Electronic Supplementary Material (ESI) for ChemComm.

Supporting Information

Visible Light Photoredox-Catalyzed Arylative Cyclization to Access Benzimidazo[2,1-*a*]isoquinolin-6(5*H*)-ones

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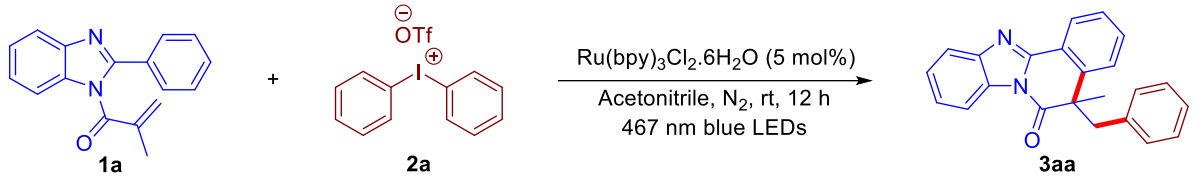
1. Table S1: Detailed optimization table.



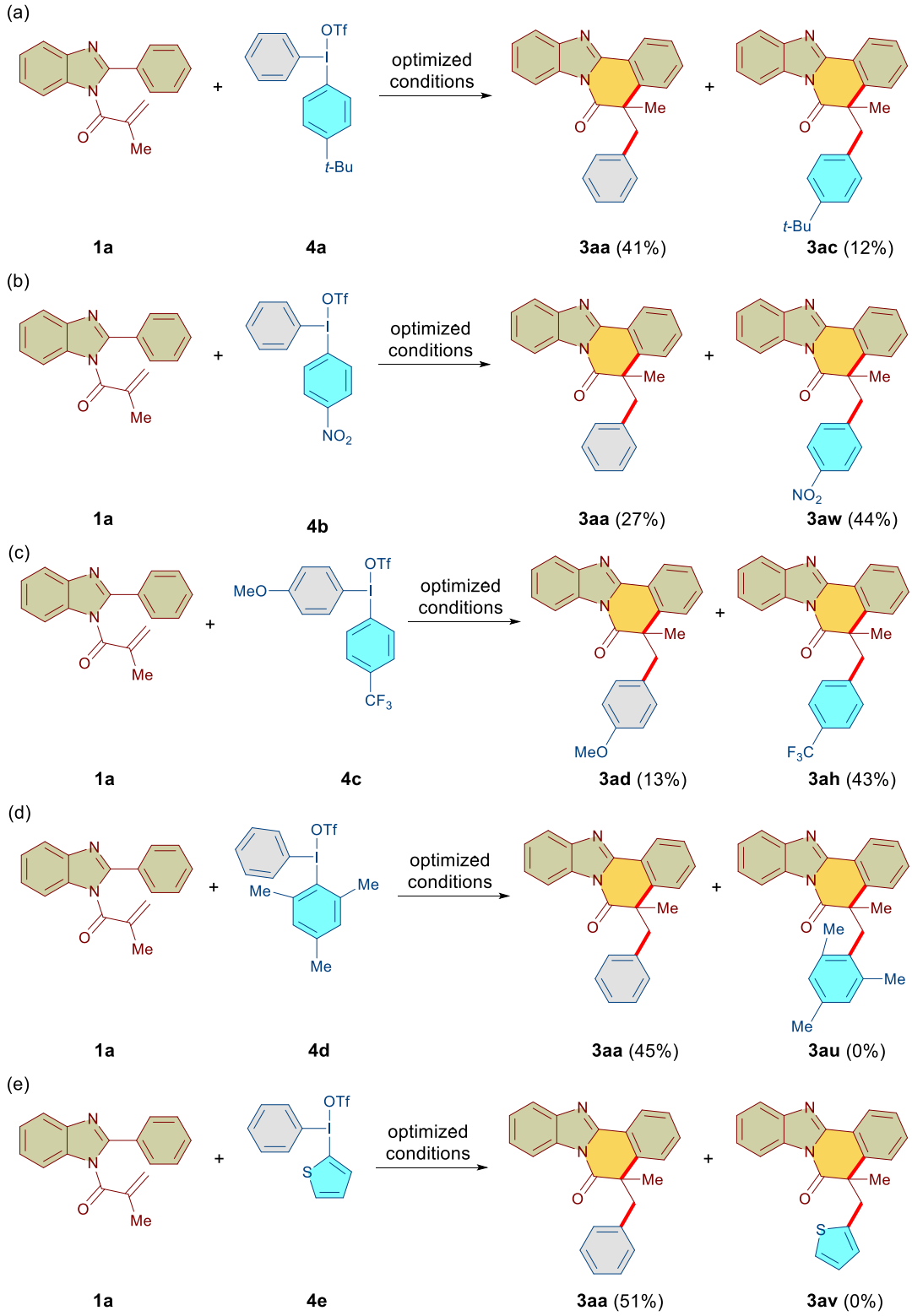
1a 2a 3aa

S. No.	Catalyst	Solvent	Yield (%)[b]
1	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	77%
2	Eosin Y	Acetonitrile	9 %
3	Eosin B	Acetonitrile	n.d
4	Fluorescein	Acetonitrile	n.d
5	Rhodamine B	Acetonitrile	n.d
6	Rose bengal	Acetonitrile	12%
7	Methylene Blue	Acetonitrile	n.d
8	Na_2Eosin Y	Acetonitrile	n.d
9	9-Mesityl-10-methylacridinium perchlorate	Acetonitrile	n.d
10	$Ru(bpy)_3Cl_2.6H_2O$	DCE	62%
12	$Ru(bpy)_3Cl_2.6H_2O$	Methanol	53%
14	$Ru(bpy)_3Cl_2.6H_2O$	Toluene	38%
16	$Ru(bpy)_3Cl_2.6H_2O$	THF	21%
17	$Ru(bpy)_3Cl_2.6H_2O$	DMF	24%
18	$Ru(bpy)_3Cl_2.6H_2O$	DMSO	30%
19	$Ru(bpy)_3Cl_2.6H_2O$	DCM	36%
20[c]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	59%
21[d]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	58%
22[e]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	74%
23[f]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	44%
24[g]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	39%
25[h]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	68%
26[i]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	75%
27[j]	----------	Acetonitrile	Trace

28[k]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	50%
29[l]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	47%
30[m]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	75%
31[n]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	55%
32[o]	$Ru(bpy)_3Cl_2.6H_2O$	Acetonitrile	n.d.

[a]Reaction Condition = **1a** (0.15 mmol), **2a** (0.225 mmol), catalyst (5 mol%), [b]Isolated yield, [c]Using 1.2 equiv **2a**, [d]Using 2.5 mol% catalyst, [e]Using 2 equiv HVI, [f]440 nm Blue LEDs was used, [g]427 nm Blue LEDs was used, [h]456 nm Blue LEDs, [i]525 nm Blue LEDs was used, [j]In the absence of photocatalyst, [k]Reaction was stirred for 6 h, [l]Under air, [m]Instead of OTf counter ion BF_4 was used, [n]Instead of OTf counter ion Br was used, [o]In the absence of light.

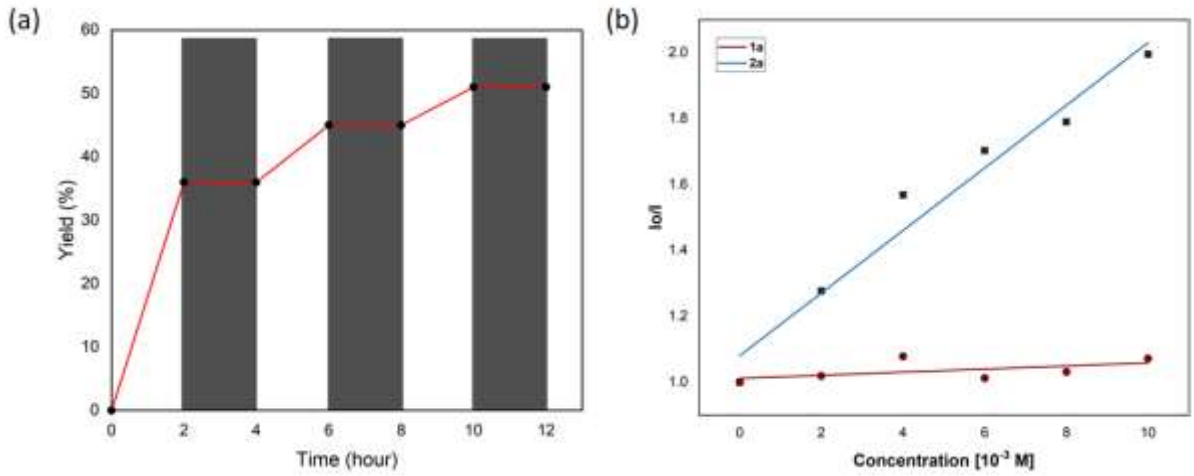
2. Scheme S1: Selectivity Studies for Unsymmetrical Diaryliodonium Triflates



We synthesized a set of unsymmetrical DAIRs (**4a**-**4e**) and reacted those with **1a** under the established conditions to evaluate if chemoselective transfer of one aryl moiety over the other is possible or not (Scheme S1). In case of DAIRs **4a**-**4c**, a selective transfer of electron-deficient aryl group providing corresponding compounds **3aa** (41%, Ph over 4-*t*-Bu-Ph), **3aw**

(44%, 4-NO_2-Ph over Ph), and **3ah** (43%, 4-CF_3-Ph over 4-OMe-Ph) was observed (Scheme S1, a-c). Expectedly, an exclusive transfer of sterically less demanding phenyl group was observed (**3aa**, 45%) when mesityliodonium salt **4d** was employed (d). Moreover, in the case of DAIR **4e**, phenyl group was transferred in exclusivity (**3aa**, 51%), and electron-rich thienyl moiety served as a dummy group

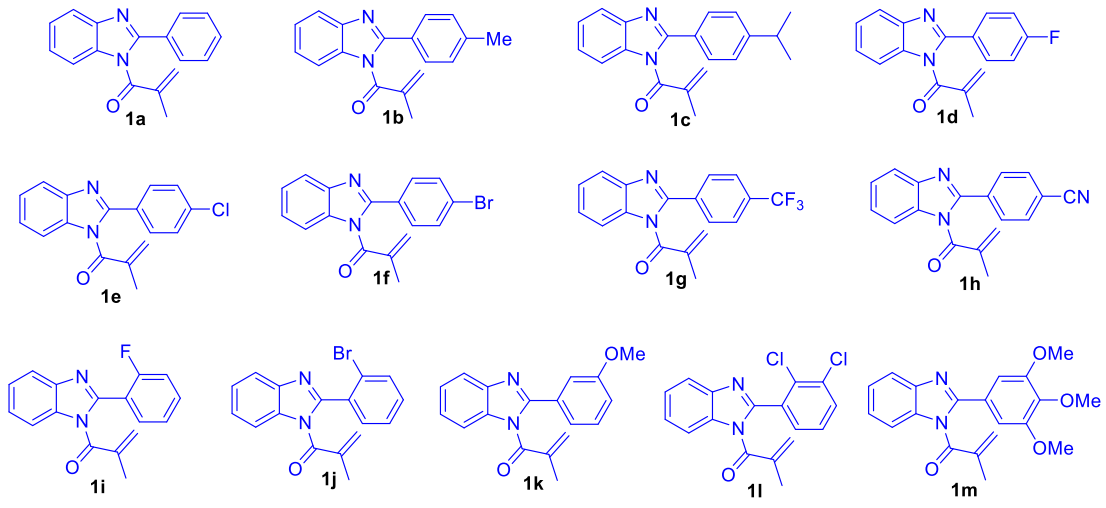
3. Figure S1: (a) Light-dark experiment. (b) Stern-Volmer plot

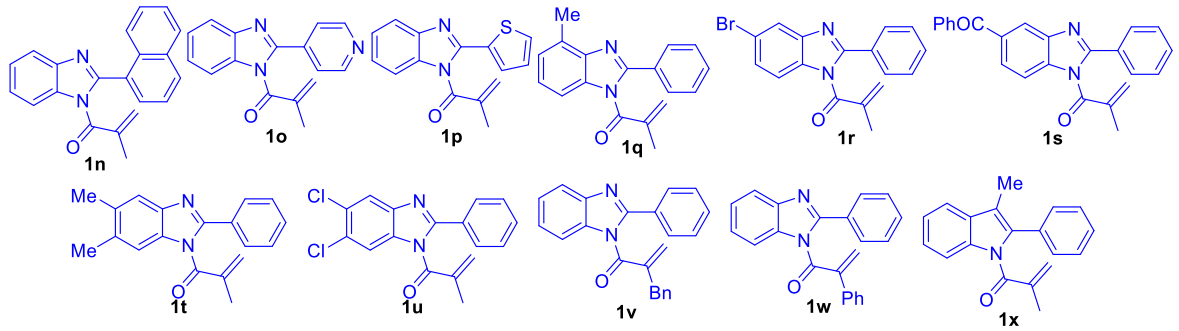


4. General Information: Photoredox reactions were performed under N_2 atmosphere for all the reaction sets, using pre-dried glassware and standard schlenk tubes. All the solvents were dried with calcium hydride and freshly distilled under nitrogen. The following starting materials and reaction components arenes, iodoarenes, diamine derivatives, aldehydes, acyl chlorides, $NaHSO_3$, DMAP, all the photocatalysts and TEMPO, BHT were obtained from commercial sources and were used without further purification. Both symmetrical and unsymmetrical diaryliodonium salts and *N*-acyl-2-arylbenzoimidazole derivatives were synthesized following the procedures given below. Yields refer to isolated compounds, estimated to be >95% pure as determined by ^{1}H NMR and ^{13}C NMR. All optimized photoredox reactions were conducted under the photo-irradiation using a 40 W Kessil blue LED (467 nm) lamp. Thin layer chromatography (TLC) was performed on Merck pre-coated silica gel 60 F_{254} aluminium sheets with detection under UV light at 254 nm. Chromatographic separations were carried out on Avra silica gel (100-200 mesh or 230-400 mesh). Nuclear magnetic resonance (NMR) spectroscopy was performed using Bruker 500 MHz spectrometers. If not otherwise specified, chemical shifts (δ) are provided in ppm. HRMS spectra were recorded using Agilent 6546 LC/Q-TOF spectrometer. The fluorescence experiment was recorded with a "JASCO FP-8300" Scientific Spectrofluorometer. UV experiment was performed on LABINDIA ANALYTICAL-2000 U UV/VIS Spectrophotometer instrument.

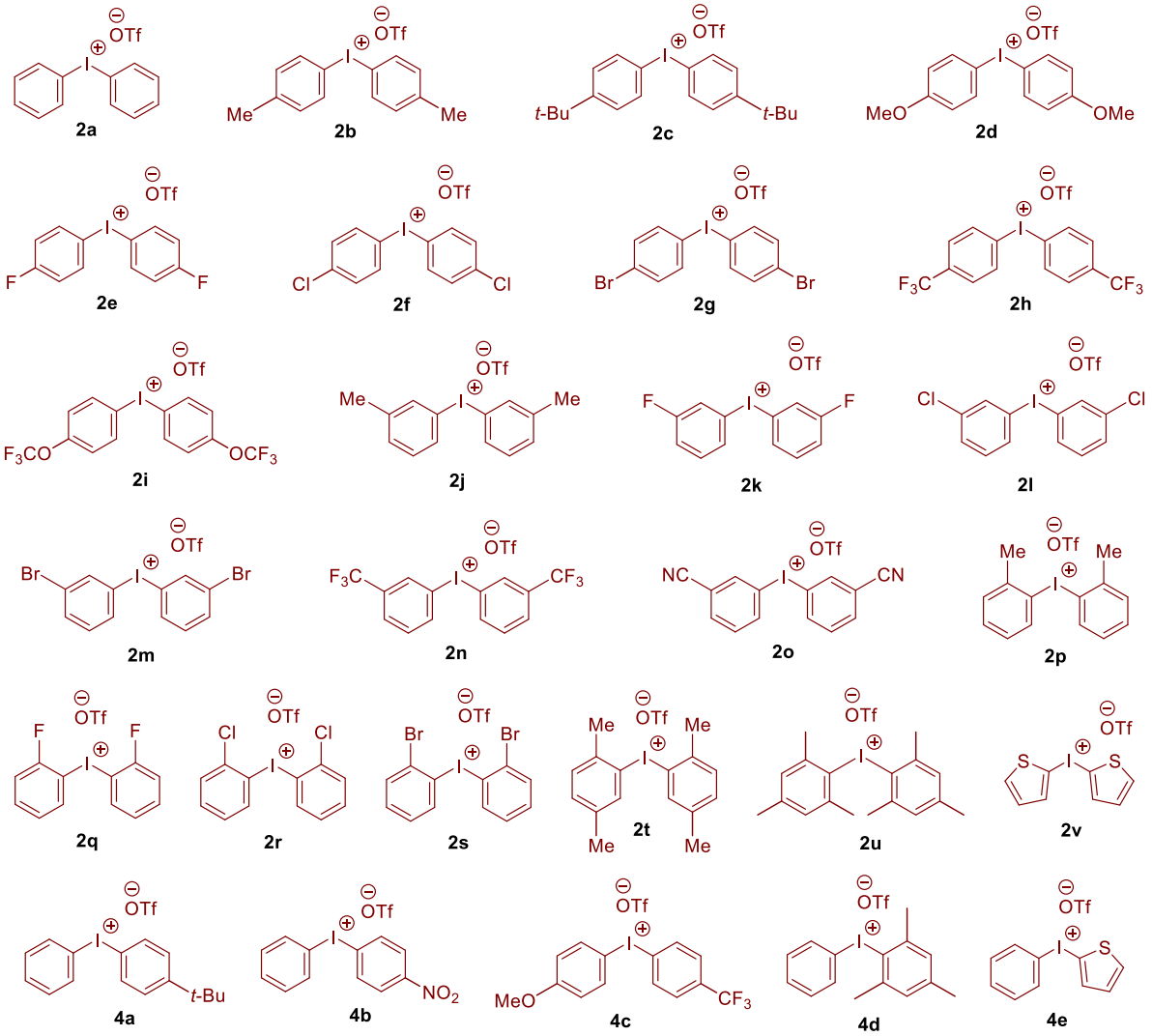
5. Preparation of Starting Materials:

Various *N*-acryloyl-2-arylbenzoimidazoles **(1a, 1b, 1c, 1d, 1e, 1g, 1h, 1n, 1t),**[1] **(1f, 1j, 1k, 1p),**[2] **1l,**[3] **1m,**[4] **(1i, 1l),**[5] **(1v, 1w),**[6] **1o**[7] were prepared following previous literature procedures and obtained characterization data were in alignment with the literature reported data.

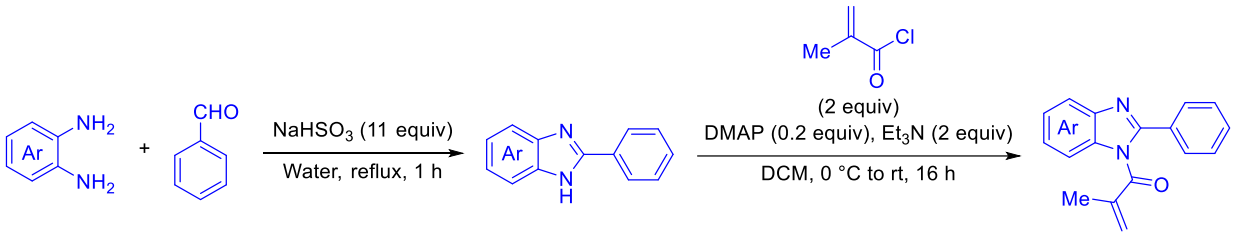




The diaryliodonium salts **(2a, 2b, 2c, 2e, 2f, 2g, 2t, 2u, 4a, 4b, 4d, 4e),**[8] **(2d, 2h, 2o),**[9] **(2i),**[10] **(2j, 2k, 2p, 2q, 2r, 2s),**[11] **2l,**[12] **2m,**[13] **2n,**[14] **2v,**[15] **4c,**[16] were prepared following previous literature procedures and obtained characterization data were in alignment with the literature reported data.



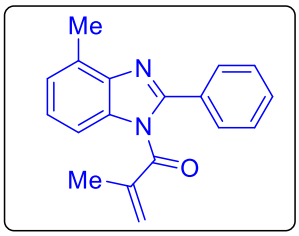
6. General Procedure (GP) for the synthesis of New *N*-acryloyl-2-arylbenzoimidazoles 1:



Step-1: In a round-bottomed flask (50 mL) equipped with a magnetic stirrer, a mixture of benzaldehyde (5.0 mmol, 1 equiv) and $NaHSO_3$ (5.73 g, 11.0 equiv) in H_2O (20 mL) was prepared. When the mixture reached refluxing temperature, phenylenediamine derivative (5.0 mmol, 1 equiv) was added. The resulting mixture was stirred for 1h. After completion of the reaction, the reaction mixture was vacuum filtered after cooling to room temperature by a glass funnel. The residues were washed by water (20 mL × 2), dried in air dry oven to give the corresponding products.

Step-2: To the solution of 2-phenyl-1*H*-benzo[d]imidazoles (1 equiv) and DMAP (0.2 equiv) in DCM (0.8 M) was added Et_3N (2 equiv) and methacryloyl chloride (2 equiv) at 0 °C. The solution was warmed up to room temperature and stirred for 16 h. The reaction was completed according to TLC analysis, and water (20 mL) was added to the mixture, which was extracted with DCM (15 mL × 3). Then the organic solvent was concentrated in vacuo and the residue was purified by flash column chromatography with 2-10% ethyl acetate/hexane as eluent to give the corresponding products.

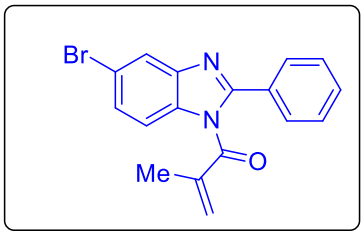
2-Methyl-1-(4-methyl-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (1q)



The compound was prepared according to GP using 4-methyl-2-phenyl-1*H*-benzo[*d*]imidazole (0.5 g, 2.4 mmol), methacryloyl chloride (0.5 g, 4.8 mmol), DMAP (0.058 g, 0.48 mmol) and Et_3N (0.49 g, 4.8 mmol). After 12 h, purification by column chromatography (0-10% ethyl acetate in hexane) gave **1q** as a white solid (0.48 g, 72%).

^{1}H NMR (400 MHz, $CDCl_3$) δ 7.58 (dd, *J* = 6.5, 2.8 Hz, 2H), 7.55 (d, *J* = 8.1 Hz, 1H), 7.44 – 7.40 (m, 3H), 7.24 (t, *J* = 7.8 Hz, 1H), 7.17 (d, *J* = 7.3 Hz, 1H), 5.53 (s, 1H), 5.32 (s, 1H), 2.71 (s, 3H), 1.97 (s, 3H). **^{13}C{^{1}H} NMR** (101 MHz, $CDCl_3$) δ 170.5, 152.9, 142.1, 140.4, 134.4, 131.8, 130.4, 123.0, 129.0, 128.8, 128.5, 125.0, 124.7, 110.3, 18.2, 16.9. **HRMS-ESI** (m/z): calcd for $C_{18}H_{17}N_2O$ $[M + H]^+$ 277.1335; found 277.1351.

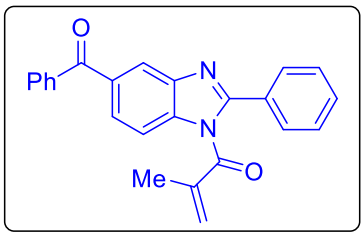
1-(5-Bromo-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (1r)



The compound was prepared according to GP using 5-Bromo-2-phenyl-1*H*-benzo[*d*]imidazole (0.5 g, 2.88 mmol), methacryloyl chloride (0.60 g, 5.76 mmol), DMAP (0.071 g, 0.58 mmol) and Et_3N (0.58 g, 5.76 mmol). After 12 h, purification by column chromatography (0-10% ethyl acetate in hexane) gave **1q** as a white solid (0.78 g, 79%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 7.83 (d, *J* = 1.7 Hz, 1H), 7.62 (d, *J* = 8.5 Hz, 1H), 7.52 (dd, *J* = 6.3, 2.8 Hz, 2H), 7.44 – 7.41 (m, 1H), 7.39 (dd, *J* = 5.1, 1.5 Hz, 3H), 5.52 (d, *J* = 0.9 Hz, 1H), 5.25 (s, 1H), 1.93 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 169.9, 154.1, 141.8, 140.1, 135., 131.0, 130.3, 129.0, 128.9, 128.8, 127.9, 121.4, 118.0, 115.9, 18.1. **HRMS-ESI** (m/z): calcd for $C_{17}H_{14}BrN_2O$ $[M + H]^+$ 341.0284; found 341.0290.

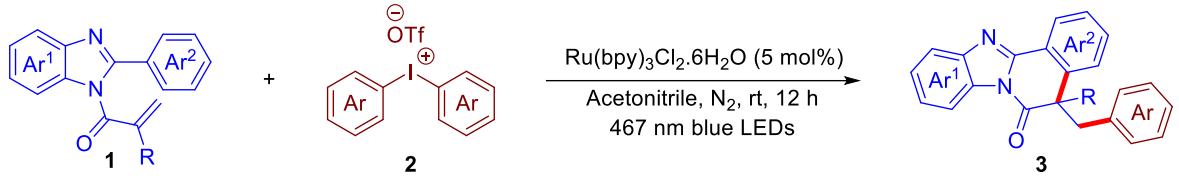
1-(5-Benzoyl-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (1s)



The compound was prepared according to GP using phenyl(2-Phenyl-1*H*-benzo[*d*]imidazol-5-yl)methanone (0.5 g, 1.68 mmol), methacryloyl chloride (0.35 g, 3.36 mmol), DMAP (0.041 g, 0.34 mmol) and Et_3N (0.34 g, 3.36 mmol). After 12 h, purification by column chromatography (0-10% ethyl acetate in hexane) gave **1q** as a white solid (0.46 g, 75%).

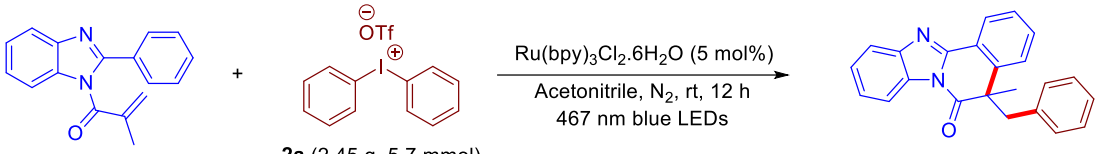
^{1}H NMR (500 MHz, $CDCl_3$) δ 8.18 (s, 1H), 7.84 (d, *J* = 8.5 Hz, 1H), 7.75 (d, *J* = 8.1 Hz, 2H), 7.70 (d, *J* = 8.5 Hz, 1H), 7.55 – 7.52 (m, 2H), 7.51 – 7.47 (m, 1H), 7.38 (dd, *J* = 11.7, 6.1 Hz, 5H), 5.55 (s, 1H), 5.29 (s, 1H), 1.93 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 196.2, 169.9, 155.1, 142.2, 140.1, 137.9, 137.5, 134.1, 132.4, 130.8, 130.5, 130.1, 129.4, 128.9, 128.8, 128.3, 126.7, 123.2, 112.7, 18.1. **HRMS-ESI** (m/z): calcd for $C_{24}H_{19}N_2O_2$ $[M + H]^+$ 367.1441; found 367.1436.

7. General Procedure (GP) for the visible light-mediated photoredox-catalyzed cascade cyclization of *N*-acryloyl-2-arylbenzoimidazoles 1 with diaryliodonium salts 2:



N-acryloyl-2-arylbenzoimidazole derivatives **1** (0.25 mmol, 1.0 equiv), diaryliodonium trifluoromethanesulfonates **2** (0.375 mmol, 1.5 equiv) and $Ru(bpy)_3Cl_2 \cdot 6H_2O$ (0.0125 mmol, 5 mol%) were added in a pre-dried 10 ml Schlenk tube under N_2 atmosphere. The tube was degassed and purged with N_2 three times. Then dry acetonitrile (2.5 mL) was added under the N_2 atmosphere and the mixture was allowed to stir for 12 h under irradiation of 40 W Kessil blue LED (467 nm) lamp. After completion, the reaction mixture was concentrated under vacuum, and purified by silica gel column chromatography using 0-5% ethyl acetate in hexane to afford arylated-benzimidazo[2,1-*a*]isoquinolin-6(5*H*)-ones **3**.

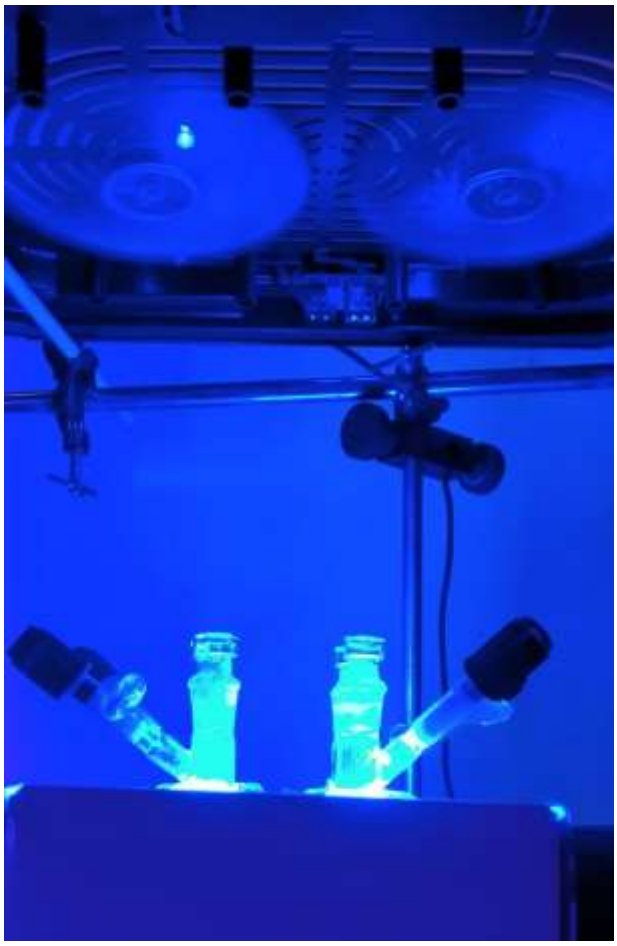
8. Gram scale synthesis of 5-Benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one 3aa

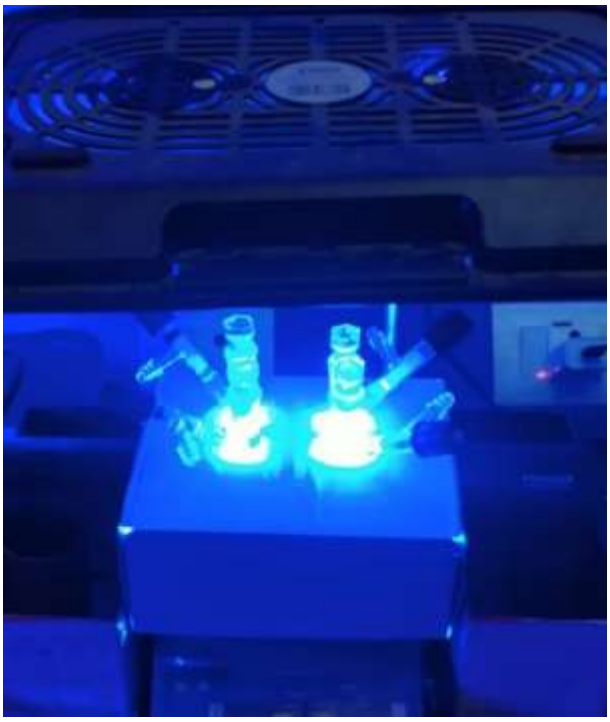


1a (1 g, 3.8 mmol) **2a** (2.45 g, 5.7 mmol) **3aa** (0.91 g, 71%)

2-Methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one **1a** (1 g, 3.8 mmol, 1.0 equiv), diphenyliodonium trifluoromethanesulfonate **2a** (2.45 g, 5.7 mmol, 1.5 equiv) and $Ru(bpy)_3Cl_2.6H_2O$ (187 mg, 0.25 mmol, 5 mol%) were added in a pre-dried 50 mL Schlenk fask under N_2 atmosphere. The flask was degassed and purged with N_2 three times. Then dry acetonitrile (15 mL) were added under the N_2 atmosphere. Then the mixture was allowed to stir for 12 h under irradiation of 40 W Kessil blue LED (467 nm) lamp. After completion, the reaction mixture was concentrated under vacuum, and purified by silica gel column chromatography using 0-5% ethyl acetate in hexane to afford the 5-Benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one **3aa** (71%).

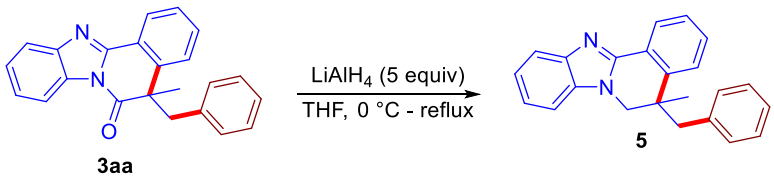
9. Image of setup for Photoredox-catalyzed reaction:





10. Post-Synthetic Modifications:

a)

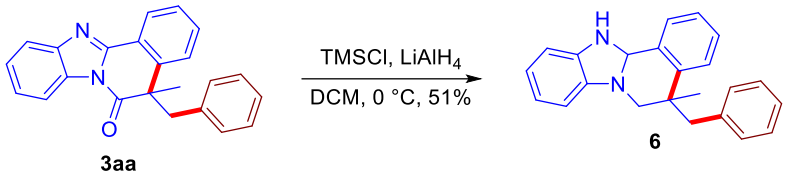


3aa **5**

$LiAlH_4$ (0.028 g, 5 equiv) was slowly added to a solution of 5-Benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one **3aa** (0.051 g, 0.15 mmol) in dry THF (3 ml) at 0 ^oC and the resulting reaction mixture was refluxed for 5h. Following which, the solution was cooled to 0 ^oC, EtOAc (10 ml) was added and product was extracted. The reaction mixture was concentrated and purified by column chromatography using silica gel. The product 5-Benzyl-5-methyl-5,6-dihydrobenzo[4,5]imidazo[2,1-*a*]isoquinoline **5** was isolated as a white solid (0.016 g, 34%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.42 (dd, *J* = 7.2, 1.7 Hz, 1H), 7.92 – 7.88 (m, 1H), 7.50 – 7.44 (m, 2H), 7.37 – 7.32 (m, 4H), 7.25 – 7.20 (m, 3H), 6.83 (dd, *J* = 7.4, 1.9 Hz, 2H), 4.21 (d, *J* = 12.6 Hz, 1H), 3.83 (d, *J* = 12.6 Hz, 1H), 2.76 (s, 2H), 1.53 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 142.60, 142.58, 136.27, 130.65, 130.62, 128.16, 128.14, 128.06, 126.98, 126.97, 126.95, 125.42, 125.41, 125.40, 112.02, 109.41, 109.39, 49.12, 46.65, 39.16, 22.23. **HRMS-ESI** (m/z): calcd for $C_{23}H_{21}N_2$ $[M + H]^+$ 325.1699; found 325.1742.

b)



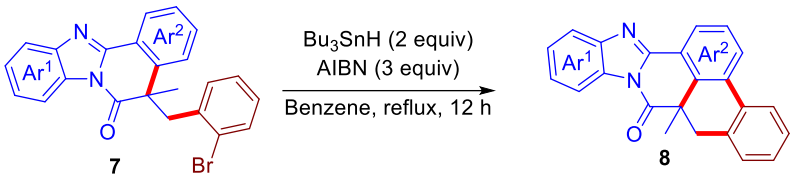
3aa **6**

5-Benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one **3aa** (0.051 g, 0.15 mmol) and DCM (2 ml) were charged into a round bottomed flask and stirred under nitrogen atmosphere for 10-15 minutes and cooled to 0-5˚C. Trimethylsilyl chloride (0.18 mmol, 1.2 equiv) was then added to the mixture over 10-15 minutes and stirred for 10-15 minutes at same temperature. Lithium aluminum hydride (0.008 g, 0.21 mmol, 1.4 equiv) in THF solution is added dropwise at 0˚C. After complete addition, the stirring was continued for 1-2 hours. After completion of reaction (TLC), the reaction was quenched by slow dropwise addition of 2M sodium hydroxide and stirred for 10-15 minutes. Then, EtOAc (10 ml) was

added and the product was extracted. The reaction mixture was concentrated and purified by column chromatography using silica gel. The product 5-benzyl-5-methyl-5,6,12,12a-tetrahydrobenzo[4,5]imidazo[2,1-*a*]isoquinoline **6** was isolated as a white solid (0.025 g, 51%).

^{1}H NMR (400 MHz, DMSO) δ 8.18 (d, *J* = 7.2 Hz, 1H), 7.69 (d, *J* = 7.9 Hz, 1H), 7.54 (d, *J* = 7.9 Hz, 1H), 7.44 – 7.41 (m, 1H), 7.39 (td, *J* = 7.6, 1.4 Hz, 1H), 7.30 (t, *J* = 7.0 Hz, 1H), 7.24 (t, *J* = 7.6 Hz, 1H), 7.16 (d, *J* = 7.5 Hz, 1H), 7.12 – 7.07 (m, 3H), 6.74 (d, *J* = 5.7 Hz, 1H), 6.61 (d, *J* = 6.4 Hz, 2H), 5.69 (d, *J* = 6.0 Hz, 1H), 3.29 (s, 2H), 2.60 (d, *J* = 13.2 Hz, 1H), 2.52 (d, *J* = 13.2 Hz, 1H), 1.48 (s, 3H). **^{13}C{^{1}H} NMR** (101 MHz, DMSO) δ 147.9, 144.2, 141.3, 136.9, 134.4, 130.9, 130.7, 128.2, 127.7, 127.5, 127.0, 125.4, 125.4, 123.1, 122.9, 119.6, 110.7, 79.8, 47.0, 44.6, 20.0. **HRMS-ESI** (m/z): calcd for $C_{23}H_{22}N_2$ $[M]^+$ 326.1783; found 326.1750.

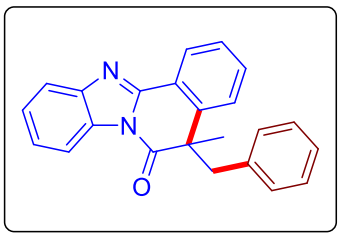
c) Homolytic Aromatic Substitution (HAS):



A solution of Bu_3SnH (2 equiv) and AIBN (3 equiv) in benzene was added to 2-arylated-benzimidazo[2,1-*a*]isoquinolin-6(5*H*)-ones derivatives **7** (0.15 mmol) in benzene at reflux under an atmosphere of nitrogen over 8 h by using a syringe pump. After completion of addition, the reaction was stirred for an additional 4 h at the same temperature. After cooling to room temperature, the mixture was evaporated to dryness to afford a yellow oil, which was re-dissolved in ethyl acetate. Saturated aqueous potassium fluoride solution was added and the mixture stirred vigorously for 15 min. The resultant solution was filtered and extracted with EtOAc. The organic fractions were dried and evaporated to dryness to yield a crude yellow oil. The oil was purified by column chromatography using light 20-40% DCM/hexane as eluent to give the product **8**.

11. Report of NMR Spectra:

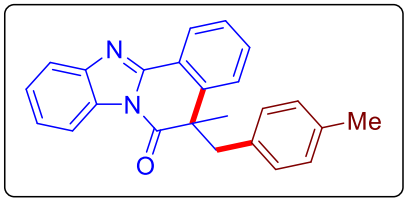
5-Benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3aa)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3aa** as a white solid (0.065 g, 77%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 – 8.34 (m, 1H), 8.30 (d, *J* = 7.5 Hz, 1H), 7.72 – 7.69 (m, 1H), 7.65 – 7.60 (m, 2H), 7.49 (ddd, *J* = 8.2, 6.4, 2.1 Hz, 1H), 7.44 – 7.39 (m, 2H), 6.92 (t, *J* = 7.4 Hz, 1H), 6.82 (t, *J* = 7.6 Hz, 2H), 6.55 (d, *J* = 7.2 Hz, 2H), 3.61 – 3.56 (m, 1H), 3.20 (d, *J* = 13.1 Hz, 1H), 1.96 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.6, 149.5, 143.8, 140.7, 135.0, 131.5, 131.0, 129.0, 127.8, 127.8, 127.0, 126.6, 125.7, 125.64, 125.4, 123.6, 119.6, 115.4, 51.1, 50.8, 26.0.

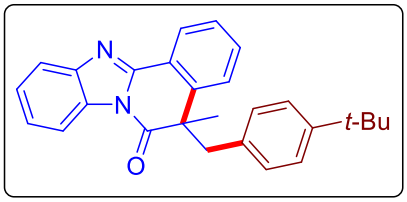
5-Methyl-5-(4-methylbenzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ab)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), di-*p*-tolyl-iodonium trifluoromethanesulfonate (0.172 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ab** as a white solid (0.053 g, 60%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 – 8.34 (m, 1H), 8.33 – 8.30 (m, 1H), 7.74 – 7.70 (m, 1H), 7.64 – 7.59 (m, 2H), 7.48 (ddd, *J* = 8.3, 6.7, 1.8 Hz, 1H), 7.44 – 7.39 (m, 2H), 6.64 (d, *J* = 7.8 Hz, 2H), 6.46 (d, *J* = 8.0 Hz, 2H), 3.55 (d, *J* = 13.2 Hz, 1H), 3.17 (d, *J* = 13.2 Hz, 1H), 2.05 (s, 3H), 1.93 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.8, 149.6, 143.8, 140.9, 136.6, 131.9, 131.5, 131.1, 128.9, 128.5, 127.8, 126.7, 125.7, 125.6, 125.4, 123.5, 119.6, 115.5, 51.1, 50.2, 26.2, 20.9.

5-(4-(Tert-butyl)benzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ac)

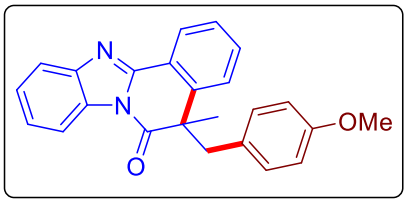


The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1H-benzo[d]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(4-(tert-

butyl)phenyl)iodonium trifluoromethanesulfonate (0.203 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ac** as a white solid (0.055 g, 56%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.34 – 8.29 (m, 2H), 7.70 – 7.66 (m, 1H), 7.65 – 7.60 (m, 2H), 7.51 – 7.47 (m, 1H), 7.43 – 7.36 (m, 2H), 6.83 – 6.78 (m, 2H), 6.48 – 6.43 (m, 2H), 3.51 (d, *J* = 13.0 Hz, 1H), 3.16 (d, *J* = 13.1 Hz, 1H), 1.95 (s, 3H), 1.05 (s, 9H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.7, 149.9, 149.5, 143.7, 140.8, 131.8, 131.5, 131.1, 128.6, 127.8, 126.5, 125.6, 125.5, 125.3, 124.6, 123.7, 119.5, 115.3, 51.2, 50.6, 34.1, 31.0, 25.6. **HRMS-ESI** (m/z): calcd for $C_{27}H_{27}N_2O$ $[M + H]^+$ 395.2118; found 395.2124.

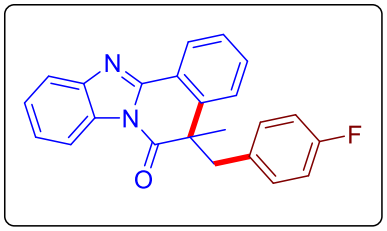
5-(4-Methoxybenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ad)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(4-methoxyphenyl)iodonium trifluoromethanesulfonate (0.184 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ad** as a white solid (0.046 g, 50%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.37 – 8.34 (m, 1H), 8.31 (dd, *J* = 7.9, 1.0 Hz, 1H), 7.73 – 7.70 (m, 1H), 7.65 – 7.61 (m, 1H), 7.59 (dd, *J* = 7.9, 1.1 Hz, 1H), 7.48 (ddd, *J* = 8.3, 7.0, 1.5 Hz, 1H), 7.44 – 7.39 (m, 2H), 6.49 – 6.46 (m, 2H), 6.37 – 6.34 (m, 2H), 3.55 (s, 3H), 3.52 (d, *J* = 13.3 Hz, 1H), 3.14 (d, *J* = 13.3 Hz, 1H), 1.93 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.8, 158.5, 149.6, 143.8, 140.8, 131.5, 131.1, 130.1, 127.8, 127.1, 126.6, 125.7, 125.7, 125.4, 123.5, 119.6, 115.4, 113.2, 55.0, 51.2, 50.0, 25.9. **HRMS-ESI** (m/z): calcd for $C_{24}H_{21}N_2O_2$ $[M + H]^+$ 369.1598; found 369.1617.

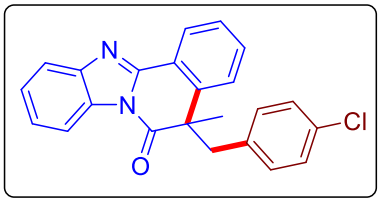
5-(4-Fluorobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ae)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(4-fluorophenyl)-iodonium trifluoromethanesulfonate (0.174 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ae** as a white solid (0.042 g, 47%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.30 (m, 2H), 7.74 – 7.71 (m, 1H), 7.65 – 7.59 (m, 2H), 7.49 (ddd, *J* = 8.3, 7.0, 1.5 Hz, 1H), 7.43 – 7.40 (m, 2H), 6.52 (d, *J* = 7.1 Hz, 4H), 3.56 (d, *J* = 13.2 Hz, 1H), 3.16 (d, *J* = 13.3 Hz, 1H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.5, 161.8 (d, *J* = 246.0 Hz), 149.4, 143.8, 140.4, 131.6, 131.0, 130.9 (d, *J* = 3.3 Hz), 130.5 (d, *J* = 8.1 Hz), 128.0, 126.6, 125.8 (d, *J* = 12.9 Hz), 125.5, 123.5, 119.8, 115.4, 114.8, 114.6, 51.1, 49.8, 26.1. **^{19}F{^{1}H} NMR** (471 MHz, $CDCl_3$) δ -115.2. **HRMS-ESI** (m/z): calcd for $C_{23}H_{18}FN_2O$ $[M + H]^+$ 357.1398; found 357.1447.

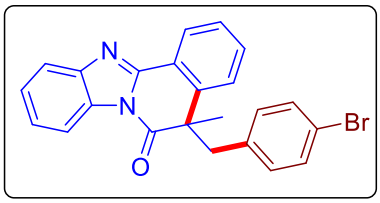
5-(4-Chlorobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3af)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(4-chlorophenyl)-iodonium trifluoromethanesulfonate (0.187 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3af** as a white solid (0.060 g, 64%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.32 (m, 2H), 7.76 – 7.72 (m, 1H), 7.62 (dtd, *J* = 8.9, 8.0, 1.2 Hz, 2H), 7.52 – 7.48 (m, 1H), 7.45 – 7.40 (m, 2H), 6.84 – 6.80 (m, 2H), 6.54 – 6.49 (m, 2H), 3.57 (d, *J* = 13.2 Hz, 1H), 3.18 (d, *J* = 13.2 Hz, 1H), 1.93 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.4, 149.3, 143.8, 140.3, 133.7, 133.0, 131.6, 130.9, 130.4, 128.0, 126.6, 125.9, 125.8, 125.6, 123.4, 119.8, 115.4, 50.9, 49.5, 26.6.

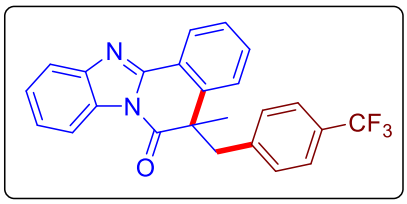
5-(4-Bromobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ag)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(4-bromophenyl)-iodonium trifluoromethanesulfonate (0.220 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ag** as a white solid (0.065 g, 62%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.34 (dd, *J* = 6.0, 2.5 Hz, 2H), 7.77 – 7.73 (m, 1H), 7.65 – 7.61 (m, 1H), 7.61 – 7.58 (m, 1H), 7.51 – 7.47 (m, 1H), 7.45 – 7.41 (m, 2H), 6.99 – 6.95 (m, 2H), 6.49 – 6.44 (m, 2H), 3.55 (d, *J* = 13.2 Hz, 1H), 3.17 (d, *J* = 13.2 Hz, 1H), 1.92 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.3, 149.3, 143.8, 140.3, 134.2, 131.6, 131.0, 130.9, 130.7, 128.1, 126.6, 125.9, 125.9, 125.6, 123.4, 121.2, 119.8, 115.4, 50.9, 49.4, 26.7.

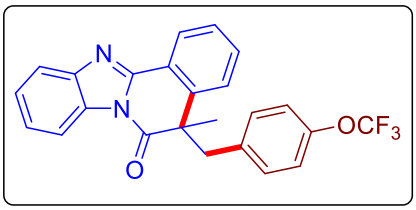
5-Methyl-5-(4-(trifluoromethyl)benzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ah)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(4-(trifluoromethyl)phenyl)-iodonium trifluoromethanesulfonate (0.212 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ah** as a white solid (0.051 g, 50%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.32 (m, 2H), 7.74 – 7.72 (m, 1H), 7.67 – 7.61 (m, 2H), 7.51 (ddd, *J* = 8.2, 6.9, 1.6 Hz, 1H), 7.44 – 7.41 (m, 2H), 7.11 (d, *J* = 8.1 Hz, 2H), 6.72 (d, *J* = 8.1 Hz, 2H), 3.66 (d, *J* = 13.2 Hz, 1H), 3.27 (d, *J* = 13.2 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.2, 149.2, 143.7, 140.1, 139.3 (d, *J* = 0.9 Hz), 131.7, 130.9, 129.4, 128.2, 127.5, 126.5, 126.0, 125.9, 125.7, 124.8 (q, *J* = 3.7 Hz), 123.8 (q, *J* = 272.1 Hz), 123.3, 119.8, 115.4, 50.9, 49.5, 26.9. **^{19}F{^{1}H} NMR** (471 MHz, $CDCl_3$) δ -63.2. **HRMS-ESI** (m/z): calcd for $C_{24}H_{17}F_3N_2O$ $[M]^+$ 406.1293; found 406.1269.

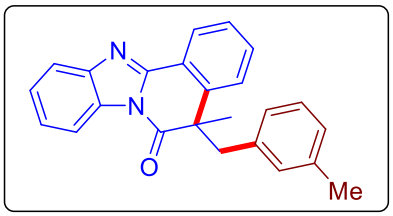
5-Methyl-5-(4-(trifluoromethoxy)benzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ai)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(4-(trifluoromethoxy)phenyl)-iodonium trifluoromethanesulfonate (0.224 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ai** as a white solid (0.052 g, 49%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.33 (dd, *J* = 6.1, 1.8 Hz, 2H), 7.74 – 7.70 (m, 1H), 7.65 – 7.59 (m, 2H), 7.52 – 7.48 (m, 1H), 7.43 – 7.40 (m, 2H), 6.68 (d, *J* = 8.1 Hz, 2H), 6.59 – 6.56 (m, 2H), 3.58 (d, *J* = 13.2 Hz, 1H), 3.19 (d, *J* = 13.2 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.3, 149.2, 148.2, 143.8, 140.2, 133.8, 131.6, 130.9, 130.3, 128.1, 126.5, 125.9, 125.8, 125.6, 123.6, 122.2 (q, *J* = 256.8 Hz),120.2, 119.8, 115.3, 51.0, 49.6, 26.2. **^{19}F{^{1}H} NMR** (471 MHz, $CDCl_3$) δ -58.1. **HRMS-ESI** (m/z): calcd for $C_{24}H_{18}F_3N_2O_2$ $[M + H]^+$ 423.1315; found 423.1301.

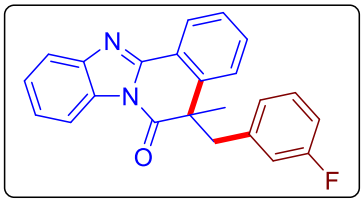
5-Methyl-5-(4-methylbenzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3aj)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), di-p-tolyl-iodonium trifluoromethanesulfonate (0.171 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3aj** as a white solid (0.062 g, 70%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 – 8.35 (m, 1H), 8.28 – 8.24 (m, 1H), 7.72 – 7.67 (m, 1H), 7.65 – 7.59 (m, 2H), 7.48 (ddd, *J* = 8.1, 6.6, 1.9 Hz, 1H), 7.44 – 7.38 (m, 2H), 6.72 – 6.66 (m, 2H), 6.35 (s, 1H), 6.24 – 6.19 (m, 1H), 3.50 (d, *J* = 12.9 Hz, 1H), 3.10 (d, *J* = 12.9 Hz, 1H), 1.98 (s, 3H), 1.79 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.7, 149.5, 143.8, 140.6, 137.3, 134.7, 131.4, 131.0, 129.7, 127.8, 127.7, 127.5, 126.5, 125.9, 125.6, 125.5, 125.4, 123.8, 119.6, 115.3, 51.6, 51.2, 25.1, 20.7.

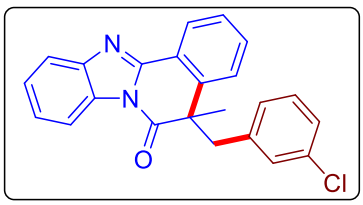
5-(3-Fluorobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ak)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(3-fluorophenyl)iodonium trifluoromethanesulfonate (0.175 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ak** as a white solid (0.046 g, 52%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.37 – 8.32 (m, 2H), 7.74 – 7.71 (m, 1H), 7.67 – 7.60 (m, 2H), 7.53 – 7.49 (m, 1H), 7.45 – 7.40 (m, 2H), 6.80 (td, *J* = 8.0, 6.1 Hz, 1H), 6.65 – 6.59 (m, 1H), 6.38 (d, *J* = 7.7 Hz, 1H), 6.29 – 6.23 (m, 1H), 3.60 (d, *J* = 13.1 Hz, 1H), 3.20 (d, *J* = 13.1 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.4, 162.1 (d, *J* = 246.2 Hz), 149.4, 143.8, 140.3, 137.6 (d, *J* = 7.4 Hz), 131.6, 131.0, 129.3, 129.2, 128.1, 126.5, 125.8 (d, *J* = 7.2 Hz), 125.6, 124.7 (d, *J* = 2.9 Hz), 123.5, 119.7, 116.0 (d, *J* = 21.5 Hz), 115.4, 114.0 (d, *J* = 20.9 Hz), 50.9, 49.9, 26.5. **^{19}F{^{1}H} NMR** (377 MHz, $CDCl_3$) δ -113.5.

5-(3-Chlorobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3al)[7]

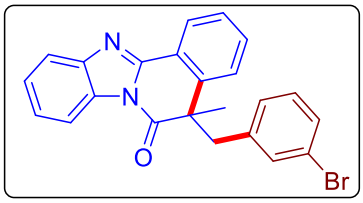


The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(3-chlorophenyl)iodonium

trifluoromethanesulfonate (0.187 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3al** as a white solid (0.073 g, 78%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.37 – 8.31 (m, 2H), 7.75 – 7.71 (m, 1H), 7.67 – 7.63 (m, 1H), 7.60 (d, *J* = 7.2 Hz, 1H), 7.53 – 7.49 (m, 1H), 7.45 – 7.40 (m, 2H), 6.89 (ddd, *J* = 8.0, 2.0, 0.9 Hz, 1H), 6.75 (t, *J* = 7.8 Hz, 1H), 6.53 (t, *J* = 1.8 Hz, 1H), 6.42 (d, *J* = 7.7 Hz, 1H), 3.54 (d, *J* = 13.1 Hz, 1H), 3.14 (d, *J* = 13.1 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.3, 149.3, 143.7, 140.2, 137.0, 133.6, 131.7, 130.9, 129.3, 129.0, 128.1, 127.2, 127.0, 126.4, 125.9, 125.8, 125.6, 123.5, 119.7, 115.4, 51.0, 50.2, 25.9.

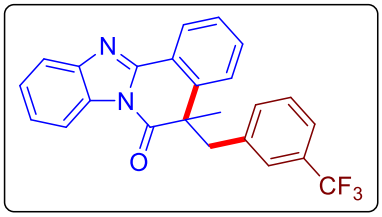
5-(3-Bromobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3am)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(3-bromophenyl)iodonium trifluoromethanesulfonate (0.220 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3am** as a white solid (0.084 g, 81%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 – 8.34 (m, 1H), 8.32 (dd, *J* = 7.8, 1.0 Hz, 1H), 7.75 – 7.71 (m, 1H), 7.66 – 7.62 (m, 1H), 7.59 (d, *J* = 7.3 Hz, 1H), 7.53 – 7.48 (m, 1H), 7.45 – 7.40 (m, 2H), 7.05 – 7.02 (m, 1H), 6.68 (dt, *J* = 9.5, 5.2 Hz, 2H), 6.44 (d, *J* = 7.7 Hz, 1H), 3.54 – 3.49 (m, 1H), 3.12 (d, *J* = 13.1 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.3, 149.3, 143.7, 140.1, 137.2, 132.2, 131.7, 130.9, 130.1, 129.3, 128.1, 127.5, 126.4, 125.9, 125.8, 125.6, 123.5, 121.8, 119.7, 115.4, 51.0, 50.3, 25.7.

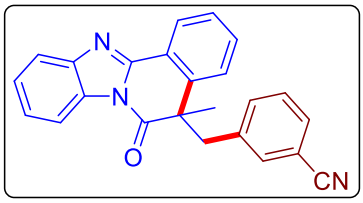
5-Methyl-5-(3-(trifluoromethyl)benzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3an)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(3-(trifluoromethyl)phenyl)iodonium trifluoromethanesulfonate (0.212 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3an** as a white solid (0.062 g, 61%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.35 – 8.32 (m, 1H), 8.28 (dd, *J* = 7.8, 0.9 Hz, 1H), 7.69 (ddd, *J* = 7.4, 5.1, 1.2 Hz, 1H), 7.64 (ddd, *J* = 9.0, 7.4, 1.2 Hz, 2H), 7.50 (ddd, *J* = 8.3, 6.9, 1.6 Hz, 1H), 7.44 – 7.38 (m, 2H), 7.17 (d, *J* = 7.8 Hz, 1H), 6.93 (t, *J* = 7.8 Hz, 1H), 6.77 (s, 1H), 6.70 (d, *J* = 7.8 Hz, 1H), 3.62 (d, *J* = 13.0 Hz, 1H), 3.21 (d, *J* = 13.0 Hz, 1H), 1.99 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.2, 149.1, 143.7, 139.9, 135.9, 132.2, 131.7, 130.8, 129.8 (q, *J* = 32.7 Hz), 128.2, 128.2, 126.4, 125.9, 125.8, 125.6, 123.9, 123.9, 123.5 (q, *J* = 272.8 Hz), 123.5, 119.7, 115.4, 51.0, 50.6, 25.6. **^{19}F{^{1}H} NMR** (471 MHz, $CDCl_3$) δ -62.7. **HRMS-ESI** (m/z): calcd for $C_{24}H_{17}F_3N_2O$ $[M]^+$ 406.1293; found 406.1229.

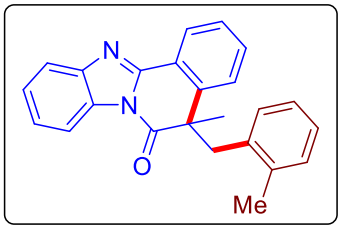
3-((5-Methyl-6-oxo-5,6-dihydrobenzo[4,5]imidazo[2,1-*a*]isoquinolin-5-yl)methyl)benzonitrile (3ao)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(3-cyanophenyl)iodonium trifluoromethanesulfonate (0.180 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (5-20% ethyl acetate in hexane) gave **36** as a white solid (0.036 g, 40%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.30 (m, 2H), 7.74 – 7.66 (m, 2H), 7.64 (dd, *J* = 7.9, 0.8 Hz, 1H), 7.56 – 7.51 (m, 1H), 7.47 – 7.41 (m, 2H), 7.22 (dt, *J* = 7.7, 1.3 Hz, 1H), 6.93 (t, *J* = 7.7 Hz, 1H), 6.84 – 6.78 (m, 2H), 3.62 (d, *J* = 13.2 Hz, 1H), 3.21 (d, *J* = 13.2 Hz, 1H), 1.97 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.0, 149.0, 143.7, 139.7, 136.7, 133.3, 132.6, 131.9, 130.8, 130.8, 128.6, 128.4, 126.4, 126.1, 125.9 (X 2C), 123.5, 119.8, 118.1, 115.3, 111.9, 50.9, 49.8, 26.3. **HRMS-ESI** (m/z): calcd for $C_{24}H_{17}N_3O$ $[M]^+$ 363.1372; found 363.1369.

5-Methyl-5-(2-methylbenzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ap)[7]

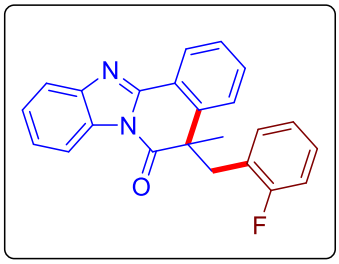


The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), di-o-tolyliodonium trifluoromethanesulfonate (0.172 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ap** as a white solid (0.043 g, 49%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.31 (m, 2H), 7.75 – 7.71 (m, 1H), 7.56 – 7.48 (m, 2H), 7.45 – 7.40 (m, 3H), 6.87 (td, *J* = 7.4, 1.2 Hz, 1H), 6.82 (d, *J* = 7.0 Hz, 1H), 6.65 (dd, *J* =

10.8, 4.1 Hz, 1H), 6.48 (d, *J* = 7.3 Hz, 1H), 3.53 (d, *J* = 13.7 Hz, 1H), 3.23 (d, *J* = 13.8 Hz, 1H), 1.99 (s, 3H), 1.79 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.9, 149.6, 143.8, 140.6, 136.9, 133.1, 131.5, 131.2, 130.2, 129.8, 128.0, 127.1, 126.8, 125.8, 125.7, 125.5, 125.1, 123.6, 119.7, 115.3, 50.7, 47.0, 24.9, 19.0.

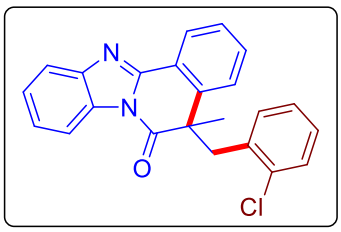
5-(2-Fluorobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3aq)[7]



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(2-fluorophenyl)-iodonium trifluoromethanesulfonate (0.174 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3aq** as a white solid (0.042 g, 47%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 – 8.35 (m, 1H), 8.30 (d, *J* = 7.8 Hz, 1H), 7.76 – 7.73 (m, 1H), 7.58 (d, *J* = 3.8 Hz, 2H), 7.49 – 7.45 (m, 1H), 7.45 – 7.42 (m, 2H), 6.95 (ddd, *J* = 15.3, 5.3, 1.8 Hz, 1H), 6.67 – 6.62 (m, 2H), 6.59 (td, *J* = 7.5, 1.8 Hz, 1H), 3.56 (d, *J* = 13.6 Hz, 1H), 3.29 (d, *J* = 13.6 Hz, 1H), 1.97 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.5, 160.8 (d, *J* = 246.8 Hz), 149.6, 143.9, 140.3, 131.5, 131.2, 131.0 (d, *J* = 4.1 Hz), 129.0 (d, *J* = 8.3 Hz), 128.0, 126.7, 125.7, 125.6, 125.5, 123.5 (d, *J* = 3.6 Hz),123.2, 122.3 (d, *J* = 15.6 Hz), 119.7, 115.5, 114.9 (d, *J* = 22.7 Hz), 50.5, 43.1, 25.3. **^{19}F{^{1}H} NMR** (377 MHz, $CDCl_3$) δ -116.3.

5-(2-Chlorobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ar)[7]

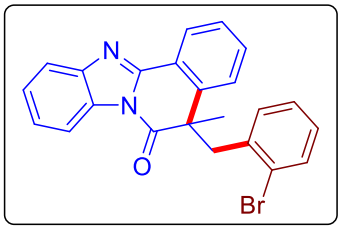


The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(2-chlorophenyl)iodonium trifluoromethanesulfonate (0.187 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ar** as a white solid (0.065 g, 70%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.32 (m, 2H), 7.78 – 7.73 (m, 1H), 7.56 – 7.49 (m, 2H), 7.49 – 7.45 (m, 1H), 7.45 – 7.41 (m, 2H), 7.05 (dd, *J* = 8.0, 1.1 Hz, 1H), 6.93 (td, *J* = 7.8, 1.6 Hz, 1H), 6.75 (td, *J* = 7.6, 1.2 Hz, 1H), 6.61 (dd, *J* = 7.7, 1.4 Hz, 1H), 3.66 (d, *J* = 14.0 Hz, 1H), 3.42 (d, *J* = 14.0 Hz, 1H), 1.96 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.4,

149.6, 143.9, 140.2, 134.7, 133.1, 131.6, 131.3, 130.9, 129.4, 128.4, 128.1, 126.9, 126.1, 125.8, 125.7, 125.5, 123.3, 119.7, 115.5, 50.4, 46.1, 25.2.

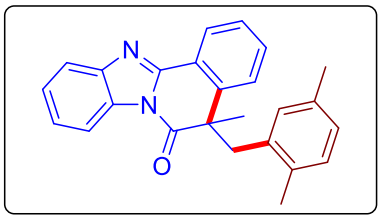
5-(2-Bromobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3as)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(2-bromophenyl)iodonium trifluoromethanesulfonate (0.220 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3as** as a white solid (0.068 g, 65%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.32 (m, 2H), 7.78 – 7.73 (m, 1H), 7.56 – 7.49 (m, 2H), 7.49 – 7.45 (m, 1H), 7.45 – 7.41 (m, 2H), 7.05 (dd, J = 8.0, 1.1 Hz, 1H), 6.93 (td, *J* = 7.8, 1.6 Hz, 1H), 6.75 (td, *J* = 7.6, 1.2 Hz, 1H), 6.61 (dd, *J* = 7.7, 1.4 Hz, 1H), 3.66 (d, *J* = 14.0 Hz, 1H), 3.42 (d, *J* = 14.0 Hz, 1H), 1.96 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.4, 149.6, 143.9, 140.2, 134.9, 132.8 (X 2C), 131.7, 131.3, 130.7, 128.6, 128.1, 127.0, 126.8, 125.9, 125.8, 125.5, 123.3, 119.7, 115.6, 50.3, 48.0, 25.6. **HRMS-ESI** (m/z): calcd for calcd for $C_{23}H_{21}BrN_3O$ $[M + NH_4]^+$ 436.0843; found 436.0873.

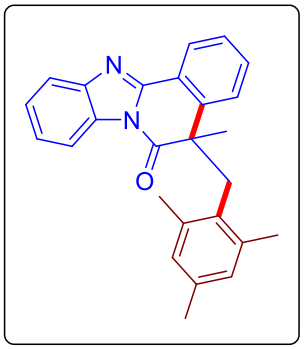
5-(2,5-Dimethylbenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-oneone (3at)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), bis(2,5-dimethylphenyl)iodonium trifluoromethanesulfonate (0.182 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3at** as a white solid (0.037 g, 40%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.33 (m, 1H), 8.28 (dd, *J* = 7.8, 0.9 Hz, 1H), 7.73 – 7.69 (m, 1H), 7.58 – 7.52 (m, 2H), 7.49 (ddd, J = 8.4, 6.8, 1.7 Hz, 1H), 7.44 – 7.38 (m, 2H), 6.63 (dt, J = 7.8, 4.5 Hz, 2H), 6.29 (s, 1H), 3.41 (d, *J* = 13.3 Hz, 1H), 3.17 (d, *J* = 13.4 Hz, 1H), 2.03 (s, 3H), 1.66 (s, 3H), 1.58 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.9, 149.5, 143.7, 140.4, 134.6, 133.6, 132.6, 131.4, 131.1, 130.6, 130.0, 128.0, 127.8, 126.8, 125.7, 125.6, 125.5, 123.9, 119.6, 115.2, 51.1, 48.3, 23.8, 20.0, 18.2. **HRMS-ESI** (m/z): calcd for $C_{25}H_{23}N_2O$ $[M + H]^+$ 367.1805; found 367.1778.

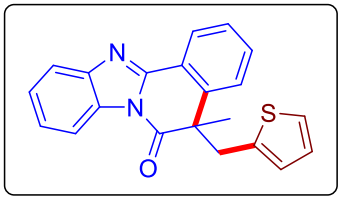
5-Methyl-5-(2,4,6-trimethylbenzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3au)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), dimesityliodonium trifluoromethanesulfonate (0.192 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3au** as a white solid (0.033 g, 35%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.39 (dd, *J* = 7.8, 1.3 Hz, 1H), 8.33 – 8.28 (m, 1H), 7.80 – 7.76 (m, 1H), 7.50 (td, *J* = 7.6, 1.1 Hz, 1H), 7.42 (ddd, *J* = 11.8, 6.8, 2.3 Hz, 3H), 7.10 (d, *J* = 7.4 Hz, 1H), 6.60 (s, 2H), 3.36 – 3.28 (m, 2H), 2.14 (s, 3H), 1.93 (s, 3H), 1.78 (s, 6H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.4, 149.8, 144.1, 140.7, 138.3, 136.3, 131.5, 131.1, 129.0, 128.7, 128.0, 127.5, 125.9, 125.6, 125.4, 123.3, 119.7, 115.2, 51.5, 42.7, 22.4, 20.7, 20.2. **HRMS-ESI** (m/z): calcd for $C_{26}H_{25}N_2O$ $[M + H]^+$ 381.1961; found 381.1958.

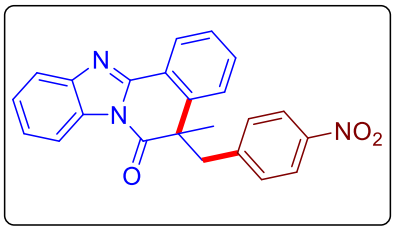
5-Methyl-5-(thiophen-2-ylmethyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3av)



The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), di(thiophen-2-yl)-iodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3av** as a yellow solid (0.026 g, 30%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 (ddd, *J* = 9.1, 4.0, 1.9 Hz, 2H), 7.78 – 7.74 (m, 1H), 7.68 – 7.62 (m, 2H), 7.52 (ddd, *J* = 8.3, 6.8, 1.7 Hz, 1H), 7.45 – 7.41 (m, 2H), 6.80 (dd, *J* = 5.1, 1.1 Hz, 1H), 6.56 (dd, *J* = 5.1, 3.5 Hz, 1H), 6.40 – 6.38 (m, 1H), 3.95 (d, *J* = 14.4 Hz, 1H), 3.48 (d, *J* = 14.4 Hz, 1H), 1.91 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.4, 149.6, 143.8, 140.6, 137.0, 131.8, 131.2, 128.1, 126.7, 126.5, 126.4, 125.9, 125.8, 125.6, 124.6, 123.5, 119.7, 115.6, 51.2, 43.2, 27.9. **HRMS-ESI** (m/z): calcd for $C_{21}H_{16}N_2OS$ $[M + H]^+$ 345.1056; found 345.1056.

5-Methyl-5-(4-nitrobenzyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3aw)

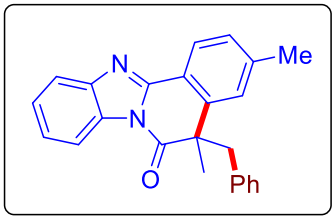


The compound was prepared according to GP using 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.065 g, 0.25 mmol), (4-nitrophenyl)(phenyl)iodonium

trifluoromethanesulfonate (0.178 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (5-15% ethyl acetate in hexane) gave **3aw** as a white solid (0.042 g, 44%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.33 (m, 2H), 7.74 – 7.71 (m, 3H), 7.67 (dt, *J* = 7.9, 3.8 Hz, 2H), 7.53 (ddd, *J* = 8.2, 4.9, 1.7 Hz, 1H), 7.46 – 7.41 (m, 2H), 6.77 (d, *J* = 8.7 Hz, 2H), 3.74 (d, *J* = 13.1 Hz, 1H), 3.33 (d, *J* = 13.1 Hz, 1H), 1.98 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 171.9, 149.0, 146.9, 143.7, 142.9, 139.7, 131.8, 130.8, 130.0, 128.4, 126.5, 126.2, 126.0, 125.8, 123.3, 123.1, 119.9, 115.4, 50.9, 49.3, 27.2. **HRMS-ESI** (m/z): calcd for $C_{26}H_{25}N_2O$ $[M]^+$ 383.1270; found 383.1303.

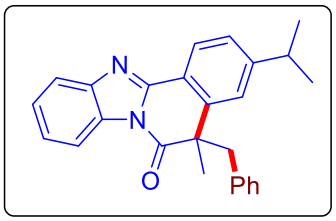
5-Benzyl-3,5-dimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ba)[7]



The compound was prepared according to GP using 2-methyl-1-(2-(*p*-tolyl)-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.069 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ba** as a white solid (0.057 g, 65%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.32 (m, 1H), 8.19 (d, *J* = 8.0 Hz, 1H), 7.72 – 7.68 (m, 1H), 7.41 – 7.37 (m, 3H), 7.30 (d, *J* = 8.0 Hz, 1H), 6.92 (t, *J* = 7.4 Hz, 1H), 6.84 (t, *J* = 7.6 Hz, 2H), 6.56 (d, *J* = 7.2 Hz, 2H), 3.54 (d, *J* = 13.0 Hz, 1H), 3.19 (d, *J* = 13.0 Hz, 1H), 2.53 (s, 3H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.8, 149.8, 143.7, 142.1, 140.7, 135.1, 131.0, 129.0, 128.9, 127.7, 127.1, 127.0, 125.6 (X 2C), 125.2, 120.8, 119.4, 115.3, 51.0, 50.8, 25.9, 22.0.

5-Benzyl-3-isopropyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ca)

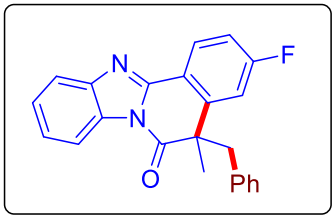


The compound was prepared according to GP using 1-(2-(4-isopropylphenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.076 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ca** as a white solid (0.064 g, 67%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.37 – 8.33 (m, 1H), 8.23 (d, *J* = 8.4 Hz, 1H), 7.74 – 7.69 (m, 1H), 7.42 – 7.38 (m, 2H), 7.37 – 7.34 (m, 2H), 6.93 (ddd, *J* = 6.8, 3.9, 1.2 Hz, 1H), 6.85 (dd,

J = 10.4, 4.7 Hz, 2H), 6.58 – 6.54 (m, 2H), 3.55 (d, *J* = 13.0 Hz, 1H), 3.20 (d, *J* = 13.1 Hz, 1H), 3.09 – 3.01 (m, 1H), 1.94 (s, 3H), 1.36 (d, *J* = 6.9 Hz, 6H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.0, 152.9, 149.8, 143.7, 140.6, 135.1, 131.0, 129.1, 127.8, 127.0, 126.2, 125.8, 125.7, 125.2, 124.7, 121.0, 119.4, 115.4, 51.1, 50.7, 34.6, 25.8, 24.0, 23.7. **HRMS-ESI** (m/z): calcd for $C_{26}H_{25}N_2O$ $[M + H]^+$ 381.1961; found 381.1969.

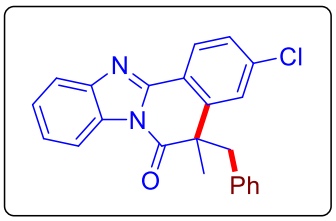
5-Benzyl-3-fluoro-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3da)



The compound was prepared according to GP using 1-(2-(4-fluorophenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.070 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3da** as a white solid (0.040 g, 45%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.28 (m, 2H), 7.71 – 7.68 (m, 1H), 7.44 – 7.38 (m, 2H), 7.30 – 7.27 (m, 1H), 7.22 – 7.17 (m, 1H), 6.93 (t, *J* = 7.4 Hz, 1H), 6.84 (dd, *J* = 10.5, 4.7 Hz, 2H), 6.60 – 6.56 (m, 2H), 3.59 (d, *J* = 13.1 Hz, 1H), 3.14 (d, *J* = 13.1 Hz, 1H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.1, 164.8 (d, J = 252.8 Hz), 148.7, 143.6, 143.5 (d, *J* = 7.7 Hz), 134.6, 130.9, 128.9, 128.1 (d, *J* = 9.1 Hz), 127.9, 127.3, 125.8, 125.5, 120.0, 119.6, 115.8 (d, *J* = 22.4 Hz), 115.4, 113.6 (d, *J* = 23.1 Hz), 51.3, 50.9, 25.8. **^{19}F{^{1}H} NMR** (471 MHz, $CDCl_3$) δ -106.6. **HRMS-ESI** (m/z): calcd for $C_{23}H_{18}FN_2O$ $[M + H]^+$ 357.1398; found 357.1409.

5-Benzyl-3-chloro-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ea)[7]

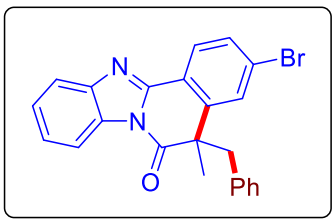


The compound was prepared according to GP using 1-(2-(4-chlorophenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.074 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ea** as a white solid (0.065 g, 70%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.36 – 8.33 (m, 1H), 8.22 (d, *J* = 8.4 Hz, 1H), 7.71 – 7.68 (m, 1H), 7.57 (d, *J* = 1.9 Hz, 1H), 7.46 (dd, *J* = 8.4, 2.0 Hz, 1H), 7.44 – 7.39 (m, 2H), 6.94 (dd, *J* = 10.5, 4.2 Hz, 1H), 6.84 (dd, *J* = 10.4, 4.7 Hz, 2H), 6.58 – 6.55 (m, 2H), 3.57 (d, *J* = 13.1

Hz, 1H), 3.15 (d, *J* = 13.1 Hz, 1H), 1.95 (s, 3H). **$^{13}C\{^{1}H\}$ NMR** (126 MHz, $CDCl_3$) δ 171.9, 148.6, 143.7, 142.4, 137.7, 134.6, 131.0, 129.0, 128.4, 127.9, 127.3, 127.0, 126.8, 125.9, 125.7, 122.2, 119.7, 115.4, 51.2, 50.9, 25.7.

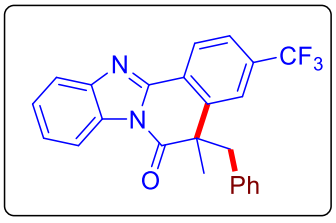
5-Benzyl-3-bromo-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3fa)



The compound was prepared according to GP using 1-(2-(4-bromophenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.085 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3fa** as a white solid (0.070 g, 67%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.34 (dd, *J* = 6.8, 2.2 Hz, 1H), 8.14 (d, *J* = 8.4 Hz, 1H), 7.73 (d, *J* = 1.7 Hz, 1H), 7.69 (dd, *J* = 6.6, 2.1 Hz, 1H), 7.62 (dd, *J* = 8.4, 1.7 Hz, 1H), 7.45 – 7.39 (m, 2H), 6.94 (t, *J* = 7.4 Hz, 1H), 6.85 (t, *J* = 7.6 Hz, 2H), 6.57 (d, *J* = 7.4 Hz, 2H), 3.57 (d, *J* = 13.1 Hz, 1H), 3.15 (d, *J* = 13.1 Hz, 1H), 1.95 (s, 3H). **$^{13}C\{^{1}H\}$ NMR** (126 MHz, $CDCl_3$) δ 171.8, 148.6, 143.7, 142.5, 134.5, 131.3, 131.0, 129.8, 129.0, 127.9, 127.3, 127.1, 126.0, 125.9, 125.7, 122.6, 119.7, 115.4, 51.1, 50.9, 25.6. **HRMS-ESI** (m/z): calcd for $C_{23}H_{18}BrN_2O$ $[M + H]^+$ 417.0597; found 419.0595.

5-Benzyl-5-methyl-3-(trifluoromethyl)benzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ga)

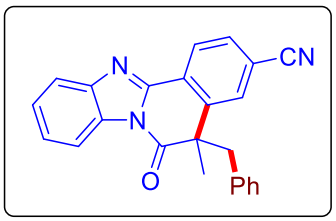


The compound was prepared according to GP using 2-methyl-1-(2-(4-(trifluoromethyl)phenyl)-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.083 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ga** as a white solid (0.079 g, 78%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.41 – 8.35 (m, 2H), 7.78 (s, 1H), 7.72 (dd, *J* = 6.8, 1.4 Hz, 2H), 7.44 (pd, *J* = 7.3, 1.4 Hz, 2H), 6.92 (t, *J* = 7.4 Hz, 1H), 6.83 (t, *J* = 7.6 Hz, 2H), 6.51 (d, *J* = 7.3 Hz, 2H), 3.58 (d, *J* = 13.2 Hz, 1H), 3.17 (d, *J* = 13.2 Hz, 1H), 1.98 (s, 3H). **$^{13}C\{^{1}H\}$ NMR** (126 MHz, $CDCl_3$) δ 171.8, 148.0, 143.6, 141.2, 132.9 (q, *J* = 32.7 Hz), 134.4, 131.0, 128.9, 128.2, 127.9, 127.3, 126.2, 126.2, 126.0, 125.9 (q, *J* = 272.9 Hz), 124.6 (q, *J* = 3.6

Hz), 123.8 (q, *J* = 3.8 Hz), 120.0, 115.5, 51.3, 50.9, 25.4. **^{19}F{^{1}H} NMR** (471 MHz, $CDCl_3$) δ -62.8. **HRMS-ESI** (m/z): calcd for $C_{24}H_{18}F_3N_2O$ $[M + H]^+$ 407.1366; found 407.1379.

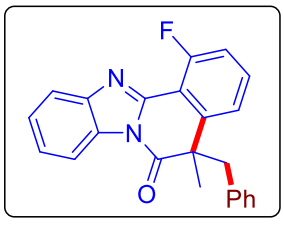
5-Benzyl-5-methyl-6-oxo-5,6-dihydrobenzo[4,5]imidazo[2,1-*a*]isoquinoline-3-carbonitrile (3ha)



The compound was prepared according to GP using 4-(1-methacryloyl-1*H*-benzo[*d*]imidazol-2-yl)benzonitrile (0.072 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ha** as a white solid (0.068 g, 75%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.37 – 8.32 (m, 1H), 8.22 (d, *J* = 8.4 Hz, 1H), 7.71 – 7.67 (m, 1H), 7.57 (d, *J* = 1.9 Hz, 1H), 7.46 (dd, *J* = 8.4, 2.0 Hz, 1H), 7.45 – 7.39 (m, 2H), 6.94 (t, *J* = 7.4 Hz, 1H), 6.85 (t, *J* = 7.6 Hz, 2H), 6.57 (d, *J* = 7.3 Hz, 2H), 3.58 (d, *J* = 13.1 Hz, 1H), 3.15 (d, *J* = 13.1 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 171.9, 148.6, 143.7, 142.3, 137.7, 134.6, 130.9, 129.0, 128.4, 127.9 (X 2C), 127.3, 127.0, 126.8, 125.9, 125.7, 122.2, 119.7, 115.4, 51.2, 50.9, 25.7. **HRMS-ESI** (m/z): calcd for $C_{24}H_{18}N_3O$ $[M + H]^+$ 364.1444; found 364.1440.

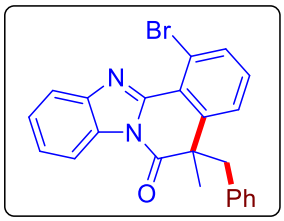
5-Benzyl-1-fluoro-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ia)



The compound was prepared according to GP using 1-(2-(2-fluorophenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.070 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ia** as a white solid (0.040 g, 45%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.38 (dd, *J* = 7.2, 1.3 Hz, 1H), 7.83 – 7.79 (m, 1H), 7.59 (td, *J* = 8.1, 5.3 Hz, 1H), 7.43 (ddd, *J* = 9.2, 6.0, 1.4 Hz, 3H), 7.27 – 7.21 (m, 1H), 6.93 (t, *J* = 7.4 Hz, 1H), 6.83 (t, *J* = 7.6 Hz, 2H), 6.54 (d, *J* = 7.3 Hz, 2H), 3.58 (d, *J* = 13.1 Hz, 1H), 3.16 (d, *J* = 13.1 Hz, 1H), 1.97 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.0, 160.2 (d, *J* = 262.4 Hz), 145.6 (d, *J* = 8.0 Hz), 144.0, 143.2, 134.6, 132.1 (d, *J* = 9.5 Hz), 130.0, 128.9, 127.9, 127.3, 125.9 (d, *J* = 26.7 Hz), 122.5 (d, *J* = 3.4 Hz), 120.4, 115.8, 115.6, 115.3, 113.0 (d, *J* = 9.7 Hz), 51.2, 51.1, 26.1. **^{19}F{^{1}H} NMR** (471 MHz, $CDCl_3$) δ -107.9. **HRMS-ESI** (m/z): calcd for $C_{23}H_{18}FN_2O$ $[M + H]^+$ 357.1398; found 357.1395.

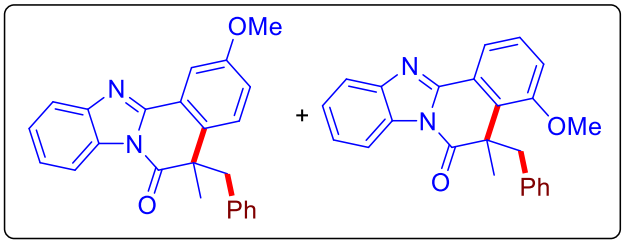
5-Benzyl-1-bromo-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ja)



The compound was prepared according to GP using 1-(2-(2-bromophenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.085 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ja** as a white solid (0.057 g, 55%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.39 – 8.36 (m, 1H), 7.83 – 7.80 (m, 2H), 7.58 (dd, *J* = 7.9, 0.9 Hz, 1H), 7.47 – 7.39 (m, 3H), 6.94 (t, *J* = 7.4 Hz, 1H), 6.82 (t, *J* = 7.7 Hz, 2H), 6.50 (d, *J* = 7.2 Hz, 2H), 3.53 (d, *J* = 13.1 Hz, 1H), 3.13 (d, *J* = 13.1 Hz, 1H), 1.96 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 171.7, 146.9, 143.6, 143.3, 135.2, 134.5, 130.9, 130.4, 128.9, 127.8, 127.4, 126.2, 126.0, 125.7, 123.1, 121.1, 120.6, 115.3, 51.5, 51.4, 25.7. **HRMS-ESI** (m/z): calcd for $C_{23}H_{18}BrN_2O$ $[M + H]^+$ 417.0597; found 417.0595.

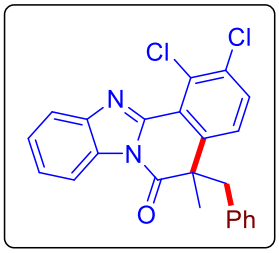
5-Benzyl-2-methoxy-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ka)



The compound was prepared according to GP using 1-(2-(3-methoxyphenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.073 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (5-15% ethyl acetate in hexane) gave **3ka and 3ka'** as a white solid in 1.1:1 ratio (0.072 g, 78%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.44 – 8.39 (m, 1H), 8.38 – 8.34 (m, 1H), 8.01 (dd, *J* = 7.8, 0.9 Hz, 1H), 7.76 (d, *J* = 2.8 Hz, 1H), 7.74 – 7.70 (m, 2H), 7.48 (d, *J* = 8.7 Hz, 1H), 7.45 (d, *J* = 8.0 Hz, 1H), 7.44 – 7.39 (m, 4H), 7.19 (dd, *J* = 8.7, 2.8 Hz, 1H), 7.15 (d, *J* = 8.2 Hz, 1H), 6.91 (t, *J* = 7.4 Hz, 1H), 6.88 – 6.80 (m, 5H), 6.70 – 6.65 (m, 2H), 6.57 (d, *J* = 7.4 Hz, 2H), 4.06 (s, 3H), 4.02 (d, *J* = 13.1 Hz, 1H), 3.94 (s, 3H), 3.55 (dd, *J* = 13.1, 7.4 Hz, 2H), 3.15 (d, *J* = 13.0 Hz, 1H), 2.05 (s, 3H), 1.92 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.8, 172.9, 159.0, 157.4, 149.8, 149.6, 143.8, 143.5, 136.9, 135.1, 132.9, 131.1, 131.0, 129.0, 128.7, 128.3, 128.2, 128.0, 127.8, 127.8, 127.0, 126.6, 125.8, 125.7, 125.5, 125.4, 124.6, 124.4, 120.2, 119.6, 119.5, 118.5, 115.7, 115.4, 114.1, 107.5, 55.7, 55.6, 51.4, 50.8, 50.7, 44.2, 26.1, 24.8. **HRMS-ESI** (m/z): calcd for $C_{24}H_{21}N_2O_2$ $[M + H]^+$ 369.1598; found 369.1600.

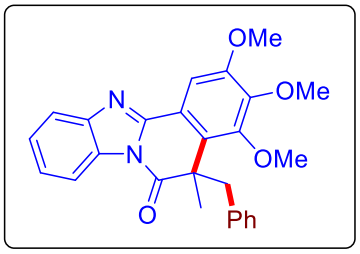
5-Benzyl-1,2-dichloro-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3la)



The compound was prepared according to GP using 1-(2-(2,3-dichlorophenyl)-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.083 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3la** as a white solid (0.065 g, 64%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.37 (d, *J* = 7.4 Hz, 1H), 7.82 (d, *J* = 7.4 Hz, 1H), 7.69 (d, *J* = 8.6 Hz, 1H), 7.51 – 7.39 (m, 3H), 6.95 (t, *J* = 7.4 Hz, 1H), 6.82 (dd, *J* = 10.6, 4.7 Hz, 2H), 6.51 (d, *J* = 7.1 Hz, 2H), 3.53 (d, *J* = 13.1 Hz, 1H), 3.10 (d, *J* = 13.1 Hz, 1H), 1.96 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 171.4, 146.2, 143.4, 141.5, 134.8, 134.3, 131.6, 131.3, 130.2, 128.8, 127.9, 127.5, 126.5, 125.8, 125.8, 123.8, 120.7, 115.3, 51.3, 51.2, 25.6. **HRMS-ESI** (m/z): calcd for $C_{23}H_{17}Cl_2N_2O$ $[M + H]^+$ 407.0712; found 407.0716.

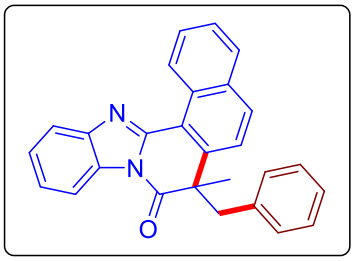
5-Benzyl-2,3,4-trimethoxy-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ma)



The compound was prepared according to GP using 2-methyl-1-(2-(3,4,5-trimethoxyphenyl)-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.088 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ma** as a white solid (0.073 g, 68%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.39 – 8.35 (m, 1H), 7.72 – 7.65 (m, 1H), 7.61 (s, 1H), 7.39 (p, *J* = 6.0 Hz, 2H), 6.89 – 6.81 (m, 3H), 6.70 (d, *J* = 7.1 Hz, 2H), 4.16 (s, 3H), 4.00 (d, *J* = 7.4 Hz, 6H), 3.80 (d, *J* = 13.1 Hz, 1H), 3.53 (d, *J* = 13.1 Hz, 1H), 2.02 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.7, 153.5, 152.4, 149.7, 145.5, 143.7, 136.7, 131.0, 128.8, 127.8, 126.6, 126.3, 125.7, 125.2, 119.3, 119.0, 115.6, 103.4, 61.1, 60.73, 56.2, 51.2, 45.8, 25.7. **HRMS-ESI** (m/z): calcd for $C_{26}H_{24}N_2O_4$ $[M]^+$ 428.1736; found 428.1745.

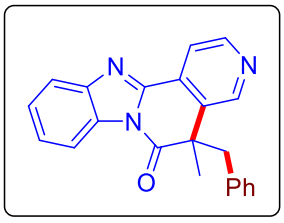
7-Benzyl-7-methylbenzo[h]benzo[4,5]imidazo[2,1-*a*]isoquinolin-8(7*H*)-one (3na)



The compound was prepared according to GP using 2-methyl-1-(2-(naphthalen-1-yl)-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.078 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3na** as a white solid (0.056 g, 58%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 10.19 (d, *J* = 8.7 Hz, 1H), 8.35 – 8.31 (m, 1H), 8.01 (d, *J* = 8.7 Hz, 1H), 7.85 (d, *J* = 8.0 Hz, 1H), 7.73 – 7.70 (m, 1H), 7.69 – 7.65 (m, 2H), 7.55 (ddd, *J* = 8.0, 6.9, 1.1 Hz, 1H), 7.38 – 7.32 (m, 2H), 6.77 – 6.73 (m, 1H), 6.63 (dd, *J* = 10.6, 4.8 Hz, 2H), 6.47 – 6.43 (m, 2H), 3.57 (d, *J* = 13.2 Hz, 1H), 3.24 (d, *J* = 13.2 Hz, 1H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.67, 149.6, 143.8, 141.0, 135.0, 132.7, 132.4, 130.0, 130.0, 128.8, 128.7, 128.3, 128.0, 127.7, 127.0, 126.8, 125.7, 125.6, 123.5, 119.9, 119.0, 115.4, 51.4, 50.2, 26.5. **HRMS-ESI** (m/z): calcd for $C_{26}H_{25}N_2O$ $[M + H]^+$ 389.1648; found 389.1679.

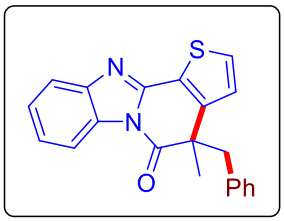
5-Benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*][2,6]naphthyridin-6(5*H*)-one (3oa)[7]



The compound was prepared according to GP using 2-methyl-1-(2-(pyridin-4-yl)-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.066 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (15-20% ethyl acetate in hexane) gave **3oa** as a white solid (0.030 g, 35%).

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.92 (s, 1H), 8.70 (d, *J* = 5.1 Hz, 1H), 8.38 – 8.34 (m, 1H), 8.04 (d, *J* = 5.1 Hz, 1H), 7.75 – 7.71 (m, 1H), 7.45 (dtd, *J* = 16.6, 7.4, 1.3 Hz, 2H), 6.89 (ddd, *J* = 6.7, 3.9, 1.3 Hz, 1H), 6.83 – 6.77 (m, 2H), 6.55 – 6.51 (m, 2H), 3.63 (d, *J* = 13.1 Hz, 1H), 3.22 (d, *J* = 13.1 Hz, 1H), 2.01 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 171.6, 148.9, 148.6, 147.0, 143.6, 134.9, 134.4, 131.1, 130.5, 129.0, 128.0, 127.3, 126.7, 126.2, 120.4, 118.0, 115.6, 50.7, 49.9, 25.4.

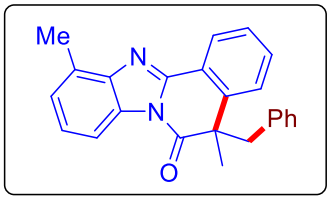
4-Benzyl-4-methylbenzo[4,5]imidazo[1,2-*a*]thieno[2,3-*c*]pyridin-5(4*H*)-one (3pa)



The compound was prepared according to GP using 2-methyl-1-(2-(thiophen-2-yl)-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.067 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3pa** as a yellow solid (0.026 g, 30%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.31 (dd, *J* = 6.6, 2.1 Hz, 1H), 7.66 – 7.63 (m, 2H), 7.41 – 7.35 (m, 2H), 7.20 (d, *J* = 5.1 Hz, 1H), 6.96 (dd, *J* = 8.3, 6.3 Hz, 1H), 6.90 (dd, *J* = 10.0, 4.6 Hz, 2H), 6.67 – 6.64 (m, 2H), 3.55 (d, *J* = 13.0 Hz, 1H), 3.17 (d, *J* = 13.0 Hz, 1H), 1.89 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.2, 146.7, 146.4, 143.5, 135.0, 130.6, 130.4, 128.9, 127.9, 127.2, 125.9, 125.7, 125.4, 124.3, 119.5, 115.0, 51.2, 49.6, 26.0. **HRMS-ESI** (m/z): calcd for $C_{21}H_{17}N_2OS$ $[M + H]^+$ 345.1056; found 345.1063.

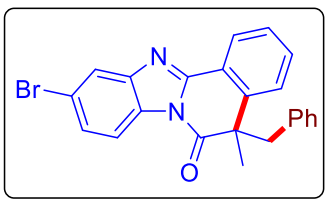
5-Benzyl-5,11-dimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3qa)



The compound was prepared according to GP using 2-methyl-1-(4-methyl-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.069 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3qa** as a white solid (0.048 g, 55%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.37 – 8.35 (m, 1H), 8.19 (d, *J* = 8.0 Hz, 1H), 7.63 – 7.58 (m, 2H), 7.47 (ddd, *J* = 8.2, 6.3, 2.2 Hz, 1H), 7.31 (t, *J* = 7.7 Hz, 1H), 7.21 (d, *J* = 7.4 Hz, 1H), 6.95 – 6.91 (m, 1H), 6.85 (dd, *J* = 10.4, 4.7 Hz, 2H), 6.61 – 6.57 (m, 2H), 3.60 (d, *J* = 13.1 Hz, 1H), 3.21 (d, *J* = 13.1 Hz, 1H), 2.66 (s, 3H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.7, 148.7, 143.0, 140.5, 135.2, 131.2, 130.8, 129.8, 129.1, 127.8, 127.7, 127.0, 126.6, 126.3, 125.8, 125.3, 123.7, 112.8, 51.0, 50.5, 26.4, 16.6. **HRMS-ESI** (m/z): calcd for $C_{24}H_{21}N_2O$ $[M + H]^+$ 353.1648; found 353.1654.

5-Benzyl-10-bromo-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ra)

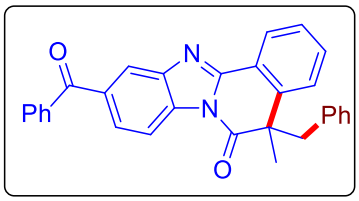


The compound was prepared according to GP using 1-(5-bromo-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.085 g, 0.25 mmol), diphenyliodonium

trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ra** as a white solid (0.063 g, 60%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.25 (dd, *J* = 7.9, 1.0 Hz, 1H), 8.21 (d, *J* = 8.5 Hz, 1H), 7.82 (d, *J* = 1.8 Hz, 1H), 7.68 – 7.61 (m, 2H), 7.53 – 7.47 (m, 2H), 6.94 – 6.90 (m, 1H), 6.81 (dd, *J* = 10.5, 4.8 Hz, 2H), 6.52 – 6.48 (m, 2H), 3.55 (d, *J* = 13.0 Hz, 1H), 3.17 (d, *J* = 13.0 Hz, 1H), 1.97 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.5, 150.5, 145.1, 140.8, 134.7, 131.9, 130.0, 128.9, 128.3, 128.0, 127.8, 127.2, 126.6, 125.8, 123.2, 122.6, 118.8, 116.4, 51.2 (X 2C), 25.7. **HRMS-ESI** (m/z): calcd for $C_{23}H_{17}BrN_2O$ $[M]^+$ 418.0504; found 418.0533.

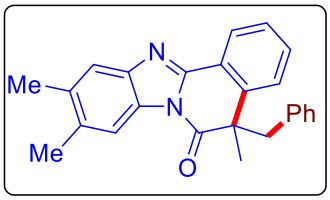
10-Benzoyl-5-benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3sa)



The compound was prepared according to GP using 1-(5-benzoyl-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.092 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3sa** as a white solid (0.067 g, 61%).

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.42 – 8.39 (m, 1H), 8.28 – 8.24 (m, 1H), 8.09 – 8.07 (m, 1H), 7.95 – 7.91 (m, 1H), 7.83 – 7.79 (m, 2H), 7.65 – 7.59 (m, 2H), 7.59 – 7.56 (m, 1H), 7.50 – 7.45 (m, 3H), 6.91 – 6.86 (m, 1H), 6.79 (t, *J* = 7.5 Hz, 2H), 6.52 – 6.47 (m, 2H), 3.54 (d, *J* = 13.0 Hz, 1H), 3.17 (d, *J* = 13.0 Hz, 1H), 1.95 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 196.2, 172.7, 151.0, 143.3, 140.7, 137.7, 135.1, 134.8, 133.8, 132.4, 132.1, 130.1, 128.9, 128.3, 128.1, 127.8, 127.5, 127.2, 126.7, 125.8, 123.2, 122.2, 115.2, 51.3, 51.2, 25.8. **HRMS-ESI** (m/z): calcd for $C_{30}H_{23}N_2O_2$ $[M + H]^+$ 443.1754; found 443.1726.

5-Benzyl-5,9,10-trimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ta)[7]

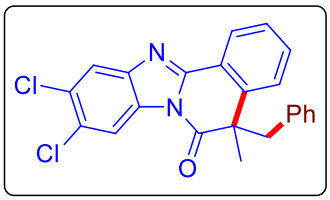


The compound was prepared according to GP using 1-(5,6-dimethyl-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.073 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ta** as a white solid (0.064 g, 70%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.27 (d, *J* = 7.8 Hz, 1H), 8.14 (s, 1H), 7.62 – 7.58 (m, 2H), 7.48 – 7.44 (m, 2H), 6.94 – 6.90 (m, 1H), 6.84 (dd, *J* = 10.3, 4.7 Hz, 2H), 6.58 – 6.54 (m,

2H), 3.59 (d, J = 13.1 Hz, 1H), 3.19 (d, *J* = 13.1 Hz, 1H), 2.44 (s, 3H), 2.39 (s, 3H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.6, 148.7, 140.5, 135.1, 134.7, 134.7, 131.1, 129.3, 129.0, 127.8, 127.8, 127.0, 126.6, 125.5, 123.6, 119.8, 115.7, 51.0, 50.6, 26.3, 20.6, 20.5.

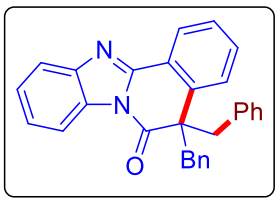
5-Benzyl-9,10-dichloro-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3ua)



The compound was prepared according to GP using 1-(5,6-dichloro-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.083 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3ua** as a white solid (0.082 g, 81%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.44 (s, 1H), 8.20 (dd, *J* = 7.9, 1.1 Hz, 1H), 7.72 (s, 1H), 7.65 (dtd, *J* = 9.0, 8.0, 1.2 Hz, 2H), 7.50 – 7.46 (m, 1H), 6.93 – 6.89 (m, 1H), 6.81 (dd, *J* = 10.5, 4.7 Hz, 2H), 6.50 – 6.46 (m, 2H), 3.53 (d, *J* = 13.0 Hz, 1H), 3.16 (d, *J* = 13.0 Hz, 1H), 1.96 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.4, 151.1, 143.2, 140.8, 134.6, 132.2, 129.9, 129.7, 129.1, 128.8, 128.0, 127.8, 127.2, 126.7, 125.8, 123.0, 120.8, 116.7, 51.3, 51.2, 25.6. **HRMS-ESI** (m/z): calcd for $C_{23}H_{17}Cl_2N_2O$ $[M + H]^+$ 407.0712; found 407.0710.

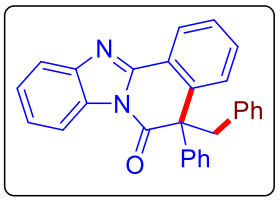
5,5-Dibenzylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3va)



The compound was prepared according to GP using 2-benzyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.085 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3va** as a white solid (0.057 g, 55%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.42 – 8.39 (m, 1H), 8.18 (dd, *J* = 7.9, 1.1 Hz, 1H), 7.84 (d, *J* = 7.9 Hz, 1H), 7.71 – 7.67 (m, 1H), 7.65 – 7.62 (m, 1H), 7.47 – 7.43 (m, 1H), 7.43 – 7.39 (m, 1H), 7.39 – 7.35 (m, 1H), 6.95 – 6.86 (m, 6H), 6.77 – 6.72 (m, 4H), 3.94 (d, *J* = 13.4 Hz, 2H), 3.50 (d, *J* = 13.4 Hz, 2H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 171.6, 149.3, 143.5, 138.5, 135.2, 131.2, 130.8, 129.3, 128.0, 127.9, 127.3, 126.9, 125.7, 125.7, 125.4, 124.8, 119.5, 115.5, 57.6, 48.0. **HRMS-ESI** (m/z): calcd for $C_{28}H_{21}N_2O$ $[M + H]^+$ 415.1805; found 415.1806.

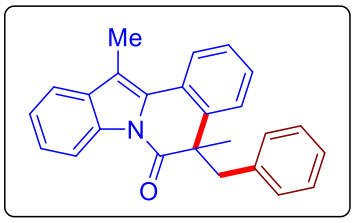
5-Benzyl-5-phenylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (3wa)



The compound was prepared according to GP using 2-phenyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.081 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3wa** as a white solid (0.060 g, 60%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.33 – 8.26 (m, 2H), 7.73 – 7.69 (m, 1H), 7.53 (td, *J* = 7.6, 1.5 Hz, 1H), 7.48 – 7.45 (m, 1H), 7.39 (ddd, *J* = 11.7, 9.1, 4.5 Hz, 7H), 7.26 – 7.23 (m, 1H), 6.91 (t, *J* = 7.4 Hz, 1H), 6.80 (t, *J* = 7.7 Hz, 2H), 6.58 (d, *J* = 7.2 Hz, 2H), 4.34 (d, *J* = 12.4 Hz, 1H), 3.62 (d, *J* = 12.4 Hz, 1H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 171.2, 149.5, 143.8, 142.6, 141.3, 134.3, 131.4, 131.0, 129.3, 129.2, 128.9, 128.1, 128.0, 127.9, 127.8, 127.2, 125.8, 125.6, 125.2, 124.2, 119.7, 115.6, 59.5, 47.0. **HRMS-ESI** (m/z): calcd for $C_{28}H_{21}N_2O$ $[M + H]^+$ 401.1648; found 401.1655.

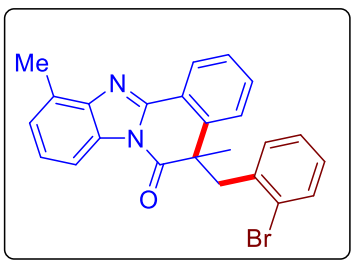
5-benzyl-5,12-dimethylindolo[2,1-*a*]isoquinolin-6(5*H*)-one (3xa)



The compound was prepared according to GP using 2-methyl-1-(3-methyl-2-phenyl-1*H*-indol-1-yl)prop-2-en-1-one (0.069 g, 0.25 mmol), diphenyliodonium trifluoromethanesulfonate (0.166 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **3wa** as a white solid (0.031 g, 35%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.59 (d, *J* = 8.1 Hz, 1H), 7.83 – 7.78 (m, 1H), 7.50 (dd, *J* = 10.6, 5.1 Hz, 2H), 7.42 – 7.37 (m, 3H), 7.34 (t, *J* = 7.4 Hz, 1H), 6.92 (t, *J* = 7.4 Hz, 1H), 6.79 (t, *J* = 7.6 Hz, 2H), 6.53 (d, *J* = 7.5 Hz, 2H), 3.44 (d, *J* = 13.0 Hz, 1H), 3.04 (d, *J* = 13.0 Hz, 1H), 2.43 (s, 3H), 1.91 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.1, 137.6, 135.8, 134.1, 132.3, 129.7, 129.3, 127.6, 127.3 (X 2C), 127.1, 126.7, 126.5, 125.5, 124.8, 123.9, 118.2, 116.3, 113.7, 50.9, 50.3, 24.9, 11.0. **HRMS-ESI** (m/z): calcd for $C_{25}H_{21}NO$ $[M + H]^+$ 352.1696; found 352.1700.

5-(2-Bromobenzyl)-5,11-dimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (7b)

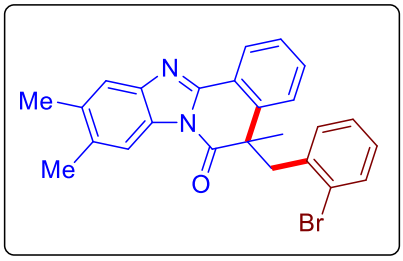


The compound was prepared according to GP using 2-methyl-1-(4-methyl-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-

one (0.069 g, 0.25 mmol), bis(2-bromophenyl)iodonium trifluoromethanesulfonate (0.220 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **7b** as a white solid (0.085 g, 79%).

^{1}H NMR (500 MHz, CDCl3) δ 8.43 (dd, *J* = 7.6, 1.5 Hz, 1H), 8.19 (d, *J* = 8.0 Hz, 1H), 7.53 – 7.44 (m, 3H), 7.33 (t, *J* = 7.7 Hz, 2H), 7.24 (d, *J* = 7.4 Hz, 1H), 6.90 – 6.83 (m, 2H), 6.67 (dd, *J* = 7.6, 1.8 Hz, 1H), 3.70 (d, *J* = 14.3 Hz, 1H), 3.49 (d, *J* = 14.3 Hz, 1H), 2.71 (s, 3H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.5, 148.8, 143.2, 140.0, 135.1, 132.9, 131.4, 131.1, 130.7, 129.9, 128.5, 127.9, 126.9, 126.8, 126.4, 126.0, 125.6, 125.4, 123.4, 113.00, 50.2, 47.7, 25.8, 16.7. **HRMS-ESI** (m/z): calcd for $C_{24}H_{20}BrN_2O$ $[M + H]^+$ 431.0754; found 431.0762.

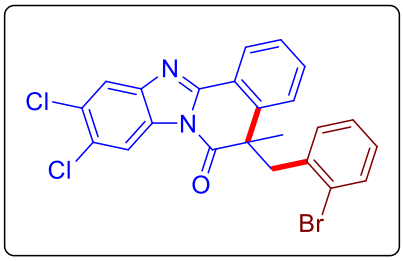
5-(2-Bromobenzyl)-5,9,10-trimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (7c)



The compound was prepared according to GP using 1-(5,6-dimethyl-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.073 g, 0.25 mmol), bis(2-bromophenyl)iodonium trifluoromethanesulfonate (0.220 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **7c** as a white solid (0.088 g, 79%).

^{1}H NMR (500 MHz, CDCl3) δ 8.36 – 8.32 (m, 1H), 8.14 (s, 1H), 7.53 (s, 1H), 7.52 – 7.44 (m, 3H), 7.31 (dd, *J* = 7.8, 1.3 Hz, 1H), 6.89 – 6.82 (m, 2H), 6.64 (dd, *J* = 7.6, 1.7 Hz, 1H), 3.70 (d, *J* = 14.3 Hz, 1H), 3.48 (d, *J* = 14.3 Hz, 1H), 2.45 (s, 3H), 2.42 (s, 3H), 1.94 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.3, 148.9, 142.4, 140.0, 135.1, 134.8, 134.7, 132.8, 131.2, 130.6, 129.7, 128.5, 128.0, 127.0, 126.8, 125.7, 125.6, 123.5, 119.9, 115.9, 50.1, 47.7, 25.9, 20.6, 20.5. **HRMS-ESI** (m/z): calcd for $C_{25}H_{22}BrN_2O$ $[M + H]^+$ 445.0910; found 445.0921.

5-(2-Bromobenzyl)-9,10-dichloro-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (7d)

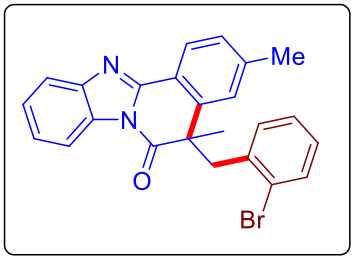


The compound was prepared according to GP using 1-(5,6-dichloro-2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)-2-methylprop-2-en-1-one (0.083 g, 0.25 mmol), bis(2-

bromophenyl)iodonium trifluoromethanesulfonate (0.220 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **7d** as a white solid (0.081 g, 67%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.43 (s, 1H), 8.28 – 8.24 (m, 1H), 7.77 (s, 1H), 7.59 – 7.55 (m, 1H), 7.53 – 7.46 (m, 2H), 7.29 – 7.25 (m, 1H), 6.88 – 6.81 (m, 2H), 6.57 (dd, *J* = 7.5, 1.6 Hz, 1H), 3.65 (d, *J* = 14.0 Hz, 1H), 3.40 (d, *J* = 14.0 Hz, 1H), 1.96 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.0, 151.2, 143.3, 140.3, 134.5, 132.9, 132.3, 130.7, 130.3, 129.7, 129.3, 128.8, 128.3, 127.1, 126.8, 126.1, 125.5, 122.8, 120.8, 116.9, 50.4, 48.6, 24.9. **HRMS-ESI** (m/z): calcd for $C_{23}H_{16}BrCl_2N_2O$ $[M + H]^+$ 486.9798; found 486.9797.

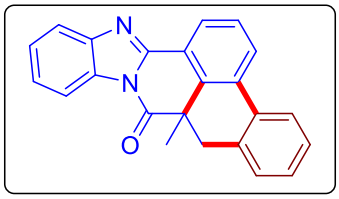
5-(2-Bromobenzyl)-3,5-dimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one (7e)



The compound was prepared according to GP using 2-methyl-1-(2-(*p*-tolyl)-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (0.069 g, 0.25 mmol), bis(2-bromophenyl)iodonium trifluoromethanesulfonate (0.220 g, 0.375 mmol) and $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%). After 12 h, purification by column chromatography (0-5% ethyl acetate in hexane) gave **7e** as a white solid (0.070 g, 64%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.35 (dd, *J* = 6.4, 2.8 Hz, 1H), 8.24 (d, *J* = 7.9 Hz, 1H), 7.76 (dd, *J* = 6.3, 2.8 Hz, 1H), 7.44 – 7.40 (m, 2H), 7.31 – 7.27 (m, 2H), 7.26 (s, 1H), 6.88 – 6.81 (m, 2H), 6.65 (dd, *J* = 7.6, 1.7 Hz, 1H), 3.68 (d, *J* = 14.2 Hz, 1H), 3.48 (d, *J* = 14.2 Hz, 1H), 2.43 (s, 3H), 1.93 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 172.6, 149.9, 143.8, 142.21, 140.1, 135.1, 132.8, 131.2, 130.6, 129.1, 128.5, 127.6, 126.8, 125.9, 125.8, 125.5, 125.3, 120.4, 119.5, 115.5, 50.2, 47.8, 25.8, 22.0. **HRMS-ESI** (m/z): calcd for $C_{24}H_{20}BrN_2O$ $[M + H]^+$ 433.0734; found 433.0736.

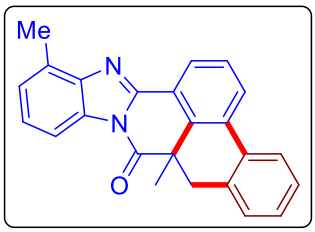
8a-Methyl-8,8a-dihydro-9*H*-dibenzo[*de,g*]benzo[4,5]imidazo[1,2-*b*]isoquinolin-9-one (8a)



The compound was prepared according to HAS procedure using 5-(2-bromobenzyl)-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one **3as** (0.063 g, 0.15 mmol). After 12 h, purification by column chromatography (20-40% DCM in hexane) gave **8a** as a white solid (0.031 g, 62%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.32 (dd, *J* = 7.8, 1.0 Hz, 1H), 8.28 – 8.25 (m, 1H), 7.93 – 7.89 (m, 1H), 7.81 – 7.75 (m, 2H), 7.51 (t, *J* = 7.8 Hz, 1H), 7.39 – 7.36 (m, 2H), 7.34 (dd, *J* = 10.9, 4.3 Hz, 2H), 7.31 – 7.28 (m, 1H), 3.44 (d, *J* = 15.6 Hz, 1H), 3.23 (d, *J* = 15.5 Hz, 1H), 1.37 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.4, 149.8, 144.2, 137.0, 133.5, 133.2, 132.1, 131.4, 129.9, 129.0, 128.7, 127.8, 127.0, 125.9, 125.7, 125.5, 123.8, 122.1, 119.9, 115.4, 44.0, 35.9, 26.4. **HRMS-ESI** (m/z): calcd for $C_{23}H_{17}N_2O$ $[M + H]^+$ 337.1335; found 337.1337.

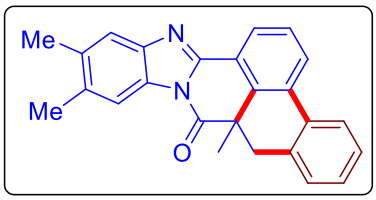
8a,14-Dimethyl-8,8*a*-dihydro-9*H*-dibenzo[*de,g*]benzo[4,5]imidazo[1,2-*b*]isoquinolin-9-one (8b)



The compound was prepared according to HAS procedure using 5-(2-bromobenzyl)-5,11-dimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one **7b** (0.065 g, 0.15 mmol). After 12 h, purification by column chromatography (20-40% DCM in hexane) gave **8b** as a white solid (0.031 g, 62%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.54 (d, *J* = 7.6 Hz, 1H), 8.19 (d, *J* = 8.0 Hz, 1H), 8.02 – 7.99 (m, 1H), 7.89 (d, *J* = 7.2 Hz, 1H), 7.61 (t, *J* = 7.8 Hz, 1H), 7.46 – 7.35 (m, 5H), 3.53 (d, *J* = 15.6 Hz, 1H), 3.33 (d, *J* = 15.6 Hz, 1H), 2.79 (s, 3H), 1.46 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.5, 149.0, 136.9, 133.4, 133.2, 132.1, 131.4, 131.1, 130.1, 129.9, 128.9, 128.5, 127.8, 126.8, 126.4, 125.7, 125.6, 123.8, 122.2, 112.7, 44.0, 35.9, 26.4, 16.7. **HRMS-ESI** (m/z): calcd for $C_{24}H_{19}N_2O$ $[M + H]^+$ 351.1492; found 351.1493.

8a,12,13-Trimethyl-8,8a-dihydro-9*H*-dibenzo[*de,g*]benzo[4,5]imidazo[1,2-*b*]isoquinolin-9-one (8c)

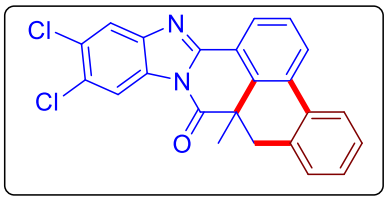


The compound was prepared according to HAS procedure using 5-(2-Bromobenzyl)-5,9,10-trimethylbenzo[4,5]imidazo [2,1-*a*]isoquinolin-6(5*H*)-one **7c** (0.067 g, 0.15 mmol). After 12 h, purification by column chromatography (20-40% DCM in hexane) gave **8c** as a white solid (0.031 g, 56%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.29 (dd, *J* = 7.8, 1.0 Hz, 1H), 8.04 (s, 1H), 7.90 – 7.87 (m, 1H), 7.81 – 7.77 (m, 1H), 7.50 (dd, *J* = 14.5, 6.7 Hz, 2H), 7.33 (t, *J* = 6.4 Hz, 2H), 7.31 – 7.27 (m, 1H), 3.42 (d, *J* = 15.6 Hz, 1H), 3.22 (d, *J* = 15.6 Hz, 1H), 2.35 (d, *J* = 11.1 Hz, 6H), 1.35 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, CDCl3) δ 173.4, 149.1, 142.7, 136.8, 135.1, 134.8, 133.4, 133.3, 132.2, 129.9, 129.7, 128.9, 128.6, 127.8, 126.6, 125.3, 123.8, 122.3, 120.1,

115.6, 43.9, 35.9, 26.4, 20.5, 20.5. **HRMS-ESI** (m/z): calcd for $C_{25}H_{20}N_2O$ $[M + H]^+$ 365.1648; found 365.1648.

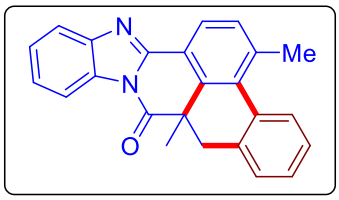
12,13-Dichloro-8a-methyl-8,8a-dihydro-9*H*-dibenzo[*de,g*]benzo[4,5]imidazo[1,2-*b*]isoquinolin-9-one (8d)



The compound was prepared according to HAS procedure using 5-(2-bromobenzyl)-9,10-dichloro-5-methylbenzo[4,5] imidazo-[2,1-*a*]isoquinolin-6(5*H*)-one **7d** (0.073 g, 0.15 mmol). After 12 h, purification by column chromatography (20-40% DCM in hexane) gave **8d** as a white solid (0.027 g, 45%).

^{1}H NMR (500 MHz, $CDCl_3$) δ 8.48 (s, 1H), 8.37 (d, *J* = 7.7 Hz, 1H), 8.04 (d, *J* = 7.7 Hz, 1H), 7.93 (s, 1H), 7.89 (d, *J* = 7.5 Hz, 1H), 7.62 (t, *J* = 7.8 Hz, 1H), 7.47 – 7.39 (m, 3H), 3.52 (d, *J* = 15.6 Hz, 1H), 3.32 (d, *J* = 15.6 Hz, 1H), 1.47 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.2, 151.5, 143.7, 137.2, 133.7, 133.0, 131.9, 130.5, 130.0 (X 2C), 129.6, 129.2, 128.9, 128.0, 127.7, 125.8, 124.0, 121.5, 121.2, 116.8, 44.2, 35.8, 26.5. **HRMS-ESI** (m/z): calcd for $C_{23}H_{15}Cl_2N_2O$ $[M + H]^+$ 405.0556; found 405.0559.

3,8a-Dimethyl-8,8a-dihydro-9*H*-dibenzo[*de,g*]benzo[4,5]imidazo[1,2-*b*]isoquinolin-9-one (8e)

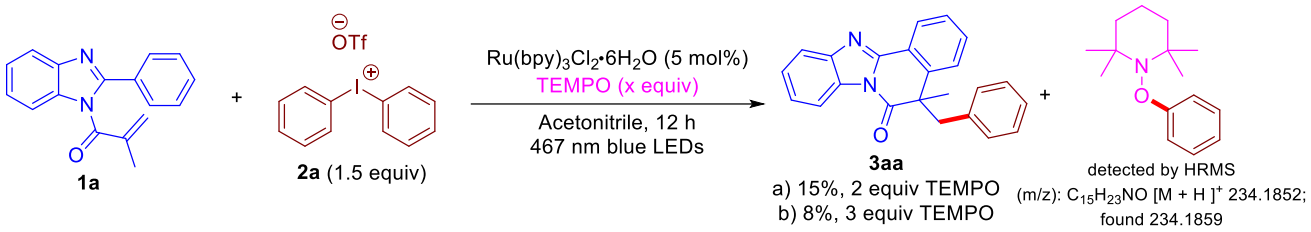


The compound was prepared according to HAS procedure using 5-(2-bromobenzyl)-3,5-dimethylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one **7e** (0.065 g, 0.15 mmol). After 12 h, purification by column chromatography (20-40% DCM in hexane) gave **8e** as a white solid (0.036 g, 69%).

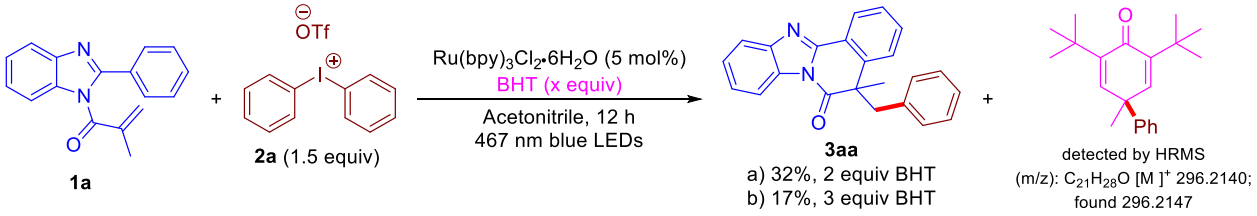
^{1}H NMR (500 MHz, $CDCl_3$) δ 8.35 – 8.30 (m, 2H), 7.85 (dd, *J* = 6.3, 2.4 Hz, 1H), 7.80 (d, *J* = 7.5 Hz, 1H), 7.50 (d, *J* = 8.1 Hz, 1H), 7.48 – 7.44 (m, 3H), 7.42 (d, *J* = 7.4 Hz, 1H), 7.39 (dd, *J* = 10.5, 4.2 Hz, 1H), 3.48 (d, *J* = 14.9 Hz, 1H), 3.22 (d, *J* = 14.9 Hz, 1H), 2.76 (s, 3H), 1.32 (s, 3H). **^{13}C{^{1}H} NMR** (126 MHz, $CDCl_3$) δ 173.5, 150.2, 144.3, 138.9, 138.9, 135.1, 133.3, 132.7, 132.5, 131.3, 129.7, 128.5, 128.2, 126.8, 125.8, 125.4, 124.8, 119.8, 119.3, 115.3, 44.9, 36.5, 24.9, 23.7. **HRMS-ESI** (m/z): calcd for $C_{24}H_{19}N_2O$ $[M + H]^+$ 351.1492; found 351.1493.

12. Control Experiments:

a) Control Experiment with TEMPO:



b) Control Experiment with BHT:

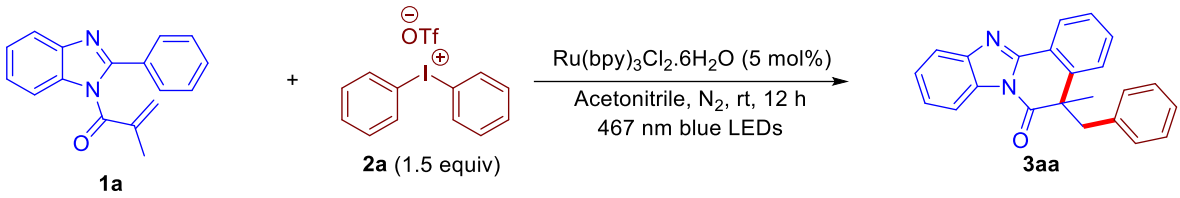


(a) 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one **1a** (40 mg, 0.25 mmol, 1.0 equiv), diphenyliodonium trifluoromethanesulfonate **2a** (166 mg, 0.75 mmol, 3.0 equiv), $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%), and 2,2,6,6-tetramethylpiperidin-1- yloxyl (TEMPO) (either 0.50 mmol, 2 equiv or 0.75 mmol, 3 equiv) were added in a pre-dried 10 ml Schlenk tube under N_2 atmosphere. The tube was degassed and purged with N_2 three times. Then dry acetonitrile (2.5 ml) was added under the N_2 atmosphere. Then the mixture was allowed to stir for 24 h under irradiation of 40 W Kessil blue LED (467 nm) lamp. After completion, the reaction mixture was concentrated under vacuum, and purified by silica gel column chromatography using 15% ethyl acetate in hexane to afford the 5-benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one. Formation of adduct was confirmed by HRMS.

(b) 2-methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one **1a** (40 mg, 0.25 mmol, 1.0 equiv), diphenyliodonium trifluoromethanesulfonate **2a** (166 mg, 0.75 mmol, 3.0 equiv) $Ru(bpy)_3Cl_2.6H_2O$ (9.35 mg, 0.0125 mmol, 5 mol%), and 2,6-di-tert-butyl-4-methylphenol (BHT) (either 0.50 mmol, 2 equiv or 0.75 mmol, 3 equiv) were added in a pre-dried 10 ml Schlenk tube under N_2 atmosphere. The tube was degassed and purged with N_2 three times. Then dry acetonitrile (2.5 ml) was added under the N_2 atmosphere. Then the mixture was allowed to stir for 24 h under irradiation of 40 W Kessil blue LED (467 nm) lamp. After completion, the reaction mixture was concentrated under vacuum, and purified by silica gel column chromatography using 15% ethyl acetate in hexane to afford the 5-

benzyl-5-methylbenzo[4,5]imidazo[2,1-*a*]isoquinolin-6(5*H*)-one. The formation of adduct was confirmed by HRMS.

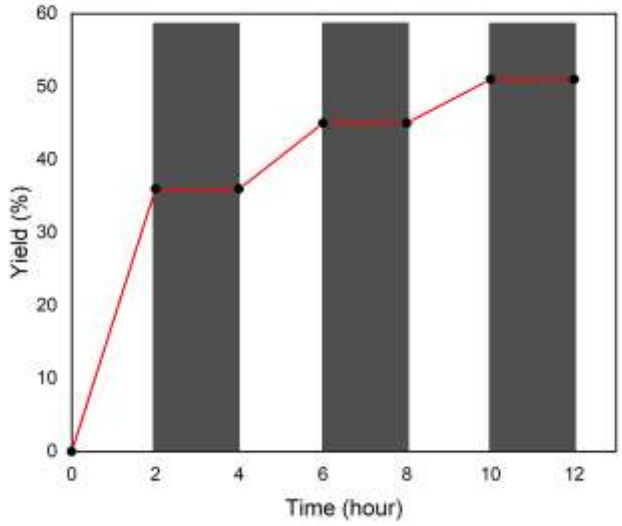
13. Light- dark experiment:



1a **2a** (1.5 equiv) **3aa**

2-Methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one **1a** (0.079 g, 0.3 mmol, 1.0 equiv), diphenyliodonium trifluoromethanesulfonate **2a** (0.193 g, 0.45 mmol, 1.5 equiv) and $Ru(bpy)_3Cl_2.6H_2O$ (11.22 mg, 0.015 mmol, 5 mol%) were added in a pre-dried 10 ml schlenk tube under N_2 atmosphere. The tube was degassed and purged with N_2 three times. Then dry acetonitrile (3 ml) was added under the N_2 atmosphere. The mixture was irradiated using a 40 W Kessil blue LED (467 nm) lamp and the reaction was placed in light and dark in every alternative 2 hour. After every time interval of 2 hour a 0.5 mL reaction aliquot was taken out by a syringe and quenched with water, organic part taken in ethyl acetate and NMR was carried out. The yield was determined using mesitylene as a NMR standard.

Entry	Time (h)	Light source	Yield (%)
1	2	on	36
2	4	off	36
3	6	on	45
4	8	off	45
5	10	on	51
6	12	off	51



14. Determination of Quantum Yield:

(A) Determination of light intensity of the Blue LED:

The photon flux was determined by using standard ferrioxalate actinometry.7 A 0.15 M solution of ferrioxalate was made by mixing 0.737 g of potassium ferrioxalate hydrate in 10 mL of 0.05 M H_2SO_4. A buffered solution of phenanthroline was made by mixing 25 mg of phenanthroline and 5.63 g of sodium acetate in 25 mL of 0.5 M H_2SO_4. Both solutions were kept in the dark. To determine the photon flux, 1.0 mL of the ferrioxalate solution was placed in a cuvette and irradiated for 60.0 seconds at λ = 467 nm placing 2 cm away from 40 W Kessil blue LED lamp. After irradiation, 0.175 mL of the phenanthroline solution was added to the cuvette. The solution was then kept for 1 h to permit the ferrous ions to completely coordinate to the phenanthroline. The absorbance of the solution was determined at 510 nm. A non-irradiated sample was also prepared and the absorbance at 510 nm was determined. Conversion was calculated using eq. 1

$$\text{mol } Fe^{2+} = \frac{V \cdot \Delta A}{\varepsilon \cdot l} \quad \text{.....} \mathbf{1}$$

Where V is the total volume (0.001175 L) of the solution after addition of the phenanthroline, ΔA is the difference in absorbance at 510 nm between the irradiated and non-irradiated solutions, l is the path length (1.000 cm), and ε is the molar absorptivity at 510 nm (11,100 L mol^{-1} cm^{-1}).

$$\text{mol } Fe^{2+} = \frac{0.001175 \text{ L. } (0.837818 - 0.213601)}{1 \text{ cm . } 11100 \text{ L.cm}^{-1}\text{. mol}^{-1}}$$

$$= \quad 6.6077 \times 10^{-8} \text{ mol}$$

The photon flux can be calculated using eq 2.

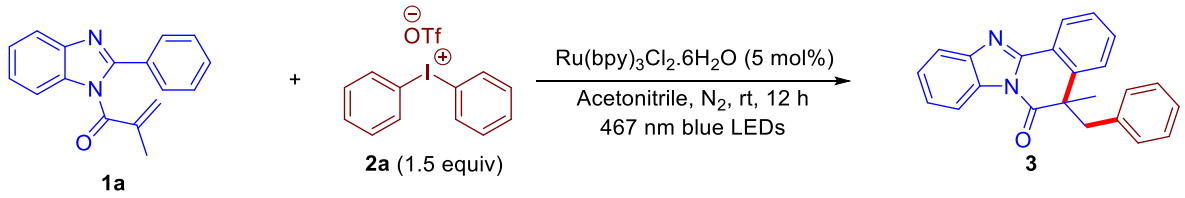
$$\text{photon flux} = \frac{\text{mol } Fe^{2+}}{\Phi \text{ . t . f}} \qquad \text{.....} \mathbf{2}$$

Where Φ is the quantum yield for the ferrioxalate actinometer (1.01 for a 0.15 M solution at λ = 467 nm), t is the time (60 s), and f is the fraction of light absorbed at λ = 467 nm, f = 1.000 -10^{-A}.

Calculated f = 1.000 – 10^{-A} = 1.000-$10^{-0.8378}$ = 0.8547

$$\text{photon flux} = \frac{6.6077 \times 10^{-8} \text{ mol}}{1.01 \times 60 \text{ s} \times 0.8547} = 12.757 \times 10^{-10} \text{ Einstein s}^{-1}$$

(B) Quantum Yield Calculation:



2-Methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one **1a** (0.079 g, 0.3 mmol, 1.0 equiv), diphenyliodonium trifluoromethanesulfonate **2a** (0.193 g, 0.45 mmol, 1.5 equiv) and $Ru(bpy)_3Cl_2.6H_2O$ (11.22 mg, 0.015 mmol, 5 mol%) were added in a pre-dried 10 ml schlenk tube under N_2 atmosphere. The tube was degassed and purged with N_2 three times. Then dry acetonitrile (3 ml) was added under the N_2 atmosphere. The mixture was irradiated for 2 h using a 40 W Kessil blue LED (467 nm) lamp. After 2 h, a 0.5 mL reaction aliquot was taken out by a syringe and quenched with water, organic part was taken in ethyl aceteate and ^1H NMR was carried out. The yield was determined using mesitylene as an internal NMR standard.

The quantum yield was calculated as follows:

$$\Phi = \frac{\text{mol product}}{\text{flux . t . f}}$$

Where, flux is the photon flux determined by ferrioxalate actinometry (12.757×10^{-10} Einstein/s), t is the time (7200 s), and f (> 0.999) is the fraction of light absorbed by $Ru(bpy)_3Cl_2.6H_2O$ at 467 nm under the reaction condition mentioned above.

$$\Phi = \frac{1.8 \times 10^{-5}}{12.757 \times 10^{-10} \times 7200 \times 1}$$

$$= 1.96$$

15. Luminescence quenching Experiments:

(A) **Preparation of the Stock Solution:**

A 0.5 mM solution of the $Ru(bpy)_3Cl_2.6H_2O$ catalyst was prepared in a sample vial by dissolving 1.87 mg of the catalyst in 5 mL of acetonitrile (spectroscopic grade, purchased from Spectrochem). The freshly prepared solution was used for the spectroscopic measurement. The required amount was taken using a micropipette from the mother solution as an aliquot and it was diluted further by dissolving in 3 mL of acetonitrile in the cuvette. Similarly, 10 mL 0.1 M solution of 2-Methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one **1a**, diphenyliodonium trifluoromethanesulfonate **2a** were prepared by dissolving the requisite amount of each substrate in acetonitrile. Freshly prepared solutions were used for the quenching experiment.

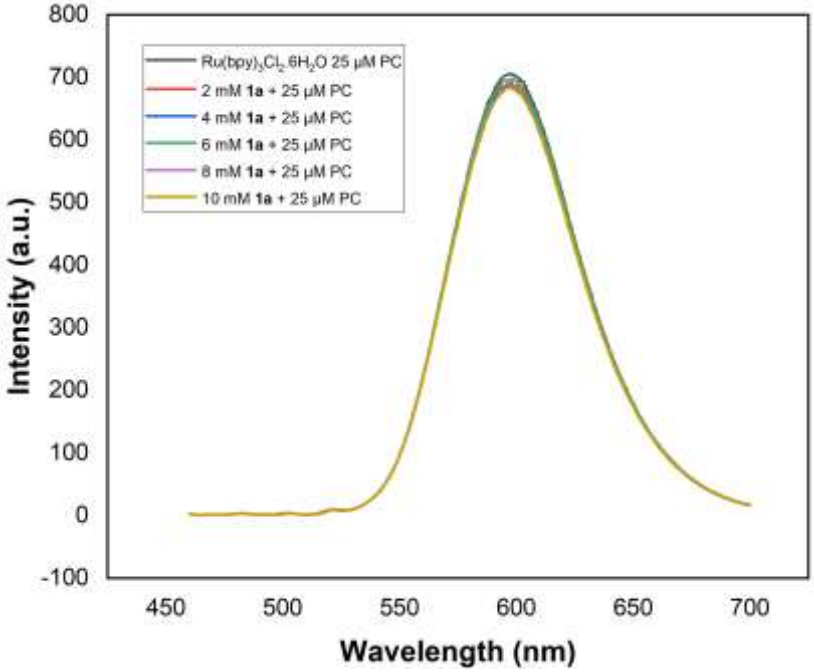
(B) Quenching studies:

Fluorescence emission spectra of the photocatalyst in presence of different reactions components (**1a, 2a**) were recorded and analyzed in detail to estimate the light emission properties of the pure catalyst system and their distractions by external interference from the substrates. Emission intensities of $Ru(bpy)_3Cl_2.6H_2O$ were recorded with a "HITACHI f-7000" Scientific Spectrofluorometer using a 10.0 mm quartz cuvette. The catalyst exhibits an absorption maximum at 451 nm (confirmed from literature). Hence, the sample solution of $Ru(bpy)_3Cl_2.6H_2O$ with a proper concentration of 25 **µ**M in acetonitrile was excited (degassed for 15 mins before recording the spectra) with a wavelength of 451 nm, and the emission maxima were found to be observed at 599 nm. The substrates **1a** was silent to show any emission feature in that region. To study the quenching behaviour of $Ru(bpy)_3Cl_2.6H_2O$, different concentration of **2a** was added to the catalyst solution and the emission spectra were measured following the aforementioned procedure. The quenching effect **2a** was quite significant on the photocatalyst; the intensity of the emission maxima decreased gradually upon increasing the concentration of **2a**. Some sets of solutions with different concentrations of the **2a** were used; the experiment was repeated and finally, the Stern-Volmer plot was depicted. On the other hand, no significant change in the emission maxima of $Ru(bpy)_3Cl_2.6H_2O$ was observed when 2-Methyl-1-(2-phenyl-1*H*-benzo[d]imidazol-1-

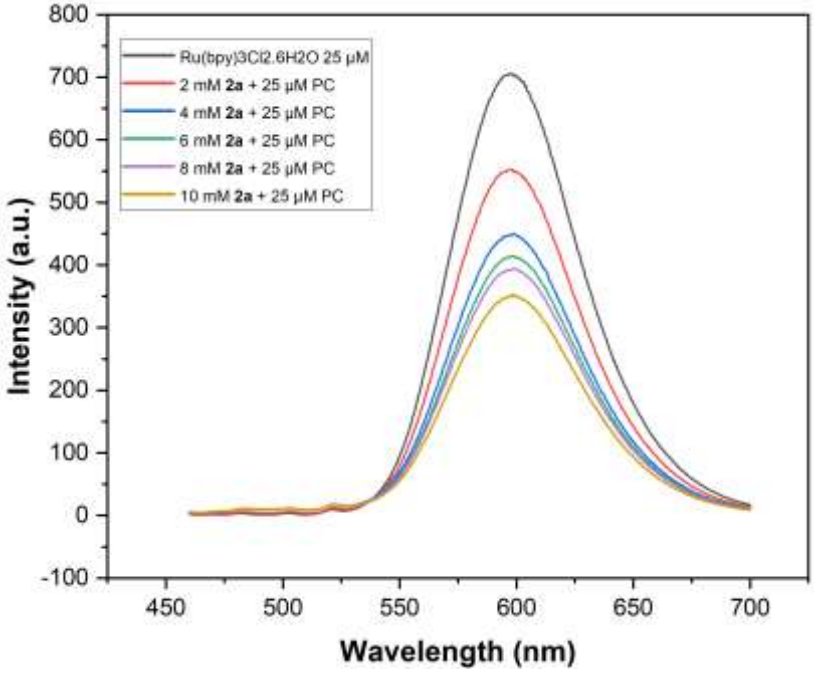
yl)prop-2-en-1-one **1a** was used as the quencher. The corresponding Stern-Volmer plot was drawn for all the cases.

Luminescence Spectra:

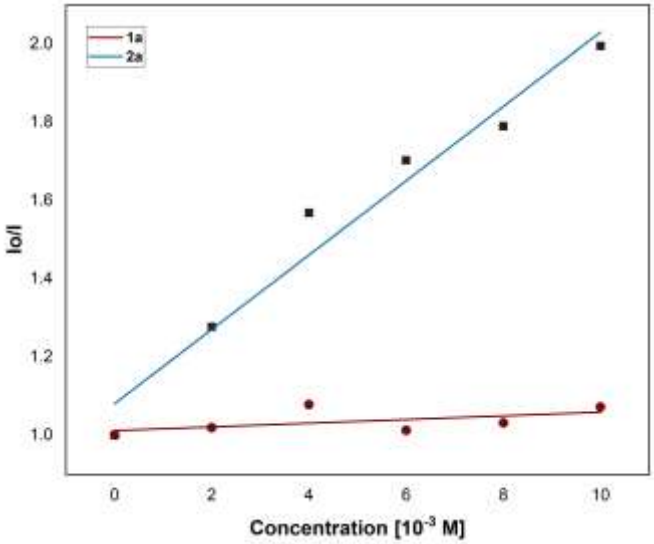
- **Luminescence spectra of $Ru(bpy)_3Cl_2.6H_2O$ (25 µM) as a function of concentration of 2-Methyl-1-(2-phenyl-1*H*-benzo[*d*]imidazol-1-yl)prop-2-en-1-one (1a) in CH_3CN with excitation at 467 nm**



- **Luminescence spectra of $Ru(bpy)_3Cl_2.6H_2O$ (25 µM) as a function of concentration of diphenyliodonium trifluoromethanesulfonate (2a) in CH_3CN with excitation at 467 nm**



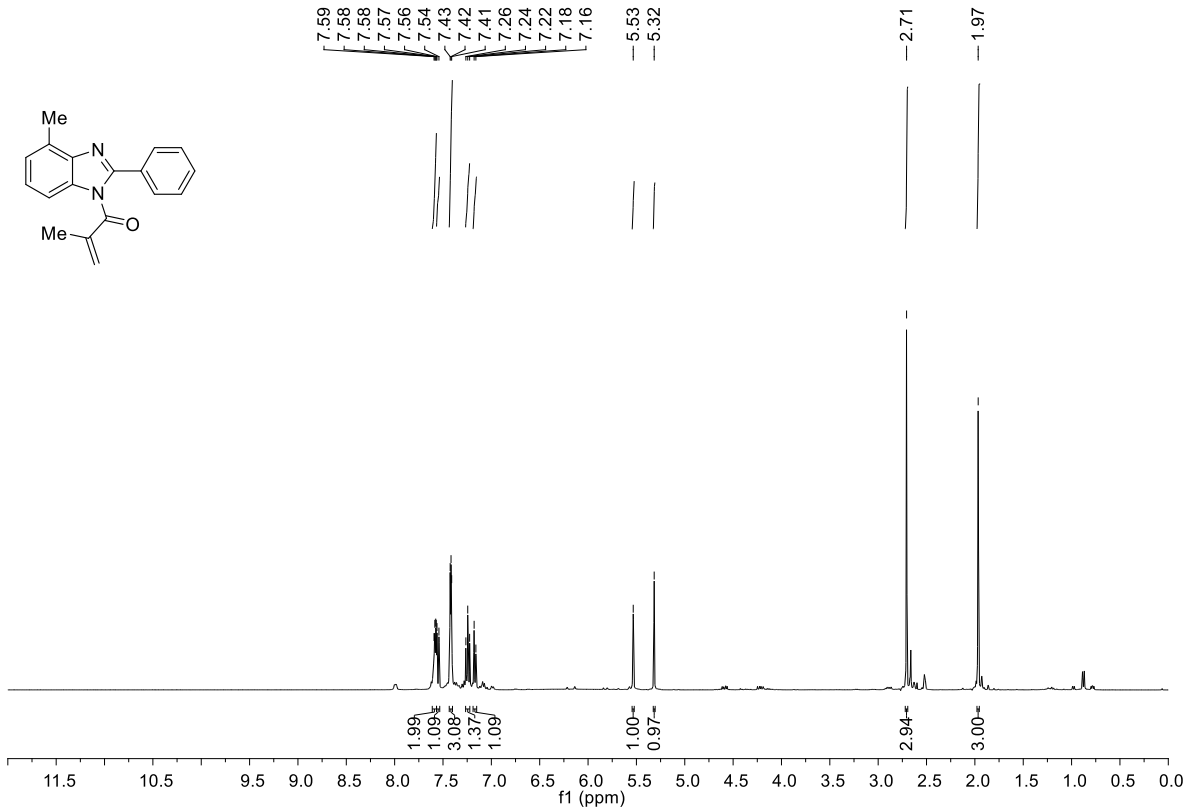
- **Stern-Volmer plot**



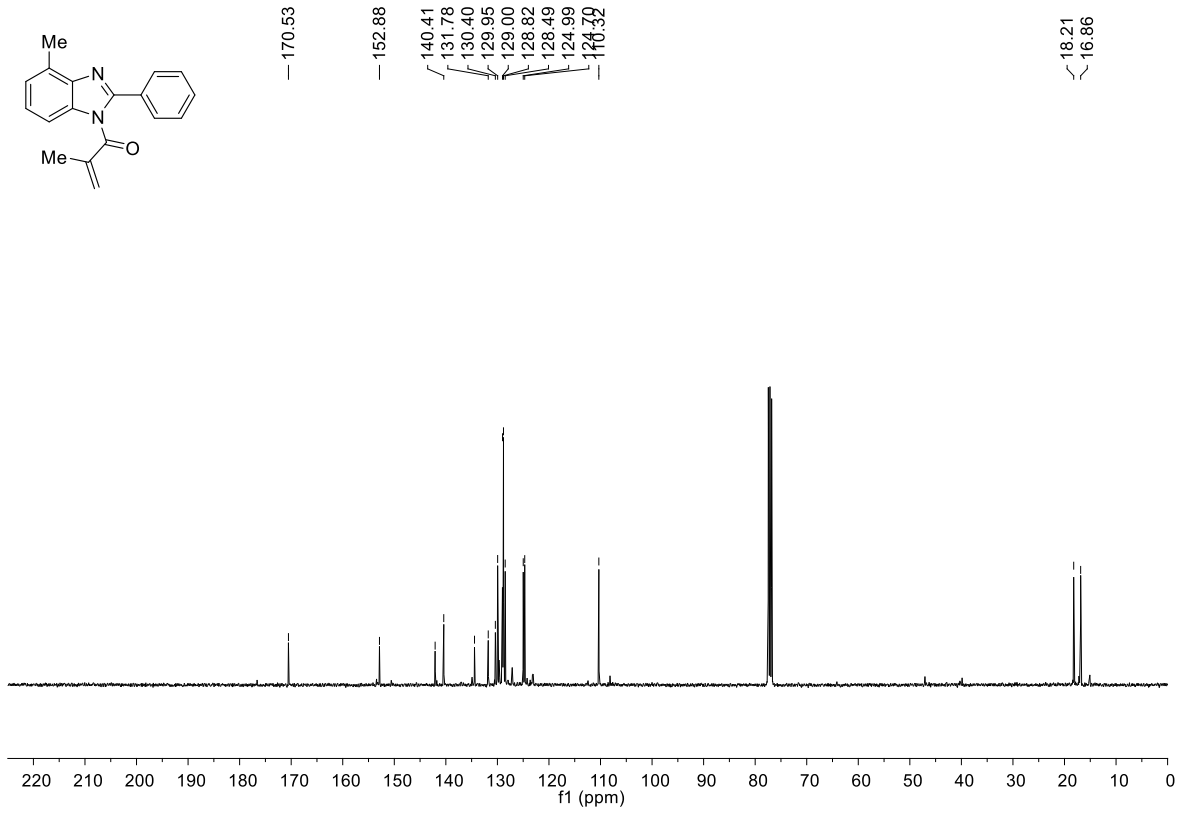
16. References:

1. Z. Chen, Zhu, M. Zhu, X. Huang, J. Sun, Y. Liu, Z. Li, *Asian J. Org. Chem.*, 2022, **11**, 8.
2. V. Ramesh, M. Gangadhar, J. B. Nanubolu, P. R. Adiyala, *J. Org. Chem.*, 2021, **86**, 12908-12921.
3. L. Liu, D. Y. Yang, Y. H. He, Z. Guan, *J. Org. Chem.*, 2020, **85**, 11892-11901.
4. B. Zhao, G. B. Hammond, B. Xu, *J. Org. Chem.*, 2021, **86**, 12851-12861.
5. C. Wang, G. Sun, H. L. Huang, J. Liu, H. Tang, Y. Li, H. Hu, S. He, F. Gao, *Chem. Asian J.*, 2021, **16**, 2618-2621.
6. C. Pan, C. Yuan, J. T. Yu, *Org. Bimol. Chem.*, 2021, **19**, 619-616.
7. B. Wang, L. Zou, L. Wang, M. Sun, P. Li, *Chin. Chem. Lett.*, 2021, **32**, 1229-1232.
8. M. Bielawski, M. Zhu, B. Olofsson, *Adv. Synth. Catal.*, 2007, **349**, 2610-2618.
9. P. K. Pramanick, Z. Zhou, Z. L. Hou, B. Yao, *J. Org. Chem.*, 2019, **84**, 5684-5694.
10. P. Villo, G. Kervefors, B. Olofsson, *Chem Commun.*, 2018, **54**, 8810-8813.
11. G. An, L. Wang, J. Han, *Org. Lett.*, 2021, **23**, 8688-8693.
12. X. Wu, J. Han, L. Wang, *J. Org. Chem.* 2018, **83**, 49-56.
13. X. Hong, Q. Tan, B. Liu, B. Xu, *Angew. Chem., Int. Ed.*, 2017, **56**, 3961-3965.
14. F. Zhang, S. Das, A. J. Walkinshaw, A. Casitas, M. Taylor, M. G. Suero, M. J. Gaunt, *J. Am. Chem. Soc.*, 2014, **136**, 8851-8854.
15. X. Zhou, Q. Wu, Y. Yu, C. Yu, E. Hao, Y. Wei, X. Mu, L. Jiao, *Org. Lett.*, 2016, **18**, 736-739.
16. N. Purkait, G. Kervefors, E. Linde, B. Olofsson, *Angew. Chem., Int. Ed.*, 2018, **57**, 11427-11431.

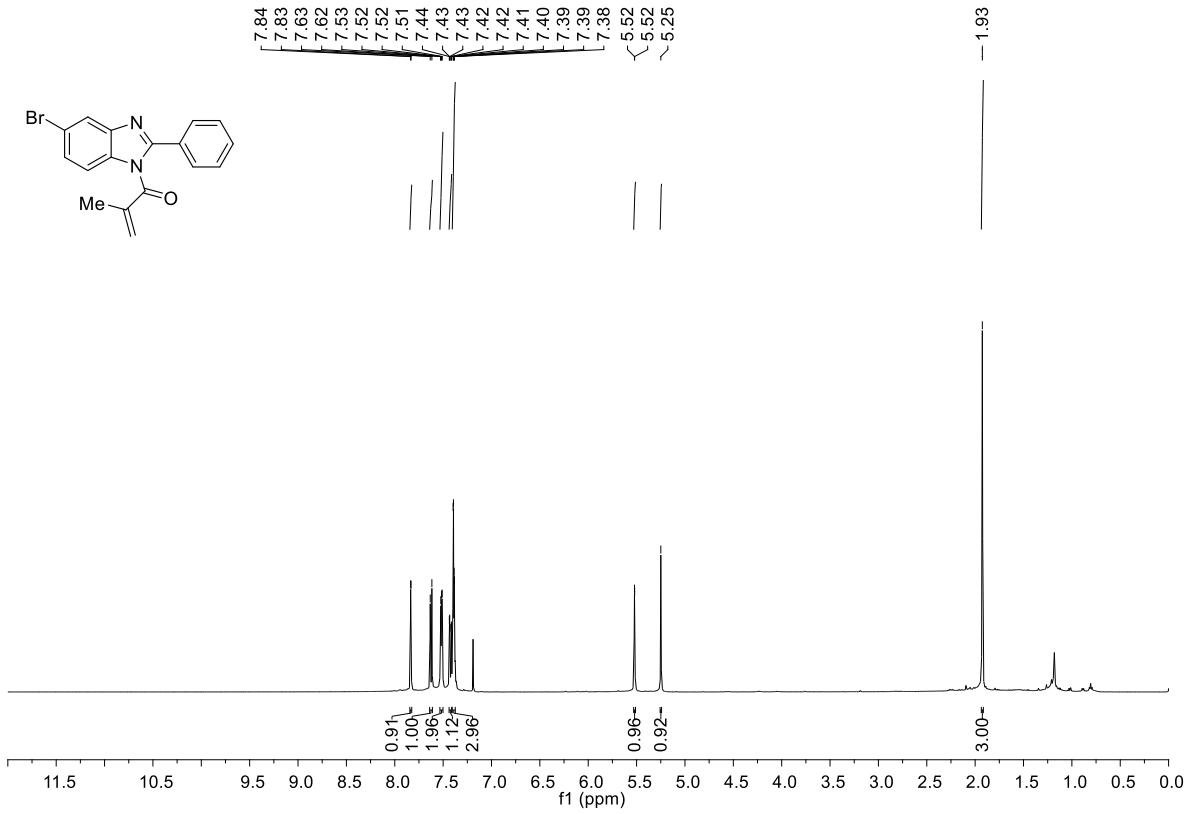
17. NMR Spectra of Compounds: ^{1}H NMR spectrum of 1q ($CDCl_3$, 400 MHz)



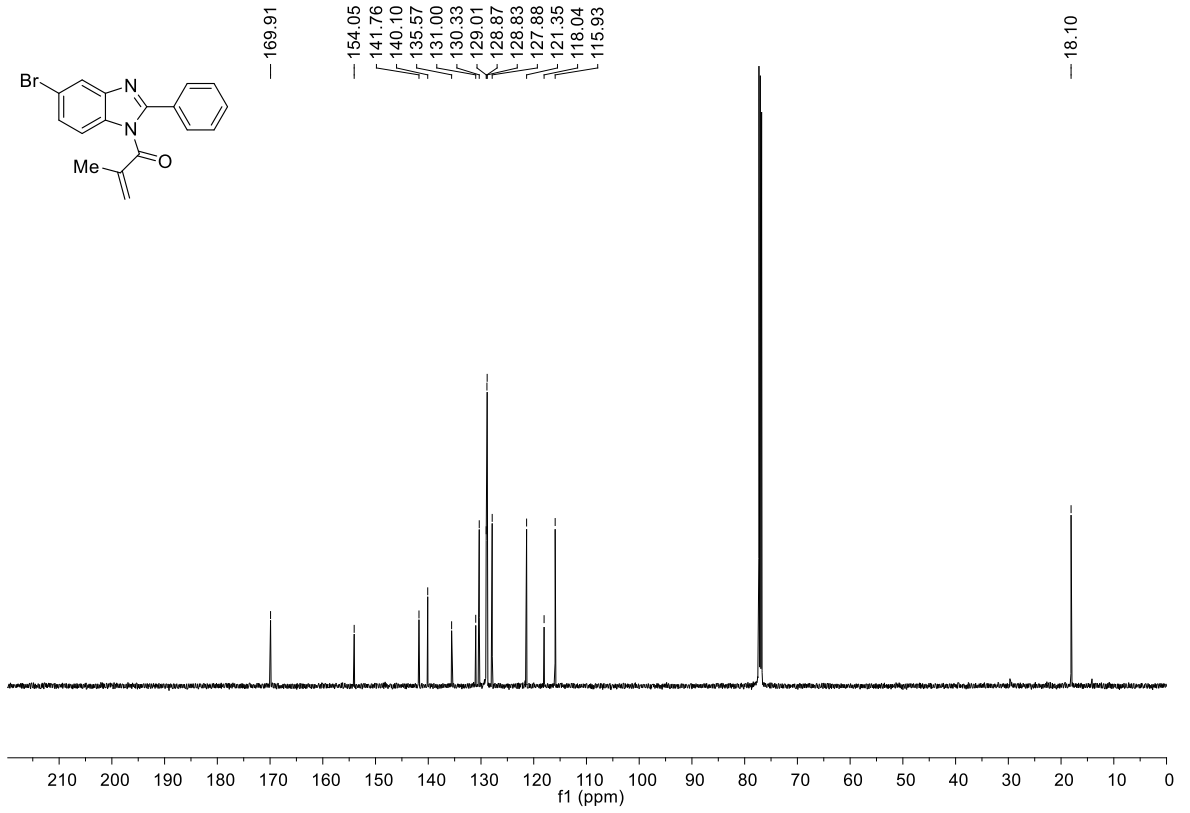
^{13}C{^{1}H} NMR spectrum of 1q ($CDCl_3$, 101 MHz)



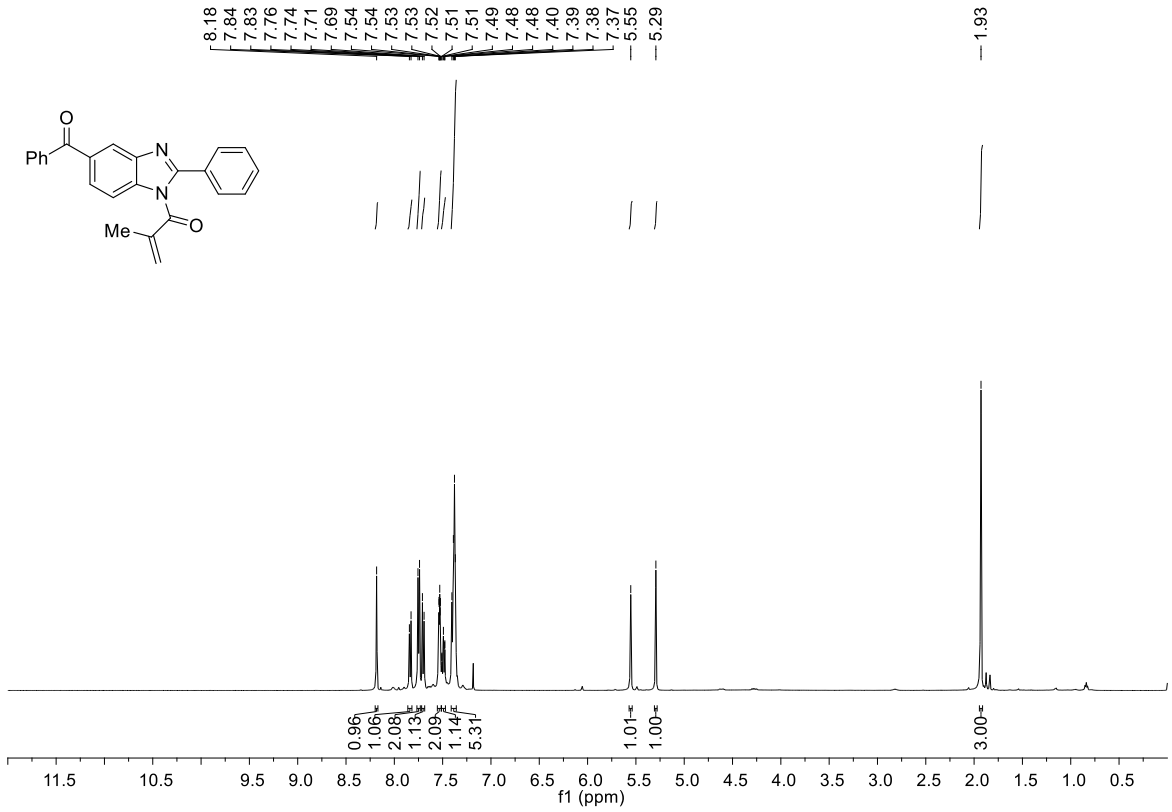
^{1}H NMR spectrum of 1r ($CDCl_3$, 400 MHz)



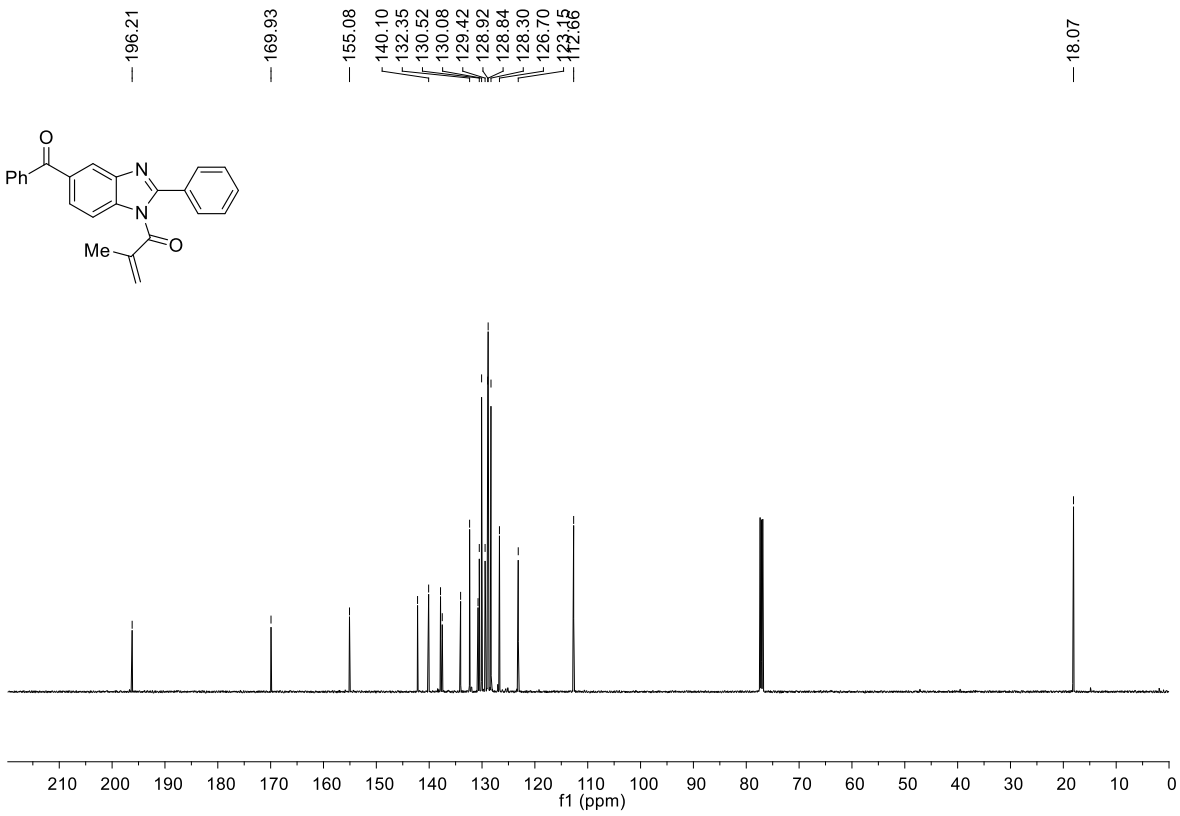
^{13}C{^{1}H} NMR spectrum of 1r ($CDCl_3$, 101 MHz)



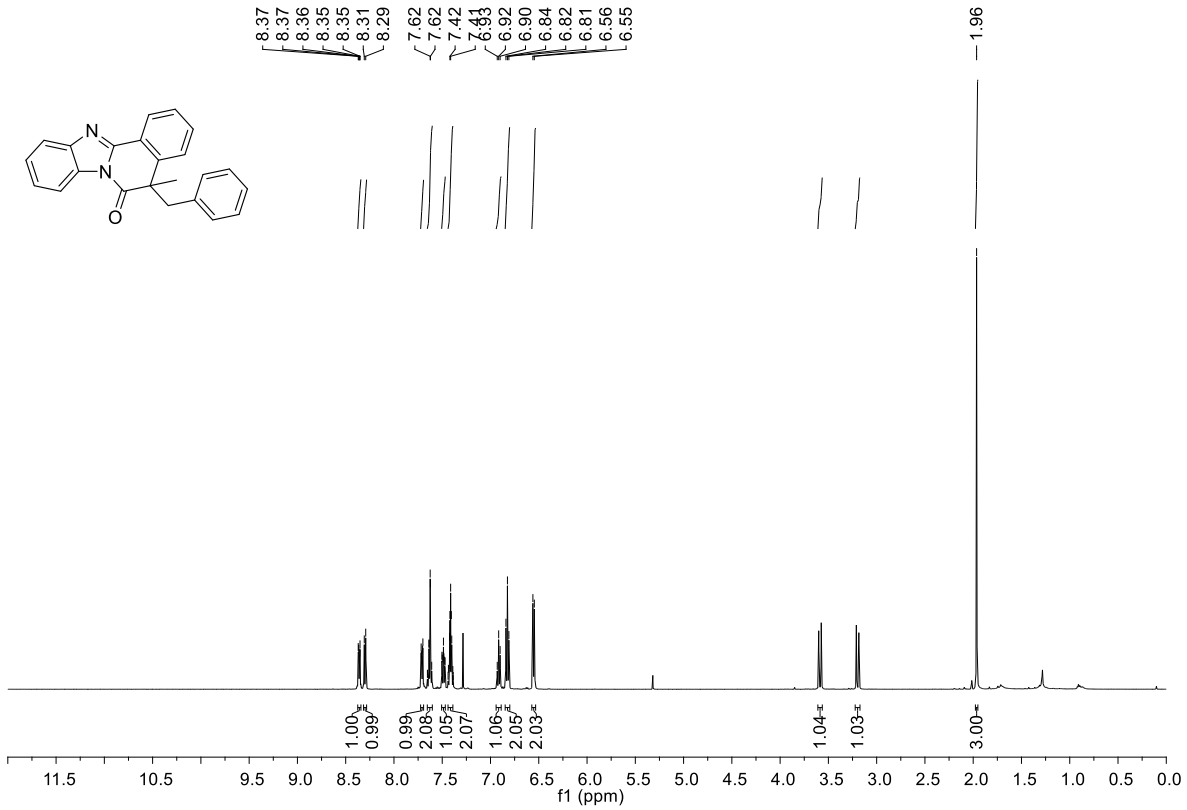
^{1}H NMR spectrum of 1s ($CDCl_3$, 500 MHz)



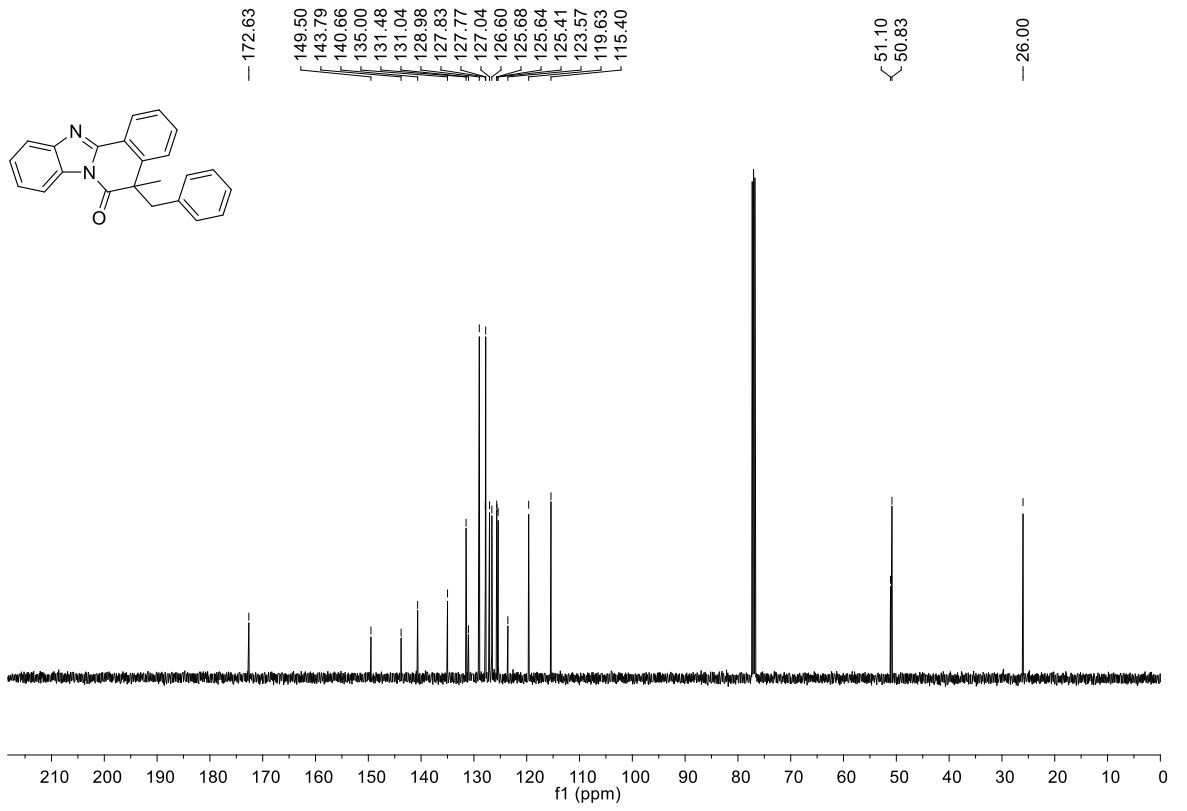
^{13}C{^{1}H} NMR spectrum of 1s ($CDCl_3$, 126 MHz)



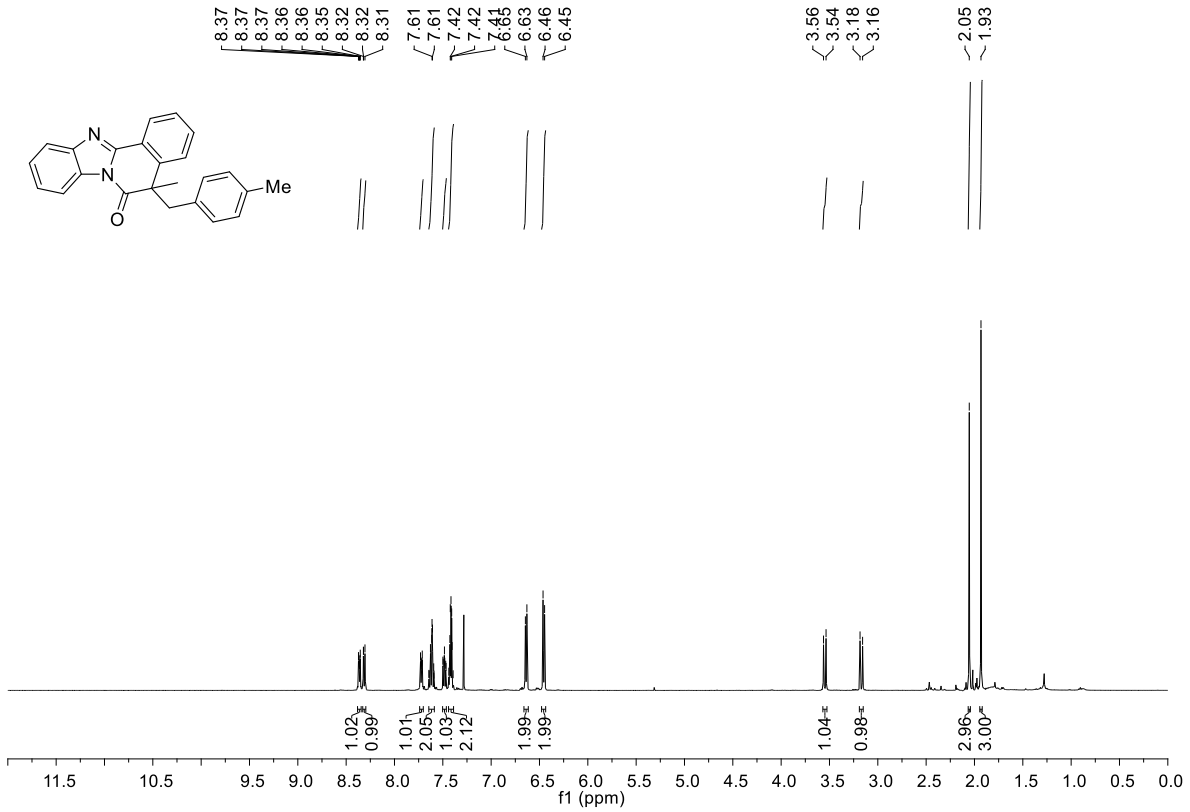
^{1}H NMR spectrum of 3aa ($CDCl_3$, 500 MHz)



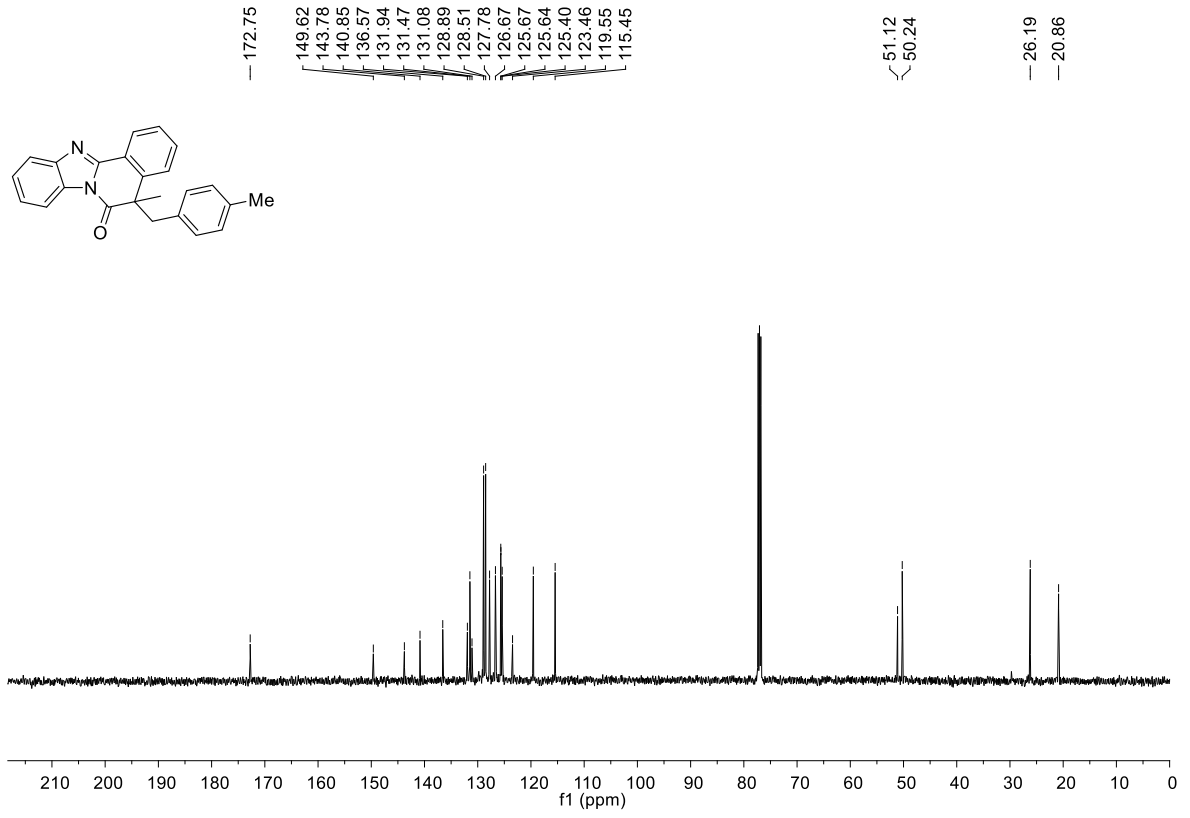
^{13}C{^{1}H} NMR spectrum of 3aa ($CDCl_3$, 126 MHz)



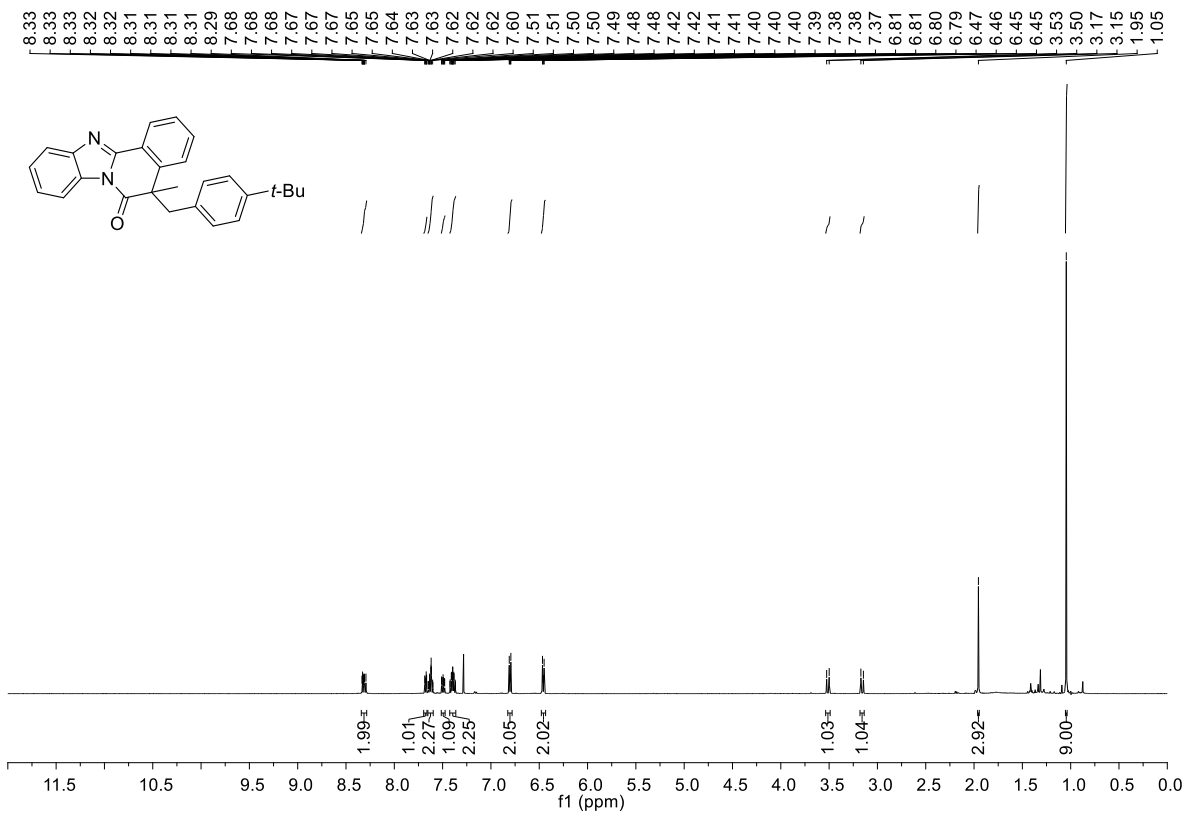
^{1}H NMR spectrum of 3ab ($CDCl_3$, 500 MHz)



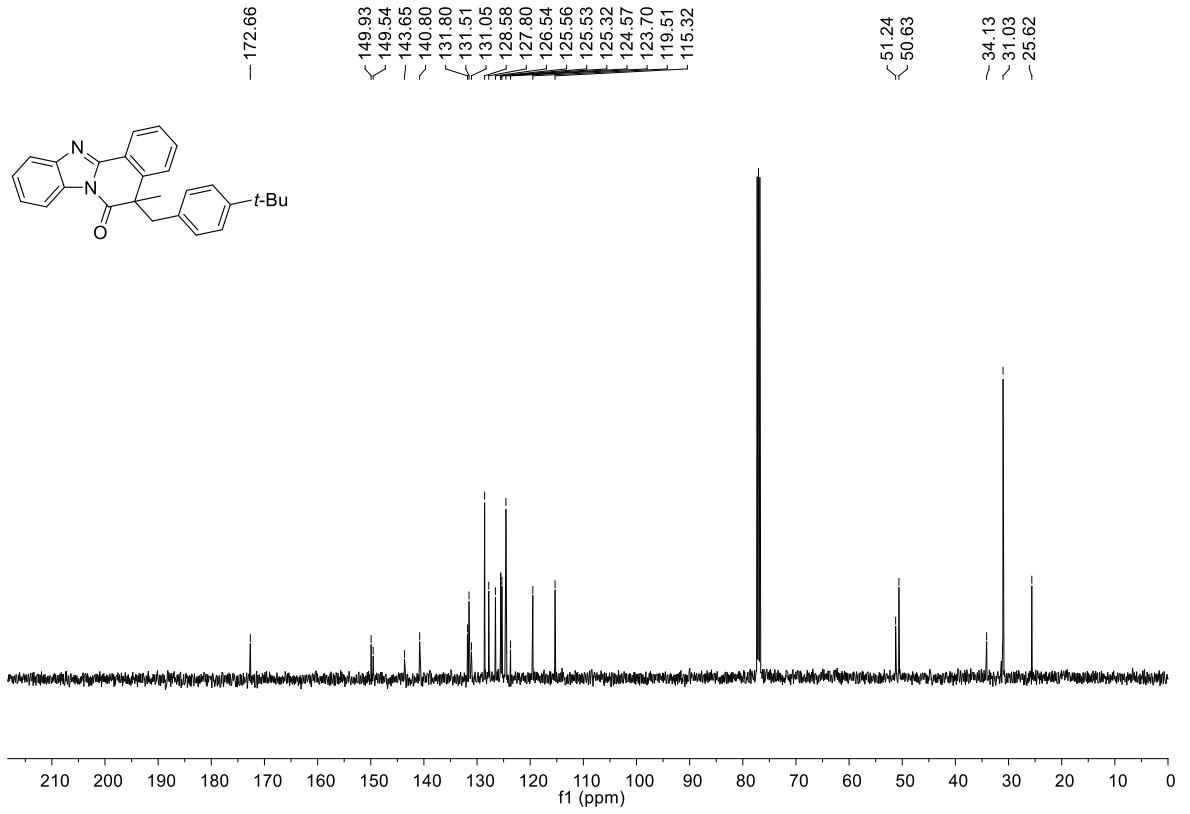
^{13}C{^{1}H} NMR spectrum of 3ab ($CDCl_3$, 126 MHz)



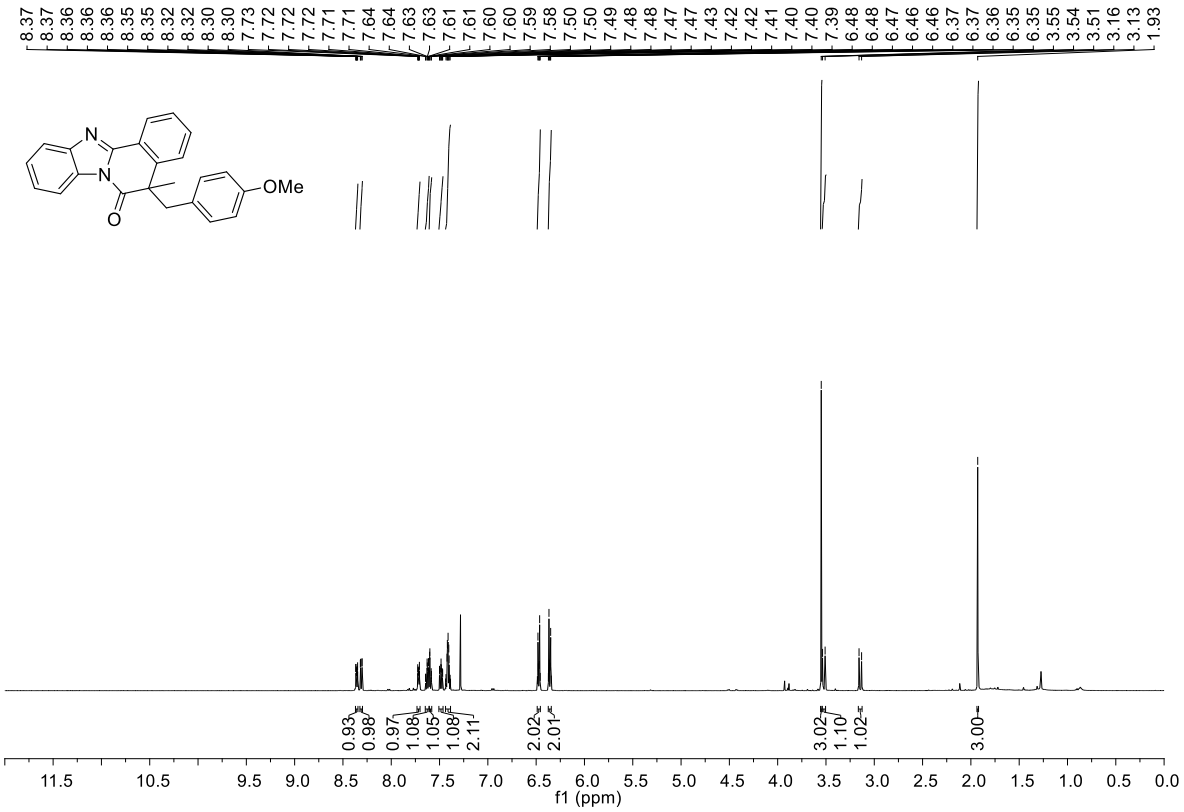
^{1}H NMR spectrum of 3ac ($CDCl_3$, 500 MHz)



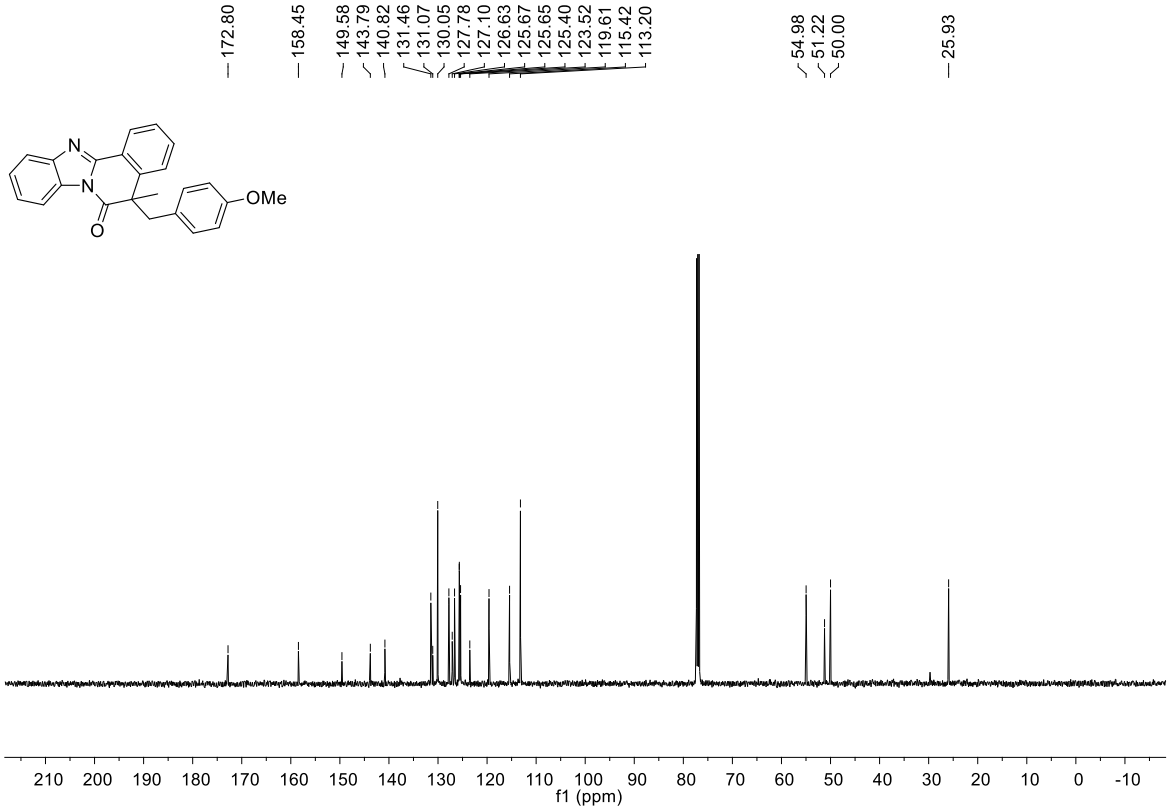
^{13}C{^{1}H} NMR spectrum of 3ac ($CDCl_3$, 126 MHz)



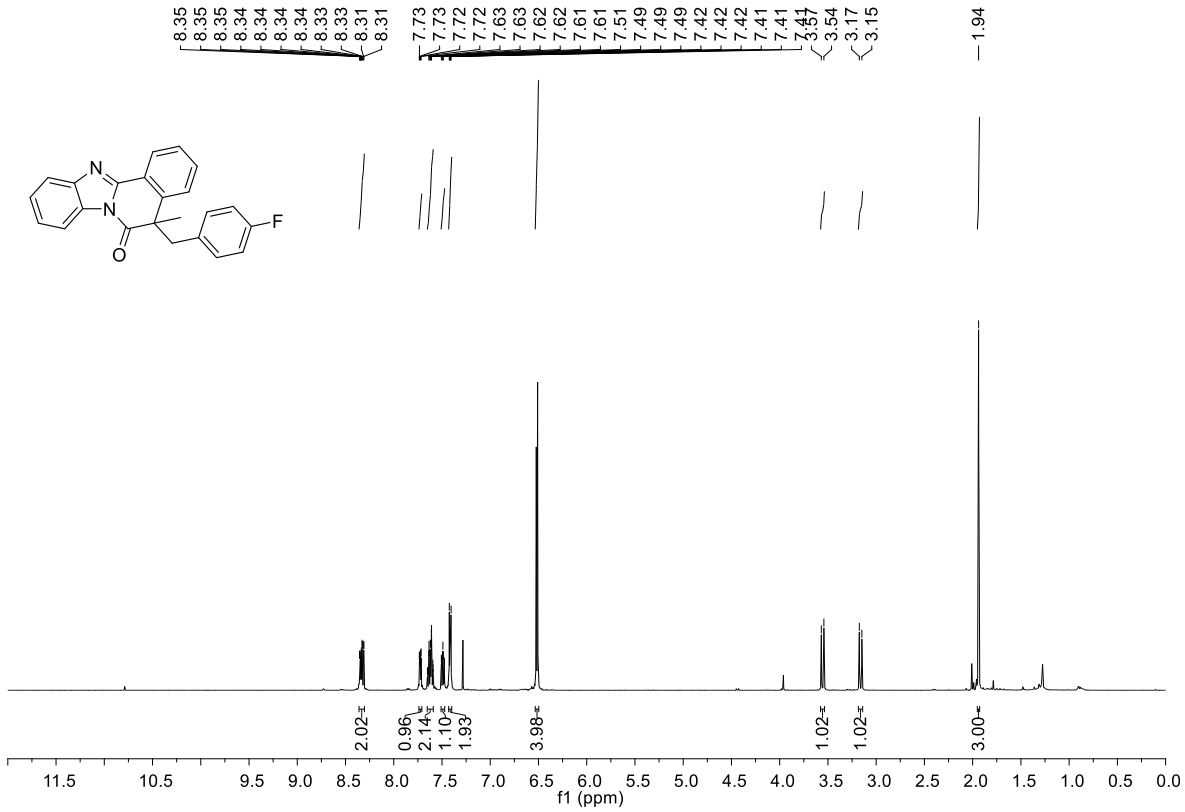
^{1}H NMR spectrum of 3ad ($CDCl_3$, 500 MHz)



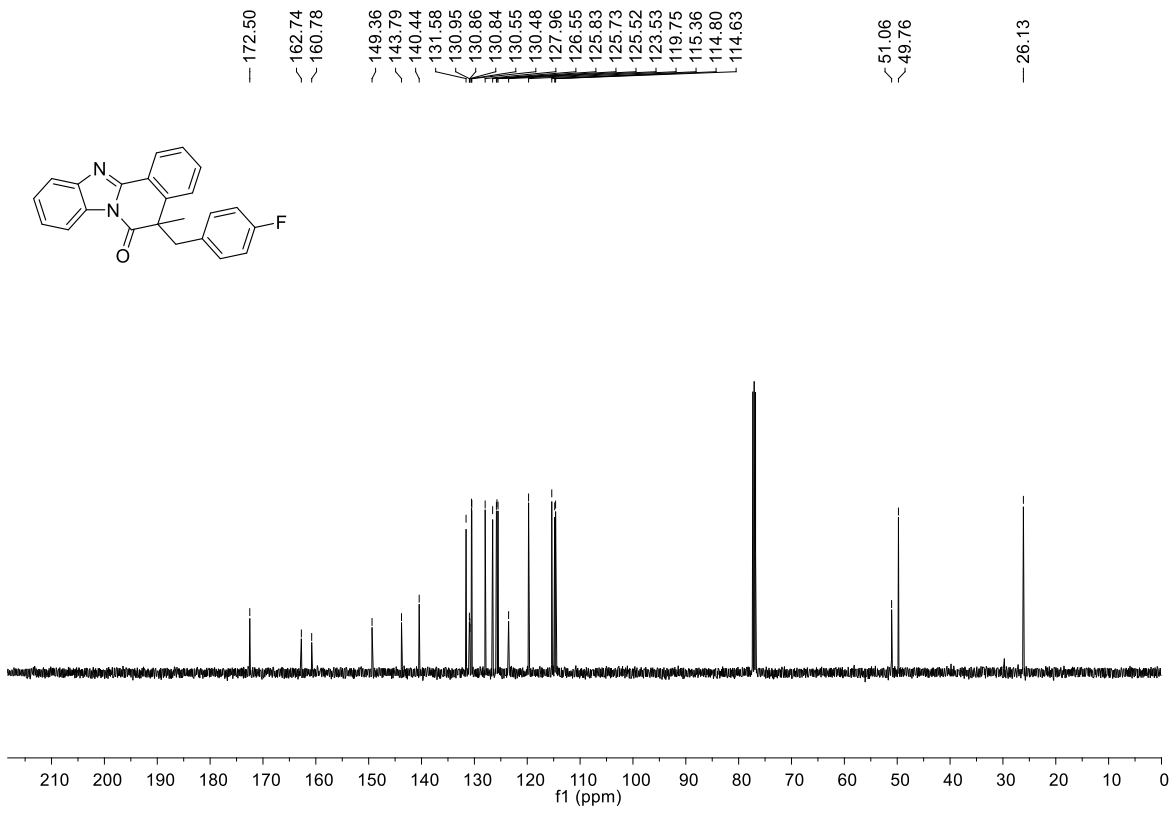
^{13}C{^{1}H} NMR spectrum of 3ad ($CDCl_3$, 126 MHz)



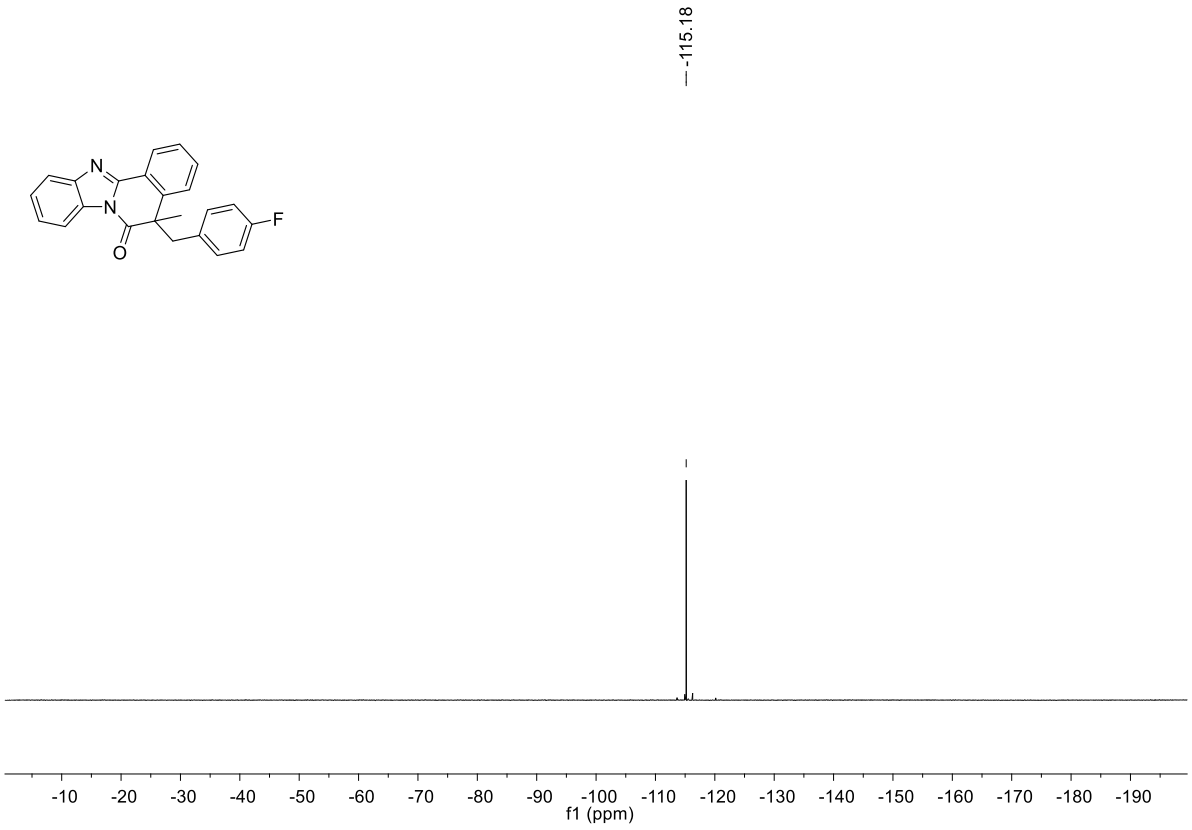
^{1}H NMR spectrum of 3ae ($CDCl_3$, 500 MHz)



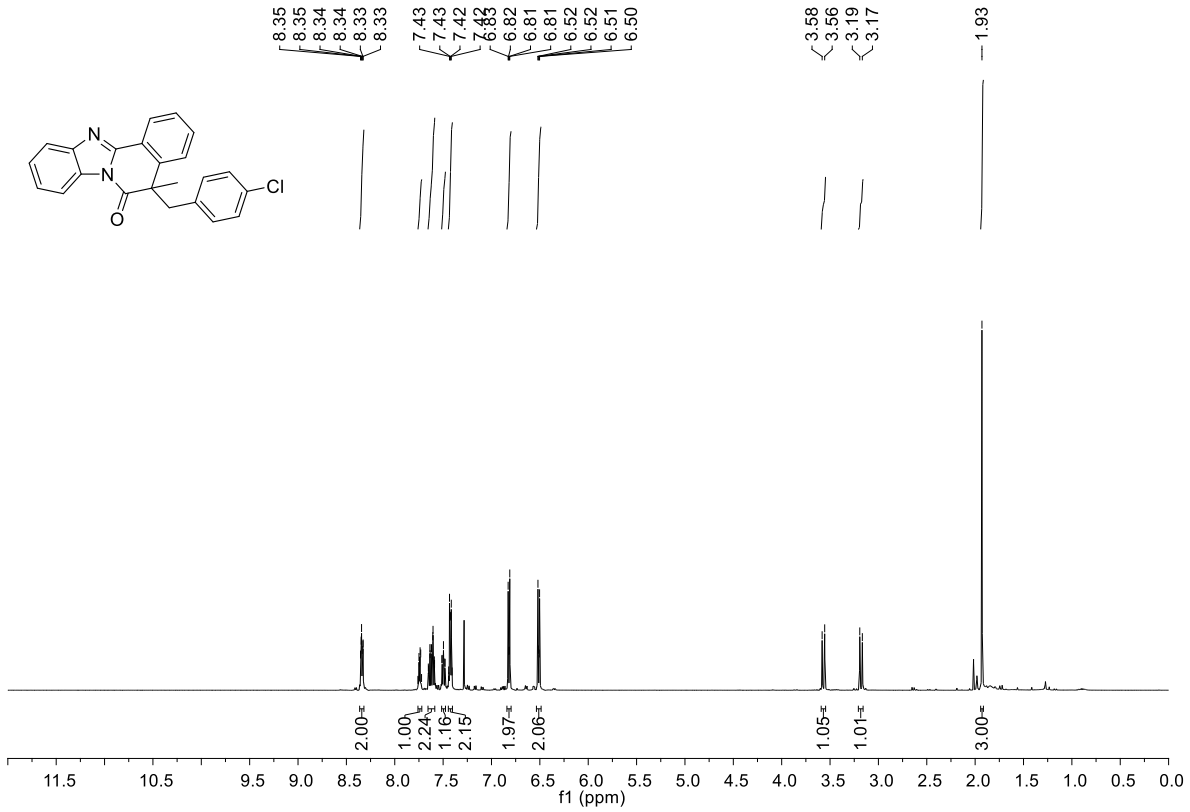
^{13}C{^{1}H} NMR spectrum of 3ae ($CDCl_3$, 126 MHz)



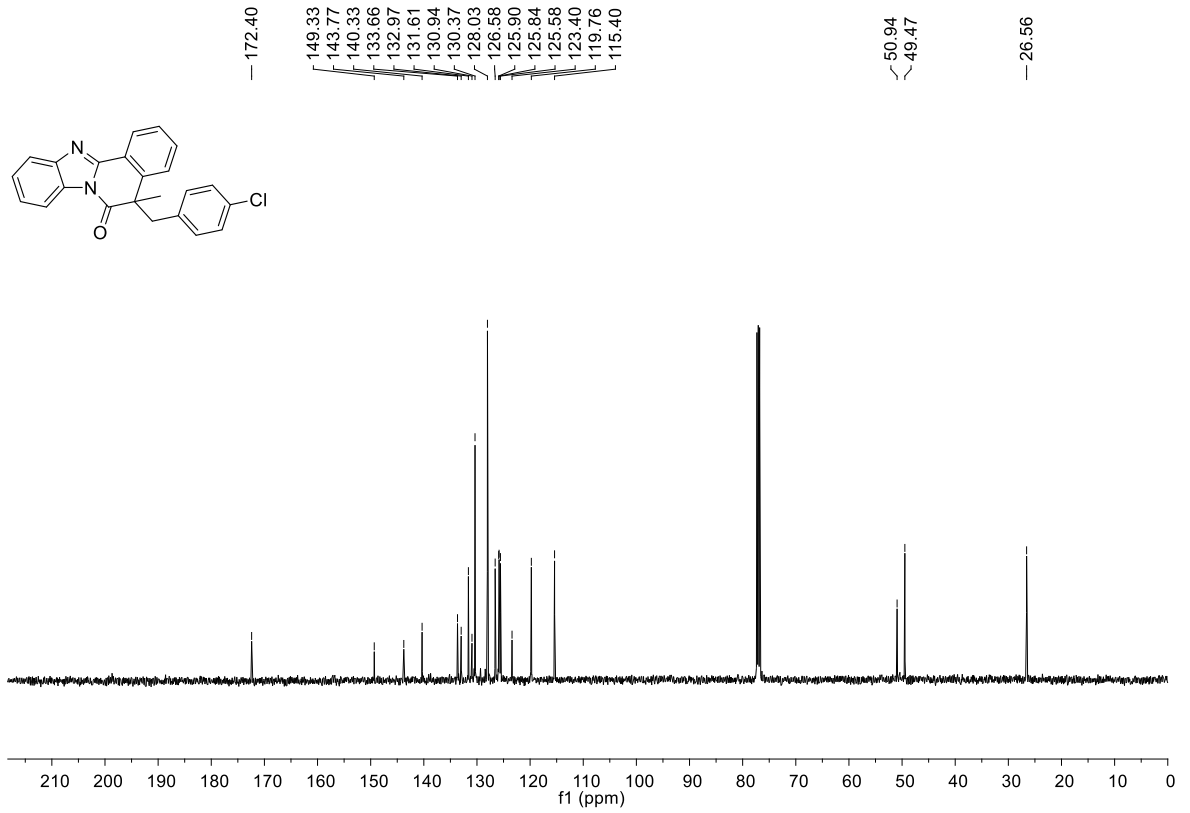
$^{19}F\{^{1}H\}$ NMR spectrum of 3ae ($CDCl_3$, 471 MHz)



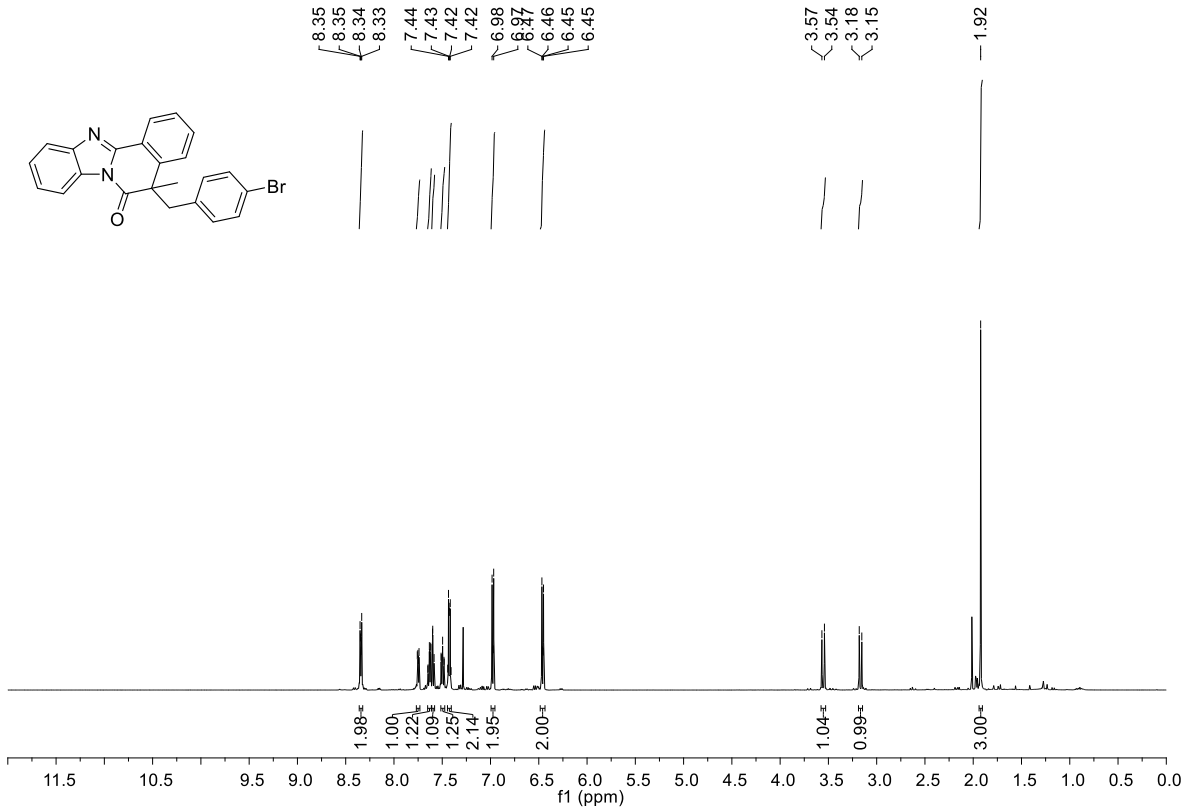
^{1}H NMR spectrum of 3af ($CDCl_3$, 500 MHz)



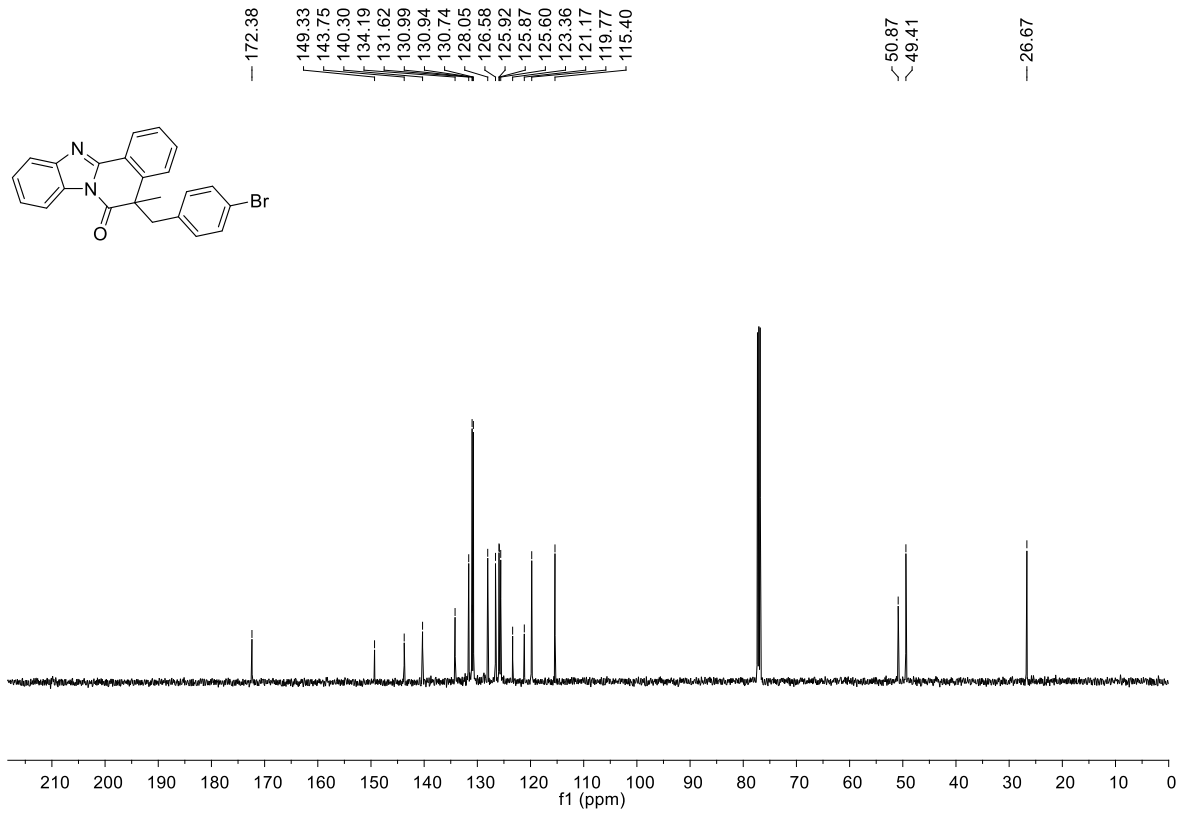
^{13}C{^{1}H} NMR spectrum of 3af ($CDCl_3$, 126 MHz)



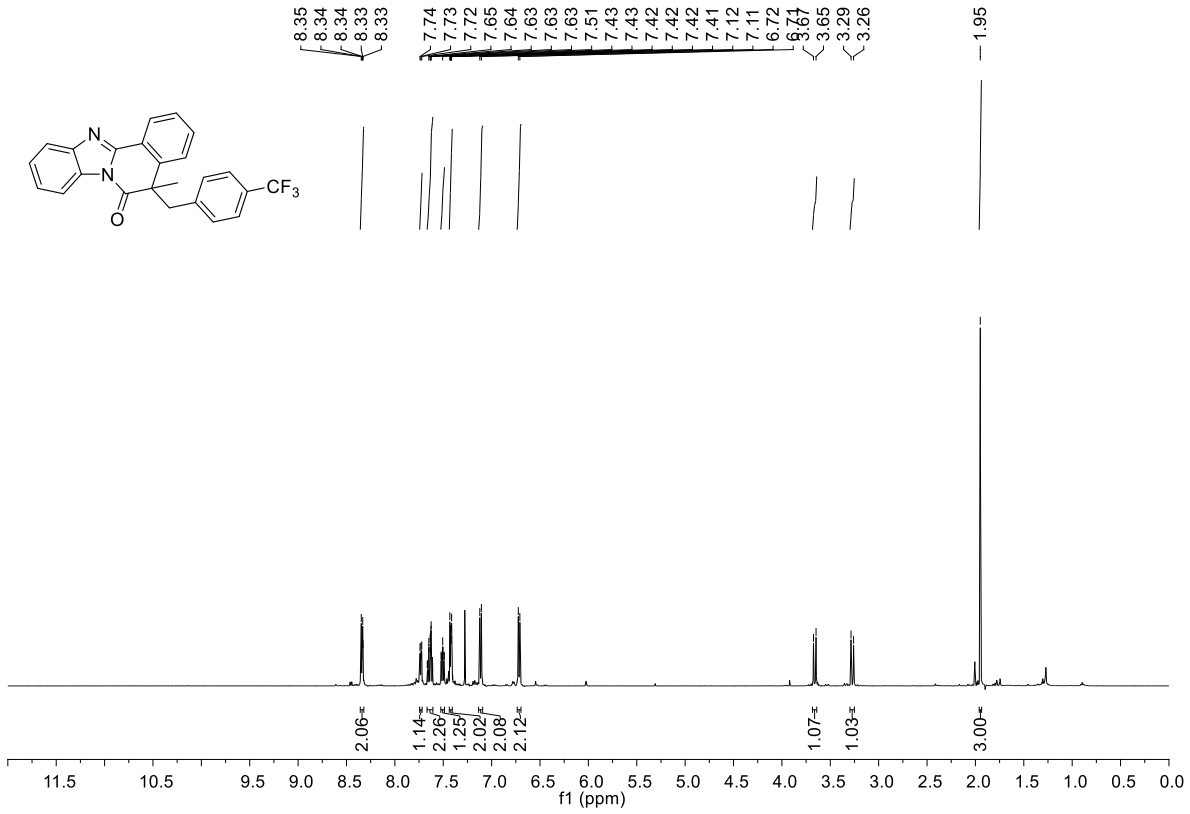
^{1}H NMR spectrum of 3ag ($CDCl_3$, 500 MHz)



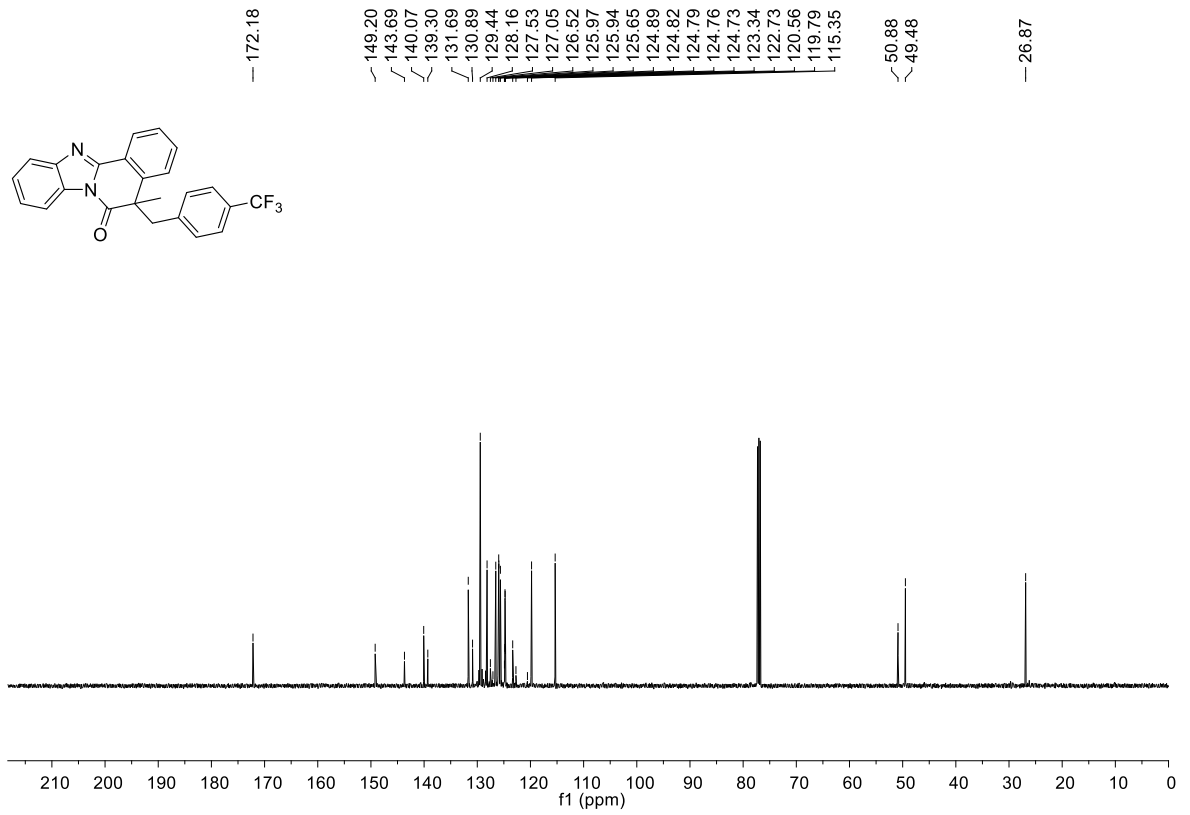
^{13}C{^{1}H} NMR spectrum of 3ag ($CDCl_3$, 126 MHz)



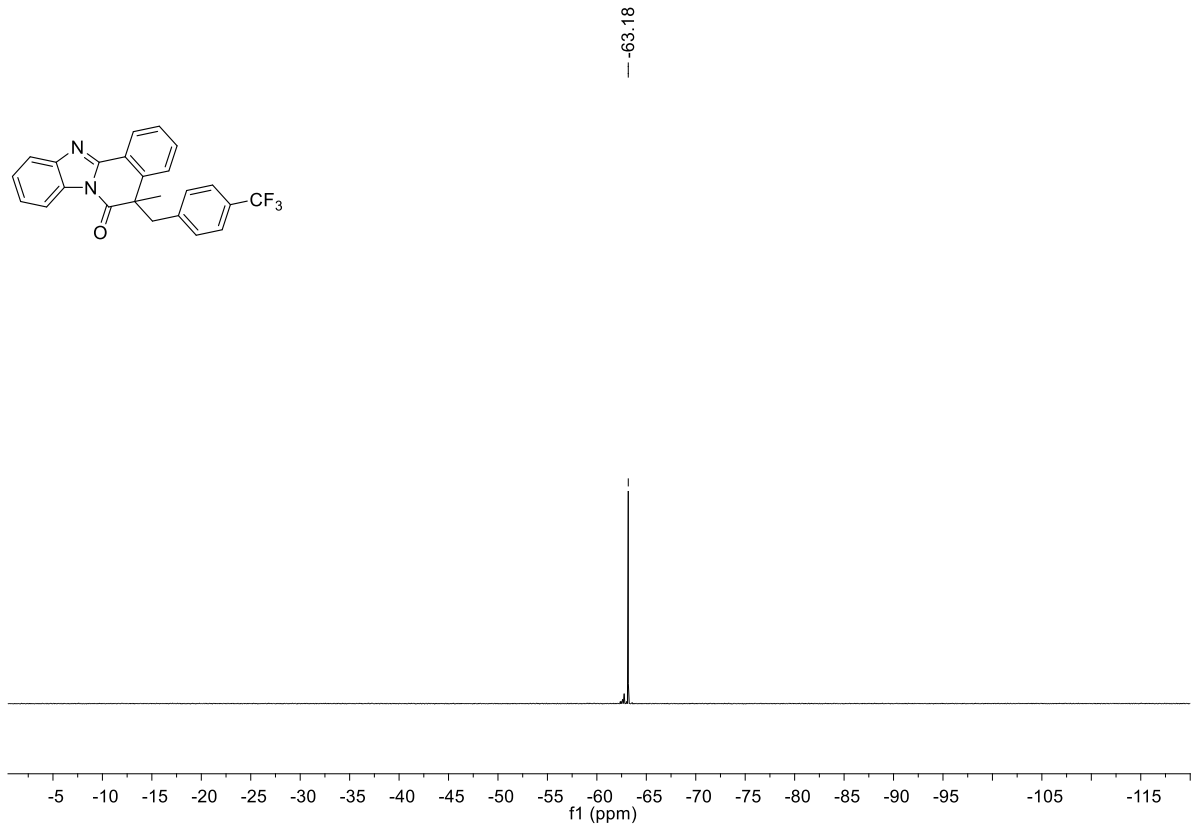
^{1}H NMR spectrum of 3ah ($CDCl_3$, 500 MHz)



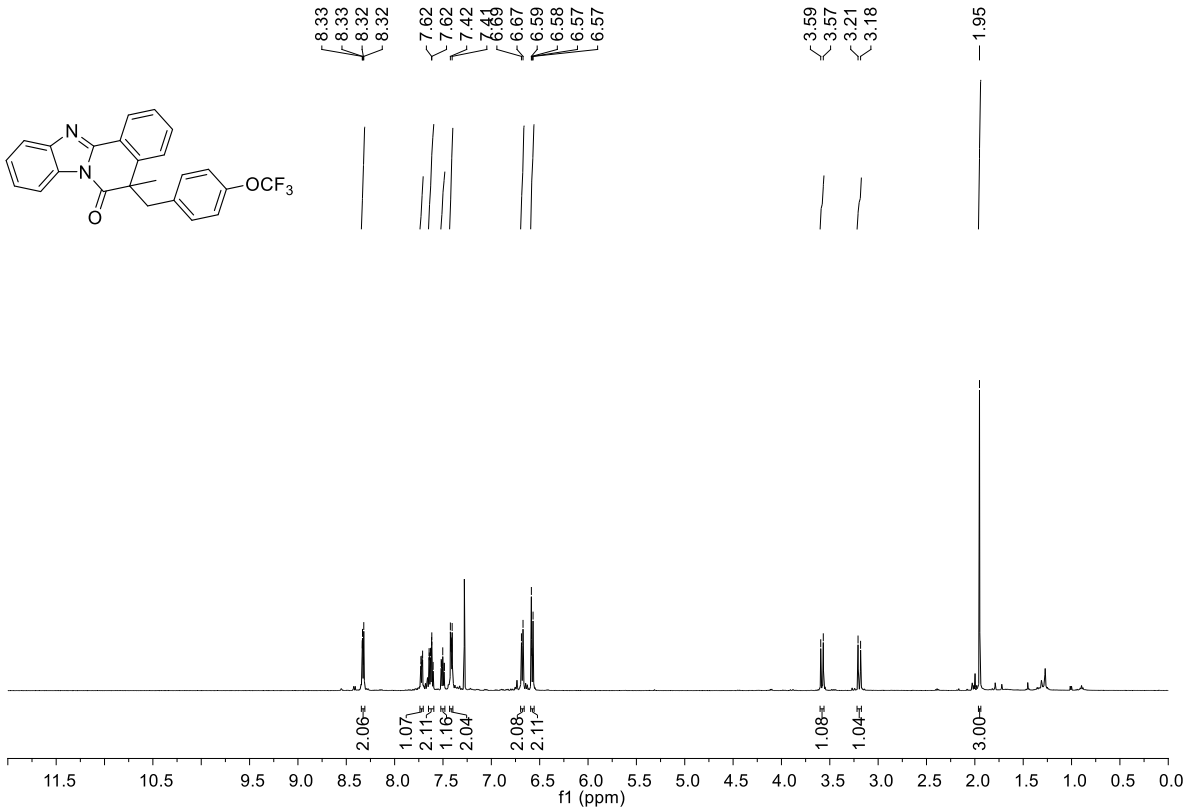
^{13}C{^{1}H} NMR spectrum of 3ah ($CDCl_3$, 126 MHz)



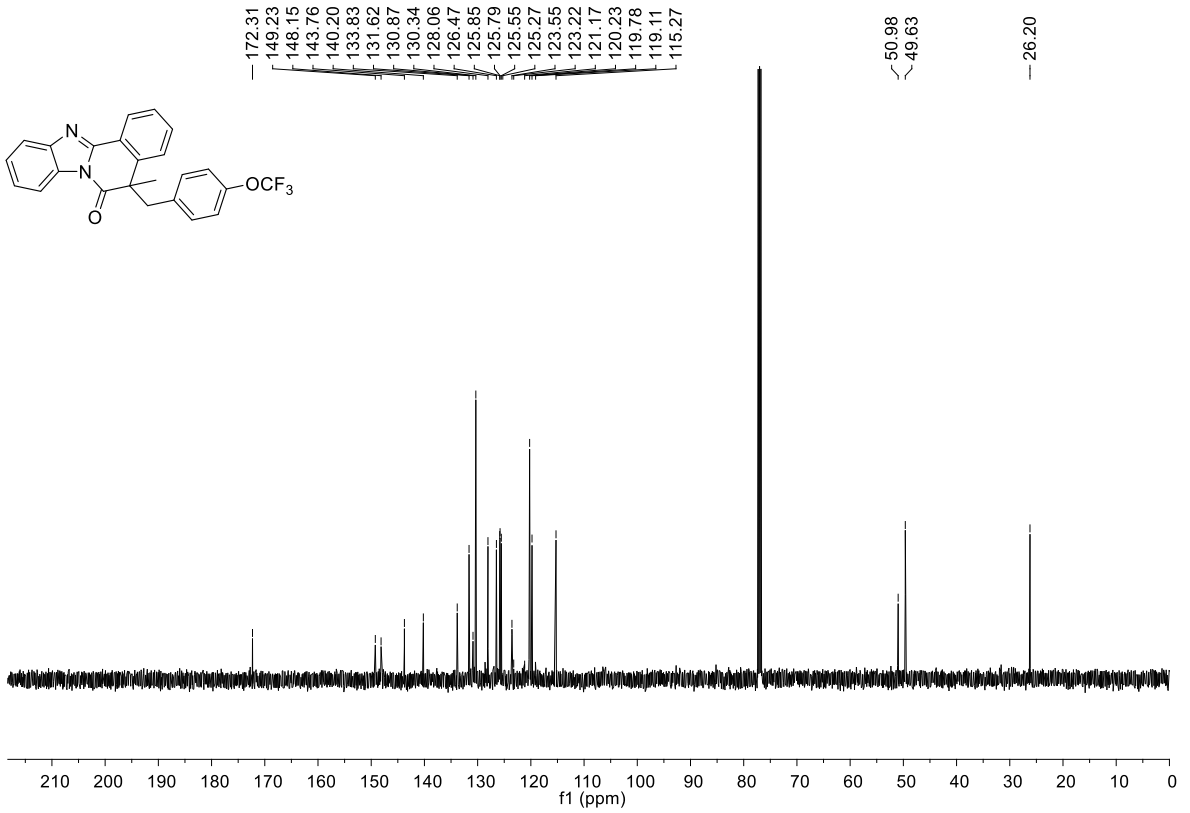
$^{19}F\{^{1}H\}$ NMR spectrum of 3ah ($CDCl_3$, 471 MHz)



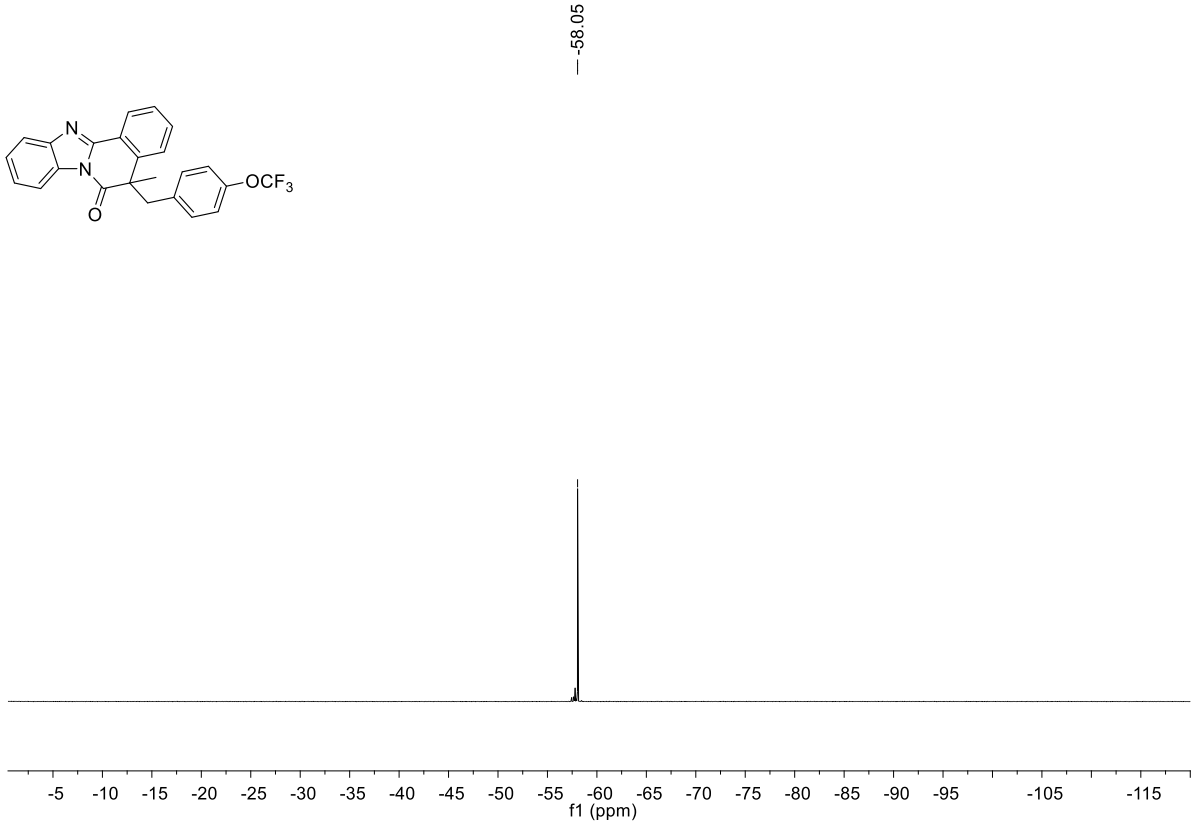
^{1}H NMR spectrum of 3ai ($CDCl_3$, 500 MHz)



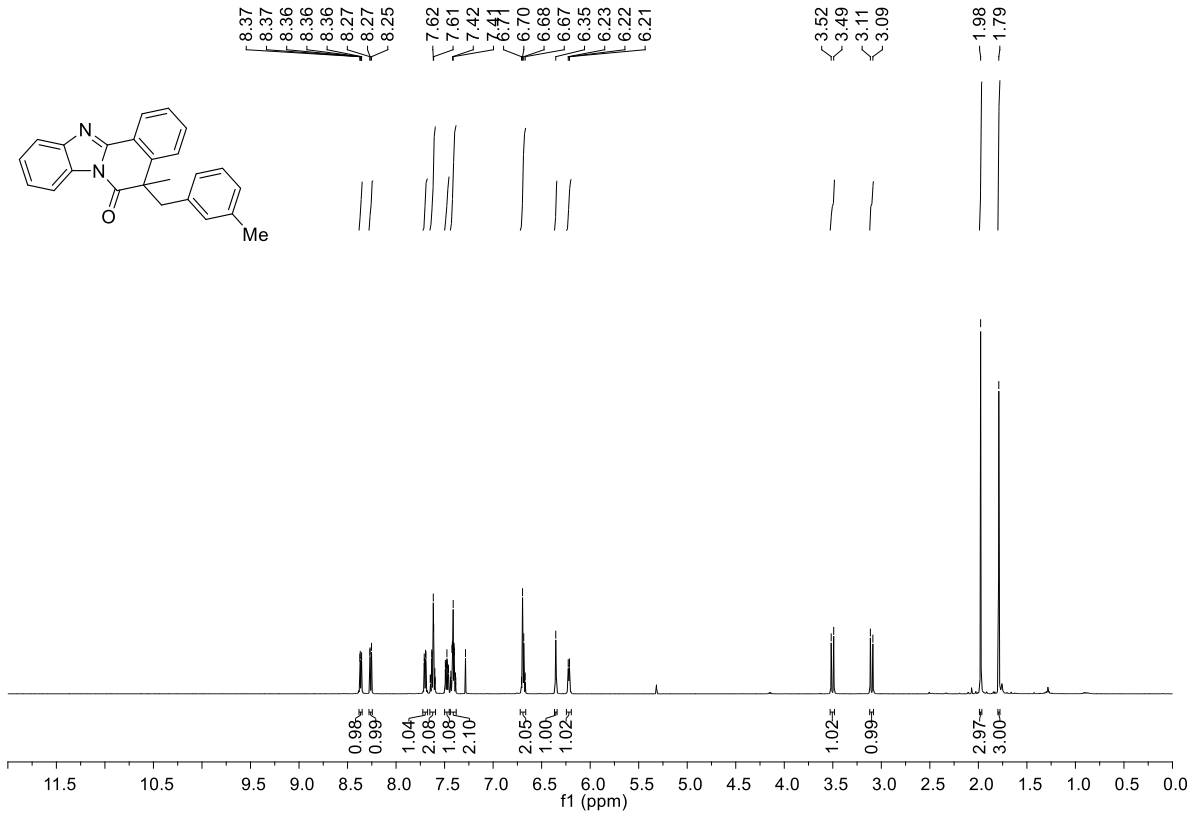
^{13}C{^{1}H} NMR spectrum of 3ai ($CDCl_3$, 126 MHz)



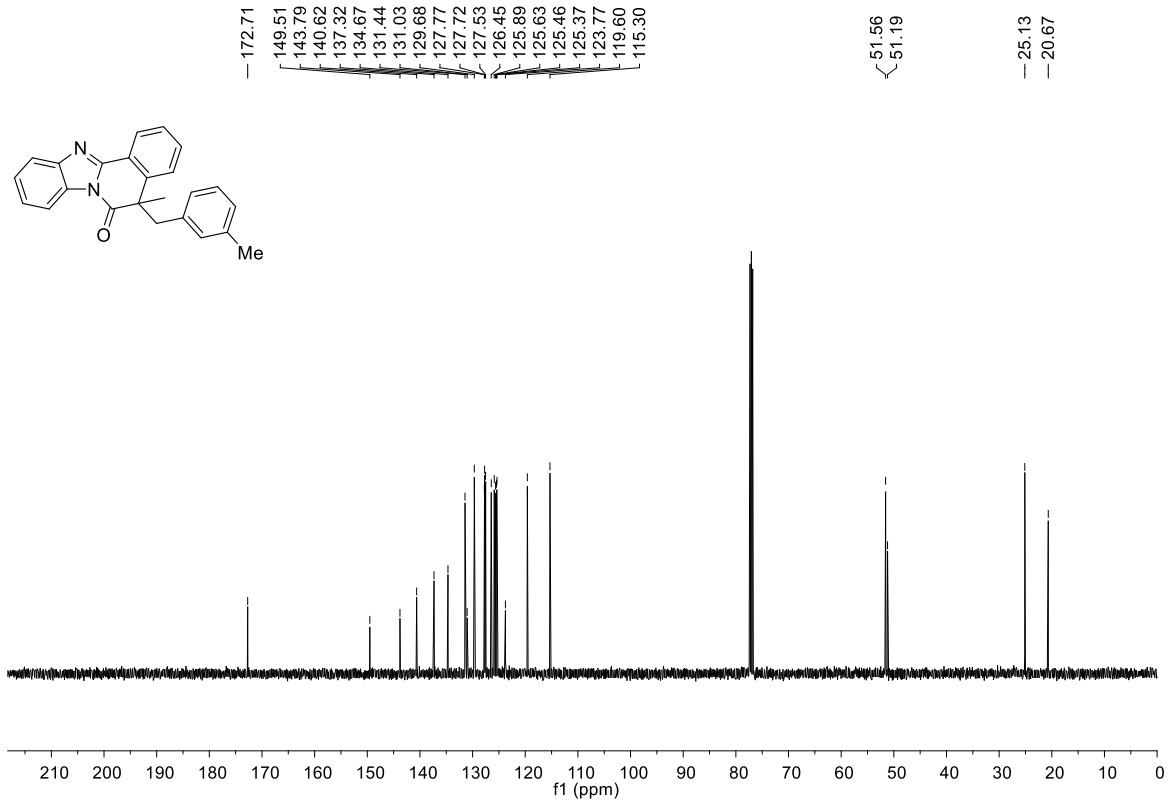
$^{19}F\{^{1}H\}$ NMR spectrum of 3ai ($CDCl_3$, 471 MHz)



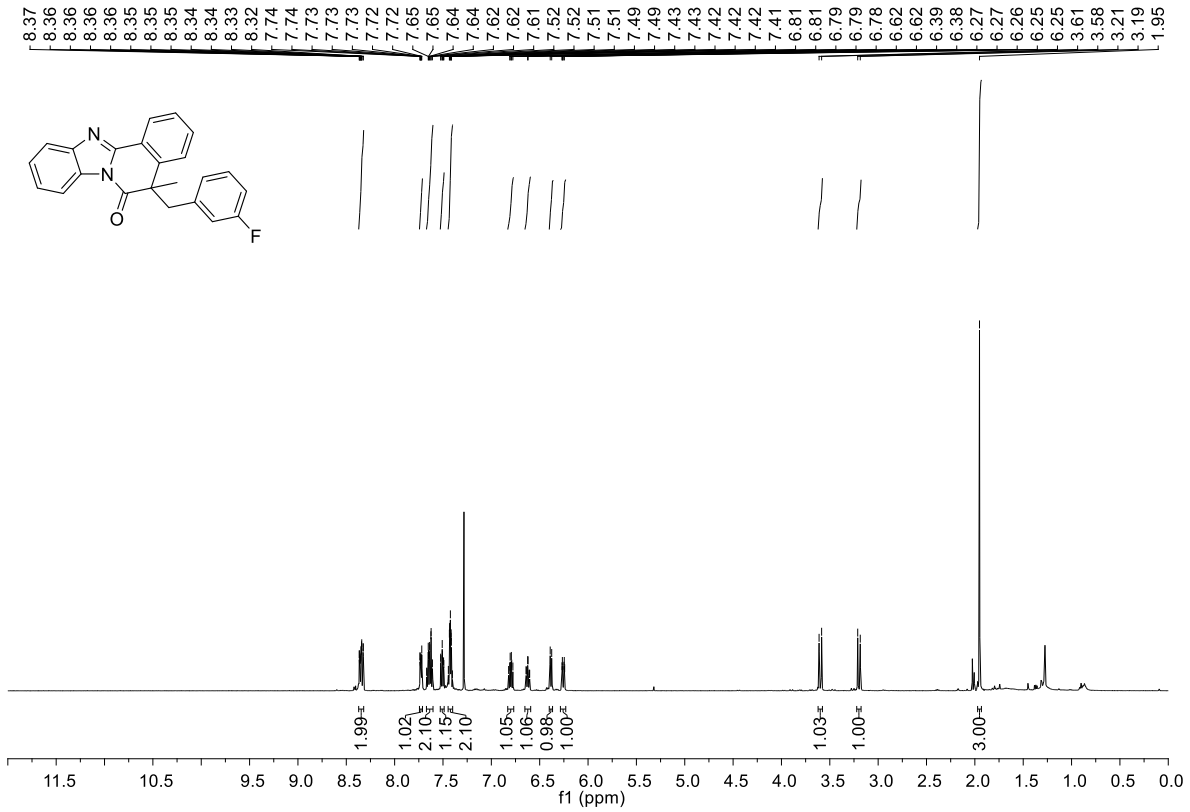
^{1}H NMR spectrum of 3aj ($CDCl_3$, 500 MHz)



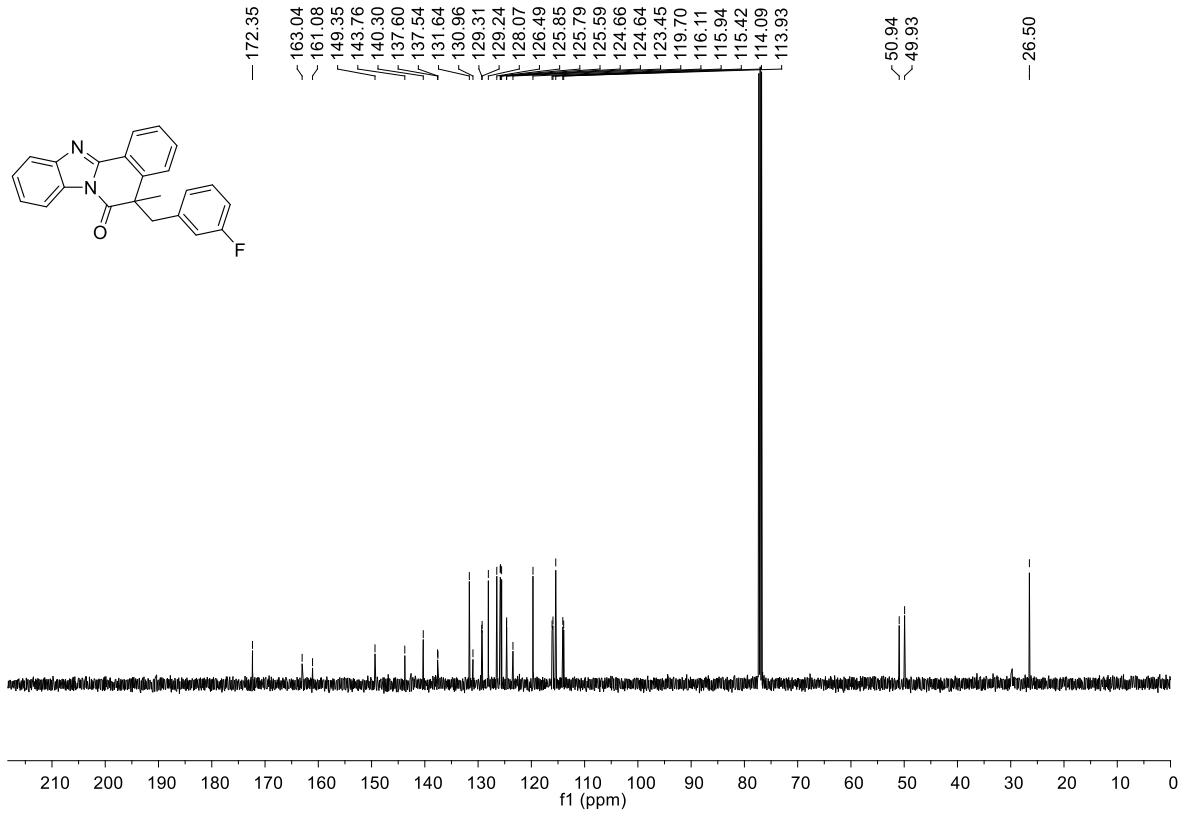
^{13}C{^{1}H} NMR spectrum of 3aj ($CDCl_3$, 126 MHz)



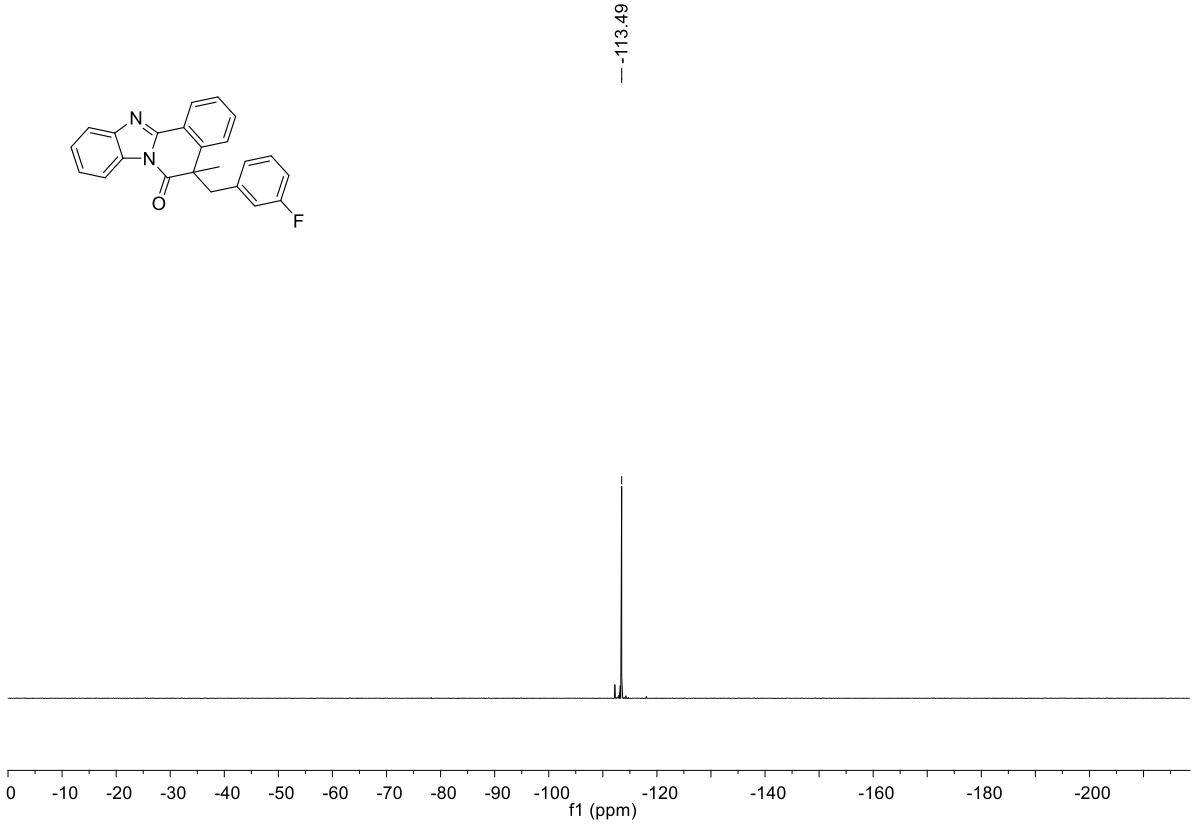
^{1}H NMR spectrum of 3ak ($CDCl_3$, 500 MHz)



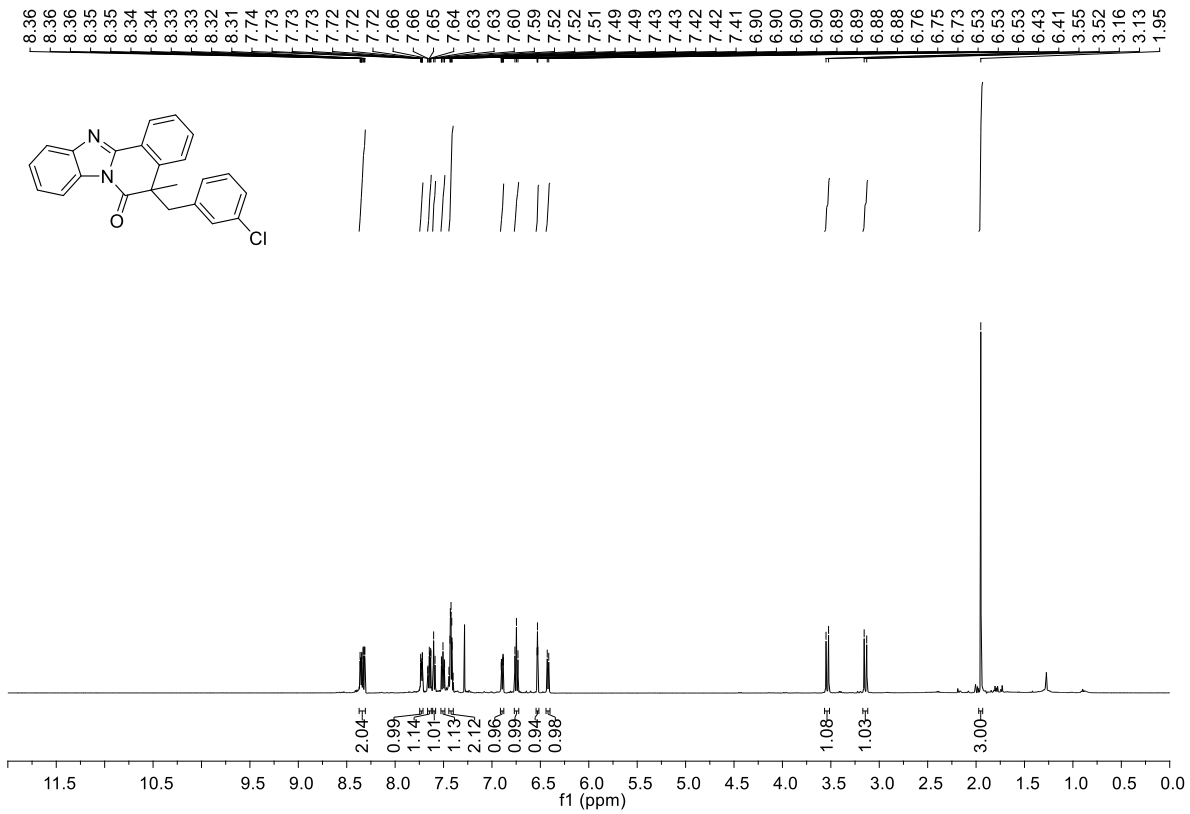
^{13}C{^{1}H} NMR spectrum of 3ak ($CDCl_3$, 126 MHz)



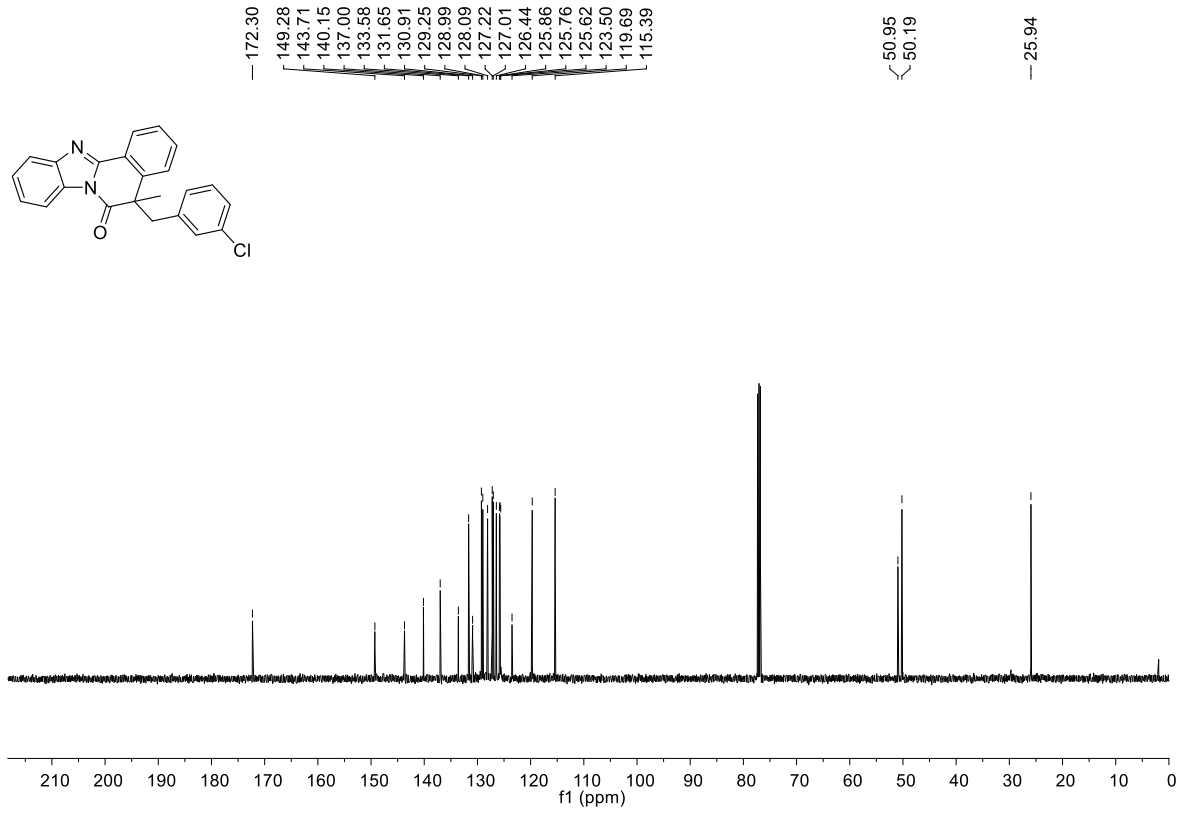
$^{19}F\{^{1}H\}$ NMR spectrum of 3ak ($CDCl_3$, 377 MHz)



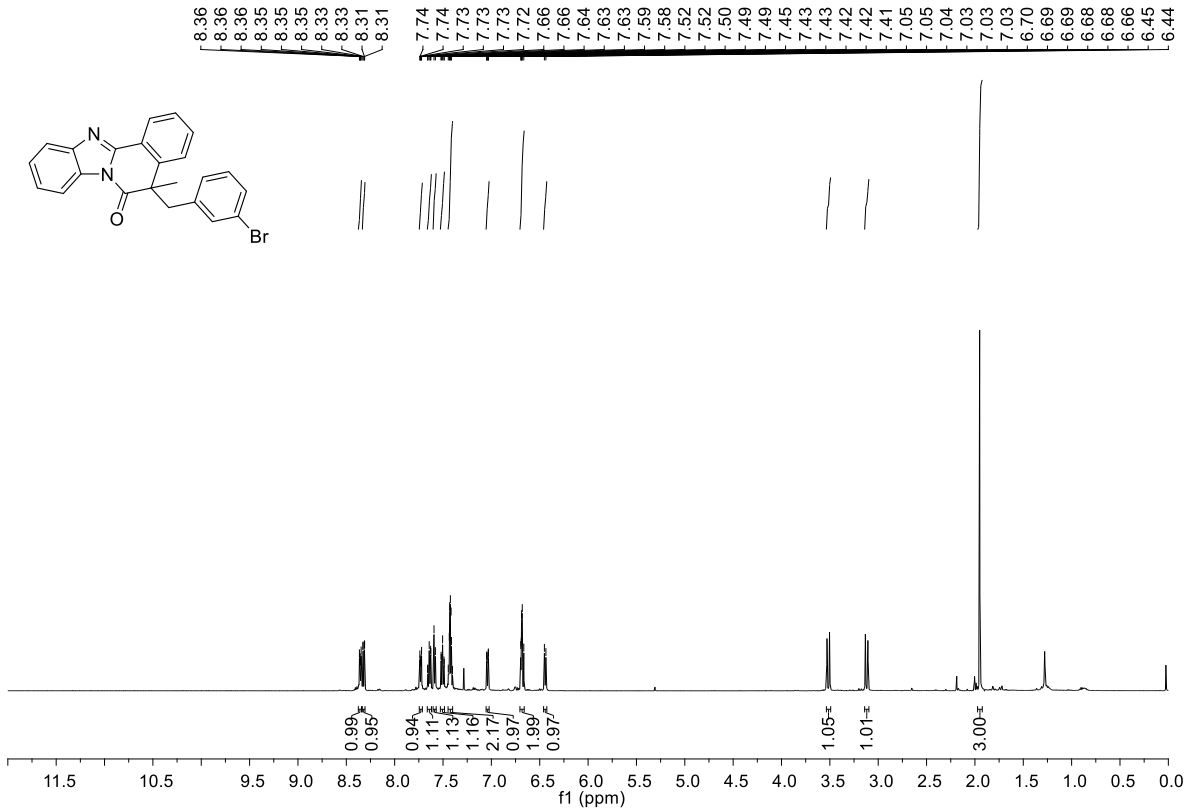
^{1}H NMR spectrum of 3al ($CDCl_3$, 500 MHz)



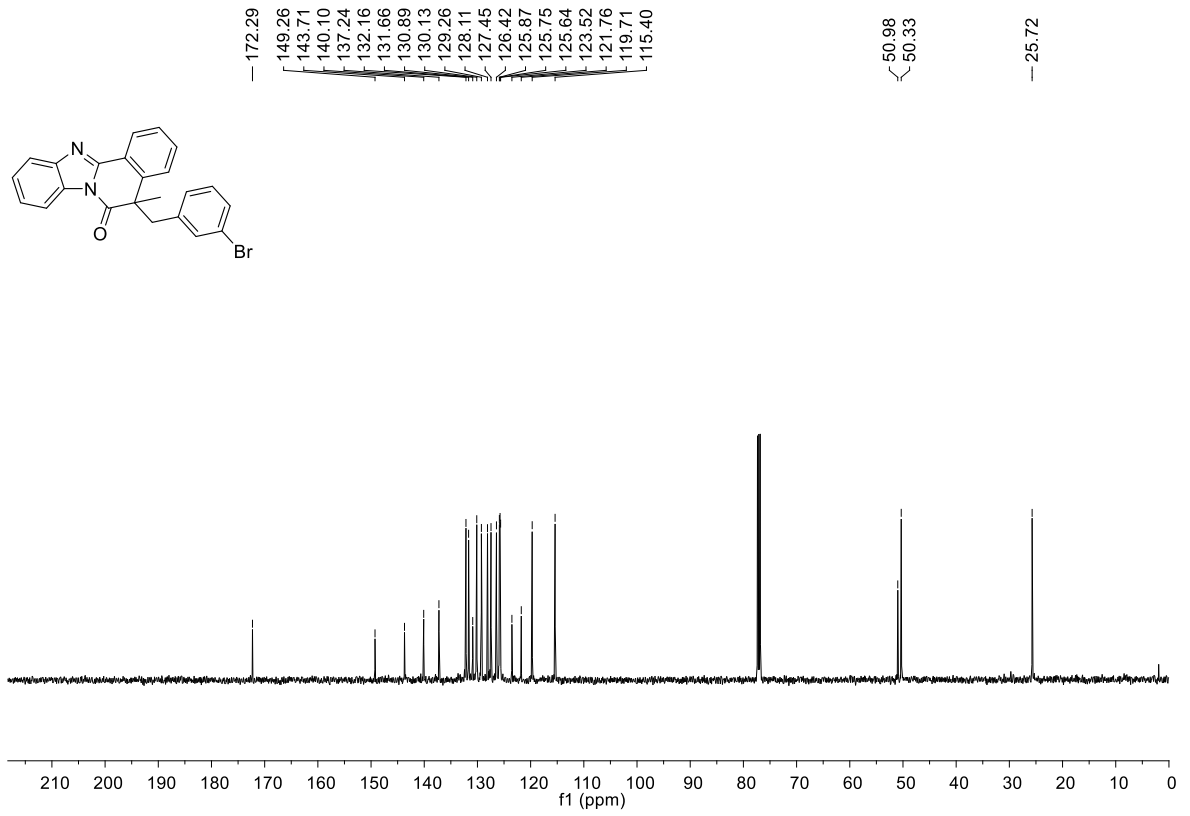
^{13}C{^{1}H} NMR spectrum of 3al ($CDCl_3$, 126 MHz)



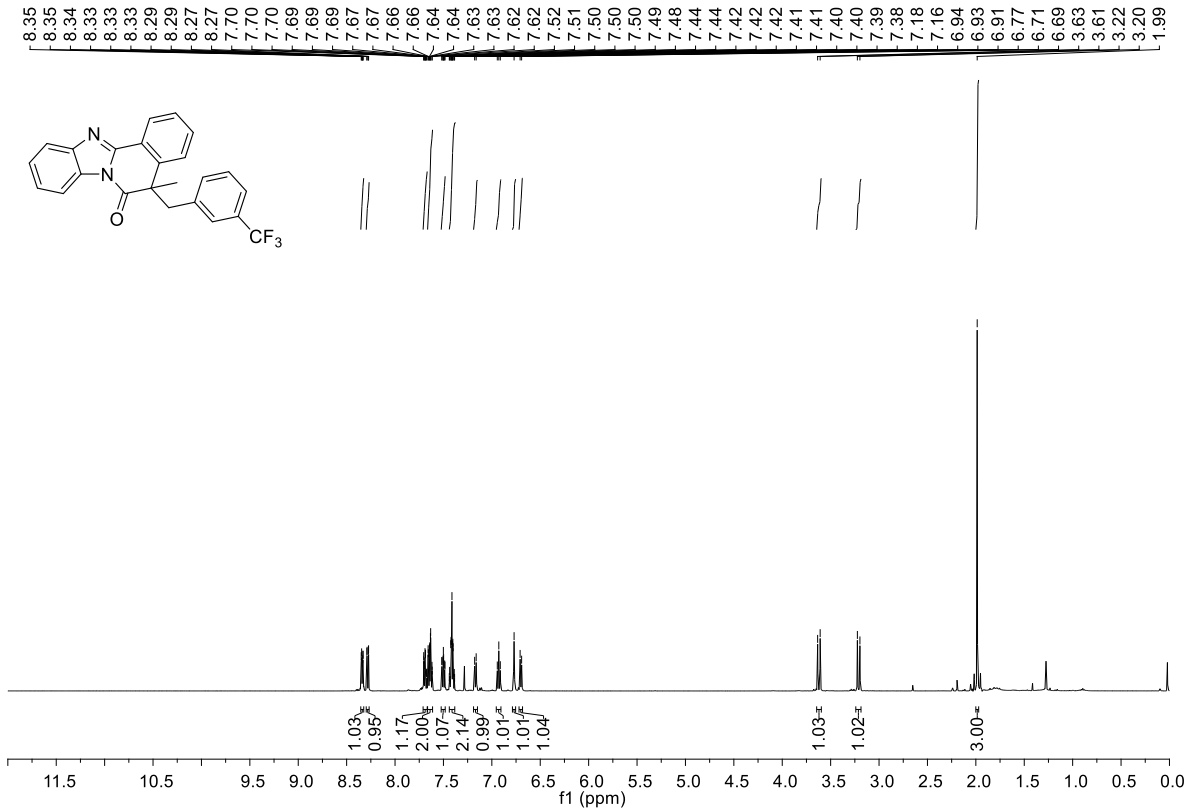
^{1}H NMR spectrum of 3am ($CDCl_3$, 500 MHz)



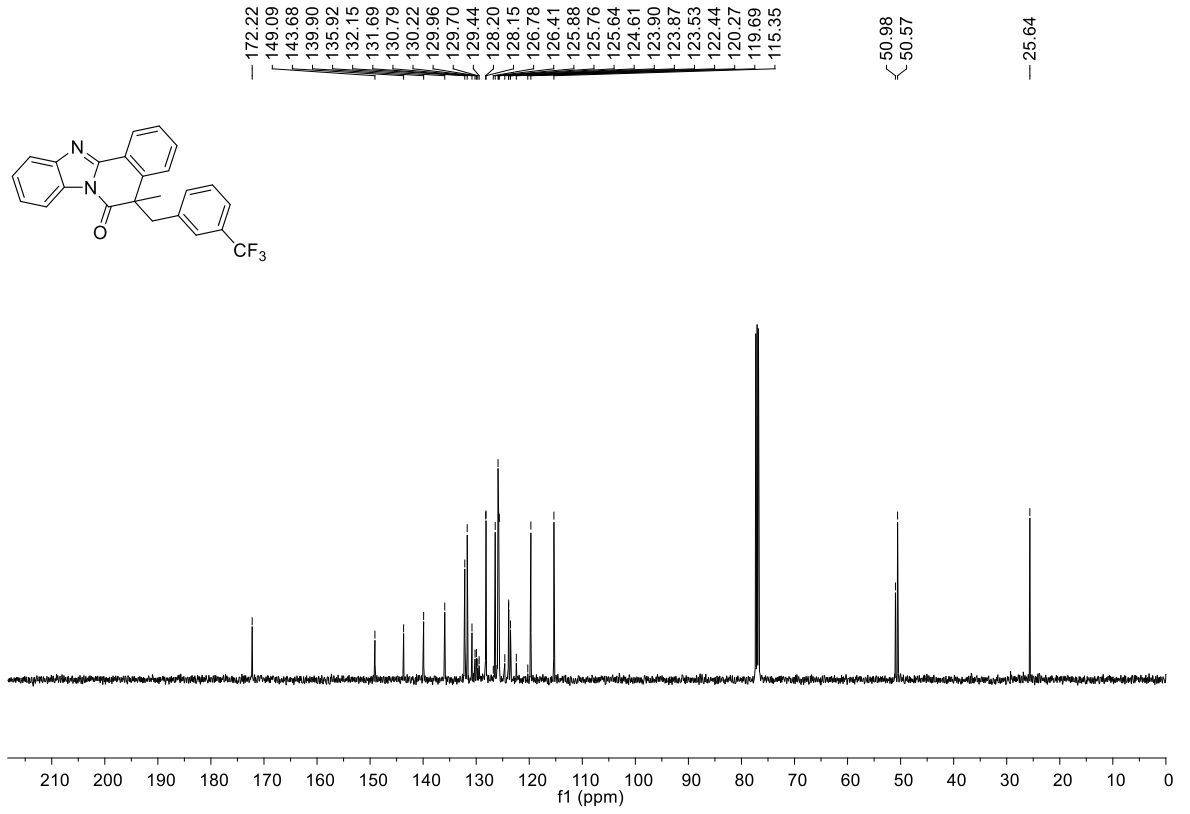
^{13}C{^{1}H} NMR spectrum of 3am ($CDCl_3$, 126 MHz)



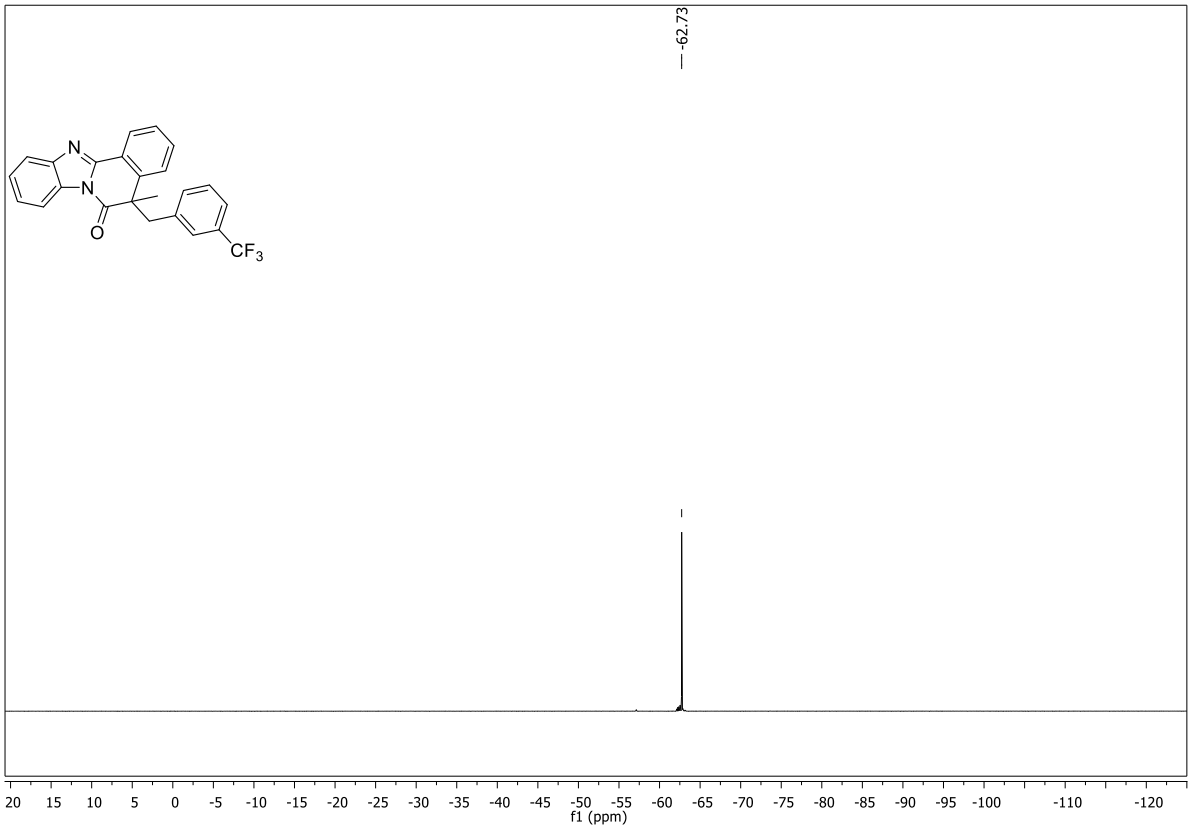
^{1}H NMR spectrum of 3an ($CDCl_3$, 500 MHz)



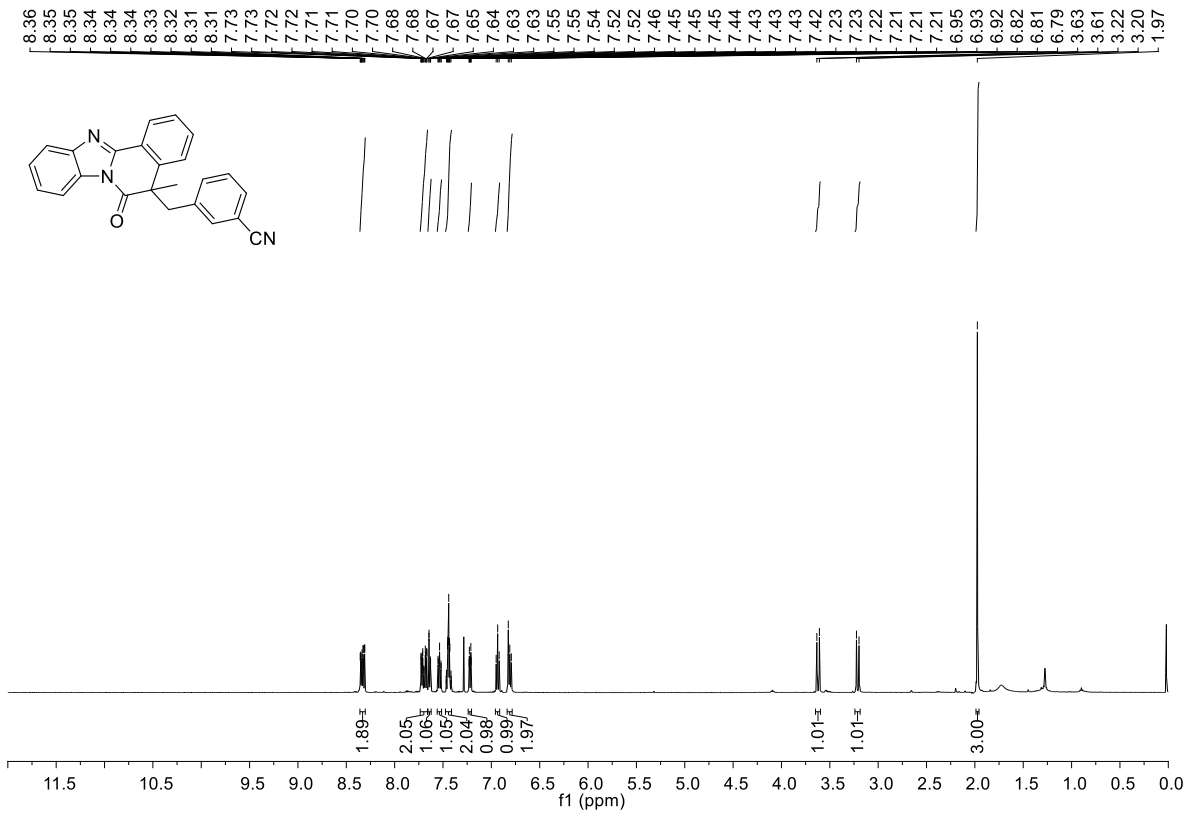
^{13}C{^{1}H} NMR spectrum of 3an ($CDCl_3$, 126 MHz)



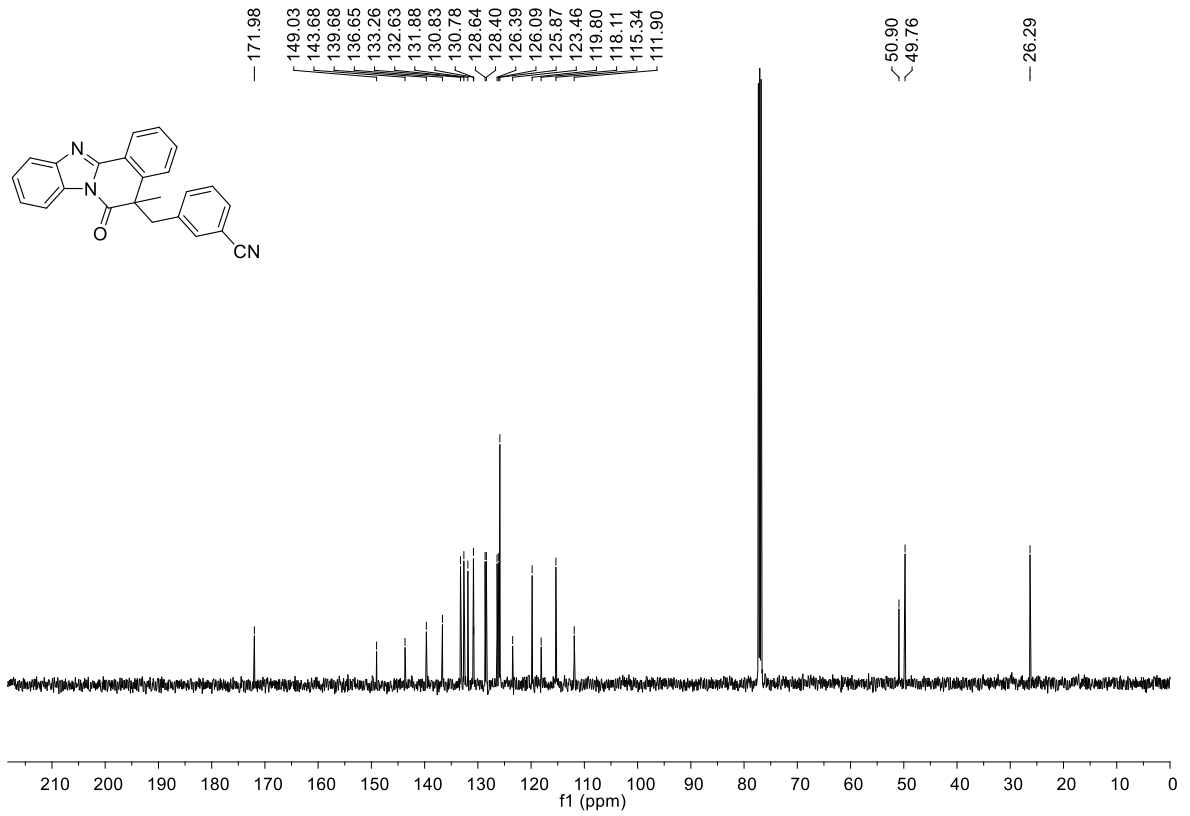
$^{19}F\{^{1}H\}$ NMR spectrum of 3an ($CDCl_3$, 471 MHz)



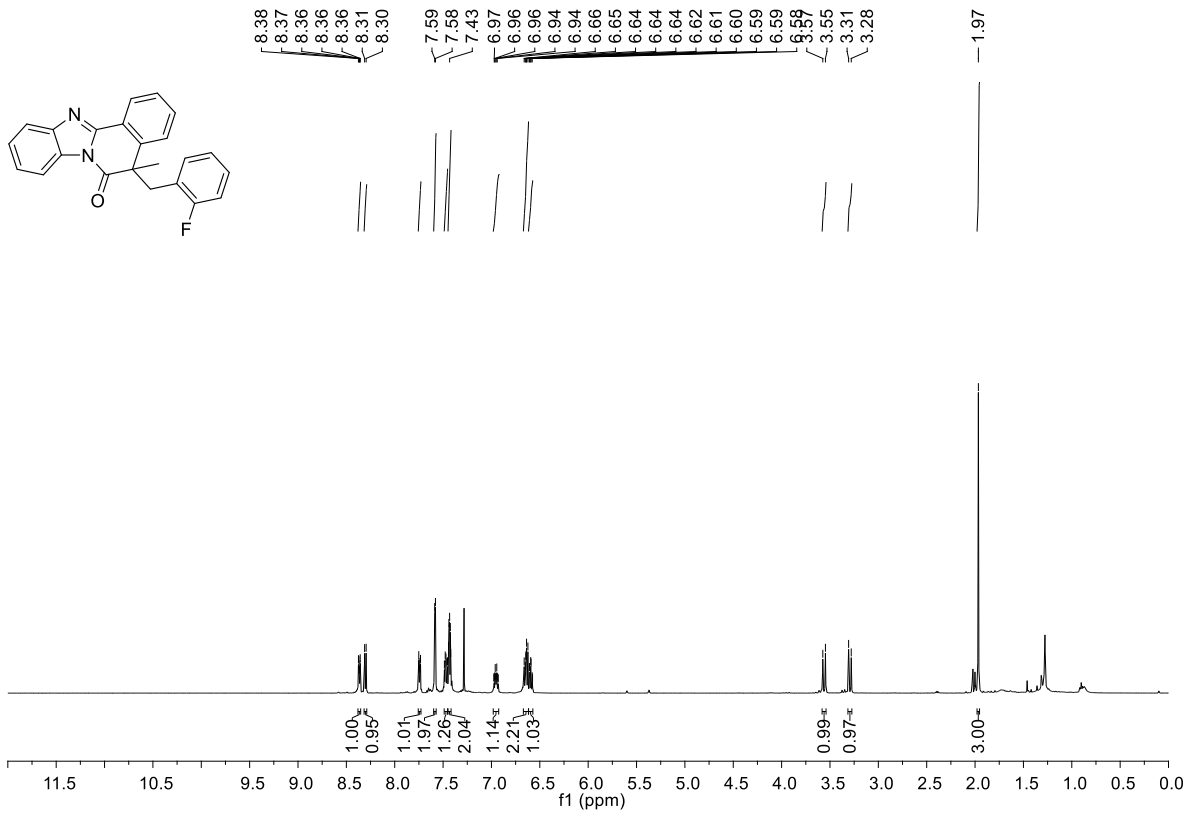
^{1}H NMR spectrum of 3ao ($CDCl_3$, 500 MHz)



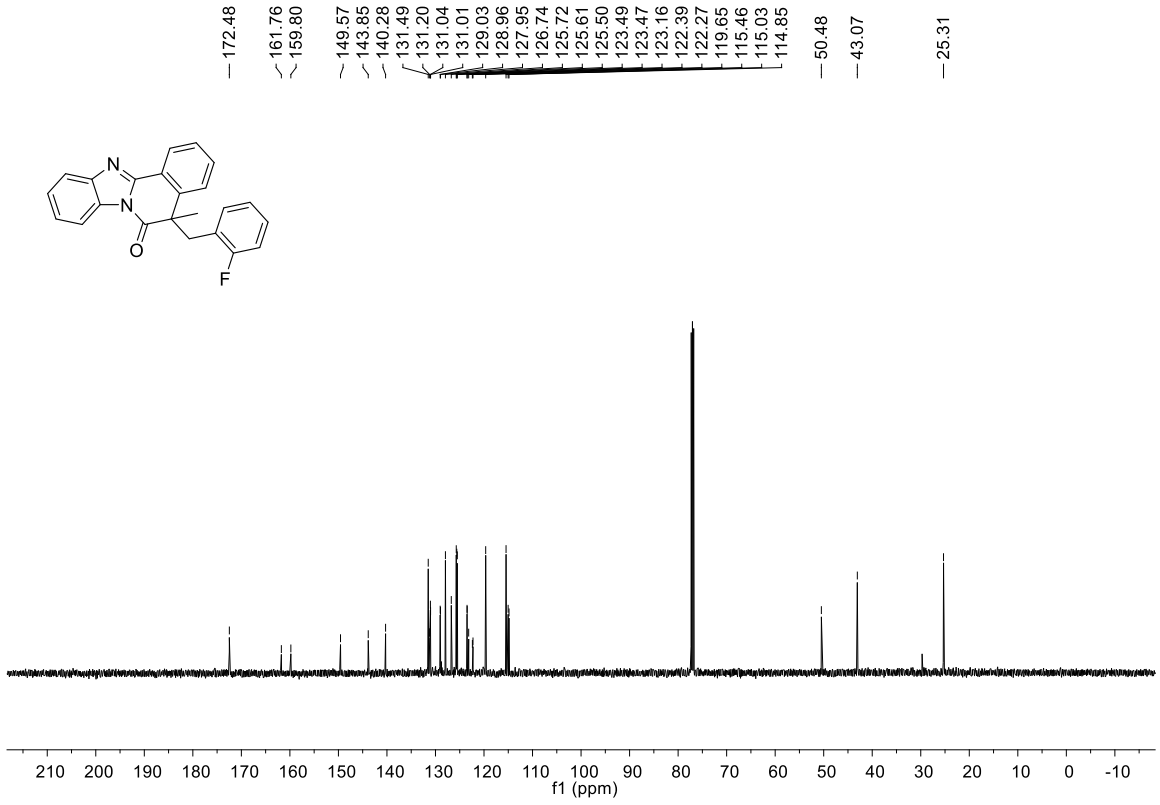
^{13}C{^{1}H} NMR spectrum of 3ao ($CDCl_3$, 126 MHz)



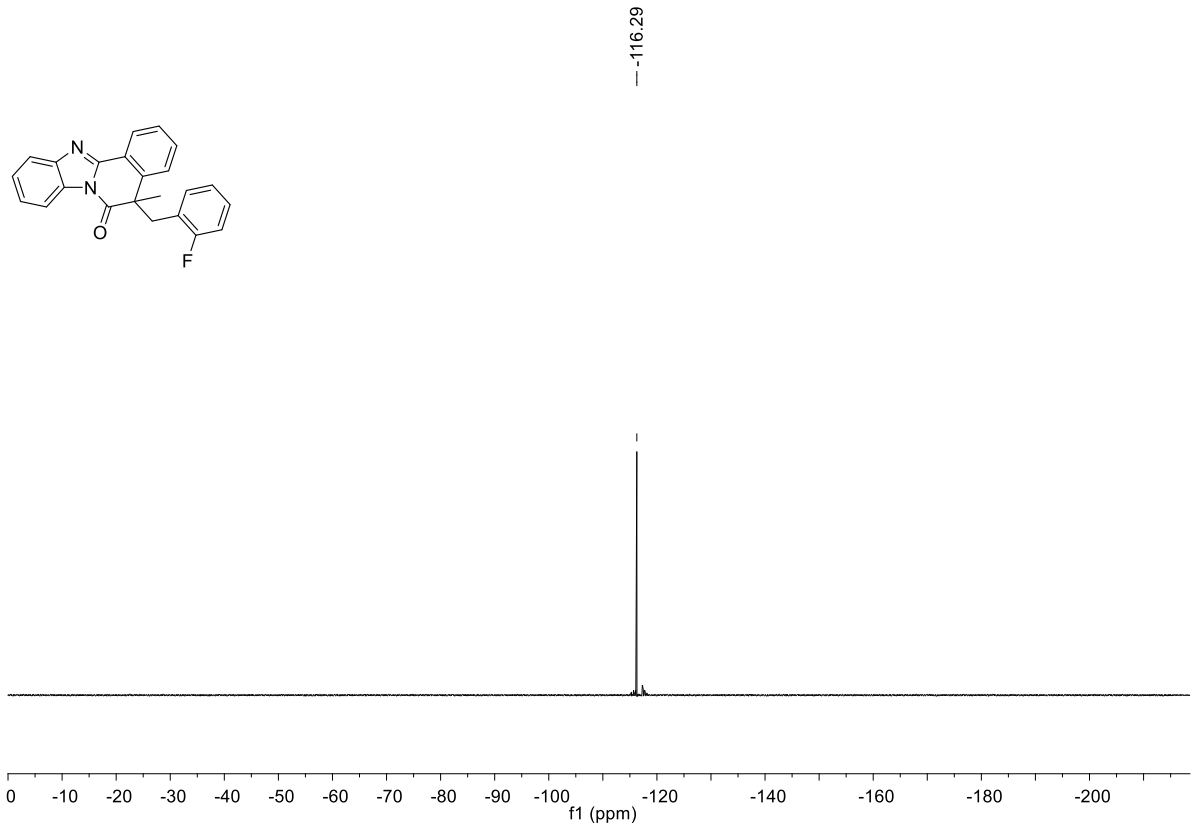
^{1}H NMR spectrum of 3ap ($CDCl_3$, 500 MHz)



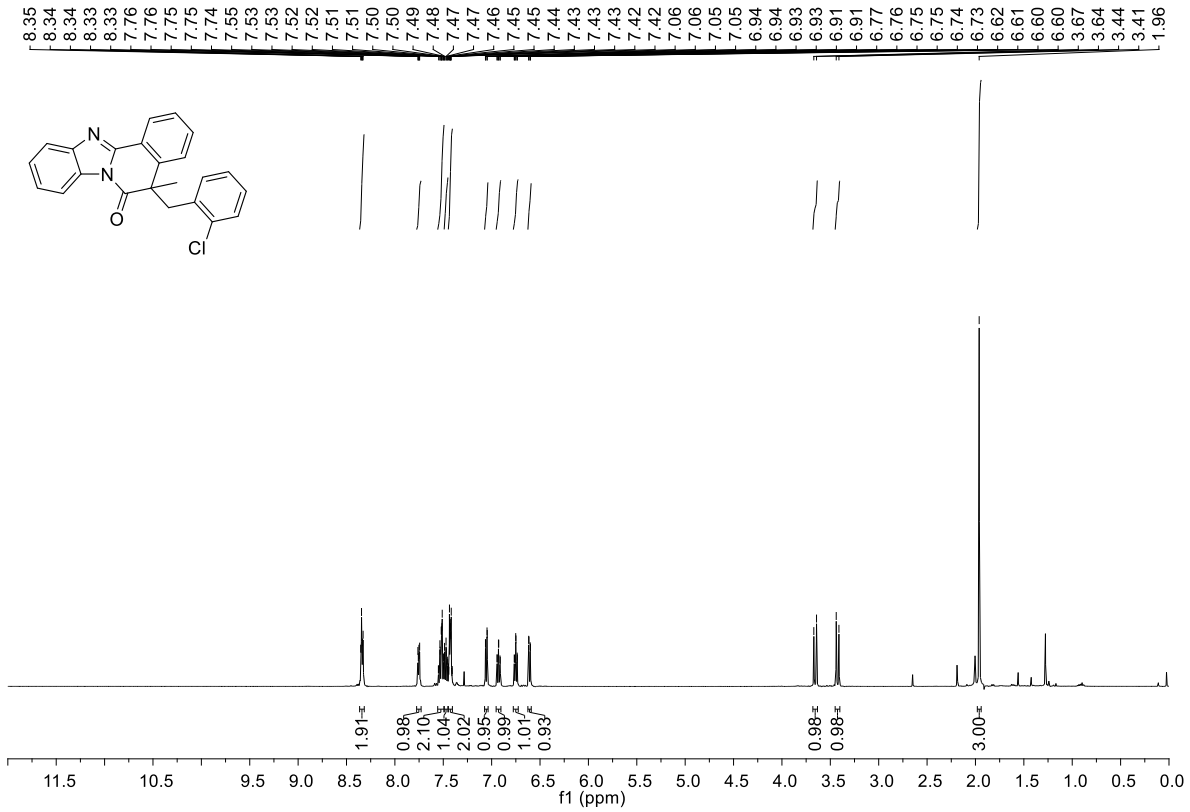
^{13}C{^{1}H} NMR spectrum of 3ap ($CDCl_3$, 126 MHz)



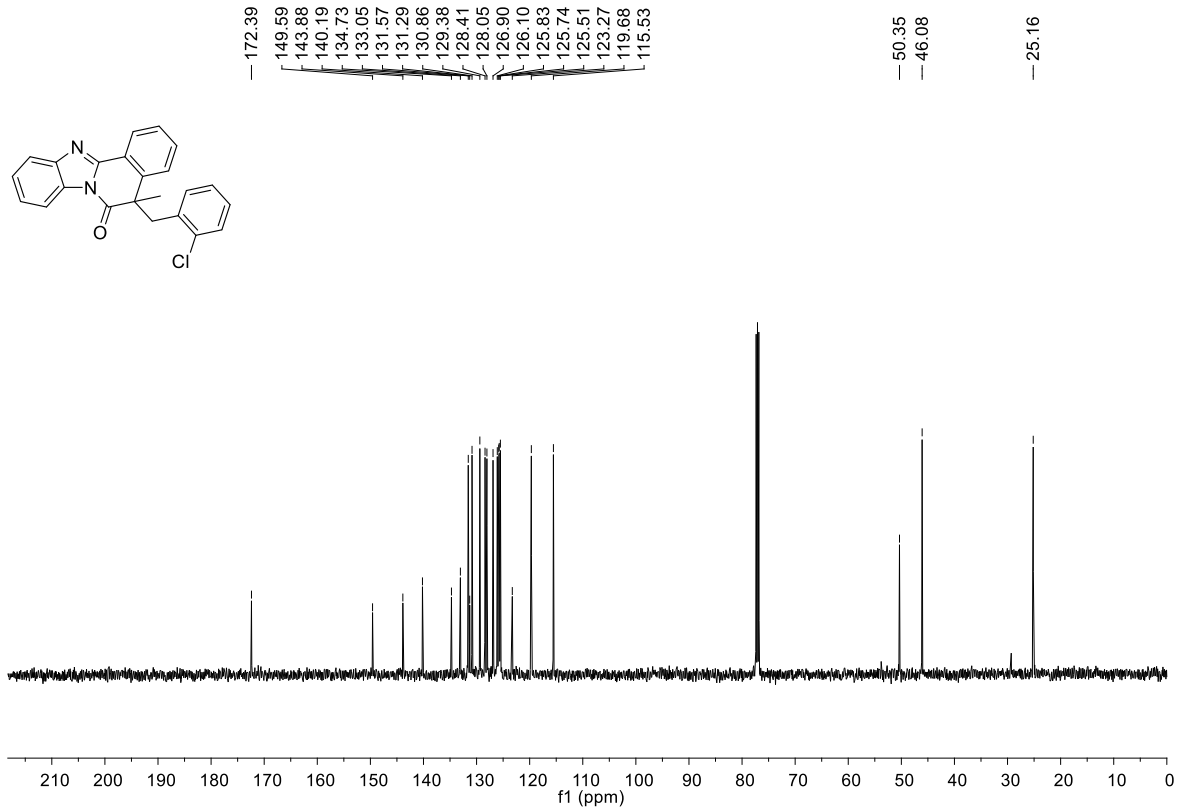
$^{19}F\{^{1}H\}$ NMR spectrum of 3ap ($CDCl_3$, 377 MHz)



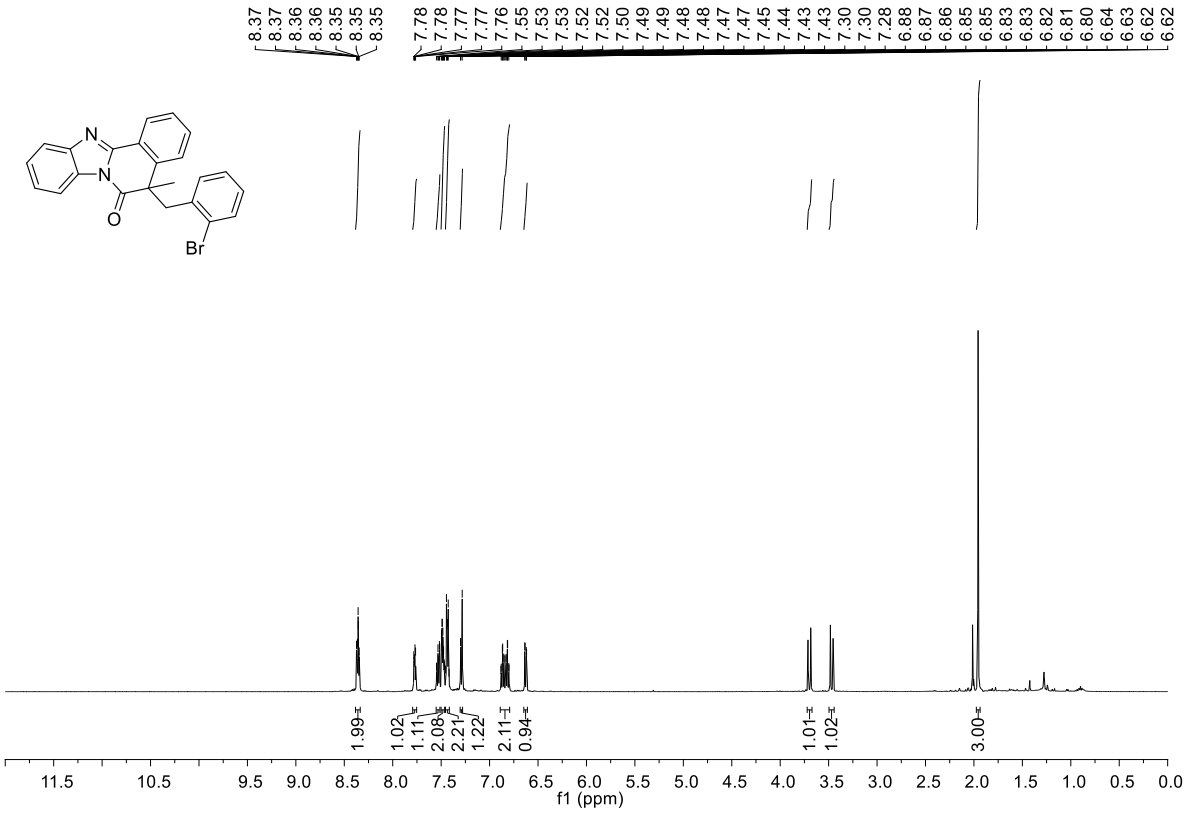
^{1}H NMR spectrum of 3aq ($CDCl_3$, 500 MHz)



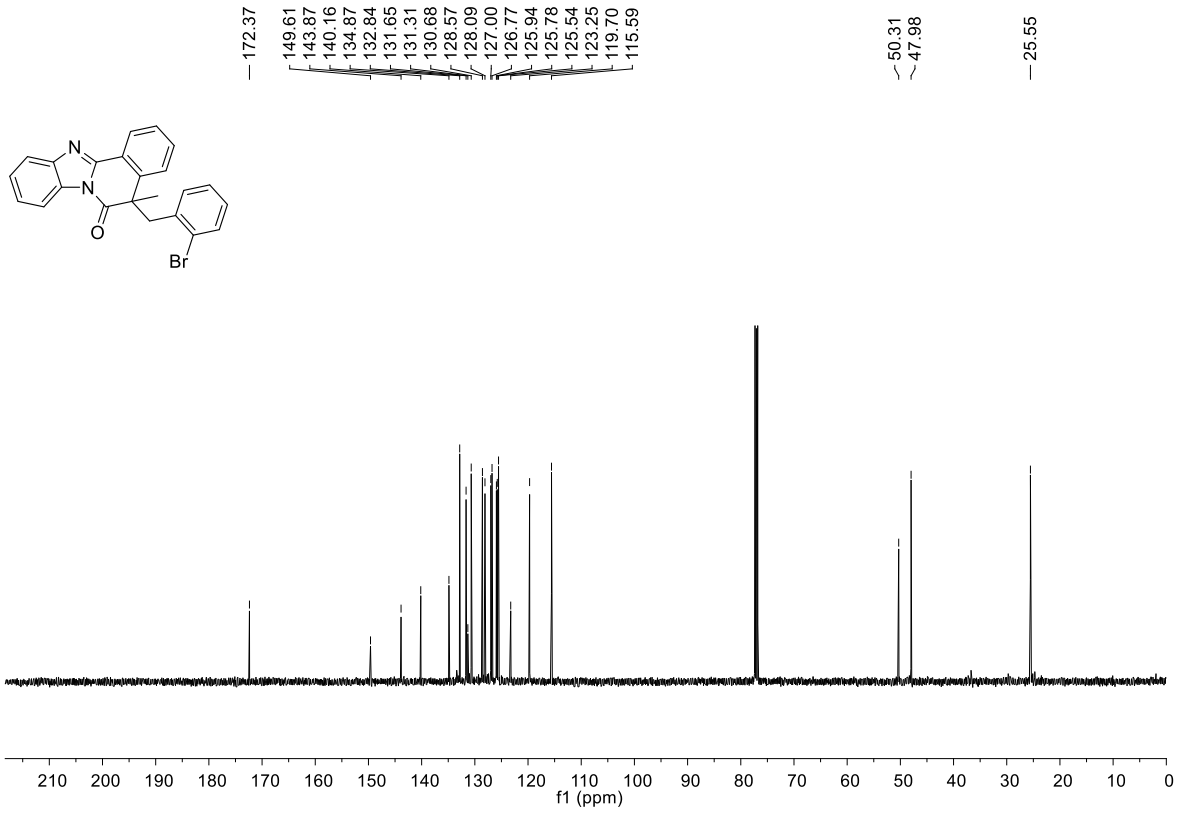
^{13}C{^{1}H} NMR spectrum of 3aq ($CDCl_3$, 126 MHz)



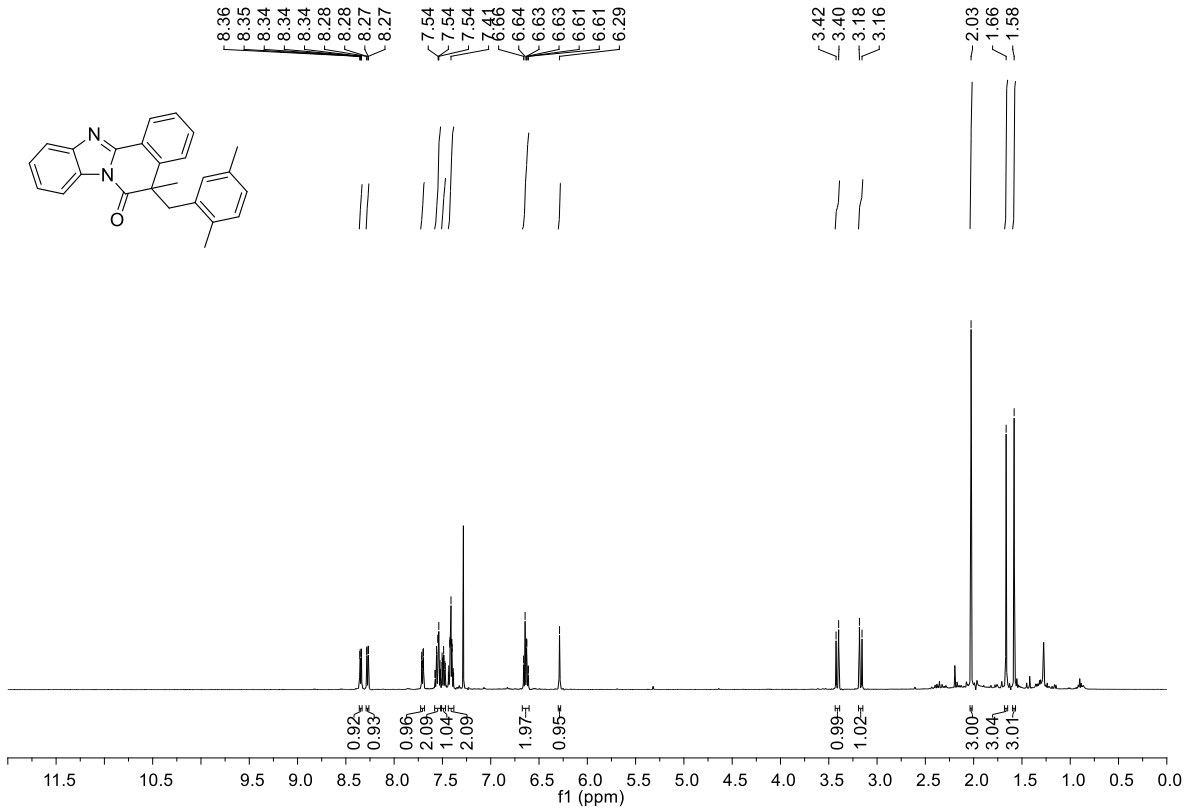
^{1}H NMR spectrum of 3ar ($CDCl_3$, 500 MHz)



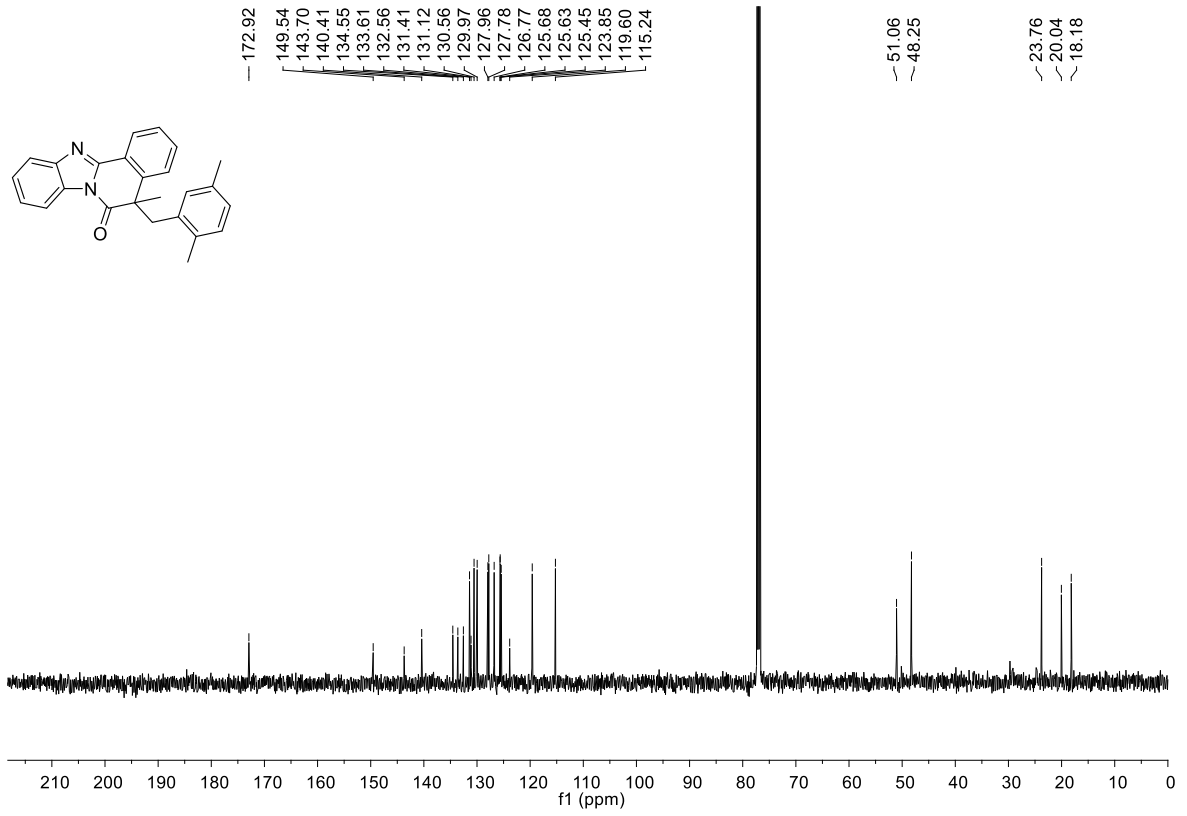
^{13}C{^{1}H} NMR spectrum of 3ar ($CDCl_3$, 126 MHz)



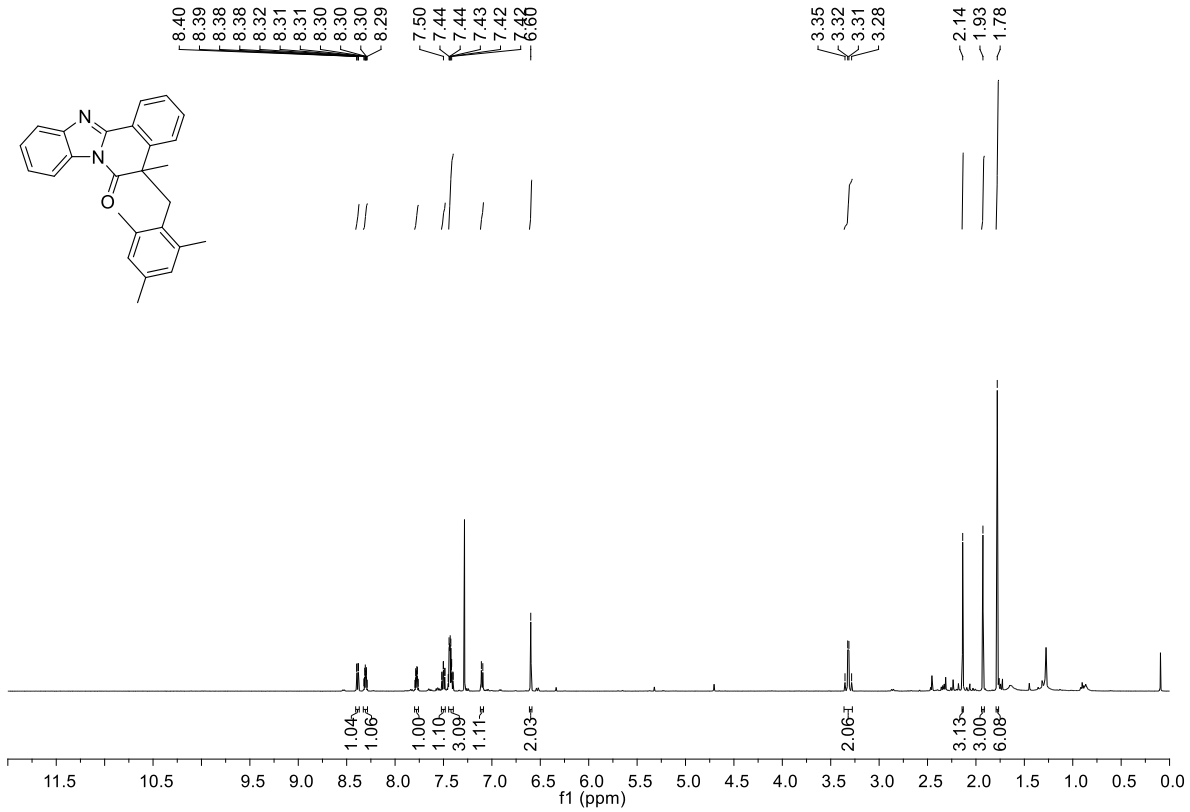
^{1}H NMR spectrum of 3as ($CDCl_3$, 500 MHz)



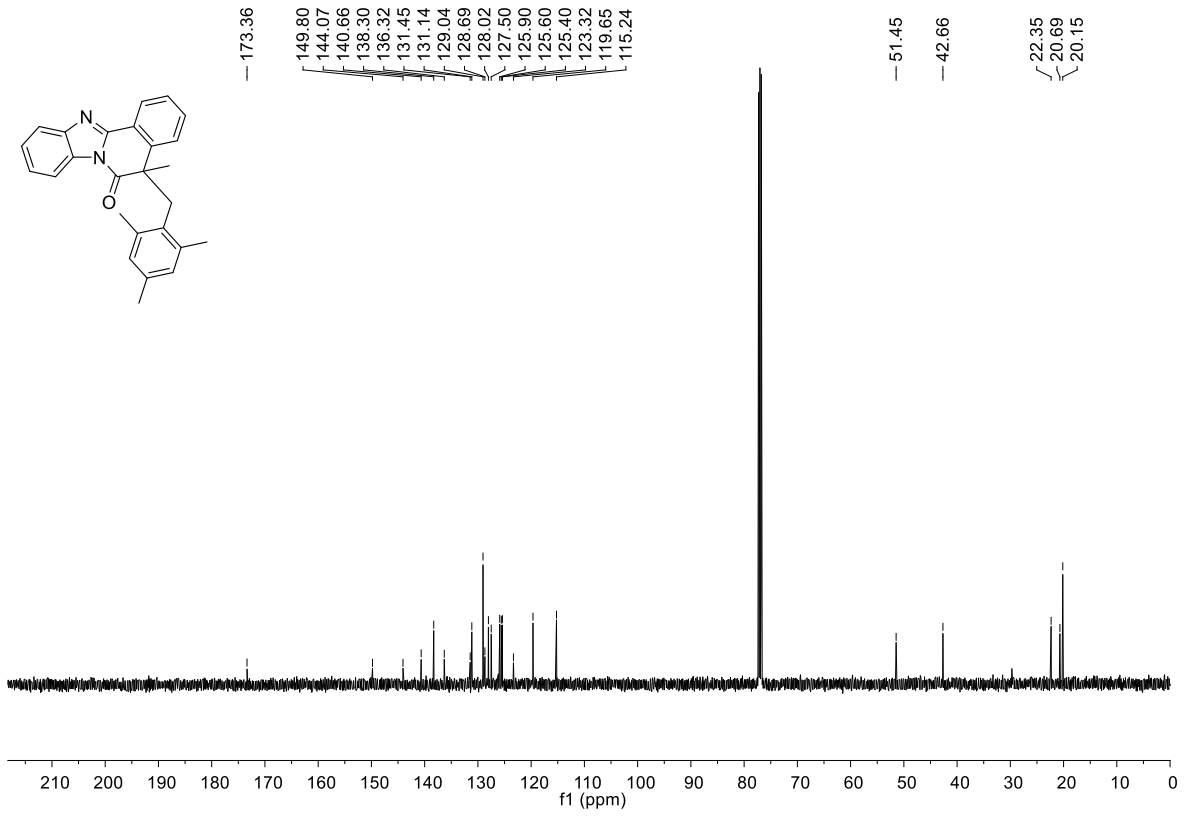
^{13}C{^{1}H} NMR spectrum of 3as ($CDCl_3$, 126 MHz)



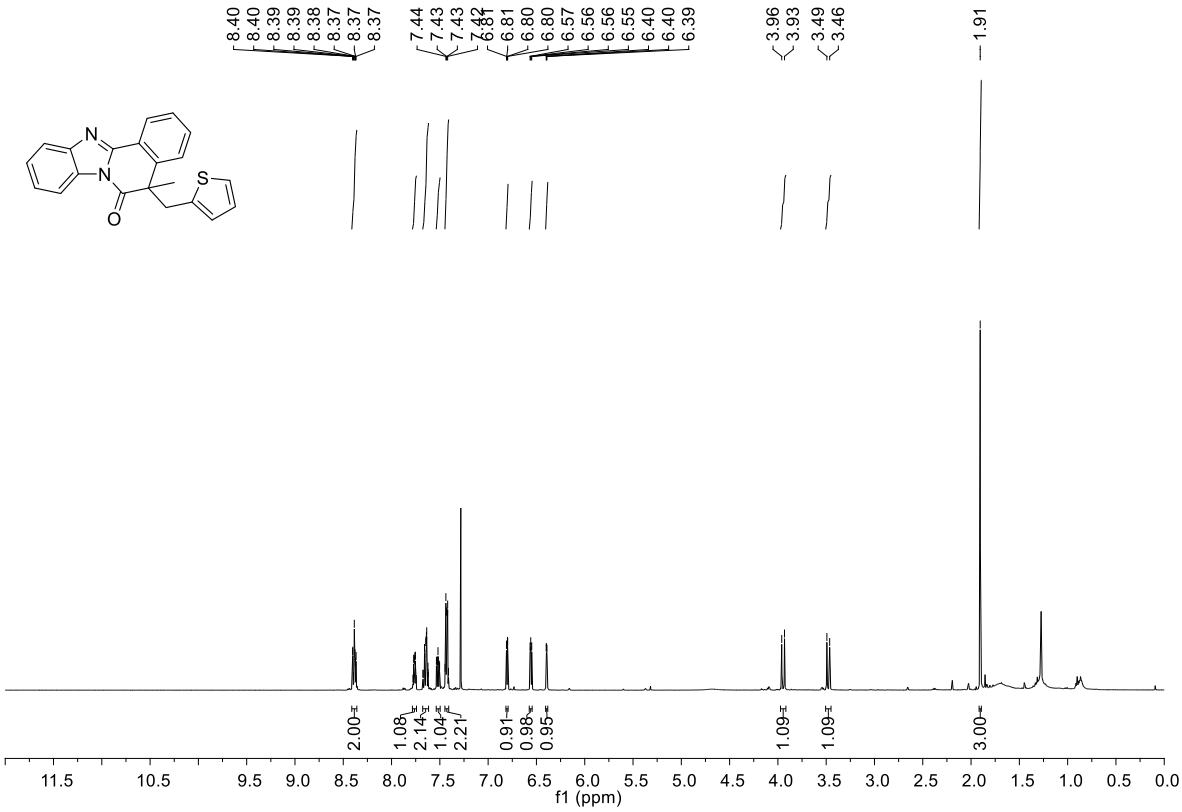
^{1}H NMR spectrum of 3at ($CDCl_3$, 500 MHz)



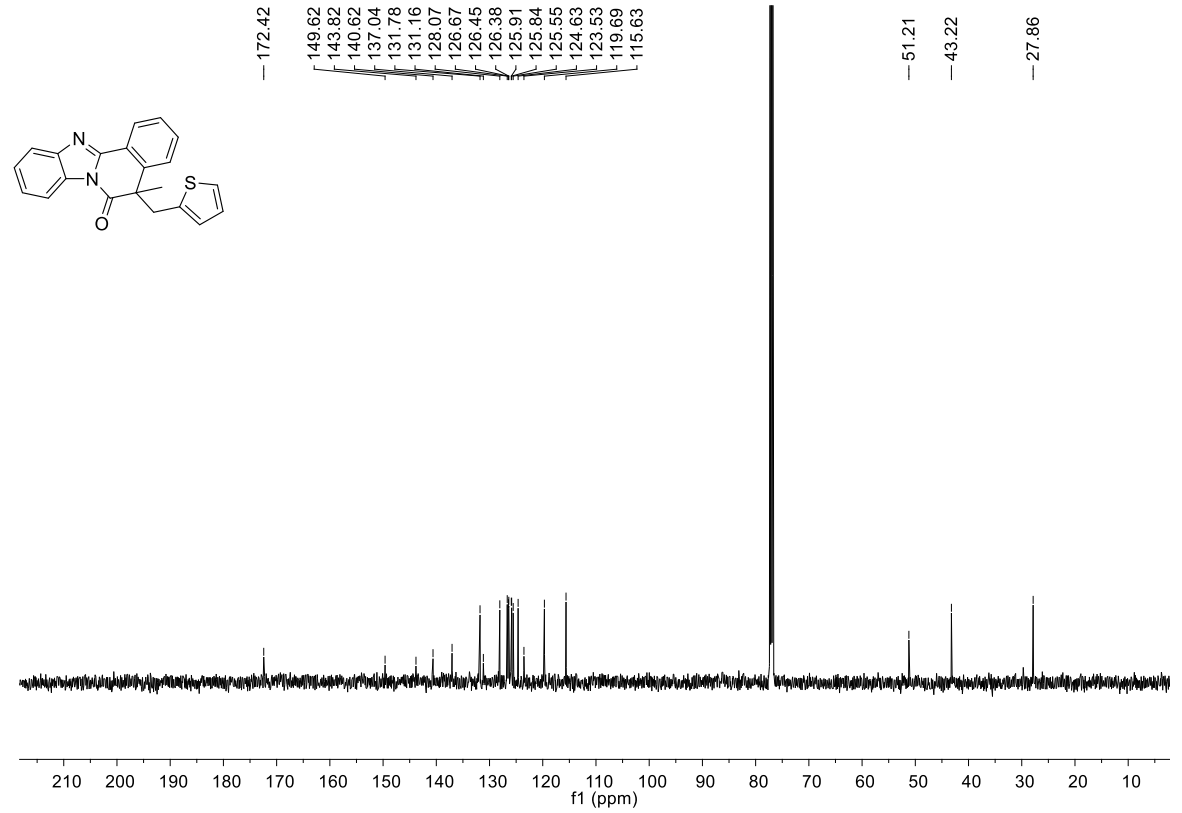
^{13}C{^{1}H} NMR spectrum of 3at ($CDCl_3$, 126 MHz)



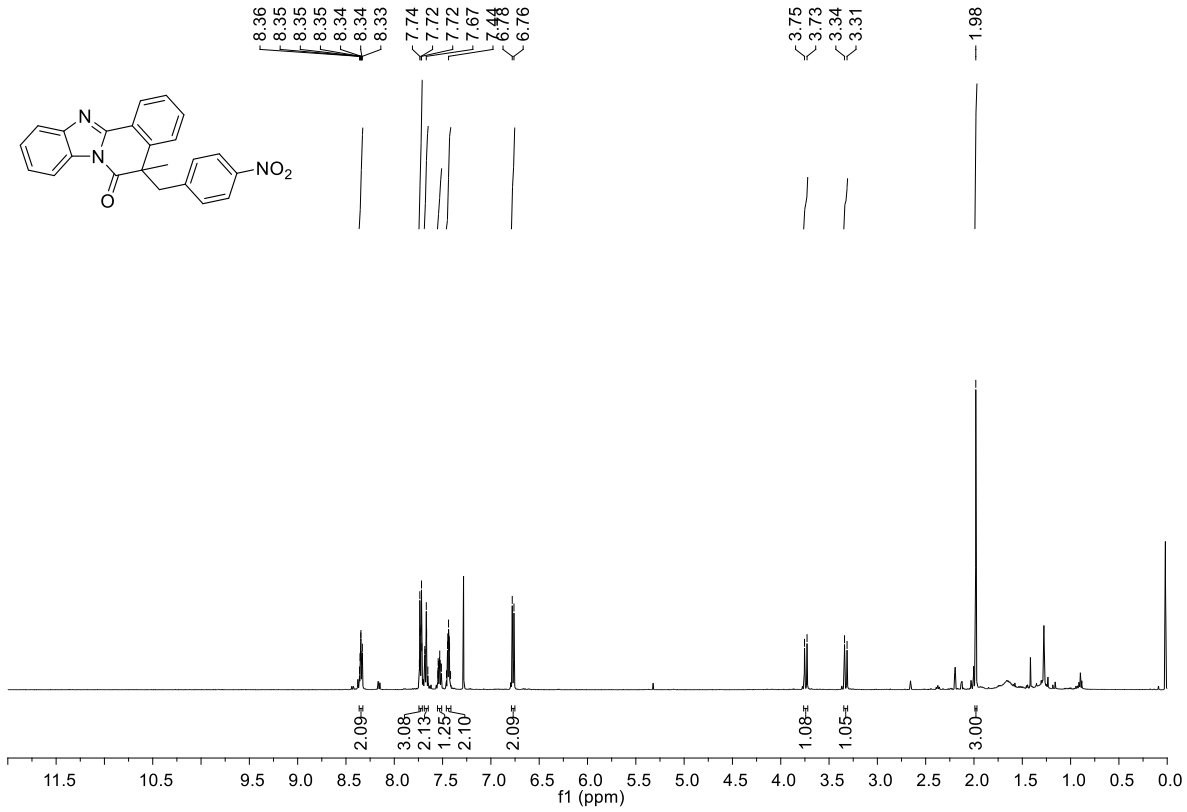
^{1}H NMR spectrum of 3au ($CDCl_3$, 500 MHz)



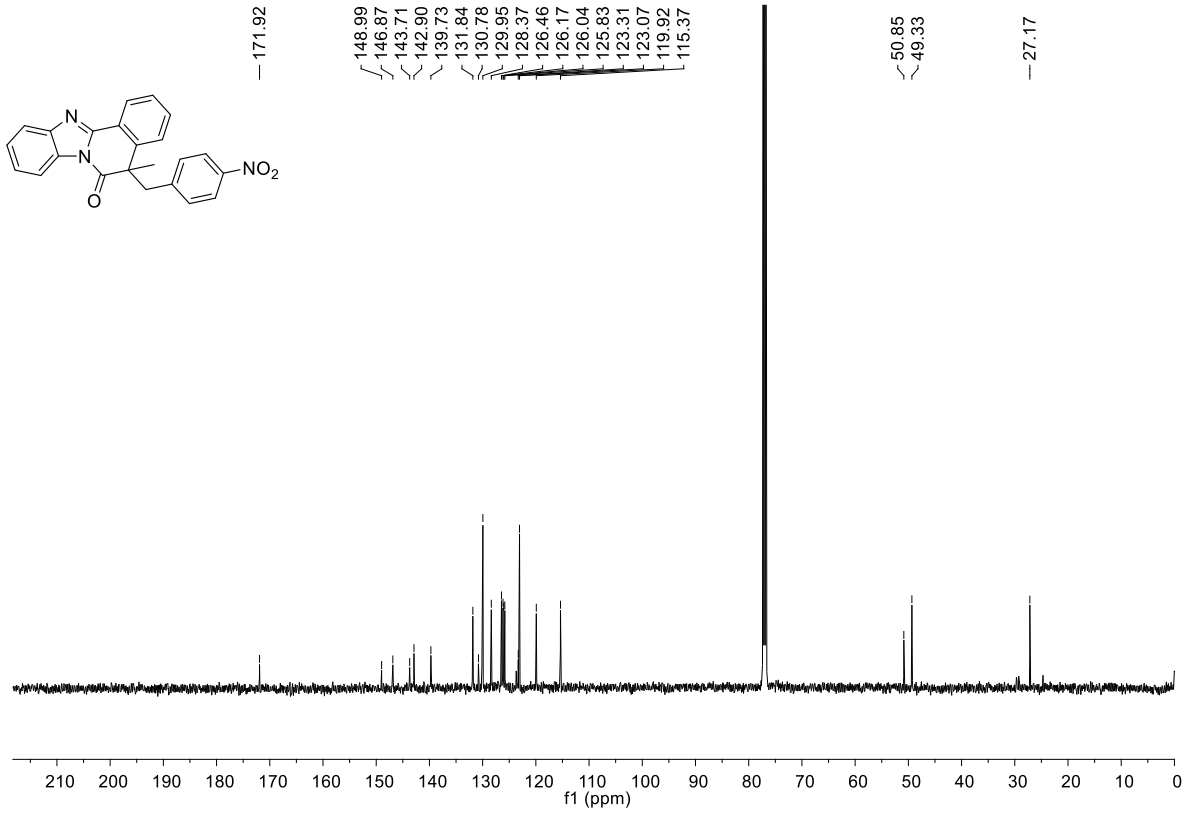
^{13}C{^{1}H} NMR spectrum of 3au ($CDCl_3$, 126 MHz)



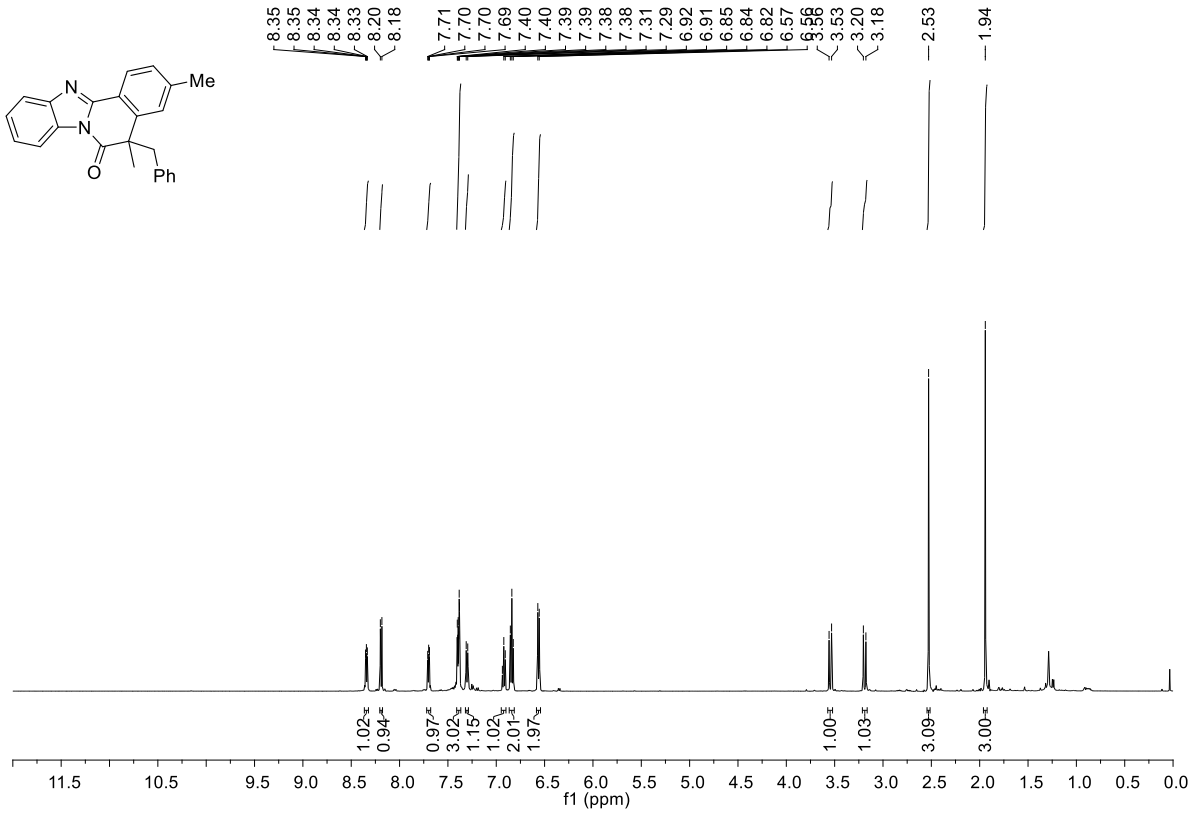
^{1}H NMR spectrum of 3av ($CDCl_3$, 500 MHz)



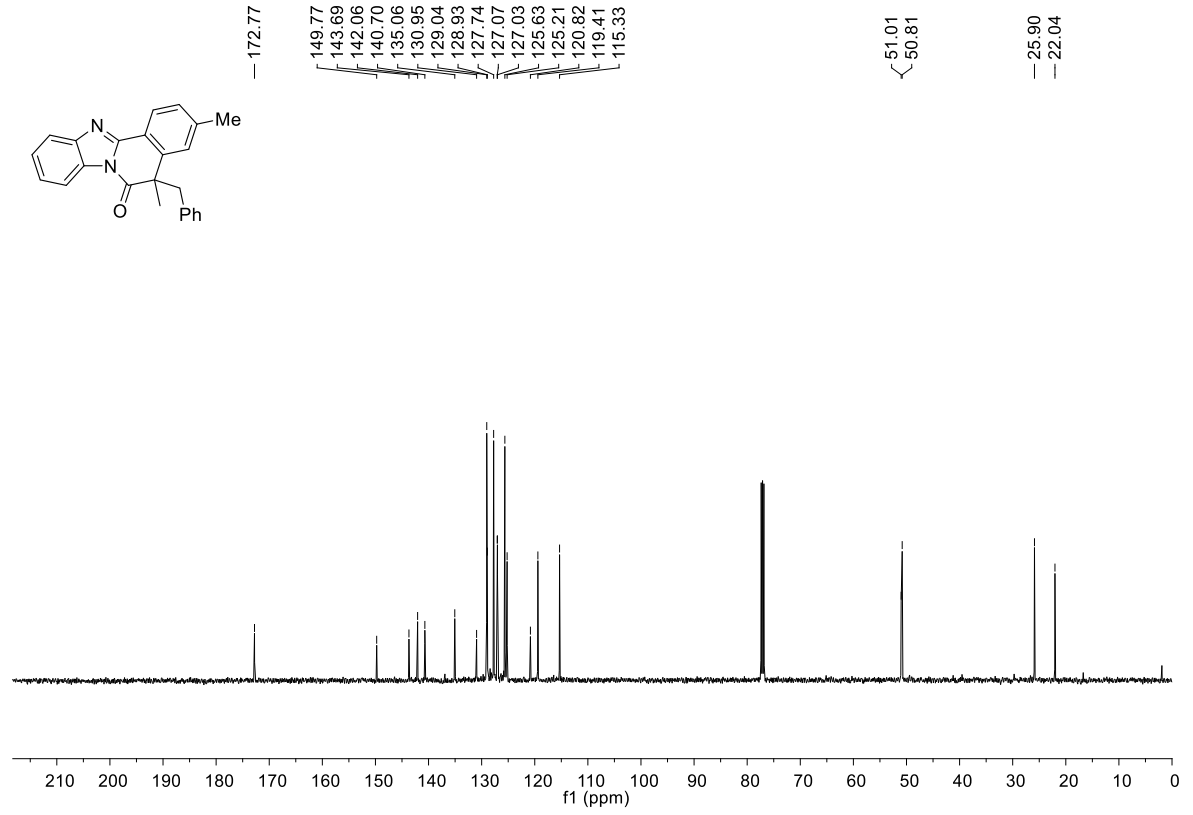
^{13}C{^{1}H} NMR spectrum of 3av ($CDCl_3$, 126 MHz)



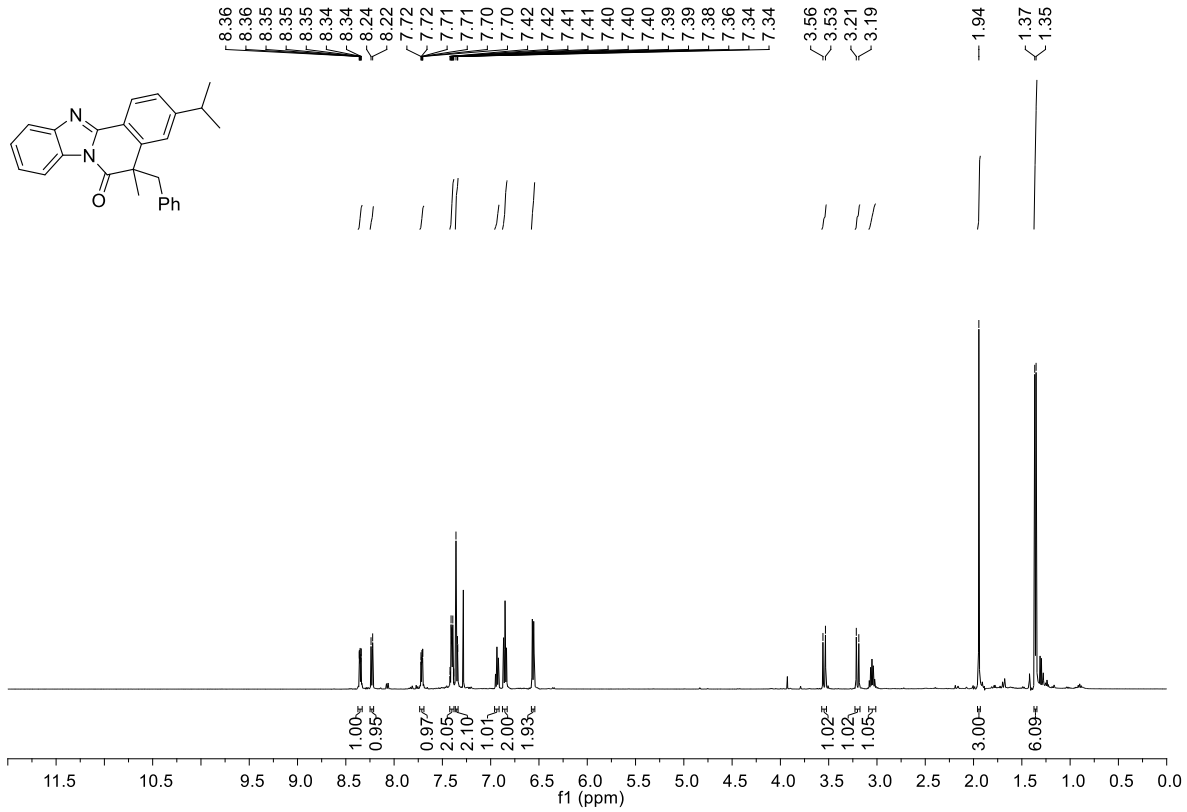
^{1}H NMR spectrum of 3ba ($CDCl_3$, 500 MHz)



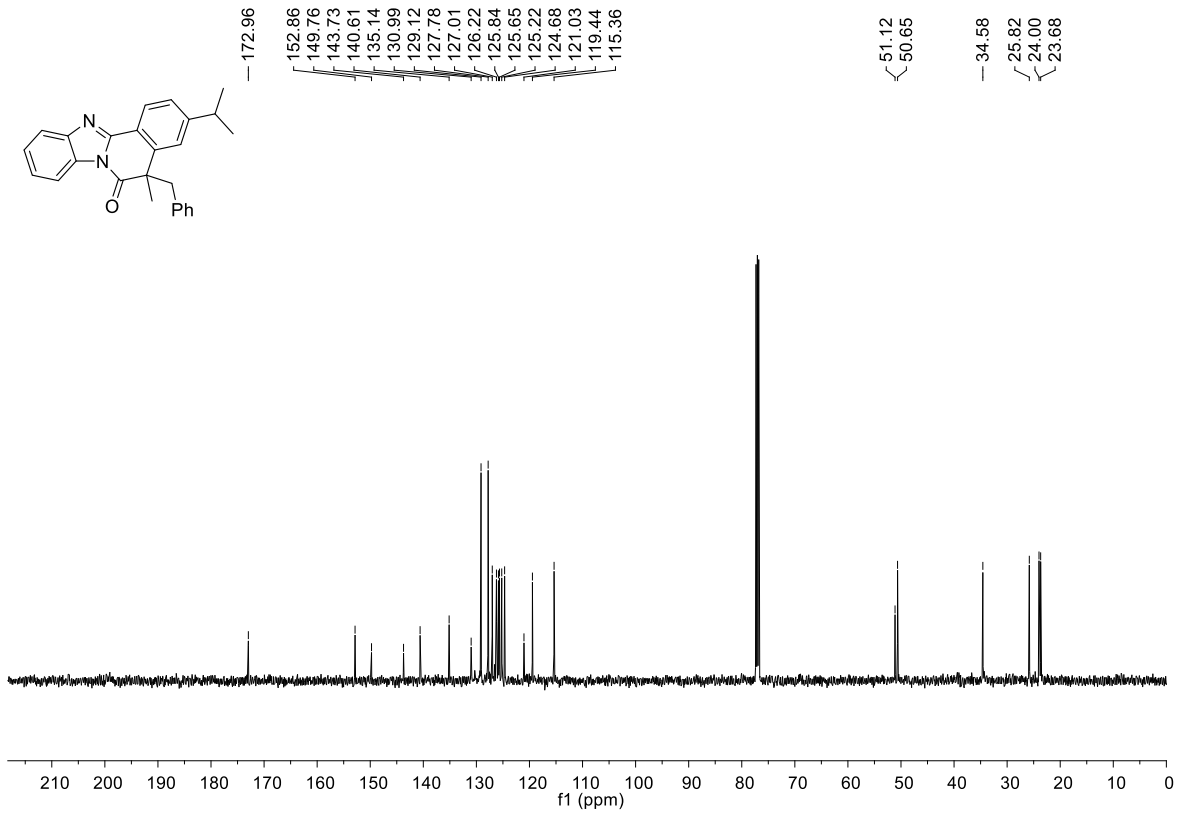
^{13}C{^{1}H} NMR spectrum of 3ba ($CDCl_3$, 126 MHz)



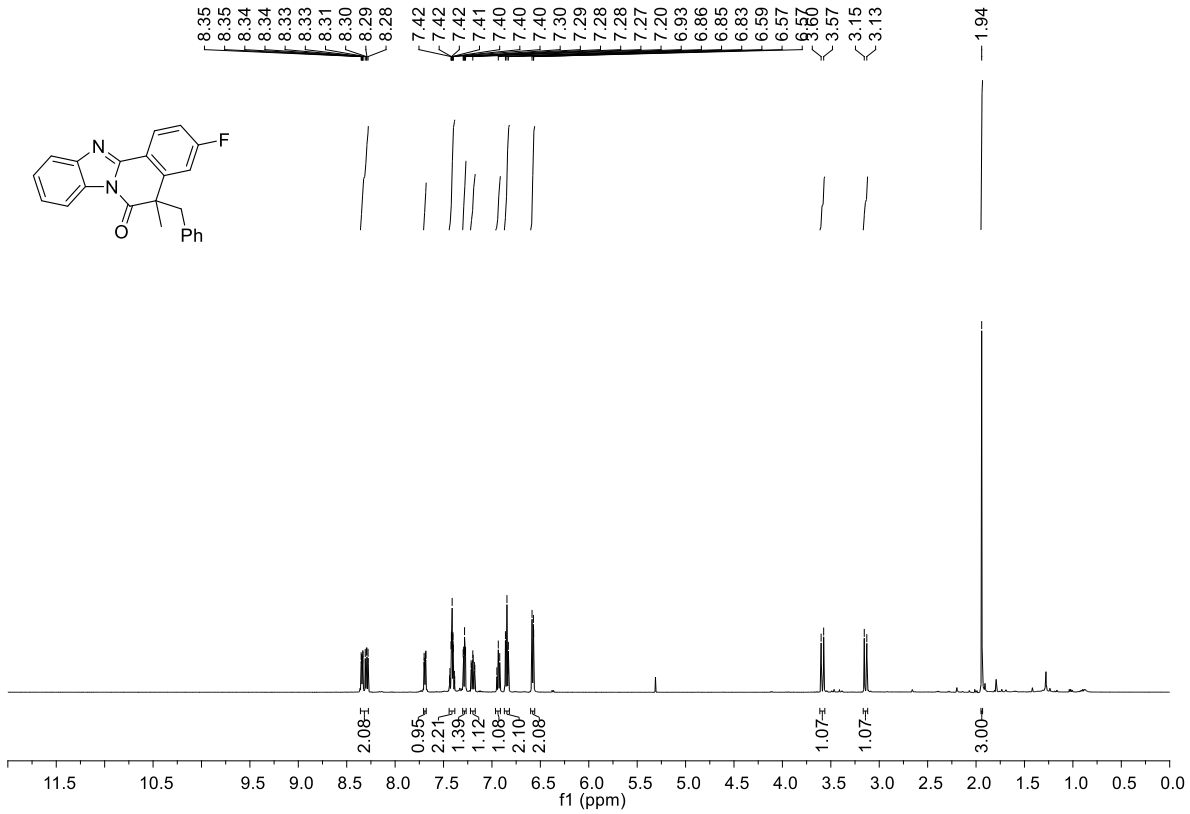
^{1}H NMR spectrum of 3ca ($CDCl_3$, 500 MHz)



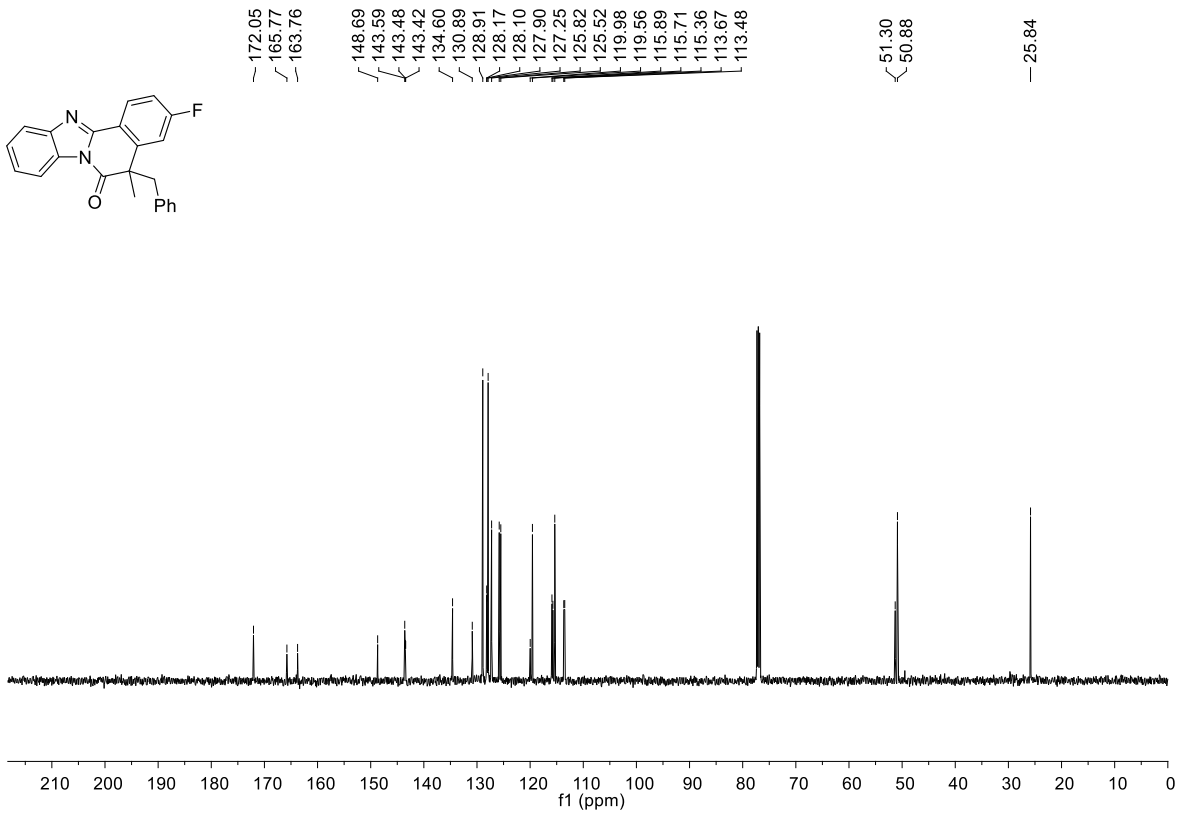
^{13}C{^{1}H} NMR spectrum of 3ca ($CDCl_3$, 126 MHz)



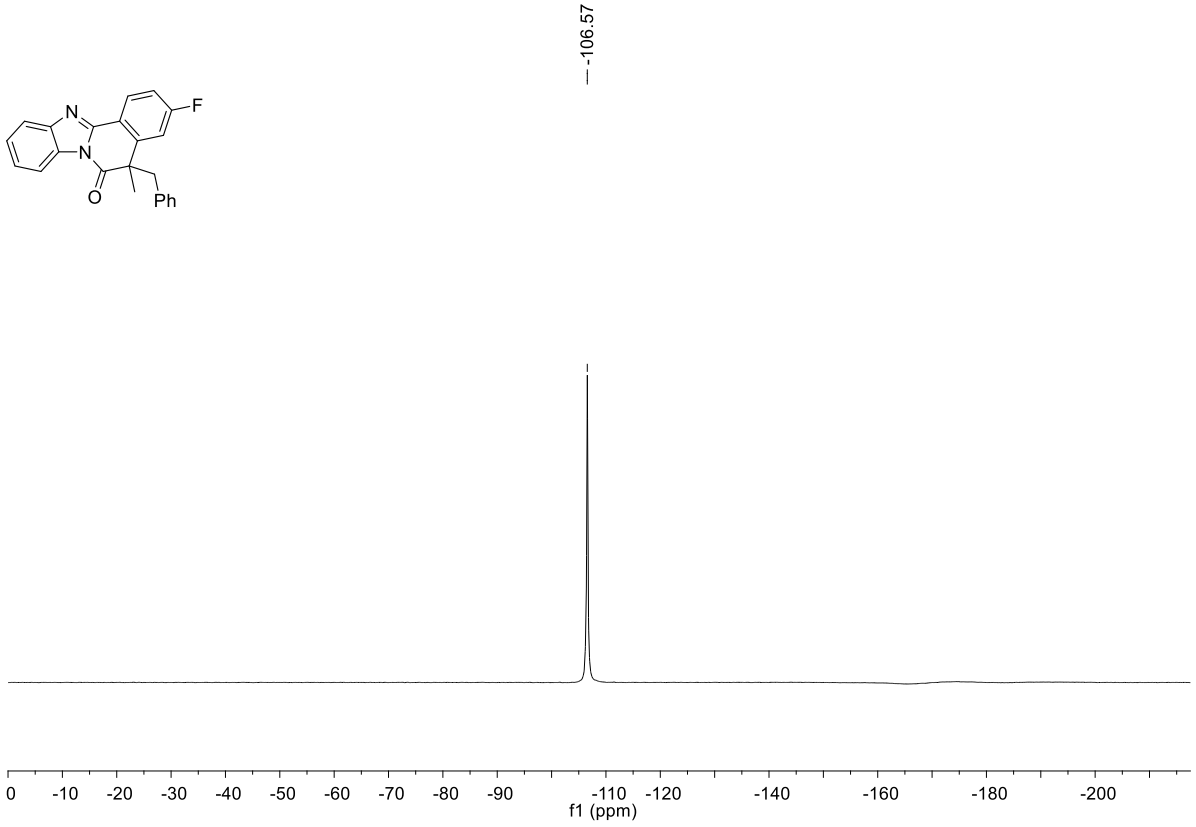
^{1}H NMR spectrum of 3da ($CDCl_3$, 500 MHz)



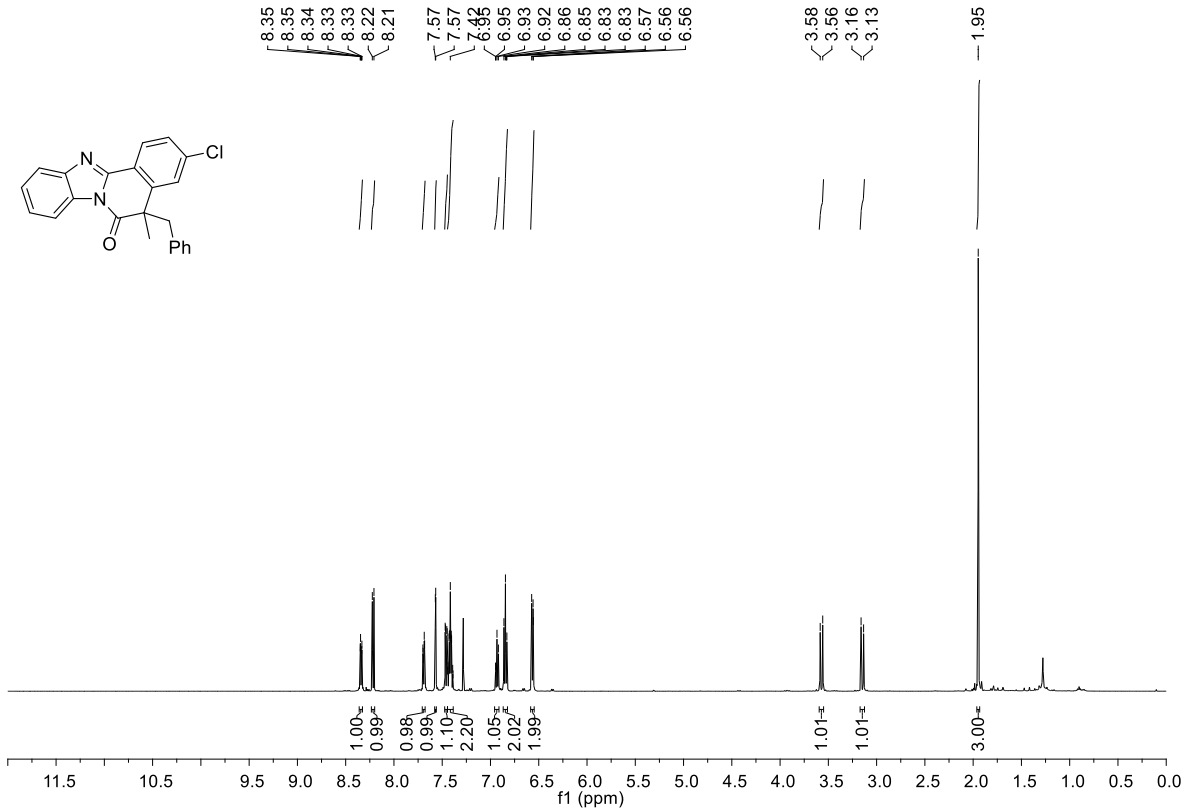
^{13}C{^{1}H} NMR spectrum of 3da ($CDCl_3$, 126 MHz)



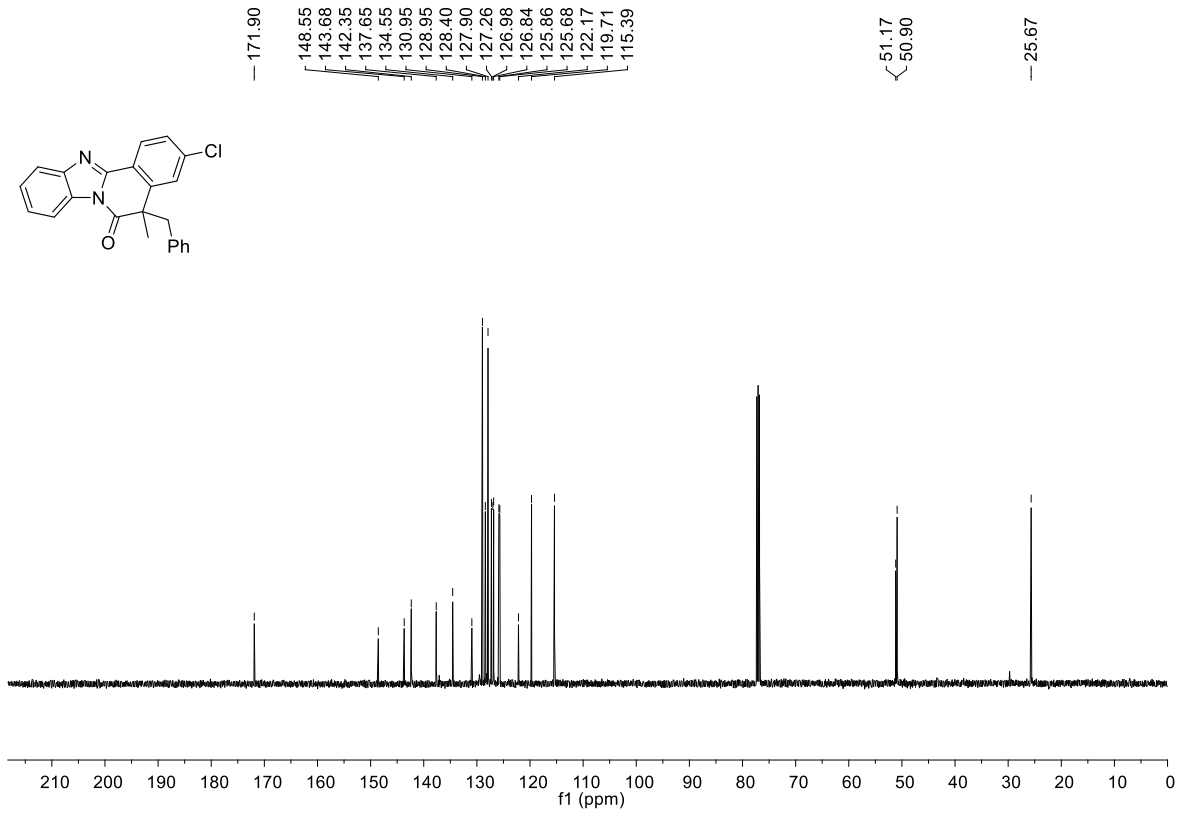
$^{19}F\{^{1}H\}$ NMR spectrum of 3da ($CDCl_3$, 471 MHz)



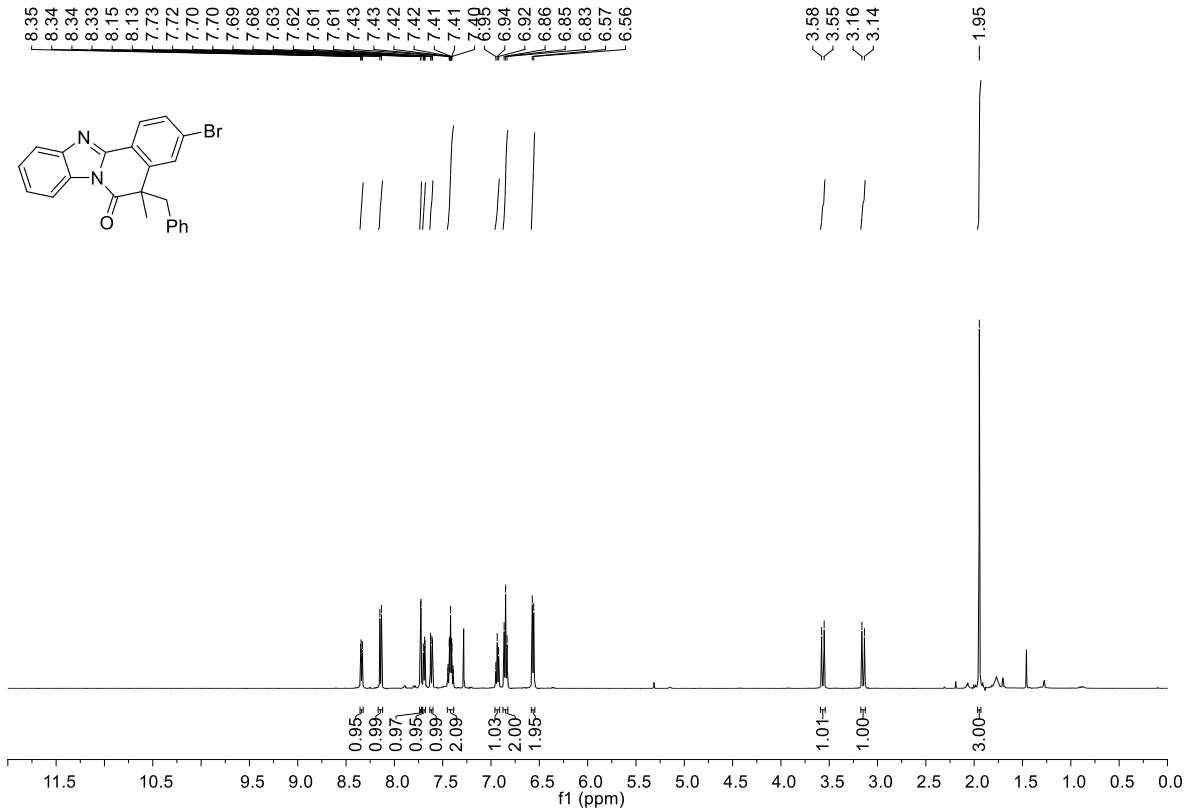
^{1}H NMR spectrum of 3ea ($CDCl_3$, 500 MHz)



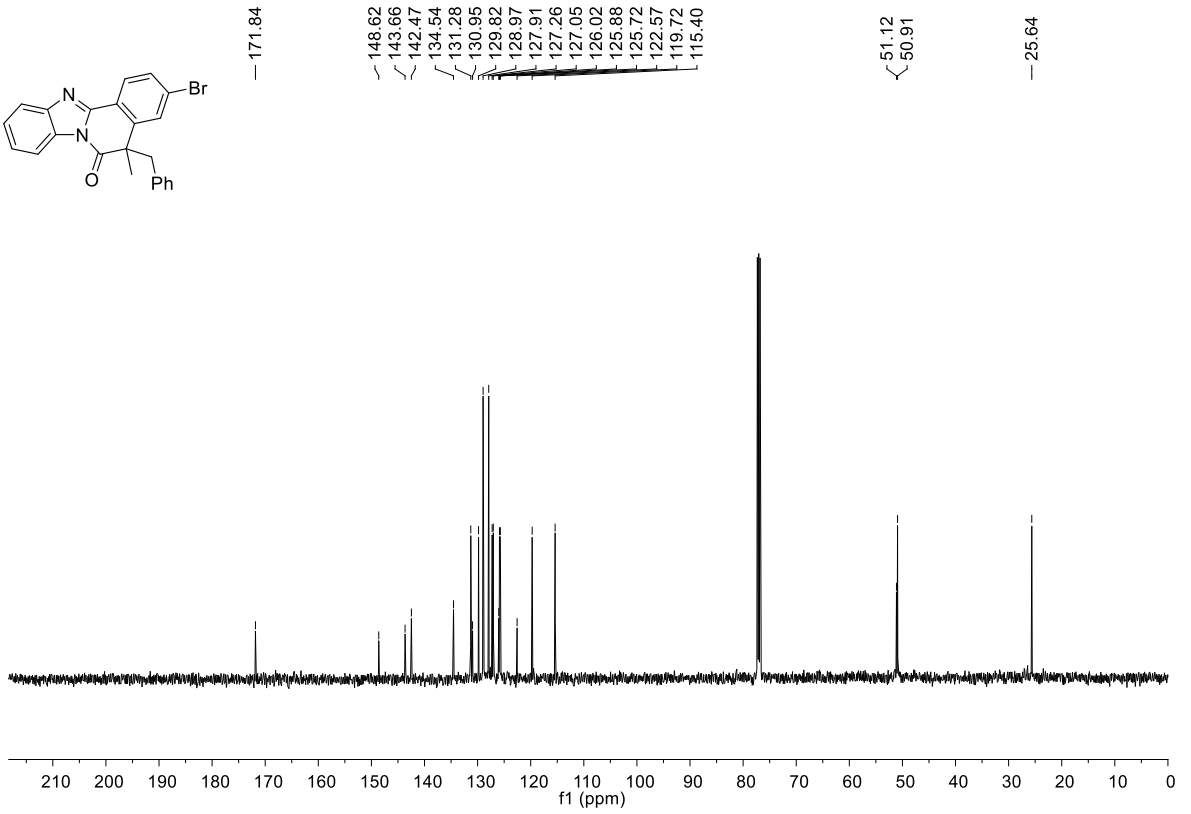
^{13}C{^{1}H} NMR spectrum of 3ea ($CDCl_3$, 126 MHz)



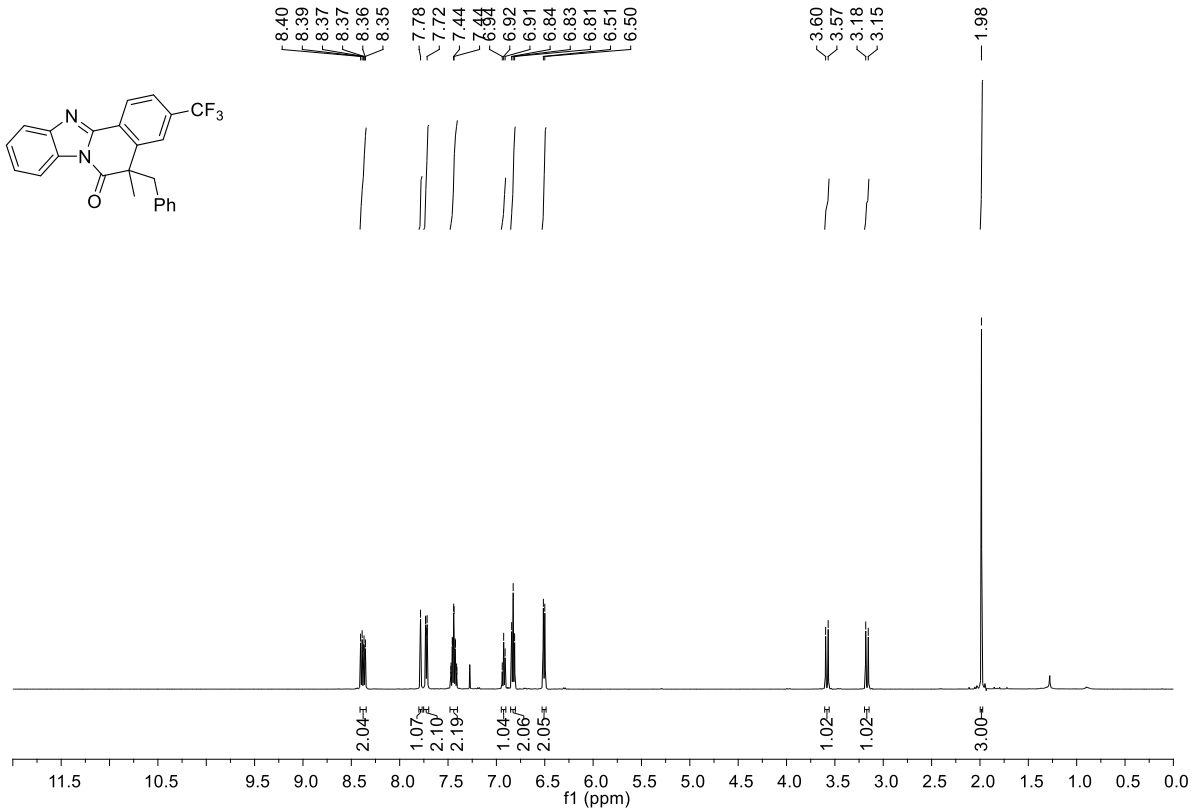
^{1}H NMR spectrum of 3fa ($CDCl_3$, 500 MHz)



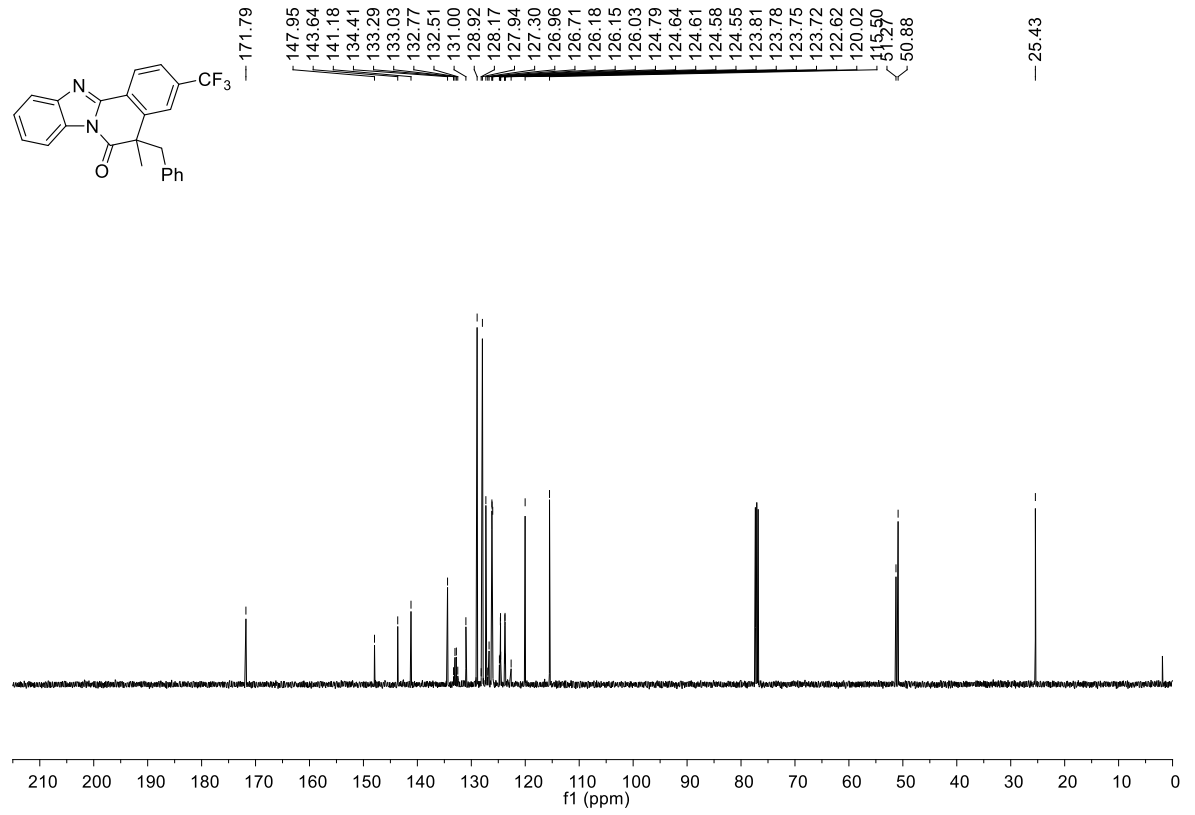
^{13}C{^{1}H} NMR spectrum of 3fa ($CDCl_3$, 126 MHz)



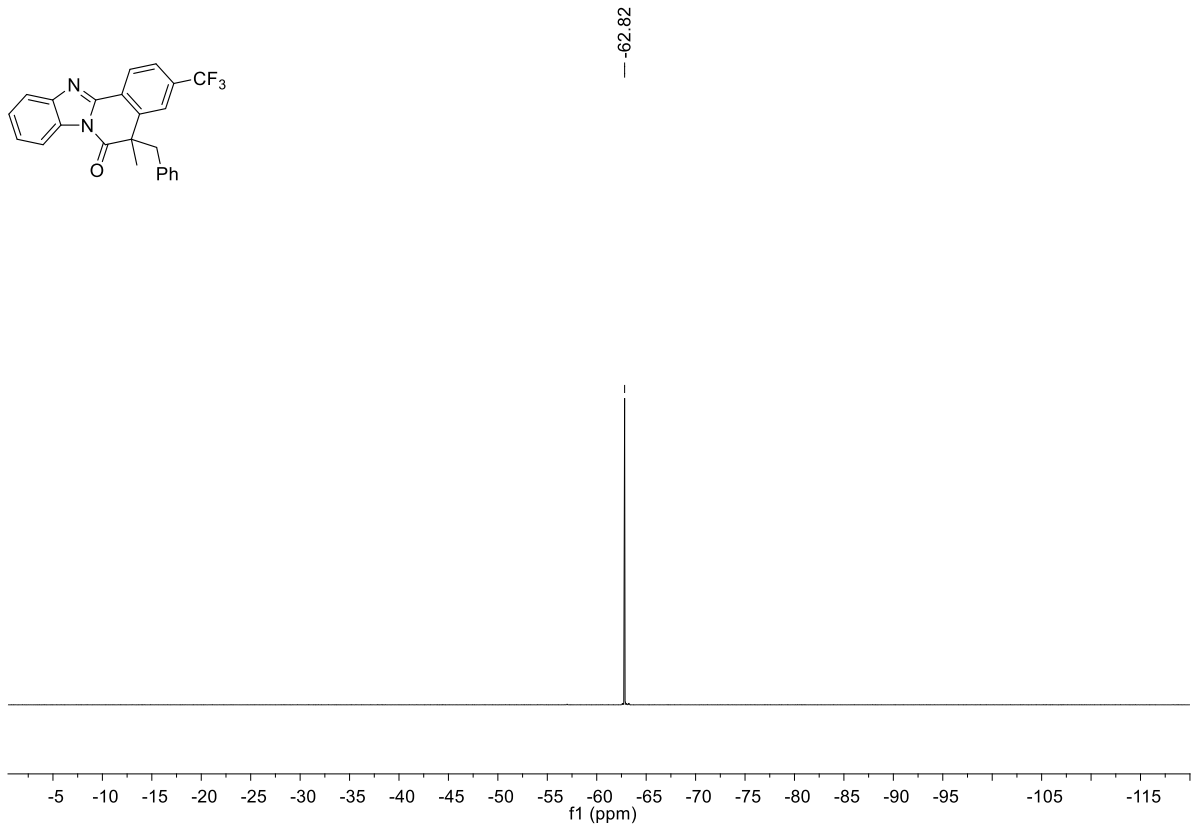
^{1}H NMR spectrum of 3ga ($CDCl_3$, 500 MHz)



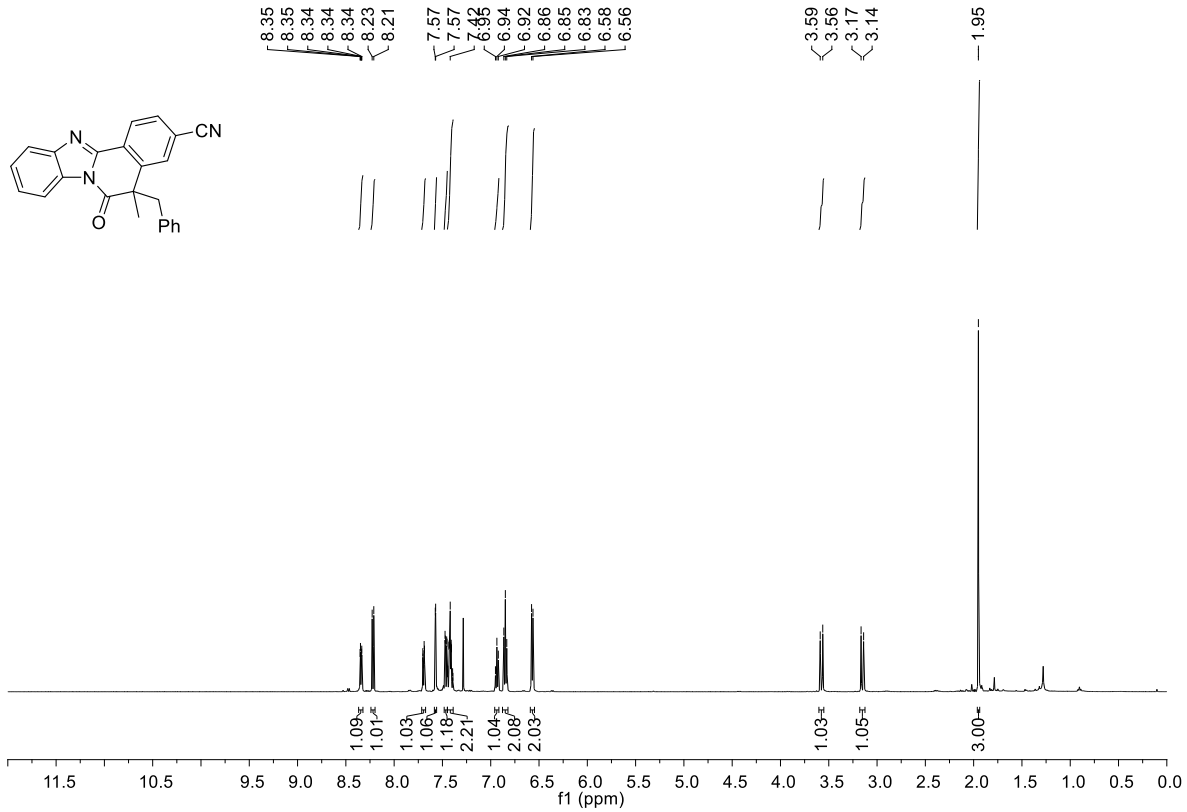
^{13}C{^{1}H} NMR spectrum of 3ga ($CDCl_3$, 126 MHz)



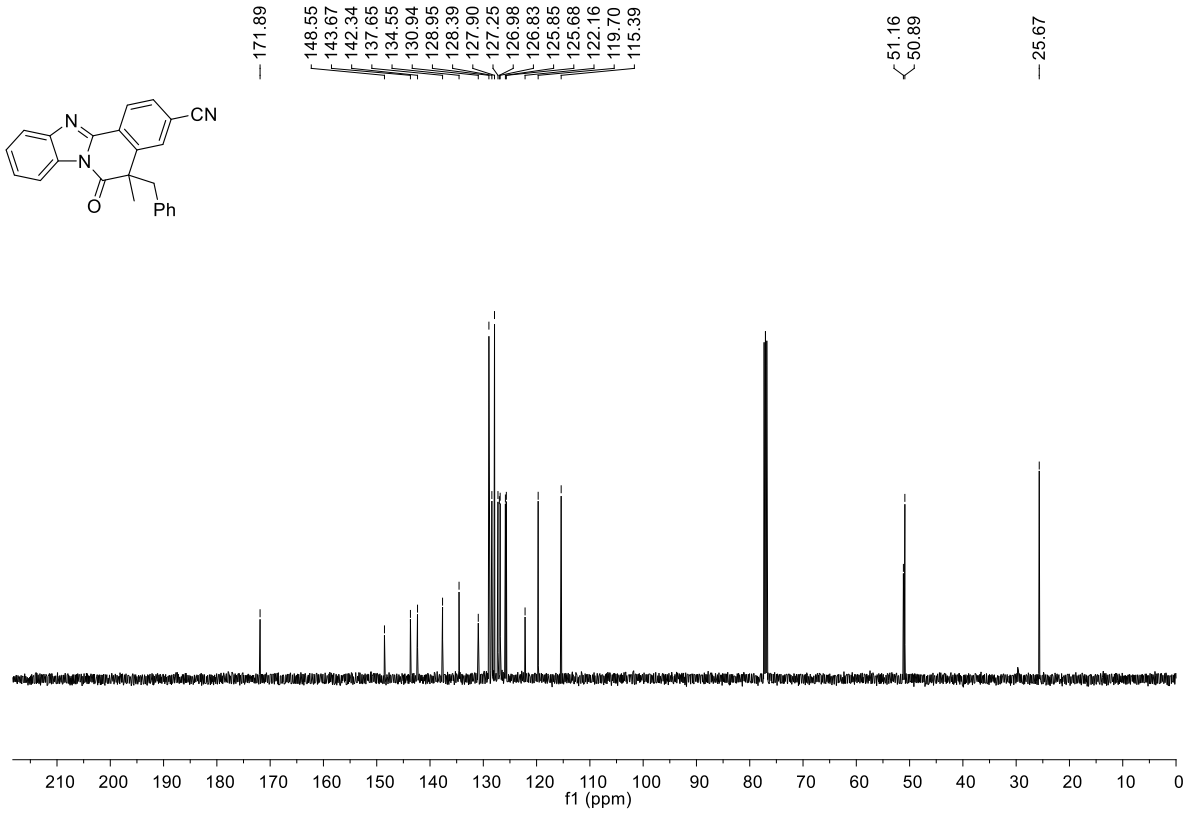
$^{19}F\{^{1}H\}$ NMR spectrum of 3ga ($CDCl_3$, 471 MHz)



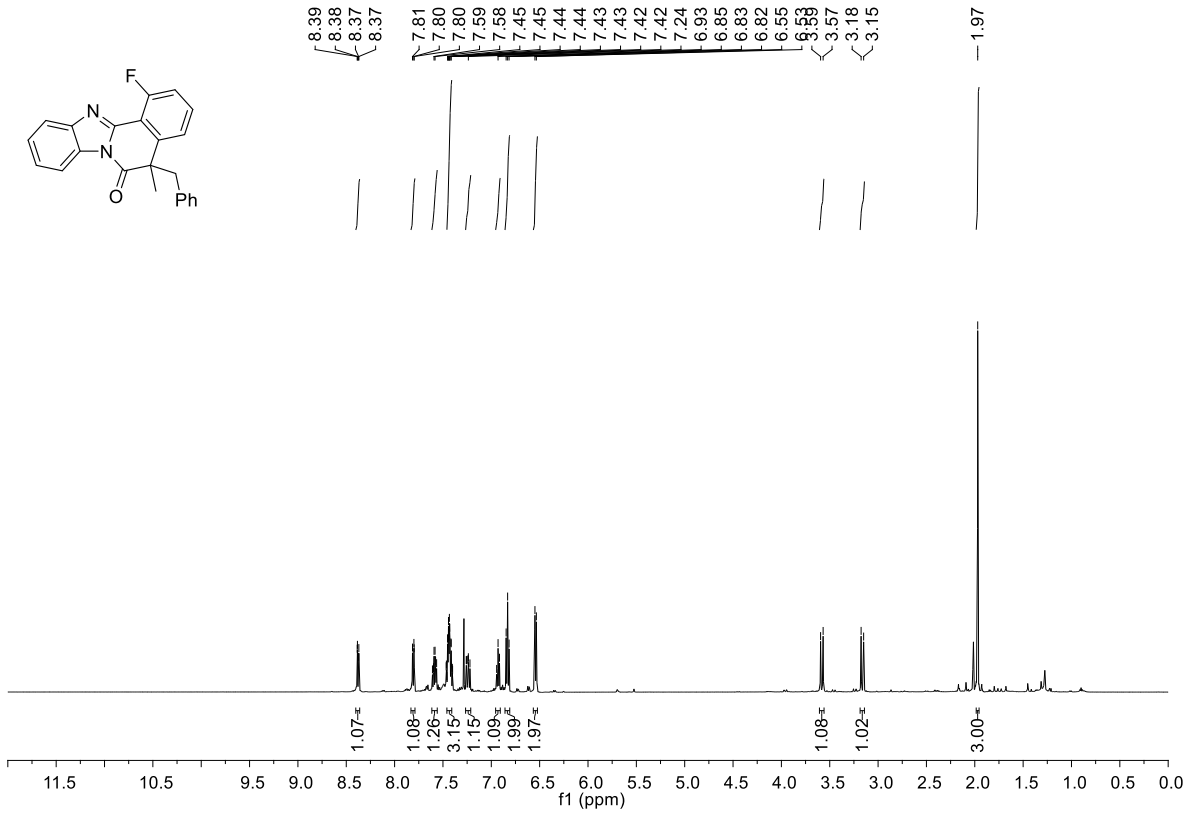
^{1}H NMR spectrum of 3ha ($CDCl_3$, 500 MHz)



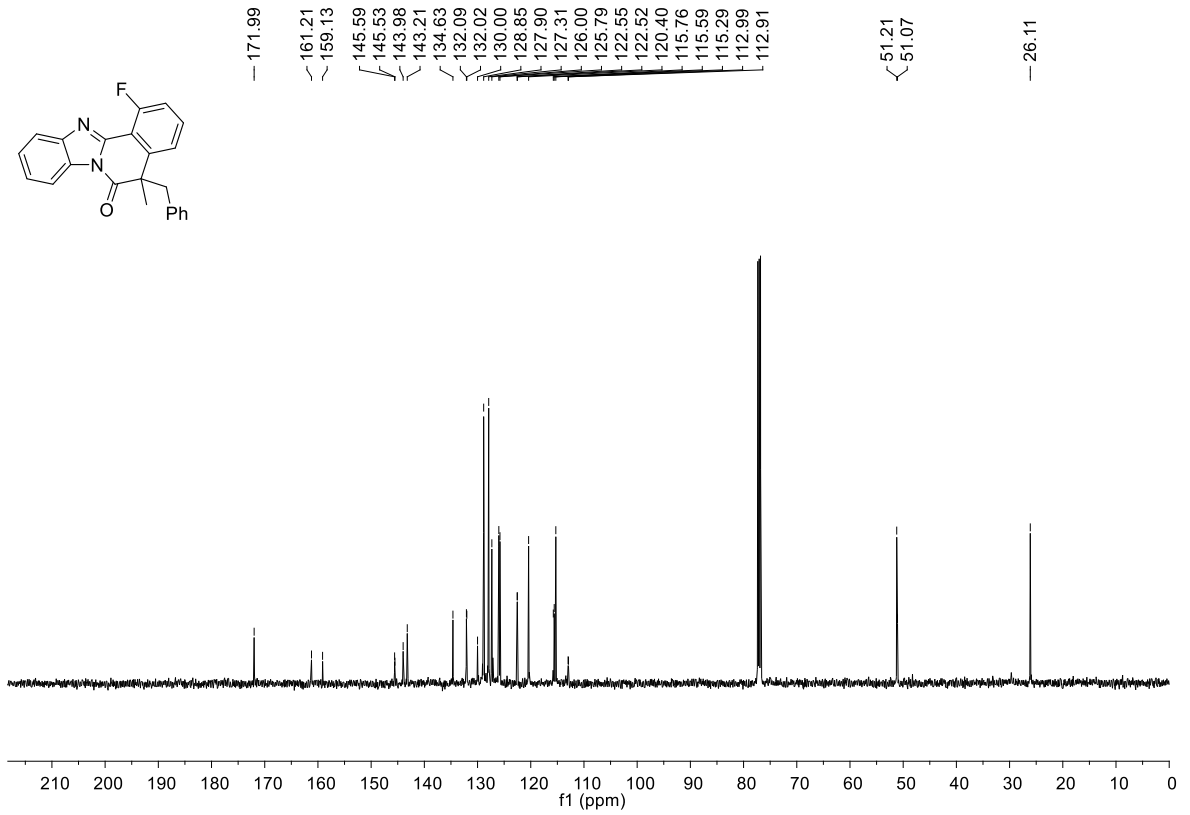
^{13}C{^{1}H} NMR spectrum of 3ha ($CDCl_3$, 126 MHz)



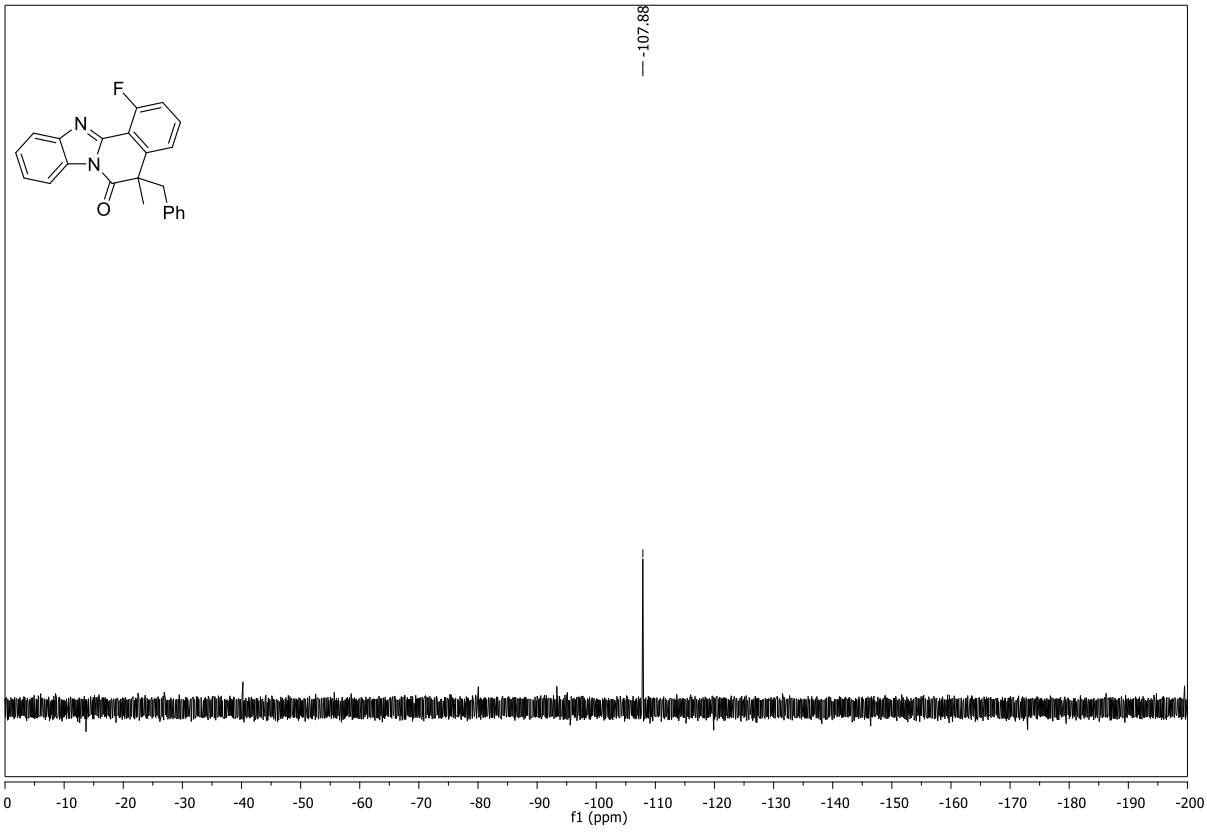
^{1}H NMR spectrum of 3ia ($CDCl_3$, 500 MHz)



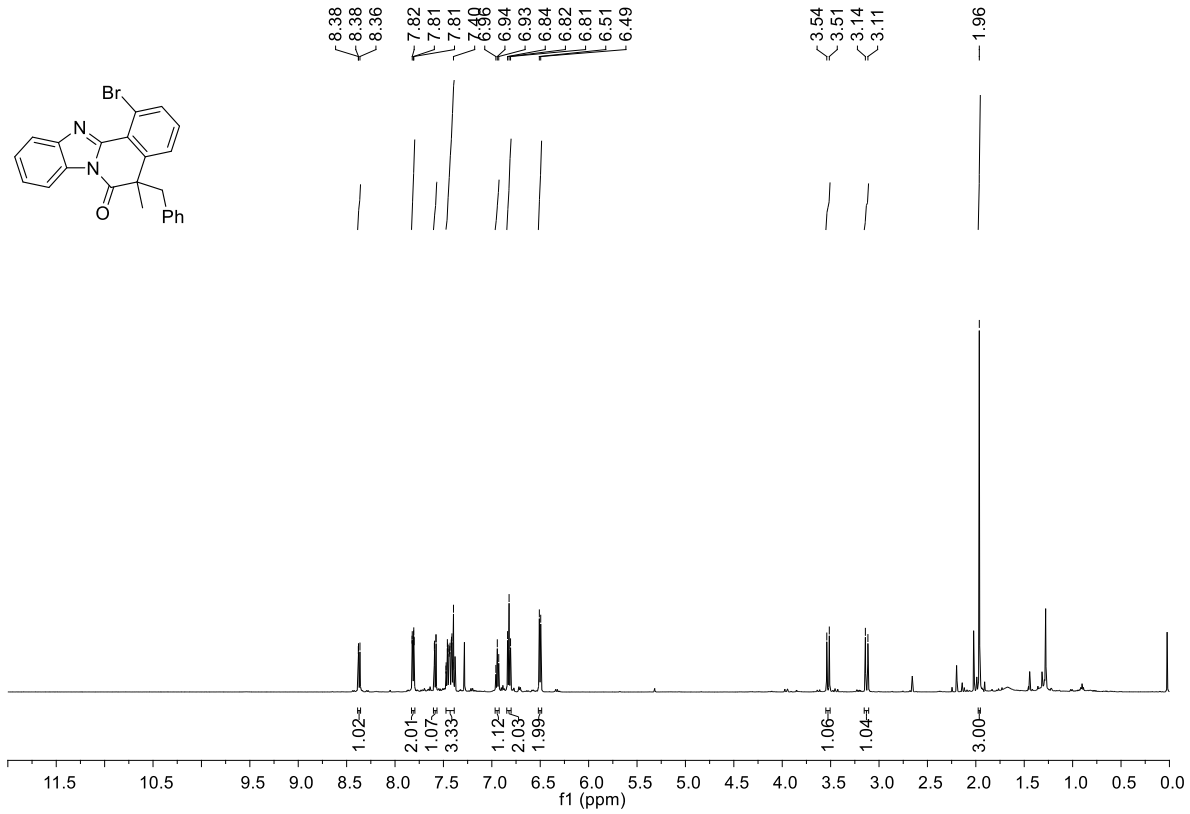
^{13}C{^{1}H} NMR spectrum of 3ia ($CDCl_3$, 126 MHz)



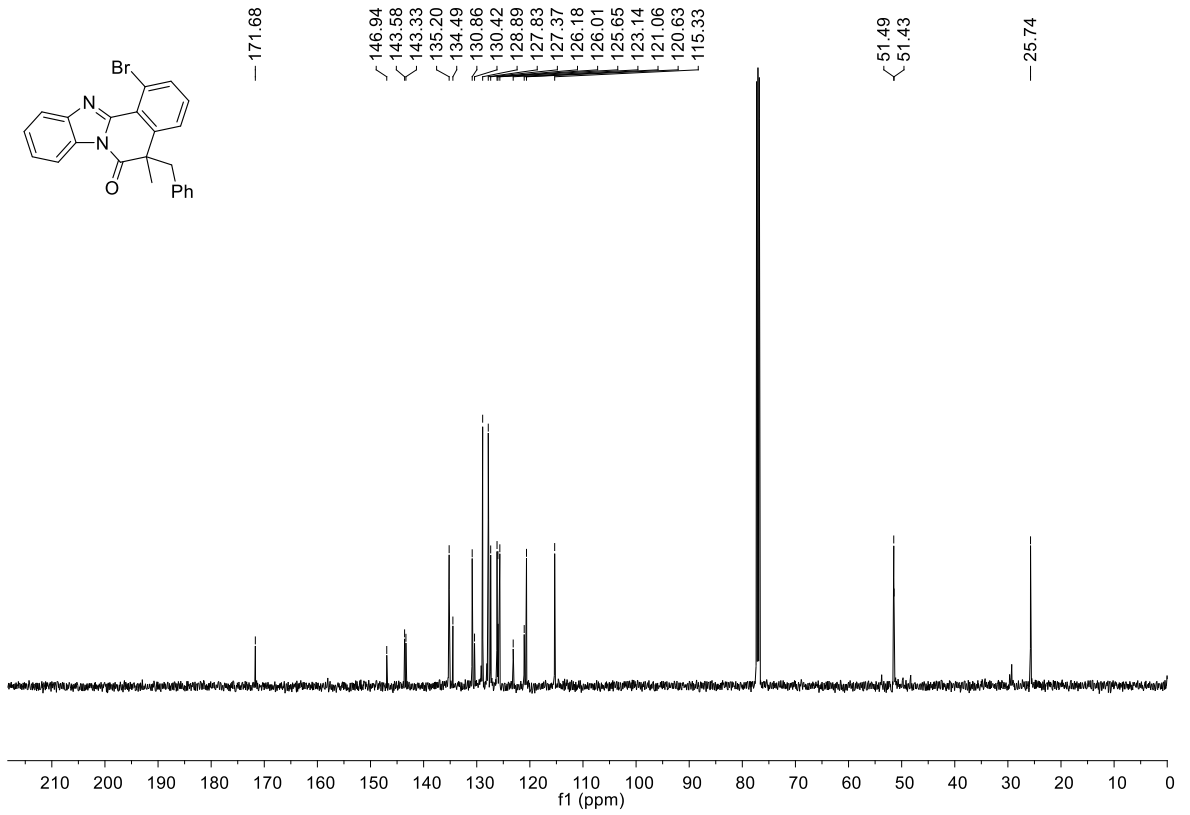
$^{19}F\{^{1}H\}$ NMR spectrum of 3ia ($CDCl_3$, 471 MHz)



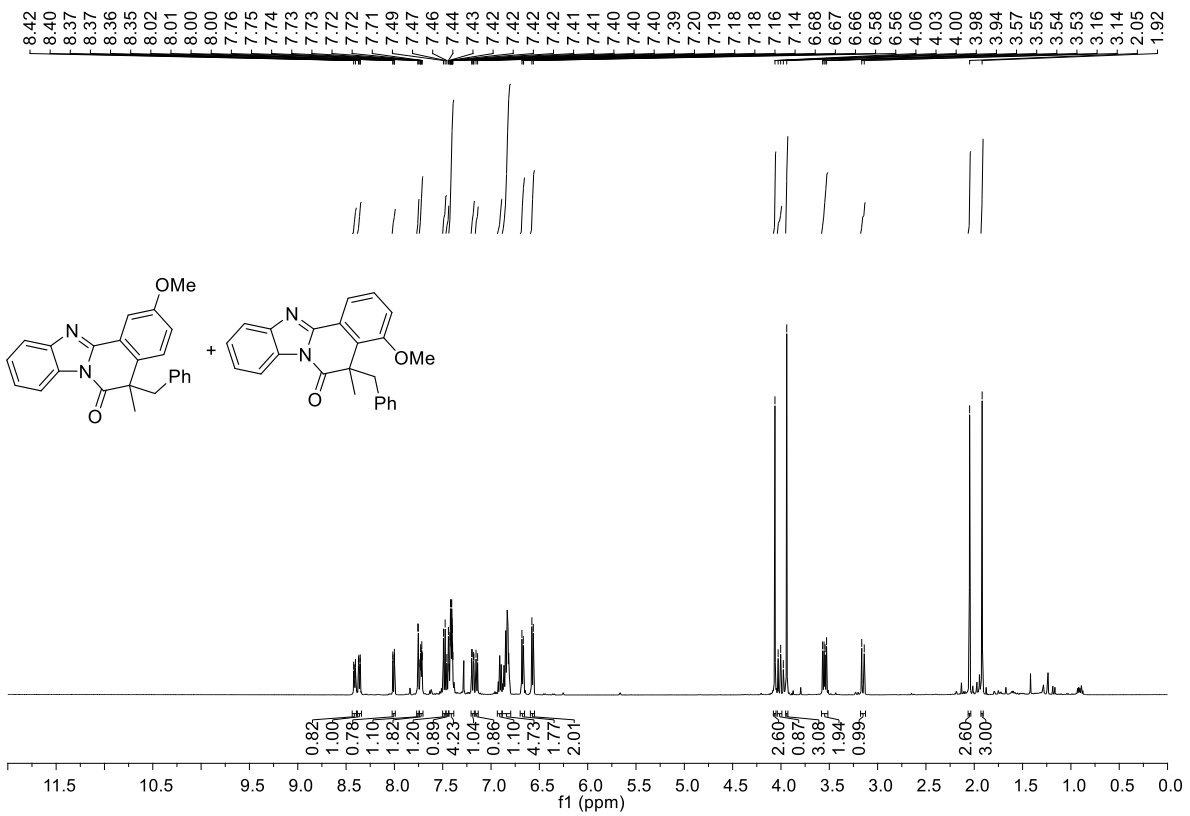
^{1}H NMR spectrum of 3ja ($CDCl_3$, 500 MHz)



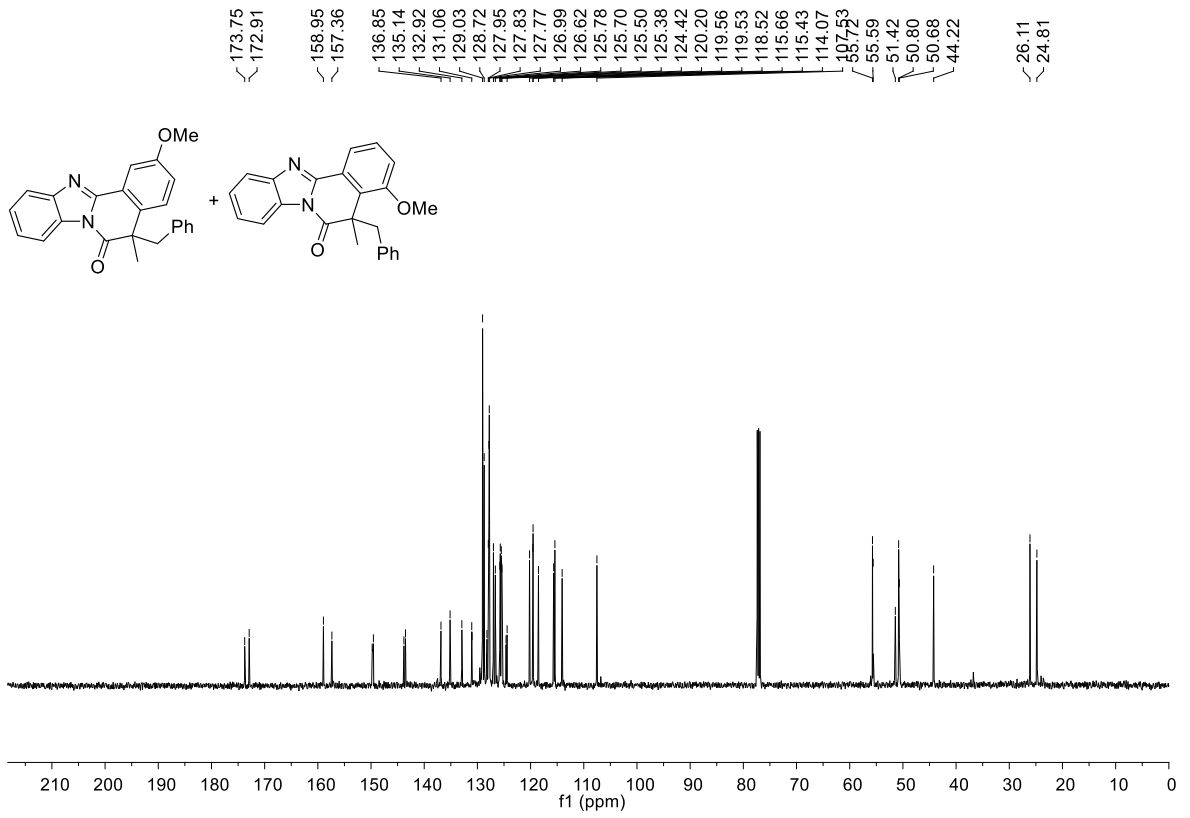
^{13}C{^{1}H} NMR spectrum of 3ja ($CDCl_3$, 126 MHz)



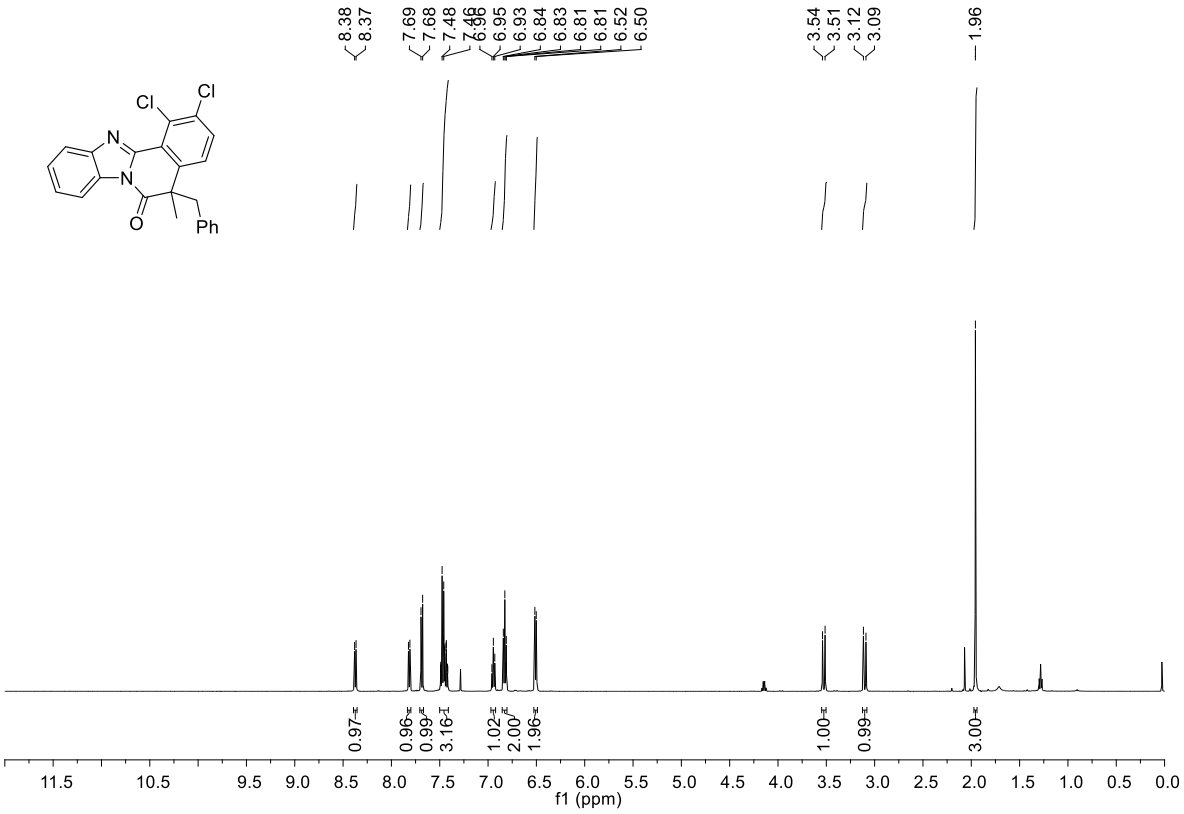
^{1}H NMR spectrum of 3ka ($CDCl_3$, 500 MHz)



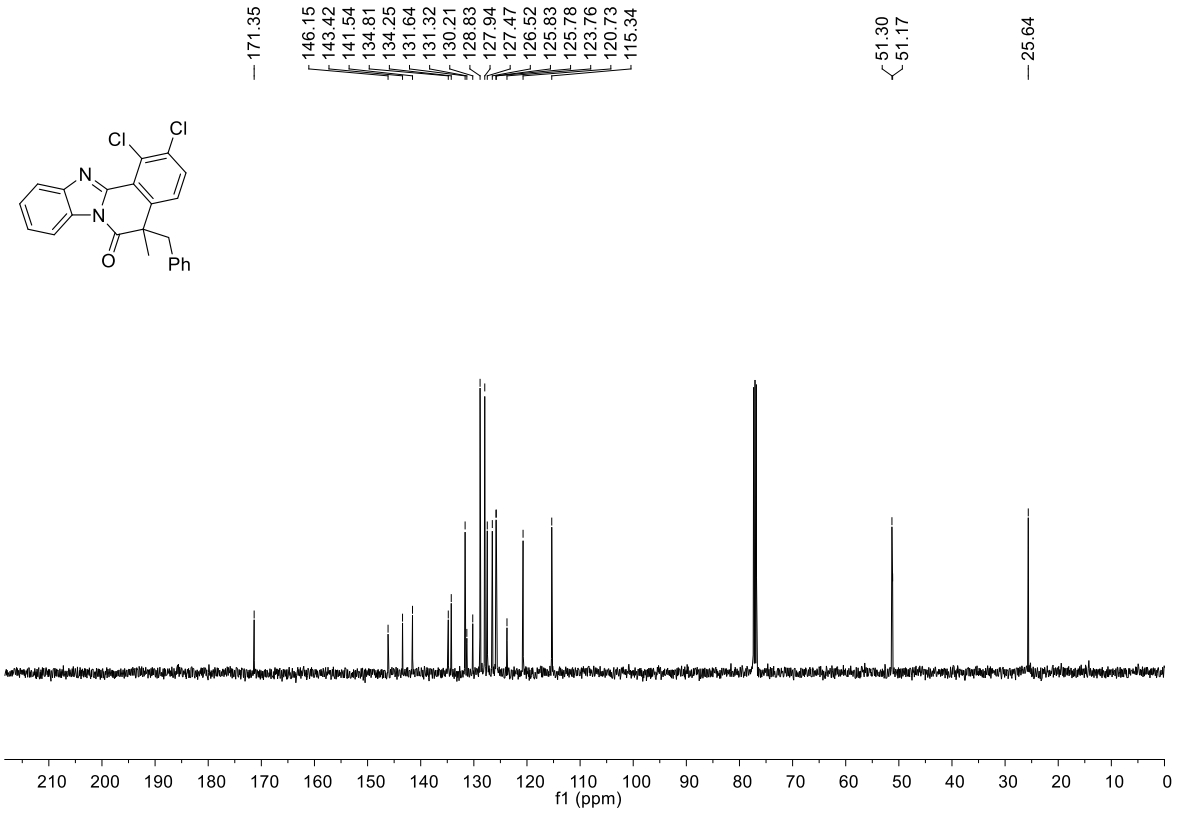
^{13}C{^{1}H} NMR spectrum of 3ka ($CDCl_3$, 126 MHz)



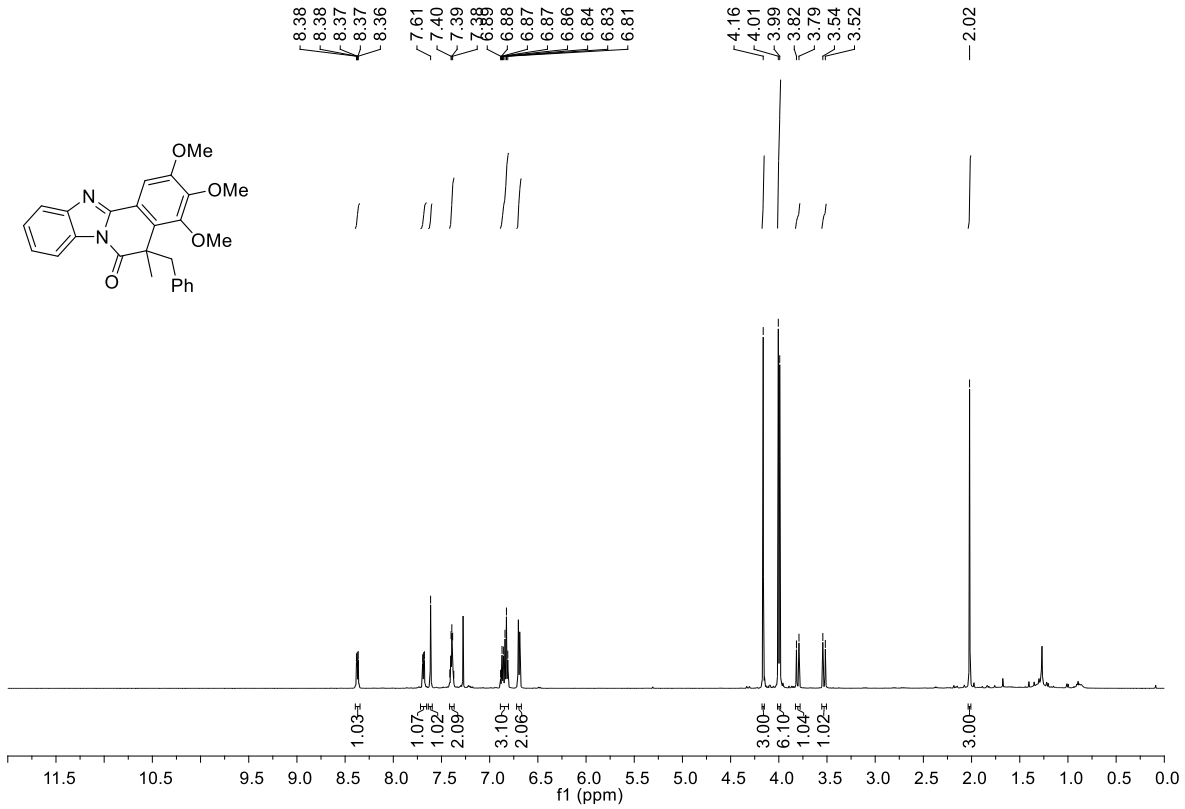
^{1}H NMR spectrum of 3la ($CDCl_3$, 500 MHz)



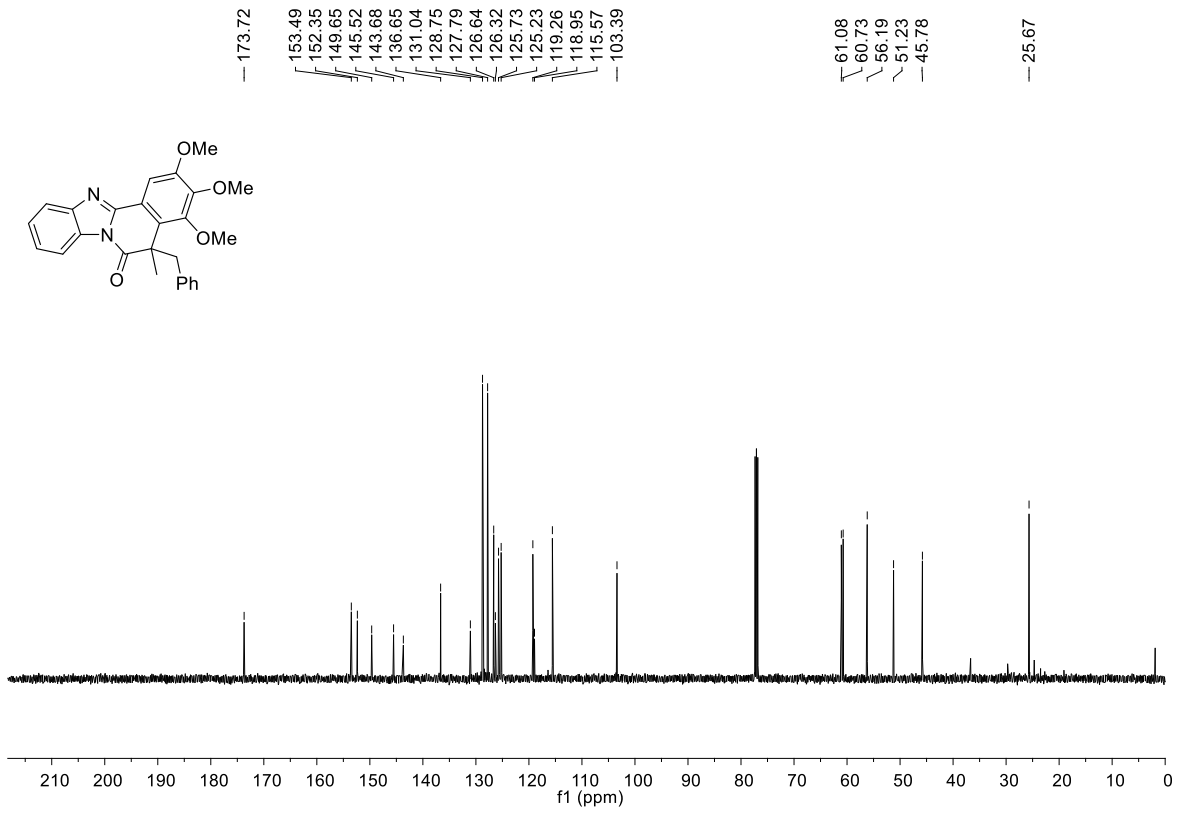
^{13}C{^{1}H} NMR spectrum of 3la ($CDCl_3$, 126 MHz)



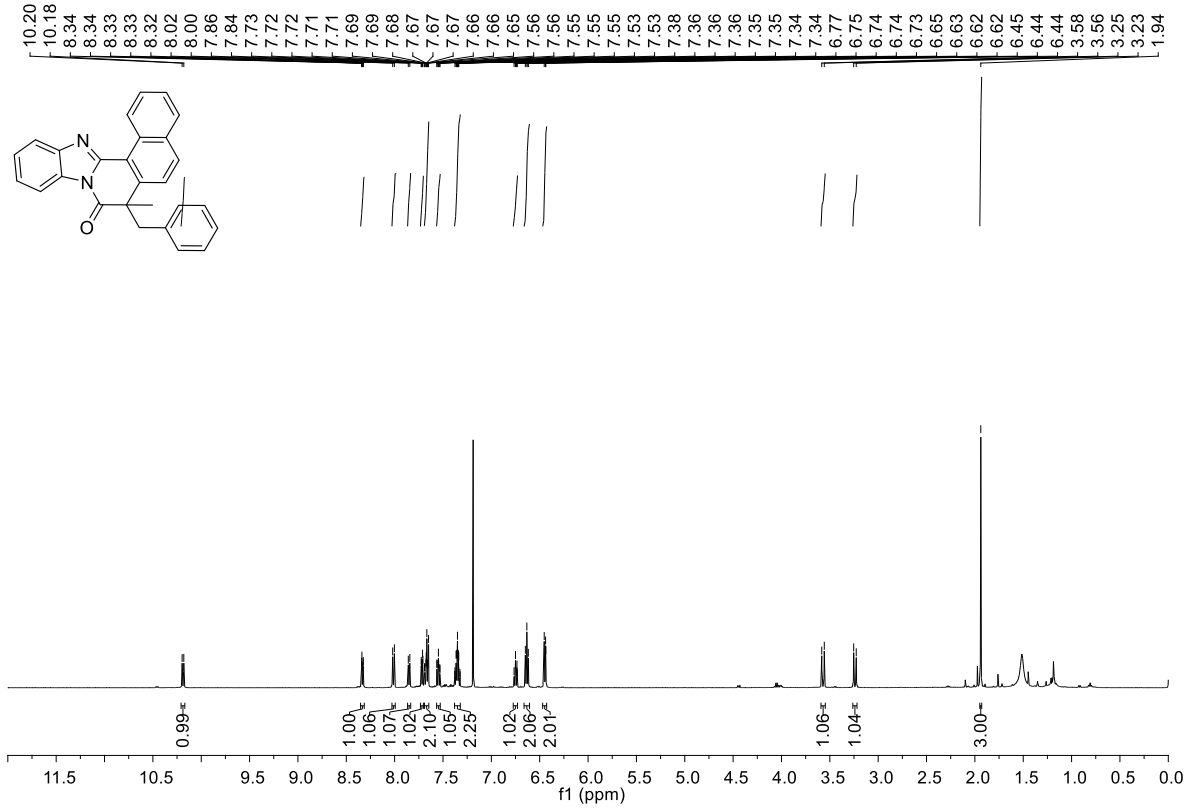
^{1}H NMR spectrum of 3ma ($CDCl_3$, 500 MHz)



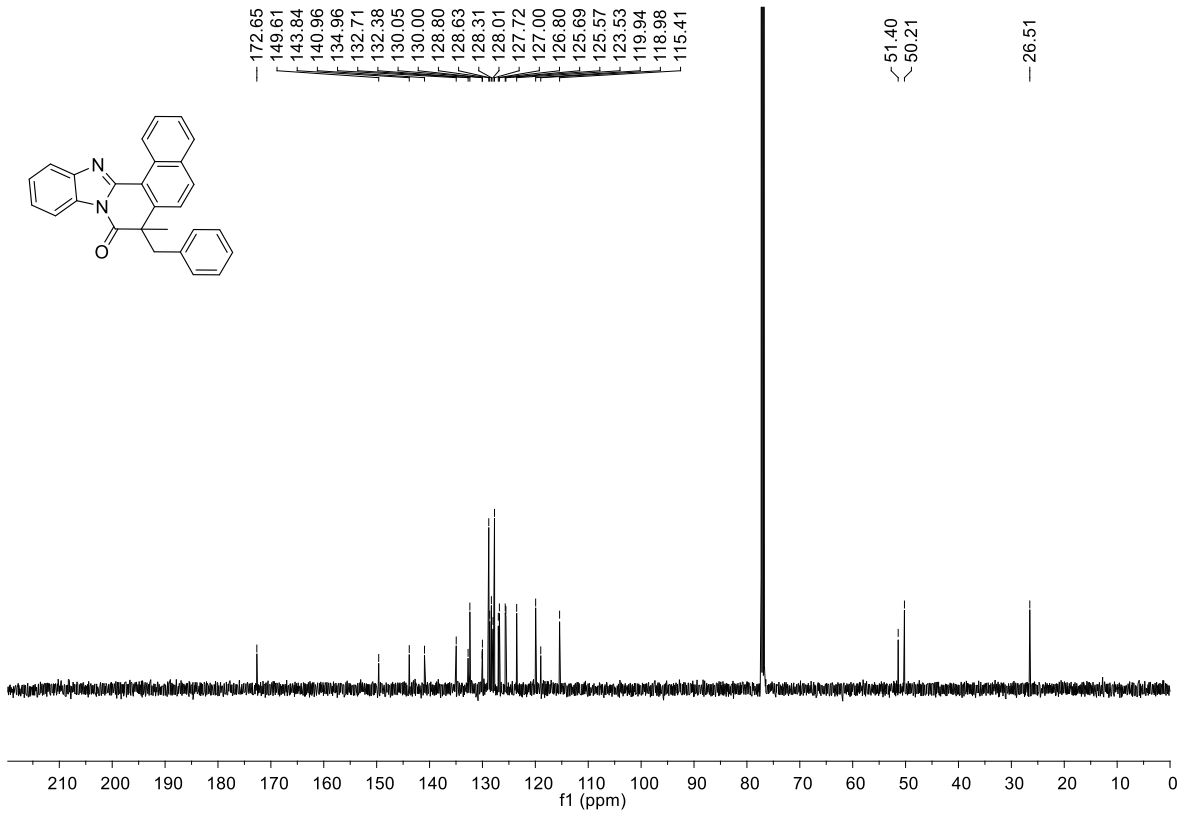
^{13}C{^{1}H} NMR spectrum of 3ma ($CDCl_3$, 126 MHz)



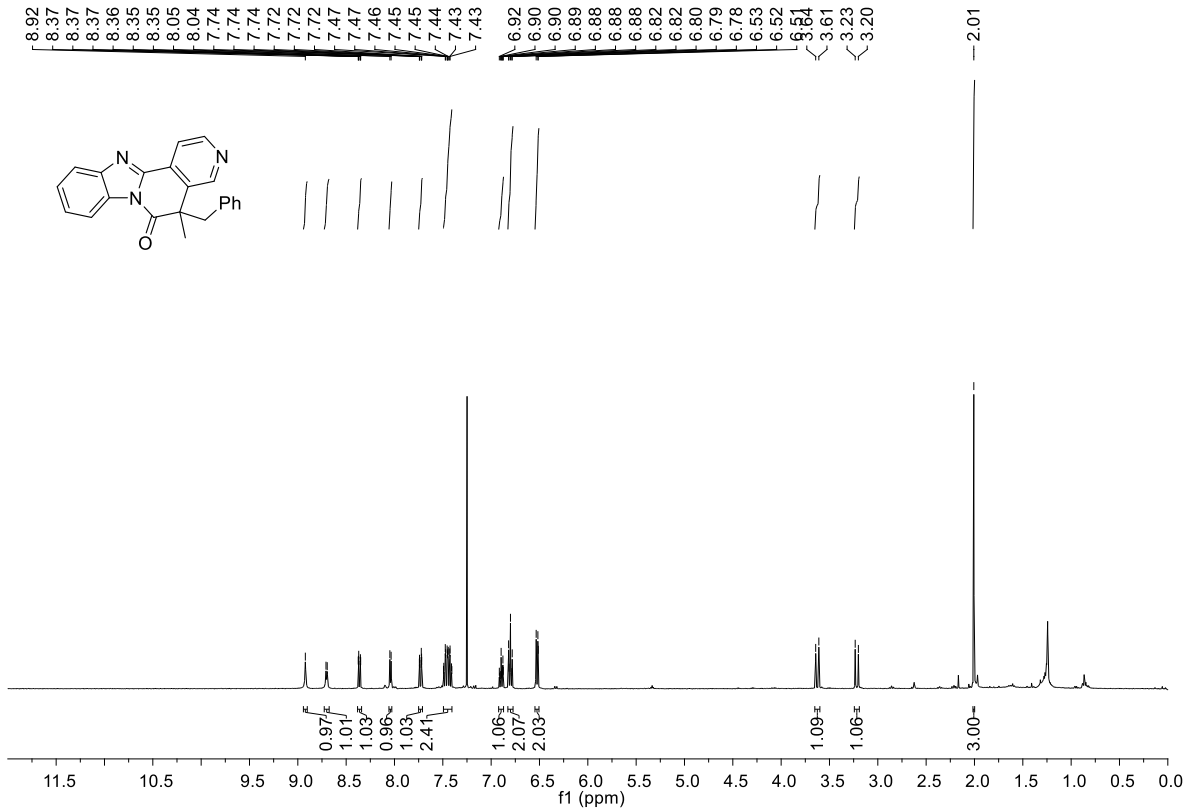
^{1}H NMR spectrum of 3na ($CDCl_3$, 500 MHz)



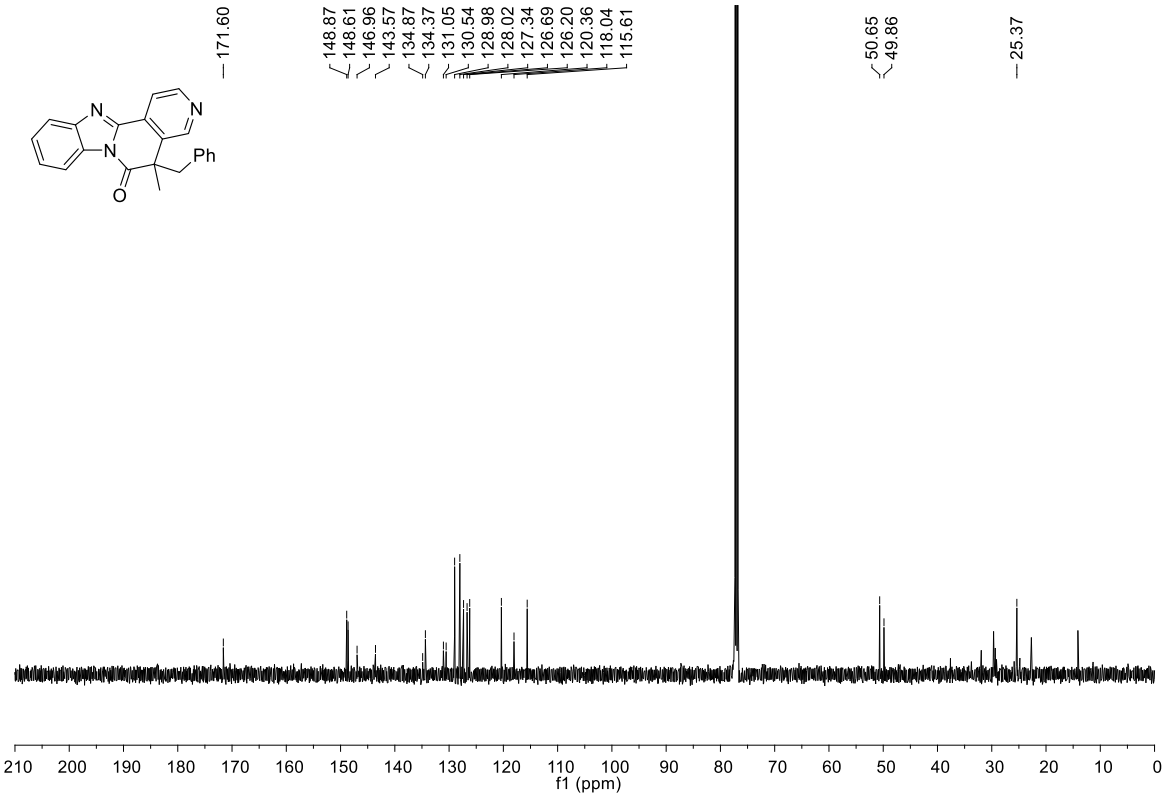
^{13}C{^{1}H} NMR spectrum of 3na ($CDCl_3$, 126 MHz)



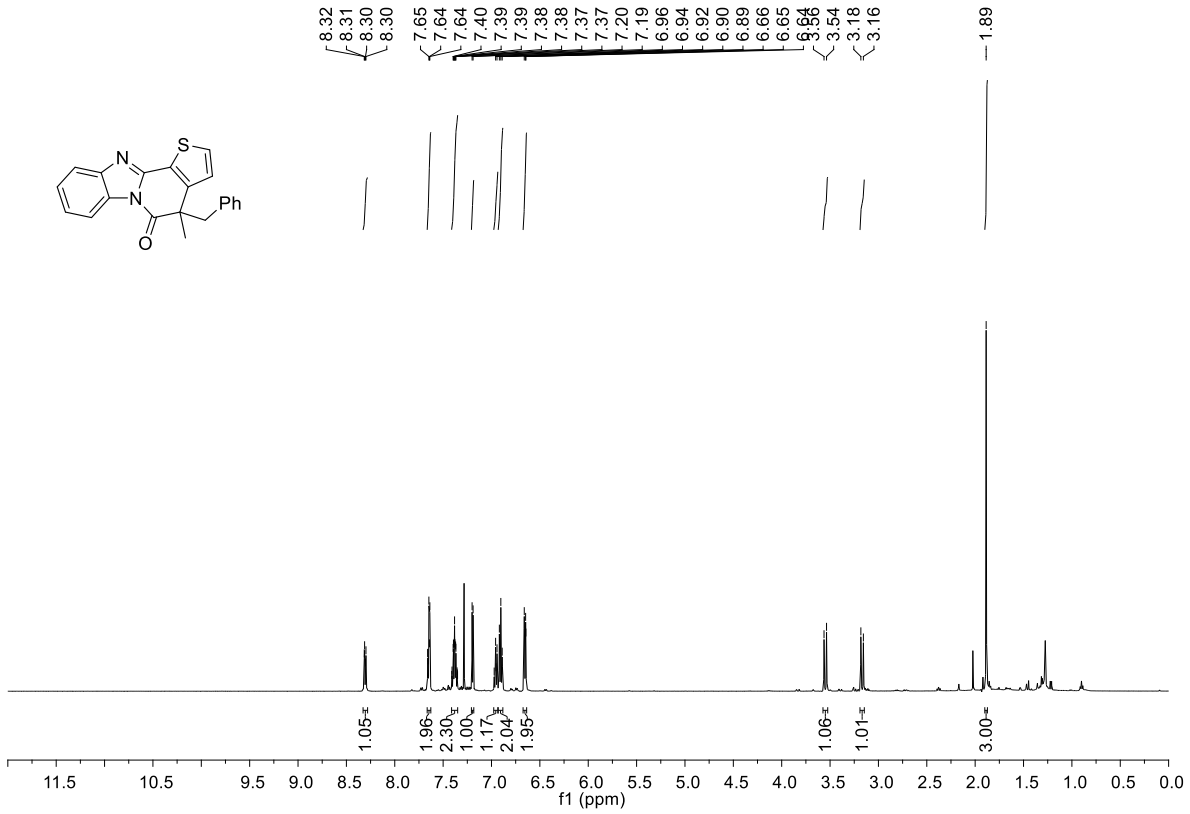
^{1}H NMR spectrum of 3oa ($CDCl_3$, 400 MHz)



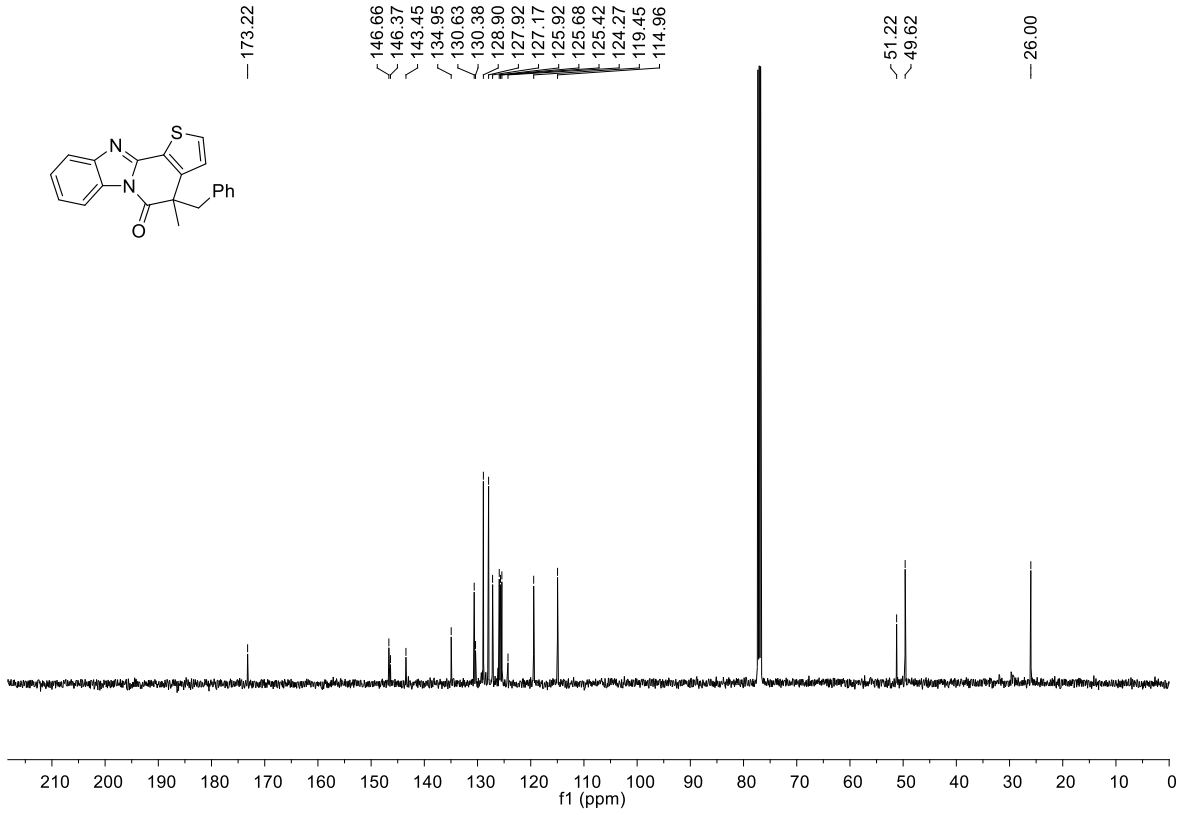
^{13}C{^{1}H} NMR spectrum of 3oa ($CDCl_3$, 126 MHz)



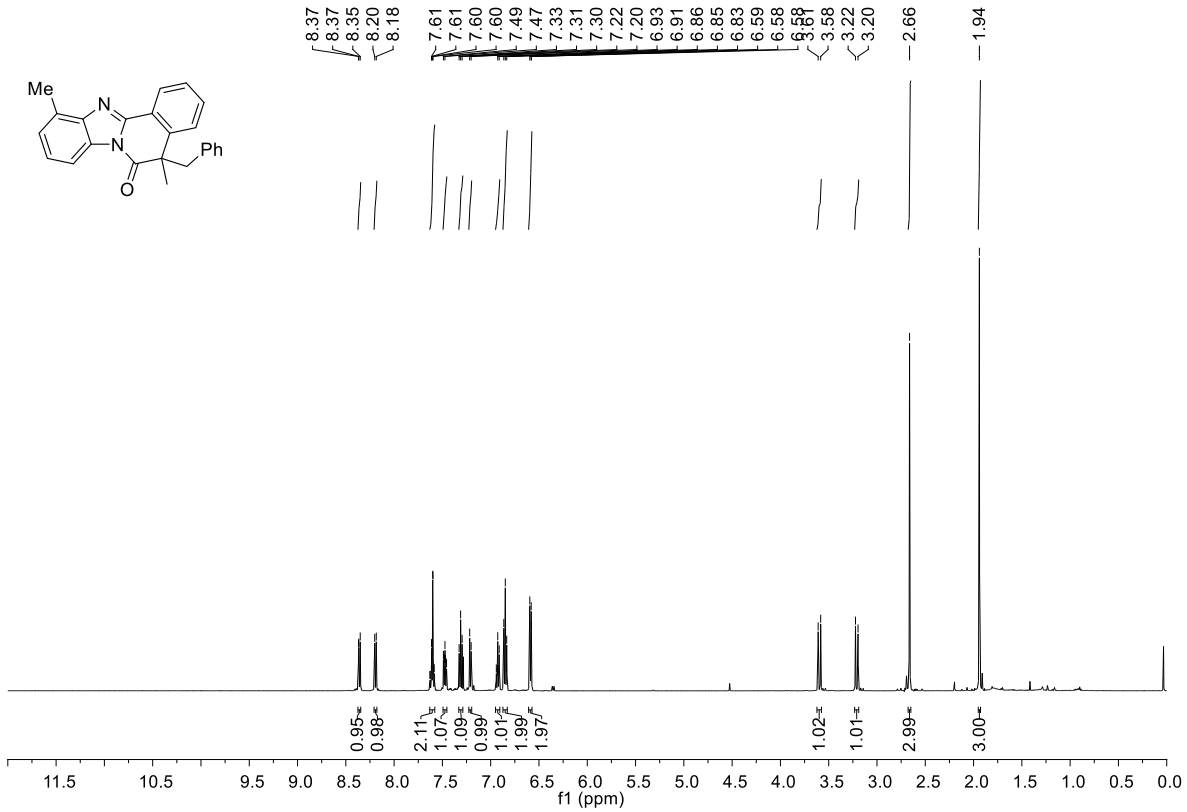
^{1}H NMR spectrum of 3pa ($CDCl_3$, 500 MHz)



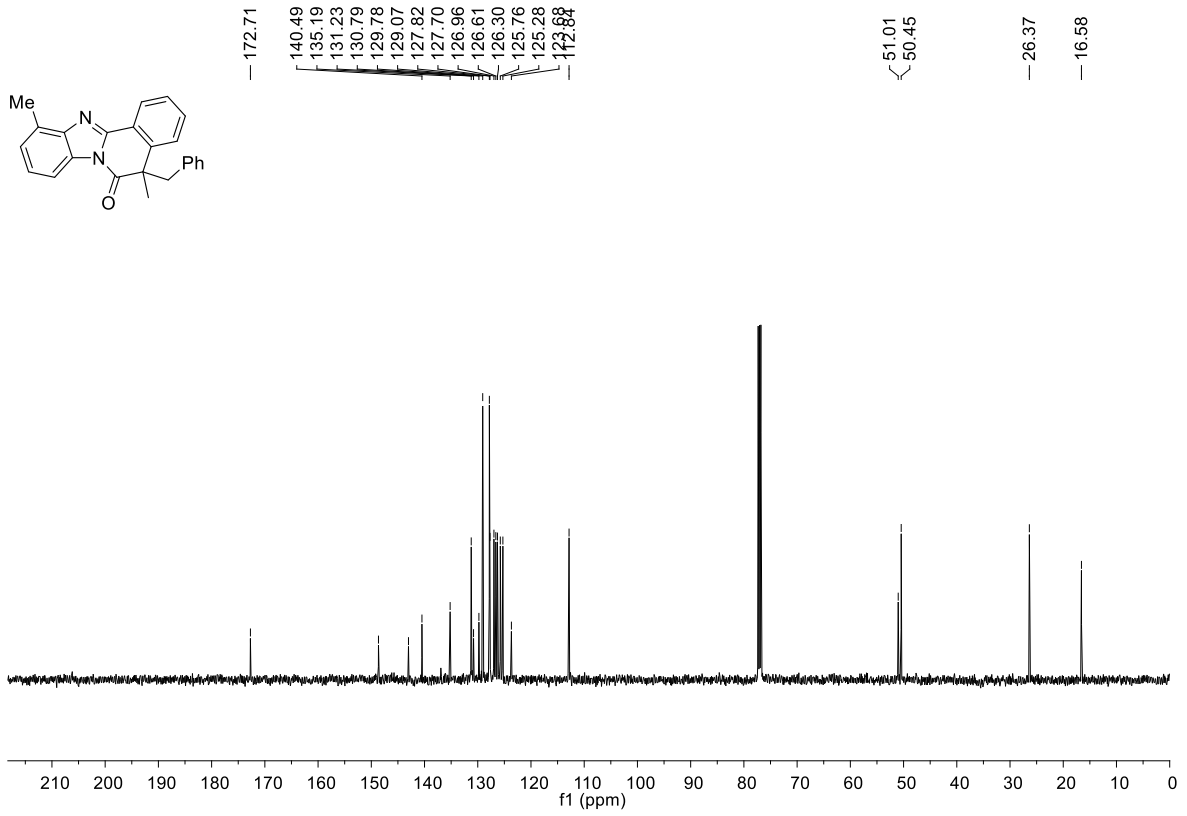
^{13}C{^{1}H} NMR spectrum of 3pa ($CDCl_3$, 126 MHz)



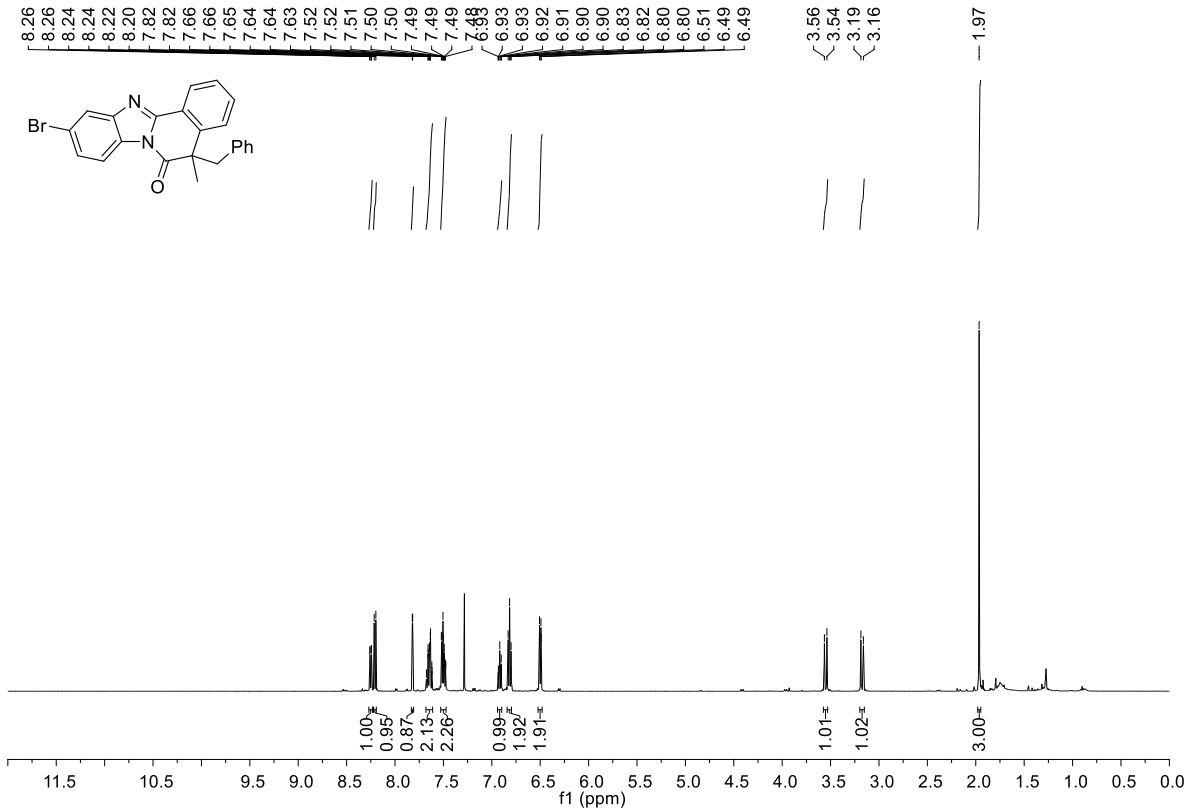
^{1}H NMR spectrum of 3qa ($CDCl_3$, 500 MHz)



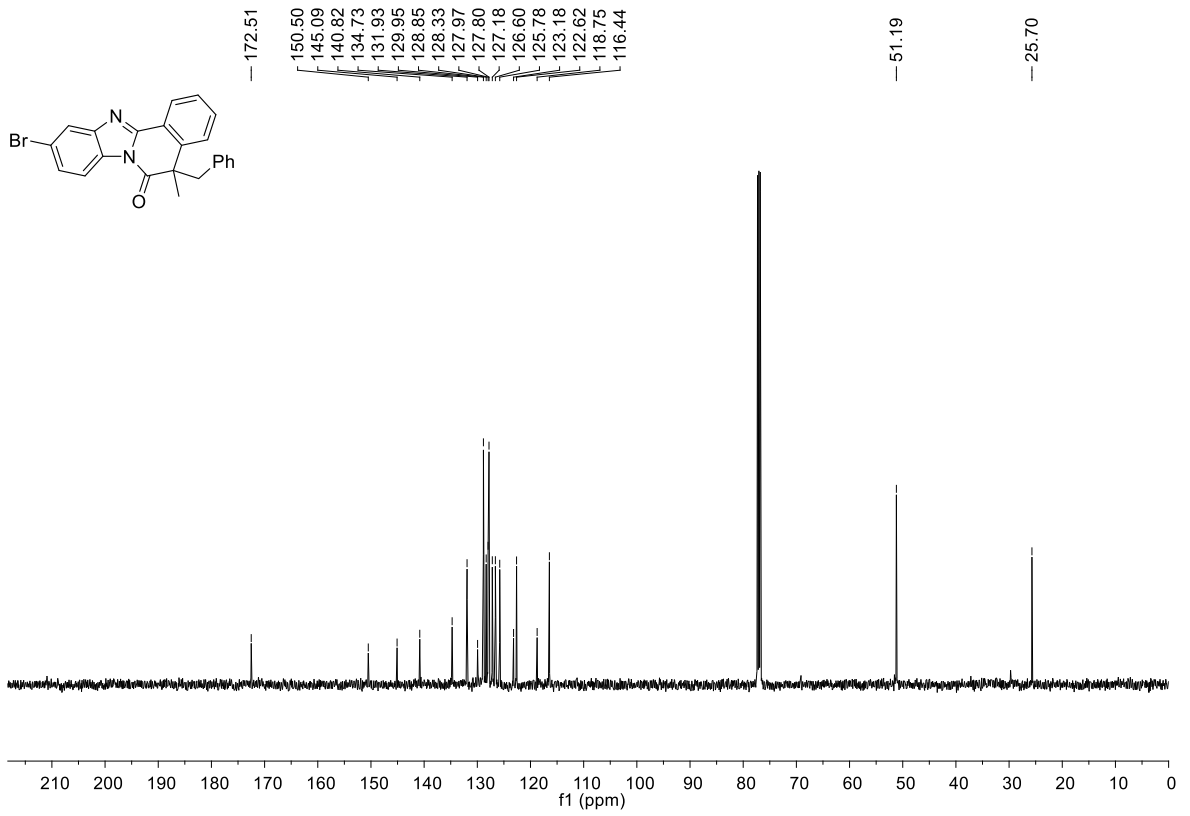
^{13}C{^{1}H} NMR spectrum of 3qa ($CDCl_3$, 126 MHz)



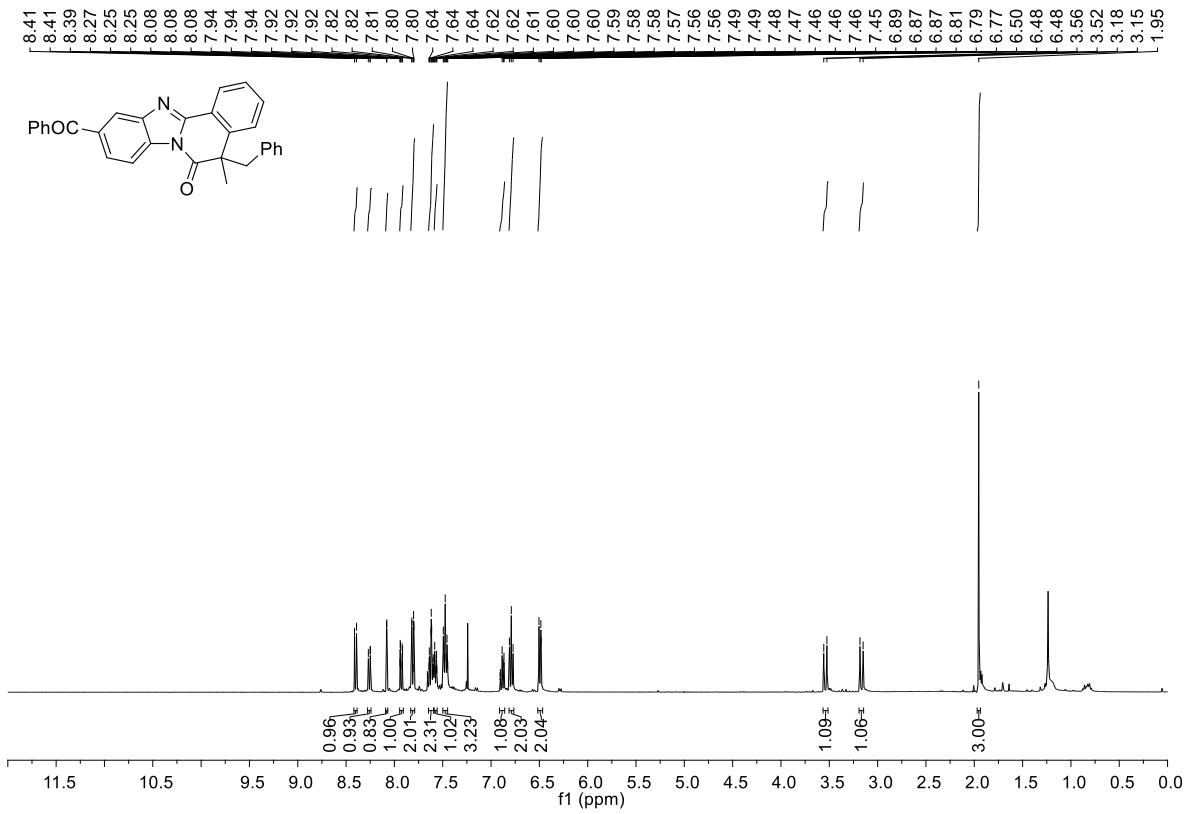
^{1}H NMR spectrum of 3ra ($CDCl_3$, 500 MHz)



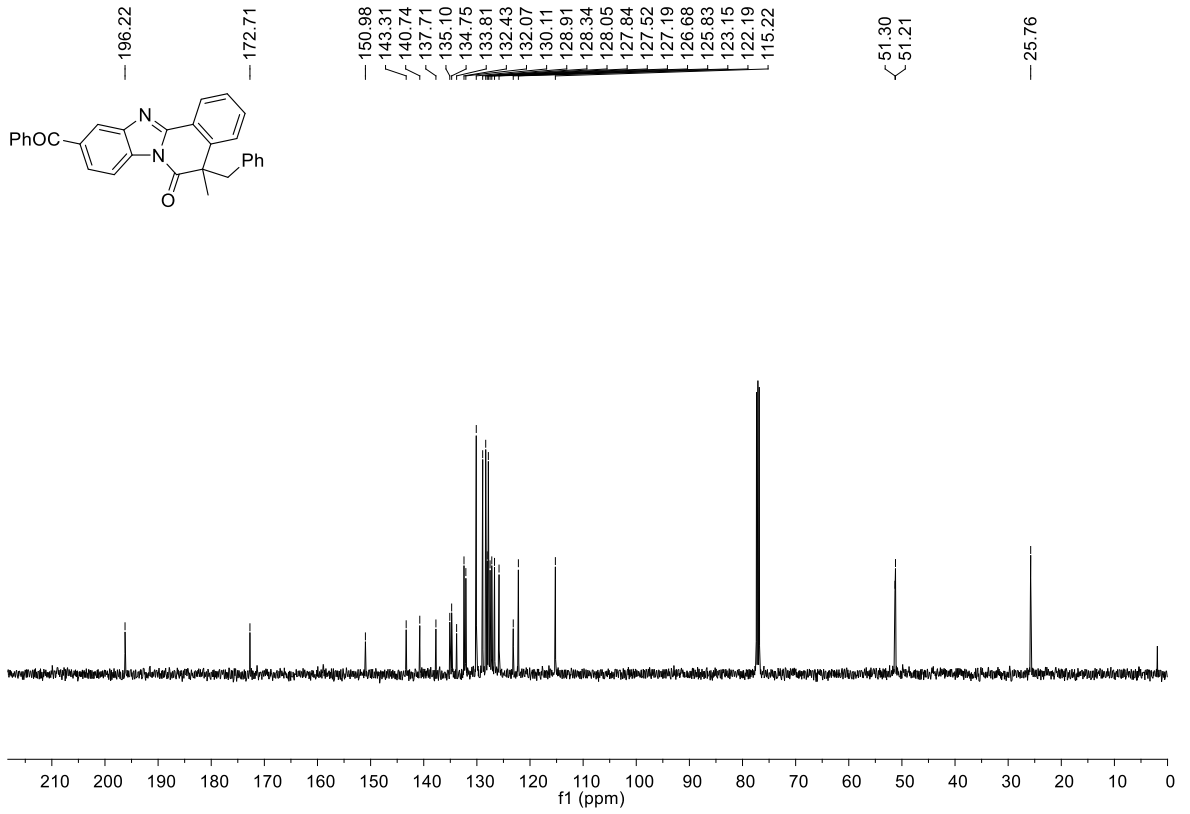
^{13}C{^{1}H} NMR spectrum of 3ra ($CDCl_3$, 126 MHz)



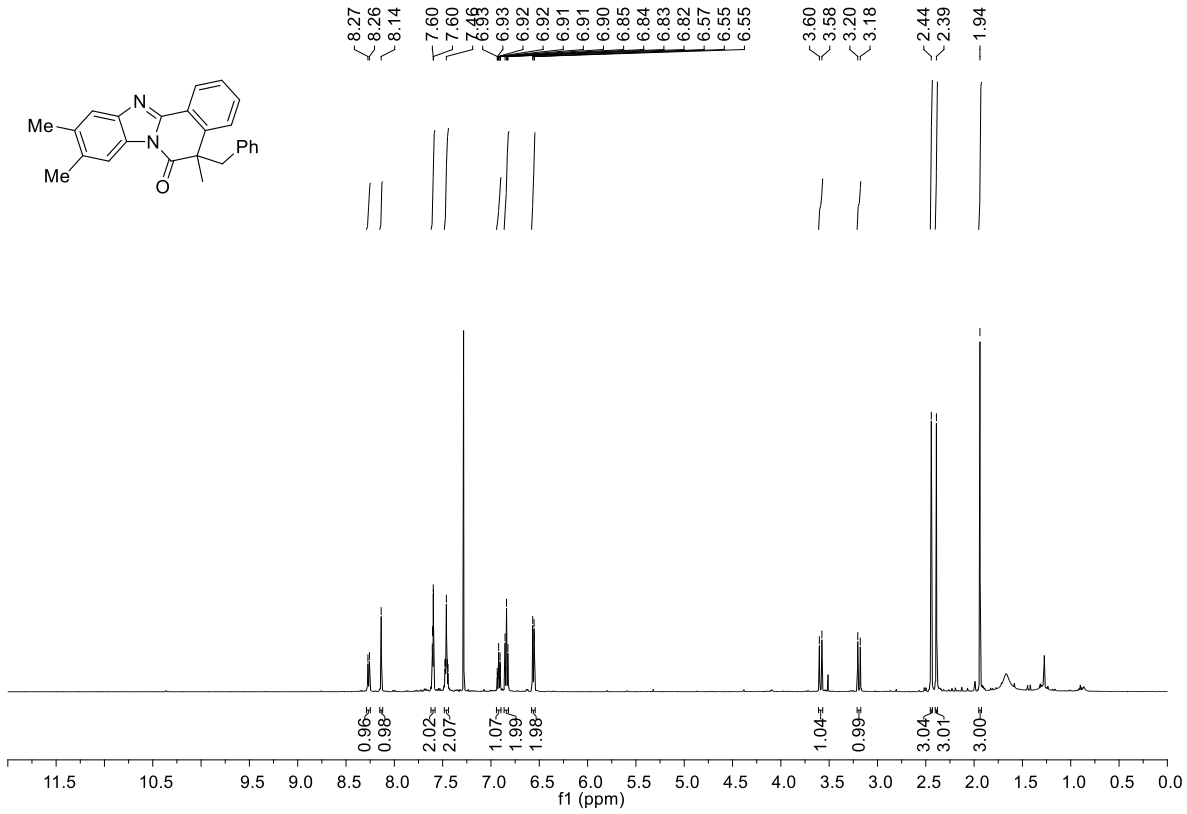
^{1}H NMR spectrum of 3sa ($CDCl_3$, 400 MHz)



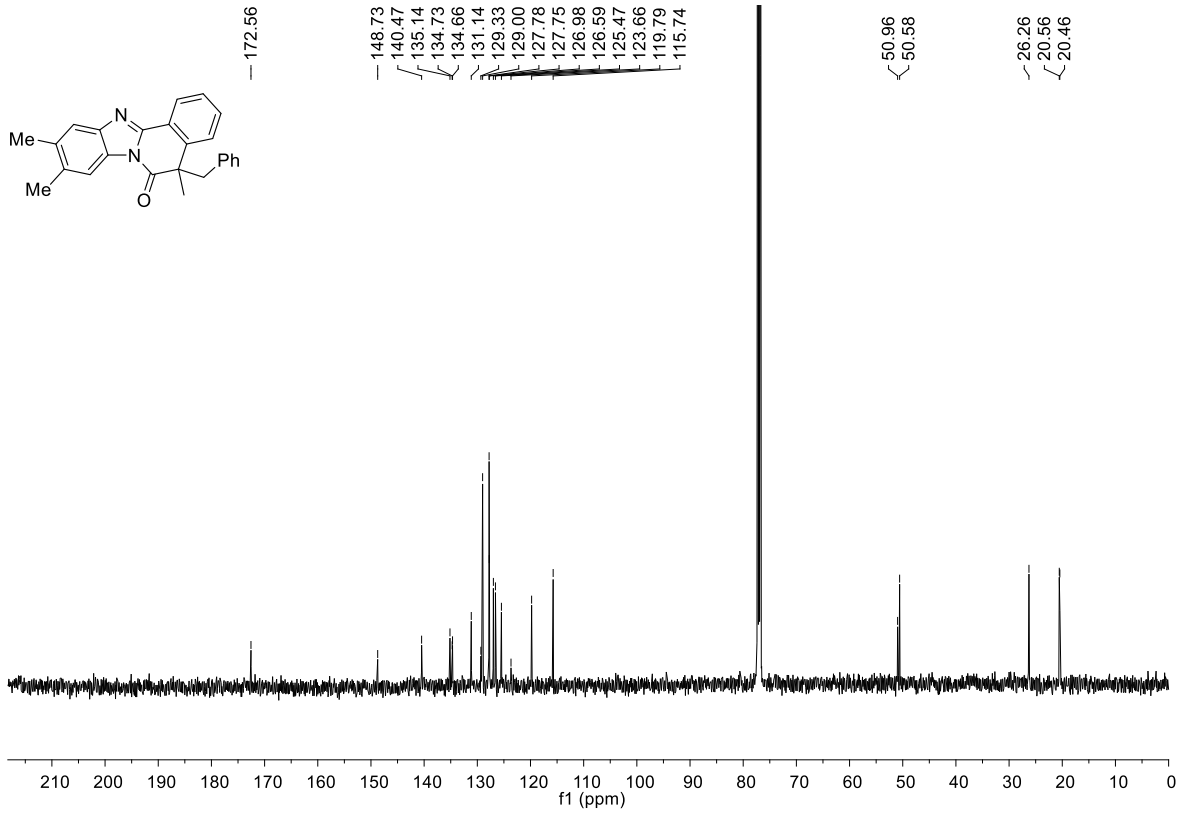
^{13}C{^{1}H} NMR spectrum of 3sa ($CDCl_3$, 126 MHz)



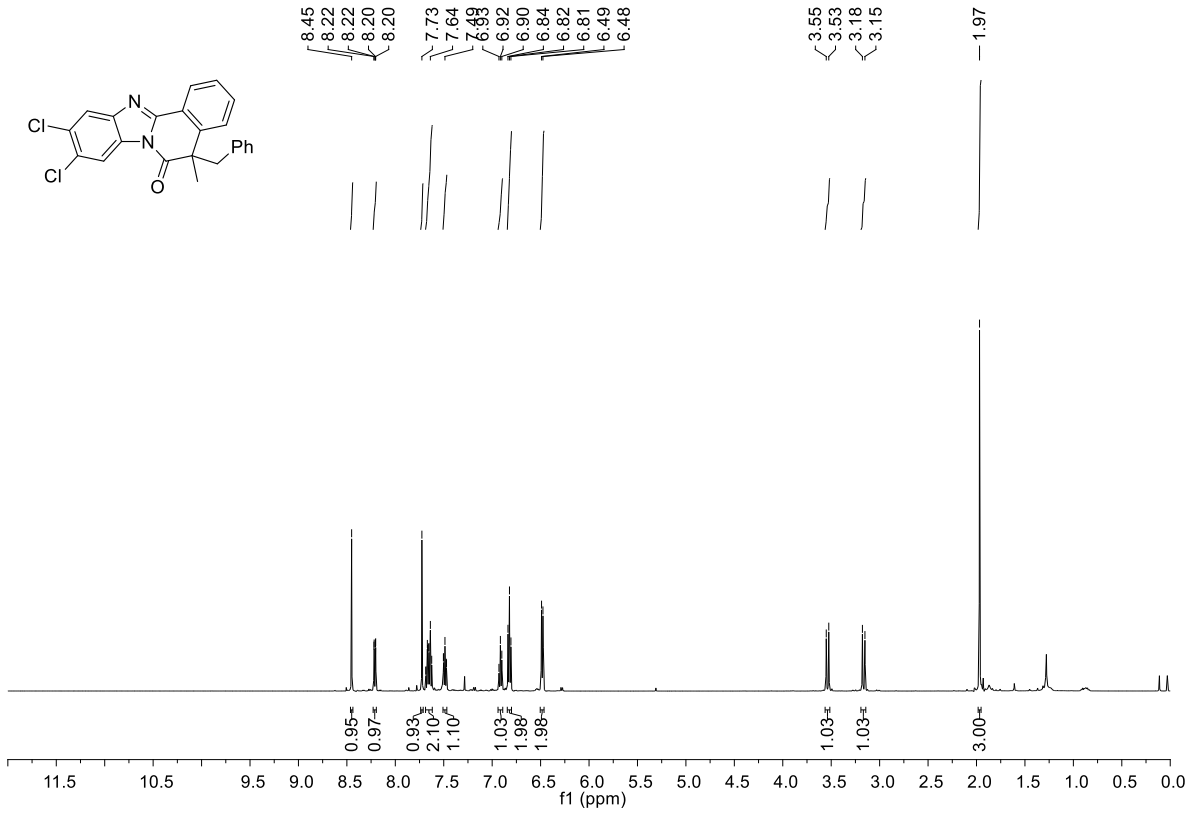
^{1}H NMR spectrum of 3ta ($CDCl_3$, 500 MHz)



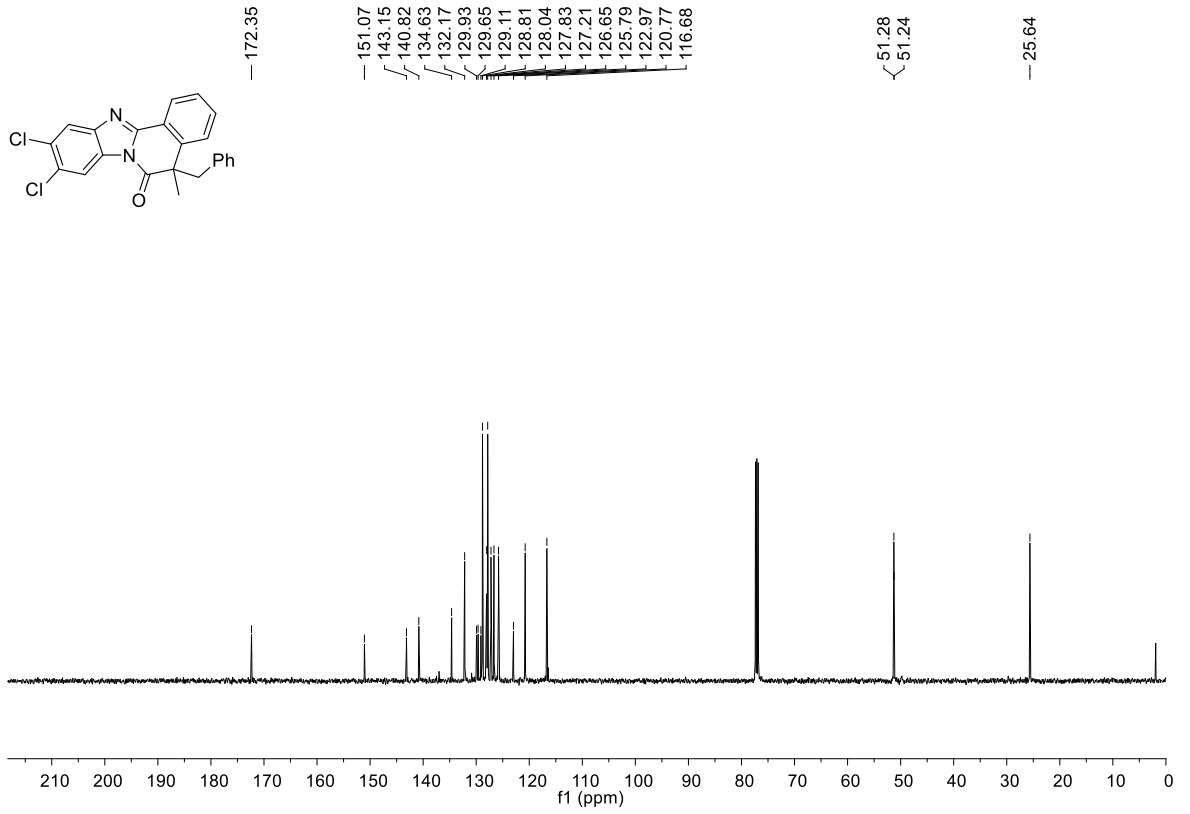
^{13}C{^{1}H} NMR spectrum of 3ta ($CDCl_3$, 126 MHz)



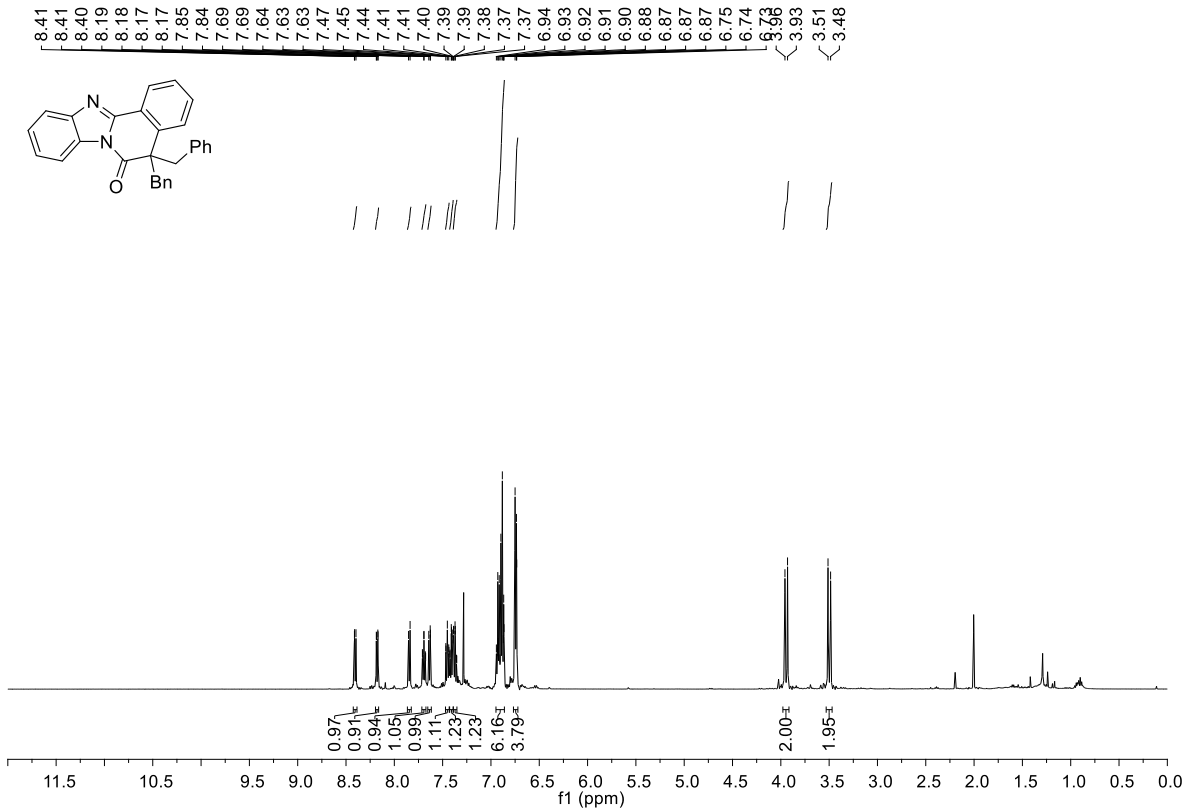
^{1}H NMR spectrum of 3ua ($CDCl_3$, 500 MHz)



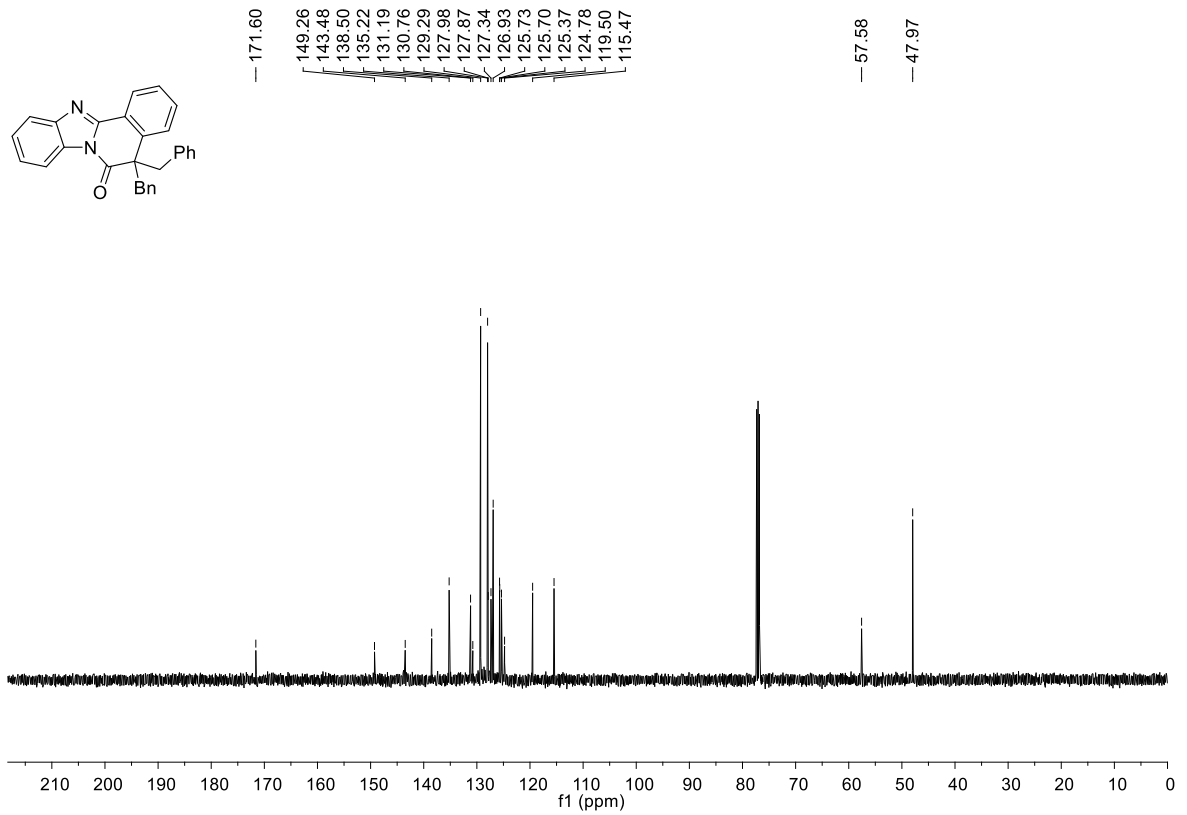
^{13}C{^{1}H} NMR spectrum of 3ua ($CDCl_3$, 126 MHz)



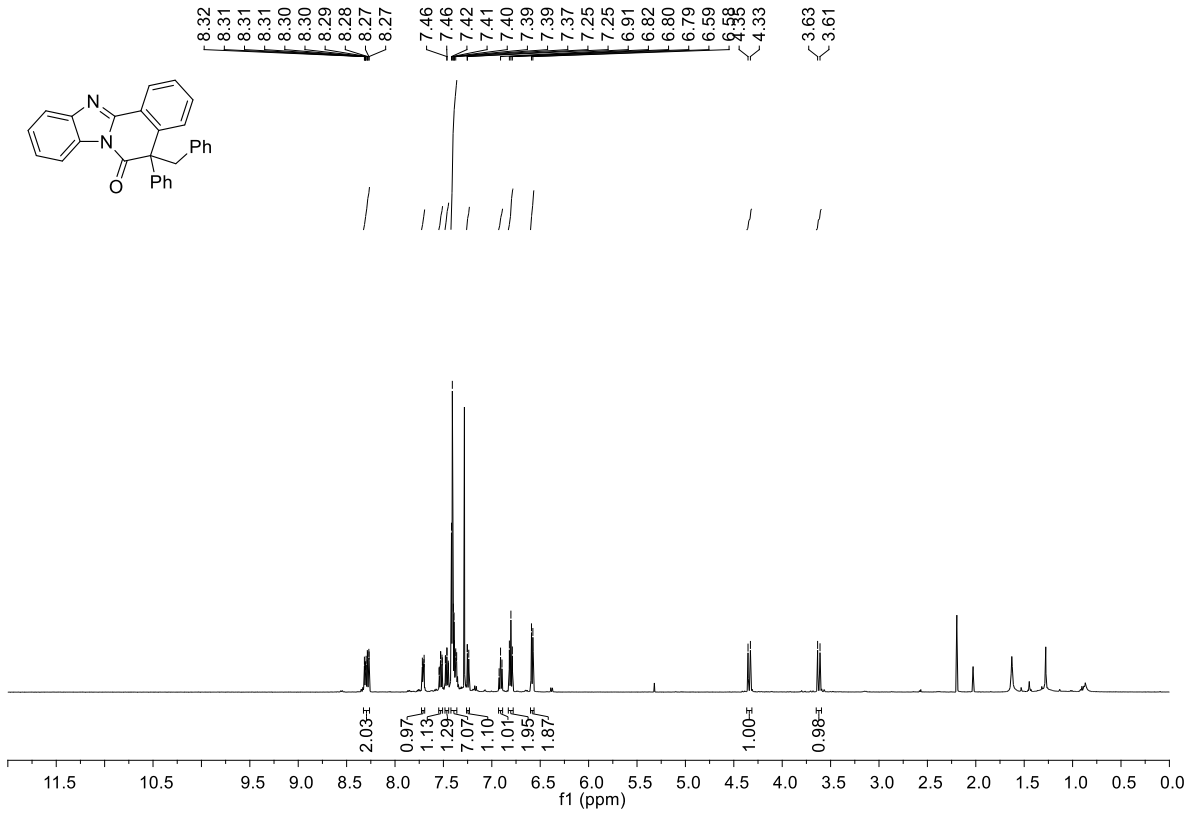
^{1}H NMR spectrum of 3va ($CDCl_3$, 500 MHz)



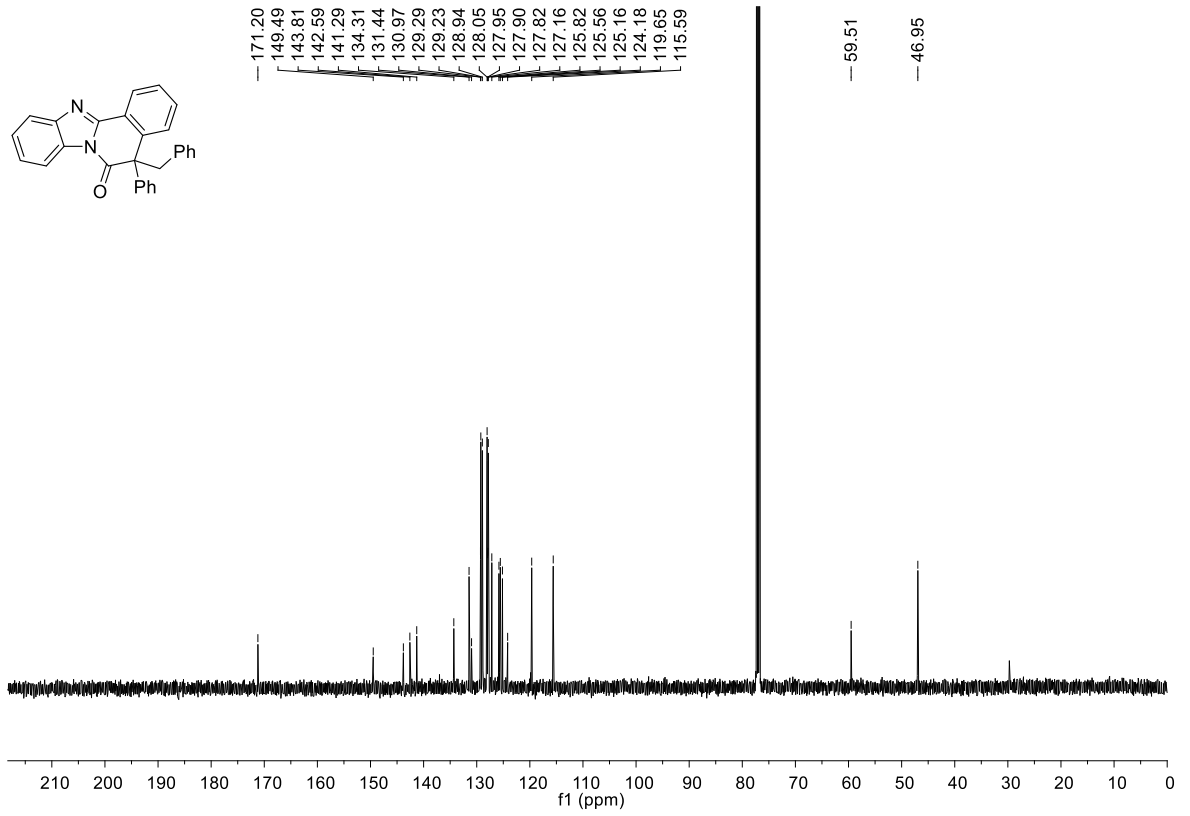
^{13}C{^{1}H} NMR spectrum of 3va ($CDCl_3$, 126 MHz)



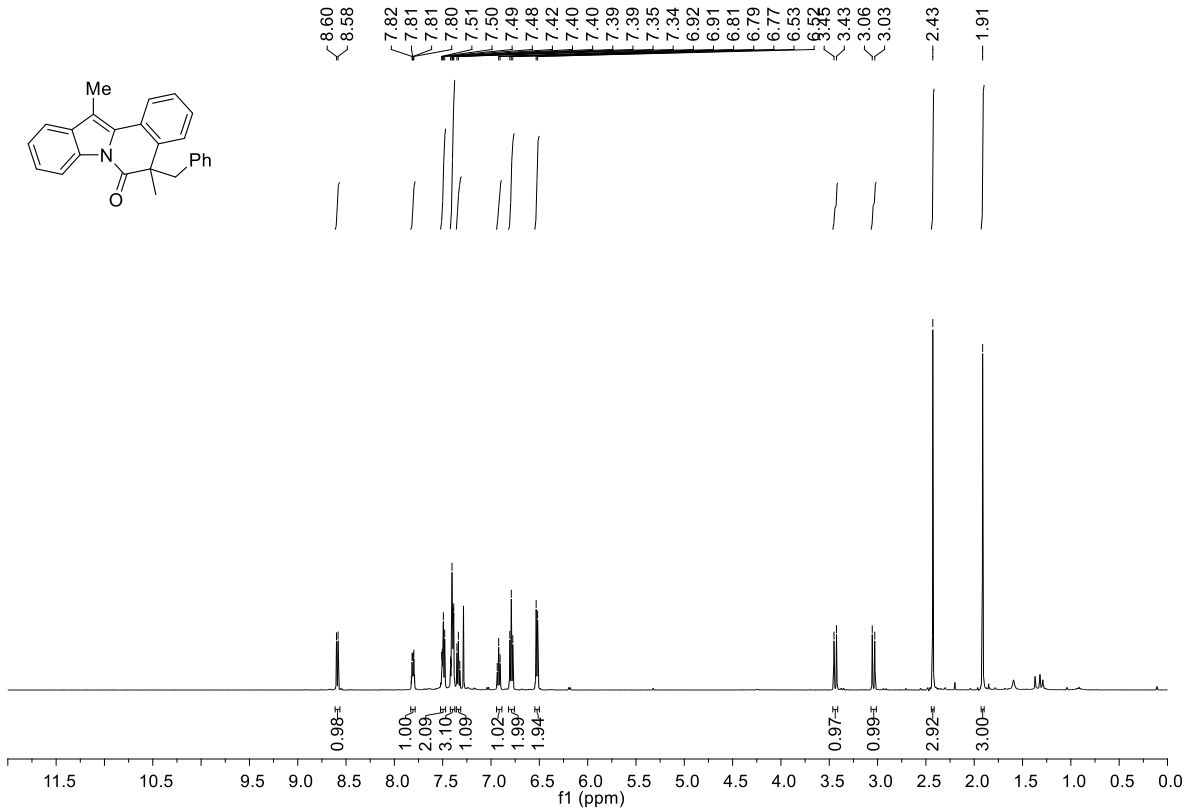
^{1}H NMR spectrum of 3wa ($CDCl_3$, 500 MHz)



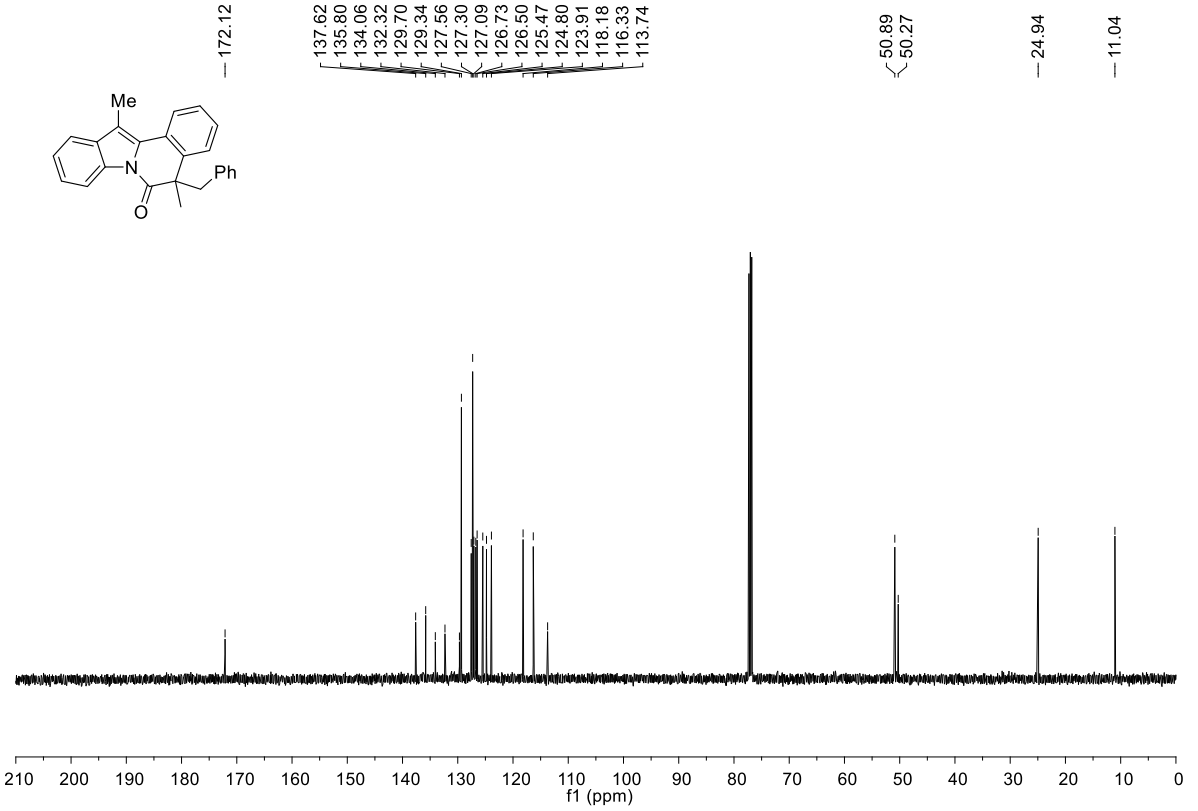
^{13}C{^{1}H} NMR spectrum of 3wa ($CDCl_3$, 126 MHz)



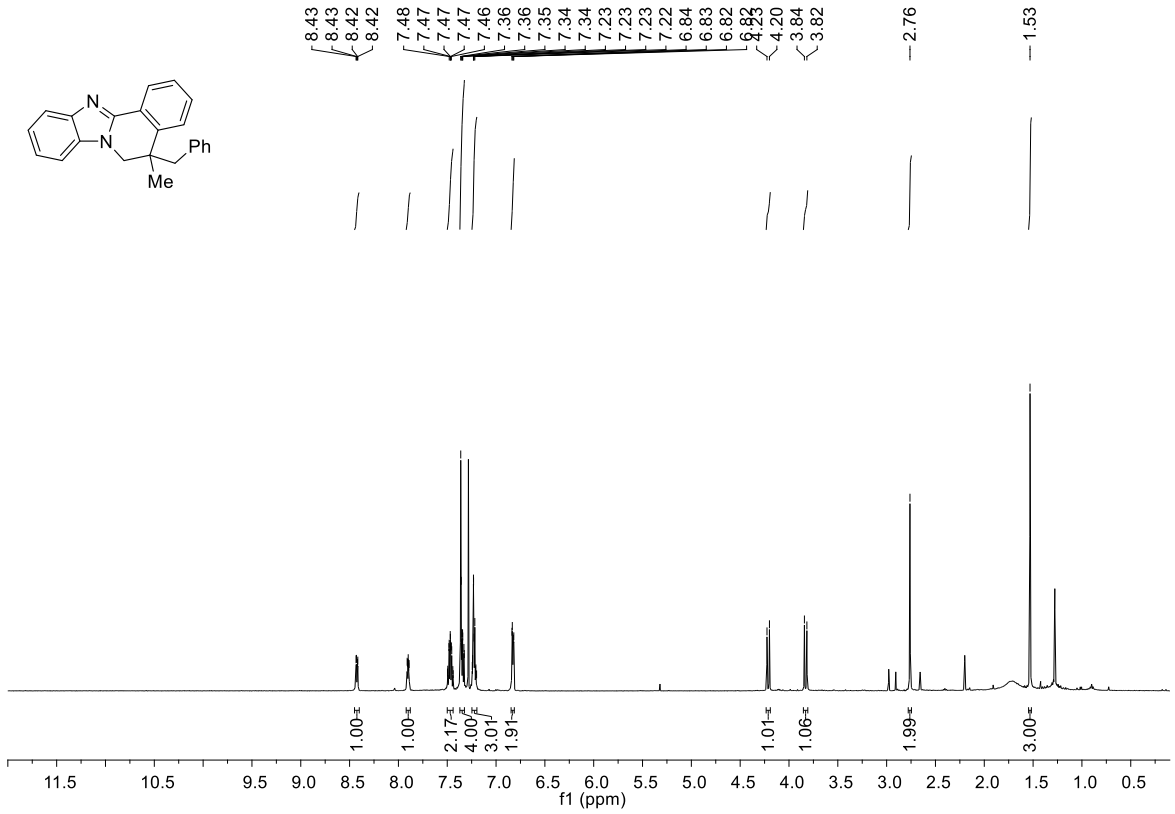
^{1}H NMR spectrum of 3xa ($CDCl_3$, 500 MHz)



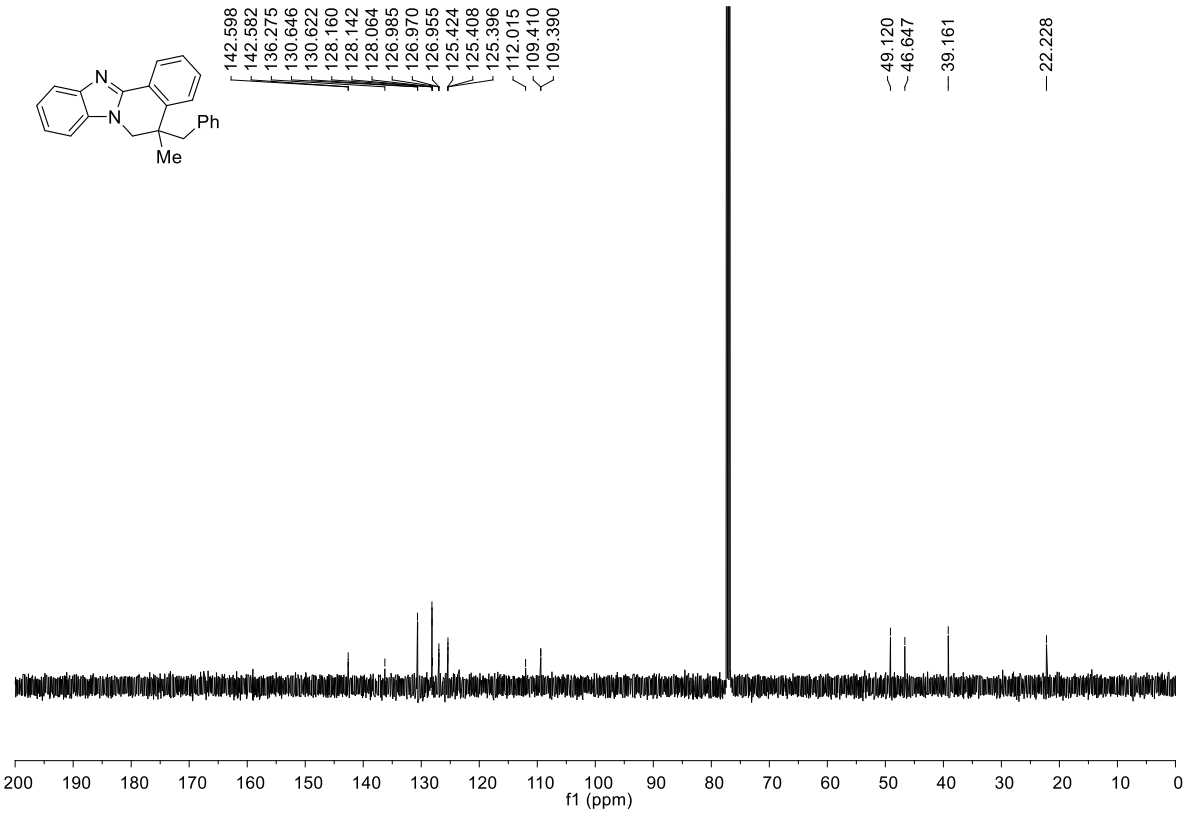
^{13}C{^{1}H} NMR spectrum of 3xa ($CDCl_3$, 126 MHz)



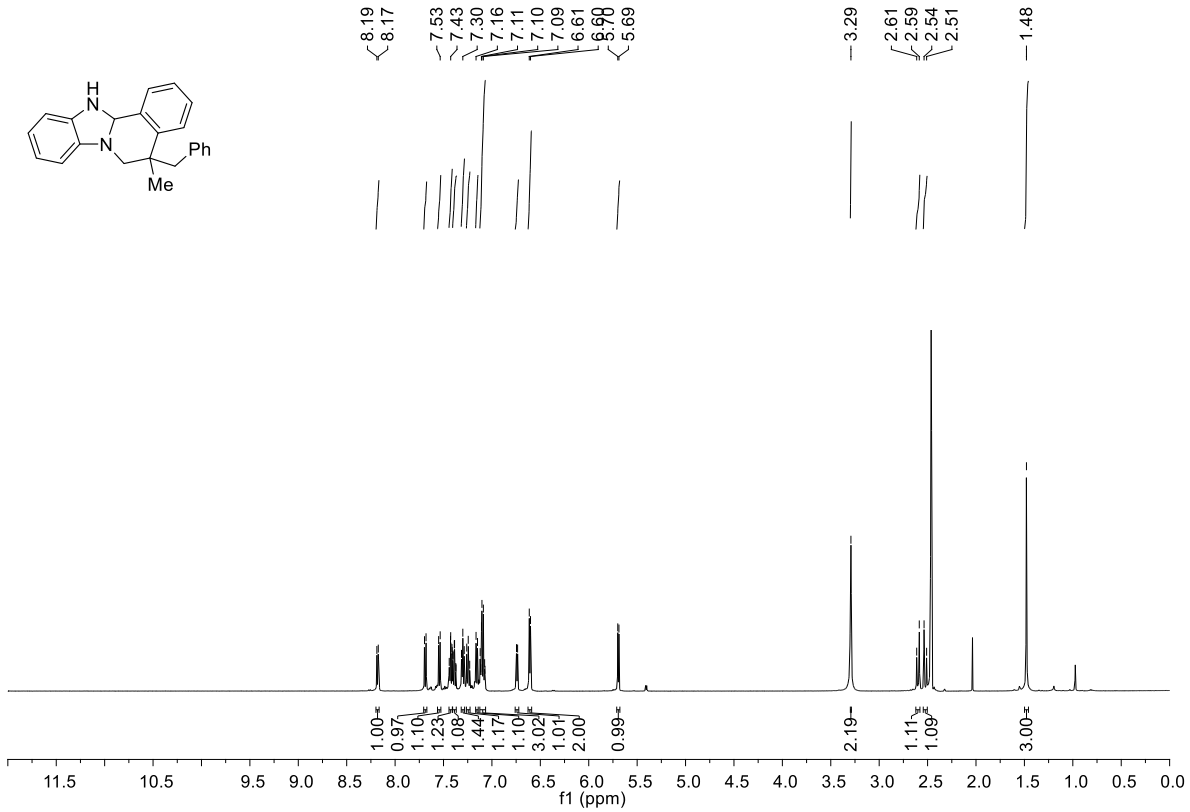
^{1}H NMR spectrum of 5 ($CDCl_3$, 500 MHz)



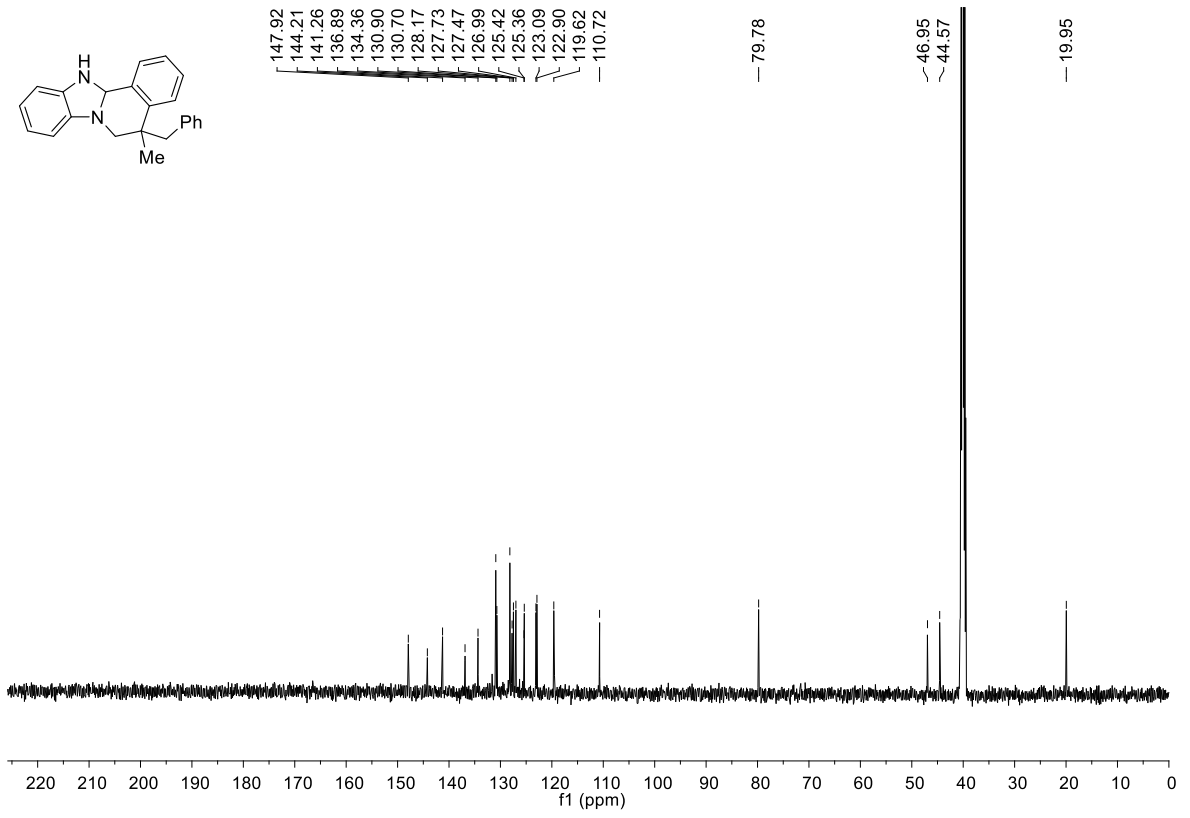
^{13}C{^{1}H} NMR spectrum of 5 ($CDCl_3$, 126 MHz)



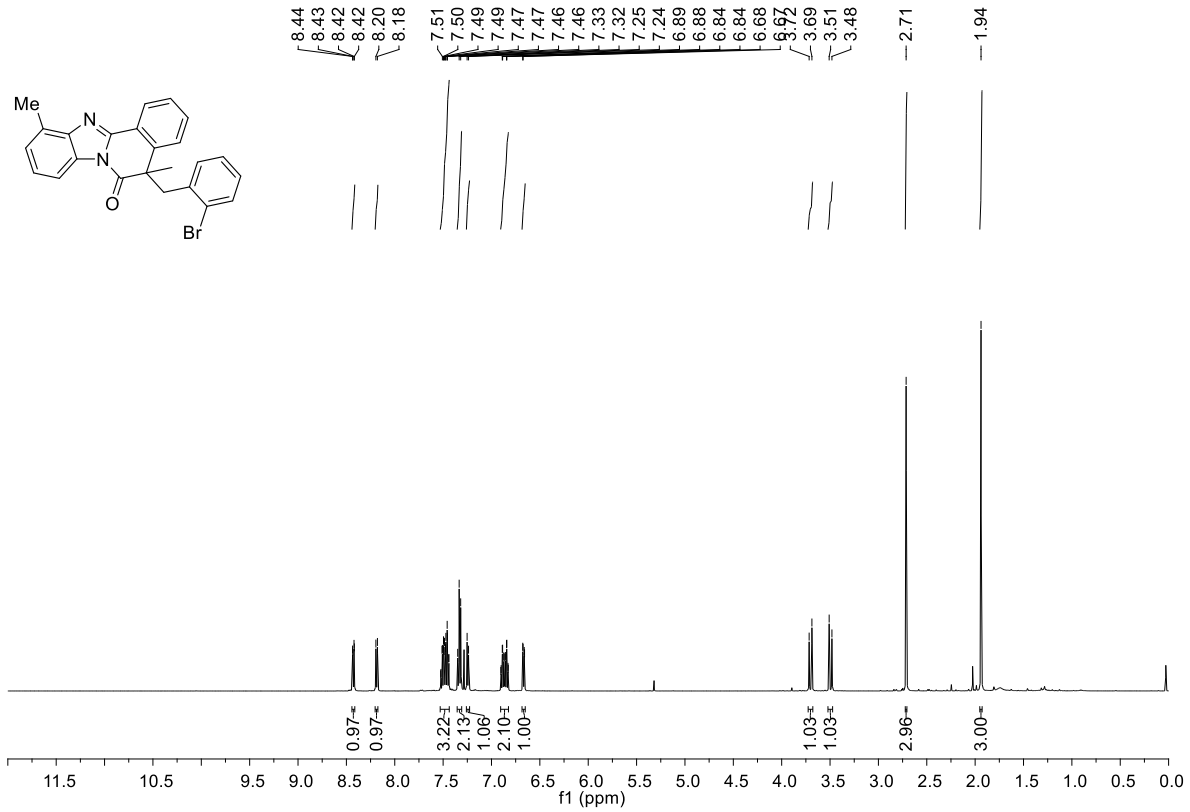
^{1}H NMR spectrum of 6 (DMSO, 400 MHz)



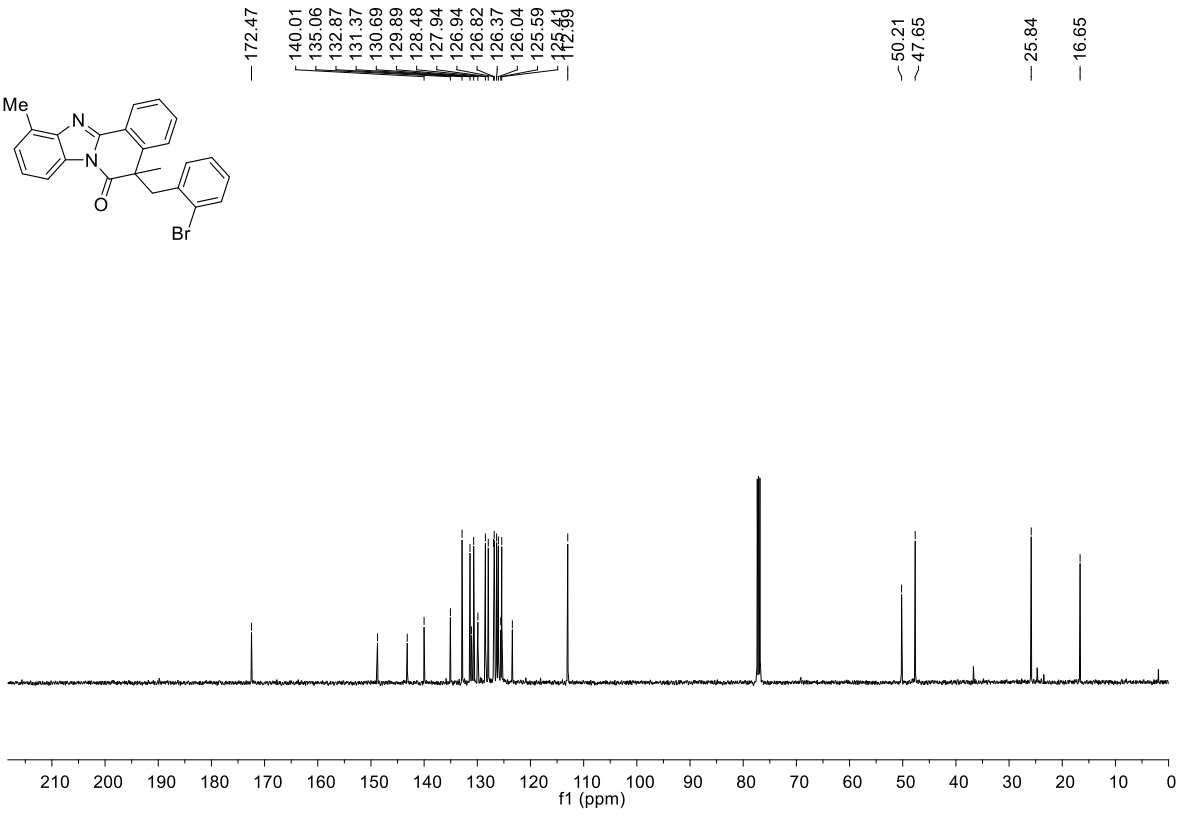
^{13}C{^{1}H} NMR spectrum of 6 (DMSO, 101 MHz)



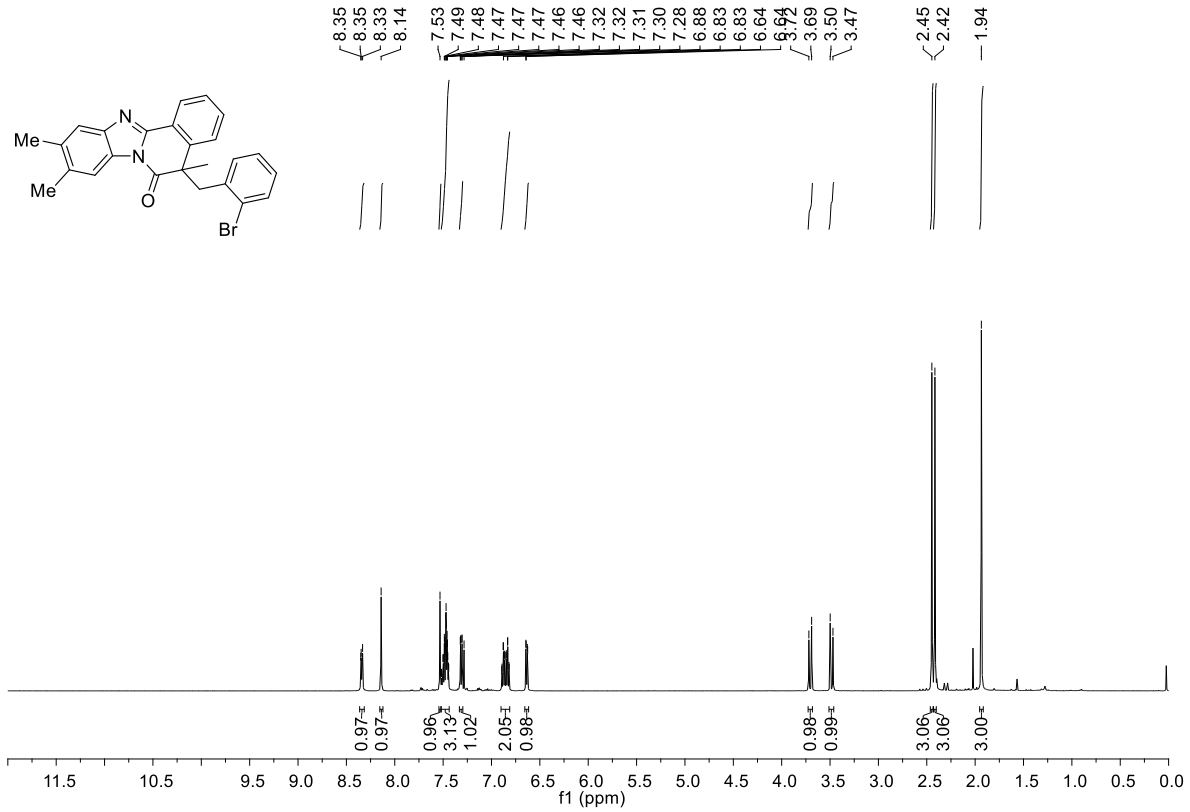
^{1}H NMR spectrum of 7b ($CDCl_3$, 500 MHz)



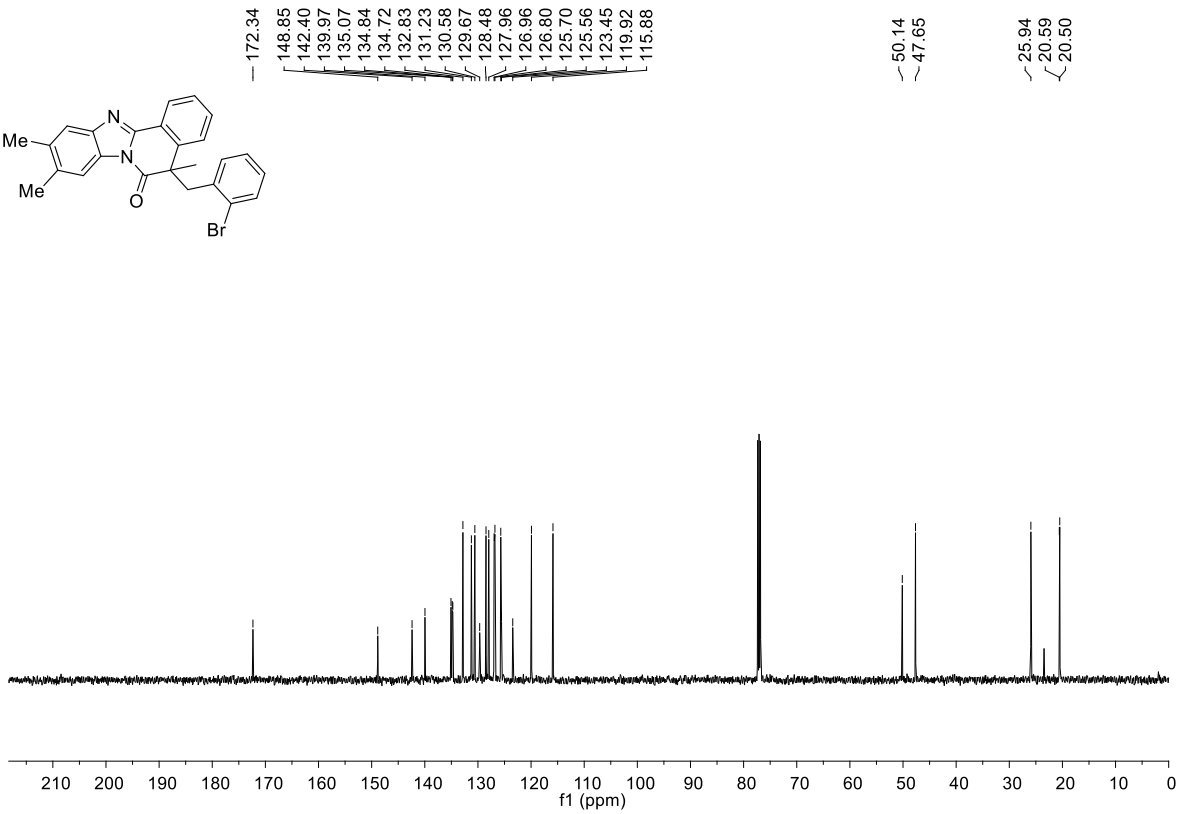
^{13}C{^{1}H} NMR spectrum of 7b ($CDCl_3$, 126 MHz)



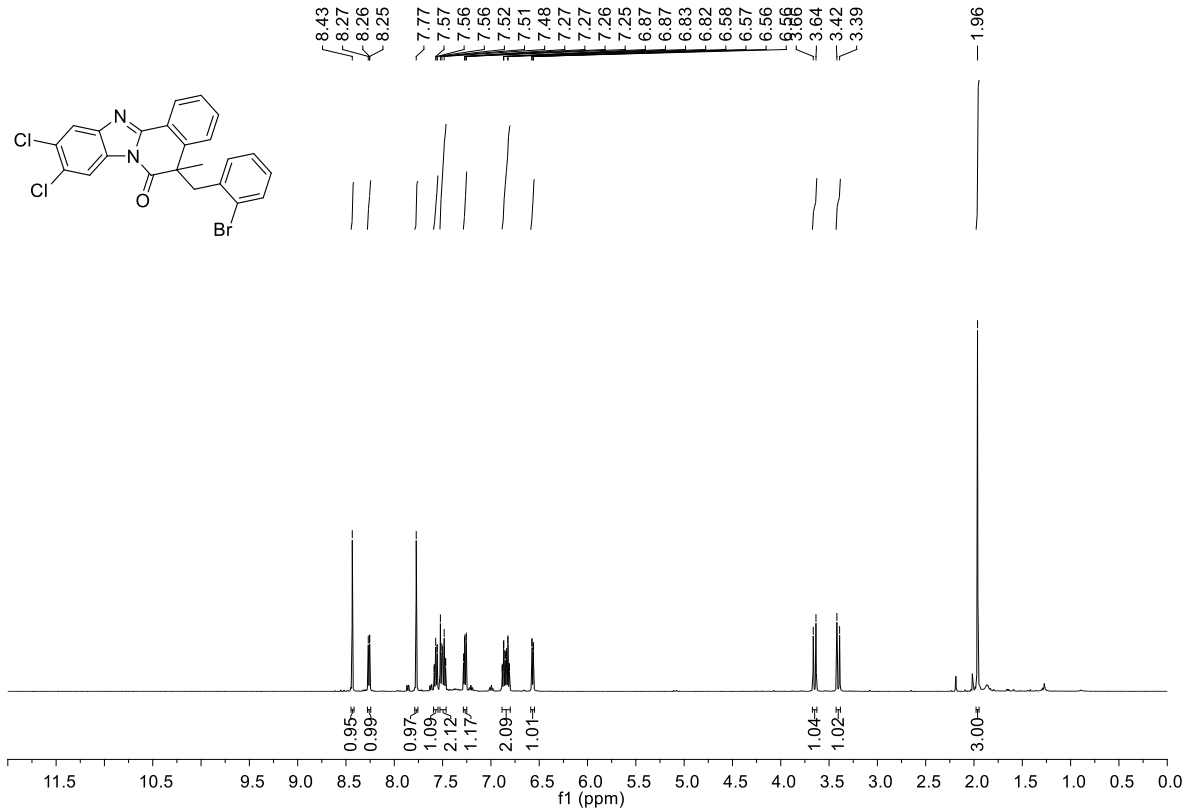
^{1}H NMR spectrum of 7c ($CDCl_3$, 500 MHz)



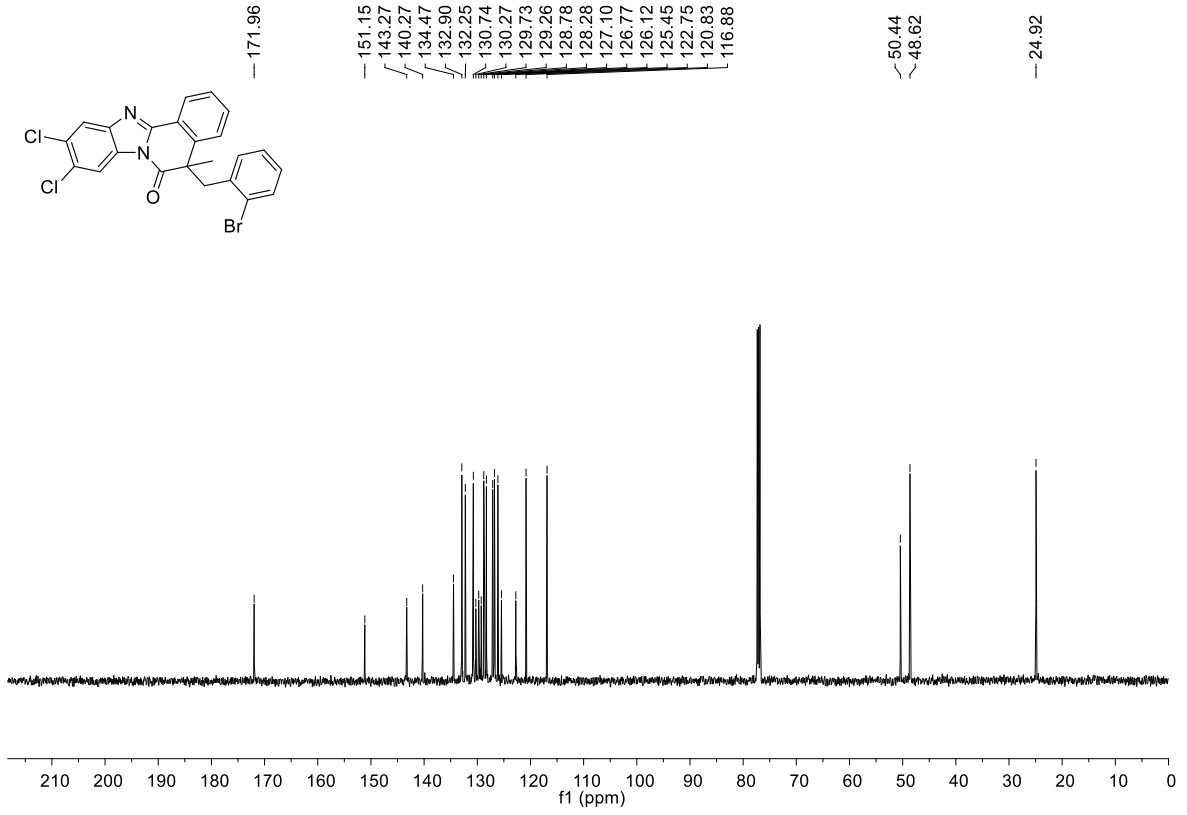
^{13}C{^{1}H} NMR spectrum of 7c ($CDCl_3$, 126 MHz)



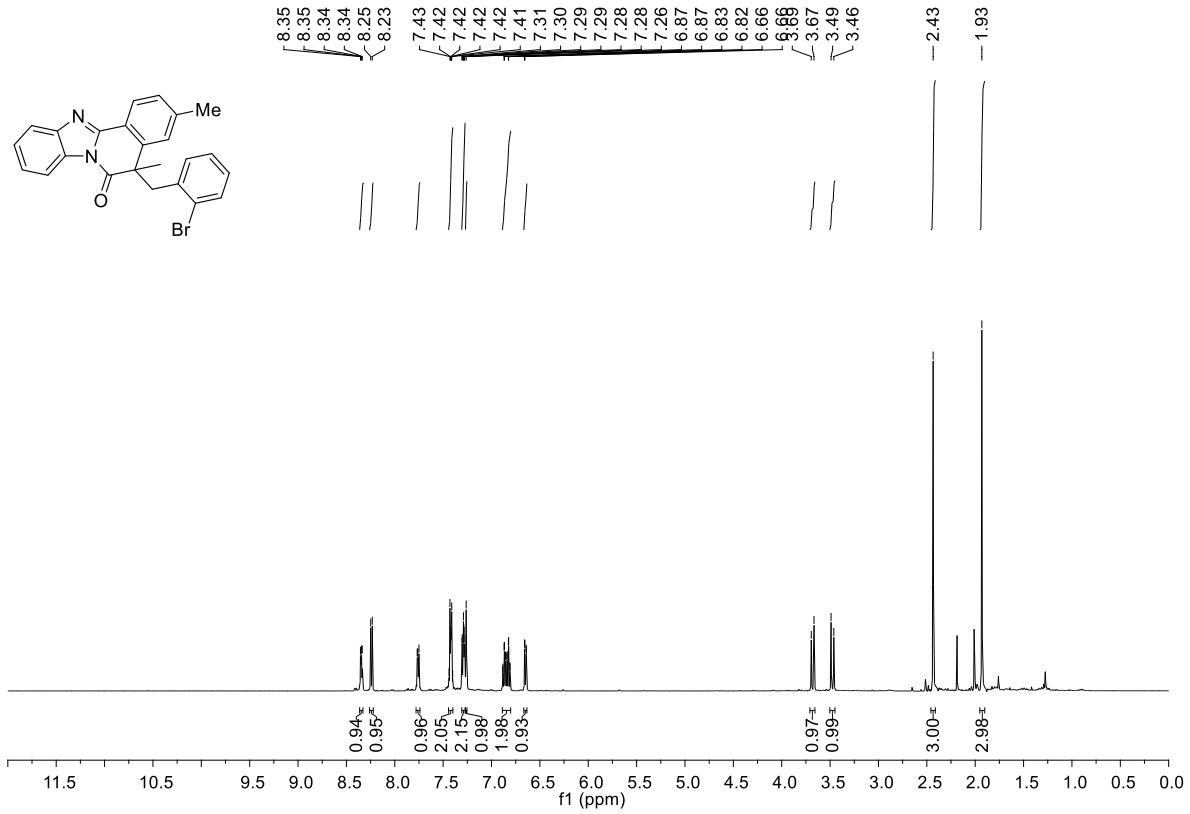
^{1}H NMR spectrum of 7d ($CDCl_3$, 500 MHz)



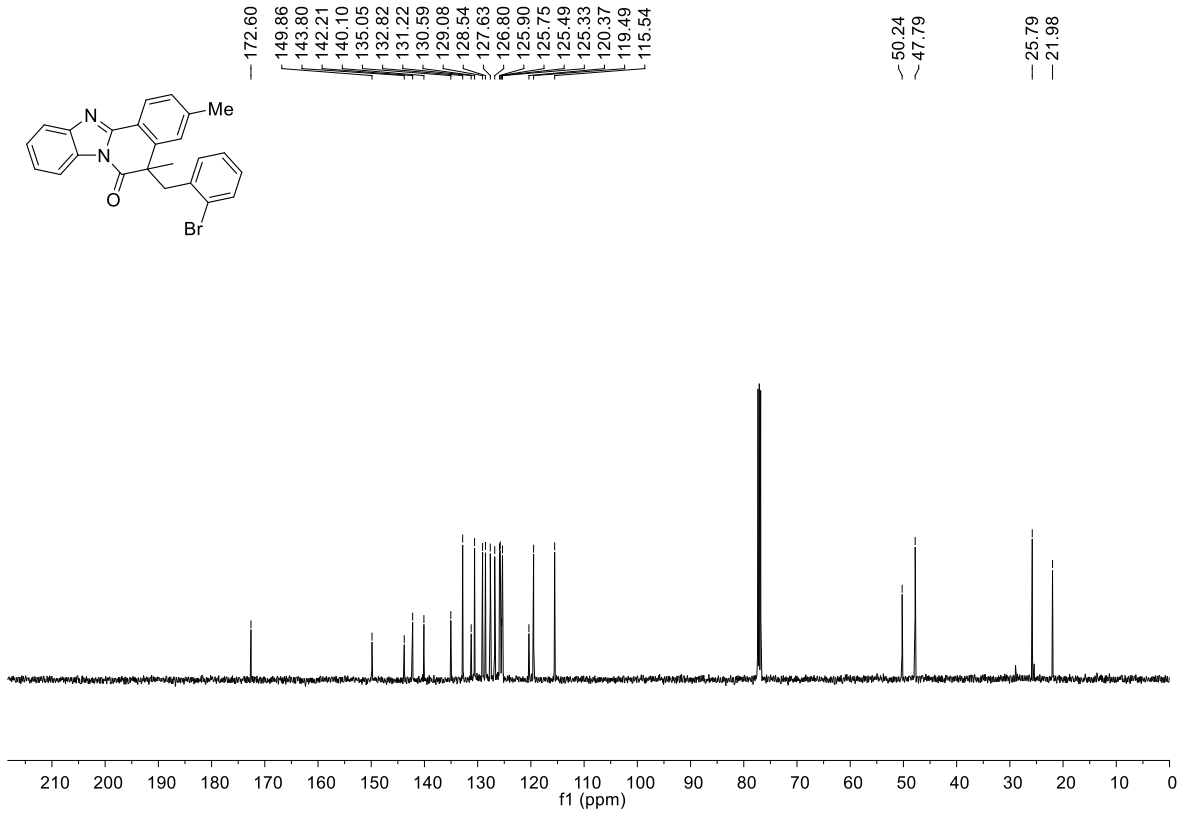
^{13}C{^{1}H} NMR spectrum of 7d ($CDCl_3$, 126 MHz)



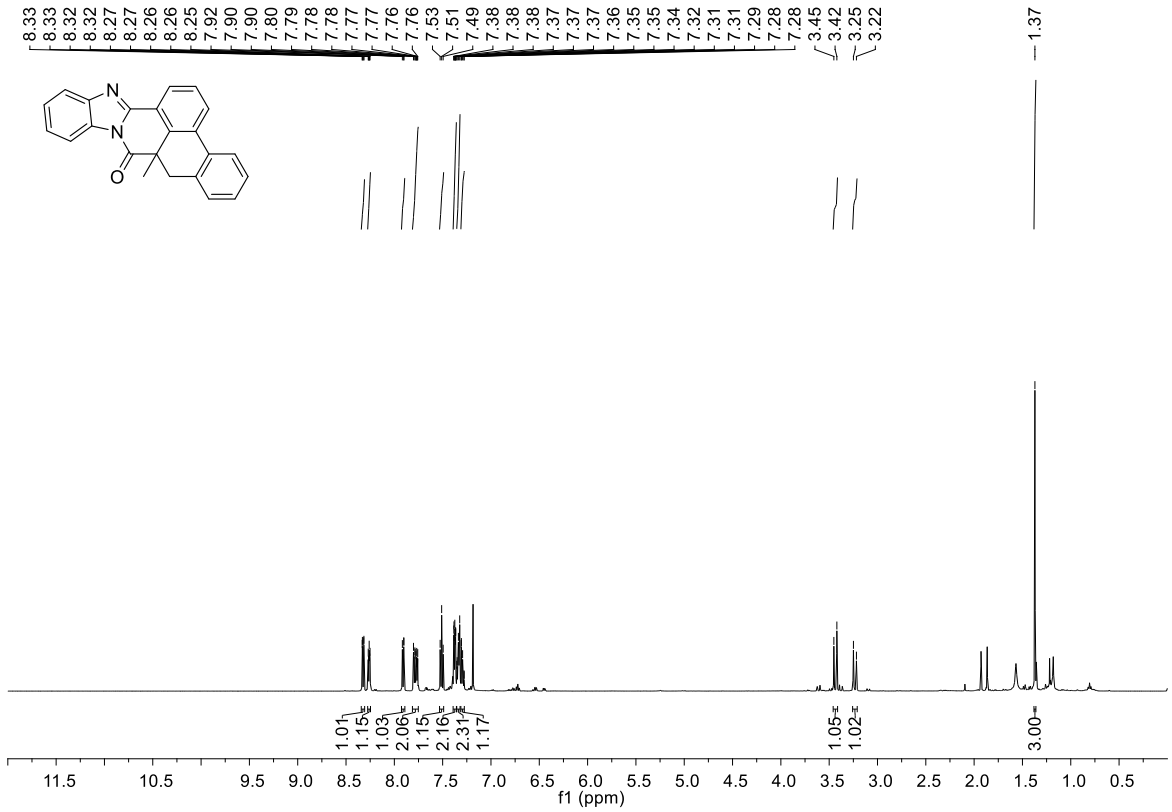
^{1}H NMR spectrum of 7e ($CDCl_3$, 500 MHz)



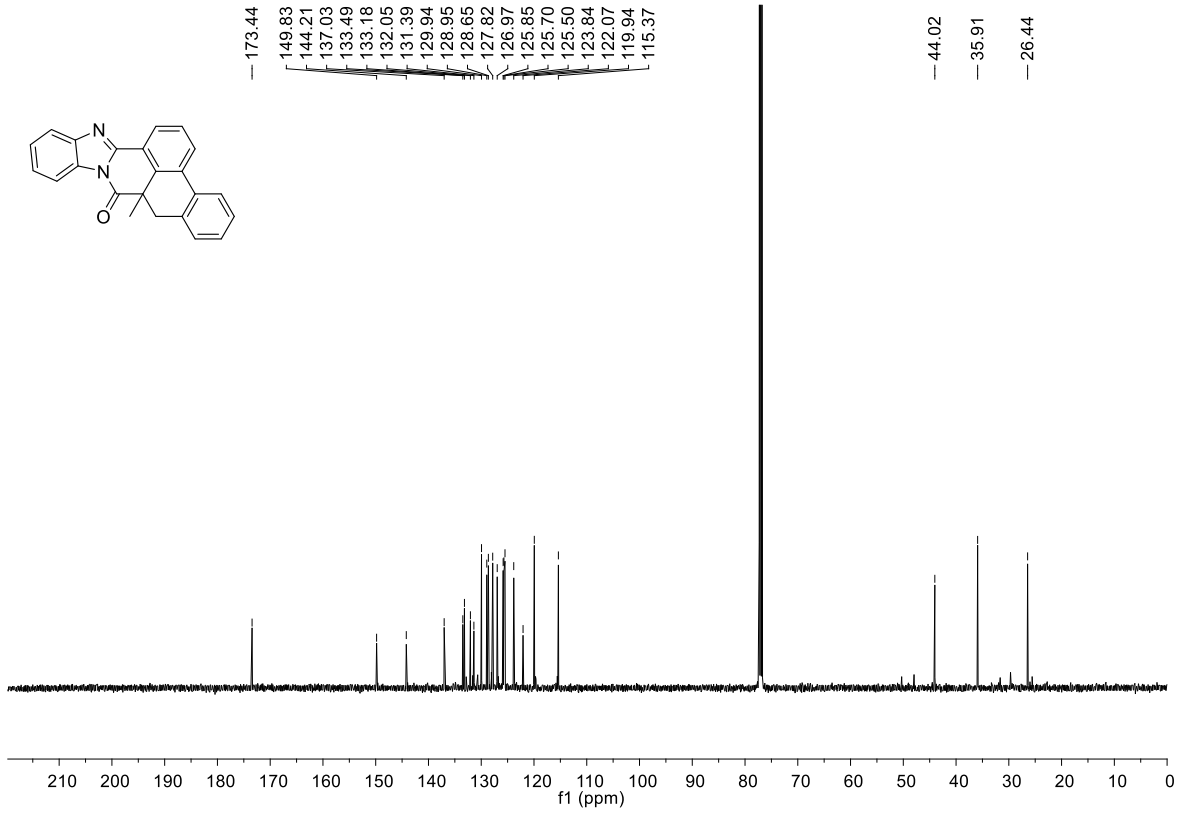
^{13}C{^{1}H} NMR spectrum of 7e ($CDCl_3$, 126 MHz)



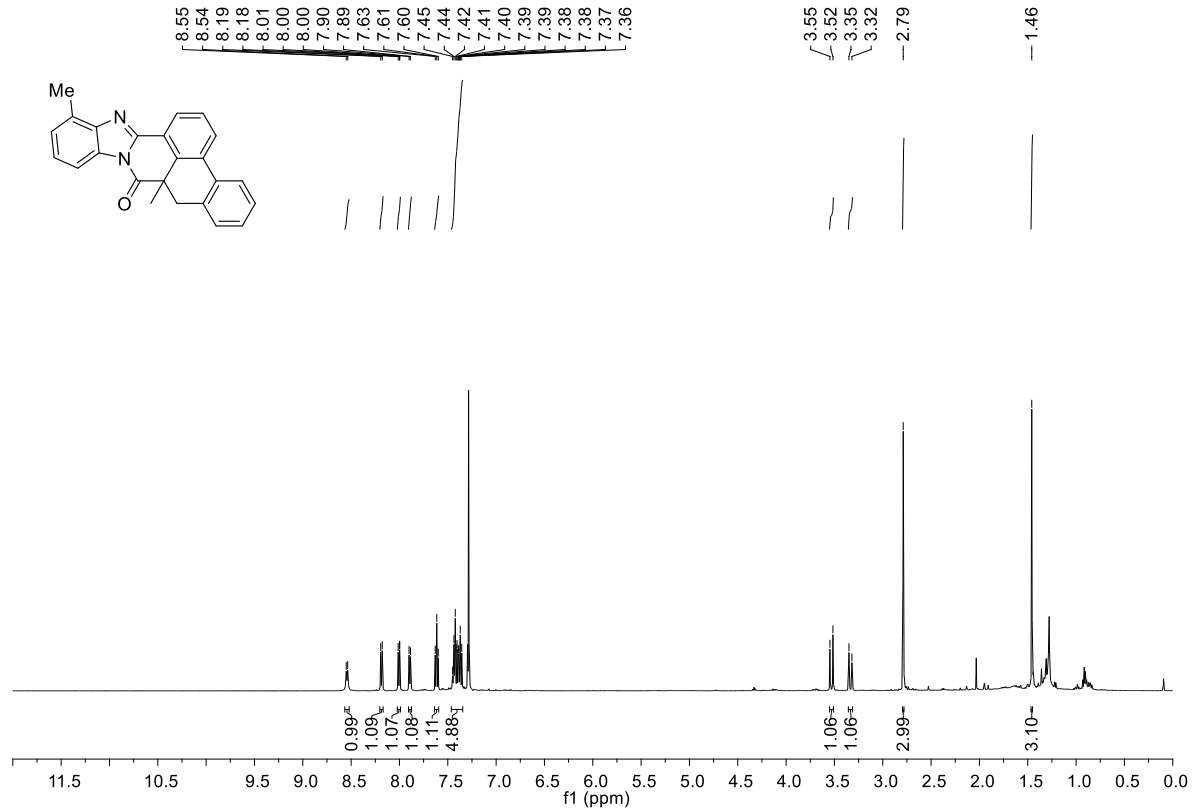
^{1}H NMR spectrum of 8a ($CDCl_3$, 500 MHz)



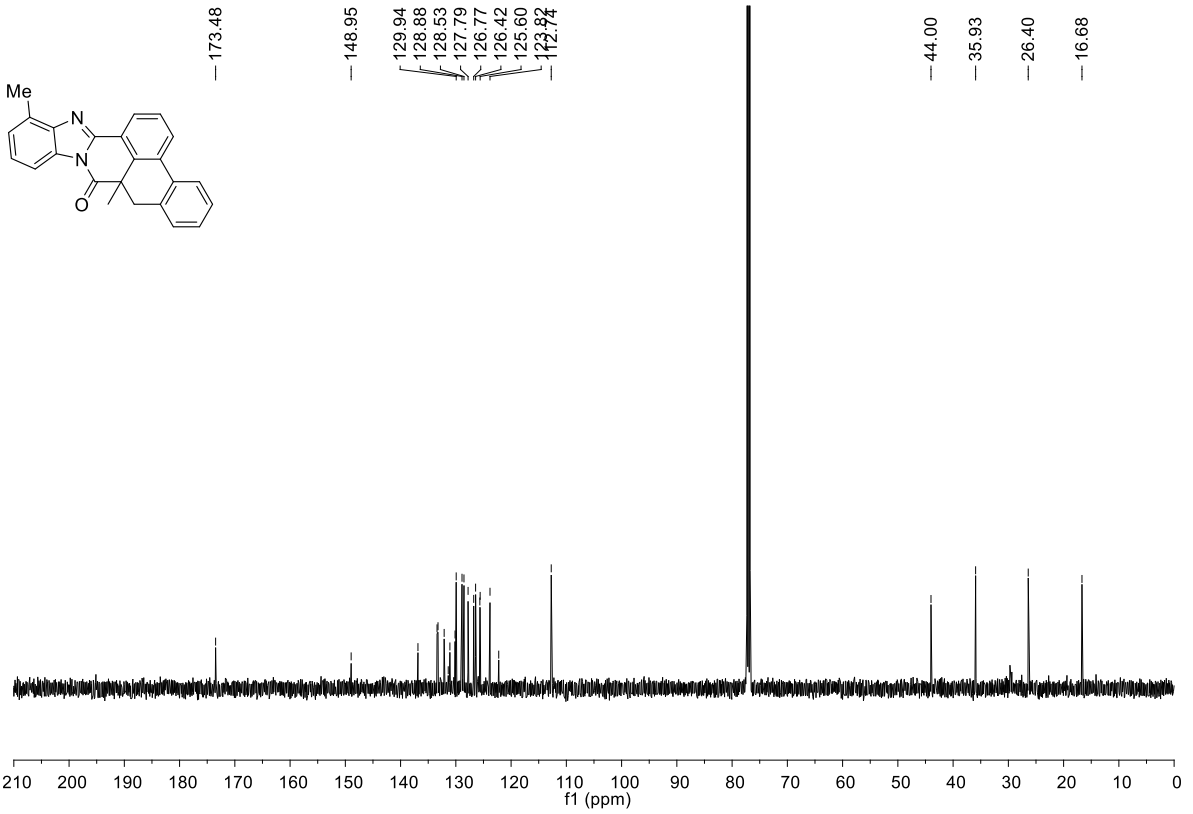
^{13}C{^{1}H} NMR spectrum of 8a ($CDCl_3$, 126 MHz)



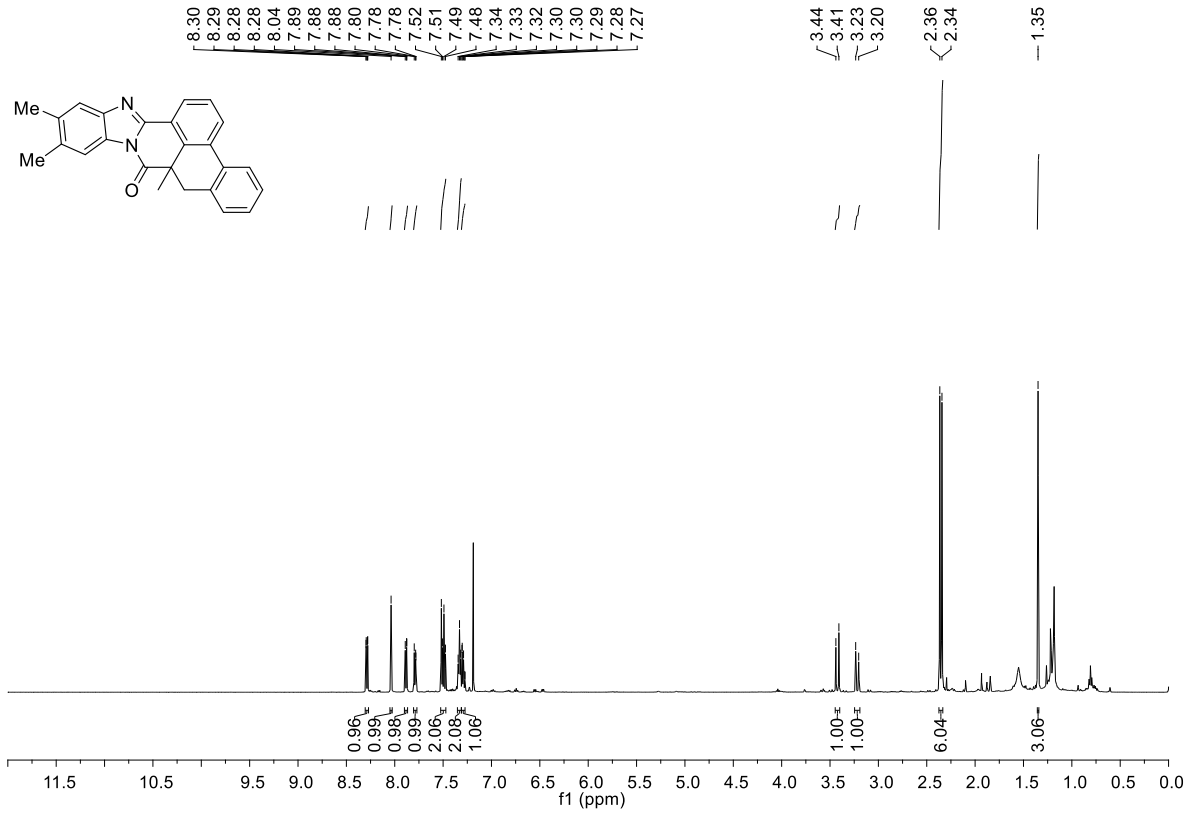
^{1}H NMR spectrum of 8b ($CDCl_3$, 500 MHz)



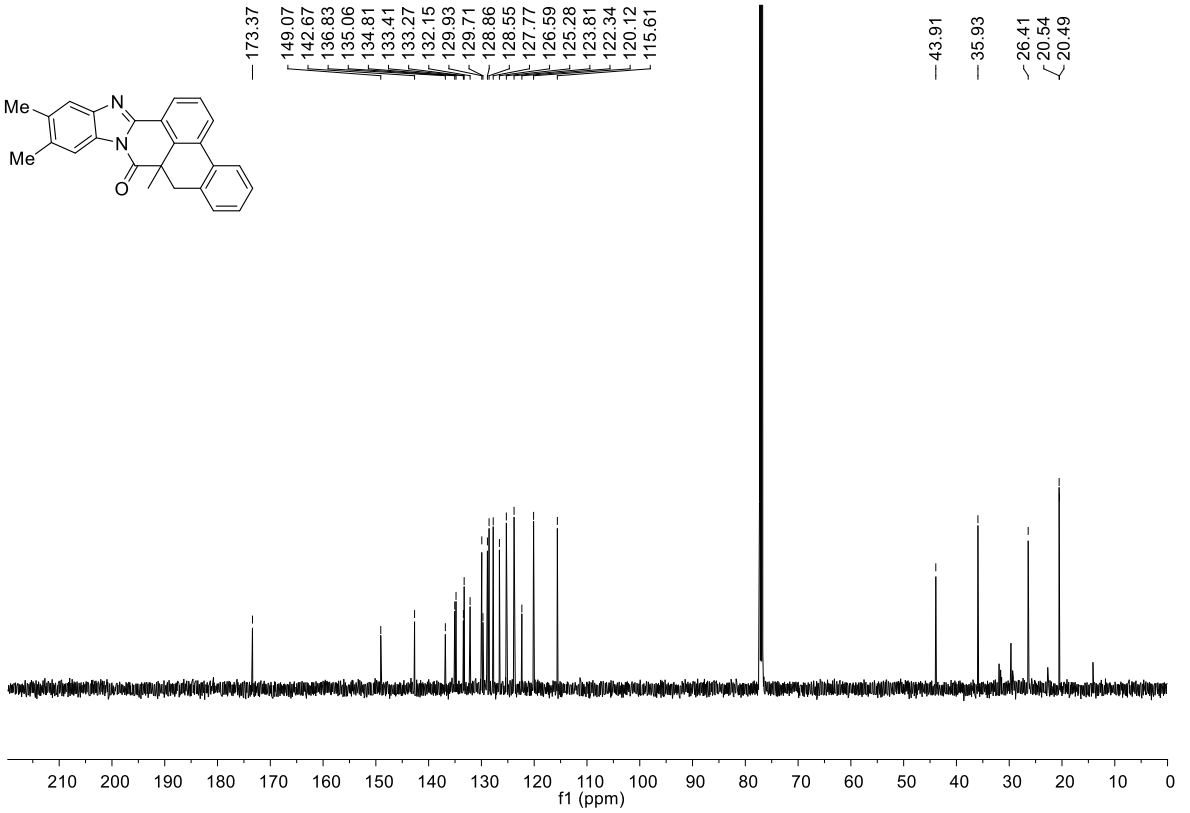
^{13}C{^{1}H} NMR spectrum of 8b ($CDCl_3$, 126 MHz)



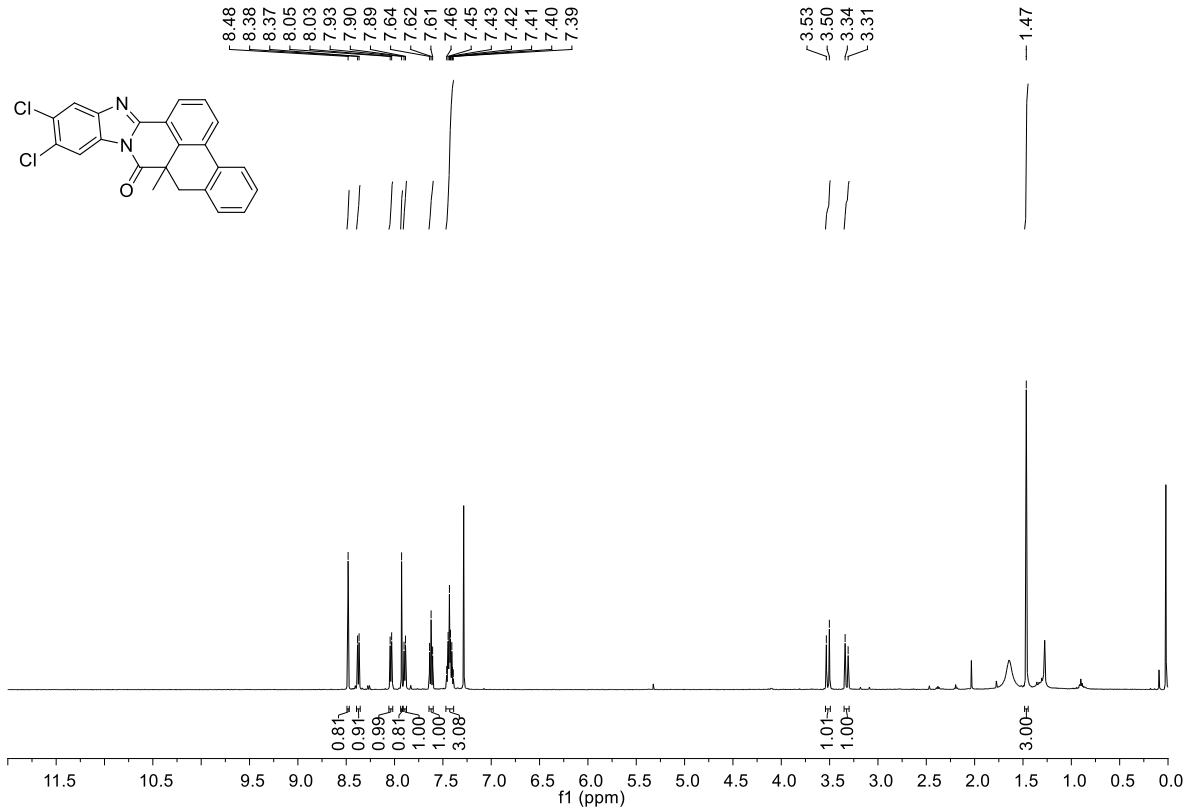
^{1}H NMR spectrum of 8c ($CDCl_3$, 500 MHz)



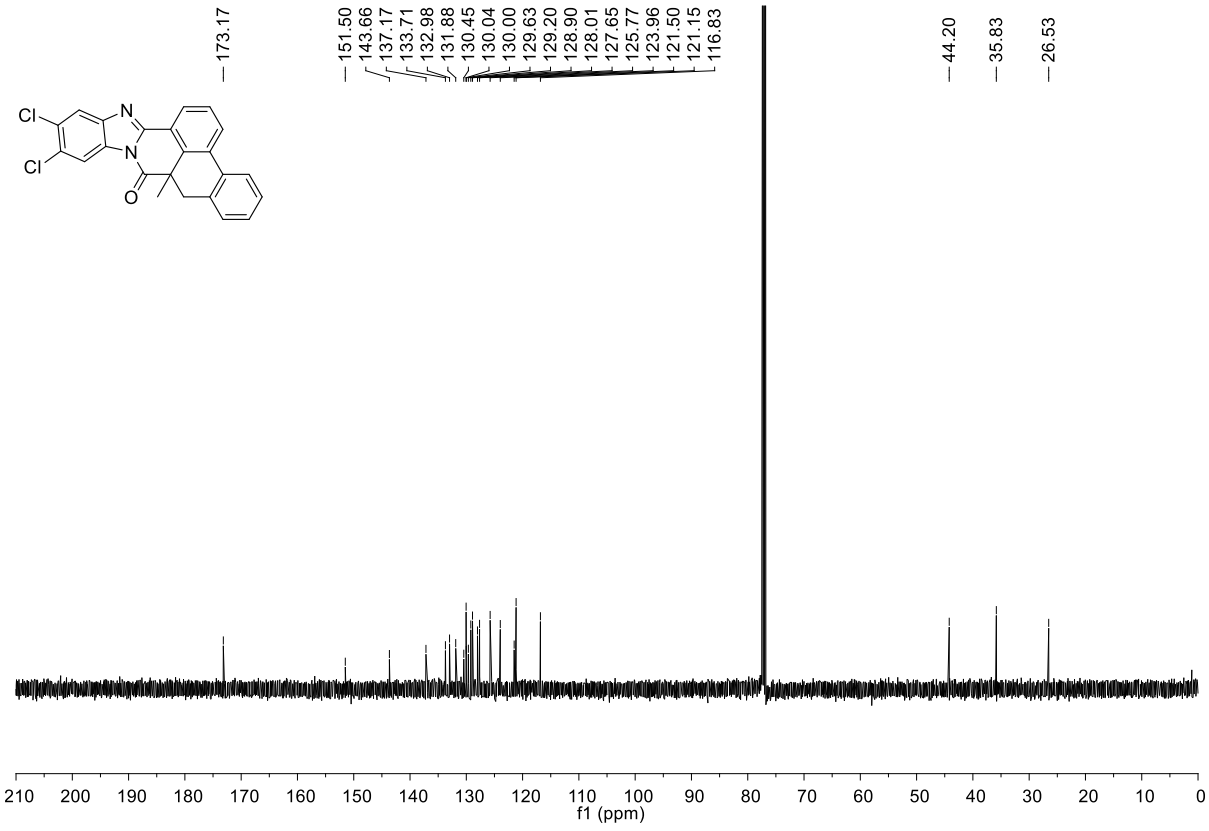
^{13}C{^{1}H} NMR spectrum of 8c ($CDCl_3$, 126 MHz)



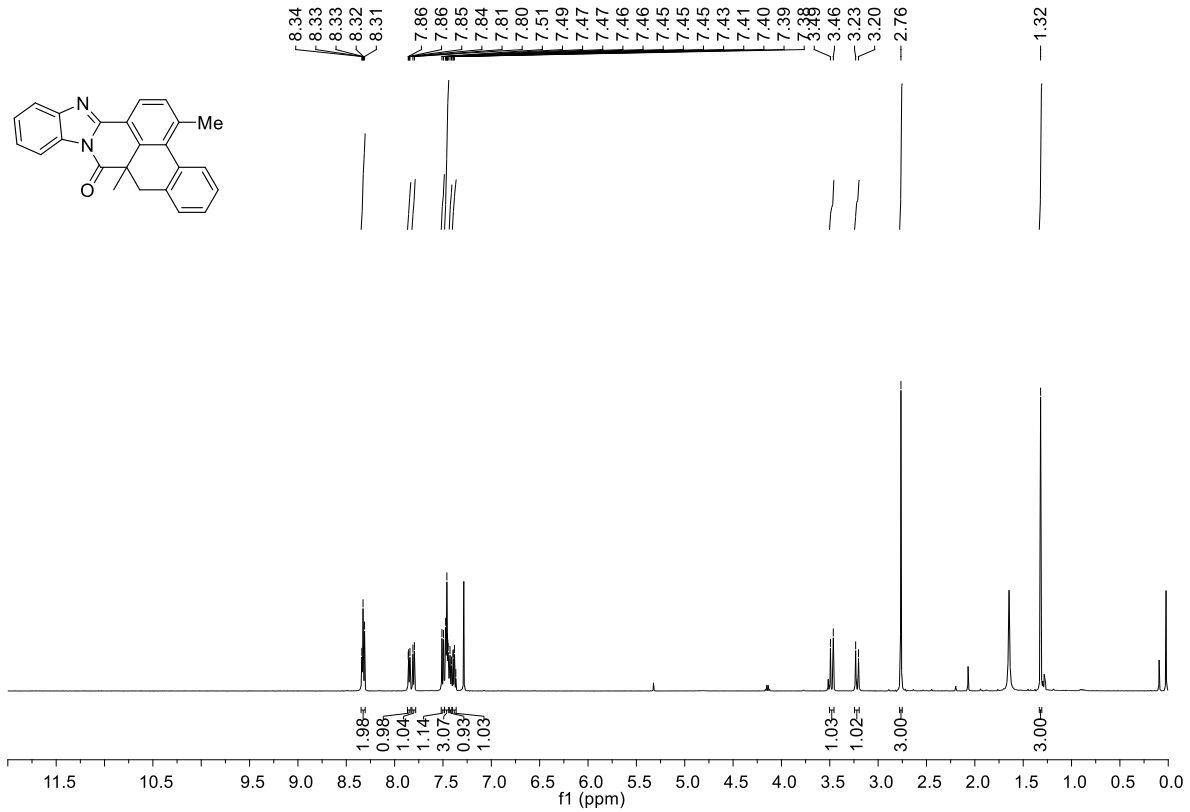
^{1}H NMR spectrum of 8d ($CDCl_3$, 500 MHz)



^{13}C{^{1}H} NMR spectrum of 8d ($CDCl_3$, 126 MHz)



^{1}H NMR spectrum of 8e ($CDCl_3$, 500 MHz)



^{13}C{^{1}H} NMR spectrum of 8e ($CDCl_3$, 126 MHz)