Supplementary Information

Visible-light-enabled spirocyclization of alkynes leading to 3-sulfonyl and 3-sulfenyl azaspiro[4,5]trienones

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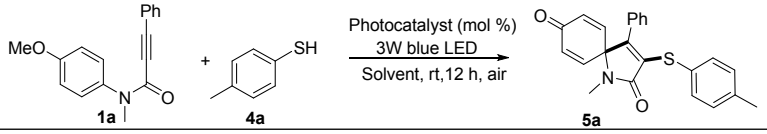
1. General information

All commercially available reagent grade chemicals were purchased from Aldrich, Acros, Alfa Aesar and Beijing Ouhe Chemical Company and used as received without further purification unless otherwise stated. All solvents were dried according to standard procedures. ^{1}H NMR and ^{13}C NMR were recorded in $CDCl_3$ on a Bruker Avance III 400 spectrometer with TMS as internal standard (500 MHz ^{1}H, 125 MHz ^{13}C) at room temperature, the chemical shifts (δ) were expressed in ppm and J values were given in Hz. The following abbreviations are used to indicate the multiplicity: singlet (s), doublet (d), triplet (t), quartet (q), doublet of doublets (dd), doublet of triplets (dt), and multiplet (m). All first order splitting patterns were assigned on the basis of the appearance of the multiplet. Splitting patterns that could not be easily interpreted were designated as multiplet (m). Mass analyses and HRMS were obtained on a Finnigan-LCQDECA mass spectrometer and a Bruker Daltonics Bio-TOF-Q mass spectrometer by the ESI method, respectively. Column chromatography was performed on silica gel (200-300 mesh). There is 3.0 cm distance between the reactor and LEDs.

2. Optimization of conditions for the synthesis of 3-sulfenyl azaspiro[4,5]trienone 5a[a] (Table S1).



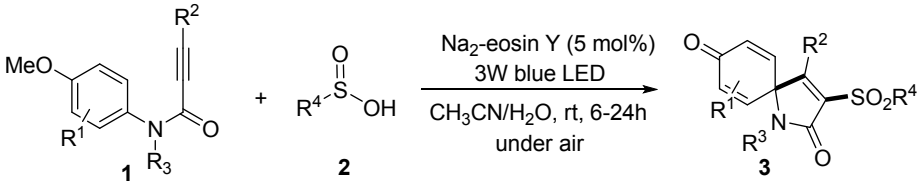
Entry	Photocatalyst (mol%)	Solvent	Yield(%)[b]
1	Eosin Y (5)	CH_3CN	66
2	Eosin Y (2)	CH_3CN	74
3	Eosin Y (1)	CH_3CN	87
4	Eosin Y (1)	1,4-dioxane	59
5	Eosin Y (1)	Toluene	32
6	Eosin Y (1)	$CHCl_3$	58
7	Eosin Y (1)	Acetone	54
8	Eosin Y (1)	DCE	58
9	Eosin Y (1)	DMSO	25
10	Eosin Y (1)	DME	14
11	Eosin Y (1)	THF	15
12	Eosin Y (1)	CH_3CN/H_2O	52
13	Eosin Y (1)	H_2O	14
14	Eosin B (1)	CH_3CN	24
15	Rose Bengal (1)	CH_3CN	42
16	Rhodamine B (1)	CH_3CN	46

17	Acridine Red (1)	CH_3CN	33
18	Na_2-Eosin Y (1)	CH_3CN	46
19	$Ru(bpy)_3Cl_2$(1)	CH_3CN	65
20	--	CH_3CN	0
21	Eosin Y (1)	CH_3CN	0[c]

[a] Reaction conditions: **1a** (0.125 mmol), **4a** (0.25 mmol), catalyst (1-5 mol%), Solvent 2 mL, at room temperature, 12h, under air. DME: 1,2-dimethoxyethane; DCE: 1,2-dichloroethane; THF: tetrahydrofuran; [b] Isolated yields based on **1a**. [c] Without visible-light irradiation.

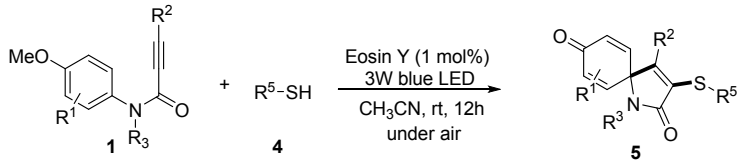
3. General procedure

3.1 The procedure for visible-light-induced difunctionalization of alkynes with sulfinic acids leading to 3-sulfonyl azaspiro[4,5]trienones.



To a solution of sulfinic acid **2** (0.375 mmol) and Na_2-eosin Y (0.00625mmol, 5 mol%) in CH_3CN/H_2O (v_1/v_2=1/1) 2 mL was added N-(*p*-methoxyaryl)-propiolamide **1** (0.125 mmol). The reaction vessel was allowed to stir at room temperature under the irradiation of 3W blue LED lamps for 6-24h. After the reaction, the resulting mixture was extracted with EtOAc and the solvent was then removed under vacuum. The residue was purified by flash column chromatography using a mixture of petroleum ether and ethyl acetate as eluent to give the desired product **3**.

3.2 The procedure for visible-light-induced difunctionalization of alkynes with thiols leading to 3-sulfenyl azaspiro[4,5]trienones.

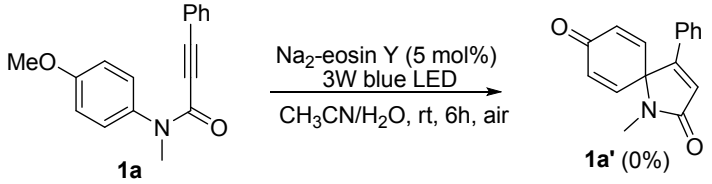


To a solution of thiol **5** (0.25 mmol) and Eosin Y (0.00125mmol, 1 mol %) in CH_3CN 2 mL was added N-(*p*-methoxyaryl)-propiolamide **1** (0.125 mmol). The reaction mixture was open to the air and stirred under the irradiation of 3W blue LED lamps at room temperature for 12h. After completion of the reaction, the solution was concentrated in vacuum. The residue was purified by flash column chromatography using

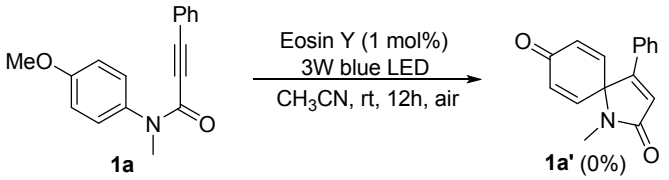
a mixture of petroleum ether and ethyl acetate as eluent to give the desired product **5**.

4. Preliminary mechanistic studies

4.1 N-(p-methoxyaryl)-propiolamide 1a was added independently under the standard conditions.

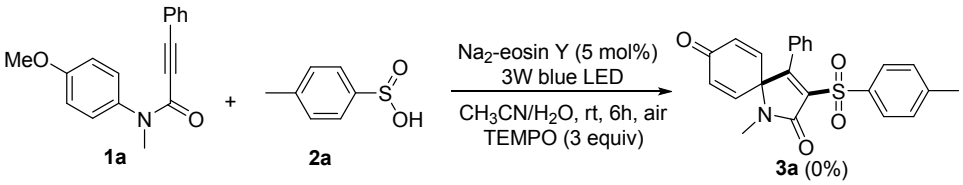


To a solution of Na_2-Eosin Y (0.00625mmol, 5 mol%) in CH_3CN/H_2O (v_1/v_2=1/1) 2 mL was added N-(*p*-methoxyaryl)-propiolamide **1a** (0.125 mmol). The reaction mixture was open to the air and stirred under the irradiation of 3W blue LED lamps at room temperature for 6h. After completion of the reaction, the solution was concentrated in vacuum, none of the desired product **1a'** was detected.



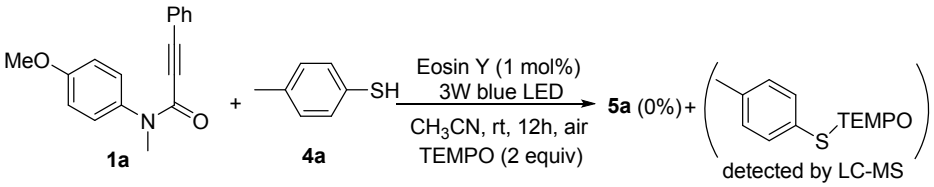
To a solution of Eosin Y (0.00125mmol, 1 mol %) in CH_3CN 2 mL was added N-(*p*-methoxyaryl)-propiolamide **1a** (0.125 mmol). The reaction mixture was open to the air and stirred under the irradiation of 3W blue LED lamps at room temperature for 12h. After completion of the reaction, the solution was concentrated in vacuum, none of the desired product **1a'** was detected.

4.2 The addition of TEMPO in the model reaction systems.

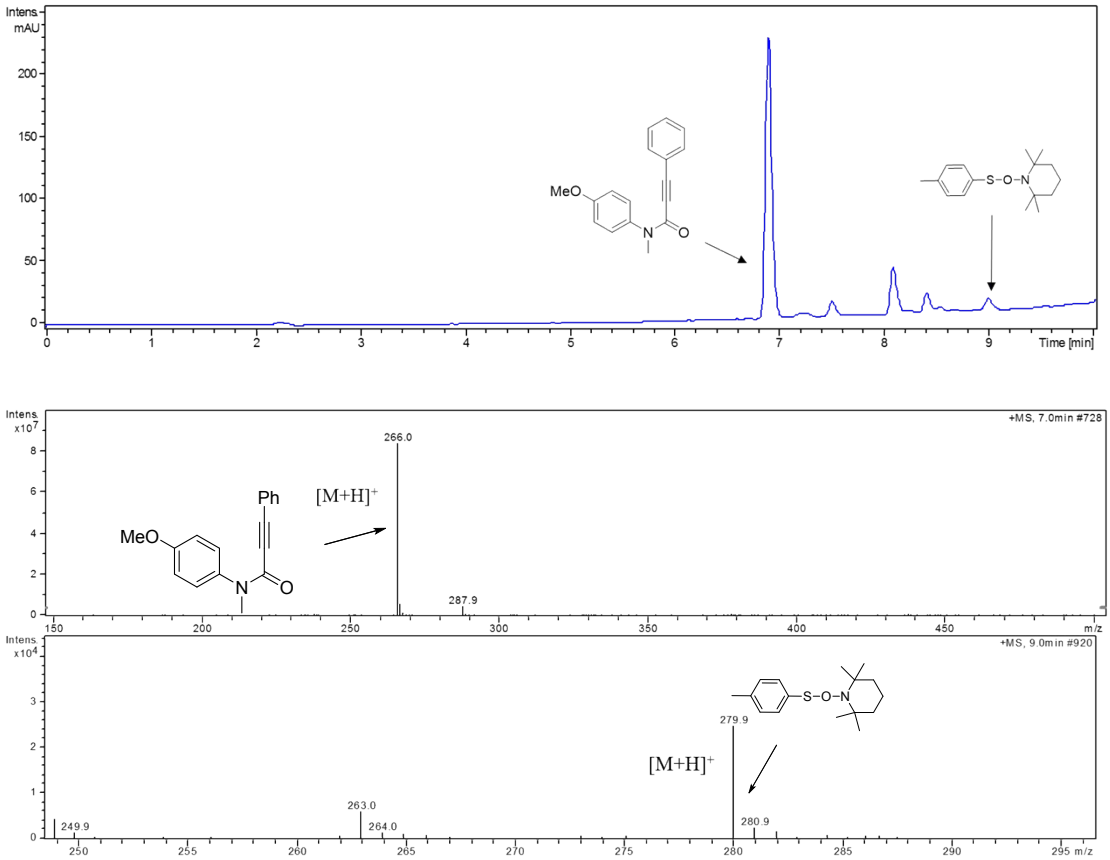


To a solution of 4-methylbenzenesulfinic acid **2a** (0.375 mmol), TEMPO (0.375 mmol), and Na_2-Eosin Y (0.00625mmol, 5 mol%) in CH_3CN/H_2O (v_1/v_2=1/1) 2 mL was added N-(*p*-methoxyaryl)-propiolamide **1** (0.125 mmol) (0.125 mmol). The reaction

mixture was open to air and stirred under the irradiation of 3W blue LED lamps at room temperature for 6h. After completion of the reaction, the solution was concentrated in vacuum, none of desired product **3a** was detected.



To a solution of 4-methylbenzenethiol **2a** (0.25 mmol), TEMPO (0.25 mmol), and Eosin Y (0.001 mmol, 1 mol %) in CH_3CN 2 mL was added N-(*p*-methoxyaryl)-propiolamide **1** (0.125 mmol) (0.125 mmol). The reaction mixture was open to the air and stirred under the irradiation of 3W blue LED lamps at room temperature for 12h. After completion of the reaction, the solution was concentrated in vacuum, TEMPO-trapped thiyl radical complex (p-MePhS–TEMPO) was also detected by LC-MS experiment and none of desired product **5a** was obtained.

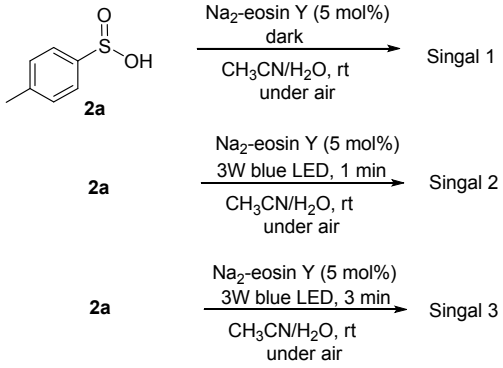


LC-MS spectra

4.3 Investigation of mechanism by ESR

Electron spin resonance (ESR) spectra were recorded with a JEOL JES FA200 (X-band). The sample was bubbled with Ar for over 20 min, then small amount of the sample was transferred to a flat cell, and ESR spectra were recorded under different conditions.

(1)



Mixture of Na_2-eosin Y (2.5 mM), 4-methylbenzenesulfinic acid **2a** (50 mM), and DMPO (100 mM) in CH_3CN/H_2O was transferred to a capillary, and the ESR spectra were recorded under different conditions (Figure S1.). A very weak radical signal was trapped without irradiation of visible light. Notably, when the reaction mixture of **2a** was irradiated with visible light (1 min and 3 min), a distinct signal of a trapped radical was observed, suggesting the involvement of a sulfonyl radical in the transformation. These results indicated that photocatalysis plays an essential role in the formation of the sulfonyl radical.

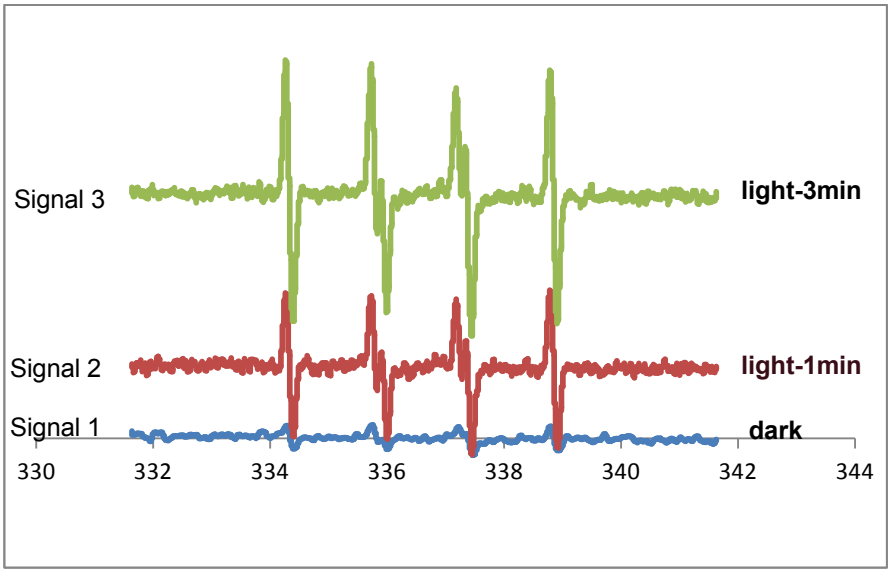
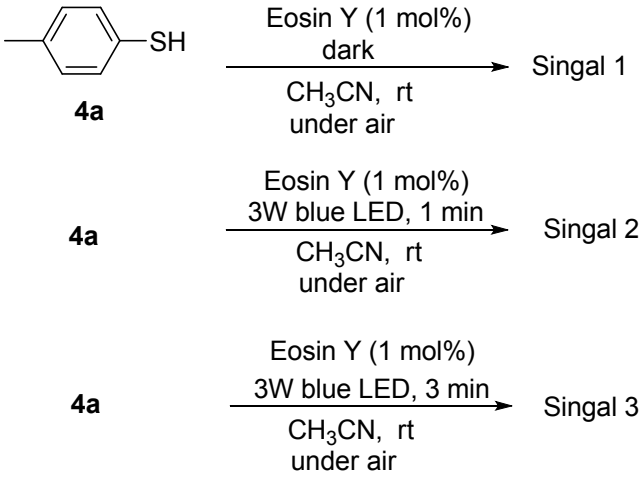


Figure S1. ESR spectra of mixture of Na_2-eosin Y (2.5 mM), 4-methylbenzenesulfinic acid **2a** (50 mM), and DMPO (100 mM) in CH_3CN/H_2O under different conditions. ESR conditions: frequency (9.43 GHz), power (1 mW), modulation width (0.1 mT), center field (336.7 mT), sweep width (10 mT), sweep time (1 min), time constant (0.1 s).

(2)



Mixture of Eosin Y (0.25mM), 4-methylbenzenethiol **4a** (25mM), and DMPO (50 mM) in CH_3CN was transferred to a flat cell, and the ESR spectra were recorded under different conditions (Figure S2). No radical was trapped without irradiation of visible light. Notably, when the reaction mixture of **4a** was irradiated with visible light (1 min and 3 min), a distinct signal of a trapped radical was observed, suggesting the involvement of a thiyl radical in the transformation. These results indicated that photocatalysis plays an essential role in the formation of the thiyl radical.

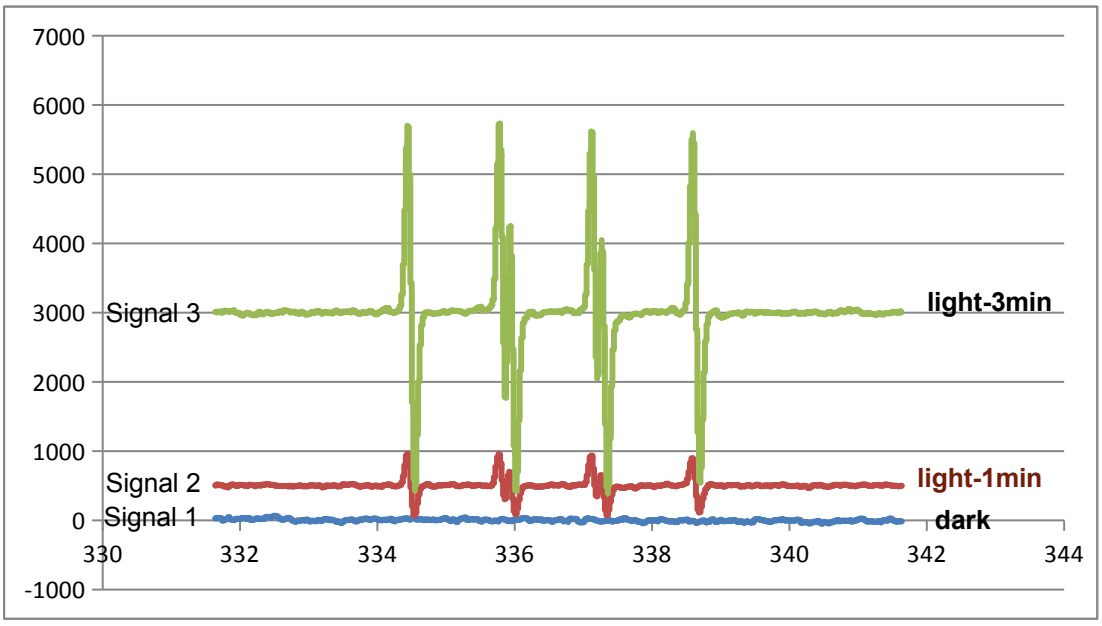


Figure S2. ESR spectra of mixture of Eosin Y (0.25 mM), 4-methylbenzenethiol **4a** (25 mM), and DMPO (50 mM) in CH_3CN under different conditions. ESR conditions: frequency (9.43 GHz), power (1 mW), modulation width (0.1 mT), center field (336.7 mT), sweep width (10 mT), sweep time (1 min), time constant (0.1 s).

4.3 The UV-visible spectroscopy and Fluorescence quenching studies for the synthesis of 3-sulfonyl azaspiro[4,5]trienone (Stern–Volmer Studies)

UV-visible spectroscopy of reaction solution was recorded on a SHIMADZU UV-3600 UV-visible spectrophotometer. The sample was prepared by mixing N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a** with solvent CH_3CN (M[N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide] = 1.25×10^{-2}mol/L in a light path quartz UV cuvette. The absorption was collected and the result was listed in Figure S3.

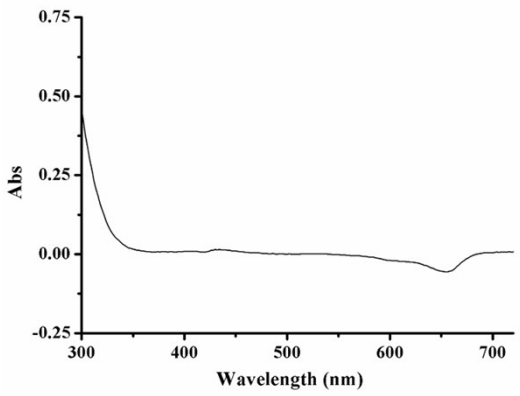


Figure S3. UV–vis spectra of **1a**.

UV-visible spectroscopy of reaction solution was recorded on a SHIMADZU UV-3600 UV-visible spectrophotometer. The sample was prepared by mixing 4-methylbenzenesulfinic acid **2a** with solvent CH_3CN (M[4-methylbenzenesulfinic acid] = 3.75×10^{-2}mol/L in a light path quartz UV cuvette. The absorption was collected and the result was listed in Figure S4.

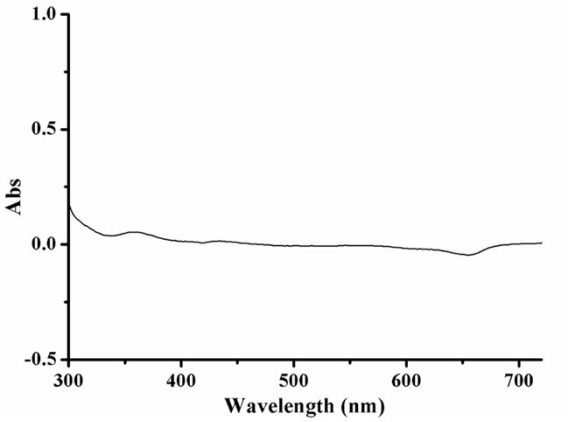


Figure S4. UV–vis spectra of the **2a**.

The fluorescence emission intensities were recorded on a Fluormax-4600 spespectrofluorimeter. The excitation wavelength was fixed at 530nm, and the emission wavelength was measured at 549 nm (emission maximum). The samples were prepared by mixing N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a** in CH_3CN (total volume = 0.1 mL) in a light path quartz fluorescence cuvette. The concentration of N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a** stock solution is 1.25×10^{-2}mol/L in CH_3CN. Then the emission intensity was collected and the result was presented in Figure S5.

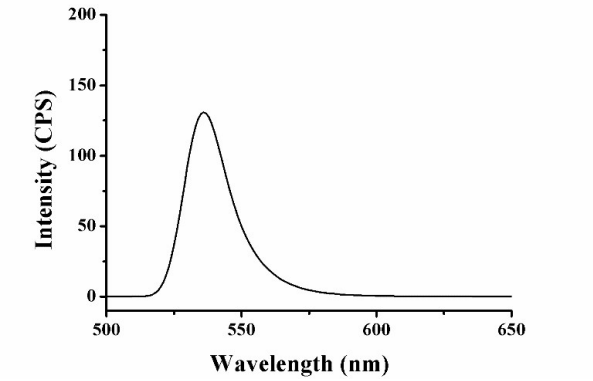


Figure S5. Fluorescence spectra of **1a**

The fluorescence emission intensities were recorded on a Fluormax-4600 spespectrofluorimeter. The excitation wavelength was fixed at 530nm, and the emission wavelength was measured at 549 nm (emission maximum). The samples were prepared by mixing 4-methylbenzenesulfinic acid **2a** in CH_3CN (total volume = 0.1 mL) in a light path quartz fluorescence cuvette. The concentration of 4-methylbenzene sulfonic acid stock solution is 3.75×10^{-2}mol/L in CH_3CN. The concentration of 4-methylbenzenesulfinic acid **2a** stock solution is 3.75×10^{-2}mol/L in CH_3CN. Then, the emission intensity was collected and the result was presented in Figure S6.

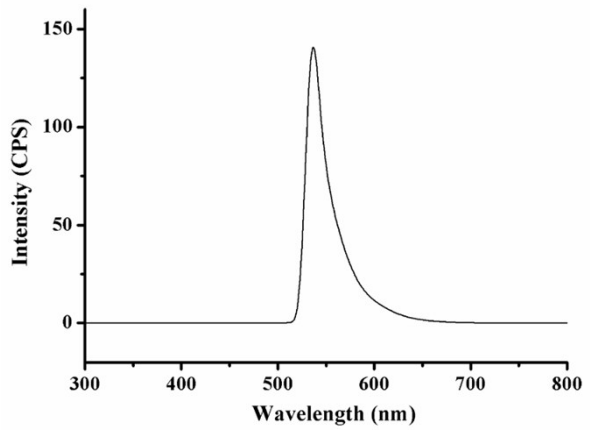


Figure S6 Fluorescence spectra of **2a**.

UV-visible spectroscopy of reaction solution was recorded on a SHIMADZU UV-3600 UV-visible spectrophotometer. The sample was prepared by mixing Na_2-EosinY, N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a** and 4-methylbenzenesulfinic acid **2a** with solvent CH_3CN/H_2O （V_1/V_2=1:1） (M[Na_2-EosinY] = 6.25×10^{-4}mol/L, M[N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide] = 1.25×10^{-2}mol/L, M[4-methylbenzenesulfinic acid] = 3.75×10^{-2}mol/L in a light path quartz UV cuvette. The UV-visible spectroscopy indicated that the maximum absorption wavelength of reaction solution was found to be 530 nm. The absorption was collected and the result was listed in Figure S7.

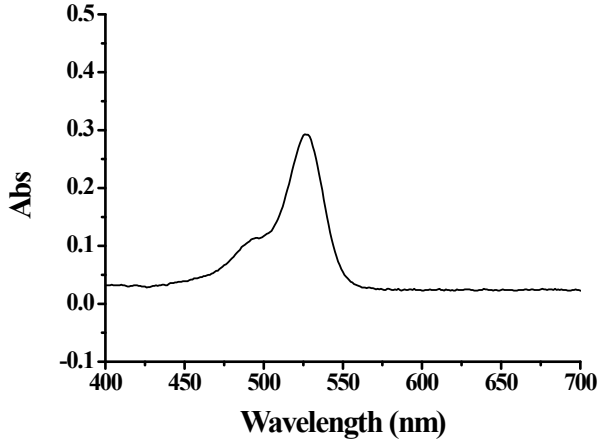


Figure S7. UV–vis spectra of the reaction mixture.

The fluorescence emission intensity of reaction solution was recorded on a Fluoromax-4600 spectrofluorimeter. The excitation wavelength was fixed at 530nm, and the emission wavelength was measured at 549 nm. The sample was prepared by mixing Na_2-EosinY, N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a**, 4-methylbenzenesulfinic acid **2a** with solvent CH_3CN/H_2O (V_1/V_2=1:1) (M[Na_2-EosinY] = 6.25×10^{-6}mol/L, M[N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide] = 1.25×10^{-4}mol/L, M[4-methylbenzenesulfinic acid] = 3.75×10^{-4}mol/L) in a light path quartz fluoresence cuvette. The emission intensity was collected and the result was listed in Figure S8.

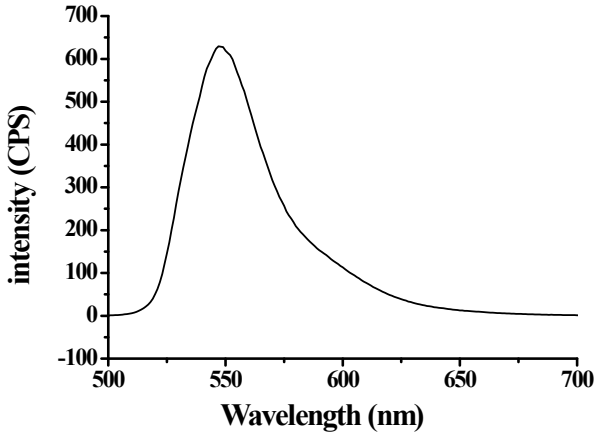


Figure S8. Fluorescence spectra of the reaction mixture.

The fluorescence emission intensities were recorded on a Fluormax-4600 spespectrofluorimeter. The excitation wavelength was fixed at 530nm, and the emission wavelength was measured at 549 nm (emission maximum). The samples were prepared by mixing by Na_2-EosinY (6.25×10^{-6}mol/L) and different amount of 4-methylbenzenesulfinic acid **2a** in CH_3CN (total volume = 0.1 mL) in a light path quartz fluorescence cuvette. The concentration of 4-methylbenzene sulfonic acid stock solution is 3.75×10^{-8}mol/L in CH_3CN. For each quenching experiment, 0.1mL of 4-methylbenzenesulfinic acid **2a** stock solution was titrated to a mixed solution of Na_2-EosinY (0.1mL, in a total volume = 1.0 mL). Then the emission intensity was collected and the results were presented in Figure S9.

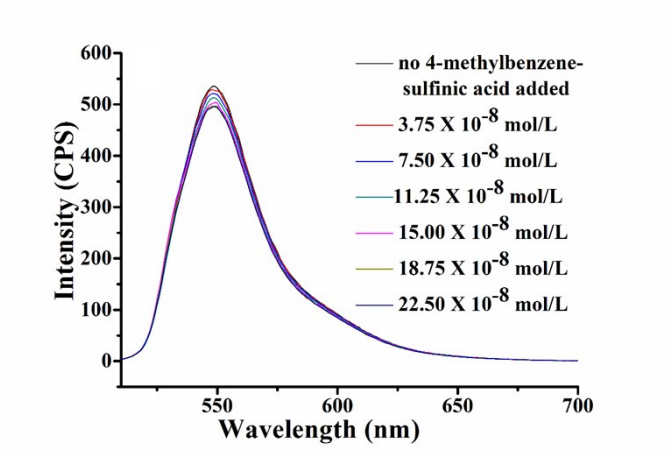


Figure S9. Quenching of Na_2-EosinY fluorescence emission in the presence of 4-methylbenzenesulfinic acid **2a**

An indeed fluorescence quenching phenomenon of Na_2-EosinY under various concentrations of 4-methylbenzenesulfinic acid **2a** was demonstrated in a curve of $[I_0/I]$ vs C [4-methylbenzenesulfinic acid], as shown in Figure S10 (Stern-Volmer plots).

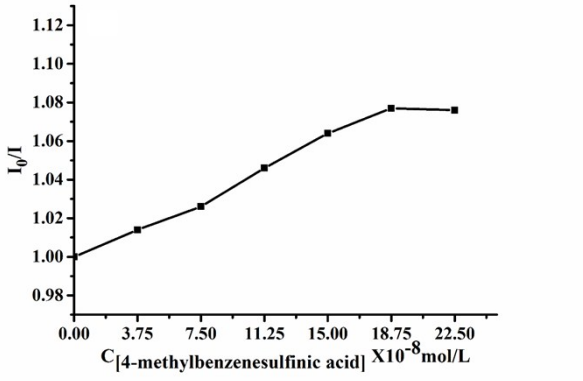


Figure S10. Stern-volmer plots

The fluorescence emission intensities were recorded on a Fluormax-4600 spespectrofluorimeter. The excitation wavelength was fixed at 530nm, and the emission wavelength was measured at 549nm (emission maximum). The samples were prepared by mixing by Na_2-EosinY (6.25×10^{-6}mol/L) and different amount of N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a** in CH_3CN (total volume = 0.1 mL) in a light path quartz fluorescence cuvette. The concentration of N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a** stock solution is 1.25×10^{-8}mol/L in CH_3CN. For each quenching experiment, 0.1mL of alkynes stock solution was titrated to a mixed solution of Na_2-EosinY (0.1mL, in a total volume = 1.0 mL). Then the emission intensity was collected and the results were presented in Figure S11. An fluorescence quenching phenomenon of Na_2-EosinY under various concentrations of alkyne **1a** was shown in Figure S12 (Stern-Volmer plots).

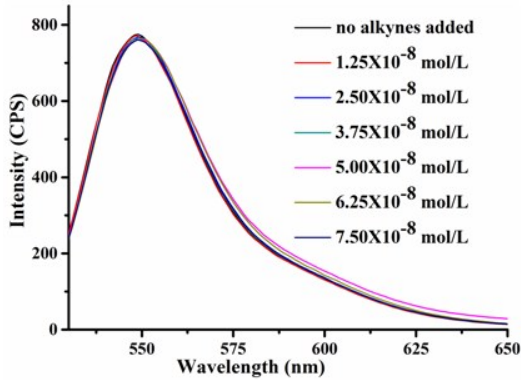


Figure S11. Quenching of Na_2-EosinY fluorescence emission in the presence of alkyne **1a**

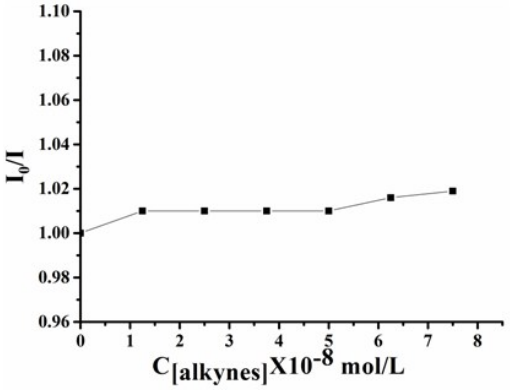


Figure S12. Stern-volmer plots.

4.4 The UV-visible spectroscopy and Fluorescence quenching studies for the

synthesis of 3-sulfenyl azaspiro[4,5]trienone (Stern–Volmer Studies)

UV-visible spectroscopy of reaction solution was recorded on a SHIMADZU UV-3600 UV-visible spectrophotometer. The sample was prepared by mixing of Eosin Y, N-(4-methoxyphenyl)-N-methyl-3-phenylpropiolamide **1a** and thiophenol **4a** with solvent (CH_3CN) (M[EosinY] = 1.25×10^{-5}mol/L, M[alkynes] = 1.25×10^{-3}mol/L, M[thiophenol] = 2.5×10^{-3}mol/L in a light path quartz UV cuvette. The UV-visible spectroscopy indicated that the maximum absorption wavelength of reaction solution was found to be 530 nm. The absorption was collected and the result was listed in Figure S13.

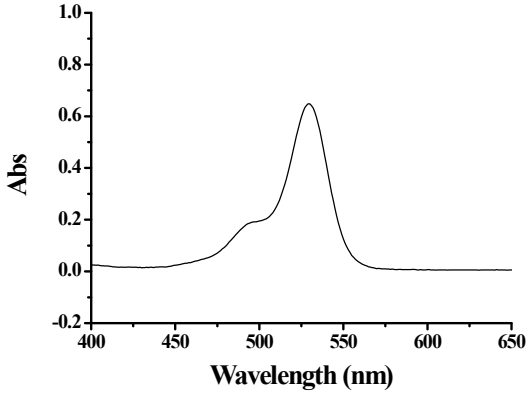


Figure S13. UV–vis spectra of the reaction mixture.

The fluorescence emission intensity of reaction solution was recorded on a Fluoromax-4600 spectrofluorimeter. The excitation wavelength was fixed at 500nm, and the emission wavelength was masured at 558 nm. The sample was prepared by mixing EosinY, alkynes, thiophenol with solvent (CH_3CN) (M[EosinY] = 1.25×10^{-6}mol/L, M[alkyne] = 1.25×10^{-4}mol/L, M[thiophenol] = 2.5×10^{-4}mol) in a light path quartz fluoresence cuvette. The emission intensity was collected and the result was listed in Figure S14.

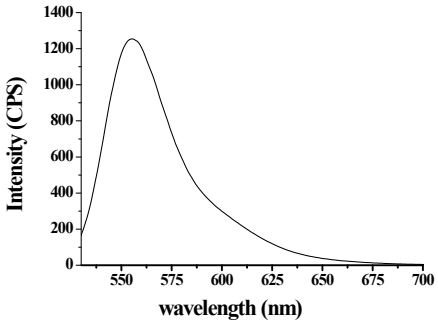


Figure S14. Fluorescence spectra of the reaction mixture

The fluorescence emission intensities were recorded on a Fluormax-4600 spespectrofluorimeter. The excitation wavelength was fixed at 500nm, and the emission wavelength was measured at 558 nm (emission maximum). The samples were prepared by mixing by EosinY (1.25×10^{-6}mol/L) and different amount of Thiophenol in CH_3CN (total volume = 0.1 mL) in a light path quartz fluorescence cuvette. The concentration of Thiophenol stock solution is 2.5×10^{-8}mol/L in CH_3CN. For each quenching experiment, 0.1mL of Thiophenol stock solution was titrated to a mixed solution of EosinY (0.1mL, in a total volume = 1.0 mL). Then the emission intensity was collected and the results were presented in Figure S15.

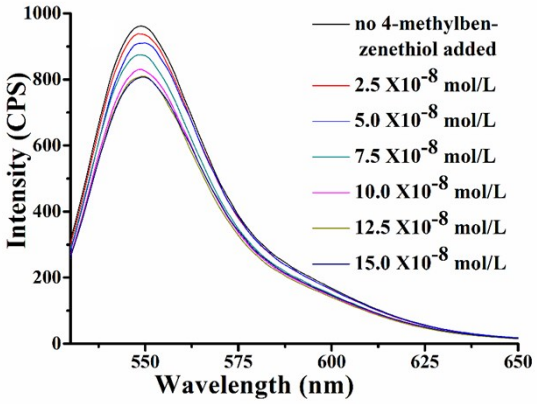


Figure S15. Quenching of EosinY fluorescence emission in the presence of thiophenol **4a**.

An indeed fluorescence quenching phenomenon of EosinY under various concentrations of thiophenol was demonstrated in a curve of $[I_0/I]$ vs $C_{[Thiophenol]}$, as shown in Figure S16 (Stern-Volmer plots).

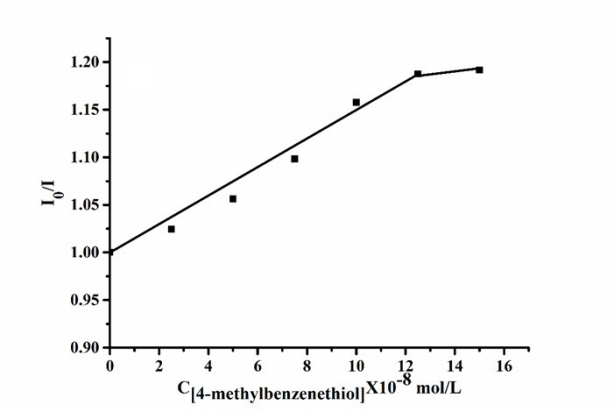


Figure S16. Stern-volmer plots.

The fluorescence emission intensities were recorded on a Fluormax-4600 spespectrofluorimeter. The excitation wavelength was fixed at 500 nm, and the emission wavelength was measured at 557nm (emission maximum). The samples were prepared by mixing by Eosin Y (1.25×10^{-6}mol/L) and different amount of styrene in CH_3CN (total volume = 0.1 mL) in a light path quartz fluorescence cuvette. The concentration of alkynes stock solution is 1.25×10^{-8}mol/L in CH_3CN. For each quenching experiment, 0.1mL of alkynes stock solution was titrated to a mixed solution of Eosin Y (0.1mL, in a total volume = 1.0 mL). Then the emission intensity was collected and the results were presented in Figure S17. An fluorescence quenching phenomenon of EosinY under various concentrations of alkynes was shown in Figure S18 (Stern-Volmer plots).

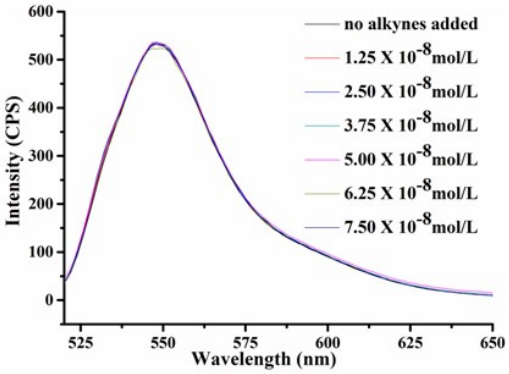


Figure S17. Quenching of Eosin Y fluorescence emission in the presence of alkyne **1a**.

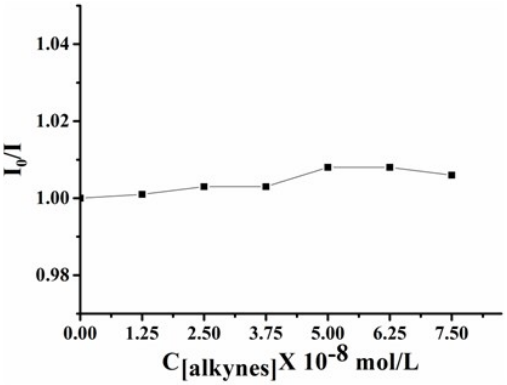
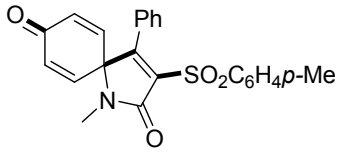
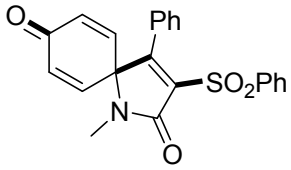


Figure S18. Stern-volmer plots.

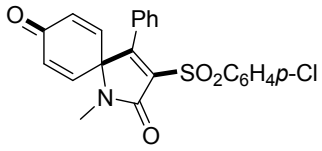
5.Characterization data of products 3a-5r



1-methyl-4-phenyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3a), Compound **3a** was obtained in 76% yield according to the general procedure (6h). White solid; mp 273.2-274.6 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.93 (d, J = 8.3 Hz, 2H), 7.44 (t, J = 7.5 Hz, 1H), 7.39-7.33 (m, 4H), 7.15 (d, J = 7.2 Hz, 2H), 6.43 (s, 4H), 2.83 (s, 3H), 2.44 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.1, 163.6, 161.5, 145.6, 142.3, 137.0, 136.2, 134.3, 130.3, 129.7, 129.3, 128.6, 128.0, 127.8, 68.2, 26.4, 21.8; HRMS calc. for $C_{23}H_{19}NO_4SNa$ $(M+Na)^+$, 428.0932; found, 428.0935.

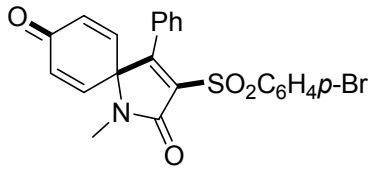


1-methyl-4-phenyl-3-(phenylsulfonyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3b), Compound **3b** was obtained in 60% yield according to the general procedure (8h). White solid; mp 215.7-217.0 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 8.05 (d, J = 8.1 Hz, 2H), 7.66 (d, J = 7.5 Hz, 1H), 7.55 (t, J = 8.1 Hz, 2H), 7.44 (t, J = 8.5 Hz, 1H), 7.38 (t, J = 7.8 Hz, 2H), 7.15 (d, J = 7.1 Hz, 2H), 6.44 (s, 4H), 2.83 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.1, 163.5, 162.1, 142.2, 139.2, 136.7, 134.4, 130.4, 129.2, 129.1, 128.6, 128.0, 127.8, 68.3, 26.4; HRMS calc. for $C_{22}H_{17}NO_4SNa$ $(M+Na)^+$, 414.0776, found, 414.0777.

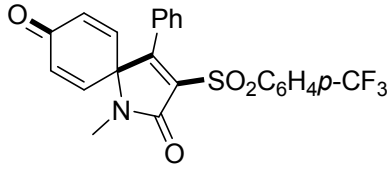


3-(4-chlorophenylsulfonyl)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3c), Compound **3c** was obtained in 55% yield according to the general procedure (10 h). Yellow solid; mp 267.9-268.6 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.99 (d, J = 8.7 Hz, 2H), 7.52 (d, J = 8.7 Hz, 2H), 7.46 (t, J = 7.5 Hz, 1H), 7.39 (t, J = 7.9 Hz, 2H), 7.15 (d, J = 7.1 Hz, 2H), 6.47-6.42 (m, 4H), 2.84 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.0, 163.4, 162.4, 142.0, 141.3, 137.5, 136.4, 134.5, 130.8, 130.6, 129.4, 128.4,

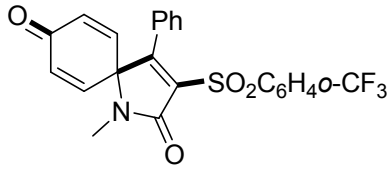
128.1, 127.8, 68.4, 26.4; HRMS calc. for $C_{22}H_{16}{}^{35}ClNO_4SNa$ $(M+Na)^+$, 448.0386; found, 448.0384.



3-(4-bromophenylsulfonyl)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3d), Compound **3d** was obtained in 85% yield according to the general procedure (12h). Yellow solid; mp 274.4-275.2 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.91 (d, J = 8.7 Hz, 2H), 7.69 (d, J = 8.7 Hz, 2H), 7.46 (t, J = 7.5Hz, 1H), 7.39 (t, J = 7.9 Hz, 2H), 7.14 (d, J = 7.1 Hz, 2H), 6.47-6.42 (m, 4H), 2.84 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 182.9, 163.4, 162.5, 142.0, 138.1, 136.3, 134.5, 132.4, 130.8, 130.6, 130.1, 128.4, 128.1, 127.8, 68.4, 26.4; HRMS calc. for $C_{22}H_{16}{}^{79}BrNO_4SNa$ $(M+Na)^+$, 491.9881; found, 491.9883.

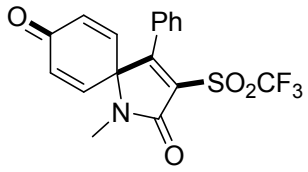


1-methyl-4-phenyl-3-(4-(trifluoromethyl)phenylsulfonyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3e), Compound **3e** was obtained in 81% yield according to the general procedure (6h). Yellow solid; mp 254.7-255.5 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 8.19 (d, J = 8.2 Hz, 2H), 7.81 (d, J = 8.4 Hz, 2H), 7.47 (t, J = 7.5 Hz, 1H), 7.40 (t, J = 7.9 Hz, 2H), 7.16 (d, J = 7.4 Hz, 2H), 6.48-6.42 (m, 4H), 2.84 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 182.9, 163.3 (d, J = 6.8 Hz), 142.6, 141.8, 136.0, 135.7, 134.5, 130.7, 129.9, 128.2, 128.1, 127.8, 126.2 (q, J = 3.7 Hz), 123.1 (d, J = 271.6 Hz), 68.5, 26.4; HRMS calc. for $C_{23}H_{16}F_3NO_4SNa$ $(M+Na)^+$, 482.0650; found, 482.0655.

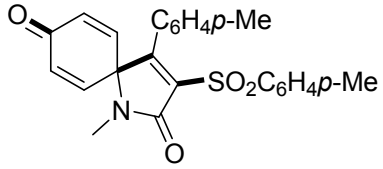


1-methyl-4-phenyl-3-(2-(trifluoromethyl)phenylsulfonyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3f), Compound **3f** was obtained in 80% yield according to the general procedure (8h). White solid; mp 219.6-220.9 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ

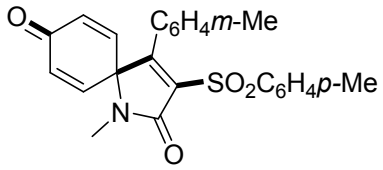
8.52 (d, J = 7.4 Hz, 1H), 7.82-7.78 (m, 3H), 7.44 (t, J = 7.5 Hz, 1H), 7.38 (t, J = 7.9 Hz, 2H), 7.30 (d, J = 7.3 Hz, 2H), 6.51-6.45 (m, 4H), 2.79 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.2, 162.3, 161.4, 142.2, 137.4, 136.0, 134.4, 134.2, 132.2, 130.8, 128.7 (d, J = 33.3 Hz), 128.3, 128.1, 128.1, 128.0 (q, J = 6.0 Hz), 122.9 (d, J = 272.9 Hz), 68.0, 26.2; HRMS calc. for $C_{23}H_{16}F_3NO_4SNa$ $(M+Na)^+$, 482.0650; found, 482.0653.



1-methyl-4-phenyl-3-(trifluoromethylsulfonyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3g), Compound **3g** was obtained in 37% yield according to the general procedure (24h). White solid; mp 198.7-200.4 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.41-7.38 (m, 5H), 6.54-6.49 (m, 4H), 2.95 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.7, 165.8, 151.3, 144.1, 133.5, 130.3, 130.2, 128.8, 127.8, 119.9, 68.3, 26.7; HRMS calc. for $C_{17}H_{12}F_3NO_4SNa$ $(M+Na)^+$, 406.0337; found, 406.0341.

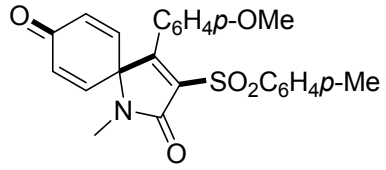


1-methyl-4-p-tolyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3h), Compound **3h** was obtained in 52% yield according to the general procedure (6h). Yellow solid; mp 213.5-215.1 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.93 (d, J = 8.3 Hz, 2H), 7.34 (d, J = 8.1 Hz, 2H), 7.17 (d, J = 7.9 Hz, 2H), 7.07 (d, J = 8.2 Hz, 2H), 6.45-6.40 (m, 4H), 2.81 (s, 3H), 2.44 (s, 3H), 2.37 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.2, 163.7, 161.9, 145.5, 142.5, 140.8, 136.5, 136.3, 134.2, 129.7, 129.3, 128.7, 127.8, 125.7, 68.2, 26.3, 21.8, 21.5; HRMS calc. for $C_{24}H_{21}NO_4SNa$ $(M+Na)^+$, 442.1089; found, 442.1091.

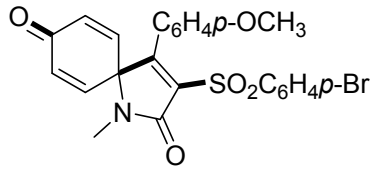


1-methyl-4-m-tolyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione(3i), Compound **3i** was obtained in 60% yield according to the general procedure (20h). Yellow solid; mp 266.7-268.4 ^{o}C. ^{1}H NMR ($CDCl_3$, 400 MHz, ppm): δ 7.92 (d, J = 8.3 Hz, 2H), 7.34 (d, J

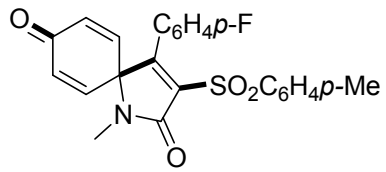
= 8.1 Hz, 2H), 7.25-7.22 (m, 2H), 6.94-6.91 (m, 2H), 6.45-6.40 (m, 4H), 2.82 (s, 3H), 2.44 (s, 3H), 2.35 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.2, 163.7, 161.8, 145.5, 142.4, 137.6, 136.7, 136.3, 134.2, 131.2, 130.0, 129.3, 128.6, 128.4, 127.8, 124.8, 68.2, 26.3, 21.8, 21.4; HRMS calc. for $C_{24}H_{21}NO_4SNa$ $(M+Na)^+$, 442.1089; found, 442.1091.



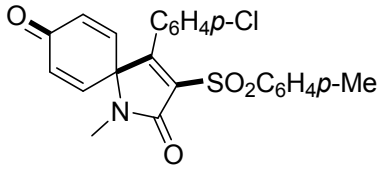
4-(4-methoxyphenyl)-1-methyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3j), Compound **3j** was obtained in 81% yield according to the general procedure (6h). White solid; mp 216.5-218.3 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.94 (d, J = 8.3 Hz, 2H), 7.34 (d, J = 8.0 Hz, 2H), 7.19 (d, J = 8.9 Hz, 2H), 6.88 (d, J = 8.9 Hz, 2H), 6.48-6.40 (m, 4H), 3.83 (s, 3H), 2.80 (s, 3H), 2.44 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.3, 163.8, 161.5, 161.4, 145.4, 142.8, 136.4, 135.9, 134.1, 129.8, 129.7, 129.3, 120.7, 113.6, 68.1, 55.3, 26.2, 21.8; HRMS calc. for $C_{24}H_{21}NO_5SNa$ $(M+Na)^+$, 458.1038; found, 458.1037.



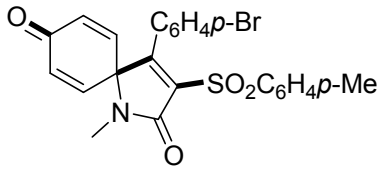
3-(4-bromophenylsulfonyl)-4-(4-methoxyphenyl)-1-methyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3k), Compound **3k** was obtained in 53% yield according to the general procedure (10h). White solid; mp 223.0-224.1 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.92 (d, J = 8.7 Hz, 2H), 7.68 (d, J = 8.7 Hz, 2H), 7.19 (d, J = 8.9 Hz, 2H), 6.89 (d, J = 8.3 Hz, 2H), 6.48 (d, J = 10.3 Hz, 2H), 6.41 (d, J = 10.3 Hz, 2H), 3.83 (s, 3H), 2.80 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.1, 163.6, 162.4, 161.6, 142.5, 138.2, 135.2, 134.2, 132.3, 130.9, 130.0, 129.8, 120.5, 113.7, 68.2, 55.3, 26.2; HRMS calc. for $C_{23}H_{18}{}^{79}BrNO_5SNa$ $(M+Na)^+$, 521.9987; found, 521.9989.



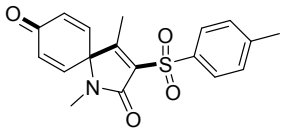
4-(4-fluorophenyl)-1-methyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3l), Compound **3l** was obtained in 61% yield according to the general procedure (6h). White solid; mp 267.8-268.1 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.92 (d, J = 8.3 Hz, 2H), 7.35 (d, J = 8.1 Hz, 2H), 7.19-7.16 (m, 2H), 7.08 (t, J = 8.5 Hz, 2H), 6.46 (t, J = 8.2 Hz, 2H), 6.42 (t, J = 8.6 Hz, 2H), 2.82 (s, 3H), 2.45 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 182.9, 163.8 (d, J = 250.5 Hz), 163.4, 160.3, 145.8, 142.2, 137.3, 136.0, 134.5, 130.1 (d, J = 8.6 Hz), 129.8, 129.3, 124.5 (d, J = 3.6 Hz), 115.5 (d, J = 22.0 Hz), 68.2, 26.4, 21.8; HRMS calc. for $C_{23}H_{18}FNO_4SNa$ $(M+Na)^+$, 446.0838; found, 446.0839.



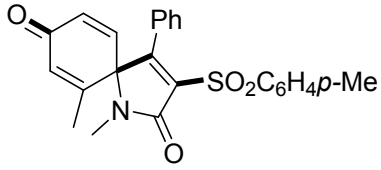
4-(4-chlorophenyl)-1-methyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3m), Compound **3m** was obtained in 53% yield according to the general procedure (6h). Yellow solid; mp 231.2-232.7 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.92 (d, J = 8.4 Hz, 2H), 7.37-7.34 (m, 4H), 7.11 (d, J = 8.6 Hz, 2H), 6.47-6.40 (m, 4H), 2.82 (s, 3H), 2.45 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 182.9, 163.3, 160.1, 145.8, 142.1, 137.5, 136.8, 135.9, 134.5, 129.8, 129.3, 129.3, 128.5, 127.0, 68.1, 26.4, 21.8; HRMS calc. for $C_{23}H_{18}{}^{35}ClNO_4SNa$ $(M+Na)^+$, 462.0543; found, 462.0544.



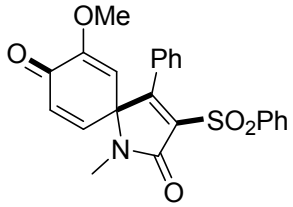
4-(4-bromophenyl)-1-methyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3n), Compound **3n** was obtained in 49% yield according to the general procedure (7h). Yellow solid; mp 258.1-259.4 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.91 (d, J = 8.1 Hz, 2H), 7.51 (d, J = 8.3 Hz, 2H), 7.35 (d, J = 8.1 Hz, 2H), 7.04 (d, J = 8.4 Hz, 2H), 6.46 (d, J = 10.1 Hz, 2H), 6.40 (d, J = 10.1 Hz, 2H), 2.82 (s, 3H), 2.45 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 182.8, 163.3, 160.0, 145.8, 142.0, 137.5, 135.9, 134.6, 131.4, 129.8, 129.4, 129.4, 127.5, 125.2, 68.0, 26.4, 21.8; HRMS calc. for $C_{23}H_{18}{}^{79}BrNO_4SNa$ $(M+Na)^+$, 506.0038; found, 506.0041.



1,4-dimethyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **3o** was obtained in 25% yield according to the general procedure (24h). White solid; mp 231.2-232.4 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 8.01 (d, J = 8.2 Hz, 2H), 7.37 (d, J = 8.1 Hz, 2H), 6.61 (d, J = 10.0 Hz, 2H), 6.29 (d, J = 10.1 Hz, 2H), 2.77 (s, 3H), 2.45 (s, 3H), 2.31 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.3, 163.8, 161.2, 145.6, 143.4, 136.4, 135.6, 134.5, 129.8, 129.0, 68.2, 26.3, 21.8, 12.1; HRMS calc. for $C_{18}H_{17}NO_4SNa$ $(M+Na)^+$, 366.0776; found, 366.0779.

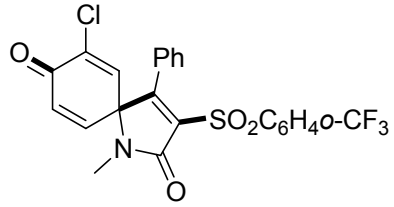


1,6-dimethyl-4-phenyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3p), Compound **3o** was obtained in 80% yield according to the general procedure (10h). Yellow solid; mp 219.1-220.7 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.92 (d, J = 8.3 Hz, 2H), 7.45 (t, J = 8.4 Hz, 1H), 7.37 (t, J = 8.0 Hz, 2H), 7.34 (d, J = 8.1 Hz, 2H), 7.16 (d, J = 7.2 Hz, 2H), 6.44-6.31 (m, 3H), 2.73 (s, 3H), 2.44 (s, 3H), 1.73 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.8, 164.0, 161.7, 150.7, 145.5, 142.4, 137.4, 136.3, 133.9, 133.0, 130.7, 129.7, 129.2, 128.4, 128.1, 127.8, 70.5, 26.0, 21.8, 17.7; HRMS calc. for $C_{24}H_{21}NO_4SNa$ $(M+Na)^+$, 442.1089; found, 442.1092.

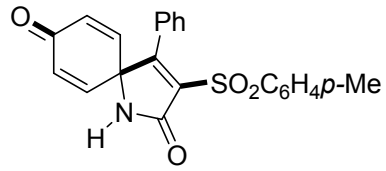


7-methoxy-1-methyl-4-phenyl-3-(phenylsulfonyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3q), Compound **3p** was obtained in 64% yield according to the general procedure (8h). Yellow solid; mp 238.1-239.8 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 8.06 (t, J = 7.3 Hz, 2H), 7.66 (d, J = 7.4 Hz, 1H), 7.55 (t, J = 8.1 Hz, 2H), 7.42 (d, J = 7.5 Hz, 1H), 7.37 (t, J = 7.8 Hz, 2H), 7.12 (t, J = 7.2 Hz, 2H), 6.42 (s, 2H), 5.28 (s, 1H), 3.66 (s, 3H), 2.82 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 178.7, 163.3, 163.2, 154.6,

142.9, 139.3, 136.1, 134.3, 133.8, 130.3, 129.3, 129.1, 128.9, 128.7, 128.0, 127.7, 108.5, 69.7, 55.7, 26.2; HRMS calc. for $C_{23}H_{19}NO_5SNa$ $(M+Na)^+$, 444.0882; found, 444.0883.

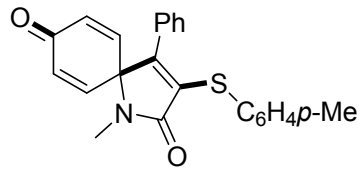


7-chloro-1-methyl-4-phenyl-3-(2-(trifluoromethyl)phenylsulfonyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3q), Compound **3r** was obtained in 71% yield according to the general procedure (10h). White solid; mp 196.7-198.5 ^oC. ^1H NMR ($CDCl_3$, 500 MHz, ppm): δ 8.50 (d, J = 7.2 Hz, 1H), 7.83-7.79 (m, 3H), 7.46 (t, J = 7.5 Hz, 1H), 7.39 (t, J = 7.9 Hz, 2H), 7.27-7.25 (m, 2H), 6.66 (d, J = 2.8 Hz, 1H), 6.56-6.51 (m, 2H), 2.81 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 176.4, 162.9, 160.5, 142.9, 137.9, 137.8, 136.7 (d, J = 119.6 Hz), 134.3 (d, J = 16.7 Hz), 133.3, 132.2, 131.0, 128.8, 128.7 (d, J = 33.2 Hz) , 128.3, 128.0, 127.9, 122.9 (d, J = 272.9 Hz), 69.7, 26.4; HRMS calc. for $C_{23}H_{15}{}^{35}ClF_3NO_4SNa$ $(M+Na)^+$, 516.0260; found, 516.0261.



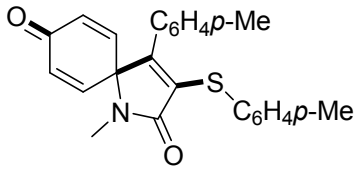
4-phenyl-3-tosyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (3s), Compound **3r** was obtained in 24% yield according to the general procedure (24h). Grey oil. ^1H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.87 (d, J = 8.3 Hz, 2H), 7.44 (t, J = 7.4 Hz, 1H), 7.37 (t, J = 7.8 Hz, 2H), 7.32 (d, J = 8.2 Hz, 2H), 7.11 (d, J = 7.4 Hz, 2H), 6.56 (s, 1H), 6.55 (d, J = 10.0 Hz, 2H), 6.31 (d, J = 10.0 Hz, 2H), 2.44 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.2, 165.5, 164.6, 145.6, 142.5, 136.3, 136.3, 132.6, 130.4, 129.8, 129.2, 128.4, 128.0, 127.8, 64.0, 21.8; HRMS calc. for $C_{22}H_{17}NO_4SNa$ $(M+Na)^+$, 414.0770; found, 414.0764.

Characterization data of products 5a-5q



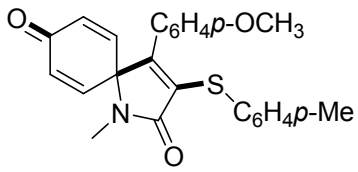
1-methyl-4-phenyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione

Compound **5a** was obtained in 87% yield according to the general procedure (12h). Yellow solid; mp 159.3-160.2 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.31-7.28 (m, 1H), 7.25-7.20 (m, 2H), 7.23-7.19 (m, 4H), 6.99 (d, J = 8.0 Hz, 2H), 6.51 (d, J = 10.2 Hz, 2H), 6.45 (d, J = 10.3 Hz, 2H), 2.88 (s, 3H), 2.26 (s, 3H); ^{13}C NMR ($CDCl_3$, 125MHz, ppm): δ 184.0, 167.8, 151.7, 145.3, 137.8, 133.1, 132.8, 131.7, 130.8, 129.7, 129.5, 128.3, 128.2, 127.7, 67.7, 26.3, 21.1; HRMS calc. for $C_{23}H_{19}NO_2SNa$ $(M+Na)^+$, 396.1034; found, 396.1036.



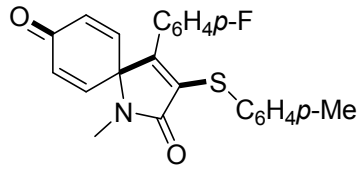
1-methyl-4-p-tolyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione,

Compound **5b** was obtained in 69% yield according to the general procedure (12h). Yellow solid; mp 183.9-184.5 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.21 (d, J = 7.9 Hz, 2H), 7.15 (d, J = 7.9 Hz, 2H), 7.07 (d, J = 7.9 Hz, 2H), 7.00 (d, J = 7.9 Hz, 2H), 6.51 (d, J = 10.1 Hz, 2H), 6.46 (d, J = 10.1 Hz, 2H), 2.86 (s, 3H), 2.31 (s, 3H), 2.27 (s, 3H); ^{13}C NMR ($CDCl_3$, 125MHz, ppm): δ 184.1, 167.8, 152.3, 145.5, 139.9, 137.7, 133.1, 132.0, 131.5, 129.7, 129.0, 128.1, 128.0, 127.8, 67.6, 26.2, 21.3, 21.1; HRMS calc. for $C_{24}H_{21}NO_2SNa$ $(M+Na)^+$, 410.1191; found, 410.1195.

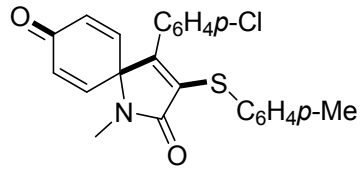


4-(4-methoxyphenyl)-1-methyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5c** was obtained in 65% yield according to the general procedure (12h). Yellow solid; mp 145.1-145.9 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.28 (t, J = 8.6 Hz, 2H), 7.21 (d, J = 7.9 Hz, 2H), 7.02 (d, J = 7.8 Hz, 2H), 6.78 (d, J = 8.6 Hz, 2H), 6.51 (d, J = 10.2 Hz, 2H), 6.48 (d, J = 10.2 Hz, 2H), 3.78 (s, 3H), 2.86 (s, 3H), 2.28 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 184.1, 167.9, 160.7, 152.2, 145.7, 137.6, 133.0,

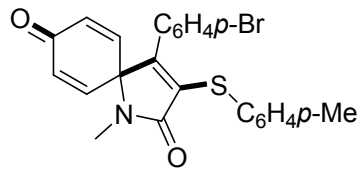
131.1, 130.9, 130.0, 129.7, 128.3, 123.1, 113.8, 67.4, 55.3, 26.1, 21.1; HRMS calc. for $C_{24}H_{21}NO_3SNa$ $(M+Na)^+$, 426.1140; found, 426.1143.



4-(4-fluorophenyl)-1-methyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5d** was obtained in 70% yield according to the general procedure (12h). Yellow solid; mp 144.4-145.7 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.18 (d, J = 7.9 Hz, 4H), 6.98 (d, J = 7.9 Hz, 2H), 6.91 (t, J = 8.5 Hz, 2H), 6.50 (d, J = 10.1 Hz, 2H), 6.50 (d, J = 10.1 Hz, 2H), 2.88 (s, 3H), 2.27 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.8, 167.7, 163.1 (d, J = 249.4 Hz), 149.7, 145.2, 138.1, 133.3, 133.2, 131.9, 130.2 (d, J = 33.5 Hz), 129.7, 127.2, 126.7 (d, J = 13.7 Hz), 115.5 (d, J = 21.7 Hz), 67.7, 26.3, 21.1; HRMS calc. for $C_{23}H_{18}FNO_2SNa$ $(M+Na)^+$, 414.0940; found, 414.0937.

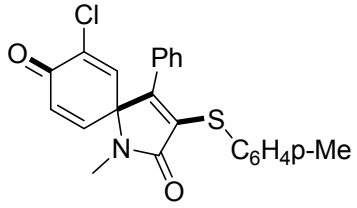


4-(4-chlorophenyl)-1-methyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5e** was obtained in 57% yield according to the general procedure (12h). Yellow oil; ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.10 (d, J = 7.6 Hz, 4H), 7.01 (d, J = 8.3 Hz, 2H), 6.90 (d, J = 7.9 Hz, 2H), 6.42 (d, J = 10.0 Hz, 2H), 6.38 (d, J = 10.1 Hz, 2H), 2.81 (s, 3H), 2.20 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 182.7, 166.5, 147.7, 144.0, 137.3, 134.5, 132.8, 132.2, 131.2, 128.7, 128.5, 128.0, 127.5, 125.7, 66.6, 25.3, 20.1; HRMS calc. for $C_{23}H_{18}{}^{35}ClNO_2SNa$ $(M+Na)^+$, 430.0644; found, 430.0649.

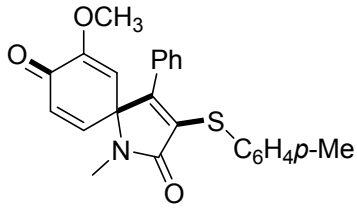


4-(4-bromophenyl)-1-methyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5f** was obtained in 53% yield according to the general procedure (12h).

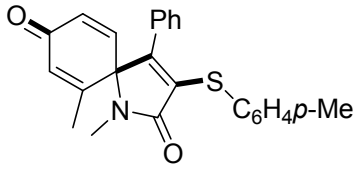
Yellow oil; ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.32 (d, J = 8.4 Hz, 2H), 7.16 (d, J = 8.0 Hz, 2H), 7.00 (d, J = 8.4 Hz, 2H), 6.96 (d, J = 7.9 Hz, 2H), 6.48 (d, J = 10.2 Hz, 2H), 6.45 (d, J = 10.2 Hz, 2H), 2.88 (s, 3H), 2.28 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.7, 167.5, 148.5, 145.0, 138.4, 133.9, 133.3, 132.3, 131.4, 129.8, 129.7, 129.5, 126.6, 123.8, 67.6, 26.3, 21.2; HRMS calc. for $C_{23}H_{18}{}^{79}BrNO_2SNa$ $(M+Na)^+$, 474.0139; found, 474.0141.



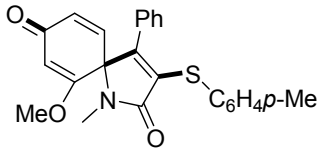
7-chloro-1-methyl-4-phenyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5g** was obtained in 71% yield according to the general procedure (12h). Yellow solid; mp 169.8-171.6 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.30 (t, J = 7.1 Hz, 1H), 7.24 (d, J = 7.3 Hz, 2H), 7.19 (d, J = 8.0 Hz, 2H), 7.13 (d, J = 7.5 Hz, 2H), 6.97 (d, J = 7.8 Hz, 2H), 6.72 (d, J = 2.2 Hz, 1H), 6.55-6.49 (m, 2H), 2.90 (s, 3H), 2.26 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 177.2, 167.4, 150.0, 145.9, 141.0, 138.1, 136.3, 133.5, 132.2, 132.0, 130.2, 129.7, 129.6, 128.4, 128.2, 127.1, 69.5, 26.5, 21.1; HRMS calc. for $C_{23}H_{18}{}^{35}ClNO_2SNa$ $(M+Na)^+$, 430.0644; found, 430.0641.



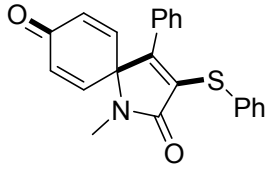
(R)-7-methoxy-1-methyl-4-phenyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5h** was obtained in 58% yield according to the general procedure (12h). Yellow oil. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.29 (t, J = 7.2 Hz, 1H), 7.25-7.21 (m, 4H), 7.18 (d, J = 7.5 Hz, 2H), 6.99 (d, J = 7.9 Hz, 2H), 6.52-6.44 (m, 2H), 5.35 (d, J = 2.1 Hz, 1H), 3.66 (s, 3H), 2.87 (s, 3H), 2.27 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 179.5, 167.5, 153.8, 152.9, 146.0, 137.8, 132.3, 132.1, 131.8, 130.9, 129.7, 129.5, 128.3, 128.2, 127.8, 111.4, 69.1, 55.5, 26.0, 21.1; HRMS calc. for $C_{24}H_{21}NO_3SNa$ $(M+Na)^+$, 426.1140; found, 426.1147.



(S)-1,6-dimethyl-4-phenyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione,Compound **5i** was obtained in 87% yield according to the general procedure (12h). Yellow oil. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.34-7.32 (m, 1H), 7.29-7.25 (m, 4H), 7.23 (d, J = 8.0 Hz, 2H), 7.02 (d, J = 7.9 Hz, 2H), 6.49 (s, 2H), 6.37 (s, 1H), 2.79 (s, 3H), 2.29 (s, 3H), 1.78 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 184.7, 168.1, 153.5, 152.1, 145.5, 137.8, 133.0, 132.6, 132.0, 131.5, 130.6, 129.8, 128.4, 128.1, 127.9, 69.7, 25.8, 21.1,17.9; HRMS calc. for $C_{24}H_{21}NO_2SNa$ $(M+Na)^+$, 410.1191; found, 410.1196.

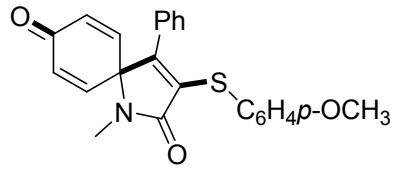


6-methoxy-1-methyl-4-phenyl-3-(p-tolylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5j** was obtained in 68% yield according to the general procedure (12h). Yellow oil. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.30-7.22 (m, 3H), 7.19-7.14 (m, 4H), 6.99 (d, J = 8.0 Hz, 2H), 6.40-6.37 (dd, J_1 = 1.3 Hz, J_2 = 9.8 Hz, 1H), 6.30 (d, J = 9.8 Hz, 1H), 5.76 (d, J = 1.1 Hz, 1H), 3.74 (s, 3H), 2.78 (s, 3H), 2.26 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 186.2, 168.7, 168.5, 152.8, 141.0, 137.3, 132.4, 132.2, 130.8, 130.6, 129.7, 129.5, 128.5, 128.3, 128.0, 106.2, 69.0, 56.3, 26.0, 21.1; HRMS calc. for $C_{24}H_{21}NO_3SNa$ $(M+Na)^+$, 426.1140; found, 426.1146.

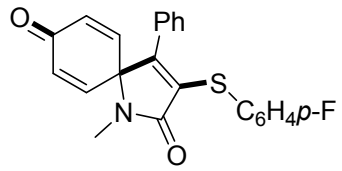


1-methyl-4-phenyl-3-(phenylthio)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione,
Compound **5k** was obtained in 56% yield according to the general procedure (12h). Yellow oil. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.29-7.23 (m, 7H), 7.20-7.17 (m, 3H), 6.53 (d, J = 10.0 Hz, 2H), 6.47 (d, J = 10.1 Hz, 2H), 2.89 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.9, 167.7, 152.8, 145.1, 133.2, 132.2, 131.7, 131.0, 130.7, 129.7, 128.9,

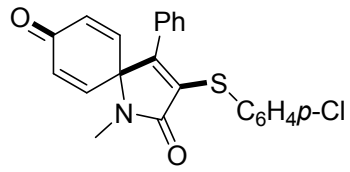
128.3, 128.1, 127.5, 67.7, 26.3; HRMS calc. for $C_{22}H_{17}NO_2SNa$ $(M+Na)^+$, 382.0878; found, 382.0879.



3-(4-methoxyphenylthio)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5l** was obtained in 50% yield according to the general procedure (12h). Yellow solid; mp 156.3-157.6 ^oC. ^1H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.28-7.27 (m, 2H), 7.24-7.22 (m, 3H), 7.17-7.15 (m, 2H), 6.69 (d, J = 8.9 Hz, 2H), 6.49 (d, J = 10.3 Hz, 2H), 6.43 (d, J = 10.2 Hz, 2H), 3.74 (s, 3H), 2.87 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 184.0, 167.9, 159.8, 150.2, 145.3, 134.6, 133.5, 133.1, 130.7, 129.4, 128.3, 128.2, 121.1, 114.5, 67.7, 55.3, 26.3; HRMS calc. for $C_{23}H_{19}NO_3SNa$ $(M+Na)^+$, 412.0983; found, 412.0985.

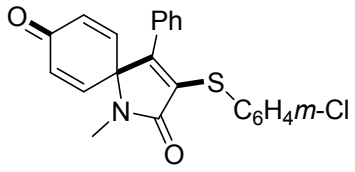


3-(4-fluorophenylthio)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dion, Compound **5m** was obtained in 62% yield according to the general procedure (12h). Yellow solid; mp 126.7-127.6 ^oC. ^1H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.31-7.30 (m, 3H), 7.24 (d, J = 7.4 Hz, 2H), 7.17 (d, J = 7.5 Hz, 2H), 6.87 (d, J = 8.5 Hz, 2H), 6.51 (d, J = 10.0 Hz, 2H), 6.45 (d, J = 10.3 Hz, 2H), 2.88 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.9, 167.6, 162.5, 151.6, 145.0, 134.2 (d, J = 8.3 Hz), 133.2, 132.7, 130.5, 129.7, 128.3, 128.2, 126.1 (d, J = 3.3 Hz), 116.1 (d, J = 22.0 Hz), 67.7, 26.3; HRMS calc. for $C_{22}H_{16}FNO_2SNa$ $(M+Na)^+$, 400.0783; found, 400.0788.



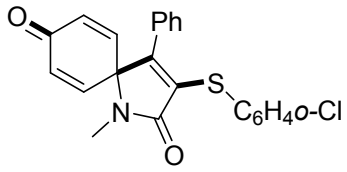
3-(4-chlorophenylthio)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dio,

Compound **5n** was obtained in 71% yield according to the general procedure (12h). Yellow solid; mp 180.7-181.3 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.35-7.32 (m, 1H), 7.29-7.26 (m, 2H), 7.24-7.19 (m, 4H), 7.15 (d, J = 8.7 Hz, 2H), 6.52-6.46 (m, 4H), 2.89 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.8, 167.5, 152.9, 144.9, 133.9, 133.3, 132.7, 132.0, 130.5, 130.0, 129.9, 129.1, 128.4, 128.1, 67.8, 26.3; HRMS calc. for $C_{22}H_{16}{}^{35}ClNO_2SNa$ $(M+Na)^+$, 416.0488; found, 416.0489.



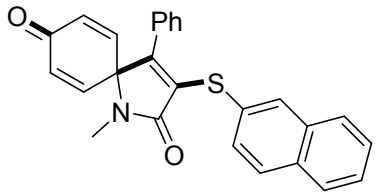
3-(3-chlorophenylthio)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione,

Compound **5o** was obtained in 54% yield according to the general procedure (12h). Yellow solid; mp 187.9-189.3 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.33-7.27 (m, 1H), 7.28-7.25 (m, 2H), 7.22-7.17 (m, 4H), 7.13-7.11 (m, 2H), 6.53 (d, J = 10.3 Hz, 2H), 6.48 (d, J = 10.3 Hz, 2H), 2.91 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.8, 167.4, 153.4, 144.8, 134.6, 133.5, 133.3, 131.5, 130.7, 130.4, 129.9, 129.0, 128.4, 128.0, 127.7, 67.9, 26.4; HRMS calc. for $C_{22}H_{16}{}^{35}ClNO_2SNa$ $(M+Na)^+$, 416.0488; found, 416.0486.

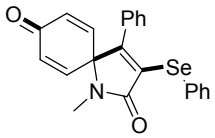


3-(2-chlorophenylthio)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione,

Compound **5p** was obtained in 53% yield according to the general procedure (12h). Yellow solid; mp 138.4-140.0 °C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.32-7.30 (m, 1H), 7.27-7.26 (m, 1H), 7.25-7.21 (m, 5H), 7.12-7.07 (m, 2H), 6.53 (d, J = 10.2 Hz, 2H), 6.46 (d, J = 10.2 Hz, 2H), 2.90 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 184.0, 167.4, 152.0, 145.1, 135.8, 133.2, 131.1, 130.5, 130.2, 130.0, 129.7, 129.0, 128.3, 127.9, 127.0, 67.9, 26.3; HRMS calc. for $C_{22}H_{16}{}^{35}ClNO_2SNa$ $(M+Na)^+$, 416.0488; found, 416.0489.



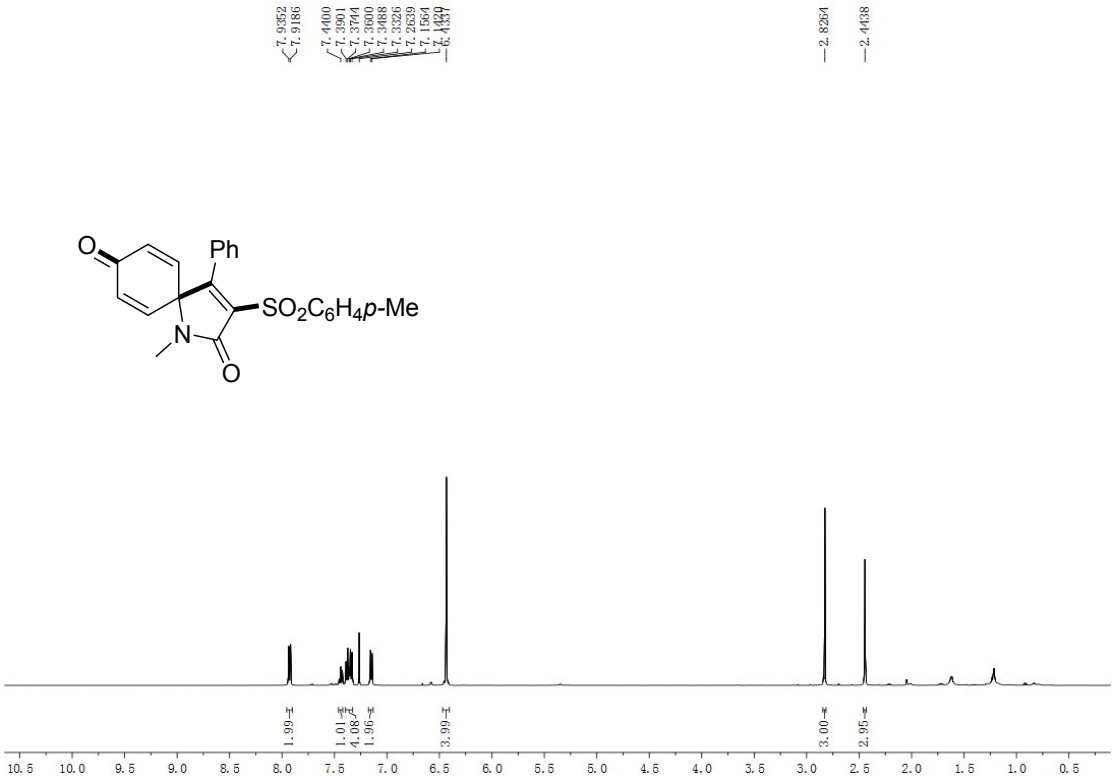
1-methyl-3-(naphthalen-2-ylthio)-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5q** was obtained in 60% yield according to the general procedure (12h). Yellow solid; mp 153.4-154.0 ^{o}C. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.79 (d, J = 1.5 Hz, 1H), 7.74-7.72 (m, 1H), 7.68-7.66 (m, 1H), 7.63 (d, J = 8.6 Hz, 1H), 7.45-7.43 (m, 2H), 7.34-7.32 (m, 1H), 7.23-7.16 (m, 5H), 6.54 (d, J = 10.2 Hz, 2H), 6.46 (d, J = 10.2 Hz, 2H), 2.89 (s, 3H); ^{13}C NMR ($CDCl_3$, 125 MHz, ppm): δ 183.9, 167.8, 152.5, 145.1, 133.4, 133.2, 132.5, 132.4, 130.7, 130.6, 130.0, 128.7, 128.6, 128.4, 128.2, 128.1, 127.7, 127.5, 126.5, 126.4, 67.8, 26.3; HRMS calc. for $C_{26}H_{19}NO_2SNa$ $(M+Na)^+$, 432.1034; found, 432.1037.

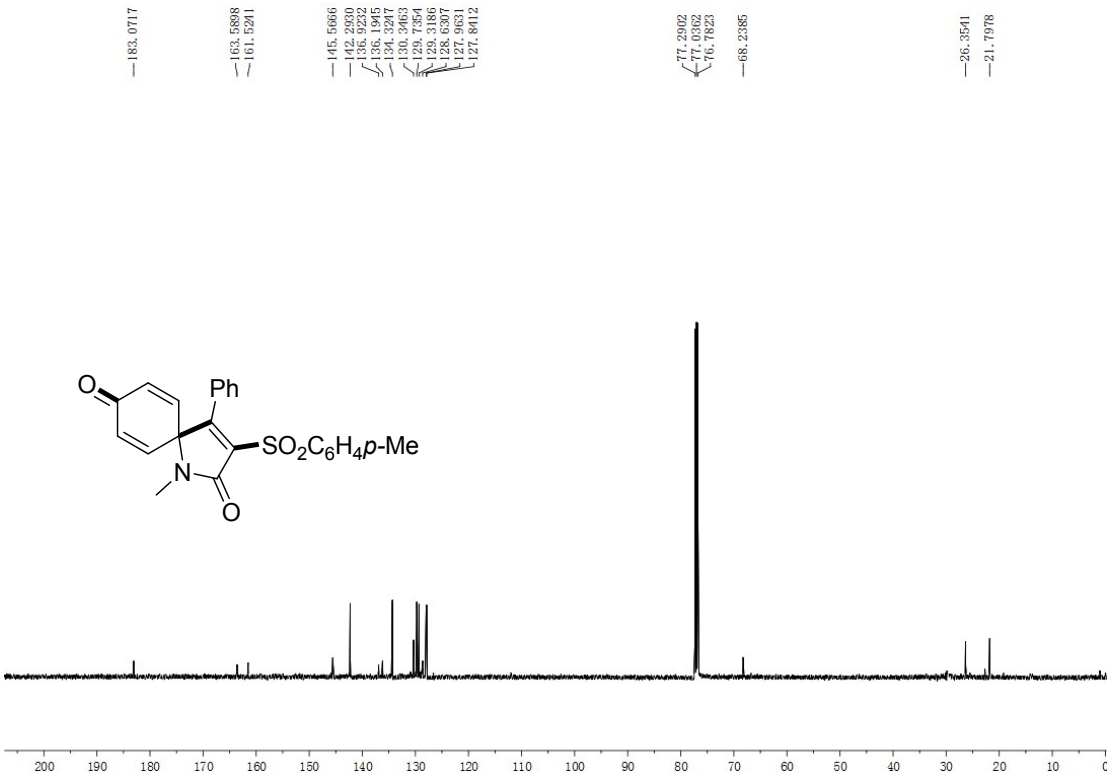


1-methyl-4-phenyl-3-(phenylselanyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione, Compound **5r** was obtained in 63% yield according to the general procedure (12h). White solid; mp:128.1-129.4. ^{1}H NMR ($CDCl_3$, 500 MHz, ppm): δ 7.31 (d, J = 7.3 Hz, 2H), 7.18 (t, J = 7.4 Hz, 1H), 7.13-7.08 (m, 3H), 7.05-7.01 (m, 4H), 6.43 (d, J = 10.2 Hz, 2H), 6.35 (d, J = 10.2 Hz, 2H), 2.83 (s, 3H); ^{13}C NMR ($CDCl_3$, 125MHz, ppm): δ 182.9, 167.8, 153.1, 144.0, 132.9, 132.1, 130.2, 129.2, 128.4, 128.0, 127.2, 127.0, 126.9, 126.1, 68.0, 25.4; HRMS calc. for $C_{22}H_{17}NO_2SeNa$ $(M+Na)^+$, 430.0322; found, 430.0318.

6.Copies of NMR spectra for 3a−5r

3a

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127.8412
77.2902
77.0362
76.7823
68.2385
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O
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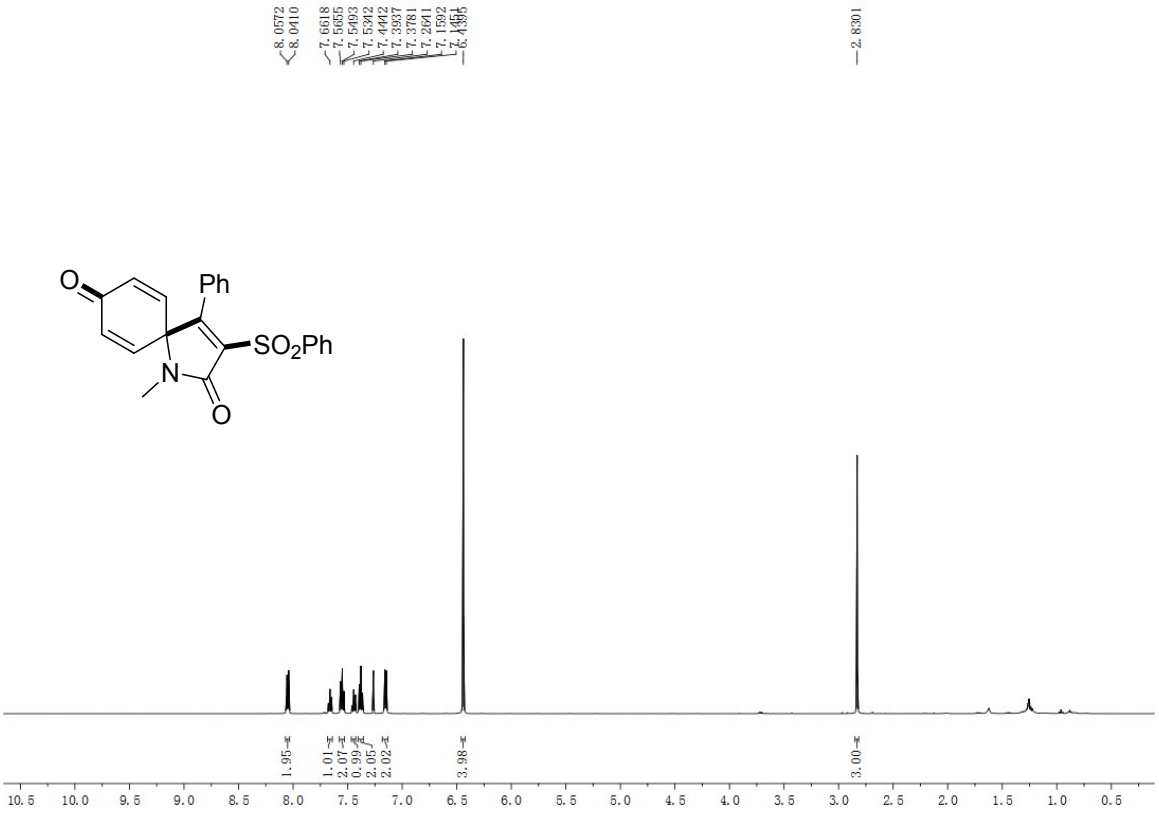
3b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7.4442
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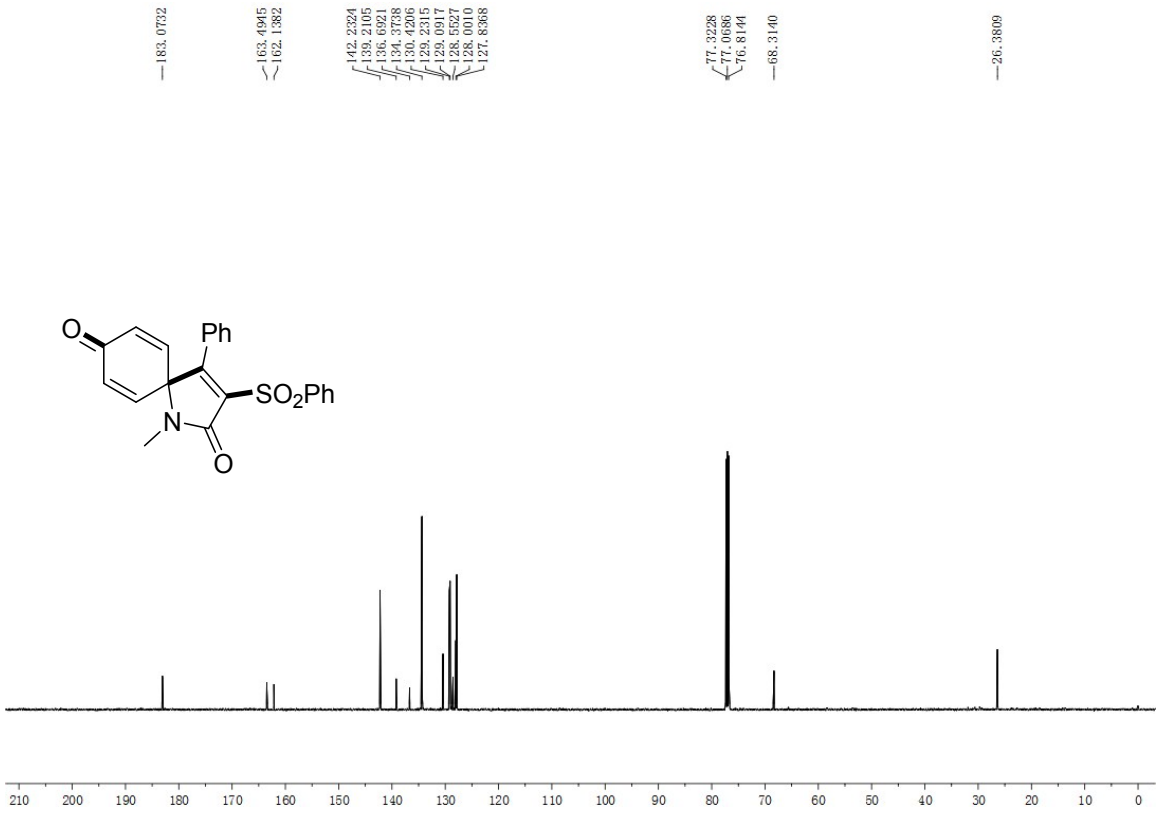
7.3781
7.2661
7.1592
7.1451
6.4395
2.8301
O
Ph
SO2Ph
N
O
1.95
1.01
2.07
0.99
2.05
2.02
3.98
3.00
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5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139.2105
136.6921
134.3738
130.4206
129.2315
129.0917
128.5527
128.0010
127.8368
77.3228
77.0686
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Ph
SO2Ph
N
O
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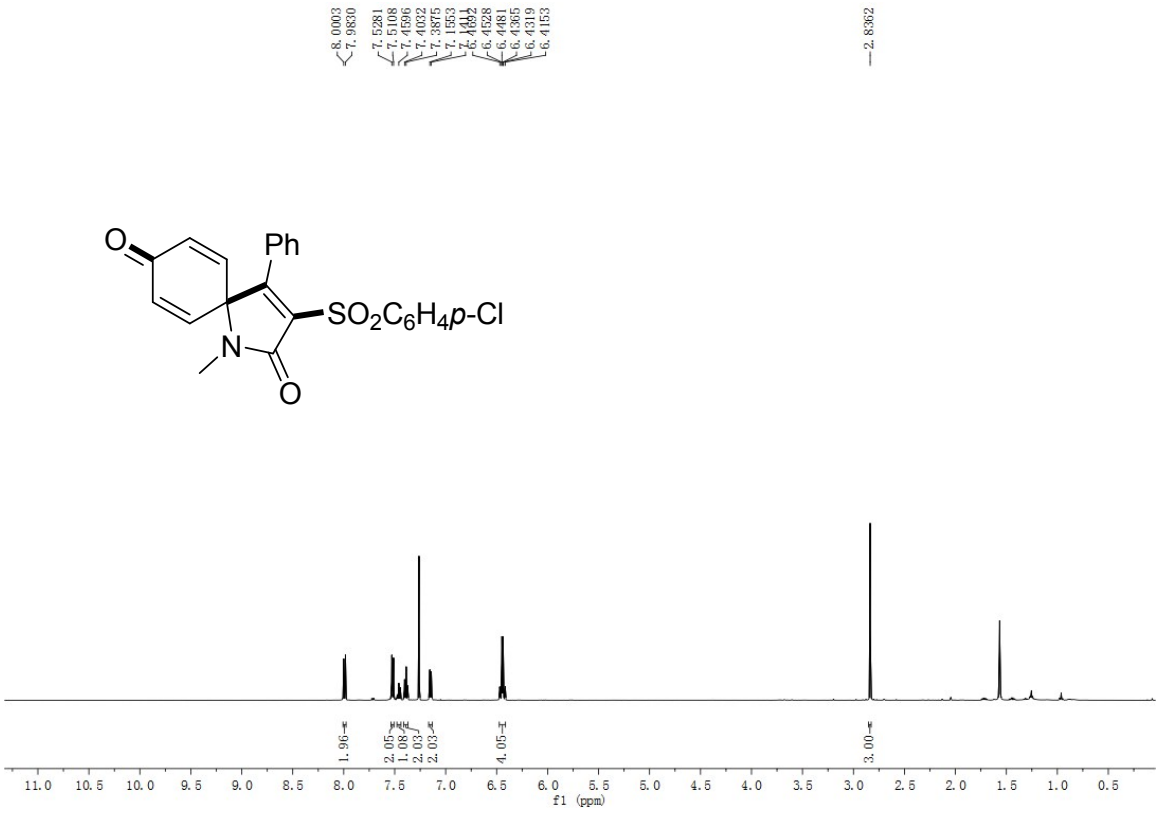
3c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6.4365
6.4319
6.4153
2.8362
O
Ph
SO2C6H4p-Cl
N
O
1.96
2.05
1.08
2.03
2.03
4.05
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
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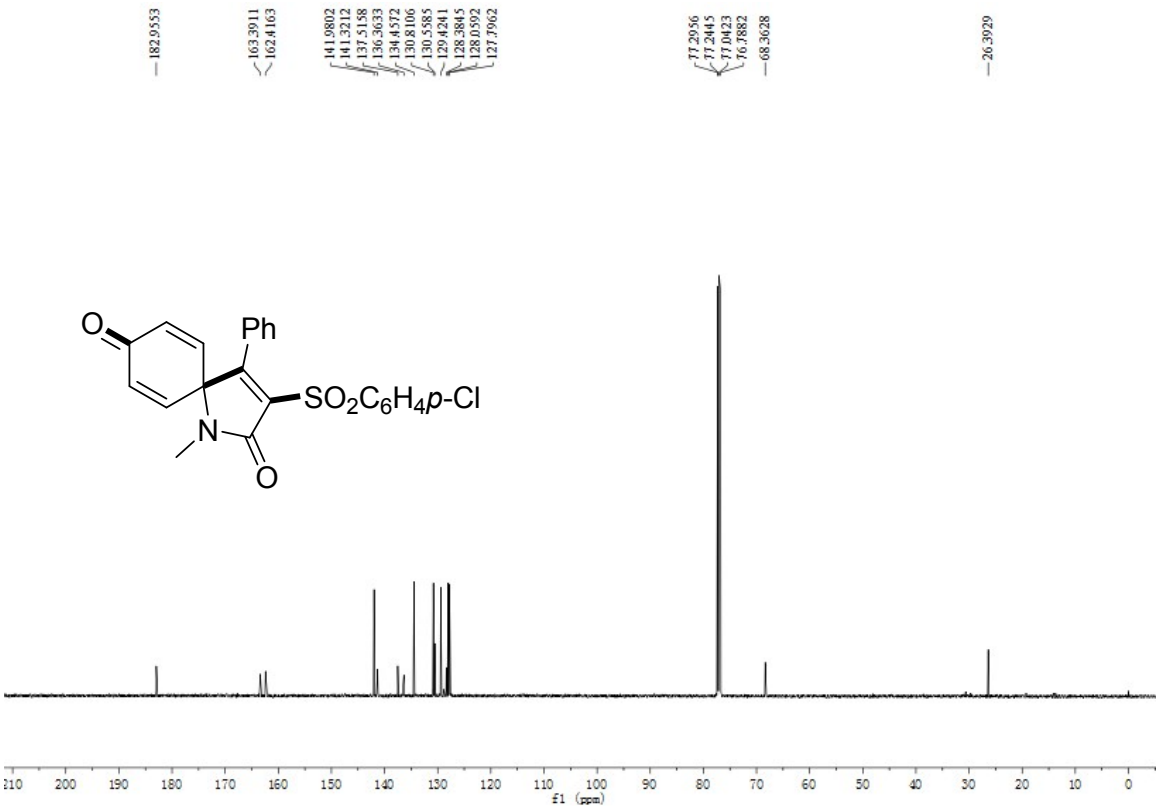
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136.3633
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130.8106
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128.3845
128.0592
127.7962
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77.2445
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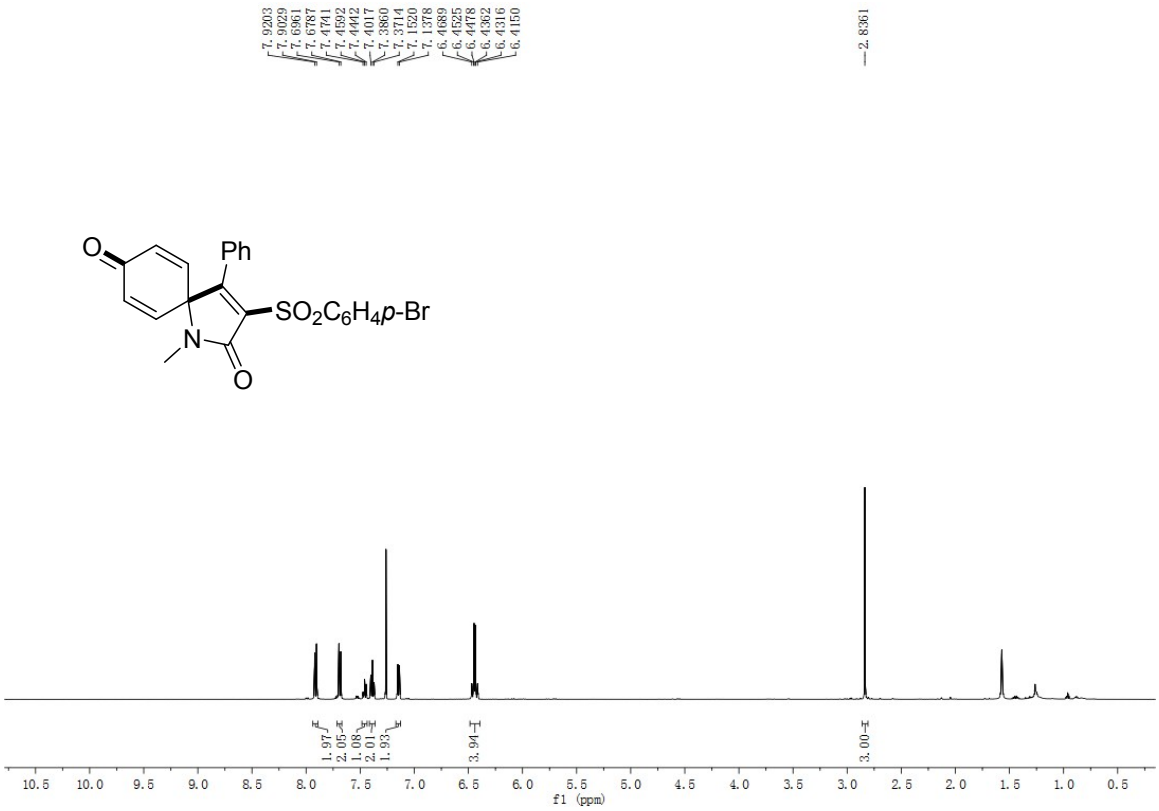
N
O
f1 (ppm)

3d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7.1520
7.1378
6.4689
6.4525
6.4478
6.4362
6.4316
6.4150
2.8361
O
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O
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1.08
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1.93
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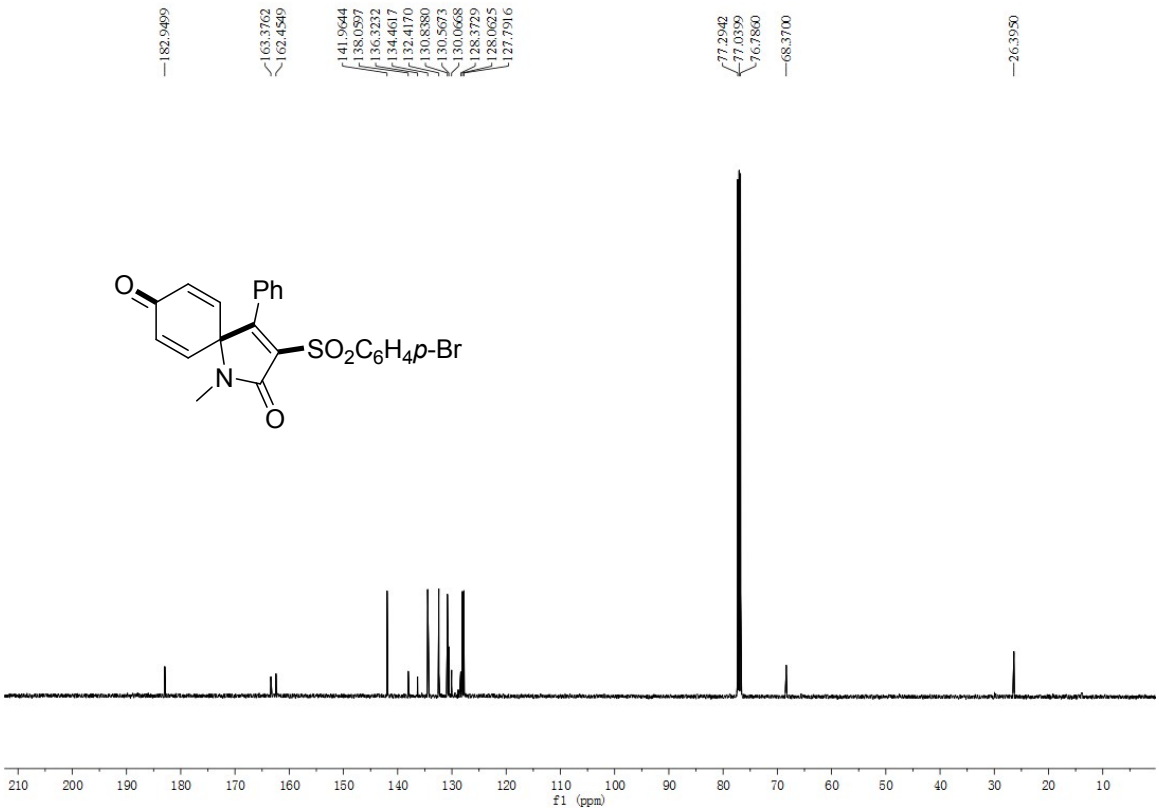
3.00
f1 (ppm)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128.0625
127.7916
77.2942
77.0399
76.7860
68.3700
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N
O
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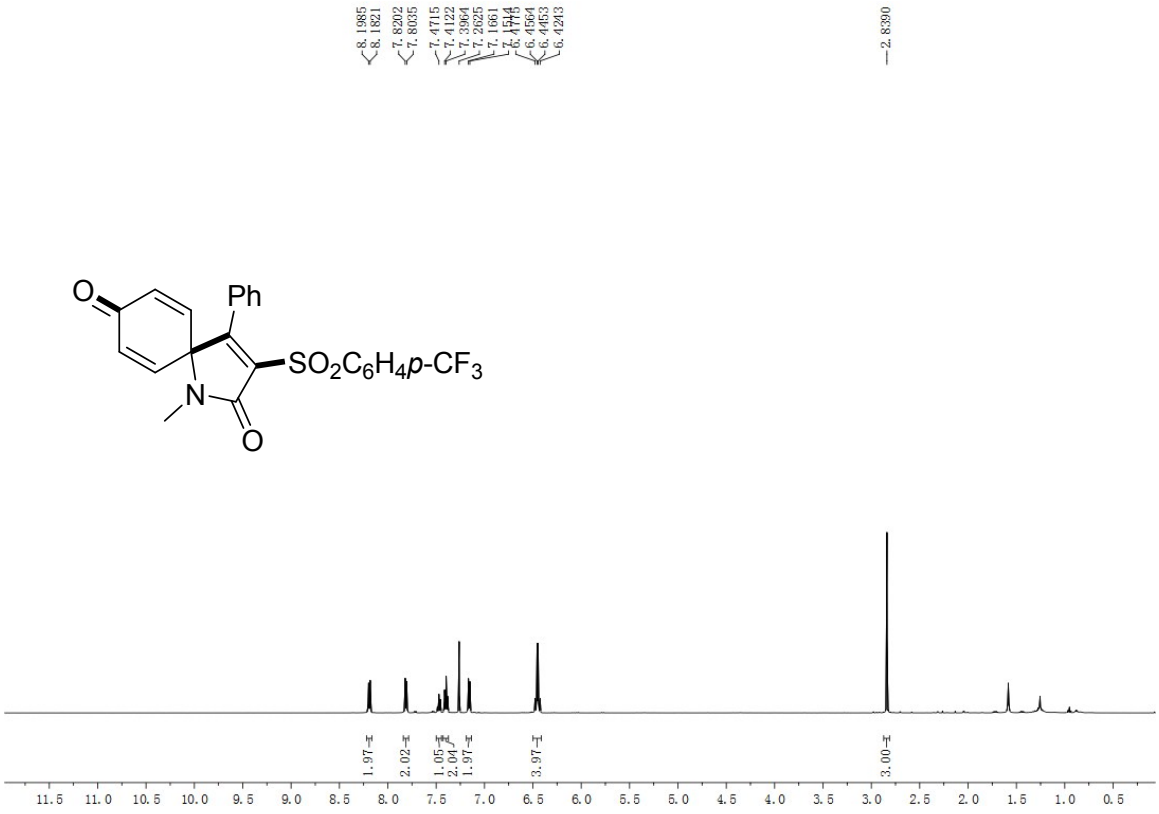
f1 (ppm)

3e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O
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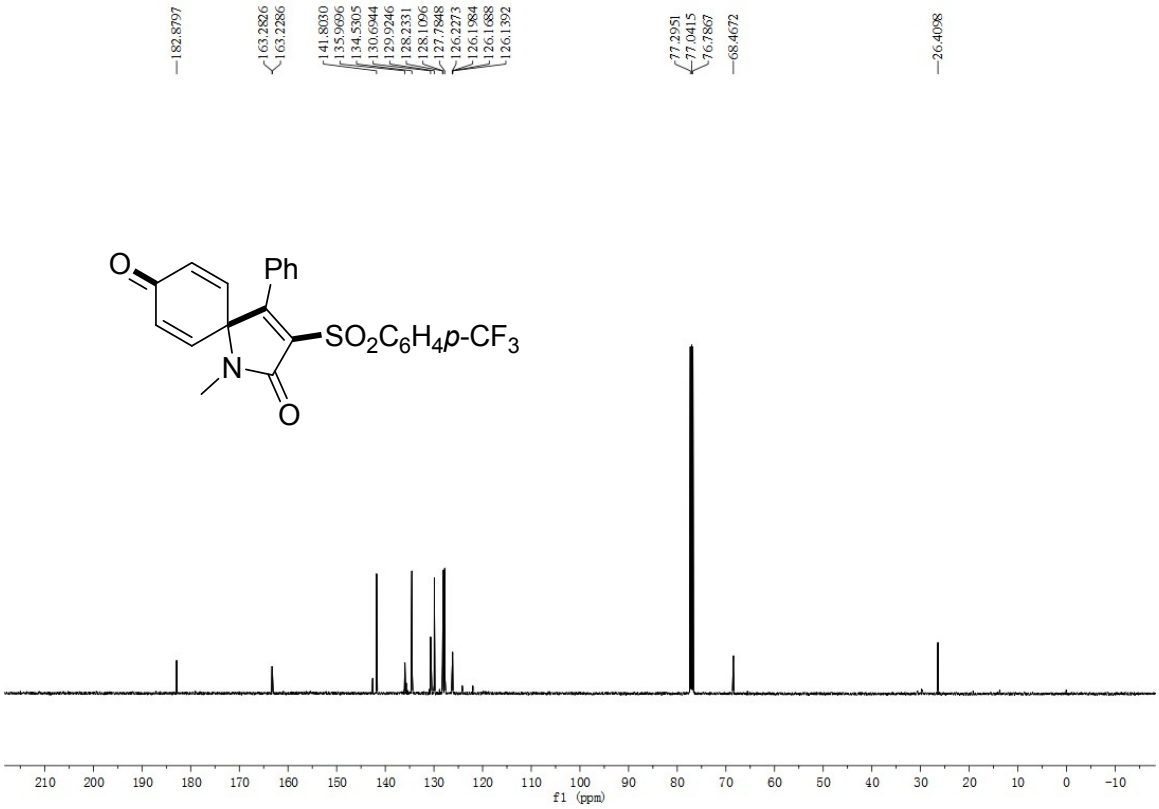
2.02
1.05
2.04
1.97
3.97
3.00
11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5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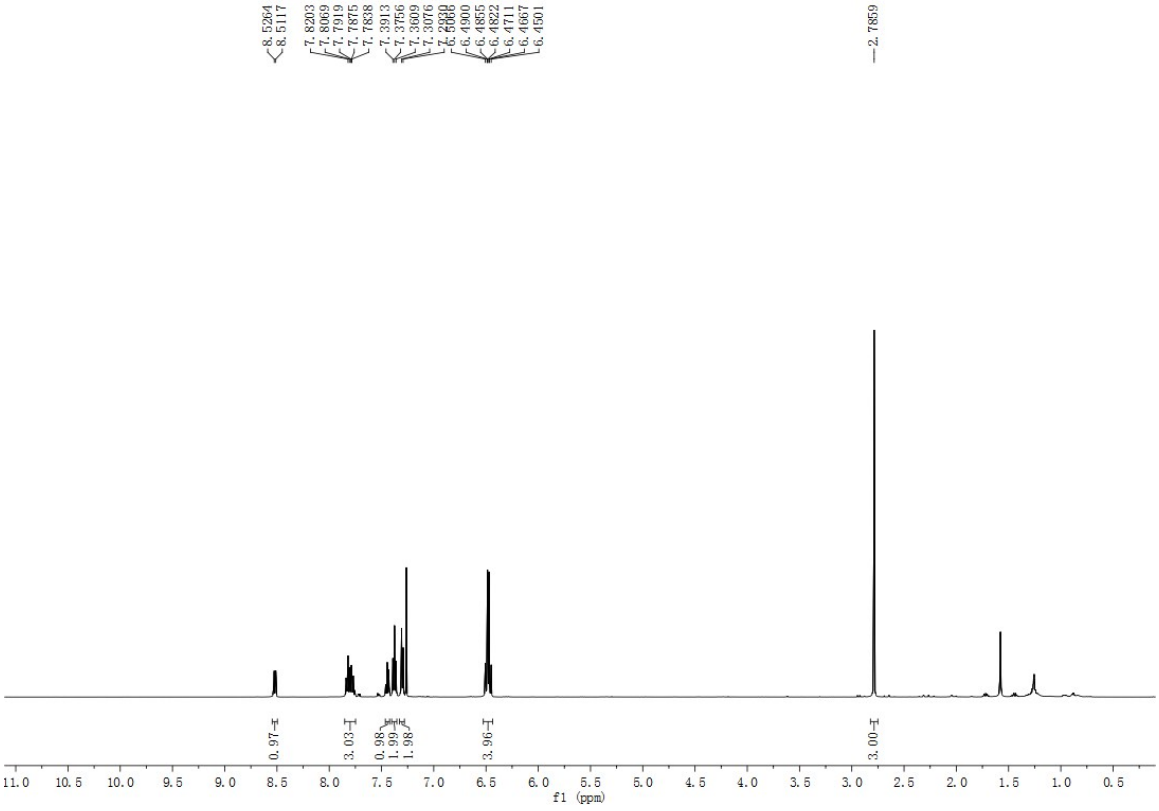
N
O
f1 (ppm)

3f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6.4855
6.4822
6.4711
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1.98
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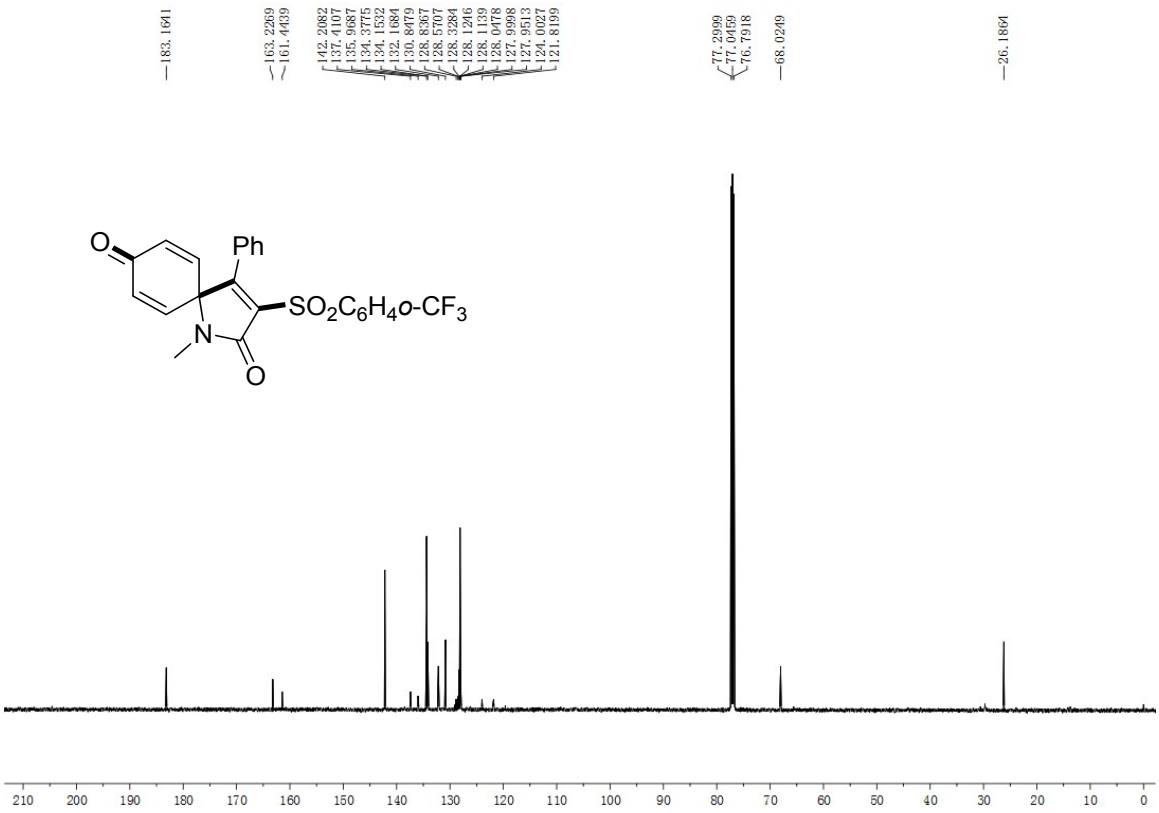
3.00
f1 (ppm)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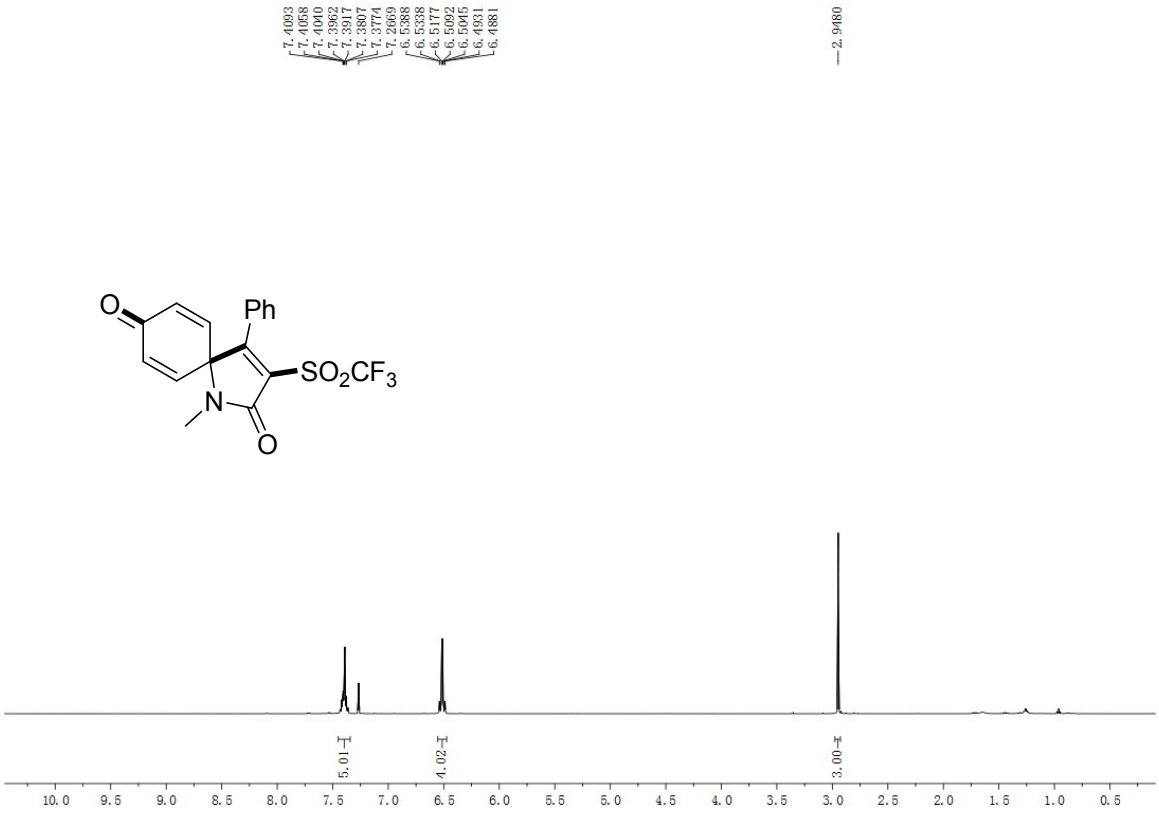
3g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O
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4.02
3.00
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9.5
9.0
8.5
8.0
7.5
7.0
6.5
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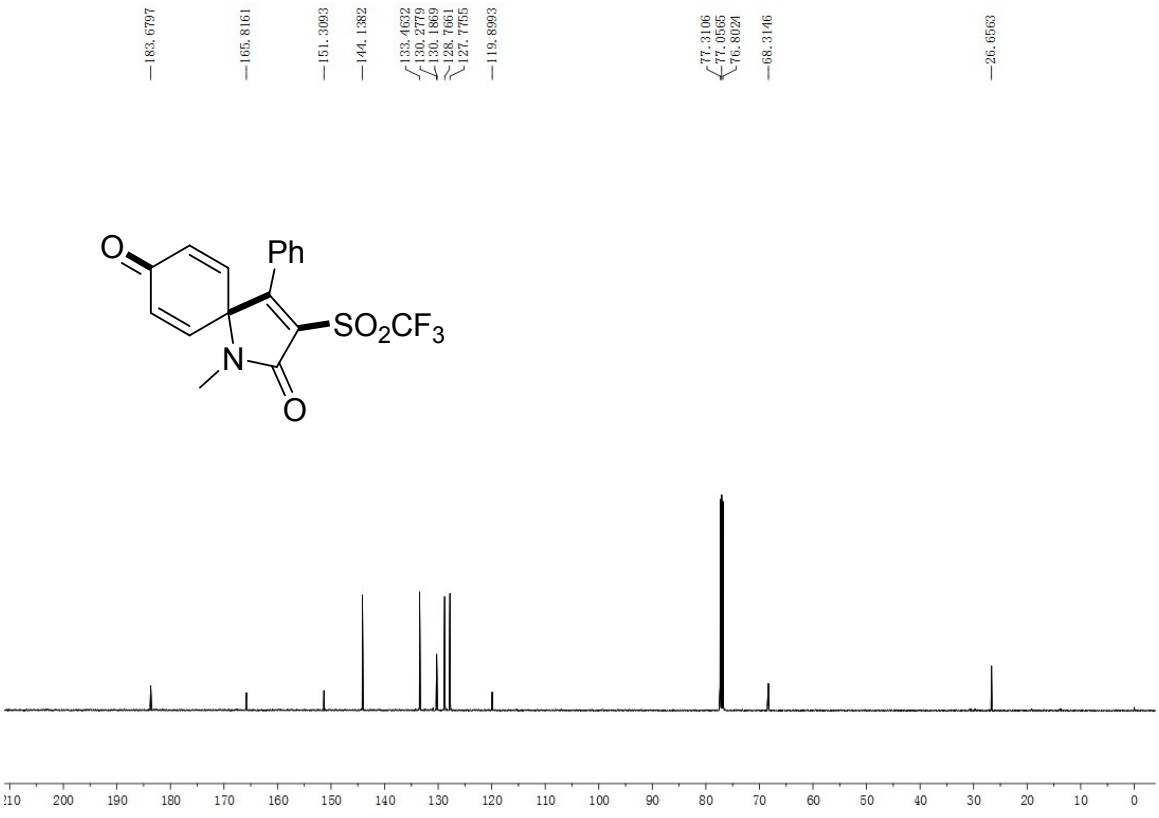
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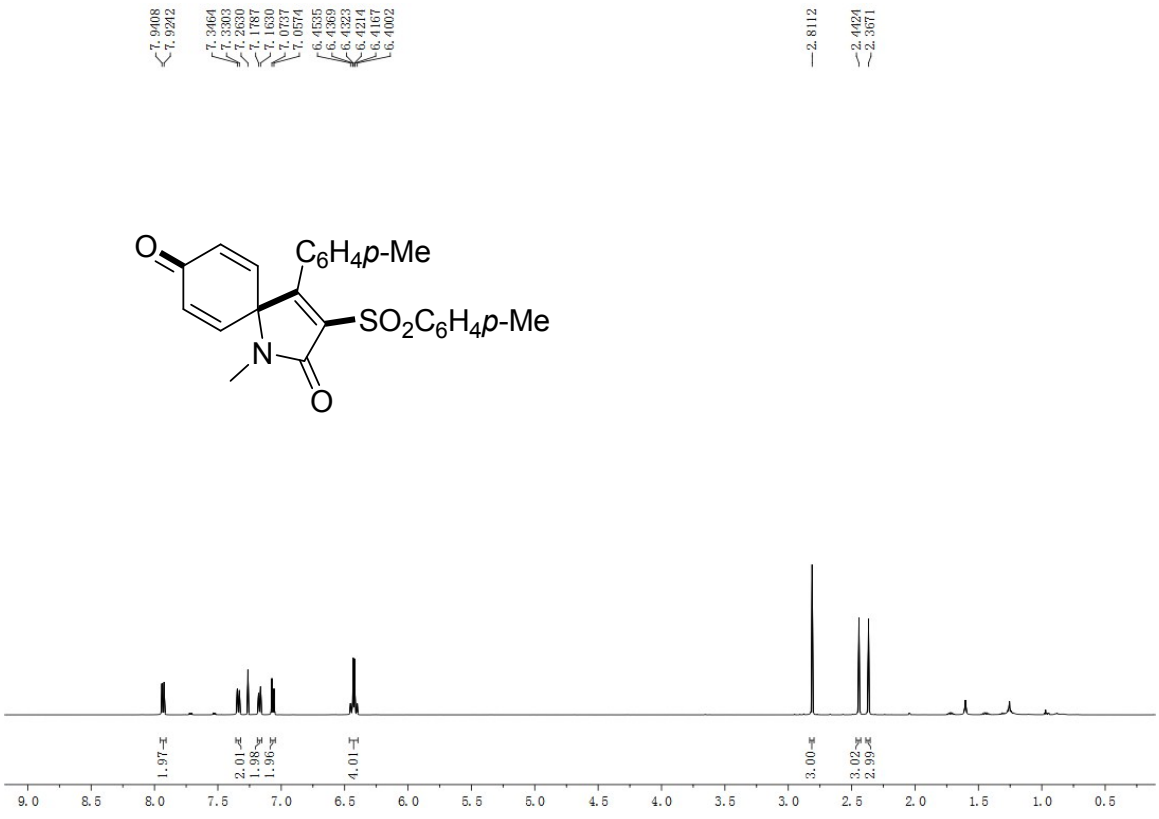
N
O

3h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6.4214
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1.96
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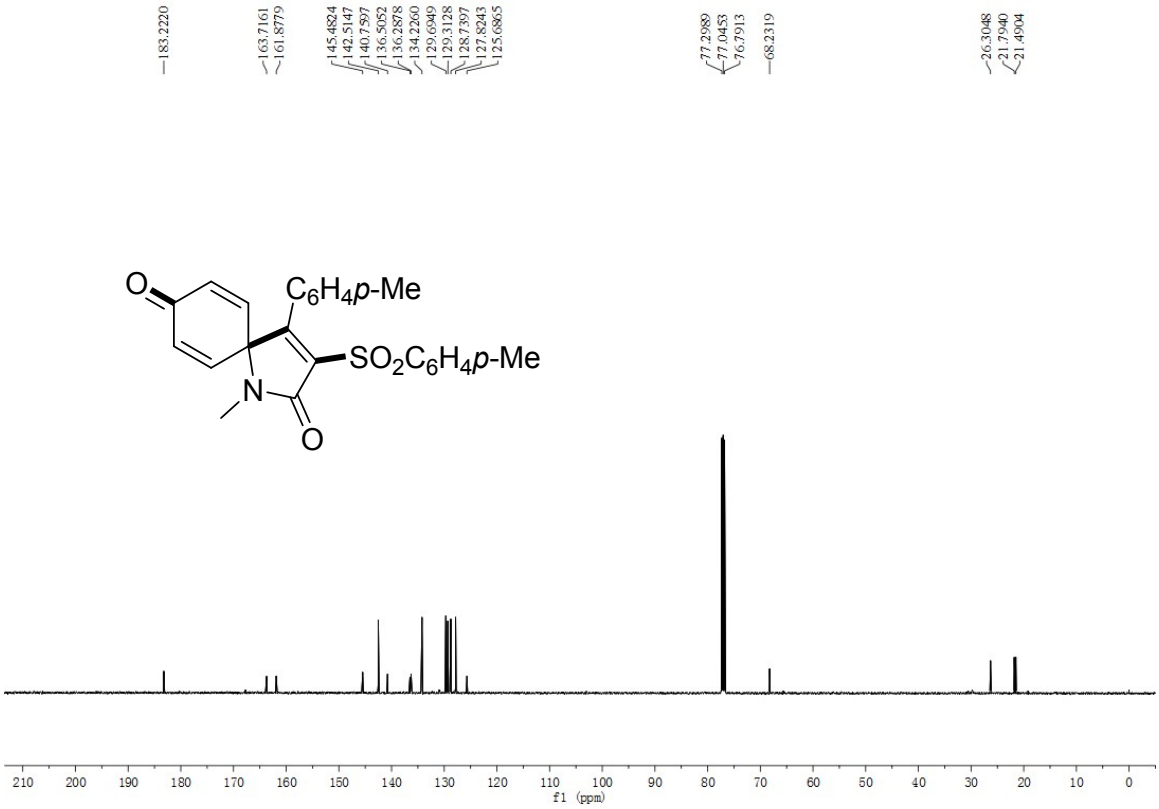
3.02
2.99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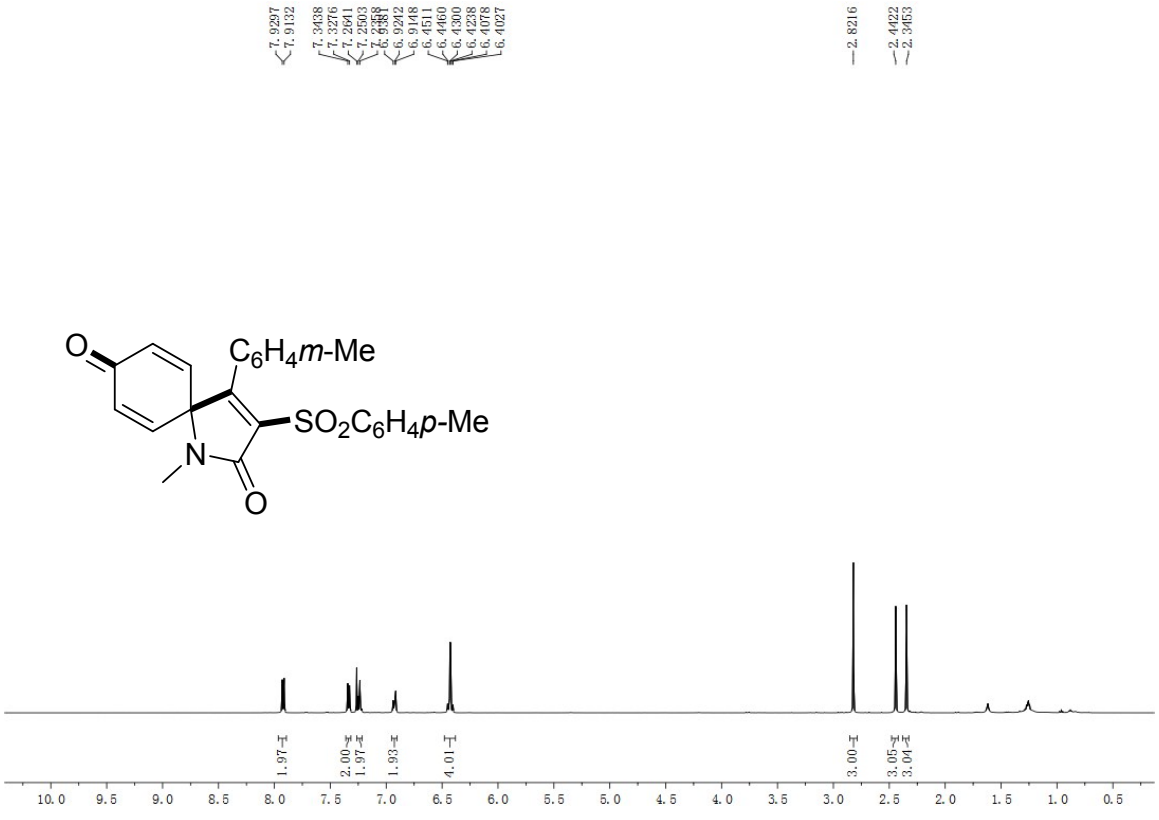
3i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6.4078
6.4027
2.8216
2.4422
2.3453
O
C6H4m-Me
SO2C6H4p-Me
N
O
1.97
2.00
1.97
1.93
4.01
3.00
3.05
3.04
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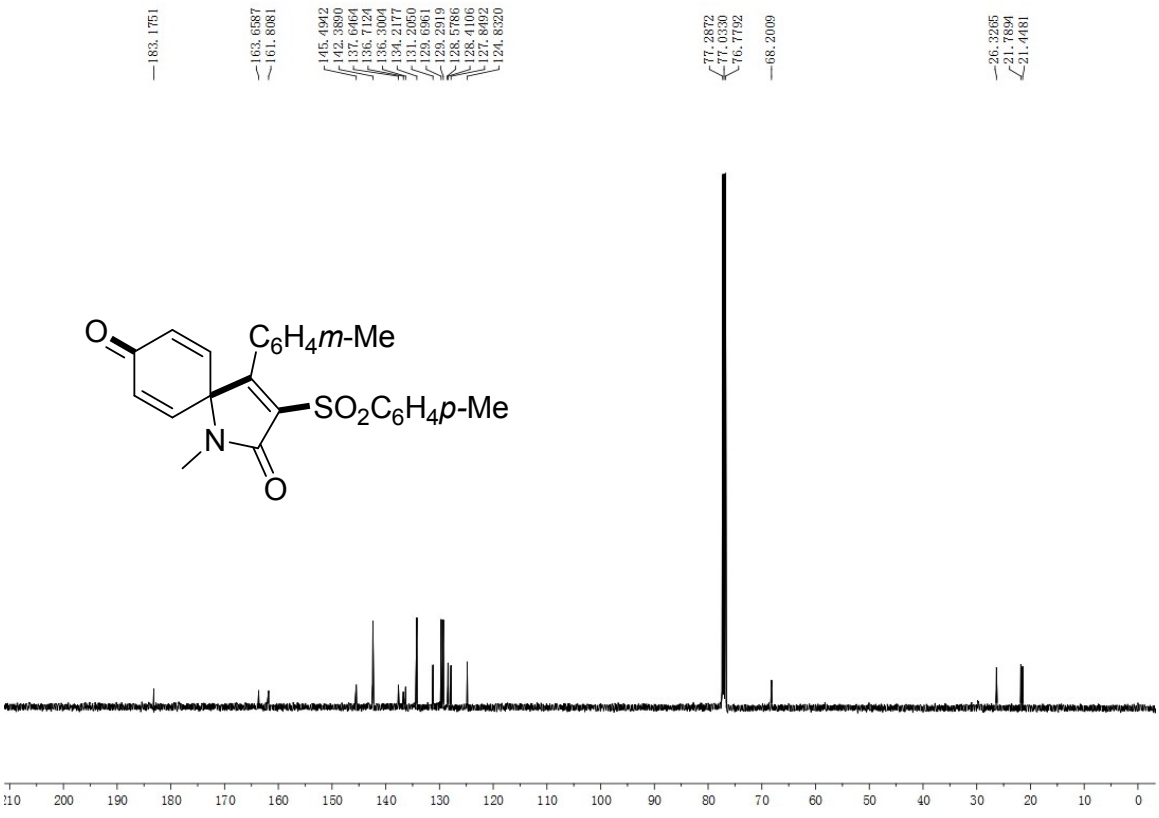
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129.6961
129.2919
128.5786
128.4106
127.8492
124.8320
77.2872
77.0330
76.7792
68.2009
26.3265
21.7894
21.4481
O
C6H4m-Me
SO2C6H4p-Me
N
O
210
200
190
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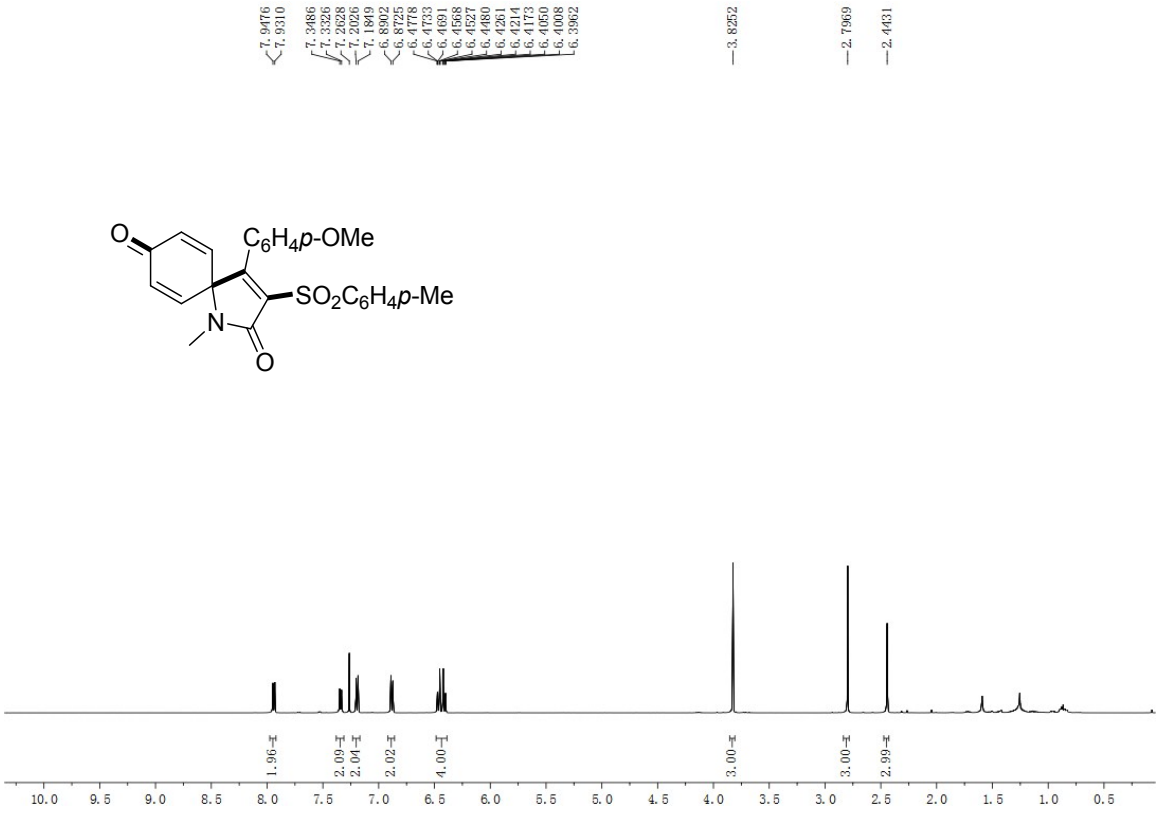
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7.1849
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6.4733
6.4691
6.4568
6.4527
6.4480
6.4261
6.4214
6.4173
6.4050
6.4008
6.3962
3.8252
2.7969
2.4431
O
C6H4p-OMe
SO2C6H4p-Me
N
O
1.96
2.09
2.04
2.02
4.00
3.00
3.00
2.99
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
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5.0
4.5
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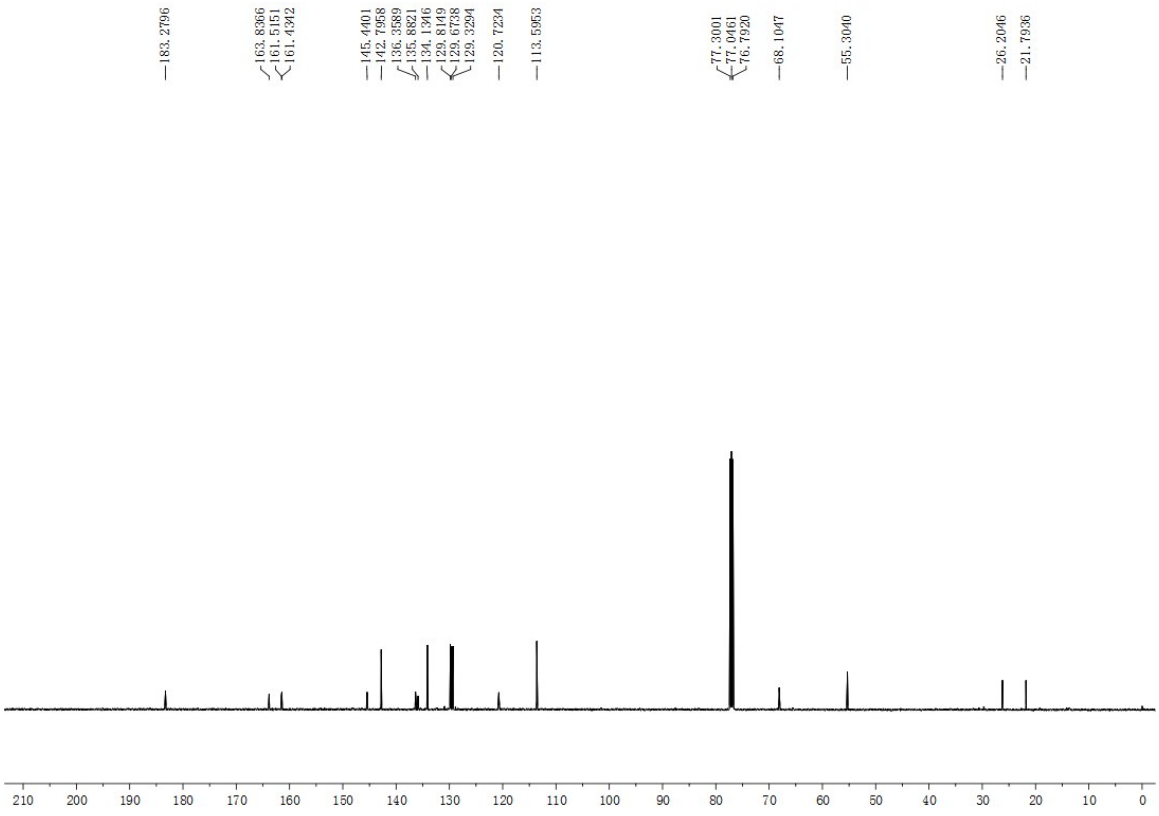
3.5
3.0
2.5
2.0
1.5
1.0
0.5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76.7920
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26.2046
21.7936
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200
190
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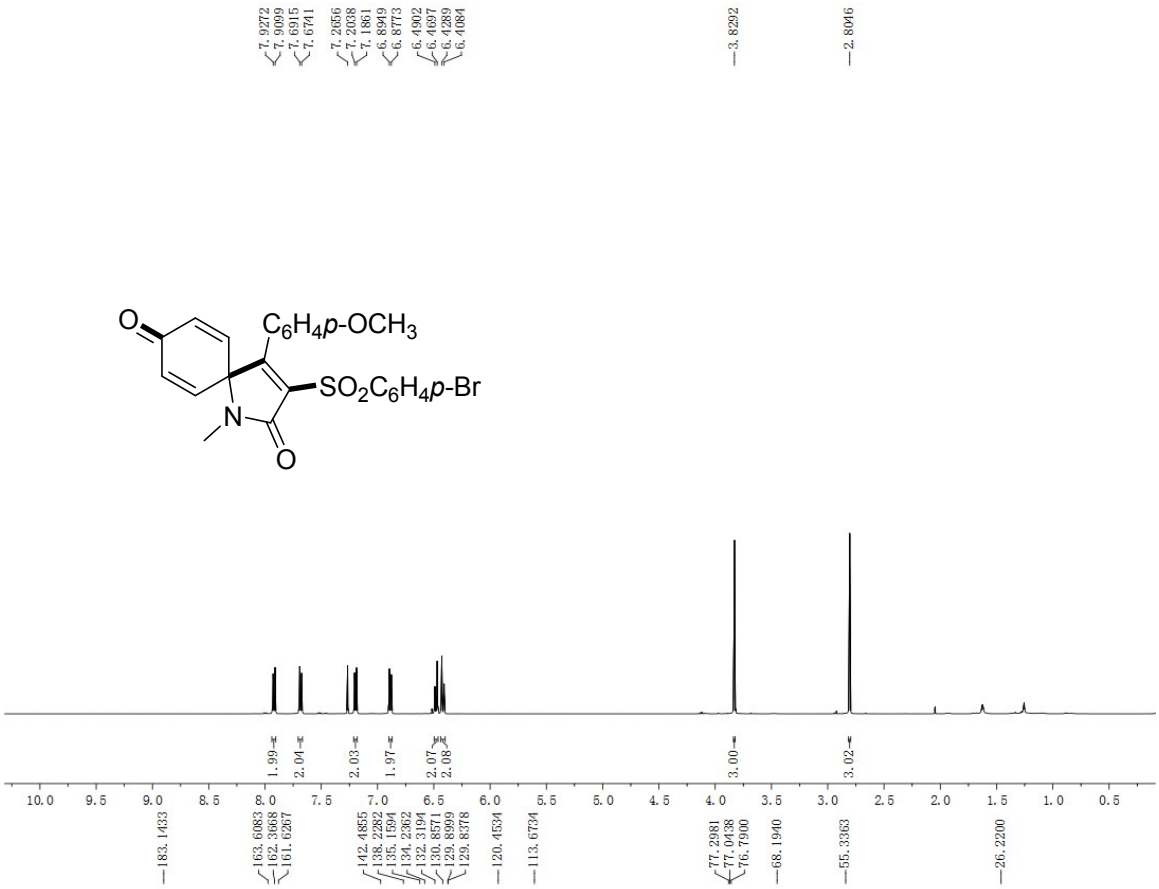
3k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7.1861
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6.4697
6.4289
6.4084
3.8292
2.8046
C6H4p-OCH3
SO2C6H4p-Br
N
O
O
1.99
2.04
2.03
1.97
2.07
2.08
3.00
3.02
183.1433
163.6083
162.3668
161.6267
142.4855
138.2282
135.1594
134.2362
132.3194
130.8571
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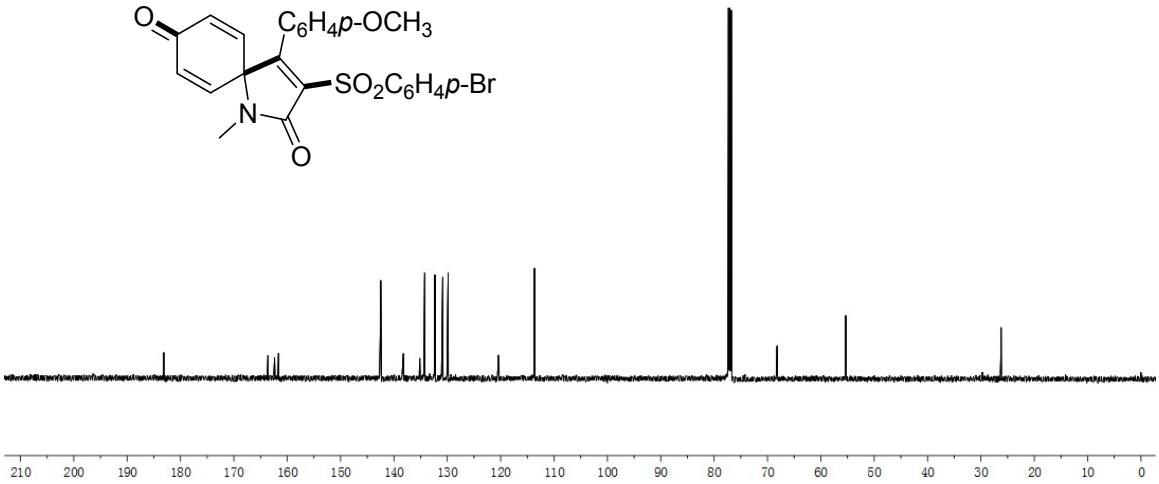
76.7900
68.1940
55.3363
26.2200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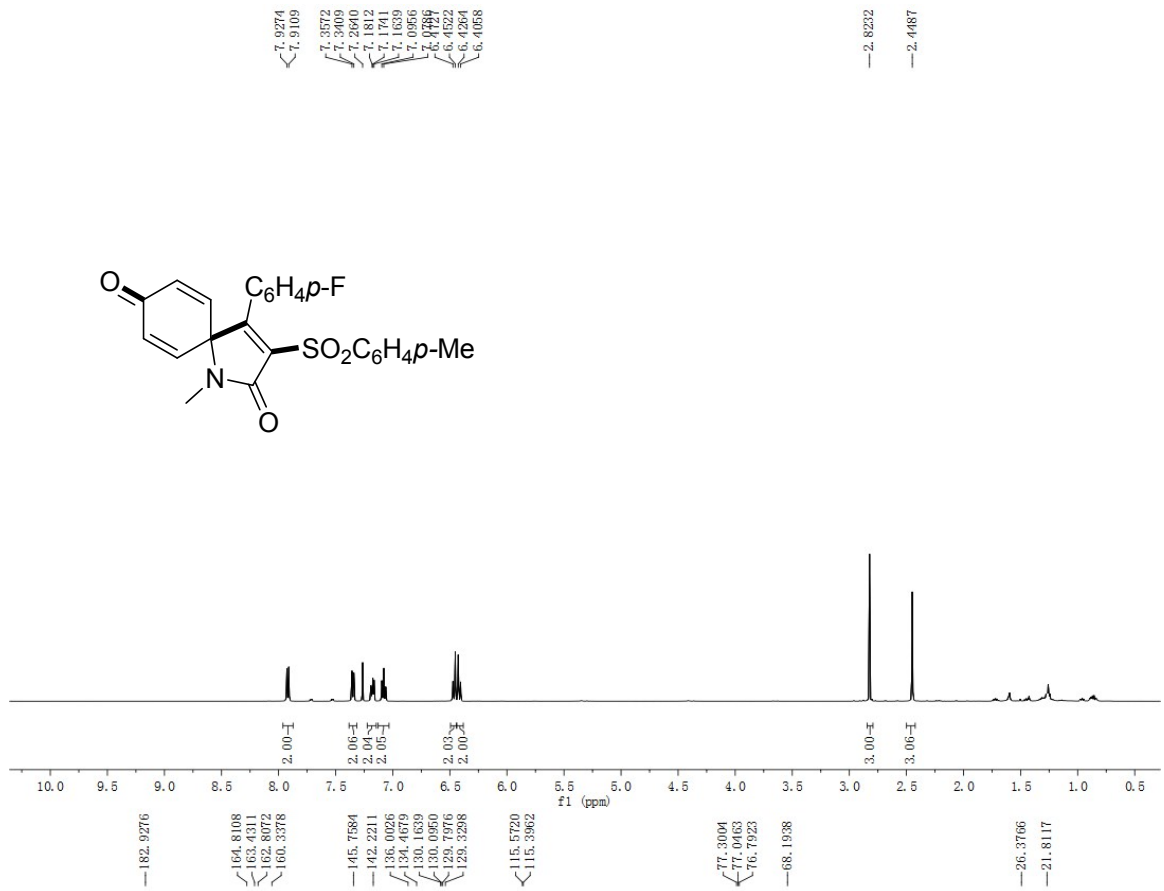
SO2C6H4p-Br
N
O
O

3l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7.0956
6.4522
6.4264
6.4058
2.8232
2.4487
O
C6H4p-F
SO2C6H4p-Me
N
O
2.00
2.06
2.04
2.05
2.03
2.00
3.00
3.06
f1 (ppm)
182.9276
164.8108
163.4311
162.8072
160.3378
145.7584
142.2211
136.0026
134.4679
130.1639
130.0950
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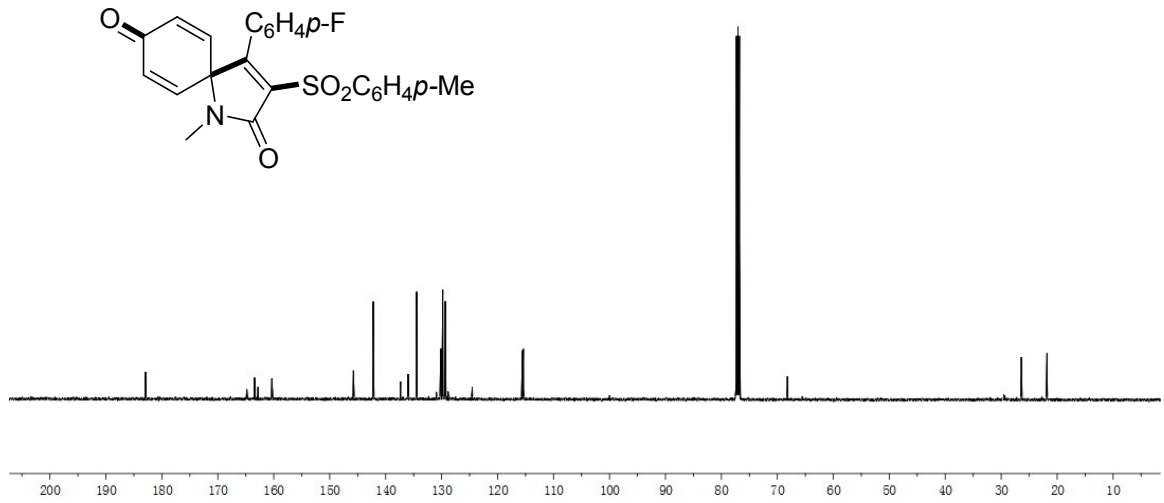
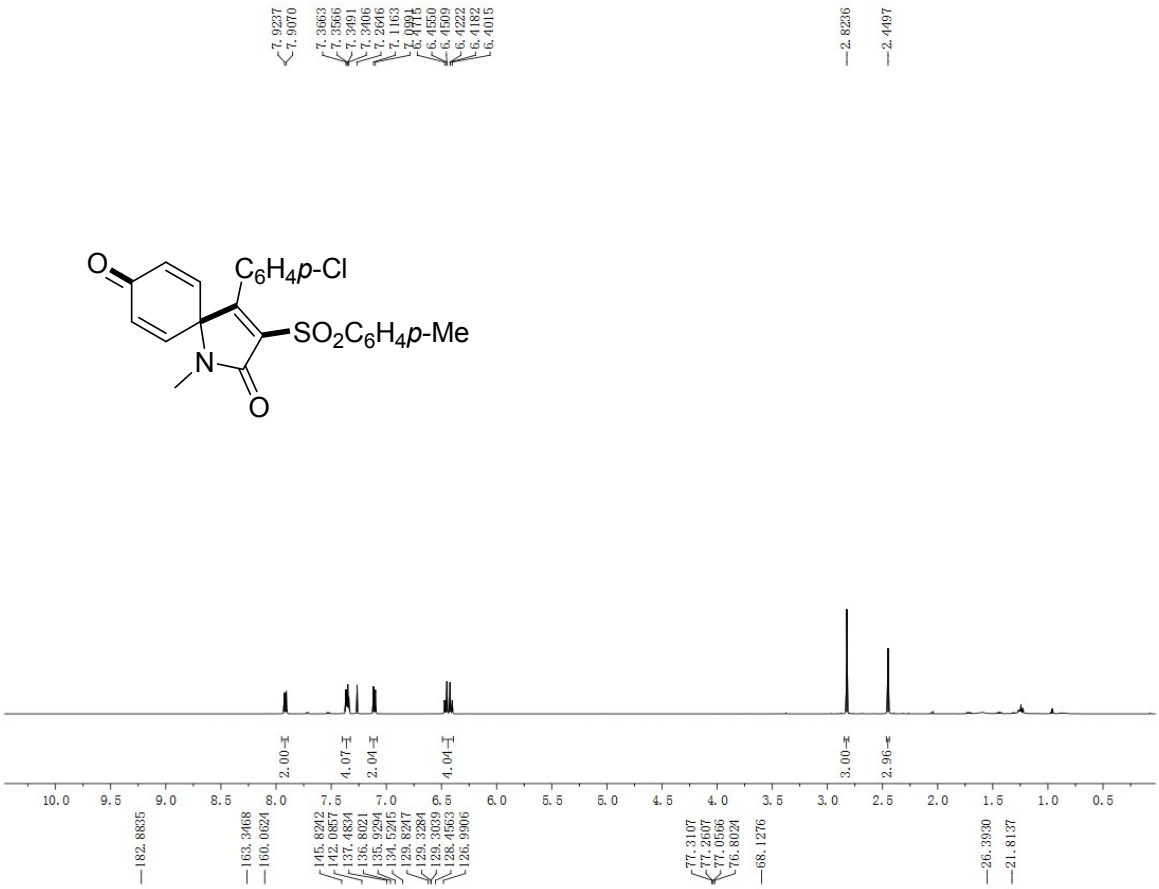
26.3766
21.8117


O
C6H4p-F
SO2C6H4p-Me
N
O

3m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6.4509
6.4222
6.4182
6.4015
2.8236
2.4497
O
C6H4p-Cl
SO2C6H4p-Me
N
O
2.00
4.07
2.04
4.04
3.00
2.96
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
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163.3468
160.0624
145.8242
142.0857
137.4834
136.8021
135.9294
134.5245
129.8247
129.3284
129.3039
128.4563
126.9906
77.3107
77.2607
77.0566
76.8024
68.1276
26.3930
21.8137
O
C6H4p-Cl
SO2C6H4p-Me
N
O
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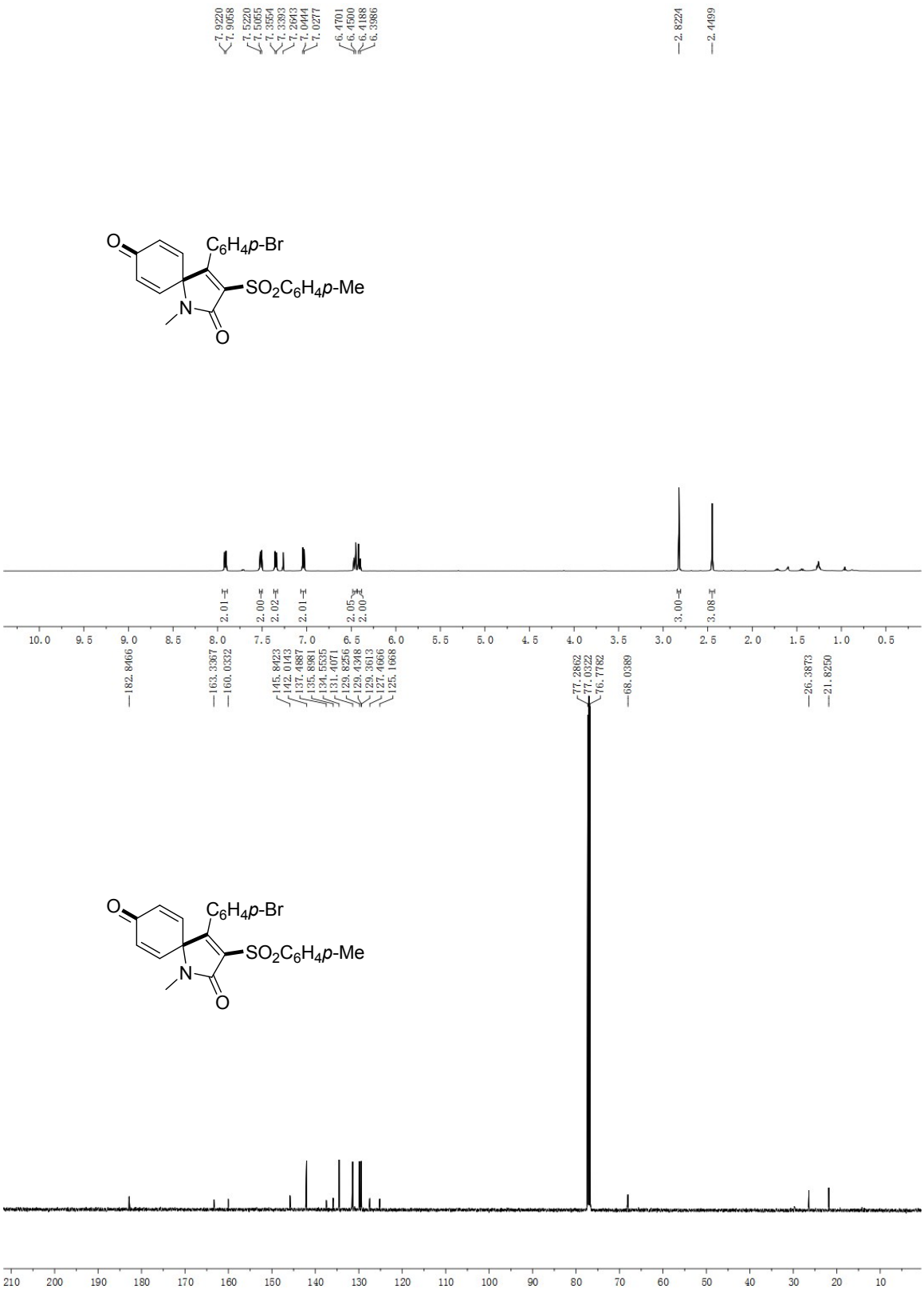
3n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7.3554
7.3393
7.2643
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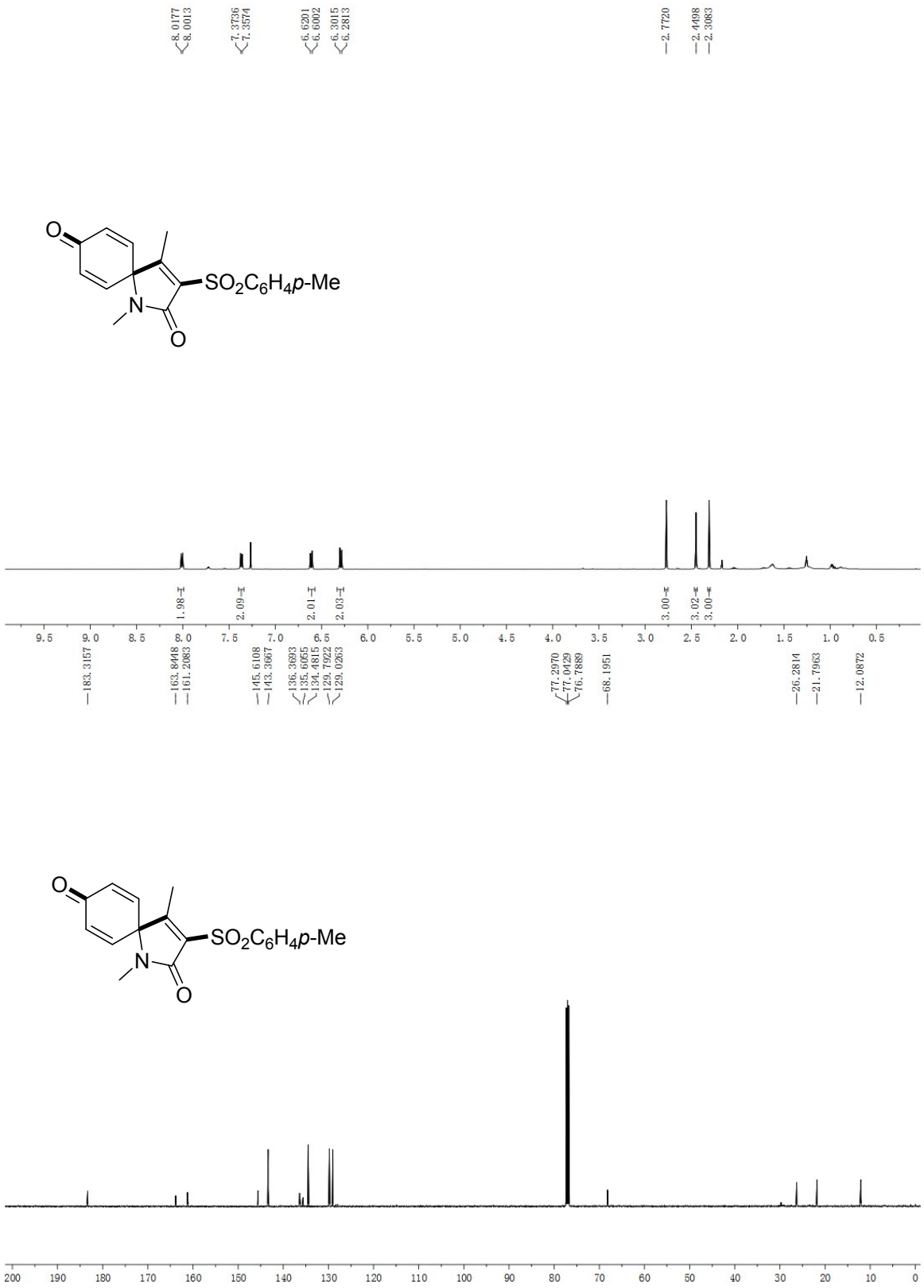
7.0277
6.4701
6.4500
6.4188
6.3986
2.8224
2.4499
O
C6H4p-Br
SO2C6H4p-Me
N
O
2.01
2.00
2.02
2.01
2.05
2.00
3.00
3.08
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
182.8466
163.3367
160.0332
145.8423
142.0143
137.4887
135.8981
134.5535
131.4071
129.8256
129.4348
129.3613
127.4966
125.1668
77.2862
77.0322
76.7782
68.0389
26.3873
21.8250
O
C6H4p-Br
SO2C6H4p-Me
N
O
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10

3o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6.6002
6.3015
6.2813
2.7720
2.4498
2.3083
O
N
O
SO2C6H4p-Me
1.98
2.09
2.01
2.03
3.00
3.02
3.00
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
183.3157
163.8948
161.2083
145.6108
143.3667
136.3693
135.6055
134.4815
129.7922
129.0263
77.2970
77.0429
76.7889
68.1951
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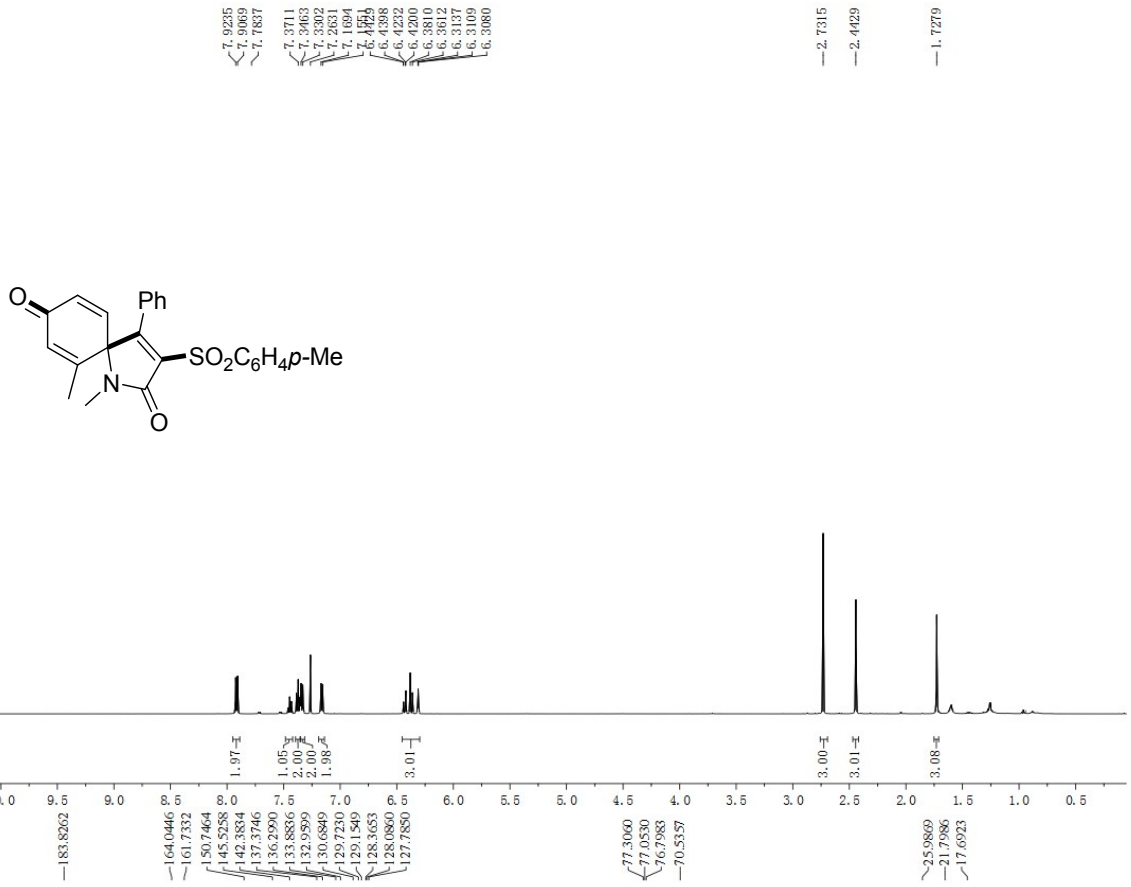
21.7963
12.0872
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0

3p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7.3463
7.3302
7.2631
7.1694
6.4398
6.4232
6.4200
6.3810
6.3612
6.3137
6.3109
6.3080
2.7315
2.4429
1.7279
Ph
SO2C6H4p-Me
N
O
1.97
1.05
2.00
2.00
1.98
3.01
3.00
3.01
3.08
183.8262
164.0446
161.7332
150.7464
145.5258
142.3834
137.3746
136.2990
133.8836
132.9999
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128.3653
128.0860
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77.0530
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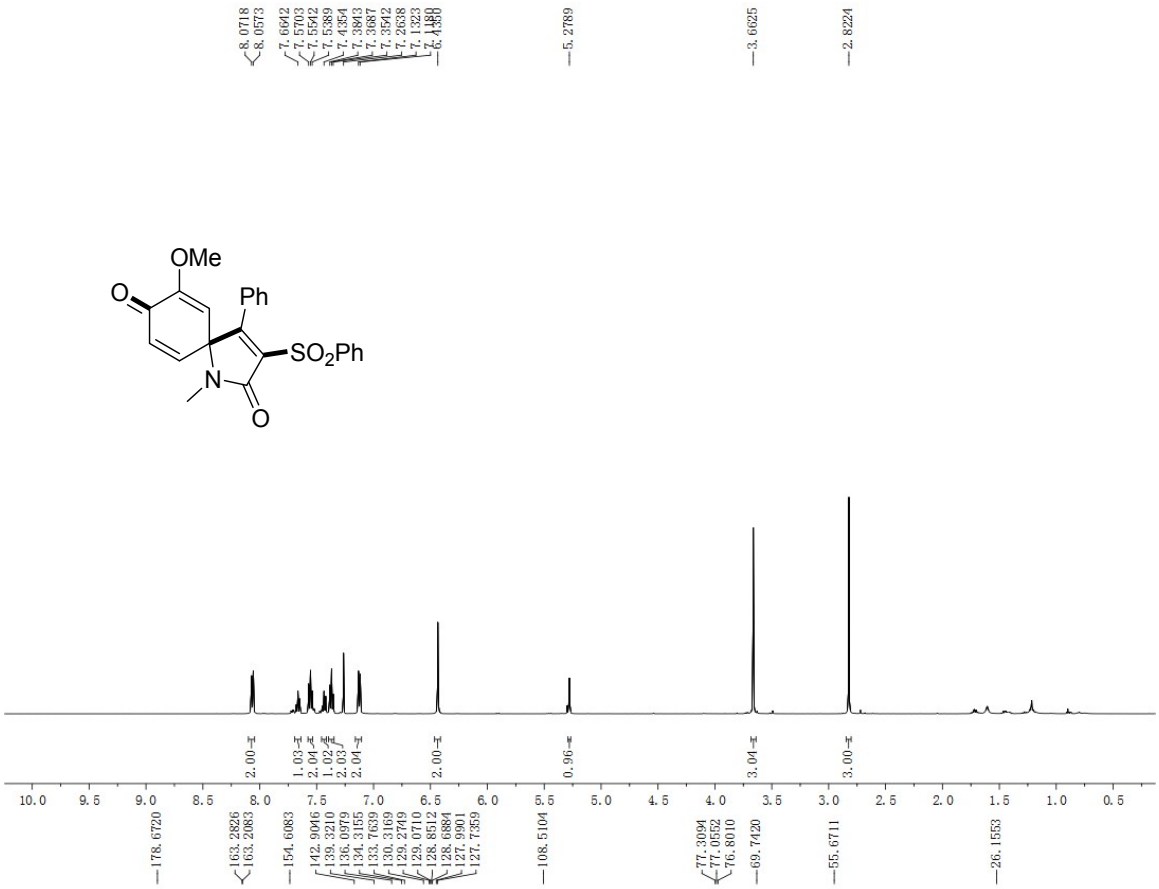
21.7986
17.6923
f1 (ppm)

3q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Ph
SO2Ph
N
O
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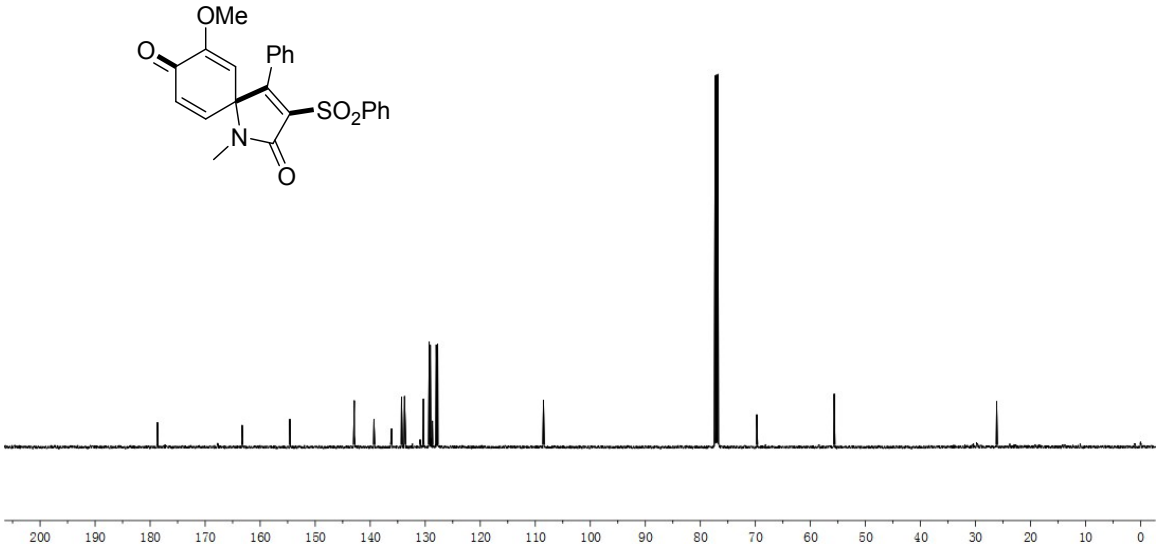
7.5542
7.5389
7.4354
7.3843
7.3687
7.3542
7.2638
7.1323
6.4350
5.2789
3.6625
2.8224
2.00
1.03
2.04
1.02
2.03
2.04
2.00
0.96
3.04
3.00


OMe
O
Ph
SO2Ph
N
O
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142.9046
139.3210
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134.3155
133.7639
130.3169
129.2749
129.0710
128.8512
128.6884
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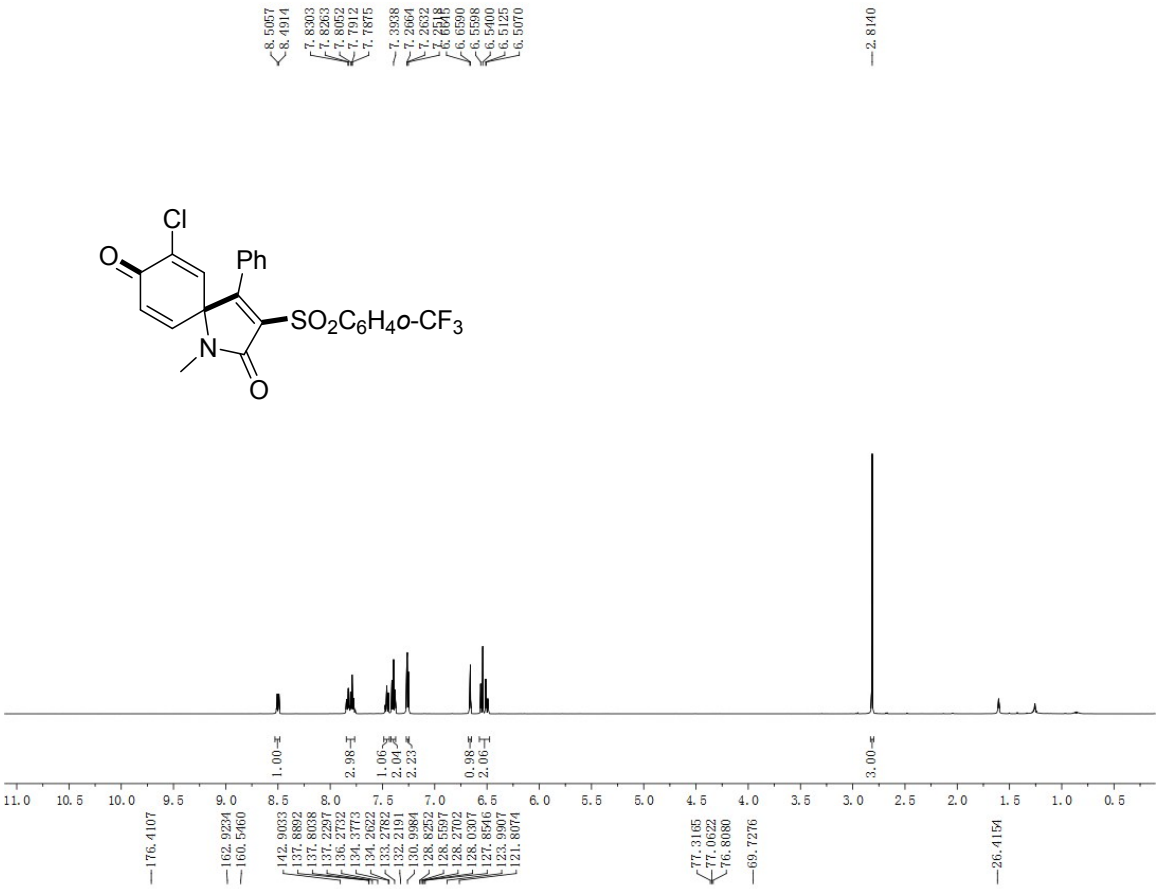
26.1553

3r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6.5598
6.5400
6.5125
6.5070
2.8140
Cl
O
Ph
SO2C6H4o-CF3
N
O
1.00
2.98
1.06
2.04
2.23
0.98
2.06
3.00
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
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160.5460
142.9033
137.8892
137.8038
137.2297
136.2732
134.3773
134.2622
133.2782
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130.9984
128.8252
128.5597
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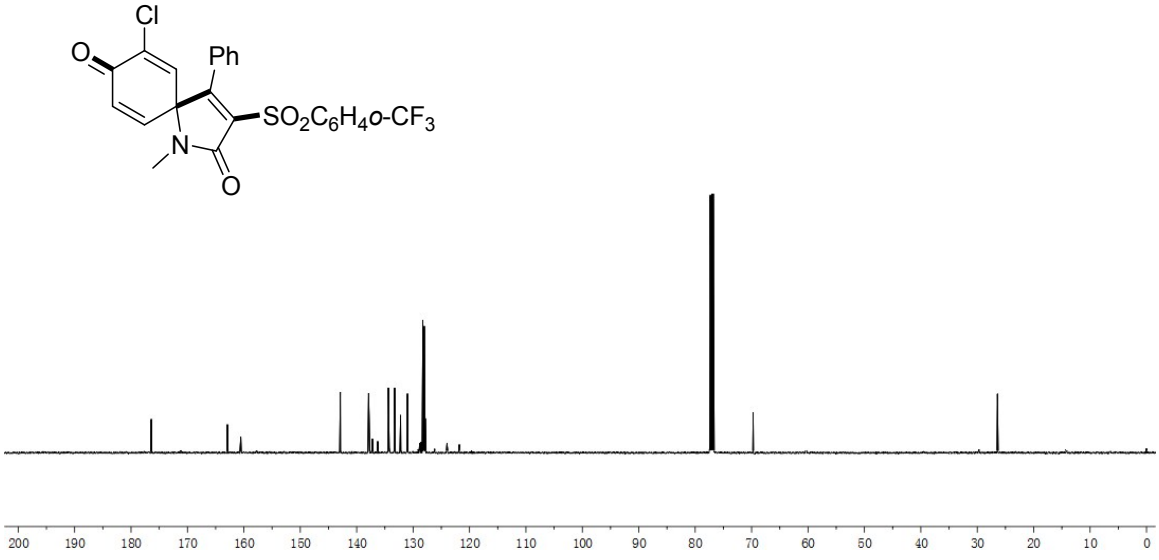
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O
Ph
SO2C6H4o-CF3
N
O
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3s


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7.4356
7.3811
7.3655
7.3247
7.3084
7.2614
7.1180
6.5425
6.5426
6.3163
6.2964
2.4402
Ph
SO2C6H4p-Me
N
H
O
2.01
1.02
2.04
2.02
1.96
0.96
2.06
1.99
3.00
183.2478
165.5374
164.5515
145.6407
142.5274
136.3359
136.2532
132.6472
132.4200
129.7690
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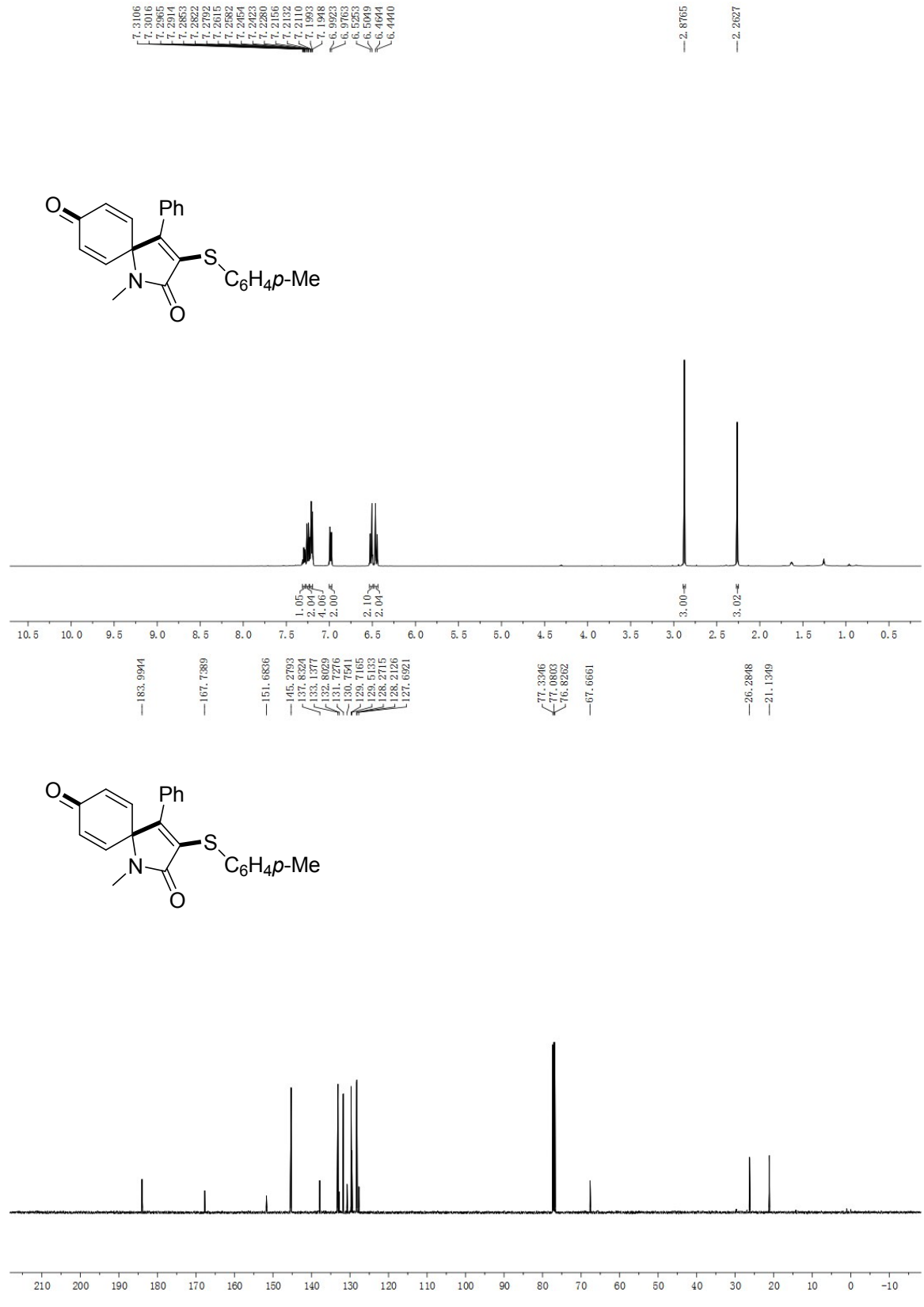
21.7774

5a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7.2454
7.2423
7.2280
7.2156
7.2132
7.2110
7.1993
7.1948
6.9923
6.9763
6.5253
6.5049
6.4664
6.4440
2.8765
2.2627
Ph
S
C6H4p-Me
N
O
1.05
2.04
4.06
2.00
2.10
2.04
3.00
3.02
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145.2793
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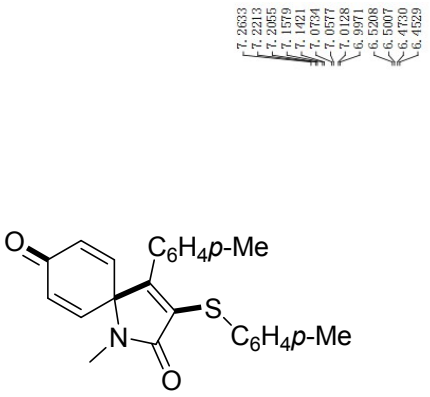
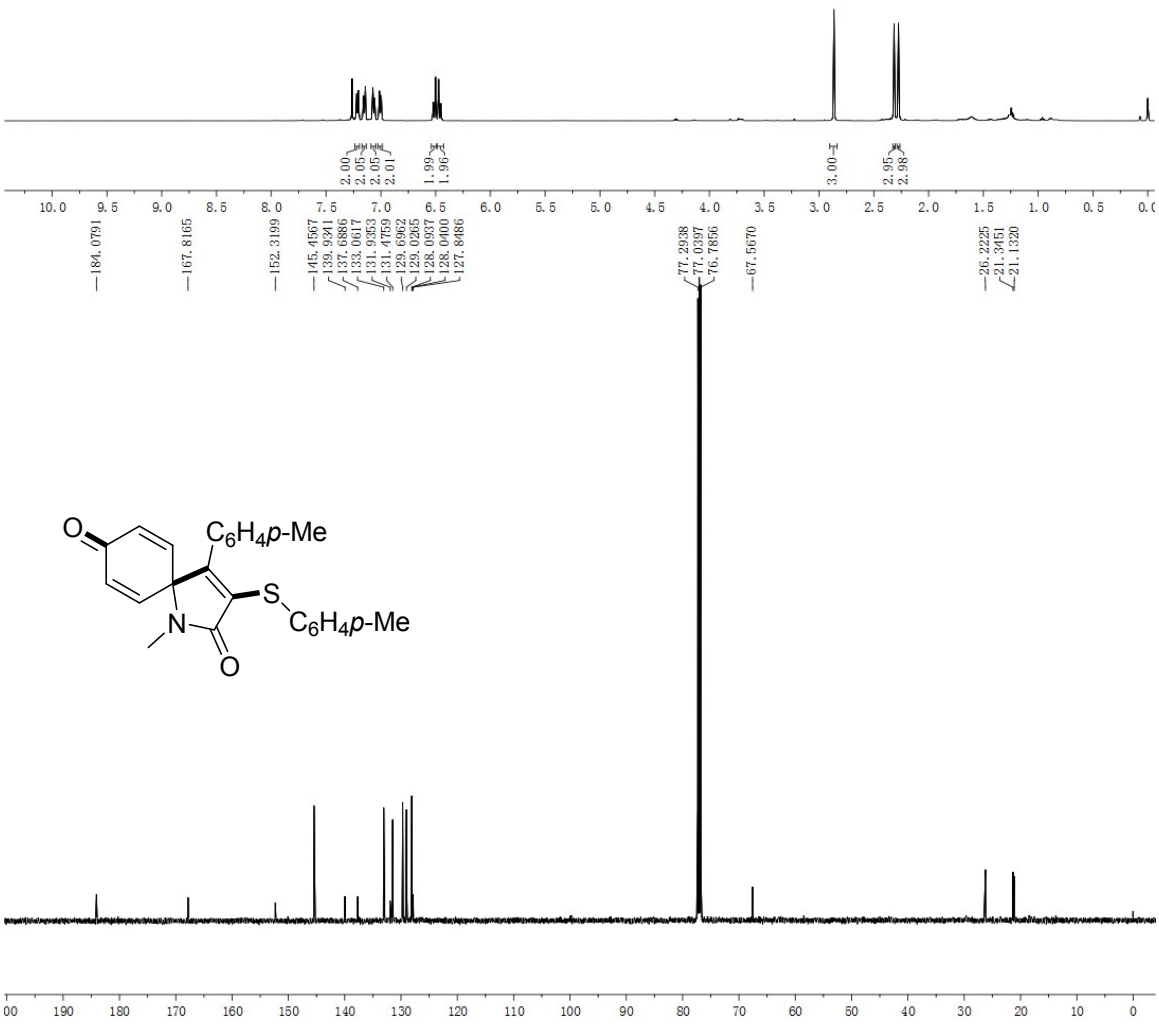
5b





5c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7.0224
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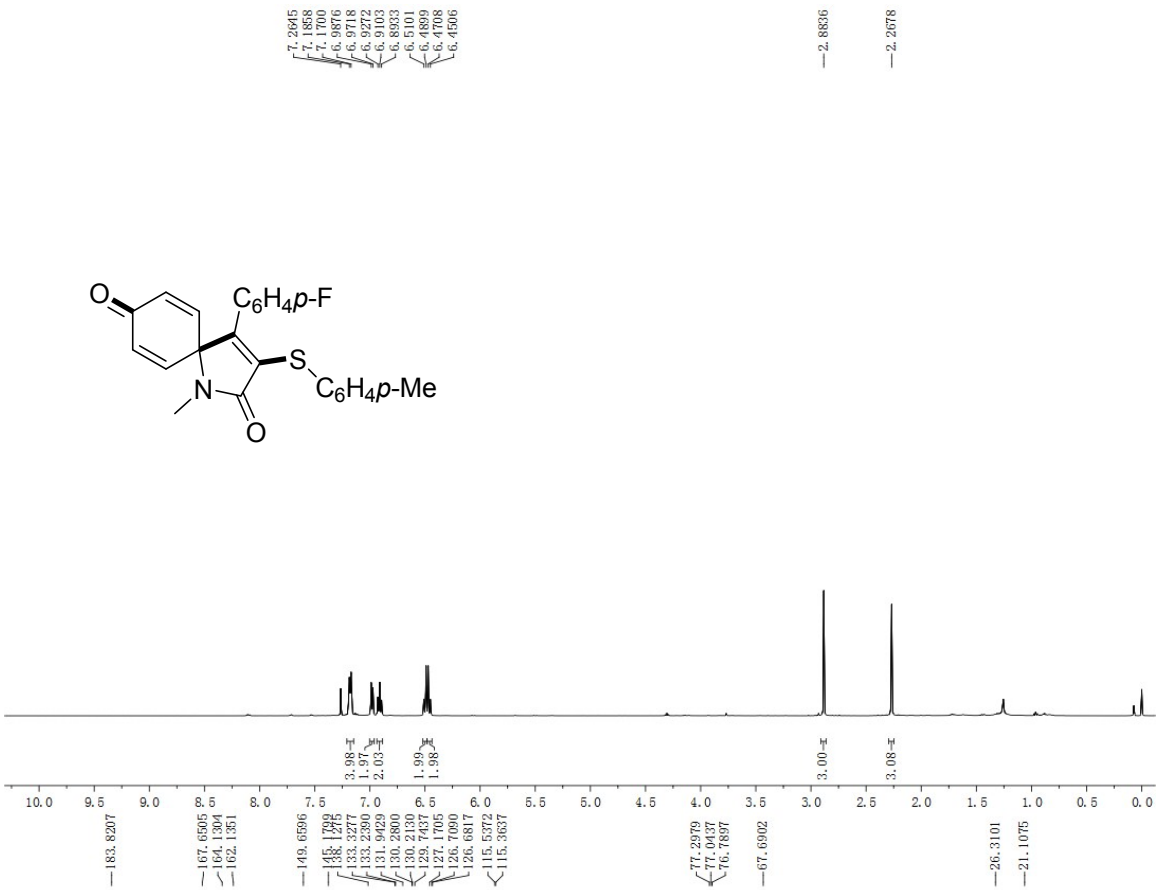
6.5293
6.5090
6.4932
6.4729
3.7841
2.8575
2.2763
O
C6H4p-OCH3
S
C6H4p-Me
N
O
1.97
1.94
1.99
1.99
1.96
1.98
3.00
2.93
3.04
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
184.1039
167.9453
160.6571
152.1709
145.7016
137.5656
132.9886
131.1441
130.8764
129.7326
129.6923
128.3217
123.0942
113.7971
77.2968
77.0427
76.7886
67.4285
55.2902
26.1335
21.1202
O
C6H4p-OCH3
S
C6H4p-Me
N
O
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10

5d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6.4506
2.8836
2.2678
O
C6H4p-F
S
N
C6H4p-Me
O
3.98
1.97
2.03
1.99
1.98
3.00
3.08
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
183.8207
167.6505
164.1304
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145.1798
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133.2290
131.9929
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126.6817
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77.0437
76.7897
67.6902
26.3101
21.1075
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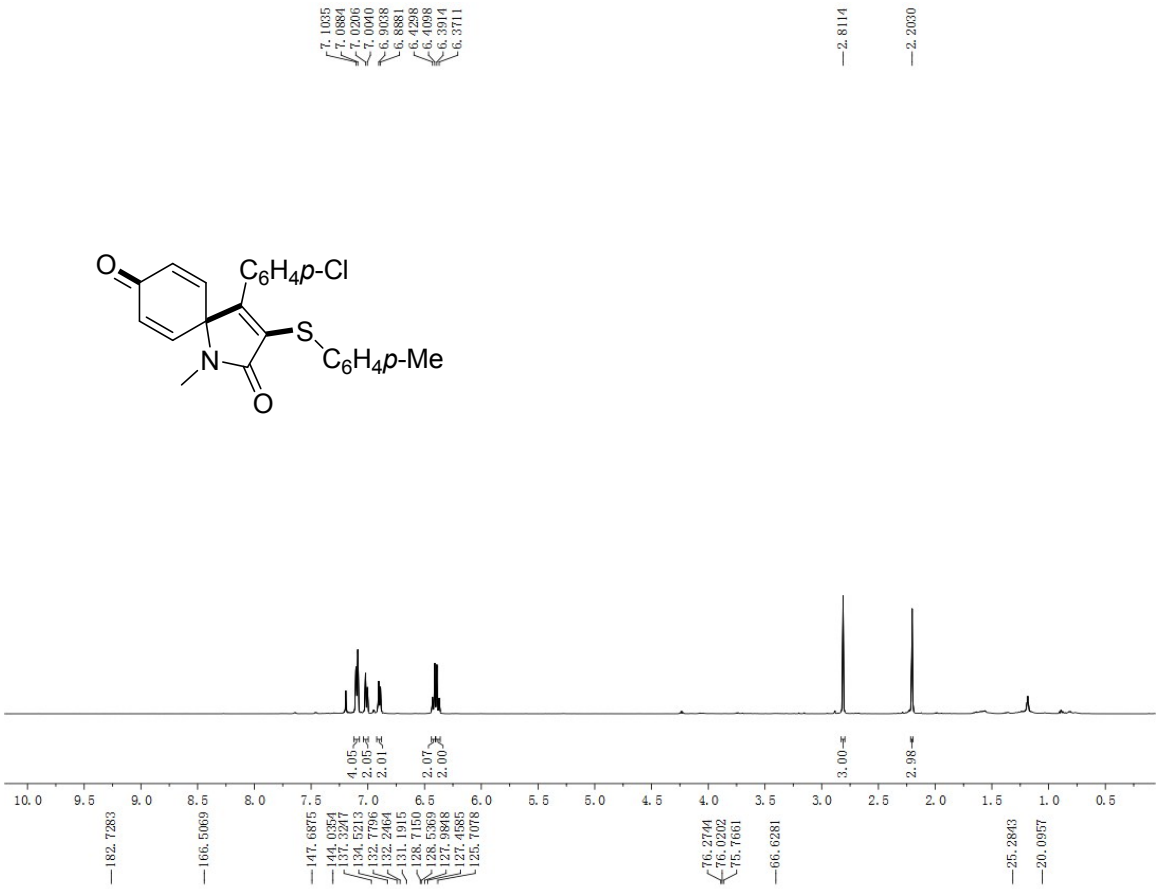
S
N
C6H4p-Me
O
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0

5e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6.3711
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2.2030
O
C6H4p-Cl
S
C6H4p-Me
N
O
4.05
2.05
2.01
2.07
2.00
3.00
2.98
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5
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147.6875
144.0354
137.3247
134.5213
132.7796
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66.6281
25.2843
20.0957
O
C6H4p-Cl
S
C6H4p-Me
N
O
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10

5f


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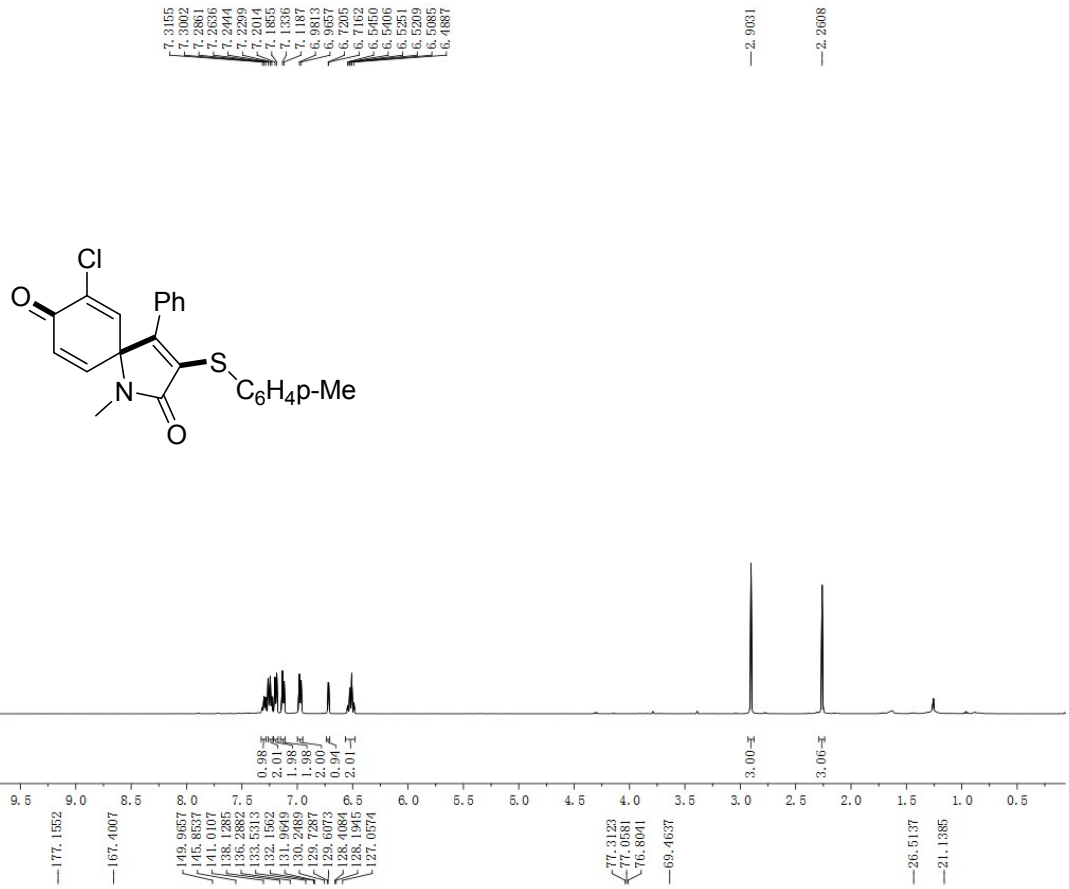
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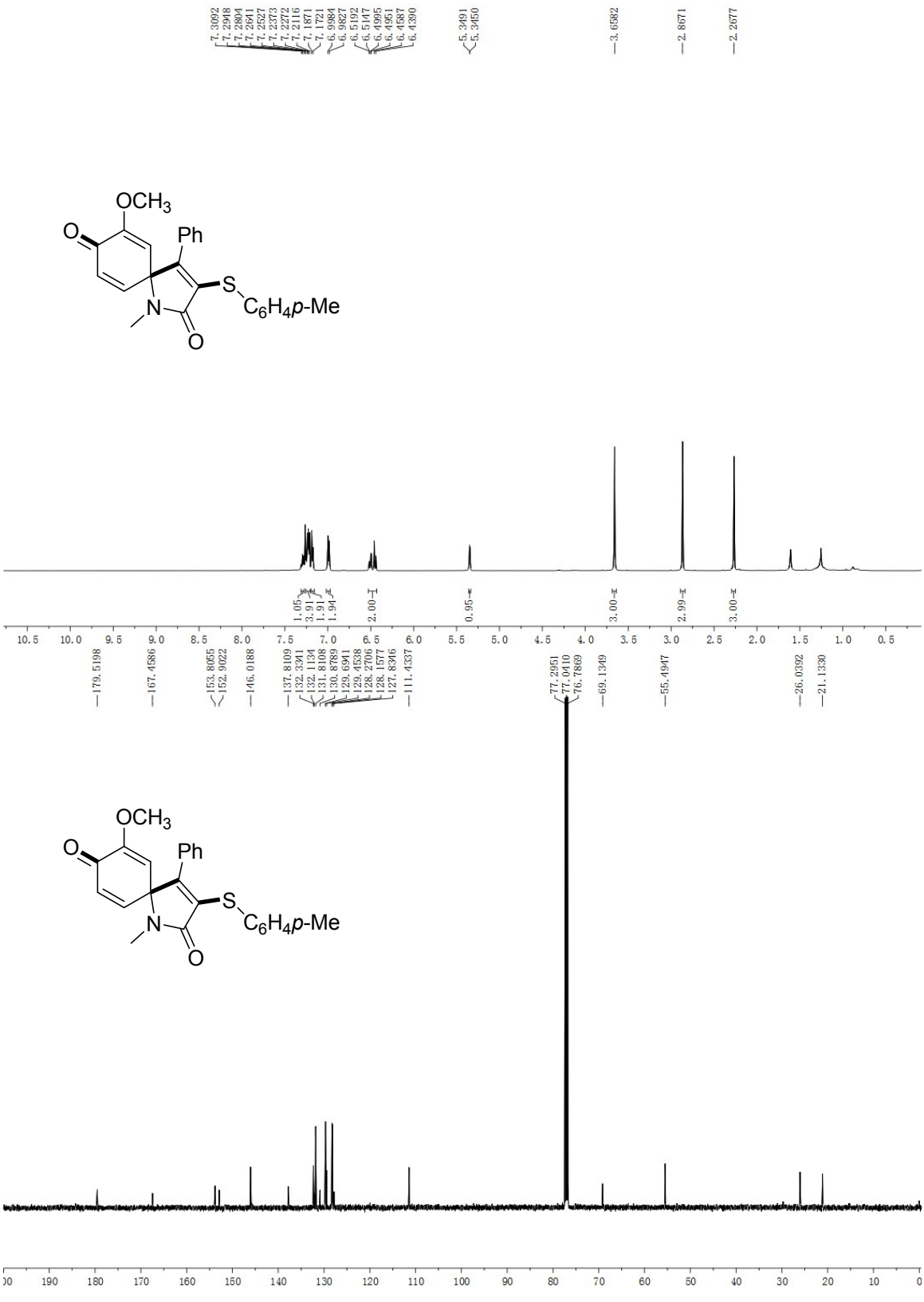
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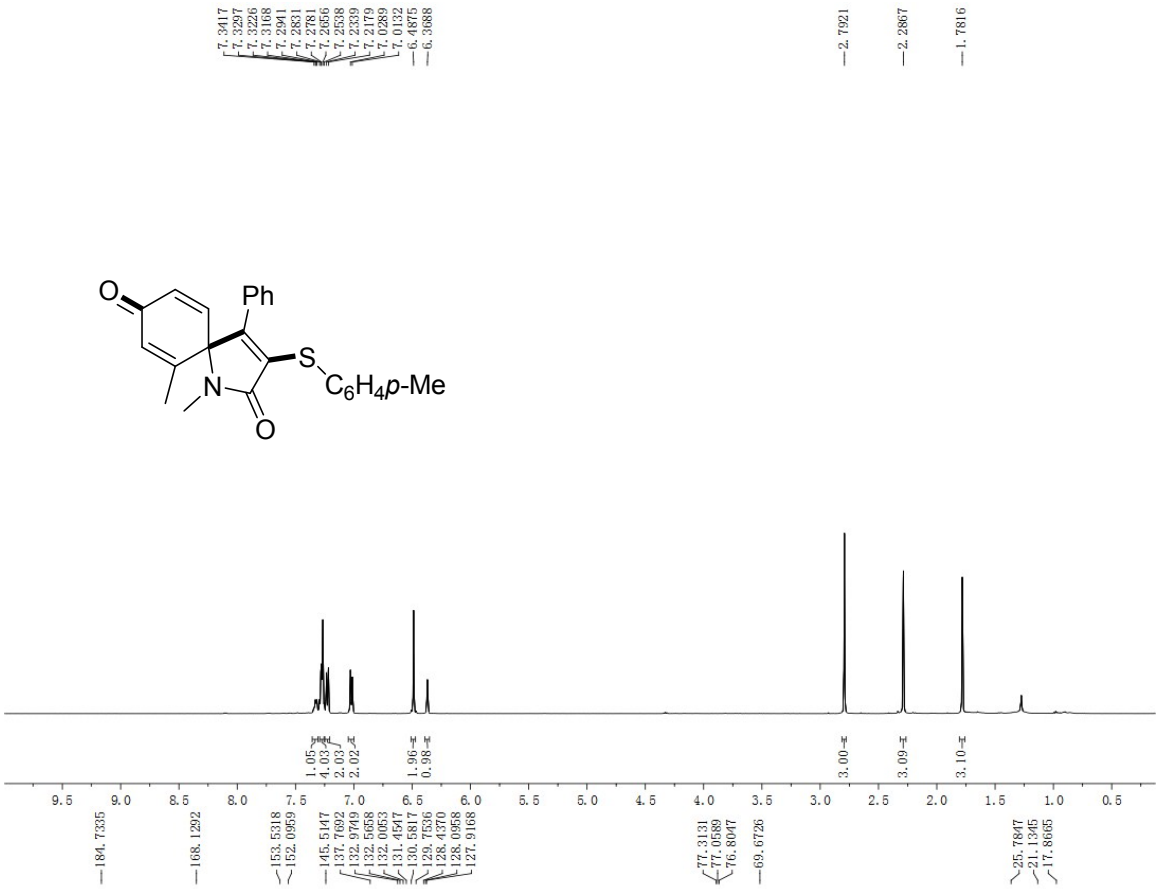
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3.10
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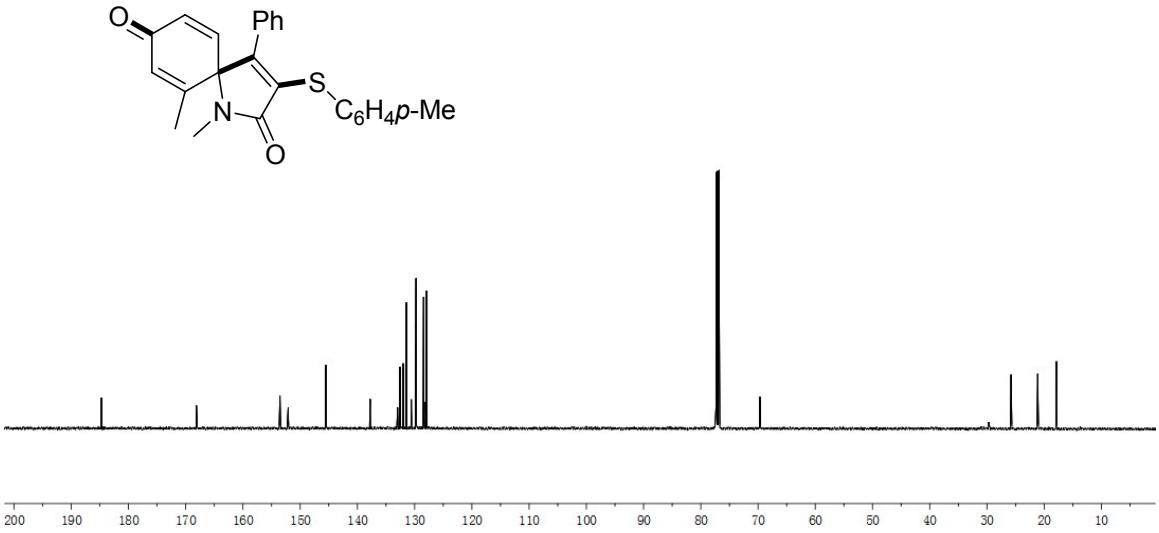
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5j


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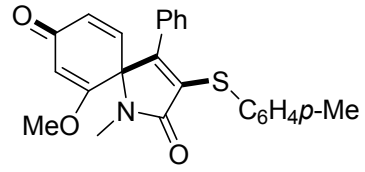
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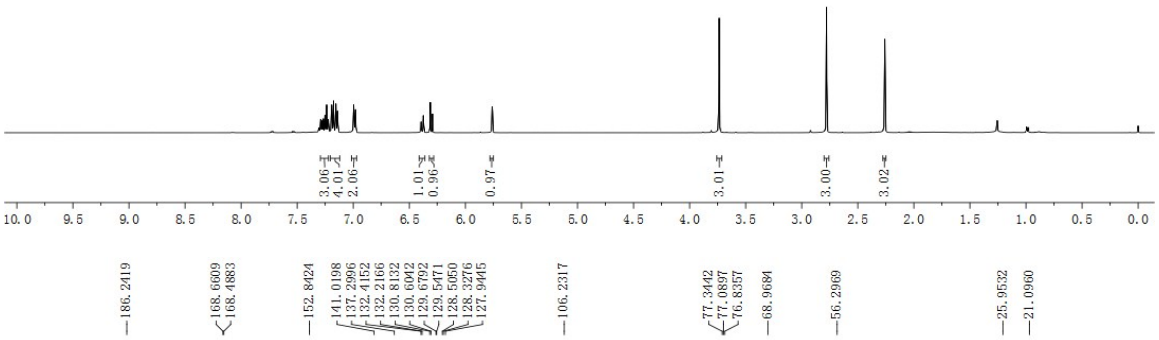
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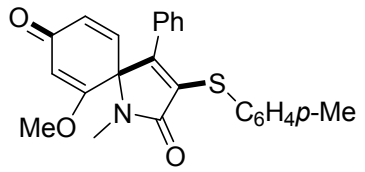
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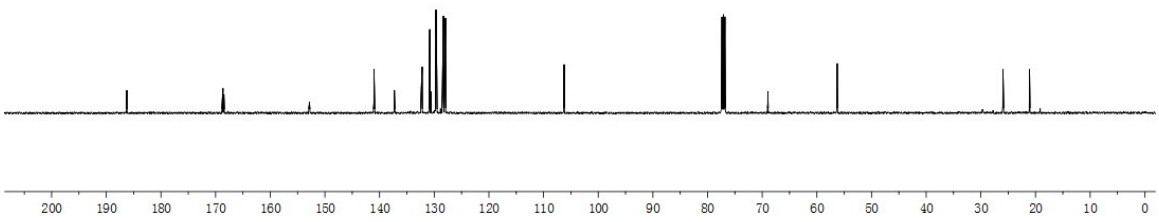
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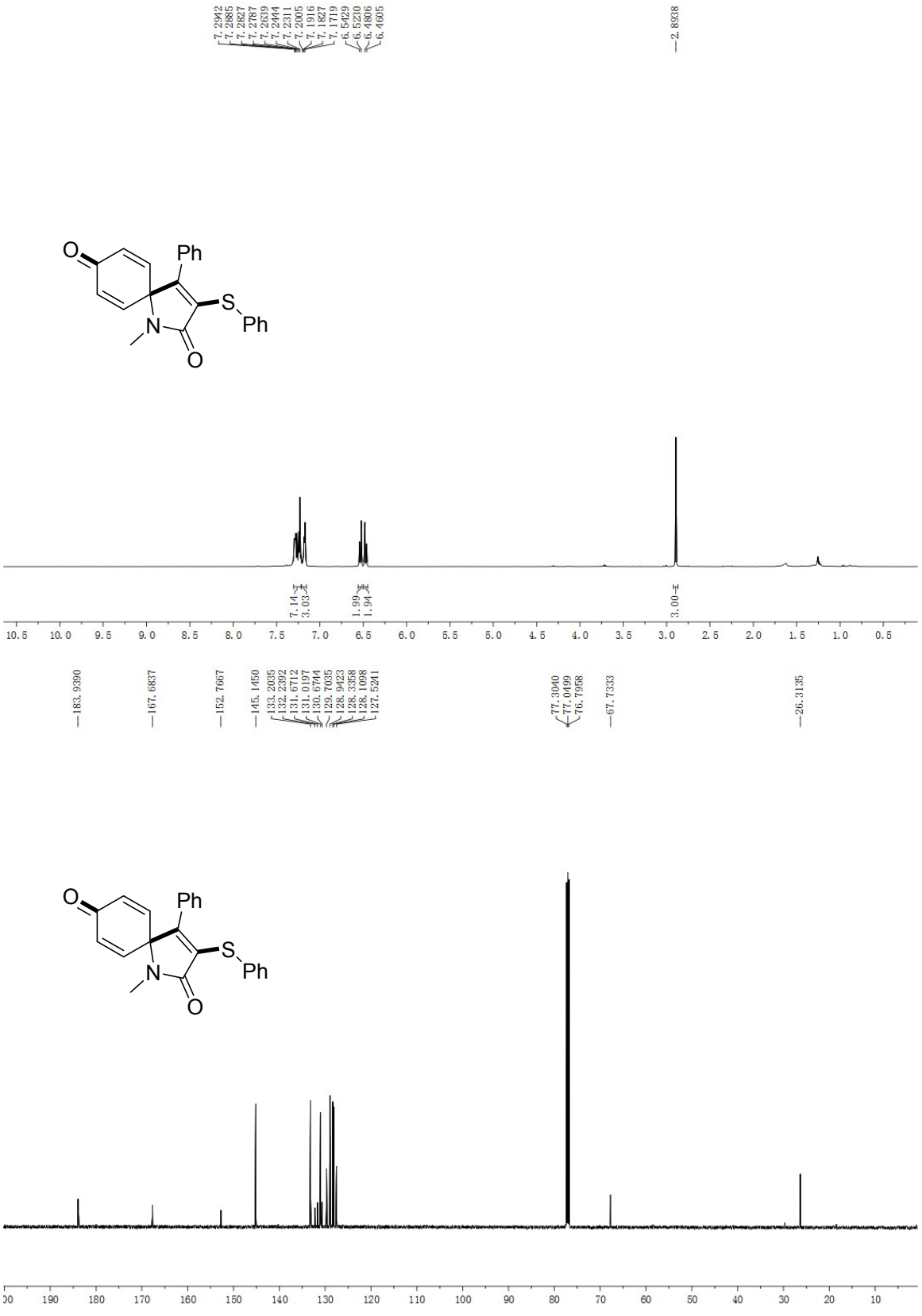
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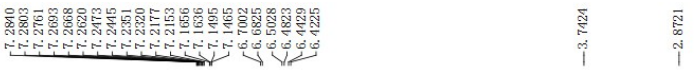
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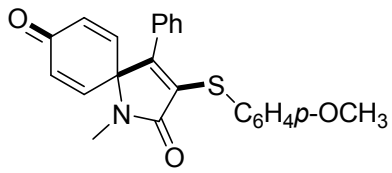
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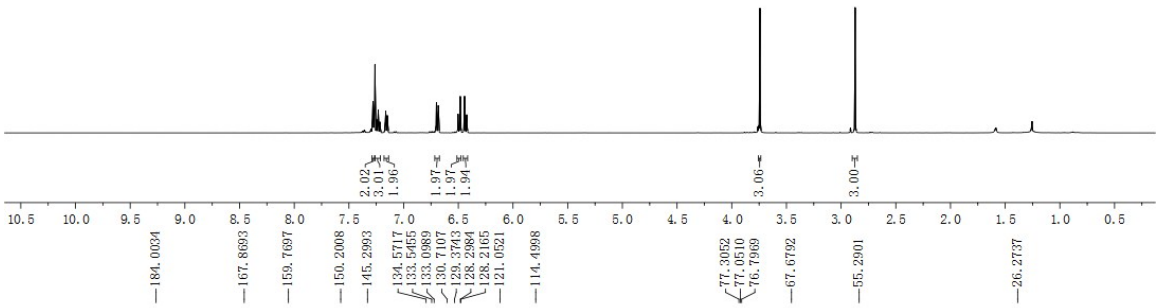
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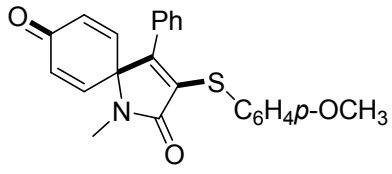
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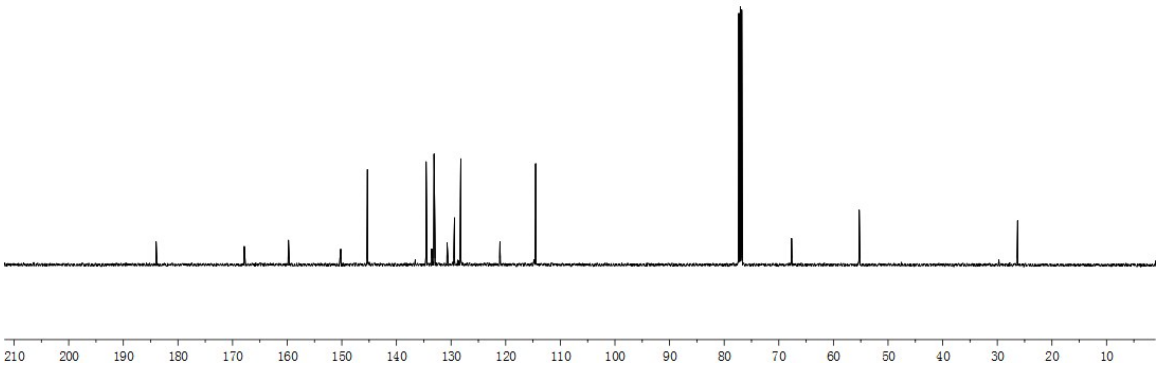
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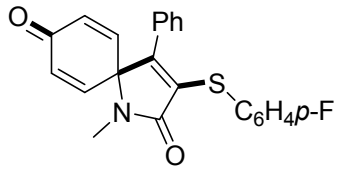
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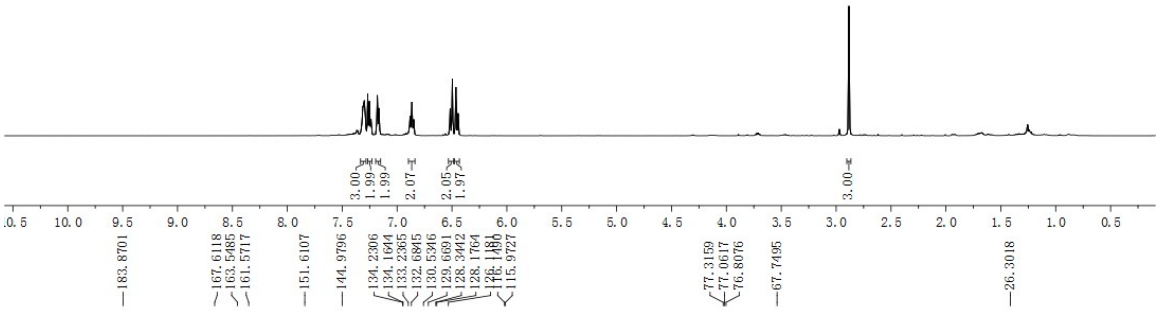
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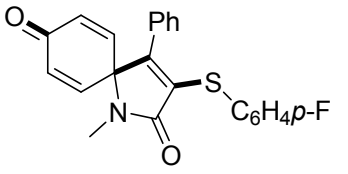
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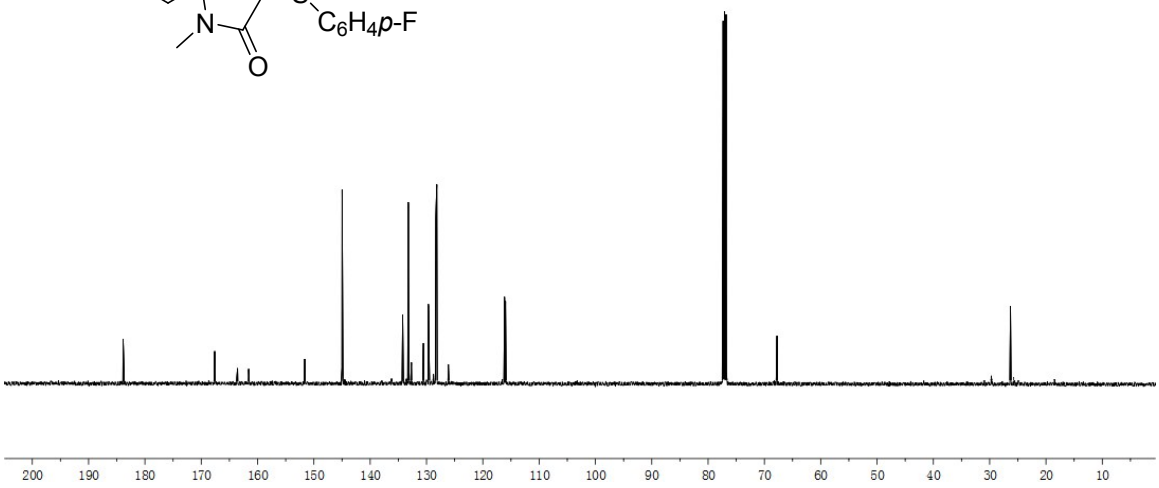
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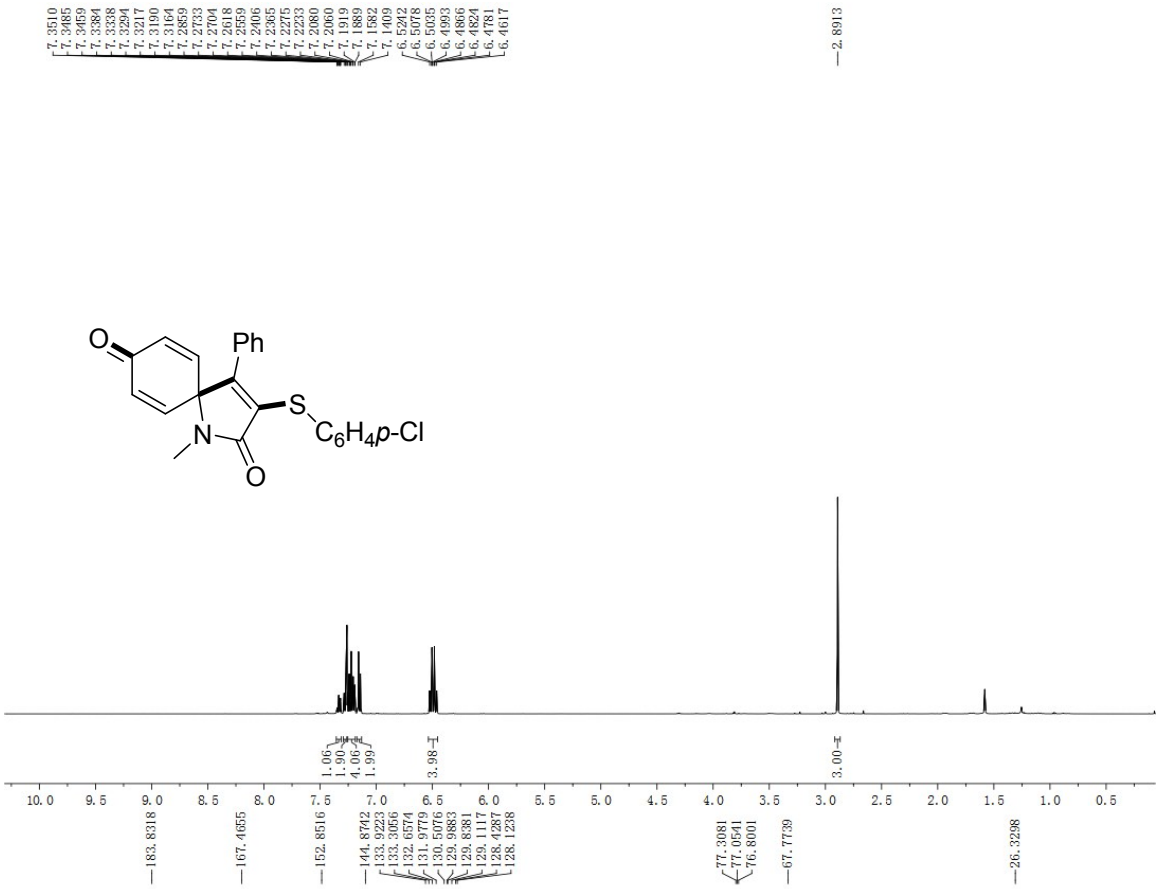
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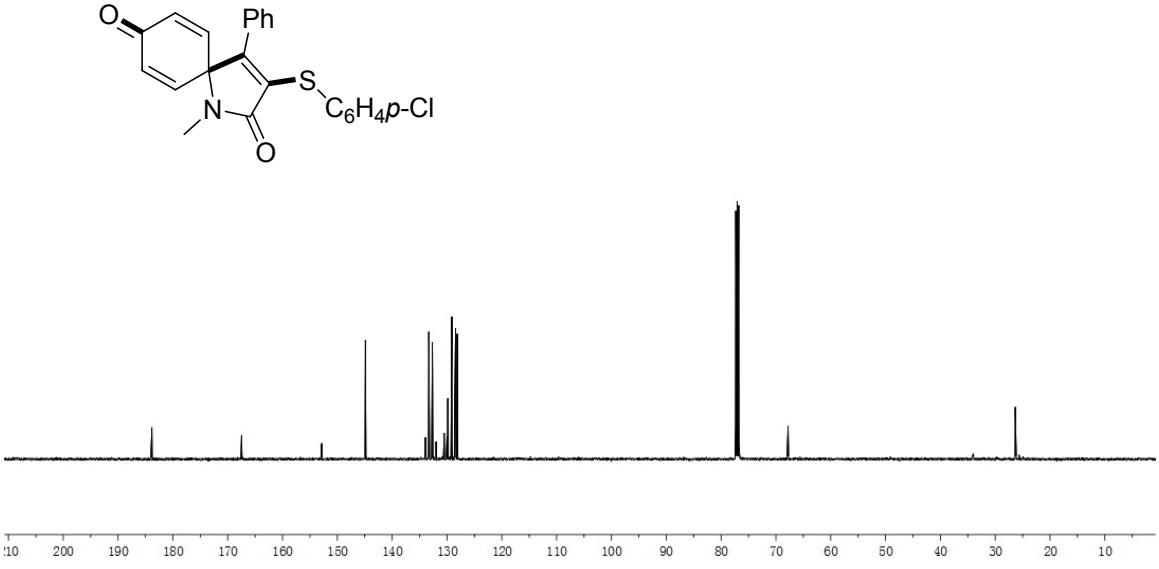
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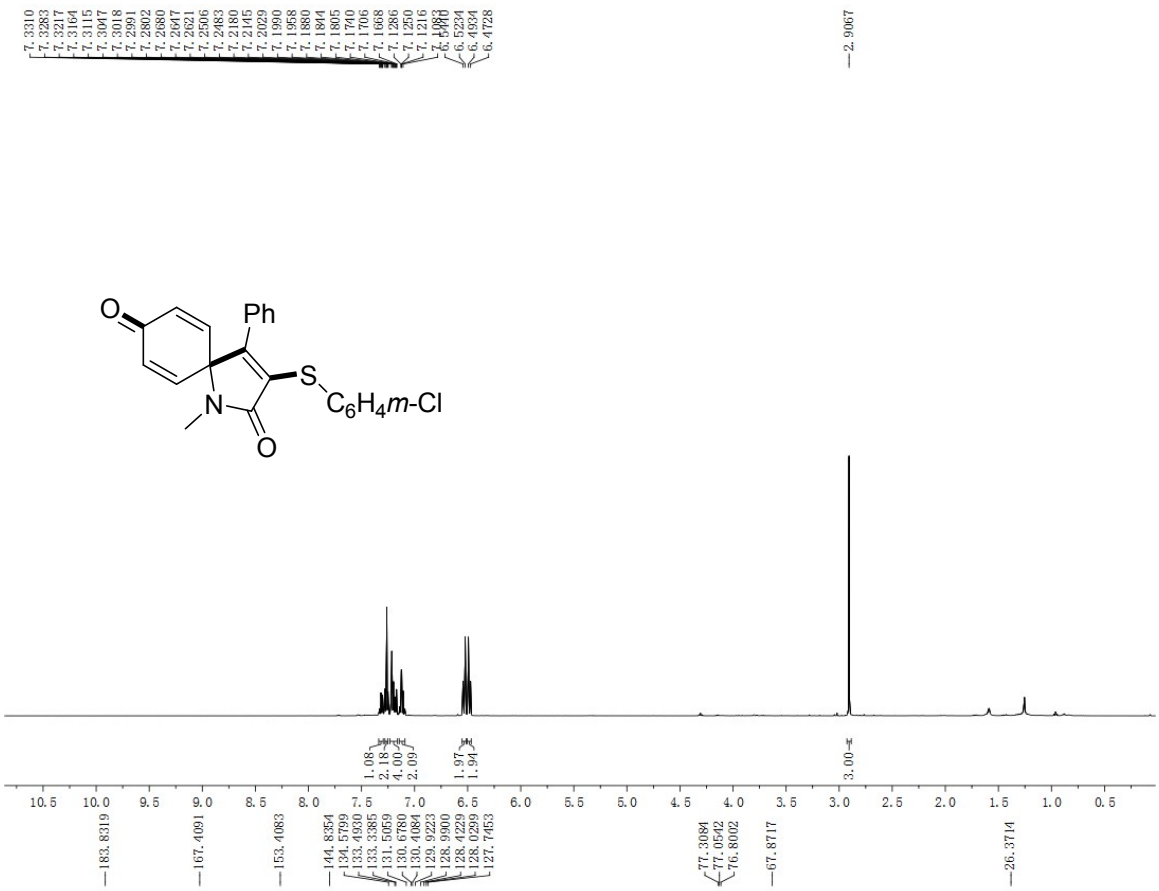
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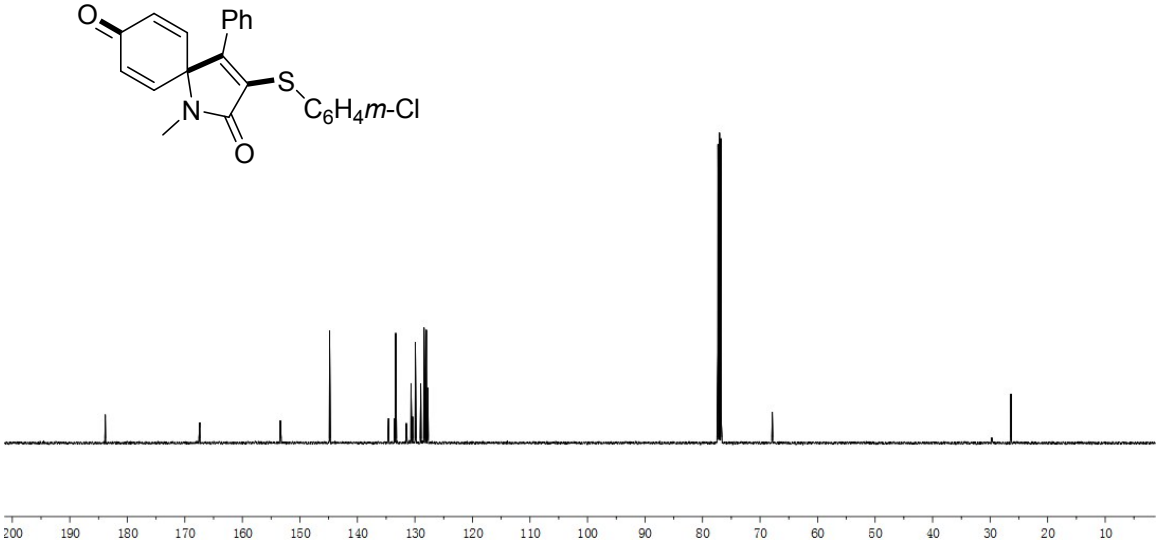
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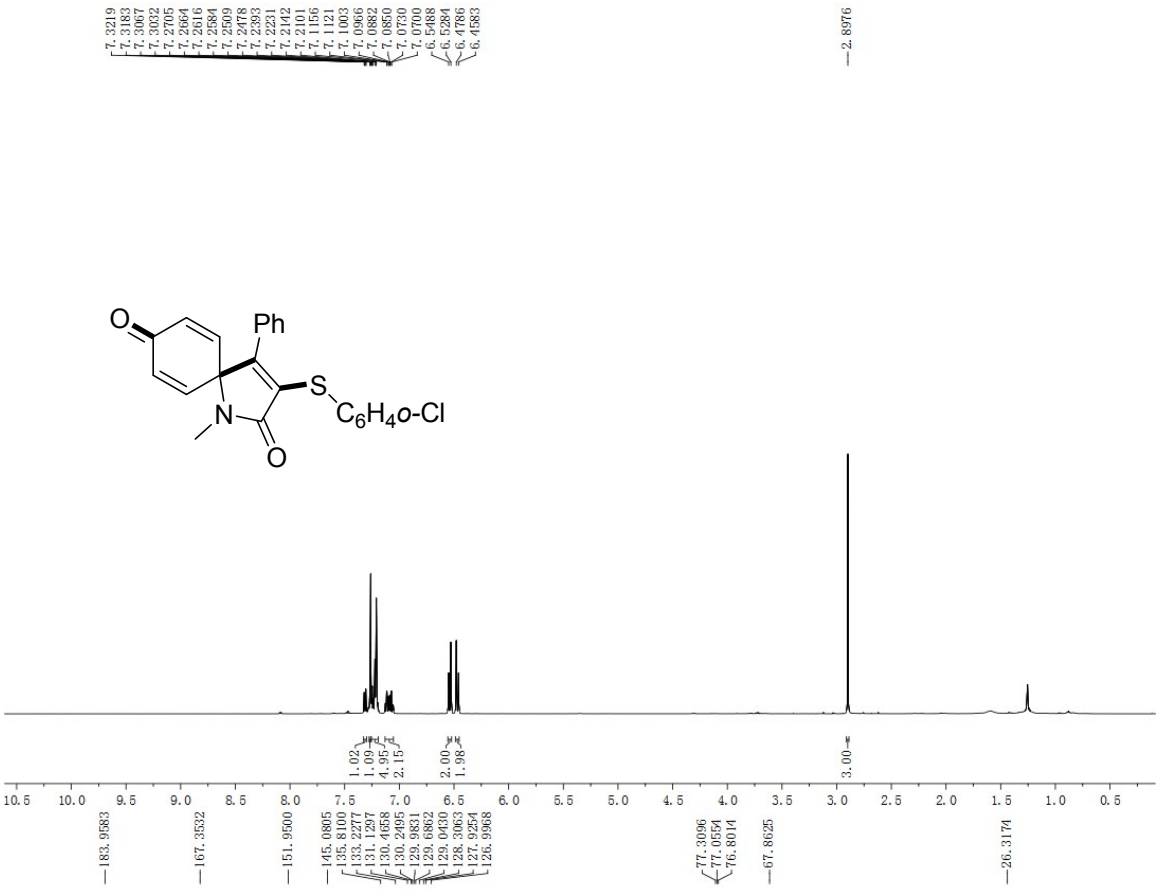
5p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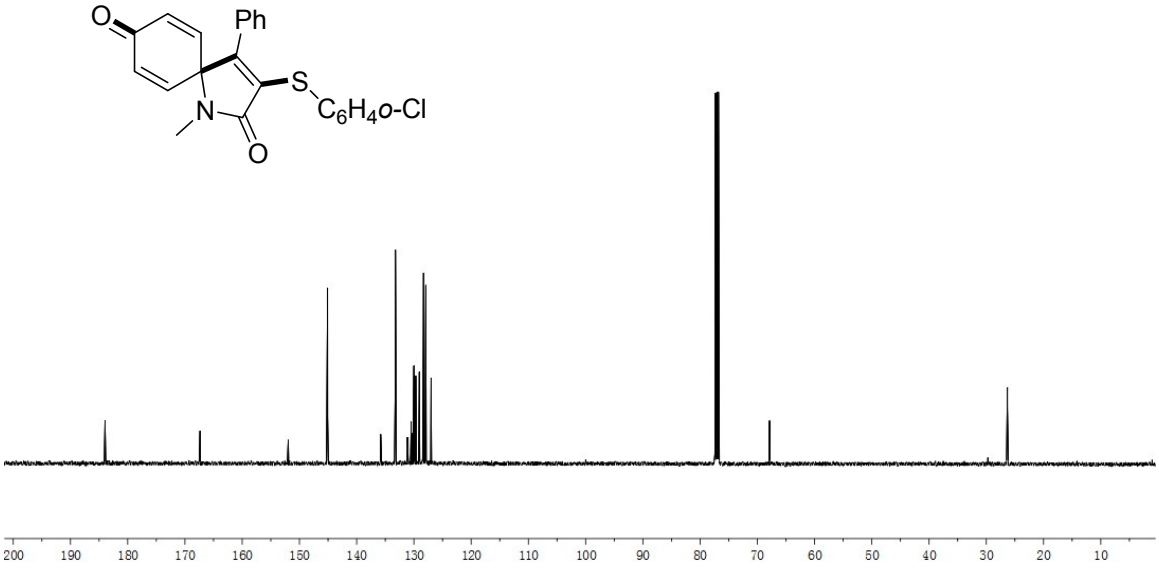
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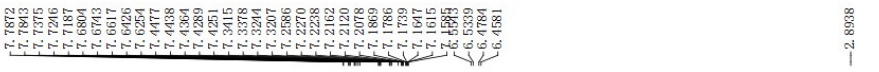
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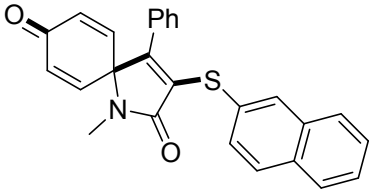
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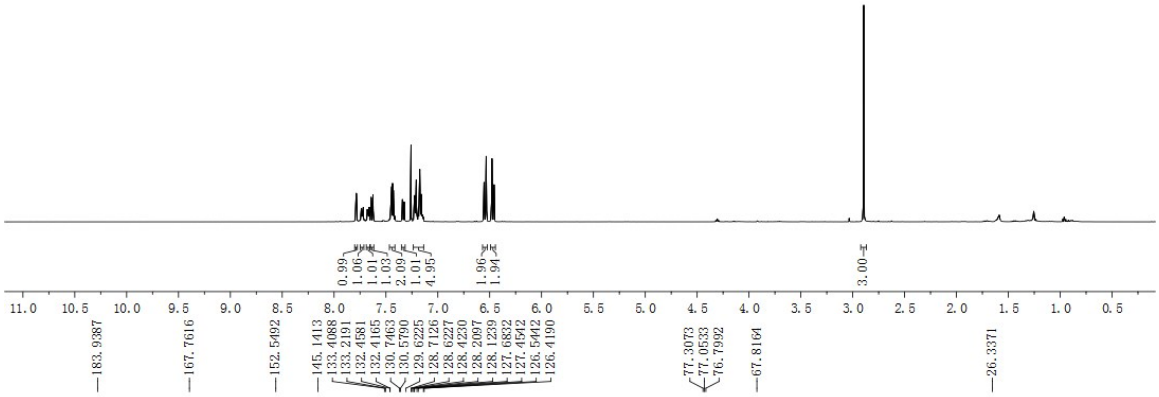
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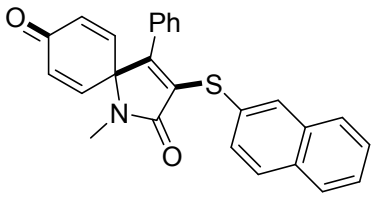
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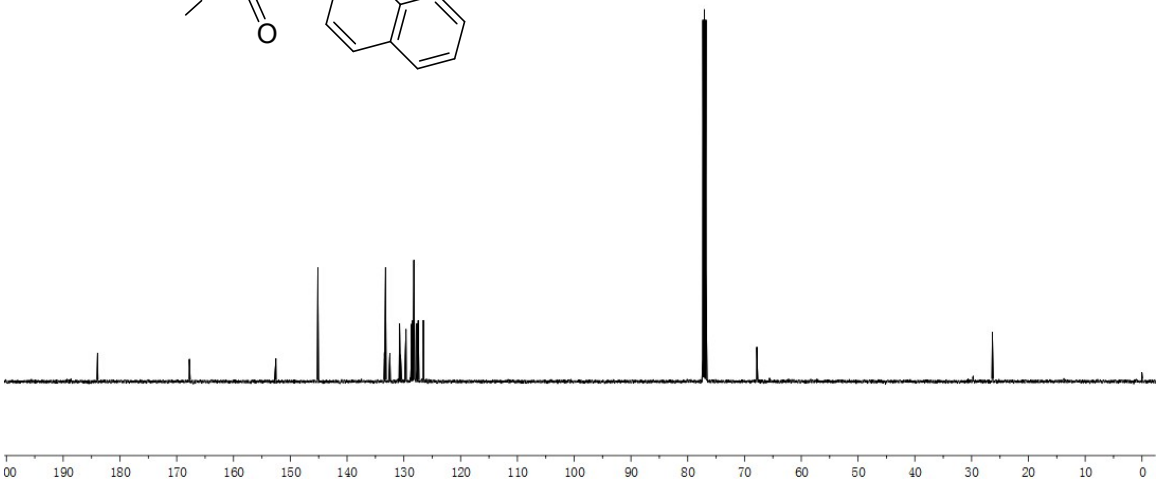
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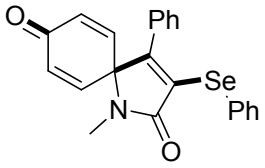
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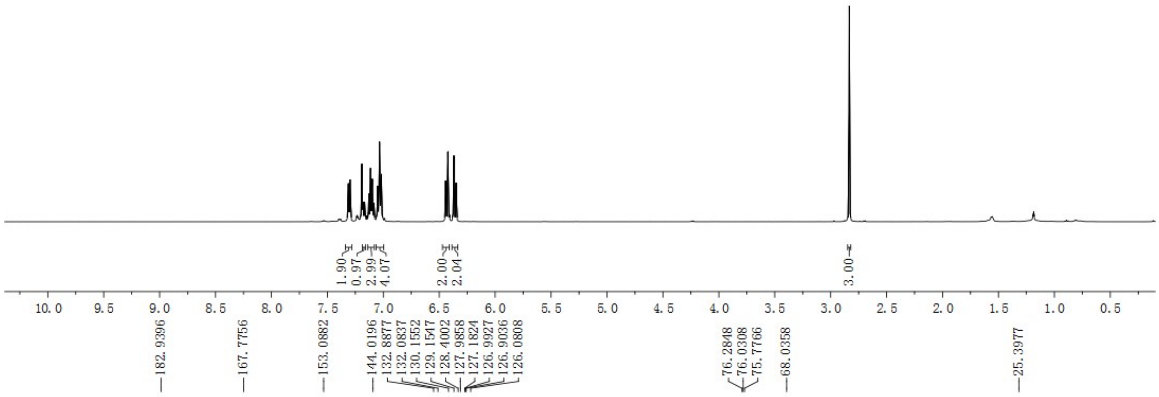
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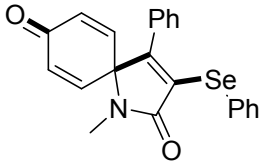
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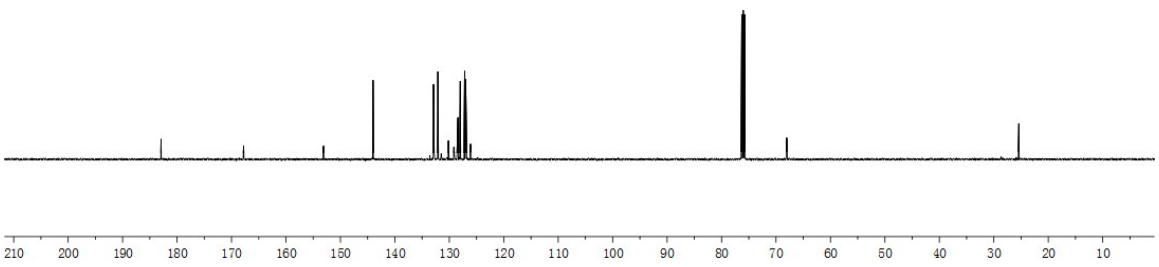
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