Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.


**Supporting Information for**

# Divergent Synthesis of Fused N-Heterocycles via Rhodium-Catalyzed [4+2] Cyclization of Pyrazolidinones with Iodonium Ylides

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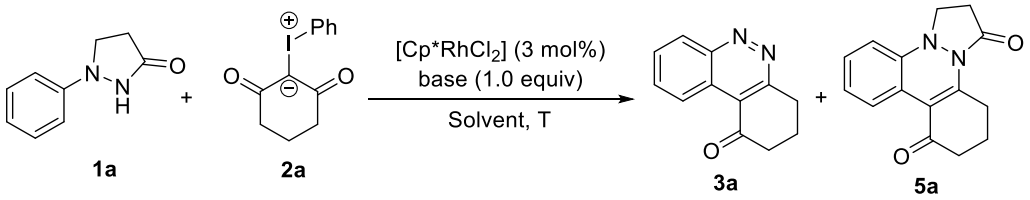
## 1. General information

Unless otherwise noted, materials were purchased from commercial suppliers (Alfa, TCI and Sigma-Aldrich etc.), and used without further purification. All the solvents were treated according to general methods. All reactions were monitored by thin-layer chromatography (TLC) on silica gel plates using UV light as visualizing agent (if applicable). Flash column chromatography was performed using 200-300 mesh silica gel. $^{1}$H NMR spectra were recorded on 400 and 600 MHz spectrophotometers. Chemical shifts are reported in delta (δ (ppm)) units in parts per million (ppm) relative to the singlet (0 ppm) for tetramethylsilane (TMS). Data are reported as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, dd = doublet of doublets, m = multiplet), coupling constants (Hz) and integration. $^{13}$C NMR spectra were recorded on Varian Mercury 100 MHz with complete proton decoupling spectrophotometers ($CDCl_3$: 77.0 ppm). The high resolution mass spectra (HRMS) were measured on a Shimadzu LCMS-IT-TOF mass spectrometer or DIONEX UltiMate 3000 & Bruker Compact TOF mass spectrometer by ESI. Measured values are reported to 4 decimal places of the calculated value. The calculated values are based on the most abundant isotope. An oil bath was used for the synthesis of 1,2-oxazetidines, and a heating module was used for preparation of compounds **3a**-**3z**, **5a**-**5u**, **6**, **7** and **8**.

## 2. Table S1: Screening of the reaction conditions.

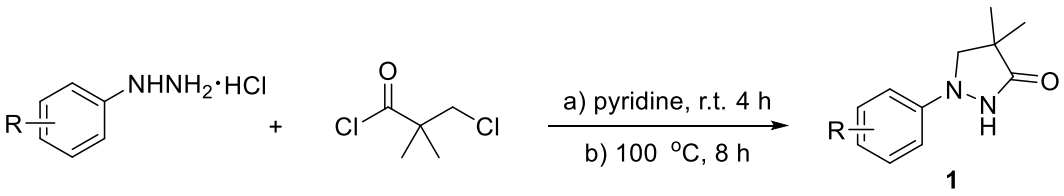



| Entry | Solvent | T (℃) | Base (1.0 equiv) | **5a/3a** (Yield %) |
|---|---|---|---|---|
| 1 | DCE | 100 | NaOAc | 30/trace |
| 2[b] | DCE | 100 | NaOAc | 64/trace |
| 3[b] | DCE | 100 | AgOAc | 44/trace |
| 4[b] | DCE | 100 | $CH_3COOK$ | 71/trace |
| 5[b] | DCE | 100 | $Cs_2CO_3$ | 70/trace |
| 6[b] | DCE | 100 | $Et_3N$ | 72/trace |
| 7[b.c] | DCE | 100 | $Et_3N$ | 82/trace |

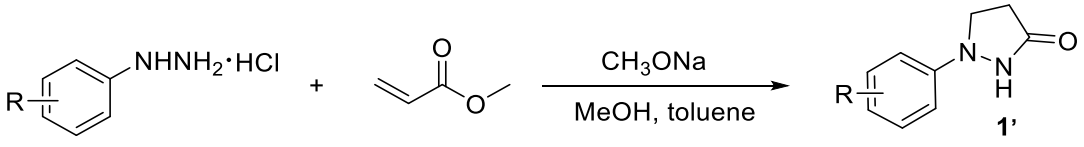
[a]Reaction conditions: 0.2 mmol **1a**, 0.24 mmol **4a**, 3 mol% $[Cp^*RhCl_2]_2$, 2.0 mL solvent, 100 °C, 5 h. [b]Under nitrogen atmosphere. [c]4 mol% $[Cp^*RhCl_2]_2$ was used, 0.3 mmol **4a**, 9 h under nitrogen atmosphere. HFIP = hexafluoro-2-propanol. DCE = 1, 2-dichloroethane. DMF = N, N-dimethylformamide.

## 3. Preparation of substrates

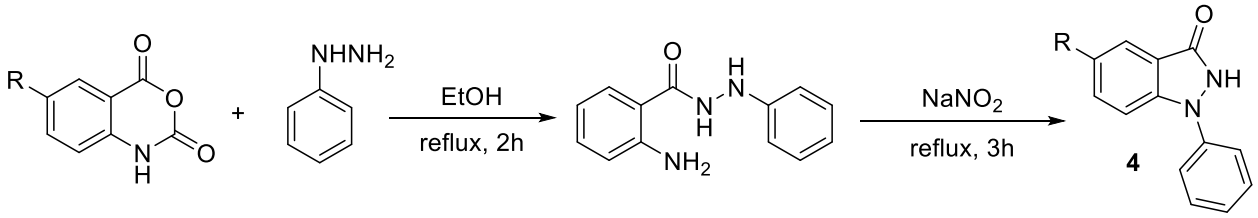
### 3.1 General procedure for preparation of product 1, 2, 4.



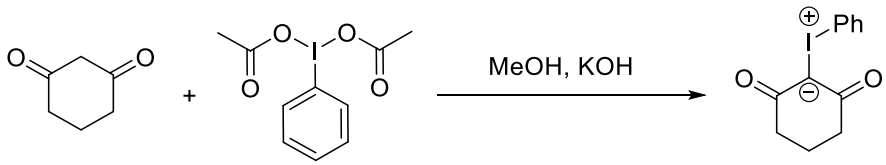

Phenylhydrazine (5 mmol) hydrochloride was added to pyridine (15 mL), then 3-chloro-2,2-dime-thpropa -noyl chloride (5 mmol, 0.77 g) was added dropwisely within 5 minutes at 0 °C. Warm the reaction to room temperature and stirring at room temperature for 4 h. Then stirring at 100 °C for 8 h. After cooling to room temperature, the reaction mixture was poured into 3.0 M HCl solution and extracted with DCM. Purification by flash column chromatography (EtOAc/PE) afforded the product[1].



Substituted phenyl hydrazine hydrochloride (20 mmol) was added to a mixture of sodium methoxide (50 mol), anhydrous methanol (6 mL) and toluene (21 mL). Then, a solution of the α,β-unsaturated acid esters (0.06 mol) in anhydrous methanol (6 mL) was added dropwisely at 30–35 ℃ for 0.5 h, after which the mixture was refluxed until the starting material was completely consumed as judged by TLC. After reaction completion, the mixture was evaporated under reduced pressure. Water (20 mL) was added to the residue and the pH was adjusted to 6.5. The solvent was cooled to 1 ℃, allowed to stand and filtered. The solid was recrystallized from ethyl acetate to give the expected compound [2].



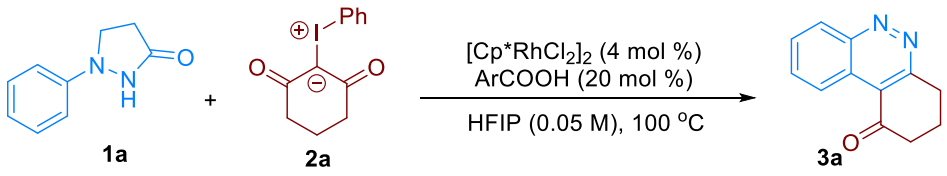

Add 2H-benzo[d][1,3]oxazine-2,4(1H)-dione (6 mmol) and phenylhydrazine (6 mmol, 0.65 g) to ethanol (10 mL), refluxed for 2 hours. After filtering out the solid hydrazine, washed with ethanol and proceed to the next reaction. Dissolved it in 1.0 M hydrochloric acid (12.5 mL), then add an aqueous solution of sodium nitrite (9 mmol, 0.62 g) and ethanol (12.5 mL), refluxed for 3 hours. After cooling, a white solid precipitated [3].




To a solution of cyclic the 1, 3-dione (14 mmol) in 30 mL methanol, added 20 mL 10% aq solution of KOH, followed by the addition of a solution of diacetoxy iodobenzene (15 mmol) in 40 mL methanol. The reaction mixture was stirred for 2 h at room temperature and then quenched with ice cold water. The resulting white precipitate was filtered and mother liquor was extracted with dichloromethane, then washed with water, dried over anhydrous sodium sulfate, filtered and concentrated in vaccuo. The resultant white solid was mixed with the first crop and the mixture recrystallized from DCM/hexane[4].

## 4. General Procedure and Spectral Data of the Products

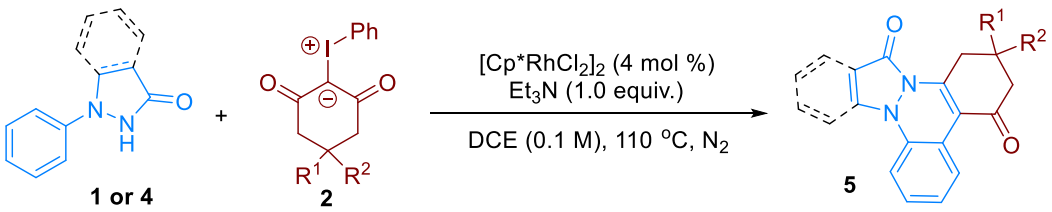
### 4.1 General procedure for the synthesis of 3a-3z.




$ArCO_2H$ = 2,4,6-trimethylbenzoic acid

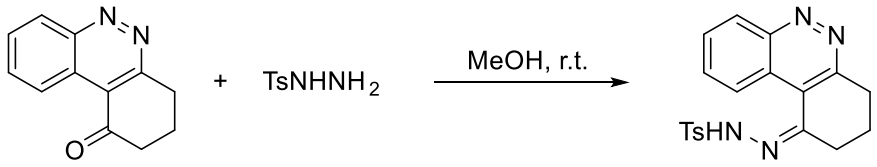
**1a** (0.2 mmol), **2a** (0.3 mmol), $[Cp^*RhCl_2]_2$ (4.9 mg, 4 mol %) and ArCOOH (6.6 mg, 20 mol%) were dissolved in HFIP (2.0 mL). Then, the mixture was stirred at 100 °C for 11 h, as monitored by TLC analysis. The crude product was purified by flash chromatography on silica gel (petroleum ether/ethyl acetate = 2:1) directly to give the desired product **3a** in 79% isolated yield as a yellow solid. Other products **3b-3z** were prepared according to the above procedure. (Note: a heating module was used as the heating source).

### 4.2 General procedure the synthesis of 5a-5u.



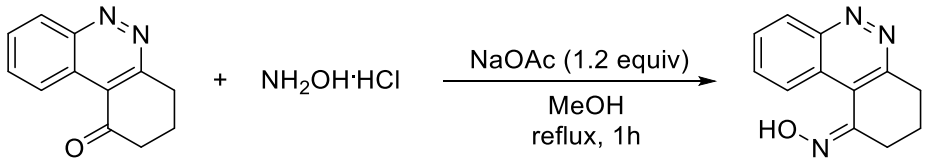

**1a** or **4a** (0.2 mmol), **2a** (0.3 mmol), $[Cp^*RhCl_2]_2$ (4.9 mg, 4 mol %) and $Et_3N$ (27.6 uL, 0.2 mmol) were dissolved in DCE (2 mL). Then, the mixture was stirred at 110 °C for 9 h under the atmosphere of nitrogen, as monitored by TLC analysis. The crude product was purified by flash chromatography on silica gel (petroleum ether/ethyl acetate = 2:1) directly to give the desired product **5a** in 82% isolated yield as a yellow solid. Other products **5b-5u** were prepared according to the above procedure. (Note: a heating module was used as the heating source).

**4.3 General procedure for the synthesis of 6, 7, 8.**



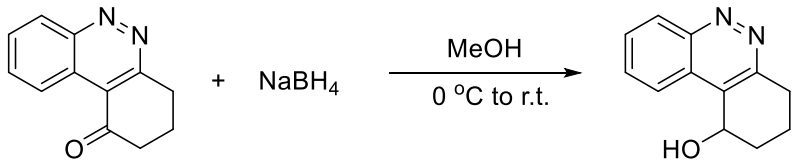

**3a** **6**

A mixture of a **3a** (0.2 mmol, 39.6 mg) and tosylhydrazide (0.2 mmol, 37.2 mg) in methanol (MeOH, 1 mL) was stirred at room temperature overnight. A yellow solid was precipitated, and the compound **6** was obtained by recrystallization as a white solid in 71% yield.




**3a** **7**

Strried the solution of **3a** (0.2 mmol, 39.6 mg), NaOAc (0.24 mmol, 19.7 mg), hydroxylamine hydrochloride (0.26 mmol, 18.1 mg) and MeOH (1mL) at reflux for 1h. Then NaOH (2M, 4 mL) was added to neutralize extra Grignard reagent. Next, the resulting mixture was extracted by EtOAc/$H_2O$. Organic phase was dried over $Na_2SO_4$ and concentrated. The resulting mixture was purified by chromatography on silica gel to afford pure product **7** as a white solid in 70% yield.

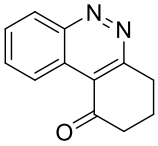



**3a** **8**

**3a** (0.2 mmol, 39.6 mg) was dissolved in MeOH (0.5 mL) and cooled to 0 ℃. Then $NaBH_4$ (0.4 mmol, 15.1 mg) is slowly added and the mixture is stirred for 8 h while warming up to room temperature. The reaction is quenched with $H_2O$ (2 mL) and methanol is removed under reduced pressure. The aqueous phase is extracted three times with DCM or EA, the combined organic phases are washed with brine and dried over $MgSO_4$. The solution is concentrated under reduced pressure. The resulting mixture was purified by chromatography on silica gel to afford pure product **8** as a white solid in 59% yield.

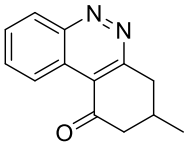
## 4.4 Spectral data of the products 3a-3z, 5a-5u, 6, 7 and 8

**Product 3a**



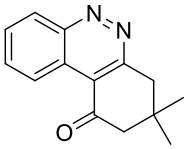
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1)($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3a** as a kelly solid (31.4 mg, 79% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.24 (d, *J* = 8.5 Hz, 1H), 8.56 (d, *J* = 8.2 Hz, 1H), 7.90 – 7.77 (m, 2H), 3.66 (t, *J* = 6.2 Hz, 2H), 2.85 (t, *J* = 6.7 Hz, 2H), 2.39 – 2.29 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.2, 157.0, 151.1, 134.2, 130.6, 129.7, 125.2, 121.3, 118.6, 40.3, 30.7, 21.7. M.P.: 90.0 – 90.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{12}H_{11}N_2O^+$: 199.0866; found: 199.0869.

**Product 3b**



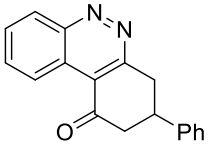
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3b** as a green solid (23.4 mg, 55% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.28 (d, *J* = 8.3 Hz, 1H), 8.60 (d, *J* = 7.6 Hz, 1H), 7.93 – 7.81 (m, 2H), 3.89 – 3.77 (m, 1H), 3.28 (dd, *J* = 17.4, 10.7 Hz, 1H), 2.98 – 2.87 (m, 1H), 2.62 – 2.51 (m, 2H), 1.29 (d, *J* = 6.1 Hz, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ 201.3, 156.4, 151.2, 134.2, 130.7, 129.8, 125.2, 121.4, 118.3, 48.4, 39.0, 29.4, 21.1. M.P.: 112.0 – 112.5 °C. HRMS (ESI-TOF) m/z: $[M+Na]^+$ calcd for $C_{13}H_{12}N_2ONa^+$: 235.0842; found: 235.0838.

**Product 3c**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3c** as a tan solid (23.7 mg, 52% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.28 (d, *J* = 8.5 Hz, 1H), 8.60 (d, *J* = 8.3 Hz, 1H), 7.88 – 7.83 (m, 2H), 3.58 (s, 2H), 2.73 (s, 2H), 1.22 (s, 6H).$^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.5, 155.7, 151.2, 134.2, 130.7, 129.8, 125.2, 121.2, 117.9, 54.0, 44.6, 33.3, 28.1. M.P.: 112.0 – 112.5 °C. HRMS (ESI-TOF) m/z: $[M+Na]^+$ calcd for $C_{14}H_{14}N_2ONa^+$: 249.0998; found: 249.0996.

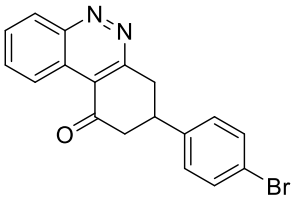
**Product 3d**



The residue was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding the corresponding product **3d** as a yellow solid (40.1 mg,

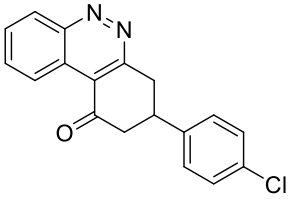
73% yield). $^{1}$H NMR (400 MHz, $CDCl_3$) δ = 9.31 (d, *J* = 8.5 Hz, 1H), 8.62 (d, *J* = 8.3 Hz, 1H), 7.94 – 7.85 (m, 2H), 7.45 – 7.36 (m, 4H), 7.33 (t, *J* = 6.9 Hz, 1H), 4.11 – 4.06 (m, 1H), 3.81 – 3.63 (m, 2H), 3.19 – 3.05 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.6, 156.1, 151.3, 142.0, 134.5, 130.8, 130.0, 129.0, 127.3, 126.6, 125.2, 121.3, 118.3, 47.2, 39.6, 38.4. M.P.: 96.0 – 96.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{18}H_{15}N_2ON^+$: 275.1179; found: 275.1180.

**Product 3e**



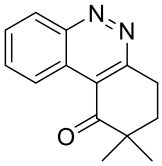

The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3e** as a orange solid (37.9 mg, 54% yield). $^{1}$H NMR (400 MHz, $CDCl_3$) δ = 9.21 (d, *J* = 8.4 Hz, 1H), 8.53 (d, *J* = 8.2 Hz, 1H), 7.85 – 7.77 (m, 2H), 7.45 (d, *J* = 8.3 Hz, 2H), 7.18 (d, *J* = 8.3 Hz, 2H), 3.99 – 3.95 (m, 1H), 3.68 – 3.54 (m, 2H), 3.07 – 2.92 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.1, 155.7, 151.3, 141.0, 134.6, 132.1, 130.8, 130.0, 128.4, 125.1, 121.2, 121.1, 118.2, 47.0, 39.1, 38.2. M.P.: 200.0 – 200.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{18}H_{14}BrN_2O^+$: 353.0284; found: 353.0285.

**Product 3f**




The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3f** as a yellow solid (24.9 mg, 40% yield). $^{1}$H NMR (400 MHz, $CDCl_3$) δ = 9.20 (d, *J* = 8.1 Hz, 1H), 8.53 (d, *J* = 7.9 Hz, 1H), 7.90 – 7.73 (m, 2H), 7.31 – 7.22 (m, 4H), 3.98 – 3.95 (m, 1H), 3.68 – 3.58 (m, 2H), 3.11 – 2.90 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.1, 155.7, 151.3, 140.4, 134.6, 133.1, 130.8, 130.0, 129.1, 128.0, 125.1, 121.2, 118.2, 47.1, 39.0, 38.2. M.P.: 186.0 –186.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{18}H_{14}ClN_2O^+$: 309.0789; found: 309.0786.

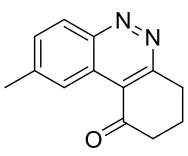
**Product 3g**




The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3g** as a yellow solid (32.6 mg, 72% yield). $^{1}$H NMR (400 MHz, $CDCl_3$) δ = 9.21 (d, *J* = 8.7 Hz, 1H), 8.60 (d, *J* = 8.1 Hz, 1H), 7.92 – 7.80 (m, 2H), 3.69 (t, *J* = 6.4 Hz, 2H), 2.21 (t, *J* = 6.4 Hz, 2H), 1.31 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 205.8, 156.0, 151.1, 134.0, 130.8, 129.7, 125.3, 122.2, 118.0, 43.0, 34.9, 27.0, 24.0. M.P.: 86.0 – 86.5 °C.

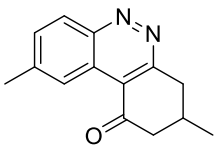
HRMS (ESI-TOF) m/z: $[M+Na]^+$ calcd for $C_{14}H_{14}N_2ONa^+$: 249.0998; found: 249.0994.

**Product 3h**



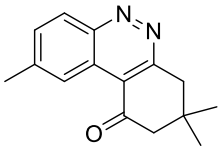
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3h** as a dark solid (26.9 mg, 63% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.06 (s, 1H), 8.46 (d, *J* = 8.7 Hz, 1H), 7.67 (d, *J* = 8.4 Hz, 1H), 3.65 (t, *J* = 6.2 Hz, 2H), 2.89 – 2.83 (m, 2H), 2.64 (s, 3H), 2.37 – 2.32 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.3, 157.1, 150.4, 145.8, 132.2, 130.4, 123.8, 121.9, 118.5, 40.5, 30.8, 22.8, 21.9. M.P.: 84.0 – 84.5 ºC. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{13}H_{13}N_2O^+$: 213.1022; found: 213.1023.

**Product 3i**



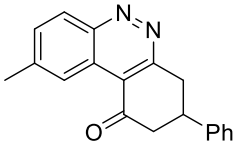
The crude products were purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding a mixture of **3i** as a kelly solid (33.3 mg, 74% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.06 (s, 1H), 8.46 (d, *J* = 8.7 Hz, 1H), 7.66 (d, *J* = 7.9 Hz, 1H), 3.80 (d, *J* = 19.3 Hz, 1H), 3.27 – 3.20 (m, 1H), 2.95 – 2.87 (m, 1H), 2.63 (s, 3H), 2.55 (d, *J* = 9.1 Hz, 2H), 1.28 (d, *J* = 5.8 Hz, 3H).$^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.6, 156.6, 150.4, 145.7, 132.2, 130.4, 123.6, 121.7, 117.9, 48.5, 39.0, 29.4, 22.8, 21.1. M.P.: 104.0 – 104.5 ºC. HRMS (ESI-TOF) m/z: $[M+Na]^+$ calcd for $C_{14}H_{14}N_2ONa^+$: 249.0998; found: 249.0996.

**Product 3j**



The crude products were purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding a mixture of **3j** as a yellow solid (34.1 mg, 71% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.05 (s, 1H), 8.45 (d, *J* = 8.7 Hz, 1H), 7.66 (d, *J* = 9.7 Hz, 1H), 3.53 (s, 2H), 2.70 (s, 2H), 2.63 (s, 3H), 1.20 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.6, 155.7, 150.4, 145.6, 132.1, 130.3, 123.6, 121.5, 117.47, 54.0, 44.6, 33.2, 28.1, 22.7. M.P.: 123.0 – 123.5 ºC. HRMS (ESI-TOF) m/z: $[M+Na]^+$ calcd for $C_{15}H_{16}N_2ONa^+$: 263.1155; found: 263.1152.

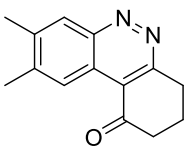
**Product 3k**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3k** as a green solid (51.8 mg, 90% yield).

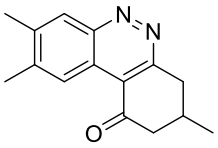
$^{1}$H NMR (400 MHz, $CDCl_3$) δ = 9.08 (s, 1H), 8.47 (d, *J* = 8.7 Hz, 1H), 7.67 (d, *J* = 8.7 Hz, 1H), 7.43 – 7.33 (m, 4H), 7.32 (t, *J* = 6.9 Hz, 1H), 4.03 (d, *J* = 15.0 Hz, 1H), 3.78 – 3.64 (m, 2H), 3.15 – 3.01 (m, 2H), 2.64 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.7, 156.2, 150.5, 146.0, 142.1, 132.3, 130.4, 128.9, 127.3, 126.6 123.6, 121.6, 117.9, 47.3, 39.6, 38.4, 22.8. M.P.: 90.0 – 90.5 ºC. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{19}H_{17}N_2O^+$: 289.1335; found: 289.1340.

**Product 3l**



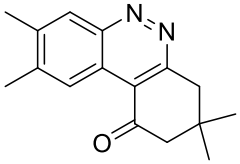
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3l** as a yellow solid (42.2 mg, 93% yield). $^{1}$H NMR (600 MHz, $CDCl_3$) δ = 9.02 (s, 1H), 8.30 (s, 1H), 3.62 (t, *J* = 6.2 Hz, 2H), 2.86 – 2.82 (m, 2H), 2.54 (d, *J* = 5.3 Hz, 6H), 2.35 – 2.31 (m, 2H). $^{13}$C NMR (150 MHz, $CDCl_3$) δ = 201.6, 156.6, 151.1 146.2, 140.4, 129.5, 124.0, 120.4, 118.3, 40.5, 30.8, 21.9, 21.2, 20.3. M.P.: 163.0 – 163.5 ºC. HRMS (ESI-TOF) m/z: $[M+Na]^+$ calcd for $C_{14}H_{14}N_2ONa^+$: 249.0998; found: 249.0994.

**Product 3m**



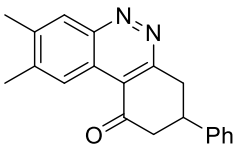
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3m** as a yellow solid (41.3 mg, 86% yield). $^{1}$H NMR (600 MHz, $CDCl_3$) δ = 9.01 (s, 1H), 8.29 (s, 1H), 3.77 – 3.75 (m, 1H), 3.21 (dd, *J* = 17.2, 10.4 Hz, 1H), 2.89 (q, *J* = 11.7 Hz, 1H), 2.53 (d, *J* = 3.7 Hz, 8H), 1.27 (d, *J* = 5.9 Hz, 3H). $^{13}$C NMR (150 MHz, $CDCl_3$) δ = 201.7, 155.9, 151.0, 146.1, 140.4, 129.4, 123.9, 120.2, 117.8, 48.5, 38.9, 29.4, 21.2, 21.1, 20.3. M.P.: 124.0 – 124.5 ºC. HRMS (ESI-TOF) m/z: $[M+Na]^+$ calcd for $C_{15}H_{16}N_2ONa^+$: 263.1155; found: 263.1155.

**Product 3n**



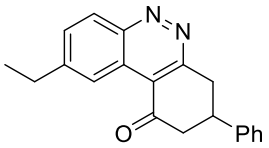
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3n** as a yellow solid (39.9 mg, 78% yield). $^{1}$H NMR (400 MHz, $CDCl_3$) δ = 9.02 (s, 1H), 8.29 (s, 1H), 3.51 (s, 2H), 2.69 (s, 2H), 2.54 (d, *J* = 2.2 Hz, 6H), 1.19 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.8, 155.2, 151.0, 146.1, 140.4, 129.4, 123.9, 120.1, 117.4, 54.0 44.52, 33.3, 28.1, 21.1, 20.3. M.P.: 142.0 – 142.5 ºC. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{16}H_{19}N_2O^+$: 255.1492; found: 255.1492.

**Product 3o**



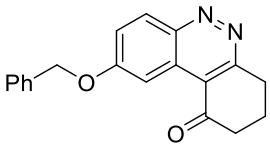
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3o** as a yellow solid (57.5 mg, 95% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.06 (s, 1H), 8.32 (s, 1H), 7.43 – 7.36 (m, 4H), 7.34 – 7.29 (m, 1H), 4.04 – 4.00 (m, 1H), 3.77 – 3.64 (m, 2H), 3.18 – 3.04 (m, 2H), 2.55 (d, *J* = 3.9 Hz, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200. 9, 155.6, 151.2, 146.4, 142.2, 140.6, 129.5, 128.9, 127.3, 126.7, 124.0, 120.2, 117.8, 47.3, 39.7, 38.4, 21.2, 20.3. M.P.: 138.0 – 138.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{20}H_{19}N_2O^+$: 303.1492; found: 303.1486.

**Product 3p**



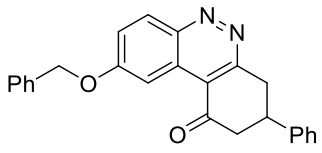
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3p** as a green solid (27.3 mg, 90% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.04 (s, 1H), 8.44 (d, *J* = 8.7 Hz, 1H), 7.65 (dd, *J* = 8.7, 1.6 Hz, 1H), 7.35 – 7.29 (m, 4H), 7.27 – 7.25 (mf, 1H), 4.02 – 3.93 (m, 1H), 3.72 – 3.57 (m, 2H), 3.09 – 2.98 (m, 2H), 2.87 (q, *J* = 7.6 Hz, 2H), 1.31 (t, *J* = 7.6 Hz, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.8, 156.2, 152.0, 150.7, 142.2, 131.3, 130.6, 129.0, 127.3, 126.7, 122.5, 121.8, 118.1, 47.4, 39.6, 38.4, 30.0, 15.1. M.P.: 94.0 – 94.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{20}H_{19}N_2O^+$: 303.1492; found: 303.1494.

**Product 3q**



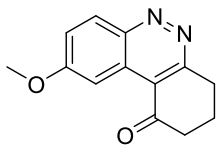
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3q** as a yellow solid (35.6 mg, 59% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.68 (d, *J* = 2.5 Hz, 1H), 8.34 (d, *J* = 9.3 Hz, 1H), 7.47 – 7.39 (m, 3H), 7.35 (t, *J* = 7.3 Hz, 2H), 7.29 (t, *J* = 7.2 Hz, 1H), 5.20 (s, 2H), 3.51 (t, *J* = 6.2 Hz, 2H), 2.80 – 2.72 (m, 2H), 2.28 – 2.20 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.6, 163.2, 157.6, 148.8, 135.5, 132.6, 128.7, 128.4, 127.9, 124.3, 123.9, 117.9, 102.6, 70.7, 40.5, 30.9, 21.8. M.P.:206.0 – 206.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{19}H_{17}N_2O_2^+$: 305.1285; found: 305.1288.

**Product 3r**



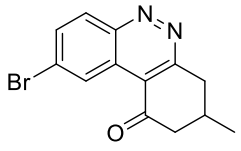
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3r** as a green solid (51.9 mg, 68% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.73 (d, *J* = 2.6 Hz, 1H), 8.38 (d, *J* = 9.3 Hz, 1H), 7.48 – 7.42 (m, 3H), 7.38 – 7.34 (m, 3H), 7.30 (t, *J* = 6.5 Hz, 4H), 7.26 – 7.23 (m, 1H), 5.22 (s, 2H), 3.94 – 3.90 (m, 1H), 3.66 – 3.55 (m, 2H), 3.09 – 2.95 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.9, 163.5, 156.6, 149.0, 142.2, 135.4, 132.7, 129.0, 128.7, 128.5, 128.0, 127.3, 126.7, 124.2, 124.2, 117.5, 102.6, 70.8, 47.4, 39.6, 38.5. M.P.: 142.0 – 142.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{25}H_{21}N_2O_2^+$: 381.1598; found: 381.1594.

**Product 3s**



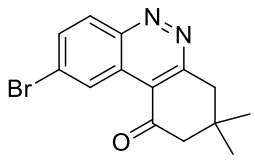
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3s** as a yellow solid (31.2 mg, 68% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.60 (d, *J* = 2.6 Hz, 1H), 8.37 (d, *J* = 9.3 Hz, 1H), 7.39 (dd, *J* = 9.3, 2.6 Hz, 1H), 4.00 (s, 3H), 3.57 (t, *J* = 6.2 Hz, 2H), 2.85 – 2.79 (m, 2H), 2.35 – 2.26 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.6, 164.2, 157.5, 148.9, 132.5, 124.4, 123.6, 117.8, 101.5, 56.0, 40.5, 30.9, 21.8. M.P.: 151.0 – 151.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{13}H_{13}N_2O_2^+$: 229.0972; found: 229.0967.

**Product 3t**



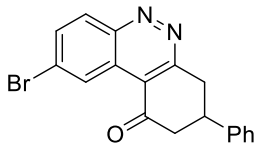
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3t** as a dark solid (28.3 mg, 58% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.54 (d, *J* = 1.7 Hz, 1H), 8.45 (d, *J* = 9.0 Hz, 1H), 7.92 (dd, *J* = 9.0, 1.8 Hz, 1H), 3.86 – 3.81 (m, 1H), 3.27 (dd, *J* = 17.6, 10.3 Hz, 1H), 2.97 – 2.88 (m, 1H), 2.60 – 2.52 (m, 2H), 1.29 (d, *J* = 5.9 Hz, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.9, 157.0, 149.6, 133.8, 132.1, 130.4, 127.7, 122.2, 116.9, 48.2, 38.9, 29.3, 21.1. M.P.: 112.0 – 112.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{13}H_{12}BrN_2O^+$: 291.0128; found: 291.0127.

**Product 3u**



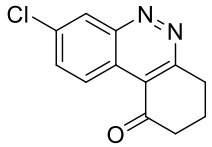
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3u** as a brown solid (43.8 mg, 72% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.54 (s, 1H), 8.45 (d, *J* = 9.0 Hz, 1H), 7.92 (d, *J* = 8.9 Hz, 1H), 3.57 (s, 2H), 2.72 (s, 2H), 1.21 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 201.0, 156.3, 149.7, 133.8, 132.1, 130.3, 127.7, 122.0, 116.5, 53.8, 44.6, 33.3, 28.1. M.P.: 96.0 – 96.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{14}H_{14}BrN_2O^+$: 305.0284; found: 305.0283.

**Product 3v**



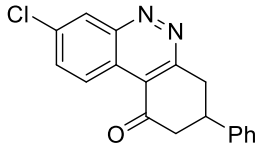
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3v** as a dark solid (44.6 mg, 63% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.57 (d, *J* = 1.8 Hz, 1H), 8.47 (d, *J* = 9.0 Hz, 1H), 7.95 (dd, *J* = 9.0, 1.9 Hz, 1H), 7.44 – 7.33 (m, 5H), 4.12 – 4.05 (m, 1H), 3.81 – 3.77 (m, 1H), 3.73 – 3.64 (m, 1H), 3.20 – 3.19 (m, 1H), 3.16 – 3.05 (m, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.1, 156.7, 149.7, 141.8, 133.9, 132.1, 130.6, 129.0, 127.6, 127.4, 126.6, 122.0, 116.9, 47.0, 39.5, 38.4. M.P.: 110.0 – 110.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{18}H_{14}BrN_2O^+$: 353.0284; found: 353.0272.

**Product 3w**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3w** as a yellow solid (38.6 mg, 83% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.27 (d, *J* = 9.3 Hz, 1H), 8.59 (d, *J* = 2.1 Hz, 1H), 7.81 (dd, *J* = 9.3, 2.2 Hz, 1H), 3.68 (t, *J* = 6.2 Hz, 2H), 2.90 – 2.85 (m, 2H), 2.40 – 2.33 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.9, 157.3, 151.4, 135.8, 135.1, 129.2, 127.2, 119.9, 118.6, 40.3, 30.7, 21.7. M.P.: 100.0 – 100.5 °C. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{12}H_{10}ClN_2O^+$: 233.0476; found: 233.0477.

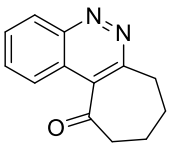
**Product 3x**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3x** as a yellow solid (35.4 mg, 57% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 9.30 (d, *J* = 9.2 Hz, 1H), 8.61 (d, *J* = 2.1 Hz, 1H), 7.84 (dd, *J* = 9.2, 2.2 Hz, 1H), 7.45 – 7.33 (m, 5H), 4.12 – 4.04 (m, 1H), 3.81 – 3.66 (m, 2H), 3.20 –

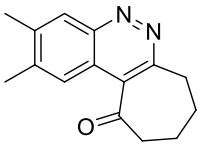
3.15 (m, 1H), 3.09 – 3.05 (m, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 200.3, 156.3, 151.5, 141.8, 136.0, 135.3, 129.2, 129.0, 127.4, 127.0, 126.6, 119.7, 118.1, 47.2, 39.5, 38.3. M.P.: 220.0 – 220.5 ºC. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{18}H_{14}ClN_2O^+$: 309.0789; found: 309.0808.

**Product 3y**



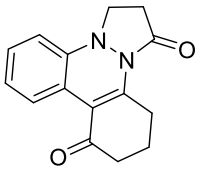
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3y** as a green liquid (27.3 mg, 55% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.49 (d, *J* = 8.1 Hz, 1H), 8.02 (d, *J* = 8.0 Hz, 1H), 7.77 – 7.69 (m, 2H), 3.54 – 3.49 (m, 2H), 2.84 – 2.78 (m, 2H), 2.06 – 2.00 (m, 2H), 1.94 – 1.90 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 206.5, 151.8, 150.3, 132.6, 130.2, 130.0, 129.5, 123.8, 121.4, 42.6, 33.7, 24.5, 23.5. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{13}H_{13}N_2O^+$: 213.1022; found: 213.1023.

**Product 3z**



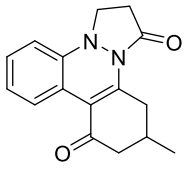
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **3z** as a yellow solid (27.3 mg, 57% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.26 (s, 1H), 7.80 (s, 1H), 3.55 – 3.49 (m, 2H), 2.87 – 2.82 (m, 2H), 2.51 (s, 3H), 2.46 (s, 3H), 2.10 – 2.03 (m, 2H), 1.97 – 1.91 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 207.0, 151.1, 150.1, 144.0, 140.8, 128.9, 128.7, 122.4, 120.3, 42.6, 33.7, 24.6, 23.4, 20.9, 20.4. M.P.: 116.0 – 116.5 ºC. HRMS (ESI-TOF) m/z: $[M+H]^+$ calcd for $C_{15}H_{17}N_2O^+$: 241.1335; found: 241.1337.

**Product 5a**



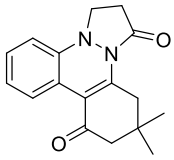
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5a** as a orange red solid (31 mg, 82% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.01 (d, *J* = 7.8 Hz, 1H), 7.11 (t, *J* = 7.8 Hz, 1H), 6.94 (t, *J* = 7.7 Hz, 1H), 6.49 (d, *J* = 8.0 Hz, 1H), 3.58 (t, *J* = 8.3 Hz, 2H), 3.20 (t, *J* = 6.2 Hz, 2H), 2.81 (t, *J* = 8.3 Hz, 2H), 2.54 – 2.48 (m, 2H), 2.05 – 1.99 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.7, 168.0, 153.1, 146.3, 128.5, 126.3, 123.2, 121.1, 116.0, 110.9, 46.9, 38.2, 32.0, 24.9, 20.4. M.P.: 152.0 – 152.5 ℃. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{15}H_{15}N_2O_2^+$: 255.1128; found: 255.1126.

**Product 5b**



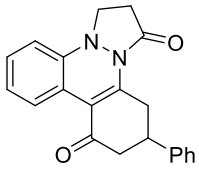
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5b** as a yellow solid (55 mg, 92% yield). 1H NMR (400 MHz, CDCl3) δ = 8.04 (d, *J* = 7.8 Hz, 1H), 7.12 (t, *J* = 7.7 Hz, 1H), 6.95 (t, *J* = 7.7 Hz, 1H), 6.50 (d, *J* = 7.4 Hz, 1H), 3.69 – 3.53 (m, 3H), 2.83 (t, *J* = 8.3 Hz, 2H), 2.61 – 2.47 (m, 2H), 2.29 – 2.18 (m, 2H), 1.13 (d, *J* = 6.1 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl3) δ = 195.8, 167.8, 152.5, 146.4, 128.6, 126.3, 123.3, 121.1, 115.8, 110.9, 47.1, 46.5, 33.0, 32.1, 28.3, 21.0. M.P.: 118.0 – 118.5 °C. HRMS (ESI): m/z [M + H]+ calcd for $C_{16}H_{17}N_2O_2^+$: 269.1285; found: 269.1284.

**Product 5c**



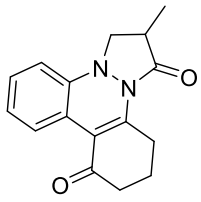
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5c** as a yellow solid (40.7 mg, 72% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.05 (d, *J* = 7.8 Hz, 1H), 7.12 (t, *J* = 7.8 Hz, 1H), 6.95 (t, *J* = 7.7 Hz, 1H), 6.50 (d, *J* = 8.0 Hz, 1H), 3.61 (t, *J* = 8.2 Hz, 2H), 3.05 (s, 2H), 2.83 (t, *J* = 8.2 Hz, 2H), 2.39 (s, 2H), 1.16 – 1.11 (m, 6H) $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.8, 167.7, 151.2, 146.4, 128.6, 126.1, 123.2, 120.9, 115.1, 110.9, 52.0, 47.1, 38.6, 32.2, 32.0, 28.3. M.P.: 153.0 – 153.5 °C. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{17}H_{19}N_2O_2^+$: 283.1441; found: 283.1439.

**Product 5d**



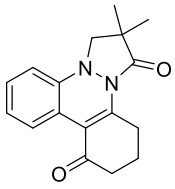
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5d** as a yellow solid (54.8 mg, 83% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.09 – 8.05 (m, 1H), 7.37 – 7.31 (m, 2H), 7.28 – 7.25 (m, 3H), 7.15 – 7.10 (m, 1H), 6.96 (t, *J* = 7.3 Hz, 1H), 6.49 (d, *J* = 7.9 Hz, 1H), 3.88 (dd, *J* = 19.0, 4.2 Hz, 1H), 3.65 – 3.50 (m, 2H), 3.40 – 3.31 (m, 1H), 2.94 (dd, *J* = 19.0, 11.3 Hz, 1H), 2.84 – 2.73 (m, 4H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ= 194.9, 167.8, 152.2, 146.3, 142.2, 128.7, 128.7, 127.1, 126.7, 126.3, 123.3, 120.9, 115.7, 111.0, 47.0, 44.9, 38.5, 32.5, 31.9. M.P.: 137.0 – 137.5 °C. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{21}H_{19}N_2O_2^+$ : 331.1441; found: 331.1433.

**Product 5e**



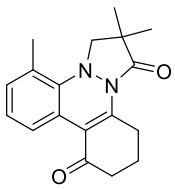
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5e** as a yellow solid (32.5 mg, 61% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.02 (d, *J* = 7.8 Hz, 1H), 7.11 (t, *J* = 7.7Hz, 1H), 6.95 (t, *J* = 7.7 Hz, 1H), 6.50 (d, *J* = 8.0 Hz, 1H), 3.92 (t, *J* = 8.8 Hz, 1H), 3.54 – 3.46 (m, 1H), 3.04 (t, *J* = 8.9 Hz, 1H), 2.97 – 2.87 (m, 2H), 2.60 – 2.43 (m, 2H), 2.13 – 1.93 (m, 2H), 1.35 (d, *J* = 6.9 Hz, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ= 195.7, 170.9, 153.0, 146.2, 128.5, 126.4, 123.2, 121.1, 116.0, 111.0, 54.5, 38.2, 37.5, 24.9, 20.5, 13.6. M.P.: 134.0 – 134.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{16}H_{17}N_2O_2^+$: 269.1285; found: 269.1287.

**Product 5f**



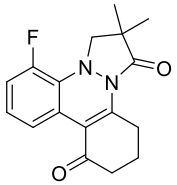
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5f** as a yellow solid (34.3 mg, 61% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.02 (d, *J* = 7.8 Hz, 1H), 7.10 (t, *J* = 7.7 Hz, 1H), 6.94 (t, *J* = 7.6 Hz, 1H), 6.47 (d, *J* = 8.0 Hz, 1H), 3.35 (s, 2H), 3.20 (t, *J* = 6.1 Hz, 2H), 2.55 – 2.50 (m, 2H), 2.07 – 2.01 (m, 2H), 1.33 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ= 195.7, 173.5, 153.0, 146.2, 128.4, 126.3, 123.1, 121.0, 116.0, 110.9, 60.4, 41.7, 38.2, 24.8, 23.0, 20.4. M.P.: 74.0 – 74.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{19}N_2O_2^+$: 283.1411; found: 283.1462.

**Product 5g**



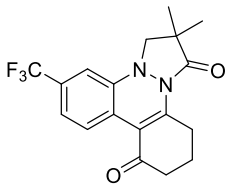
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5g** as a yellow solid (27.9 mg, 80% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 7.86 (d, *J* = 7.6 Hz, 1H), 6.86 (t, *J* = 7.5 Hz, 2H), 3.60 (s, 2H), 3.15 (t, *J* = 6.2 Hz, 2H), 2.54 – 2.49 (m, 2H), 2.29 (s, 3H), 2.09 – 2.03 (m, 2H), 1.30 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.7, 173.3, 152.4, 147.5, 132.9, 124.8, 122.6, 121.3, 119.9, 117.4, 64.4, 40.6, 38.6, 25.6, 22.6, 22.5, 21.1. M.P.: 112.0 – 112.5 ºC. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{18}H_{20}N_2O_2Na^+$: 319.1417; found: 319.1422.

**Product 5h**



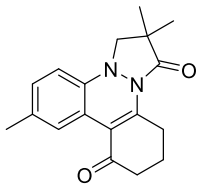
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5h** as a yellow solid (36.8 mg, 61% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 7.83 (d, *J* = 7.0 Hz, 1H), 6.90 – 6.81 (m, 2H), 3.70 (d, *J* = 5.1 Hz, 2H), 3.20 (t, *J* = 6.2 Hz, 2H), 2.55 – 2.50 (m, 2H), 2.08 – 2.01 (m, 2H), 1.30 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.3, 173.6, 153.7, 148.9 (d, *J* = 240.3 Hz), 133.8 (d, *J* = 7.6 Hz), 123.2 (d, *J* = 2.3 Hz), 123.1 (d, *J* = 2.3 Hz), 122.4 (d, *J* = 3.0 Hz), 116.5 (d, *J* = 21.9 Hz), 116.0 (d, *J* = 2.9 Hz), 62.8 (d, *J* = 13.9 Hz), 41.2 (d, *J* = 3.3 Hz), 38.3, 25.2, 22.5, 20.5. M.P.: 145.0 – 145.5 °C. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{18}H_{17}FN_2O_2^+$ : 301.1347; found: 301.1351.

**Product 5i**



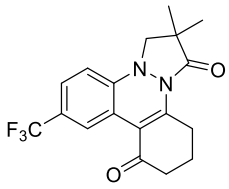
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5i** as a yellow solid (59.5 mg, 85% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.14 (d, *J* = 8.1 Hz, 1H), 7.17 (d, *J* = 8.2 Hz, 1H), 6.61 (s, 1H), 3.38 (s, 2H), 3.22 (t, *J* = 6.2 Hz, 2H), 2.53 (t, *J* = 6.6 Hz, 2H), 2.08 – 2.01 (m, 2H), 1.35 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.2, 173.4, 154.5, 146.5, 130.1 (q, *J* = 32.4 Hz), 126.4, 124.7, 123.8 (q, *J* = 270.4 Hz), 120.1 (q, *J* = 3.9 Hz), 114.6, 107.4 (q, *J* = 3.7 Hz), 60.2, 41.7, 38.0, 24.9, 23.0, 20.3. M.P.: 164.0 – 164.5 °C. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{18}H_{18}F_3N_2O_2^+$ : 351.1315; found: 351.1318.

**Product 5j**



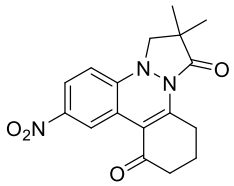
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5j** as a yellow solid (30.3 mg, 51% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 7.84 (s, 1H), 6.91 (d, *J* = 8.0 Hz, 1H), 6.39 (d, *J* = 8.1 Hz, 1H), 3.33 (s, 2H), 3.20 (t, *J* = 6.1 Hz, 2H), 2.55 – 2.49 (m, 2H), 2.25 (s, 3H), 2.05 (s, 2H), 1.32 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.8, 173.6, 153.0, 143.8, 132.6, 128.6, 127.1, 120.9, 116.1, 110.9, 60.6, 41.8, 38.3, 24.9, 23.0, 20.9, 20.5. M.P.: 132.0 – 132.5 °C. HRMS (ESI): m/z [M + Na]$^+$ calcd for $C_{18}H_{20}N_2O_2Na^+$ : 319.1417; found: 319.1418.

**Product 5k**



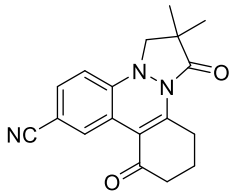
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5k** as a yellow solid (39.8 mg, 68% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.35 (s, 1H), 7.34 (d, *J* = 7.7 Hz, 1H), 6.48 (d, *J* = 8.4 Hz, 1H), 3.37 (s, 2H), 3.20 (t, *J* = 6.2 Hz, 2H), 2.56 – 2.50 (m, 2H), 2.05 (p, *J* = 6.3 Hz, 2H), 1.34 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.2, 173.2, 153.9, 148.8, 125.6 (q, *J* = 4.0 Hz), 124.9 (q, *J* = 32.4 Hz), 124.2 (q, *J* = 269.9 Hz), 123.3 (q, *J* = 4.0 Hz), 121.5, 114.5, 110.5, 60.1, 41.6, 38.1, 24.9, 23.0, 20.3. M.P.: 122.0 – 122.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{18}H_{18}F_3N_2O_2^+$ : 351.1315; found: 351.1317.

**Product 5l**



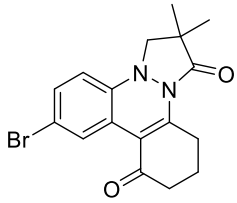
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5l** as a yellow solid (30.9 mg, 47% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.93 (s, 1H), 7.95 (d, *J* = 8.8 Hz, 1H), 6.39 (d, *J* = 8.9 Hz, 1H), 3.41 (s, 2H), 3.20 (t, *J* = 6.2 Hz, 2H), 2.54 (t, *J* = 6.7 Hz, 2H), 2.09 – 2.02 (m, 2H), 1.36 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 194.8, 172.8, 154.0, 151.1, 142.9, 124.9, 121.5, 121.2, 113.3, 109.9, 59.6, 41.5, 37.9, 25.0, 23.1, 20.2. M.P.: 108.0 – 108.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{18}N_3O_4^+$ : 328.1292; found: 328.1291.

**Product 5m**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5m** as a yellow solid (25 mg, 57% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.37 (s, 1H), 7.35 (d, *J* = 8.3 Hz, 1H), 6.42 (d, *J* = 8.3 Hz, 1H), 3.36 (s, 2H), 3.19 (t, *J* = 6.2 Hz, 2H), 2.58 – 2.48 (m, 2H), 2.09 – 1.99 (m, 2H), 1.34 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.0, 172.9, 154.2, 149.5, 133.1, 129.3, 121.7, 119.1, 113.7, 110.8, 106.0, 59.6, 41.5, 37.9, 25.0, 23.0, 20.2. M.P.: 130.0 – 130.5 ºC. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{18}H_{17}N_3NaO_2^+$ : 330.1213; found: 330.1217.

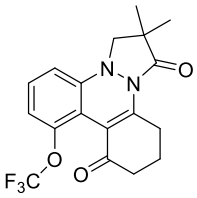
**Product 5n**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5n** as a yellow solid (36.8 mg, 63% yield).

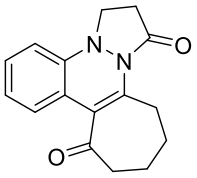
$^1$H NMR (400 MHz, $CDCl_3$) δ = 8.20 (s, 1H), 7.19 (d, *J* = 8.5 Hz, 1H), 6.31 (d, *J* = 8.5 Hz, 1H), 3.31 (s, 2H), 3.20 (t, *J* = 6.2 Hz, 2H), 2.55 – 2.47 (m, 2H), 2.03 (p, *J* = 6.3 Hz, 2H), 1.33 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.2, 173.4, 153.9, 145.0, 130.9, 129.0, 123.0, 116.2, 114.6, 112.3, 60.3, 41.7, 38.1, 24.9, 23.0, 20.3. M.P.: 126.0 – 126.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{17}H_{18}BrN_2O_2^+$ : 361.0546; found: 361.0549.

**Product 5o**



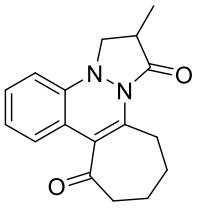
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5o** as a kelly solid (33.8 mg, 46% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 7.17 (t, *J* = 8.2 Hz, 1H), 6.89 (d, *J* = 8.4 Hz, 1H), 6.47 (d, *J* = 8.0 Hz, 1H), 3.34 (s, 2H), 3.18 (t, *J* = 6.2 Hz, 2H), 2.53 (t, *J* = 6.7 Hz, 2H), 2.06 – 1.99 (m, 2H), 1.33 (s, 6H) $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 192.6, 173.8, 154.7, 150.4, 144.2, 129.3, 120.4 (d, *J* = 256.4 Hz), 117.2, 116.6, 114.9, 109.5, 60.2, 42.0, 37.0, 24.2, 23.0, 19.8. M.P.: 162.0 – 162.5 ºC. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{18}H_{17}F_3N_2O_3Na^+$ : 389.1083; found: 389.1080.

**Product 5p**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5p** as a yellow solid (18 mg, 33% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 7.35 (d, *J* = 7.8 Hz, 1H), 7.11 (t, *J* = 8.4 Hz, 1H), 6.89 (t, *J* = 7.7 Hz, 1H), 6.54 (d, *J* = 7.6 Hz, 1H), 3.69 (t, *J* = 8.4 Hz, 2H), 3.08 (t, *J* = 6.0 Hz, 2H), 2.78 (t, *J* = 8.4 Hz, 2H), 2.66 (t, *J* = 6.1 Hz, 2H), 1.93 – 1.85 (m, 4H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 204.4, 167.6, 147.1, 145.8, 128.8, 124.9, 123.6, 122.9, 122.2, 111.1, 46.8, 42.4, 31.7, 26.0, 23.1, 21.1. M.P.: 147.0 – 147.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{16}H_{17}N_2O_2^+$ : 269.1285; found: 269.1284.

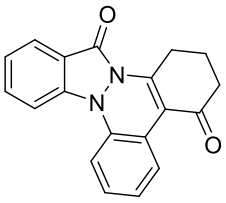
**Product 5q**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5q** as a yellow solid (22 mg, 39% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 7.33 (d, *J* = 7.0 Hz, 1H), 7.10 (t, *J* = 7.2 Hz, 1H), 6.88 (t, *J* = 7.6 Hz, 1H), 6.53 (d, *J* = 8.0 Hz, 1H), 3.99 (t, *J* = 9.2 Hz, 1H), 3.18 – 3.07 (m, 2H), 3.06 – 2.84 (m, 2H), 2.71 – 2.58 (m, 2H), 1.96 – 1.83 (m, 4H), 1.33 (d, *J* = 7.0 Hz, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 204.5, 170.2, 147.1, 145.7, 128.7, 124.9, 123.3, 122.9, 122.1, 111.2, 54.5, 42.4, 37.1, 25.8, 23.0, 21.0,

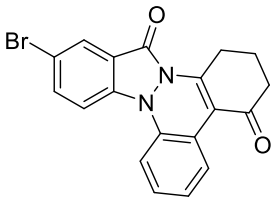
13.8. M.P.: 126.0 – 126.5 °C. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{17}H_{18}N_2O_2Na^+$ : 305.1260; found: 305.1269.

**Product 5r**



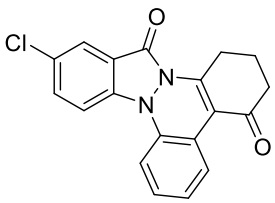
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5r** as a yellow solid (27.9 mg, 91% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.40 (d, *J* = 7.5 Hz, 1H), 7.97 (d, *J* = 7.9 Hz, 1H), 7.75 – 7.66 (m, 2H), 7.52 (d, *J* = 8.0 Hz, 1H), 7.30 (t, *J* = 7.2 Hz, 1H), 7.23 (t, *J* = 7.3 Hz, 1H), 7.19 – 7.13 (m, 1H), 3.61 (t, *J* = 6.1 Hz, 2H), 2.64 – 2.55 (m, 2H), 2.17 – 2.11 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.6, 158.7, 152.5, 139.7, 136.4, 133.6, 128.2, 127.7, 124.8, 124.6, 122.7, 120.9, 117.2, 113.5, 113.4, 111.1, 38.3, 24.6, 20.4. M.P.: 174.0 – 174.5 °C. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{19}H_{15}N_2O_2^+$ : 303.1128; found: 303.1132.

**Product 5s**



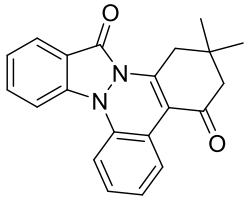
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5s** as a yellow solid (46.7 mg, 61% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.43 (dd, *J* = 7.9, 1.3 Hz, 1H), 8.12 (d, *J* = 2.0 Hz, 1H), 7.82 (d, *J* = 8.9 Hz, 1H), 7.65 (d, *J* = 8.8 Hz, 1H), 7.49 (d, *J* = 8.0 Hz, 1H), 7.29 (d, *J* = 7.6 Hz, 1H), 7.21 (d, *J* = 7.6 Hz, 1H), 3.63 (t, *J* = 6.2 Hz, 2H), 2.62 (dd, *J* = 7.5, 5.8 Hz, 2H), 2.20 – 2.14 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 195.6, 157.4, 152.1, 138.3, 136.6, 136.1, 128.5, 128.0, 127.4, 125.2, 120.9, 118.9, 115.5, 114.9, 113.8, 111.2, 38.4, 24.7, 20.5. M.P.: 261.0 – 261.5. °C. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{19}H_{14}BrN_2O_2^+$ : 381.0233; found: 381.0227.

**Product 5t**



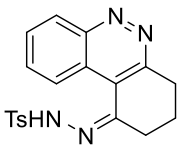
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5t** as a yellow solid (45.4 mg, 68% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.43 (d, *J* = 9.2 Hz, 1H), 7.95 (s, 1H), 7.70 (s, 2H), 7.49 (d, *J* = 8.1 Hz, 1H), 7.29 (d, *J* = 9.3 Hz, 1H), 7.21 (t, *J* = 7.9 Hz, 1H), 3.63 (t, *J* = 6.2 Hz, 2H), 2.61 (d, *J* = 6.1 Hz, 2H), 2.24 – 2.10 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ= 195.6, 157.6, 152.1, 138.0, 136.2, 134.0, 128.5, 128.4, 128.0, 125.2, 124.2, 120.9, 118.5, 114.7, 113.8, 111.2, 38.4, 24.7, 20.5. M.P.: 221.0 – 221.5 °C. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{19}H_{14}ClN_2O_2^+$ : 337.0738; found: 337.0749.

**Product 5u**



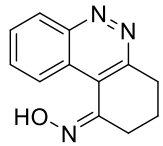
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **5u** as a yellow solid (46.3 mg, 70% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.45 (d, *J* = 7.9 Hz, 1H), 7.99 (d, *J* = 7.8 Hz, 1H), 7.74 (s, 2H), 7.57 (d, *J* = 8.0 Hz, 1H), 7.35 – 7.31 (m, 1H), 7.26 (t, *J* = 7.7 Hz, 1H), 7.21 – 7.15 (m, 1H), 3.49 (s, 2H), 2.49 (s, 2H), 1.20 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ= 195.9, 158.7, 150.7, 139.9, 136.5, 133.6, 128.3, 127.5, 124.9, 124.7, 122.8, 120.8, 117.4, 113.5, 112.6, 111.1, 52.1, 38.1, 32.4, 28.3. M.P.: 185.0 – 185.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{21}H_{19}N_2O_2^+$ : 331.1441; found: 331.1446.

**Product 6**



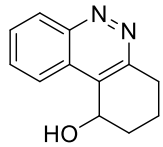
The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 2:1), yielding **6** as a yellow solid (52.3 mg, 71% yield). $^1$H NMR (400 MHz, $CDCl_3$) δ = 8.80 (d, *J* = 8.6 Hz, 1H), 8.42 (d, *J* = 8.4 Hz, 1H), 7.85 (d, *J* = 8.2 Hz, 2H), 7.75 – 7.61 (m, 2H), 7.24 (d, *J* = 8.1 Hz, 2H), 3.42 – 3.18 (m, 2H), 2.66 (t, *J* = 6.6 Hz, 2H), 2.34 (s, 3H), 2.03 – 1.91 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) δ = 154.3, 150.7, 150.2, 144.7, 135.1, 132.2, 130.5, 129.8, 129.3, 128.3, 126.3, 122.3, 122.0, 30.6, 26.4, 21.6, 20.3. M.P.: 133.0 – 133.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{19}H_{19}N_4O_2S^+$ : 367.1223; found: 367.1224.

**Product 7**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to1:1), yielding **7** as a white solid (29.7 mg, 70% yield). $^1$H NMR (400 MHz, DMSO) δ = 12.26 (d, *J* = 4.0 Hz, 1H), 9.02 (s, 1H), 8.46 (s, 1H), 7.88 (s, 2H), 3.36 – 3.29 (m, 2H), 2.89 (s, 2H), 1.95 (s, 2H). $^{13}$C NMR (100 MHz, DMSO) δ= 154.2, 153.5, 149.9, 132.1, 129.9, 129.6, 126.8, 121.7, 121.4, 30.6, 24.3, 20.0. M.P.: 133.0 – 133.5 ºC. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{19}H_{19}N_4O_2S^+$ : 367.1223; found: 367.1224.

**Product 8**



The crude product was purified by column chromatography ($SiO_2$, petroleum ether/ethyl acetate = 10:1 to 1:1), yielding **8** as a white solid (23.7 mg, 59% yield). $^1$H NMR (400 MHz, DMSO) δ = 8.46 – 8.36 (m, 1H), 8.35 – 8.26 (m, 1H), 7.86 – 7.83 (m, 2H), 5.61 (d, *J* = 6.7 Hz, 1H), 5.33 (s, 1H), 3.35 (s, 1H), 3.18 – 3.09 (m, 1H), 2.12 – 2.01 (m, 2H), 1.91 - 1.86 (m, 2H). $^{13}$C

NMR (100 MHz, DMSO) δ = 152.4, 149.1, 130.9, 129.4, 129.4, 129.3, 124.8, 124.1, 61.3, 31.2, 29.9, 17.0. M.P.: 134.0 – 134.5 $^{o}$C. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{12}H_{13}N_2O^+$ : 201.1022; found: 201.1022.

**References:**

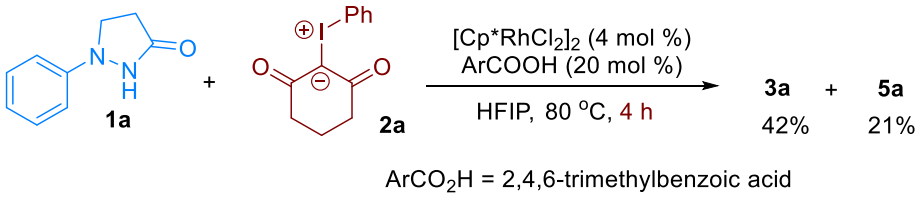

[1] Hu, S.; Han, X.; Xie, X.; Fang, F.; Synthesis of Pyrazolo[1,2-a]cinnolines via Rhodium(III)-Catalyzed [4+2] Annulation Reactions of Pyrazolidinones with Sulfoxonium Ylides. *Adv. Synth. Catal.* **2021**, *13*, 3311-3317.

[2] Liu, J.; Yang, S.; Dong, R.; Jin, Z.; Wang, M. A Convenient One-Pot Synthesis of 1-Aryl-Substituted 4-Iodopyrazol-3-Ols via Aromatisation and Oxidative Iodination Reactions. *J. Chem. Res.* **2018**, *42*, 24-27.

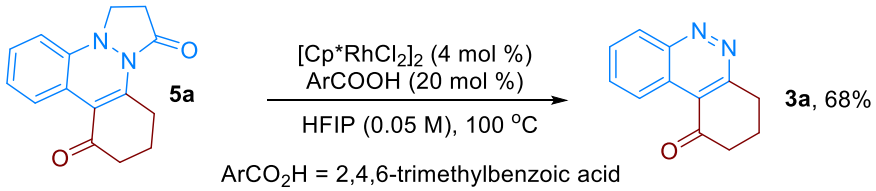
[3] Gogoi, K.; Bora, B. R.; Borah, G.; Sarma, B.; Gogoi, S. Synthesis of Quaternary Carbon-centered Indolo[1,2-a]quinazolinones and Indazolo[1,2-a]indazolones via C-H Functionalization. *Chem. Commun.* **2021**, *57*, 1388-1391.

[4] Jiang, Y.; Li, P.; Zhao, J.; Liu, B.; Li, X. Iodonium Ylides as Carbene Precursors in Rh(III)-Catalyzed C-H Activation. *Org. Lett.* **2020**, *22*, 7475-7479.

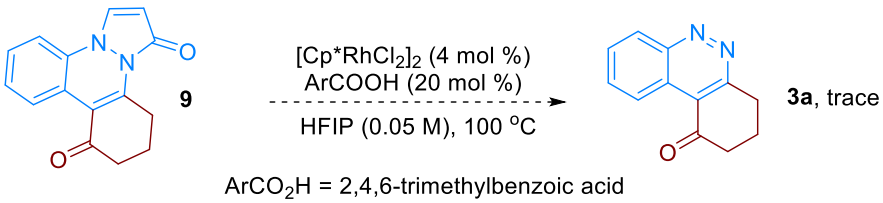
## 5. Mechanistic studies




**1a** (32.4 mg, 0.2 mmol), **2a** (93.9 mg, 0.3 mmol), $[Cp^*RhCl_2]_2$ (4 mol %) and ArCOOH (6.6 mg, 20 mol %) were dissolved in HFIP (2 mL). Then, the mixture was stirred at 80 °C for 4 h by using a heating module as heating source. A mixture of **3a** and **5a** was observed.



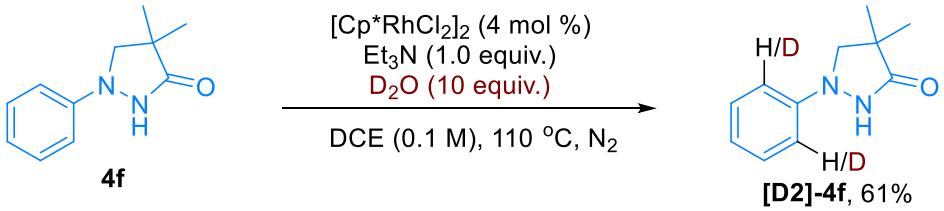

**5a** (17.5 mg), $[Cp^*RhCl_2]_2$ (4 mol %), ArCOOH (20 mol %) were dissolved in HFIP (2 mL). Then, the mixture was stirred at 100 °C for 9 h by using a heating module as heating source. The desired product **3a** can be obtained in 68% yield.



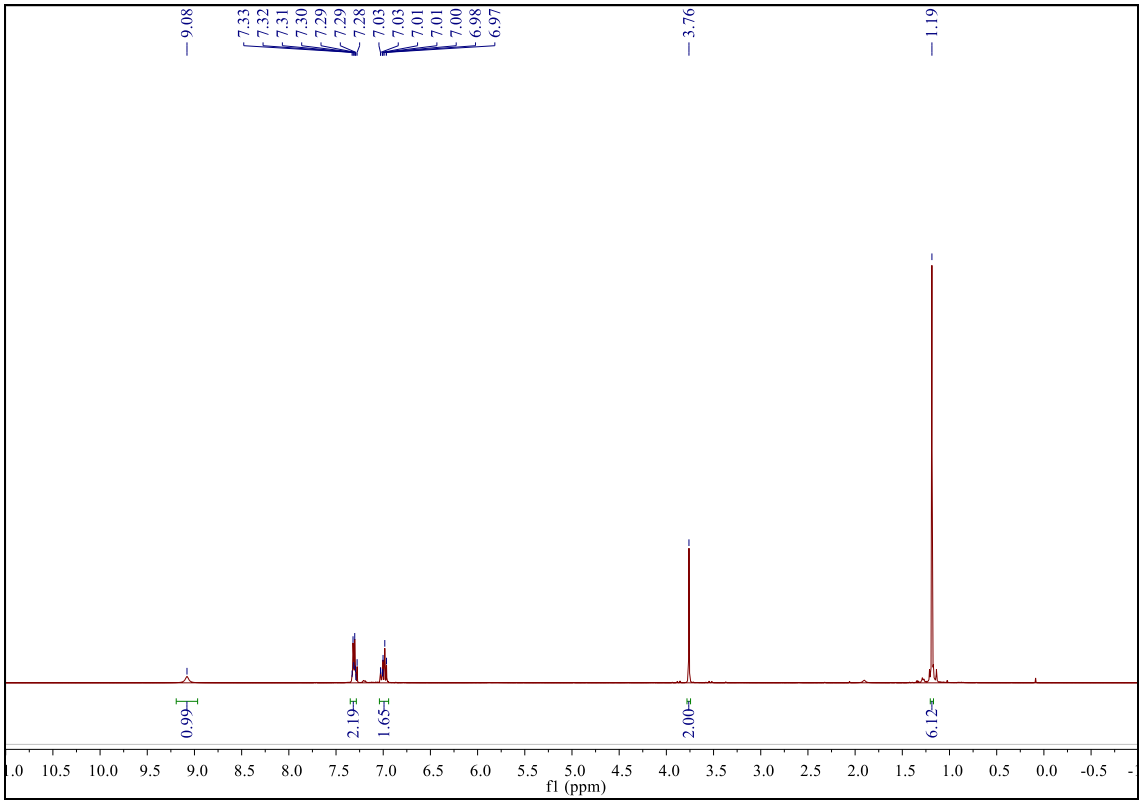

**9** (14.9 mg), $[Cp^*RhCl_2]_2$ (4 mol %), ArCOOH (20 mol %) were dissolved in HFIP (2 mL). Then, the mixture was stirred at 100 °C for 9 h. The expected **3a** can not be detected in this reaction.

***Compound 5a was easily converted into cinnoline 3a under standard conditions. In some case, we can detect the oxidized byproduct 9, which proved to be ineffective for the generation of 3a. These results indicated that pyrazo-lo[1,2-a]cinnoline 5a is the key intermediate for the generation of cinnoline product 3a in HFIP system.***

***H/D exchange experiment***

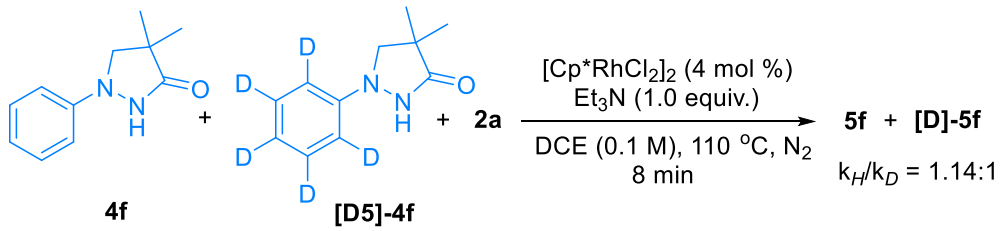



**4f** **[D2]-4f**, 61%




***The reversibility of the C−H activation was determined by running the reaction with 1a in the presence of [Cp*RhCl2]2 and D2O. As a result, ortho-deuteration of 1a (61% D) can be observed, revealing that the C−H activation is reversible.***

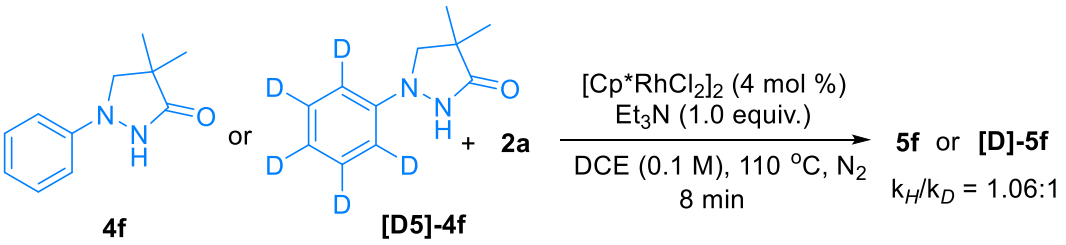
***competitive experiment***




**4f** **[D5]-4f**

**4f** (19.0 mg, 0.1 mmol), **[D]-1a** (19.5 mg, 0.1 mmol), and **2a** (93.9 mg, 0.3 mmol), $[Cp^*RhCl_2]_2$ (4.9 mg, 4 mol %) and $Et_3N$ (27.6 uL, 0.2 mmol) were dissolved in DCE (2.0 mL). Then, the mixture was stirred at 110 °C for 8 min by using a heating module as heating source. Yields determined by GC using tetradecane as the internal standard: $k_H/k_D$ = 1.14:1.

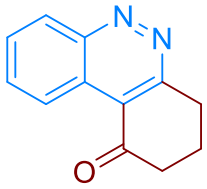
***parallel experiment***




**1a** (19.0 mg, 0.1 mmol) or **[D]-1a** (19.5 mg, 0.1 mmol), and **2a** (93.9 mg, 0.3 mmol), $[Cp^*RhCl_2]_2$ (4.9 mg, 4 mol %) and $Et_3N$ (13.8 uL, 0.1 mmol) were dissolved in DCE (1.0 mL). Then, the mixture was stirred at 110 °C for 8 min by using a heating module as heating source. Yields determined by GC using tetradecane as the internal standard: $k_H/k_D$ = 1.06:1.

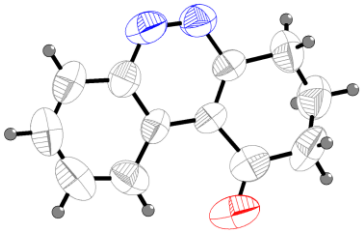
***There are no significant kinetic isotope effects (KIEs) were observed in competitive ($k_H/k_D$ = 1.14:1) and parallel ($k_H/k_D$ = 1.06:1) experiments at the early stage of this reaction (8 min), which indicated the C–H bond activation might not be the rate-determining step for this reaction.***

## 6. X-Ray structures of 3fa and 3g

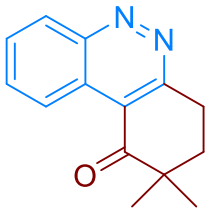



3a

≡

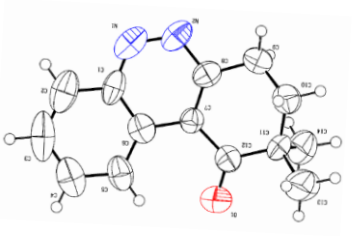


CCDC: 2133553




3g

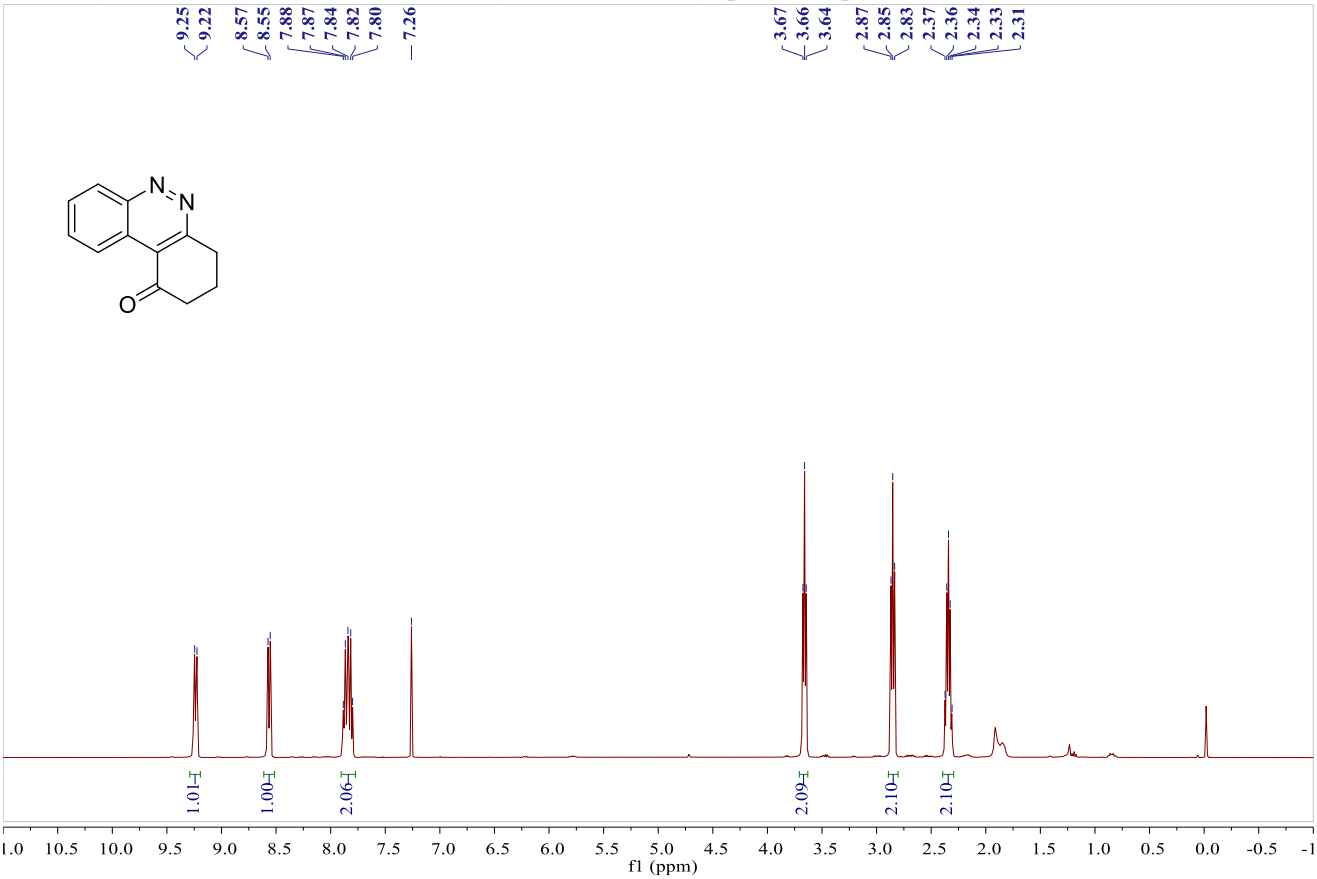
≡

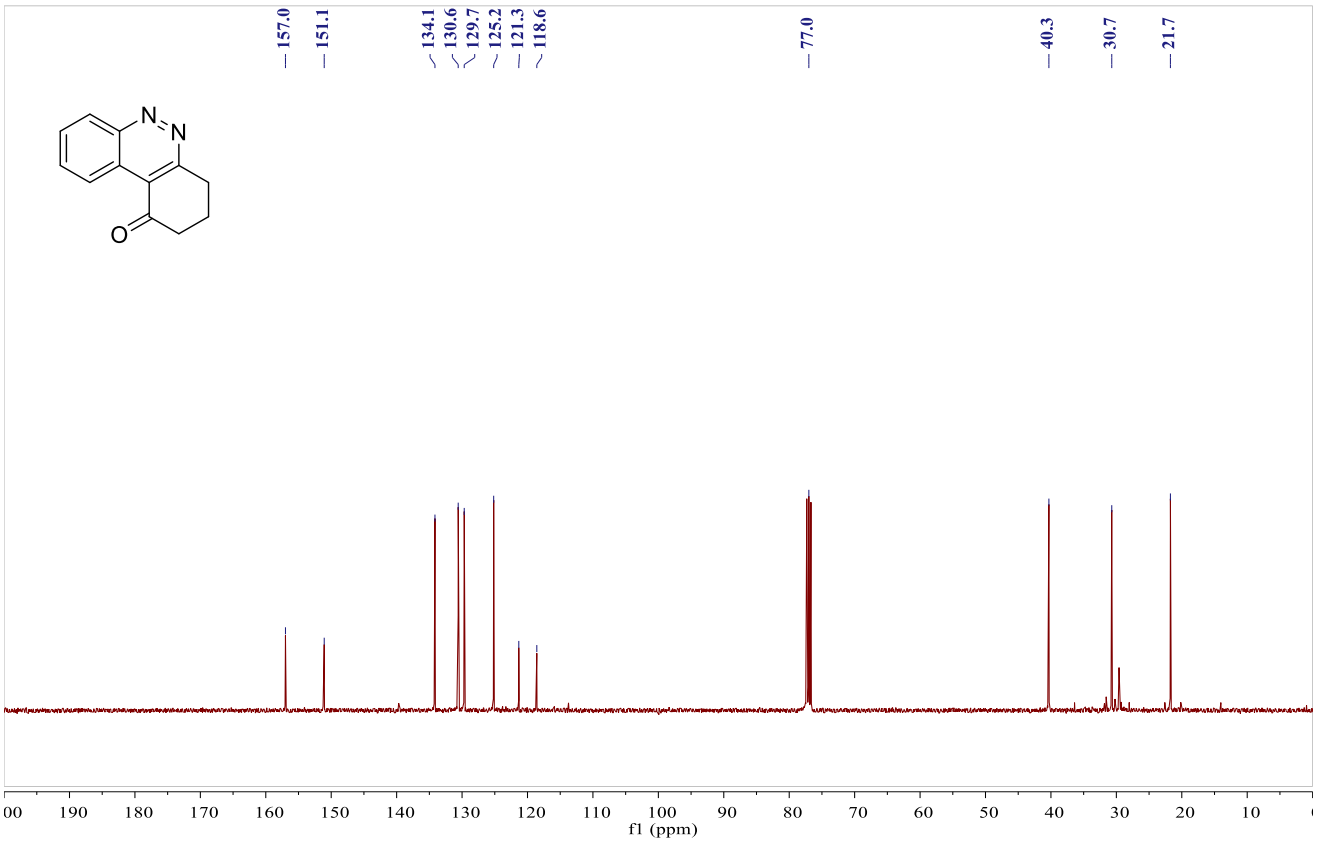


CCDC: 2133551

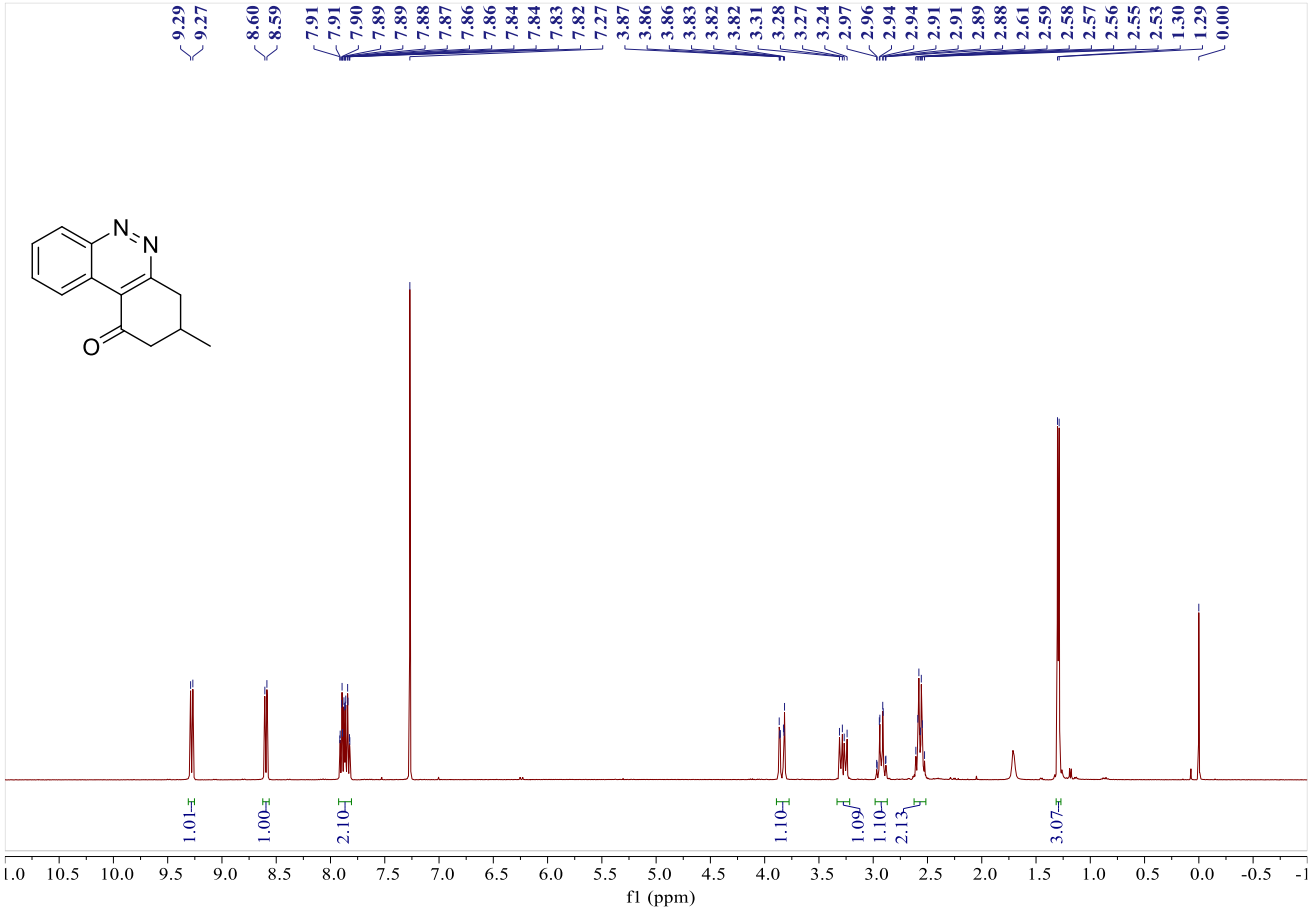
## 7. NMR Spectra of products 3a-3z, 5a-5u, 6, 7 and 8

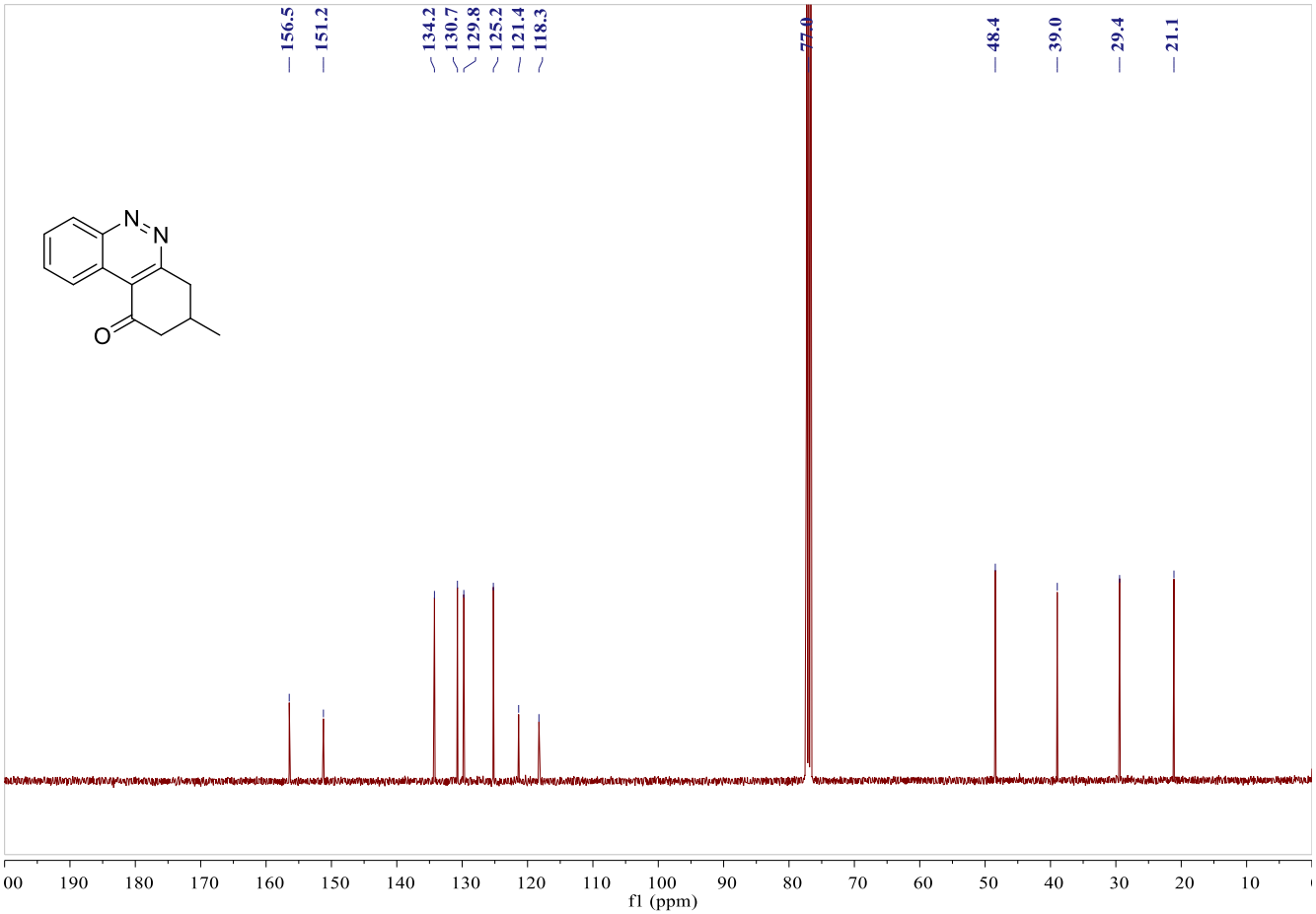
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3a**



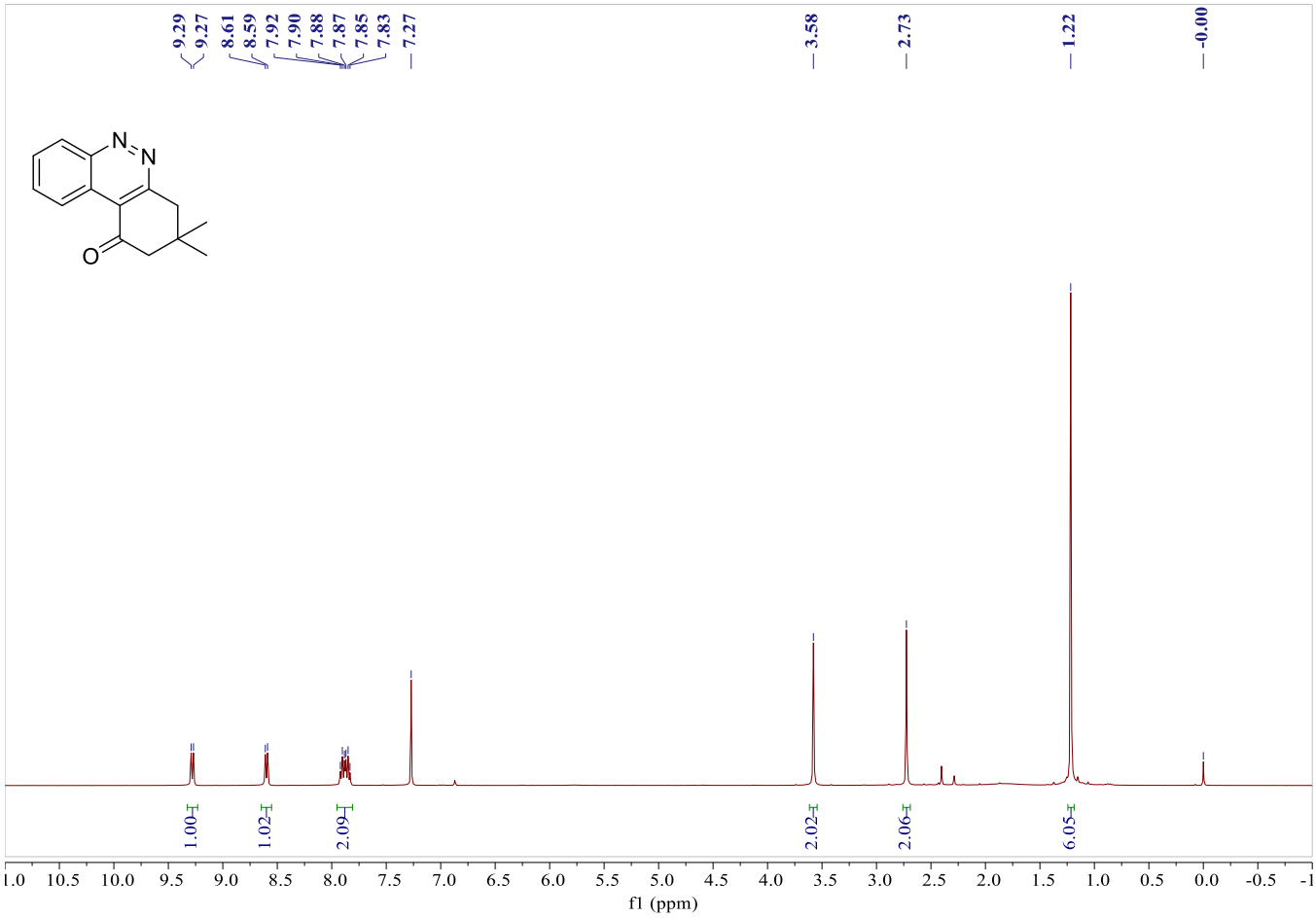



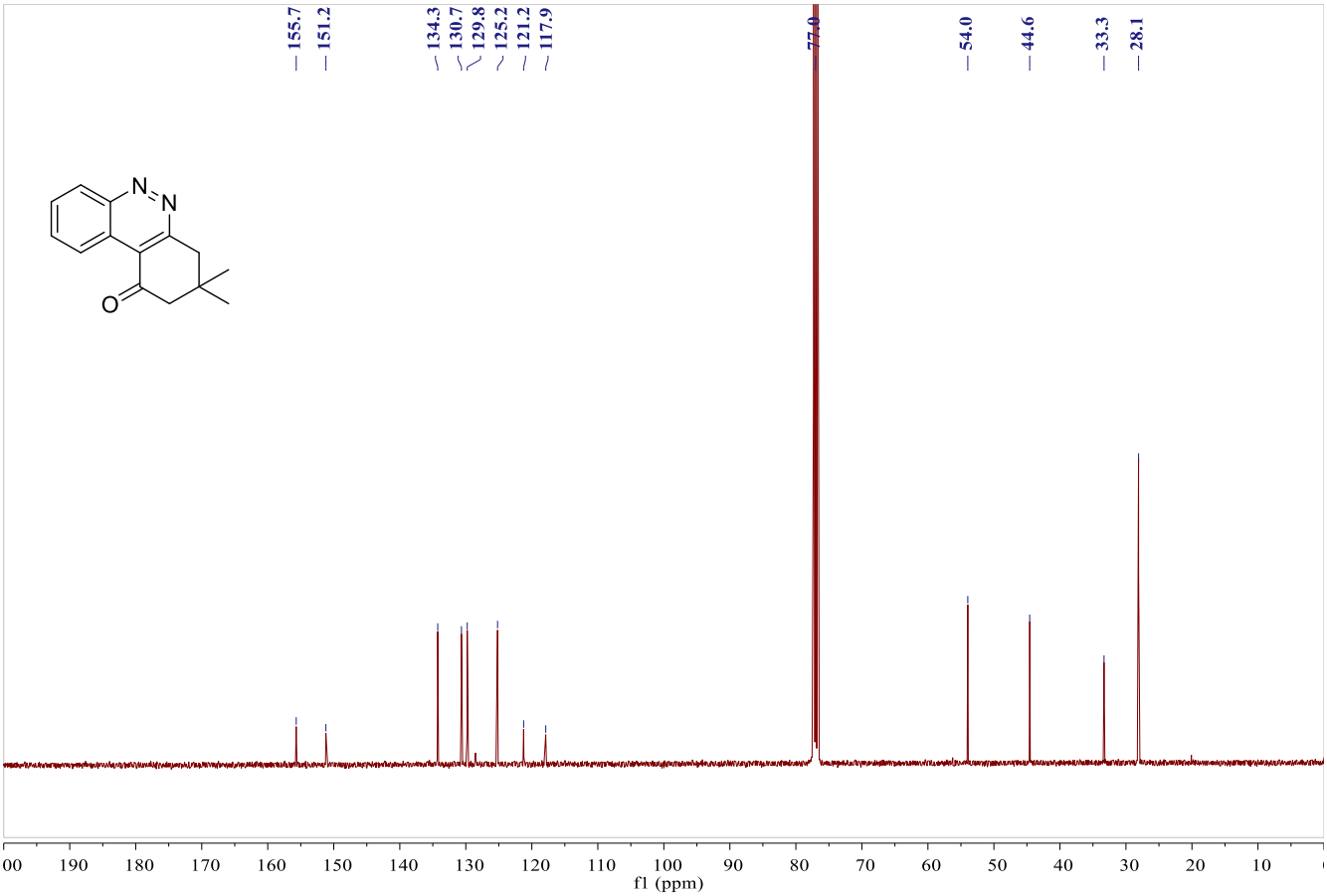
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3b**



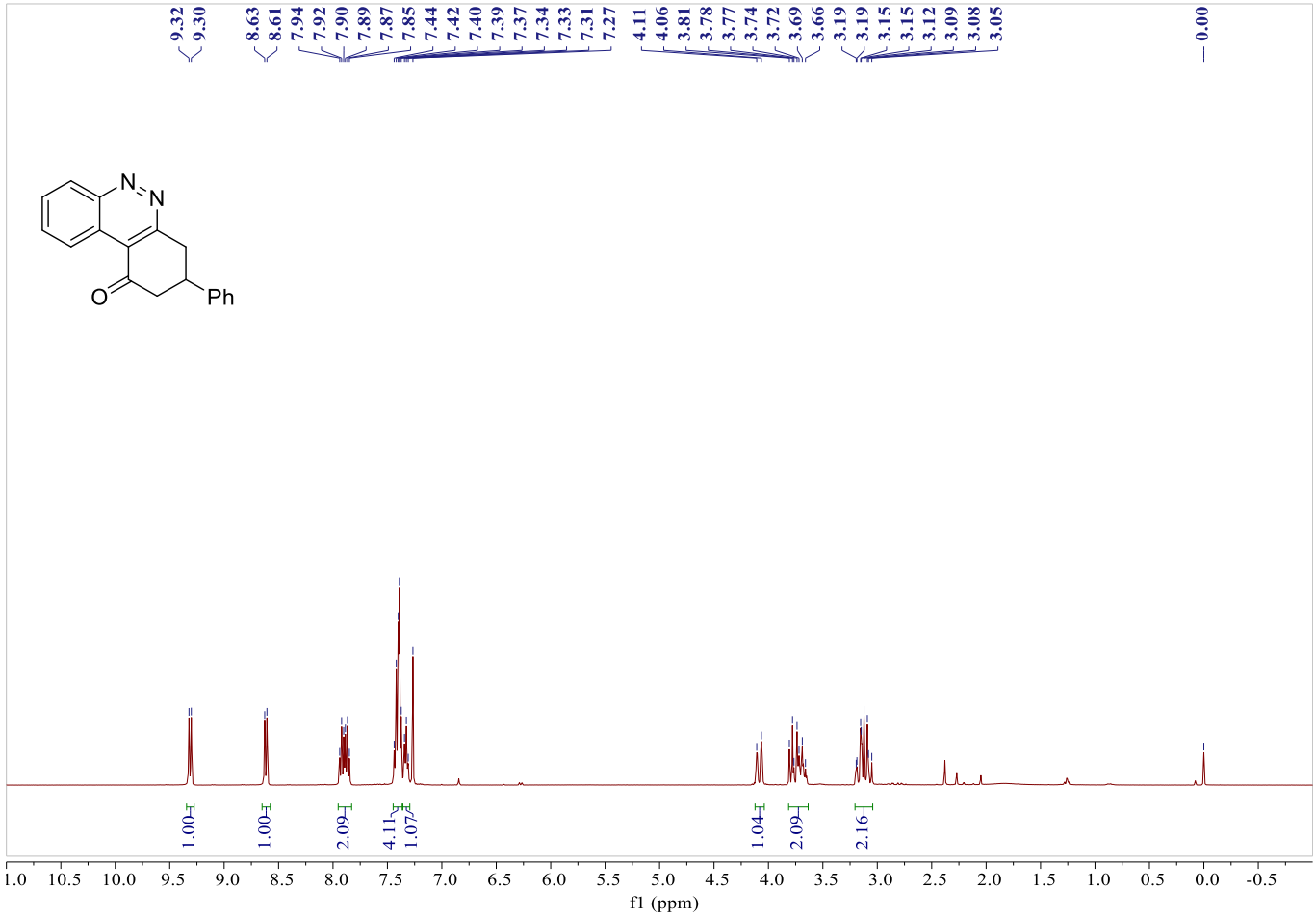



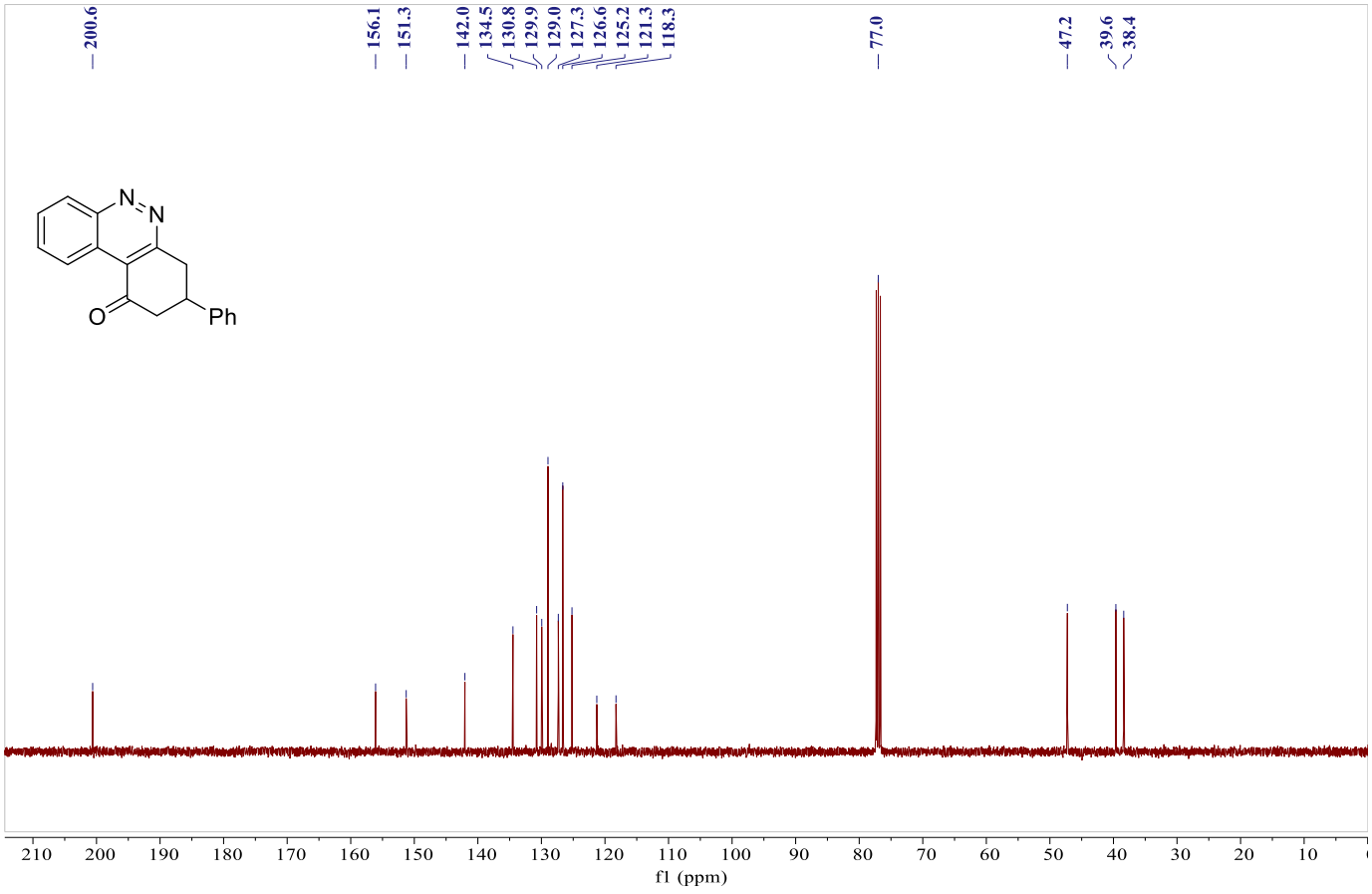
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3c**



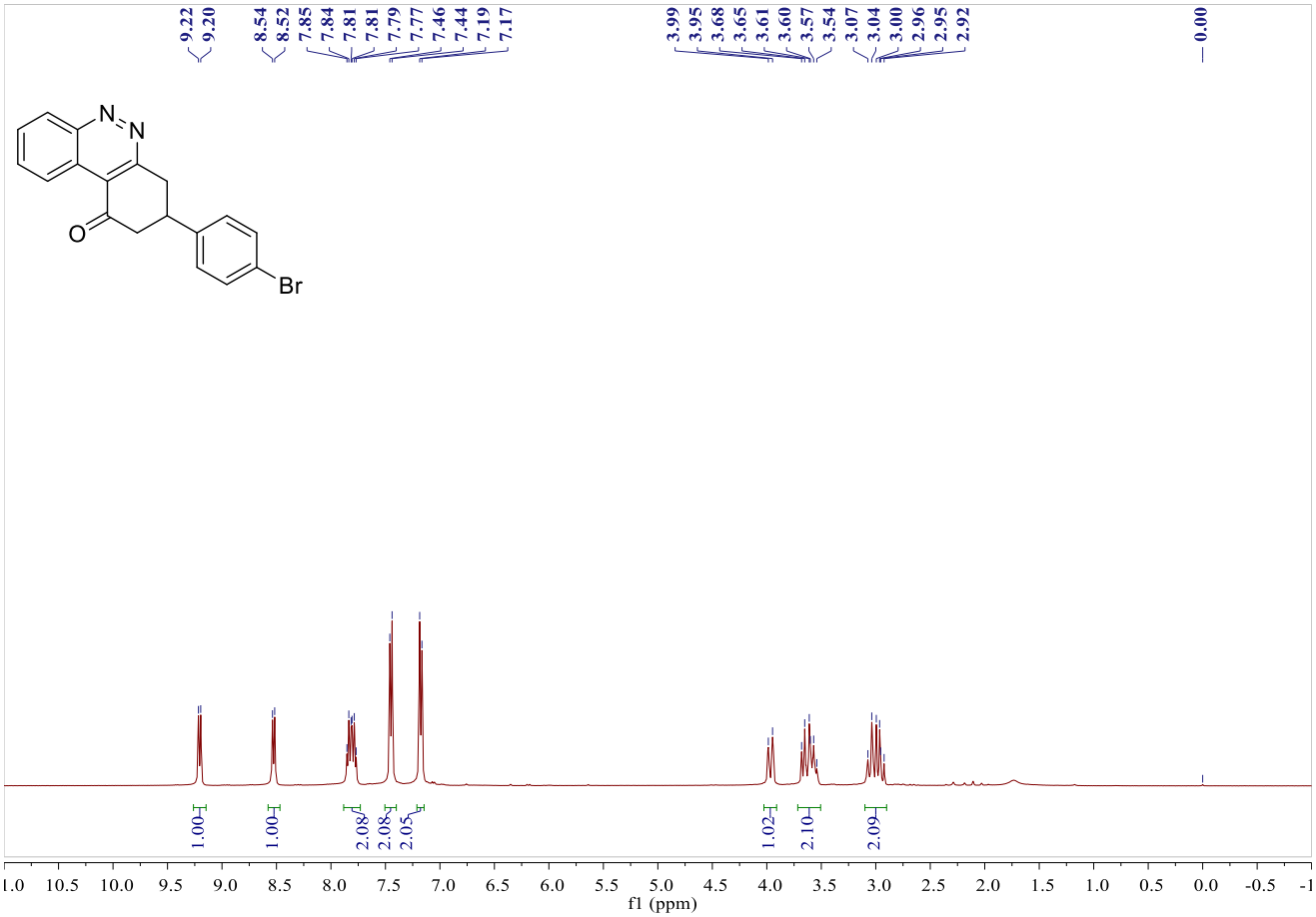



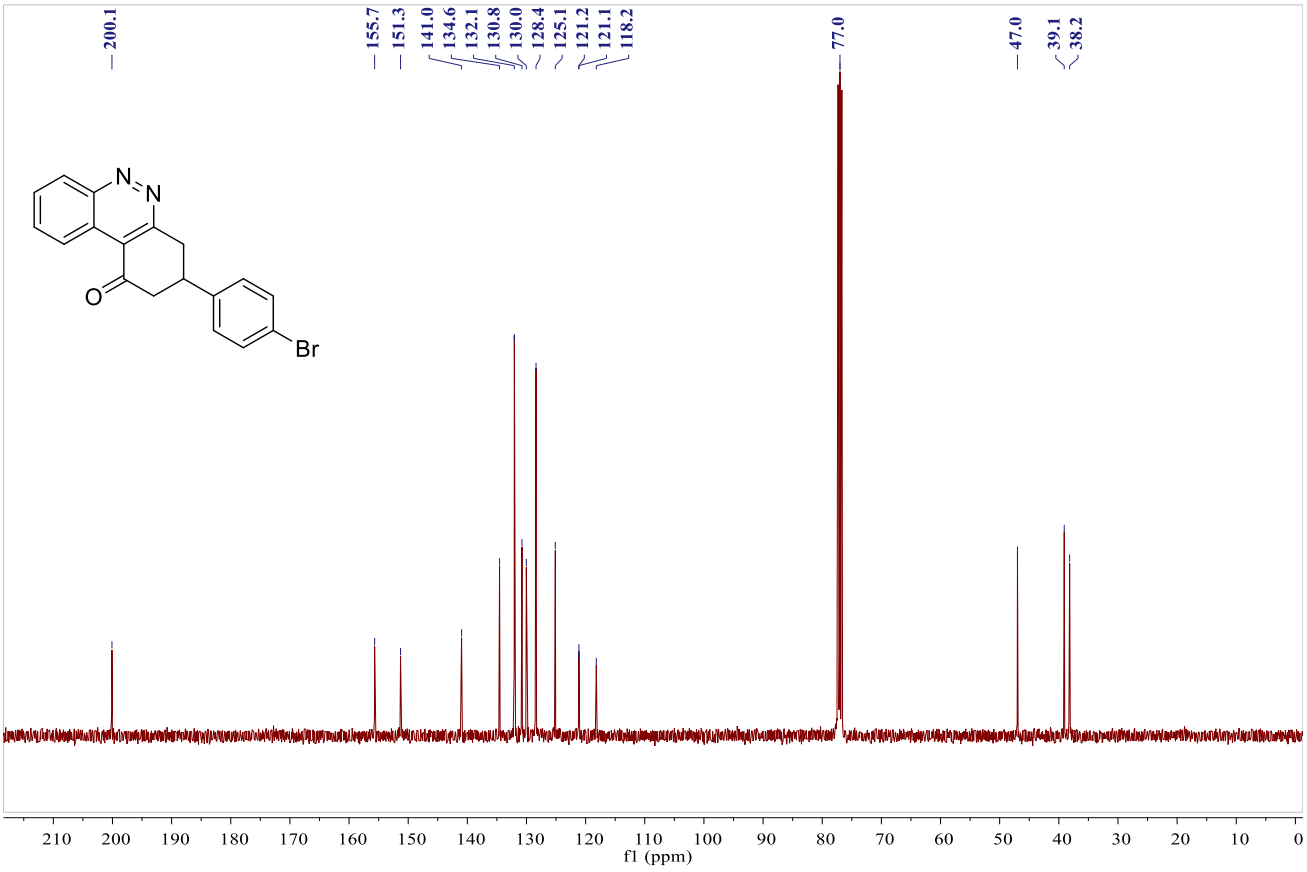
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3d**



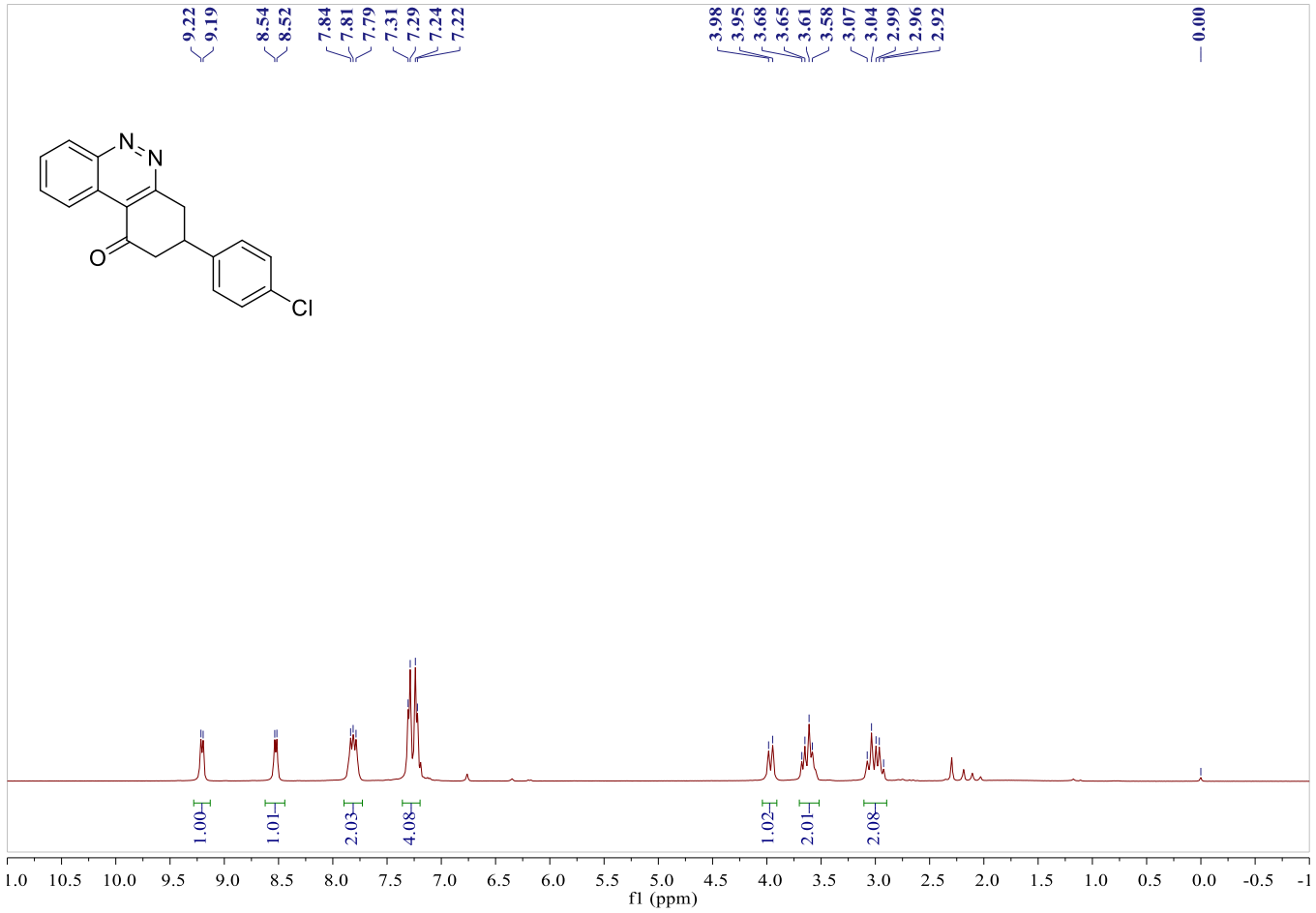



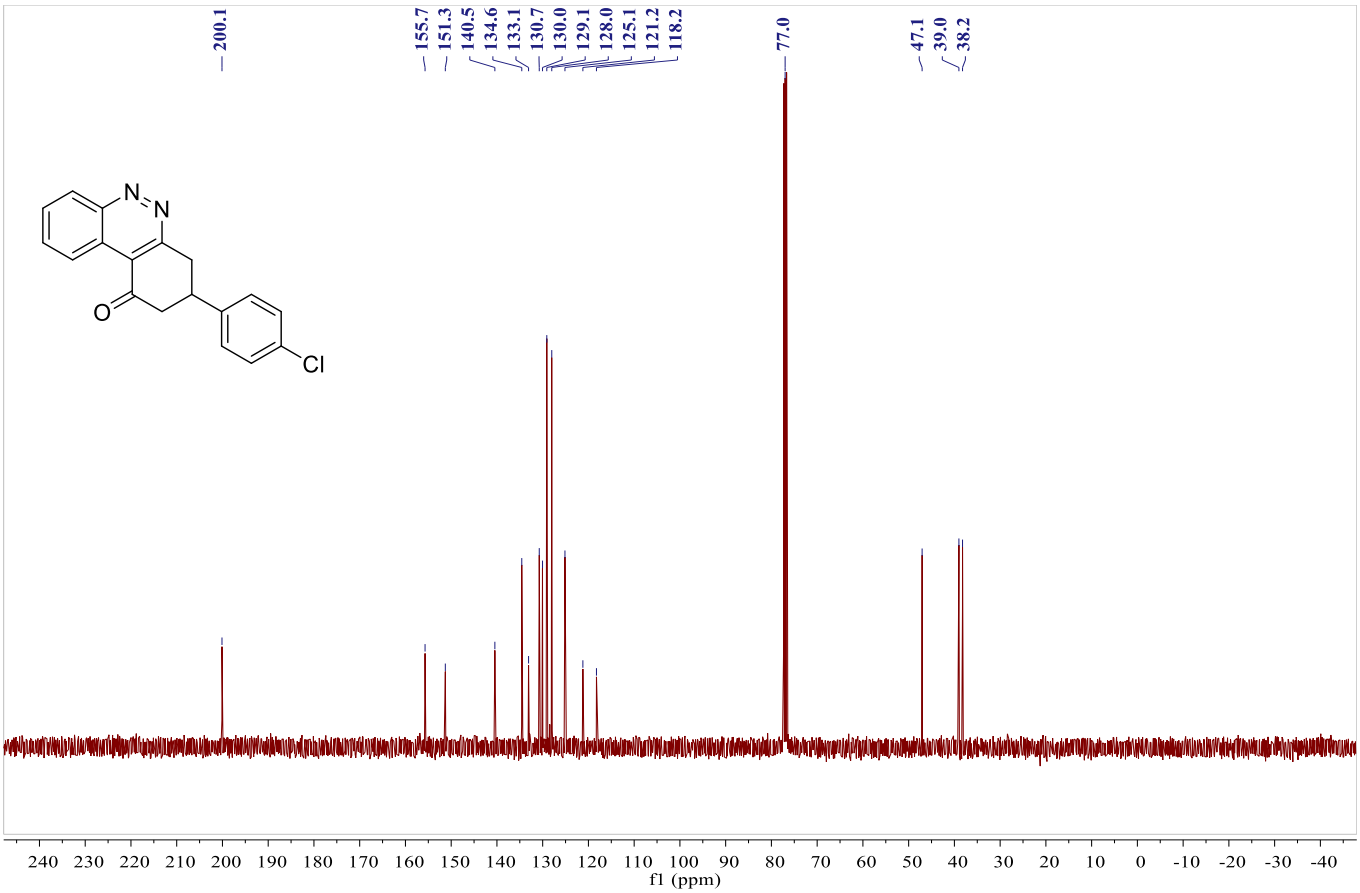
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3e**



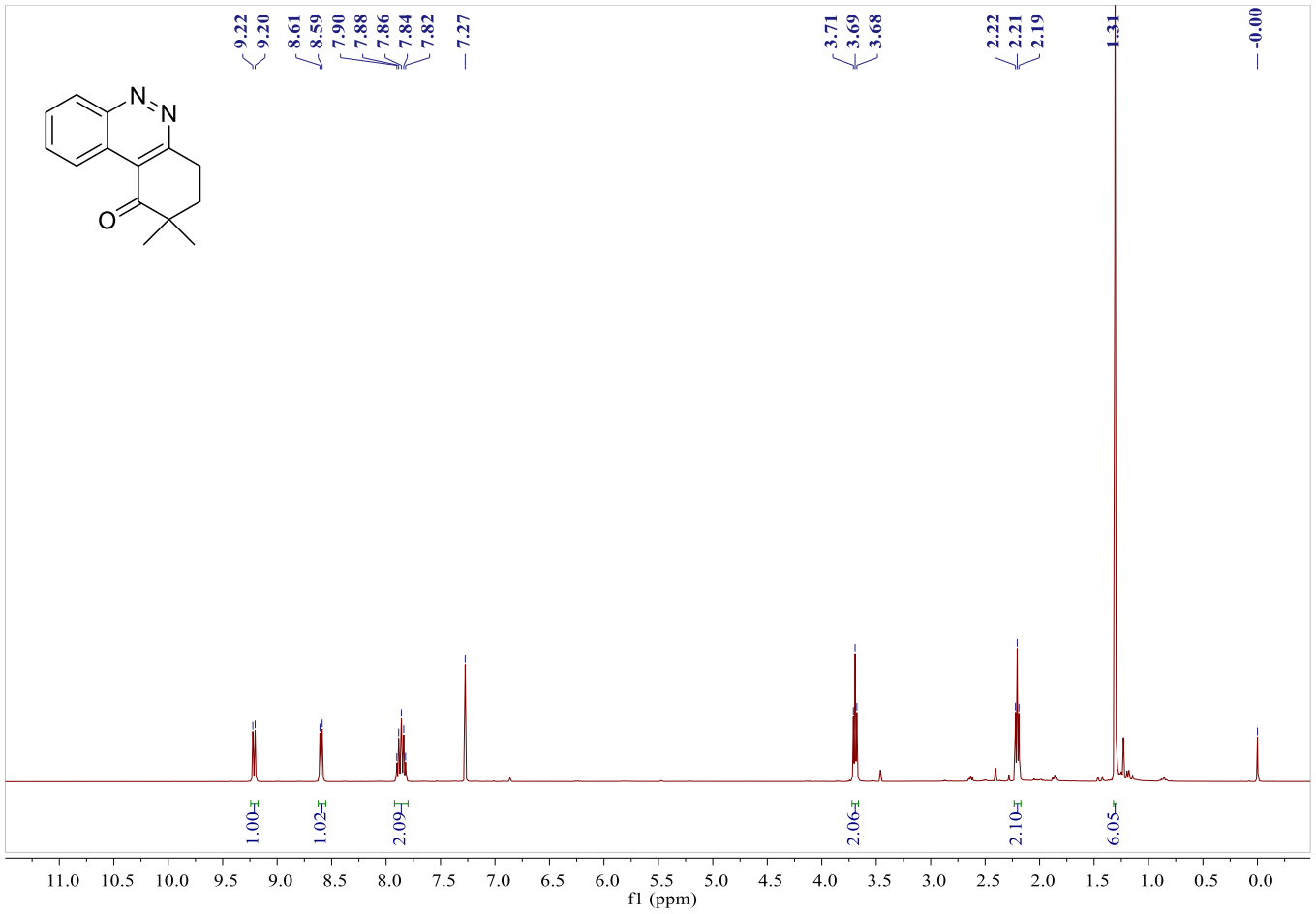



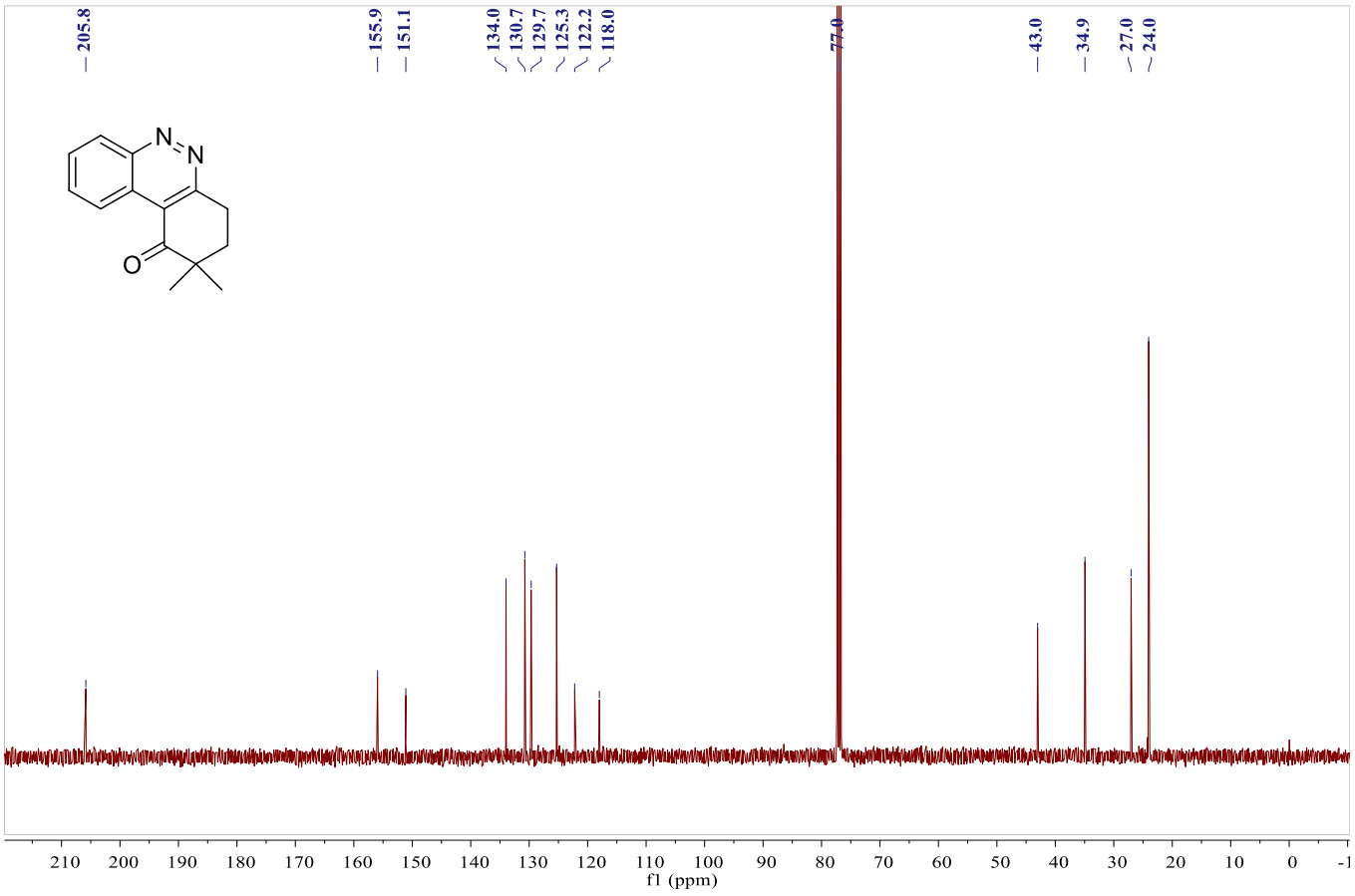
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3f**



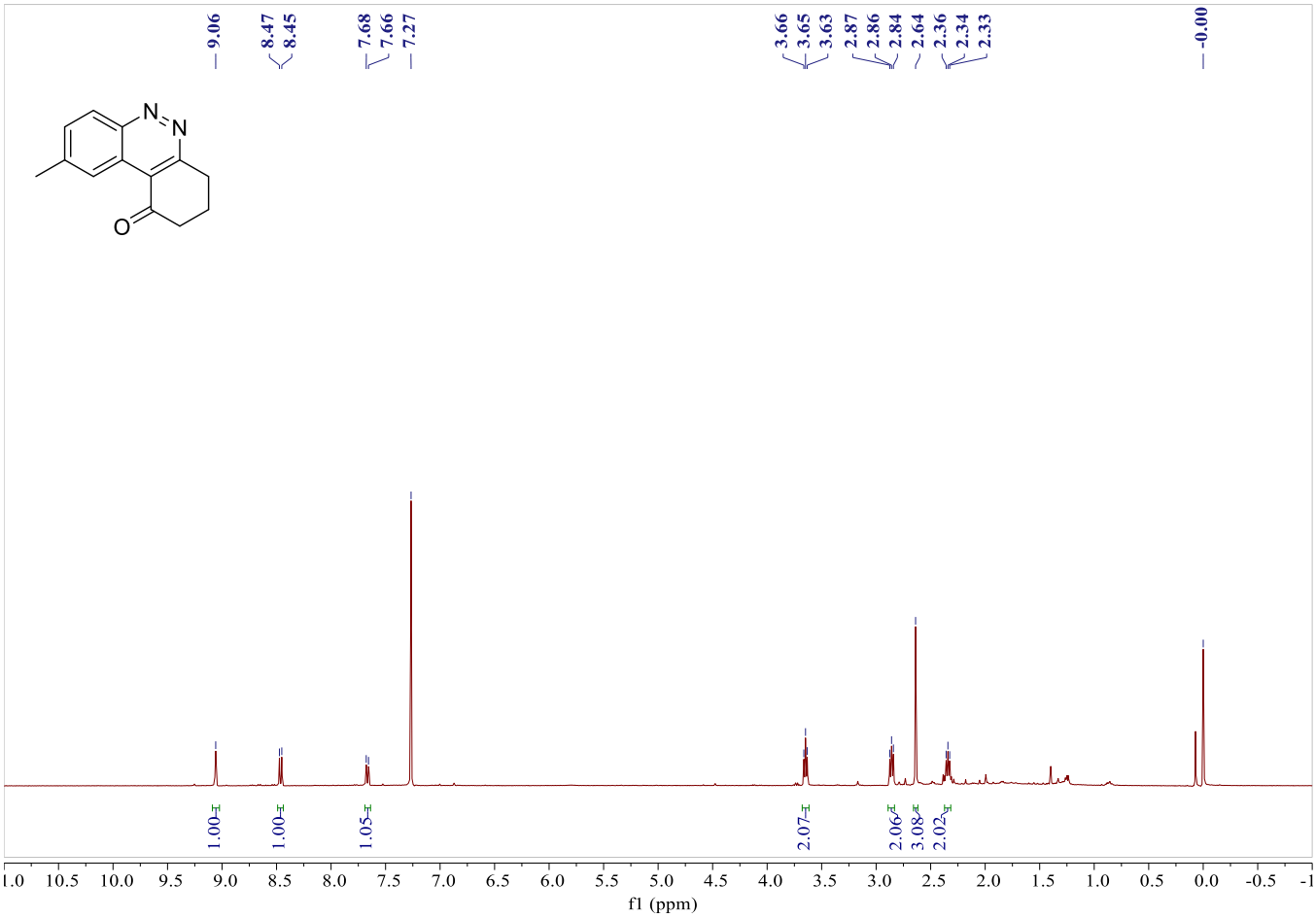



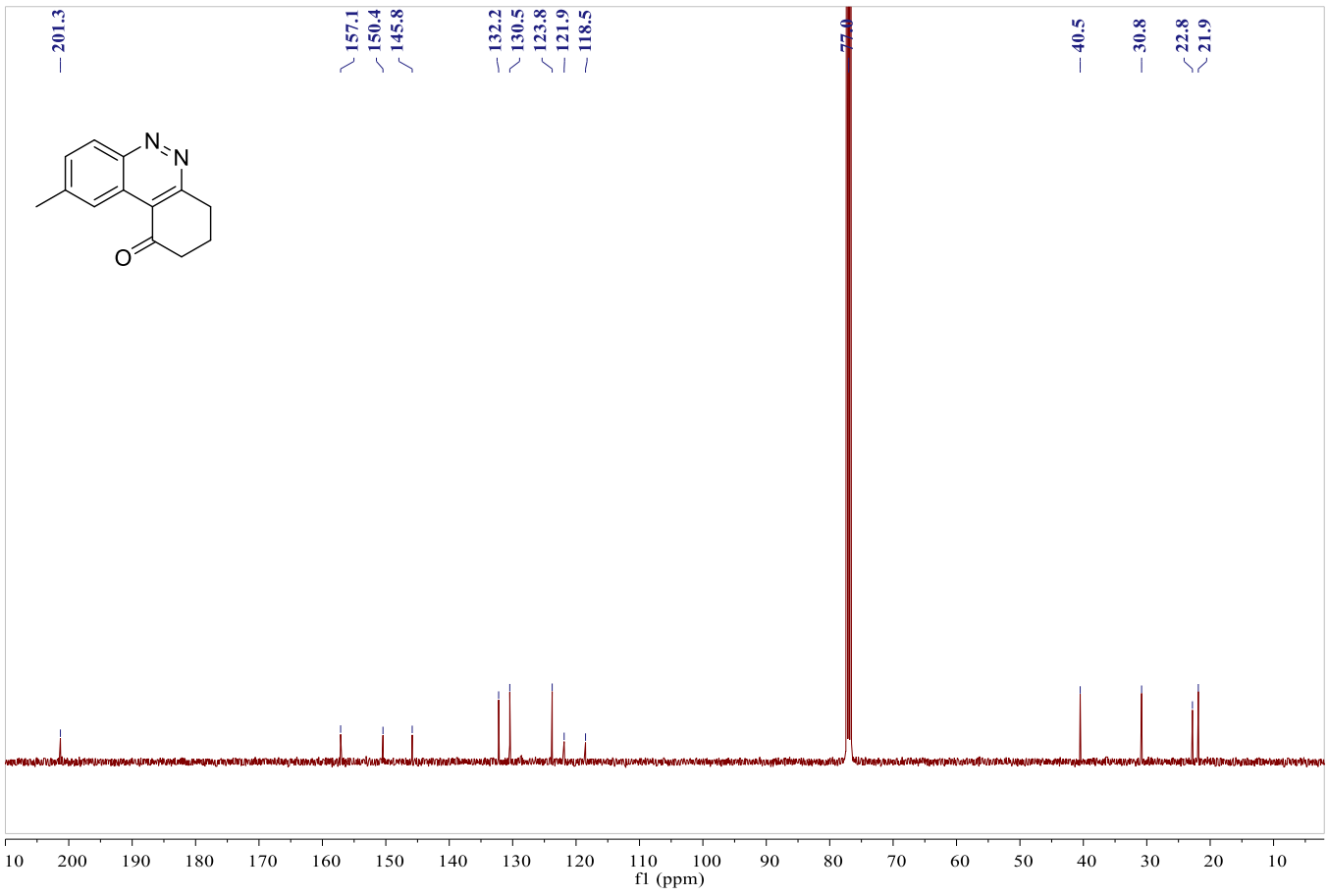
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3g**



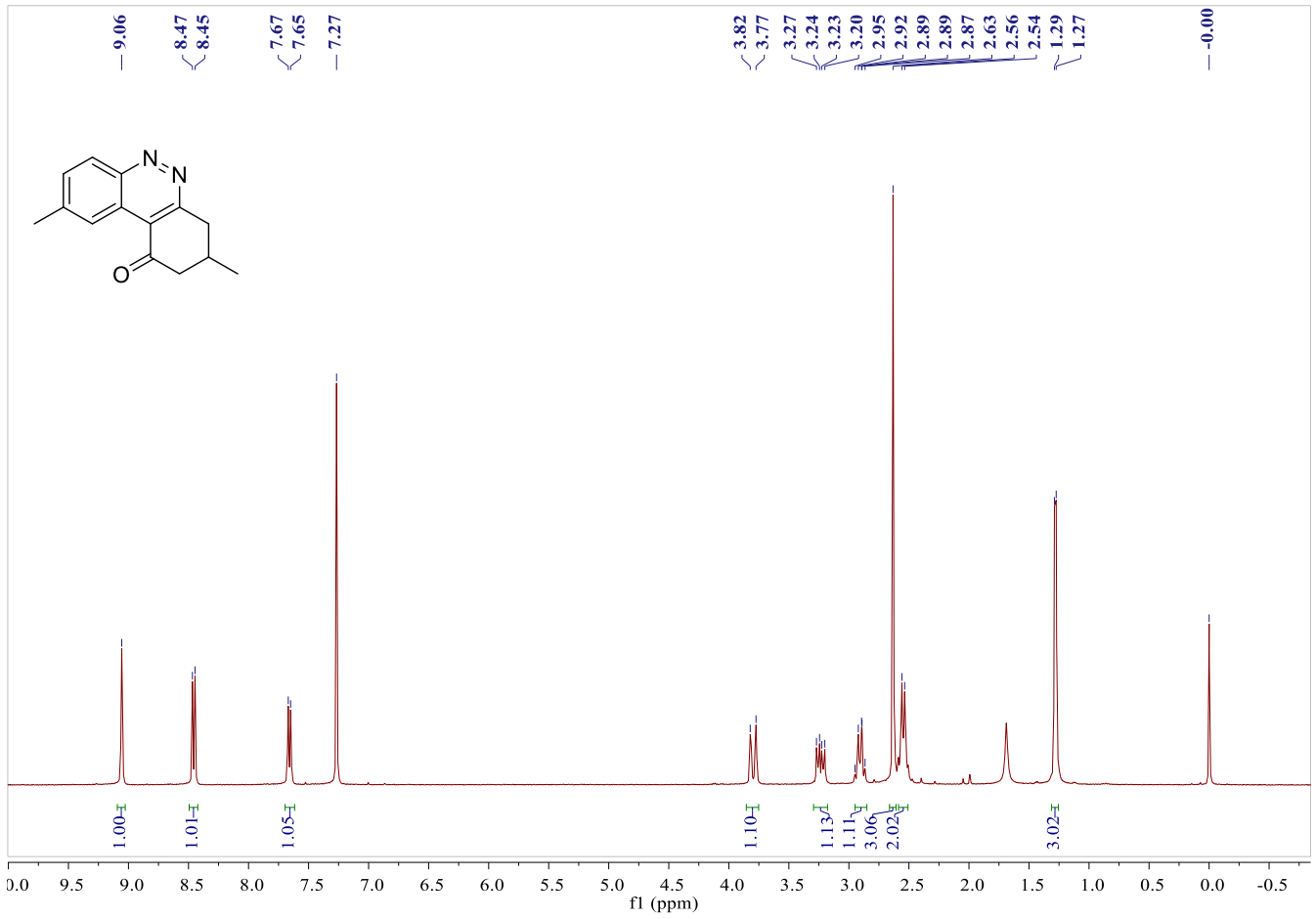



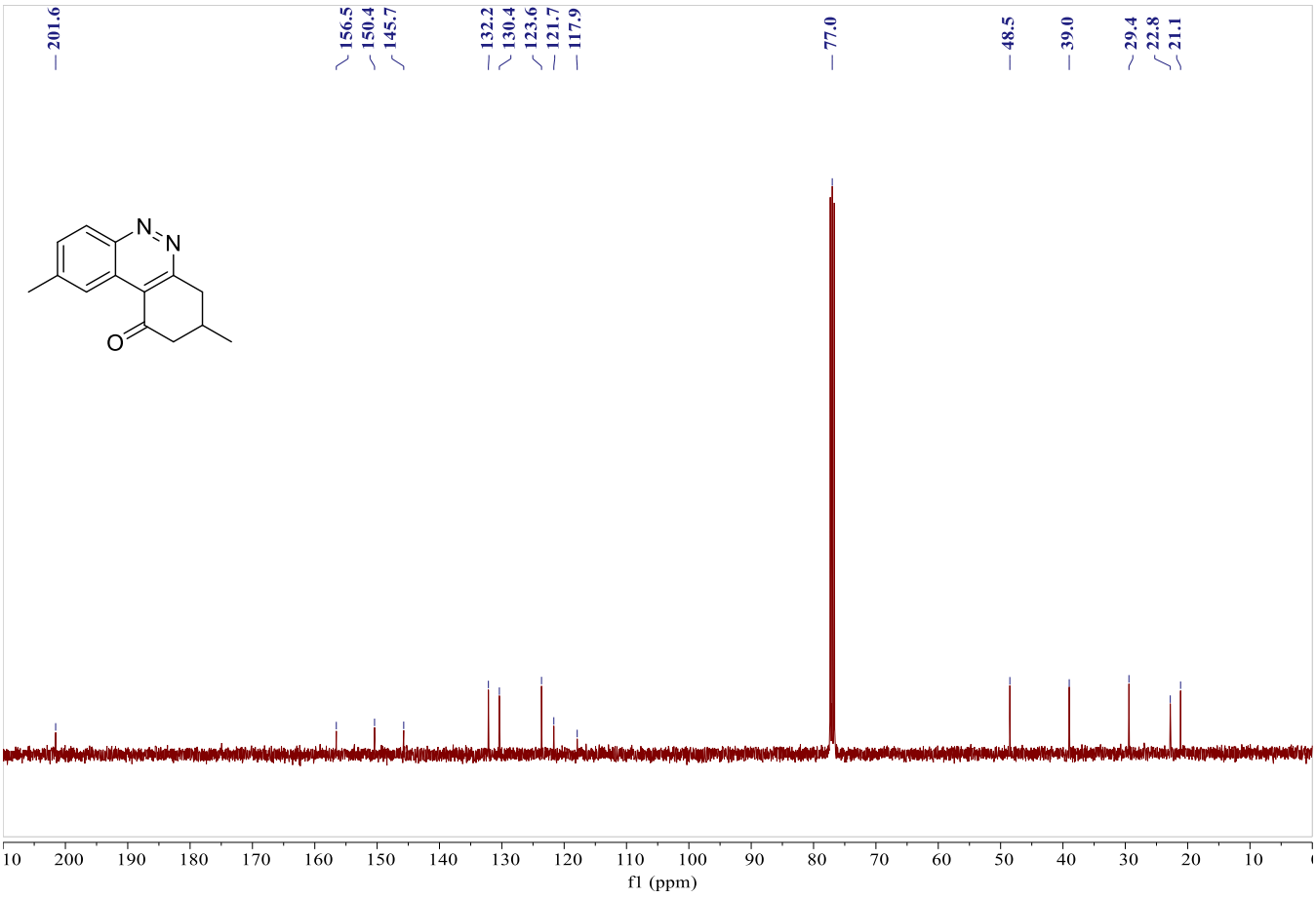
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3h**



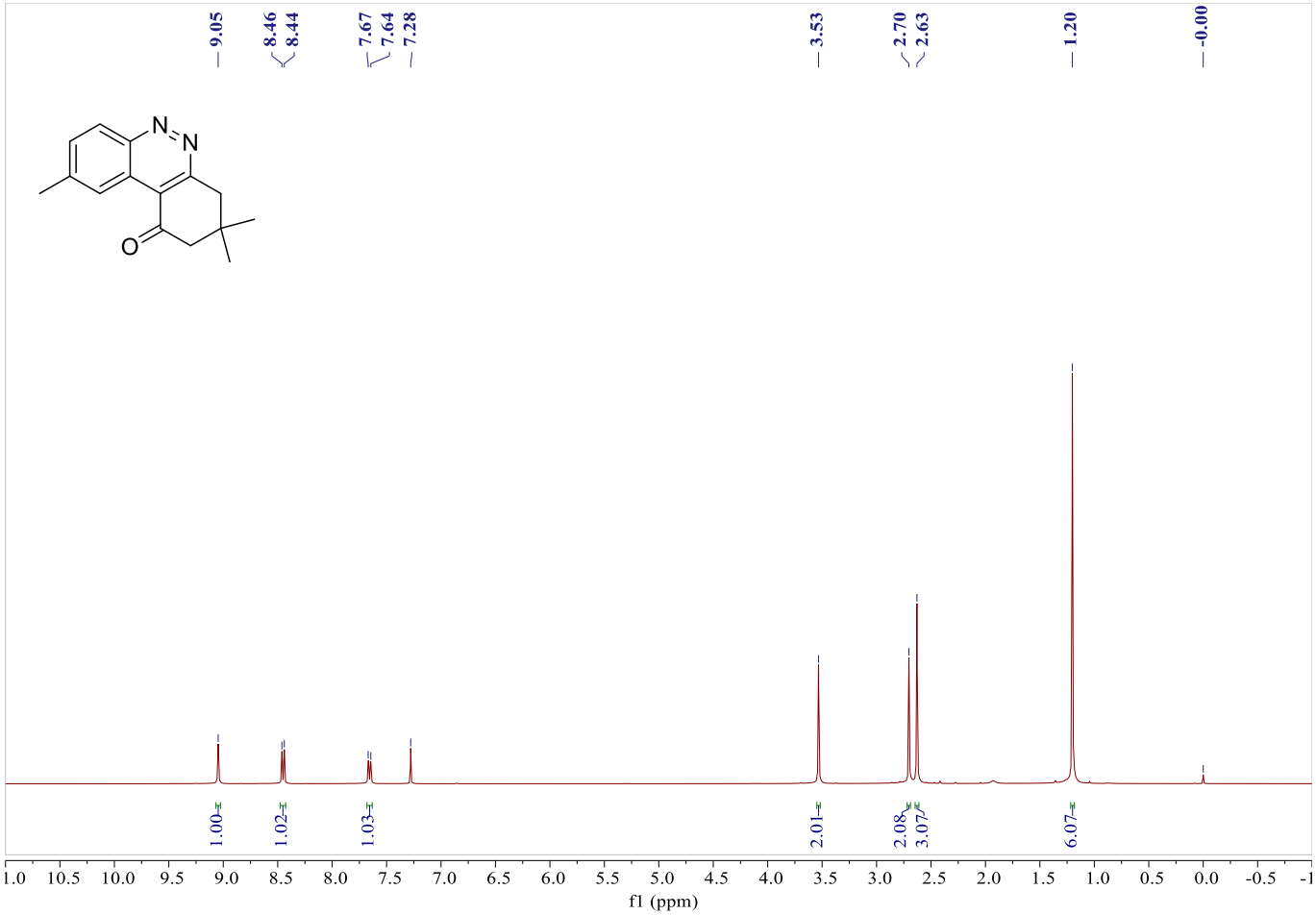



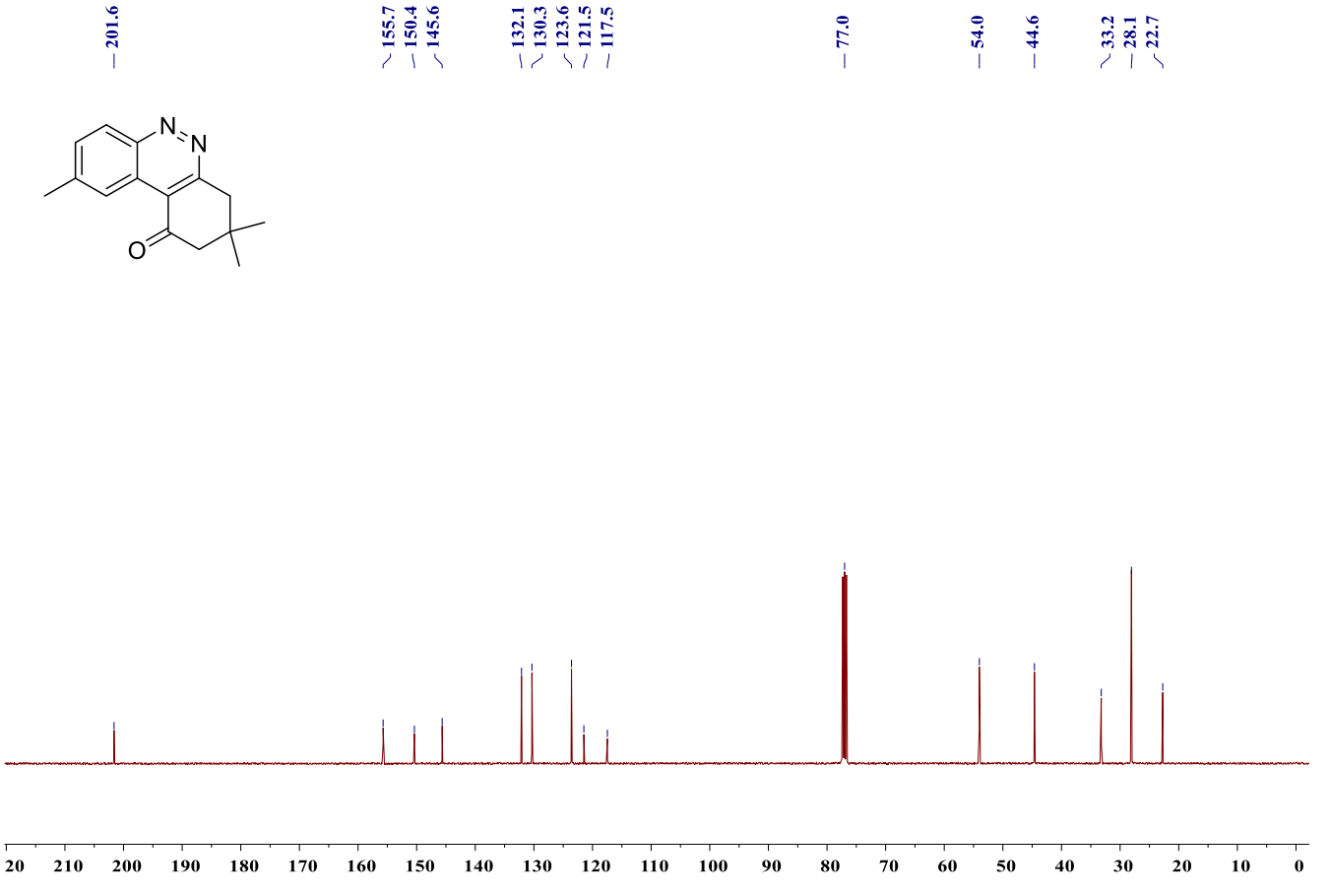
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3i**



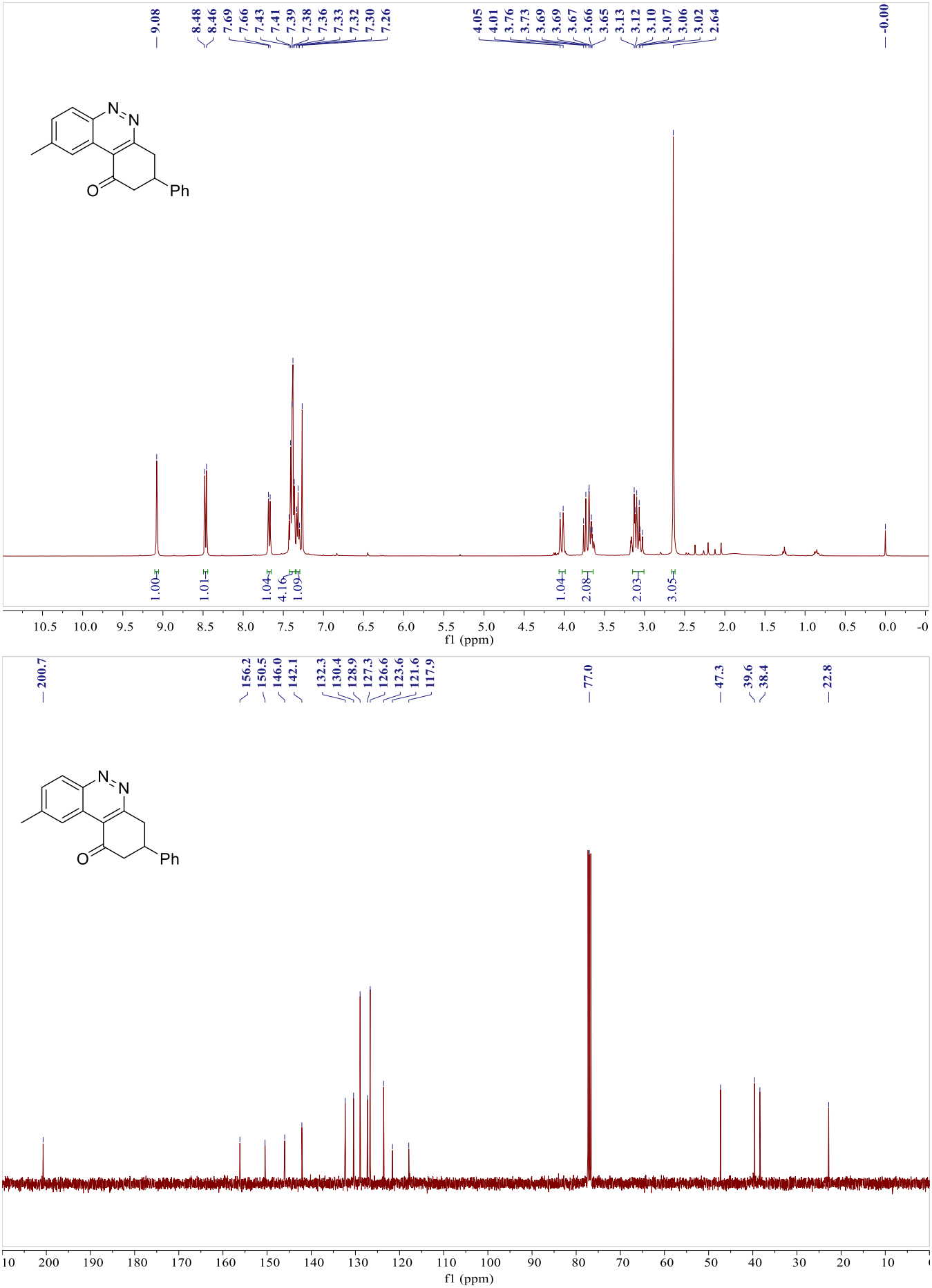



**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3j**

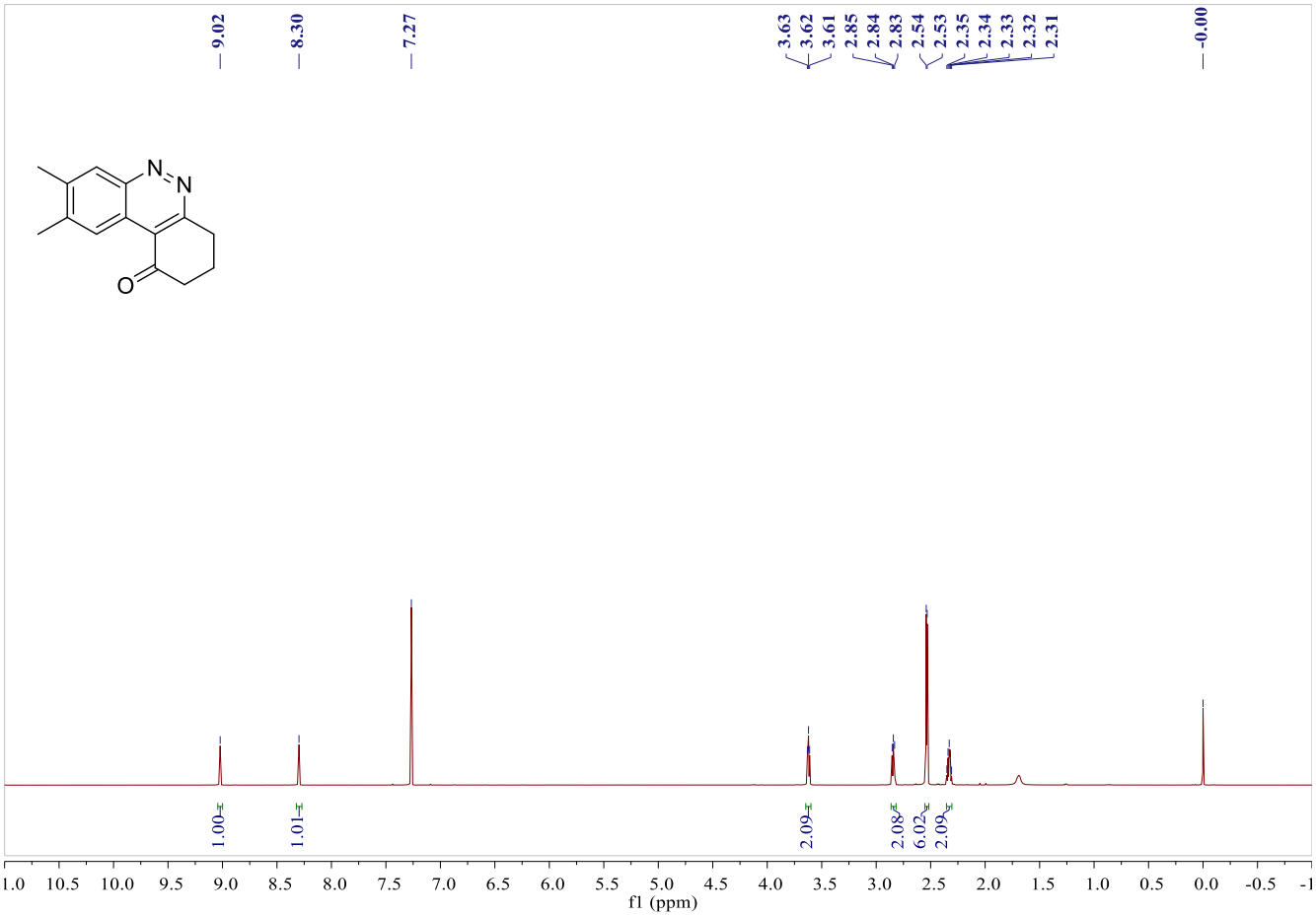


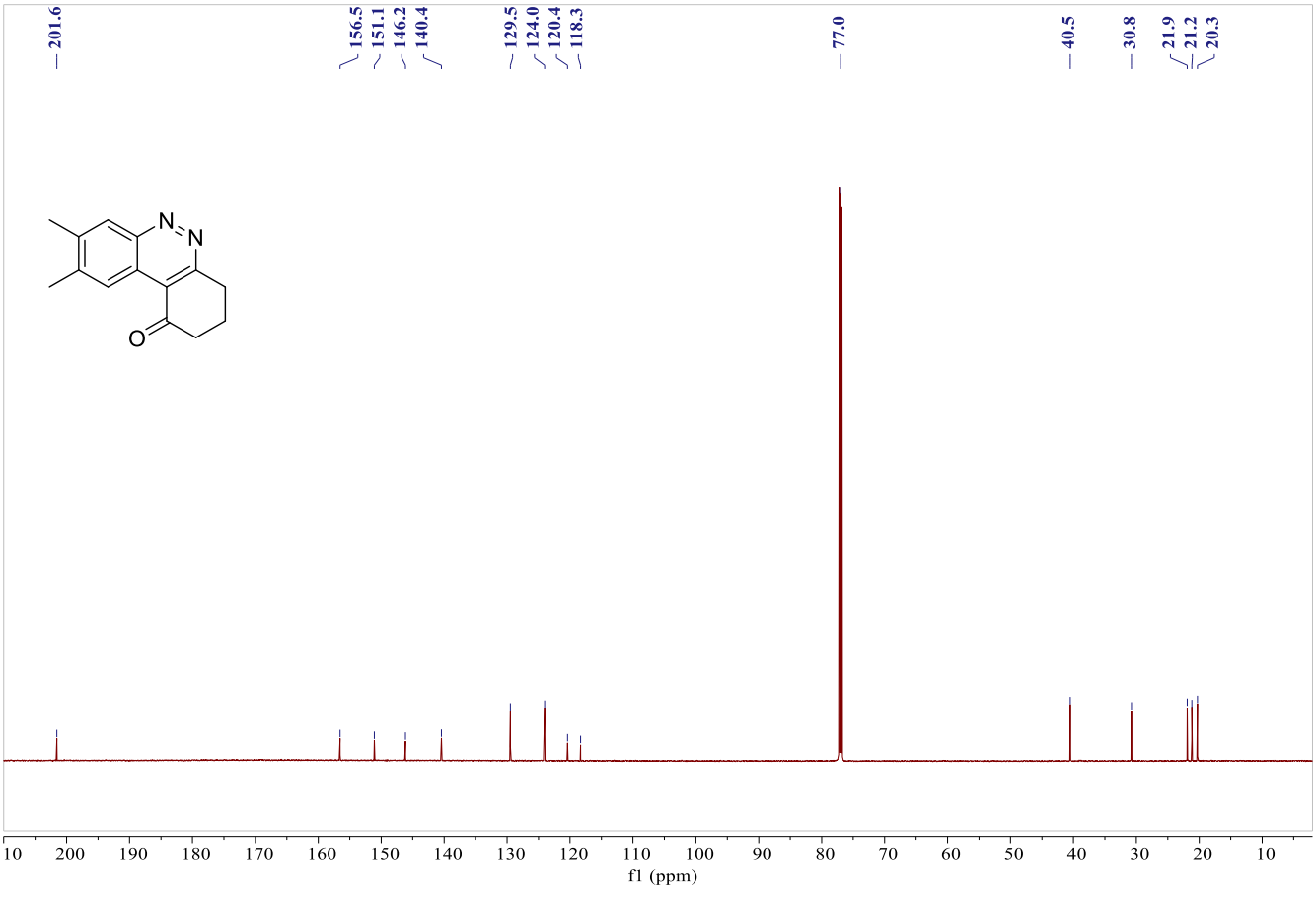



**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3k**

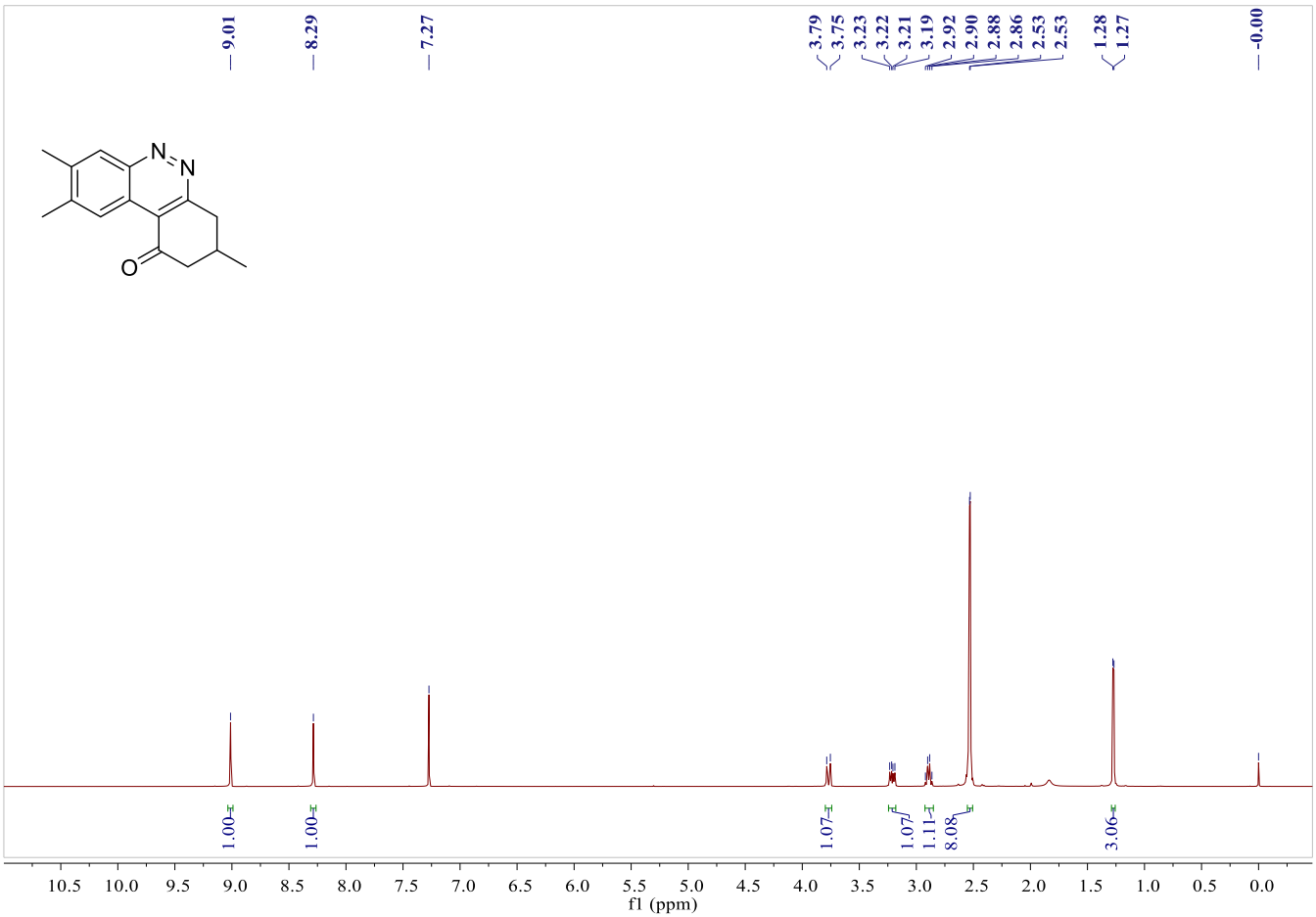


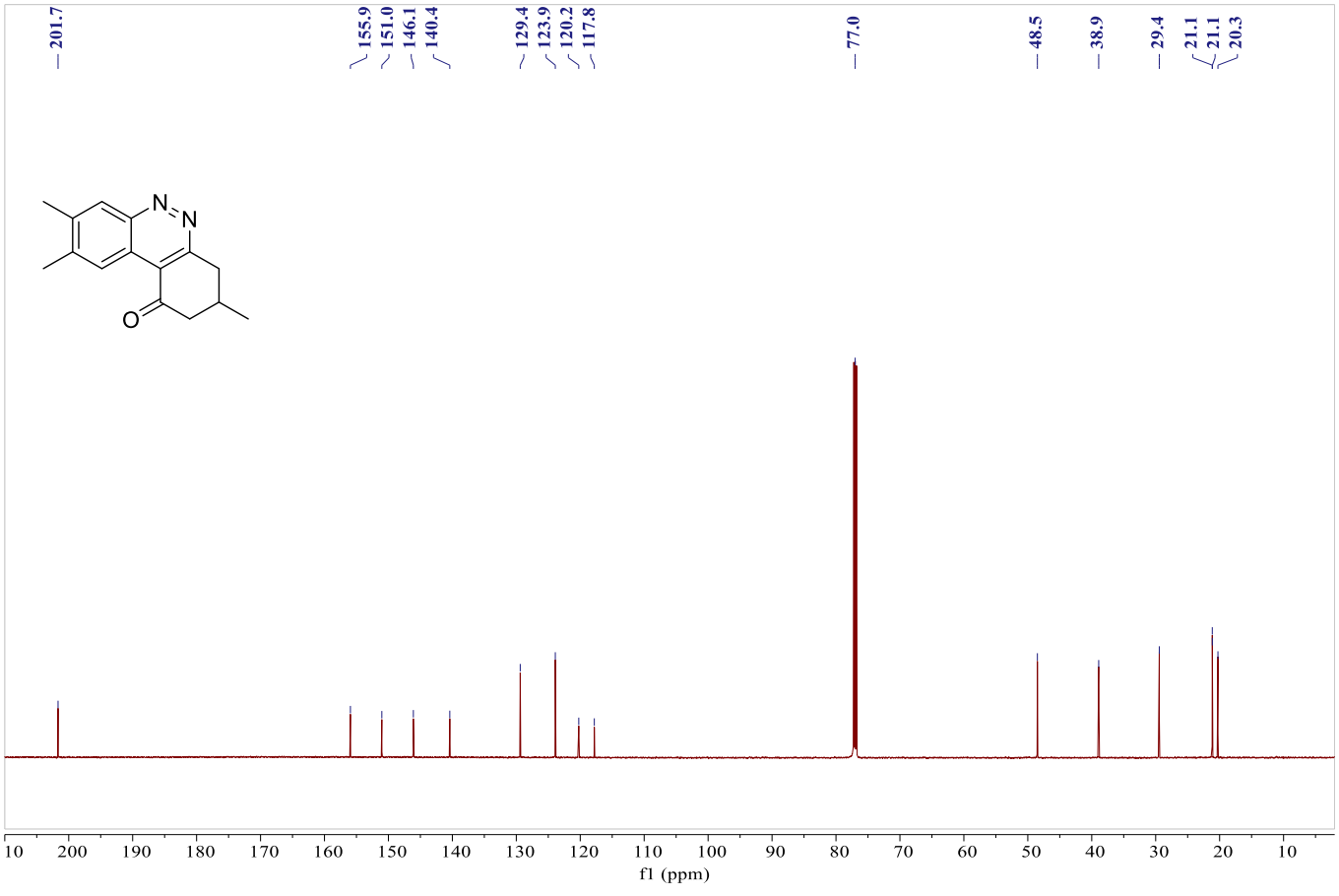
**$^{1}$H NMR (600 MHz, $CDCl_3$) and $^{13}$C NMR (150 MHz, $CDCl_3$) spectra of product 3l**



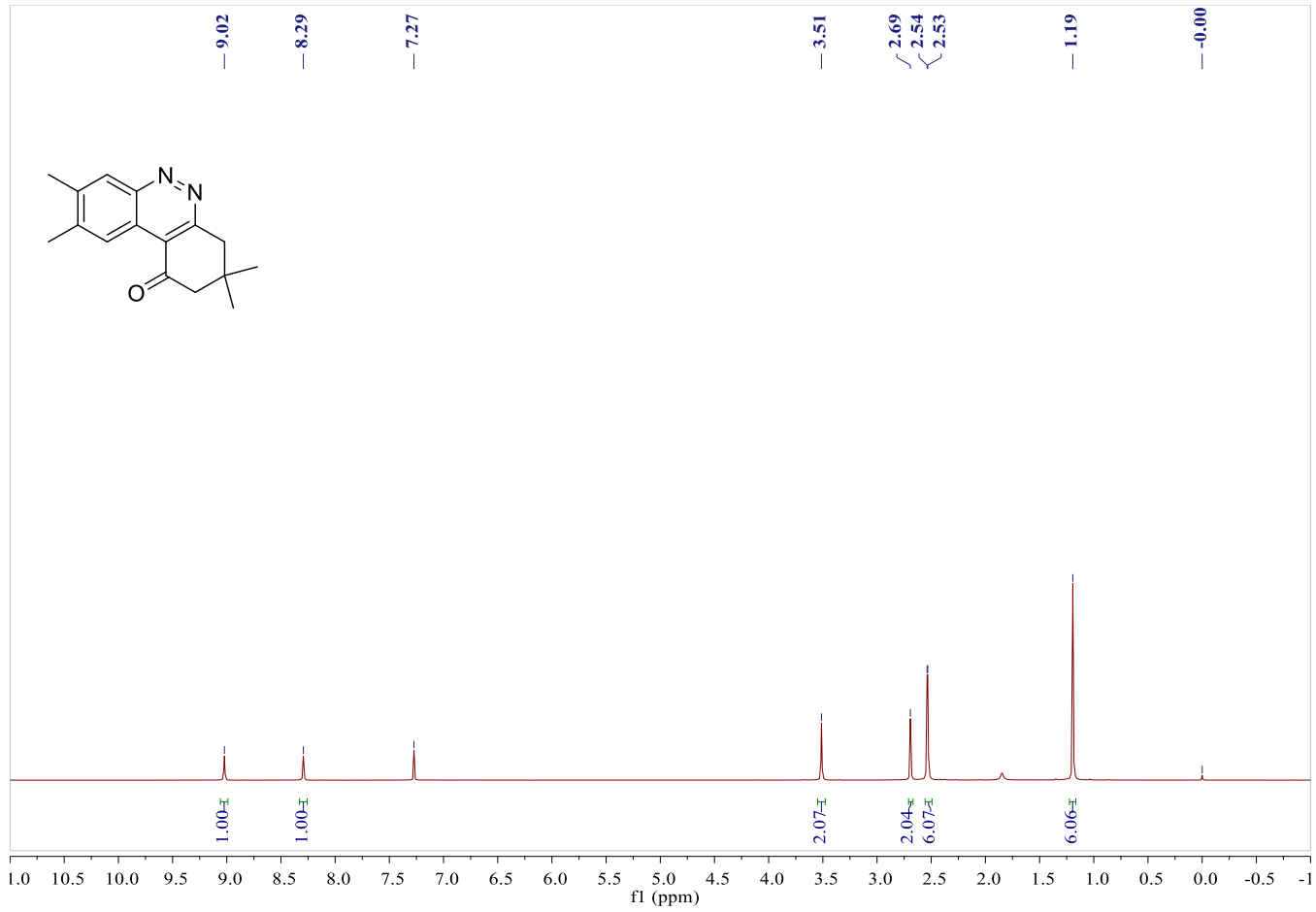



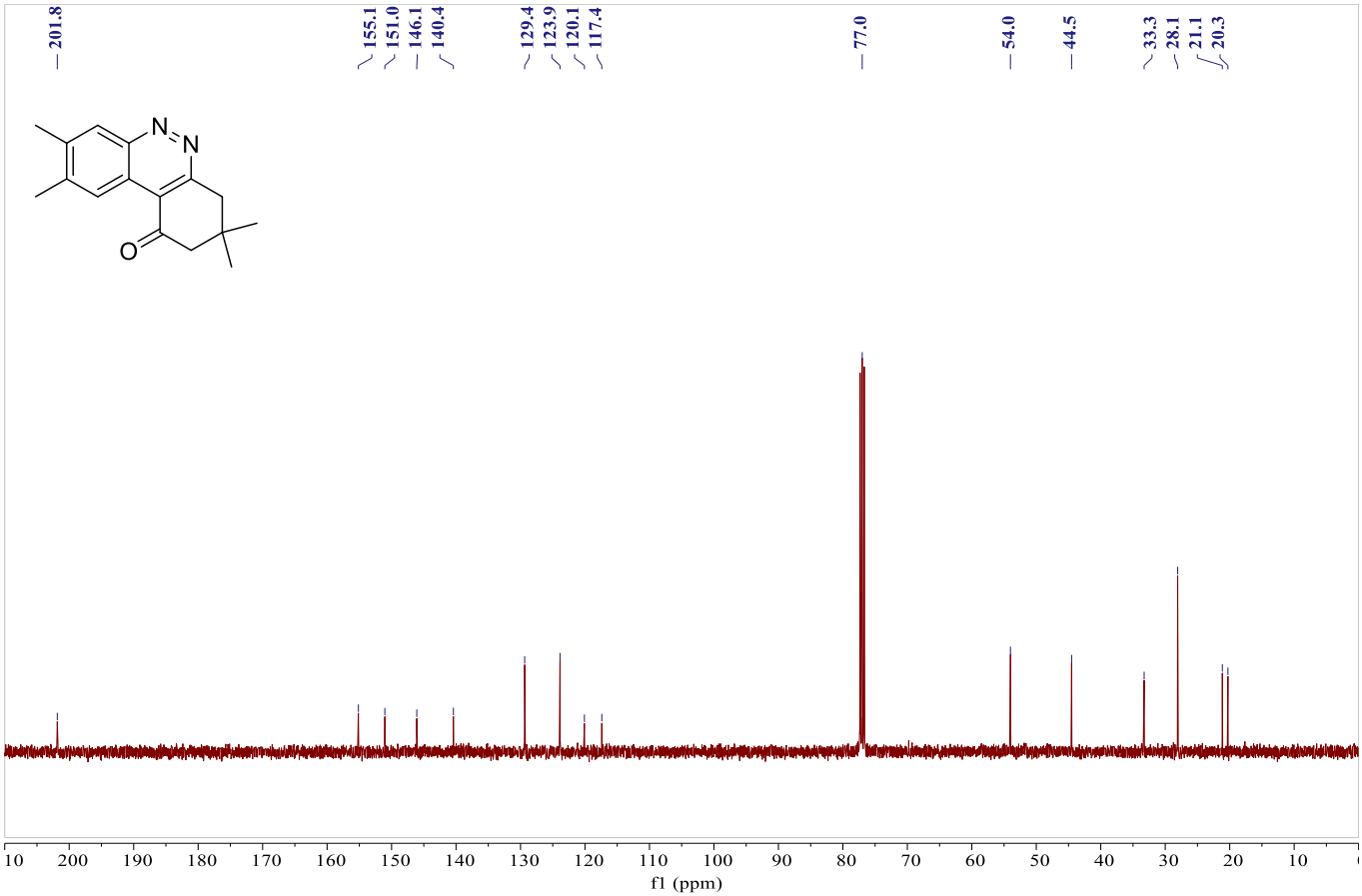
**$^{1}$H NMR (600 MHz, $CDCl_3$) and $^{13}$C NMR (150 MHz, $CDCl_3$) spectra of product 3m**



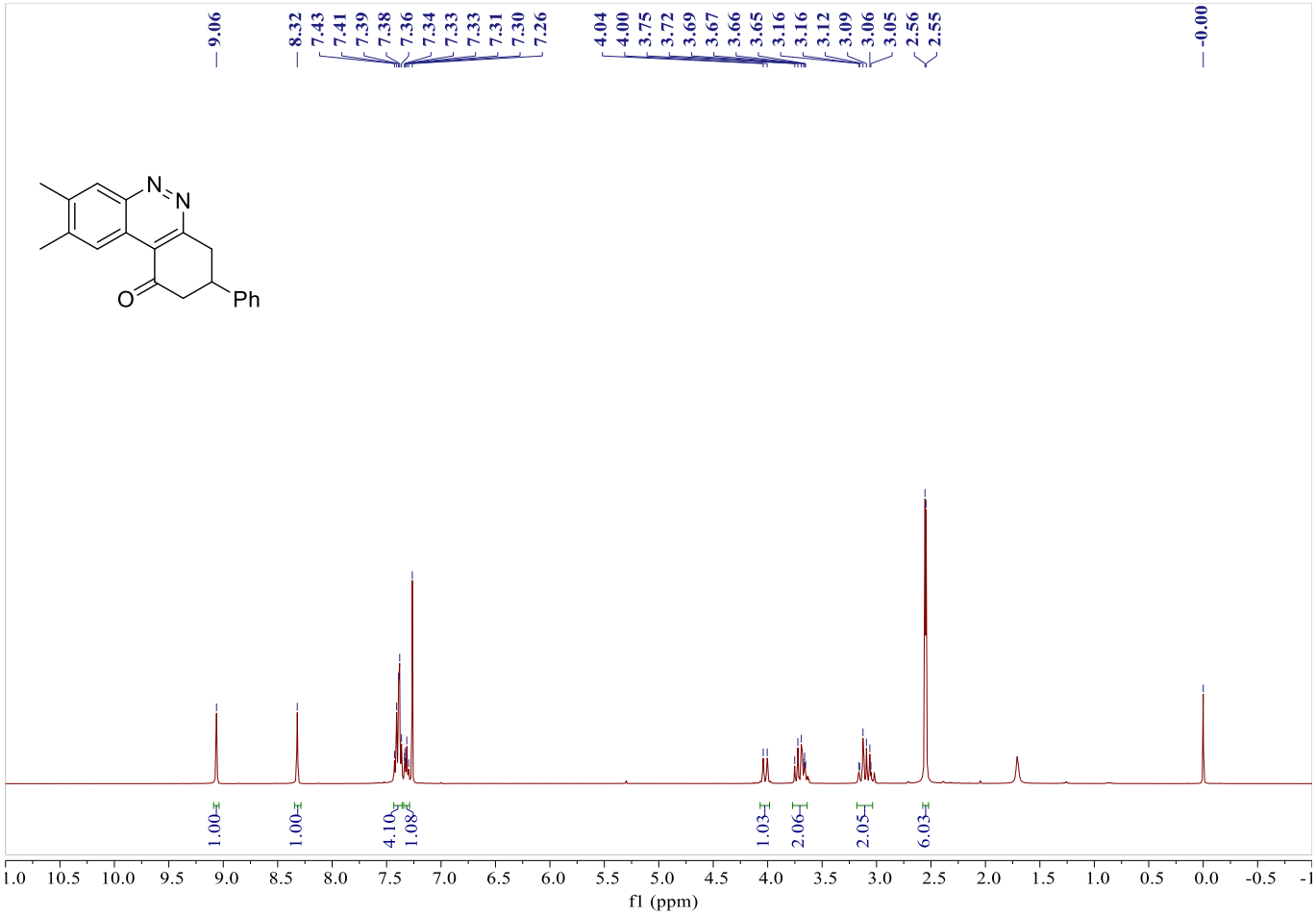



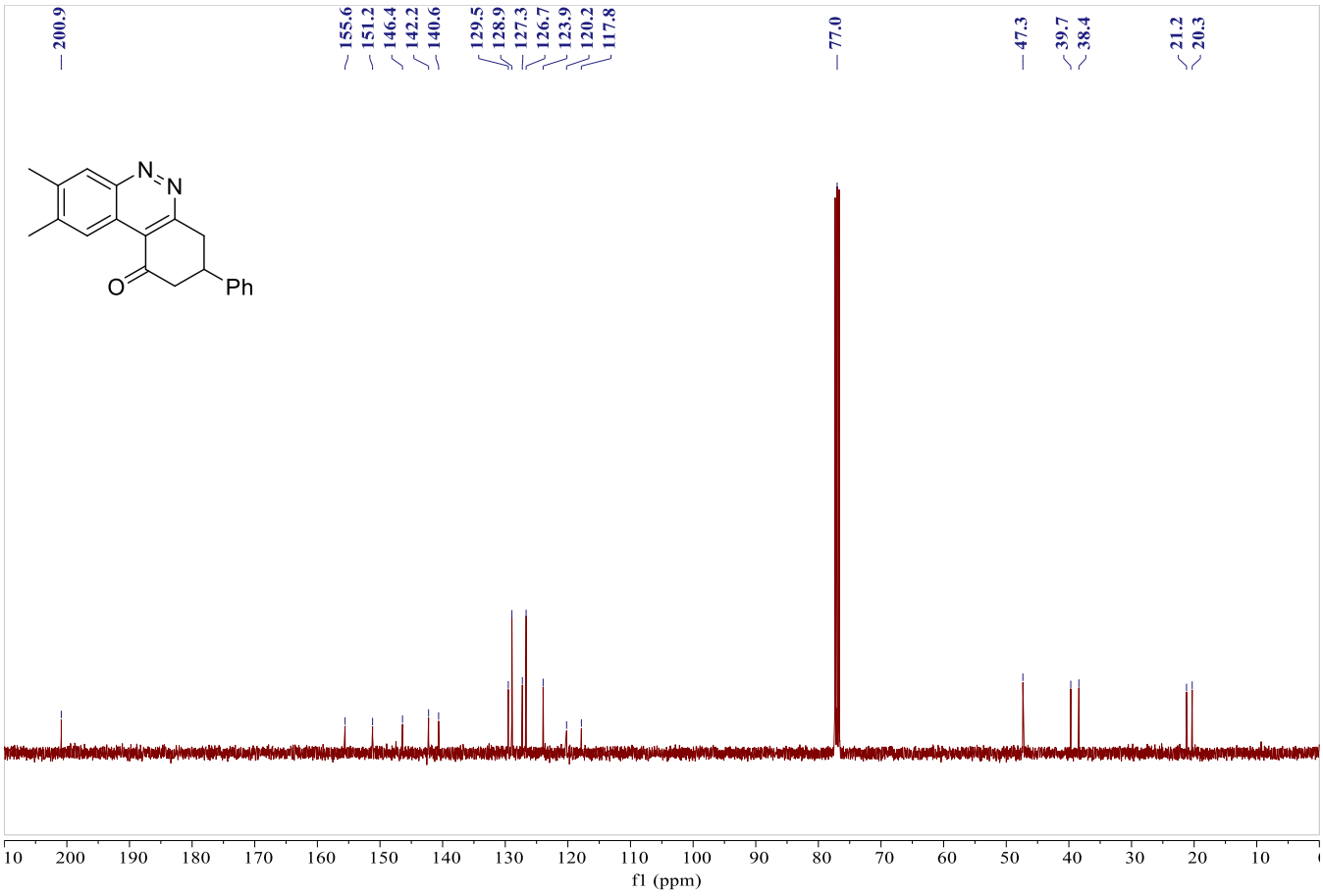
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3n**



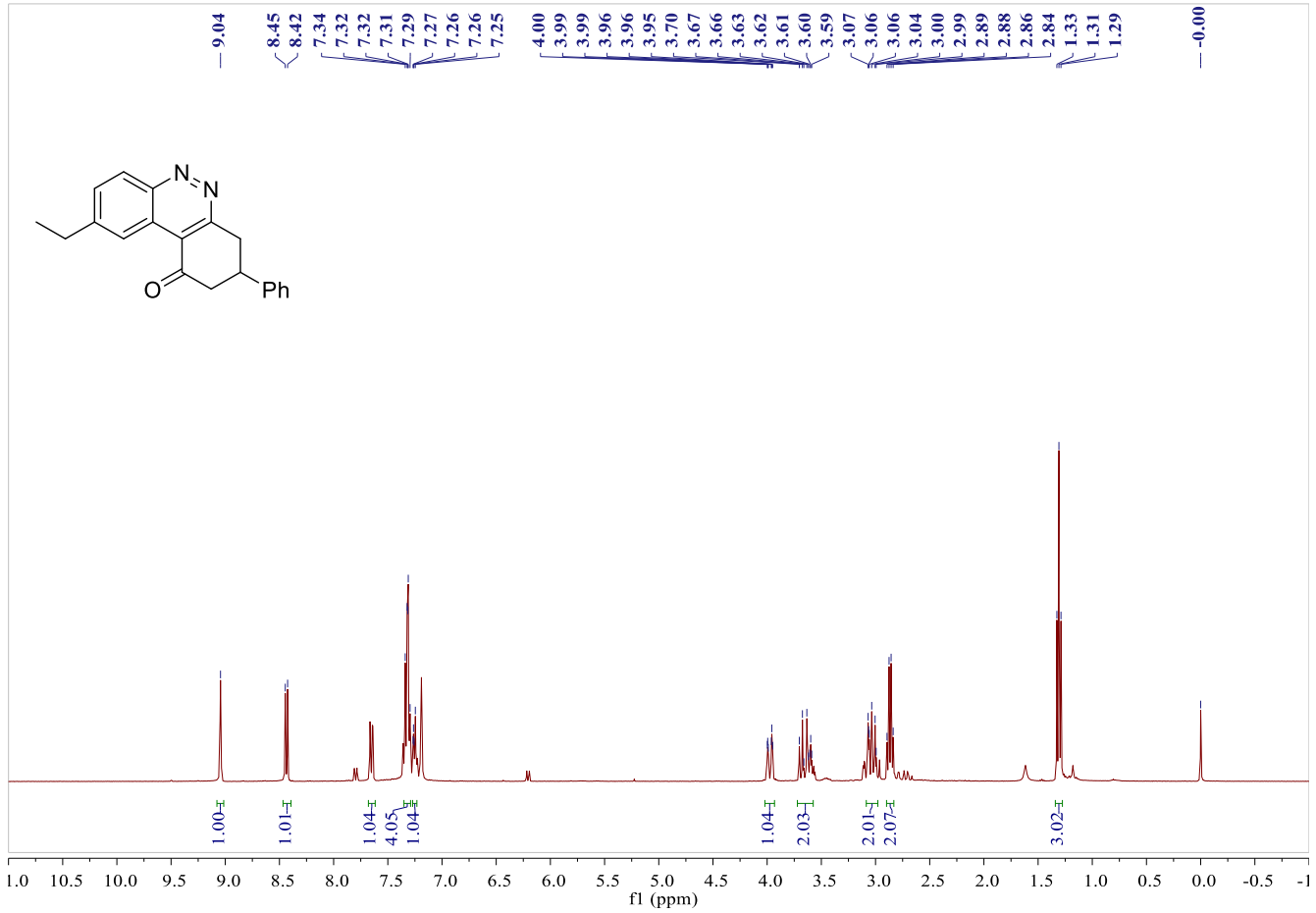



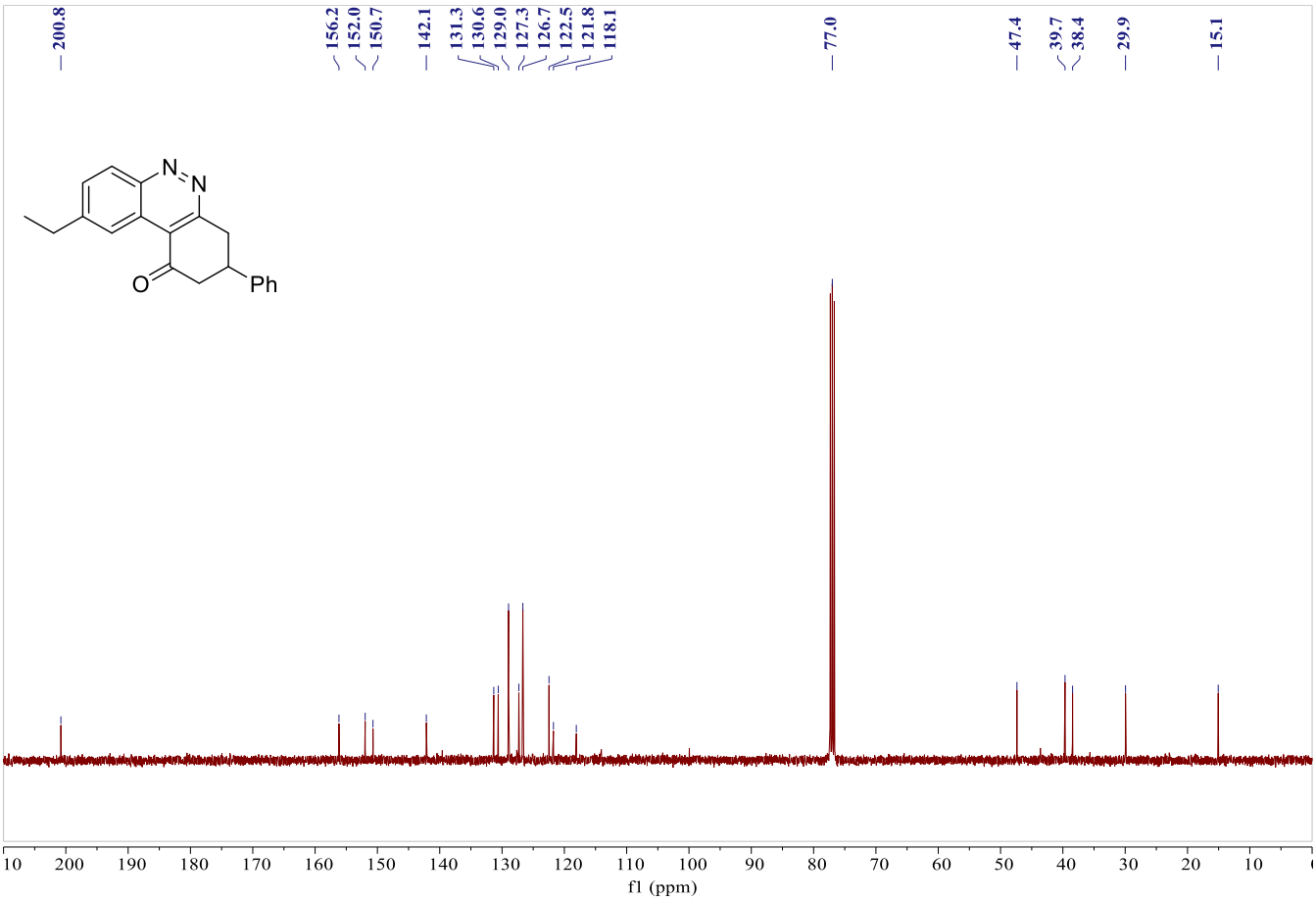
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3o**



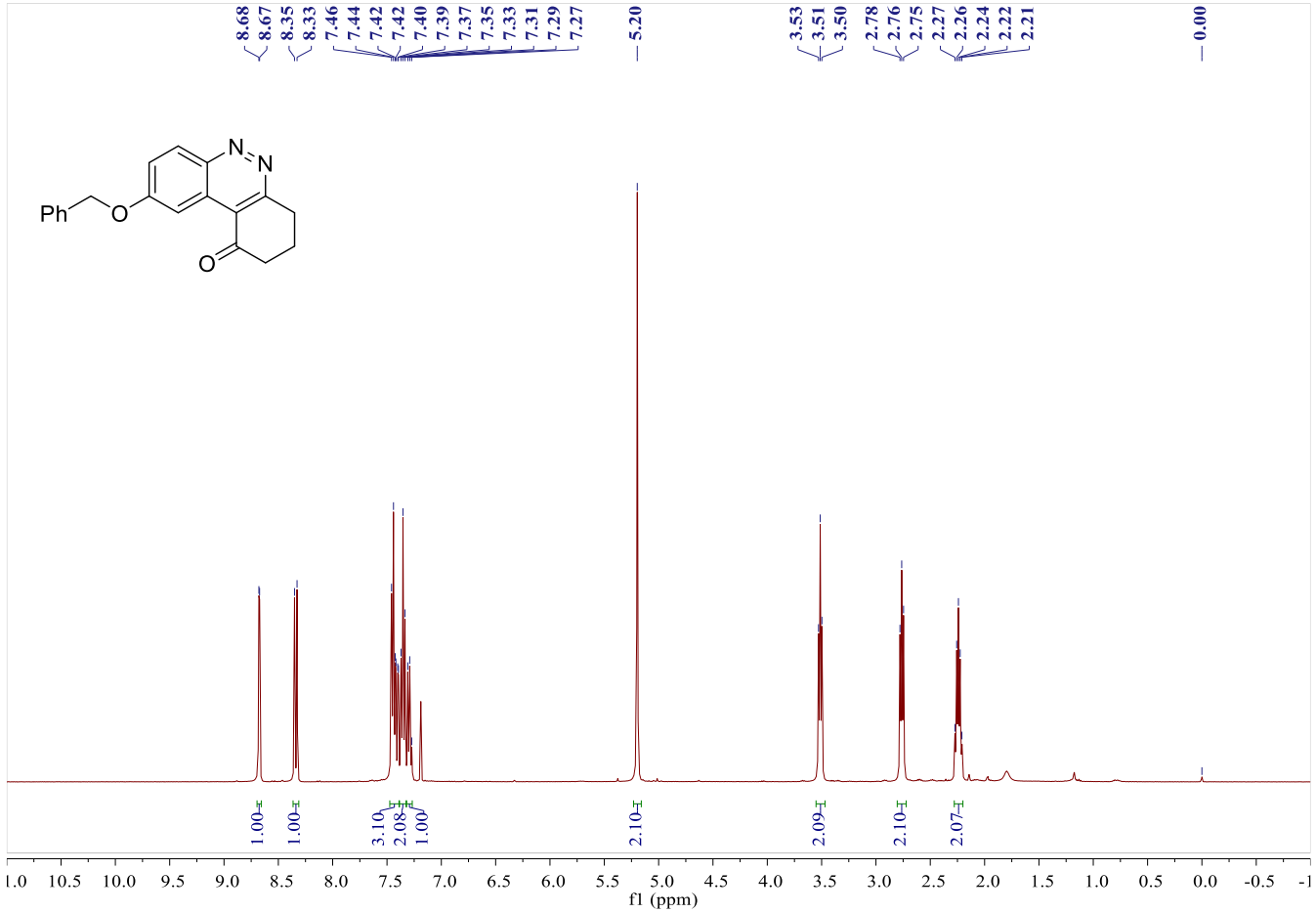



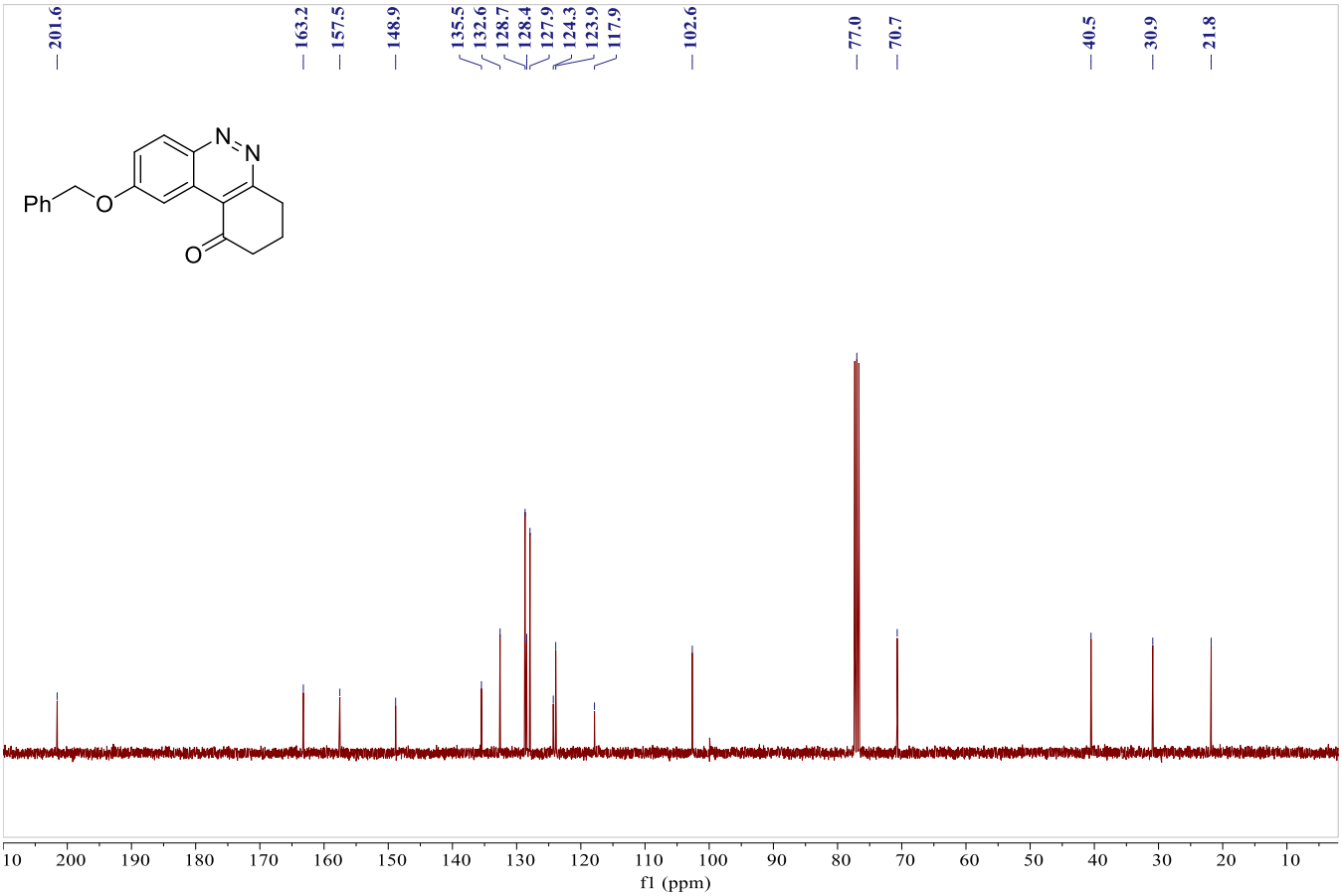
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3p**



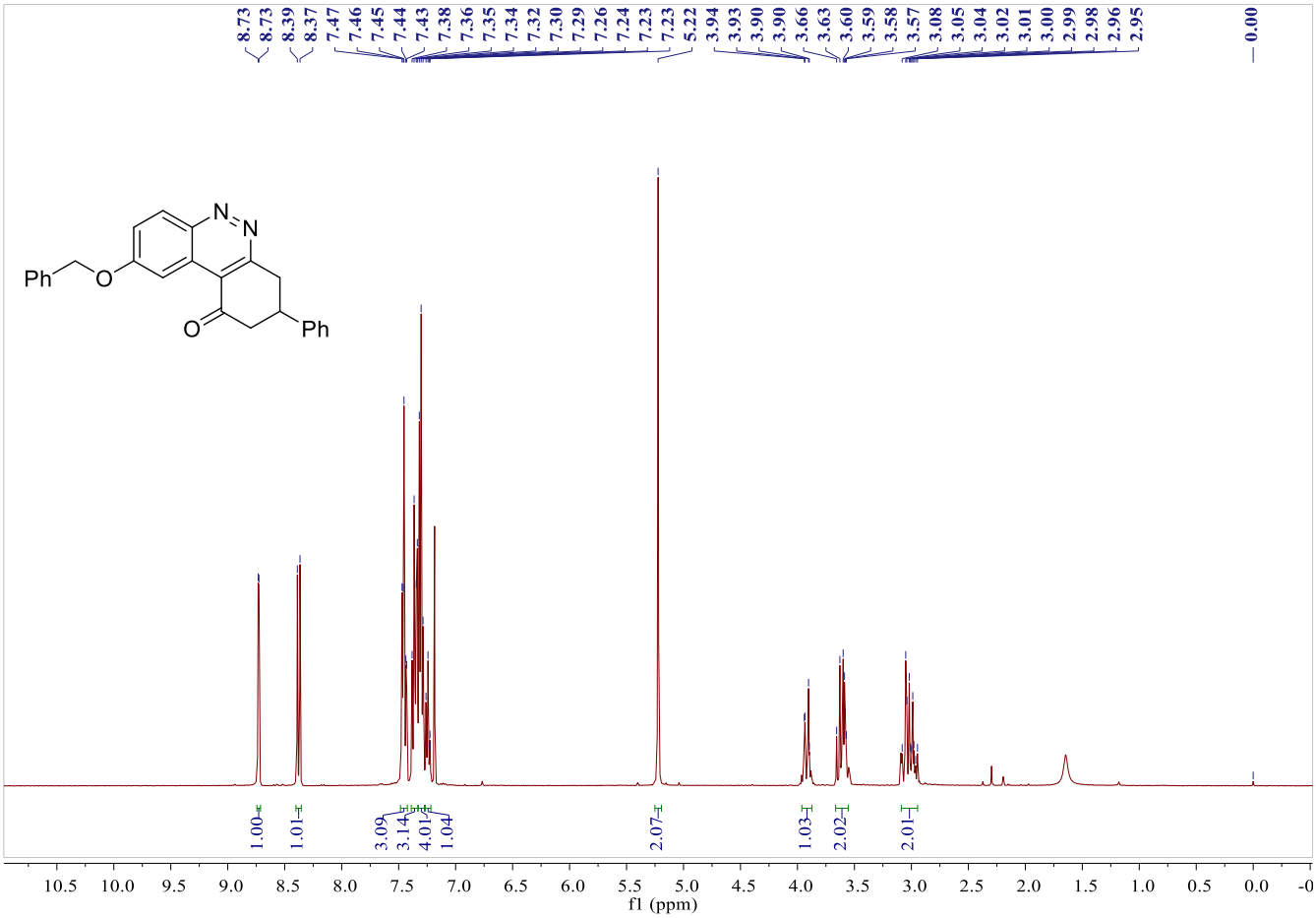



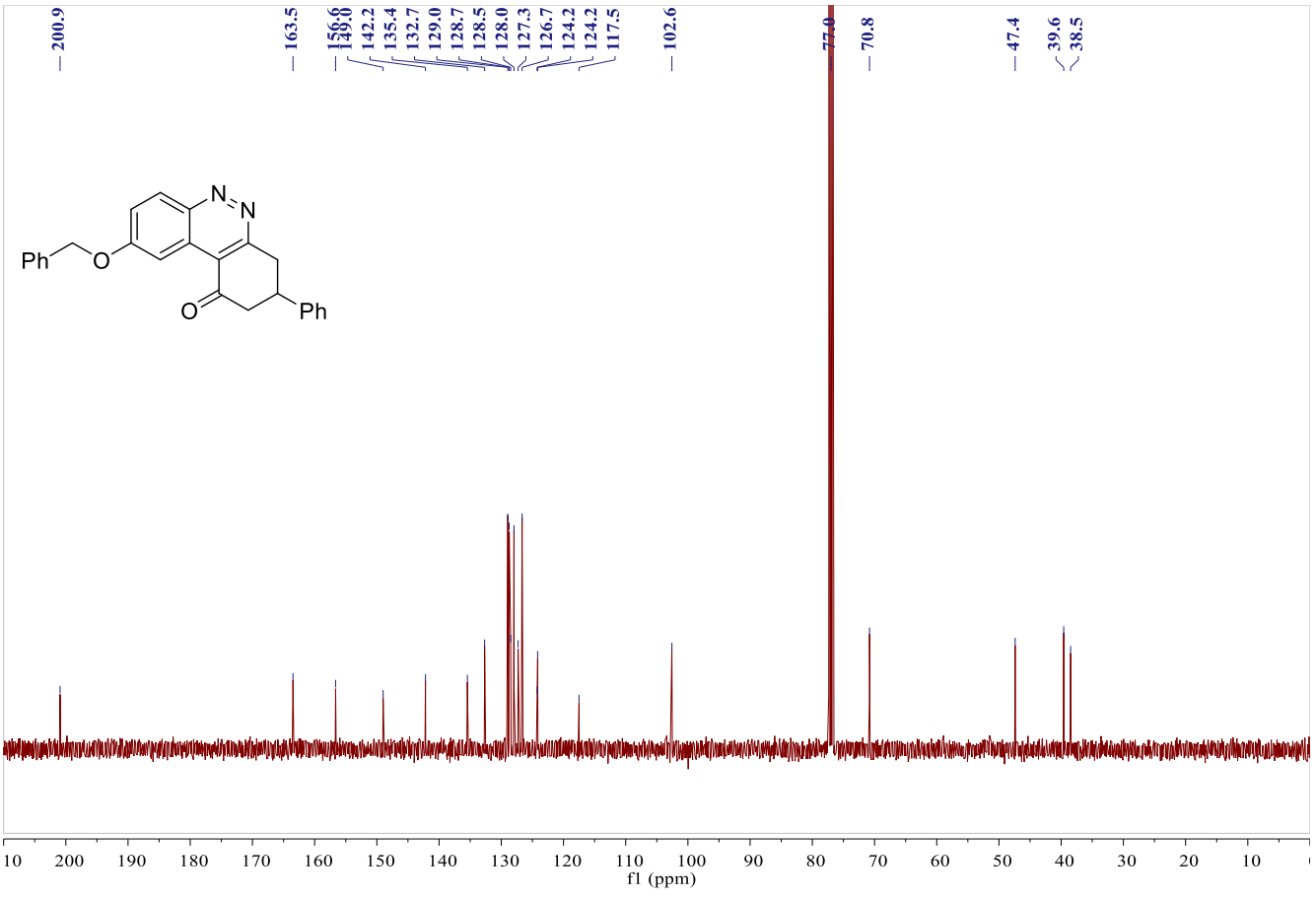
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3q**



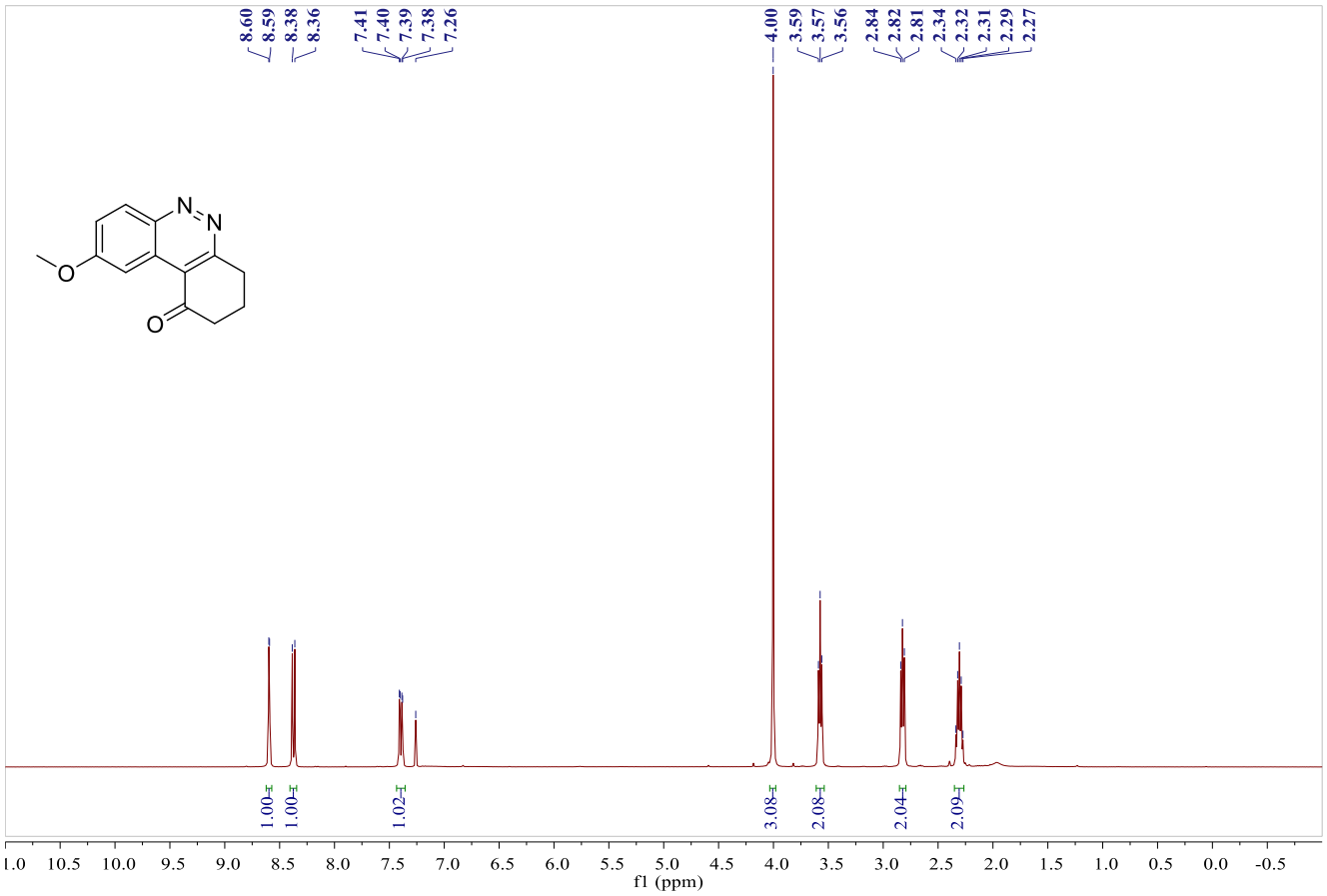



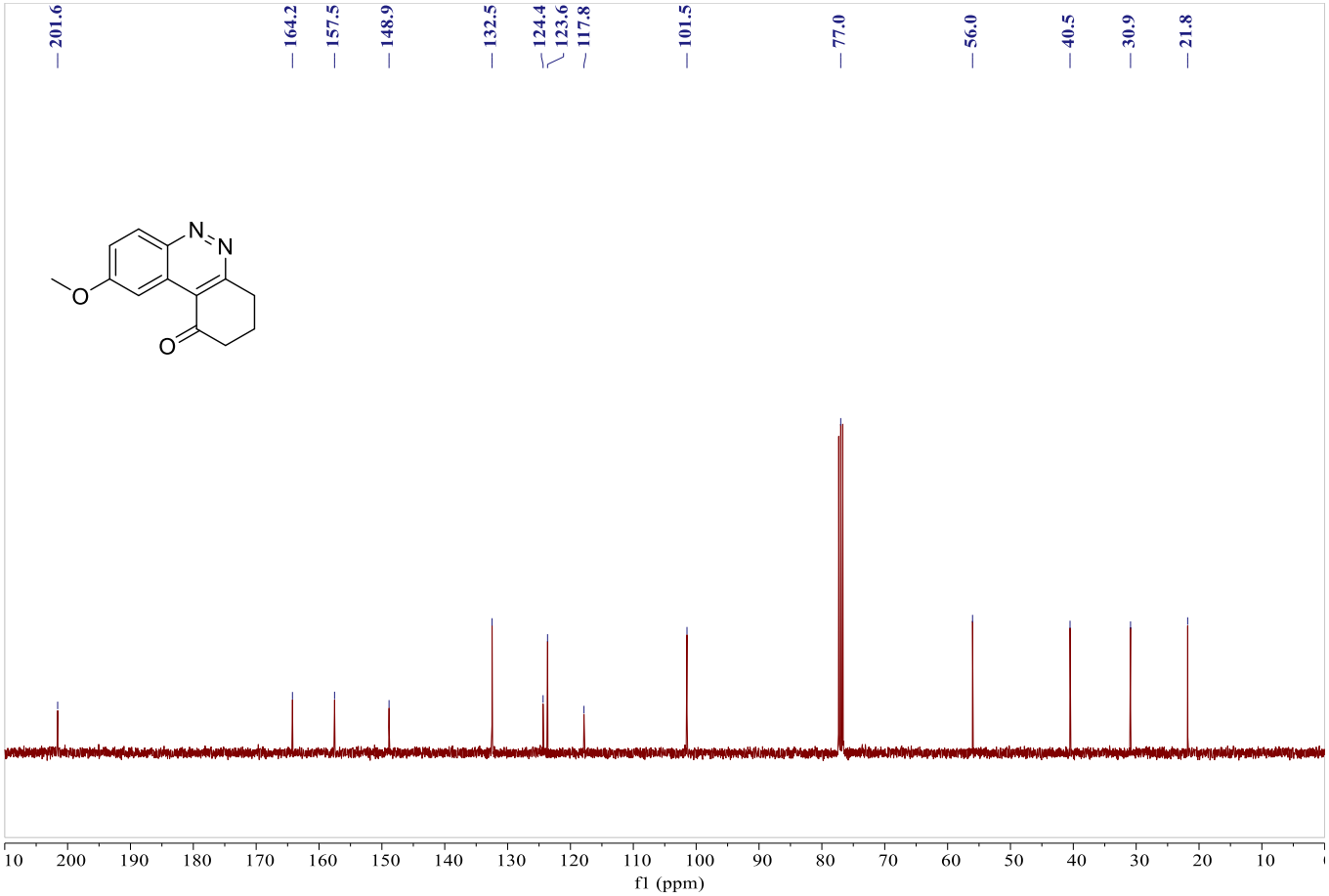
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3r**



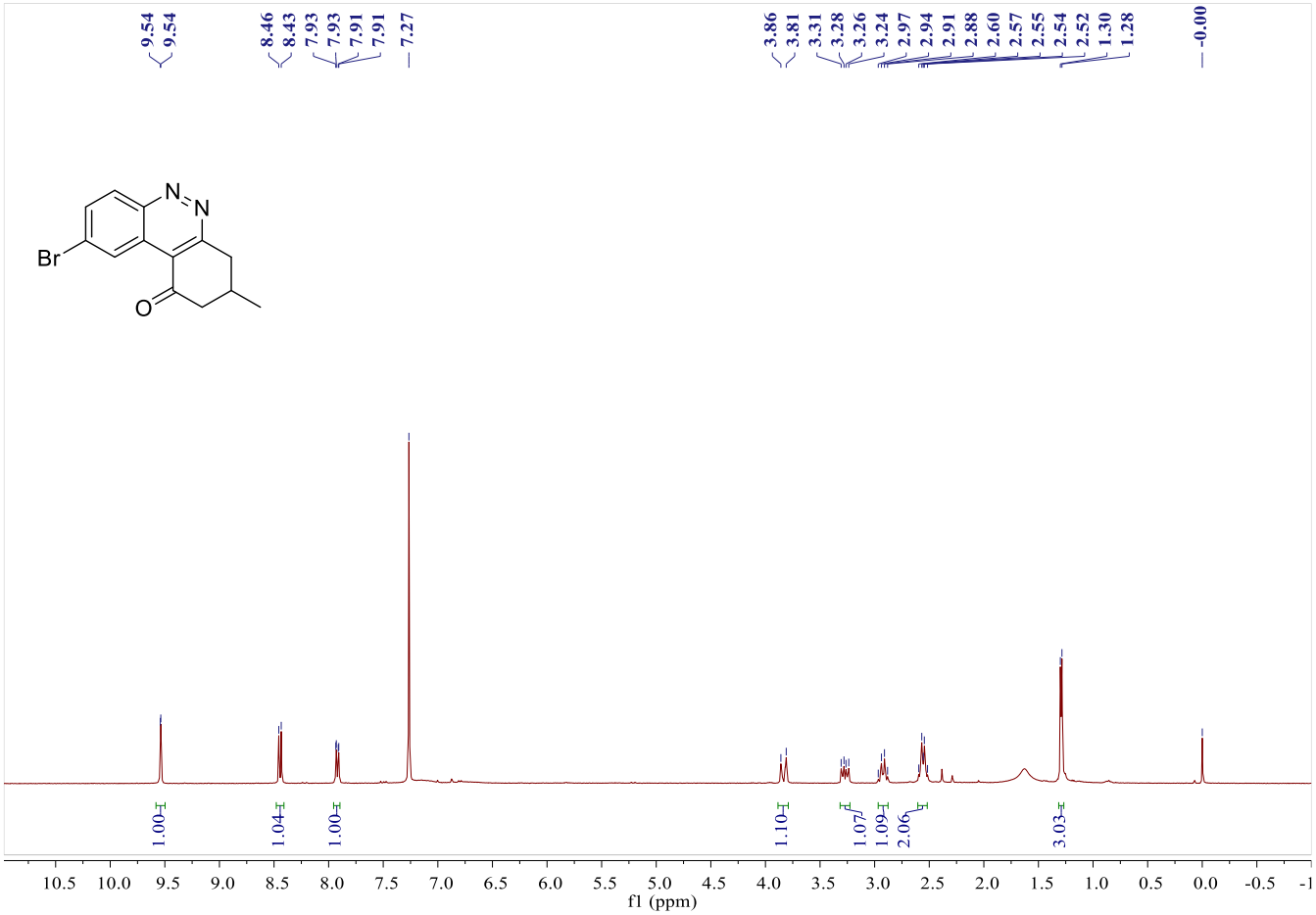



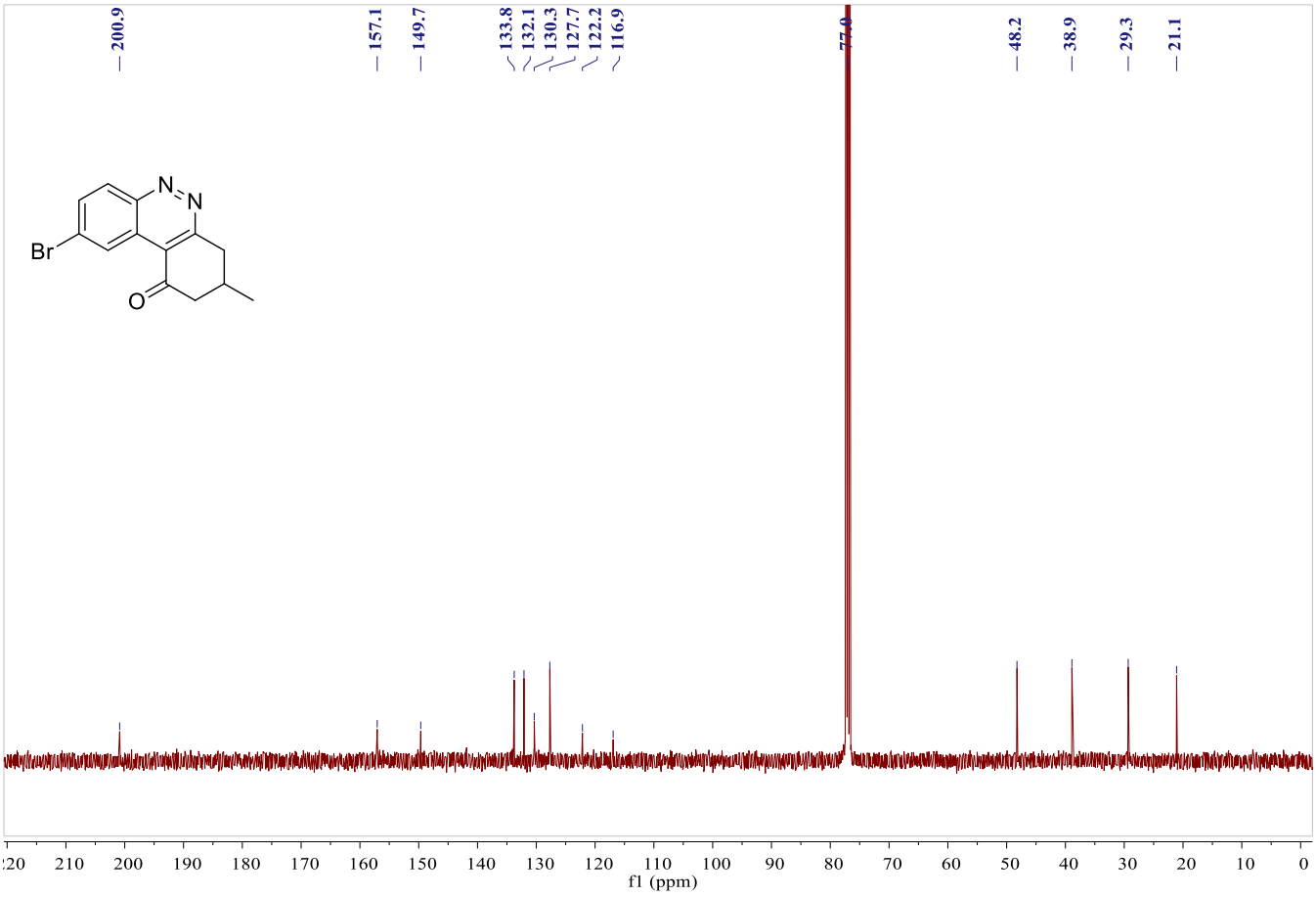
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3s**



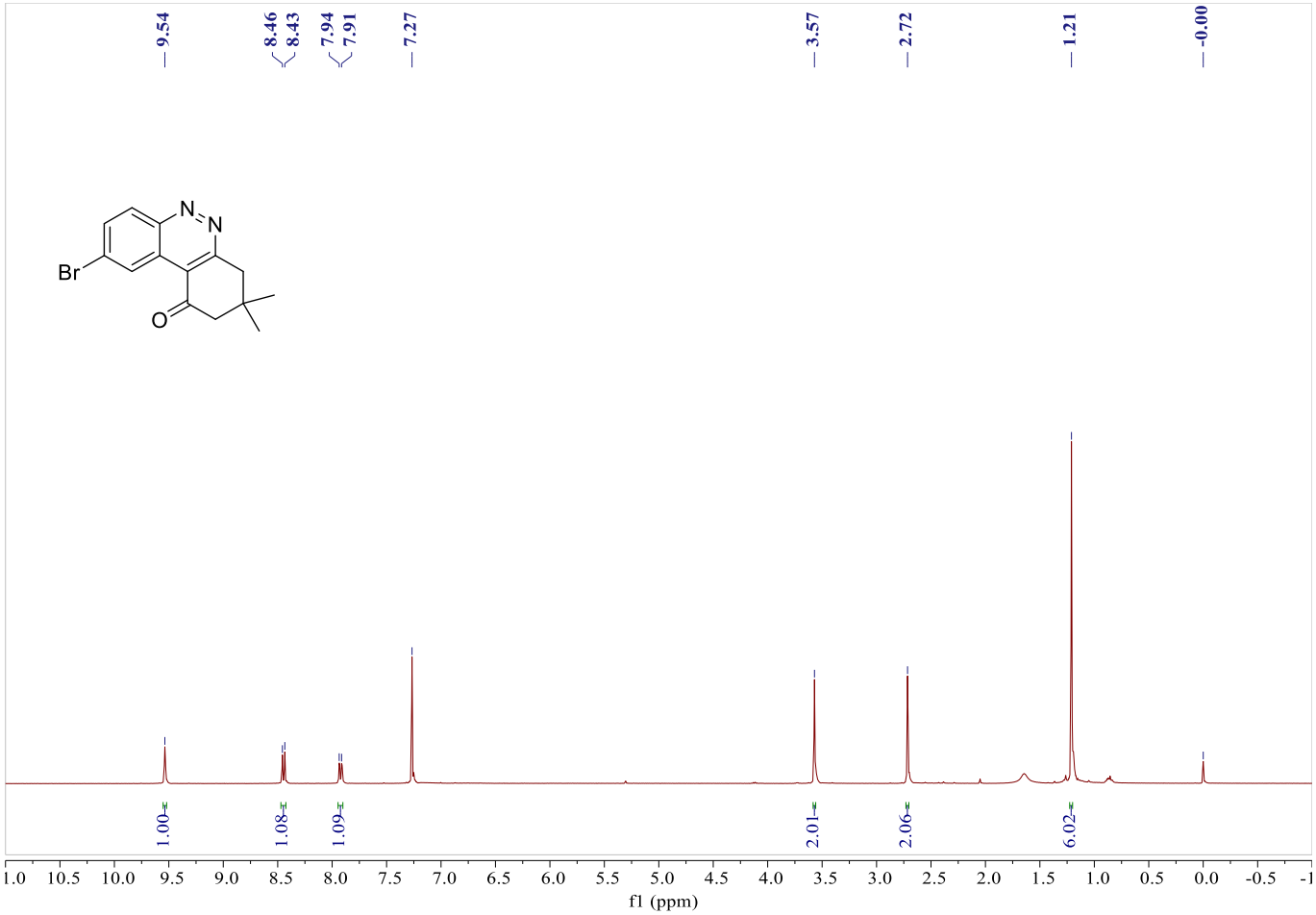



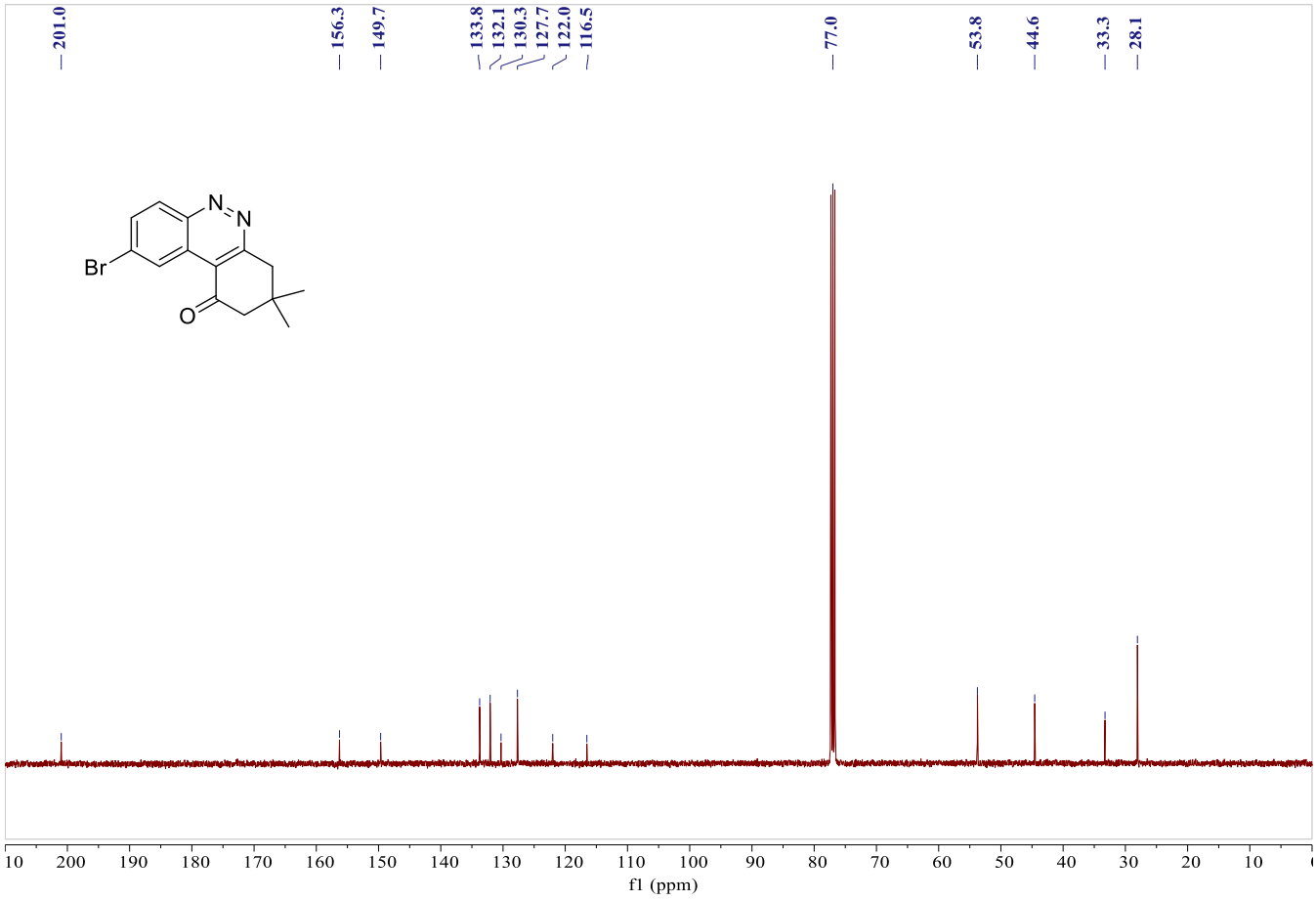
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3t**



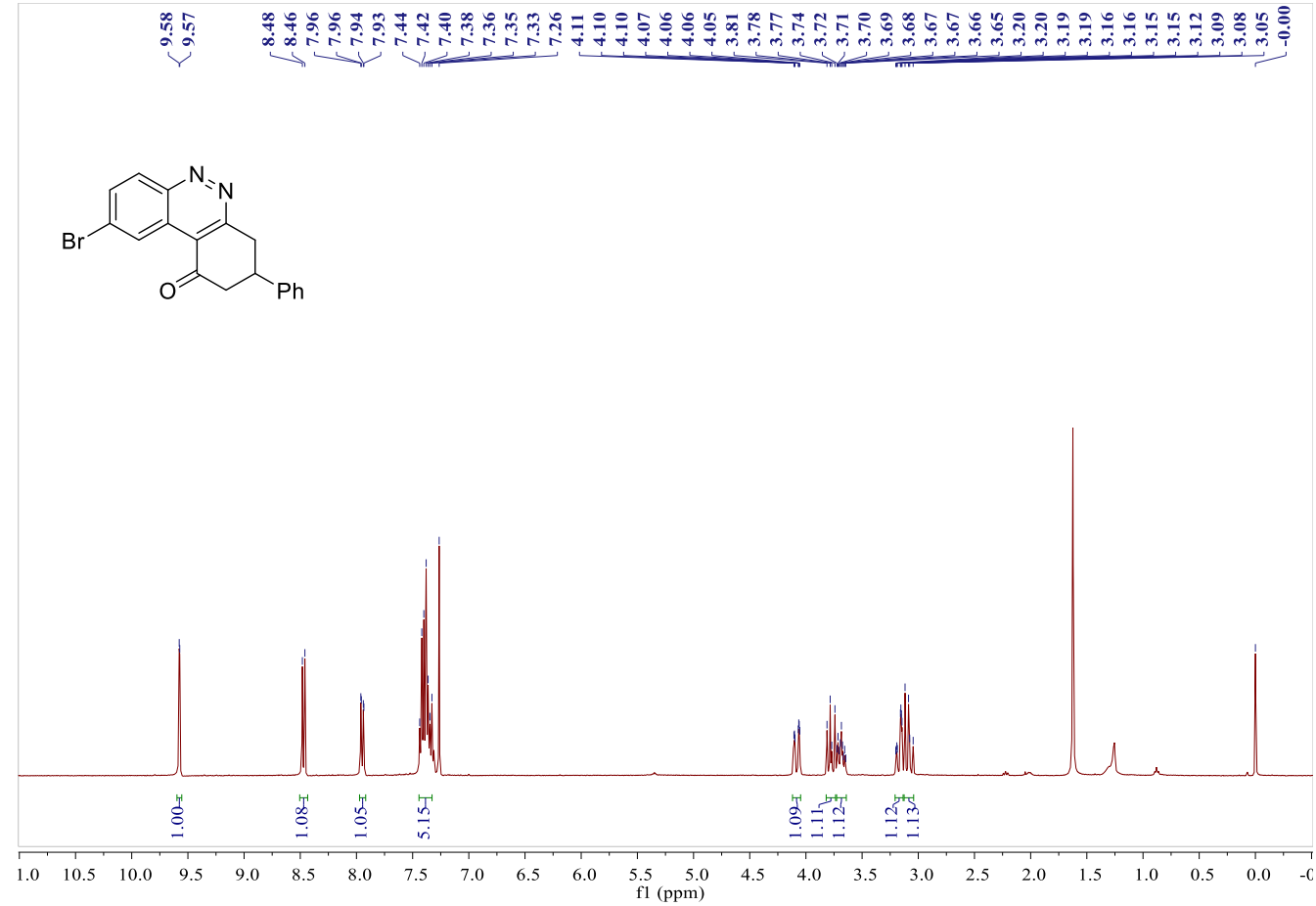



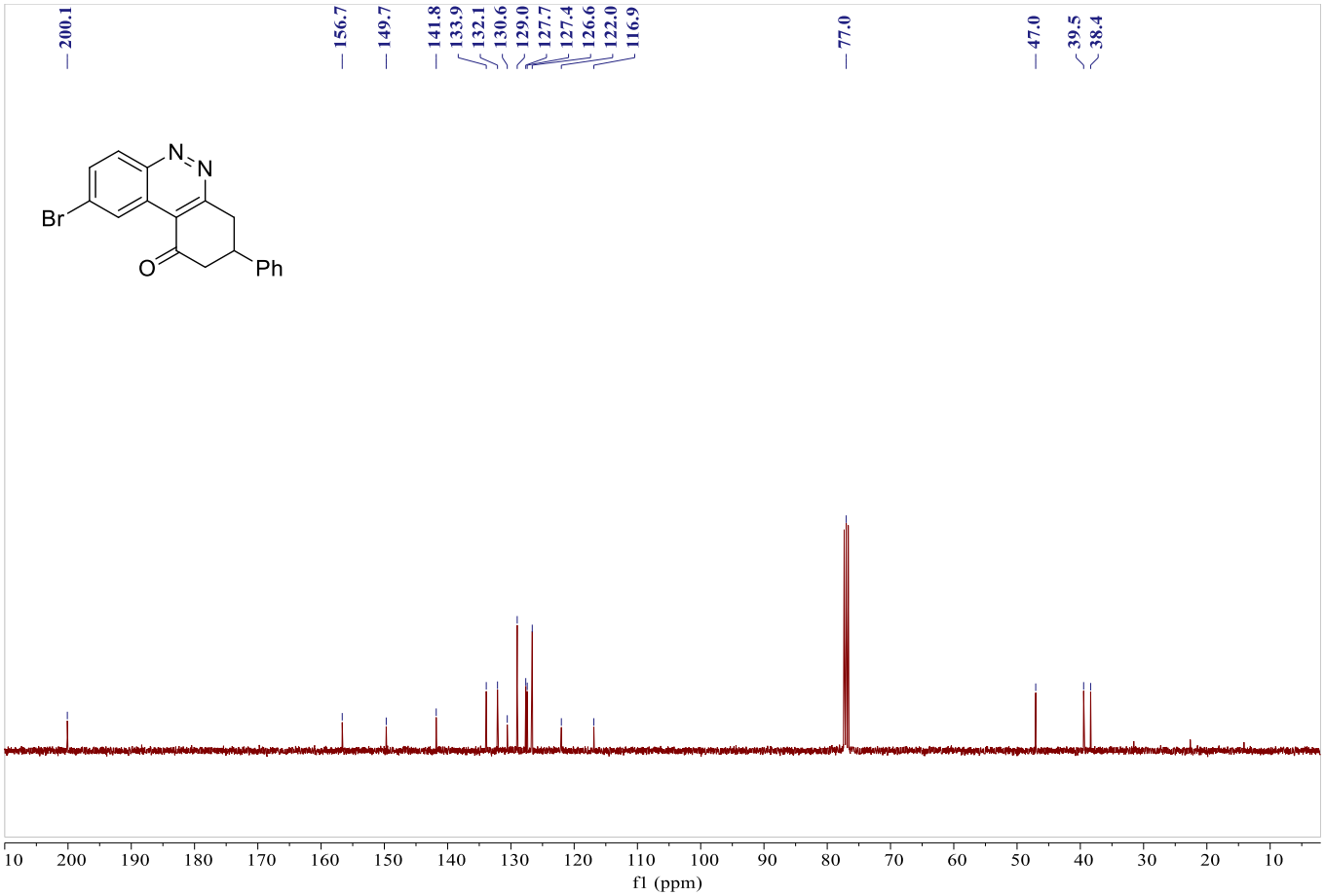
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3u**



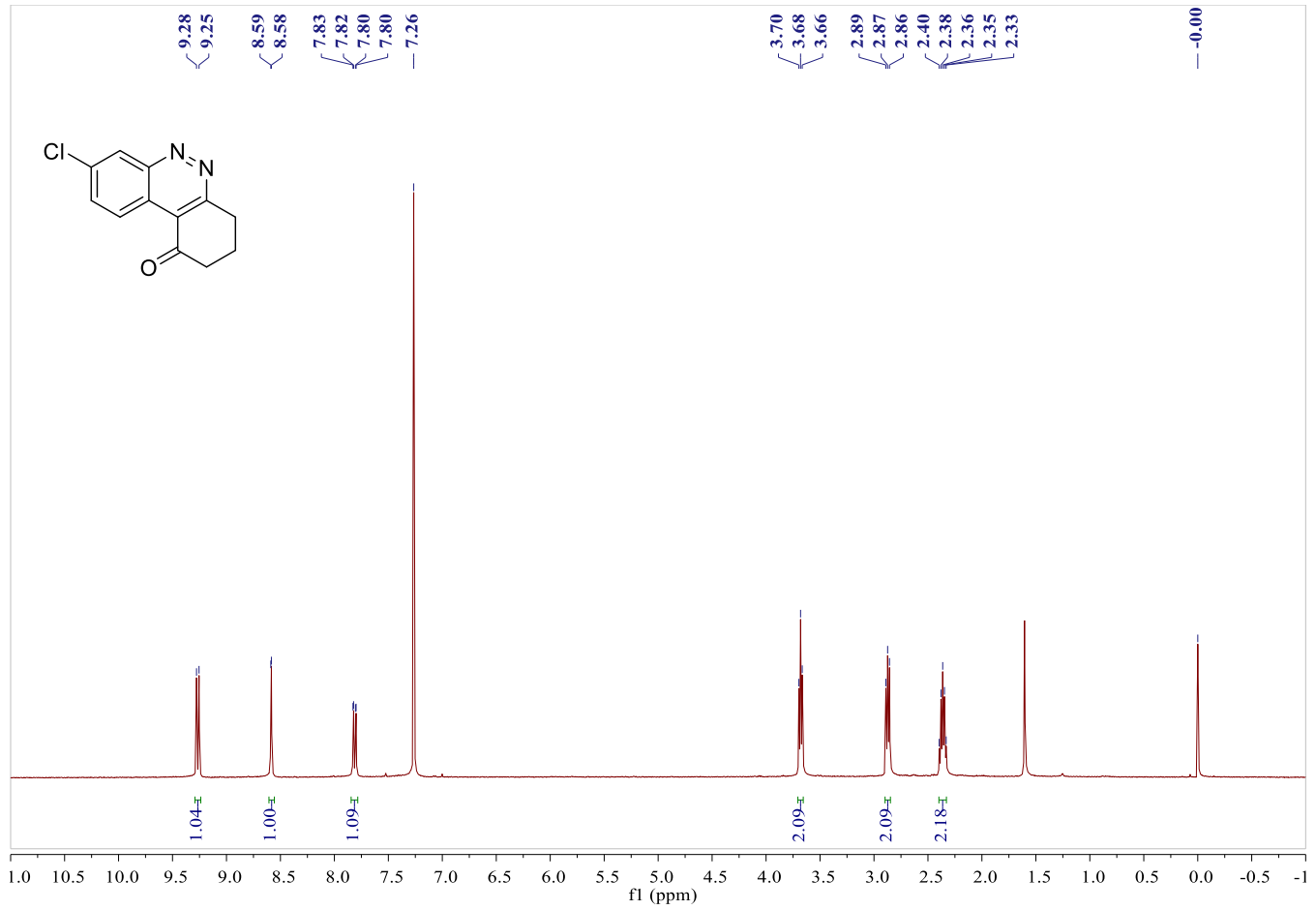



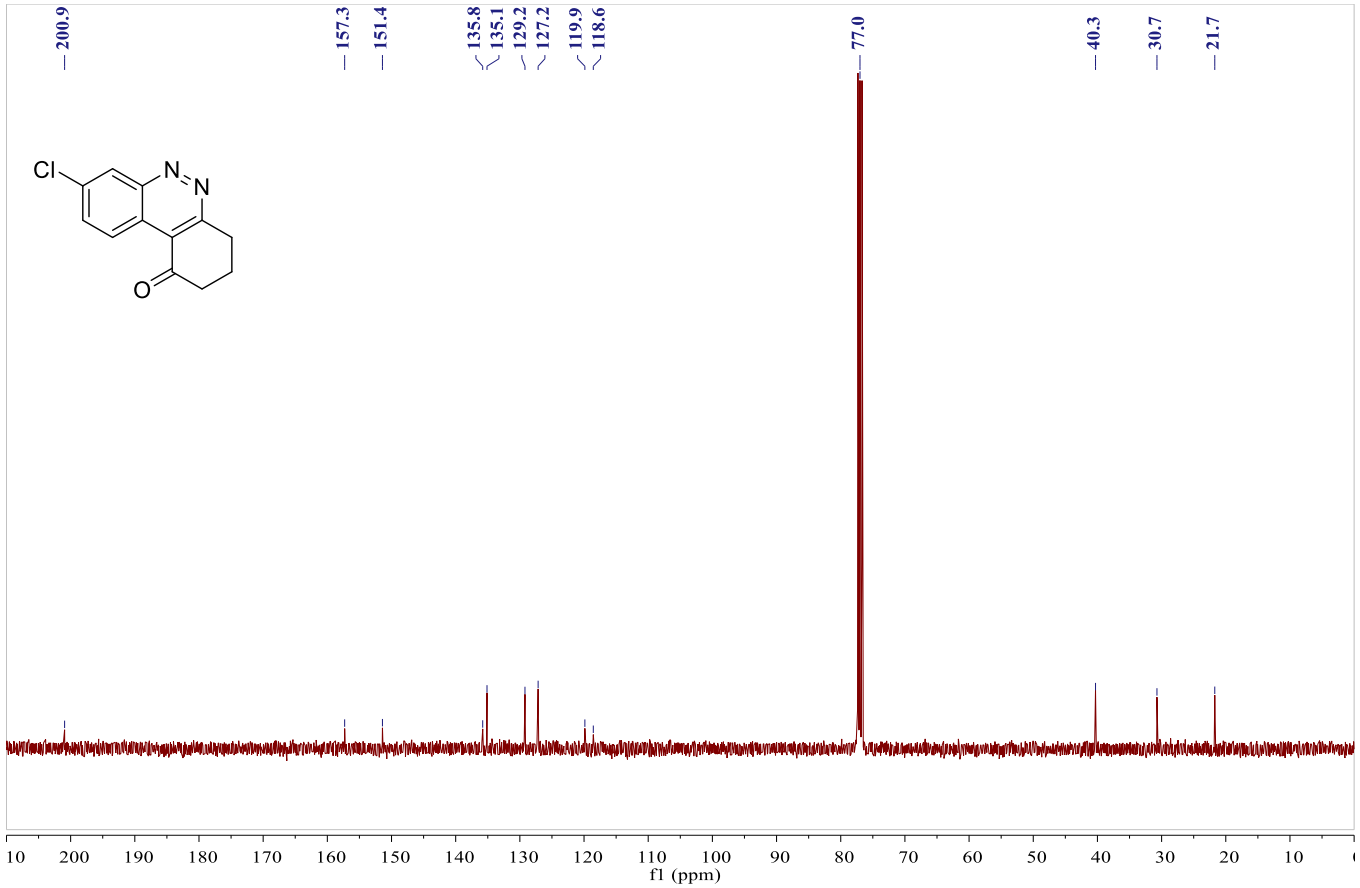
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3v**



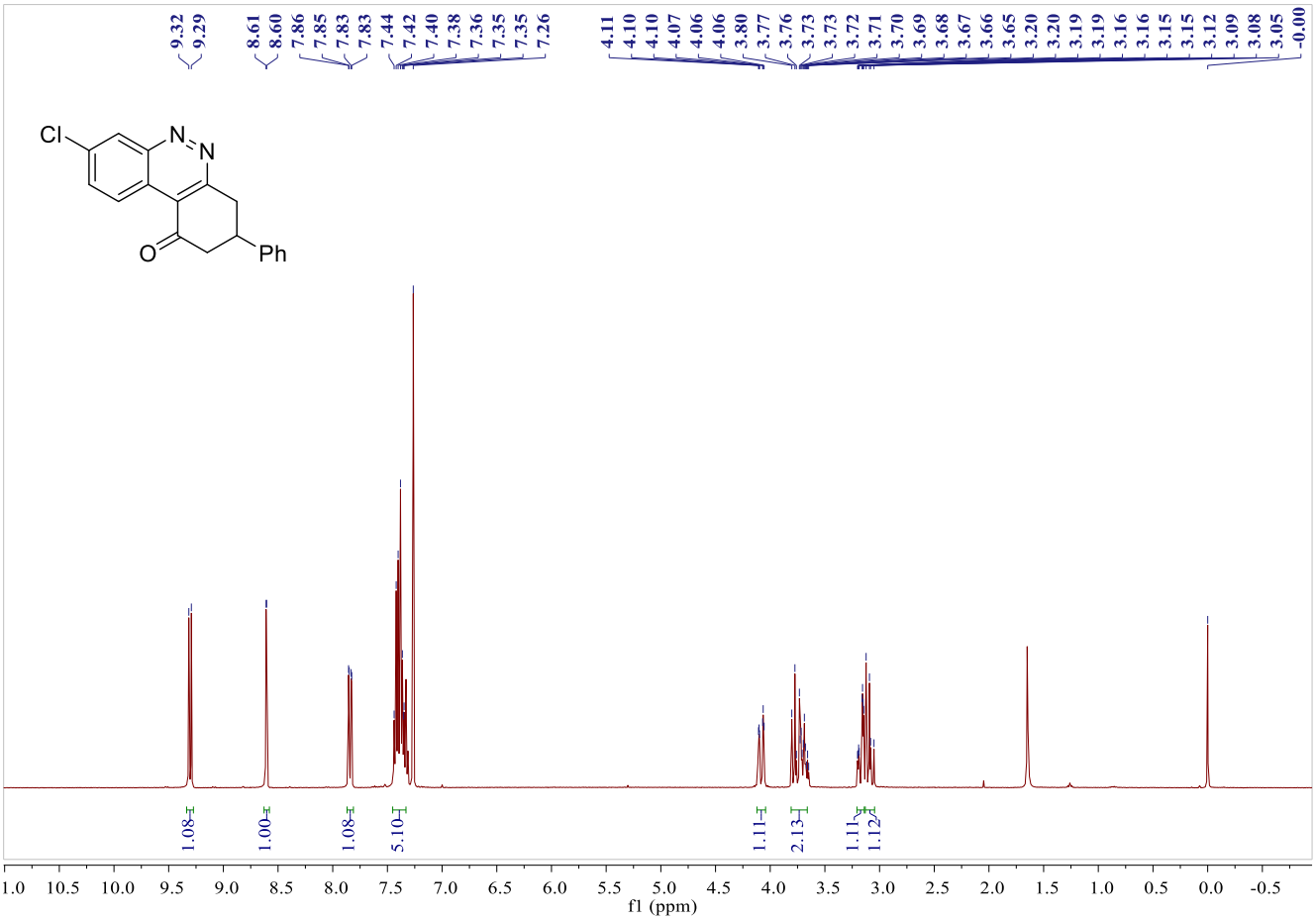



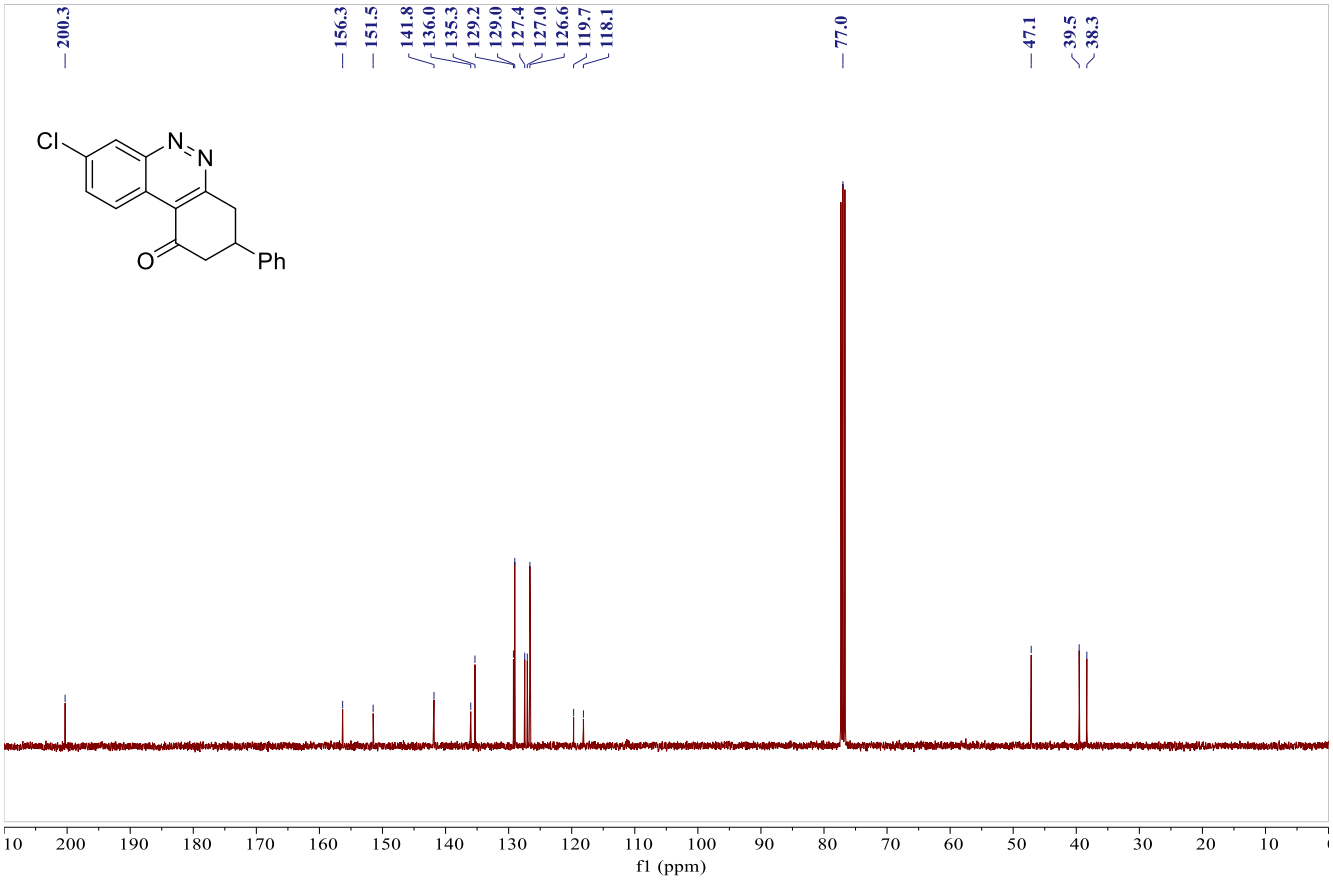
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3w**



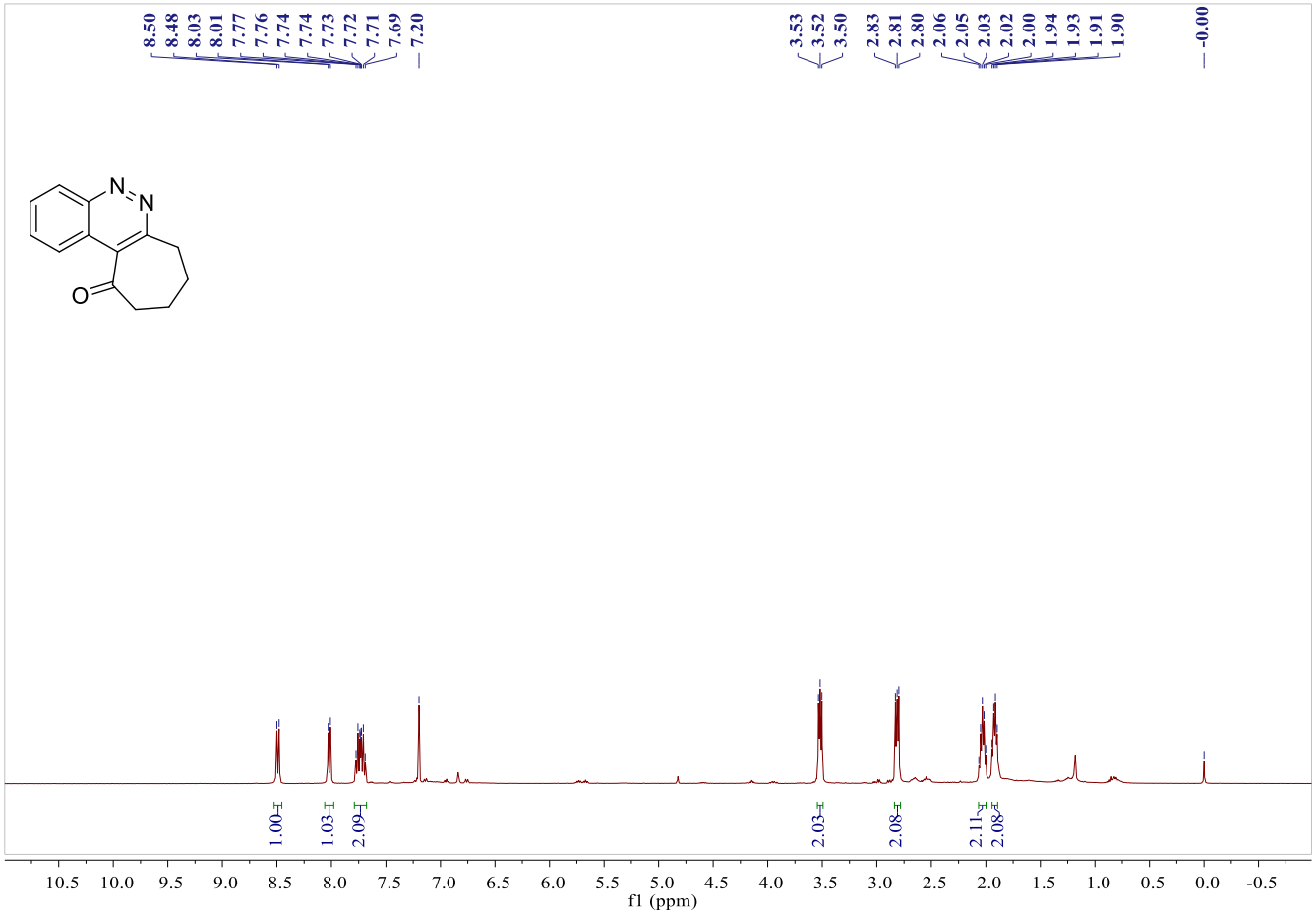



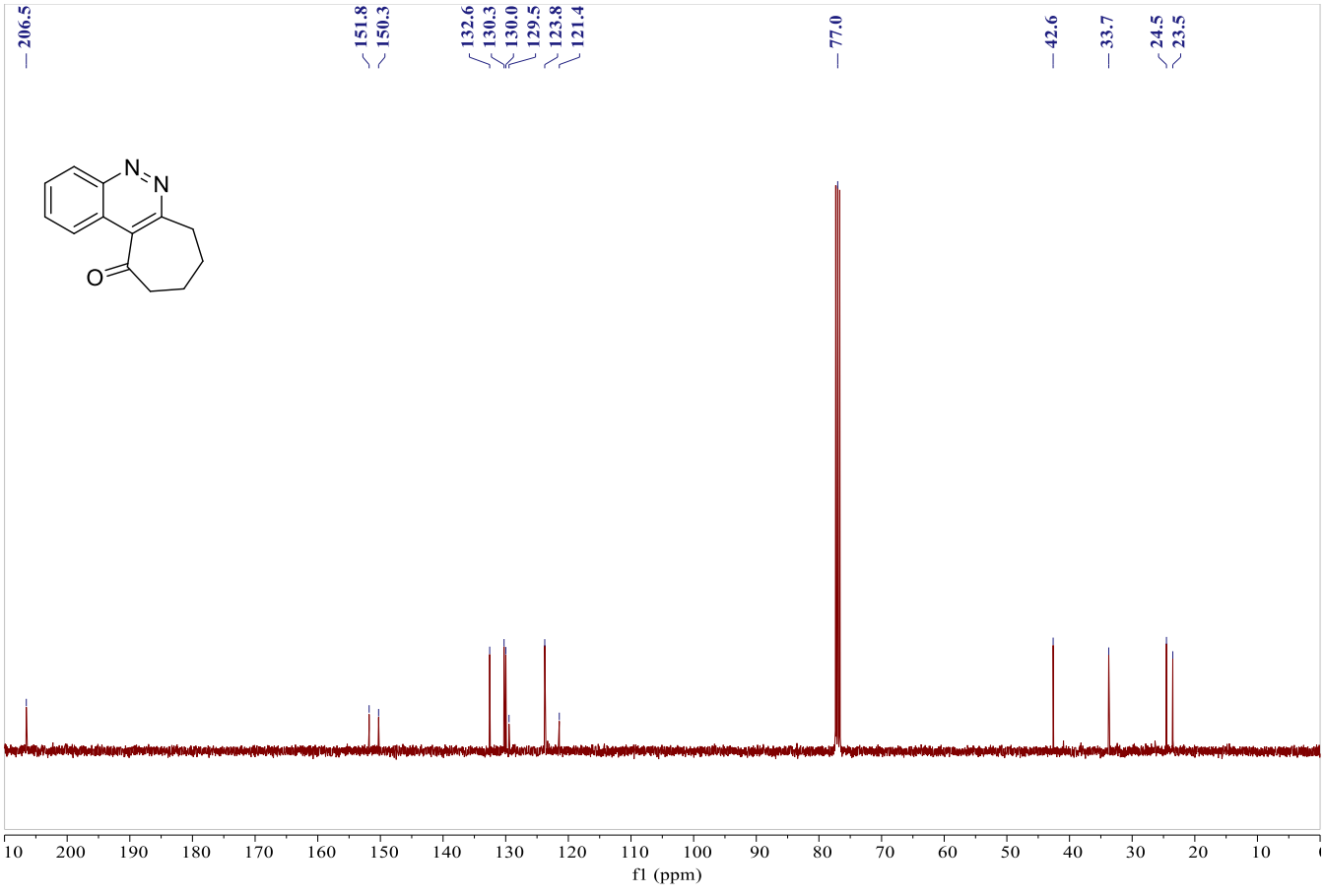
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3x**



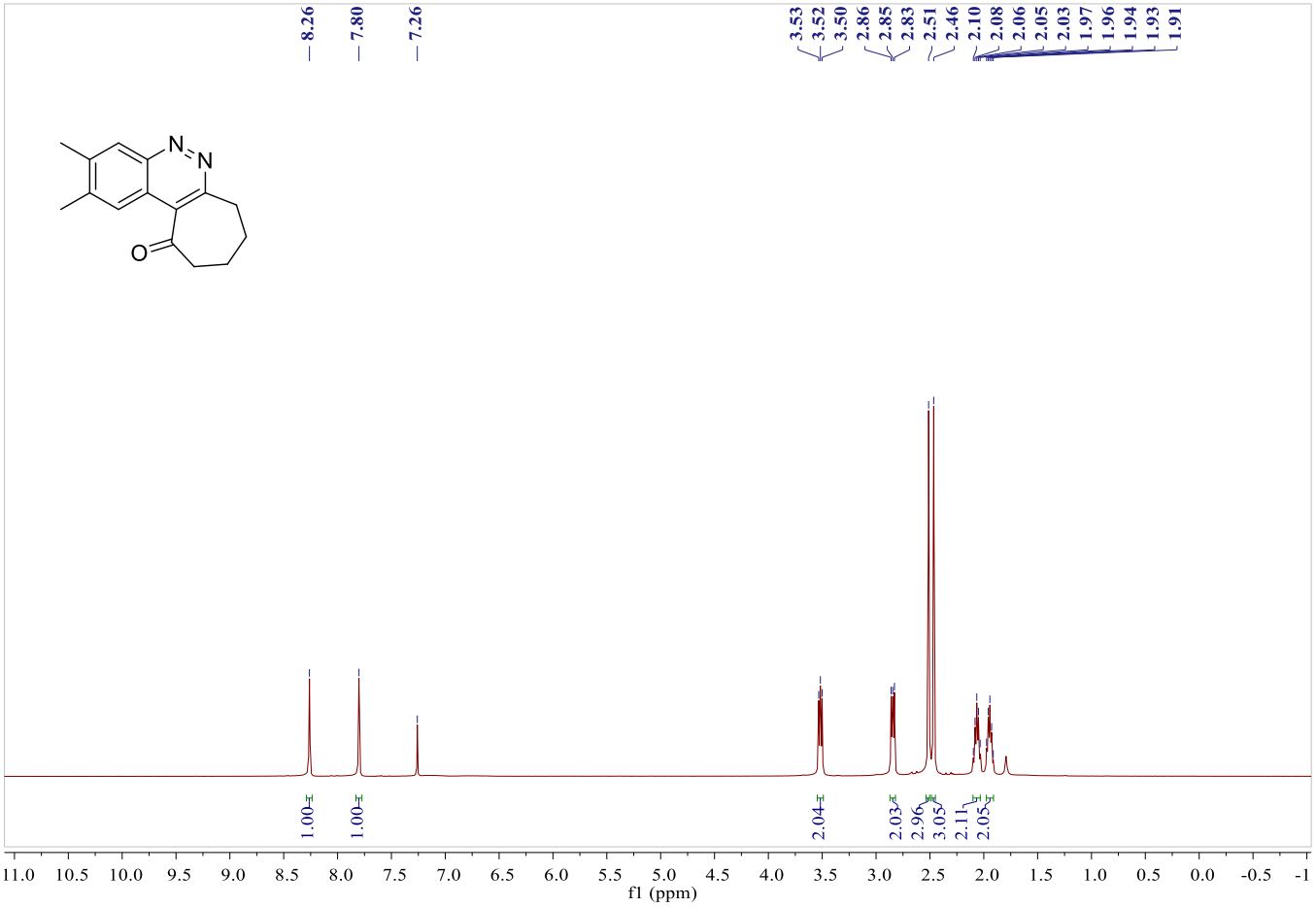



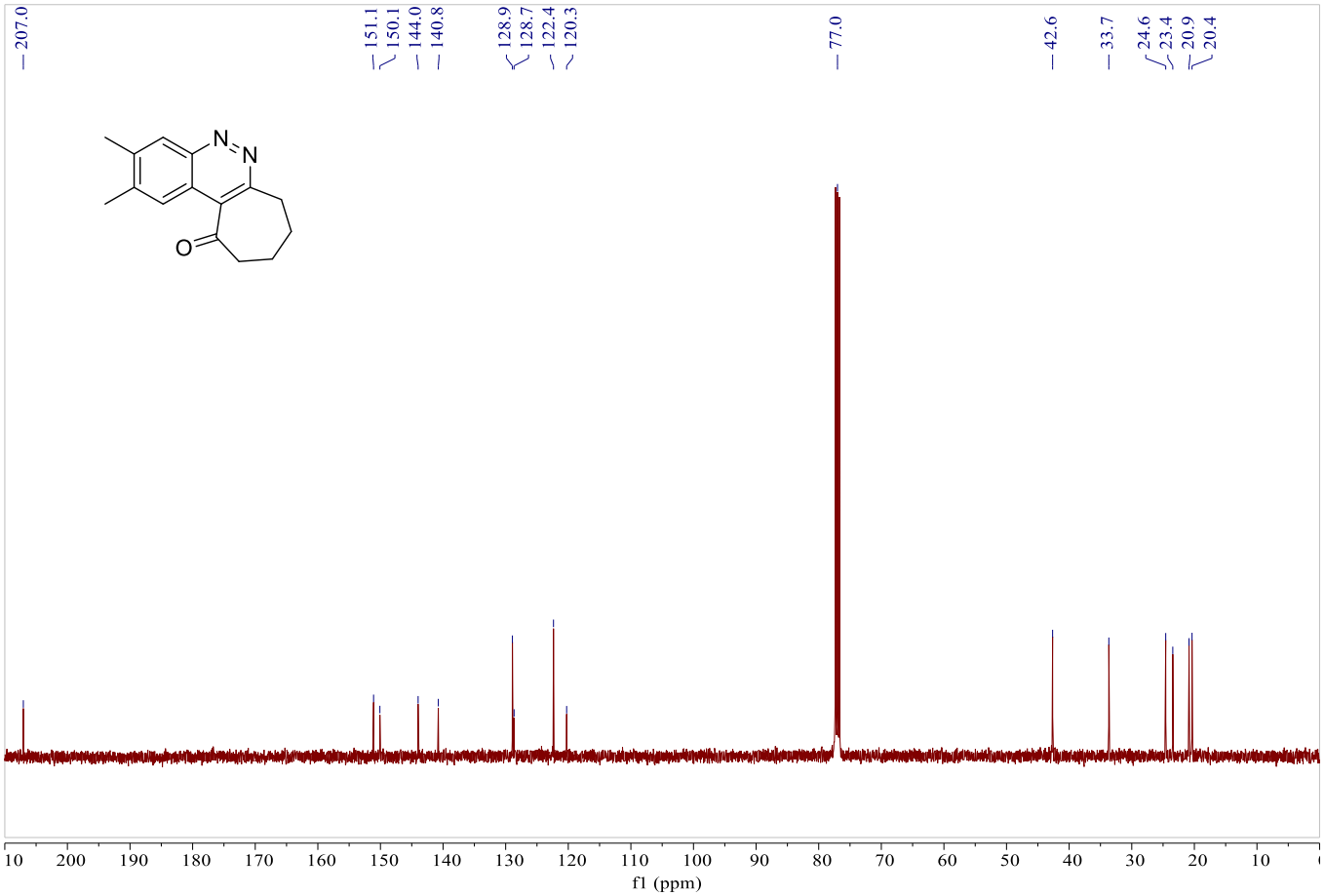
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3y**



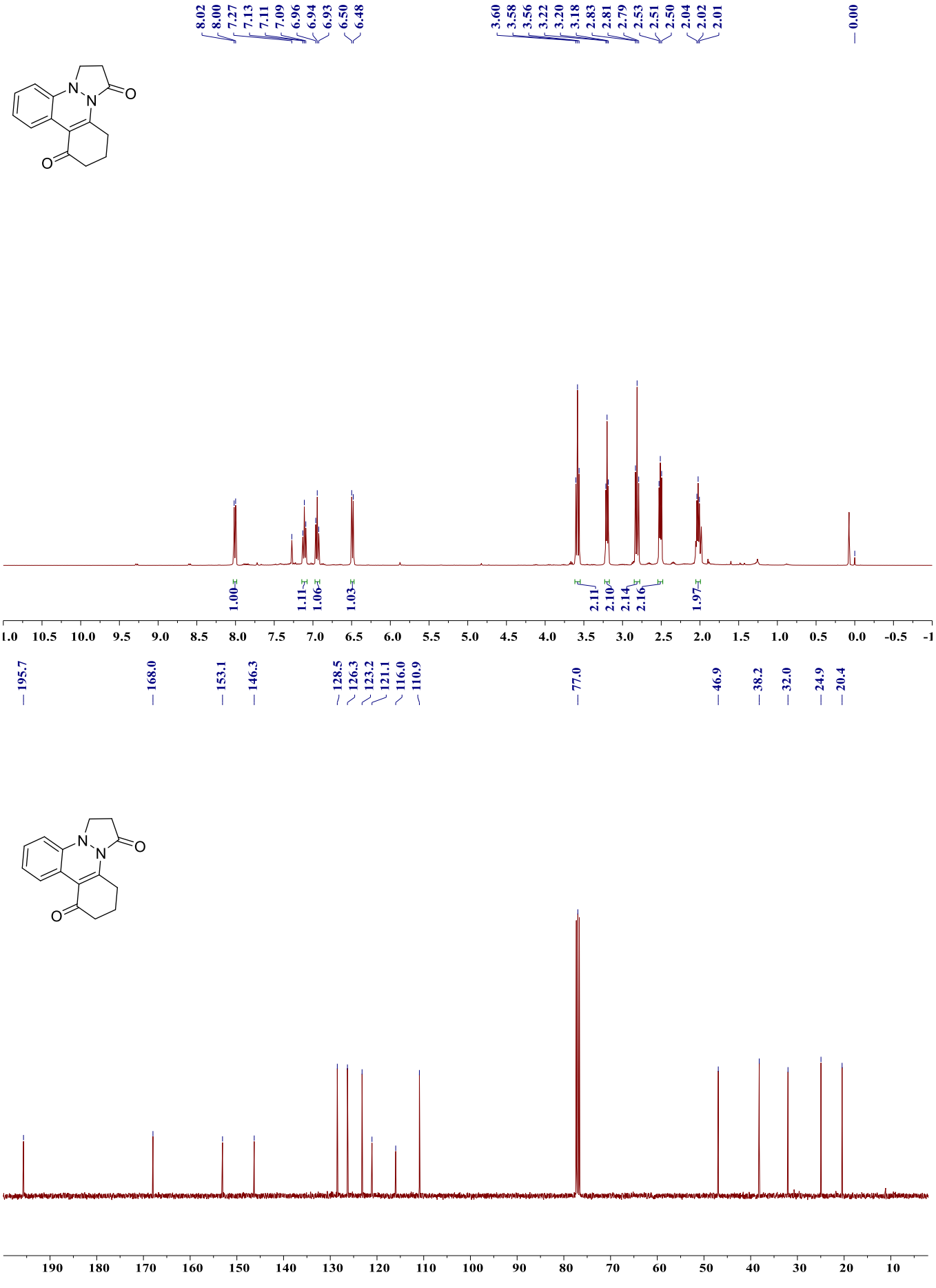



**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 3z**

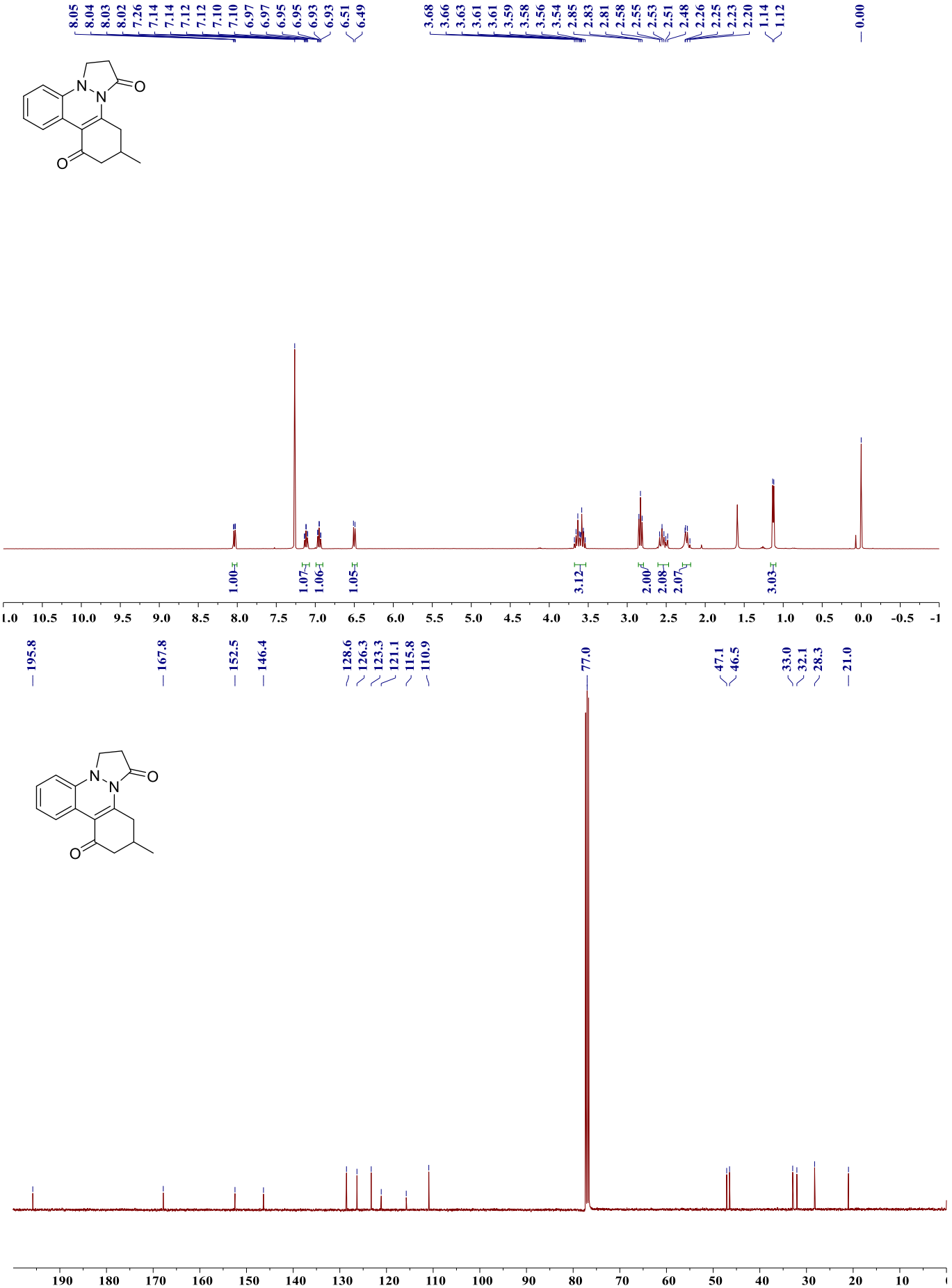


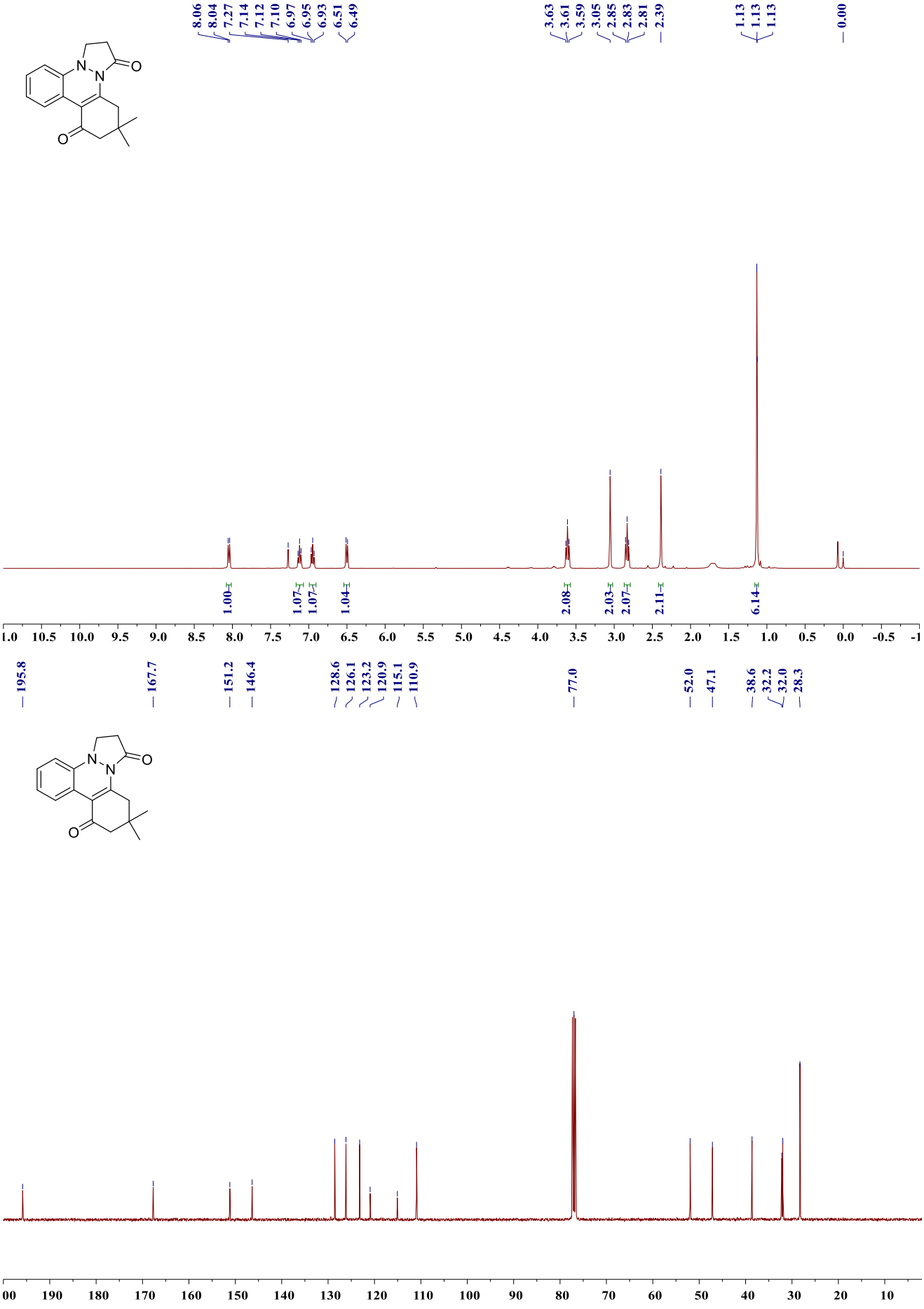
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5a**



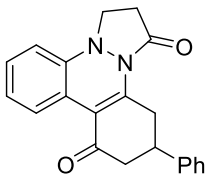
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5b**

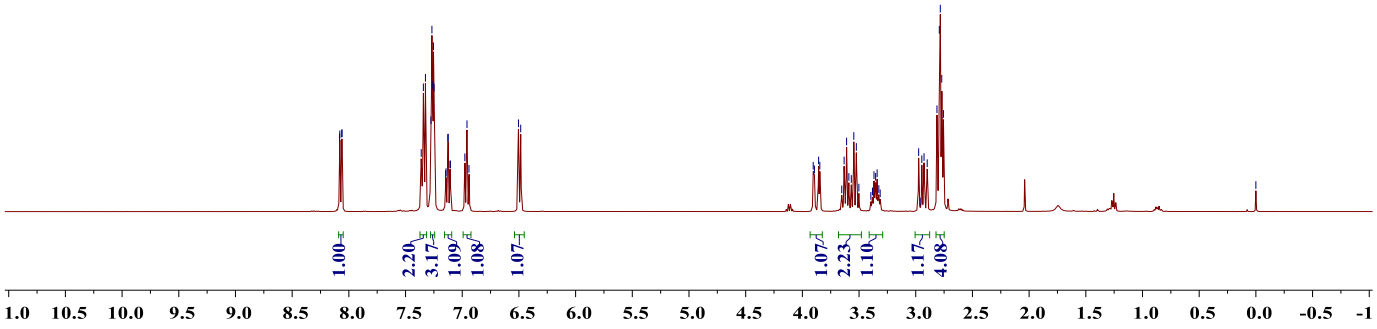


**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5c**

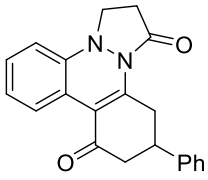


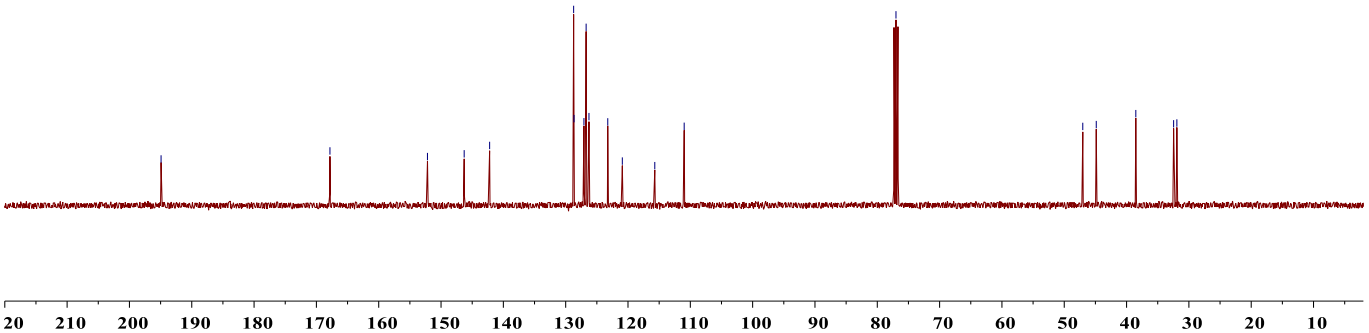
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5d**

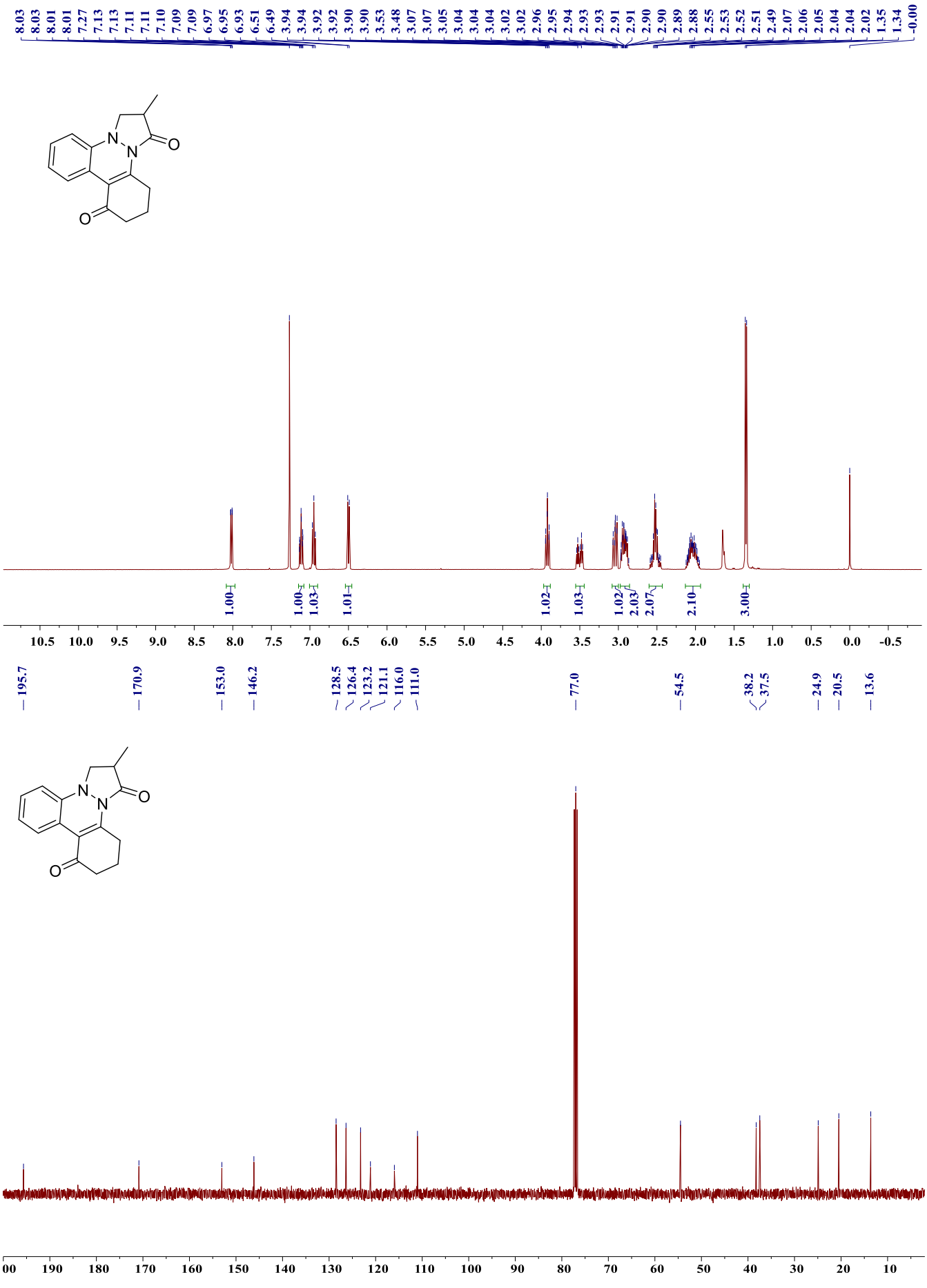





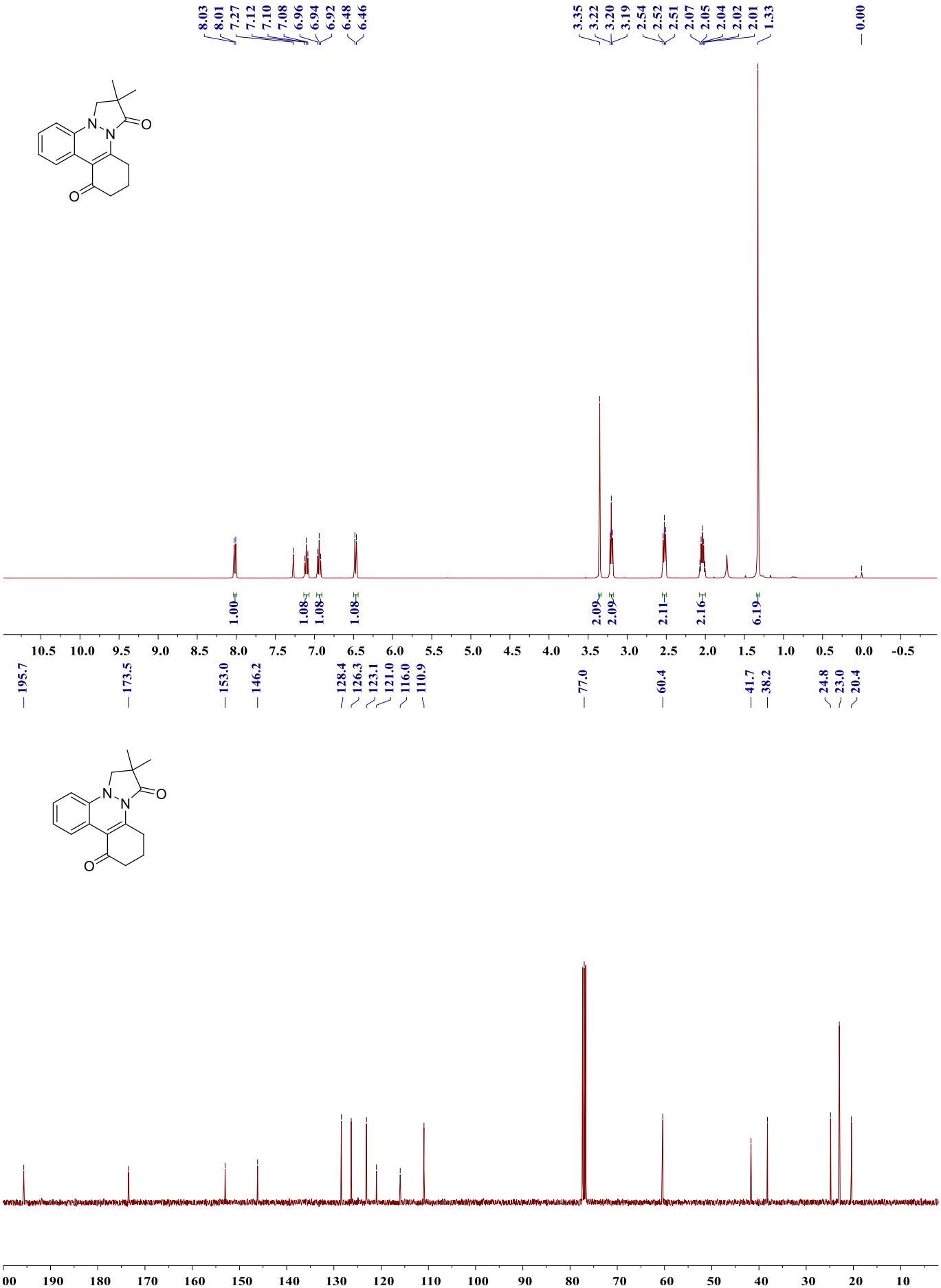


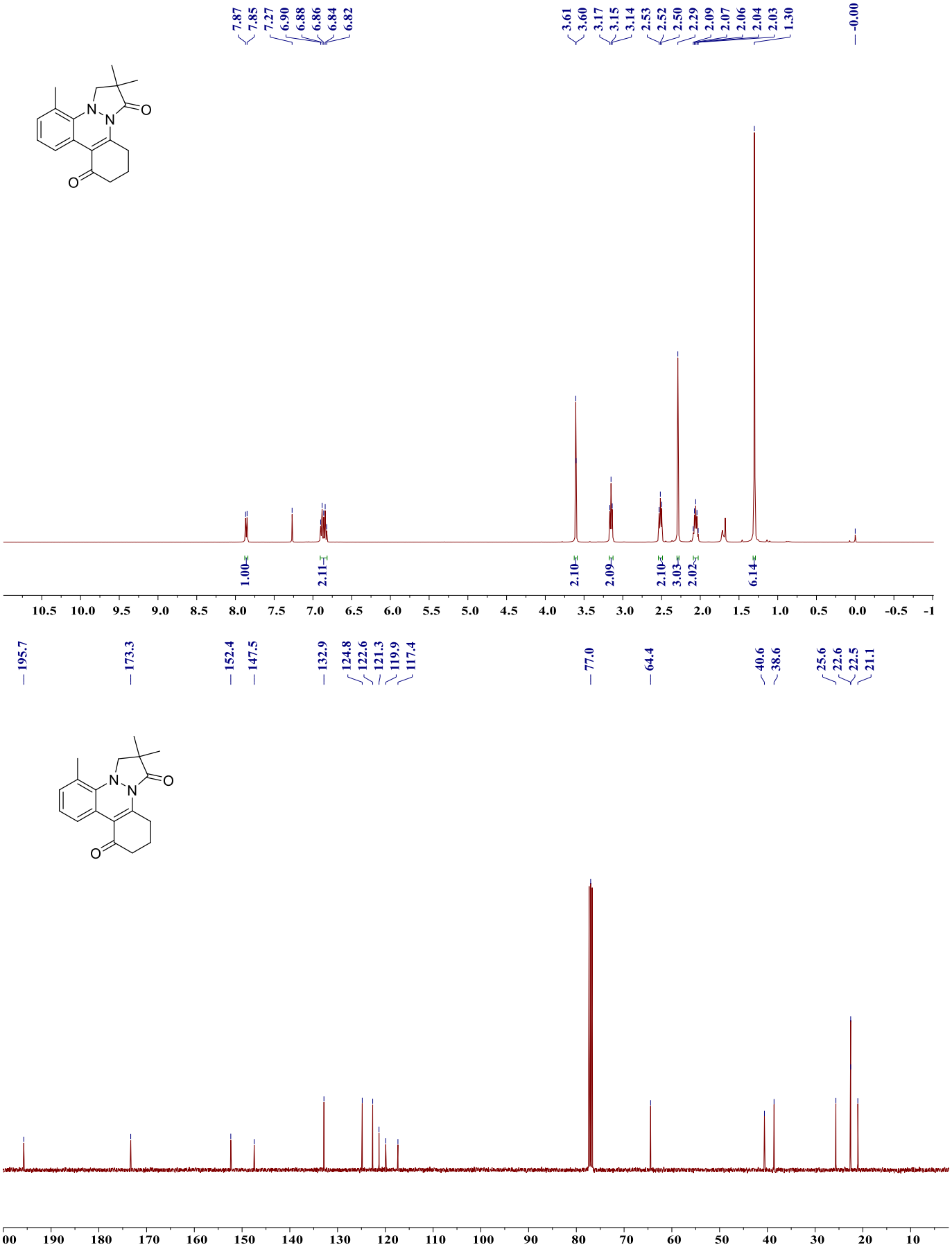
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5e**



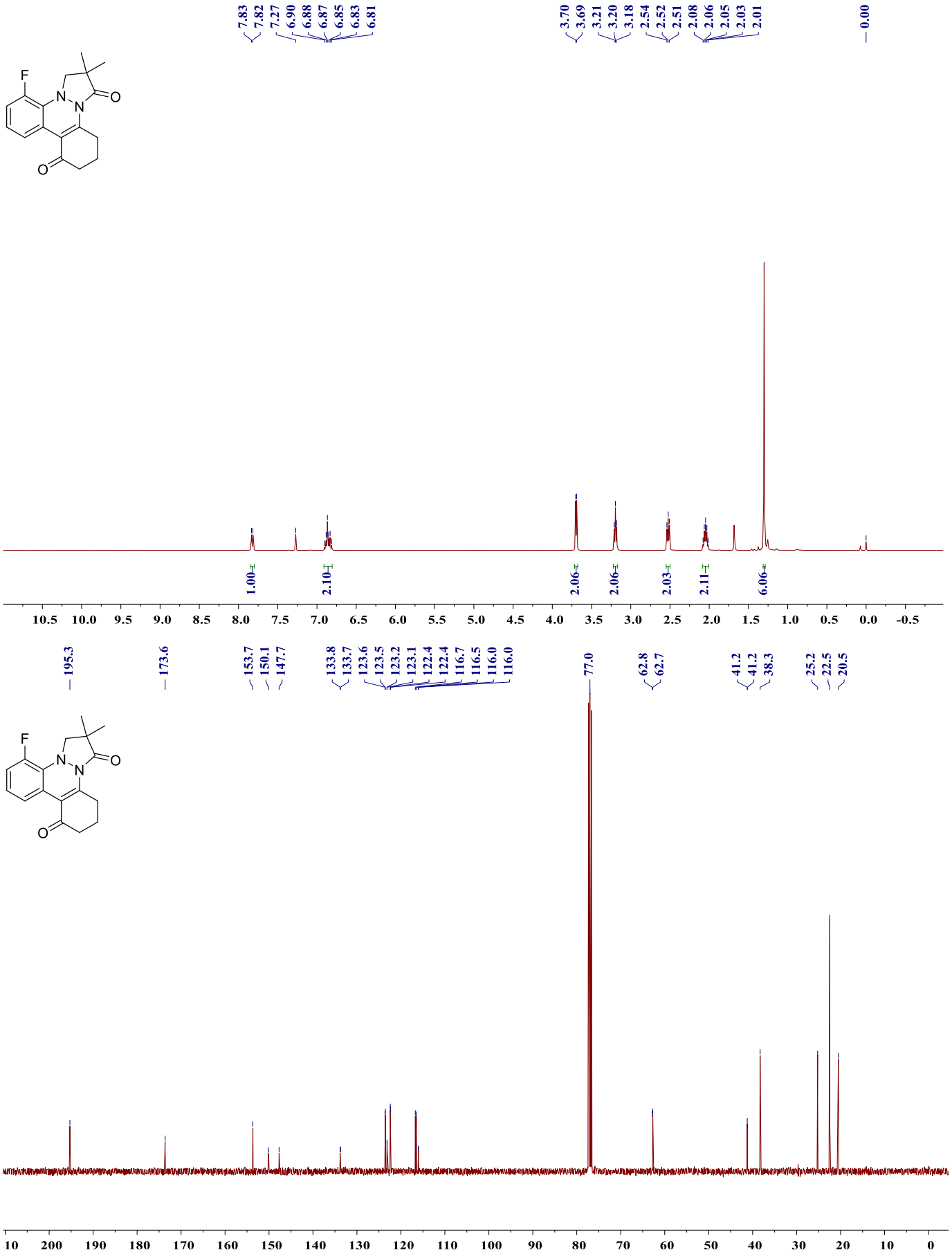
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5f**



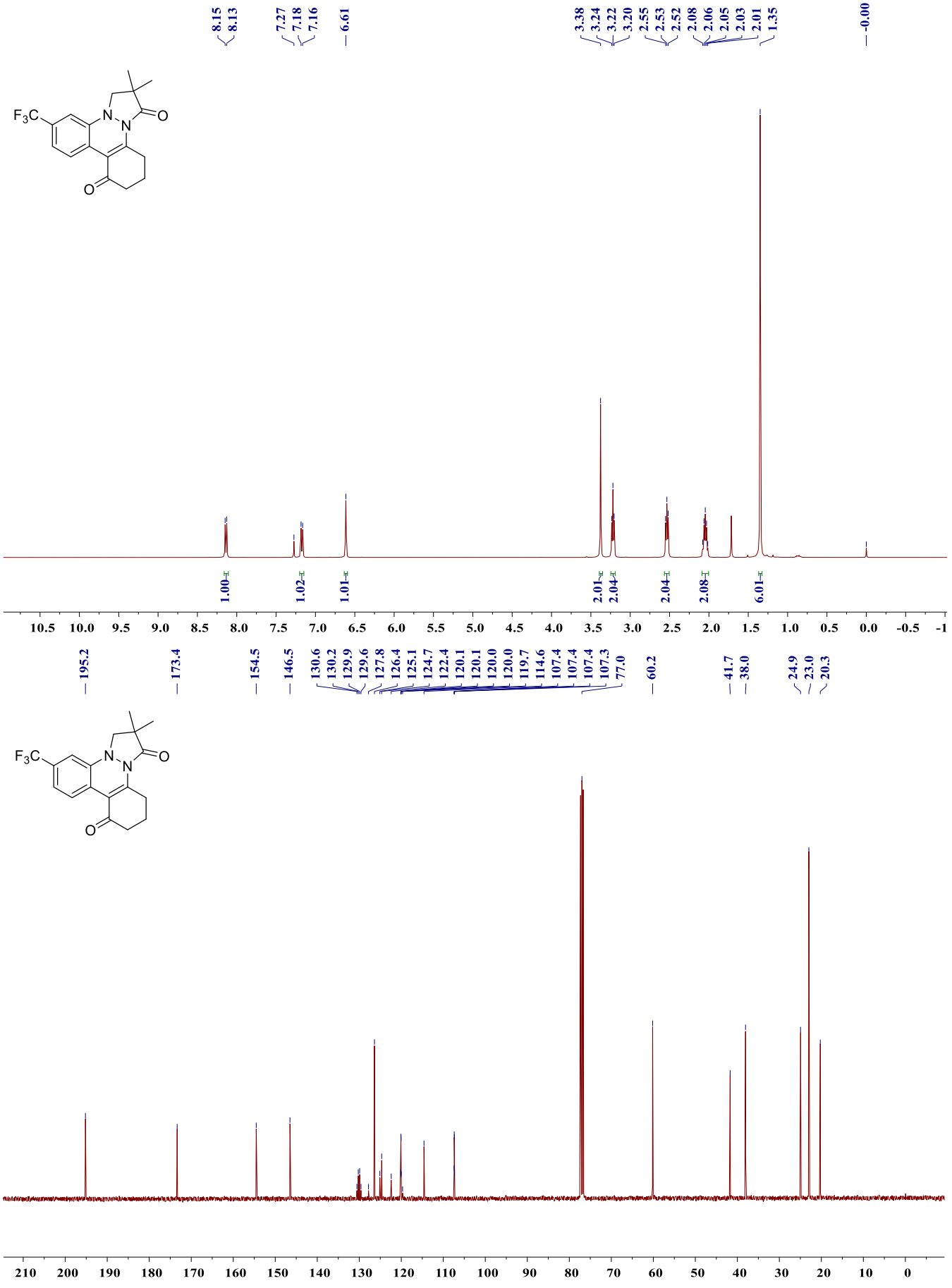
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5g**



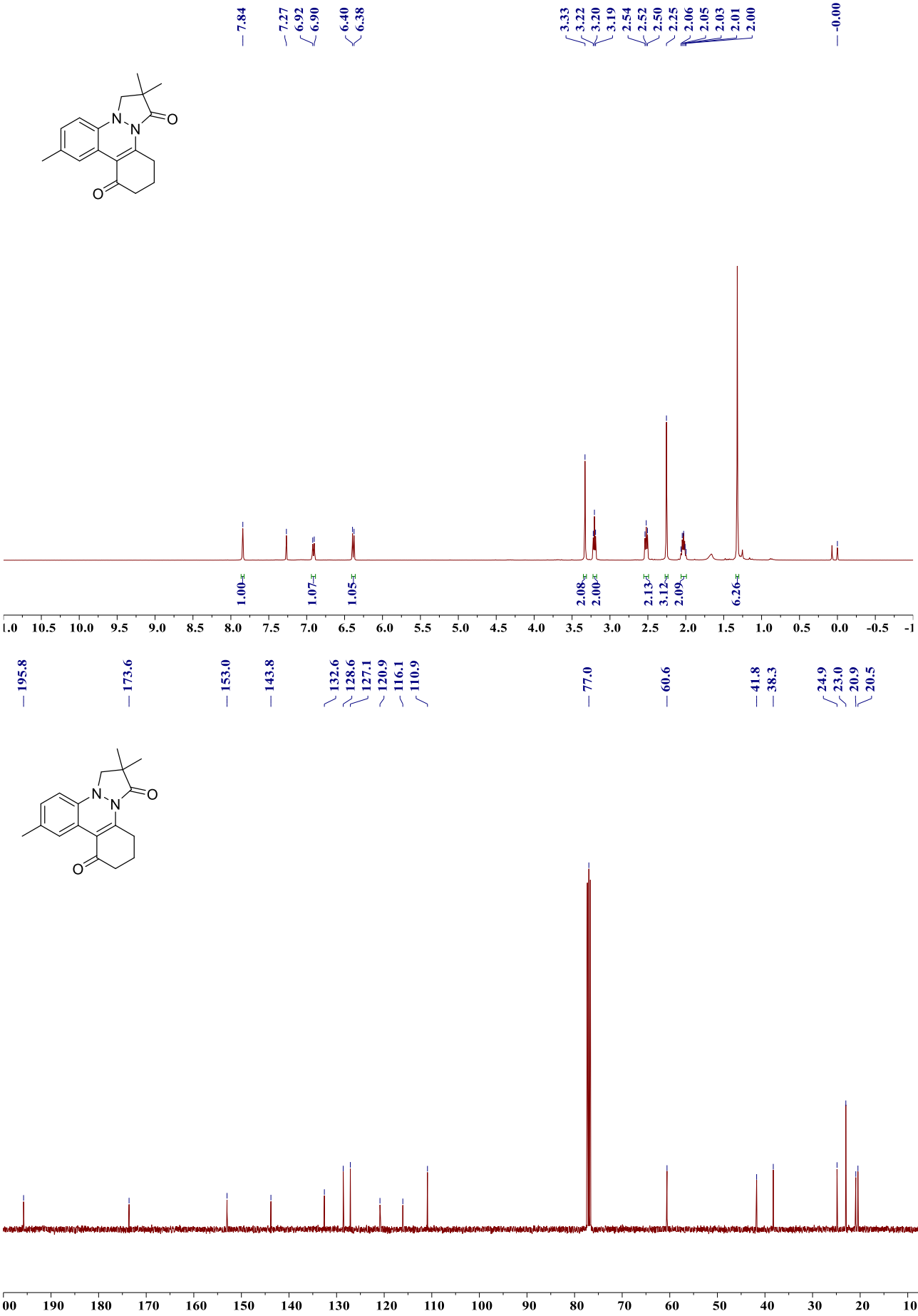
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5h**



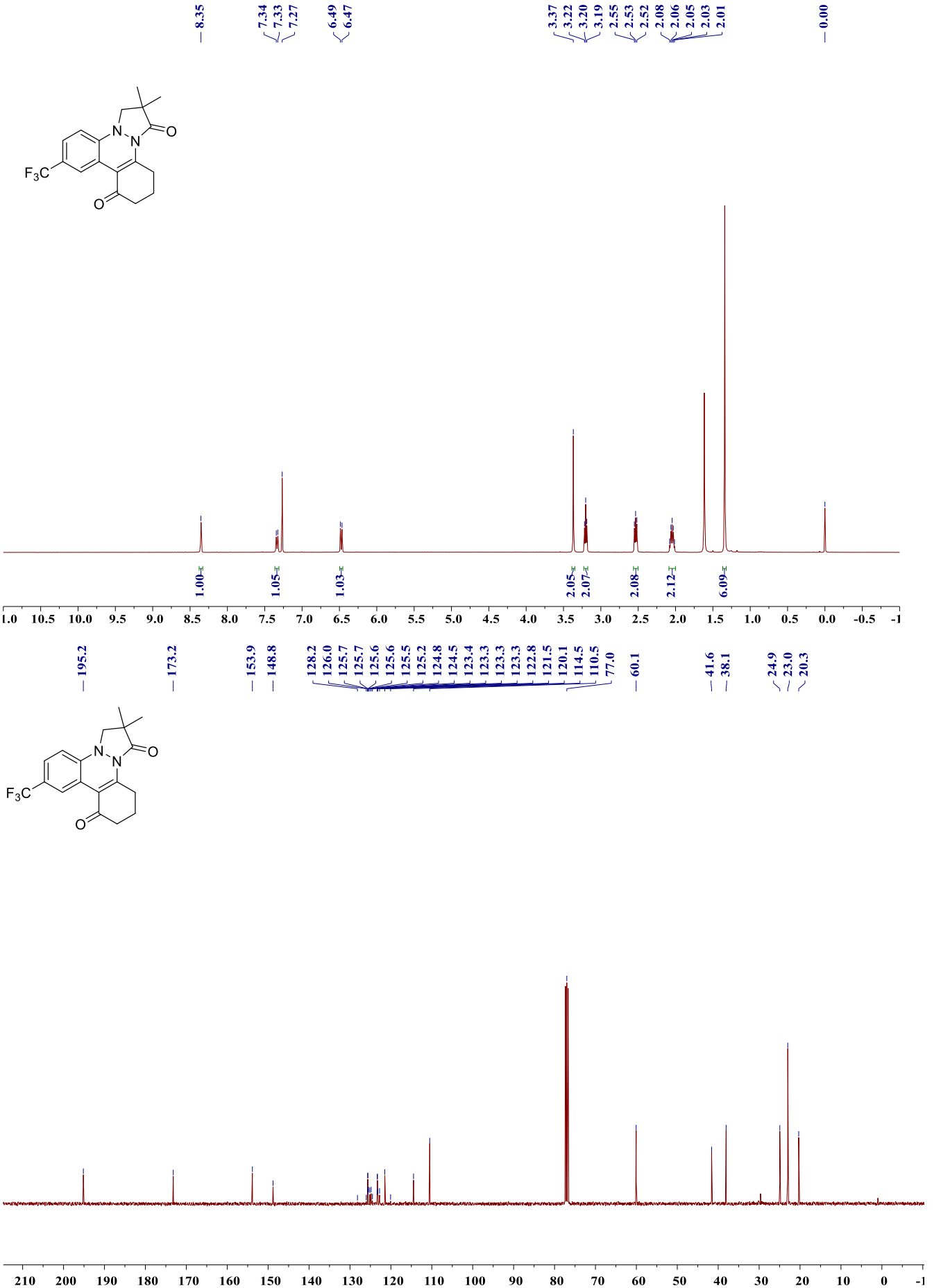
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5i**



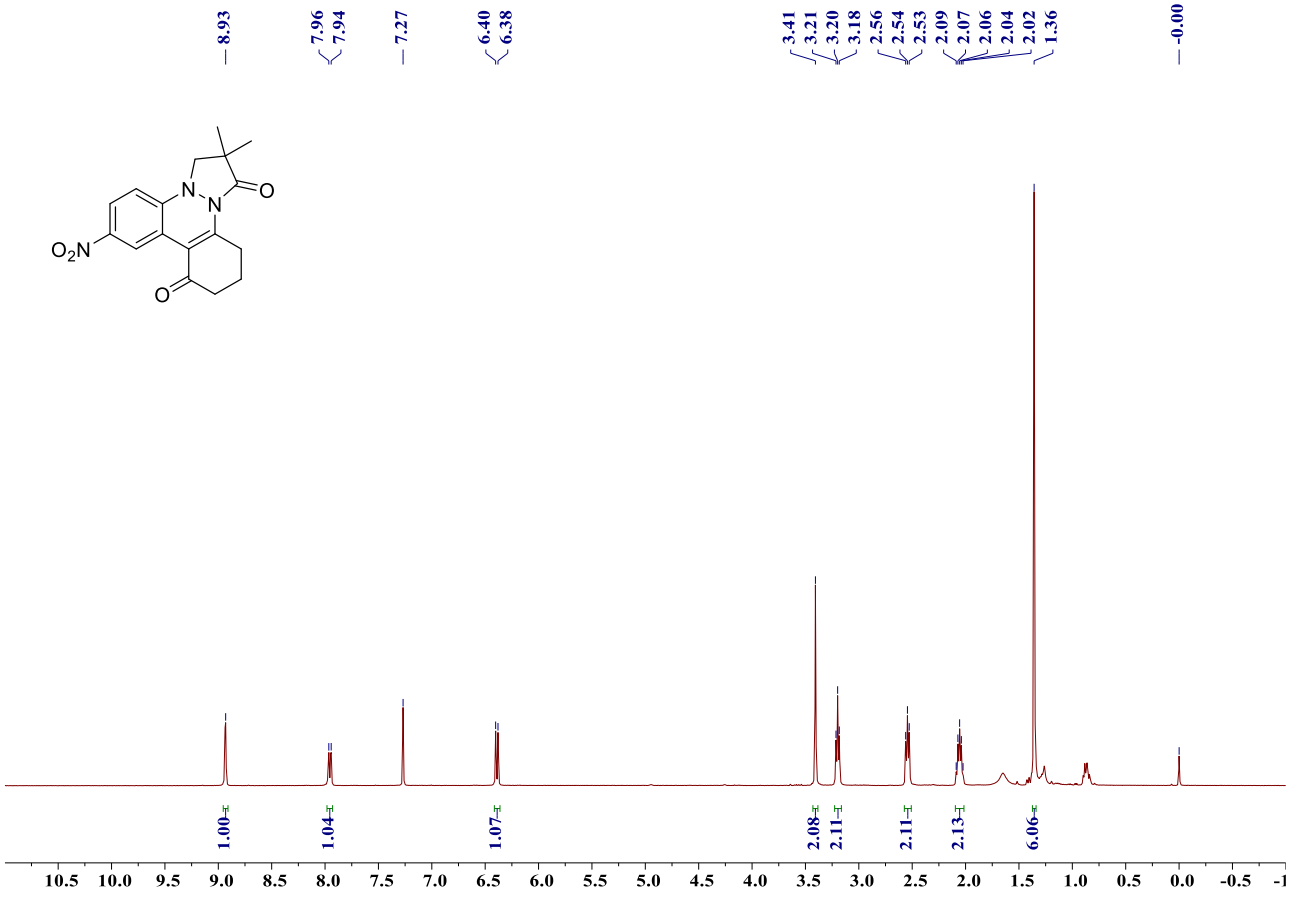
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5j**

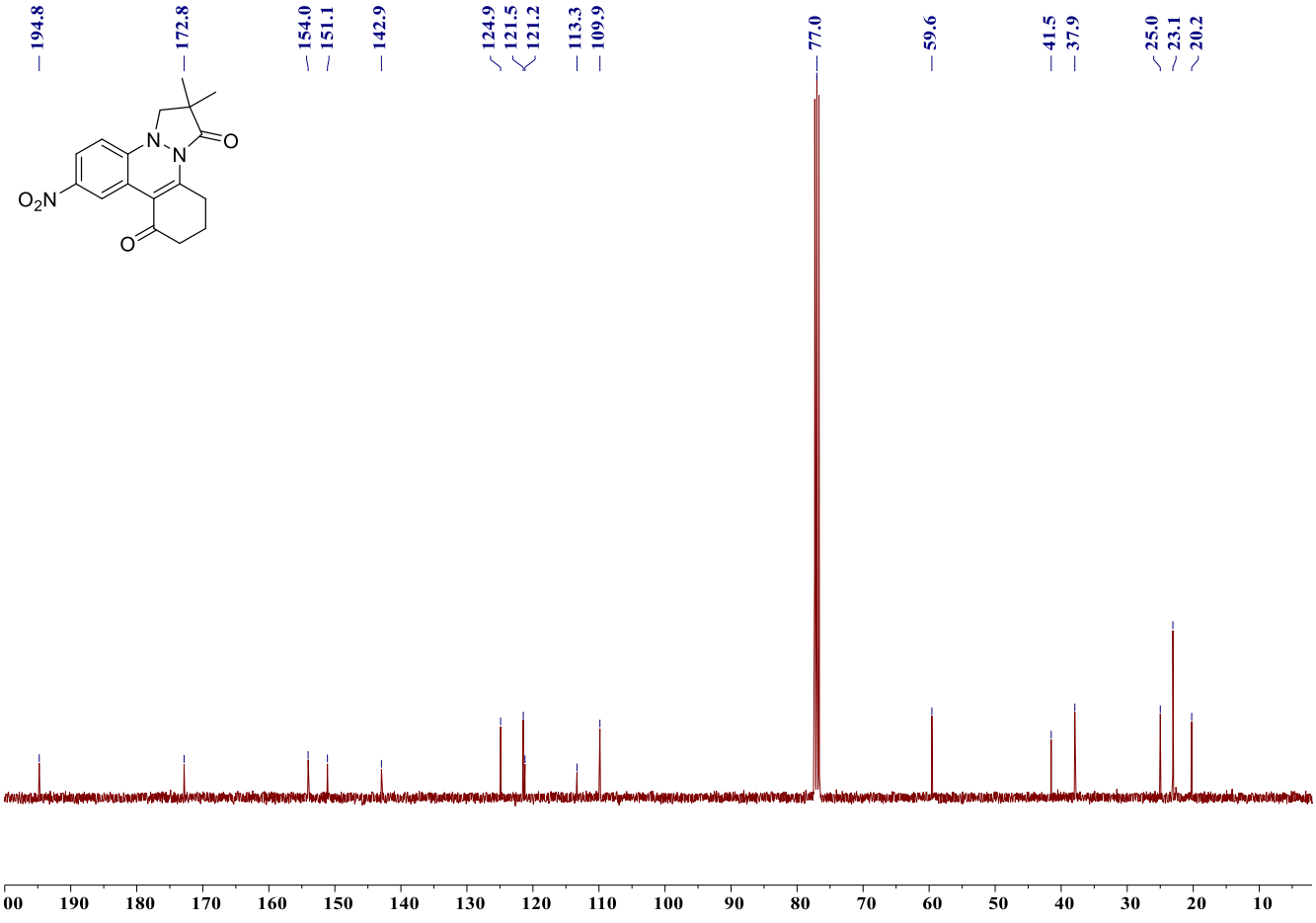


**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5k**

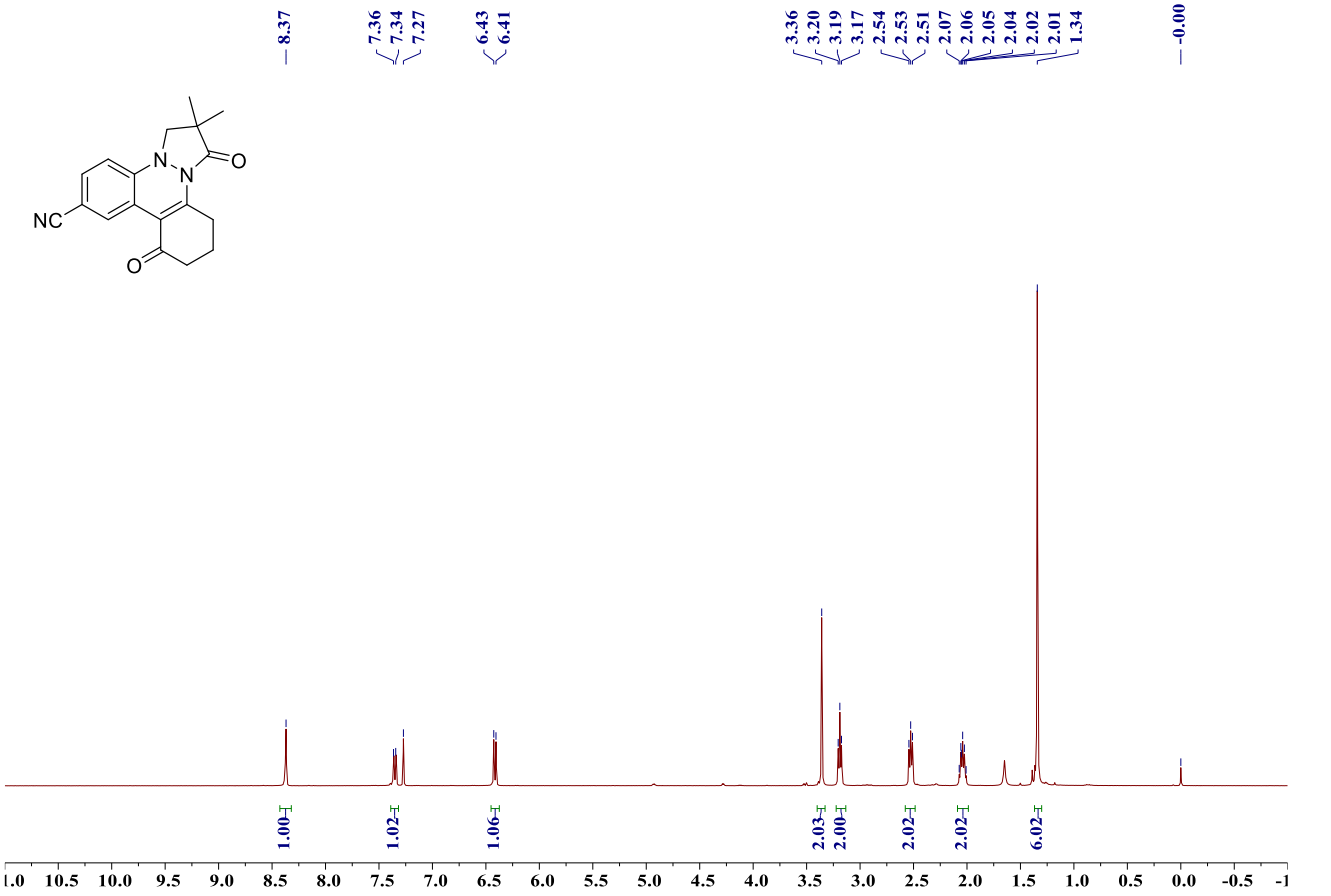


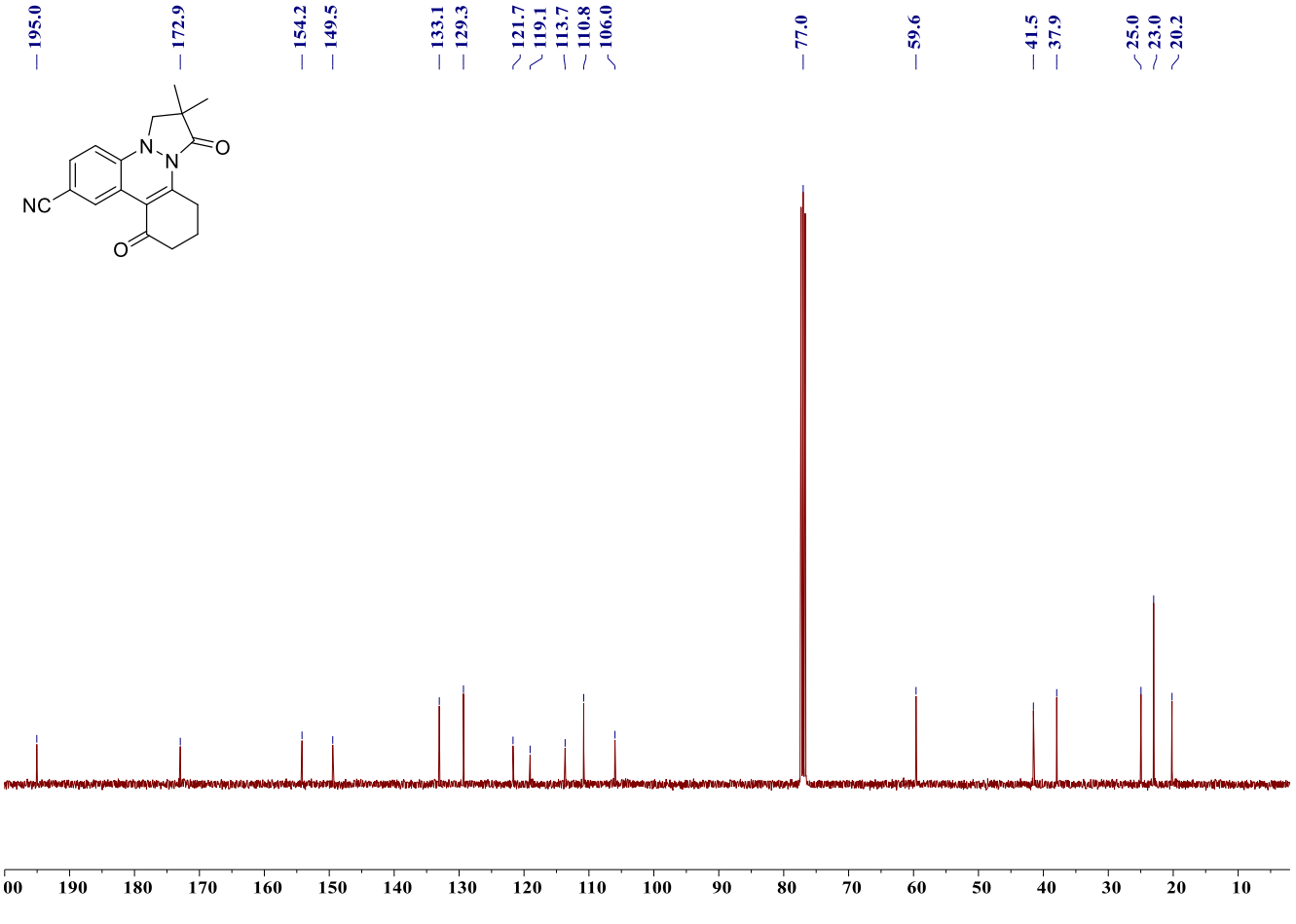
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5l**



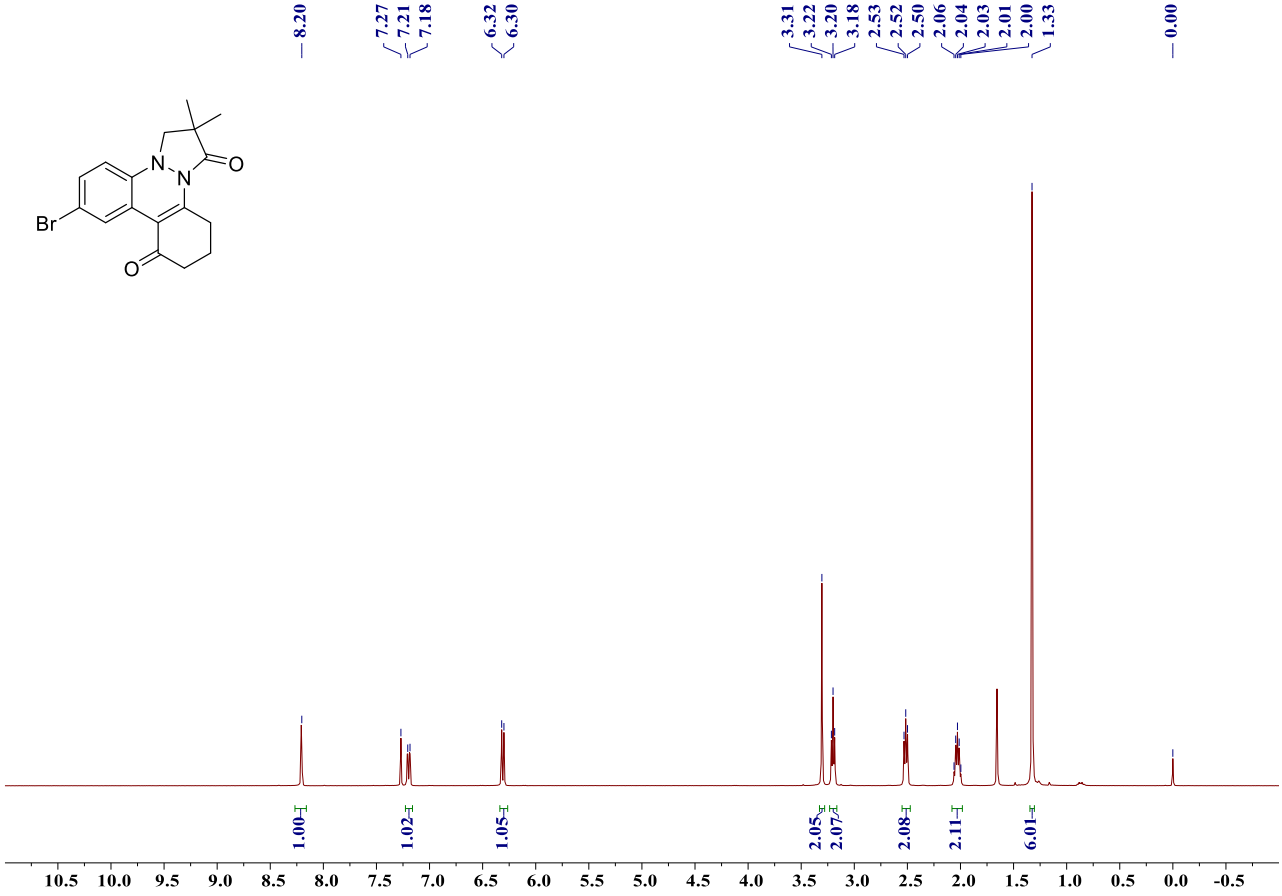



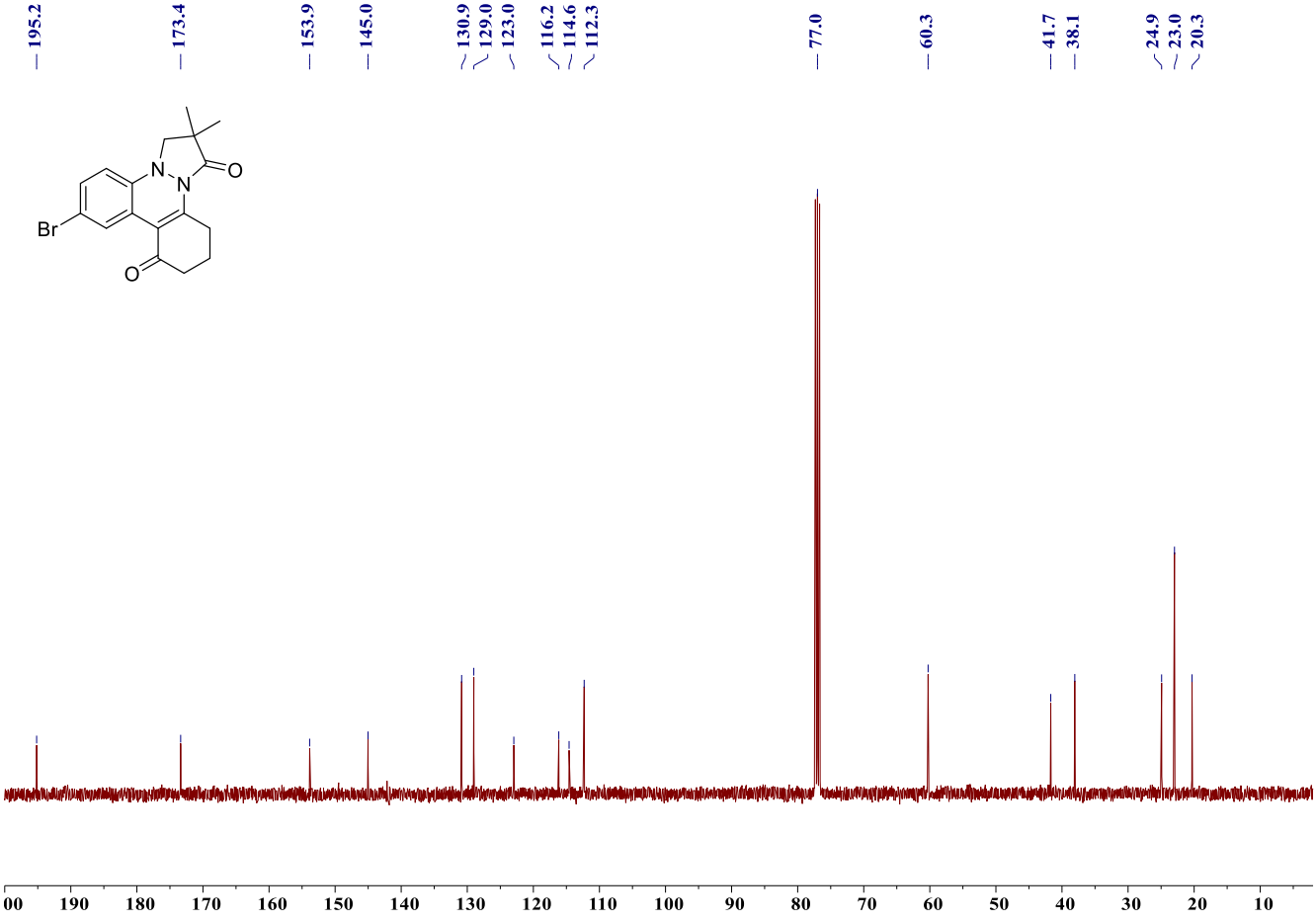
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5m**



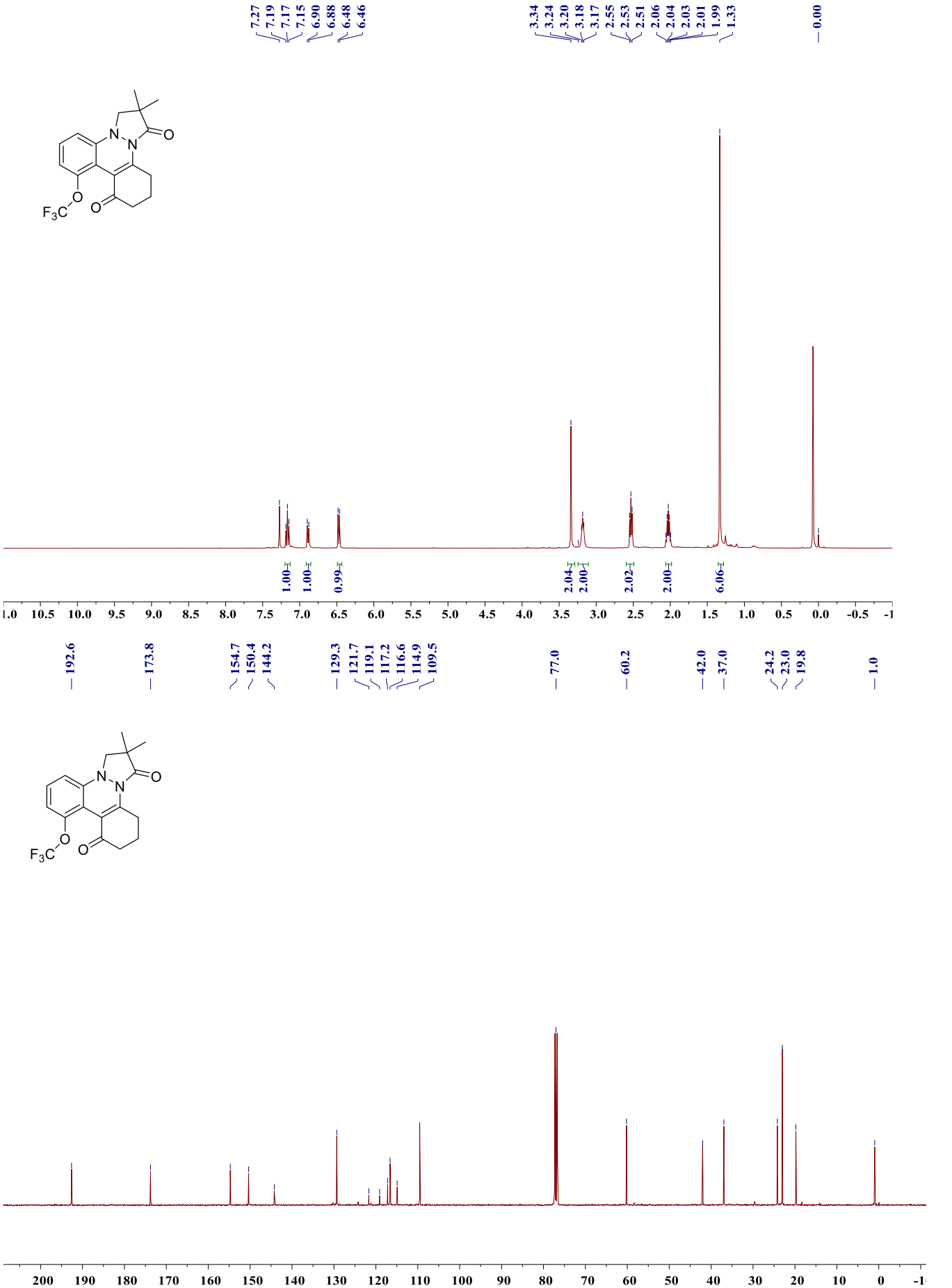



**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5n**

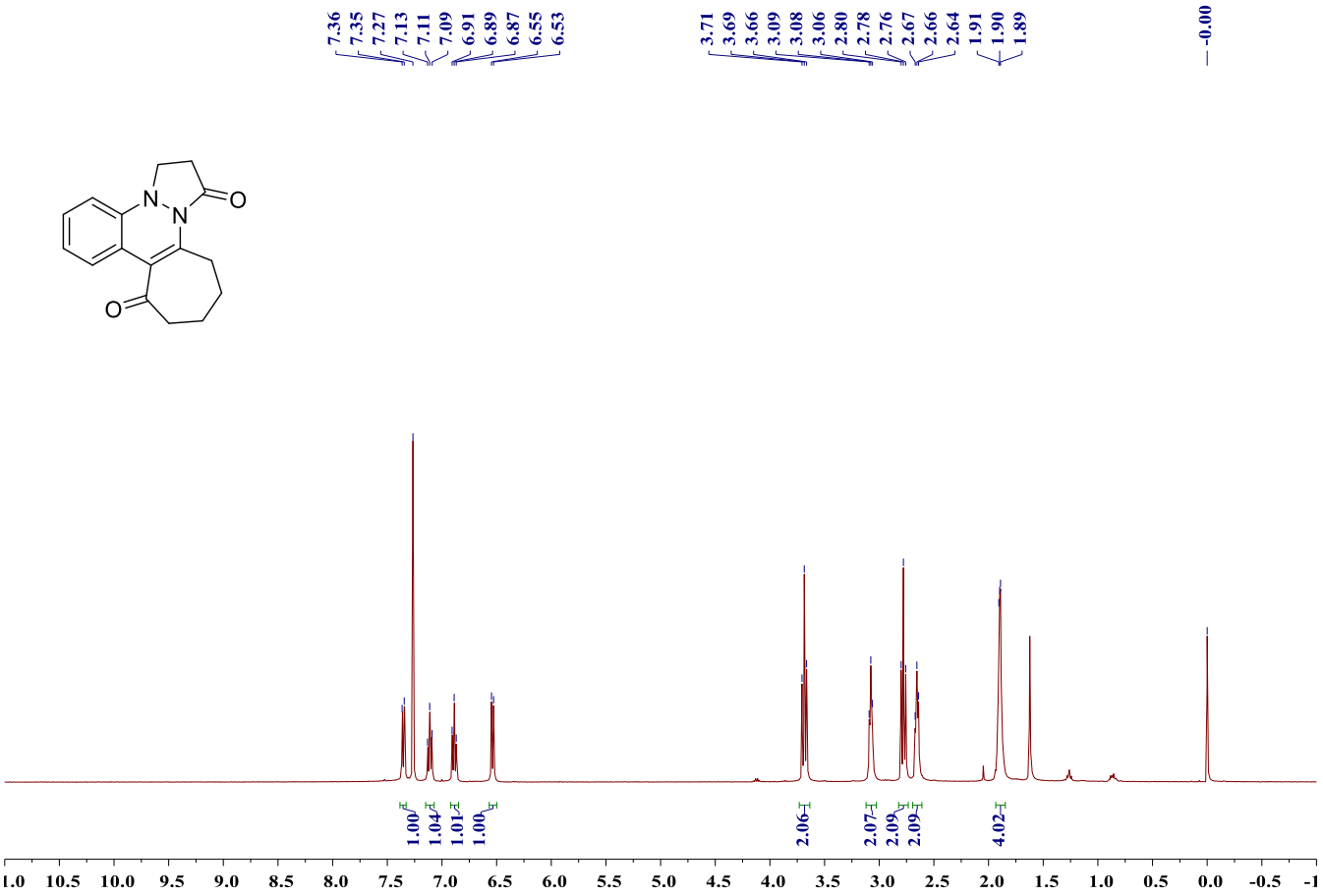


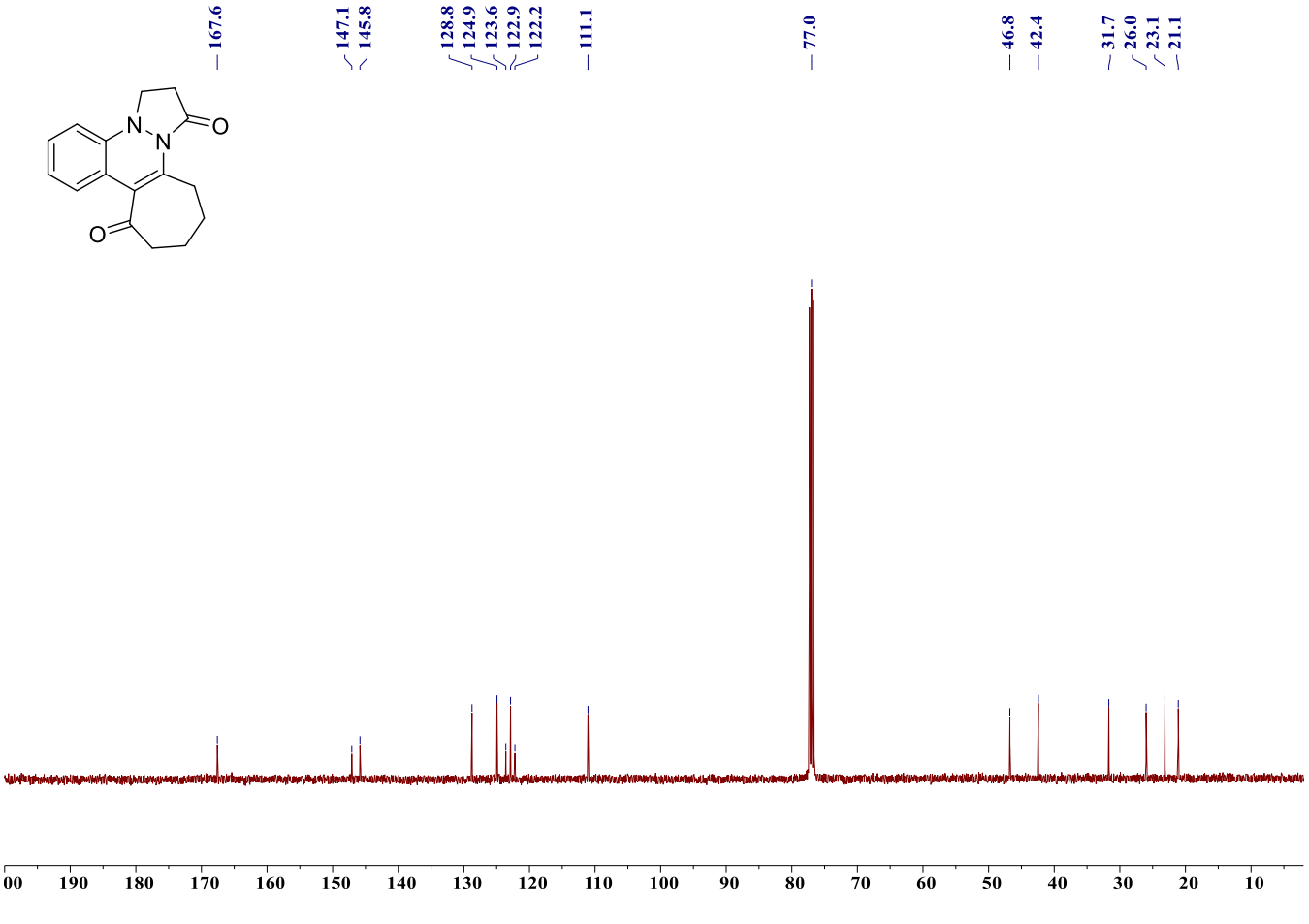



**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5o**

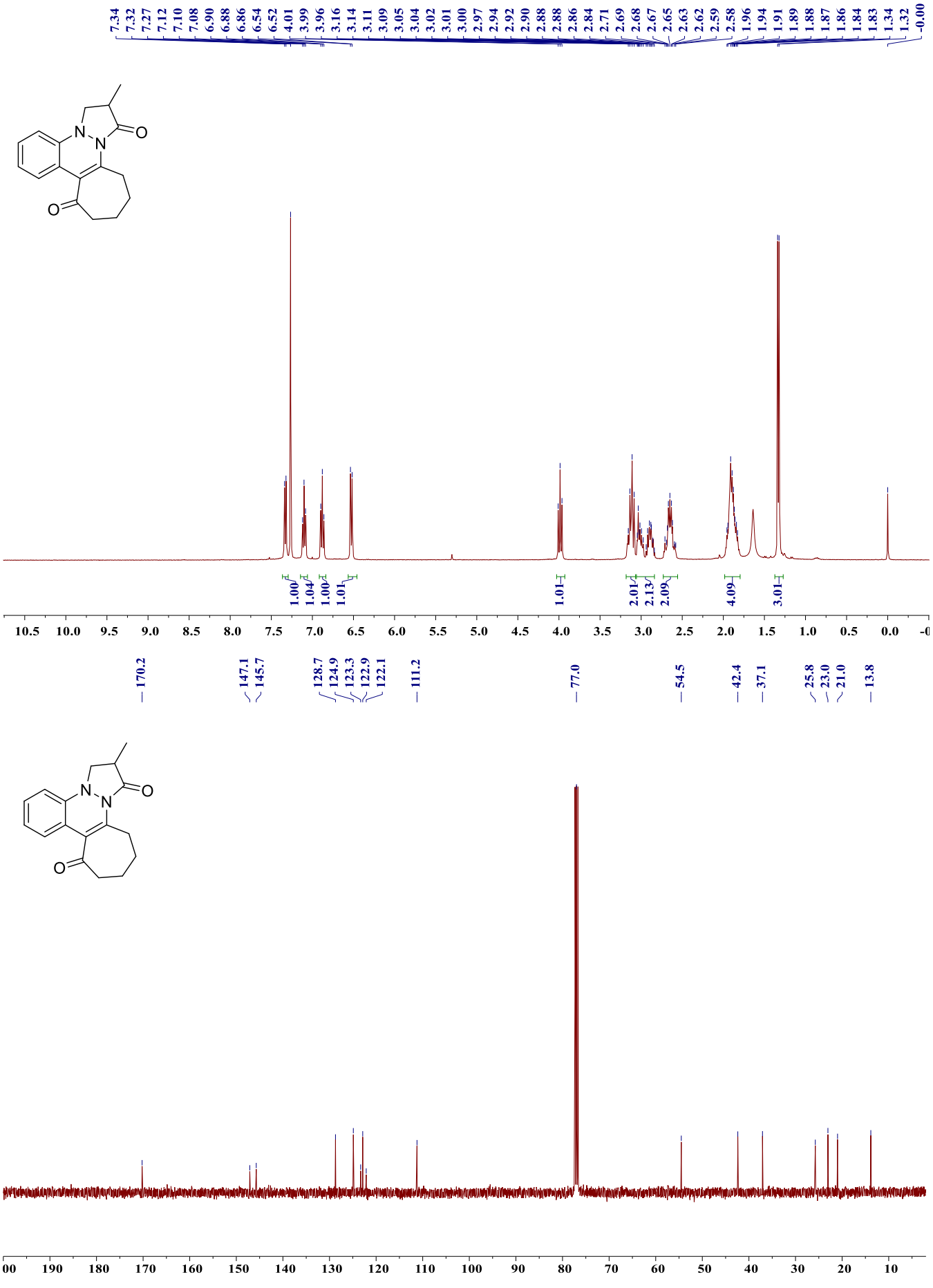


**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5p**

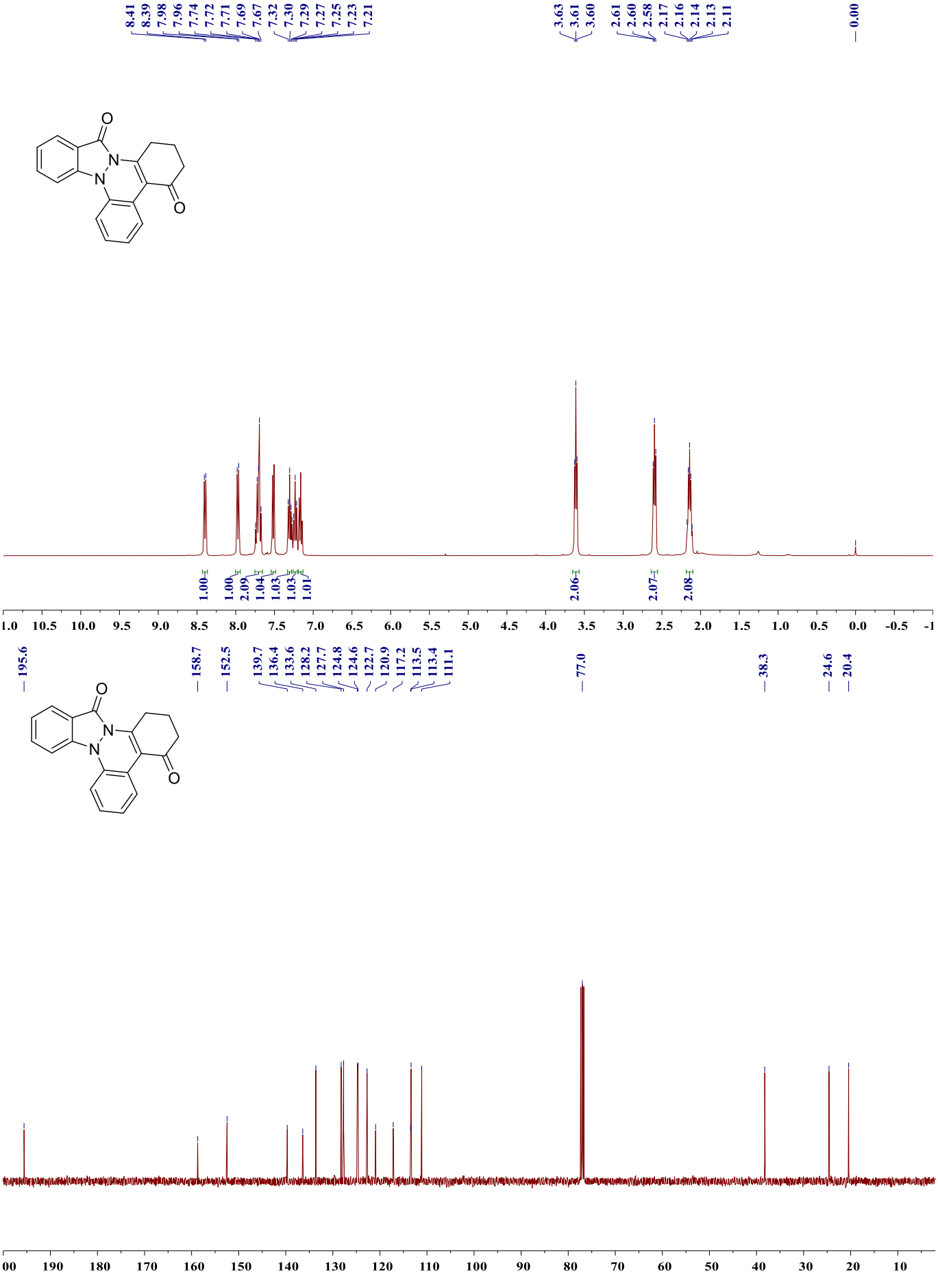


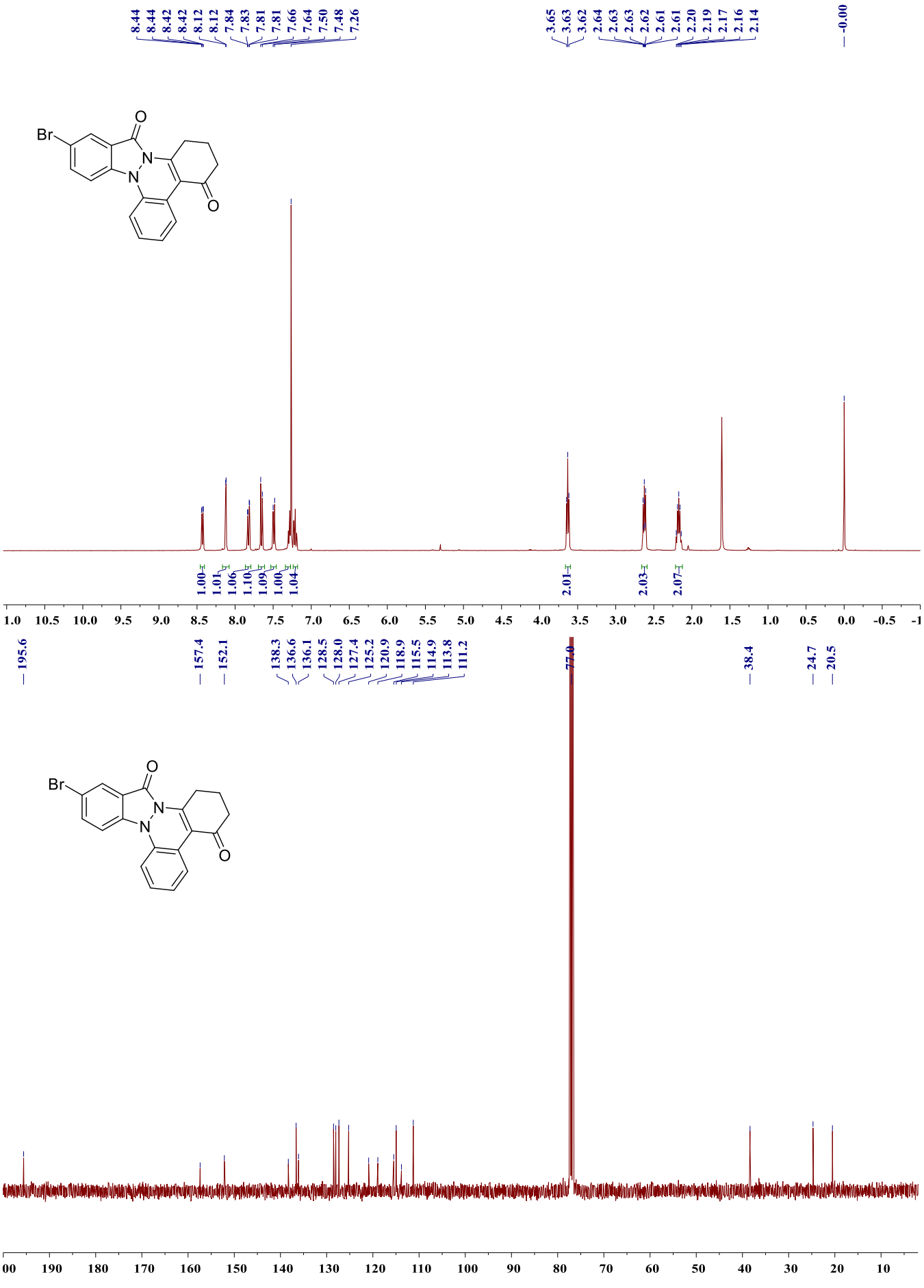
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5q**



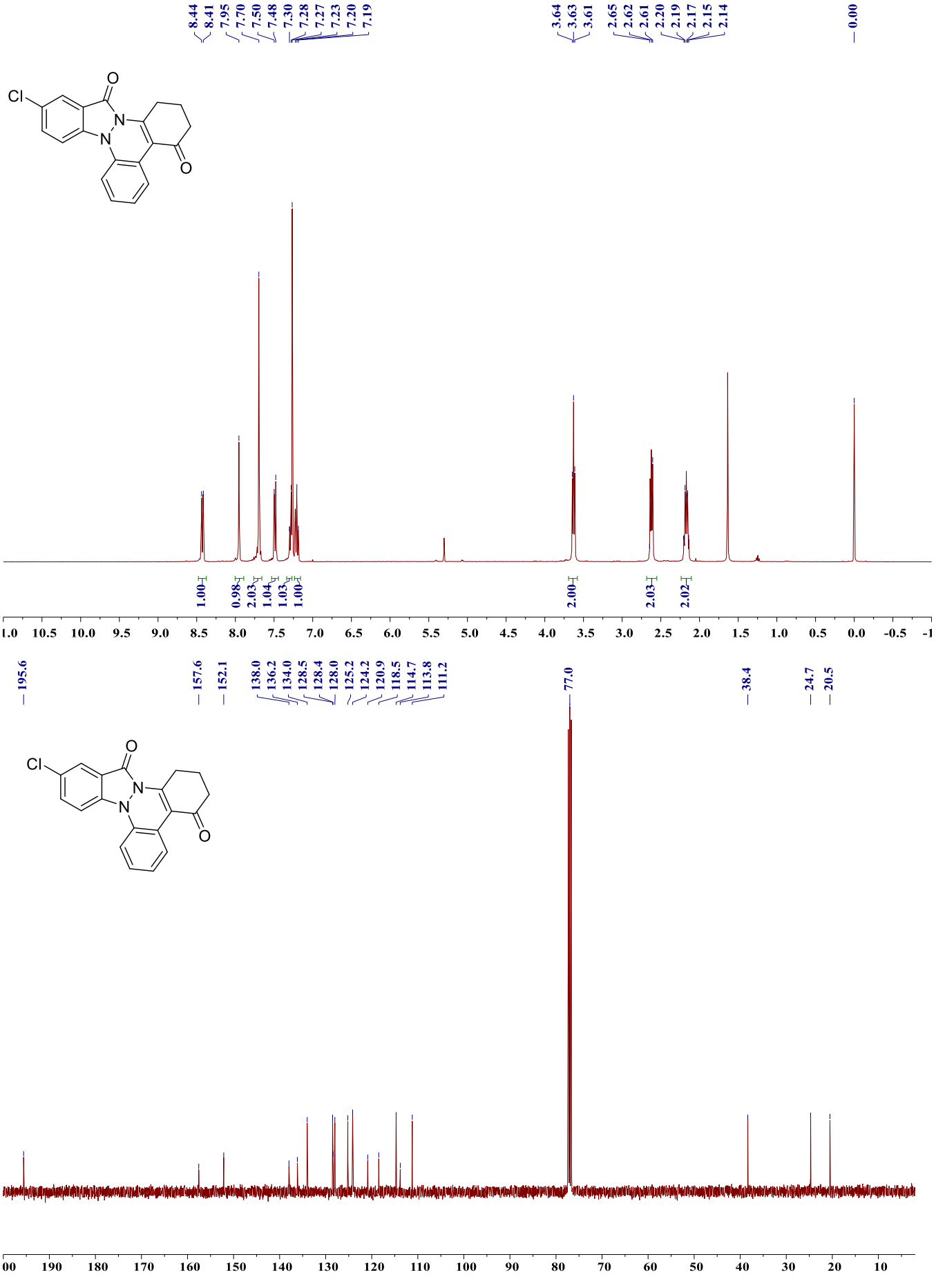
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5r**



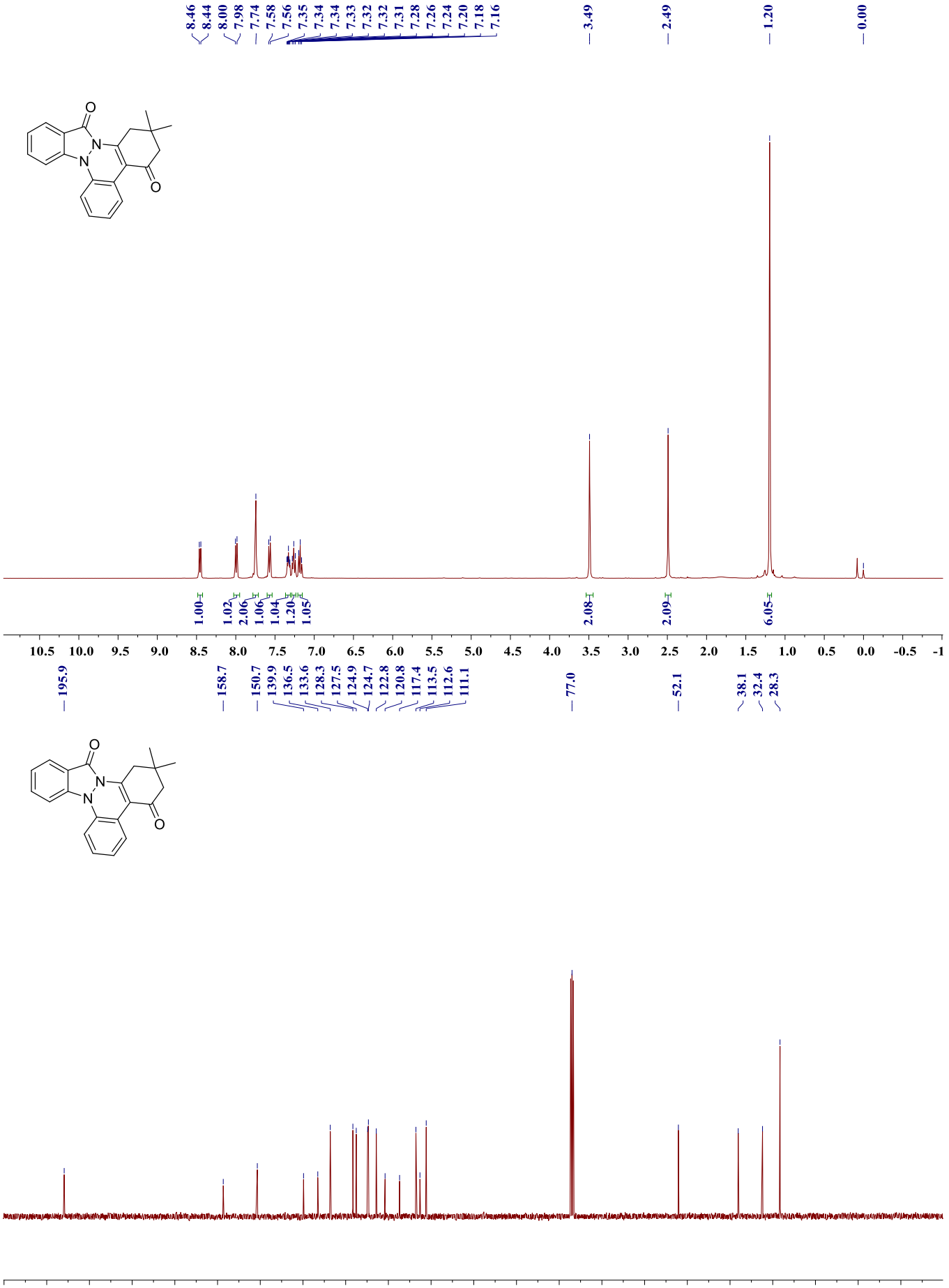
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5s**



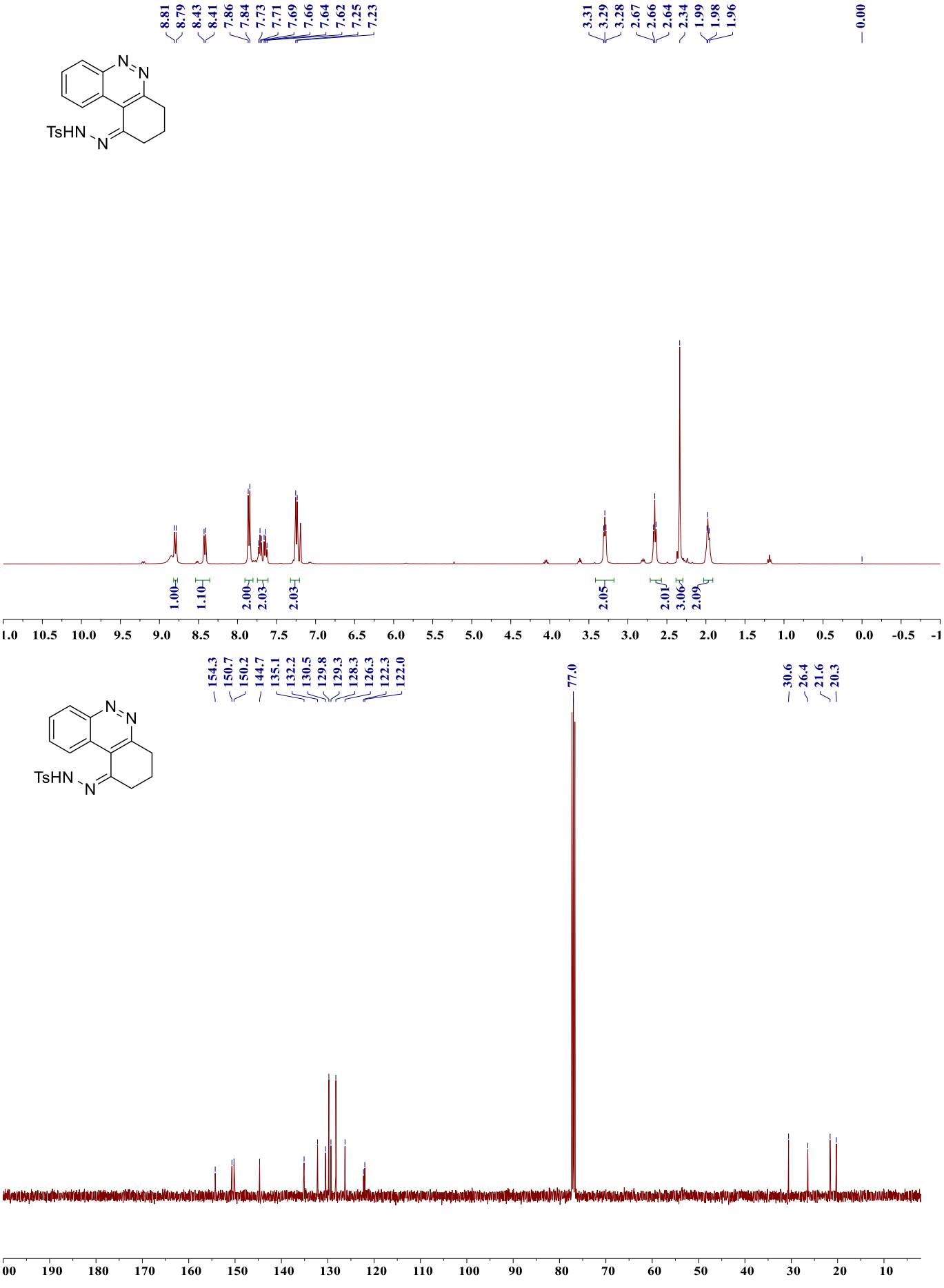
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5t**



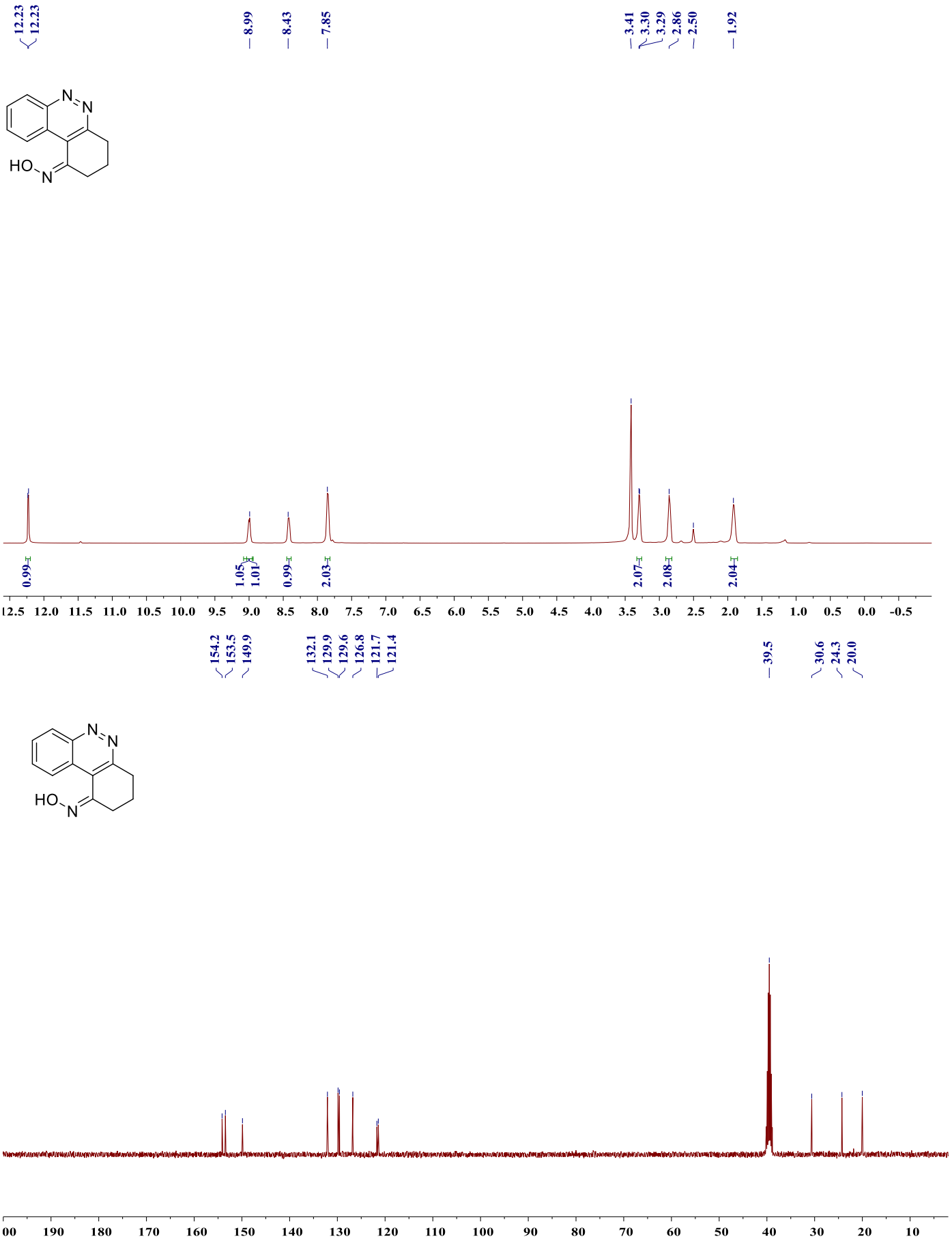
**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 5u**



**$^{1}$H NMR (400 MHz, $CDCl_3$) and $^{13}$C NMR (100 MHz, $CDCl_3$) spectra of product 6**



**$^{1}$H NMR (400 MHz, DMSO) and $^{13}$C NMR (100 MHz, DMSO) spectra of product 7**



**$^{1}$H NMR (400 MHz, DMSO) and $^{13}$C NMR (100 MHz, DMSO) spectra of product 8**