Supporting Information

KOBu^t/DMSO Mediated α-C–H Vinylation of N-Benzyl Ketimines with Acetylene Gas: Stereoselective Synthesis of (*E*,*Z*) 2-Azadienes

Ivan A. Bidusenko, Elena Yu. Schmidt, Nadezhda I. Protsuk, Igor A. Ushakov, Alexander V. Vashchenko, Andrei V. Afonin, and Boris A. Trofimov

A. E. Favorsky Irkutsk Institute of Chemistry, Siberian Branch, Russian Academy of Sciences, 1 Favorsky Str., 664033 Irkutsk, Russia

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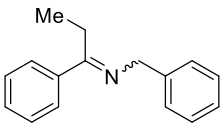
1. General information

^{1}H (400.1 MHz), ^{13}C (100.6 MHz), and ^{15}N (40.5 MHz) NMR spectra were recorded on a Bruker AV400 instrument in $CDCl_3$. The assignment of signals in the ^{1}H NMR spectra was made using COSY and NOESY experiments. Resonance signals of carbon atoms were assigned based on ^{1}H-^{13}C HSQC and ^{1}H-^{13}C HMBC experiments. The ^{1}H and ^{13}C chemical shifts (δ) were referenced to hexamethyldisiloxane (0.05 ppm and 2.0 respectively). The chemical shifts were recorded in ppm. Coupling constants (*J*) in hertz (Hz) were measured from one-dimensional spectra and multiplicities were abbreviated as following: s (singlet), d (doublet), dd (doublet of doublets), dt (doublet of triplets), dq (doublet of quartets), t (triplet), q (quartet), m (multiplet). The values of the δ ^{15}N were measured through the 2D ^{1}H-^{15}N HMBC experiment. The ^{15}N chemical shifts were referenced to CH_3NO_2. IR spectra were taken with FT-IR. Melting points (uncorrected) were measured on a Kofler micro hot-stage apparatus. The microanalyses were performed on a Flash EA 1112 Series elemental analyzer. Thin layer chromatography was carried out on Merck silica gel 60 F_{254} pre-coated aluminium foil sheets (eluent: hexane/diethyl ether = 1:3) and were visualized using UV light (254 nm). Column chromatography was carried out using slurry packed Sigma Aldrich silica gel, 70-230 mesh, pore size 60 Å (eluent: hexane/diethyl ether). Aldehydes, amines and all other chemicals and solvents are commercially available and were used without further purification.

2. Starting materials

Imines **1** were synthesized by published procedure[1] from ketones and amines in the presence of PCl_5. Physical-chemical characteristics of imines **1a,d-i,k,n,p,s** were identical to the literature data.[2]

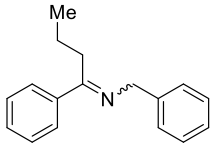
N-benzyl-1-phenylpropan-1-imine (1b)



1b was prepared from propiophenone (50 mmol, 6.709 g) and benzylamine (50 mmol, 5.358 g) as a mixture of *E*/*Z* isomers in a 2:1 molar ratio (9.192 g, 82% yield). Light yellow oil. B.p. = 111-117 ^{o}C (1 mm Hg). Elemental analysis calcd (%) for $C_{16}H_{17}N$ (235.33): C, 86.05; H, 7.67; N, 6.27; found: C, 86.17; H, 7.61; N, 6.19. **IR** (film): ν_{max} 3059, 3028, 2973, 2932, 2880, 1632, 1577, 1492, 1450, 1347, 1322, 1286, 1231, 1179, 1079, 1036, 919, 769, 732, 697. *E*-**1b**. **^{1}H NMR**: δ 7.98-7.73 (m, 2H), 7.44-7.15 (m, 8H), 4.78 (s, 2H), 2.80 (q, ^{3}J = 7.7 Hz, 2H), 1.14 (t, ^{3}J

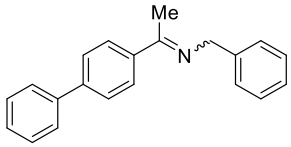
= 7.7 Hz, 3H). **^{13}C NMR**: δ 170.4, 140.7, 139.7, 129.5, 128.3, 128.2, 127.6, 127.0, 126.5, 54.8, 22.0, 11.6. *Z*-**1b**. **^{1}H NMR**: δ 7.44-7.16 (m, 8H), 7.12-7.08 (m, 2H), 4.41 (s, 2H), 2.62 (q, ^{3}J = 7.4 Hz, 2H), 1.13 (t, ^{3}J = 7.4 Hz, 3H). **^{13}C NMR**: δ 173.8, 140.7, 138.5, 128.5, 128.3, 128.1, 127.5, 126.4, 126.3, 56.7, 35.2, 10.9.

_N_-benzyl-1-phenylpropan-1-imine (1c)



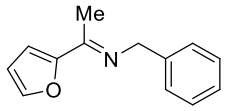
1c was prepared from 1-phenylbutan-1-one (50 mmol, 7.411 g) and benzylamine (50 mmol, 5.358 g) as a mixture of *E*/*Z* isomers in a 2:1 molar ratio (10.216 g, 86% yield). Light yellow oil. B.p. = 116-120 ^{o}C (1 mm Hg). Elemental analysis calcd (%) for $C_{17}H_{19}N$ (235.33): C, 86.03; H, 8.07; N, 5.90; found: C, 86.14; H, 8.12; N, 5.62. **IR** (film): ν_{max} 3059, 3028, 2961, 2933, 2873, 1683, 1634, 1579, 1492, 1451, 1375, 1347, 1307, 1287, 1216, 1180, 1061, 1028, 917, 900, 735, 698. *E*-**1c**. **^{1}H NMR**: δ 7.94-7.85 (m, 2H), 7.51-7.21 (m, 8H), 4.84 (s, 2H), 2.82 (dd, ^{3}J = 6.5 Hz, ^{3}J = 7.5 Hz, 2H), 1.70 -1.56 (m, 2H), 1.03 (t, ^{3}J = 7.6 Hz, 3H). **^{13}C NMR**: δ 169.5, 140.8, 140.1, 129.4, 128.3, 128.2, 127.6, 127.0, 126.5, 55.1, 30.8, 20.5, 14.3. *Z*-**1c**. **^{1}H NMR**: δ 7.51-7.21 (m, 8H, 7.20-7.13 (m, 2H), 4.48 (s, 2H), 2.66 (dd, ^{3}J = 7.6 Hz, ^{3}J = 7.1 Hz, 2H), 1.71-1.54 (m, 2H), 1.01 (t, ^{3}J = 6.8 Hz, 3H). **^{13}C NMR**: δ 173.0, 140.6, 138.5, 128.5, 128.3, 128.1, 127.5, 126.4, 126.3, 56.8, 44.2, 19.7, 13.9.

1-([1,1'-Biphenyl]-4-yl)-_N_-benzylethan-1-imine (1j)



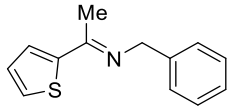
1j was prepared from 1-([1,1'-biphenyl]-4-yl)ethan-1-one (50 mmol, 9.813 g) and benzylamine (50 mmol, 5.358 g) as a mixture of *E*/*Z* isomers in a 17:1 molar ratio (11.678 g, 82% yield). Light yellow solid. M.p. = 108-109 ^{o}C. Elemental analysis calcd (%) for $C_{21}H_{19}N$ (285.39): C, 88.38; H, 6.71; N, 4.91; found: C, 88.54; H, 6.55; N, 4.85. **IR** (film): ν_{max} 3060, 3029, 2884, 1679, 1628, 1603, 1552, 1488, 1446, 1403, 1371, 1348, 1276, 1236, 1160, 1125, 1082, 1047, 1005, 906, 841, 763, 727, 693. *E*-**1j**. **^{1}H NMR**: δ 7.96-7.90 (m, 2H), 7.63-7.57 (m, 4H), 7.46-7.39 (m, 4H), 7.37-7.30 (m, 3H), 7.27-7.21 (m, 2H), 4.75 (s, 2H), 2.34 (s, 3H). **^{13}C NMR**: δ 165.5, 142.4, 140.7, 140.6, 140.0, 128.9, 128.5, 127.8, 127.6, 127.3, 127.2, 127.0, 126.6, 55.8, 15.8.

(*E*)-*N*-Benzyl-1-(furan-2-yl)ethan-1-imine (1l)



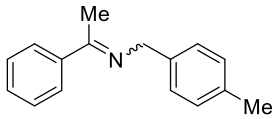
1l was prepared from 2-acetylfuran (50 mmol, 5.506 g) and benzylamine (50 mmol, 5.358 g). Yellow solid (6.878 g, 69% yield). M.p. = 37-39 ^{o}C. Elemental analysis calcd (%) for $C_{13}H_{13}NO$ (199.25): C, 78.36; H, 6.58; N, 7.03; found: C, 78.52; H, 6.46; N, 6.98. **IR** (film): ν_{max} 3140, 3111, 3084, 3060, 3028, 2957, 2920, 2862, 1628, 1575, 1487, 1450, 1365, 1349, 1291, 1223, 1162, 1103, 1071, 1050, 1015, 945, 886, 820, 739, 698. **^{1}H NMR**: δ 7.47 (dd, ^{3}J = 1.8 Hz, ^{4}J = 0.8 Hz, 1H), 7.38-7.28 (m, 4H), 7.25-7.18 (m, 1H), 6.81 (dd, ^{3}J = 3.4 Hz, ^{4}J = 0.8 Hz, 1H), 6.43 (dd, ^{3}J = 3.4 Hz, ^{3}J = 1.8 Hz, 1H), 4.72 (s, 2H), 2.23 (s, 3H). **^{13}C NMR**: δ 157.3, 154.3, 144.0, 140.1, 128.4, 127.8, 126.6, 111.4, 111.2, 55.3, 14.9.

(*E*)-*N*-Benzyl-1-(thiophen-2-yl)ethan-1-imine (1m)



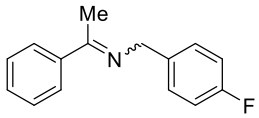
1m was prepared from 2-acetylthiophene (50 mmol, 6.309 g) and benzylamine (50 mmol, 5.358 g). Light yellow oil (8.983 g, 83% yield). B.p. = 127-128 ^{o}C (1 mm Hg). Elemental analysis calcd (%) for $C_{13}H_{13}NS$ (215.31): C, 72.52; H, 6.09; N, 6.51; S, 14.89; found: C, 72.78; H, 5.92; N, 6.46; S, 14.71. **IR** (film): ν_{max} 3065, 3028, 2919, 2861, 1621, 1529, 1494, 1432, 1367, 1347, 1279, 1234, 1063, 1030, 845, 707. **^{1}H NMR**: δ 7.39-7.34 (m, 2H), 7.33-7.26 (m, 4H), 7.22-7.16 (m, 1H), 6.99 (dd, ^{3}J = 5.2 Hz, ^{3}J = 3.7 Hz, 1H), 4.68 (s, 2H), 2.25 (s, 3H). **^{13}C NMR**: δ 160.6, 147.7, 140.2, 128.6, 128.2, 127.4, 127.1, 126.6, 126.4, 54.8, 15.5.

***N*-(4-Methylbenzyl)-1-phenylethan-1-imine (1o)**



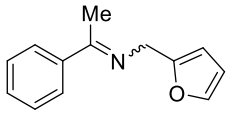
1o was prepared from acetophenone (50 mmol, 6.008 g) and *p*-tolylmethanamine (50 mmol, 6.059 g) as a mixture of *E*/*Z* isomers in a 13:1 molar ratio (9.487 g, 85% yield). Colorless solid. M.p. = 46-50 ^{o}C. B.p. = 140-145 ^{o}C (1 mm Hg). Elemental analysis calcd (%) for $C_{16}H_{17}N$ (223.32): C, 86.05; H, 7.67; N, 6.27; found: C, 86.29; H, 7.51; N, 6.13. **IR** (film): ν_{max} 3084, 3051, 3021, 2919, 2864, 1682, 1634, 1576, 1514, 1446, 1415, 1369, 1345, 1279, 1179, 1110, 1032, 801, 763, 694. *E*-**1o**. **^{1}H NMR**: δ 7.89-7.80 (m, 2H), 7.40-7.34 (m, 3H), 7.31-7.26 (m, 2H), 7.16-7.12 (m, 2H), 4.69 (s, 2H), 2.32 (s, 3H), 2.30 (s, 3H). **^{13}C NMR**: δ 165.7, 141.2, 137.6, 136.0, 129.6, 129.1, 128.2, 127.7, 126.8, 55.5, 21.2, 15.8.

***N*-(4-Fluorobenzyl)-1-phenylethan-1-imine (1q)**



1q was prepared from acetophenone (50 mmol, 6.008 g) and (4-fluorophenyl)methanamine (50 mmol, 6.258 g) as a mixture of *E*/*Z* isomers in a 13:1 molar ratio (8.418 g, 74% yield). Colorless solid. M.p. = 33-36 ^{o}C. B.p. = 123-124 ^{o}C (1 mm Hg). Elemental analysis calcd (%) for $C_{15}H_{14}FN$ (227.28): C, 79.27; H, 6.21; F, 8.36; N, 6.16; found: C, 79.36; H, 6.33; F, 8.24; N, 6.02. **IR** (film): ν_{max} 3058, 3029, 2869, 1684, 1634, 1605, 1577, 1508, 1444, 1420, 1368, 1347, 1277, 1222, 1155, 1086, 1034, 1023, 845, 822, 762, 694. *E*-**1q**. **^{1}H NMR**: δ 7.92-7.75 (m, 2H), 7.44-7.32 (m, 5H), 7.04-6.98 (m, 2H), 4.66 (s, 2H), 2.31 (s, 3H). **^{13}C NMR**: δ 165.9, 161.7 (d, ^{1}J = 243.9 Hz), 140.9, 136.4 (d, ^{4}J = 2.5 Hz), 129.7, 129.2 (d, ^{3}J = 7.8 Hz), 128.2, 126.7, 115.1 (d, ^{2}J = 21.2 Hz), 54.9, 15.7.

***N*-(Furan-2-ylmethyl)-1-phenylethan-1-imine (1r)**



1r was prepared from acetophenone (50 mmol, 6.008 g) and furan-2-ylmethanamine (50 mmol, 4.856 g) as a mixture of *E*/*Z* isomers in a 10:1 molar ratio (8.173 g, 82% yield). Yellow oil. Elemental analysis calcd (%) for $C_{13}H_{13}NO$ (199.25): C, 78.36; H, 6.58; N, 7.03; found: C, 78.52; H, 6.44; N, 6.86. **IR** (film): ν_{max} 3143, 3113, 3058, 3029, 2913, 2872, 1684, 1634, 1600, 1583, 1501, 1445, 1368, 1335, 1282, 1333, 1183, 1146, 1078, 1008, 922, 890, 807, 761, 735, 695. *E*-**1r**. **^{1}H NMR**: δ 7.83-7.77 (m, 2H), 7.38-7.31 (m, 4H), 6.32 (dd, ^{3}J = 2.8 Hz, ^{3}J = 1.5 Hz, 1H), 6.26 (d, ^{3}J = 2.8 Hz, 1H), 4.67 (s, 2H), 2.33 (s, 3H). **^{13}C NMR**: δ 167.1, 153.8, 141.6, 140.8, 129.7, 128.1, 126.8, 110.3, 106.4, 49.3, 15.7.

3. Synthesis of 2-azadienes 2 (*general procedure*)

Acetylene gas was bubbled through a mixture of imine **1** (5 mmol) and KOBu^{t} (5 mmol, 0.561 g) in DMSO (50 mL) at 40 ^{o}C (oil bath) for 30 min (acetylene flow rate ~15 cm^{3}/min). Then, the reaction mixture was diluted with H_2O (100 mL) and extracted with Et_2O (20 mL × 7). The combined organic extract was washed with H_2O (20 mL × 3) and dried over K_2CO_3 for 1 h. Et_2O was evaporated under reduced pressure, and the pure 2-azadiene **2** was obtained by column chromatography [SiO_2, eluent: hexane/Et_2O = 20:1].

Note:

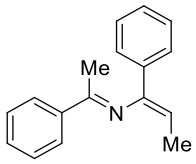
Acetylene gas was delivered to the reaction mixture from commercially available acetylene cylinder or from Kipp apparatus charged with CaC_2. In the experiment under pressure (~2 atm, Table 1, entry 11), acetylene was fed (from commercially available acetylene cylinder) into 250-mL steel Parr reactor equipped with mechanical stirrer and manometer. During all the above operations with acetylenes special safety requirements[3] should be carefully and rigorously observed. Especially no any contacts with acetylenes with copper items should be mandatory avoided.

Precautions:

Please be aware of potential safety hazards associated with the on scale use of DMSO as solvent, especially in the presence of basic additives at increased temperatures.[4]

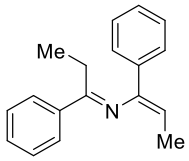
Physical-chemical characteristics of 2-azadiene **2a** were identical to the literature data.[5]

(*E*)-1-Phenyl-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2a)



Following the general procedure, **2a** was prepared from **1a** (5 mmol, 1.046 g). **2a** was isolated as a yellow oil (0.847 g, 72% yield). R_f = 0.48. Elemental analysis calcd (%) for $C_{17}H_{17}N$ (235.33): C, 86.77; H, 7.28; N, 5.95; found: C, 86.83; H, 7.16; N, 5.81. **IR** (film): ν_{max} 3055, 3033, 2963, 2917, 2855, 1638, 1601, 1581, 1443, 1367, 1333, 1285, 1250, 1180, 1153, 1112, 1078, 1029, 969, 926, 756, 741, 692. **^{1}H NMR**: δ 8.39-7.91 (m, 2H), 7.52-7.43 (m, 3H), 7.43-7.39 (m, 2H), 7.31-7.25 (m, 2H), 7.24-7.15 (m, 1H), 5.50 (q, ^3J = 7.0 Hz, 1H), 2.18 (s, 3H), 1.58 (d, ^3J = 7.0 Hz, 3H). **^{13}C NMR**: δ 165.7, 147.6, 139.4, 138.1, 130.5, 128.5, 128.4, 127.3, 127.2, 125.1, 104.2, 17.5, 12.8. **^{15}N NMR**: δ -53.5.

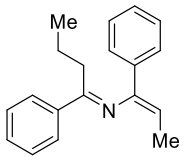
(*E*)-1-Phenyl-*N*-((*Z*)-1-phenylprop-1-en-1-yl)propan-1-imine (2b)



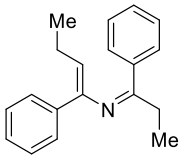
Following the general procedure, **2b** was prepared from **1b** (5 mmol, 1.117 g). **2b** was isolated as a yellow oil (0.773 g, 62% yield). R_f = 0.43. Elemental analysis calcd (%) for $C_{18}H_{19}N$ (249.36): C, 86.70; H, 7.69; N, 5.61; found: C, 86.92; H, 7.76; N, 5.28. **IR** (film): ν_{max} 3054, 3033, 2973, 2929, 2871, 1636, 1580, 1492, 1449, 1378, 1320, 1300, 1238, 1180, 1108, 1078, 1033, 985, 922,

803, 761, 731, 694. **^{1}H NMR**: δ 7.97-7.88 (m, 2H), 7.46-7.40 (m, 5H), 7.29-7.24 (m, 2H), 7.22-7.17 (m, 1H), 5.48 (q, ^{3}J = 6.9 Hz, 1H), 2.59 (q, ^{3}J = 7.7 Hz, 2H), 1.60 (d, ^{3}J = 6.9 Hz, 3H), 0.91 (t, ^{3}J = 7.7 Hz, 3H). **^{13}C NMR**: δ 171.4, 147.3, 138.5, 138.2, 130.3, 128.6, 128.4, 127.7, 127.3, 125.1, 103.8, 23.9, 13.1, 11.6.

(*E*)-1-Phenyl-*N*-((*Z*)-1-phenylprop-1-en-1-yl)butan-1-imine (2c)



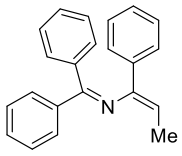
2c



2c'

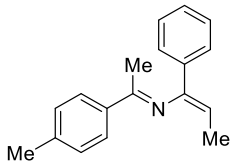
Following the general procedure, **2c** was prepared from **1c** (5 mmol, 1.187 g). **2c** was isolated with admixture of isomer **2c'** (~20%) as a yellow oil (1.040 g, 79% yield). Elemental analysis calcd (%) for $C_{19}H_{21}N$ (263.38): C, 86.64; H, 8.04; N, 5.32; found: C, 86.91; H, 7.86; N, 5.22. **IR** (film): ν_{max} 3056, 3030, 2962, 2930, 2872, 1635, 1577, 1489, 1448, 1378, 1292, 1239, 1176, 1121, 1076, 1027, 752, 693. **2c**: **^{1}H NMR**: δ 7.97-7.84 (m, 2H), 7.48-7.40 (m, 5H), 7.31-7.23 (m, 2H), 7.24-7.16 (m, 1H), 5.47 (q, ^{3}J = 6.9 Hz, 1H), 2.58-2.52 (m, 2H), 1.61 (d, ^{3}J = 6.9 Hz, 3H), 1.43-1.29 (m, 2H), 0.74 (t, ^{3}J = 7.3 Hz, 3H). **^{13}C NMR**: δ 170.5, 147.4, 138.8, 138.7, 130.2, 128.5, 128.4, 127.6, 127.3, 125.2, 104.0, 32.9, 20.5, 14.4, 11.7. **^{15}N NMR**: δ -52.0. **2c'**: **^{1}H NMR**: δ 7.98-7.88 (m, 2H), 7.49-7.39 (m, 5H), 7.30-7.24 (m, 1H), 7.22-7.18 (m, 2H), 5.38 (t, ^{3}J = 7.0 Hz, 1H), 2.61 (q, ^{3}J = 7.8 Hz, 2H), 2.00 (dq, ^{3}J = 7.0 Hz, ^{3}J = 7.4 Hz, 2H), 1.03 (t, ^{3}J = 7.4 Hz, 3H), 0.90 (t, ^{3}J = 7.8 Hz, 3H). **^{13}C NMR**: δ 171.2, 145.9, 138.5, 138.2, 130.3, 128.5, 128.4, 127.7, 127.3, 125.2, 111.6, 23.9, 20.9, 14.1, 13.1. **^{15}N NMR**: δ -52.8.

(*Z*)-1,1-Diphenyl-*N*-(1-phenylprop-1-en-1-yl)methanimine (2d)



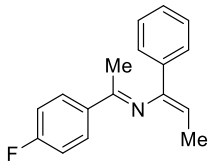
Following the general procedure, **2d** was prepared from **1d** (5 mmol, 1.357 g). **2d** was isolated as a yellow solid (0.996 g, 67% yield). R_f = 0.53. M.p. = 122-124 ^{o}C. Elemental analysis calcd (%) for $C_{22}H_{19}N$ (297.40): C, 88.85; H, 6.44; N, 4.71; found: C, 88.96; H, 6.32; N, 4.63. **IR** (film): ν_{max} 3056, 3030, 2958, 2917, 2854, 1622, 1571, 1517, 1491, 1443, 1316, 1288, 1178, 1069, 1032, 1003, 757, 695. **^{1}H NMR**: δ 7.81-7.72 (m, 2H), 7.50-7.42 (m, 1H), 7.42-7.33 (m, 4H), 7.32-7.27 (m, 1H), 7.26-7.20 (m, 4H), 7.20-7.14 (m, 1H), 7.07-7.01 (m, 2H), 5.14 (q, ^{3}J = 7.0 Hz, 1H), 1.44 (d, ^{3}J = 7.0 Hz, 3H). **^{13}C NMR**: δ 168.5, 148.3, 139.7, 139.4, 137.3, 130.7, 129.3, 128.8, 128.3, 128.2, 127.9, 127.8, 127.1, 125.2, 103.8, 13.3.

(*E*)-*N*-((*Z*)-1-Phenylprop-1-en-1-yl)-1-(*p*-tolyl)ethan-1-imine (2e)



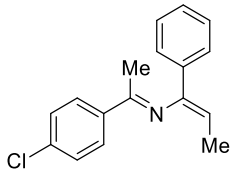
Following the general procedure, **2e** was prepared from **1e** (5 mmol, 1.117 g). **2e** was isolated as a light yellow solid (0.810 g, 65% yield). R_f = 0.49. M.p. = 51-53 ^{o}C. Elemental analysis calcd (%) for $C_{18}H_{19}N$ (249.36): C, 86.70; H, 7.68; N, 5.62; found: C, 86.87; H, 7.53; N, 5.55. **IR** (film): ν_{max} 3079, 3051, 3031, 2960, 2918, 2856, 1637, 1613, 1567, 1511, 1494, 1441, 1404, 1366, 1335, 1288, 1250, 1182, 1078, 1032, 969, 817, 756, 723, 694. **^{1}H NMR**: δ 7.93-7.87 (m, 2H), 7.46-7.36 (m, 2H), 7.31-7.24 (m, 2H), 7.26-7.22 (m, 2H), 7.22-7.15 (m, 1H), 5.49 (q, ^{3}J = 7.0 Hz, 1H), 2.39 (s, 3H), 2.15 (s, 3H), 1.57 (d, ^{3}J = 7.0 Hz, 3H). **^{13}C NMR**: δ 165.5, 147.6, 140.7, 138.1, 136.7, 129.1, 128.3, 127.3, 127.2, 125.1, 104.1, 21.4, 17.3, 12.8.

(*E*)-1-(4-Fluorophenyl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2f)



Following the general procedure, **2f** was prepared from **1f** (5 mmol, 1.136 g). **2f** was isolated as a yellow oil (1.013 g, 80% yield). R_f = 0.49. Elemental analysis calcd (%) for $C_{17}H_{16}FN$ (253.32): C, 80.60; H, 6.37; F, 7.50; N, 5.53; found: C, 80.84; H, 6.24; F, 7.48; N, 5.41. **IR** (film): ν_{max} 3056, 2962, 2917, 2856, 1639, 1599, 1550, 1505, 1440, 1411, 1367, 1289, 1231, 1157, 1099, 1079, 1031, 969, 936, 838, 759, 726, 695. **^{1}H NMR**: δ 8.05-7.94 (m, 2H), 7.43-7.38 (m, 2H), 7.32-7.24 (m, 2H), 7.24-7.18 (m, 1H), 7.16-7.04 (m, 2H), 5.49 (q, ^{3}J = 6.9 Hz, 1H), 2.15 (s, 3H), 1.56 (d, ^{3}J = 6.9 Hz, 3H). **^{13}C NMR**: δ 164.4 (d, ^{1}J = 250.5 Hz), 164.4, 147.5, 138.0, 135.5 (d, ^{4}J = 3.1 Hz), 129.3 (d, ^{3}J = 8.4 Hz), 128.4, 127.4, 125.1, 115.3 (d, ^{2}J = 21.6 Hz), 104.3, 17.3, 12.8. **^{15}N NMR**: δ -53.5.

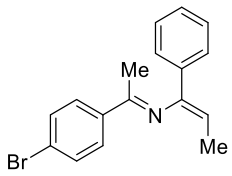
(*E*)-1-(4-Chlorophenyl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2g)



Following the general procedure, **2g** was prepared from **1g** (5 mmol, 1.219 g). **2g** was isolated as a yellow oil (0.917 g, 68% yield). R_f = 0.49. Elemental analysis calcd (%) for $C_{17}H_{16}ClN$ (269.77): C, 75.69; H, 5.98; Cl, 13.14; N, 5.19; found: C, 75.88; H, 5.92; Cl, 13.09; N, 5.02. **IR**

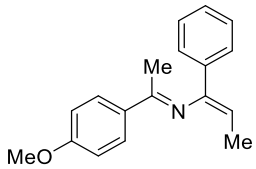
(film): ν_{max} 3054, 2967, 2915, 2855, 1638, 1593, 1568, 1489, 1441, 1398, 1367, 1334, 1286, 1250, 1178, 1095, 1030, 1019, 830, 778, 748, 696. **^{1}H NMR**: δ 8.01-7.90 (m, 2H), 7.44-7.34 (m, 4H), 7.30-7.25 (m, 2H), 7.24-7.18 (m, 1H), 5.49 (q, ^{3}J = 6.9 Hz, 1H), 2.15 (s, 3H), 1.56 (d, ^{3}J = 6.9 Hz, 3H). **^{13}C NMR**: δ 164.5, 147.4, 137.9, 137.7, 136.6, 128.7, 128.6, 128.4, 127.4, 125.1, 104.4, 17.3, 12.8.

(*E*)-1-(4-Bromophenyl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2h)



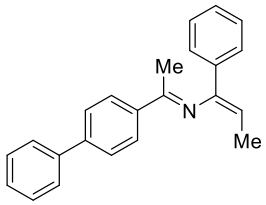
Following the general procedure, **2h** was prepared from **1h** (5 mmol, 1.441 g). **2h** was isolated as a yellow oil (0.864 g, 55% yield). R_f = 0.48. Elemental analysis calcd (%) for $C_{17}H_{16}BrN$ (314.23): C, 64.98; H, 5.13; Br, 25.43; N, 4.46; found: C, 65.12; H, 5.06; Br, 25.51; N, 4.28. **IR** (film): ν_{max} 3051, 2961, 2917, 2855, 1637, 1589, 1550, 1488, 1440, 1393, 1367, 1334, 1286, 1251, 1178, 1078, 1032, 1011, 825, 767, 745, 693. **^{1}H NMR**: δ 7.91-7.83 (m, 2H), 7.59-7.52 (m, 2H), 7.43-7.38 (m, 2H), 7.31-7.25 (m, 2H), 7.25-7.16 (m, 1H), 5.49 (q, ^{3}J = 7.0 Hz, 1H), 2.15 (s, 3H), 1.55 (d, ^{3}J = 7.0 Hz, 3H). **^{13}C NMR**: δ 164.7, 147.4, 138.1, 137.9, 131.6, 128.8, 128.4, 127.5, 125.1, 125.0, 104.4, 17.3, 12.9.

(*E*)-1-(4-Methoxyphenyl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2i)



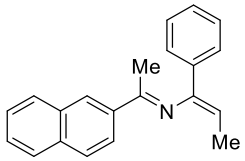
Following the general procedure, **2i** was prepared from **1i** (5 mmol, 1.197 g). **2i** was isolated as a light yellow solid (0.995 g, 75% yield). R_f = 0.44. M.p. = 71-72 ^{o}C. Elemental analysis calcd (%) for $C_{18}H_{19}NO$ (265.36): C, 81.47; H, 7.22; N, 5.28; found: C, 81.54; H, 7.19; N, 5.23. **IR** (film): ν_{max} 3050, 3005, 2958, 2927, 2914, 2844, 1635, 1603, 1574, 1510, 1444, 1422, 1366, 1303, 1251, 1176, 1032, 836, 807, 757, 723, 694. **^{1}H NMR**: δ 7.97-7.86 (m, 2H), 7.42-7.34 (m, 2H), 7.27-7.18 (m, 2H), 7.20-7.11 (m, 1H), 6.96-6.84 (m, 2H), 5.44 (q, ^{3}J = 6.9 Hz, 1H), 3.80 (s, 3H), 2.09 (s, 3H), 1.52 (d, ^{3}J = 6.9 Hz, 3H). **^{13}C NMR**: δ 164.7, 161.6, 147.6, 138.2, 132.1, 128.8, 128.3, 127.2, 125.1, 113.7, 104.2, 55.4, 17.2, 12.8. **^{15}N NMR**: δ -58.8.

(*E*)-1-([1,1'-Biphenyl]-4-yl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2j)



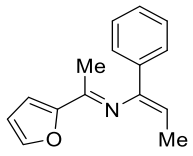
Following the general procedure, **2j** was prepared from **1j** (5 mmol, 1.427 g). **2j** was isolated as a light yellow solid (0.950 g, 61% yield). R_f = 0.50. M.p. = 93-95 ^oC. Elemental analysis calcd (%) for $C_{23}H_{21}N$ (311.43): C, 88.71; H, 6.80; N, 4.50; found: C, 88.87; H, 6.71; N, 4.39. **IR** (film): ν_{max} 3053, 3033, 2959, 2917, 2854, 1636, 16.04, 1552, 1489, 1442, 1401, 1367, 1334, 1287, 1252, 1187, 1122, 1074, 1036, 843, 791, 757, 720, 694. **^{1}H NMR**: δ 8.14-8.03 (m, 2H), 7.73-7.60 (m, 4H), 7.50-7.41 (m, 4H), 7.41-7.32 (m, 1H), 7.33-7.24 (m, 2H), 7.24-7.16 (m, 1H), 5.51 (q, ^3J = 7.0 Hz, 1H), 2.21 (s, 3H), 1.59 (d, ^3J = 7.0 Hz, 3H). **^{13}C NMR**: δ 165.3, 147.6, 143.3, 140.6, 138.2, 138.1, 129.0, 128.4, 127.8, 127.9, 127.4, 127.3, 127.2, 125.2, 104.2, 17.4, 12.9. **^{15}N NMR**: δ -52.8.

(*E*)-1-(Naphthalen-2-yl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2k)



Following the general procedure, **2k** was prepared from **1k** (5 mmol, 1.297 g). **2k** was isolated as a light yellow solid (0.985 g, 69% yield). R_f = 0.50. M.p. = 91-93 ^oC Elemental analysis calcd (%) for $C_{21}H_{19}N$ (285.39): C, 88.38; H, 6.71; N, 4.91; found: C, 88.38; H, 6.71; N, 4.91. **IR** (film): ν_{max} 3070, 3038, 2964, 2916, 2854, 1624, 1552, 1529, 1492, 1432, 1367, 1328, 1285, 1255, 1239, 1060, 1023, 968, 915, 848, 758, 714. **^{1}H NMR**: δ 8.36-8.31 (m, 1H), 8.30-8.25 (m, 1H), 7.94-7.81 (m, 3H), 7.53-7.49 (m, 2H), 7.48-7.44 (m, 2H), 7.32-7.26 (m, 2H), 7.24-7.19 (m, 1H), 5.52 (q, ^3J = 7.0 Hz, 1H), 2.29 (s, 3H), 1.60 (d, ^3J = 7.0 Hz, 3H). **^{13}C NMR**: δ 165.5, 147.7, 138.1, 136.7, 134.5, 133.1, 129.0, 128.4, 128.1, 127.8, 127.5, 127.4, 127.2, 126.4, 125.1, 124.4, 104.2, 17.4, 12.9. **^{15}N NMR**: δ -51.2.

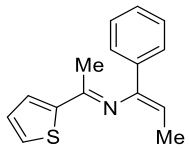
(*E*)-1-(Furan-2-yl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-imine (2l)



Following the general procedure, **2l** was prepared from **1l** (5 mmol, 0.996 g). **2i** was isolated as a yellow oil (0.755 g, 67% yield). R_f = 0.44. Elemental analysis calcd (%) for $C_{15}H_{15}NO$ (225.29):

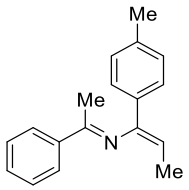
C, 79.97; H, 6.71; N, 6.22; found: C, 80.15; H, 6.62; N, 6.13. **IR** (film): ν_{max} 3111, 3041, 1963, 2918, 2857, 1680, 1632, 1576, 1485, 1441, 1389, 1368, 1331, 1296, 1254, 1227, 1165, 1105, 1077, 1020, 950, 898, 815, 752, 712, 646, 596. **^{1}H NMR**: δ 7.54 (d, ^3J = 1.8 Hz, 1H), 7.45-7.39 (m, 2H), 7.31-7.25 (m, 2H), 7.23-7.17 (m, 1H), 6.94 (d, ^3J = 3.3 Hz, 1H), 6.50 (dd, ^3J = 3.3 Hz, ^3J = 1.8 Hz, 1H), 5.49 (q, ^3J = 7.0 Hz, 1H), 2.07 (s, 3H), 1.59 (d, ^3J = 7.0 Hz, 3H). **^{13}C NMR**: δ 157.0, 153.7, 146.8, 144.6, 138.1, 128.4, 127.3, 125.2, 112.5, 111.8, 105.1, 16.7, 12.9.

(*E*)-*N*-((*Z*)-1-Phenylprop-1-en-1-yl)-1-(thiophen-2-yl)ethan-1-imine (2m)



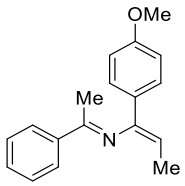
Following the general procedure, **2m** was prepared from **1m** (5 mmol, 1.077 g). **2m** was isolated as a yellow oil (0.772 g, 64% yield). R_f = 0.43. Elemental analysis calcd (%) for $C_{15}H_{15}NS$ (241.35): C, 74.65; H, 6.26; N, 5.80; S, 13.28; found: C, 74.86; H, 6.14; N, 5.75; S, 13.16. **IR** (film): ν_{max} 3070, 3038, 2964, 2916, 2854, 1624, 1529, 1492, 1432, 1367, 1328, 1285, 1255, 1239, 1060, 1023, 968, 915, 848, 758, 714. **^{1}H NMR**: δ 7.45-7.38 (m, 4H), 7.30-7.24 (m, 2H), 7.22-7.16 (m, 1H), 7.07 (dd, ^3J = 5.0 Hz, ^3J = 3.7 Hz, 1H), 5.52 (q, ^3J = 7.0 Hz, 1H), 2.15 (s, 3H), 1.59 (d, ^3J = 7.0 Hz, 3H). **^{13}C NMR**: δ 160.7, 146.6, 146.3, 138.0, 129.7, 128.3, 128.2, 127.5, 127.3, 125.1, 105.3, 17.5, 12.9.

(*E*)-1-Phenyl-*N*-((*Z*)-1-(*p*-tolyl)prop-1-en-1-yl)ethan-1-imine (2o)



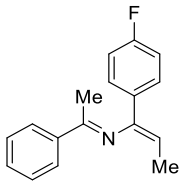
Following the general procedure, **2o** was prepared from **1o** (5 mmol, 1.117 g). **2o** was isolated as a yellow oil (0.536 g, 43% yield). R_f = 0.50. Elemental analysis calcd (%) for $C_{18}H_{19}N$ (249.36): C, 86.70; H, 7.68; N, 5.62; found: C, 86.82; H, 7.61; N, 5.56. **IR** (film): ν_{max} 3052, 3028, 2965, 2918, 2858, 1637, 1576, 1509, 1443, 1367, 1332, 1286, 1253, 1182, 1115, 1081, 1029, 970, 931, 827, 798, 764, 694. **^{1}H NMR**: δ 8.03-7.97 (m, 2H), 7.46-7.39 (m, 3H), 7.36-7.27 (m, 2H), 7.12-7.03 (m, 2H), 5.45 (q, ^3J = 7.0 Hz, 1H), 2.31 (s, 3H), 2.17 (s, 3H), 1.56 (d, ^3J = 7.0 Hz, 3H). **^{13}C NMR**: δ 165.5, 147.5, 139.4, 137.0, 135.2, 130.4, 129.1, 128.4, 127.2, 125.0, 103.2, 21.2, 17.4, 12.8.

(*E*)-*N*-((*Z*)-1-(4-Methoxyphenyl)prop-1-en-1-yl)-1-phenylethan-1-imine (2p)



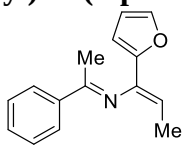
Following the general procedure, **2p** was prepared from **1p** (5 mmol, 1.197 g). **2p** was isolated as a light yellow oil (0.643 g, 48% yield). R_f = 0.31. Elemental analysis calcd (%) for $C_{18}H_{19}NO$ (265.36): C, 81.47; H, 7.22; N, 5.28; found: C, 81.64; H, 7.14; N, 5.17. **IR** (film): ν_{max} 3056, 3036, 3000, 2955, 2932, 2909, 2853, 2835, 1638, 1606, 1577, 1509, 1463, 1446, 1417, 1365, 1333, 1284, 1246, 1178, 1112, 1081, 1034, 970, 934, 837, 800, 764, 751, 693. **^{1}H NMR**: δ 8.06-7.83 (m, 2H), 7.46-7.41 (m, 3H), 7.39-7.33 (m, 2H), 6.85-6.37 (m, 2H), 5.37 (q, ^3J = 6.9 Hz, 1H), 3.77 (s, 3H), 2.17 (s, 3H), 1.55 (d, ^3J = 6.9 Hz, 3H). **^{13}C NMR**: δ 165.4, 159.0, 147.1, 139.3, 130.7, 130.4, 128.4, 127.1, 126.2, 113.7, 102.2, 55.2, 17.3, 12.7.

(*E*)-*N*-((*Z*)-1-(4-Fluorophenyl)prop-1-en-1-yl)-1-phenylethan-1-imine (2q)

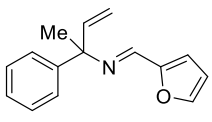


Following the general procedure, **2q** was prepared from **1q** (5 mmol, 1.136 g). **2q** was isolated as a yellow oil (0.646 g, 51% yield). R_f = 0.50. Elemental analysis calcd (%) for $C_{17}H_{16}FN$ (253.32): C, 80.60; H, 6.37; F, 7.50; N, 5.53; found: C, 80.79; H, 6.28; F, 7.42; N, 5.46. **IR** (film): ν_{max} 3056, 2963, 2916, 2856, 1638, 1602, 1549, 1505, 1442, 1406, 1367, 1333, 1287, 1229, 1158, 1091, 1029, 970, 932, 841, 809, 763, 694. **^{1}H NMR**: δ 8.03-7.96 (m, 2H), 7.47-7.42 (m, 3H), 7.42-7.35 (m, 2H), 6.99-6.93 (m, 2H), 5.41 (q, ^3J = 7.0 Hz, 1H), 2.17 (s, 3H), 1.56 (d, ^3J = 7.0 Hz, 3H). **^{13}C NMR**: δ 165.9, 162.4 (d, ^1J = 246.1 Hz), 146.7, 139.2, 134.3 (d, ^4J = 2.6 Hz), 130.6, 128.5, 127.2, 126.7 (d, ^3J = 7.9 Hz), 115.2 (d, ^2J = 21.2 Hz), 104.0, 17.4, 12.8.

(*E*)-*N*-((*Z*)-1-(Furan-2-yl)prop-1-en-1-yl)-1-phenylethan-1-imine (2r) and (*E*)-1-(furan-2-yl)-*N*-(2-phenylbut-3-en-2-yl)methanimine (2r')



2r

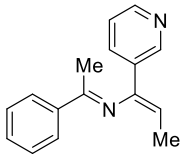


2r'

Following the general procedure, **2r** and **2r'** were prepared from **1r** (5 mmol, 0.996 g). **2r** was isolated as a yellow oil (0.687 g, 61% yield). R_f = 0.47. Elemental analysis calcd (%) for $C_{15}H_{15}NO$ (225.29): C, 79.97; H, 6.71; N, 6.22; found: C, 80.11; H, 6.65; N, 6.13. **IR** (film): ν_{max}

3142, 3112, 3057, 3033, 2965, 2915, 2855, 1638, 1572, 1548, 1493, 1443, 1368, 1284, 1219, 1159, 1032, 1010, 944, 888, 766, 735, 691. **^{1}H NMR**: δ 8.04-7.84 (m, 2H), 7.54-7.39 (m, 3H), 7.34 (d, ^{3}J = 1.7 Hz, 1H), 6.31 (dd, ^{3}J = 3.2 Hz, ^{3}J = 1.7 Hz, 1H), 6.02 (d, ^{3}J = 3.2 Hz, 1H), 5.58 (q, ^{3}J = 7.1 Hz, 1H), 2.21 (s, 3H), 1.57 (d, ^{3}J = 7.1 Hz, 3H). **^{13}C NMR**: δ 167.9, 151.3, 141.8, 139.7, 139.3, 130.6, 128.5, 127.3, 111.1, 105.5, 103.4, 17.2, 12.0. **2r'** was isolated as a yellow oil (0.034 g, 3% yield). R_f = 0.39. **^{1}H NMR**: δ 7.99 (s, 1H), 7.52 (d, ^{3}J = 1.8 Hz, 1H), 7.45-7.39 (m, 2H), 7.35-7.28 (m, 2H), 7.26-7.19 (m, 1H), 6.74 (d, ^{3}J = 3.4 Hz, 1H), 6.46 (dd, ^{3}J = 3.4 Hz, ^{3}J = 1.8 Hz, 1H), 6.06 (dd, ^{3}J = 17.4 Hz, ^{3}J = 10.6 Hz, 1H), 5.31 (d, ^{3}J = 10.7 Hz, 1H), 5.22 (d, ^{3}J = 17.4 Hz, 1H), 1.69 (s, 3H). **^{13}C NMR**: δ 152.5, 148.3, 145.5, 144.9, 144.0, 128.3, 127.2, 126.9, 114.9, 114.3, 111.8, 68.1, 27.5. **^{15}N NMR**: δ -43.1.

(*E*)-1-Phenyl-*N*-((*Z*)-1-(pyridin-3-yl)prop-1-en-1-yl)ethan-1-imine (2s)



Following the general procedure, **2s** was prepared from **1s** (5 mmol, 1.051 g). **2s** was isolated as a yellow oil (0.628 g, 53% yield). R_f = 0.26. Elemental analysis calcd (%) for $C_{16}H_{16}N_2$ (236.32): C, 81.32; H, 6.82; N, 11.85; found: C, 81.47; H, 6.69; N, 11.78. **IR** (film): ν_{max} 3070, 3048, 2999, 2958, 2938, 2835, 1621, 1598, 1582, 1561, 1487, 1462, 1434, 1362, 1321, 1279, 1270, 1249, 1224, 1191, 1185, 1153, 1043. 994, 981, 876, 864, 795, 741, 688. **^{1}H NMR**: δ 8.72 (s, 1H), 8.45 (d, ^{3}J = 4.7 Hz, 1H), 8.05-7.94 (m, 2H), 7.68 (d, ^{3}J = 8.0 Hz, 1H), 7.51-7.37 (m, 3H), 7.19 (dd, ^{3}J = 8.0 Hz, ^{3}J = 4.7 Hz, 1H), 5.55 (q, ^{3}J = 6.9 Hz, 1H), 2.19 (s, 3H), 1.60 (d, ^{3}J = 6.9 Hz, 3H). **^{13}C NMR**: δ 166.4, 148.2, 146.6, 144.9, 138.8, 133.6, 132.1, 130.6, 128.4, 127.1, 123.1, 105.8, 17.4, 12.7.

4. X-Ray diffraction analysis of 2-azadiene 2k

The determination of the unit cell and the data collection for (*E*)-1-(naphtalen-2-yl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-mine (**2k**) was performed on a Bruker D8 Venture Photon 100 CMOS diffractometer with MoK_α radiation (λ = 0.71073) at 297.0(2) K using the ω–φ scan technique. A specimen of $C_{21}H_{19}N$, approximate dimensions 0.37 mm x 0.08 mm x 0.05 mm, was used for the X-ray crystallographic analysis. The X-ray intensity data were measured. The integration of the data using a monoclinic unit cell with *P2$_1$/n* space group yielded a total of 27864 reflections to a maximum θ angle of 26.0° (0.81 Å resolution), of which 3174 were independent (completeness = 100%, Rint = 7.11%, Rsig = 4.43%) and 1663 were greater than

2σ(F2). The final cell constants of **a** = 12.154(12) Å, **b** = 6.023(6) Å, **c** = 22.30(3) Å, β = 99.32(3)°, Z= 4, volume = 1611(3) Å^3. Data were corrected for absorption effects using the multi-scan method (SADABS). The ratio of minimum to maximum apparent transmission was 0.922. The structure was solved and refined using the Bruker SHELXTL Software Package.[6] The H atoms were determined by rider method.

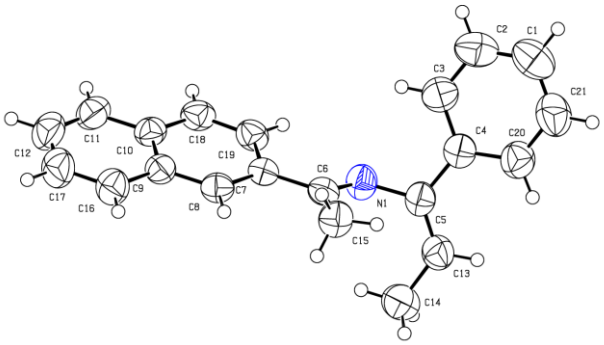


Figure 2. X-ray structure of (*E*)-1-(naphtalen-2-yl)-*N*-((*Z*)-1-phenylprop-1-en-1-yl)ethan-1-mine (**2k**). Thermal ellipsoids set at 50% probability.

The final anisotropic full-matrix least-squares refinement on F^2 with 201 variables converged at R1 = 5.61%, for the observed data and wR2 = 15.16% for all data. The goodness-of-fit was 1.073. The largest peak in the final difference electron density synthesis was 0.130 e-/Å^3 and the deepest hole was -0.190 e-/Å^3 . On the basis of the final model, the calculated density was 1.177 g/cm^3 and F(000), 608 e-.

Atomic coordinates, bond lengths, bond angles and thermal parameters have been deposited at the Cambridge Crystallographic Data Centre (CCDC) and allocated the deposition numbers CCDC **1968359.** These data can be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

5. References

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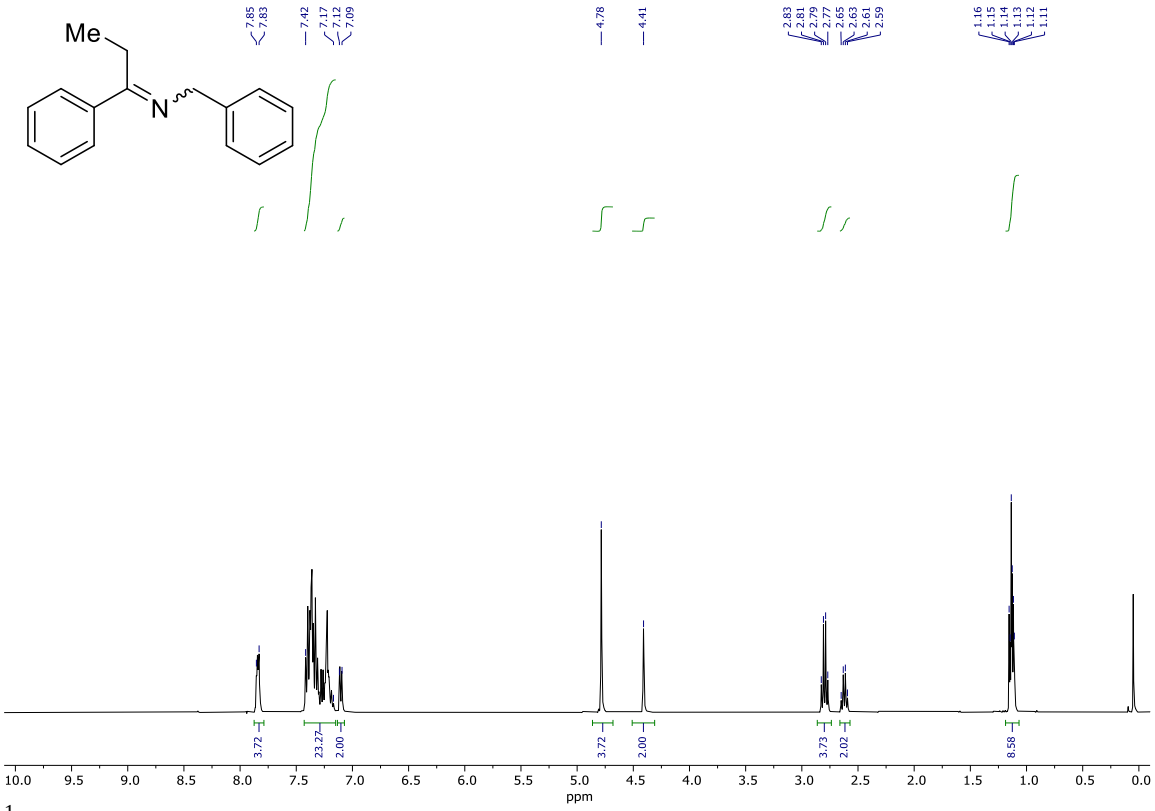
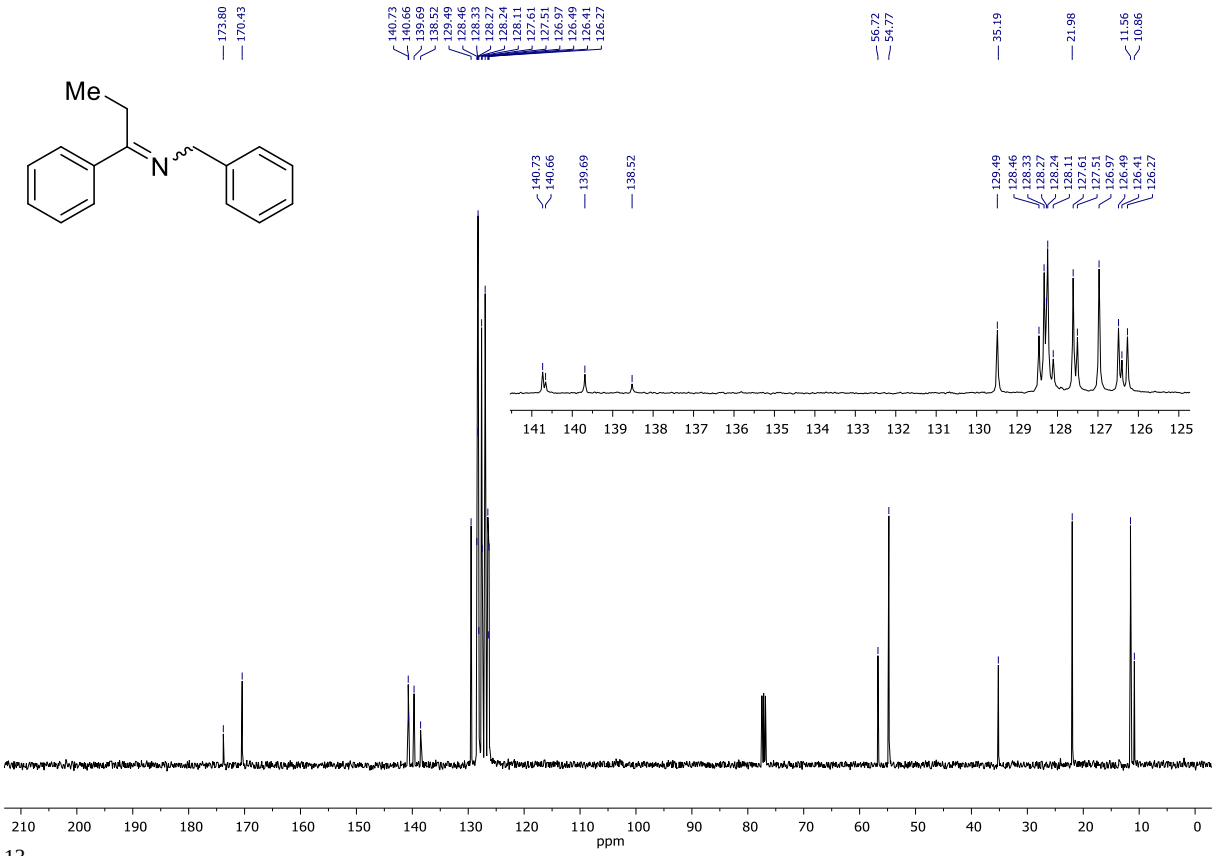
6. Sheldrick G. M. Acta Crystallogr. **2008**, *64*, 112-122.

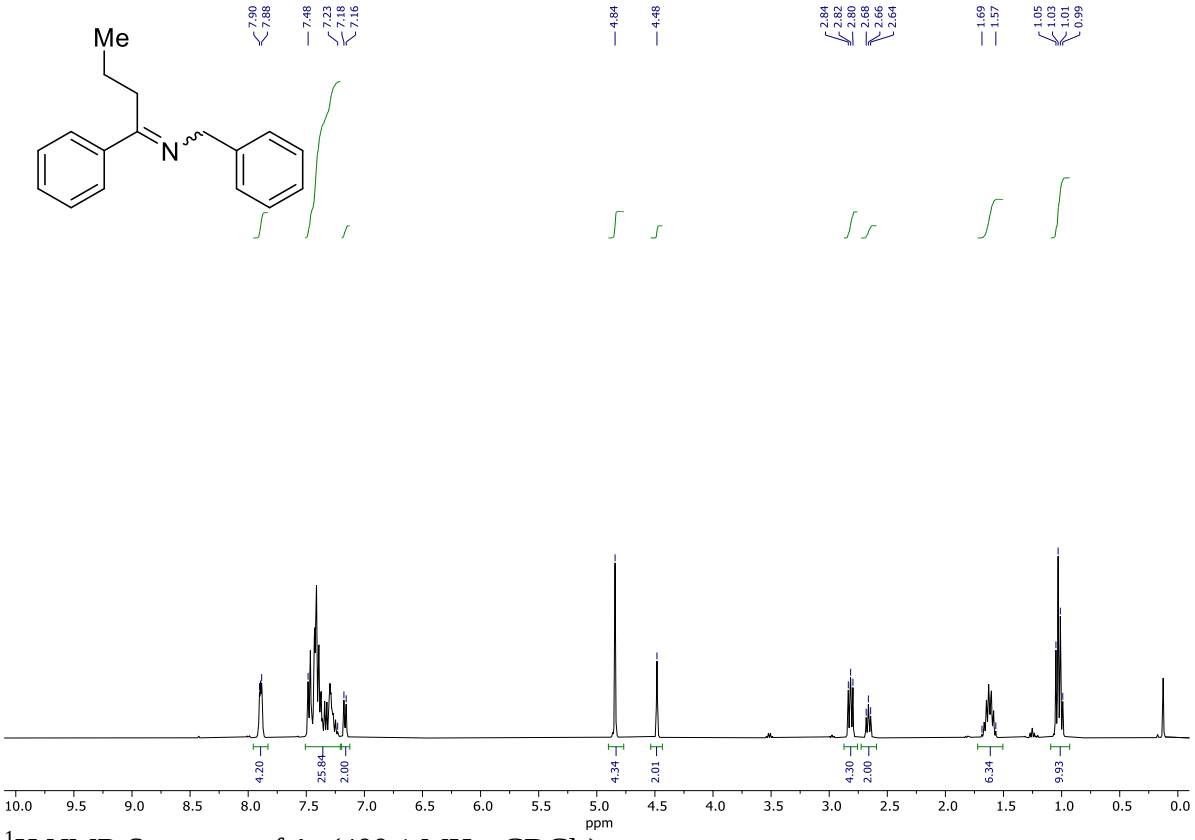
6. NMR Spectra



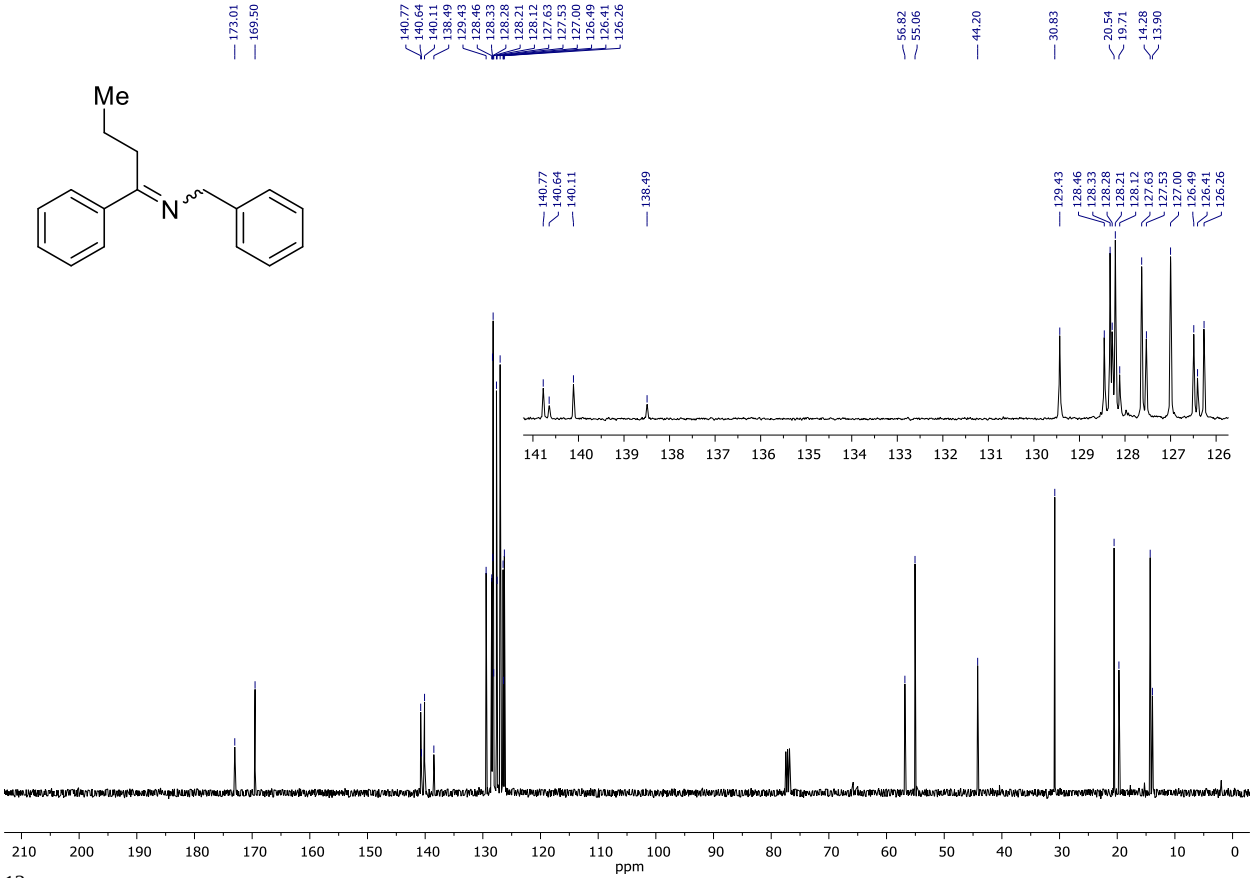
^{1}H NMR Spectrum of **1b** (400.1 MHz, $CDCl_3$)



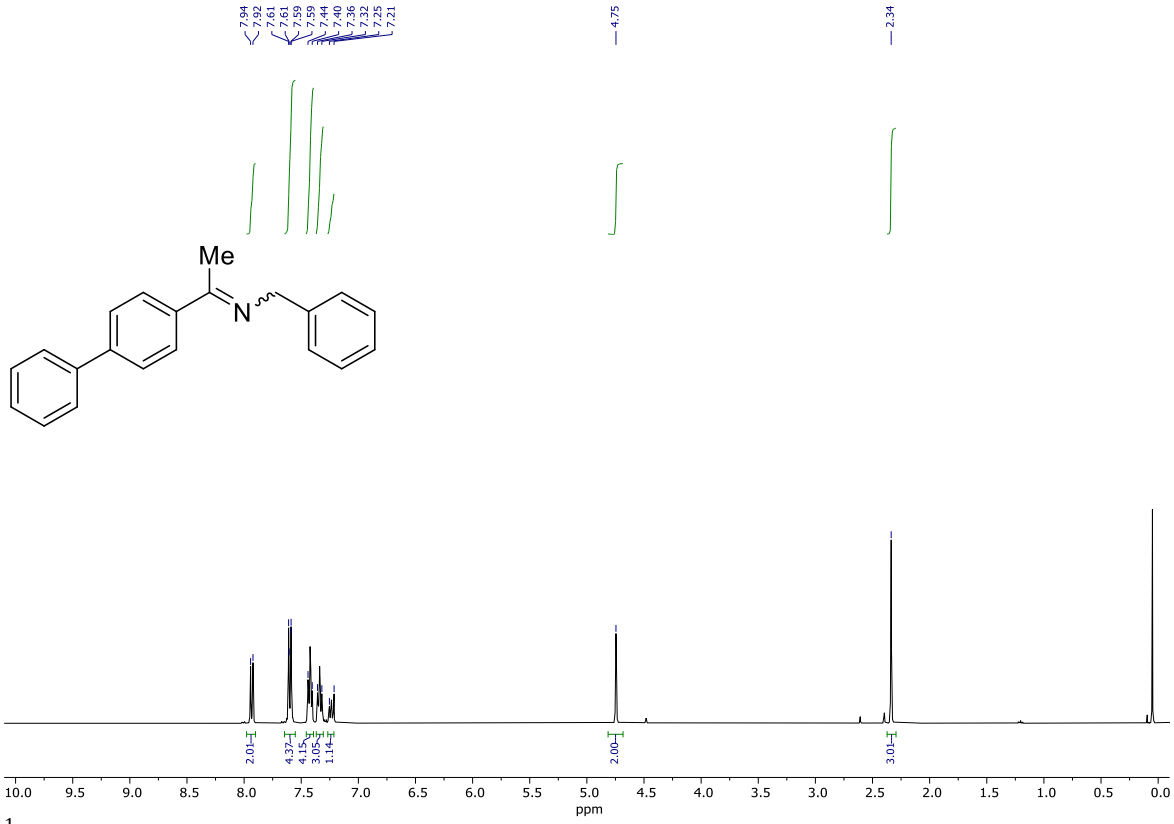
^{13}C NMR Spectrum of **1b** (100.6 MHz, $CDCl_3$)



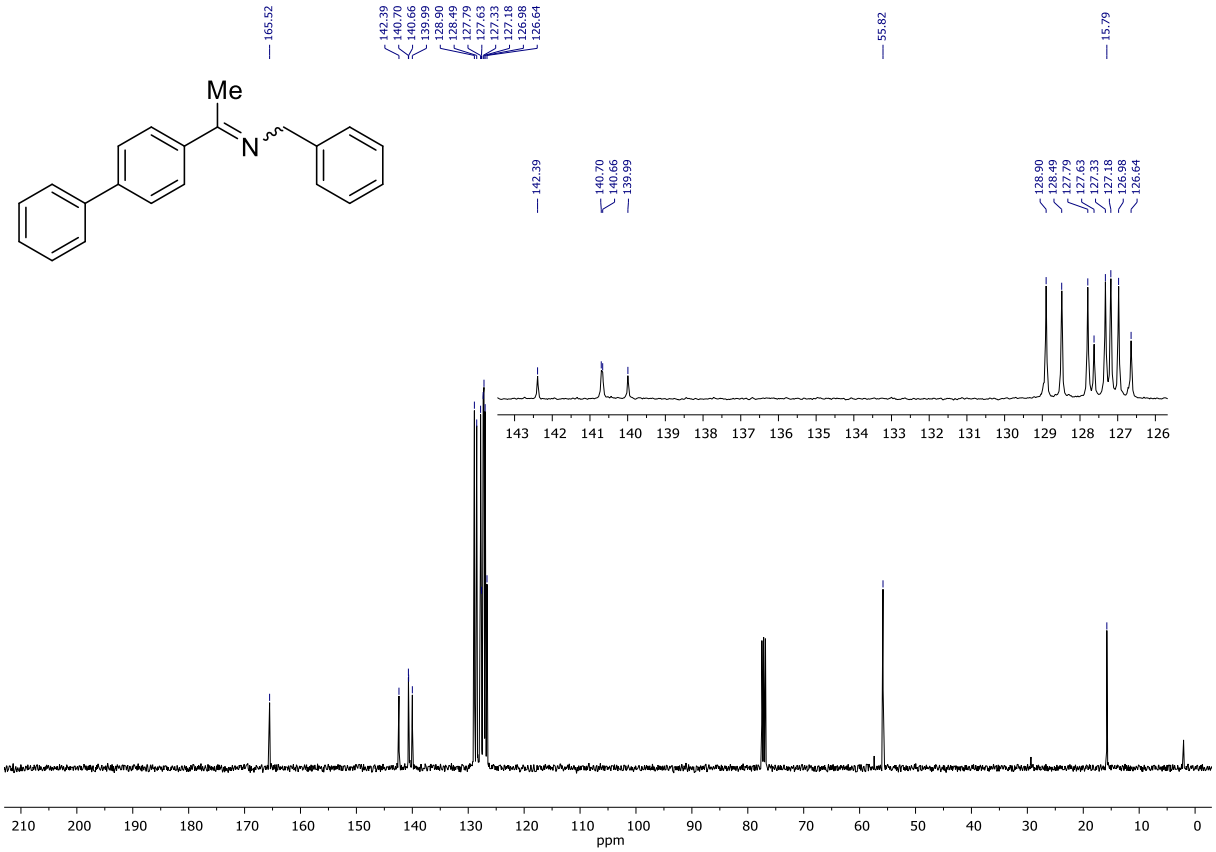
^{1}H NMR Spectrum of **1c** (400.1 MHz, $CDCl_3$)



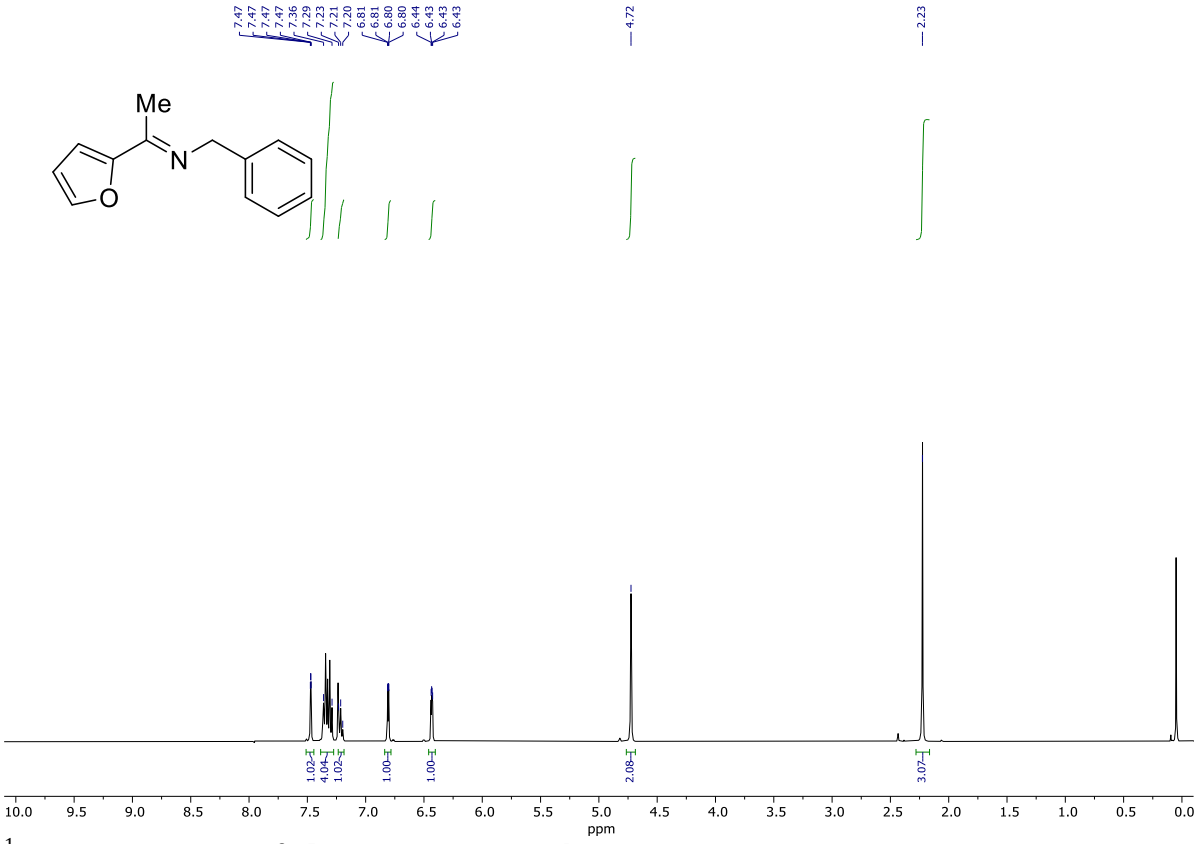
^{13}C NMR Spectrum of **1c** (100.6 MHz, $CDCl_3$)



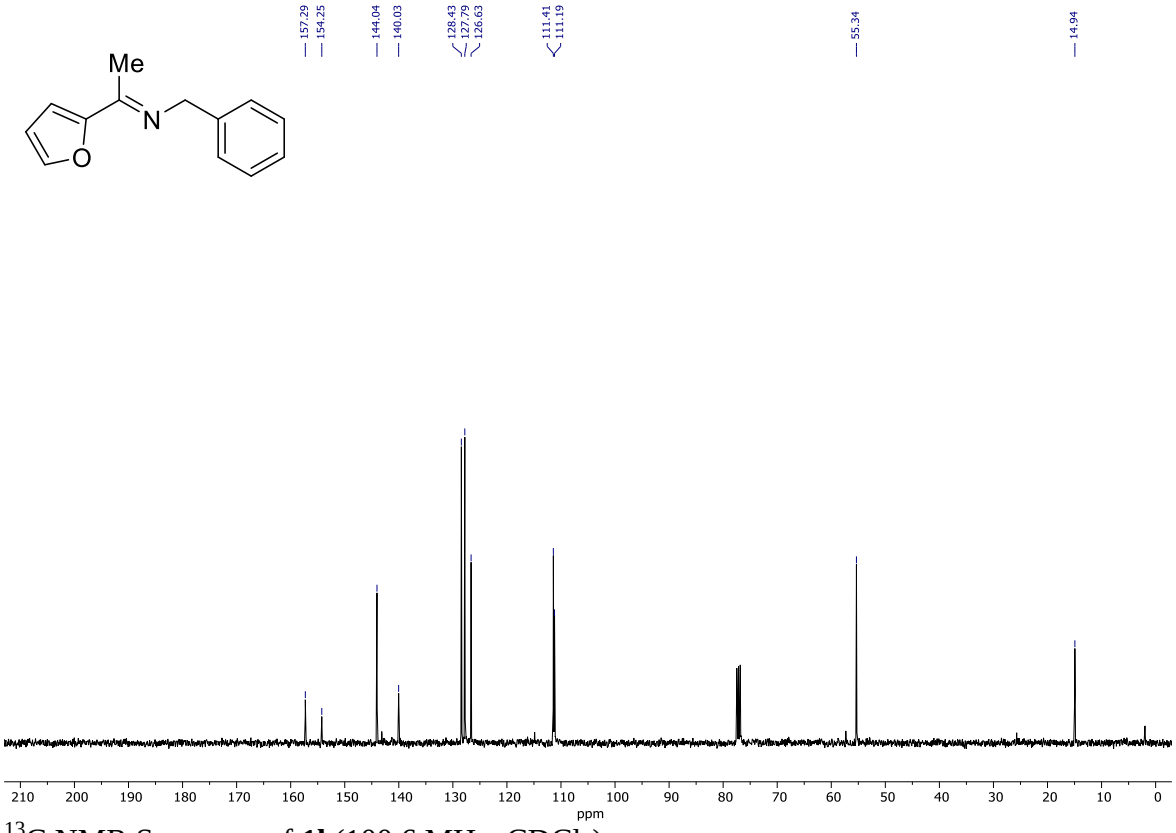
^{1}H NMR Spectrum of **1j** (400.1 MHz, $CDCl_3$)



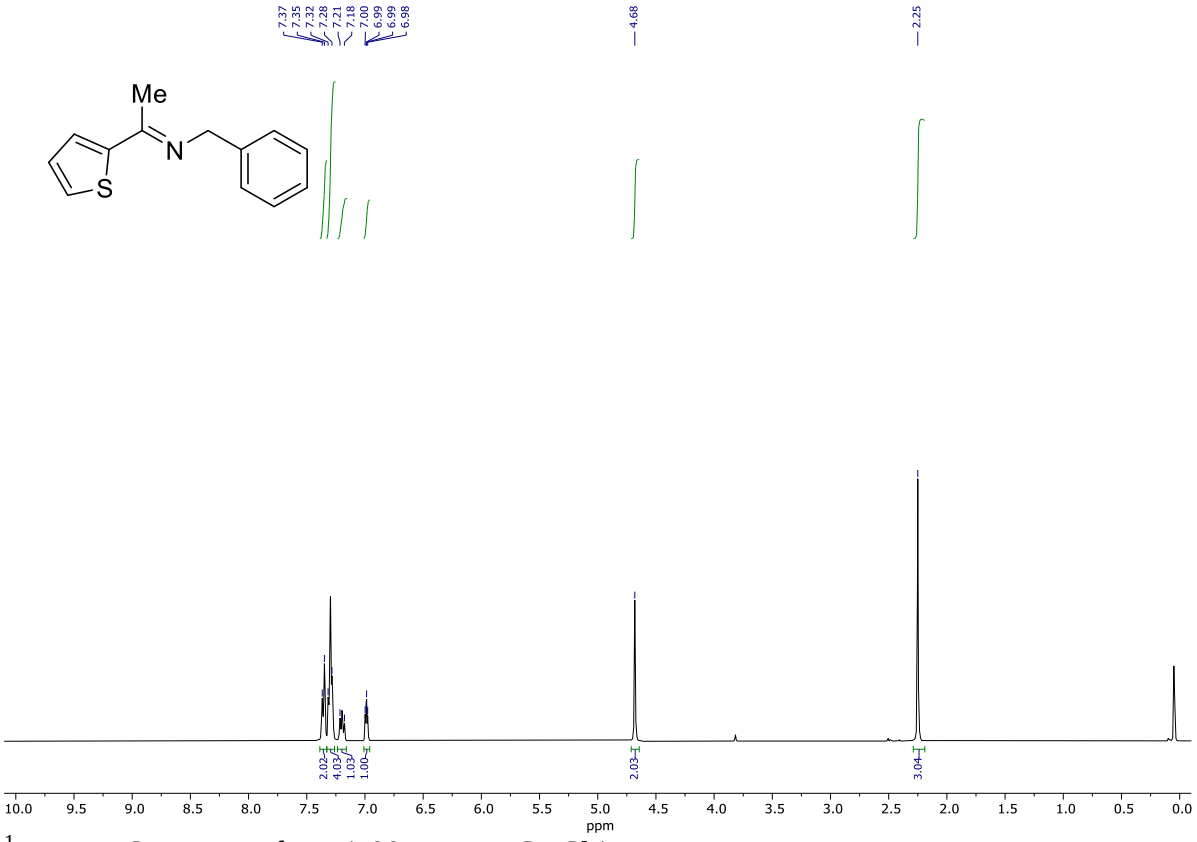
^{13}C NMR Spectrum of **1j** (100.6 MHz, $CDCl_3$)



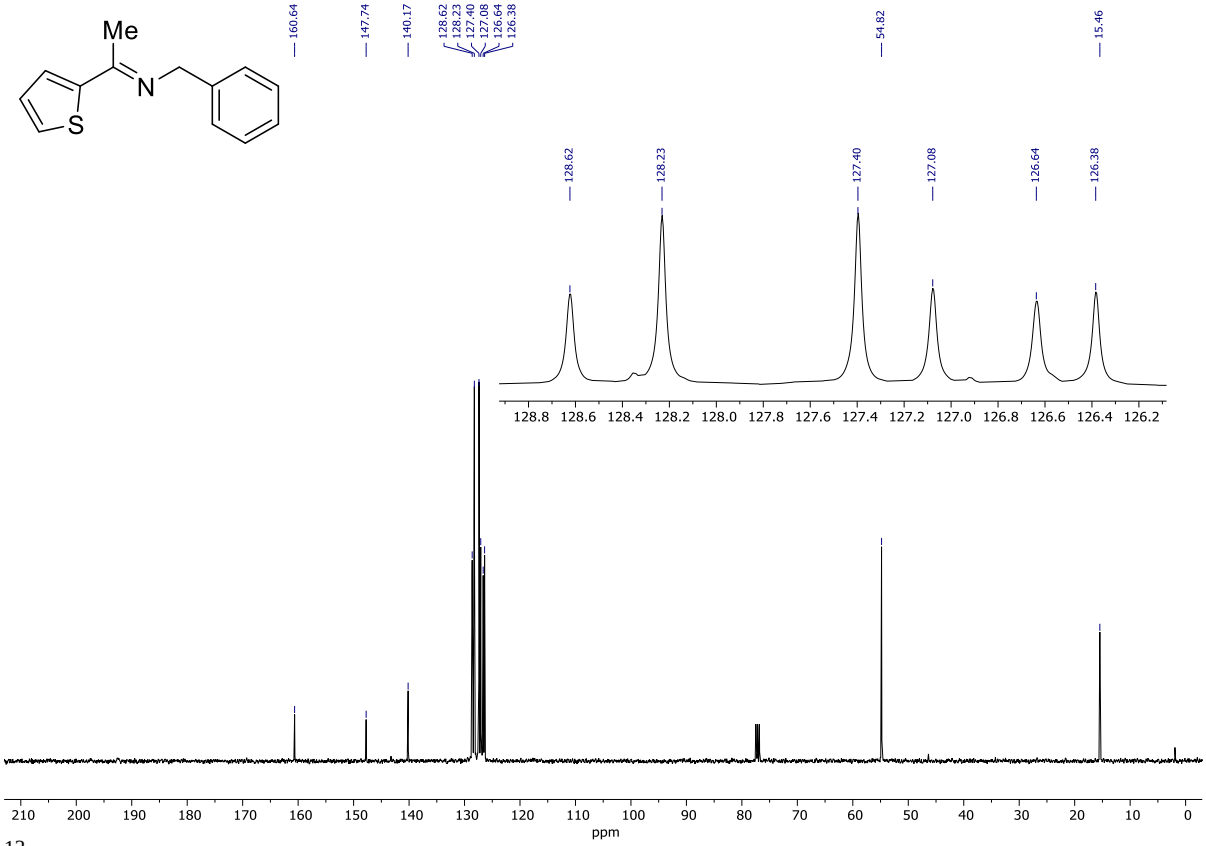
^{1}H NMR Spectrum of **1l** (400.1 MHz, $CDCl_3$)



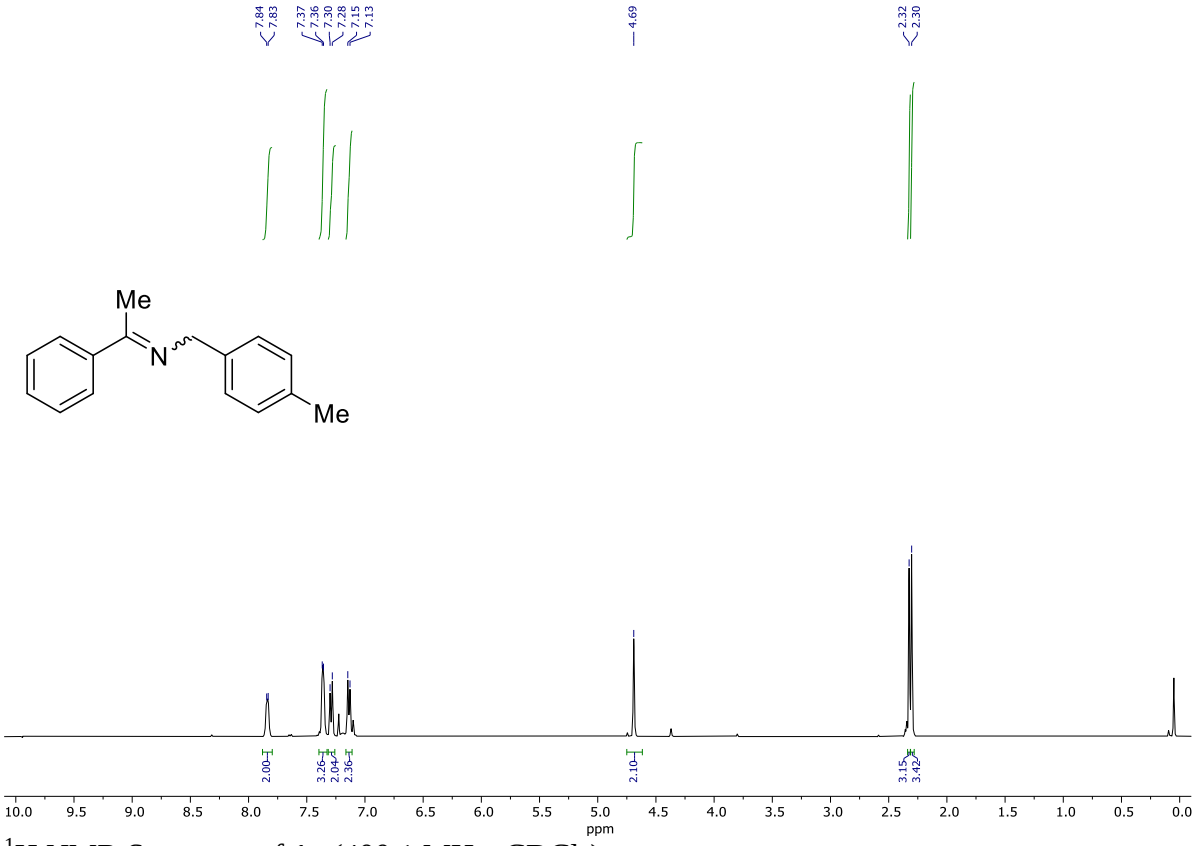
^{13}C NMR Spectrum of **1l** (100.6 MHz, $CDCl_3$)



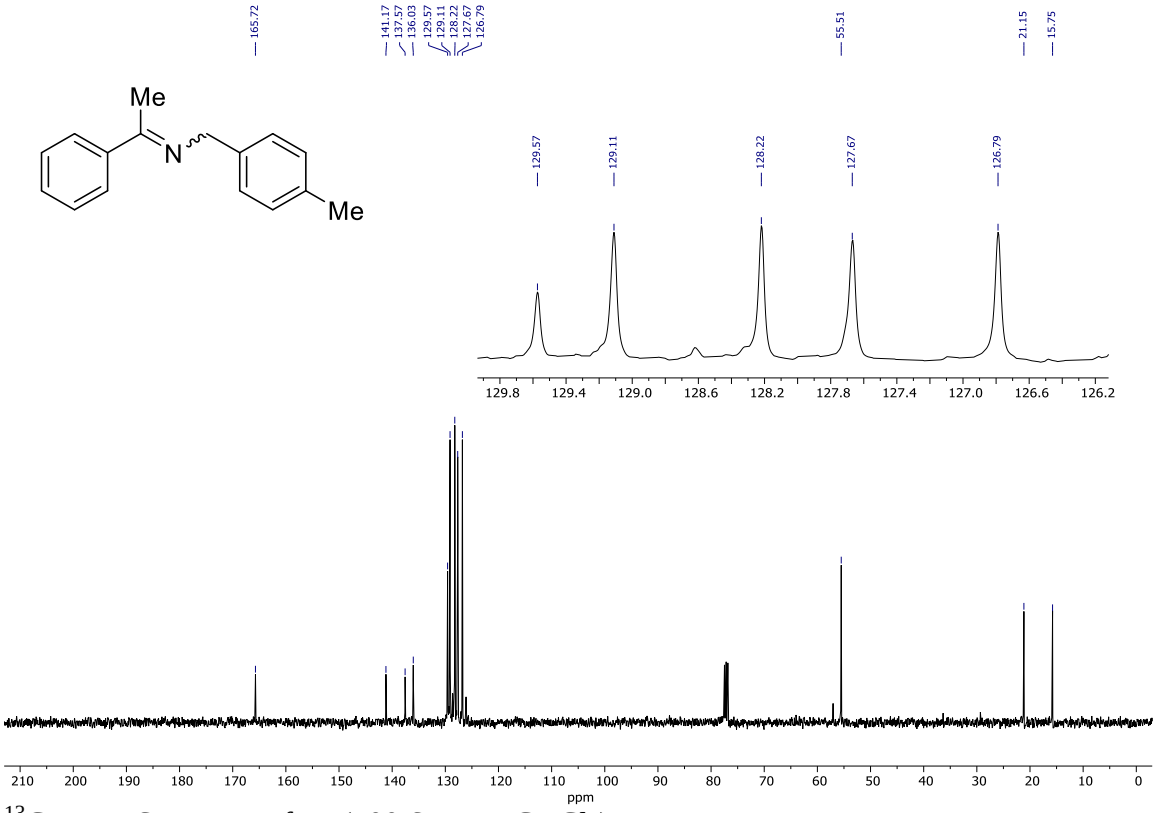
^{1}H NMR Spectrum of **1m** (400.1 MHz, $CDCl_3$)



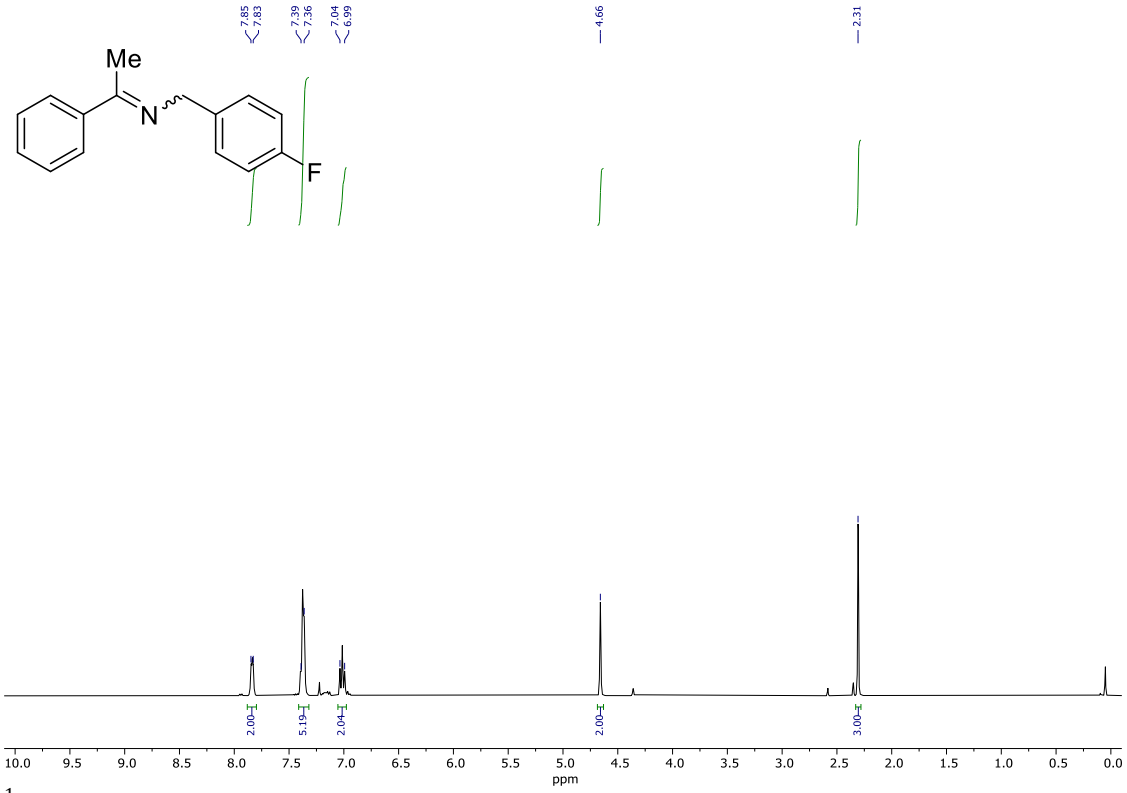
^{13}C NMR Spectrum of **1m** (100.6 MHz, $CDCl_3$)



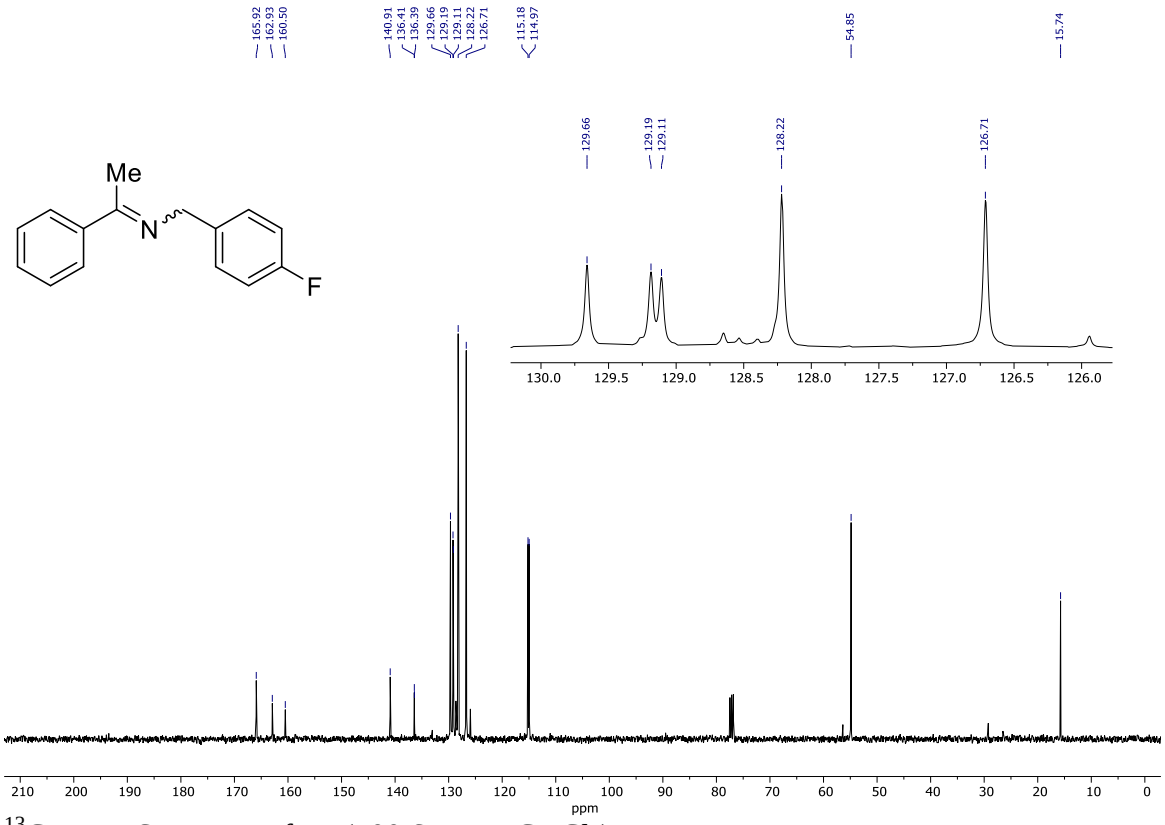
^{1}H NMR Spectrum of **1o** (400.1 MHz, $CDCl_3$)



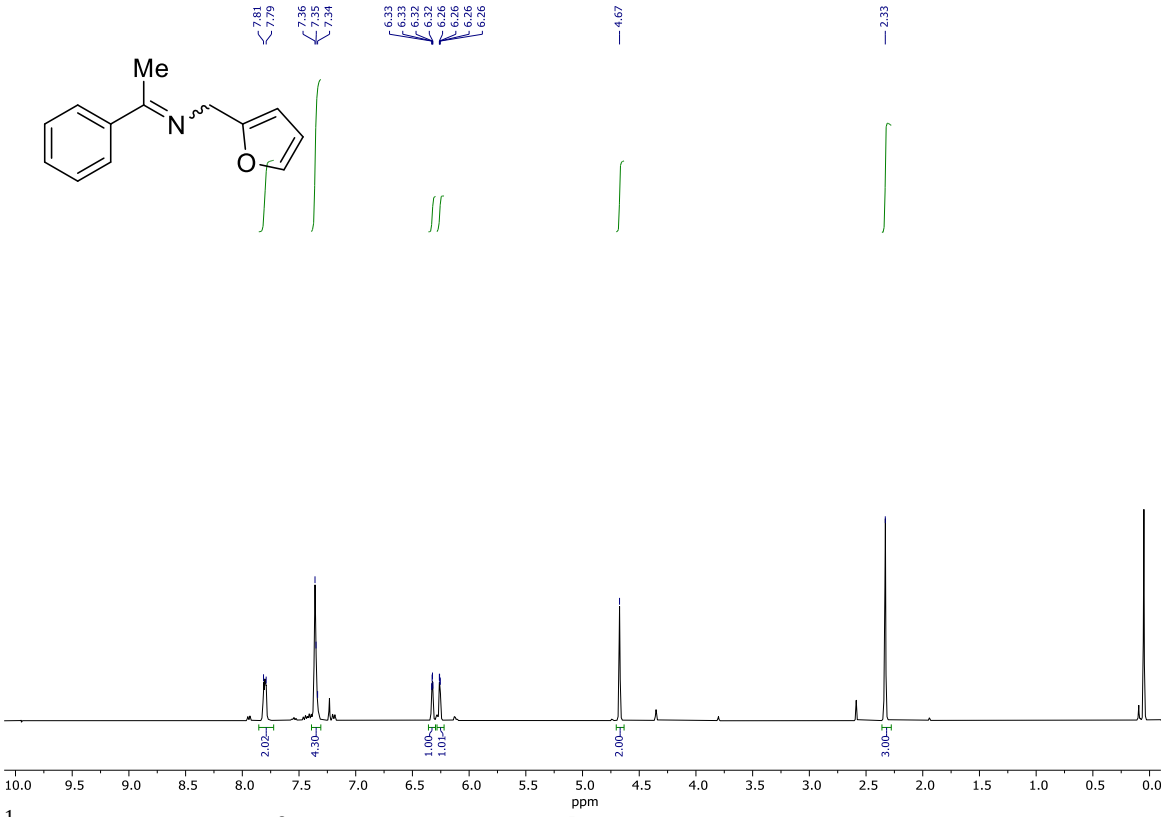
^{13}C NMR Spectrum of **1o** (100.6 MHz, $CDCl_3$)



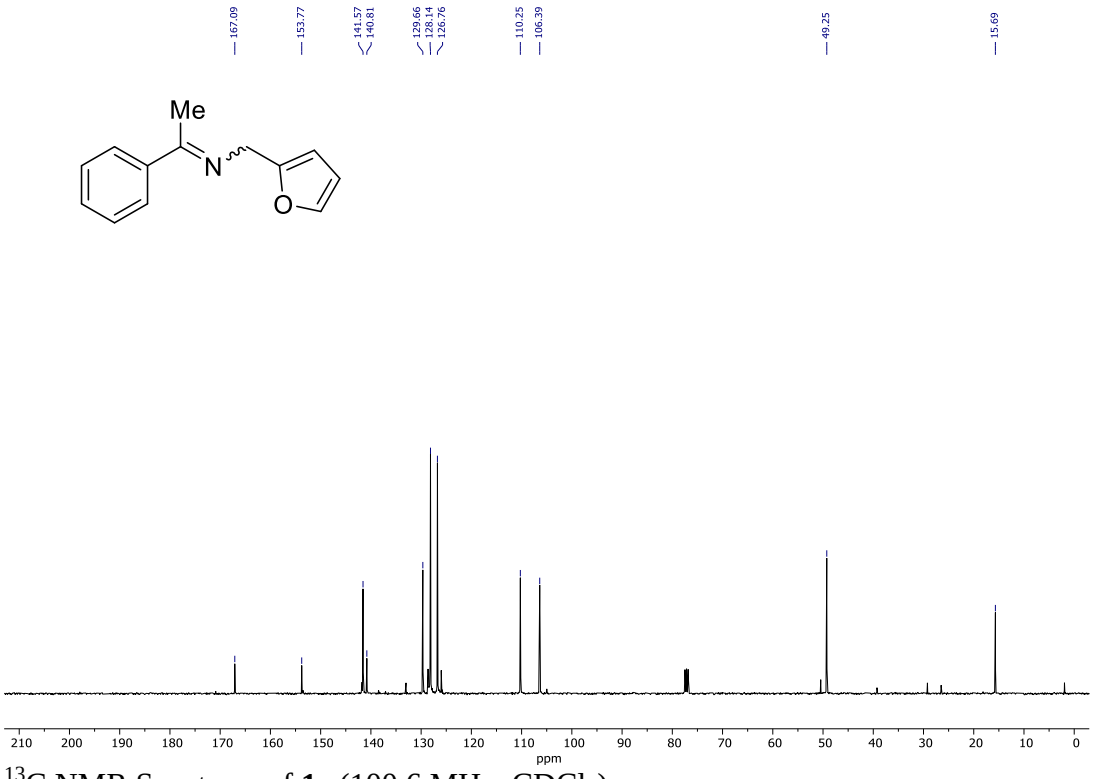
^{1}H NMR Spectrum of **1q** (400.1 MHz, $CDCl_3$)



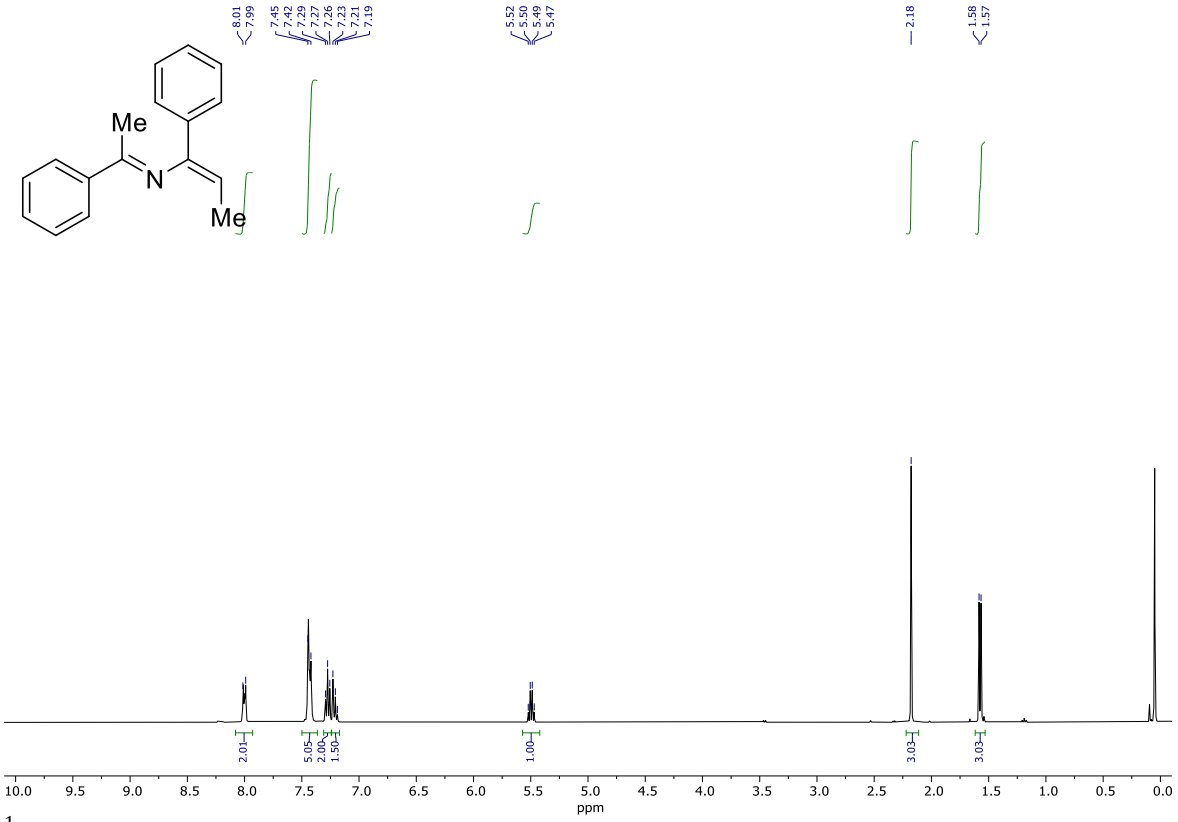
^{13}C NMR Spectrum of **1q** (100.6 MHz, $CDCl_3$)



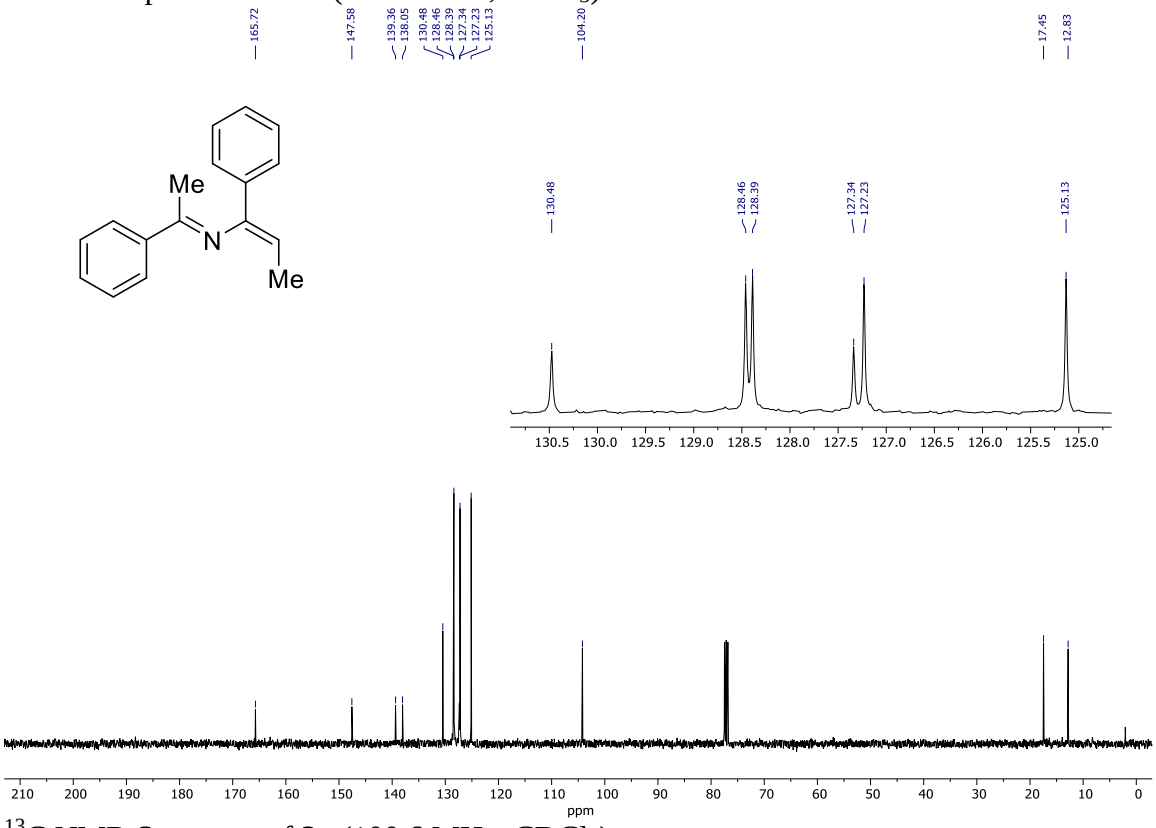
^{1}H NMR Spectrum of **1r** (400.1 MHz, $CDCl_3$)



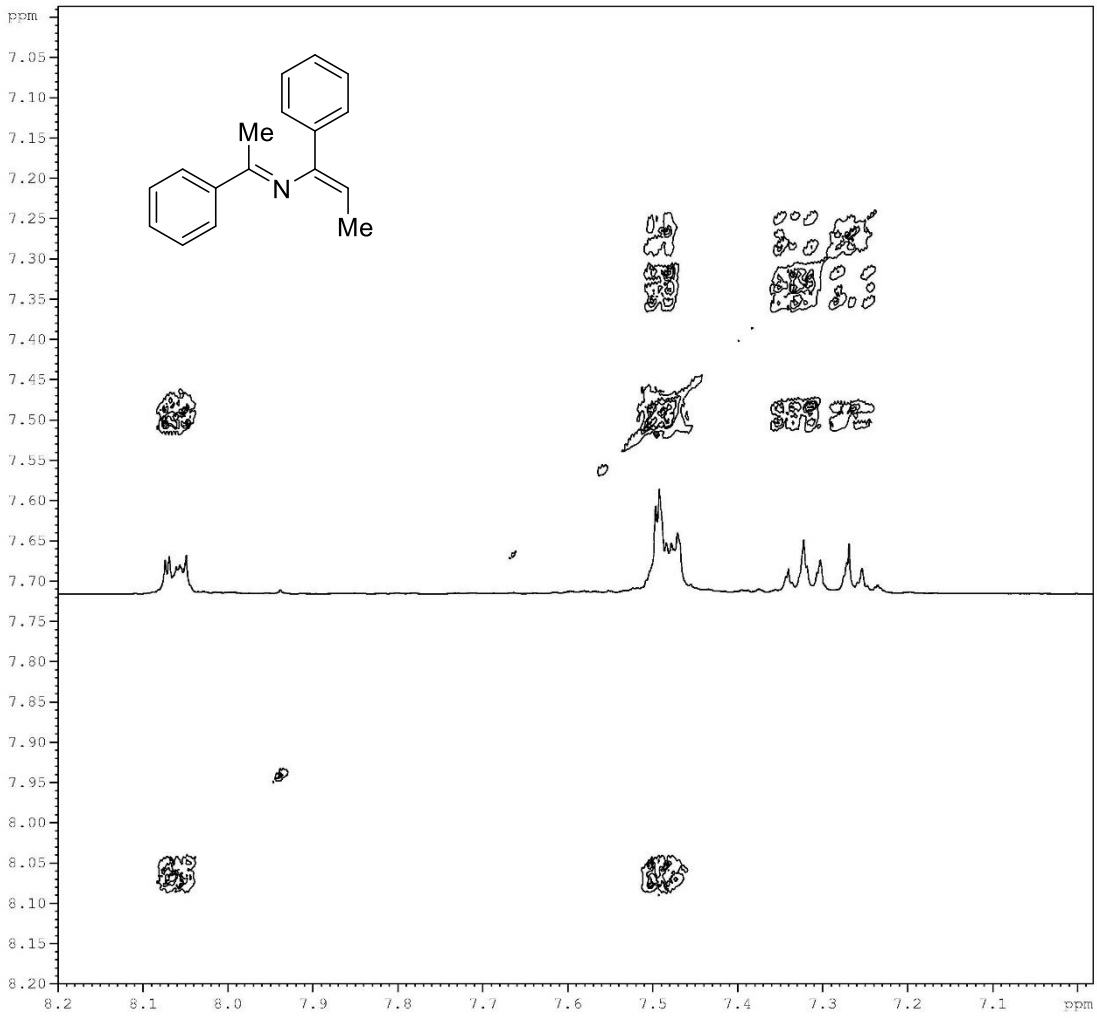
^{13}C NMR Spectrum of **1r** (100.6 MHz, $CDCl_3$)



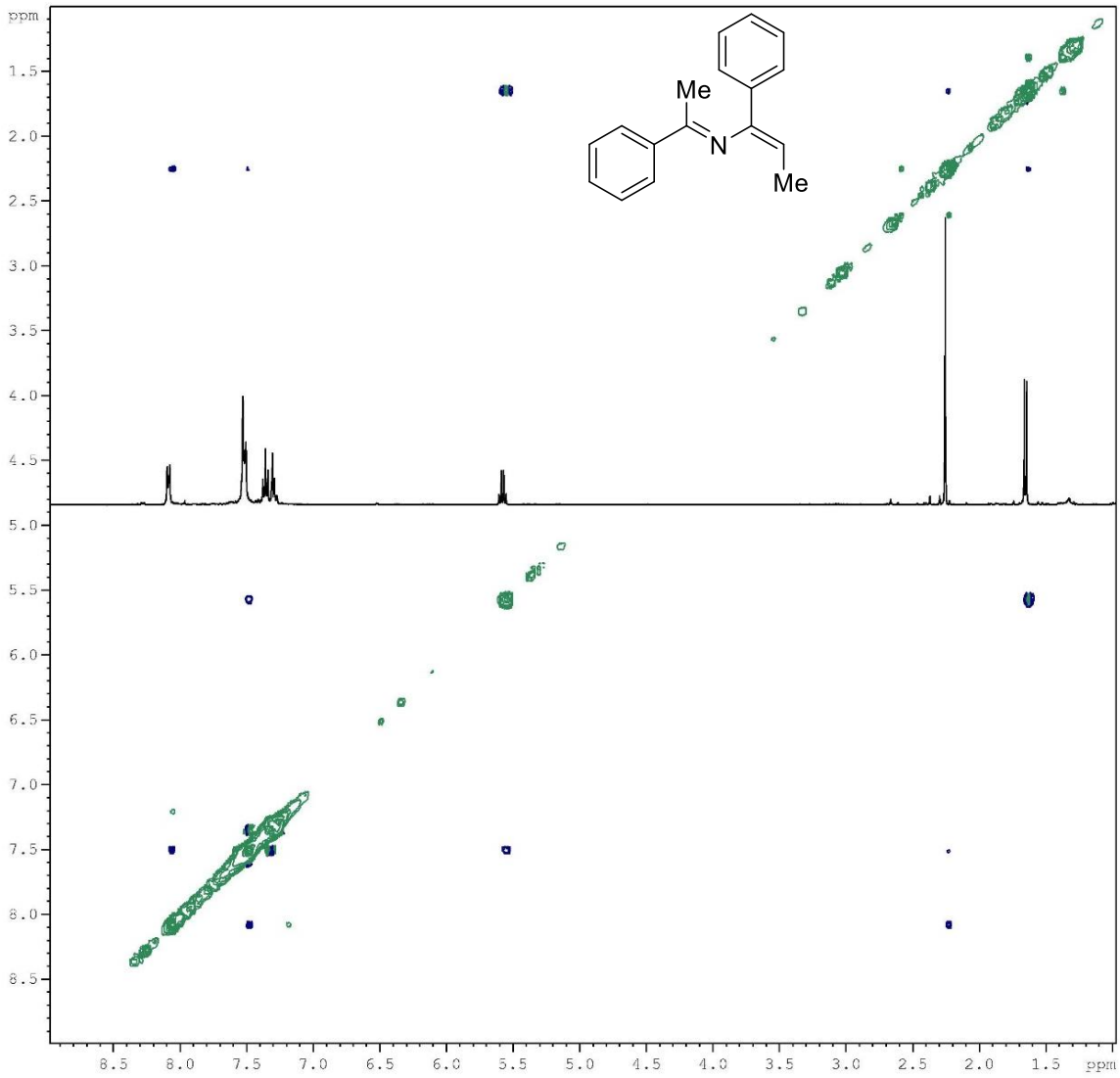
^{1}H NMR Spectrum of **2a** (400.1 MHz, $CDCl_3$)



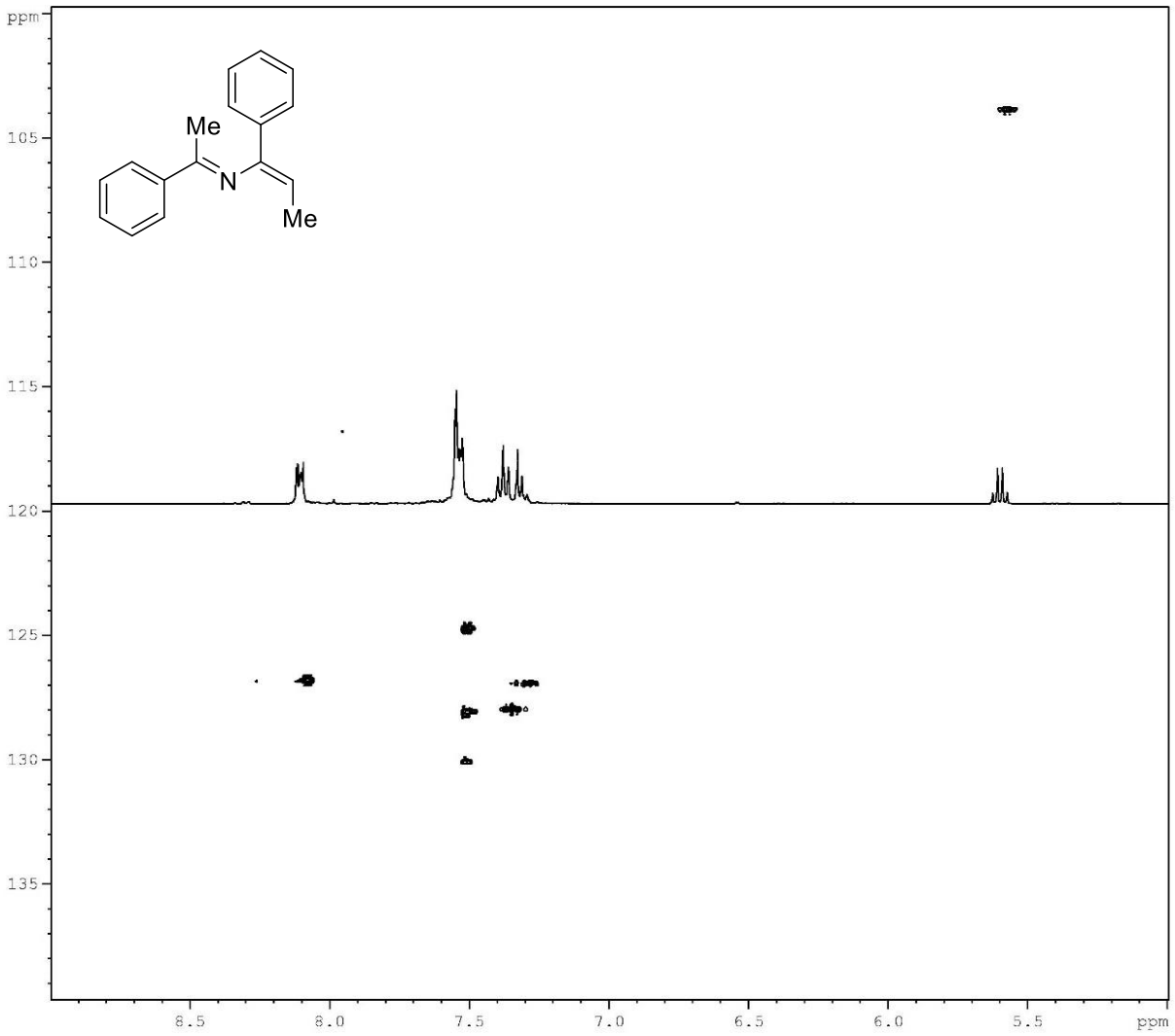
^{13}C NMR Spectrum of **2a** (100.6 MHz, $CDCl_3$)



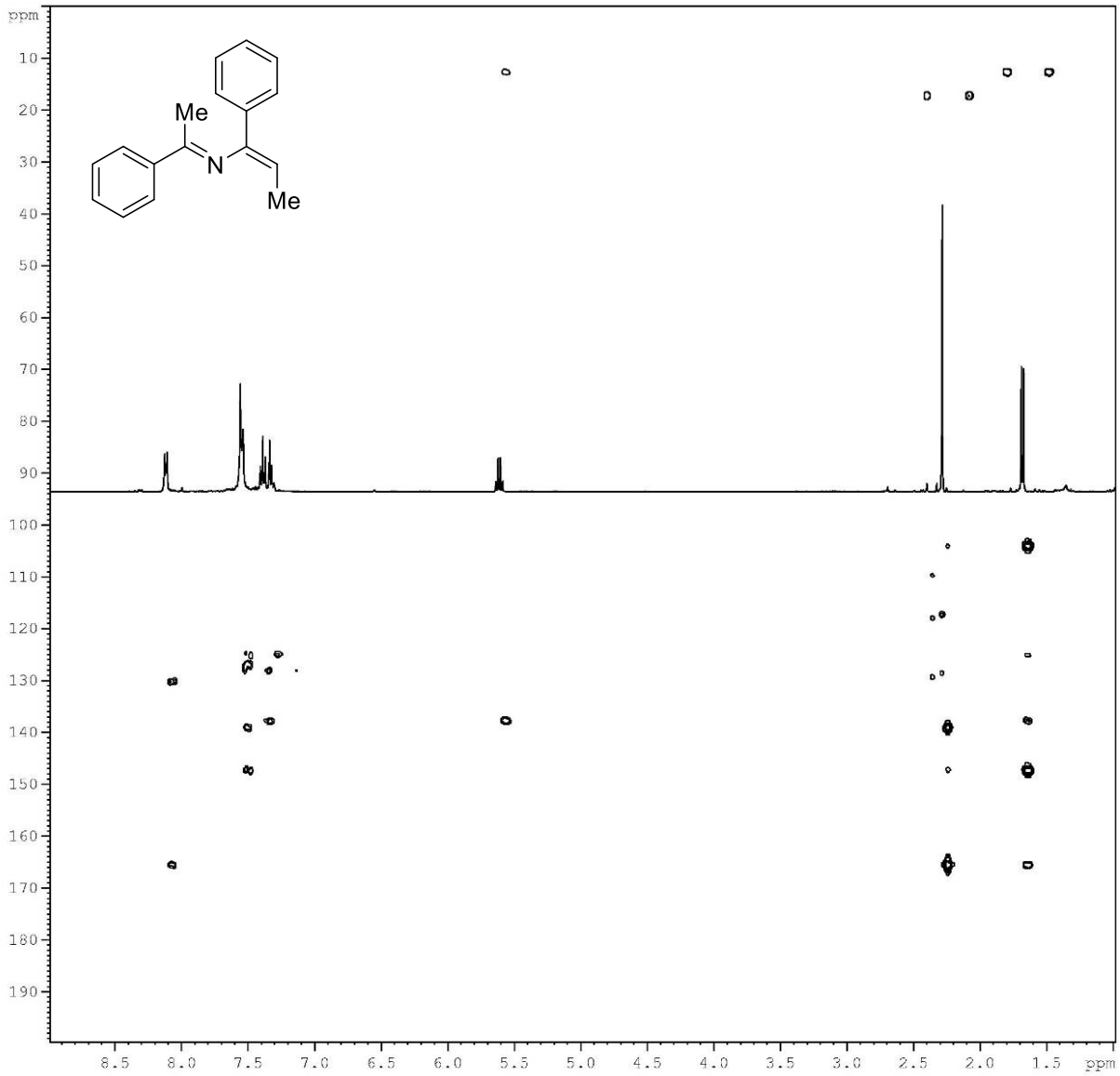
2D COSY Spectrum of **2a**



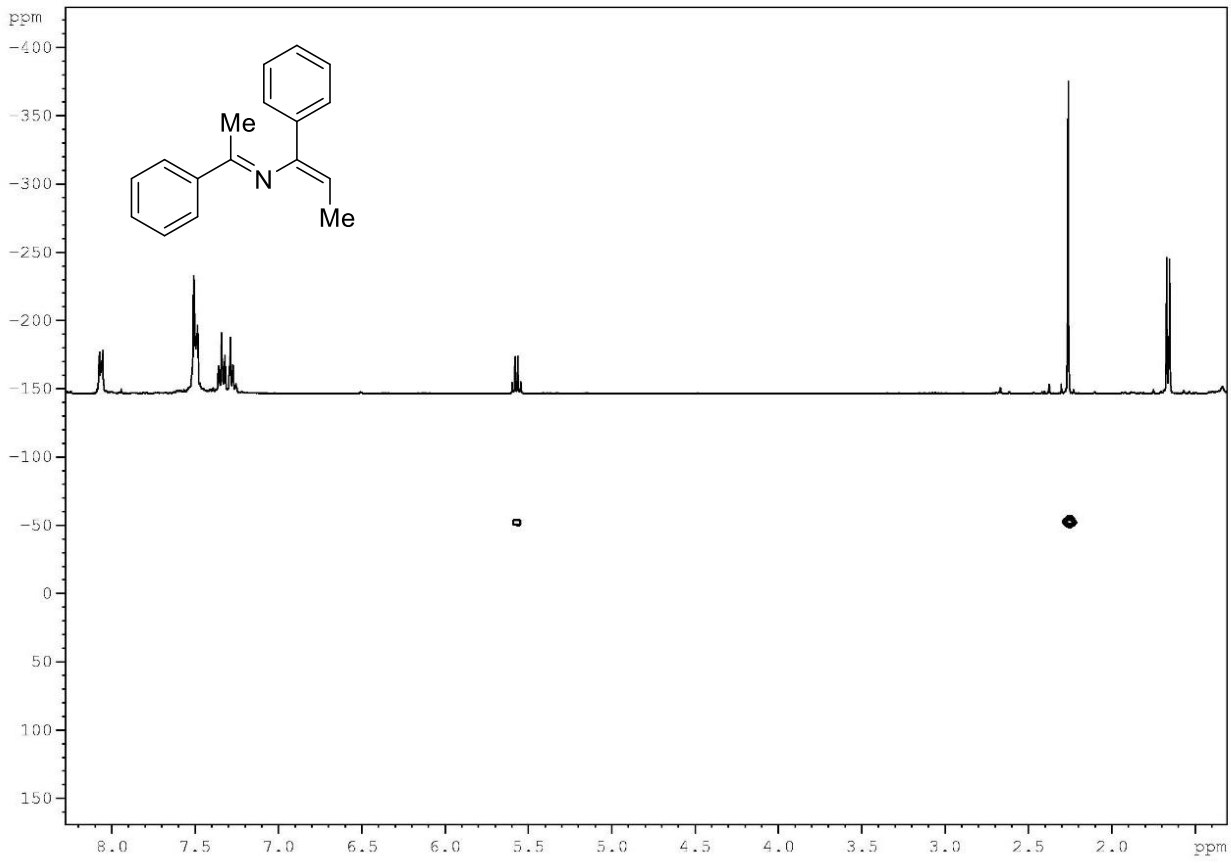
2D NOESY Spectrum of **2a**



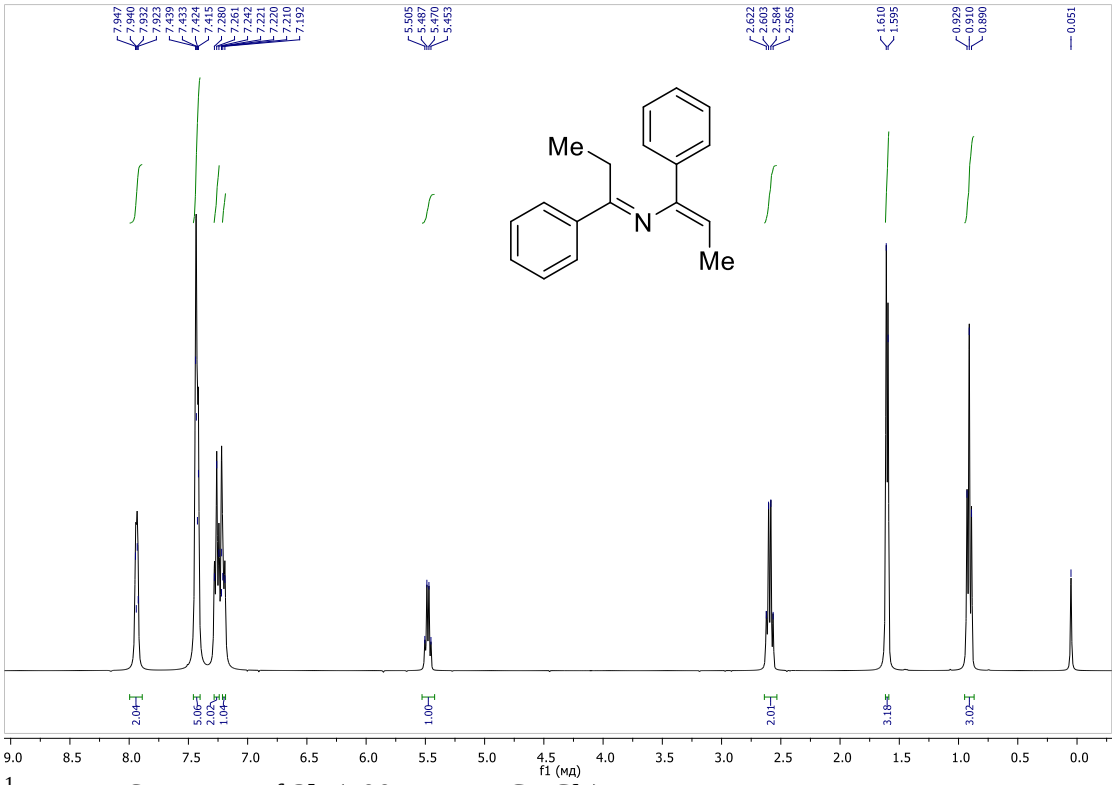
2D ^{1}H-^{13}C HSQC Spectrum of **2a**



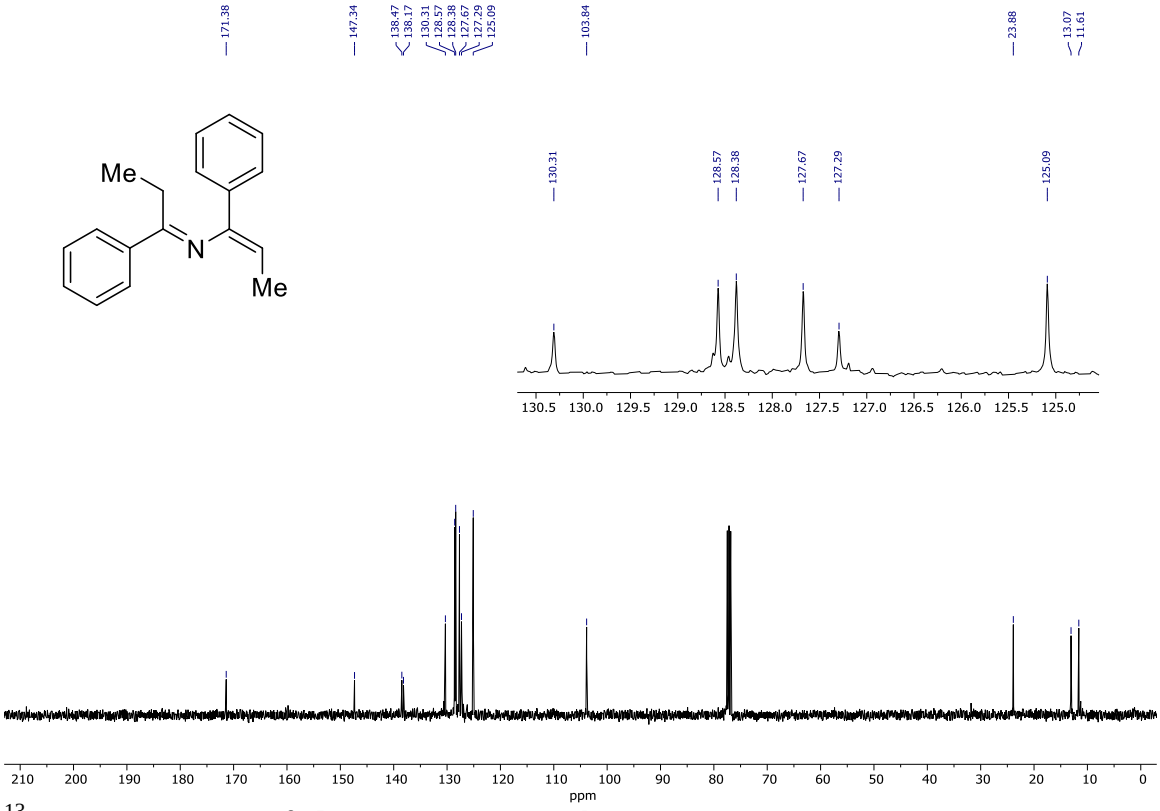
2D ^{1}H-^{13}C HMBC Spectrum of **2a**



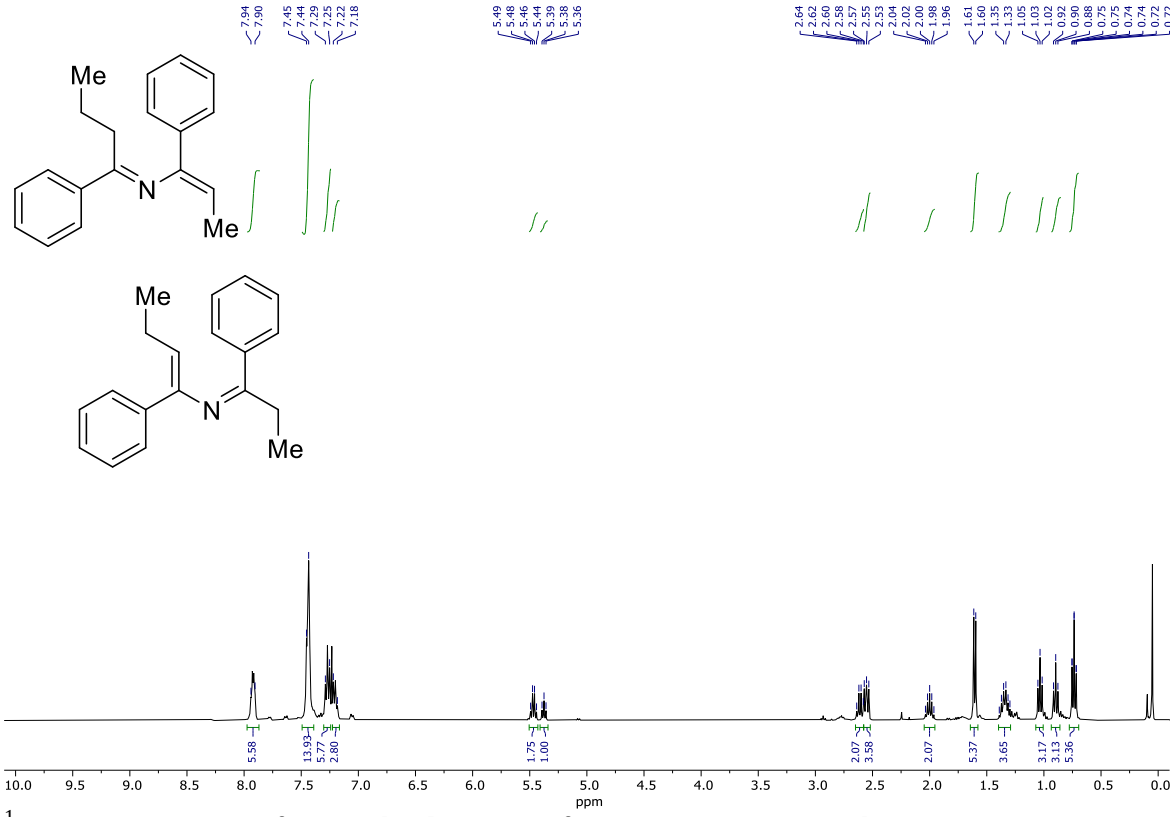
2D ^{1}H-^{15}N HMBC Spectrum of **2a**



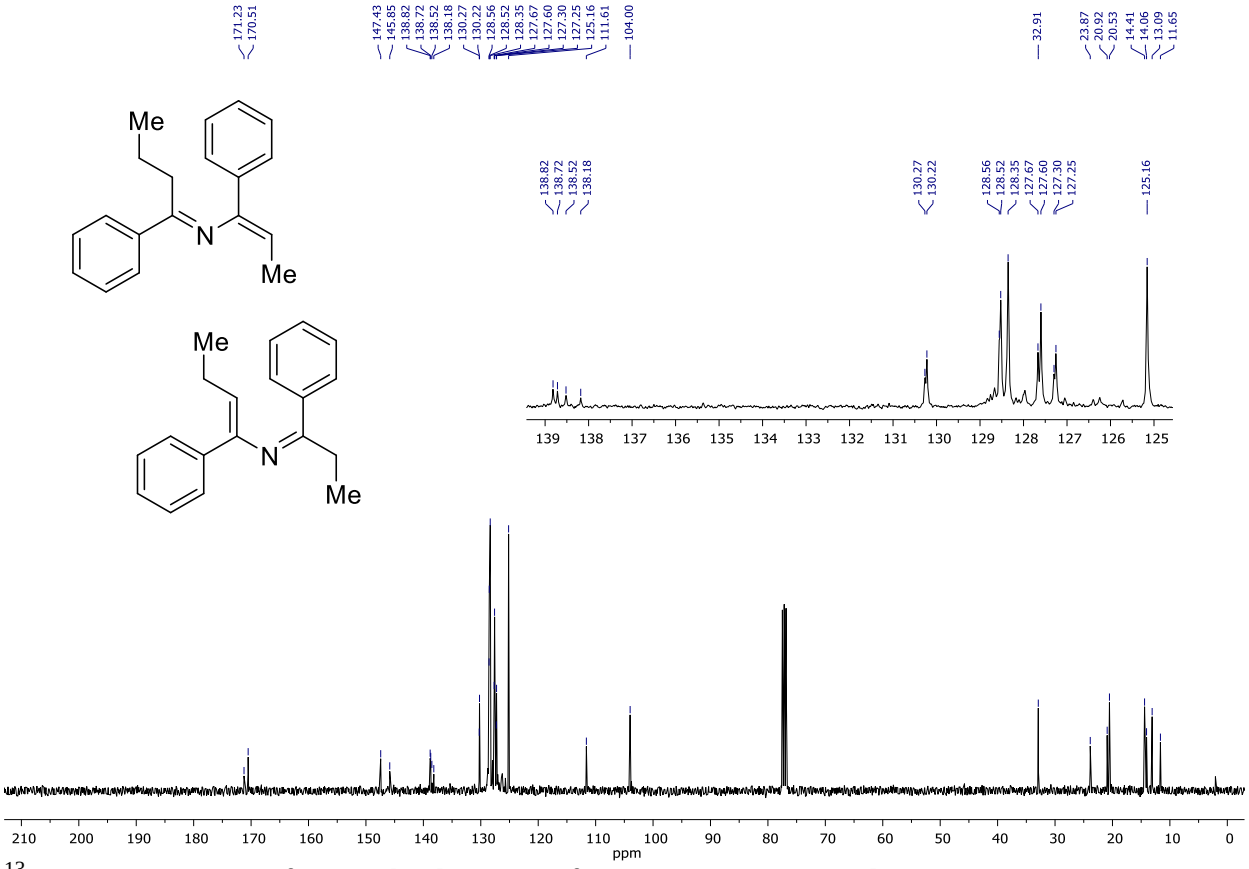
^{1}H NMR Spectrum of **2b** (400.1 MHz, $CDCl_3$)



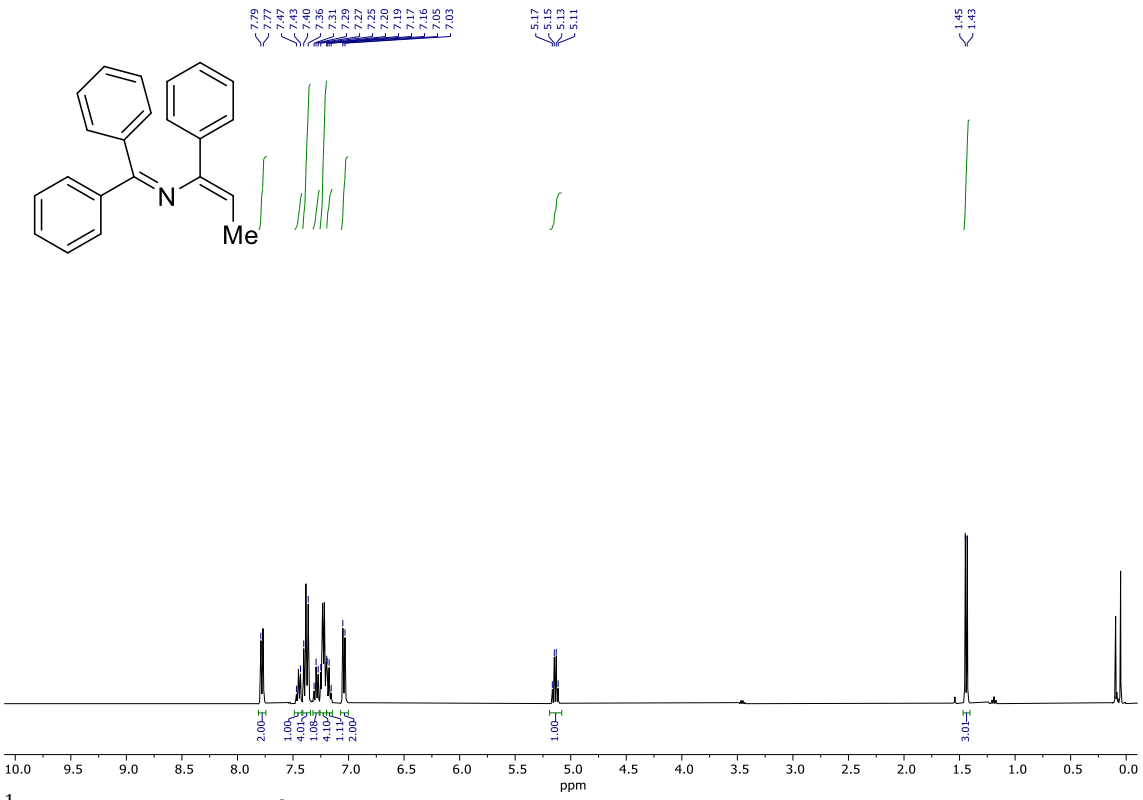
^{13}C NMR Spectrum of **2b** (100.6 MHz, $CDCl_3$)



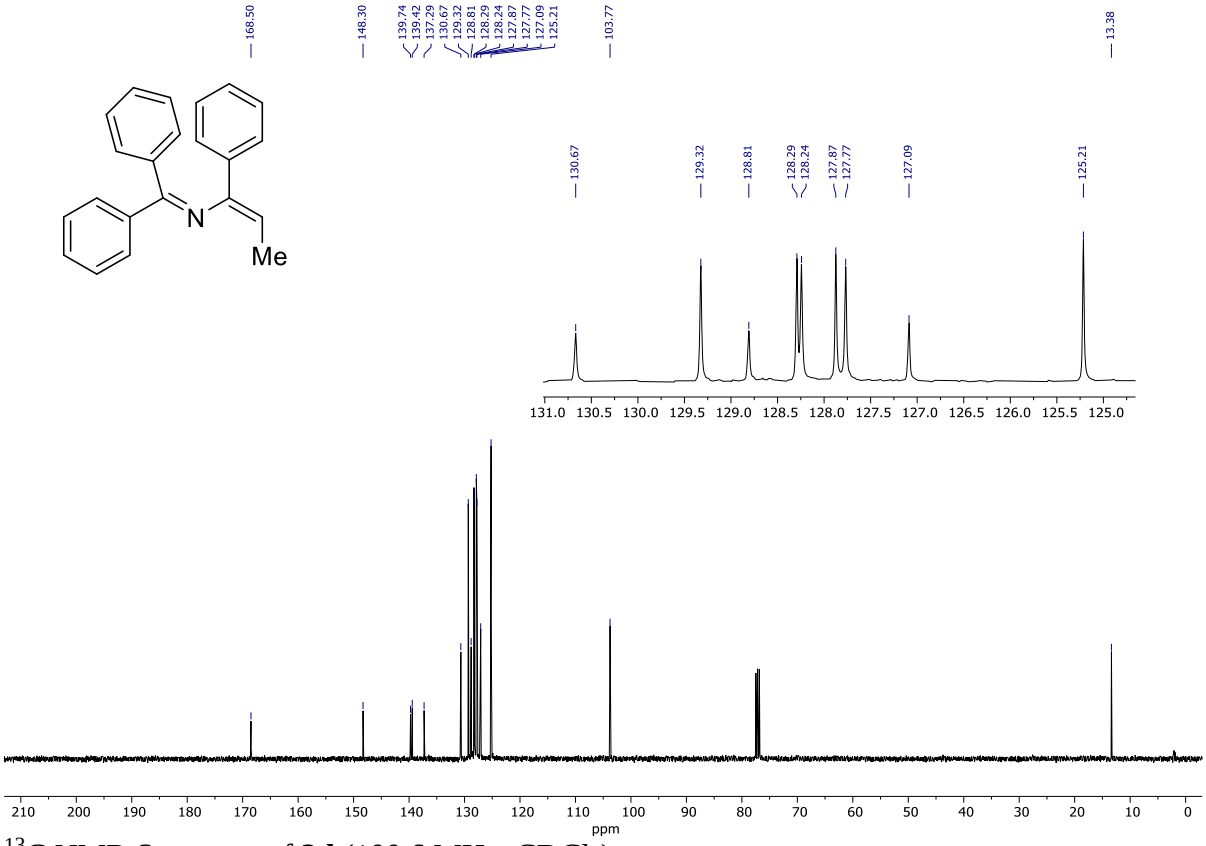
^{1}H NMR Spectrum of **2c** (with admixture of **2c'**, 400.1 MHz, $CDCl_3$)



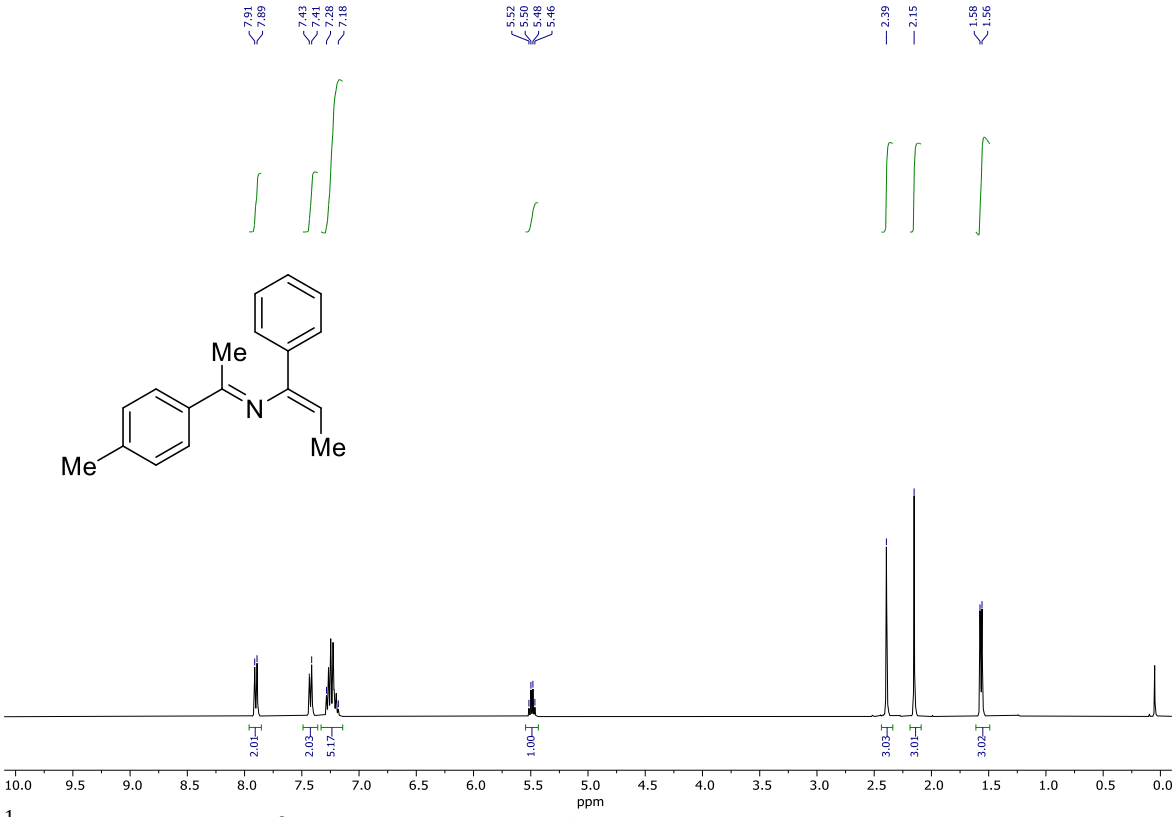
^{13}C NMR Spectrum of **2c** (with admixture of **2c'**, 100.6 MHz, $CDCl_3$)



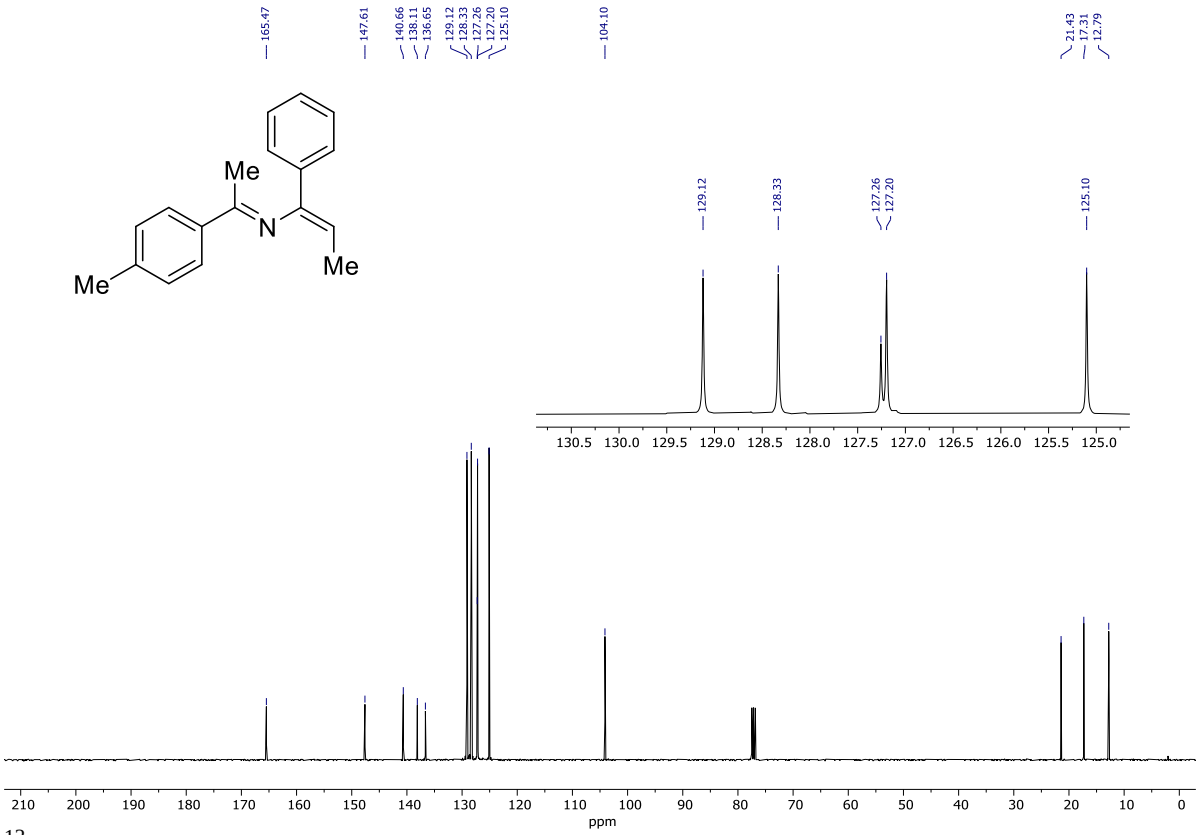
^{1}H NMR Spectrum of **2d** (400.1 MHz, $CDCl_3$)



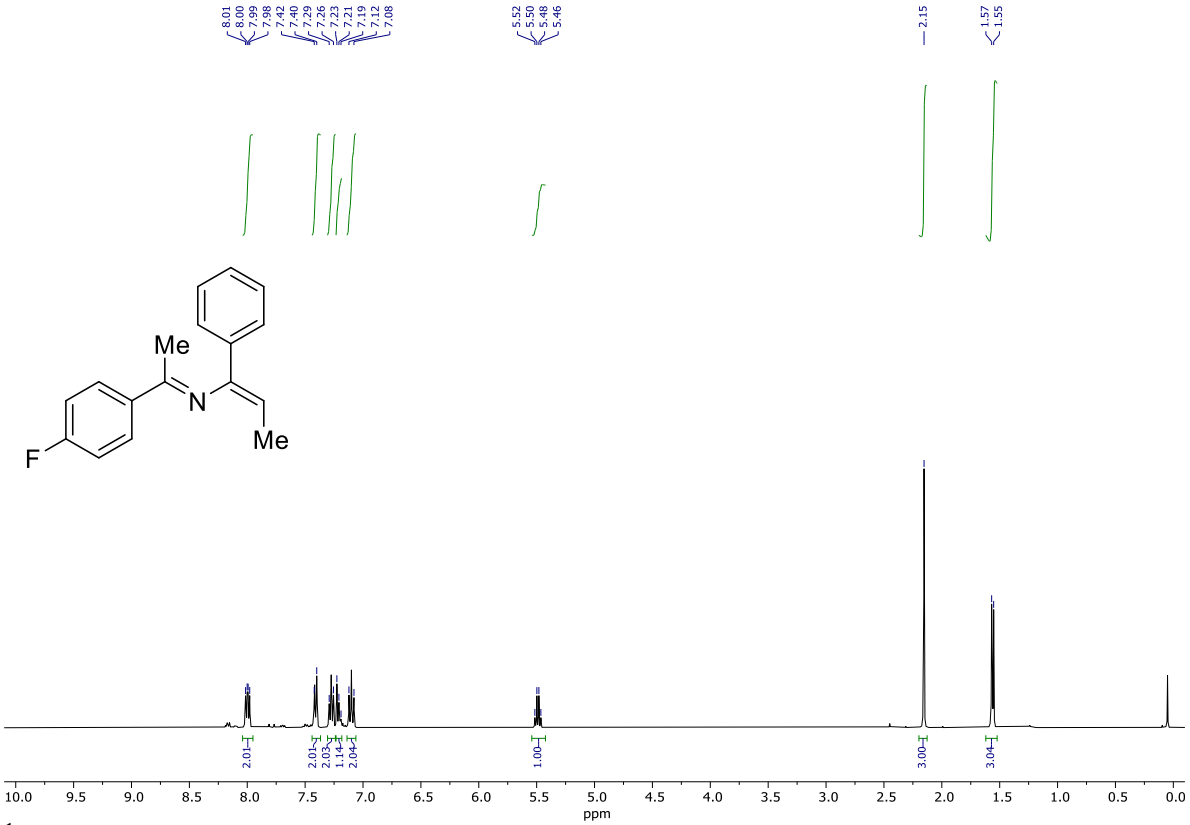
^{13}C NMR Spectrum of **2d** (100.6 MHz, $CDCl_3$)



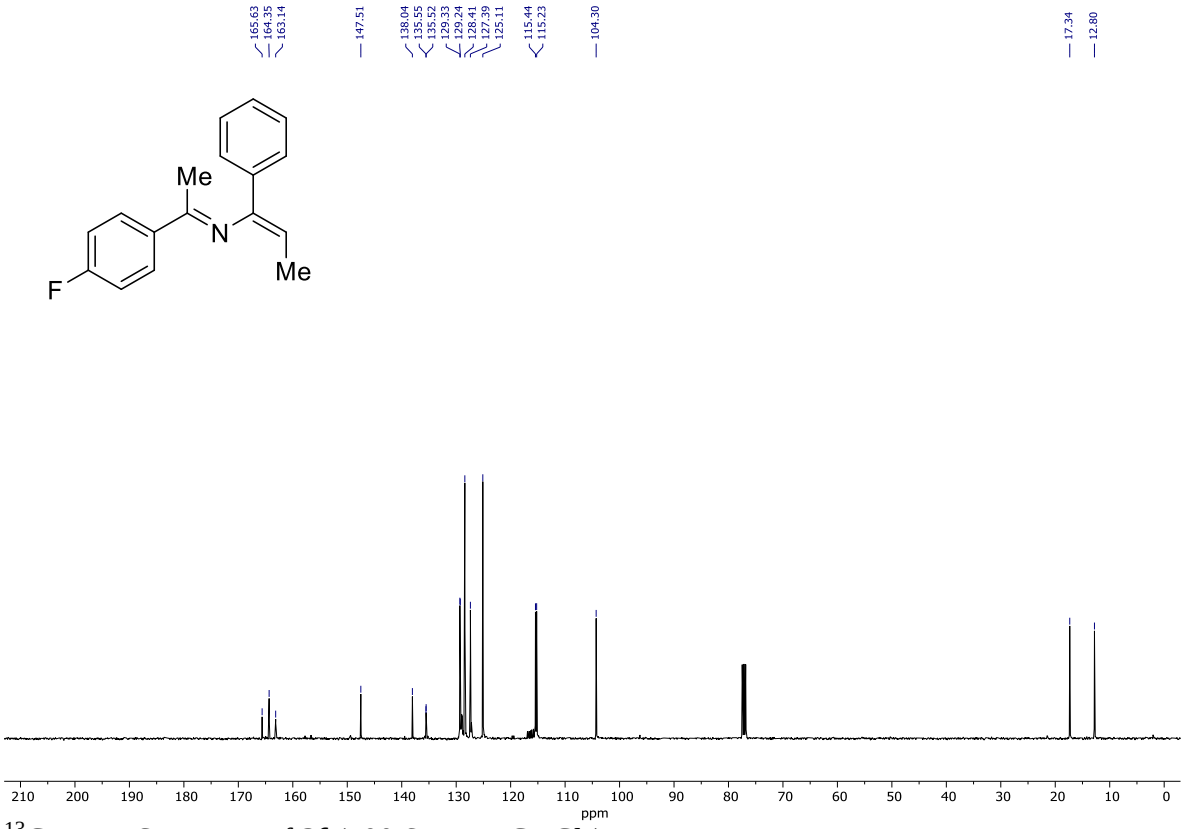
^{1}H NMR Spectrum of **2e** (400.1 MHz, $CDCl_3$)



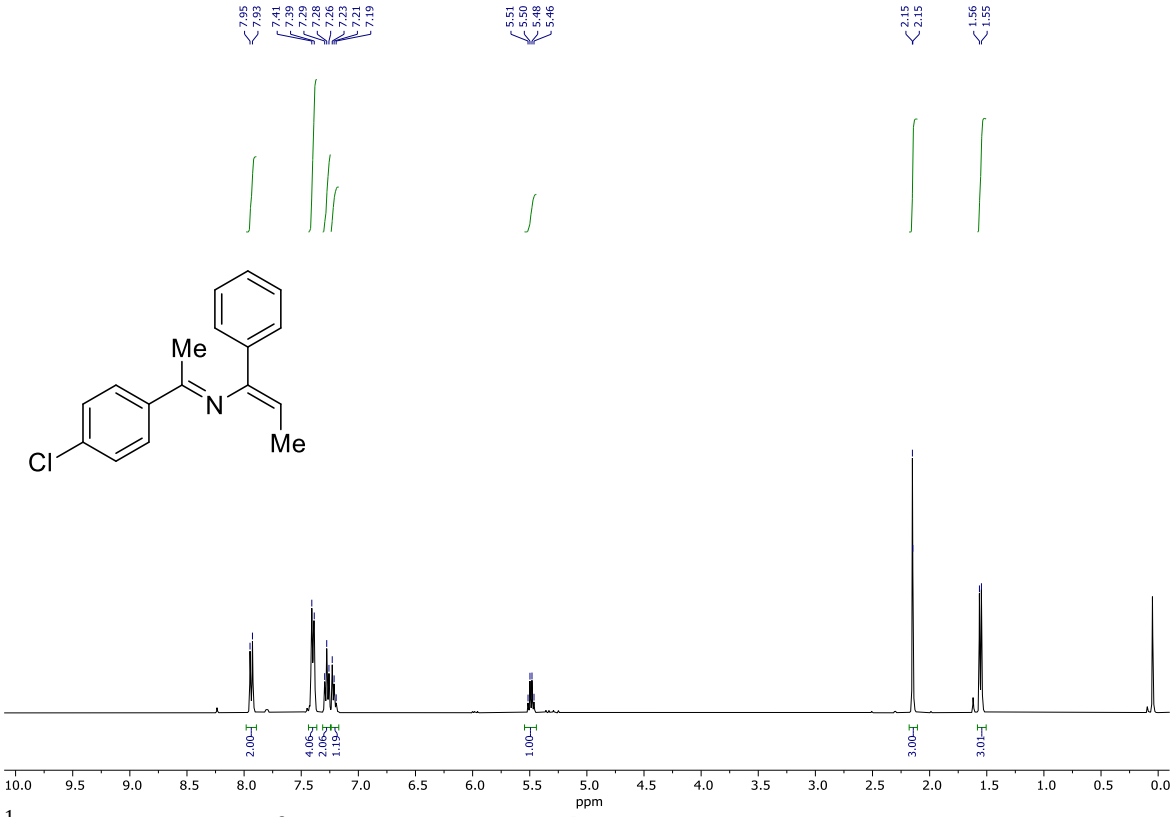
^{13}C NMR Spectrum of **2e** (100.6 MHz, $CDCl_3$)



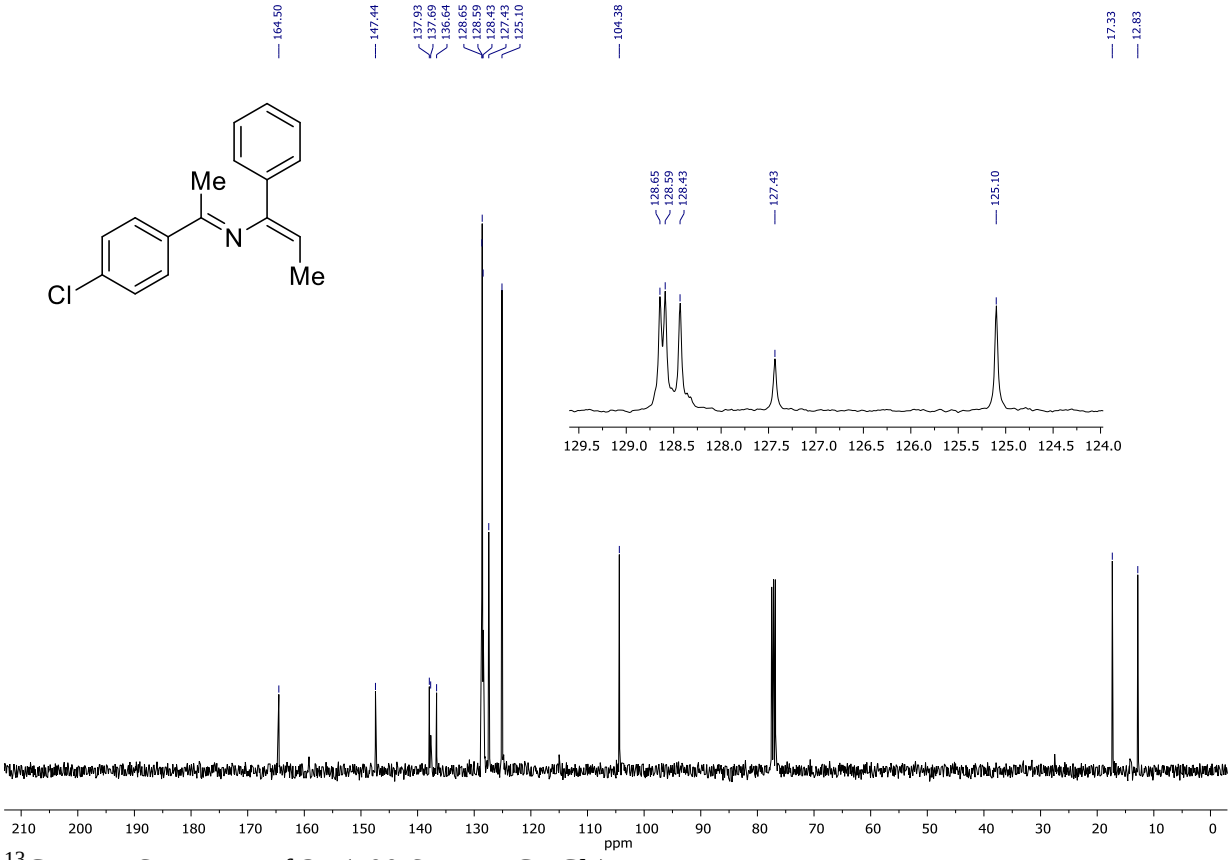
^{1}H NMR Spectrum of **2f** (400.1 MHz, $CDCl_3$)



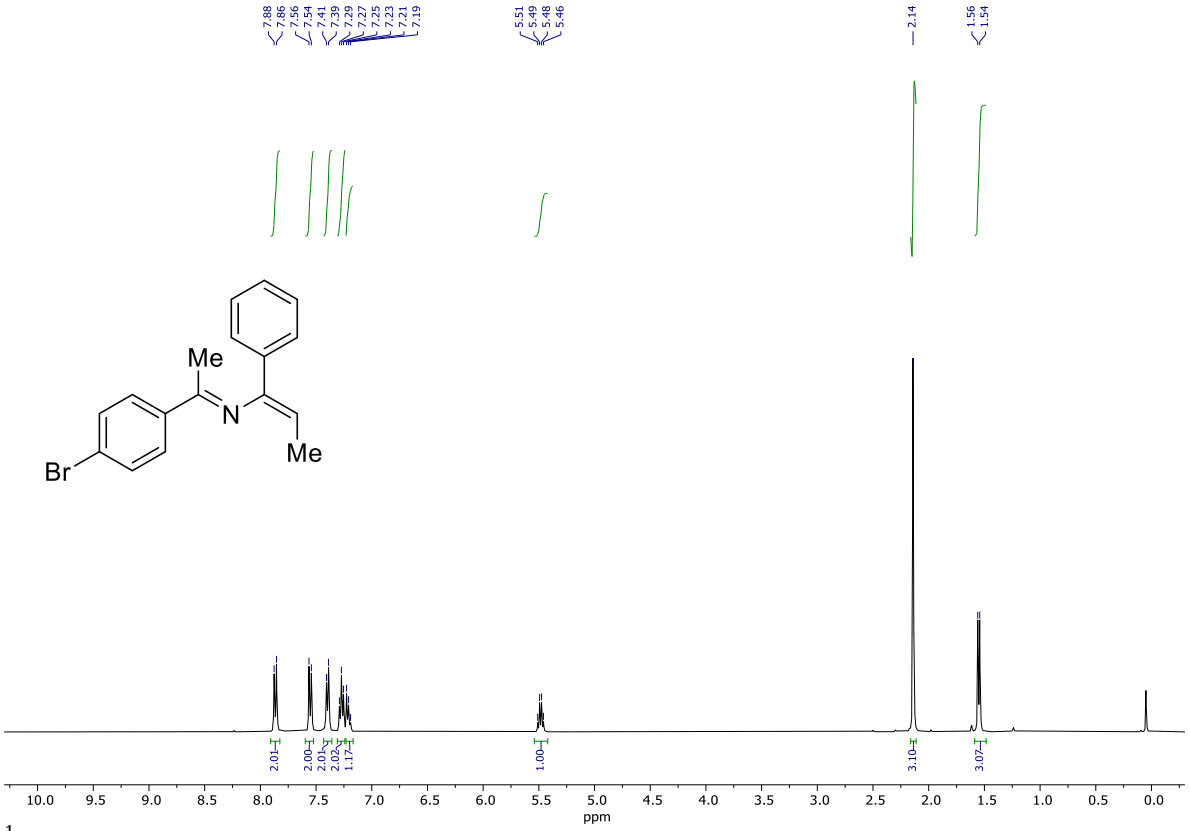
^{13}C NMR Spectrum of **2f** (100.6 MHz, $CDCl_3$)



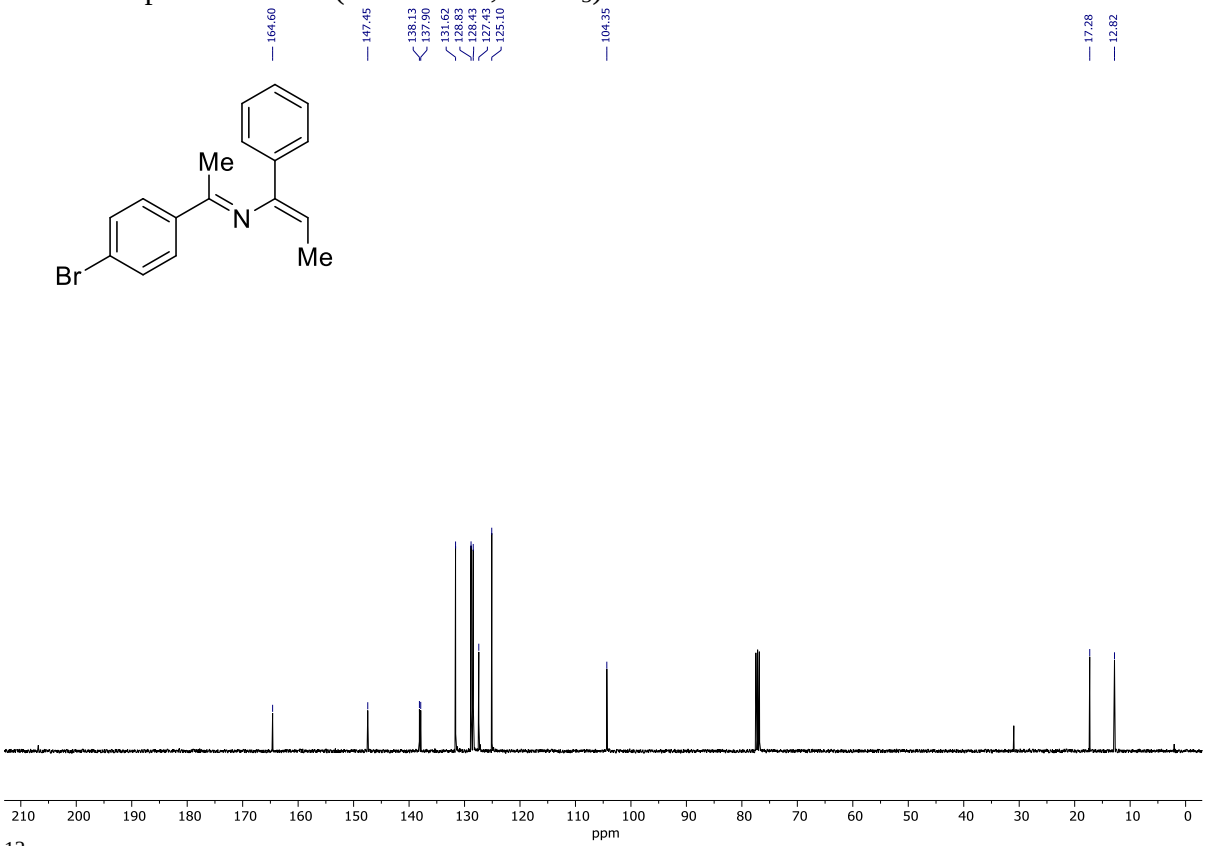
^{1}H NMR Spectrum of **2g** (400.1 MHz, $CDCl_3$)



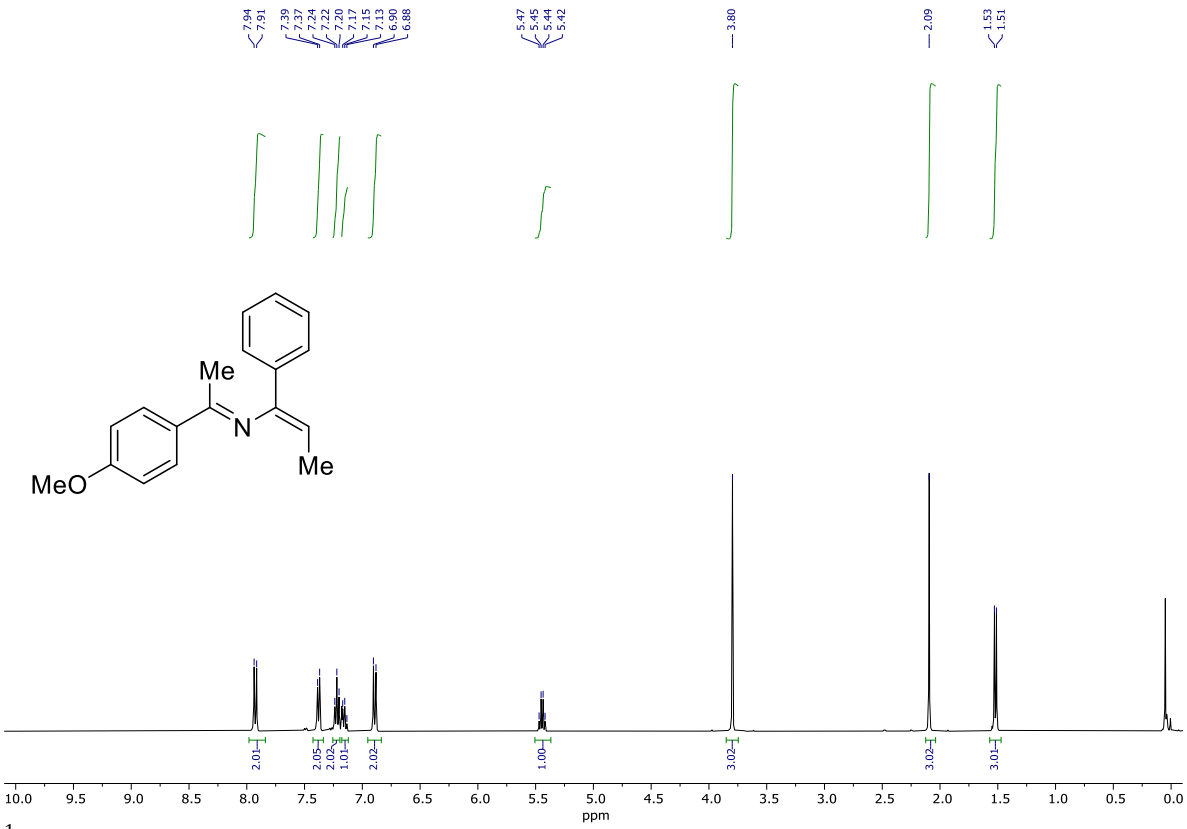
^{13}C NMR Spectrum of **2g** (100.6 MHz, $CDCl_3$)



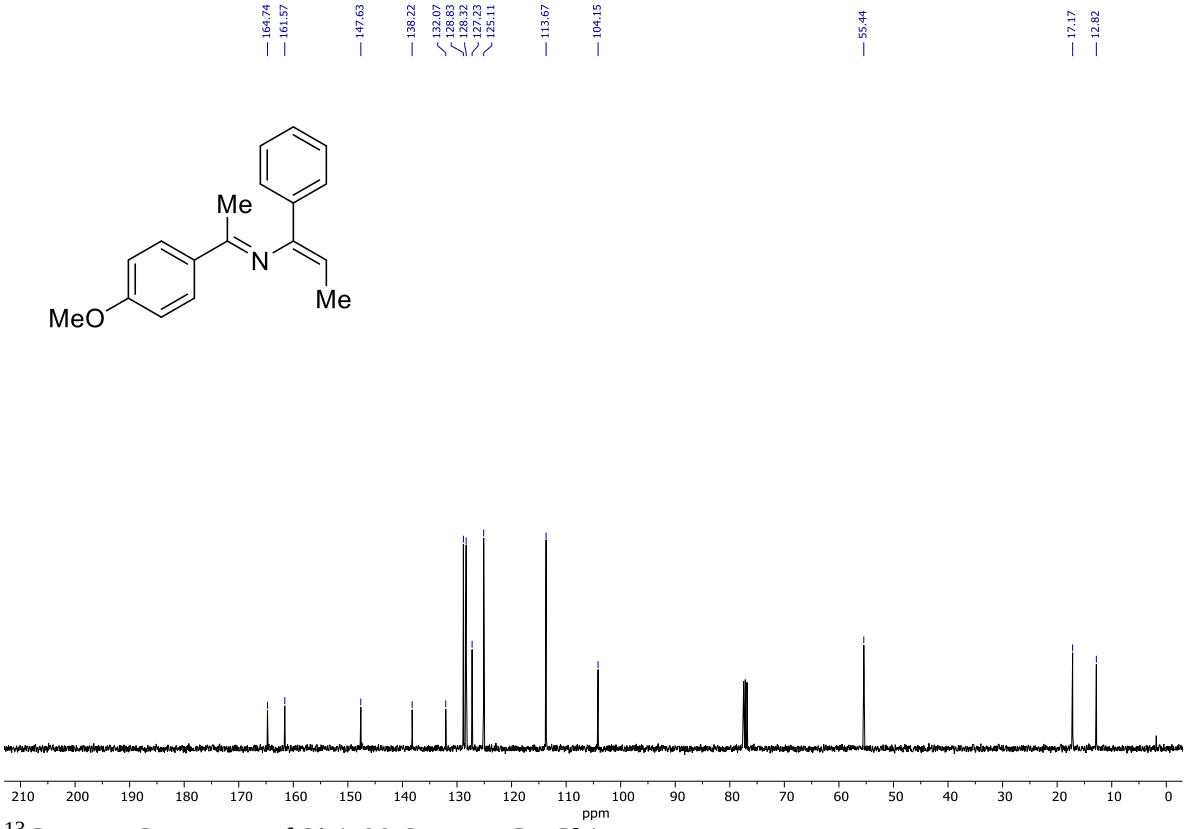
^{1}H NMR Spectrum of **2h** (400.1 MHz, $CDCl_3$)



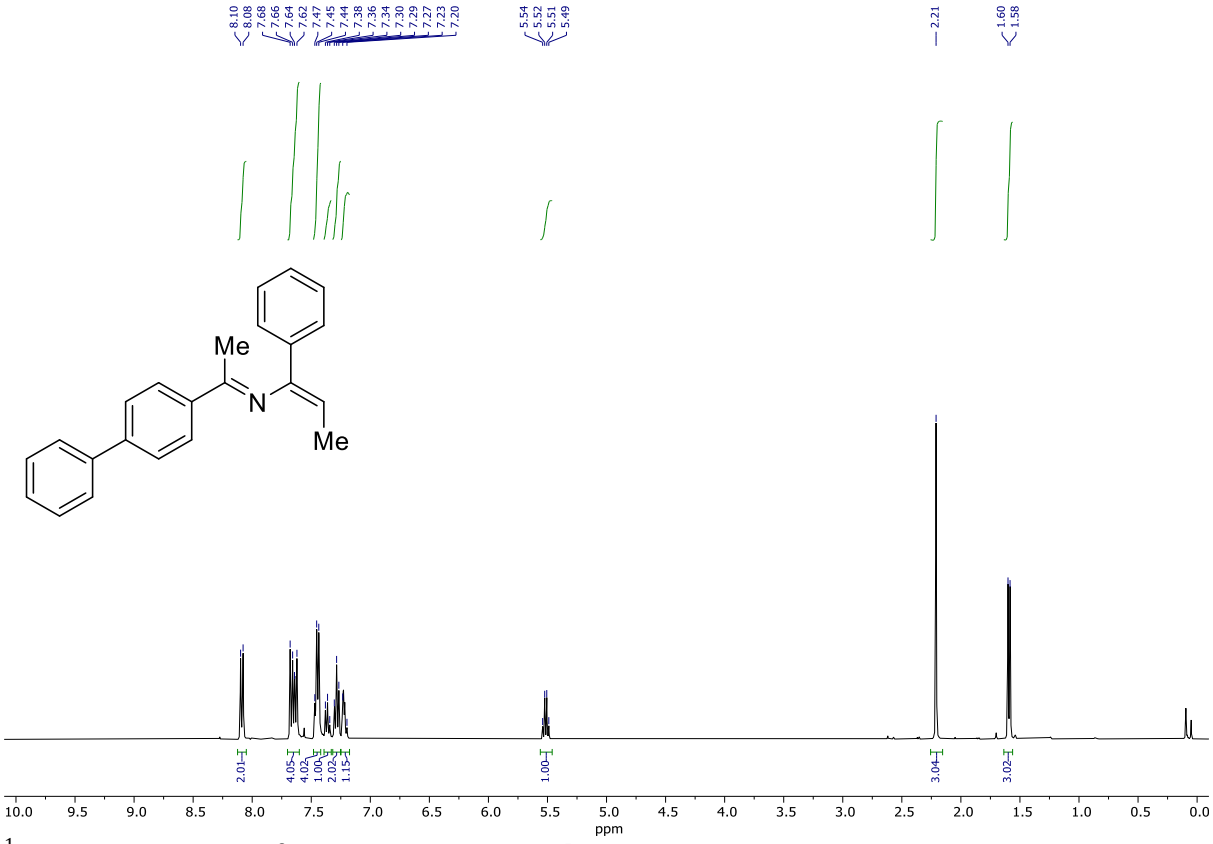
^{13}C NMR Spectrum of **2h** (100.6 MHz, $CDCl_3$)



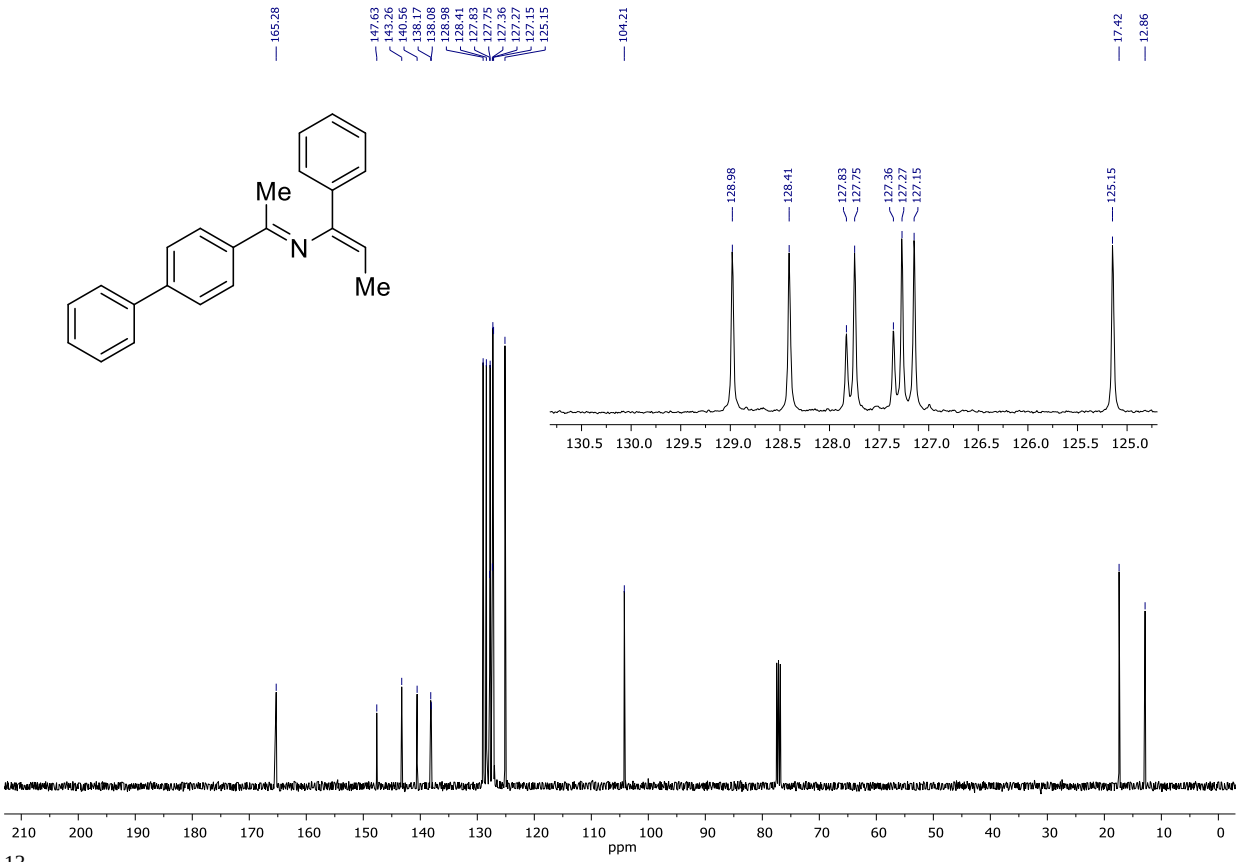
^{1}H NMR Spectrum of **2i** (400.1 MHz, $CDCl_3$)



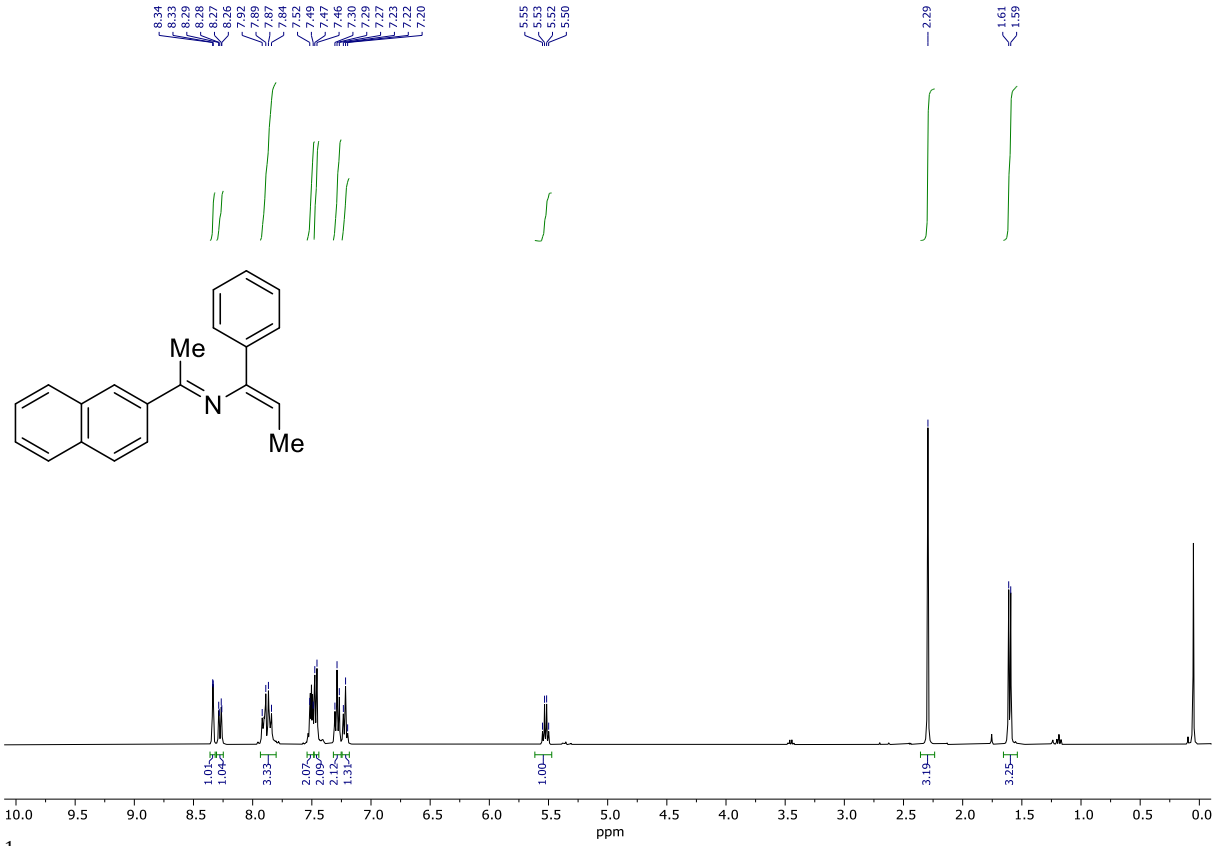
^{13}C NMR Spectrum of **2i** (100.6 MHz, $CDCl_3$)



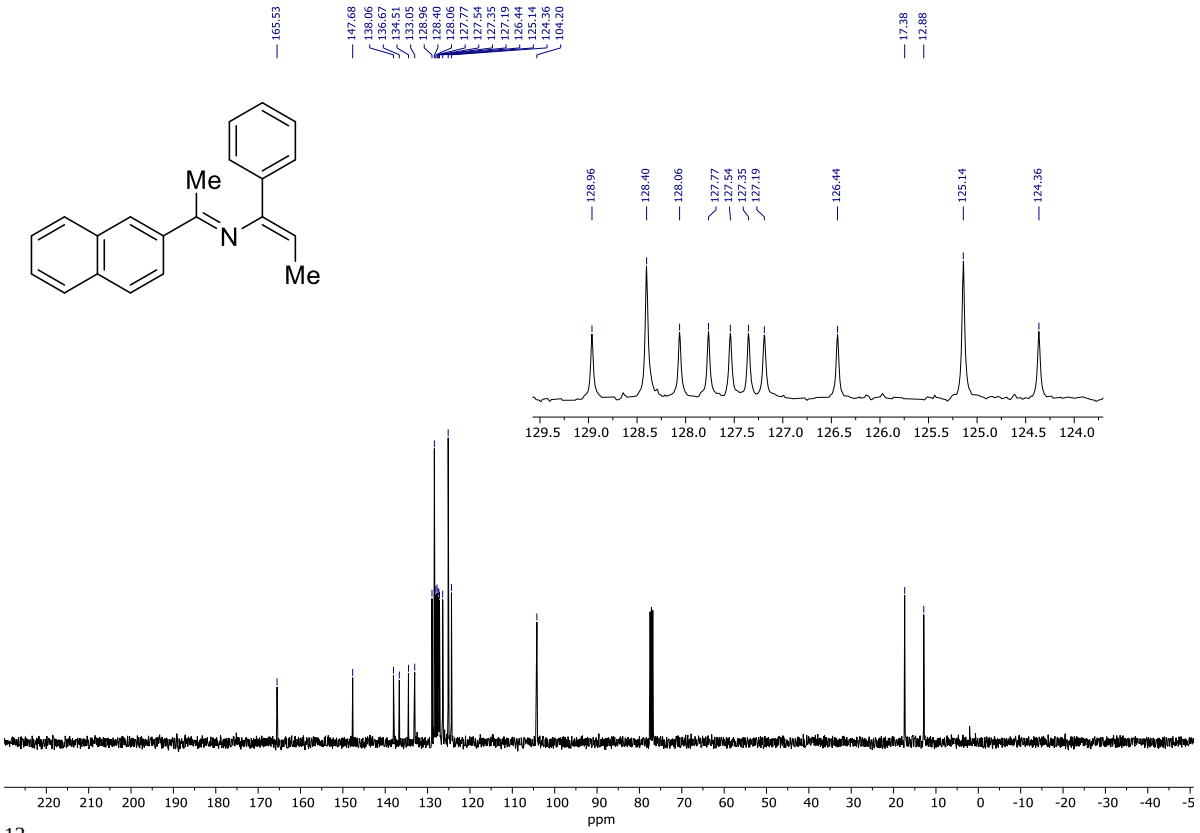
^{1}H NMR Spectrum of **2j** (400.1 MHz, $CDCl_3$)



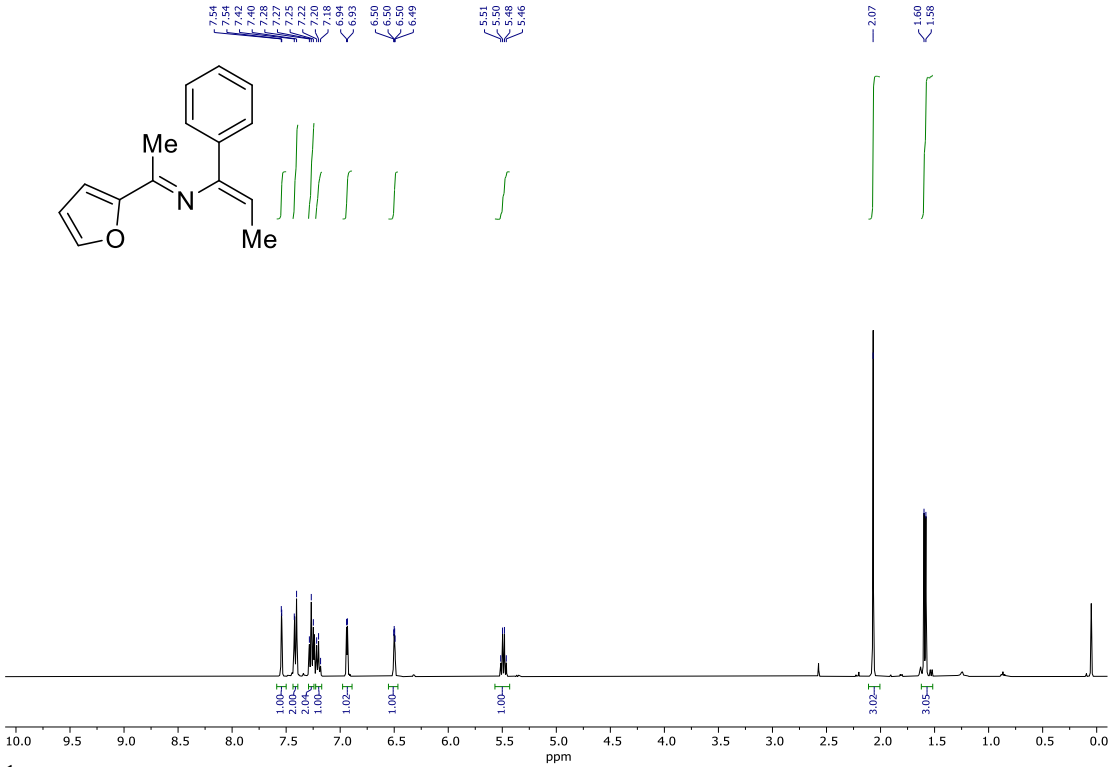
^{13}C NMR Spectrum of **2j** (100.6 MHz, $CDCl_3$)



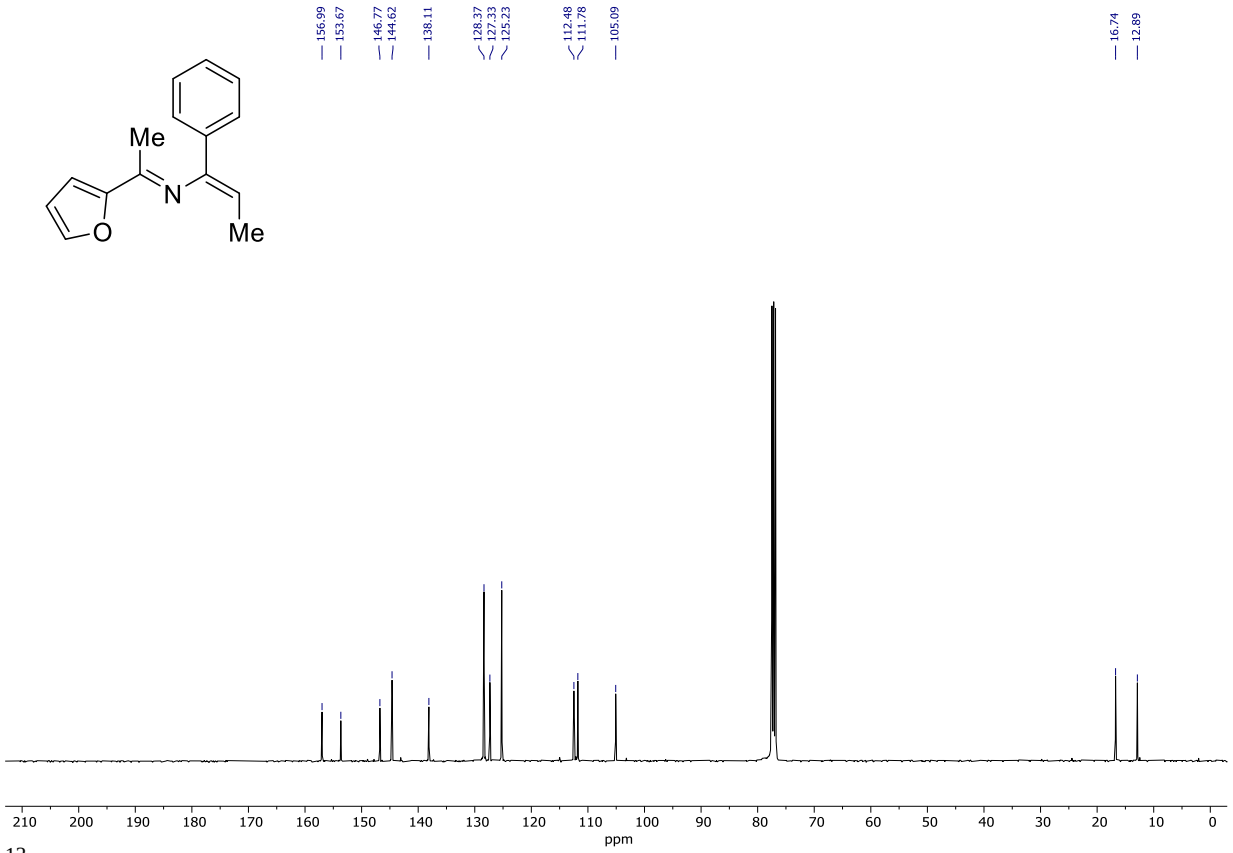
^{1}H NMR Spectrum of **2k** (400.1 MHz, $CDCl_3$)



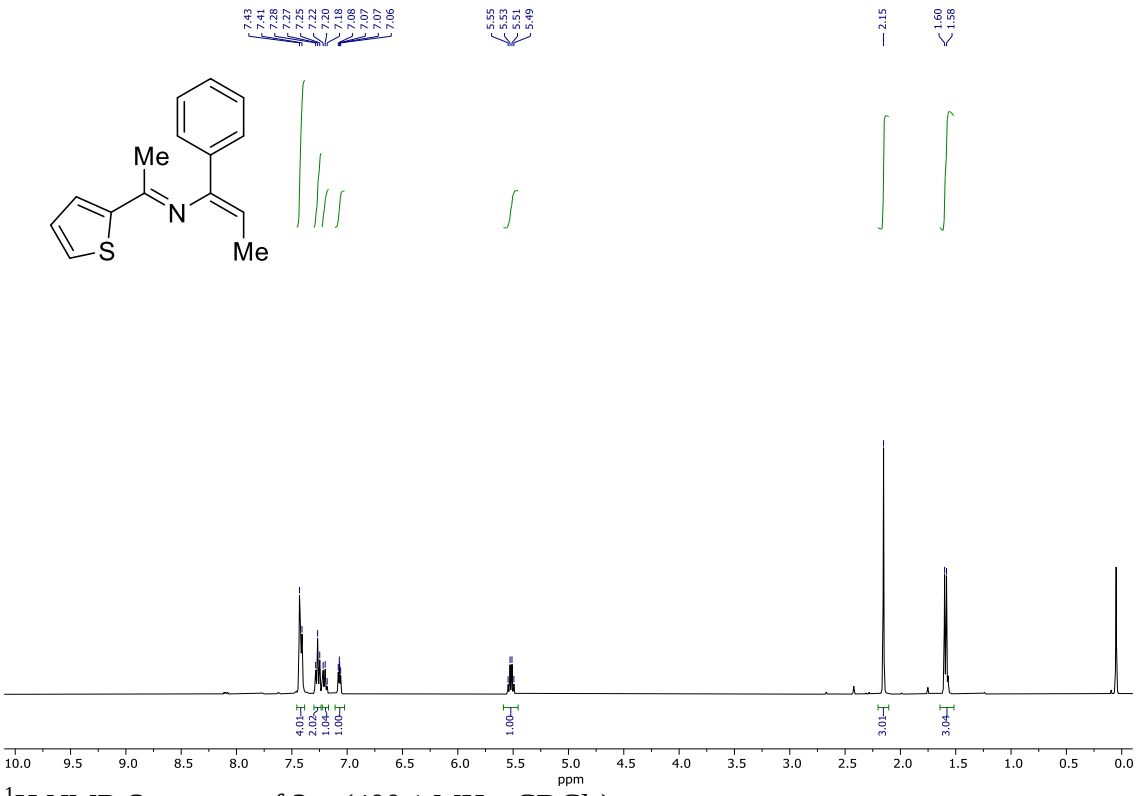
^{13}C NMR Spectrum of **2k** (100.6 MHz, $CDCl_3$)



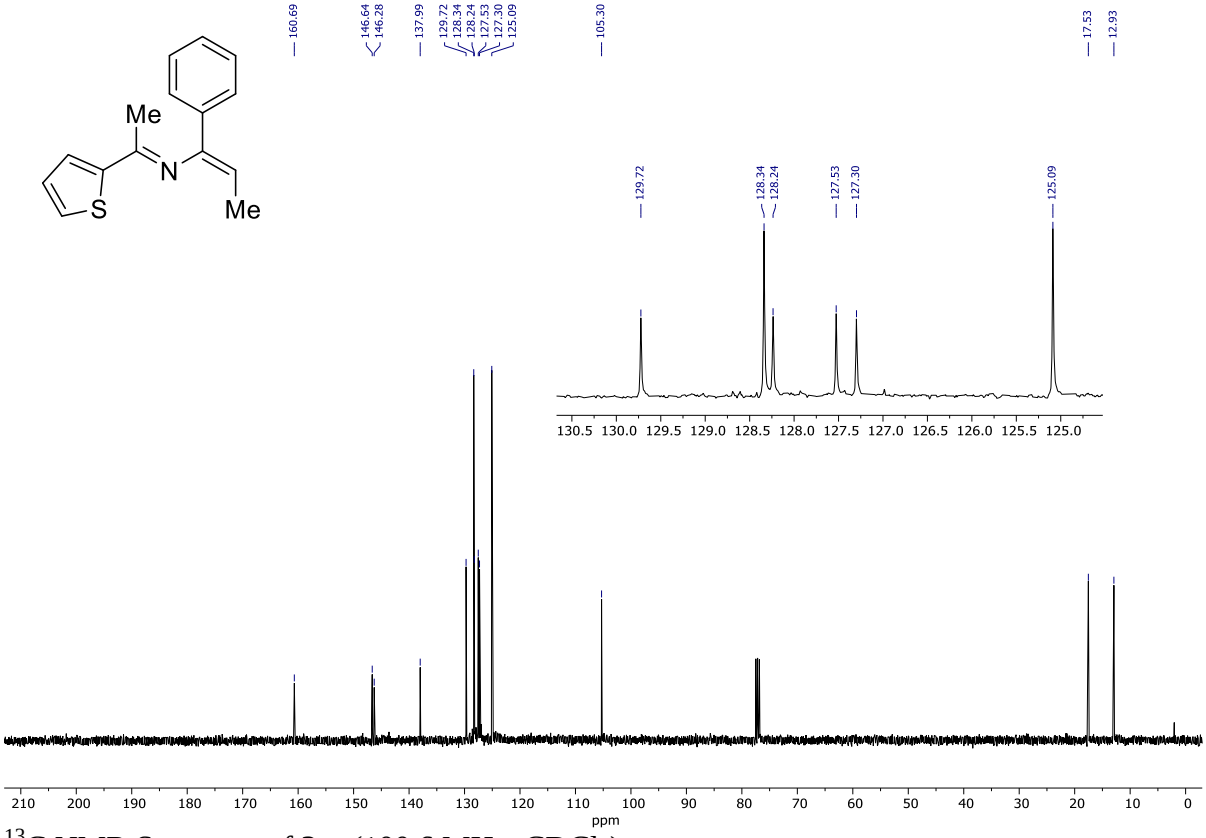
^{1}H NMR Spectrum of **2l** (400.1 MHz, $CDCl_3$)



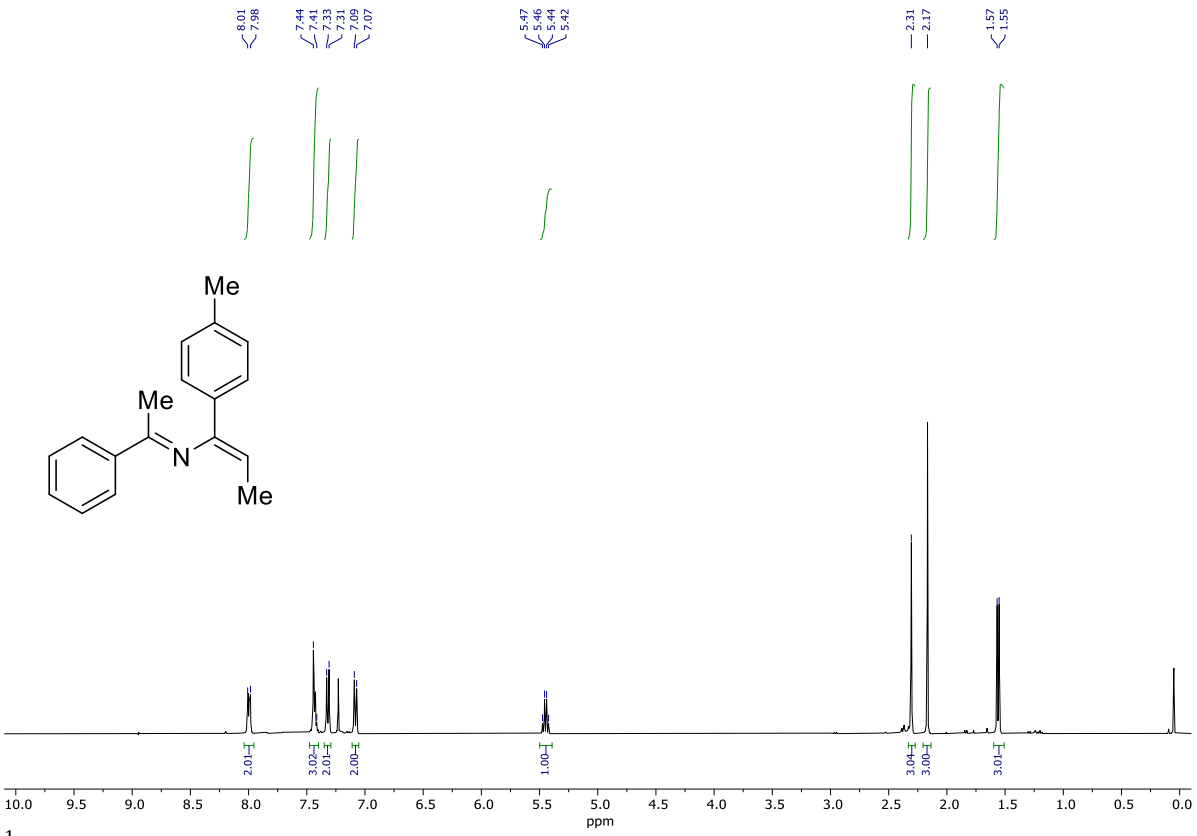
^{13}C NMR Spectrum of **2l** (100.6 MHz, $CDCl_3$)



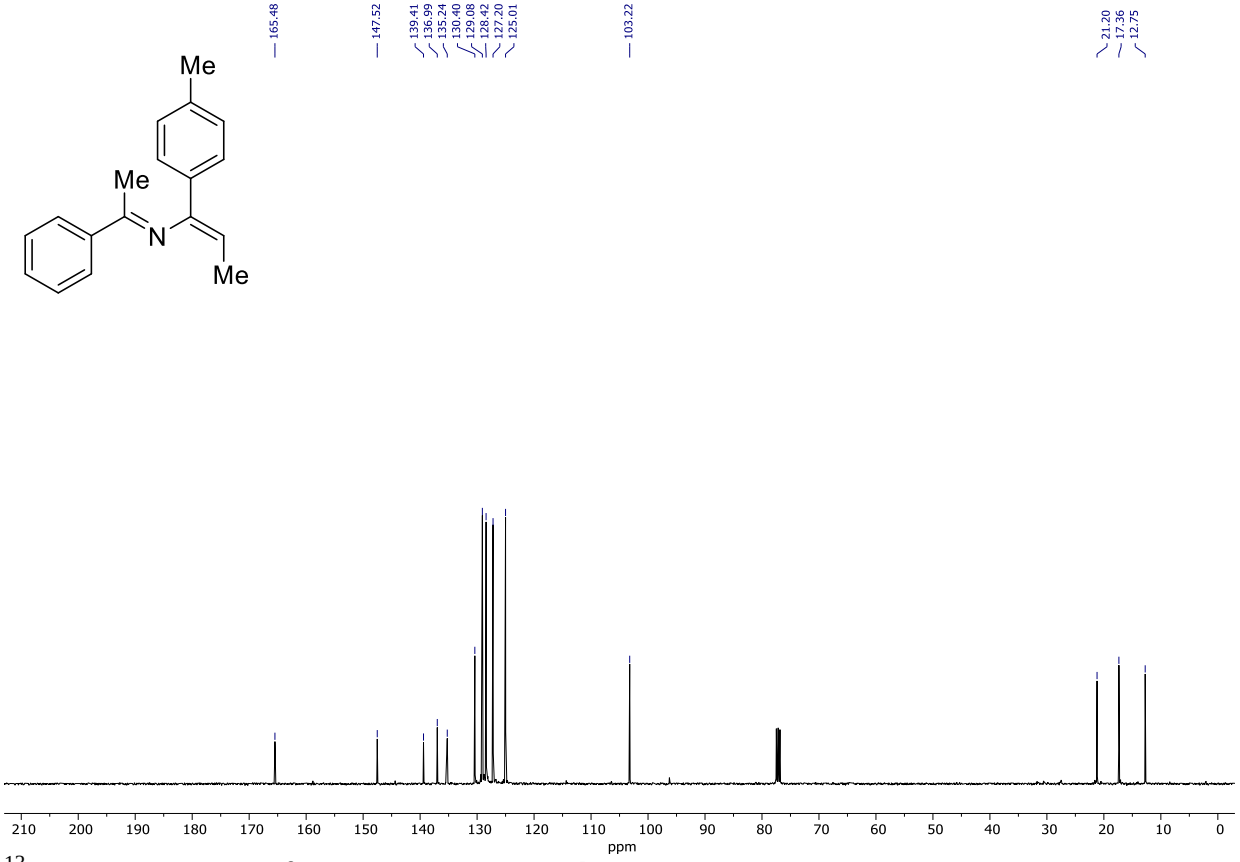
^{1}H NMR Spectrum of **2m** (400.1 MHz, $CDCl_3$)



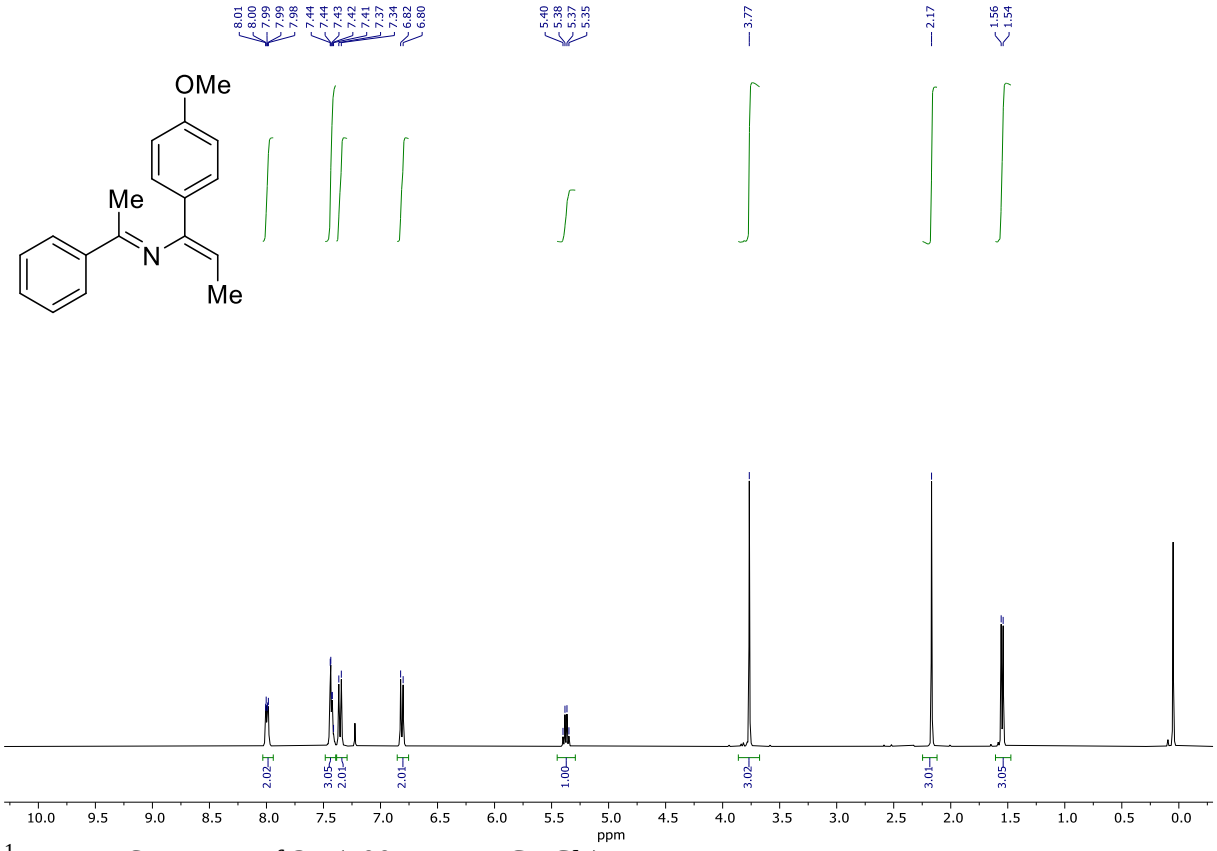
^{13}C NMR Spectrum of **2m** (100.6 MHz, $CDCl_3$)



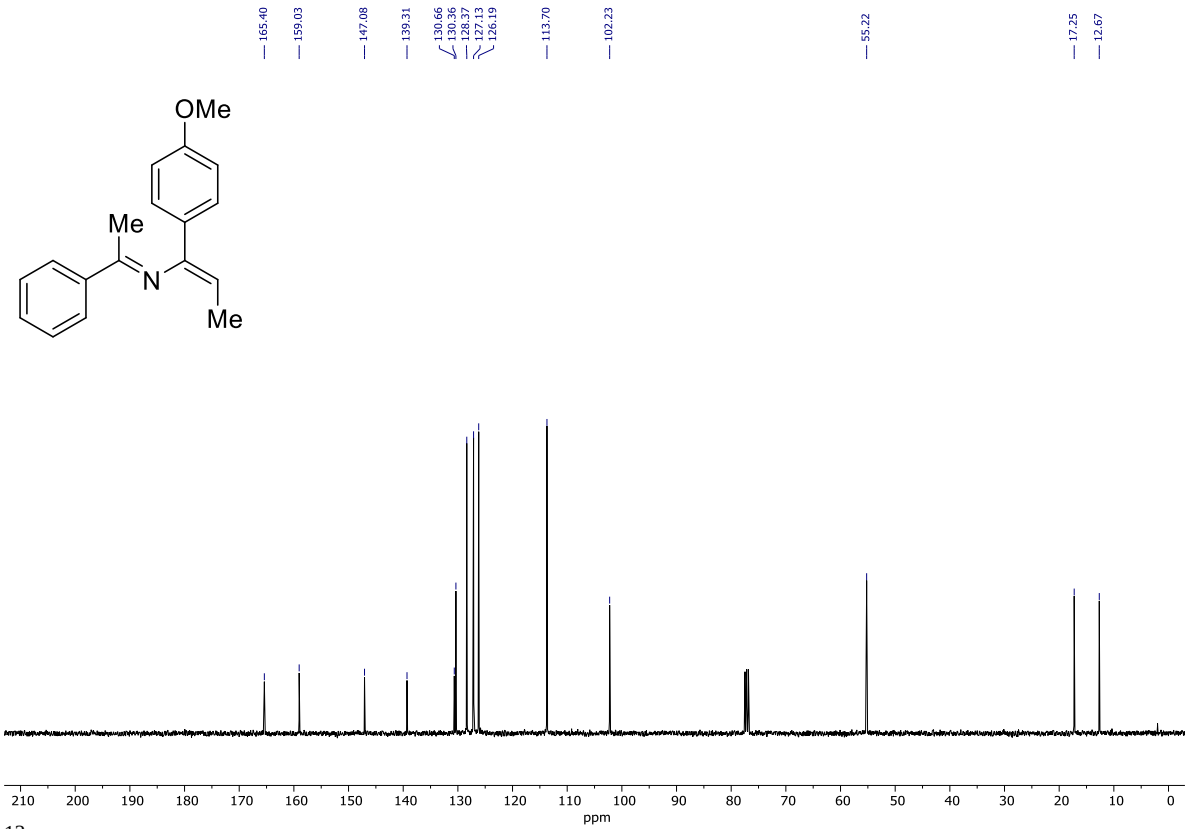
^{1}H NMR Spectrum of **2o** (400.1 MHz, $CDCl_3$)



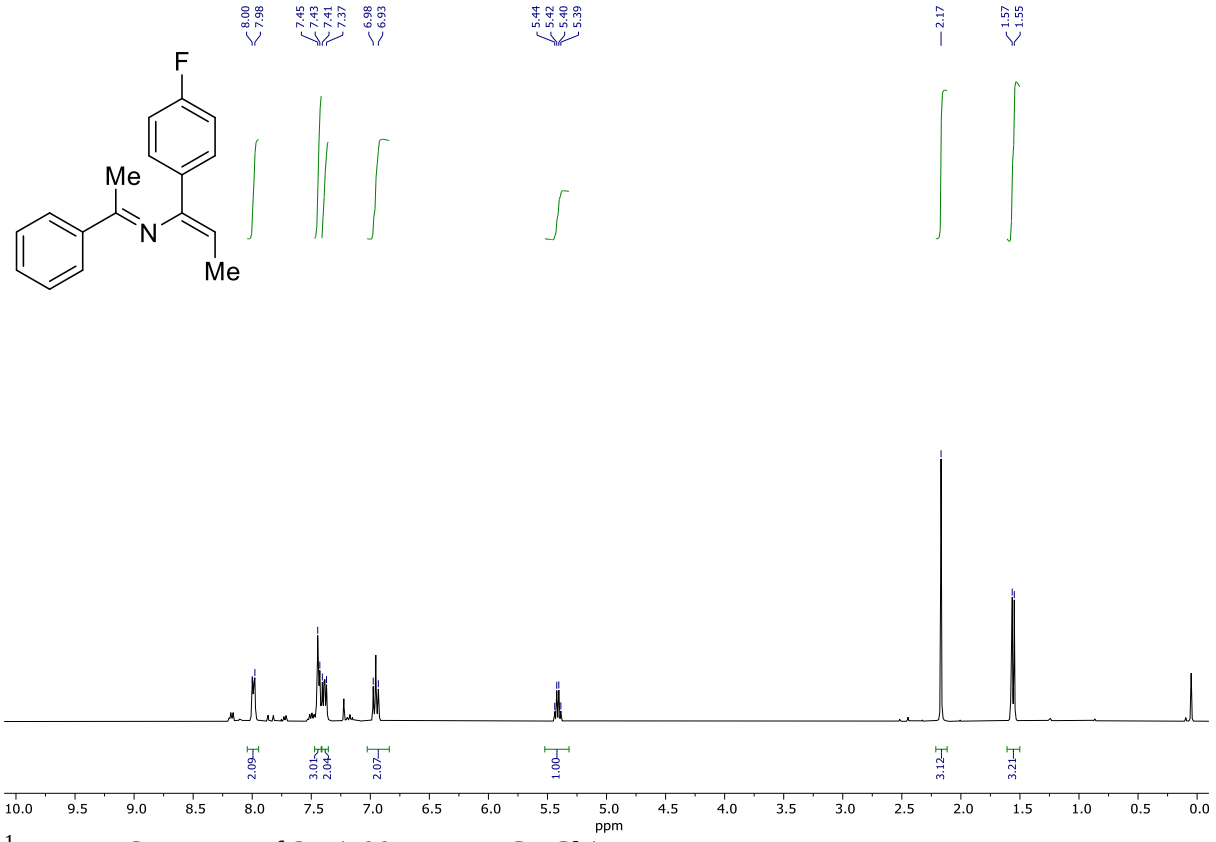
^{13}C NMR Spectrum of **2o** (100.6 MHz, $CDCl_3$)



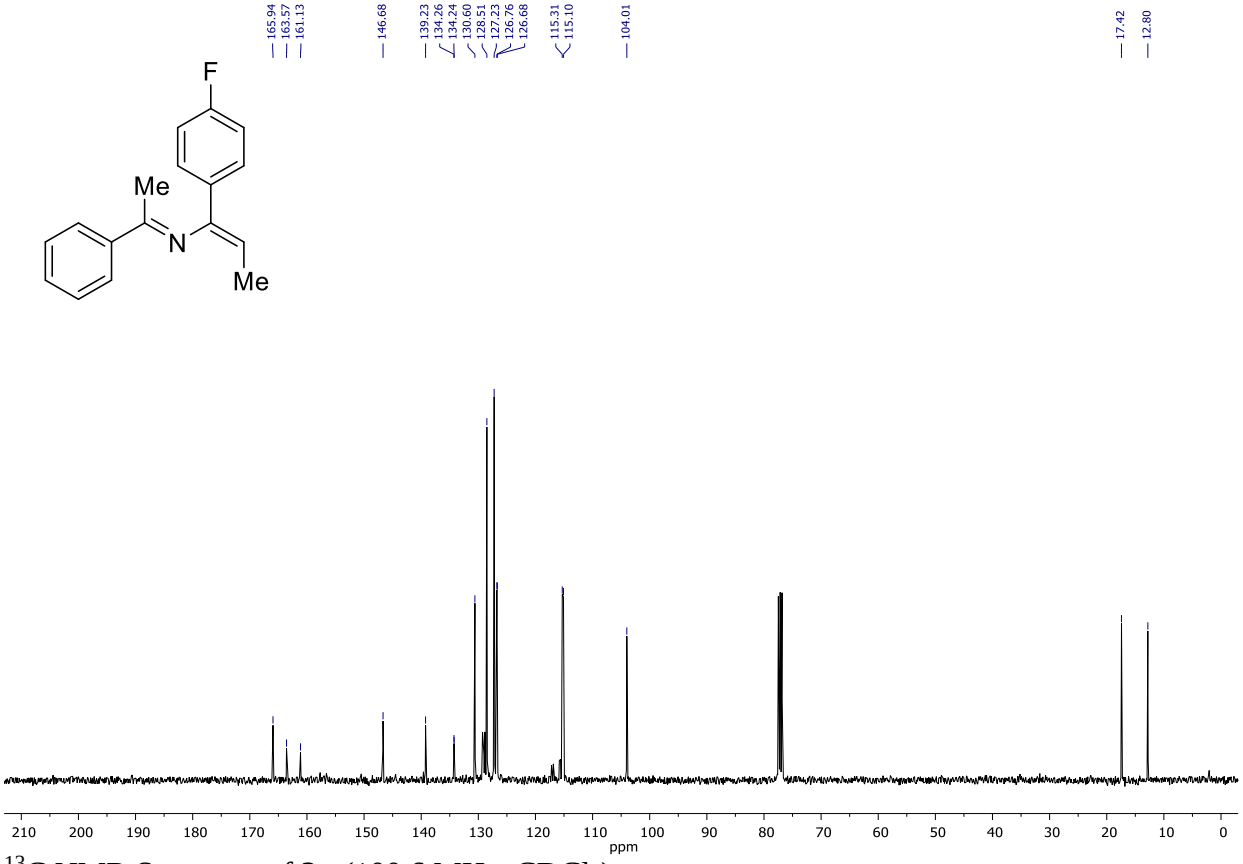
^{1}H NMR Spectrum of **2p** (400.1 MHz, $CDCl_3$)



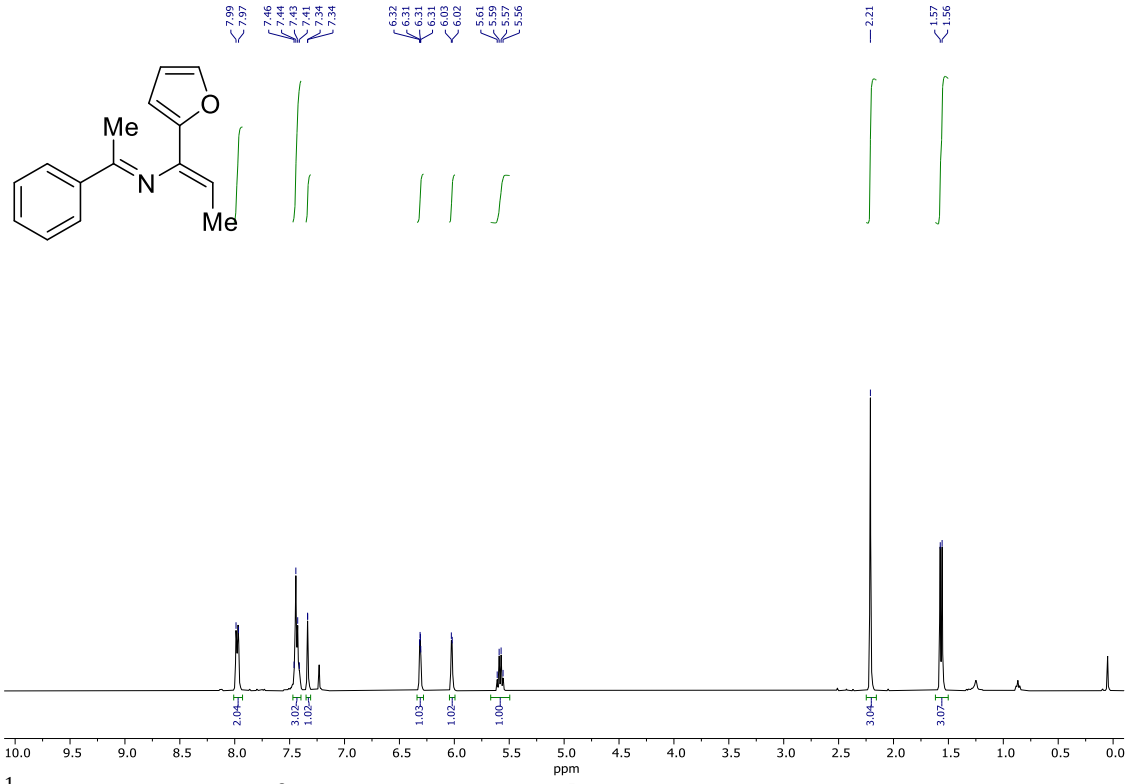
^{13}C NMR Spectrum of **2p** (100.6 MHz, $CDCl_3$)



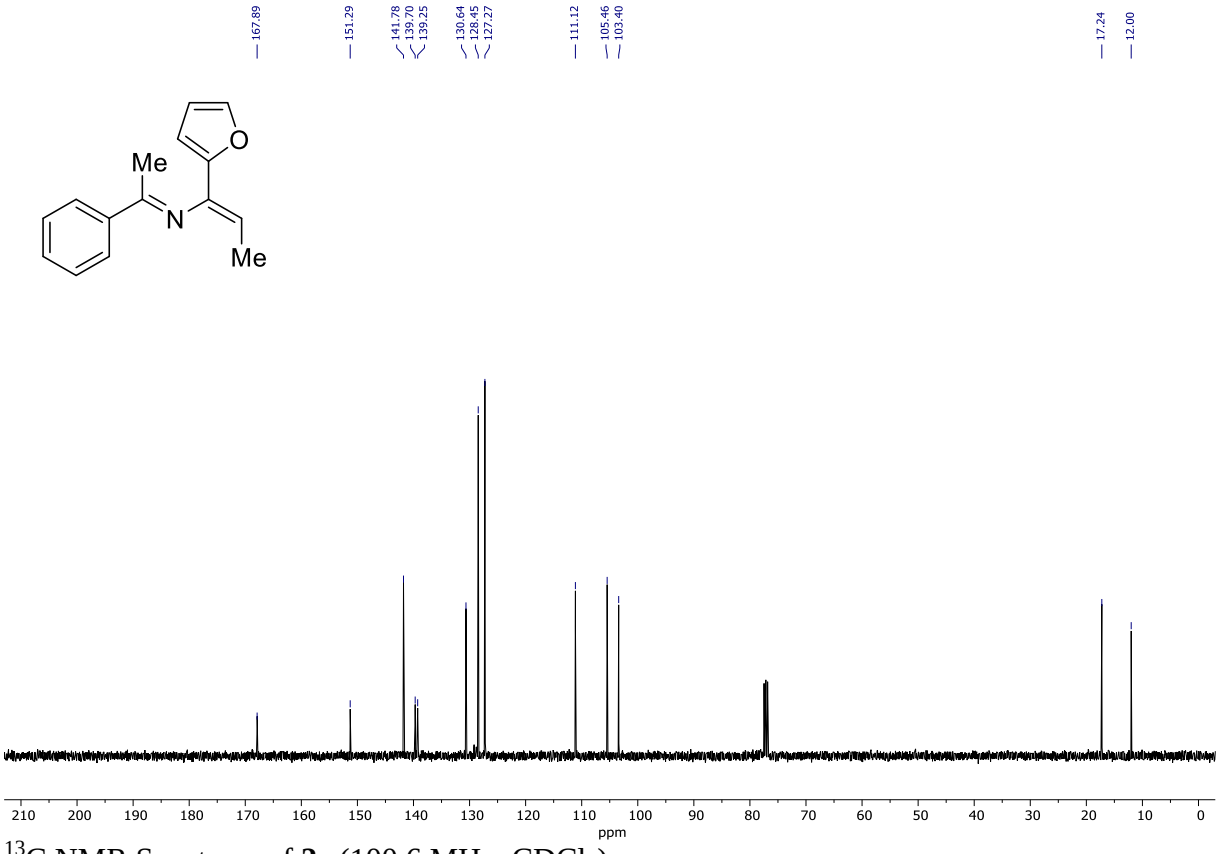
^{1}H NMR Spectrum of **2q** (400.1 MHz, $CDCl_3$)



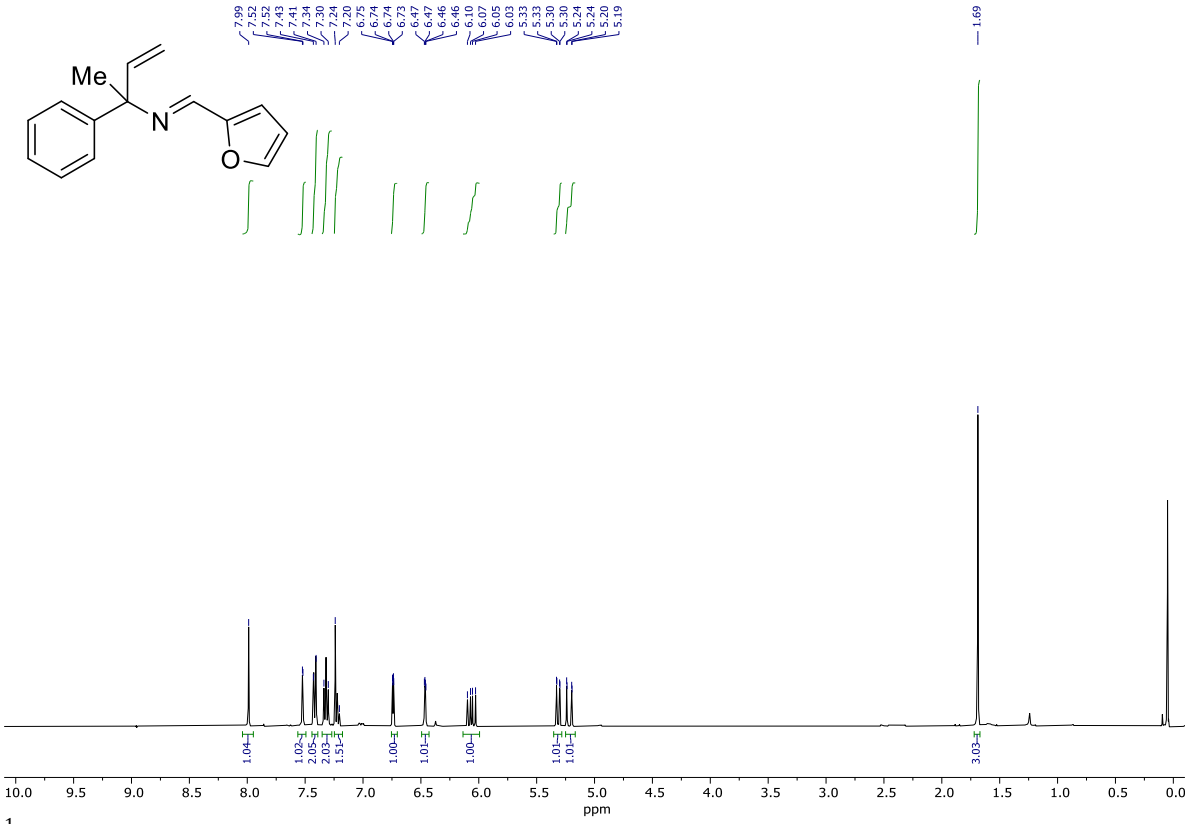
^{13}C NMR Spectrum of **2q** (100.6 MHz, $CDCl_3$)



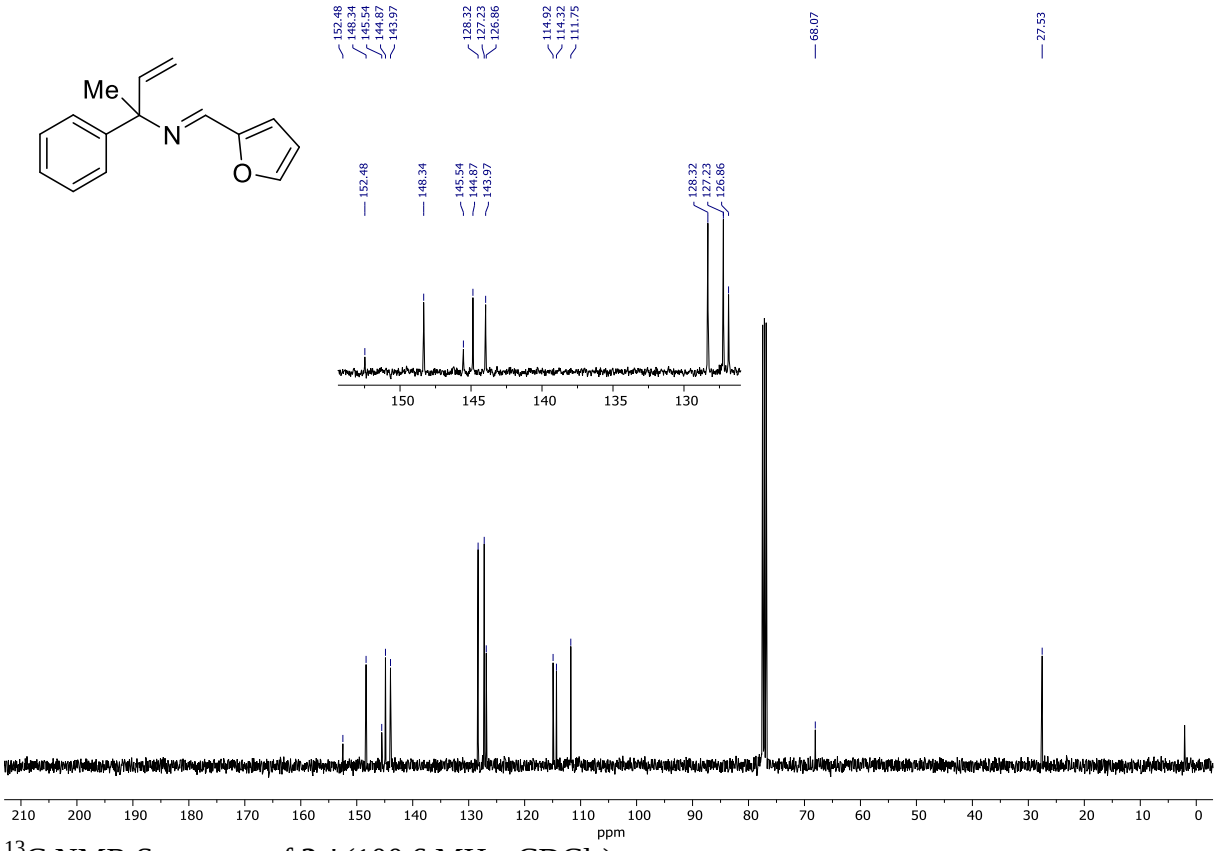
^{1}H NMR Spectrum of **2r** (400.1 MHz, $CDCl_3$)



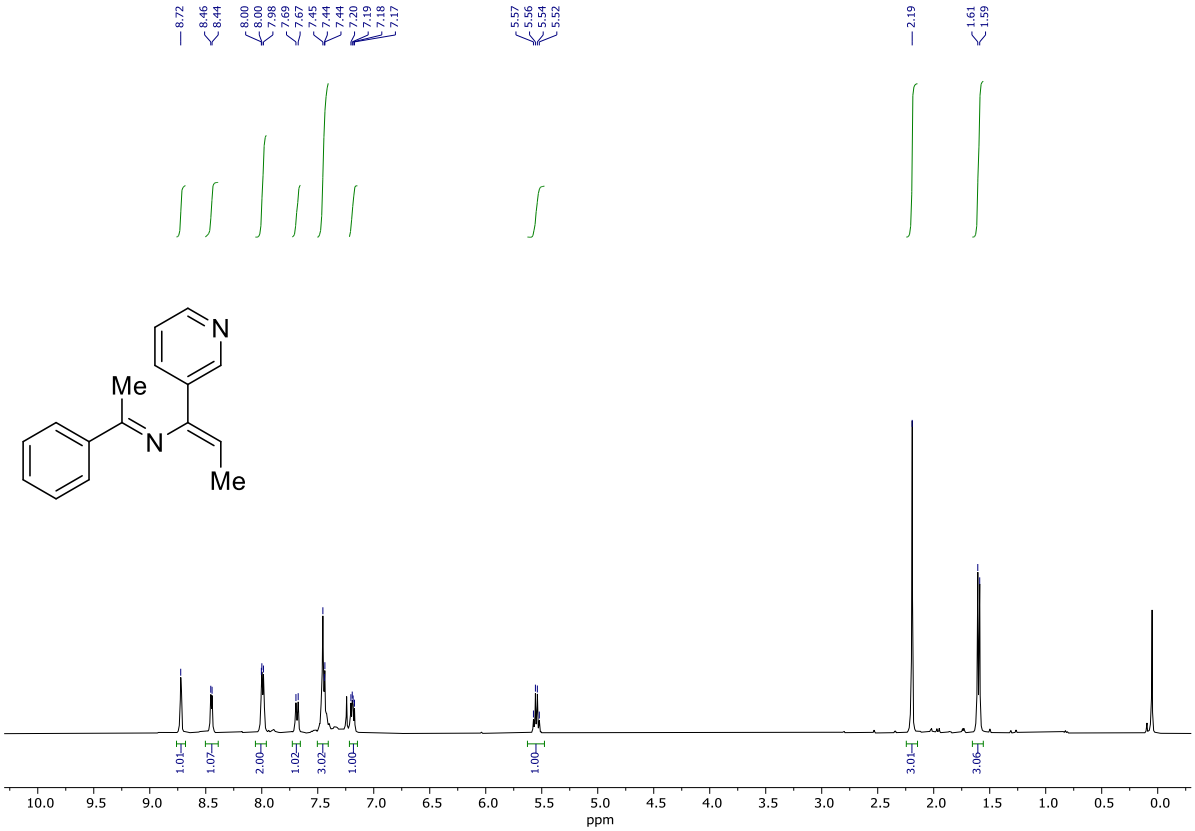
^{13}C NMR Spectrum of **2r** (100.6 MHz, $CDCl_3$)



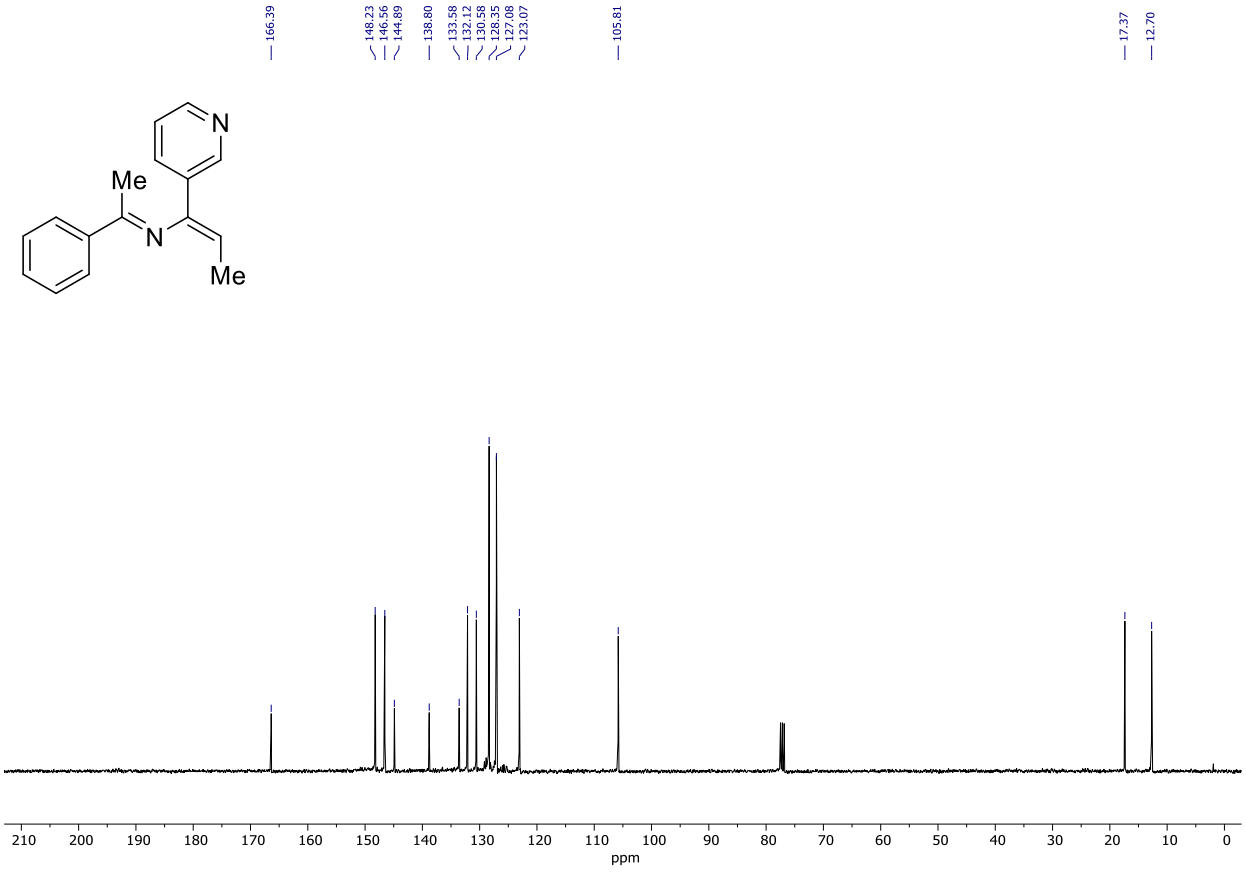
^{1}H NMR Spectrum of **2r'** (400.1 MHz, $CDCl_3$)



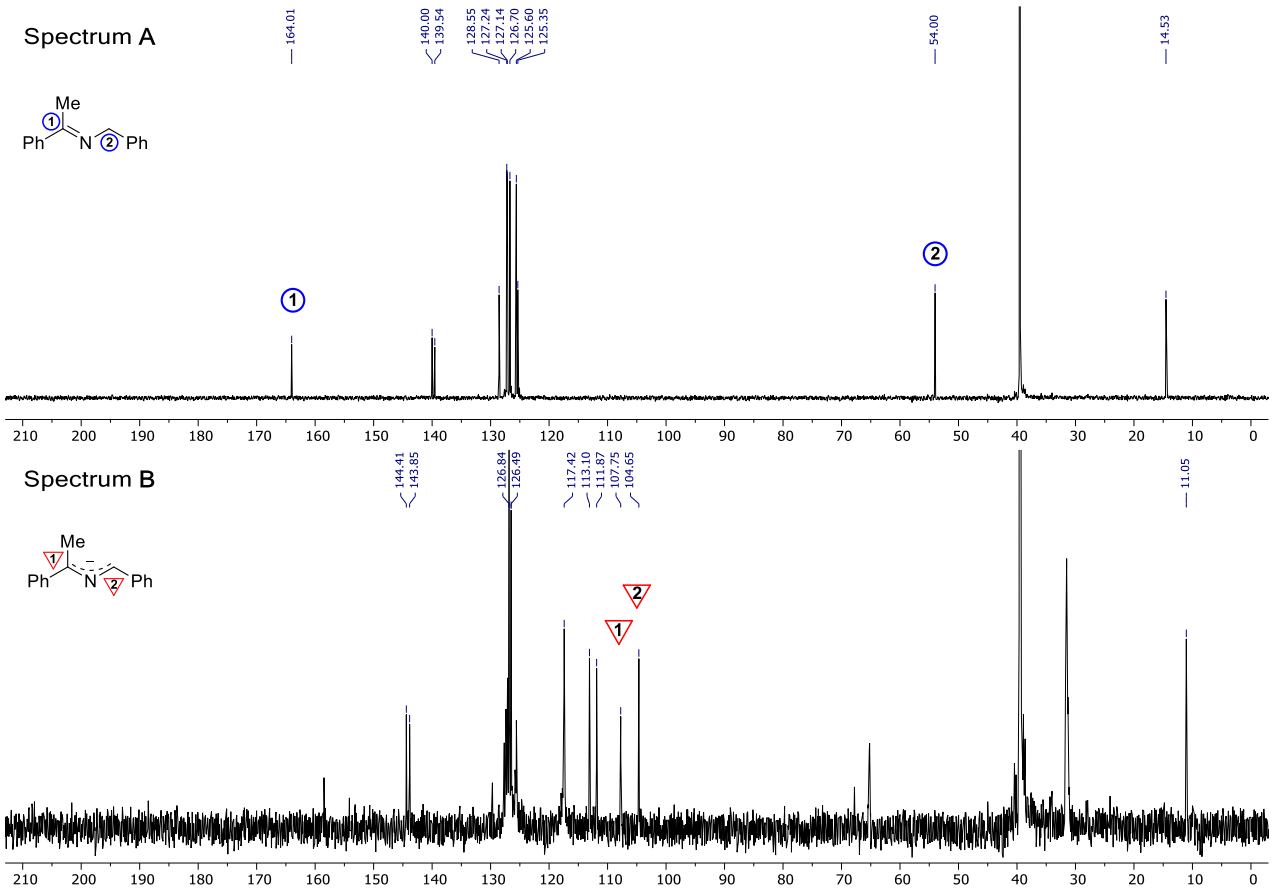
^{13}C NMR Spectrum of **2r'** (100.6 MHz, $CDCl_3$)



^{1}H NMR Spectrum of **2s** (400.1 MHz, $CDCl_3$)



^{13}C NMR Spectrum of **2s** (100.6 MHz, $CDCl_3$)



^{13}C NMR Spectra (100.6 MHz, DMSO). Spectrum **A**: imine **1a** in DMSO; Spectrum **B**: imine **1a** in the KOBu^{t}/DMSO system