

SUPPORTING INFORMATION

“All-water” chemistry of tandem *N*-alkylation-reduction-condensation for synthesis of *N*-arylmethyl-2-substituted benzimidazoles

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Spectral data

***N*-Benzyl-4-methyl-2-nitroaniline (Entry 2, Table 3)**

Red solid (232 mg, 96%); mp = 92-94 °C; IR (Neat) ν : 3445, 2895, 1612, 1573, 1523, 1359, 1162 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 8.34 (brs, 1H), 7.80 (s, 1H), 7.34-7.25 (m, 5H), 7.25-7.19 (m, 1H), 6.72 (d, J = 8.64 Hz, 1H), 4.53 (d, J = 5.28 Hz, 2H), 2.25 (s, 3H).¹⁶

***N*-Benzyl-4-chloro-2-nitroaniline (Entry 3, Table 3)**

Red solid (249 mg, 95%); mp = 68-70 °C; IR (Neat) ν : 3428, 2932, 1618, 1571, 1518, 1358, 1162 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 8.40 (brs, 1H), 8.17 (d, J = 2.56 Hz, 1H), 7.39-7.28 (m, 6H), 6.76 (d, J = 9.20 Hz, 1H), 4.53 (d, J = 5.68 Hz, 2H).¹⁶

***N*-Benzyl-5-methyl-2-nitroaniline (Entry 4, Table 3)**

Red solid (232 mg, 96%); mp = 101-103 °C; IR (Neat) ν : 3443, 2885, 1609, 1575, 1528, 1360, 1169 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 8.43 (brs, 1H), 8.14 (d, J = 9.12 Hz, 1H), 7.39-7.29 (m, 5H), 6.60 (s, 1H), 6.49-6.47 (m, 1H), 4.53 (d, J = 5.60 Hz, 2H), 2.29 (s, 3H).¹⁶

***N*-Benzyl-5-chloro-2-nitroaniline (Entry 5, Table 3)**

Red solid (249 mg, 95%); mp = 88-90 °C; IR (Neat) ν : 3425, 2935, 1616, 1575, 1520, 1350, 1270, 1160 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 8.43 (brs, 1H), 8.14 (d, J = 9.12 Hz, 1H), 7.41-7.30 (m, 5H), 6.83 (d, J = 2.08 Hz, 1H), 6.65-6.62 (m, 1H), 4.51 (d, J = 5.52 Hz, 2H).¹⁴

***N*-(3-Methoxybenzyl)-2-nitroaniline (Entry 6, Table 3)**

yellow solid (247 mg, 96%); mp = 110-112 °C; IR (Neat) ν : 3440, 2945, 1610, 1560, 1542, 1268, 1165 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 8.43 (brs, 1H), 8.21-8.19 (m, 1H), 7.38 (t, J = 8.44 Hz, 1H), 7.30-7.26 (m, 1H), 6.93 (d, J = 7.52 Hz, 1H), 6.88-6.85 (m, 1H), 6.85-6.79 (m, 2H), 6.69-6.65 (m, 1H), 5.52 (d, J = 5.60 Hz, 2H), 3.80 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ :

160.1, 145.3, 139.1, 136.2, 130.0, 126.9, 119.2, 115.8, 114.2, 112.9, 112.7, 55.3, 47.1. HRMS (+pESI) m/z calcd for $C_{14}H_{15}N_2O_3$ [$M + H^+$], 259.1083; Found 259.1085.

***N*-(4-Fluorobenzyl)-2-nitroaniline (Entry 7, Table 3)**

Yellow solid (236 mg, 96%); mp = 75-76 °C; IR (Neat) ν : 3420, 2945, 1610, 1570, 1511, 1352, 1151 cm^{-1} ; 1H NMR ($CDCl_3$, 400 MHz) δ : 8.39 (brs, 1H), 8.20 (dd, J = 1.52 & 8.56 Hz, 1H), 7.41-7.37 (m, 1H), 7.34-7.29 (m, 2H), 7.80-7.02 (m, 2H), 6.79-6.77 (m, 1H), 6.70-6.66 (m, 1H), 4.51 (d, J = 5.56 Hz, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ : 163.5, 161.1, 145.0, 136.3, 133.1, 133.0, 132.4, 128.7, 128.6, 126.9, 116.0, 115.9, 115.7, 114.1, 46.4.¹⁷

***N*-(4-Iodobenzyl)-2-nitroaniline (Entry 8, Table 3)**

yellow solid (336 mg, 95%); mp = 88-90 °C; IR (Neat) ν : 3432, 2939, 1612, 1565, 1538, 1252, 1155 cm^{-1} ; 1H NMR ($CDCl_3$, 400 MHz) δ : 8.42 (brs, 1H), 8.20 (dd, J = 1.56 & 8.56 Hz, 1H), 7.70-7.67 (m, 2H), 7.40-7.35 (m, 2H), 7.10 (d, J = 8.44 Hz, 2H), 6.74-6.67 (m, 2H), 4.50 (d, J = 5.64 Hz, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ : 144.9, 138.0, 137.2, 136.3, 128.9, 126.9, 116.0, 114.1, 92.9, 46.57. HRMS (+pESI) m/z calcd for $C_{13}H_{12}IN_2O_2$ [$M + H^+$], 354.9943; Found 354.9946

1-Benzyl-2-(4-methylphenyl)-1H-benzimidazole (Entry 2, Table 5)

White solid (262 mg, 88%); mp = 266-268 °C; IR (Neat) ν : 3033, 2947, 1611, 1462, 1451, 1362, 1253, 1163 cm^{-1} ; 1H NMR (DMSO, 400 MHz) δ : 7.85 (d, J = 8.0 Hz, 1H), 7.57 (d, J = 8.04 Hz, 2H), 7.34-7.18 (m, 8H), 7.10 (d, J = 6.96 Hz, 2H), 5.44 (s, 2H), 2.40 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ : 154.3, 143.2, 140.0, 136.5, 136.1, 129.4, 129.1, 129.0, 127.7, 127.1, 125.9, 122.8, 122.5, 119.9, 110.4, 48.3, 21.4.¹⁸

1-Benzyl-2-(3,5-dimethoxyphenyl)-1H-benzimidazole (Entry 3, Table 5)

White solid (295 mg, 86%); mp = 119-121 °C; IR (Neat) ν : 2969, 1635, 1603, 1454, 1386, 1227, 1205, 1156, 1063 cm^{-1} ; ^1H NMR (DMSO, 400 MHz) δ : 7.89-7.87 (m, 1H), 7.34-7.28 (m, 4H), 7.27-7.23 (m, 2H), 7.10 (d, J = 6.84 Hz, 2H), 6.80 (d, J = 2.32 Hz, 2H), 6.54 (t, J = 2.28 Hz, 1H), 5.46 (s, 2H), 3.66 (s, 6H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 160.8, 154.0, 142.9, 136.6, 136.2, 131.7, 129.1, 127.8, 125.9, 123.2, 122.8, 120.0, 110.4, 106.9, 102.9, 55.4, 48.4. HRMS (+pESI) m/z calcd for $\text{C}_{22}\text{H}_{21}\text{N}_2\text{O}_2$ [$\text{M} + \text{H}^+$], 345.1603; Found 345.1603

1-Benzyl-2-(2,3-dimethoxyphenyl)-1H-benzimidazole (Entry 4, Table 5)

White solid (292 mg, 85%); mp = 162-164 °C; IR (Neat) ν : 2970, 1636, 1455, 1365, 1229, 1217, 1129, 1050 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.86-7.84 (m, 1H), 7.29-7.24 (m, 1H), 7.19-7.17 (m, 5H), 7.14-7.11 (m, 2H), 7.05-7.03 (m, 1H), 6.97-6.95 (m, 2H), 5.30 (s, 2H), 3.89 (s, 3H), 3.65 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 152.7, 151.7, 147.4, 143.3, 136.3, 135.1, 128.6, 127.5, 126.7, 125.2, 124.6, 123.7, 122.7, 122.2, 119.9, 114.1, 110.8, 61.5, 55.9, 48.4. HRMS (+pESI) m/z calcd for $\text{C}_{22}\text{H}_{21}\text{N}_2\text{O}_2$ [$\text{M} + \text{H}^+$], 345.1603; Found 345.1603.

5-(1-Benzyl-1H-benzimidazol-2-yl)-2-methoxyphenol (Entry 5, Table 5)

White solid (287 mg, 87%); mp = 160-162 °C; IR (Neat) ν : 3440, 1635, 1403, 1241, 1157, 1056 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.84 (d, J = 7.96 Hz, 1H), 7.50 (d, J = 2.08 Hz, 1H), 7.30-7.24 (m, 4H), 7.17-7.14 (m, 2H), 7.07-7.03 (m, 3H), 6.82 (d, J = 8.36 Hz, 1H), 5.42 (s, 2H), 3.84 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 154.3, 149.1, 146.7, 142.4, 136.3, 135.6, 129.0, 127.7, 126.0, 122.9, 122.7, 122.0, 120.9, 119.5, 116.6, 110.9, 110.5, 55.8, 48.3. HRMS (+pESI) m/z calcd for $\text{C}_{21}\text{H}_{19}\text{N}_2\text{O}_2$ [$\text{M} + \text{H}^+$], 331.1447; Found 331.1447.

1-Benzyl-2-(4-(benzyloxy)phenyl)-1H-benzimidazole (Entry 6, Table 5)

White solid (343 mg, 88%); mp = 167-169 °C; IR (Neat) ν : 3033, 2971, 1610, 1481, 1453, 1420, 1385, 1277, 1251, 1176 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.85 (d, J = 7.88 Hz, 1H), 7.61 (d,

$J = 8.48$ Hz, 2H), 7.42-7.35 (m, 4H), 7.33-7.23 (m, 5H), 7.21-7.15 (m, 2H), 7.09 (d, $J = 6.64$ Hz, 2H), 7.01 (d, $J = 8.48$ Hz, 2H), 5.41 (s, 2H), 5.08 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 160.1, 154.1, 143.1, 136.5, 136.4, 136.1, 130.7, 129.0, 128.6, 128.5, 128.1, 127.7, 127.5, 125.9, 122.8, 122.6, 119.7, 115.1, 110.4, 70.0, 48.3. HRMS (+pESI) m/z calcd for $\text{C}_{27}\text{H}_{23}\text{N}_2\text{O}$ [$\text{M} + \text{H}^+$], 391.1810; Found 391.1810.

1-Benzyl-2-(4-chlorophenyl)-1H-benzimidazole (Entry 7, Table 5)

(286 mg, 90%); IR (Neat) ν : 2990, 1634, 1472, 1384, 1275, 1015 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.87-7.85 (m, 1H), 7.63-7.60 (m, 2H), 7.44-7.40 (m, 2H), 7.38-7.30 (m, 4H), 7.29-7.23 (m, 2H), 7.21-7.10 (m, 2H), 5.43 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 152.9, 143.0, 136.2, 136.1, 136.1, 130.5, 129.1, 129.0, 128.5, 127.9, 125.8, 123.3, 122.8, 120.0, 110.5, 48.3, 29.7.¹⁸

1-(4-Fluorobenzyl)-2-phenyl-1H-benzimidazole (Entry 8, Table 5)

Brown liquid (265 mg, 88%); IR (Neat) ν : 3021, 2939, 2868, 1616, 1465, 1425, 1362, 1255, 1160, 1085 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.91-7.88 (m, 1H), 7.71-7.68 (m, 2H), 7.53-7.46 (m, 2H), 7.37-7.33 (m, 2H), 7.30-7.28 (m, 2H), 7.26-7.22 (m, 2H), 7.11-7.09 (m, 1H), 7.08-7.06 (m, 1H), 7.05-7.02 (m, 1H), 5.45 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 163.5, 161.0, 154.1, 143.2, 135.9, 132.1, 132.0, 130.0, 129.9, 129.2, 128.9, 128.8, 128.5, 127.7, 127.6, 126.7, 123.2, 122.8, 120.1, 116.2, 115.9, 110.4, 47.8; HRMS (+pESI) m/z calcd for $\text{C}_{20}\text{H}_{15}\text{FN}_2$ [$\text{M} + \text{H}^+$], 302.1219; Found 302.1222.

1-(3-Methoxybenzyl)-2-phenyl-1H-benzimidazole (Entry 9, Table 5)

Off white solid (270 mg, 86%); mp = 125-127 °C; IR (Neat) ν : 3025, 2920, 2859, 1615, 1470, 1452, 1398, 1259, 1162, 1095 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.76 (d, $J = 8.80$ Hz, 1H), 7.68-7.66 (m, 2H), 7.48-7.42 (m, 3H), 7.38-7.29 (m, 3H), 7.12 (d, $J = 6.12$ Hz, 2H), 6.94 (dd, $J = 2.40$ & 8.76 Hz, 1H), 6.67 (d, $J = 2.32$ Hz, 1H), 5.42 (s, 2H), 3.79 (s, 3H); ^{13}C NMR (CDCl_3 , 100

MHz) δ : 156.8, 153.5, 137.7, 136.8, 136.3, 130.2, 129.6, 129.1, 129.0, 128.7, 127.8, 125.9, 120.5, 111.76, 94.3, 55.8, 48.4; HRMS (+pESI) m/z calcd for $C_{21}H_{18}N_2NaO$ [$M + Na^+$], 337.1317; Found 337.1314.

1-(4-Iodobenzyl)-2-phenyl-1H-benzimidazole (Entry 10, Table 5)

Brown solid (356 mg, 87%); mp = 180-182 °C; IR (Neat) ν : 2990, 2870, 1615, 1480, 1369, 1280, 1156 cm^{-1} ; 1H NMR (DMSO, 400 MHz) δ : 7.86 (d, J = 8.0 Hz, 1H), 7.65-7.62 (m, 4H), 7.47-7.41 (m, 3H), 7.32-7.28 (m, 1H), 7.25-7.21 (m, 1H), 7.16 (d, J = 7.92 Hz, 1H), 6.82 (d, J = 8.36 Hz, 1H), 5.36 (s, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ : 154.1, 143.2, 138.2, 136.1, 135.8, 130.1, 129.9, 129.2, 128.9, 128.8, 127.9, 123.3, 122.9, 120.1, 110.4, 47.9; HRMS (+pESI) m/z calcd for $C_{20}H_{16}IN_2$ [$M + H^+$], 411.0358; Found 411.0359.

1-Benzyl-5-methyl-2-phenyl-1H-benzimidazole (Entry 11, Table 5)

White solid (268 mg, 90%); mp = 135-137 °C; IR (KBr) ν_{max} = 3250, 1645, 1610, 1479, 1289 cm^{-1} ; 1H NMR (400 MHz, $CDCl_3$): δ = 7.69 (m, 3H), 7.47 (m, 3 H), 7.32 (m, 3 H), 7.09 (m, 4 H), 5.44 (s, 2 H), 2.51 (s, 3 H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 154.1, 143.5, 136.5, 134.1, 132.4, 130.2, 129.8, 129.2, 129.0, 128.7, 127.7, 125.9, 124.5, 119.7, 110.0, 48.40, 21.60; MS (+pAPCI) m/z : 299.5 ($M+H^+$).¹⁹

1-Benzyl-5-methyl-2-(4-methylphenyl)-1H-benzimidazole (Entry 12, Table 5)

White solid (280 mg, 90%); mp = 183-185 °C; IR (Neat) ν : 3030, 2920, 2853, 1615, 1478, 1452, 1384, 1354, 1324, 1259, 1169 cm^{-1} ; 1H NMR (DMSO, 400 MHz) δ : 7.64 (s, 1H), 7.55 (d, J = 7.68 Hz, 2H), 7.30-7.26 (m, 3H), 7.23-7.22 (m, 2H), 7.08-7.01 (s, 4H), 5.39 (s, 2H), 2.47 (s, 3H), 2.38 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ : 154.2, 143.5, 139.9, 136.6, 134.2, 132.2, 129.4, 129.1, 129.0, 127.6, 127.2, 125.9, 124.3, 119.6, 109.9, 48.3, 21.6, 21.4. HRMS (+pESI) m/z calcd for $C_{22}H_{21}N_2$ [$M + H^+$], 313.1705; Found 313.1697.¹⁸

1-Benzyl-2-(4-(benzyloxy)phenyl)-5-methyl-1H-benzimidazole (Entry 13, Table 5)

White solid (363 mg, 90%); mp = 187-188 °C; IR (Neat) ν : 3032, 2972, 1611, 1488, 1459, 1428, 1395, 1281, 1258, 1170 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.61-7.59 (m, 3H), 7.43-7.25 (m, 8H), 7.10-7.01 (m, 6H), 5.45 (s, 2H), 5.09 (s, 2H), 2.48 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 159.9, 154.0, 143.4, 136.6, 136.4, 134.1, 132.2, 130.6, 129.0, 128.6, 128.1, 127.6, 127.4, 125.9, 124.2, 122.7, 119.5, 115.0, 109.8, 70.0, 48.3, 21.6. HRMS (+pESI) m/z calcd for $\text{C}_{28}\text{H}_{25}\text{N}_2\text{O}$ [$\text{M} + \text{H}^+$], 405.1967; Found 405.1967.

1-Benzyl-2-(4-chlorophenyl)-5-methyl-1H-benzimidazole (Entry 14, Table 5)

White solid (298 mg, 90%); mp = 138-139 °C; IR (Neat) ν : 3030, 2970, 1634, 1601, 1471, 1378, 1228, 1093 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.63-7.58 (m, 3H), 7.40-7.38 (m, 2H), 7.34-7.24 (m, 3H), 7.09-7.05 (m, 4H), 5.38 (s, 2H), 2.48 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 152.8, 143.3, 136.2, 136.1, 134.2, 132.6, 130.4, 129.1, 129.0, 128.6, 127.8, 125.8, 124.8, 119.7, 110.0, 48.3, 21.6. HRMS (+pESI) m/z calcd for $\text{C}_{21}\text{H}_{18}\text{ClN}_2$ [$\text{M} + \text{H}^+$], 333.1159; Found 333.1153

1-Benzyl-5-chloro-2-phenyl-1H-benzimidazole (Entry 15, Table 5)

White solid (268 mg, 88%); mp = 179-181 °C; IR (KBr) ν_{max} = 3242, 1645, 1601, 1493, 1270 cm^{-1} ; ^1H NMR (400 MHz, CDCl_3): δ = 7.85 (d, J = 1.92 Hz, 1 H), 7.69 (m, 2 H), 7.48 (m, 3 H), 7.35 (m, 3 H), 7.21 (dd, J = 1.96 Hz & 8.6 Hz, 1 H), 7.12 (d, J = 8.80 Hz, 1 H), 7.09 (m, 2 H), 5.46 (s, 2 H); ^{13}C NMR (100 MHz, CDCl_3): δ = 155.4, 144.0, 135.9, 134.6, 130.2, 129.7, 129.2, 129.1, 128.8, 128.3, 128.0, 125.9, 123.4, 119.8, 111.3, 48.5; MS (+pAPCI) m/z : 319.5 ($\text{M} + \text{H}^+$).¹⁹

1-Benzyl-5-chloro-2-(4-methylphenyl)-1H-benzimidazole (Entry 16, Table 5)

White solid (308 mg, 87%); mp = 127-129 °C; IR (Neat) ν : 2995, 2913, 1631, 1609, 1454, 1432, 1382, 1381, 1246, 1057 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.81 (s, 1H), 7.55 (d, J = 7.72 Hz,

2H), 7.33-7.31 (m, 3H), 7.26-7.24 (m, 2H), 7.17-7.15 (m, 1H), 7.07-7.06 (m, 3H), 5.41 (s, 2H), 2.40 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 155.5, 143.9, 140.4, 136.0, 134.6, 129.5, 129.1, 128.8, 128.1, 127.9, 126.6, 125.9, 123.2, 119.6, 111.2, 48.5, 21.4. HRMS (+pESI) m/z calcd for $\text{C}_{21}\text{H}_{17}\text{ClN}_2\text{Na}$ [$\text{M} + \text{H}^+$], 355.0978; Found 355.0977.

1-Benzyl-2-(4-(benzyloxy)phenyl)-5-chloro-1H-benzimidazole (Entry 17, Table 5)

White solid (369 mg, 88%); mp = 173-175 °C; IR (Neat) ν : 2935, 2920, 1608, 1609, 1454, 1383, 1251, 1176 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.80 (s, 1H), 7.60 (d, J = 8.6 Hz, 2H), 7.43-7.40 (m, 2H), 7.39-7.34 (m, 3H), 7.32-7.30 (m, 3H), 7.18-7.15 (m, 1H), 7.08-7.06 (m, 3H), 7.04-7.02 (m, 2H), 5.42 (s, 2H), 5.10 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 160.3, 155.3, 144.0, 136.3, 136.0, 134.6, 130.7, 129.1, 128.8, 128.6, 128.1, 127.9, 127.4, 125.8, 123.1, 122.5, 119.5, 115.2, 111.5, 70.0, 48.5. HRMS (+pESI) m/z calcd for $\text{C}_{27}\text{H}_{22}\text{ClN}_2\text{O}$ [$\text{M} + \text{H}^+$], 425.1421; Found 425.1420.

1-Benzyl-5-chloro-2-(4-chlorophenyl)-1H-benzimidazole (Entry 18, Table 5)

White solid (313 mg, 89%); IR (Neat) ν : 3028, 2968, 1620, 1600, 1461, 1388, 1218, 1094 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.82 (d, J = 1.72 Hz, 1H), 7.63-7.58 (m, 2H), 7.45-7.41 (m, 2H), 7.37-7.30 (m, 3H), 7.21 (dd, J = 1.92 & 8.60 Hz, 1H), 7.10-7.08 (m, 3H), 7.07-7.04 (m, 2H), 5.41 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 154.1, 143.8, 136.5, 135.7, 134.7, 130.4, 129.2, 129.1, 128.4, 128.1, 128.0, 125.7, 123.7, 119.8, 111.3, 48.5. HRMS (+pESI) m/z calcd for $\text{C}_{20}\text{H}_{15}\text{Cl}_2\text{N}_2$ [$\text{M} + \text{H}^+$], 353.0612; Found 353.0607.

1-Benzyl-6-methyl-2-phenyl-1H-benzimidazole (Entry 19, Table 5)

White solid (259 mg, 90%); mp = 195-197 °C; IR (KBr) ν_{max} = 3260, 1649, 1616, 1486, 1277 cm^{-1} ; ^1H NMR (400 MHz, CDCl_3): δ = 7.74 (d, J = 8.24 Hz, 1 H), 7.66 (m, 2 H), 7.43 (m, 3 H), 7.34 (m, 3 H), 7.12 (m, 3 H), 7.00 (s, 1 H), 5.42 (s, 2 H), 2.44 (s, 3 H); ^{13}C NMR (100 MHz,

CDCl_3): δ = 153.7, 141.3, 136.6, 136.4, 133.1, 130.2, 129.7, 129.2, 129.0, 128.7, 127.7, 125.9, 124.3, 119.9, 110.3, 48.24, 21.8; MS (+pAPCI) m/z : 299.4 ($\text{M}+\text{H}^+$).²⁰

1-Benzyl-6-methyl-2-(4-methylphenyl)-1H-benzimidazole (Entry 20, Table 5)

White solid (274 mg, 88%); mp = 193-195 °C; IR (Neat) ν : 3031, 2957, 1614, 1452, 1441, 1372, 1263, 1143 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.73 (d, J = 8.20 Hz, 1H), 7.56-7.54 (m, 2H), 7.36-7.29 (m, 3H), 7.27-7.22 (m, 2H), 7.13-7.10 (m, 3H), 6.98 (s, 1H), 5.40 (s, 2H), 2.42 (s, 3H), 2.32 (s, 1H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 153.9, 141.2, 139.8, 136.6, 136.3, 132.9, 129.4, 129.1, 129.0, 127.6, 127.2, 125.9, 124.1, 119.3, 110.2, 48.2, 21.8, 21.4. HRMS (+pESI) m/z calcd for $\text{C}_{22}\text{H}_{21}\text{N}_2$ [$\text{M} + \text{H}^+$], 313.1705; Found 313.1697.

1-Benzyl-2-(4-(benzyloxy)phenyl)-6-methyl-1H-benzimidazole (Entry 21, Table 5)

White solid (359 mg, 89%); mp = 214-216 °C; IR (Neat) ν : 3030, 2982, 1616, 1495, 1455, 1438, 1385, 1271, 1268, 1172 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.71 (d, J = 8.20 Hz, 1H), 7.61-7.57 (m, 2H), 7.43-7.35 (m, 4H), 7.34-7.28 (m, 4H), 7.12-7.10 (m, 3H), 7.03-6.99 (m, 2H), 6.97 (s, 1H), 5.40 (s, 2H), 5.09 (s, 2H), 2.42 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 159.9, 153.6, 141.3, 136.6, 136.5, 136.4, 132.8, 130.6, 129.0, 128.6, 128.1, 127.6, 127.4, 125.8, 124.1, 122.8, 119.2, 115.0, 110.1, 70.0, 48.2, 21.8. HRMS (+pESI) m/z calcd for $\text{C}_{28}\text{H}_{25}\text{N}_2\text{O}$ [$\text{M} + \text{H}^+$], 405.1967; Found 405.1967.

1-benzyl-2-(4-chlorophenyl)-6-methyl-1H-benzimidazole (Entry 22, Table 5)

White solid (285 mg, 86%); mp = 169-172 °C; IR (Neat) ν : 3031, 2975, 1624, 1475, 1378, 1238, 1125 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.73 (d, J = 8.20 Hz, 1H), 7.60-7.55 (m, 2H), 7.41-7.36 (m, 2H), 7.35-7.29 (m, 3H), 7.14 (d, J = 8.24 Hz, 1H), 7.09 (d, J = 7.08 Hz, 1H), 7.01 (m, 1H), 5.39 (s, 2H), 2.44 (s, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 141.2, 136.4, 136.3, 136.0,

133.4, 130.4, 129.1, 129.0, 128.6, 127.8, 125.7, 124.5, 119.5, 110.2, 48.2, 21.8. HRMS (+pESI) m/z calcd for $C_{21}H_{18}ClN_2$ $[M + H^+]$, 333.1159; Found 333.1153.

1-Benzyl-6-chloro-2-phenyl-1H-benzimidazole (Entry 23, Table 5)

White Solid (283 mg, 89%); mp = 230-232 °C; IR (KBr) $\nu_{\max}$ = 3245, 1640, 1605, 1485, 1273 cm^{-1} ; 1H NMR (400 MHz, $CDCl_3$): δ = 7.77 (d, J = 8.56 Hz, 1 H), 7.67 (m, 2 H), 7.46 (m, 3 H), 7.35 (m, 3 H), 7.28 (m, 1 H), 7.20 (d, J = 1.84 Hz, 1 H), 7.09 (m, 2 H), 5.42 (s, 2 H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 155.0, 141.8, 136.7, 135.8, 130.2, 129.6, 129.2, 128.8, 128.7, 128.0, 125.9, 123.4, 120.8, 110.5, 48.5; MS (+pAPCI) m/z : 319.8 ($M+H^+$).²¹

1-Benzyl-6-chloro-2-(4-methylphenyl)-1H-benzimidazole (Entry 24, Table 5)

White solid (308 mg, 87%); mp = 146-148 °C; IR (Neat) ν : 2995, 1638, 1609, 1457, 1381, 1245 cm^{-1} ; 1H NMR ($CDCl_3$, 400 MHz) δ : 7.74 (s, 1H), 7.56-7.54 (m, 2H), 7.37-7.29 (m, 3H), 7.26-7.24 (m, 3H), 7.18-7.17 (m, 1H), 7.09-7.07 (m, 2H), 5.40 (s, 2H), 2.40 (s, 3H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ : 155.1, 141.8, 140.3, 136.7, 135.9, 129.5, 129.1, 129.0, 128.5, 127.9, 126.6, 125.8, 123.3, 120.7, 110.4, 48.4, 21.42. HRMS (+pESI) m/z calcd for $C_{21}H_{17}ClN_2Na$ $[M + H^+]$, 355.0978; Found 355.0977.

1-Benzyl-2-(4-(benzyloxy)phenyl)-6-chloro-1H-benzimidazole (Entry 25, Table 5)

White solid (374 mg, 88%); mp = 138-141 °C; IR (Neat) ν : 2998, 1636, 1625, 1455, 1365, 1217, 1157 cm^{-1} ; 1H NMR ($CDCl_3$, 400 MHz) δ : 7.74 (d, J = 8.56 Hz, 1H), 7.62-7.58 (m, 2H), 7.44-7.40 (m, 2H), 7.38-7.37 (m, 3H), 7.36-7.33 (m, 3H), 7.26-7.24 (m, 1H), 7.16 (d, J = 1.84 Hz, 1H), 7.10-7.08 (m, 2H), 7.04-7.02 (m, 2H), 5.40 (s, 2H), 5.10 (s, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz) δ : 160.2, 154.9, 141.7, 136.7, 136.3, 135.8, 130.6, 129.2, 128.6, 128.4, 128.1, 127.9, 127.4, 125.8, 123.3, 122.0, 120.5, 115.1, 110.4, 70.0, 48.4. HRMS (+pESI) m/z calcd for $C_{27}H_{22}ClN_2O$ $[M + H^+]$, 425.1421; Found 425.1420.

1-Benzyl-6-chloro-2-(4-chlorophenyl)-1H-benzimidazole (Entry 26, Table 5)

White solid (313 mg, 89%); mp = 146-148 °C; IR (Neat) ν : 3030, 2972, 1616, 1465, 1391, 1226, 1119 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.75 (d, J = 8.60 Hz, 1H), 7.61-7.59 (m, 2H), 7.44-7.41 (m, 2H), 7.38-7.32 (m, 3H), 7.29-7.27 (m, 1H), 7.18 (d, J = 1.88 Hz, 1H), 7.07-7.06 (m, 2H), 5.35 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 153.7, 141.7, 136.7, 136.4, 135.6, 130.4, 129.3, 129.1, 129.0, 128.1, 128.0, 125.7, 123.6, 120.6, 110.5, 48.4. HRMS (+pESI) m/z calcd for $\text{C}_{20}\text{H}_{15}\text{Cl}_2\text{N}_2$ [$\text{M} + \text{H}^+$], 353.0612; Found 353.0607.

1-Benzyl-2-(1H-indol-3-yl)-1H-benzimidazole (Entry 27, Table 5)

White solid (271 mg, 84%); mp = 232-234 °C; IR (Neat) ν : 3340, 3025, 2985, 2855, 1614, 1602, 1495, 1460, 1350, 1268, 1265, 1155 cm^{-1} ; ^1H NMR (DMSO, 400 MHz) δ : 11.64 (s, 1 H), 8.36 (d, J = 7.40 Hz, 1H), 7.71 (d, J = 7.04 Hz, 1H), 7.66 (s, 1H), 7.47-7.43 (m, 2H), 7.29-7.27 (m, 2H), 7.22-7.17 (m, 5H), 7.06-7.05 (m, 2H), 5.72 (s, 2H); ^{13}C NMR, DMSO, 100 MHz) δ : 149.6, 143.1, 137.1, 135.9, 135.5, 128.8, 127.3, 126.3, 125.9, 125.8, 122.4, 121.7, 121.7, 121.4, 120.3, 118.4, 117.3, 110.0, 104.6, 46.97. HRMS (+pESI) m/z calcd for $\text{C}_{22}\text{H}_{17}\text{N}_3\text{K}$ [$\text{M} + \text{K}^+$], 362.1060; Found 362.1054.

1-Benzyl-2-(thiophen-3-yl)-1H-benzimidazole (Entry 28, Table 5)

White solid (246 mg, 85%); mp = 165-167 °C; IR (Neat) ν : 3030, 2965, 2875, 1616, 1485, 1368, 1288, 1256, 1155 cm^{-1} ; ^1H NMR (DMSO, 400 MHz) δ : 7.87 (d, J = 7.68 Hz, 1H), 7.65-7.60 (m, 1H), 7.54-7.51 (m, 1H), 7.42-7.40 (m, 1H), 7.36-7.18 (m, 6H), 7.16 (d, J = 7.08 Hz, 2H), 5.49 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 149.7, 143.0, 136.2, 135.9, 130.9, 129.2, 128.2, 127.8, 126.5, 126.3, 125.8, 123.0, 122.7, 119.8, 110.0, 48.1. HRMS (+pESI) m/z calcd for $\text{C}_{18}\text{H}_{15}\text{N}_2\text{S}$ [$\text{M} + \text{H}^+$], 291.0956; Found 291.0953.

1-Benzyl-2-cyclohexyl-1H-benzimidazole (Entry 29, Table 5)

White solid; (255 mg, 88%); IR (Neat) ν : 3034, 2931, 2853, 1614, 1497, 1454, 1418, 1354, 1281, 1250, 1155 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.80 (d, J = 7.88 Hz, 1H), 7.47-7.28 (m, 3H), 7.25-7.20 (m, 1H), 7.17-7.16 (m, 2H), 7.03 (d, J = 6.48 Hz, 2H), 5.36 (s, 2H), 2.82-2.76 (m, 1H), 1.85-1.82 (m, 6H), 1.72-1.71(m, 1H), 1.40-1.21 (m, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 159.3, 142.4, 136.2, 134.9, 128.8, 127.8, 126.0, 122.2, 122.0 119.2, 109.6, 46.7, 36.4, 31.8, 26.3, 25.7. HRMS (+pESI) m/z calcd for $\text{C}_{20}\text{H}_{23}\text{N}_2$ [$\text{M} + \text{H}^+$], 291.1861; Found 291.1862.

1-Benzyl-2-cyclohexyl-5-methyl-1H-benzimidazole (Entry 30, Table 5)

White solid (270 mg, 89%); mp = 144-145 $^{\circ}\text{C}$; IR (Neat) ν : 3035, 2975, 2865, 1616, 1495, 1464, 1360, 1288, 1255, 1158 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.57 (s, 1H), 7.29-7.25 (m, 3H), 7.05-6.98 (m, 4H), 5.32 (s, 2H), 2.77-2.76 (m, 1H), 2.44 (s, 1H), 1.90-1.84 (m, 6H), 1.84-1.71(m, 1H), 1.38-1.26 (m, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 159.2, 142.7, 136.3, 133.0, 131.66, 128.9, 127.7, 126.0, 123.6, 119.1, 109.1, 46.7, 36.5, 31.8, 26.3, 25.7, 21.5. HRMS (+pESI) m/z calcd for $\text{C}_{21}\text{H}_{25}\text{N}_2$ [$\text{M} + \text{H}^+$], 305.2018; Found 305.2017.

1-Benzyl-5-chloro-2-cyclohexyl-1H-benzimidazole (Entry 31, Table 5)

White solid (278 mg, 86%); mp = 139-141 $^{\circ}\text{C}$; IR (Neat) ν : 3030, 2978, 1618, 1469, 1398, 1235, 1084 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.74 (d, J = 1.84 Hz, 1H), 7.33-7.27 (m, 3H), 7.13-7.11 (m, 1H), 7.06-7.04 (m, 1H), 7.01-6.99 (m, 2H), 5.33 (s, 2H), 2.81-2.74 (m, 1H), 1.88-1.75 (m, 6H), 1.74-1.72 (m, 1H), 1.39-1.25 (m, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 160.6, 143.5, 135.7, 133.6, 129.0, 128.0, 127.5, 125.9, 122.5, 119.1, 110.3, 46.8, 36.4, 31.8, 26.2, 25.6. HRMS (+pESI) m/z calcd for $\text{C}_{20}\text{H}_{22}\text{ClN}_2$ [$\text{M} + \text{H}^+$], 325.1472; Found 325.1472.

1-Benzyl-2-cyclohexyl-6-methyl-1H-benzimidazole (Entry 32, Table 5)

White solid; (264 mg, 87%); IR (Neat) ν : 3033, 2985, 2855, 1618, 1498, 1454, 1368, 1278, 1259, 1159 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.65 (d, J = 8.16 Hz, 1H), 7.32-7.25 (m, 3H), 7.06-

7.02 (m, 3H), 6.97-6.96 (m, 1H), 5.32 (m, 2H), 2.79-2.71 (m, 1H), 2.41 (s, 3H), 1.87-1.71 (m, 7H), 1.38-1.25 (m, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 158.9, 140.7, 136.4, 135.3, 132.1, 128.9, 127.7, 126.0, 123.4, 118.8, 109.5, 46.5, 36.4, 31.9, 26.3, 25.7, 21.7. HRMS (+pESI) m/z calcd for $\text{C}_{21}\text{H}_{25}\text{N}_2$ [$\text{M} + \text{H}^+$], 305.2018; Found 305.2017.

1-Benzyl-6-chloro-2-cyclohexyl-1H-benzimidazole (Entry 33, Table 5)

White solid (285 mg, 88%); mp = 143-145 °C; IR (Neat) ν : 3028, 2988, 2869, 1615, 1479, 1388, 1255, 1154 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.67 (d, J = 10.28 Hz, 1H), 7.35-7.28 (m, 3H), 7.19-7.15 (m, 2H), 7.03-7.00 (m, 2H), 2.80-2.73 (m, 1H), 1.88-1.78 (m, 6H), 1.75-1.72 (m, 1H), 1.35-1.25 (m, 3H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 160.2, 141.3, 136.0, 135.7, 129.0, 128.0, 127.9, 125.9, 122.5, 120.1, 109.7, 46.8, 36.4, 31.8, 26.2, 25.6; HRMS (+pESI) m/z calcd for $\text{C}_{20}\text{H}_{22}\text{ClN}_2$ [$\text{M} + \text{H}^+$], 325.1472; Found 325.1472.

(1-Benzyl-1H-benzimidazol-2-yl)methanol (Entry 34, Table 5)

White solid (229 mg, 82%); mp = 184-185 °C; IR (Neat) ν : 3455, 3030, 2983, 2861, 1612, 1485, 1454, 1355, 1277, 1251, 1150 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ : 7.71 (d, J = 7.52 Hz, 1H), 7.26-7.23 (m, 4H), 7.22-7.20 (m, 2H), 7.09-7.08 (m, 2H), 5.45 (s, 2H), 4.86 (s, 2H); ^{13}C NMR (CDCl_3 , 100 MHz) δ : 153.9, 141.6, 135.7, 135.4, 128.9, 127.9, 126.4, 123.1, 122.4, 119.4, 110.0, 57.1, 47.2. HRMS (+pESI) m/z calcd for $\text{C}_{15}\text{H}_{15}\text{N}_2\text{O}$ [$\text{M} + \text{H}^+$], 239.1184; Found 239.1182.²²

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Scanned NMR spectra

Table 3, Entry 1: ^1H NMR

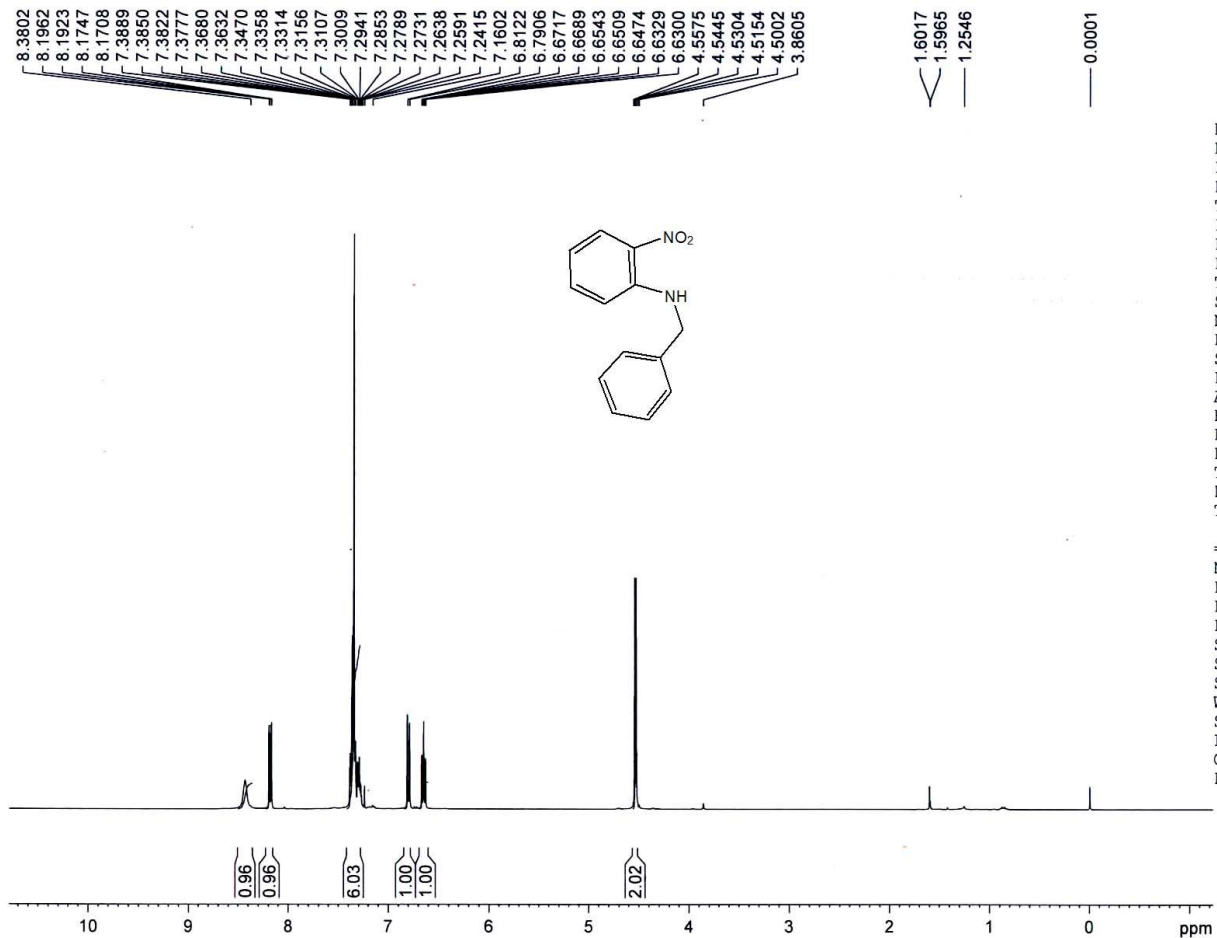


Table 3, Entry 2: ^1H NMR

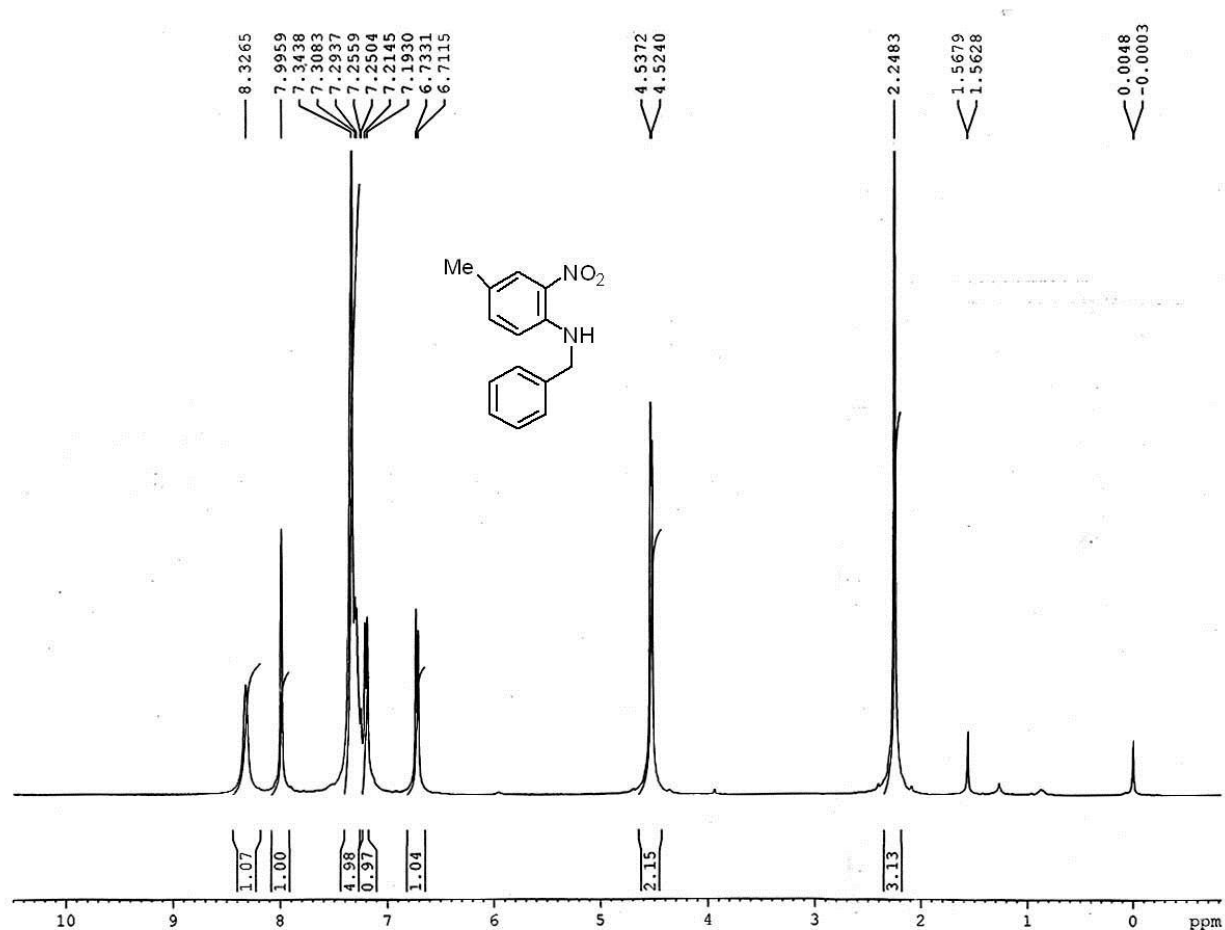


Table 3, Entry 3: ¹H NMR

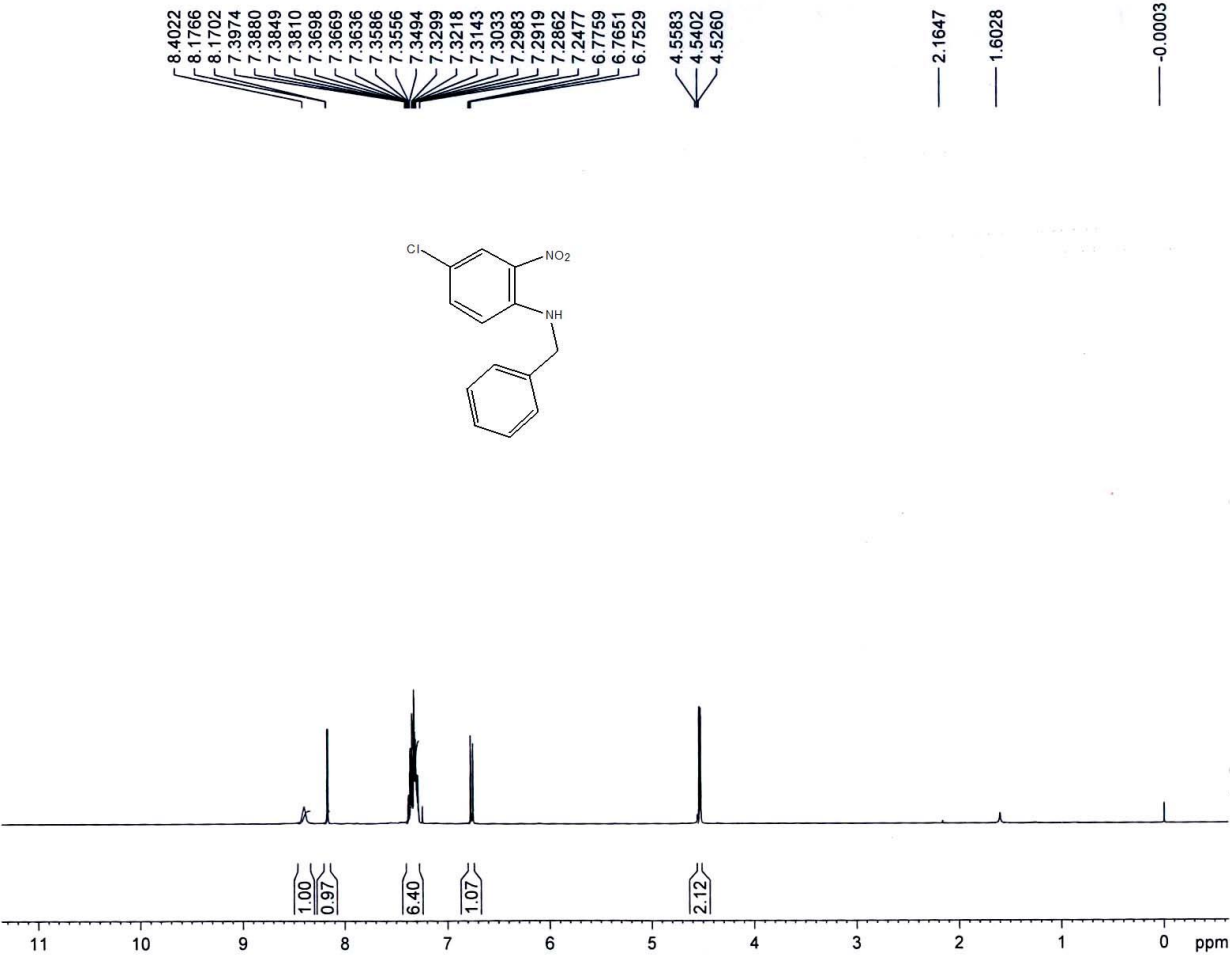


Table 3, Entry 4: ^1H NMR

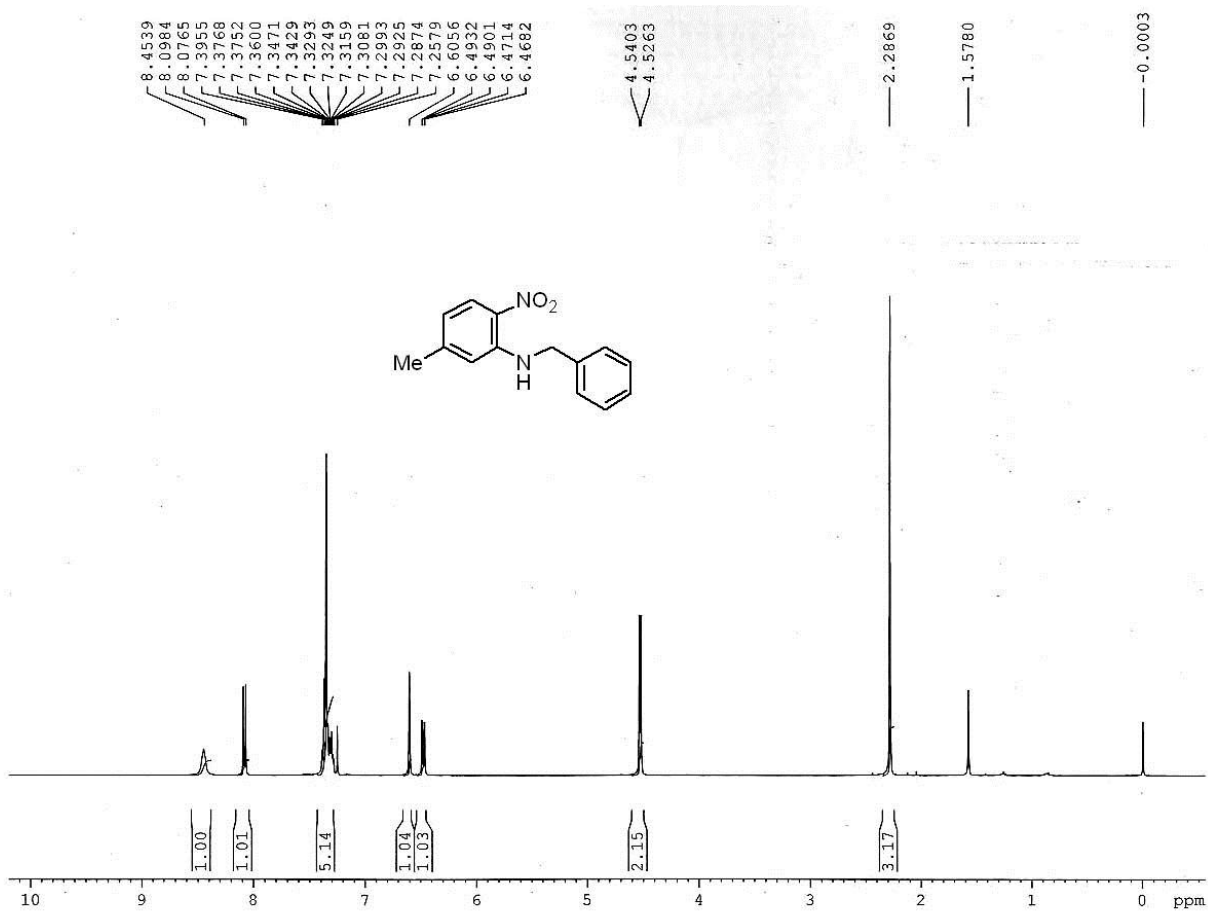


Table 3, Entry 5: ^1H NMR

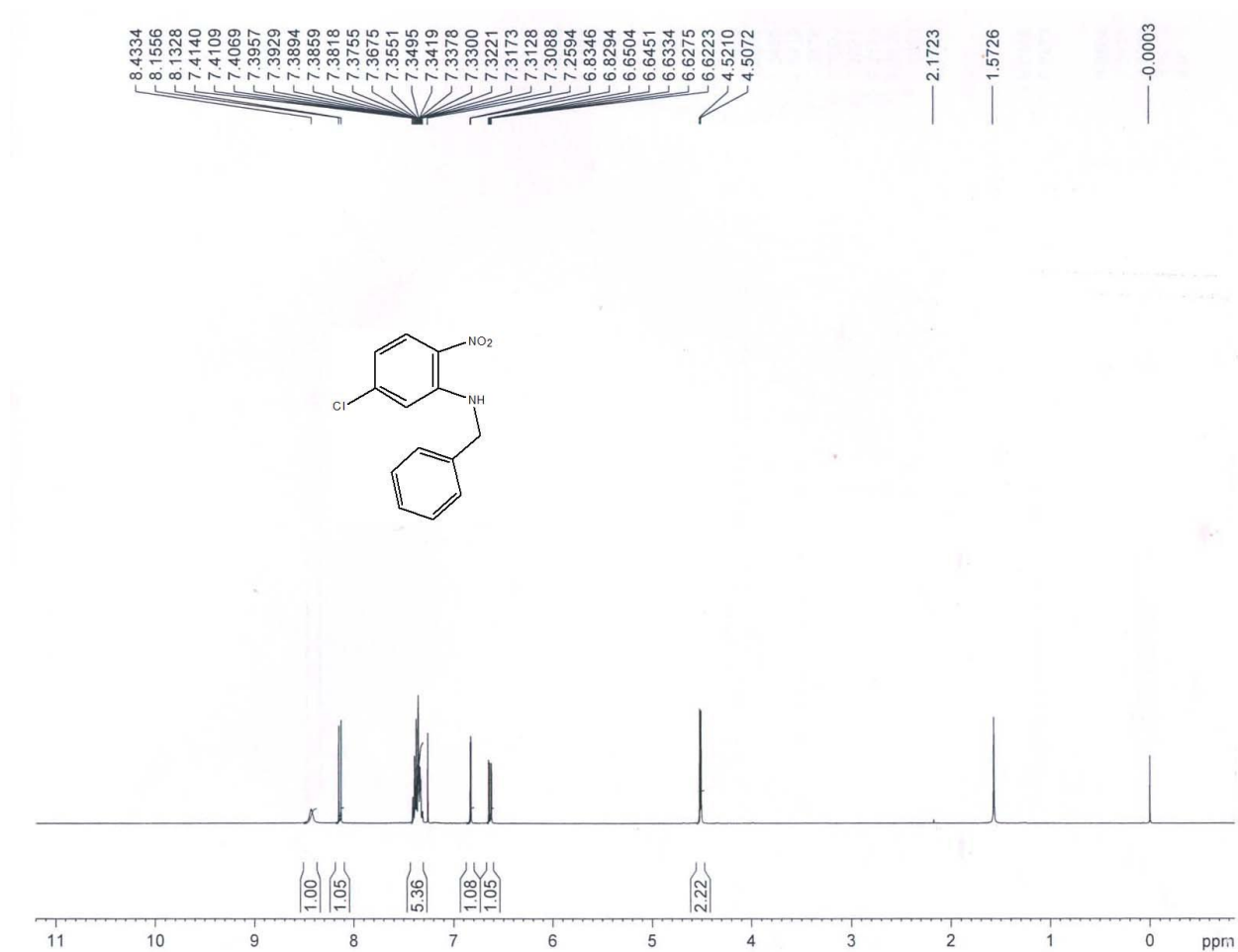


Table 3, Entry 6: ^1H NMR

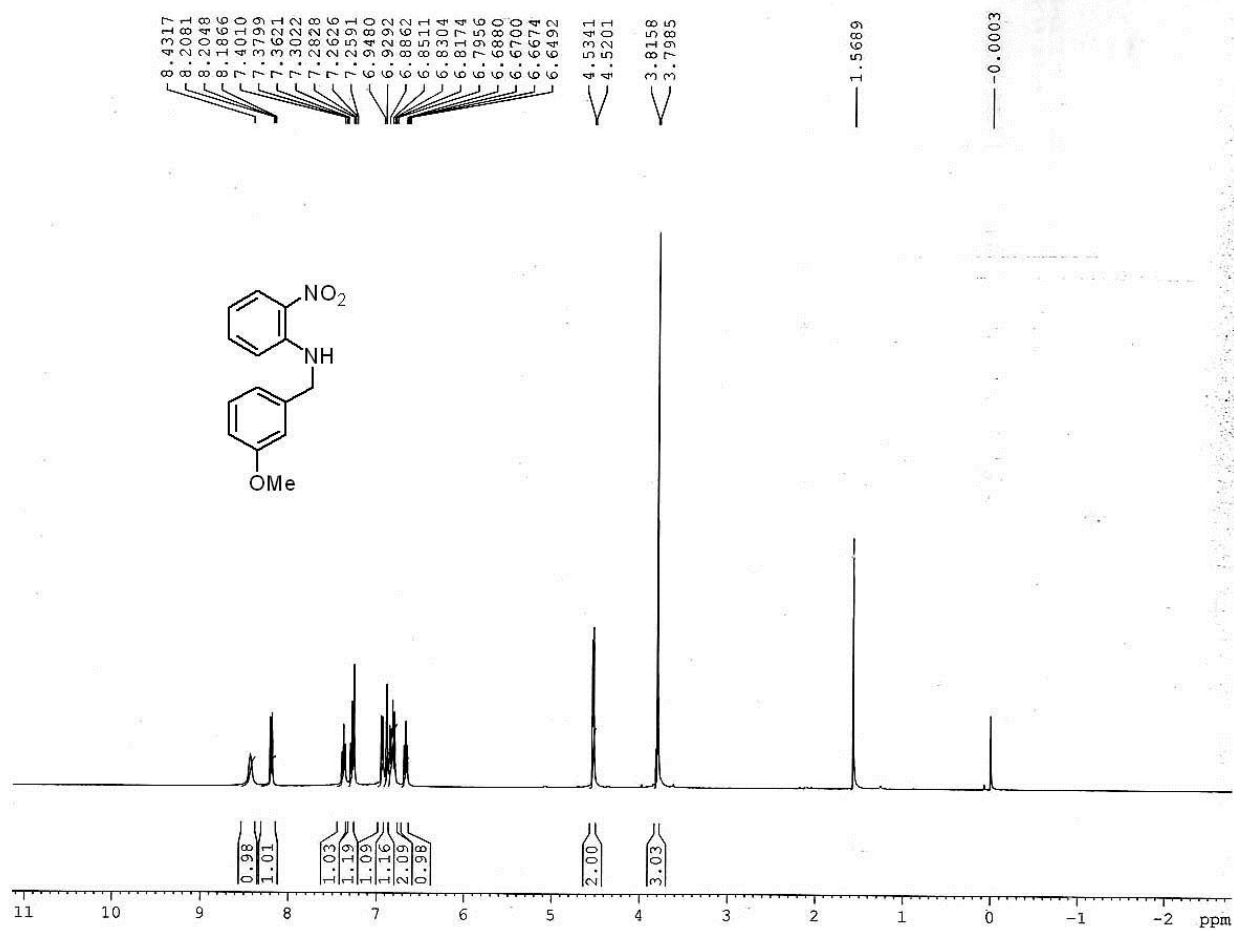


Table 3, Entry 6: ^{13}C NMR

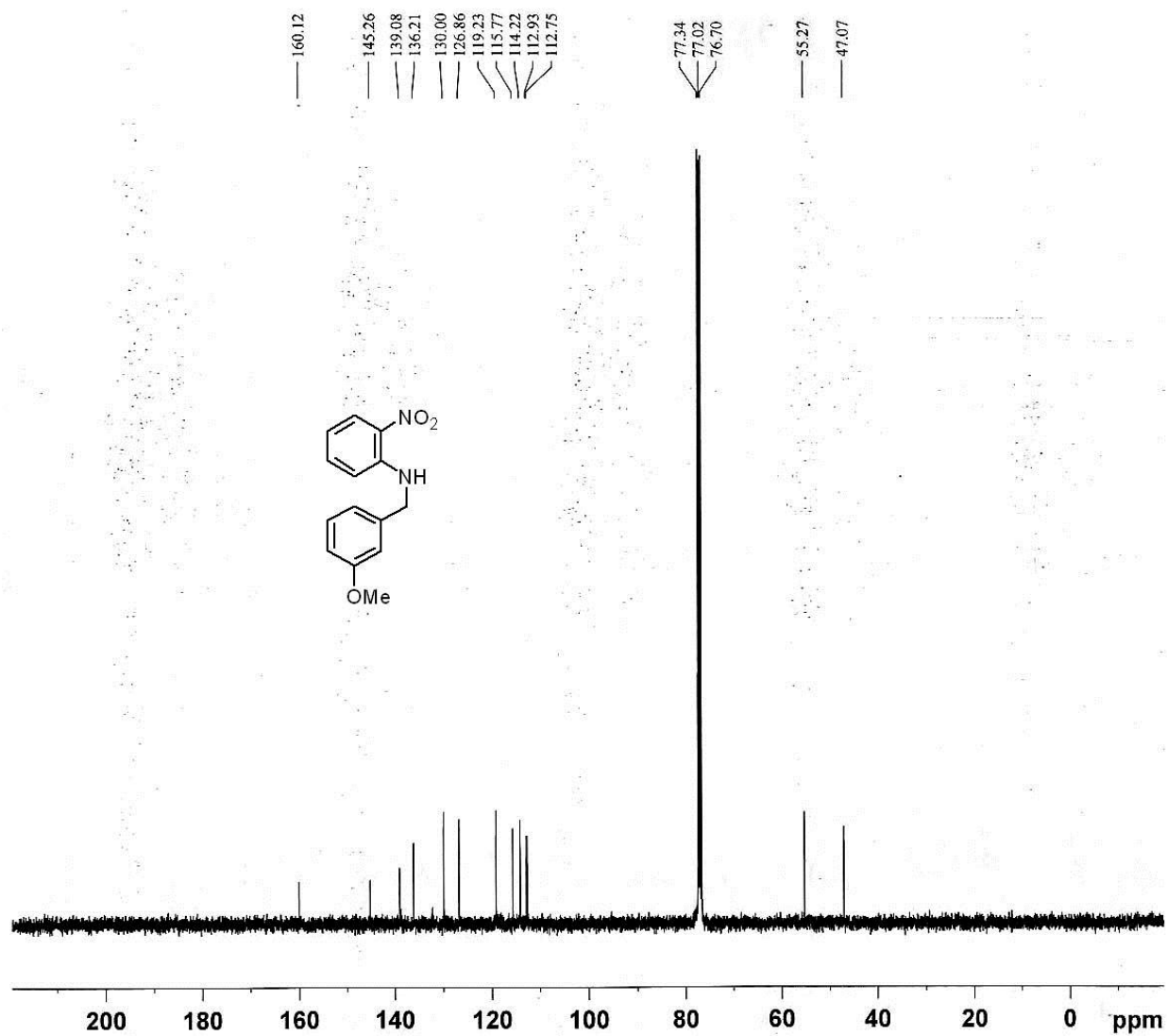


Table 3, Entry 7: ^1H NMR

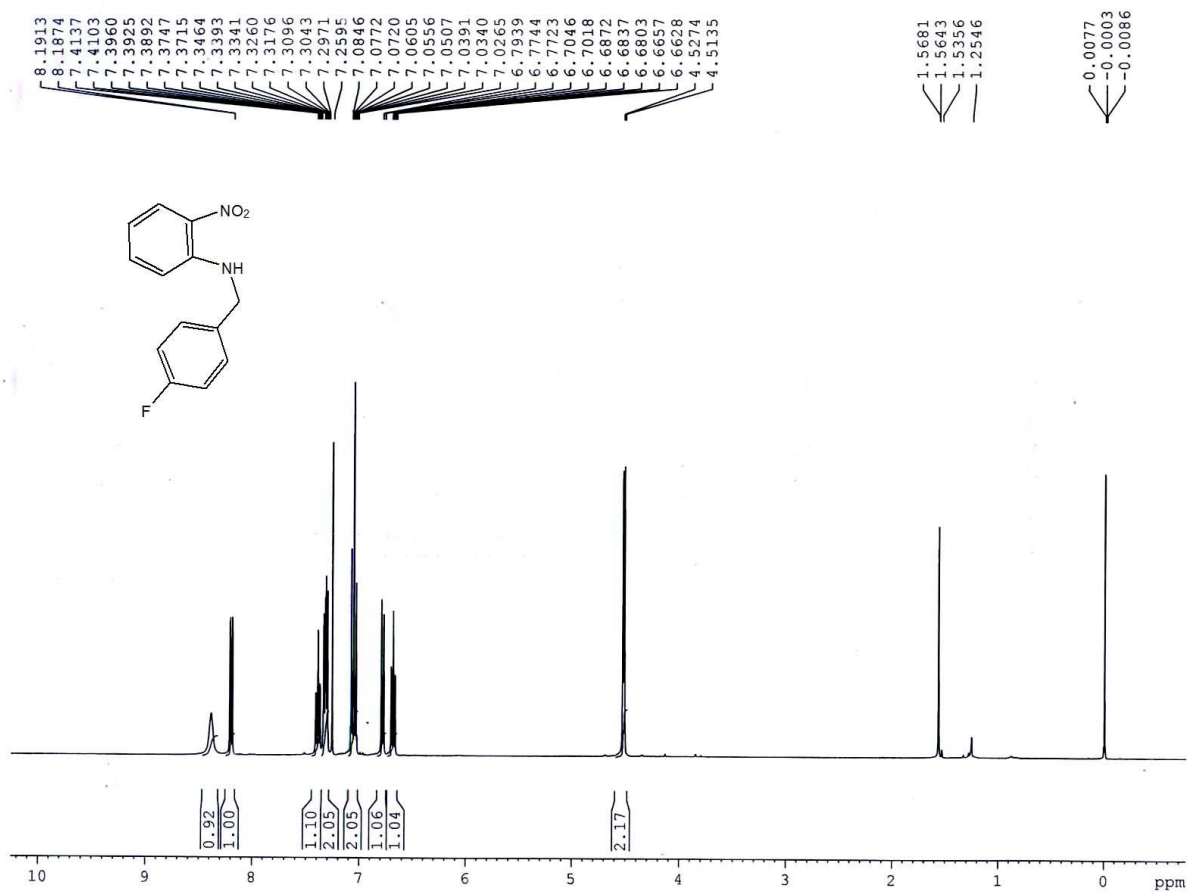


Table 3, Entry 8: ^1H NMR

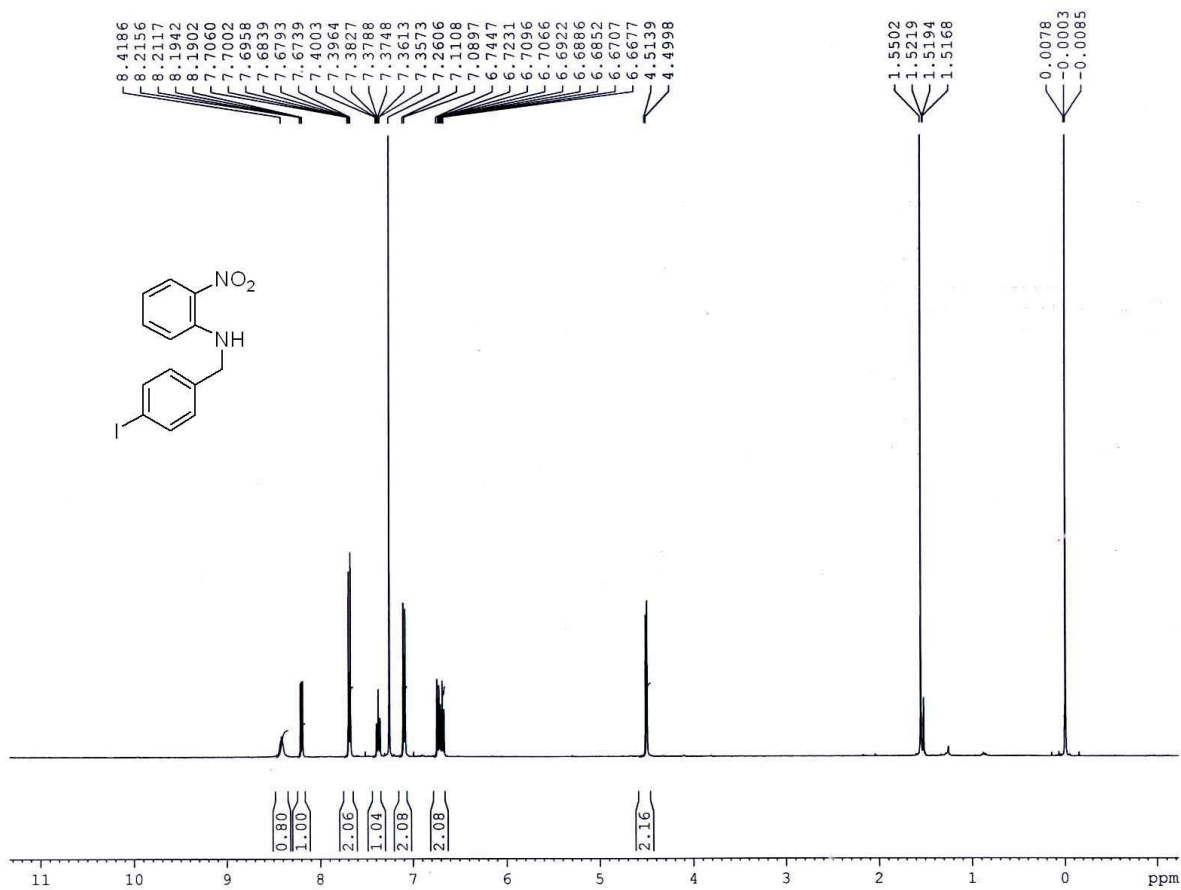
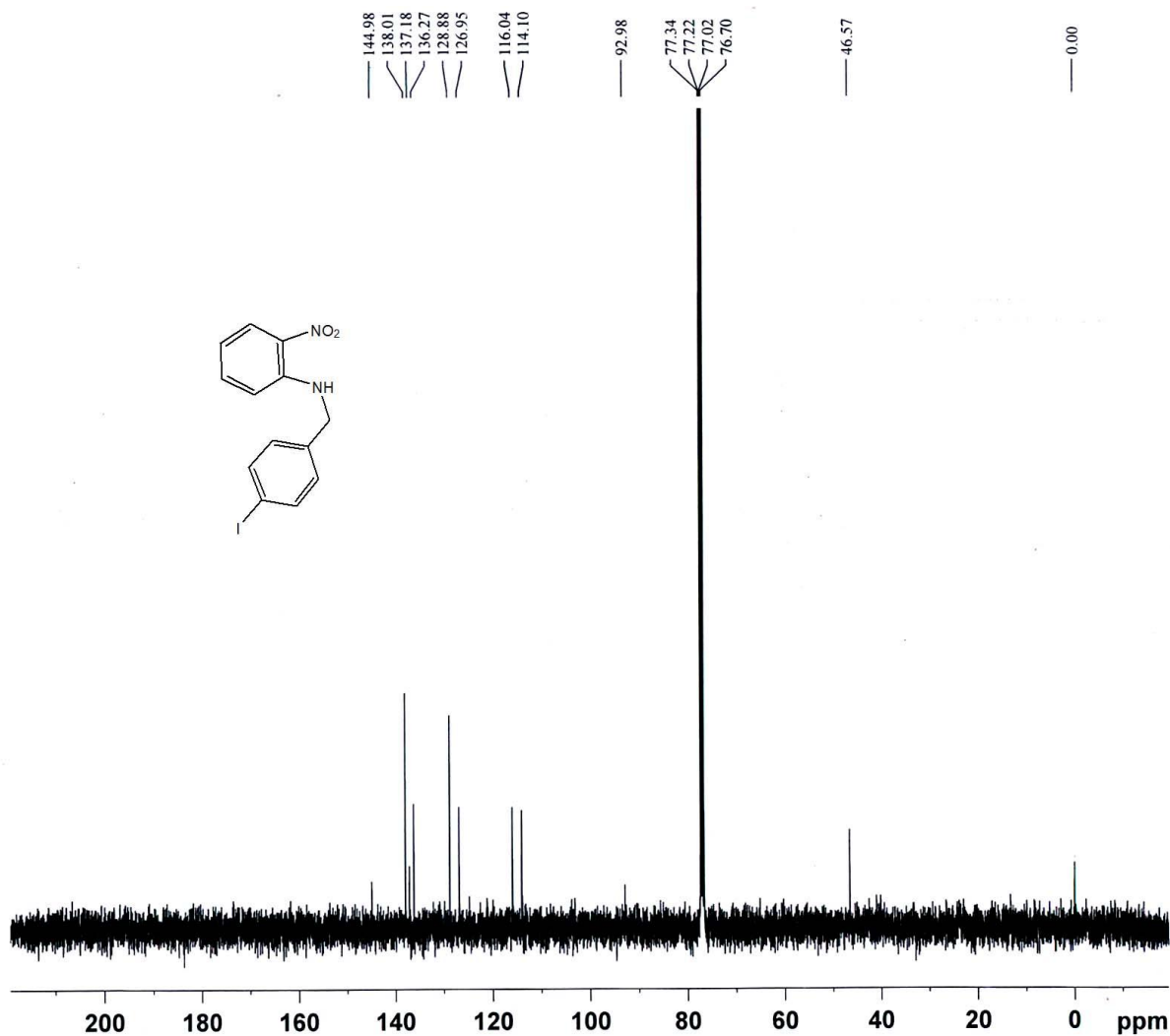


Table 3, Entry 8: ¹³C NMR



¹H NMR spectra of *N*¹-benzylbenzene-1,2-diamine

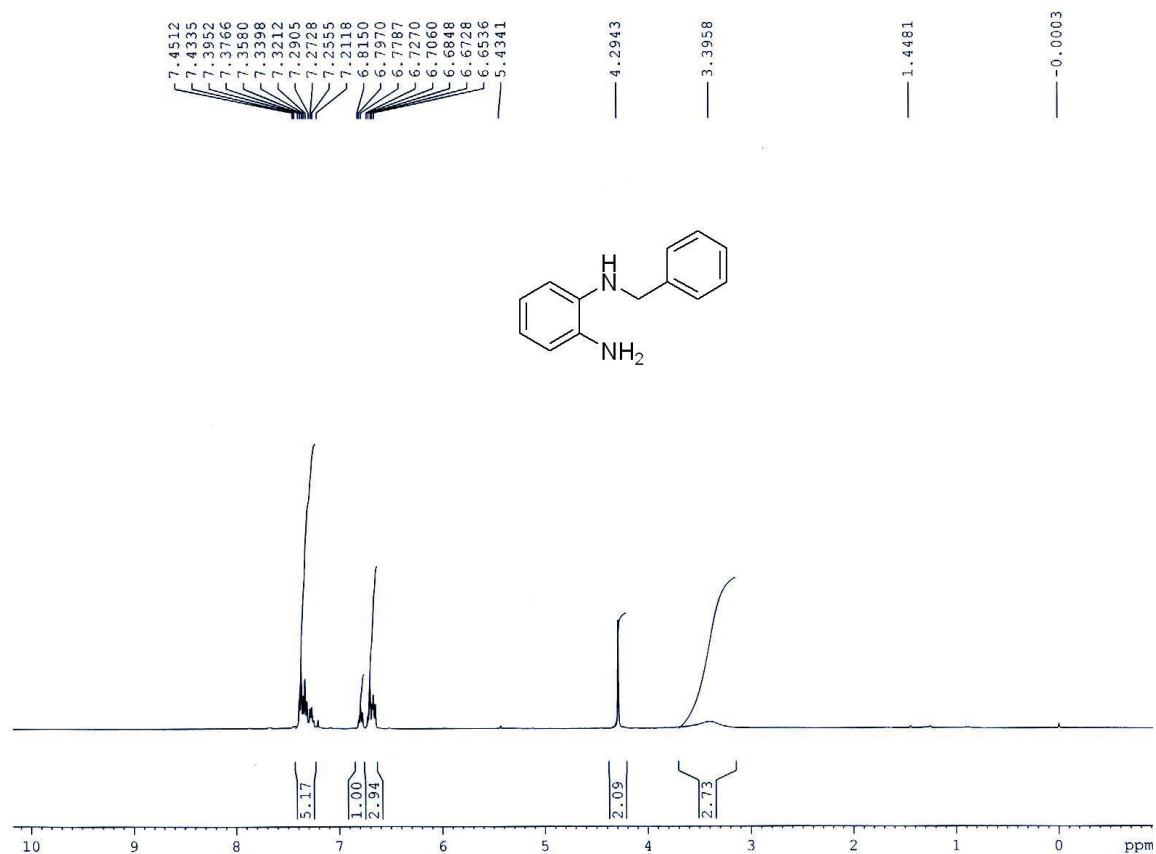


Table 5, Entry 1: ¹H NMR

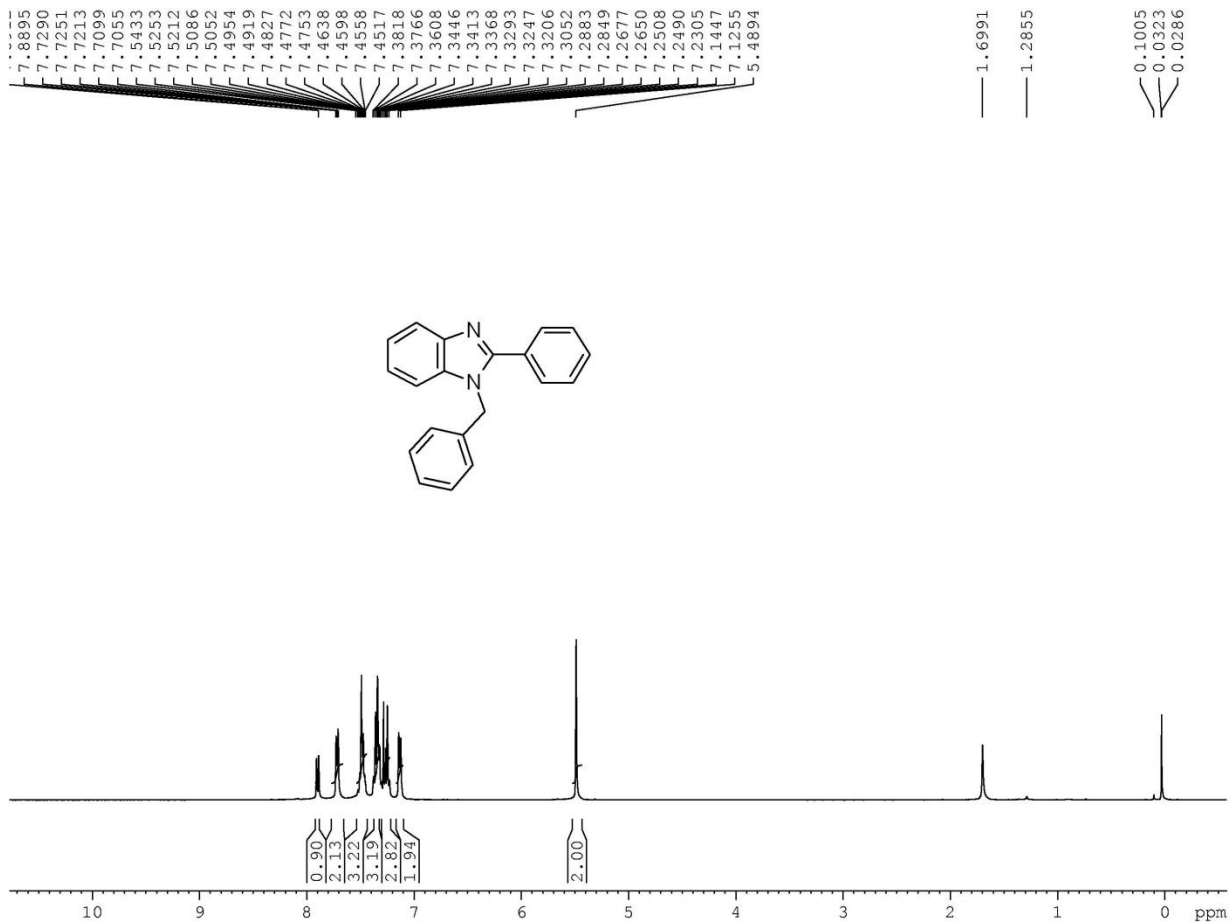


Table 5, Entry 2: ^1H NMR

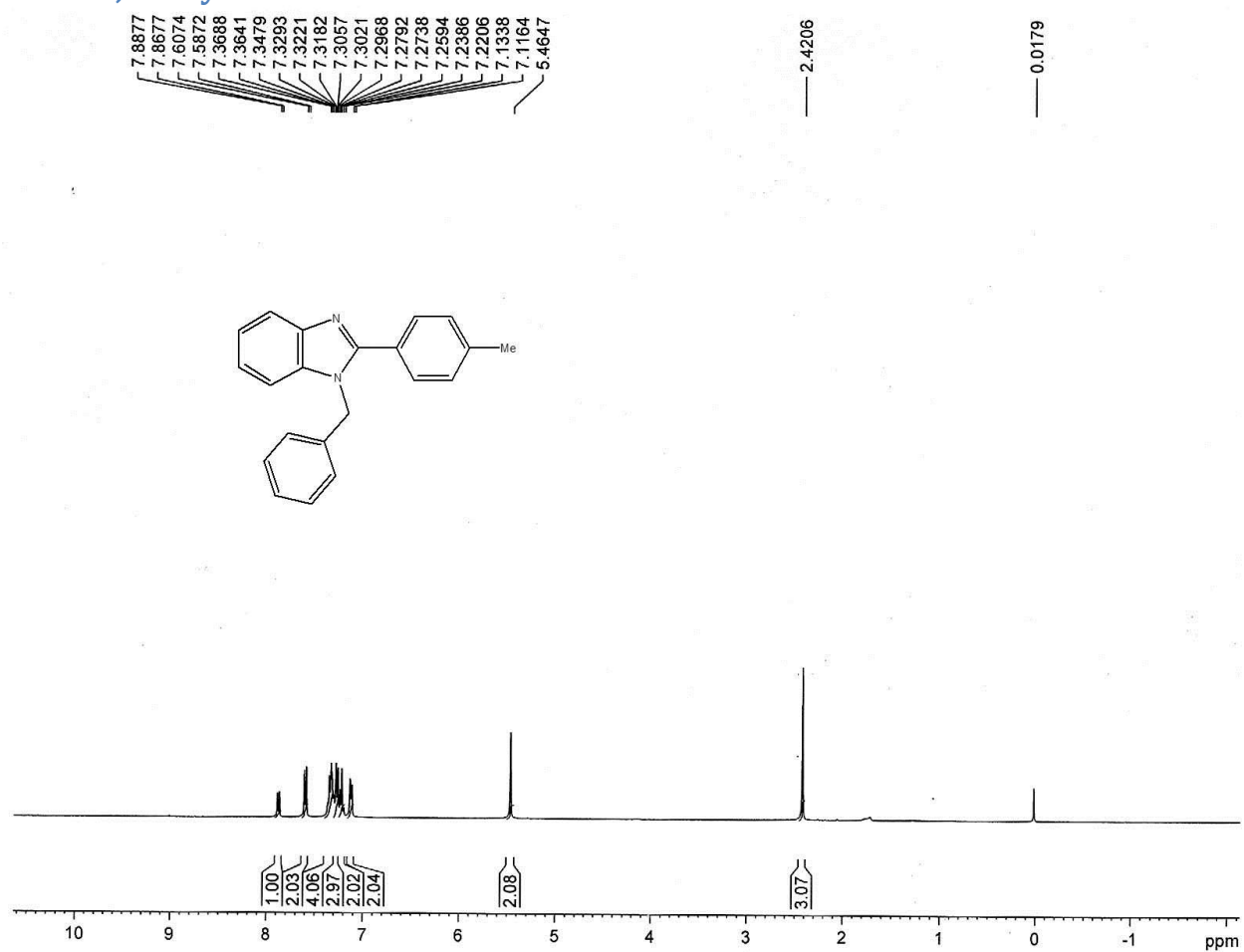


Table 5, Entry 3: ¹HNMR

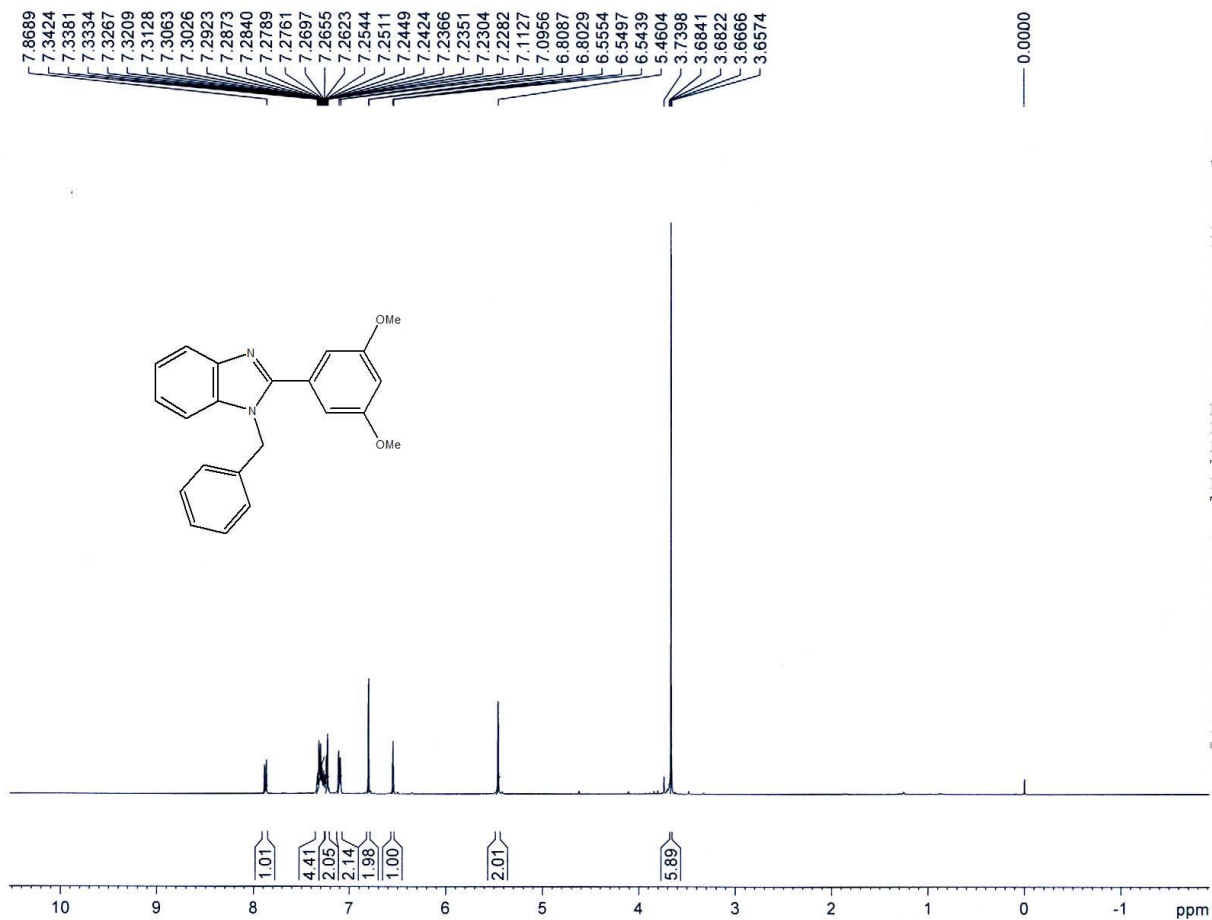


Table 5, Entry 3: ^{13}C NMR

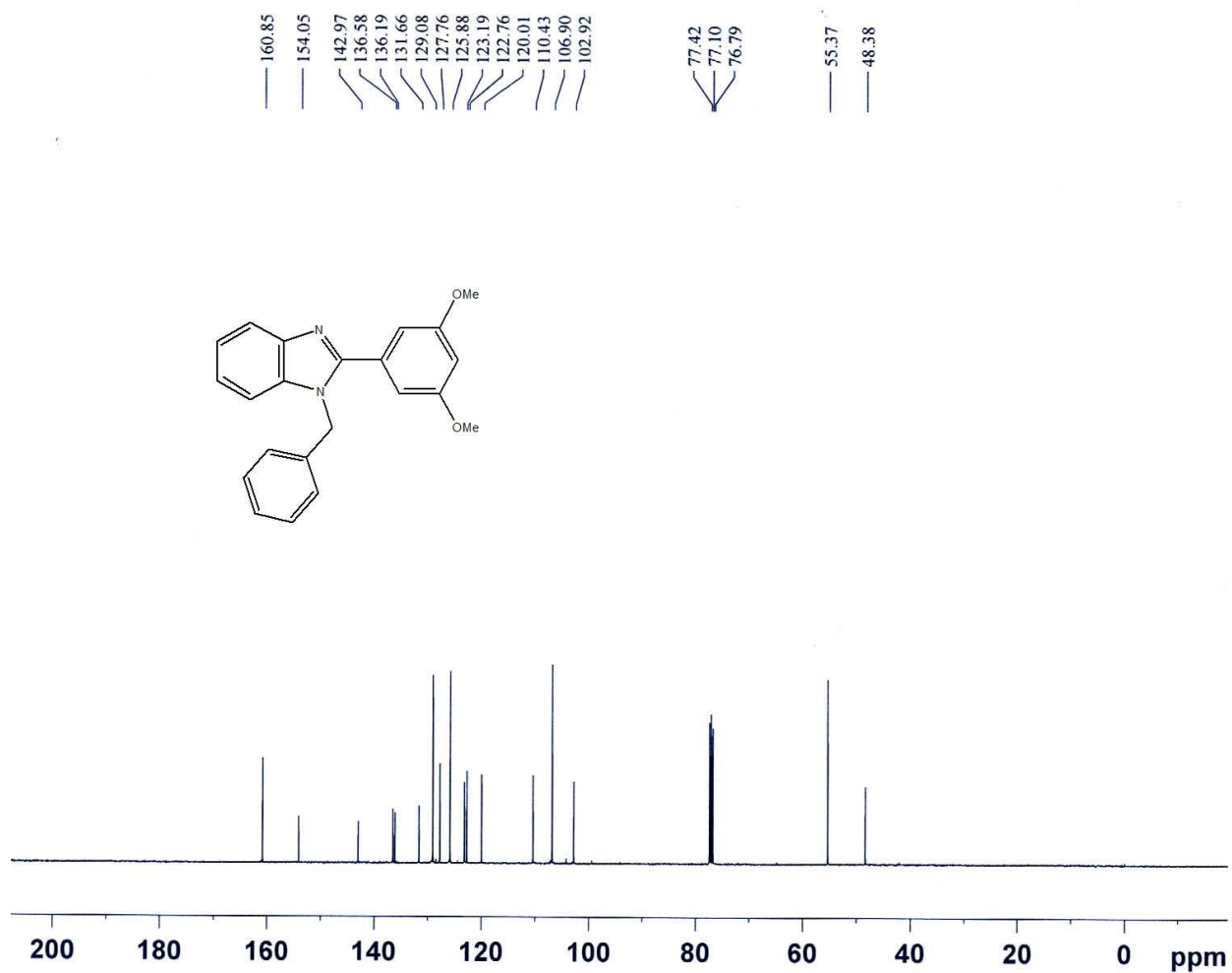


Table 5, Entry 4: ^1H NMR

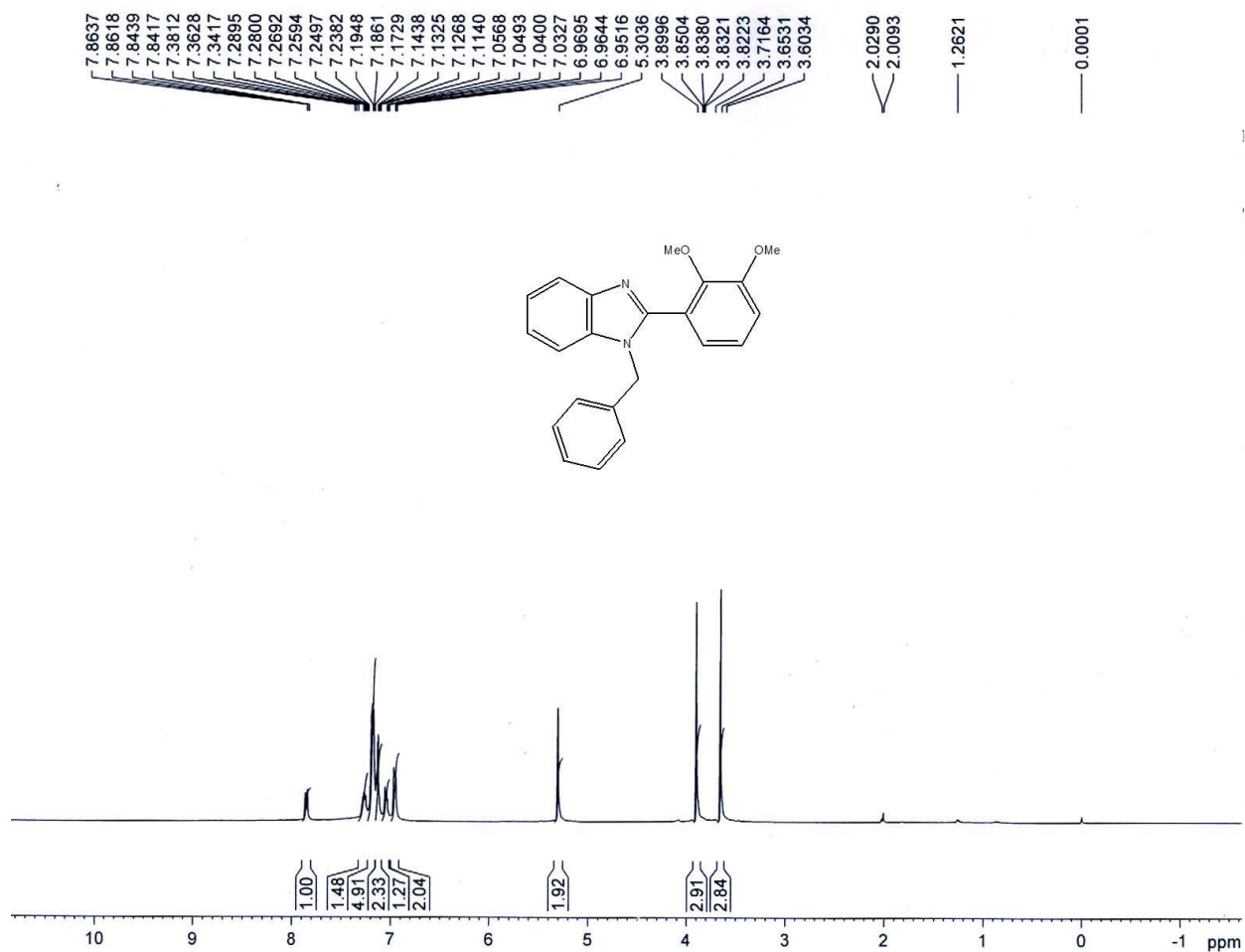


Table 5, Entry 4: ¹³C NMR

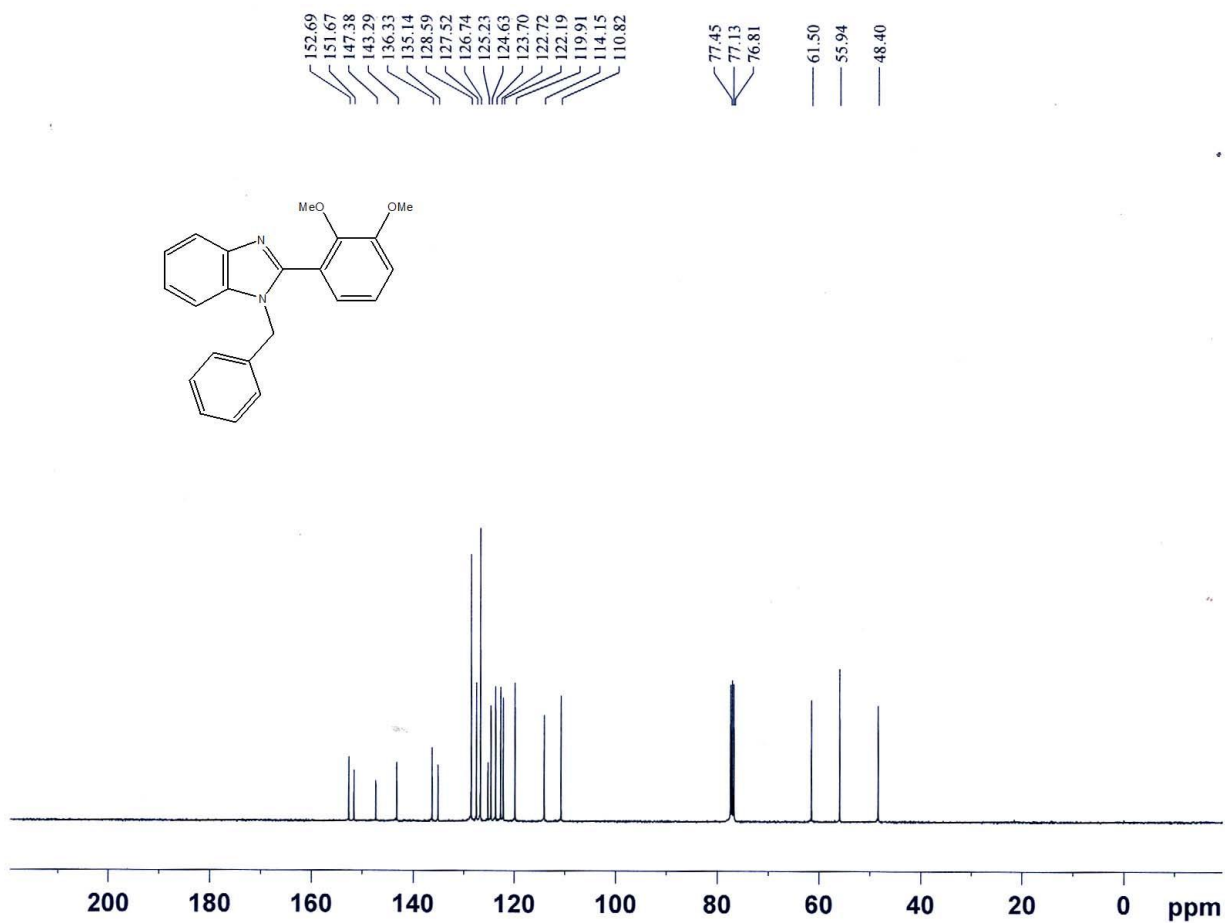


Table 5, Entry 5: ¹HNMR

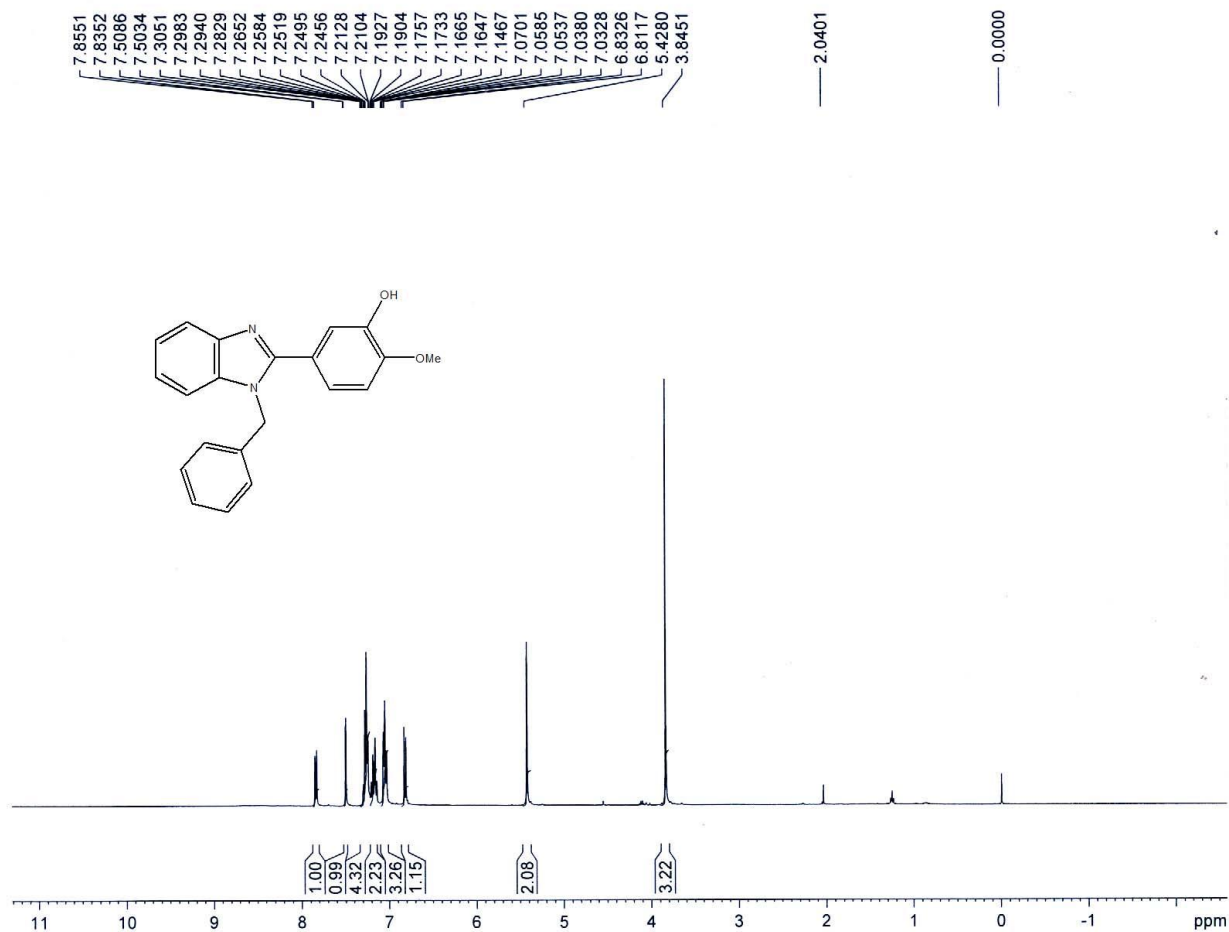


Table 5, Entry 5: ¹³C NMR

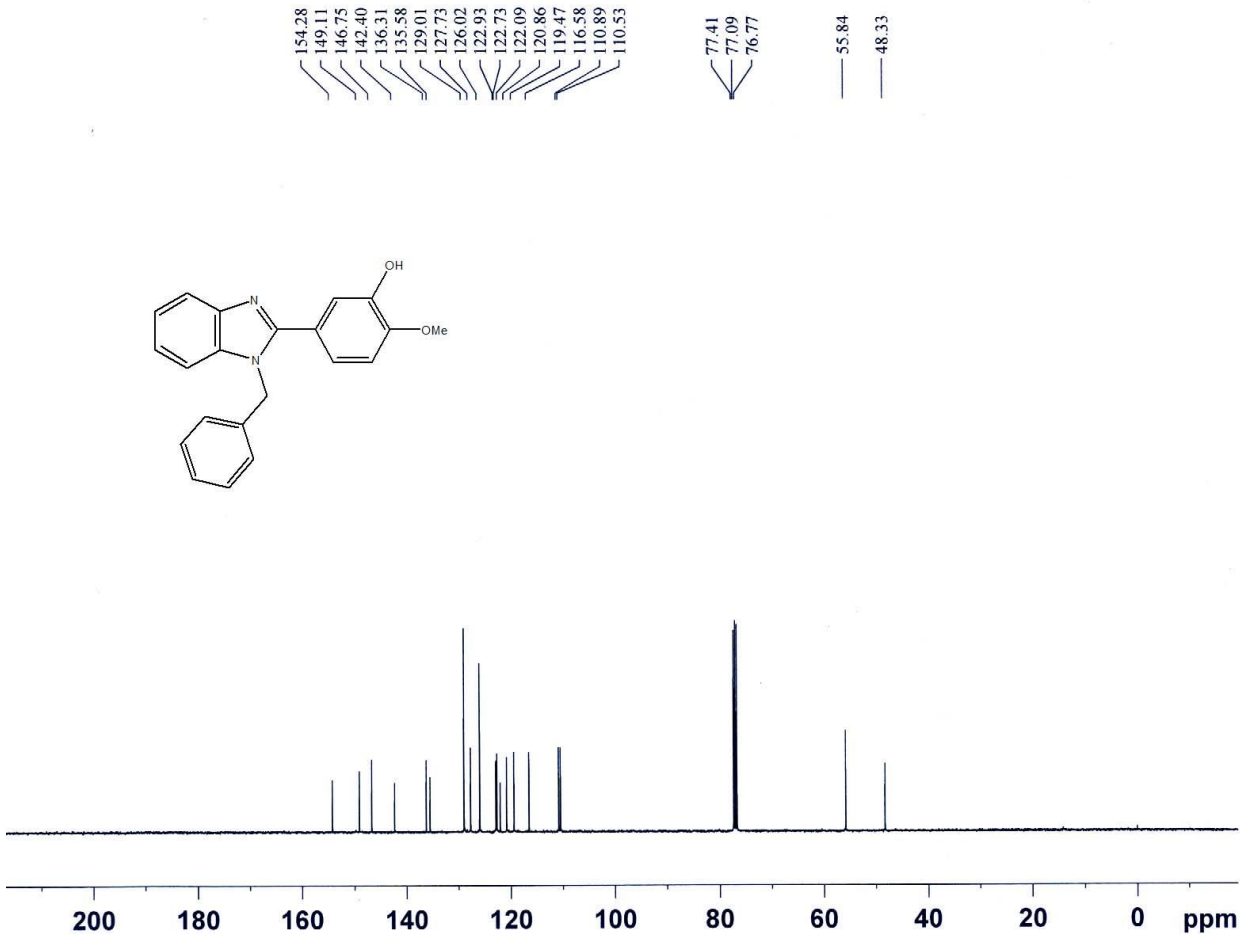


Table 5, Entry 6: ^1H NMR

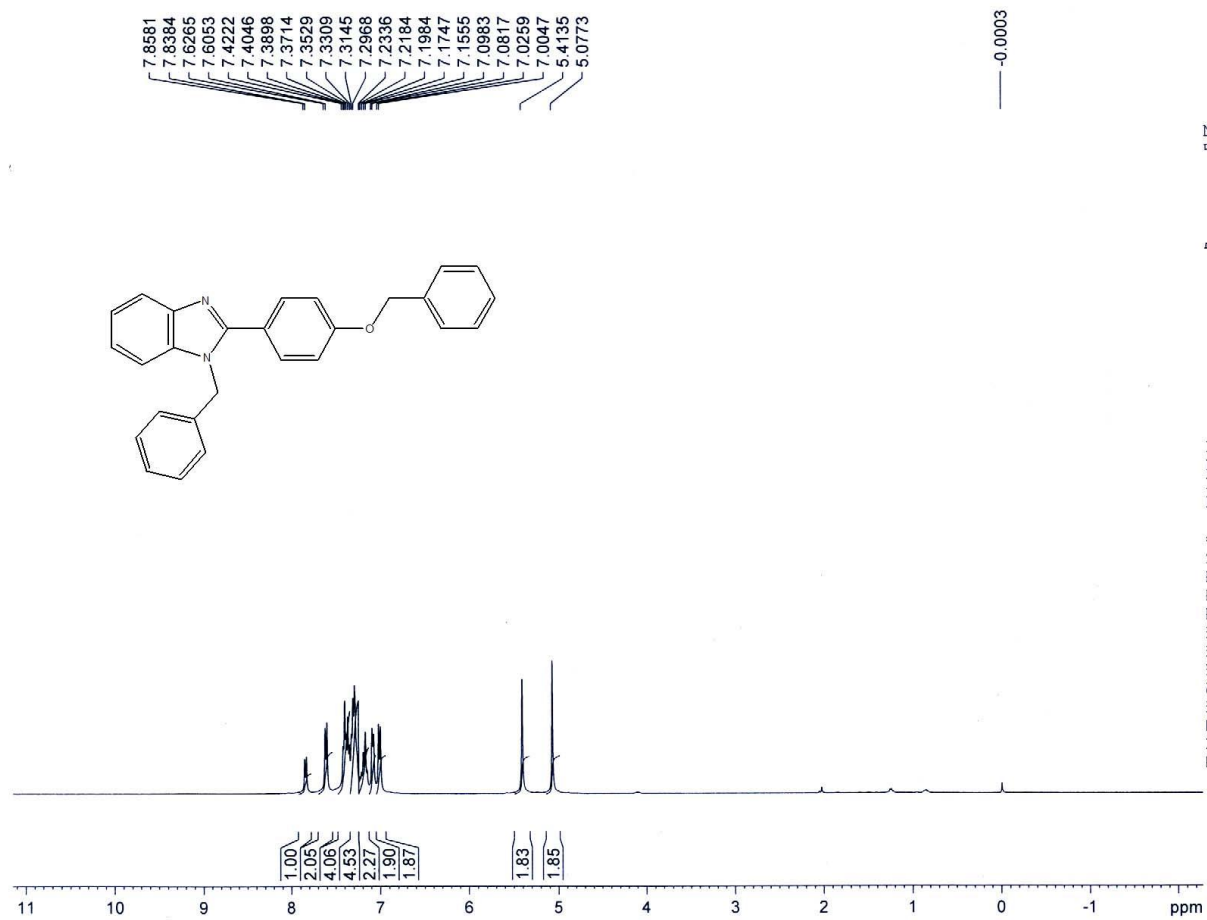


Table5, Entry 6: ^{13}C NMR

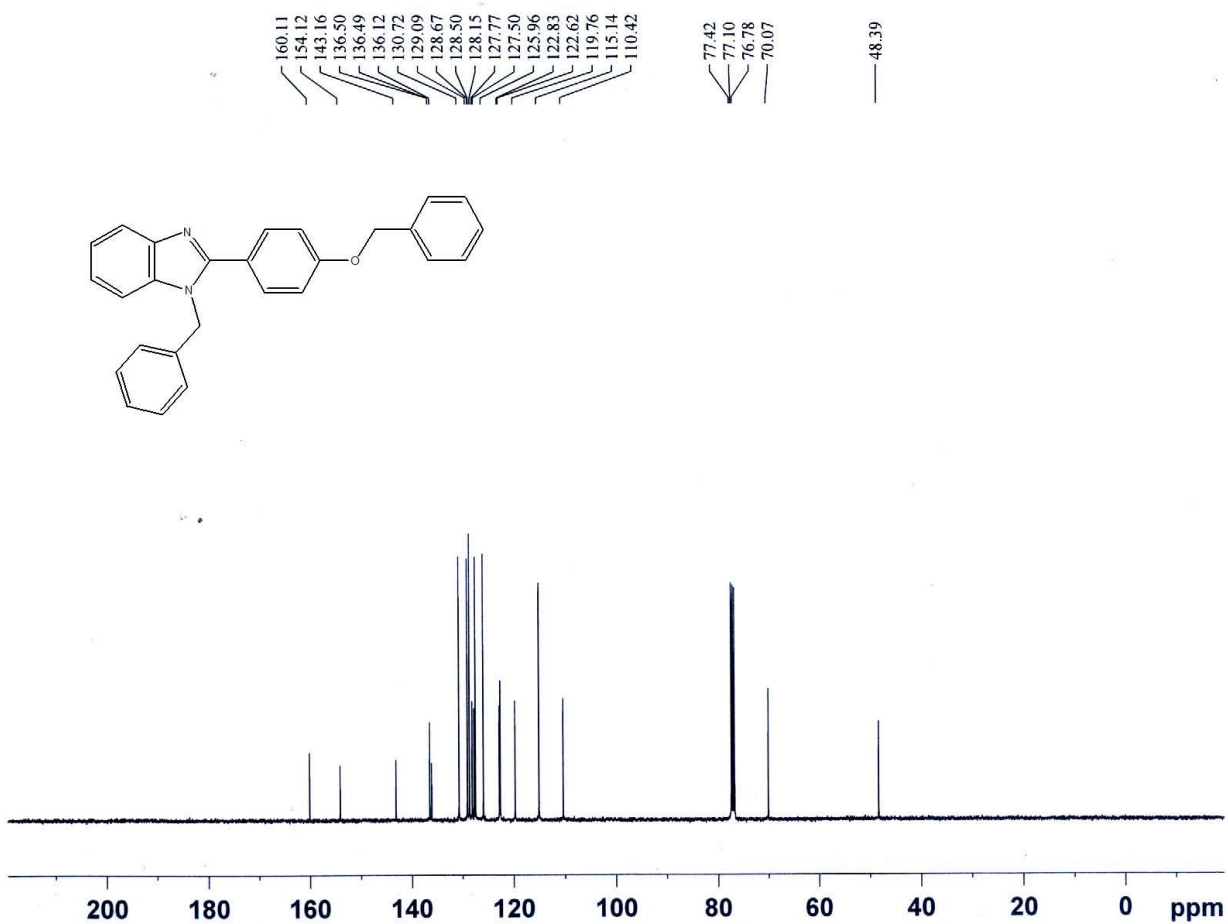


Table 5, Entry 7: ^1H NMR

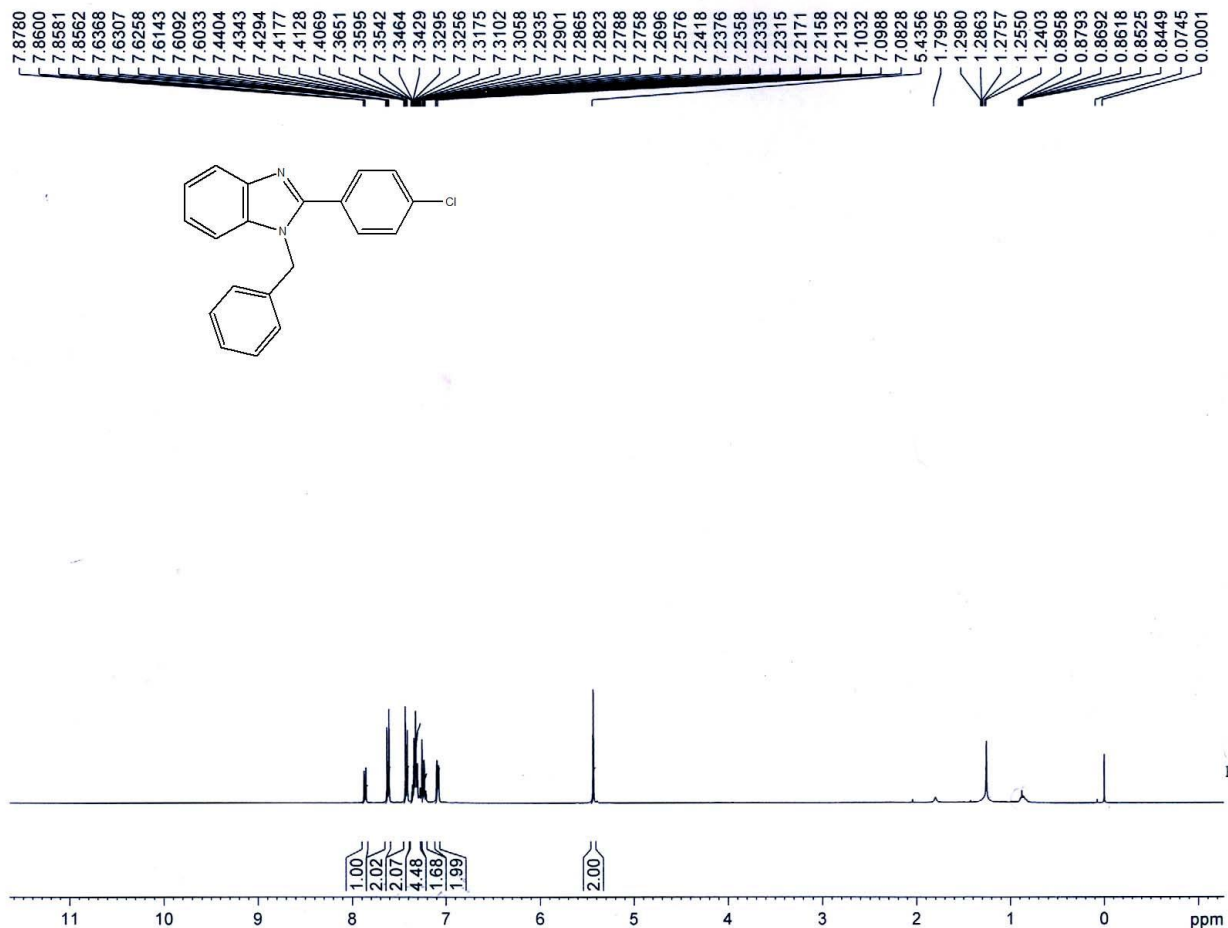


Table 5, Entry 8: ^1H NMR

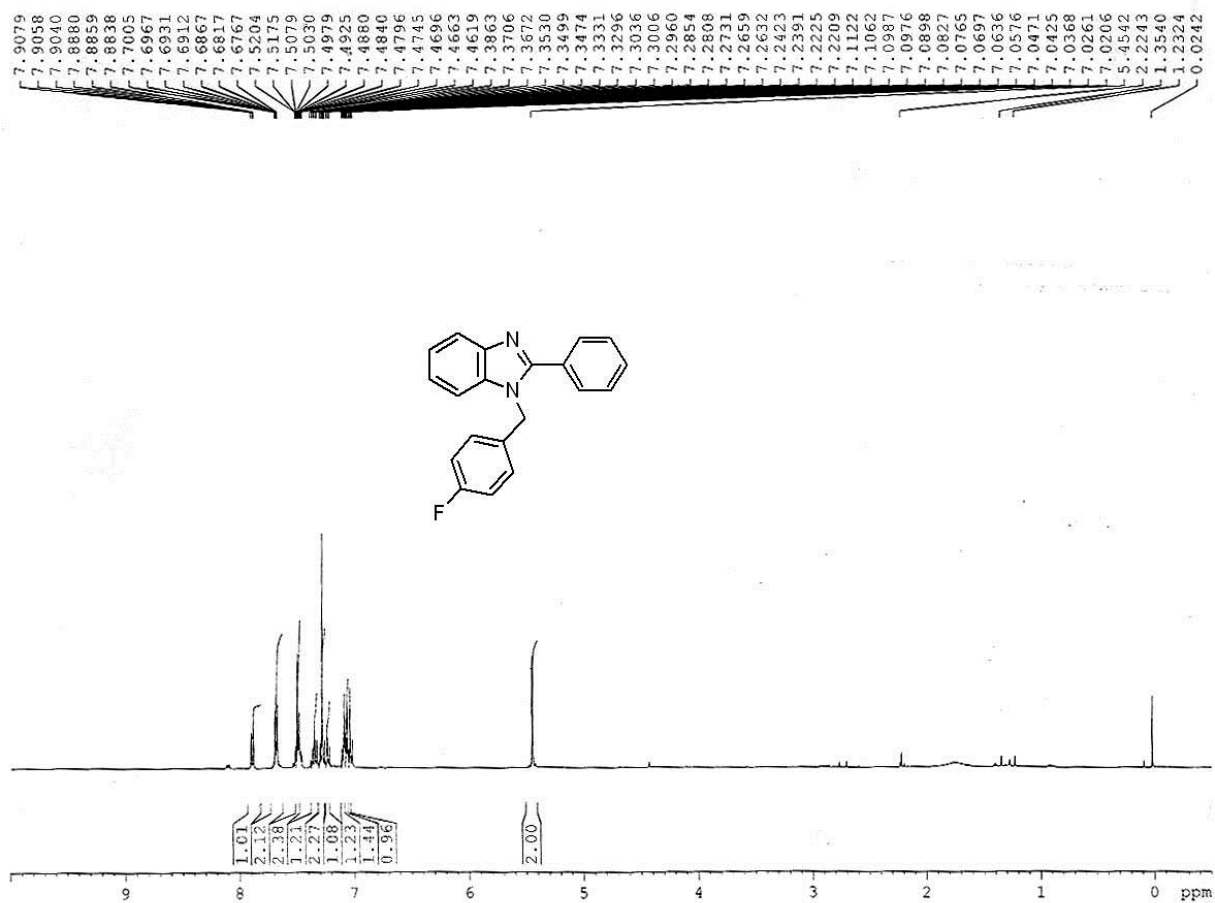


Table 5, Entry 8: ^{13}C NMR

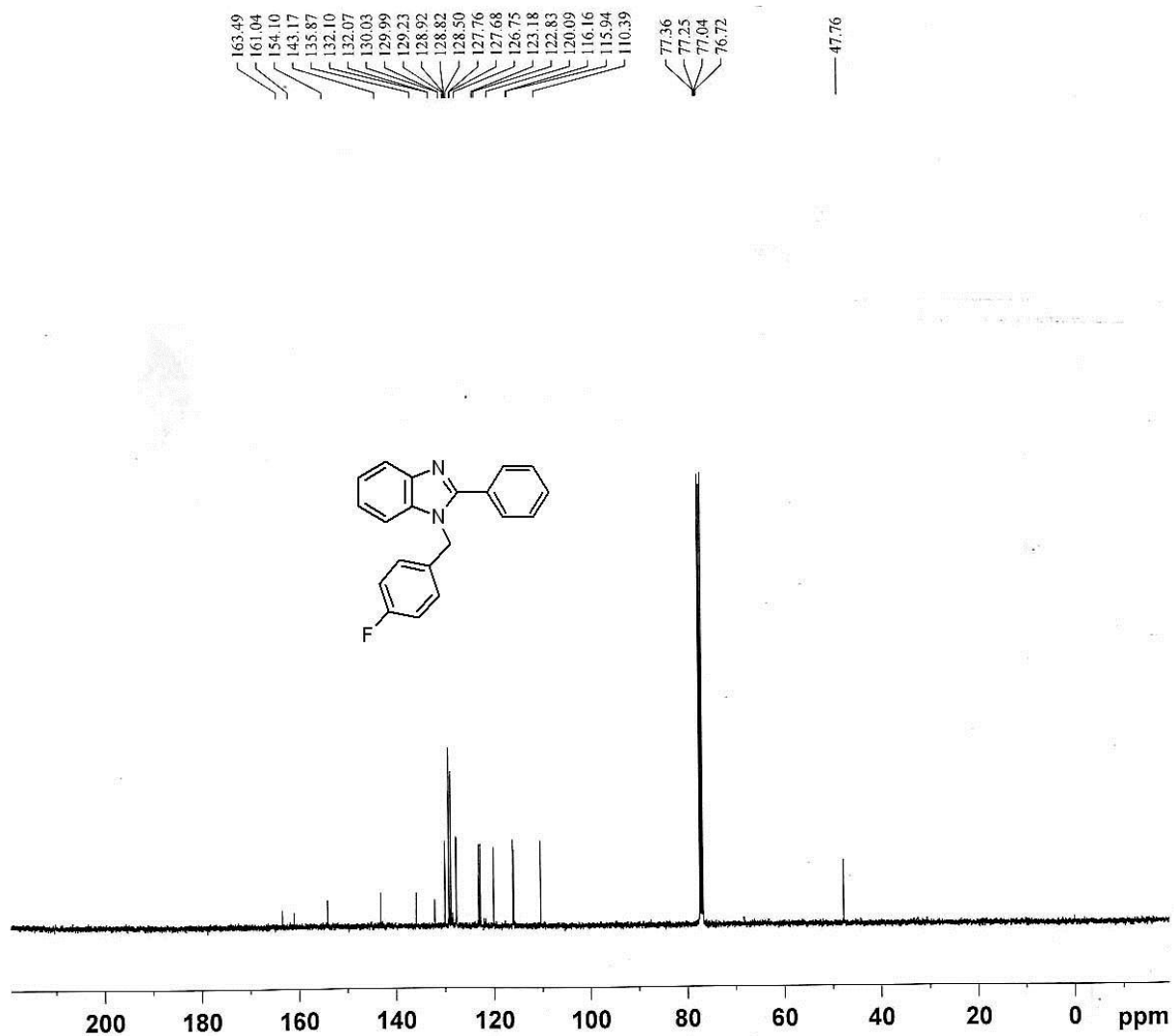


Table 5, Entry 9: ¹H NMR

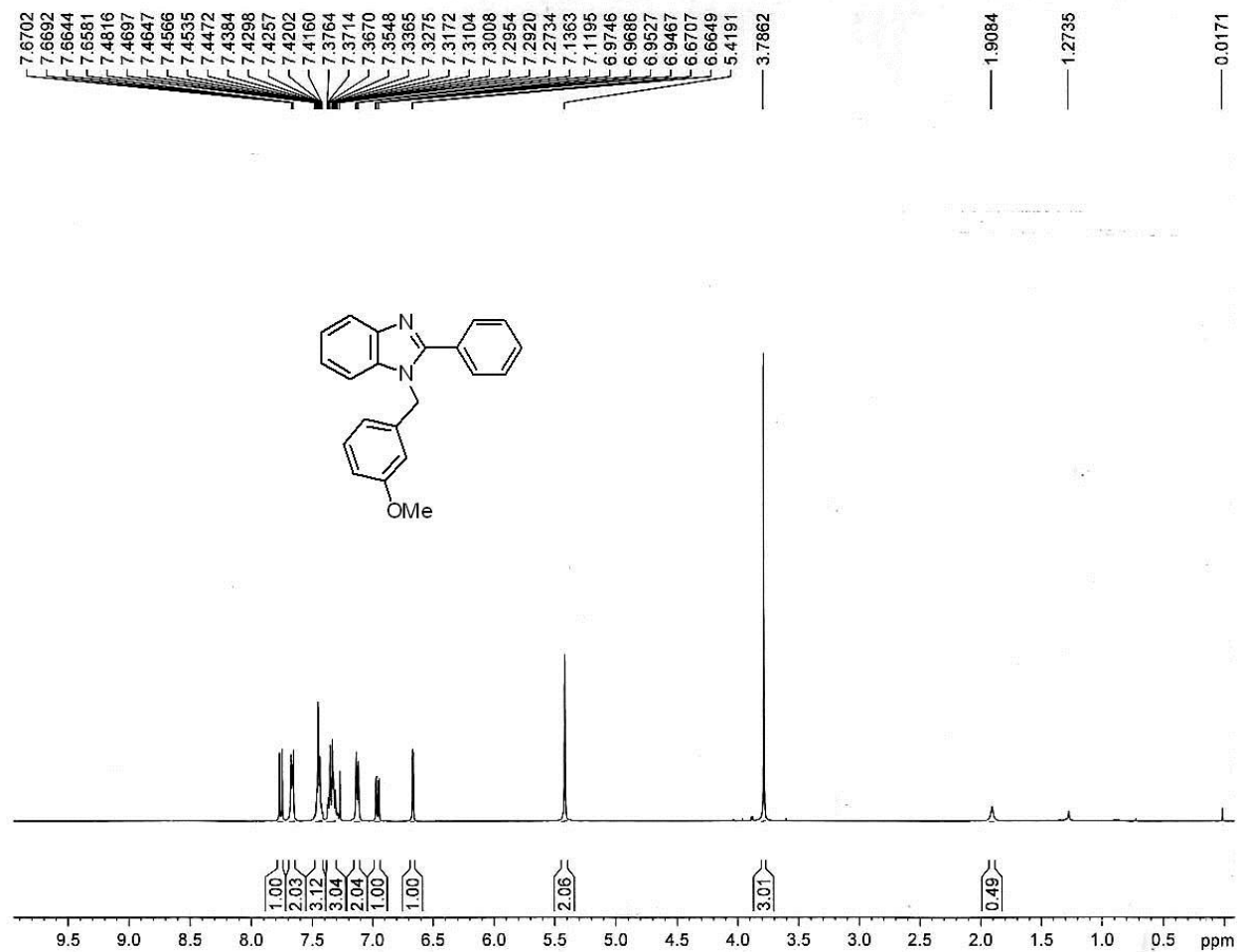


Table 5, Entry 9: ^{13}C NMR

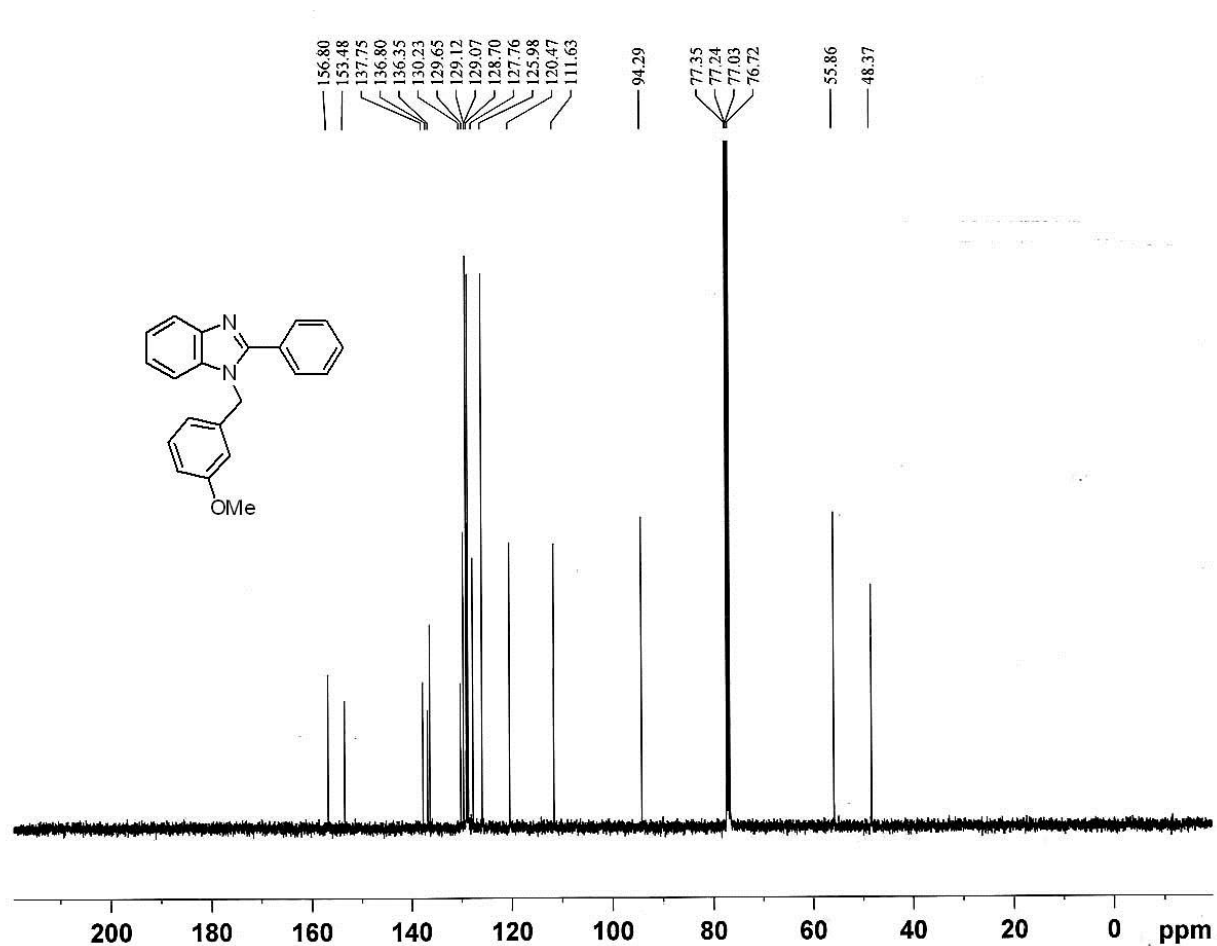


Table 5, Entry 10: ¹H NMR

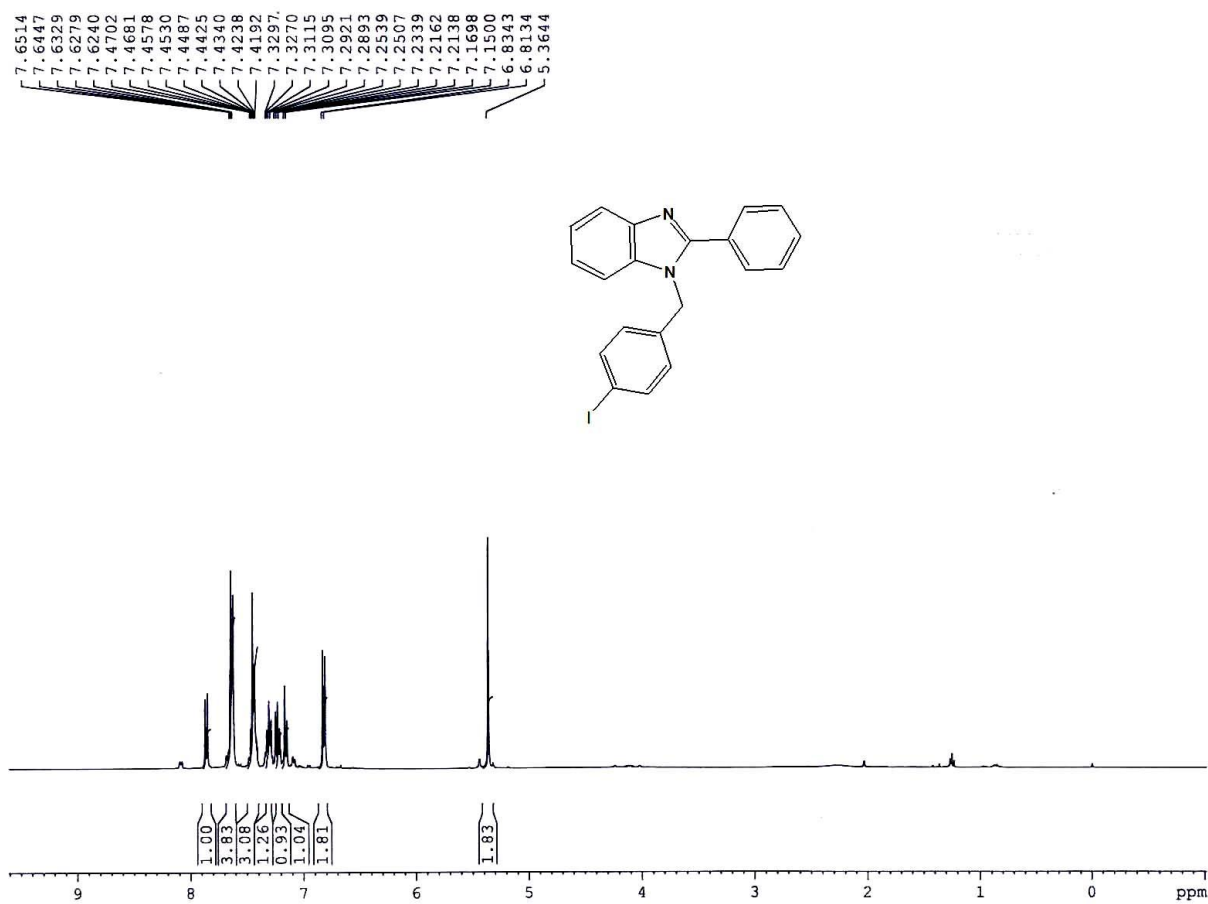


Table 5, Entry 10: ^{13}C NMR

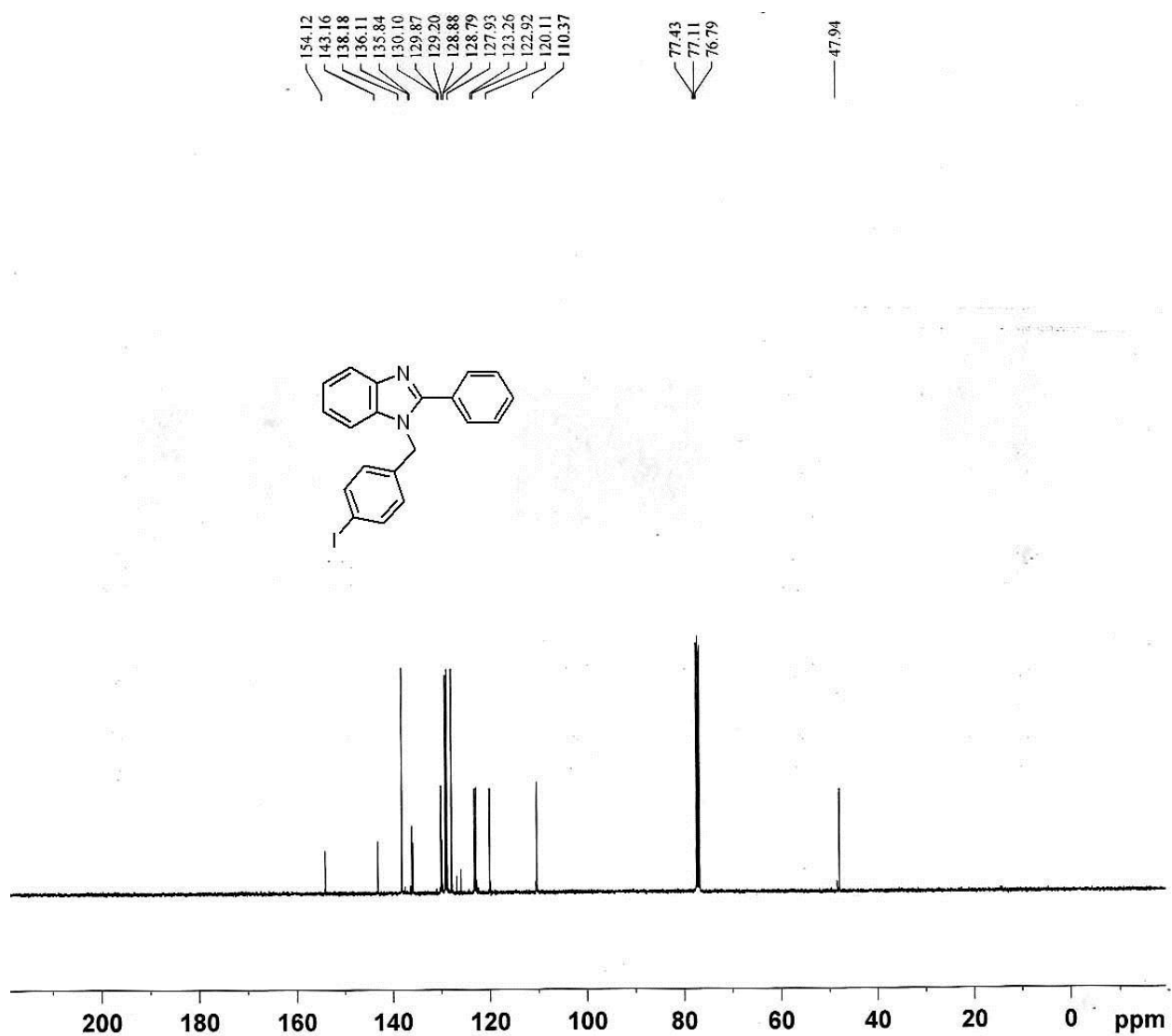


Table 5, Entry 11: ¹H NMR

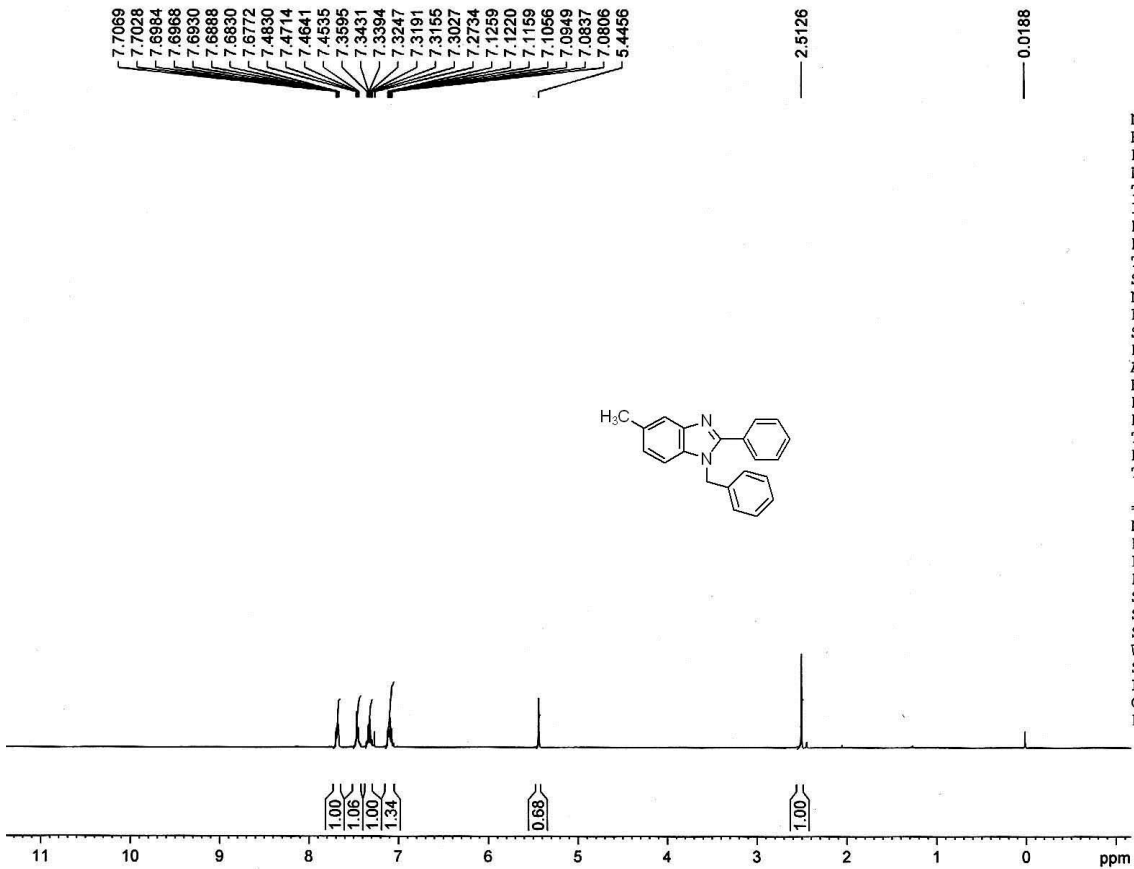


Table 5, Entry 11: ¹³C NMR

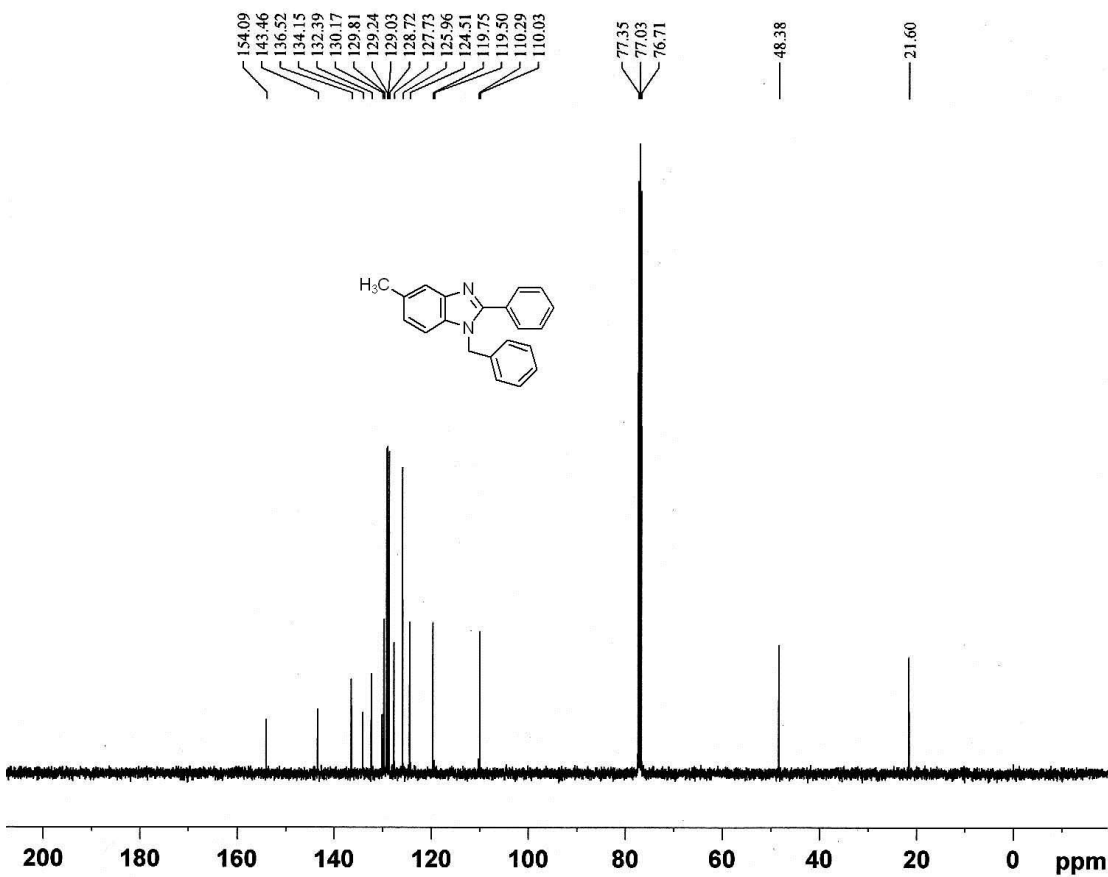


Table 5, Entry 12: ^1H NMR

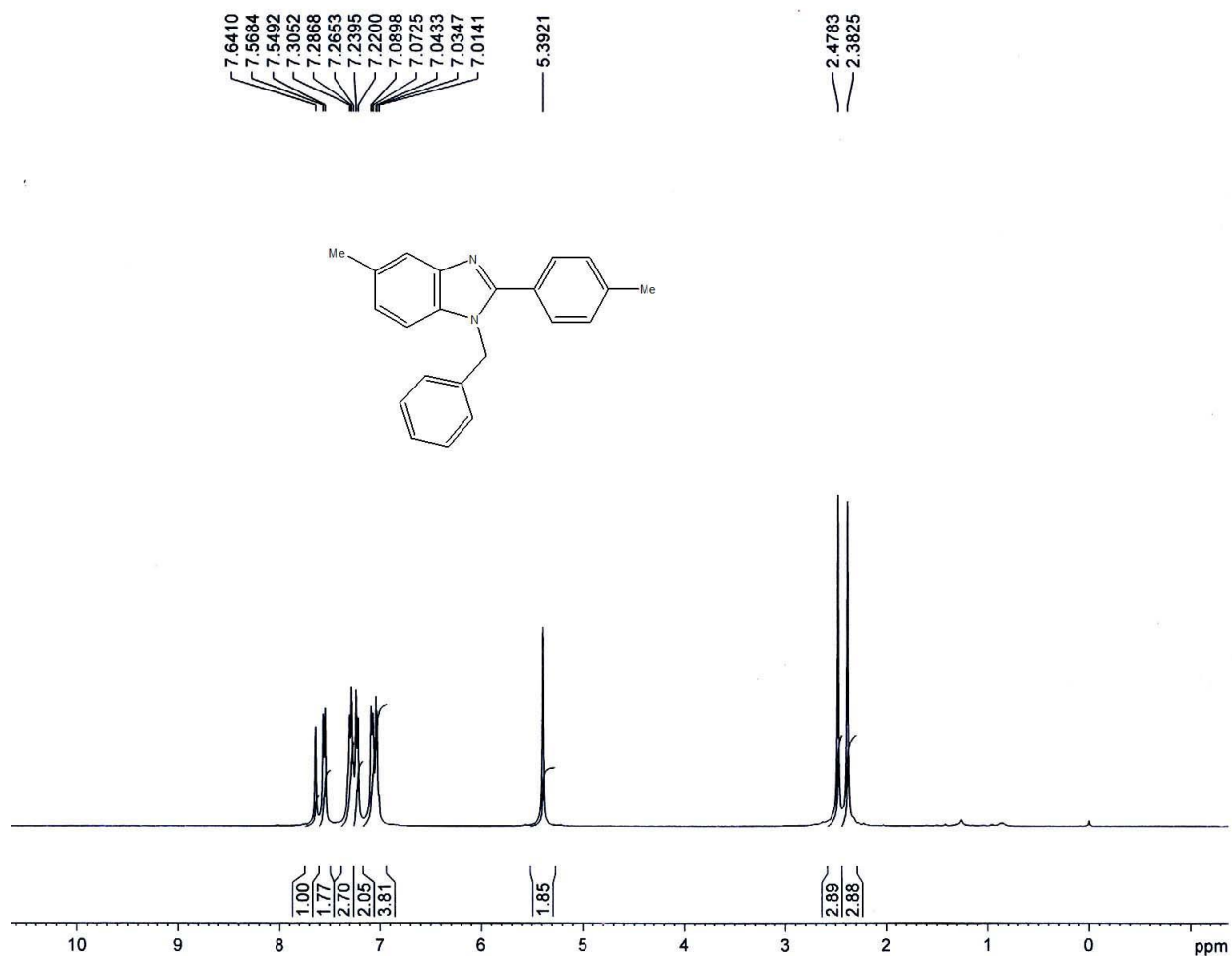


Table 5, Entry 12: ¹³C NMR

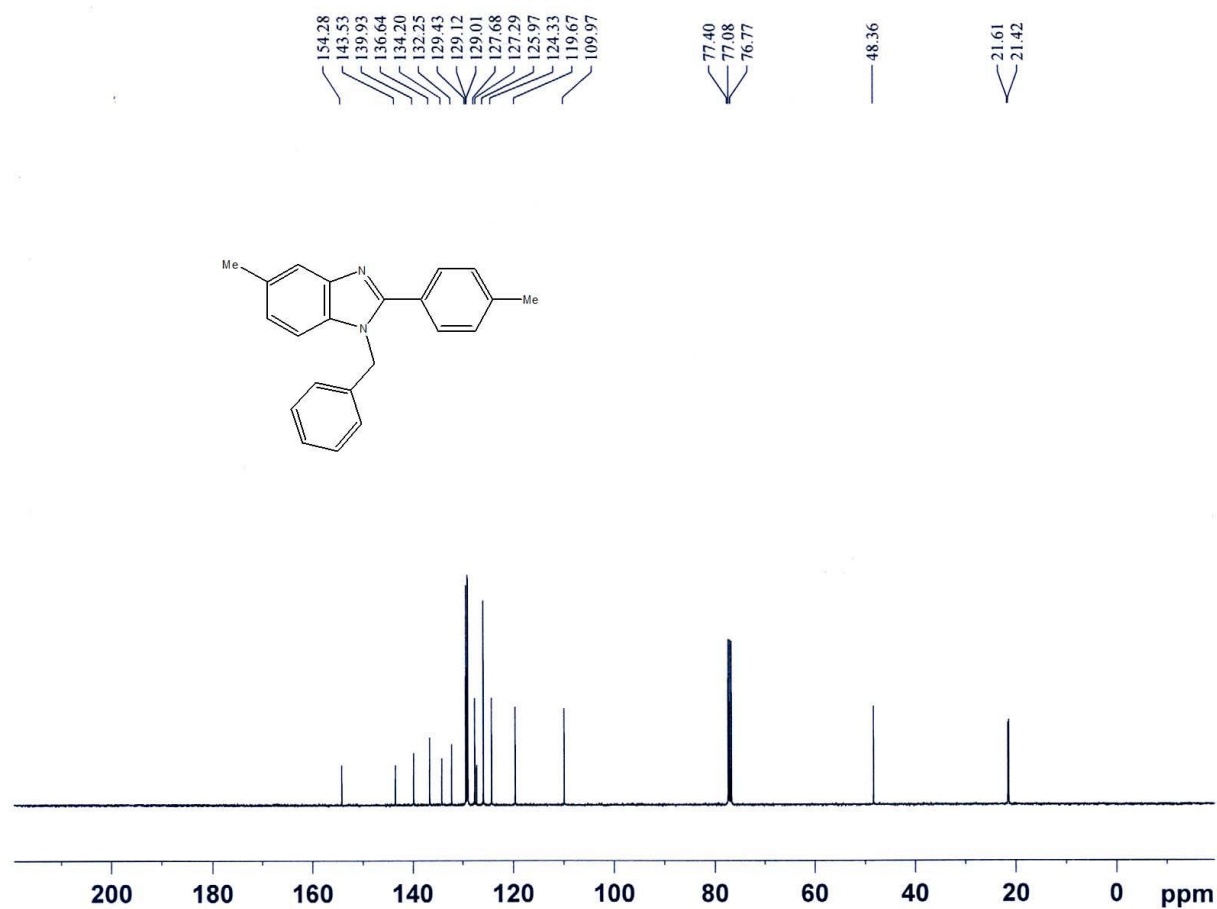


Table 5, Entry 13: ¹H NMR

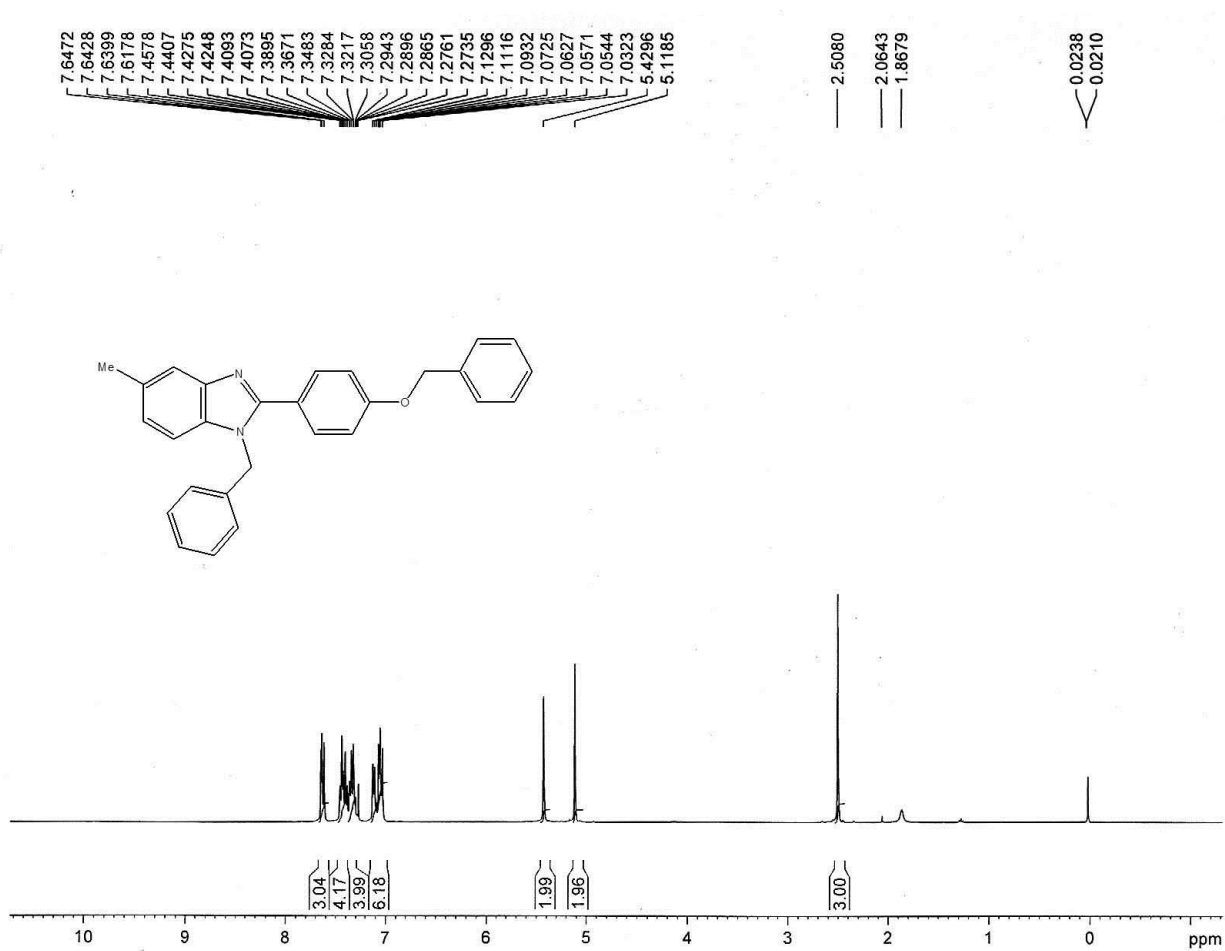


Table 5, Entry 13: ^{13}C NMR

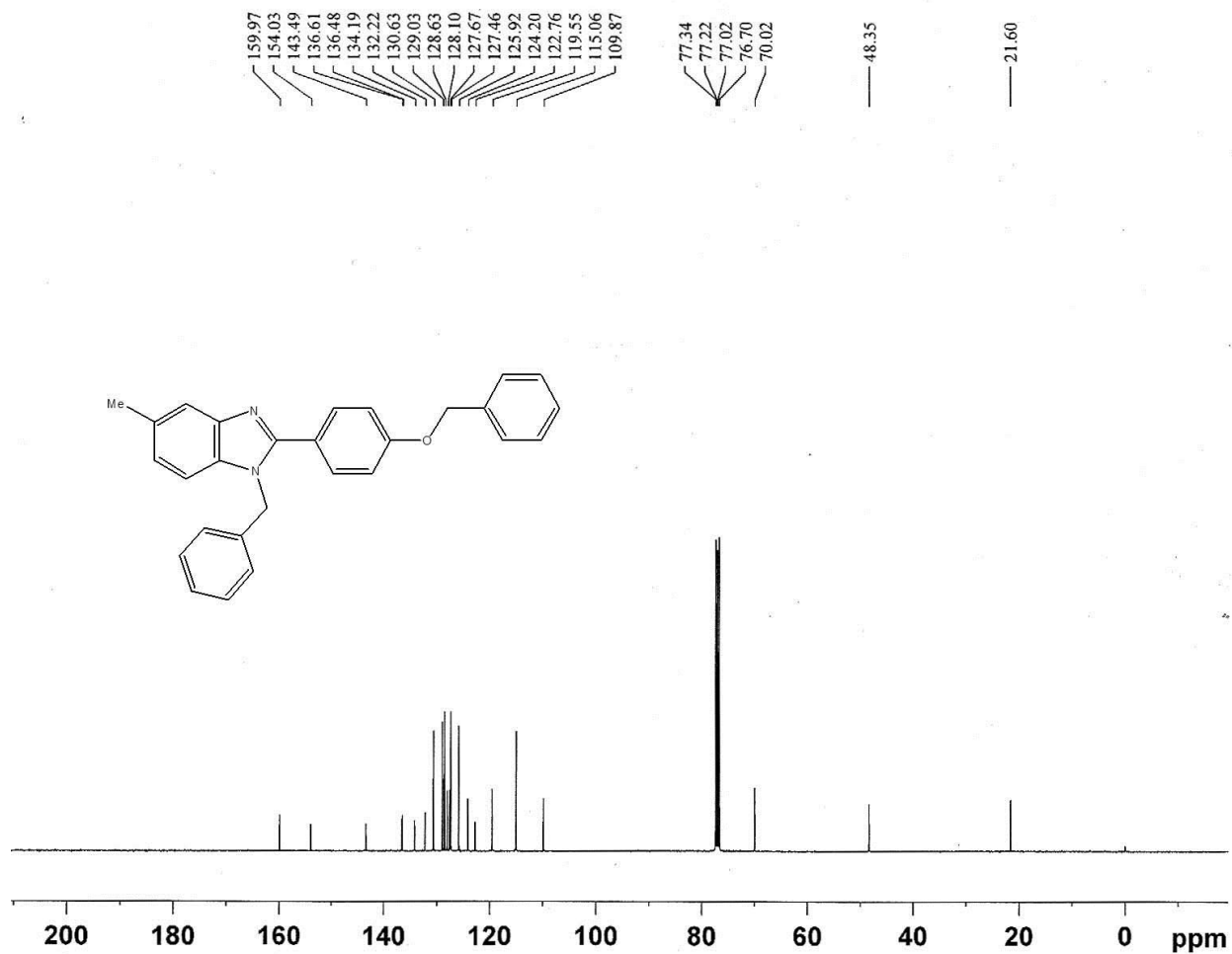


Table 5, Entry 14: ^1H NMR

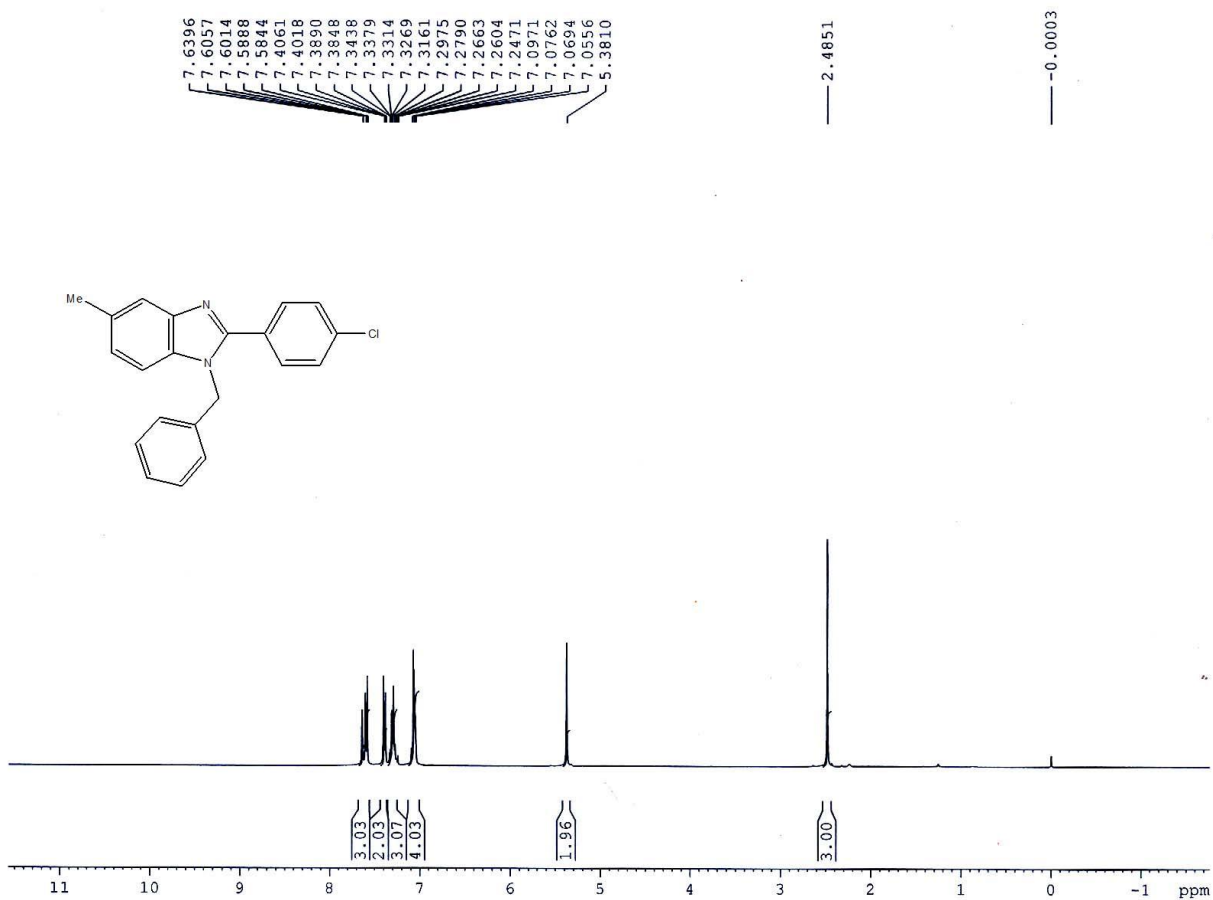


Table 5, Entry 14: ¹³C NMR

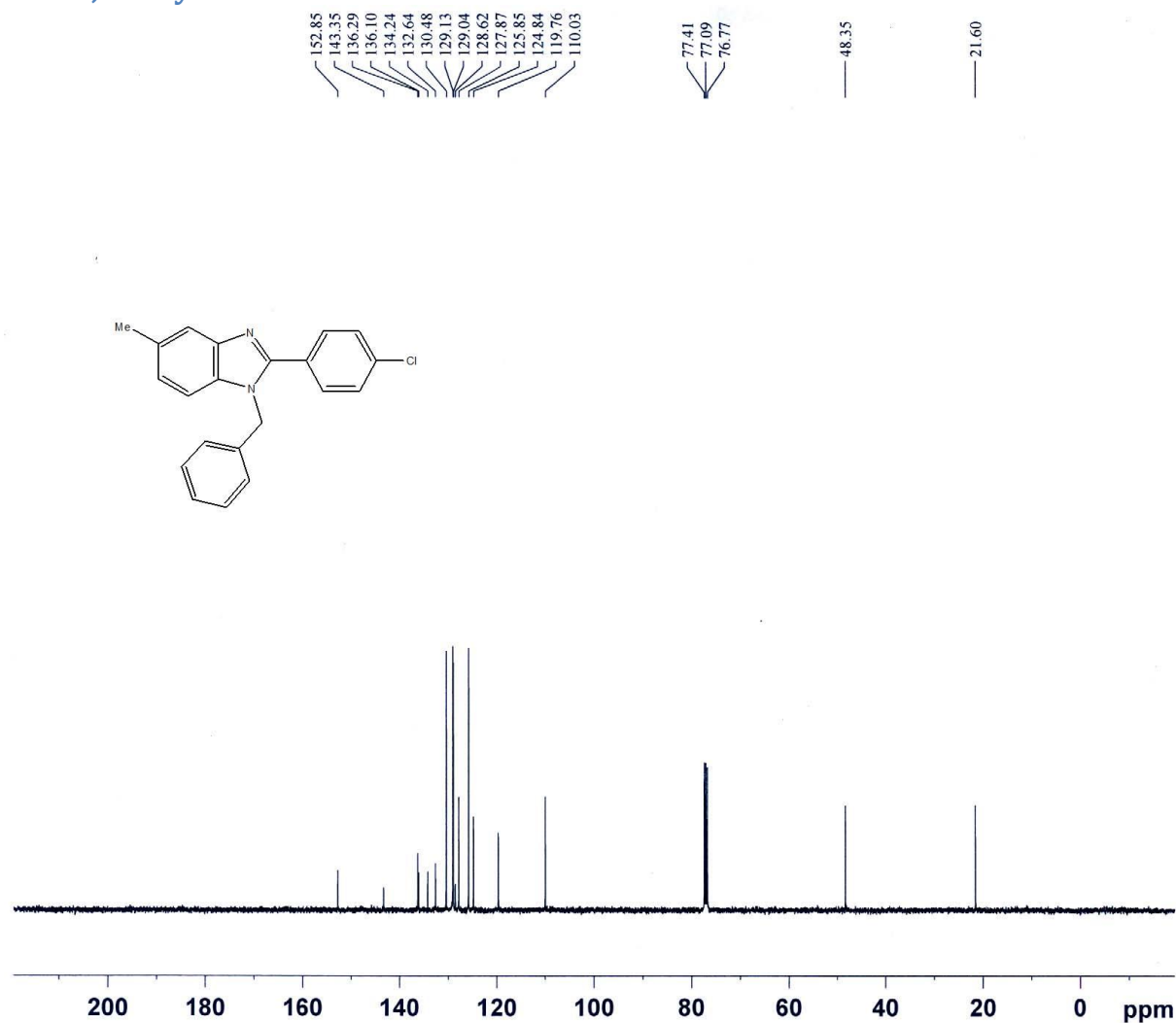


Table 5, Entry 15: ¹H NMR

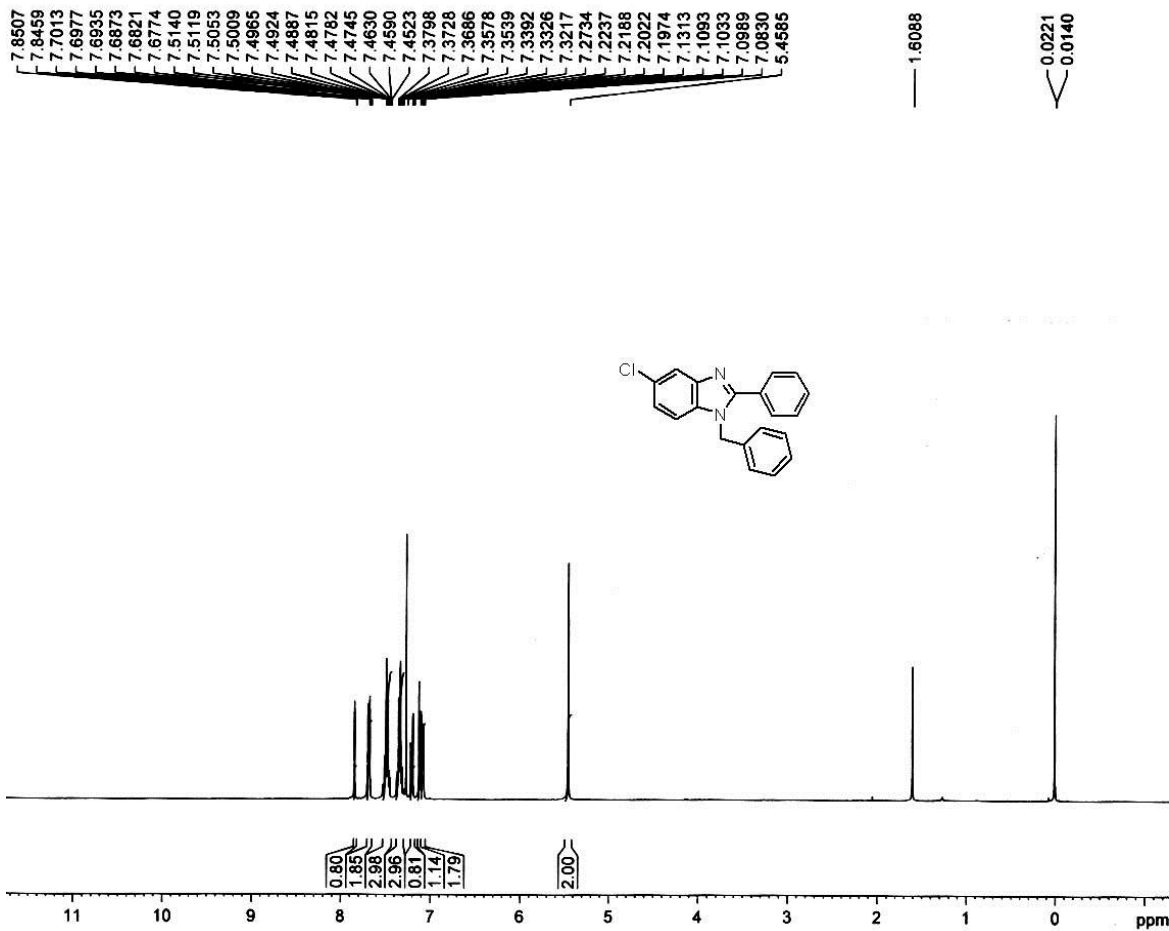


Table 5, Entry 15: ¹H NMR

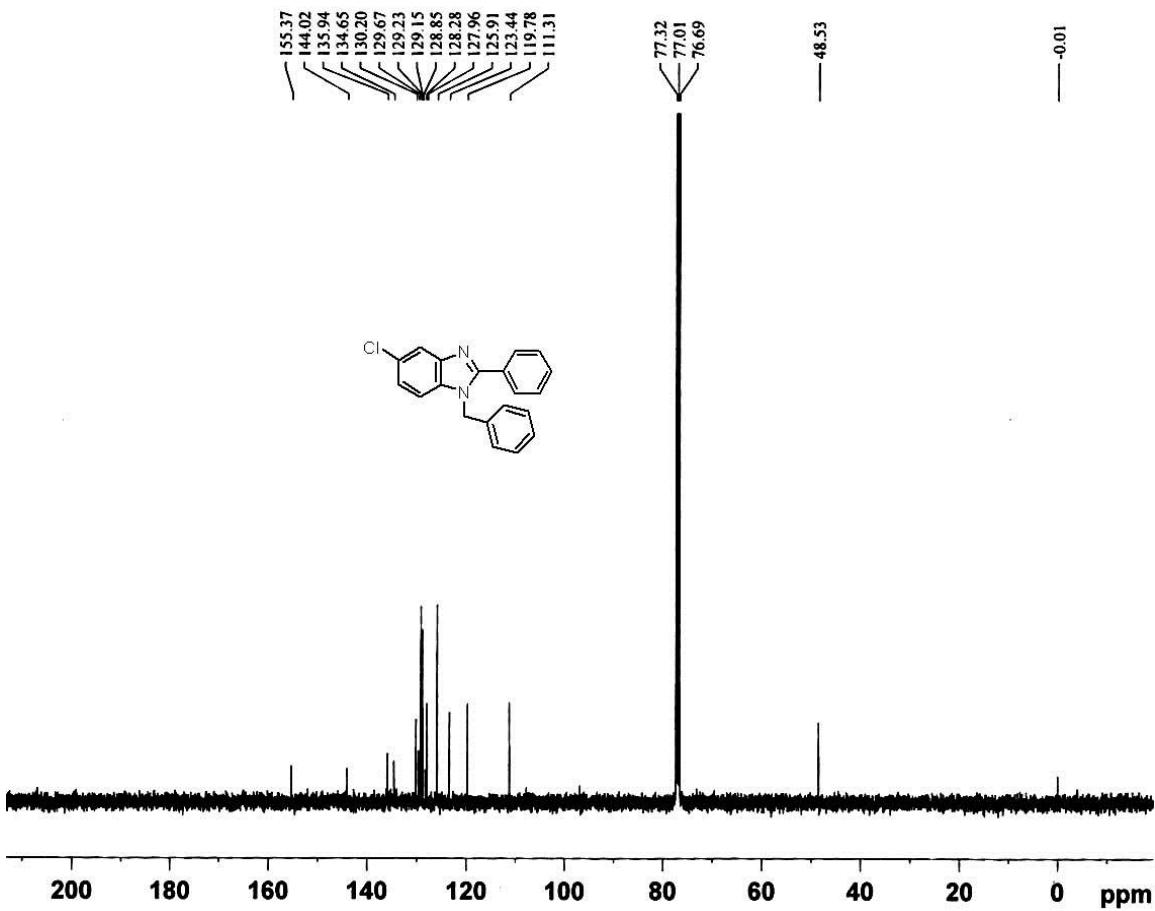


Table 5, Entry 16: ¹H NMR

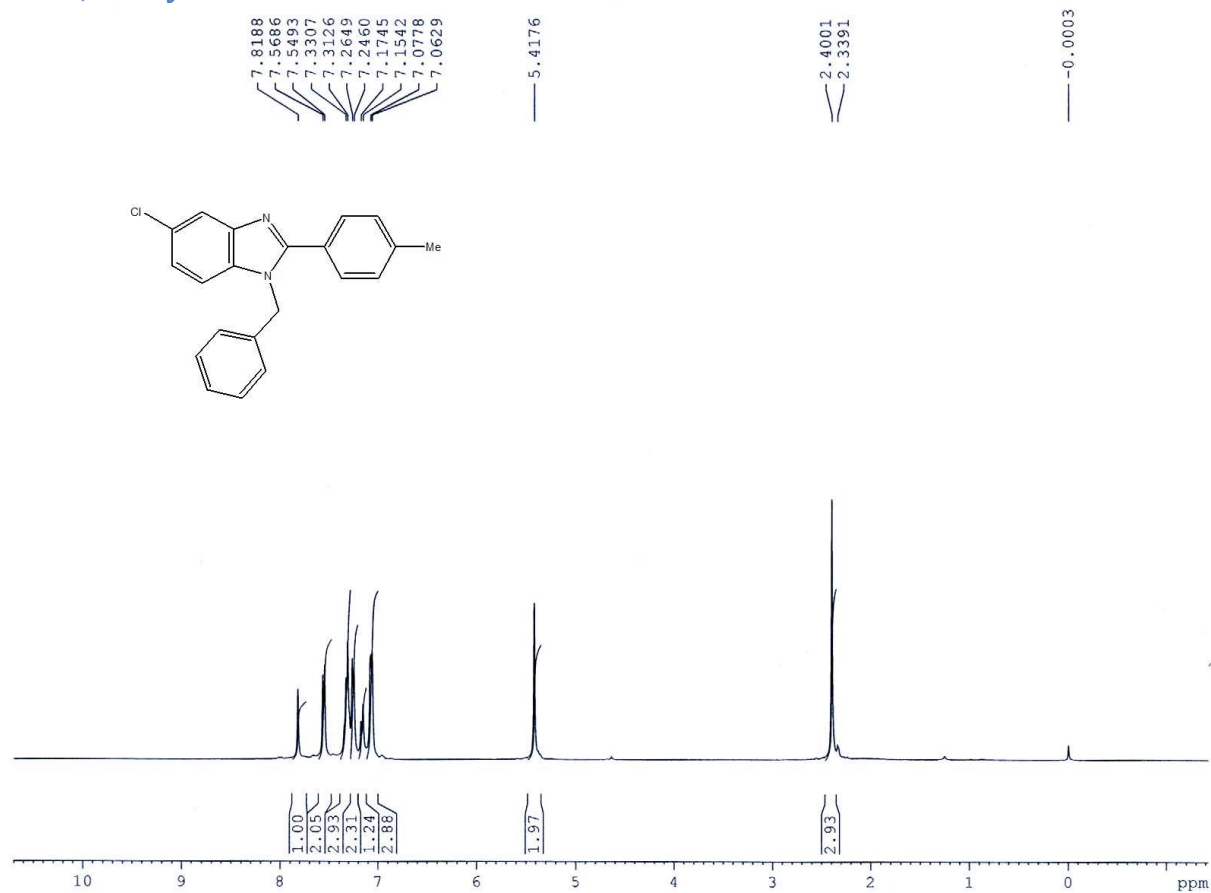


Table 5, Entry 16: ¹³C NMR

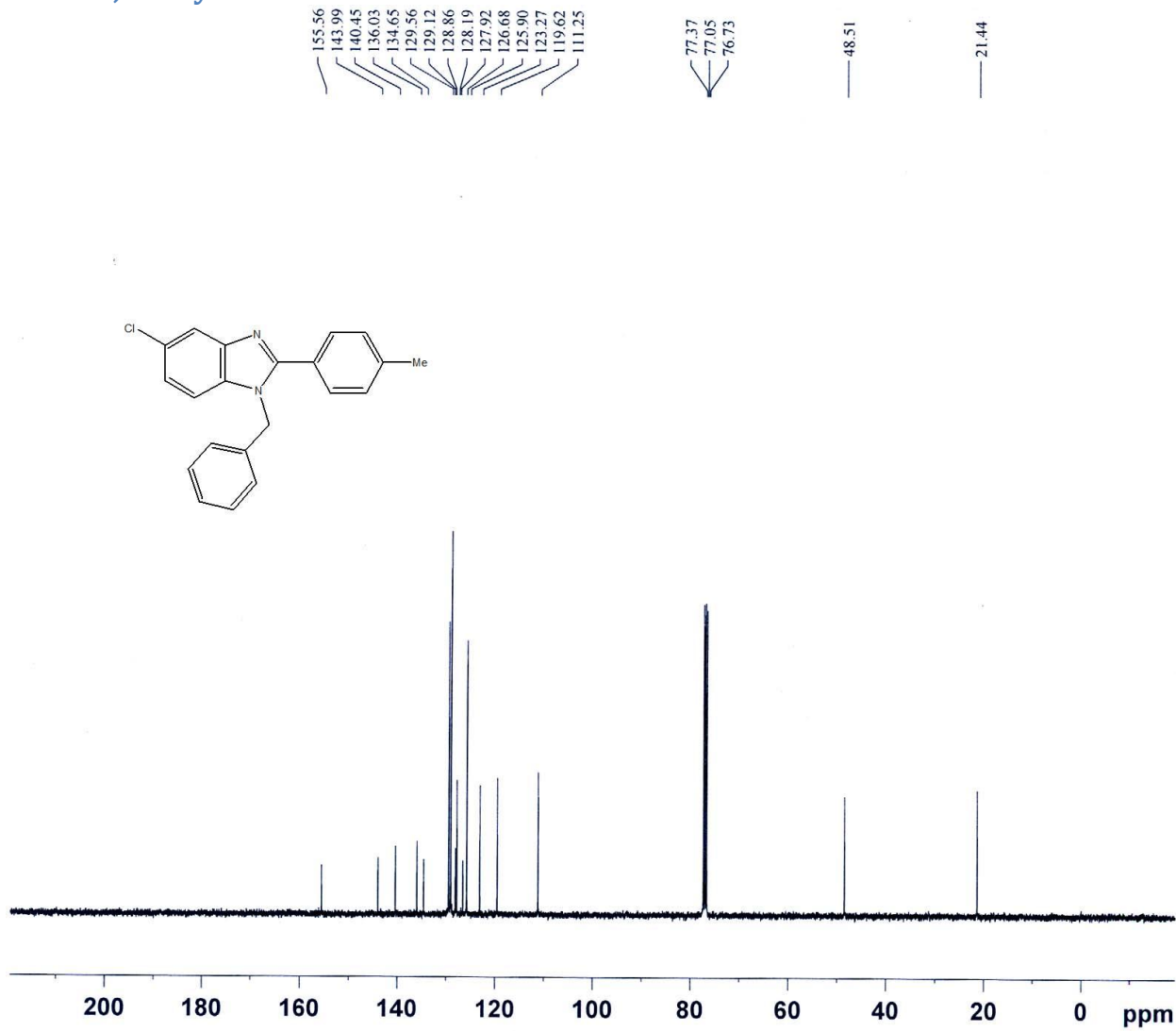


Table 5, Entry 17: ¹H NMR

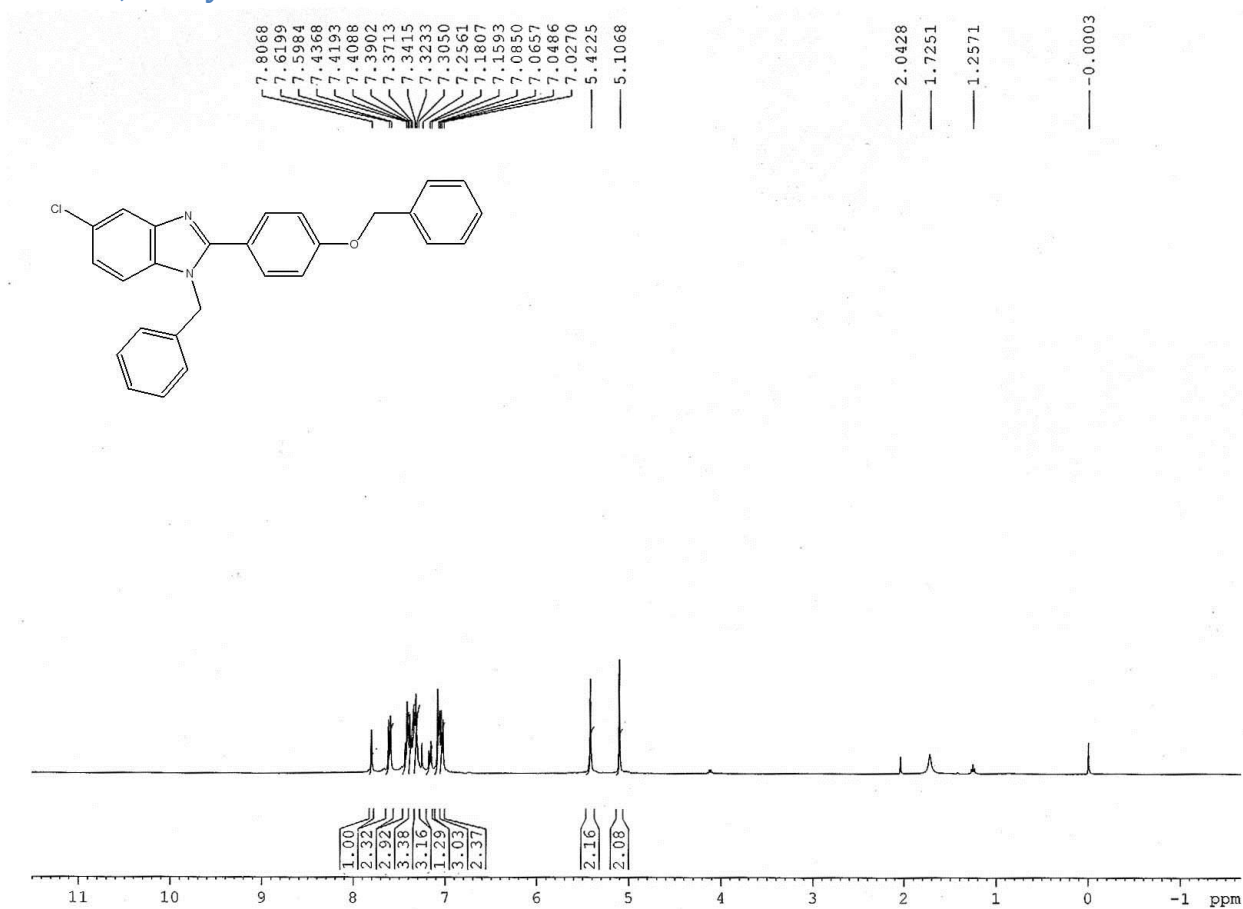


Table 5, Entry 17: ^{13}C NMR

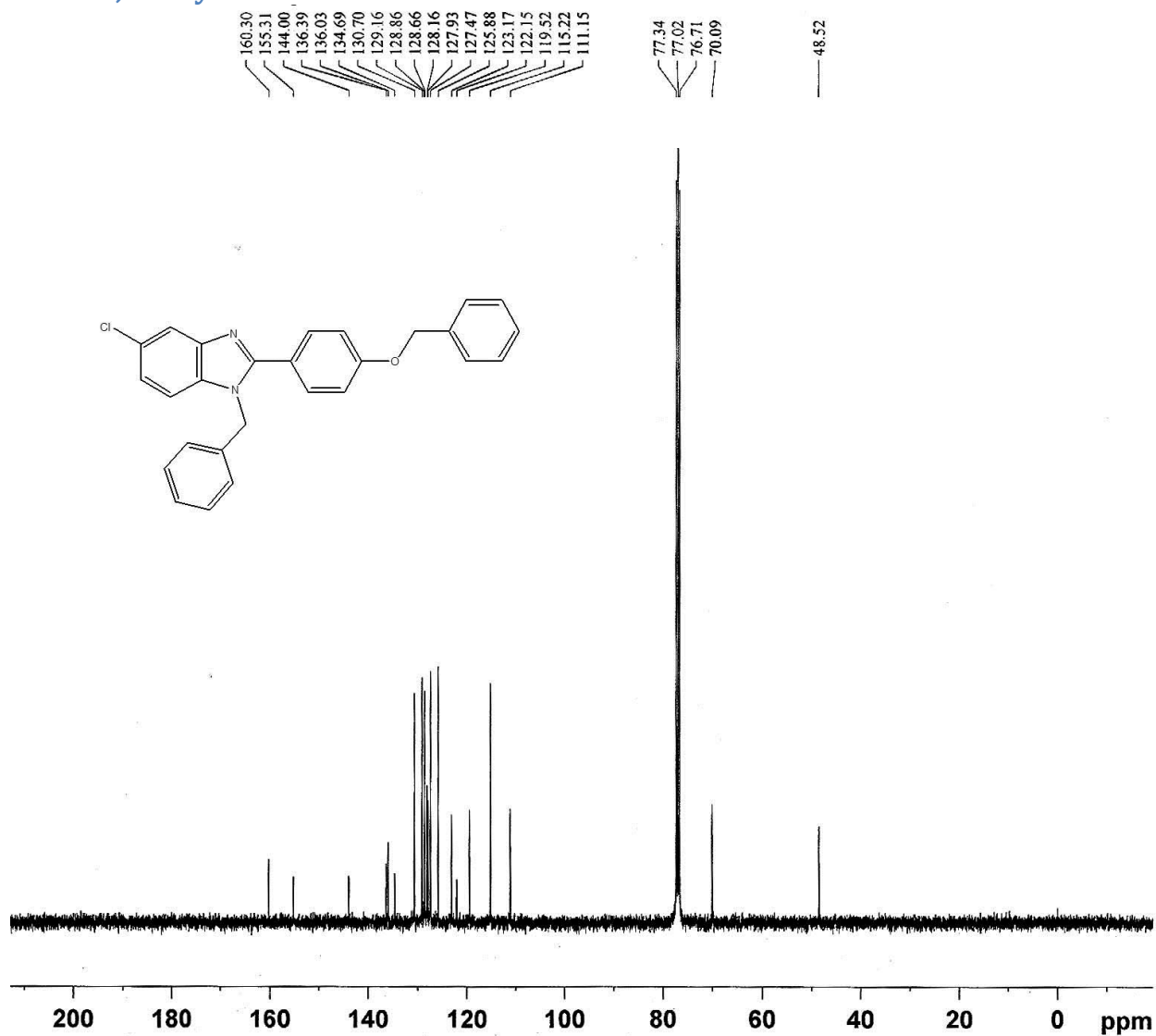


Table 5, Entry 18: ¹H NMR

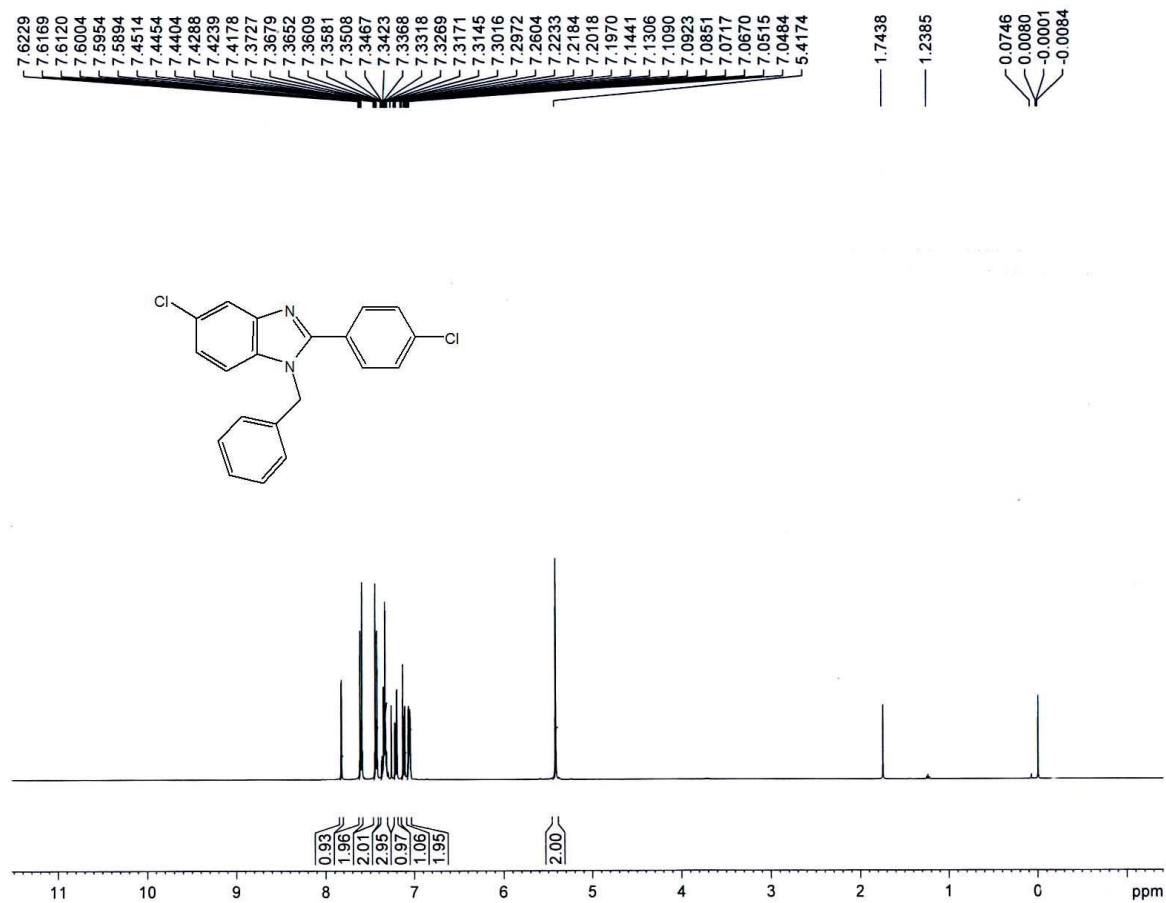


Table 5, Entry 18: ¹³C NMR

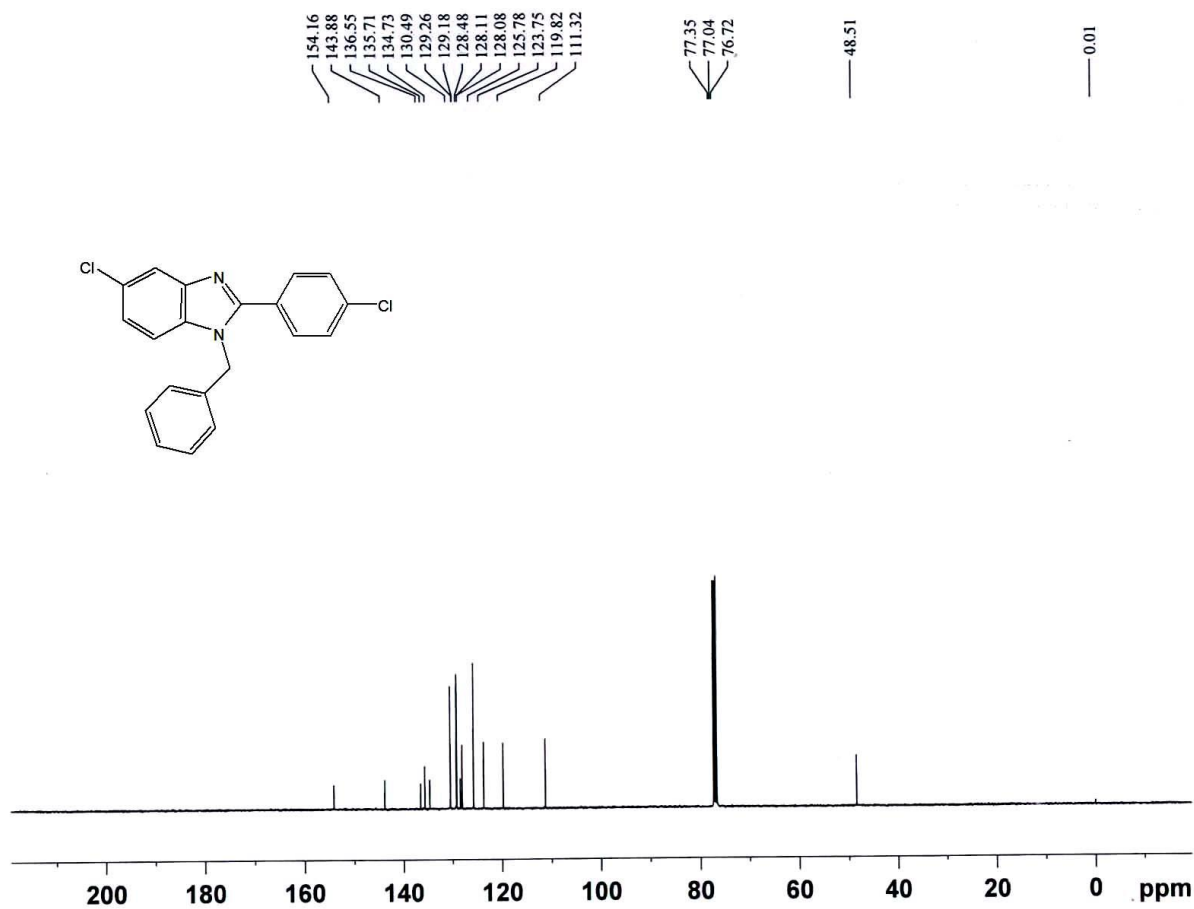


Table 5, Entry 19: ¹H NMR

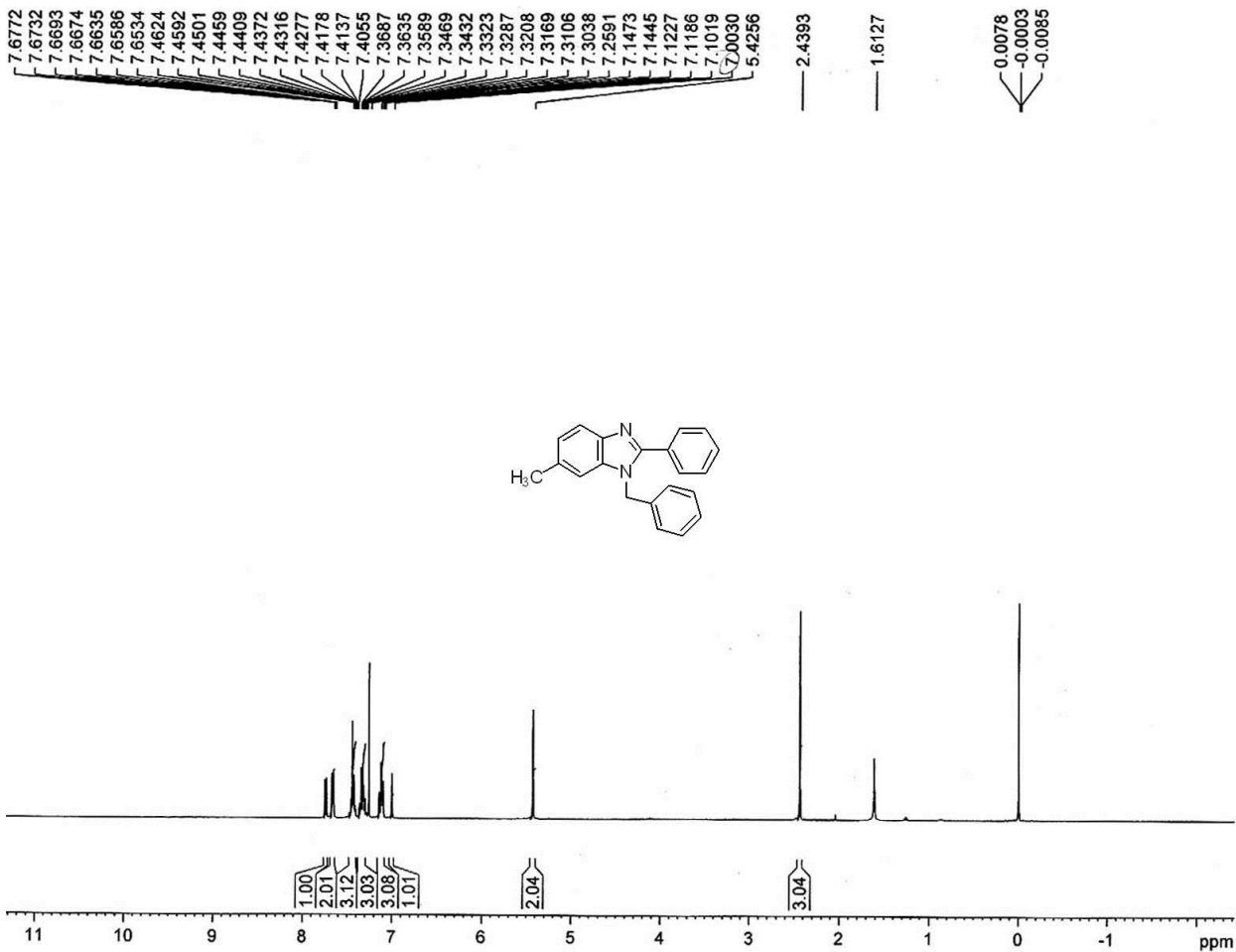


Table 5, Entry 19: ^{13}C NMR

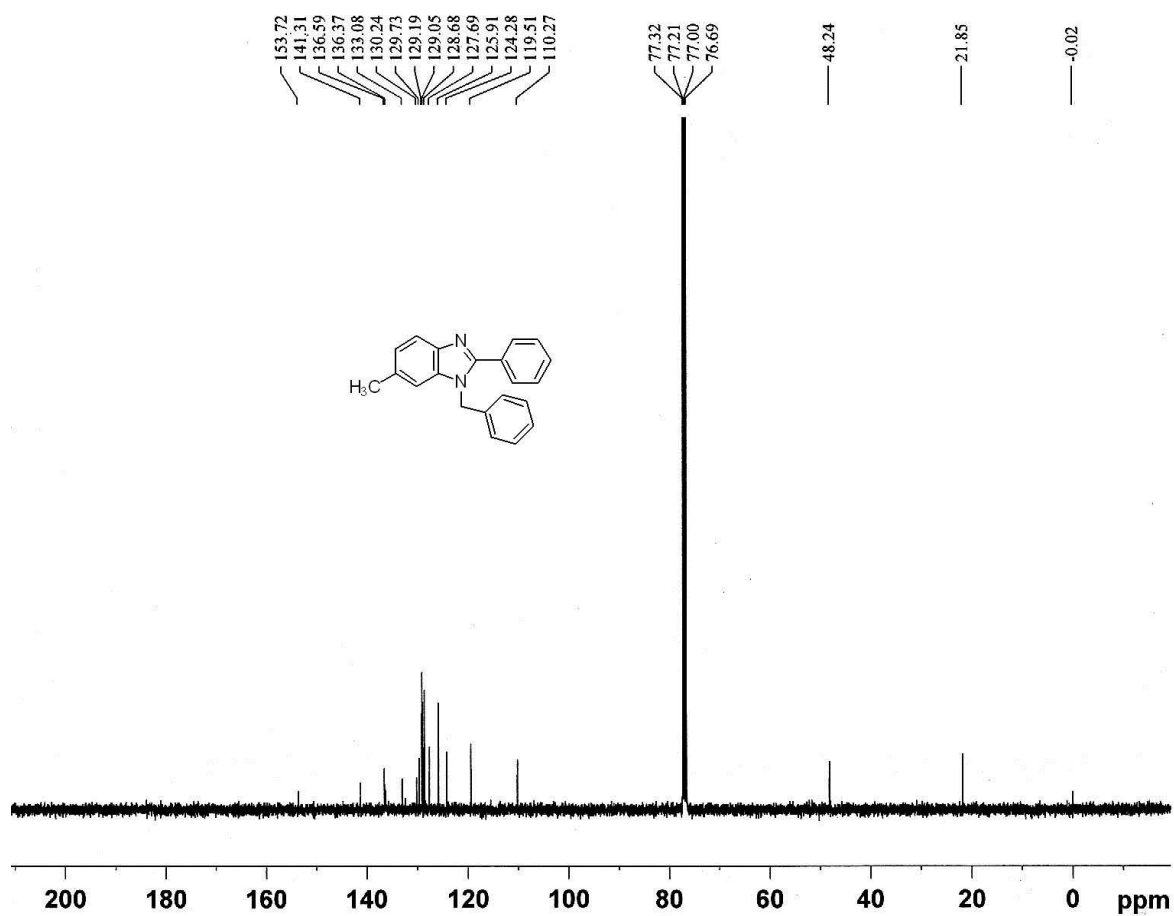


Table 5, Entry 20: ¹H NMR

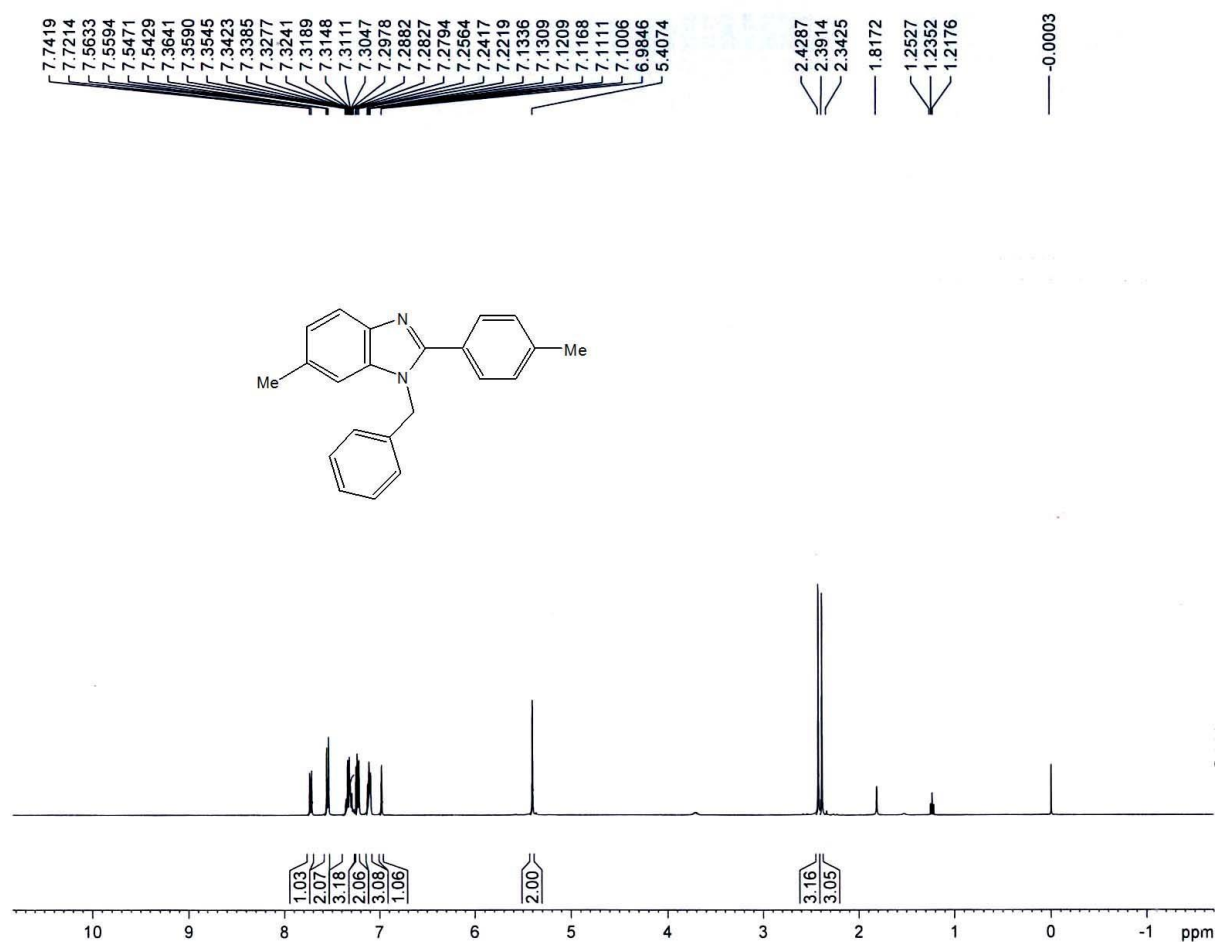


Table 5, Entry 20: ^{13}C NMR

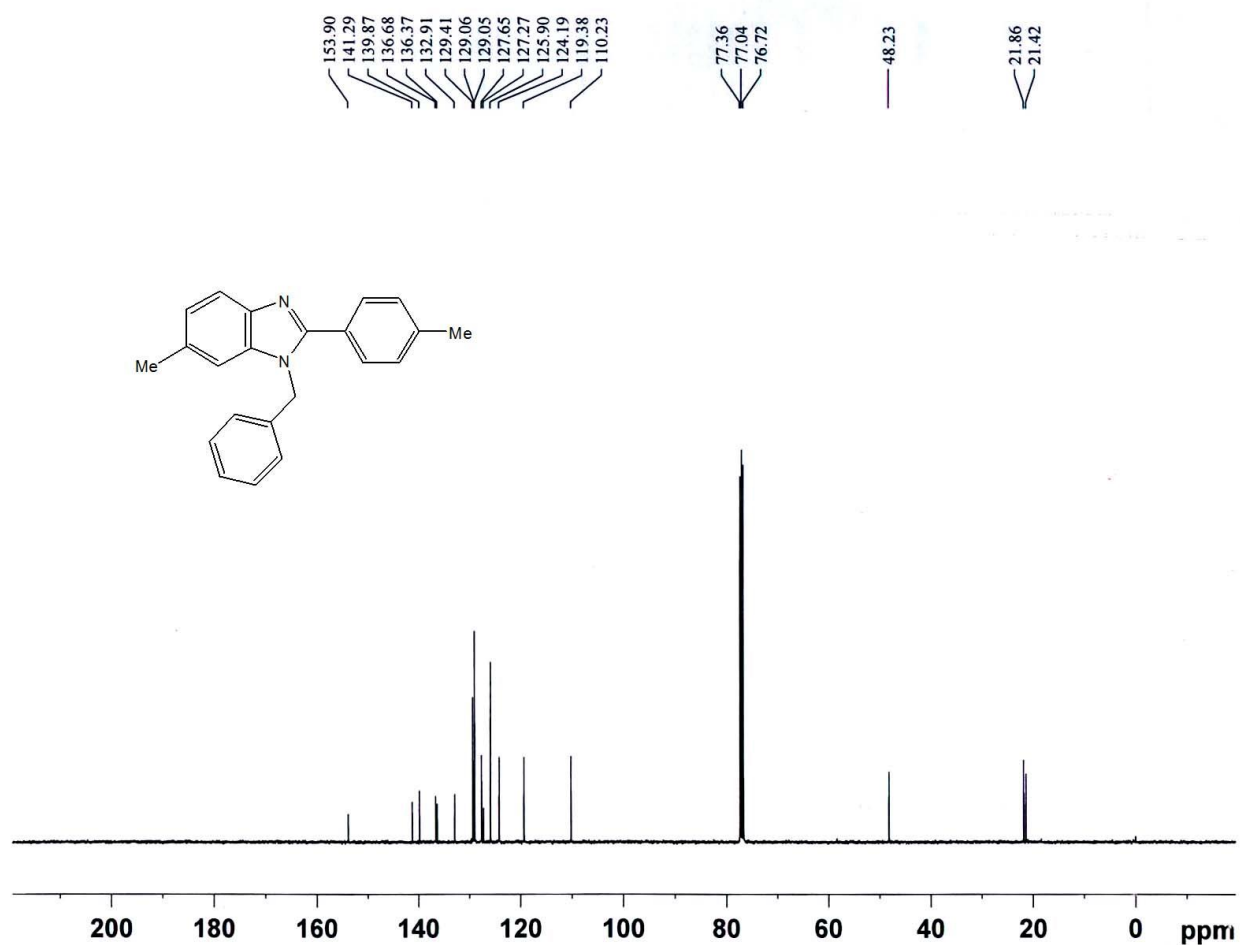


Table 5, Entry 21: ¹H NMR

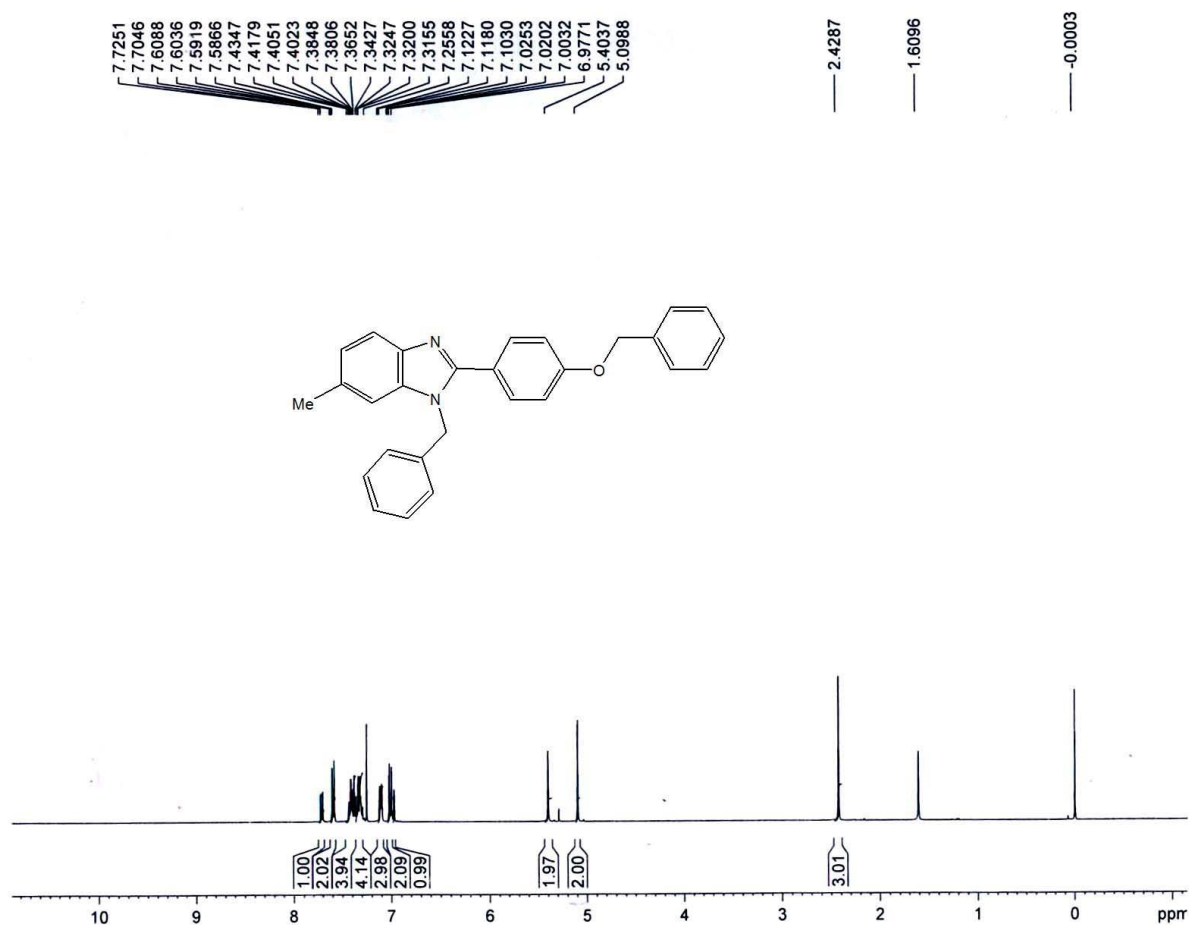


Table 5, Entry 21: ¹³C NMR

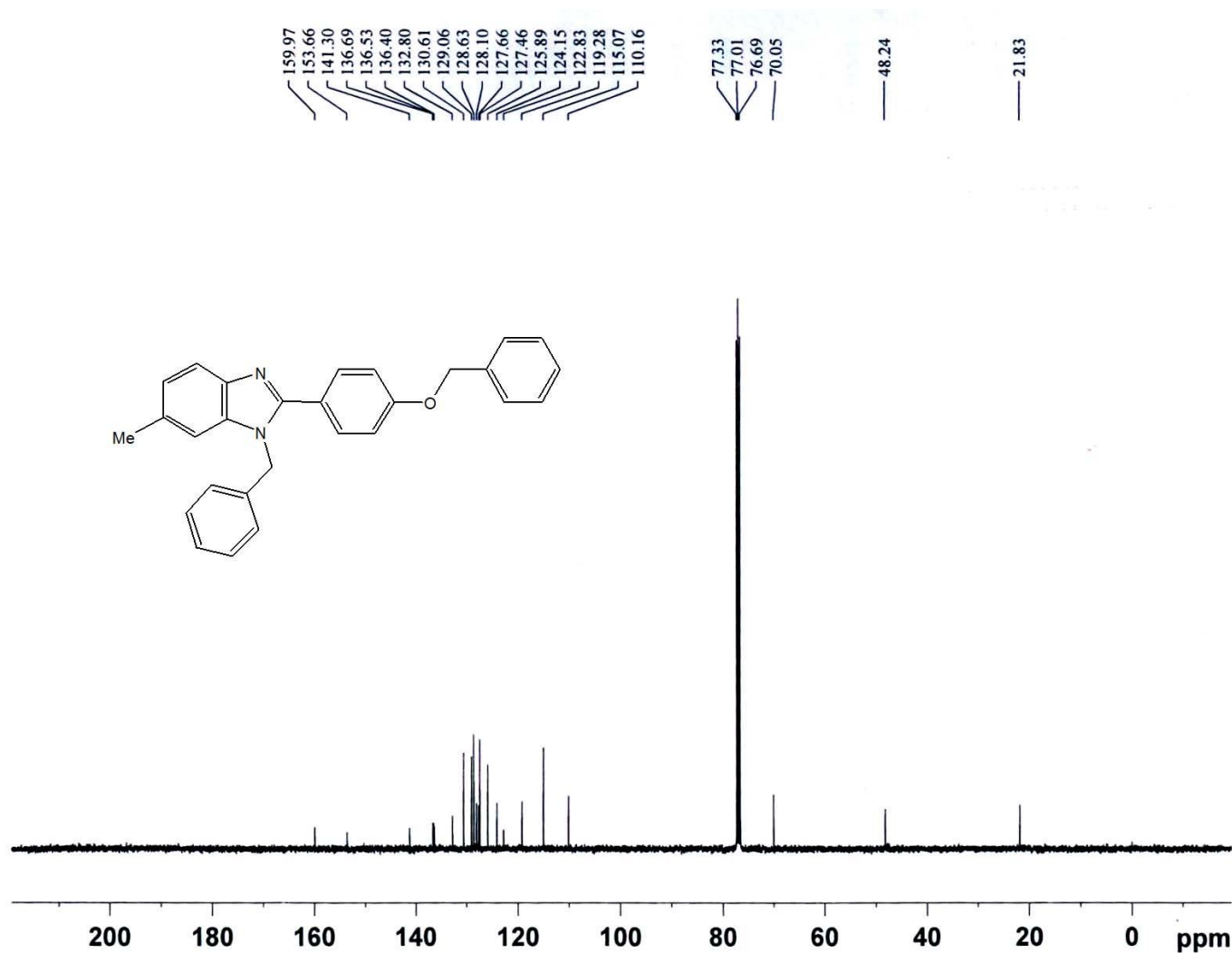


Table 5, Entry 22: ¹H NMR

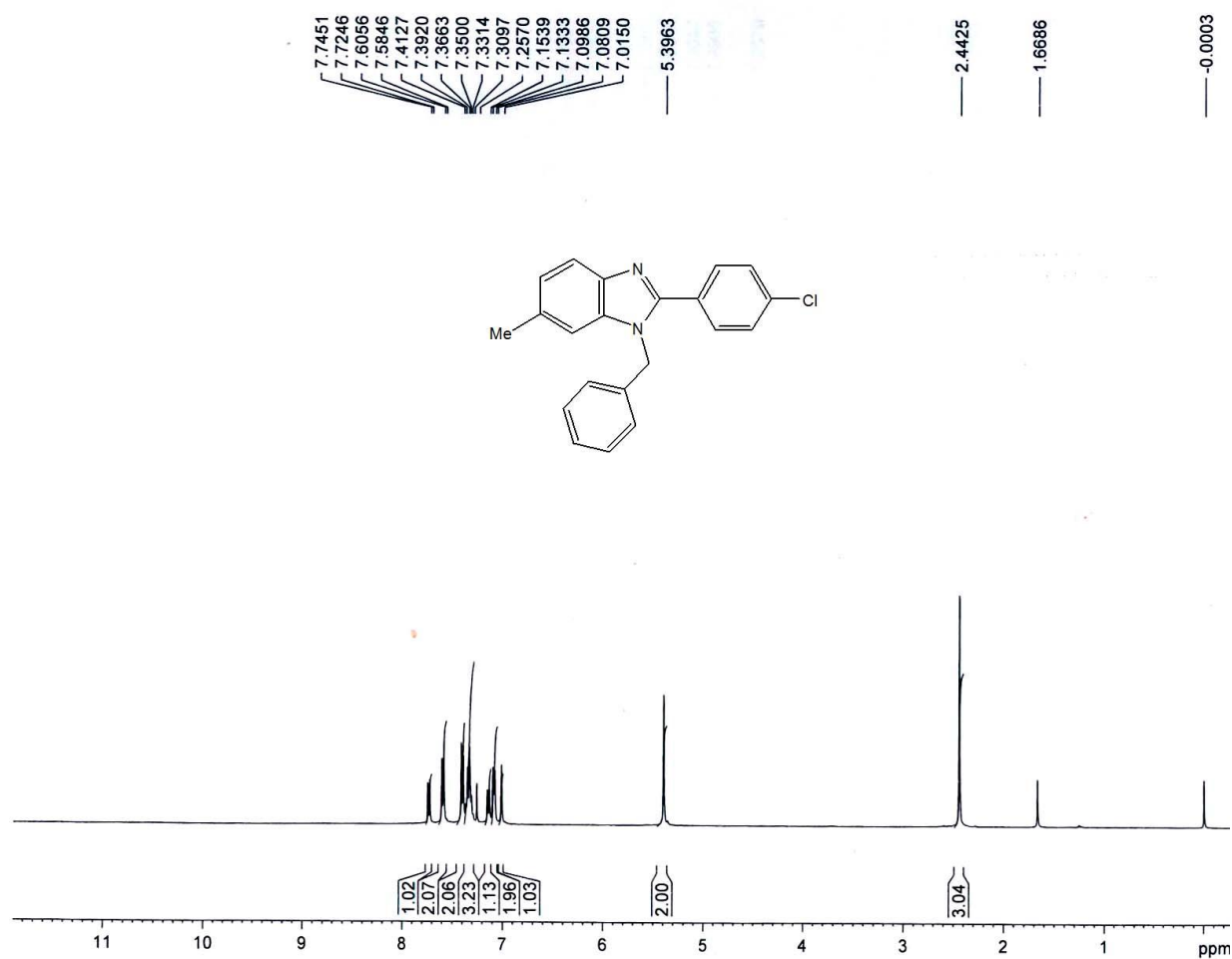


Table 5, Entry 22: ¹³C NMR

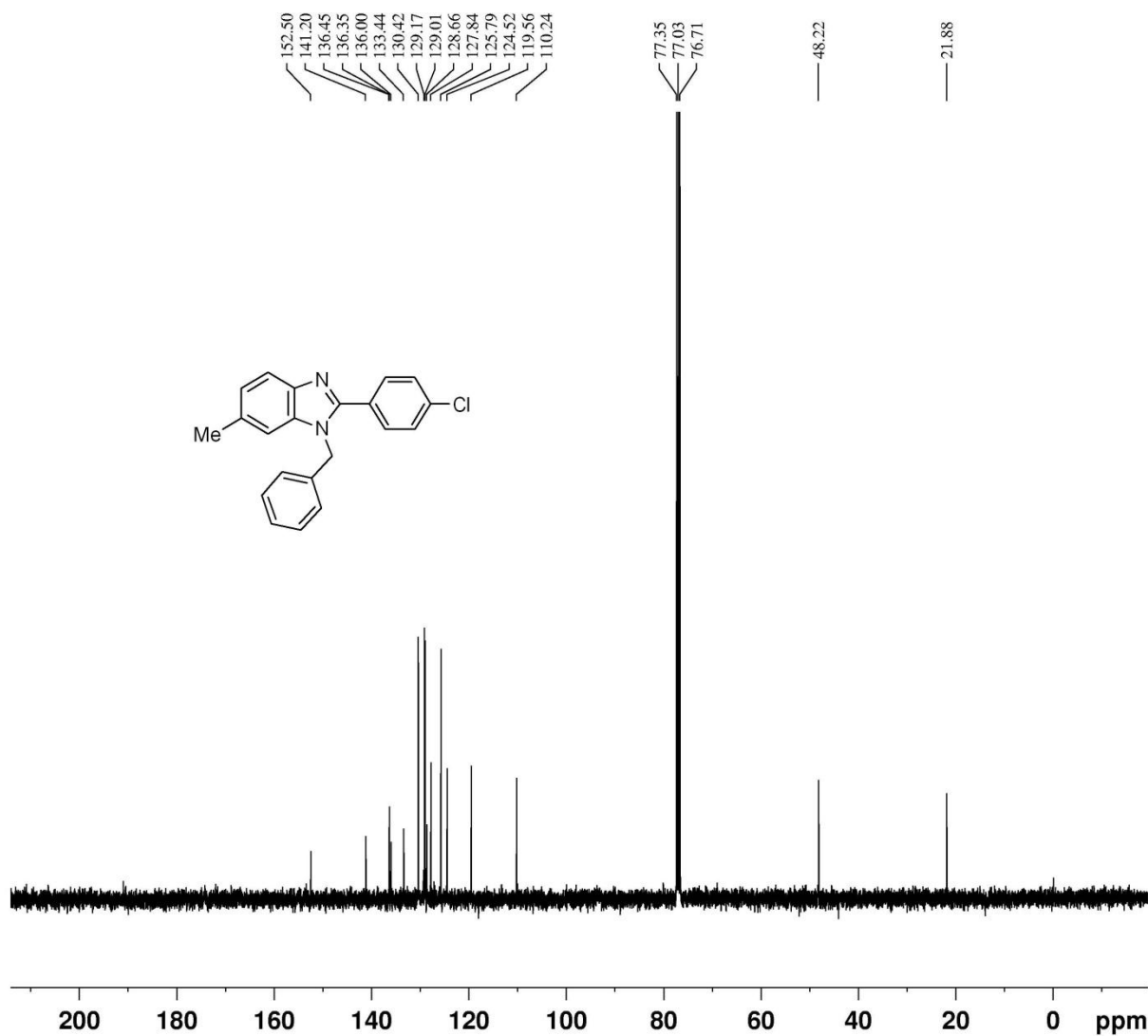


Table 5, Entry 23: ¹H NMR

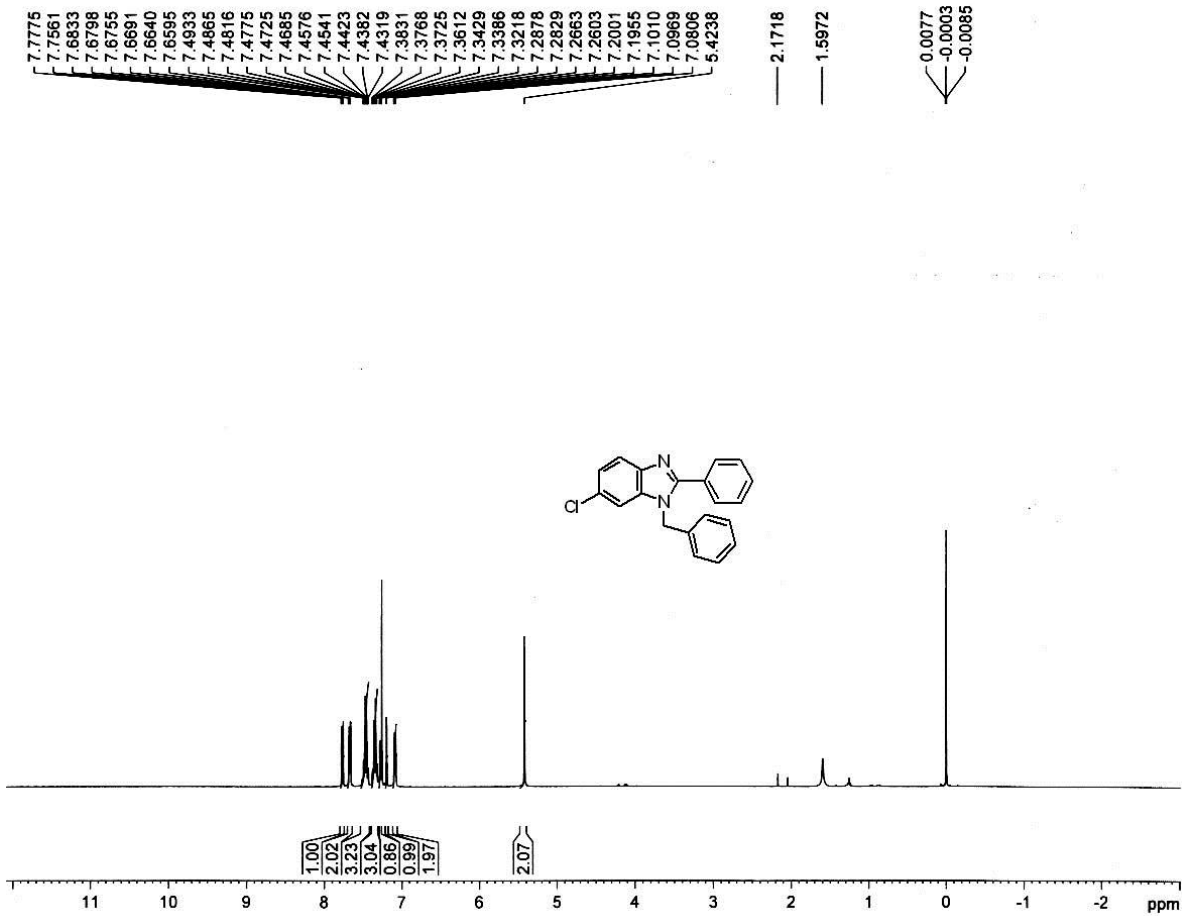


Table 5, Entry 23: ¹³C NMR

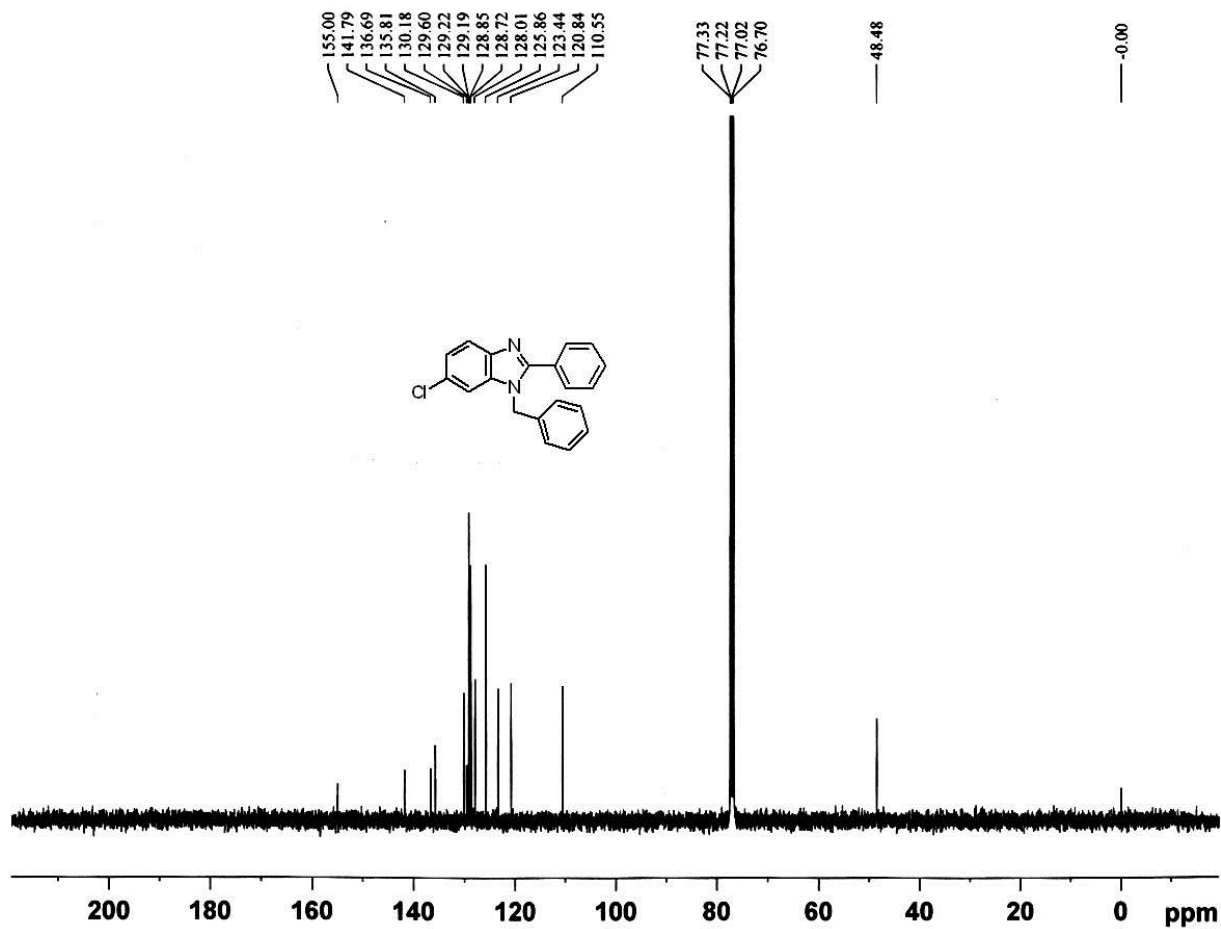


Table 5, Entry 24: ¹H NMR

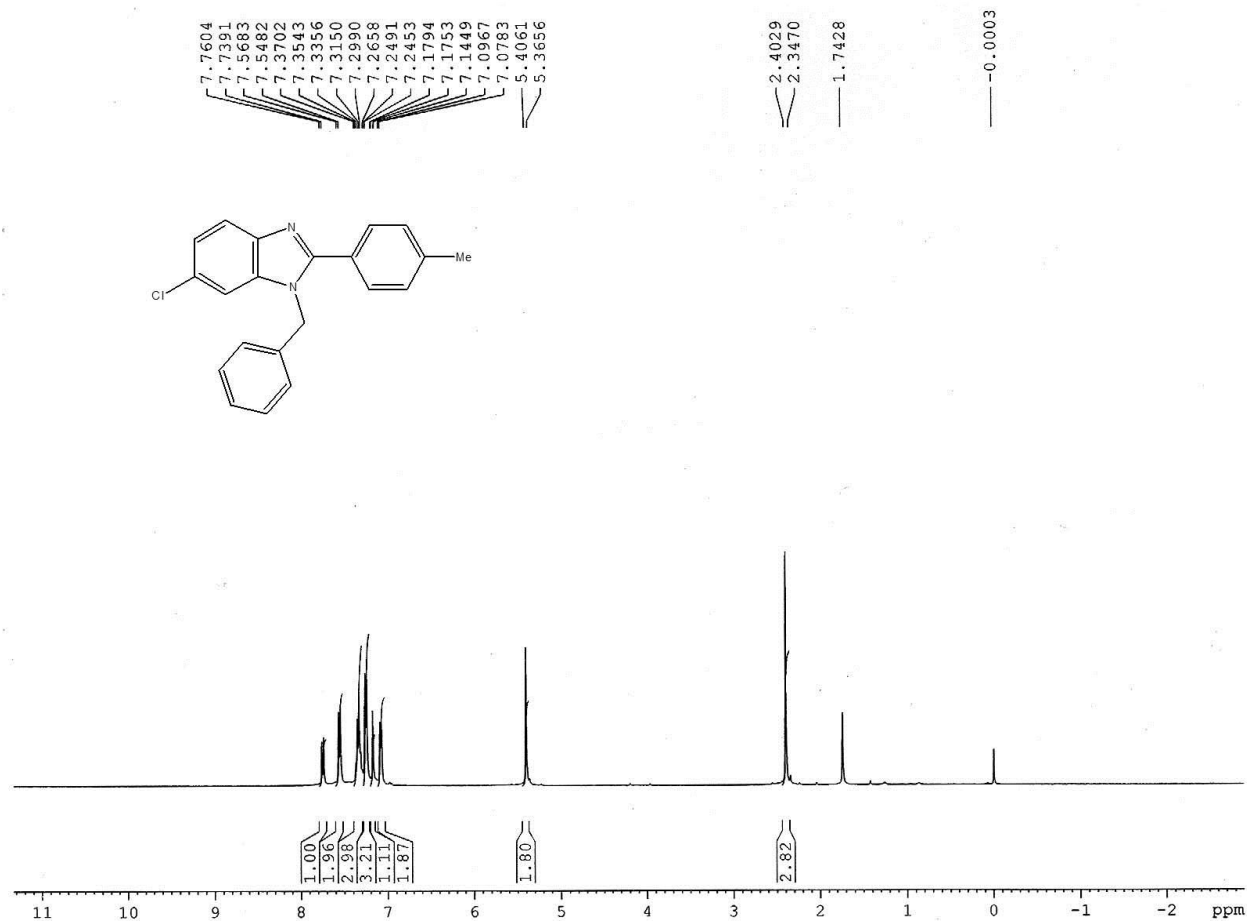


Table 5, Entry 24: ^{13}C NMR

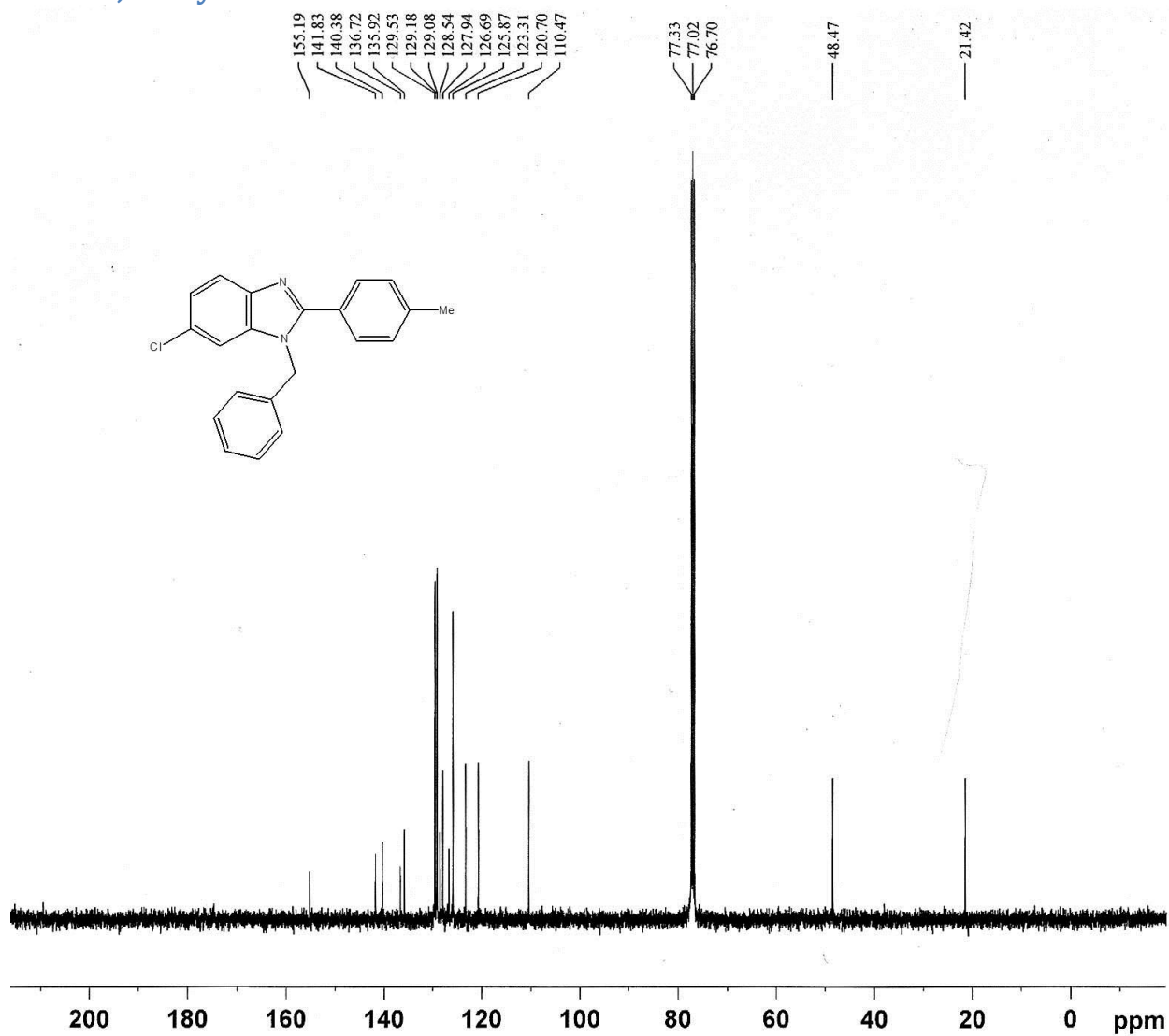


Table 5, Entry 25: ^1H NMR

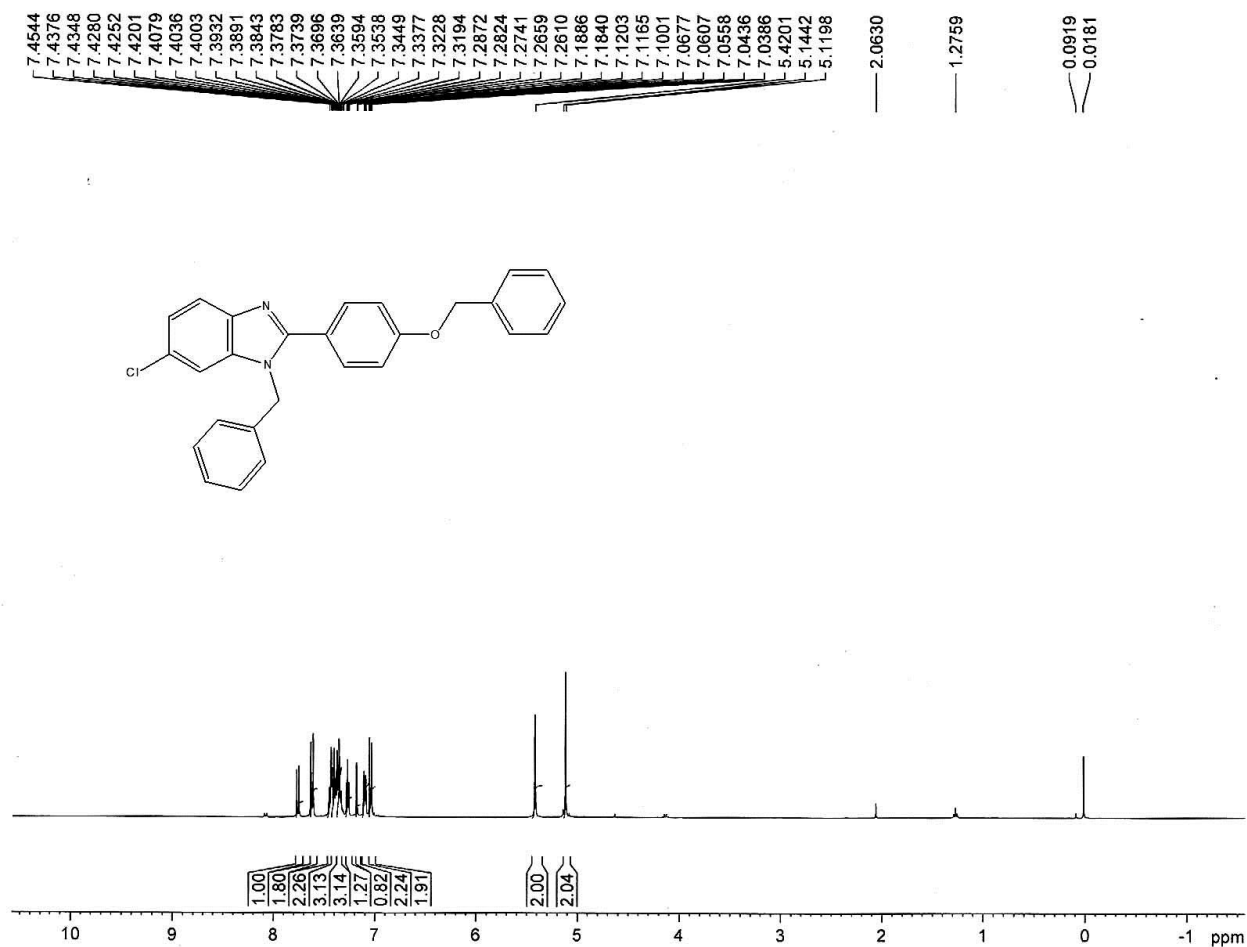


Table 5, Entry 25: ^{13}C NMR

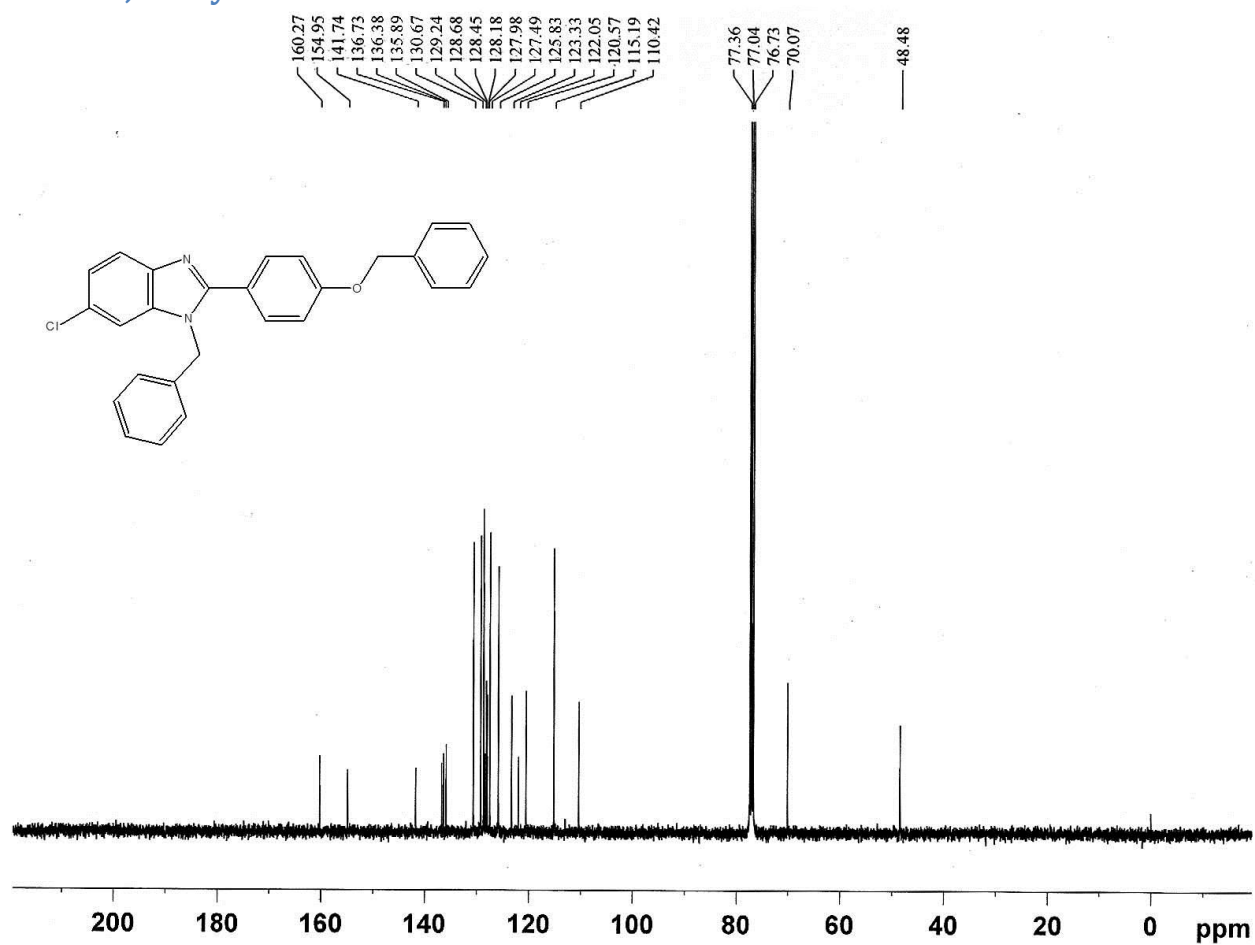


Table 5, Entry 26: ¹H NMR

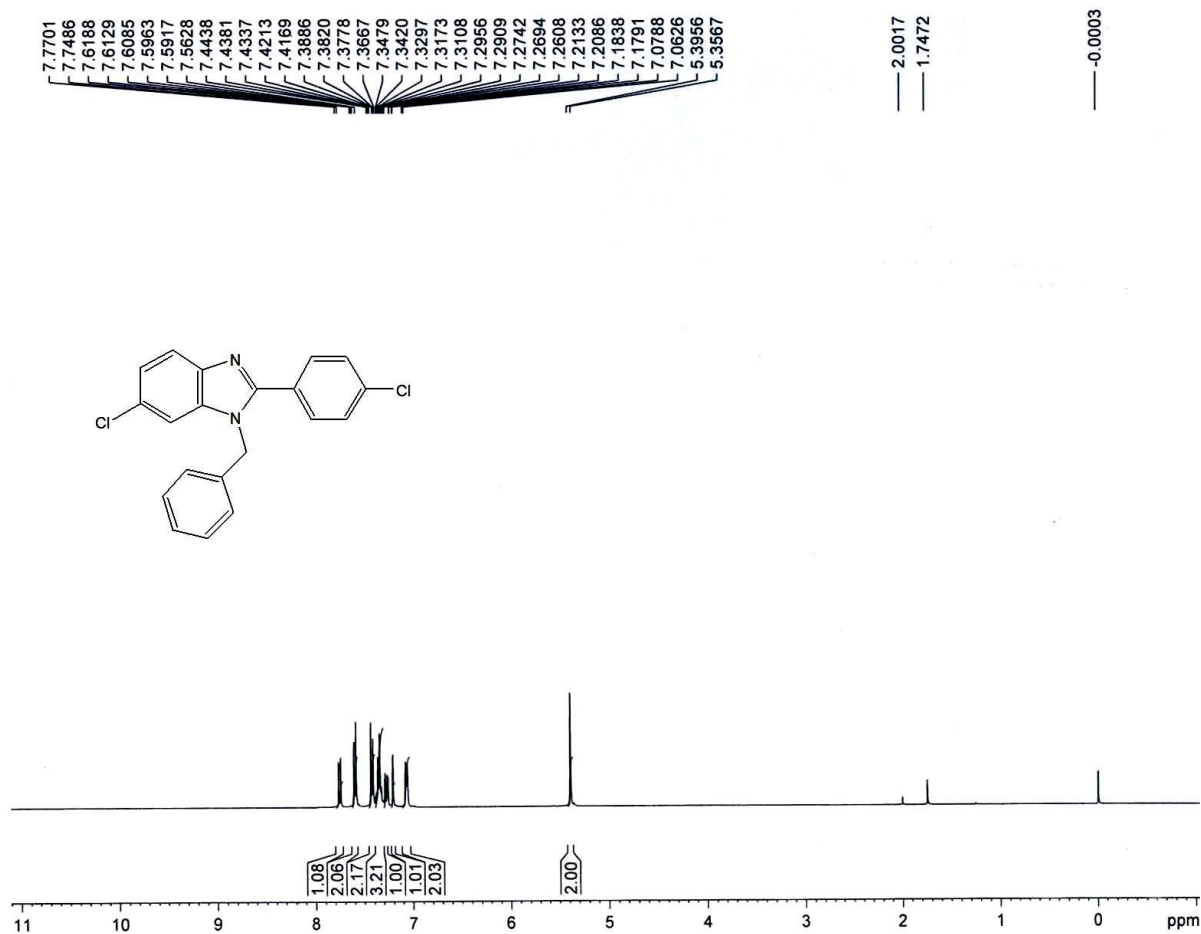


Table 5, Entry 26: ¹³C NMR

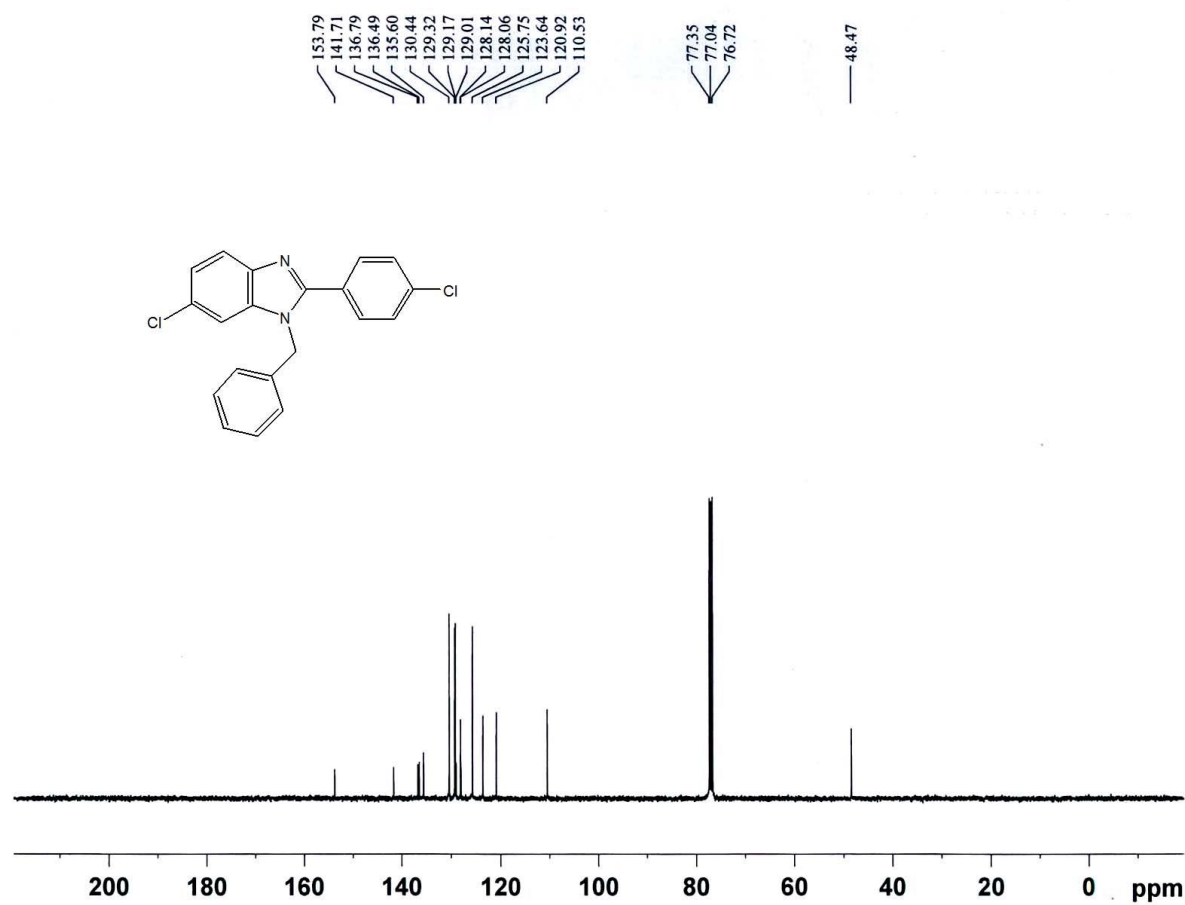


Table 5, Entry 27: ^1H NMR

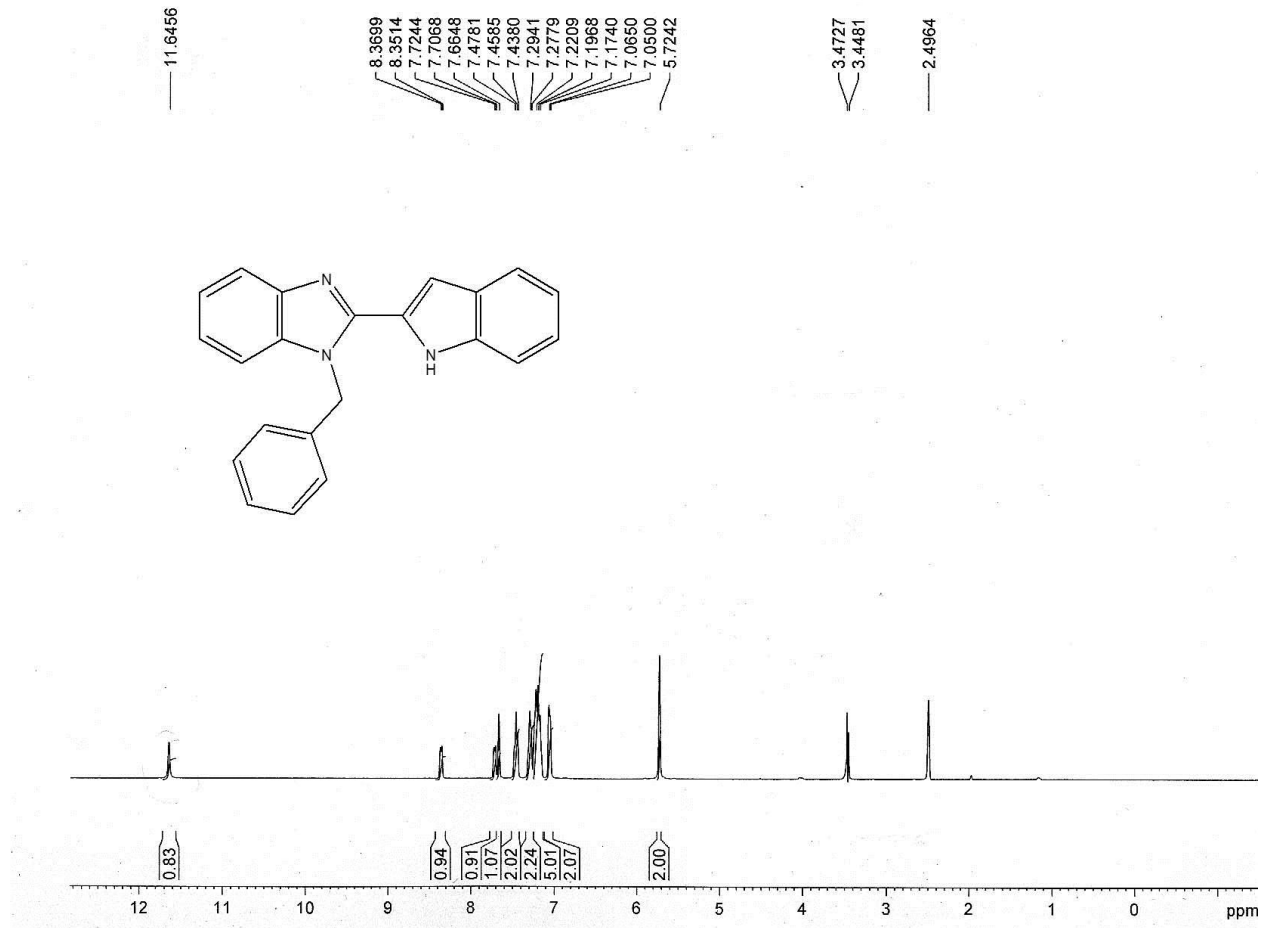


Table 5, Entry 27: ¹³C NMR

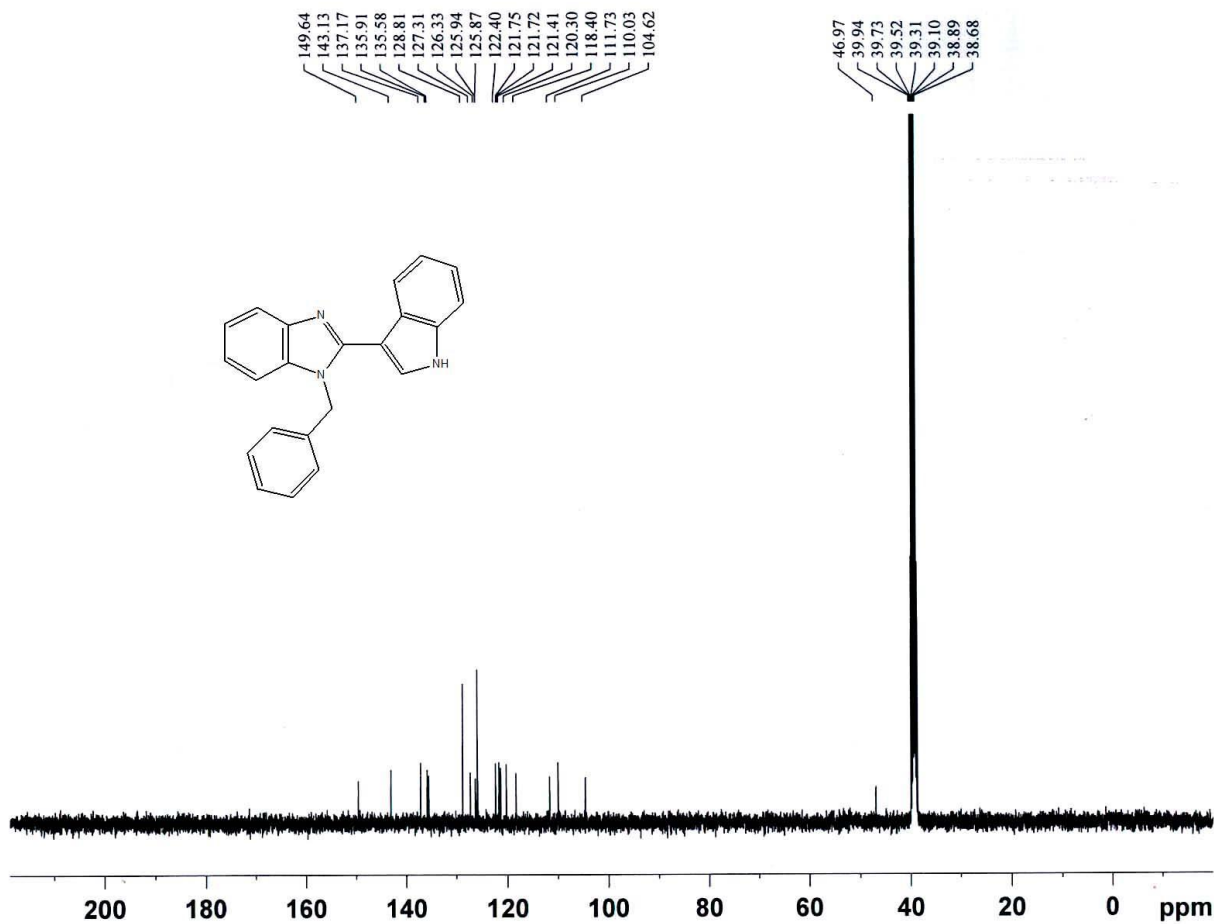


Table 5, Entry 28: ¹H NMR

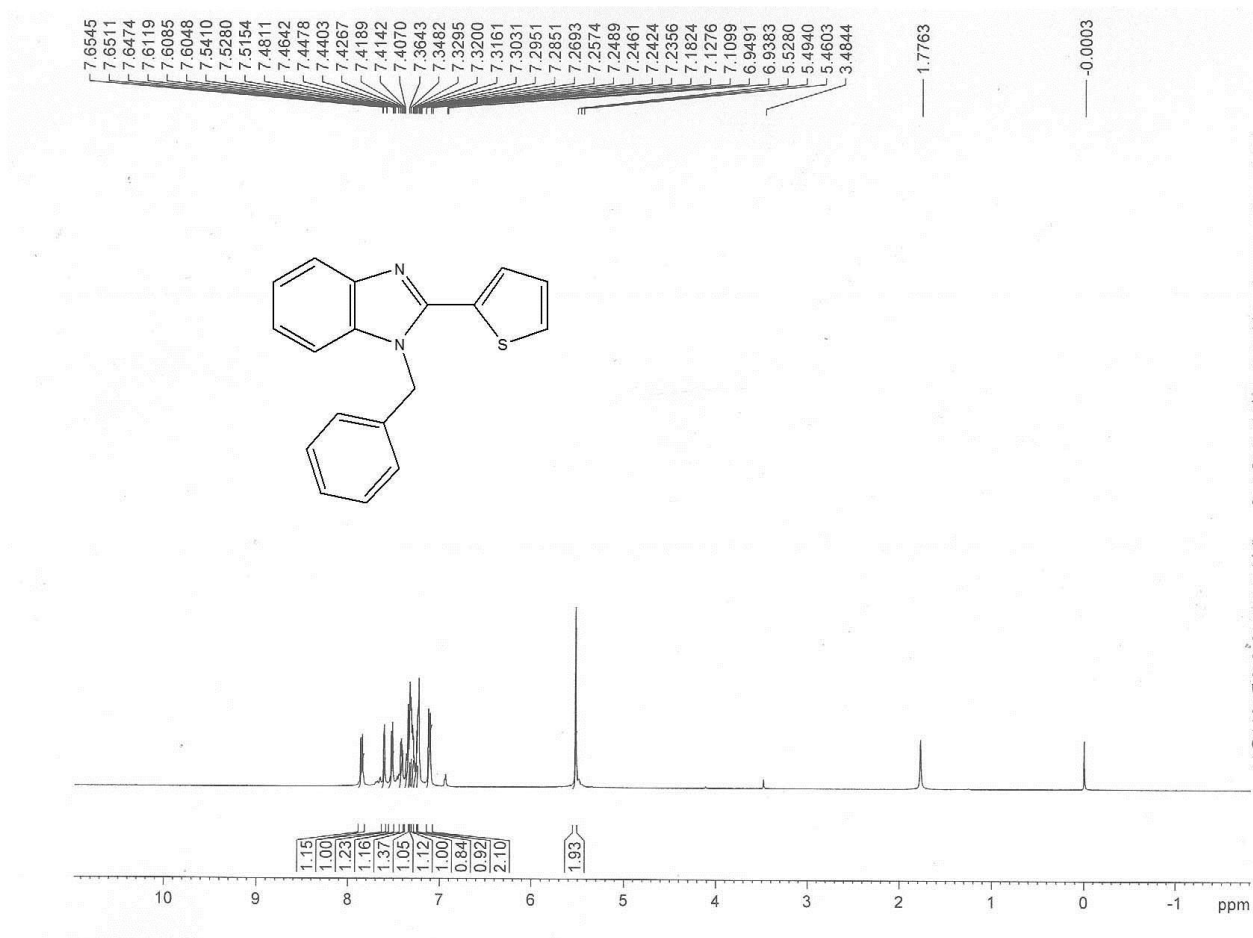


Table 5, Entry 28: ^{13}C NMR

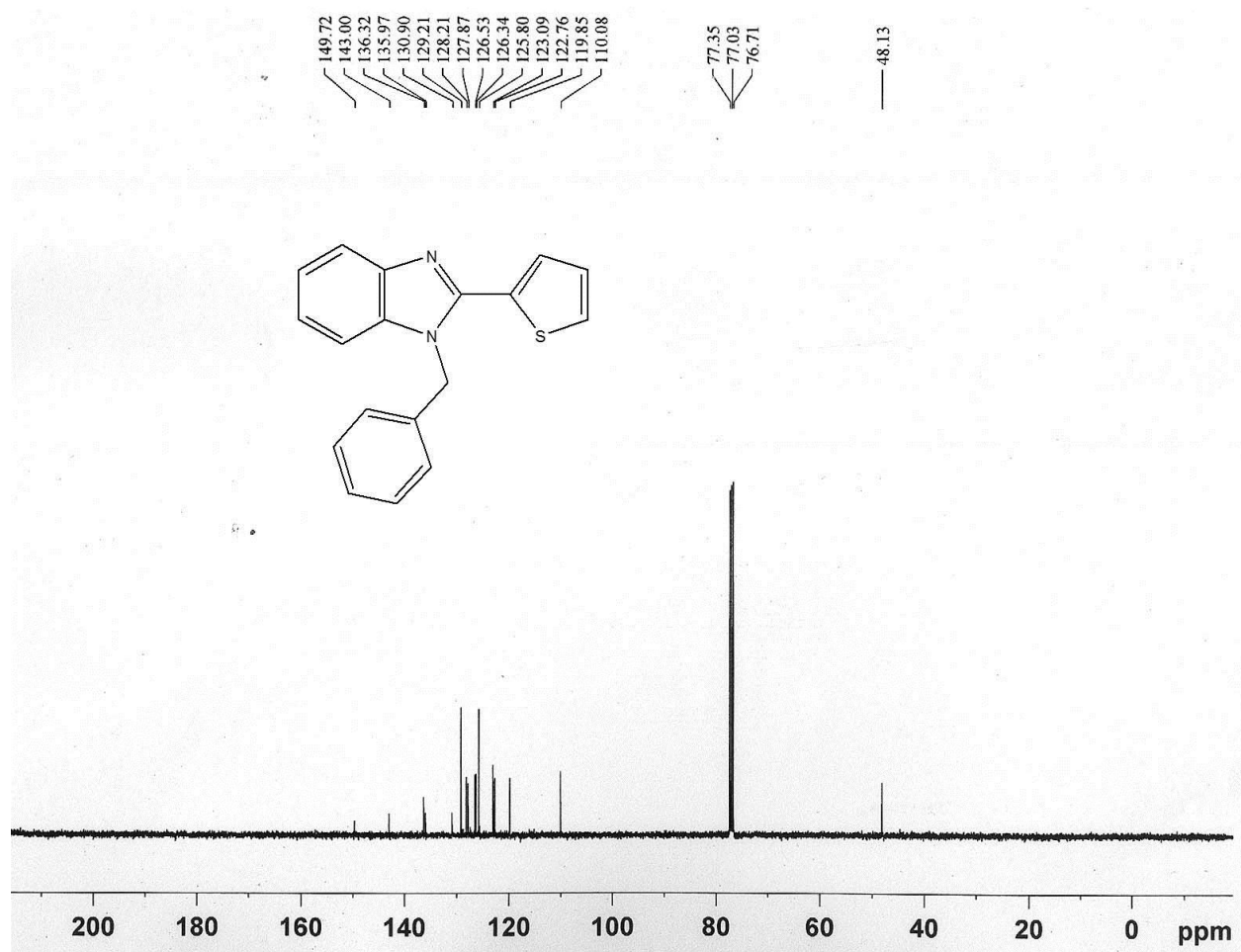


Table 5, Entry 29: ^1H NMR

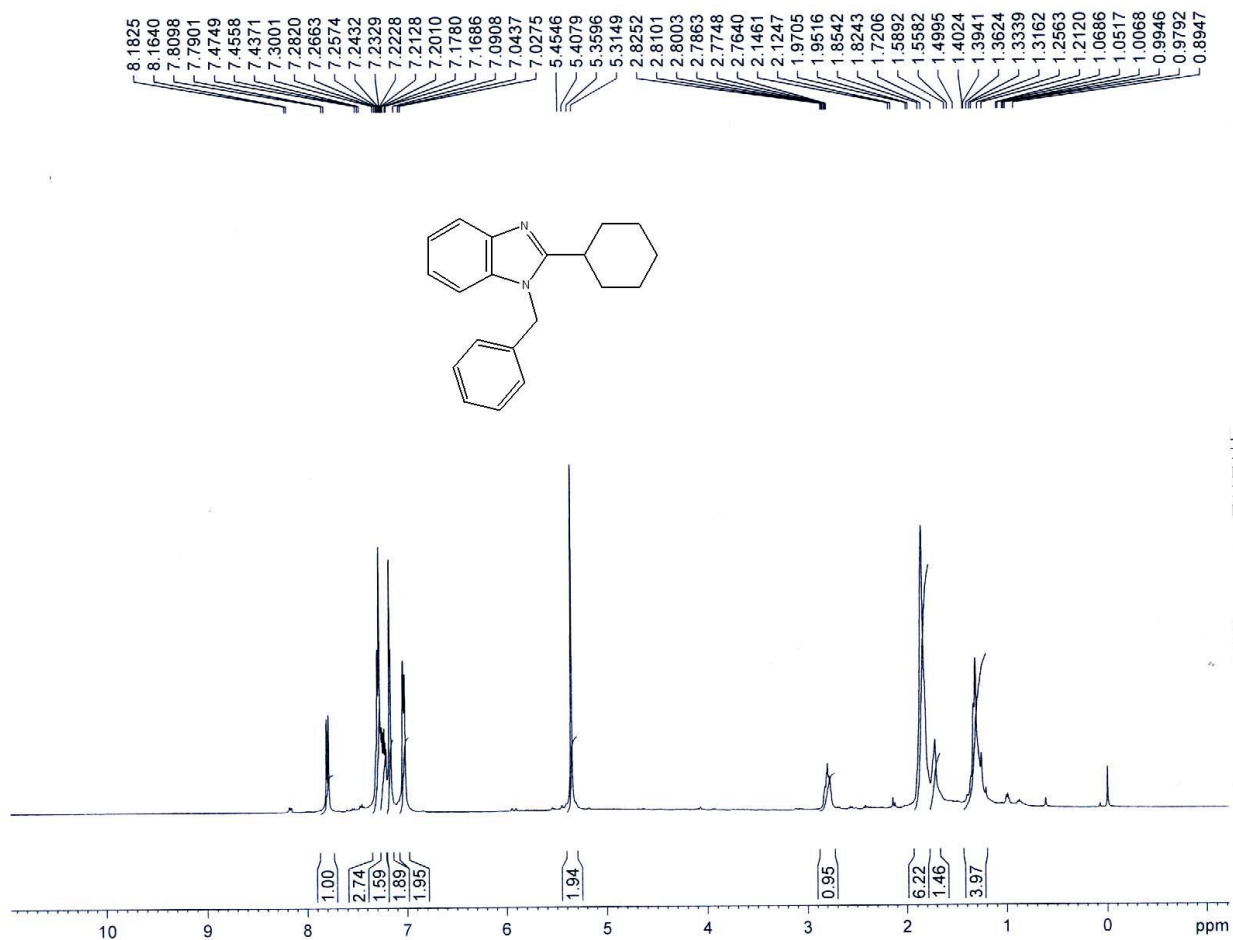


Table 5, Entry 29: ^{13}C NMR

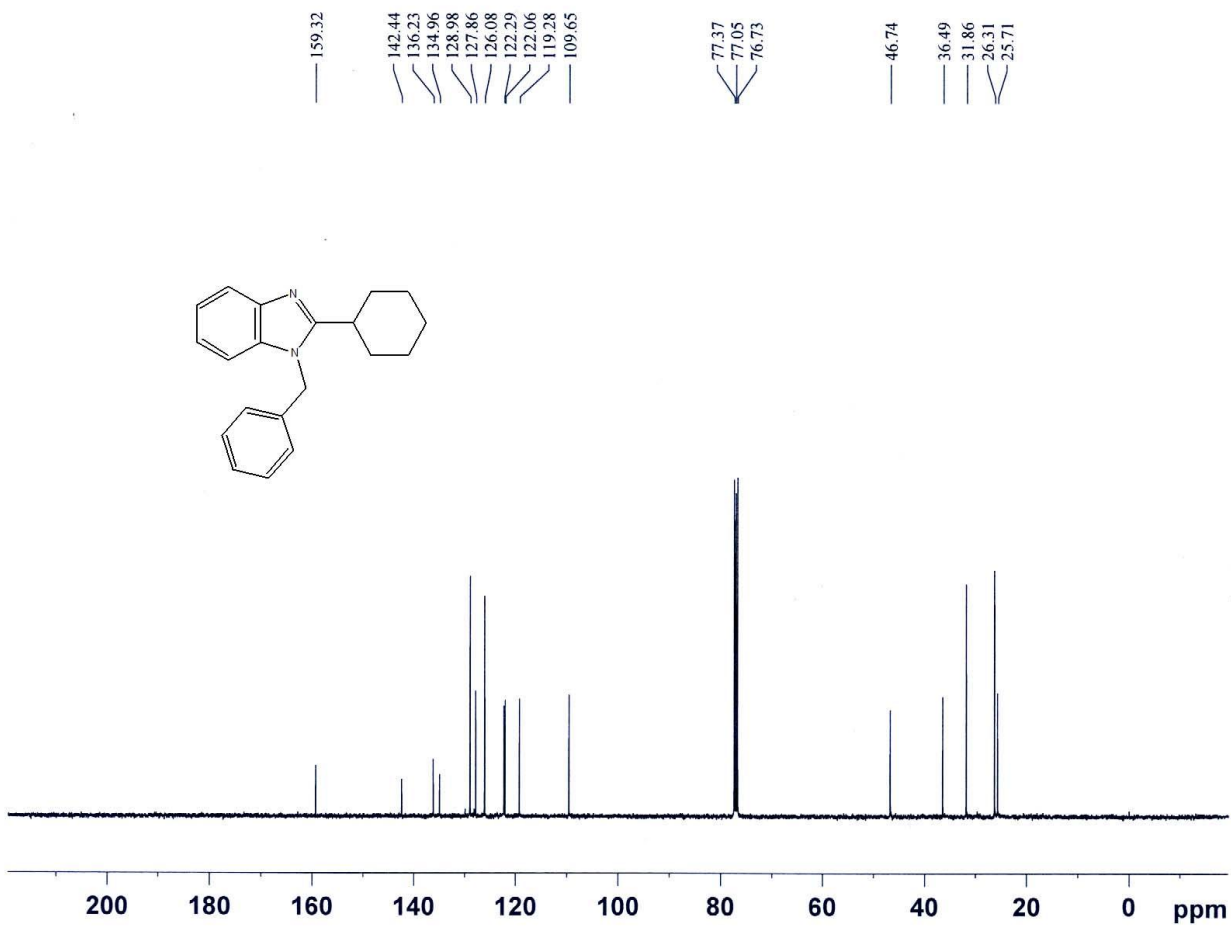


Table 5, Entry 30: ¹H NMR

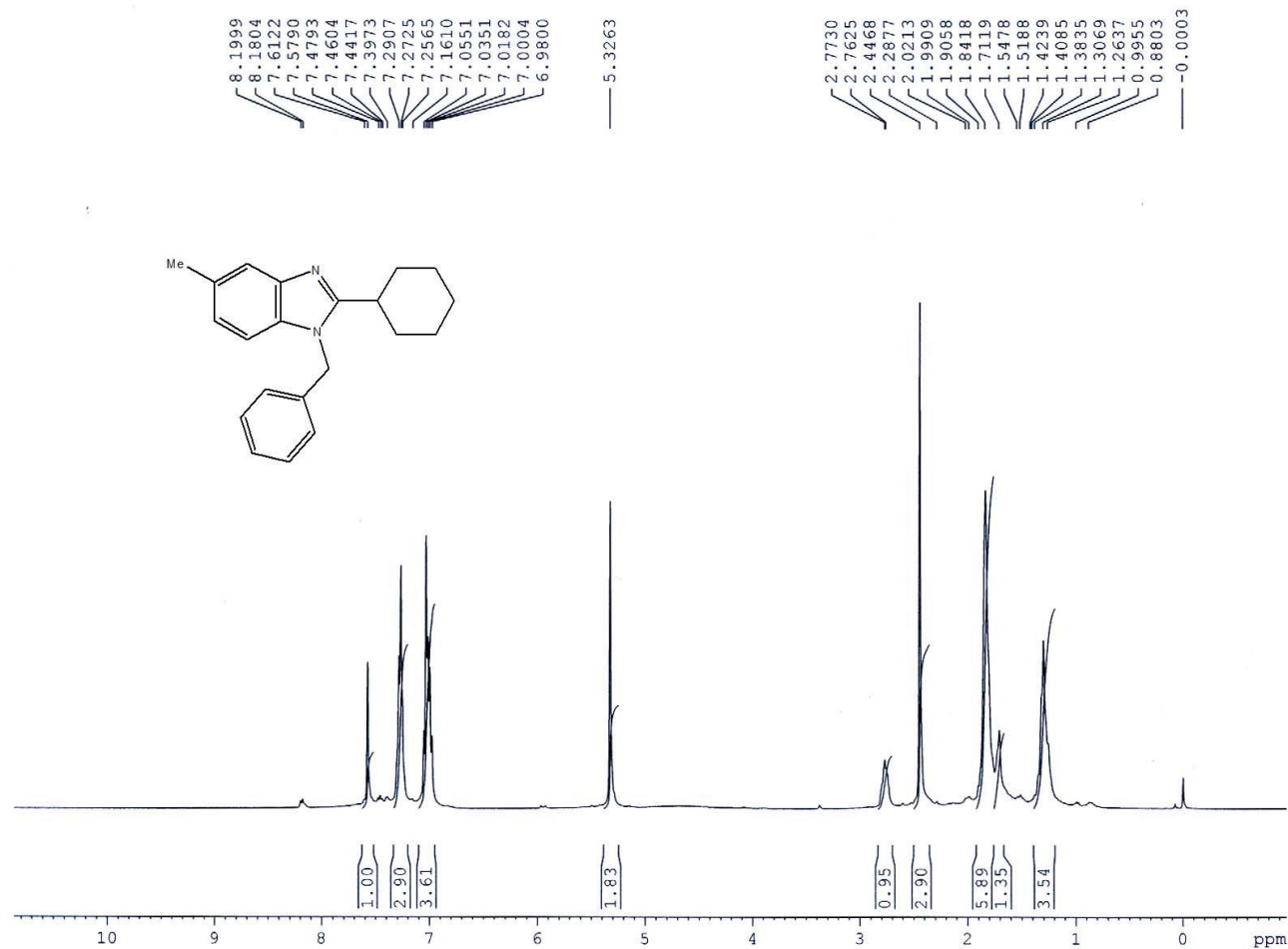


Table 5, Entry 30: ^{13}C NMR

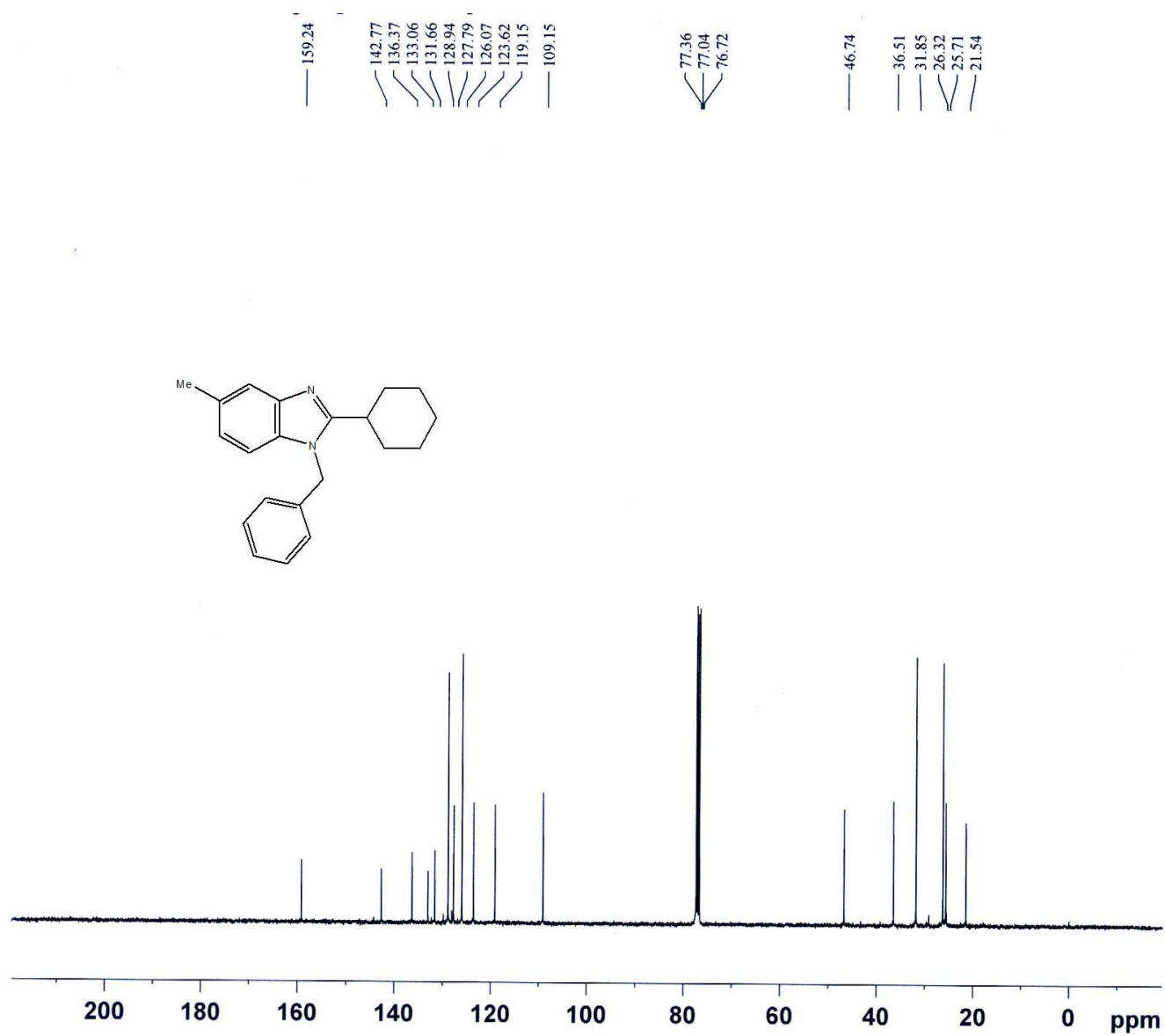


Table 5, Entry 31: ^1H NMR

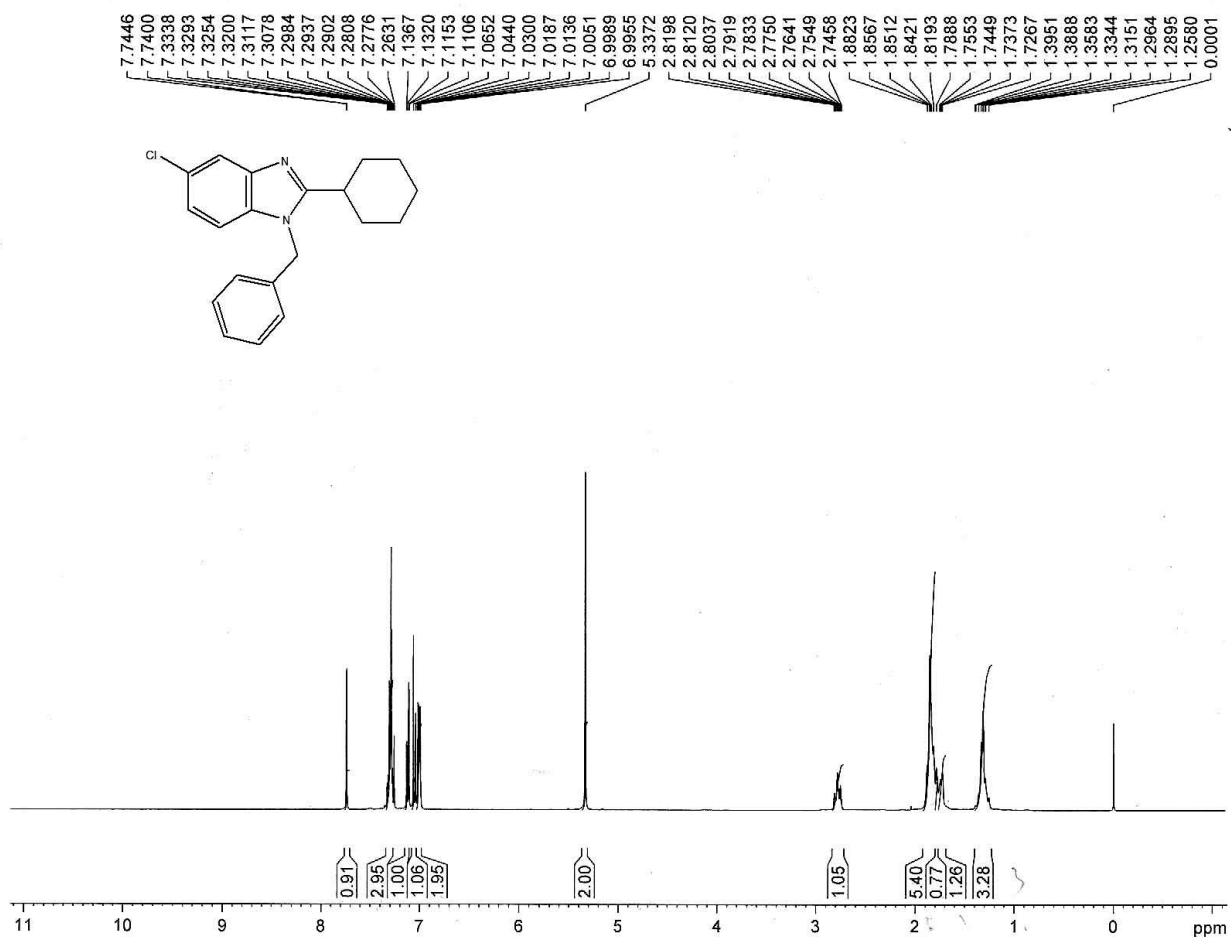


Table 5, Entry 31: ^{13}C NMR

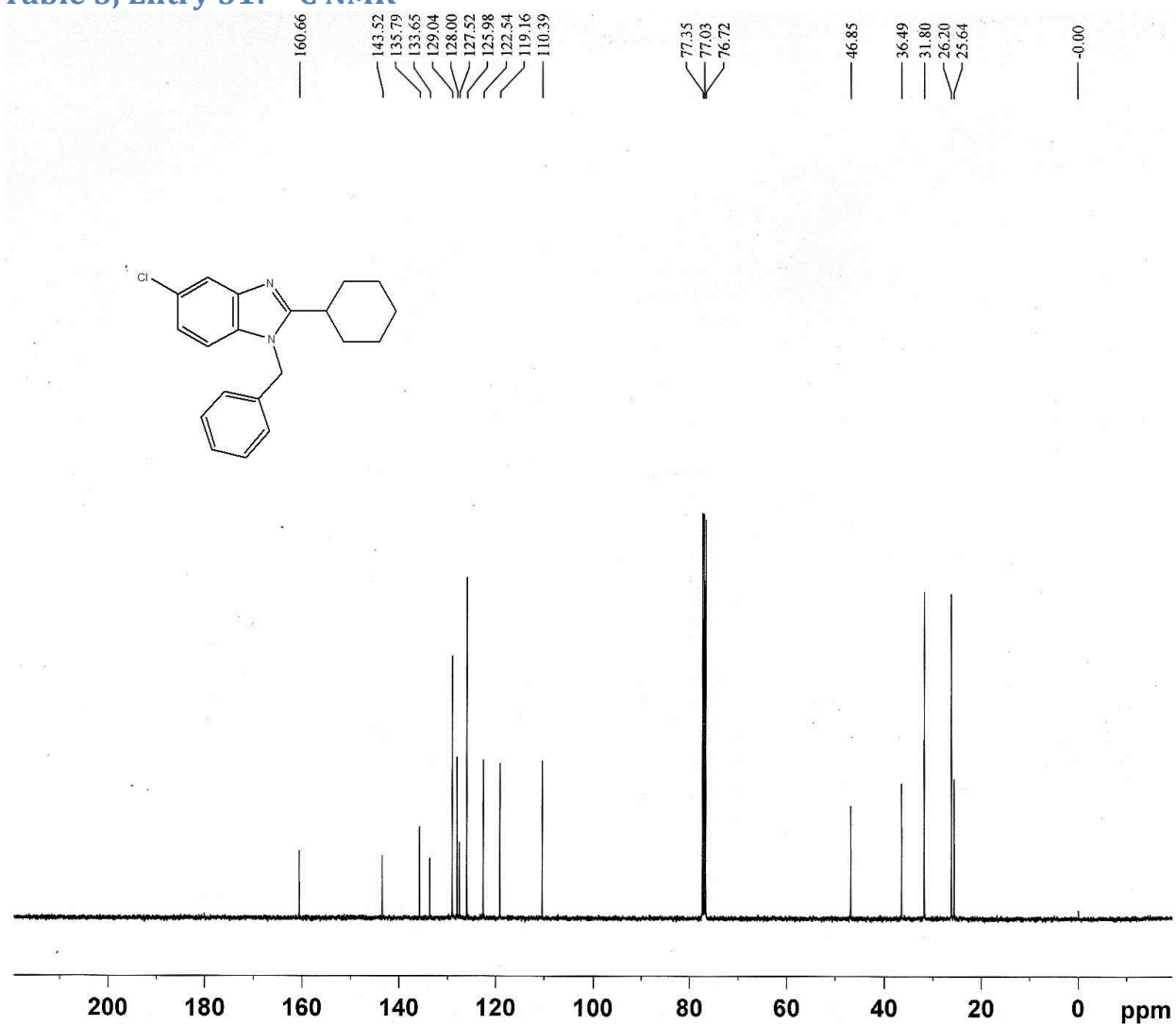


Table 5, Entry 32: ¹H NMR

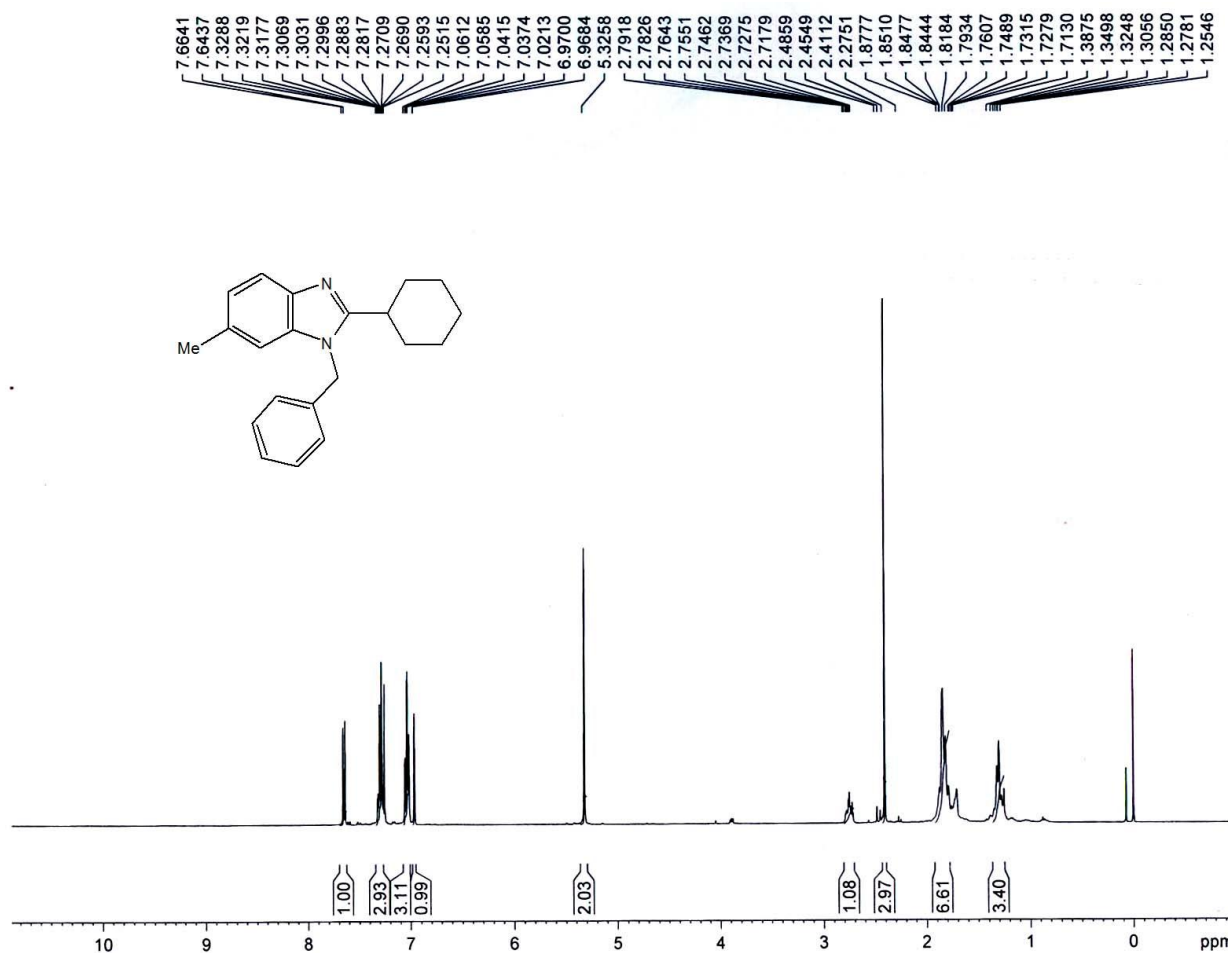


Table 5, Entry 32: ^{13}C NMR

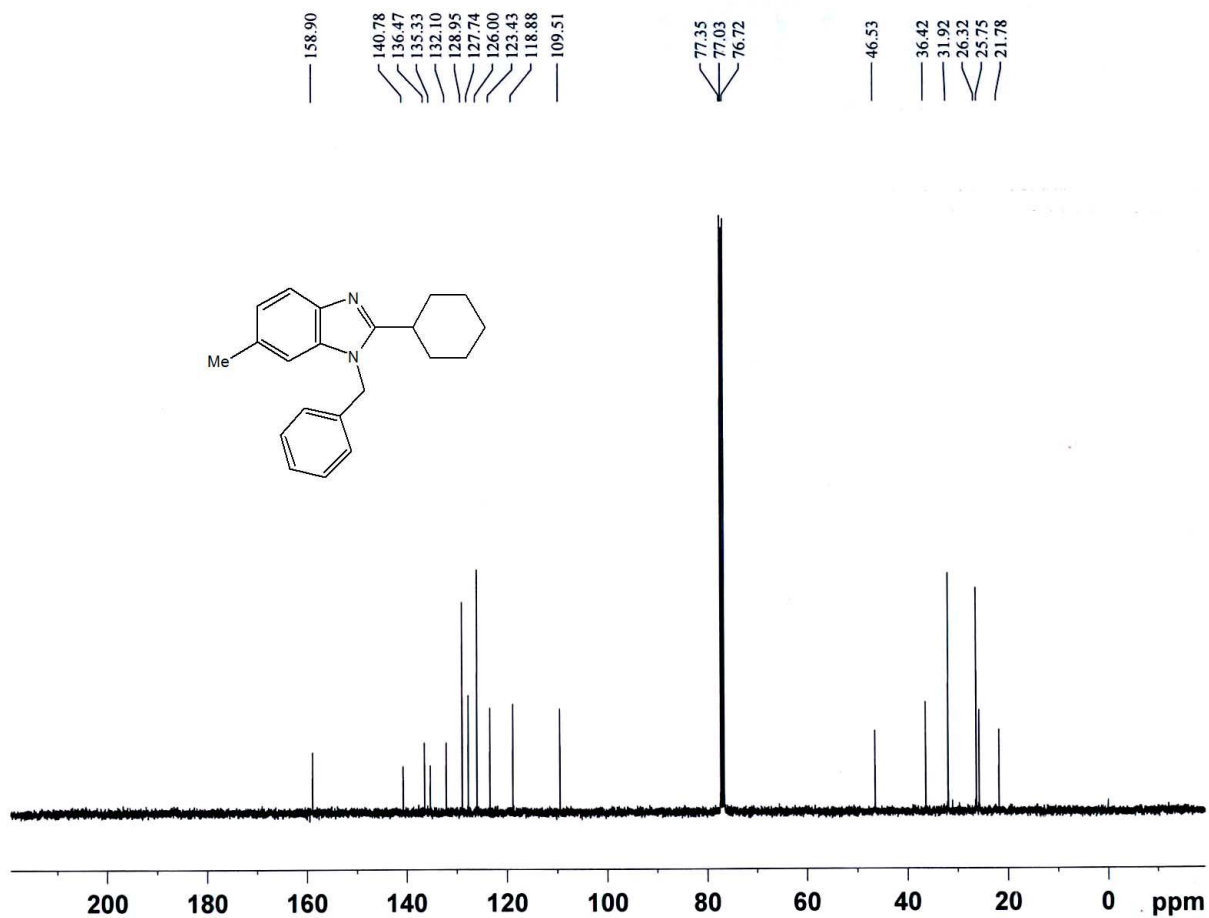


Table 5, Entry 33: ^1H NMR

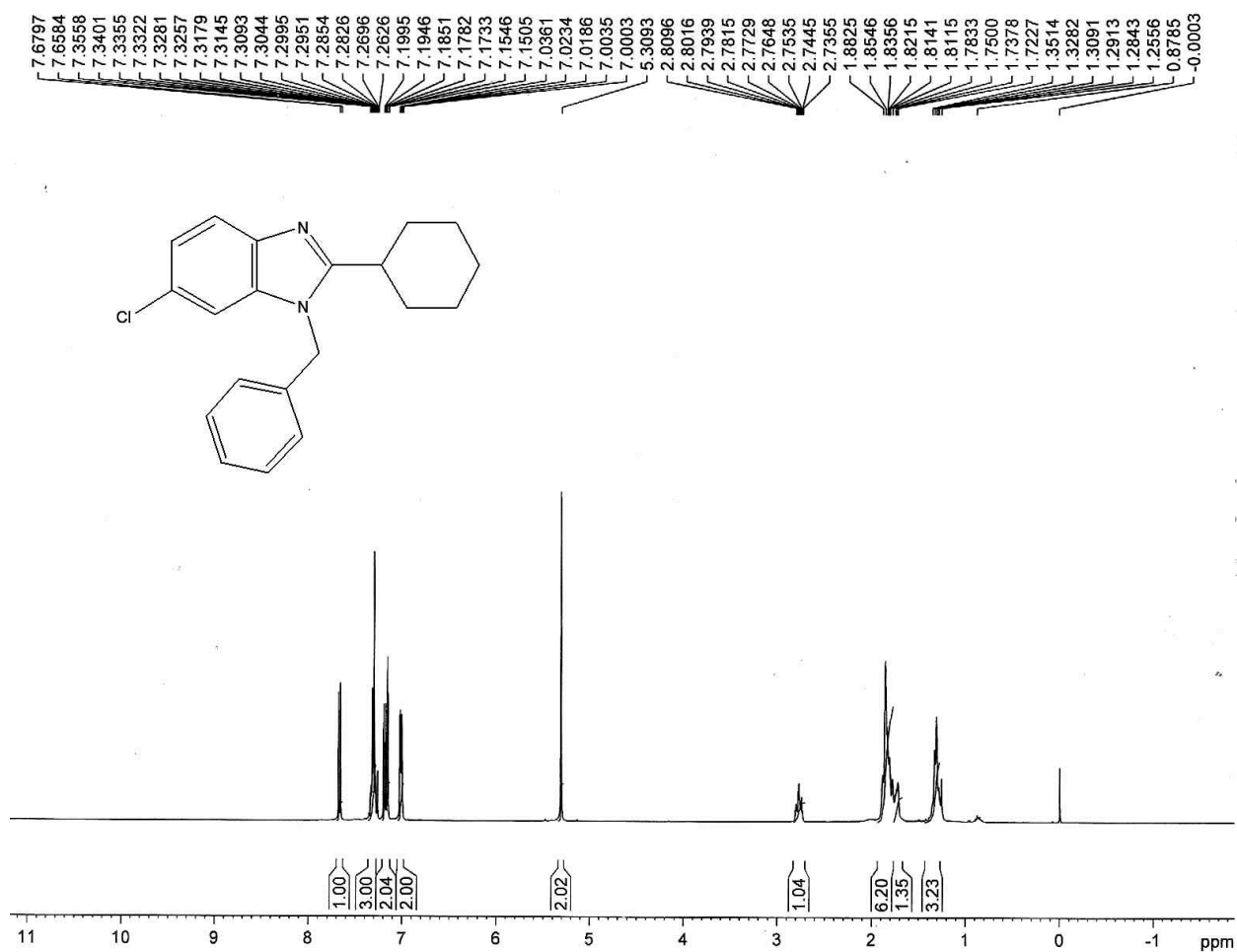


Table 5, Entry 33: ^{13}C NMR

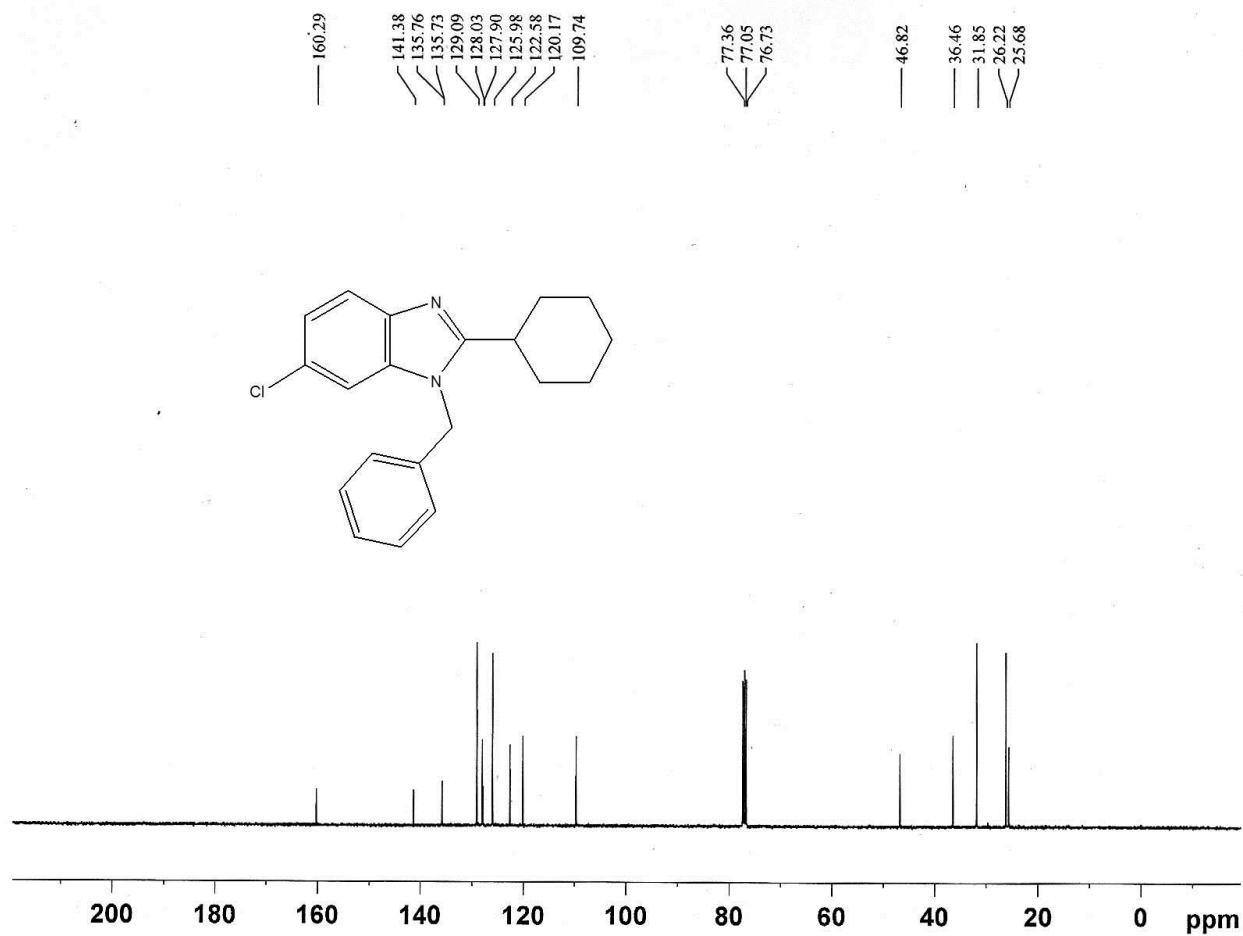
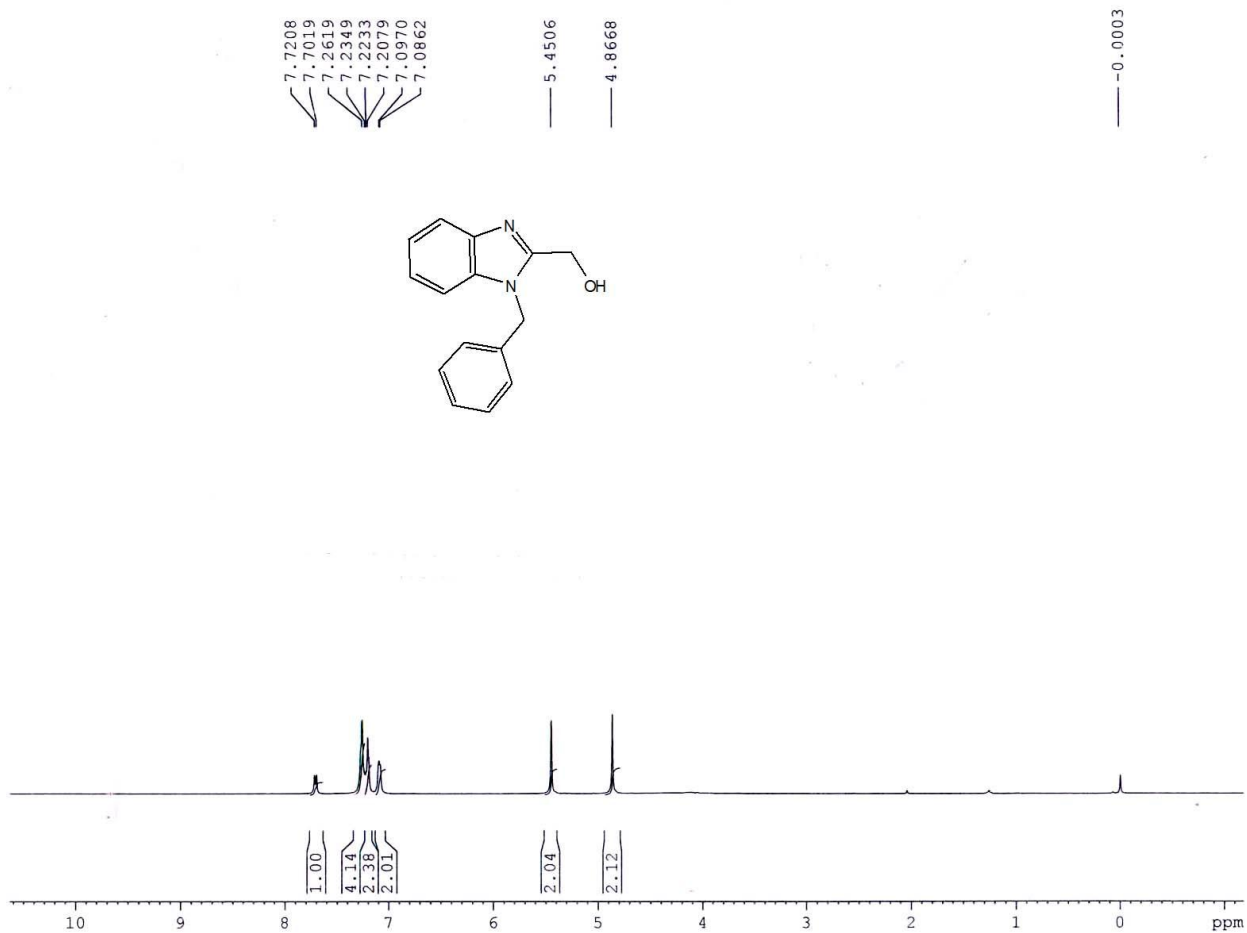
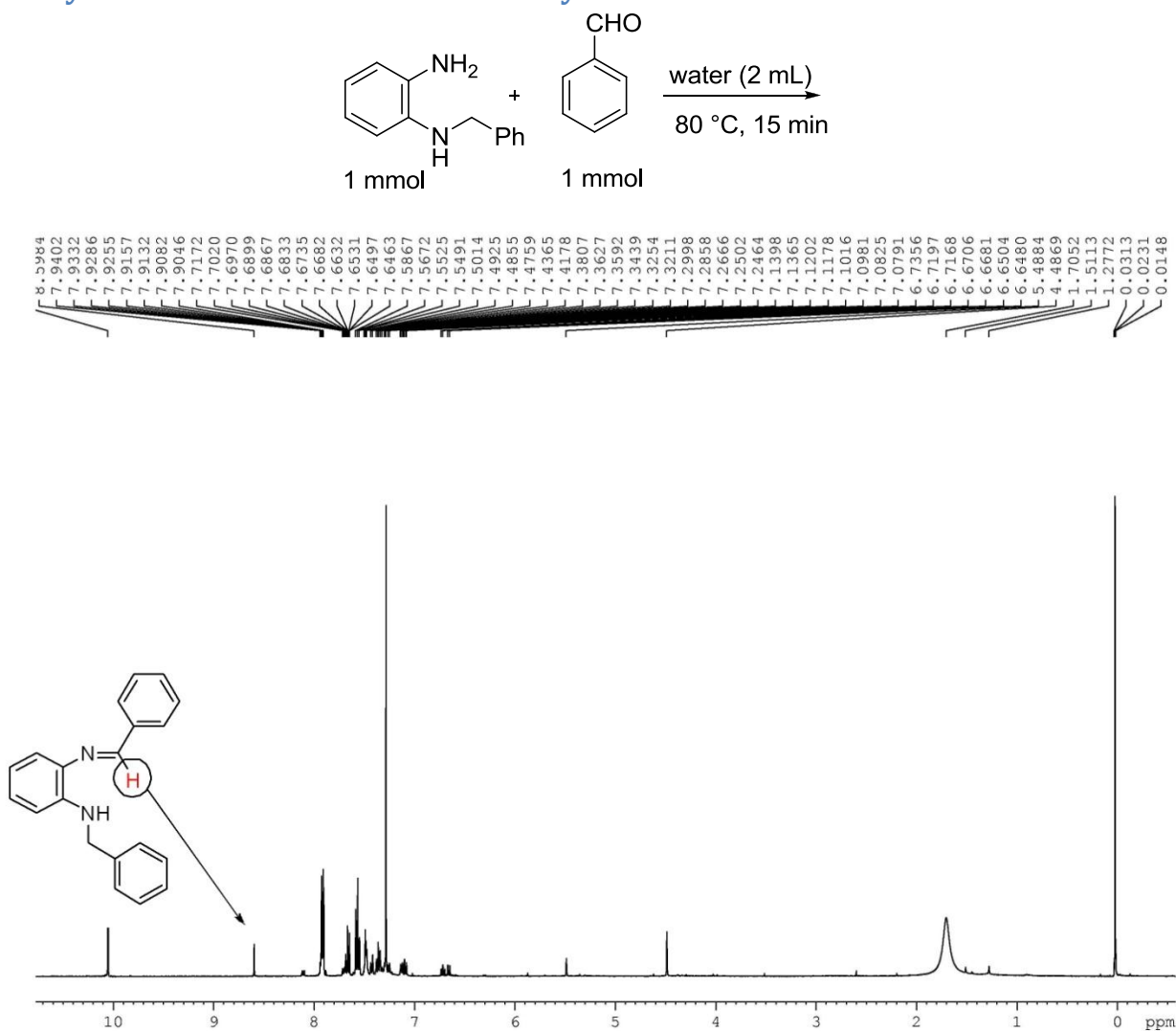


Table 5, Entry 34: ¹H NMR

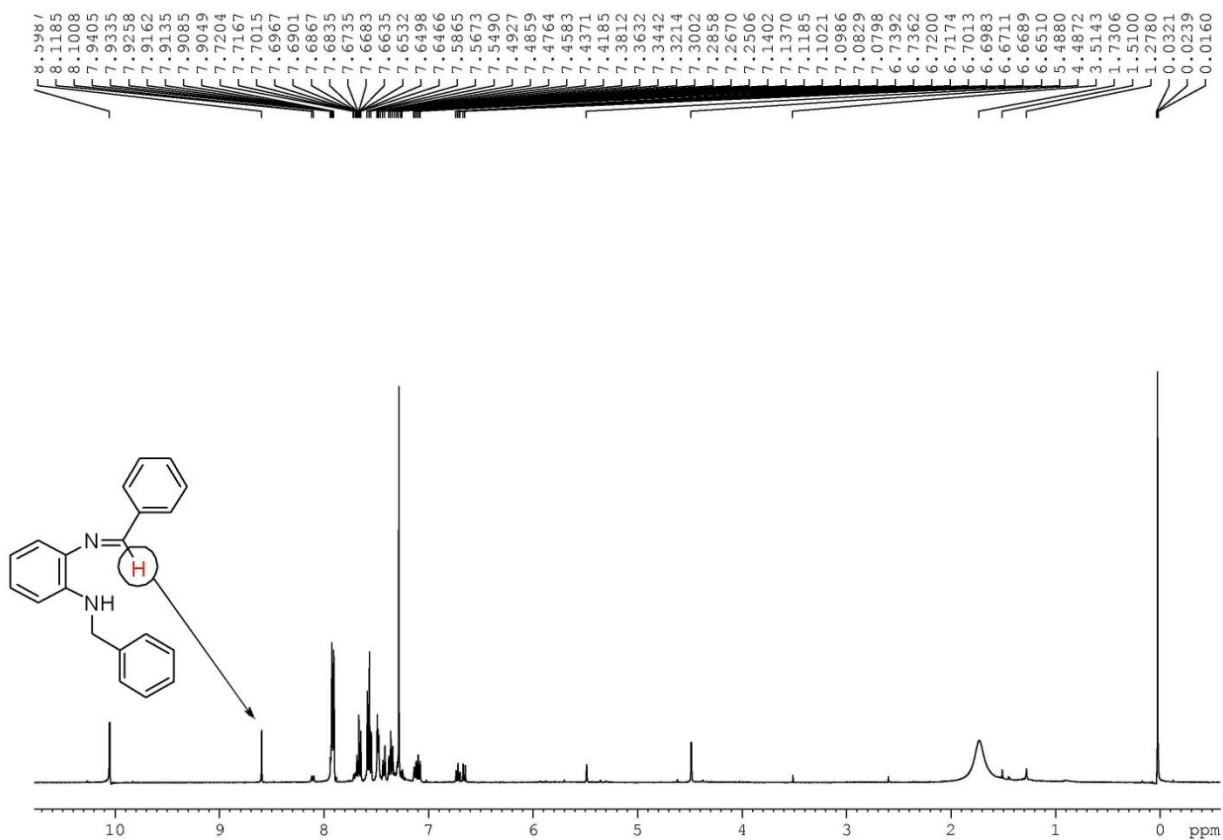
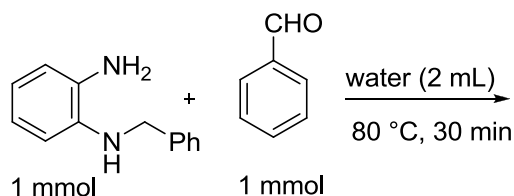


NMR study

¹H NMR of an aliquot sample of reaction mixture of *N*-methylphenyl-*o*-phenylenediamine 4a with benzaldehyde 5a in water at 80 °C after 15 min

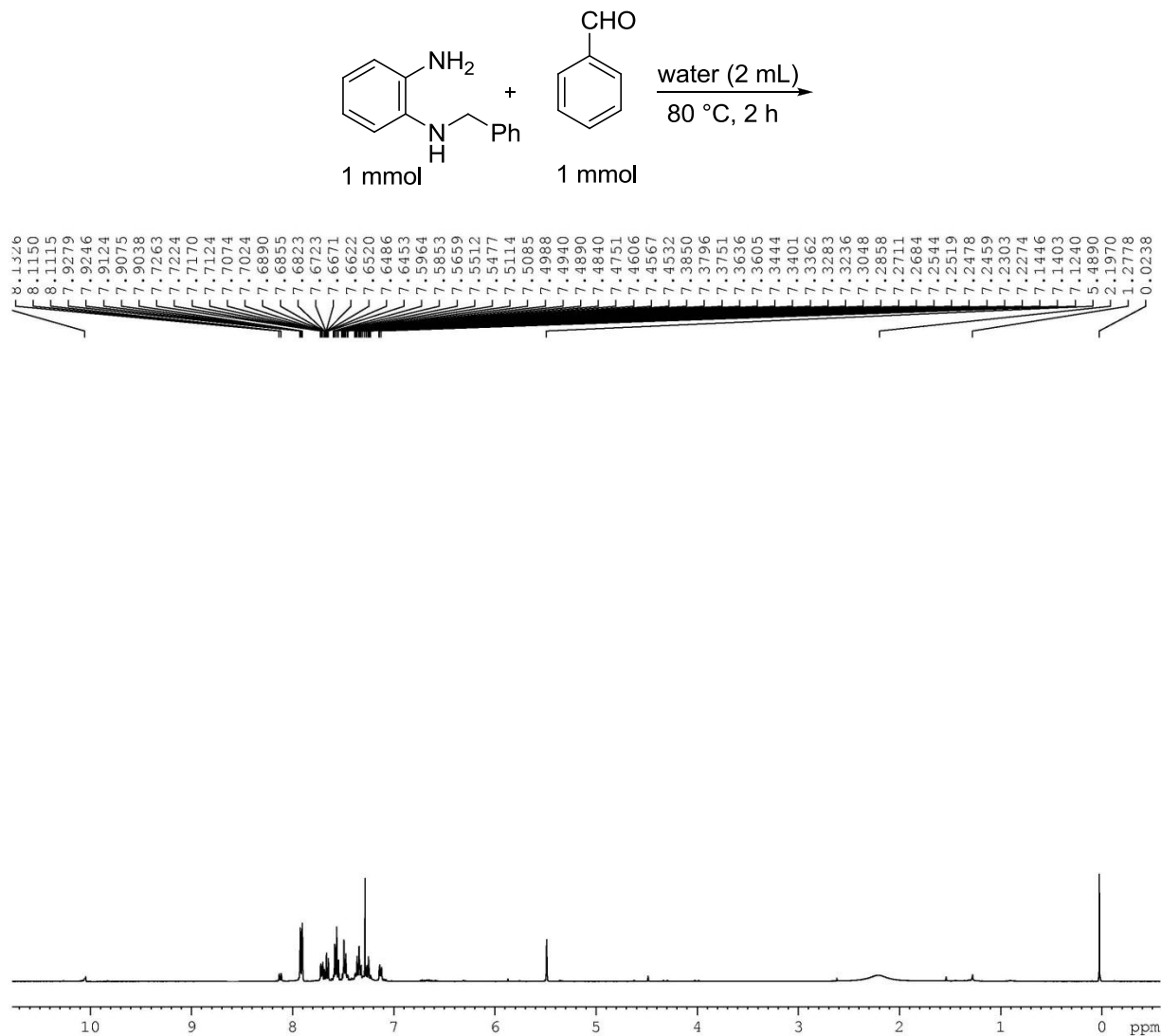


¹H NMR of an aliquot sample of reaction mixture of *N*-methylphenyl-o-phenylenediamine 4a with benzaldehyde 5a in water at 80 °C after 30 min



¹H NMR of an aliquot sample of reaction mixture of *N*-methylphenyl-*o*-phenylenediamine 4a with benzaldehyde 5a in water at 80 °C after 2 h

The imine peak at δ 8.6 was completely disappeared



¹H NMR Spectra of *N*¹,*N*²-dibenzylbenzene-1,2-diamine

