Electronic Supplementary Information

Modular synthesis of spirocyclic carbonates: Unravelling the synergistic interplay of electronic and electrostatic sites on phenolic catalyst

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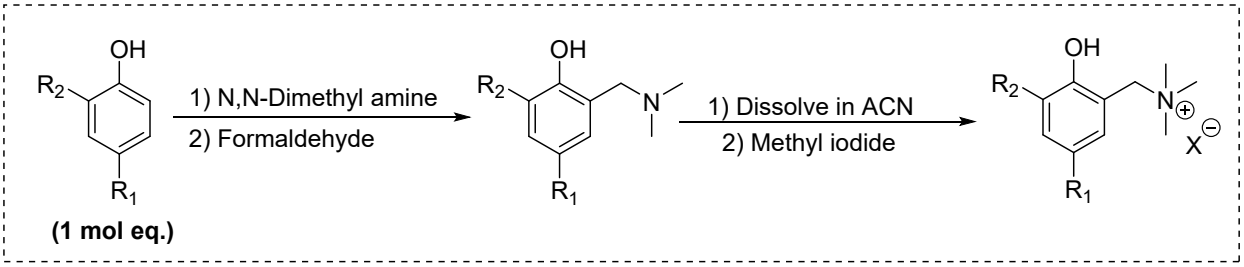
1. General Methods

Reagents and solvents were obtained from commercial sources and used as received. Air- and moisture-sensitive liquids were transferred via a syringe and a stainless-steel needle. Reactions were magnetically stirred and monitored by thin-layer chromatography. All work-up and purification procedures were carried out with reagent-grade solvents under ambient atmosphere. For separation of products column chromatography was carried out using Finar 100-200 mess silica as stationary phase.

Nuclear magnetic resonance (NMR) spectra were acquired on a 500 MHz and 600 MHz Bruker Avance III spectrometer. ^{1}H and ^{13}C NMR chemical shifts are reported in ppm and referenced to tetramethylsilane or residual solvent peaks as internal standards (for $CDCl_3$, tetramethylsilane) ppm for ^{1}H and $CDCl_3$ 77.16 ppm for ^{13}C; for DMSO-d6, 2.50 ppm for ^{1}H and 39.52 ppm for ^{13}C. NMR data are reported as follows: chemical shifts, multiplicity (s, singlet; d, doublet; dd, doublet of doublet; t, triplet; td, triplet of doublet; q, quartet; m, multiplet; br, broad signal), coupling constants (Hz), and integration. HPLC analysis was carried out using a Shimadzu SPD-M20A diode array detector. HRMS spectra was recorded on LCMS Spectrometer Model Q-ToF. FT-IR spectra was recorded using a IRAffinity-1S Shimadzu instrument.

2. General procedure for catalyst synthesis

General synthesis



Scheme S1: General scheme for synthesis of **PDC** catalysts

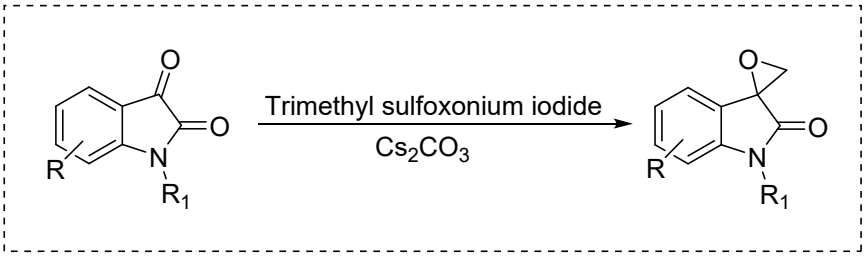
The procedure for preparing the phenol-alkylammonium salt was adapted from the literature.[1] In a round-bottom flask, an equivalent amount of phenol was placed, and then aqueous solutions containing 1.5 equivalents of dimethylamine (in a 33% solution) and 1.5 equivalents of formaldehyde (in a 37% solution) were added. The resulting mixture was stirred at room

temperature overnight. Afterward, the reaction mixture was subjected to an ethyl acetate/water workup to extract the product. The solvent was subsequently removed under reduced pressure. The resulting crude product was dissolved in acetonitrile, and then either methyl iodide, trimethyloxonium tetrafluoroborate, or methyltrifluoromethane sulfonate (all in 1.5 equivalents) was added. The reaction mixture was allowed to stir for 24 hours at room temperature. The precipitated product was then filtered and washed with diethyl ether to obtain the pure product of PDCs.

Synthetic procedure for *N*-methyl quininium iodide (*N*-Me-QI)

The *N*-methyl quininium iodide was synthesized by adopting the literature procedure.{McNeice, 2020 #1565} Appropriate amount of quinine (1.04 g) was taken in a round bottom flask and 30 mL of methanol was added to obtain a transparent solution. To this solution, methyl iodide (0.6 g) was added and allow the reaction mixture to stir for 12 h at 28 ^{o}C. The solvent is evaporated under reduced pressure to yield a yellow solid product. The obtained solid was recrystallized using methanol and the crystalline solid was allowed to wash with diethyl ether and dried under vacuum condition for 12 h. The product was confirmed using NMR spectra.

3. General procedure for the synthesis of spiro-epoxyoxindole

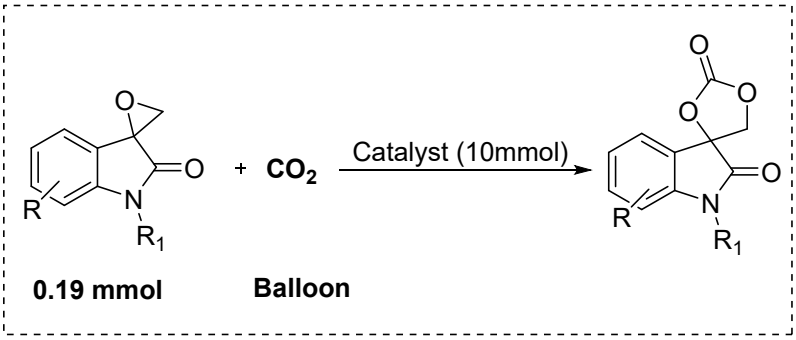


Scheme S2: General procedure for the synthesis of spiro-epoxyoxindole

Trimethylsulfoxonium iodide (2.0 mmol) and cesium carbonate (4.0 mmol) was taken in round bottom flask with dry CH_3CN and stirred at 50 °C under inert atmosphere for 1h which will generate sulphur ylide.[2] Next, a solution of 2.0 mmol of isatin in 10 mL of dry CH_3CN was added dropwise over a period of 10 minutes. The progress of the reaction was monitored by thin-layer chromatography (TLC). Once the reaction was complete, the reaction mixture was filtered through a Celite bed, and the filtrate was then evaporated to concentrate the product. The final purification

of the product was achieved by column chromatography using silica gel as the stationary phase and a mobile phase consisting of n-hexane/ethyl acetate in a 90:10 ratio.

4. General procedure for ring expansion reaction



Scheme S3: General procedure for cycloaddition reaction

In a 10 mL Pyrex tube, spiro-epoxyoxindoles (0.19 mmol) were placed along with PDC catalyst (10 mol%) and 1 mL of solvent. The reaction tube was promptly sealed, a CO_2 balloon was attached, and the mixture was stirred at 40°C. The progress of the reaction was monitored for completion using TLC with a eluent mixture of n-hexane and ethyl acetate. Upon completion of the reaction, the crude compound was subjected to purification through column chromatography on silica gel, resulting in the isolation of the pure compound.

5. Density Measurements

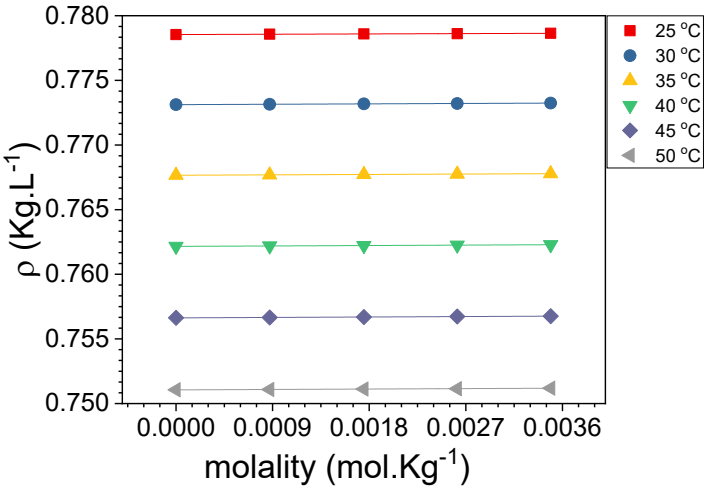


Fig. S1 Density of PDC-3 in acetonitrile

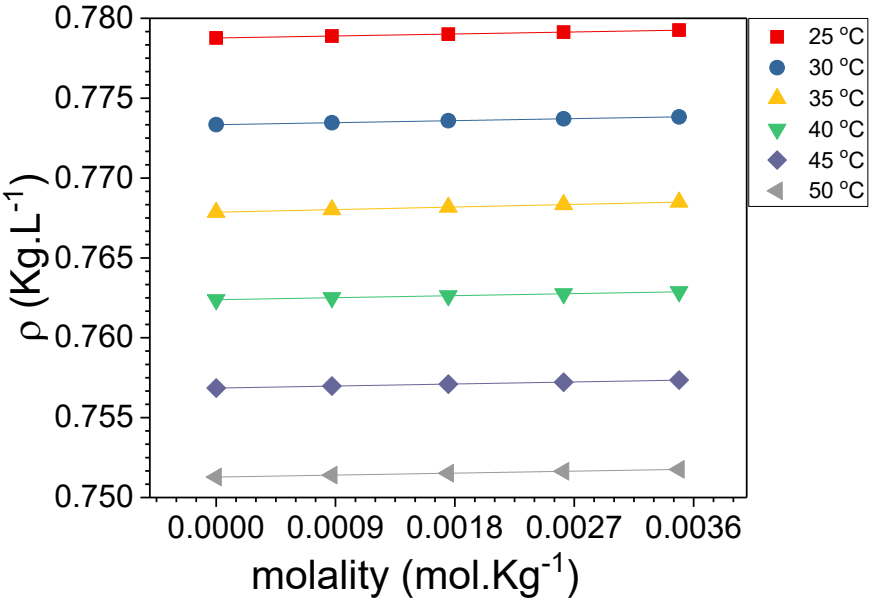


Fig. S2 Density of PDC-3 and 1a in acetonitrile

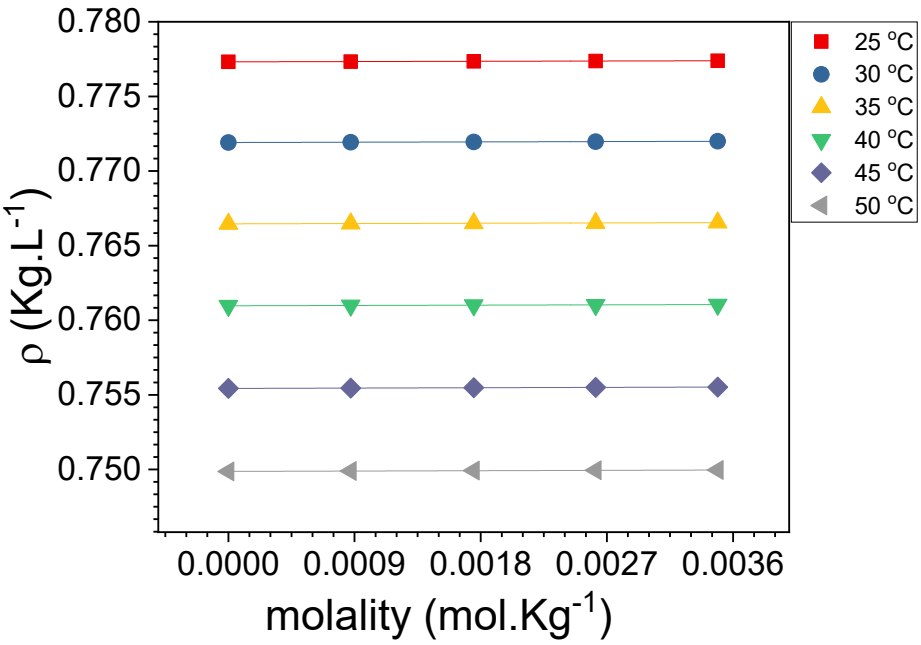


Fig. S3 Density of PDC-7 in acetonitrile

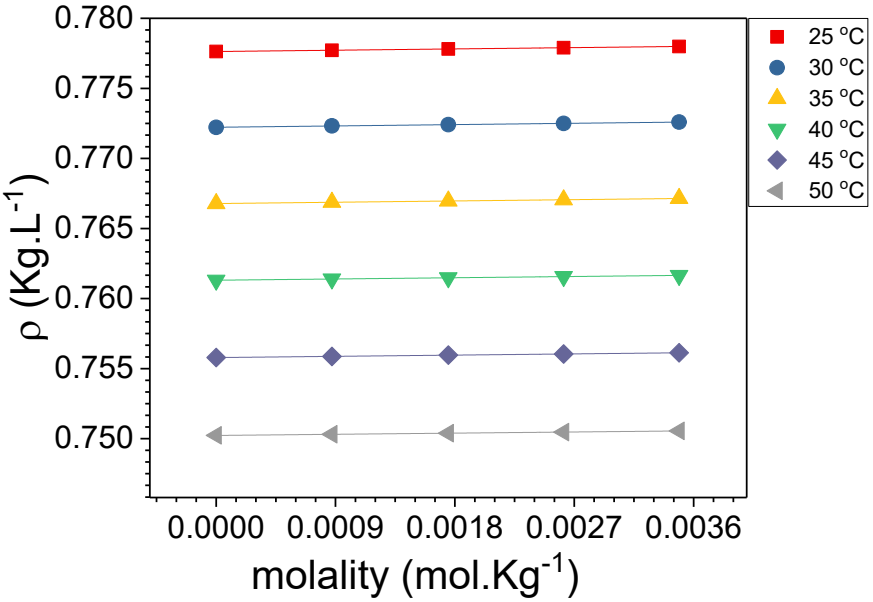


Fig. S4 Density of PDC-7 and 1a in acetonitrile

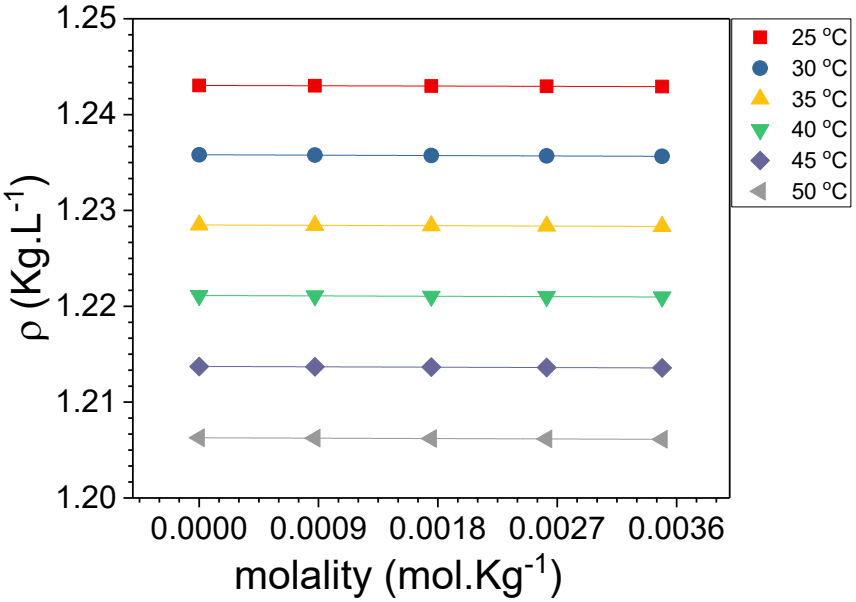


Fig. S5 Density of PDC-3 in dichloroethane

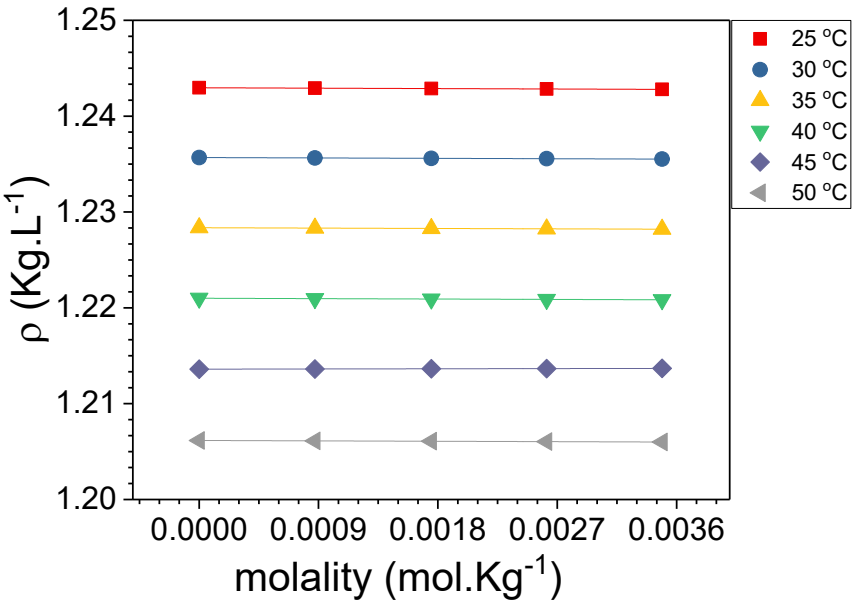


Fig. S6 Density of PDC-3 and 1a in dichloroethane

6. Equations followed to calculate volumetric parameters[3]

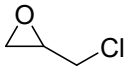
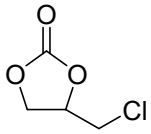
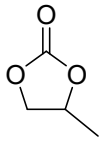
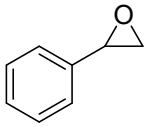
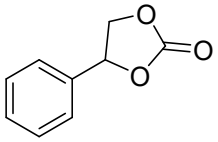
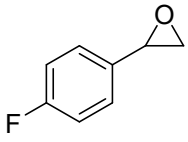
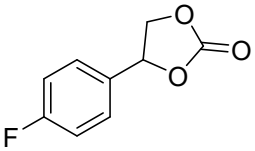
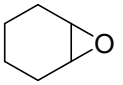
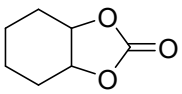
Eq. S1

$$V_{\emptyset} = {}^{M}/_{\rho} - (\rho - \rho_{\circ})/m_A \rho \rho_{\circ}$$

Eq. S2

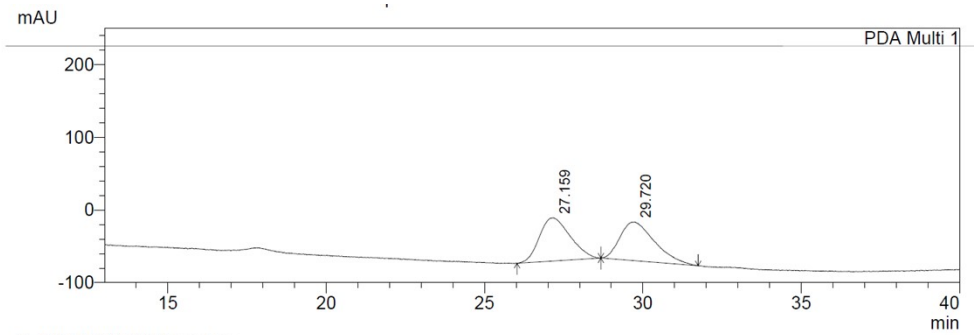
$$V_{\emptyset} = V°_{\emptyset} + S_V^* m_a$$

7. Extended substrate scope for the PDC-3 catalyzed chemical fixation of CO_2 Cyclic carbonate

Entry	Substrate (Epoxide)	Reaction product	Conversion %	Selectivity %
1			99	98
2			99	99
3			96	98
4			91	99
5[a]			26	99

[a]The reactions are carried out with catalyst (10 mol%), substrate (1 mmol), CO_2 (balloon) using solvent (1 mL) at 40 [o]C for 13 h. [b]Time 24 h.

8. HPLC profile of chiral spiroepoxy oxindole



1 PDA Multi 1/254nm 4nm

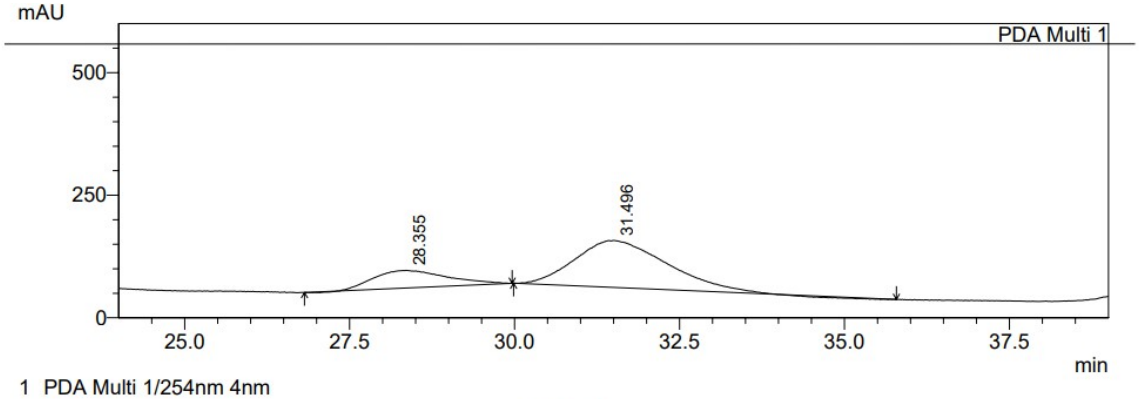
PeakTable

PDA Ch1 254nm 4nm

Peak#	Ret. Time	Area	Height	Area %	Height %
1	27.159	4008018	59703	49.706	52.883
2	29.720	4055415	53194	50.294	47.117
Total		8063433	112897	100.000	100.000

Chiral HPLC analysis: Chiralpak AD-H, hexane/iPrOH=90/10, flow rate 1.0 mL/min.

Fig. S7. HPLC chromatogram of Spectrum 1'-benzylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



1 PDA Multi 1/254nm 4nm

PeakTable

PDA Ch1 254nm 4nm

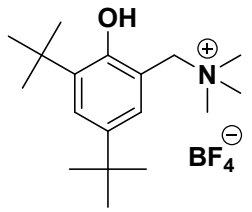
Peak#	Ret. Time	Area	Height	Area %	Height %
1	28.355	2775240	36108	23.154	27.342
2	31.496	9210746	95953	76.846	72.658
Total		11985985	132060	100.000	100.000

Chiral HPLC analysis: Chiralpak AD-H, hexane/iPrOH=90/10, flow rate 1.0 mL/min.

Fig. S8. HPLC chromatogram of Spectrum 1'-benzylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione (chiral)

9. Characterization data of catalysts[1]

1. 1-(2-hydroxy-3,5-dimethylphenyl)-*N*,*N*,*N*-trimethylmethanaminium tetrafluoroborate (PDC-1)

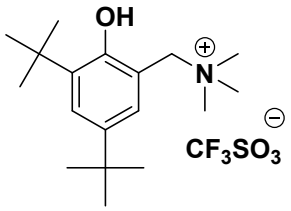


Yield: 85%

^{1}H NMR (500 MHz, CD_3CN) δ 7.72 (d, *J* = 2.5 Hz, 1H), 7.50 (d, *J* = 2.5 Hz, 1H), 6.51 (s, 1H), 4.51 (d, *J* = 5.7 Hz, 2H), 3.04 (d, *J* = 5.1 Hz, 6H), 2.36 (s, 3H), 1.68 (s, 9H), 1.56 (s, 9H).

^{13}C NMR (126 MHz, CD_3CN) δ 151.87, 145.21, 139.23, 127.86, 127.11, 120.16, 59.26, 43.52, 35.23, 34.97, 31.66, 31.50, 30.80, 30.16, 29.65.

2. 1-(3,5-di-tert-butyl-2-hydroxyphenyl)-*N*,*N*,*N*-trimethylmethanaminium trifluoromethanesulfonate (PDC-2)

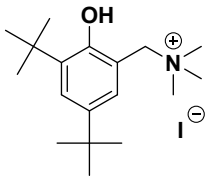


Yield : 86%

^{1}H NMR (500 MHz, CD_3CN) δ 7.50 (d, *J* = 2.5 Hz, 1H), 7.25 (d, *J* = 2.5 Hz, 1H), 6.50 (s, 1H), 4.46 (s, 2H), 2.99 (s, 9H), 1.41 (s, 9H), 1.31 (s, 9H).

^{13}C NMR (126 MHz, CD_3CN) δ 153.33, 144.66, 139.91, 130.13, 127.96, 65.85, 53.49, 53.45, 53.42, 35.55, 34.99, 31.56, 30.21.

3. 1-(2-hydroxy-3,5-dimethylphenyl)-N,N,N-trimethylmethanaminium iodide (PDC-3)

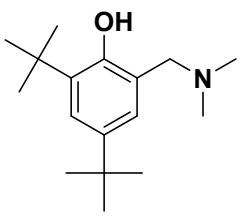


Yield : 89%

^{1}H NMR (500 MHz, DMSO-d6) δ 8.66 (s, 1H), 7.39 (s, 1H), 7.32 (s, 1H), 4.68 (s, 2H), 3.12 (s, 6H), 2.76 (s, 3H), 1.40 (s, 9H), 1.29 (s, 9H).

^{13}C NMR (126 MHz, DMSO-d6) δ 153.23, 142.06, 138.82, 129.55, 125.90, 116.86, 63.40, 54.39, 48.06, 34.83, 33.99, 31.28, 29.79.

4. 2,4-di-tert-butyl-6-((dimethylamino)methyl)phenol (DMAMP) (PDC-4)

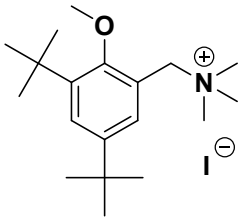


^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.21 (d, *J* = 2.5 Hz, 1H), 6.81 (d, *J* = 2.5 Hz, 1H), 3.60 (s, 2H), 2.31 (s, 6H), 1.42 (s,

9H), 1.28 (s, 8H).

^{13}C NMR (126 MHz, Chloroform-*d*) δ 154.61, 140.41, 135.51, 123.24, 122.91, 121.44, 63.74, 44.45, 34.97, 34.26, 31.84, 29.74

5. 1-(3,5-di-tert-butyl-2-methoxyphenyl)-*N,N,N*-trimethylmethanaminium iodide (PDC-5)

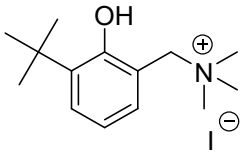


Yield : 48%

^{1}H NMR (500 MHz, DMSO-d6) δ 7.48 (d, *J* = 2.6 Hz, 1H), 7.42 (d, *J* = 2.6 Hz, 1H), 4.51 (s, 2H), 3.78 (s, 3H), 2.97 (s, 9H), 1.38 (s, 9H), 1.30 (s, 9H).

^{13}C NMR (126 MHz, DMSO-d6) δ 140.02, 136.23, 88.29, 73.70, 61.41, 44.50, 43.74, 9.61

6. 1-(3-(tert-butyl)-2-hydroxyphenyl)-N,N,N-trimethylmethanaminium iodide (PDC-6)

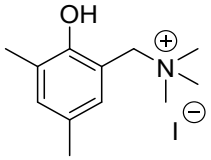


Yield : 75%

^{1}H NMR (500 MHz, DMSO-d6) δ 8.96 (s, 1H), 7.39 (d, *J* = 9.7 Hz, 1H), 7.28 (d, *J* = 7.5 Hz, 1H), 6.96 (s, 1H), 4.64 (s, 2H), 3.02 (s, 9H), 1.38 (s, 9H).

^{13}C NMR (126 MHz, DMSO-d6) δ 155.66, 139.25, 132.46, 129.29, 120.22, 116.85, 63.47, 54.51, 51.86, 34.71, 29.66

7. 1-(3,5-di-tert-butyl-2-methoxyphenyl)-*N,N,N*-trimethylmethanaminium iodide (PDC-7)

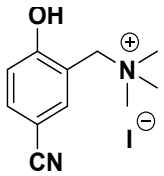


Yield : 49%

^{1}H NMR (500 MHz, DMSO-d6) δ 8.73 (s, 1H), 7.12 (s, 3H), 4.38 (s, 3H), 3.13 (s, 6H), 2.19 (s, 9H).

^{13}C NMR (126 MHz, DMSO-d6) δ 154.30, 132.41, 123.82, 117.48, 66.16, 53.78, 53.74, 46.98, 15.96.

8. 1-(5-cyano-2-hydroxyphenyl)-*N,N,N*-trimethylmethanaminium iodide (PDC-8)

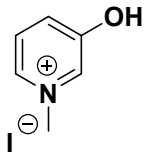


Yield: 82%

^{1}H NMR (500 MHz, DMSO-d6) δ 11.59 (s, 1H), 7.91 (d, J = 2.2 Hz, 1H), 7.82 (dd, J = 8.5, 2.1 Hz, 1H), 7.11 (d, J = 8.6 Hz, 1H), 4.48 (s, 2H), 3.06 (s, 9H).

^{13}C NMR (126 MHz, DMSO-d6) δ 161.57, 139.22, 136.09, 118.92, 117.29, 116.31, 101.54, 61.93, 52.28.

9. 3-hydroxy-1-methylpyridin-1-ium iodide (PDC-9)



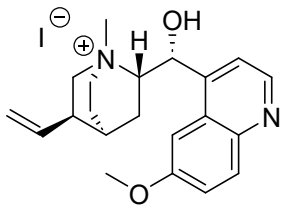
Yield: 80%

^{1}H NMR (600 MHz, DMSO-d6) δ 8.21 – 8.15 (m, 2H), 7.72 (dd, J = 8.9, 5.7 Hz, 1H), 7.64 (dd, J = 8.8, 2.6 Hz, 1H), 4.51 (s, 3H).

^{13}C NMR (151 MHz, DMSO-d6) δ 160.83, 134.47, 133.55, 131.72, 128.48, 48.16.

10. Characterization data of chiral N-Me-QI catalyst{McNeice, 2020 #1565}

***N*-methyl Quininium iodide**



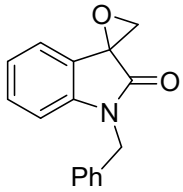
^{1}H NMR (600 MHz, DMSO-d6) δ 1H NMR (600 MHz, DMSO-D6) δ 8.75 (d, J = 4.5 Hz, 1H), 7.96 (d, J = 9.2 Hz, 1H), 7.66 (d, J = 4.6 Hz, 1H), 7.43 (dd, J = 9.4, 2.5 Hz, 1H), 7.17 (d, J = 2.7 Hz, 1H), 6.47 (d, J = 3.9 Hz, 1H), 6.19 (d, J = 4.0 Hz, 1H), 5.71 (ddd, J = 17.0, 10.3, 6.6 Hz, 1H), 5.11 (s, 1H), 4.97 (d, J = 10.4 Hz, 1H), 3.97 (s, 3H), 3.79 – 3.69 (m, 1H), 3.62 (dtd, J = 26.2, 8.3, 4.6 Hz, 2H), 3.38 (s, 3H), 3.12 (s, 1H), 2.78 (q, J = 7.7 Hz, 1H), 2.18 – 2.07 (m, 2H), 2.01 (q, J = 3.1 Hz, 1H), 1.90 (dt, J = 14.8, 7.5 Hz, 1H), 1.36 – 1.24 (m, 1H).

^{13}C NMR (151 MHz, DMSO-d6) δ 13C NMR (151 MHz, DMSO-D6) δ 157.96, 148.01, 144.39, 144.19, 138.57,

132.00, 125.71, 122.15, 120.51, 117.07, 102.12, 67.42, 64.46, 56.17, 54.76, 49.40, 49.14, 46.30, 38.08, 26.44, 25.15, 19.92.

11. Characterization data of spiro-epoxyoxindole[2{Parmar, 2019 #1564}]

1. 1-benzylspiro[indoline-3,2'-oxiran]-2-one

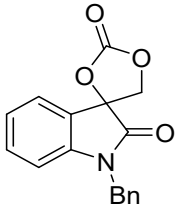


^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.39 – 7.22 (m, 6H), 7.11 (d, *J* = 7.7 Hz, 1H), 7.04 (t, *J* = 7.5 Hz, 1H), 6.80 (d, *J* = 8.0 Hz, 1H), 5.04 – 4.90 (m, 2H), 3.65 (d, *J* = 6.8 Hz, 1H), 3.47 (d, *J* = 6.8 Hz, 1H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.74, 160.21, 158.60, 140.17, 135.08, 129.04,127.47, 124.55, 116.86, 116.70, 110.77, 110.54, 110.37, 56.49, 54.58, 44.56.

12. Characterization data of products[2{Parmar, 2019 #1564}]

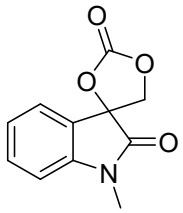
1. 1'-benzylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; Melting point 153-155 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.46 (d, *J* = 7.6 Hz, 1H), 7.32 (dt, *J* = 28.0, 7.8 Hz, 7H), 7.15 (t, *J* = 7.5 Hz, 1H), 6.80 (d, *J* = 7.5 Hz, 1H), 4.95 (d, *J* = 15.3 Hz, 1H), 4.84 (d, *J* = 15.8 Hz, 1H), 4.78 (d, *J* = 8.6 Hz, 1H), 4.58 (d, *J* = 8.9 Hz, 1H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.65, 153.78, 143.67, 134.53, 132.52, 129.20, 128.32, 127.49, 125.11, 124.28, 123.50, 110.55, 79.27, 70.93, 44.43

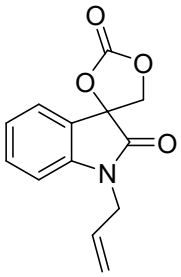
2. 1'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; Melting point 171-173 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.47 (dd, *J* = 9.9, 7.6 Hz, 2H), 7.19 (t, *J* = 7.5 Hz, 1H), 6.91 (d, *J* = 7.2 Hz, 1H), 4.72 (d, *J* = 9.1 Hz, 1H), 4.55 (d, *J* = 8.5 Hz, 1H), 3.23 (s, 3H).

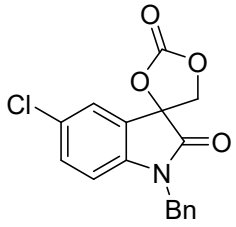
^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.40, 153.81, 144.48, 132.62, 124.98, 124.25, 123.56, 109.52, 79.24, 70.90, 26.80.

3. 1'-allylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; Melting point 128-120 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.52 – 7.41 (m, 2H), 7.19 (t, *J* = 7.7 Hz, 1H), 6.91 (d, *J* = 7.8 Hz, 1H), 5.83 (ddt, *J* = 16.1, 10.4, 5.3 Hz, 1H), 5.33 – 5.24 (m, 2H), 4.74 (d, *J* = 9.3 Hz, 1H), 4.56 (d, *J* = 9.1 Hz, 1H), 4.34 (qd, *J* = 16.3, 5.6 Hz, 2H). ^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.18, 153.77, 143.76, 132.52, 130.32, 125.08, 124.22, 123.53, 118.87, 110.43, 79.18, 70.91, 42.98.

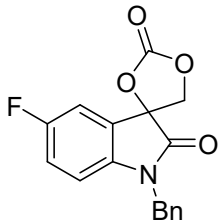
4. 1'-benzyl-5'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.38 (dd, *J* = 13.8, 7.9 Hz, 3H), 7.33 (d, *J* = 6.9 Hz, 1H), 7.30 – 7.26 (m, 3H), 7.13 (dd, *J* = 8.0, 2.0 Hz, 1H), 6.79 (d, *J* = 2.0 Hz, 1H), 4.92 (s, 1H), 4.83 (s, 1H), 4.78 (d, *J* = 8.3 Hz, 1H), 4.56 (d, *J* = 9.2 Hz, 1H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.14, 145.22, 134.11, 133.53, 132.83, 129.27, 128.47, 127.43, 124.78, 108.98, 79.02, 77.37, 77.16, 76.95, 68.57, 44.71.

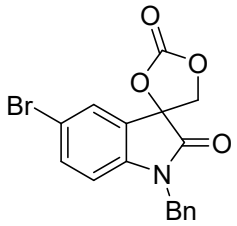
5. 1'-benzyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



Light pink solid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.38 – 7.29 (m, 4H), 7.29 – 7.25 (m, 2H), 7.22 (d, *J* = 7.0 Hz, 1H), 7.05 (t, *J* = 8.7 Hz, 1H), 6.73 (dd, *J* = 8.9, 3.6 Hz, 1H), 4.95 (d, *J* = 15.6 Hz, 1H), 4.86 – 4.78 (m, 2H), 4.57 (d, *J* = 8.0 Hz, 1H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 134.18, 129.29, 128.47, 127.46, 119.11, 118.95, 113.33, 113.17, 111.57, 111.52, 77.37, 77.16, 76.95, 70.82, 44.62.

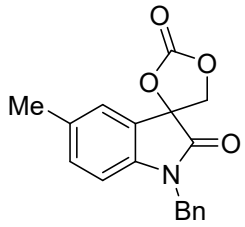
6. 1'-benzyl-5'-bromospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



Pale yellow solid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.46 (d, *J* = 7.6 Hz, 1H), 7.35 (t, *J* = 7.8 Hz, 3H), 7.29 (d, *J* = 7.5 Hz, 3H), 7.15 (t, *J* = 7.5 Hz, 1H), 6.80 (d, *J* = 7.5 Hz, 1H), 4.95 (d, *J* = 15.3 Hz, 1H), 4.84 (d, *J* = 15.8 Hz, 1H), 4.78 (d, *J* = 8.6 Hz, 1H), 4.58 (d, *J* = 8.9 Hz, 1H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.62, 153.75, 143.64, 134.50, 132.49, 129.17, 128.28, 127.45, 125.08, 124.24, 123.47, 110.51, 79.24, 70.89, 44.40.

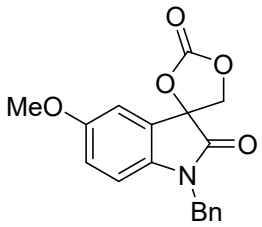
7. 1'-benzyl-5'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; Melting point 198-200 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.33 (t, *J* = 7.1 Hz, 2H), 7.30 (d, *J* = 5.2 Hz, 2H), 7.28 (d, *J* = 4.7 Hz, 3H), 7.13 (d, *J* = 8.2 Hz, 1H), 6.64 (dd, *J* = 32.2, 8.0 Hz, 1H), 4.93 (d, *J* = 15.1 Hz, 1H), 4.86 – 4.73 (m, 2H), 4.57 (d, *J* = 9.2 Hz, 1H), 2.32 (s, 3H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 132.74, 129.16, 128.96, 128.25, 127.47, 127.33, 125.75, 110.32, 77.37, 77.16, 76.95, 70.98, 44.43, 21.07.

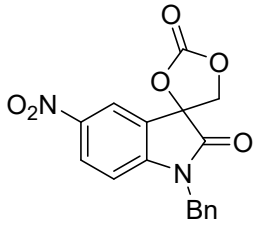
8. 1'-benzyl-5'-methoxyspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; Melting point 152-154 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.37 – 7.32 (m, 2H), 7.32 – 7.26 (m, 4H), 7.04 (d, *J* = 2.4 Hz, 1H), 6.85 (dd, *J* = 8.6, 2.7 Hz, 1H), 6.68 (d, *J* = 8.6 Hz, 1H), 4.93 (d, *J* = 15.5 Hz, 1H), 4.84 – 4.76 (m, 2H), 4.57 (d, *J* = 9.1 Hz, 1H), 3.78 (s, 3H).

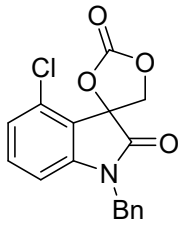
^{13}C NMR (151 MHz, Chloroform-*d*) δ 134.57, 129.14, 128.24, 127.43, 117.15, 111.68, 111.23, 71.00, 56.05, 44.47.

9. 1'-benzyl-5'-nitrospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



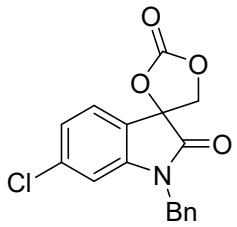
Light yellow solid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.46 (d, *J* = 7.6 Hz, 1H), 7.35 (t, *J* = 7.8 Hz, 3H), 7.29 (d, *J* = 7.5 Hz, 3H), 7.15 (t, *J* = 7.5 Hz, 1H), 6.80 (d, *J* = 7.5 Hz, 1H), 4.95 (d, *J* = 15.3 Hz, 1H), 4.84 (d, *J* = 15.8 Hz, 1H), 4.78 (d, *J* = 8.6 Hz, 1H), 4.58 (d, *J* = 8.9 Hz, 1H).
^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.62, 153.75, 143.64, 134.50, 132.49, 129.17, 128.28, 127.45, 125.08, 124.24, 123.47, 110.51, 79.24, 70.89, 44.40.

10. 1'-benzyl-4'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



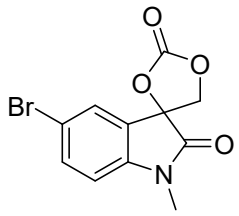
White solid. ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.37 – 7.33 (m, 2H), 7.32 – 7.27 (m, 4H), 7.08 (d, *J* = 8.8 Hz, 1H), 6.71 (d, *J* = 8.2 Hz, 1H), 4.96 (d, *J* = 15.6 Hz, 1H), 4.86 – 4.81 (m, 2H), 4.75 (d, *J* = 8.6 Hz, 1H).
^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.14, 145.22, 134.11, 133.53, 132.83, 129.27, 128.47, 127.43, 124.78, 108.98, 79.02, 77.37, 77.16, 76.95, 68.57, 44.71.

11. 1'-benzyl-6'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; Melting point 124-126 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.38 (dd, *J* = 13.8, 7.9 Hz, 3H), 7.33 (d, *J* = 6.9 Hz, 1H), 7.28 (d, *J* = 7.1 Hz, 2H), 7.13 (dd, *J* = 8.0, 2.0 Hz, 1H), 6.79 (d, *J* = 2.0 Hz, 1H), 4.93 (d, *J* = 15.5 Hz, 1H), 4.84 – 4.76 (m, 2H), 4.56 (d, *J* = 9.2 Hz, 1H).
^{13}C NMR (151 MHz, Chloroform-*d*) δ 153.48, 144.91, 138.65, 133.99, 129.36, 128.57, 127.46, 126.11, 124.30, 121.79, 111.28, 78.73, 77.37, 77.16, 76.95, 70.73, 44.60.

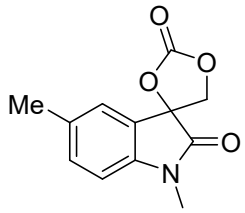
12. 1'-methyl-5'-bromospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



Pale-yellow solid. ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.48 – 7.46 (m, 2H), 7.19 (t, *J* = 7.5 Hz, 1H), 6.91 (d, *J* = 7.5 Hz, 1H), 4.72 (d, *J* = 9.1 Hz, 1H), 4.55 (d, *J* = 9.1 Hz, 1H), 3.23 (s, 3H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.36, 153.77, 144.45, 132.58, 124.94, 124.22, 123.53, 109.49, 79.21, 70.87, 26.77

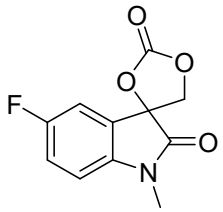
13. 1'-methyl-5'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione



White solid; Melting point 159-161 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.30 – 7.24 (m, 2H), 6.79 (d, *J* = 8.0 Hz, 1H), 4.71 (d, *J* = 9.1 Hz, 1H), 4.53 (d, *J* = 9.0 Hz, 1H), 3.21 (s, 3H), 2.37 (s, 3H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.32, 153.87, 142.00, 134.17, 132.79, 125.65, 123.57, 109.28, 79.43, 77.37, 77.16, 76.95, 70.95, 26.81, 21.10.

14. 1'-methyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione

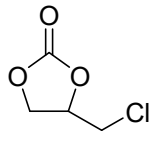


Pale pink solid; Melting point 155-157 ^{o}C; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.49 – 7.45 (m, 2H), 7.19 (t, *J* = 7.5 Hz, 1H), 6.93 (d, *J* = 7.5 Hz, 1H), 4.72 (d, *J* = 9.1 Hz, 1H), 4.55 (d, *J* = 9.1 Hz, 1H), 3.23 (s, 3H).

^{13}C NMR (151 MHz, Chloroform-*d*) δ 171.36, 153.77, 144.45, 132.58, 124.94, 124.22, 123.53, 109.49, 79.21, 70.87, 26.77

Characterization of cyclic carbonate products{Subramanian, 2018 #1408}

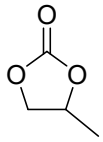
15. 4-(chloromethyl)-1,3-dioxolan-2-one



Light yellow liduid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 5.05 – 5.01 (m, 1H), 4.62 (t, J = 8.6 Hz, 1H), 4.42 (dd, J = 9.0, 5.7 Hz, 1H), 3.83 (dd, J = 12.6, 5.7 Hz, 1H), 3.74 (dd, J = 12.5, 3.5 Hz, 1H).

^{13}C NMR (151 MHz, CDCl3) δ 154.29, 74.30, 66.83, 43.85.

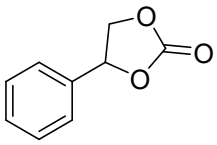
16. Propylene carbonate



Tranparent water like liquid; ^{1}H NMR (600 MHz, Chloroform-*d*): δ = 4.92 – 4.89 (m, 1H), 4.61 – 4.58 (m, 1H), 4.06 (dd, J = 8.5, 7.2 Hz, 1H), 1.47 (d, J = 6.4 Hz, 3H).

^{13}C NMR (151 MHz, CDCl3): δ = 154.70, 73.30, 70.19, 18.48.

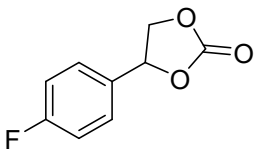
17. Styrene carbonate



Dark red liquid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.47 – 7.35 (m, 5H), 5.67 (t, J = 8.0 Hz, 1H), 4.80 (t, J = 8.1 Hz, 1H), 4.33 (t, J = 8.5 Hz, 1H).

^{13}C NMR (151 MHz, CDCl3) δ 154.80, 135.71, 129.62, 129.12, 125.80, 71.09.

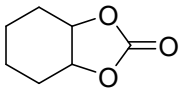
18. 2-(4-Fluorophenyl)oxirane



Yellowish liquid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 7.39 – 7.34 (m, 2H), 7.14 (t, *J* = 8.6 Hz, 2H), 5.67 (t, *J* = 8.0 Hz, 1H), 4.80 (t, *J* = 8.5 Hz, 1H), 4.35 – 4.30 (m, 1H).

^{13}C NMR (600 MHz, Chloroform-*d*) δ 162.65, 154.71, 131.72, 128.14, 128.09, 116.51, 116.36, 71.18.

19. Hexahydrobenzo[d][1,3]dioxol-2-one



Pale yellow liquid; ^{1}H NMR (600 MHz, Chloroform-*d*) δ 4.72 (s, 2H), 1.88 (s, 4H), 1.59 (s, 2H), 1.42 (s, 2H).

^{13}C NMR (151 MHz, CDCl3) δ 155.50, 75.99, 26.36, 19.06.

13. ^{1}H and ^{13}C NMR spectra of Catalyst[1]

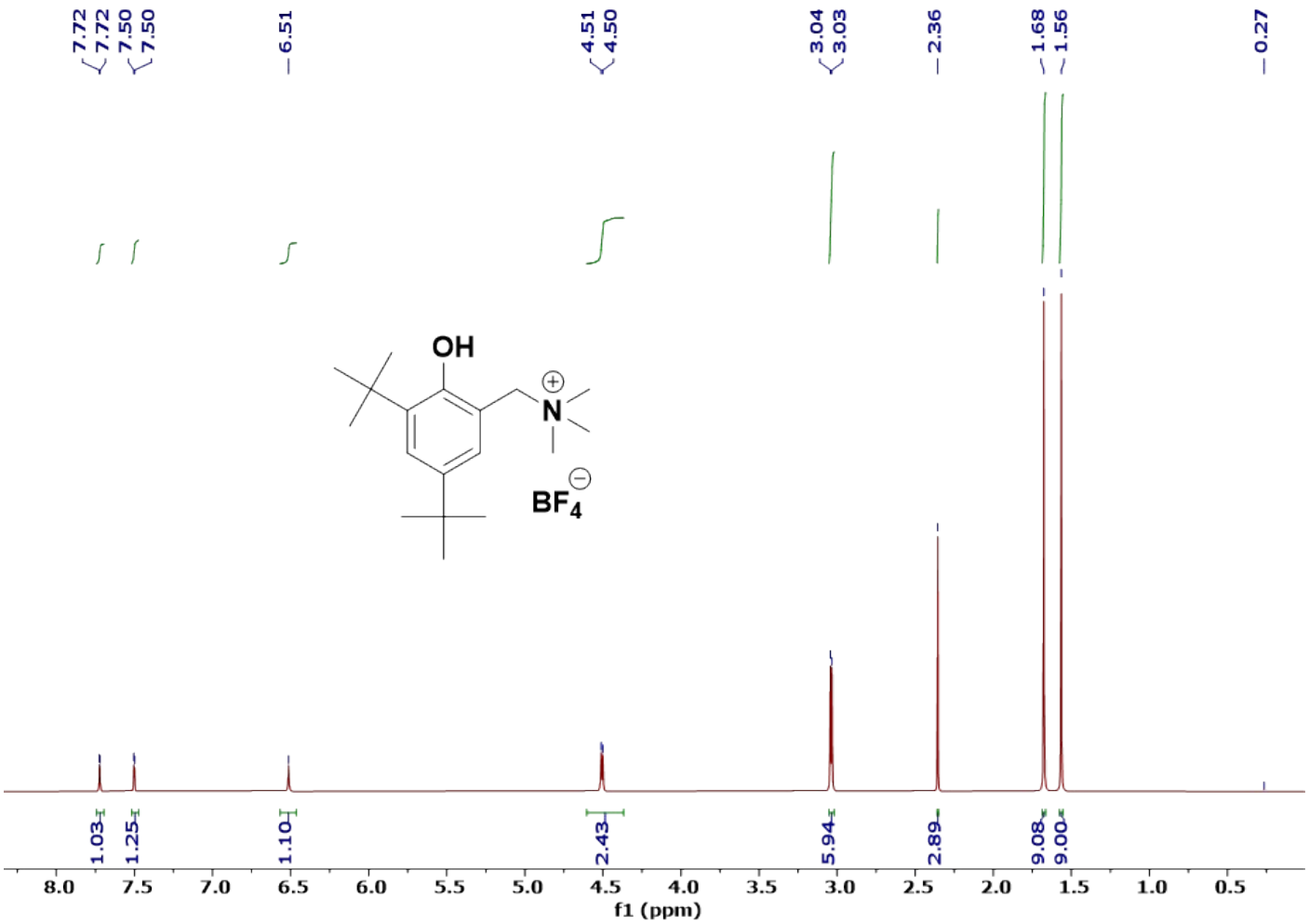


Fig. S9 ^{1}H NMR Spectrum of PDC-1 (CD_3CN, 500 MHz, 298K)

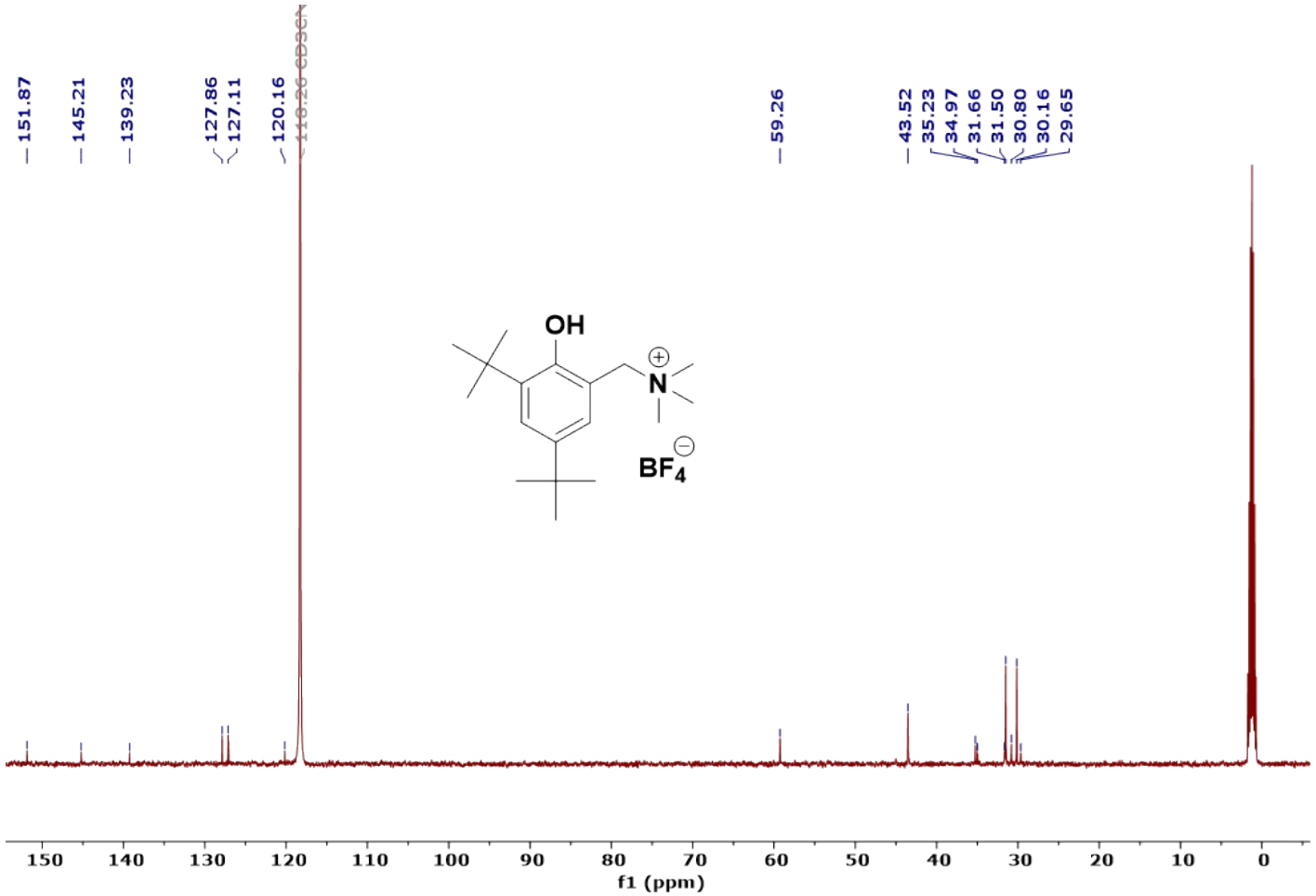


Fig. S10 ^{13}C NMR Spectrum of PDC-1 (CD_3CN, 126 MHz, 298K)

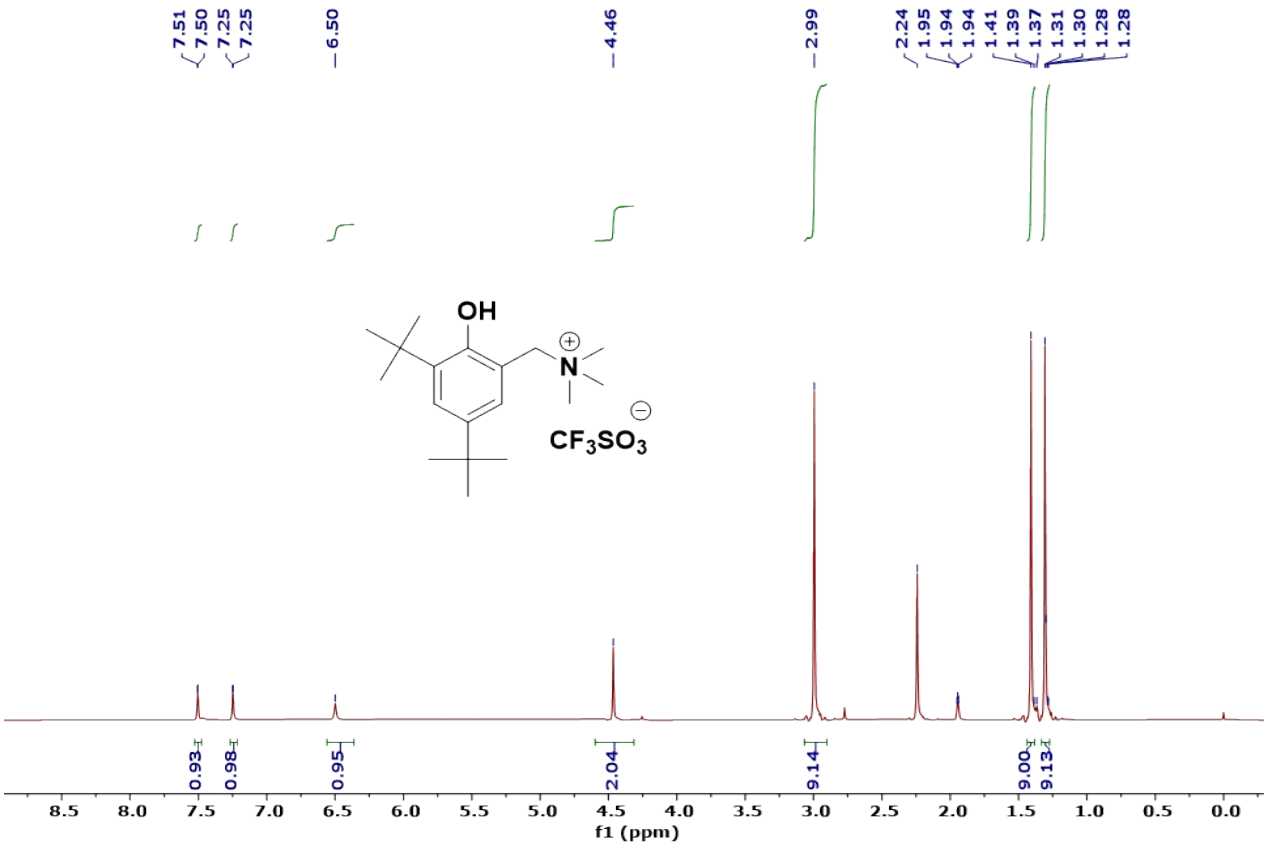


Fig. S11 ^{1}H NMR Spectrum of PDC-2 (CD_3CN, 500 MHz, 298K)

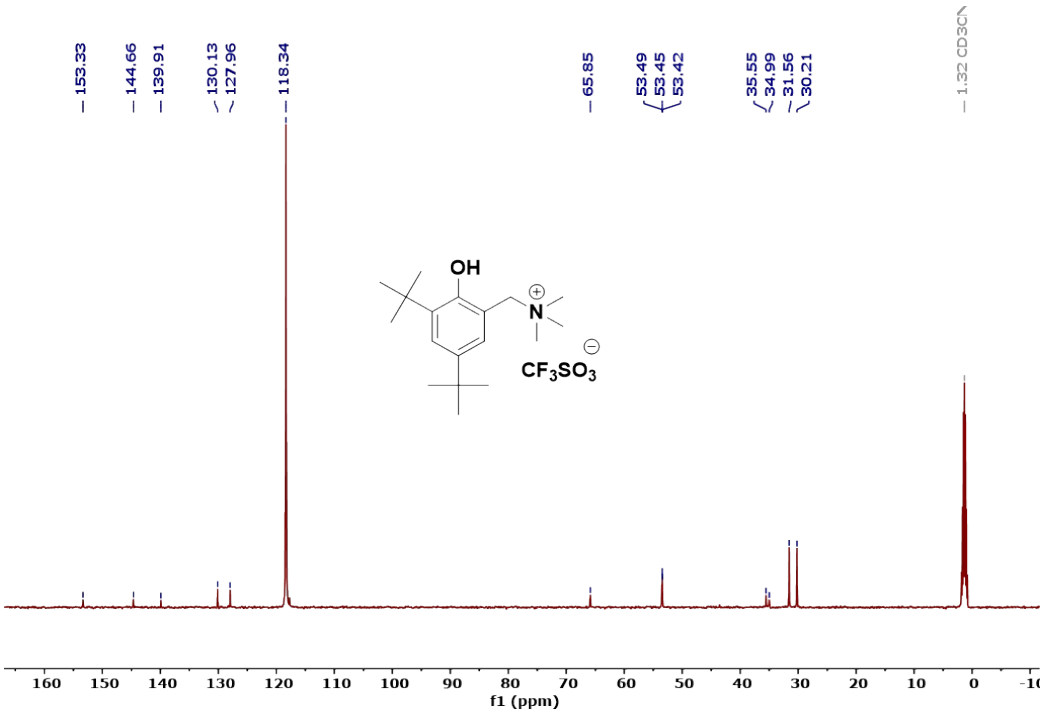


Fig. S12 ^{13}C NMR Spectrum of PDC-2 (CD_3CN, 126 MHz, 298K)

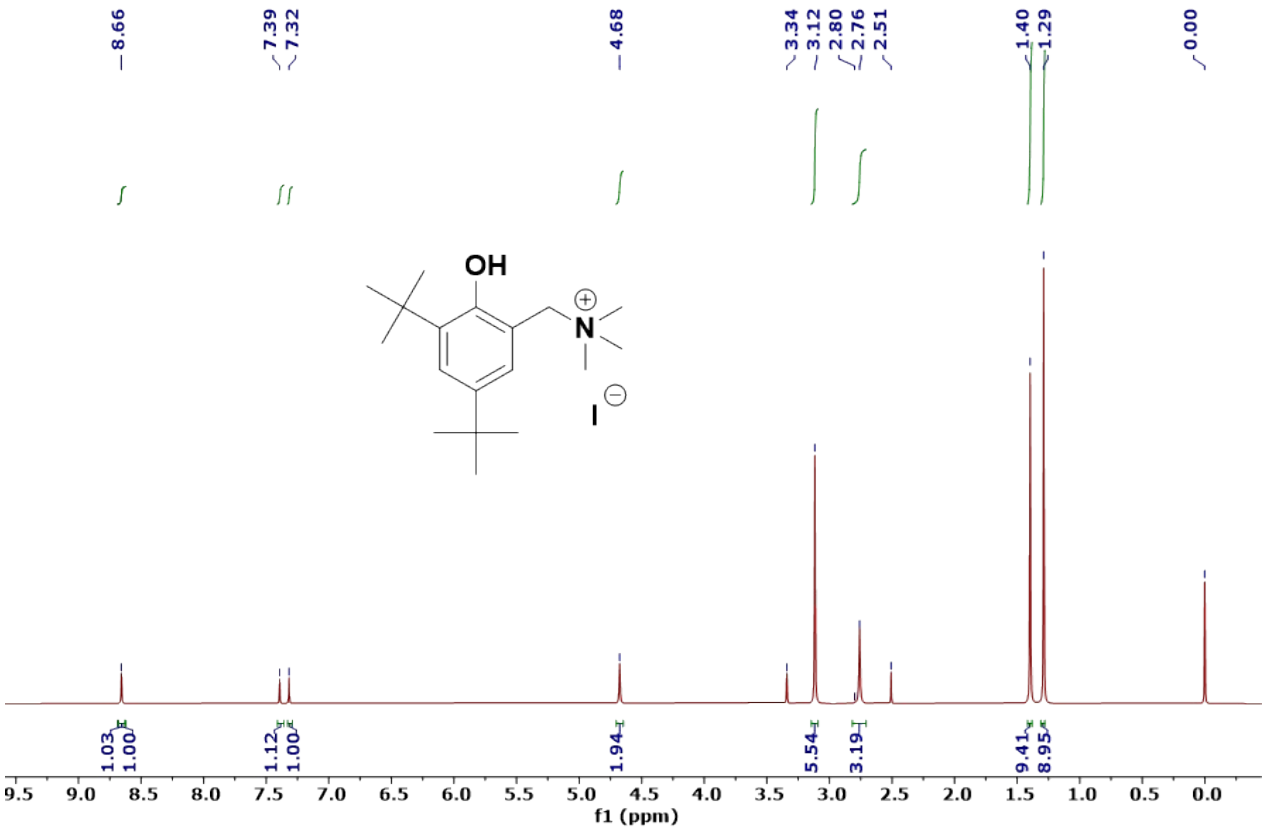


Fig. S13 ^{1}H NMR Spectrum of PDC-3 (DMSO, 500 MHz, 298K)

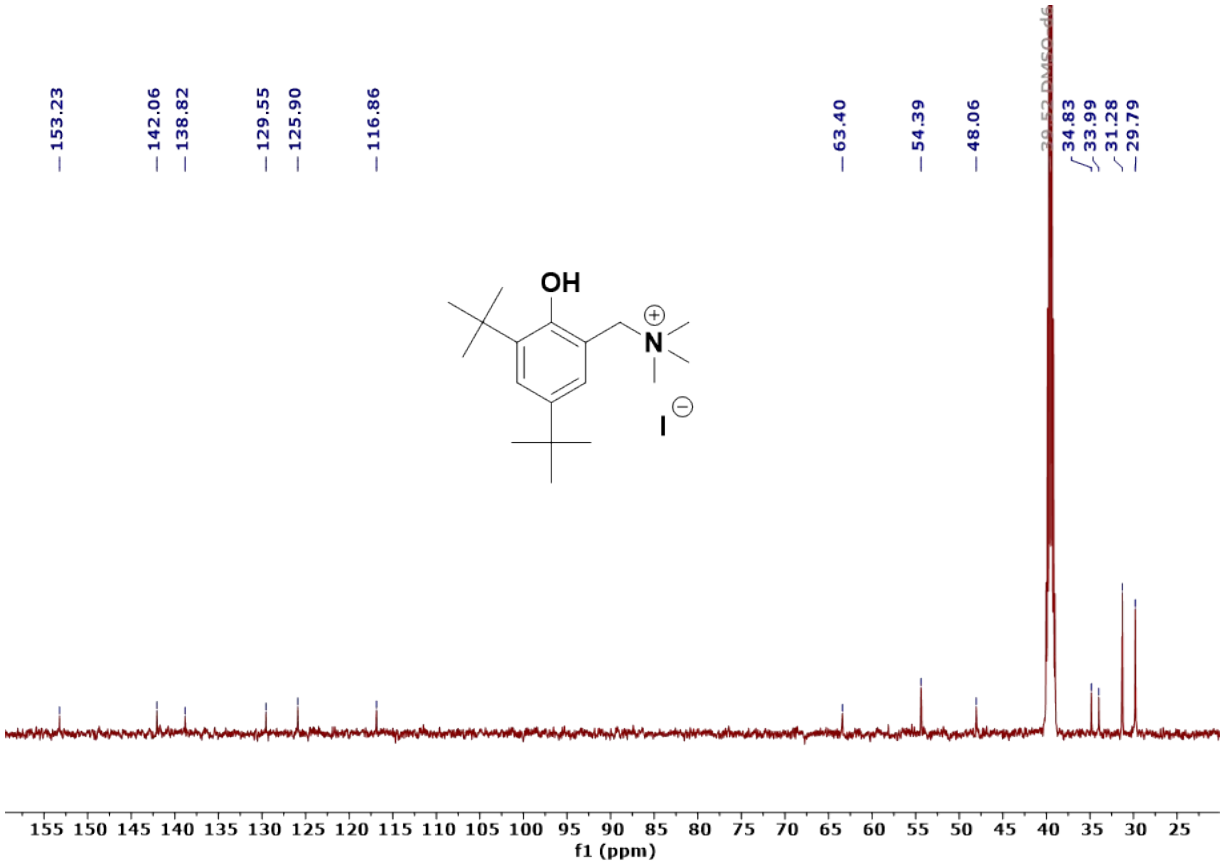


Fig. S14 ^{13}C NMR Spectrum of PDC-3 (DMSO, 126 MHz, 298K)

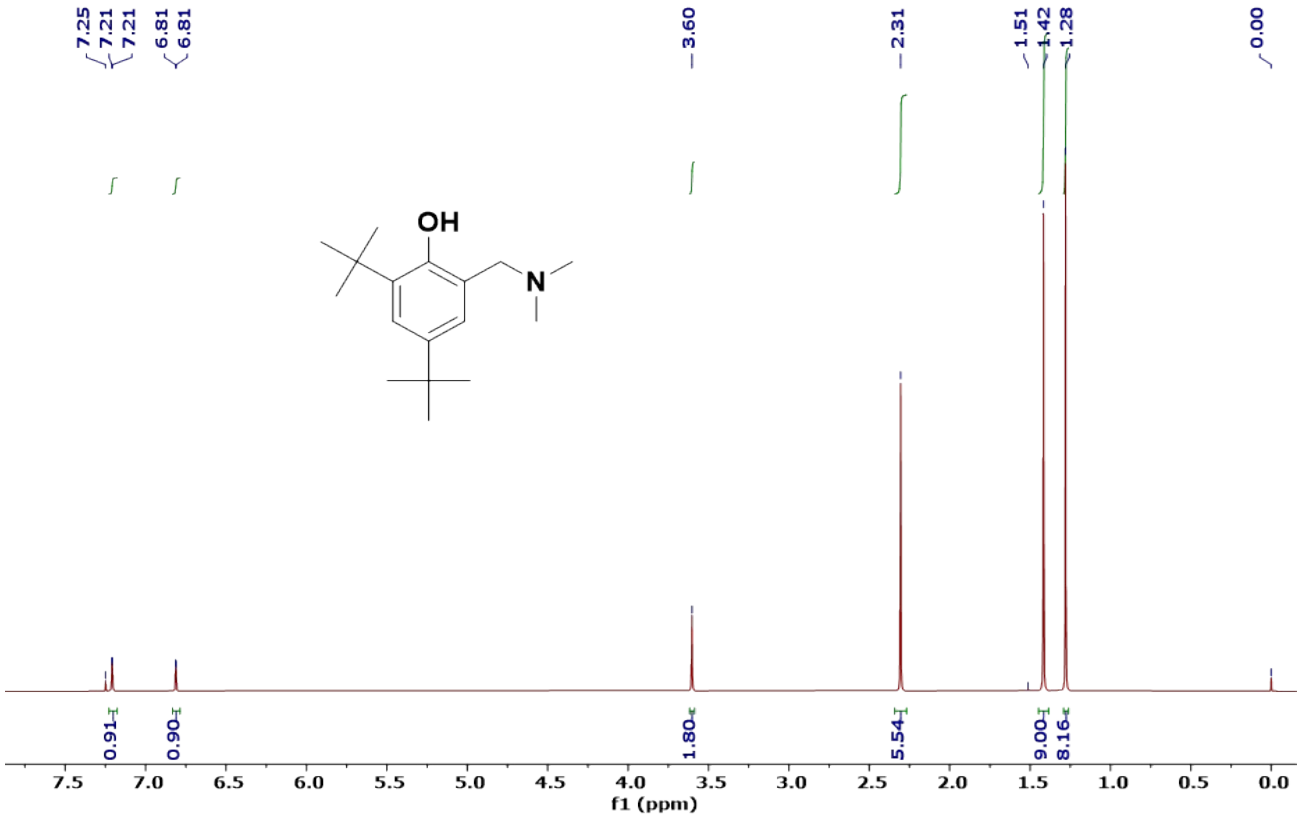


Fig. S15 ^{1}H NMR Spectrum of DMAMP moiety in PDC-4 (CD3Cl, 126 MHz, 298K)

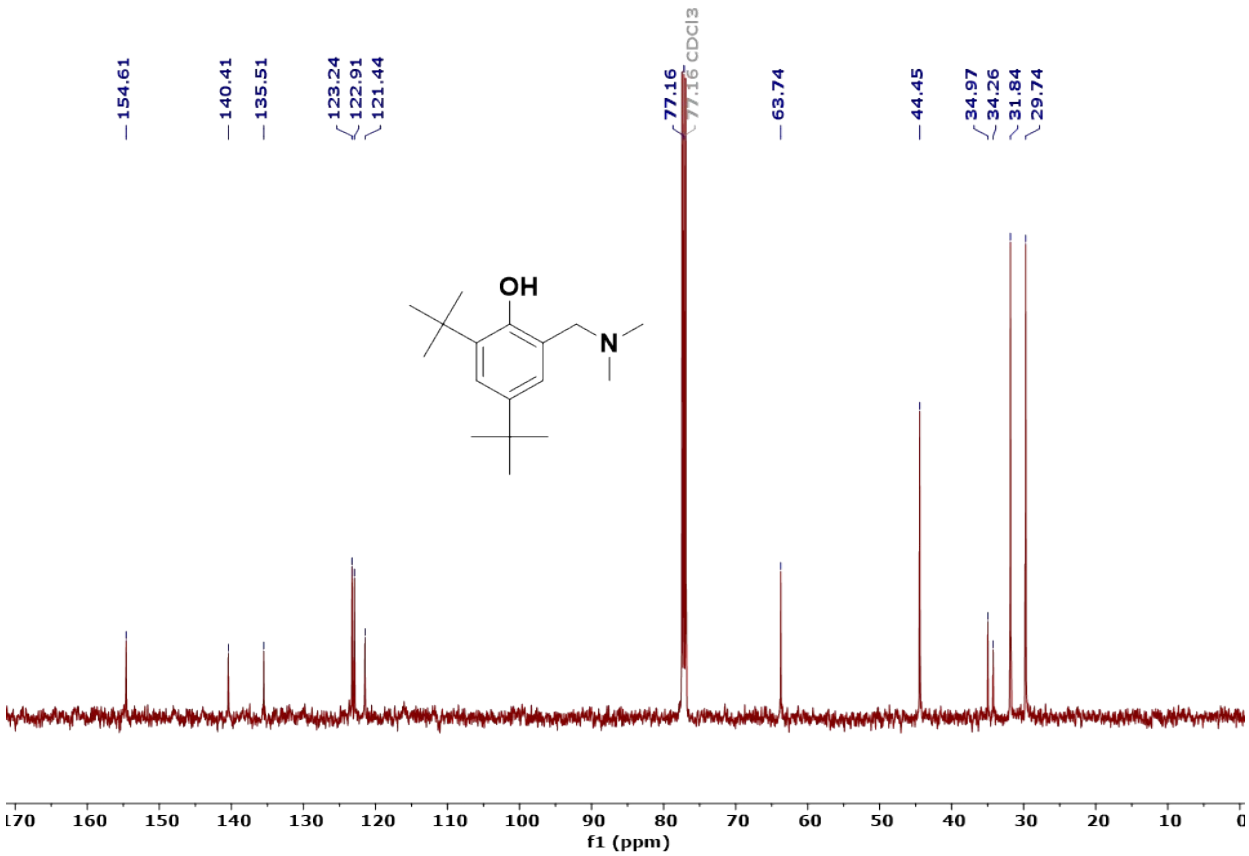


Fig. S16 ^{13}C NMR Spectrum of DMAMP moiety in PDC-4 (CD3Cl, 126 MHz, 298K)

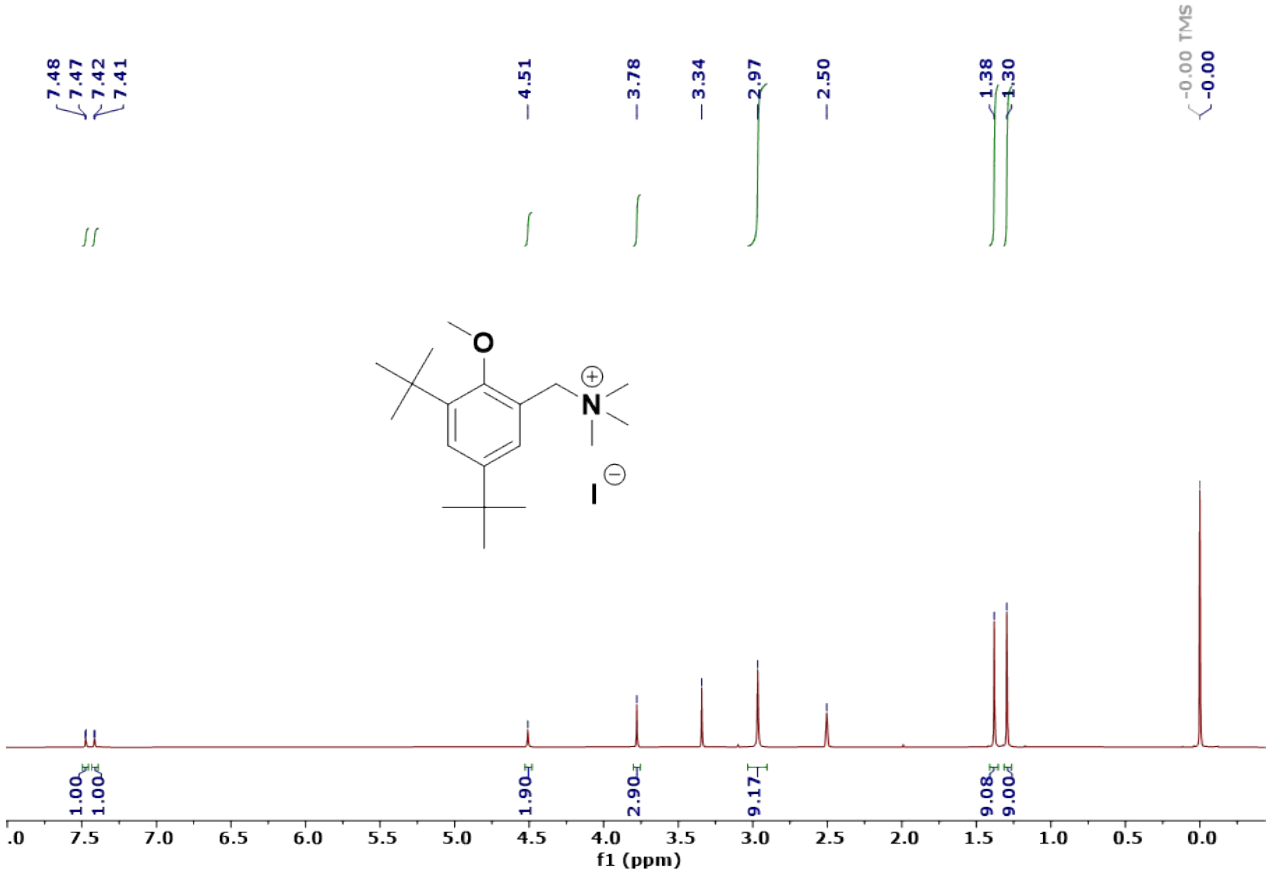


Fig. S17 ^{1}H NMR Spectrum PDC-5 (DMSO, 500 MHz, 298K)

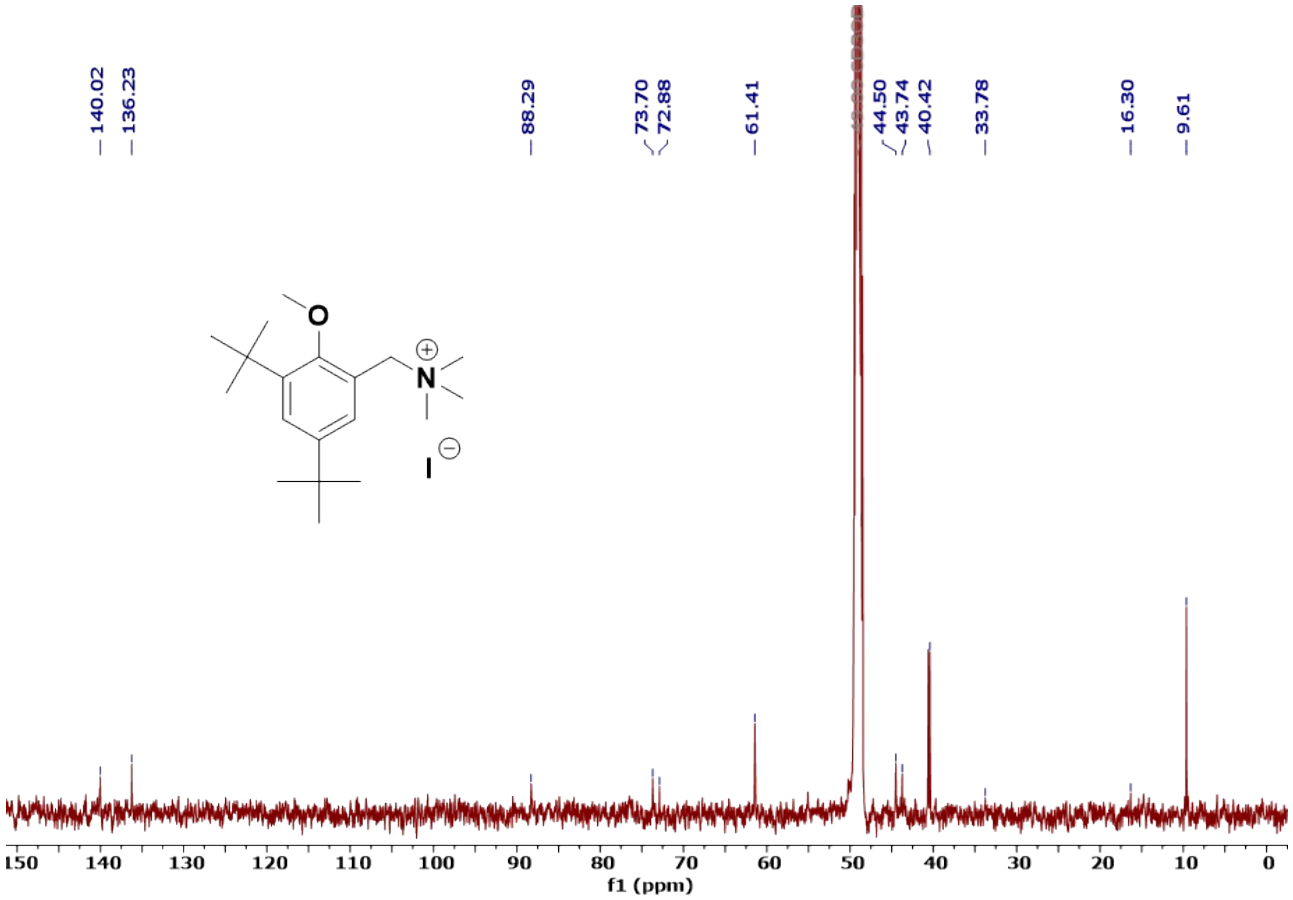


Fig. S18 ^{13}C NMR Spectrum of PDC-5 (DMSO, 126 MHz, 298K)

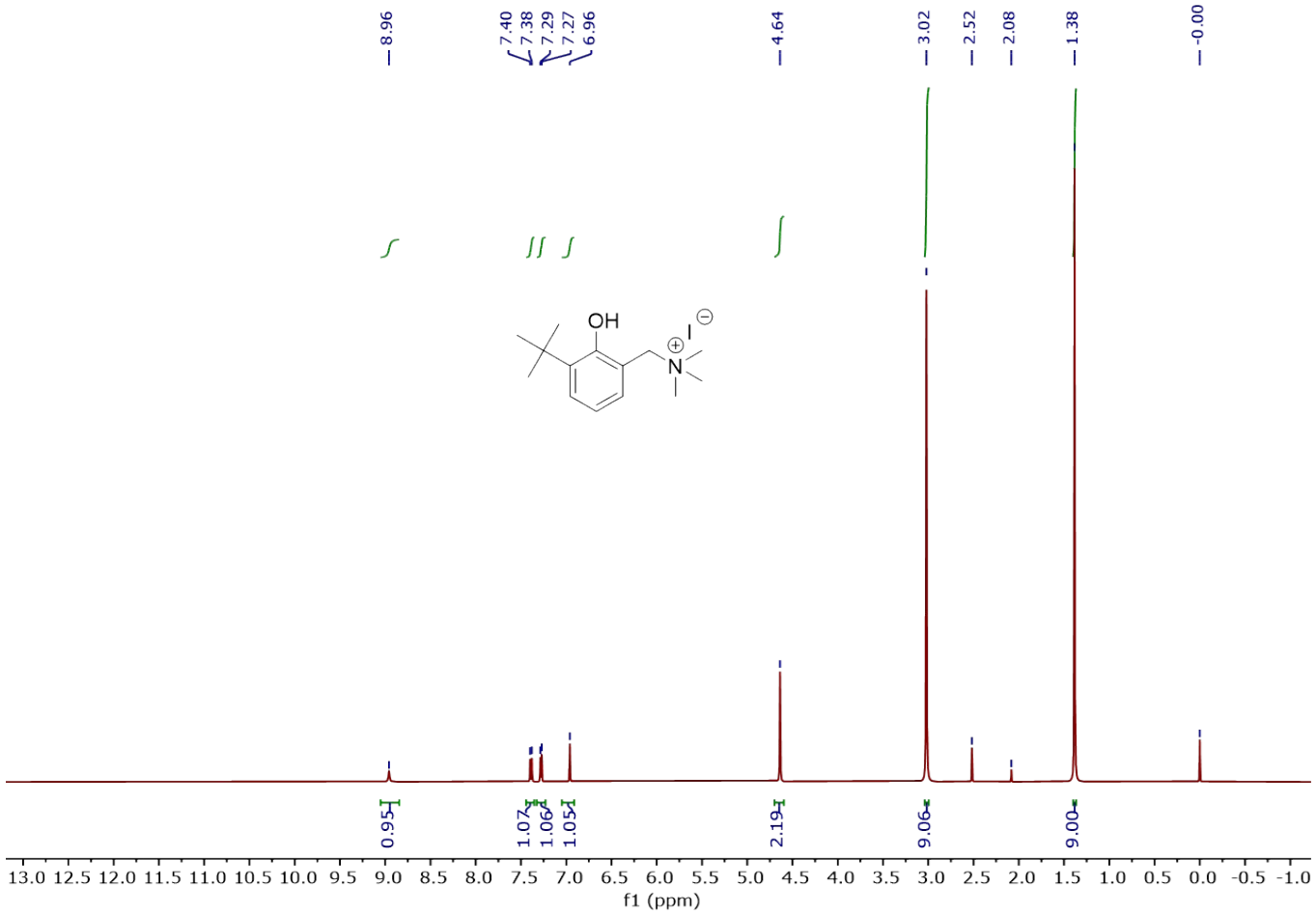


Fig. S19 ^{1}H NMR Spectrum PDC-6 (DMSO, 500 MHz, 298K)

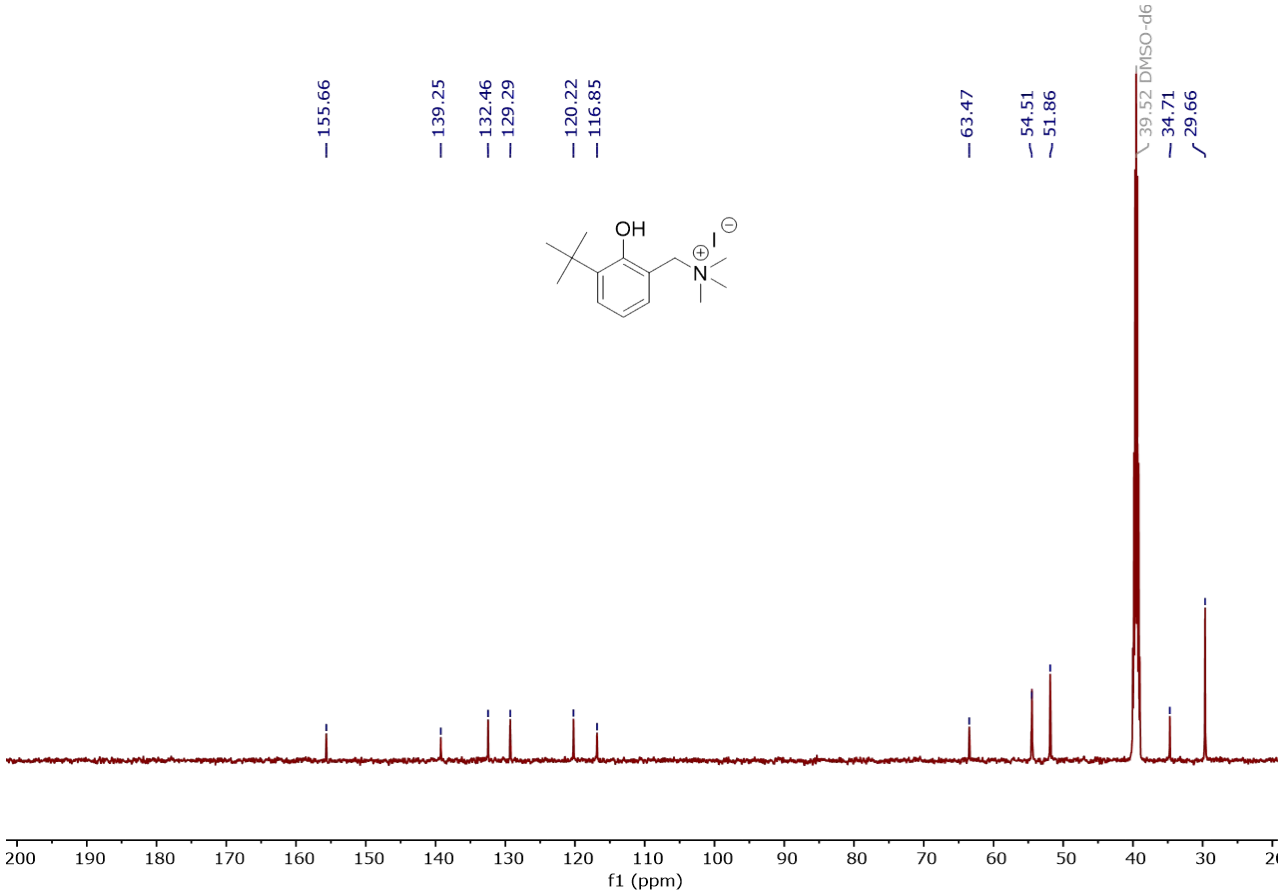


Fig. S20 ^{13}C NMR Spectrum of PDC-6 (DMSO, 126 MHz, 298K)

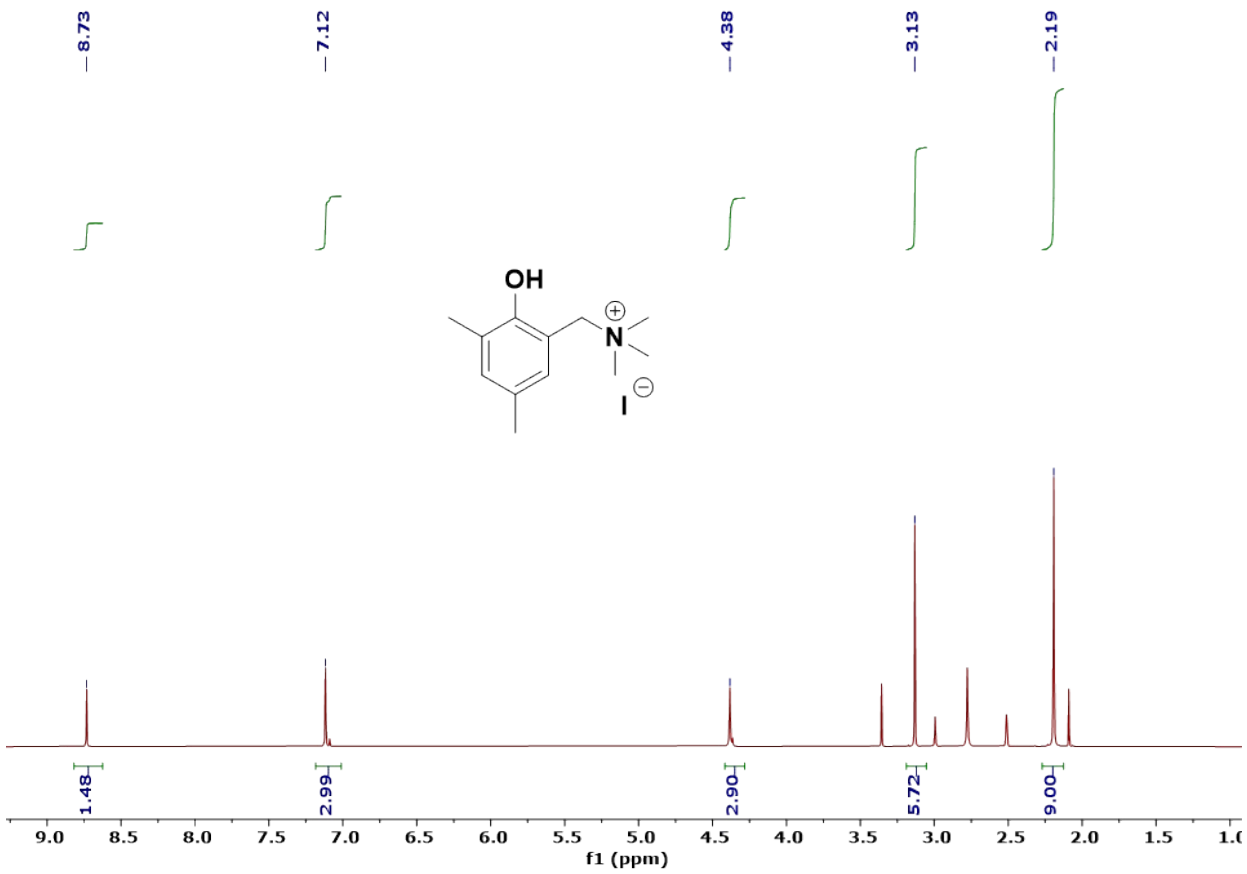


Fig. S21 ^{1}H NMR Spectrum PDC-7 (DMSO, 500 MHz, 298K)

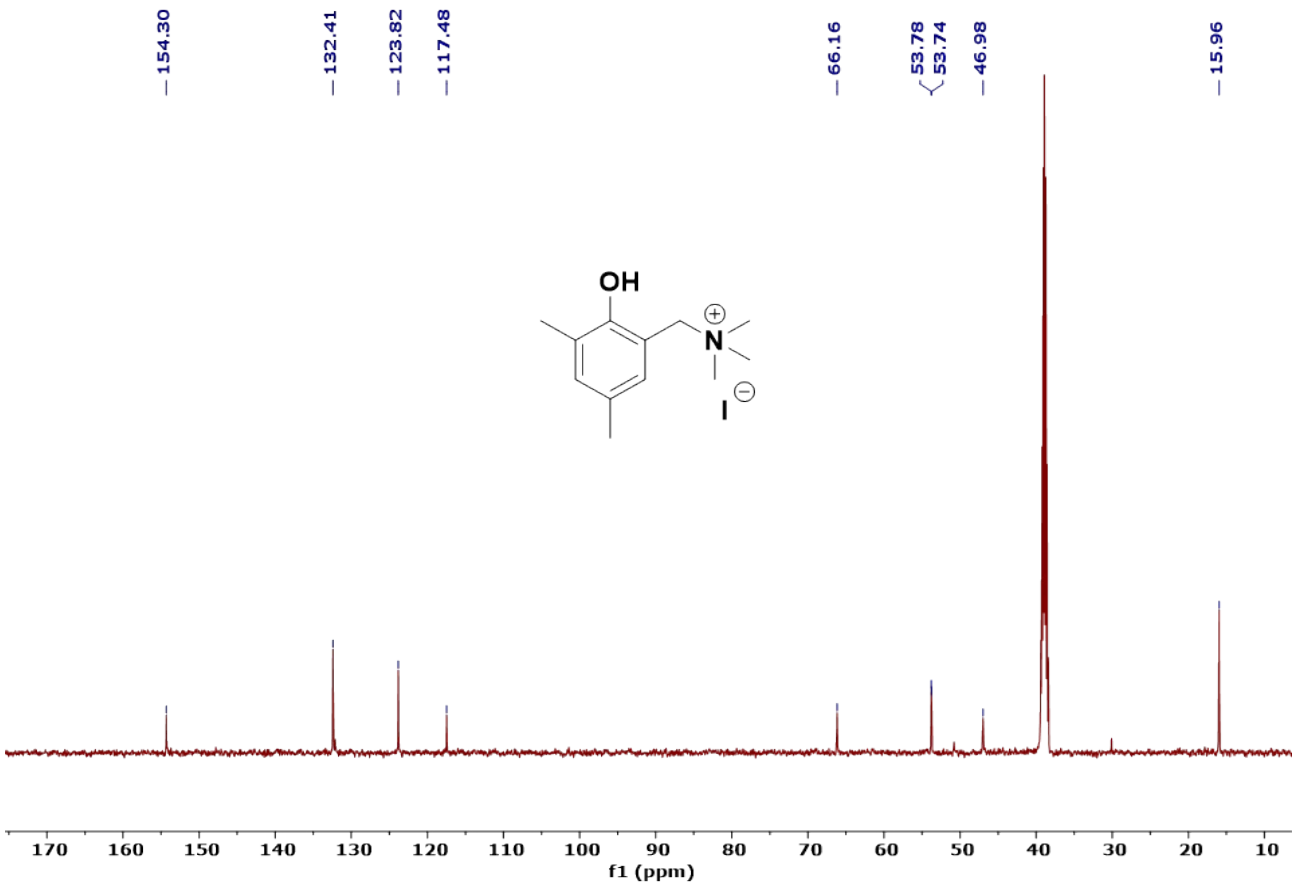


Fig. S22 ^{13}C NMR Spectrum of PDC-7 (DMSO, 126 MHz, 298K)

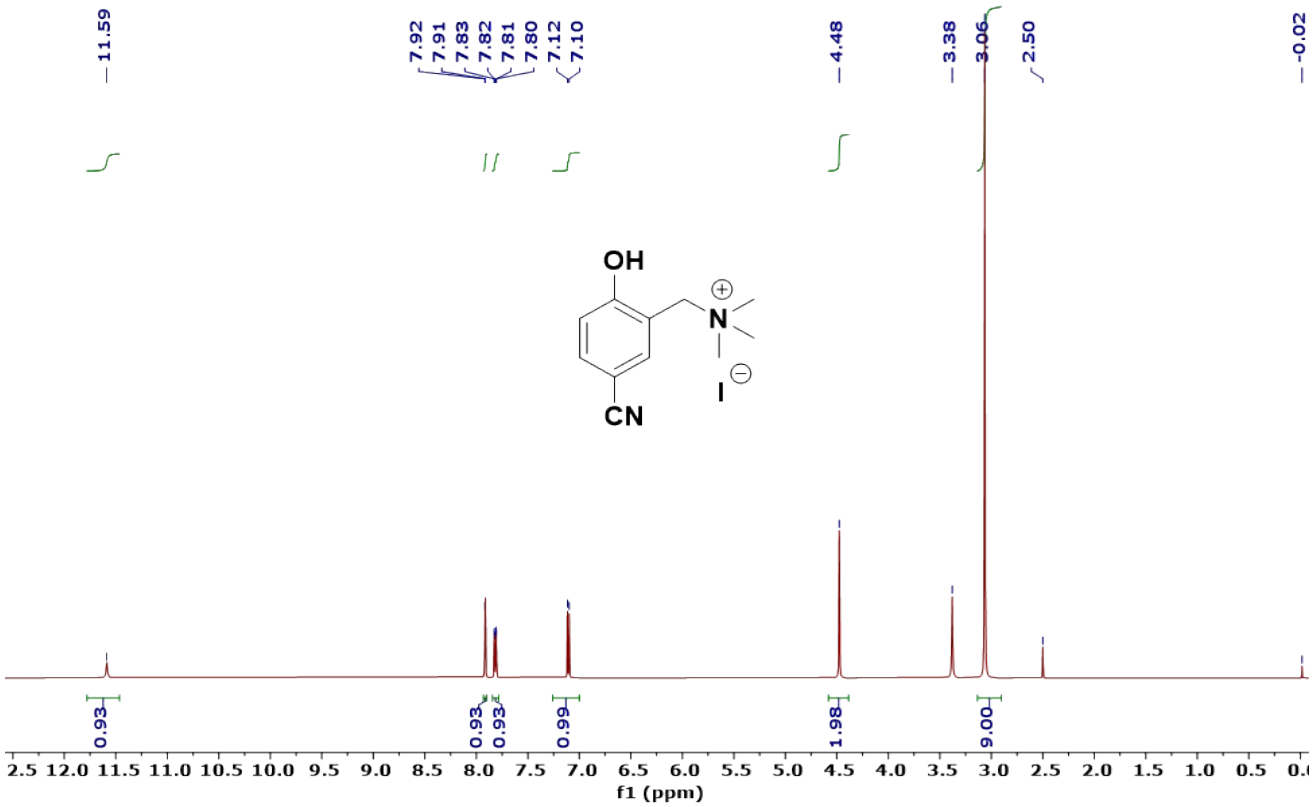


Fig. S23 ^{1}H NMR Spectrum PDC-8 (DMSO, 500 MHz, 298K)

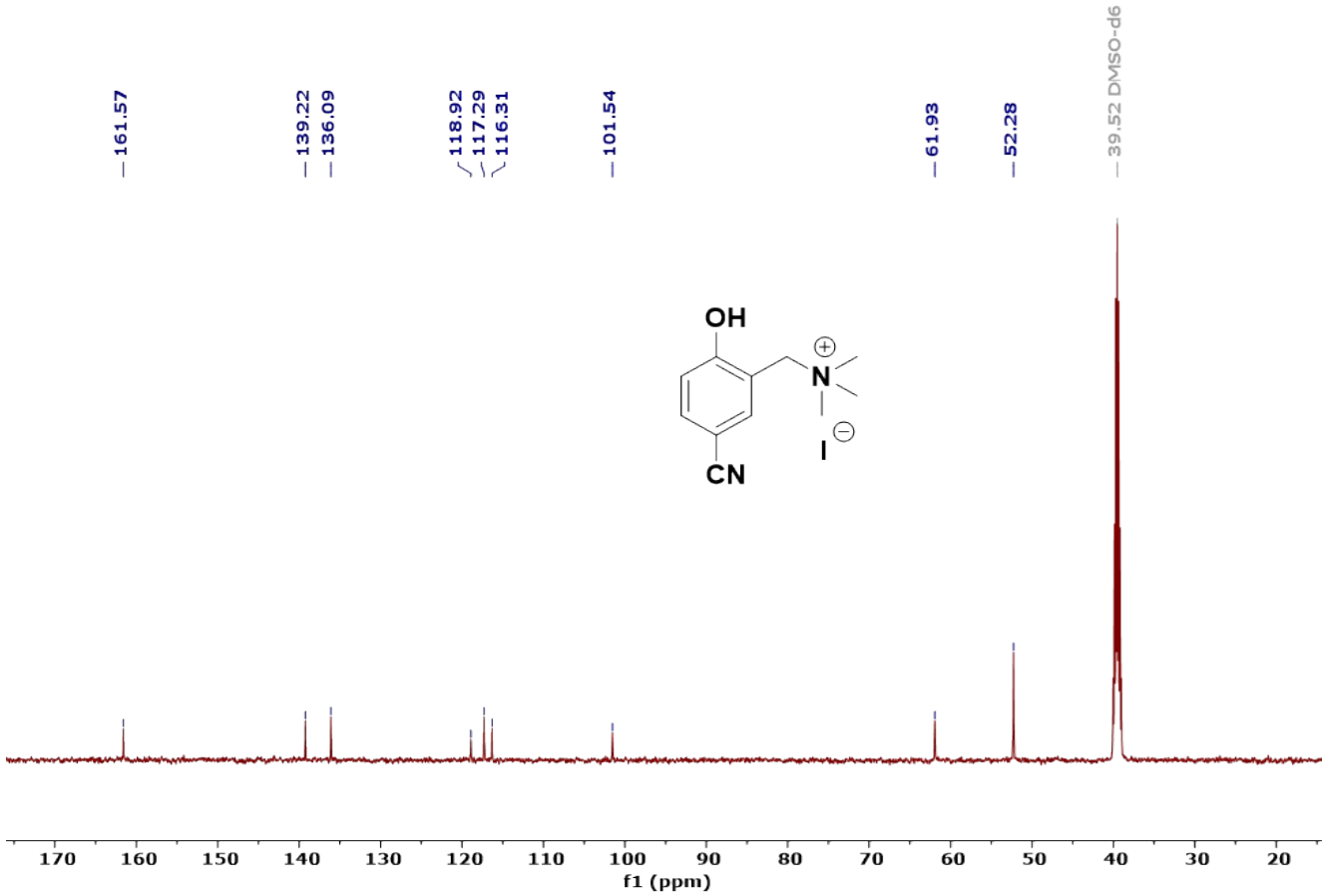


Fig. S24 ^{13}C NMR Spectrum of PDC-8 (DMSO, 126 MHz, 298K)

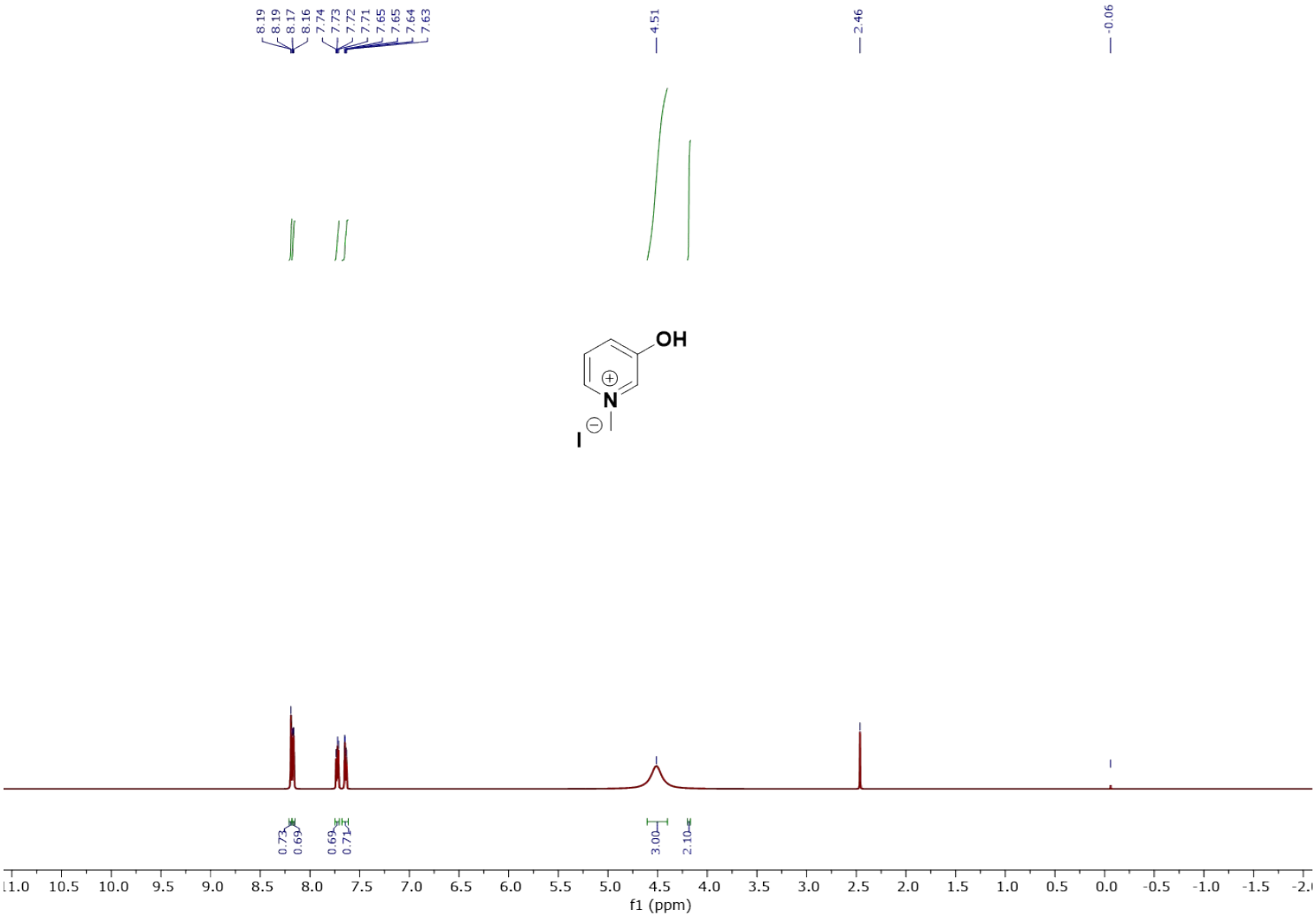


Fig. S25 ^{1}H NMR Spectrum PDC-9 (DMSO, 600 MHz, 298K)

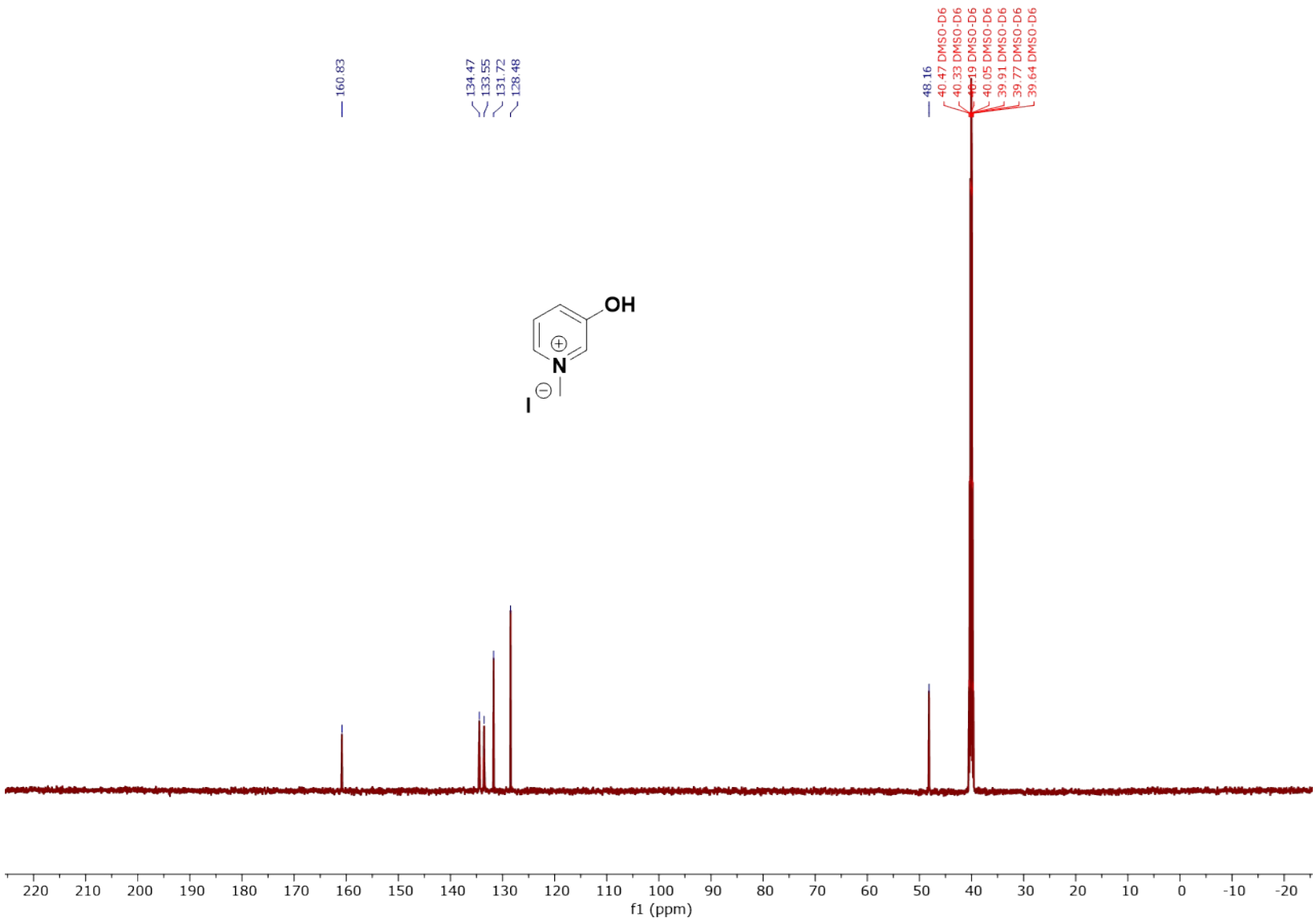


Fig. S26 ^{13}C NMR Spectrum of PDC-9 (DMSO, 151 MHz, 298K)

14. ^{1}H and ^{13}C NMR spectra of chiral *N*-Me-Q-I catalyst[5]

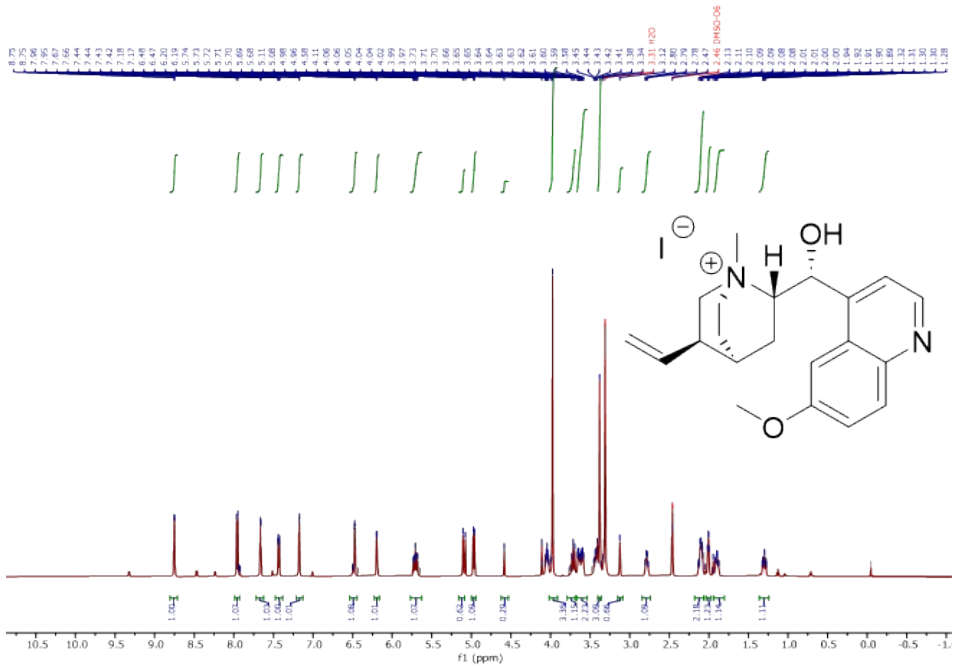


Fig. S27 ^{1}H NMR Spectrum of N-methyl quininium iodide (DMSO-d6, 600 MHz, 298K)

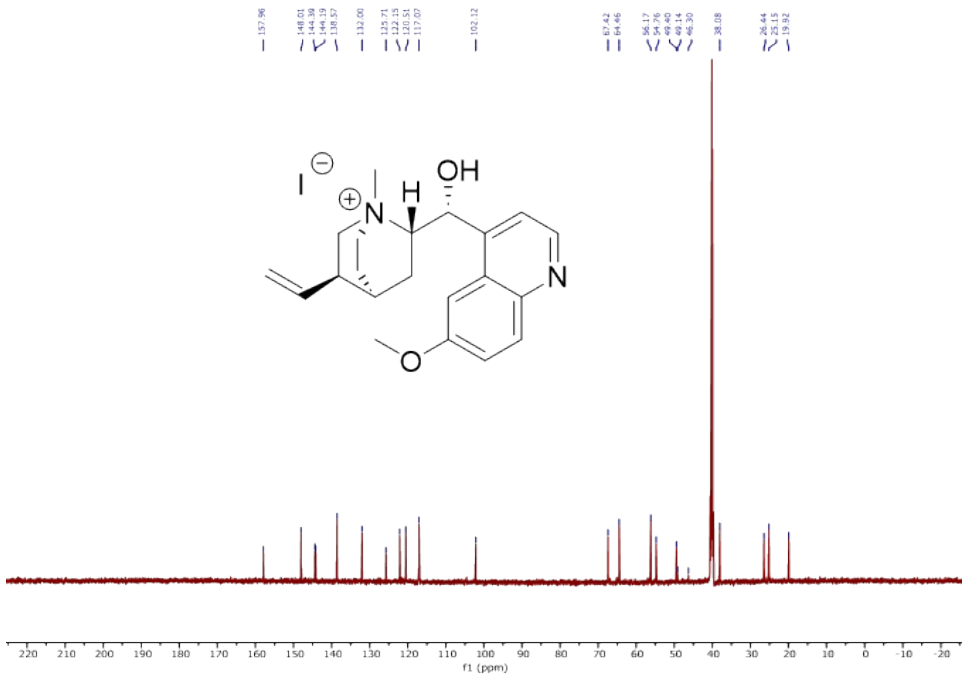


Fig. S28 ^{13}C NMR Spectrum of N-methyl quininium iodide (DMSO-d6, 151 MHz, 298K)

15. ^{1}H and ^{13}C NMR spectra of spiro-epoxyoxindole

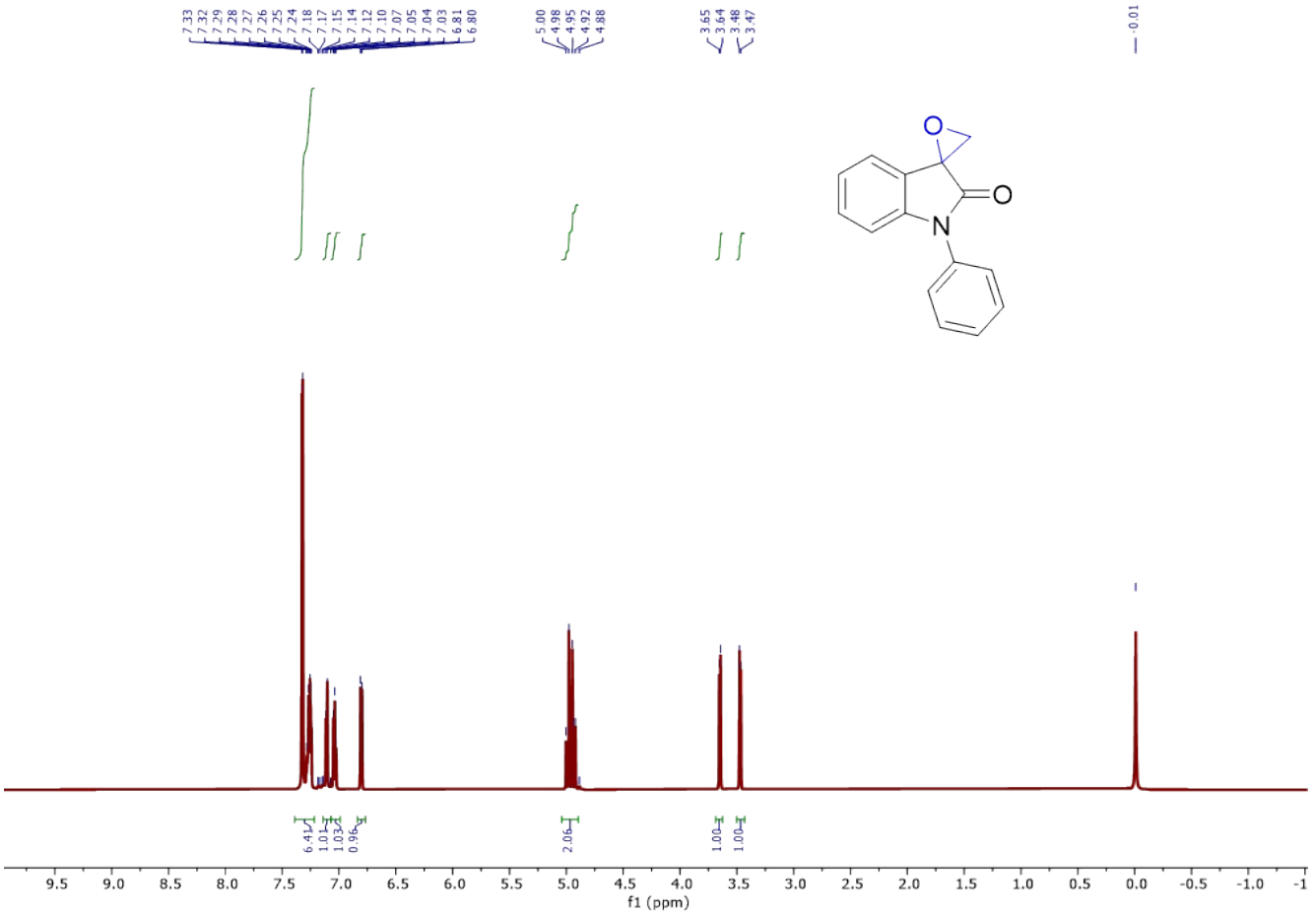


Fig. S29 ^{1}H NMR Spectrum of 1-phenylspiro[indoline-3,2'-oxiran]-2-one ($CDCl_3$, 600 MHz, 298K)

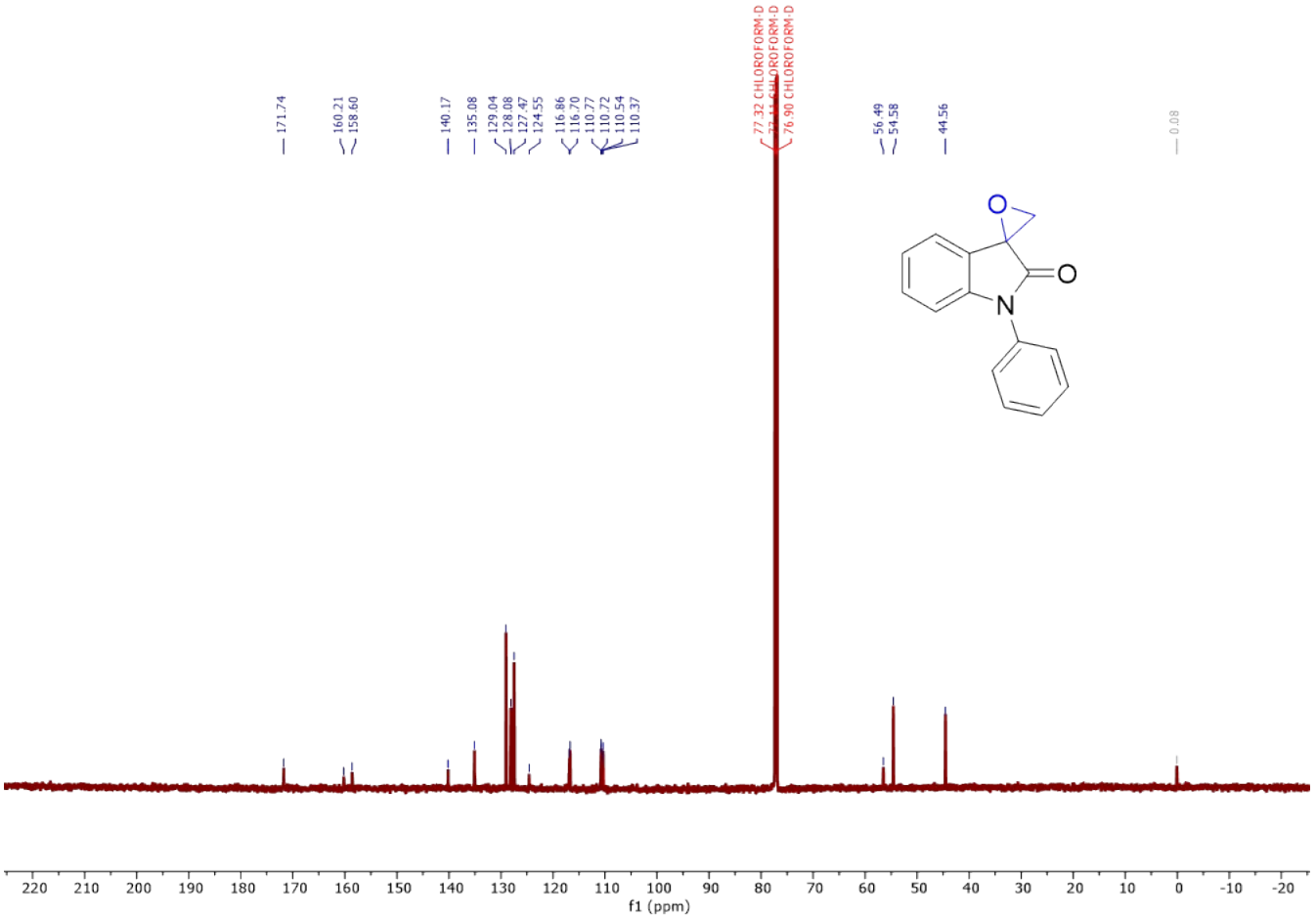


Fig. S30 ^{13}C NMR Spectrum of 1-phenylspiro[indoline-3,2'-oxiran]-2-one ($CDCl_3$, 151 MHz, 298K)

16. ^{1}H and ^{13}C spectra NMR of Product

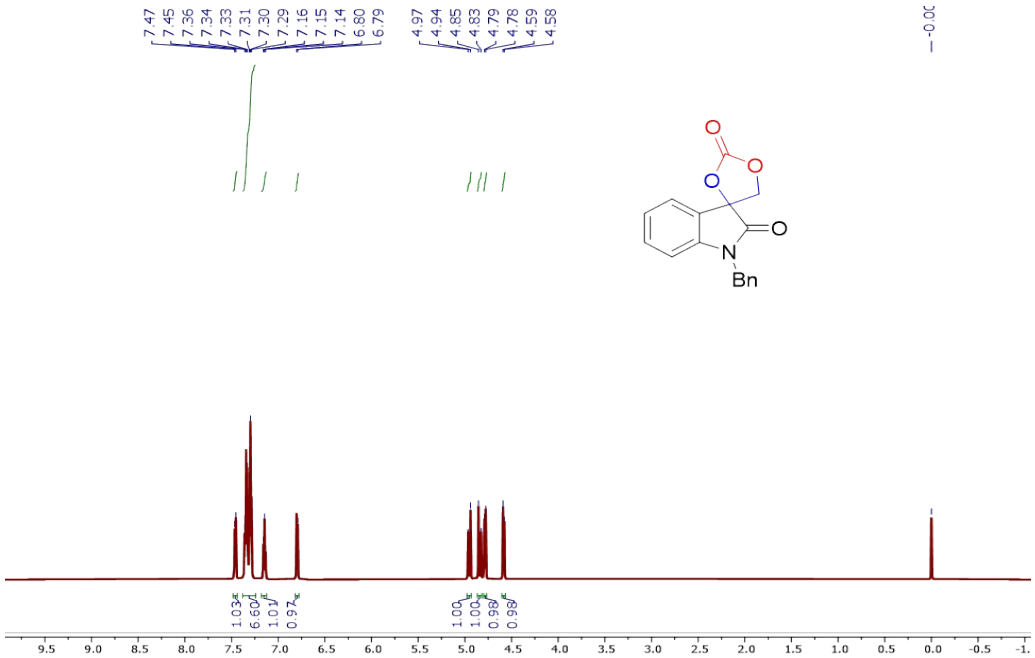


Fig. S31 ^{1}H NMR Spectrum of 1'-benzylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

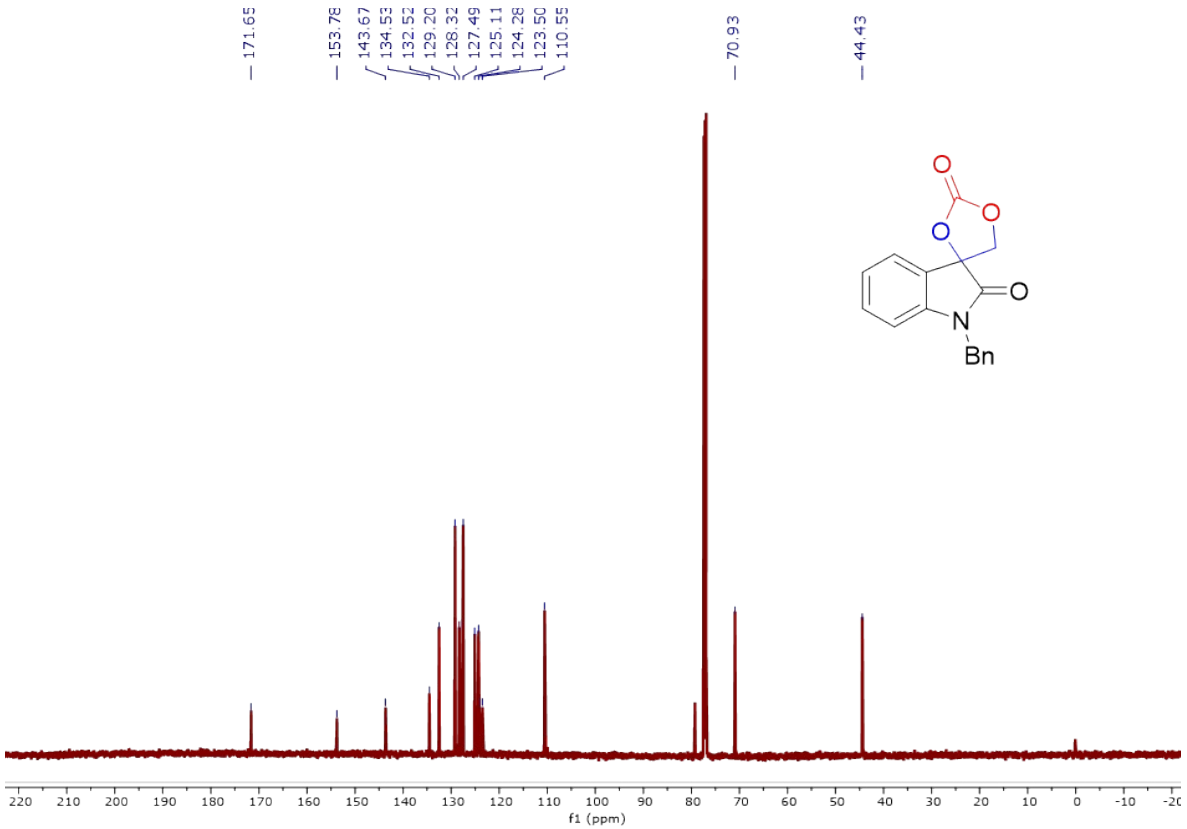


Fig. S32 ^{13}C NMR Spectrum of 1'-benzylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

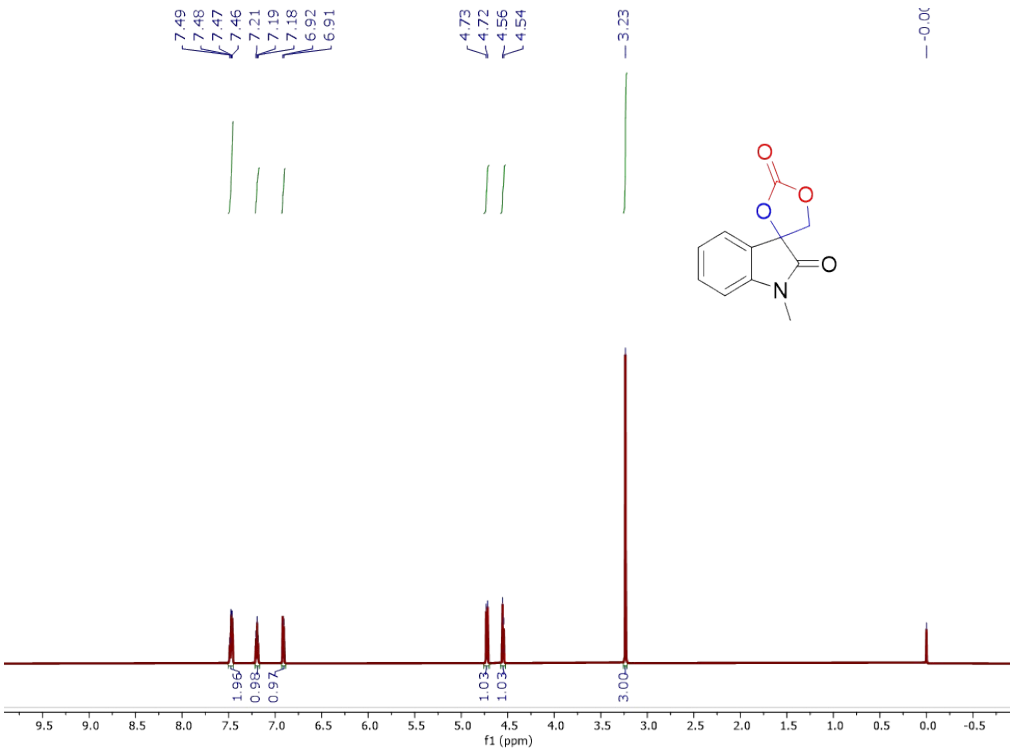


Fig. S33 ^{1}H NMR Spectrum of 1'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

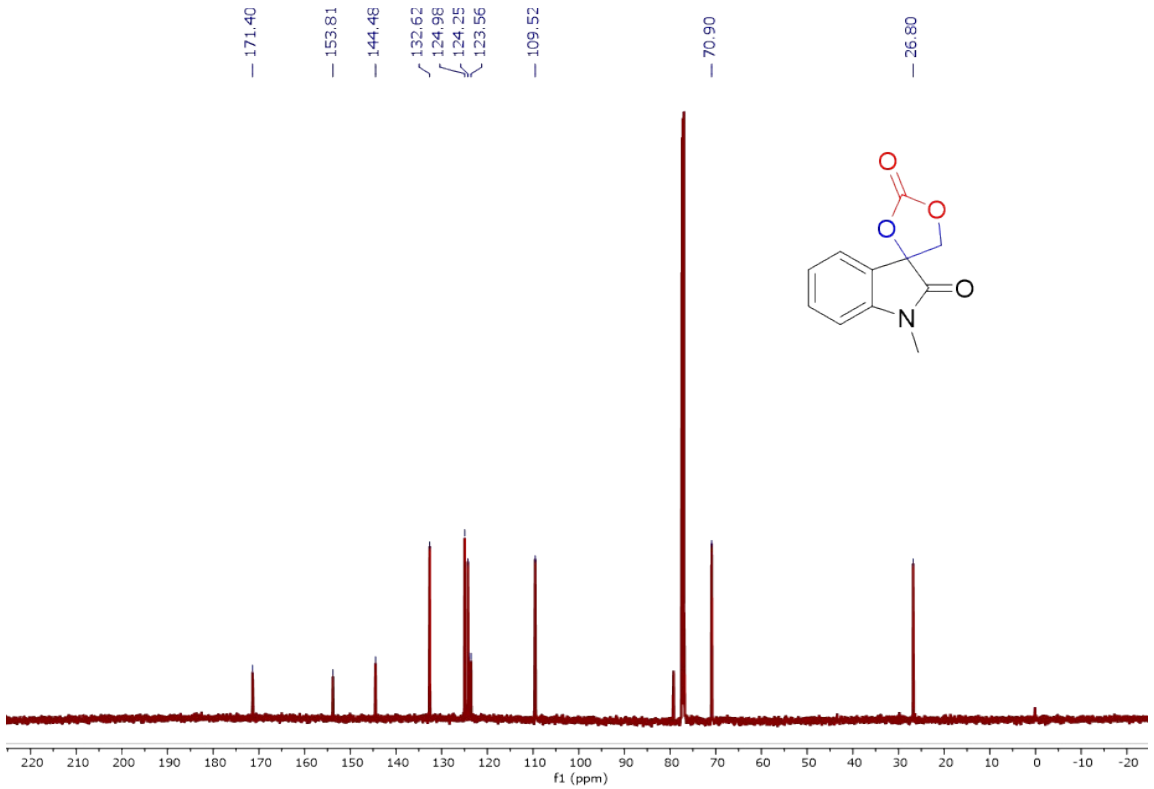


Fig. S34 ^{13}C NMR Spectrum of 1'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

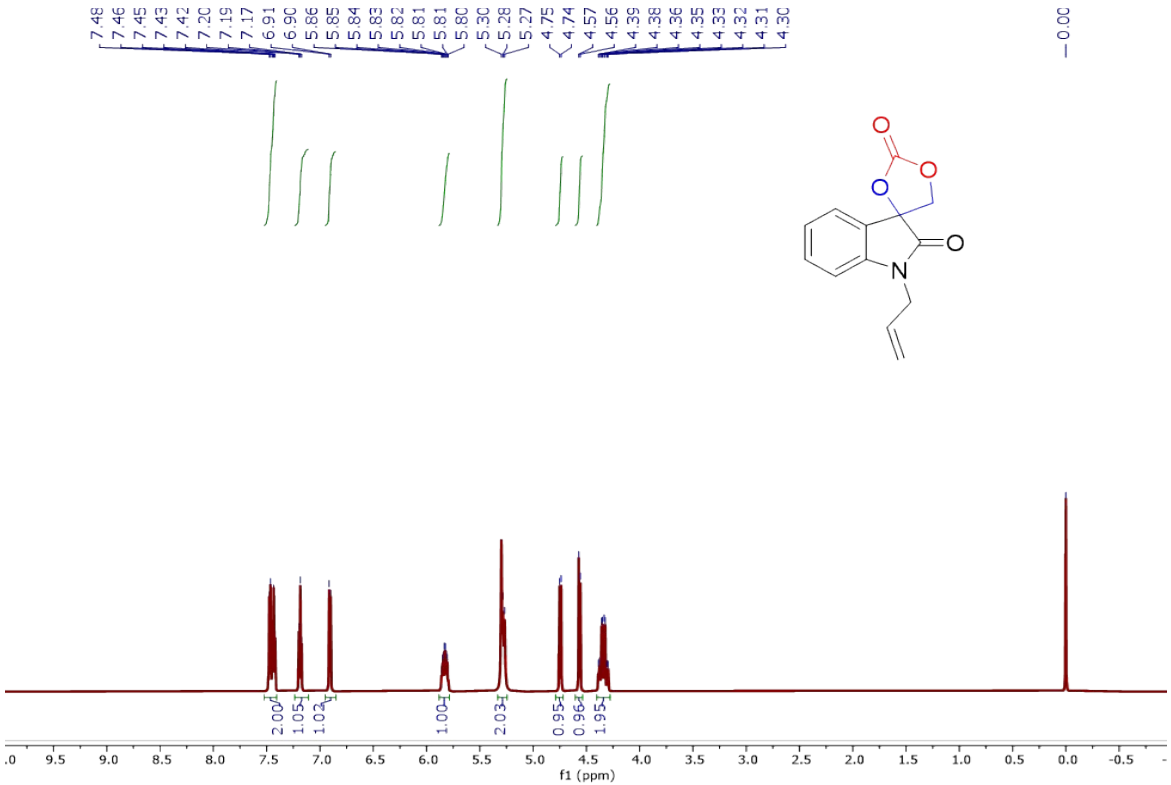


Fig. S35 ^{1}H NMR Spectrum of 1'-allylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

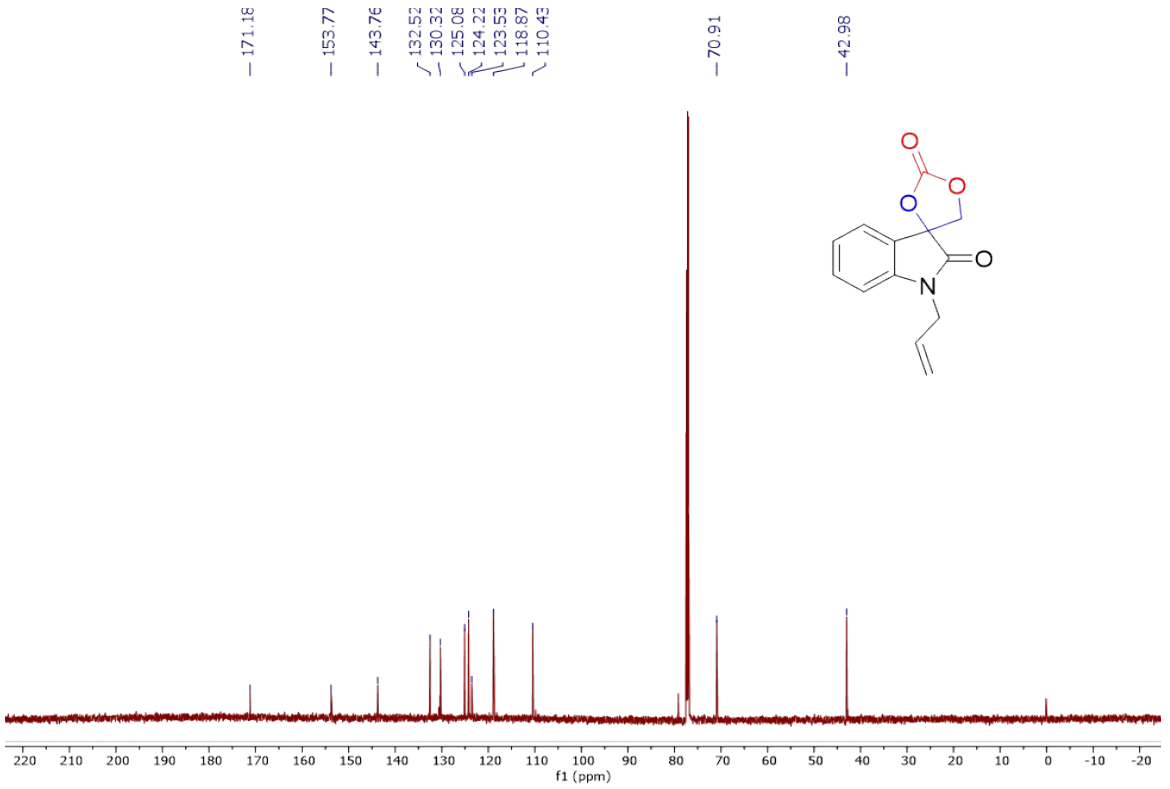


Fig. S36 ^{13}C NMR Spectrum of 1'-allylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

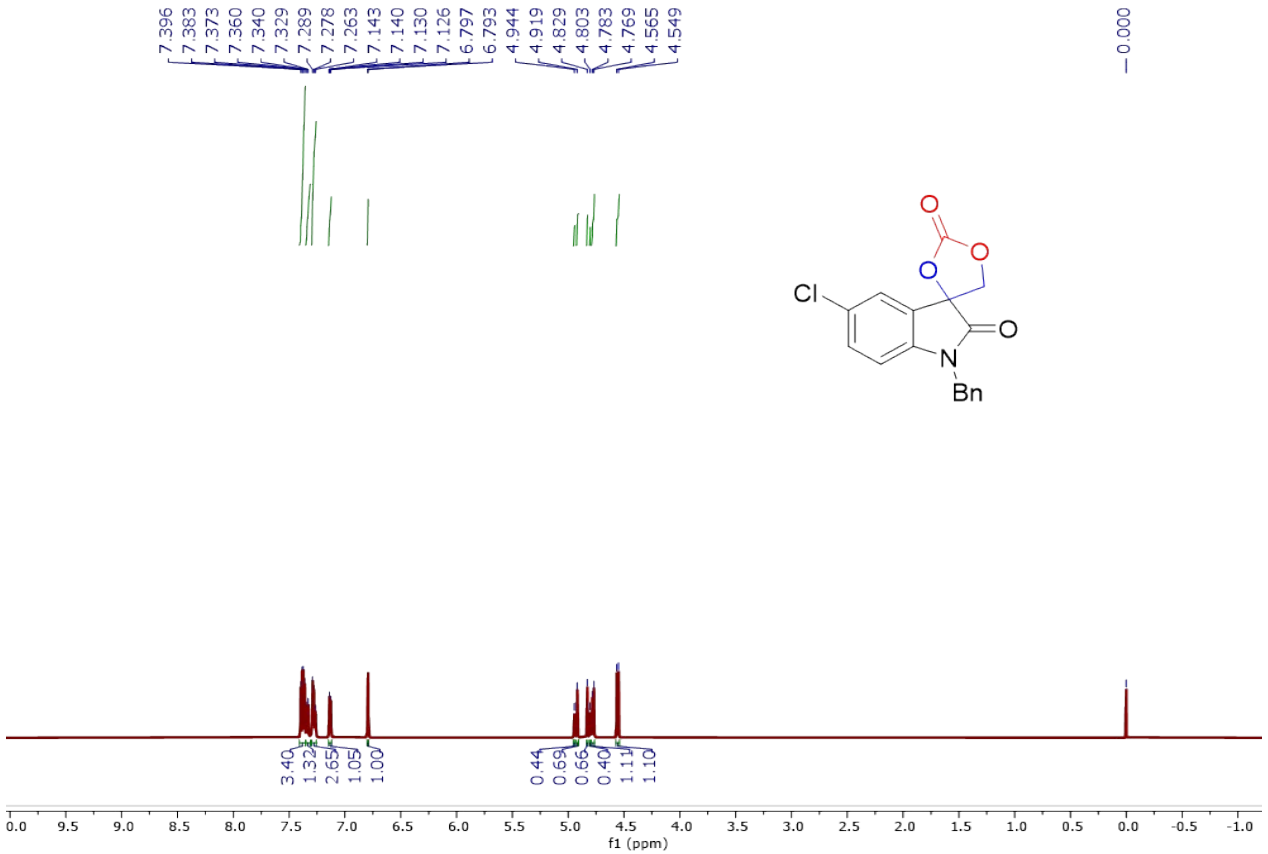


Fig. S37 ^{1}H NMR Spectrum of 1'-benzyl-5'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

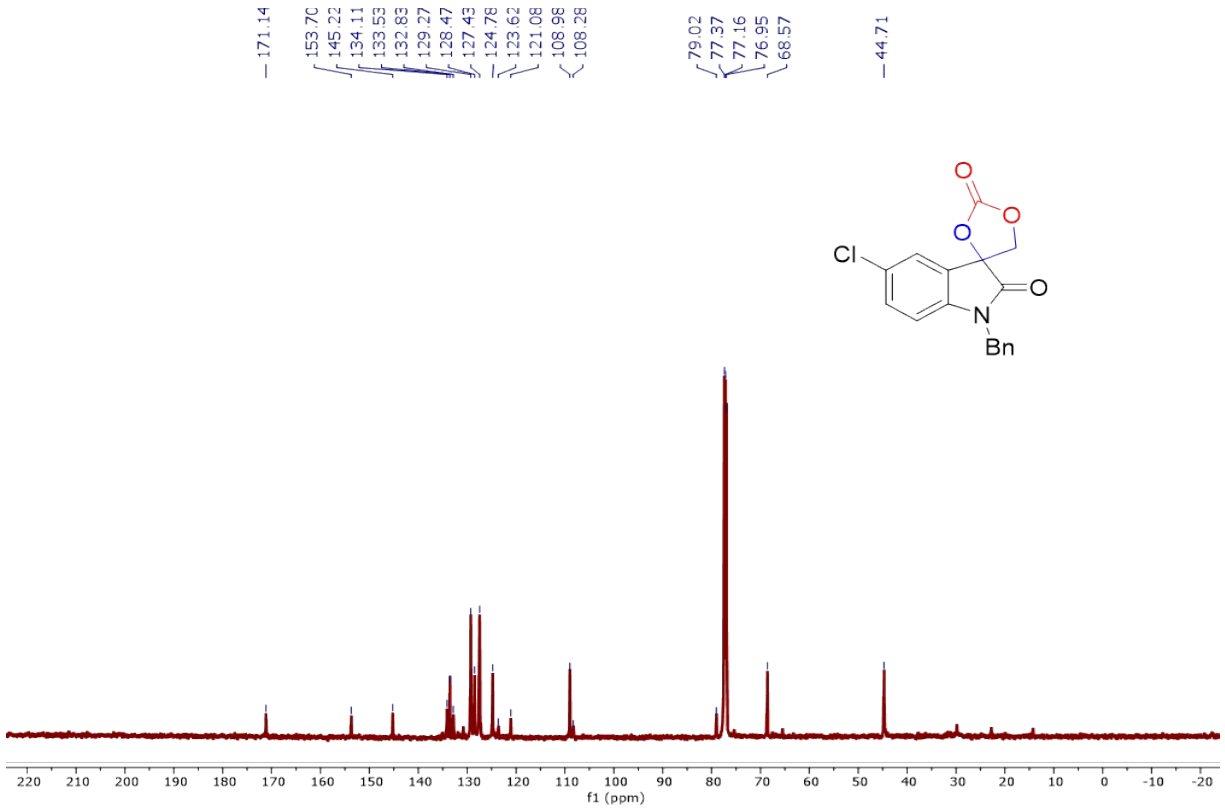


Fig. S38 ^{13}C NMR Spectrum of 1'-benzyl-5'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

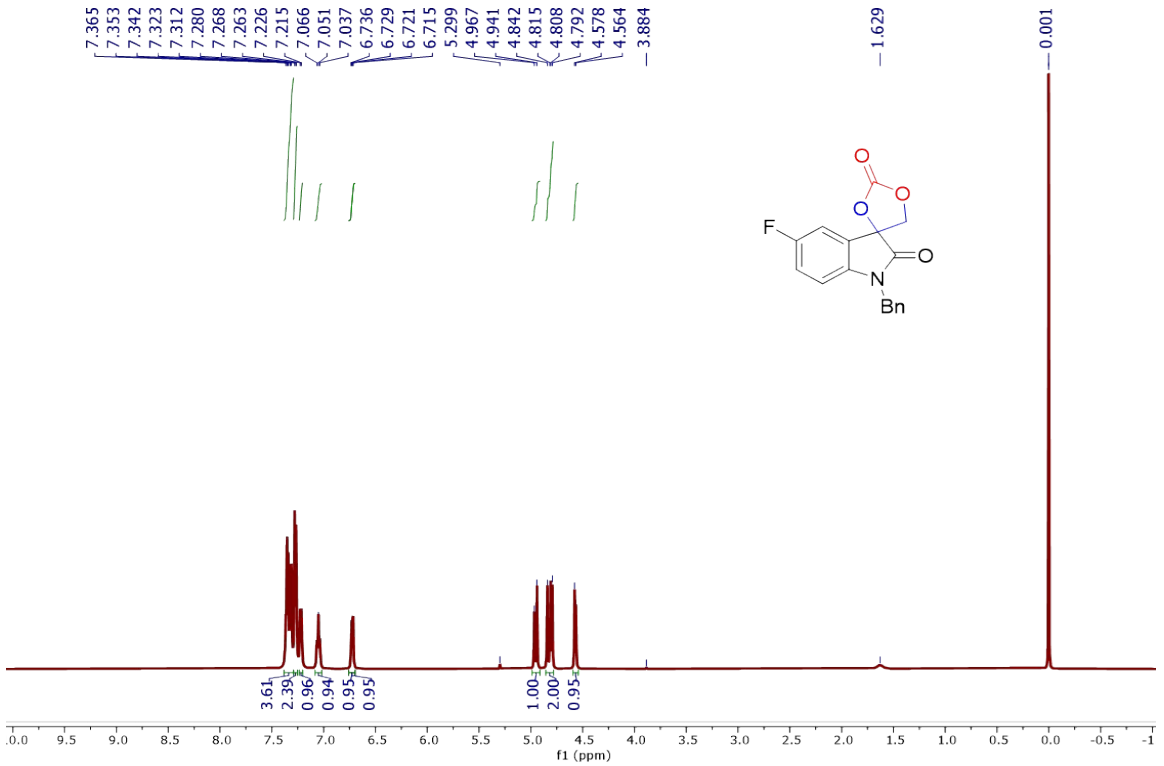


Fig. S39 ^{1}H NMR Spectrum of 1'-benzyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

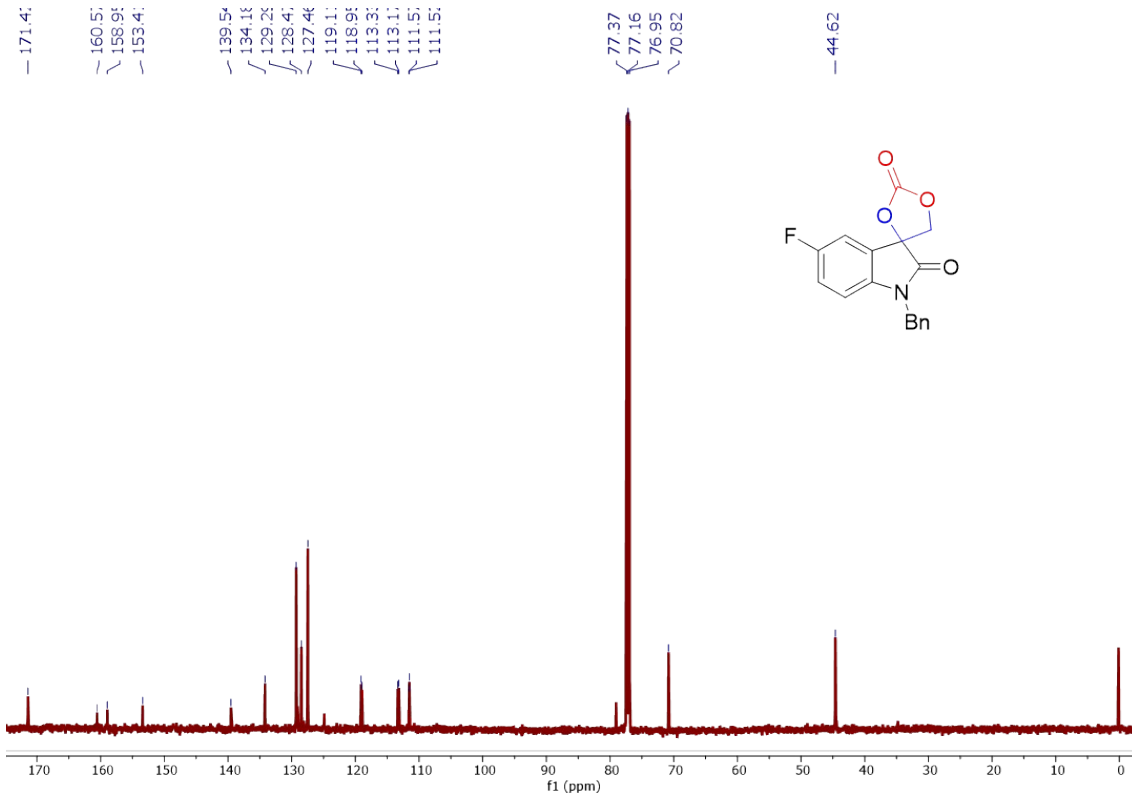


Fig. S40 ^{13}C NMR Spectrum of 1'-benzyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

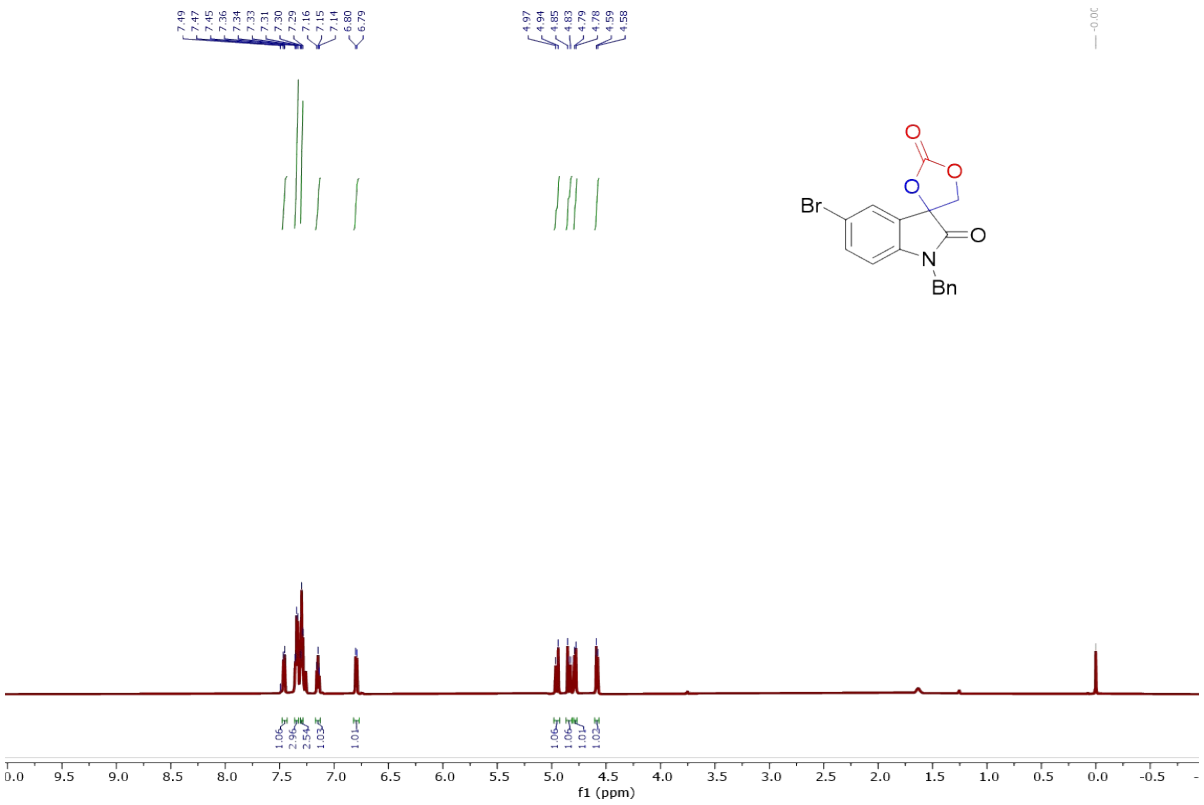


Fig. S41 ^{1}H NMR Spectrum of 1'-benzyl-5'-bromospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

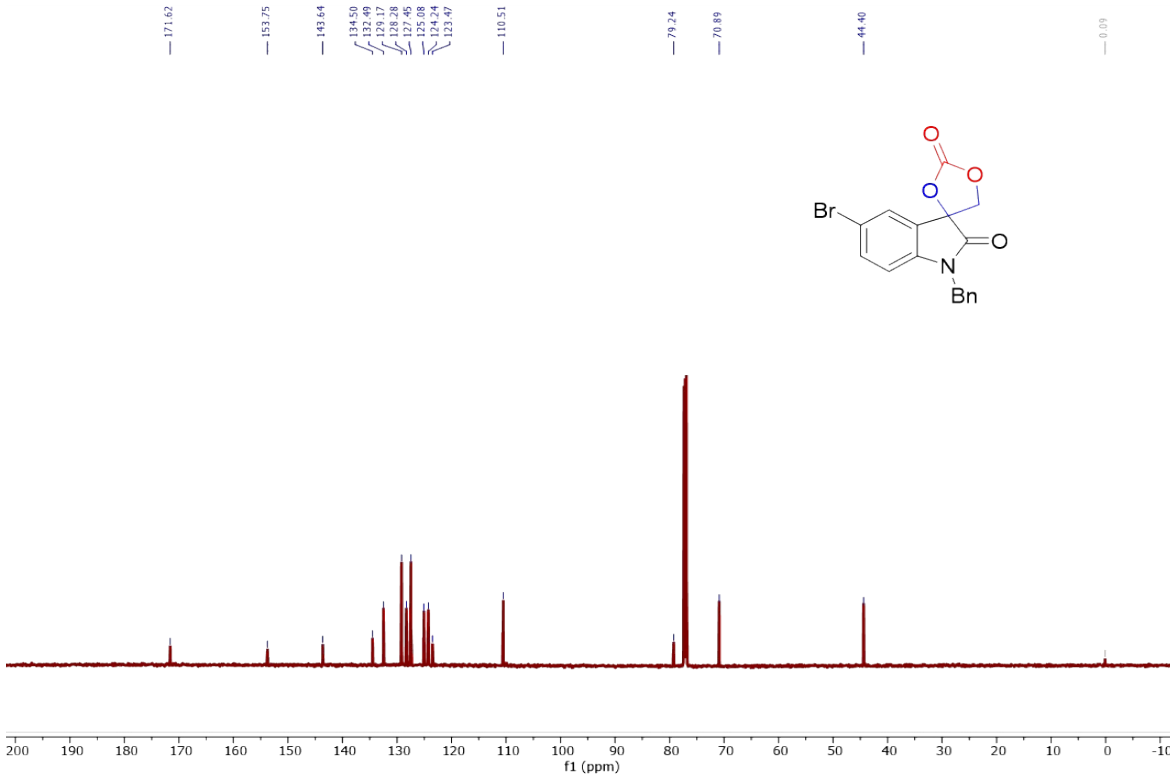


Fig. S42 ^{13}C NMR Spectrum of 1'-benzyl-5'-bromospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

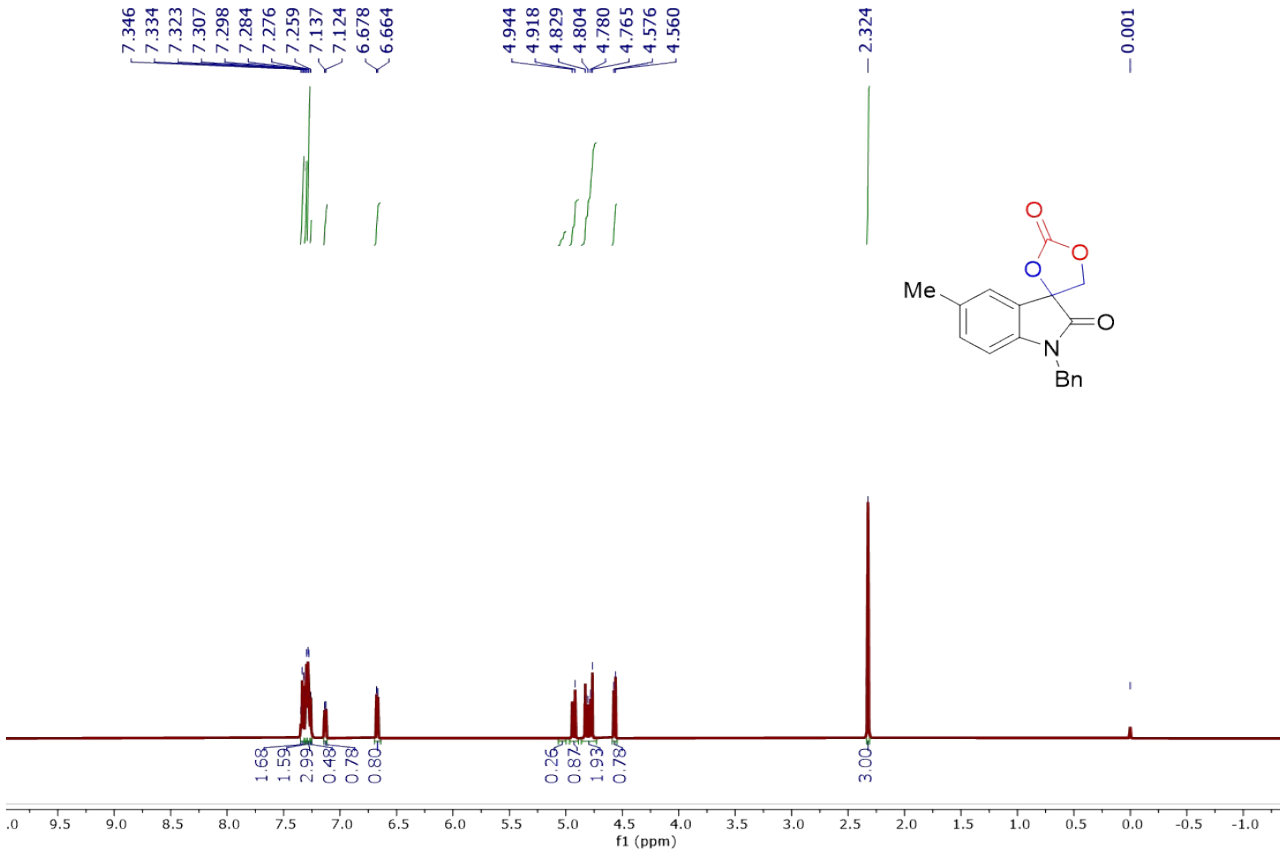


Fig. S43 ^{1}H NMR Spectrum of 1'-benzyl-5'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

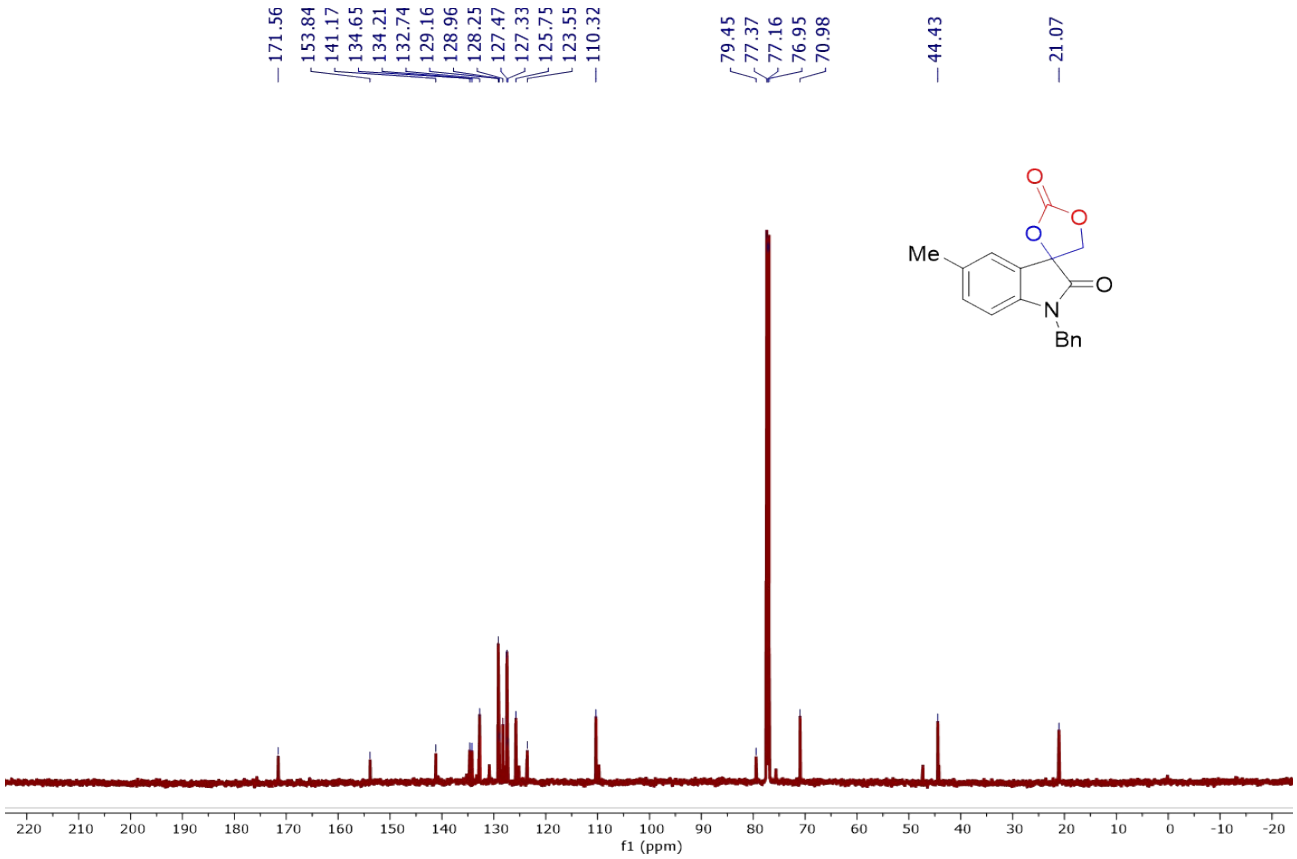


Fig. S44 ^{13}C NMR Spectrum of 1'-benzyl-5'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

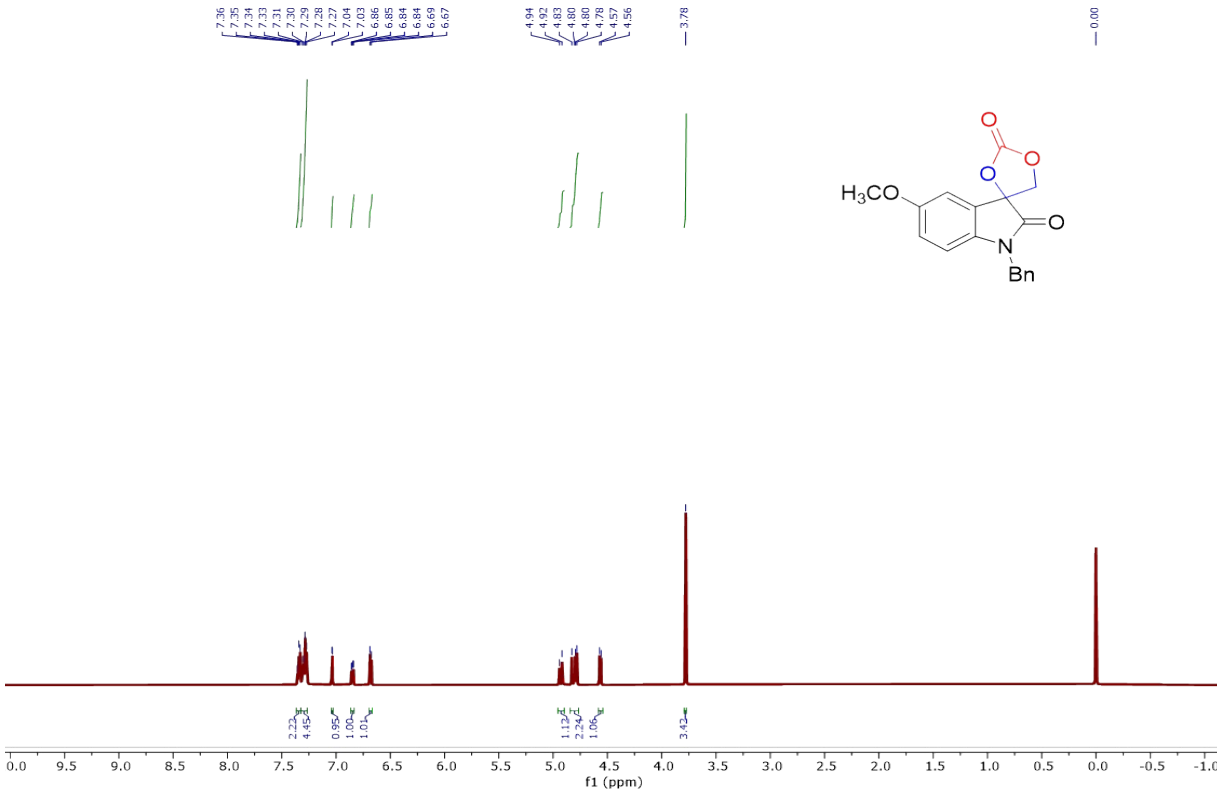


Fig. S45 ^{1}H NMR Spectrum of 1'-benzyl-5'-methoxyspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

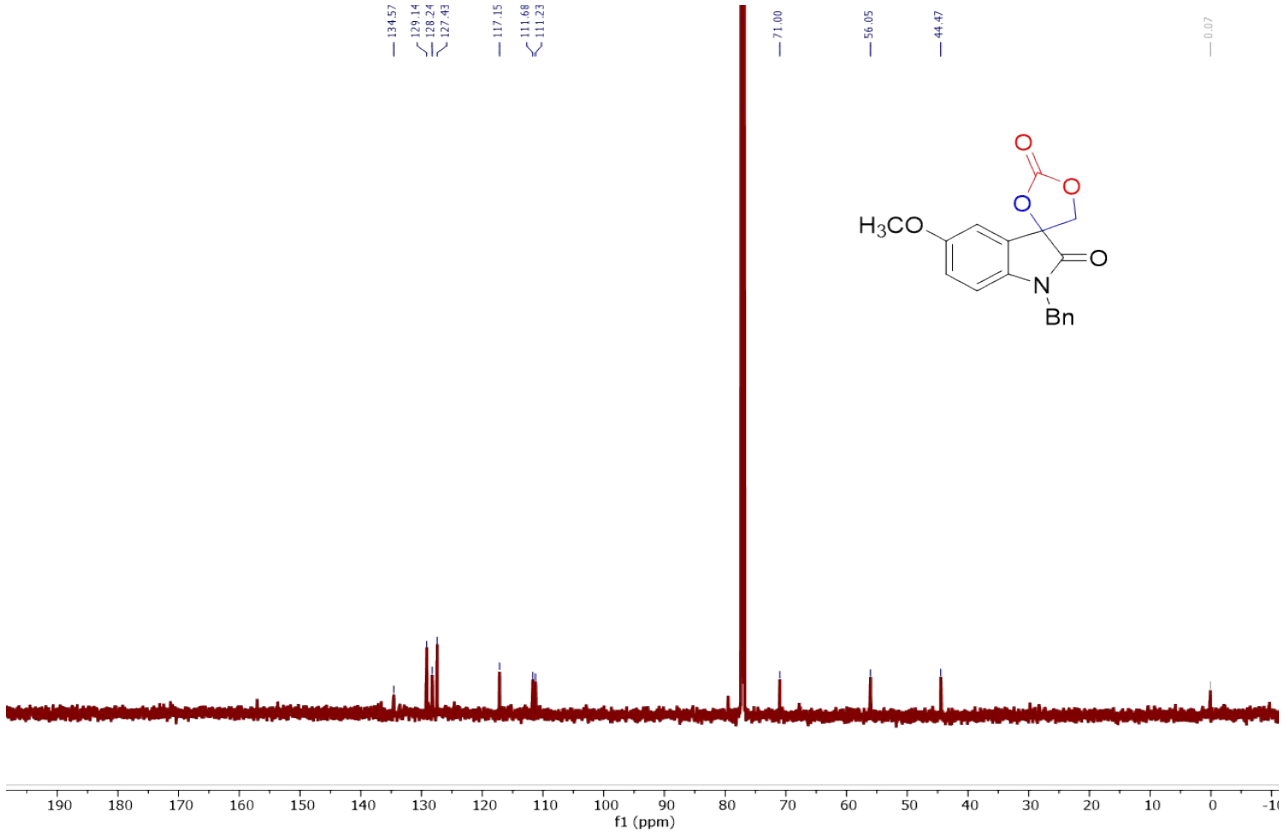


Fig. S46 ^{13}C NMR Spectrum of 1'-benzyl-5'-methoxyspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

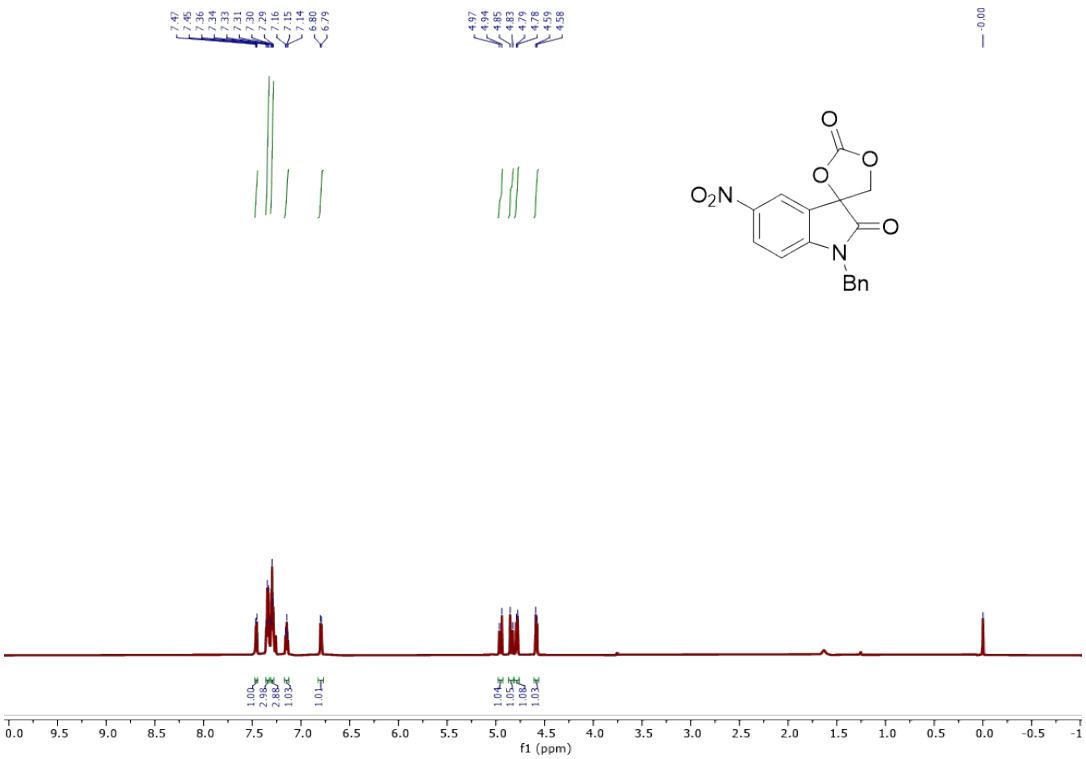


Fig. S47 ^{1}H NMR Spectrum of 1'-benzyl-5'-nitrospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

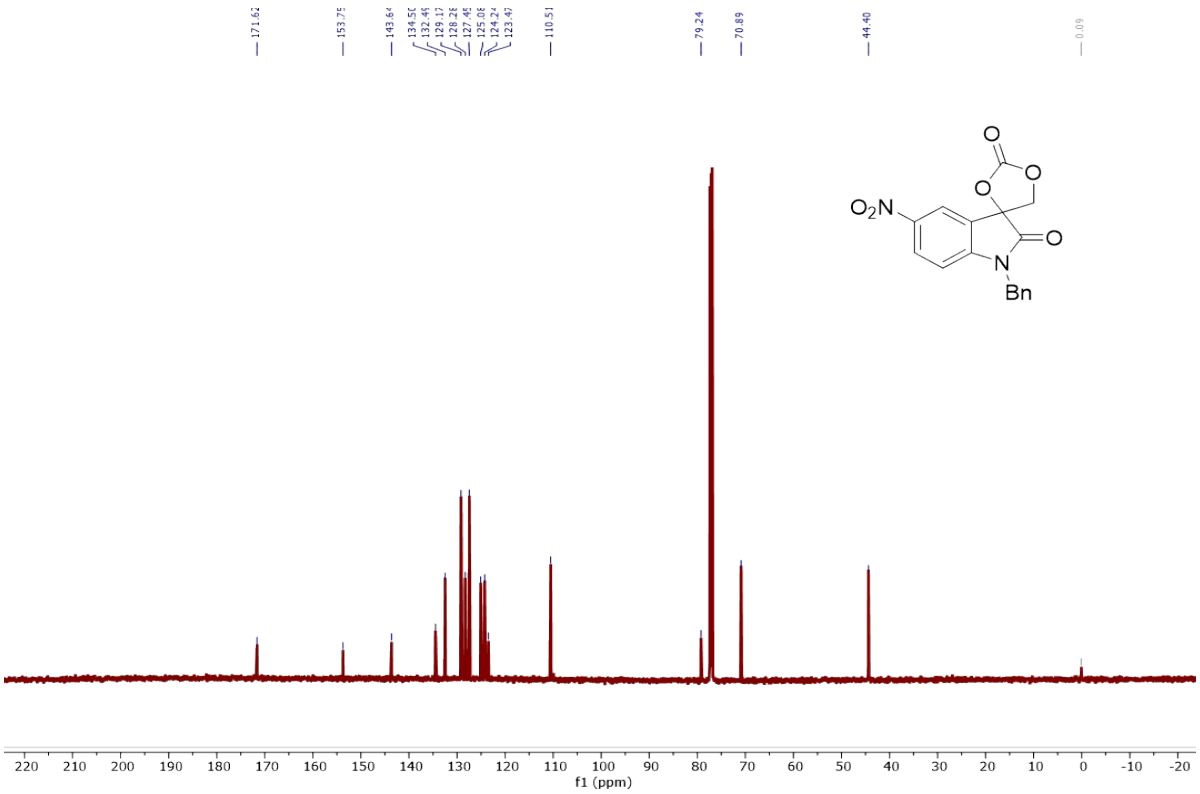


Fig. S48 ^{13}C NMR Spectrum of 1'-benzyl-5'-nitrospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

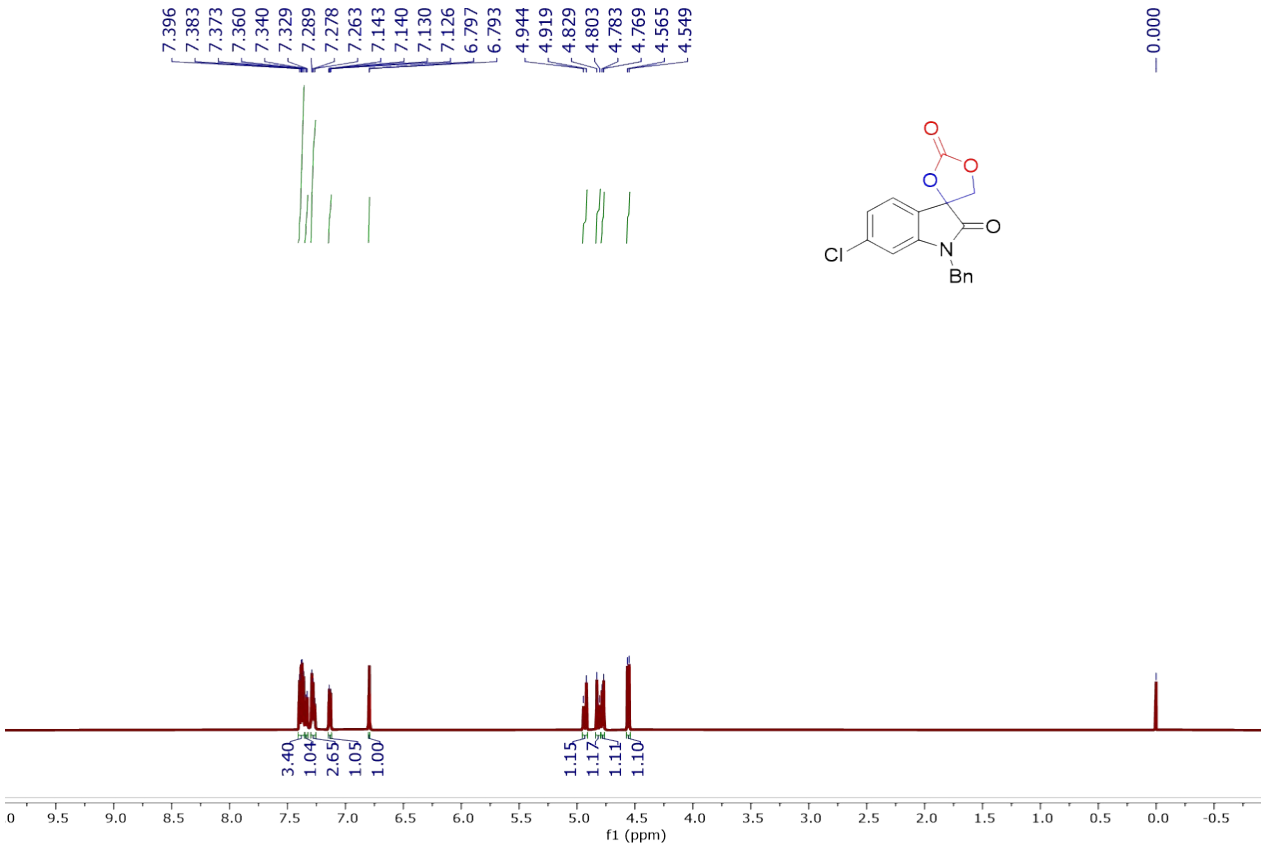


Fig. S49 ^{1}H NMR Spectrum of 1'-benzyl-6'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'- ($CDCl_3$, 600 MHz, 298K)

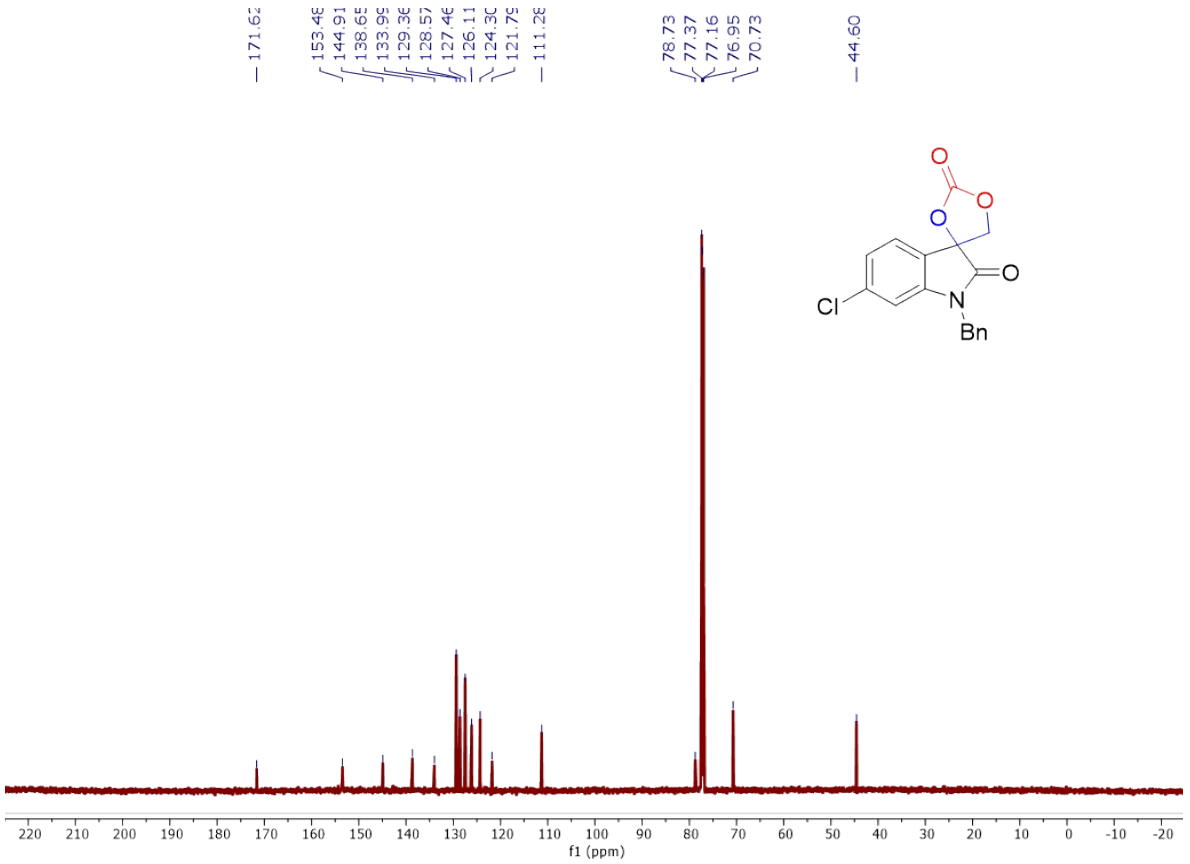


Fig. S50 ^{13}C NMR Spectrum of 1'-benzyl-6'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

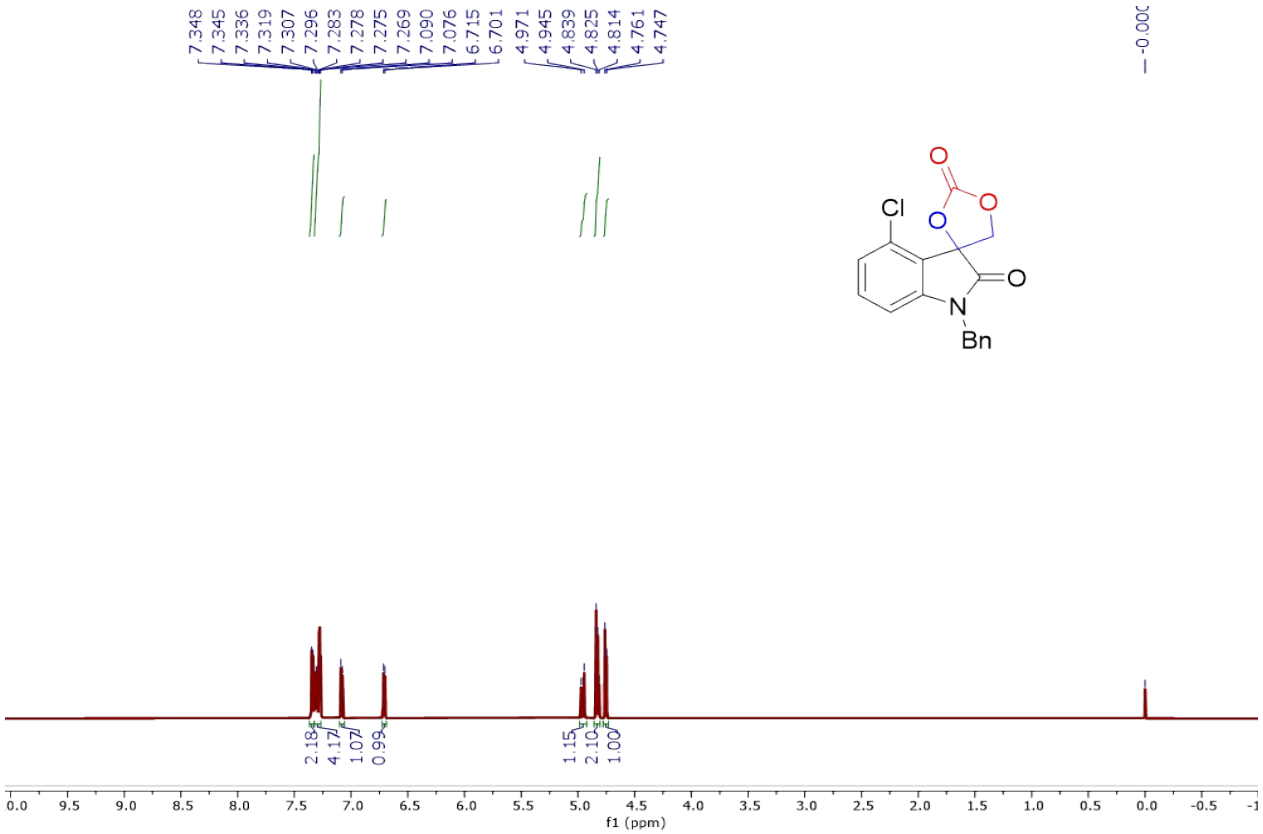


Fig. S51 ^{1}H NMR Spectrum of 1'-benzyl-4'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

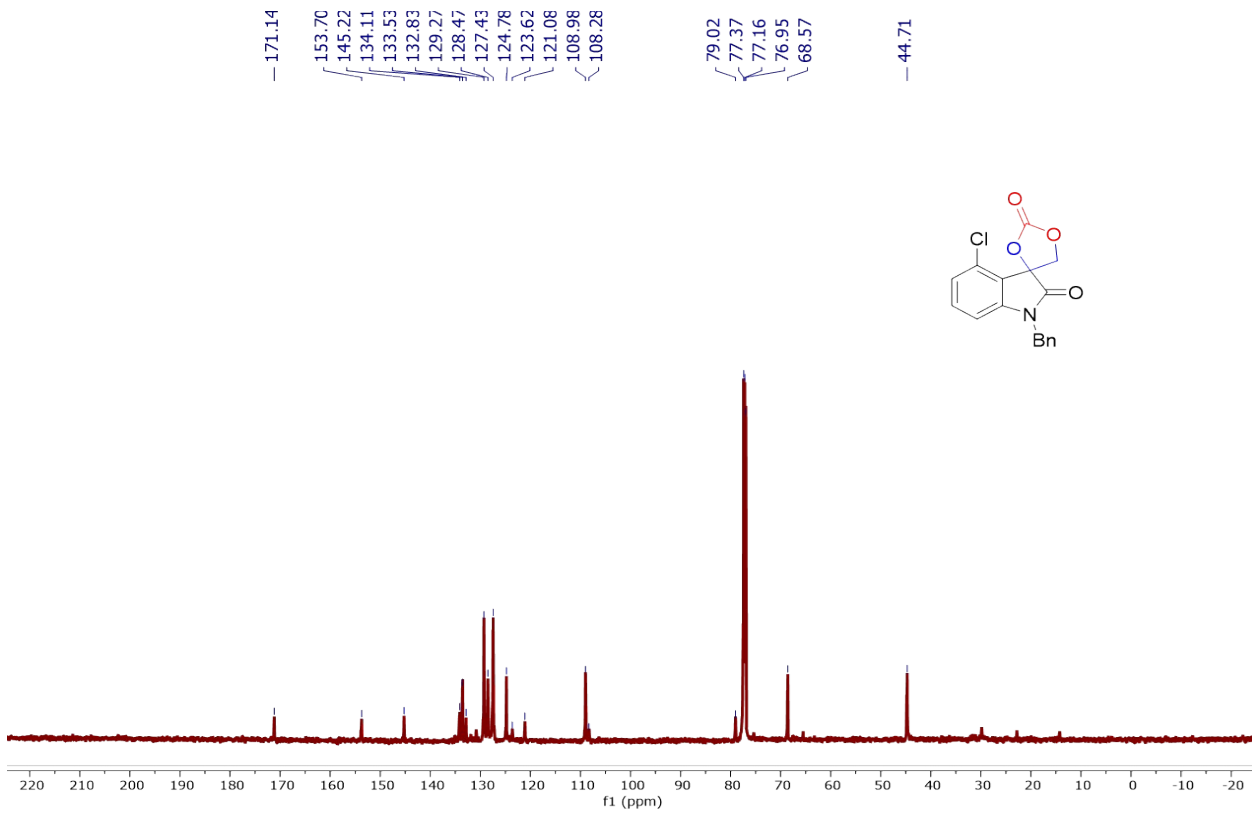


Fig. S52 ^{13}C NMR Spectrum of 1'-benzyl-4'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

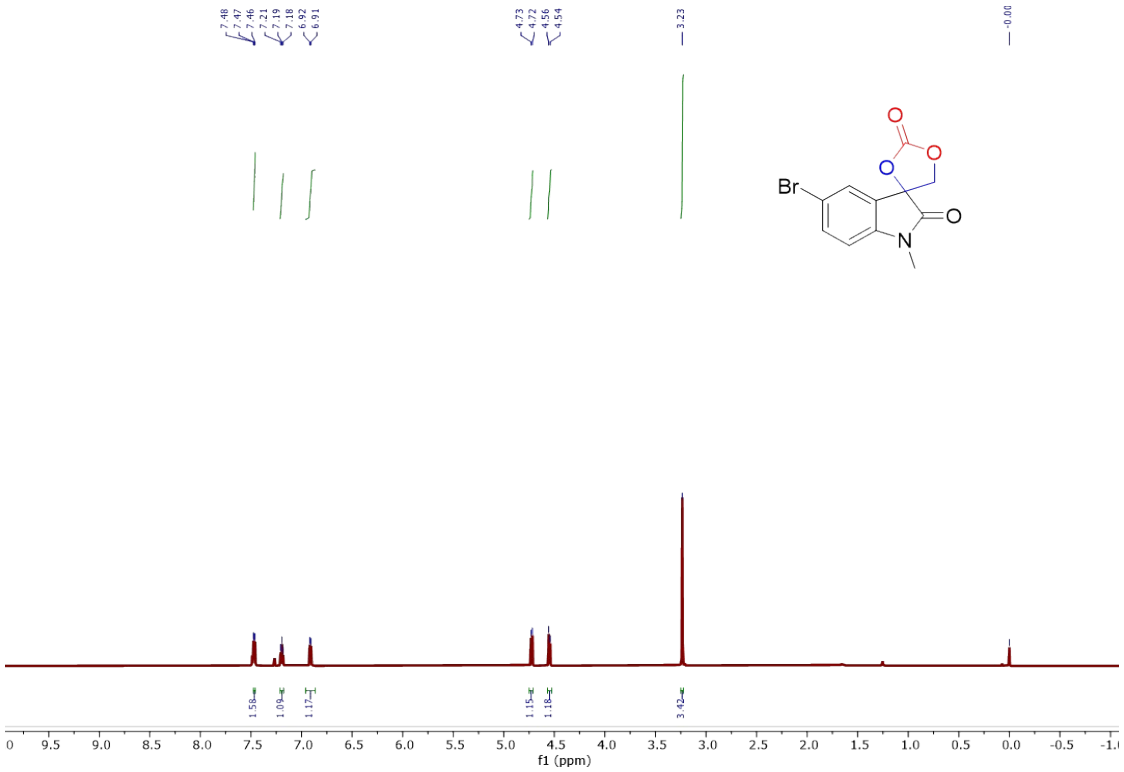


Fig. S53 ^{1}H NMR Spectrum of 1'-methyl-5'-bromospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

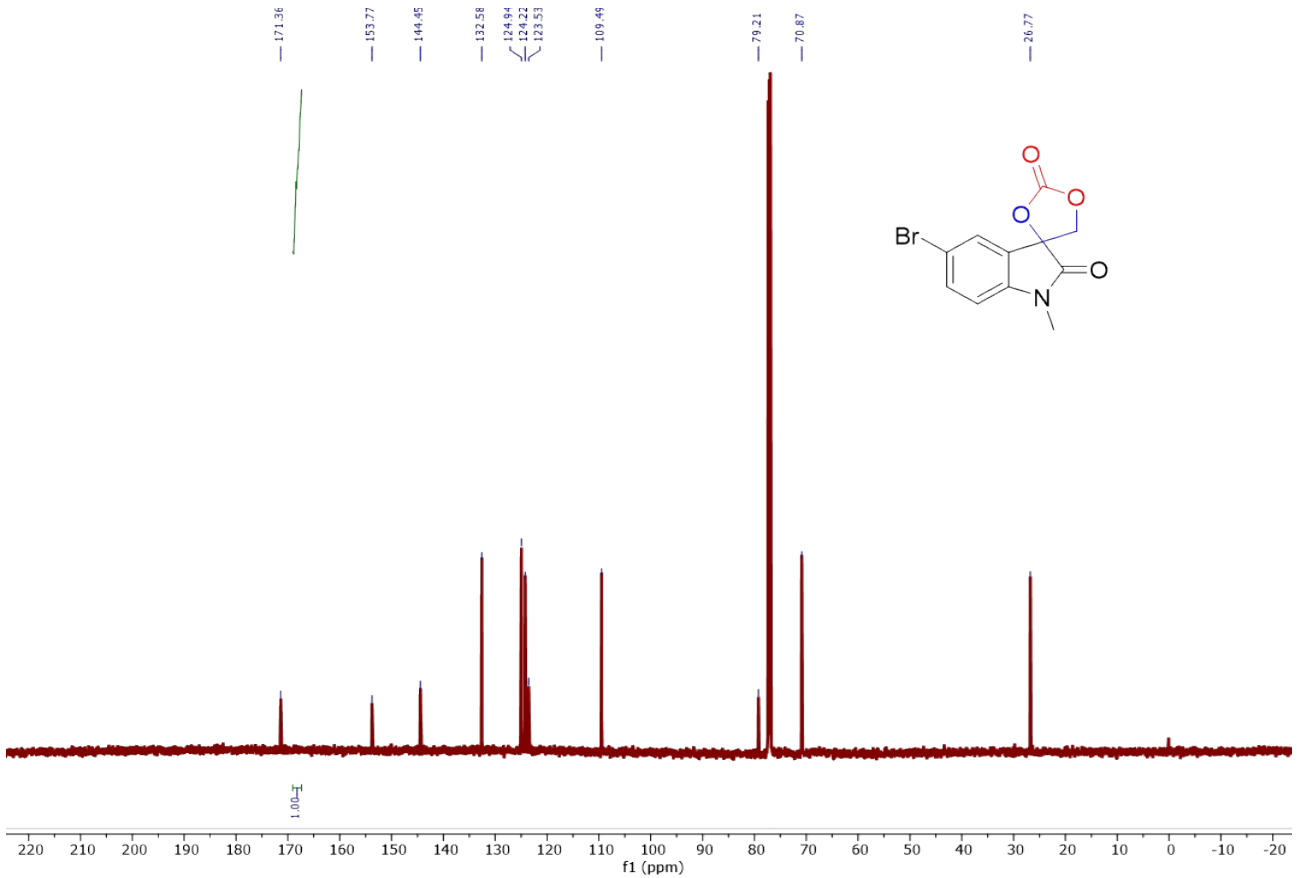


Fig. S54 ^{13}C NMR Spectrum of 1'-methyl-5'-bromospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

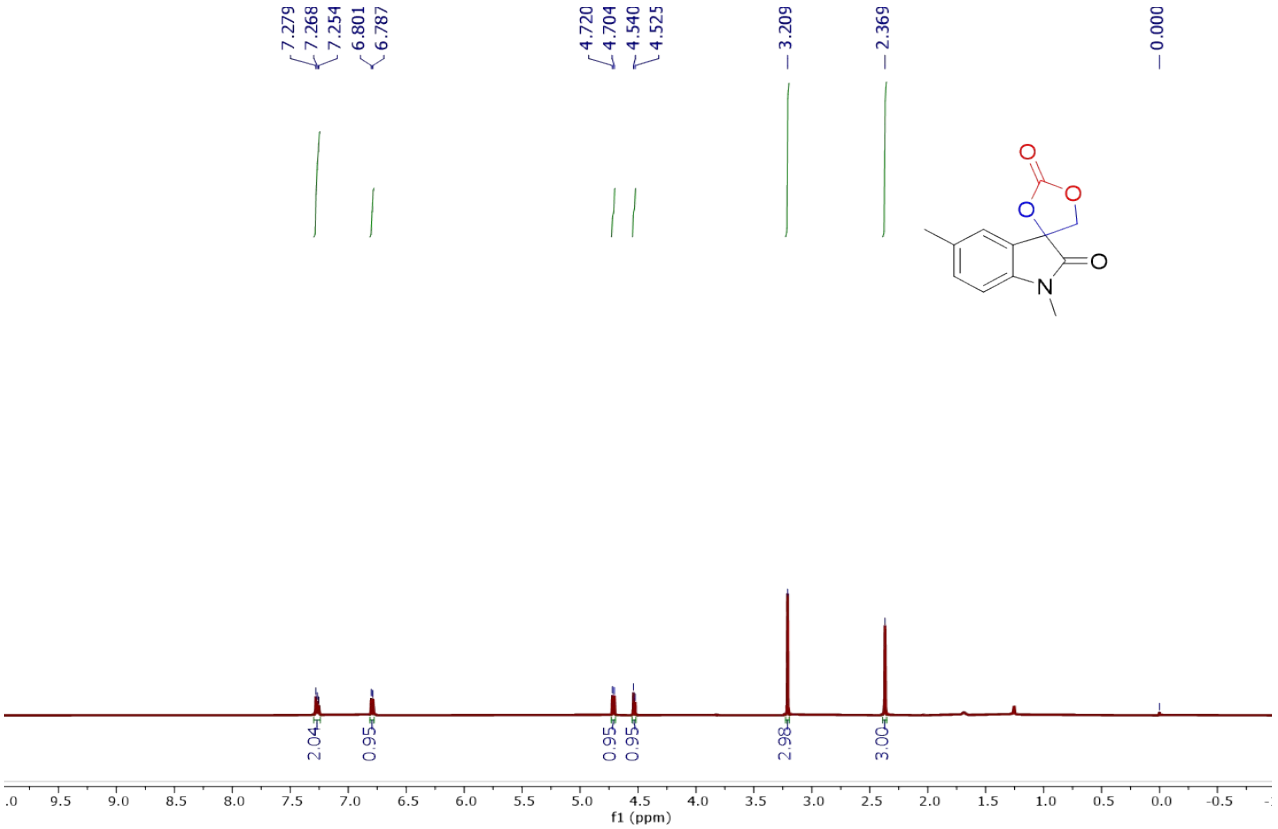


Fig. S55 ^{1}H NMR Spectrum of 1'-methyl-5'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

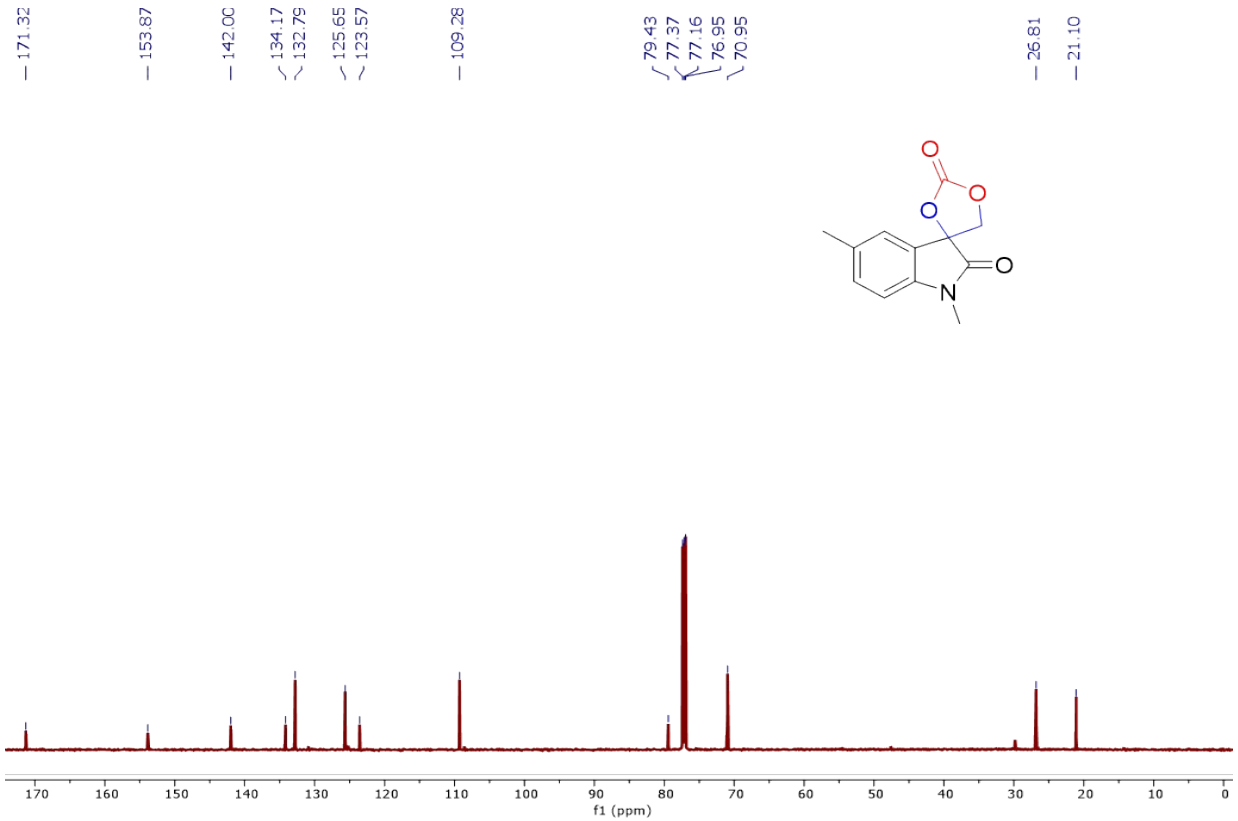


Fig. S56 ^{13}C NMR Spectrum of 1'-methyl-5'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

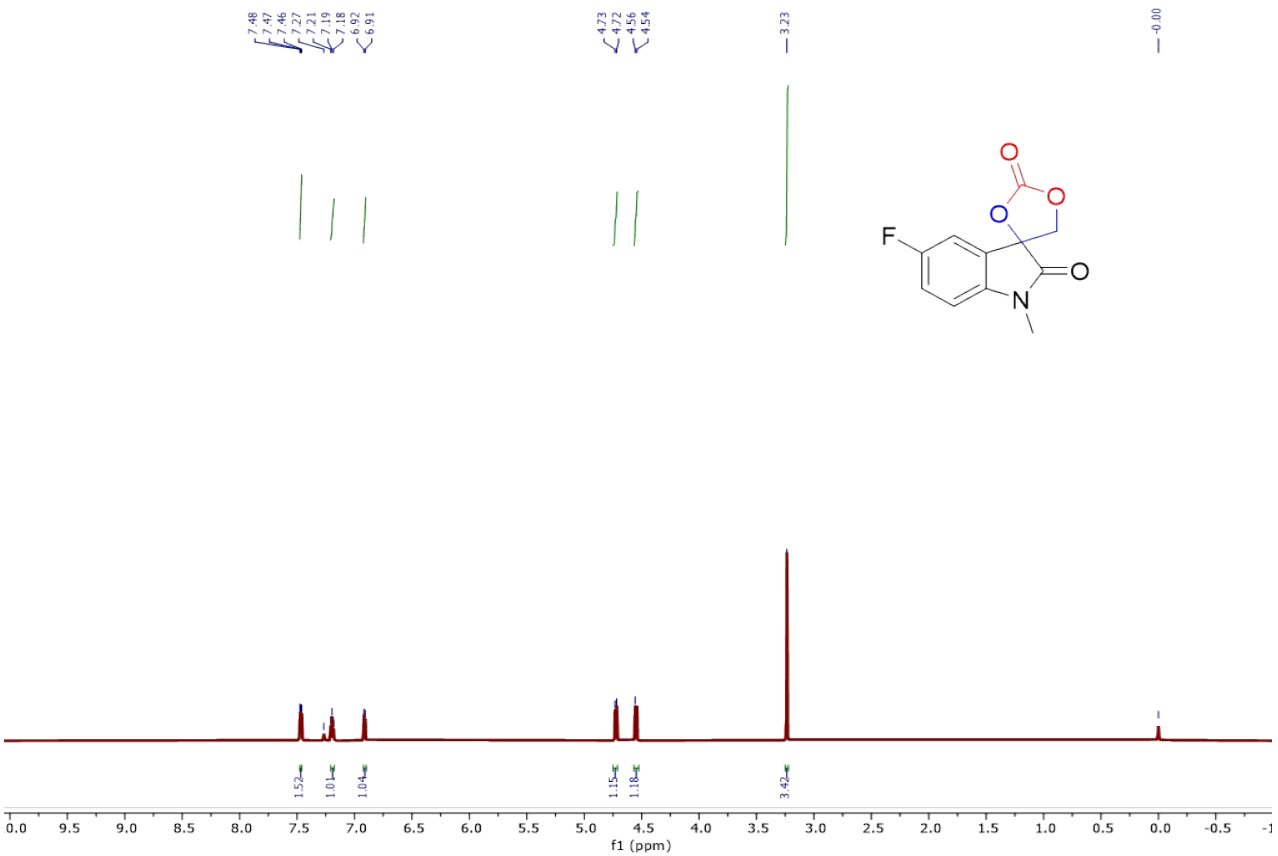


Fig. S57 ^{1}H NMR Spectrum of 1'-methyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

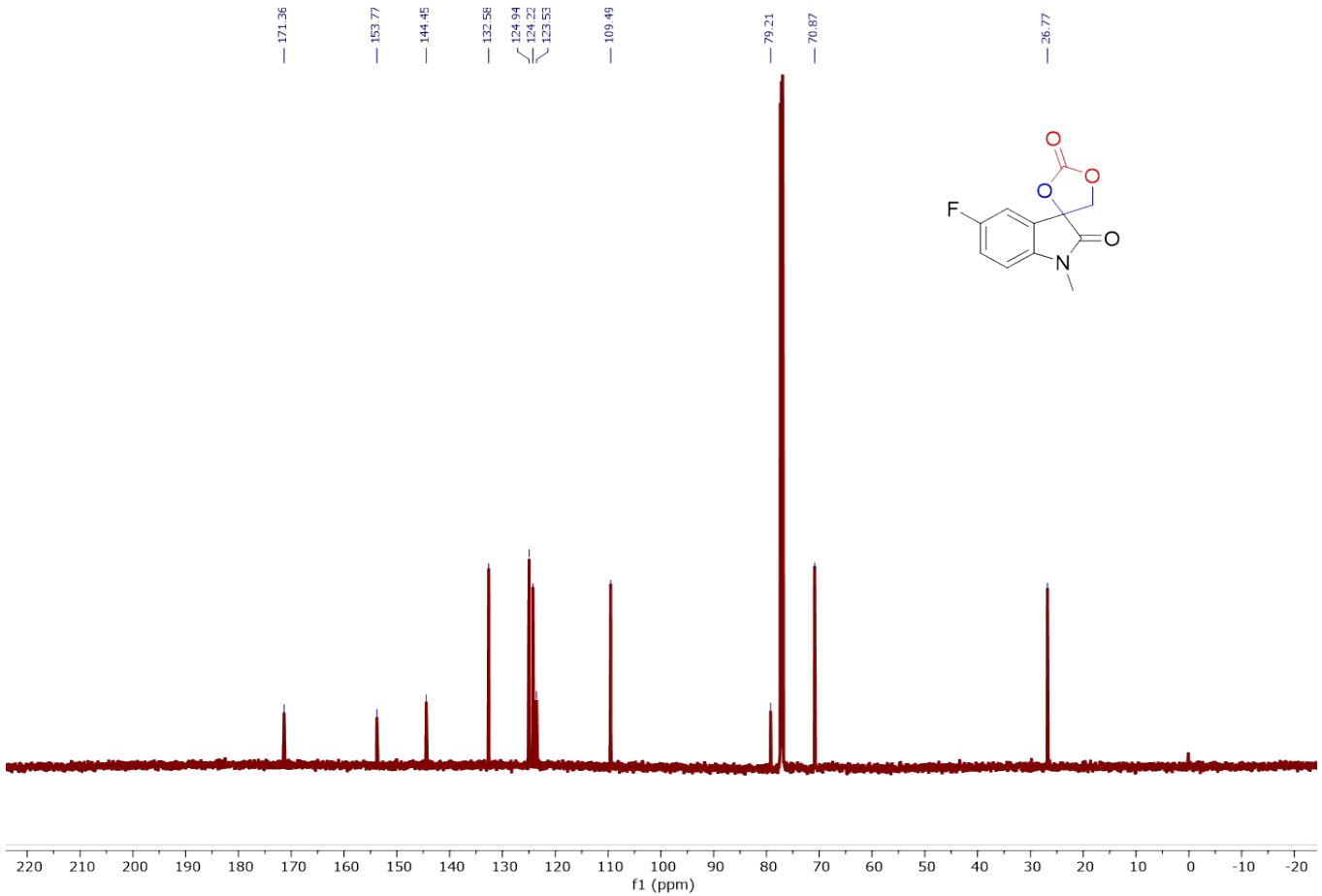


Fig. S58 ^{13}C NMR Spectrum of 1'-methyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 151 MHz, 298K)

17. ^{19}F NMR spectra of product

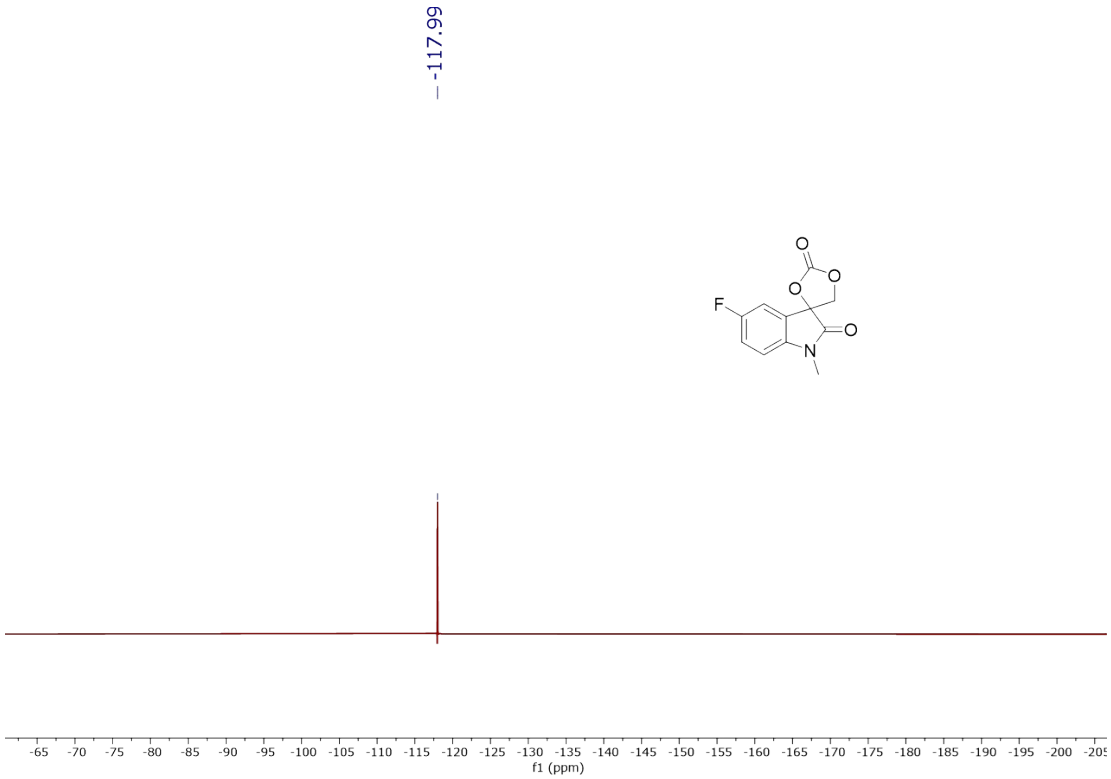


Fig. S59 ^{19}F NMR Spectrum of 1'-methyl-5'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

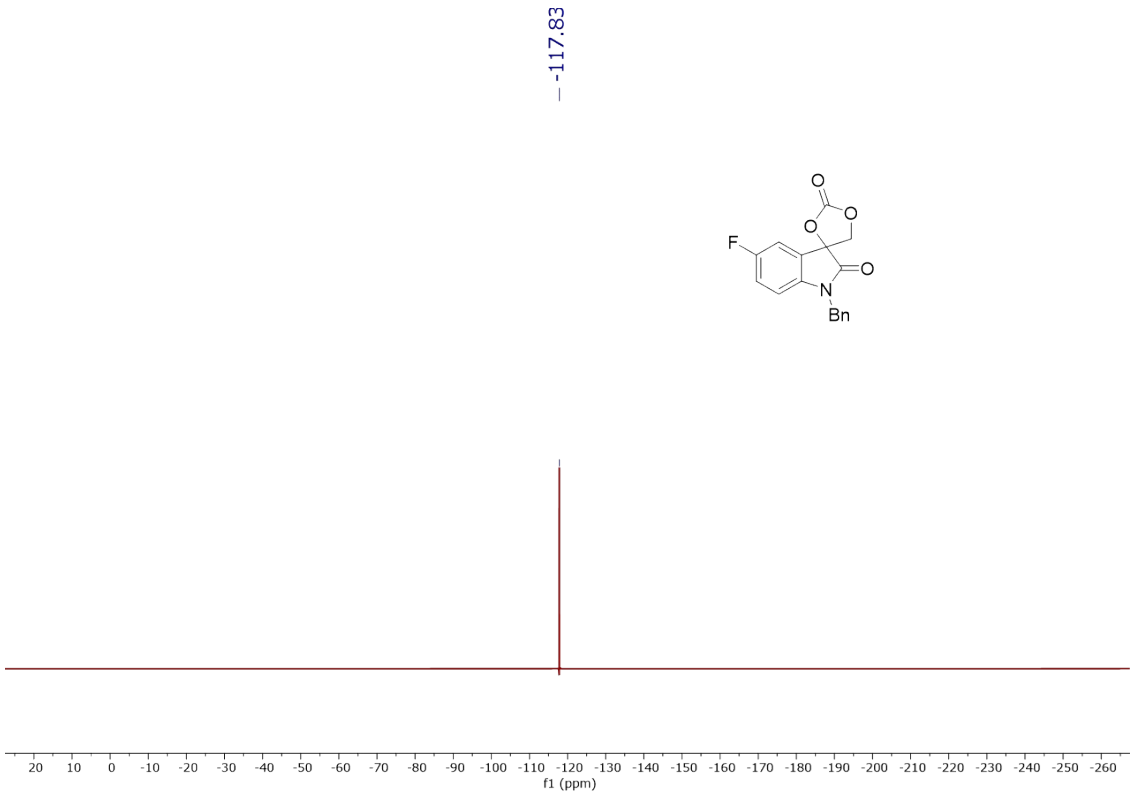


Fig. S60 ^{19}F NMR Spectrum of 1'-benzyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione ($CDCl_3$, 600 MHz, 298K)

18. ^{1}H and ^{13}C NMR spectra of Cyclic carbonate products

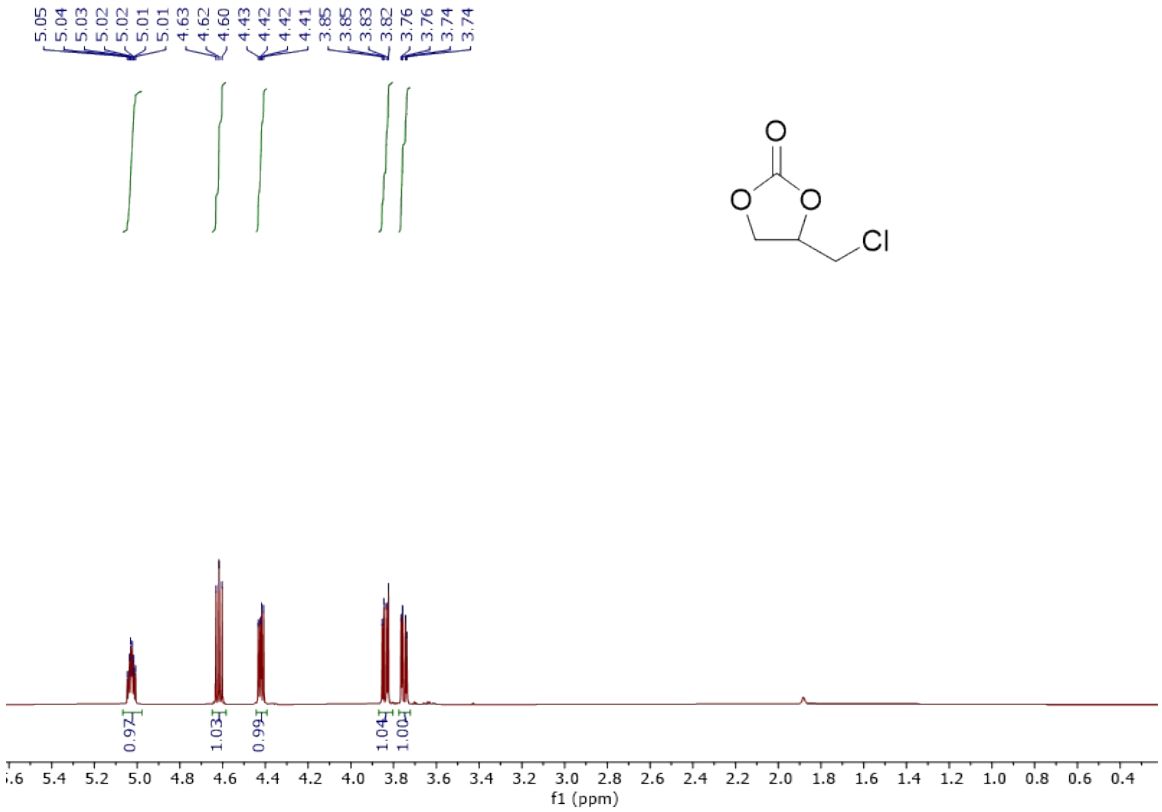


Fig. S61 ^{1}H NMR Spectrum of 4-(chloromethyl)-1,3-dioxolan-2-one ($CDCl_3$, 600 MHz, 298K)

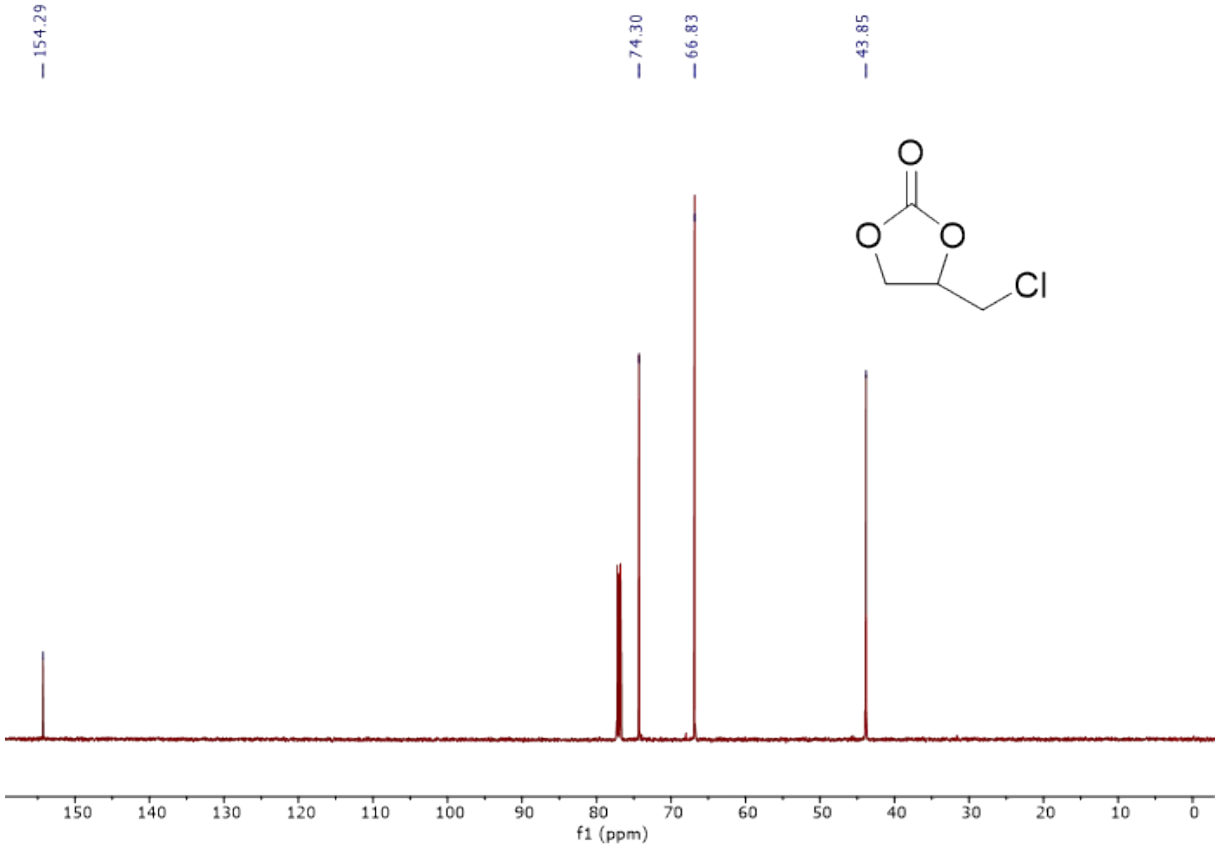


Fig. S62 ^{13}C NMR Spectrum of 4-(chloromethyl)-1,3-dioxolan-2-one ($CDCl_3$, 151 MHz, 298K)

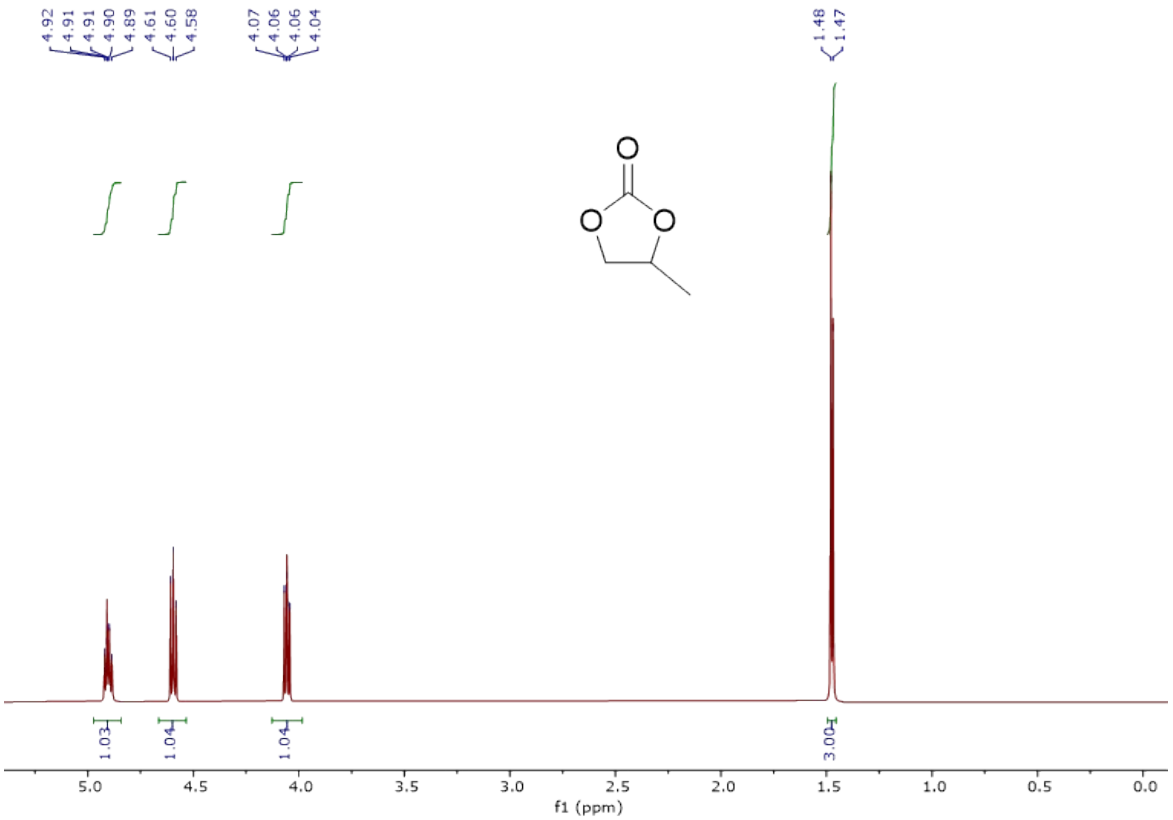


Fig. S63 ^{1}H NMR Spectrum of 4-methyl-1,3-dioxolan-2-one ($CDCl_3$, 600 MHz, 298K)

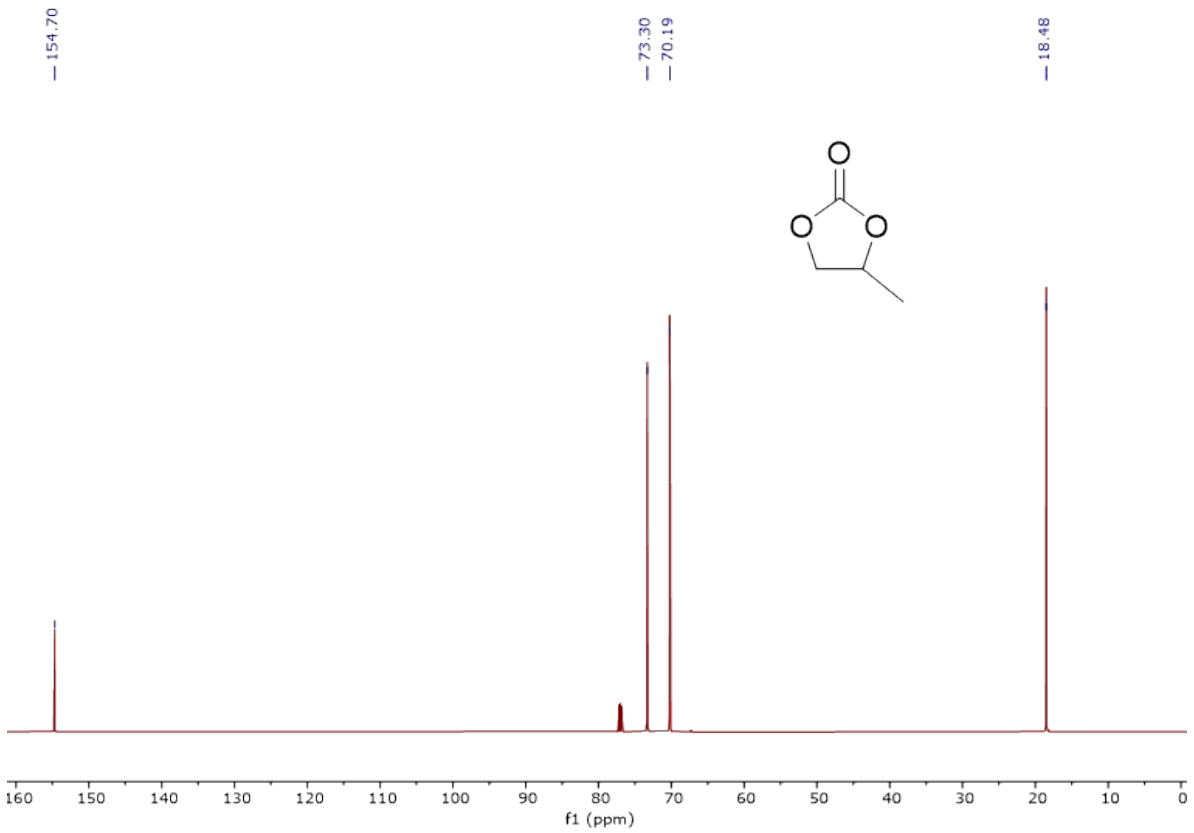


Fig. S64 ^{13}C NMR Spectrum of 4-methyl-1,3-dioxolan-2-one ($CDCl_3$, 151 MHz, 298K)

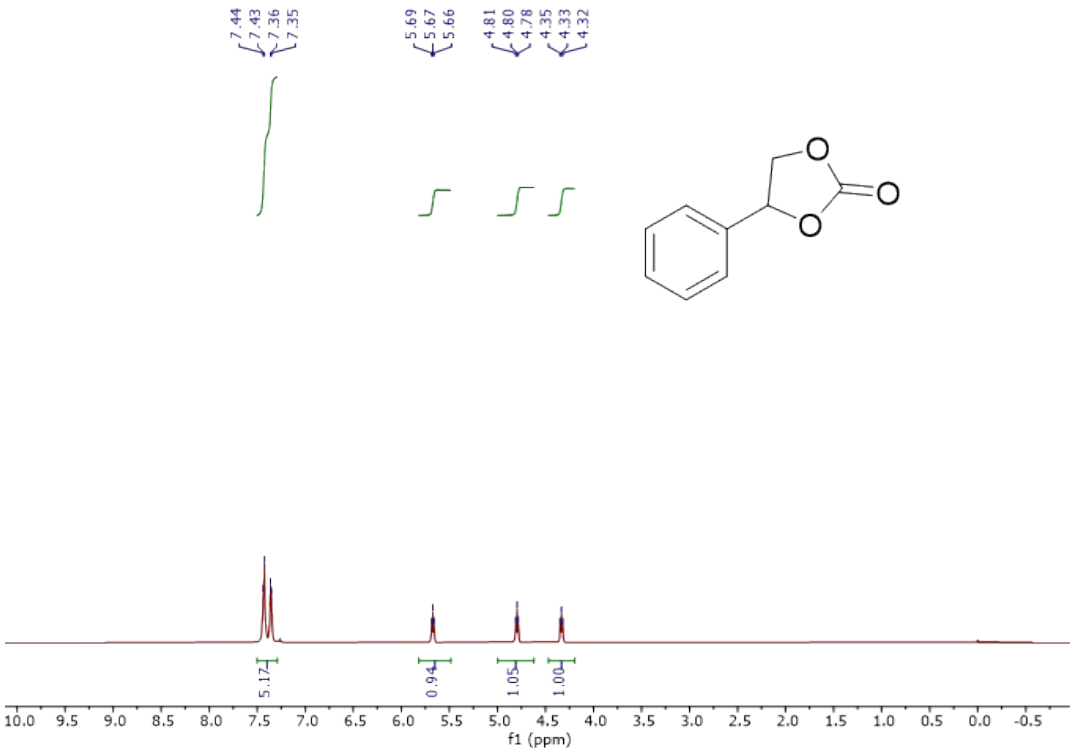


Fig. S65 ^{13}C NMR Spectrum of styrene carbonate ($CDCl_3$, 151 MHz, 298K)

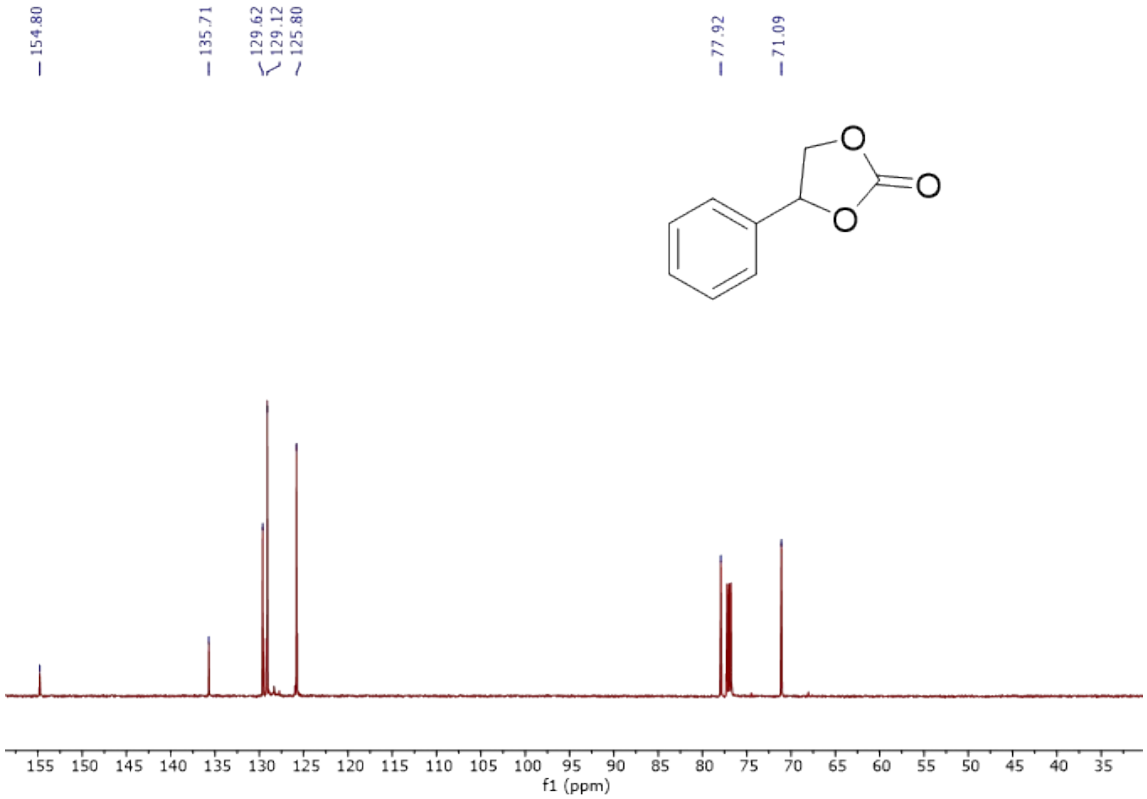


Fig. S66 ^{1}H NMR Spectrum of styrene carbonate ($CDCl_3$, 600 MHz, 298K)

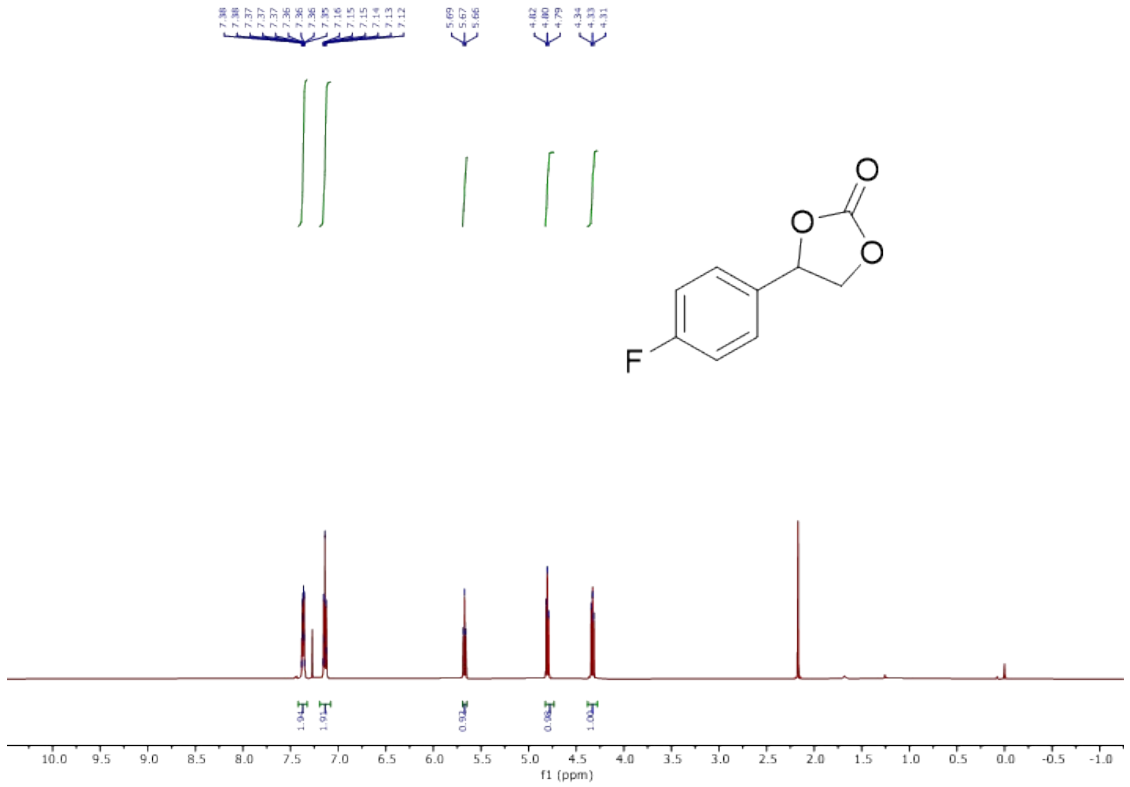


Fig. S67 ^{1}H NMR Spectrum of 4-(4-fluorophenyl)-1,3-dioxolan-2-one ($CDCl_3$, 600 MHz, 298K)

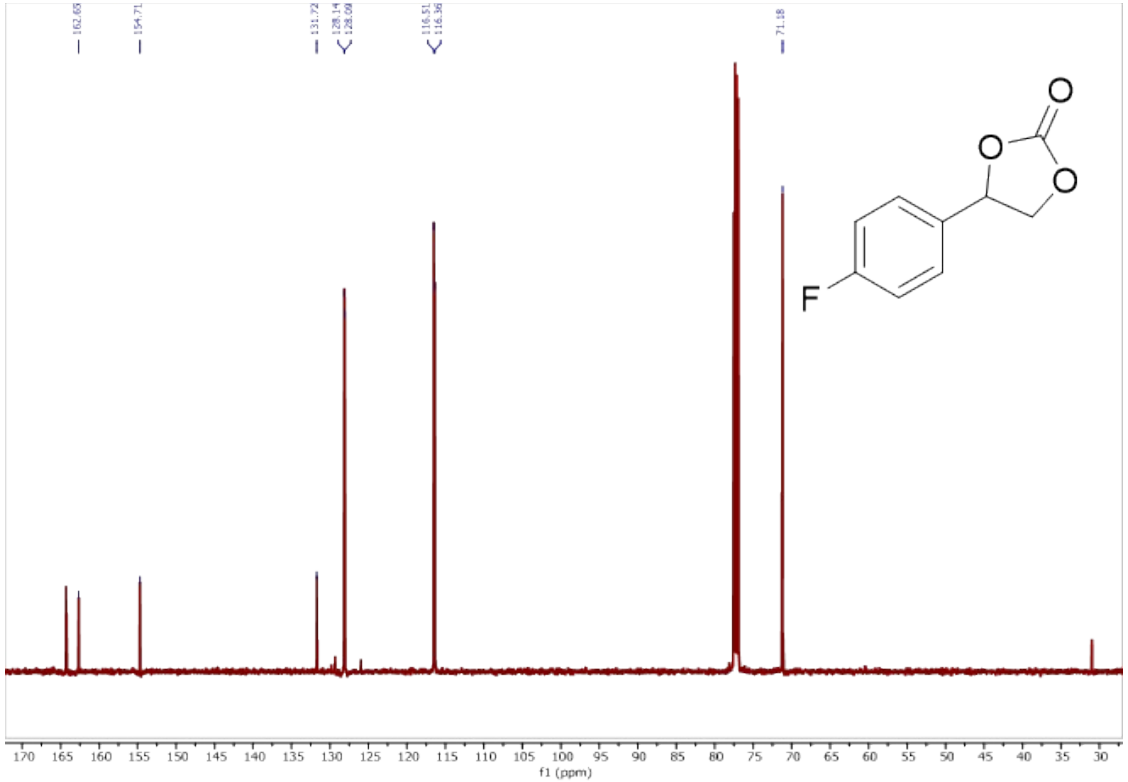


Fig. S68 ^{13}C NMR Spectrum of 4-(4-fluorophenyl)-1,3-dioxolan-2-one ($CDCl_3$, 151 MHz, 298K)

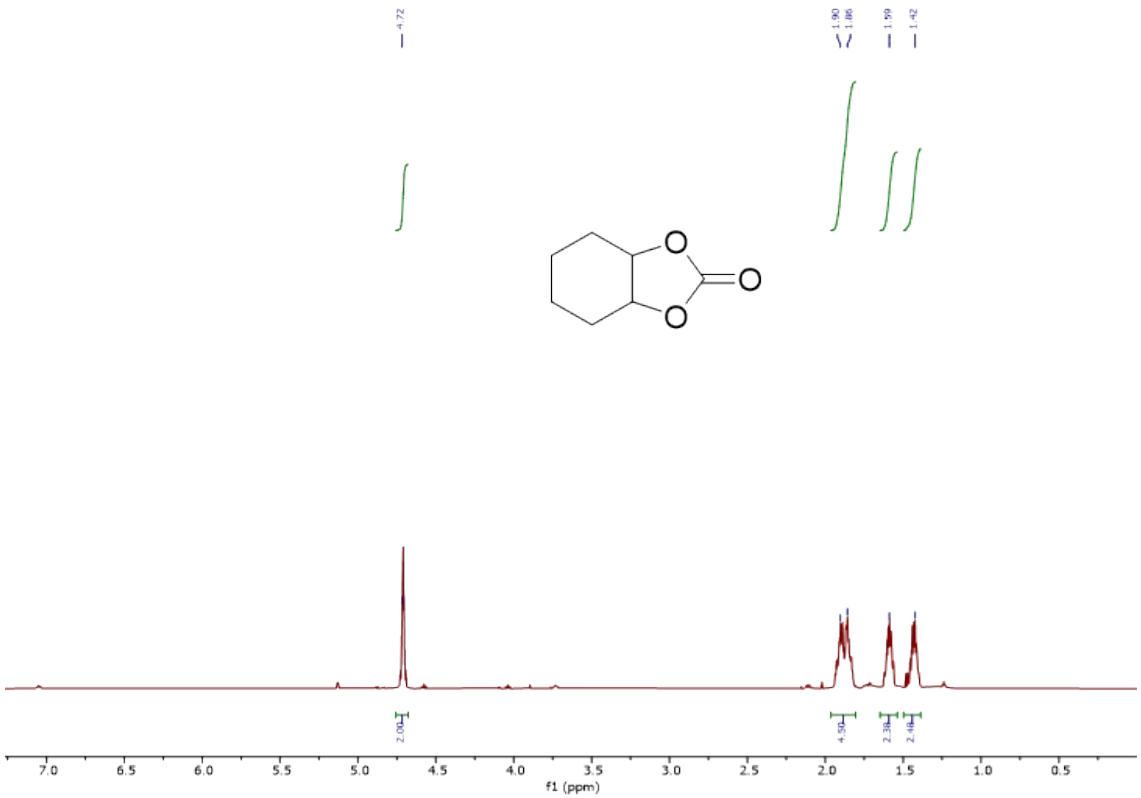


Fig. S69 ^{1}H NMR Spectrum of hexahydrobenzo[d][1,3]dioxol-2-one ($CDCl_3$, 600 MHz, 298K)

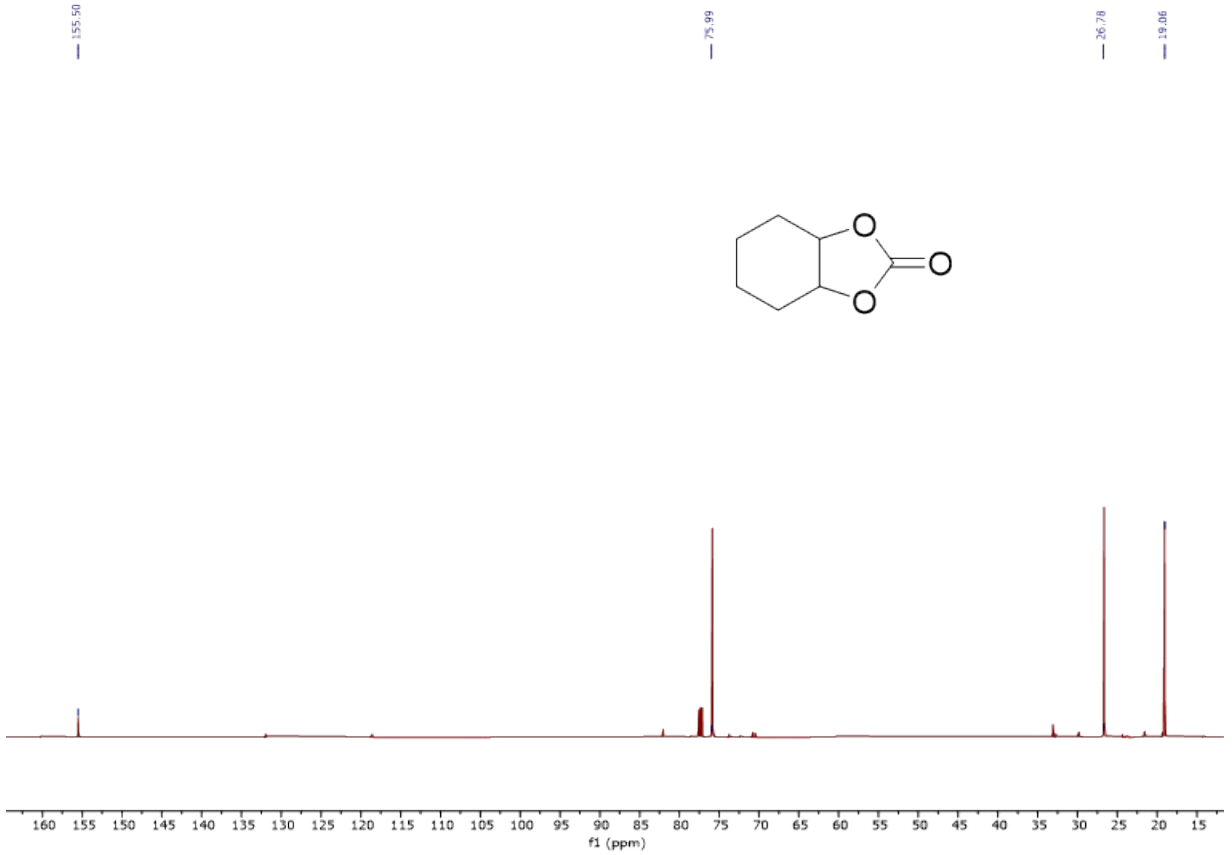


Fig. S70 ^{13}C NMR Spectrum of hexahydrobenzo[d][1,3]dioxol-2-one ($CDCl_3$, 151 MHz, 298K)

19. Equations used to calculate green metrics[4]

$$\text{AE \%} = \frac{\text{Mol wt of product} * 100}{\text{sum of Mol wts of reactants}}$$

$$\text{E-factor (E)} = \frac{\text{total mass of waste}}{\text{mass of waste final product}}$$

$$\text{MI} = \frac{\text{total mass in process}}{\text{mass product}}$$

$$\text{MI} = \frac{\text{mass of product}}{\text{mass of hazardous reactants}}$$

$$\text{Mp} = \frac{\text{mass of product} * 100}{\text{total mass (incl. sol.)}}$$

$$\text{Mp} = \frac{\text{mass of solvent}}{\text{mass of product}}$$

$$\text{Process mass intensity (PMI)} = \frac{\text{total mass in process}}{\text{mass of product}}$$

s

20. FT-IR spectra of products{Tak, 2018 #2}

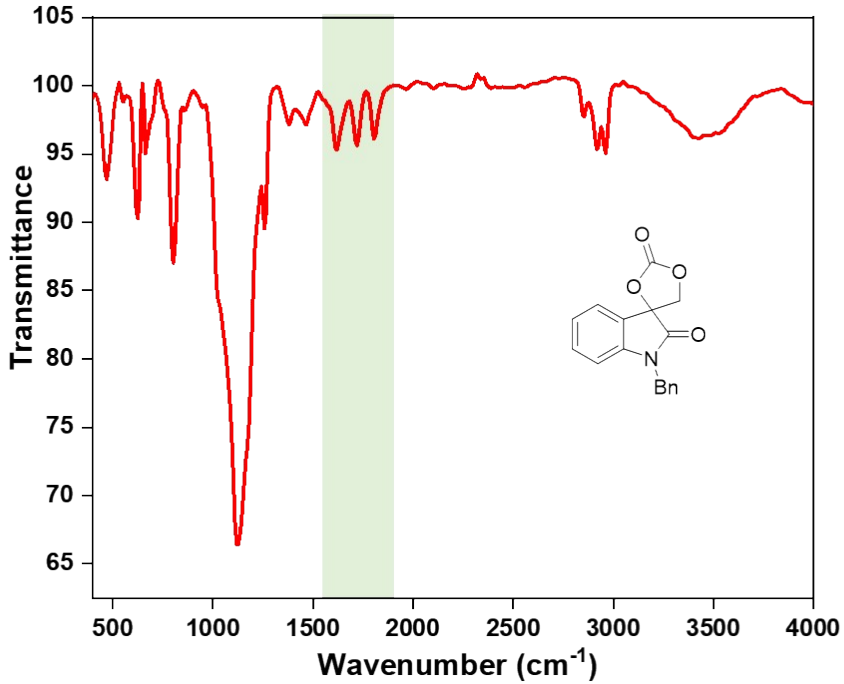


Fig.S72 FT-IR Spectrum of 1'-benzylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione.

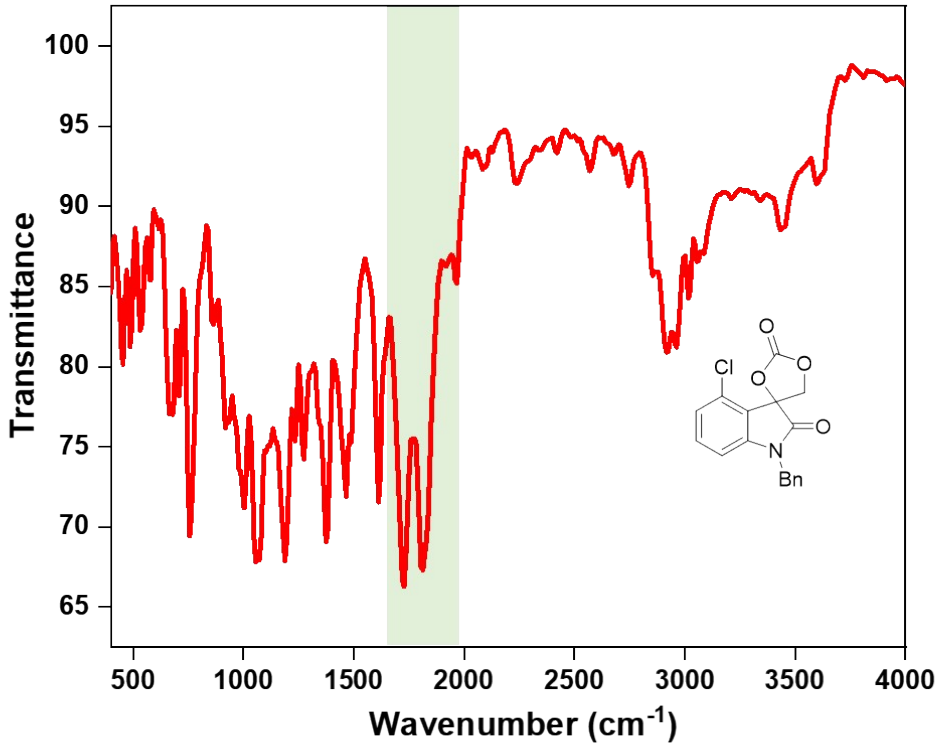


Fig.S73 FT-IR Spectrum of 1'-benzyl-4'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione.

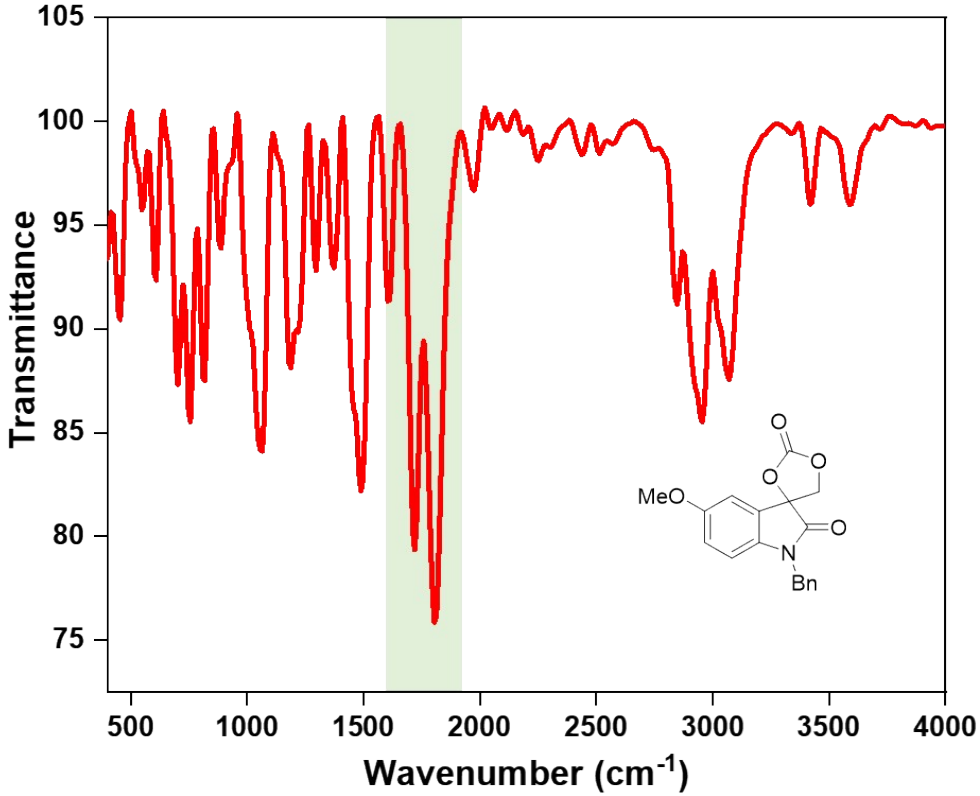


Fig.S74 FT-IR Spectrum of 1'-benzyl-5'-methoxyspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione

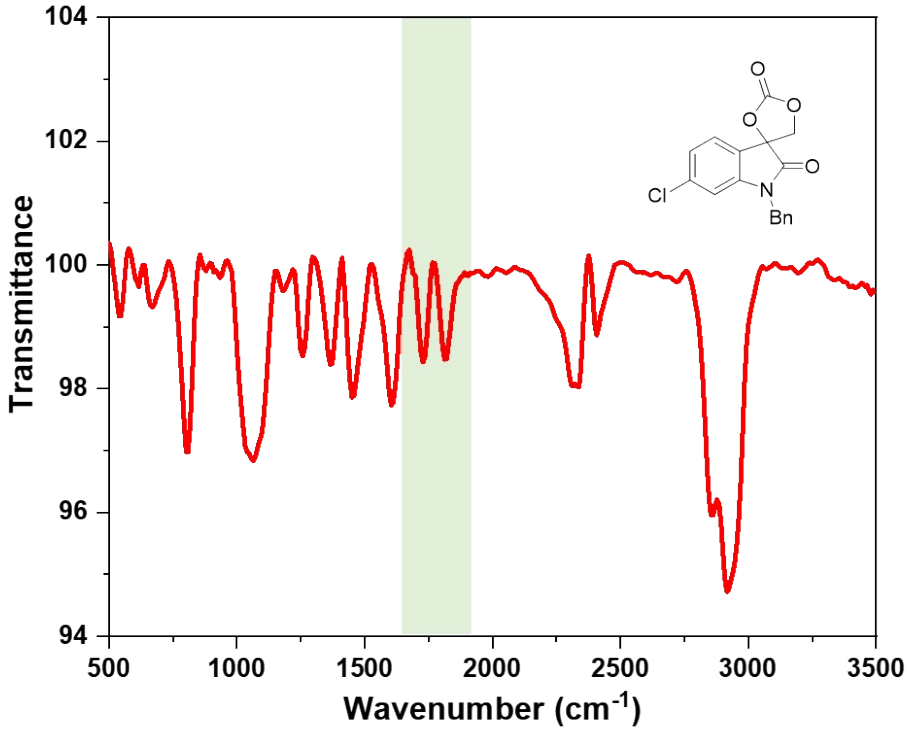


Fig.S75 FT-IR Spectrum of 1'-benzyl-6'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione.

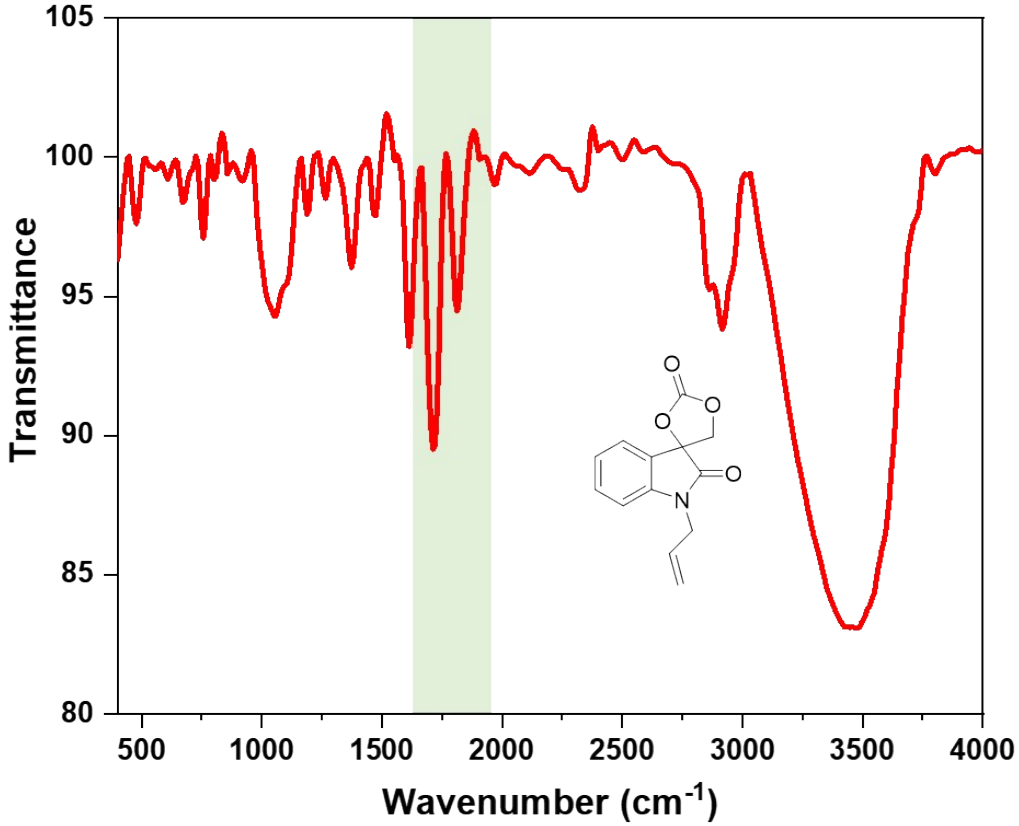


Fig.S76 FT-IR Spectrum of 1'-allylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione.

21. HRMS data of products{Parmar, 2019 #1564}

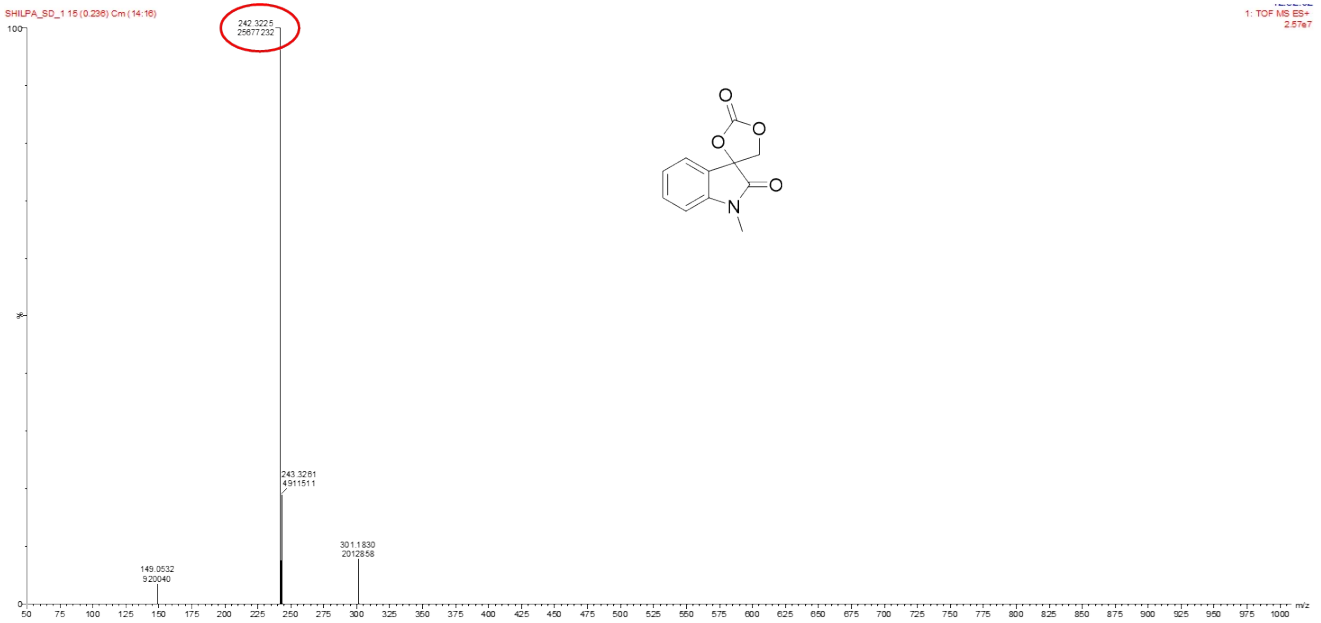


Fig.S77 HRMS Spectrum of 1'-methylspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione (Product + Na).

Fig.S78 HRMS Spectrum of 1'-benzyl-5'-chlorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione (Product + Na).

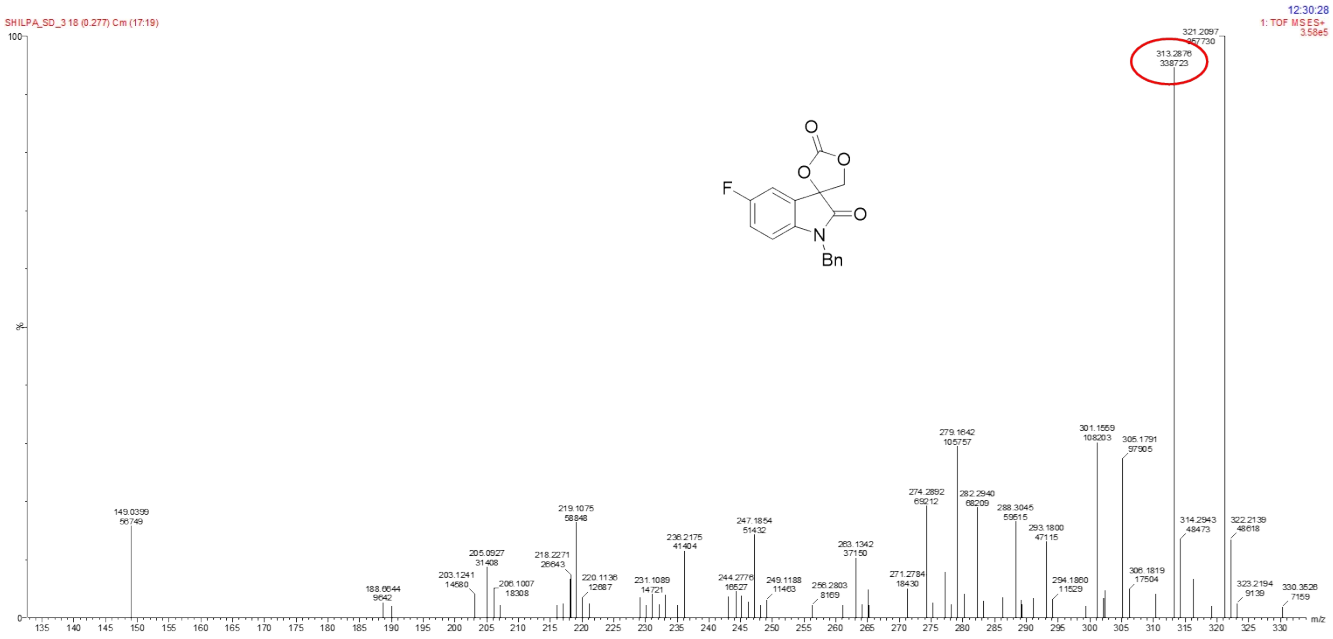


Fig.S79 HRMS Spectrum of 1'-benzyl-5'-fluorospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione (Product + Na).

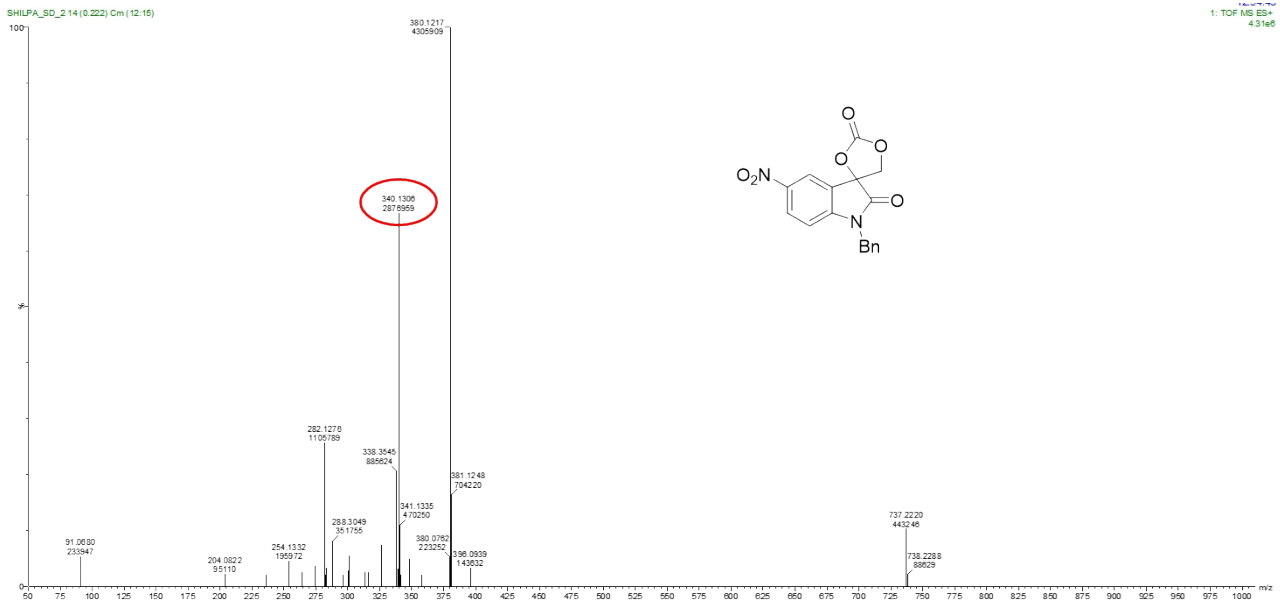


Fig.S80 HRMS Spectrum of 1'-benzyl-5'-nitrospiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione (Product + Na).

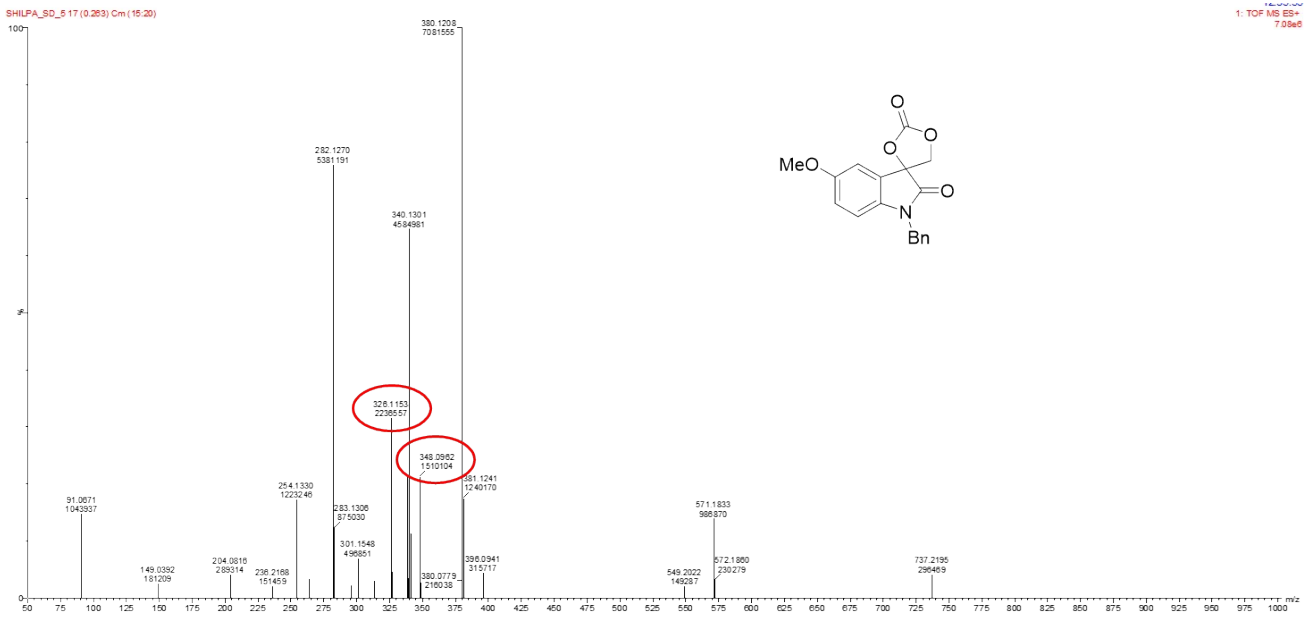


Fig.S81 HRMS Spectrum of 1'-benzyl-5'-methoxyspiro[[1,3]dioxolane-4,3'-indoline]-2,2'-dione (Product + Na and (Product + H).

22. References

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   B. Parmar, P. Patel, R. S. Pillai, R. Tak, R. I. Kureshy, N.-u. H. Khan, E. Suresh, Cycloaddition of CO2 with an Epoxide-Bearing Oxindole Scaffold by a Metal–Organic Framework-Based Heterogeneous Catalyst under Ambient Conditions, Inorg. Chem. 2019, **58**, 15, 10084–10096.
3. P. Kaur, K. Juglan, H. Kumar, M. J. J. o. C. Singla and E. Data, Volumetric and Ultrasonic Studies of PEG200, PEG400, and Ethanol–Chlorhexidine Solutions at Various Temperatures, 2023, **68**, 1123-1132.
4. R. A. Sheldon, Metrics of Green Chemistry and Sustainability: Past, Present, and Future, *ACS Sustainable Chemistry & Engineering*, 2018, **6**, 32-48.