Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information

Visible-Light-Induced Metal-Free Cascade Cyclization of *N*-arylpropiolamides to 3-Phosphorylated, Trifluoromethylated and Thiocyanated azaspiro[4.5]trienones

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1. General Information

1.1 Materials and instruments

All commercially available reagents were used directly used without further purification. All reactions were monitored by Thin Layer Chromatography (TLC) using silica gel F254 plates. Products were purified by column chromatography using 200-300 mesh silica gel as the stationary phase. All the ^{1}H, ^{13}C, ^{31}P, and ^{19}F NMR spectra were recorded on Bruker Avance 400 or 600 spectrometers. All NMR spectra were recorded in $CDCl_3$ at room temperature (20 ± 2 °C). Proton chemical shifts δ were given in ppm using TMS as the internal standard. High-resolution mass spectra (HRMS) were obtained with a 3000-mass spectrometer, using Waters Q-Tof MS/MS system with the ESI technique. Emission intensities were recorded using an F-4600 FL spectrophotometer. Cyclic voltammetry was performed on the CHI-660E electrochemical workstation (Shanghai Chenhua Instrument Co., Ltd., China).

1.2 The spectrum of our lamp and the visible-light irradiation instrument.

All reactions have been studied in borosilicate glass vessels irradiated by blue light from a photoreactor manufactured by Beijing Roger Technology Co., Ltd. without using filters.

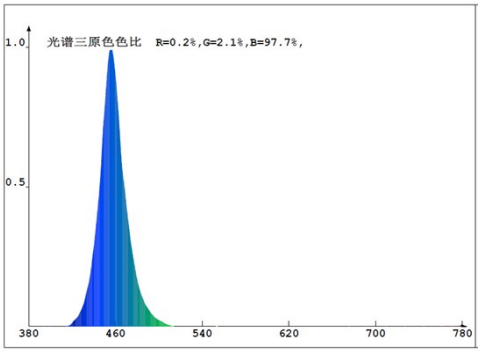


Figure S1a. The spectrum of our lamp (blue LED)

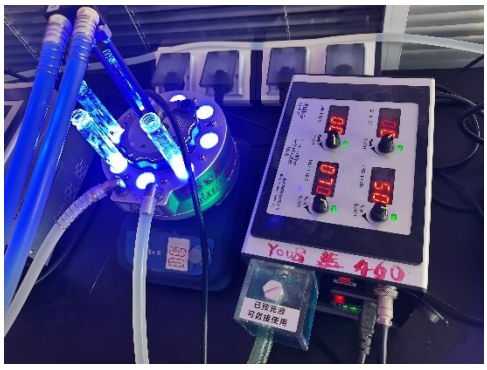
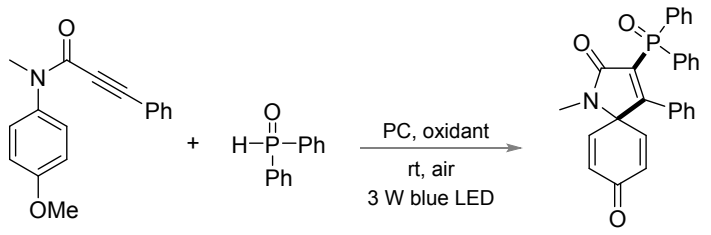


Figure S1b. The visible-light irradiation instrument

2. Experimental procedures

2.1 Optimization of reaction conditions

Table S1. Optimization of reaction conditions of **3a**



1a **2a** **3a**

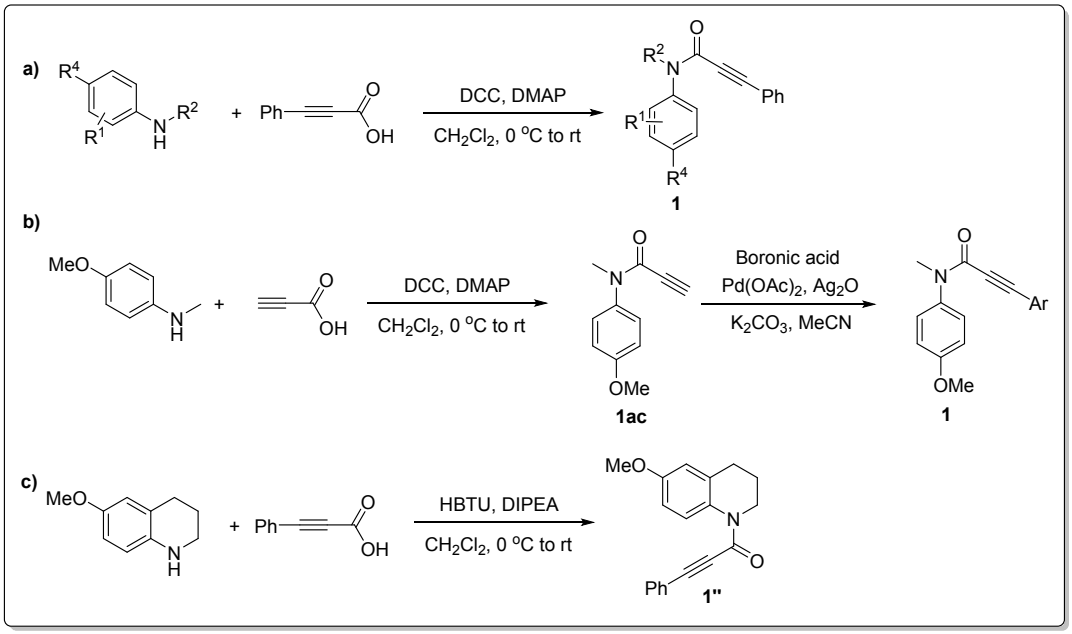
Entry	PC (mol%)	Solvent (v/v)	Oxidant (equiv)	Yield (%)
1	4CzIPN (5)	MeCN	LPO (2)	60
2	4CzIPN (5)	DMC	LPO (2)	40
3	4CzIPN (5)	H_2O	LPO (2)	20
4	4CzIPN (5)	DMF	LPO (2)	N.D.
5	4CzIPN (5)	DCE	LPO (2)	N.D.
6	4CzIPN (5)	DMSO	LPO (2)	N.D.
7	4CzIPN (5)	MeCN/H_2O (100:1)	LPO (2)	50
8	4CzIPN (5)	MeCN/H_2O (50:1)	LPO (2)	55
9	4CzIPN (5)	MeCN/H_2O (30:1)	LPO (2)	70
10	**4CzIPN (5)**	**MeCN/H_2O (20:1)**	**LPO (2)**	**83**
11	4CzIPN (5)	MeCN/H_2O (15:1)	LPO (2)	60
12	Na_2EosinY (5)	MeCN/H_2O (20:1)	LPO (2)	15
13	Rose Bengal (5)	MeCN/H_2O (20:1)	LPO (2)	trace
14	Eosin B (5)	MeCN/H_2O (20:1)	LPO (2)	20
15	Eosin Y (5)	MeCN/H_2O (20:1)	LPO (2)	15
16	4CzIPN (5)	MeCN/H_2O (20:1)	$K_2S_2O_8$ (2)	25
17	4CzIPN (5)	MeCN/H_2O (20:1)	H_2O_2 (2)	trace
18	4CzIPN (5)	MeCN/H_2O (20:1)	TBHP (2)	trace
19	4CzIPN (5)	MeCN/H_2O (20:1)	BPO (2)	30
20	4CzIPN (5)	MeCN/H_2O (20:1)	DDQ (2)	trace
21	4CzIPN (5)	MeCN/H_2O (20:1)	TBPB (2)	trace
22[b]	4CzIPN (5)	MeCN/H_2O (20:1)	LPO (2)	N.D.
23	--	MeCN/H_2O (20:1)	LPO (2)	N.D.

[a] Reaction conditions: **1a** (0.2 mmol), **2a** (0.4 mmol), solvent (2 mL) under air for 3 h, room temperature, blue LED (3 W), PC = photocatalyst, N.D. = not detected, DMC = dimethyl carbonate. Yields were determined by ^{1}H NMR using 1,1,2,2-tetrachloroethane as the internal standard. [b] In dark.

We started with establishing a suitable reaction condition for accessing 3-phosphorylazaspiro[4.5]trienone using the model reaction of *N*-arylpropiolamide **1a** with diphenylphosphine oxide (**2a**) in the presence of different photocatalysts and oxidants under irradiation of blue LED at room temperature in open air, as summarized in Table S1. The influence of a variety of solvents on the model reaction was first examined (entries 1-11). Among them, the

target product **3a** was obtained in the highest yield of (83%) in a mixed solvent (MeCN-H_2O, v/v: 20/1) (entry 10). Encouraged by this satisfied result, some other dyes as the photocatalysts (Na_2EosinY, Rose Bengal, Eosin B, and Eosin Y) were further examined (entries 12-15). However, in sharp contrast, much less yields were observed with them. Among them, the highest yield was produced by using Eosin B and only up to 20% (entry 14). After that, some other usually used oxidants including $K_2S_2O_8$, H_2O_2, tert-Butyl hydroperoxide (TBHP), benzoyl peroxide (BPO), 2,3-dichloro-5,6-dicyano-1,4-benzoquinone (DDQ), and tert-butyl peroxybenzoate (TBPB) on the model reaction were tried (entries 16-21). No any satisfied yields were observed in those cases, among which, the highest yield was gained with BPO and was up to 30% (entry 19). At last, the model reaction was carried out in dark and no 4CzIPN, as predicted, no any product of **3a** was obtained (entry 22-23). After a wide exploration, the optimal conditions were finally established as follows: **1a** (0.2 mmol), **2a** (0.4 mmol), 4CzIPN (5 mol%), LPO (2 equiv), MeCN:H_2O (2 mL:100 uL) in air atmosphere under the irradiation of 3 W blue LED at room temperature for 3 h.

2.2 Preparation of starting materials

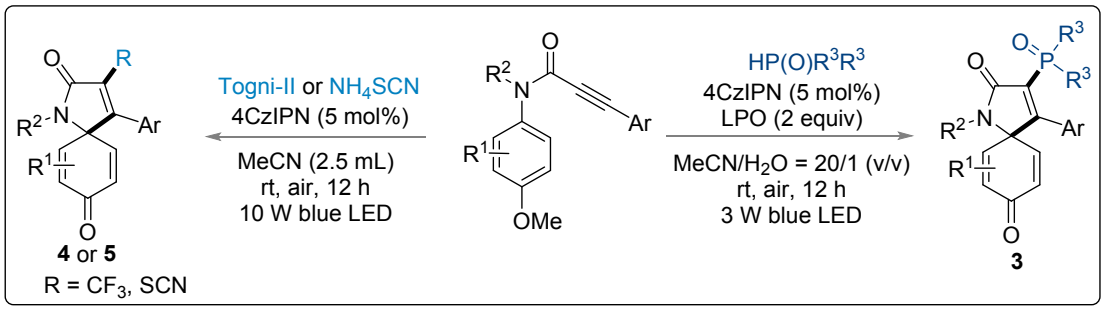


Scheme S1. General experimental procedures for substrates *N*-arylpropiolamides **1**

General procedure a): The mixture of *N*-methylaniline (or the relative aniline) (2.5 mmol, 1.0 equiv.) in CH_2Cl_2 (15 mL) was added the corresponding propynoic acid (2.75 mmol, 1.1 equiv) at 0 ℃, then a mixture of dicyclohexylcarbodiimide (DCC) (3.75 mmol, 1.5 equiv) and 4-dimethylaminopyridine (DMAP) (0.25 mmol, 0.1 equiv) in CH_2Cl_2 (10 mL) was added dropwise, stirred at room temperature for 12 h. Then, the mixture was filtered and washed with CH_2Cl_2 (3 x 50 mL) and concentrated. The residue was purified by a silica gel column chromatography (ether/ethyl acetate (v/v = 5/1)) to give the products (**1a-j**).[1, 2 5]

General procedure b): Following the general procedure **a)** to get **1ac,** then *N*-methyl-*N*-phenylpropiolamide (0.5 mmol, 80 mg), boronic acid (0.6 mmol), $Pd(OAc)_2$ (1 mol%, 1.2 mg), Ag_2O (0.75 mmol, 175 mg), K_2CO_3 (1 mmol, 138 mg) and MeCN (3 mL) was added to a Schlenk tube (25 mL). Then the solution was stirred at 70 ℃ for 12 h. After the reaction was finished, the mixture was filtered, extracted with ethyl acetate, and evaporated in a vacuum. The residue was purified by flash column chromatography (ether/ethyl acetate (v/v = 5/1)) on silica gel to afford the desired product (**1k-o**).[5]

General procedure c): The mixture of propynoic acid (3.3 mmol, 1.1 equiv) in CH_2Cl_2 under nitrogen condition at 0 ℃ then add *O*-Benzotriazole-N,N,N',N'-tetraMethyl-uroniuM-hexafluorophosphate (HBTU) (4.5 mmol, 1.5 equiv). After 30 mins, 6-methoxy-1,2,3,4-tetrahydroquinoline (3 mmol, 1 equiv) was added, followed by the addition of *N,N*-diisopropylethylamine (DIPEA) (9 mmol, 3 equiv) dropwise period of 5 mins at 0 ℃. The reaction mixture was allowed to stir for 12 h at room temperature. The reaction quenched with water. The aqueous solution was extracted with CH_2Cl_2 (3 x 50 mL). The combined organic layers were washed with brine solution (50 mL). After the organic layer was dried by Na_2SO_4 and concentrated. The residue was purified by a silica gel column chromatography (ether/ethyl acetate (v/v = 3/1)) to give the product (**1''**).[2]



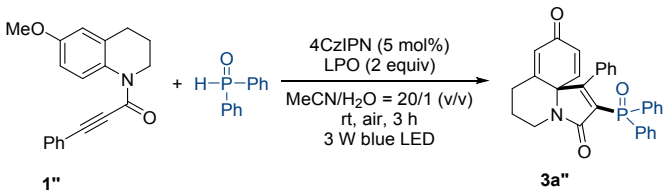
Scheme S2. General experimental procedures for the azaspiro[4.5]trienones

General procedure: The mixture of *N*-arylpropiolamides **1** (0.2 mmol), diarylphosphine oxides (0.4 mmol), 4CzIPN (5 mol%), LPO (2 qeuiv) and MeCN/H_2O (2 mL/0.1 mL) were sequentially added in a 25 mL reaction vessel. Then the reaction vessel was exposed to the irradiation of 3 W blue LED at room temperature for 3 h (open-air). After the reaction, the solvent was evaporated under vacuum, all the crude products were purified by thin-layer chromatography using petroleum ethyl acetate as eluting solvent to give the desired products **3**.

The mixture of *N*-arylpropiolamides **1** (0.2 mmol), togni-II (0.4 mmol), 4CzIPN (5 mol%) and MeCN (2.5 mL) were sequentially added in a 25 mL reaction vessel. Then the reaction vessel was exposed to the irradiation of 10 W blue LED at room temperature for 12 h (open-air). After the reaction was completed, 20 ml of CH_2Cl_2 was added, and then the reaction solution was combined and washed by a saturated $NaHCO_3$ solution (25 mL x 3), the aqueous solution was extracted with CH_2Cl_2 (3 x 15 mL). After the organic layer was dried by Na_2SO_4 and concentrated under vacuum, the crude products were further purified by silica gel chromatography using petroleum ether/ethyl

acetate (v/v = 3/1) as eluting solvent to give the desired products **4**.

The mixture of *N*-arylpropiolamides **1** (0.2 mmol), NH_4SCN (0.4 mmol), 4CzIPN (5 mol%) and MeCN (2.5 mL) were sequentially added in a 25 mL reaction vessel. Then the reaction vessel was exposed to 10 W blue LED irradiation at room temperature with stirring for 12 h (open air). After the reaction, the solvent was evaporated under vacuum, all the crude products were purified by silica gel chromatography using petroleum ether/ethyl acetate (v/v = 3/1) as eluting solvent to give the desired products **5**.

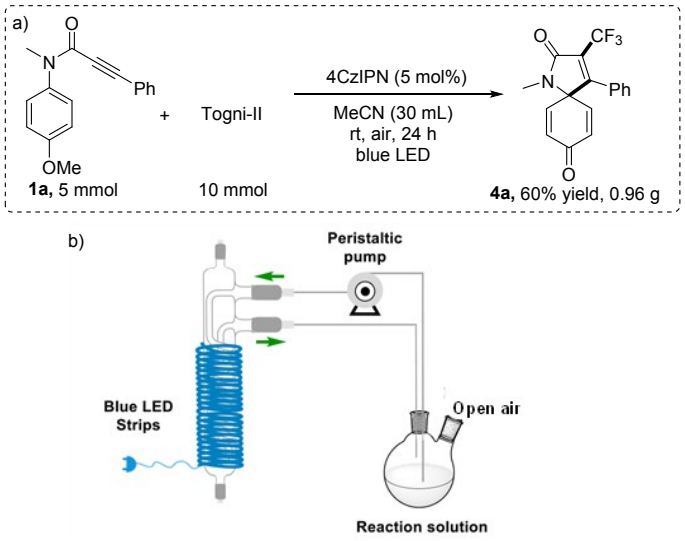


Scheme S3. General experimental procedures for the pyrrolo-[2,1-j]-quinolones

General procedure: The mixture of **1''** (0.2 mmol), diphenylphosphorus Oxide (0.4 mmol), 4CzIPN (5 mol%), LPO (2 qeuiv) and MeCN/H_2O (2 mL/0.1 mL) were sequentially added in a 25 mL reaction vessel. Then the reaction vessel was exposed to 3 W blue LED irradiation at room temperature for 3 h (open-air). After the reaction was completed, the solvent was evaporated under vacuum, all the crude products were purified by thin-layer chromatography using petroleum ethyl acetate as eluting solvent to give the desired products **3a''**.

2.3 The gram-scale synthesis

The mixture of *N*-arylpropiolamides **1''** (0.2 mmol), Togni-II (0.4 mmol), 4CzIPN (5 mol%) and MeCN (2.5 mL) were sequentially added in a 25 mL reaction vessel. Then the reaction vessel was exposed to 10 W blue LED irradiation at room temperature with stirring for 12 h (open-air). After the reaction, the solvent was evaporated under vacuum, all the crude products were purified by silica gel chromatography using petroleum ether/ethyl acetate (v/v = 3/1) as eluting solvent to give the desired products **4a**.

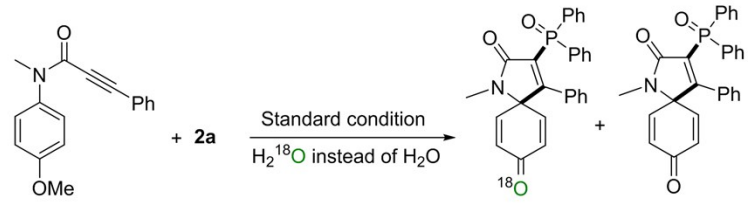


Scheme S4. Specially designed reactor for the gram-scale synthesis

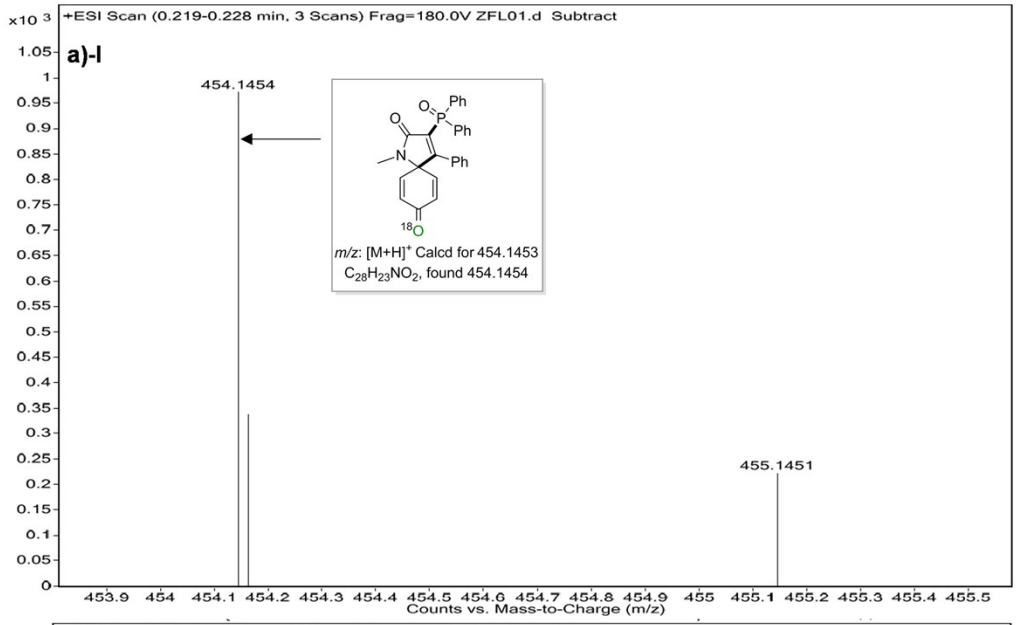
Specially designed reactor for the gram-scale synthesis: Gram-scale synthesis of **4a** with blue LEDs light irradiation in air atmosphere: **1a** (5 mmol), Togni-II (10 mmol), 4CzIPN (5 mol%) in 30 mL MeCN at room temperature for 24 h with the assistance of specially designed reactor. An isolated yield of **4a** (60%, 0.96 g) was given.

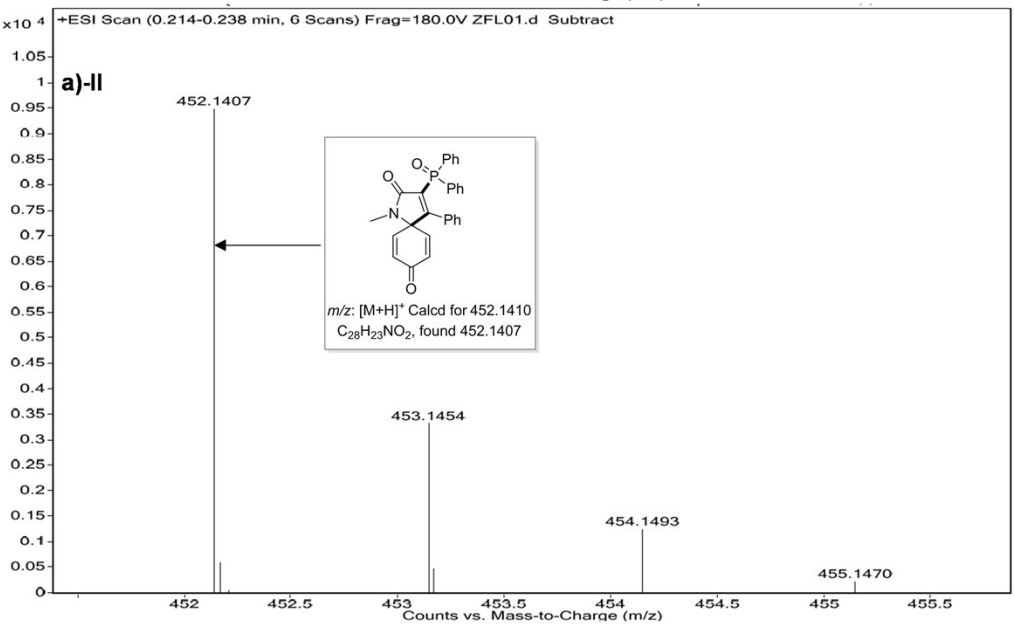
2.4 HRMS data analysis

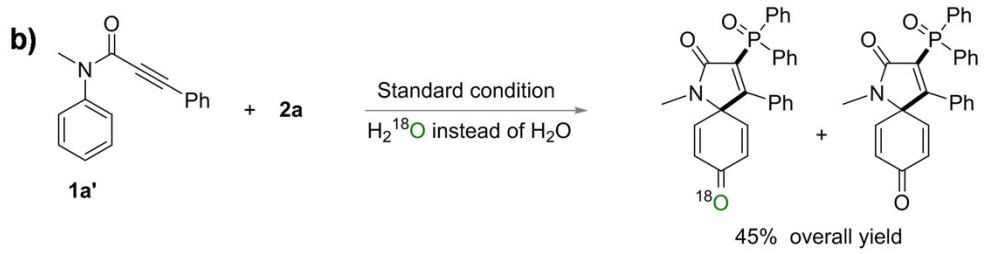
a)

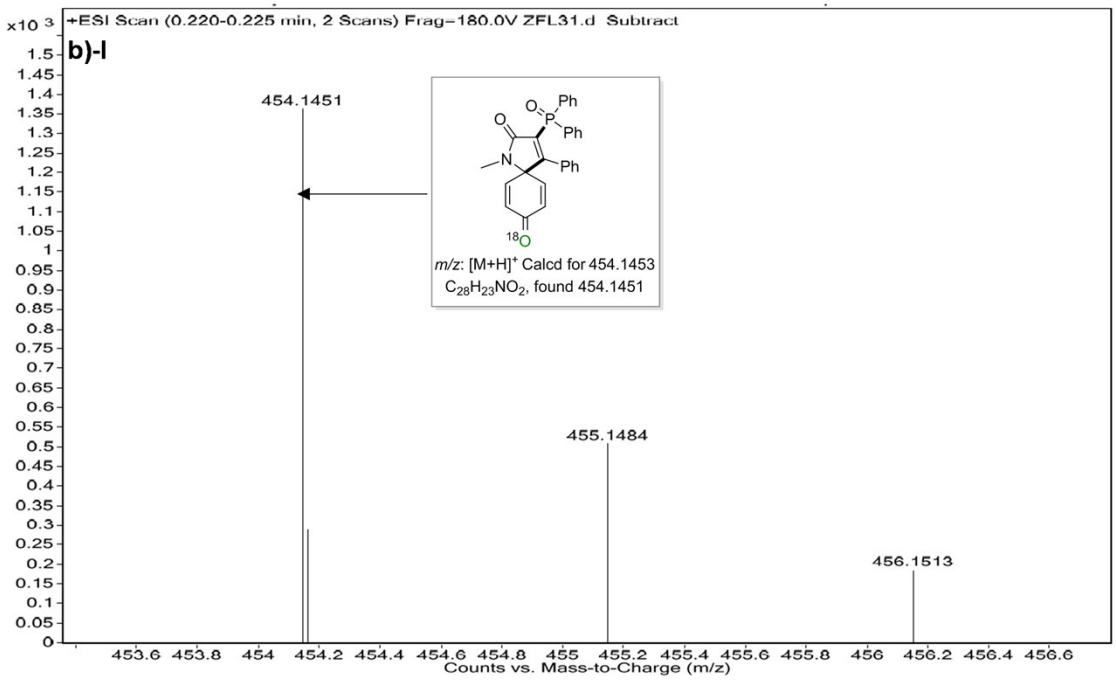


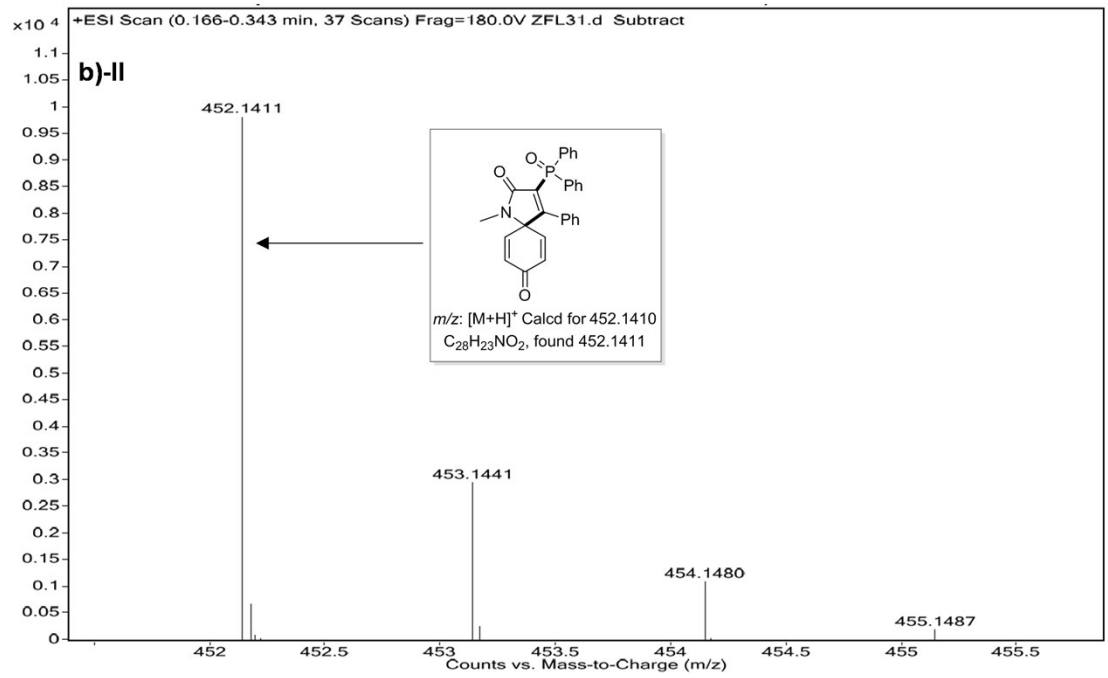
77% , overall yield

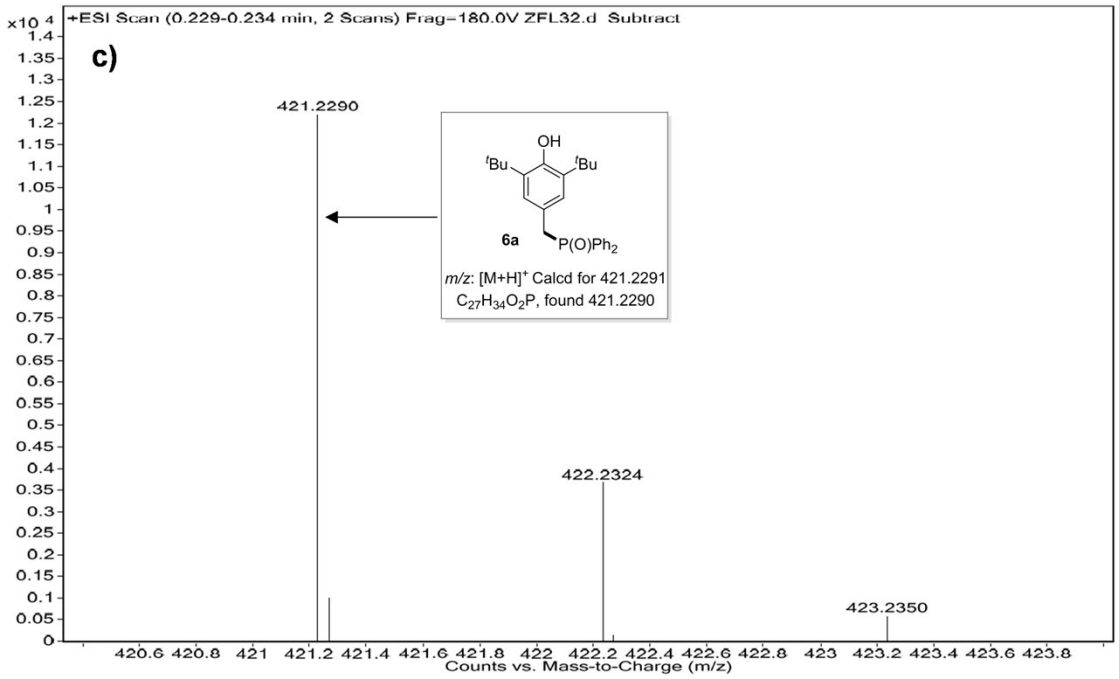


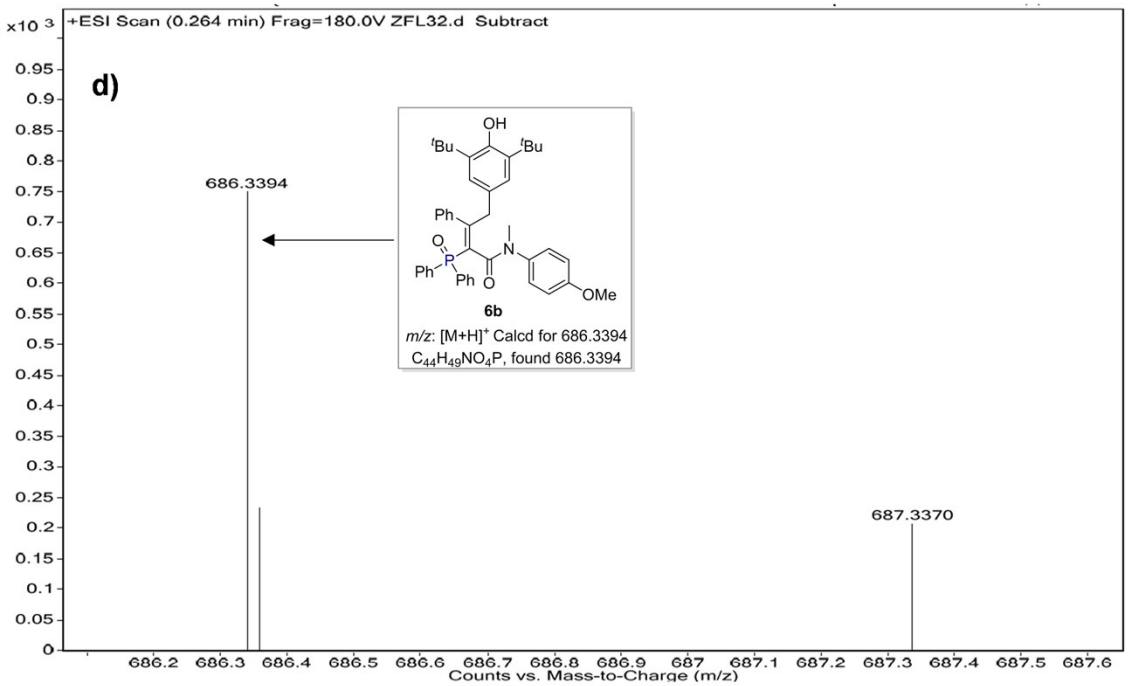



b)
Ph
1a'
+ 2a
Standard condition
H2 18O instead of H2O
Ph
Ph
Ph
18O
+
Ph
Ph
Ph
O
45% overall yield


+ESI Scan (0.220-0.225 min, 2 Scans) Frag=180.0V ZFL31.d Subtract
b)-I
454.1451
455.1484
456.1513
m/z: [M+H]+ Calcd for 454.1453
C28H23NO2, found 454.1451
Counts vs. Mass-to-Charge (m/z)


+ESI Scan (0.166-0.343 min, 37 Scans) Frag=180.0V ZFL31.d Subtract
b)-II
452.1411
453.1441
454.1480
455.1487
m/z: [M+H]+ Calcd for 452.1410
C28H23NO2, found 452.1411
Counts vs. Mass-to-Charge (m/z)





2.5 Procedure for emission quenching experiments

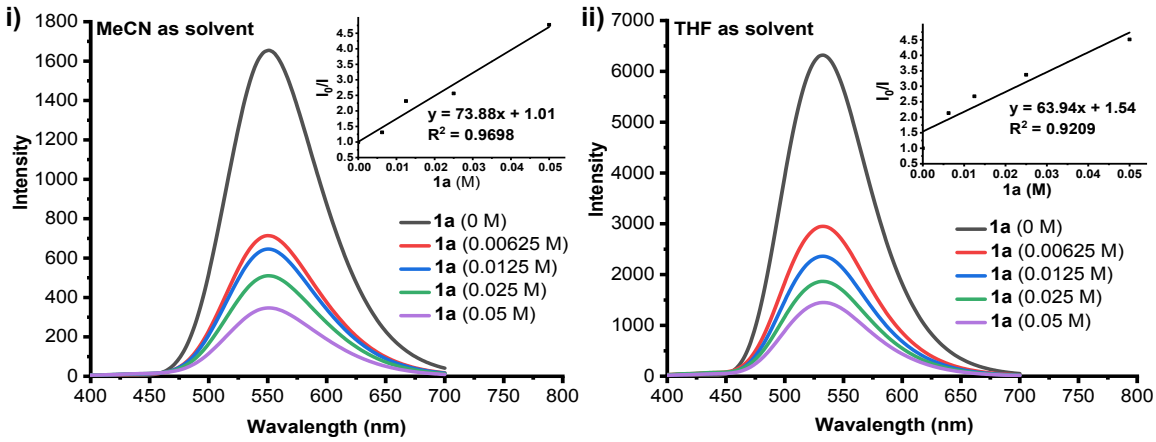


Figure S2a. The Stern-Volmer fluorescence quenching of **1a**

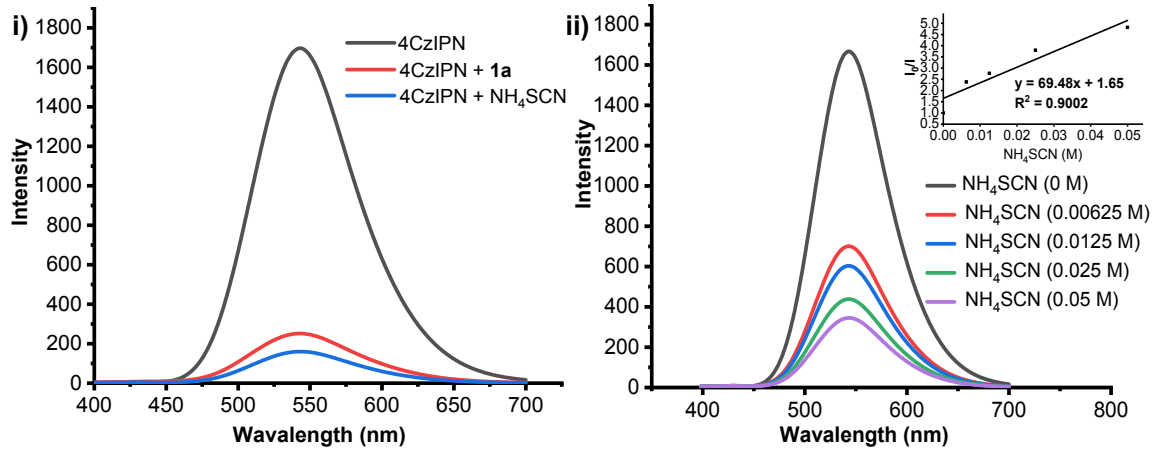


Figure S2b. The Stern-Volmer fluorescence quenching of NH_4SCN

Emission intensities were recorded using an F-4600 FL Spectrophotometer. First, the emission intensity of 4CzIPN solutions was observed at 550 nm. The solutions were irradiated at 378 nm (Maximum absorption wavelength of 4CzIPN) and fluorescence was measured from 400 nm to 700 nm. In a typical experiment, the emission spectrum of a 5×10^{-5} M solution of 4CzIPN with different concentration of **1a**, **2a**, Togni-II, and LPO in degassed anhydrous CH_3CN or THF in 10 mm path length quartz cuvette was collected: Figure 2-i) the emission spectra of 5×10^{-5} M solutions of 4CzIPN with reactants (**1a**, **2a**, and LPO) in degassed anhydrous CH_3CN; ii) the emission spectra of 5×10^{-5} M solutions of 4CzIPN with reactants (**1a** and Togni-II) in degassed anhydrous CH_3CN; Figure S2ai-ii) the emission spectra of a 5×10^{-5} M solution of 4CzIPN with various concentrations of **1a** in degassed anhydrous CH_3CN/THF and the linear relationship between I_0/I and the increasing concentration of **1a** (I_0 and I are the fluorescence intensities before and after the increasing the concentration of **1a**, respectively); Figure S2b: i) the emission spectra of 5×10^{-5} M solutions of 4CzIPN with reactants (**1a** and NH_4SCN) in degassed anhydrous CH_3CN; ii) The emission spectra of a 5×10^{-5} M solution of 4CzIPN with various concentrations of NH_4SCN in degassed anhydrous CH_3CN; and the linear relationship between I_0/I and the increasing

concentration of NH_4SCN (I_0 and I are the fluorescence intensities before and after the increasing the concentration of NH_4SCN, respectively).

2.6 Cyclic voltammograms of NH_4SCN

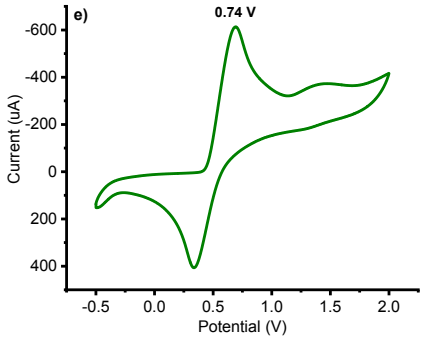


Figure S3. Cyclic voltammograms of 0.1 M $LiClO_4$ and related compounds in CH_3CN using Pt working electrode, Pt wire, and saturated calomel electrode (SCE) as counter and reference electrode at 100 mV/s scan rate. NH_4SCN (0.02 M).

2.7 Proposed Reaction Mechanisms

We proposed a reasonable mechanism for the synthesis of **4a** (Figure S4-i). Initially, 4CzIPN is excited to form 4CzIPN*, which subsequently transfers its energy to **1a**, leading to **1a***. Then a SET process from **1a*** to Togni-II occurs, rendering *N*-arylpropiolamide radical cation **1ab**, 2-iodobenzonate anion (**C**) as well as CF_3 radical. After that, CF_3 radical regioselectively adds to the triple bond of **1a**, resulting in the formation of alkenyl radical intermediate **7a**, which then undergoes an intramolecular cyclization, giving the azaspiro radical **7b**. Then by another additional SET process, **7b** is oxidized by **1ab** to resonance-stable cation **7c**, with the regeneration of a molecule of **1a**. **7c** continuously reacts with iodobenzonate **C**, leading to the desired **4a** together with methyl 2-iodobenzonate. As show as Figure S4-ii, 4CzIPN is first excited by blue light to its excited state (4CzIPN*). Then, a single-electron transfer (SET) process from SCN^- to 4CzIPN* occurs, resulting in the formation of SCN radical, meanwhile, 4CzIPN* is changed into its radical anion ($4CzIPN^{\bullet-}$), which afterwards is oxidized by O_2 in air to regenerate 4CzIPN for next photocatalytic cycle. Following that, SCN radical regioselectively adds to the triple bond of **1a**, rendering alkenyl radical **8a**, which then undergoes an intramolecular cyclization to give the corresponding azaspiro radical (**8b**). By another additional SET process, **8b** is oxidized by O_2 in air to resonance-stable cation **7c**. At last, cation **8c** reacts with a water molecule nearby, leading to the formation of target product **5a** together with a methanol molecule.

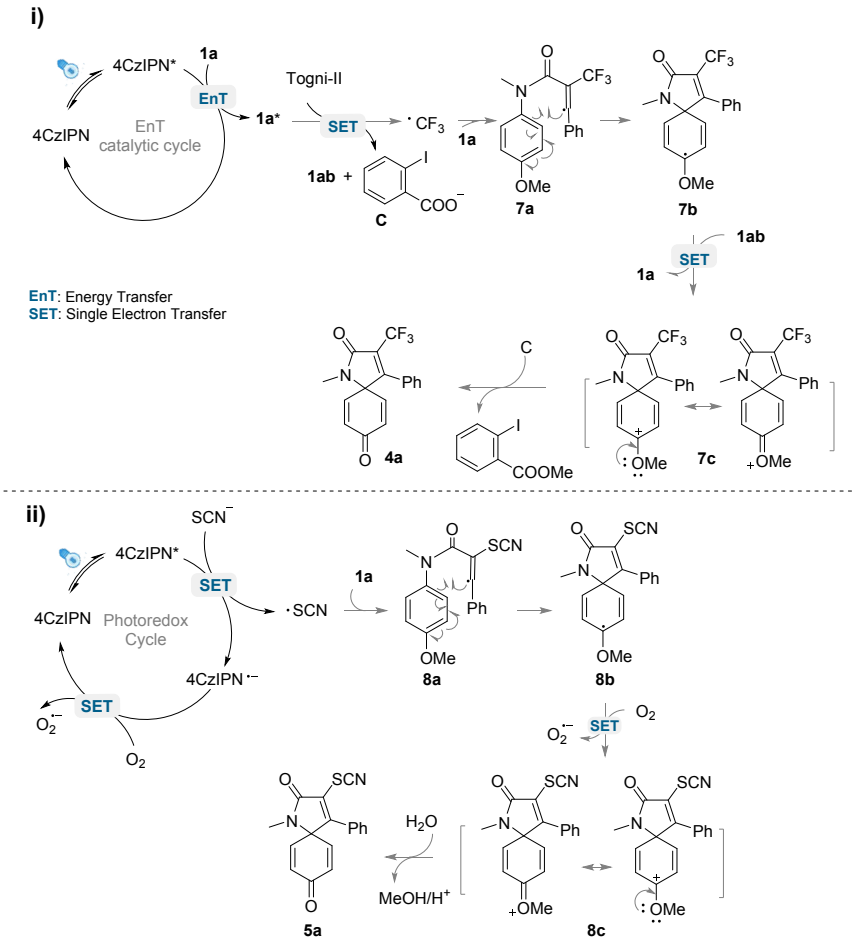
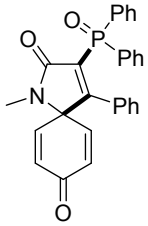


Figure S4. Proposed Reaction Mechanisms

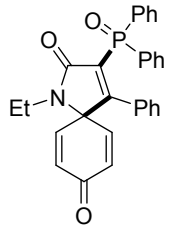
3. Characterization Data for Products

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



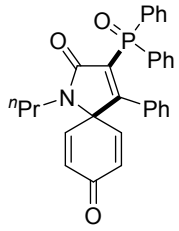
Light yellow solid (72 mg, 80% yield), mp 83 – 85 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.78 – 7.75 (m, 4H), 7.51 – 7.45 (m, 2H), 7.43 – 7.30 (m, 4H), 7.28 – 7.21 (m, 1H), 7.15 (dd, *J* = 8.6, 6.9 Hz, 2H), 7.11 – 7.05 (m, 2H), 6.51 (d, *J* = 10.2 Hz, 2H), 6.44 (d, *J* = 10.2 Hz, 2H), 2.84 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.53, 168.74 (d, *J* = 4.7 Hz), 168.20 (d, *J* = 14.1 Hz), 143.76, 133.75, 132.03 (d, *J* = 2.9 Hz), 131.66 (d, *J* = 110.0 Hz), 131.59 (d, *J* = 10.3 Hz), 131.46 (d, *J* = 100.2 Hz), 130.15 (d, *J* = 2.7 Hz), 129.93, 128.35 (d, *J* = 12.8 Hz), 128.13, 127.71, 69.63 (d, *J* = 10.9 Hz), 26.17. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.41.

*3-(diphenylphosphoryl)-1-ethyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3b**)*



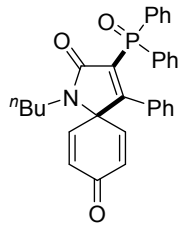
White solid (77 mg, 83% yield), mp 232 – 234 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.79 – 7.74 (m, 4H), 7.51 – 7.43 (m, 2H), 7.40 – 7.36 (m, 4H), 7.24 – 7.18 (m, 1H), 7.12 (dd, *J* = 8.6, 6.9 Hz, 2H), 7.07 – 7.01 (m, 2H), 6.57 (d, *J* = 10.1 Hz, 2H), 6.40 (d, *J* = 10.1 Hz, 2H). 3.31 (q, *J* = 7.2 Hz, 2H), 1.15 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 184.29, 169.12 (d, *J* = 4.4 Hz), 168.71 (d, *J* = 14.7 Hz), 151.83, 143.90, 133.27, 132.55, 131.86 (d, *J* = 112.0 Hz), 131.71 (d, *J* = 140.0 Hz), 130.29, 129.93 (d, *J* = 2.8 Hz), 128.40 (dd, *J* = 12.8, 3.6 Hz), 128.05, 127.86, 71.86 (d, *J* = 10.7 Hz), 25.77, 17.85. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 17.92. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{29}H_{25}NO_3P$, 466.1567; Found: 466.1568.

*3-(diphenylphosphoryl)-4-phenyl-1-propyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3c**)*



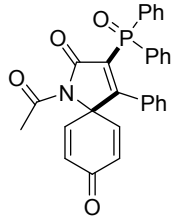
White solid (70 mg, 73% yield), mp 246 – 247 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.79 – 7.74 (m, 4H), 7.51 – 7.44 (m, 2H), 7.40 – 7.36 (m, 4H), 7.25 – 7.19 (m, 1H), 7.13 (t, *J* = 7.7 Hz, 2H), 7.09 – 7.02 (m, 2H), 6.56 (d, *J* = 10.1 Hz, 2H), 6.39 (d, *J* = 10.1 Hz, 2H), 3.27 – 3.13 (t, d, *J* = 7.8 Hz, 2H), 1.62 – 1.50 (m, 2H), 0.84 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.75, 168.55 (d, *J* = 4.8 Hz), 168.34 (d, *J* = 14.1 Hz), 144.25, 133.19, 131.98 (d, *J* = 3.0 Hz), 131.70 (d, *J* = 100.6 Hz), 131.62 (d, *J* = 110.0 Hz), 131.52 (d, *J* = 10.3 Hz), 130.08 (d, *J* = 2.8 Hz), 129.81, 128.32 (d, *J* = 12.8 Hz), 128.16, 127.65, 70.03 (d, *J* = 10.7 Hz), 43.12, 22.99, 11.33. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.01. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{30}H_{27}NO_3P$, 480.1723; Found: 480.1724.

*1-butyl-3-(diphenylphosphoryl)-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3d**)*



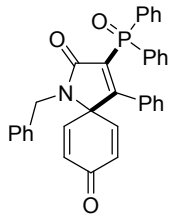
White solid (75 mg, 76% yield), mp 232 – 234 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.82 – 7.70 (m, 4H), 7.47 (td, *J* = 7.3, 1.6 Hz, 2H), 7.40 – 7.36 (m, 4H), 7.25 – 7.19 (m, 1H), 7.12 (t, *J* = 7.7 Hz, 2H), 7.04 (d, *J* = 7.4 Hz, 2H), 6.55 (d, *J* = 10.1 Hz, 2H), 6.39 (d, *J* = 10.0 Hz, 2H), 3.22 (dd, *J* = 8.9, 6.9 Hz, 2H), 1.59 – 1.45 (m, 2H), 1.31 – 1.19 (m, 3H), 0.86 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.74, 168.49 (d, *J* = 4.8 Hz), 168.29 (d, *J* = 13.9 Hz), 144.28, 133.19, 131.95 (d, *J* = 2.9 Hz), 131.79 (d, *J* = 100.4 Hz), 131.70 (d, *J* = 109.9 Hz), 131.52 (d, *J* = 10.3 Hz), 130.10 (d, *J* = 2.9 Hz), 129.80, 128.31 (d, *J* = 12.8 Hz), 128.15, 127.64, 70.04 (d, *J* = 10.8 Hz), 41.37, 31.77, 20.20, 13.66. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 17.85. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{31}H_{29}NO_3P$, 494.1880; Found: 494.1879.

*1-acetyl-3-(diphenylphosphoryl)-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3e**)*



Light yellow solid (43 mg, 45% yield), mp 241 – 242 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.70 – 7.64 (m, 4H), 7.49 – 7.44 (m, 2H), 7.39 – 7.34 (m, 4H), 7.20 – 7.14 (m, 1H), 7.05 (dd, *J* = 8.5, 7.1 Hz, 2H), 6.89 – 6.87 (m, 2H), 6.56 (d, *J* = 10.0 Hz, 2H), 6.32 (d, *J* = 10.0 Hz, 2H), 2.52 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.60, 171.15 (d, *J* = 4.6 Hz), 168.96, 166.95 (d, *J* = 13.9 Hz), 144.51, 142.82, 132.72, 132.28 (d, *J* = 3.0 Hz), 131.33 (d, *J* = 10.4 Hz), 130.60 (d, *J* = 130.9 Hz)，130.41 (d, *J* = 93.3 Hz)， 129.95, 128.54 (d, *J* = 12.9 Hz), 128.38, 127.41, 69.65 (d, *J* = 9.5 Hz), 25.73. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 17.70. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{29}H_{23}NO_4P$, 480.1359; Found: 480.1360.

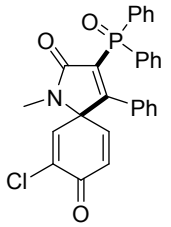
*1-benzyl-3-(diphenylphosphoryl)-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3f**)*



Light yellow solid (79 mg, 75% yield), mp 228 – 229 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.83 – 7.78 (m, 4H), 7.53 – 7.48 (m, 2H), 7.46 – 7.40 (m, 4H), 7.28 – 7.23 (m, 3H), 7.22 – 7.15 (m, 3H), 7.10 (dd, *J* = 8.5, 7.0 Hz, 2H), 7.05 – 6.96 (m, 2H), 6.34 (d, *J* = 10.1 Hz, 2H), 6.22 (d, *J* = 10.1 Hz, 2H), 4.51 (s, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.68, 168.90 (d, *J* = 4.7 Hz), 168.31 (d, *J* = 14.0 Hz), 143.94, 137.18, 132.84, 132.57, 132.00 (d, *J* = 2.9 Hz), 131.65 (d, *J* = 110.0 Hz), 131.54 (d, *J* = 10.3 Hz), 131.36 (d, *J* = 100.4 Hz), 129.96 – 129.67 (m), 128.87, 128.54 (d, *J* = 5.1 Hz), 128.35 (d, *J* = 12.8 Hz), 128.17, 127.94, 127.57, 69.96 (d, *J* = 10.7 Hz), 44.77. ^{31}P NMR (162

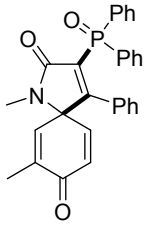
MHz, Chloroform-*d*) δ 18.04. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{34}H_{27}NO_3P$, 528.1723; Found: 528.1722.

7-chloro-3-(diphenylphosphoryl)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione ***(3g)***



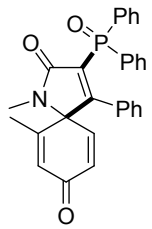
White solid (58 mg, 60% yield), mp 122 – 123 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.82 – 7.73 (m, 4H), 7.53 – 7.36 (m, 6H), 7.27 – 7.22 (m, 1H), 7.21 – 7.13 (m, 2H), 7.07 – 7.02 (m, 2H), 6.99 (d, *J* = 2.7 Hz, 1H), 6.55 (dd, *J* = 9.8, 2.8 Hz, 1H), 6.49 (d, *J* = 9.9 Hz, 1H), 2.87 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 176.62, 167.91 (d, *J* = 14.1 Hz), 167.52 (d, *J* = 4.8 Hz), 144.06 (d, *J* = 51.9 Hz), 139.52, 137.04, 132.66, 132.27 – 131.93 (m), 131.88 (d, *J* = 99.3 Hz), 131.58 (dd, *J* = 10.4, 2.4 Hz), 131.35 (dd, *J* = 110.1, 13.8 Hz), 130.10, 129.63 (d, *J* = 2.7 Hz), 128.79, 128.41 (dd, *J* = 12.8, 3.8 Hz), 127.96 (d, *J* = 18.2 Hz), 72.11 (d, *J* = 10.8 Hz), 26.45. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.04. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{28}H_{22}ClNO_3P$, 486.1020; Found: 486.1019.

3-(diphenylphosphoryl)-1,7-dimethyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione ***(3h)***



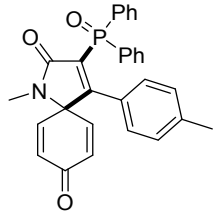
Light yellow oil (70 mg, 75% yield); ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.81 – 7.75 (m, 4H), 7.51 – 7.45 (m, 2H), 7.43 – 7.36 (m, 4H), 7.25 – 7.19 (m, 1H), 7.17 – 7.10 (m, 2H), 7.07 – 7.01 (m, 2H), 6.47 (dd, *J* = 9.8, 3.0 Hz, 1H), 6.39 (d, *J* = 9.8 Hz, 1H), 6.28 (dd, *J* = 3.0, 1.5 Hz, 1H), 2.82 (s, 3H), 1.89 (d, *J* = 1.5 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 176.75, 167.85 (d, *J* = 21.2 Hz), 167.80 (d, *J* = 2.4 Hz), 144.40, 139.52, 137.04, 132.66, 132.15 (t, *J* = 2.8 Hz), 131.89 (d, *J* = 99.3 Hz), 131.58 (dd, *J* = 10.4, 1.7 Hz), 131.37 (dd, *J* = 110.3, 12.1 Hz), 130.11, 129.65 (d, *J* = 2.8 Hz), 128.41 (dd, *J* = 12.8, 2.9 Hz), 128.03 (d, *J* = 1.3 Hz), 127.88, 71.38 (d, *J* = 11.0 Hz), 26.41. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.26. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{29}H_{25}NO_3P$, 466.1567; Found: 466.1568.

3-(diphenylphosphoryl)-1,6-dimethyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione ***(3i)***



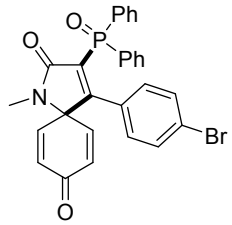
Light yellow oil (42 mg, 45% yield); ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.83 – 7.75 (m, 4H), 7.52 – 7.47 (m, 2H), 7.45 – 7.39 (m, 4H), 7.31 – 7.21 (m, 1H), 7.20 – 7.06 (m, 4H), 6.52 – 6.40 (m, 2H), 6.32 (t, *J* = 1.5 Hz, 1H), 2.74 (s, 3H), 1.80 (d, *J* = 1.4 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 184.29, 169.12 (d, *J* = 4.4 Hz), 168.71 (d, *J* = 14.7 Hz), 151.83, 143.90, 133.27, 132.55, 132.09 (d, *J* = 3.1 Hz), 131.92 (d, *J* = 100.4 Hz), 131.85 (d, *J* = 110.3 Hz), 131.52 (dd, *J* = 16.1, 10.4 Hz), 131.01, 130.29, 129.93 (d, *J* = 2.8 Hz), 128.40 (dd, *J* = 12.8, 3.6 Hz), 127.95 (d, *J* = 18.6 Hz), 71.86 (d, *J* = 10.7 Hz), 25.77, 17.85. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.43. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{29}H_{25}NO_3P$, 466.1567; Found: 466.1568.

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



White solid (84 mg, 90% yield), mp 270 – 272 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.86 – 7.74 (m, 4H), 7.52 – 7.48 (m, 2H), 7.44 – 7.39 (m, 4H), 7.09 – 6.92 (m, 4H), 6.58 – 6.40 (m, 4H), 2.84 (s, 3H), 2.26 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.66, 169.07 (d, *J* = 4.6 Hz), 168.34 (d, *J* = 14.6 Hz), 143.99, 140.19, 133.65, 131.92 (d, *J* = 3.0 Hz), 131.81 (d, *J* = 109.8 Hz), 131.62 (d, *J* = 10.4 Hz), 130.84 (d, *J* = 100.9 Hz), 128.42 (d, *J* = 6.7 Hz), 128.26, 128.10, 127.28 (d, *J* = 2.8 Hz), 69.59 (d, *J* = 10.9 Hz), 26.11, 21.35. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.33. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{29}H_{25}NO_3P$, 466.1567; Found: 466.1567.

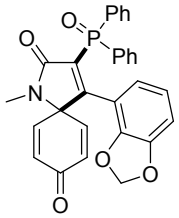
*4-(4-bromophenyl)-3-(diphenylphosphoryl)-1-methyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3k**)*



Light yellow solid (70 mg, 66% yield), mp 260 – 261 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.81 – 7.75 (m, 4H), 7.56 – 7.48 (m, 2H), 7.46 – 7.38 (m, 4H), 7.30 (d, *J* = 8.5 Hz, 2H), 6.96 (d, *J* = 8.5 Hz, 2H), 6.54 – 6.40 (m, 4H), 2.84 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.28,

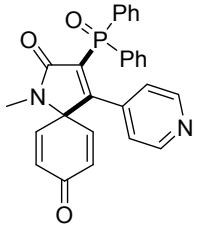
167.94 (d, *J* = 14.1 Hz), 167.43 (d, *J* = 4.4 Hz), 143.42, 133.98, 132.27 (d, *J* = 99.3 Hz), 132.22 (d, *J* = 2.9 Hz), 131.60 (d, *J* = 10.6 Hz), 131.33 (d, *J* = 110.0 Hz), 131.01, 129.69, 129.00 (d, *J* = 2.9 Hz), 128.48 (d, *J* = 12.8 Hz), 124.66, 69.52 (d, *J* = 10.7 Hz), 26.23. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.47. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{28}H_{22}BrNO_3P$, 530.0515; Found: 530.0515.

*4-(benzo[d][1,3]dioxol-4-yl)-3-(diphenylphosphoryl)-1-methyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3l**)*



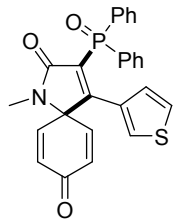
Light yellow solid (69 mg, 70% yield), mp 262 – 263 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.86 – 7.73 (m, 4H), 7.55 – 7.47 (m, 2H), 7.44 – 7.40 (m, 4H), 6.66 (dd, *J* = 8.1, 1.8 Hz, 1H), 6.60 – 6.54 (m, 2H), 6.52 – 6.44 (m, 4H), 5.90 (s, 2H), 2.82 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.65, 168.30 (d, *J* = 4.9 Hz), 168.10 (d, *J* = 5.2 Hz), 149.19, 146.95, 143.98, 133.68, 132.02 (d, *J* = 2.9 Hz), 131.55 (d, *J* = 10.4 Hz), 131.53 (d, *J* = 110.1 Hz), 130.76 (d, *J* = 101.2 Hz), 128.38 (d, *J* = 12.8 Hz), 123.64, 122.99, 108.68, 101.49, 69.46 (d, *J* = 10.7 Hz), 26.08. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.59. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{29}H_{23}NO_5P$, 496.1308; Found: 496.1306.

*3-(diphenylphosphoryl)-1-methyl-4-(pyridin-4-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3m**)*



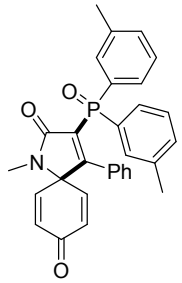
Light yellow solid (58 mg, 64% yield), mp 258 – 259 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.51 – 8.43 (m, 2H), 7.85 – 7.76 (m, 4H), 7.58 – 7.52 (m, 2H), 7.48 – 7.43 (m, 4H), 7.00 – 6.92 (m, 2H), 6.52 – 6.43 (m, 4H), 2.86 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 182.90, 167.53 (d, *J* = 13.9 Hz), 165.37 (d, *J* = 4.1 Hz), 149.11, 142.66, 138.31 (d, *J* = 2.9 Hz), 134.28, 133.66 (d, *J* = 97.0 Hz), 132.52 (d, *J* = 2.9 Hz), 131.63 (d, *J* = 10.6 Hz), 130.93 (d, *J* = 110.2 Hz), 128.58 (d, *J* = 12.9 Hz), 122.52, 69.41 (d, *J* = 10.5 Hz), 26.31. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.48. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{27}H_{22}N_2O_3P$, 453.1363; Found: 453.1361.

*3-(diphenylphosphoryl)-1-methyl-4-(thiophen-3-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3n**)*



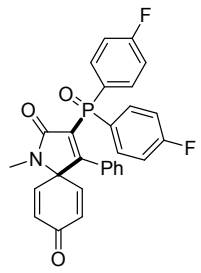
Light yellow solid (58 mg, 63% yield), mp 179 – 181 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.83 – 7.77 (m, 4H), 7.71 (t, *J* = 2.1 Hz, 1H), 7.54 – 7.49 (m, 2H), 7.46 – 7.40 (m, 4H), 7.18 – 7.07 (m, 2H), 6.53 (s, 4H), 2.79 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.70, 168.23 (d, *J* = 14.3 Hz), 162.89 (d, *J* = 4.3 Hz), 144.57, 133.49, 132.08 (d, *J* = 2.9 Hz), 131.55 (d, *J* = 10.3 Hz), 131.49 (d, *J* = 110.1 Hz), 130.45 (d, *J* = 3.2 Hz), 129.33, 129.03 (d, *J* = 101.3 Hz),128.39 (d, *J* = 12.8 Hz), 127.77, 125.70, 68.47 (d, *J* = 10.8 Hz), 25.82. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 18.49. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{26}H_{21}NO_3PS$, 458.0974; Found: 458.0975.

*3-(di-m-tolylphosphoryl)-1-methyl-4-phenyl-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**3o**)*



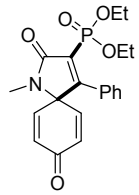
White solid (62 mg, 65% yield), mp 222 – 224 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.42 – 7.31 (m, 4H), 7.23 – 7.18 (m, 5H), 7.12 (t, *J* = 7.6 Hz, 2H), 7.01 (d, *J* = 7.1 Hz, 2H), 6.60 – 6.39 (m, 4H), 2.86 (s, 3H), 2.47 (s, 6H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.57, 168.27 (d, *J* = 4.7 Hz), 168.11 (d, *J* = 13.3 Hz), 143.86, 142.87 (d, *J* = 8.6 Hz), 133.78, 132.65 (d, *J* = 13.3 Hz), 132.10 (d, *J* = 2.8 Hz), 131.78 (d, *J* = 11.1 Hz), 130.69 (d, *J* = 123.6 Hz), 130.10, 129.76 (d, *J* = 108.6 Hz), 129.75, 127.93, 127.55, 125.41 (d, *J* = 13.4 Hz), 69.69 (d, *J* = 10.6 Hz), 26.26, 21.69 (d, *J* = 4.6 Hz). ^{31}P NMR (162 MHz, Chloroform-*d*) δ 25.02. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{30}H_{27}NO_3P$, 480.1723; Found: 480.1725.

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



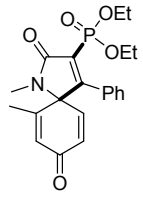
Light yellow solid (70 mg, 72% yield), mp 258 – 259 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.84 – 7.73 (m, 4H), 7.31 – 7.26 (m, 1H), 7.19 (t, *J* = 7.7 Hz, 2H), 7.21 – 7.17 (m, 6H), 6.55 – 6.40 (m, 4H), 2.85 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.36, 169.26 (d, *J* = 4.8 Hz), 168.12 (d, *J* = 14.6 Hz), 165.15 (dd, *J* = 254.1, 3.4 Hz), 143.36, 134.11 (dd, *J* = 12.0, 8.9 Hz), 133.90, 131.02 (d, *J* = 102.5 Hz), 130.09, 129.98 (d, *J* = 2.9 Hz), 127.38 (dd, *J* = 113.8, 3.3 Hz), 115.88 (dd, *J* = 21.5, 14.1 Hz), 115.92, 69.77 (d, *J* = 11.1 Hz), 26.20. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 16.66. ^{19}F NMR (376 MHz, Chloroform-*d*) δ -106.04. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{28}H_{21}F_2NO_3P$, 488.1222; Found: 488.1223.

*Diethyl (1-methyl-2,8-dioxo-4-phenyl-1-azaspiro[4.5]deca-3,6,9-trien-3-yl)phosphonate (**3q**)*[6]



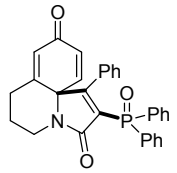
White solid (50 mg, 65% yield), mp 127-129 ℃. ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.42 – 7.30 (m, 3H), 7.26 – 7.15 (m, 2H), 6.61 – 6.38 (m, 4H), 4.17 – 3.96 (m, 4H), 2.88 (s, 3H), 1.10 (t, *J* = 7.1 Hz, 6H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 183.50, 167.79 (d, *J* = 17.9 Hz), 165.35 (d, *J* = 8.2 Hz), 143.61, 137.11, 133.72, 130.97 (d, *J* = 3.2 Hz), 129.94, 128.89 (d, *J* = 203.0 Hz), 127.97 (d, *J* = 5.1 Hz), 69.35 (d, *J* = 15.4 Hz), 62.94 (d, *J* = 6.1 Hz), 26.10, 16.07 (d, *J* = 6.7 Hz). ^{31}P NMR (162 MHz, Chloroform-*d*) δ 7.47.

*Diethyl (1,6-dimethyl-2,8-dioxo-4-phenyl-1-azaspiro[4.5]deca-3,6,9-trien-3-yl)phosphonate (**3r**)*[6]



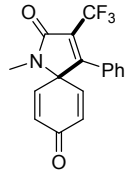
Light yellow oil (34 mg, 42% yield). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.39 (dd, *J* = 6.8, 2.0 Hz, 1H), 7.34 (dd, *J* = 8.4, 6.6 Hz, 2H), 7.27 – 7.22 (m, 2H), 6.49 (d, *J* = 2.2 Hz, 2H), 6.32 (t, *J* = 1.5 Hz, 1H), 4.16 – 4.02 (m, 4H), 2.81 (s, 3H), 1.81 (d, *J* = 1.4 Hz, 3H), 1.13 (td, *J* = 7.1, 1.8 Hz, 6H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 183.24, 167.21 (d, *J* = 17.4 Hz), 164.81 (d, *J* = 7.7 Hz), 151.01, 142.71, 131.82 (d, *J* = 141.7 Hz), 129.71 (d, *J* = 3.4 Hz), 129.22, 128.27 (d, *J* = 201.9 Hz), 127.09, 126.83, 125.52 (d, *J* = 374.9 Hz), 70.59 (d, *J* = 15.0 Hz), 61.93 (dd, *J* = 17.1, 6.0 Hz), 24.71, 16.78, 15.07 (dd, *J* = 6.7, 2.4 Hz). ^{31}P NMR (162 MHz, Chloroform-*d*) δ 7.42.

*2-(diphenylphosphoryl)-1-phenyl-6,7-dihydro-3H-pyrrolo[2,1-j]quinoline-3,9(5H)-dione (**3a''**)*



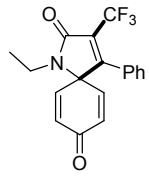
Light yellow solid (76 mg, 80% yield), mp 89 – 90 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.74 (td, *J* = 12.0, 7.0 Hz, 4H), 7.52 – 7.36 (m, 6H), 7.21 (t, *J* = 7.5 Hz, 1H), 7.11 (t, *J* = 7.6 Hz, 2H), 6.90 – 6.81 (m, 2H), 6.58 (d, *J* = 9.7 Hz, 1H), 6.32 – 6.19 (m, 2H), 4.12 (dd, *J* = 14.1, 9.0 Hz, 1H), 2.82 – 2.74 (m, 1H), 2.45 (dd, *J* = 13.0, 8.4 Hz, 2H), 2.08 – 1.95 (m, 1H), 1.83 – 1.77 (m, 1H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ. ^{13}C NMR (101 MHz, Chloroform-*d*) δ 184.06, 172.27 (d, *J* = 13.9 Hz), 170.13 (d, *J* = 5.2 Hz), 157.06, 144.87, 132.96, 132.22, 132.03 (t, *J* = 3.6 Hz), 131.77 (d, *J* = 176.4 Hz), 131.45 (dd, *J* = 16.0, 10.4 Hz), 131.40 (d, *J* = 53.3 Hz), 129.61, 129.34, 129.15 (d, *J* = 2.8 Hz), 128.42, 128.41, 128.29, 128.26, 127.39. ^{31}P NMR (162 MHz, Chloroform-*d*) δ 17.82. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{30}H_{25}NO_3P$, 478.1567; Found: 478.1566.

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



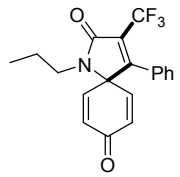
Yellow solid (45 mg, 70% yield), mp 162 – 164 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.45 – 7.39 (m, 1H), 7.35 (dd, *J* = 8.3, 6.6 Hz, 2H), 7.16 – 7.09 (m, 2H), 6.55 – 6.44 (m, 4H), 2.92 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 183.17, 164.54, 159.47 (q, *J* = 3.4 Hz), 142.62, 134.20, 130.35, 128.82, 128.32, 127.52, 126.55 (q, *J* = 33.6 Hz), 120.38 (q, *J* = 272.5 Hz), 67.94, 26.19.

*1-ethyl-4-phenyl-3-(trifluoromethyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**4b**)*



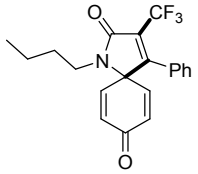
White solid (45 mg, 68% yield), mp 175 – 176 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.47 – 7.29 (m, 3H), 7.16 – 7.06 (m, 2H), 6.57 (d, *J* = 10.1 Hz, 2H), 6.44 (d, *J* = 10.1 Hz, 2H), 3.39 (q, *J* = 7.2 Hz, 2H), 1.23 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 183.38, 164.50, 159.34 (q, *J* = 3.3 Hz), 143.04, 133.66, 130.27, 128.80, 128.28, 127.56, 126.82 (q, *J* = 33.6 Hz), 120.38 (q, *J* = 272.4 Hz), 68.30, 36.35, 15.03. ^{19}F NMR (376 MHz, Chloroform-*d*) δ -60.63. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{18}H_{15}F_3NO_2$, 334.1049; Found: 334.1064.

*4-phenyl-1-propyl-3-(trifluoromethyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**4c**)*



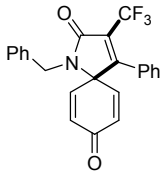
White solid (55 mg, 73% yield), mp 128 – 129 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.44 – 7.31 (m, 3H), 7.14 – 7.06 (m, 2H), 6.55 (d, *J* = 10.2 Hz, 2H), 6.43 (d, *J* = 10.2 Hz, 2H), 3.31 – 3.21 (m, 2H), 1.69 – 1.56 (m, 2H), 0.92 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 183.37, 164.78, 159.32 (q, *J* = 3.5 Hz), 143.13, 133.66, 130.27, 128.80, 128.27, 127.58, 126.81 (q, *J* = 33.7 Hz), 120.37 (q, *J* = 272.6 Hz), 68.33, 43.32, 22.91, 11.38. ^{19}F NMR (376 MHz, Chloroform-*d*) δ -60.65. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{19}H_{17}F_3NO_2$, 348.1206; Found: 348.1217.

*1-butyl-4-phenyl-3-(trifluoromethyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**4d**)*



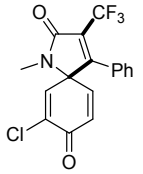
Light yellow solid (52 mg, 72% yield), mp 111 – 113 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.46 – 7.31 (m, 3H), 7.14 – 7.03 (m, 2H), 6.60 – 6.52 (m, 2H), 6.46 – 6.37 (m, 2H), 3.36 – 3.25 (m, 2H), 1.65 – 1.50 (m, 2H), 1.38 – 1.28 (m, 2H), 0.92 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.39, 164.75, 159.28 (q, *J* = 3.4 Hz), 143.13, 133.64, 130.25, 128.77, 128.25, 127.55 (d, *J* = 1.6 Hz), 126.78 (q, *J* = 33.7 Hz), 120.35 (q, *J* = 272.5 Hz), 68.33, 41.47, 31.67, 20.16, 13.62. ^{19}F NMR (376 MHz, Chloroform-*d*) δ -60.65. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{20}H_{19}F_3NO_2$, 362.1362; Found: 362.1362.

*1-benzyl-4-phenyl-3-(trifluoromethyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**4e**)*



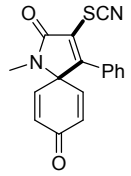
Light yellow solid (51 mg, 65% yield), mp 125 – 127 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.41 – 7.21 (m, 8H), 7.12 – 6.96 (m, 2H), 6.44 – 6.15 (m, 4H), 4.56 (s, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 183.37, 164.71, 159.84, 142.86, 136.72, 133.28, 130.27, 129.02, 128.68, 128.56, 128.19, 128.15, 127.56, 126.42 (q, *J* = 33.6 Hz), 120.40 (q, *J* = 272.0 Hz), 68.29, 44.97. ^{19}F NMR (376 MHz, Chloroform-*d*) δ -60.53. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{23}H_{17}F_3NO_2$, 396.1206; Found: 396.1207.

*7-chloro-1-methyl-4-phenyl-3-(trifluoromethyl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**4f**)*[4]



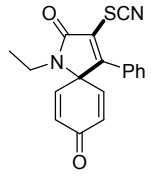
White solid (39 mg, 55% yield), mp 150 – 152 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.46 – 7.35 (m, 3H), 7.14 – 7.07 (m, 2H), 6.75 (d, *J* = 2.2 Hz, 1H), 6.61 – 6.51 (m, 2H), 2.95 (s, 3H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 176.43, 164.19, 158.74 (d, *J* = 3.4 Hz), 143.25, 138.39, 137.55, 133.14, 130.58, 128.51, 128.33, 127.45, 126.80 (q, *J* = 34.0 Hz), 120.22 (q, *J* = 272.6 Hz), 69.67, 26.43.

*1-methyl-4-phenyl-3-thiocyanato-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**5a**)*[3]



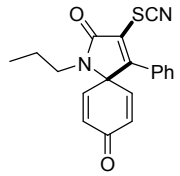
White solid (49 mg, 80% yield), mp 155 – 157 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.52 – 7.41 (m, 3H), 7.31 – 7.22 (m, 2H), 6.53 (s, 4H), 2.95 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.30, 164.95, 156.99, 143.07, 133.97, 131.13, 129.10, 128.87, 127.78, 122.34, 106.26, 68.37, 26.58.

*1-ethyl-4-phenyl-3-thiocyanato-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**5b**)*[3]



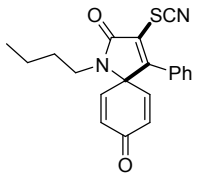
White solid (42 mg, 65% yield), mp 144 – 146 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.48 – 7.44 (m, 3H), 7.28 – 7.18 (m, 2H), 6.63 – 6.45 (m, 4H), 3.41 (q, *J* = 7.2 Hz, 2H), 1.25 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.53, 164.87, 156.72, 143.46, 133.43, 130.98, 129.02, 128.87, 127.81, 122.64, 106.29, 68.81, 36.77, 15.05.

*4-phenyl-1-propyl-3-thiocyanato-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (7**c**)*



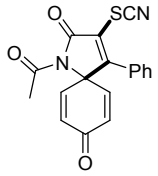
Yellow solid (50 mg, 75% yield), mp 153 – 154 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.49 – 7.39 (m, 3H), 7.27 – 7.20 (m, 2H), 6.62 – 6.54 (m, 2H), 6.52 – 6.42 (m, 2H), 3.35 – 3.23 (m, 2H), 1.70 – 1.61 (m, 2H), 0.92 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.53, 165.16, 156.68, 143.55, 133.41, 130.96, 129.00, 128.86, 127.83, 122.65, 106.28, 68.86, 43.72, 22.90, 11.39. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{19}H_{17}N_2O_2S$, 337.1005; Found: 337.1006.

*1-butyl-4-phenyl-3-thiocyanato-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**5d**)*



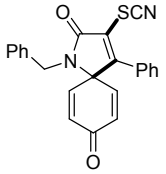
Light yellow solid (64 mg, 91% yield), mp 170 – 171 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.50 – 7.38 (m, 3H), 7.25 – 7.22 (m, 2H), 6.58 (d, *J* = 10.2 Hz, 2H), 6.48 (d, *J* = 10.1 Hz, 2H), 3.37 – 3.30 (m, 2H), 1.65 – 1.56 (m, 2H), 1.38 – 1.28 (m, 2H), 0.91 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.52, 165.11, 156.61, 143.56, 133.41, 130.96, 129.00, 128.87, 127.82, 122.66, 106.26, 68.86, 41.90, 31.65, 20.16, 13.63. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{20}H_{19}N_2O_2S$, 351.1162; Found: 351.1162.

*1-acetyl-4-phenyl-3-thiocyanato-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**5e**)*[3]



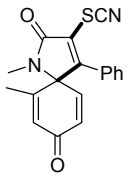
Light yellow solid (30 mg, 45% yield), mp 205 – 207 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.55 – 7.38 (m, 3H), 7.21 – 7.06 (m, 2H), 6.55 (d, *J* = 9.9 Hz, 2H), 6.51 – 6.33 (m, 2H), 2.65 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.39, 168.45, 163.76, 161.54, 141.94, 133.00, 131.47, 128.98, 127.99, 127.56, 122.28, 105.66, 68.53, 25.67.

*1-benzyl-4-phenyl-3-thiocyanato-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**5f**)*[3]



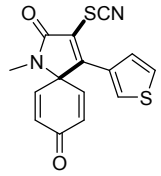
White solid (52 mg, 72% yield), mp 140 – 141 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.47 – 7.35 (m, 3H), 7.31 – 7.23 (m, 5H), 7.19 – 7.16 (m, 2H), 6.40 – 6.24 (m, 4H), 4.59 (s, 2H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.51, 165.16, 157.31, 143.26, 136.55, 133.08, 131.02, 128.99, 128.72, 128.67, 128.22, 127.82, 122.35, 106.26, 68.84, 45.47.

*1,6-dimethyl-4-phenyl-3-thiocyanato-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**5g**)*[3]



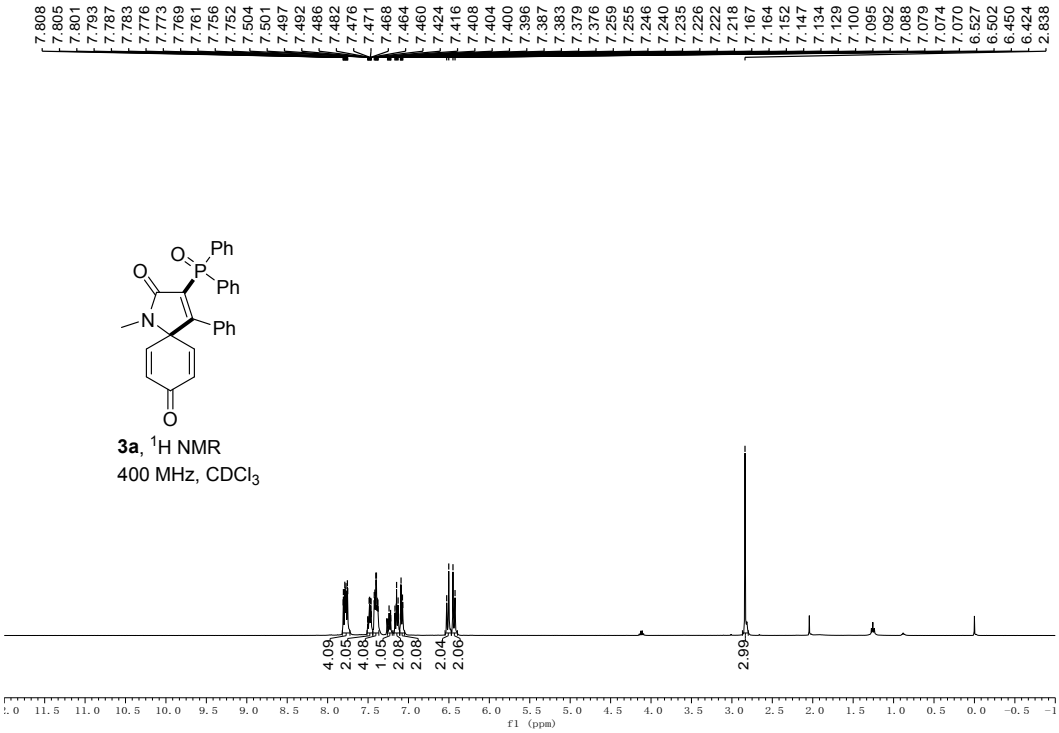
Light yellow solid (50 mg, 78% yield), mp 145 – 146 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.54 – 7.40 (m, 3H), 7.29 – 7.26 (m, 2H), 6.59 – 6.46 (m, 2H), 6.43 – 6.32 (m, 1H), 2.86 (s, 3H), 1.77 (d, *J* = 1.4 Hz, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 184.06, 165.41, 157.41, 151.58, 143.27, 133.45, 132.57, 131.33, 129.26, 128.73, 127.56, 122.33, 106.35, 70.50, 26.15, 17.71.

*1-methyl-3-thiocyanato-4-(thiophen-3-yl)-1-azaspiro[4.5]deca-3,6,9-triene-2,8-dione (**5h**)*

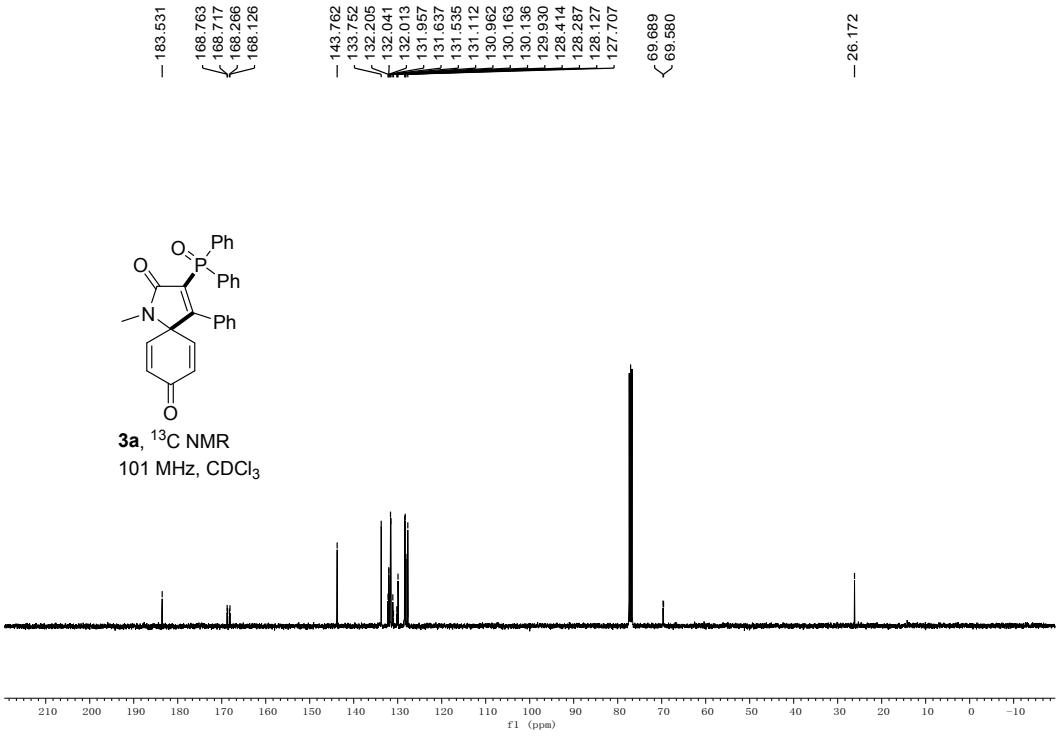


Light yellow solid (50 mg, 80% yield), mp 142 – 144 ℃; ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.75 (dd, *J* = 2.9, 1.5 Hz, 1H), 7.48 – 7.38 (m, 2H), 6.64 – 6.52 (m, 4H), 2.92 (s, 3H). ^{13}C NMR (101 MHz, Chloroform-*d*) δ 183.47, 165.23, 151.69, 144.08, 133.65, 129.45, 128.85, 127.54, 126.35, 118.88, 106.49, 67.26, 26.21. HRMS (ESI) *m/z*: $[M+H]^+$ Calcd for $C_{15}H_{11}N_2O_2S_2$, 315.0256; Found: 315.0225.

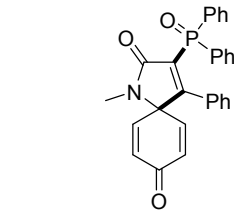
4. NMR Copies of Products



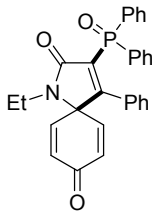
3a, ^{1}H NMR
400 MHz, $CDCl_3$



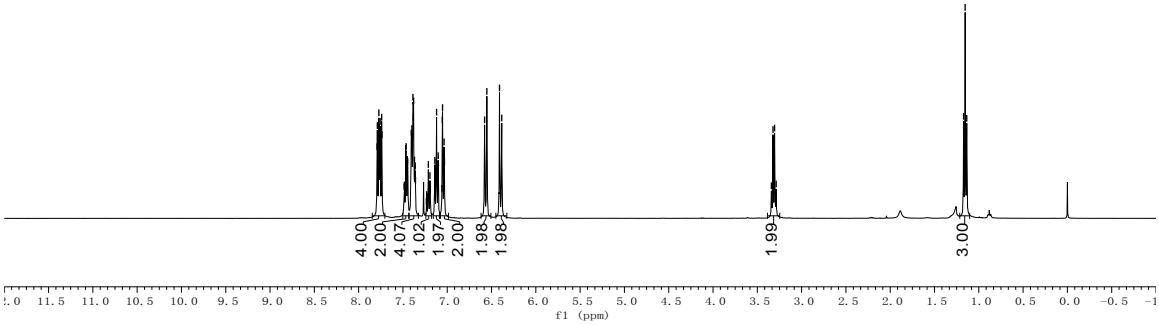
3a, ^{13}C NMR
101 MHz, $CDCl_3$

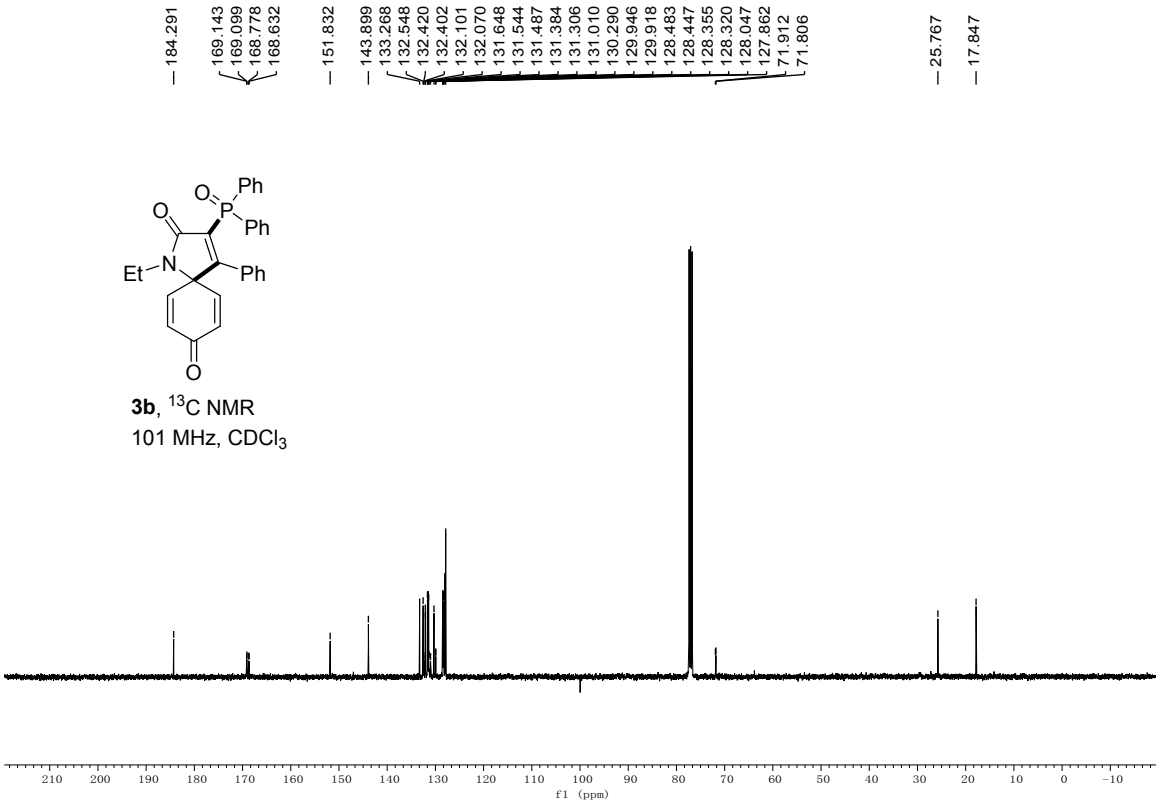


3a, ^{31}P NMR
162 MHz, $CDCl_3$

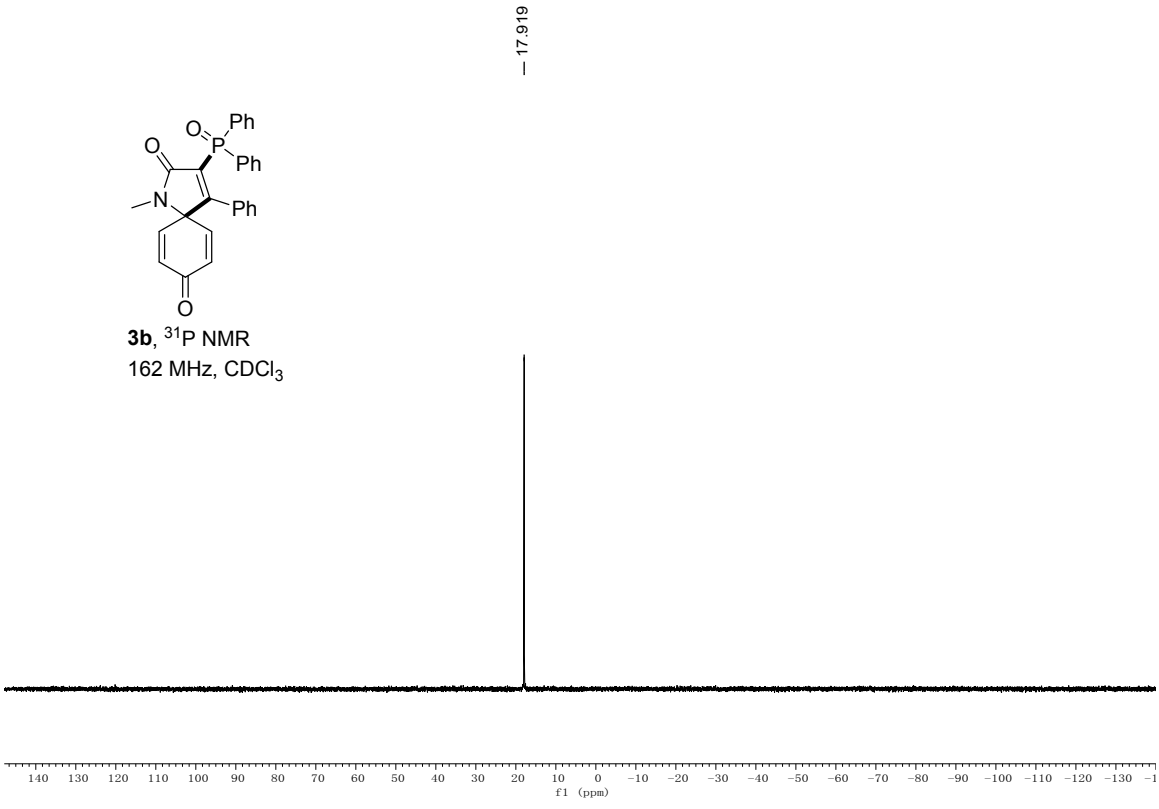


3b, ^{1}H NMR
400 MHz, $CDCl_3$

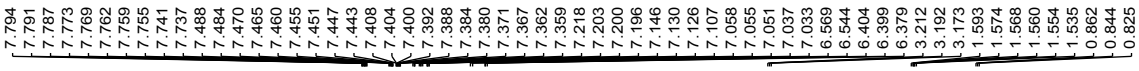


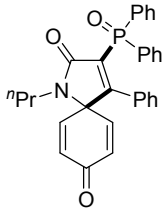


3b, ^{13}C NMR
101 MHz, $CDCl_3$

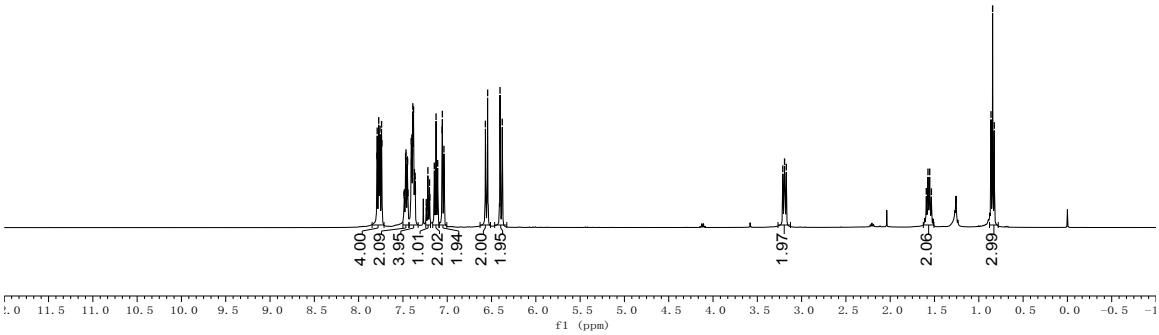


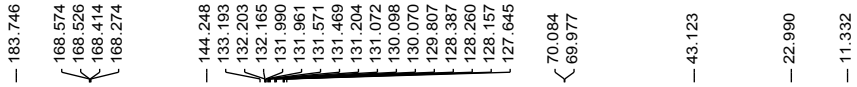
3b, ^{31}P NMR
162 MHz, $CDCl_3$

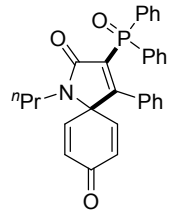




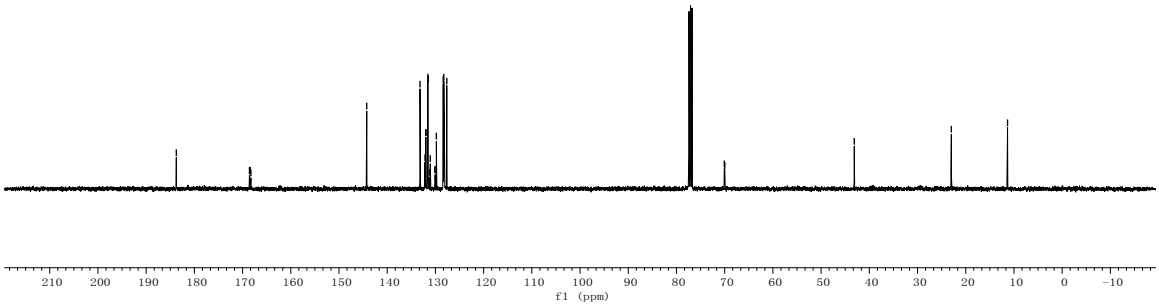
3c, ^{1}H NMR
400 MHz, $CDCl_3$

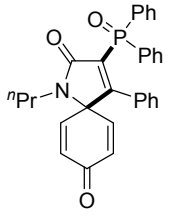




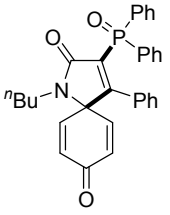


3c, ^{13}C NMR
101 MHz, $CDCl_3$

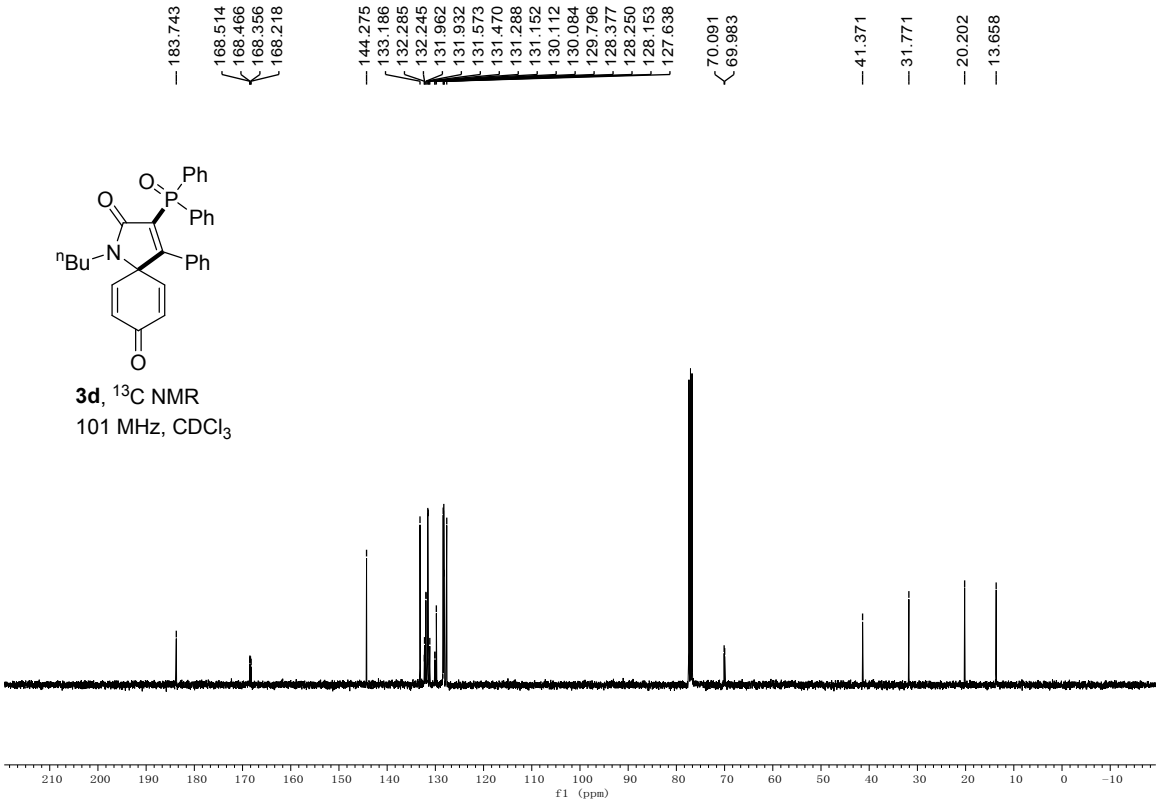




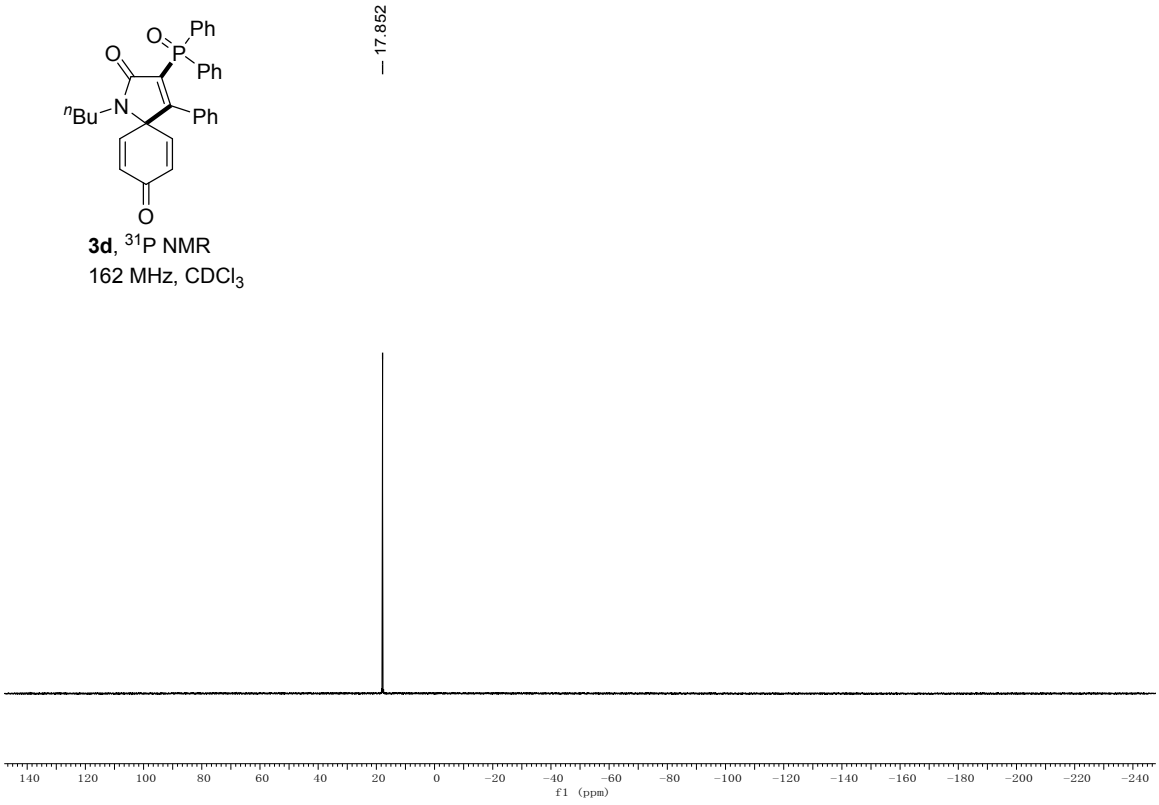
3c, ^{31}P NMR
162 MHz, $CDCl_3$



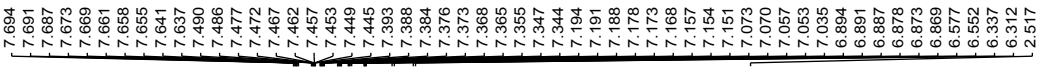
3d, ^{1}H NMR
400 MHz, $CDCl_3$

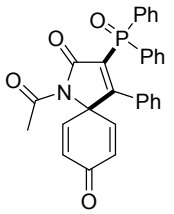


3d, ^{13}C NMR
101 MHz, $CDCl_3$

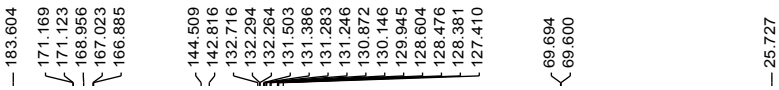


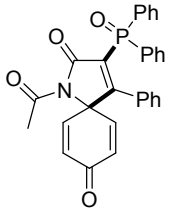
3d, ^{31}P NMR
162 MHz, $CDCl_3$



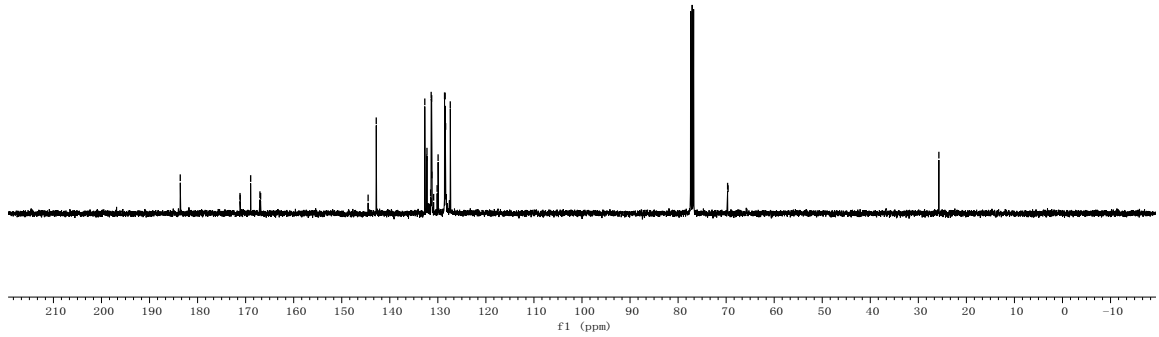


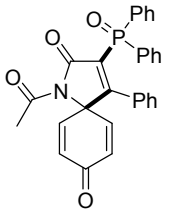
3e, ^{1}H NMR
400 MHz, $CDCl_3$



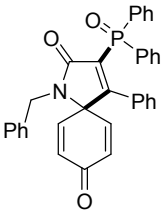


3e, ^{13}C NMR
101 MHz, $CDCl_3$

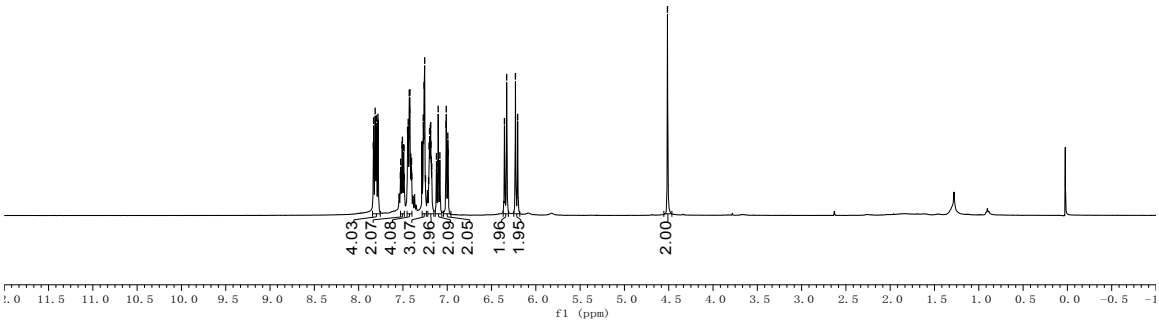


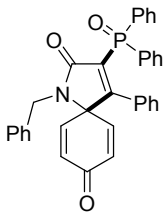


3e, ^{31}P NMR
162 MHz, $CDCl_3$

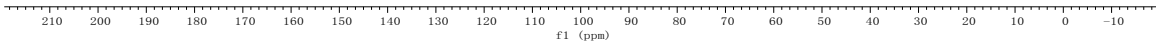


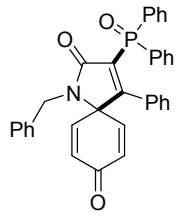
3f, ^{1}H NMR
400 MHz, $CDCl_3$



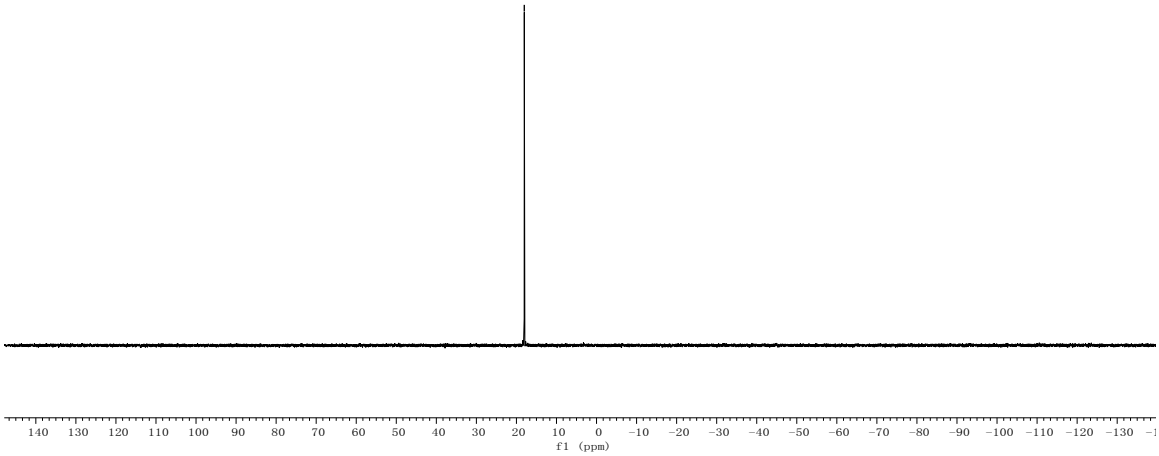


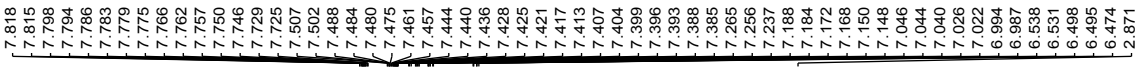
3f, ^{13}C NMR
101 MHz, $CDCl_3$

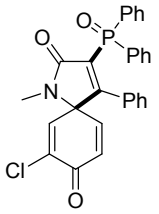




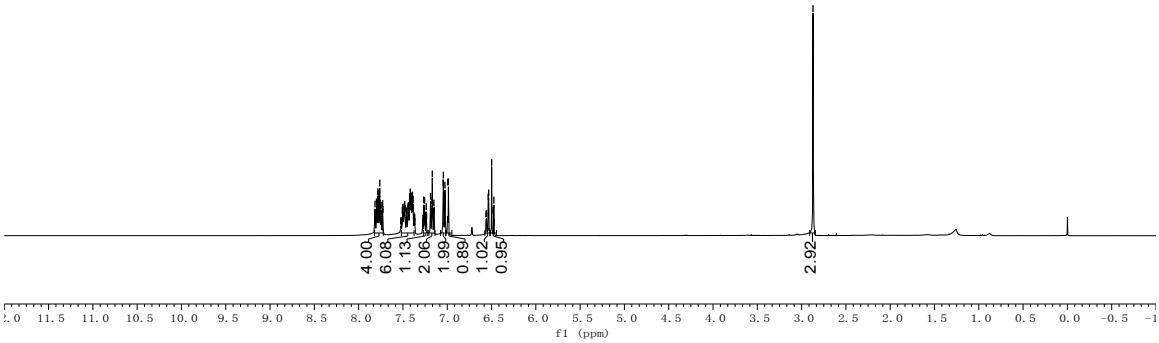
3f, ^{31}P NMR
162 MHz, $CDCl_3$

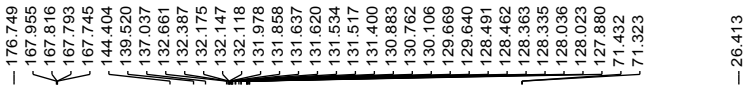


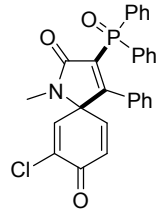




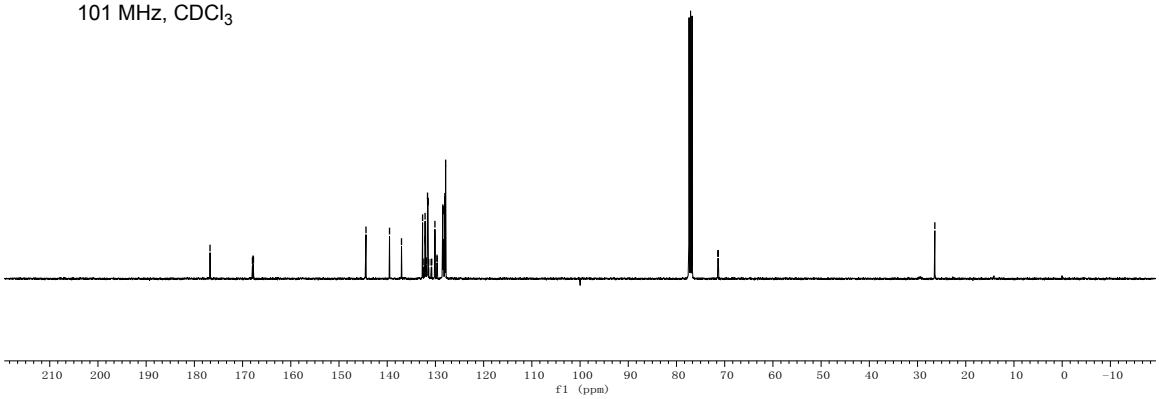
3g, ^{1}H NMR
400 MHz, $CDCl_3$

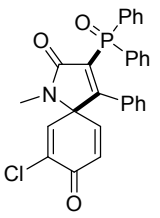




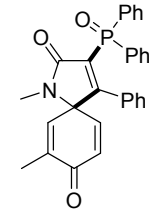


3g, ^{13}C NMR
101 MHz, $CDCl_3$

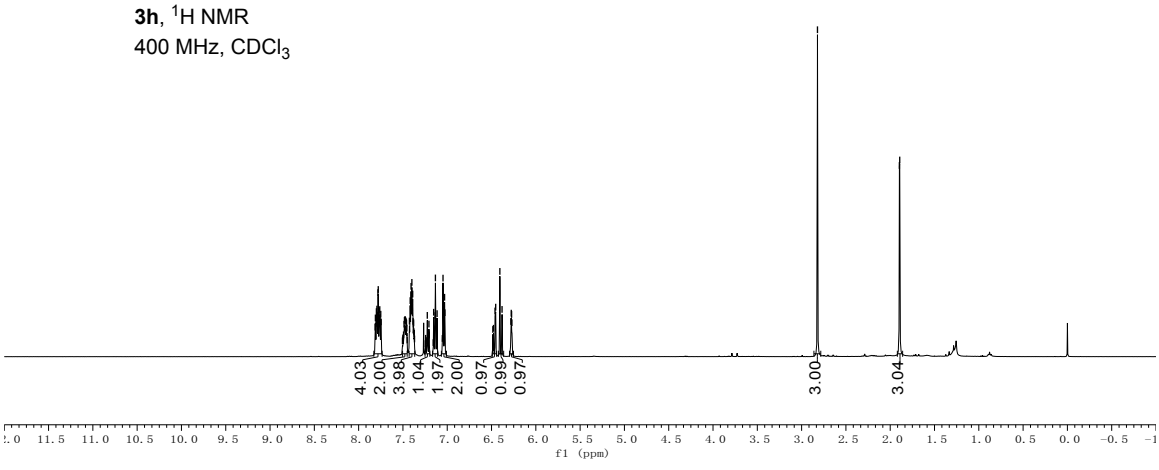


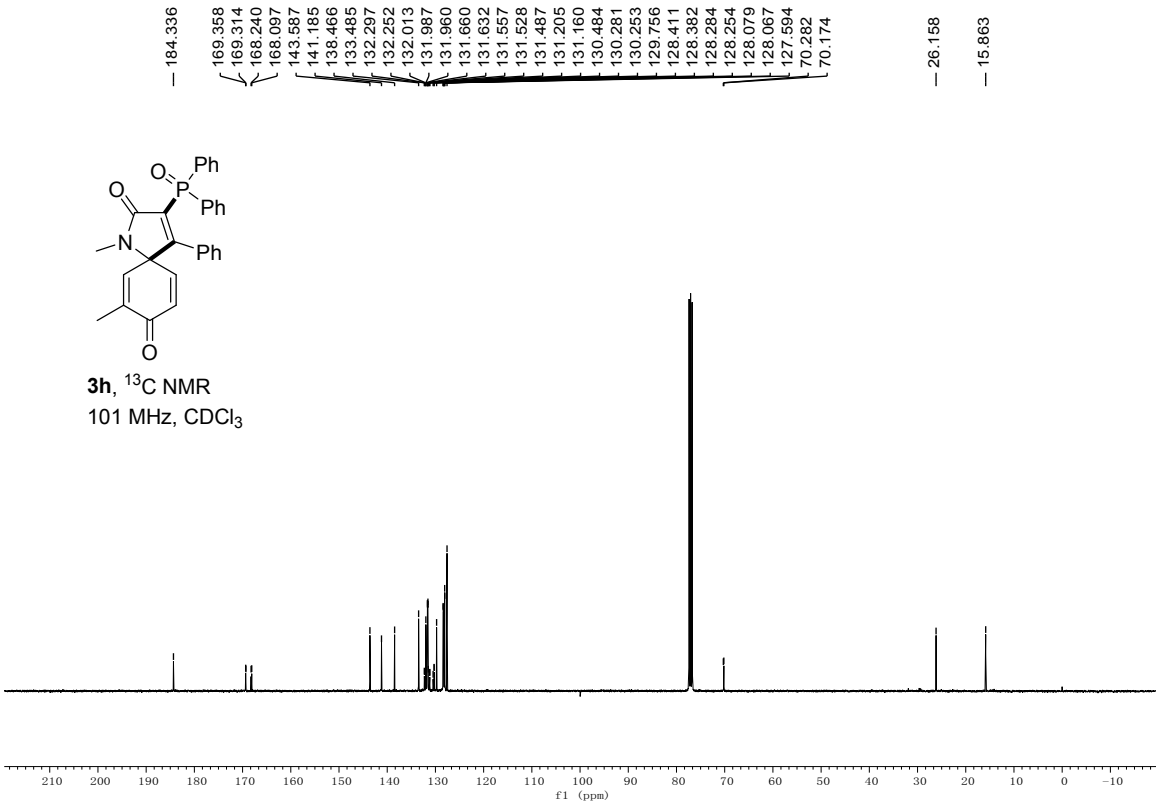


3g, ^{31}P NMR
162 MHz, $CDCl_3$

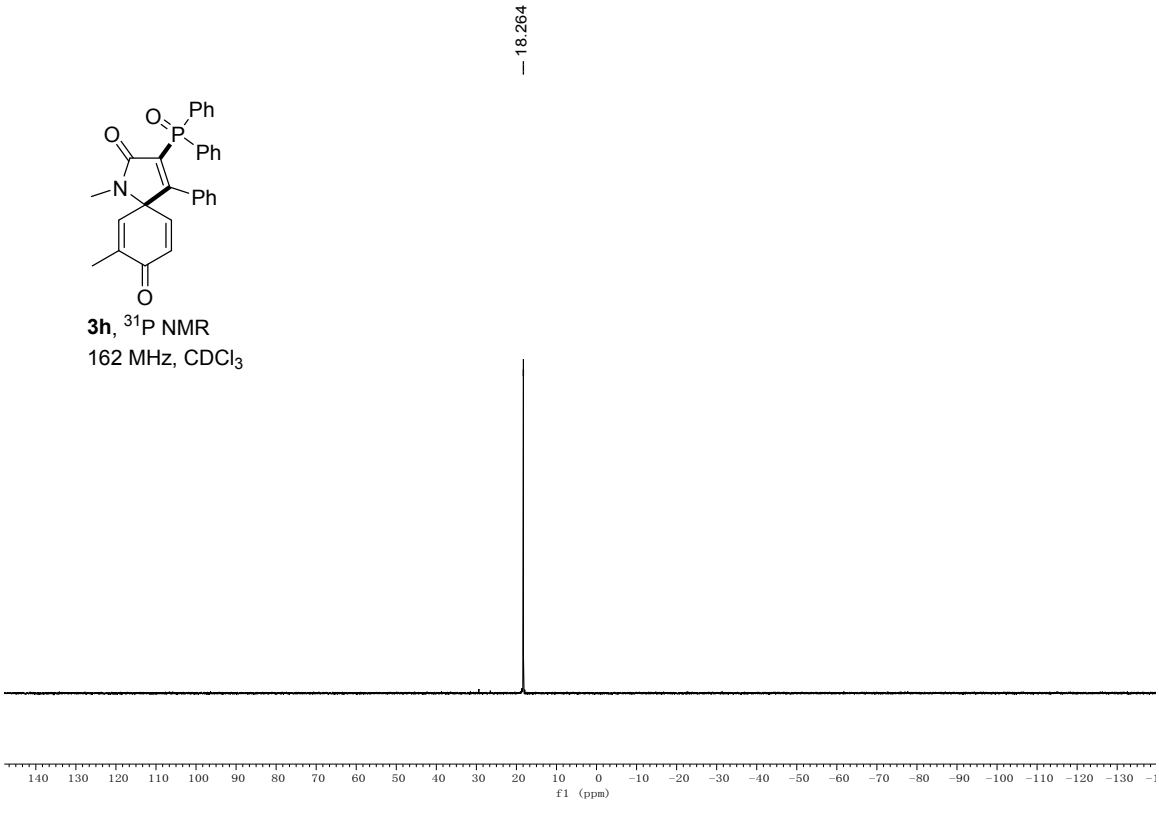


3h, ^{1}H NMR
400 MHz, $CDCl_3$

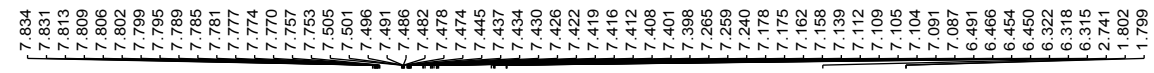


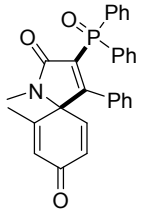


3h, ^{13}C NMR
101 MHz, $CDCl_3$

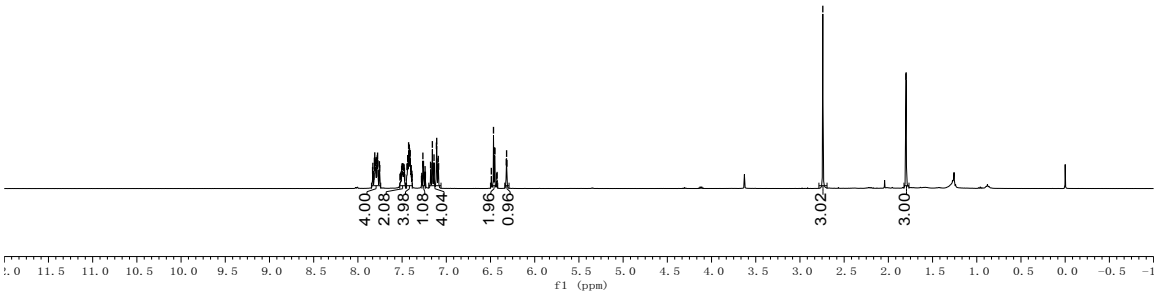


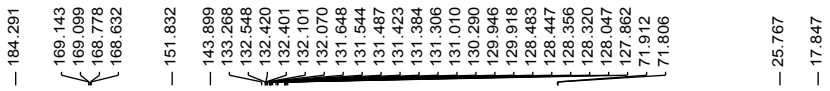
3h, ^{31}P NMR
162 MHz, $CDCl_3$

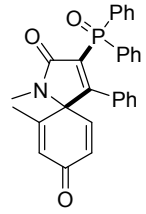




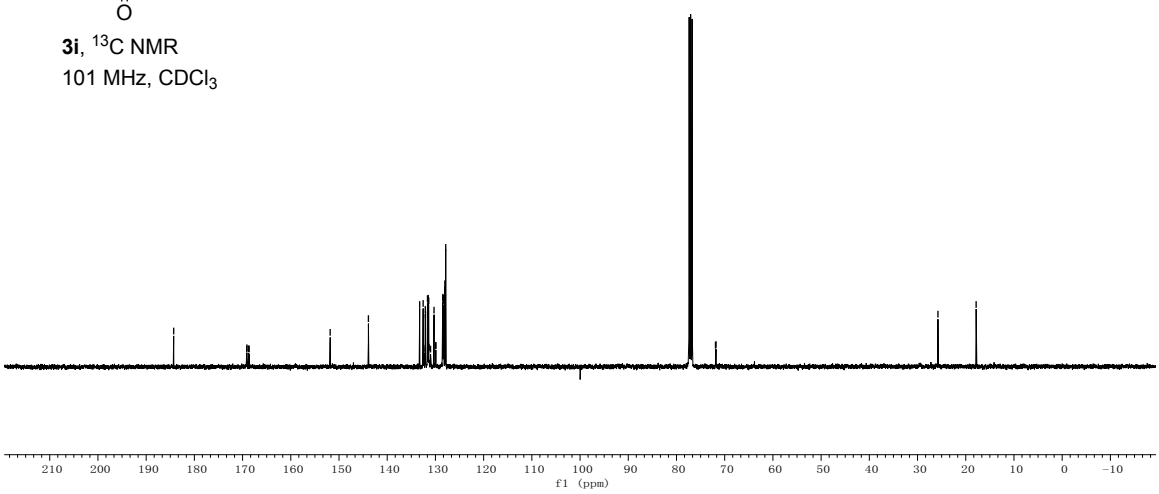
3i, ^{1}H NMR
400 MHz, $CDCl_3$

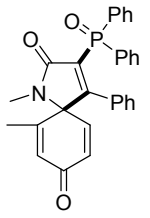




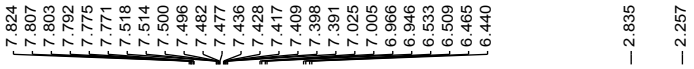


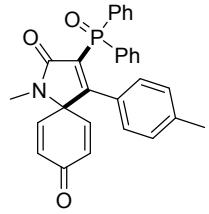
3i, ^{13}C NMR
101 MHz, $CDCl_3$



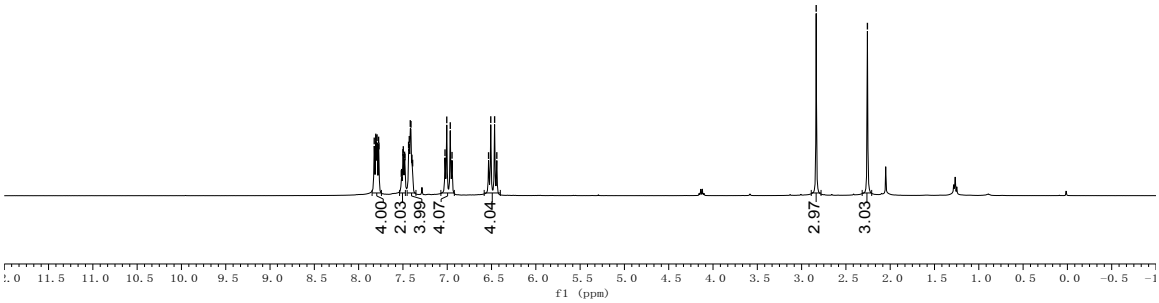


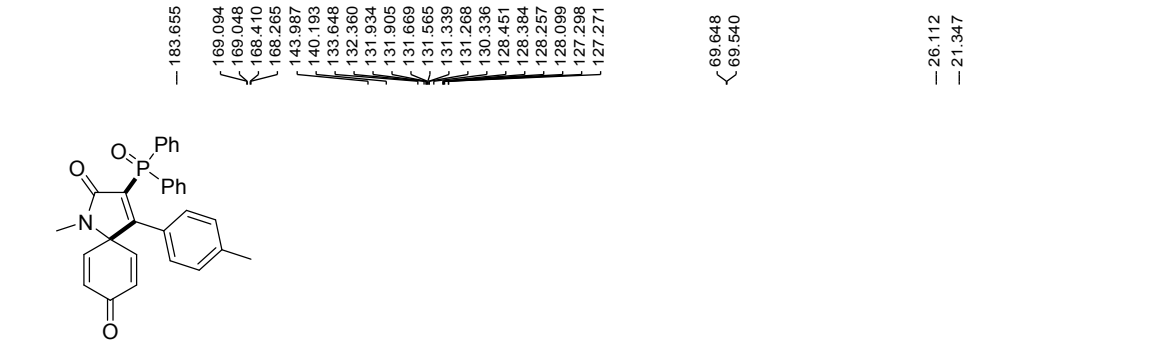
3i, ^{31}P NMR
162 MHz, $CDCl_3$



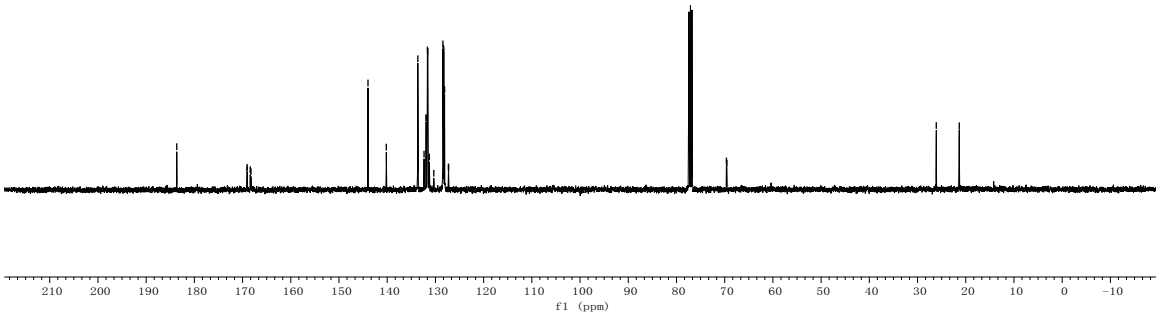


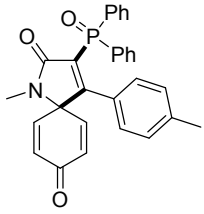
3j, ^{1}H NMR
400 MHz, $CDCl_3$



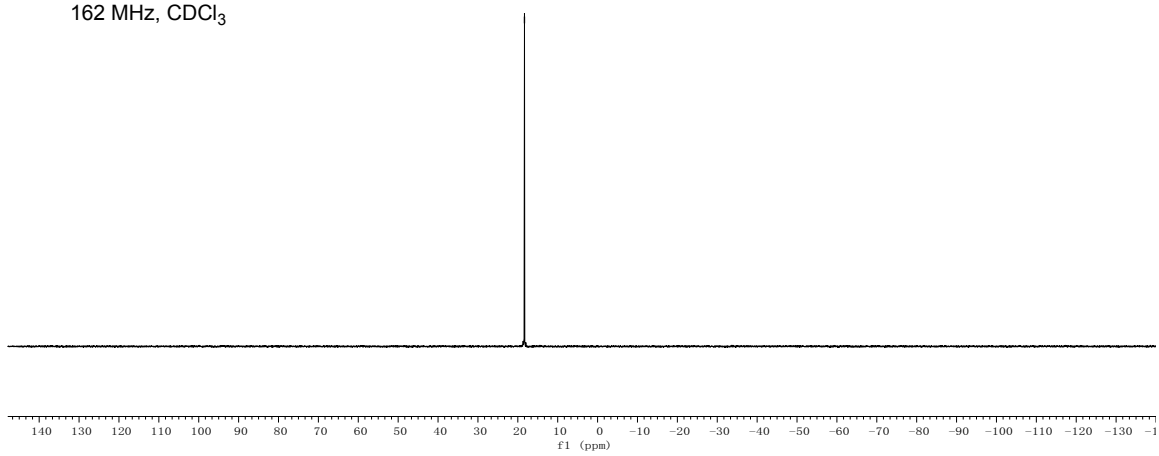


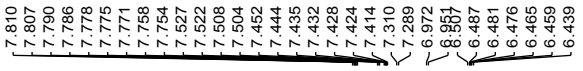
3j, ^{13}C NMR
101 MHz, $CDCl_3$

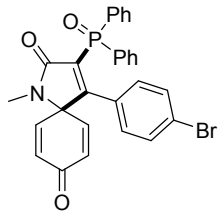




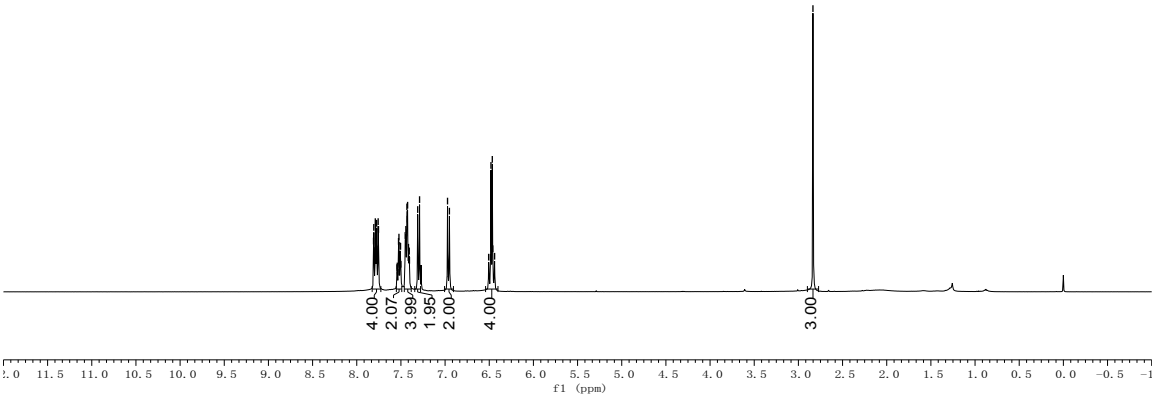
3j, ^{31}P NMR
162 MHz, $CDCl_3$

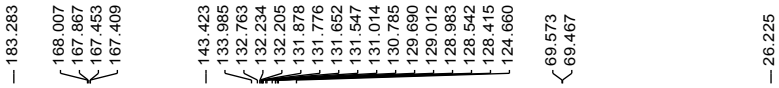


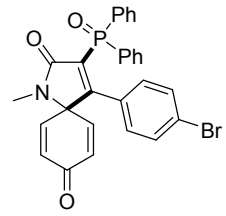




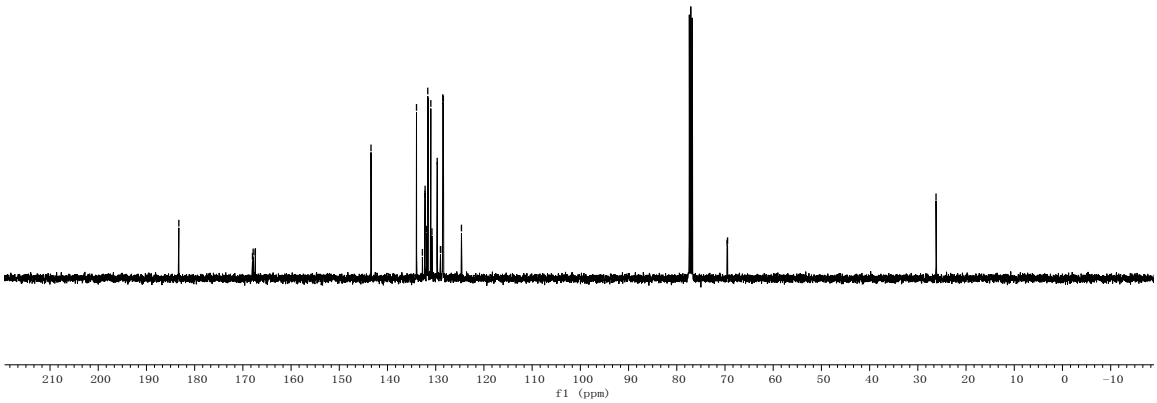
3k, ^{1}H NMR
400 MHz, $CDCl_3$

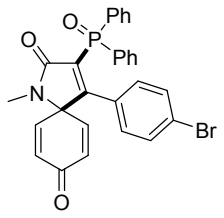




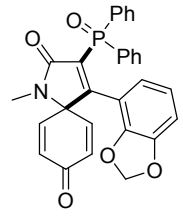


3k, ^{13}C NMR
101 MHz, $CDCl_3$

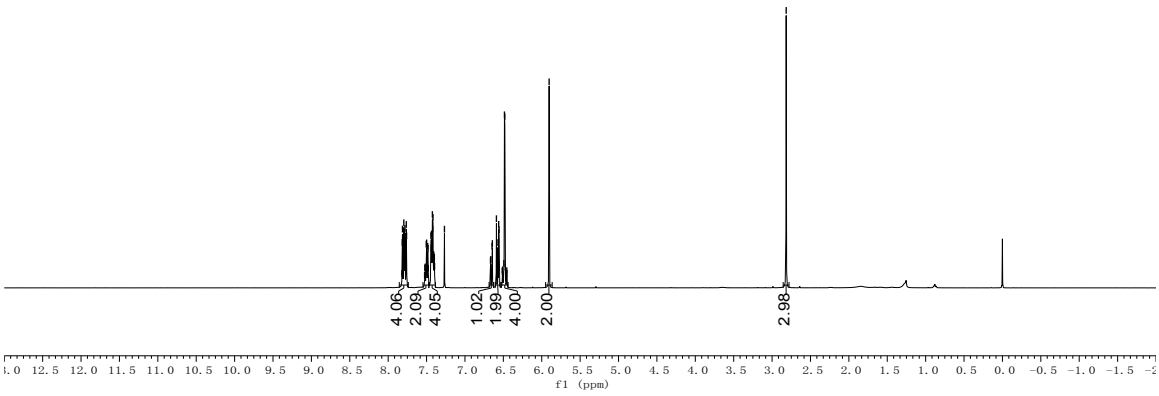


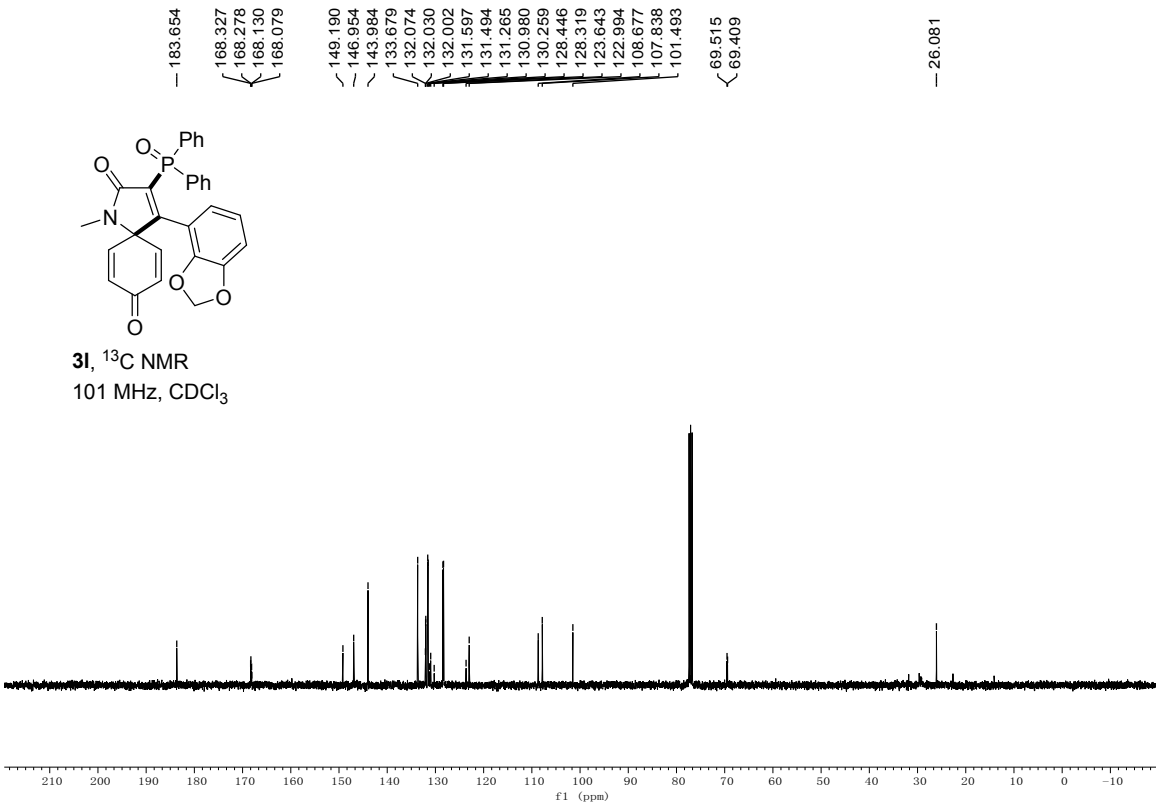


3k, ^{31}P NMR
162 MHz, $CDCl_3$

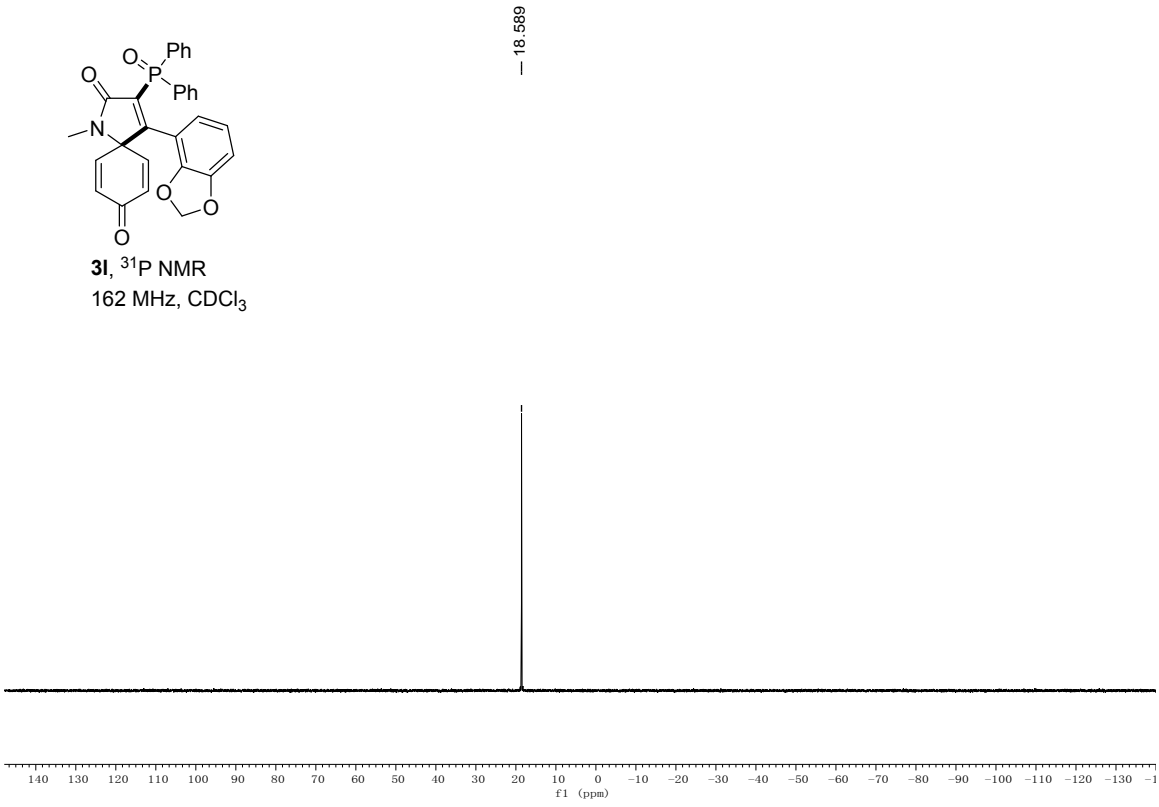


3l, ^{1}H NMR
400 MHz, $CDCl_3$

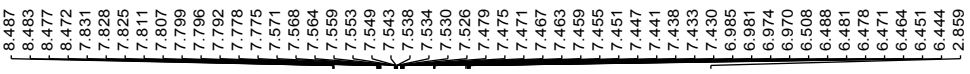


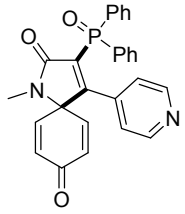


3l, ^{13}C NMR
101 MHz, $CDCl_3$

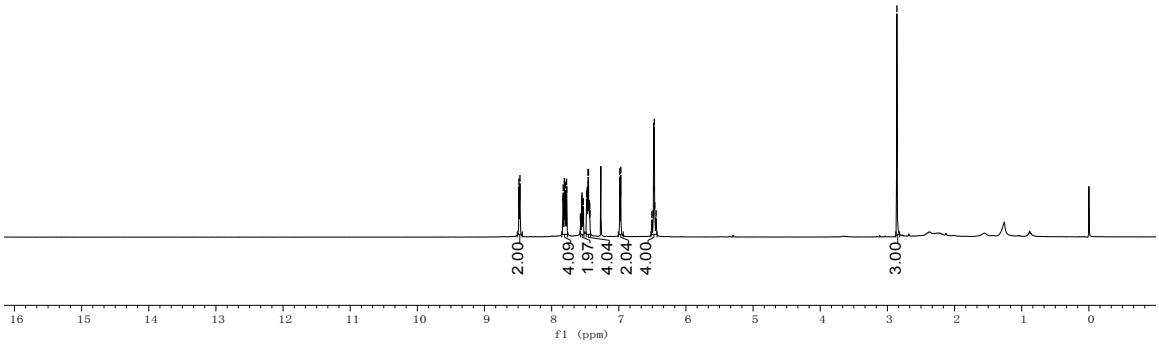


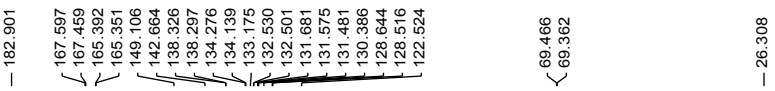
3l, ^{31}P NMR
162 MHz, $CDCl_3$

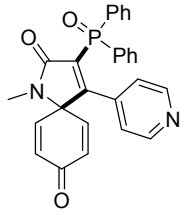




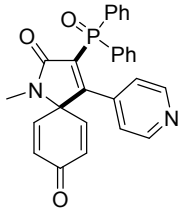
3m, ^{1}H NMR
400 MHz, $CDCl_3$



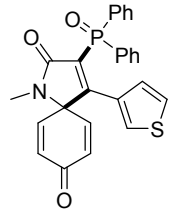




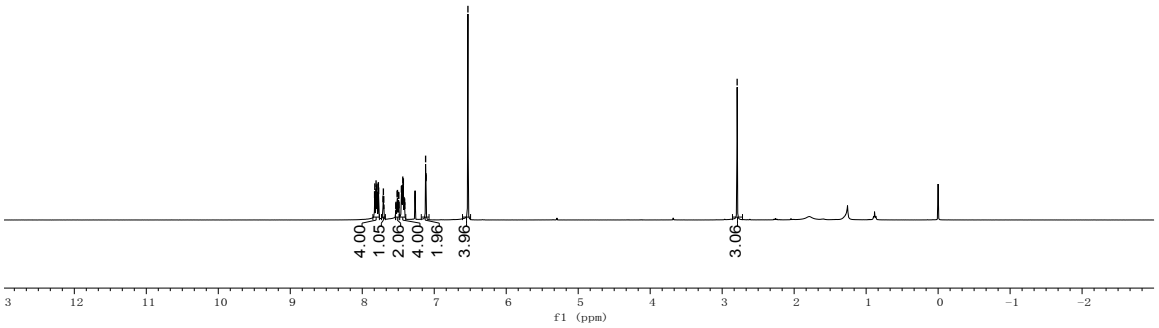
3m, ^{13}C NMR
101 MHz, $CDCl_3$

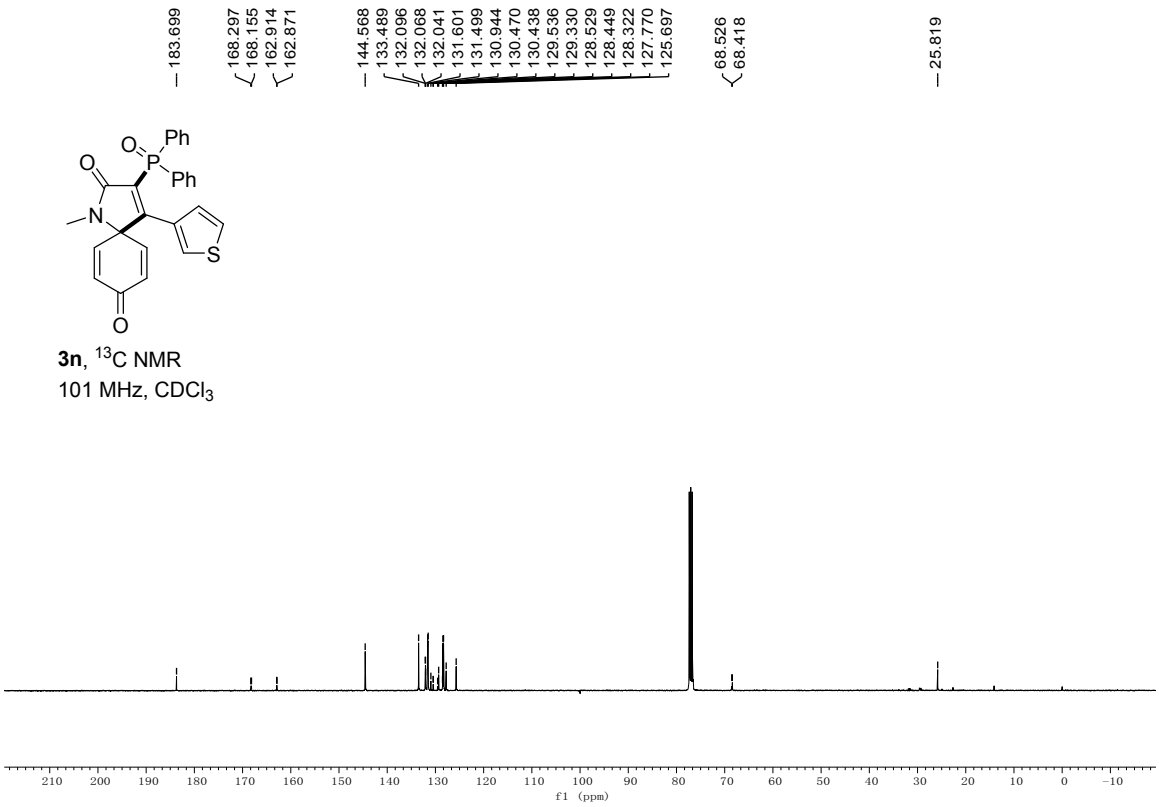


3m, ^{31}P NMR
162 MHz, $CDCl_3$

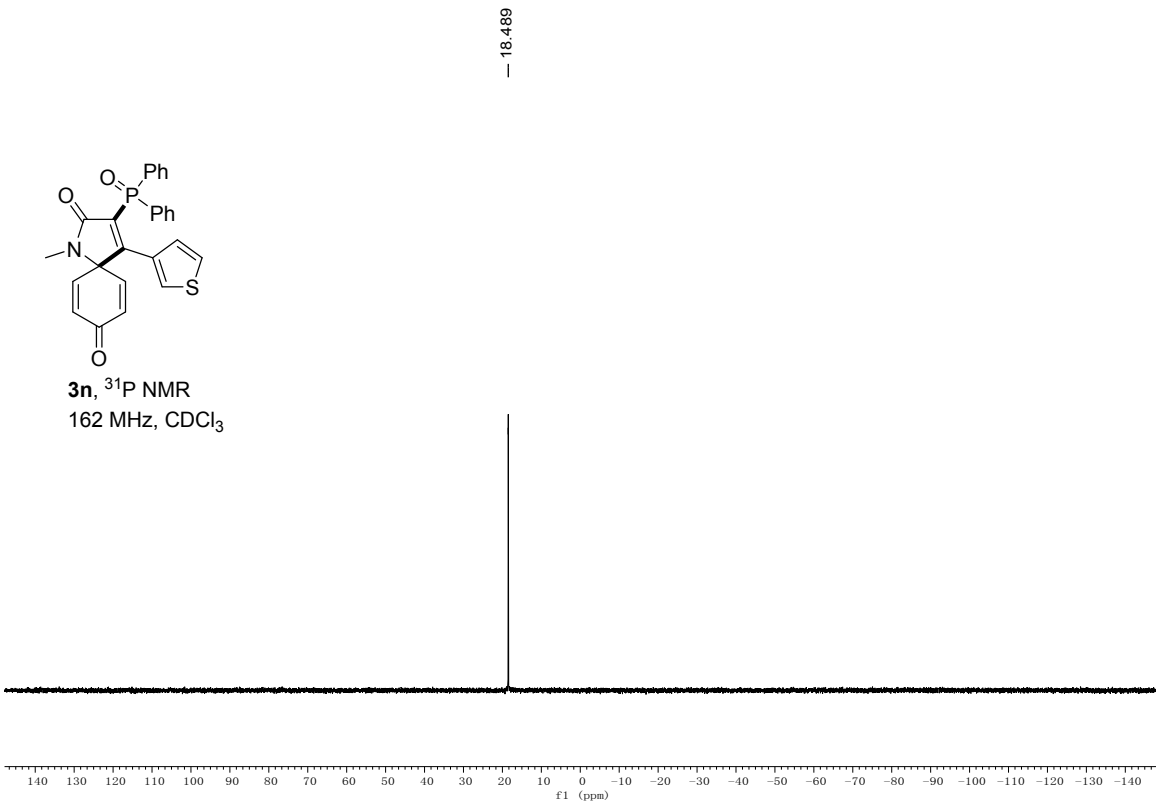


3n, ^{1}H NMR
400 MHz, $CDCl_3$

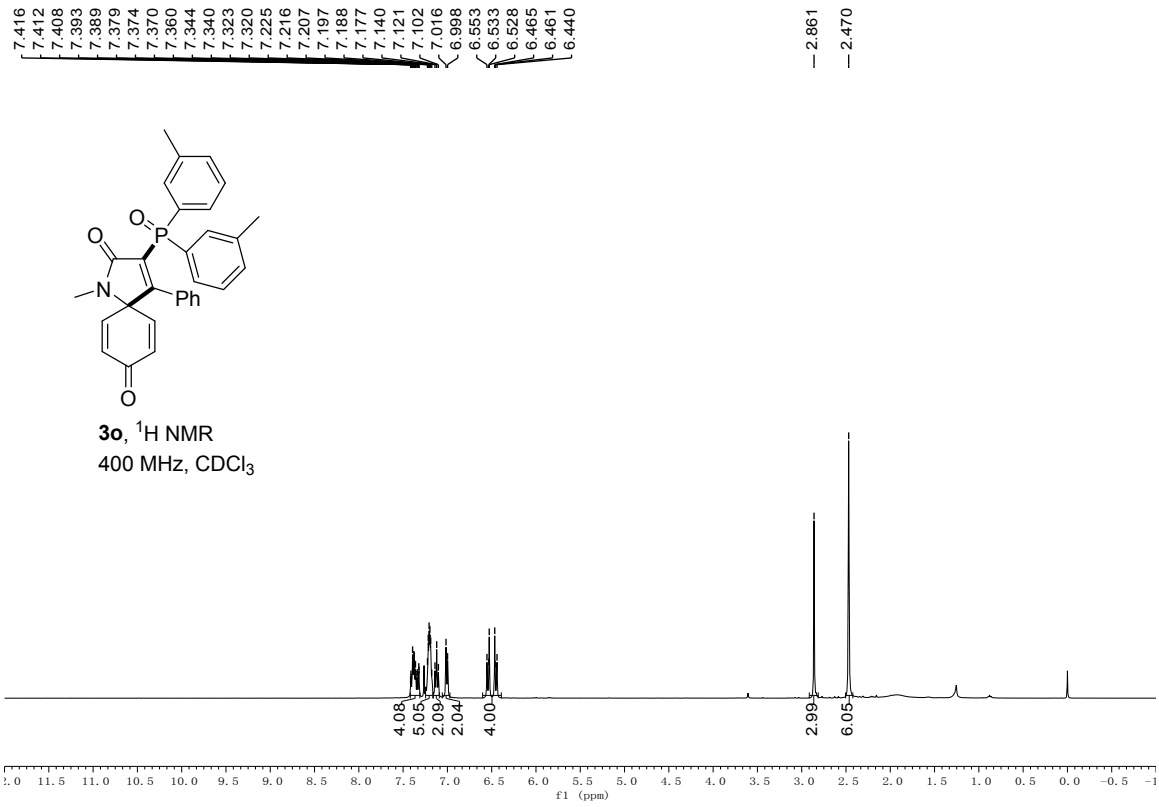




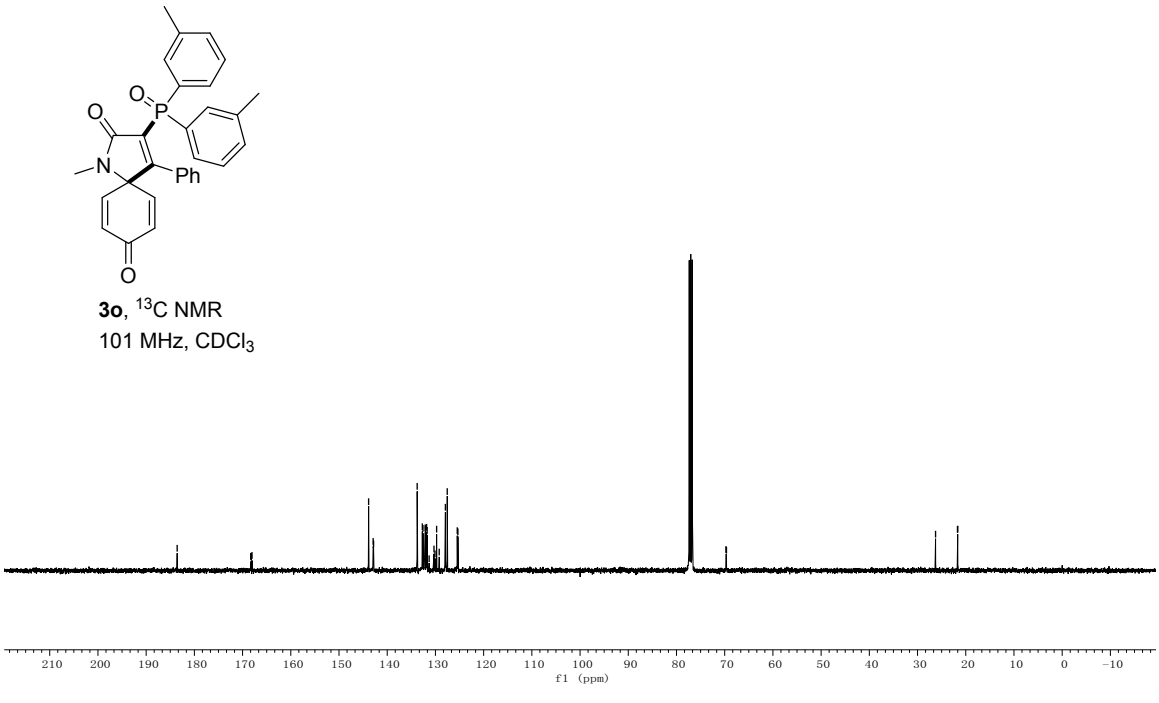
3n, ^{13}C NMR
101 MHz, $CDCl_3$



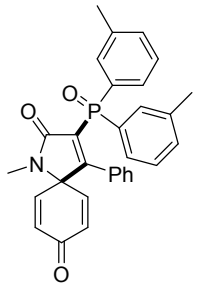
3n, ^{31}P NMR
162 MHz, $CDCl_3$



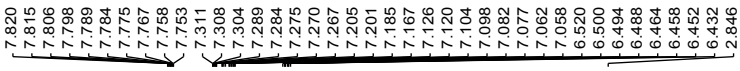
3o, ^{1}H NMR
400 MHz, $CDCl_3$

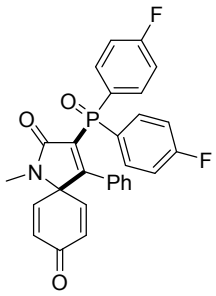


3o, ^{13}C NMR
101 MHz, $CDCl_3$

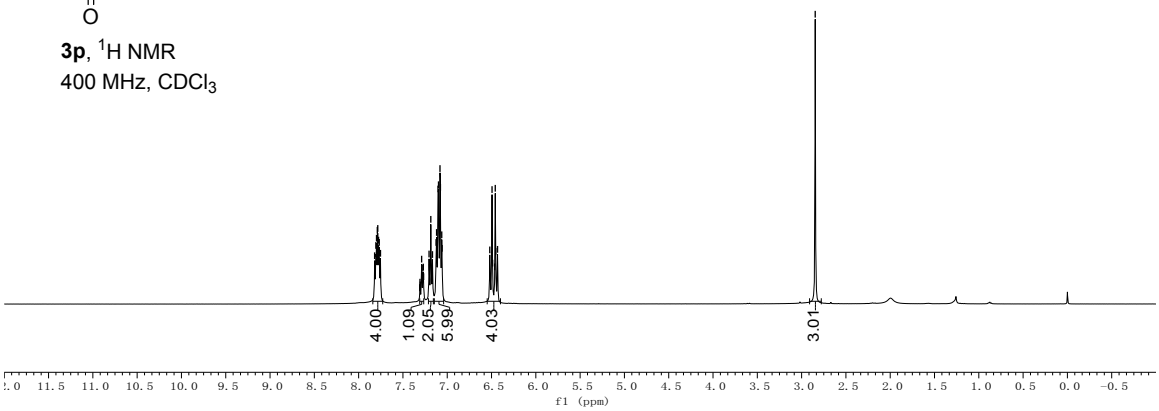


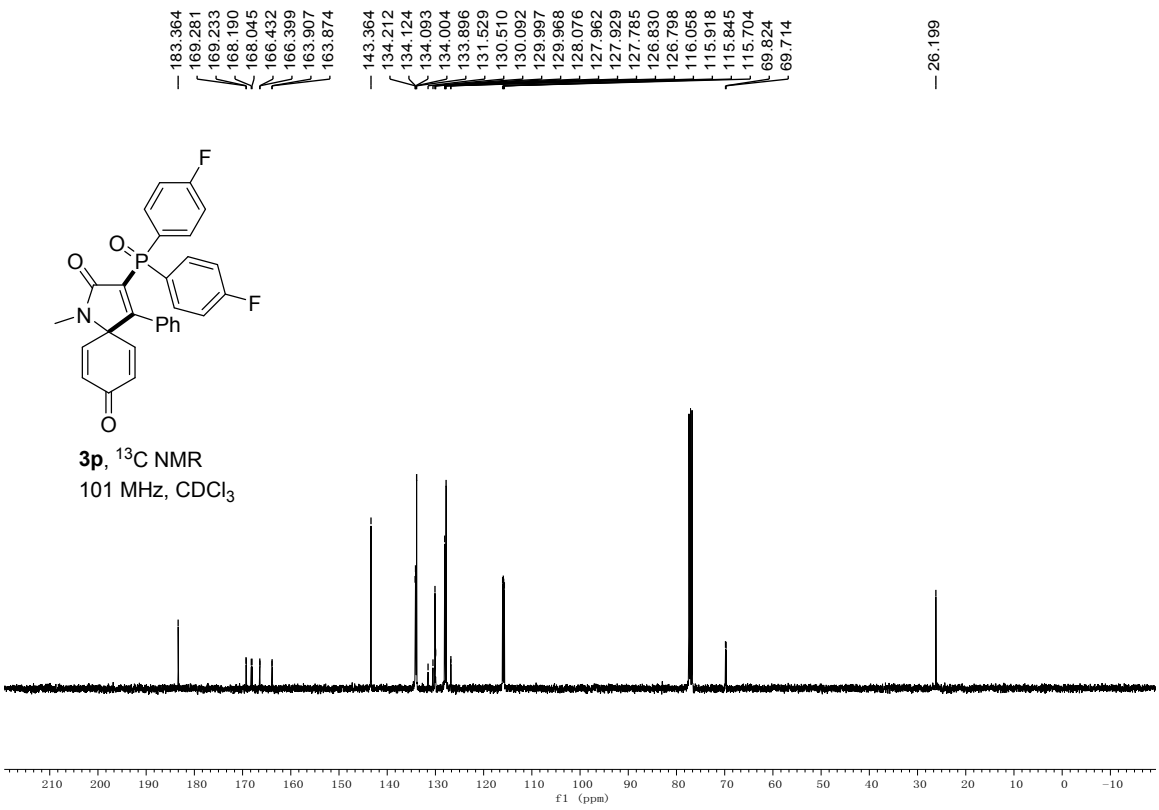
3o, ^{31}P NMR
162 MHz, $CDCl_3$



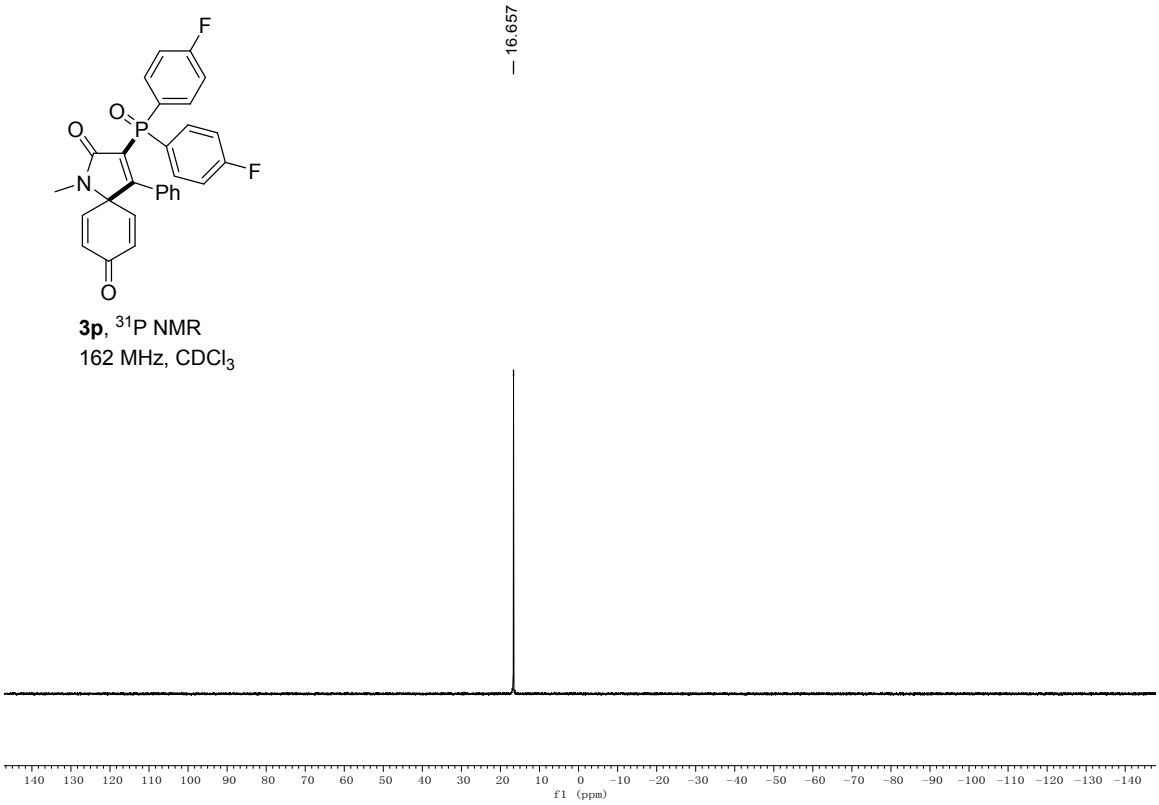


3p, ^{1}H NMR
400 MHz, $CDCl_3$

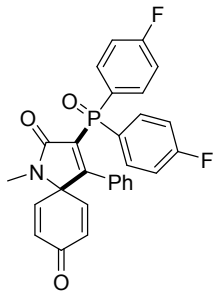




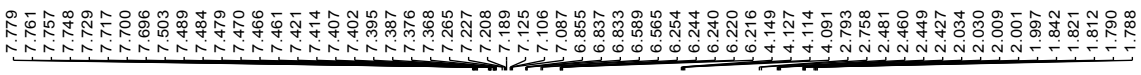
3p, ^{13}C NMR
101 MHz, $CDCl_3$

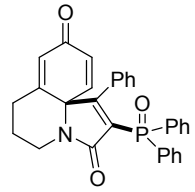


3p, ^{31}P NMR
162 MHz, $CDCl_3$

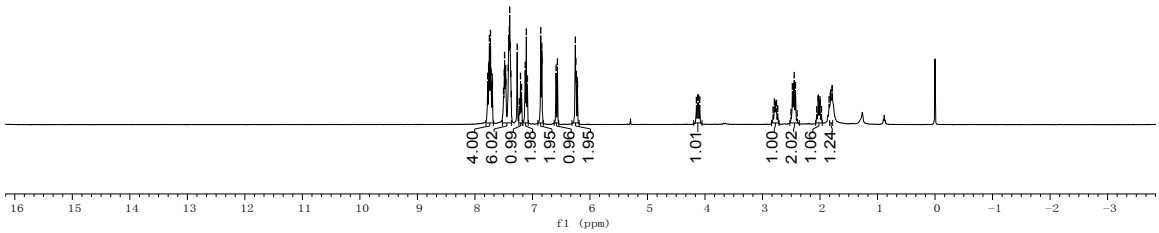


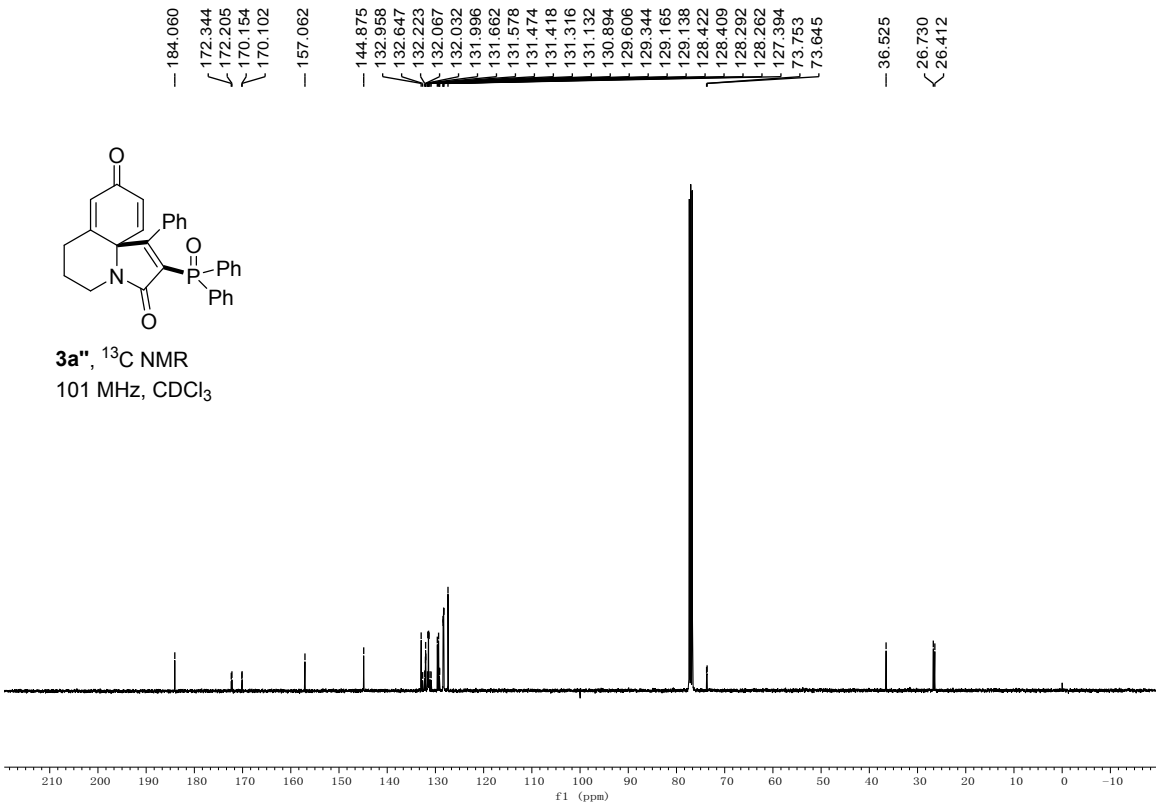
3q, ^{19}F NMR
376 MHz, $CDCl_3$



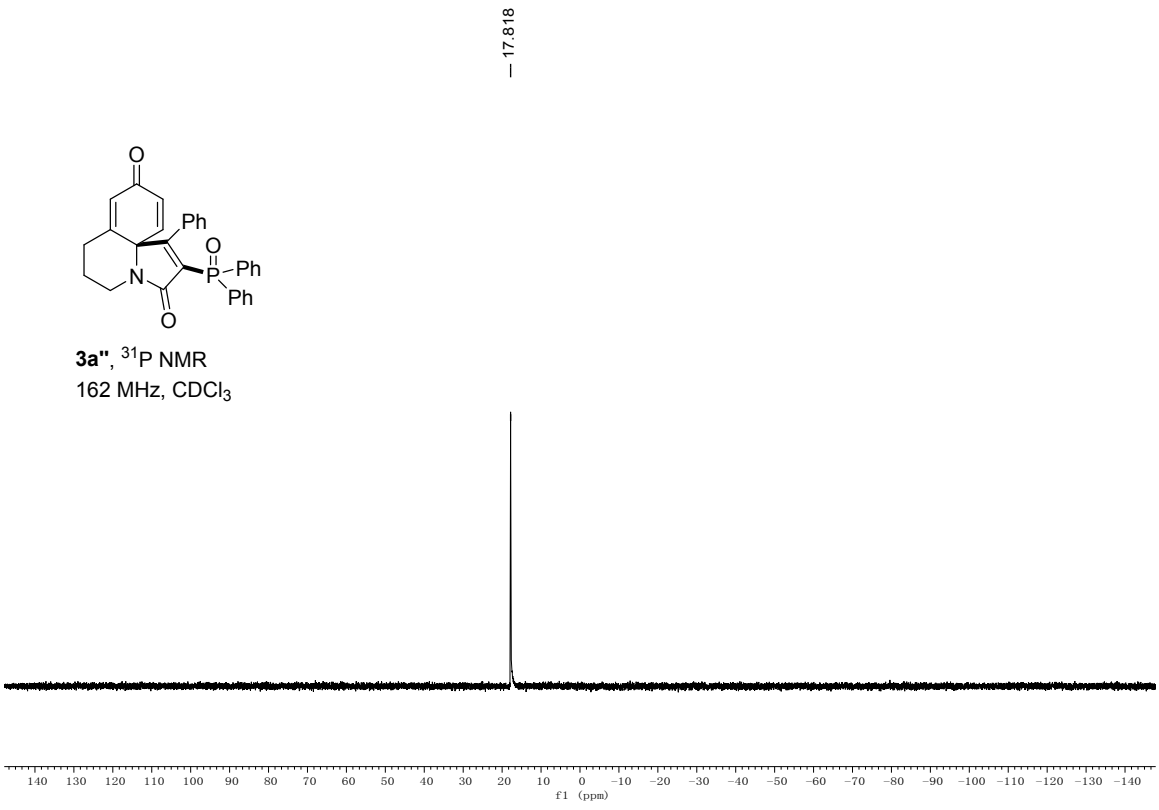


3a'', ^{1}H NMR
400 MHz, $CDCl_3$

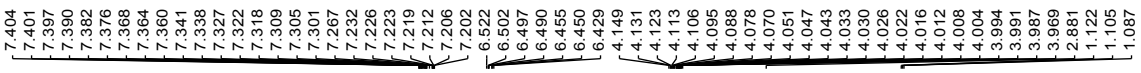


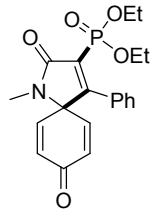


3a", ^{13}C NMR
101 MHz, $CDCl_3$

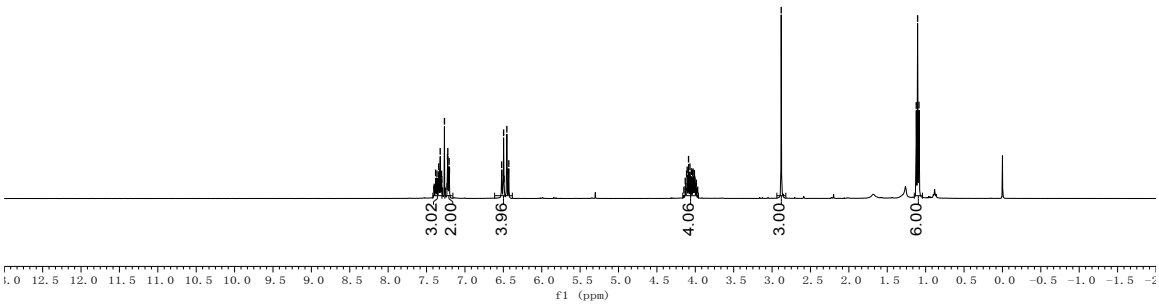


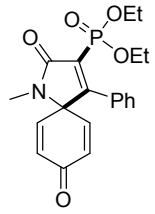
3a", ^{31}P NMR
162 MHz, $CDCl_3$



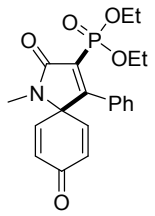


3q, ^{1}H NMR
400 MHz, $CDCl_3$

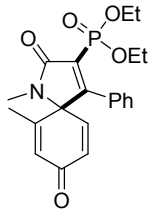




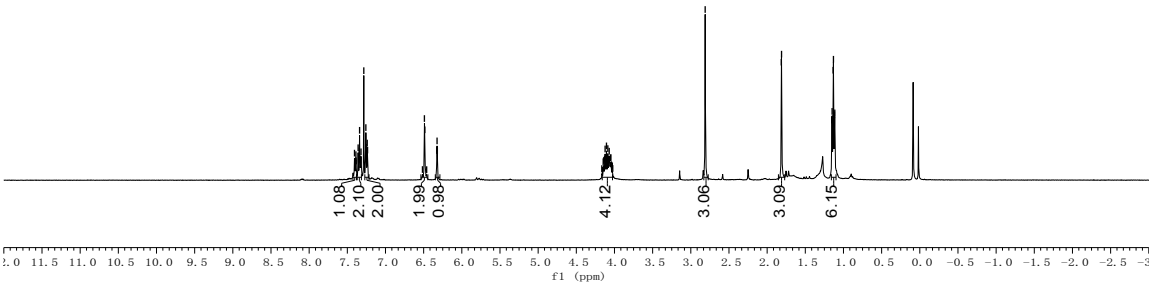
3q, ^{13}C NMR
151 MHz, $CDCl_3$

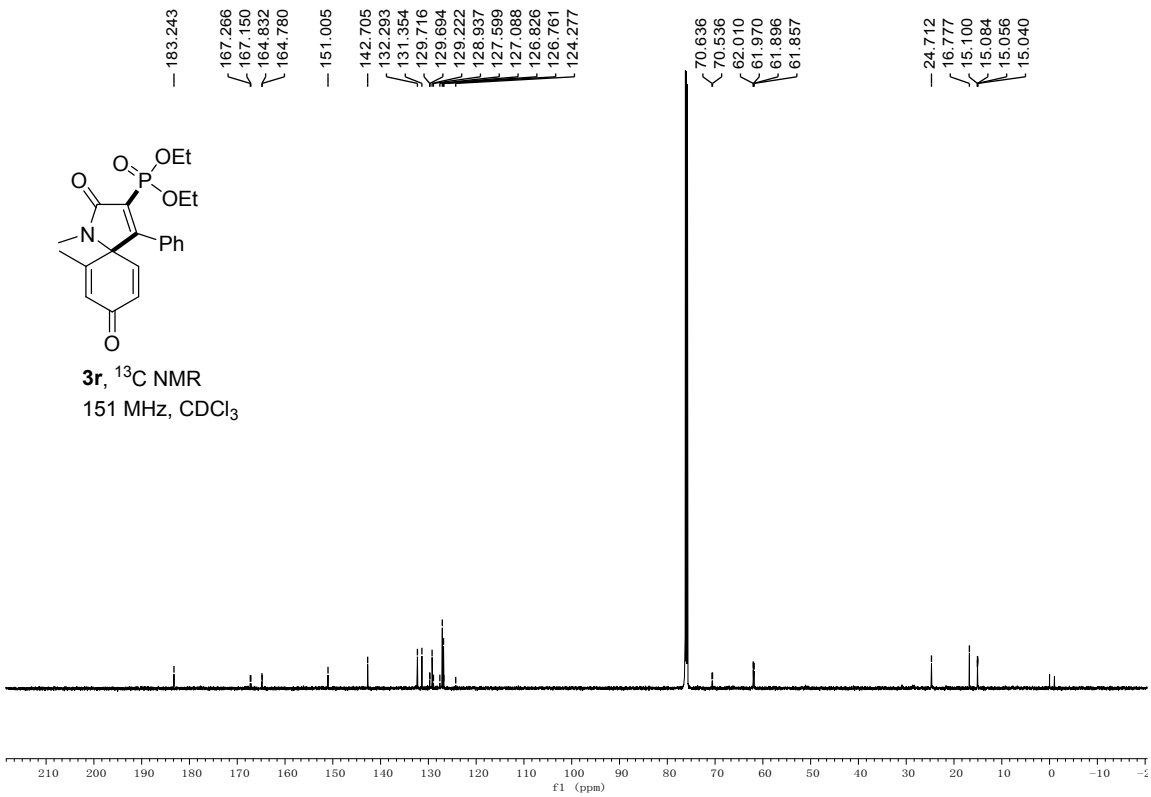


3q, ^{31}P NMR
162 MHz, $CDCl_3$

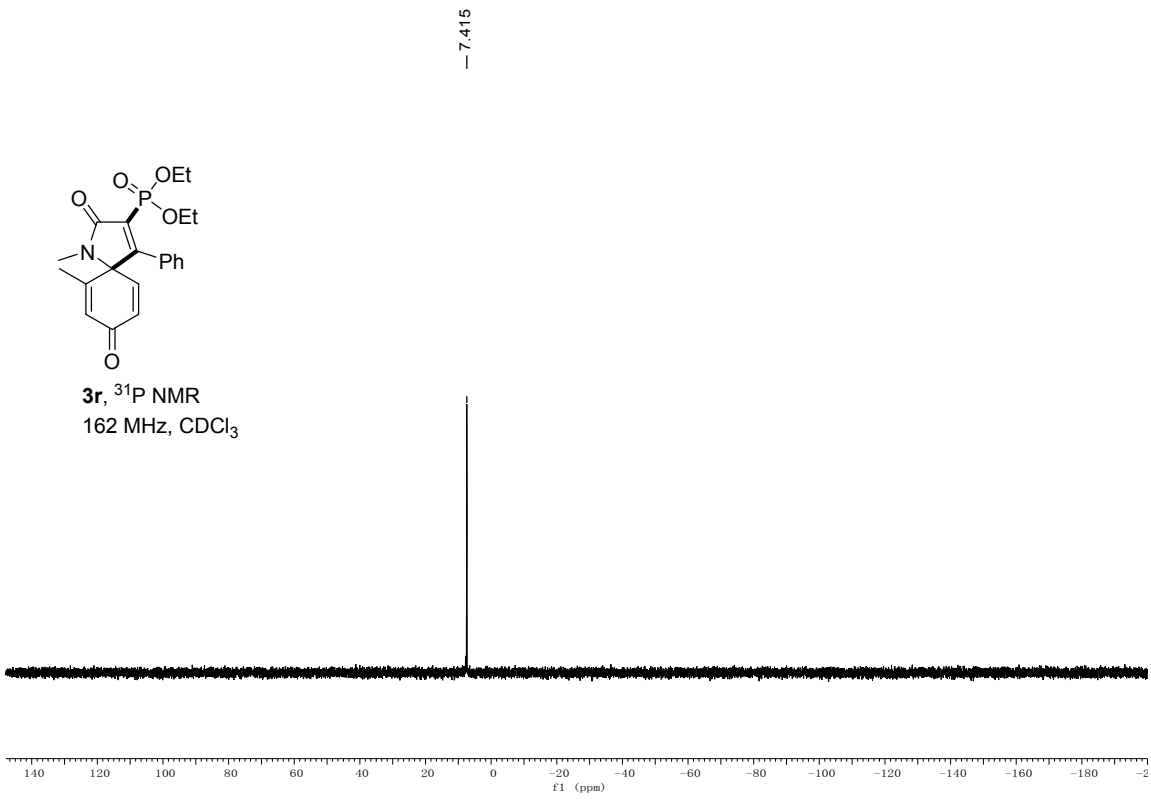


3r, ^{1}H NMR
400 MHz, $CDCl_3$

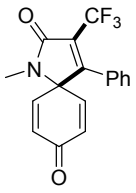




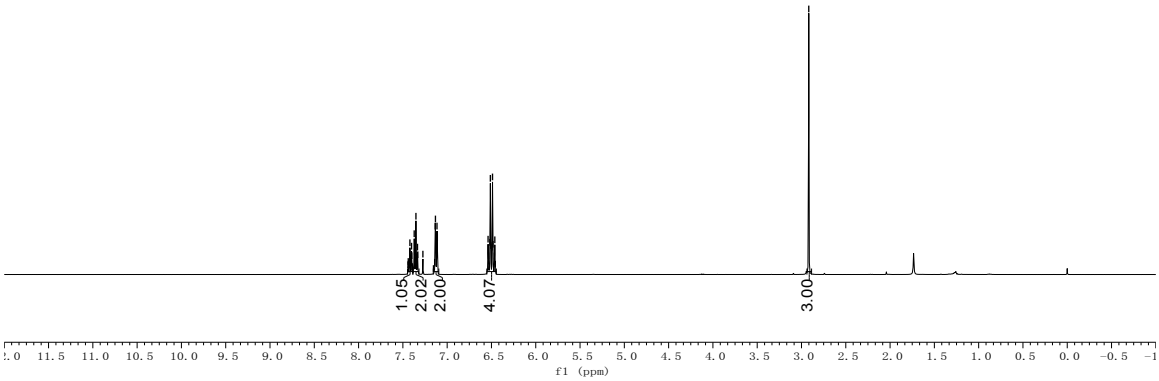
3r, ^{13}C NMR
151 MHz, $CDCl_3$

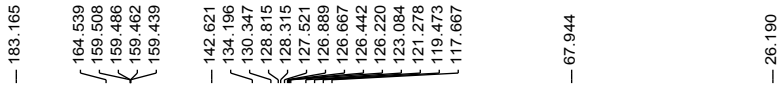


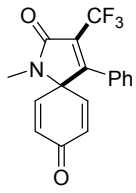
3r, ^{31}P NMR
162 MHz, $CDCl_3$



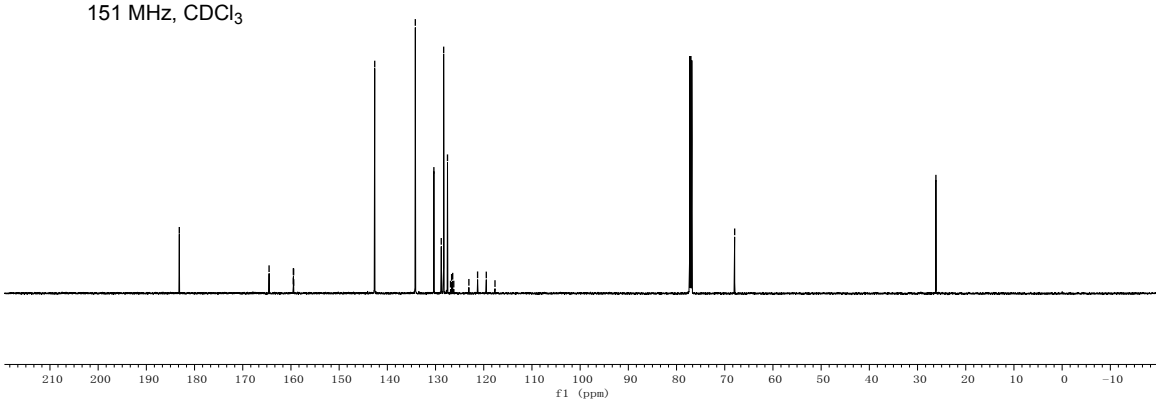
4a, ^{1}H NMR
400 MHz, $CDCl_3$

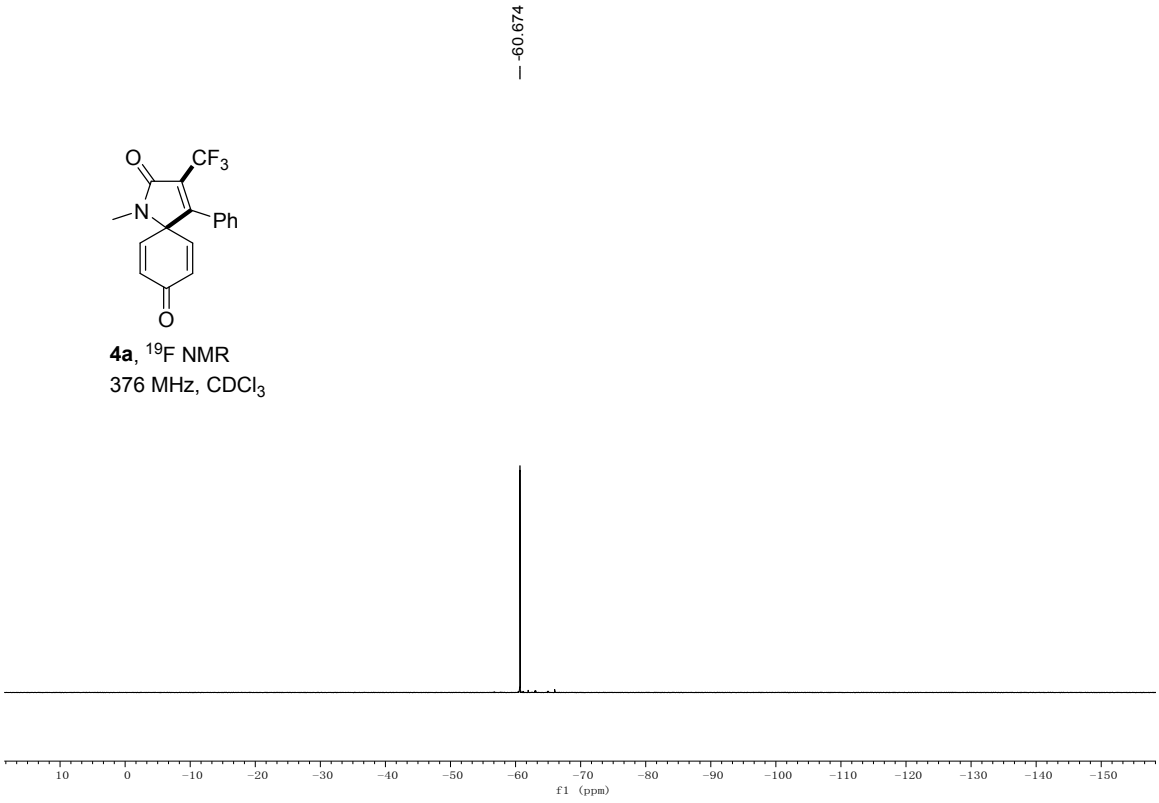




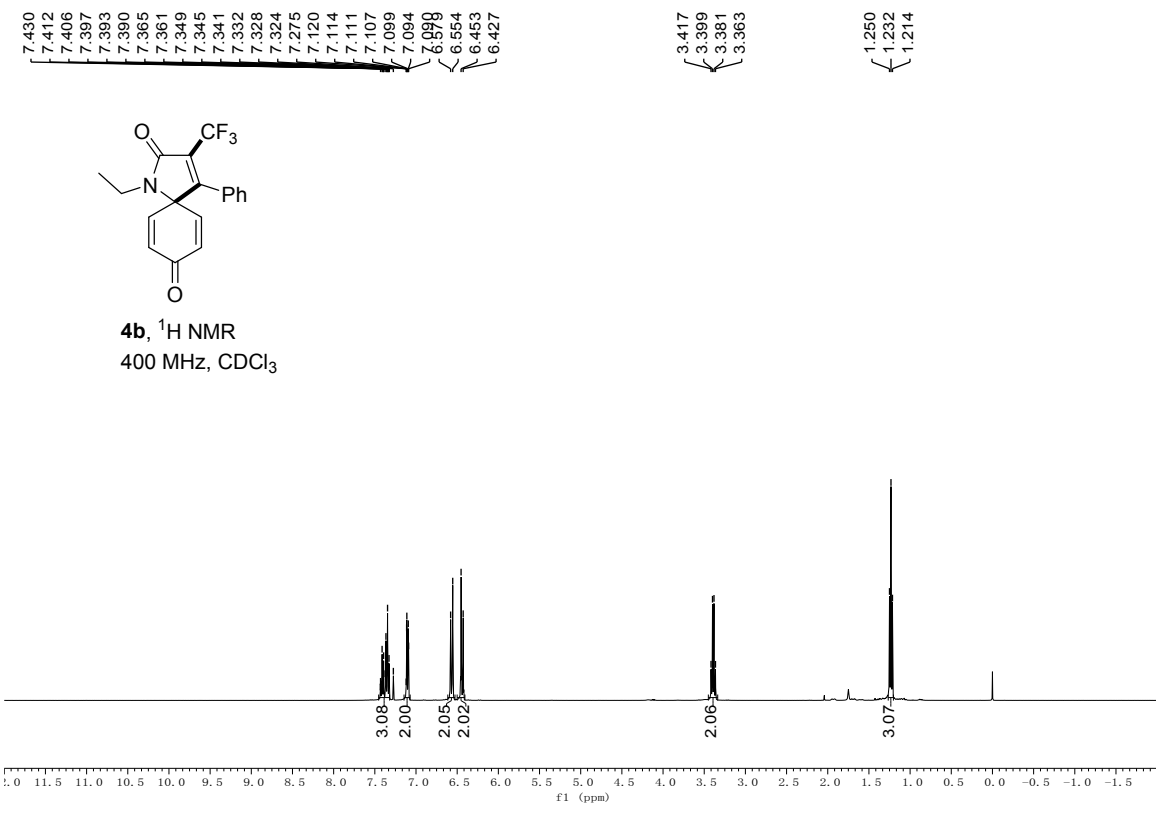


4a, ^{13}C NMR
151 MHz, $CDCl_3$

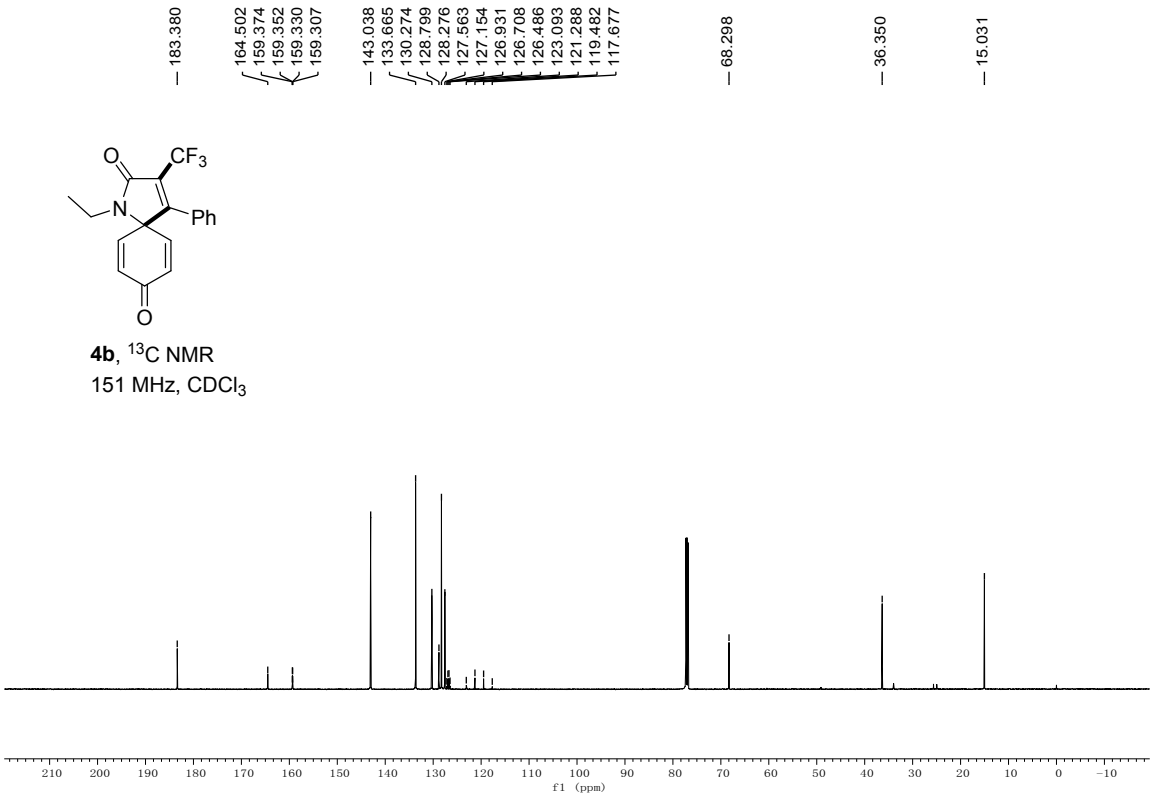




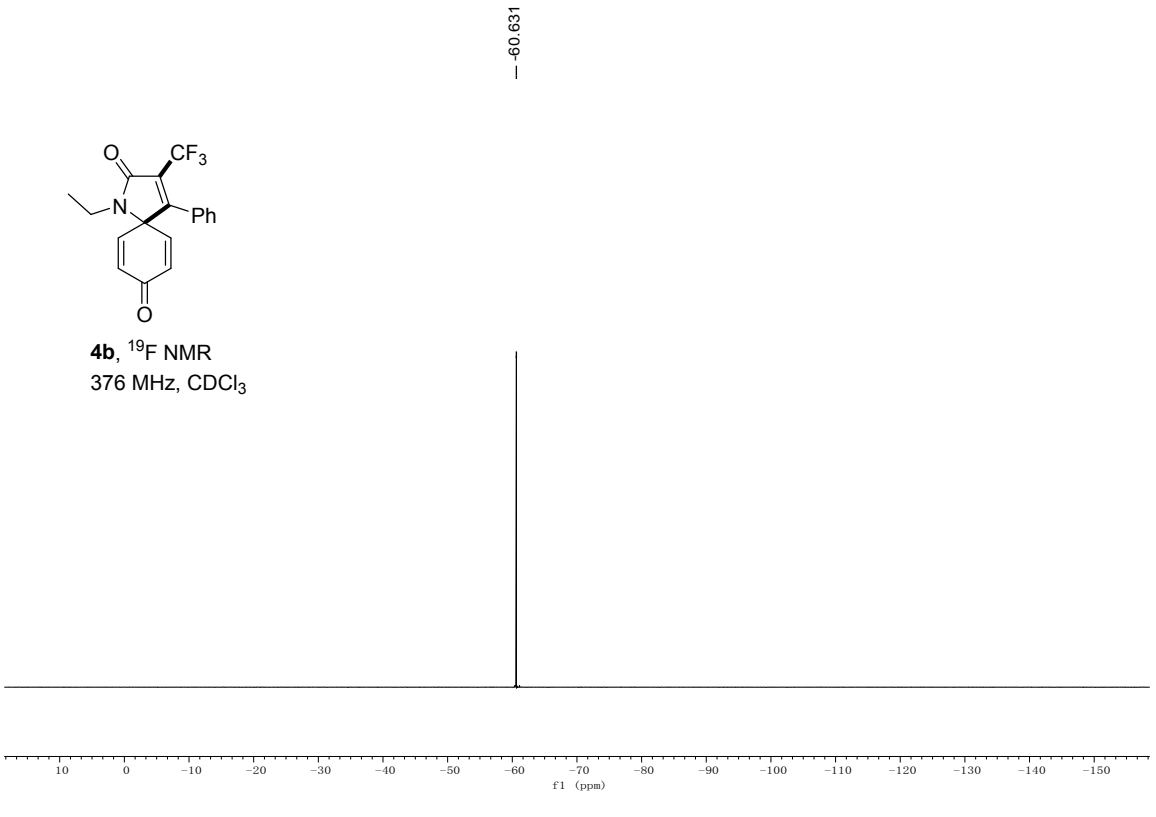
4a, ^{19}F NMR
376 MHz, $CDCl_3$



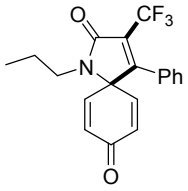
4b, ^{1}H NMR
400 MHz, $CDCl_3$



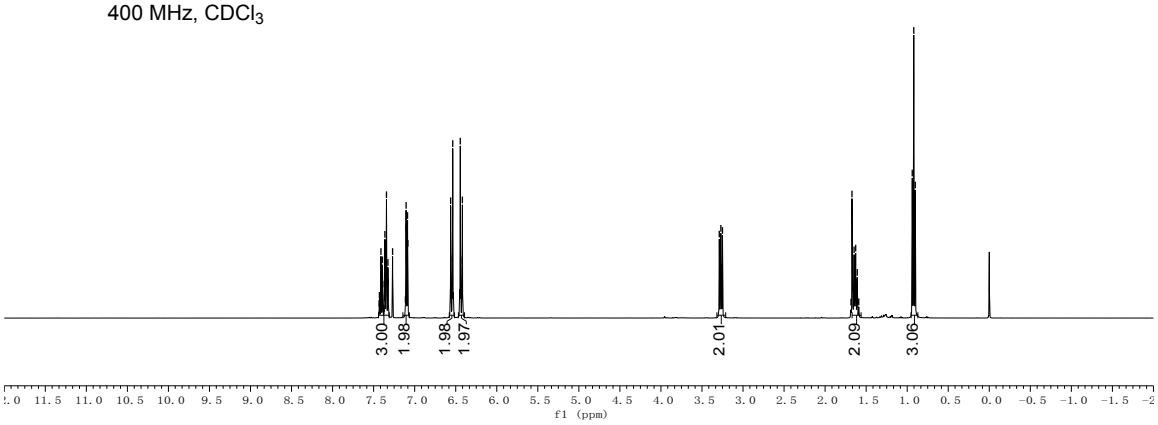
4b, ^{13}C NMR
151 MHz, $CDCl_3$

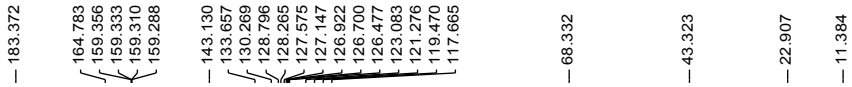


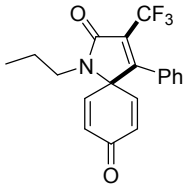
4b, ^{19}F NMR
376 MHz, $CDCl_3$



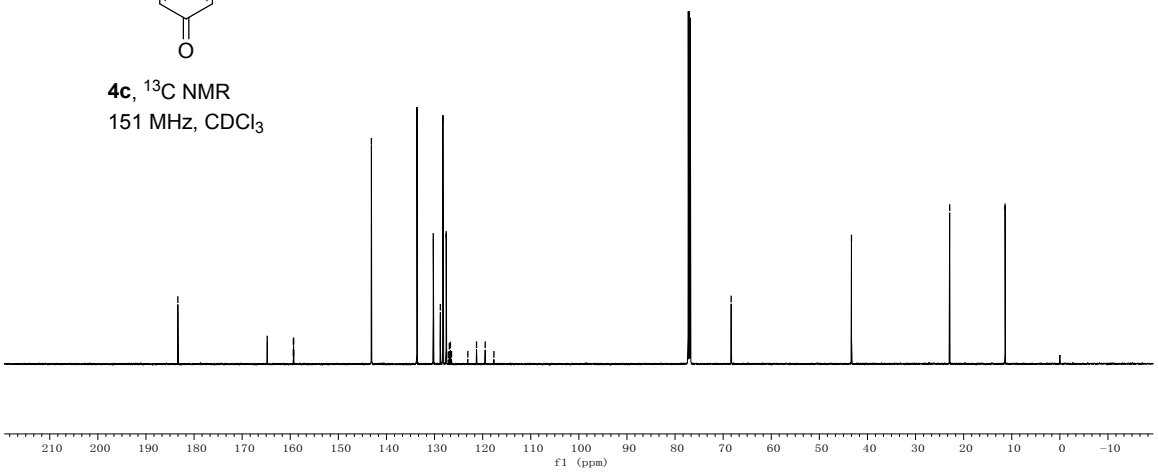
4c, ^{1}H NMR
400 MHz, $CDCl_3$

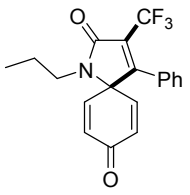




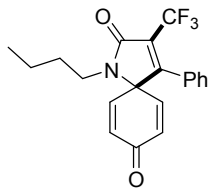


4c, ^{13}C NMR
151 MHz, $CDCl_3$

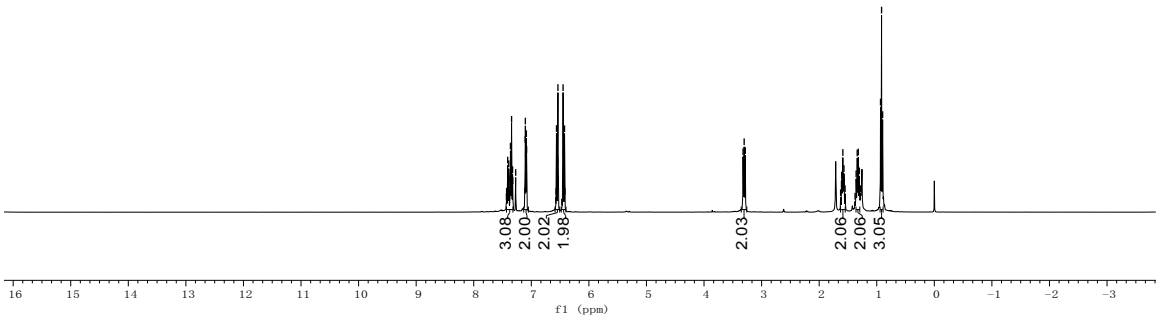


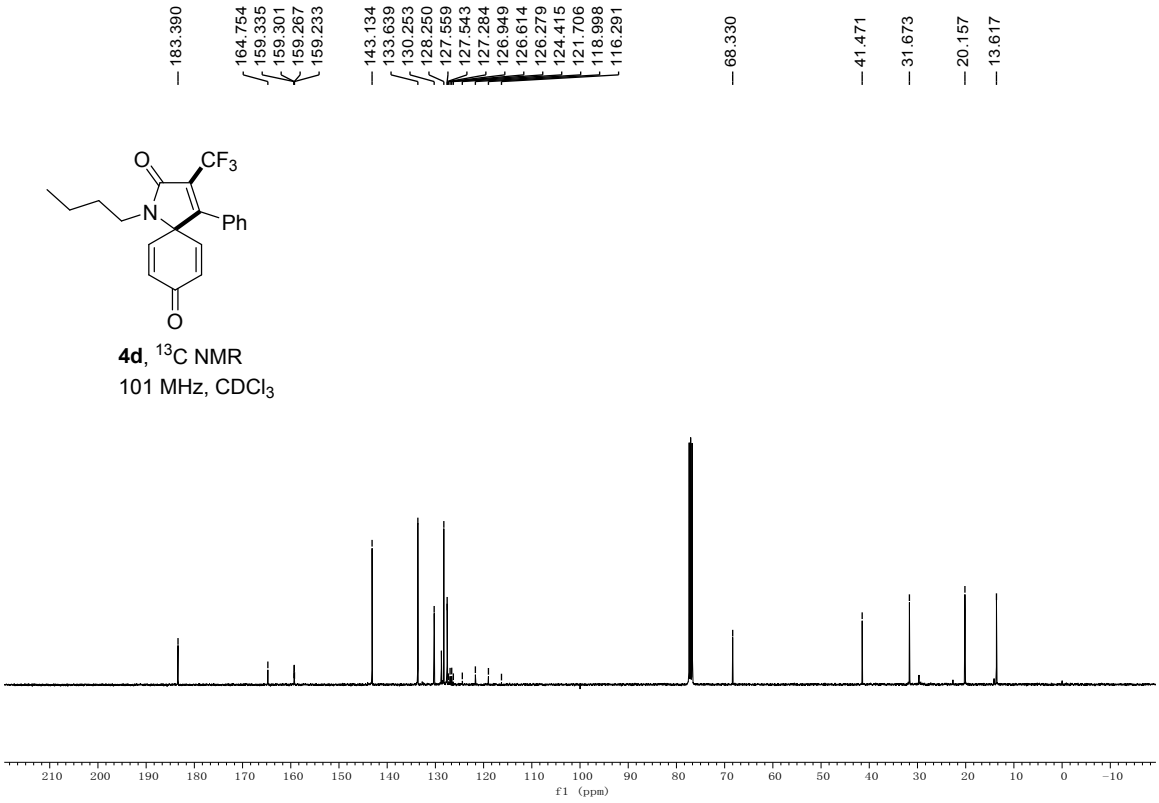


4c, ^{19}F NMR
376 MHz, $CDCl_3$

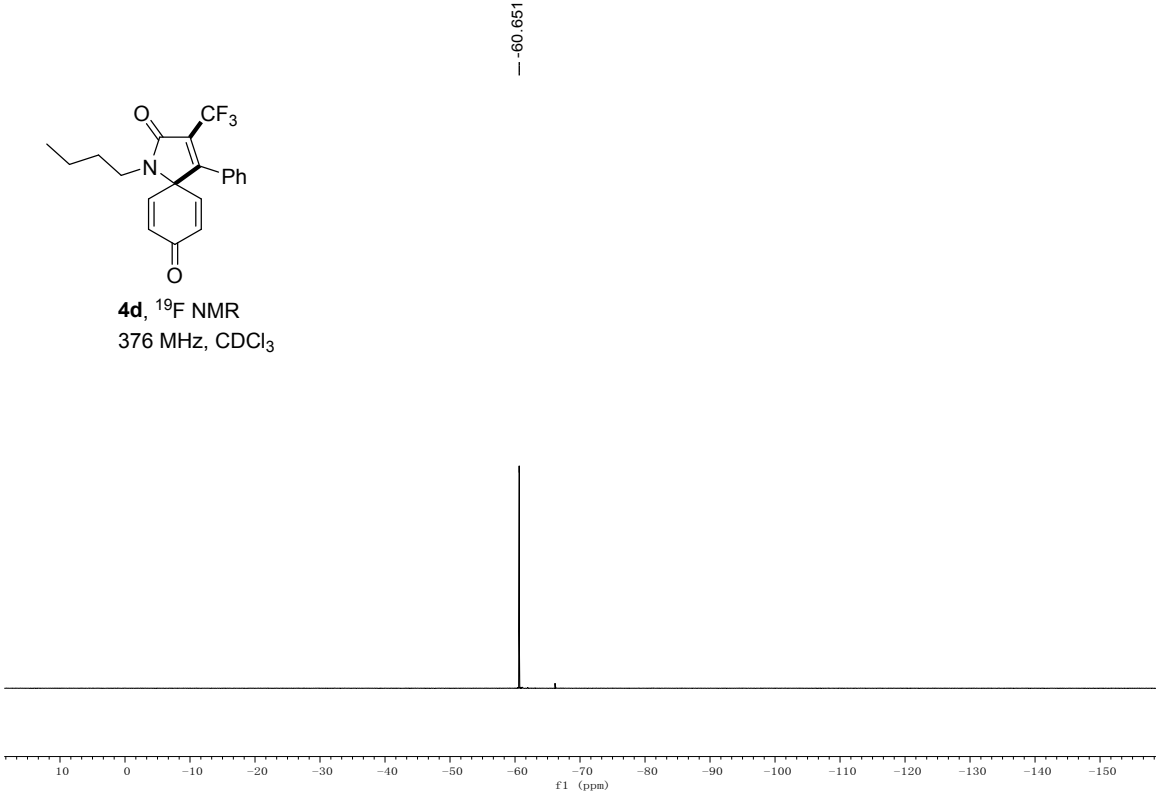


4d, ^{1}H NMR
400 MHz, $CDCl_3$

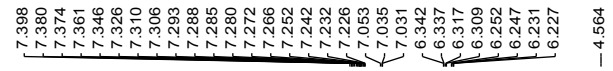


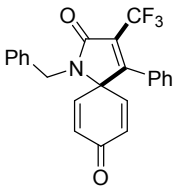


4d, ^{13}C NMR
101 MHz, $CDCl_3$

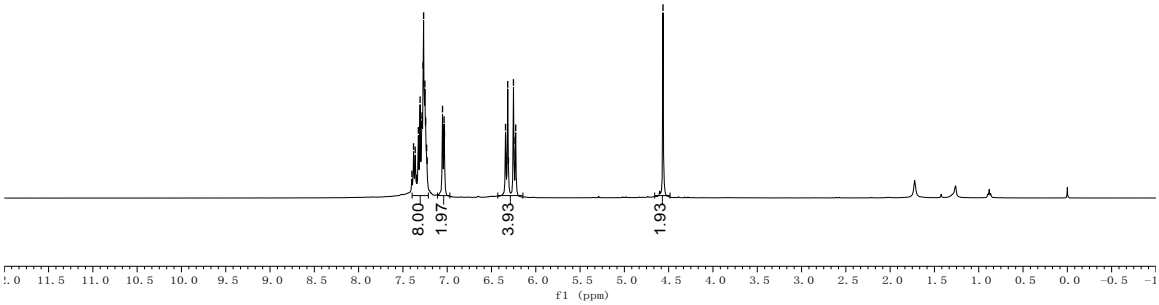


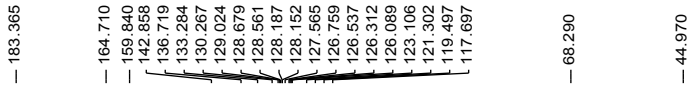
4d, ^{19}F NMR
376 MHz, $CDCl_3$

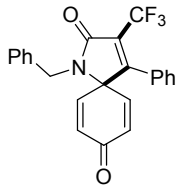




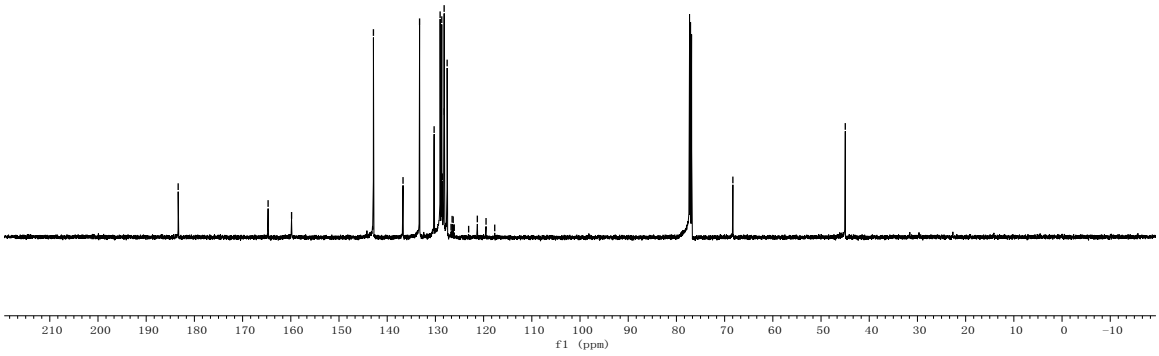
4e, ^{1}H NMR
400 MHz, $CDCl_3$

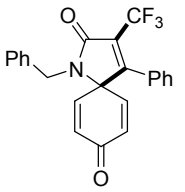




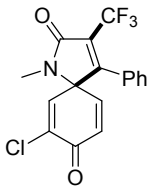


4e, ^{13}C NMR
151 MHz, $CDCl_3$

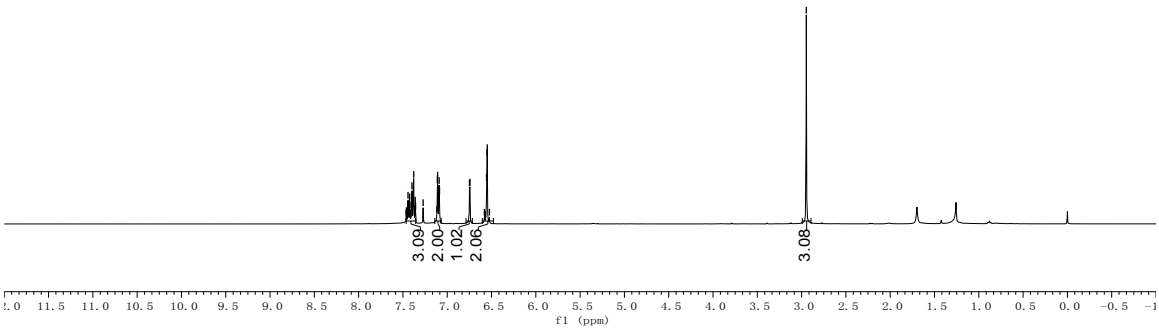


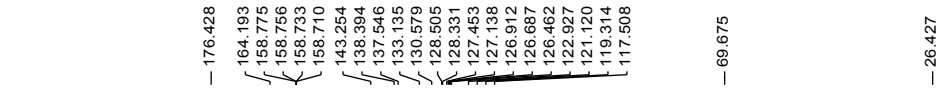


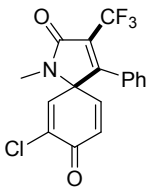
4e, ^{19}F NMR
376 MHz, $CDCl_3$



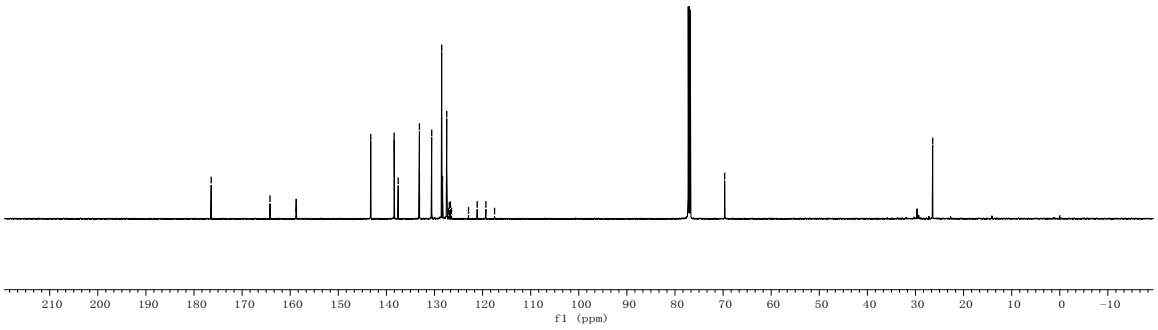
4f, ^{1}H NMR
400 MHz, $CDCl_3$

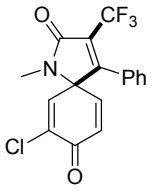




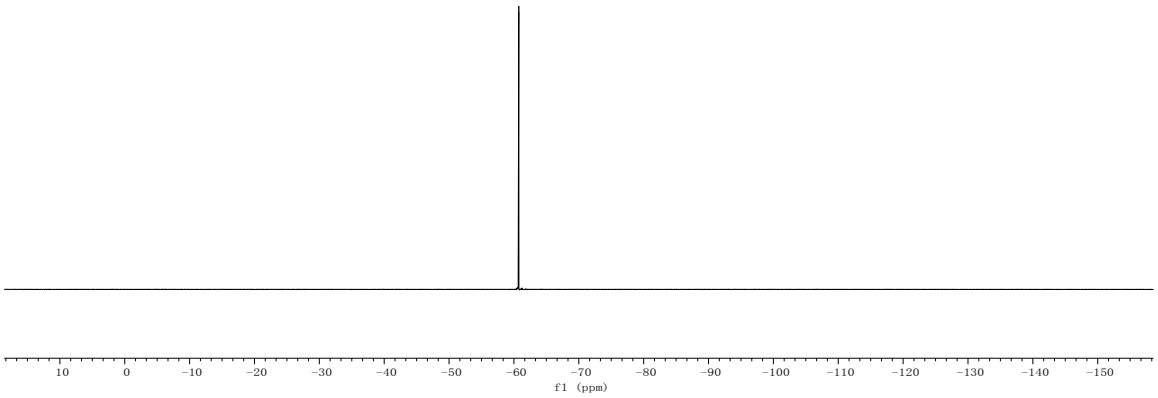


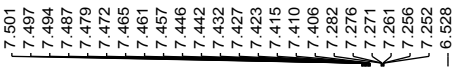
4f, ^{13}C NMR
151 MHz, $CDCl_3$

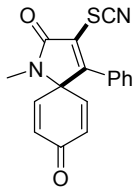




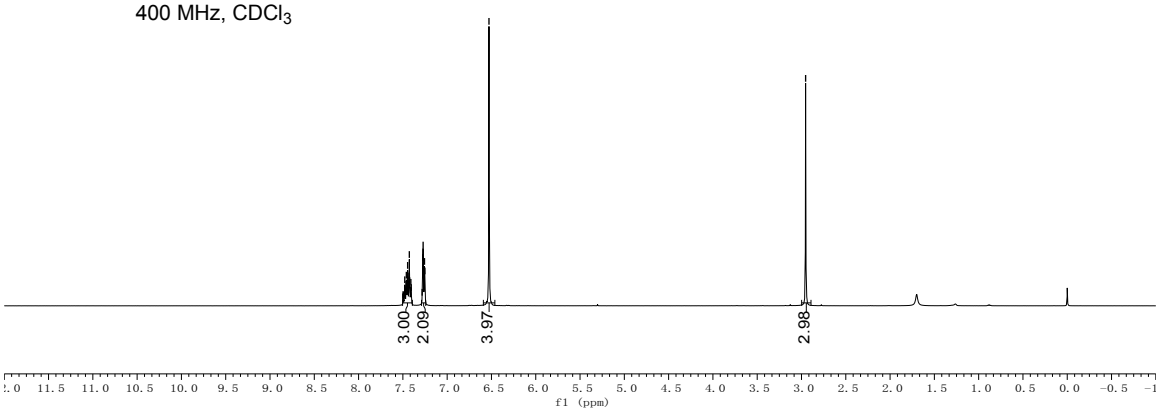
4f, ^{19}F NMR
376 MHz, $CDCl_3$

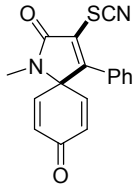




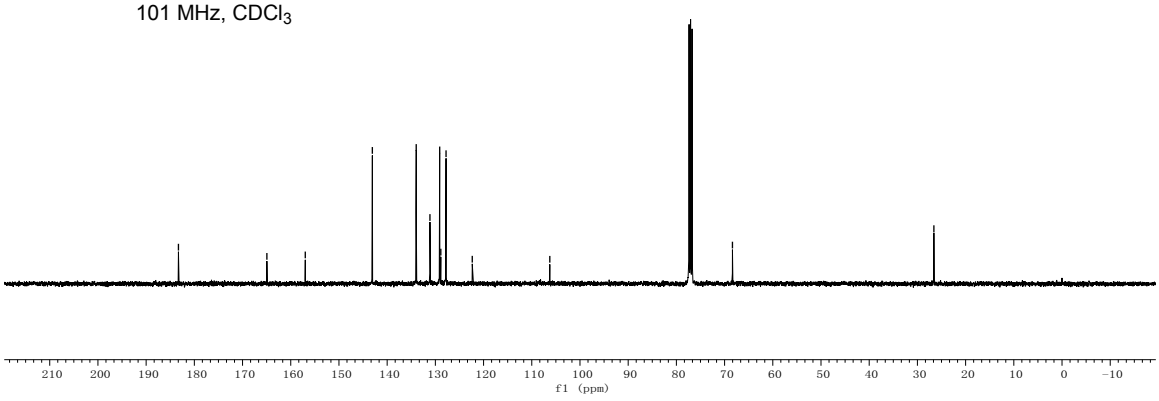


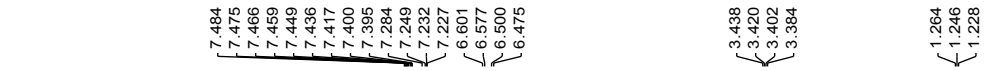
5a, ^{1}H NMR
400 MHz, $CDCl_3$

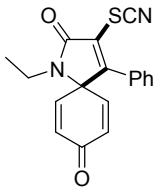




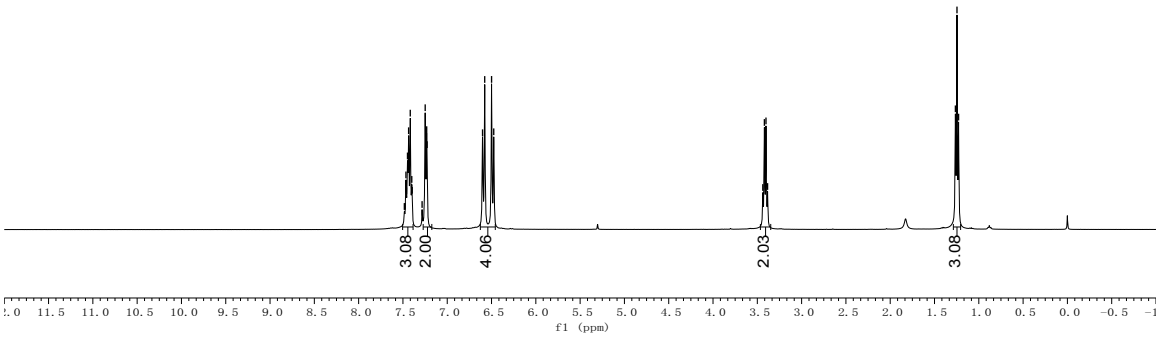
5a, ^{13}C NMR
101 MHz, $CDCl_3$

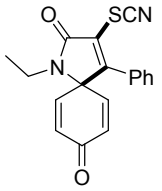




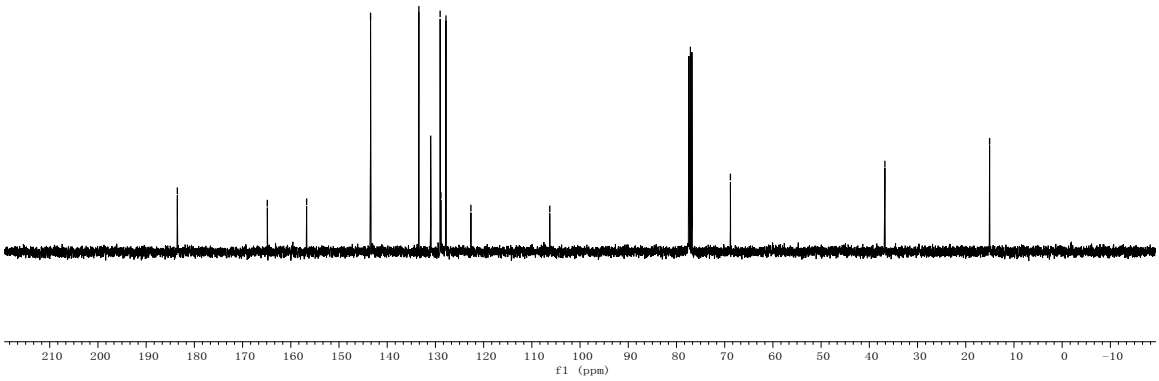


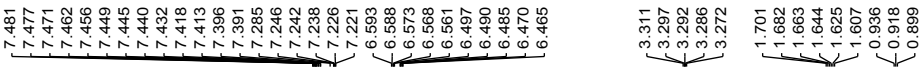
5b, ^{1}H NMR
400 MHz, $CDCl_3$

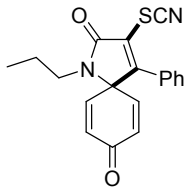




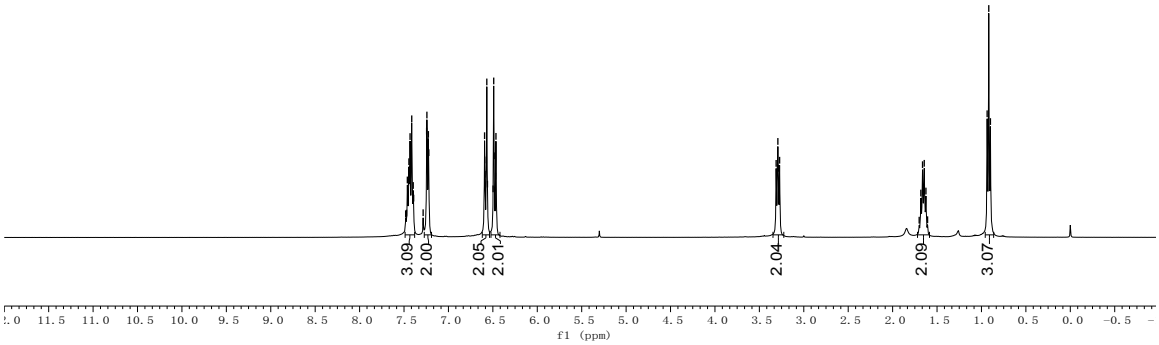
5b, ^{13}C NMR
101 MHz, $CDCl_3$

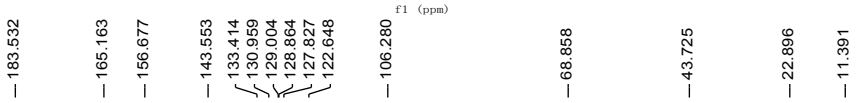


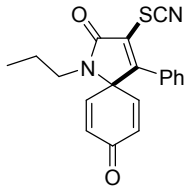




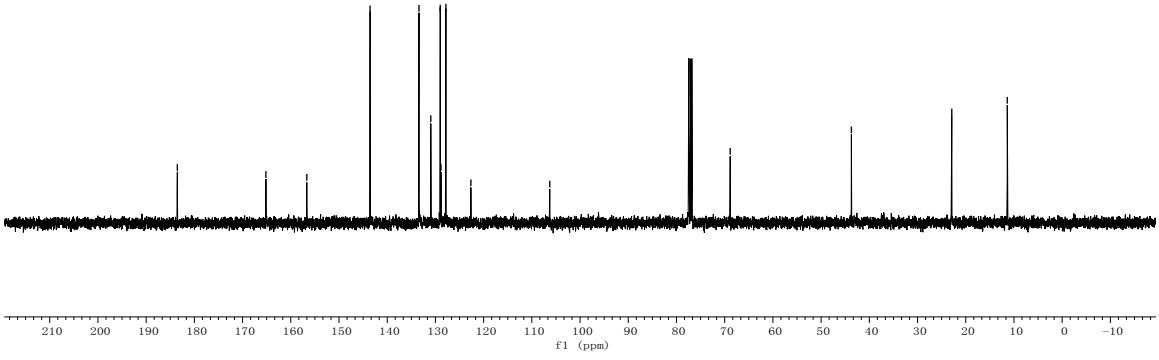
5c, ^{1}H NMR
400 MHz, $CDCl_3$

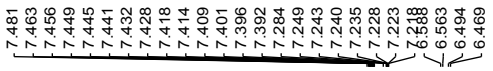


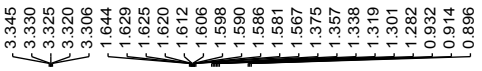


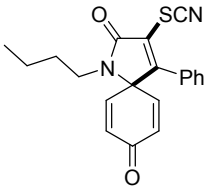


5c, ^{13}C NMR
101 MHz, $CDCl_3$

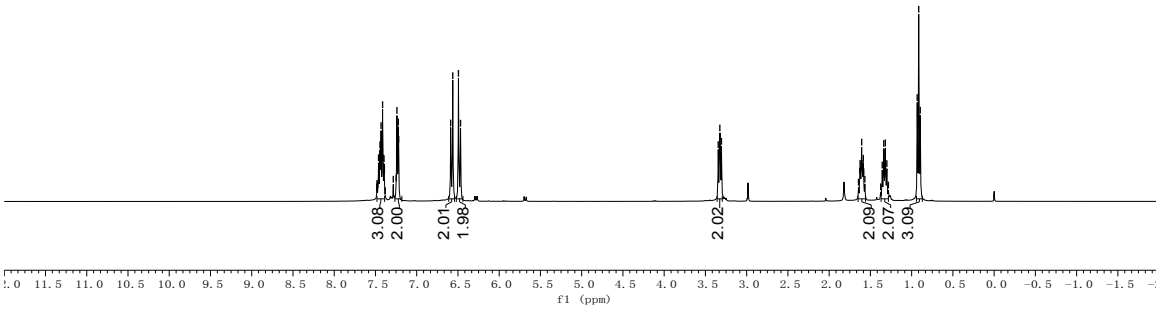


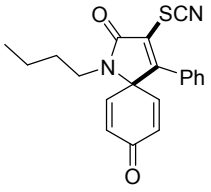




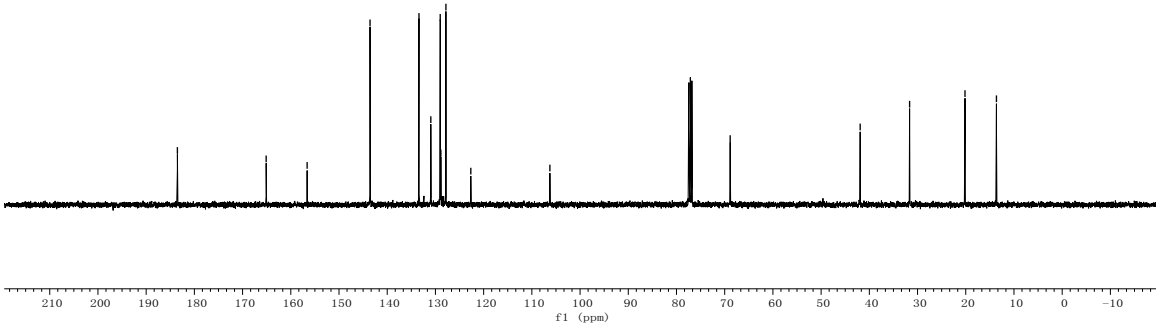


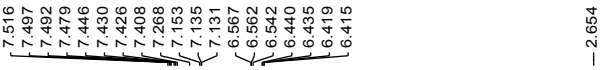
5d, ^{1}H NMR
400 MHz, $CDCl_3$

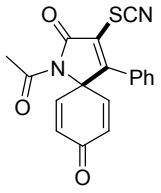




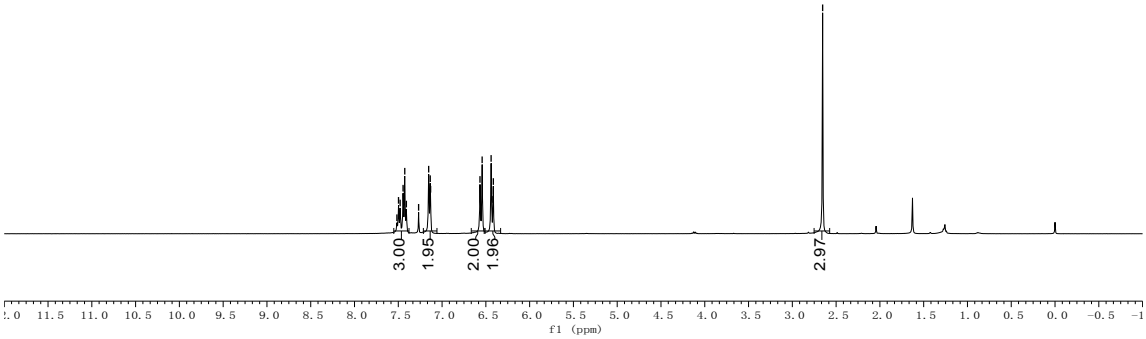
5d, ^{13}C NMR
101 MHz, $CDCl_3$

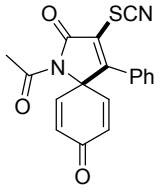




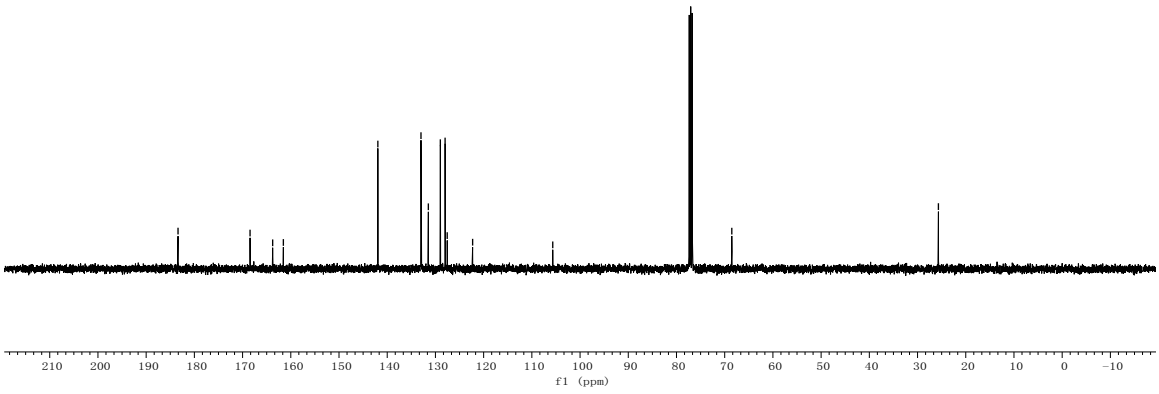


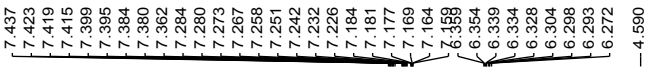
5e, ^{1}H NMR
400 MHz, $CDCl_3$

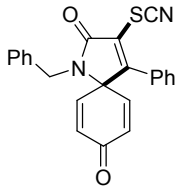




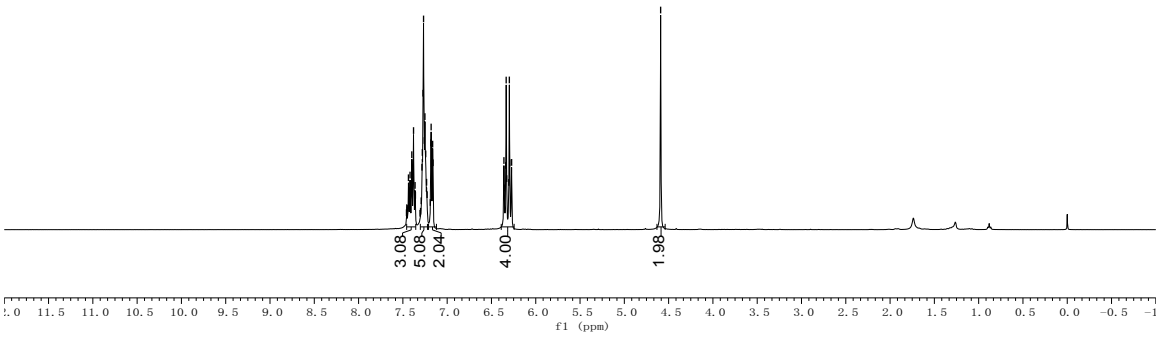
5e, ^{13}C NMR
101 MHz, $CDCl_3$

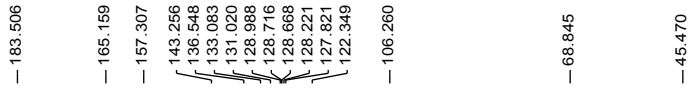


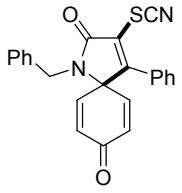




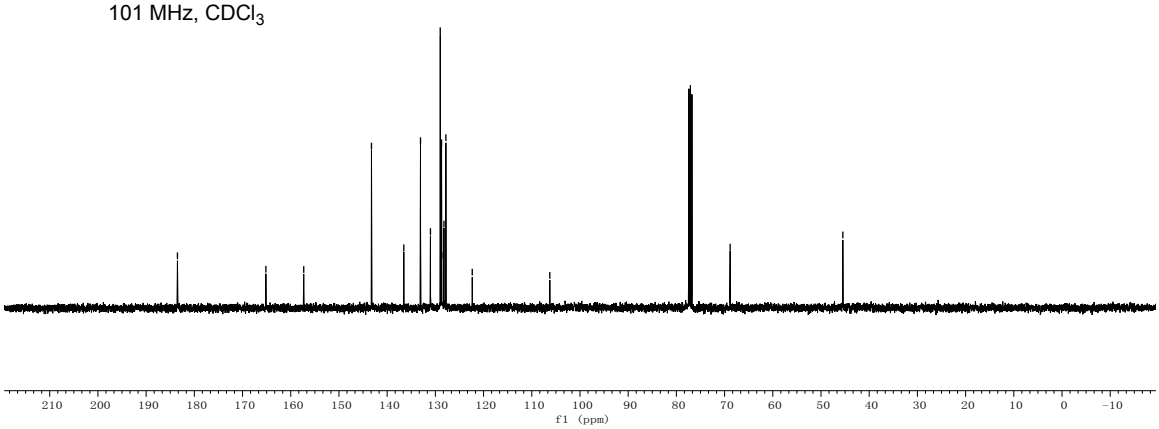
5f, ^{1}H NMR
400 MHz, $CDCl_3$

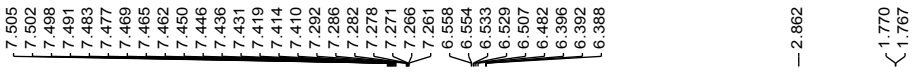


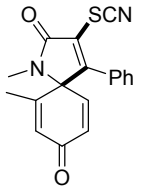




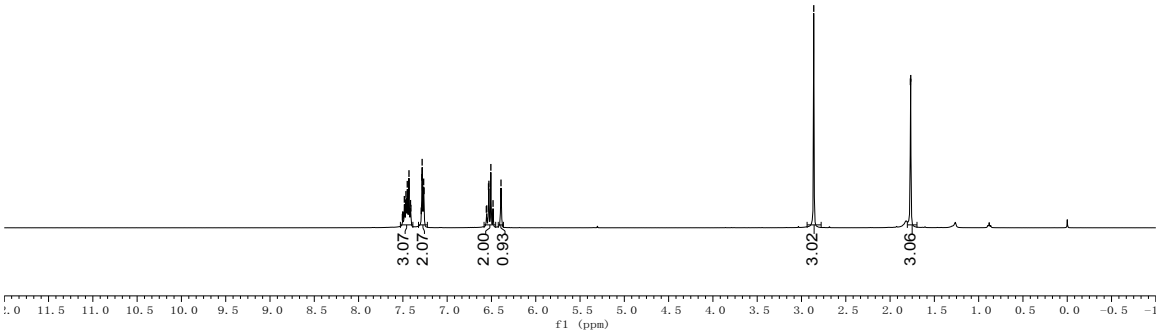
5f, ^{13}C NMR
101 MHz, $CDCl_3$

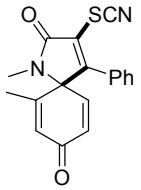




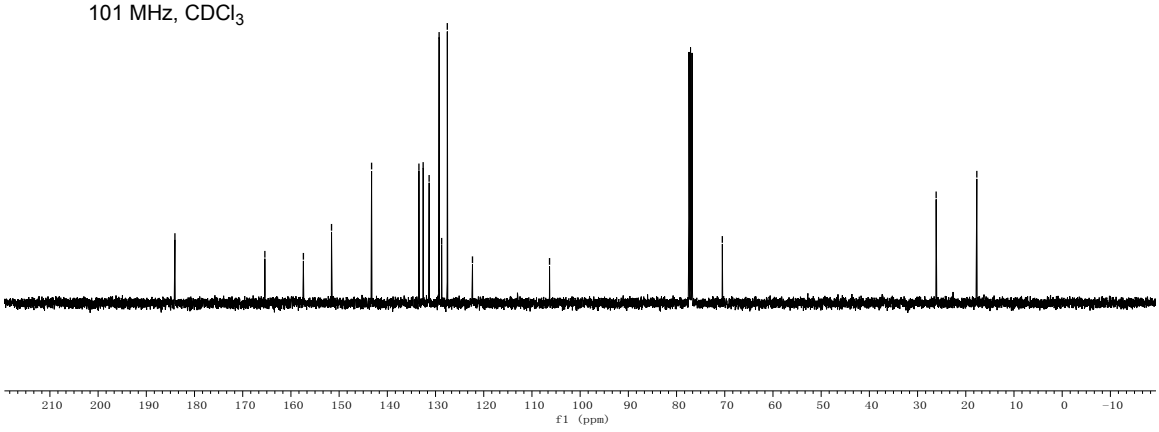


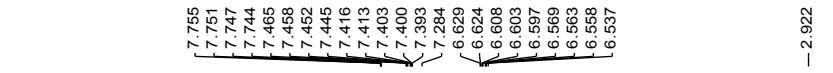
5g, ^{1}H NMR
400 MHz, $CDCl_3$

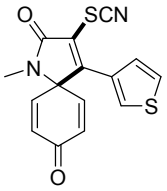




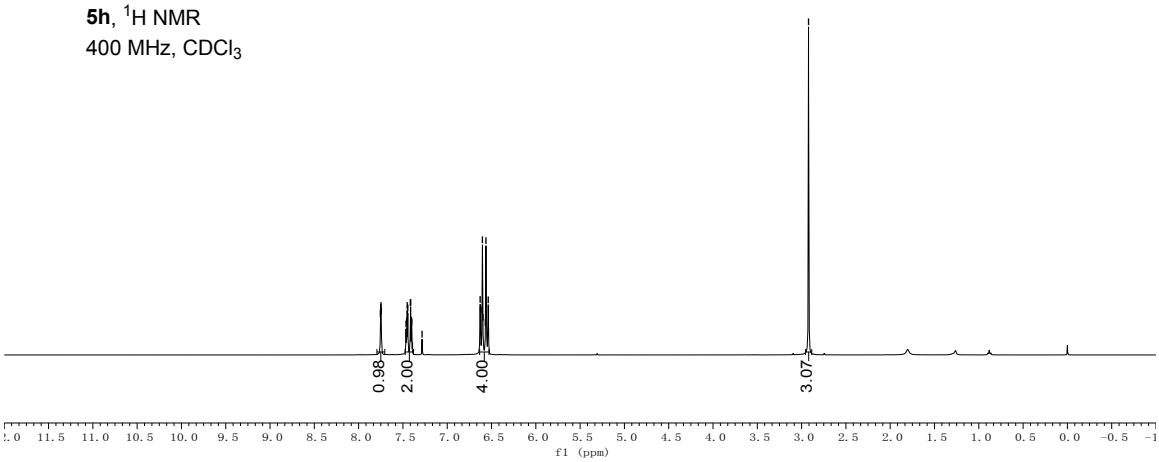
5g, ^{13}C NMR
101 MHz, $CDCl_3$

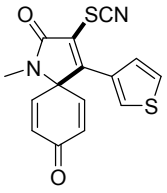




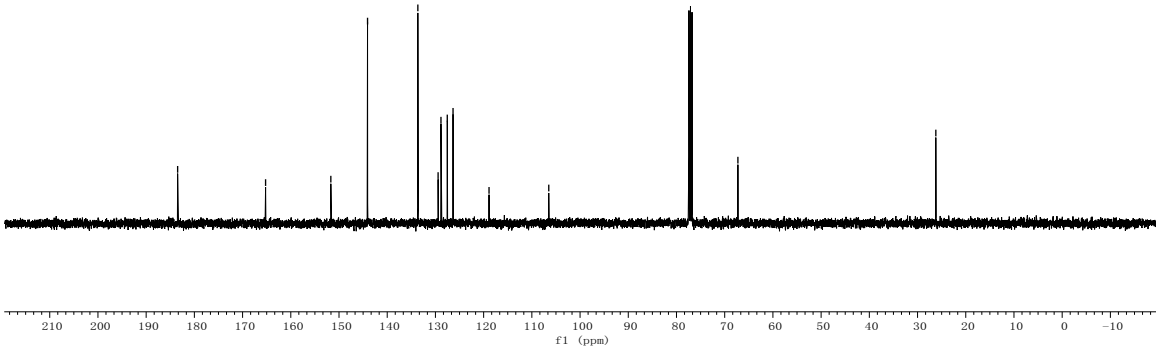


5h, ^{1}H NMR
400 MHz, $CDCl_3$





5h, ^{13}C NMR
101 MHz, $CDCl_3$



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