

Supporting Information

Synthesis of Spiroindolenine-cyclopentenedione Skeletons and Their Chemical Behaviors: First Example of Lactone-type spiroindolenine structure

Meltem Tan Uygun^{1,2*} and Nurettin Menges^{1,2*}

¹Pharmaceutical Chemistry Section, Van Yuzuncu Yil University, 65080, Van, Turkey

²SAFF Chemical Reagent R&D Laboratory, YYU-TEKNOKENT, 65080, Van, Turkey

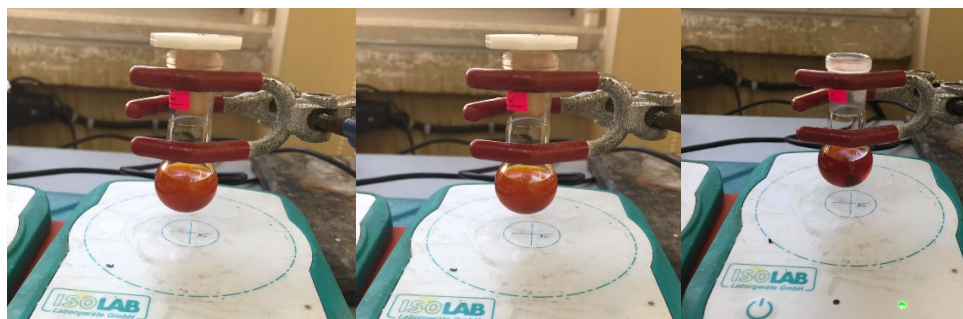
Email: meltemtan@yyu.edu.tr; nurettinmenges@yyu.edu.tr

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1. General information

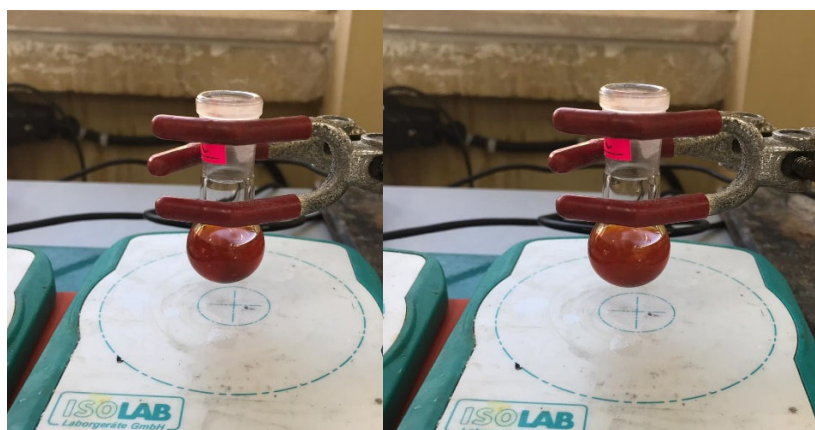
All solvents were dried and distilled using standard procedures. Unless otherwise noted, reagents were obtained from commercial sources and used without further purification. ^1H NMR and ^{13}C NMR were recorded in deuterated chloroform and dimethyl sulfoxide (CDCl_3 and $\text{d}_6\text{-DMSO}$). Coupling constants are recorded in hertz, and chemical shifts are recorded as δ values in ppm. The following abbreviations are used to describe multiplicities: s = singlet, d = doublet, dd = double doublet, t = triplet, m = multiplet. High-resolution mass spectra were carried out on a mass spectrometer with a TOF analyzer (ESI). Infrared spectra were recorded on a FT-IR spectrometer. Melting points were determined by using a local hot-stage melting point apparatus and are uncorrected.



1

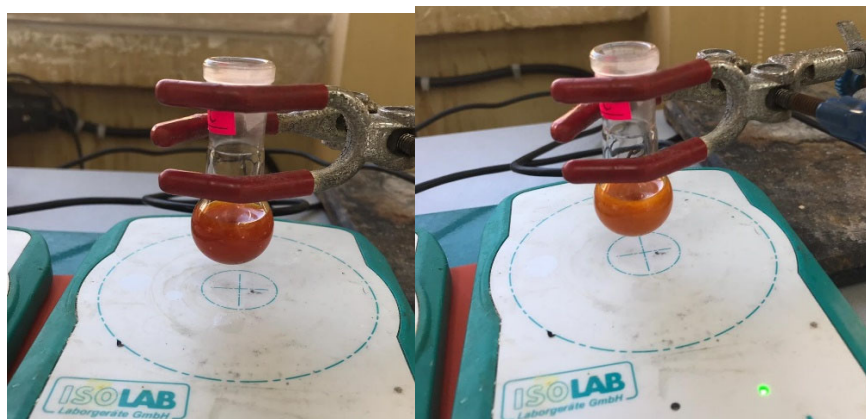
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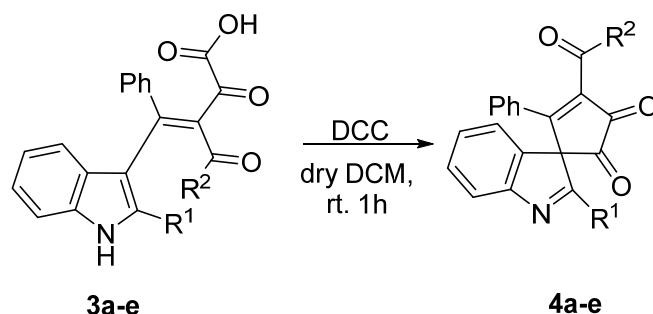
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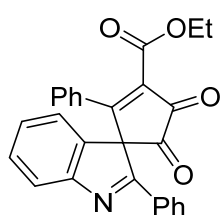
Figure S1. Visual representation of the cyclization reaction.



Scheme S1. General procedure for synthesis of **4a-e**

In a 50 mL single neck flask, the corresponding indole-3-butenoic acid derivative (1 mmol) was dissolved in anhydrous DCM (10 mL). Then DCC (1.3 mmol) was added to the mixture and the resulting solution was stirred at room temperature for 1h. After completion of the reaction, the crude reaction mixture was refrigerated for 1 h and filtered to remove DCU and then concentrated under reduced pressure. This crude residue was dissolved in dry ether (10 mL), and it was crystallized during refrigeration.

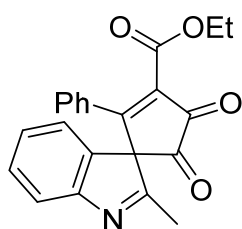
Ethyl 4,5-dioxo-2,2'-diphenylspiro[cyclopentane-1,3'-indol]-2-ene-3-carboxylate (**4a**)



Orange crystal (165-167 °C), Yield 88% (708 mg); **¹H NMR (400 MHz, CDCl₃)** δ 7.84 (dt, *J*=0.8 and 7.8 Hz, 1H, Ar-H), 7.76-7.73 (m, 2H, Ar-H), 7.54 (td, *J*=1.5 and 7.3, 1H, Ar-H), 7.40 (tt, *J*= 1.5 and 7.3, 1H, Ar-H), 7.37-7.24 (m, 5H, Ar-H), 7.19-7.14 (m, 2H, Ar-H), 6.99-6.96 (m, 2H, Ar-H), 4.39 (dq, *J*=2.2 and 7.2 Hz, 2H, OCH₂), 1.27 (t, *J*=7.2 Hz, 3H, CH₃). **¹³C NMR (100 MHz, CDCl₃)** δ 190.8, 182.4, 173.5, 171.5, 163.4, 156.0, 138.1, 137.9, 133.3, 132.0,

131.9, 131.2, 130.9, 129.3, 129.3, 128.0, 127.9, 127.5, 122.6, 121.7, 71.6, 62.6, 14.1. **HRMS** calculated for $[C_{27}H_{19}NO_4+H]^+$ 422.1387, Found 422.1393.

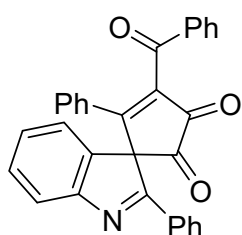
Ethyl 2'-methyl-4,5-dioxo-2-phenylspiro[cyclopentane-1,3'-indol]-2-ene-3-carboxylate (**4b**)



Green-yellow solid (152-155 °C), Yield 86% (761 mg); **¹H NMR (400 MHz, CDCl₃)** δ 7.70 (d, *J*=7.8 Hz, 1H, Ar-H), 7.49 (td, *J*=1.3 and 7.6 Hz, 1H, Ar-H), 7.44 (tt, *J*=1.2 and 7.5 Hz, 1H, Ar-H), 7.30-7.22 (m, 3H, Ar-H), 7.18 (ddd, *J*=0.6, 1.3 and 7.5 Hz, 1H, Ar-H), 7.02-7.00 (m, 2H, Ar-H), 4.39 (q, *J*=7.1 Hz, 2H, OCH₂), 2.18 (s, 3H, CH₃), 1.29 (t, *J*=7.1, 3H, CH₃).

¹³C NMR (100 MHz, CDCl₃) δ 192.1, 182.1, 175.7, 169.8, 163.4, 156.6, 139.8, 137.5, 133.5, 131.1, 130.6, 129.5, 127.9, 127.1, 122.3, 121.7, 73.5, 62.7, 17.8, 14.1. **HRMS** calculated for $[C_{22}H_{17}NO_4+H]^+$ 360.1230, Found 360.1235.

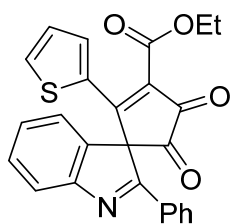
3-benzoyl-2,2'-diphenylspiro[cyclopentane-1,3'-indol]-2-ene-4,5-dione (**4c**)



Brick coloured solid (156-158 °C), Yield 82% (552 mg); **¹H NMR (400 MHz, CDCl₃)** δ 7.92-7.89 (m, 3H, Ar-H), 7.85-7.82 (m, 2H, Ar-H), 7.63-7.57 (m, 2H, Ar-H), 7.47-7.34 (m, 7H, Ar-H), 7.22 (t, *J*=1.2 and 7.5 Hz, 1H, Ar-H), 7.06-7.02 (m, 2H, Ar-H), 6.96-6.93 (m, 2H, Ar-H). **¹³C NMR (100 MHz, CDCl₃)** δ 192.1, 191.1, 183.9, 173.9, 169.8, 156.0, 144.4,

138.4, 135.3, 134.9, 133.4, 132.1, 132.0, 131.2, 130.9, 129.5, 129.5, 129.4, 129.3, 128.6, 128.0, 127.6, 122.8, 121.6, 71.9. **HRMS** calculated for $[C_{31}H_{20}NO_3 +H]^+$ = 454.1443, Found $[M+H]^+$ = 454.1439.

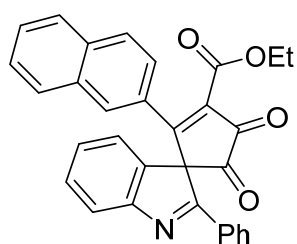
Ethyl 4,5-dioxo-2'-phenyl-2-(thiophen-2-yl)spiro[cyclopentane-1,3'-indol]-2-ene-3-carboxylate (**4d**)



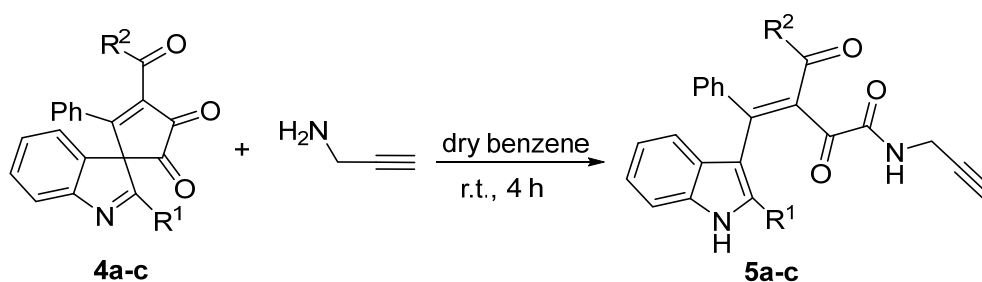
Brick coloured solid (157-159 °C), Yield 80%; **¹H NMR (400 MHz, CDCl₃)** δ 7.87 (dt, *J*=0.7, 7.8 Hz, 1H, Ar-H), 7.83-7.80 (m, 2H, Ar-H), 7.64 (dd, *J*=1.1, 5.0 Hz, 1H, Thp-H), 7.53 (ddd, *J*=2.0, 6.8, 7.8 Hz, 1H, Ar-H), 7.45-7.41 (m, 1H, Ar-H), 7.40-7.35 (m, 2H, Ar-H), 7.28-7.23 (m, 2H, Ar-H), 7.20 (dd, *J*=1.1, 4.1 Hz, 1H, Thp-H), 6.94 (dd, *J*=4.1, 5.0 Hz, 1H, Thp-H),

4.55 (q, *J*=7.12 Hz, 2H, -OCH₂), 1.46 (t, *J*=7.1 Hz, 3H, CH₃). **¹³C NMR (100 MHz, CDCl₃)** δ 190.3, 181.8, 173.9, 163.7, 159.9, 155.6, 138.1, 137.2, 135.1, 134.3, 132.9, 131.9, 131.8, 130.7, 129.4, 129.3, 127.7, 127.3, 122.5, 121.5, 71.3, 62.9, 14.1. **HRMS** Calculated for $[C_{25}H_{17}NO_4S+Na]^+$ 450.0771, Found 450.0776.

Ethyl 2-(naphthalen-2-yl)-4,5-dioxo-2'-phenylspiro[cyclopentane-1,3'-indol]-2-ene-3-carboxylate (**4e**)



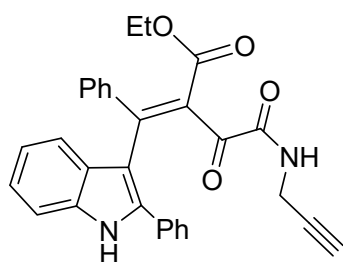
Dark yellow solid (142-144 °C), Yield 92%; ¹H NMR (400 MHz, CDCl₃) δ 7.89 (dt, *J*=0.9, 7.9 Hz, 1H, Ar-H), 7.80-7.77 (m, 2H, Ar-H), 7.70 (dd, *J*=0.6, 8.1 Hz, 1H, Ar-H), 7.63 (d, *J*=8.8 Hz, 1H, Ar-H), 7.59-7.55 (m, 2H, Ar-H), 7.54-7.49 (m, 2H, Ar-H), 7.45-7.42 (m, 1H, Ar-H), 7.39-7.34 (m, 2H, Ar-H), 7.34-7.30 (m, 3H, Ar-H), 7.04 (dd, *J*=2.0, 8.7 Hz, 1H, Ar-H), 4.47-4.39 (m, 2H, -OCH₂), 1.28 (t, *J*=7.1 Hz, 3H, CH₃). ¹³C NMR (100 MHz, CDCl₃) δ 190.6, 182.2, 173.6, 170.8, 163.6, 155.8, 138.0, 137.9, 135.1, 132.4, 131.9, 131.7, 130.8, 129.8, 129.4, 129.2, 129.1, 129.0, 128.4, 127.7, 127.6, 127.4, 127.2, 123.7, 122.5, 121.6, 71.6, 62.6, 14.1. HRMS Calculated for [C₃₁H₂₁NO₄+Na]⁺ 494.1363, Found 494.1370.



Scheme S3. General procedure for synthesis of **5a-c**

In a 50 mL single neck flask, the corresponding spiroindolenine (**4a-c**) (1 mmol) was dissolved in anhydrous benzene (10 mL). Then propargyl amine (1 mmol) was added to the mixture and the resulting solution was stirred at room temperature for 4h. After completion of the reaction, the crude reaction filtered and compounds **5a-c** obtained without any purification.

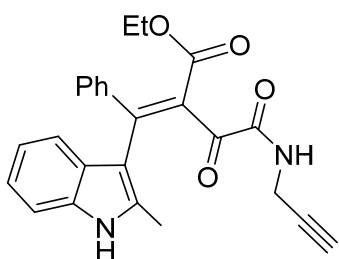
Ethyl 3,4-dioxo-2-(phenyl(2-phenyl-1*H*-indol-3-yl)methylene)-4-(prop-2-yn-1-ylamino)butanoate (**5a**)



Claret red (112-114 °C), Yield 71% (160 mg); ¹H NMR (400 MHz, CDCl₃) δ 9.17 (bs, 0.6H, NH-indole), 9.03 (bs, 0.4H, NH-indole), 7.58-7.56 (m, 1H, Ar-H), 7.49-7.47 (m, 1H, Ar-H), 7.42-7.39 (m, 1H, Ar-H), 7.34 (tt, *J*=1.3 Hz and 7.5 Hz, 1H, Ar-H), 7.30-7.15 (m, 7H, Ar-H), 7.13-7.08 (m, 1H, Ar-H), 6.99-6.92 (m,

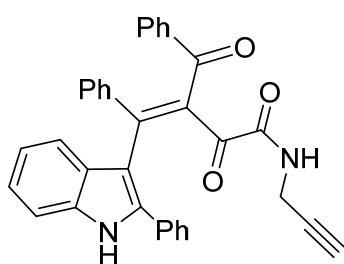
2H, Ar-H), 6.88 (t, $J=5.6$ Hz, 0.4H, NH-amide), 6.29 (t, $J=5.6$ Hz, 0.6H, NH-amide), 4.09-4.00 (m, 1.2H, OCH₂), 3.91 (ddd, $J=2.6, 5.6$ and 17.5 Hz, 0.8H, CH₂-propargyl), 3.82-3.61 (m, 0.8H, OCH₂), 3.51 (ddd, $J=2.6, 5.6$ and 17.5 Hz, 1.2H, CH₂-propargyl), 2.21 (t, $J=2.6$ Hz, 0.4H, CH), 2.19 (t, $J=2.6$ Hz, 0.6H, CH), 1.00 (t, $J=7.1$ Hz, 1.8H, CH₃), 0.81 (t, $J=7.1$ Hz, 1.2H, CH₃). ¹³C NMR (100 MHz, CDCl₃) δ 190.2, 188.3, 167.2, 166.0, 160.8, 160.5, 156.9, 155.4, 141.7, 140.4, 140.0, 139.3, 136.4, 136.2, 131.5, 130.8, 130.7, 130.3, 129.9, 129.0, 128.8, 128.7, 128.6, 128.4, 128.3, 128.3, 128.3, 128.2, 123.3, 122.8, 121.4, 121.0, 120.3, 120.0, 113.1, 113.1, 111.7, 111.6, 78.5, 78.3, 72.3, 72.1, 61.2, 61.0, 29.1, 28.8, 13.7, 13.6. HRMS calculated for [C₃₀H₂₄N₂O₄+H]⁺ 477.1809, Found 477.1812.

Ethyl 2-((2-methyl-1*H*-indol-3-yl)(phenyl)methylene)-3,4-dioxo-4-(prop-2-yn-1-ylamino)butanoate (**5b**)



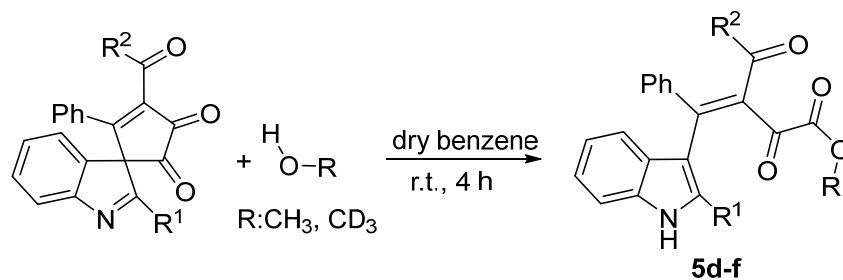
Yellow solid (182-184 °C), Yield 75% (86.6 mg); ¹H NMR (400 MHz, d₆-DMSO) δ 11.68 (bs, 0.4H, NH-indole), 11.65 (bs, 0.6H, NH-indole), 8.98 (t, $J=5.8$ Hz, 0.4H, NH-amide), 8.75 (t, $J=5.8$ Hz, 0.6H, NH-amide), 7.48-7.25 (m, 5.4 H, Ar-H), 7.12 (d, $J=7.2$ Hz, 0.6H, Ar-H), 7.04-6.98 (m, 1H, Ar-H), 6.84 (t, $J=7.3$ Hz, 0.4H, Ar-H), 6.78 (t, $J=7.4$ Hz, 0.6H, Ar-H), 6.64 (d, $J=8.0$ Hz, 0.4H, Ar-H), 6.55 (d, $J=8.0$ Hz, 0.6H, Ar-H), 3.94 (q, $J=7.1$ Hz, 2H, OCH₂), 3.82-3.80 (m, 0.8H, CH₂-propargyl), 3.46-3.38 (m, 1.2H, CH₂-propargyl), 3.11 (t, $J=2.4$ Hz, 0.4H, CH), 2.96 (t, $J=2.4$ Hz, 0.6H, CH), 2.11 (s, 2H, CH₃-indole), 2.07 (s, 1H, CH₃-indole), 0.95 (t, $J=7.1$ Hz, 1H, CH₃), 0.90 (t, $J=7.1$ Hz, 2H, CH₃). ¹³C NMR (100 MHz, d₆-DMSO) δ 189.3, 167.0, 165.9, 162.7, 162.6, 155.7, 141.5, 140.7, 140.2, 136.5, 136.0, 134.9, 130.4, 130.4, 130.3, 129.9, 128.9, 128.7, 128.0, 127.7, 127.1, 122.1, 120.4, 119.9, 119.0, 113.4, 113.2, 111.5, 80.6, 80.5, 73.5, 73.2, 60.7, 28.5, 28.2, 14.0, 13.9, 13.1, 13.0. HRMS calculated for [C₂₅H₂₂N₂O₄+H]⁺ 415.1652, Found 415.1654.

3-benzoyl-2-oxo-4-phenyl-4-(2-phenyl-1*H*-indol-3-yl)-*N*-(prop-2-yn-1-yl)but-3-enamide (**5c**)



Orange solid (204-207 °C), Yield 78% (80.6 mg); ¹H NMR (400 MHz, d₆-DMSO) δ 11.97 (bs, 0.45H, NH-indole), 11.54 (bs, 0.55H, NH-indole), 9.08 (t, $J=5.9$ Hz, 0.55H, NH-amide), 8.59 (t, $J=5.8$ Hz, 0.45H, NH-amide), 7.81-7.79 (m, 0.55H, Ar-H), 7.56-7.29 (m, 9.55H, Ar-H), 7.26-6.96 (m, 6.45H, Ar-H), 6.92-6.80 (m, 2H, Ar-H), 6.62 (d, $J=7.9$ Hz, 0.45H, Ar-H), 3.77-3.74 (m, 1H,

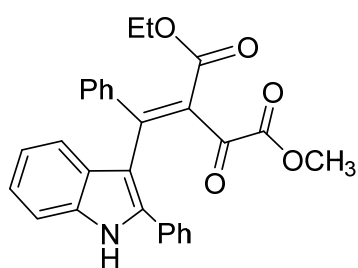
CH₂-propargyl), 3.59-3.41 (m, 1H, CH₂-propargyl), 3.10 (t, *J*=2.4 Hz, 0.55H, CH), 3.03 (t, *J*=2.4 Hz, 0.45H, CH). ¹³C NMR (100 MHz, CDCl₃) δ 195.7, 194.8, 191.3, 189.9, 160.8, 160.7, 141.6, 141.3, 139.5, 139.0, 137.5, 136.9, 136.2, 136.1, 136.1, 135.9, 132.7, 131.2, 131.0, 130.7, 130.7, 130.5, 130.2, 129.7, 129.2, 129.1, 129.1, 129.0, 129.0, 128.9, 128.9, 128.4, 128.3, 128.3, 128.2, 128.1, 127.2, 123.6, 123.2, 121.8, 121.2, 120.6, 120.3, 114.2, 113.9, 111.6, 111.0, 78.5, 78.3, 72.5, 72.2, 29.3, 29.1. HRMS calculated for [C₃₄H₂₄N₂O₃+H] 509.1860, Found 509.1867.



Scheme S4. General procedure for synthesis of **5d-f**

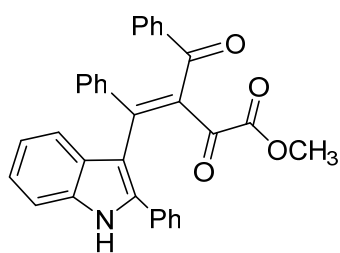
In a 50 mL single neck flask, the corresponding spiroindolenine (1 mmol) was dissolved in anhydrous benzene (10 mL). Then methanol or deuterated methanol (1 mmol) was added to the mixture and the resulting solution was stirred at room temperature for 4h. After completion of the reaction, the crude reaction filtered and compounds **5d-f** obtained without any purification.

1-Ethyl 4-methyl-3-oxo-2-(phenyl(2-phenyl-1*H*-indol-3-yl)methylene)succinate (**5d**)



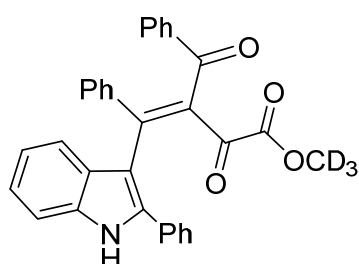
Orange solid (195-198 °C), Yield 91% (195.8 mg); ¹H NMR (400 MHz, d₆-DMSO) δ 12.19 (bs, 1H, NH), 7.55-7.50 (m, 3H, Ar-H), 7.47-7.44 (m, 4H, Ar-H), 7.42-7.35 (m, 4H, Ar-H), 7.19-7.15 (m, 1H, Ar-H), 6.95-6.91 (m, 1H, Ar-H), 6.67-6.65 (m, 1H, Ar-H), 4.06-3.91 (m, 2H, CH₂), 2.77 (s, 3H, OCH₃), 0.98 (t, *J*=7.1 Hz, 3H, CH₃). ¹³C NMR (100 MHz, d₆-DMSO) δ 183.6, 166.0, 162.5, 155.0, 144.6, 138.3, 136.8, 130.9, 129.9, 129.7, 129.4, 128.7, 128.6, 128.5, 128.4, 127.9, 123.2, 121.0, 119.6, 112.0, 110.6, 60.7, 51.5, 13.5. HRMS calculated for [C₂₃H₂₁NO₅+H] 454.1649, Found 454.1652.

Methyl 3-benzoyl-2-oxo-4-phenyl-4-(2-phenyl-1*H*-indol-3-yl)but-3-enoate (**5e**)

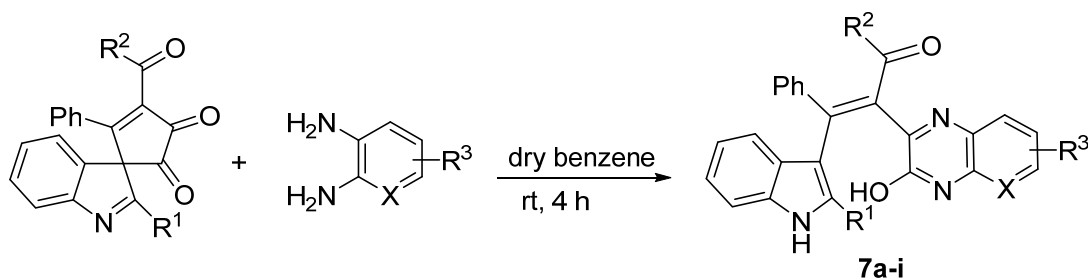


Yellow solid (268-270 °C), Yield 89% (143.5 mg); **¹H NMR (400 MHz, d₆-DMSO)** δ 12.20 (s, 1H, NH), 7.84 (d, *J* = 7.3 Hz, 1H, Ar-H), 7.64-7.57 (m, 3H, Ar-H), 7.50-7.29 (m, 12H, Ar-H), 7.22 – 7.18 (m, 1H, Ar-H), 6.99 – 6.95 (m, 1H, Ar-H), 6.75 (d, *J* = 7.6 Hz, 1H, Ar-H), 2.87 (s, 3H, OCH₃). **¹³C NMR (100 MHz, d₆-DMSO)** δ 194.2, 185.3, 162.7, 155.1, 144.7, 138.0, 136.9, 136.7, 136.2, 133.3, 130.7, 130.6, 130.0, 129.8, 128.9, 128.8, 128.7, 128.6, 128.5, 128.0, 123.2, 121.0, 119.6, 112.0, 111.6, 51.8. **HRMS** calculated for [C₃₂H₂₃NO₄+H]⁺ 486.1700, Found 486.1708.

Methyl-d₃ 3-benzoyl-2-oxo-4-phenyl-4-(2-phenyl-1*H*-indol-3-yl)but-3-enoate (**5f**)

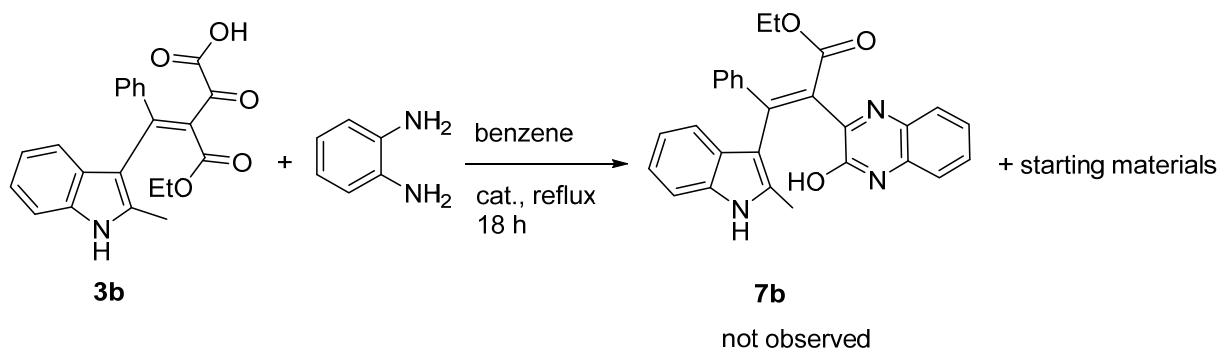


Yellow solid (260-263 °C), Yield 92% (148.9 mg); **¹H NMR (400 MHz, d₆-DMSO)** δ 12.20 (s, 0.8H, NH, %20 ND), 7.84 (d, *J* = 6.8 Hz, 1H, Ar-H), 7.64-7.56 (m, 3H, Ar-H), 7.48-7.42 (m, 6H, Ar-H), 7.38 – 7.29 (m, 6H, Ar-H), 7.22 – 7.18 (m, 1H, Ar-H), 6.98 – 6.95 (m, 1H, Ar-H), 6.76 (d, *J* = 8.1 Hz, 1H, Ar-H). **¹³C NMR (100 MHz, d₆-DMSO)** δ 194.2, 185.3, 162.7, 155.1, 144.6, 137.9, 136.8, 136.2, 133.4, 130.7, 129.9, 129.8, 128.9, 128.7, 128.7, 128.6, 128.6, 128.5, 128.0, 127.3, 123.2, 120.9, 119.7, 111.9, 111.5. **HRMS** calculated for [C₃₂H₂₀D₃NO₄+H]⁺ 489.1888, Found 489.1891.



Scheme S5. General procedure for synthesis of **7a-k**

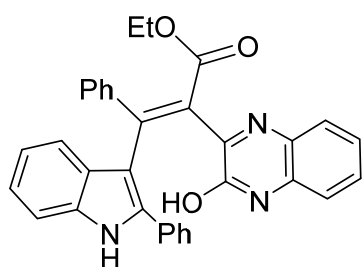
In a 50 mL single neck flask, the corresponding spiroindolenine (1 mmol) was dissolved in anhydrous benzene (10 mL). Then 1,2-diamine derivative (1 mmol) was added to the mixture and the resulting solution was stirred at room temperature for 4h. After completion of the reaction, the crude reaction filtered and compounds **7a-k** obtained without any purification.



Scheme S6. The reaction attempt of indole-3-butyric acid and 1,2-diamino benzene (cat: AcOH or TFA)

In a 50 mL single neck flask, compound 3b (1 mmol) was dissolved in benzene (10 mL). Then 1,2-diaminobenzene (1 mmol) was added to the solution and the resulting solution was stirred at room temperature for 18h. After 18h, TLC analysis was utilized, and the reaction media was analyzed with $^1\text{H-NMR}$. Same reaction was also run using catalytic amount of organic acid such as AcOH and TFA, but the result was the same.

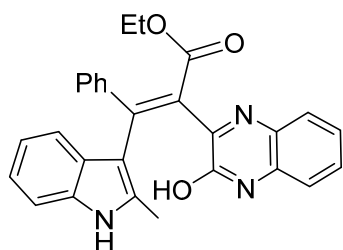
Ethyl 2-(3-hydroxyquinoxalin-2-yl)-3-phenyl-3-(2-phenyl-1*H*-indol-3-yl)acrylate (**7a**)



Yellow solid (202-205 °C), Yield 82% (99.2 mg); $^1\text{H NMR}$ (400 MHz, $\text{d}_6\text{-DMSO}$) δ 12.10 (bs, 1H, OH), 11.48 (s, 1H, NH-indole), 7.50-7.47 (m, 2H, Ar-H), 7.40-7.36 (m, 1H, Ar-H), 7.28-7.16 (m, 10H, Ar-H), 7.11-7.06 (m, 3H, Ar-H), 7.01-6.97 (m, 1H, Ar-H), 6.84-6.80 (m, 1H, Ar-H), 3.94 (q, $J=7\text{Hz}$, 2H, OCH_2), 0.90 (t, 3H, CH_3). $^{13}\text{C NMR}$ (100 MHz, $\text{d}_6\text{-DMSO}$) δ 167.4,

157.6, 153.6, 147.8, 141.2, 138.2, 135.9, 131.7, 131.7, 131.6, 130.0, 129.02, 128.6, 128.4, 128.3, 128.2, 128.0, 127.8, 127.6, 127.6, 122.9, 121.6, 119.5, 118.9, 114.9, 113.0, 111.4, 60.0, 13.6. **HRMS** calculated for $\text{C}_{33}\text{H}_{25}\text{N}_3\text{O}_3+\text{H}^+$ 512.1969, Found 512.1973.

Ethyl 2-(3-hydroxyquinoxalin-2-yl)-3-(2-methyl-1*H*-indol-3-yl)-3-phenylacrylate (**7b**)

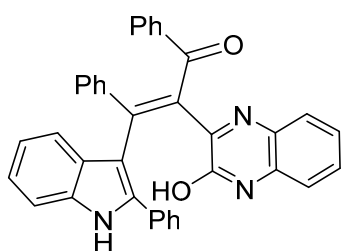


Orange solid (160-162 °C), Yield 85% (212.5 mg); $^1\text{H NMR}$ (400 MHz, CDCl_3) δ 11.94 (bs, 1H, OH), 8.31 (bs, 0.5H, NH-indole), 8.11 (bs, 0.5H, NH-indole), 7.59 (d, $J=8.3\text{ Hz}$, .05H, Ar-H), 7.52 (d, $J=7.7\text{ Hz}$, .05H, Ar-H), 7.42-7.30 (m, 3H, Ar-H), 7.25-7.18 (m, 2.5 H, Ar-H), 7.14-7.04 (m, 3.5H, Ar-H), 6.99-6.94 (m, 1.5H, Ar-H), 6.89 (t, $J=7.3\text{ Hz}$, 0.5H, Ar-H), 6.78-6.71 (m, 1H, Ar-H), 4.12-3.96 (m, 2H, OCH_2), 2.15

(H), 6.89 (t, $J=7.3\text{ Hz}$, 0.5H, Ar-H), 6.78-6.71 (m, 1H, Ar-H), 4.12-3.96 (m, 2H, OCH_2), 2.15

(s, 1.5 H, -CH₃), 1.99 (s, 1.5 H, -CH₃), 0.97 (t, *J*=7.1 Hz, 1.5 H, -CH₃), 0.88 (t, *J*=7.1 Hz, 1.5 H, -CH₃). **¹³C NMR (100 MHz, d₆-DMSO)** δ 167.6, 167.0, 159.2, 158.4, 154.5, 154.1, 149.6, 148.7, 141.8, 141.4, 136.9, 136.4, 135.5, 135.4, 131.8, 131.8, 131.7, 131.6, 130.1, 130.0, 129.7, 129.1, 128.6, 128.4, 128.4, 128.3, 128.0, 127.9, 127.8, 127.6, 126.9, 126.3, 123.2, 123.1, 120.7, 120.5, 119.2, 119.0, 118.7, 118.6, 115.2, 115.0, 113.1, 112.8, 110.8, 110.6, 59.9, 59.8, 13.7, 13.5, 12.5, 12.4. **HRMS** calculated for [C₂₈H₂₃N₃O₃+H]⁺ 450.1812, Found 450.1816.

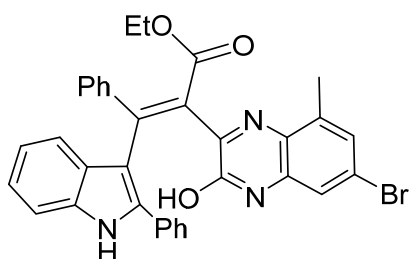
2-(3-hydroxyquinoxalin-2-yl)-1,3-diphenyl-3-(2-phenyl-1*H*-indol-3-yl)prop-2-en-1-one (**7c**)



Orange solid (280-283 °C), Yield 80% (144 mg); **¹H NMR (400 MHz, d₆-DMSO)** δ 12.34 (bs, 0.5H, OH-indole), 12.09 (bs, 0.5H, OH-quinoxaline), 11.54 (bs, 0.5H, NH-indole), 11.35 (bs, 0.5H, NH-quinoxaline), 7.86-7.84 (m, 1H, Ar-H), 7.73-7.71 (m, 1H, Ar-H), 7.54-7.45 (m, 2H, Ar-H), 7.39-6.95 (m, 16.5H, Ar-H), 6.90-

6.85 (m, 1.5H, Ar-H), 6.82-6.75 (m, 1H, Ar-H). **¹³C NMR (100 MHz, d₆-DMSO)** δ 196.3, 195.2, 160.5, 159.4, 154.3, 153.5, 148.1, 146.9, 140.7, 140.0, 138.6, 138.4, 138.2, 136.2, 136.1, 135.5, 135.1, 132.2, 131.9, 131.8, 131.7, 131.6, 131.1, 130.3, 130.2, 130.1, 129.7, 129.0, 128.8, 128.6, 128.6, 128.5, 128.4, 128.3, 128.1, 128.0, 127.9, 127.8, 127.7, 127.6, 127.2, 126.7, 123.3, 122.9, 122.0, 121.7, 119.7, 119.7, 119.6, 119.0, 115.3, 114.9, 113.5, 113.2, 111.5, 111.1. **HRMS** calculated for [C₃₇H₂₅N₃O₂+H] 544.2020, Found 544.2000.

Ethyl 2-(6-bromo-3-hydroxy-8-methylquinoxalin-2-yl)-3-phenyl-3-(2-phenyl-1*H*-indol-3-yl)acrylate (**7d**)

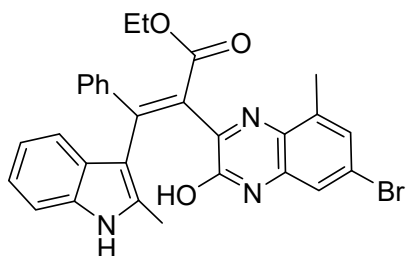


Orange solid (213-216 °C), tautomer ratio: OH (1:1), NH (1:1), Yield 72% (154.8 mg); **¹H NMR (400 MHz, d₆-DMSO)** δ 11.89 (bs, 0.5H, OH-indole), 11.76 (bs, 0.5H, OH-quinoxaline), 11.63 (bs, 0.5H, NH-indole), 11.53 (bs, 0.5H, NH-quinoxaline), 7.80-7.78 (m, 1H, Ar-H), 7.54 (bs, 1H- Ar-

H), 7.44-7.42 (m, 2H, Ar-H), 7.36-7.30 (m, 1H, Ar-H), 7.30-7.17 (m, 5H, Ar-H), 7.13-7.05 (m, 3.5H, Ar-H), 7.02-6.98 (m, 1.5H, Ar-H), 6.93-6.89 (m, 0.5H, Ar-H), 6.84-6.80 (m, 0.5H, Ar-H), 3.94 (qd, *J*=1.1, 7.0 Hz, 1H, OCH₂), 3.66-3.47 (m, 1H, OCH₂), 2.39 (s, 1.5H, CH₃-quinoxaline), 2.29 (s, 1.5H, CH₃-quinoxaline), 0.91 (t, *J*= 7.1 Hz, 1.5H, CH₃), 0.61 (t, *J*= 7.1 Hz, 1.5H, CH₃). **¹³C NMR (100 MHz, d₆-DMSO)** δ 167.3, 166.3, 159.8, 158.5, 154.6, 153.8, 149.5, 148.4, 141.0, 140.8, 138.4, 137.7, 136.3, 136.1, 133.5, 133.2, 132.8, 132.6, 131.6, 131.5, 129.7, 129.6, 129.1, 128.6, 128.6, 128.4, 128.3, 128.3, 128.1, 128.0, 127.9, 127.9, 127.8, 127.7, 126.7, 126.3, 121.9, 121.7, 119.7, 119.7, 119.1, 119.1, 114.4, 113.8, 113.0, 112.8, 111.6, 111.5,

60.1, 59.7, 16.5, 16.4, 13.6, 13.4. **HRMS** calculated for $[C_{34}H_{26}BrN_3O_3+H]^+$ = 604.1231, Found 604.1234

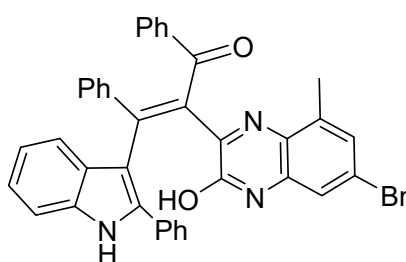
Ethyl 2-(6-bromo-3-hydroxy-8-methylquinoxalin-2-yl)-3-(2-methyl-1*H*-indol-3-yl)-3-phenylacrylate (**7e**)



Mustard coloured solid (254-258 °C), Yield 77% (174 mg); **¹H NMR (400 MHz, d₆-DMSO)** δ 11.91 (bs, 0.45H, OH-indole), 11.82 (bs, 0.55H, OH-quinoxaline), 11.45 (bs, 0.55H, NH-indole), 11.18 (bs, 0.45H, NH-quinoxaline), 7.51-7.35 (m, 3.55H, Ar-H), 7.30 (d, *J*=8.0Hz, 0.55H, Ar-H), 7.23-7.14 (m, 3H, Ar-H), 7.04-6.98 (m, 1.55H, Ar-H), 6.90-6.87 (m, 0.55H, Ar-H), 6.82 (t, *J*=7.4 Hz, 0.55H, Ar-H), 6.74-6.68 (m, 1.45H, Ar-H), 3.95-3.88 (m, 2H, OCH₂), 2.40 (s, 1.65H, CH₃-quinoxaline), 2.33 (s, 1.35H, CH₃-quinoxaline), 2.15 (s, 1.65H, CH₃-indole), 1.90 (s, 1.35H, CH₃-indole), 0.90-0.85 (m, 3H, CH₃). **¹³C NMR (100 MHz, d₆-DMSO)** δ 167.5, 166.8, 160.1, 159.3, 154.9, 154.3, 150.2, 149.4, 141.6, 141.3, 137.2, 136.6, 135.5, 133.3, 133.3, 132.8, 132.7, 129.7, 129.7, 129.6, 129.1, 128.8, 128.5, 128.1, 128.1, 128.0, 127.8, 127.5, 126.7, 126.5, 126.4,

125.6, 120.7, 120.6, 119.3, 119.2, 118.7, 118.6, 114.3, 114.1, 113.1, 112.9, 110.8, 110.7, 60.0, 59.9, 16.5, 16.4, 13.7, 13.6, 12.6. **HRMS** calculated for $[C_{29}H_{24}BrN_3O_3+H]^+$ 542.1074, Found 542.1078.

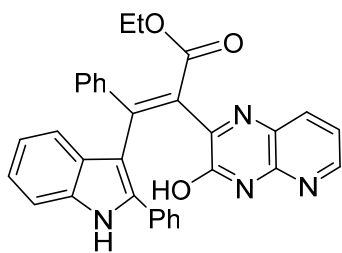
2-(6-bromo-3-hydroxy-8-methylquinoxalin-2-yl)-1,3-diphenyl-3-(2-phenyl-1*H*-indol-3-yl)prop-2-en-1-one (**7f**)



Yellow solid (235-238 °C), Yield 75% (105.4 mg); **¹H NMR (400 MHz, d₆-DMSO)** δ 11.59 (bs, 1H, OH), 11.58 (bs, 1H, NH), 7.84-7.83 (m, 2H, Ar-H), 7.47-7.45 (m, 2H, Ar-H), 7.42 (bs, 1H, Ar-H), 7.40-7.36 (m, 1H, Ar-H), 7.34-7.29 (m, 3H, Ar-H), 7.17-7.15 (m, 3H, Ar-H), 7.12-7.02 (m, 8H, Ar-H), 6.85 (t, *J*=7.5Hz, 1H, Ar-H), 2.28 (s, 3H, CH₃). **¹³C NMR (100 MHz, d₆-DMSO)** δ 195.9, 160.0, 153.5, 147.4, 140.6, 138.8, 138.2, 136.2, 134.4, 133.1, 133.0, 132.7, 132.0, 131.6, 129.9, 129.5, 128.9, 128.7, 128.4, 128.4, 128.1, 128.0, 127.9, 127.7, 126.3, 121.8, 119.8, 119.2, 113.7, 113.3, 111.6, 16.4. **HRMS** calculated for $[C_{38}H_{26}BrN_3O_2+H]^+$ 636.1281, Found 636.1293.

Ethyl 2-(3-hydroxypyrido[2,3-b]pyrazin-2-yl)-3-phenyl-3-(2-phenyl-1*H*-indol-3-yl)acrylate

(7g)

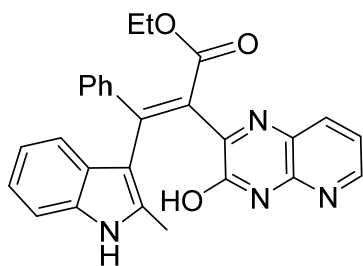


Orange solid (198-200 °C), Yield 70% (150.6 mg); ¹H NMR (400 MHz, CDCl₃) δ 8.57 (bs, 0.33H, NH-indole), 8.49 (dd, *J*=1.7 and 4.8 Hz, 0.33H, Py-H), 8.40 (bs, 0.66H, NH-indole), 8.38 (dd, *J*=1.7 and 4.8 Hz, 0.66H, Py-H), 7.95 (dd, *J*=1.7 and 8.0 Hz, 0.33H, Py-H), 7.72-7.70 (m, 0.66H, Ar-H), 7.61 (dd, *J*=1.7 and 8.0 Hz,

0.66H, Py-H), 7.47-7.44 (m, 1.33H, Ar-H), 7.39-7.37 (m, 0.33H, Ar-H), 7.35-7.26 (m, 4H, Ar-H), 7.21-7.16 (m, 2.31H, Ar-H), 7.12-6.97 (m, 5.61H, Ar-H), 6.89-6.85 (m, 0.66H, Ar-H), 4.07-4.00 (m, 1.33H, OCH₂), 3.86-3.68 (m, 0.66H, OCH₂), 0.96 (t, *J*=7.1Hz, 2H, CH₃), 0.71 (t, *J*=7.1Hz, 1H, CH₃). ¹³C NMR (100 MHz, CDCl₃) δ 168.5, 167.4, 160.4, 158.8, 155.5, 151.1, 149.4, 143.6, 143.3, 141.3, 141.0, 138.1, 137.5, 137.2, 136.2, 136.0, 131.9, 131.8, 130.4, 129.6, 129.4, 129.1, 129.1, 129.0, 128.7, 128.7, 128.6, 128.3, 128.3, 128.2, 128.1, 127.9, 127.8, 122.8, 122.7, 120.9, 120.8, 120.4, 119.9, 119.5, 119.4, 114.9, 113.9, 111.0, 61.00, 60.8, 13.8, 13.7. HRMS calculated for [C₃₂H₂₄N₄O₃+H]⁺ 513.1921, Found 513.1928.

Ethyl 2-(3-hydroxypyrido[2,3-b]pyrazin-2-yl)-3-(2-methyl-1*H*-indol-3-yl)-3-phenylacrylate

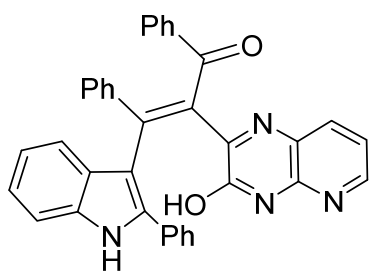
(7h)



Yellow solid (294-295 °C), Yield 74% (114.4 mg); ¹H NMR (400 MHz, d₆-DMSO) δ 12.93 (bs, 0.45H, OH-indole), 12.82 (bs, 0.55H, OH-quinoxaline), 11.47 (bs, 0.55H, NH-indole), 11.21 (bs, 0.45H, NH-quinoxaline), 8.46 (dd, *J*=1.7 and 4.7 Hz, 0.55H, Py-H), 8.41 (dd, *J*=1.7 and 4.7 Hz, 0.45H, Py-H), 7.91 (dd, *J*=1.7 and 8.0 Hz, 0.55H, Py-H), 7.87 (dd, *J*=1.7 and 8.0

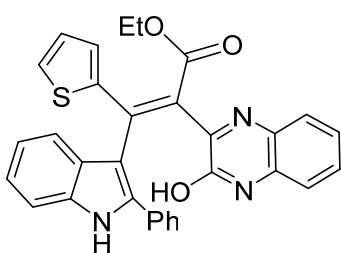
Hz, 0.45H, Py-H), 7.42-7.35 (m, 1.35H, Ar-H), 7.31-7.25 (m, 1H, Ar-H), 7.24-7.21 (m, 1.35H, Ar-H), 7.19-7.13 (m, 1.90H, Ar-H), 7.04-6.98 (m, 1.65H, Ar-H), 6.89-6.80 (m, 1H, Ar-H), 6.74-6.67 (m, 1.55H, Ar-H), 3.95-3.87 (m, 2H, OCH₂), 2.16 (s, 1.65H, CH₃-indole), 1.88 (s, 1.35H, CH₃-indole), 0.86 (t, *J*=7.1Hz, 1.35H, CH₃), 0.85 (t, *J*=7.1Hz, 1.65H, CH₃). ¹³C NMR (100 MHz, d₆-DMSO) δ 167.5, 166.7, 160.4, 159.6, 155.9, 155.3, 150.5, 150.1, 150.0, 149.6, 143.5, 143.3, 141.6, 141.2, 137.3, 136.9, 136.3, 136.2, 135.5, 135.4, 129.8, 129.1, 128.8, 128.6, 128.1, 128.1, 127.8, 127.4, 126.9, 126.8, 126.2, 125.4, 120.8, 120.7, 119.9, 119.8, 119.3, 119.2, 118.7, 118.6, 113.0, 112.8, 110.8, 110.8, 60.1, 60.0, 13.7, 13.5, 12.6. HRMS calculated for [C₂₇H₂₂N₄O₃+H]⁺ 451.1765, Found 451.1769.

2-(3-hydroxypyrido[2,3-b]pyrazin-2-yl)-1,3-diphenyl-3-(2-phenyl-1*H*-indol-3-yl)prop-2-en-1-one (7i)



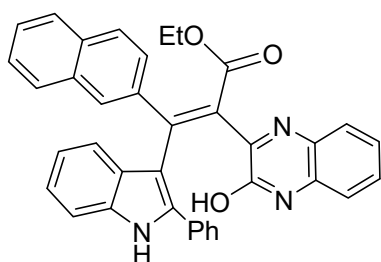
Orange solid (233-236 °C), Yield 72% (77.9 mg); **¹H NMR (400 MHz, d₆-DMSO)** δ 12.54 (bs, 1H, OH), 11.60 (bs, 1H, NH), 8.36 (dd, *J*=1.7 and 4.7 Hz, 1H, Py-H), 7.85-7.82 (m, 2H, Ar-H), 7.54 (dd, *J*=1.6 and 8.0 Hz, 1H, Py-H), 7.44-7.42 (m, 2H, Ar-H), 7.41-7.35 (m, 2H, Ar-H), 7.34-7.30 (m, 3H, Ar-H), 7.16-7.13 (m, 5H, Ar-H), 7.09-7.00 (m, 5H, Ar-H), 6.86-6.82 (m, 1H, Ar-H). **¹³C NMR (100 MHz, d₆-DMSO)** δ 196.6, 195.4, 164.2, 163.0, 154.1, 153.2, 149.3, 147.8, 145.5, 145.1, 143.0, 142.8, 140.8, 140.7, 140.5, 139.2, 138.4, 138.3, 136.4, 136.3, 135.1, 134.5, 132.2, 131.7, 131.2, 130.7, 130.3, 130.0, 129.3, 129.1, 128.7, 128.6, 128.6, 128.5, 128.2, 127.9, 127.8, 127.6, 127.5, 127.0, 125.4, 125.1, 124.5, 124.0, 122.3, 122.0, 120.1, 120.0, 119.9, 119.2, 113.4, 111.9, 111.4, 109.8. **HRMS** calculated for [C₃₆H₂₄N₄O₂+Na]⁺ 567.1792, Found 567.1794.

Ethyl 2-(3-hydroxyquinoxalin-2-yl)-3-(2-phenyl-1*H*-indol-3-yl)-3-(thiophen-2-yl)acrylate (7j)



Yellow solid (207-209 °C), Yield 94%; **¹H NMR (400 MHz, d₆-DMSO)** δ 12.44 (s, 1H, -NH), 11.78 (s, 1H, -OH), 7.80-7.78 (m, 2H, Ar-H), 7.75 (d, *J*=8.0 Hz, 1H, Ar-H), 7.57-7.53 (m, 1H, Ar-H), 7.46 (d, *J*=8.1 Hz, 1H, Ar-H), 7.41 (dd, *J*=1.1, 4.9 Hz, 1H, Ar-H), 7.36-7.28 (m, 5H, Ar-H), 7.25-7.21 (m, 1H, Ar-H), 7.16-7.12 (m, 1H, Ar-H), 7.01-6.97 (m, 1H, Ar-H), 6.76-6.71 (m, 2H, Ar-H), 3.65-3.55 (m, 2H, -OCH₂), 0.55 (t, *J*=7.1 Hz, 3H, -CH₃). **¹³C NMR (100 MHz, d₆-DMSO)** δ 166.8, 158.9, 154.4, 143.8, 141.0, 137.4, 136.4, 132.6, 132.5, 132.1, 131.3, 131.0, 130.4, 129.3, 129.2, 128.8, 128.8, 128.0, 127.5, 126.9, 123.8, 122.4, 120.1, 119.4, 115.8, 113.3, 111.9, 60.1, 13.7. **HRMS** Calculated for [C₃₁H₂₄N₃O₃S+H]⁺ 518.1538, Found 518.1522.

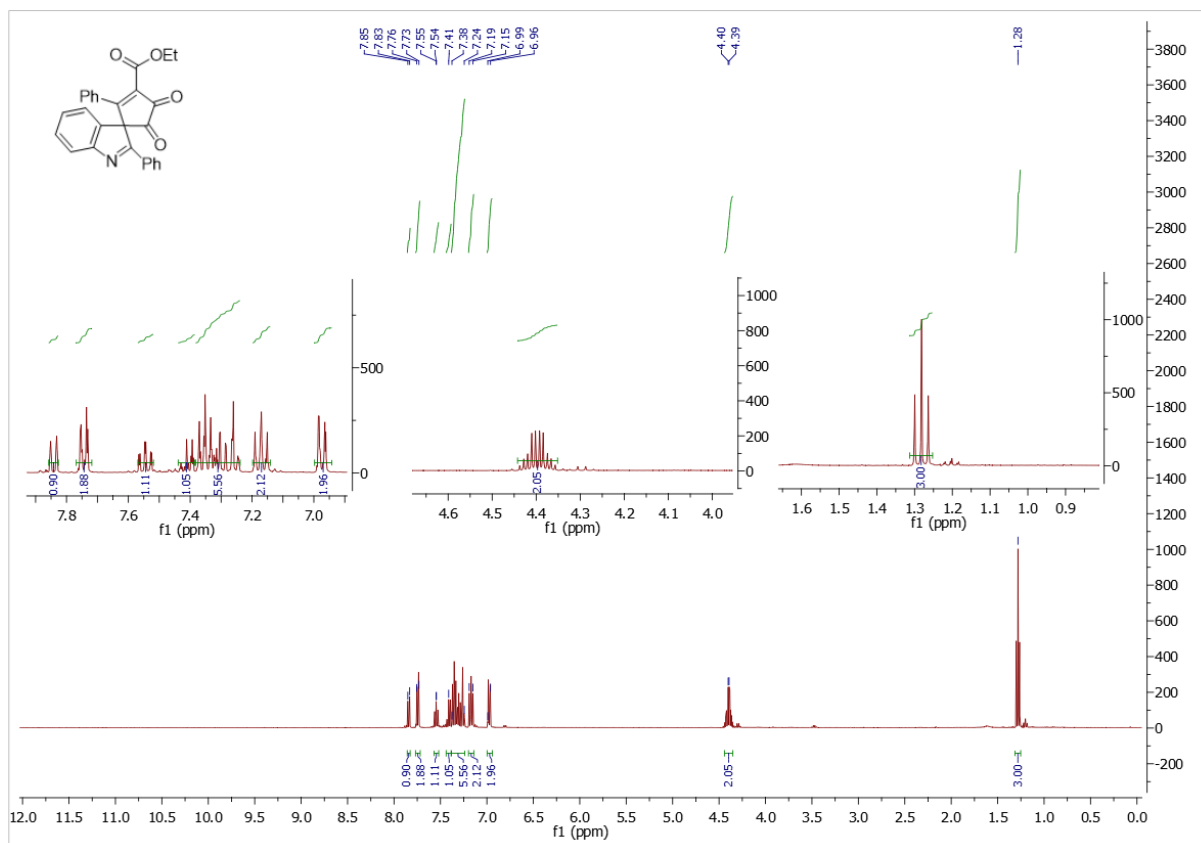
Ethyl 2-(3-hydroxyquinoxalin-2-yl)-3-(naphthalen-2-yl)-3-(2-phenyl-1*H*-indol-3-yl)acrylate (7k)



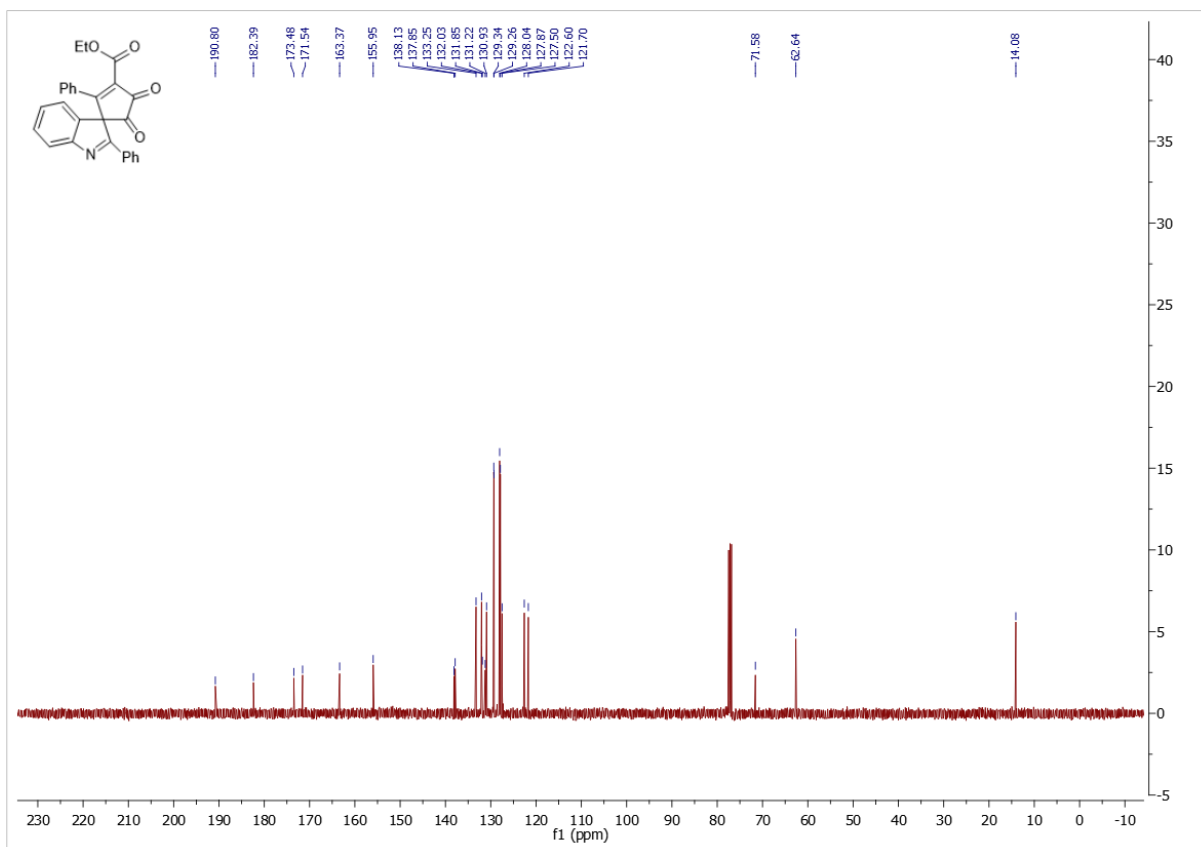
Mustard yellow solid (230-232 °C), Yield 96%; **¹H NMR (400 MHz, d₆-DMSO)** δ 12.38 (bs, 0.4H, OH-indole), 12.17 (bs, 0.6H, OH-quinoxaline), 11.81 (bs, 0.4H, NH-indole), 11.57 (bs, 0.6H, NH-quinoxaline), 7.84-7.81 (m, 1.8H, Ar-H), 7.75-7.69 (m, 1.6H, Ar-H), 7.65-7.57 (m, 1.4H, Ar-H), 7.52-7.45 (m, 3.2H, Ar-H), 7.43-7.36 (m, 1.5H, Ar-H), 7.34-7.28 (m, 2.2H, Ar-H), 7.24-7.05 (m, 6.2H, Ar-

H), 7.01-6.97 (m, 0.8H, Ar-H), 6.92-6.88 (m, 0.6H, Ar-H), 6.82-6.78 (m, 0.7H, Ar-H), 3.95-3.89 (m, 1H, -OCH₂), 3.68-3.53 (m, 1H, -OCH₂), 0.80 (t, $J=7.1$ Hz, 1.8H, -CH₃), 0.61 (t, $J=7.1$ Hz, 1.2H, -CH₃). **¹³C NMR (100 MHz, d₆-DMSO)** δ 167.8, 167.0, 159.2, 158.1, 154.6, 154.1, 149.1, 148.3, 139.2, 139.0, 138.8, 138.0, 136.7, 136.4, 133.2, 133.0, 132.8, 132.7, 132.3, 132.2, 132.2, 132.1, 132.0, 131.9, 130.7, 130.5, 129.9, 129.5, 129.2, 129.0, 128.7, 128.7, 128.6, 128.5, 128.5, 128.4, 128.3, 128.1, 127.9, 127.8, 127.7, 127.6, 127.5, 127.4, 127.0, 126.9, 126.6, 123.7, 123.4, 122.3, 122.1, 120.1, 120.0, 119.5, 119.3, 115.7, 115.4, 113.7, 113.4, 112.0, 111.9, 60.4, 60.1, 14.0, 13.8. **HRMS** Calculated for [C₃₇H₂₈N₃O₃+H]⁺ 562.2131, Found 562.2111.

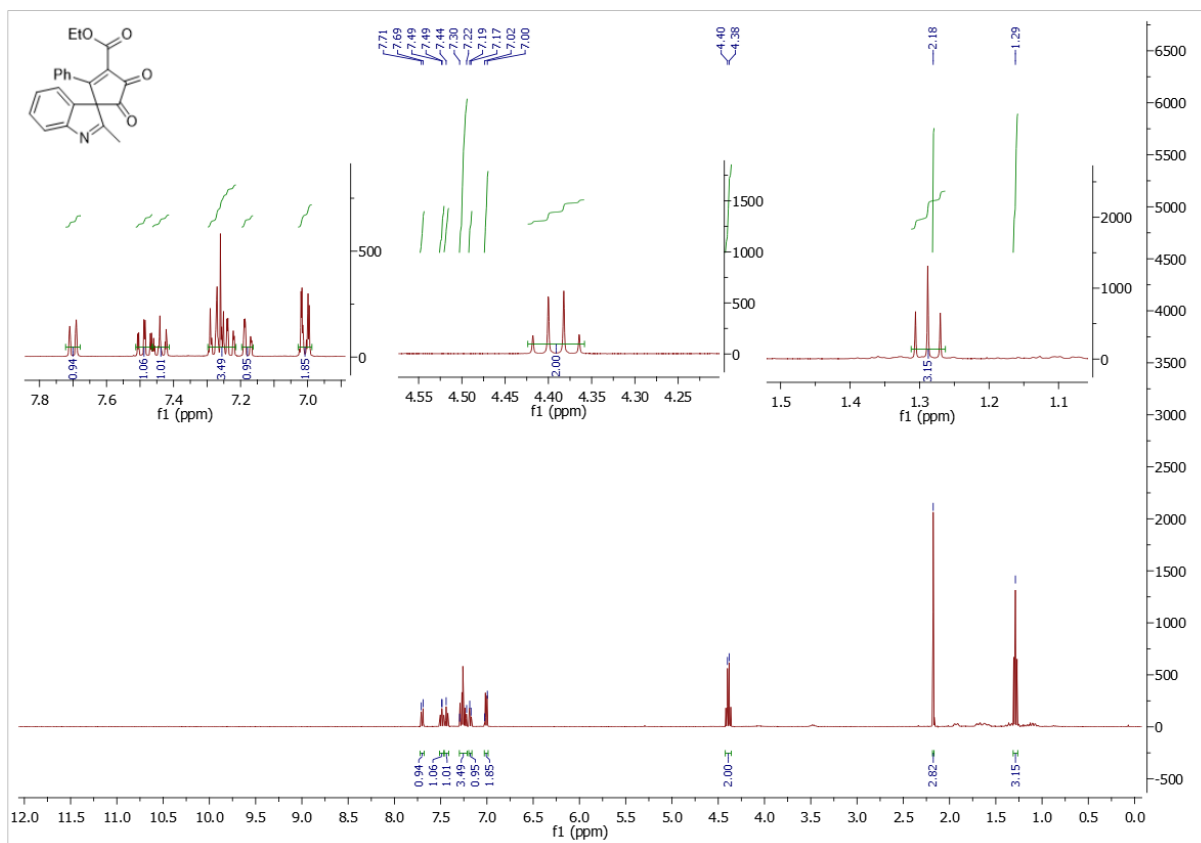
Copies of ^1H NMR and ^{13}C NMR spectra



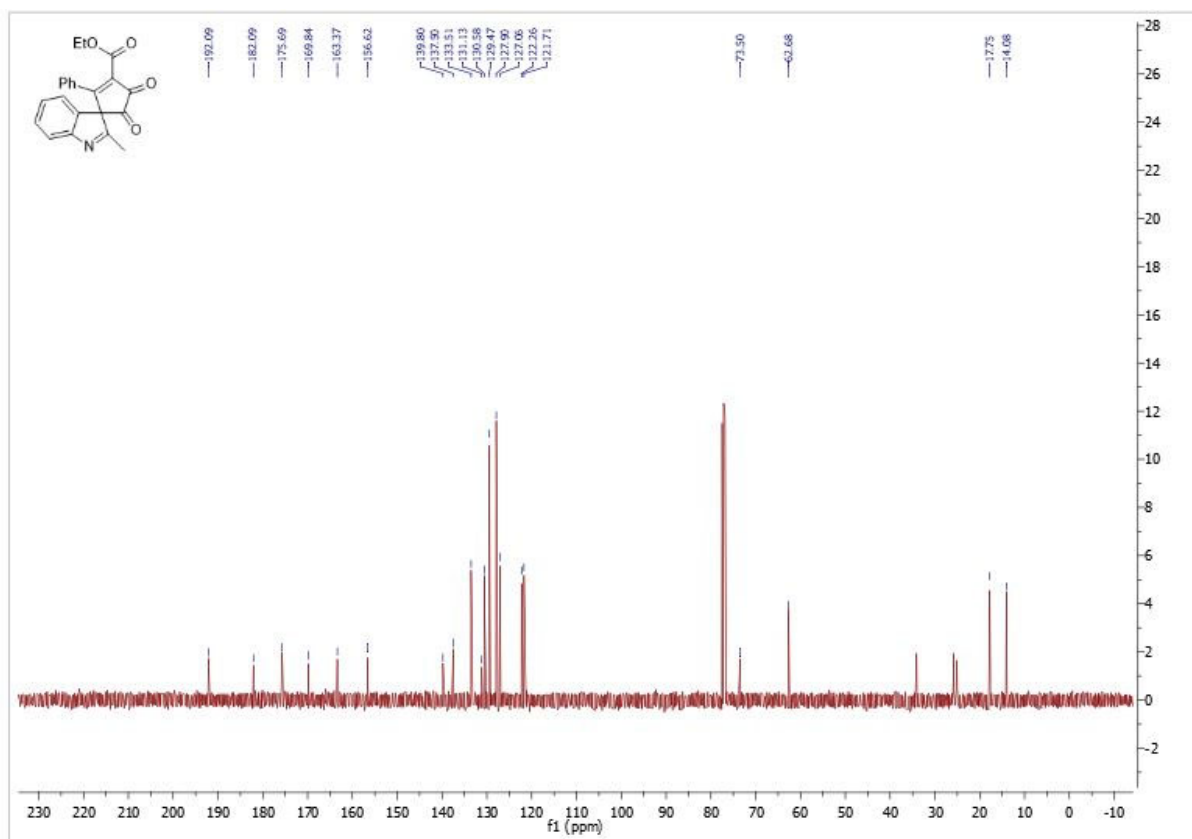
^1H -NMR spectrum of compound **4a**



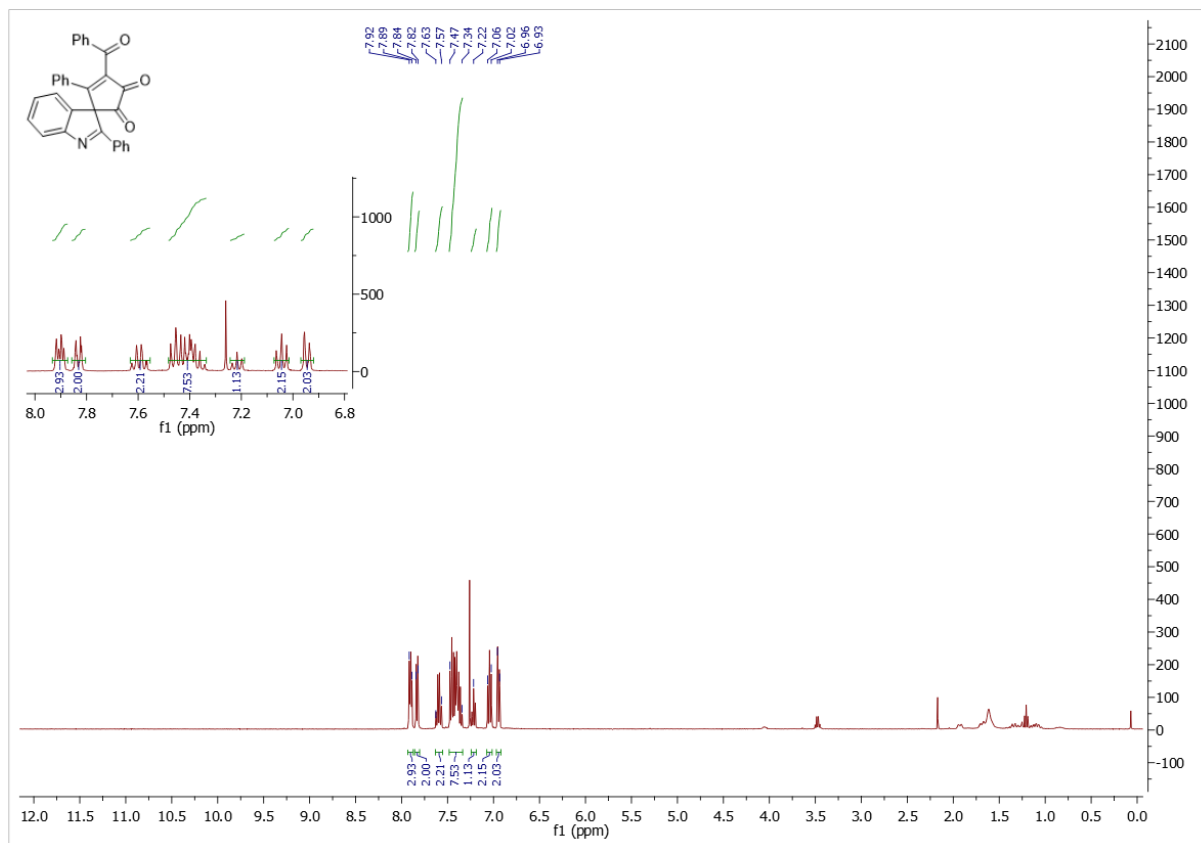
^{13}C -NMR spectrum of compound **4a**



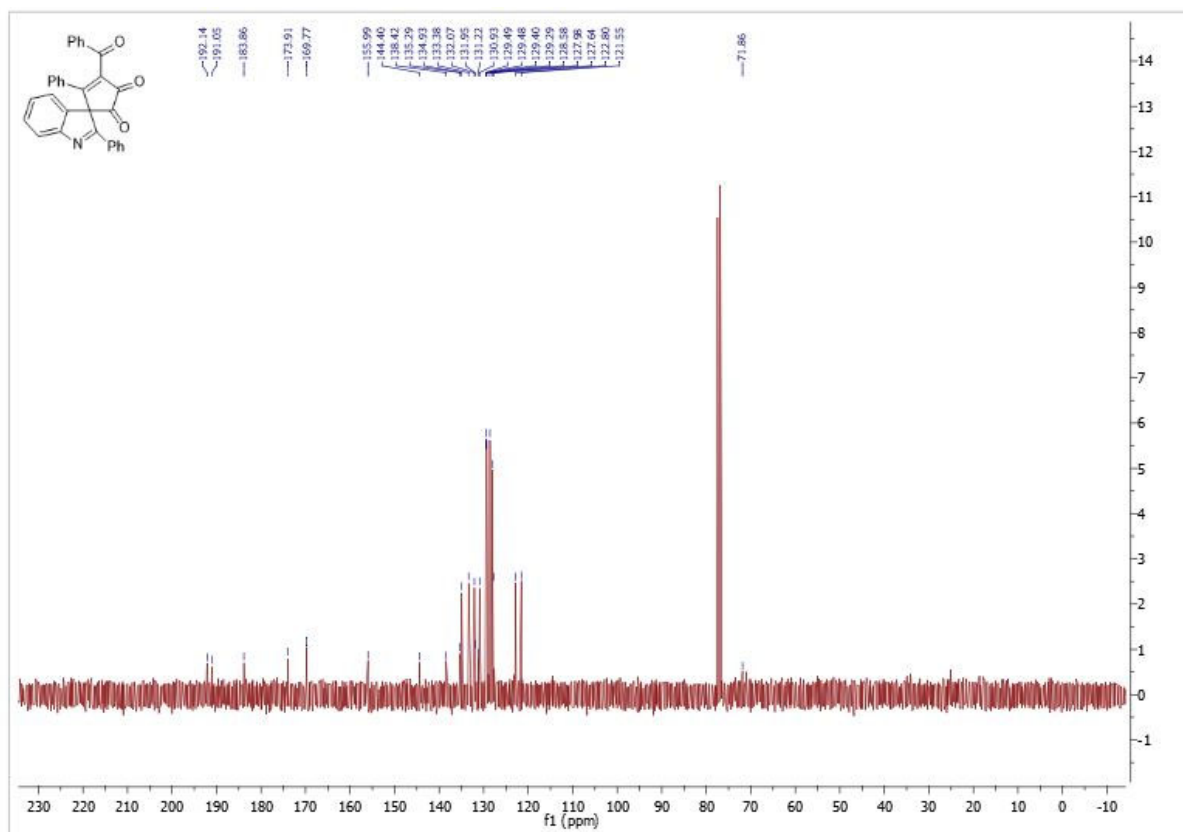
¹H-NMR spectrum of compound 4b



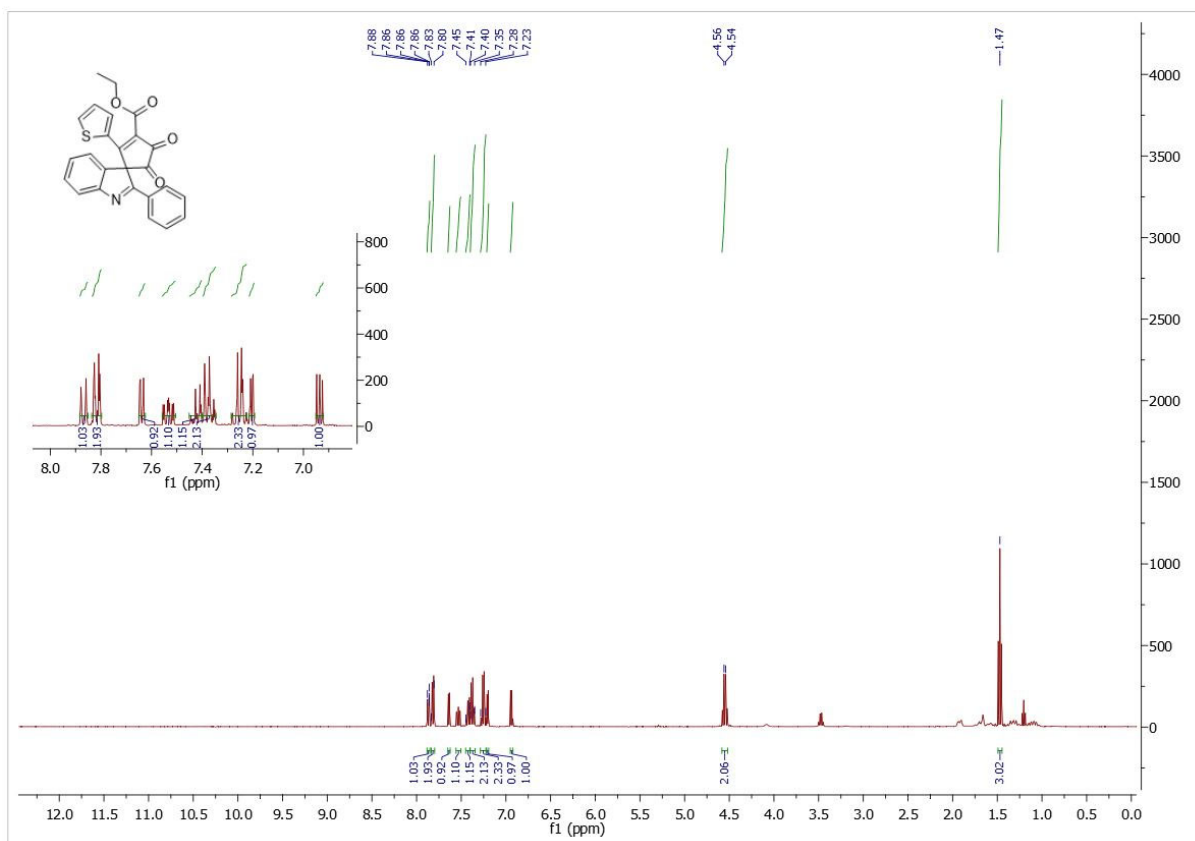
¹³C-NMR spectrum of compound 4b



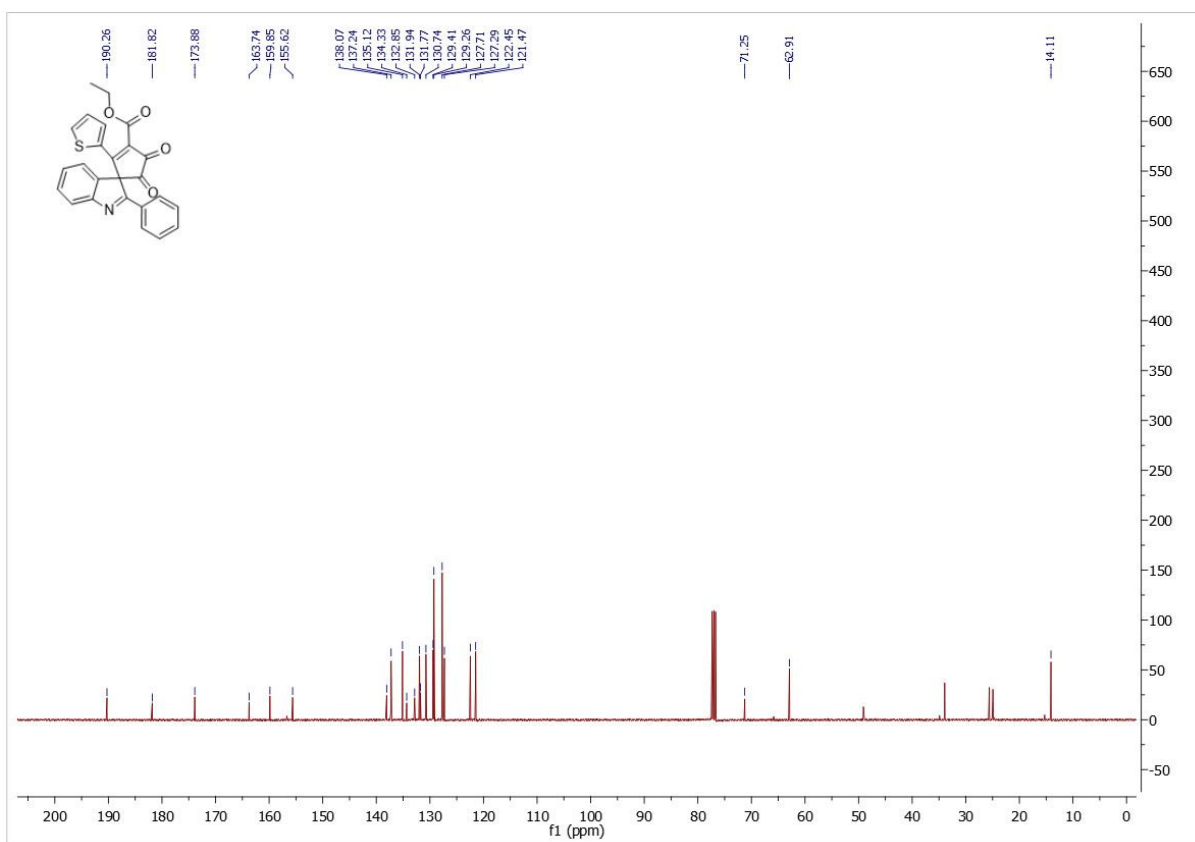
¹H-NMR spectrum of compound 4c



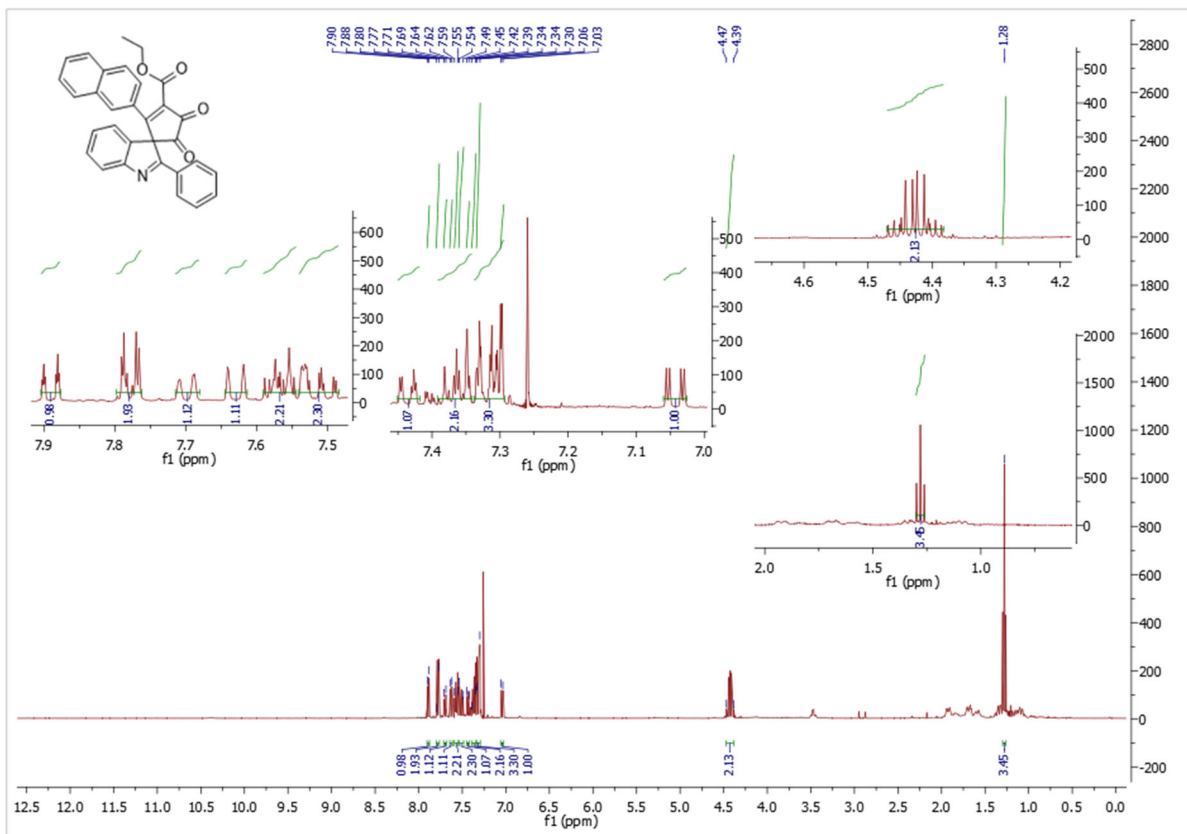
¹³C-NMR spectrum of compound 4c



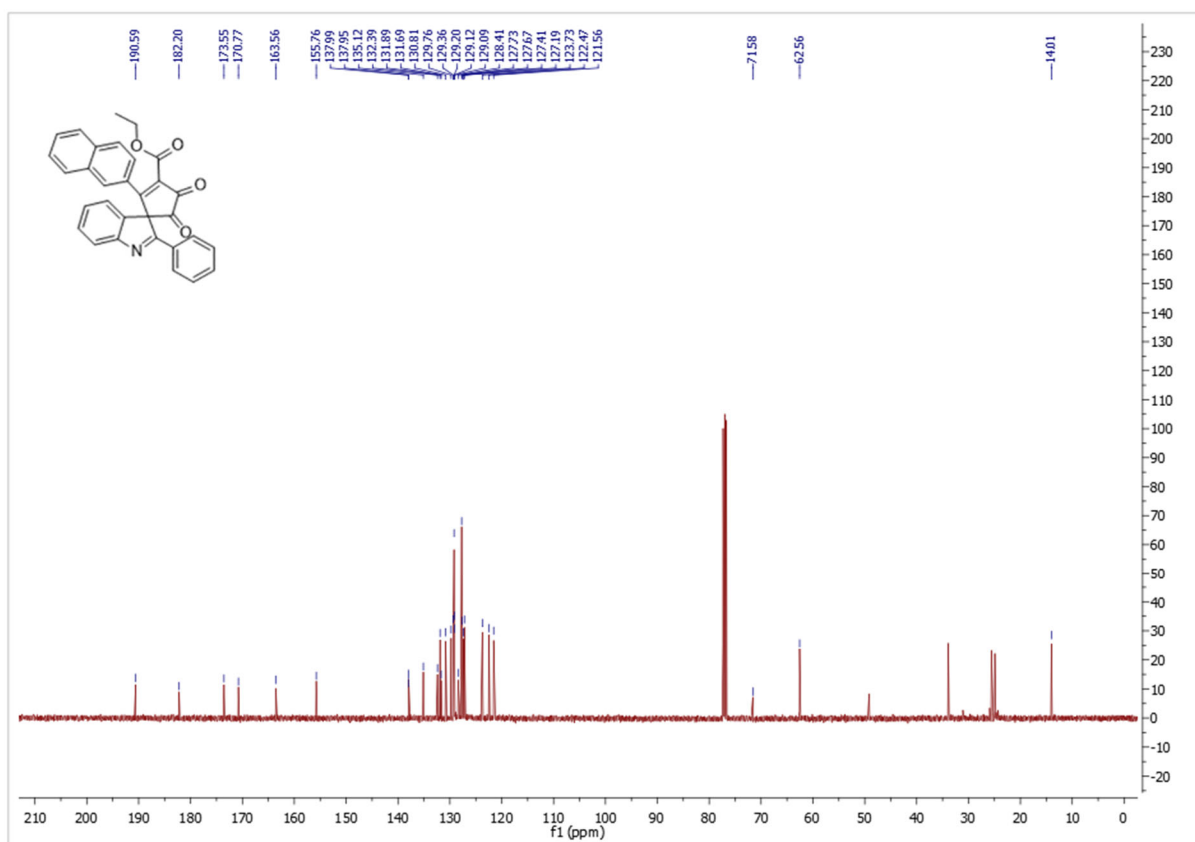
¹H-NMR spectrum of compound 4d



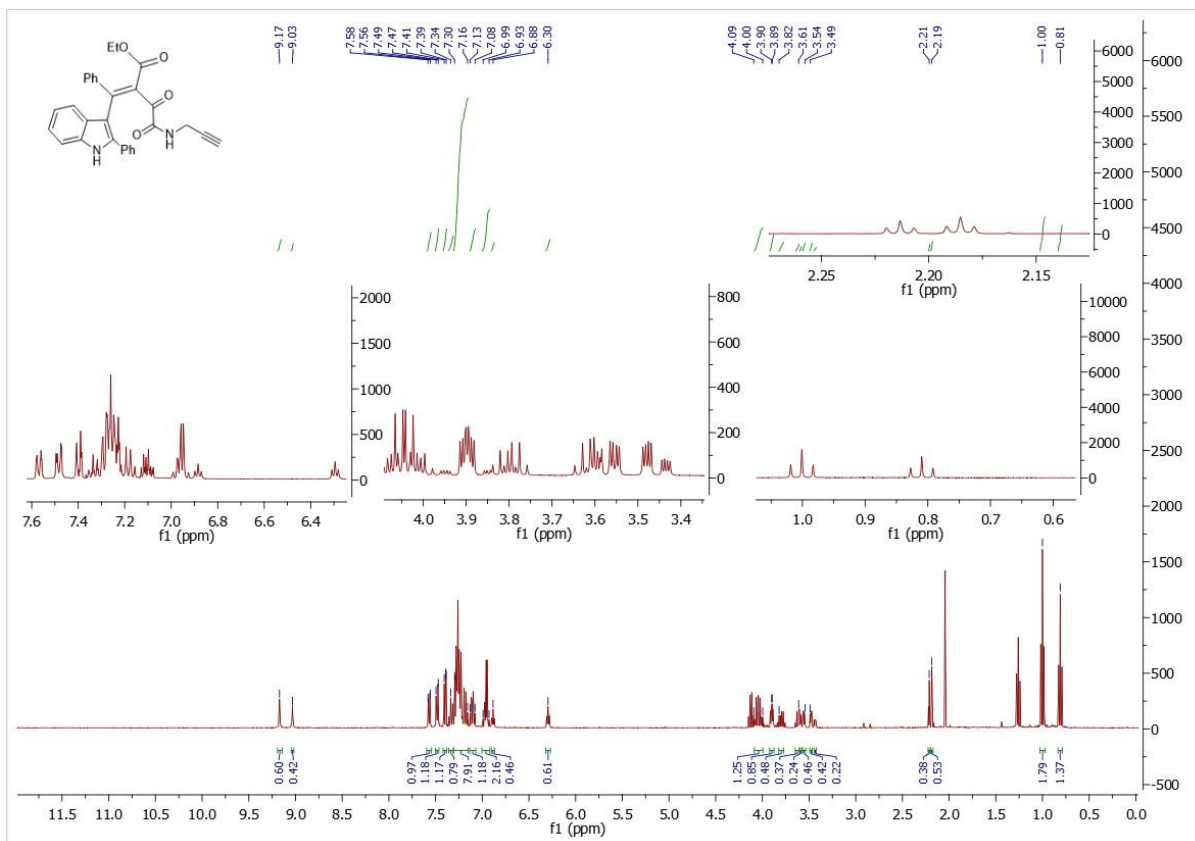
¹³C-NMR spectrum of compound 4d



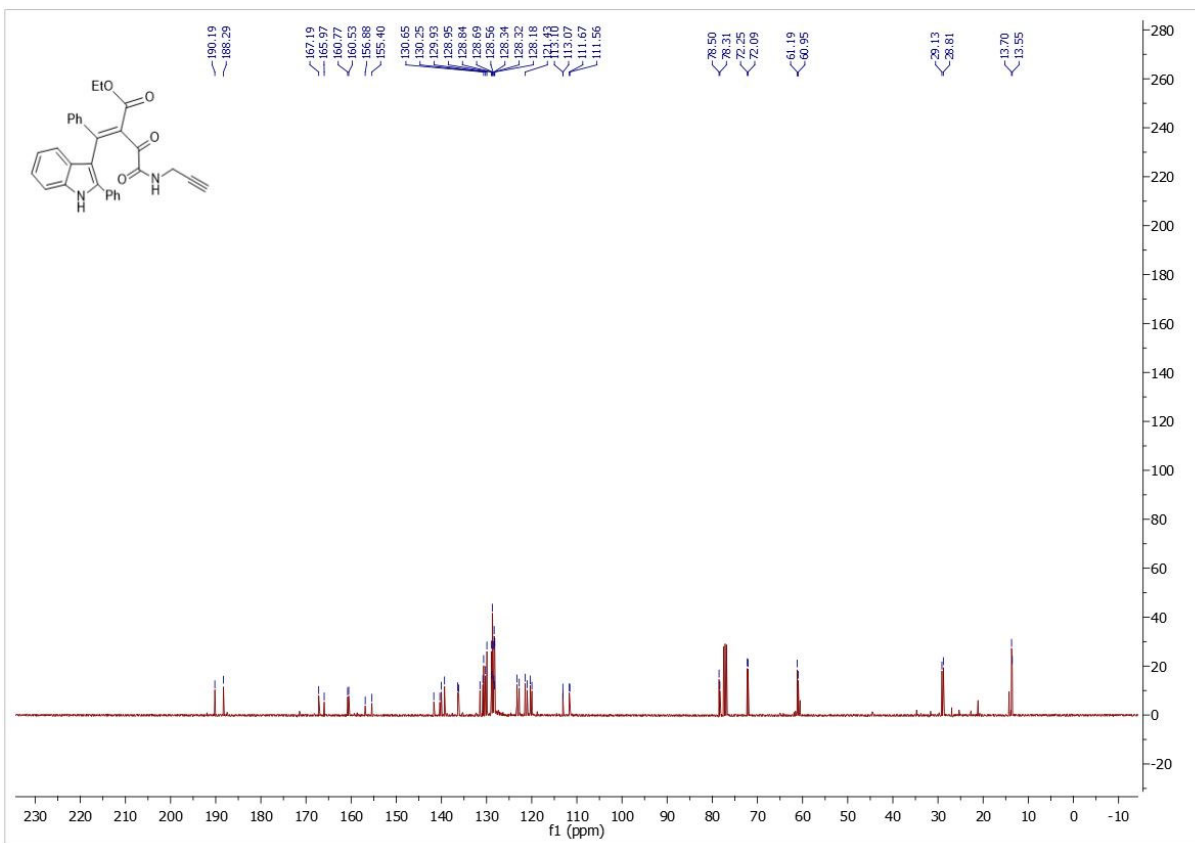
$^1\text{H-NMR}$ spectrum of compound 4e



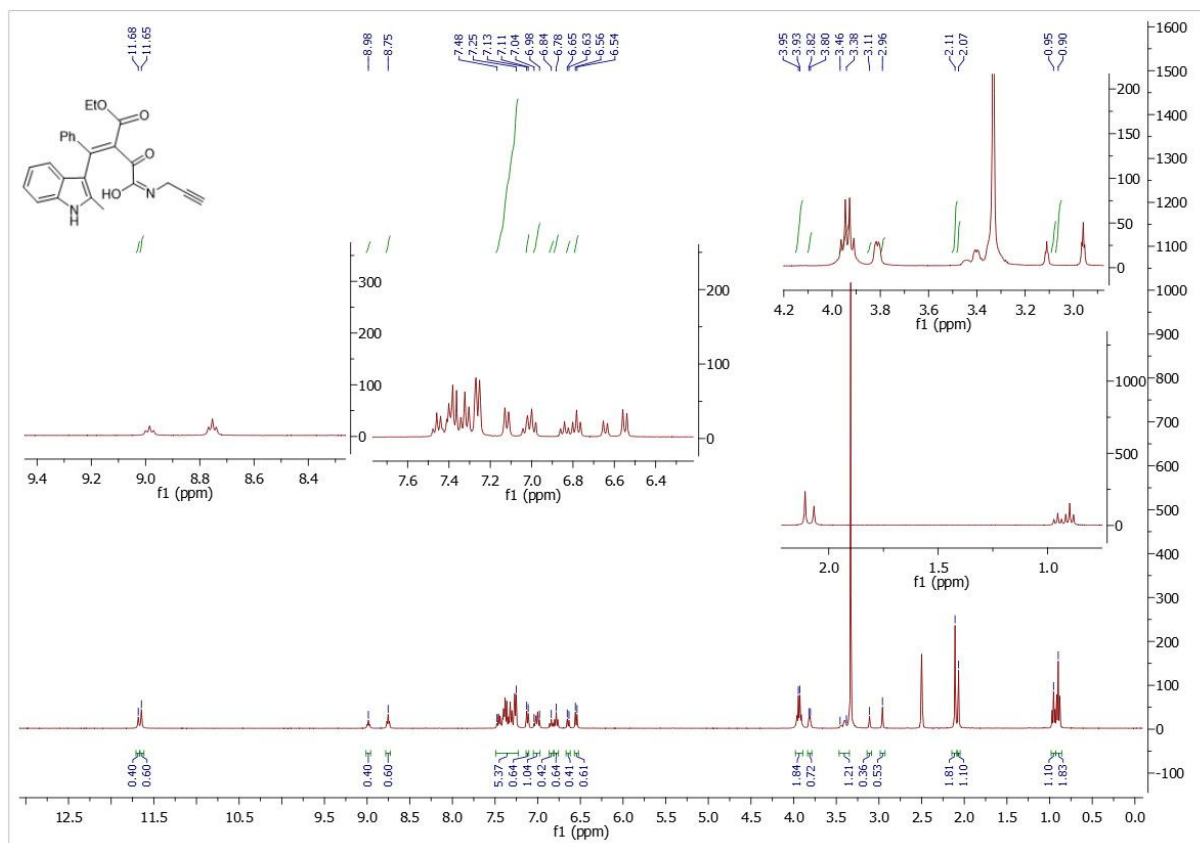
$^{13}\text{C-NMR}$ spectrum of compound 4e



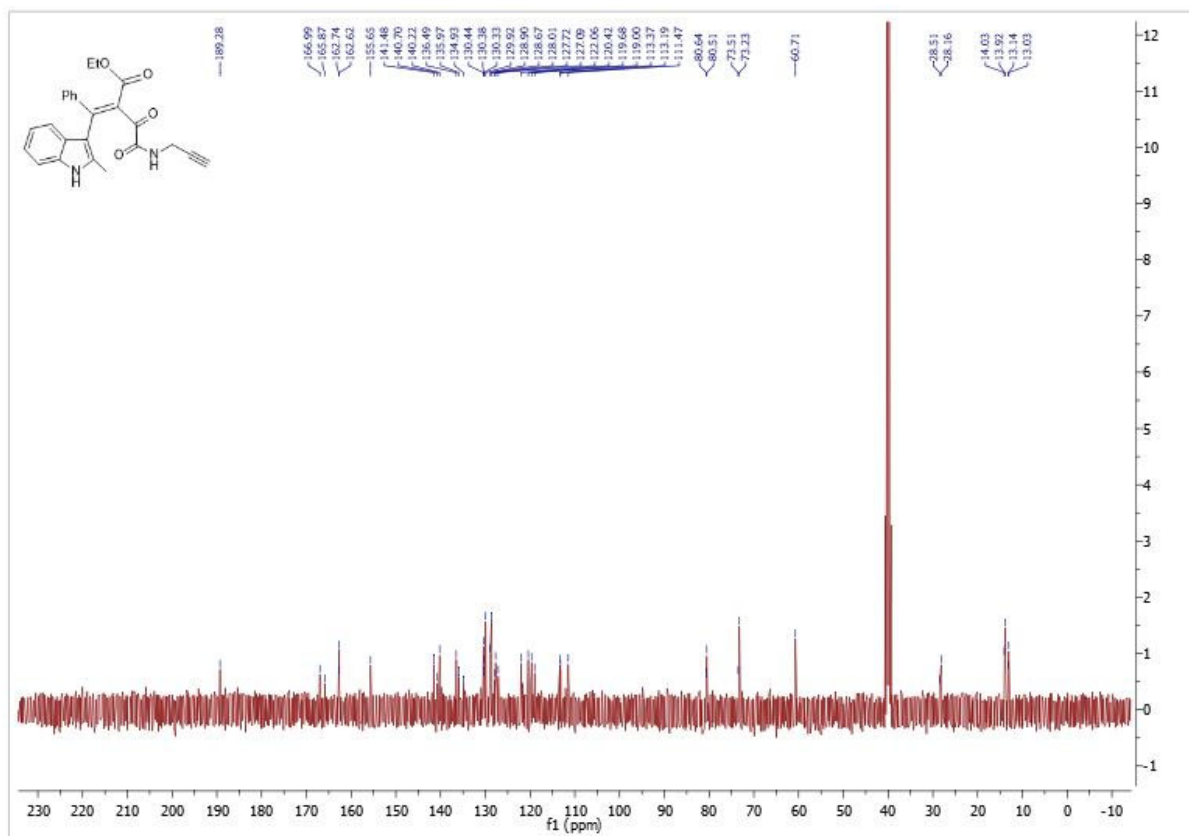
¹H-NMR spectrum of compound 5a



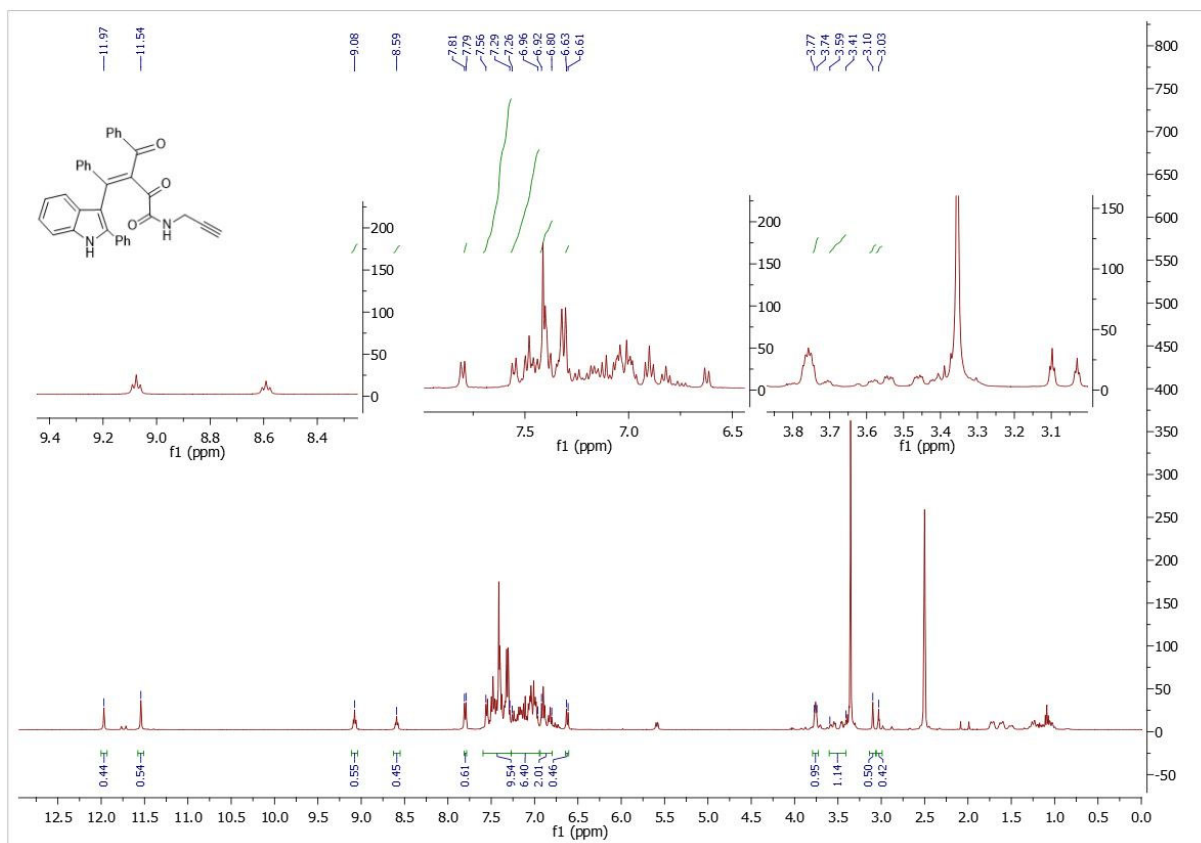
¹³C-NMR spectrum of compound 5a



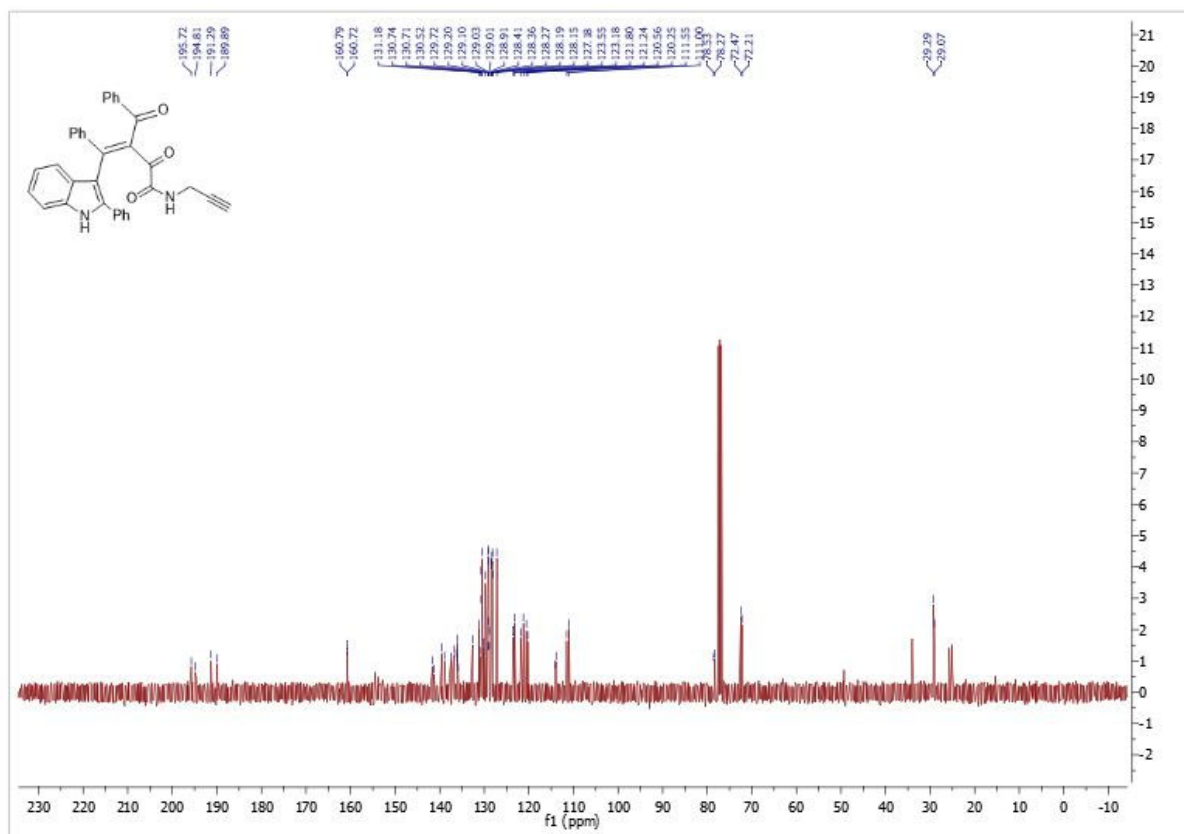
¹H-NMR spectrum of compound 5b



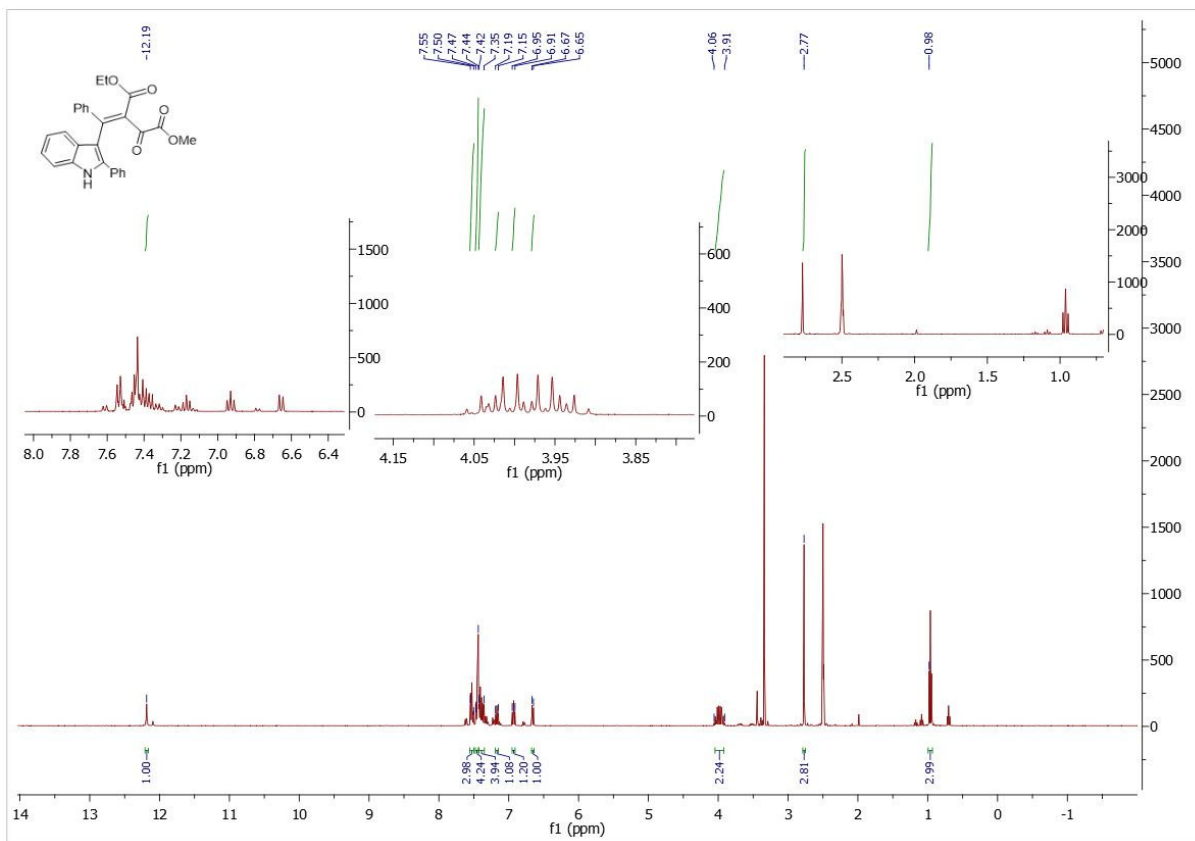
¹³C-NMR spectrum of compound 5b



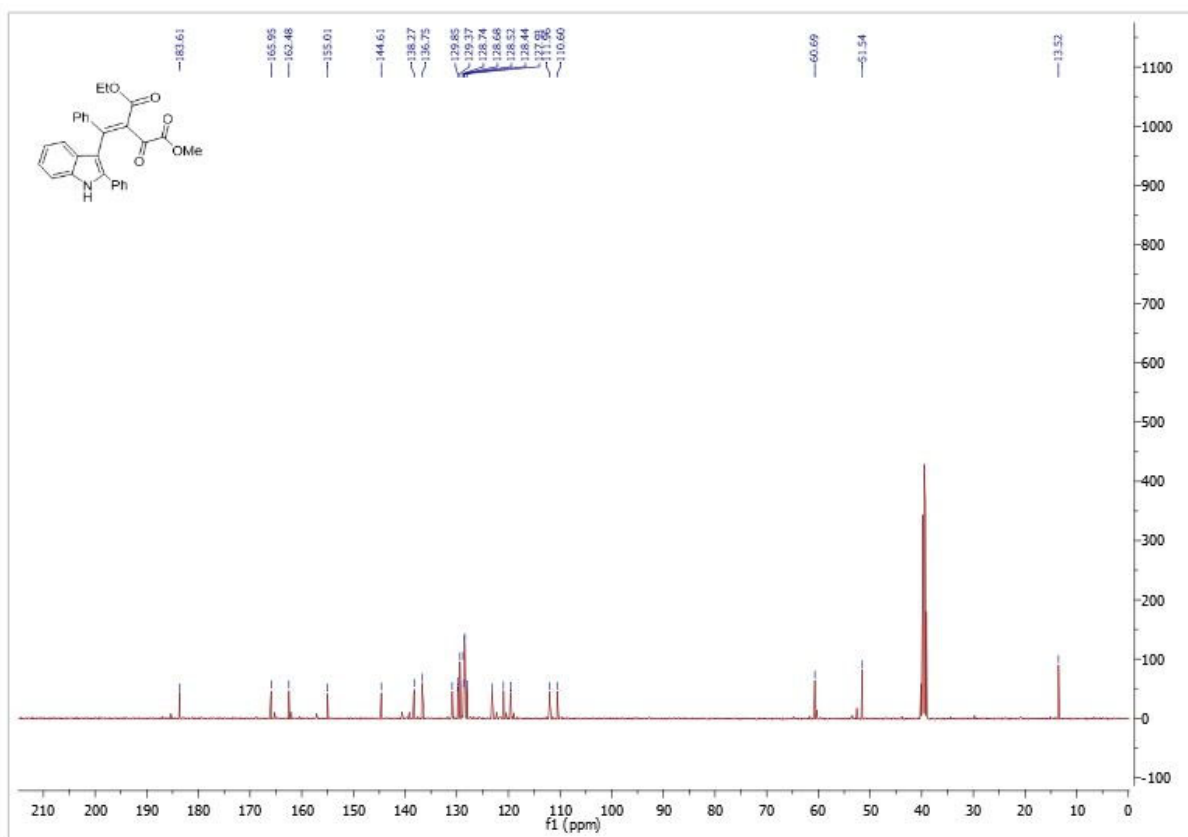
$^1\text{H-NMR}$ spectrum of compound **5c**



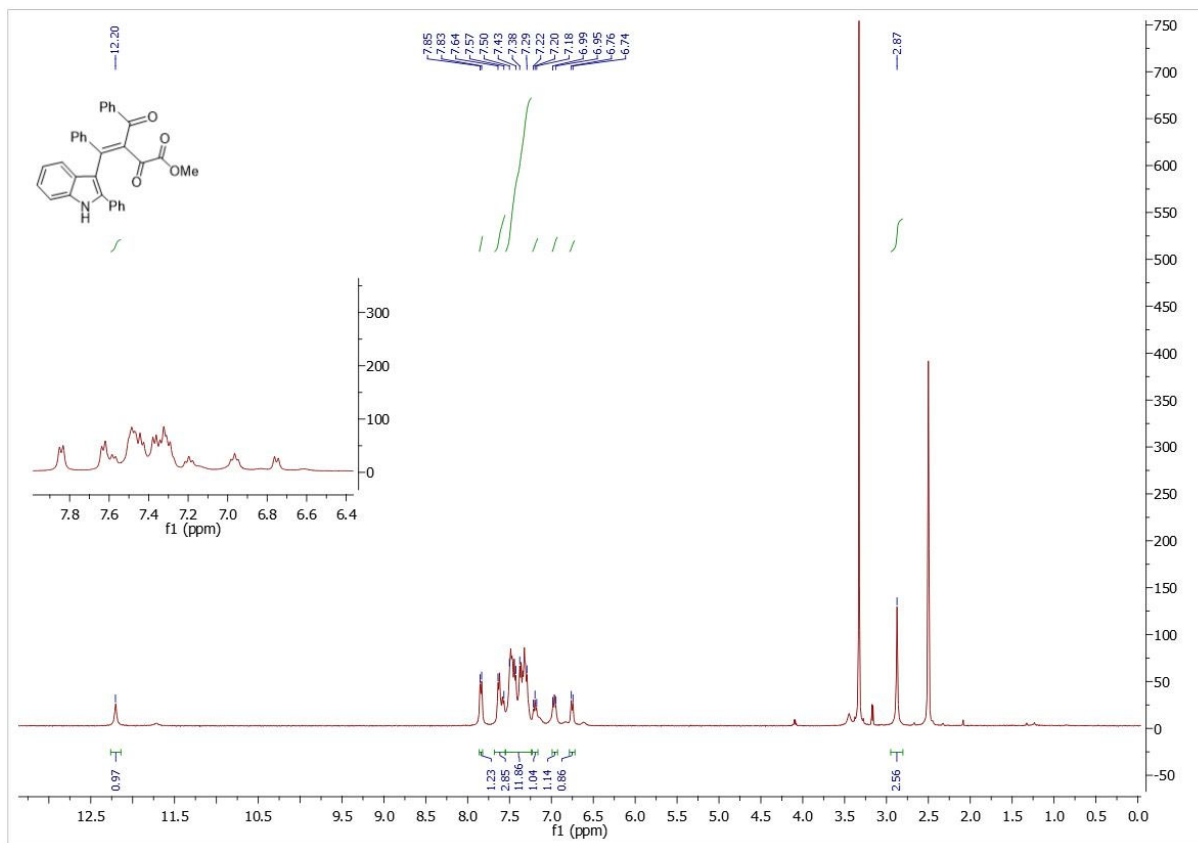
$^{13}\text{C-NMR}$ spectrum of compound **5c**



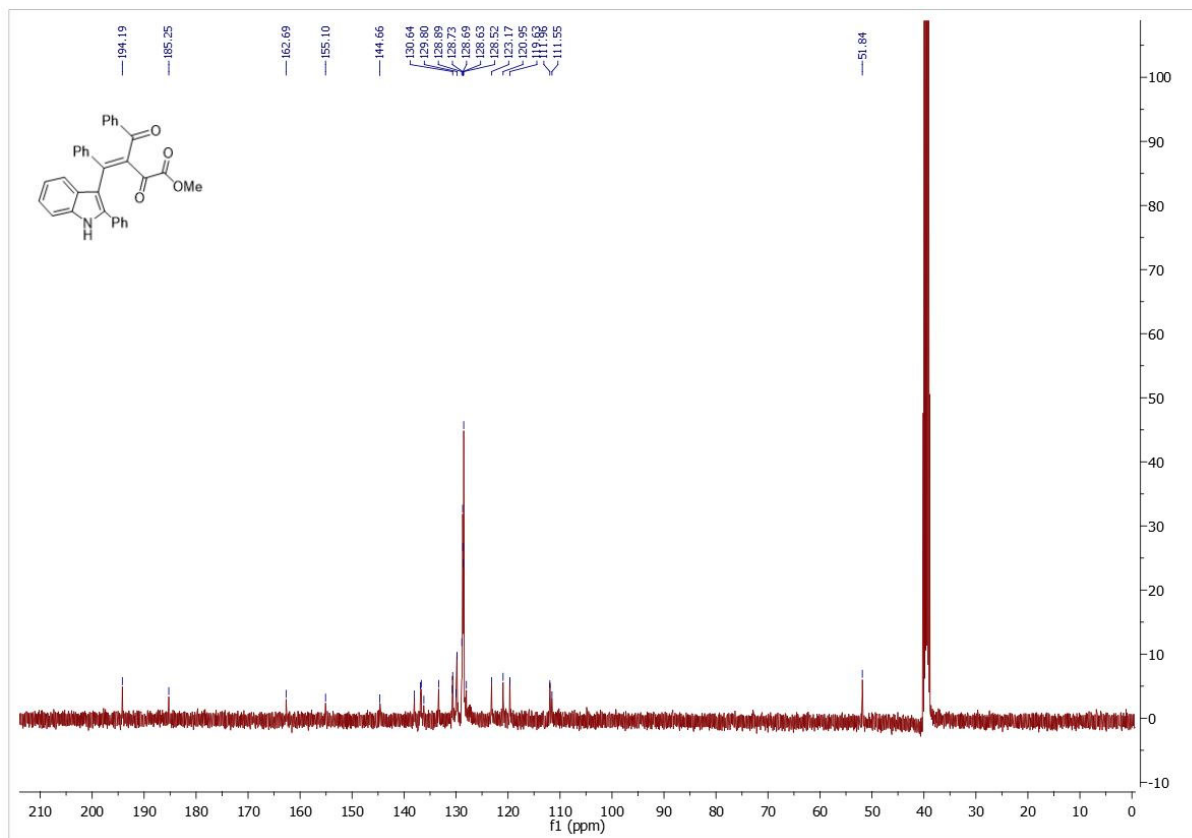
¹H-NMR spectrum of compound 5d



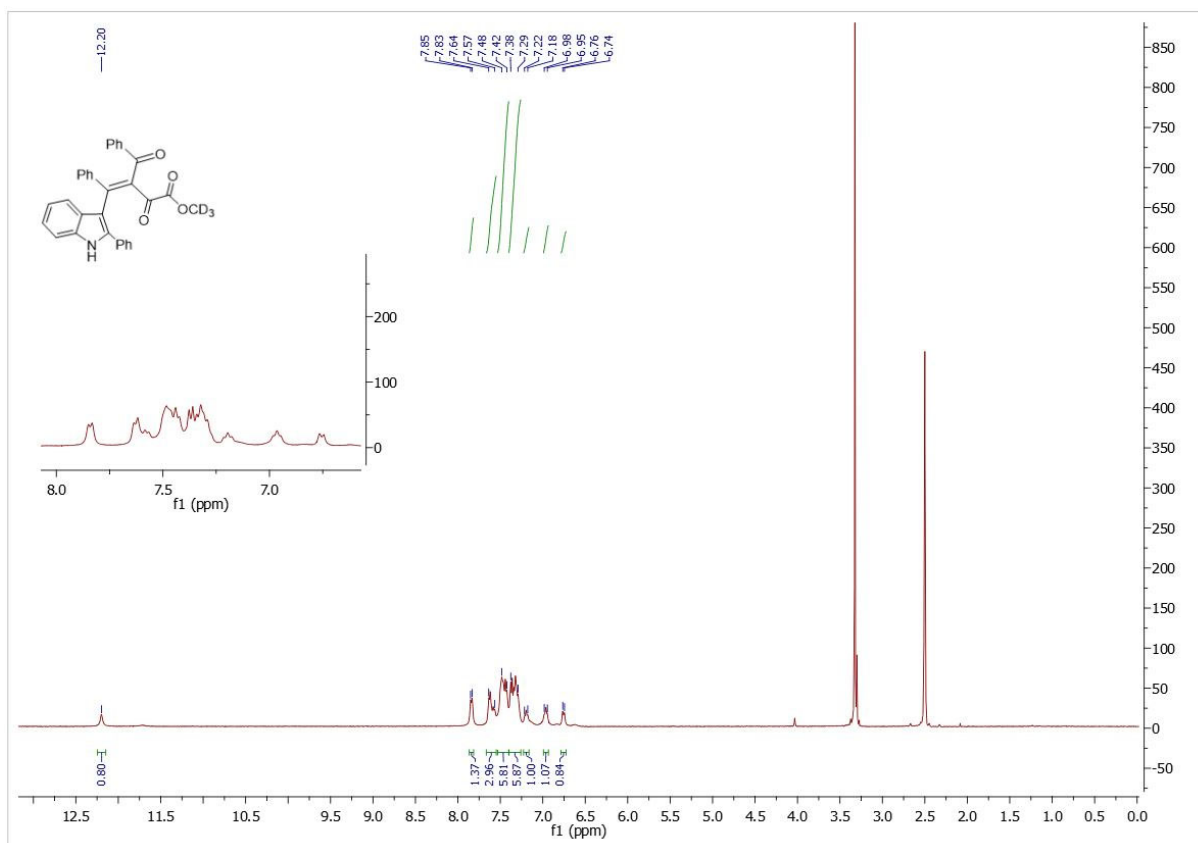
¹³C-NMR spectrum of compound 5d



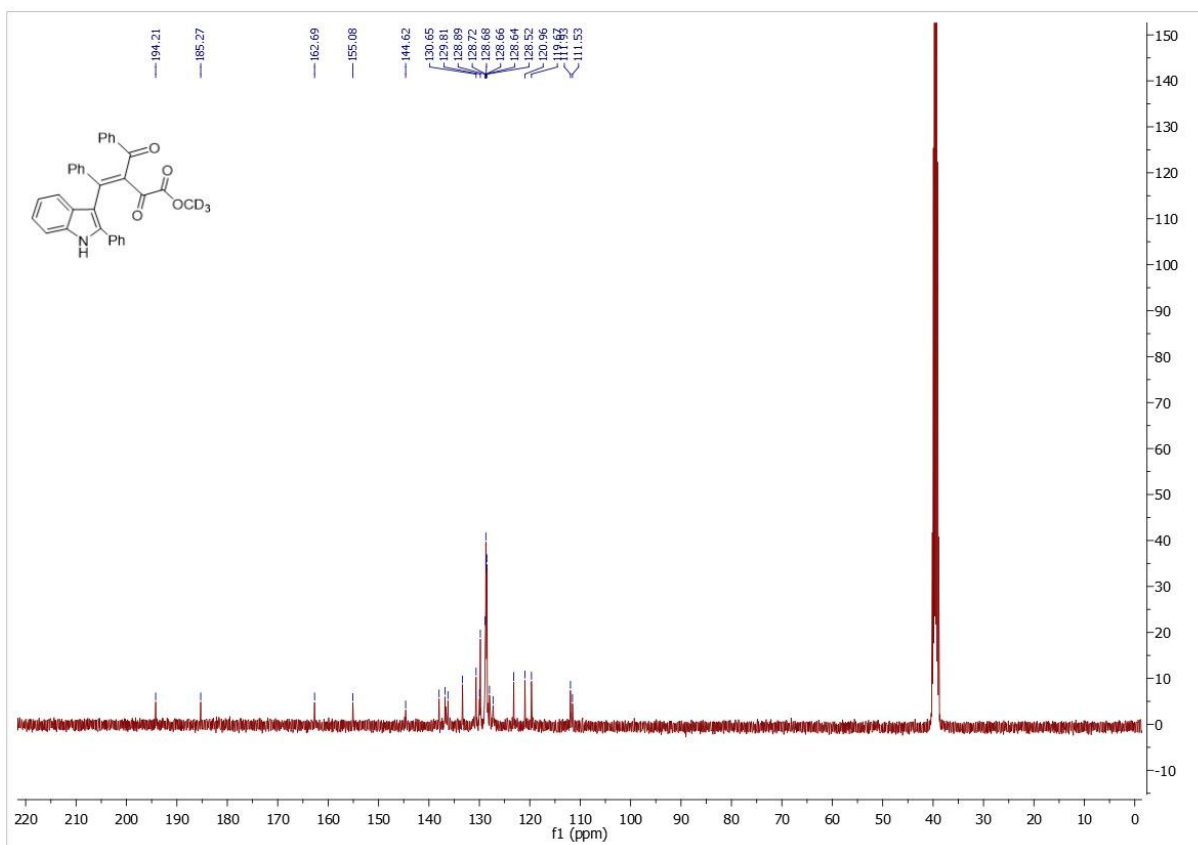
¹H-NMR spectrum of compound 5e



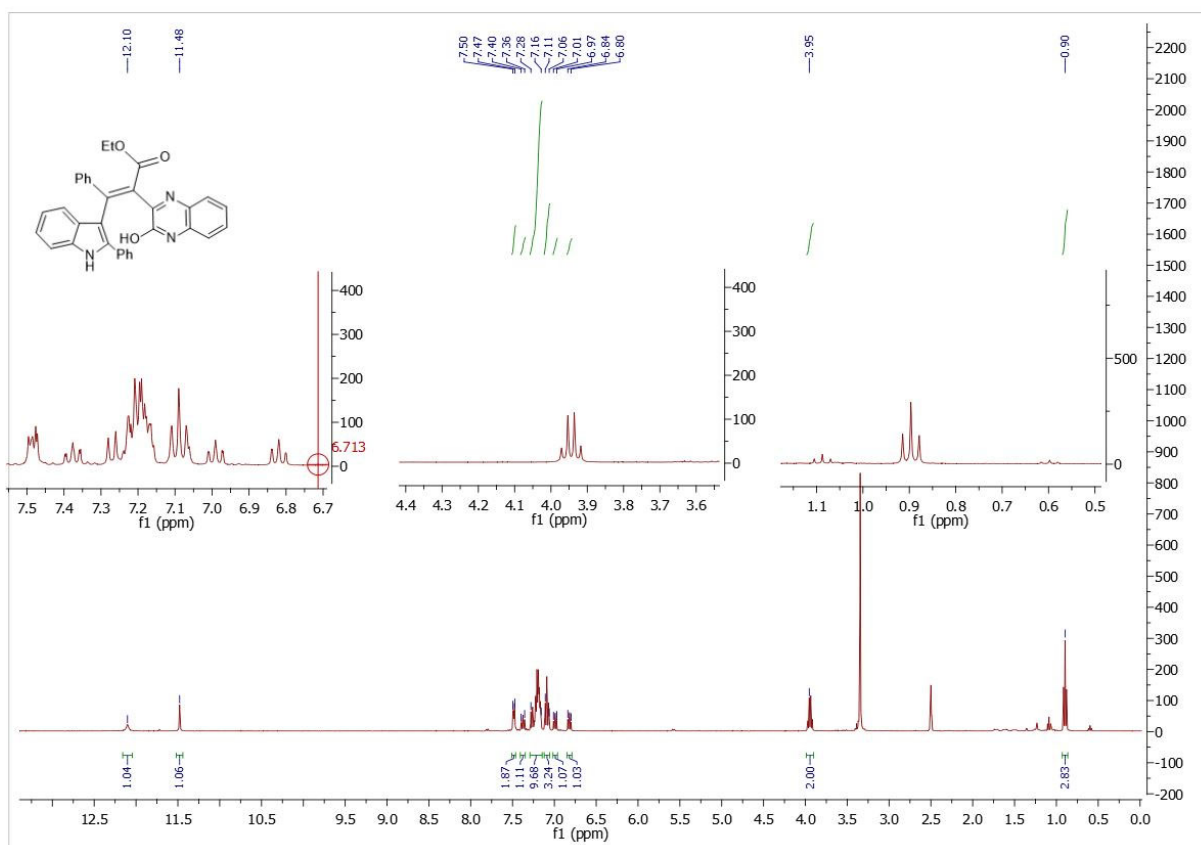
¹³C-NMR spectrum of compound 5e



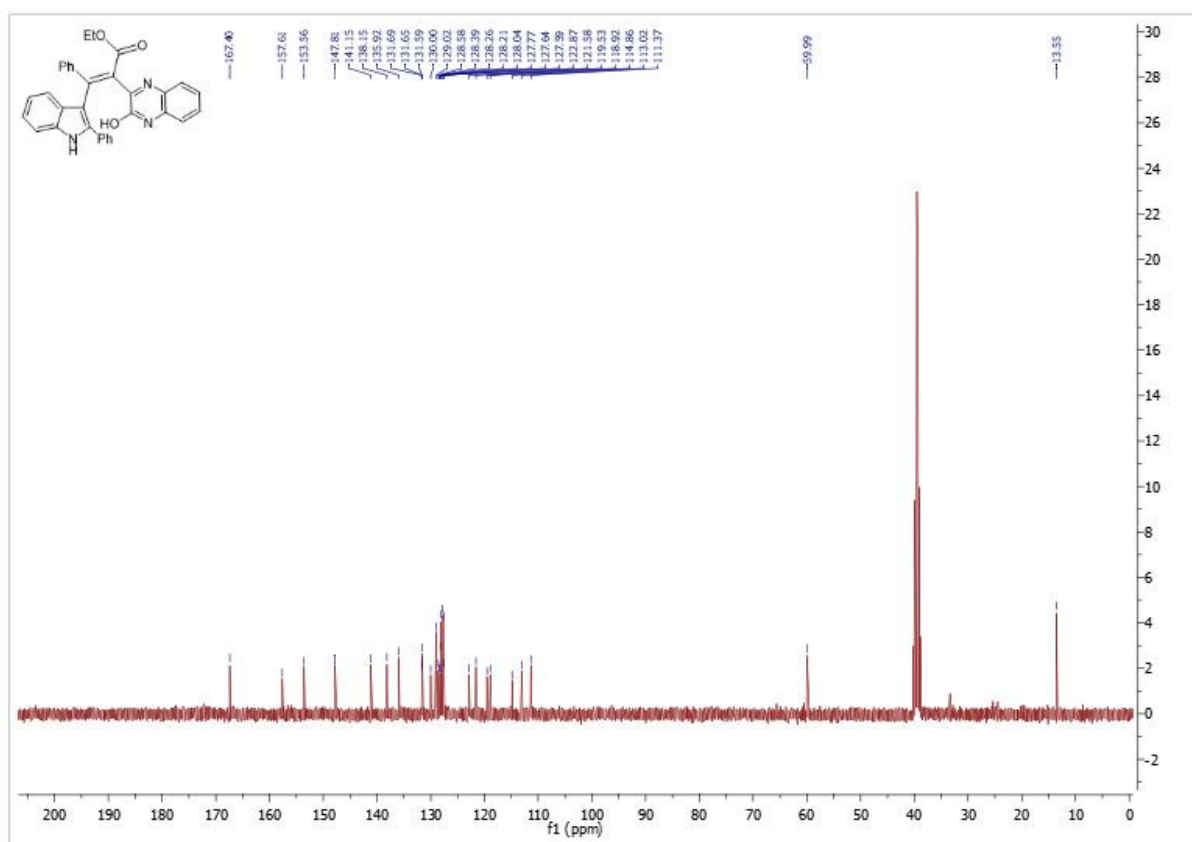
¹H-NMR spectrum of compound **5f**



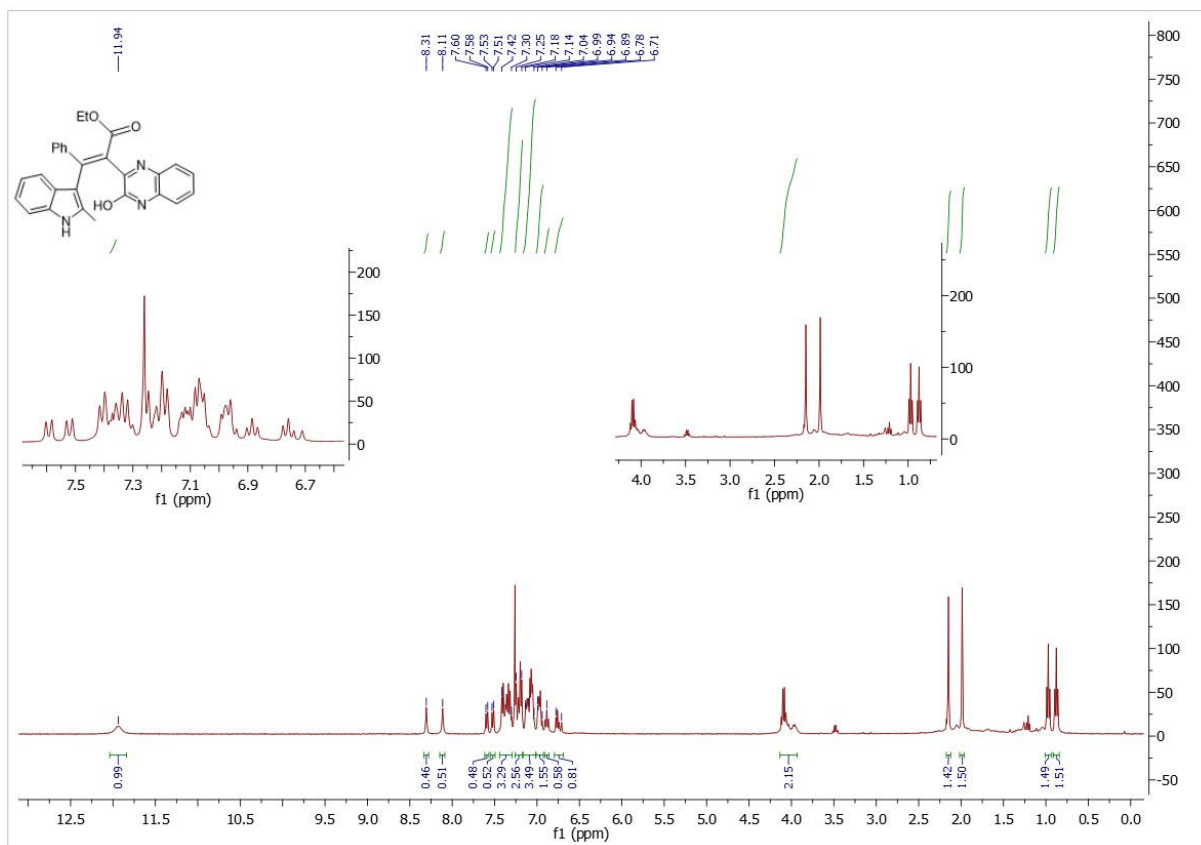
¹³C-NMR spectrum of compound **5f**



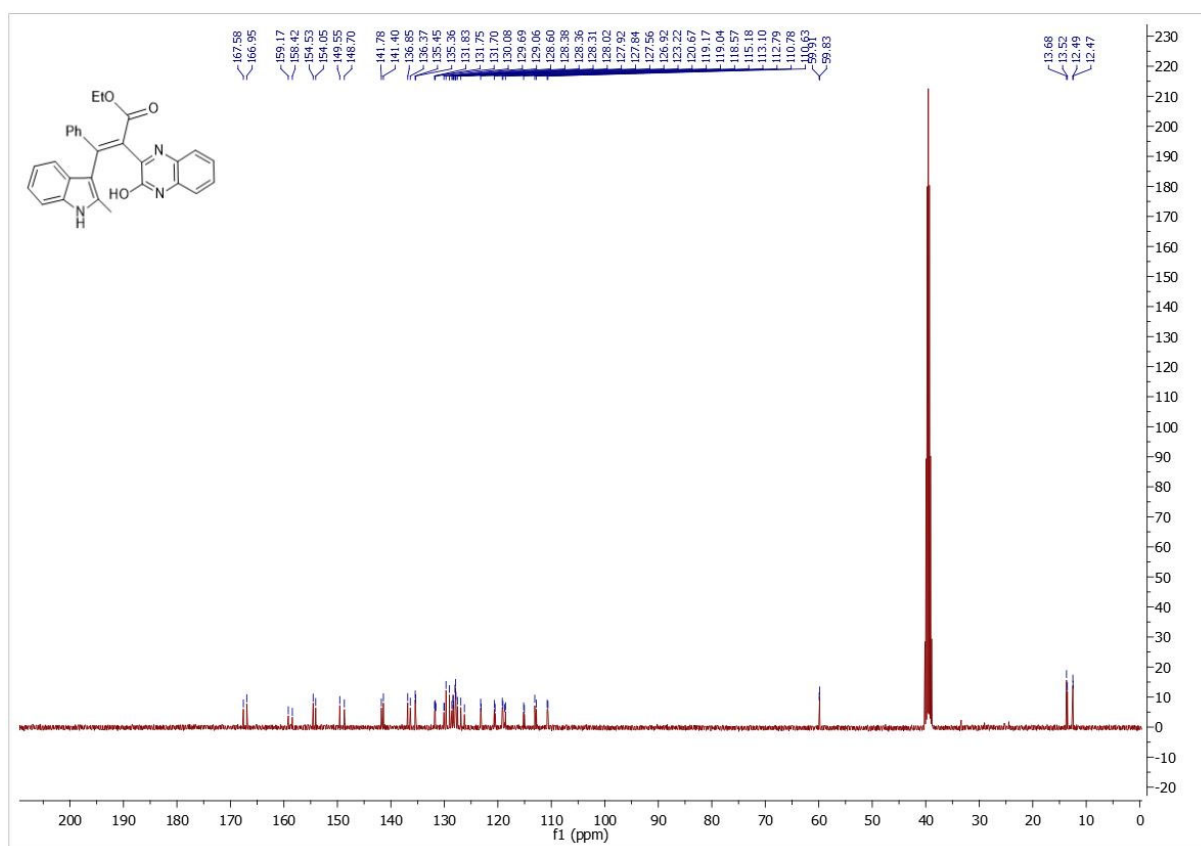
¹H-NMR spectrum of compound 7a



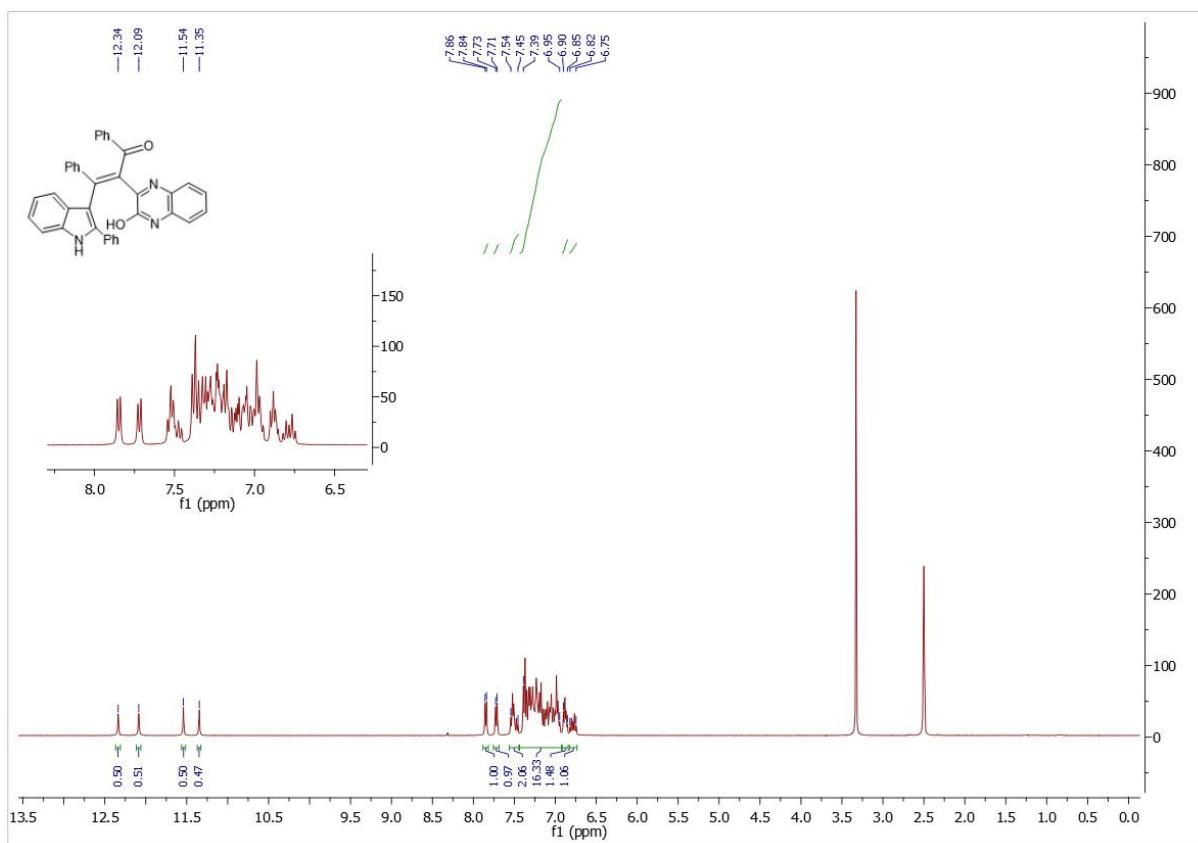
¹³C-NMR spectrum of compound 7a



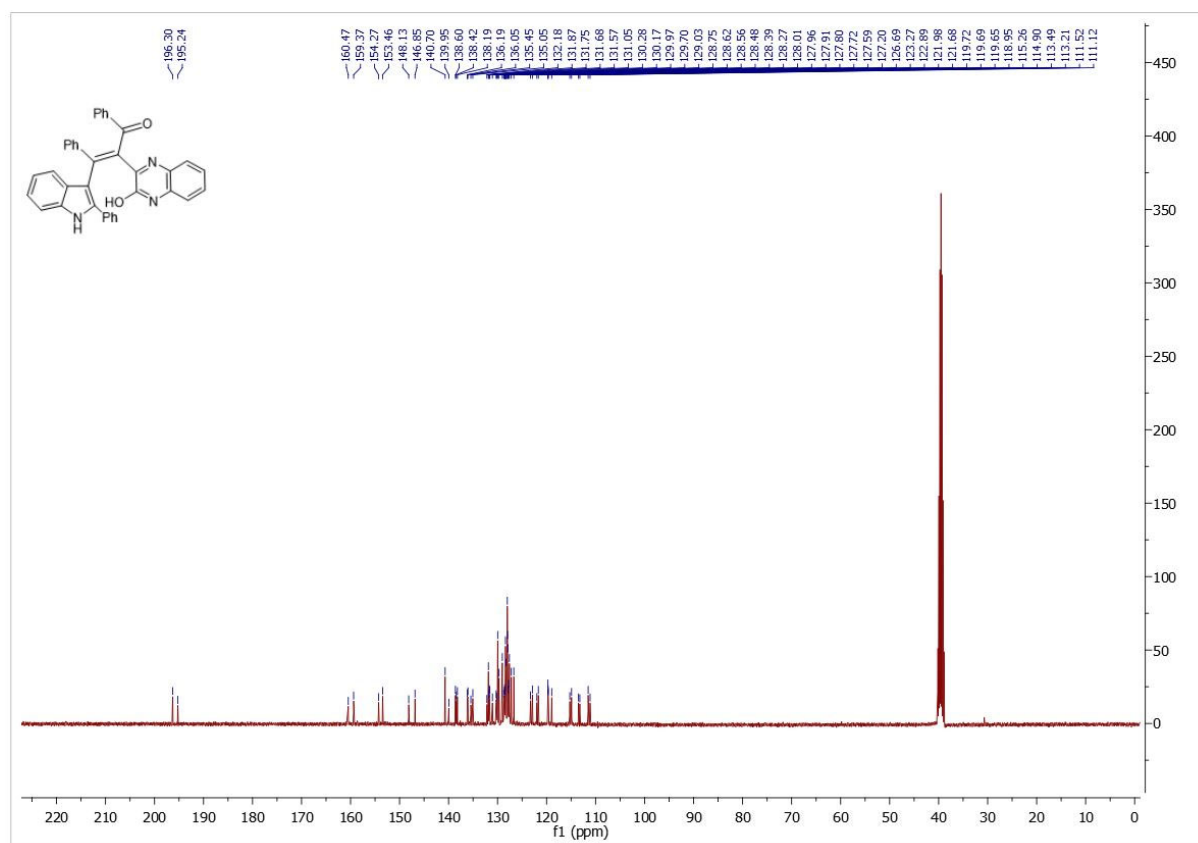
¹H-NMR spectrum of compound 7b



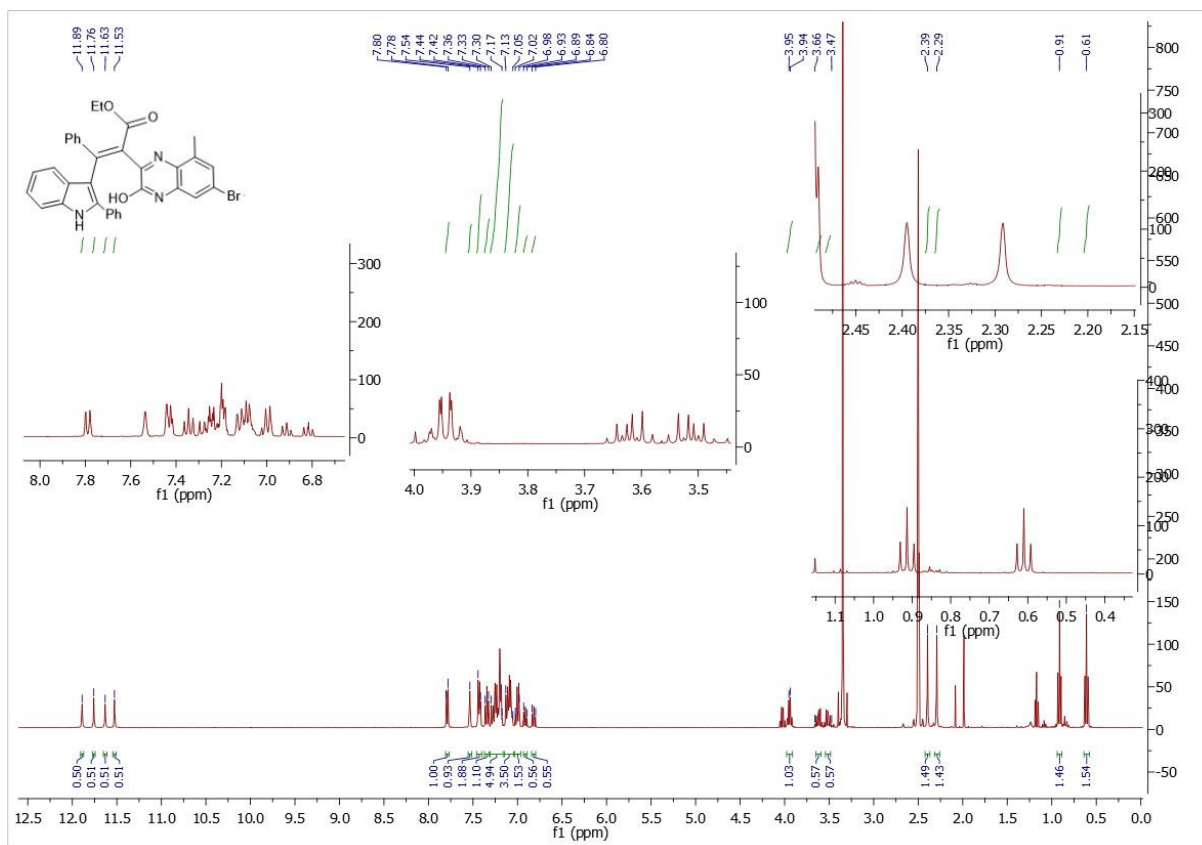
¹³C-NMR spectrum of compound 7b



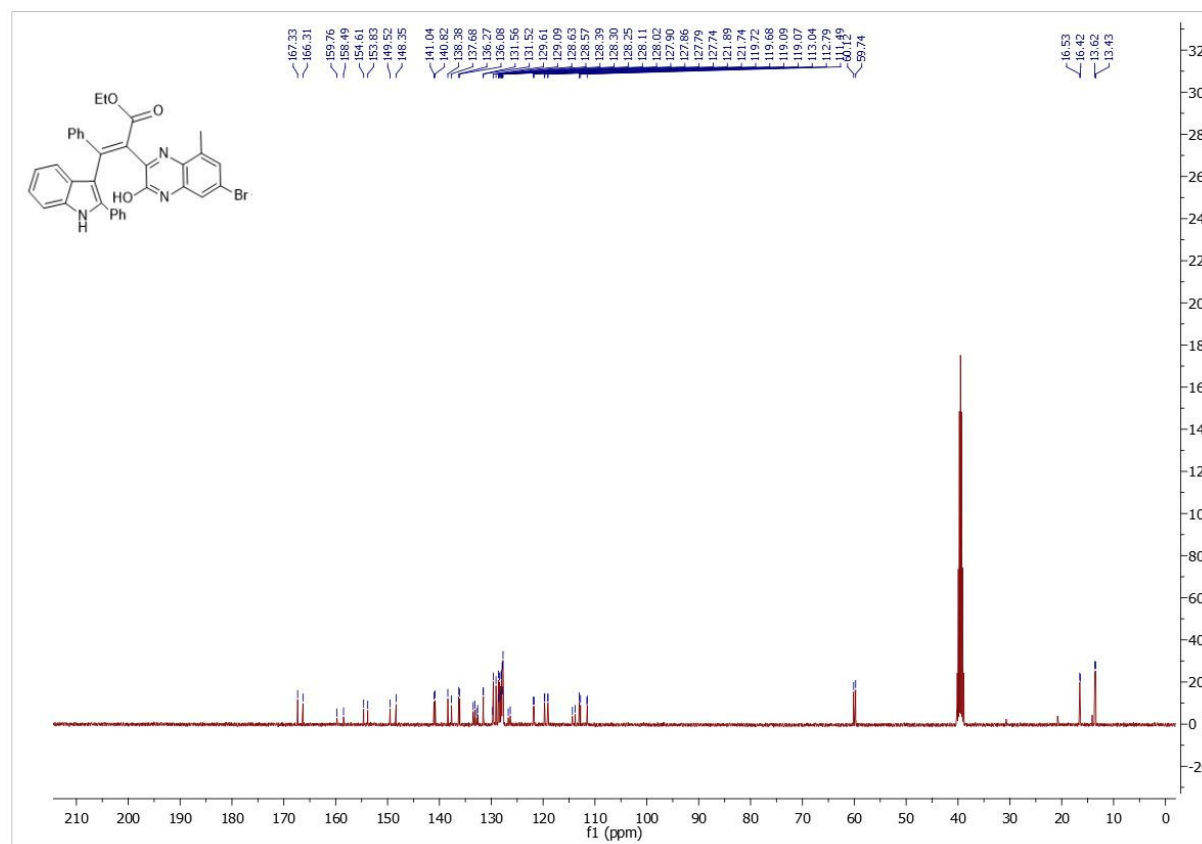
¹H-NMR spectrum of compound 7c



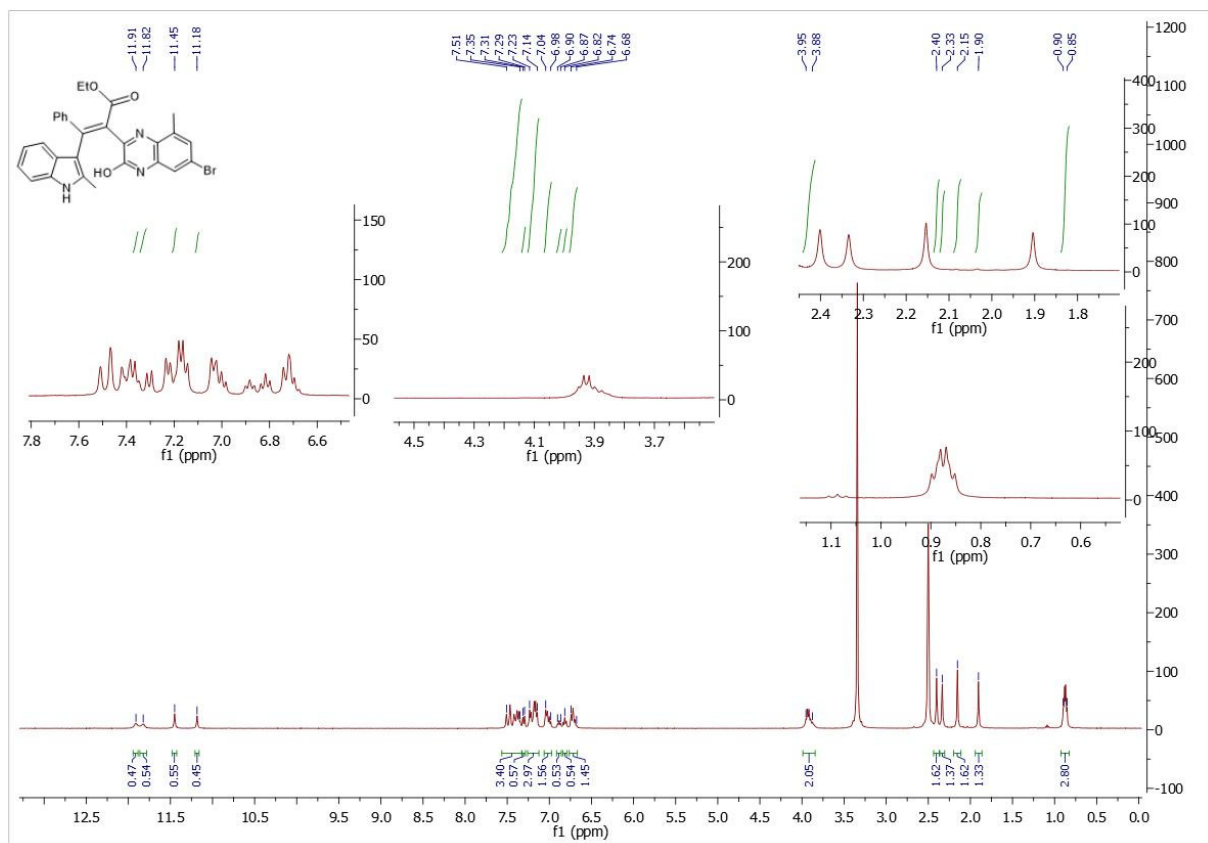
¹³C-NMR spectrum of compound 7c



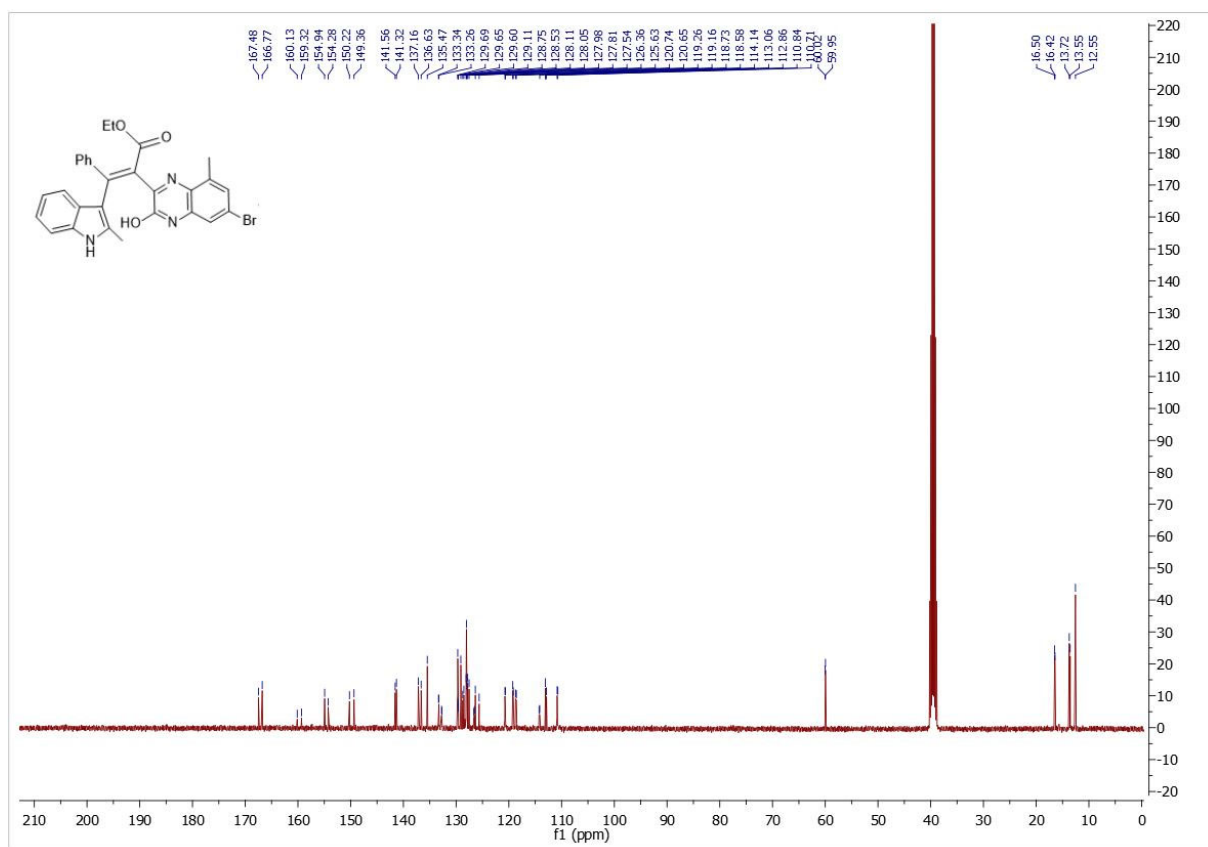
¹H-NMR spectrum of compound 7d



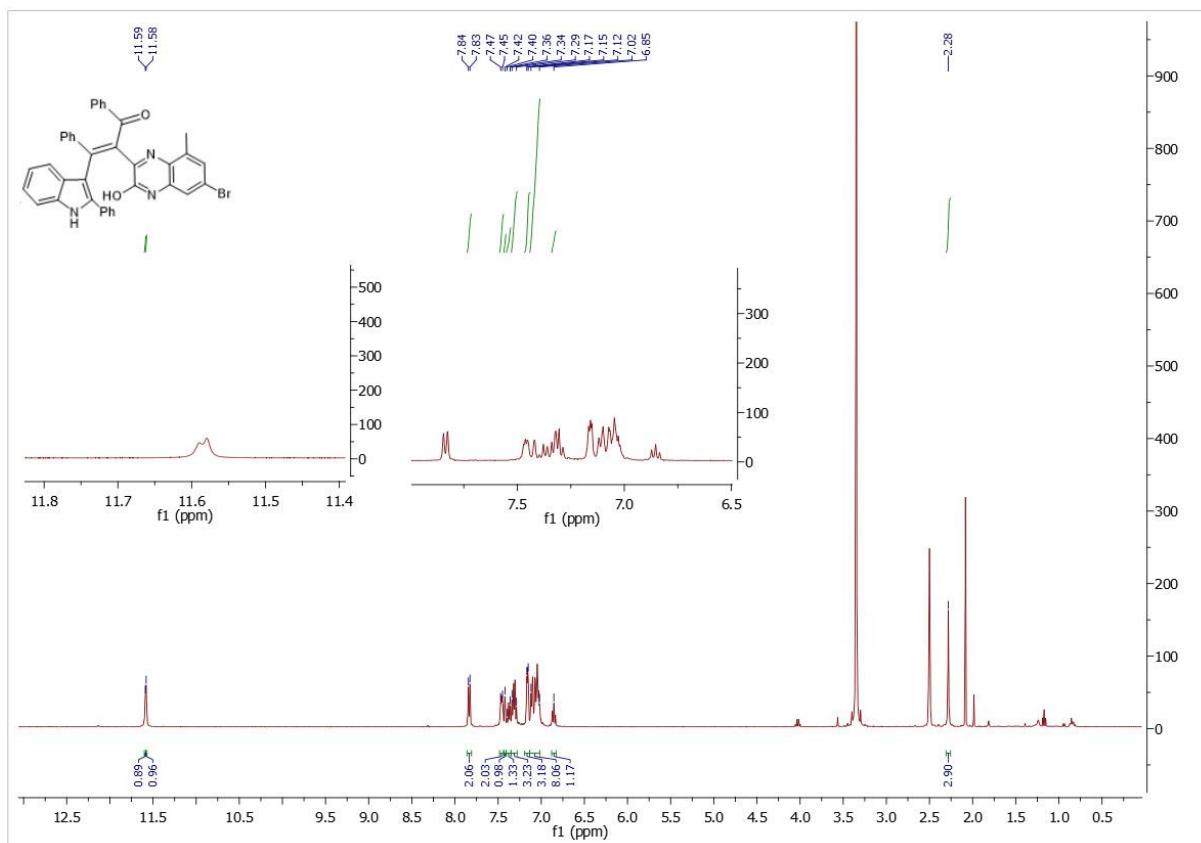
¹³C-NMR spectrum of compound 7d



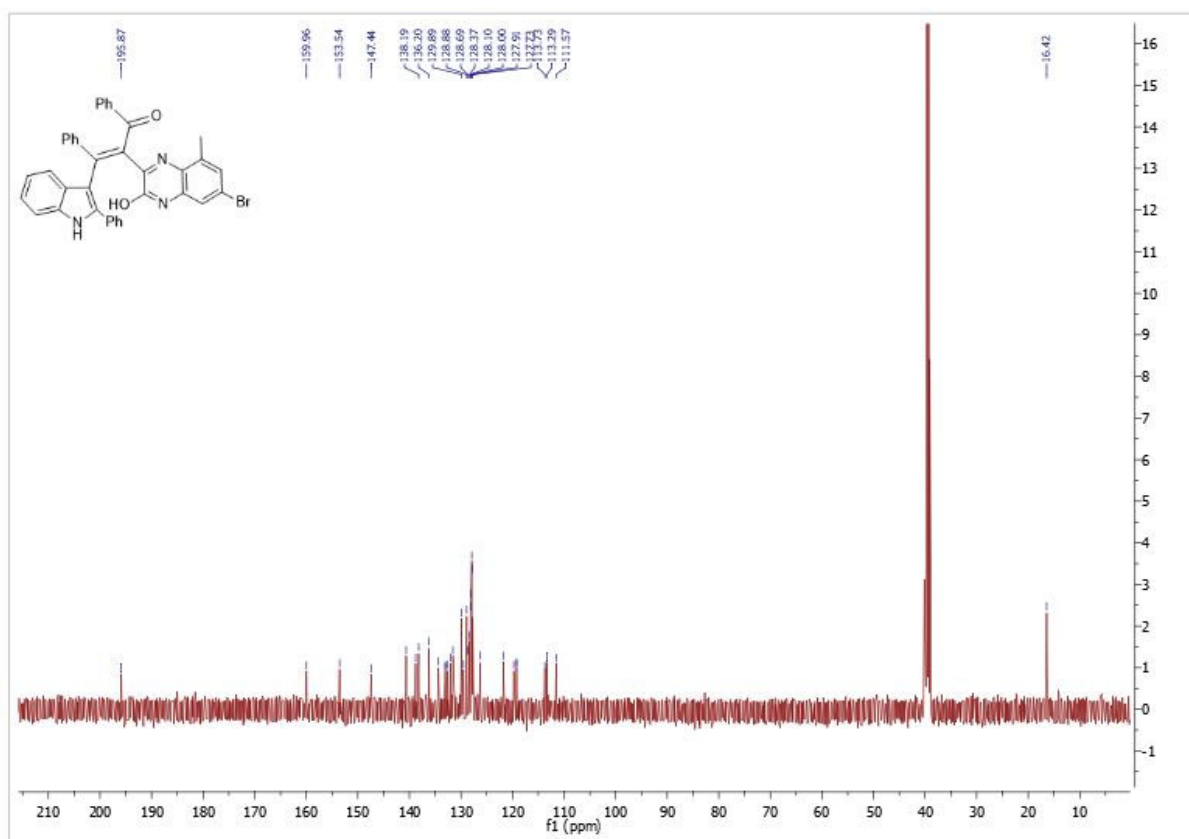
¹H-NMR spectrum of compound 7e



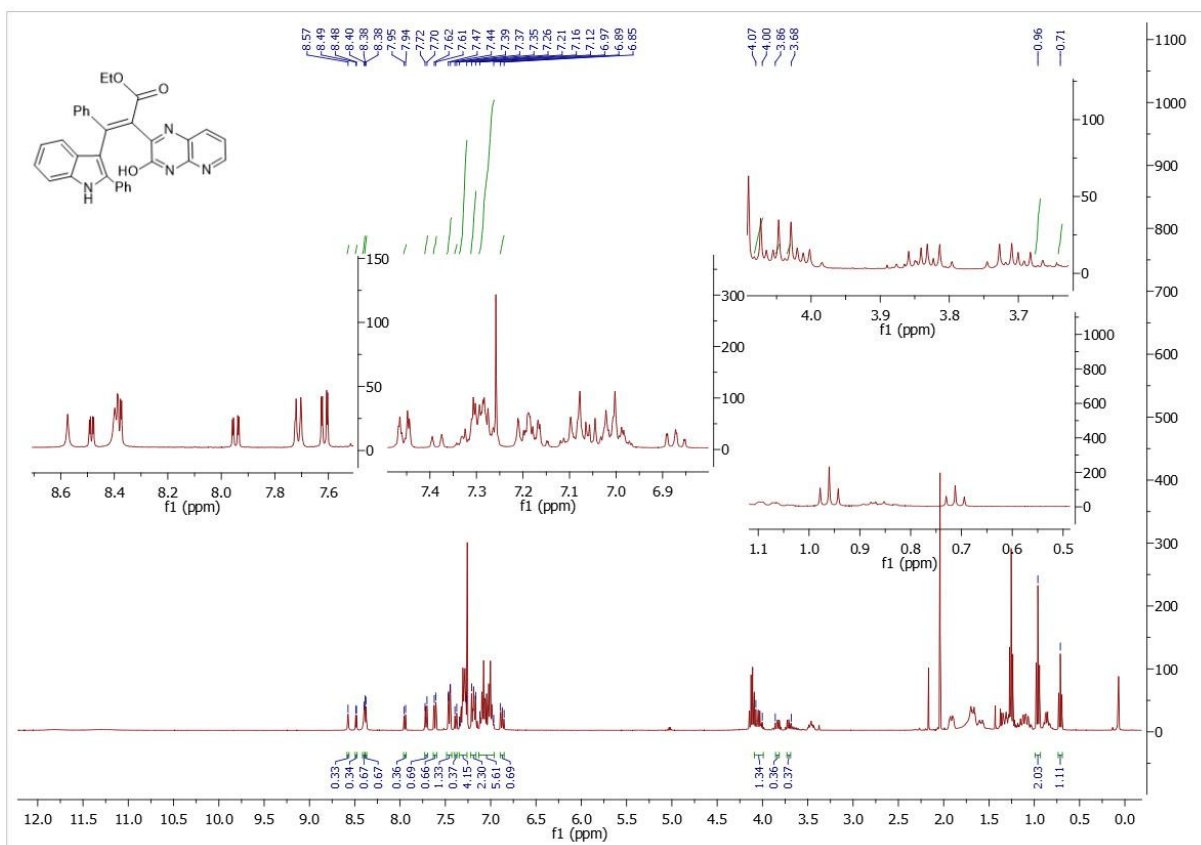
¹³C-NMR spectrum of compound 7e



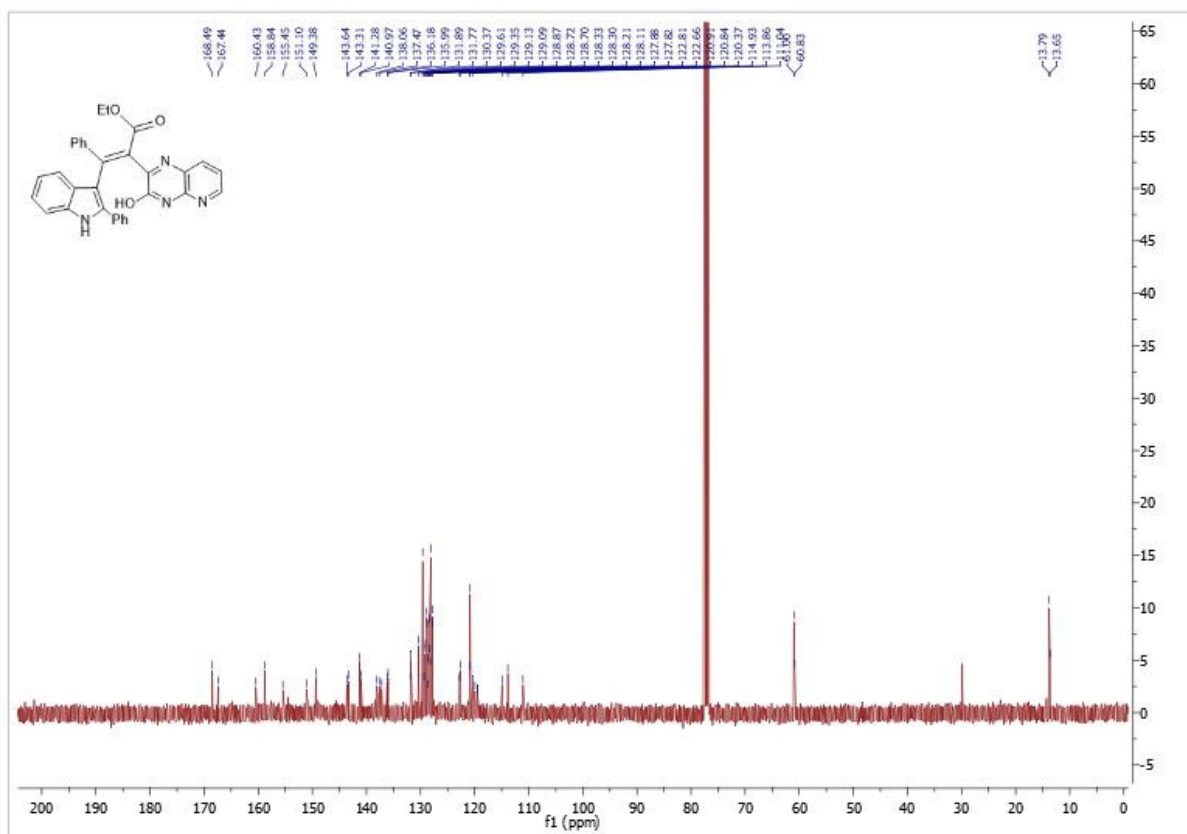
¹H-NMR spectrum of compound 7f



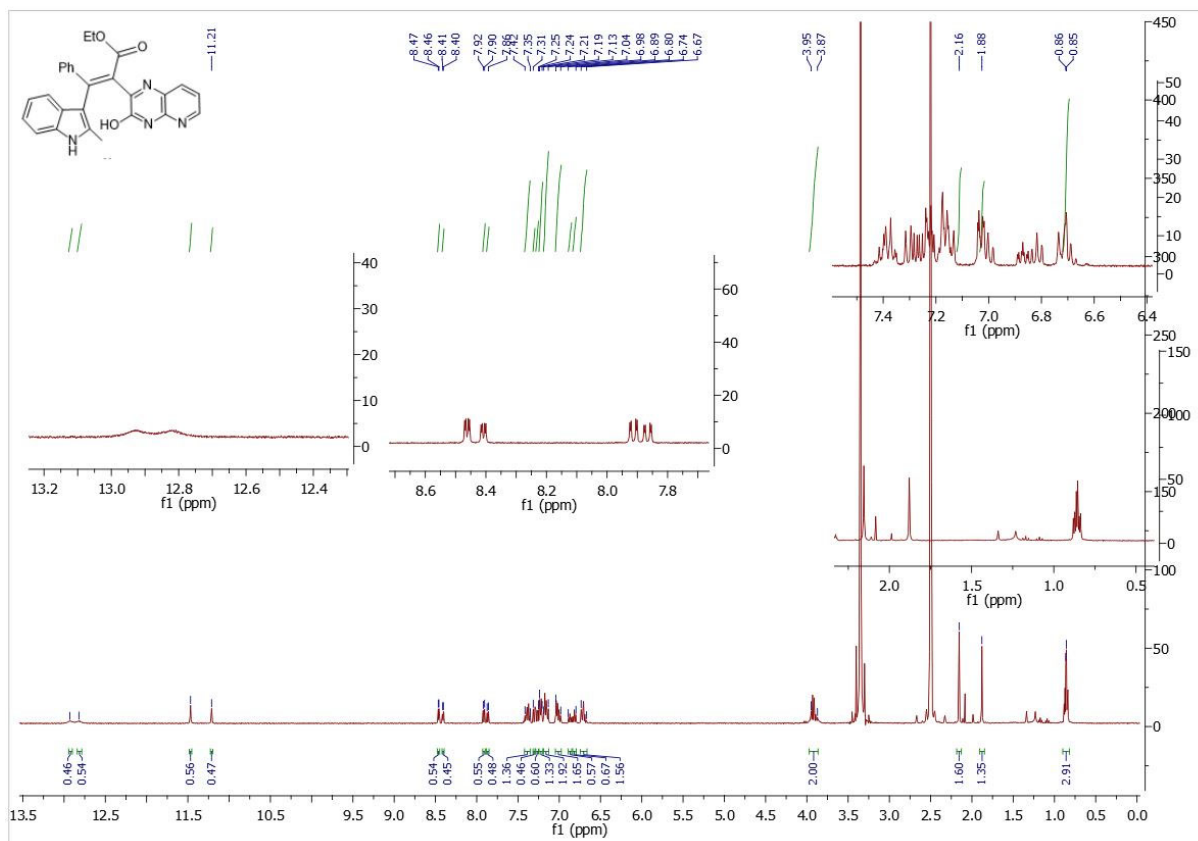
¹³C-NMR spectrum of compound 7f



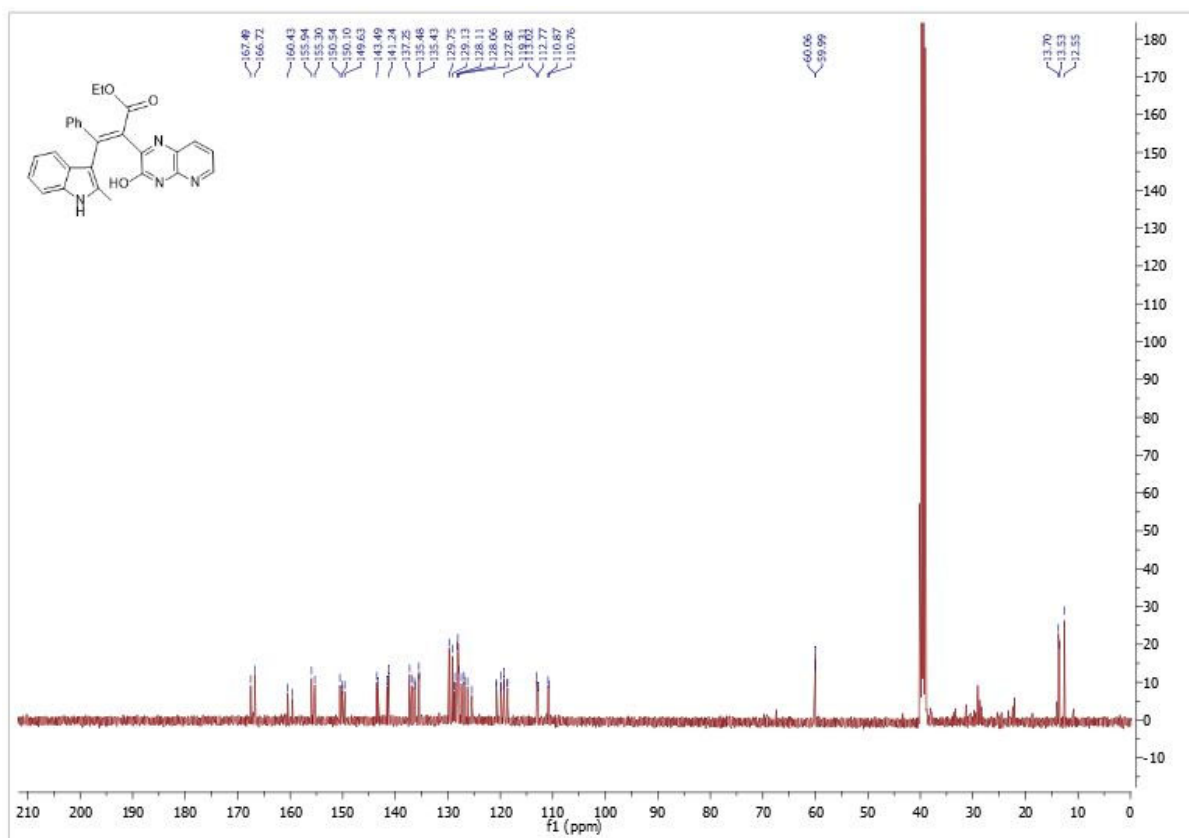
¹H-NMR spectrum of compound 7g



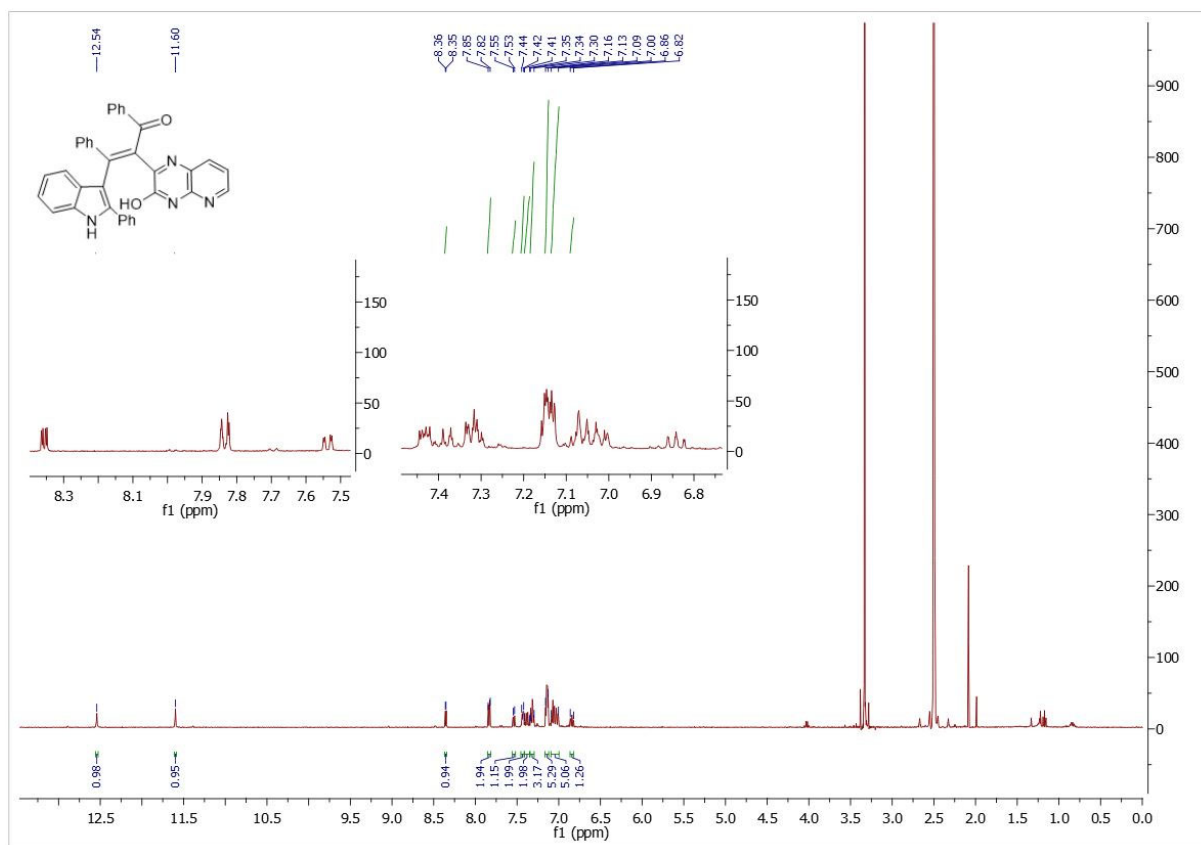
¹³C-NMR spectrum of compound 7g



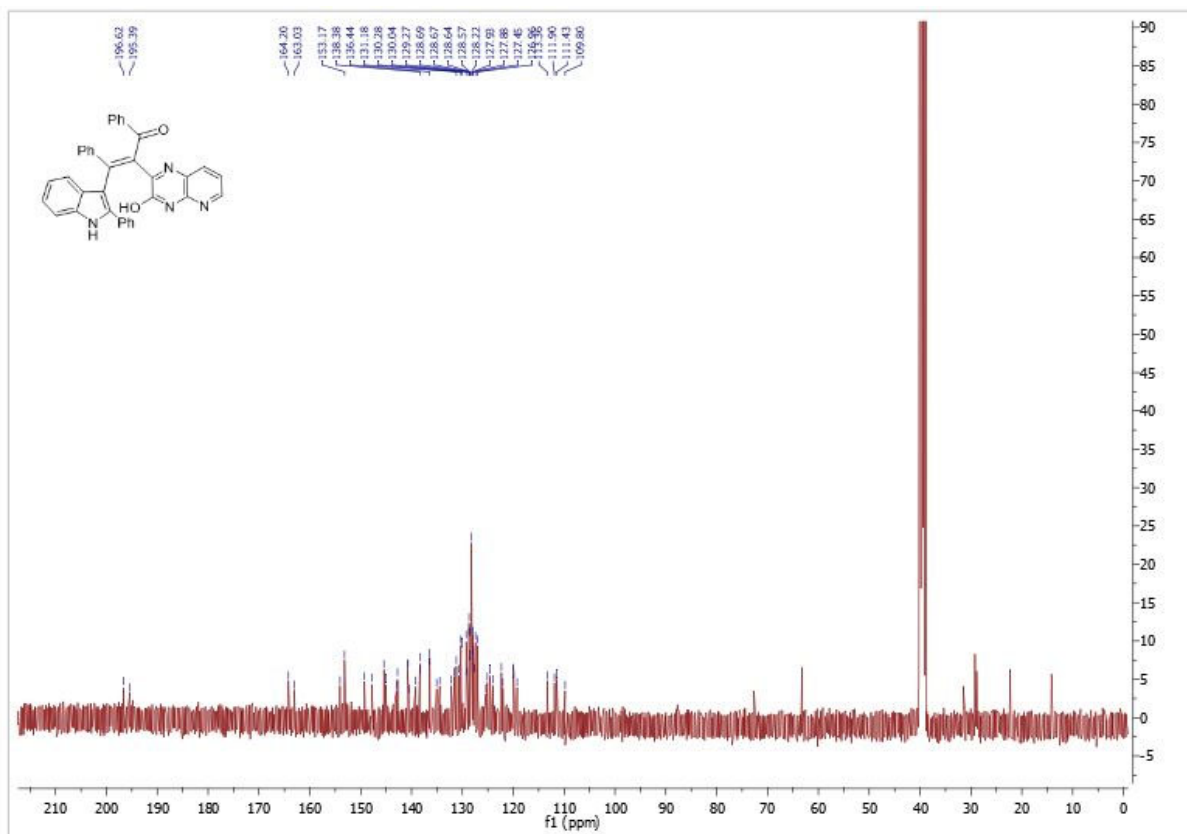
¹H-NMR spectrum of compound 7h



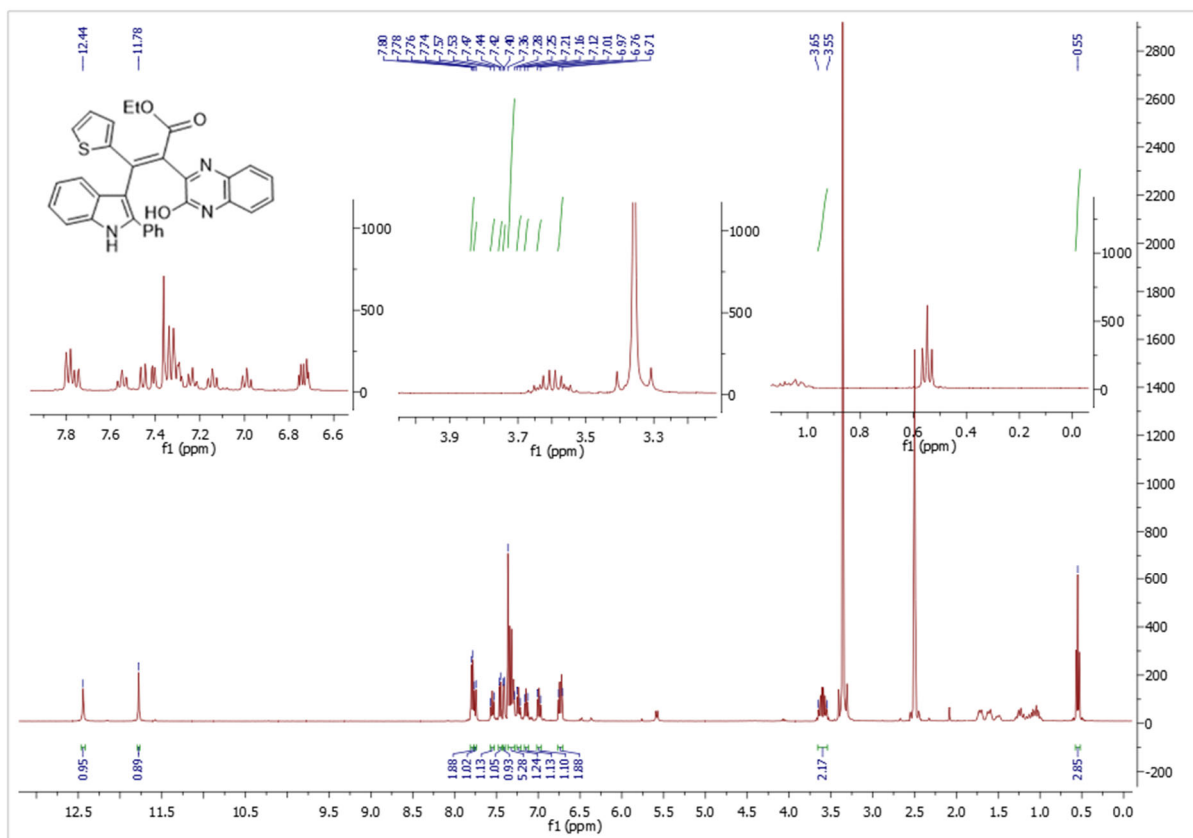
¹³C-NMR spectrum of compound 7h



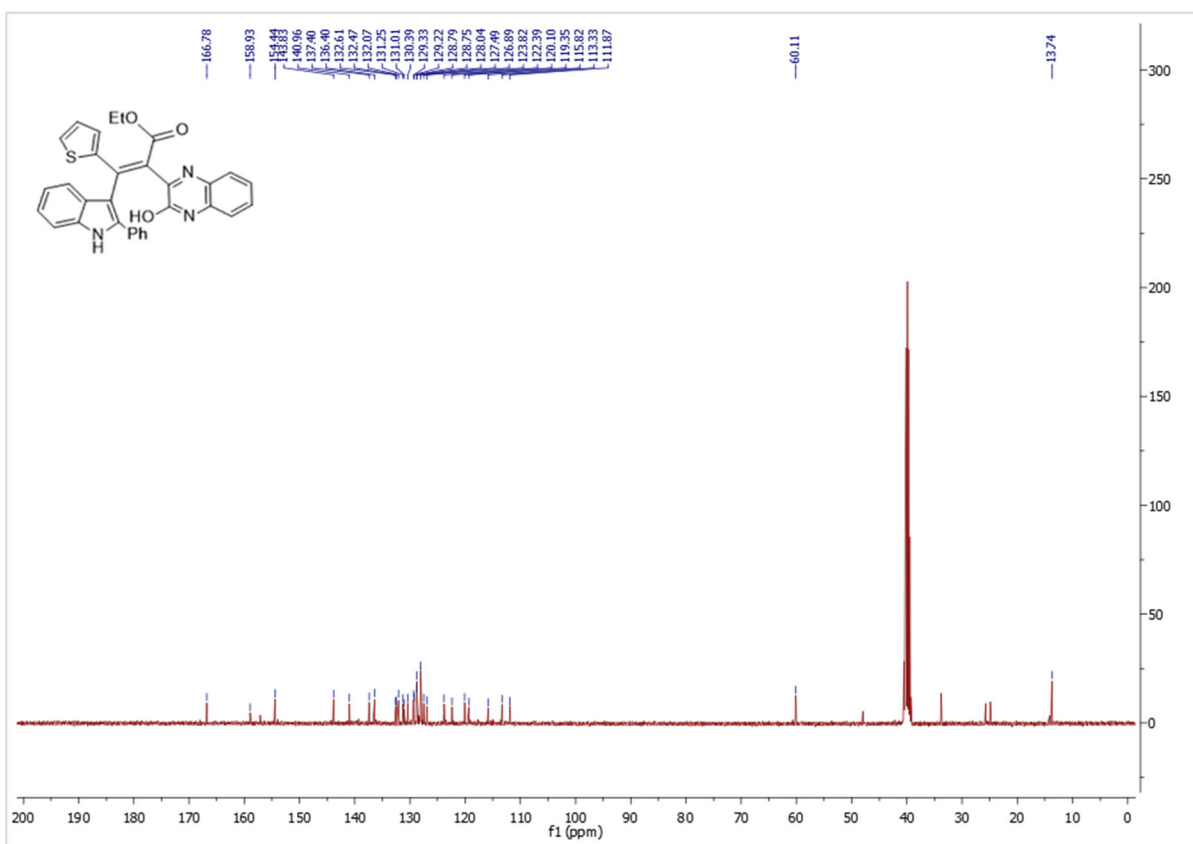
¹H-NMR spectrum of compound **7i**



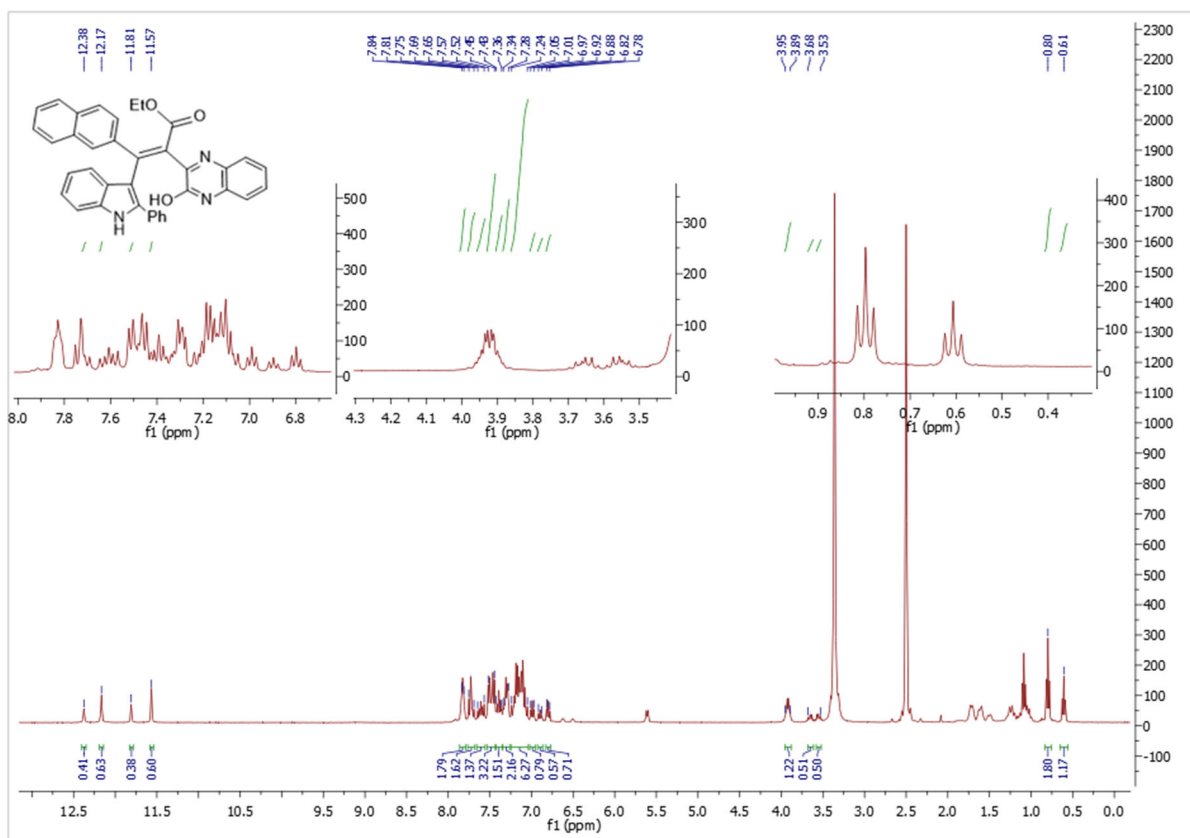
¹³C-NMR spectrum of compound **7i**



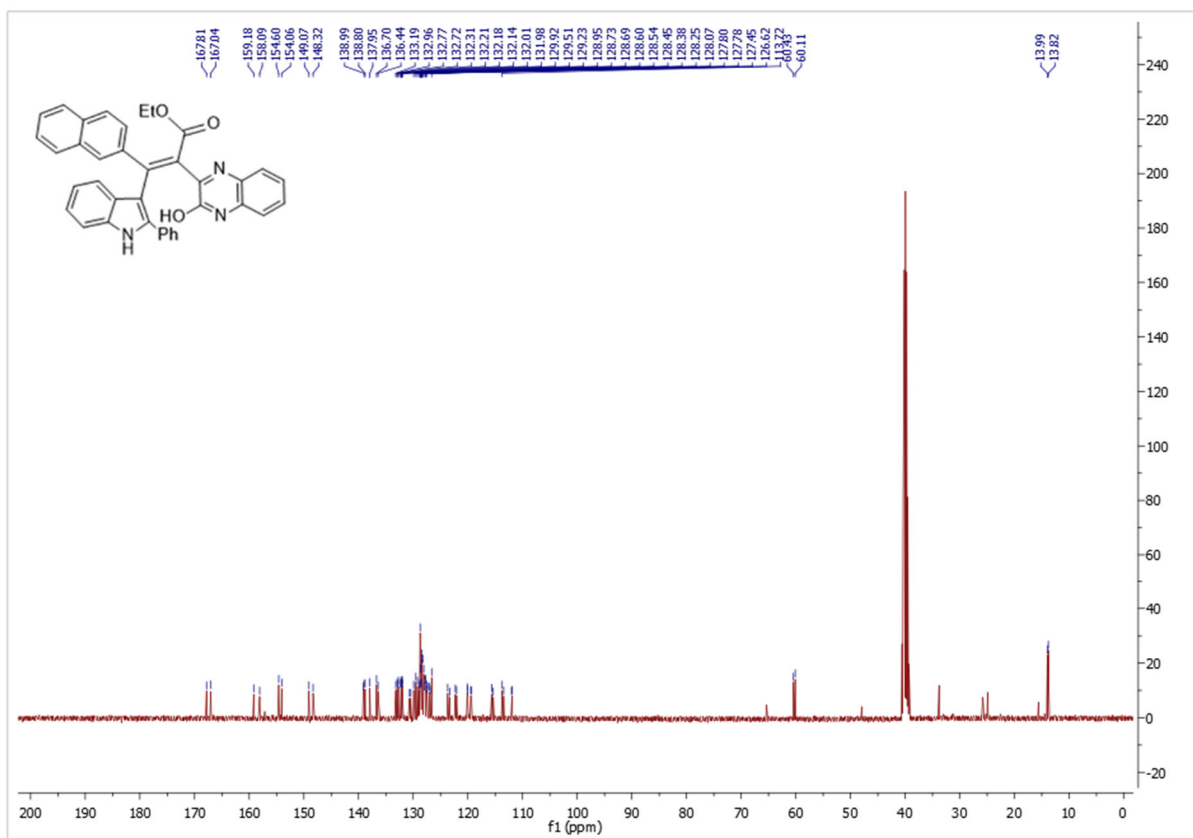
¹H-NMR spectrum of compound **7j**



¹³C-NMR spectrum of compound **7j**

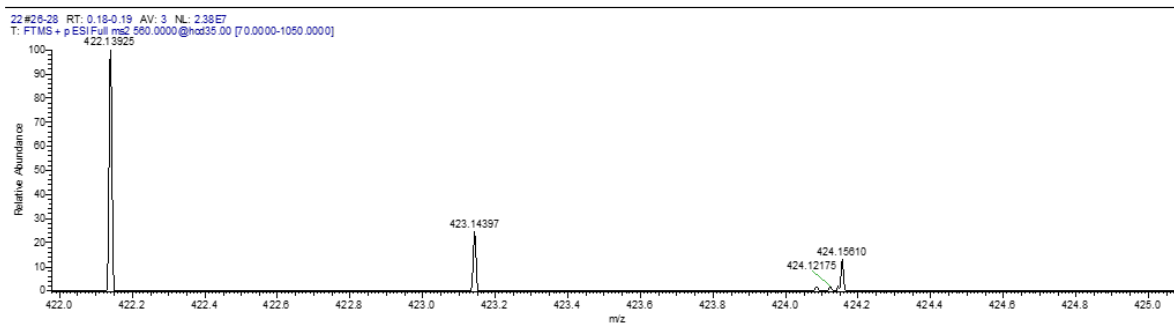


¹H-NMR spectrum of compound 7k

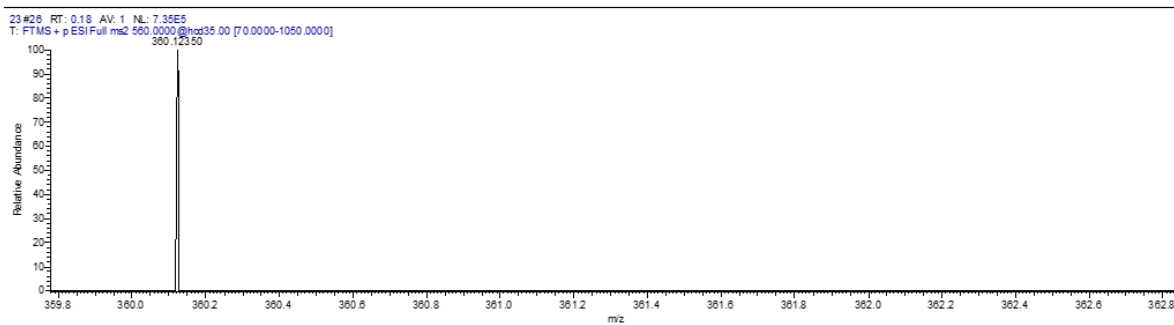


¹³C-NMR spectrum of compound 7k

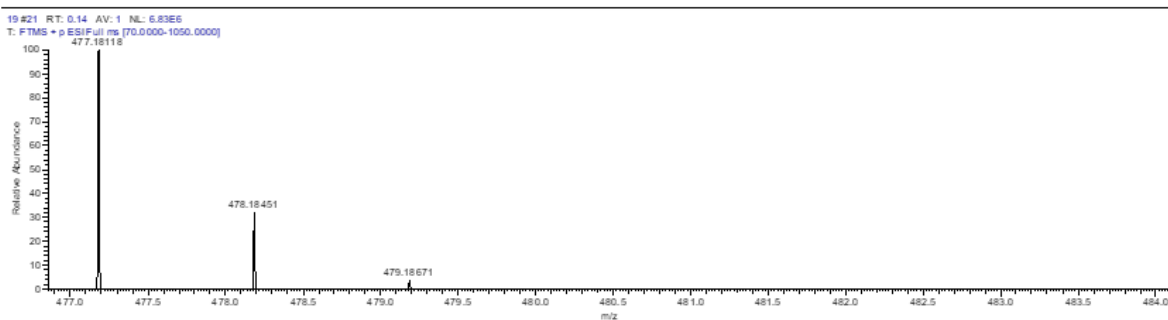
7. Copies of LC/MS spectra



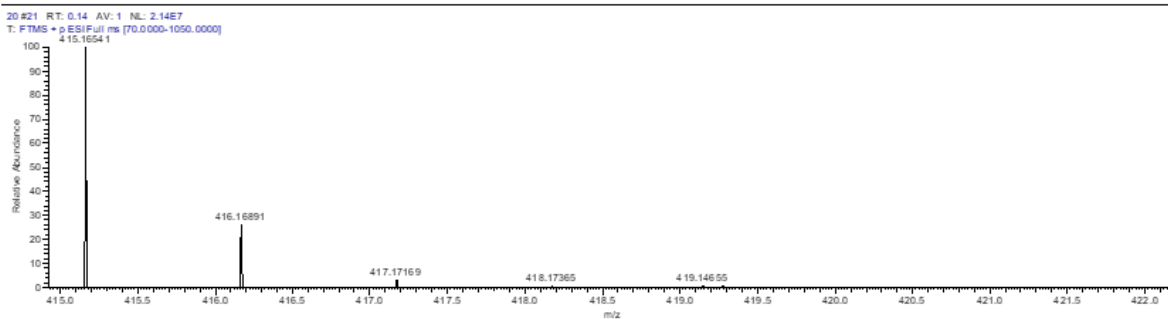
LC/MS Spectrum of compound **4a**



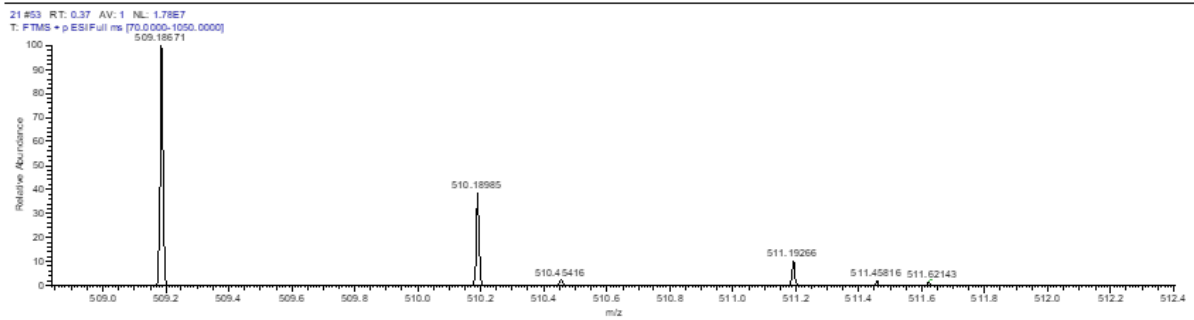
LC/MS Spectrum of compound **4b**



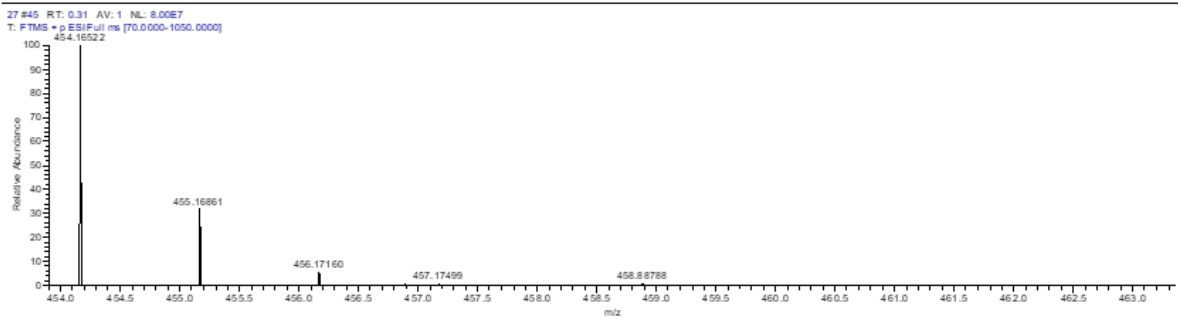
LC/MS Spectrum of compound **5a**



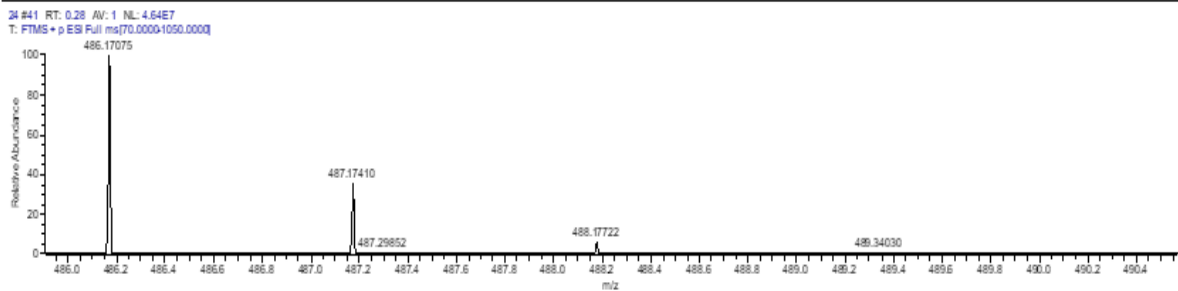
LC/MS Spectrum of compound **5b**



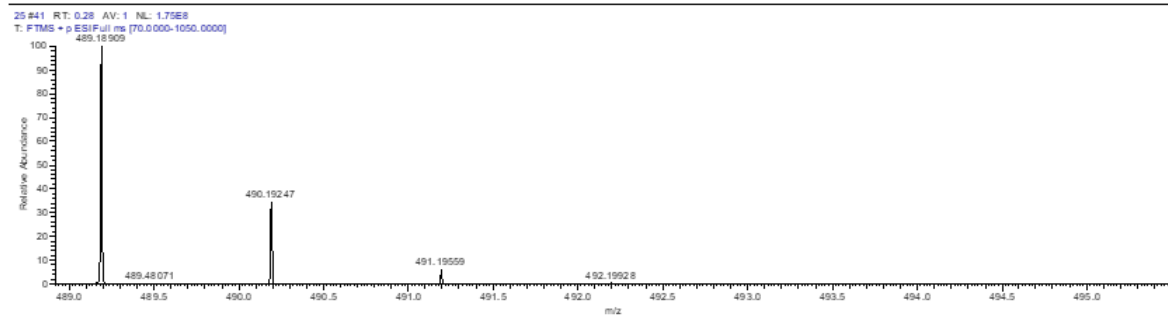
LC/MS Spectrum of compound **5c**



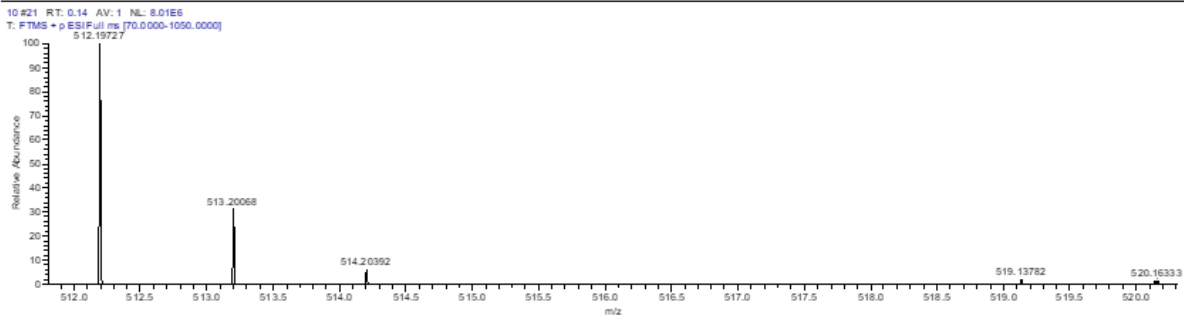
LC/MS Spectrum of compound **5d**



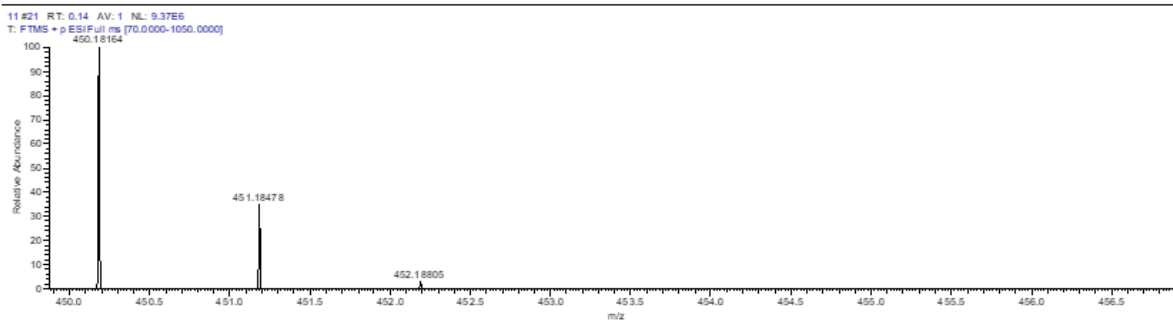
LC/MS Spectrum of compound **5e**



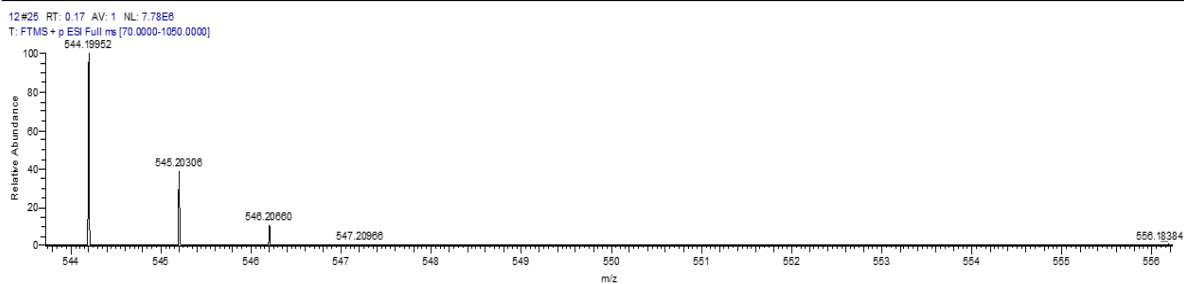
LC/MS Spectrum of compound **5f**



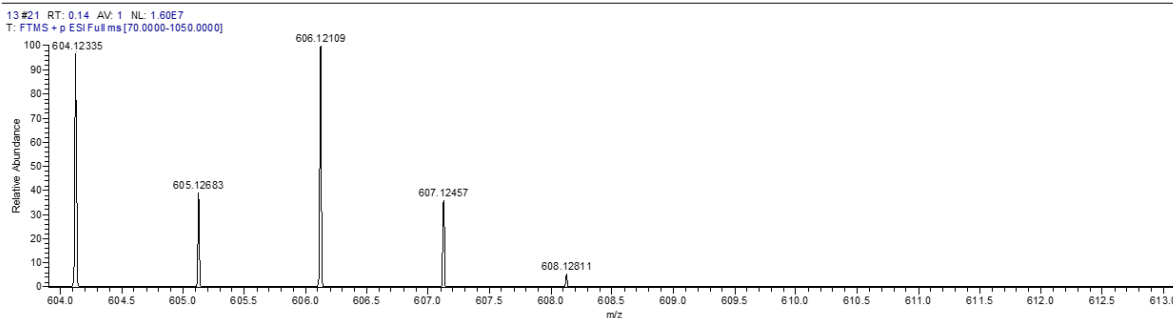
LC/MS Spectrum of compound **7a**



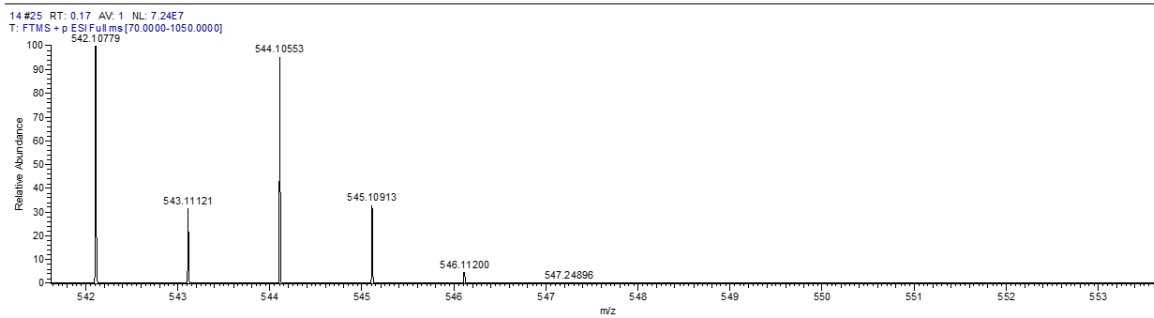
LC/MS Spectrum of compound **7b**



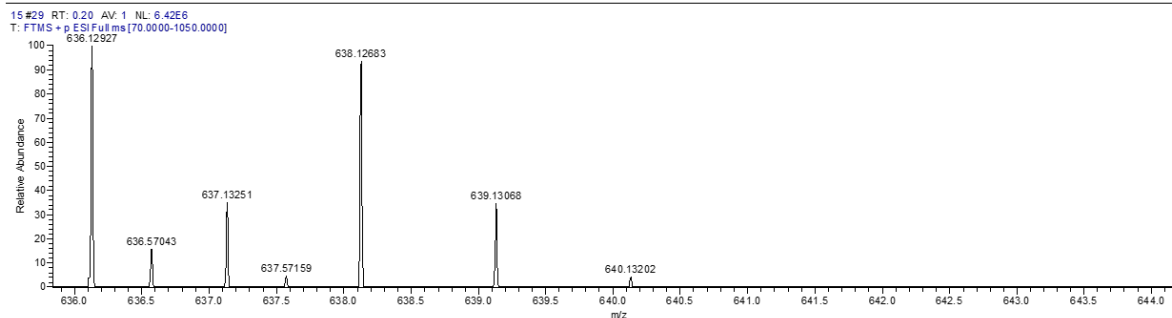
LC/MS Spectrum of compound **7c**



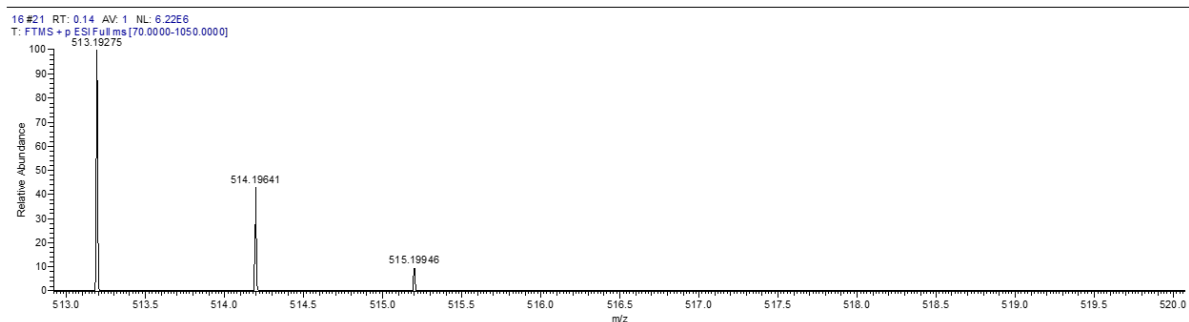
LC/MS Spectrum of compound **7d**



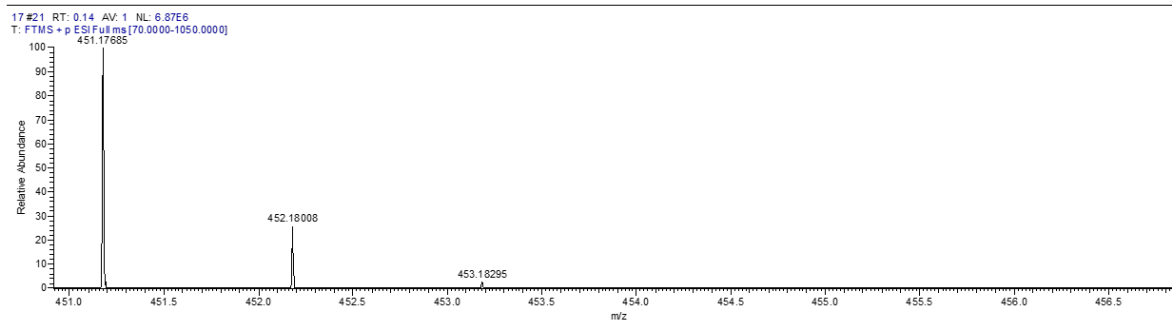
LC/MS Spectrum of compound 7e



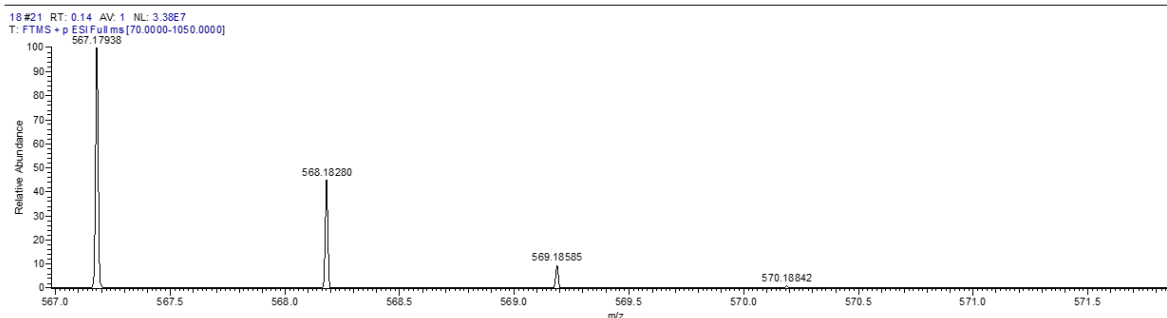
LC/MS Spectrum of compound 7f



LC/MS Spectrum of compound 7g

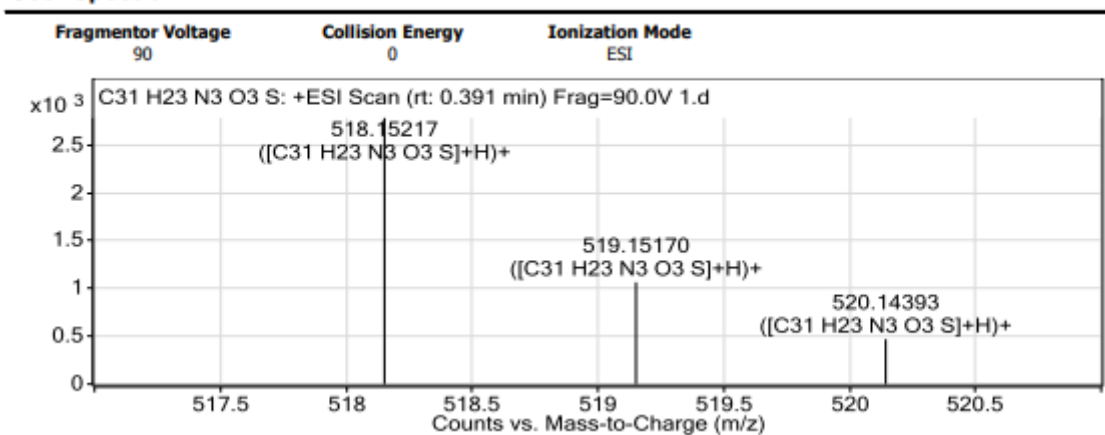


LC/MS Spectrum of compound 7h



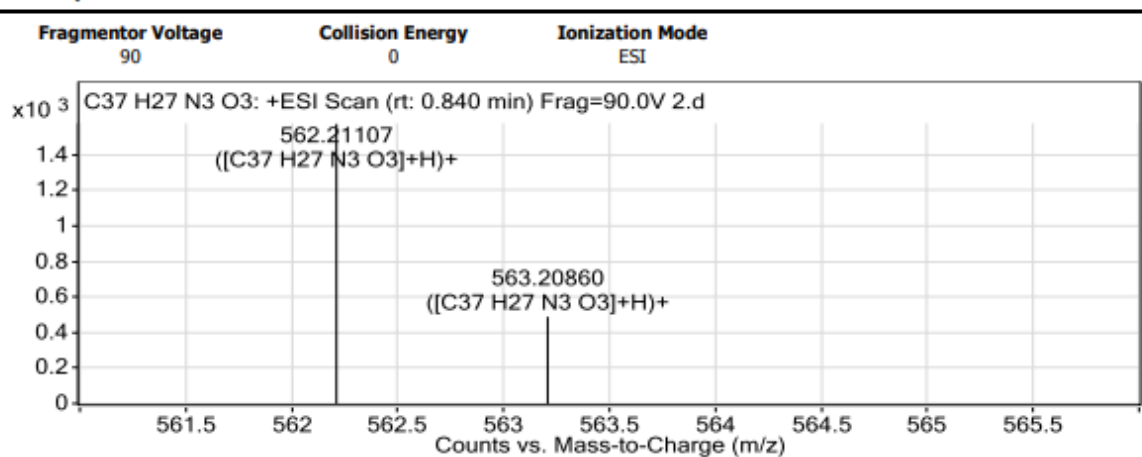
LC/MS Spectrum of compound 7i

User Spectra

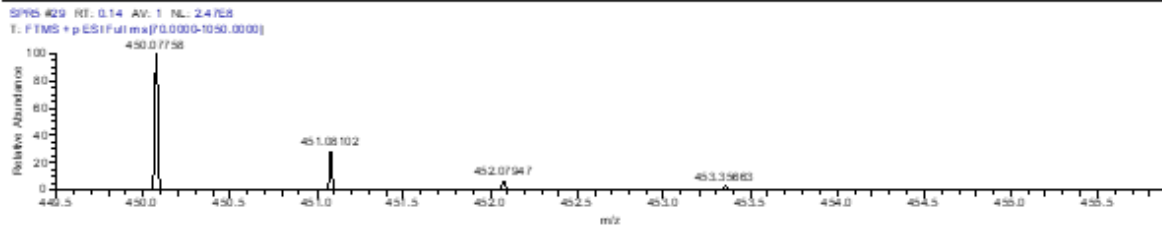


LC/MS Spectrum of compound 7j

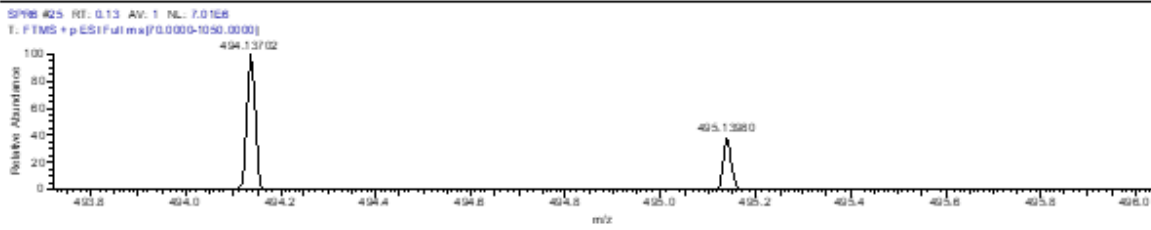
User Spectra



LC/MS Spectrum of compound 7k



LC/MS Spectrum of compound **4d**



LC/MS Spectrum of compound **4e**