***Supporting Information for:***

# Photocatalyzed Cascade Meerwein Addition/Cyclization of *N*-benzylacryamides toward Azaspirocycles

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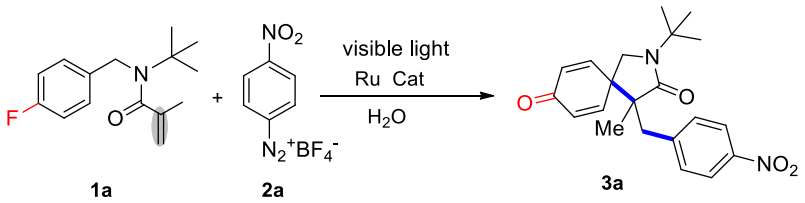
## 1、Experimental Section

**General**. All manipulations of oxygen- and moisture-sensitive materials were conducted with a Schlenk technique under a nitrogen or argon atmosphere. Photoirradiation was

carried out with a 5W blue LED (light-emitting diode). Solvent were purified and dried in a standard manner. Flash column chromatography was performed using EM Silica gel 60 (300-400 mesh). Visualization was accomplished with UV light (254 nm) and/or an aqueous alkaline $KMnO_4$ solution followed by heating. $^1H$ NMR and $^{13}C$ NMR spectra were recorded on 400 MHz NMR spectrometer with trimethylsilaneresonance as the internal standard. Unless otherwise noted, reagents were commercially available and were used without further purification. Preparation of *N*-benzyl)acrylamide (**1)** were prepared according to literature procedures.[1]

**General Procedure for the Synthesis of *N*-benzylacrylamide (1) according to known literature procedures.[1] Step 1**: To a solution of benzaldehyde (5 mmol) in ethanol (10 mL) was added alkyl amine (6 mmol), and then the resulting solution was stirred for 4 h at room temperature. Next, the mixture was added $NaBH_4$ (7.5 mmol) at 0 $^oC$, and then warmed to room temperature and continue to be stirred overnight. After related work-up and purification by flash chromatography, the *N*-alkylbenzylamine was thereby obtained (commonly about 80% yield), which is used for next synthetic step; **Step 2**: To the solution of *N*-alkylbenzylamine obtained above in dry $CH_2Cl_2$ (6 mL) was added methacryloyl chloride (1.5 equiv.) and then $Et_3N$ at 0 $^oC$. Then the resulting mixture was warmed to rt and continue to stir for 12 h. After related work-up, the residue was purified by flash chromatography (petroleum ether/ethyl acetate as the eluent) on silica gel to afford the corresponding *N*-benzylacrylamide **(1)**.

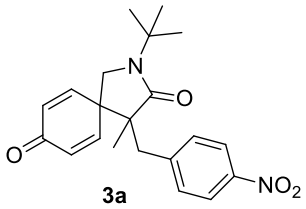
**Table S1** Screening on the Water Effect in the Addition/Cyclization Cascade




| entry | Ru catalyst | $H_2O$ (equiv.) | yield of **3a** (%)[b] |
|---|---|---|---|
| 1 | $Ru(bpy)_3Cl_2$ | — | trace |
| 2 | $Ru(bpy)_3Cl_2$ | **2** | 43 |
| 3 | $Ru(bpy)_3Cl_2$ | **4** | 75 |
| 4 | $Ru(bpy)_3Cl_2$ | **6** | 69 |
| 5 | $Ru(bpy)_3Cl_2$ | **8** | 40 |

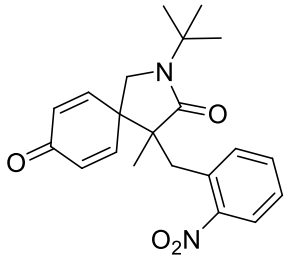
[a] Reaction conditions: **1a** (0.3 mmol), **2a** (2 equiv.), photocatalyst (5 mol%), base (3 equiv.), $H_2O$ (0~8 equiv.) and solvent (2 mL) were irradiated with a 5 W blue LEDs at room temperaturefor 24 h. [b] Yield of the isolated product.

**General Procedure for the Synthesis of 2-azaspiro[4.5]deca-6,9-diene-3,8-diones.** To a mixture of *N*-benzylacrylamide **1** (0.3 mmol), $Ru(bpy)_3Cl_2$ (0.0015mmol, 5 mol%), $H_2O$ ( 1.2 mmol, 4 equiv.), $K_3CO_3$ (0.6 mmol) in DMF (2.0 mL) was added aryldiazonium tetrafluoroborate (0.6 mmol) under $N_2$ atmosphere, and then the resulting solution was stirred under 5 W blue LED irradiation at room temperature for 24 h. Then the resulted mixture was diluted with $Et_2O$, and washed with water and then brine. The organic layer was dried over anhydrous $MgSO_4$ and concentrated in *vacuo*. The residue was purified by flash chromatography (petroleum ether/ethyl acetate 3:1 as the eluent) on silica gel to afford the corresponding 2-azaspiro[4.5]deca-6,9-diene-3,8-diones (products **3a-z**) in a yield listed in Scheme 1and Scheme 2.



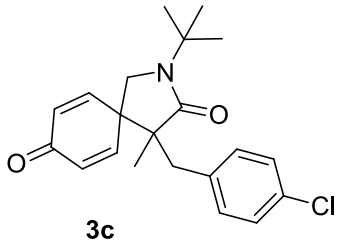

***2-(Tert-butyl)-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3a)*** **.**Yellowish oil; $^1H$ NMR (400 MHz, $CDCl_3$) δ: 8.10 (d, *J* = 1.8 Hz, 2H), 7.33 (d, *J* = 6.6 Hz, 2H),

7.13 (dd, *J* = 10.3, 3.1Hz, 1H), 6.80 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.53 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.38 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.53 (d, *J* = 10.0 Hz, 1H), 3.36 (d, *J* = 10.5 Hz, 1H), 3.14 (d, *J* = 13.7 Hz, 1H), 2.66 (d, *J* = 13.7 Hz, 1H), 1.47 (s, 9H), 1.08 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ:181.8, 175.7, 148.9, 147.6, 146.9, 144.4, 131.8, 131.0, 123.0, 54.7, 53.3, 50.0, 48.2, 39.4, 29.3, 27.6, 17.9; HRMS *m/z* (ESI-TOF) calcd for $C_{21}H_{25}N_2O_4$ $[M+H]^+$ 369.1809, found: 369.1806.



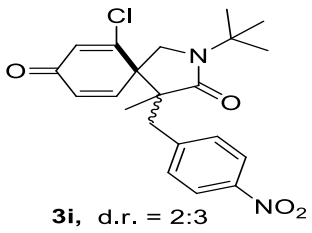
**3b**

***2-(Tert-butyl)-4-methyl-4-(2-nitrophenyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3b).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.93 (dd, *J* = 8.1, 1.1 Hz, 1H), 7.52 (dd, *J* = 3.7, 1.5 Hz, 2H), 7.38 (d, *J* = 1.5 Hz, 1H), 7.11 (d, *J* = 2.8 Hz, 1H), 6.89 (d, *J* = 3.3 Hz, 1H), 6.48 (t, *J* = 8.9 Hz, 2H), 3.76 (d, *J* = 13.7 Hz, 1H), 3.50 (d, *J* = 7.1 Hz, 2H), 2.77 (d, *J* = 13.7 Hz, 1H), 1.43 (s, 9H), 1.01 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.0, 175.6, 150.3, 148.5, 147.8, 134.6, 132.2, 131.5, 131.2, 127.9, 124.9, 54.7, 53.5, 49.8, 35.0, 27.6, 17.7; HRMS *m/z* (ESI-TOF) calcd for $C_{21}H_{25}N_2O_4$ $[M+H]^+$ 369.1809, found: 369.1806.



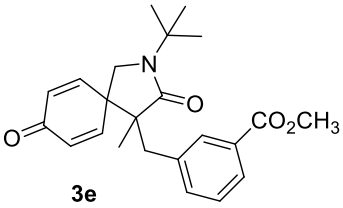
**3c**

***2-(Tert-butyl)-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3c).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.19 – 7.13 (m, 3H), 7.07 – 7.02 (m, 2H), 6.77 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.50 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.32 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.46 (d, *J* = 10.4 Hz, 1H), 3.29 (d, *J* = 10.4 Hz, 1H), 3.01 (dd, *J* = 14.3, 7.9 Hz, 1H), 2.61 (d, *J* = 14.0 Hz, 1H), 1.45 (s, 9H), 1.06 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.1, 176.0, 148.3, 148.2, 132.0, 134.8, 132.7, 132.2, 130.8, 54.6, 53.5, 50.1, 48.2 , 39.1, 37.6, 27.6, 18.0; HRMS *m/z* (ESI-TOF) calcd for $C_{21}H_{25}ClNO_2$ $[M+H]^+$ 358.1569, found: 358.1571.



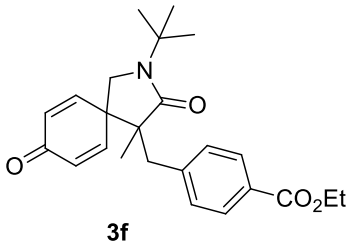
**3i**, d.r. = 2:3

***2-(Tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5] deca-6,9-diene-3,8-dione(3d)***. Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.27 (d, *J* = 8.2 Hz, 1H), 7.21 (d, *J* = 2.0 Hz, 1H), 7.12 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.97 (dd, *J* = 8.2, 2.0 Hz, 1H), 6.78 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.51 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.35 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.49 (dd, *J* = 8.7, 5.5 Hz, 1H), 3.30 (dd, *J* = 13.9, 6.4 Hz, 1H), 2.98 (d, *J* = 14.0 Hz, 1H), 2.55 (d, *J* = 6.7 Hz, 1H), 1.45 (s, 9H), 1.06 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ:184.9, 175.7, 149.0, 148.0, 136.7, 132.7, 131.4, 131.0, 130.3, 129.7, 54.7, 53.4, 50.0, 48.1, 38.8, 27.6, 17.9; HRMS *m/z* (ESI) calcd for $C_{21}H_{24}Cl_2NO_2$ $[M+H]^+$ 392.1179, found: 392.1182.



**3e**

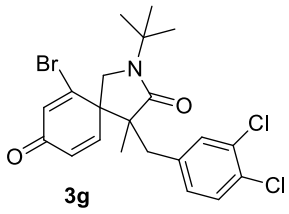
***Methyl 3-((2-(tert-butyl)-4-methyl-3,8-dioxo-2-azaspiro[4.5]deca-6,9-dien-4-yl)methyl)benzoate(3e).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.89 (dt, *J* = 7.5, 1.4 Hz, 1H), 7.33 – 7.29 (m, 2H), 7.19 (dd, *J* = 10.0, 2.7 Hz, 1H), 6.74 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.54 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.29 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.90 (s, 4H), 3.46 (d, J = 10.4 Hz, 1H), 3.46 (d, *J* = 10.4 Hz, 1H), 3.11 (d, *J* = 6.0 Hz, 1H), 2.74 (d, *J* = 8.1 Hz, 1H), 1.46 (s, 9H), 1.07 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 184.7, 175.6, 166.2, 149.6, 148.3, 137.1, 136.2, 131.1, 131.2, 129.8, 128.3, 54.4, 53.2, 51.9, 50.2, 48.3, 39.5, 27.4, 17.2; HRMS *m/z* (ESI) calcd for $C_{23}H_{28}NO_4$ $[M+H]^+$ 382.2013, found: 382.2010.



**3f**

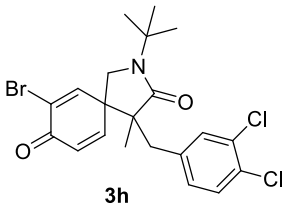
***Ethyl4-((2-(tert-butyl)-4-methyl-3,8-dioxo-2-azaspiro[4.5]deca-6,9-dien-4-yl)methyl)benzoate(3f).*** White solid; m.p. 149.3 – 149.4 °C. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.89 (d, *J* = 1.8 Hz, 2H), 7.17 (dd, *J* = 10.2, 3.1 Hz, 3H), 6.76 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.52 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.31 (dd, *J* = 10.3, 1.9 Hz, 1H), 4.36 (dd, *J* = 7.1, 0.6 Hz, 2H), 3.46 (d, *J* = 10.4 Hz, 1H), 3.30 (d, *J* = 10.4 Hz, 1H), 3.09 (d, *J* = 13.8 Hz, 1H), 2.71 (d, *J*

= 13.8 Hz, 1H), 1.46 (s, 9H), 1.39 (t, *J* = 7.1 Hz, 3H), 1.08 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.0, 175.9, 166.5, 149.2, 148.1, 141.8, 131.3, 130.9, 129.1, 60.9, 54.6, 53.5, 50.1, 48.2, 39.8, 27.6, 18.20, 14.3; HRMS *m/z* (ESI-TOF) calcd for $C_{24}H_{30}NO_4$ $[M+H]^+$ 396.2170, found: 396.2173.



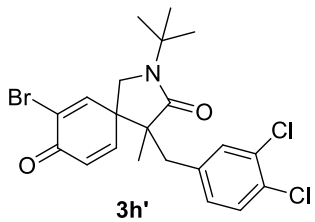
**3g**

***6-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3g).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.37 – 6.78 (m, 5H), 6.42 (ddd, J = 70.0, 10.2, 1.7 Hz, 1H), 3.78 – 3.49 (m, 2H), 3.07 (dd, J = 93.5, 13.4 Hz, 1H), 2.65 (dd, J = 64.3, 13.8 Hz, 1H), 1.51 (s, 5.4H), 1.48 (s, 3.6H), 1.15 (s, 1.2H), 1.10 (s, 1.8H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 177.1, 174.9, 148.9, 147.8, 136.5, 132.1, 128.5, 127.7, 55.3, 54.2, 52.5, 49.7, 38.9, 28.1, 19.1; HRMS *m/z* (ESI) calcd for $C_{21}H_{23}BrCl_2NO_2$ $[M+H]^+$ 470.0284, found: 470.0282.



**3h**

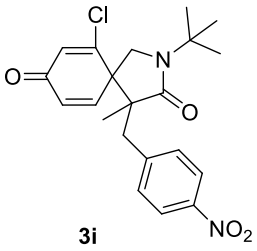
***7-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3h).*** Orange solid; m.p. 160.4 – 160.5 °C. d.r. = 3:5. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.29 (d, *J* = 3.5 Hz, 1H), 7.22 – 7.19 (m, 2H), 7.15 (dd, *J* = 10.1, 2.8 Hz, 1H), 6.95 (dd, J = 8.3, 2.1 Hz, 1H), 6.61 (d, *J* = 10.1 Hz, 1H), 3.54 (d, *J* = 10.5 Hz, 1H), 3.35 (d, J = 5.6 Hz, 1H), 2.98 (d, *J* = 14.0 Hz, 1H), 2.55 (d, J = 2.9 Hz, 1H), 1.46 (s, 9H), 1.10 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 177.8, 175.2, 149.1, 148.1, 136.3, 132.6, 132.0, 131.1, 130.1, 129.9, 129.3, 126.9, 54.9, 53.7, 51.6, 49.5, 38.9, 27.6, 18.1; HRMS *m/z* (ESI) calcd for $C_{21}H_{23}BrCl_2NO_2$ $[M+H]^+$ 470.0284, found: 470.0287.



**3h'**

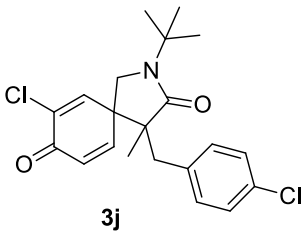
***7-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3h')*** Orange solid; m.p. 57.8 – 57.9 °C. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.50 (d, J = 2.8 Hz, 1H), 7.29 (d, J = 1.9 Hz, 1H), 7.21 (d, J = 2.1 Hz, 1H), 6.96 (dd, J = 8.3, 2.1 Hz, 1H), 6.81 (dd, J = 10.1, 2.8 Hz, 1H), 6.43 (d, J = 10.1 Hz, 1H), 3.50 (d, J = 6.7 Hz, 1H), 3.37 (d, J = 10.6 Hz, 1H), 2.97 (d, J =

8.2 Hz, 1H), 2.60 (d, J = 13.5 Hz, 1H), 1.47 (s, 9H), 1.12 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ:177.7, 175.2, 149.1, 148.0, 136.3, 131.1, 129.7, 126.7, 54.9, 53.7, 51.5, 49.5, 39.1, 27.6, 18.4; HRMS *m/z* (ESI) calcd for $C_{21}H_{23}BrCl_2NO_2$ $[M+H]^+$ 470.0284, found: 470.0287.



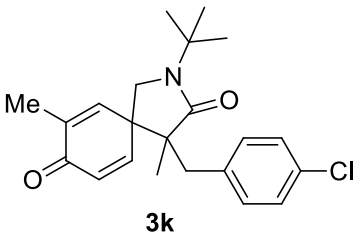
3i

***2-(Tert-butyl)-6-chloro-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5] deca-6,9-diene-3,8-dione(3i).*** Brown oil. d.r.= 2:3. $^1$H NMR (400 MHz, $CDCl_3$) δ: 8.20 – 8.03 (m, 2H), 7.43 – 7.09 (m, 3H), 6.86 – 6.30 (m, 2H), 3.81 – 3.72 (m, 1H), 3.64 (dd, J = 26.2, 11.1 Hz, 1H), 3.33 (d, J = 14.1 Hz, 0H), 3.13 (d, J = 12.9 Hz, 1H), 2.72 (dd, J = 13.5, 6.5 Hz, 1H), 1.51 (s, 5.4H), 1.41 (s, 3.6H), 1.16 (s, 1.2H), 1.08 (s, 1.8H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 178.3, 175.9, 149.2, 143.5, 134.6,.134.4, 132.8, 131.9, 130.2, 129.8,128.5, 54.7, 50.5, 49.5, 39.2, 29.6, 27.3, 18.1; HRMS *m/z* (ESI) calcd for $C_{21}H_{24}ClN_2O_4$ $[M+H]^+$ 403.1420, found: 403.1423.



3j

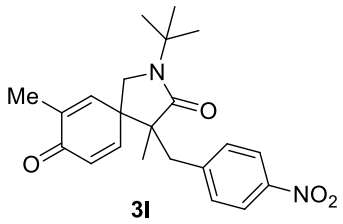
***2-(Tert-butyl)-7-chloro-4-(4-chlorobenzyl)-4-methyl-2-azaspiro [4.5]deca-6,9-diene-3,8-dione(3j).***White solid; m.p.137.2 – 137.3 °C. d.r.= 2:3. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.28 – 6.91 (m, 6H), 6.59 (d, *J* = 10.1 Hz, 1H), 3.50 (t, *J* = 10.3 Hz, 1H), 3.34 (t, *J* = 10.1 Hz, 1H), 3.00 (dd, *J* = 14.1, 2.2 Hz, 1H), 2.62 (t, *J* = 14.1 Hz, 1H), 1.46(s, 3.6H), 1.45 (s, 5.4H), 1.12 (s, 1.2H), 1.09 (s, 1.8H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 178.1, 176.5, 149.4, 143.9, 134.8, 134.5, 132.9, 132.1, 130.0, 129.7,128.1, 54.8, 50.7, 49.7, 39.1, 29.7, 27.6, 18.2; HRMS *m/z* (ESI-TOF) calcd for $C_{21}H_{24}Cl_2NO_2$ $[M+H]^+$ 392.1179, found: 392.1182.



3k

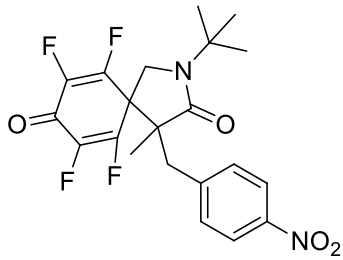
***2-(Tert-butyl)-4-(4-chlorobenzyl)-4,7-dimethyl-2-azaspiro [4.5]deca-6,9-diene-3,8-dione(3k).*** Yellowish solid; m.p. 124.4 – 124.5 °C. d.r.= 7:8. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.21 – 7.13 (m, 2H), 7.05 – 6.98 (m, 2H), 6.74 (dd, *J* = 10.1, 3.0 Hz, 1H), 6.48 (d, *J* = 10.2 Hz, 2H), 6.30 (d, *J* = 9.6 Hz, 1H), 3.44 (dd, *J* = 10.3, 2.9 Hz, 2H), 3.26 (dd, *J* = 14.5, 10.3 Hz, 2H), 2.97 (dd, *J*

= 14.0, 3.6 Hz, 2H), 2.60 (dd, *J* = 14.0, 2.1 Hz, 2H), 2.00 (d, *J* = 0.9 Hz, 3H), 1.45 (s, 3.6H), 1.44 (s, 5.4H), 1.05 (s, 1.8H), 1.04 (s, 1.2H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.3, 174.9, 148.7, 147.3, 142.3, 137.5, 131.6, 122.3, 54.8, 53.3, 50.2, 18.1, 39.0, 27.4, 18.2, 16.2; HRMS *m/z* (ESI) calcd for $C_{22}H_{28}ClNO_2$ $[M+H]^+$ 372.1725, found: 372.1728.

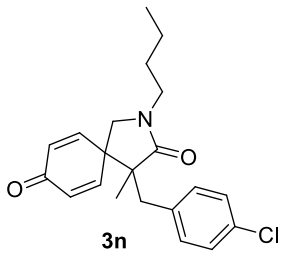


3l

***2-(Tert-butyl)-4,7-dimethyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3l).*** Brown oil. d.r.= 2:3. $^1$H NMR (400 MHz, $CDCl_3$) δ: 8.11 – 8.04 (m, 2H), 7.34 – 7.26 (m, 2H), 6.87 – 6.31 (m, 3H), 3.51 (dd, *J* = 8.3, 1.9 Hz, 1H), 3.33 (dd, *J* = 15.3, 10.4 Hz, 1H), 3.10 (dd, *J* = 13.7, 5.9 Hz, 1H), 2.64 (dd, *J* = 13.7, 3.2 Hz, 1H), 2.01 (d, *J* = 1.3 Hz, 2H), 1.86 (d, *J* = 1.3 Hz, 1H), 1.47 (s, 5.4H), 1.46 (s, 3.6H), 1.06 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.5, 175.9, 148.8, 147.5, 142.5, 137.6, 131.7, 122.9, 54.7, 53.3, 50.1, 18.0, 39.5, 27.6, 18.1, 16.4; HRMS *m/z* (ESI) calcd for $C_{22}H_{27}N_2O_4$ $[M+H]^+$ 383.1966, found: 383.1968.



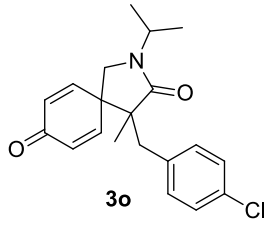

***2-(Tert-butyl)-6,7,9,10-tetrafluoro-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione（3m）.*** Orange solid; m.p. 130.0 – 130.1 °C. $^1$H NMR (400 MHz, $CDCl_3$) δ：8.16 (d, *J* = 1.9 Hz, 2H), 7.38 (d, *J* = 8.7 Hz, 2H), 3.98 (s, 2H), 3.30 (dd, *J* = 13.7, 3.0 Hz, 1H), 2.69 (dd, *J* = 13.7, 3.1 Hz, 1H), 1.51 (s, 9H), 1.27 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 172.8, 147.2, 142.8, 141.8-136.1 (m, 2C), 132.5-129.6 (m, 2C), 131.9, 125.5, 123.2, 55.5, 53.6, 45.5, 40.0, 39.9，29.7, 27.4, 10.2; HRMS *m/z* (ESI) calcd for $C_{21}H_{21}F_4N_2O_4$ $[M+H]^+$ 441.1432, found: 441.1435.11111



3n

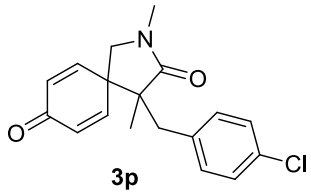
***2-Butyl-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3n).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.19 (d, *J* = 1.4 Hz, 2H), 7.14 (dd, *J* = 10.3, 3.0 Hz, 1H), 7.08 (d, *J* = 8.4 Hz, 2H), 6.80 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.51 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.34 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.49 – 3.37 (m, 2H), 3.35 – 3.22 (m, 1H), 3.13 (d, *J* = 10.3 Hz, 1H), 3.01 (d, *J* = 14.1 Hz, 1H), 2.66

(d, *J* = 14.1 Hz, 1H), 1.56 – 1.48 (m, 2H), 1.41 – 1.33 (m, 2H), 1.10 (s, 3H), 0.98 (t, *J* = 7.3 Hz, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.0, 175.8, 148.8, 148.1, 134.7, 132.8, 132.1, 131.1, 128.1, 53.1, 51.8, 49.0, 42.7, 29.3, 20.2, 18.5, 13.8; HRMS *m/z* (ESI-TOF) calcd for $C_{21}H_{25}ClNO_2$ $[M+H]^+$ 358.1569, found: 358.1571.



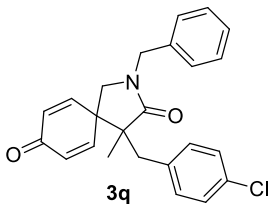
3o

***4-(4-Chlorobenzyl)-2-isopropyl-4-methyl-2-azaspiro[4.5]deca-6, 9-diene-3,8-dione(3o).*** Brown oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.19 (d, *J* = 8.4 Hz, 2H), 7.11 (dd, *J* = 10.3, 3.0 Hz, 1H), 7.06 (d, *J* = 8.4 Hz, 2H), 6.77 (dd, *J* = 10.3, 3.0 Hz, 1H), 6.49 (dd, *J* = 10.3, 1.8 Hz, 1H), 6.32 (dd, *J* = 10.3, 1.8 Hz, 1H), 4.47 (dq, *J* = 13.0, 6.5 Hz, 1H), 3.32 (d, *J* = 10.3 Hz, 1H), 3.13 (d, *J* = 10.3 Hz, 1H), 3.00 (d, *J* = 2.4 Hz, 1H), 2.64 (d, *J* = 14.1 Hz, 1H), 1.16 (dd, *J* = 6.4, 5.8 Hz, 6H), 1.08 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ:185.0, 175.0, 148.7, 148.1, 134.7, 132.7, 132.1, 131.0, 128.1, 53.3, 46.9, 43.0, 39.2, 19.5, 18.2; HRMS *m/z* (ESI) calcd for $C_{20}H_{23}ClNO_2[M+H]^+$ 344.1412, found: 344.1415.



3p

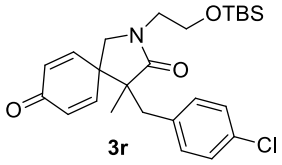
***4-(4-Chlorobenzyl)-2,4-dimethyl-2-azaspiro[4.5]deca-6,9-dien e-3,8-dione(3p)***.Yellowish solid; m.p. 149.8 – 149.9 ºC. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.20 (d, *J* = 4.8 Hz, 2H), 7.15 (dd, *J* = 10.3, 3.1 Hz, 1H), 7.08 (d, *J* = 8.4 Hz, 2H), 6.79 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.50 (dd, *J* = 7.1, 2.5 Hz, 1H), 6.33 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.33 (d, *J* = 10.3 Hz, 1H), 3.09 (d, *J* = 10.3 Hz, 1H), 2.99 (d, *J* = 14.1 Hz, 1H), 2.94 (d, *J* = 14.1 Hz, 3H), 2.67 (s, 1H), 1.09 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ:185.0, 176.0, 148.3, 134.7, 132.8, 132.0, 131.0, 128.1, 53.8, 53.1, 49.0 , 39.7, 30.1, 18.6; HRMS *m/z* (ESI) calcd for$C_{18}H_{19}ClNO_2$ $[M+H]^+$ 316.1099, found: 316.1102.



3q

***2-Benzyl-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-d iene-3,8-dione(3q).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.42 – 7.32 (m, 3H), 7.28 (d, *J* = 6.8 Hz, 2H), 7.22 – 7.17 (m, 2H), 7.09 – 7.03 (m, 3H), 6.69 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.44 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.28 (dd, *J* = 10.3, 1.9 Hz, 1H), 4.67 (d, *J* = 14.4 Hz, 1H), 4.39 (d, *J* = 6.7 Hz, 1H), 3.24 (d, *J* = 7.5 Hz, 1H), 3.02 (d, *J* = 3.9 Hz, 2H), 2.67 (d, *J* = 8.1 Hz,

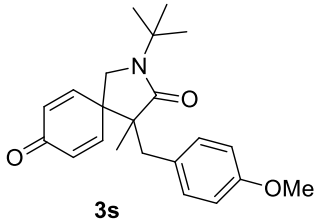
1H), 1.11 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 184.9, 175.8, 148.6, 147.8, 135.5, 134.6, 132.8, 132.1, 131.2, 129.0, 128.5, 128.2, 128.1, 53.0, 51.2, 48.8, 47.1, 39.3, 18.3; HRMS *m/z* (ESI) calcd for $C_{24}H_{23}ClNO_2$ $[M+H]^+$ 392.1412, found: 392.1414.



3r

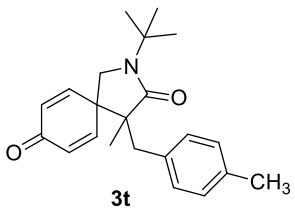
6-12-05

***2-(2-(Tert-butyldimethylsilyl)oxy)ethyl)-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3r).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ 7.16 (d, *J* = 3.0 Hz, 3H), 7.08 (d, *J* = 8.4 Hz, 2H), 6.83 (dd, *J* = 10.3, 3.0 Hz, 1H), 6.49 (dd, *J* = 10.3, 1.9 Hz, 11H), 6.33 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.87 – 3.76 (m, 3H), 3.58 – 3.53 (m, 1H), 3.35 (d, *J* = 10.4 Hz, 1H), 3.21 (d, *J* = 15.3 Hz, 1H), 3.01 (s, 1H), 2.66 (d, *J* = 14.0 Hz, 1H), 1.11 (s, 3H), 0.89 (s, 9H), 0.08 (s, 6H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.1, 176.0, 184.3, 134.7, 132.8, 132.1, 130.9, 128.1, 61.4, 53.0, 49.4, 39.3, 35.8, 18.3, -5.4; HRMS *m/z* (ESI) calcd for $C_{25}H_{35}ClNO_3Si$ $[M+H]^+$ 460.2070, found: 460.2066.



3s

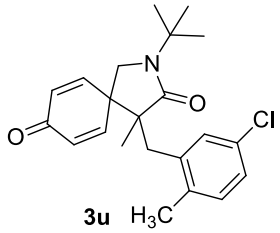
***2-(Tert-butyl)-4-(4-methoxybenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3s).***White solid; m.p. 153.5 – 153.6 $^o$C. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.20 (dd, *J* = 10.3, 3.1 Hz , 1H), 7.08 – 6.98 (dd, *J* = 6.4, 3.1 Hz, 2.0 Hz,2H), 6.82 – 6.68 (m, 3H), 6.51 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.30 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.78 (s, 3H), 3.42 (d, *J* = 10.3 Hz, 1H), 3.26 (d, *J* = 10.3 Hz, 1H), 2.97 (d, *J* = 14.2 Hz, 1H), 2.65 (d, *J* = 14.2 Hz, 1H), 1.45 (s, 9H), 1.08 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.3, 176.3, 158.4, 149.5, 148.7, 130.8, 128.2, 113.2, 55.2, 54.5, 53.9, 50.1,48.2, 39.2, 27.7, 18.1; HRMS *m/z* (ESI-TOF) calcd for $C_{22}H_{28}NO_3$ $[M+H]^+$ 354.2064, found: 354.2066.



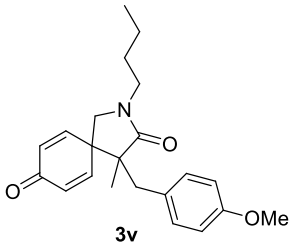
3t

***2-(Tert-butyl)-4-methyl-4-(4-methylbenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3t).***White solid; m.p. 153.3 – 153.4 $^o$C. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.21 (dd, *J* = 10.3, 3.1 Hz, 1H), 7.01 (q, *J* = 8.2 Hz, 4H), 6.76 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.51 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.29 (dd, *J* = 10.3, 1.9 Hz,

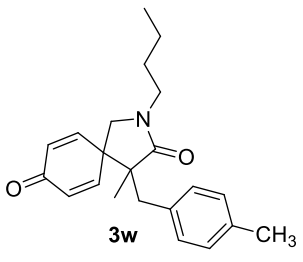
1H), 3.41 (d, *J* = 10.3 Hz, 1H), 3.27 (d, *J* = 10.3 Hz, 1H), 3.00 (d, *J* = 14.0 Hz, 1H), 2.67 (d, *J* = 14.0 Hz, 1H), 2.30 (s, 3H), 1.45 (s, 9H), 1.08 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$)δ: 185.3, 176.2, 149.5, 148.7, 136.3, 148.7, 136.3, 133.2, 130.9, 130.8, 130.7, 128.6, 54.5, 53.8, 50.1, 48.2, 39.6, 29.7, 27.7, 21.0, 18.1; HRMS *m/z* (ESI-TOF) calcd for $C_{22}H_{28}NO_2$ $[M+H]^+$ 338.2115, found: 338.2118.




***2-(Tert-butyl)-4-(5-chloro-2-methylbenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3u).*** White solid; m.p. 181.7 – 181.8 °C. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.16 – 7.01 (m, 4H), 6.87 (d, *J* = 3.3 Hz, 1H), 6.48 (dt, *J* = 4.4, 2.0 Hz, 2H), 3.51 (d, *J* = 10.4 Hz, 1H), 3.36 (d, *J* = 10.4 Hz, 1H), 3.11 (d, *J* = 14.3 Hz, 1H), 2.62 (d, *J* = 14.3 Hz, 1H), 2.23 (s, 3H), 1.47 (s, 9H), 1.00 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.0, 176.2, 149.4, 148.2, 137.2, 135.7, 130.7, 125.7, 54.5, 50.1, 48.6, 35.0, 27.6, 19.9, 17.5; HRMS *m/z* (ESI-TOF) calcd for $C_{22}H_{27}ClNO_2$ $[M+H]^+$ 372.1725, found: 372.1728.

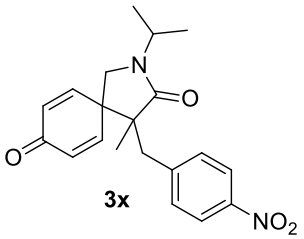



***2-Butyl-4-(4-methoxybenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3v).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.20 (dd, *J* = 10.3, 3.0 Hz, 1H), 7.05 (d, *J* = 8.6 Hz, 1H), 6.81 – 6.75 (m, 4H), 6.50 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.31 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.78 (s, 3H), 3.48 – 3.40 (m, 1H), 3.33 – 3.22 (m, 2H), 3.08 (d, *J* = 10.2 Hz, 1H), 2.97 (d, *J* = 14.2 Hz, 1H), 2.72 (d, *J* = 14.2 Hz, 1H), 1.56 – 1.47 (m, 2H), 1.42 – 1.33 (m, 2H), 1.11 (s, 3H), 0.97 (t, *J* = 7.3 Hz, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.2, 176.0, 158.4, 148.9, 131.7, 131.0, 130.6, 113.3, 55.2, 53.6, 51.8, 49.0, 42.7, 39.6, 29.3, 20.2, 16.6, 13.8; HRMS *m/z* (ESI) calcd for $C_{22}H_{28}NO_3$ $[M+H]^+$ 354.2064, found: 354.2067.



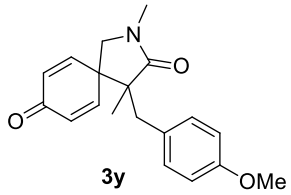

***2-Butyl-4-methyl-4-(4-methylbenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3w).*** Yellowish oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.21 (dd, *J* = 10.3, 3.0 Hz, 1H), 7.06 – 7.00 (m, 4H), 6.79 (dd, *J* = 10.3, 3.0 Hz, 1H), 6.51 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.30 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.50 – 3.40 (m, 1H), 3.33 –

3.16 (m, 2H), 3.09 (d, *J* = 10.2 Hz, 1H), 2.99 (d, *J* = 14.1 Hz, 1H), 2.74 (d, *J* = 14.1 Hz, 1H), 2.31 (s, 3H), 1.57 – 1.47 (m, 2H), 1.37 (dq, *J* = 13.8, 6.9 Hz, 2H), 1.12 (s, 3H), 0.98 (t, *J* = 7.3 Hz, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 182.0, 176.0, 148.8, 136.4, 133.0, 131.0, 130.6, 128.7, 53.5, 51.8, 49.1, 42.7, 40.1, 29.7, 21.0, 20.2, 18.7, 13.8; HRMS *m/z* (ESI-TOF) calcd for $C_{22}H_{28}NO_2$ $[M+H]^+$ 338.2115, found: 328.2113.



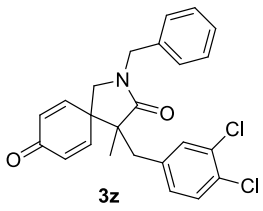
3x

***2-Isopropyl-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3x).*** Brown oil; $^1$H NMR (400 MHz, $CDCl_3$) δ: 8.08 (d, *J* = 8.7 Hz, 2H), 7.34 (d, *J* = 8.7 Hz, 2H), 7.08 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.81 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.51 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.37 (dd, *J* = 10.3, 1.9 Hz, 1H), 4.47 (dq, *J* = 13.4, 6.7 Hz, 1H), 3.41 (d, *J* = 10.4 Hz, 1H), 3.18 (dd, *J* = 29.0, 12.1 Hz, 2H), 2.67 (d, *J* = 13.7 Hz, 1H), 1.24 – 1.20 (m, 3H), 1.17 (d, *J* = 6.8 Hz, 3H), 1.09 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 184.4, 174.7, 148.4, 147.5, 146.9, 144.3, 131.8, 131.1, 123.0, 53.0, 46.7, 43.0, 39.4, 19.4, 18.1; HRMS *m/z* (ESI) calcd for $C_{20}H_{23}N_2O_4$ $[M+H]^+$ 355.1653, found: 355.1656.



3y

***4-(4-methoxybenzyl)-2,4-dimethyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3y).*** Yellowish solid; m.p. 131.8 – 131.9 °C. $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.23 (dd, *J* = 10.3, 3.0 Hz, 1H), 7.05 (dd, *J*= 11.9, 9.1 Hz, 2H), 6.87 – 6.68 (m, 3H), 6.51 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.31 (dd, *J* = 10.3, 1.9 Hz, 1H), 3.79 (s, 3H), 3.22 (d, *J* = 10.2 Hz, 1H), 3.02 (d, *J* = 10.2 Hz, 1H), 2.96 (d, *J* = 6.2 Hz, 1H), 2.93 (s, 3H), 2.76 (d, *J* = 14.3 Hz, 1H), 1.10 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 185.2, 176.2, 158.5, 149.0, 148.4, 131.6, 130.3, 128.1, 113.4, 55.2, 53.8, 49.0, 40.1, 30.1, 18.9; HRMS *m/z* (ESI) calcd for $C_{19}H_{22}NO_3$ $[M+H]^+$ 312.1595, found: 312.1598.



3z

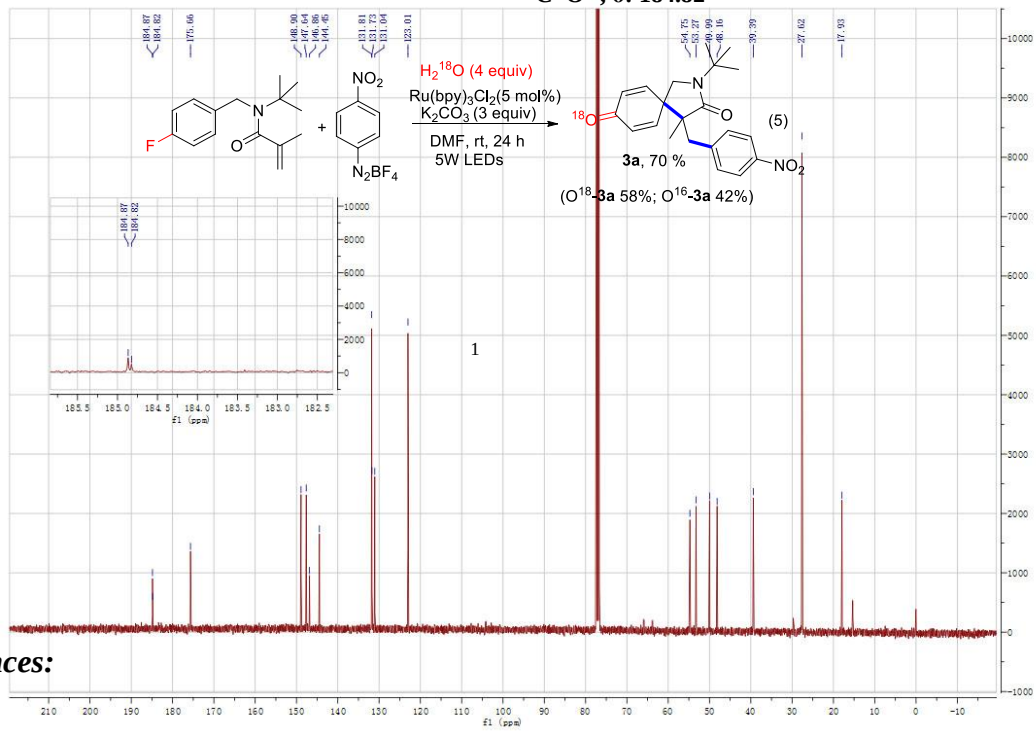
***2-Benzyl-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3z).*** $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.42 – 7.34 (m, 3H), 7.31 – 7.25 (m, 5H), 6.99 (dd, *J* = 6.5, 1.8 Hz, 1H),

6.72 (dd, *J* = 10.3, 3.1 Hz, 1H), 6.45 (dd, *J* = 10.3, 1.9 Hz, 1H), 6.32 (dd, *J* = 10.3, 1.9 Hz, 1H), 4.64 (d, *J* = 14.4 Hz, 1H), 4.44 (d, *J* = 14.4 Hz, 1H), 3.30 (d, *J* = 5.7 Hz, 1H), 3.04 (dd, *J* = 21.3, 12.3 Hz, 2H), 2.60 (d, *J* = 8.2 Hz, 1H), 1.11 (s, 3H); $^{13}$C NMR (101 MHz, $CDCl_3$) δ: 184.7, 176.6, 148.4, 147.5, 136.4, 135.4, 132.7, 131.9, 131.5, 130.9, 130.3, 129.8, 129.1, 128.5, 52.9, 51.2, 48.8, 47.2, 38.9, 18.2; HRMS *m/z* (ESI) calcd for $C_{24}H_{22}Cl_2NO_2$ $[M+H]^+$ 426.1023, found: 426.1026.

### 3. Control experiments using $H_2O^{18}$ according to known method[2]

**Experiment procedure**: To a mixture of *N*-(4-fluorobenzyl)acrylamide **1** (0.3 mmol), $Ru(bpy)_3Cl_2$ (0.0015mmol, 5 mol%), $H_2O^{18}$ (1.5 mmol, about 90% atom of $O^{18}$), $K_3CO_3$ (0.6 mmol) in dry DMF (2.0 mL) was added 4-nitroaryldiazonium tetrafluoroborate (0.6 mmol) under $N_2$ atmosphere, and then the resulting solution was stirred under 5 W blue LED irradiation at room temperature for 24 h. The residue was purified by flash chromatography (petroleum ether/ethyl acetate 3:1 as the eluent) on silica gel to afford the disired 2-azaspiro[4.5]deca-6,9-diene-3,8-diones, and then the characterization ($^{13}$NMR data, $CDCl_3$ ) of the product thereby obtained was recorded on 400 Hz NMR spectrometer (Shown as following).

**$^{13}$C NMR (101 MHz, $CDCl_3$): C=$O^{18}$, δ: 184.87**
**C=$O^{16}$, δ: 184.82**

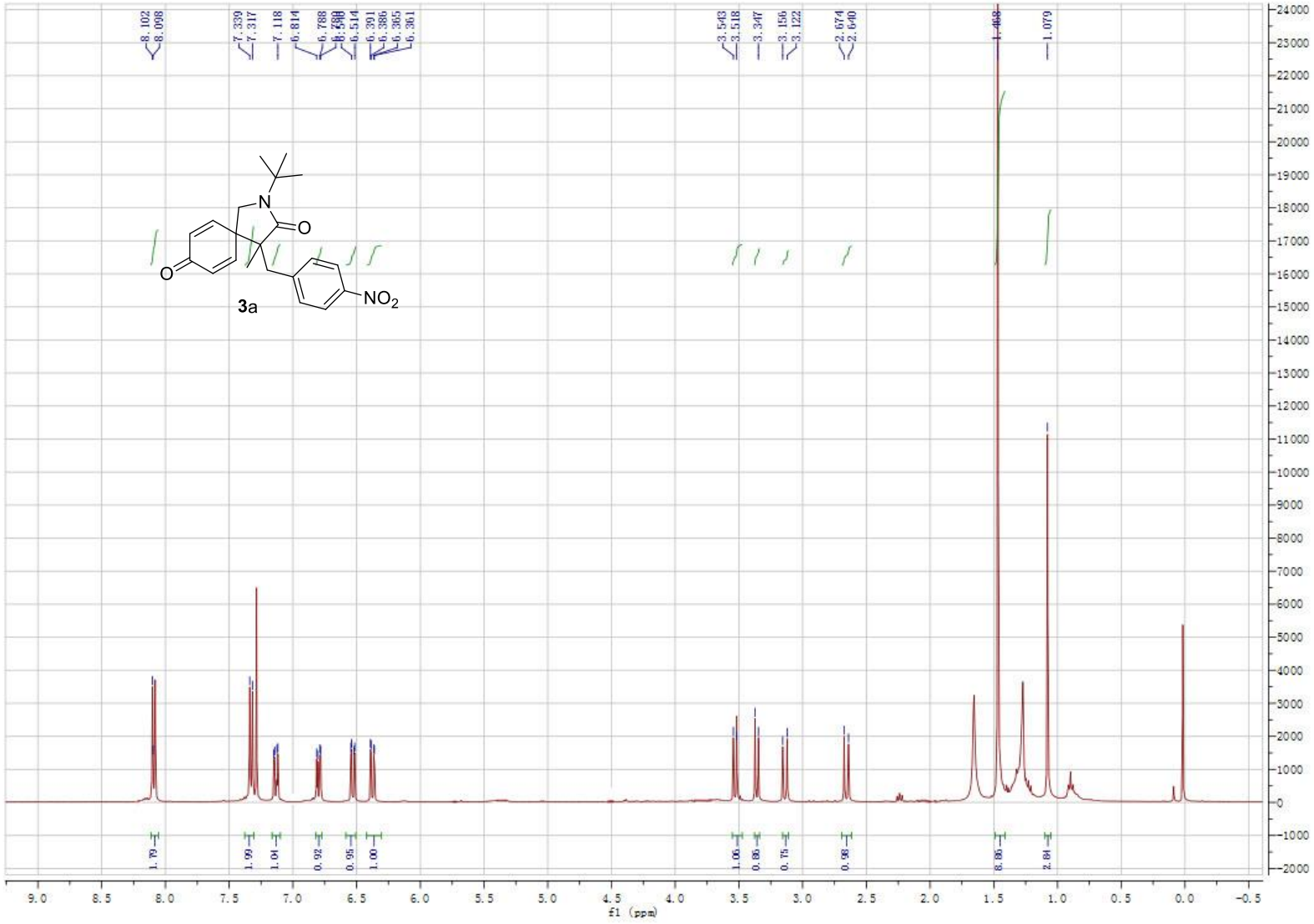



***References:***

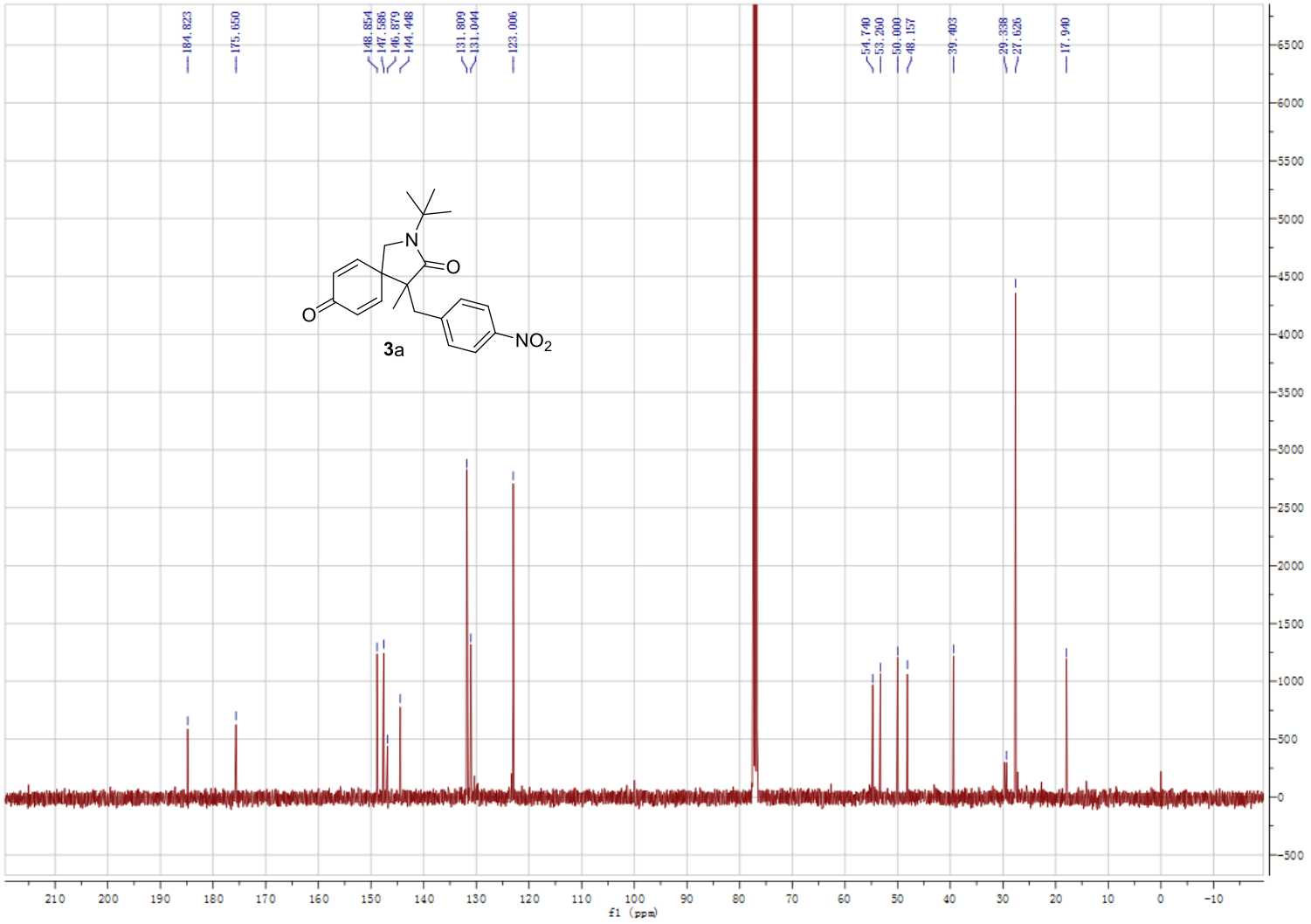
(1) Z. -X. Zhang, X.-J. Tang and W. R. Dolbier Jr, *Org. Lett.*, 2016, **18**, 1048; (d) B. Hu, Y.-Y. Li, W.-H. Dong, K. Ren, X.-M. Xie, J. Wan and Z.-G. Zhang, *Chem. Commun.*, 2016, **52**, 3709.
(2) J.M. Risley, R. L. Van Etten, *J. Am. Chem. Soc.*, **1979**, *101*, 252-253. (b) Jean E. Parente, John M. Risley, Robert L. Van Etten, *J. Am. Chem. Soc.*, 1984, **106**, 8156-8161

## 2 Copies of $^{1}$H NMR and $^{13}$C NMR

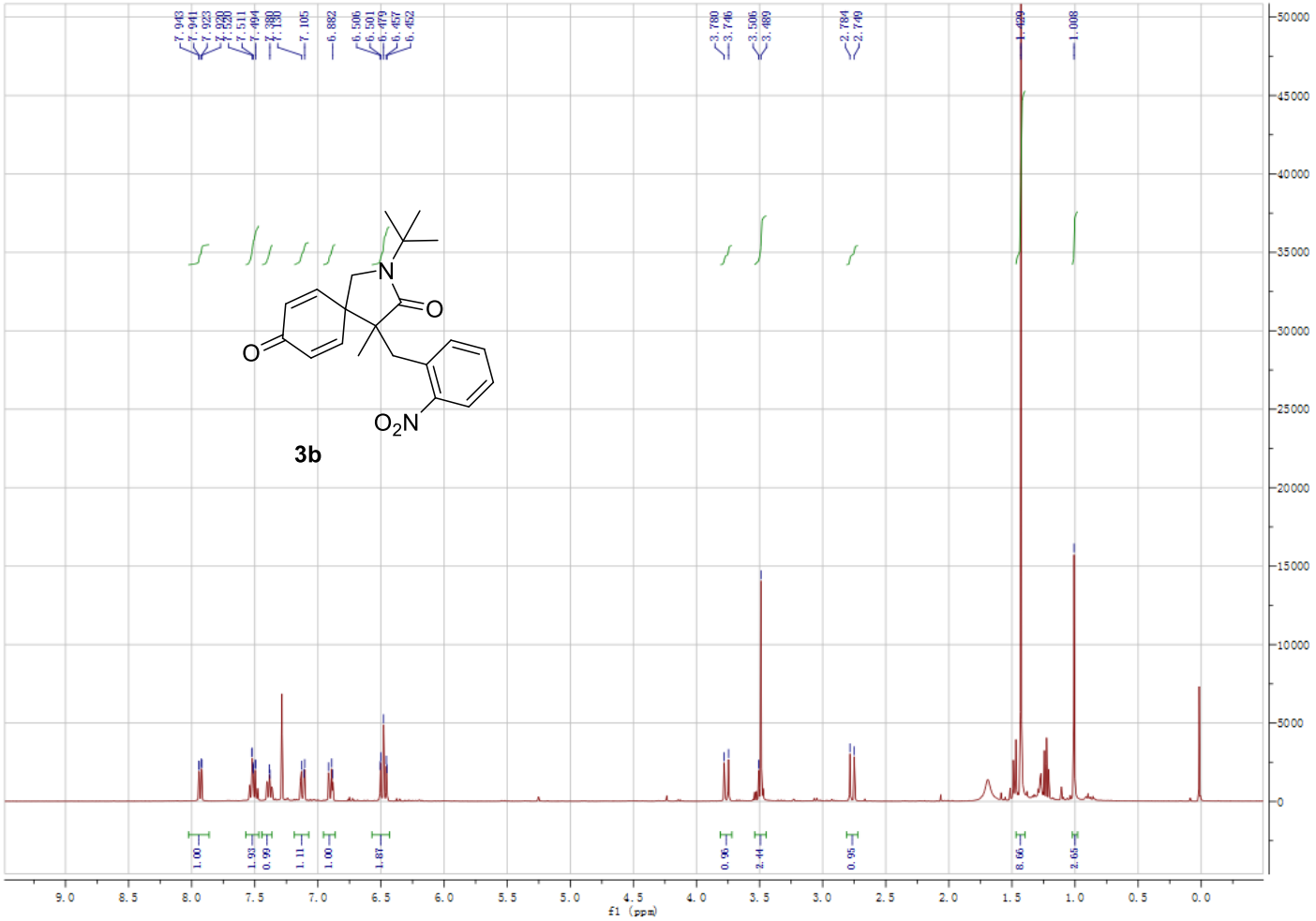
**2-(Tert-butyl)-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3a)**



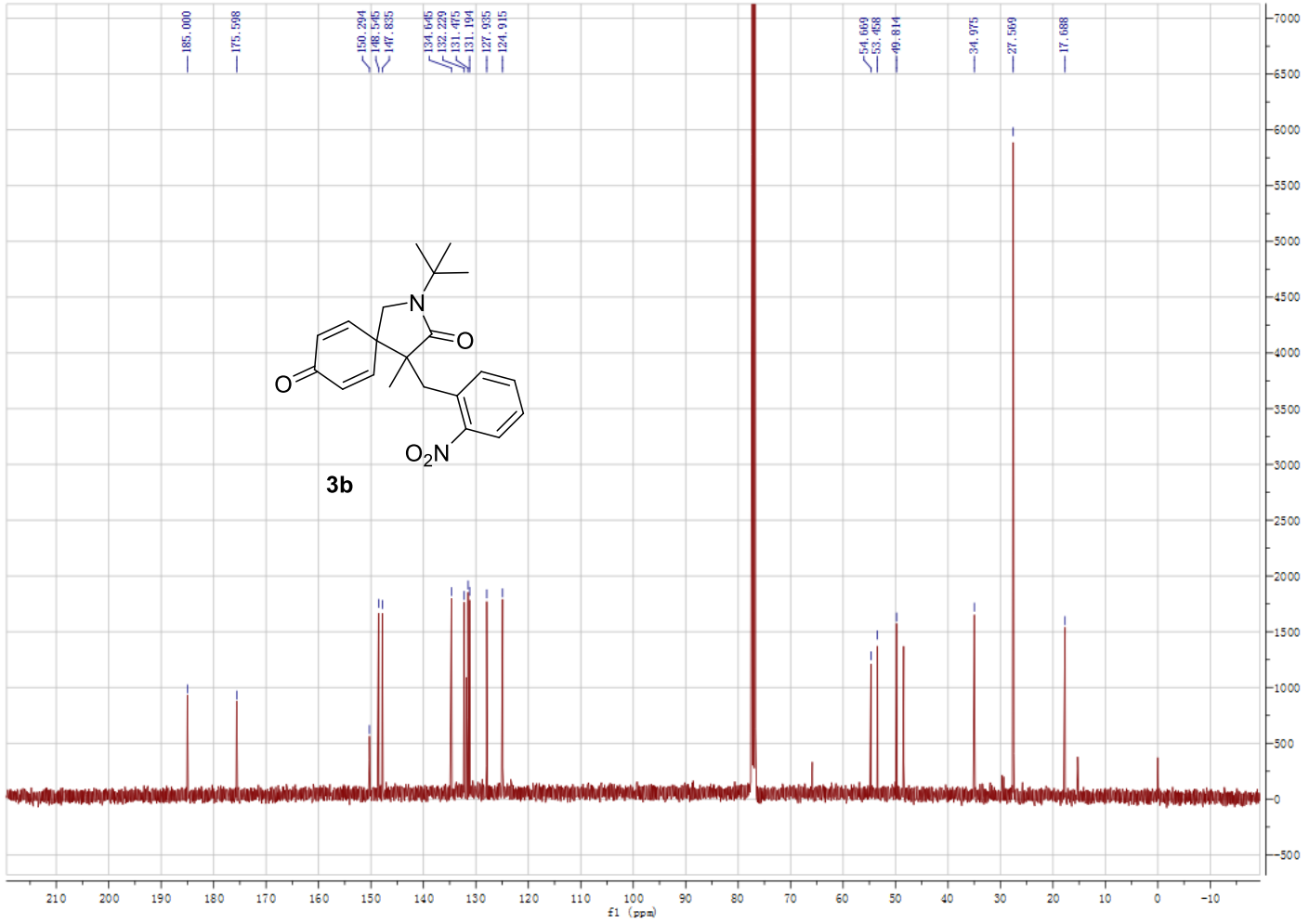
**2-(Tert-butyl)-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3a)**



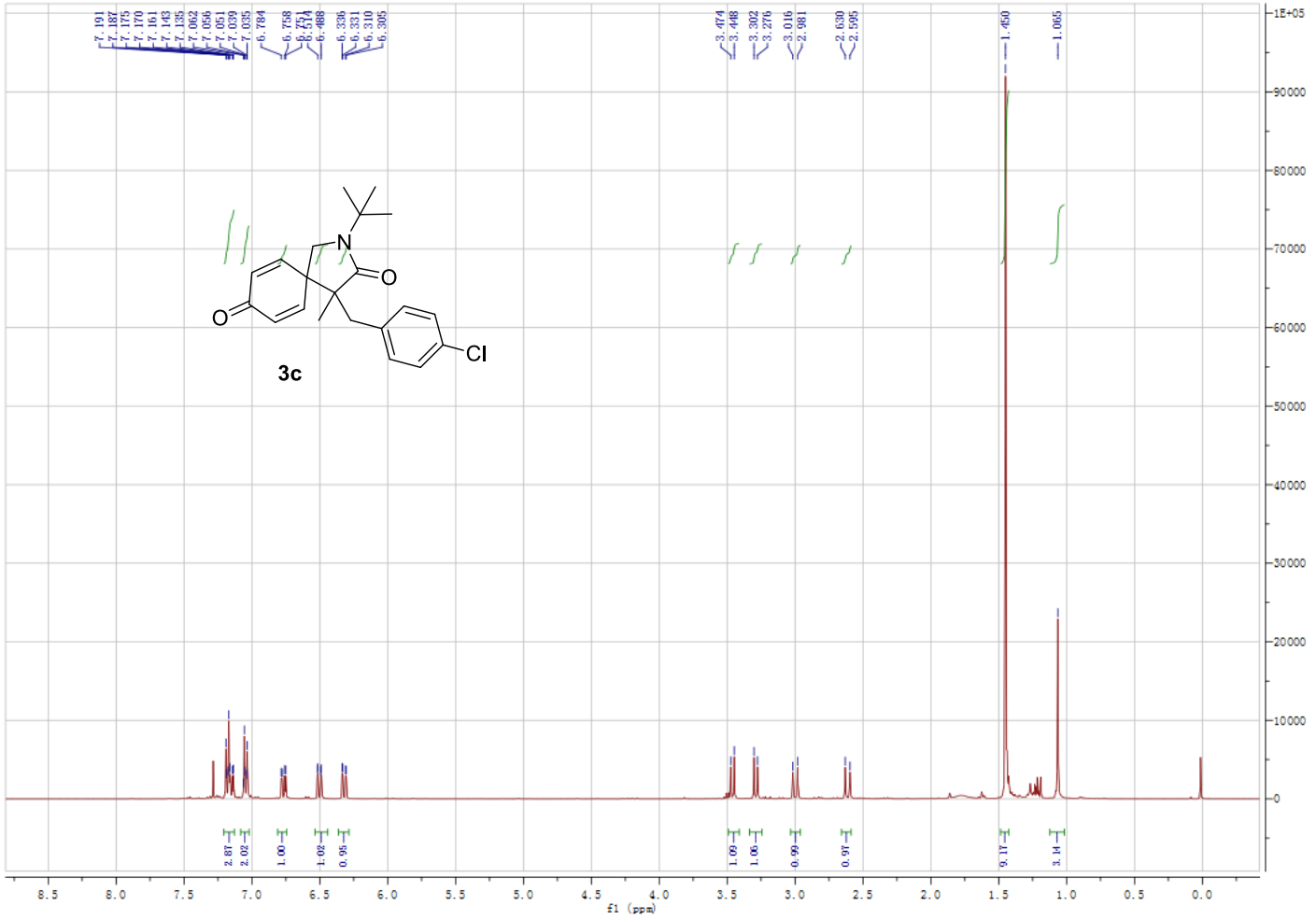
***2-(Tert-butyl)-4-methyl-4-(2-nitrophenyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3b).***



***2-(Tert-butyl)-4-methyl-4-(2-nitrophenyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3b).***



***2-(Tert-butyl)-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3c)***



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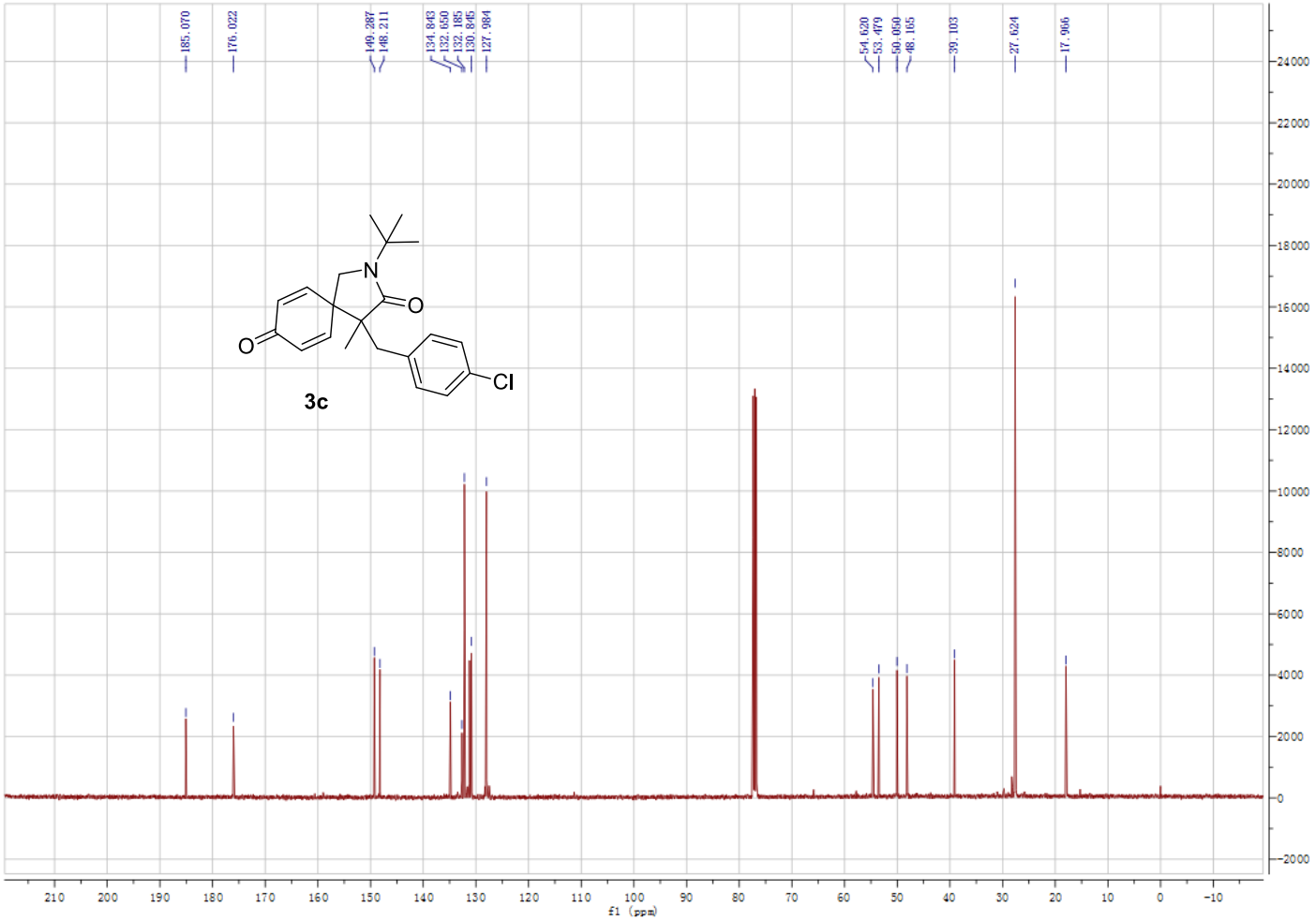
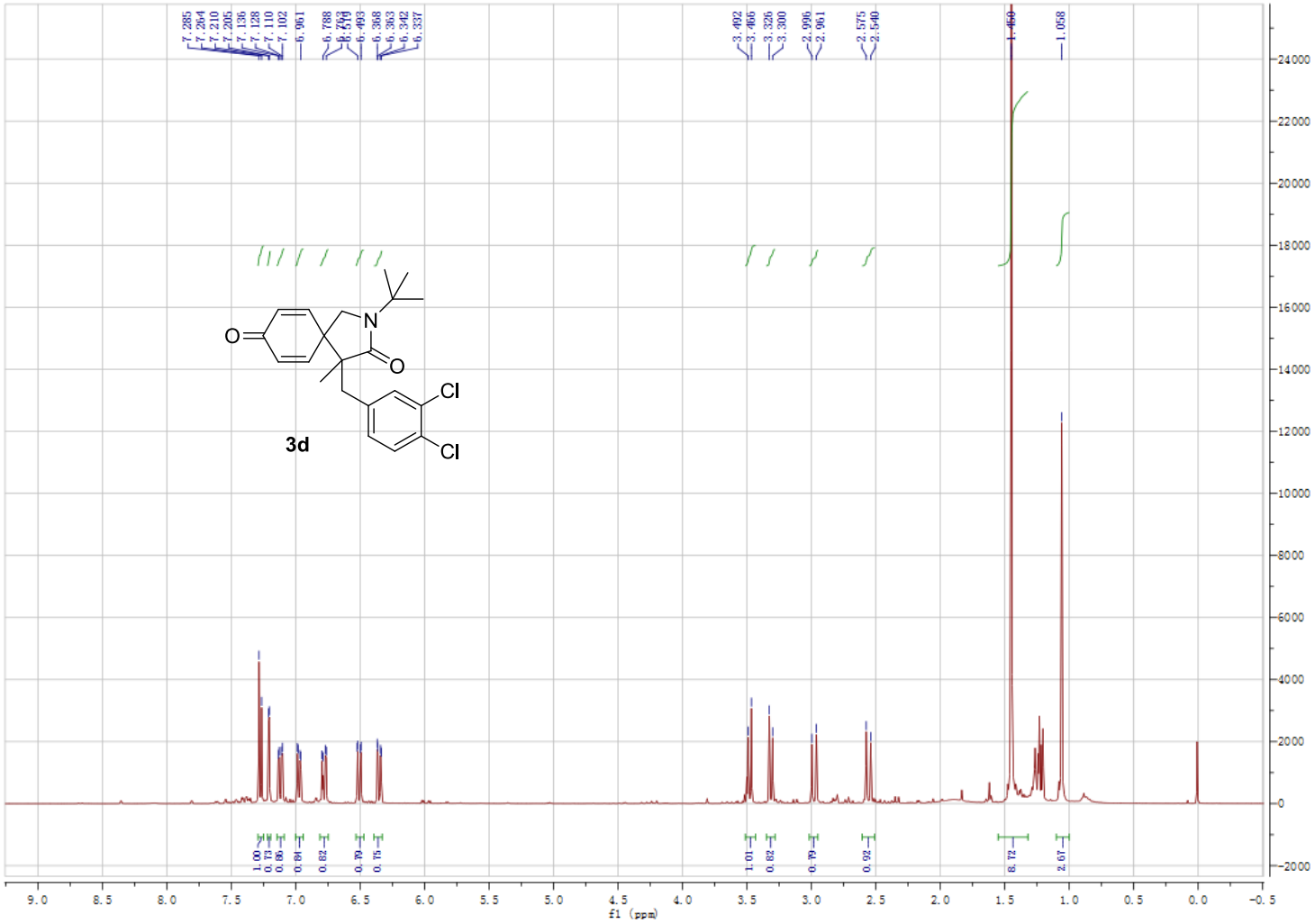
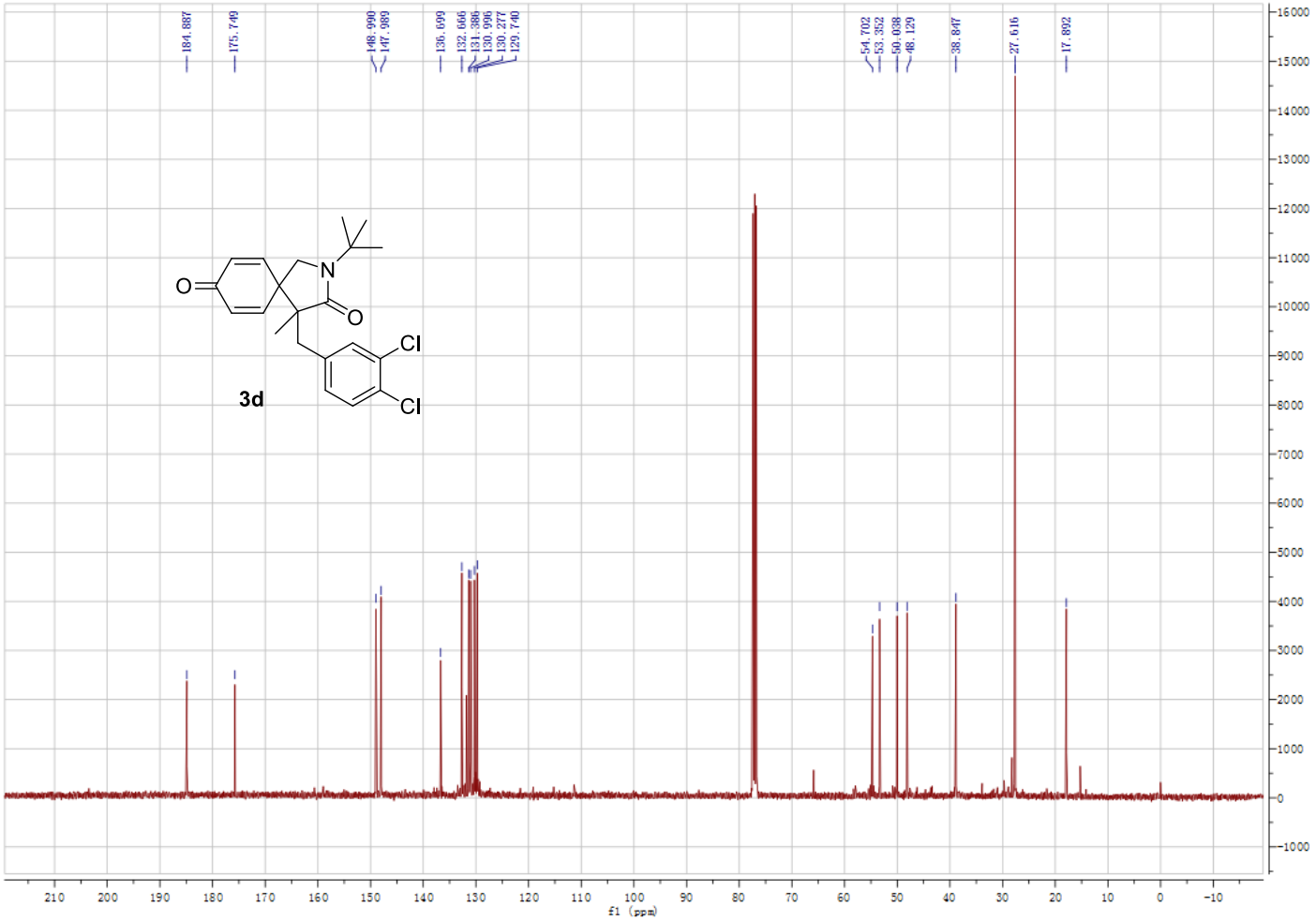
***2-(Tert-butyl)-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3c)***



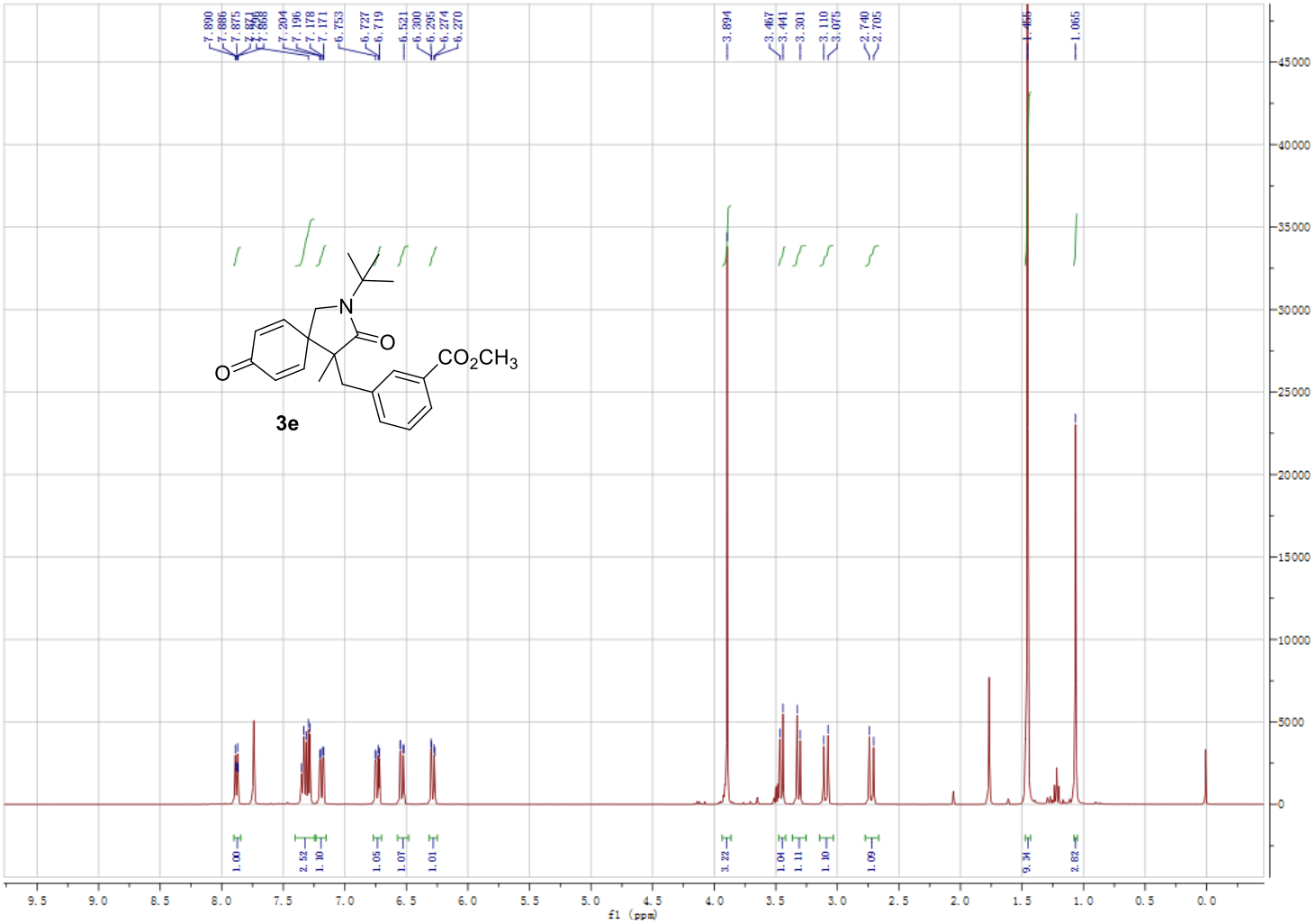
***2-(Tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3d).***



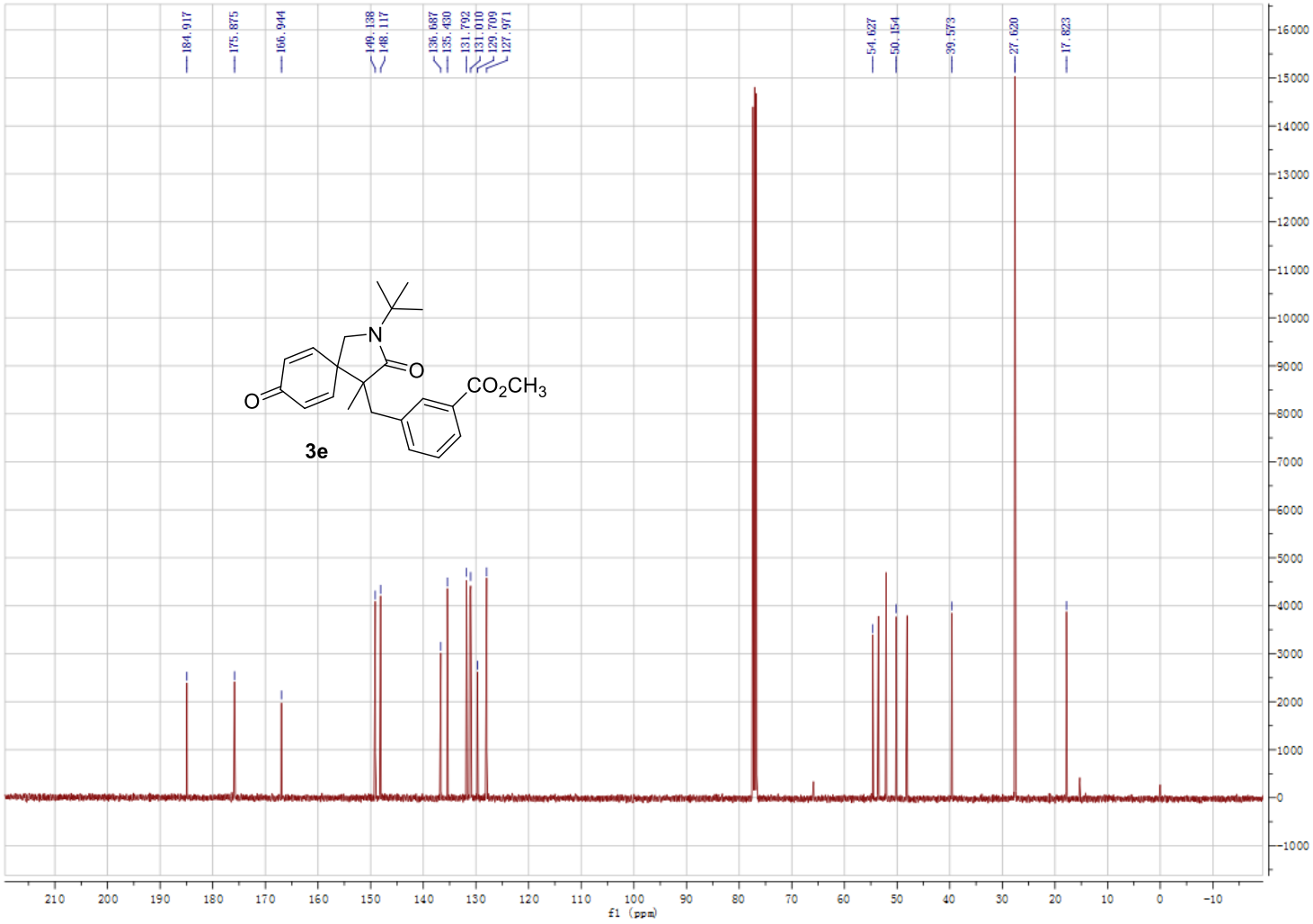
***2-(Tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3d).***



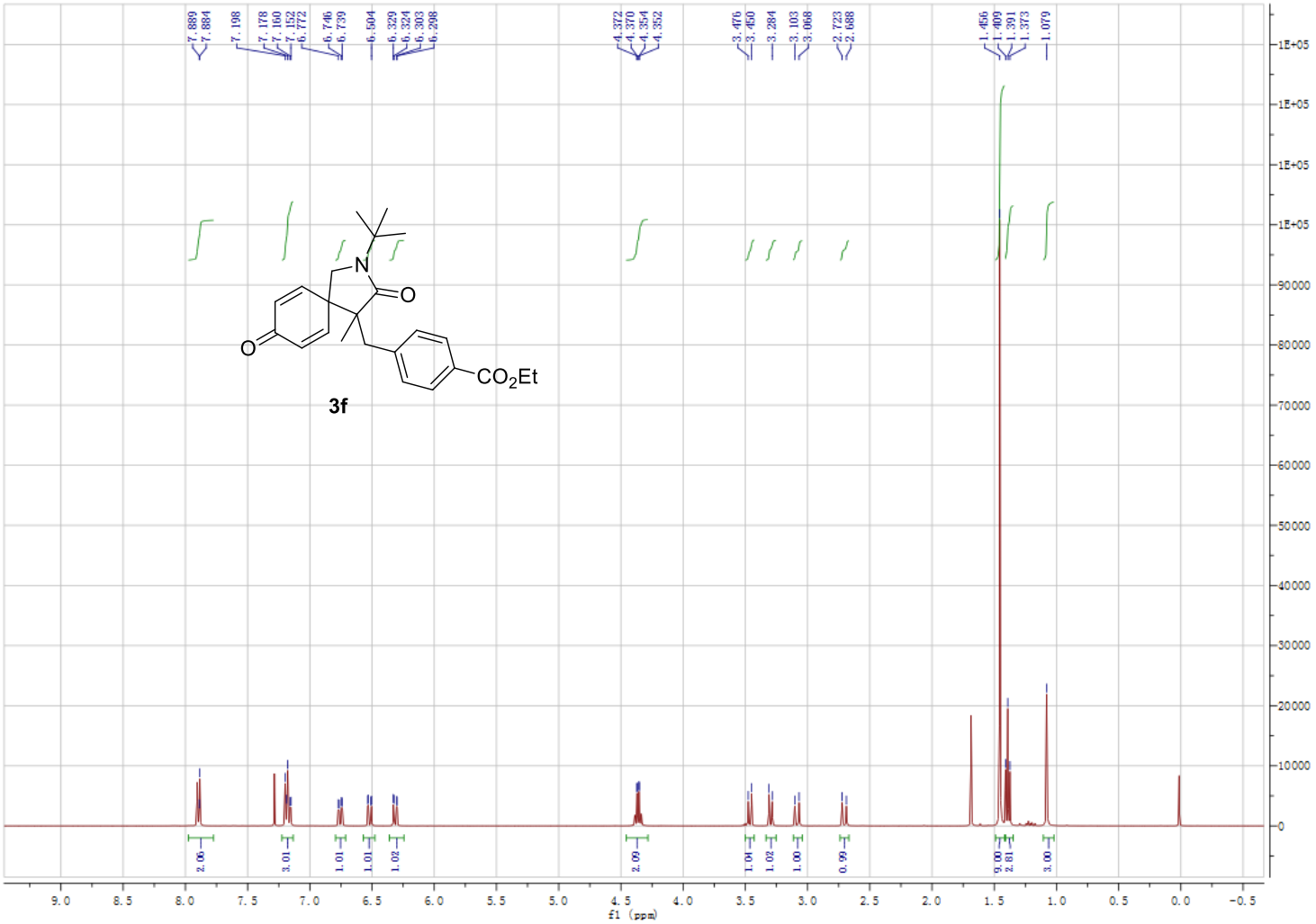
***Methyl 3-((2-(tert-butyl)-4-methyl-3,8-dioxo-2-azaspiro[4.5]deca-6,9-dien-4-yl)methyl)benzoate(3e).***



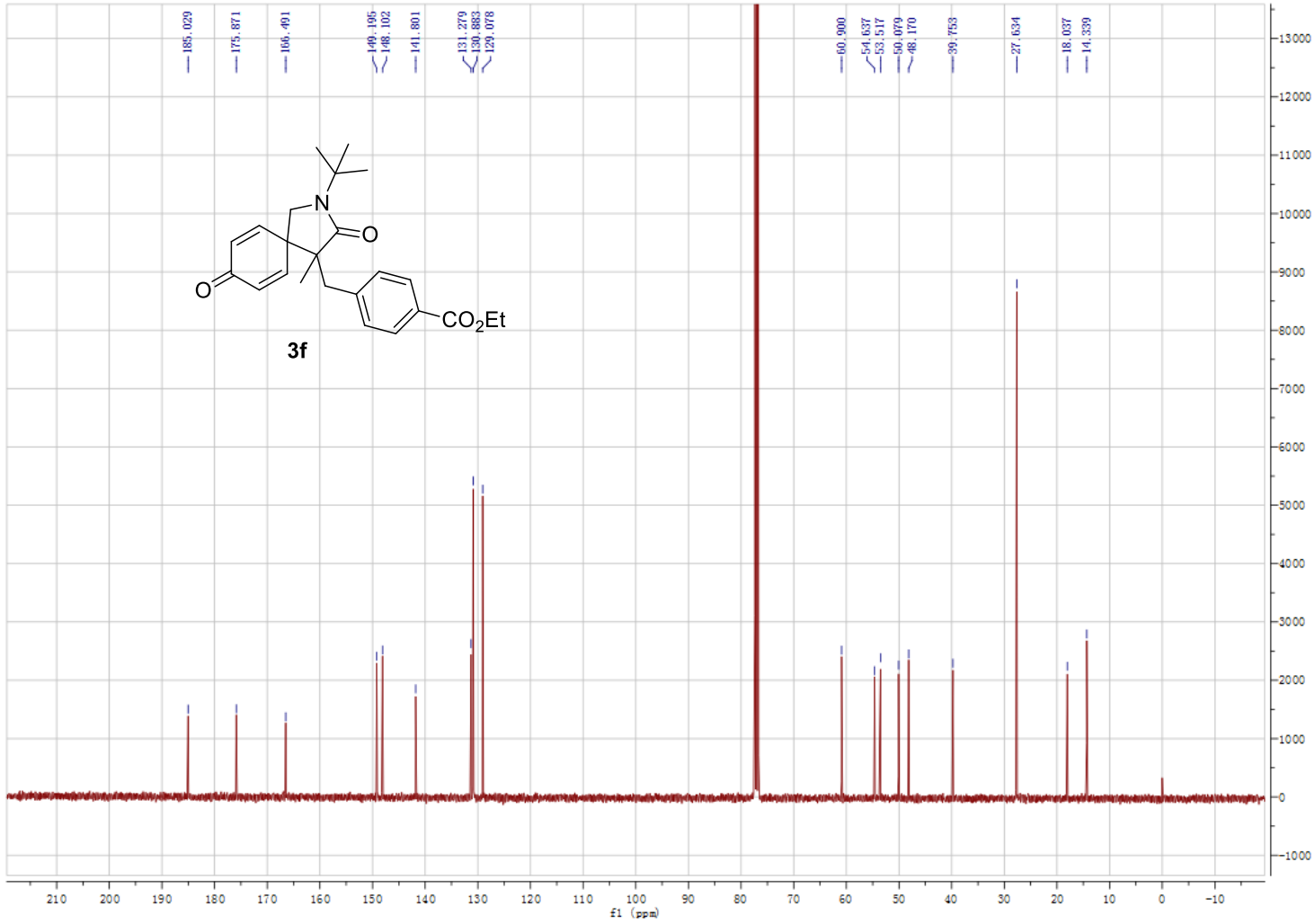
***Methyl 3-((2-(tert-butyl)-4-methyl-3,8-dioxo-2-azaspiro[4.5]deca-6,9-dien-4-yl)methyl)benzoate(3e)***



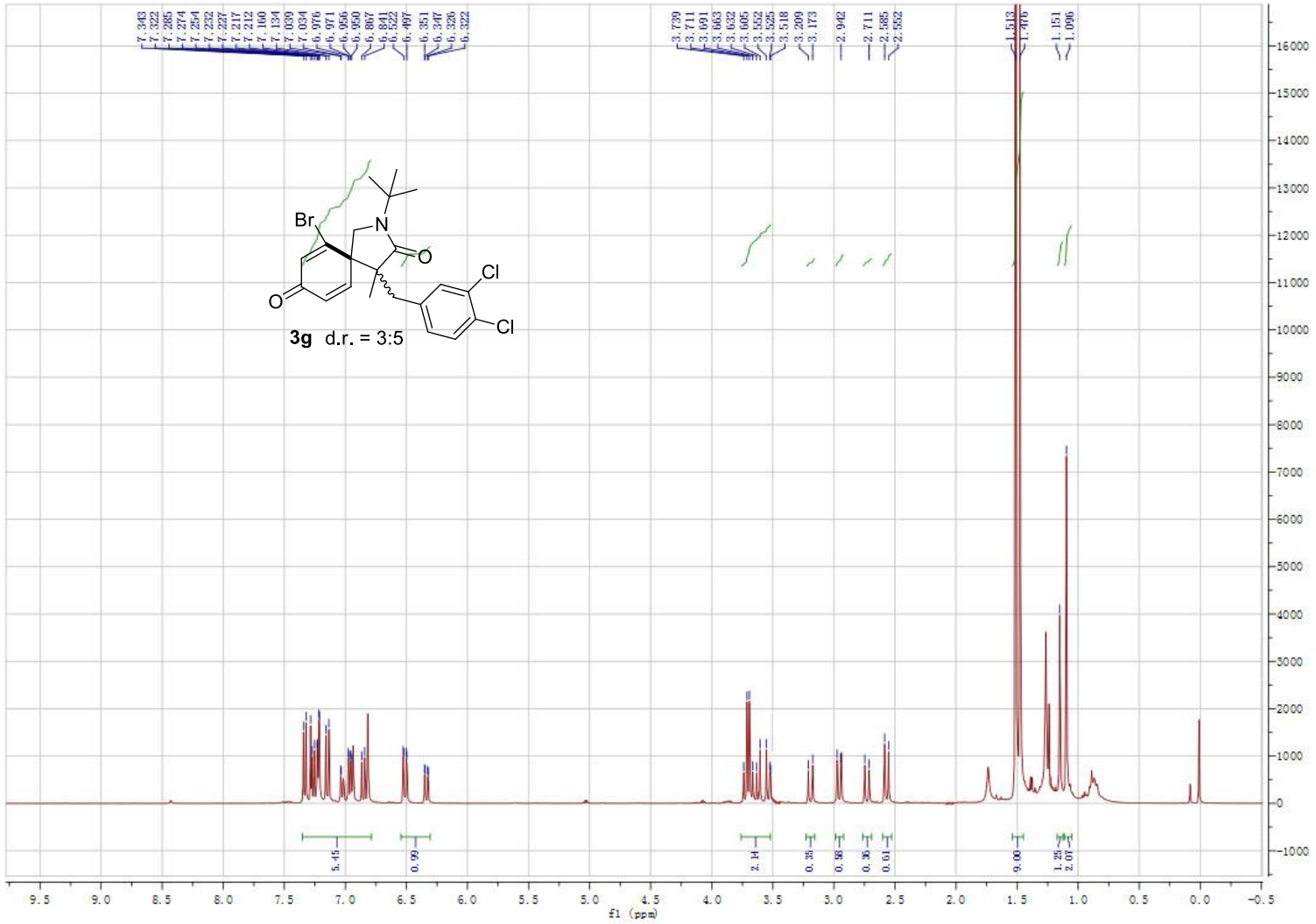
***Ethyl4-((2-(tert-butyl)-4-methyl-3,8-dioxo-2-azaspiro[4.5]deca-6,9-dien-4-yl)methyl)benzoate(3f).***



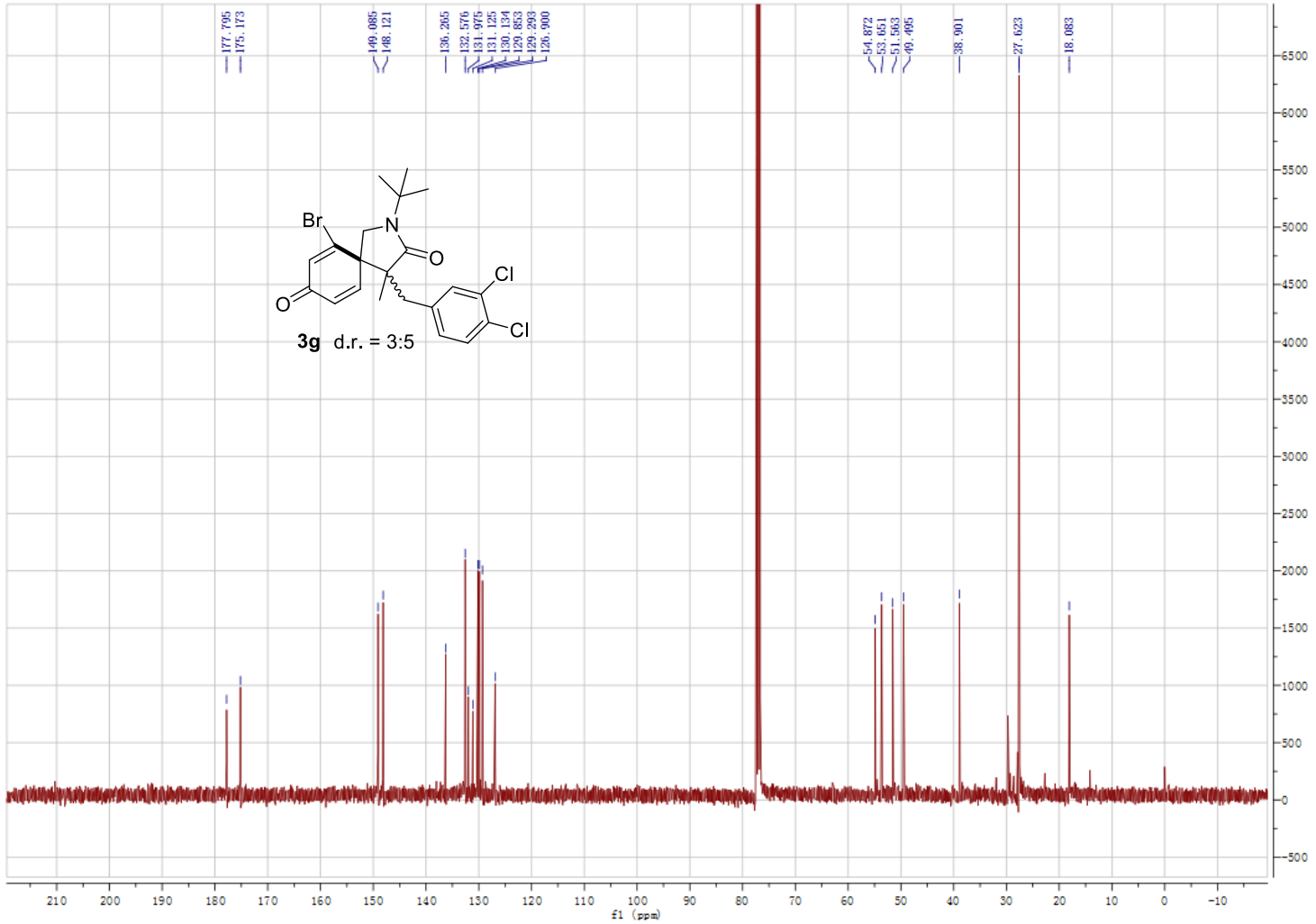
***Ethyl4-((2-(tert-butyl)-4-methyl-3,8-dioxo-2-azaspiro[4.5]deca-6,9-dien-4-yl)methyl)benzoate(3f).***



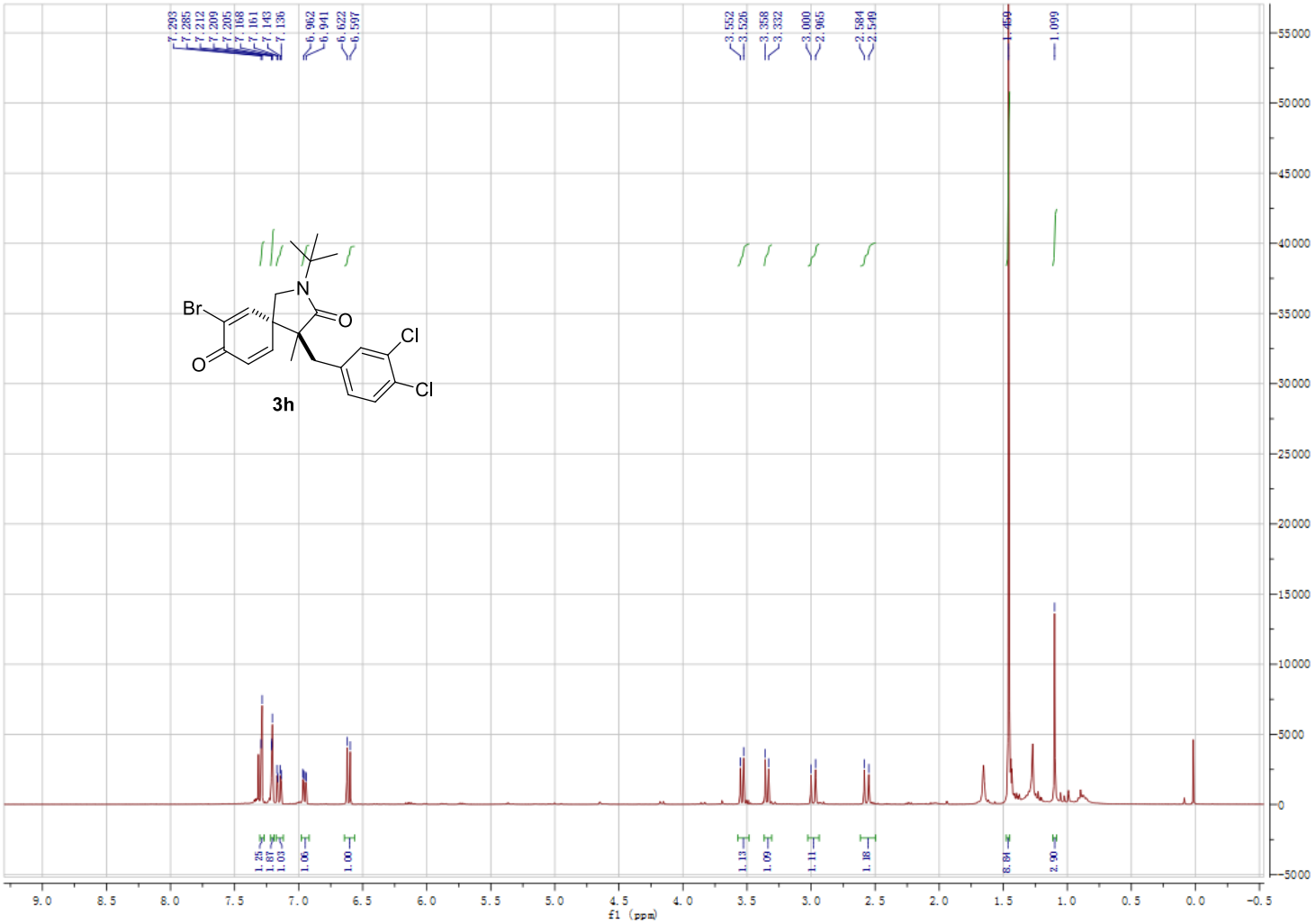
***6-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3g)***



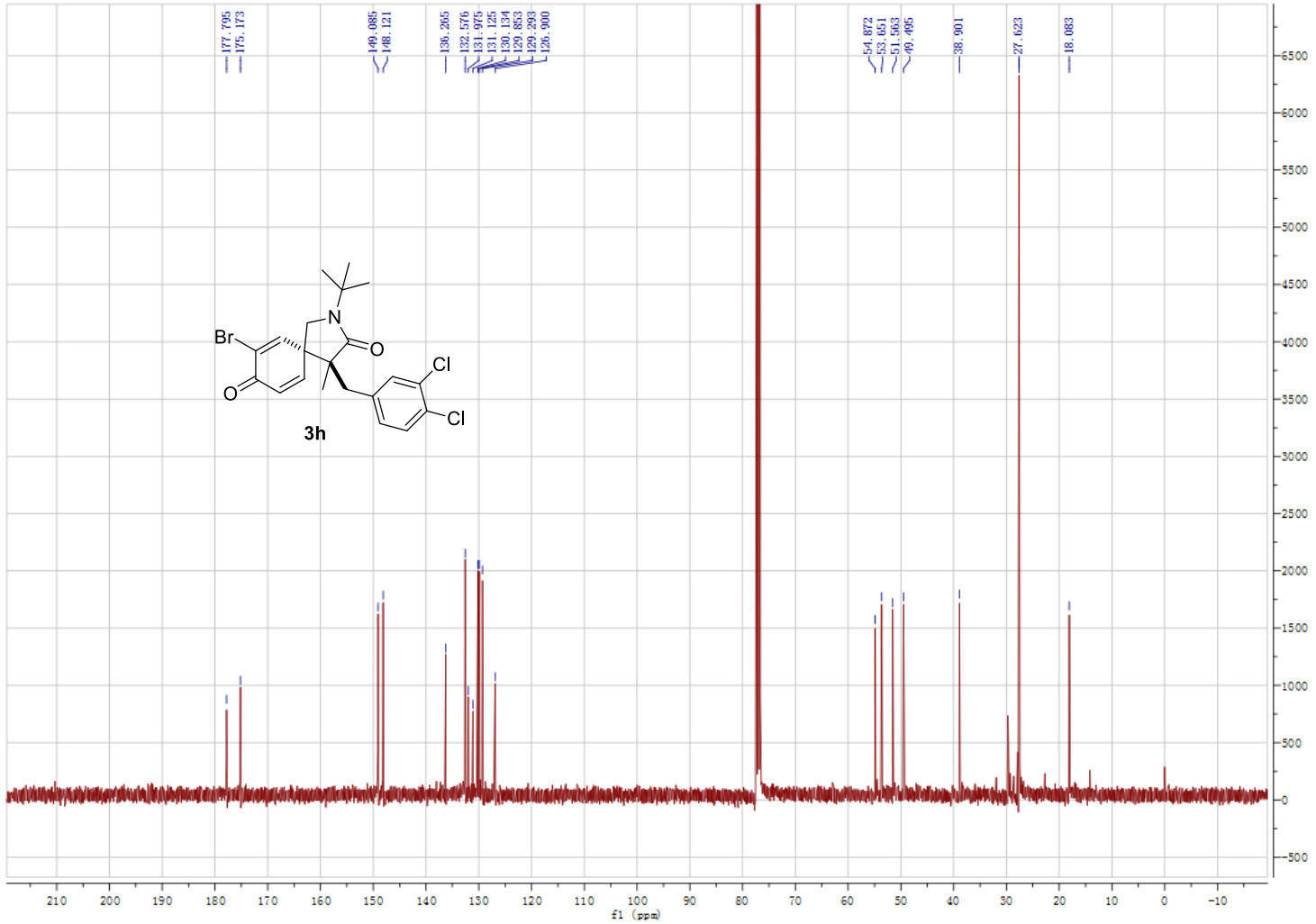
***6-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3g)***



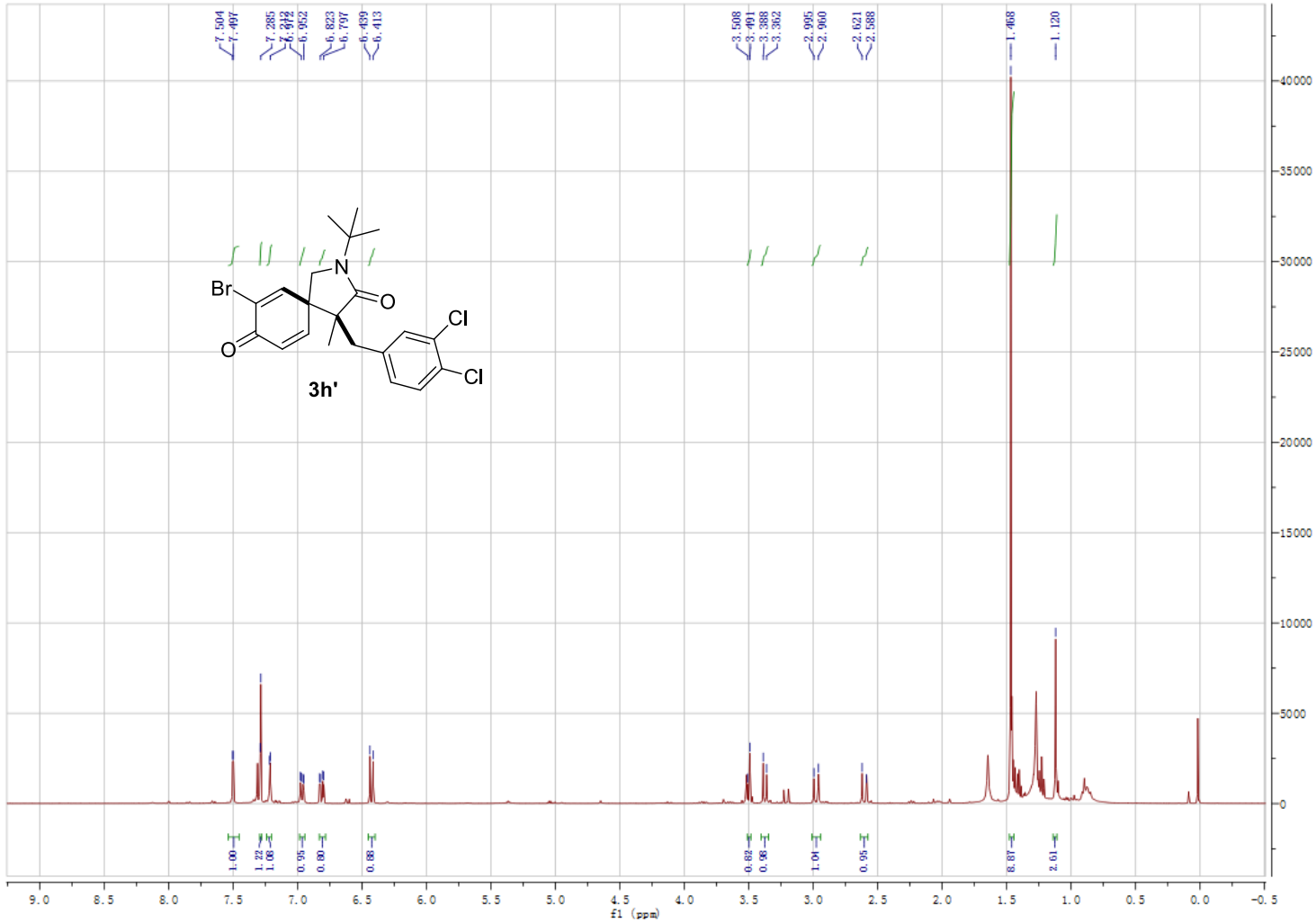
***7-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3h).***



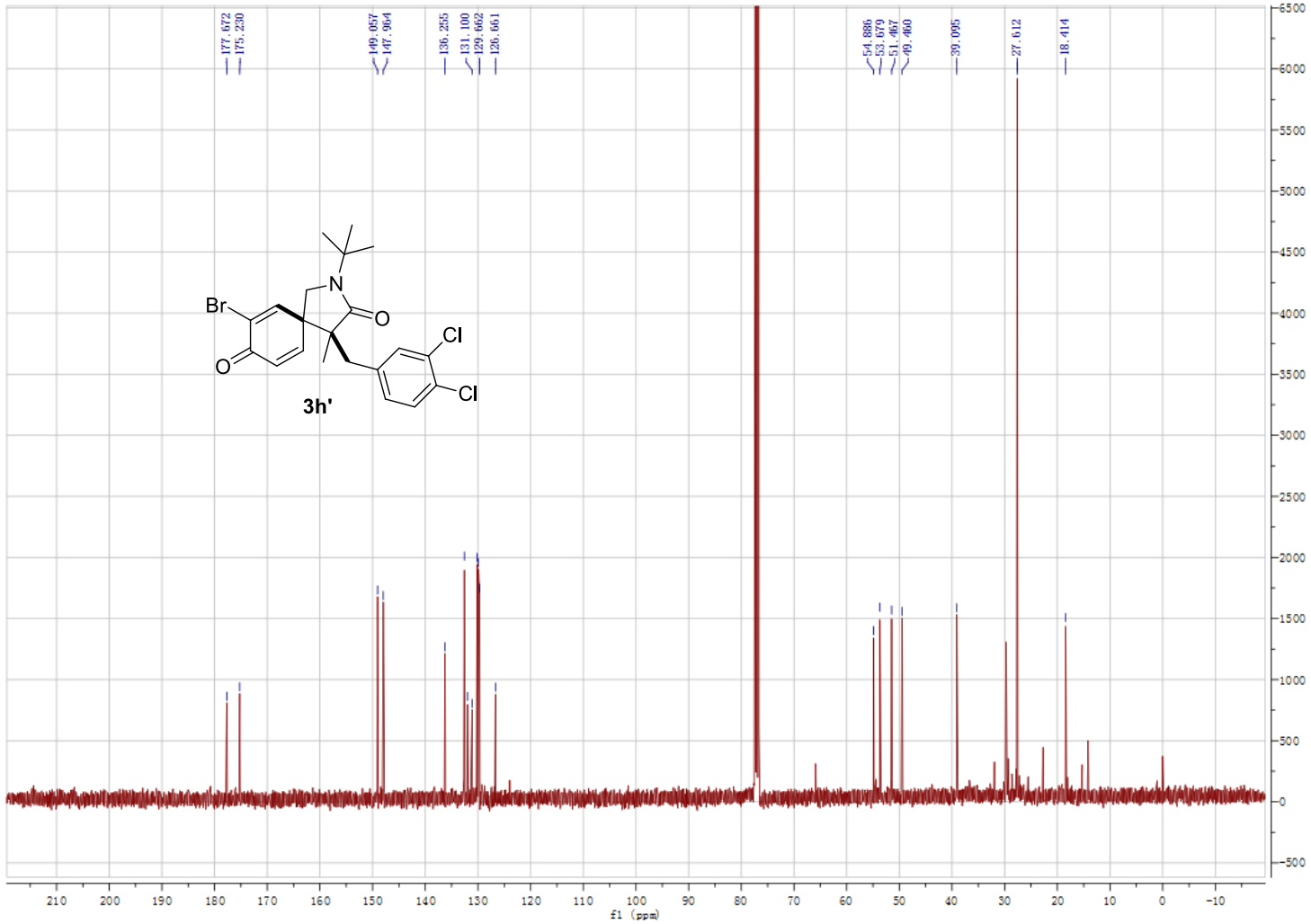
***7-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3h).***



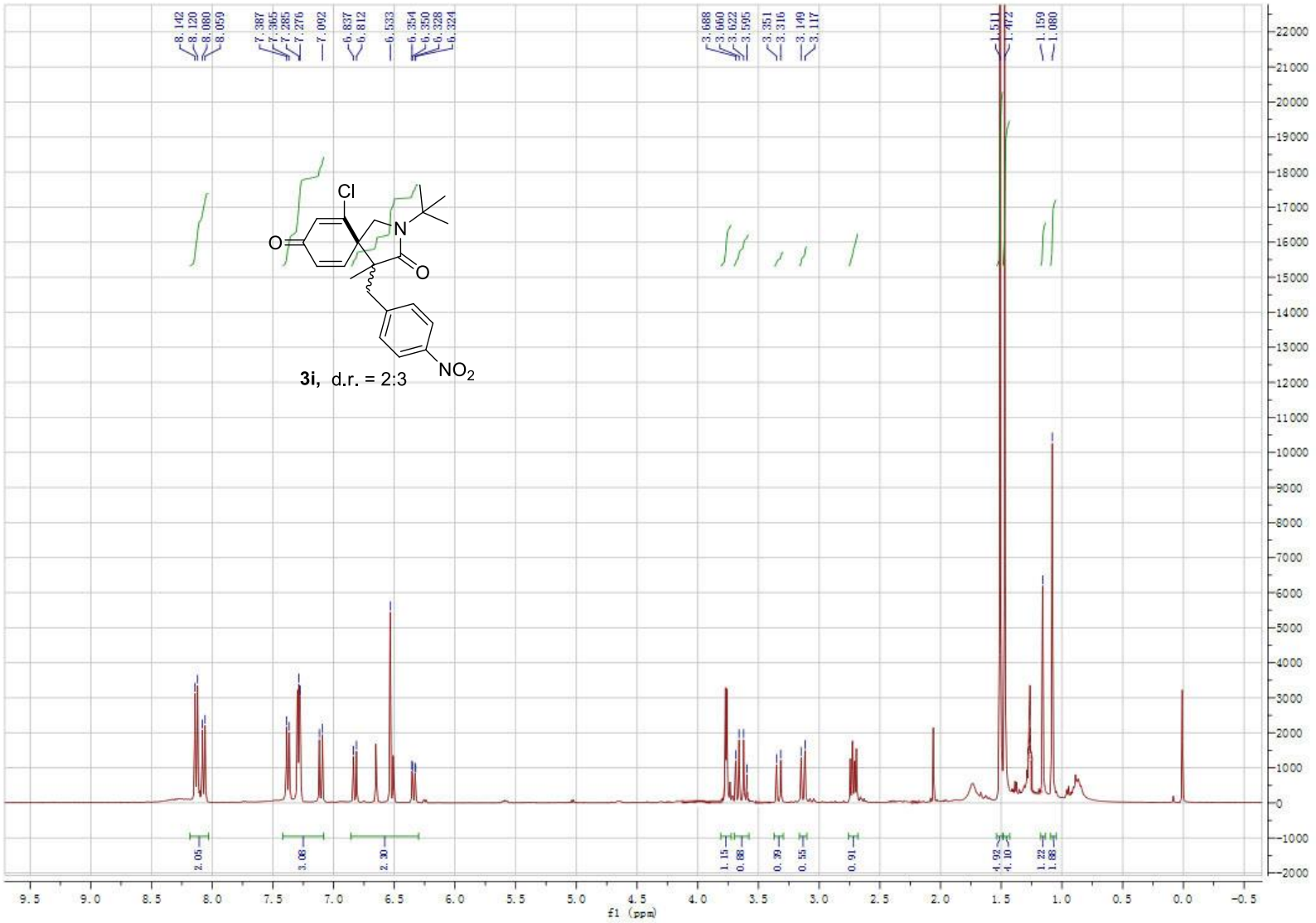
***7-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3h')***



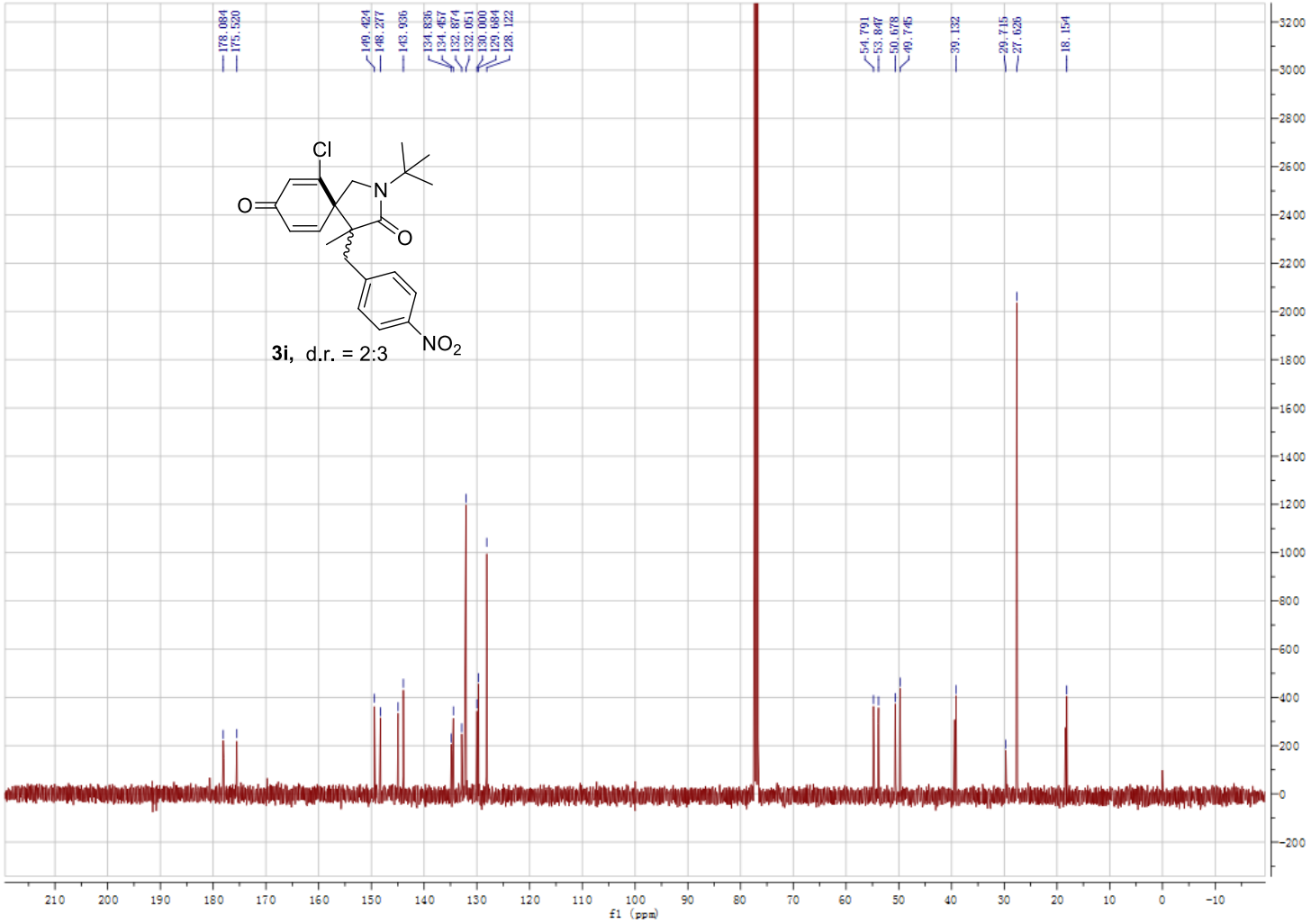
***7-Bromo-2-(tert-butyl)-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3h')***



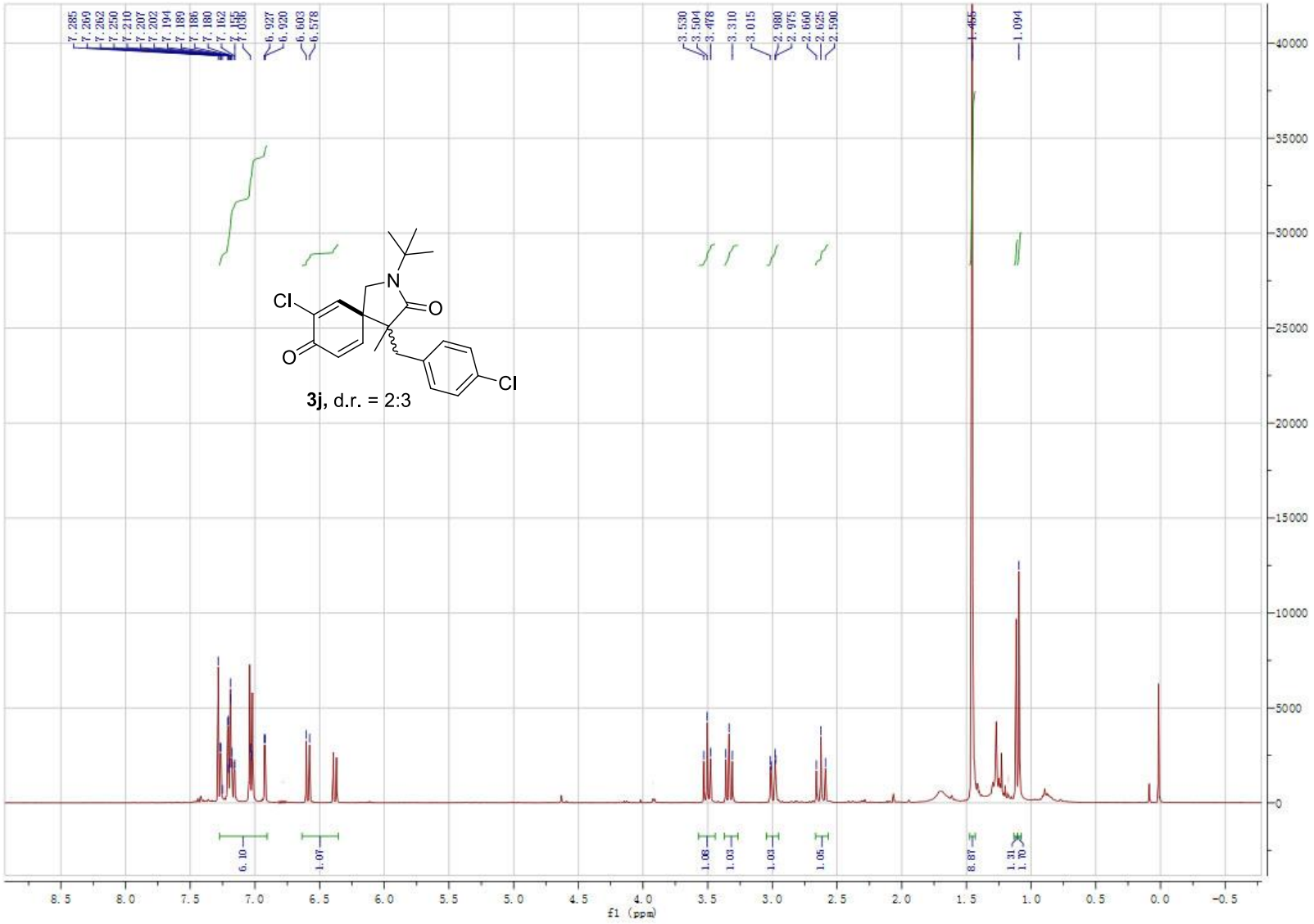
***2-(Tert-butyl)-6-chloro-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3i).***



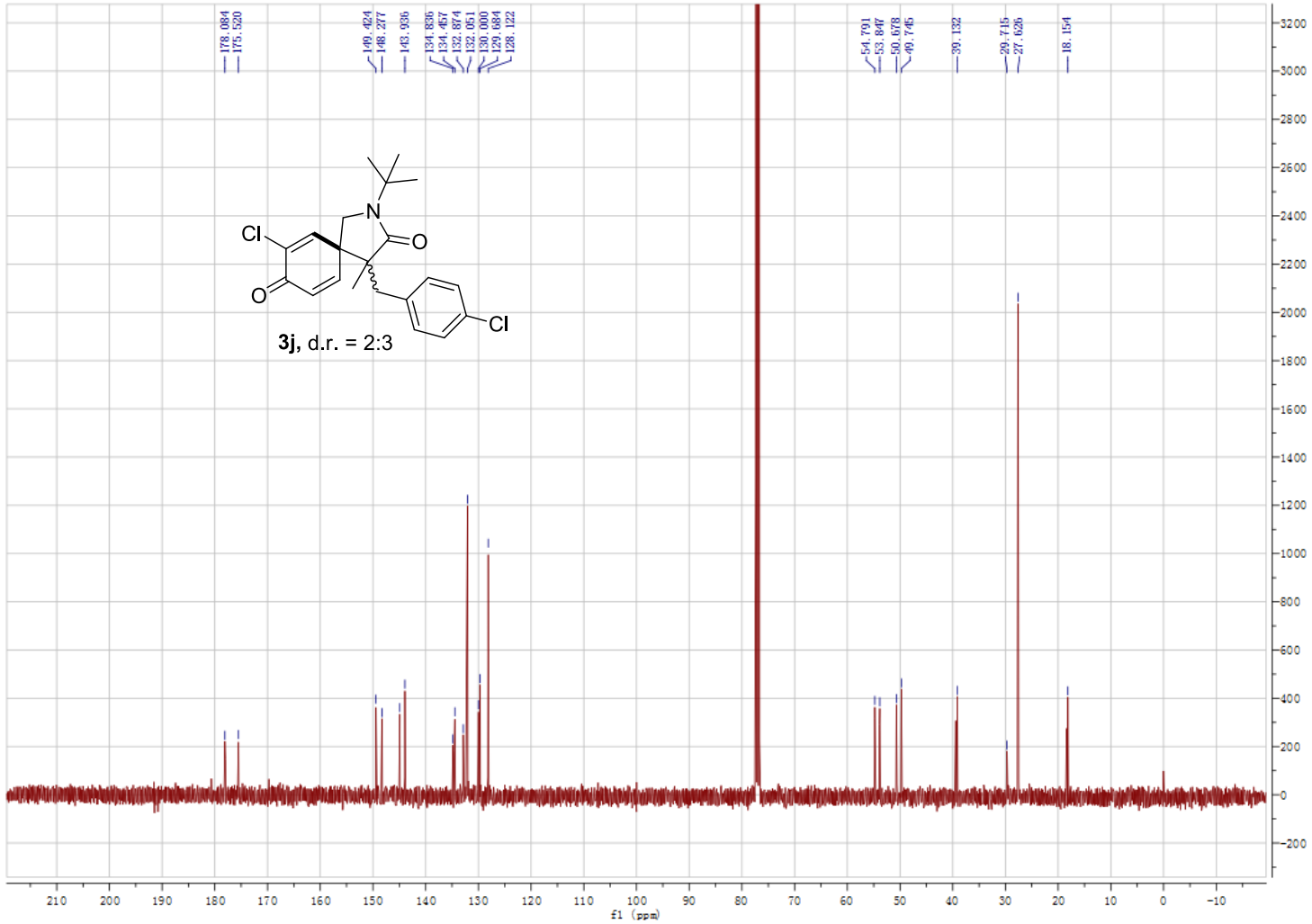
***2-(Tert-butyl)-6-chloro-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3i).***



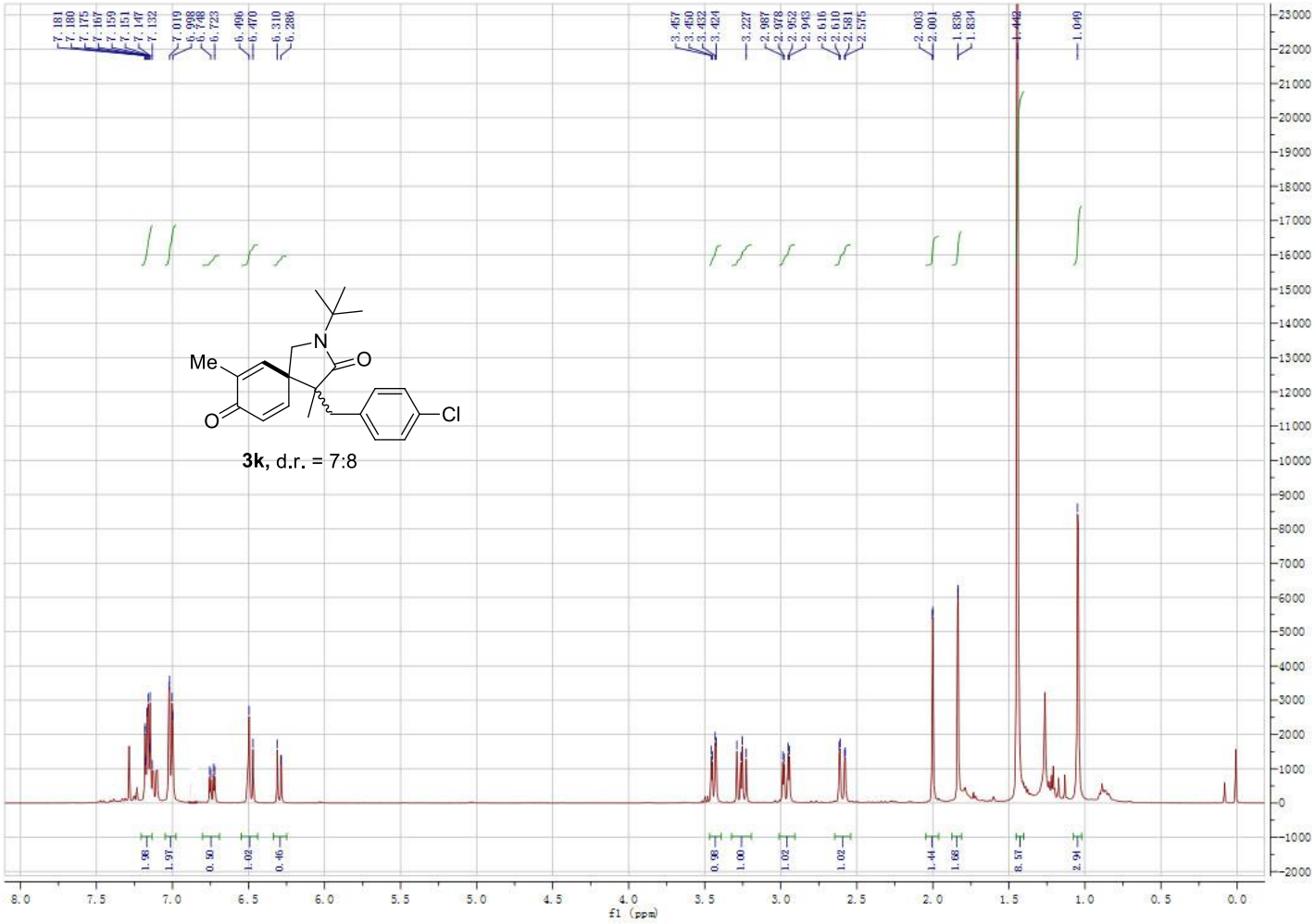
***2-(Tert-butyl)-7-chloro-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3j).***



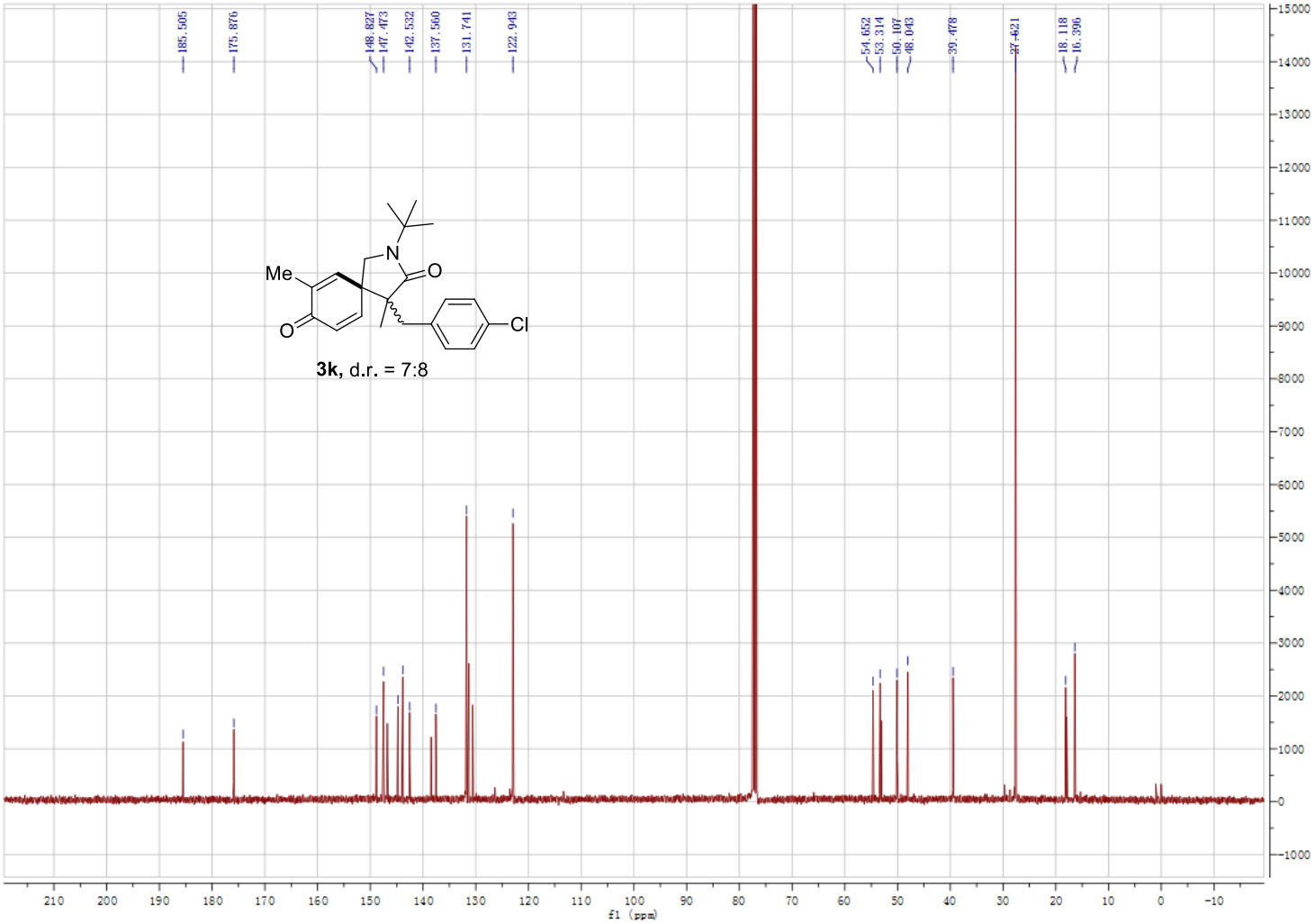
***2-(Tert-butyl)-7-chloro-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3j).***



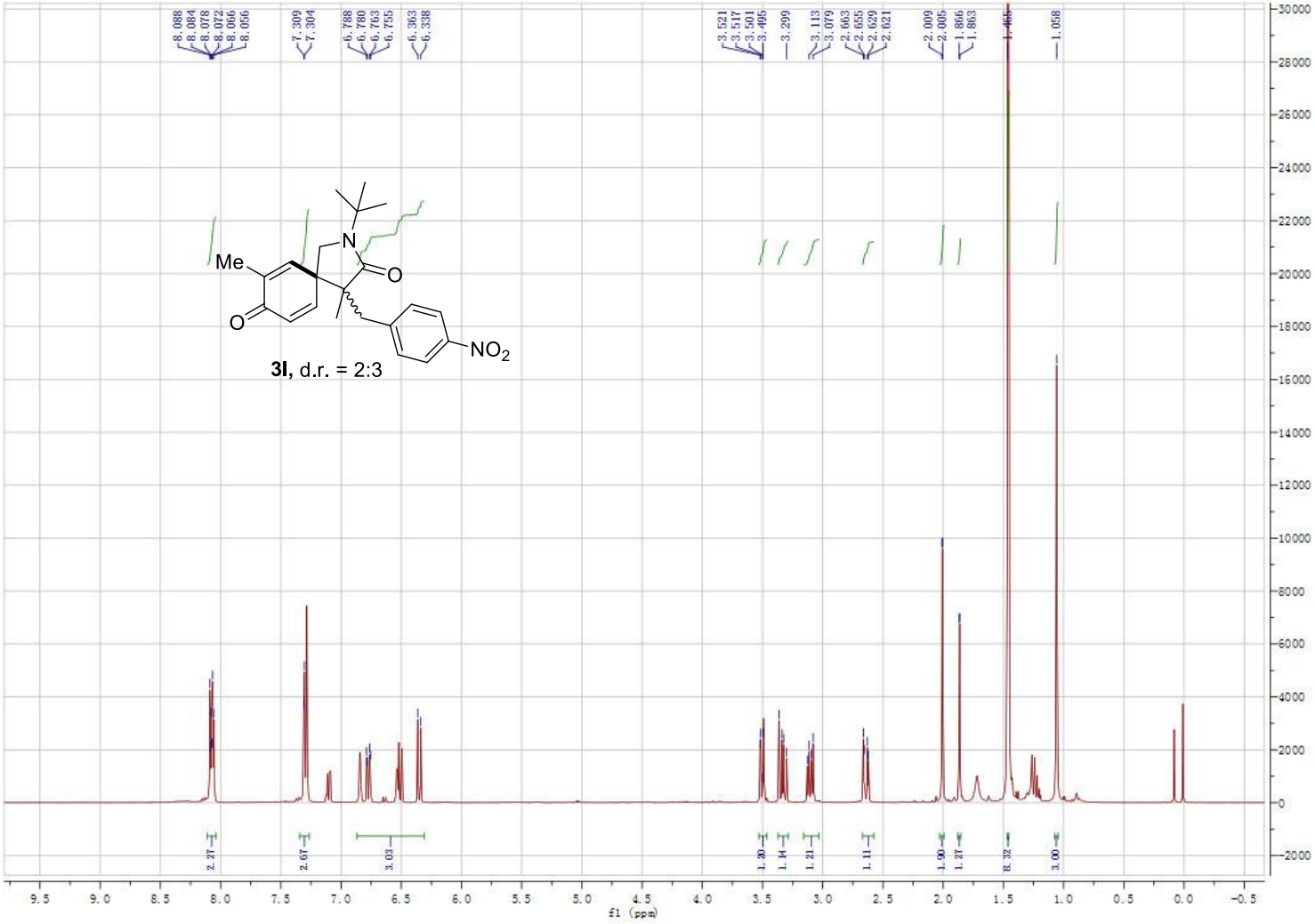
***2-(Tert-butyl)-4-(4-chlorobenzyl)-4,7-dimethyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3k)***



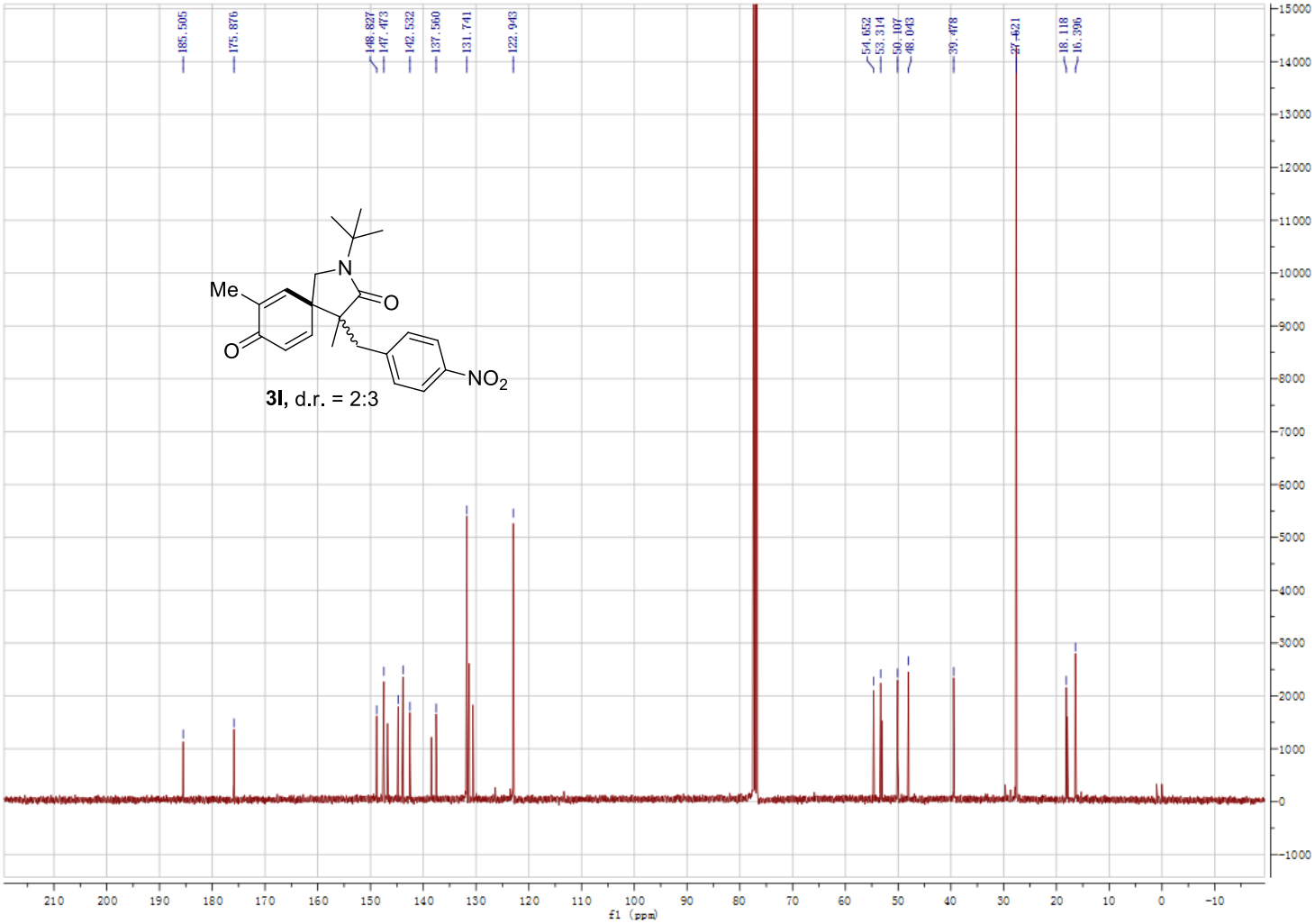
***2-(Tert-butyl)-4-(4-chlorobenzyl)-4,7-dimethyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3k)***



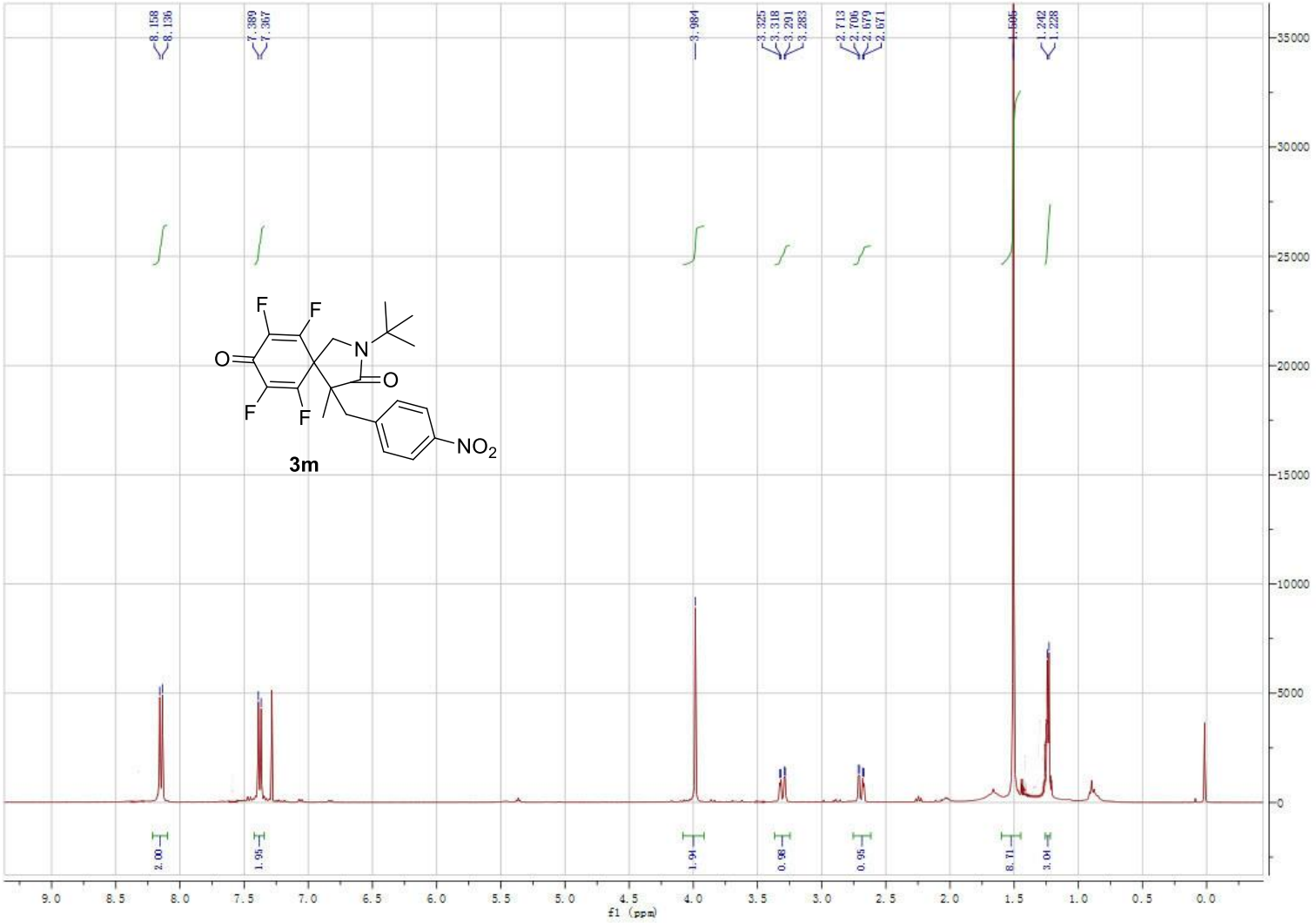
***2-(Tert-butyl)-4,7-dimethyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3l)*.**



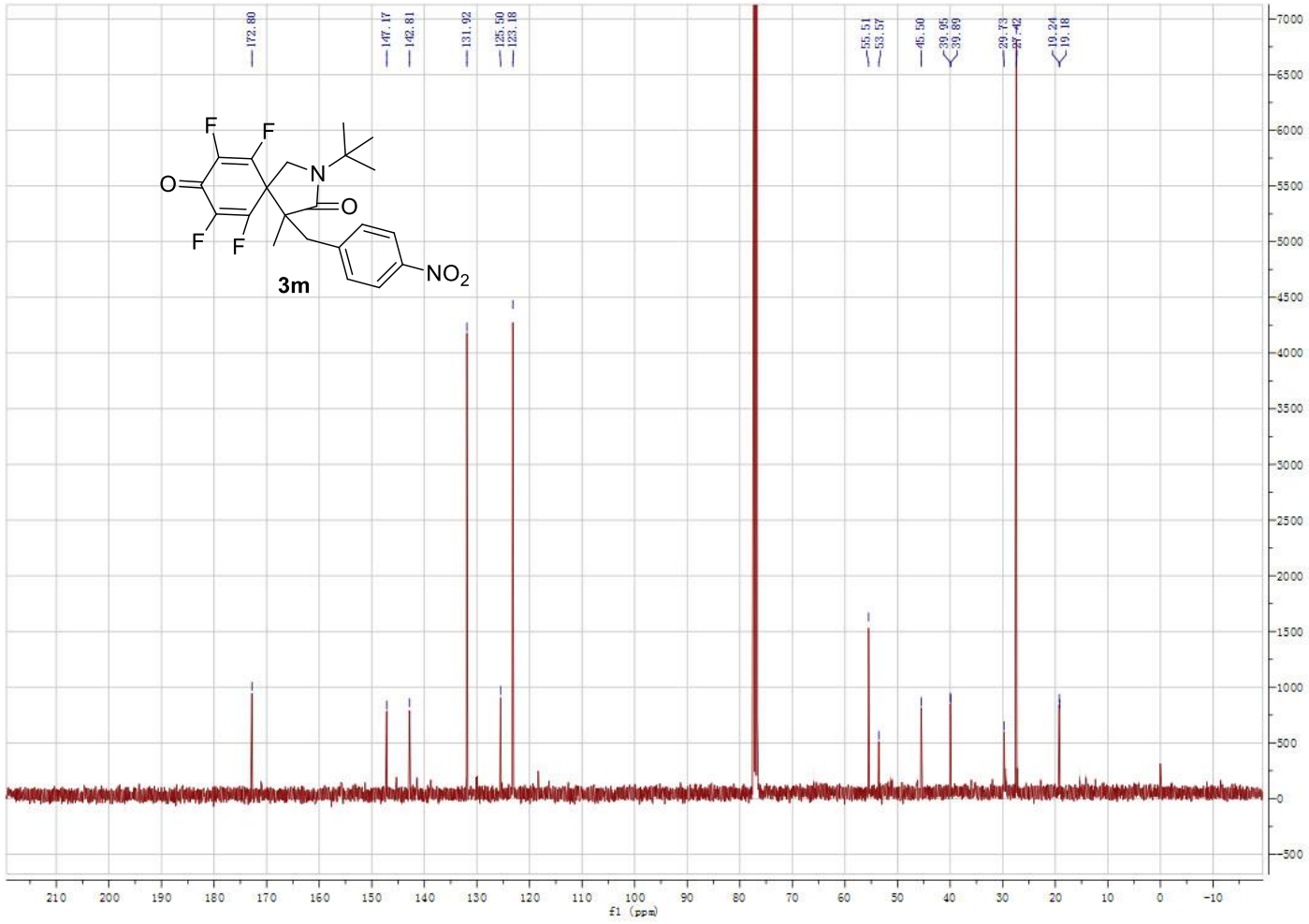
***2-(Tert-butyl)-4,7-dimethyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3l)*.**



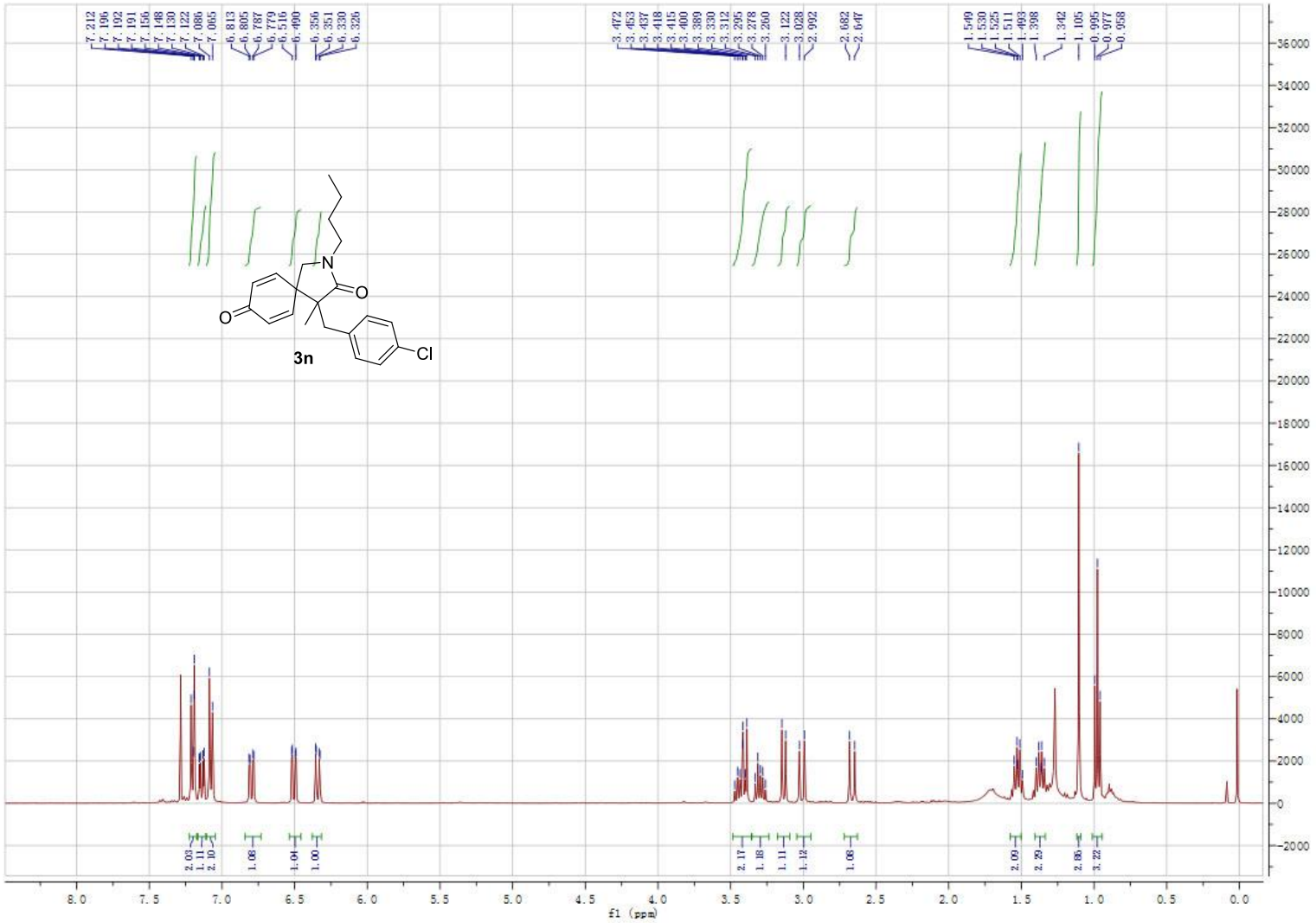
***2-(Tert-butyl)-6,7,9,10-tetrafluoro-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-di0(3m)***



***2-(Tert-butyl)-6,7,9,10-tetrafluoro-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-di0(3m)***




***2-Butyl-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3n)***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6.805
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6.516
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6.356
6.351
6.330
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3.437
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3.260
3.122
3.028
2.992
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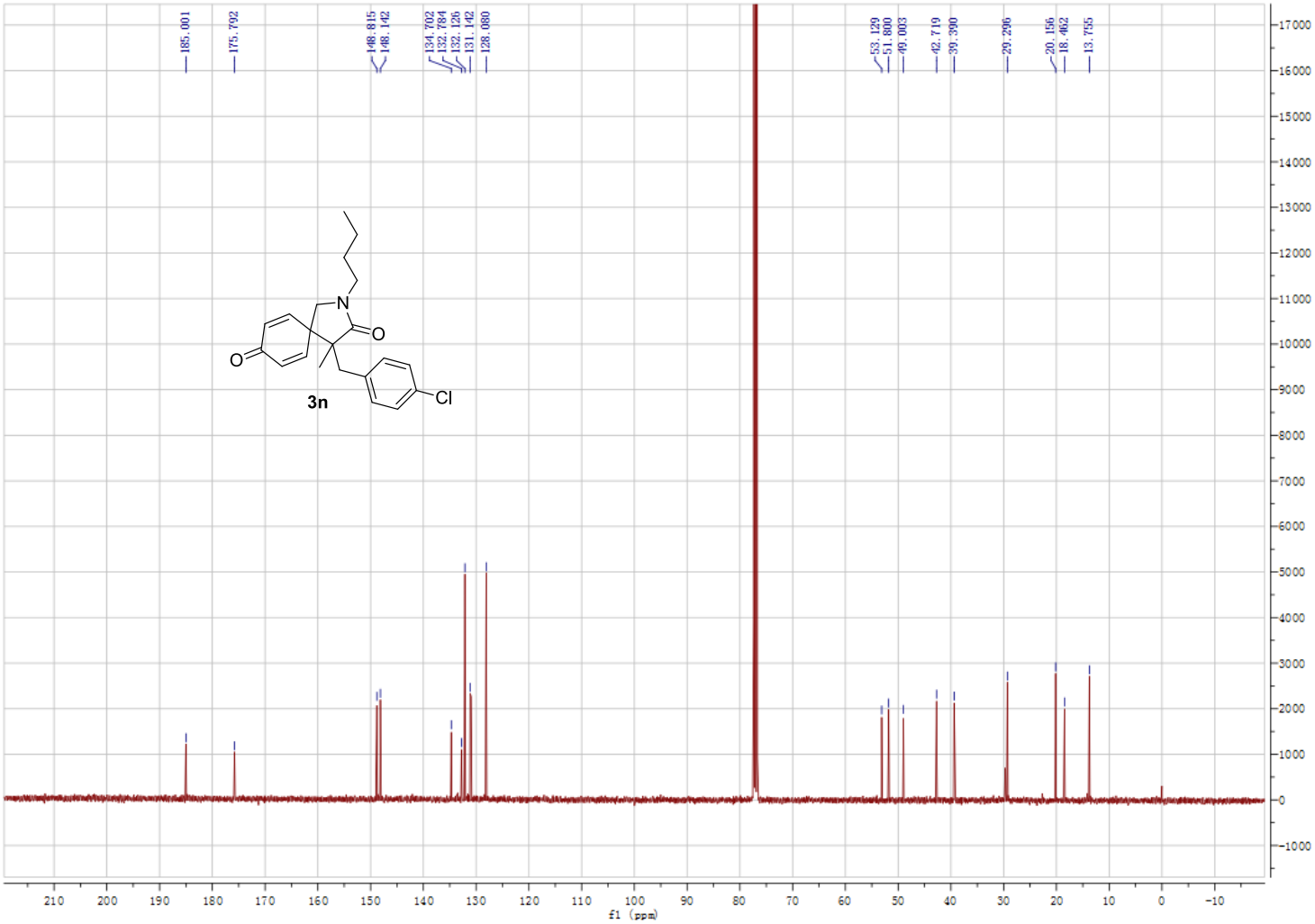
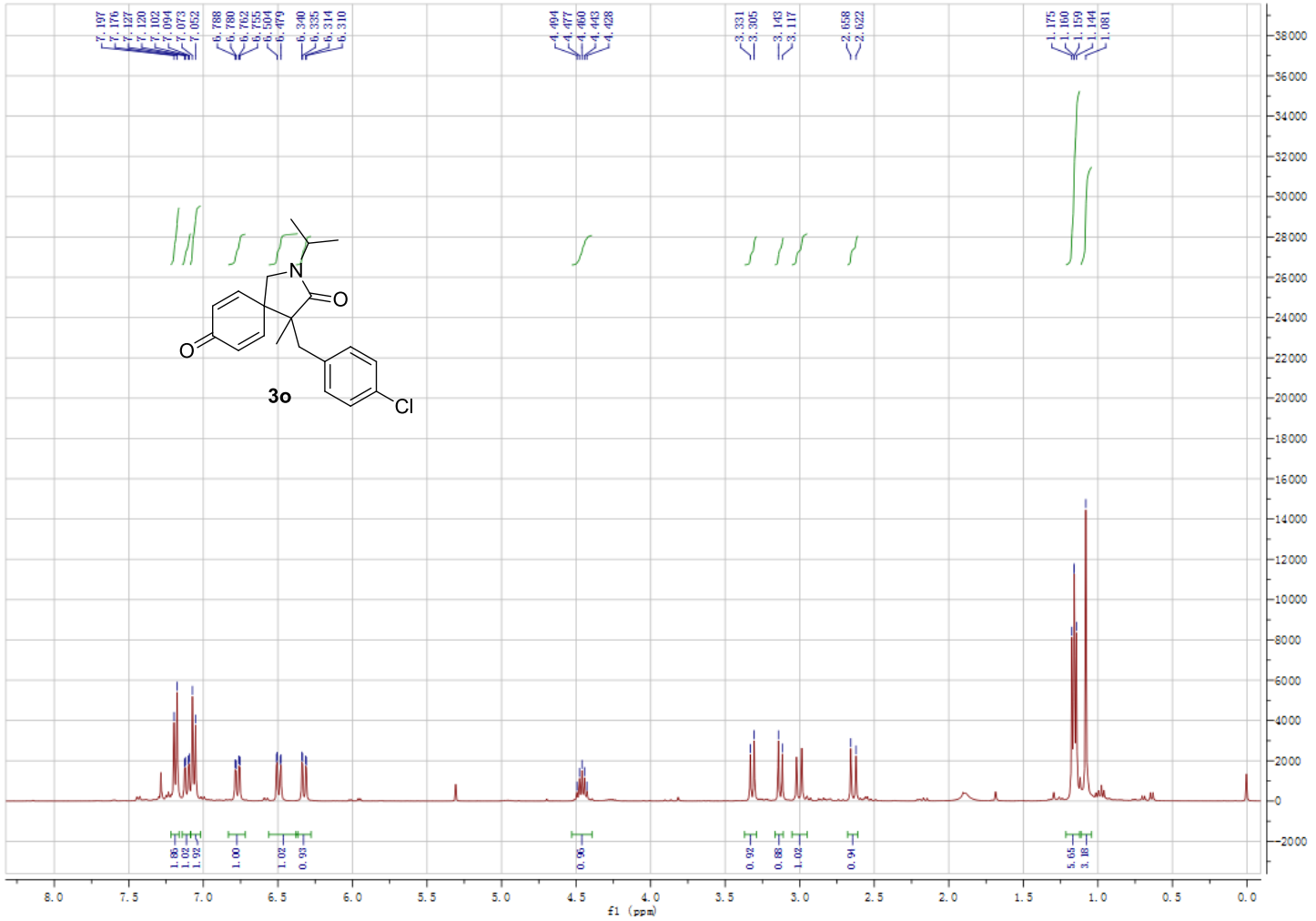
2.647
1.549
1.530
1.525
1.511
1.493
1.398
1.342
1.105
0.995
0.977
0.958
N
O
O
Cl
3n
2.03
1.11
2.10
1.00
1.04
1.00
2.17
1.18
1.11
1.12
1.06
2.09
2.29
2.86
3.22
f1 (ppm)

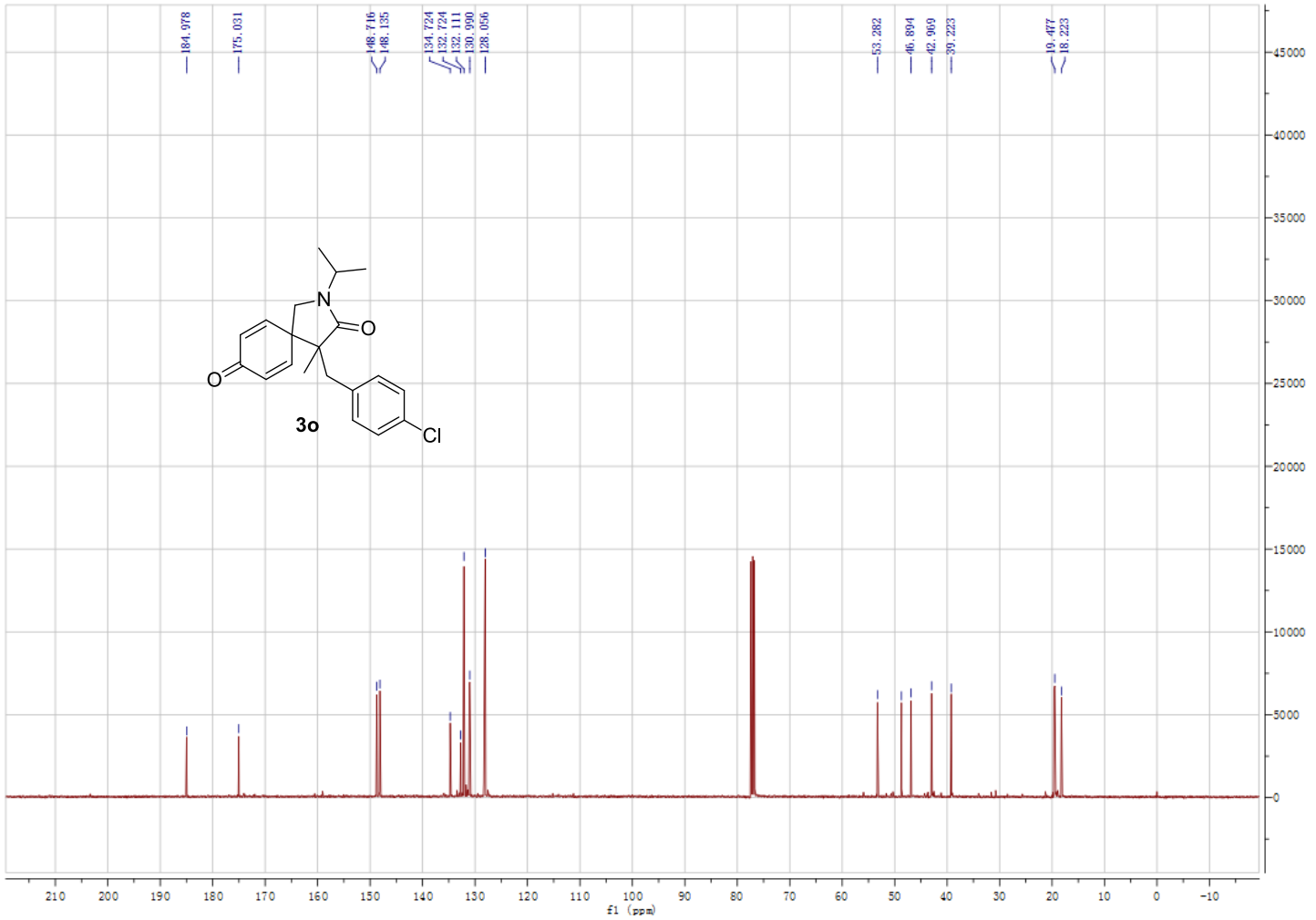
***2-Butyl-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3n)***



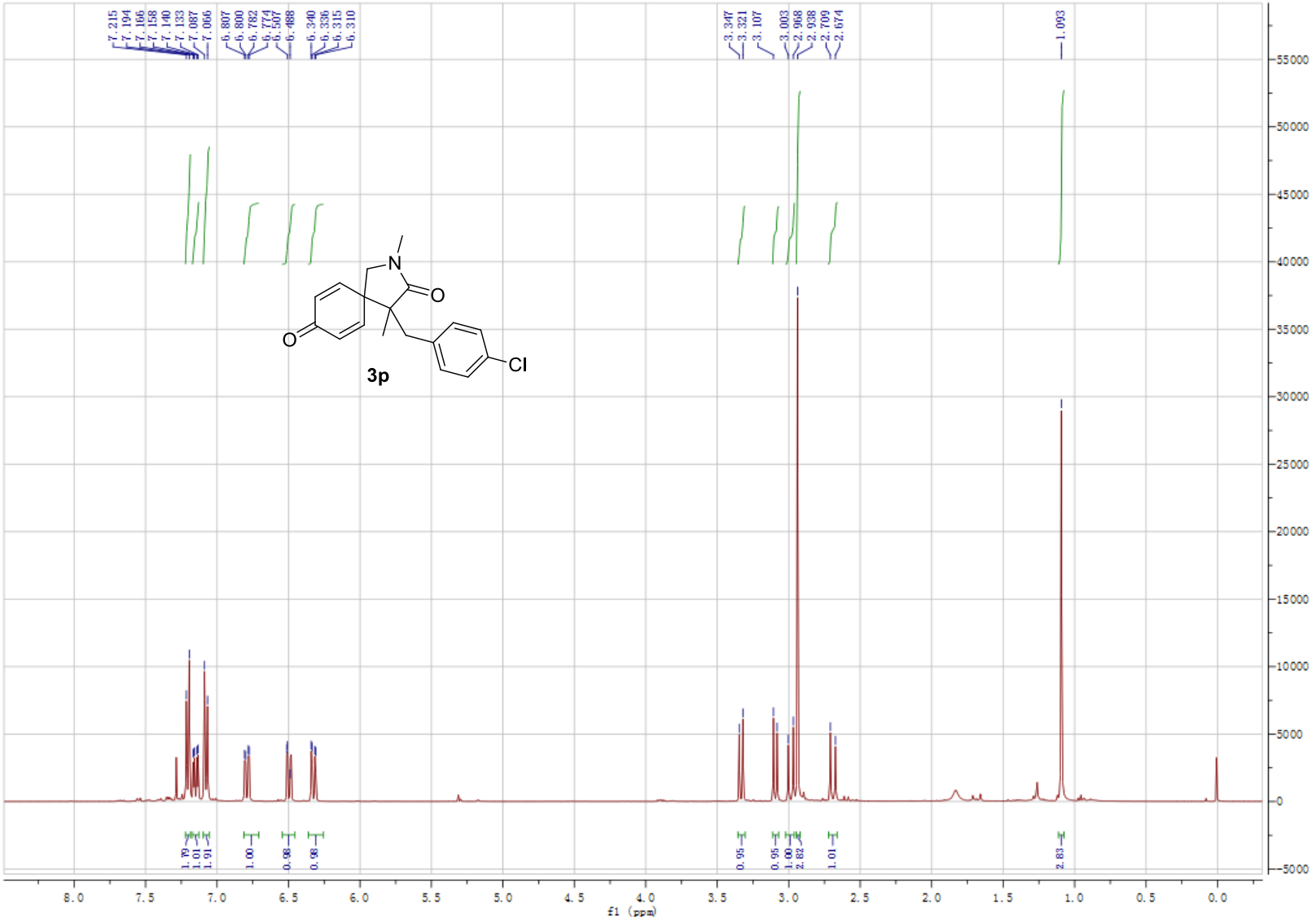
***4-(4-Chlorobenzyl)-2-isopropyl-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3o)***




***4-(4-Chlorobenzyl)-2-isopropyl-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3o)***




***4-(4-Chlorobenzyl)-2,4-dimethyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3p).***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6.340
6.336
6.315
6.310
3.347
3.321
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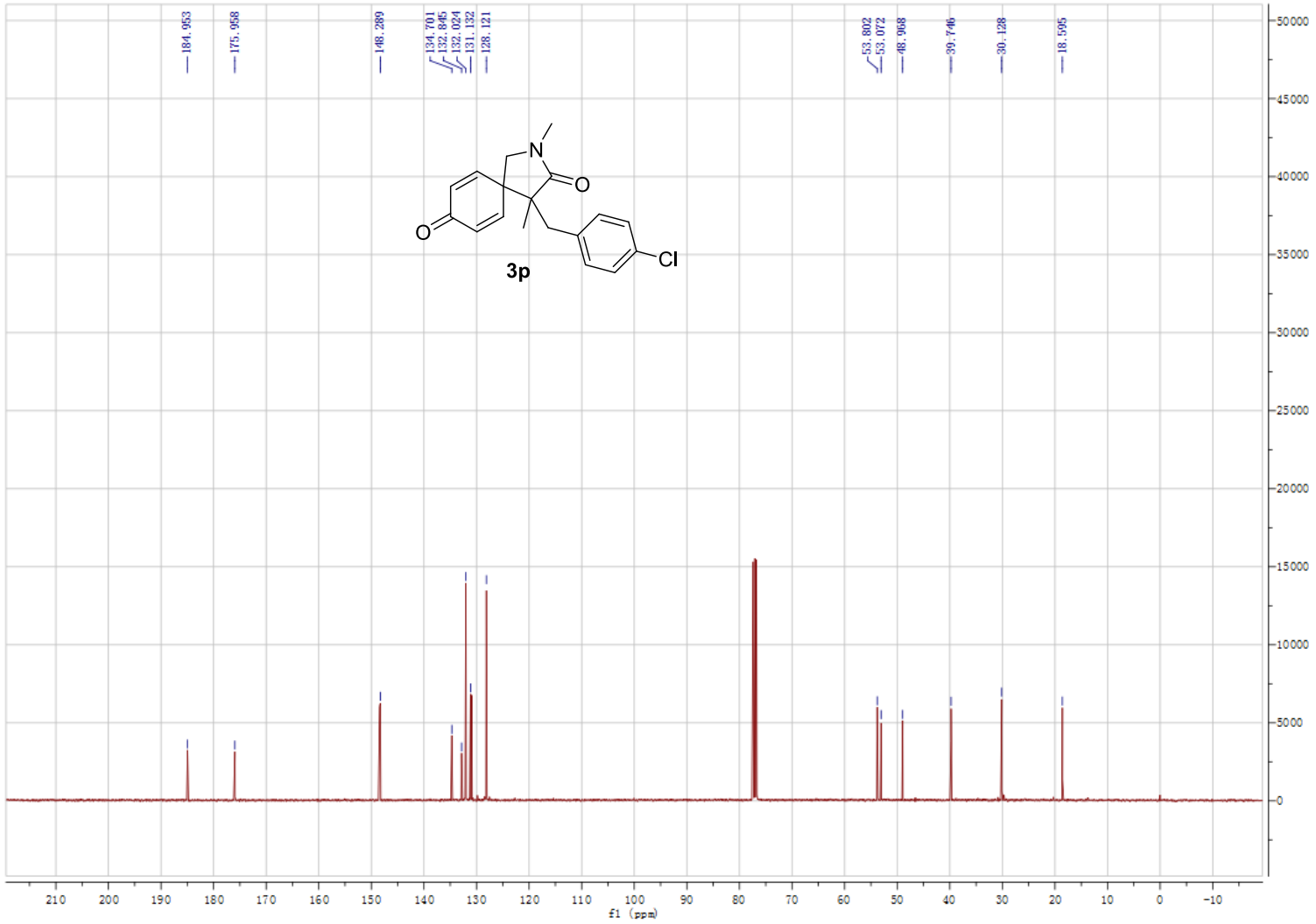
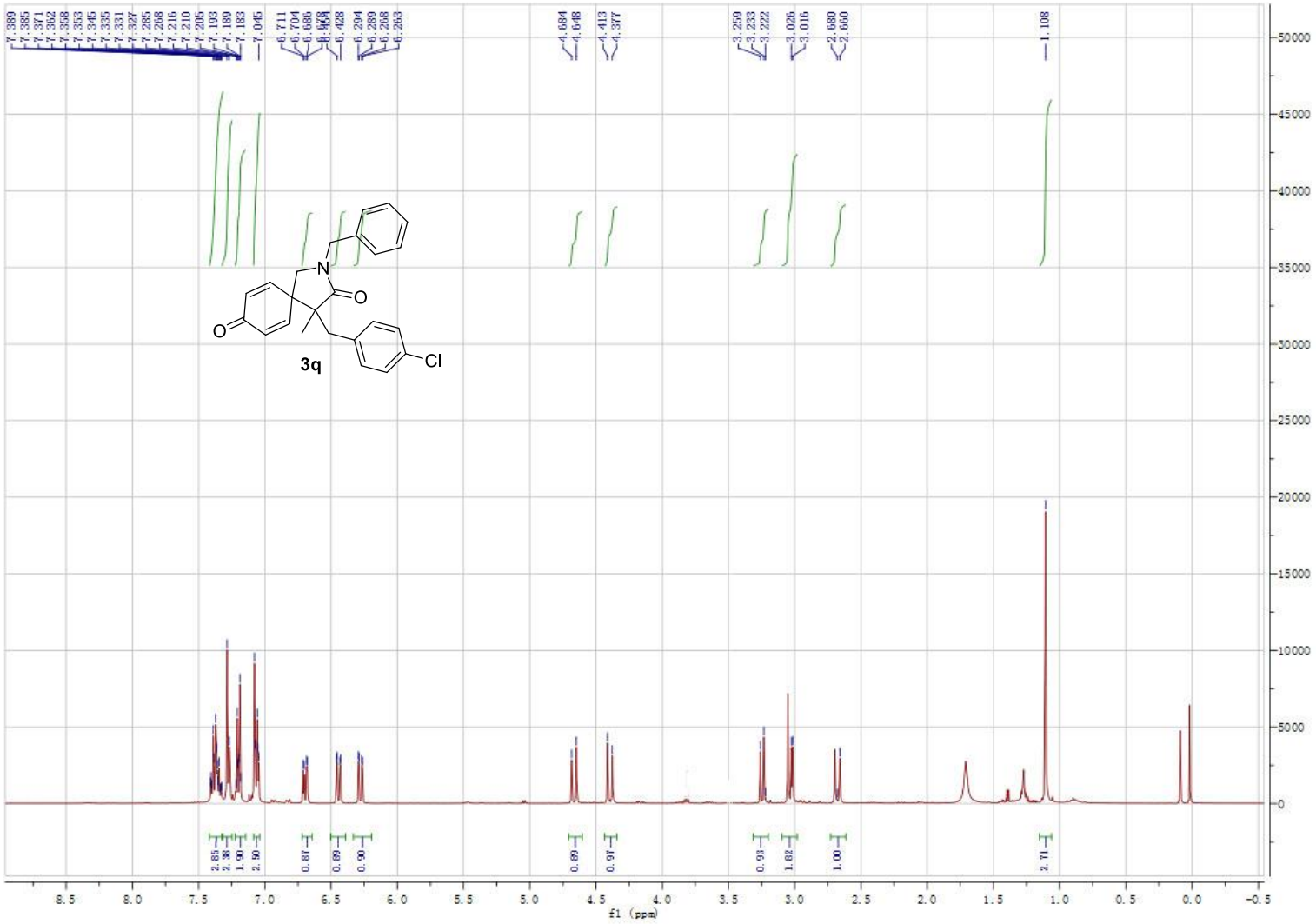
2.709
2.674
1.093
N
O
O
Cl
3p
1.79
1.01
1.91
1.00
0.98
0.98
0.95
0.95
1.00
2.82
1.01
2.83
f1 (ppm)

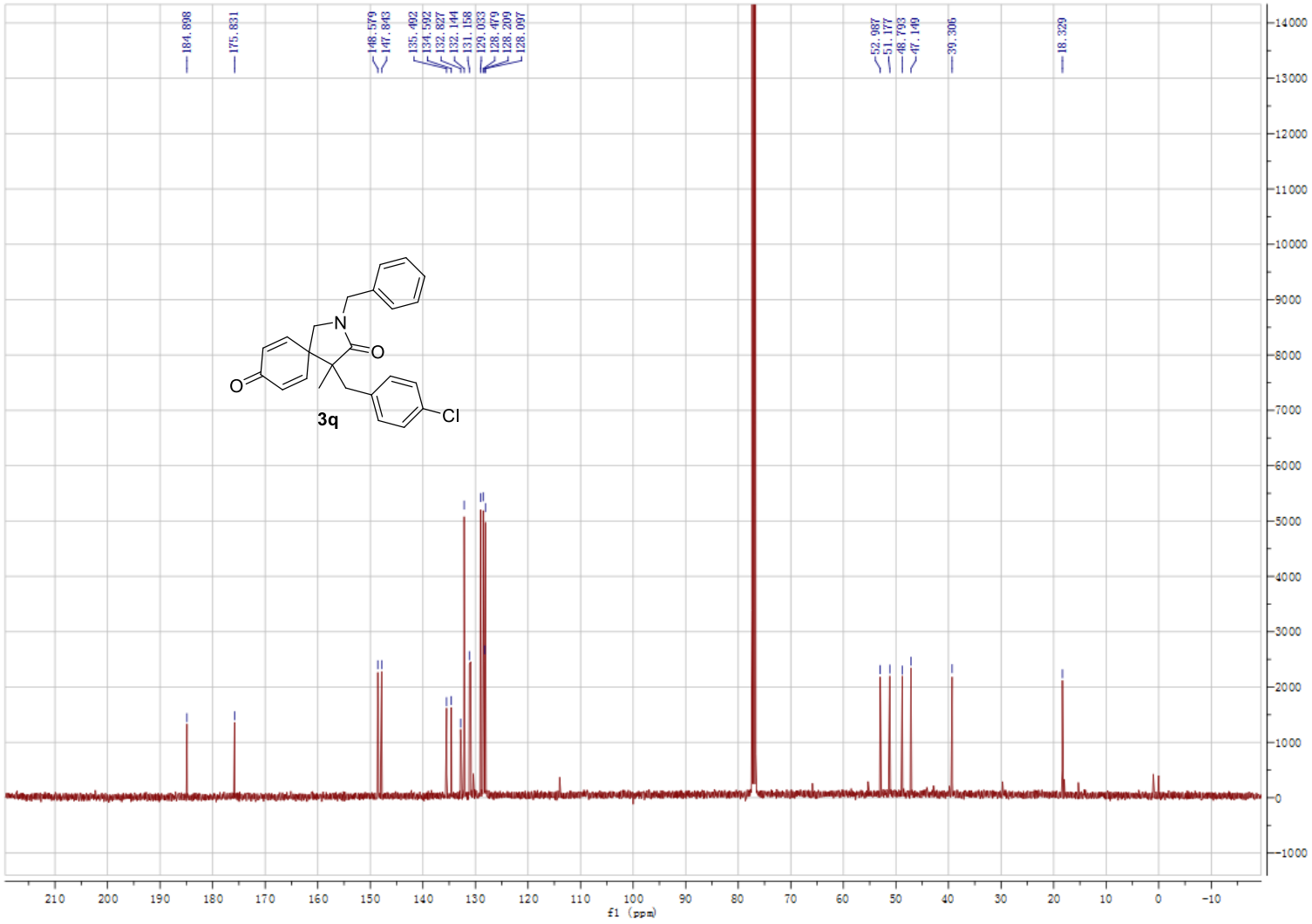
***4-(4-Chlorobenzyl)-2,4-dimethyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3p).***



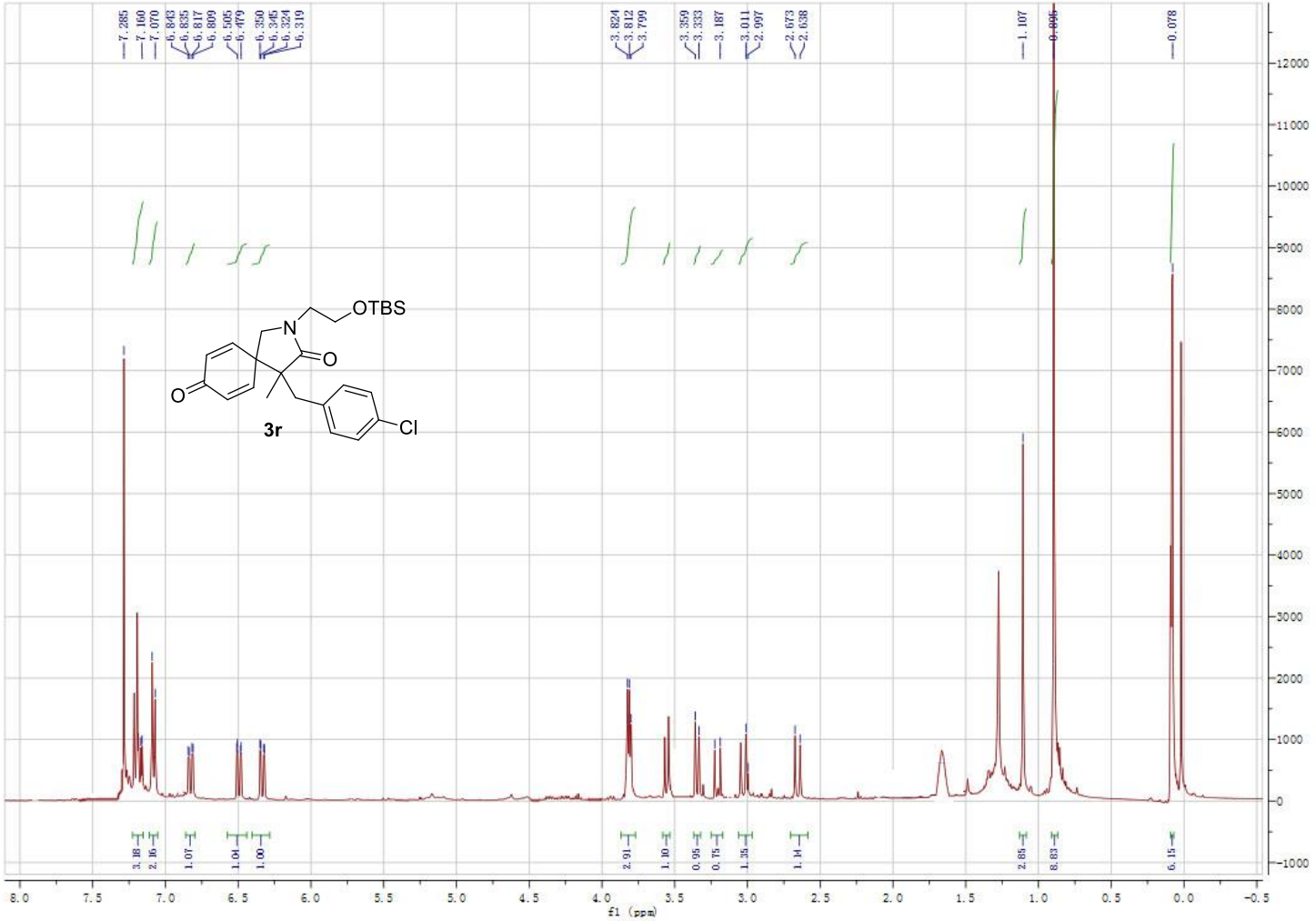
***2-Benzyl-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3q)***



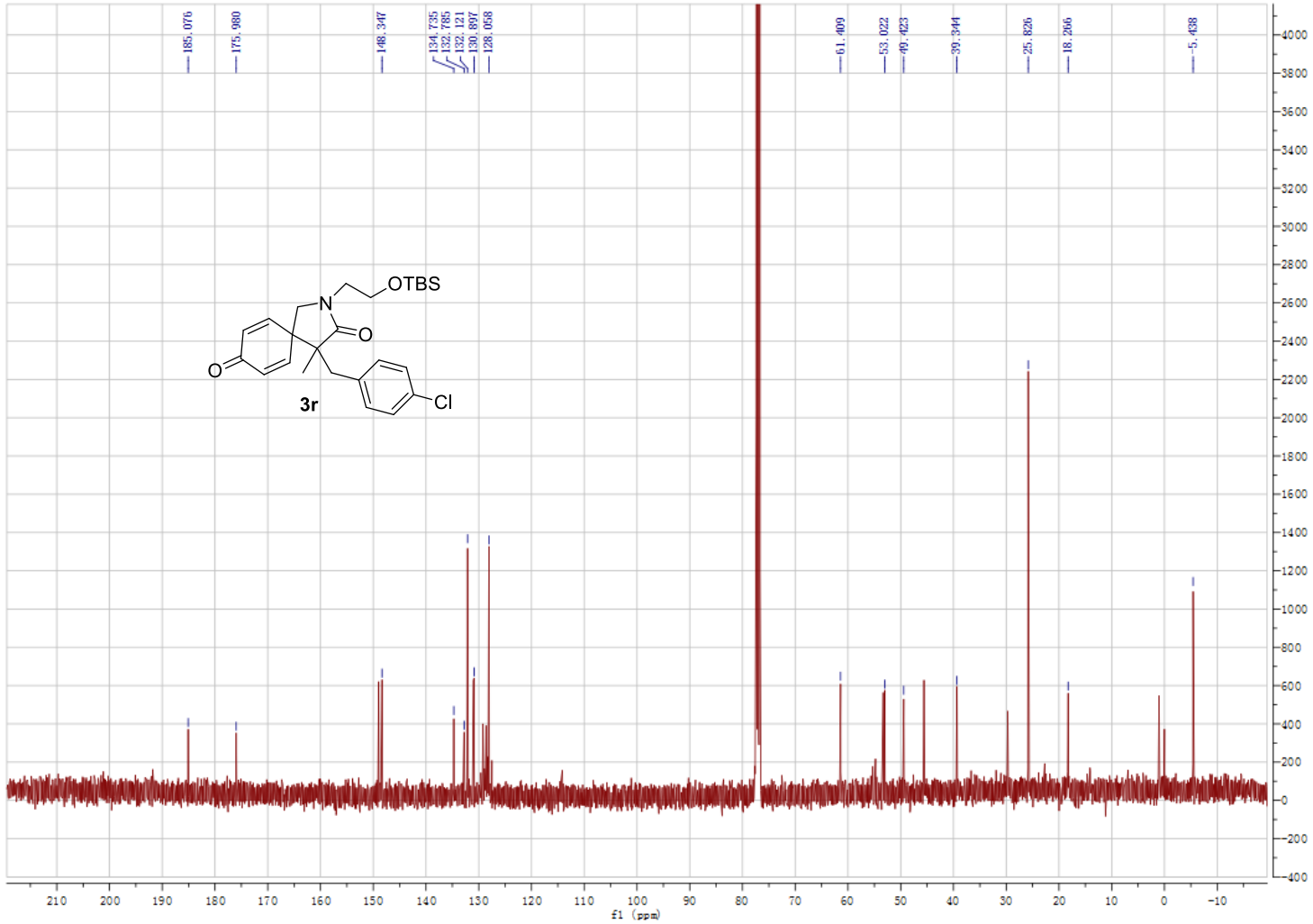
***2-Benzyl-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3q)***



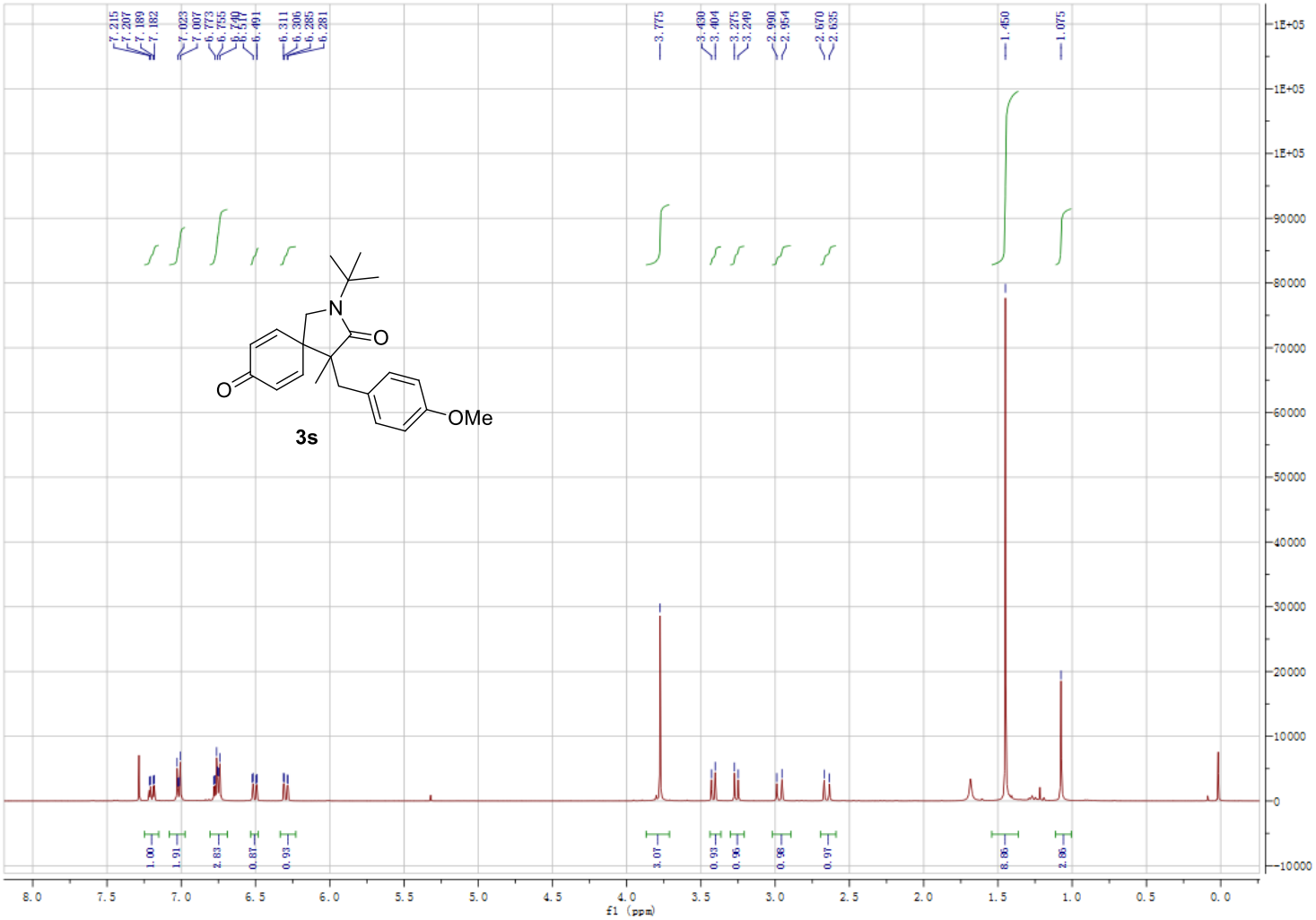
***2-(2-((Tert-butyldimethylsilyl)oxy)ethyl)-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3r)***



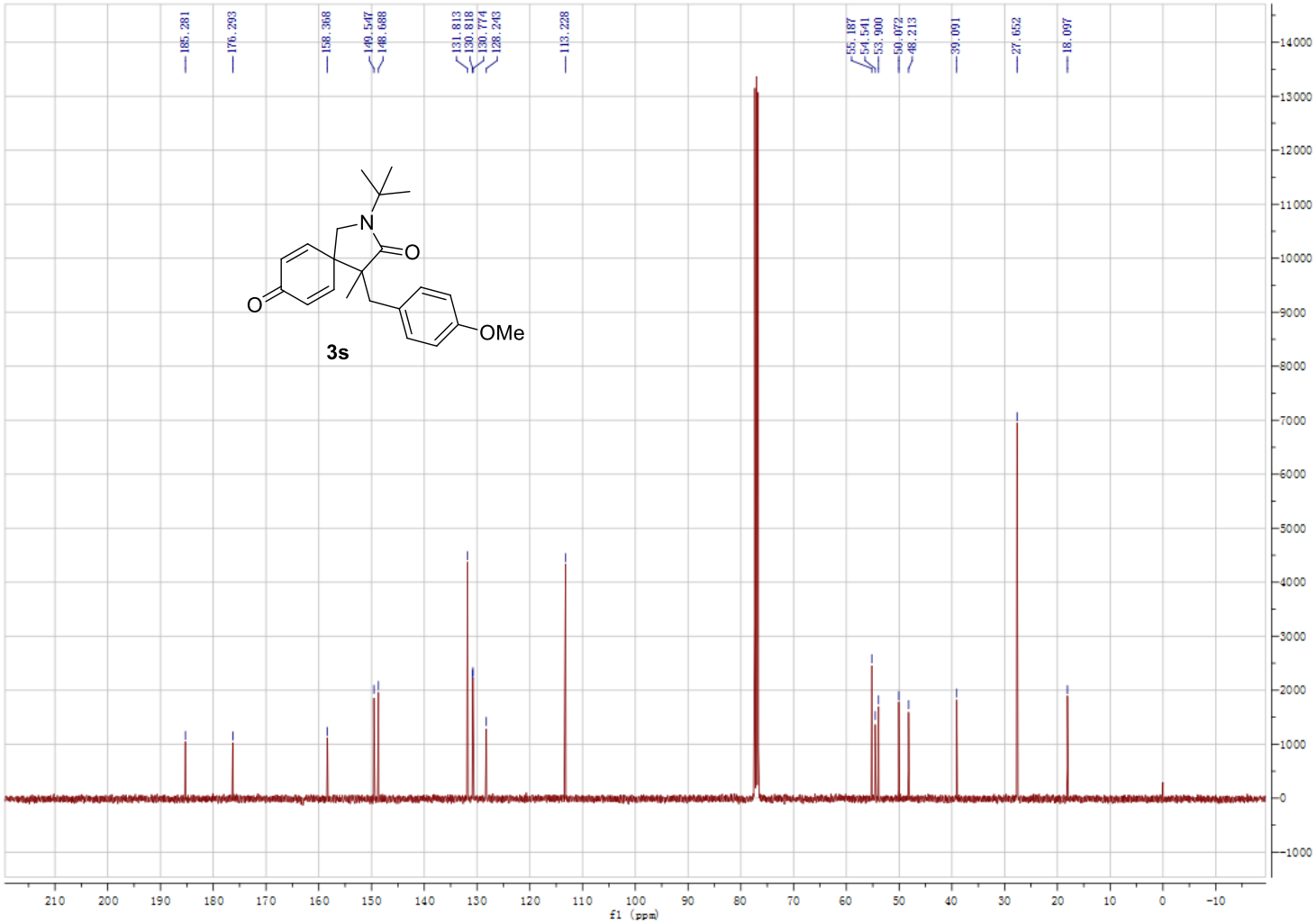
***2-(2-((Tert-butyldimethylsilyl)oxy)ethyl)-4-(4-chlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3r)***



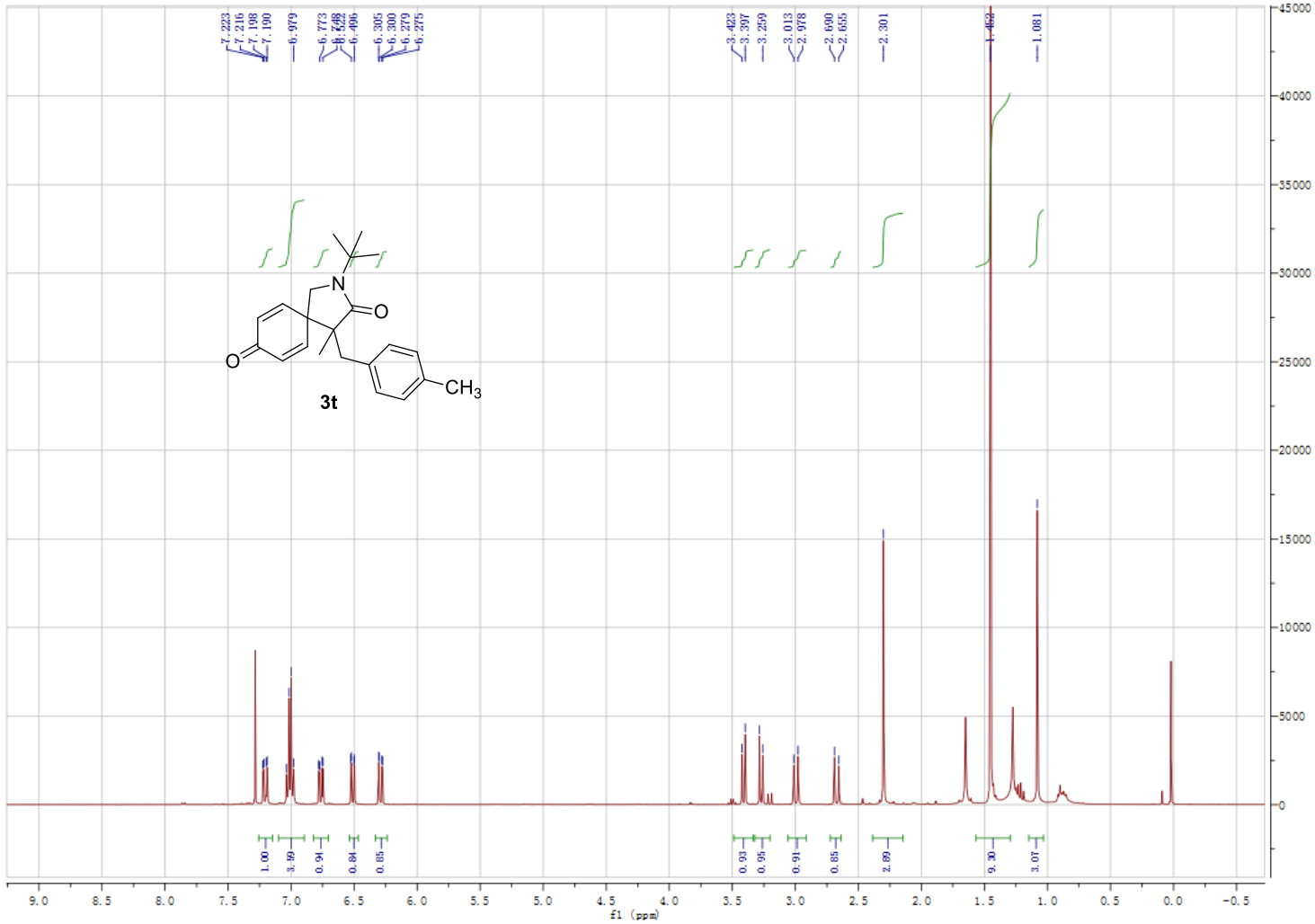
***2-(Tert-butyl)-4-(4-methoxybenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3s)***



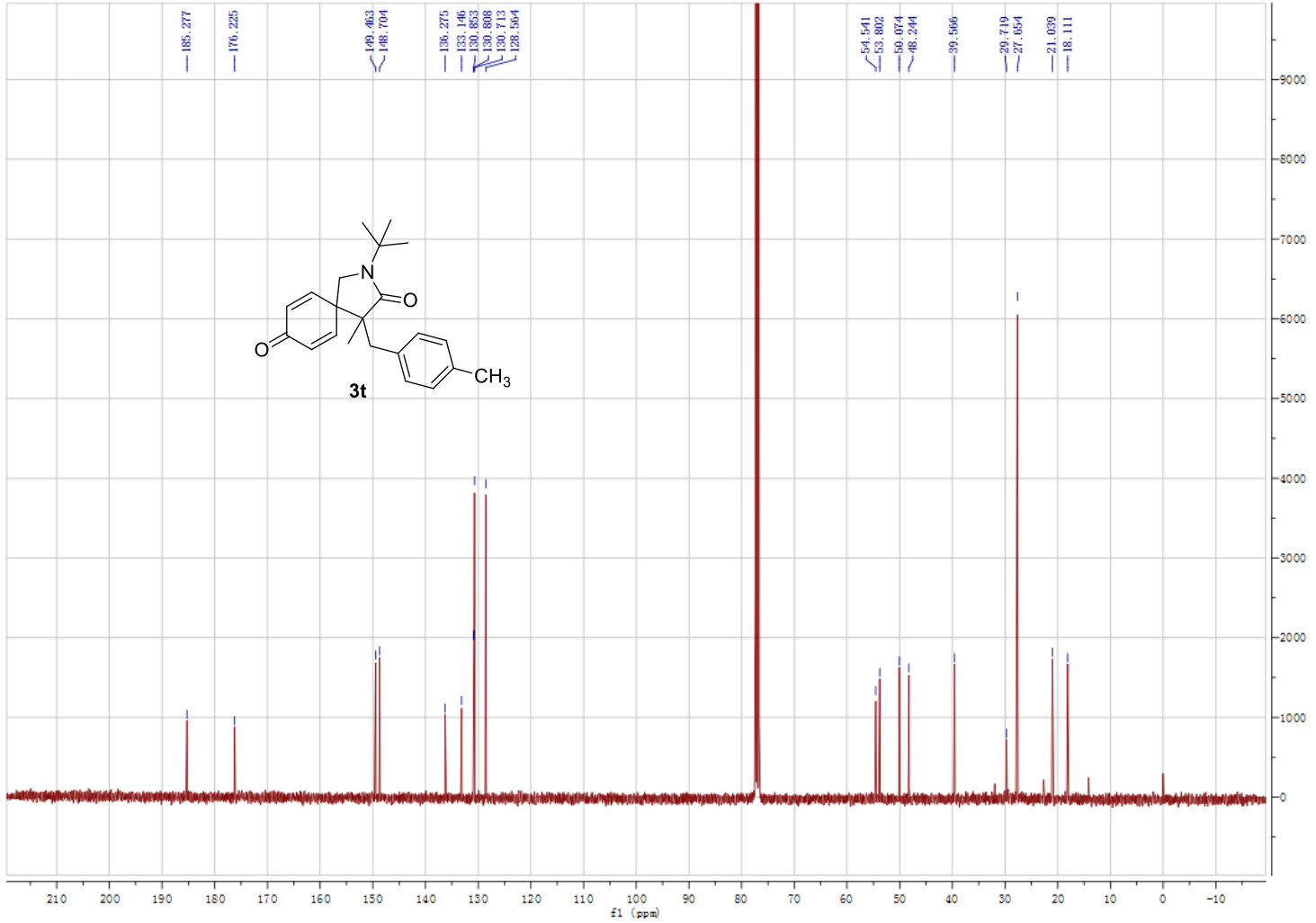
***2-(Tert-butyl)-4-(4-methoxybenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3s)***



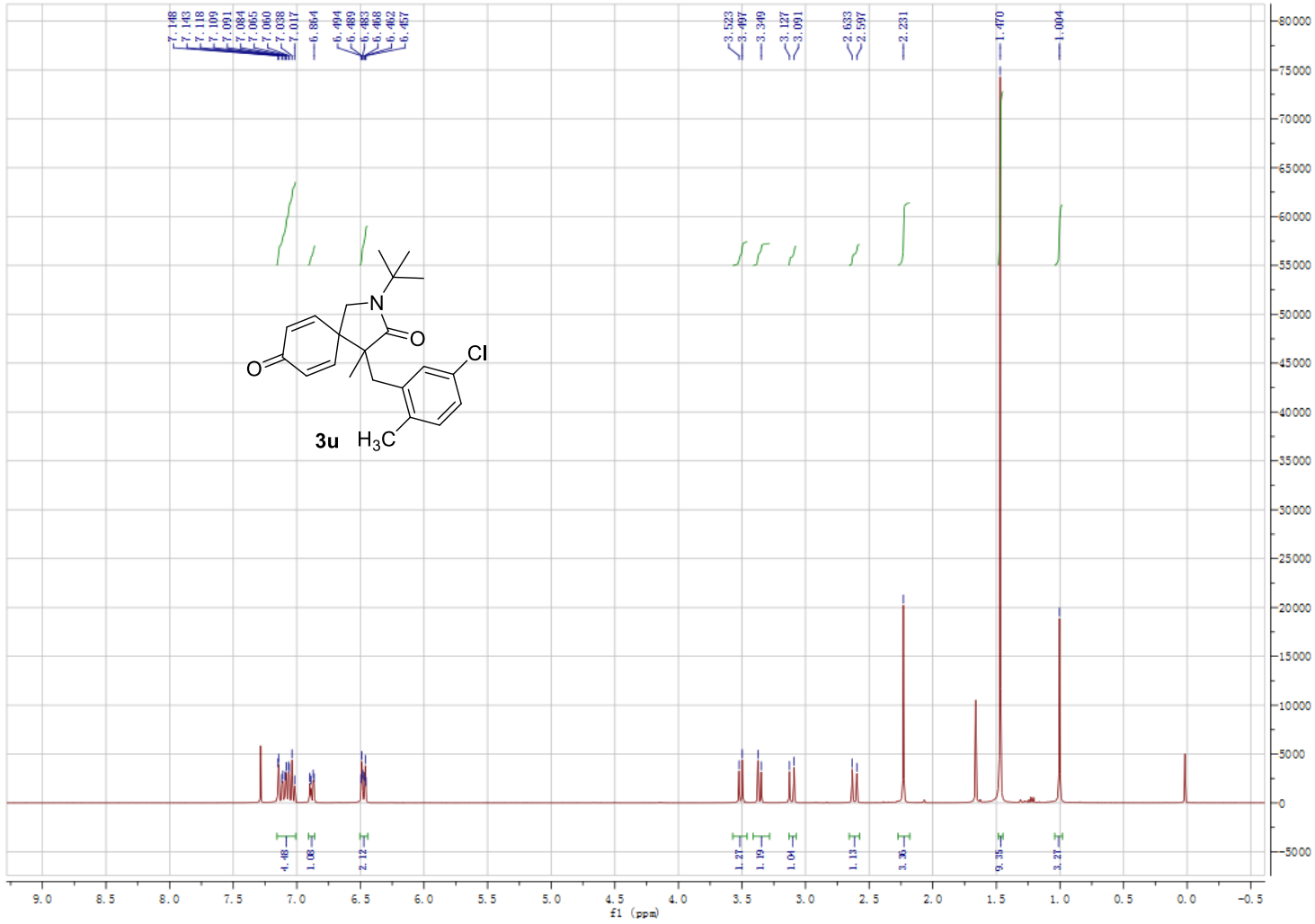
***2-(Tert-butyl)-4-methyl-4-(4-methylbenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3t)***



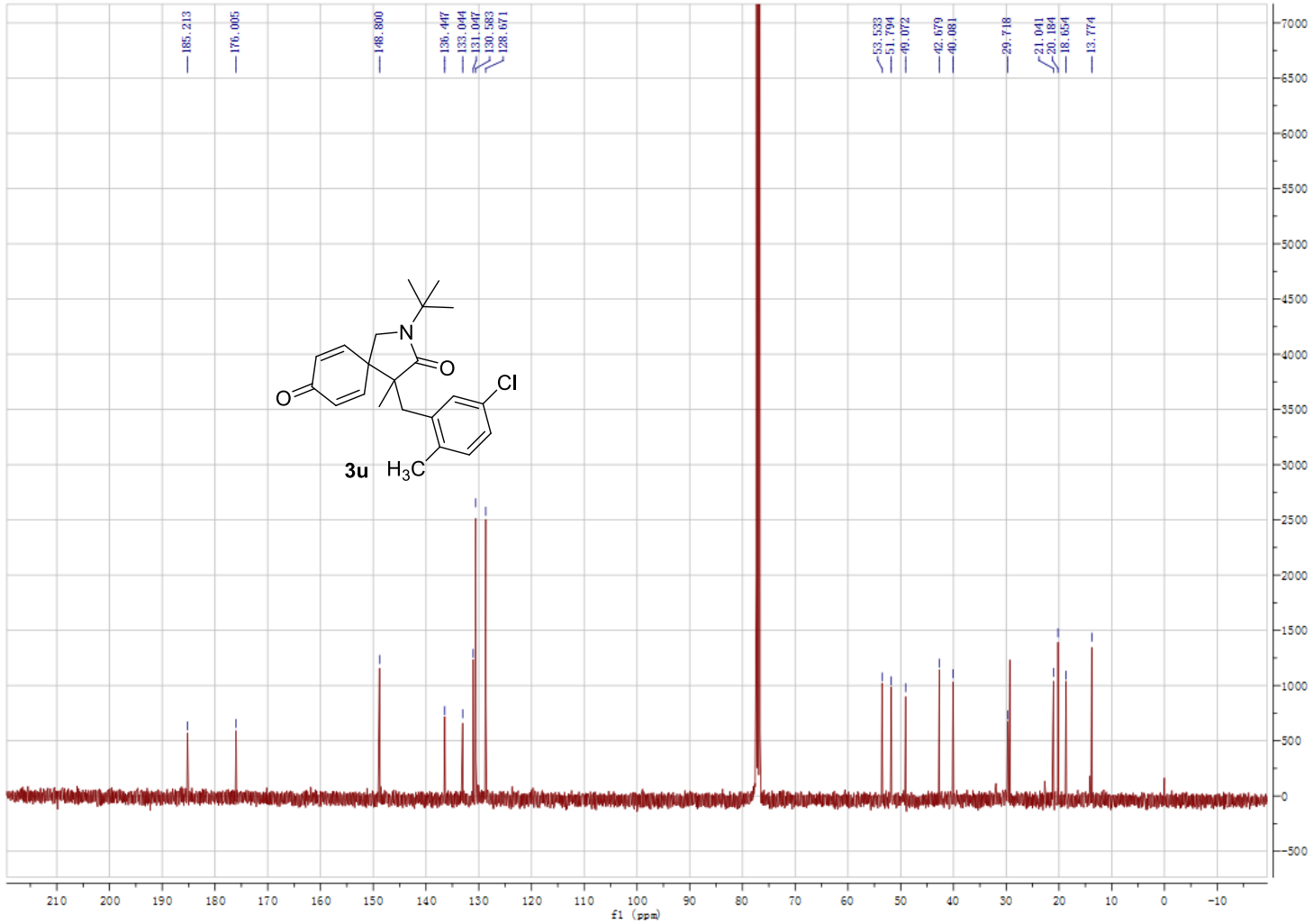
***2-(Tert-butyl)-4-methyl-4-(4-methylbenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3t)***



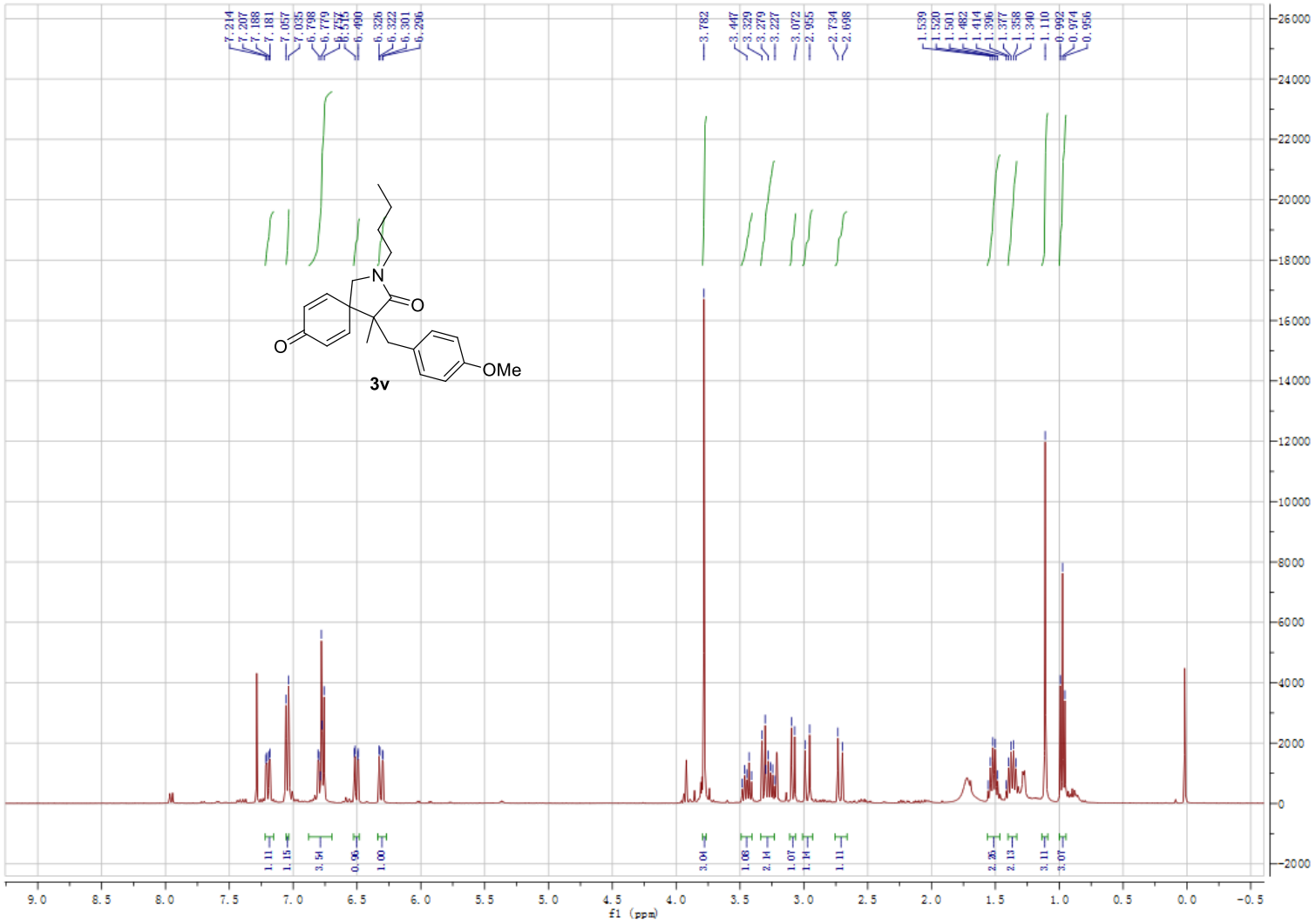
***2-(Tert-butyl)-4-(5-chloro-2-methylbenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3u)***



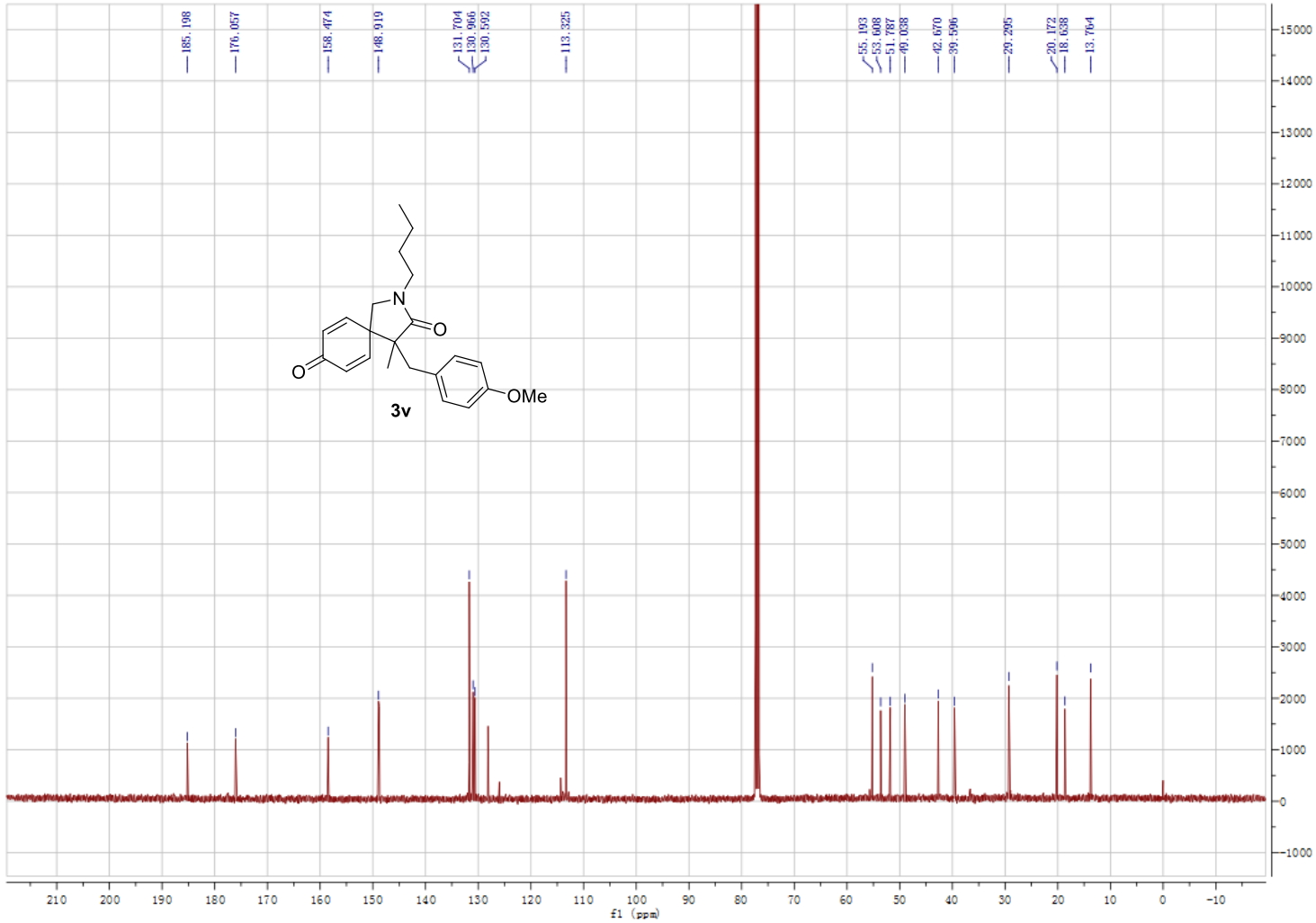
***2-(Tert-butyl)-4-(5-chloro-2-methylbenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3u)***



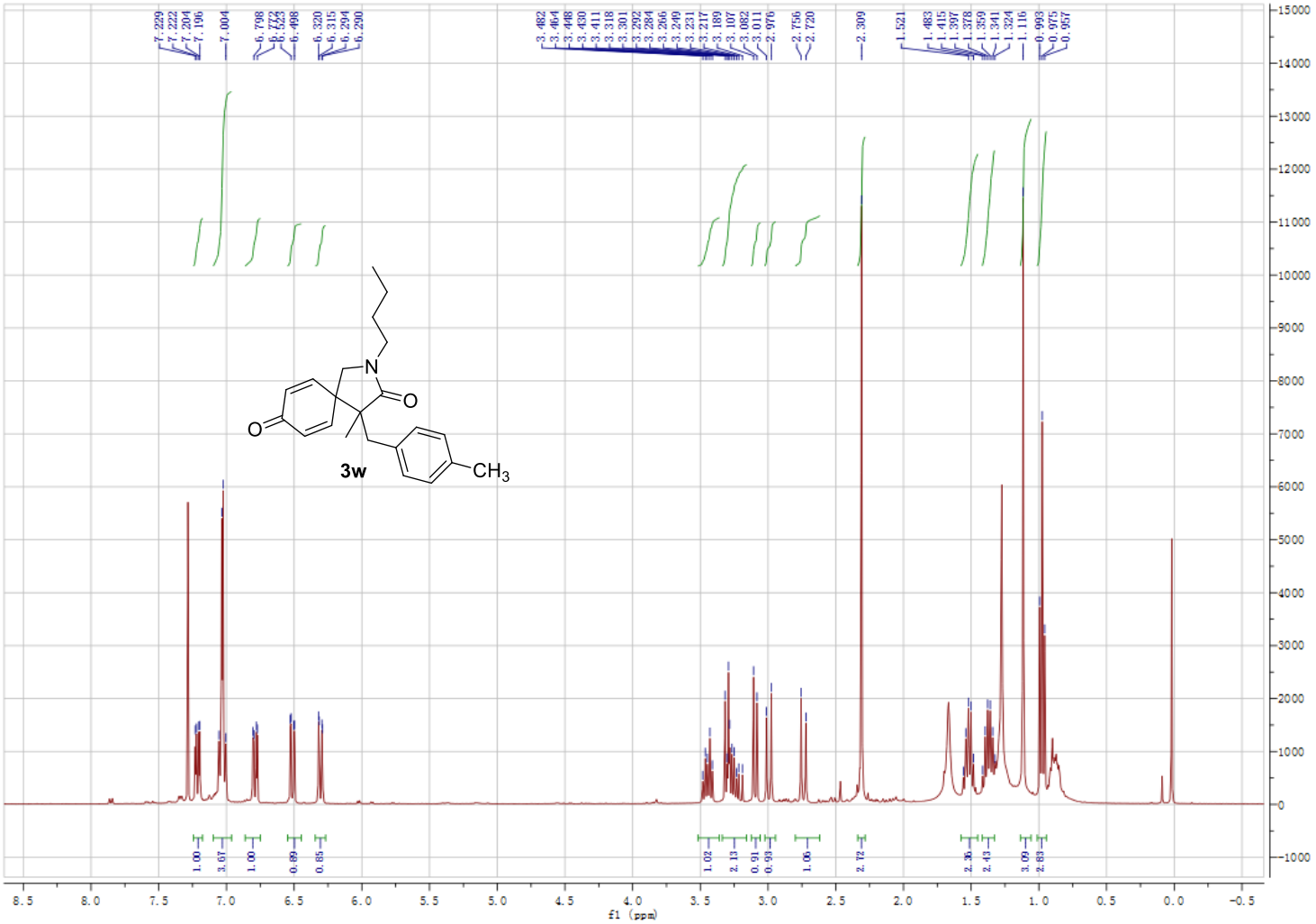
***2-Butyl-4-(4-methoxybenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3v)***



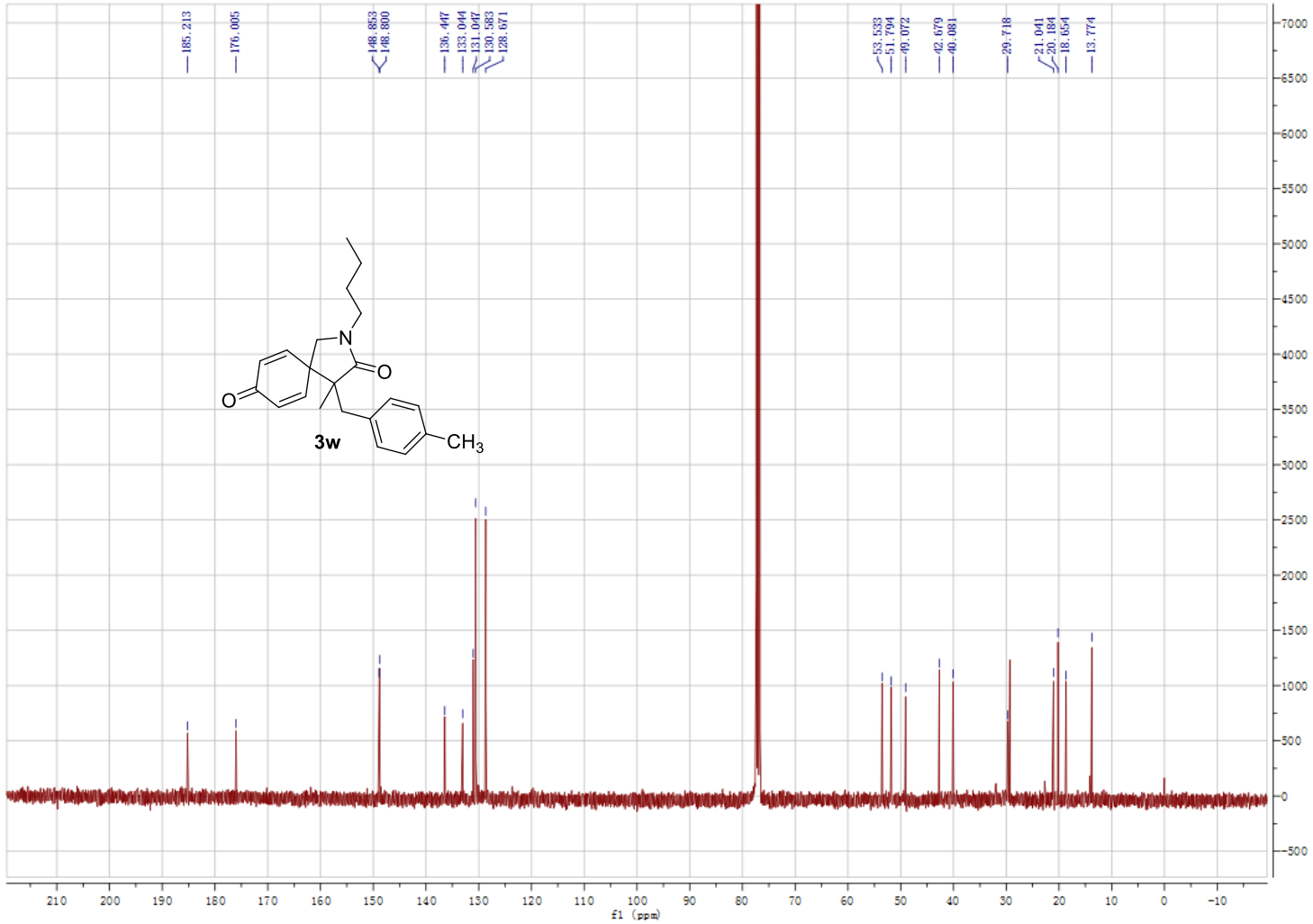
***2-Butyl-4-(4-methoxybenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3v)***



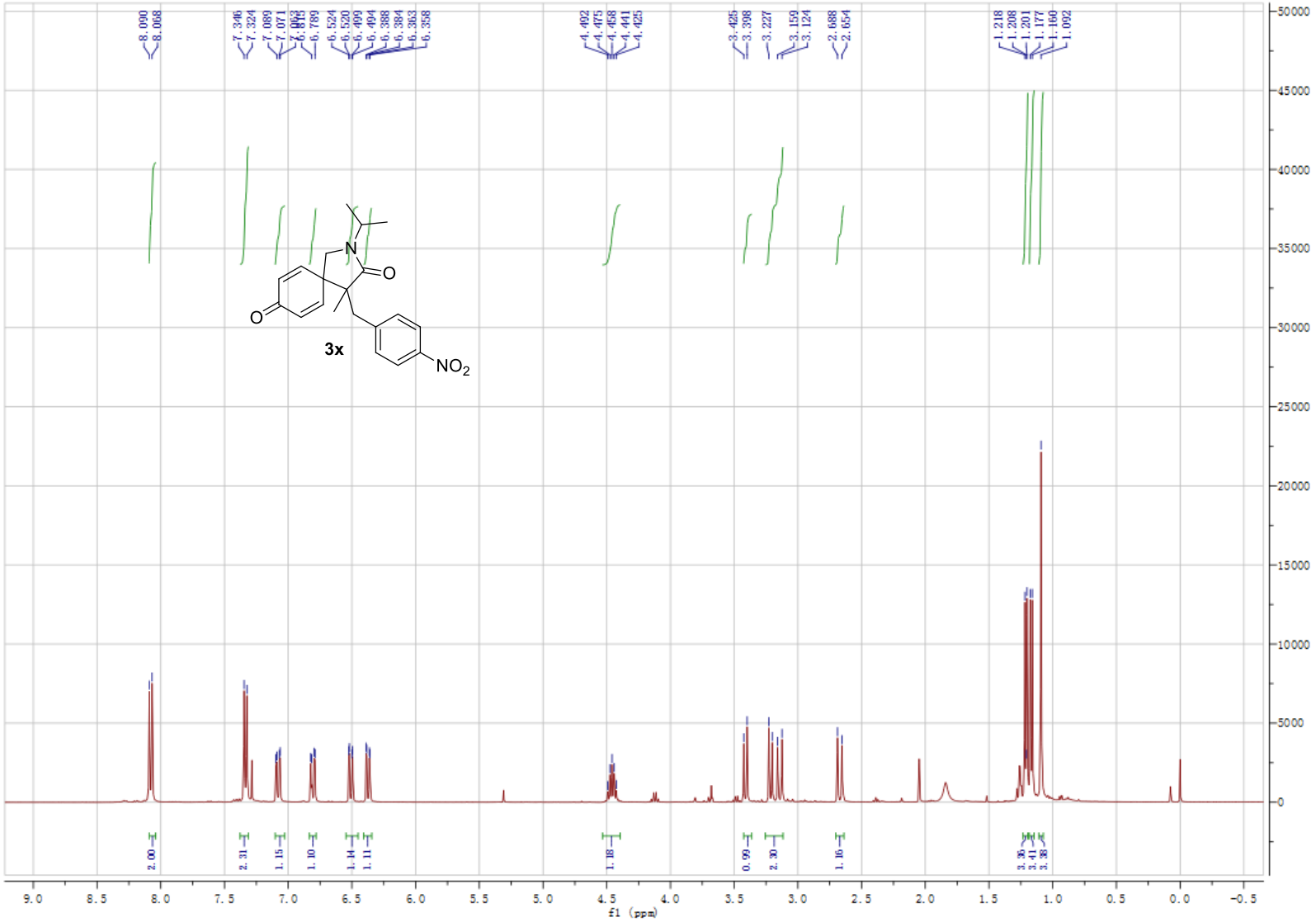
***2-Butyl-4-methyl-4-(4-methylbenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3w)***



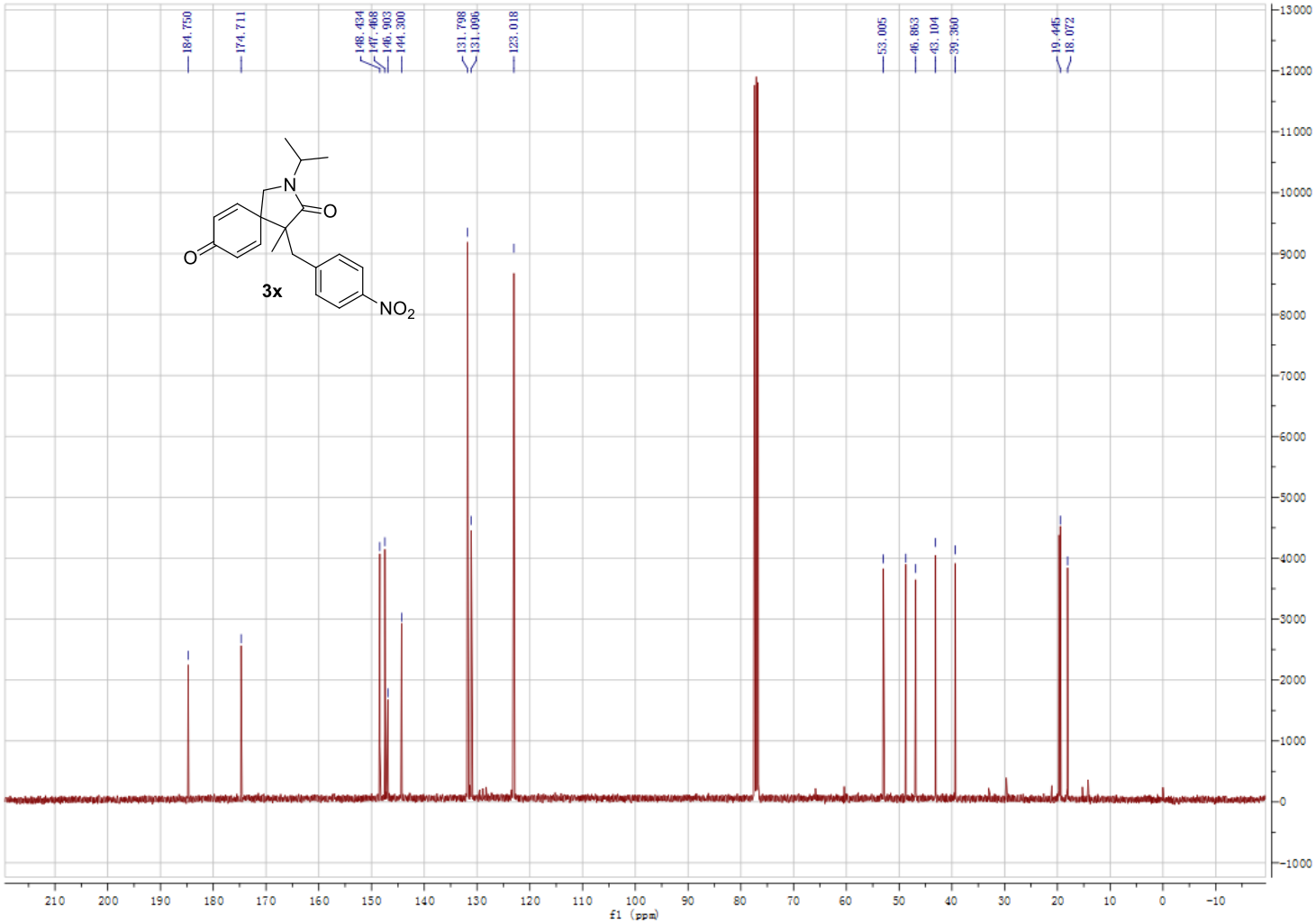
***2-Butyl-4-methyl-4-(4-methylbenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3w)***



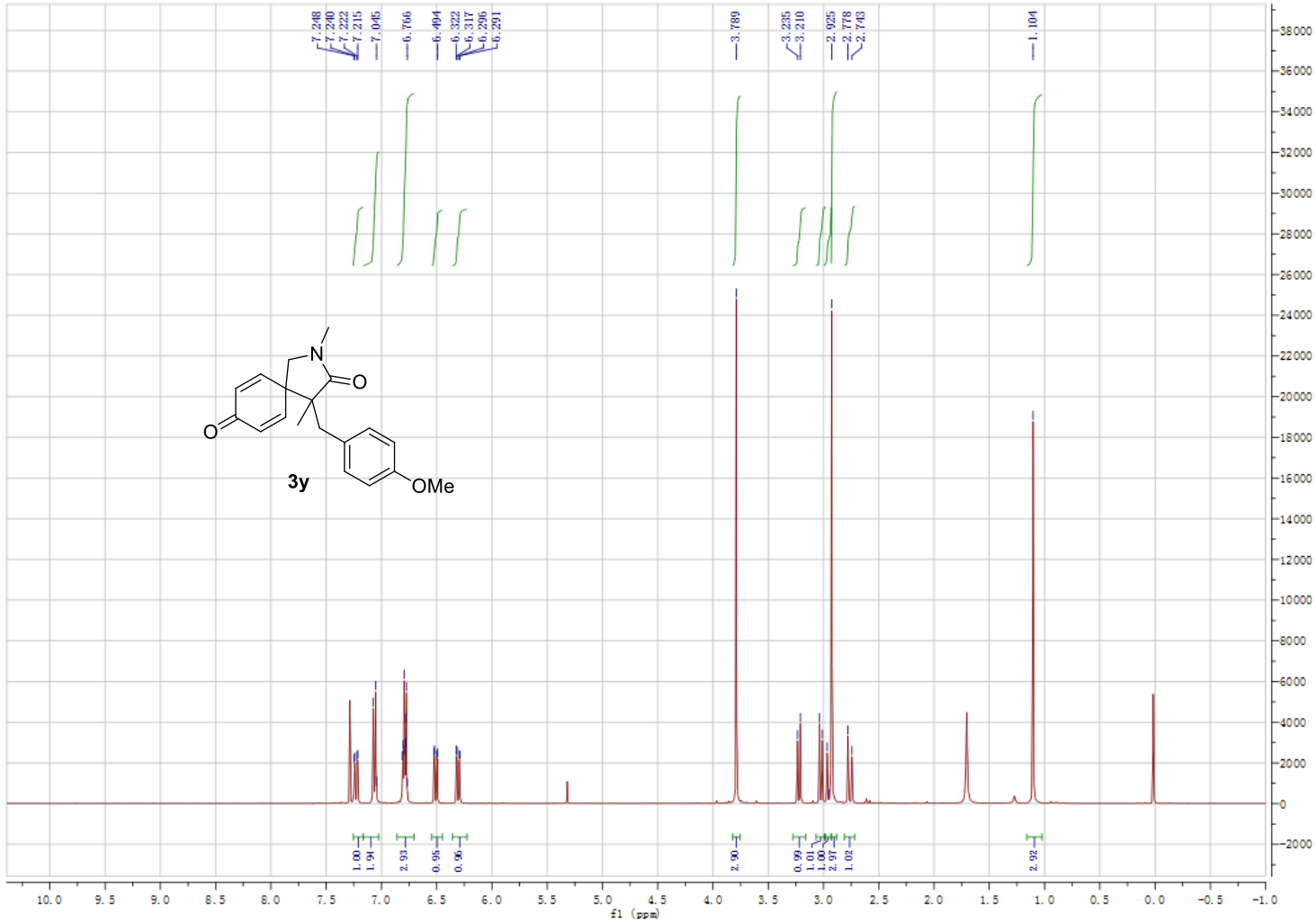
***2-Isopropyl-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3x)***



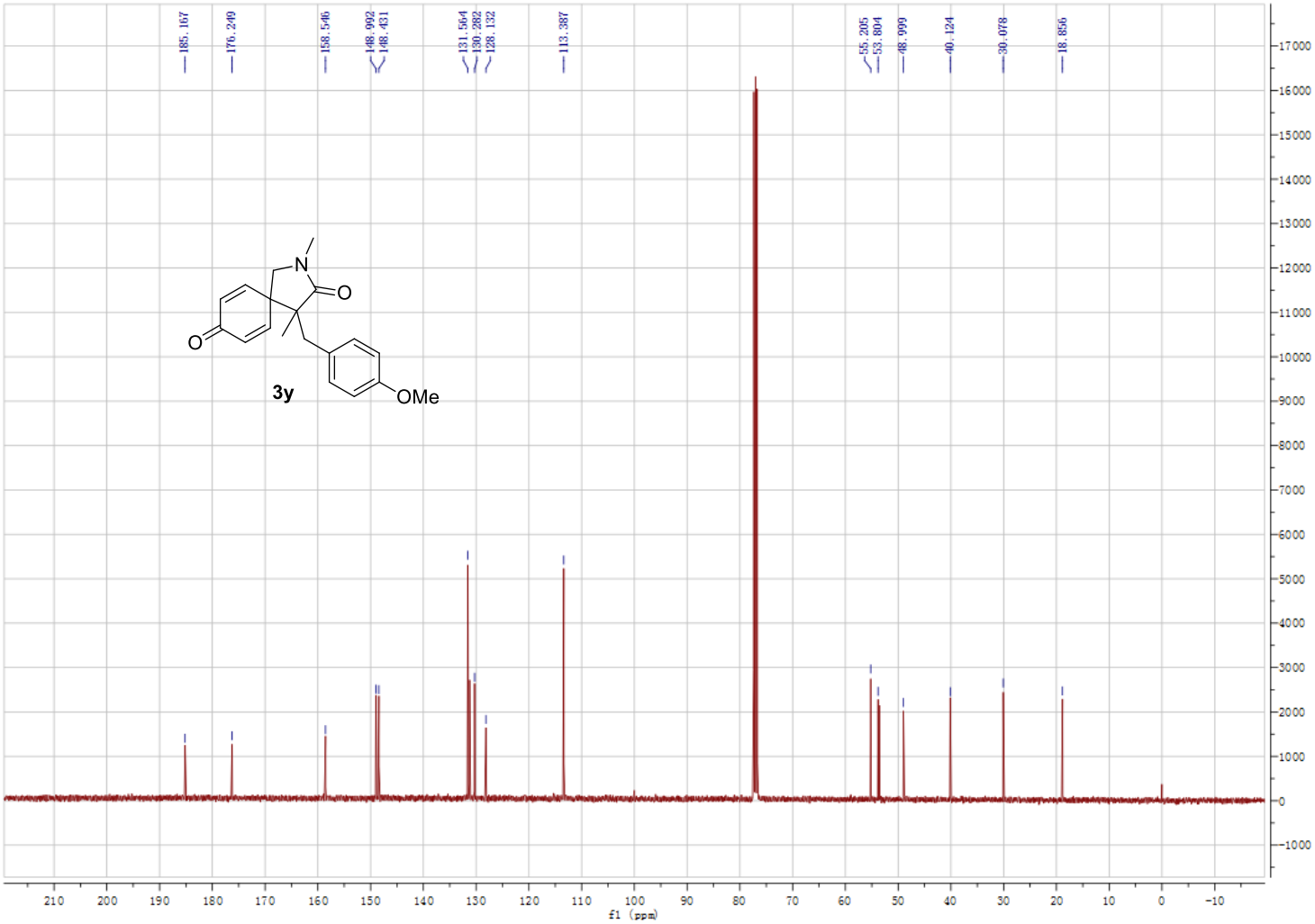
***2-Isopropyl-4-methyl-4-(4-nitrobenzyl)-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3x)***



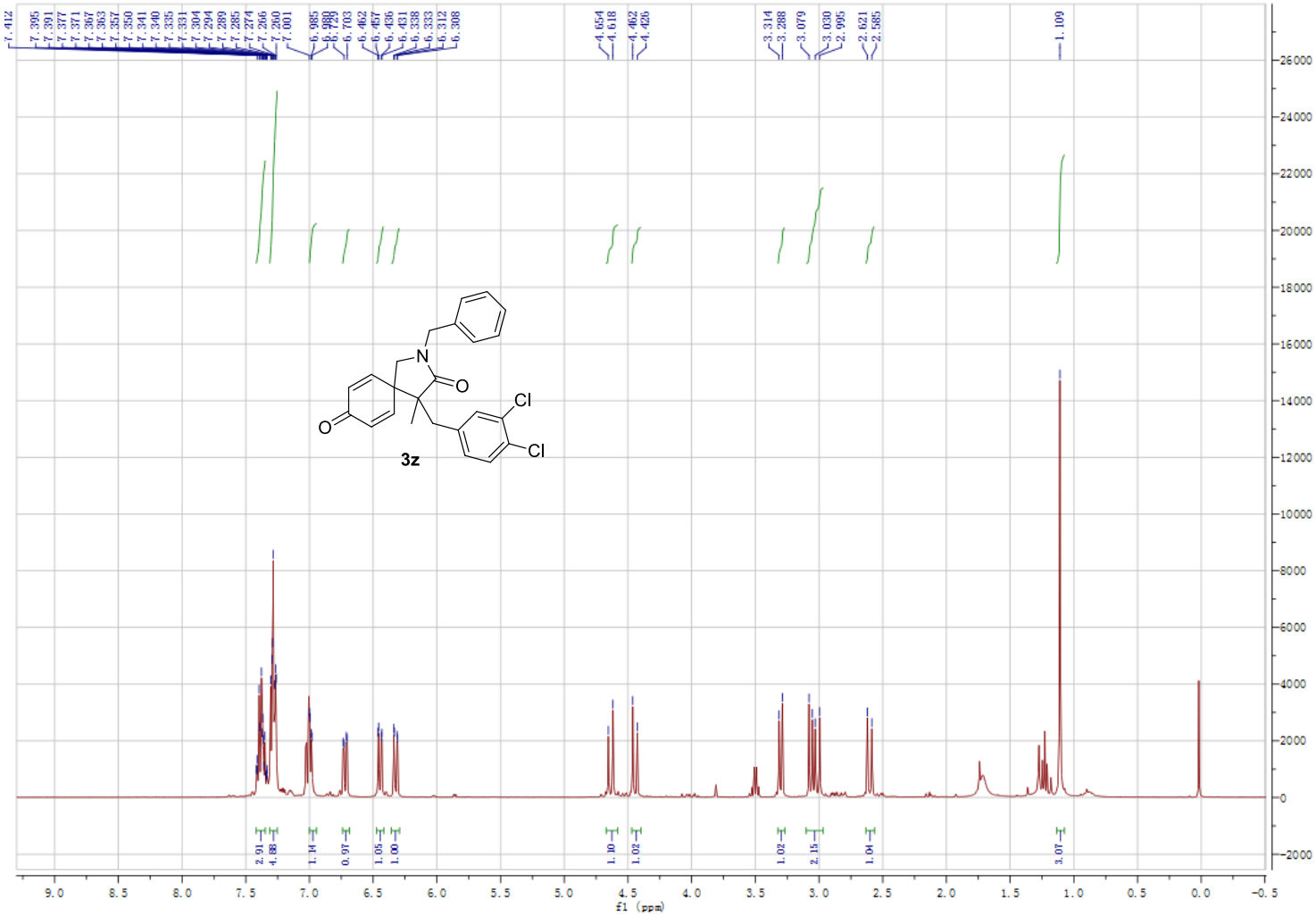
***4-(4-Methoxybenzyl)-2,4-dimethyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3y)***



***4-(4-Methoxybenzyl)-2,4-dimethyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3y)***



***2-Benzyl-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3z)***



***2-Benzyl-4-(3,4-dichlorobenzyl)-4-methyl-2-azaspiro[4.5]deca-6,9-diene-3,8-dione(3z)***