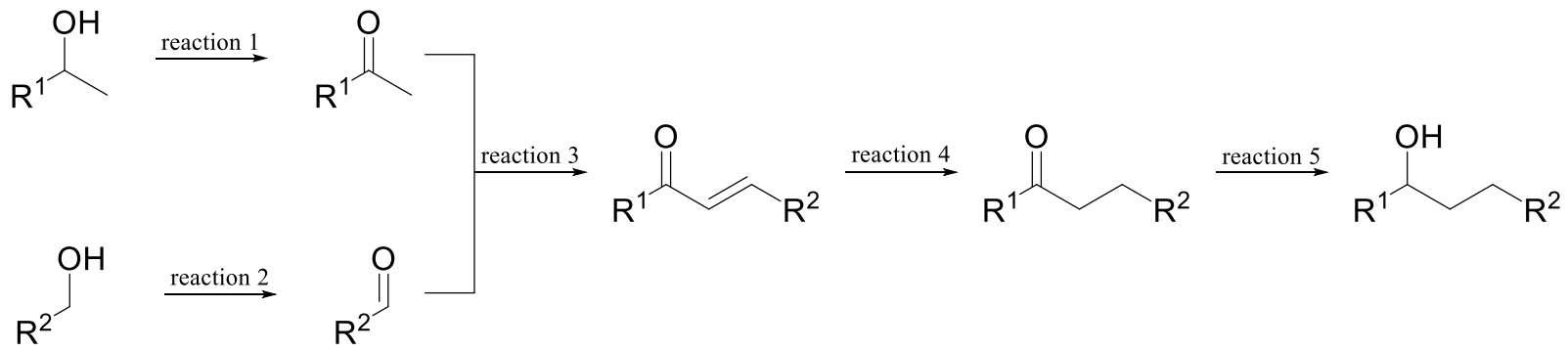
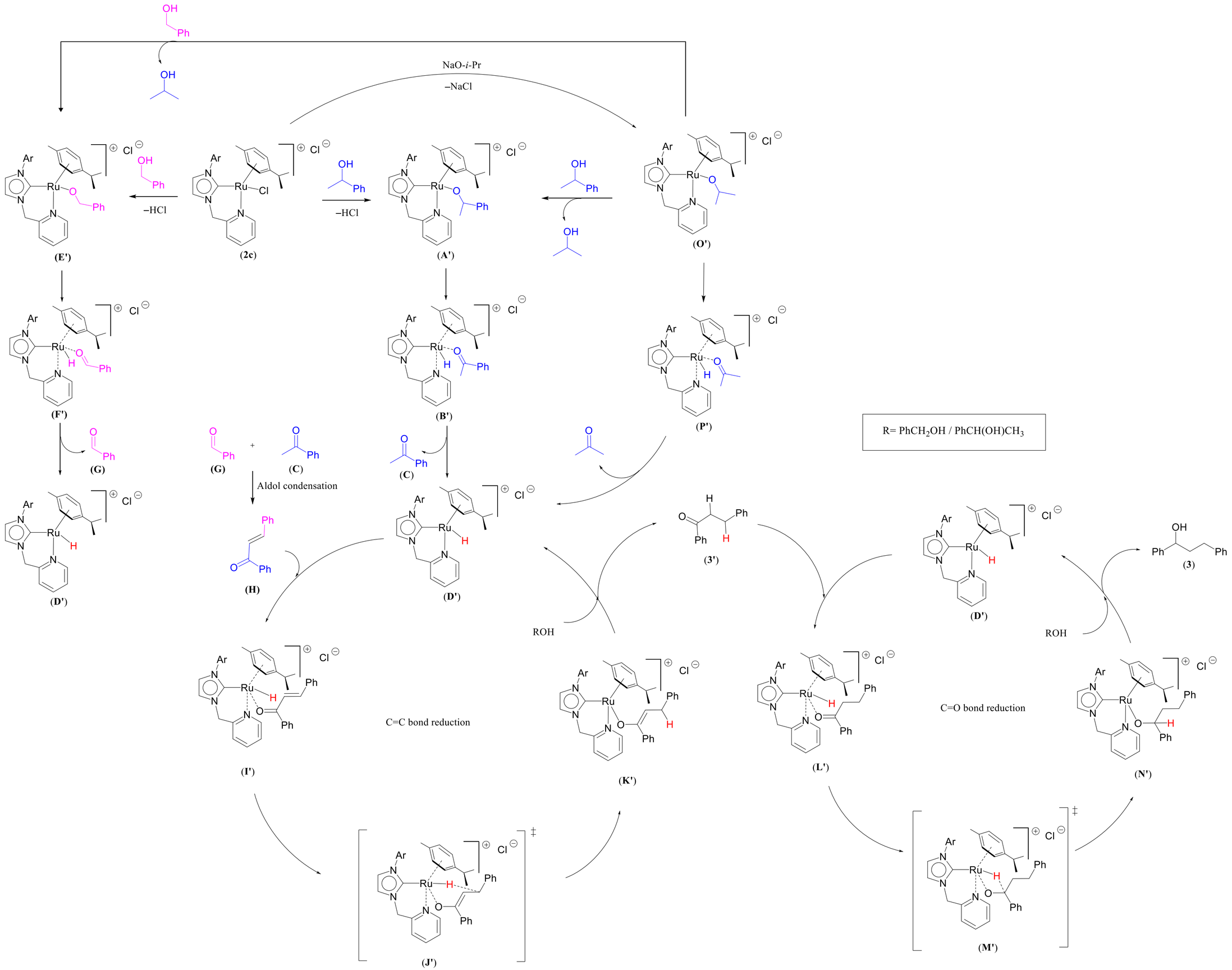
Figure S156. Elemental analysis data (**17**).



Scheme S1. One pot tandem β-alkylation reaction of secondary alcohol involving five sequential reactions.



Scheme S2. Proposed mechanism for the Ru−NHC (**2c**) catalyzed one pot tandem β-alkylation reaction for representative substrates namely 1-phenylethanol and benzyl alcohol.

Table S1. X−ray crystallographic data for Ag–NHC complex (**2b**), Ru–NHC complexes (**1c**), (**1c'**) and **2c**.

compound	(**1c**)	(**1c'**)	(**2b**)	(**2c**)
Lattice	Monoclinic	Triclinic	Monoclinic	Triclinic
Formula	$C_{32}H_{38}N_3ORuCl$	$C_{32}H_{38}N_3ORuCl$	$C_{17}H_{17}N_3AgCl$	$C_{27}H_{31}N_3RuCl_2{\bullet}2H_2O$
Formula weight	617.17	617.17	406.65	605.55
Space group	P121/n1	P–1	P121/n1	P–1
a/Å	9.0361(6)	8.0275(3)	9.8162(2)	8.2367(3)
b/Å	30.505(2)	13.1953(5)	16.0104(3)	9.1208(3)
c/Å	10.5031(6)	15.2588(6)	10.8039(2)	17.8719(5)
α/°	90.000	103.407(3)	90.000	93.836(2)
β/°	93.623(5)	104.358(3)	90.098(2)	93.825(2)
γ/°	90.000	102.686(3)	90.000	97.623(3)
V/Å^3	2889.4(3)	1455.78(10)	1697.95(6)	1323.90(8)
Z	4	2	4	2
Temperature (K)	150(2)	150(2)	150(2)	150(2)
Radiation (λ,Å)	0.71073	0.71073	0.71073	0.71073
ρ(calcd.), g cm^{-3}	1.419	1.408	1.591	1.519
μ(Mo Kα), mm^{-1}	0.665	0.660	1.343	0.824
θ max, deg.	25.00	25.00	31.101	24.999
No. Of data	3630	4556	4147	3922
No. Of parameters	351	351	201	327
R_1	0.0617	0.0554	0.0268	0.0450
wR_2	0.1500	0.1422	0.0634	0.1094
GOF	1.061	1.068	1.054	1.054

Table S2. A comparison of the metrical data of the neutral (amido-N functionalized NHC)Ru(*p*-cymene)Cl type complexes known in the literature is shown.

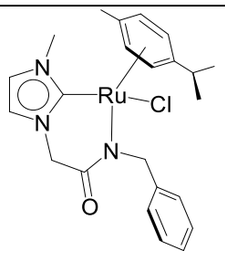
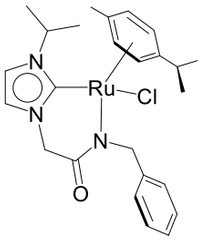
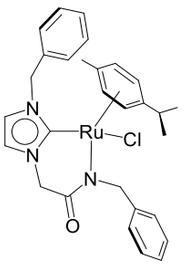
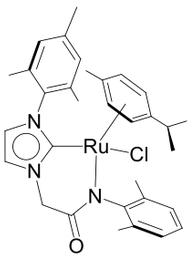
S. No.	complex	*d*(Ru–$C_{carbene}$)/ (Å)	*d*(Ru–N)/ (Å)	*d*(Ru–Cl)/ (Å)	*d*(Ru–$C_{centroid}$)/ (Å)	reference
1.		2.0172(19)	2.1074(16)	2.4404(7)	1.706	[6]
2.		2.033(5)	2.125(5)	2.4256(14)	1.712	[6]
3.		2.019(3)	2.1074(16)	2.4325(8)	1.719	[6]
4.	 (**1c**)	2.087(5)	2.153(4)	2.4299(14)	1.736	present work

Table S3. A comparison of the metrical data of the representative structurally characterized examples of neutral (unsubstituted picolyl functionalized NHC)AgCl and neutral (unsubstituted pyridyl functionalized NHC)AgCl type complexes known in the literature is shown.

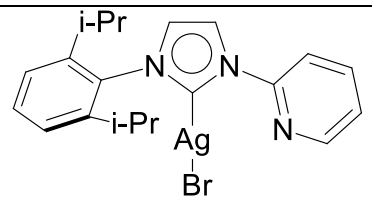
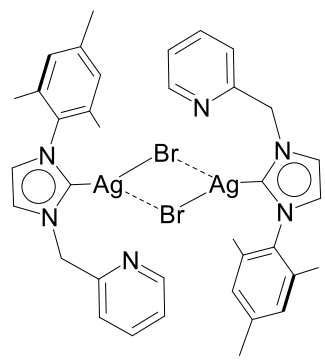
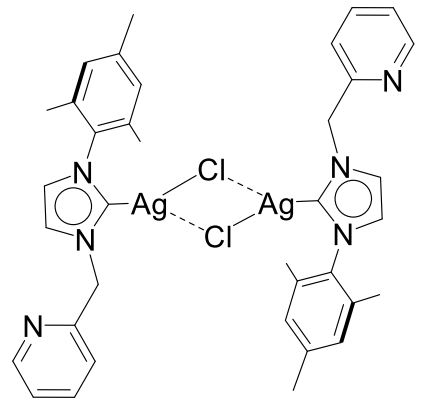
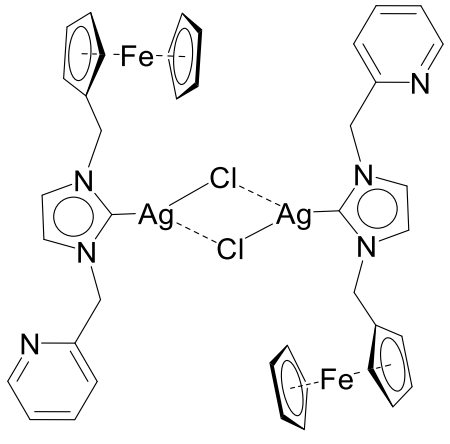
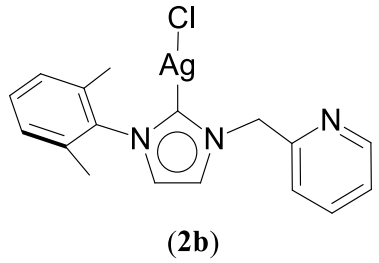
S. No.	complex	*d*(Ag–$C_{carbene}$)/ (Å)	*d*(Ag–X) (X=Br, Cl)/ (Å)	reference
1.		2.075(7)	2.421(1)	[7]
2.		2.07(2)	2.373(4) and 2.952(4)	[7]
3.		2.0762(19)	2.0763(19) and 2.9916(6)	[7]
4.		2.097(2)	2.4119(8) and 2.830(1)	[7]
5.	 (**2b**)	2.0813(17)	2.3684(5)	present work

Table S4. A comparison of the metrical data of the representative structurally characterized examples of ionic (picolyl functionalized NHC)Ru(*p*-cymene)Cl type complexes known in the literature is shown.

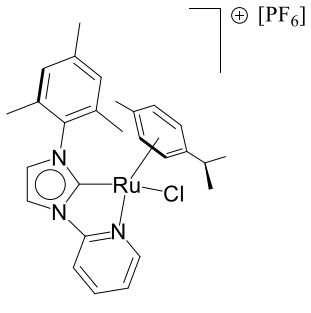
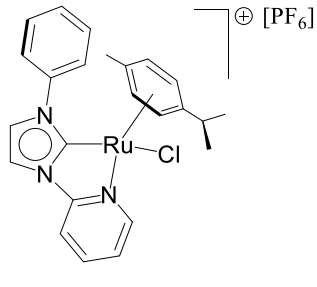
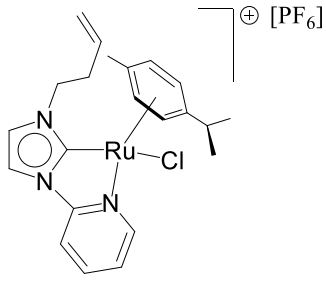
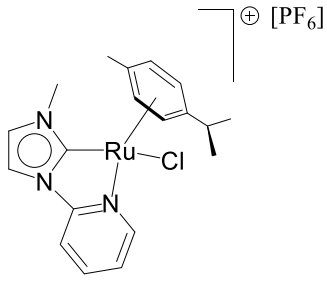
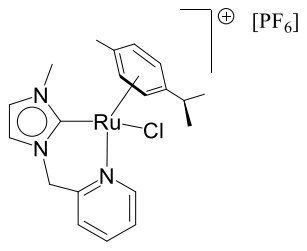
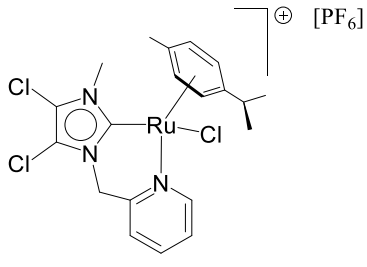
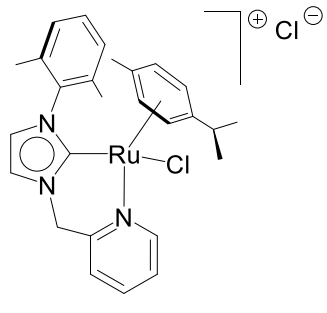
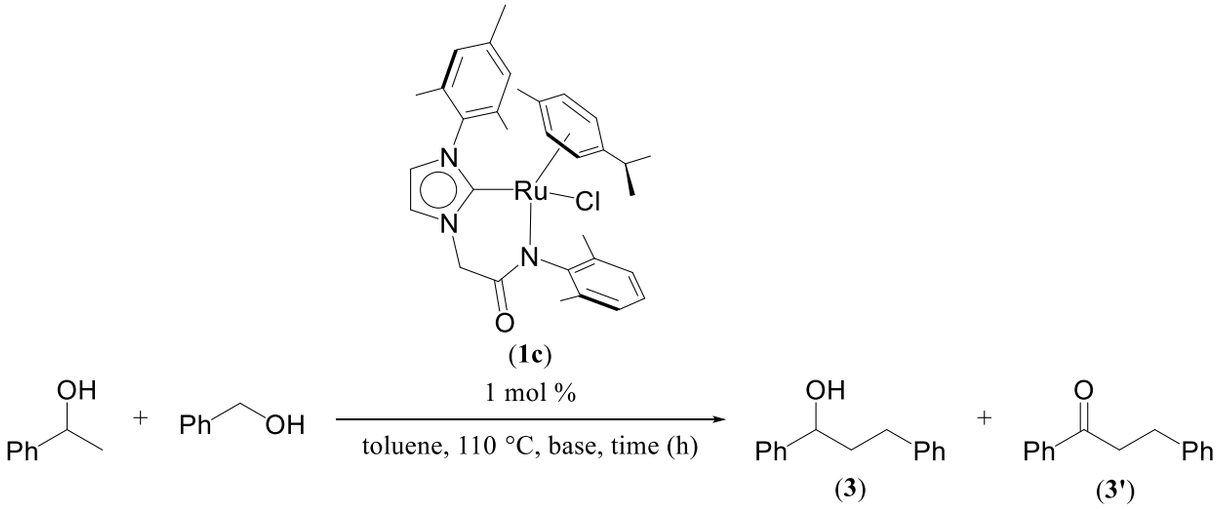
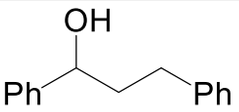
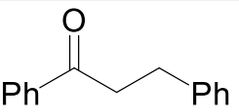
S. No.	complex	d(Ru–$C_{carbene}$)/ (Å)	d(Ru–N)/ (Å)	d(Ru–Cl)/ (Å)	d(Ru–$C_{centroid}$)/ (Å)	reference
1.		2.029(3)	2.098(3)	2.4615(7)	2.7214(15)	[8]
2.		2.0145(19)	2.1055(16)	2.4397(4)	1.7155(8)	[8]
3.		2.033(2)	2.100(2)	2.246(2)	1.712	[9]
4.		2.009(2)	2.092(18)	2.401(6)	1.710(1)	[10]
5.		2.035(7)	2.095(6)	2.389(2)	1.710(3)	[11]
6.		2.023(5)	2.111(4)	2.448(1)	1.696(2)	[11]
7.	 (**2c**)	2.052(4)	2.110(3)	2.3936(9)	1.704	present work

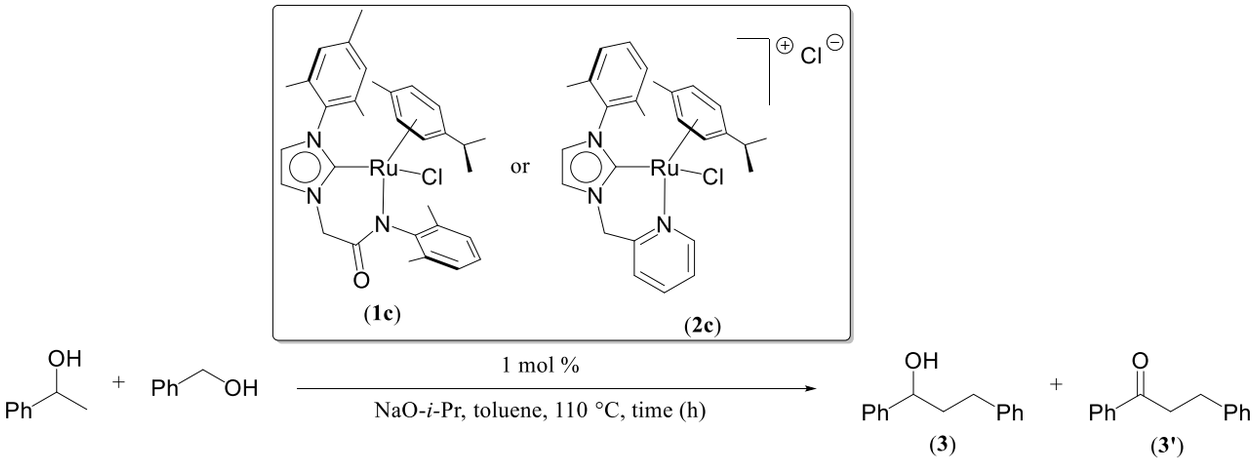
Table S5. Base variation study for the Ru−NHC (**1c**) catalyzed one pot tandem β-alkylation reaction for two representative substrates namely 1-phenylethanol and benzyl alcohol[a].

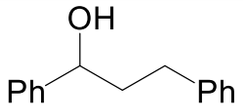
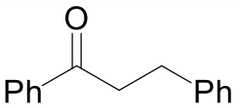


S.No	base	time (h)	yield[b]	
				
			(**3**)	(**3'**)
1	NaOH	6	37	52
2	KO-*t*-Bu	6	63	17
3	Et_3N	6	ND	ND
4	K_2CO_3	6	ND	ND
5	KOH	6	69	19
6	KOH	3	55	18
7	NaO*i*Pr	6	55	40
8	NaO*i*Pr	3	72	11

(*a*). Reaction conditions: 1:1:1 ratio of 1°-alcohol:2°-alcohol:base 1.00 mmol, 1 mol % of (**1c**), 2.0 mL of toluene at 110 °C for time (h). (*b*). Isolated yields (%).

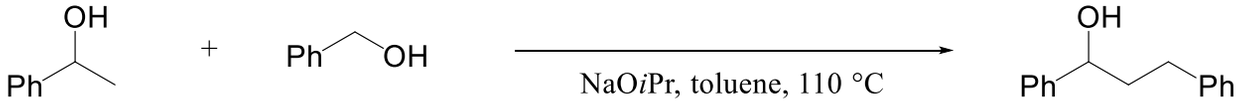
Table S6. Time variation study for the Ru−NHC (**1c**/**2c**) catalyzed one pot tandem β-alkylation reaction for two representative substrates namely 1-phenylethanol and benzyl alcohol[a].



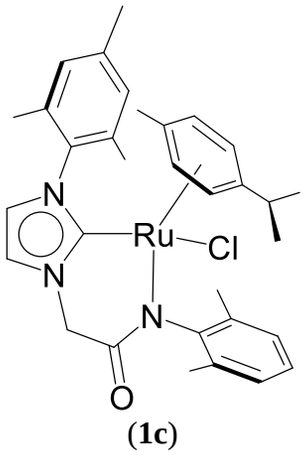
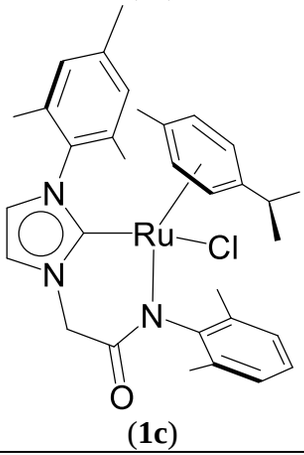
S.No	time (h)	yield[b]			
		 (**3**)		 (**3'**)	
		(**1c**)	(**2c**)	(**1c**)	(**2c**)
1	0.5	18	12	15	5
2	1	37	27	34	12
3	2	51	35	33	14
4	3	72	68	11	13
5	4	61	46	19	22
6	6	55	38	40	28
7	12	1	6	64	32
8	24	61	51	9	9
9	48	63	50	5	7
10	72	55	32	3	3
11	96	46	28	> 5	3
12	120	22	15	> 5	2

(*a*). Reaction conditions: 1:1:1 ratio of 1°-alcohol:2°-alcohol:NaO*i*Pr 1.00 mmol, 1 mol % of (**1c**/**2c**), 2.0 mL of toluene at 110 °C for T hour. (*b*) Isolated yields (%).

Table S7. Selected results of blank, control and Hg drop experiments for the one pot tandem β-alkylation reaction for two representative substrates namely 1-phenylethanol and benzyl alcohol[a].

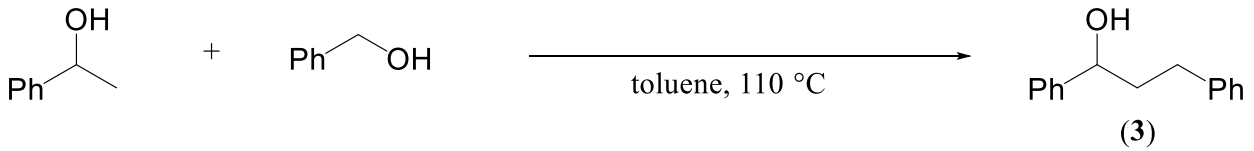


(**3**)

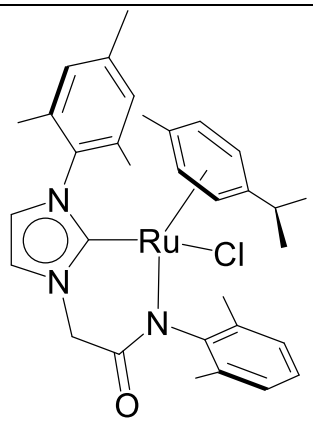
S. No	metal complex	Hg	yield[b]
1	-		ND
2	[(*p*-cymene)$RuCl_2$]$_2$		19
3	 (**1c**)		72
4	 (**1c**)	Hg	68

(*a*). Reaction conditions: 1:1:1 ratio of 1°-alcohol:2°-alcohol:NaO*i*Pr 1.00 mmol, 1 mol % of (**1c**), 2.0 mL of toluene at 110 °C for 3 hours. (*b*). Isolated yields (%).

Table S8. Selected results of blank and control experiments for the one pot tandem β-alkylation reaction for two representative substrates namely 1-phenylethanol and benzyl alcohol[a].

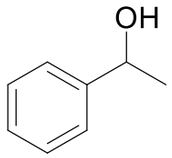
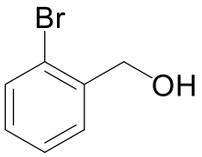
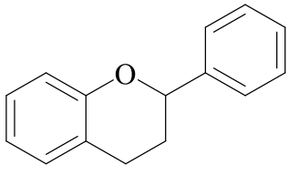
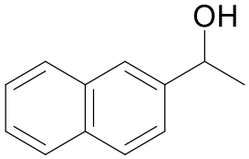
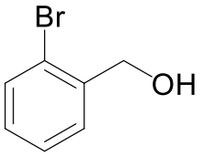
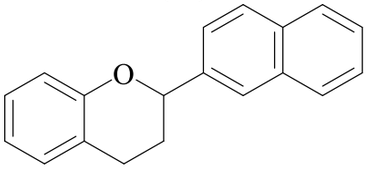
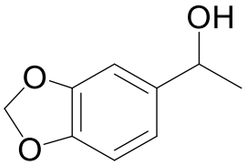
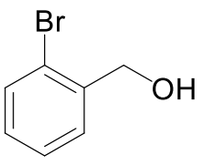
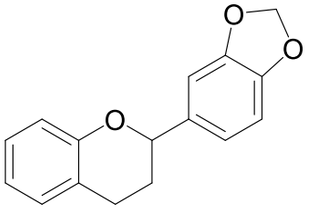
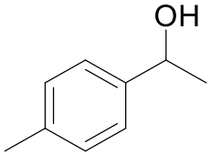
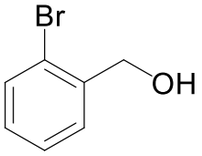
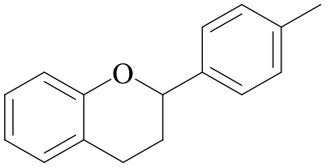
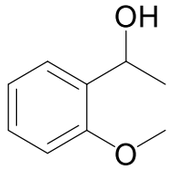
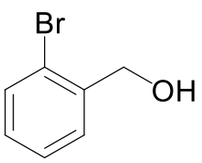
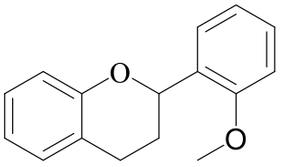


(**3**)

S. No	metal complex	base	yield[b]
1	 (**1c**)	–	ND
2	–	NaO*i*Pr	ND

(*a*). Reaction conditions: 1:1:1 ratio of 1°-alcohol:2°-alcohol:NaO*i*Pr 1.00 mmol, 1 mol % of (**1c**), 2.0 mL of toluene at 110 °C for 3 hours. (*b*). Isolated yields (%).

Table S9. Selected results for the Ru−NHC (**1**−**2**)**c** catalyzed one pot synthesis of flavan derivatives (**13**-**17**).

S. No.	2° alcohol	1° alcohol	product	Ru−NHC (**1c**) yield[b]	Ru−NHC (**2c**) yield[b]
13			 (**13**)	17	10
14			 (**14**)	36	28
15			 (**15**)	18	15
16			 (**16**)	12	11
17			 (**17**)	14	13

Reaction conditions: (*a*). Reaction conditions: 1:1:1 ratio of 1°-alcohol:2°-alcohol:base 1.00 mmol, 1 mol % of (**1c**/**2c**), 2.0 mL of toluene at 110 °C for 3 hours. (*a*). Reaction conditions: 20 mol % CuI, 20 mol % 2,2′-bipyridine, base 1.00 mmol, 1.0 mL of toluene at 110 °C for 24 hours. (*c*). Isolated yields (%).

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