

Supporting Information

Copper(I) catalyzed diastereoselective multicomponent synthesis of spiroindolo-pyrrolidines/-imidazolidines/-triazolidines from diazoamides via azomethine ylides

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Characterization of compounds **5e-i**, **7f-l**, **9f-j**

Dimethyl 1-allyl-5'-(2,3-dimethoxyphenyl)-2-oxo-1'-phenylspiro[indoline-3,2'-pyrrolidine]-3',4'-dicarboxylate (5e). According to the general procedure, to a solution of aldehyde **2c** (90 mg, 0.54 mmol), amine **3a** (50 mg, 0.54 mmol), DMAD **4c** (85 mg, 0.60 mmol) and copper(I) thiophenecarboxylate (1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1d** (100 mg, 0.50 mmol). Purification furnished the corresponding spiroindolodihydropyrrole **5e** as a white solid; yield: 211 mg (76%); $R_f = 0.42$ (hexane/EtOAc, 9:1); mp 167-169 °C; IR (neat): ν_{\max} 3042, 2824, 2804, 1788, 1673, 1623, 1592, 1432, 1310, 1261, 1176, 732, 678 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 7.46-7.56 (dd, $J_1 = 1.2$ Hz, $J_2 = 7.6$ Hz, 1H, ArH), 7.3-7.34 (td, $J_1 = 0.8$ Hz, $J_2 = 6.8$ Hz, 1H, ArH), 7.17 (d, $J_1 = 1.2$ Hz, 2H, ArH), 7.10-7.06 (td, $J_1 = 0.8$ Hz, $J_2 = 6.8$ Hz, 1H, ArH), 6.98 (t, $J = 8$ Hz, 1H, ArH), 6.89-6.84 (m, 3H, ArH), 6.75-6.79 (m, 1H, ArH), 6.63 (s, 1H, ArH), 6.57 (t, $J = 7.2$ Hz, 1H, ArH), 6.36 (d, $J = 8.0$ Hz, 2H, ArH), 5.67-5.77 (m, 1H, CH), 5.23 (ddt, 2H, $J_1 = 17.2$ Hz, $J_2 = 10.4$ Hz, $J_3 = 1.2$ Hz, CH_2), 4.31 (ddt, 2H, $J_1 = 17.2$ Hz, $J_2 = 10.4$ Hz, $J_3 = 1.2$ Hz, CH_2), 3.93 (s, 3H, OCH_3), 3.81 (s, 3H, OCH_3), 3.62 (s, 3H, OCH_3), 3.44 (s, 3H, OCH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 175.28 (C=O), 165.34 (C=O), 163.30 (C=O), 154.34 (*quat-C*), 148.92 (*quat-C*), 146.64 (*quat-C*), 145.74 (*quat-C*), 145.05 (*quat-C*), 135.00 (CH), 133.53 (CH), 133.34 (CH), 132.10 (CH), 131.09 (*quat-C*), 130.91 (CH), 126.67 (CH), 126.22 (CH), 125.25 (CH), 121.64 (CH), 121.46 (CH), 120.24 (CH), 117.45 (CH), 114.34 (CH), 111.79 (CH), 66.3 (OCH_3), 62.87 (OCH_3), 57.72 (OCH_3), 54.34 (OCH_3), 54.22 (OCH_3), 45.04 (NCH_2); HRMS (ESI) Calcd for $\text{C}_{32}\text{H}_{30}\text{N}_2\text{O}_7$ [(M+H) $^+$] 555.2131, found 555.2144.

tert-Butyl 3'-(naphthalen-1-yl)-2,4',6'-trioxo-2',5'-diphenyl-3',3a',4',5',6',6a'-hexahydro-2'H-spiro[indoline-3,1'-pyrrolo[3,4-c]pyrrole]-1-carboxylate (5f). According to the general procedure, to a solution of aldehyde **2d** (65 mg, 0.42 mmol), amine **3a** (40 mg, 0.43 mmol), *N*-phenylmaleimide **4a** (80 mg, 0.46 mmol) and copper(I) thiophenecarboxylate (0.7 mg, 1

mol %) in dichloroethane was transferred diazoamide **1e** (100 mg, 0.39 mmol). Purification furnished the corresponding spiroindolopyrrolidine **5f** as a white solid; yield: 196 mg (79%). $R_f = 0.44$ (hexane/EtOAc, 9:1); mp 167-169 °C; IR (neat): 3059, 2932, 2864, 1778, 1721, 1653, 1587, 1492, 1468, 1380, 1261, 1176, 1075, 1103, 732, 693, 667 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 7.88 (s, 1H, ArH), 7.63-7.74 (m, 6H, ArH), 7.28-7.40 (m, 9H, ArH), 6.82 (t, $J = 7.5$ Hz, 2H, ArH), 6.64-6.69 (m, 3H, ArH), 5.90 (d, $J = 6.0$ Hz, 1H, CH), 3.98 (d, $J = 10$ Hz, 1H, CH), 3.61-3.65 (dd, $J_1 = 6.0$ Hz, $J_2 = 10$ Hz, 1H, CH), 1.42 (s, 9H, *Nboc*); ^{13}C NMR (CDCl_3 , 100 MHz) δ 175.61 (C=O), 174.45 (C=O), 173.83 (C=O), 148.19 (*quat-C*), 142.28 (*quat-C*), 140.53 (*quat-C*), 137.95 (*quat-C*), 133.37 (*quat-C*), 133.11 (CH), 131.82 (*quat-C*), 130.59 (CH), 129.08 (CH), 128.89 (CH), 128.8 (CH), 128.52 (CH), 128.09 (CH), 128.03 (CH), 127.64 (CH), 126.98 (CH), 126.5 (CH), 126.17 (CH), 126.0 (CH), 125.32 (CH), 124.38 (CH), 124.14 (CH), 123.36 (CH), 115.98 (CH), 84.38 (*quat-C*), 74.74 (*quat-C*), 66.0 (CH), 53.88 (CH), 52.04 (CH), 27.93 (*Nboc*); HRMS (ESI) Calcd for $\text{C}_{40}\text{H}_{33}\text{N}_3\text{O}_5$ $[(\text{M}+\text{Na})^+]$ 658.2318, found 658.2320.

1-Methyl-3'-(4-nitrophenyl)-2',5'-diphenyl-3,3a'-dihydro-2'H-spiro[indoline-3,1'-pyrrolo[3,4-c]pyrrole]-2,4',6'(5'H,6a'H)-trione (5g). According to the general procedure, to a solution of aldehyde **2e** (95 mg, 0.63 mmol), amine **3a** (60 mg, 0.64 mmol), *N*-phenylmaleimide **4a** (120 mg, 0.69 mmol) and copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1c** (100 mg, 0.58 mmol). Purification yielded the corresponding spiroindolopyrrolidine **5g** as a white solid; yield : 132 mg (42%): $R_f = 0.41$ (hexane/EtOAc, 8:2); mp 157-159 °C; IR (neat): 3038, 2918, 2799, 1758, 1728, 1652, 1622, 1398, 1250, 1075, 1025, 732 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 7.51 (d, $J = 1.6$ Hz, 2H, ArH), 7.39-7.43 (m, 2H, ArH), 7.28-7.35 (m, 3H, ArH), 7.20-7.24 (m, 4H, ArH), 7.13-7.18 (m, 1H, ArH), 6.81-6.85 (m, 2H, ArH), 6.71-6.75 (m, 1H, ArH), 6.61-6.65 (m, 3H, ArH), 5.83 (d, $J = 6.0$ Hz, 1H, CH_2), 4.0 (d, $J = 10.0$ Hz, 1H, CH), 3.60-3.64 (dd, $J_1 = 6.0$

Hz, $J_2 = 10.0$ Hz, 1H, CH), 2.85 (s, 3H, NCH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 176.20 (C=O), 175.83 (C=O), 174.78 (C=O), 144.11 (*quat-C*), 142.88 (*quat-C*), 140.84 (*quat-C*), 131.87 (*quat-C*), 130.33 (CH), 129.27 (*quat-C*), 129.15 (CH), 128.80 (CH), 128.77 (CH), 128.32 (CH), 127.73 (CH), 127.01 (CH), 126.94 (CH), 124.15 (CH), 123.97 (CH), 123.51 (CH), 123.23 (CH), 109.04 (CH), 74.51 (*quat-C*), 65.64 (CH), 53.09 (CH), 52.29 (CH), 29.71 (CH), 25.83 (CH₃). HRMS (ESI) Calculated for C₃₂H₂₄N₄O₅ (M+H)⁺: 545.1825 found: 545.1817.

3'-(4-Bromophenyl)-1-ethyl-2',5'-diphenyl-3',3a'-dihydro-2'H-spiro[indoline-3,1'-pyrrolo[3,4-c]pyrrole]-2,4',6'(5'H,6a'H)-trione (5h). According to the general procedure, to a solution of aldehyde **2f** (110 mg, 0.59 mmol), amine **3a** (55 mg, 0.59 mmol), *N*-phenylmaleimide **4a** (110 mg, 0.64 mmol) and copper(I) thiophenecarboxylate (1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1b** (100 mg, 0.53 mmol). Purification furnished the corresponding spiroindolopyrrolidine **5h** as a white solid; yield : 229 mg (73%): $R_f = 0.42$ (hexane/EtOAc, 7:3); mp 167-169 °C; IR (neat): 3152, 2959, 2912, 2895, 1698, 1653, 1602, 1598, 1398, 1176, 1105, 732, 686 cm⁻¹. ¹H NMR (CDCl₃, 400 MHz) δ 8.13 (d, $J = 7.2$ Hz, 1H, ArH), 7.73 (d, $J = 7.6$ Hz, 2H, ArH), 7.59 (t, $J = 8$ Hz, 1H, ArH), 7.51 (d, $J = 7.6$ Hz, 1H, ArH), 7.33-7.46 (m, 3H, ArH), 7.22 (t, $J = 10$ Hz, 1H, ArH), 7.04 (d, $J = 8.4$ Hz, 2H, ArH), 6.96 (t, $J = 8.4$ Hz, 2H, ArH), 6.66-6.74 (m, 3H, ArH), 6.42 (s, 1H, ArH), 6.14 (d, $J = 6$ Hz, 2H, CH₂), 3.98 (d, $J = 10$ Hz, 1H, CH), 3.58-3.62 (dd, $J_1 = 6$ Hz, $J_2 = 10$ Hz, 1H, CH), 3.50-3.59 (m, 1H, NCH₂), 3.25-3.16 (m, 1H, NCH₂), 0.61 (t, $J = 7.2$ Hz, 3H, CH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 176.20 (C=O), 175.70 (C=O), 174.64 (C=O), 160.38 (*quat-C*), 148.73 (*quat-C*), 143.70 (*quat-C*), 142.02 (*quat-C*), 132.47 (*quat-C*), 131.72 (CH), 130.56 (*quat-C*), 130.41 (CH), 129.27 (CH), 129.03 (CH), 128.42 (CH), 127.0 (CH), 125.00 (CH), 123.75 (CH), 123.65 (CH), 119.14 (CH), 114.40 (CH), 114.32 (CH), 108.94 (CH), 93.21

(*quat-C*), 79.10 (*quat-C*), 74.56 (CH), 53.46 (CH), 52.67 (CH), 34.25 (CH₂), 12.04 (CH₃).

HRMS (ESI) Calcd for C₃₂H₂₆BrN₃O₃ [(M+Na)⁺] 614.1055, found 614.1049.

tert-Butyl 2'-methyl-2,4',6'-trioxo-3',5'-diphenyl-3',3a',4',5',6',6a'-hexahydro-2'H-spiro[indoline-3,1'-pyrrolo[3,4-c]pyrrole]-1-carboxylate (**5i**). According to the general procedure, to a solution of aldehyde **2a** (45 mg, 0.42 mmol), amine **3b** (30 mg, 0.45 mmol), *N*-phenylmaleimide **4a** (80 mg, 0.46 mmol) and copper(I) thiophenecarboxylate (0.7 mg, 1 mol %) in dichloroethane was transferred diazoamide **1e** (100 mg, 0.39 mmol). Purification furnished the corresponding spiroindolopyrrolidine **5i** as a white solid; yield: 159 mg (78%); R_f = 0.48 (hexane/EtOAc, 7:3); mp 153-154 °C. IR (neat): 3062, 2923, 2854, 1780, 1713, 1613, 1264, 1166, 1025, 1003, 731, 690, 687 cm⁻¹; ¹H NMR (CDCl₃, 400 MHz) δ 7.81 (d, *J* = 8 Hz, 1H, ArH), 7.45-7.48 (m, 3H, ArH), 7.33-7.37 (m, 6H, ArH), 7.22-7.32 (m, 4H, ArH), 4.76 (d, *J* = 6.4 Hz, 1H, CH), 3.75 (d, *J* = 10 Hz, 1H, CH), 3.43-3.48 (dd, *J*₁ = 6.4 Hz, *J*₂ = 10 Hz, 1H, CH), 1.87 (s, 3H, NCH₃), 1.54 (s, 9H, *Nboc*); ¹³C NMR (CDCl₃, 100 MHz) δ 175.69 (C=O), 174.92 (C=O), 174.07 (C=O), 148.59 (*quat-C*), 140.9 (*quat-C*), 140.51 (*quat-C*), 131.89 (*quat-C*), 130.43 (CH), 129.05 (CH), 128.99 (CH), 128.75 (CH), 128.22 (CH), 127.38 (CH), 127.02 (CH), 126.72 (CH), 125.41 (CH), 123.92 (CH), 115.47 (CH), 84.7 (*quat-C*), 73.85 (CH), 68.73 (CH), 53.66 (CH), 52.63 (CH), 32.54 (CH), 28.12 (*Nboc*); HRMS (ESI) Calcd for C₃₁H₂₉N₃O₅ [(M+Na)⁺] 546.2005, found 546.2018.

1'-Allyl-2-(3,4-dimethoxyphenyl)-1,5-bis(4-nitrophenyl)-3-phenylspiro[imidazolidine-4,3'-indolin]-2'-one (**7f**). According to the general procedure, to a solution of aldehyde **2c** (90 mg, 0.54 mmol), amine **3a** (50 mg, 0.54 mmol), (*E*)-4-nitro-*N*-(4-nitrobenzylidene)aniline **6b** (160 mg, 0.6 mmol) and copper(I) thiophenecarboxylate (1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1d** (100 mg, 0.5 mmol). Purification furnished the corresponding spiroindoloimidazolidine **7f** as a white solid; yield: 281 mg (82%); R_f = 0.42 (hexane/EtOAc, 9:1). mp 167-169 °C. IR (neat): ν_{max} 3059, 2932, 2864, 1778, 1721, 1653, 1587, 1492, 1468,

1380, 1261, 1176, 1075, 1103, 732, 693, 667 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 7.16-7.43 (dd, $J_1 = 1.2$ Hz, $J_2 = 7.6$ Hz, 2H, ArH), 6.93-6.89 (td, $J_1 = 0.8$ Hz, $J_2 = 6.8$ Hz, 2H, ArH), 6.75 (d, $J_1 = 1.2$ Hz, 3H, ArH), 6.70-6.66 (td, $J_1 = 0.8$ Hz, $J_2 = 6.8$ Hz, 2H, ArH), 6.58 (t, $J = 8$ Hz, 1H, ArH), 6.44-6.49 (m, 4H, ArH), 6.37-6.39 (m, 2H, ArH), 6.23 (s, 1H, ArH), 6.17 (t, $J = 7.2$ Hz, 1H, ArH), 5.96 (d, $J = 8.0$ Hz, 3H, ArH), 5.35-5.27 (m, 1H, CH), 4.82 (ddt, 3H, $J_1 = 17.2$ Hz, $J_2 = 10.4$ Hz, $J_3 = 1.2$ Hz, CH_2), 3.88 (ddt, 2H, $J_1 = 17.2$ Hz, $J_2 = 10.4$ Hz, $J_3 = 1.2$ Hz, CH_2), 3.53 (s, 3H, OCH_3), 3.40 (s, 3H, OCH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 175.28 (C=O), 165.34 (C=O), 163.30 (C=O), 154.34 (quat-C), 150.50 (quat-C), 149.69 (quat-C), 149.01 (quat-C), 148.92 (quat-C), 146.64 (quat-C), 145.74 (quat-C), 145.05 (quat-C), 135.00 (CH), 133.53 (CH), 133.34 (CH), 132.10 (CH), 131.09 (quat-C), 130.91 (CH), 126.67 (CH), 126.22 (CH), 125.25 (CH), 121.64 (CH), 121.46 (CH), 120.24 (CH), 117.45 (CH), 114.34 (CH), 111.79 (CH), 66.3 (CH), 62.87 (CH), 57.72 (CH), 54.34 (OCH_3), 54.22 (OCH_3), 45.04 (NCH_2); HRMS (ESI) Calcd for $\text{C}_{39}\text{H}_{33}\text{N}_5\text{O}_7$ [(M+H) $^+$] 684.2458, found 684.2463.

Ethyl 1'-methyl-2-(naphthalen-1-yl)-5-(4-nitrophenyl)-2'-oxo-3-phenylspiro[imidazolidine-4,3'-indoline]-1-carboxylate (7g). According to the general procedure, to a solution of aldehyde **2d** (100 mg, 0.64 mmol), amine **3a** (60 mg, 0.64 mmol), (*E*)-ethyl 4-nitrobenzylidene carbamate **6c** (155 mg, 0.69 mmol) and copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1c** (100 mg, 0.58 mmol). Purification furnished the corresponding spiroindoloimidazolidine **7g** as a white solid; yield: 285 mg (82%); $R_f = 0.42$ (hexane/EtOAc, 9:1). mp 167-169 $^\circ\text{C}$. IR (neat): ν_{max} 3058, 2835, 2815, 1784, 1712, 1673, 1597, 1498, 1472, 1477, 1176, 1097, 1102, 753, 685, 653 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 7.42 (d, $J = 2.8$, 1H, ArH), 7.24-7.30 (m, 1H, ArH), 7.18-7.17 (m, 9H, ArH), 7.00-7.04 (m, 1H, ArH), 6.83-6.87 (m, 2H, ArH), 6.58-6.73 (m, 6H, ArH), 5.82 (d, $J = 9.6$ Hz, 1H, CH), 4.27 (d, $J = 9.2$ Hz, 1H, CH), 3.45-3.51 (m, 2H), 2.88 (s, 3H, OCH_3),

0.89 (t, $J = 9.2$ Hz, 3H, NCH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 174.89 (C=O), 157.58 (C=O), 142.74 (C=O), 141.25 (quat-C), 135.50 (quat-C), 134.95 (quat-C), 134.33 (quat-C), 132.75 (quat-C), 131.93 (quat-C), 131.89 (quat-C), 131.23 (CH), 131.17 (CH), 130.40 (CH), 128.62 (CH), 127.90 (CH), 127.65 (CH), 127.48 (CH), 127.33 (CH), 126.90 (CH), 126.35 (CH), 122.95 (quat-C), 122.14 (CH), 121.47 (CH), 112.65 (CH), 107.14 (CH), 67.95 (quat-C), 57.62 (CH), 54.01 (CH), 28.23 (CH), 25.29 (CH), 11.01 (CH₃); HRMS (ESI) Calcd for C₃₆H₃₀N₄O₅ [(M+H)⁺] 599.2294, found 599.2311.

1'-Methyl-1,5-bis(4-nitrophenyl)-3-phenyl-2-(thiophen-2-yl)spiro[imidazolidine-4,3'-indolin]-2'-one (7h). According to the general procedure, to a solution of aldehyde **2g** (70 mg, 0.63 mmol), amine **3a** (60 mg, 0.64 mmol), (*E*)-4-nitro-*N*-(4-nitrobenzylidene)aniline **6b** (190 mg, 0.70 mmol) and copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1c** (100 mg, 0.58 mmol). Purification furnished the corresponding spiroindoloimidazolidine **7h** as a white solid; yield: 287 mg (82 %): R_f = 0.42 (hexane/EtOAc, 7:3); R_f = 0.48 (hexane/EtOAc, 7:3); mp 153-154 °C. IR (neat): ν_{\max} 3095, 2913, 2843, 1779, 1710, 1611, 1155, 1027, 1013, 690, 687 cm⁻¹; ¹H NMR (CDCl₃, 400 MHz) δ 7.61 (t, $J = 3.6$ Hz, 3H, ArH), 7.45 (t, $J = 7.6$ Hz, 2H, ArH), 7.29-7.33 (m, 6H, ArH), 7.17 (t, $J = 7.6$ Hz, 3H, ArH), 7.02-7.10 (m, 7H, ArH), 5.19 (s, 1H, CH), 3.54 (s, 3H, NCH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 173.02 (C=O), 160.38 (C=O), 155.7 (C=O), 150.67 (quat-C), 148.99 (quat-C), 148.73 (quat-C), 143.7 (quat-C), 142.02 (CH), 132.47 (CH), 131.72 (quat-C), 130.56 (CH), 130.41 (CH), 129.27 (CH), 129.03 (CH), 128.42 (CH), 127.00 (quat-C), 125.00 (CH), 123.75 (CH), 123.65 (CH), 119.14 (CH), 114.40 (CH), 114.32 (CH), 108.94 (CH), 79.1 (CH), 74.56 (quat-C), 55.41 (CH), 34.25 (CH₃); HRMS (ESI) Calcd for C₃₃H₂₅N₅O₅S [(M+Na)⁺] 626.1474, found 626.1487.

tert-Butyl 2-(4-nitrophenyl)-2'-oxo-3,5-diphenyl-1-tosylspiro[imidazolidine-4,3'-indoline]-1'-carboxylate (7i). According to the general procedure, to a solution of aldehyde **2e** (65 mg,

0.42 mmol), amine **3a** (40 mg, 0.43 mmol), *N*-benzylidene benzenesulfonamide **6a** (120 mg, 0.46 mmol) and copper(I) thiophenecarboxylate (0.7 mg, 1 mol %) catalyst in dichloroethane was transferred diazoamide **1e** (100 mg, 0.39 mmol). Purification gave the corresponding product spiroindoloimidazolidine **7i** as a white solid; yield : 229 mg (82%); mp 167-169 °C; IR (neat): ν_{\max} 3062, 2942, 2934, 1768, 1728, 1647, 1636, 1393, 1380, 1261, 1075, 1103, 732, 676 cm^{-1} ; ^1H NMR (CDCl_3 , 400MHz) δ 7.55-7.61 (m, 3H, ArH), 7.38-7.51 (m, 8H, ArH), 7.29 (s, 1H, ArH), 7.16-7.20 (m, 3H, ArH), 7.00-7.06 (m, 3H, ArH), 6.94 (d, $J = 7.6$ Hz, 2H, ArH), 6.80 (d, $J = 7.6$ Hz, 2H, ArH), 6.61-6.63 (m, 1H, ArH), 5.58 (s, 1H, CH), 2.62 (s, 3H, CH_3), 0.79 (s, 9H, *N*boc); ^{13}C NMR (CDCl_3 , 100 MHz) δ 174.10 (C=O), 148.63 (C=O), 144.76 (C=O), 144.31 (*quat-C*), 140.58 (*quat-C*), 136.39 (*quat-C*), 133.51 (*quat-C*), 131.76 (CH), 130.47 (CH), 130.36 (CH), 130.23 (*quat-C*), 130.13 (*quat-C*), 129.99 (CH), 128.29 (*quat-C*), 126.23 (CH), 124.90 (CH), 124.71 (CH), 122.43 (CH), 120.45 (CH), 115.32 (CH), 110.21 (CH), 94.63 (CH), 86.54 (*quat-C*), 78.19 (CH), 75.62 (*quat-C*), 26.96 (CH), 19.27 (*N*boc); HRMS (ESI) Calcd for $\text{C}_{40}\text{H}_{36}\text{N}_4\text{O}_7\text{S}$ [(M+Na) $^+$] 739.2202, found 739.2219.

2-(4-Bromophenyl)-1'-methyl-3,5-diphenyl-1-tosylspiro[imidazolidine-4,3'-indolin]-2'-one (**7j**). According to the general procedure, to a solution of aldehyde **2f** (120 mg, 0.65 mmol), amine **3a** (60 mg, 0.64 mmol), *N*-benzylidene benzenesulfonamide **6a** (180 mg, 0.69 mmol) and copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloromethane was transferred diazoamide **1c** (100 mg, 0.58 mmol). Purification furnished the corresponding spiroindoloimidazolidine **7j** as a white solid; yield: 297 mg (77%); Eluent: hexane/EtOAc, 90:10. mp 163-165 °C; IR (neat): ν_{\max} 3062, 2924, 2854, 1780, 1711, 1613, 1597, 1492, 1455, 1435, 1379, 1310, 1261, 1176, 1029, 732, 693, 667 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 7.75-7.83 (m, 3H, ArH), 7.53-7.71 (m, 10H, ArH), 7.49 (s, 1H, ArH), 7.20-7.40 (m, 4H, ArH), 7.12 (d, $J = 7.2$ Hz, 2H, ArH), 7.01 (d, $J = 8$ Hz, 2H, ArH), 6.81-6.83 (m, 1H, ArH), 5.78 (s, 1H, CH), 3.28 (s, 3H, CH_3), 2.81 (s, 3H, NCH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 172.16

(C=O), 146.69 (C=O), 142.83 (C=O), 142.37 (*quat-C*), 138.64 (*quat-C*), 134.45 (*quat-C*), 131.57 (CH), 129.82 (CH), 128.53 (CH), 128.42 (CH), 128.29 (*quat-C*), 128.19 (CH), 128.05 (CH), 126.35 (CH), 124.29 (*quat-C*), 122.78 (CH), 122.97 (*quat-C*), 122.78 (CH), 120.49 (CH), 118.51 (CH), 113.38 (CH), 108.27 (CH), 92.69 (CH), 84.61 (*quat-C*), 75.93 (CH), 73.68 (*quat-C*), 31.10 (CH), 25.02 (NCH₃); HRMS (ESI) Calcd for C₃₆H₃₀BrN₃O₃S [(M+H)⁺]: 664.1270 found 664.1282.

Ethyl 1'-benzyl-3-methyl-5-(4-nitrophenyl)-2'-oxo-2-phenylspiro[imidazolidine-4,3'-indoline]-1-carboxylate (7k). According to the general procedure, to a solution of aldehyde **2a** (45 mg, 0.43 mmol), amine **3b** (30 mg, 0.45 mmol), (*E*)-ethyl 4-nitrobenzylidene carbamate **6c** (107 mg, 0.48 mmol) and copper(I) thiophenecarboxylate (0.8 mg, 1 mol %) in dichloroethane was transferred diazoamide **1a** (100 mg, 0.4 mmol). Purification furnished the corresponding spiroindoloimidazolidine **7k** as a white solid; yield: 164 mg (73%). mp 153-154 °C. IR (neat): ν_{\max} 3062, 2915, 2874, 1780, 1713, 1613, 1264, 1166, 1025, 1003, 731, 690, 687 cm⁻¹. ¹H NMR (CDCl₃, 400 MHz) δ 8.12 (d, *J* = 7.2 Hz, 1H, ArH), 7.80-7.56 (m, 2H, ArH), 7.65-7.47 (m, 6H, ArH), 7.61-7.64 (m, 4H, ArH), 7.50-7.54 (m, 1H, ArH), 6.23-6.37 (m, 3H, ArH), 7.10-7.19 (m, 6H, ArH), 6.31 (d, *J* = 9.2 Hz, 1H, CH), 5.79 (m, 2H, CH), 3.45-3.51 (d, *J* = 9.2 Hz, 1H, CH), 2.13 (s, 3H, CH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 174.89 (C=O), 157.57 (C=O), 142.73 (C=O), 141.25 (*quat-C*), 135.49 (*quat-C*), 134.95 (*quat-C*), 134.33 (*quat-C*), 132.75 (*quat-C*), 131.93 (*quat-C*), 131.89 (*quat-C*), 131.22 (CH), 131.17 (CH), 130.39 (CH), 128.61 (CH), 127.90 (CH), 127.64 (CH), 127.48 (CH), 127.32 (CH), 126.90 (CH), 126.34 (*quat-C*), 122.95 (CH), 122.14 (CH), 121.46 (CH), 112.64 (CH), 107.14 (CH), 67.95 (CH), 62.81 (*quat-C*), 57.62 (CH), 54.01 (CH), 28.22 (CH₂), 11.01 (CH₃); HRMS (ESI) Calcd for C₃₃H₃₀N₄O₅ [(M+H)⁺]: 563.2294, found 563.2305.

Ethyl 1'-benzyl-3-(4-bromophenyl)-5-(4-nitrophenyl)-2'-oxo-2-phenylspiro[imidazolidine-4,3'-indoline]-1-carboxylate (7l). According to the general procedure, to a solution of aldehyde **2a** (45 mg, 0.42 mmol), amine **3c** (75 mg, 0.44 mmol), (*E*)-ethyl 4-nitrobenzylidene-carbamate **6c** (105 mg, 0.47 mmol) and copper(I) thiophenecarboxylate (0.8 mg, 1 mol %) in dichloroethane was transferred diazoamide **1a** (100 mg, 0.4 mmol). Purification furnished the corresponding spiroindoloimidazolidine **7l** as a white solid; yield: 205 mg (73%). $R_f = 0.42$ (hexane/EtOAc, 9:1); mp 167-169 °C. IR (neat): ν_{\max} 3052, 2911, 2862, 1780, 1711, 1613, 1486, 1455, 1379, 1342, 1261, 1035, 863, 685, 672 cm^{-1} . ^1H NMR (CDCl_3 , 400 MHz) δ 7.41 (d, $J = 4.8$ Hz, 1H, ArH), 7.24-7.30 (m, 2H, ArH), 7.11-7.21 (m, 10H, ArH), 7.02 (t, $J = 4.8$ Hz, 1H, ArH), 6.83-6.87 (m, 2H, ArH), 6.67-6.73 (m, 5H, ArH), 6.01 (d, $J = 5.2$ Hz, 1H, ArH), 5.81 (d, $J = 9.2$ Hz, 1H, CH), 5.29-5.28 (m, 2H, CH_2), 4.28 (d, $J = 9.6$ Hz, 1H, CH), 3.45-3.51 (m, 2H, CH), 0.89 (t, $J = 4$ Hz, 3H, CH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 175.88 (C=O), 158.57 (C=O), 143.73 (C=O), 1421.24 (*quat-C*), 136.49 (*quat-C*), 134.94 (*quat-C*), 135.32 (*quat-C*), 133.75 (*quat-C*), 132.92 (*quat-C*), 132.88 (*quat-C*), 132.22 (CH), 131.16 (CH), 131.39 (CH), 129.61 (CH), 128.89 (CH), 127.64 (CH), 127.47 (CH), 127.32 (CH), 127.89 (CH), 127.34 (*quat-C*), 123.94 (CH), 123.13 (CH), 122.46 (CH), 113.64 (CH), 108.14 (CH), 68.94 (CH), 63.81 (*quat-C*), 58.62 (CH), 28.22 (CH_2), 26.28 (CH_2), 11.99 (CH_3). HRMS (ESI) Calcd for $\text{C}_{38}\text{H}_{31}\text{BrN}_4\text{O}_5$ [(M+H) $^+$] 703.1556, found 703.1569.

Diethyl 4'-(4-nitrophenyl)-1-methyl-2-oxo-5'-phenylspiro[indoline-3,3'-[1,2,4]triazolidine]-1',2'-dicarboxylate (9f). According to the general procedure, to a solution of aldehyde **2e** (95 mg, 0.63 mmol), amine **3a** (60 mg, 0.64 mmol), DEAD **8b** (120 mg, 0.69 mmol) and copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1c** (100 mg, 0.58 mmol). Purification furnished the corresponding spiroindolotriazolidine **9f** as a white solid; yield: 237 mg (73 %); $R_f = 0.32$ (hexane/EtOAc, 8:2); mp 167-169 °C. IR (neat): ν_{\max} 3062, 2924, 2854, 1780, 1711, 1613, 1597, 1492, 1455,

1435, 1379, 1310, 1261, 1176, 1029, 732, 693, 667 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 8.03-8.05 (m, 2H, ArH), 7.81 (d, $J = 7.6$ Hz, 1H, ArH), 7.48-7.57 (m, 2H, ArH), 7.39-7.42 (m, 5H, ArH), 7.22-7.27 (m, 4H, ArH), 6.95-7.22 (m, 2H, ArH), 6.84 (d, $J = 8.0$ Hz, 2H, ArH), 4.53 (s, 1H, CH), 3.09 (s, 3H, NCH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 176.07 (C=O), 171.84 (C=O), 163.66 (C=O), 143.81 (quat-C), 137.09 (quat-C), 134.78 (quat-C), 133.99 (CH), 130.49 (quat-C), 130.07 (CH), 129.73 (quat-C), 129.56 (quat-C), 129.31 (quat-C), 129.18 (quat-C), 128.77 (quat-C), 128.70 (quat-C), 128.63 (CH), 127.82 (quat-C), 125.53 (CH), 122.85 (CH), 108.63 (CH), 52.42 (OCH_3), 35.22 (NCH_3); HRMS (ESI) Calcd for $\text{C}_{30}\text{H}_{22}\text{N}_6\text{O}_5$ [(M+Na) $^+$] 569.1549, found 569.1562.

Diethyl *4'-(4-bromophenyl)-1-ethyl-5-nitro-2-oxo-5'-phenylspiro[indoline-3,3'-[1,2,4]triazolidine]-1',2'-dicarboxylate (9g)*. According to the general procedure, to a solution of aldehyde **2f** (110 mg, 0.60 mmol), amine **3a** (55 mg, 0.59 mmol), DEAD **8b** (110 mg, 0.63 mmol) and copper(I) thiophenecarboxylate (1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1b** (100 mg, 0.53 mmol). Purification furnished the corresponding spiroindolotriazolidine **9g** as a white solid; yield: 233 mg (74 %): $R_f = 0.42$ (hexane/EtOAc, 9:1; mp 167-169 $^\circ\text{C}$. IR (neat): ν_{max} 3059, 2932, 2864, 1778, 1721, 1653, 1587, 1492, 1468, 1380, 1261, 1176, 1075, 1103, 732, 693, 667 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 8.03-8.06 (m, 2H, ArH), 7.55 (d, $J = 5.6$ Hz, 2H, ArH), 7.51-7.54 (m, 3H, ArH), 7.38-7.42 (m, 5H, ArH), 7.22-7.27 (m, 4H, ArH), 6.95-7.21 (m, 1H, ArH), 6.85 (d, $J = 8.0$ Hz, 1H, ArH), 4.53 (s, 1H, CH), 3.73 (q, $J = 7.2$ Hz, 2H, NCH_2), 1.23 (t, $J = 7.2$ Hz, 3H, CH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 175.78 (C=O), 171.56 (C=O), 161.22 (C=O), 143.53 (quat-C), 136.81 (quat-C), 133.71 (CH), 130.20 (CH), 129.78 (CH), 129.44 (quat-C), 129.28 (quat-C), 129.02 (CH), 128.89 (CH), 128.49 (CH), 128.42 (CH), 128.35 (CH), 127.54 (CH), 125.24 (CH), 122.57 (CH), 108.35 (CH), 52.14 (CH), 34.94 (CH_2), 12.72 (CH_3); HRMS (ESI) Calcd for $\text{C}_{31}\text{H}_{24}\text{BrN}_5\text{O}_3$ [(M+Na) $^+$] 616.0960, found 616.0978.

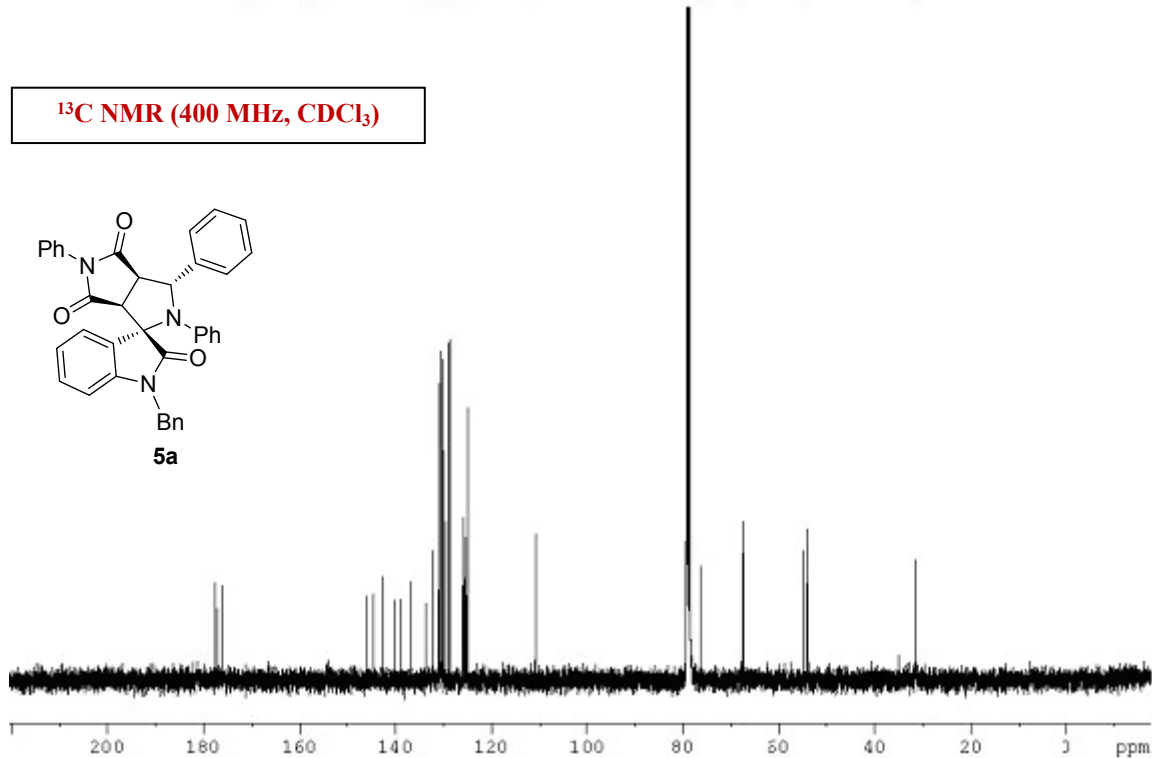
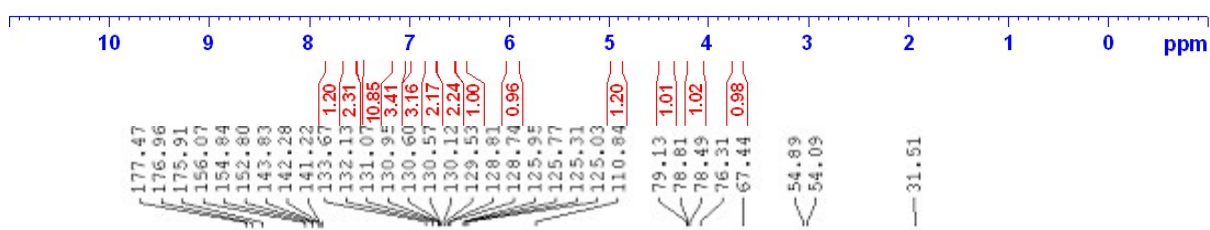
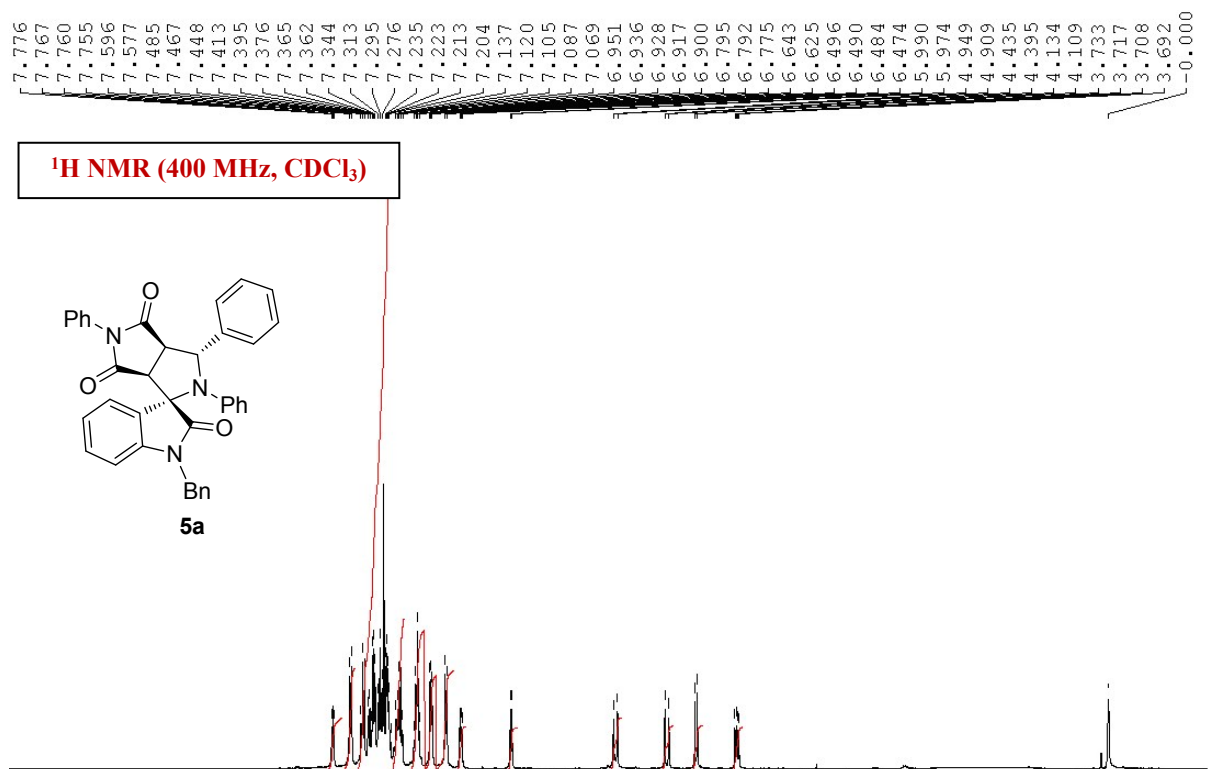
Diethyl *1,4'-dimethyl-2-oxo-5'-phenylspiro[indoline-3,3'-[1,2,4]triazolidine]-1',2'-dicarboxylate (9h)*. According to the general procedure, to a solution of aldehyde **2a** (65 mg, 0.61 mmol), amine **3b** (45 mg, 0.67 mmol), DEAD **8b** (120 mg, 0.69 mmol) and copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1c** (100 mg, 0.58 mmol). Purification furnished the corresponding spiroindolotriazolidine **9h** as a white solid; yield: 180 mg (71 %): $R_f = 0.27$ (hexane/EtOAc, 7:3); mp 153-154 °C. IR (neat): ν_{\max} 3048, 2935, 2565, 1879, 1723, 1623, 1274, 1168, 1024, 1008, 690, 687 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 7.81 (d, $J = 8$ Hz, 1H, ArH), 7.45-7.48 (m, 2H, ArH), 7.33-7.37 (m, 3H, ArH), 7.22-7.32 (m, 3H, ArH), 4.76 (d, $J = 6.4$ Hz, 1H, CH), 4.75 (q, $J = 7.2$ Hz, 2H, CH), 3.86 (d, $J = 7.2$ Hz, 1H, CH), 3.59 (d, $J = 7.2$ Hz, 1H, CH), 2.74 (s, 3H, CH_3), 1.87 (s, 3H, NCH_3), 1.24 (t, $J = 7.2$ Hz, 3H, CH_3), 1.11 (t, $J = 7.2$ Hz, 3H, CH_3); ^{13}C NMR (CDCl_3 , 100 MHz) δ 175.69 (C=O), 174.92 (C=O), 174.07 (C=O), 148.59 (*quat-C*), 140.90 (*quat-C*), 140.51 (*quat-C*), 131.89 (*quat-C*), 130.43 (CH), 129.05 (CH), 128.99 (CH), 128.75 (CH), 128.22 (CH), 127.38 (CH), 127.02 (CH), 126.72 (CH), 125.41 (CH), 123.92 (CH), 115.47 (CH), 73.85 (*quat-C*), 68.73 (CH), 53.66 (CH), 52.63 (CH), 32.54 (NCH_3), 28.12 (CH_3), 14.64 (CH), 13.27 (CH); HRMS (ESI) Calcd for $\text{C}_{23}\text{H}_{26}\text{N}_4\text{O}_5$ [(M+Na) $^+$] 461.1801, found 461.1817.

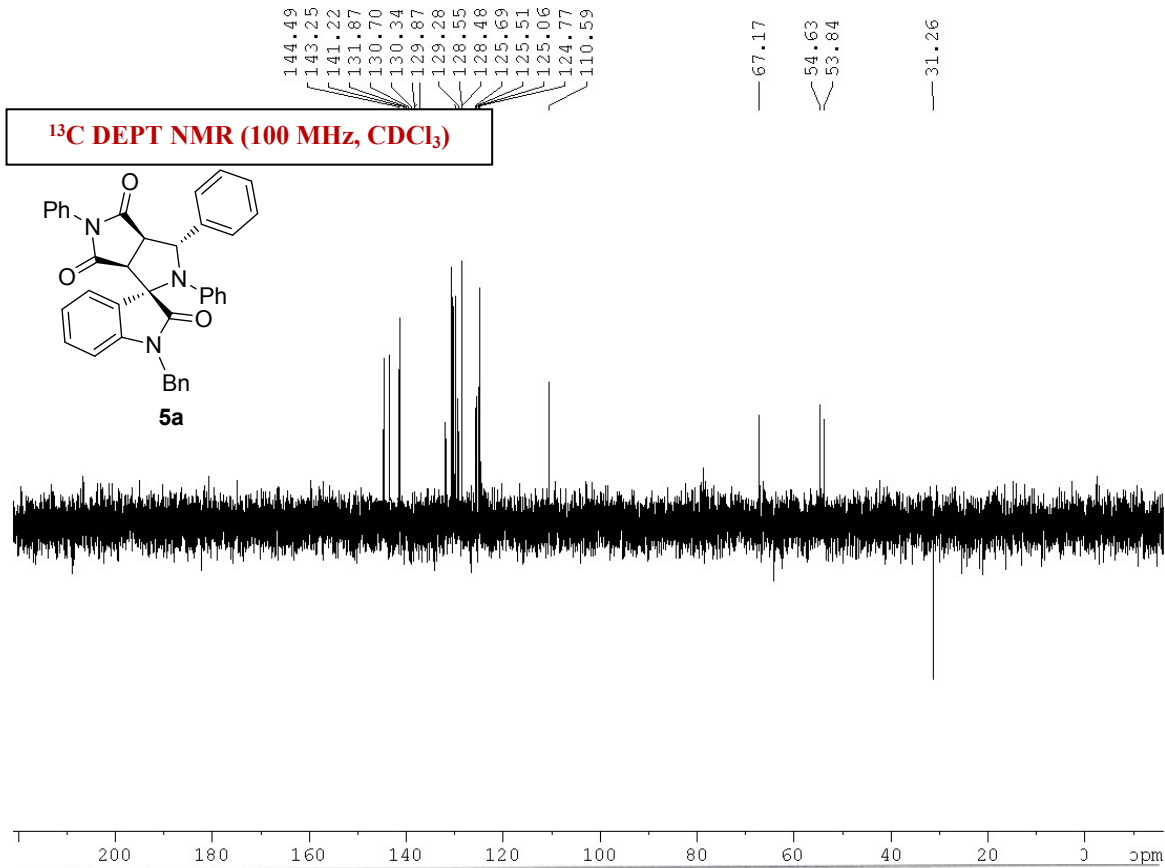
tert-Butyl *1',2'-diethyl* *4'-(4-bromophenyl)-5-nitro-2-oxo-5'-phenylspiro[indoline-3,3'-[1,2,4]triazolidine]-1,1',2'-tricarboxylate (9i)*. According to the general procedure, to a solution of aldehyde **2a** (45 mg, 0.4 mmol), amine **3c** (80 mg, 0.4 mmol), DEAD **8b** (80 mg, 0.46 mmol) and copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1i** (100 mg, 0.33 mmol). Purification furnished the corresponding spiroindolotriazolidine **9i** as a white solid; yield : 214 mg (77%): $R_f = 0.44$ (hexane/EtOAc, 9:1); mp 167-169 °C; IR (neat): ν_{\max} 3059, 2932, 2864, 1778, 1721, 1653, 1587, 1492, 1468, 1380, 1261, 1176, 1075, 1103, 732, 693, 667 cm^{-1} ; ^1H NMR (CDCl_3 , 400 MHz) δ 7.59 (t, $J =$

7.6 Hz, 2H, ArH), 7.38 (t, $J = 7.6$ Hz, 2H, ArH), 7.30 (d, $J = 8.0$ Hz, 2H, ArH), 7.09-7.17 (m, 2H), 7.90-7.09 (m, 3H), 6.75 (d, $J = 7.6$ Hz, 1H, ArH), 7.64 (d, $J = 7.6$ Hz, 1H, ArH), 3.78-3.87 (m, 4H, CH₂), 1.30-1.35 (m, 6H, CH₃), 0.80 (s, 9H, *N*boc); ¹³C NMR (CDCl₃, 100 MHz) δ 171.45 (C=O), 167.51 (C=O), 161.92 (C=O), 143.89 (*quat-C*), 137.17 (*quat-C*), 132.56 (*quat-C*), 132.15 (CH), 129.95 (CH), 129.50 (CH), 128.91 (CH), 128.66 (CH), 125.74 (CH), 125.56 (CH), 123.53 (CH), 123.19 (CH), 118.16 (CH), 110.04 (CH), 78.18 (*quat-C*), 69.66 (*quat-C*), 63.22 (CH), 36.11 (CH), 14.64 (CH), 13.27 (CH), 10.99 (CH); HRMS (ESI) Calcd for C₃₂H₃₂BrN₅O₉ [(M+Na)⁺] 732.1281, found 732.1296.

Diethyl 4'-(4-bromophenyl)-1-methyl-2-oxo-5'-phenylspiro[indoline-3,3'-[1,2,4]triazolidine]-1',2'-dicarboxylate (9j). According to the general procedure, to a solution of aldehyde **2a** (70 mg, 0.66 mmol), amine **3c** (120 mg, 0.65 mmol), DEAD **8b** (120 mg, 0.69 mmol), copper(I) thiophenecarboxylate (1.1 mg, 1 mol %) in dichloroethane was transferred diazoamide **1c** (100 mg, 0.58 mmol). Purification furnished the corresponding spiroindolotriazolidine **9j** as a white solid; yield: 282 mg (84 %). $R_f = 0.41$ (hexane/EtOAc, 9:1); mp 163-165 °C; IR (neat): ν_{\max} 3072, 2954, 2834, 1740, 1721, 1623, 1584, 1495, 1465, 1379, 1331, 116, 732, 669 cm⁻¹; ¹H NMR (CDCl₃, 400 MHz) δ 7.61 (t, $J = 7.2$ Hz, 2H, ArH), 7.46 (t, $J = 8.0$ Hz, 1H, ArH), 7.30-7.33 (m, 4H, ArH), 7.18 (t, $J = 8.0$ Hz, 2H, ArH), 7.00-7.10 (m, 5H, ArH), 4.07 (q, $J = 7.2$ Hz, 2H, NCH₂), 3.82-3.91 (m, 1H, NCH), 3.54-3.63 (m, 1H, NCH), 2.73 (s, 3H, NCH₃), 1.27 (t, $J = 7.2$ Hz, 3H, CH₃), 1.12 (t, $J = 7.2$ Hz, 3H, CH₃); ¹³C NMR (CDCl₃, 100 MHz) δ 170.65 (C=O), 166.71 (C=O), 161.12 (C=O), 143.09 (*quat-C*), 136.37 (*quat-C*), 131.76 (*quat-C*), 137.35 (*quat-C*), 129.15 (*quat-C*), 128.70 (CH), 128.11 (CH), 127.86 (CH), 124.94 (CH), 124.78 (CH), 122.73 (CH), 122.39 (CH), 117.36 (CH), 109.24 (CH), 77.38 (*quat-C*), 76.43 (CH), 68.86 (CH₂), 62.42 (CH), 35.31 (CH₂), 25.02 (CH₃), 13.84 (CH₃), 12.47 (CH₃); HRMS (ESI) Calcd for C₂₈H₂₇BrN₄O₅ [(M+H)⁺] 579.1243, found 579.1252.

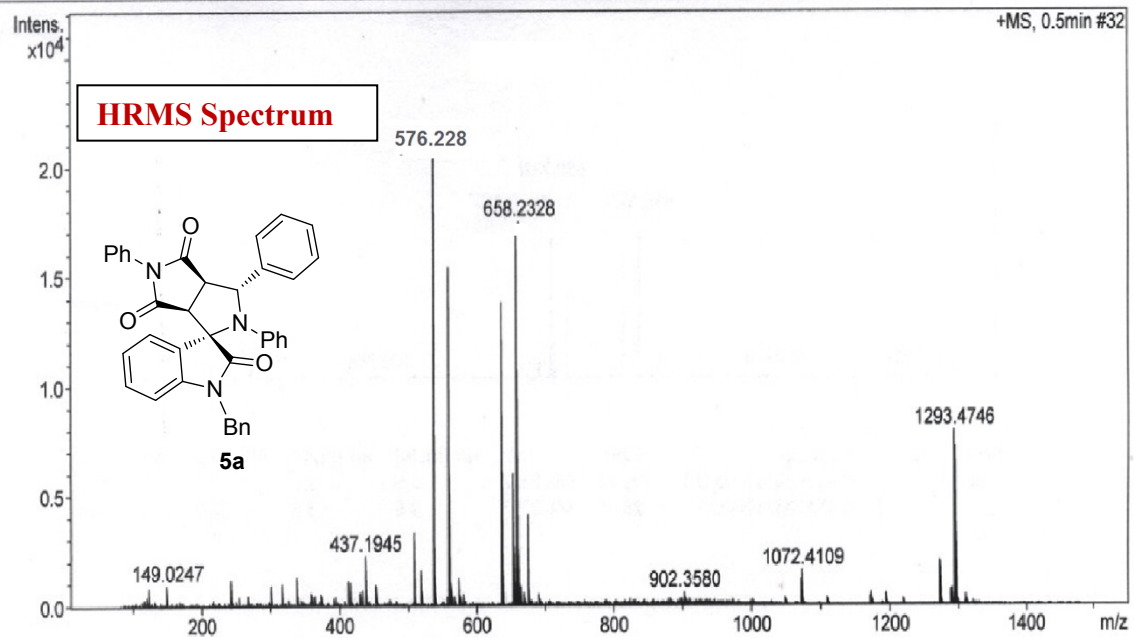
¹H NMR and ¹³C NMR, DEPT135 NMR Spectra of spiroindolopyrrolidines



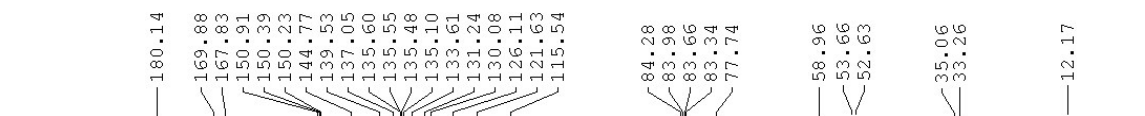
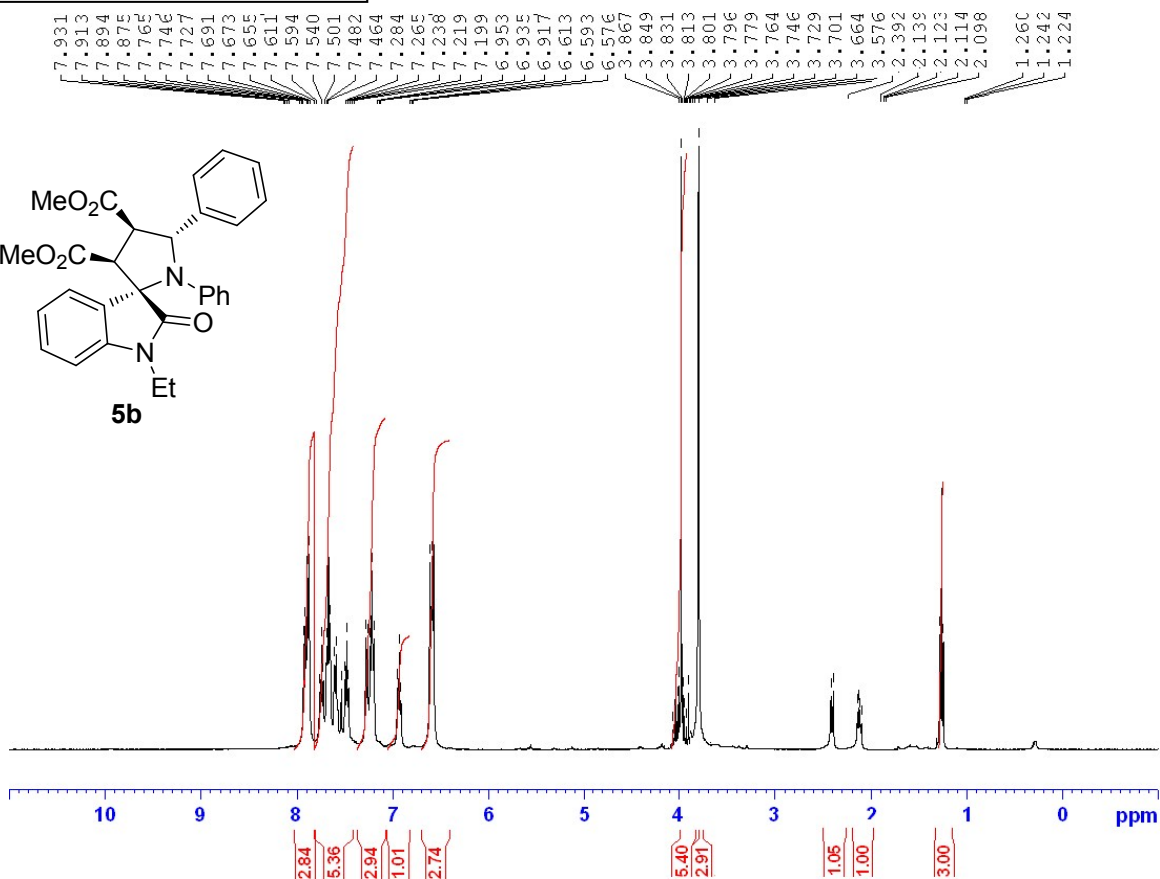
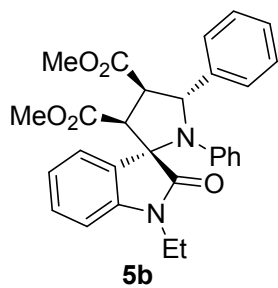


Acquisition Parameter

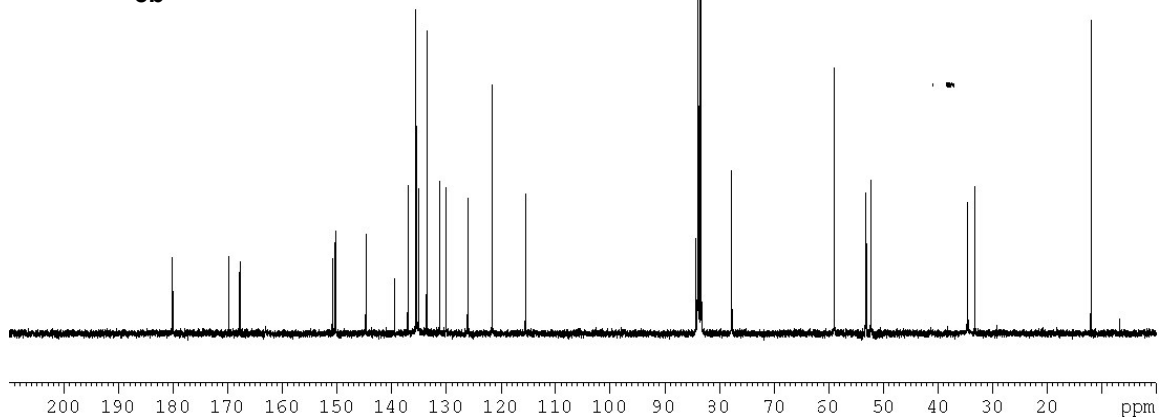
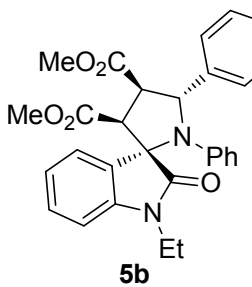
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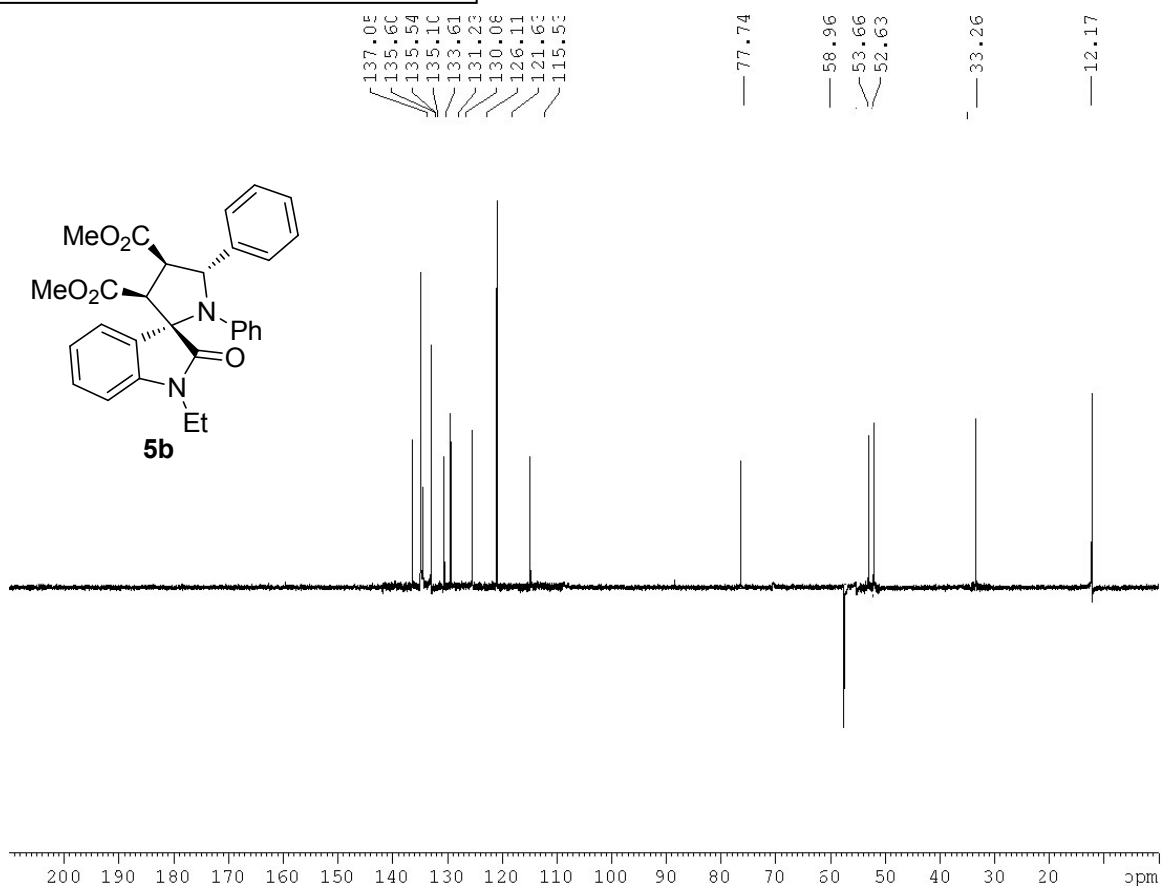
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¹³C NMR (400 MHz, CDCl₃)



¹³C DEPT NMR (100 MHz, CDCl₃)



HRMS Spectrum

Analysis Info

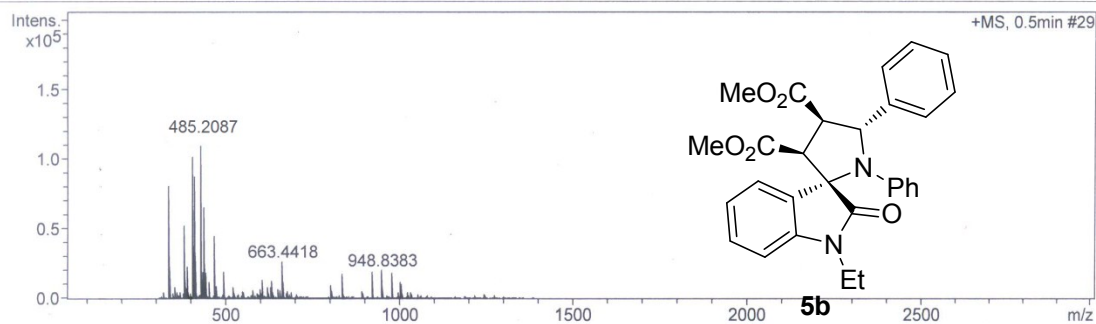
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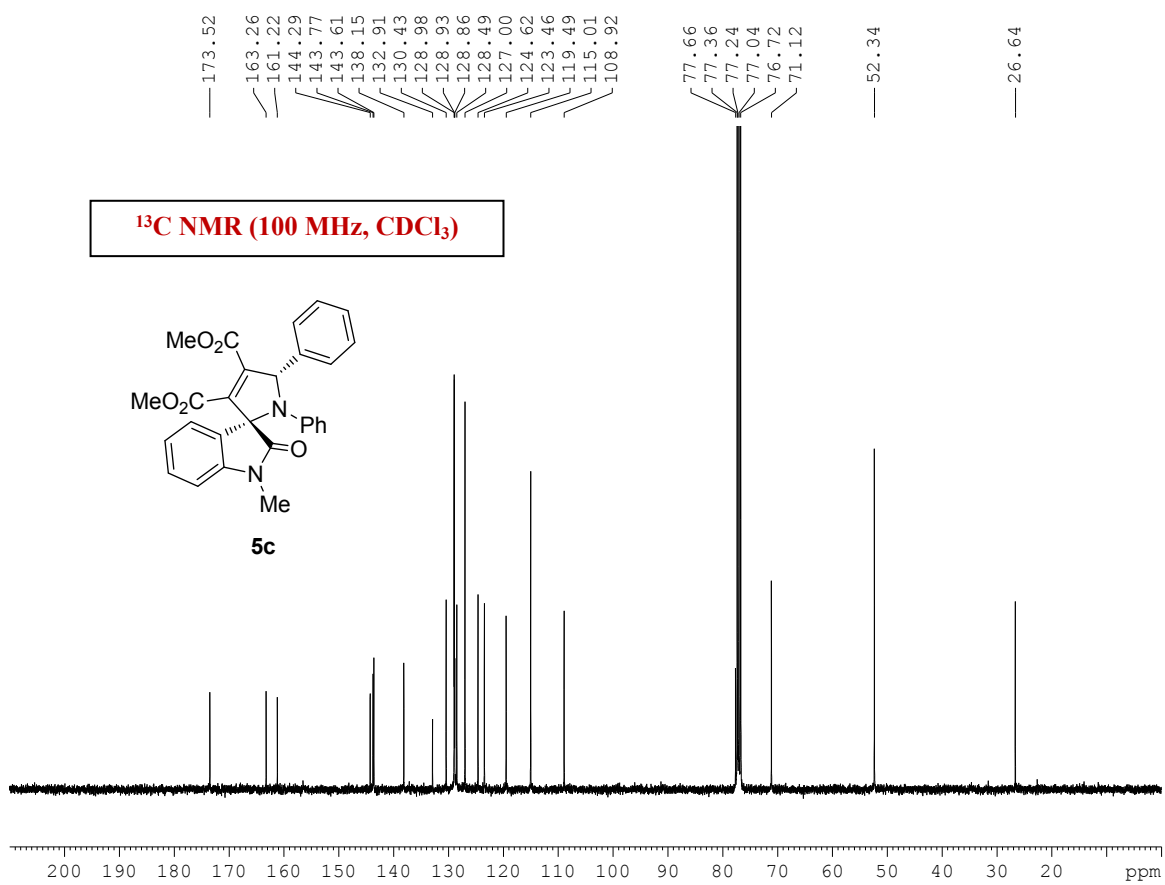
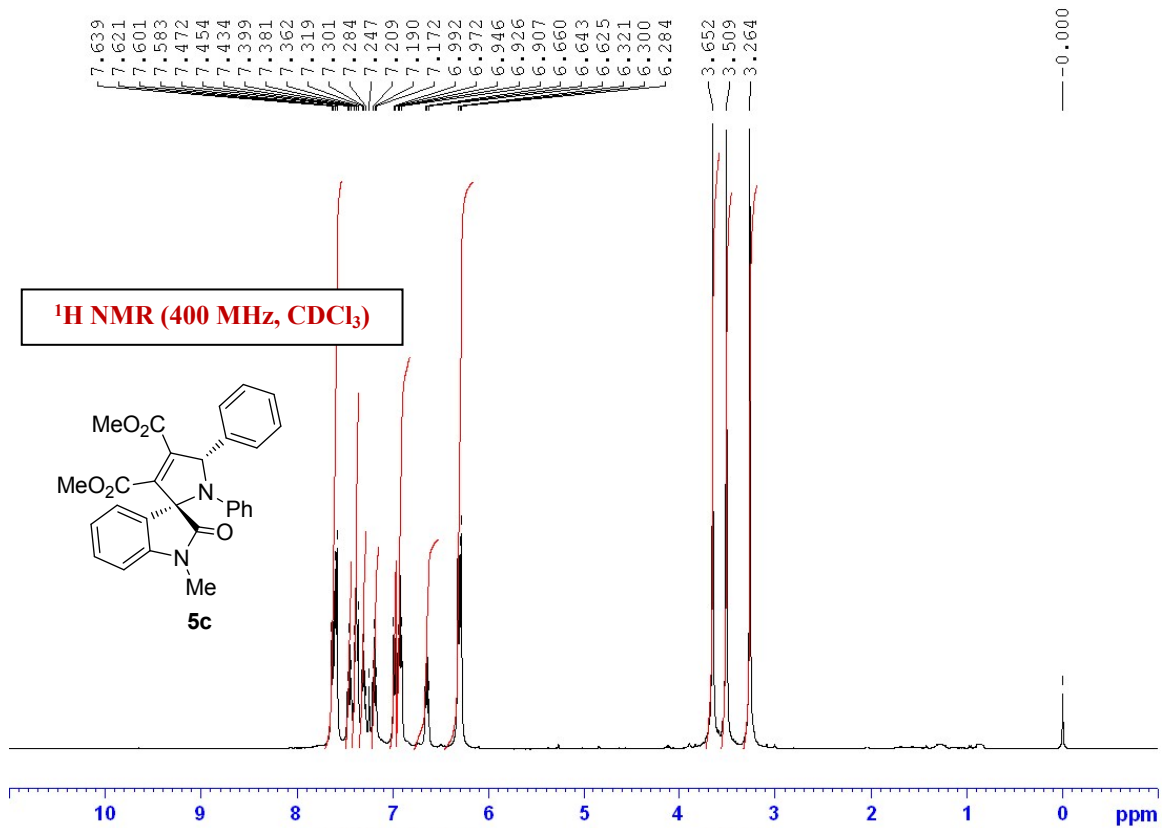
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Operator Sharma/Singh
Instrument / Ser# micrOTOF-Q II 10262

Acquisition Parameter

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Scan End	3000 m/z	Set Collision Cell RF	600.0 Vpp	Set Divert Valve	Source





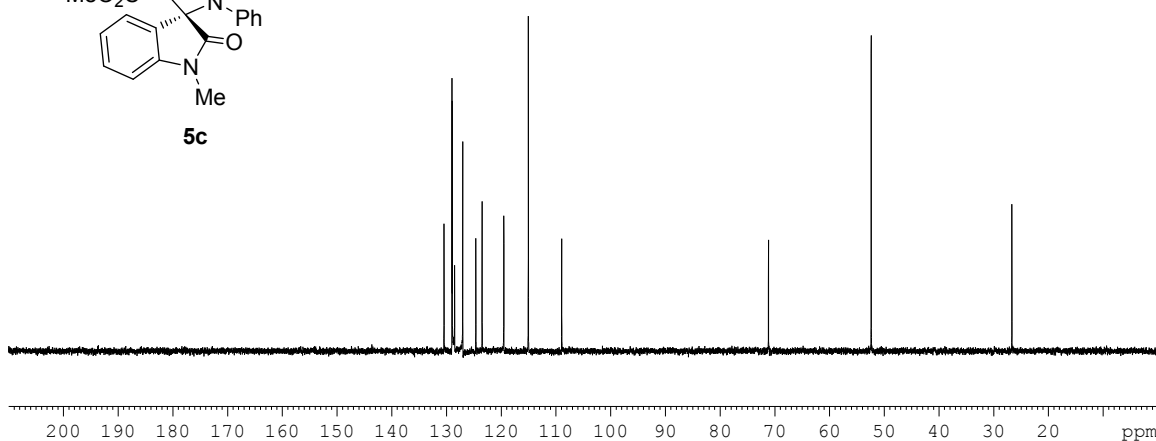
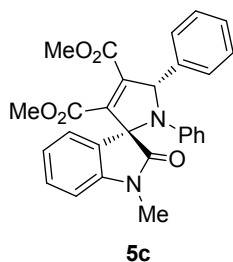
130.46
129.01
128.96
128.52
127.03
124.65
123.49
119.52
115.05
108.95

71.15

52.37

26.67

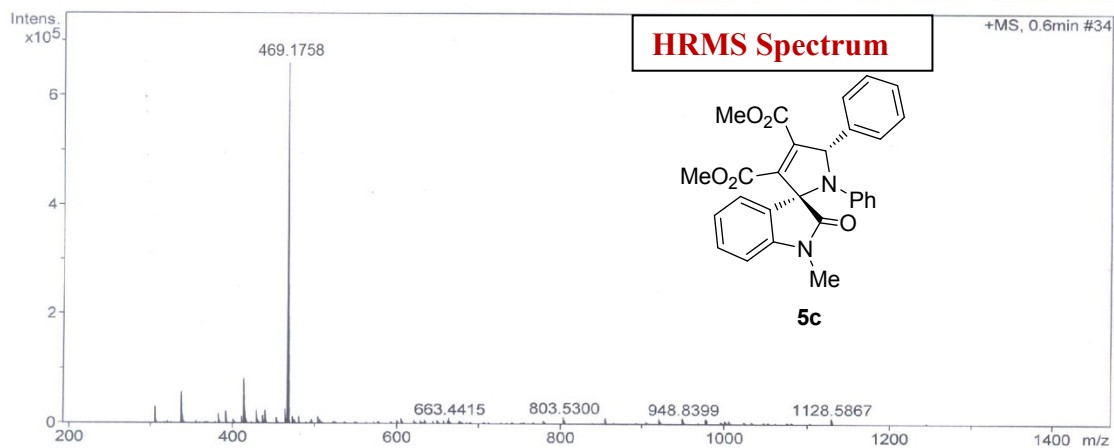
¹³C DEPT NMR (100 MHz, CDCl₃)



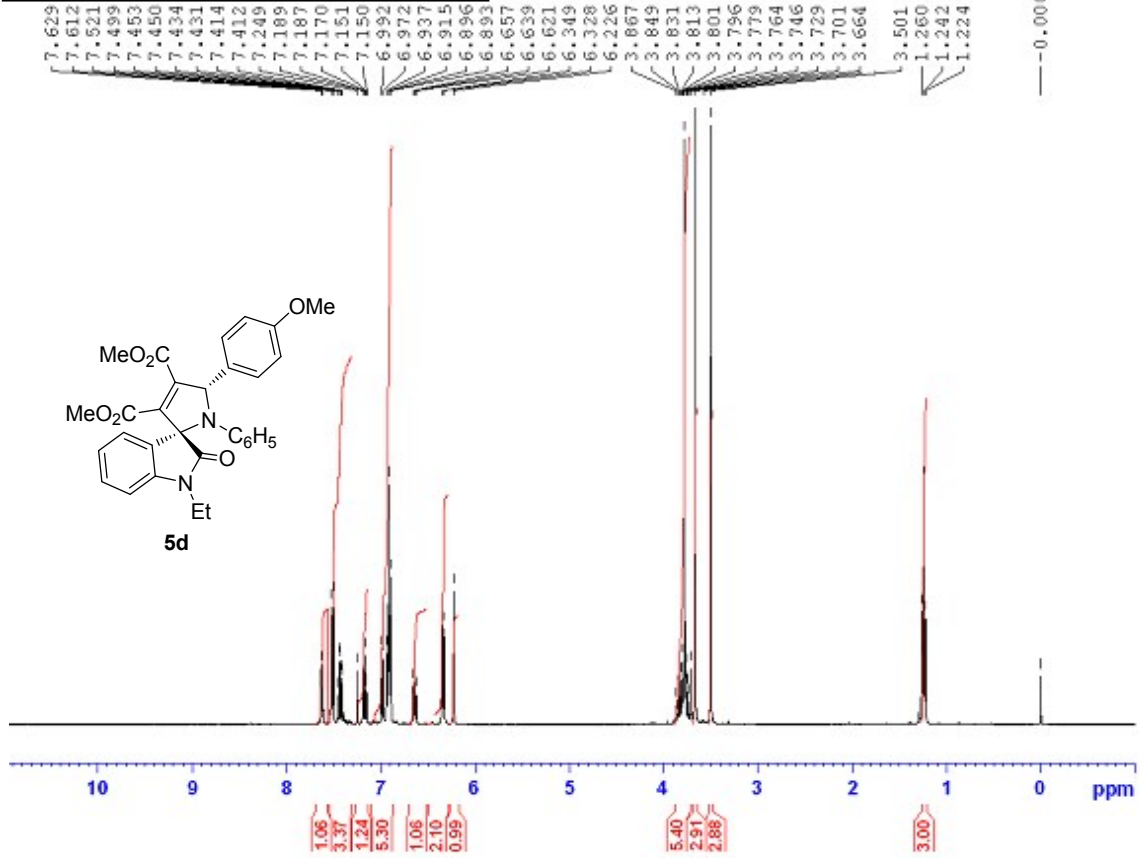
Acquisition Parameter

Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	3000 m/z	Set Collision Cell RF	600.0 Vpp	Set Divert Valve	Source

0.0 0.2 0.4 0.6 0.8 Time [min]

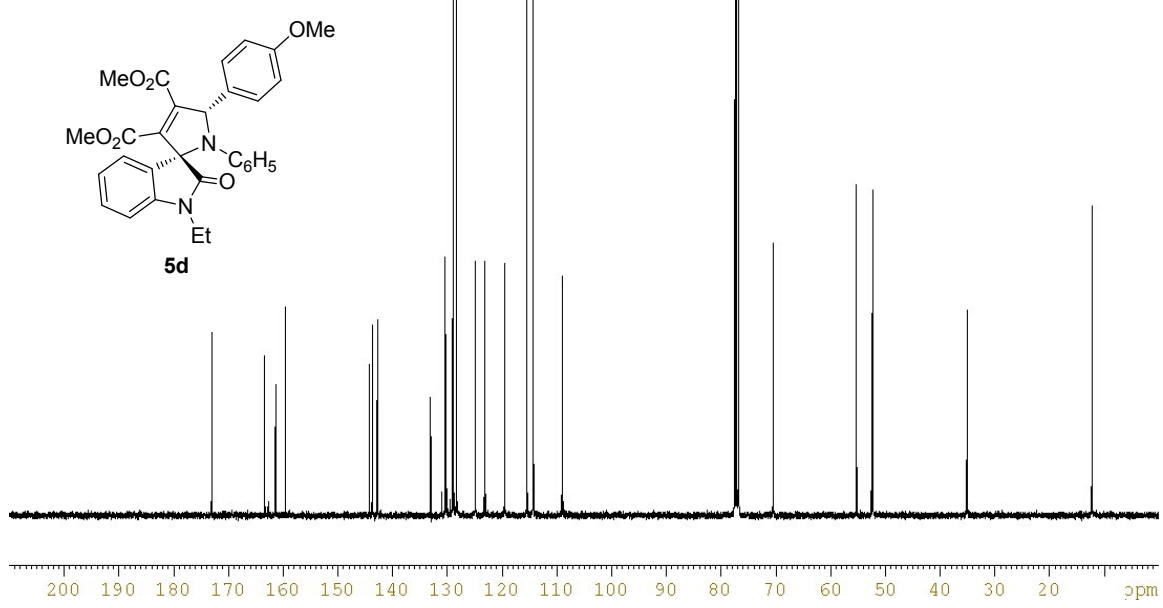


¹H NMR (400 MHz, CDCl₃)



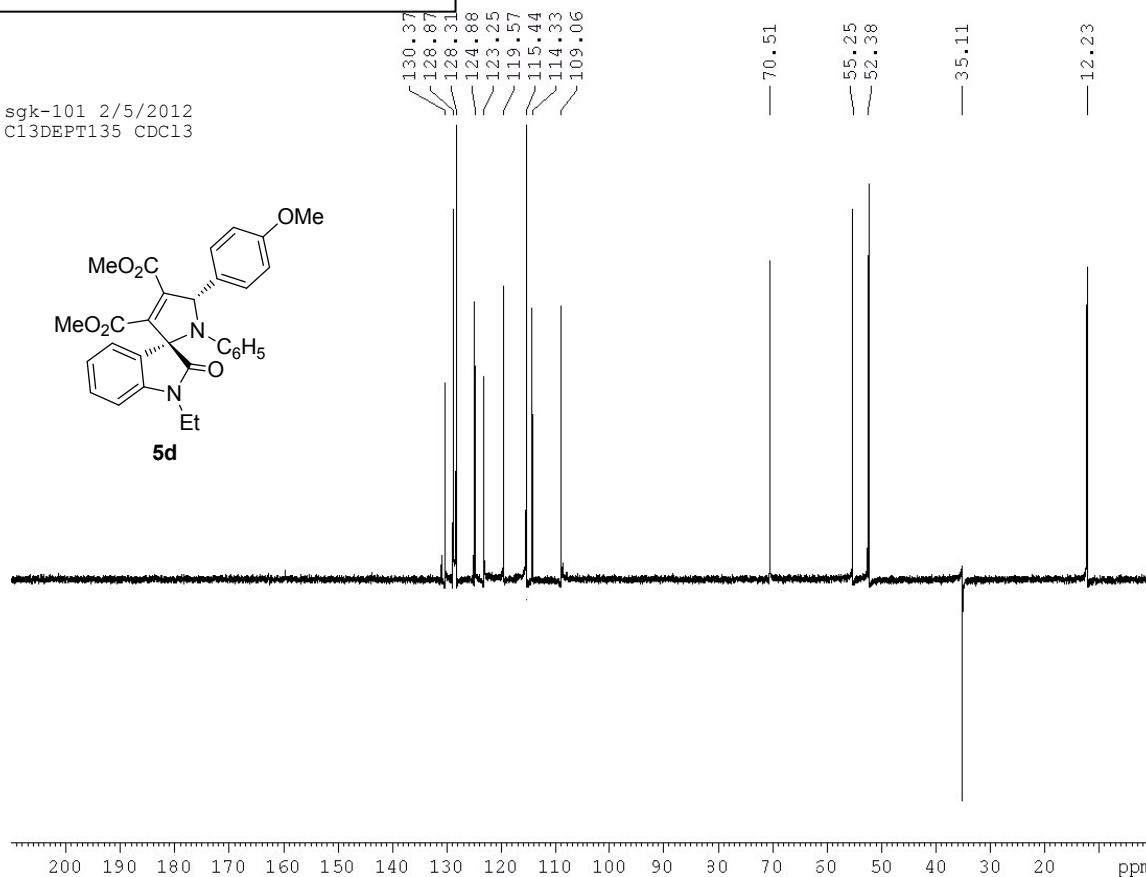
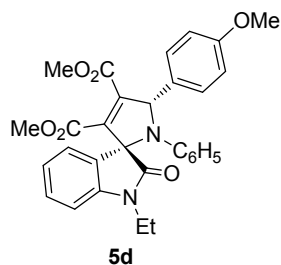
¹³C NMR (100 MHz, CDCl₃)

sgk-101 2/5/2012
C13CPD CDCl3



¹³C DEPT NMR (100 MHz, CDCl₃)

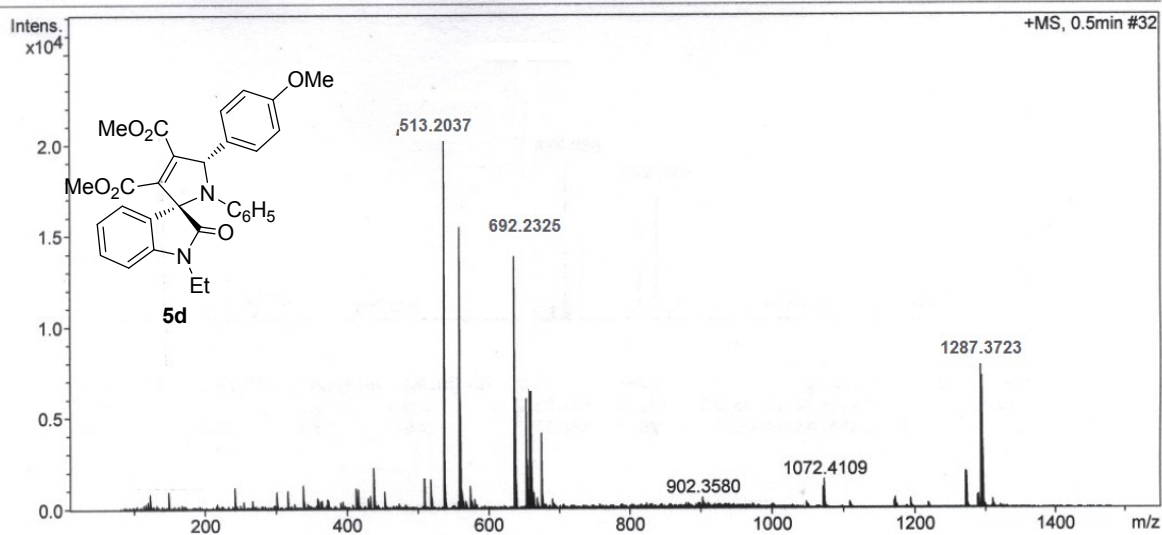
sgk-101 2/5/2012
C13DEPT135 CDCl3



HRMS Spectrum

Acquisition Parameter

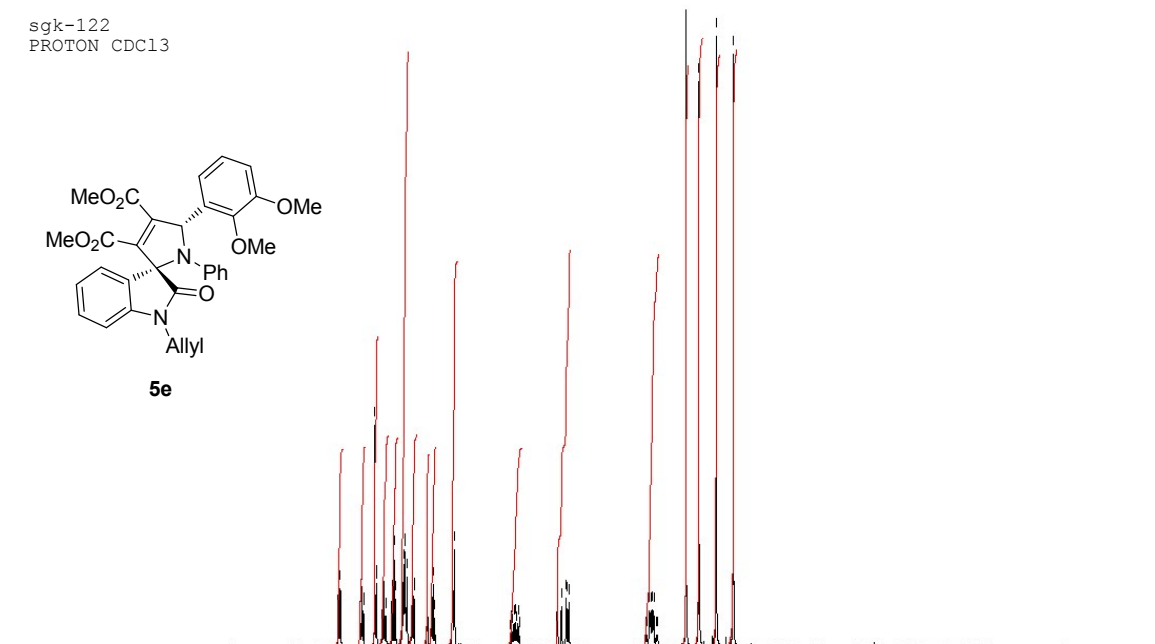
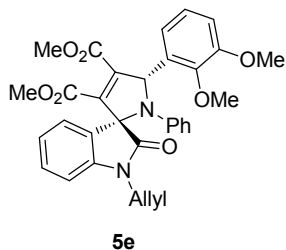
Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Not active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source



¹H NMR (400 MHz, CDCl₃)

7.566
7.565
7.548
7.344
7.341
7.324
7.322
7.305
7.302
7.188
7.170
7.167
7.107
7.106
7.088
7.069
7.068
7.007
6.987
6.967
6.901
6.890
6.882
6.871
6.868
6.850
6.799
6.796
6.779
6.776
6.639
6.598
6.580
6.561
6.375
6.355
5.734
5.717
5.282
5.279
5.239
5.236
5.189
5.186
5.163
5.160
4.320
4.307
4.290
4.275
3.938
3.805
3.623
3.445

sgk-122
PROTON CDC13

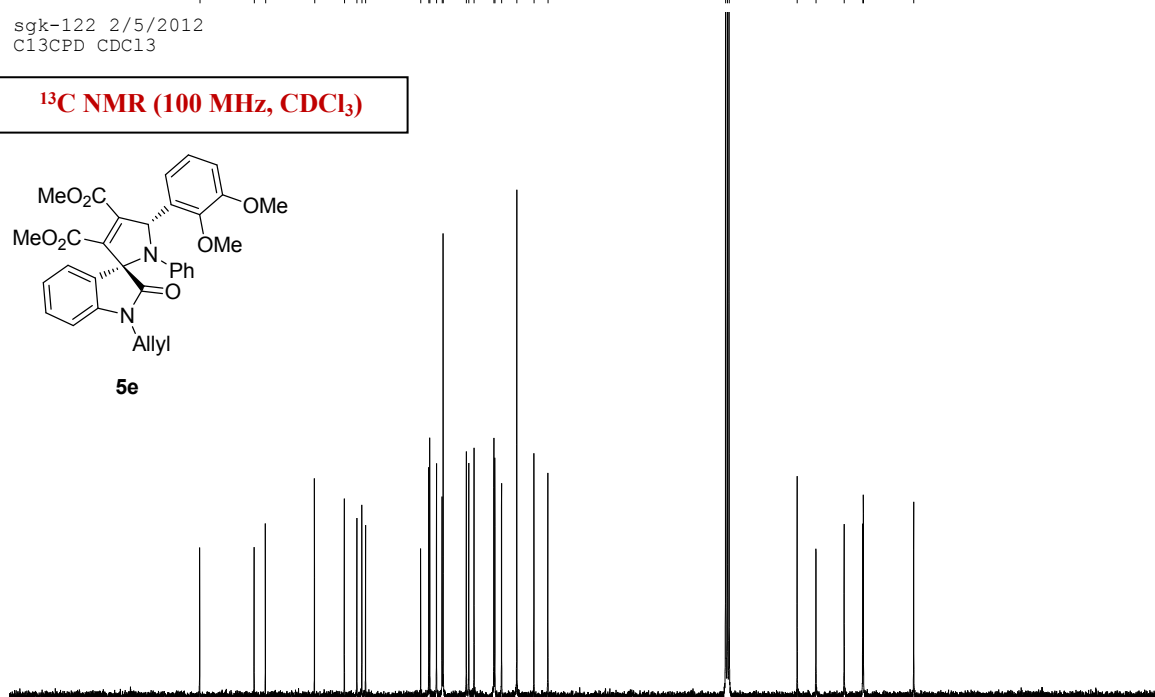
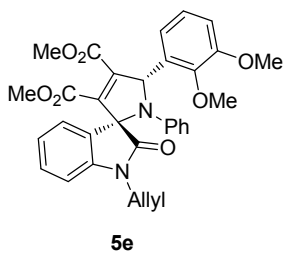


10 9 8 7 6 5 4 3 2 1 0 ppm

175.28
165.34
163.30
154.34
148.92
146.64
145.74
145.05
135.00
133.53
133.34
132.10
131.09
130.91
126.67
126.22
125.25
121.64
121.46
120.24
117.45
114.34
111.79
79.36
79.04
78.72
66.30
62.87
57.72
54.34
54.22
45.04

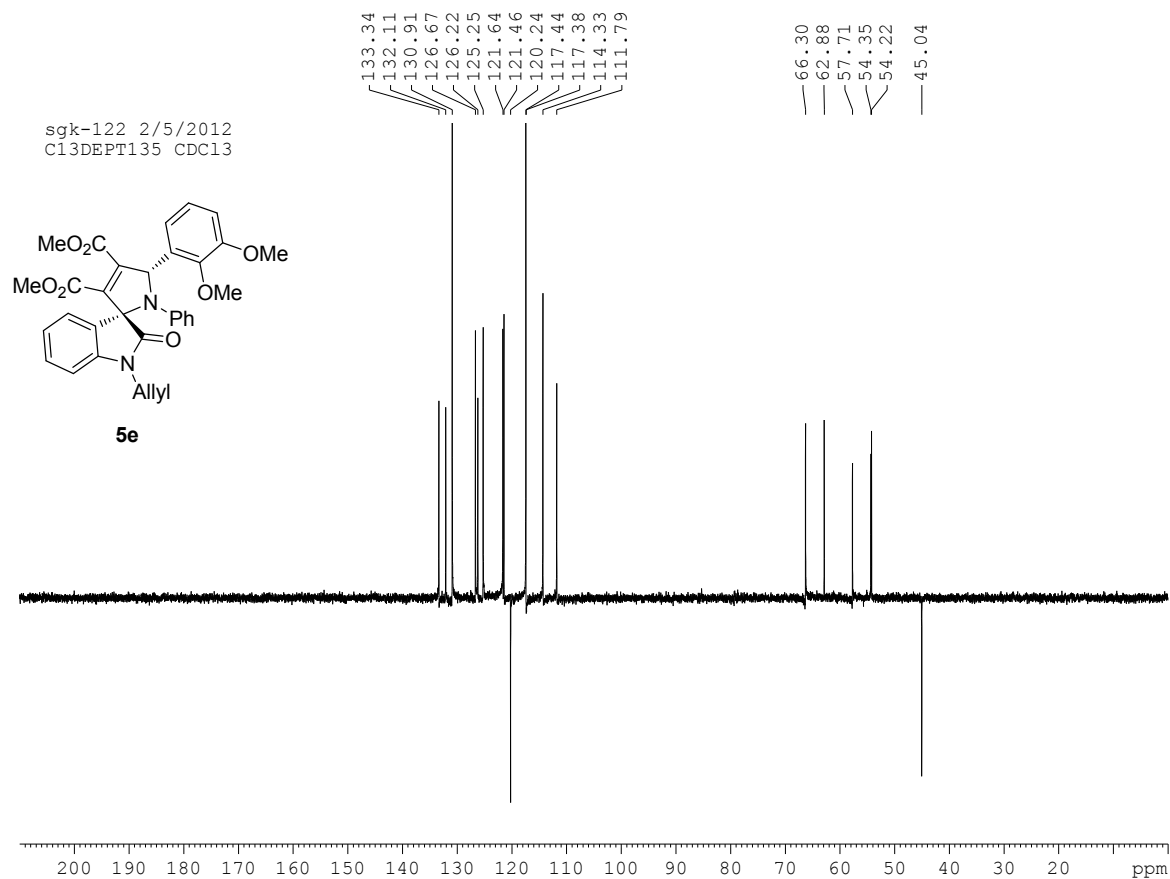
sgk-122 2/5/2012
C13CPD CDC13

¹³C NMR (100 MHz, CDCl₃)

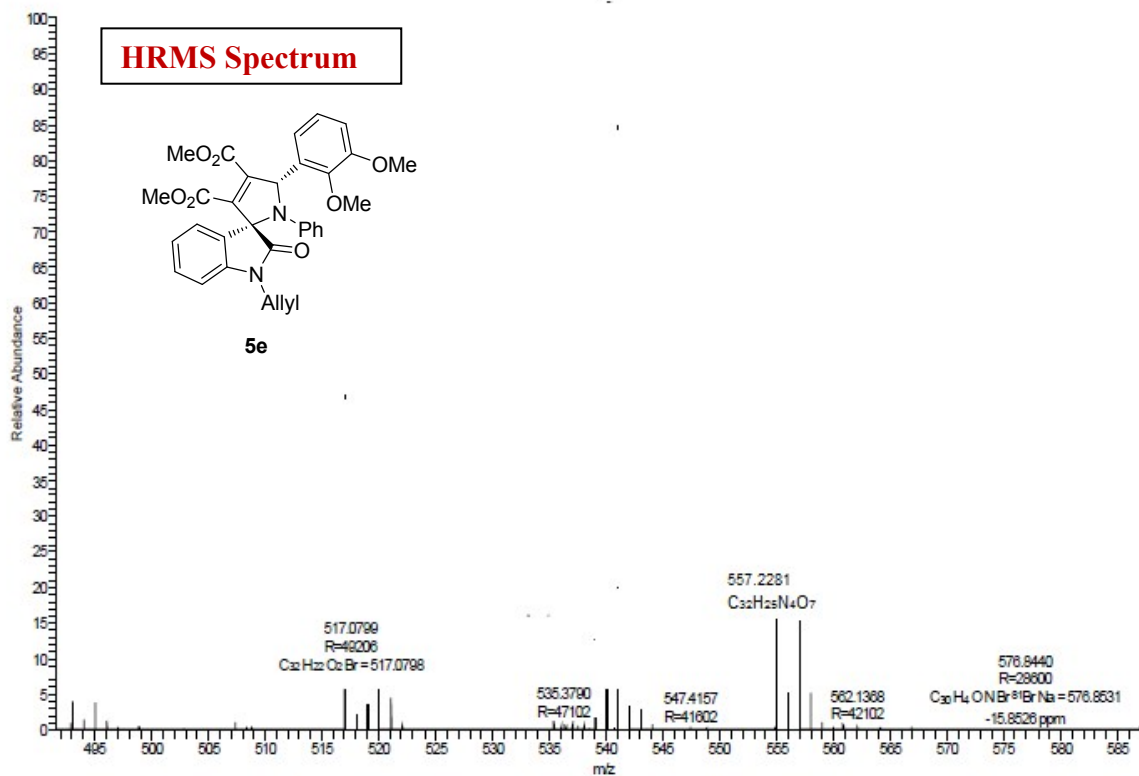


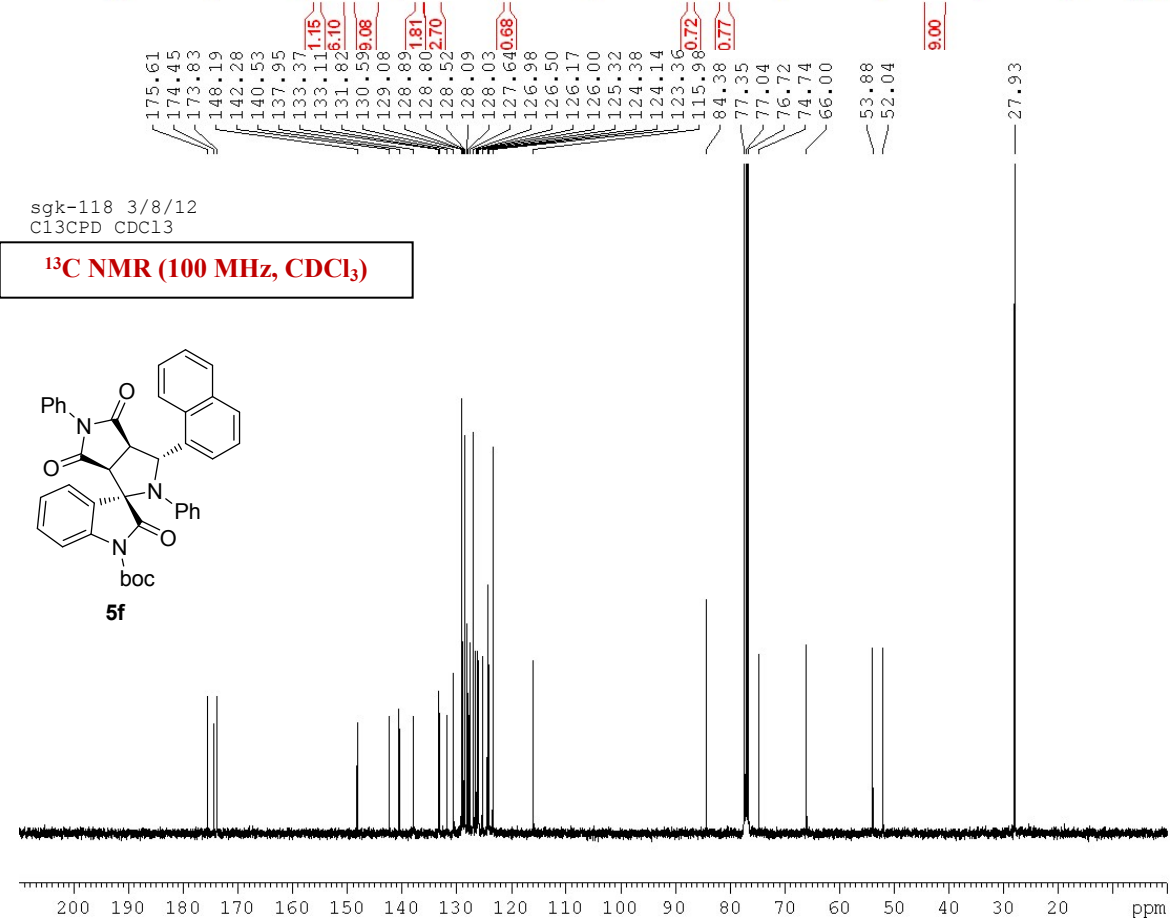
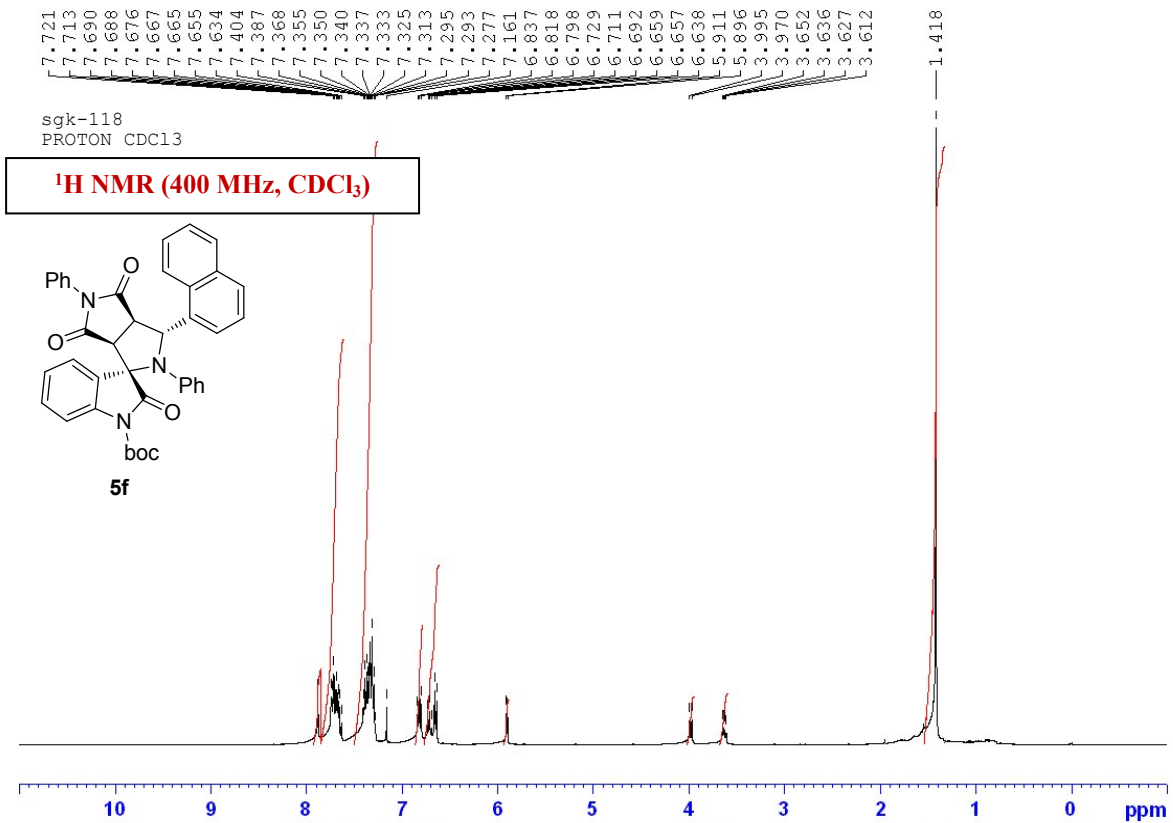
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 ppm

¹³C DEPT NMR (100 MHz, CDCl₃)



S3 #937 RT: 4.17 AM: 1 NL: 169E8
T: FTMS+pESI Full ms [100.00-1000.00]





sgk-118 3/8/12
C13DEPT135 CDC13

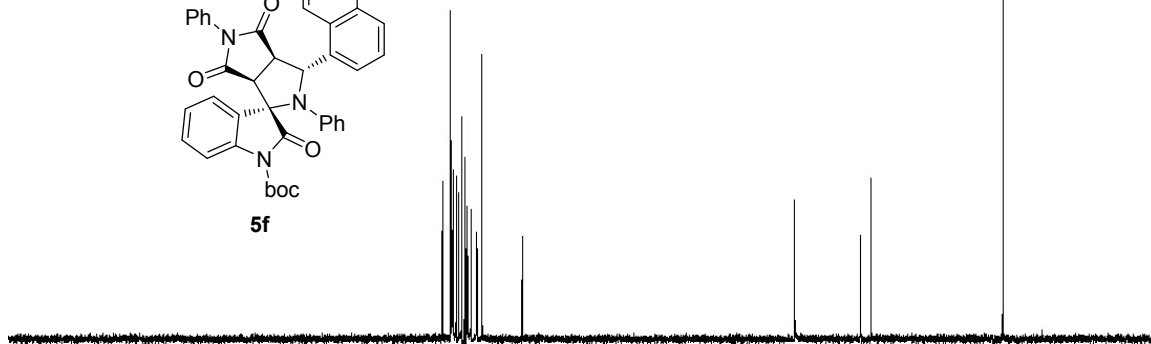
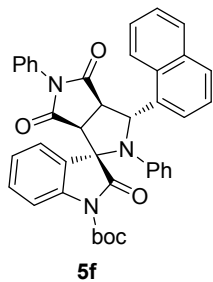
130.64
129.13
128.94
128.86
128.57
128.08
127.69
127.03
126.55
126.22
126.05
125.37
124.42
124.20
123.41
116.03

66.04

53.92
52.08

27.98

¹³C DEPT NMR (100 MHz, CDCl₃)

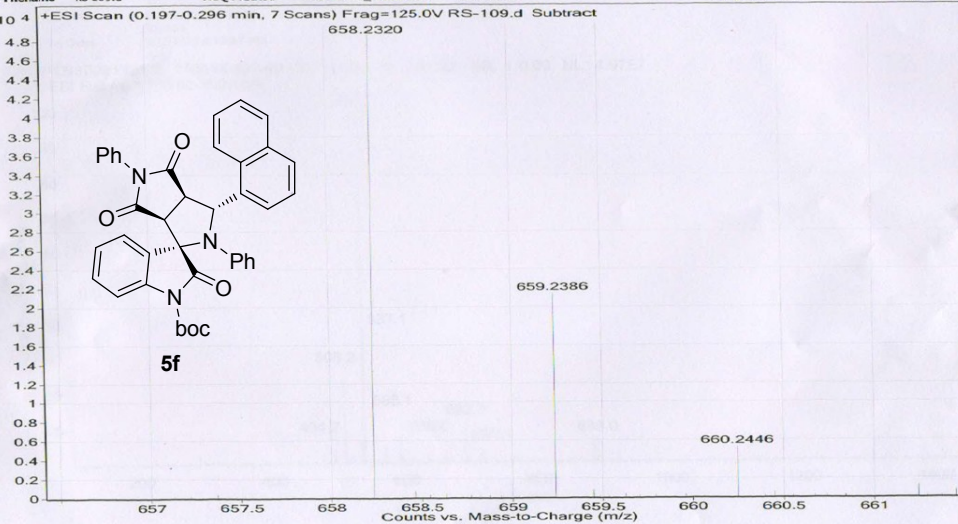
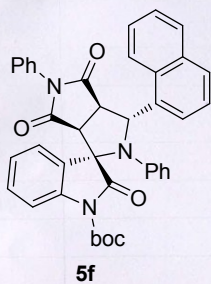


200 190 180 170 160 150 140 130 120 110 90 30 70 60 50 40 30 20 ppm

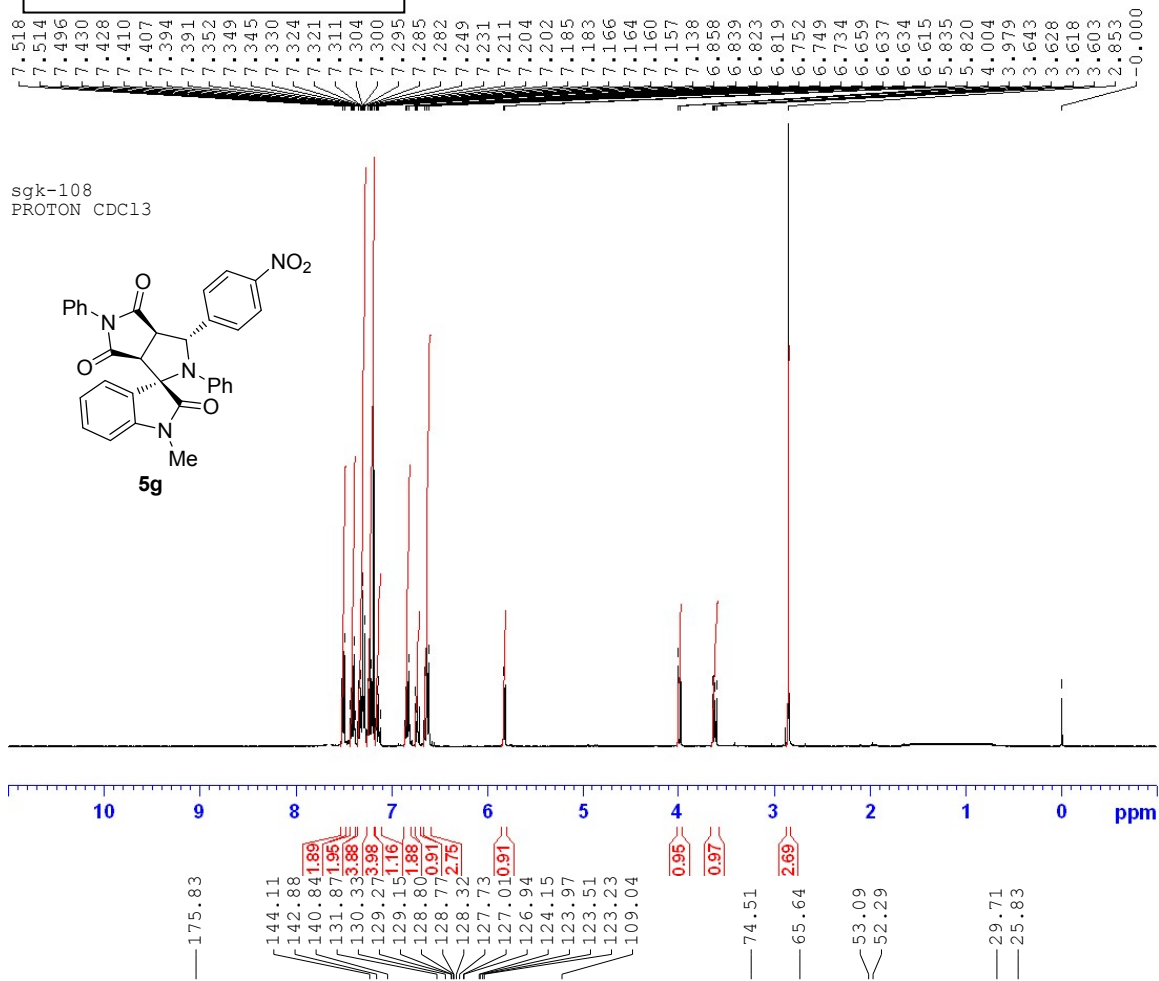
HRMS Spectrum

Sample Name	SANKALAN	Position	Vial 38	Instrument Name	Instrument 1	User Name
Inj Vol	1	InjPosition		SampleType	Sample	IRM Calibration Status
Data Filename	RS-109.d	ACQ Method	ISOCRATIC_GENERAL.m	Comment		Acquired Time

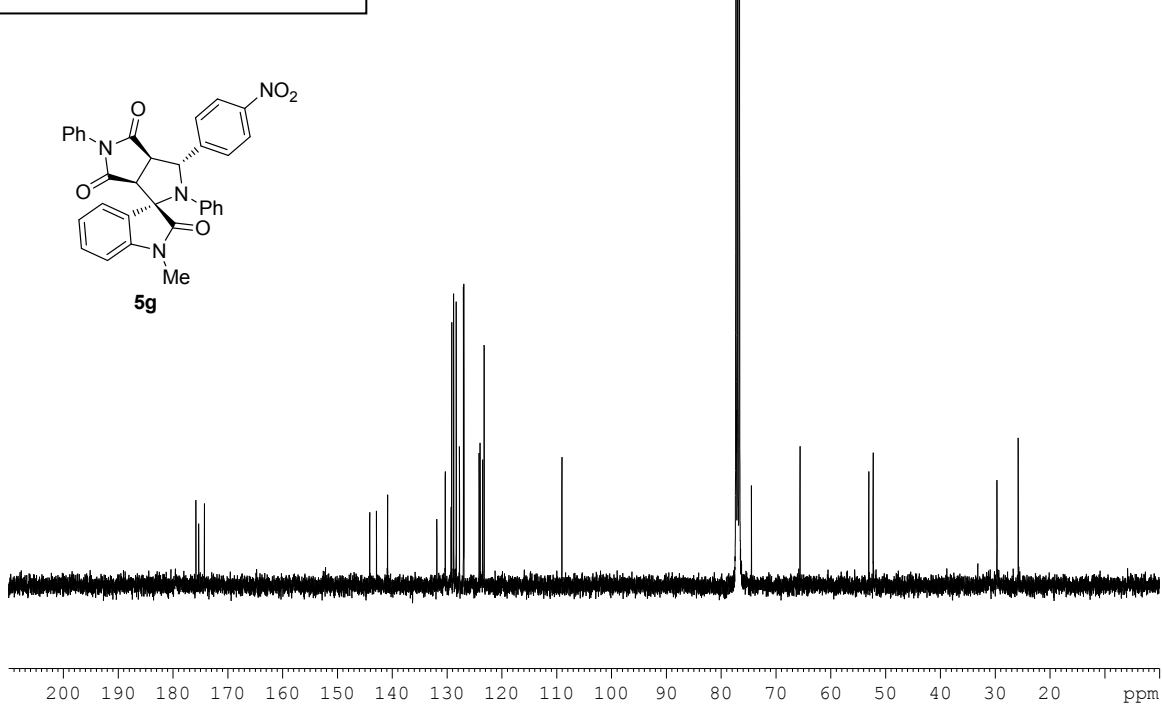
x10⁴ +ESI Scan (0.197-0.296 min, 7 Scans) Frag=125.0V RS-109.d Subtract
658.2320



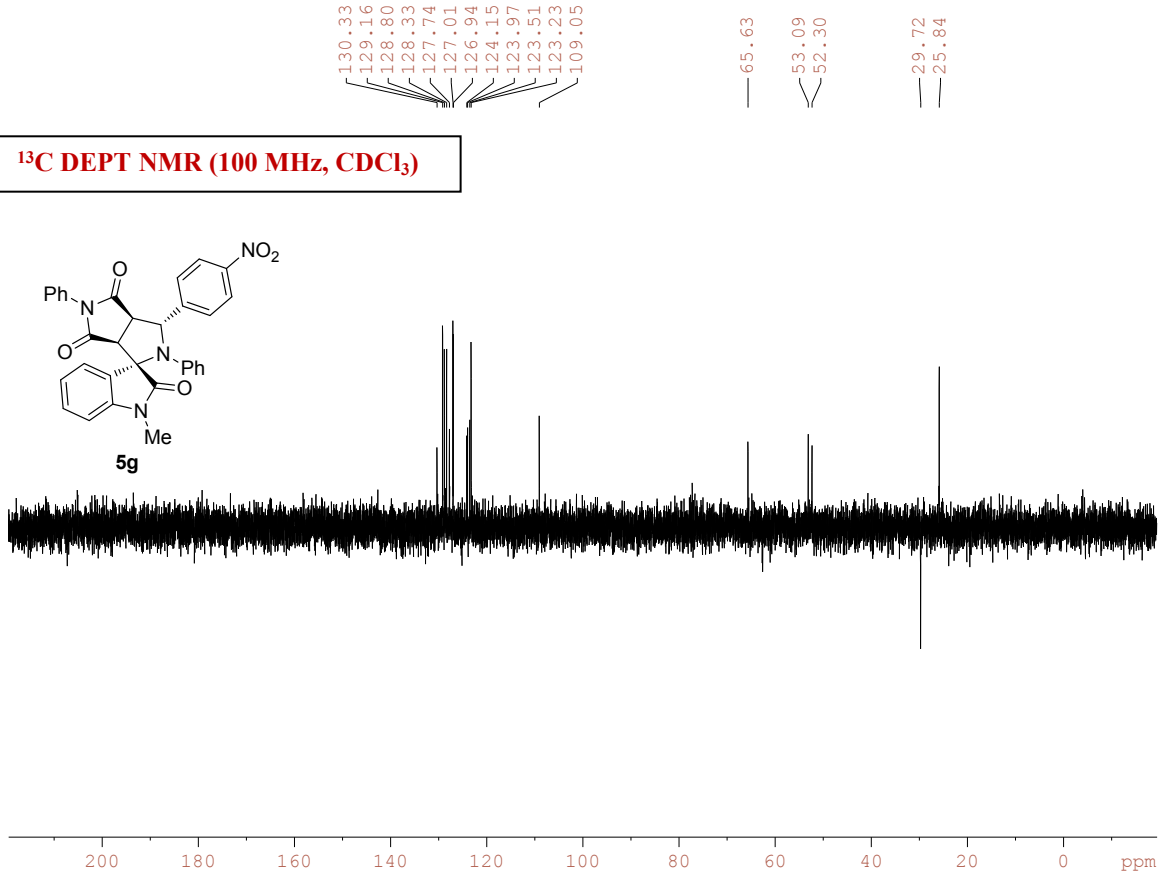
¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)

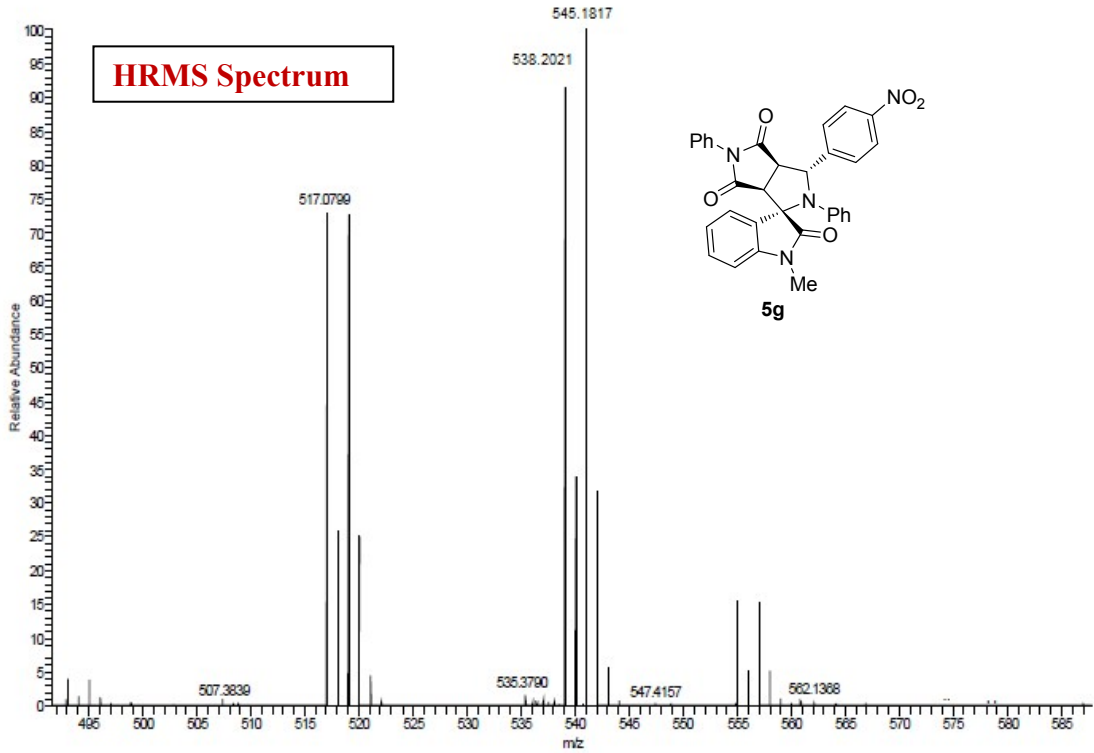


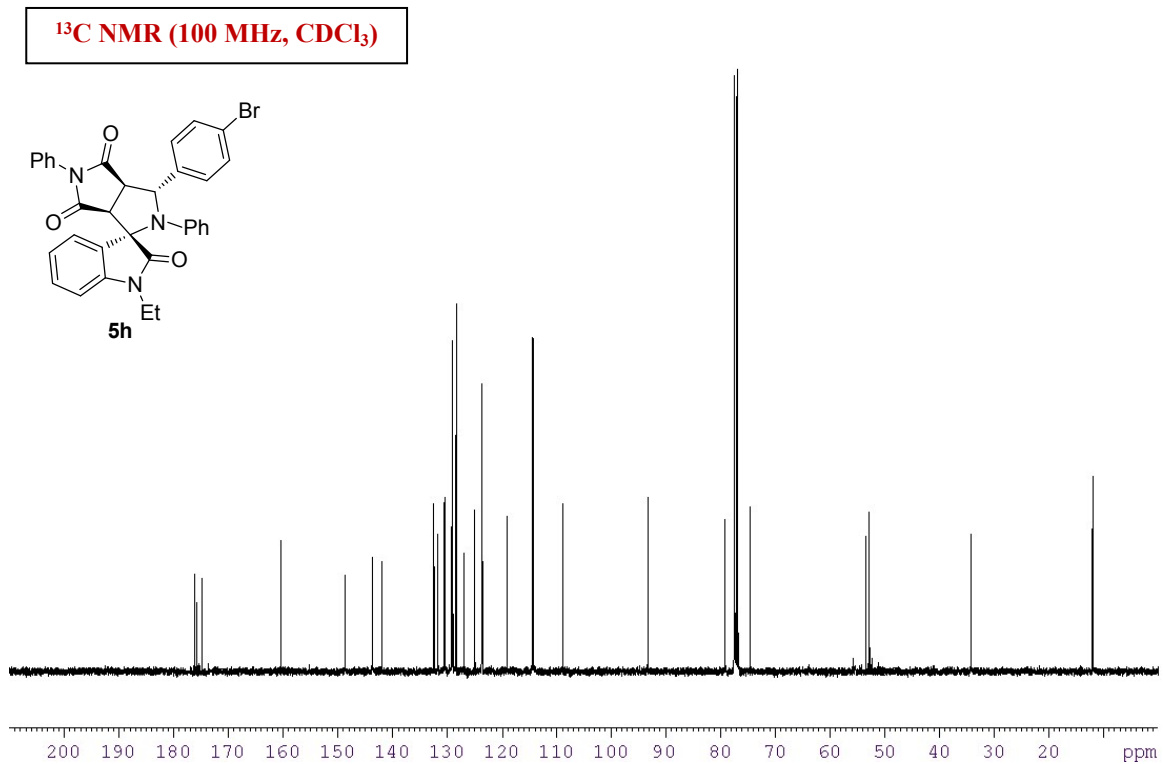
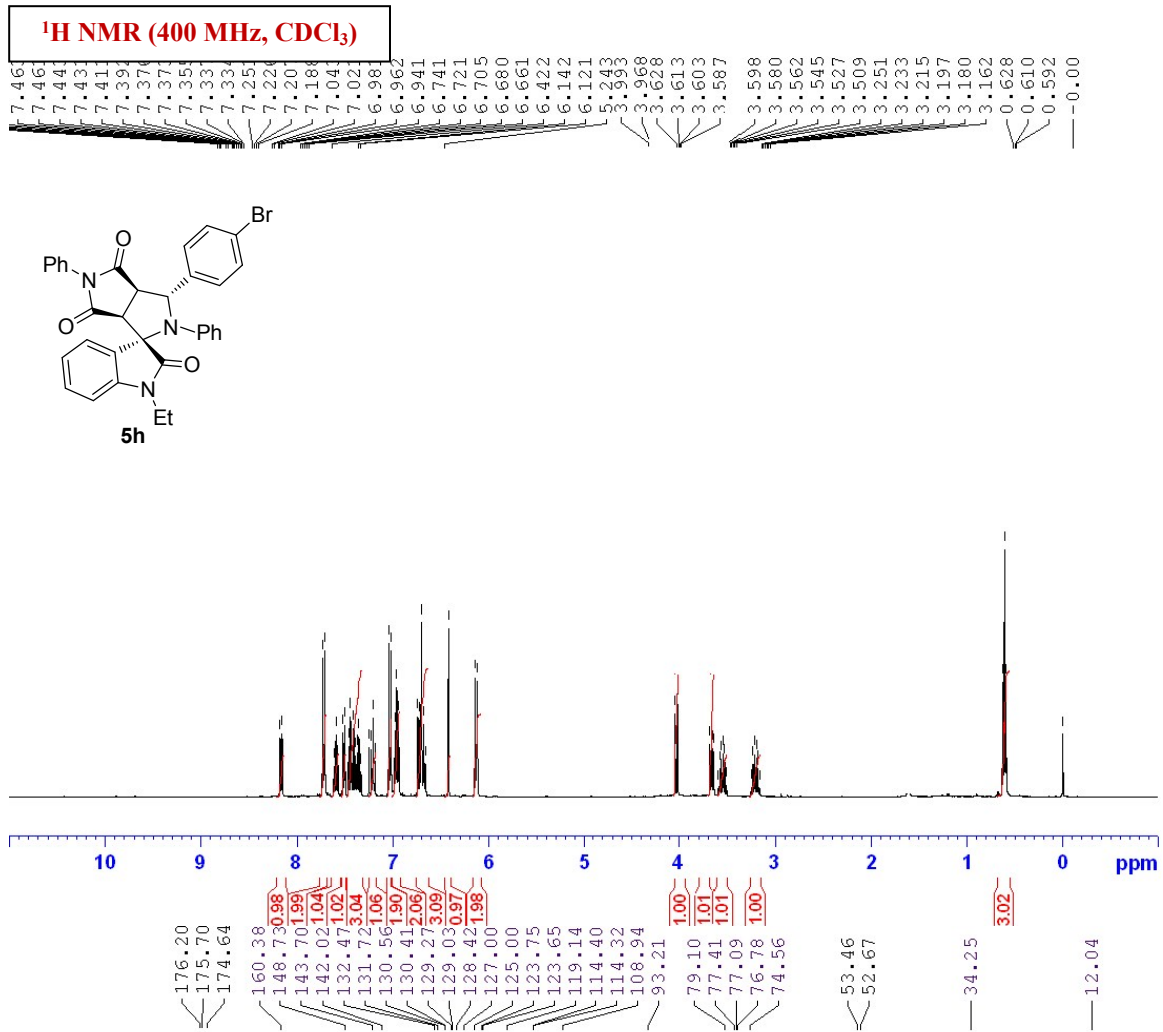
¹³C DEPT NMR (100 MHz, CDCl₃)



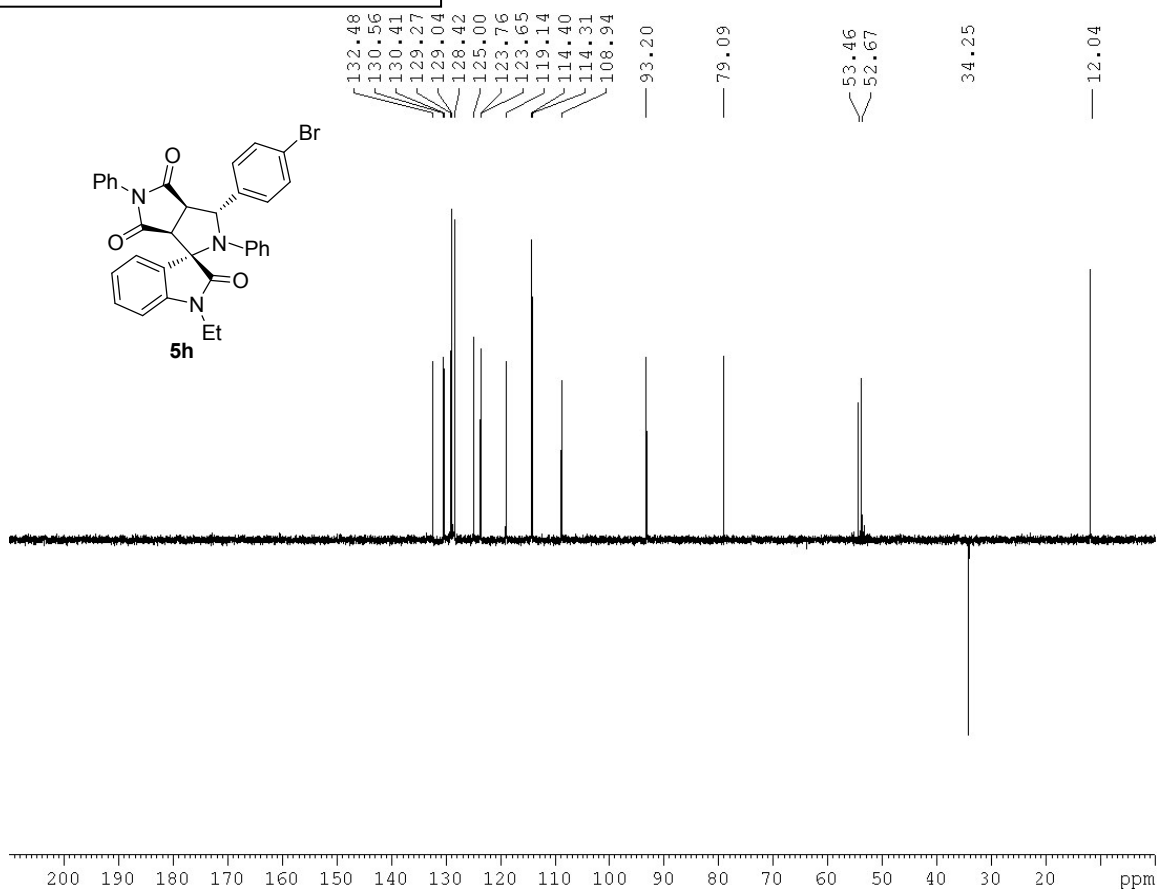
S3#937 RT: 4.17 AM: 1 NL: 1.69EB
T: FTMS+p ESI Full ms [100.00-1000.00]

HRMS Spectrum





¹³C DEPT NMR (100 MHz, CDCl₃)

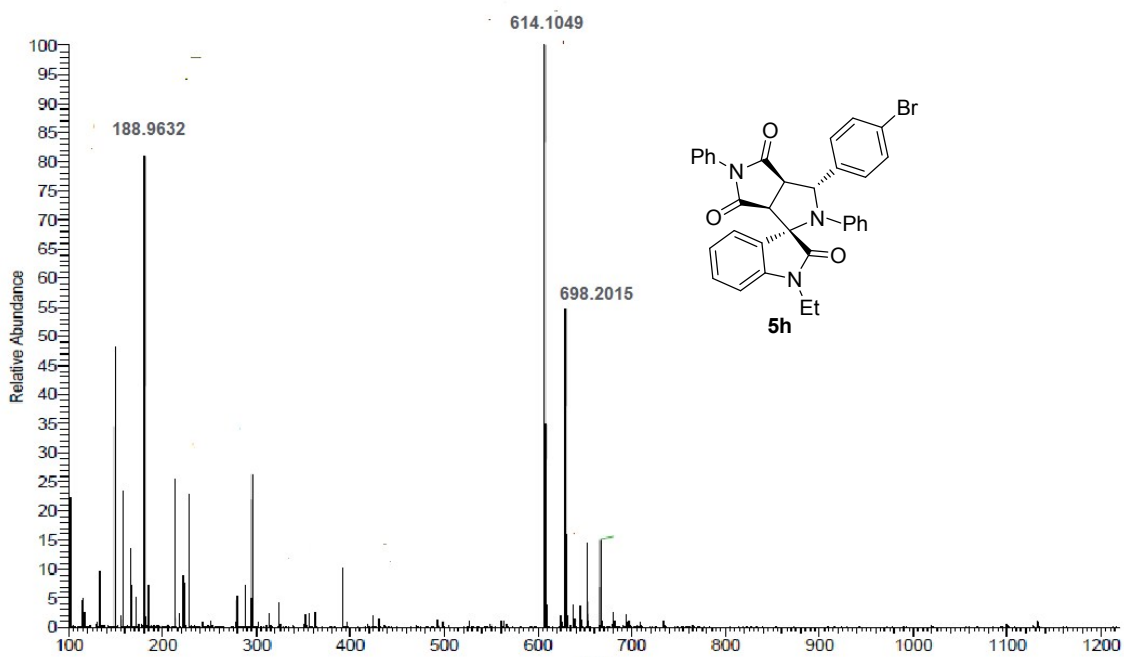


HRMS Spectrum

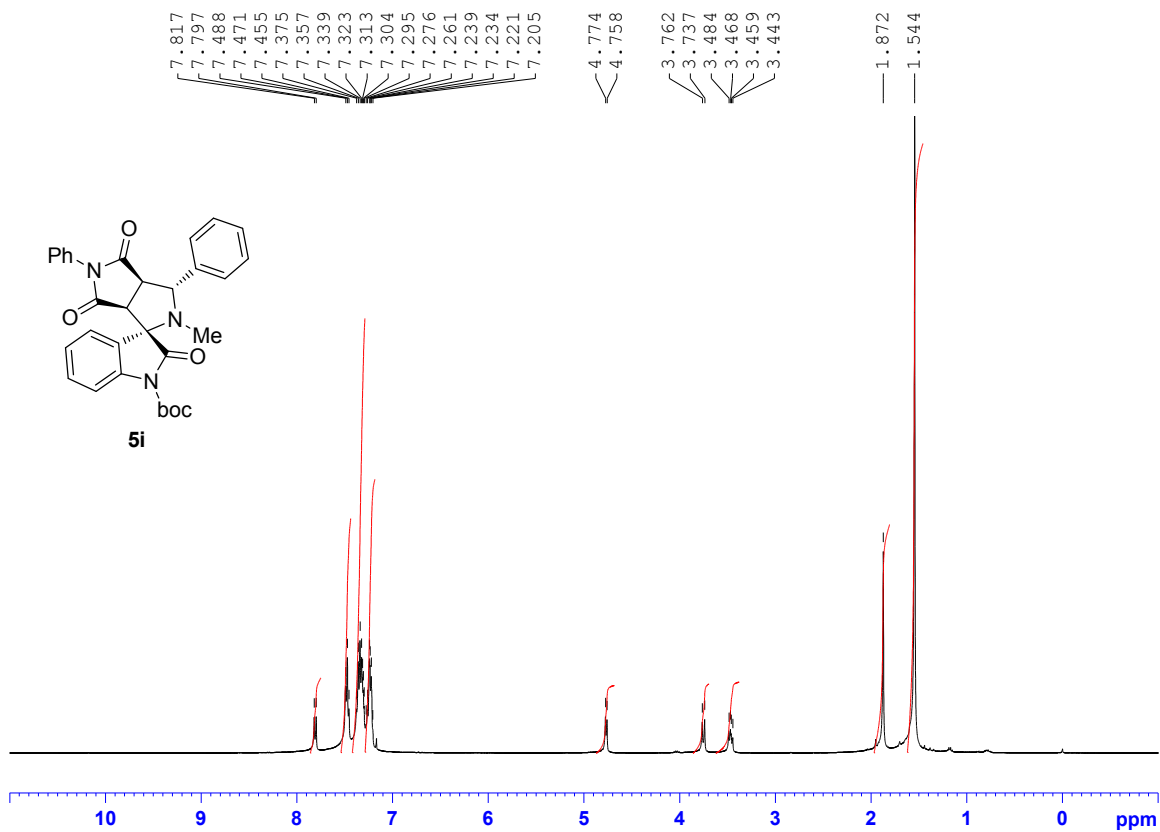
C:\xcalibur\...FEB-2014\SMGK-299

25-02-2014 11:29:36

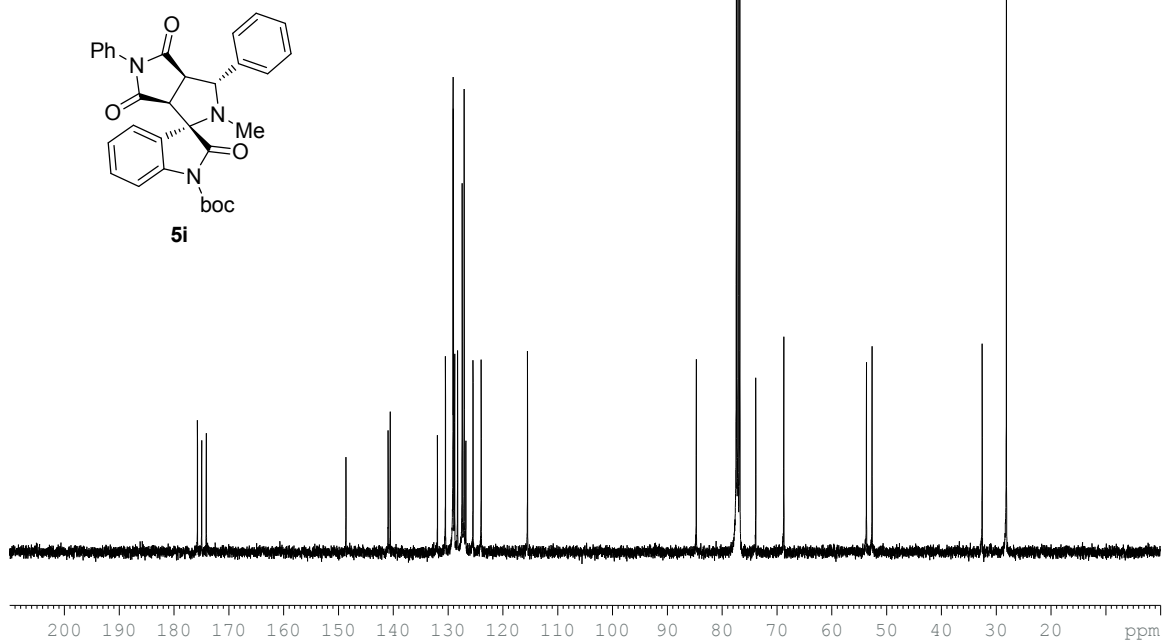
SMGK-299 #63 RT: 0.94 AV: 1 NL: 1.00E6
T: FTMS (1,1) + p ESI Full lock ms [100.00-2000.00]



¹H NMR (400 MHz, CDCl₃)

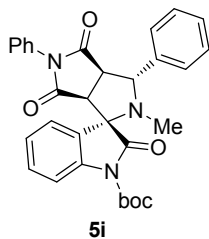
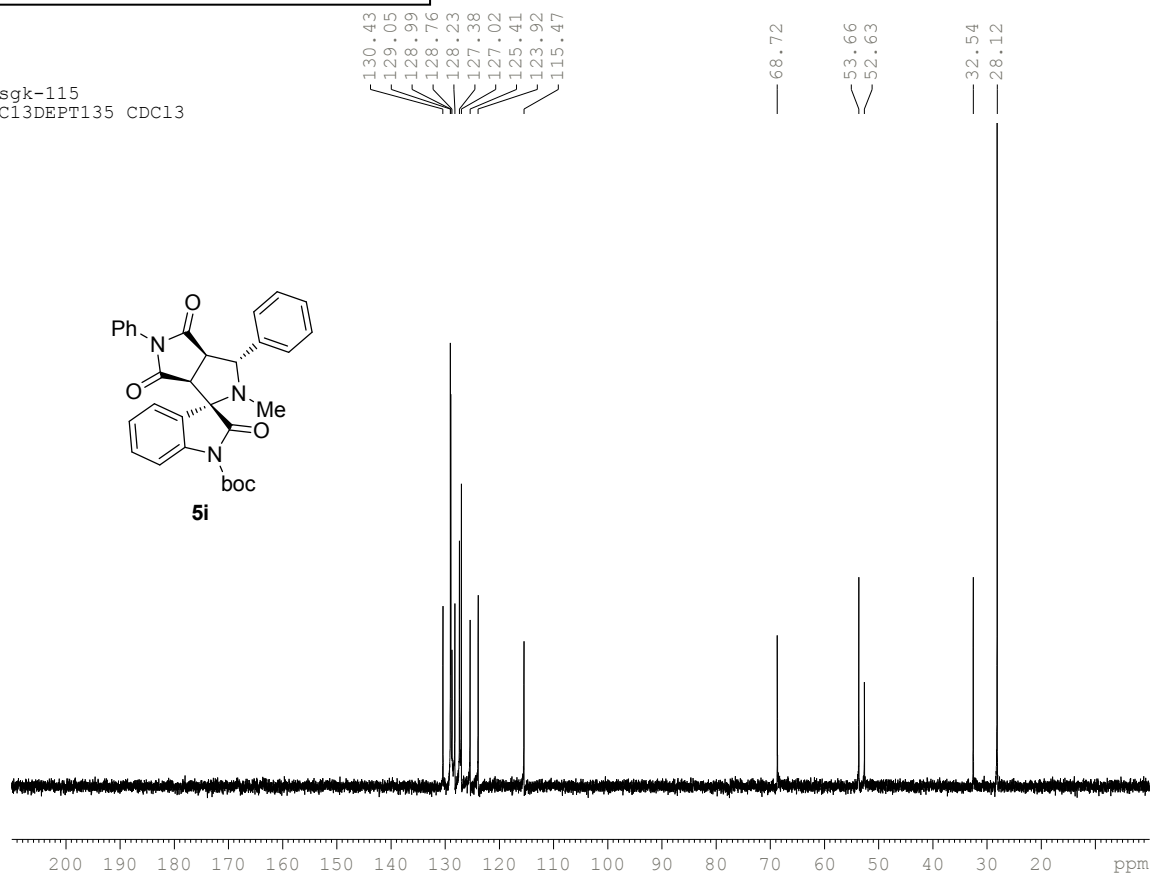


¹³C NMR (100 MHz, CDCl₃)

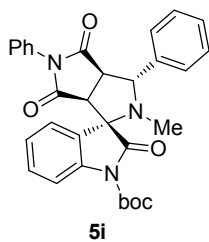
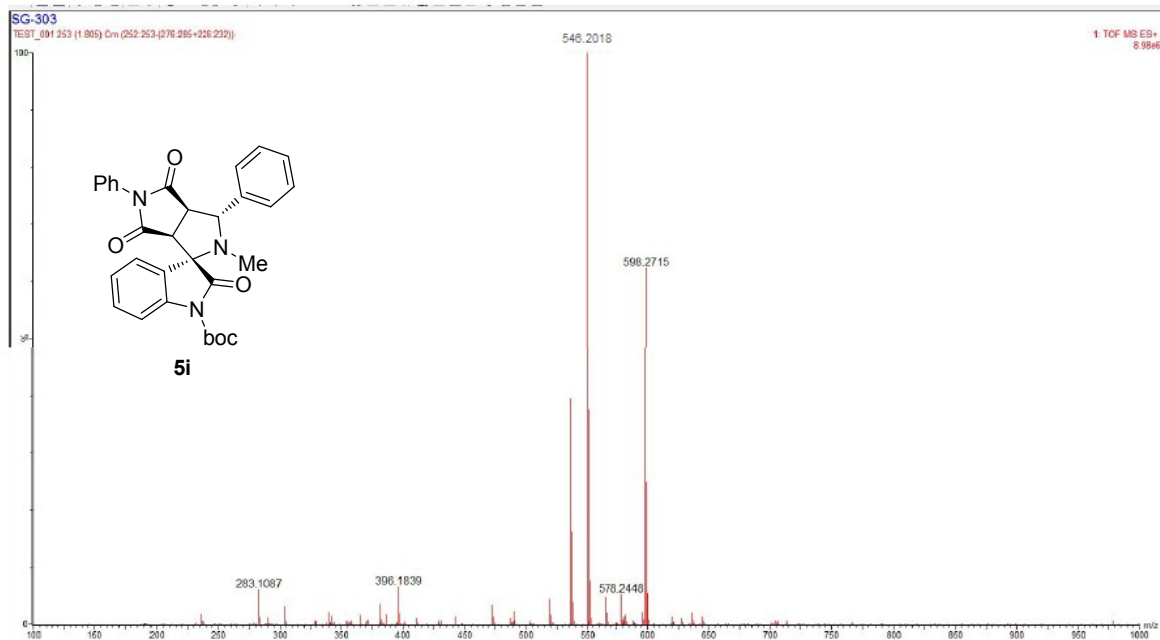


¹³C DEPT NMR (100 MHz, CDCl₃)

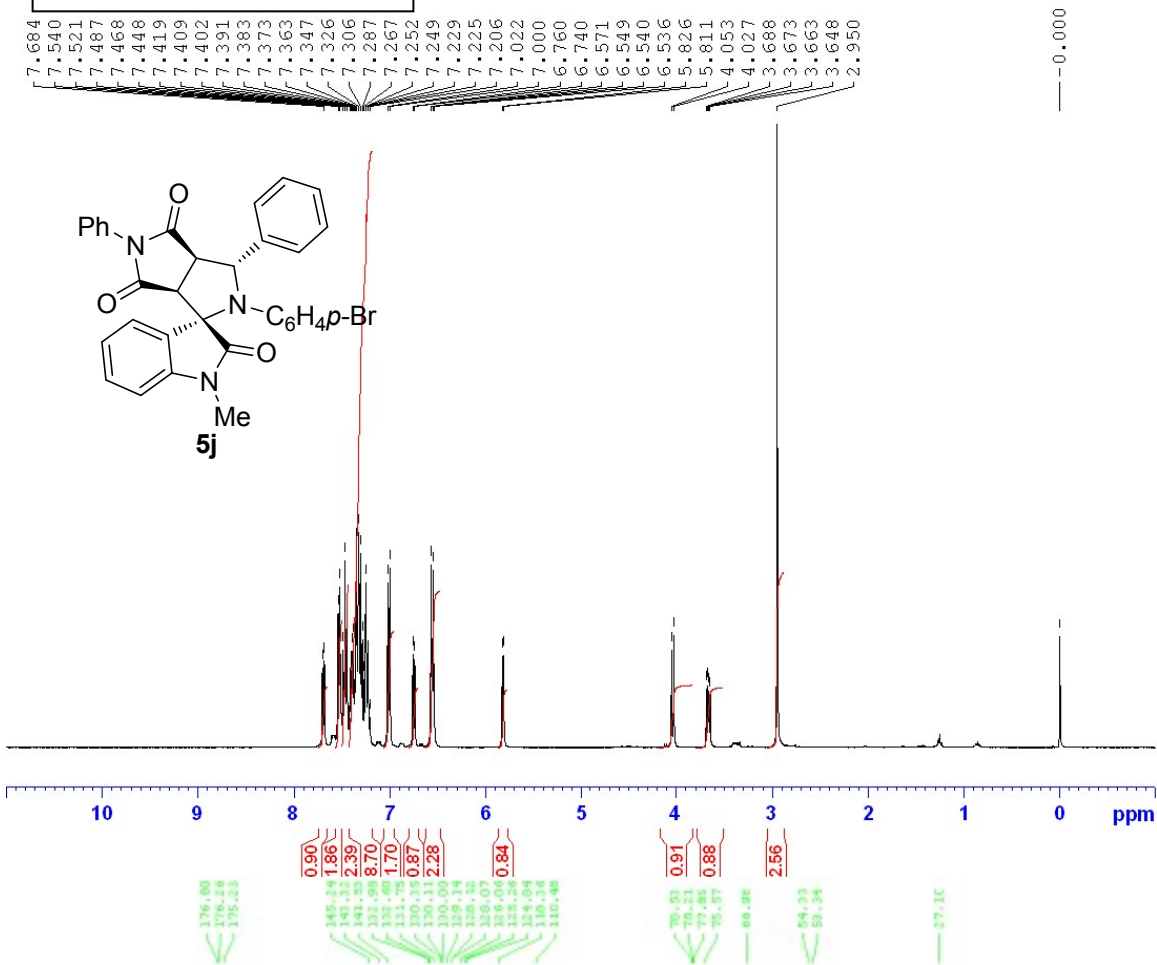
sgk-115
C13DEPT135 CDCl3



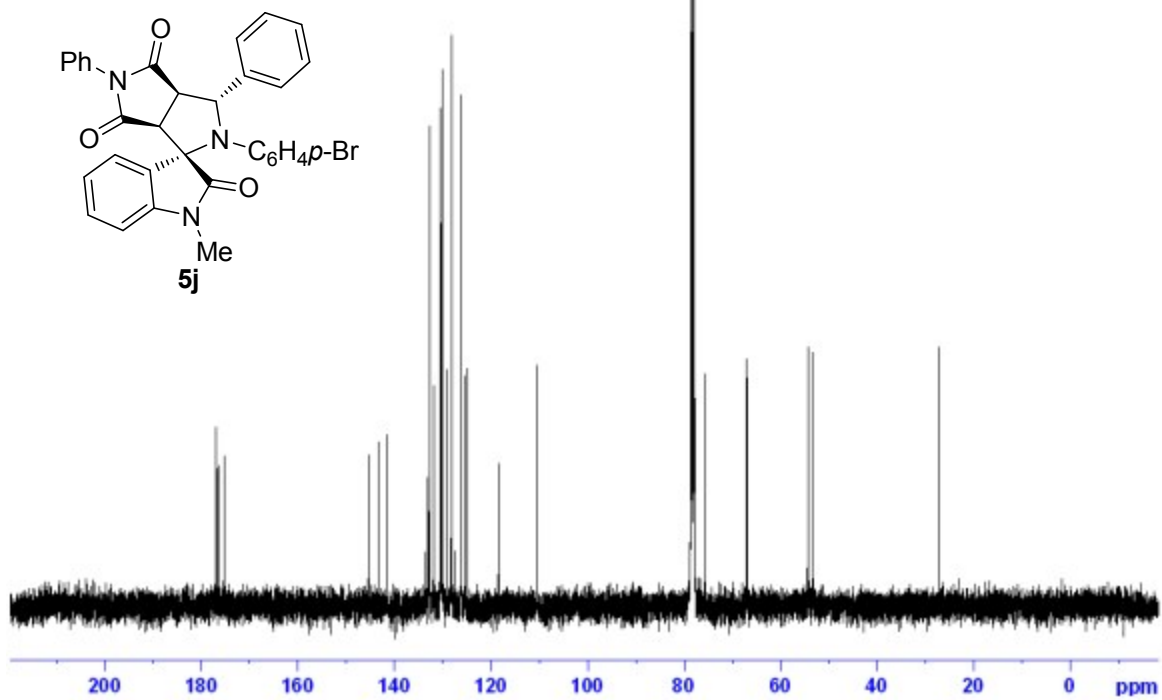
HRMS Spectrum



¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)



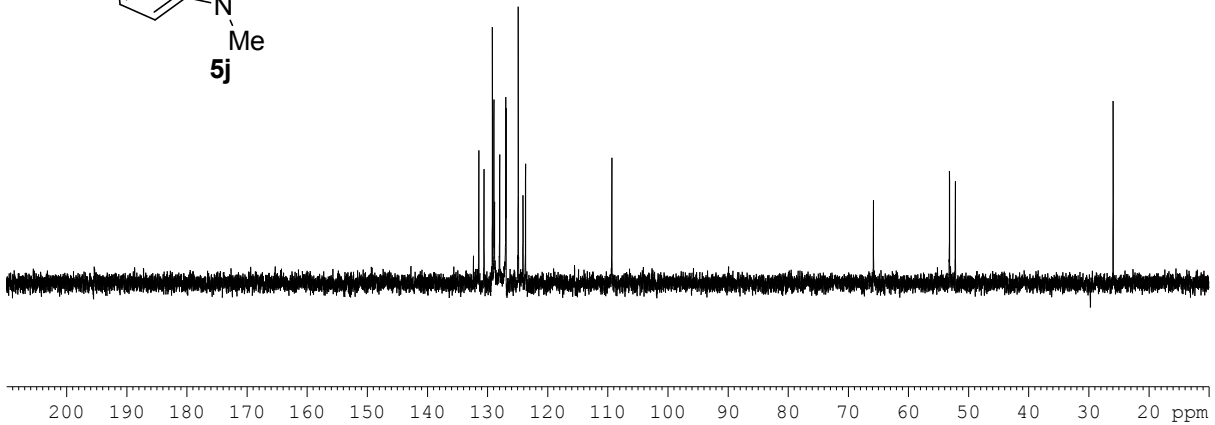
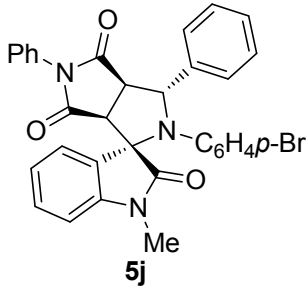
¹³C DEPT NMR (100 MHz, CDCl₃)

132.34
131.45
130.59
129.19
128.95
128.84
127.98
126.97
126.91
124.90
124.11
123.68
109.32

65.81

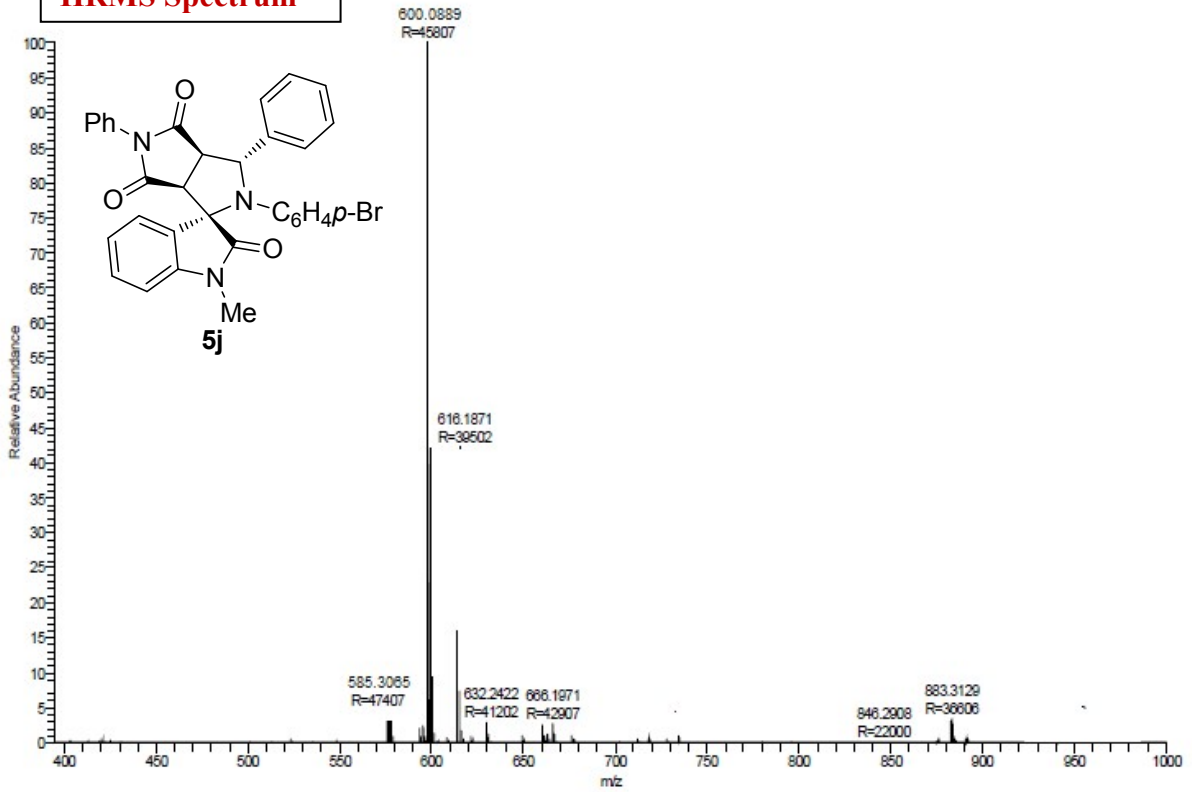
53.16
52.18

29.72
25.95



S4#457 RT: 2.03 AV: 1 NL: 2.96E8
T: FTMS+pESI Full ms [100.00-1000.00]

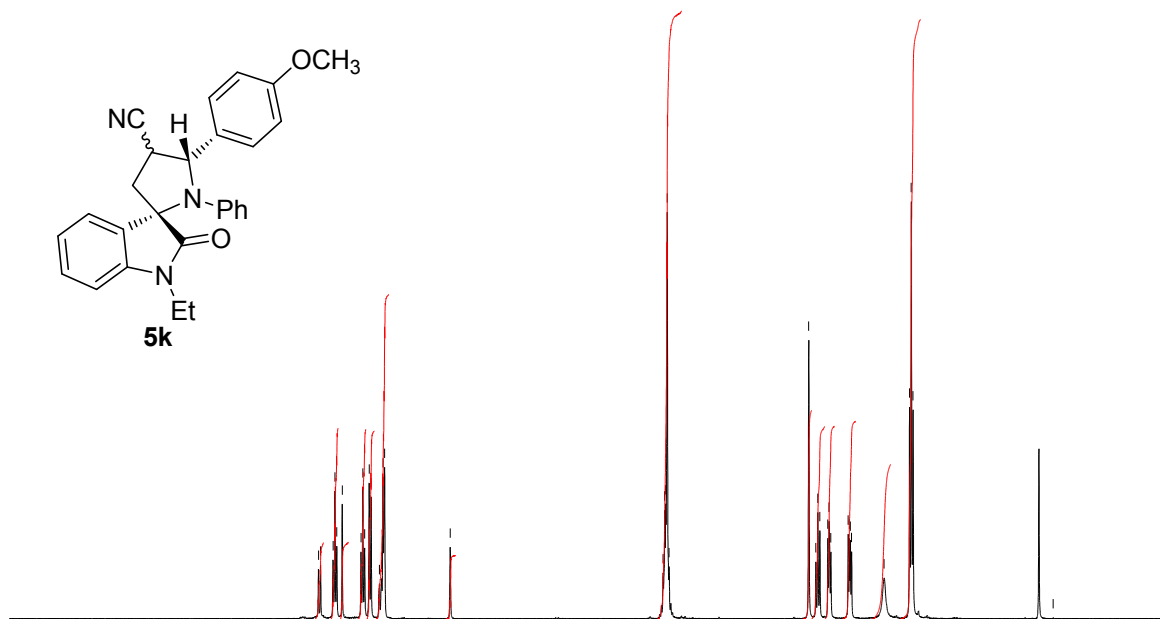
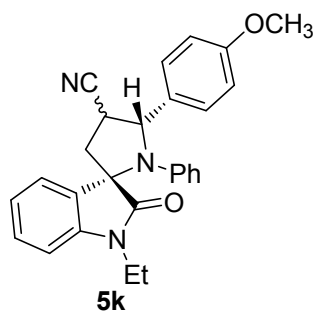
HRMS Spectrum



¹H NMR (400 MHz, CDCl₃)

sgk-307
PROTON CDCl₃

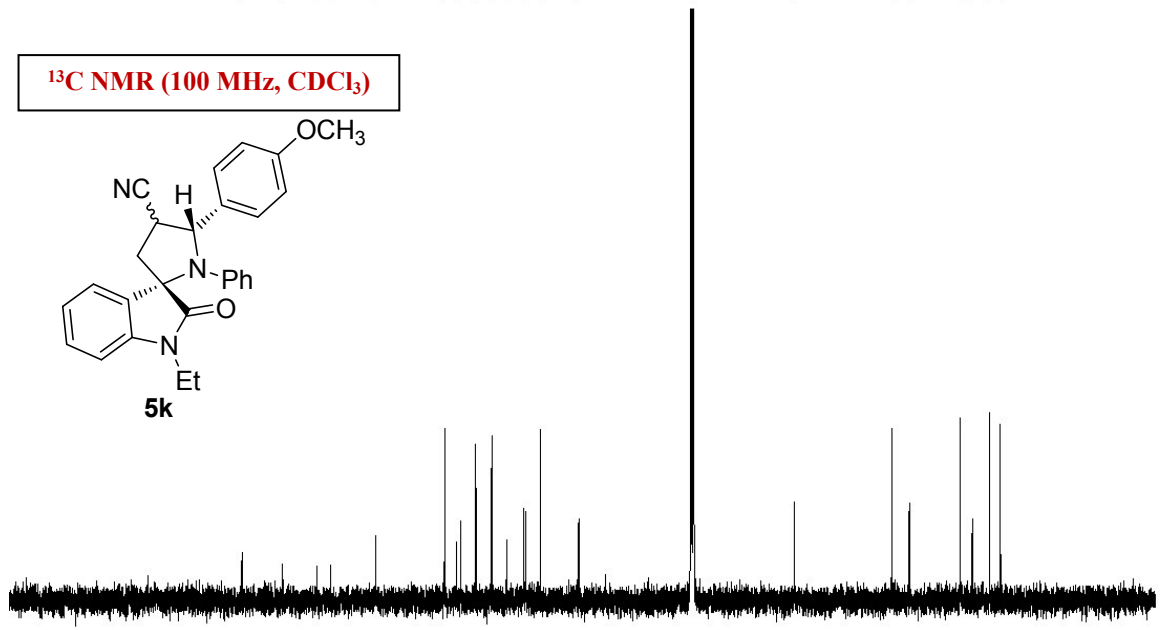
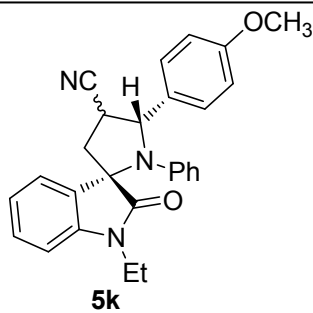
7.779, 7.758, 7.628, 7.608, 7.589, 7.532, 7.336, 7.317, 7.298, 7.251, 7.231, 7.144, 7.121, 7.108, 7.090, 6.406, 4.188, 4.170, 4.161, 4.145, 4.126, 2.670, 2.595, 2.575, 2.554, 2.470, 2.457, 2.439, 2.259, 2.246, 2.236, 2.223, 1.883, 1.618, 1.600, 1.582, 0.121



10 9 8 7 6 5 4 3 2 1 0 ppm

171.06, 162.70, 155.35, 152.58, 143.23, 128.79, 126.29, 125.55, 122.33, 119.02, 115.91, 112.32, 111.99, 108.88, 100.89, 77.36, 77.05, 76.73, 55.77, 35.42, 31.80, 21.21, 18.69, 15.12, 12.82

¹³C NMR (100 MHz, CDCl₃)

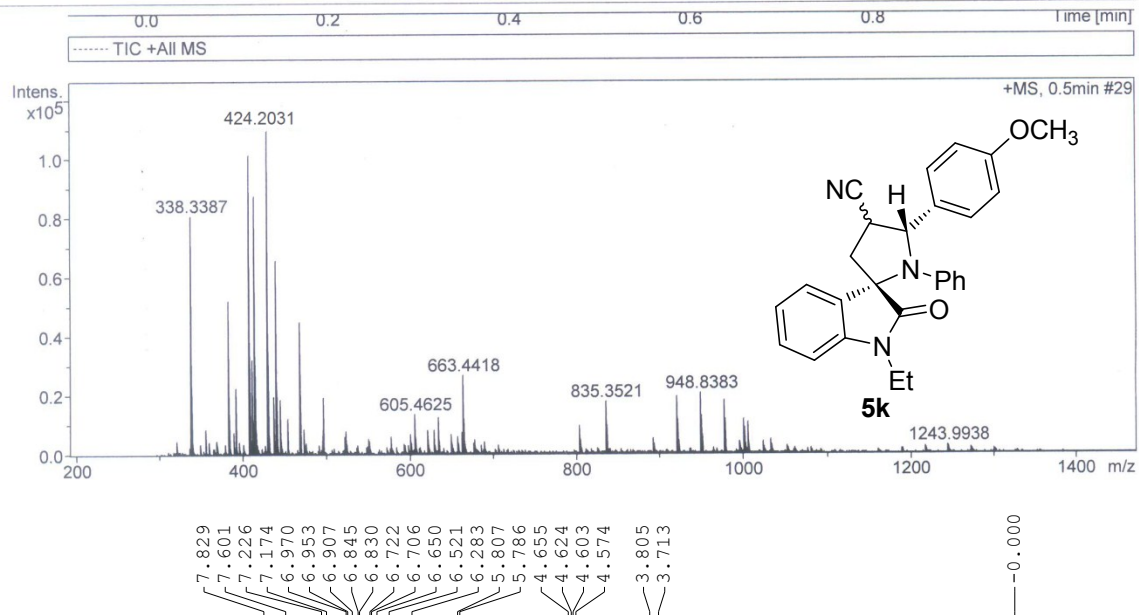


200 180 160 140 120 100 80 60 40 20 0 ppm

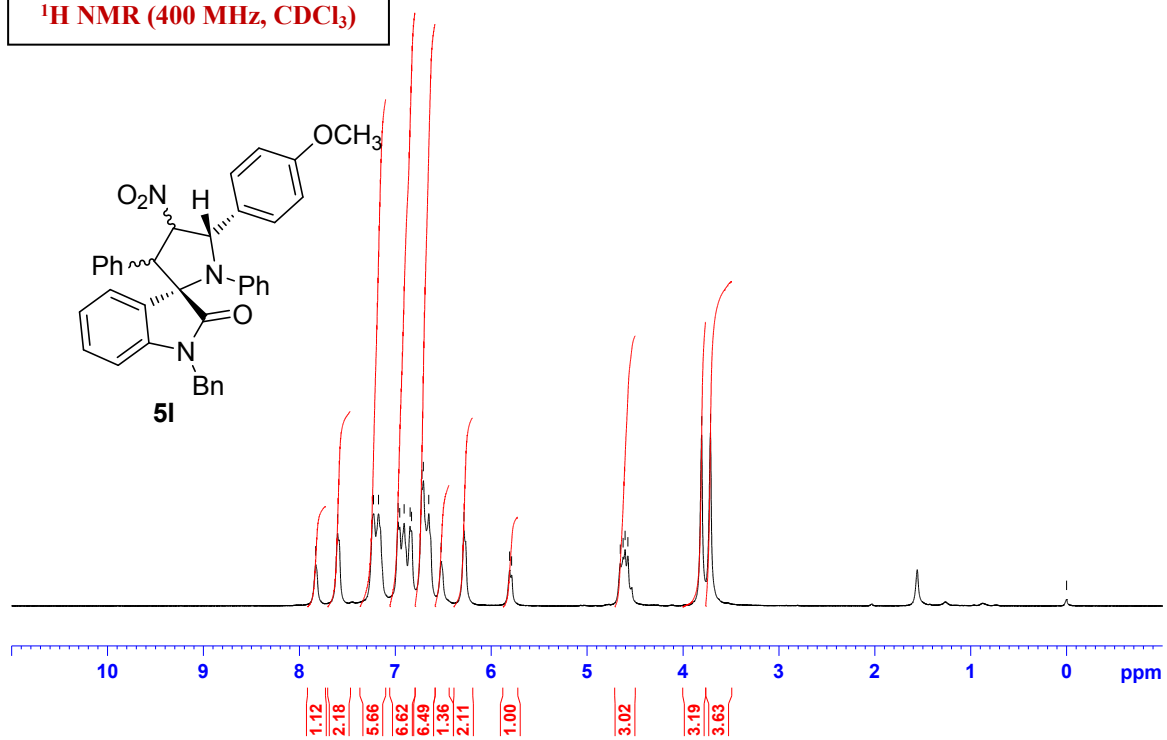
HRMS Spectrum

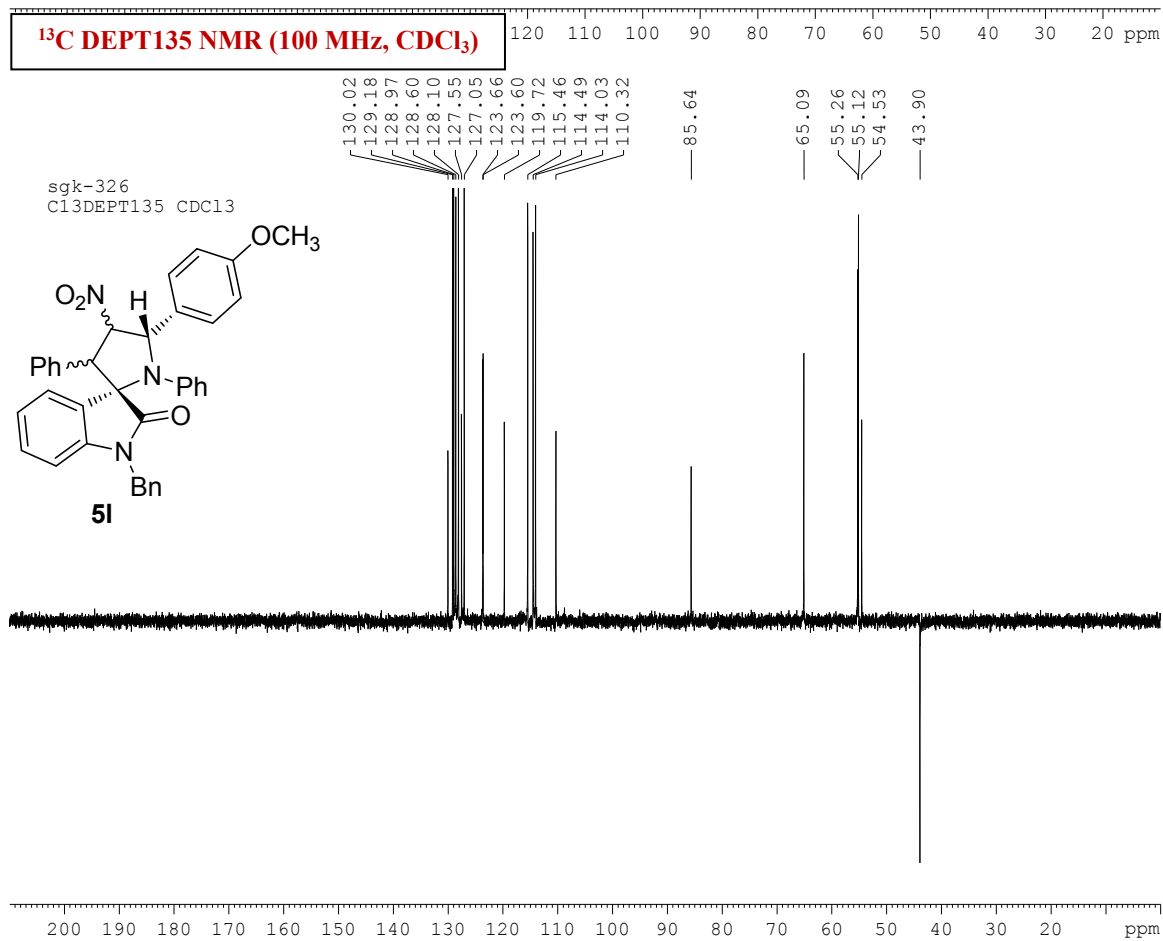
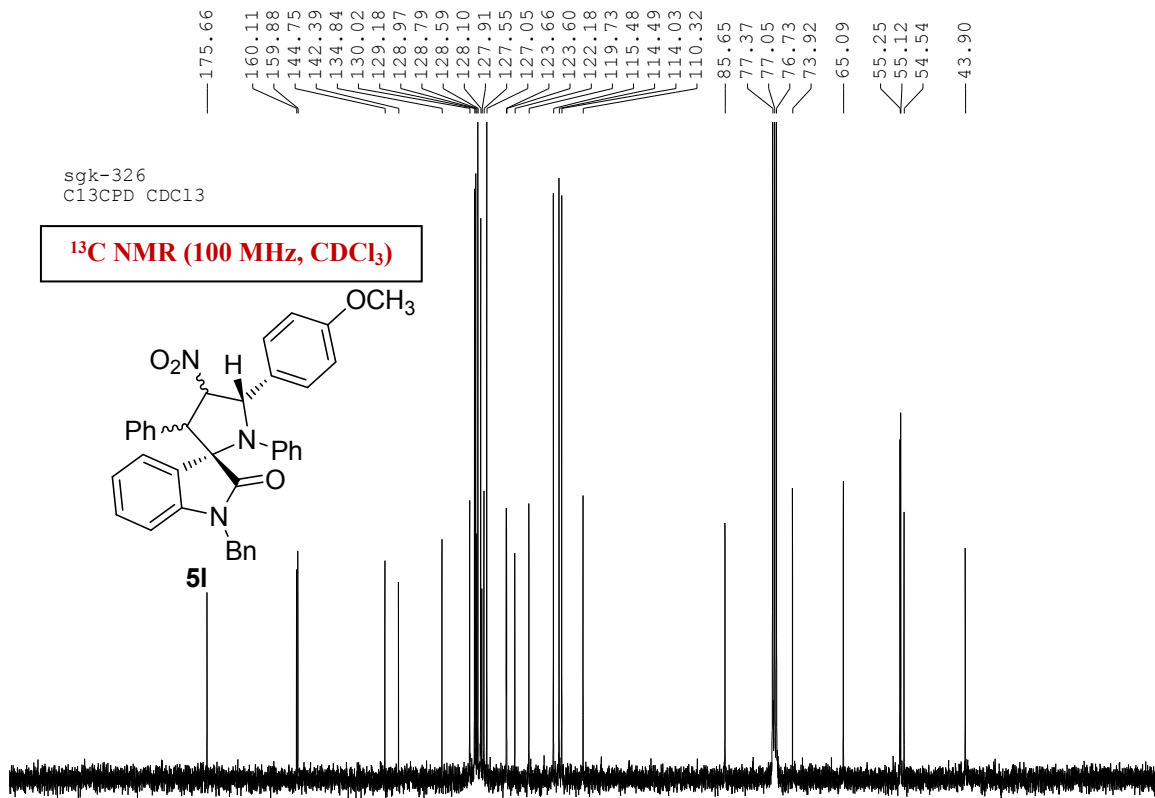
Acquisition Parameter

Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	3000 m/z	Set Collision Cell RF	600.0 Vpp	Set Divert Valve	Source



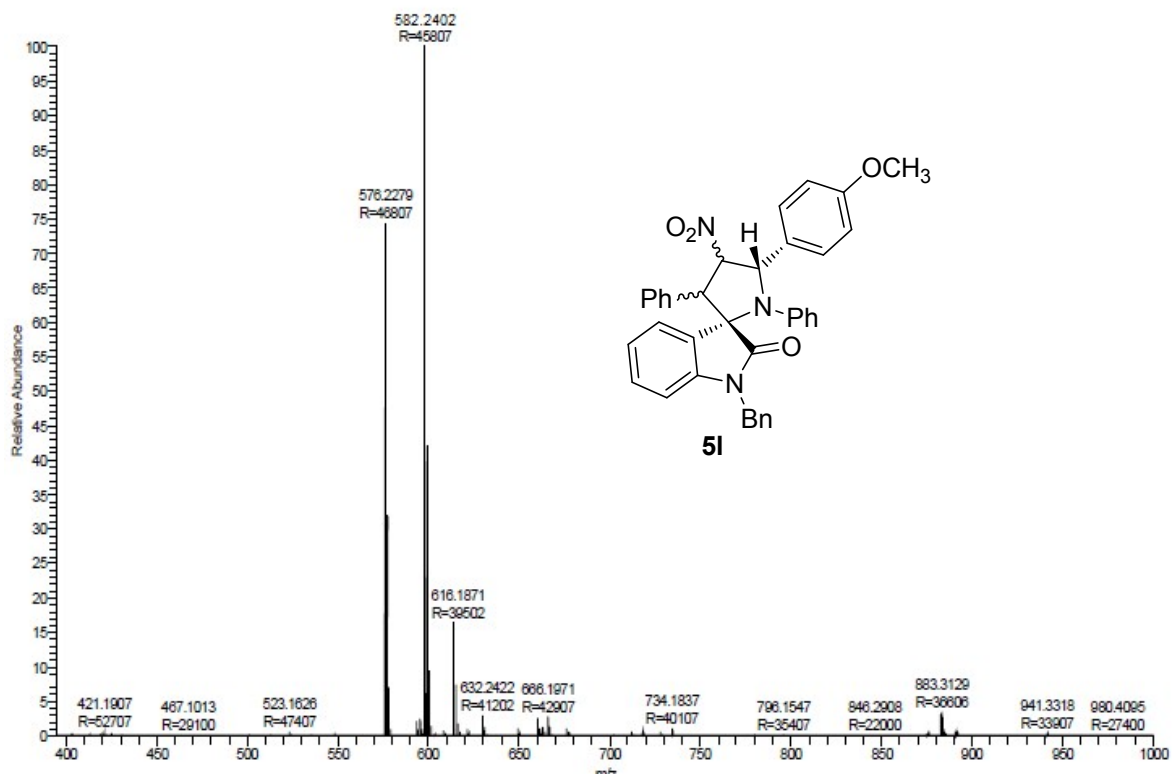
¹H NMR (400 MHz, CDCl₃)



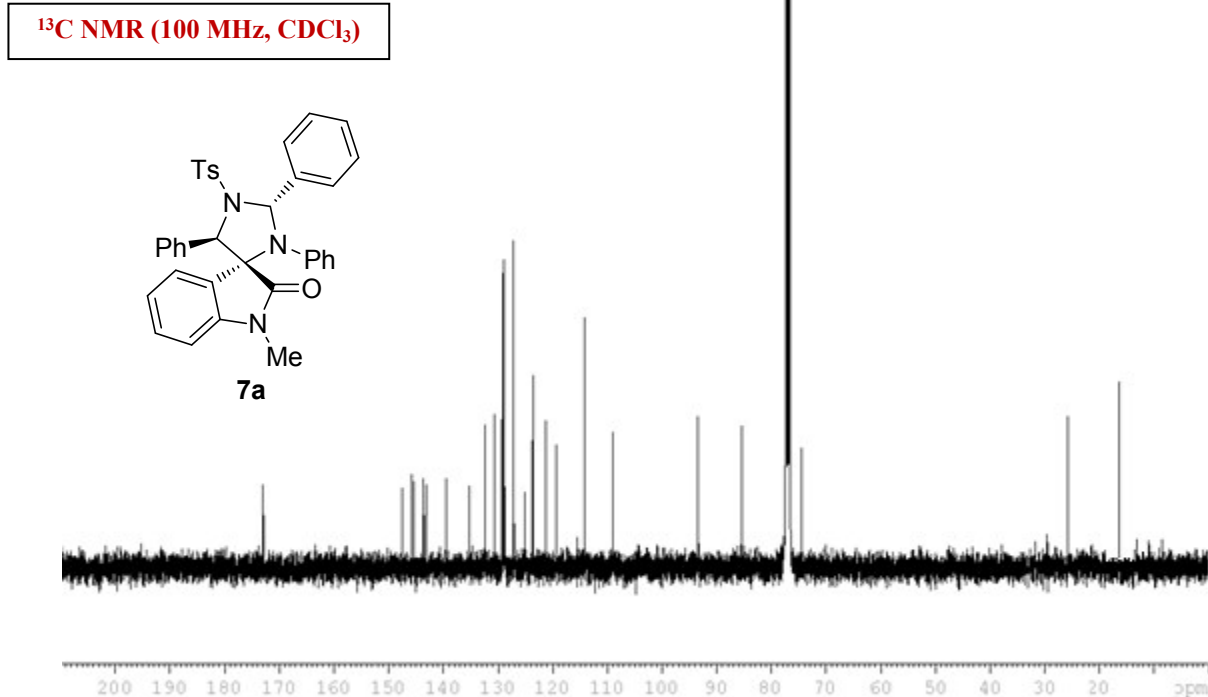
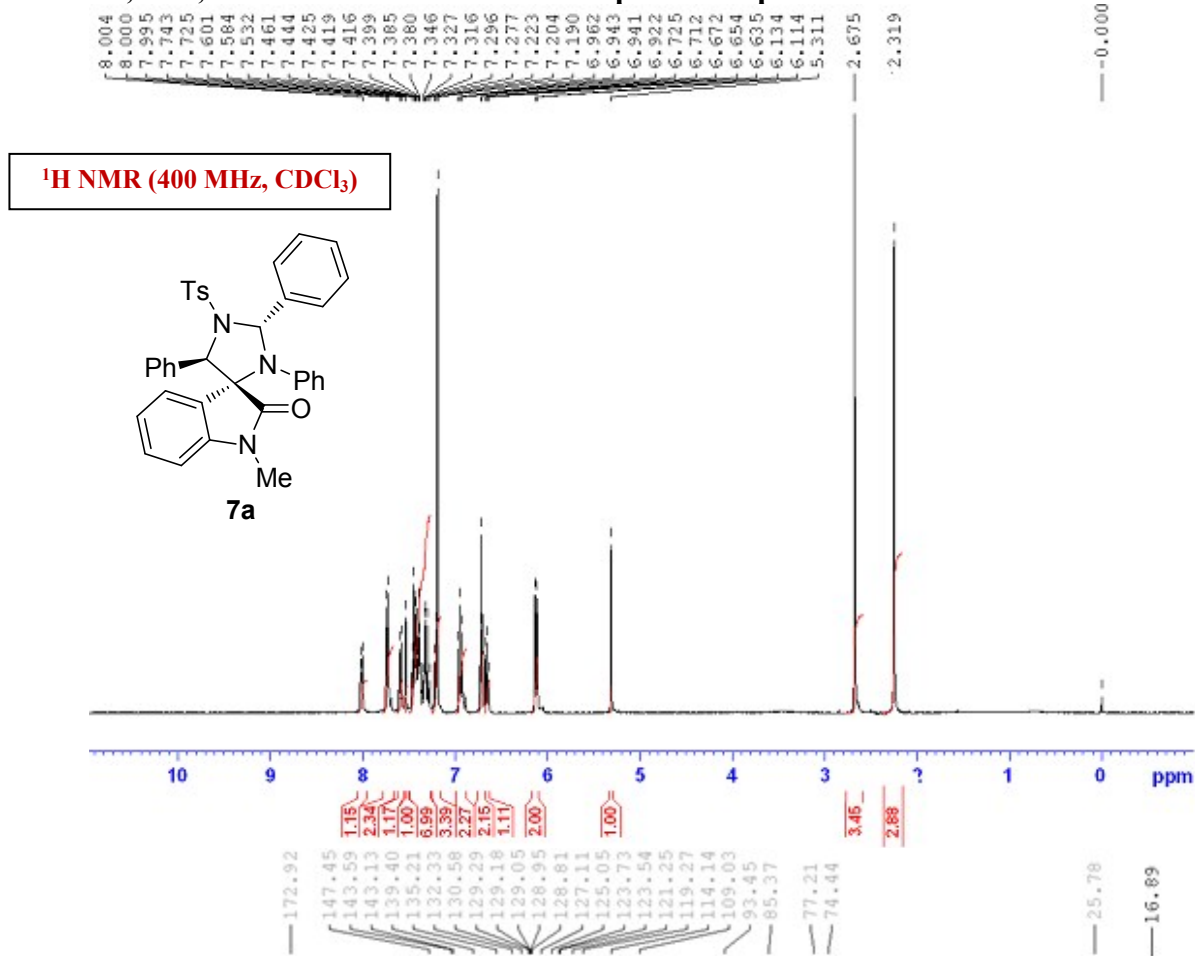


HRMS Spectrum

S4#467 RT: 2.03 A1: 1 NL: 2.96E8
T: FTMS+pESI Full ms [100.00-1000.00]

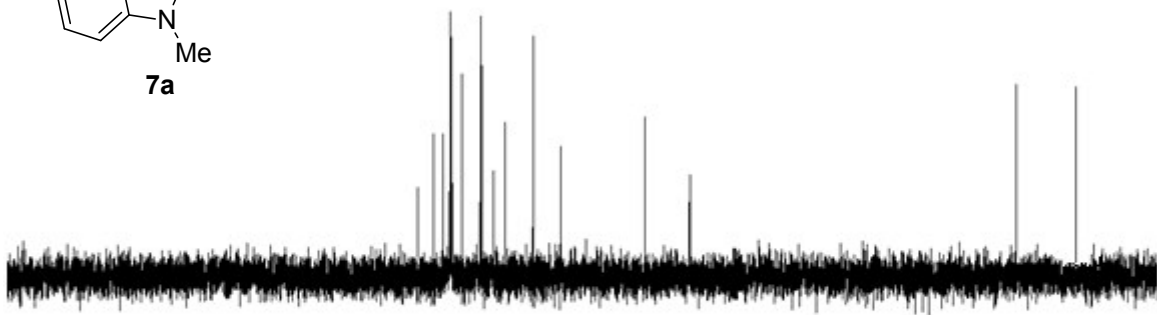
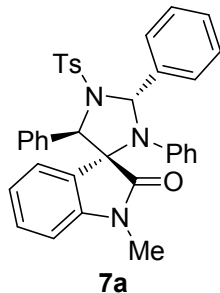


1H, 13C, DEPT135 NMR and HRMS spectra of spiroindoloimidazolidines



148.63
 132.34
 130.58
 129.30
 129.19
 128.96
 128.82
 127.11
 123.74
 123.54
 121.25
 119.27
 114.13
 109.04
 — 93.45
 — 85.36
 — 25.79
 — 16.89

¹³C DEPT135 NMR (100 MHz, CDCl₃)

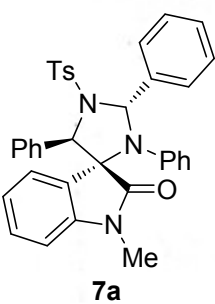
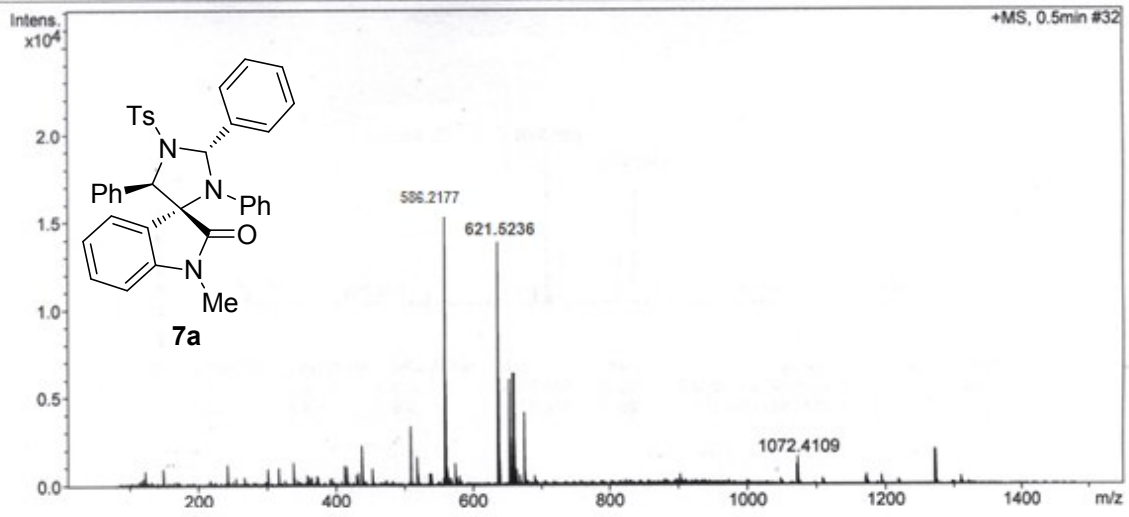


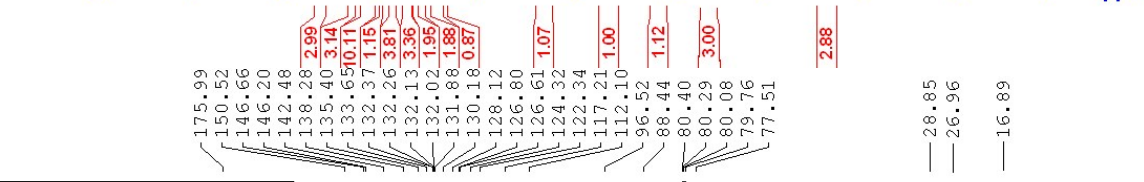
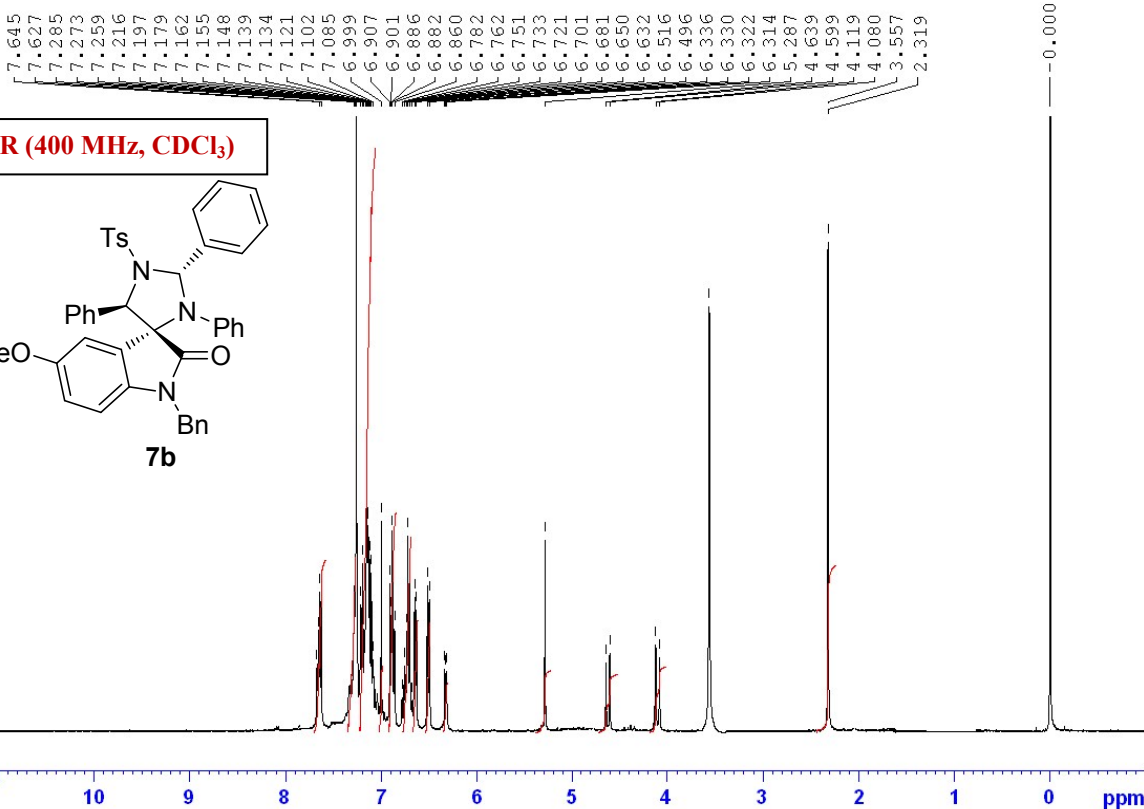
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 ppm

HRMS Spectrum

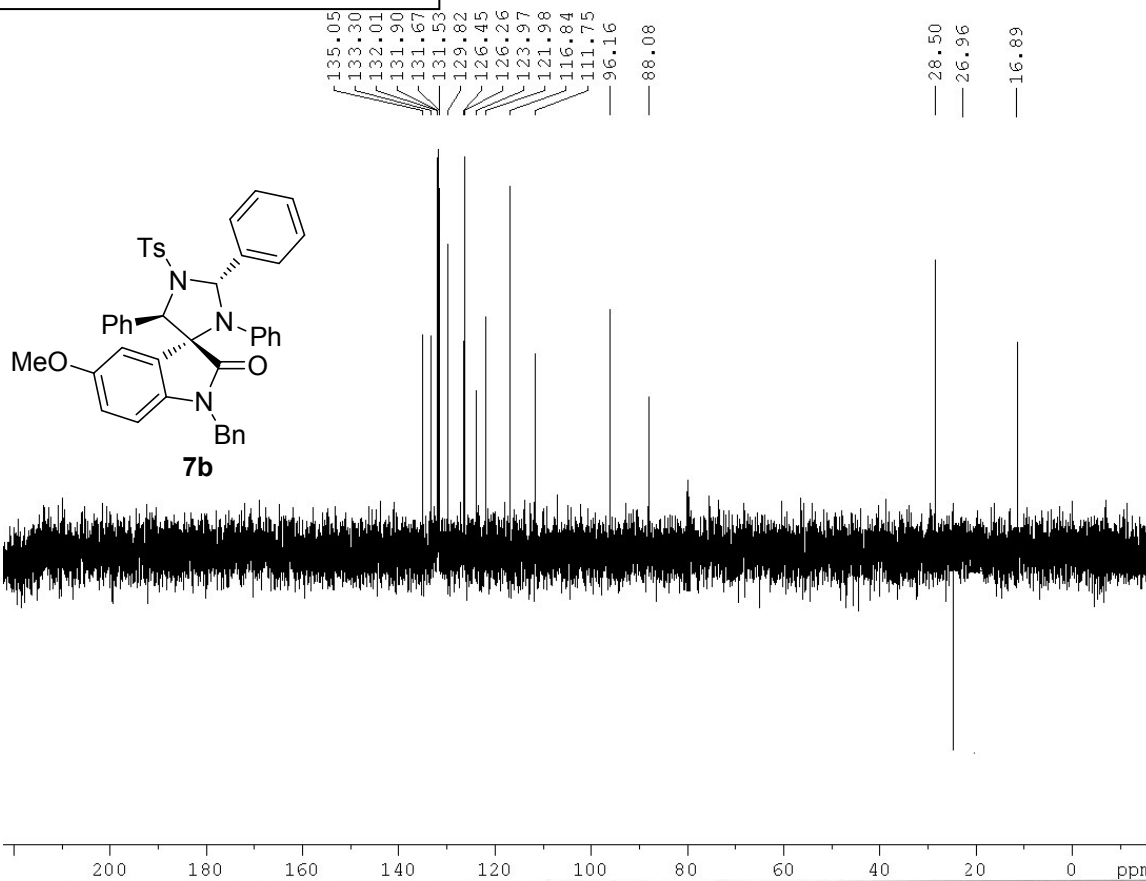
Acquisition Parameter

Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Not active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source



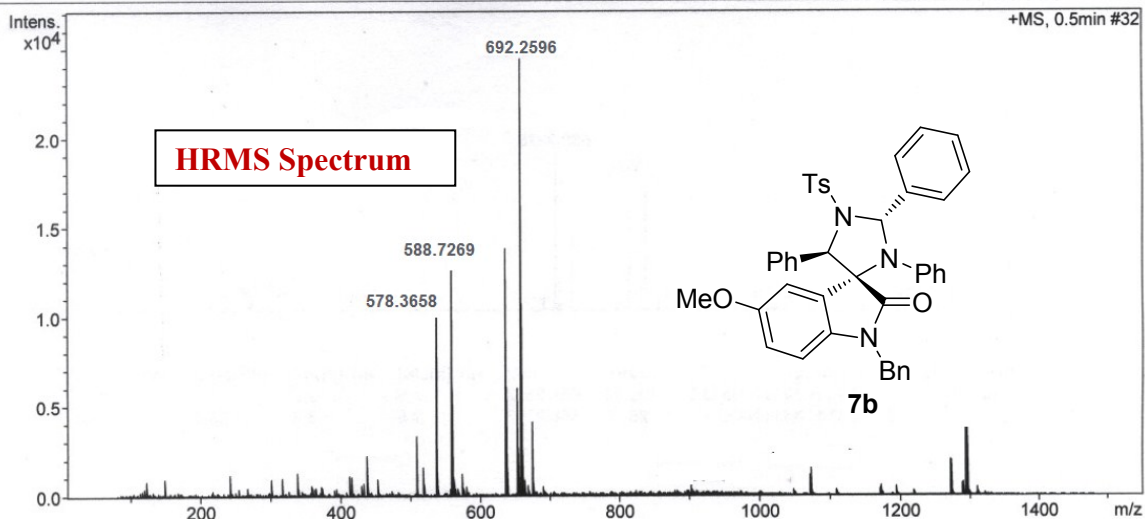


¹³C DEPT135 NMR (100 MHz, CDCl₃)

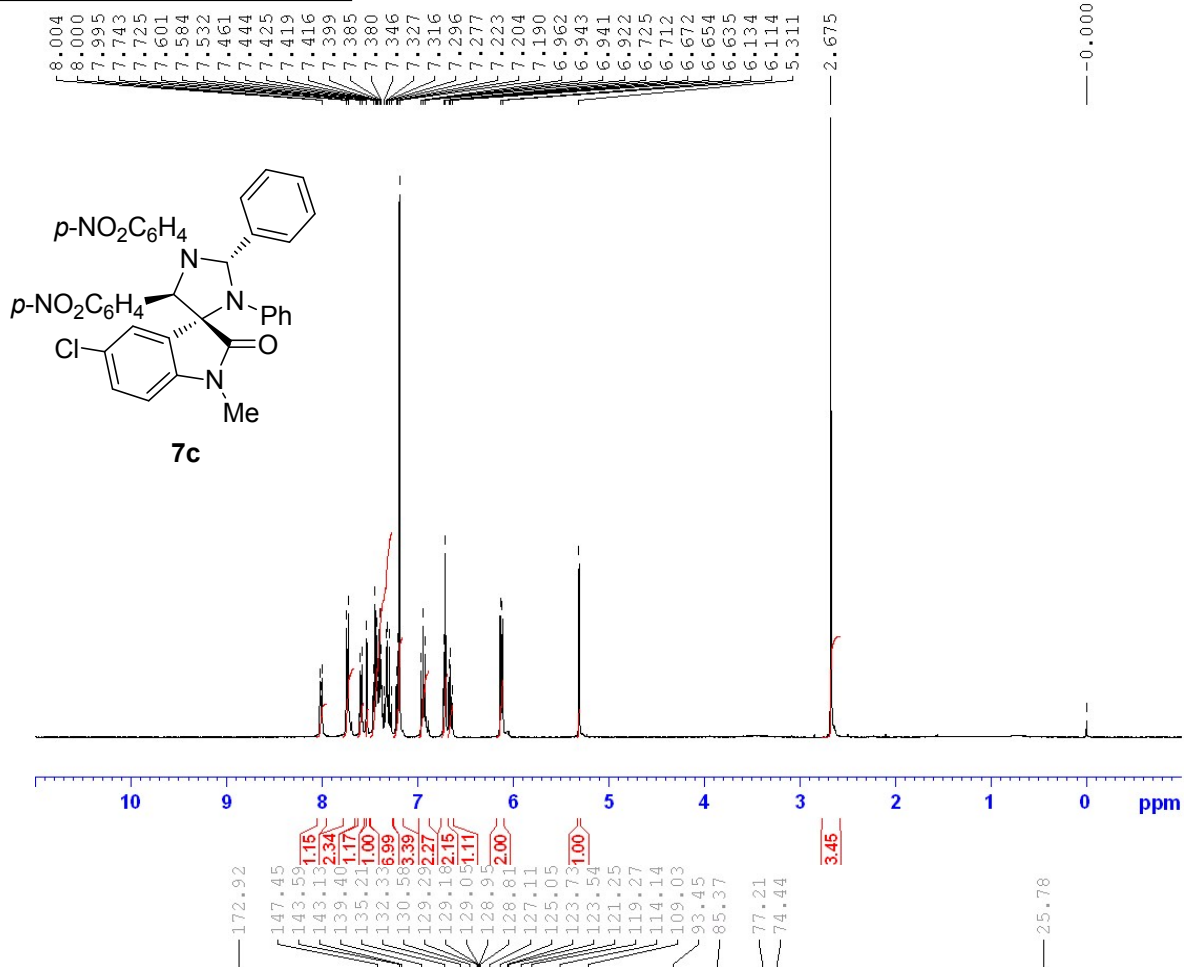


Acquisition Parameter

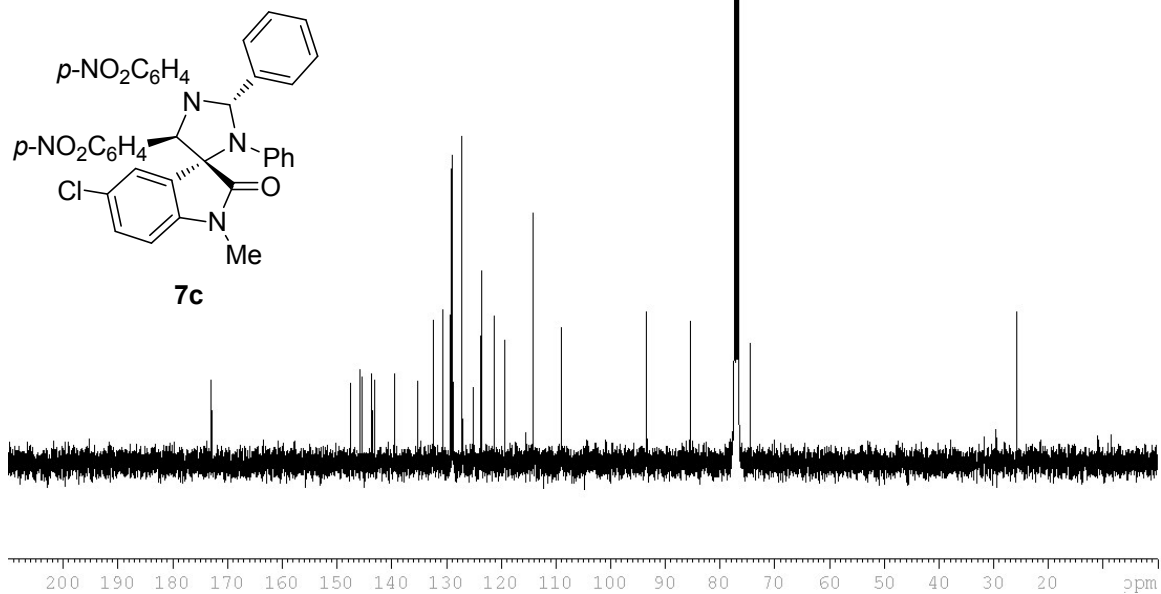
Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Not active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source



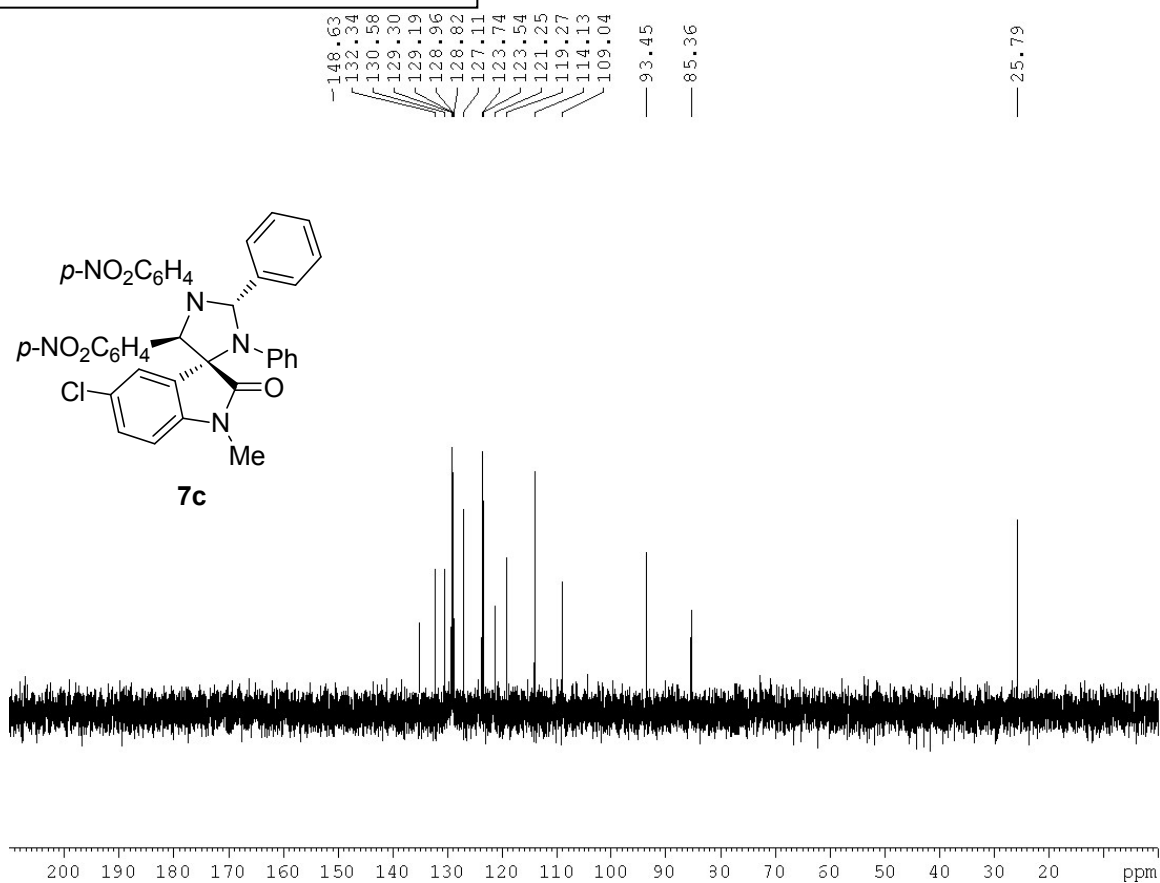
¹H NMR (400 MHz, CDCl₃)



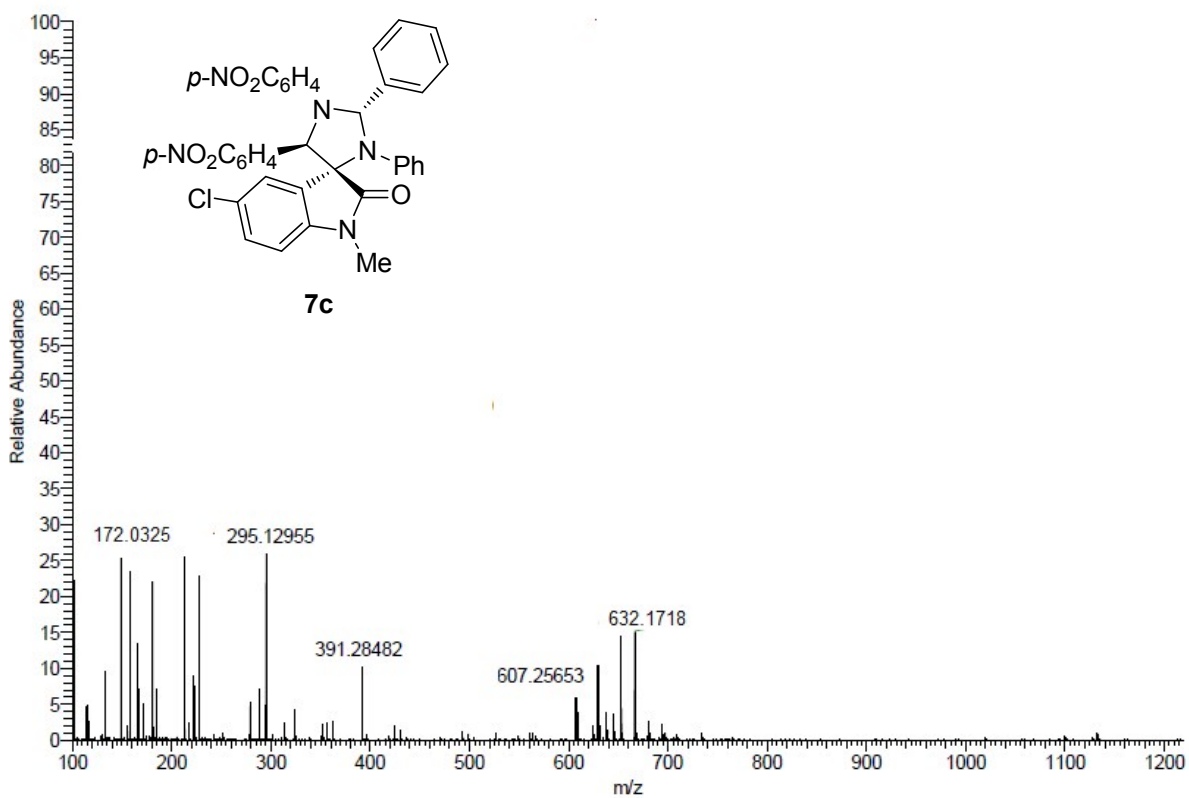
¹³C NMR (100 MHz, CDCl₃)

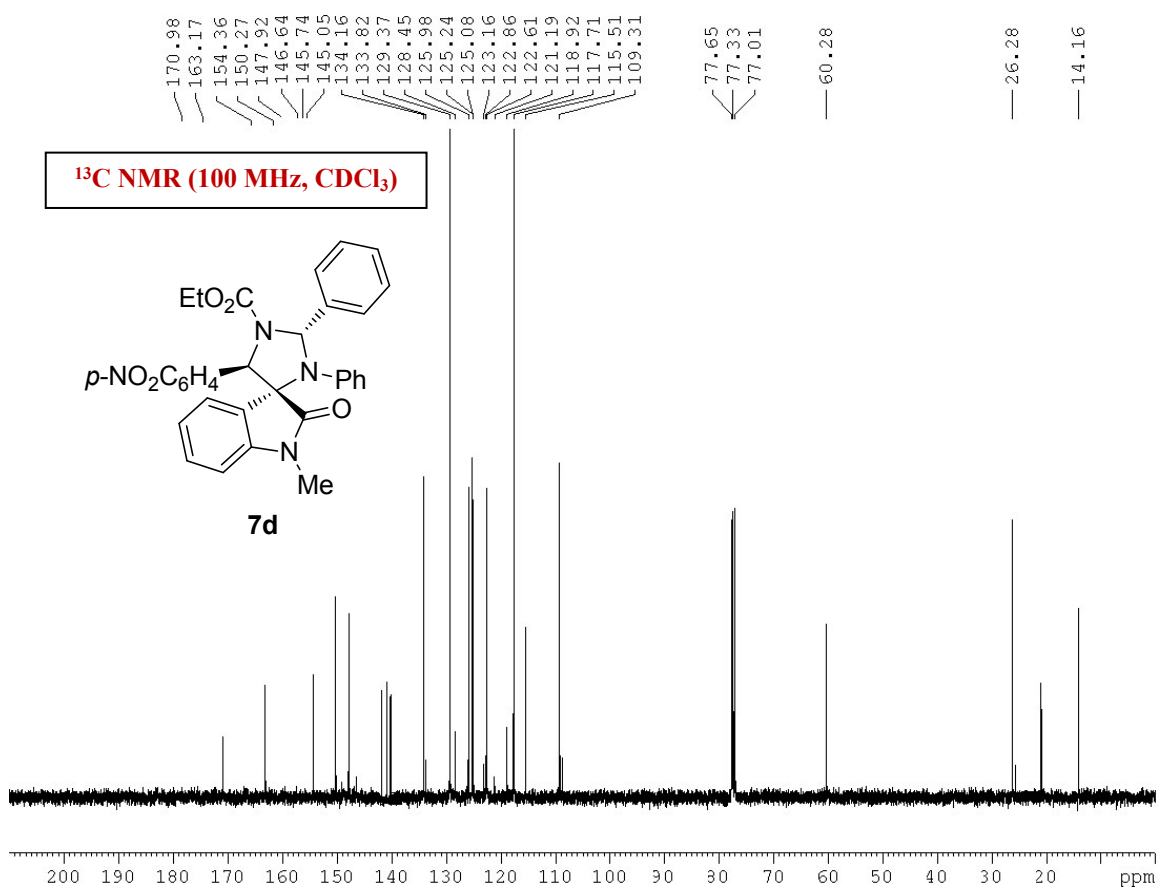
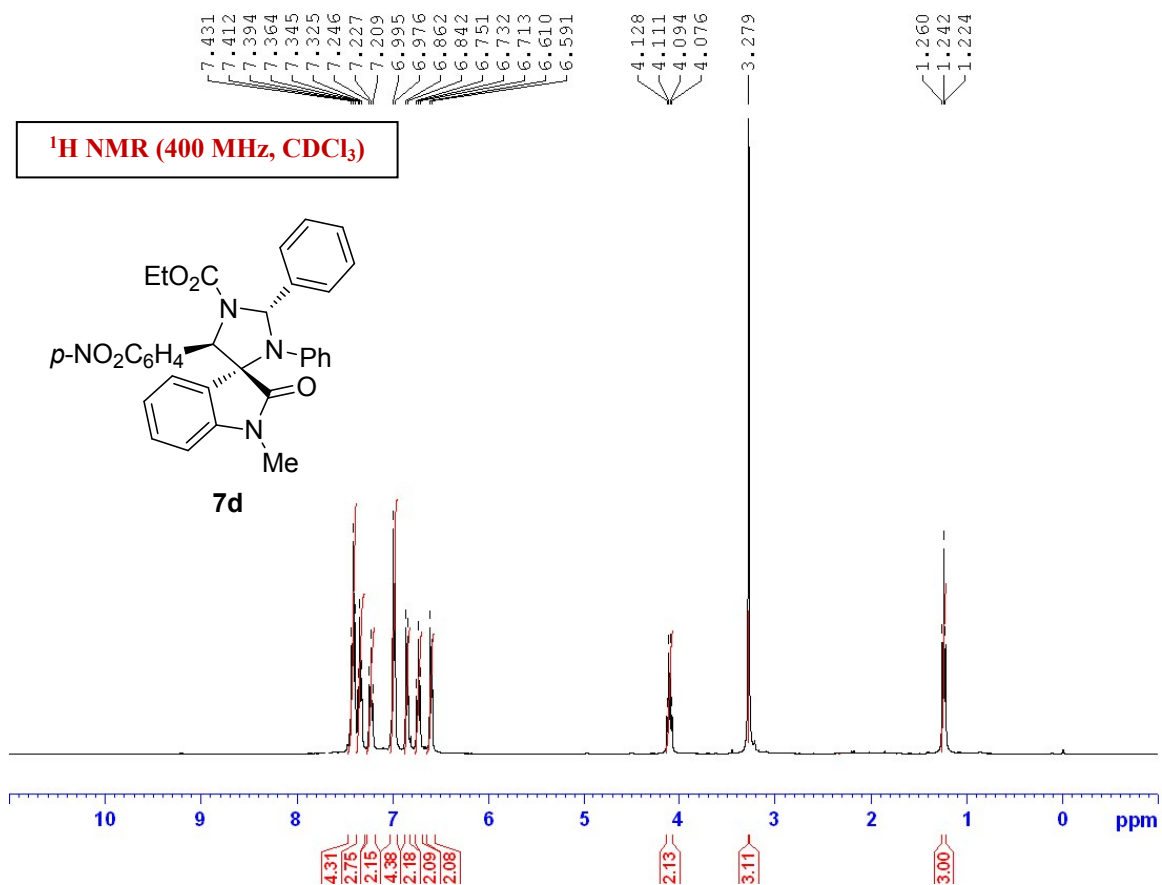


¹³C DEPT135 NMR (100 MHz, CDCl₃)

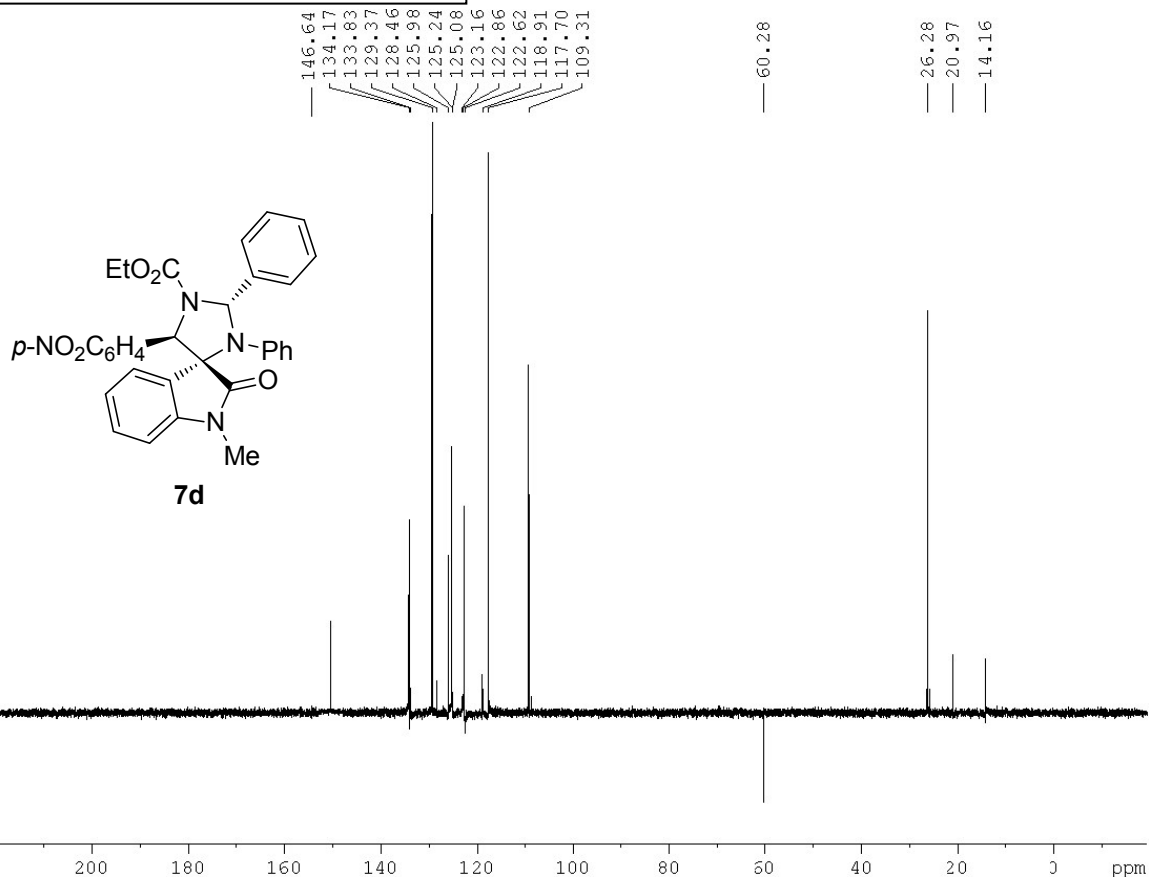


HRMS Spectrum

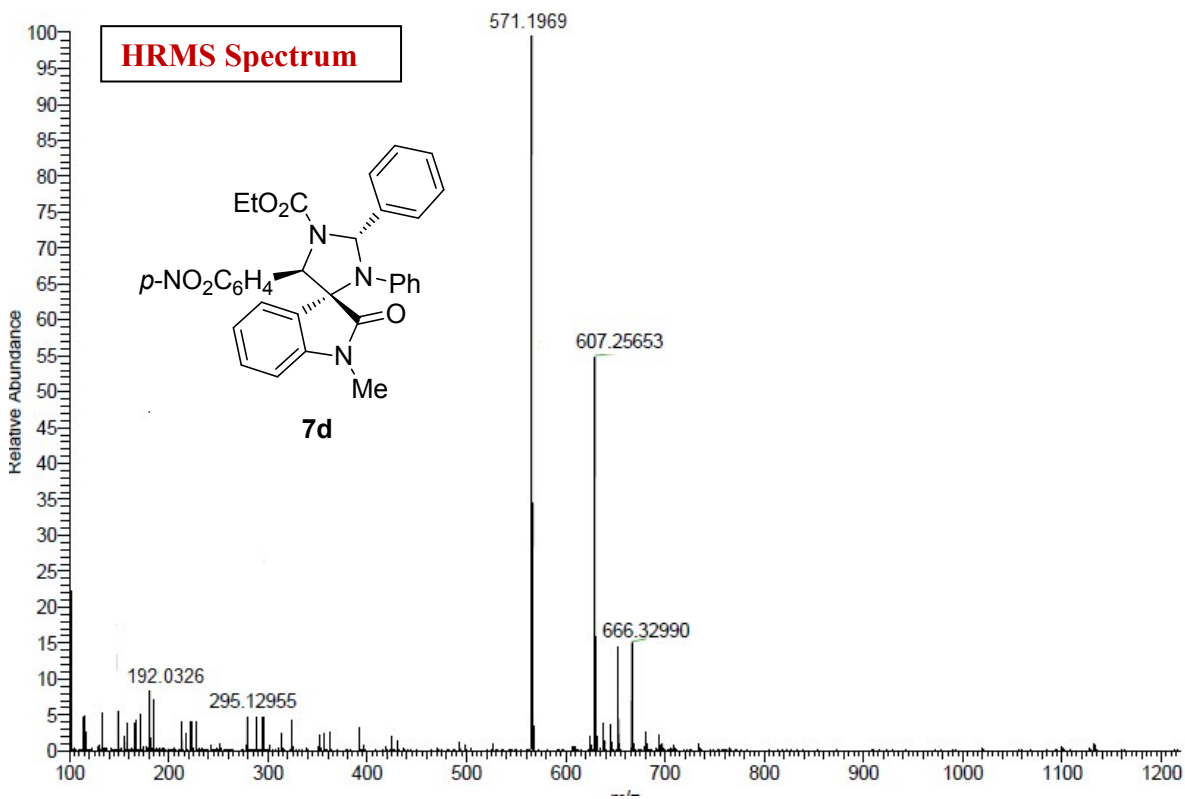


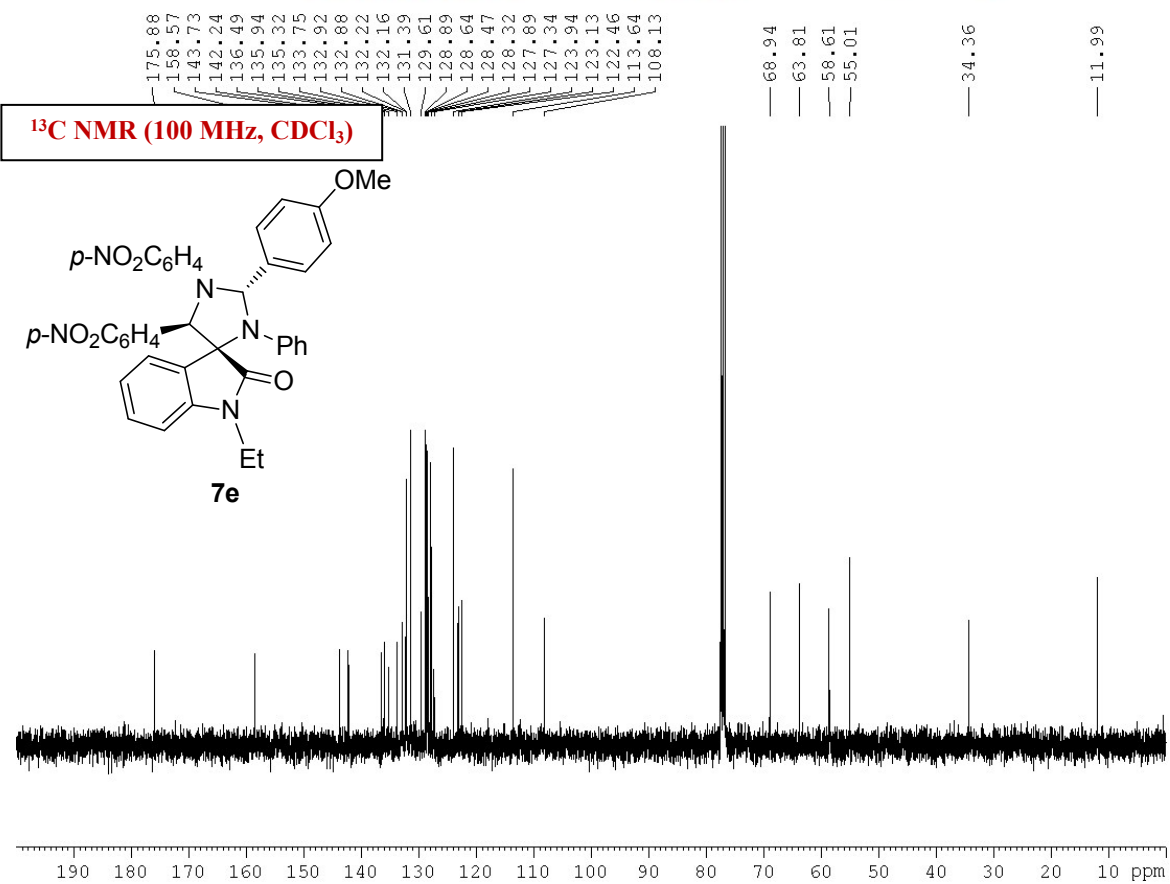
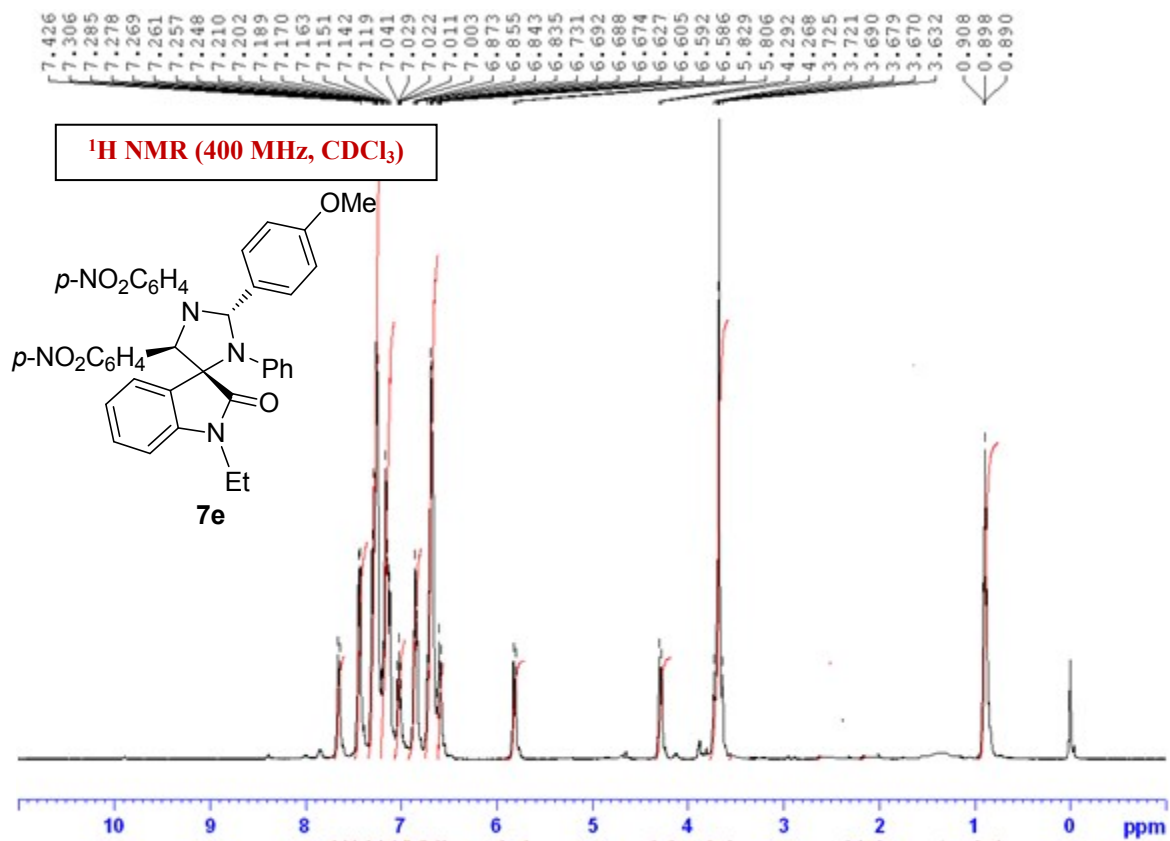


¹³C DEPT135 NMR (100 MHz, CDCl₃)

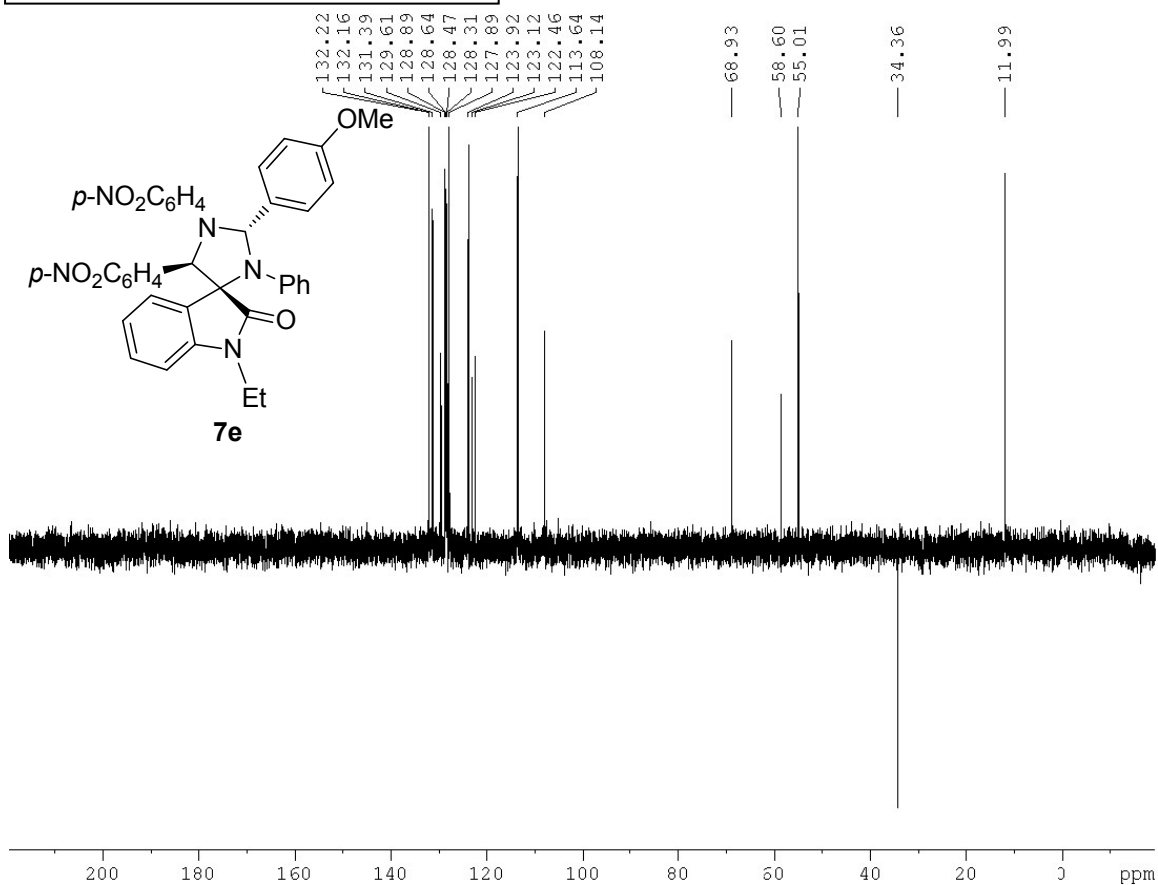


MSGK-299 #63 RT: 0.94 AV: 1 NL: 1.00E6
: FTMS {1,1} + p ESI Full lock ms [100.00-2000.00]





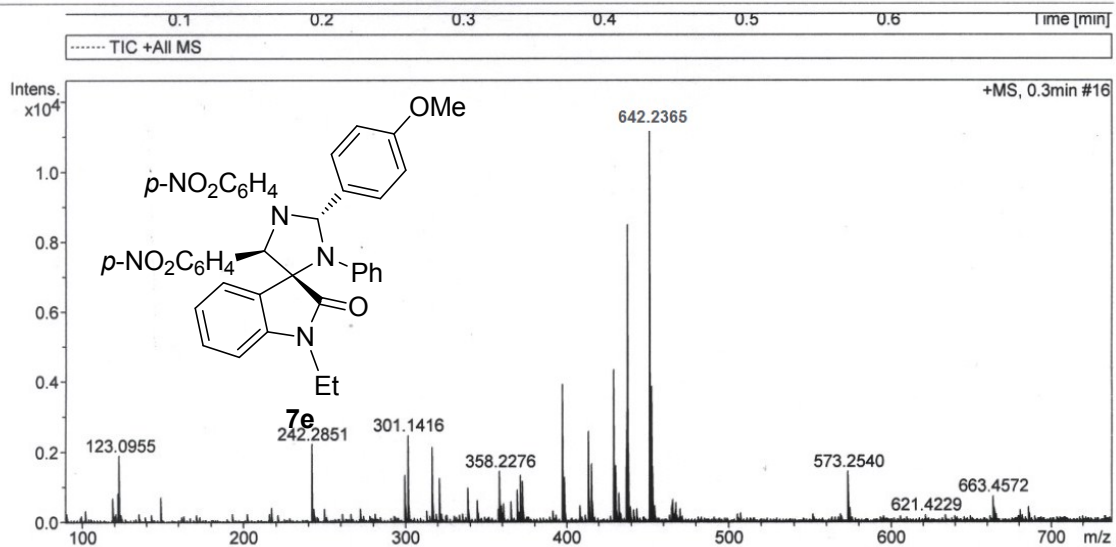
¹³C DEPT135 NMR (100 MHz, CDCl₃)



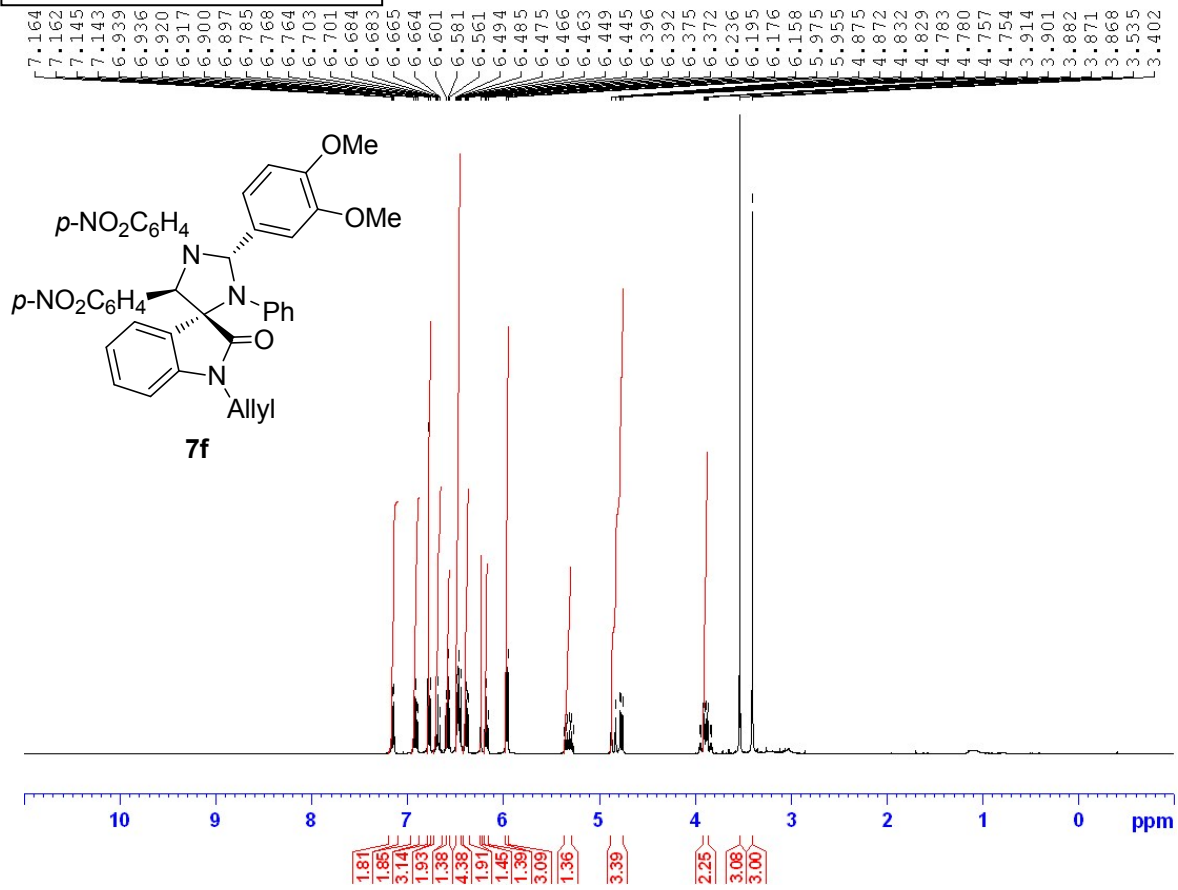
HRMS Spectrum

Acquisition Parameter

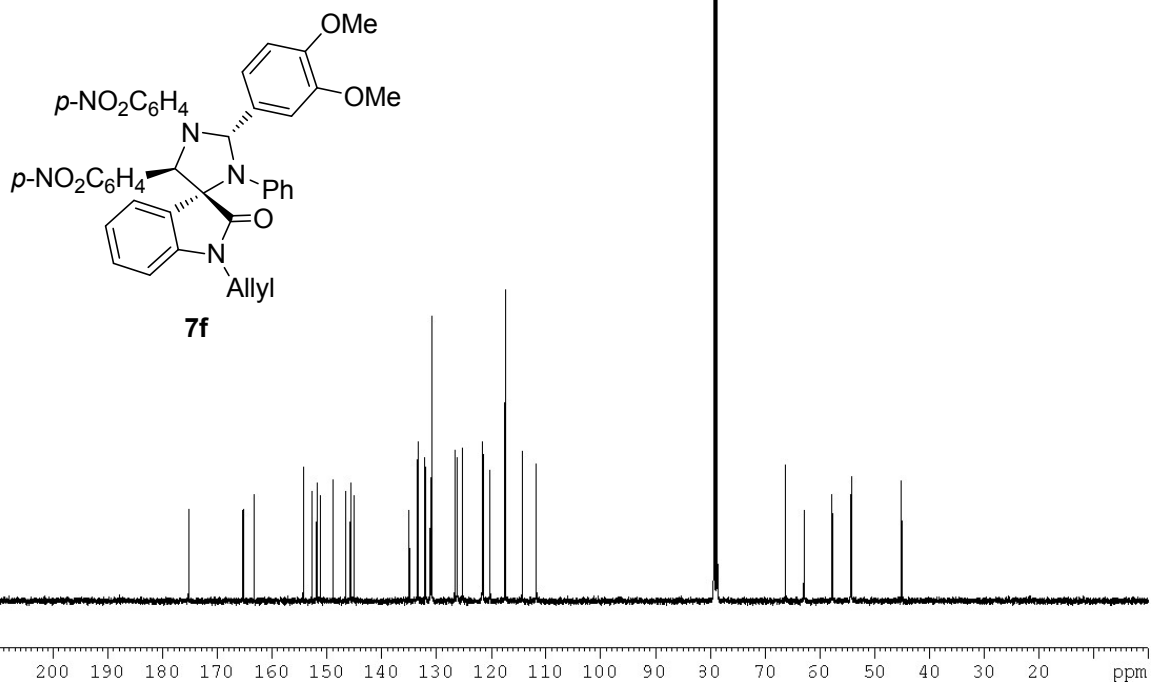
Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Not active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source



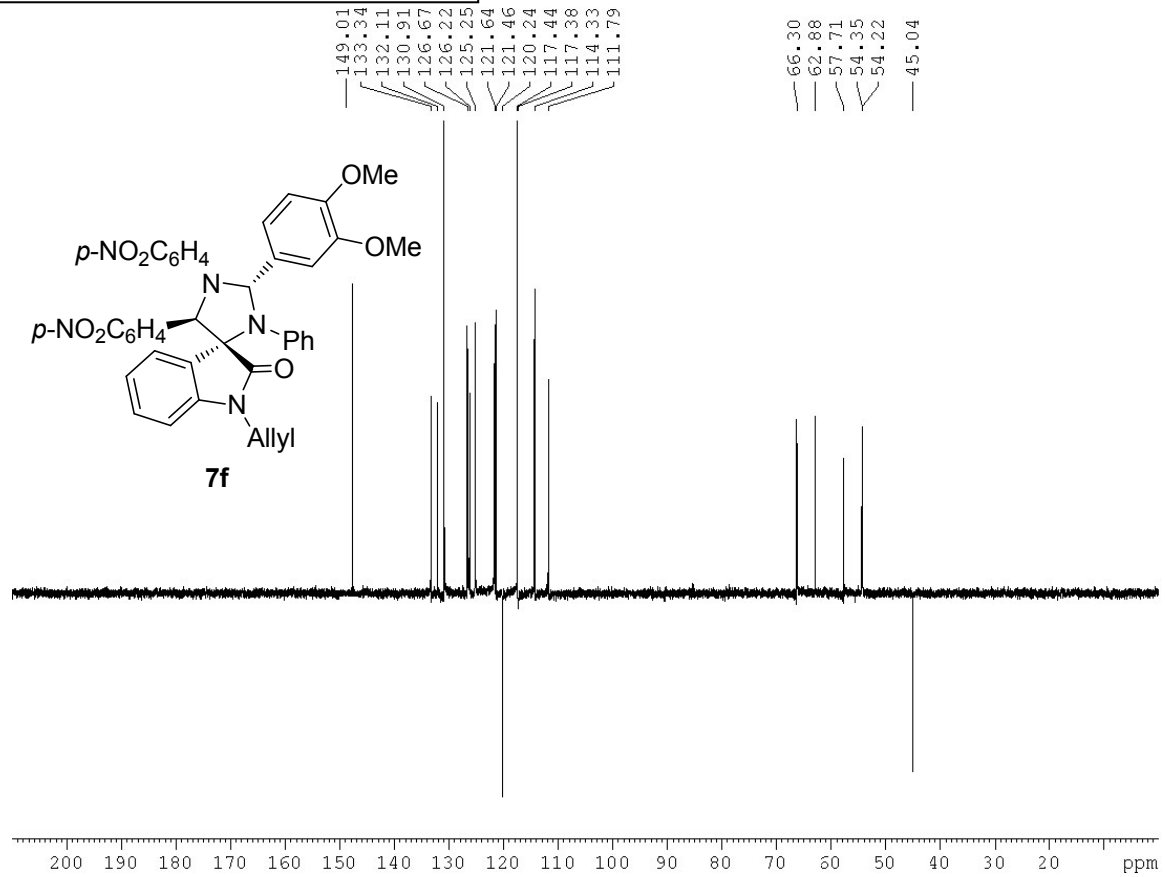
¹H NMR (400 MHz, CDCl₃)



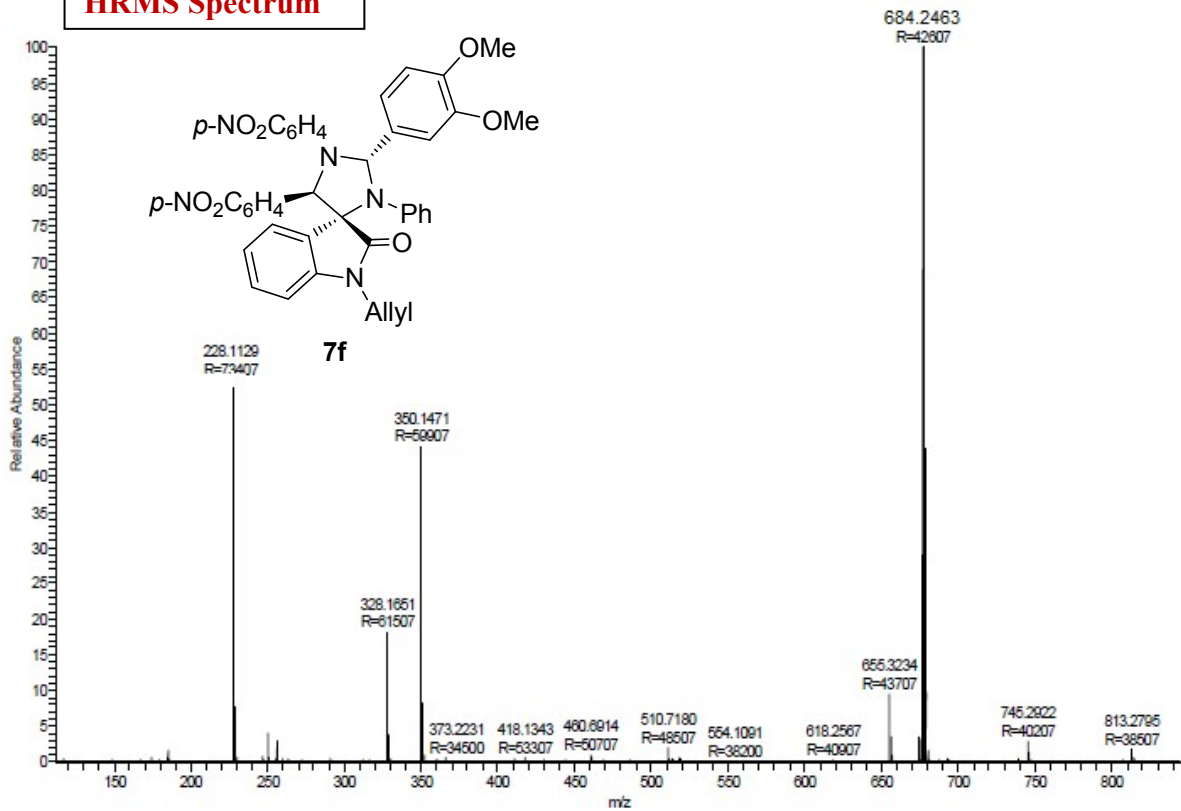
¹³C NMR (100 MHz, CDCl₃)



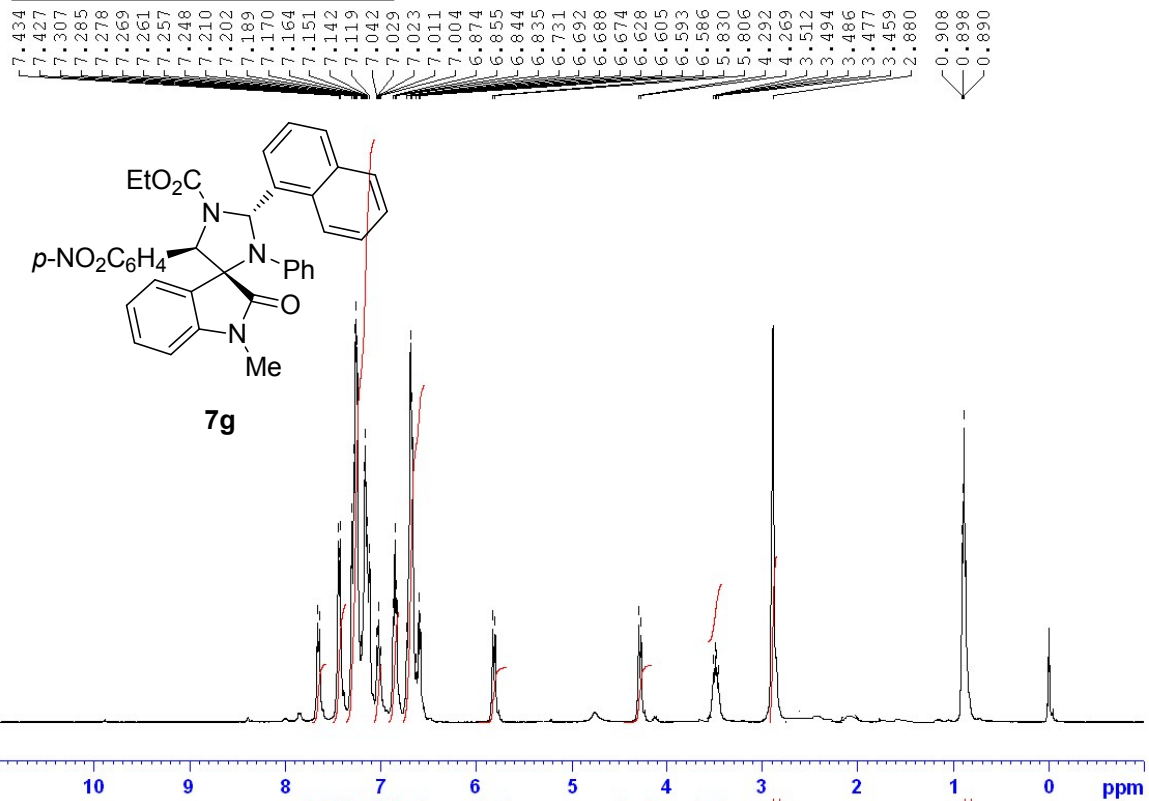
¹³C DEPT135 NMR (100 MHz, CDCl₃)



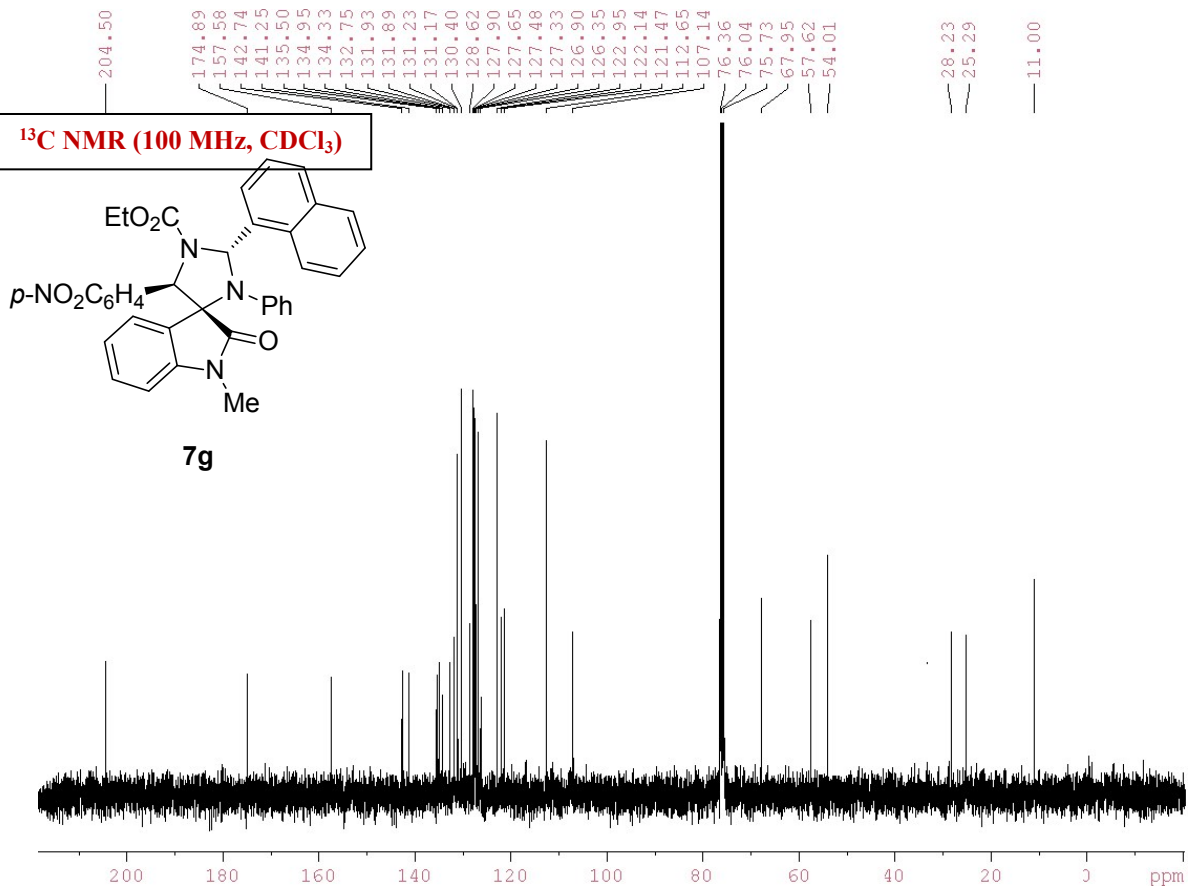
HRMS Spectrum

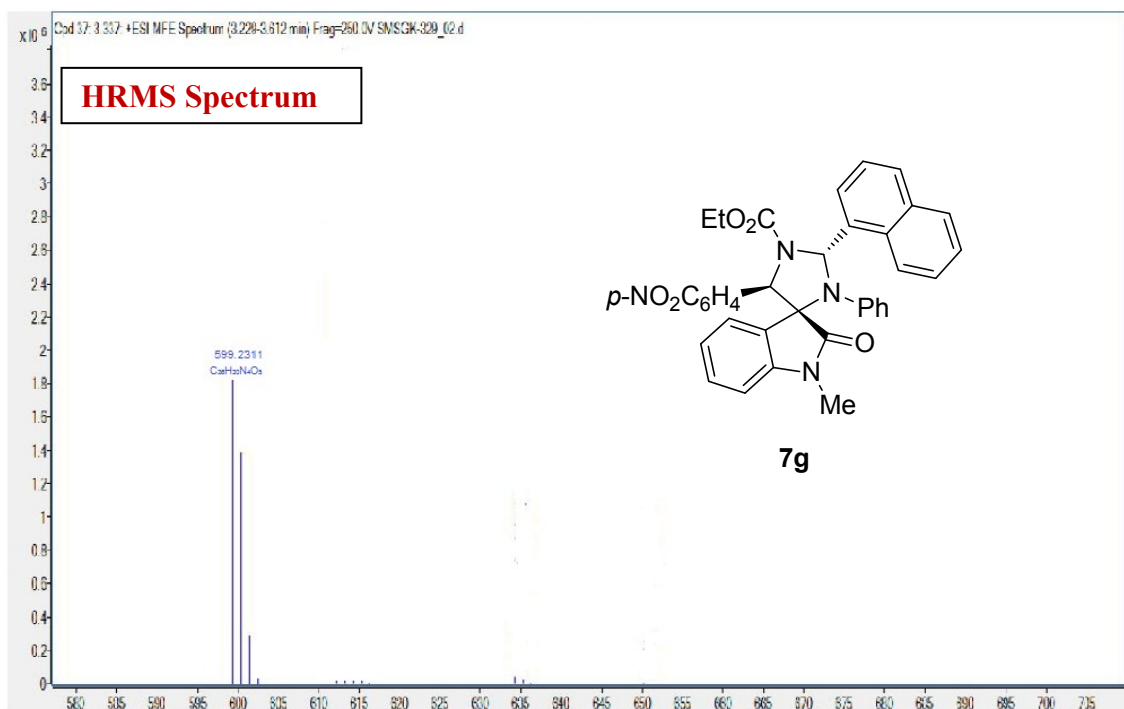
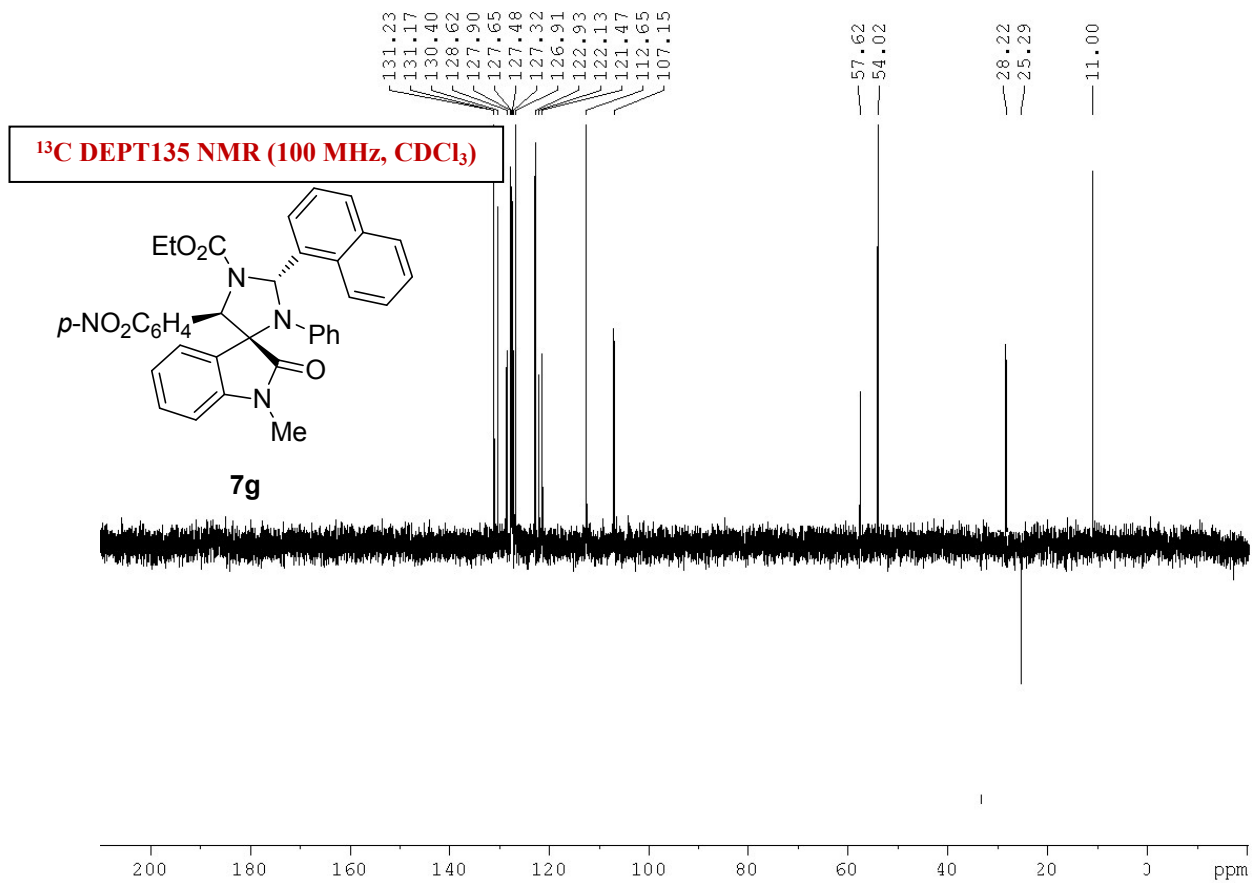


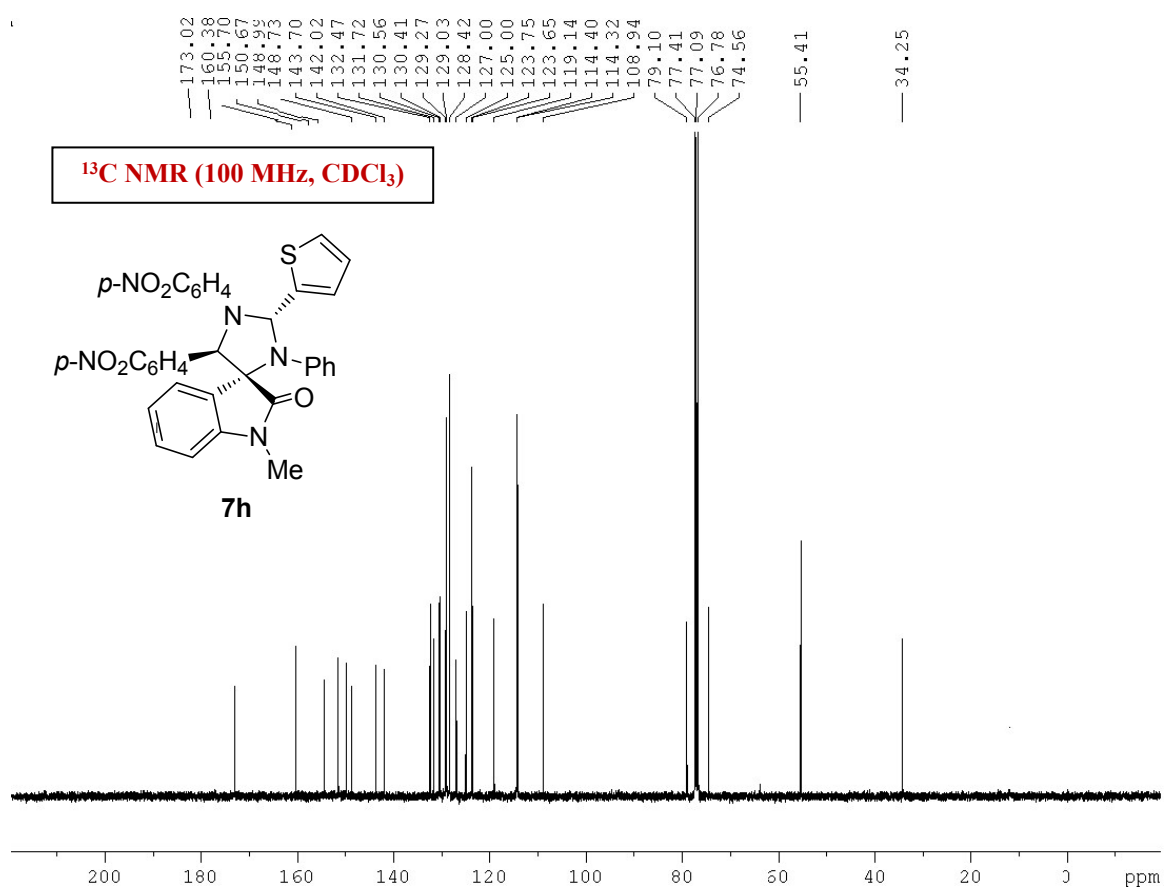
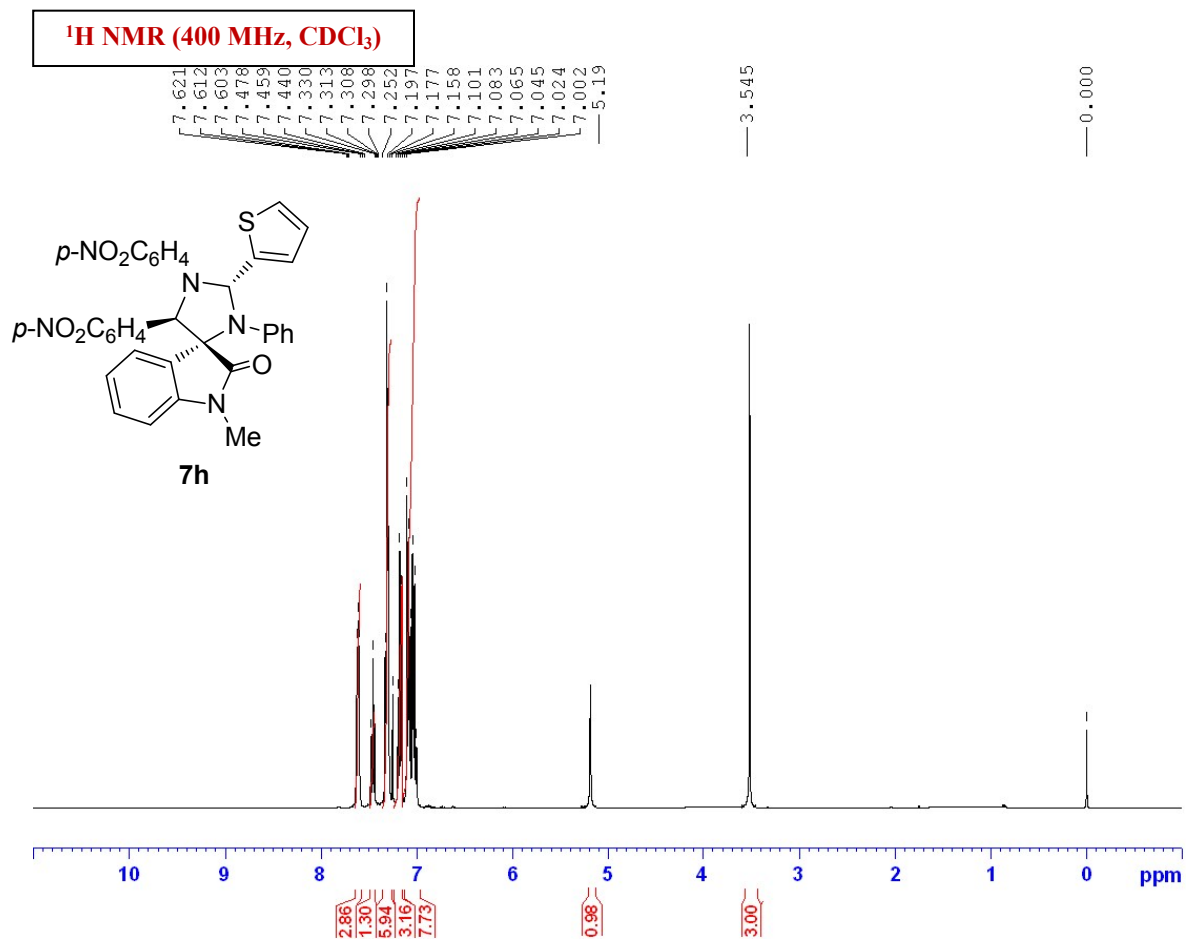
¹H NMR (400 MHz, CDCl₃)



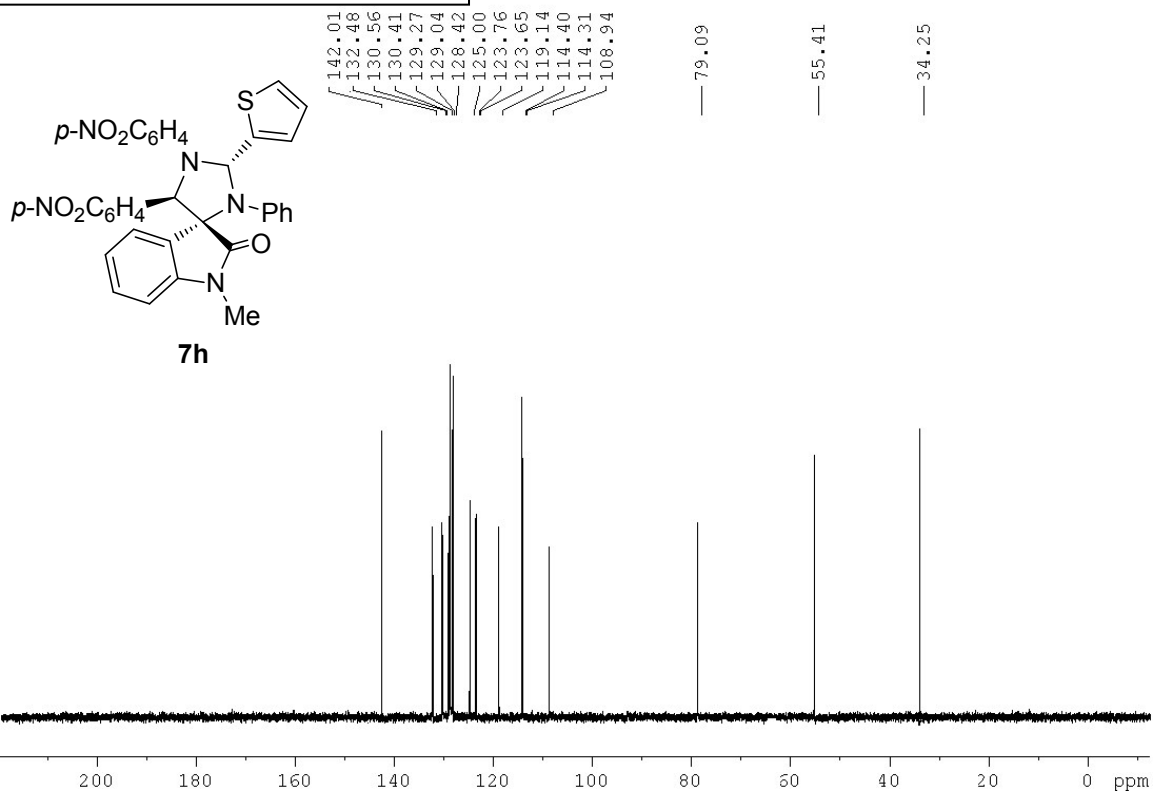
¹³C NMR (100 MHz, CDCl₃)



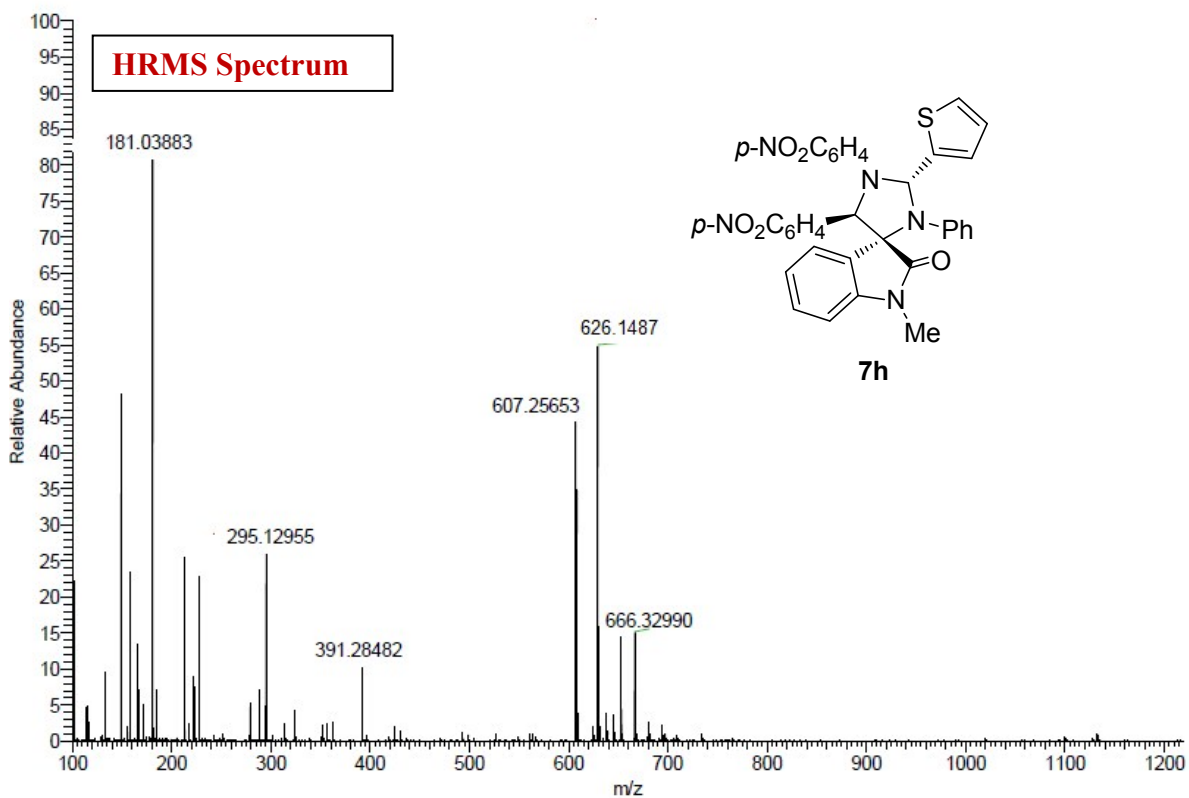




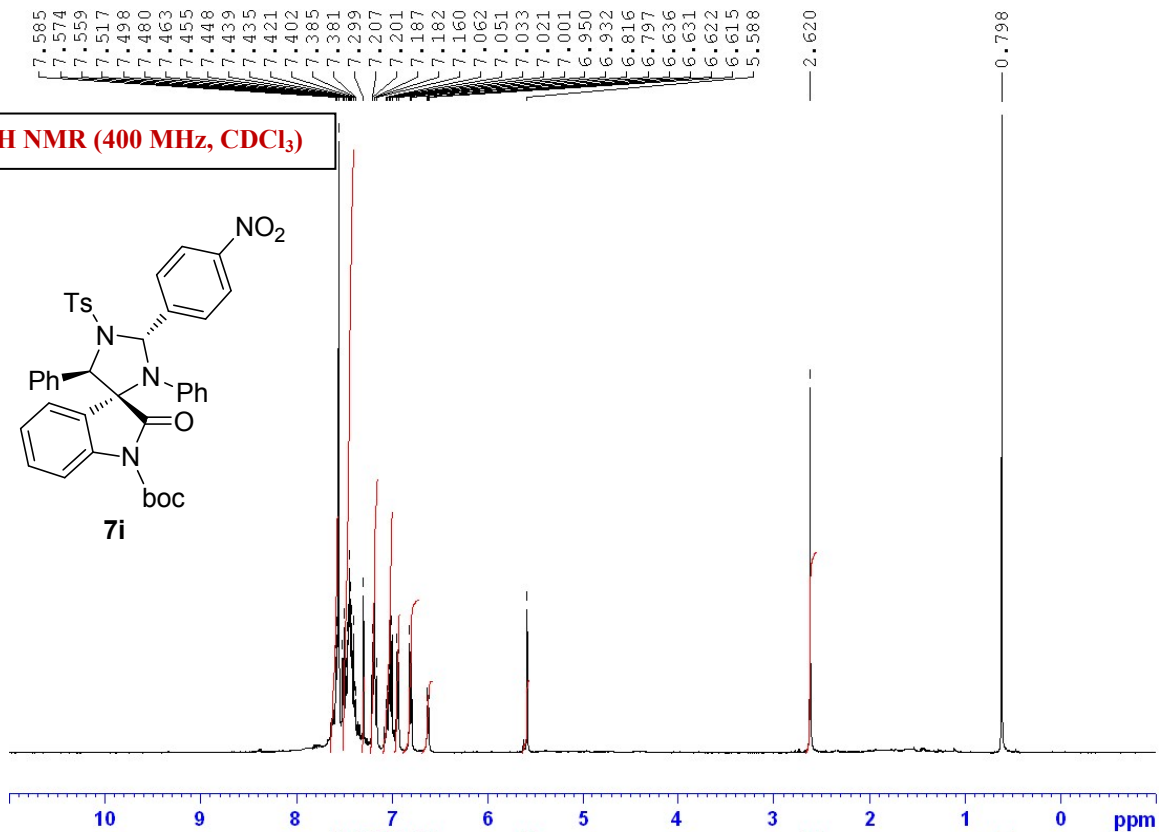
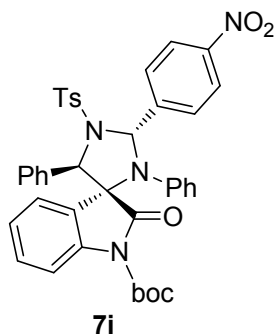
¹³C DEPT135 NMR (100 MHz, CDCl₃)



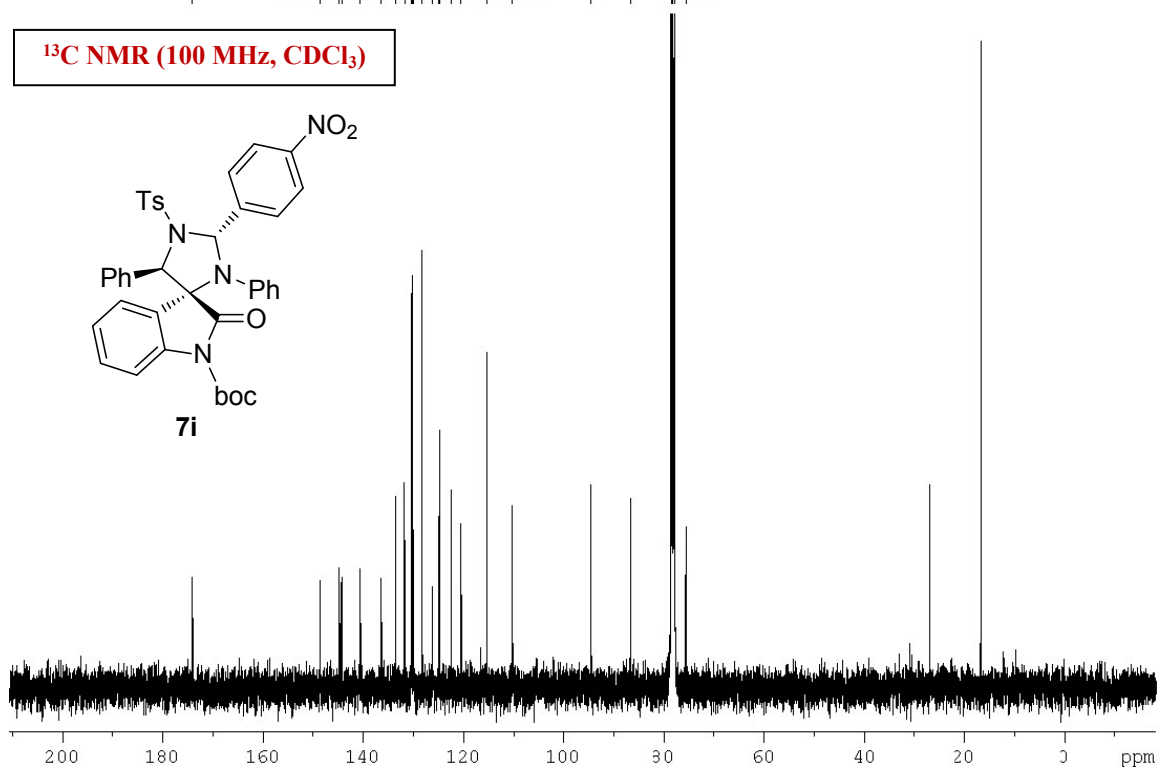
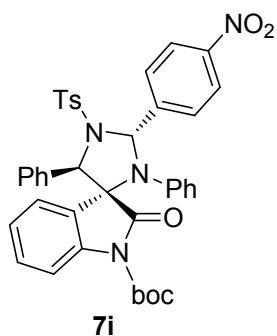
MSGK-299 #63 RT: 0.94 AV: 1 NL: 1.00E6
: FTMS (1,1) + p ESI Full lock ms [100.00-2000.00]



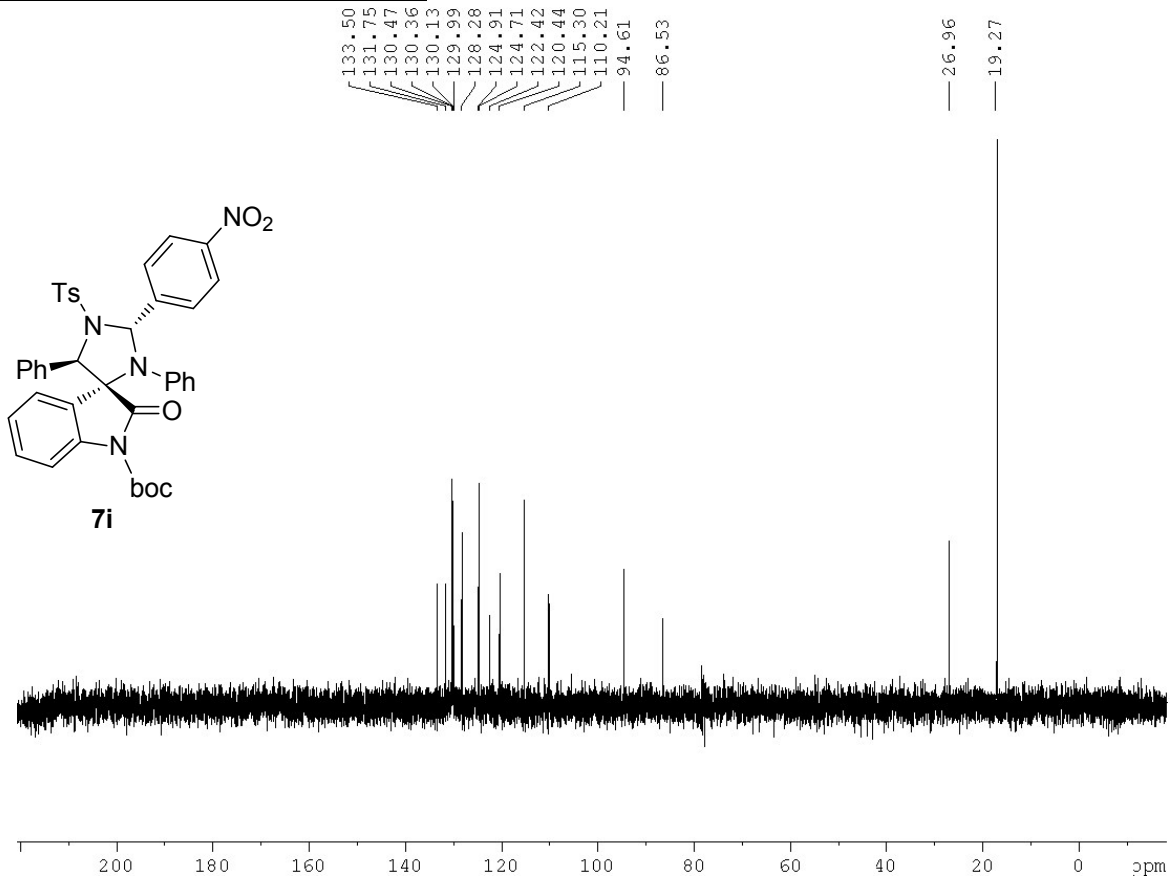
¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)

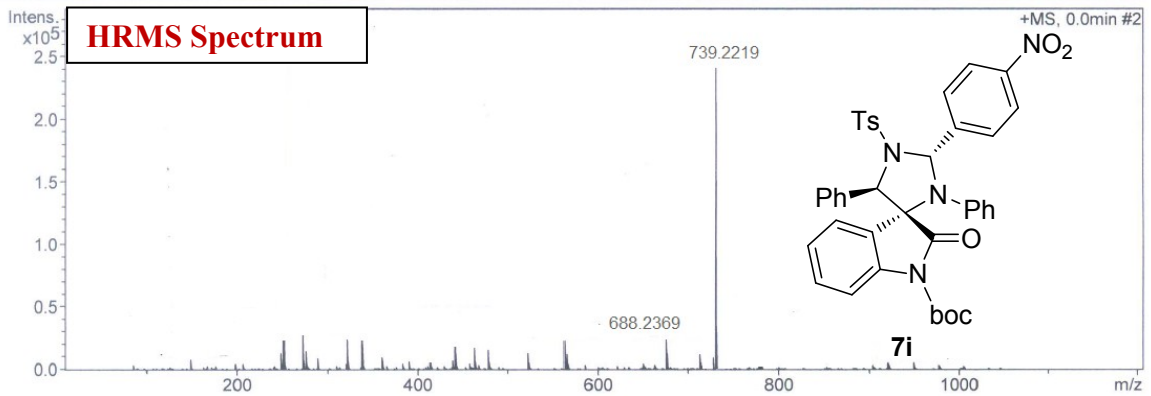


¹³C DEPT135 NMR (100 MHz, CDCl₃)

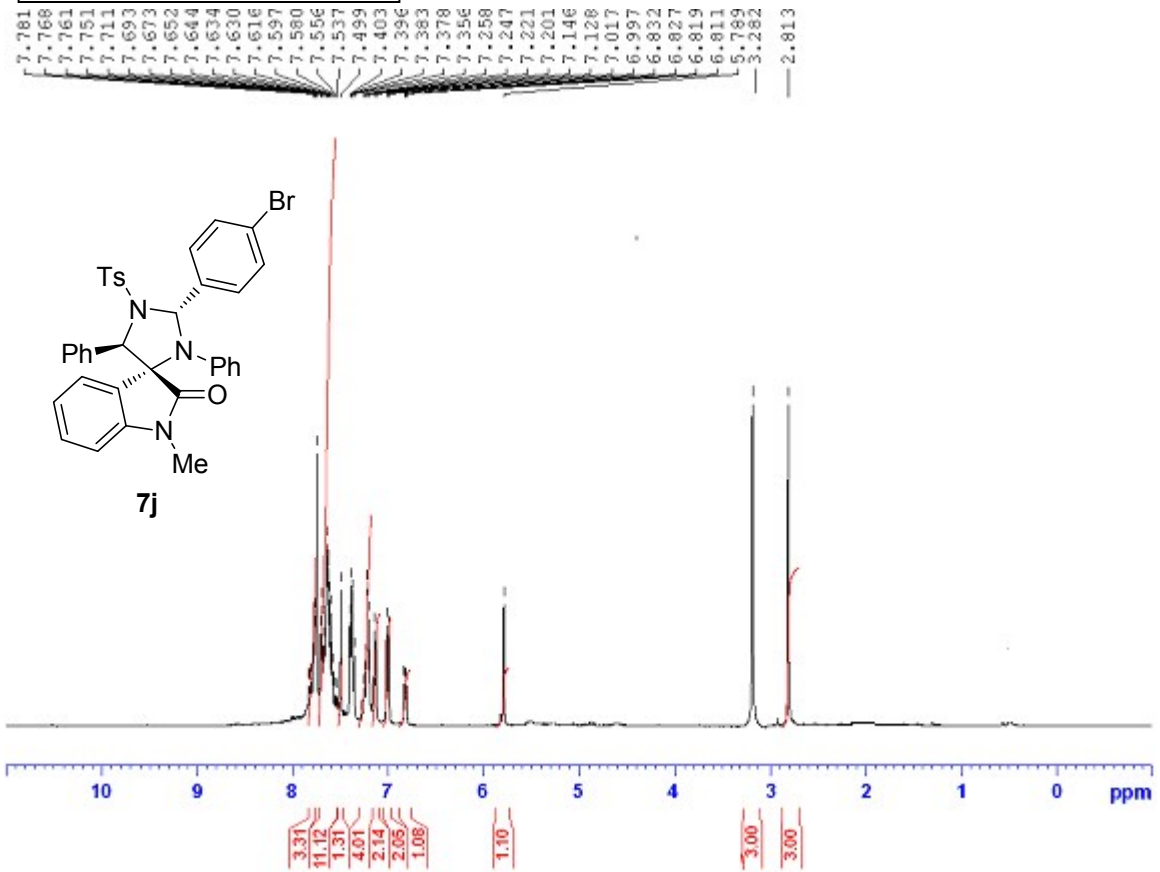


Acquisition Parameter

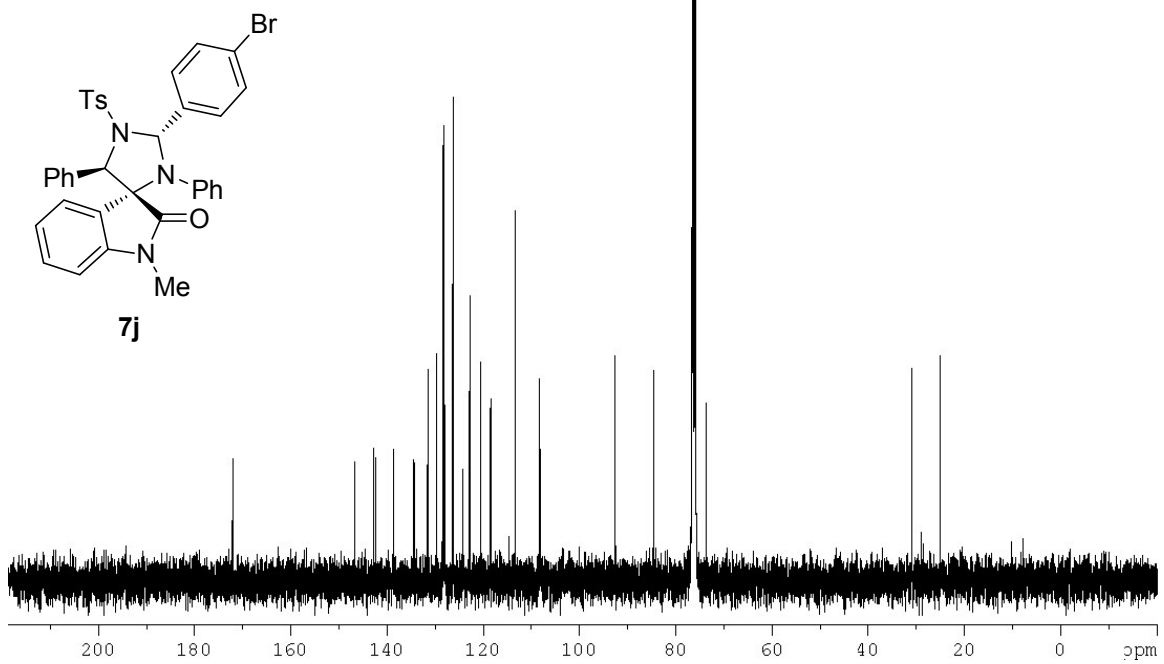
Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Active	Set Capillary	4500 V	Set Dry Heater	190 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source



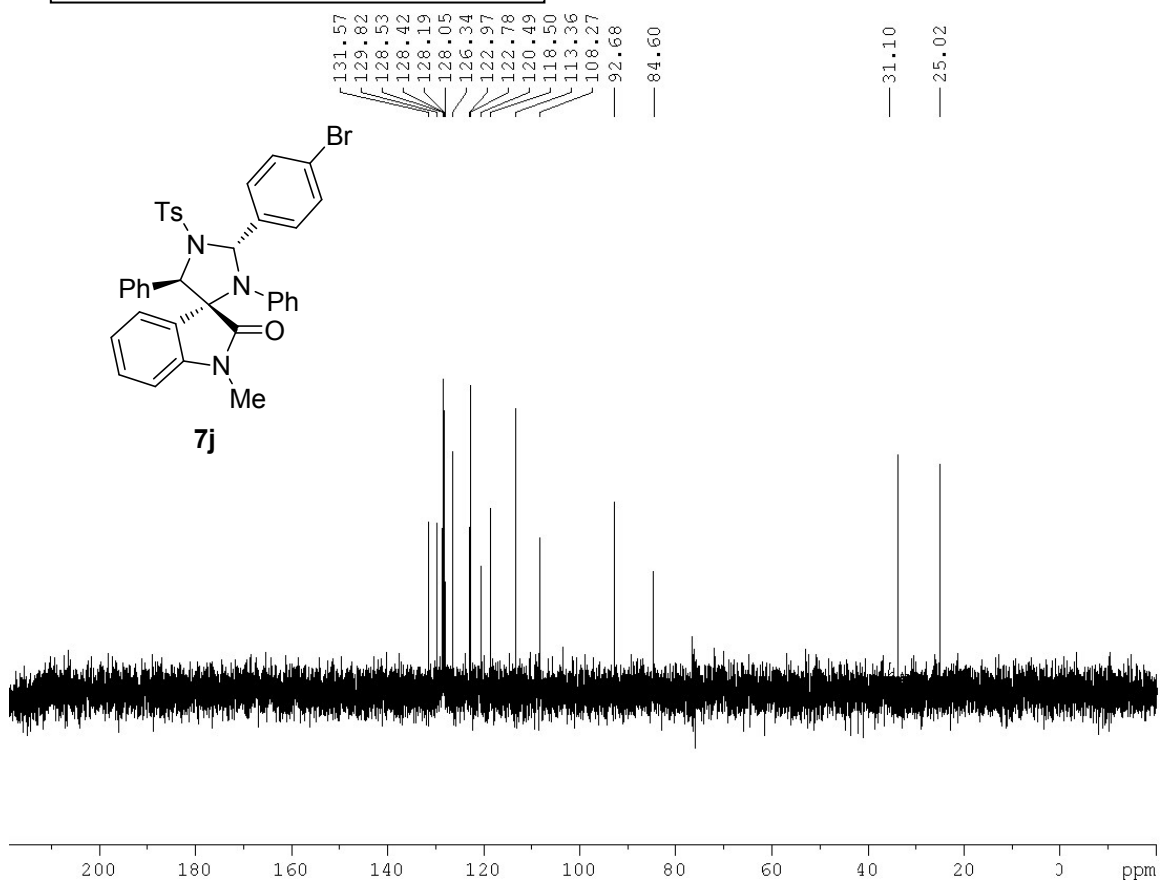
¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)



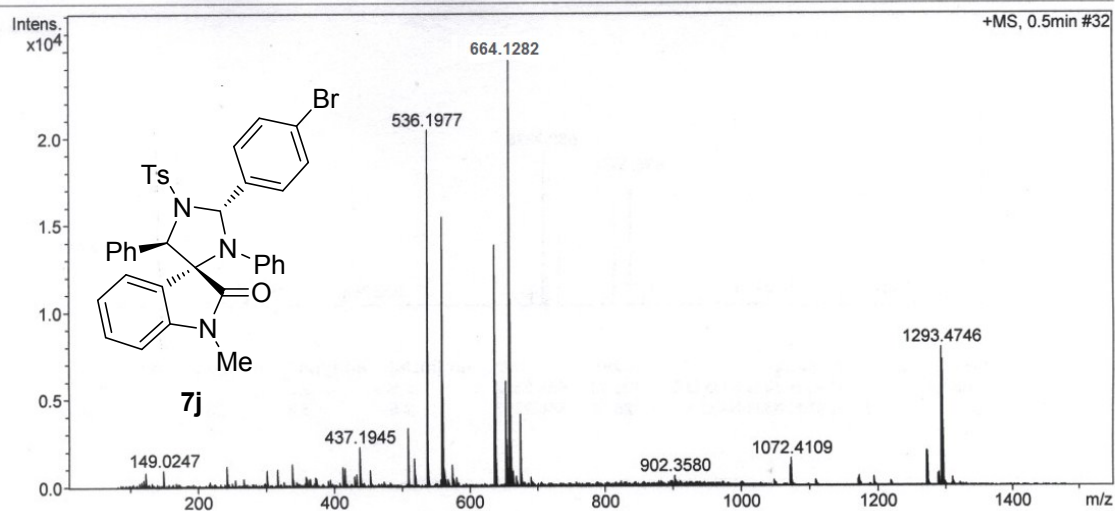
¹³C DEPT135 NMR (100 MHz, CDCl₃)



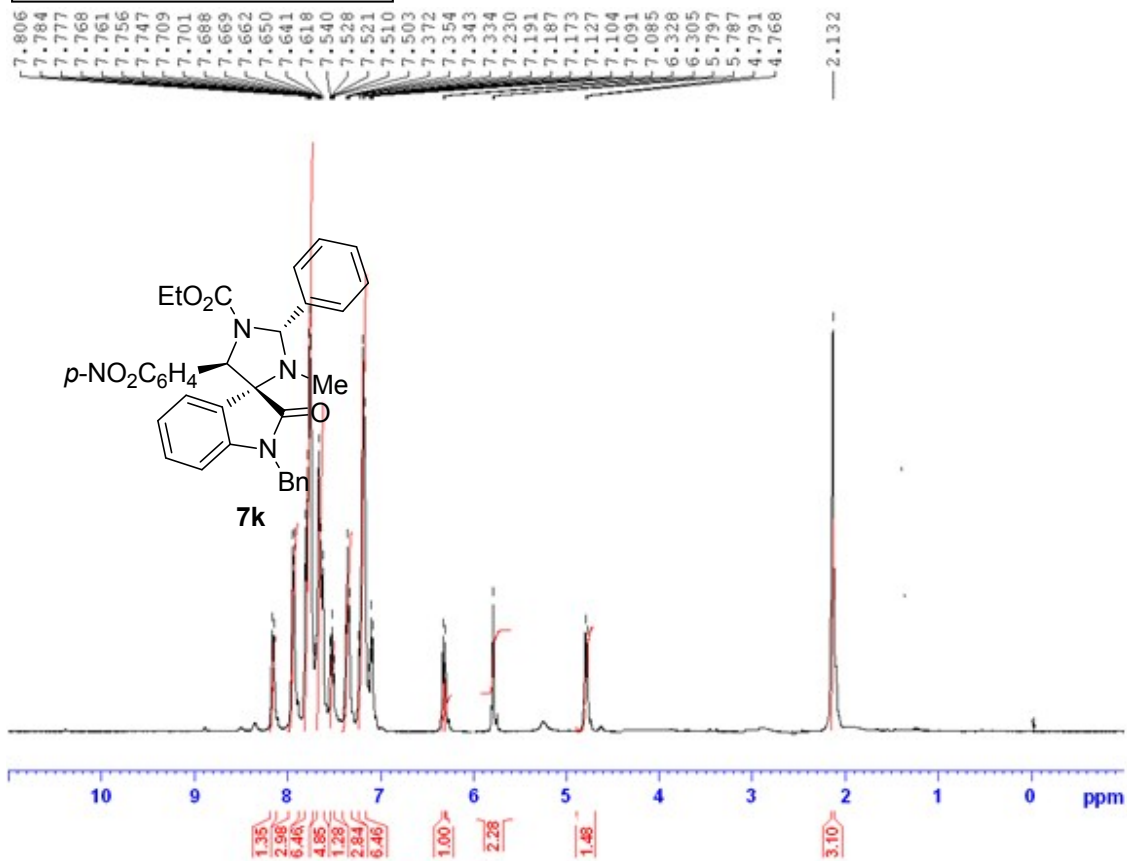
HRMS Spectrum

Acquisition Parameter

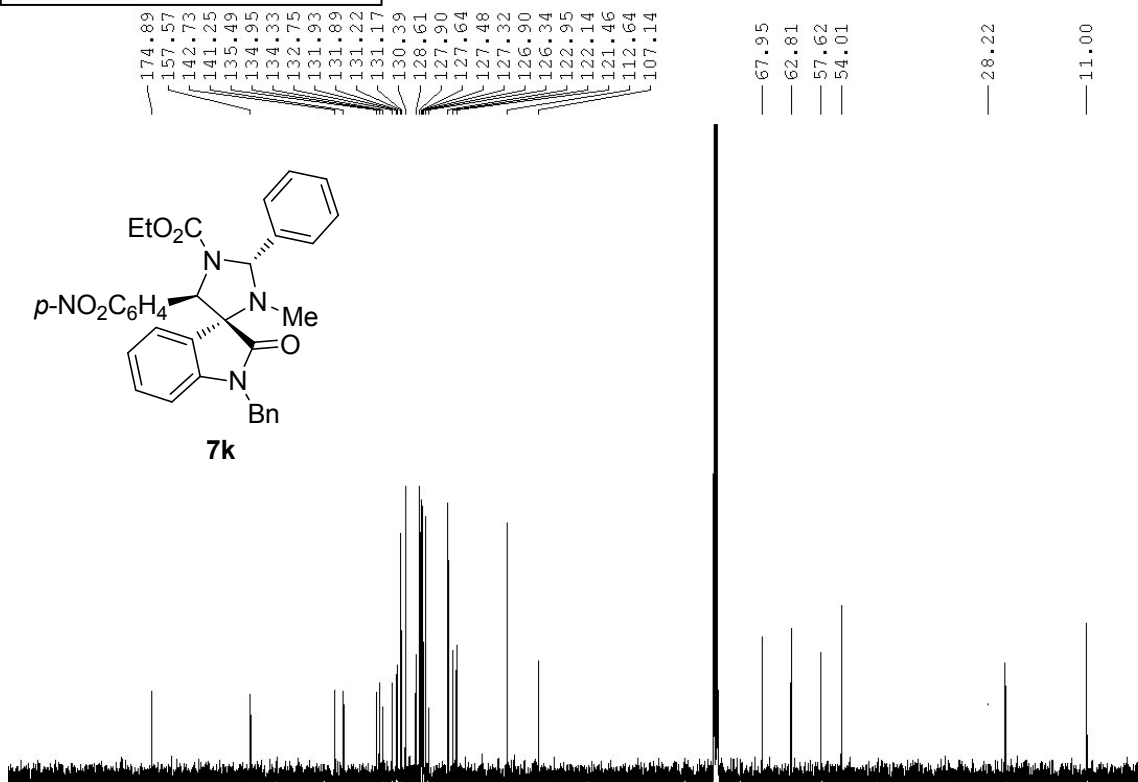
Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Not active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source



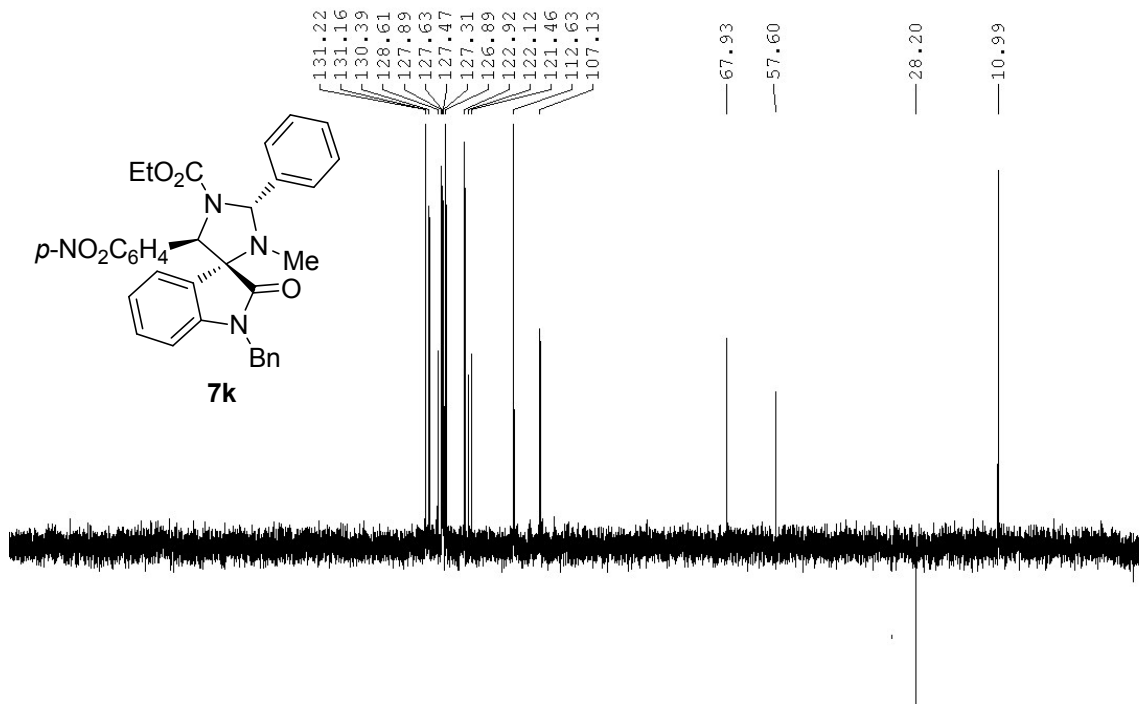
¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)



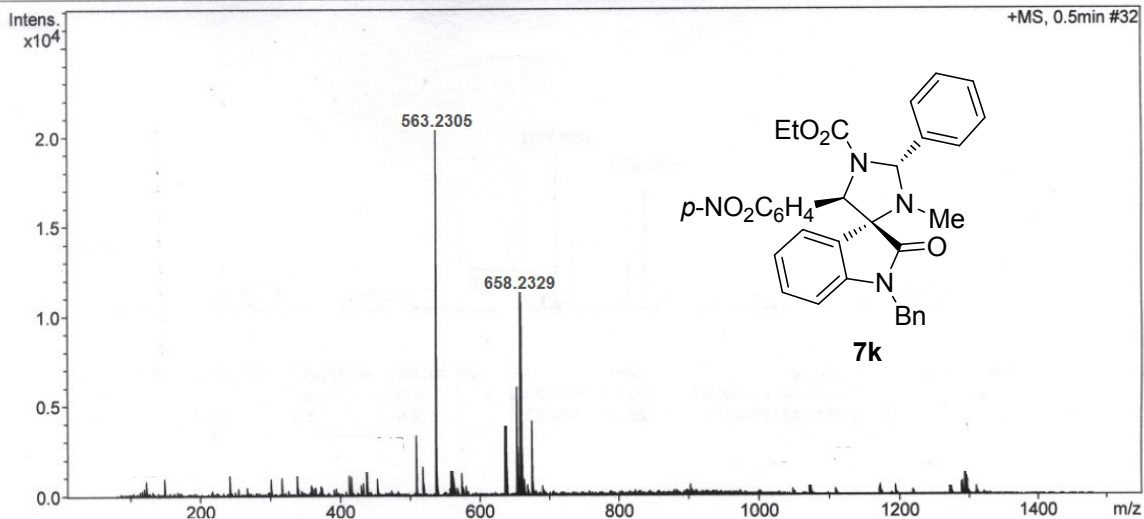
¹³C DEPT135 NMR (100 MHz, CDCl₃)



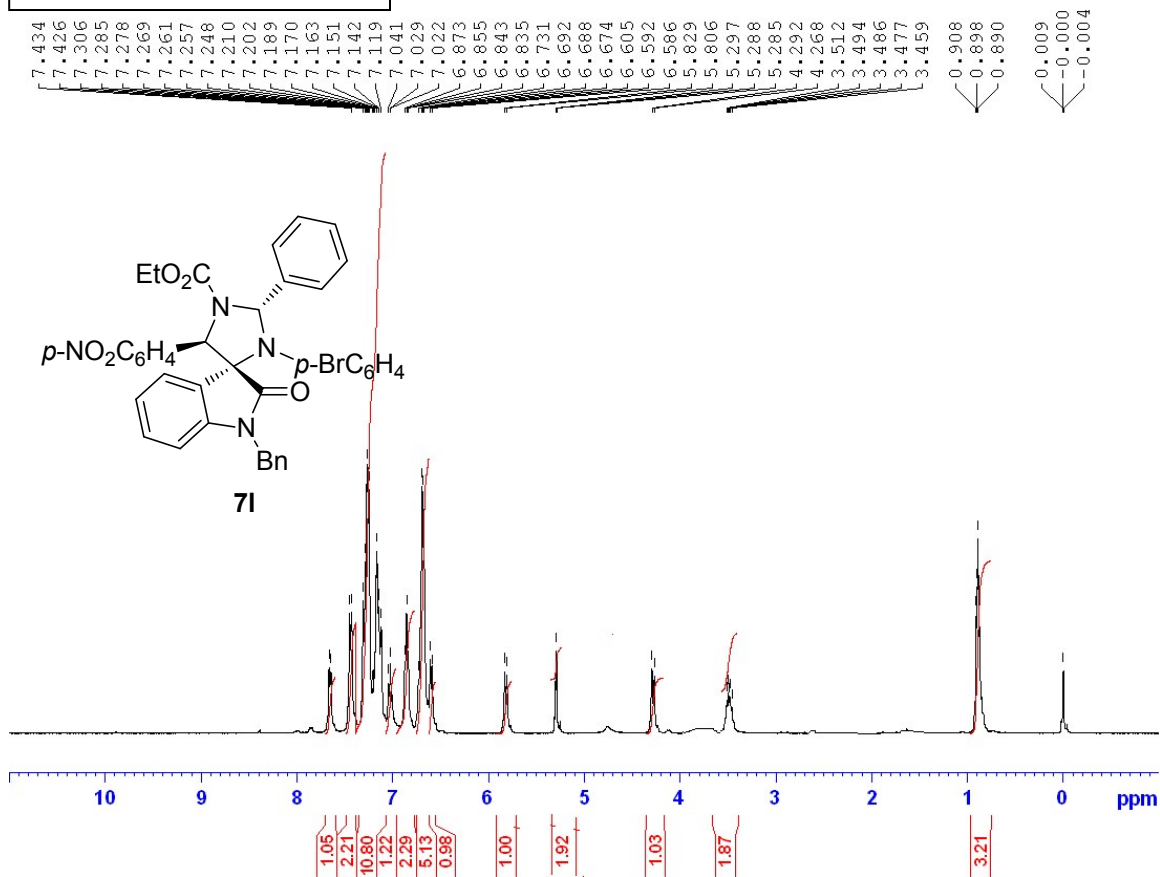
HRMS Spectrum

Acquisition Parameter

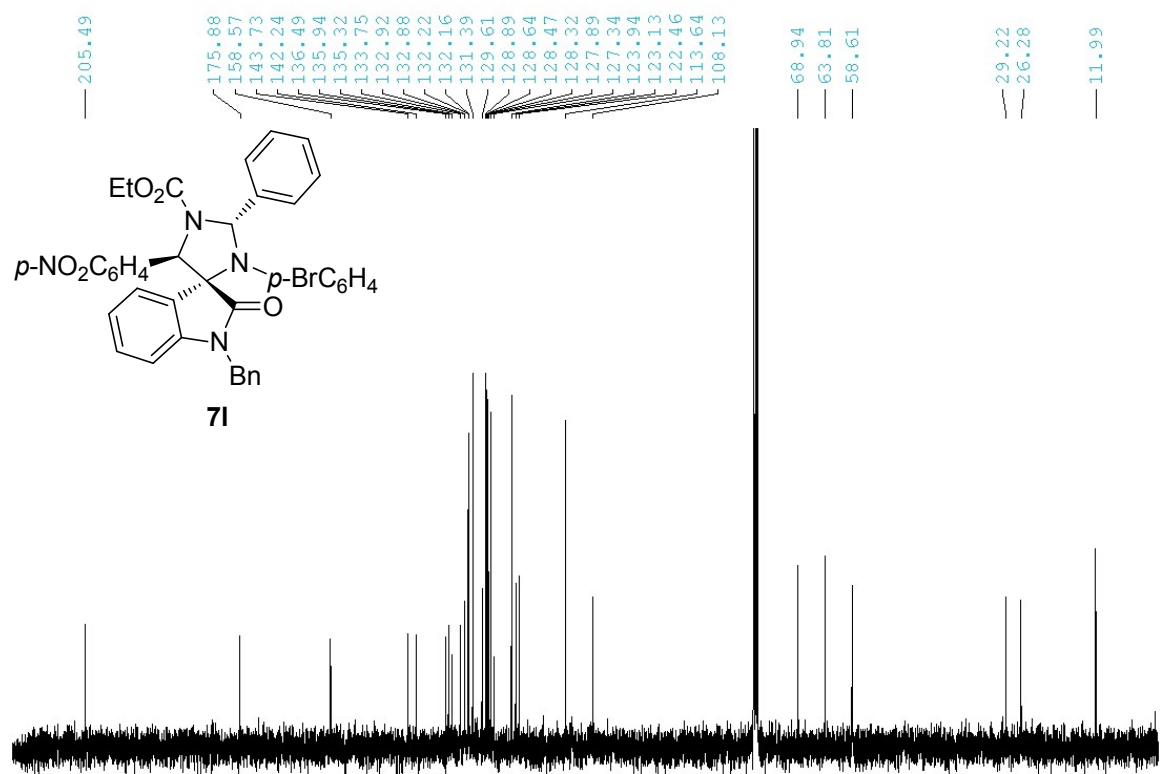
Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Not active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source



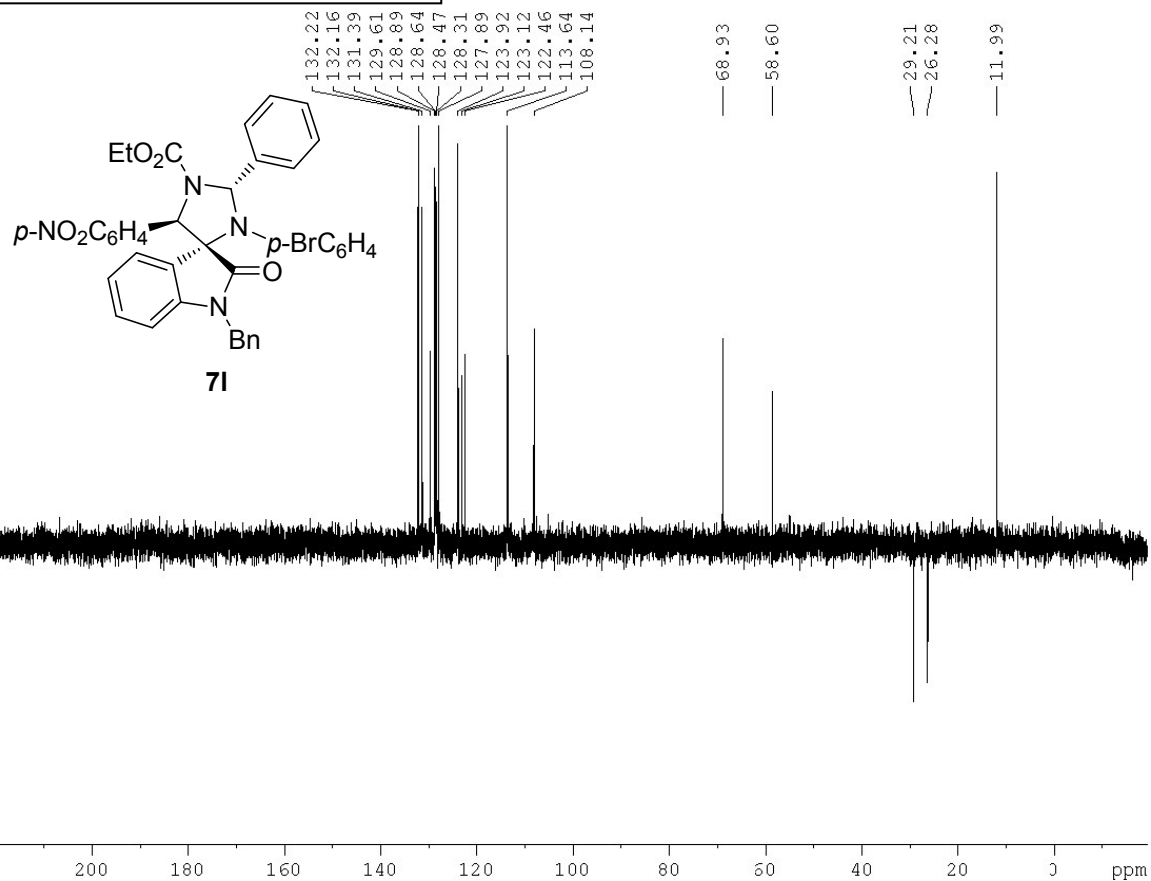
¹H NMR (100 MHz, CDCl₃)



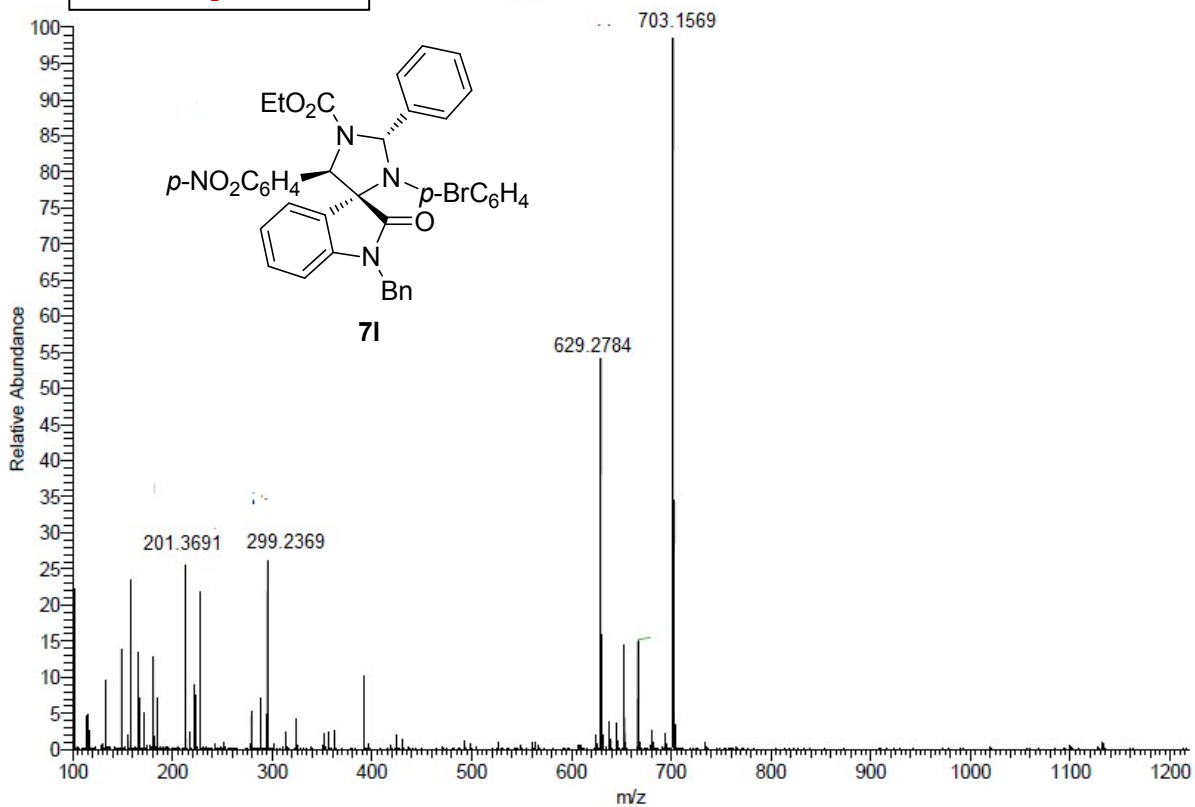
¹³C NMR (100 MHz, CDCl₃)



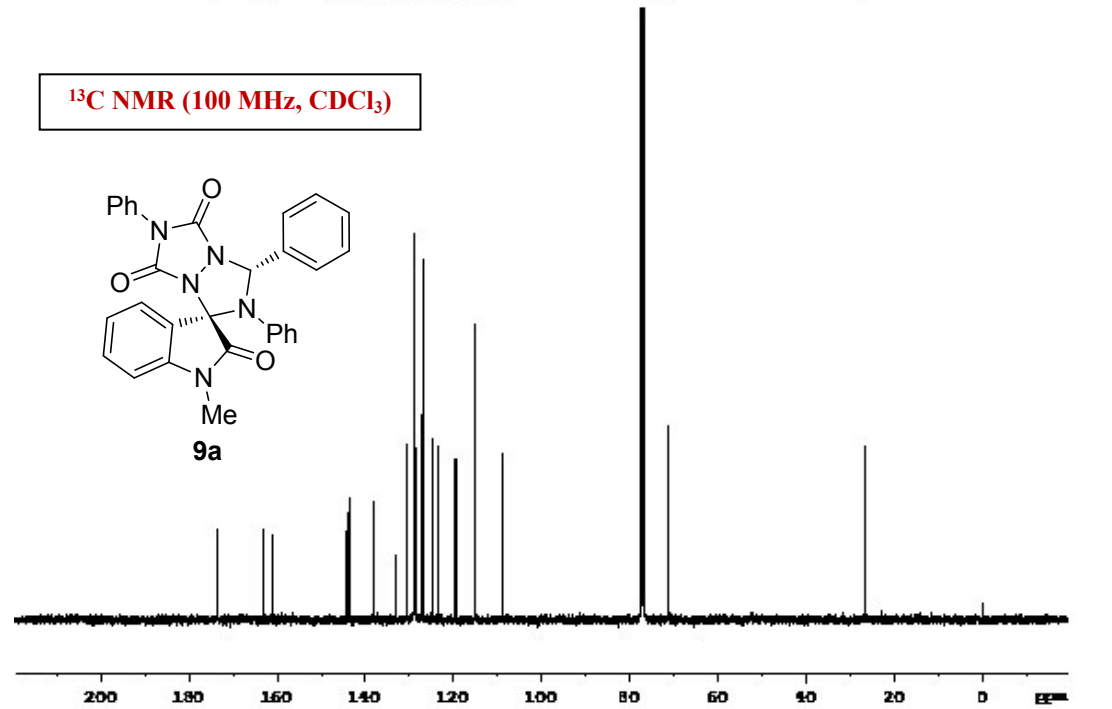
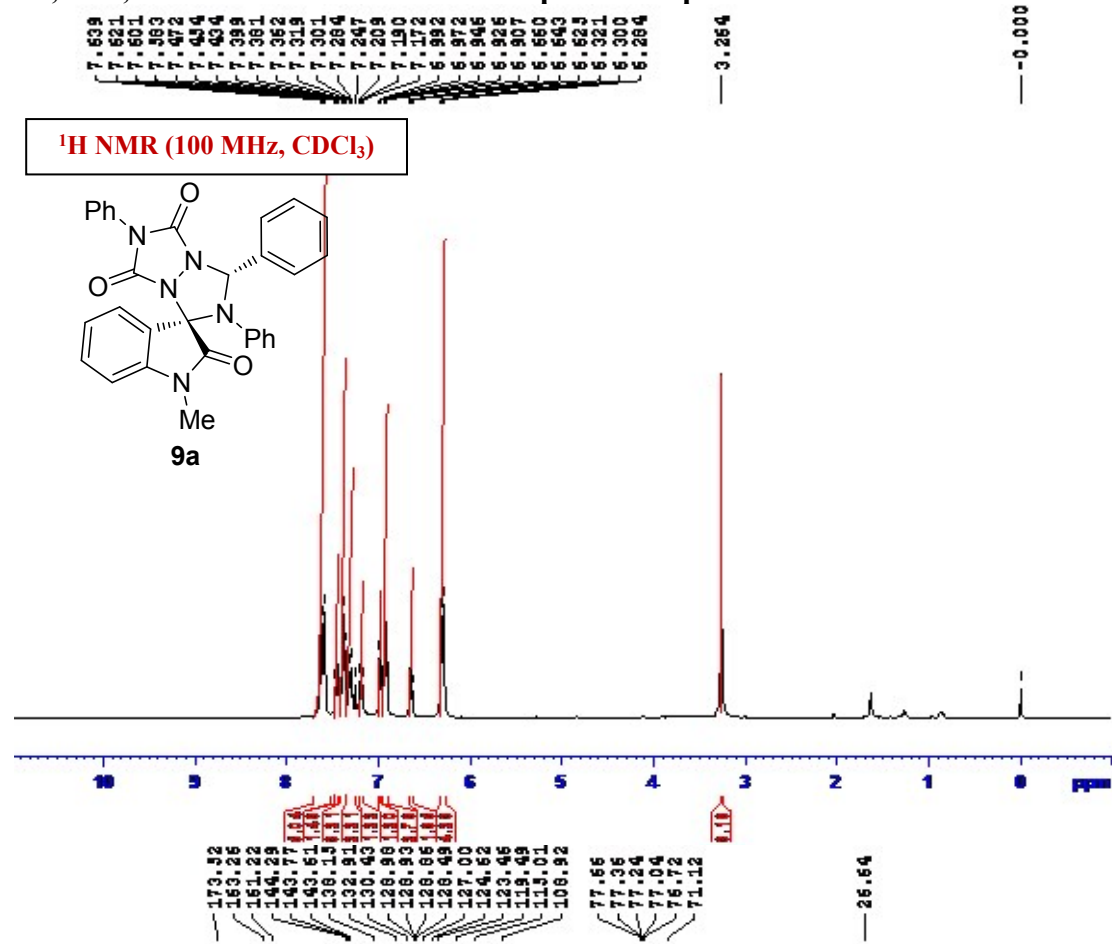
¹³C DEPT135 NMR (100 MHz, CDCl₃)



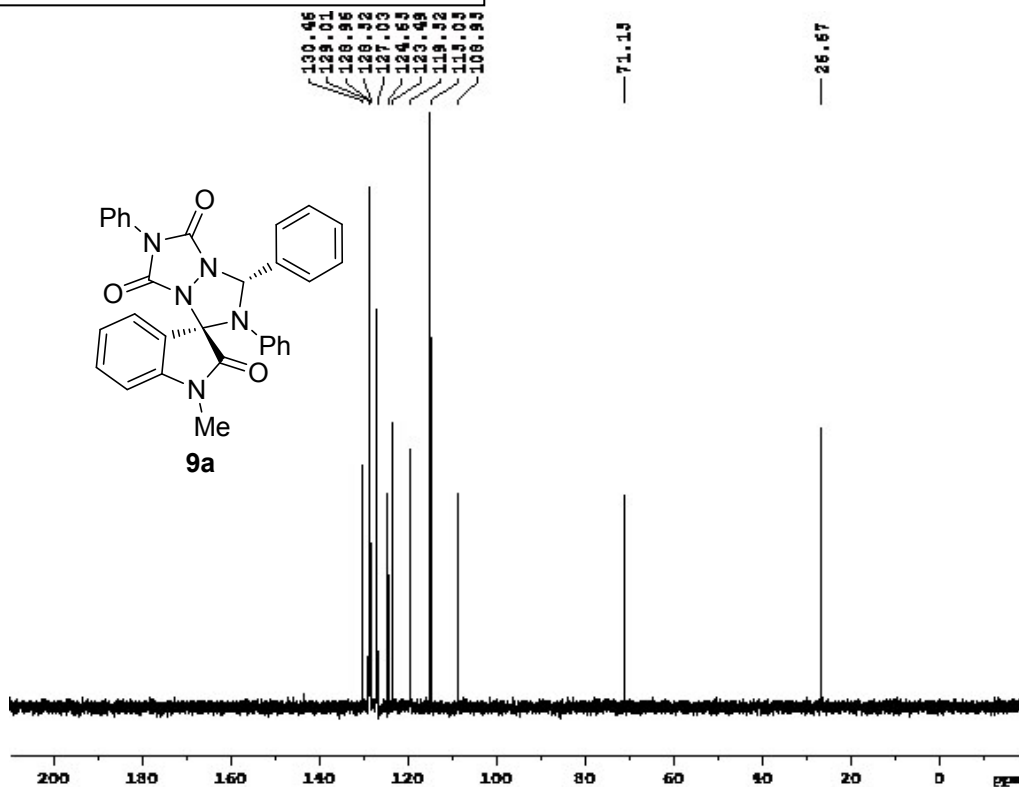
HRMS Spectrum



¹H, ¹³C, DEPT135 NMR and HRMS spectra of spiroindolotriazolines



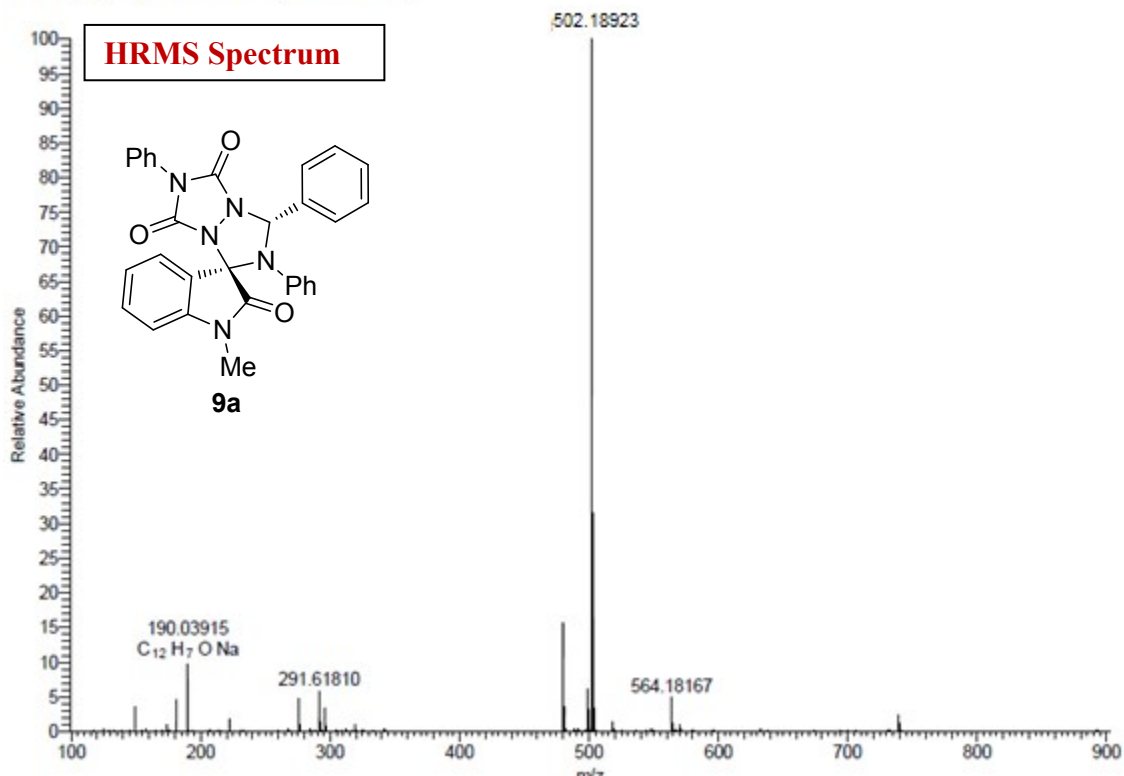
¹³C DEPT135 NMR (100 MHz, CDCl₃)

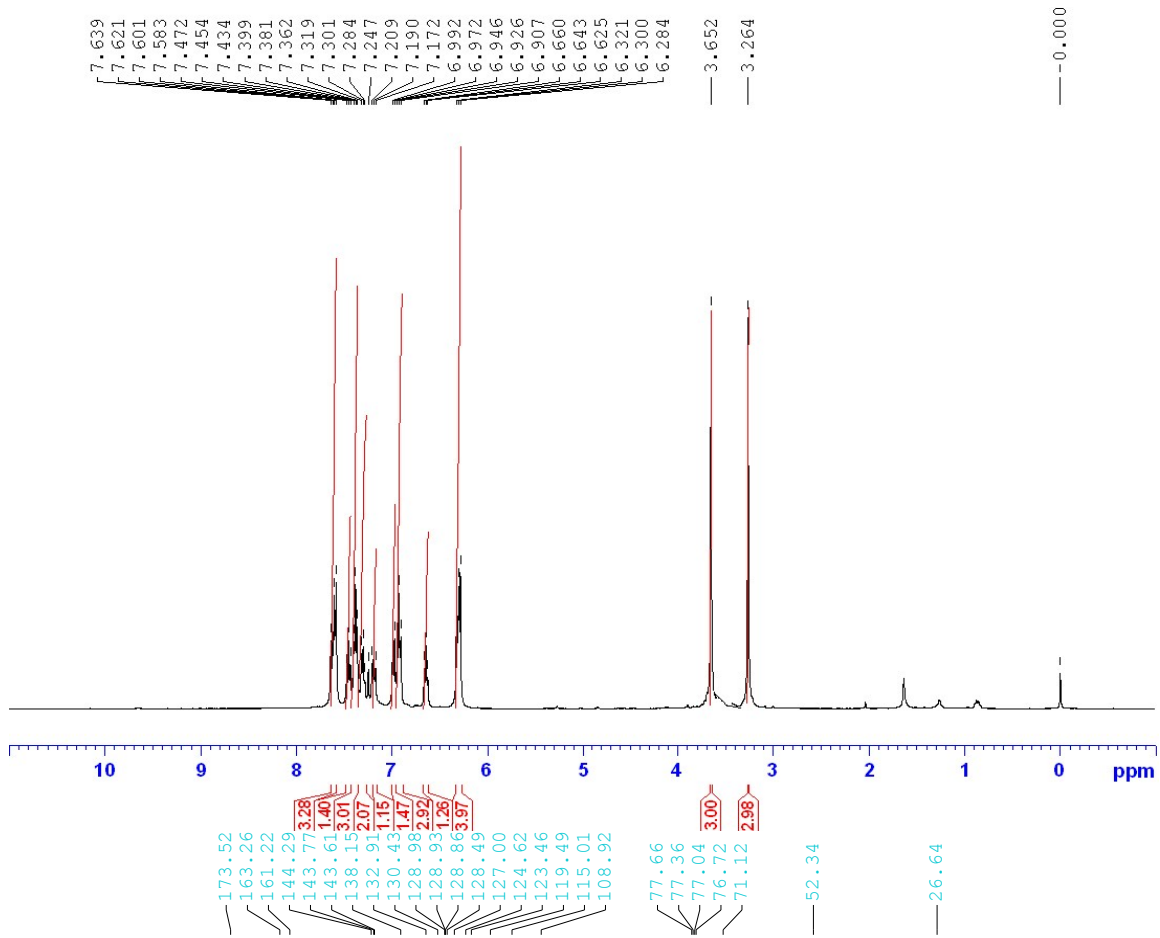


C:\xcalbur\... \SMSGK-293

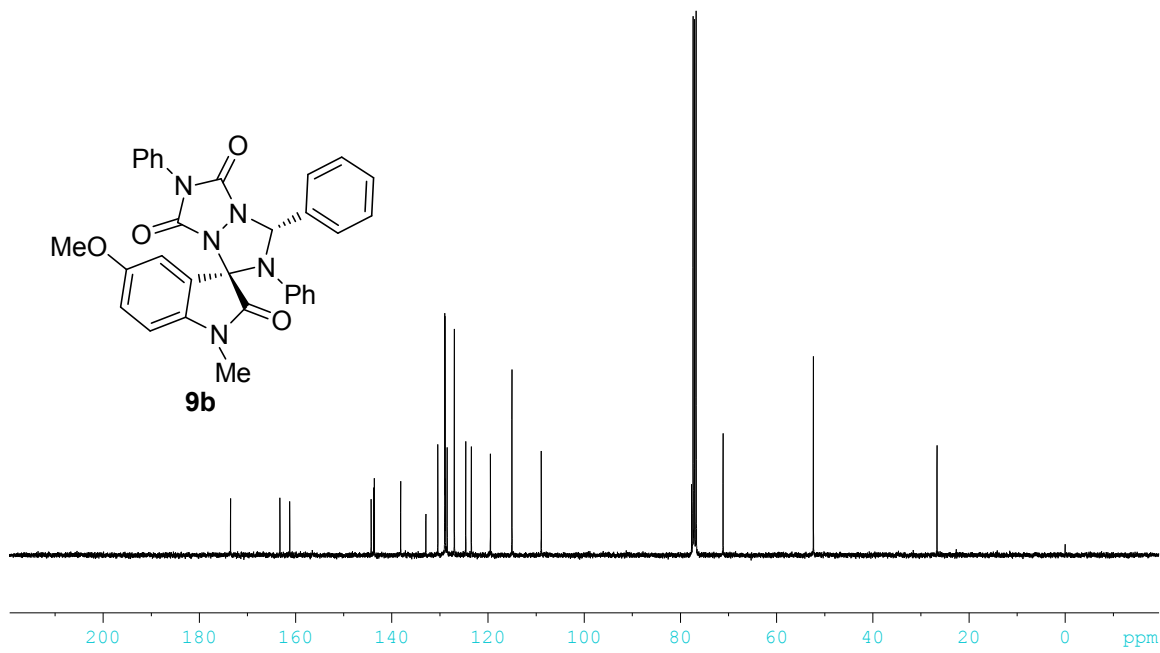
15-01-2014 14:47:22

SMSGK-293 #61-73 RT: 0.87-1.03 AV: 13 SB: 322 0.03-0.84, 1.15-4.95 NL: 8.87E6
T: FTMS (1,1) + p ESI Full lock ms [100.00-2000.00]





¹³C NMR (100 MHz, CDCl₃)



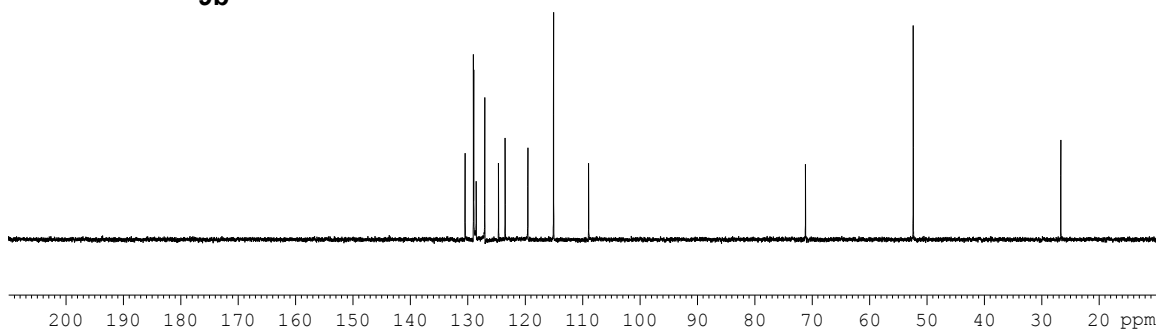
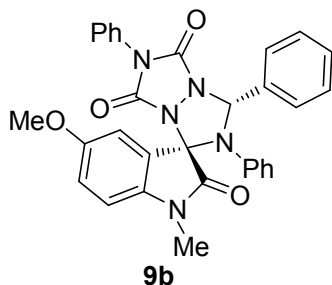
¹³C DEPT135 NMR (100 MHz, CDCl₃)

130.46
129.01
128.96
128.70
128.52
127.03
124.65
123.49
119.52
115.05
108.95

71.15

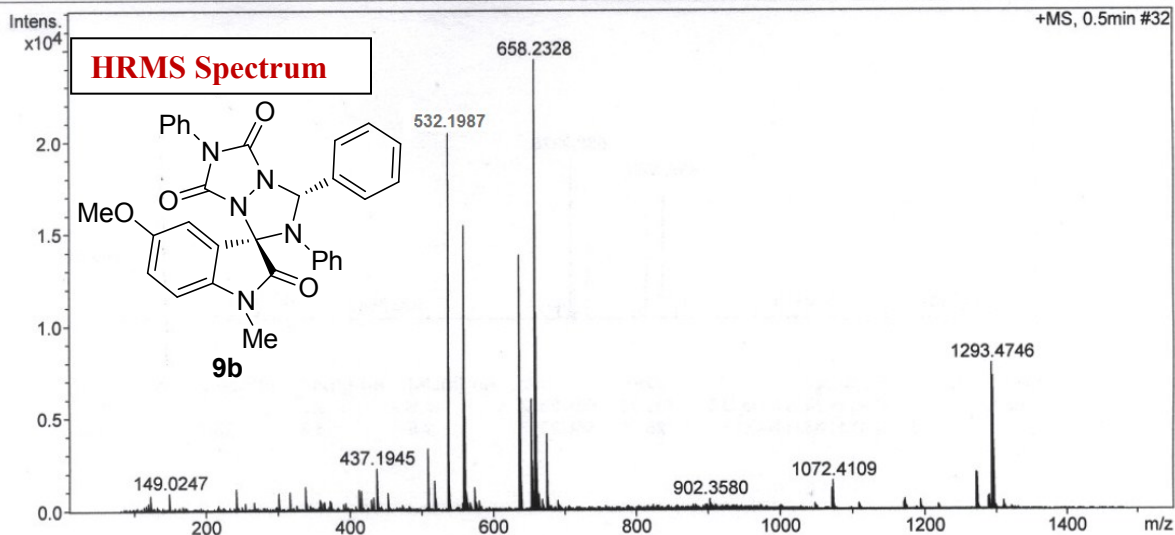
52.37

26.67



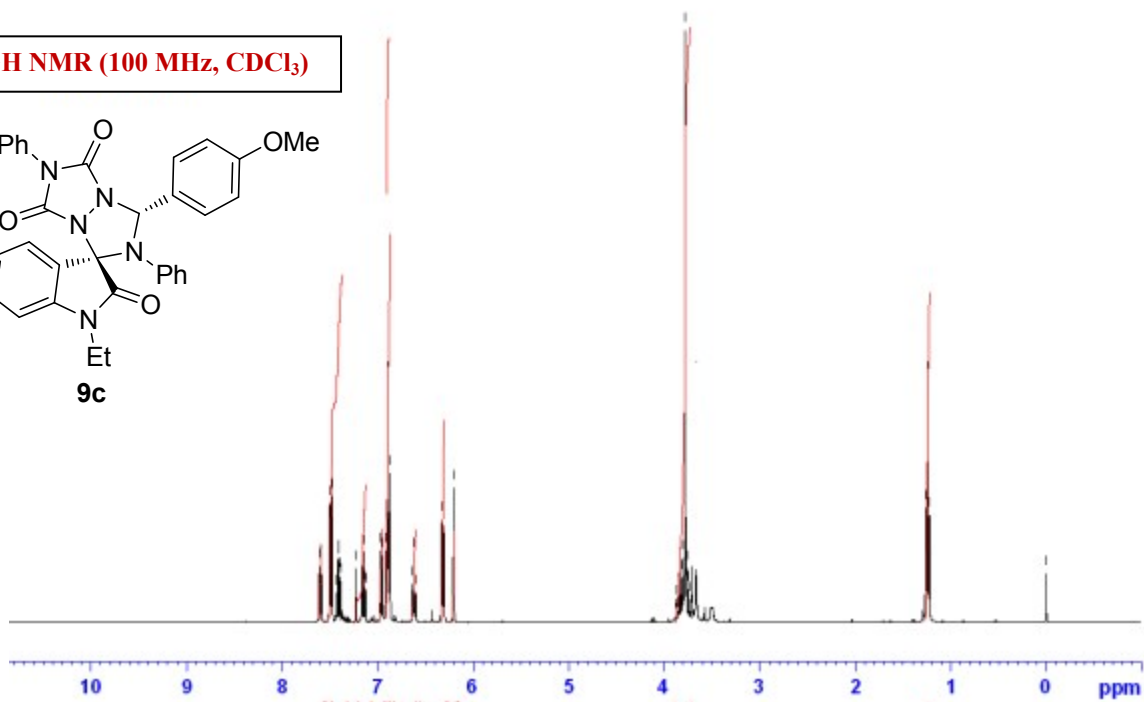
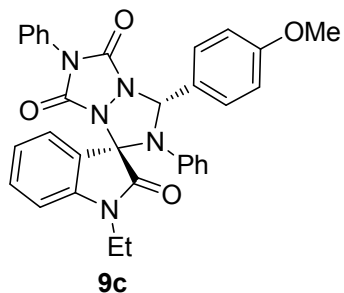
Acquisition Parameter

Source Type	ESI	Ion Polarity	Positive	Set Nebulizer	0.3 Bar
Focus	Not active	Set Capillary	4500 V	Set Dry Heater	180 °C
Scan Begin	50 m/z	Set End Plate Offset	-500 V	Set Dry Gas	4.0 l/min
Scan End	1500 m/z	Set Collision Cell RF	100.0 Vpp	Set Divert Valve	Source

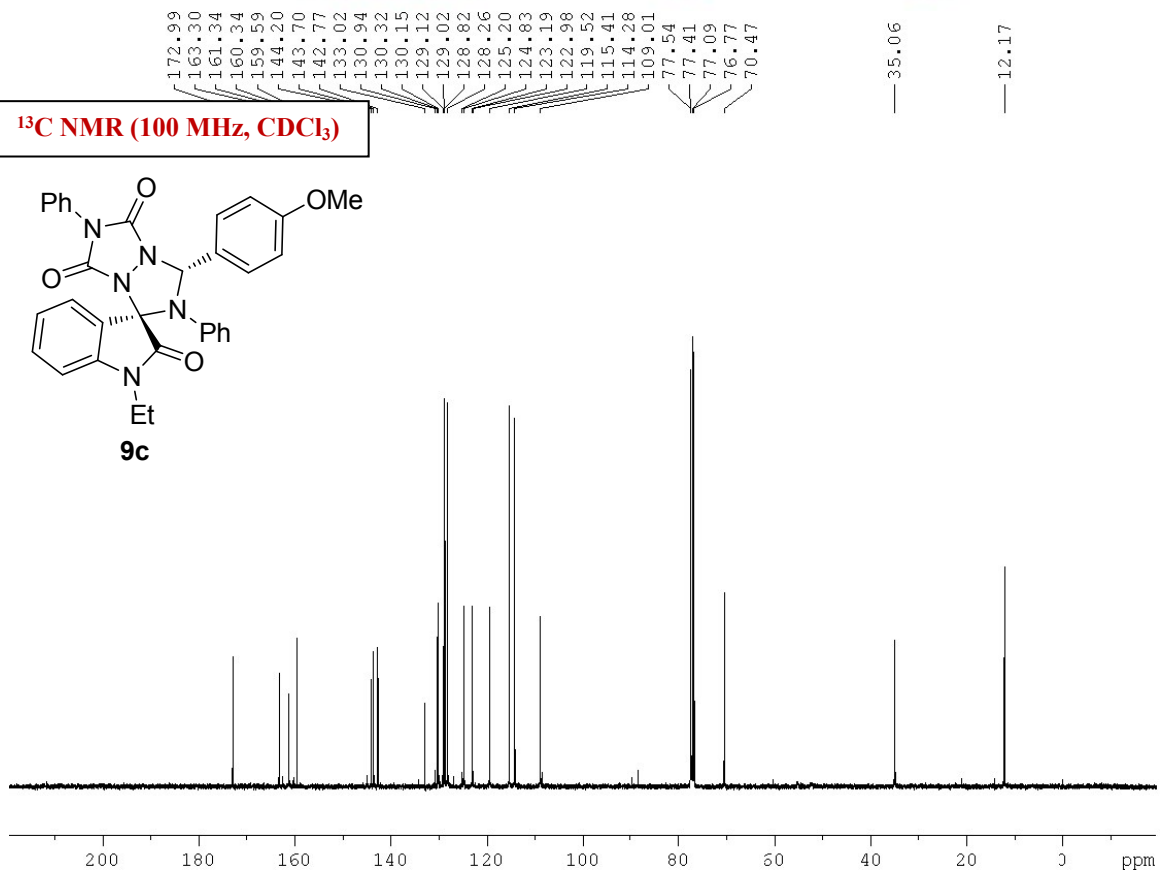
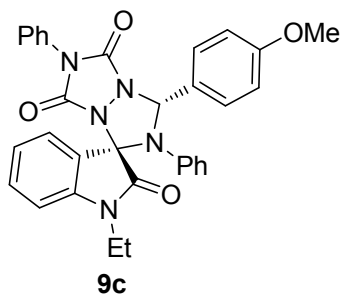


7.629
7.612
7.521
7.499
7.453
7.450
7.434
7.414
7.412
7.249
7.189
7.187
7.170
7.150
6.992
6.972
6.937
6.915
6.896
6.893
6.657
6.639
6.621
6.349
6.328
6.226
3.885
3.867
3.849
3.831
3.813
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3.729
3.710
1.260
1.242
1.224
-0.000

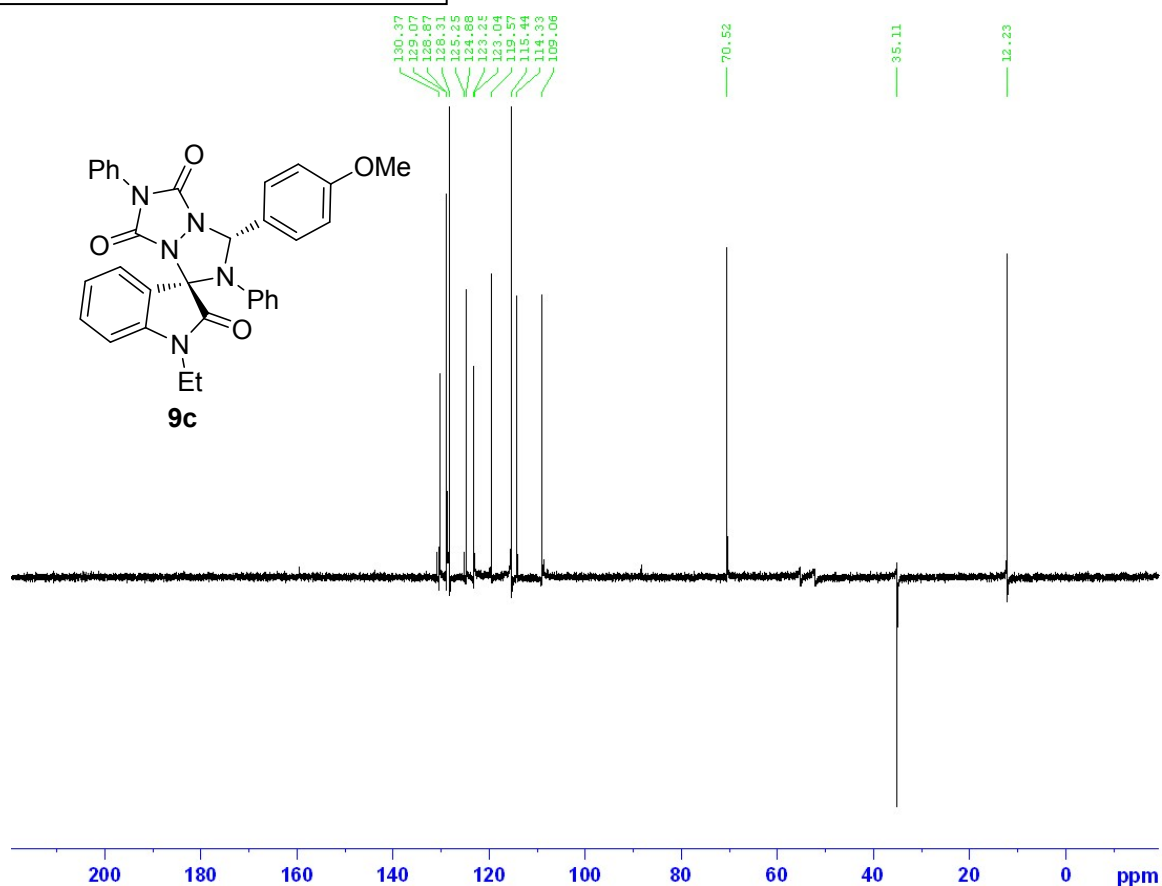
¹H NMR (100 MHz, CDCl₃)



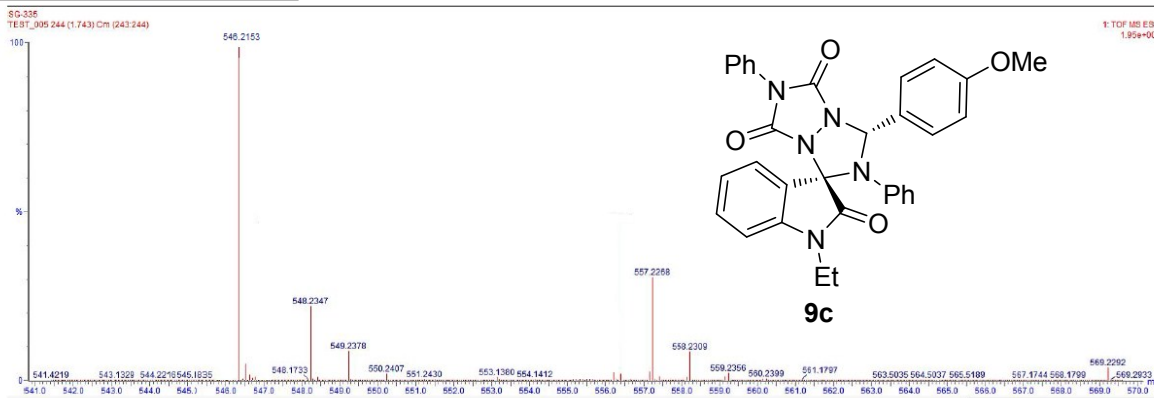
¹³C NMR (100 MHz, CDCl₃)



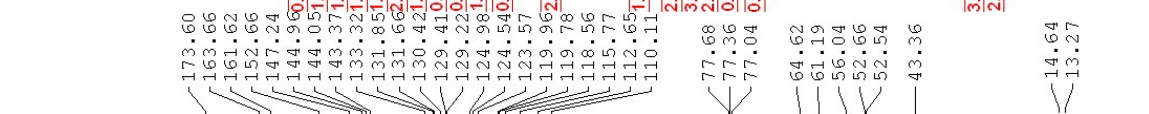
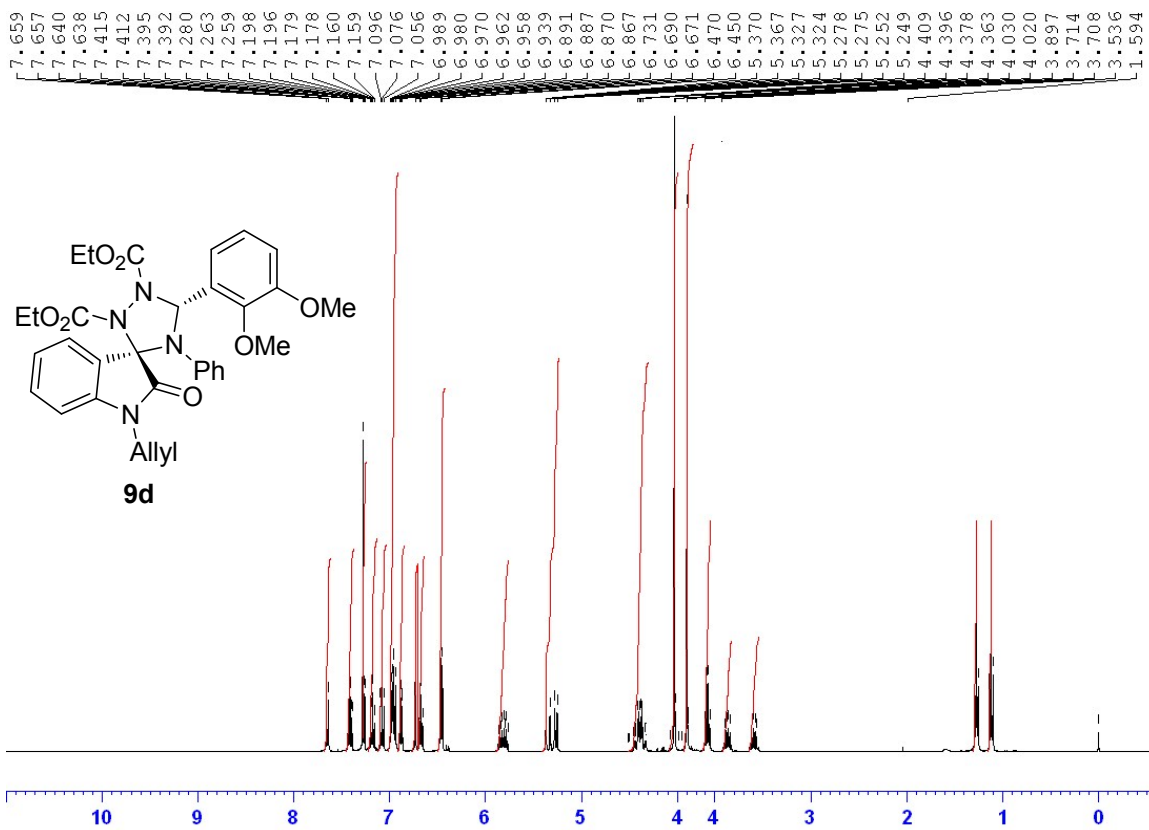
¹³C DEPT135 NMR (100 MHz, CDCl₃)



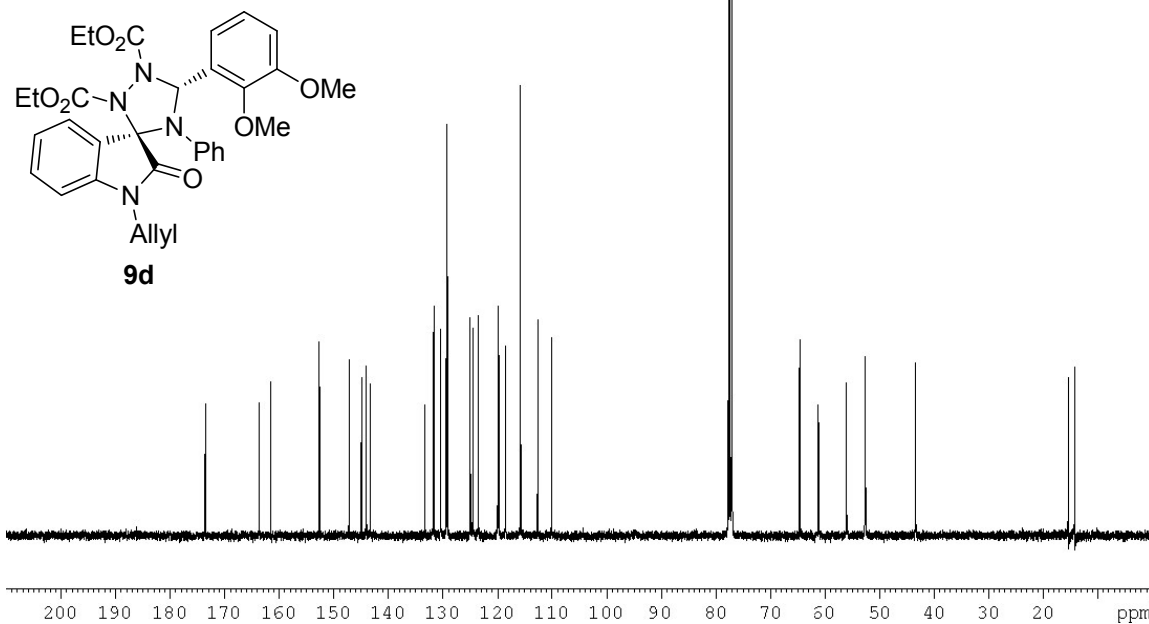
HRMS Spectrum



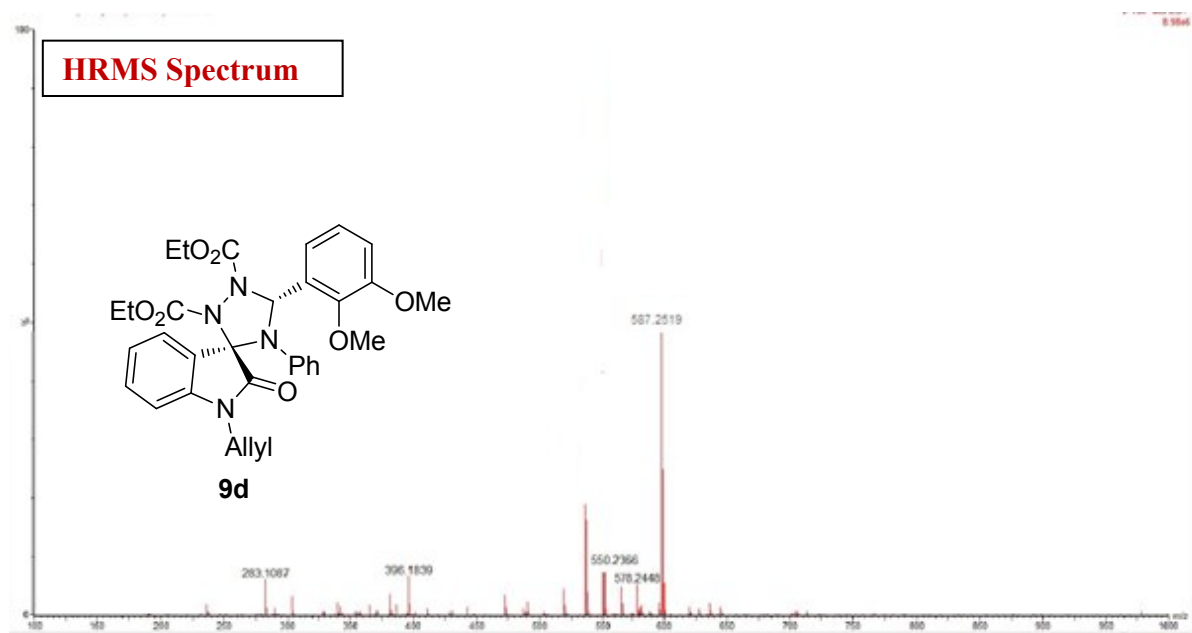
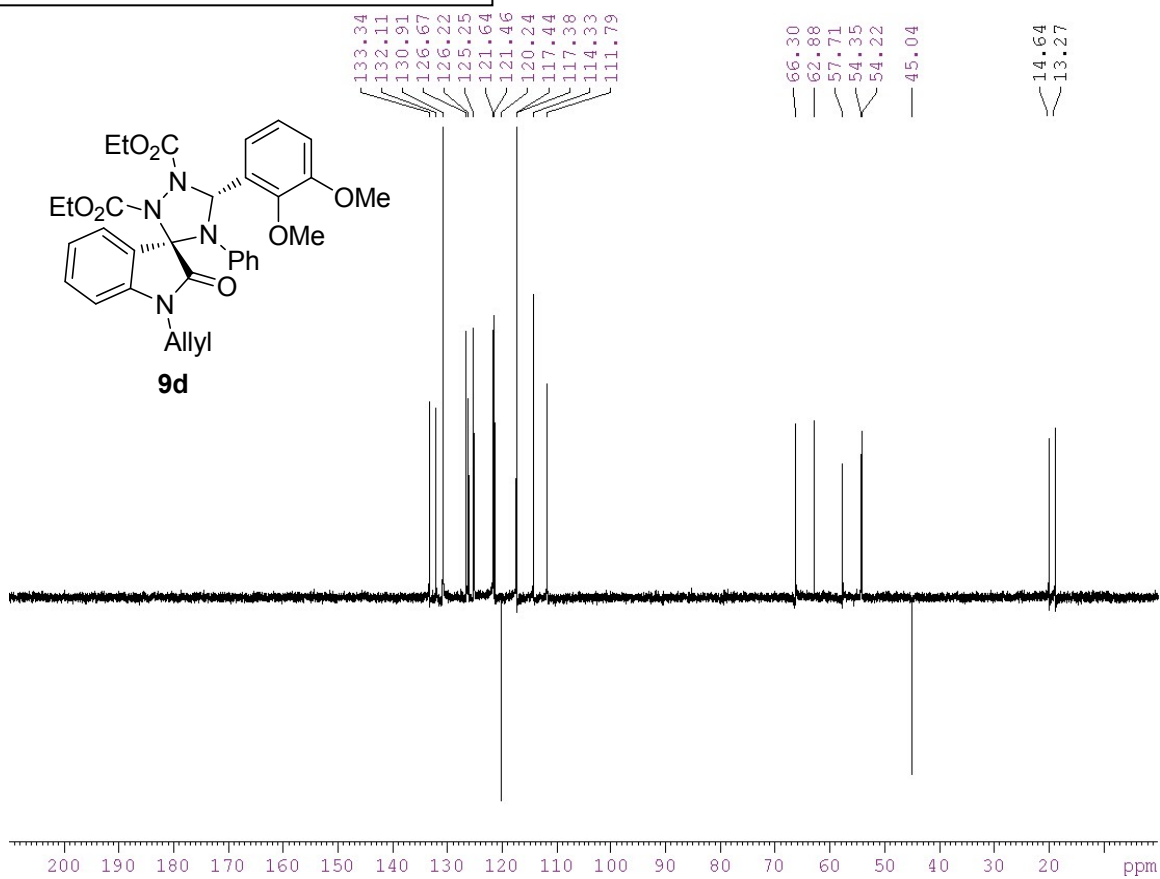
¹H NMR (100 MHz, CDCl₃)



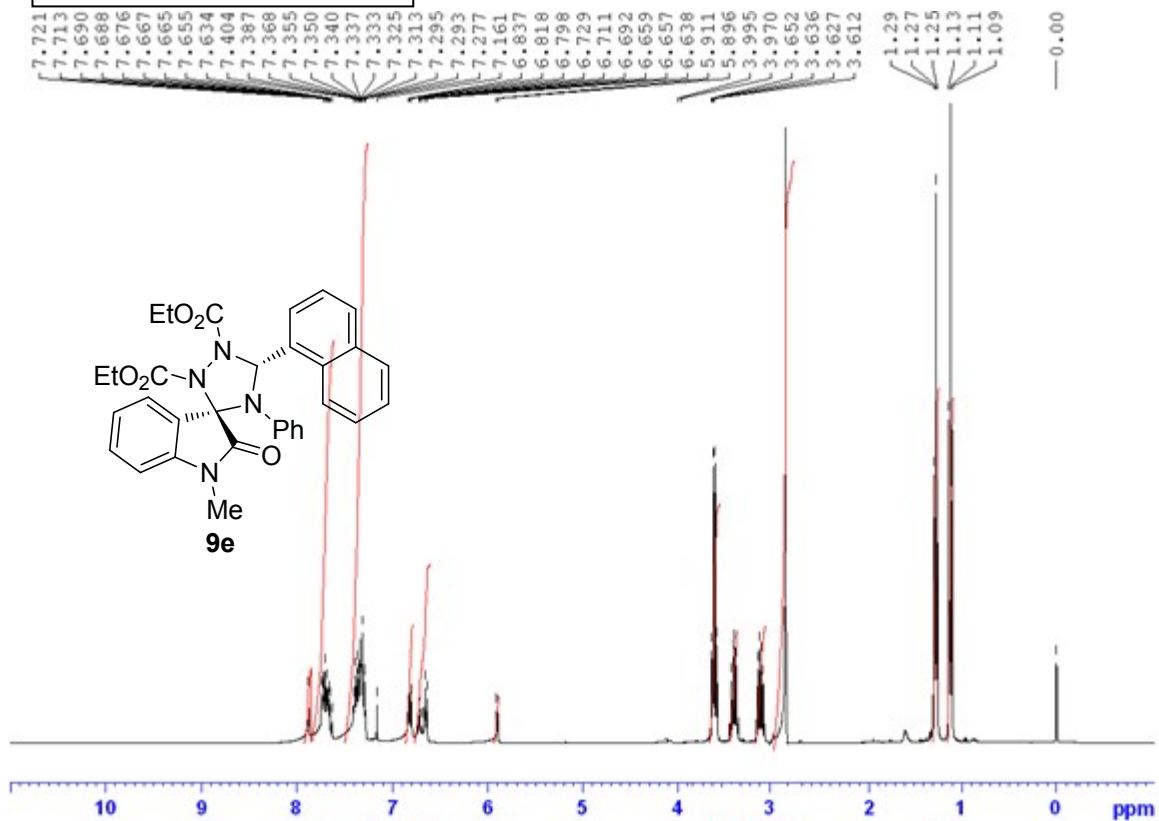
¹³C NMR (100 MHz, CDCl₃)



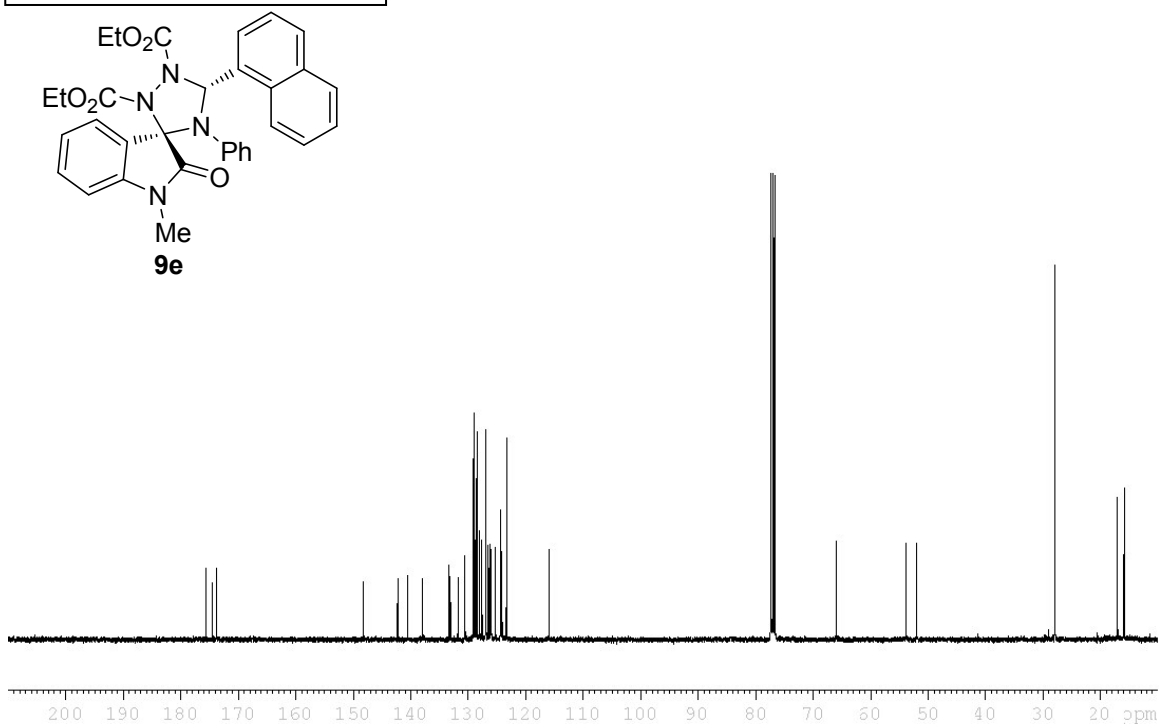
¹³C DEPT135 NMR (100 MHz, CDCl₃)



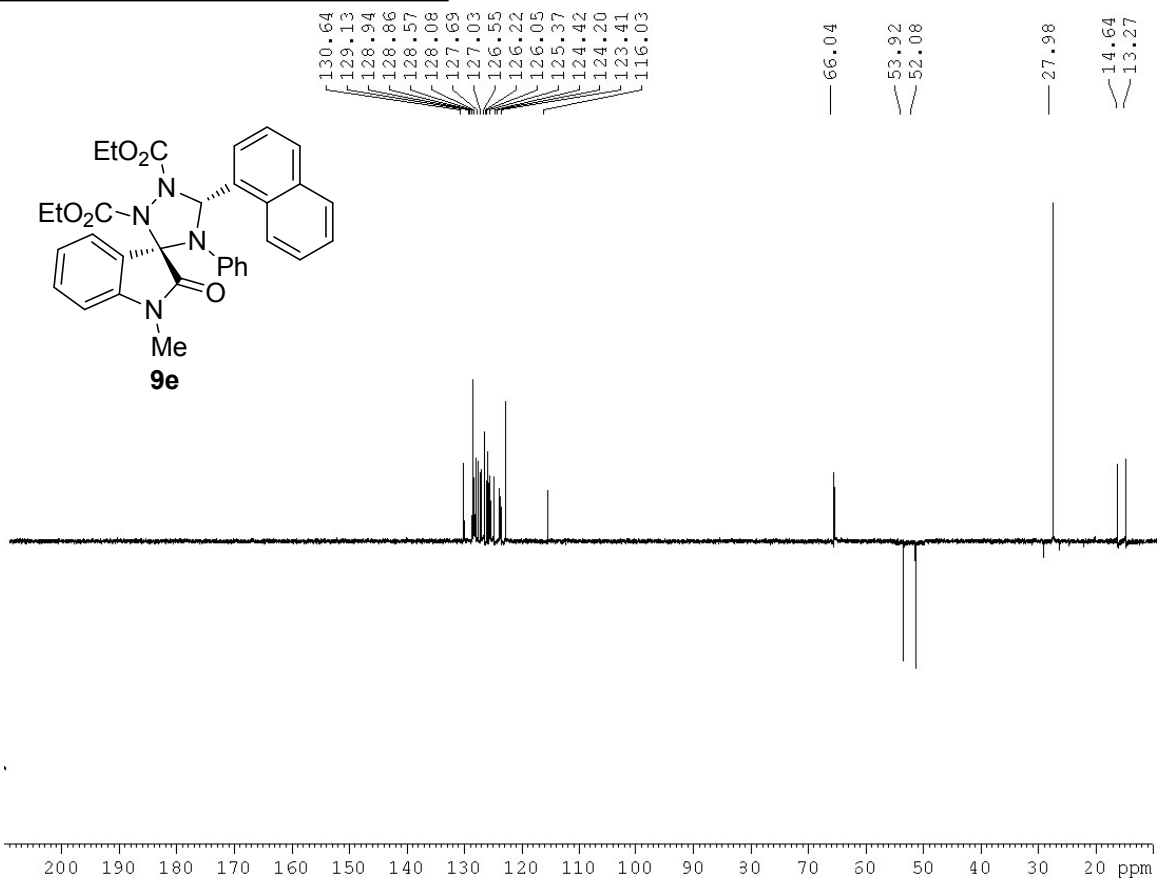
¹H NMR (100 MHz, CDCl₃)



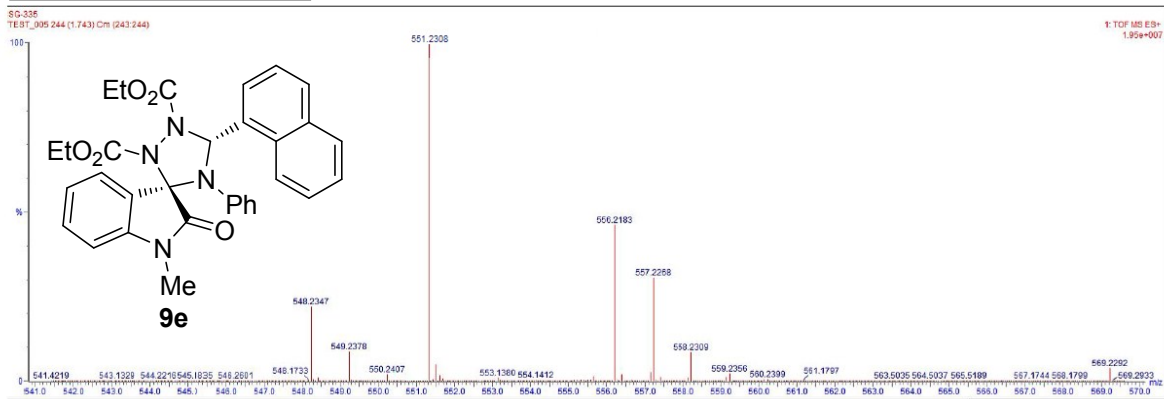
¹³C NMR (100 MHz, CDCl₃)



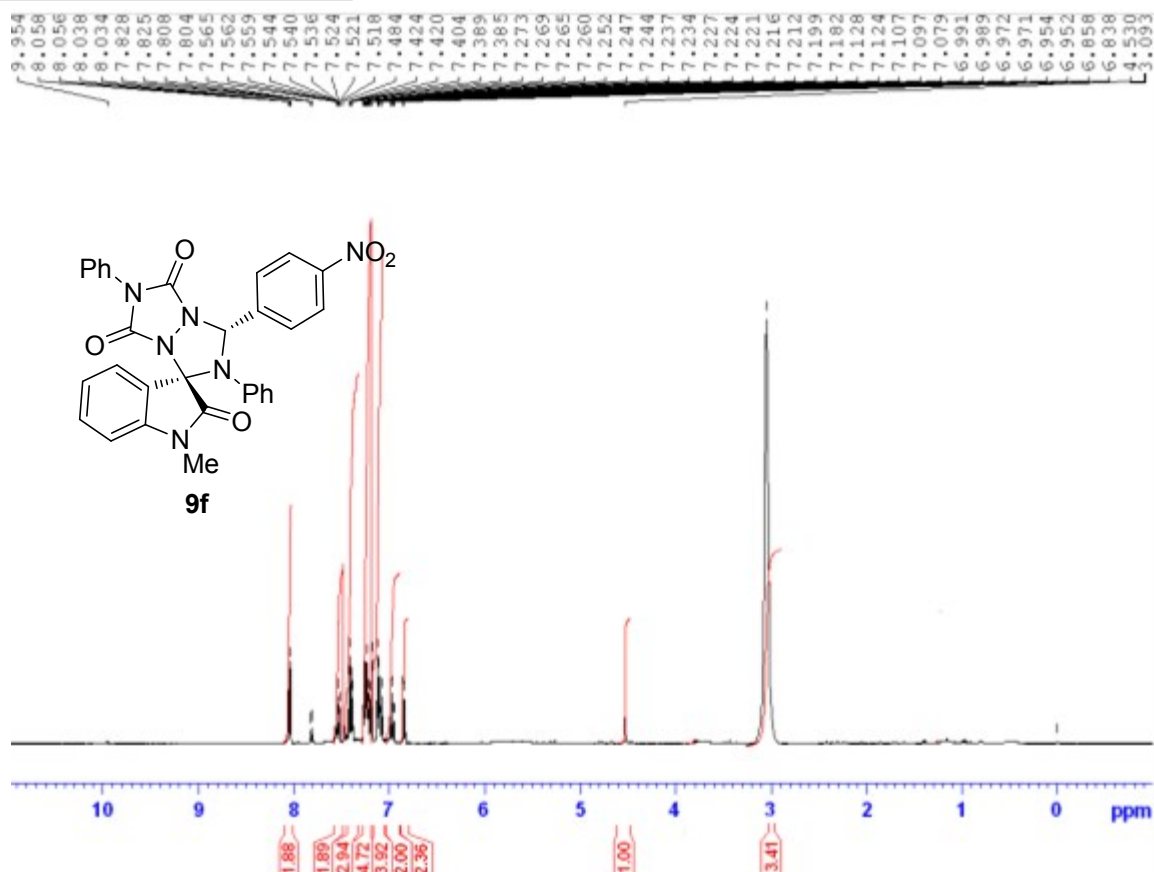
¹³C DEPT135 NMR (100 MHz, CDCl₃)



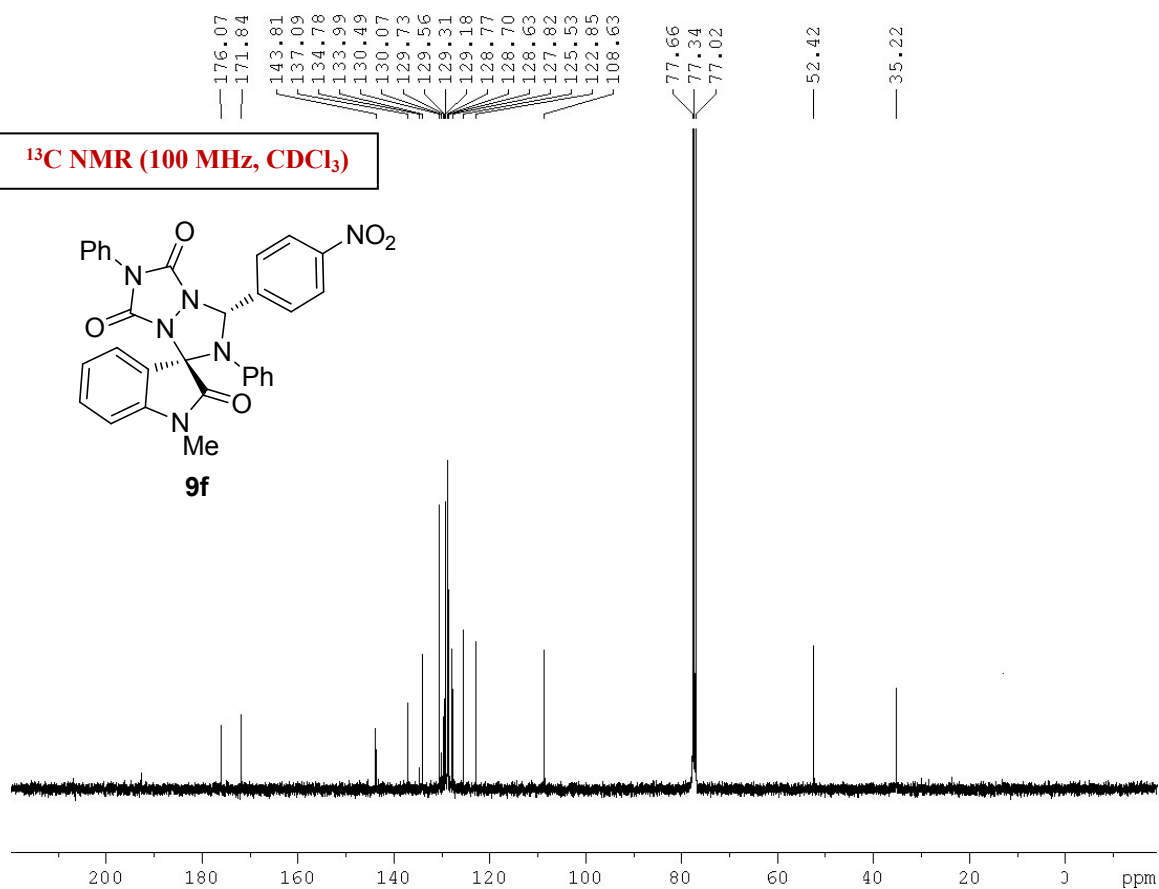
HRMS Spectrum



¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)

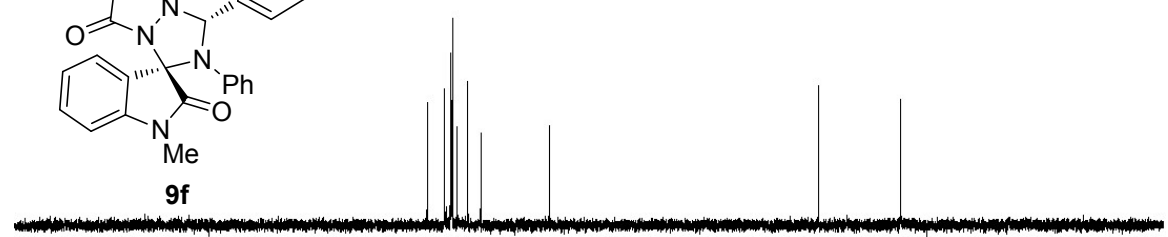
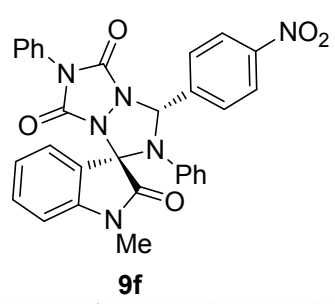


133.99
130.48
129.18
128.77
128.71
128.63
127.82
125.53
122.85
108.64

— 52.42

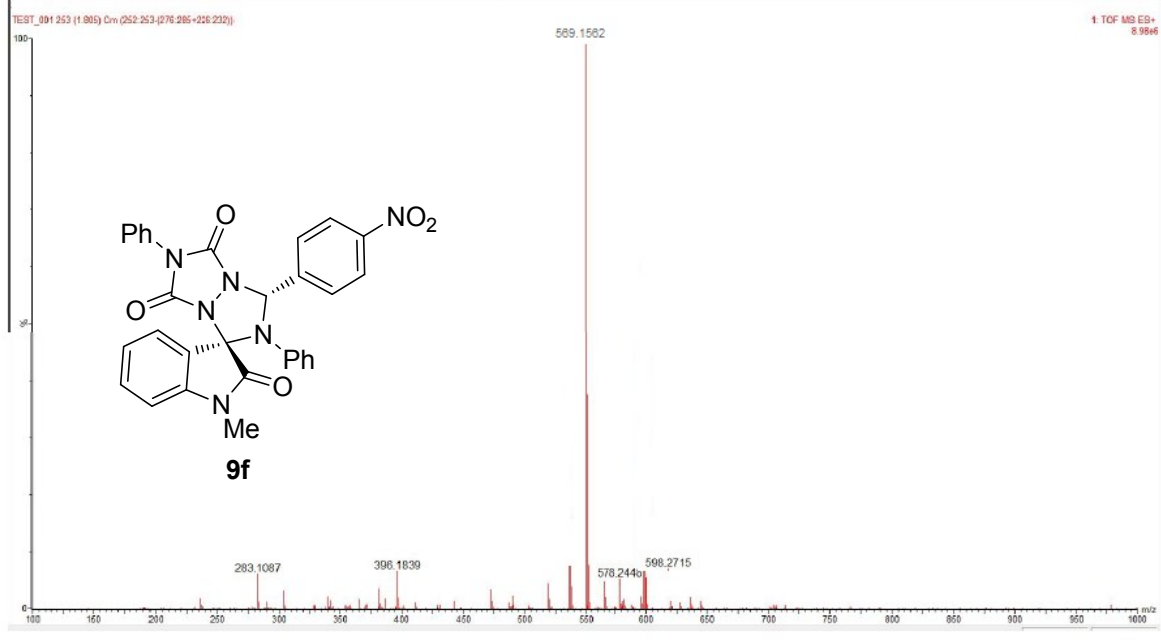
— 35.22

¹³C DEPT135 NMR (100 MHz, CDCl₃)



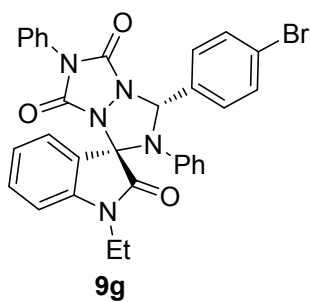
200 180 160 140 120 100 80 60 40 20 0 ppm

HRMS Spectrum



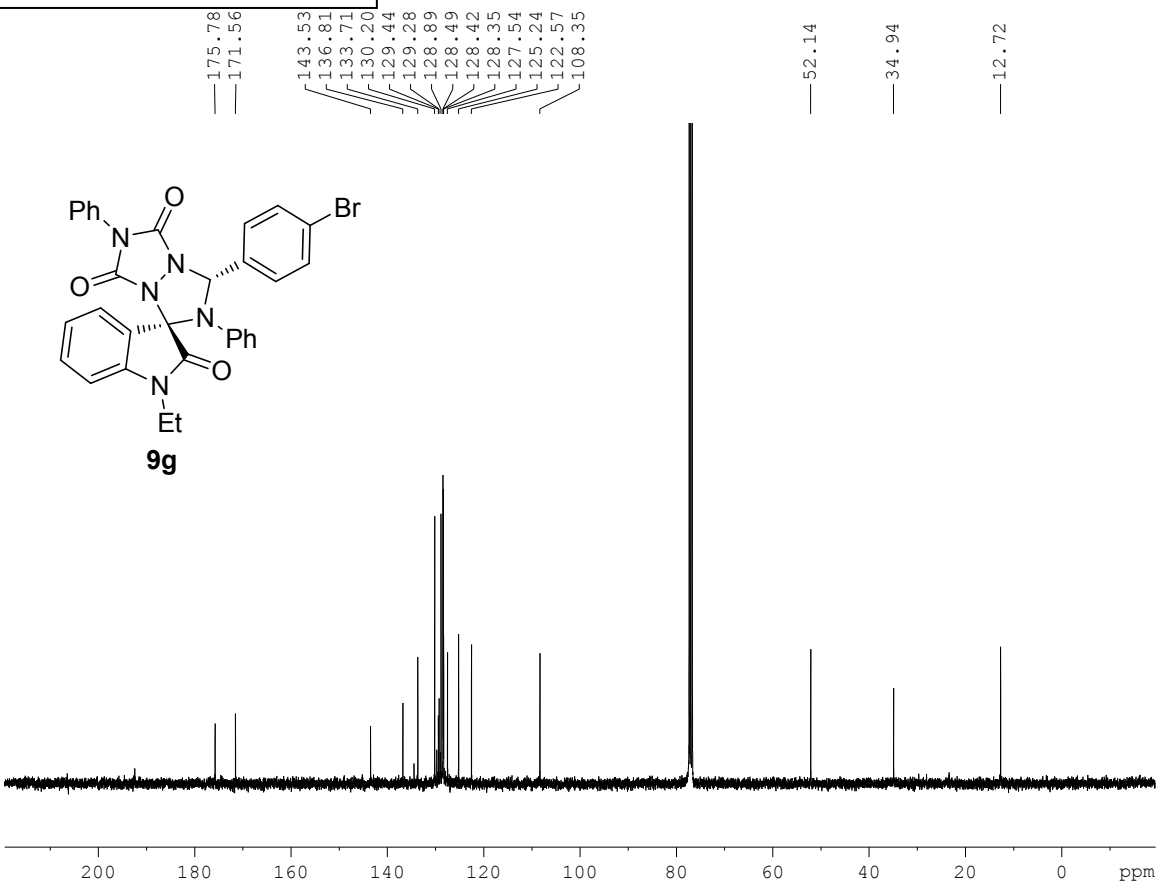
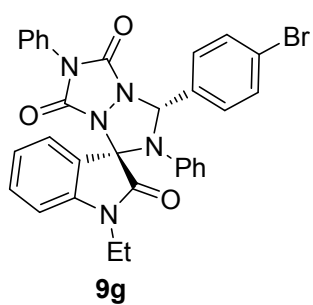
9.954
8.058
8.056
8.038
8.034
7.959
7.944
7.940
7.936
7.924
7.921
7.918
8.24
7.920
7.904
7.989
7.985
7.973
7.969
7.965
7.960
7.952
7.947
7.944
7.937
7.934
7.927
7.924
7.921
7.916
7.912
7.199
7.182
7.128
7.124
7.107
7.097
7.079
6.991
6.989
6.972
6.971
6.954
6.952
6.858
6.838
4.530
3.765
3.747
3.729
3.711
1.245
1.227
1.209

¹H NMR (400 MHz, CDCl₃)

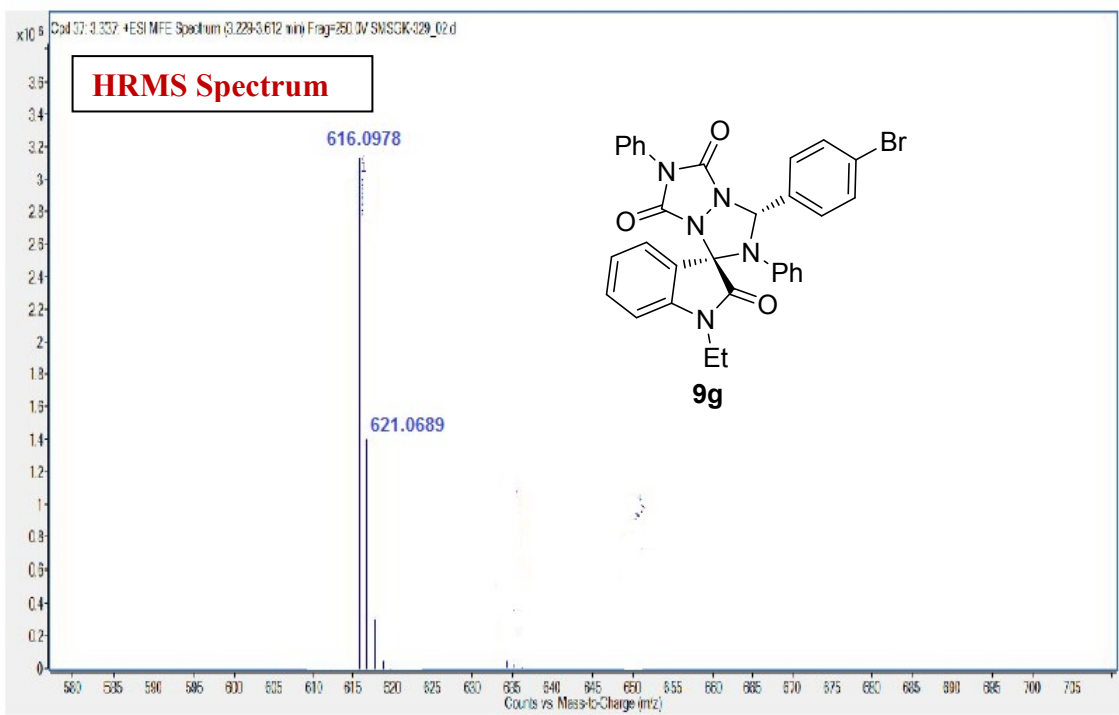
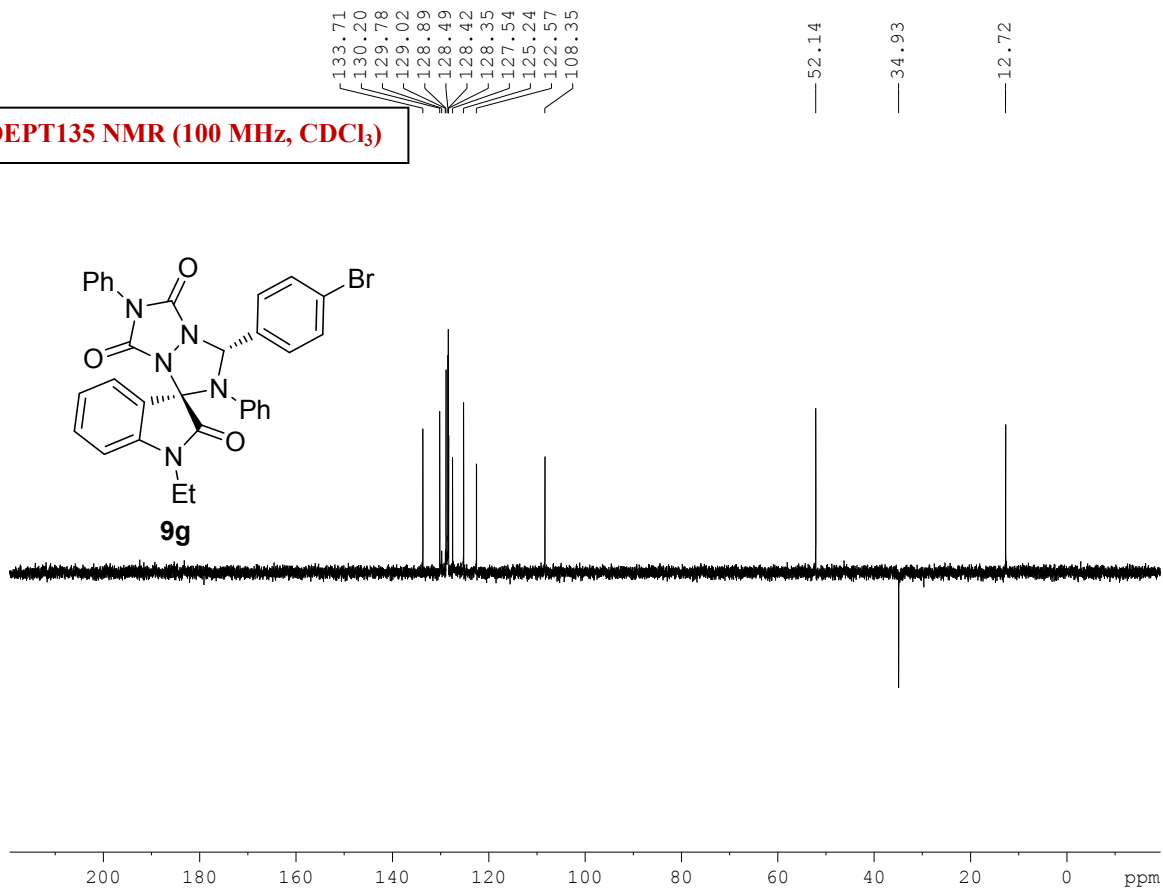


15 14 13 12 11 10 9 8 7 6 5 4 3 2 1 0 -1 ppm

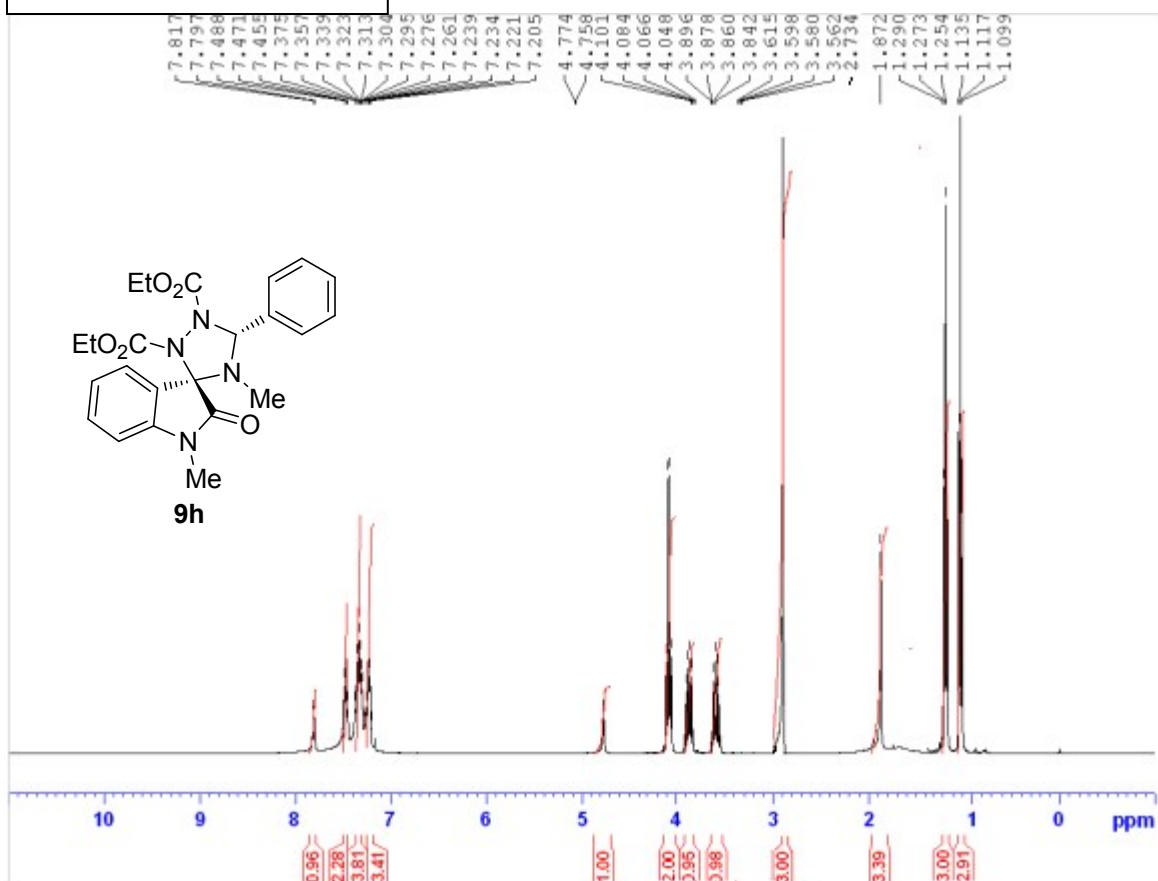
¹³C NMR (100 MHz, CDCl₃)



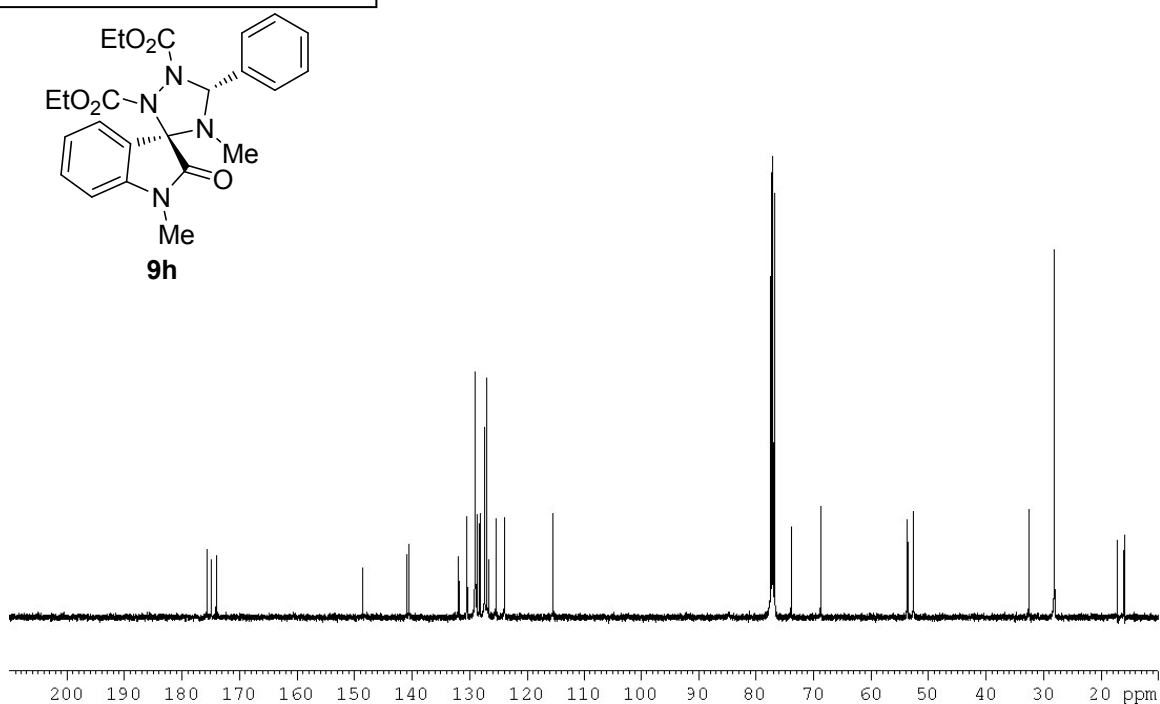
¹³C DEPT135 NMR (100 MHz, CDCl₃)



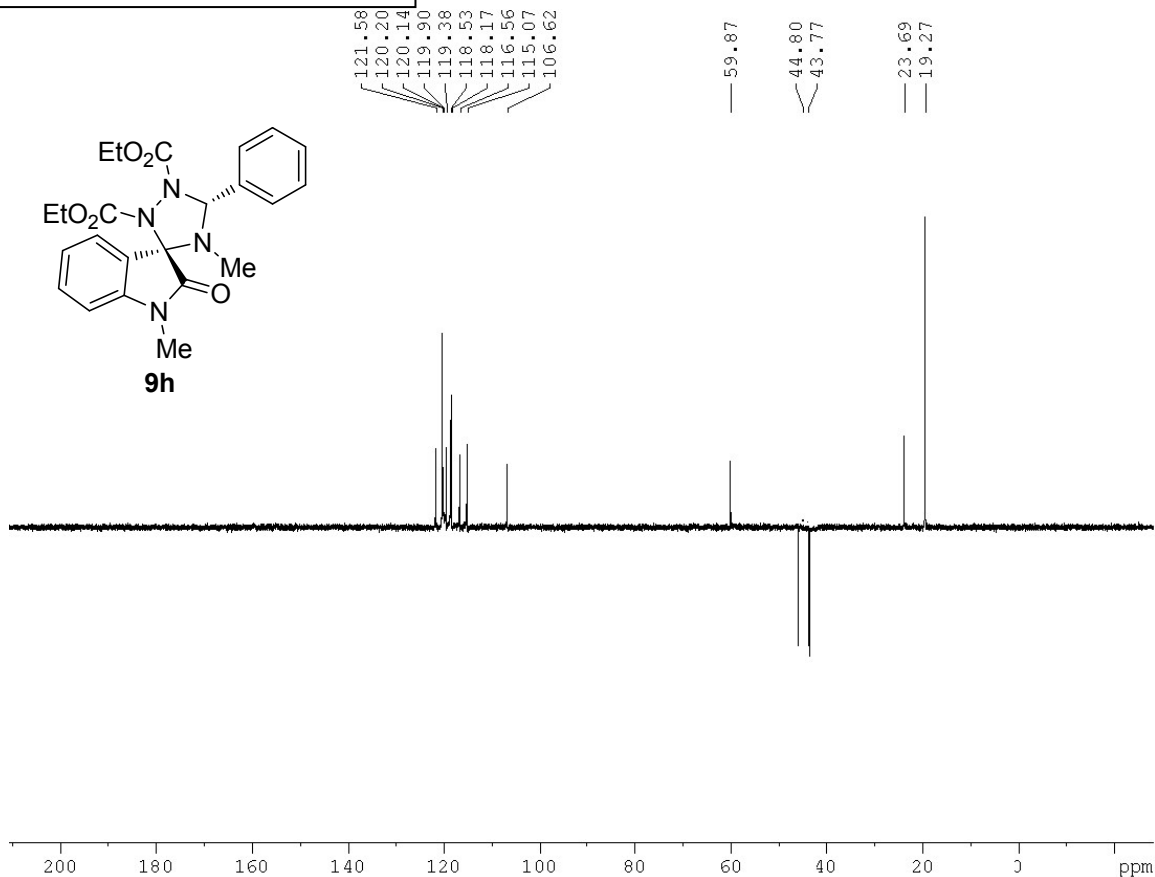
¹H NMR (400 MHz, CDCl₃)



¹³C NMR (100 MHz, CDCl₃)



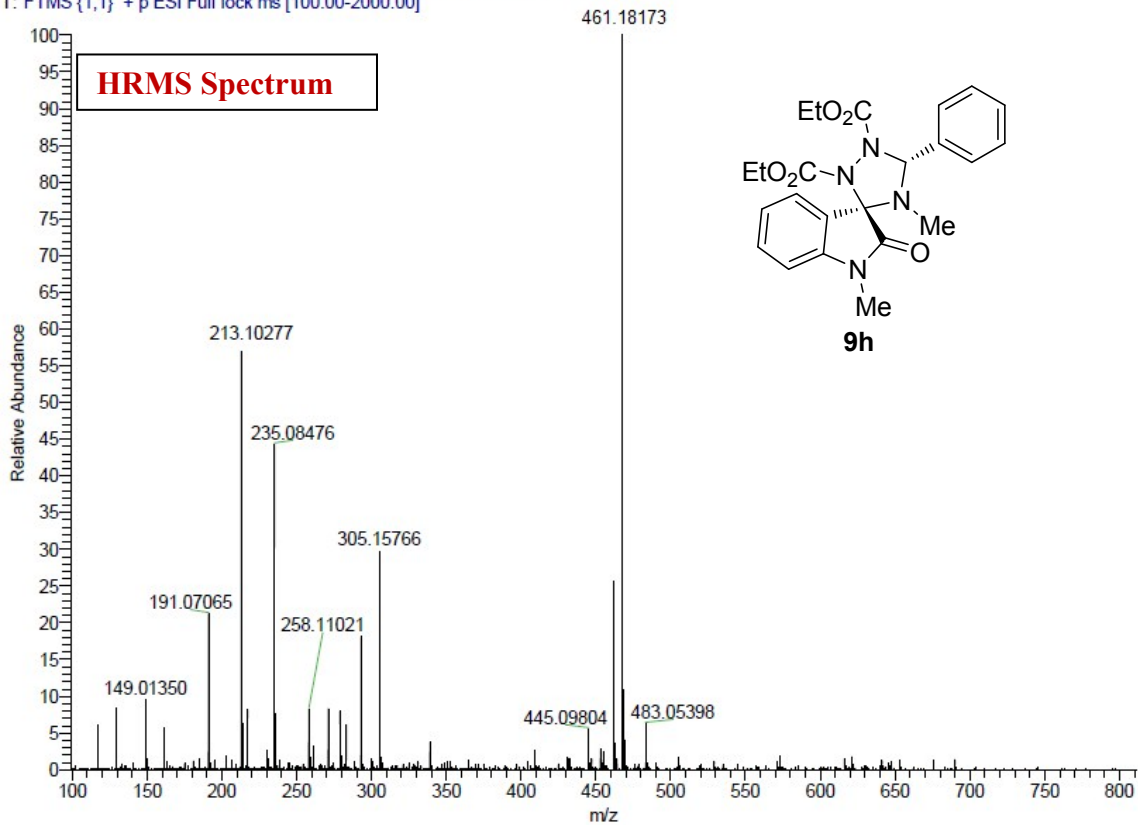
¹³C DEPT135 NMR (100 MHz, CDCl₃)

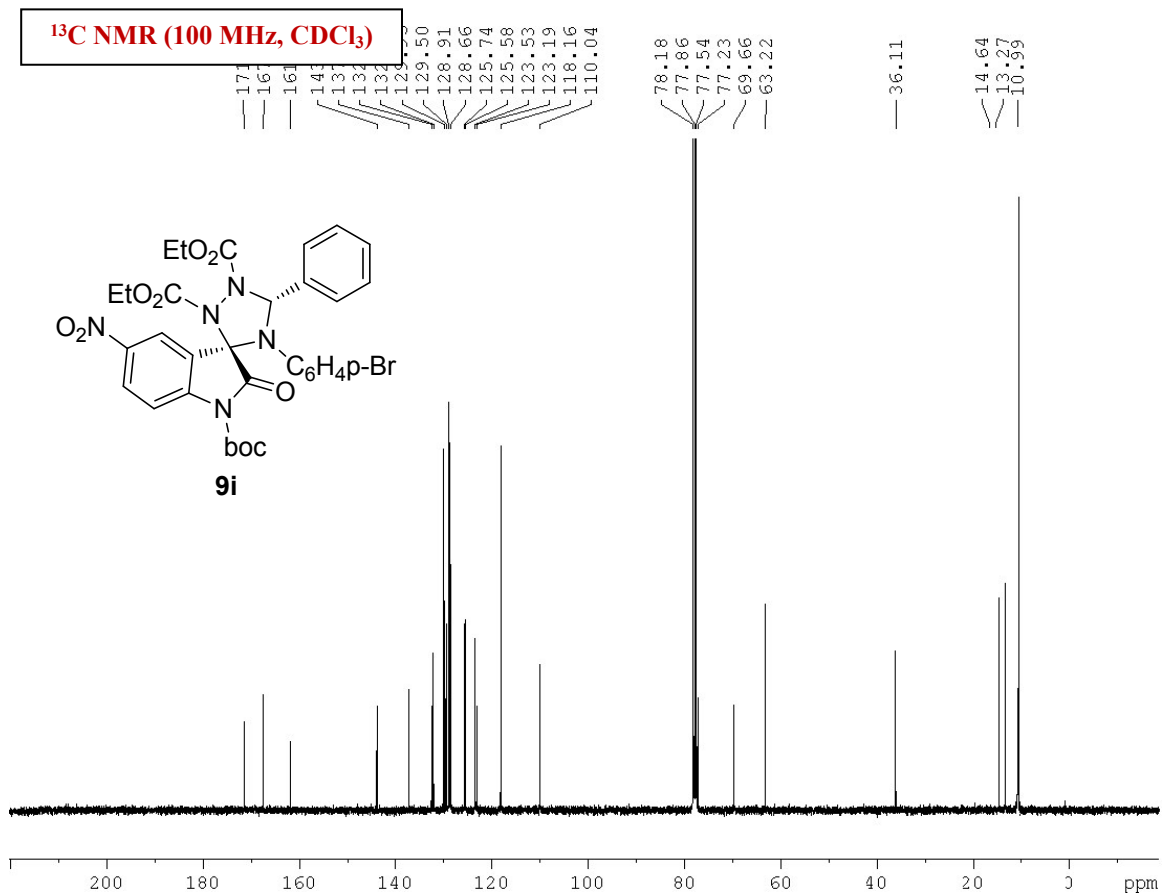
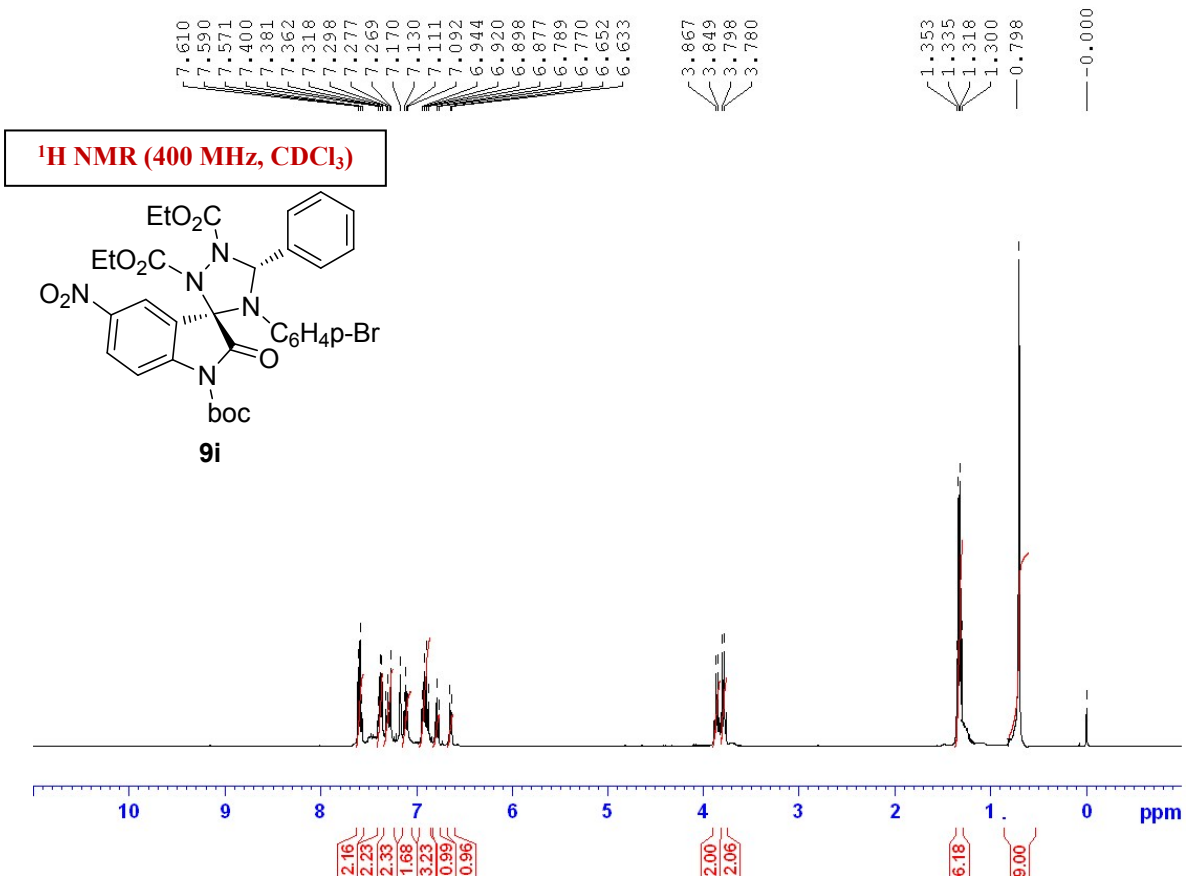


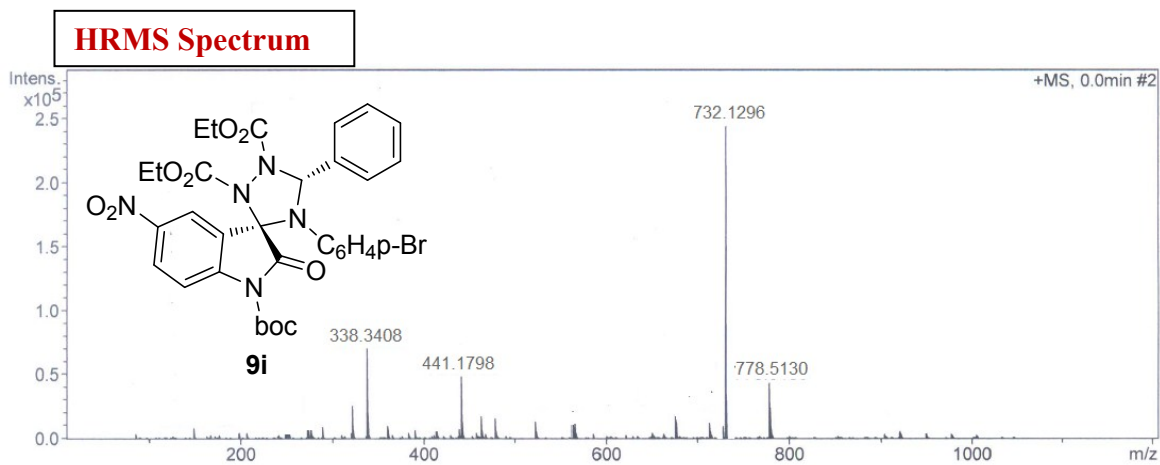
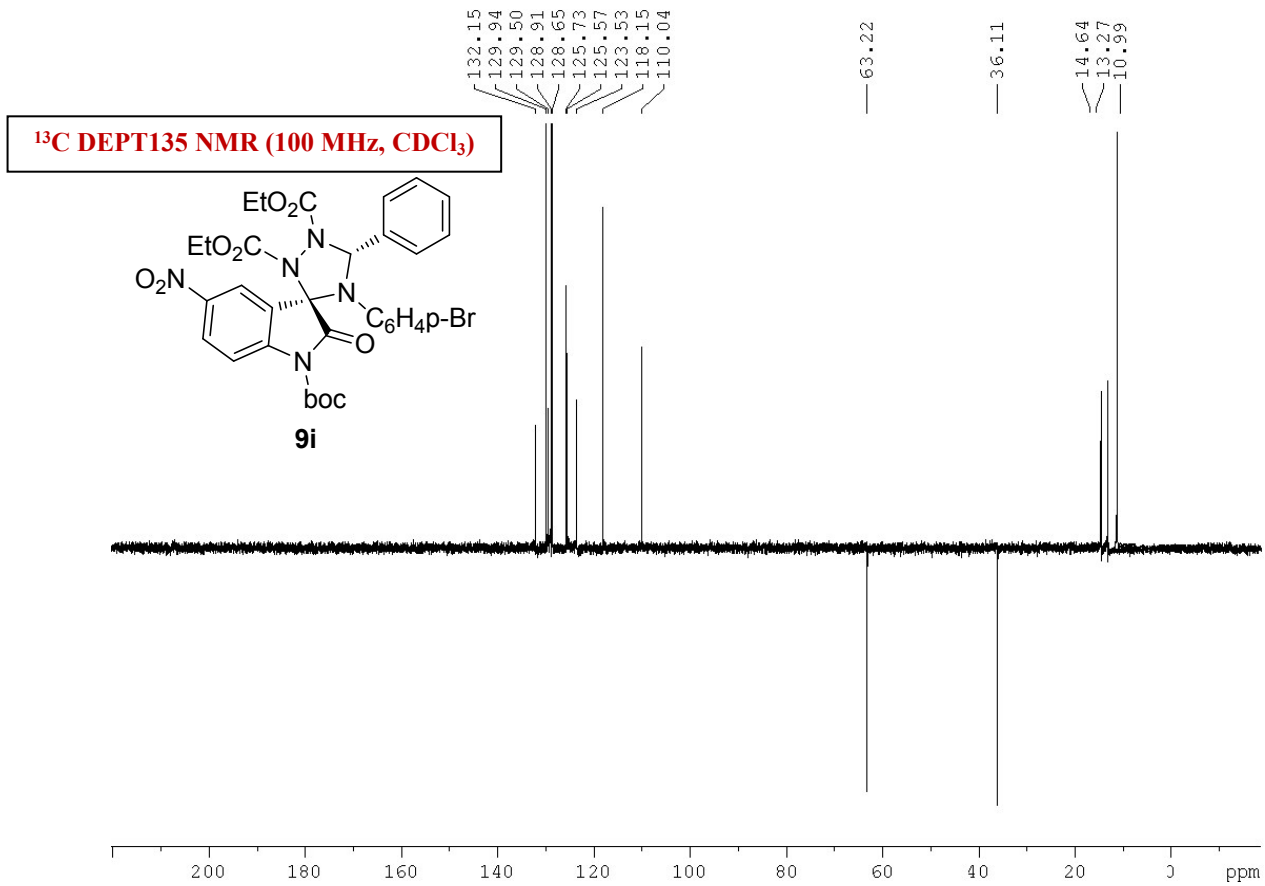
C:\Xcalibur...1-10 MAY 2014\SMGK-307

02-05-2014 13:05:11

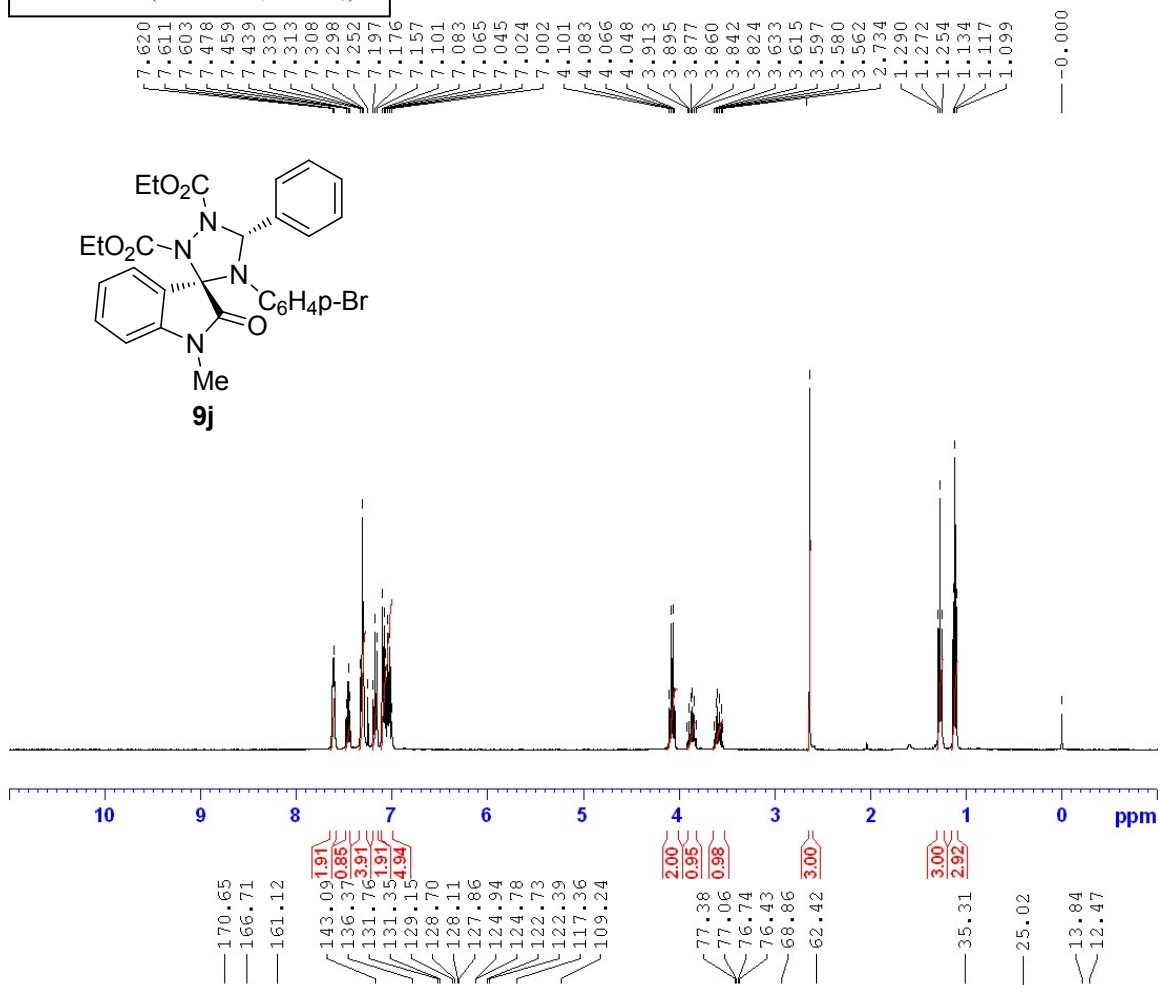
SMGK-307 #73 RT: 1.09 AV: 1 SB: 233 0.09-0.71, 2.15-5.02 NL: 3.05E5
T: FTMS {1,1} + p ESI Full lock ms [100.00-2000.00]



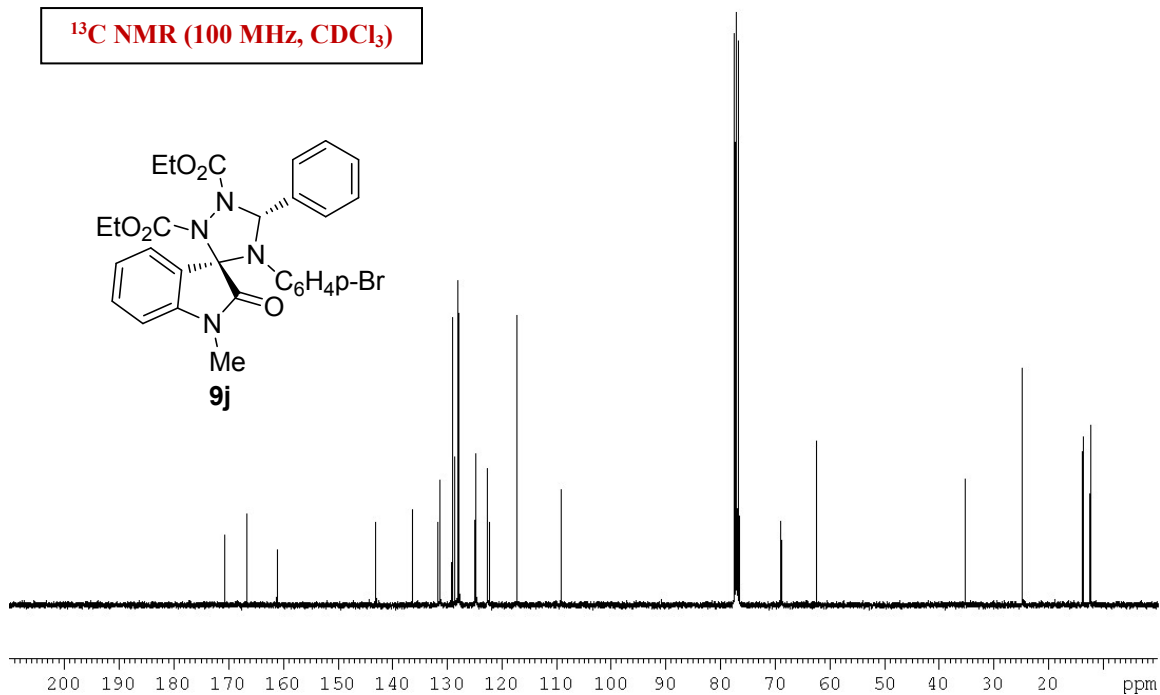




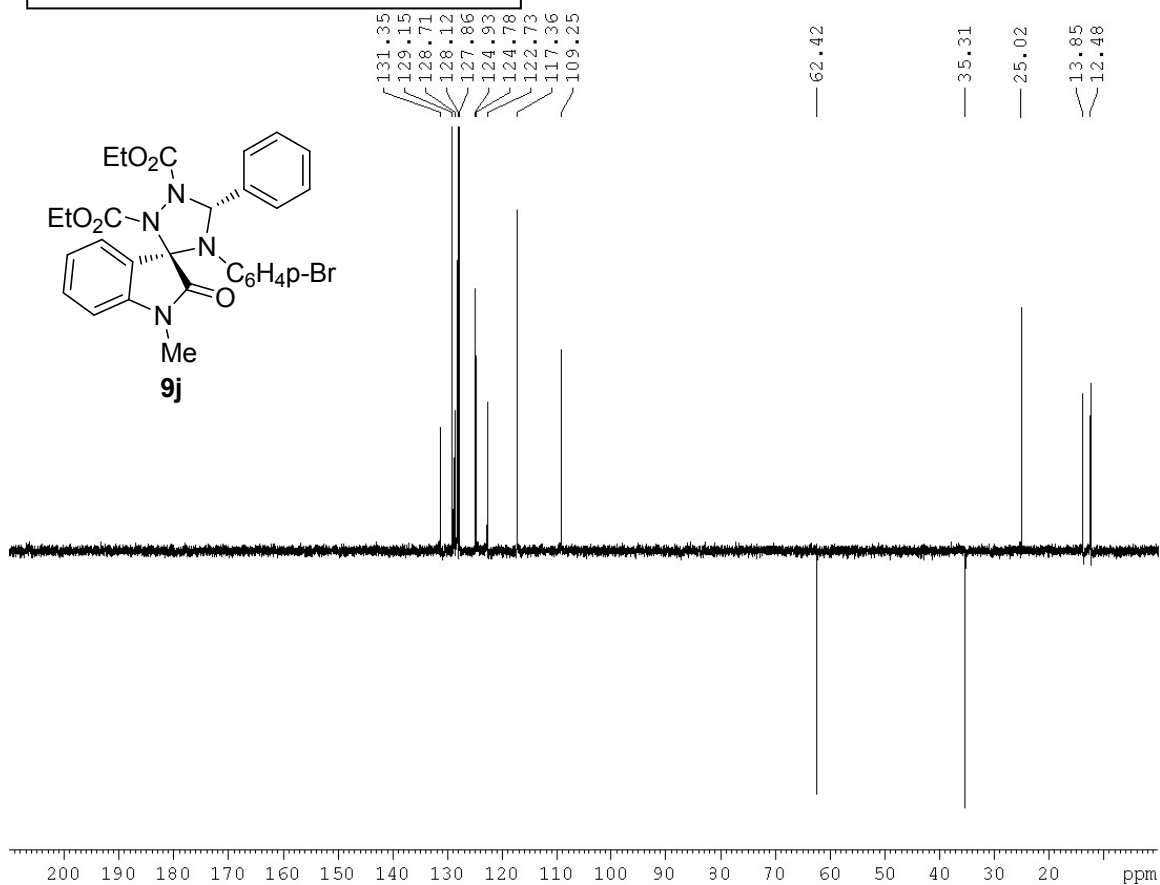
¹H NMR (400 MHz, CDCl₃)



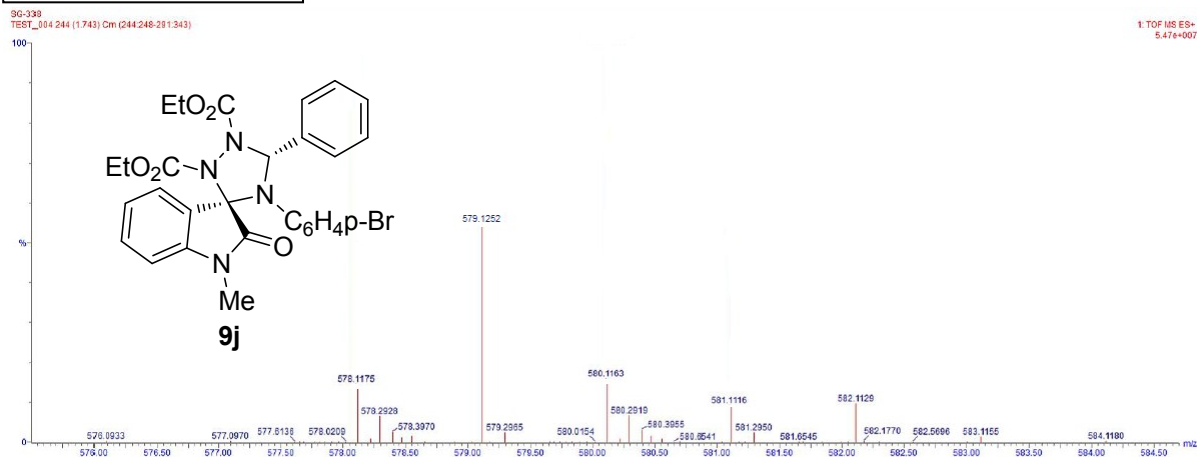
¹³C NMR (100 MHz, CDCl₃)



¹³C DEPT135 NMR (100 MHz, CDCl₃)



HRMS Spectrum

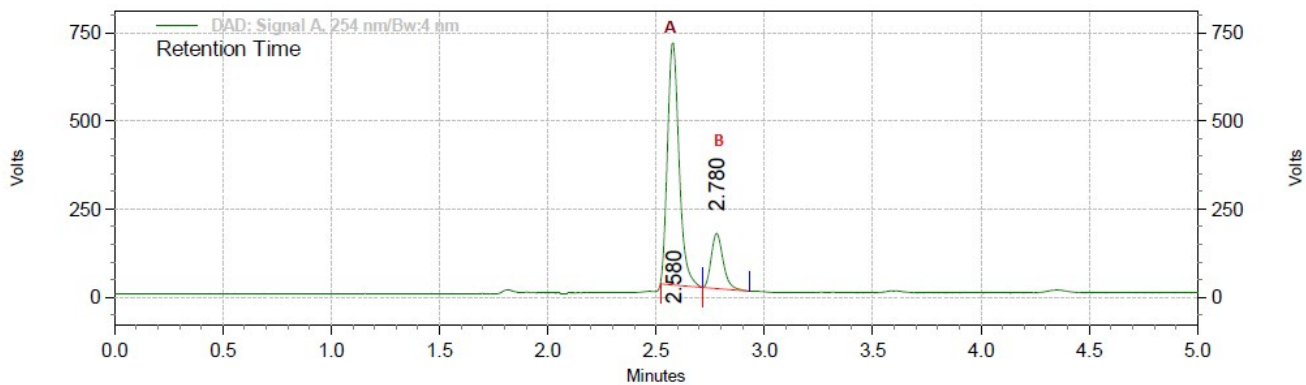


HPLC data for compound 5k

Area % Report

Data File: D:\Ezchrom\hplc data\sgk307-100acn2014_04_msk_100acn.met.rslt\186.dat

Acquired: 4/9/2014 12:21:53 PM (GMT +05:30)



DAD: Signal

A, 254 nm/Bw: 4 nm

Results

Retention Time	Area	Area %	Height	Height %	Name
2.580	5367566	81.11	1439529	81.36	A
2.780	1250371	18.89	329901	18.64	B
Totals	6617937	100.00	1769430	100.00	