Electronic Supplementary Material (ESI) for New Journal of Chemistry.


# Supporting Information

## Facile Synthesis of Spiro-Pyrazolone-Tetrahydrofurans: *ipso*-Cyclization of arylidene pyrazolones with haloalcohols

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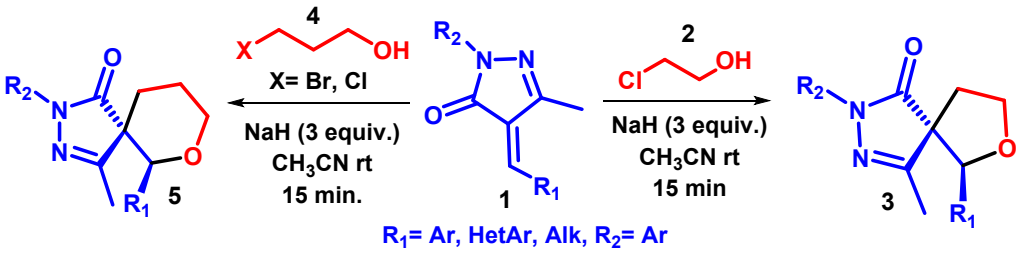
## Material and Methods General Information

All reactions were carried out in oven-dried glassware and the compounds synthesized were fully characterized by spectroscopic data. The NMR spectra were recorded on JEOL - 400 spectrometers, ($^1$H-NMR 400 MHz, $^{13}$C-NMR 100 MHz) and were referenced to the residual peaks of $CDCl_3$ at 7.26 ppm ($^1$H-NMR) and $CDCl_3$ at 77.23 ppm ($^{13}$C-NMR). Chemical shifts (δ) are expressed in ppm, and *J* values are given in Hz. Data are reported as follows: Chemical shift in ppm (δ), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet), coupling constant (Hz), and integration. The reactions were monitored by thin layer chromatography (TLC) using silica gel GF254. The melting points (m.p.) were determined on the digital melting point apparatus and are uncorrected. Micromass Q-TOF mass spectrometer was used to record high-resolution mass spectra 60-70 eV in ESI mode. Materials unless otherwise indicated, all reagents were obtained from commercial suppliers used without further purification.

All the Arylidene pyrazolone was synthesized using the procedure given in the literature.[1]

**General Procedure for the preparation of Spiro-Pyrazolone-Tetrahydrofurans/Pyrans**




To a 25 mL reaction flask, was added Arylidene pyrazolone **1** (100 mg, 1 equiv., 0.381 mmol) 2-chloro ethanol **2** (61 mg, 2 equiv. 0.763 mmol) or 3-Bromo propanol **4** (105 mg, 2 equiv. 0.763 mmol), Sodium hydride (45 mg, 3.0 equiv., 1.145mmol) and Acetonitrile (3 mL). The reaction mixture was stirred at room temperature for 15 minutes. The progress of the reaction was monitored by TLC. After the completion of the reaction, the mixture was quenched with water and extracted with ethyl acetate. The combined organic layers were dried over sodium sulfate, concentrated under reduced pressure and purified by column chromatography to afford product **3** or **5** as white solid.

### References

1 R. Maity, C. Gharui, AK Sil, S.C. Pan, Organocatalytic asymmetric Michael/hemiketalization/retro-aldol reaction of α-nitroketones with unsaturated pyrazolones: Synthesis of 3-acyloxy pyrazoles *Org. Lett.*, 2017, **19**, 3, 665.

**Table 1: Structures of both major(3c) and minor(3l) diastereomers were confirmed by single crystal X-ray analysis. Crystal structure details are given below.**

| Parameters | **3c** | **3l** |
|---|---|---|
| CCDC Number | 2325753 | 2346737 |
| Empirical formula | $C_{20}H_{20}N_2O_3$ | $C_{19}H_{16}BrN_2O_2$ |
| Formula weight | 336.38 | 384.24 |
| Crystal system | Orhorhombic | Monoclinic |
| Space group | P-21 | C 2/c |
| Temperature (K) | 293 K | 293 K |
| Unit cell dimensions | a = 7.7915 (1) Å | a=21.8407(5) Å |
| | b = 12.8038 (1) Å | b=6.7167(1) Å |
| | c = 17.2663 (2) Å | c=24.6680(7) Å |
| | α = 90 | α = 90 |
| | β = 90 | β = 109.105 (3) |
| | γ = 90 | γ = 90 |
| volume ($Å^3$ ) | 1722.50(3) | 3419.41(15) |
| Z | 4 | 8 |
| Radiation type (Cu-Kα)/Å | 1.54184 | 1.54184 |
| No. measured reflections | 17933 | 2733 |
| Calculated density (mg/m3 ) | 1.297 | 1.493 |
| Absorption coefficient (mm-1) | 0.713 | 3.386 |
| F(000) | 712 | 1560.0 |
| θ range for data collection | 4.299 to 72.062° | 0.996 to 72.166° |
| Limiting indices | $-9 \le h \le 9$<br>$-15 \le h \le 15$<br>$-21 \le h \le 20$ | $26 \le h \le 26$<br>$-6 \le k \le 8$<br>$-30 \le l \le 30$ |
| Refinement method | Full-matrix least squares on $F^2$ | Full-matrix least squares on $F^2$ |
| Data / restraints / parameter | 3395/0/228 | 3369/0/218 |
| Final R Indices[I > 2σ(I)] | R1 = 0.0349<br>wR2 = 0.0908 | R1 = 0.0676, wR2 = 0.1780 |
| R indices (all data) | R1 = 0.0385<br>wR2 = 0.0942 | R1 = 0.0582, wR2 = 0.1700 |
| Goodness of fit on $F^2$ | 1.060 | 1.064 |
| Largest diff. peak and hole ($eÅ^{-3}$) | 0.095 to -0.152 | 0.76/-0.47 |
| Reflections collected / unique | 17933/3395 [R(int)=0.0295] | 18443/3369 [Rint = 0.0482, Rsigma = 0.0288] |

**Sample preparation for crystal growth**: Compound **3c&3l** was dissolved in ethanol, while slow evaporation of solvent at room temperature needle type crystals were grown.

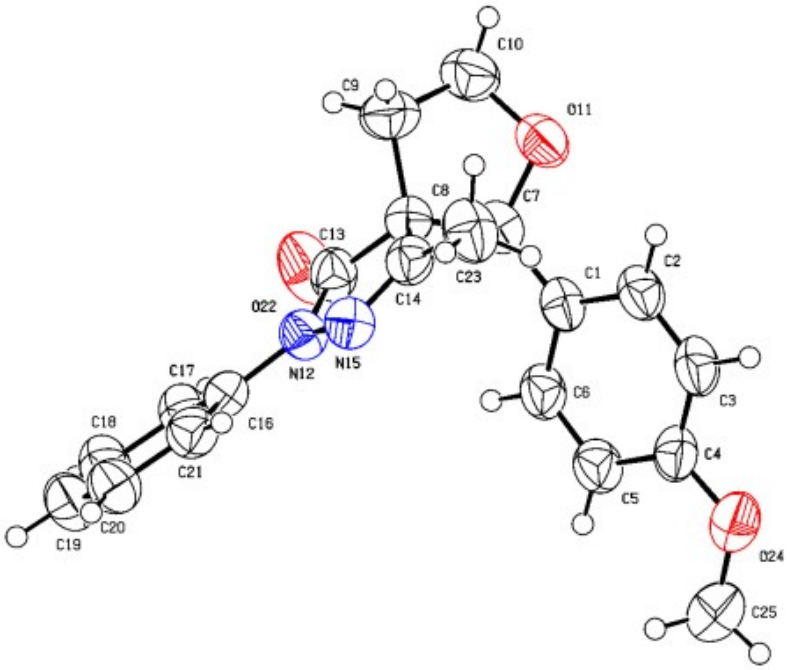



**Figure 1**: ORTEP diagram of the compound of **3c (major isomer)** with 50% ellipsoid probability.

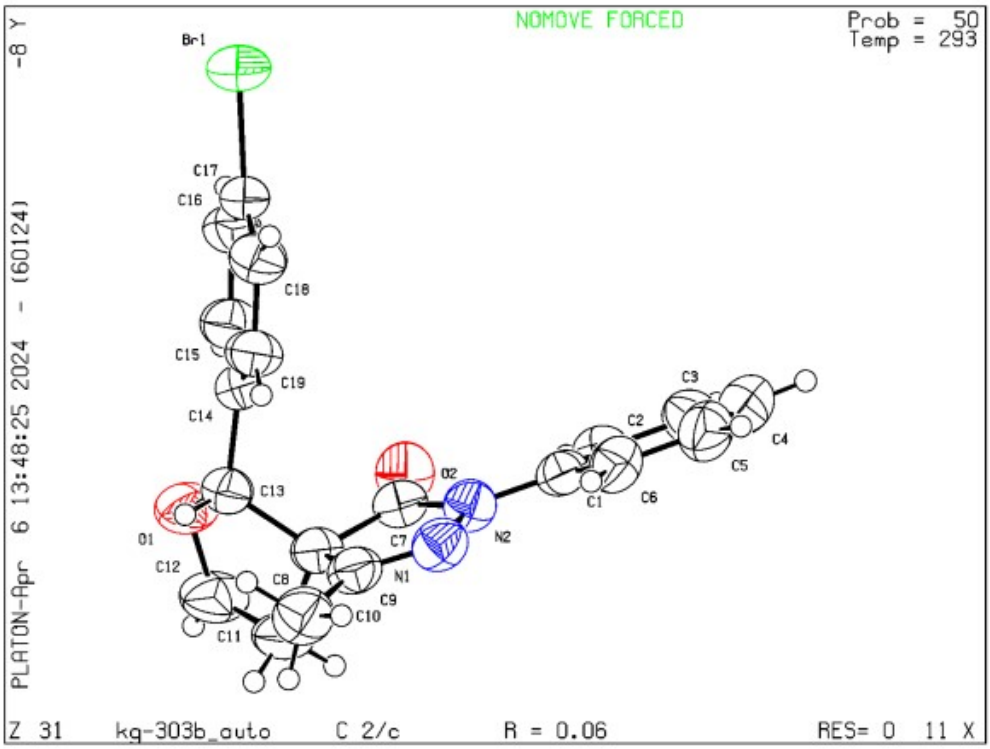
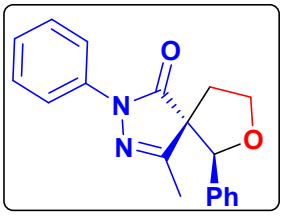



**Figure 2**: ORTEP diagram of the compound of **3l (minor isomer)** with 50% ellipsoid probability.

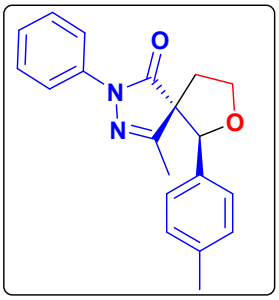
## ***Analytical Data***

**(5*S*,6*S*)-4-methyl-2,6-diphenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3a):**



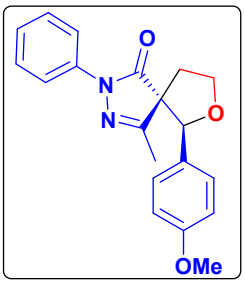

White solid; m.p. 145 - 147 °C; Yield – 76% (88 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.83 (s, 3H), 2.29 - 2.38 (m, 1H), 2.58 – 2.67 (m, 1H), 4.24 – 4.34 (m, 1H), 4.34 – 4.42 (m , 1H), 5.17 (s, 1H), 7.15 – 7.27 (m, 6H), 7.35 – 7.42 (m, 2H), 7.83 – 7.87 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.18, 34.58, 64.46, 67.39, 87.55, 118.89, 124.18, 125.12, 128.02, 128.32, 128.77, 125.65, 127.83, 161.73, 173.22; HRMS (ESI): m/z calcd for $C_{19}H_{18}N_2O_2$ [M+1] 329.1257 found 329.1260.

**(5*S*,6*S*)-4-methyl-2-phenyl-6-(p-tolyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3b):**



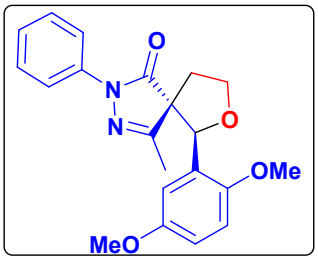

White solid; m.p. 129 - 131 °C; Yield – 78% (90 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.85 (s, 3H), 2.27 (s, 3H), 2.31 – 2.40 (m, 1H), 2.60 – 2.68 (m, 1H), 4.25 – 4.33 (m,1H), 4.37 – 4.43 (m, 1H), 5.14 (s, 1H), 7.02 – 7.10 (m, 4H), 7.16 – 7.22 (m, 1H), 7.36 – 7.43 (m, 2H), 7.82 – 7.87 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.34, 21.09, 34.64, 64.57, 67.42, 87.73, 118.97, 124.14, 125.16, 128.85, 129.07, 132.63, 137.75, 137.92, 161.97, 173.35; HRMS (ESI): m/z calcd for $C_{20}H_{20}N_2O_2$ [M+Na] 344.1448 found 344.1450.

**(5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3c) :**



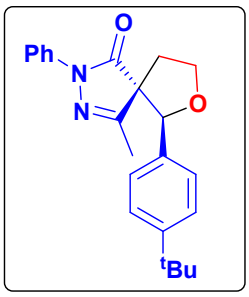

White solid; m.p. 134 - 136 °C; Yield – 72% (82 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.88 (s, 3H), 2.31 – 2.40 (m, 1H), 2.60 – 2.68 (m, 1H), 3.74 (s, 3H), 4.24 – 4.32 (m,1H), 4.35 – 4.42 (m, 1H), 5.12 (s, 1H), 6.75 – 6.79 (m, 2H), 7.10 – 7.22 (m, 3H), 7.36 – 7.42 (m, 2H),7.82 – 7.87 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.36, 34.51, 55.12, 64.60, 67.39, 87.66, 113.75, 118.92, 125.14, 125.46, 127.61, 128.84, 137.89, 159.26, 161.98, 173.30; HRMS (ESI): m/z calcd for $C_{20}H_{20}N_2O_3$ [M+Na] 359.1361 found 359.1366.

**(5*S*,6*S*)-6-(2,5-dimethoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3d) :**

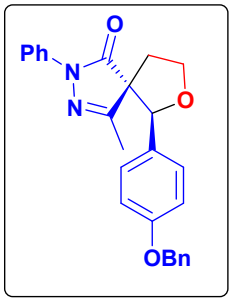



White solid; m.p. 144 - 146 °C; Yield – 75% (85 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.59 (s, 3H), 2.22 - 2.32 (m, 1H), 2.43 – 2.51 (m, 1H), 3.33 (s, 3H), 3.8 (s, 3H), 4.16 – 4.24 (m, 1H), 4.35 – 4.42 (m , 1H), 5.28 (s, 1H), 6.60 – 6.64 (m, 1H), 6.72 – 6.77 (m, 1H), 7.21 – 7.24 (m, 1H), 7.36 – 7.42 (m, 2H), 7.12 – 7.18 (m, 1H), 7.94 - 7.99 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 14.74, 36.82, 55.05, 55.87, 63.11, 67.73, 83.45, 110.17, 112.47, 113.28, 118.01, 124.39, 126.65, 128.81, 138.56, 149.98, 153.38, 161.48, 175.58; HRMS (ESI): m/z calcd for $C_{21}H_{22}N_2O_4$ [M+1] 361.1620 found 361.1623.

**(5*S*,6*S*)-6-(4-(tert-butyl)phenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3e)** White solid; m.p. 152 - 154 °C; Yield – 87% (99 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.25 (s, 9H), 1.85 (s, 3H), 2.32 – 2.41 (m, 1H), 2.60 – 2.68 (m, 1H), 4.25 – 4.33 (m, 1H), 4.36 – 4.43 (m, 1H), 5.15 (s, 1H), 7.00 – 7.12 (m, 2H), 7.17 – 7.27 (m, 3H), 7.37 – 7.44 (m, 2H), 7.82 – 7.87 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.34, 31.24, 34.50, 34.84, 64.43, 67.40, 87.56, 119.04, 123.99, 125.21, 125.29, 128.88, 132.58, 137.90, 150.96, 162.13, 173.41; HRMS (ESI): m/z calcd for $C_{23}H_{26}N_2O_2$ [M+Na] 385.1720 found 385.1725.

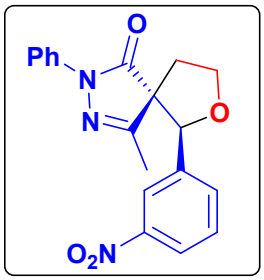


**(5*S*,6*S*)-6-(4-(benzyloxy)phenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3f) :**



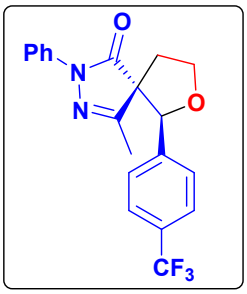

White solid; m.p. 147 - 149 °C; Yield – 74% (82 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.81 (s, 3H), 2.26 – 2.35 (m, 1H), 2.53 – 2.62 (m, 1H), 4.18 – 4.27 (m, 1H), 4.30 – 4.36 (m, 1H), 4.92 (s, 2H), 5.05 (s, 1H), 6.75 – 6.81 (m, 2H), 7.04 – 7.16 (m, 3H), 7.21 – 7.55 (m, 7H), 7.75 – 7.79 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.41, 34.52, 64.62, 67.44, 69.94, 87.66, 114.70, 118.98, 125.20, 125.52, 127.52, 127.92, 127.98, 128.55, 128.88, 136.73, 137.88, 158.42, 162.02, 173.22; HRMS (ESI): m/z calcd for $C_{26}H_{24}N_2O_3$ [M+Na] 435.1682 found 435.1679.

**(5*S*,6*S*)-4-methyl-6-(3-nitrophenyl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3g):**



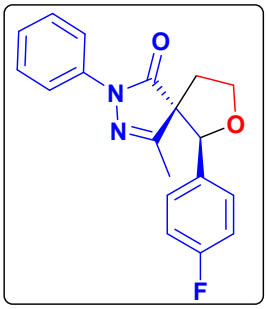

White solid; m.p. 141 - 143 °C; Yield – 92% (105 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.85 (s, 3H), 2.37 – 2.46 (m, 1H), 2.66 – 2.75 (m, 1H), 4.31 – 4.39 (m, 1H), 4.41 – 4.49 (m, 1H), 5.19 (s, 1H), 7.18 – 7.24 (m, 1H), 7.34 – 7.42 (m, 4H), 8.08 – 8.13 (m, 1H), 8.23 – 8.25 (m, 1H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.40, 34.49, 64.38, 67.73, 86.41, 119.05, 119.69, 123.21, 125.60, 128.97, 129.60, ; HRMS (ESI): m/z calcd for $C_{19}H_{17}N_3O_4$ [M+Na] 374.1126 found 374.1128.

**(5*S*,6*S*)-4-methyl-2-phenyl-6-(4-(trifluoromethyl)phenyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3h) :**



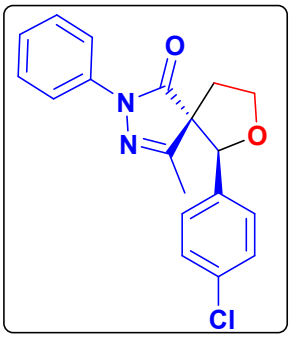

White solid; m.p. 140 - 142 °C; Yield – 90% (102 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.83 (s, 3H), 2.33 – 2.42 (m, 1H), 2.63 – 2.72 (m, 1H), 4.27 – 4.35 (m, 1H), 4.37 – 4.45 (m, 1H), 5.17 (s, 1H), 7.17 – 7.23 (m, 1H), 7.30 – 7.33 (m, 2H), 7.37 – 7.43 (m, 2H), 7.48 – 7.52 (m, 2H), 7.81 – 7.86 (M, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.26, 34.73, 64.39, 67.61, 86.79, 118.89, 122.50, 124.73, 125.42, 128.95, 130.48, 137.70, 139.81, 161.30, 172.87; HRMS (ESI): m/z calcd for $C_{20}H_{17}F_3N_2O_2$ [M+Na] 397.1132 found 397.1134.

**(5*S*,6*S*)-6-(4-fluorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3i) :**



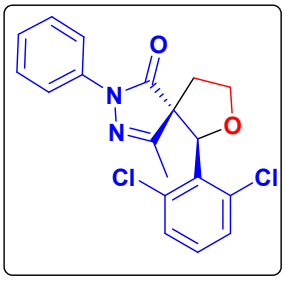

White solid; m.p. 151 - 153 °C; Yield – 52% (60 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.86 (s, 3H), 2.33 – 2.42 (m, 1H), 2.61 – 2.69 (m, 1H), 4.26 – 4.35 (m, 1H), 4.36 – 4.44 (m, 1H), 5.13 (s, 1H), 6.90 – 7.98 (m, 2H), 7.15 – 7.21 (m, 3H), 7.37 – 7.43 (m, 2H), 7.82 – 7.86 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.36, 34.51, 64.40, 67.52, 87.17, 115.32, 115.53, 118.91, 125.33, 125.93, 126.05, 128.92, 131.40, 161.72, 173.06; HRMS (ESI): m/z calcd for $C_{19}H_{17}FN_2O_2$ [M+Na] 347.1326 found 347.1329.

**(5*S*,6*S*)-6-(4-chlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3j) :**



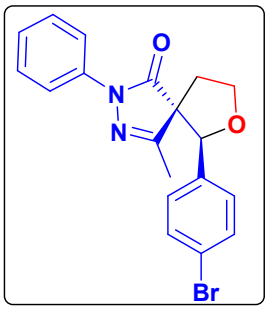

White solid; m.p. 137 - 139 °C; Yield – 55% (63 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.86 (s, 3H), 2.33 – 2.42 (m, 1H), 2.62 – 2.71 (m, 1H), 4.26 – 4.35 (m, 1H), 4.36 – 4.44 (m, 1H), 5.12 (s, 1H), 7.11 – 7.16 (m, 2H), 7.18 – 7.25 (m, 3H), 7.37 – 7.42 (m, 2H), 7.82 – 7.86 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.36, 34.59, 64.47, 67.55, 87.01, 118.91, 125.36, 125.70, 128.66, 128.93, 133.90, 134.23, 137.75, 161.54, 172.99; HRMS (ESI): m/z calcd for $C_{19}H_{17}ClN_2O_2$ [M+Na] 363.0864 found 363.0866.

**(5*S*,6*R*)-6-(2,6-dichlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3k) :**



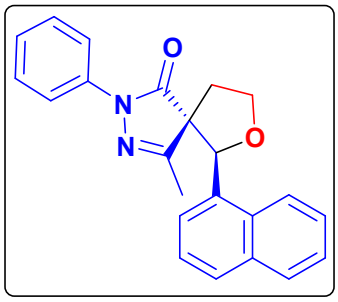

White solid; m.p. 136 - 138 °C; Yield – 69% (78 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.56 (s, 3H), 2.21 – 2.27 (m, 1H), 2.47 – 2.56 (m, 1H), 4.32 – 4.47 (m, 2H), 5.85 (s, 1H), 7.07 – 7.16 (m, 2H), 7.17 – 7.22 (m, 2H), 7.31 – 7.37 (m, 2H), 7.78 – 7.82 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 14.14, 36.61, 64.40, 68.32, 83.92, 118.84, 125.16, 128.87, 129.52, 129.75, 132.95, 134.71, 137.76, 158.91, 175.83; HRMS (ESI): m/z calcd for $C_{19}H_{16}Cl_2N_2O_2$ [M+Na] 397.0490 found 390.0494.

**(5*S*,6*S*)-6-(4-bromophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3l):**



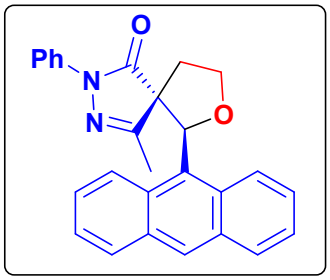

White solid; m.p. 158 - 160 °C; Yield – 81% (91 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.84 (s, 3H), 2.30 – 2.39 (m, 1H), 2.60 – 2.68 (m, 1H), 4.23 – 4.41 (m, 2H), 5.09 (s, 1H), 7.04 – 7.09 (m, 2H), 7.16 – 7.22 (m, 1H), 7.34 – 7.43 (m, 4H), 7.82 – 7.87 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.25, 34.54, 64.33, 6.47, 86.94, 118.82, 121.98, 125.26, 125.91, 128.84, 131.50, 134.77, 137.72, 161.39, 172.90; HRMS (ESI): m/z calcd for $C_{19}H_{17}BrN_2O_2$ [M+Na] 407.0361 found 407.0366.

**(5*S*,6*S*)-4-methyl-6-(naphthalen-1-yl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3m) :**



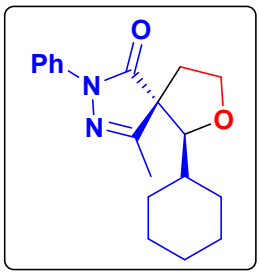

White solid; m.p. 142 - 144 °C; Yield – 70% (79 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.54 (s, 3H), 2.36 – 2.46 (m, 1H), 2.56 – 2.64 (m, 1H), 4.35 – 4.43 (m, 1H), 4.50 – 4.56 (m, 1H), 5.90 (s, 1H), 7.13 – 7.21 (m, 2H), 7.30 – 7.38 (m, 3H), 7.43 – 7.49 (m, 1H), 7.56 – 7.59 (m, 1H), 7.65 – 7.69 (m, 2H), 7.76 – 7.80 (m, 2H), 7.87 – 7.91 (m, 1H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.01, 36.31, 64.19, 67.95, 85.32, 119.11, 122.57, 123.31, 124.73, 125.21, 125.81, 126.14, 128.71, 128.82, 129.92, 132.40, 133.46, 13.74, 161.43, 174.98; HRMS (ESI): m/z calcd for $C_{23}H_{20}N_2O_2$ [M+Na] 379.1424 found 380.1427.

**(5S,6S)-6-(anthracen-9-yl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3n) :**



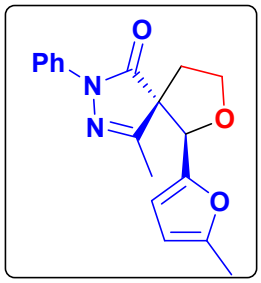

White solid; m.p. 139 - 141 °C; Yield – 67% (75 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.18 (s, 3H), 2.50 – 2.56 (m, 1H), 2.60 – 2.69 (m, 1H), 4.43 – 4.51 (m, 1H), 4.72 – 4.76 (m, 1H), 6.70 (s, 1H), 7.15 – 7.24 (m, 3H), 7.34 – 7.41 (m, 6H), 7.69 – 7.72 (m, 2H), 7.93 – 7.98 (m, 2H), 8.42 (s, 3H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 14.99, 29.68, 36.92, 65.55, 68.26, 88.15, 119.11, 124.83, 125.25, 126.06, 126.58, 128.77, 129.02, 129.34, 129.49, 130.05, 130.13, 131.33, 135.93, 136.00, 137.77, 148.33, 150.48, 161.16, 176.27; HRMS (ESI): m/z calcd for $C_{27}H_{22}N_2O_2$ [M+Na] 429.1579 found 429.1575.

**(5*S*,6*S*)-6-cyclohexyl-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3o) :**



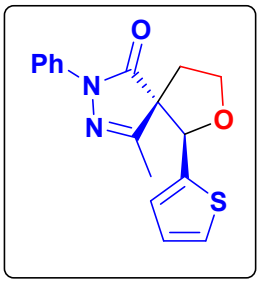

White solid; m.p. 123 - 125 °C; Yield – 62% (68 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 0.84 – 1.69 (m, 9H), 2.19 – 2.52 (m, 6H), 2.45 – 2.52 (m, 1H), 3.68 – 3.72 (m, 1H), 4.03 – 4.15 (m, 2H), 7.15 – 7.21 (m, 1H), 7.38 – 7.42 (m, 2H), 7.84 – 7.89 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.78, 25.93, 27.62, 31.28, 36.35, 39.50, 61.72, 66.29, 91.07, 118.82, 125.06, 128.88, 138.04, 162.52, 173.50; HRMS (ESI): m/z calcd for $C_{18}H_{22}N_2O_2$ [M+Na] 321.1884 found 321.1886.

**(5*S*,6*R*)-4-methyl-6-(5-methylfuran-2-yl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3p) :**



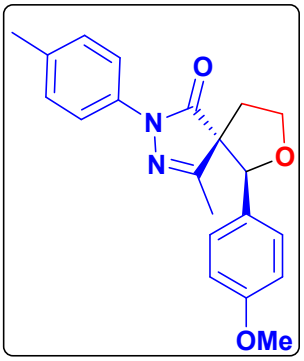

White solid; m.p. 146 - 148 °C; Yield – 82% (95 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 2.14(s, 3H), 2.15 (s, 3H), 2.29 – 2.40 (m, 1H), 2.58 – 2.67 (m, 1H), 4.24 – 4.38 (m, 2H), 4.36 – 4.43 (m, 1H), 5.02 (s, 1H), 5.82 – 5.85 (m, 1H), 6.16 (d, *J* = 2.8 Hz, 1H), 7.15 – 7.21 (m, 1H), 7.35 – 7.42 (m, 2H), 7.82 – 7.87 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 13.3, 15.34, 34.17, 63.11, 67.51, 82.49, 106.11, 108.95, 118.96, 125.12, 128.79, 137.88, 146.18, 152.74, 161.89, 172.27; HRMS (ESI): m/z calcd for $C_{18}H_{18}N_2O_3$ [M+Na] 333.1227 found 333.1230.

**(5*S*,6*R*)-4-methyl-2-phenyl-6-(thiophen-2-yl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3q) :**



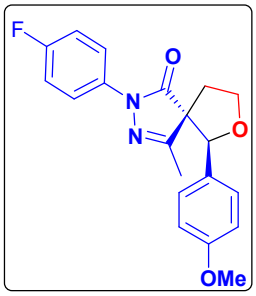

White solid; m.p. 149 - 151 °C; Yield – 77% (89 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 2.01 (s, 3H), 2.36 – 2.45 (m, 1H), 2.63 – 2.72 (m, 1H), 4.29 – 4.43 (m, 2H), 5.34 (s, 1H), 6.77 – 6.80 (m, 1H), 6.88 – 6.91 (m, 1H), 7.18 – 7.23 (m, 2H), 7.37 – 7.43 (m, 2H), 7.84 – 7.89 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.58, 34.34, 64.60, 67.70, 85.20, 118.96, 123.29, 124.63, 125.26, 127.05, 128.89, 137.68, 137.84, 161.52, 172.31; HRMS (ESI): m/z calcd for $C_{17}H_{16}N_2O_2S$ [M+Na] 335.0976 found 335.0977.

**(5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-(p-tolyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3r) :**



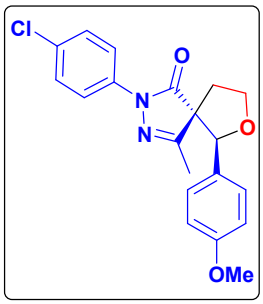

White solid; m.p. 153 - 155 °C; Yield – 63% (72 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.86 (s, 3H), 2.30 – 2.39 (m, 4H), 2.58 – 2.67 (m, 1H), 3.73 (s, 3H), 4.24 – 4.32 (m, 1H), 4.35 – 4.41 (m, 1H), 5.10 (s, 1H), 6.73 – 6.79 (m, 2H), 7.07 – 7.21 (m, 2H), 7.76 – 7.74 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.33, 20.91, 34.45, 55.11, 64.53, 67.39, 87.64, 113.75, 119.00, 125.48, 127.69, 129.34, 134.86, 135.51, 159.26, 161.82, 173.08; HRMS (ESI): m/z calcd for $C_{21}H_{22}N_2O_3$ [M+Na] 343.1403 found 343.1407.

**(5*S*,6*S*)-2-(4-fluorophenyl)-6-(4-methoxyphenyl)-4-methyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3s):**



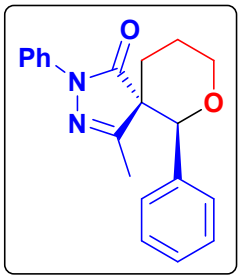

White solid; m.p. 155 - 157 °C; Yield – 65% (74 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.88 (s, 3H), 2.32 – 2.40 (m, 1H), 2.60 – 2.68 (m, 1H), 3.75 (s, 3H), 4.25 – 4.33 (m,1H), 4.35 – 4.43 (m, 1H), 5.11 (s, 1H), 6.75 – 6.80 (m, 2H), 7.05 – 7.13 (m, 4H), 7.78 – 7.85 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.35, 34.45, 55.15, 64.61, 67.14, 87.74, 113.79, 115.43, 115.66, 120.62, 120.69, 125.48, 127.56, 134.08, 159.35, 162.15, 173.12; HRMS (ESI): m/z calcd for $C_{20}H_{19}FN_2O_3$ [M+Na] 377.1246 found 377.1248.

**(5*S*,6*S*)-2-(4-chlorophenyl)-6-(4-methoxyphenyl)-4-methyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3t)**



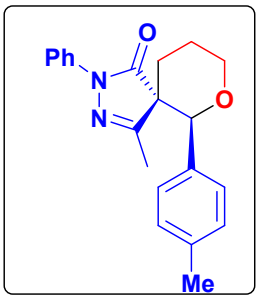

White solid; m.p. 133 - 135 °C; Yield – 61% (69 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.88 (s, 3H), 2.31 – 2.34 (m, 1H), 2.60 – 2.68 (m, 1H), 3.74 (s, 1H), 4.26 – 4.33 (m, 1H), 4.36 – 4.43 (m, 1H), 5.10 (s, 1H), 6.74 – 6.79 (m, 2H), 7.07 – 7.11 (m, 2H), 7.32 – 7.38 (m, 2H), 7.80 – 7.86 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 15.38, 34.49, 55.16, 64.72, 67.43, 87.80, 113.82, 119.94, 125.46, 127.49, 128.91, 130.22, 136.51, 159.37, 162.31, 173.30; HRMS (ESI): m/z calcd for $C_{20}H_{19}ClN_2O_2$ [M+Na] 393.0976 found 393.0976.

**(5*S*,6*S*)-4-methyl-2,6-diphenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5a) :**



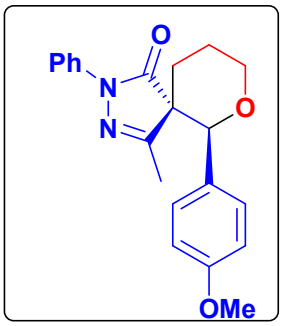

White solid; m.p. 166 - 168 °C; Yield – 74% (90 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.72 – 1.79 (m, 1H), 1.92 – 1.99 (m, 1H), 2.07 – 2.19 (m, 1H), 2.32 – 2.42 (m, 4H), 3.80 – 3.88 (m, 1H), 4.33 – 4.39 (m, 1H), 4.74 (s, 1H), 7.08 – 7.23 (m, 6H), 7.26 – 7.33 (m, 2H), 7.55 – 7.60 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 18.70, 20.91, 28.73, 59.07, 68.77, 82.80, 119.33, 125.18, 125.31, 128.03, 128.09, 128.65, 137.05, 137.41, 161.98, 173.54; HRMS (ESI): m/z calcd for $C_{20}H_{20}N_2O_2$ [M+Na] 343.1526 found 343.1528.

**(5*S*,6*S*)-4-methyl-2-phenyl-6-(p-tolyl)-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5b) :**



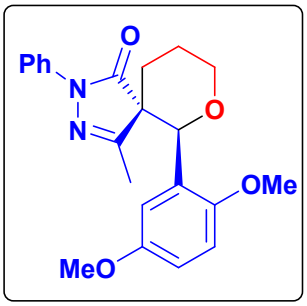

White solid; m.p. 175 - 177 °C; Yield – 72% (87 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.73 – 1.78 (m, 1H), 1.92 – 1.99 (m, 1H), 2.05 – 2.23 (m, 4H), 2.30 – 2.42 (m, 4H), 3.80 – 3.88 (m, 1H), 4.33 – 4.37 (m, 1H), 4.74 (s, 1H), 6.96 – 6.99 (m, 2H), 7.11 – 7.17 (m, 3H), 7.30 – 7.36 (m, 2H), 7.61 – 7.66 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 18.63, 20.89, 20.97, 28.78, 59.08, 68.70, 82.63, 119.24, 125.05, 125.12, 128.587, 128.66, 134.08, 137.45, 137.58, 162.07, 173.59; HRMS (ESI): m/z calcd for $C_{21}H_{22}N_2O_2$ [M+Na] 357.1776 found 357.1779.

**(5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5c) :**



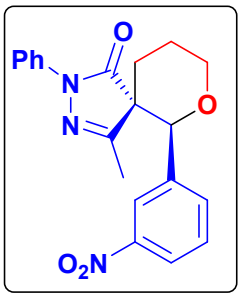

White solid; m.p. 168 - 170 °C; Yield – 74% (88 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.73 – 1.80 (m, 1H), 1.92 – 1.99 (m, 1H), 2.06 – 2.21 (m, 1H), 2.31 – 2.41 (m, 4H), 3.69 (s, 3H), 3.80 – 3.88 (m, 1H), 4.33 – 4.39 (m, 1H), 4.70 (s, 1H), 6.67 – 6.73 (m, 2H), 7.10 – 7.18 (m, 3H), 7.28 – 7.35 (m, 2H), 7.58 – 7.63 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 18.75, 20.97, 28.72, 55.10, 59.23, 68.89, 82.65, 113.41, 119.34, 125.16, 126.60, 128.67, 129.34, 137.41, 159.03, 162.19, 173.72; HRMS (ESI): m/z calcd for $C_{21}H_{22}N_2O_3$ [M+Na] 373.1652 found 373.1654.

**(5*S*,6*S*)-6-(2,5-dimethoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5d) :**



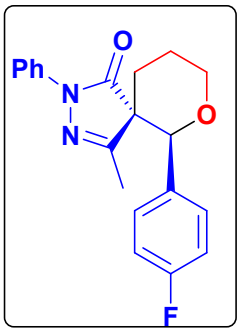

White solid; m.p. 179 - 181 °C; Yield – 69% (81 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.75 – 1.82 (m, 1H), 1.90 – 1.96 (m, 1H), 2.04 – 2.20 (m, 1H), 2.35 – 2.43 (m, 1H), 3.48 (s, 1H), 3.70 – 3.88 (m, 4H), 4.27 – 4.33 (m, 1H), 5.11 (s, 1H), 6.52 – 6.57 (m, 1H), 6.64 – 6.69 (m, 1H), 6.95 – 6.98 (m, 1H), 7.05 – 7.09 (m, 1H), 7.25 – 7.30 (m, 2H), 7.62 – 7.66 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 18.84, 20.86, 28.57, 55.41, 55.67, 58.12, 68.66, 110.96, 113.12, 114.10, 118.62, 124.38, 124.56, 126.48, 128.56, 137.96, 150.04, 153.61, 161.92, 173.07; HRMS (ESI): m/z calcd for $C_{22}H_{24}N_2O_4$ [M+Na] 403.1776 found 403.1777.

**(5*S*,6*S*)-4-methyl-6-(3-nitrophenyl)-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5e) :**



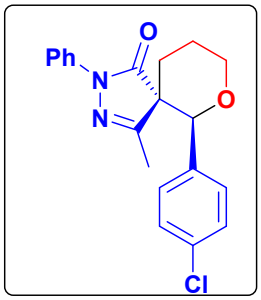

White solid; m.p. 169 - 171 °C; Yield – 89% (105 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.79 – 1.85 (m, 1H), 1.97 – 2.04 (m, 1H), 2.11 – 2.23 (m, 1H), 2.32 – 2.45 (m, 4H), 3.83 – 3.92 (m, 1H), 4.39 – 4.45 (m, 1H), 4.84 (s, 1H), 7.12 – 7.18 (m, 1H), 7.29 – 7.35 (m, 3H), 7.47 – 7.52 (m, 1H), 7.59 - 7.64 (m, 2H), 8.01 – 8.06 (m, 1H), 8.23 – 8.26 (m, 1H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 14.19, 16.24, 24.08, 54.38, 64.34, 77.00, 114.62, 116.07, 118.66, 121.04, 124.35, 124.54, 127.13, 132.62, 134.88, 143.54, 156.99, 168.60; HRMS (ESI): m/z calcd for $C_{20}H_{19}N_3O_4$ [M+Na] 388.1488 found 388.1490.

**(5*S*,6*S*)-6-(4-fluorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5f) :**



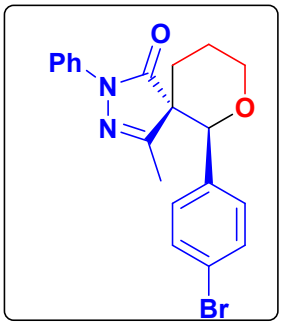

White solid; m.p. 181 - 183 °C; Yield – 56% (67 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.74 – 1.79 (m, 1H), 1.93 – 1.99 (m, 1H), 2.06 – 2.19 (m, 1H), 2.32 – 2.41 (m, 4H), 3.80 – 3.88 (m, 1H), 4.33 – 4.39 (m, 1H), 4.73 (s, 1H), 6.83 – 6.91 (m, 2H), 7.12 – 7.17 (m, 1H), 7.20 – 7.27 (m, 2H), 7.29 – 7.36 (m, 2H), 7.60 – 7.64 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 18.65, 20.80, 28.57, 59.04, 82.19, 114.81, 115.02, 119.15, 125.24, 127.06, 127.14, 128.69, 132.92, 137.31, 161.02, 161.76, 163.47, 163.47, 173.40; HRMS (ESI): m/z calcd for $C_{20}H_{19}FN_2O_2$ [M+Na] 361.1423 found 361.1424.

**(5*S*,6*S*)-6-(4-chlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5g) :**




White solid; m.p. 186 - 188 °C; Yield – 58% (69 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.74 – 1.79 (m, 1H), 1.93 – 1.99 (m, 1H), 2.05 – 2.19 (m, 1H), 2.30 – 2.41 (m, 4H), 3.80 – 3.87 (m, 1H), 4.33 – 4.39 (m, 1H), 4.73 (s, 1H), 7.13 – 7.21 (m, 5H), 7.31 – 7.36 (m, 2H), 7.61 – 7.65 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 18.60, 20.79, 28.73, 58.93, 68.73, 81.93, 119.16, 125.29, 126.71, 128.20, 128.72, 133.77, 135.70, 137.30, 161.72, 173.35; HRMS (ESI): m/z calcd for $C_{20}H_{19}ClN_2O_2$ [M+Na] 377.0256 found 377.0258.

**(5*S*,6*S*)-6-(4-bromophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5h) :**




White solid; m.p. 173 - 175 °C; Yield – 78% (138 mg); $^{1}$H-NMR (400 MHz, $CDCl_3$); δ = 1.66 – 1.72 (m, 1H), 1.85 – 1.92 (m, 1H), 1.97 – 2.11 (m, 1H), 2.23 – 2.33 (m, 1H), 4.63 (s, 1H), 7.02 – 7.11 (m, 3H), 7.17 – 7.30 (m, 4H), 7.53 – 7.58 (m, 2H); $^{13}$C-NMR (100 MHz, $CDCl_3$); δ = 18.60, 20.80, 28.77, 58.86, 68.71, 81.91, 119.17, 122.02, 125.30, 127.02, 128.73, 131.14, 136.24, 137.30, 161.71, 173.35; HRMS (ESI): m/z calcd for $C_{20}H_{19}BrN_2O_2$ [M+Na] 321.1866 found 321.1869.

***Spectral data:***

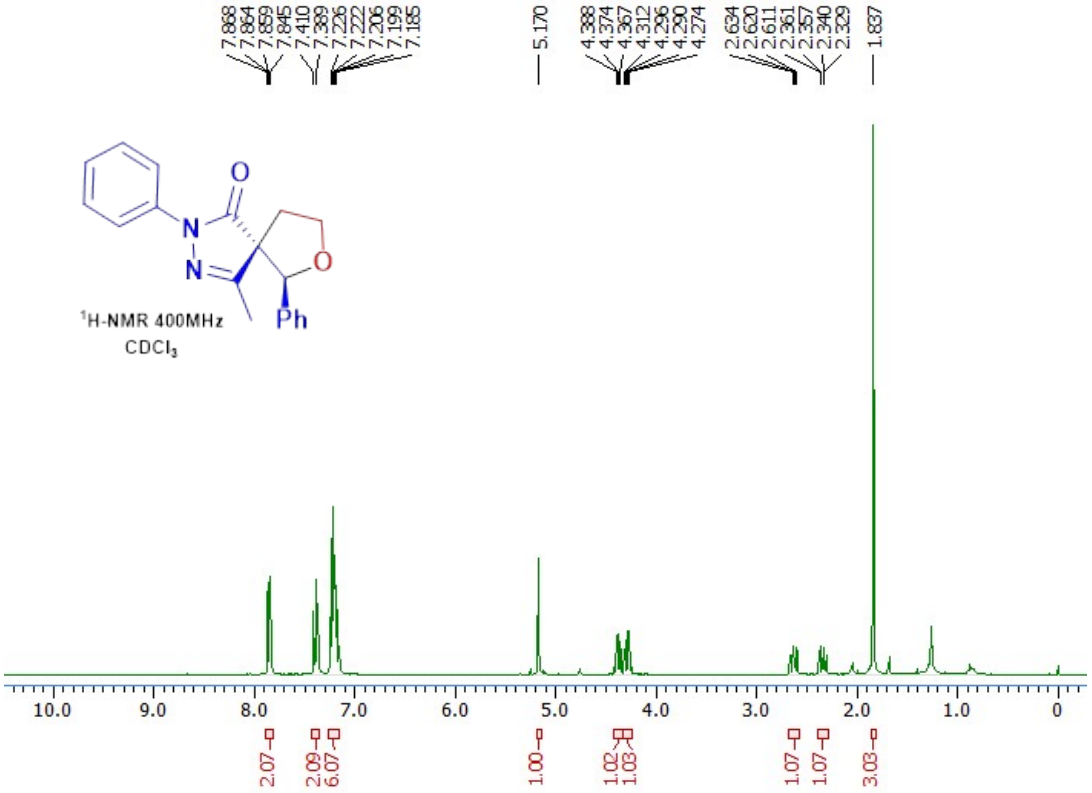

7.868
7.864
7.859
7.845
7.410
7.389
7.226
7.222
7.206
7.199
7.185
5.170
4.388
4.374
4.367
4.312
4.296
4.290
4.274
2.634
2.620
2.611
2.361
2.357
2.340
2.329
1.837
O
N
N
O
Ph
1H-NMR 400MHz
CDCl3
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
2.07
2.09
6.07
1.00
1.02
1.03
1.07
1.07
3.03

**Fig. 3: $^{1}$H-NMR spectrum of (5*S*,6*S*)-4-methyl-2,6-diphenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3a)**

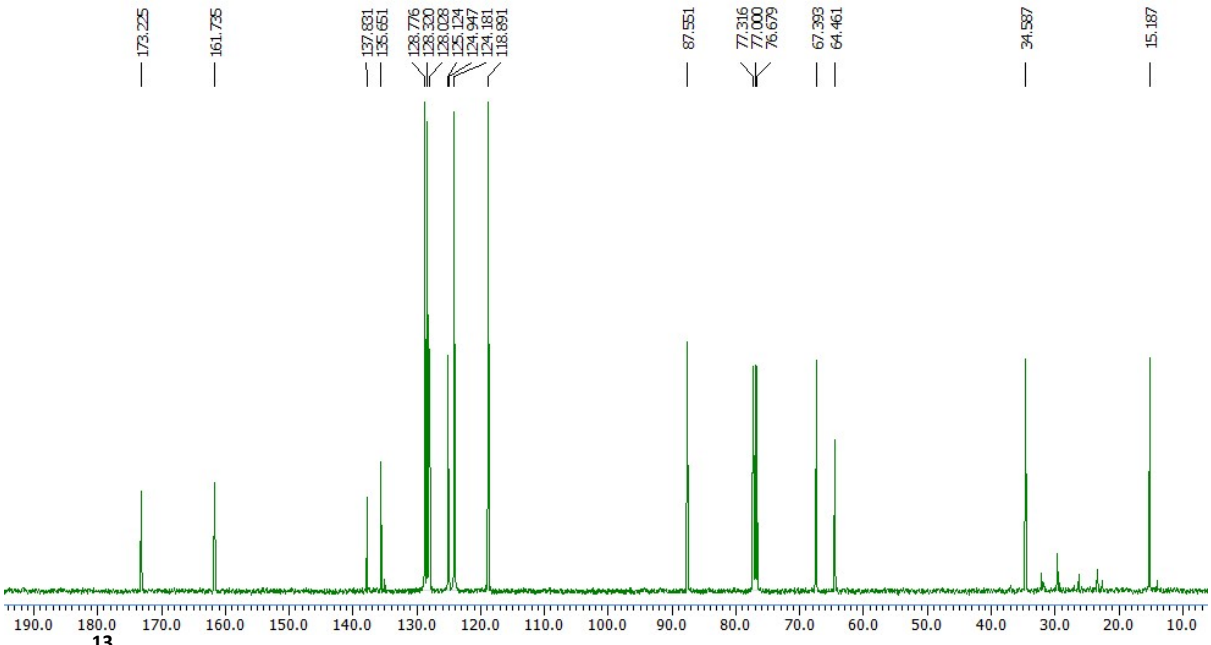



**Fig. 4: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-2,6-diphenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3a)**

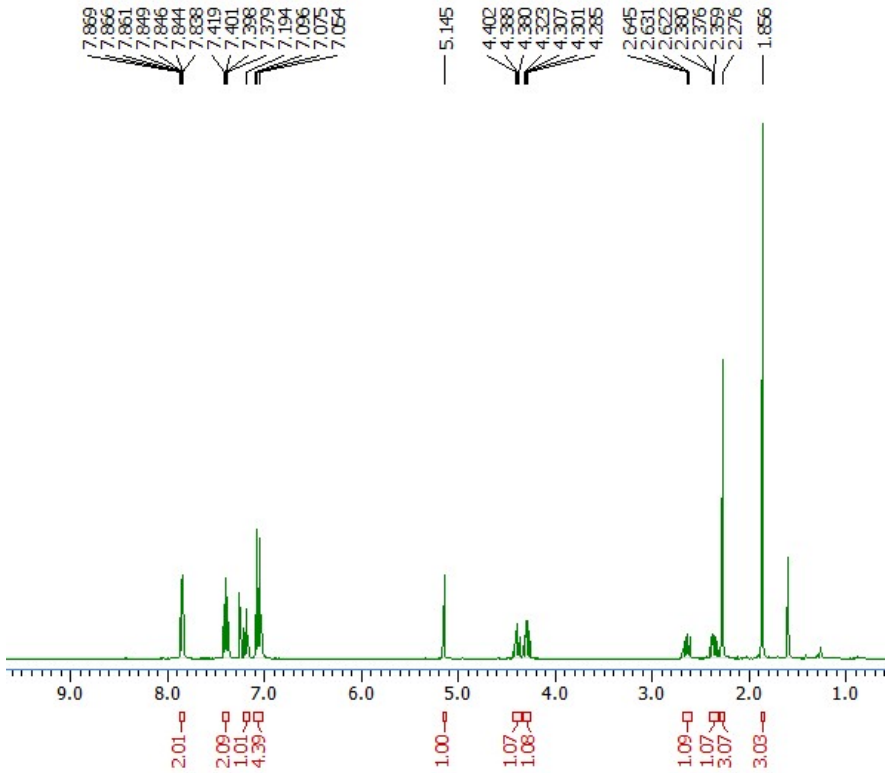



**Fig. 5: $^{1}$H-NMR spectrum of (5*S*,6*S*)-4-methyl-2-phenyl-6-(p-tolyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3b)**

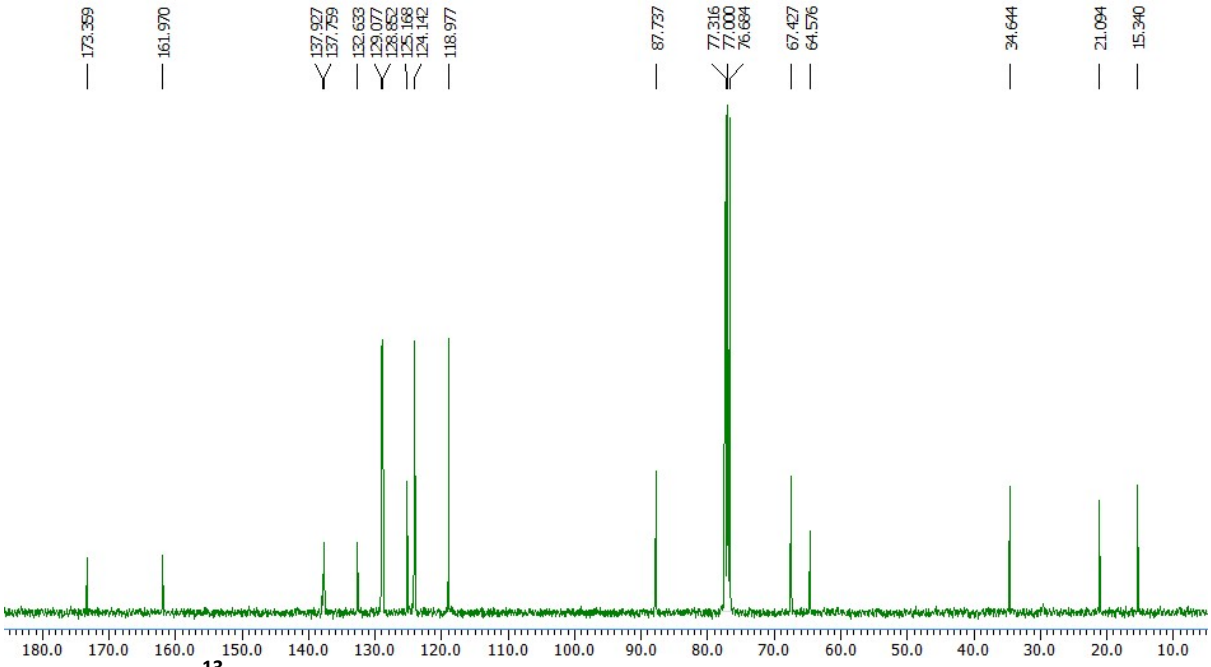
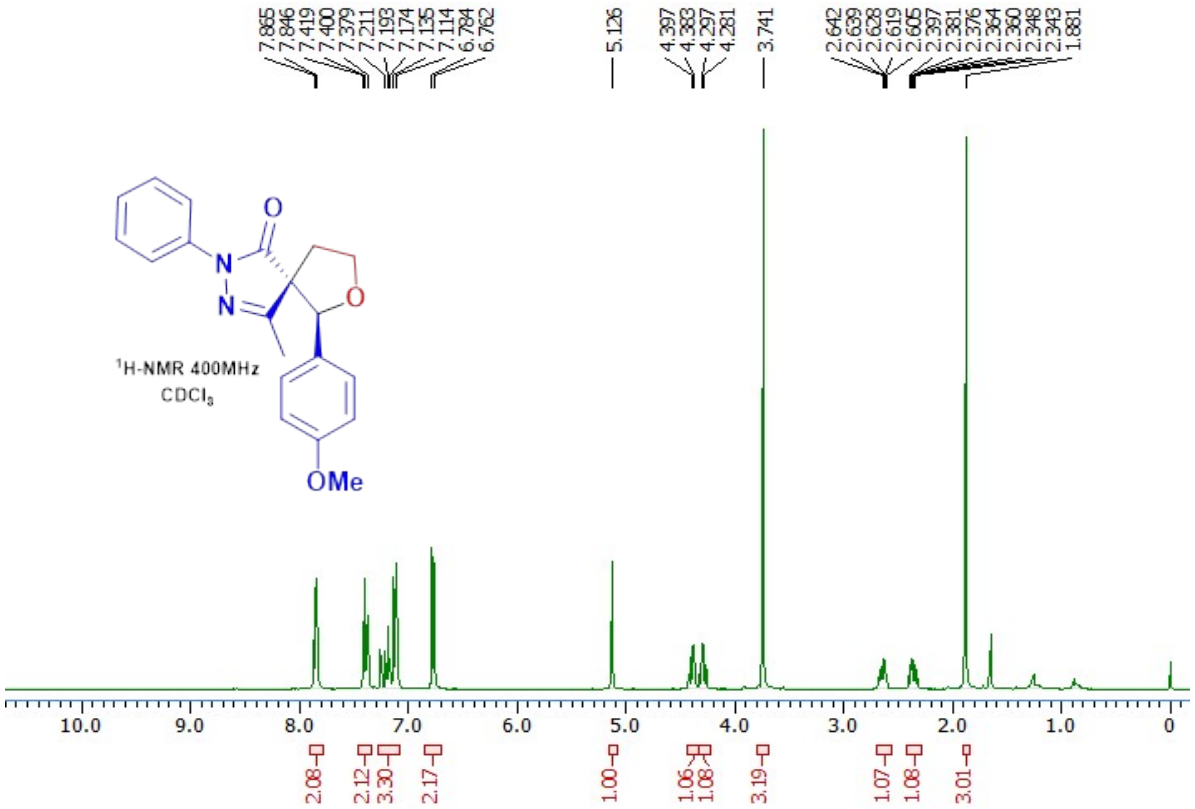



**Fig. 6: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-2-phenyl-6-(p-tolyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3b)**


7.885
7.846
7.419
7.400
7.379
7.211
7.193
7.174
7.135
7.114
6.784
6.762
5.126
4.397
4.383
4.297
4.281
3.741
2.642
2.639
2.628
2.619
2.605
2.397
2.381
2.376
2.364
2.360
2.348
2.343
1.881
O
N
N
O
1H-NMR 400MHz
CDCl3
OMe
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0
2.08
2.12
3.30
2.17
1.00
1.06
1.08
3.19
1.07
1.08
3.01

**diazaspiro[4.4]non-3-en-1-one (3c)**

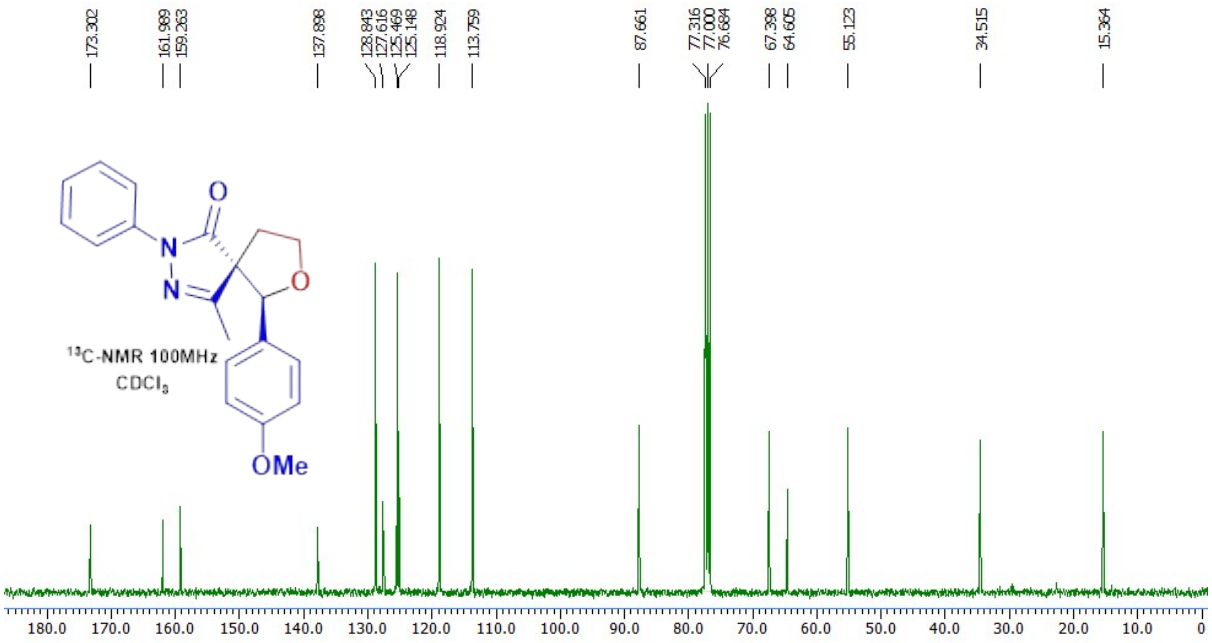


**Fig. 8: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3c)**

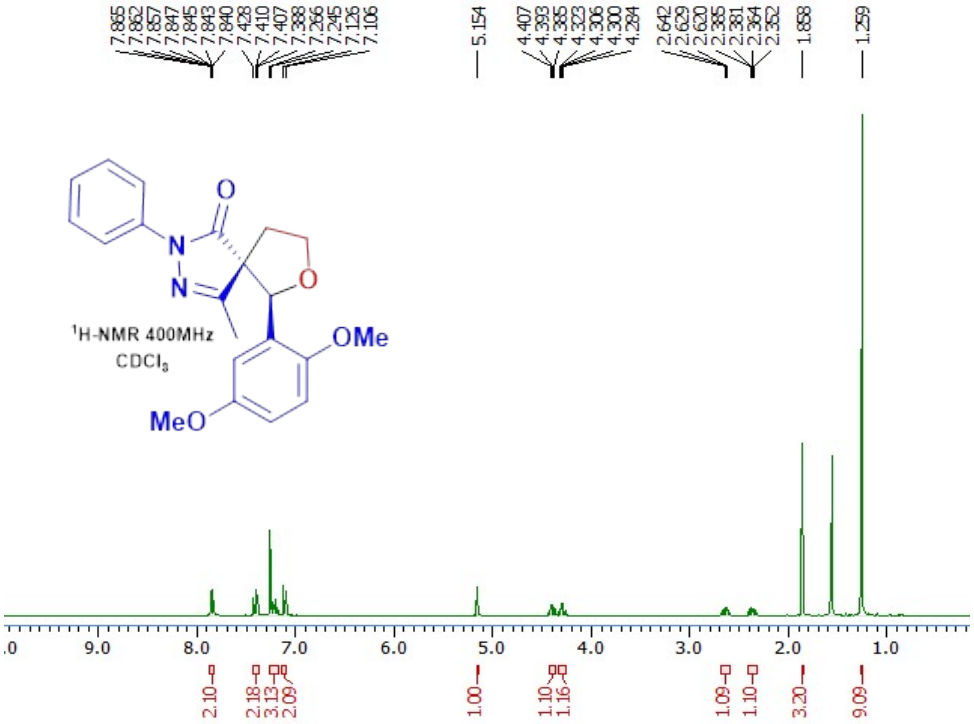


**Fig. 9: $^{1}$H-NMR spectrum of (5*S*,6*S*)-6-(2,5-dimethoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3d)**

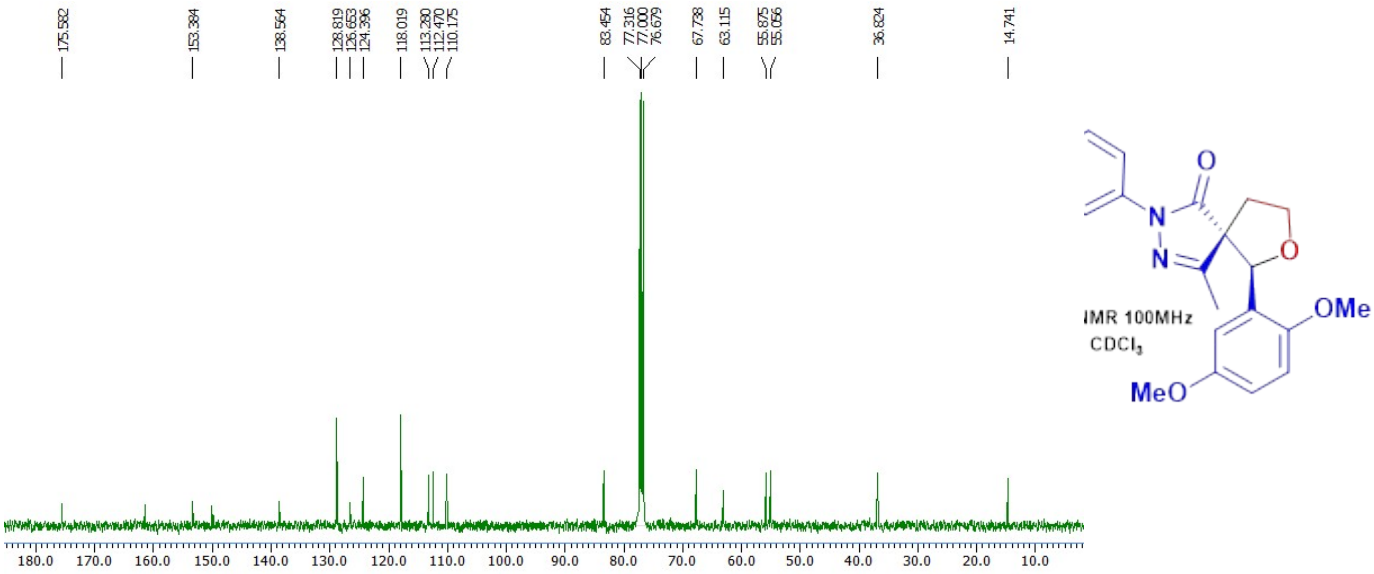



**Fig. 10: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(2,5-dimethoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3d)**

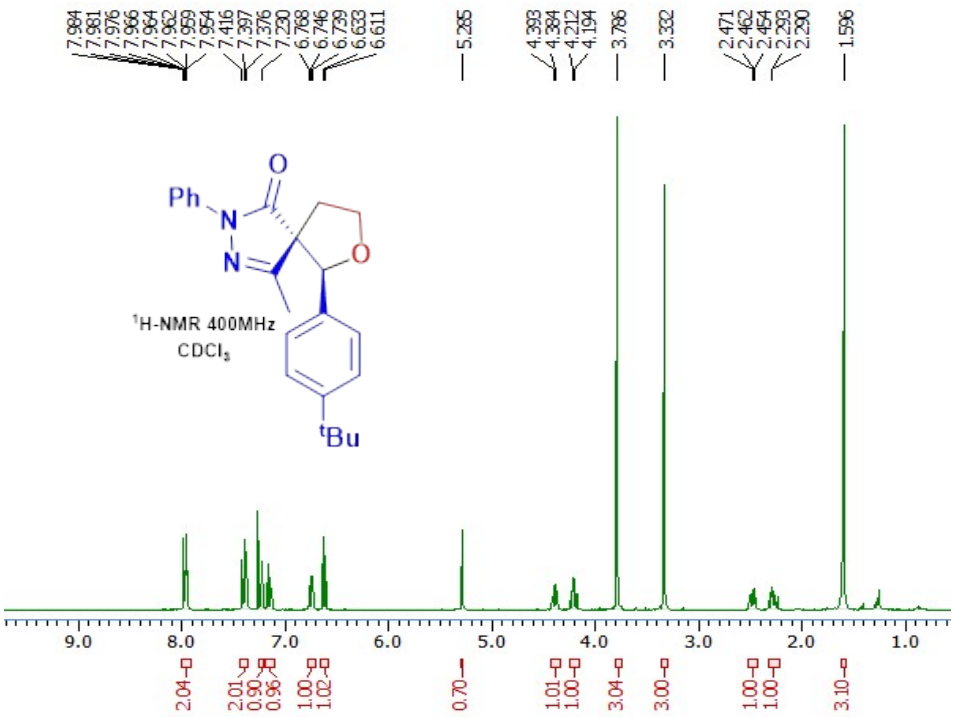


**Fig. 11: $^{1}$H-NMR spectrum of (5*S*,6*S*)-6-(4-(tert-butyl)phenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3e)**

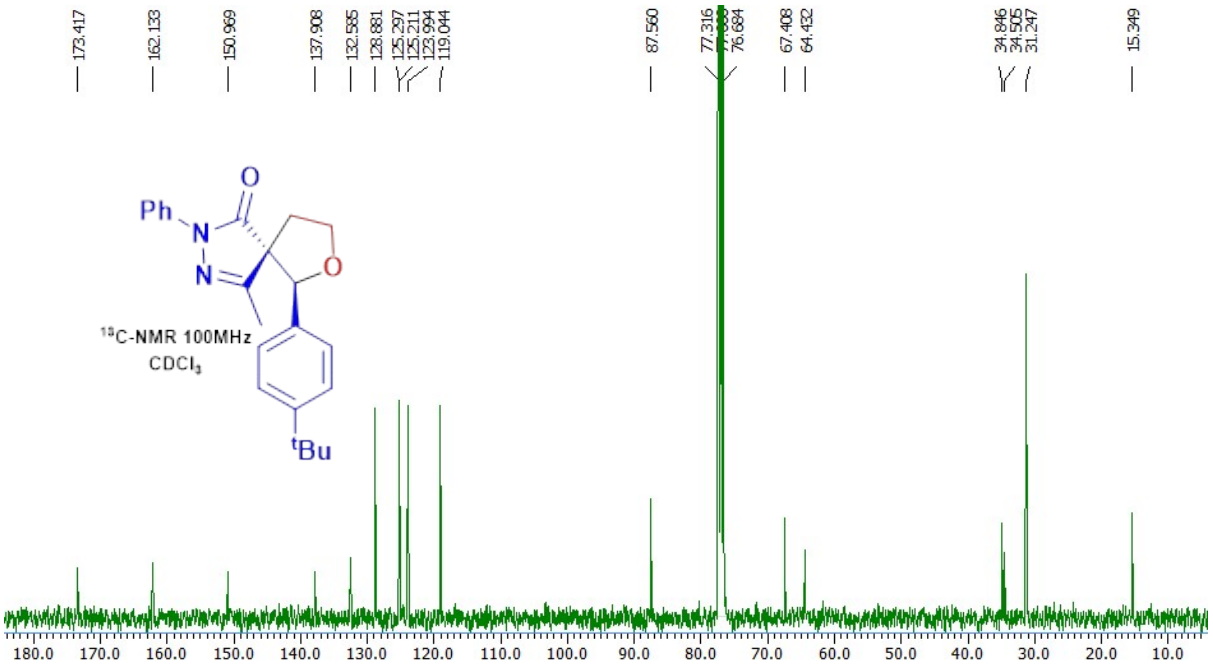
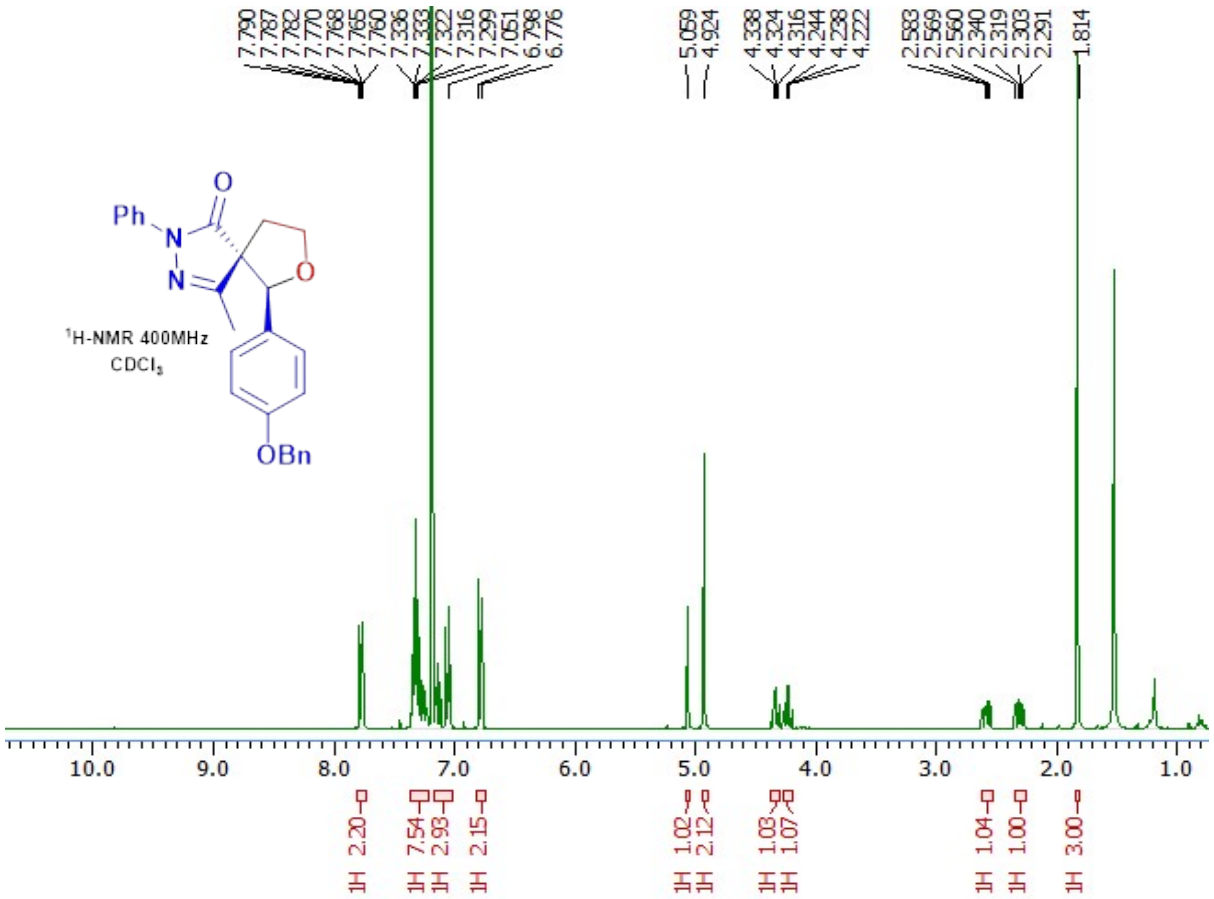
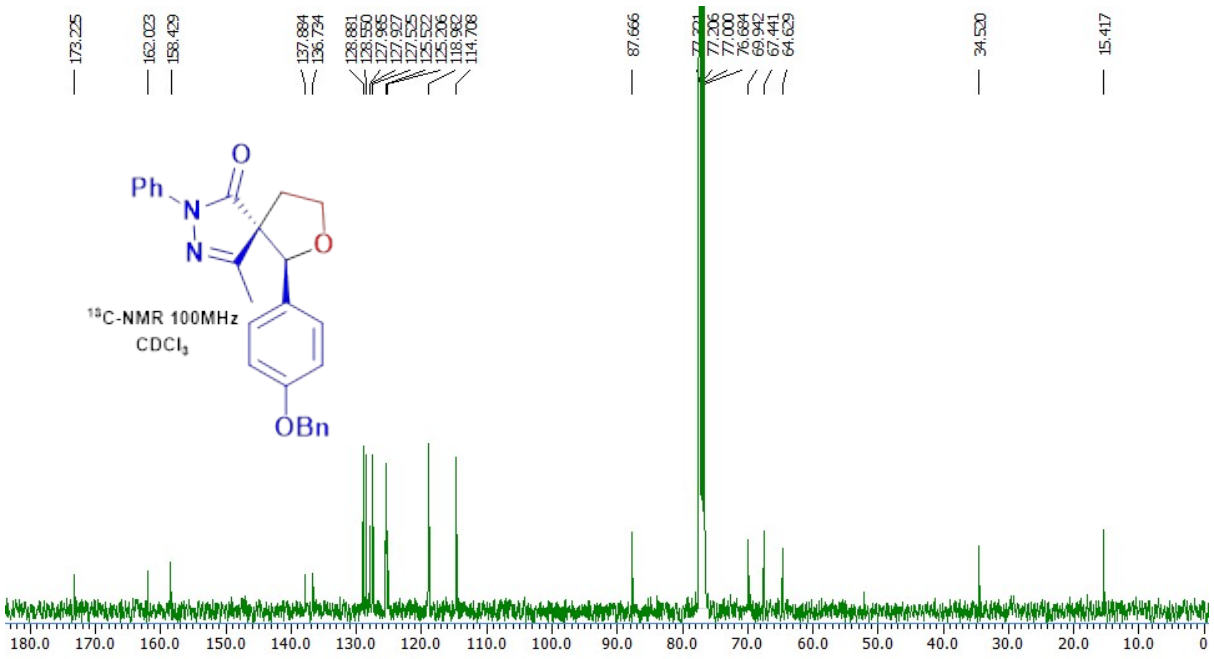


**Fig. 12: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-(tert-butyl)phenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3e)**


7.790
7.787
7.782
7.770
7.768
7.765
7.760
7.336
7.333
7.322
7.316
7.299
7.051
6.798
6.776
5.059
4.924
4.338
4.324
4.316
4.244
4.238
4.222
2.583
2.569
2.560
2.340
2.319
2.303
2.291
1.814
Ph
N
N
O
O
OBn
$^1$H-NMR 400MHz
$CDCl_3$
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
2.20
7.54
2.93
2.15
1.02
2.12
1.03
1.07
1.04
1.00
3.00
1H


173.225
162.023
158.429
137.884
136.734
128.881
128.590
127.985
127.927
127.525
125.522
125.206
118.982
114.708
87.666
77.221
77.206
77.000
76.684
69.942
67.441
64.629
34.520
15.417
Ph
N
N
O
O
OBn
$^{13}$C-NMR 100MHz
$CDCl_3$
180.0
170.0
160.0
150.0
140.0
130.0
120.0
110.0
100.0
90.0
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0

**diazaspiro[4.4]non-3-en-1-one (3f)**

**Fig. 14: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-(benzyloxy)phenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3f)**

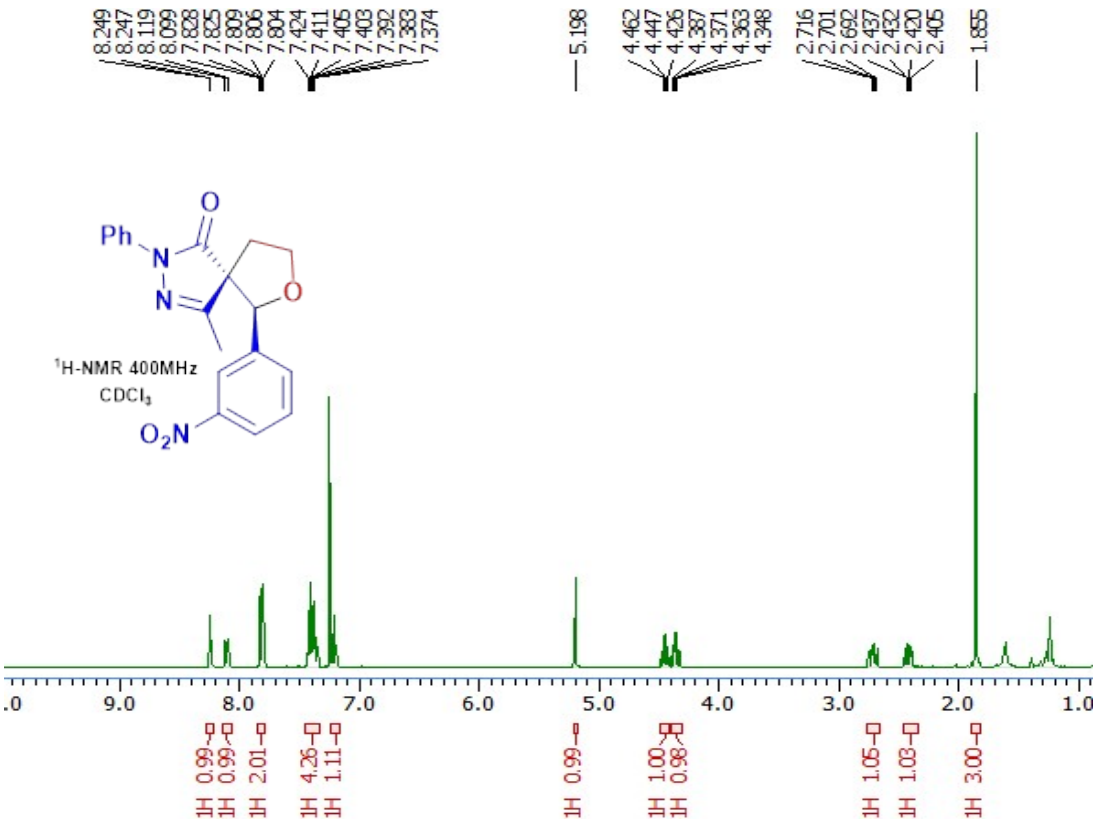
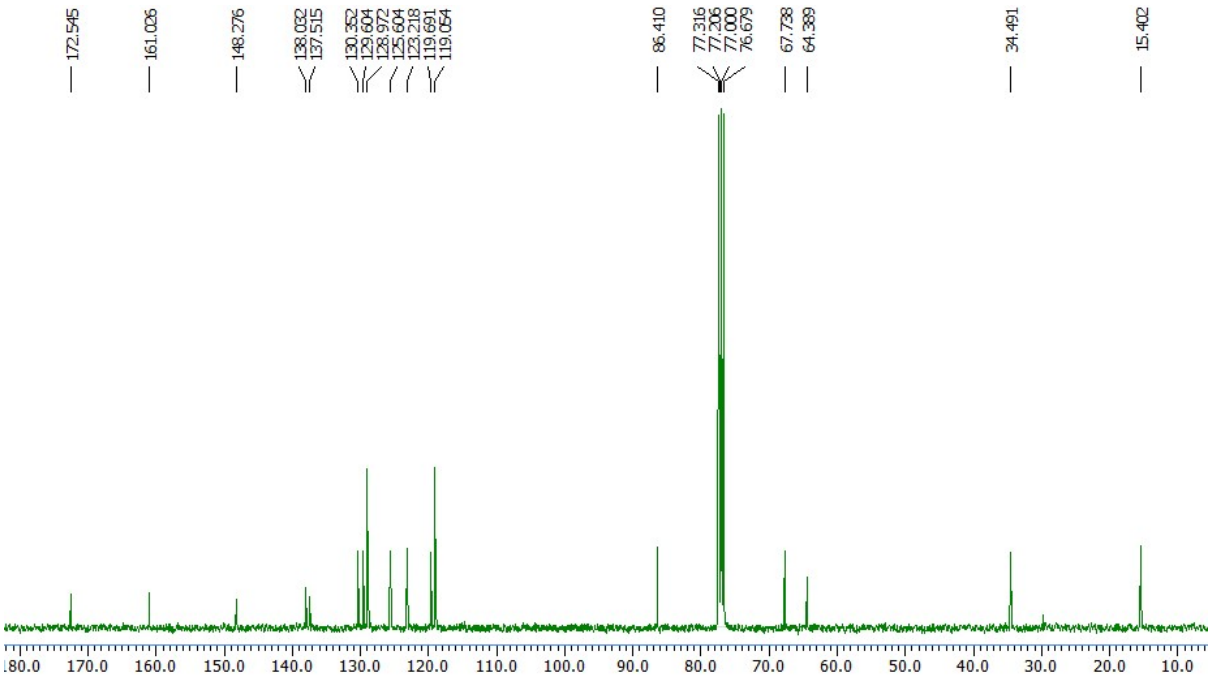



**Fig. 15: $^1$H-**



**NMR spectrum of (5*S*,6*S*)-4-methyl-6-(3-nitrophenyl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3g)**

**Fig. 16: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-6-(3-nitrophenyl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3g)**

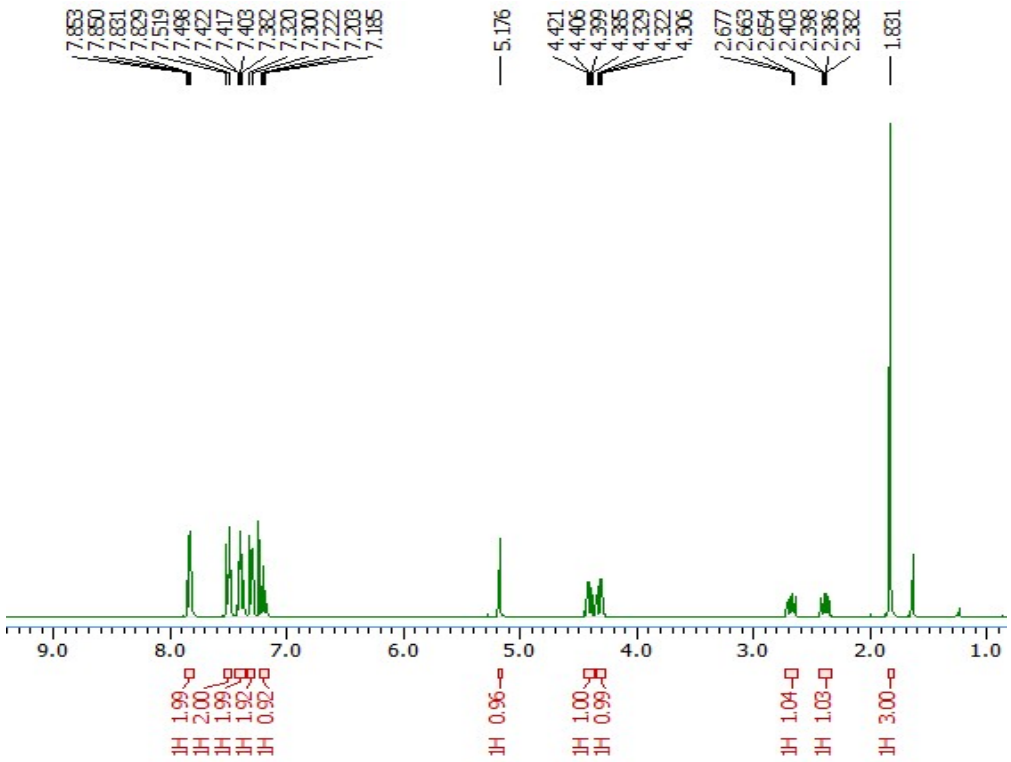



**Fig. 17: $^{1}$H-NMR spectrum of (5*S*,6*S*)-4-methyl-2-phenyl-6-(4-(trifluoromethyl)phenyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3h)**

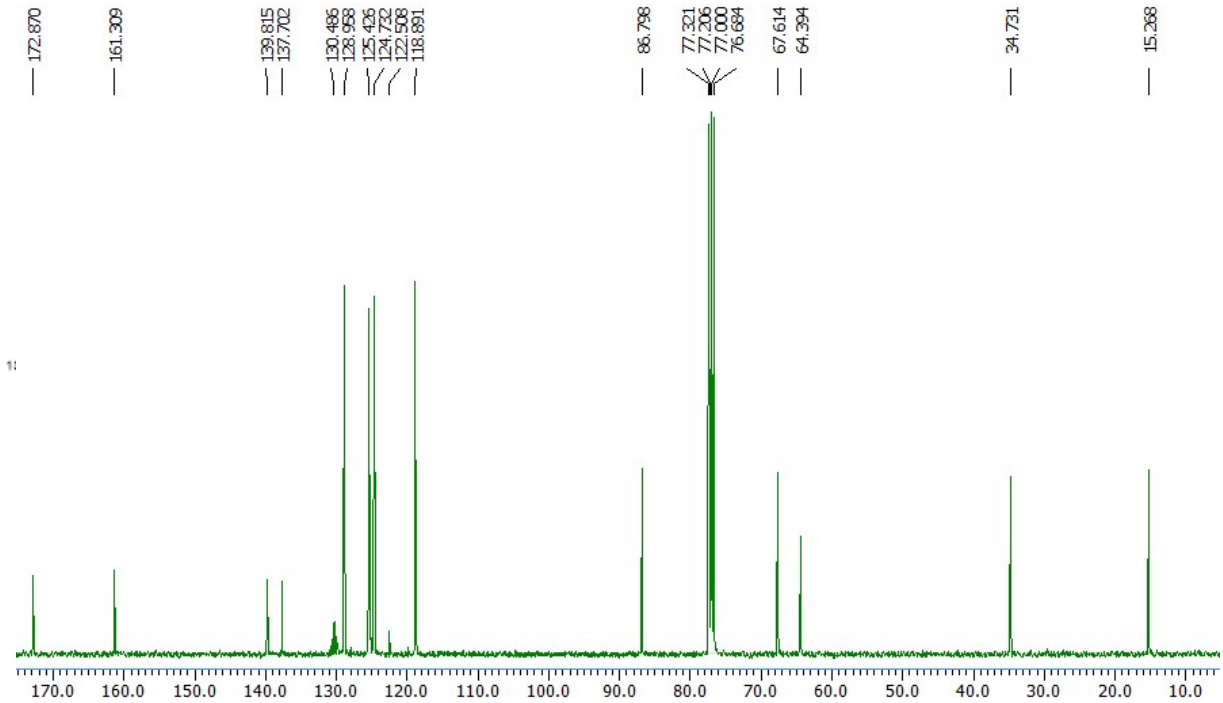


**Fig. 18: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-2-phenyl-6-(4-(trifluoromethyl)phenyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3h)**

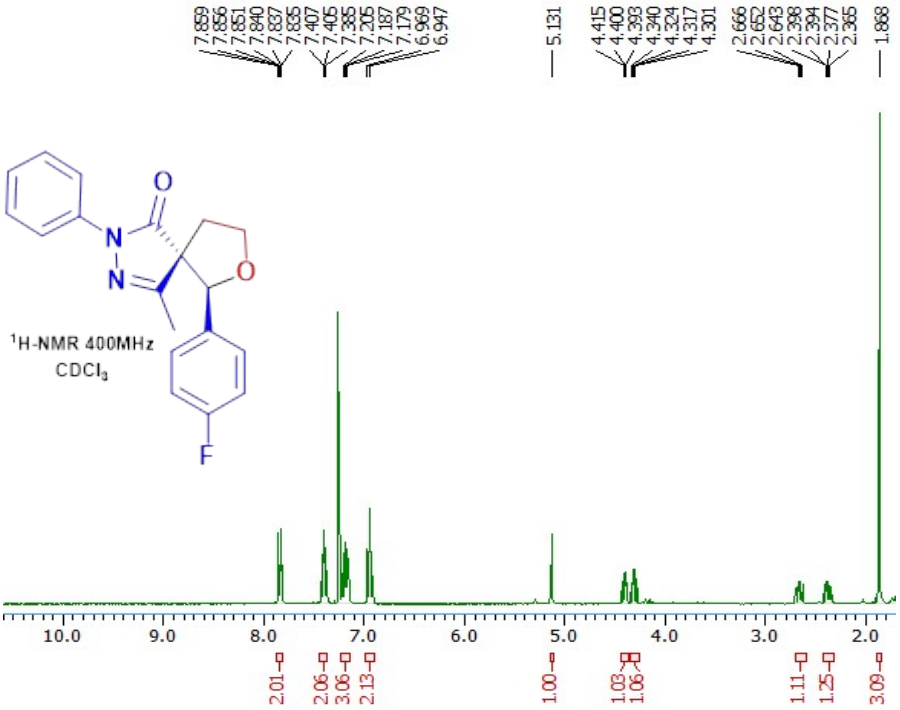
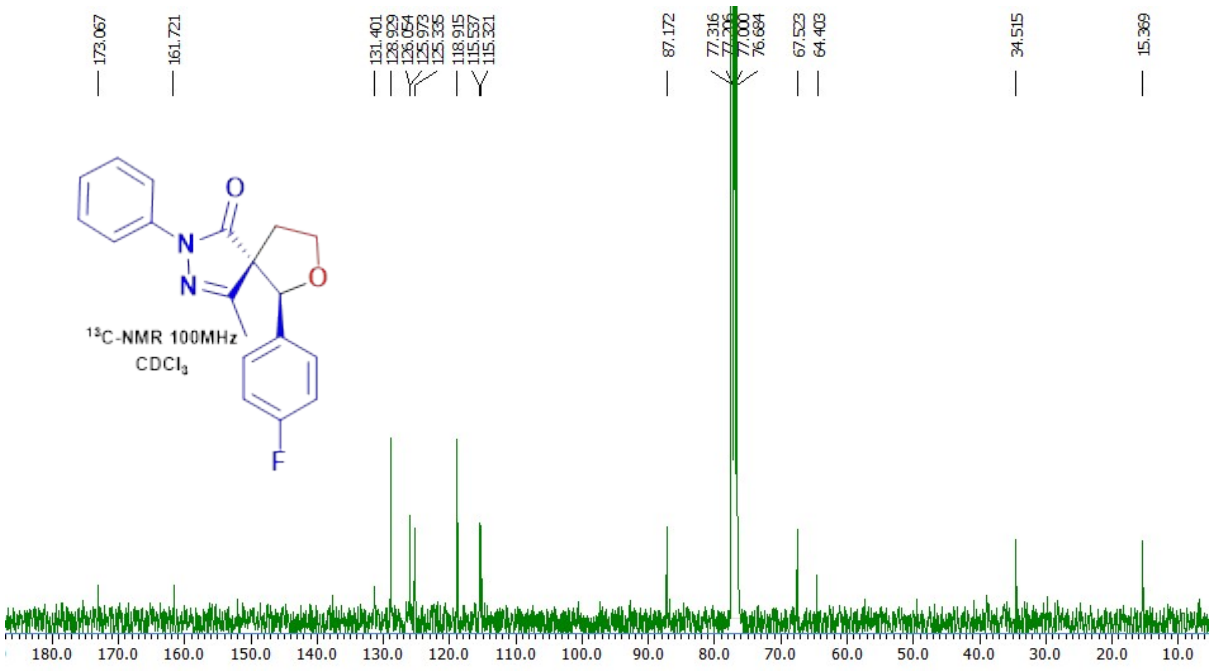



**Fig. 19:**

**$^{1}$H-NMR**



**spectrum of (5*S*,6*S*)-6-(4-fluorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one(3i)**

**Fig. 20: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-fluorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3i)**

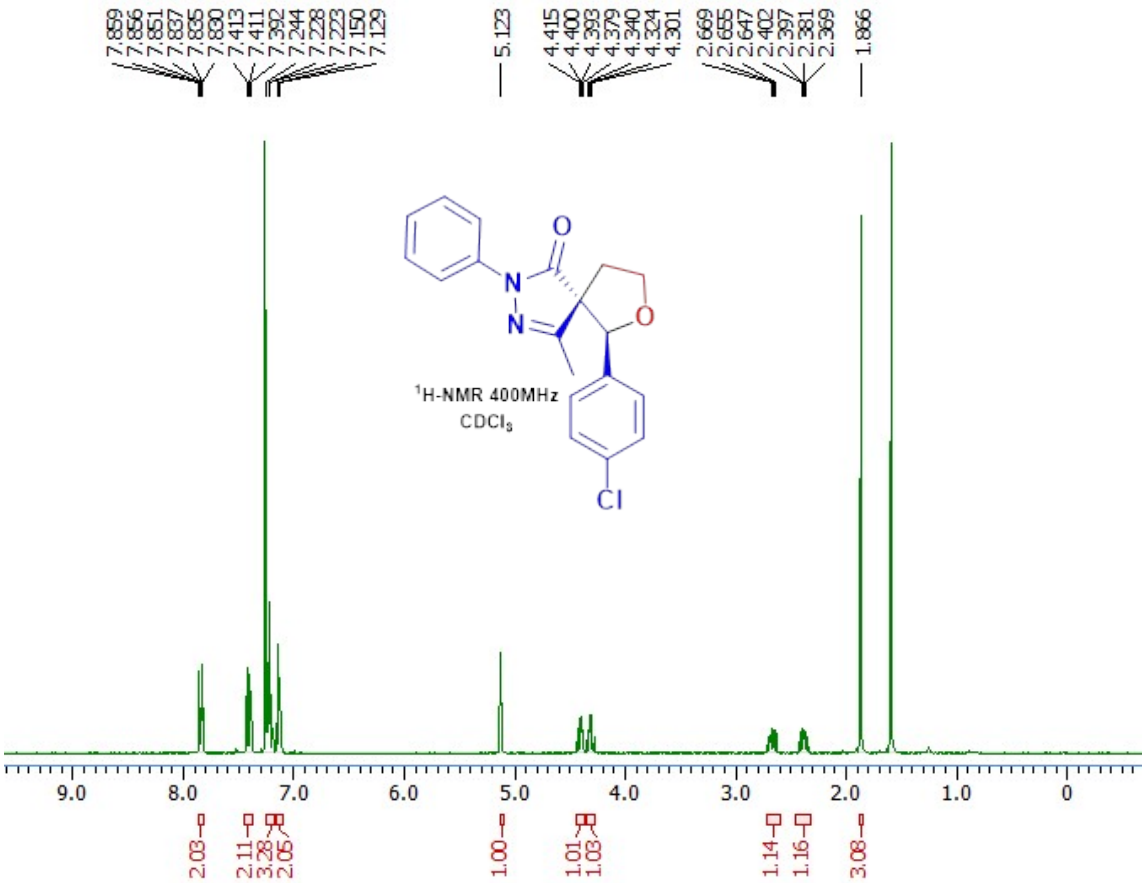


**Fig. 21: $^{1}$H-NMR spectrum of (5*S*,6*S*)-6-(4-chlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one(3j)**

**Fig. 22: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-chlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one(3j)**

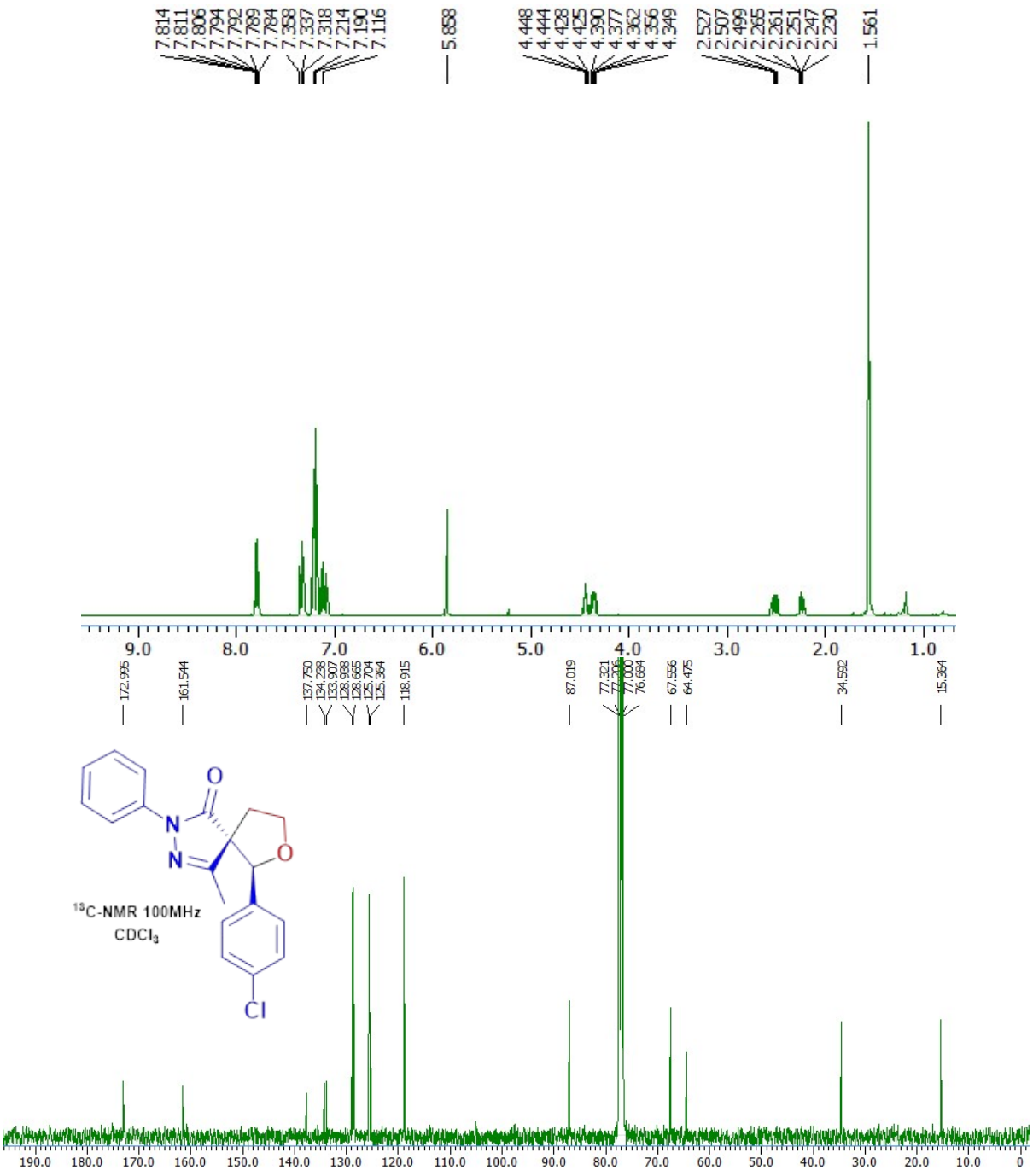


**Fig. 23: $^{1}$H-NMR spectrum of (5*S*,6*R*)-6-(2,6-dichlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3k)**

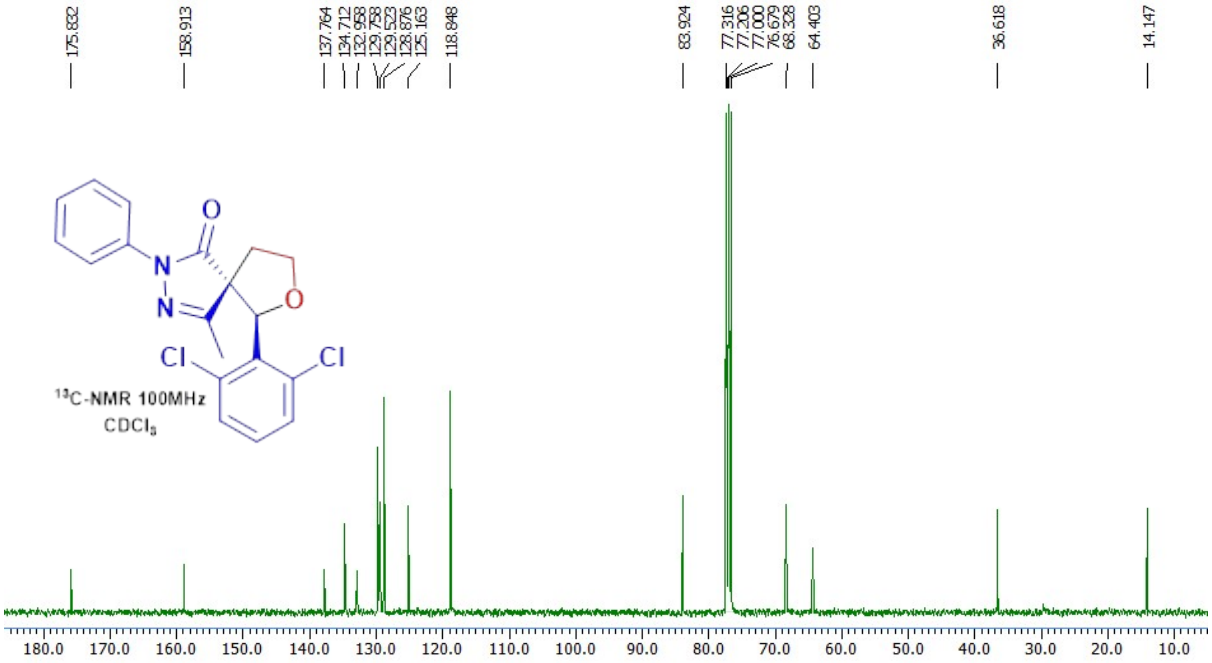


**Fig. 24: $^{13}$C-NMR spectrum of (5*S*,6*R*)-6-(2,6-dichlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3k)**

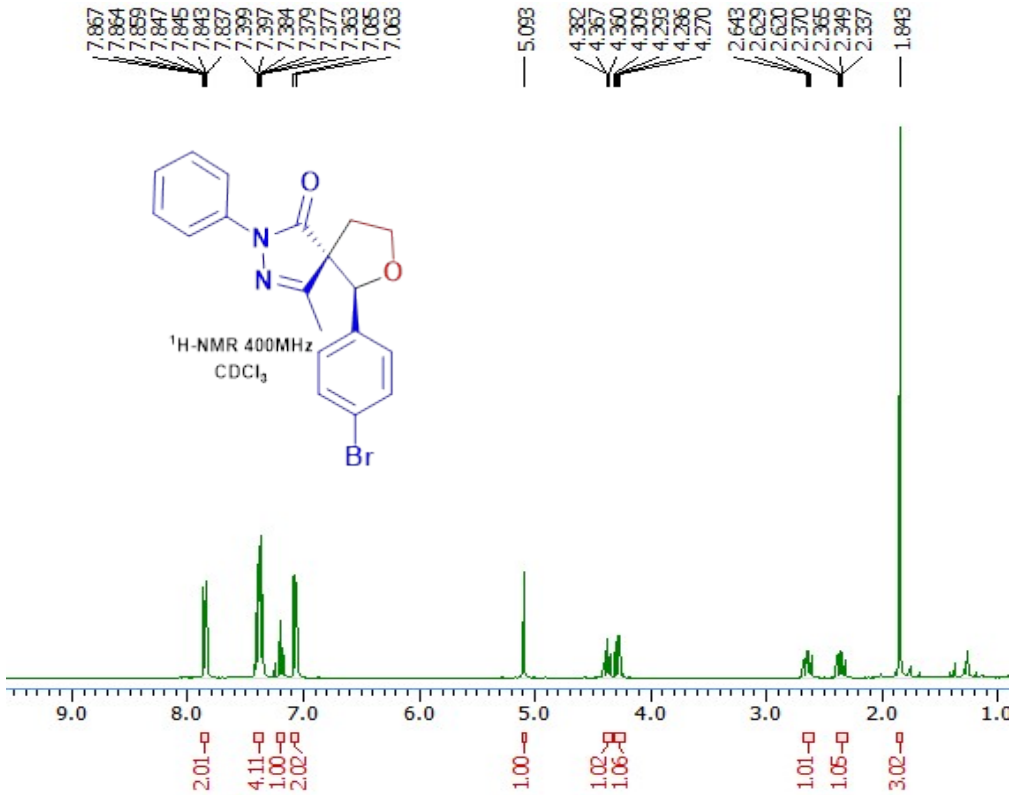



**Fig. 25: $^1$H-NMR**

**spectrum of (5*S*,6*S*)-6-(4-bromophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one(3l)**

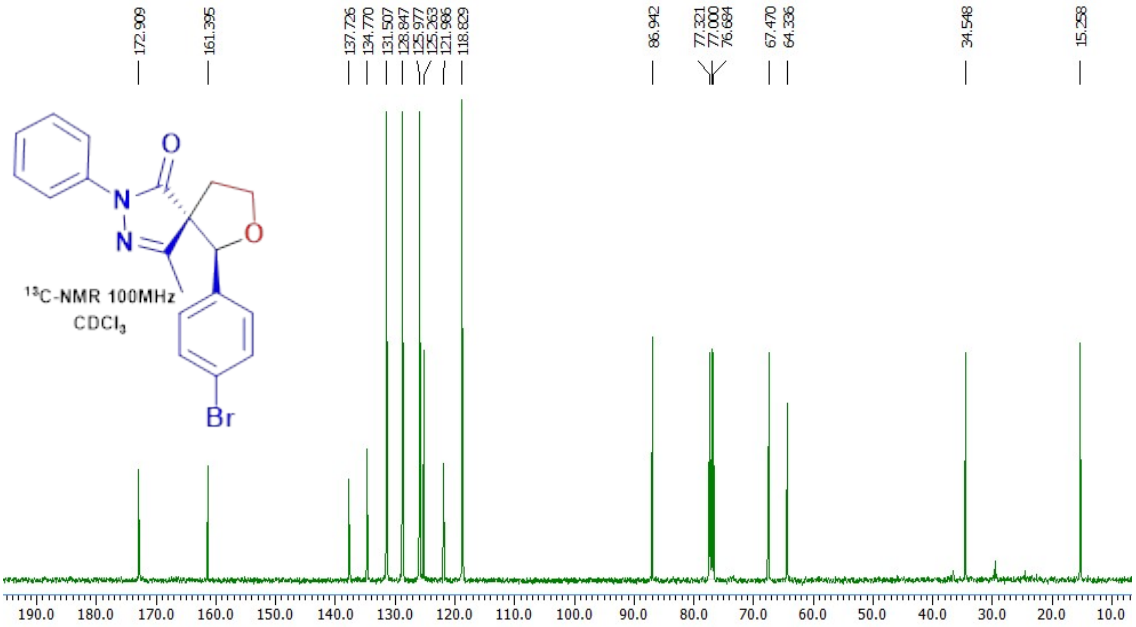



**Fig. 26: $^{13}$C-**

**NMR spectrum of (5*S*,6*S*)-6-(4-bromophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3l)**

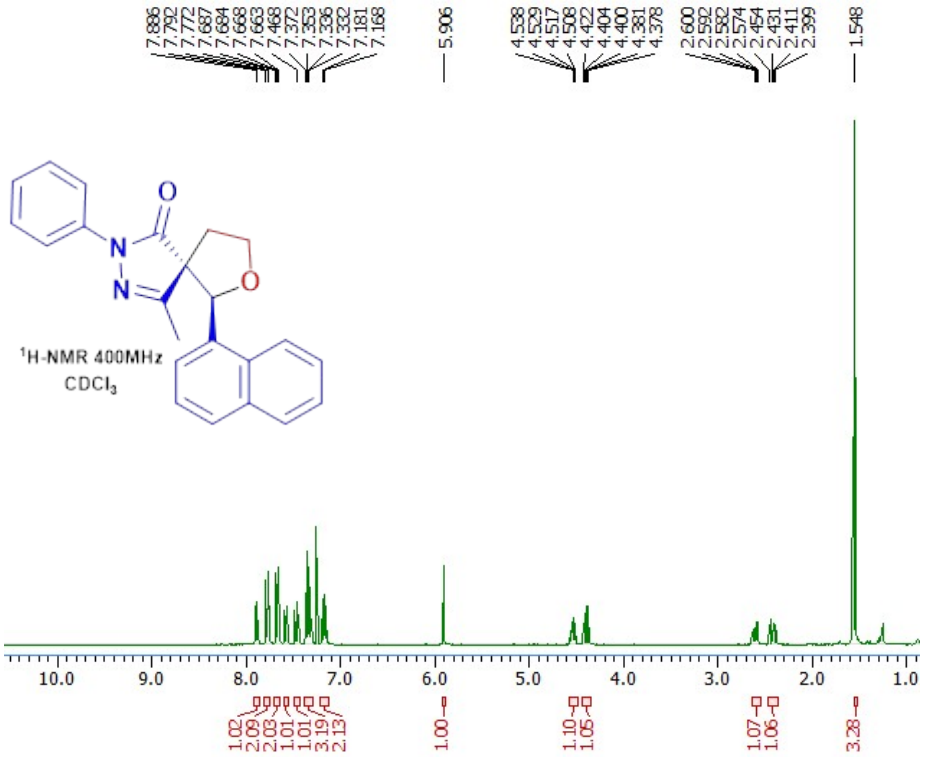
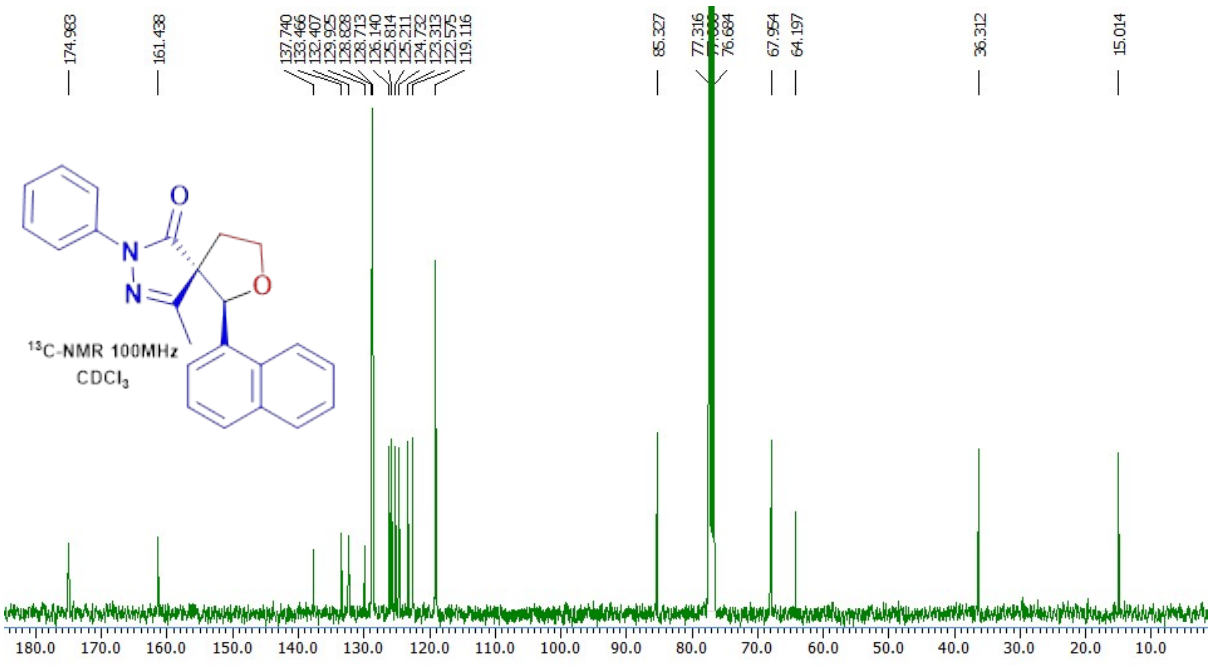
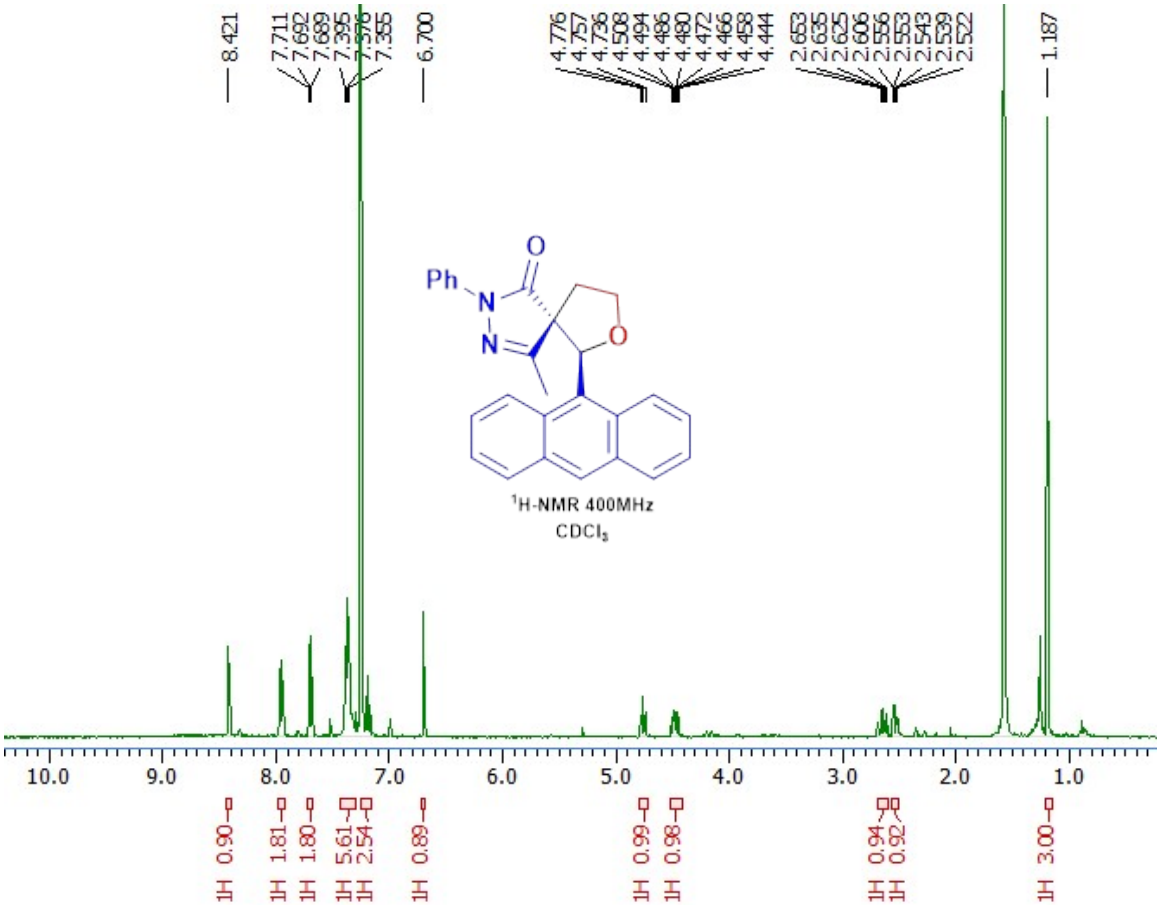


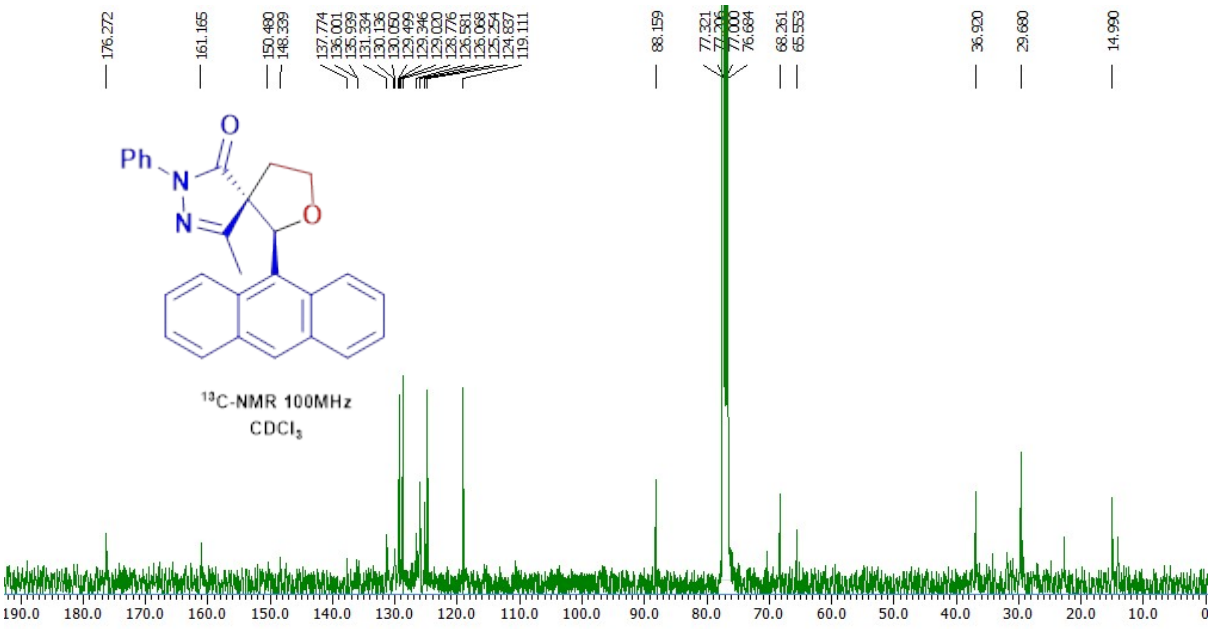

**Fig. 27: $^1$H-NMR spectrum**



**of (5*S*,6*S*)-4-methyl-6-(naphthalen-1-yl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3m)**

**Fig. 28: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-6-(naphthalen-1-yl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3m)**


8.421
7.711
7.692
7.689
7.395
7.376
7.355
6.700
4.776
4.757
4.736
4.508
4.494
4.486
4.480
4.472
4.466
4.458
4.444
2.653
2.635
2.625
2.606
2.556
2.553
2.543
2.539
2.522
1.187
Ph
N
N
O
O
$^{1}$H-NMR 400MHz
$CDCl_3$
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0.90
1.81
1.80
5.61
2.54
0.89
0.99
0.98
0.94
0.92
3.00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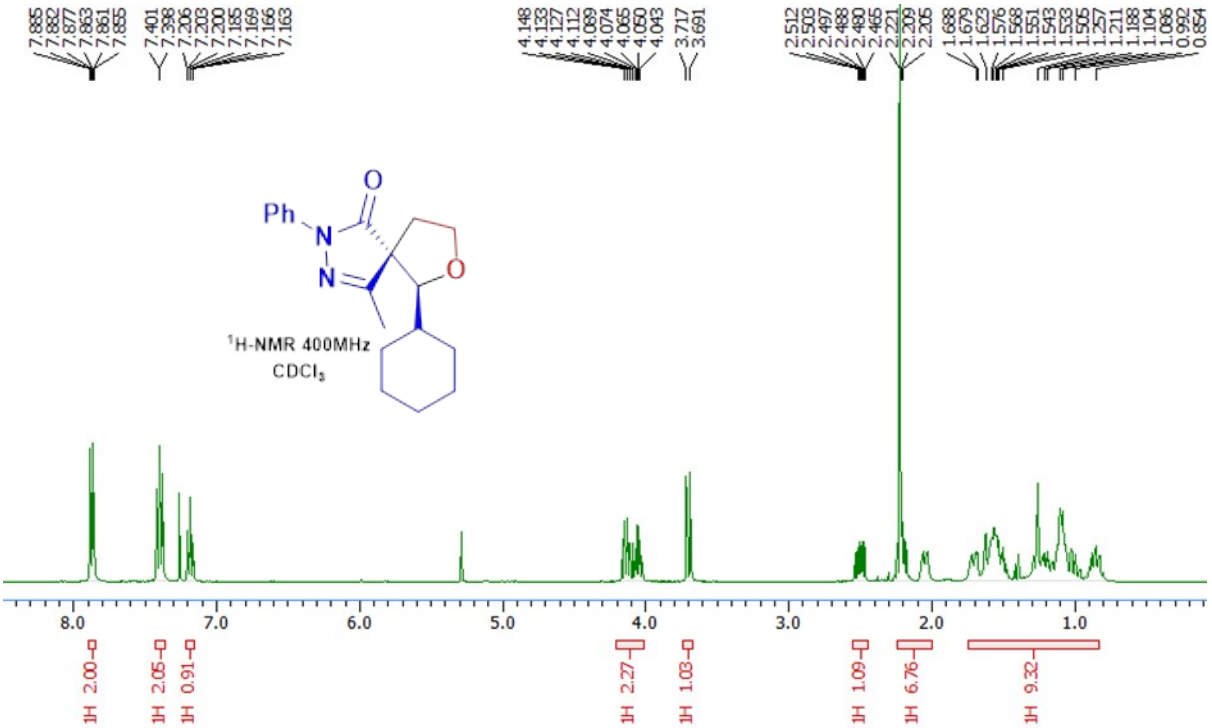
161.165
150.480
148.339
137.774
136.001
135.939
131.334
130.136
130.050
129.499
129.346
129.020
128.776
126.581
126.068
125.254
124.837
119.111
88.159
77.321
77.000
76.684
68.261
65.553
36.920
29.680
14.990
Ph
N
N
O
O
$^{13}$C-NMR 100MHz
$CDCl_3$
190.0
180.0
170.0
160.0
150.0
140.0
130.0
120.0
110.0
100.0
90.0
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0
0

**diazaspiro[4.4]non-3-en-1-one (3n)**

**Fig. 30: $^{13}$C-NMR spectrum of (5S,6S)-6-(anthracen-9-yl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3n)**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7.855
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7.169
7.166
7.163
4.148
4.133
4.127
4.112
4.089
4.074
4.065
4.050
4.043
3.717
3.691
2.512
2.503
2.497
2.488
2.480
2.465
2.221
2.209
2.205
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1.679
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1.576
1.568
1.551
1.543
1.533
1.505
1.257
1.211
1.188
1.104
1.066
0.992
0.854
Ph
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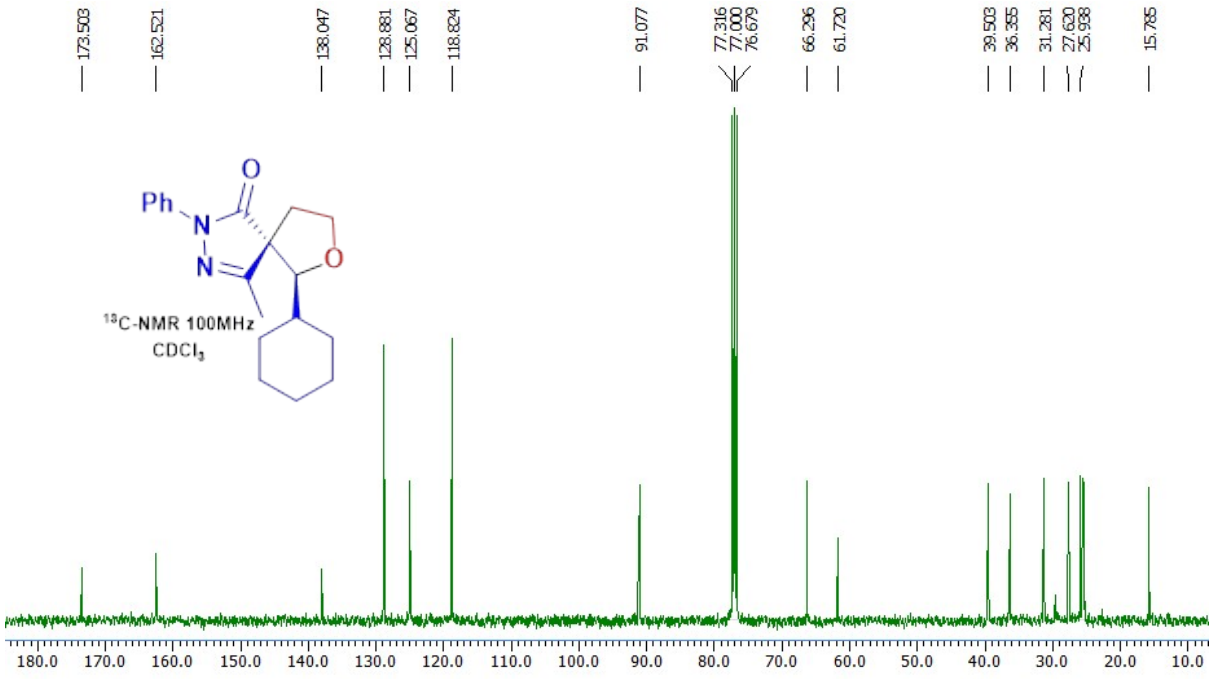
N
O
O
$^1$H-NMR 400MHz
$CDCl_3$
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
1H 2.00
1H 2.05
1H 0.91
1H 2.27
1H 1.03
1H 1.09
1H 6.76
1H 9.32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138.047
128.881
125.067
118.824
91.077
77.316
77.000
76.679
66.296
61.720
39.503
36.355
31.281
27.620
25.938
15.785
Ph
N
N
O
O
$^{13}$C-NMR 100MHz
$CDCl_3$
180.0
170.0
160.0
150.0
140.0
130.0
120.0
110.0
100.0
90.0
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0

**diazaspiro[4.4]non-3-en-1-one (3o)**

**Fig. 32: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-cyclohexyl-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3o)**

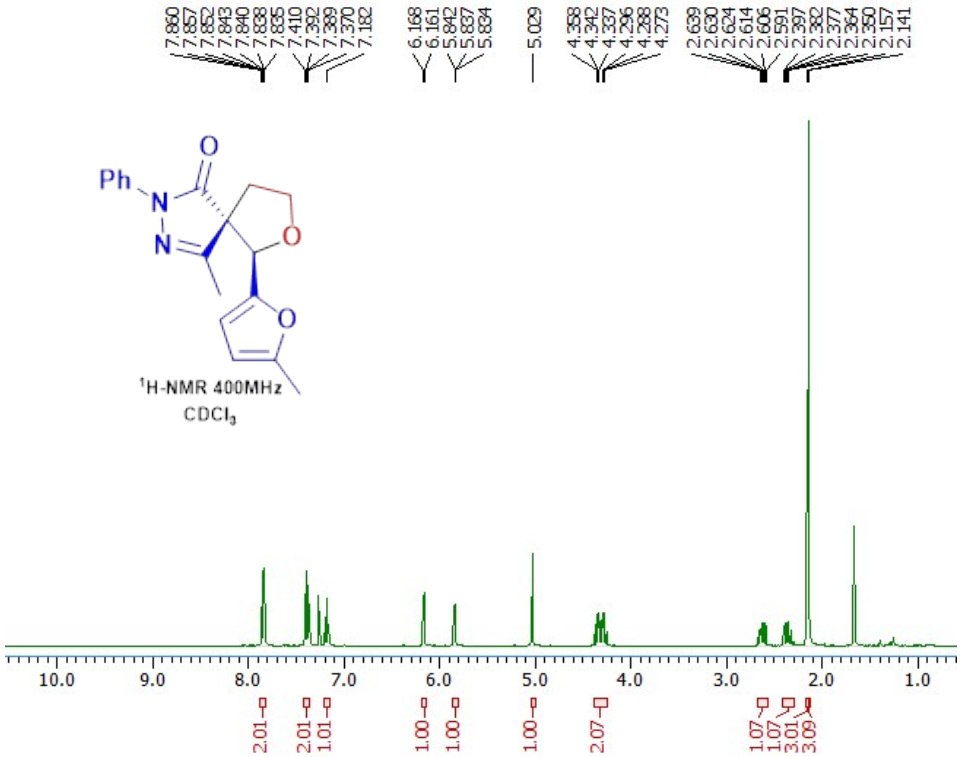



**Fig. 33: $^1$H-NMR**

**spectrum of (5*S*,6*R*)-4-methyl-6-(5-methylfuran-2-yl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3p)**

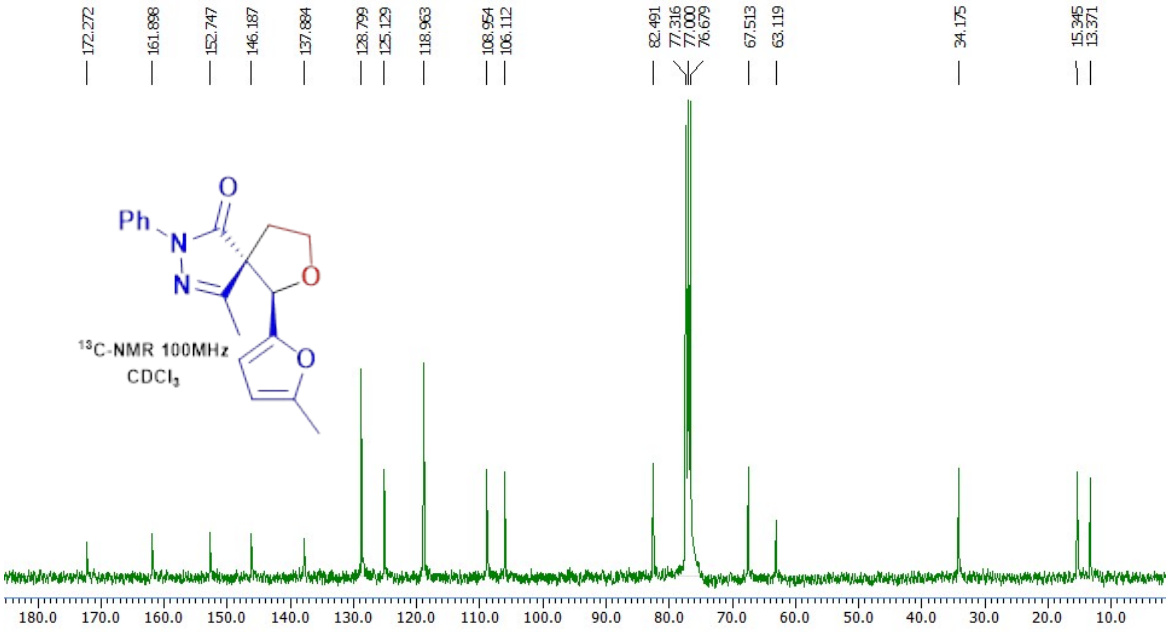



**Fig. 34: $^{13}$C-NMR spectrum of (5*S*,6*R*)-4-methyl-6-(5-methylfuran-2-yl)-2-phenyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3p)**

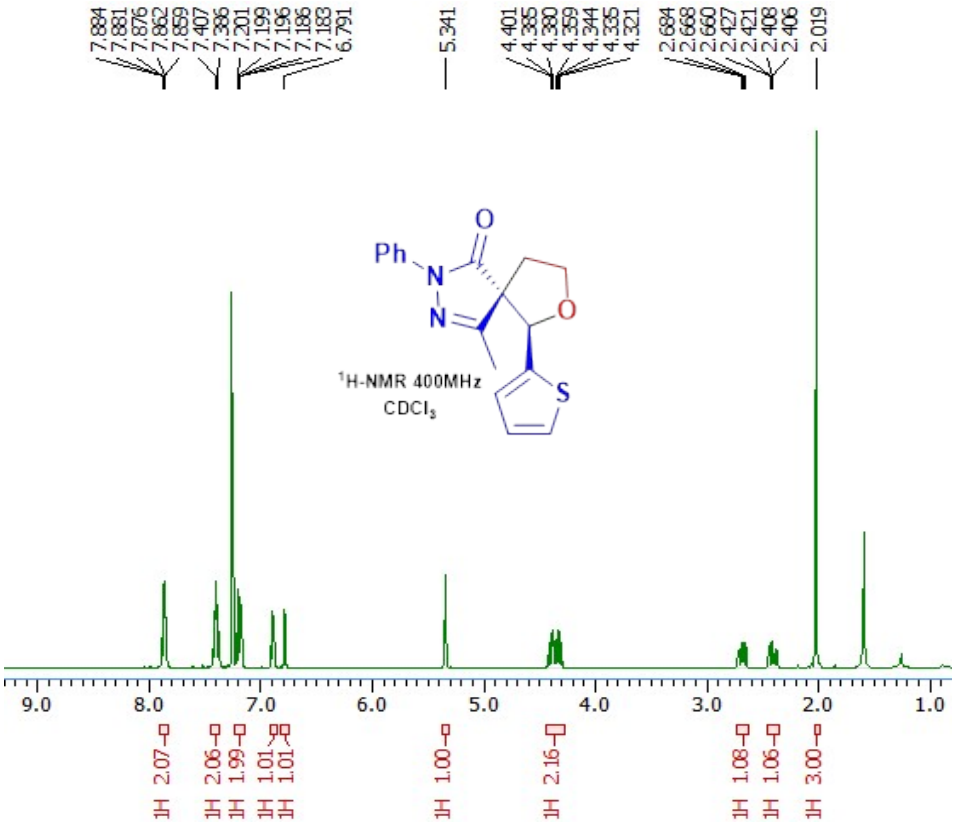


**Fig. 35: $^{1}$H-NMR spectrum of (5*S*,6*R*)-4-methyl-2-phenyl-6-(thiophen-2-yl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3q)**

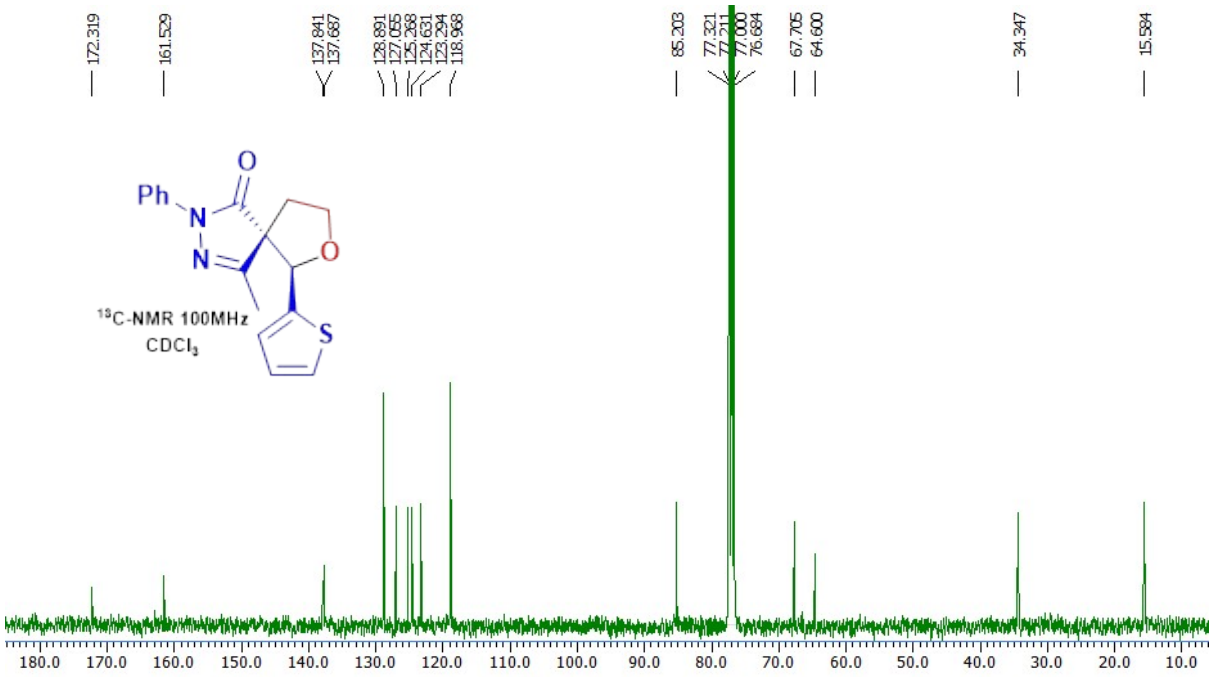


**Fig. 36: $^{13}$C-NMR spectrum of (5*S*,6*R*)-4-methyl-2-phenyl-6-(thiophen-2-yl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3q)**

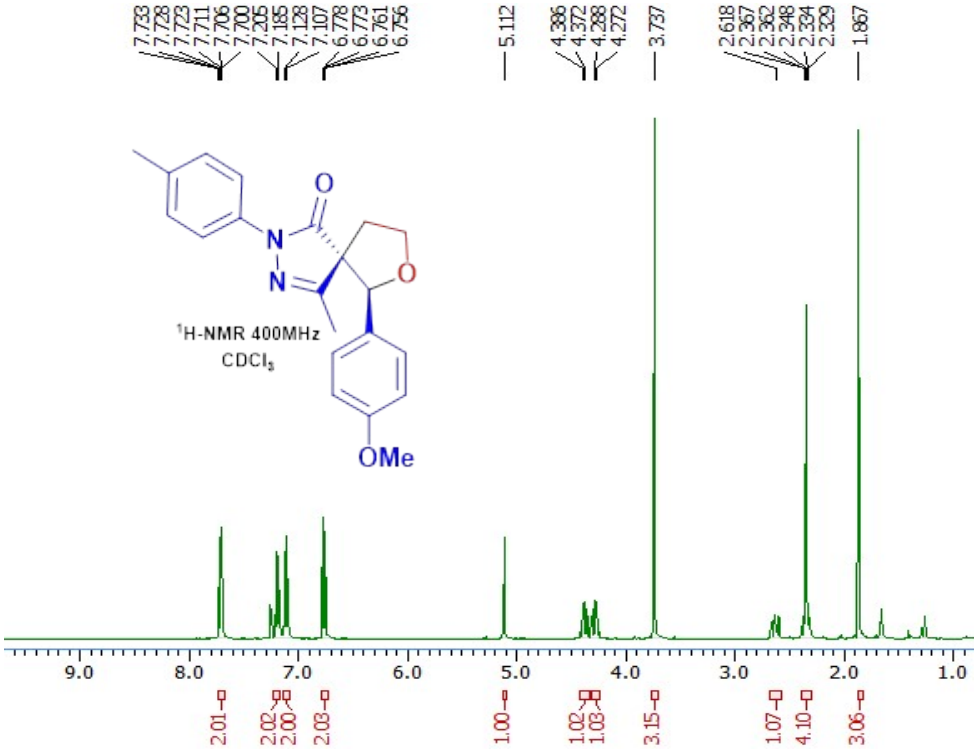



**Fig. 37: $^1$H-NMR**

**spectrum of (5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-(p-tolyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one]non-3-en-1-one(3r)**

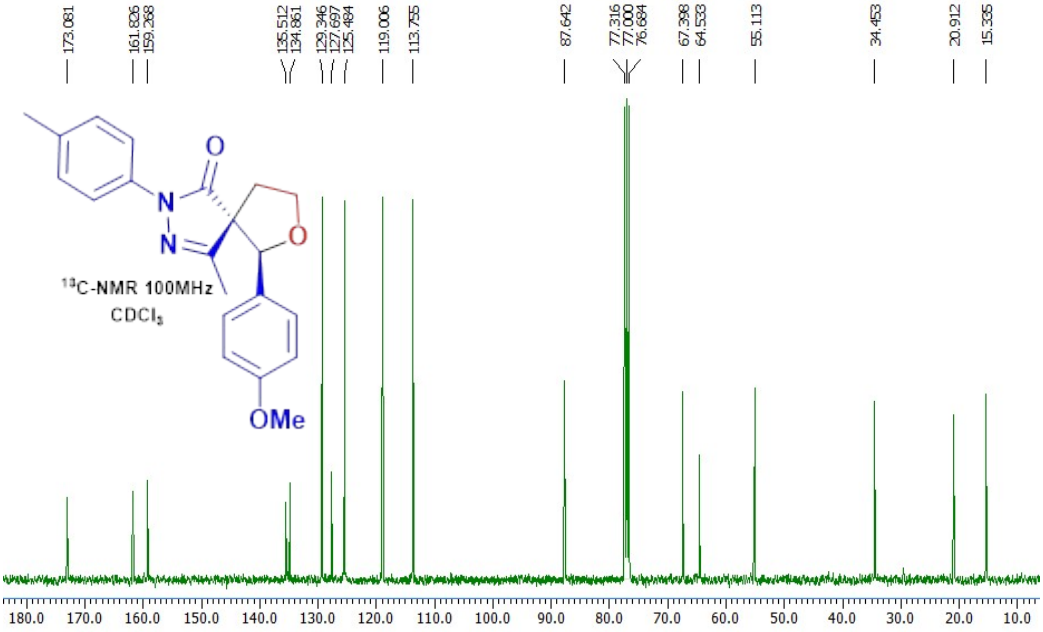



**Fig. 38: $^{13}$C-**

**NMR spectrum of (5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-(p-tolyl)-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one]non-3-en-1-one(3r)**

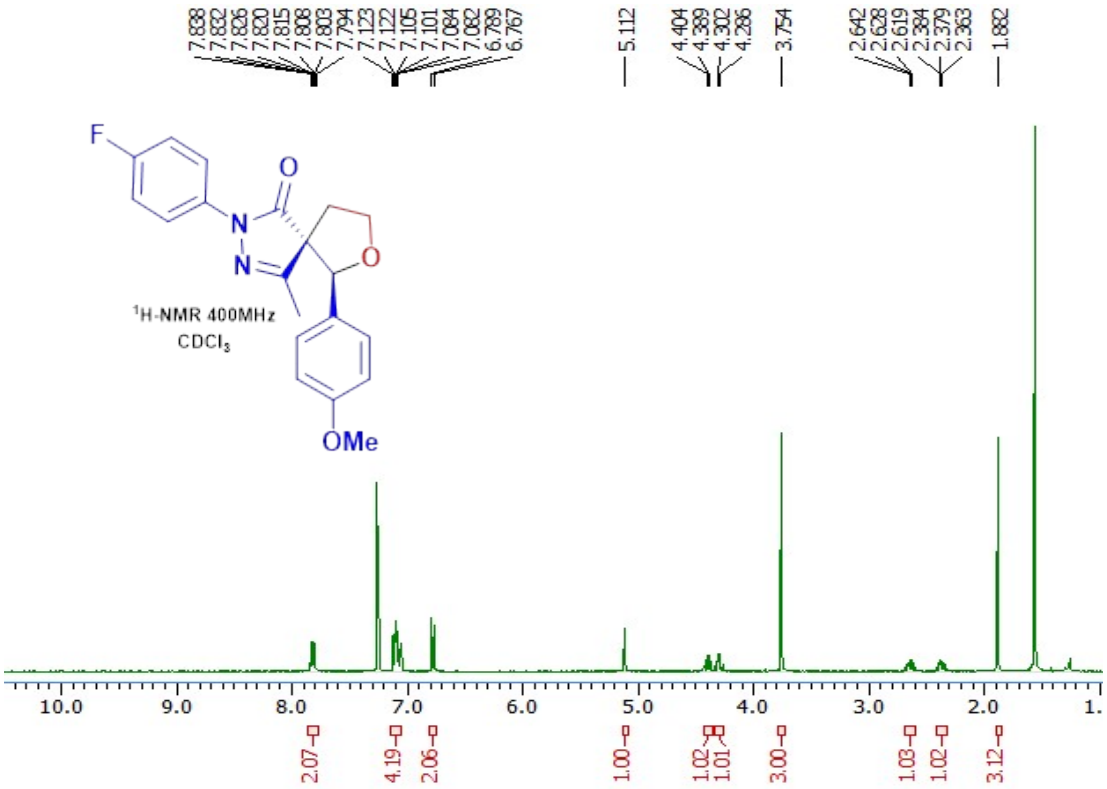



**Fig. 39: $^{1}$H-**

**NMR spectrum of (5S,6S)-2-(4-fluorophenyl)-6-(4-methoxyphenyl)-4-methyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one(3s)**

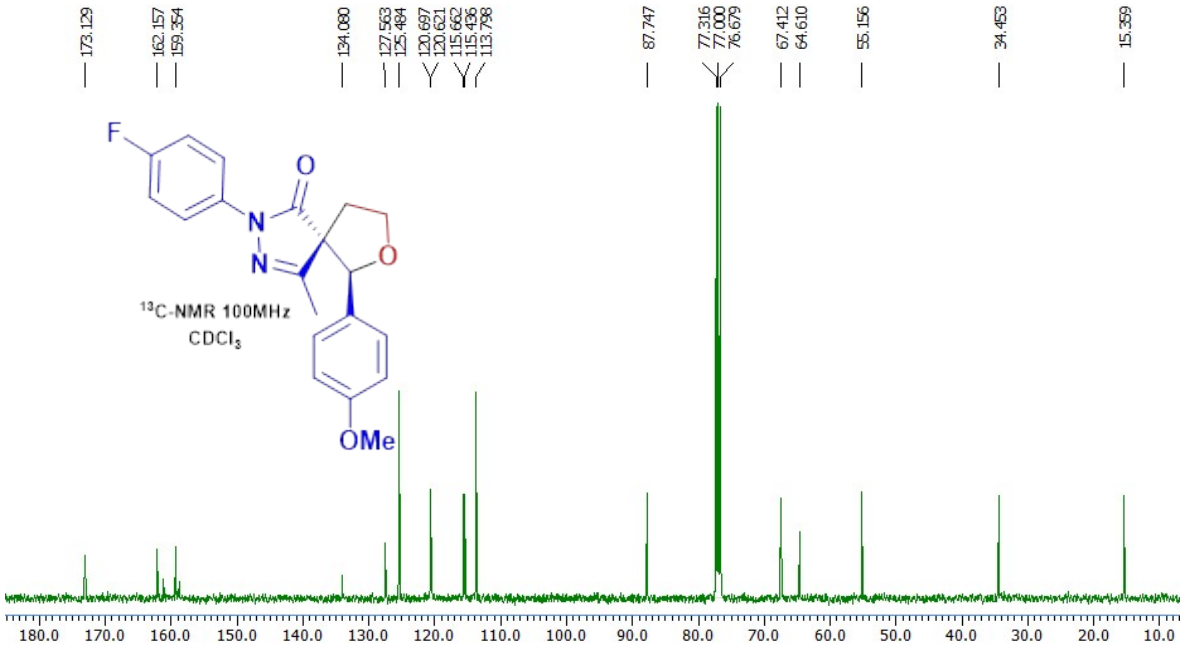


**Fig. 40: $^{13}$C-NMR spectrum of (5*S*,6*S*)-2-(4-fluorophenyl)-6-(4-methoxyphenyl)-4-methyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3s)**

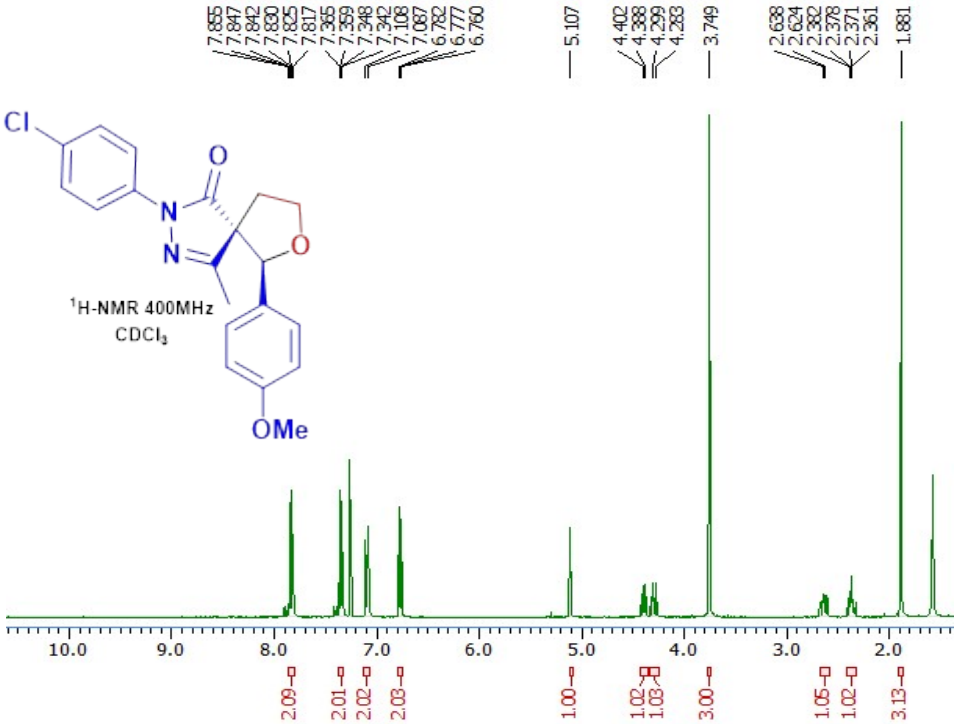
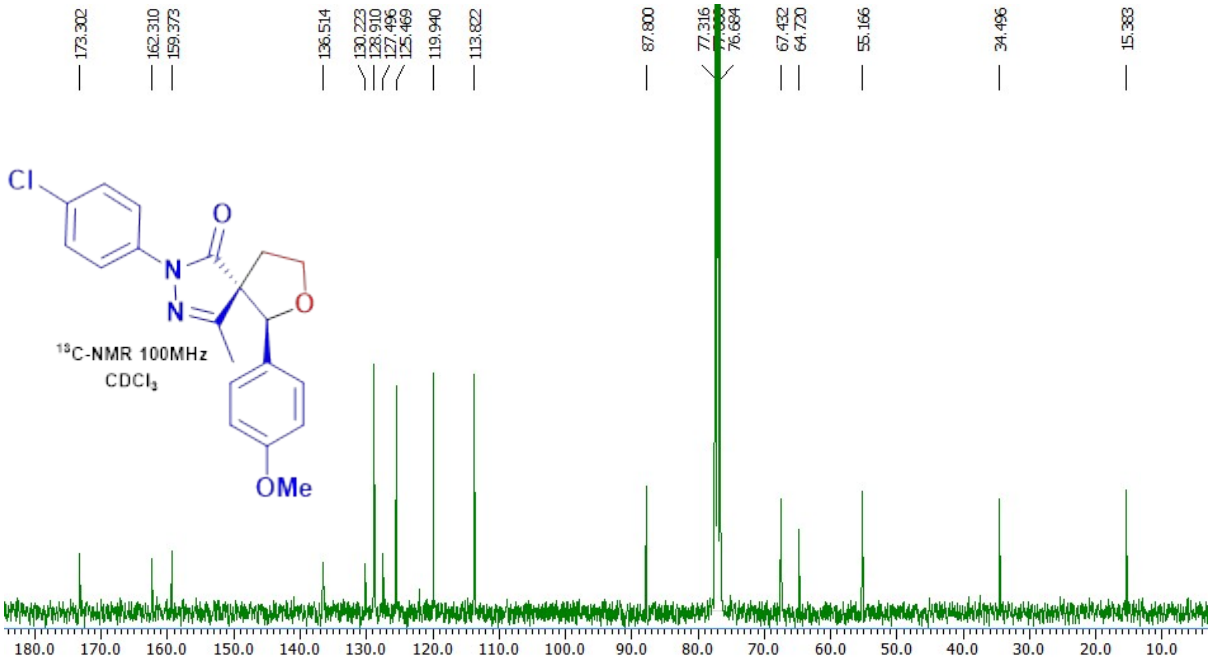



**Fig. 41:**



**$^{1}$H-NMR spectrum of (5*S*,6*S*)-2-(4-chlorophenyl)-6-(4-methoxyphenyl)-4-methyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one(3t)**

**Fig. 42: $^{13}$C-NMR spectrum of (5*S*,6*S*)-2-(4-chlorophenyl)-6-(4-methoxyphenyl)-4-methyl-7-oxa-2,3-diazaspiro[4.4]non-3-en-1-one (3t)**

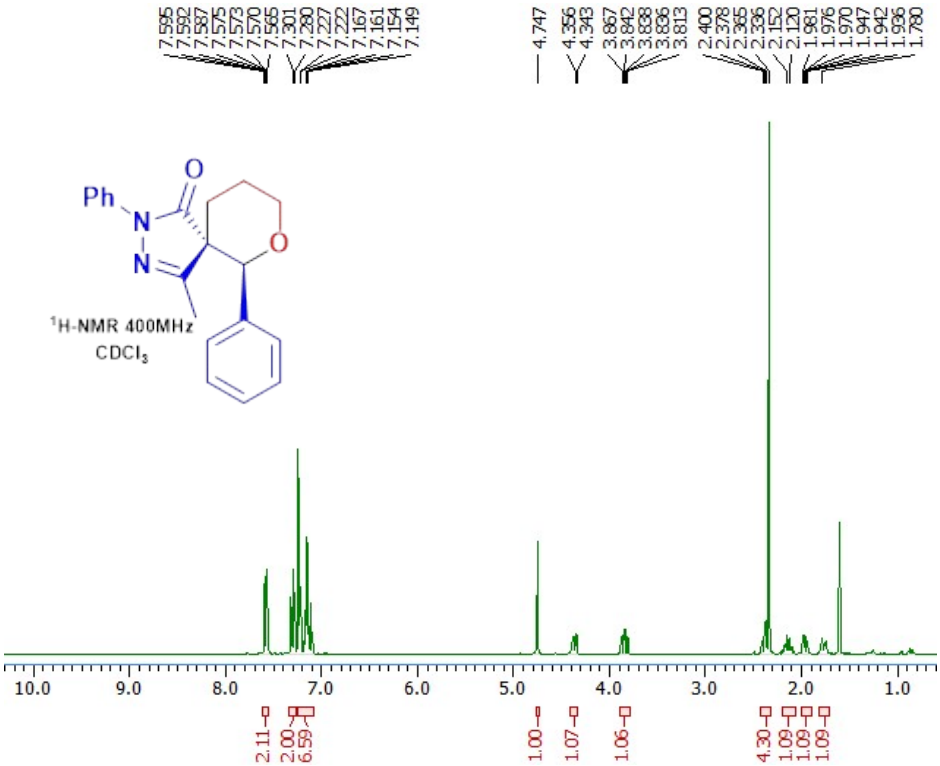
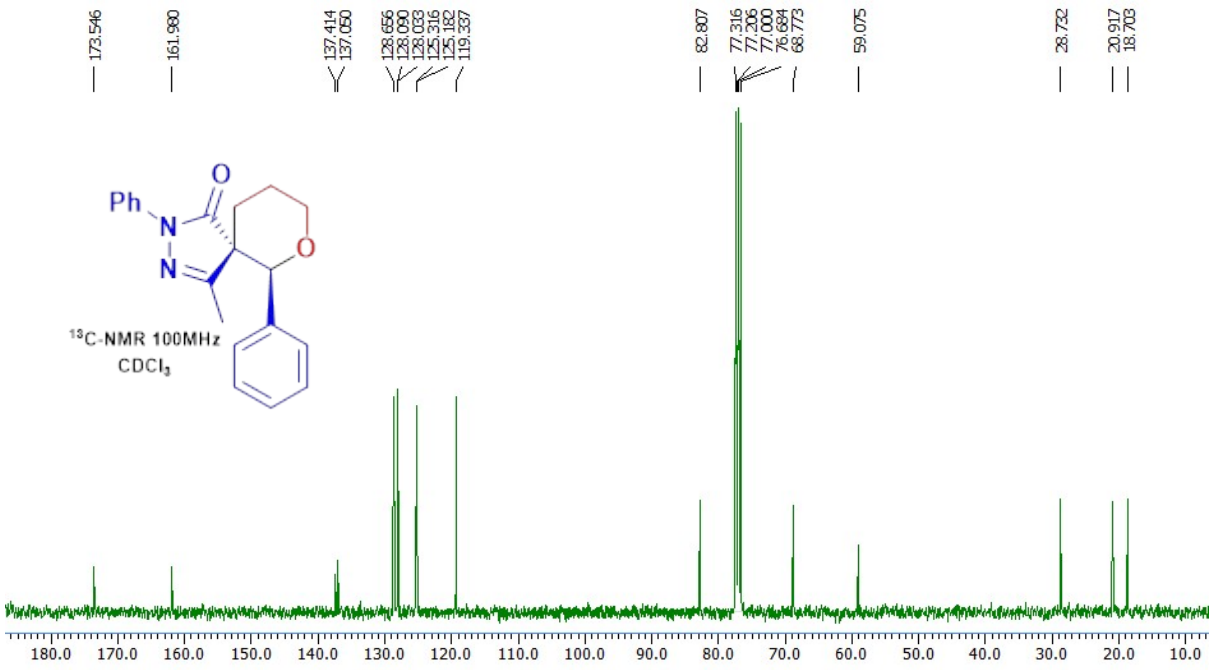



**Fig. 43:**



**$^{1}$H-NMR spectrum of (5*S*,6*S*)-4-methyl-2,6-diphenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5a)**

**Fig. 44: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-2,6-diphenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5a)**

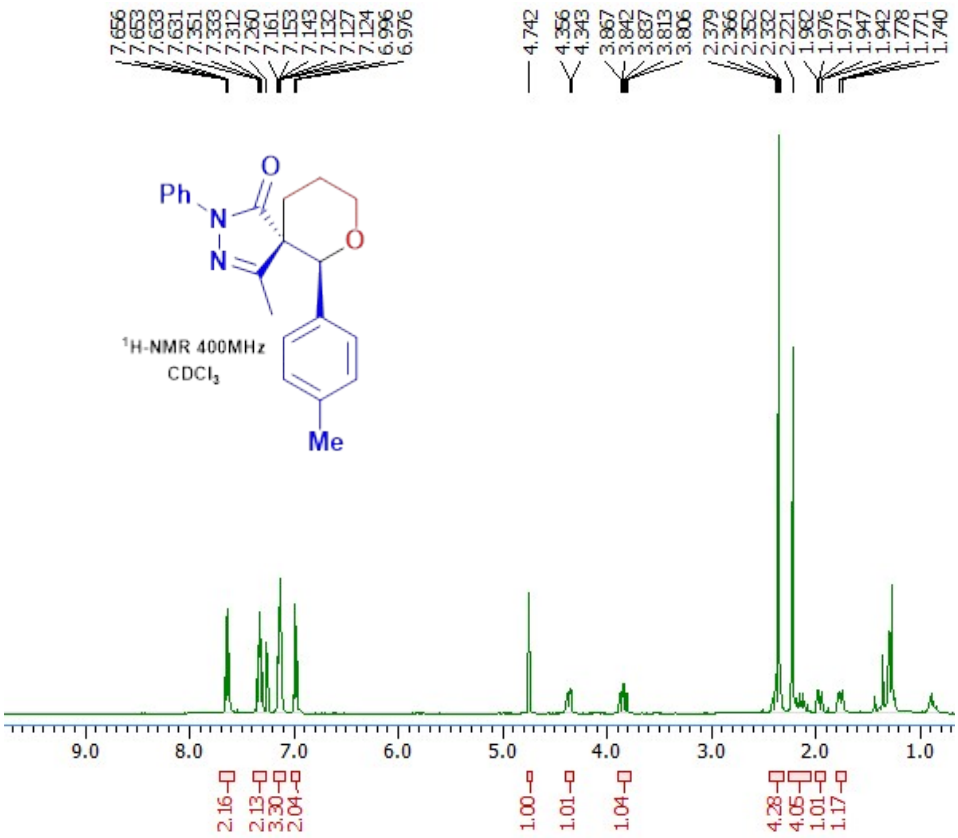
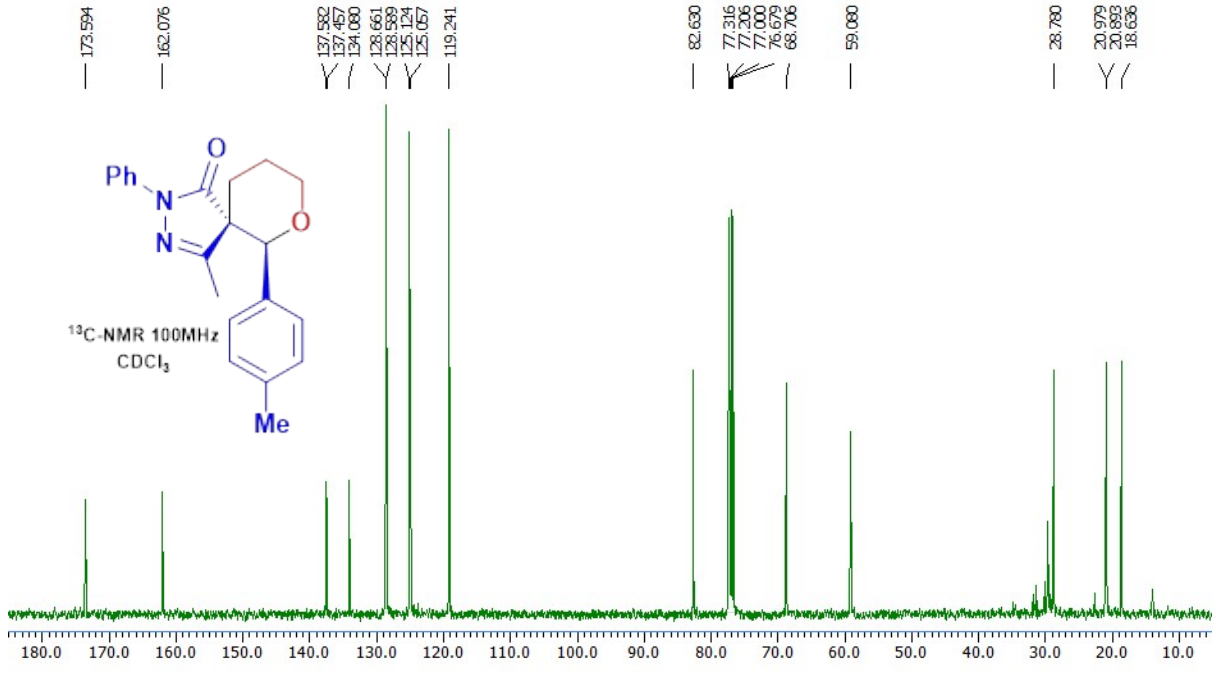



**Fig. 45:**



**$^{1}$H-NMR spectrum of (5*S*,6*S*)-4-methyl-2-phenyl-6-(p-tolyl)-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5b)**

**Fig. 46: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-2-phenyl-6-(p-tolyl)-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5b)**

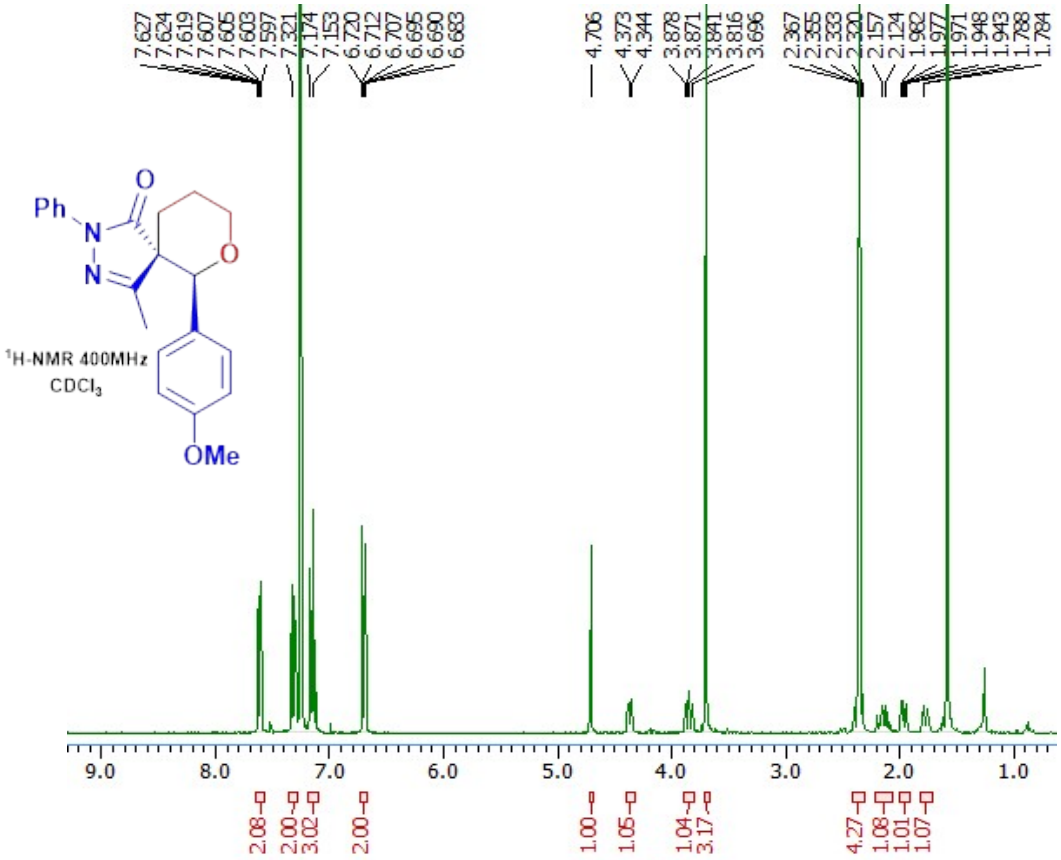


**Fig. 47: $^{1}$H-NMR spectrum of (5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5c)**

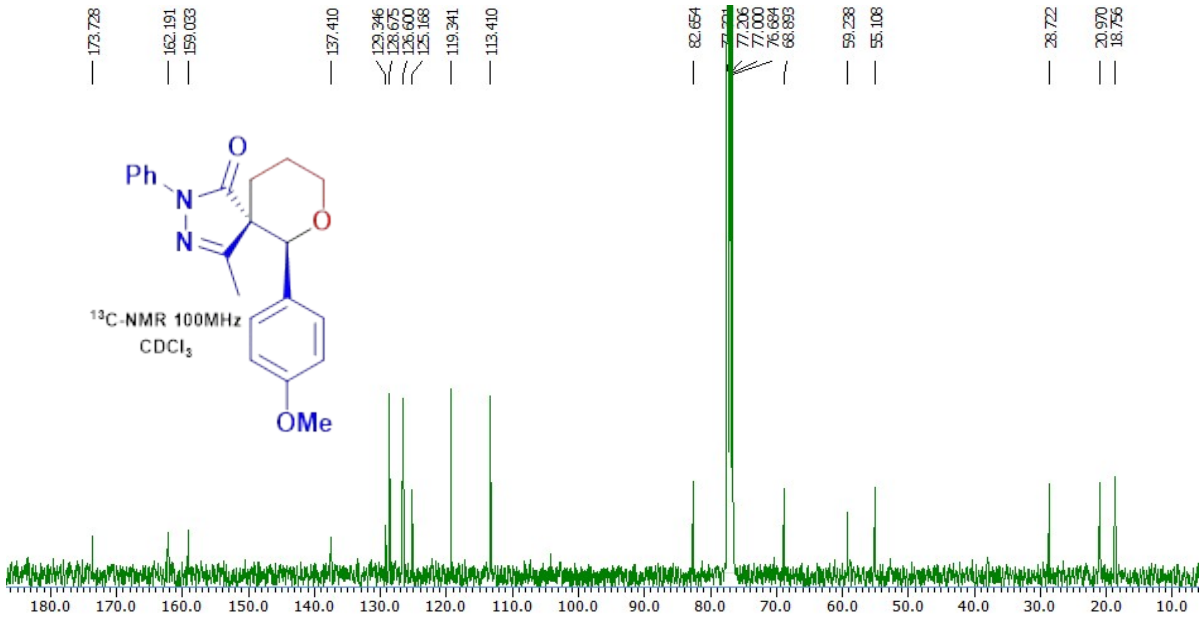


**Fig. 48: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-methoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5c)**

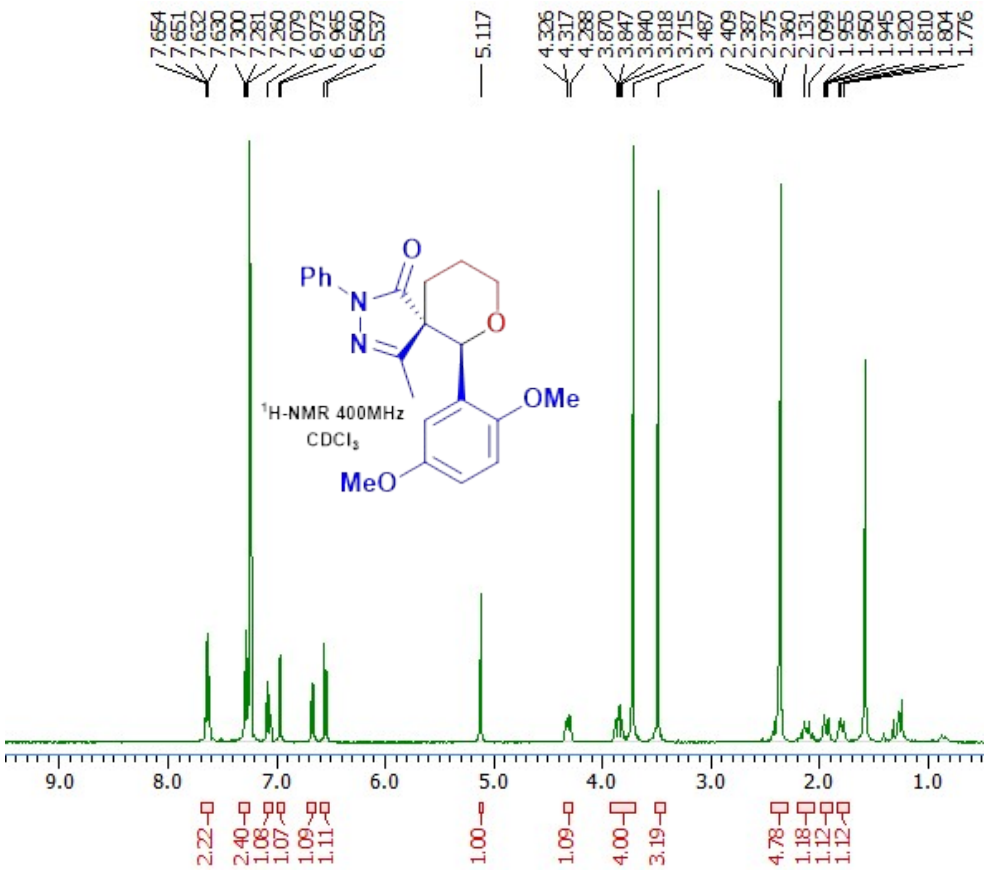



**Fig. 49: $^1$H-NMR**

**spectrum of (5*S*,6*S*)-6-(2,5-dimethoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one(5d)**

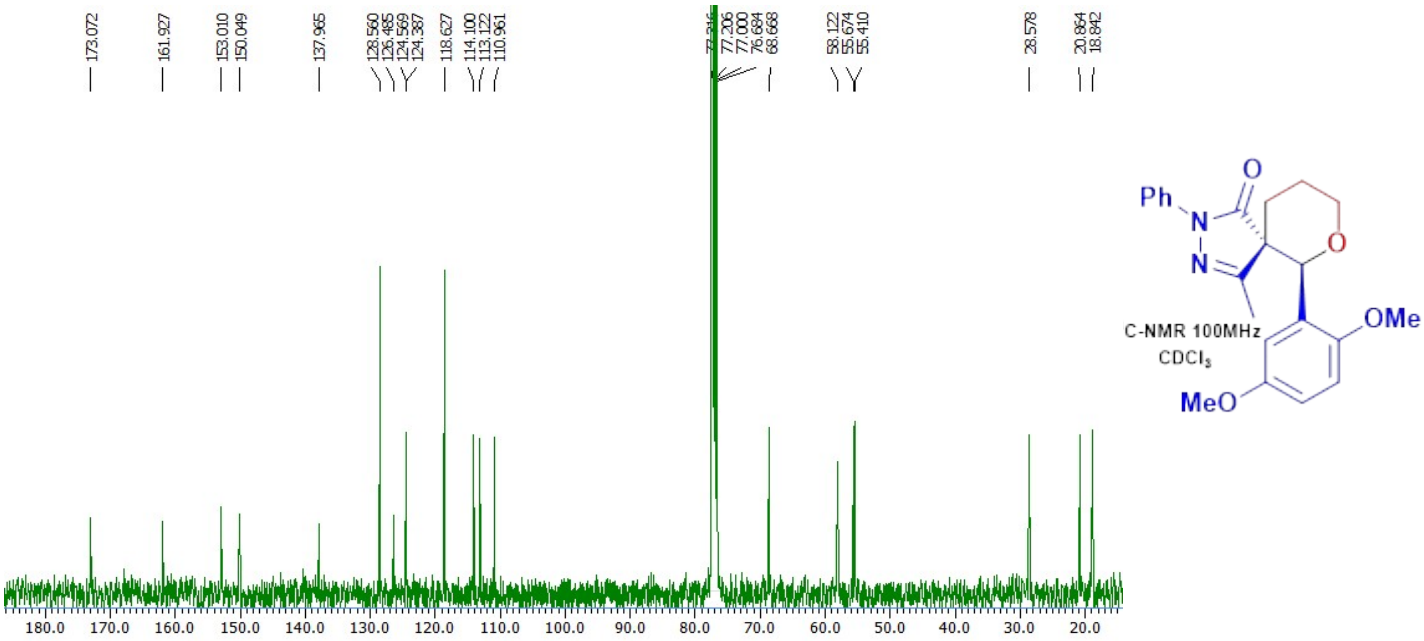



**Fig. 50: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(2,5-dimethoxyphenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one(5d)**

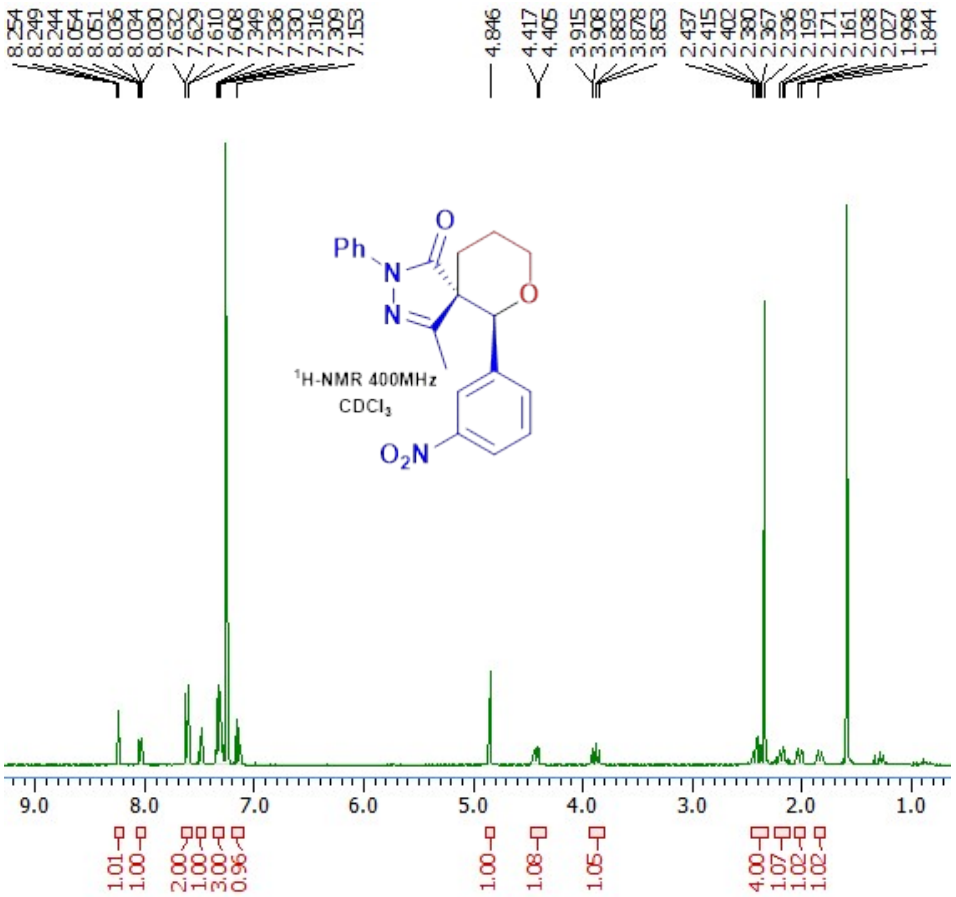

8.254
8.249
8.244
8.054
8.051
8.036
8.034
8.030
7.632
7.629
7.610
7.608
7.349
7.336
7.330
7.316
7.309
7.153
4.846
4.417
4.405
3.915
3.908
3.883
3.878
3.853
2.437
2.415
2.402
2.380
2.367
2.336
2.193
2.171
2.161
2.088
2.027
1.998
1.844
Ph
N
N
O
O
$^1$H-NMR 400MHz
$CDCl_3$
$O_2N$
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
1.01
1.00
2.00
1.88
3.00
0.96
1.00
1.08
1.05
4.00
1.07
1.02
1.02

**Fig. 51: $^1$H-**

**NMR spectrum (5*S*,6*S*)-4-methyl-6-(3-nitrophenyl)-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5e)**

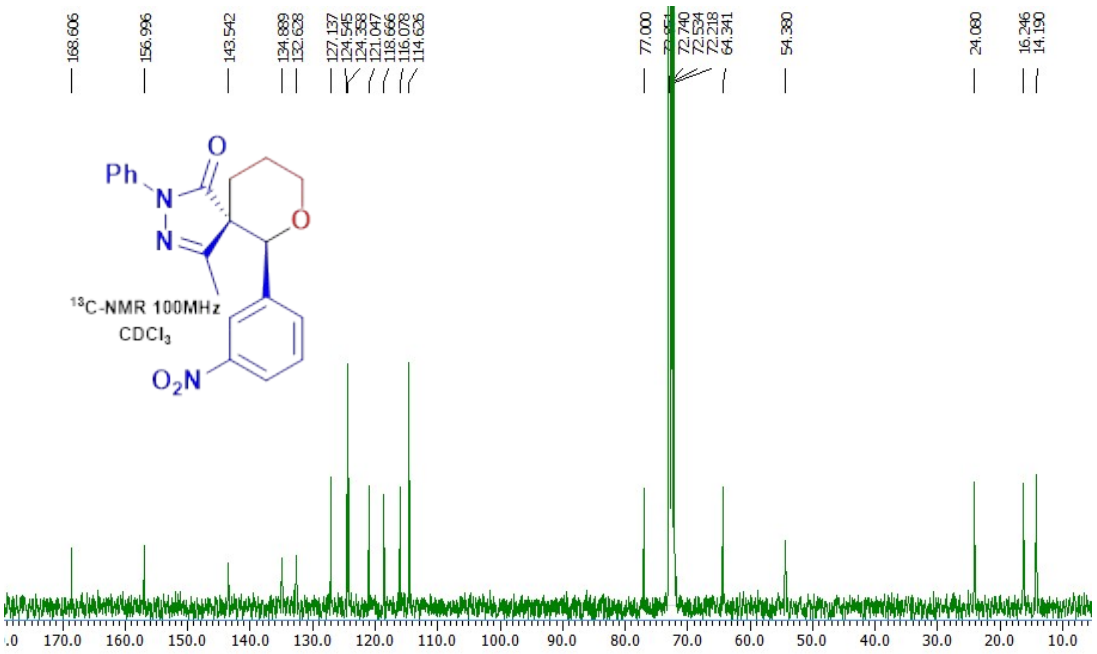

168.606
156.996
143.542
134.889
132.628
127.137
124.545
124.338
121.047
118.666
116.078
114.626
77.000
72.740
72.534
72.218
64.341
54.380
24.080
16.246
14.190
Ph
N
N
O
O
$^{13}$C-NMR 100MHz
$CDCl_3$
$O_2N$
170.0
160.0
150.0
140.0
130.0
120.0
110.0
100.0
90.0
80.0
70.0
60.0
50.0
40.0
30.0
20.0
10.0

**Fig. 52: $^{13}$C-NMR spectrum of (5*S*,6*S*)-4-methyl-6-(3-nitrophenyl)-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5e)**

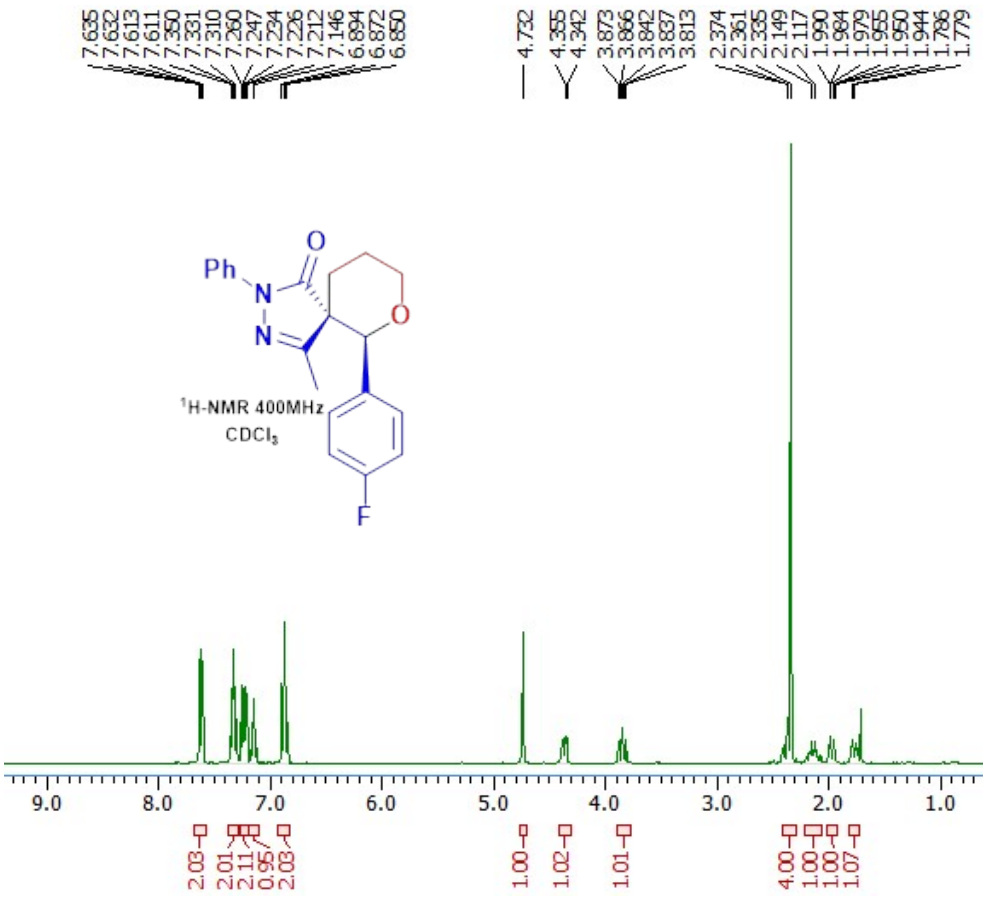


**Fig. 53: $^{1}$H-NMR spectrum of (5*S*,6*S*)-6-(4-fluorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5f)**

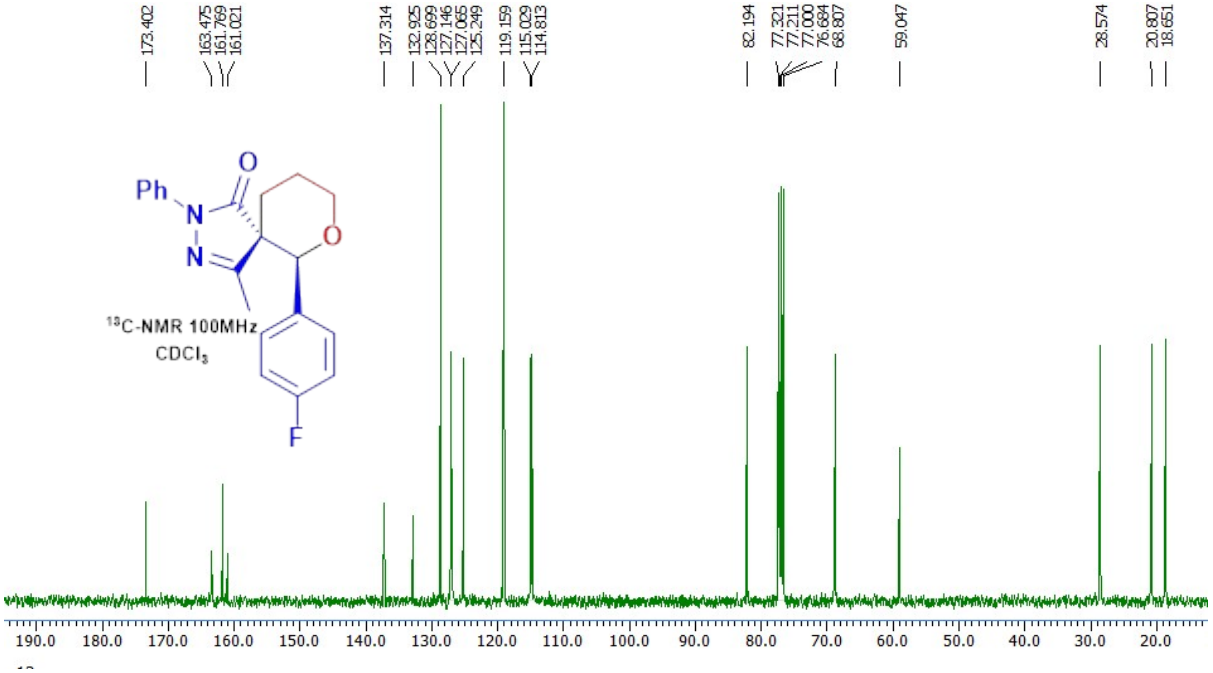


**Fig. 54: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-fluorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5f)**

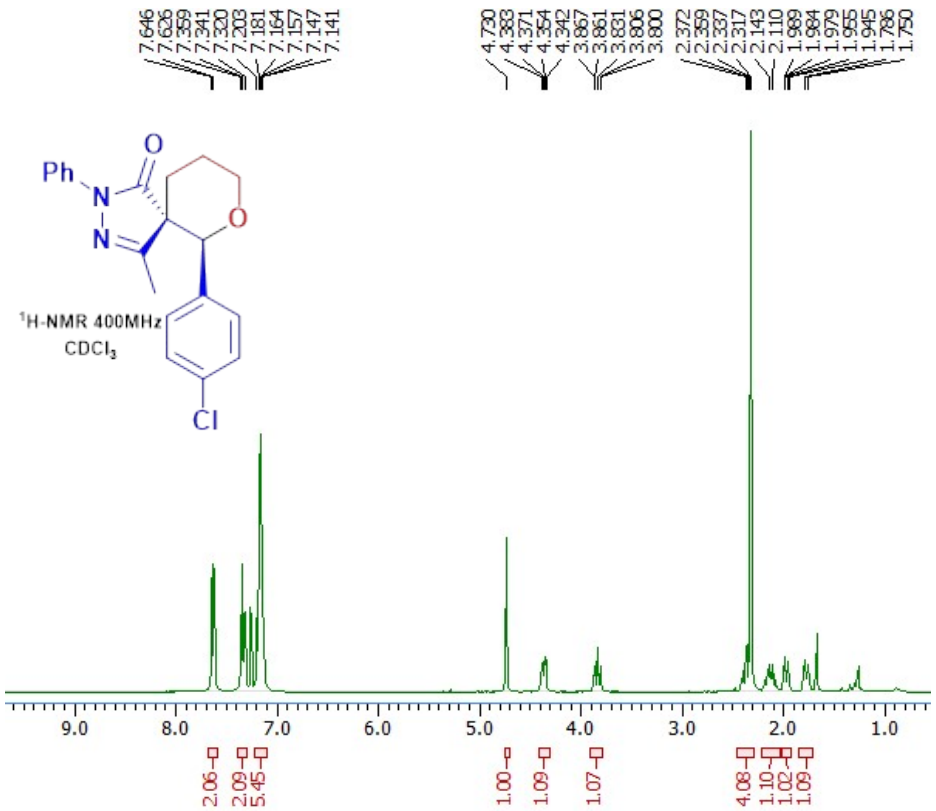
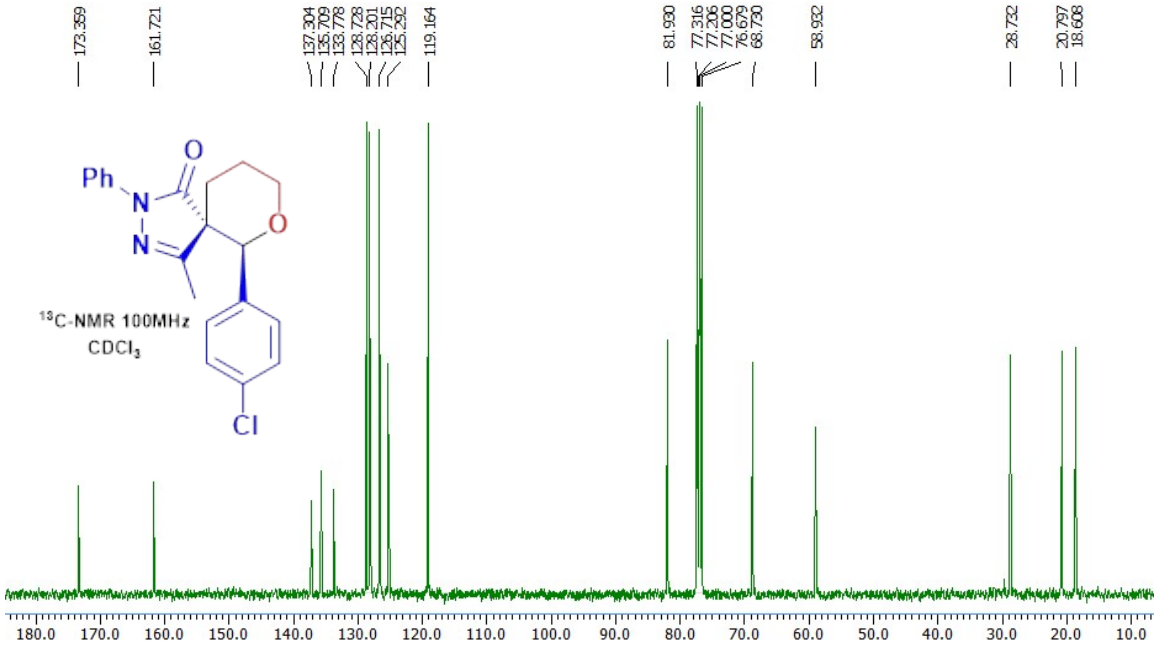



**Fig. 55: $^{1}$H-NMR**



**spectrum of (5*S*,6*S*)-6-(4-chlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5g)**

**Fig. 56: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-chlorophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5g)**

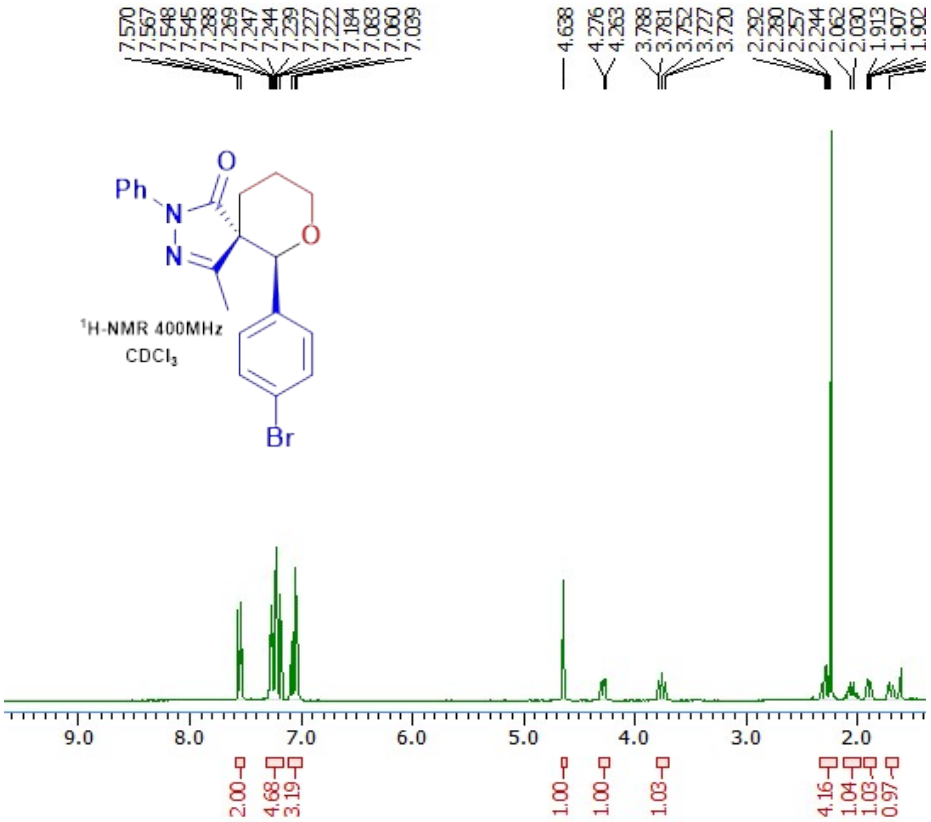
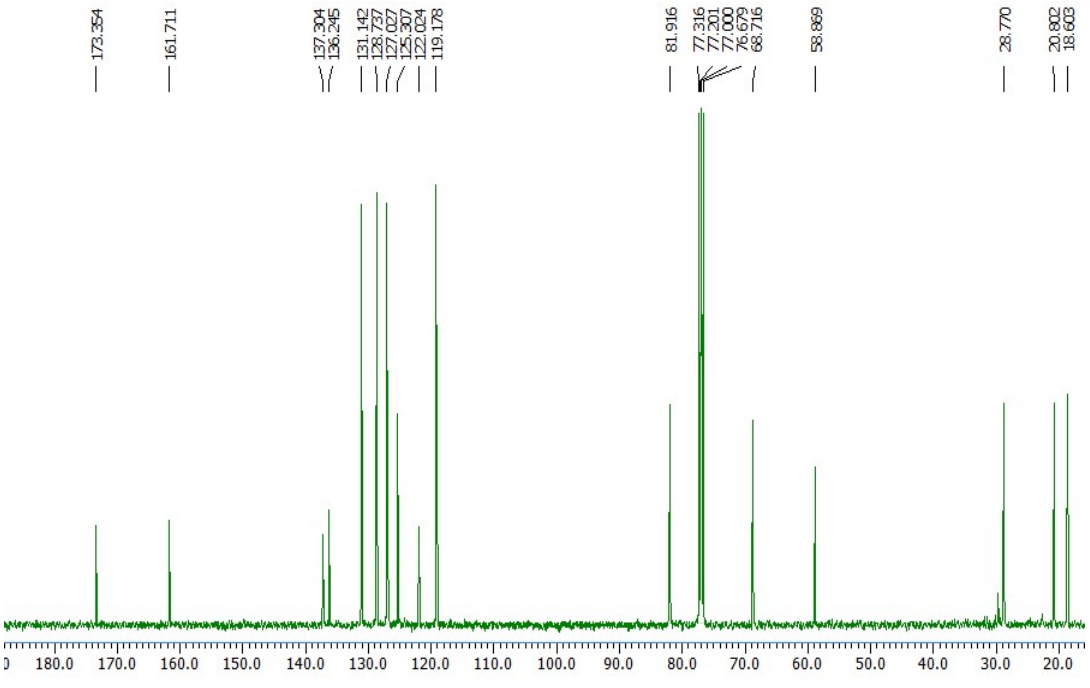



**Fig. 57: $^1$H-**



**NMR spectrum of (5*S*,6*S*)-6-(4-bromophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5h)**

**Fig. 58: $^{13}$C-NMR spectrum of (5*S*,6*S*)-6-(4-bromophenyl)-4-methyl-2-phenyl-7-oxa-2,3-diazaspiro[4.5]dec-3-en-1-one (5h)**