

Supporting Information

Chlorophyll-Catalyzed Photochemical Regioselective Coumarin C–H Arylation with Diazonium Salts

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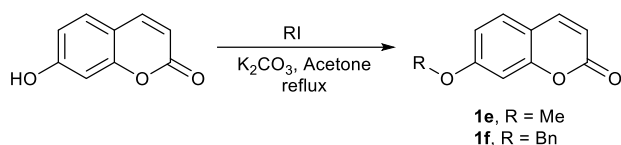
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1 General remarks:

The following includes general experimental procedures, specific details for representative reactions, isolation and spectroscopic information for the compounds. All reagents were commercially available and used as received. Chlorophyll was extracted from the Spinach. Coumarins **1a-1i** and diazonium salt derivatives were synthesized following the procedures described in the literature. The reactions were carried out in a pyrex tube. Column chromatography was carried out on silica gel (230–400 mesh). TLC was conducted on silica gel 250 micron, F254 plates. ¹H NMR spectra were recorded at room temperature on Bruker 400 and 500 MHz spectrometers, using CDCl₃ and DMSO as solvent. Chemical shifts are reported in ppm with TMS as an internal standard. ¹³C NMR spectra are referenced from the solvent central peak (77.23 ppm). Chemical shifts are given in ppm.

2 General experimental details

2-1 Synthesis of 7-alkoxycoumarins¹



A suspension of 7-hydroxycoumarin (1.0 mmol), EtI or MeI (1.5 equiv.) and anhydrous K₂CO₃ (2 equiv.) in anhydrous N,N-dimethylformamide (10 mL) was stirred at rt for 24 h. The solution was diluted with EtOAc (20 mL) and the organic phase was washed with H₂O (3 × 10 mL), dried over anhydrous MgSO₄ and concentrated in vacuo to give 7-alkoxycoumarins **1e** and **1f** as white solids.

2-2 General procedure for the preparation of aryl diazonium tetrafluoroborates²

The appropriate aniline (10 mmol) was dissolved in a mixture of distilled water (4 mL) and 50% hydrofluoroboric acid (3.4 mL). After cooling the reaction mixture to 0°C using ice bath, sodium nitrite (0.69 g in 1.5 mL) was added dropwise in 5 min interval of time. The resulting mixture was stirred for 40 min and the precipitate was collected by filtration and re-dissolved in minimum amount of acetone. Diethyl ether was added until precipitation of diazonium tetrafluoroborate, which is filtered, washed three times with diethyl ether and dried under vacuum.

2.3 General procedure for the extraction of spinach³

2.3.1. Sample Preparation and characterization

Fresh or frozen spinach (0.5 g) was combined with anhydrous magnesium sulfate (0.5 g) and sand (1.0 g). The mixture was ground in a mortar and pestle until a light green powder was obtained (5–10 minutes). The light green solid was transferred to a small test tube containing 2.0 mL of acetone. This heterogeneous mixture was agitated to ensure complete mixing of the acetone and the solid. This mixture was allowed to stand for 10 minutes and green acetone solution was removed by pipette and transferred to a micro centrifuge tube.

After centrifugation, the supernatant (green liquid) is poured in a plate. After evaporation of the solvent, the product is obtained as an oil.

¹(a) A. Sánchez-Recillas, G. Navarrete-Vázquez, S. Hidalgo-Figueroa, M. Y. Rios, M. Ibarra-Barajas and S. Estrada-Soto, *Eur. J. Med. Chem.*, 2014, **77**, 400.

²P. Hanson, J. R. Jones, A. B. Taylor, P. H. Walton and Timms, A. W., *J. Chem. Soc., Perkin Trans. 2.*, 2002, **2**, 1135.

³H. T. Quach and R. L. Steeper and G. W. Griffin, *J. Chem. Educ.*, 2004, **81**, 385.

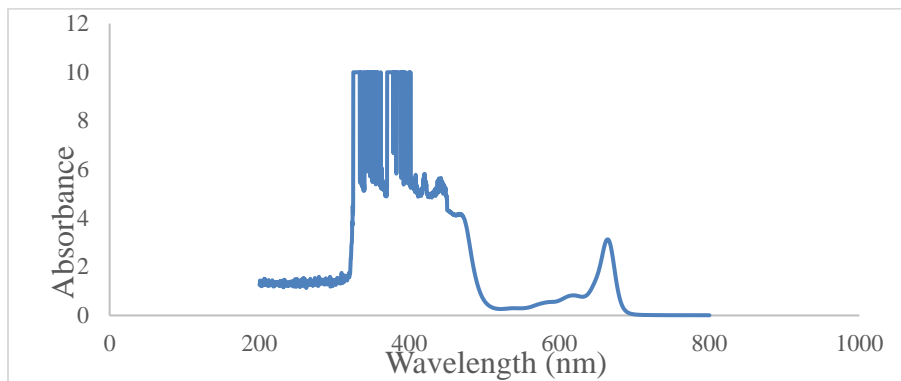


Figure 1. UV Visible Absorption Spectrum of mixture of chlorophyll in Acetone.

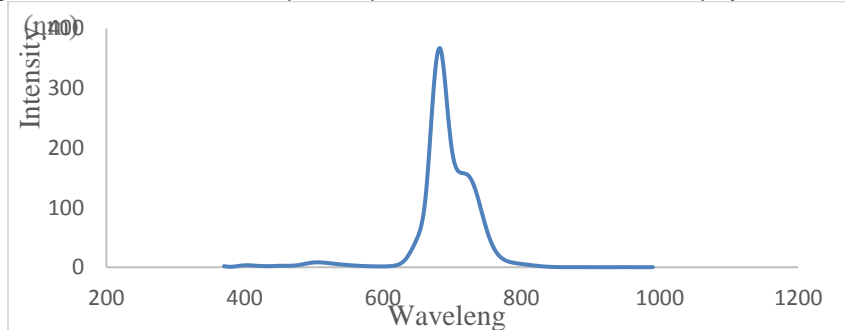


Figure 2. PL Spectrum of mixture of chlorophyll in Acetone.

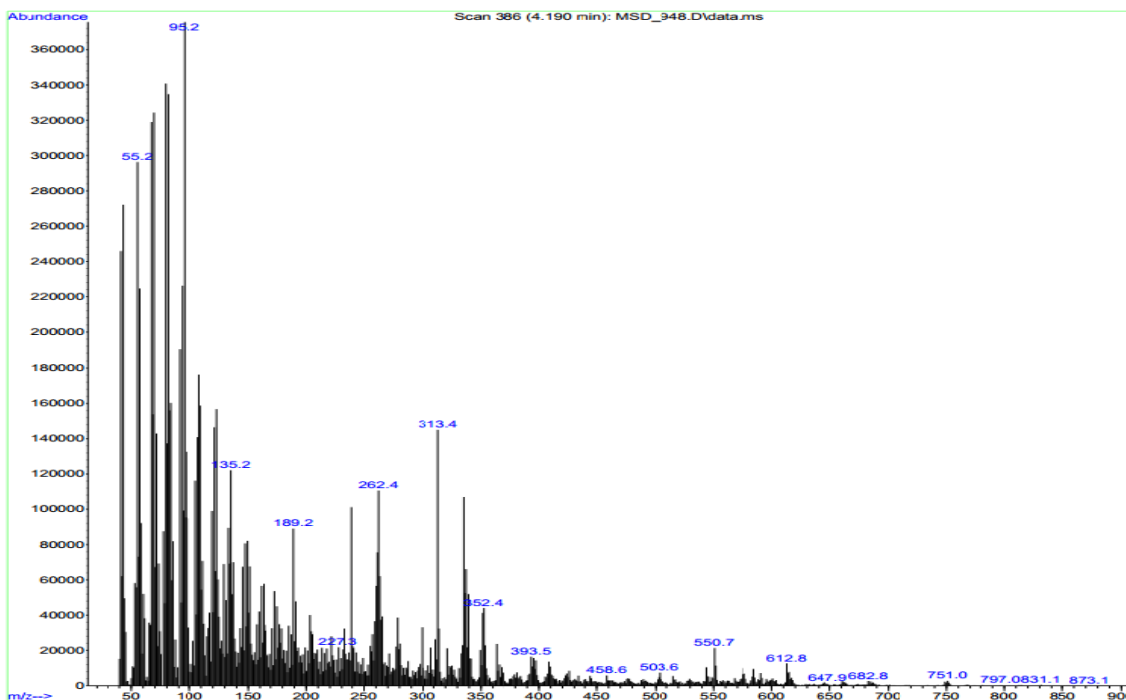
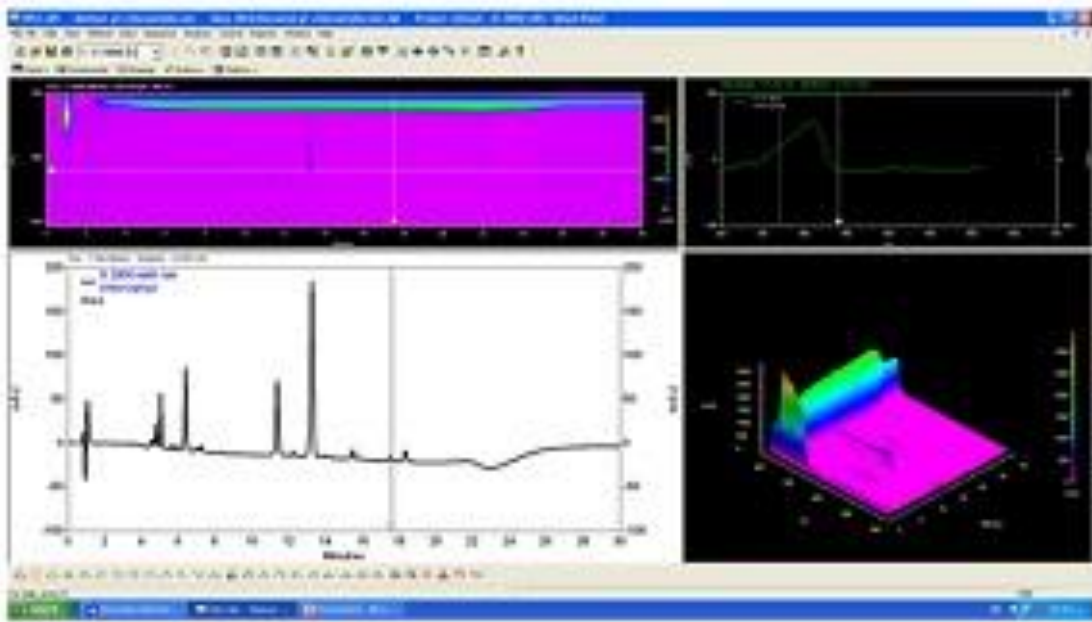


Figure 3. Mass Spectrum of mixture of chlorophyll.



ylhs.mh1.020

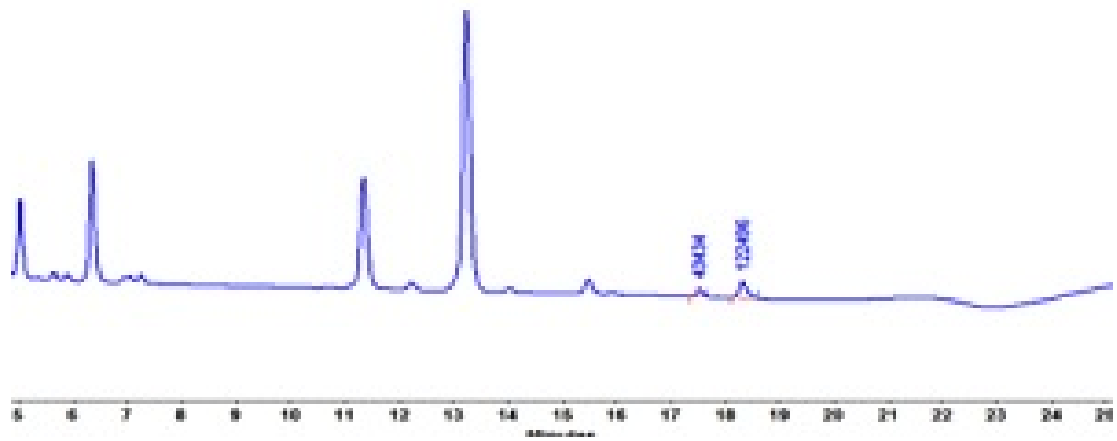


Figure 4. HPLC Spectrum (chromatogram) of mixture of chlorophyll.

2.3.2. TLC Separation

TLC plates (3.5 cm × 9.0 cm) were cut from the commercially available sheets. The acetone extract was transferred in the standard manner and the plates were eluted in a closed chamber with the following mobile phase: 60% petroleum ether (bp 35–60 C), 16% cyclohexane, 10% ethyl acetate, 10% acetone, 4% methanol. The elution order using this elution solvent system was β -carotene ($R_f = 0.95$), chlorophyll a ($R_f = 0.44$), chlorophyll b ($R_f = 0.32$), and xanthophyll ($R_f = 0.16$).

Amount of chlorophyll a = 112 mg

Amount of chlorophyll b = 78 mg

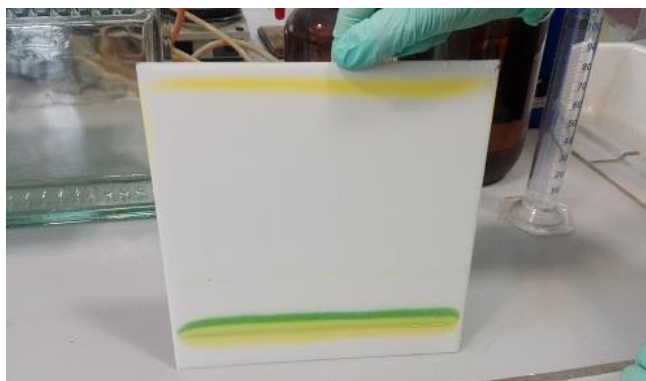


Figure 5. TLC Separation of mixture of chlorophyll.

2.4 General procedure for the reaction of aryl diazonium salts and coumarins

In a 10 mL pyrex tube with magnetic stirring bar the oil of chlorophyll (45 mg), aryl diazonium tetrafluoroborate (0.1 mmol, 1 equiv) and coumarin (4 equiv) were dissolved in DMSO (0.4 mL) and the resulting mixture was stirred.

The pyrex tube was irradiated using 5 W white LED. After 14 h of irradiation the reaction mixture was transferred to separating funnel, diluted with diethyl ether and washed with 15 mL of water. The aqueous layer was washed three times with diethyl ether. The combined organic layers were dried over $MgSO_4$, filtered and concentrated in vacuum.

Purification of the crude product was achieved by flash column chromatography using n-hexane/ethyl acetate as eluent.



Figure 6. Photocatalytic system

3 Emission spectra of White LED

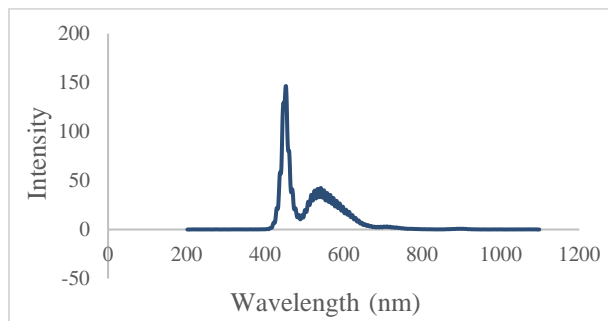
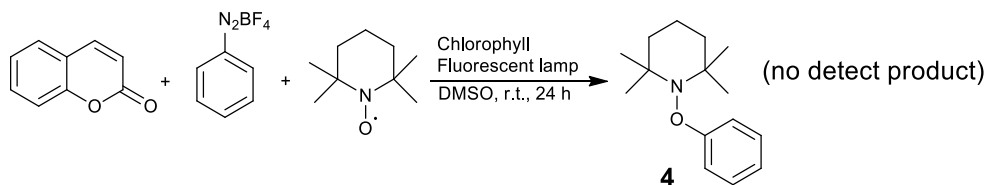


Figure 7. White LED and the emission spectrum of white LED

4 Radical trapping experiment



In a 10 mL pyrex tube with magnetic stirring bar the oil of chlorophyll (45 mg), aryl diazonium tetrafluoroborate (0.1 mmol, 1 equiv) and coumarin (4 equiv) were dissolved in DMSO (0.4 mL) and 2,2,6,6-tetramethylpiperidinyl-1-oxyl (TEMPO) (2.0 equiv) was added. Then the resulting mixture was irradiated using white LED for 14 h. While no desired product **3a** was found, the TEMPO-phenyl adduct **4** was detected.

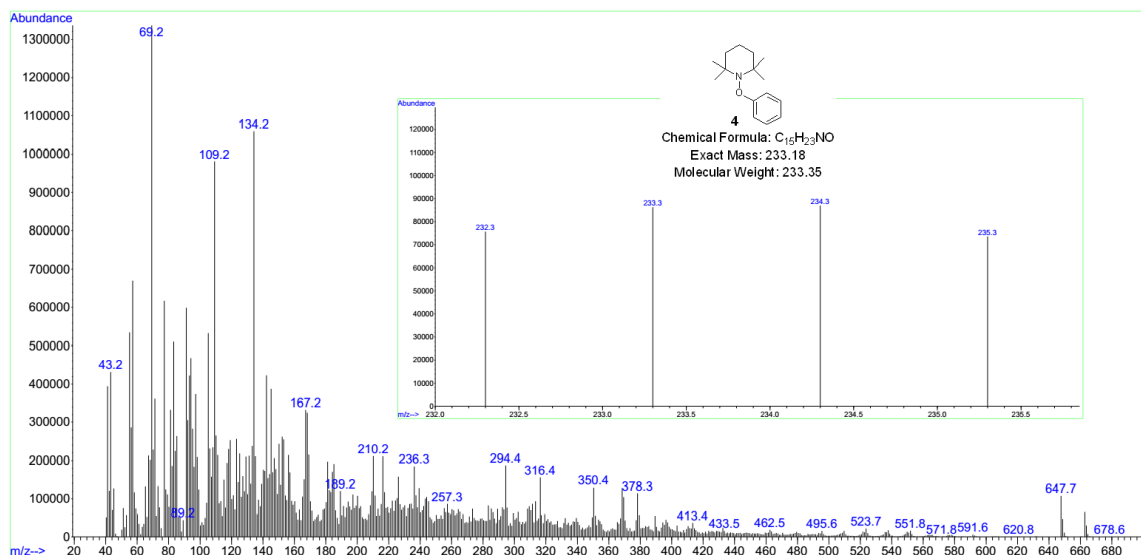


Figure 8. MS of TEMPO-Phenyl adduct

5 Gram scale experiments

In a 50 mL flask with magnetic stirring bar the oil of chlorophyll (4.5 gr), aryl diazonium tetrafluoroborate (10 mmol, 1.90 gr) and coumarin (4 equiv) were dissolved in DMSO (20 mL) and the resulting mixture was stirred.

The flask was irradiated under different conditions. After irradiation the reaction mixture was transferred to separating funnel, diluted with diethyl ether and washed with 50 mL of water. The aqueous layer was washed three times with diethyl ether. The combined organic layers were dried over MgSO_4 , filtered and concentrated in vacuum.

Purification of the crude product was achieved by flash column chromatography using n-hexane/ethyl acetate as eluent.

Condition A: irradiation with 5 W white LED for 14 h, 34% isolated yield.

Condition B: irradiation with 5 W white LED for 24 h, 40% isolated yield.



Figure 9. Gram scale photocatalytic system with 5 W white LED

Condition C: irradiation with 16W Fluorescent lamp for 14 h, 40% isolated yield.

Condition D: irradiation with 16W Fluorescent lamp for 24 h, 46% isolated yield.



Figure 10. Gram scale photocatalytic system with 16W Fluorescent lamp

Condition E: irradiation with 5W Sun light LED for 14 h, 16% isolated yield.



Figure 11. Gram scale photocatalytic system with 5W Sun light LED

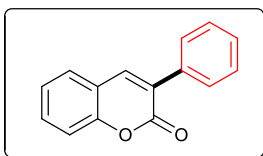
Condition F: irradiation with 5 W white LED for 14 h, dropping flow, 29% isolated yield (use of flow technique).



Figure 12. Gram scale photocatalytic system with 5 W white LED and dropping flow

6 Compounds characterization data

6.1. 3-Phenyl-2H-chromen-2-one (3a)⁴

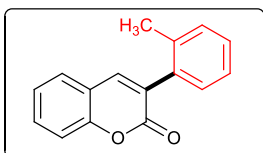


White solid; Yield 62% (14 mg); m.p. 137-139 °C.

¹H NMR (400 MHz, CDCl₃): δ 7.73 (s, 1H), 7.62 (dd, *J* = 7.8, 2.0 Hz, 2H), 7.47 (td, *J* = 8.0, 1.2 Hz, 2H), 7.32-7.39 (m, 3H), 7.28 (d, *J* = 8.0 Hz, 1H), 7.21 (td, *J* = 7.8, 0.8 Hz, 1H).

¹³C NMR (100 MHz, CDCl₃): δ 159.6, 152.4, 138.9, 133.6, 130.4, 127.8, 127.5, 127.4, 126.9, 123.5, 118.6, 115.4.

6.2. 3-(*o*-Tolyl)-2H-chromen-2-one (3b)⁵

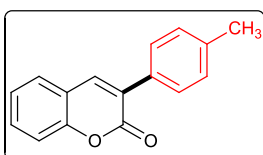


White solid; Yield 65% (15 mg); m.p. 142-144 °C.

¹H NMR (500 MHz, CDCl₃): δ 7.65 (s, 1H), 7.57-7.51 (m, 2H), 7.40 (d, *J* = 5.0 Hz, 1H), 7.34-7.26 (m, 5H), 2.31 (s, 3H).

¹³C NMR (125 MHz, CDCl₃): δ 160.3, 153.8, 141.6, 136.9, 134.7, 131.5, 130.4, 129.8, 128.9, 128.4, 127.9, 125.9, 124.5, 119.3, 116.6, 20.0.

6.3. 3-(*p*-Tolyl)-2H-chromen-2-one (3c)⁴

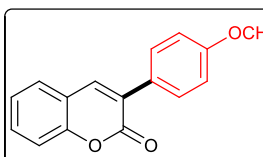


White solid; Yield 70% (17 mg); m.p. 149-151 °C.

¹H NMR (400 MHz, CDCl₃): δ 7.72 (s, 1H), 7.54 (d, *J* = 8.0 Hz, 2H), 7.47-7.42 (m, 2H), 7.29 (d, *J* = 8.0 Hz, 1H), 7.24-7.18 (m, 3H), 2.33 (s, 3H).

¹³C NMR (100 MHz, CDCl₃): δ 159.7, 152.4, 138.2, 137.9, 130.8, 130.2, 128.5, 128.2, 127.4, 126.8, 123.4, 118.7, 115.4, 20.3.

6.4. 3-(4-Methoxyphenyl)-2H-chromen-2-one (3d).⁴

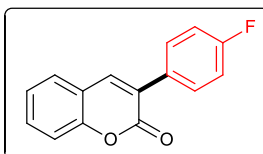


Colorless solid; Yield 81% (20 mg); m.p. 139-141 °C.

¹H NMR (500 MHz, CDCl₃): δ 7.77 (s, 1H), 7.68 (d, *J* = 8.8 Hz, 2H), 7.54-7.49 (m, 2H), 7.36 (dd, *J* = 8.3, 1.0 Hz, 1H), 7.29 (td, *J* = 7.5, 1.1 Hz, 1H), 6.68 (d, *J* = 8.8 Hz, 2H), 3.86 (s, 3H).

¹³C NMR (125 MHz, CDCl₃): δ 160.7, 160.2, 153.3, 138.4, 130.9, 129.8, 127.9, 127.6, 127.1, 124.4, 119.8, 116.4, 113.9, 55.4.

6.5. 3-(4-Fluorophenyl)-2H-chromen-2-one (3e).⁴



White solid; Yield 73% (18 mg); m.p. 159-161 °C.

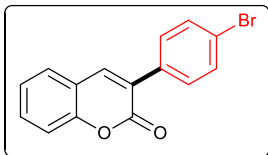
¹H NMR (500 MHz, CDCl₃): δ 7.80 (s, 1H), 7.72-7.69 (m, 2H), 7.56-7.52 (m, 2H), 7.38-7.36 (m, 1H), 7.31 (tt, *J* = 7.5, 1.0 Hz, 1H), 7.14 (td, *J* = 8.8, 1.0 Hz, 2H).

¹³C NMR (125 MHz, CDCl₃): δ 163.1 (d, ¹*J*_{C-F} = 247.5 Hz), 160.5, 153.5, 139.6, 131.5, 130.7, 130.7 (d, ⁴*J*_{C-F} = 3.7 Hz), 130.4 (d, ³*J*_{C-F} = 7.5 Hz), 127.9, 127.3, 124.6, 119.6, 116.5, 115.6 (d, ²*J*_{C-F} = 21.2 Hz).

⁴M. Golshani, M. Khoobi, N. Jalalimanesh, F. Jafarpour and A. Ariaifard, *Chem. Commun.*, 2017, **53**, 10676.

⁵F. Jafarpour, H. Hazrati, N. Mohasselyazdi, M. Khoobi and A. Shafiee, *Chem. Commun.*, 2013, **49**, 10935.

6.6. 3-(4-Bromophenyl)-2H-chromen-2-one (3f)⁴

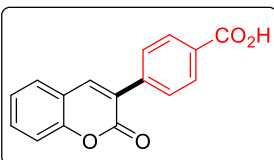


Colorless solid; Yield 69% (21 mg); m.p. 189-191°C.

¹H NMR (500 MHz, CDCl₃): δ 7.82 (s, 1H), 7.61-7.53 (m, 6H), 7.37 (d, *J* = 8.6 Hz, 1H), 7.31 (t, *J* = 8.0 Hz, 1H).

¹³C NMR (125 MHz, CDCl₃): δ 160.3, 153.5, 139.9, 133.5, 131.7, 131.6, 130.1, 128.0, 127.2, 124.6, 123.2, 119.5, 116.5.

6.7. 4-(2-Oxo-2H-chromen-3-yl)benzoic acid (3g)⁶

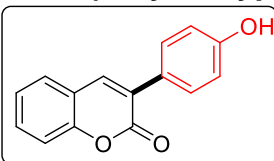


White solid; Yield 52% (14 mg); m.p. 289-290°C.

¹H NMR (500 MHz, DMSO-d₆): δ 8.39 (s, 1H), 8.03 (d, *J* = 8.2 Hz, 2H), 7.88 (d, *J* = 8.2 Hz, 2H), 7.82 (d, *J* = 7.7 Hz, 1H), 7.66 (t, *J* = 7.8 Hz, 1H), 7.47 (d, *J* = 8.3 Hz, 1H), 7.41 (t, *J* = 7.5 Hz, 1H).

¹³C NMR (125 MHz, DMSO-d₆): δ 167.4, 159.9, 153.6, 142.1, 139.3, 132.6, 130.4, 129.61, 129.3, 129.1, 126.3, 125.1, 119.8, 116.4.

6.8. 3-(4-Hydroxyphenyl)-2H-chromen-2-one (3h)⁷

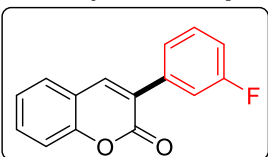


White solid; Yield 79% (19 mg); m.p. 201-203°C.

¹H NMR (500 MHz, CDCl₃): δ 7.79 (s, 1H), 7.64 (d, *J* = 8.5 Hz, 2H), 7.58 – 7.48 (m, 2H), 7.38 (d, *J* = 8.2 Hz, 1H), 7.34 – 7.21 (m, 4H), 6.93 (d, *J* = 8.4 Hz, 3H).

¹³C NMR (125 MHz, CDCl₃): δ 159.9, 156.7, 150.1, 138.5, 132.8, 132.5, 131.04, 130.0, 127.7, 126.8, 124.4, 116.4, 115.4, 109.9.

6.9. 3-(3-Fluorophenyl)-2H-chromen-2-one (3i)⁸

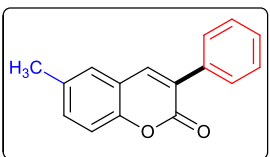


White solid; Yield 67% (16 mg); m.p. 152-153°C.

¹H NMR (500 MHz, CDCl₃): δ 7.86 (s, 1H), 7.57 (td, *J* = 7.2, 1.6 Hz, 2H), 7.54 – 7.47 (m, 2H), 7.46 – 7.37 (m, 2H), 7.34 (td, *J* = 7.5, 1.1 Hz, 1H), 7.13 (td, *J* = 8.4, 2.6 Hz, 1H).

¹³C NMR (125 MHz, CDCl₃): δ 163.6 (d, ¹*J*_{C-F} = 240 Hz), 160.2, 153.6, 140.40, 136.7 (d, ³*J*_{C-F} = 8.28 Hz), 131.8, 130.0 (d, ³*J*_{C-F} = 8.37 Hz), 128.0, 127.0, 124.6, 124.1 (d, ⁴*J*_{C-F} = 2.86 Hz), 119.4, 116.5, 115.8 (d, ²*J*_{C-F} = 21.04 Hz), 115.7 (d, ²*J*_{C-F} = 22.85 Hz).

6.11. 6-Methyl-3-phenyl-2H-chromen-2-one (3j)⁵

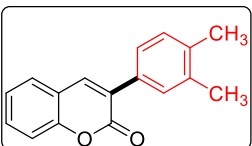


Light brown solid; Yield 63% (15 mg); m.p. 142-144 °C.

¹H NMR (400 MHz, CDCl₃): δ 7.66 (s, 1H), 7.61 (d, *J* = 7.3 Hz, 2H), 7.37-7.29 (m, 3H), 7.24 (d, *J* = 7.8 Hz, 2H), 7.16 (d, *J* = 8.4 Hz, 1H), 2.33 (s, 3H).

¹³C NMR (100 MHz, CDCl₃): δ 160.8, 151.7, 139.9, 134.9, 134.2, 132.5, 128.8, 128.6, 128.5, 128.2, 127.7, 119.4, 116.2, 20.8.

6.10. 3-(3,4-Dimethylphenyl)-2H-chromen-2-one (3k)⁹



White solid; Yield 75% (18 mg); m.p. 110-112°C.

¹H NMR (500 MHz, CDCl₃): δ 7.80 (s, 1H), 7.58 – 7.50 (m, 3H), 7.47 (dd, *J* = 7.8, 1.9 Hz, 1H), 7.38 (d, *J* = 8.2 Hz, 1H), 7.31 (td, *J* = 7.6, 1.1 Hz, 1H), 7.24 (d, *J* = 7.8 Hz, 1H), 2.35 (s, 3H), 2.33 (s, 3H).

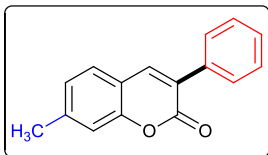
⁶ H. Meerwein, E. Büchner and K. V. Emster, *J. Prakt. Chem.*, 1939, **2**, 237.

⁷ Y. Hu, B. Wang, J. Yang, T. Liu, J. Sun and X. Wang, *J. Enzyme Inhib. Med. Chem.*, 2019, **34**, 15.

⁸ P. Chauhan, M. Ravi, S. Singh, P. Prajapati and P. P. Yadav, *RSC Adv.*, 2016, **6**, 109.

¹³C NMR (125 MHz, CDCl₃): δ 160.7, 153.4, 139.1, 137.6, 136.69, 132.2, 131.1, 129.7, 129.6, 128.5, 127.7, 125.9, 124.4, 119.8, 116.4, 19.9, 19.6.

6.12. 7-Methyl-3-phenyl-2H-chromen-2-one (3l)⁹

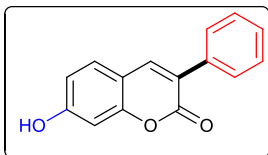


Colorless solid; Yield 65% (15 mg); m.p. 175-177 °C.

¹H NMR (400 MHz, CDCl₃): δ 7.68 (s, 1H), 7.60 (dd, *J* = 8.2, 1.2 Hz, 2H), 7.41-7.27 (m, 4H), 7.07 (s, 1H), 7.01 (dd, *J* = 7.7, 1.4 Hz, 1H), 2.37 (s, 3H).

¹³C NMR (100 MHz, CDCl₃): δ 160.8, 153.6, 142.7, 139.9, 134.9, 128.7, 128.5, 128.5, 127.6, 127.1, 125.7, 117.3, 116.6, 21.9.

6.13. 7-Hydroxy-3-phenyl-2H-chromen-2-one (3m)⁴

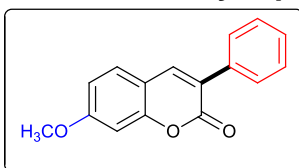


Light yellow solid; Yield 82% (20 mg); m.p. 186-189 °C.

¹H NMR (500 MHz, DMSO-*d*₆): δ 10.61 (s, 1H), 8.11 (s, 1H), 7.68 (d, *J* = 7.7 Hz, 2H), 7.58 (dd, *J* = 8.6, 3.2 Hz, 1H), 7.48-7.31 (m, 3H), 6.82 (d, *J* = 8.6 Hz, 1H), 6.76 (s, 1H).

¹³C NMR (125 MHz, DMSO-*d*₆): δ 161.7, 160.5, 155.4, 141.5, 135.6, 130.4, 128.7, 128.6, 128.4, 122.7, 113.8, 112.5, 102.2.

6.14. 7-Methoxy-3-phenyl-2H-chromen-2-one (3n)⁴

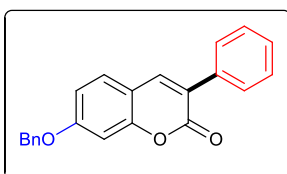


Yellow solid; Yield 83% (21 mg); m.p. 109-111 °C.

¹H NMR (400 MHz CDCl₃): δ 7.67 (s, 1H), 7.60 (d, *J* = 8 Hz, 2H), 7.36-7.27 (m, 4H), 6.79-6.75 (m, 2H), 3.79 (s, 3H).

¹³C NMR (100 MHz, CDCl₃): δ 161.6, 159.8, 154.3, 139.0, 133.9, 127.8, 127.4, 127.4, 127.3, 123.7, 112.3, 111.7, 99.4, 54.8.

6.15. 7-(Benzyloxy)-3-phenyl-2H-chromen-2-one (3o)⁴

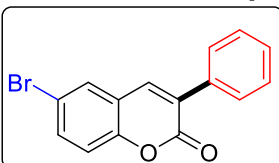


Light Yellow solid; Yield 83% (28 mg); m.p. 136-139 °C.

¹H NMR (400 MHz, CDCl₃): δ 7.67 (s, 1H), 7.60 (dd, *J* = 8.6, 1.6 Hz, 2H), 7.36-7.27 (m, 9H), 6.87-6.8 (m, 2H), 5.06 (s, 2H).

¹³C NMR (100 MHz, CDCl₃): δ 160.6, 159.8, 154.2, 138.9, 134.8, 133.9, 127.9, 127.8, 127.7, 127.6, 127.4, 127.4, 127.4, 126.5, 123.9, 112.5, 112.4, 100.5, 69.5.

6.17. 6-Bromo-3-phenyl-2H-chromen-2-one (3p)¹⁰



White solid; Yield 38% (11 mg); m.p. 171-173°C.

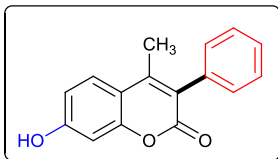
¹H NMR (500 MHz, CDCl₃): δ 7.74 (s, 1H), 7.72 – 7.69 (m, 3H), 7.63 (dd, *J* = 8.8, 2.3 Hz, 1H), 7.70 – 7.43 (m, 3H), 7.27 (d, *J* = 9.0 Hz, 1H).

¹³C NMR (125 MHz, CDCl₃): δ 159.8, 152.3, 138.2, 134.2, 134.0, 130.1, 129.5, 129.2, 128.5, 128.5, 121.2, 118.1, 117.0.

⁹W. Shen, J. Mao, J. Sun, M. Sun and C. Zhang, *Med. Chem. Res.*, 2013, **22**, 1630.

¹⁰H. J. Yoo and S. W. Youn, *Org. Lett.* 2019, **21**, 3422.

6.16. 7-Hydroxy-4-methyl-3-phenyl-2H-chromen-2-one (3q)¹¹

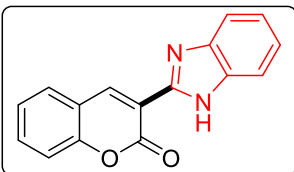


White solid; Yield 89% (25 mg); m.p. 230–232 °C.

¹H NMR (500 MHz, DMSO-*d*₆): δ 10.51 (s, 1H), 7.61 (dd, *J* = 8.6, 3.3 Hz, 1H), 7.43-7.26 (m, 5H), 6.81 (d, *J* = 8.6 Hz, 1H), 6.73 (s, 1 H), 2.16 (s, 3H).

¹³C NMR (125 MHz, DMSO-*d*₆): δ 161.3, 160.7, 154.3, 148.6, 135.4, 130.7, 128.5, 128.0, 127.5, 122.8, 113.4, 112.8, 102.4, 16.7.

6.19. 3-(1H-Benzo[d]imidazol-2-yl)-2H-chromen-2-one (3s)¹²

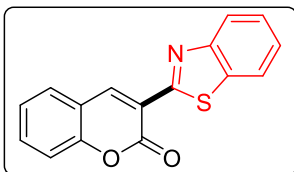


Colorless solid; Yield 30% (8 mg); m.p. 230-231°C.

¹H NMR (500 MHz, CDCl₃, 1 drop DMSO-*d*₆): δ 12.62 (s, 1H), 7.77 (dd, *J* = 7.9, 1.6 Hz, 1H), 7.38-7.40 (m, 3H), 7.15 – 7.04 (m, 1H), 7.06 – 6.98 (m, 3H), 6.83 (dd, *J* = 8.3, 1.1 Hz, 1H), 6.76 – 6.64 (td, *J* = 8.3, 1.1 Hz 1H).

¹³C NMR (125 MHz, CDCl₃, DMSO-*d*₆): δ 158.5, 151.7, 136.7, 131.4, 125.9, 125.8, 122.6, 122.66, 118.7, 117.2, 114.5, 114.5, 114.4, 114.4, 112.6, 77.7, 77.4, 77.2, 40.2, 40.0, 39.9, 39.75, 39.5.

6.20. 3-(Benzo[d]thiazol-2-yl)-2H-chromen-2-one (3t)¹³



Light green solid; Yield 36% (10 mg); m.p. 213-214°C.

¹H NMR (500 MHz, CDCl₃): δ 9.12 (s, 1H), 8.10 (d, *J* = 8.1 Hz, 1H), 7.98 (d, *J* = 8.0 Hz, 1H), 7.73 (dd, *J* = 7.8, 1.5 Hz, 1H), 7.66 (t, *J* = 10, Hz, 1H), 7.56 (t, *J* = 10, Hz, 1H), 7.44 (m, 2H), 7.38 (t, *J* = 7.5 Hz, 1H).

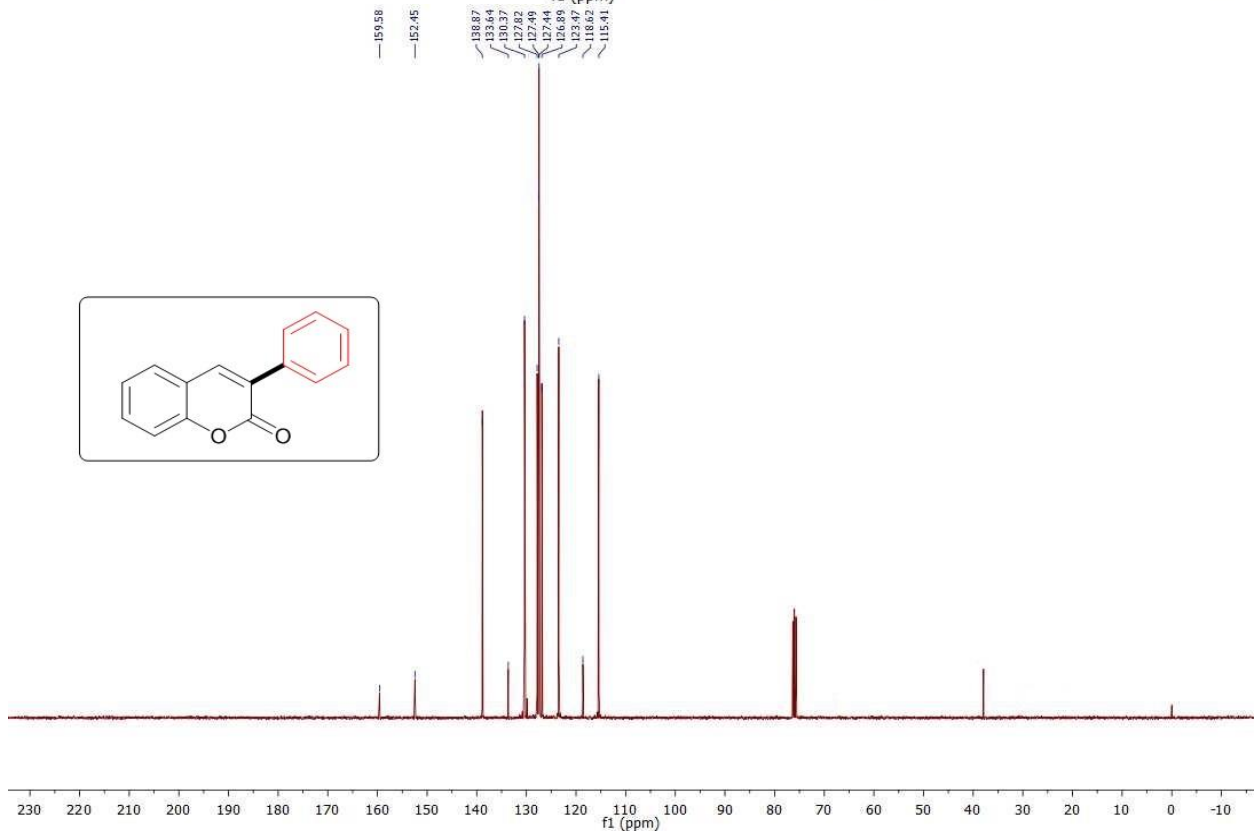
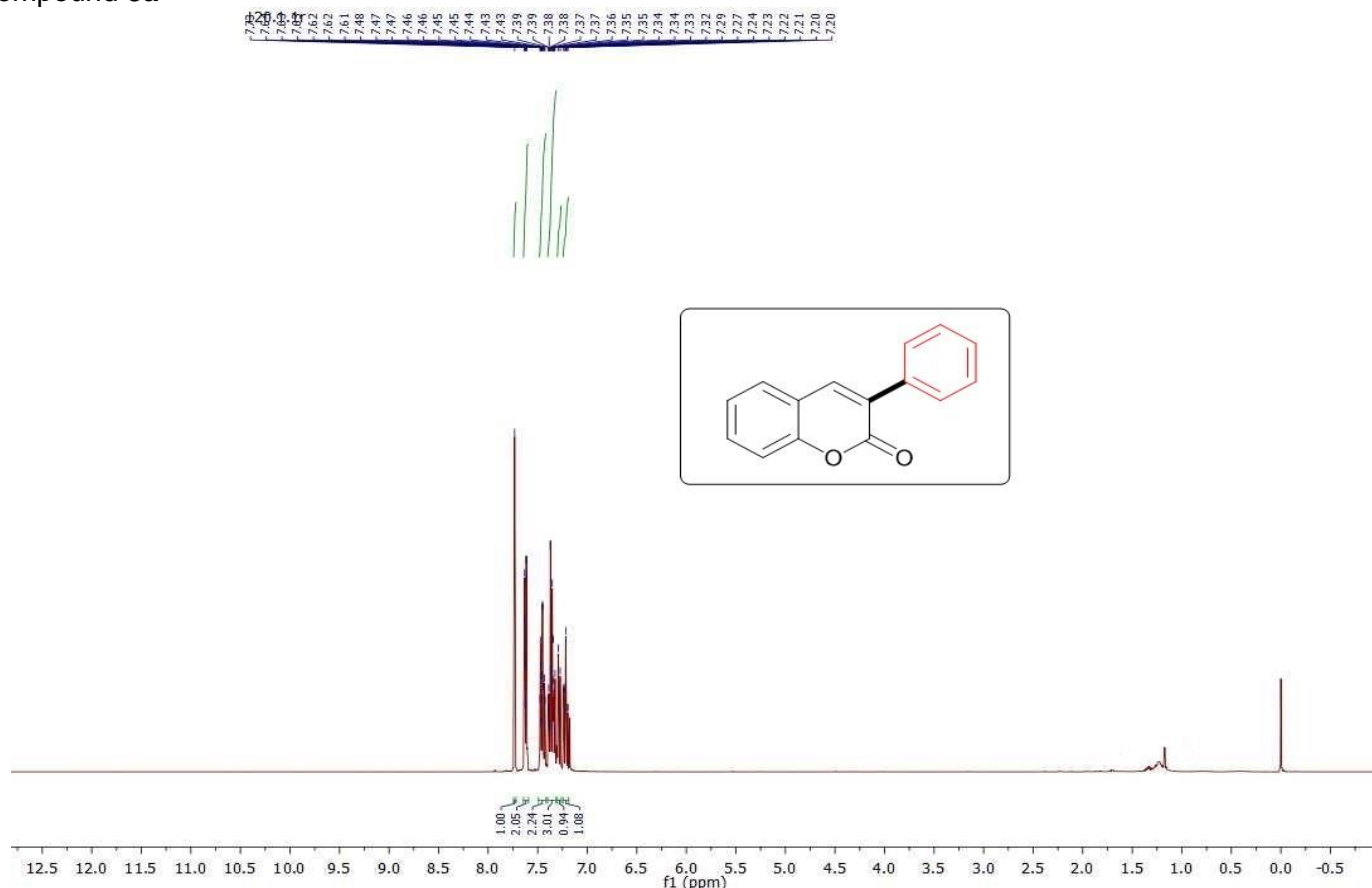
¹³C NMR (125 MHz, CDCl₃): δ 159.9, 159.8, 153.8, 152.0, 141.7, 136.6, 133.3, 129.4, 126.6, 125.5, 125.2, 122.7, 121.7, 120.0, 118.9, 116.7.

¹¹J. W. Yuan, L. R. Yang, Q.-Y. Yin, P. Mao and L.B. Qu, *RSC Adv.*, 2016, **6**, 35936.

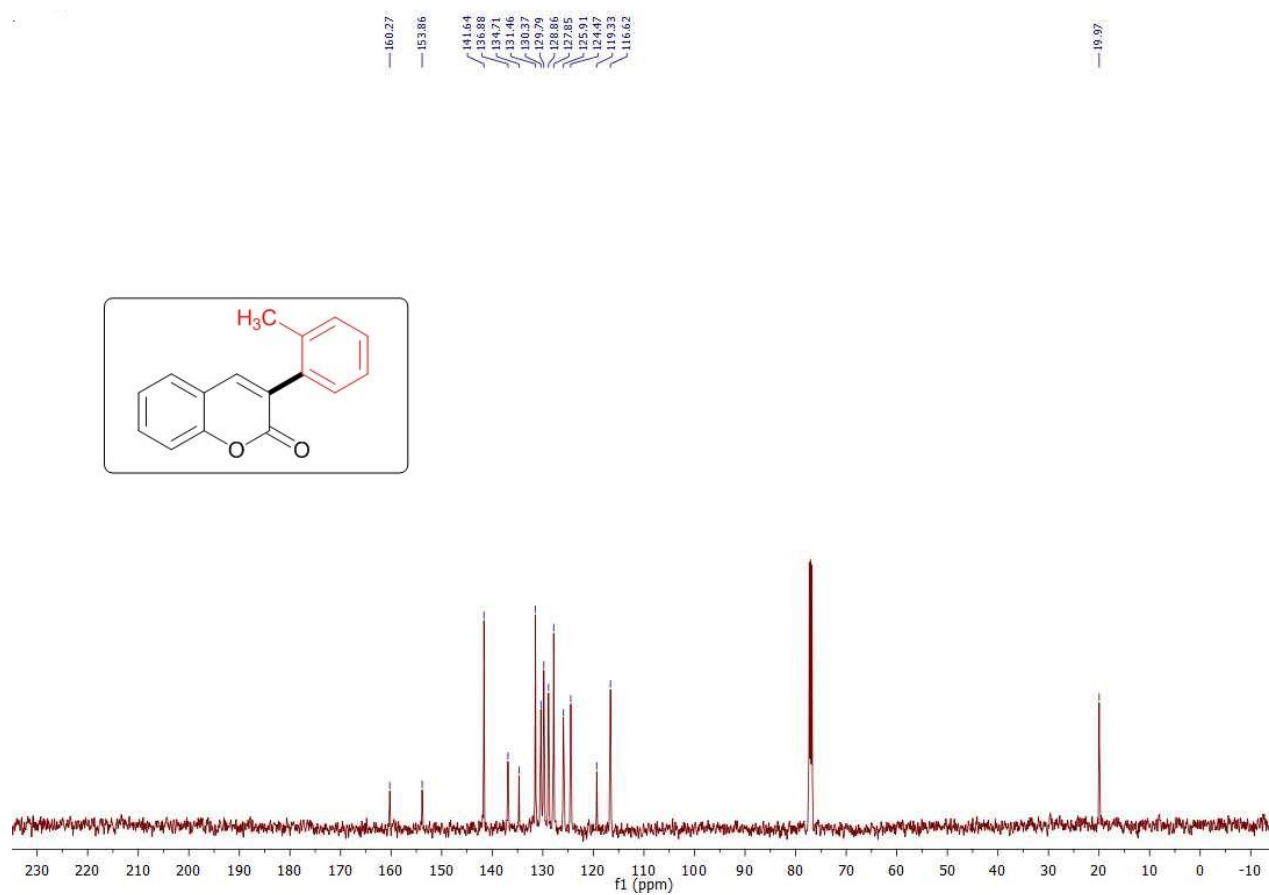
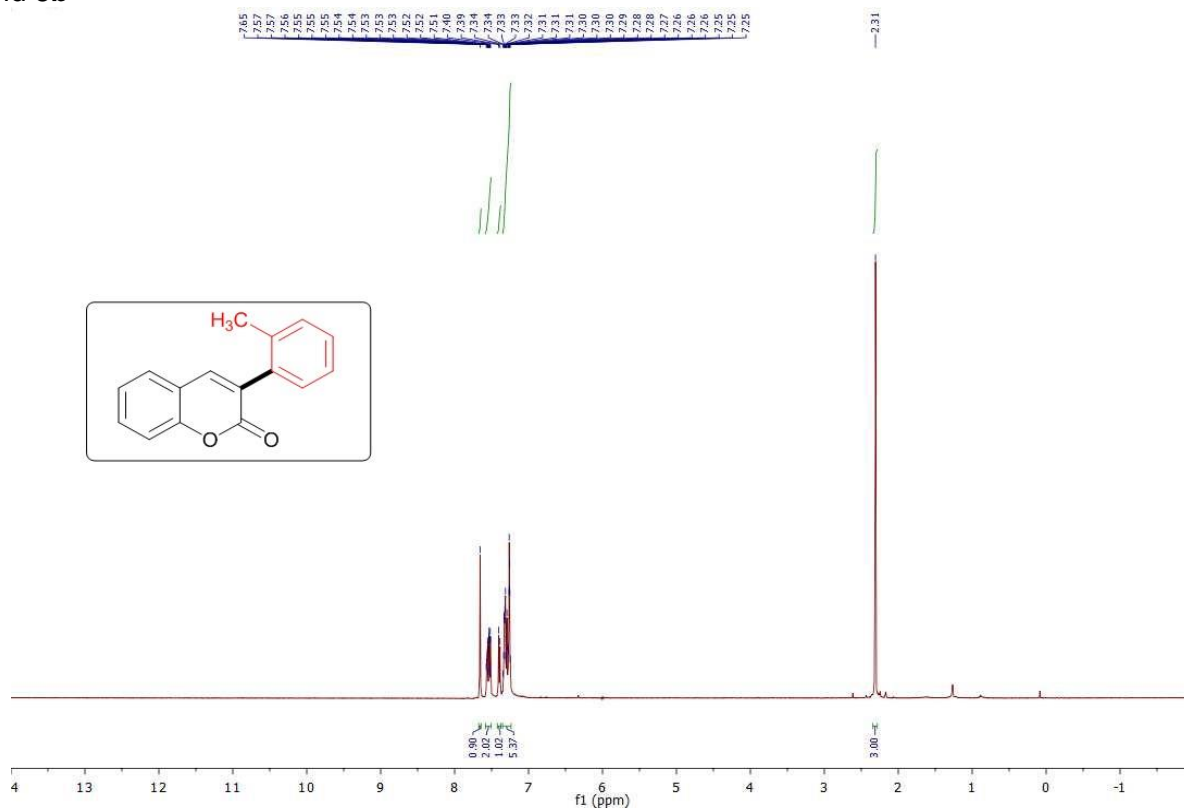
¹²V. Kumar, A. Kalabandi and J. Seetharamappa, *Synth. Commun.*, 2016, **46**, 626.

¹³M. Khoobia, A. Ramazania, A. R. Foroumadib, H. Hamadic, Z. Hojjatia and A. Shafieeb, *J. Iran. Chem. Soc.*, 2011, **8**, 1036.

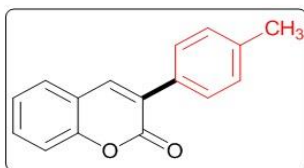
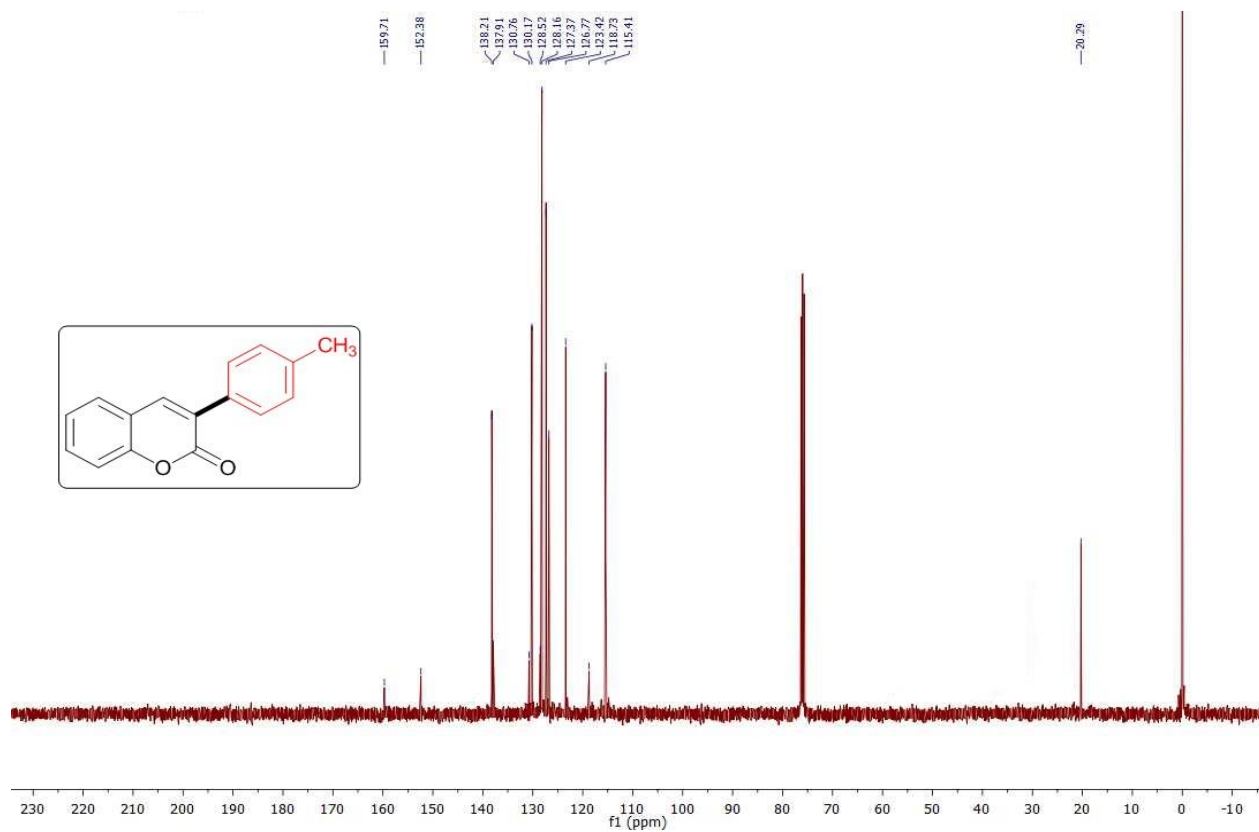
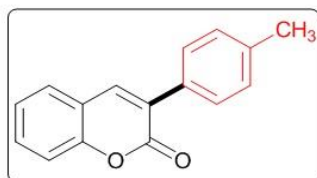
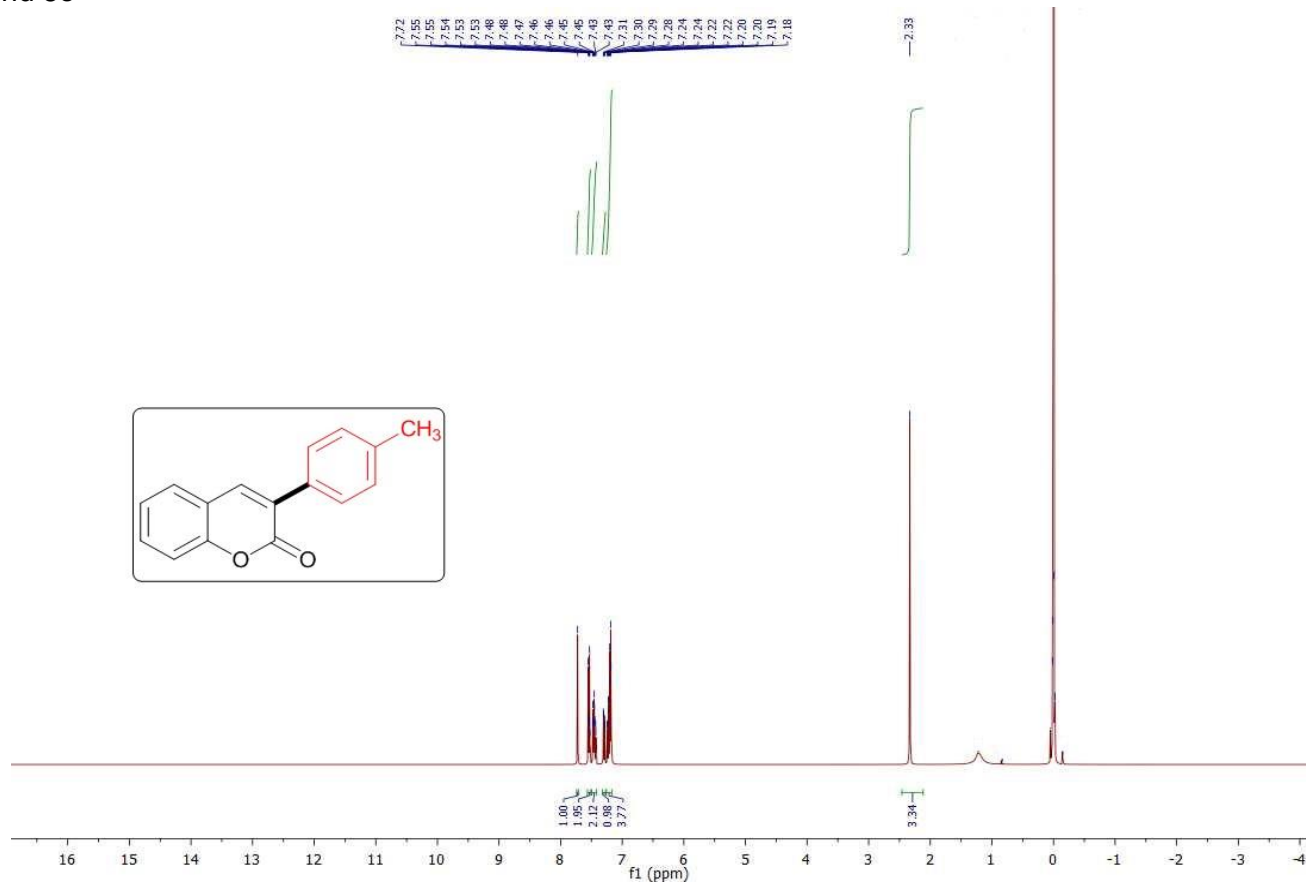
Compound 3a



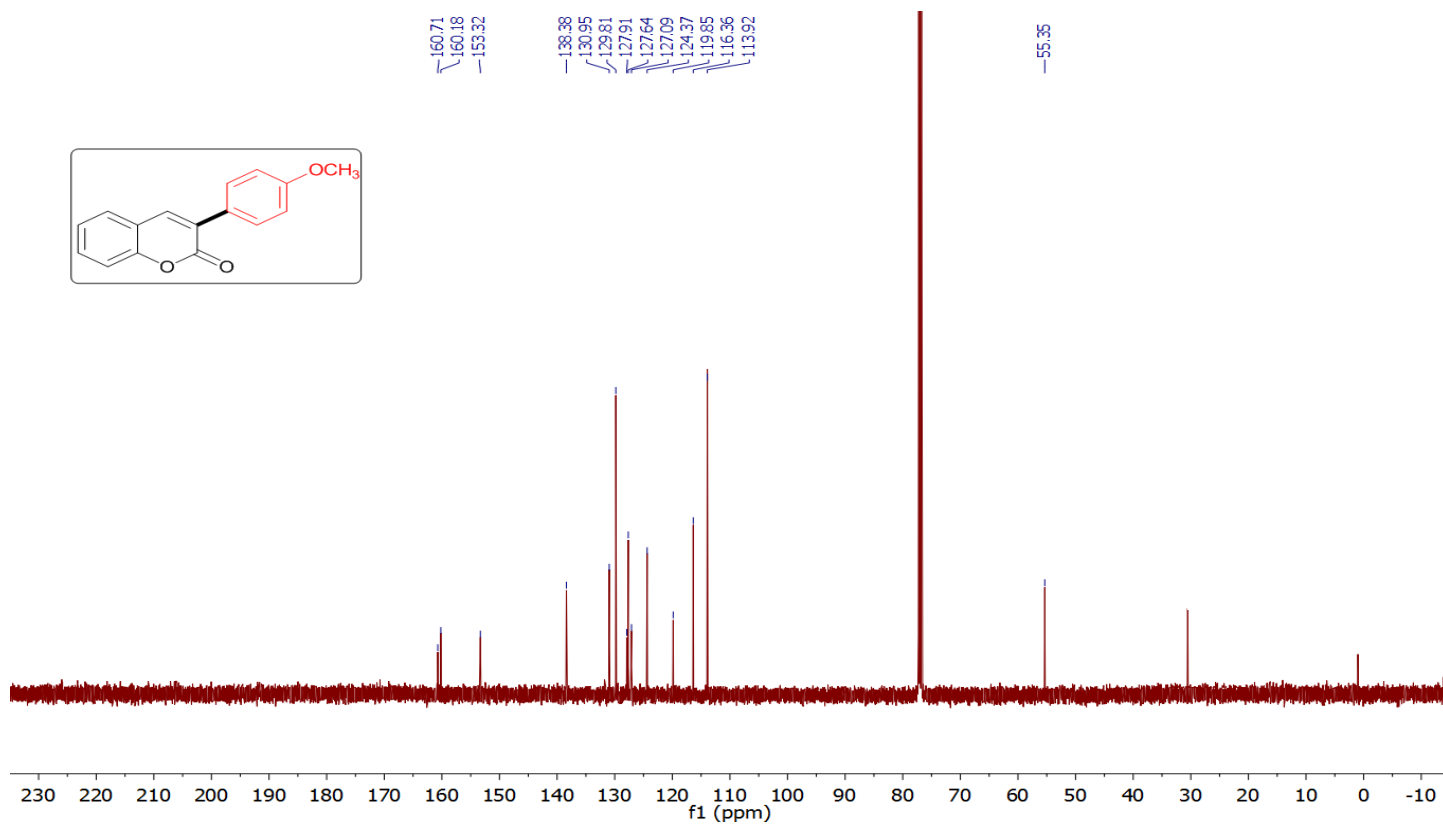
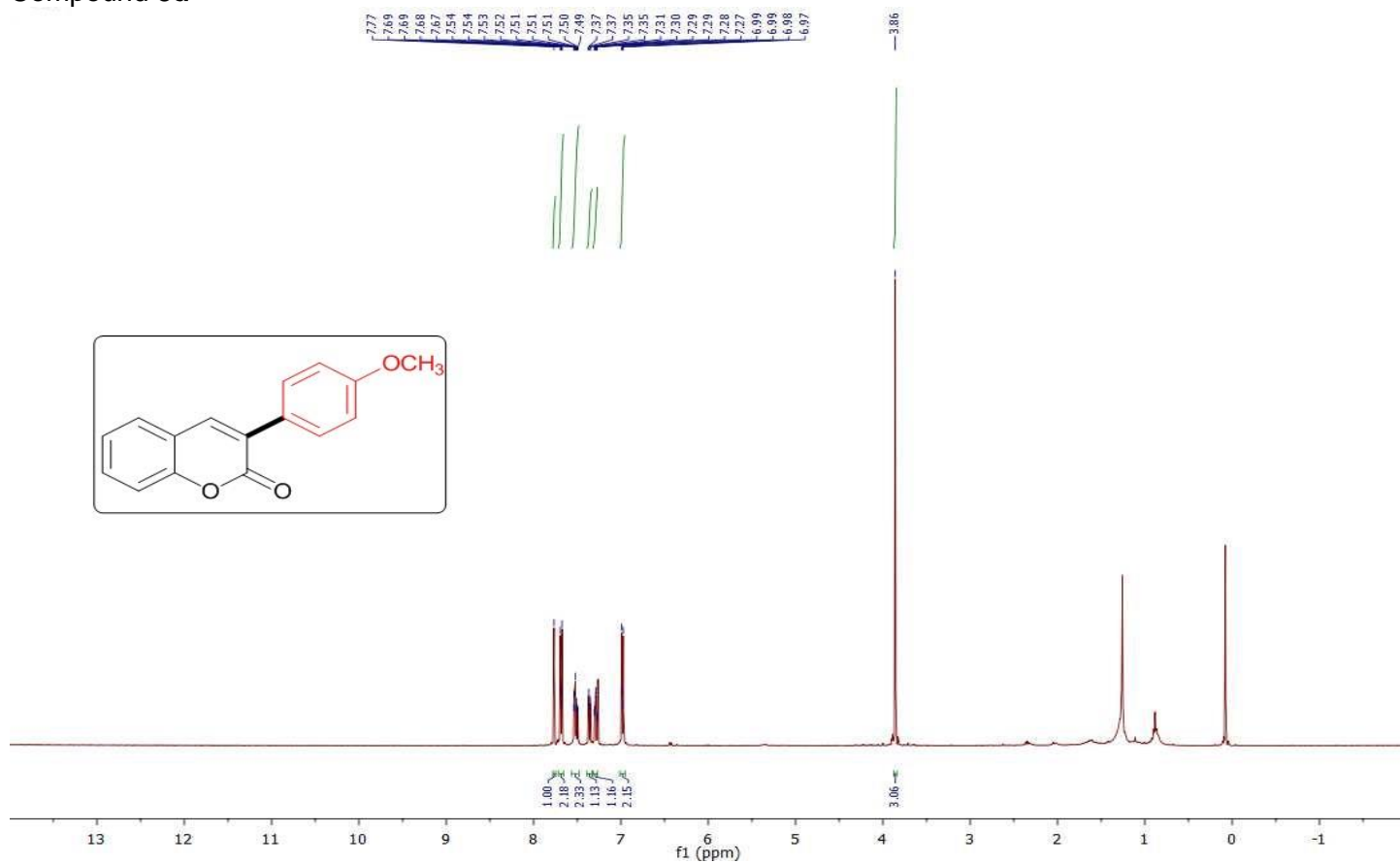
Compound **3b**



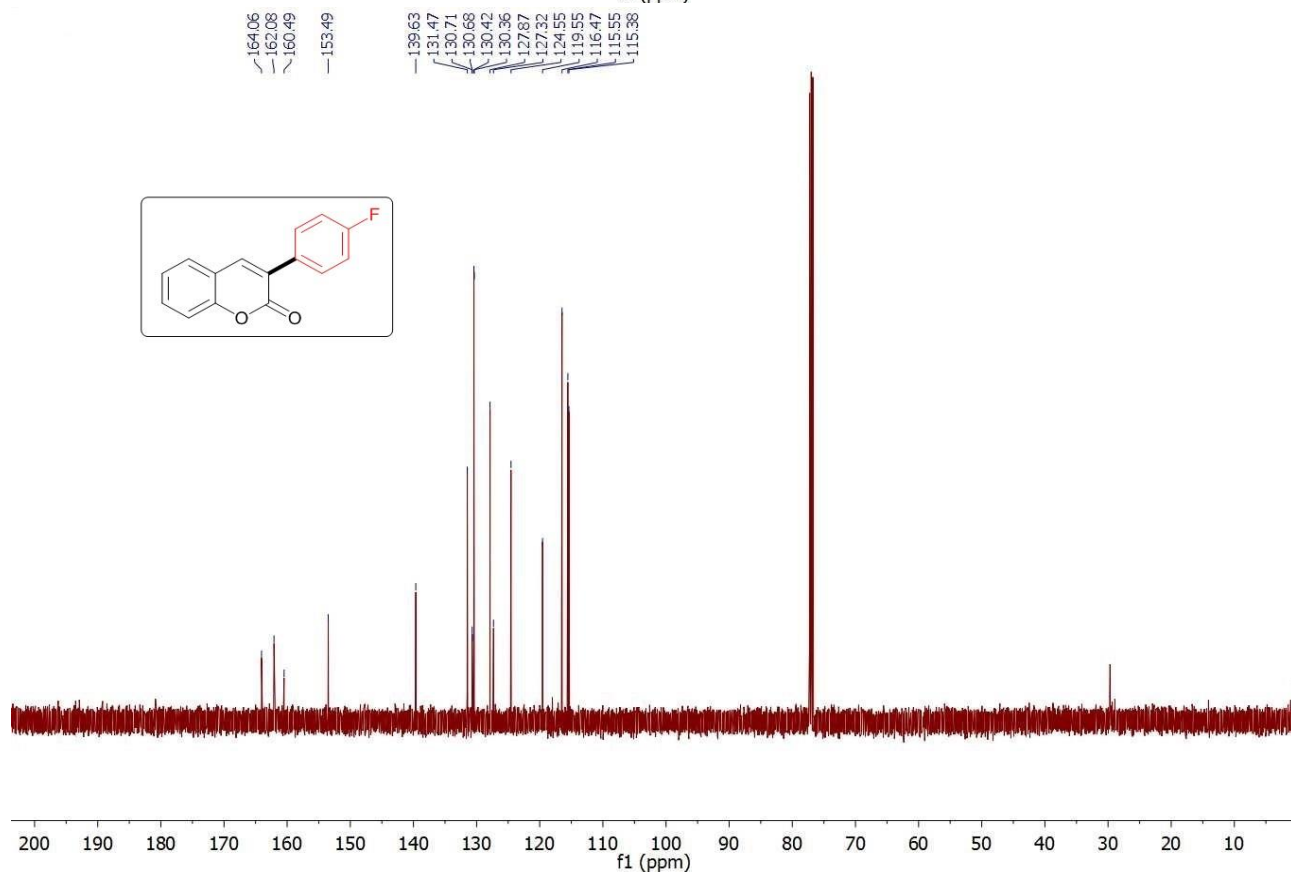
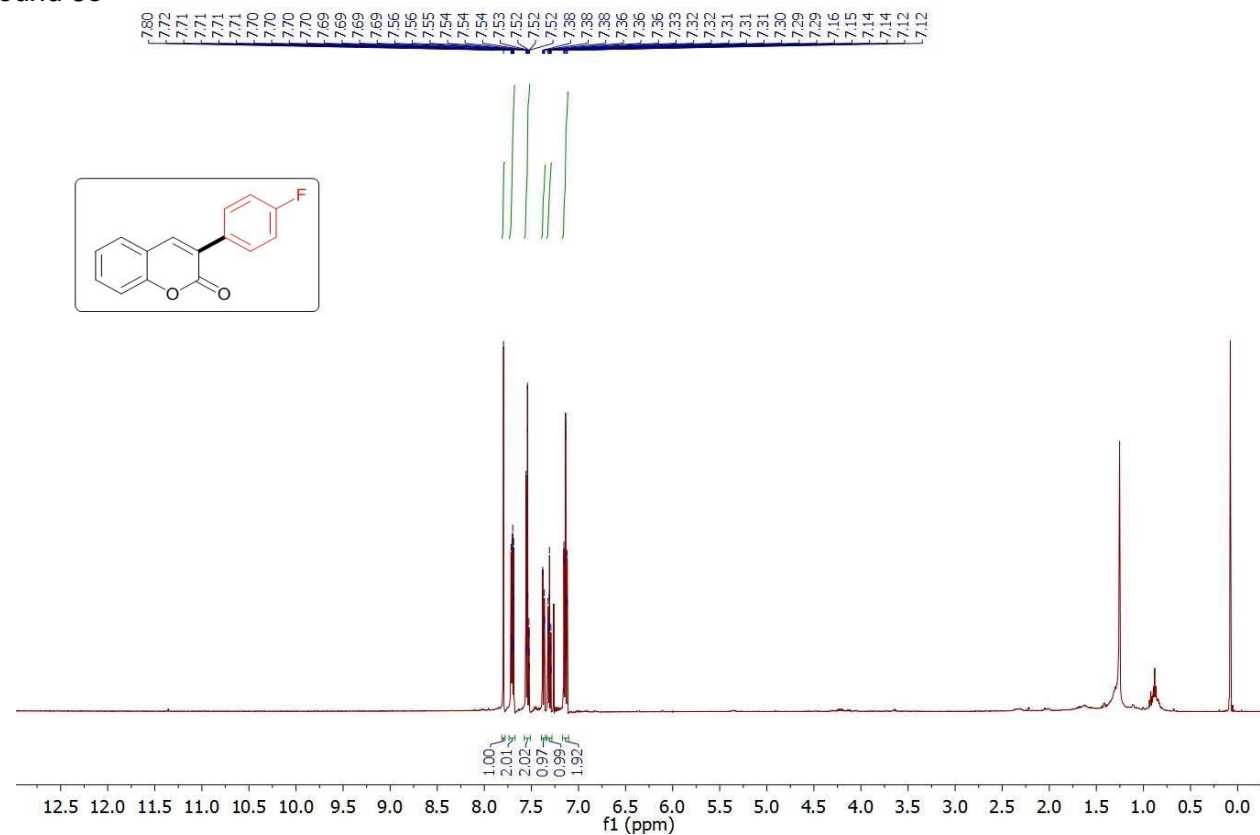
Compound 3c



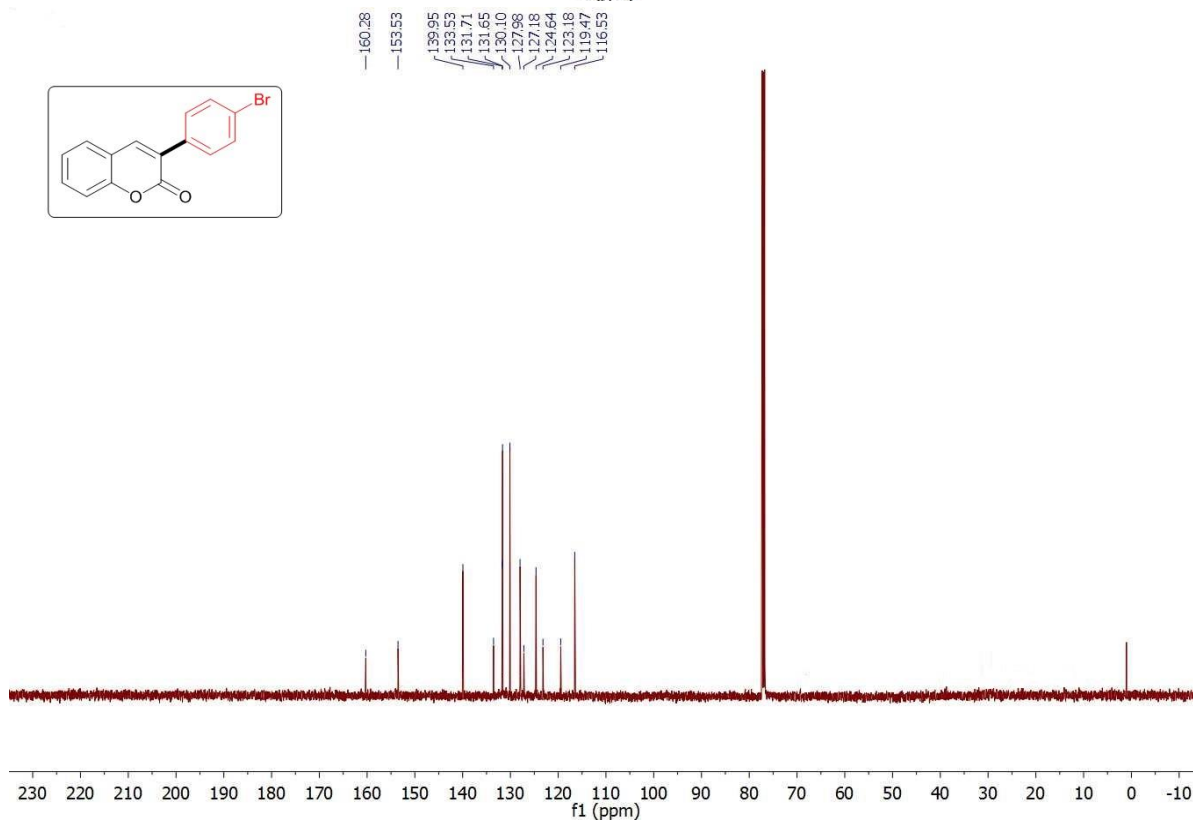
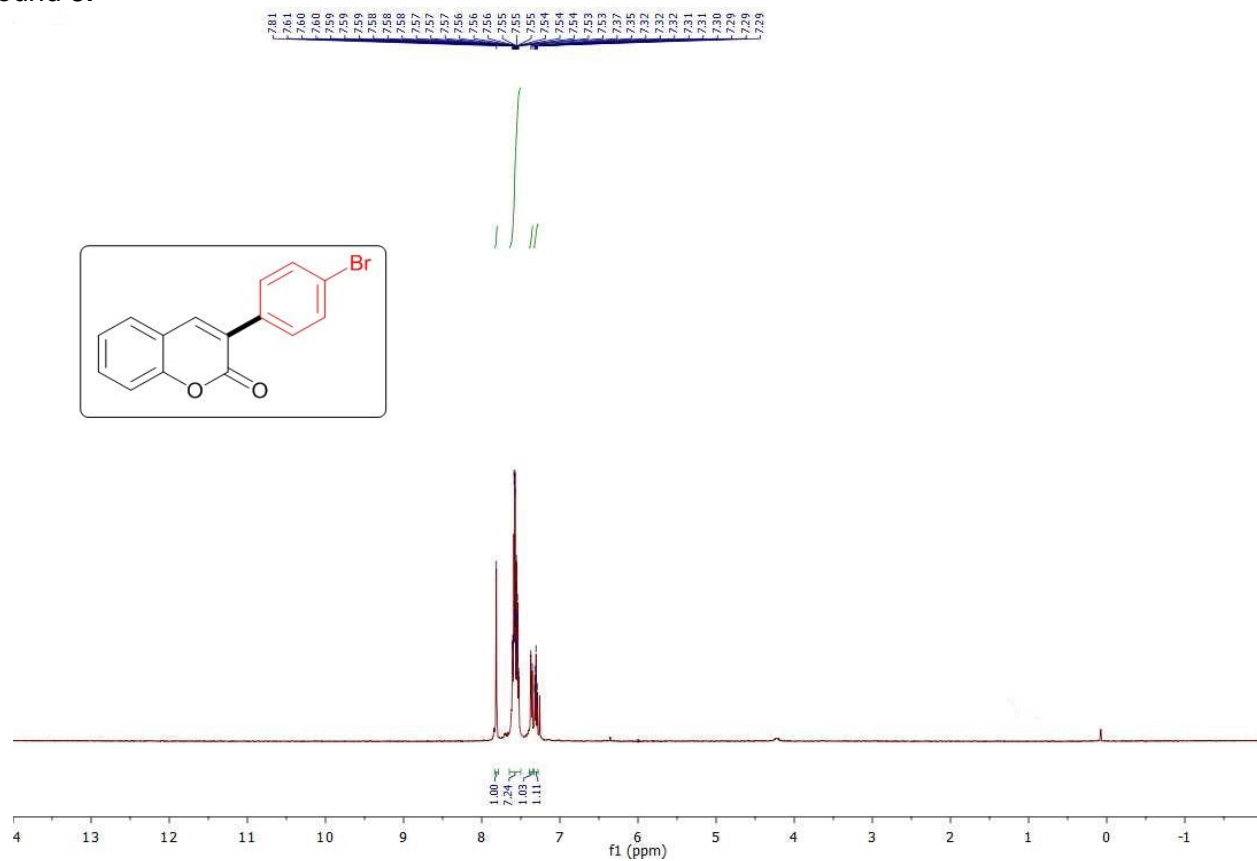
Compound 3d



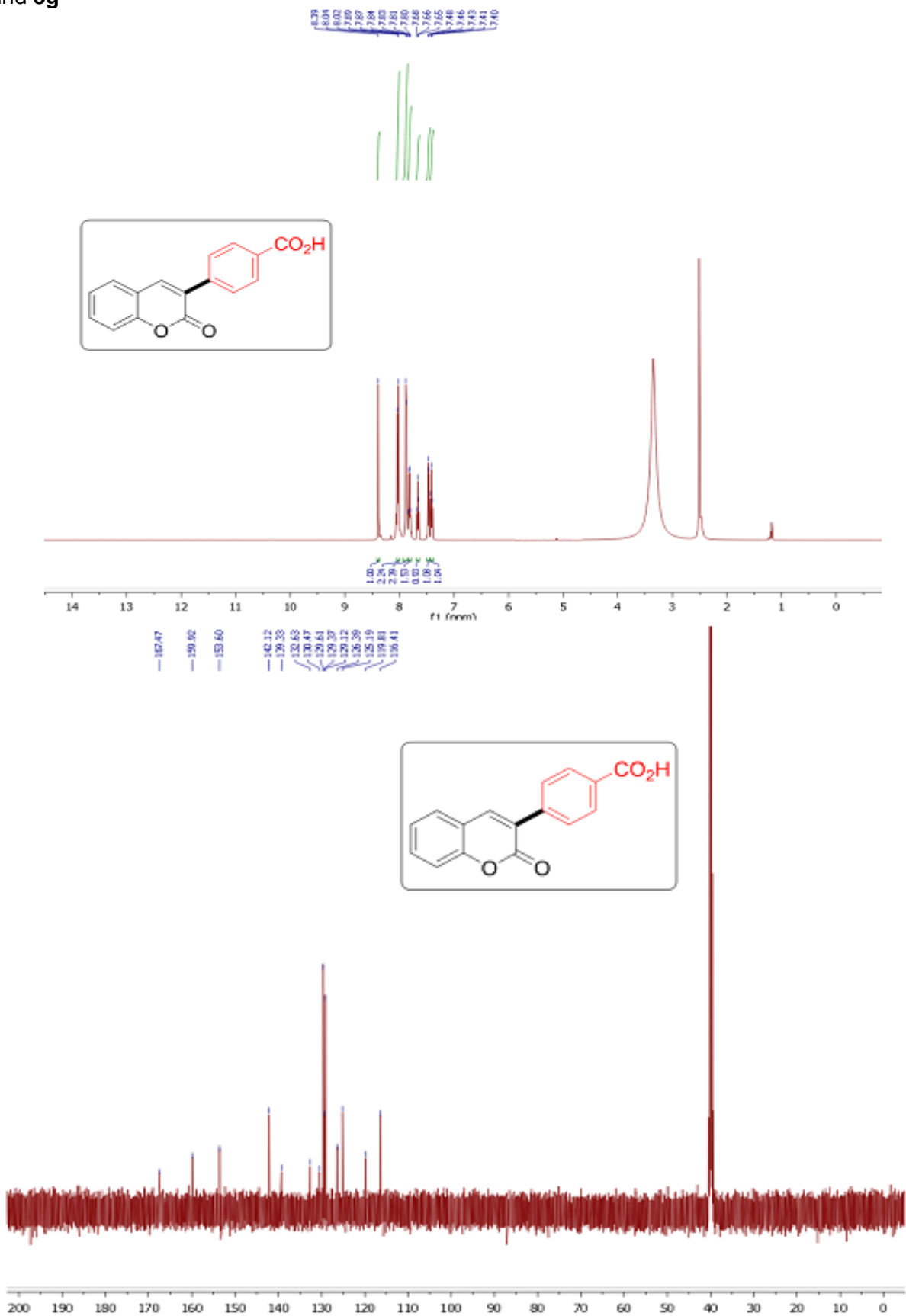
Compound 3e



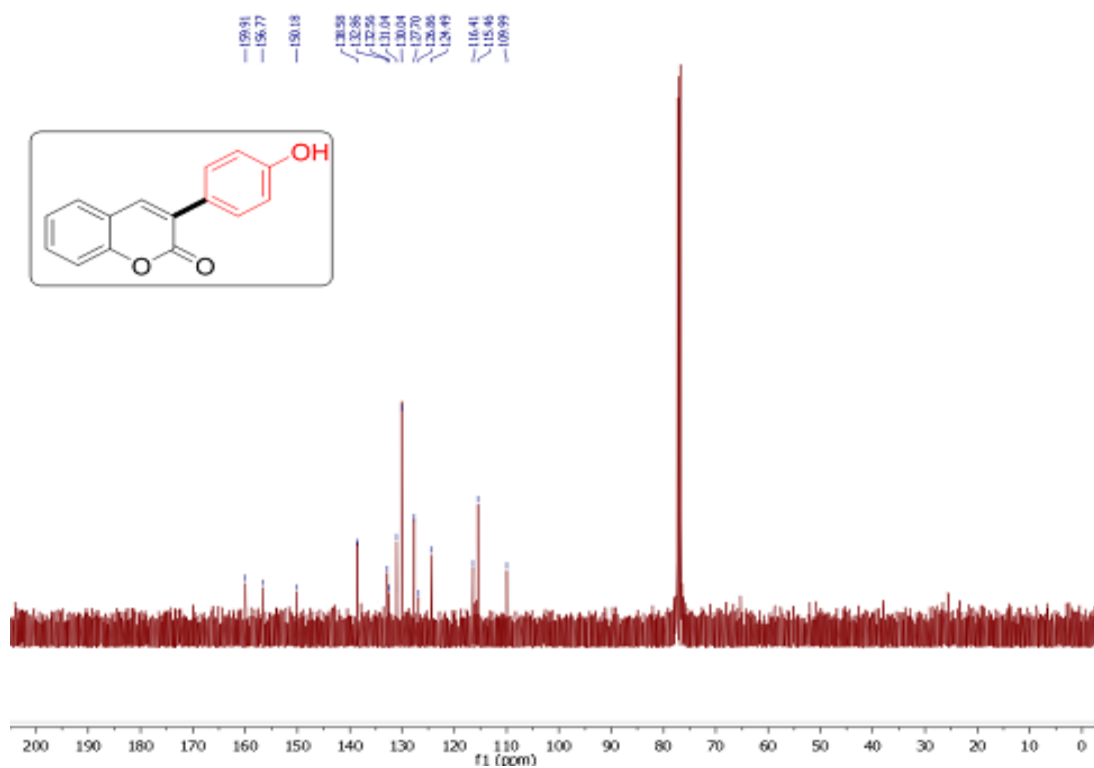
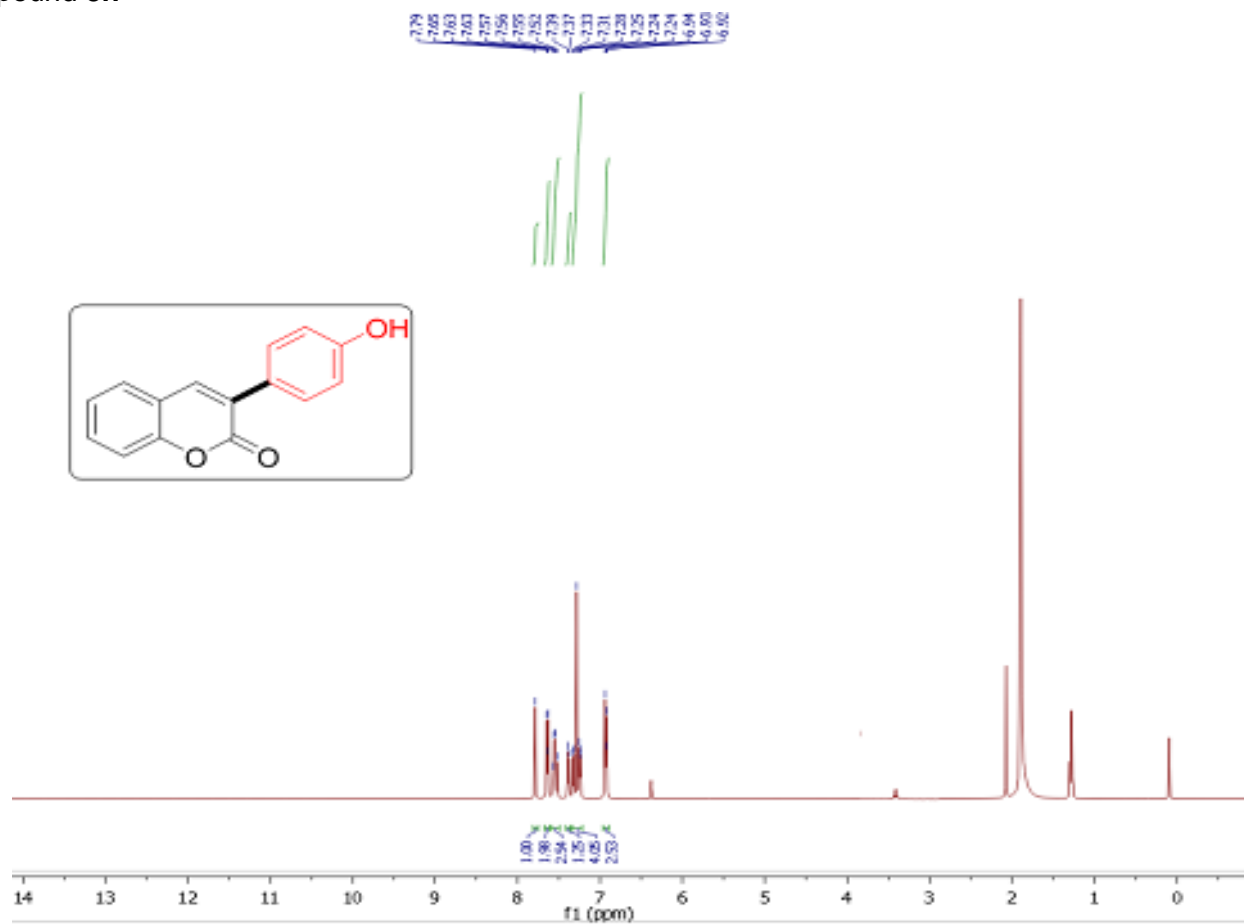
Compound **3f**



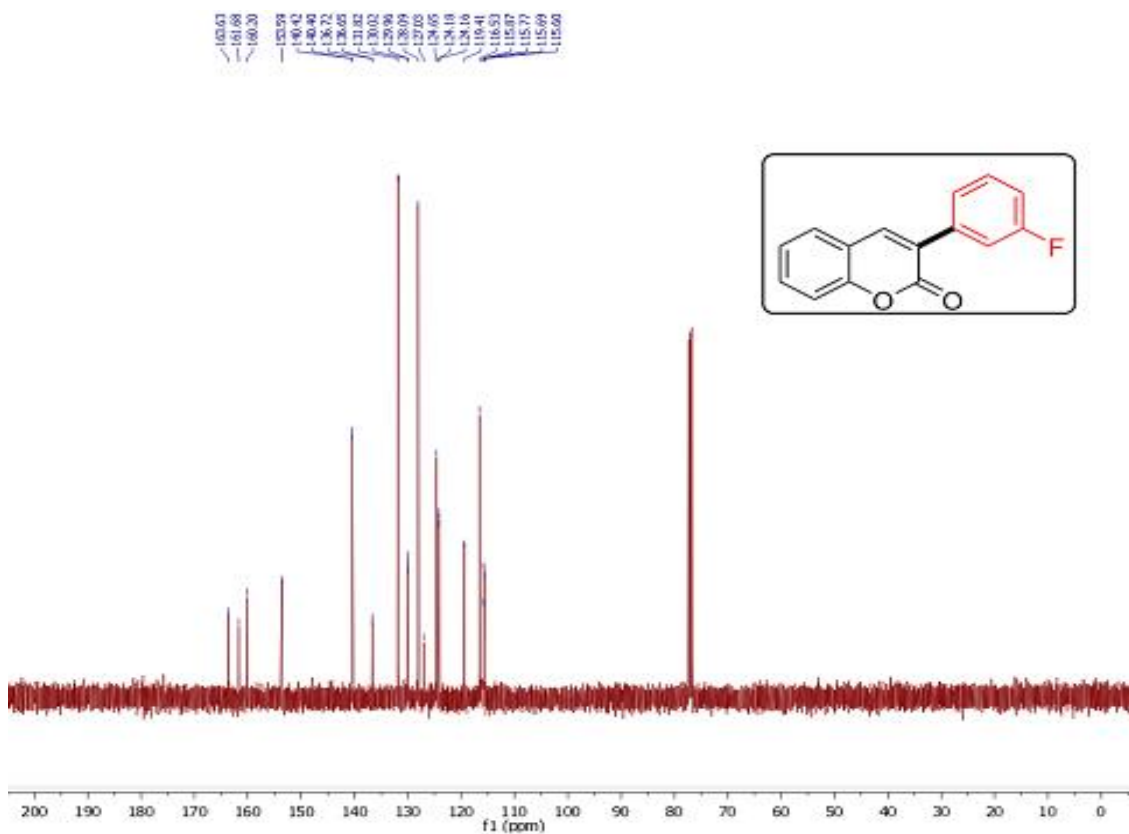
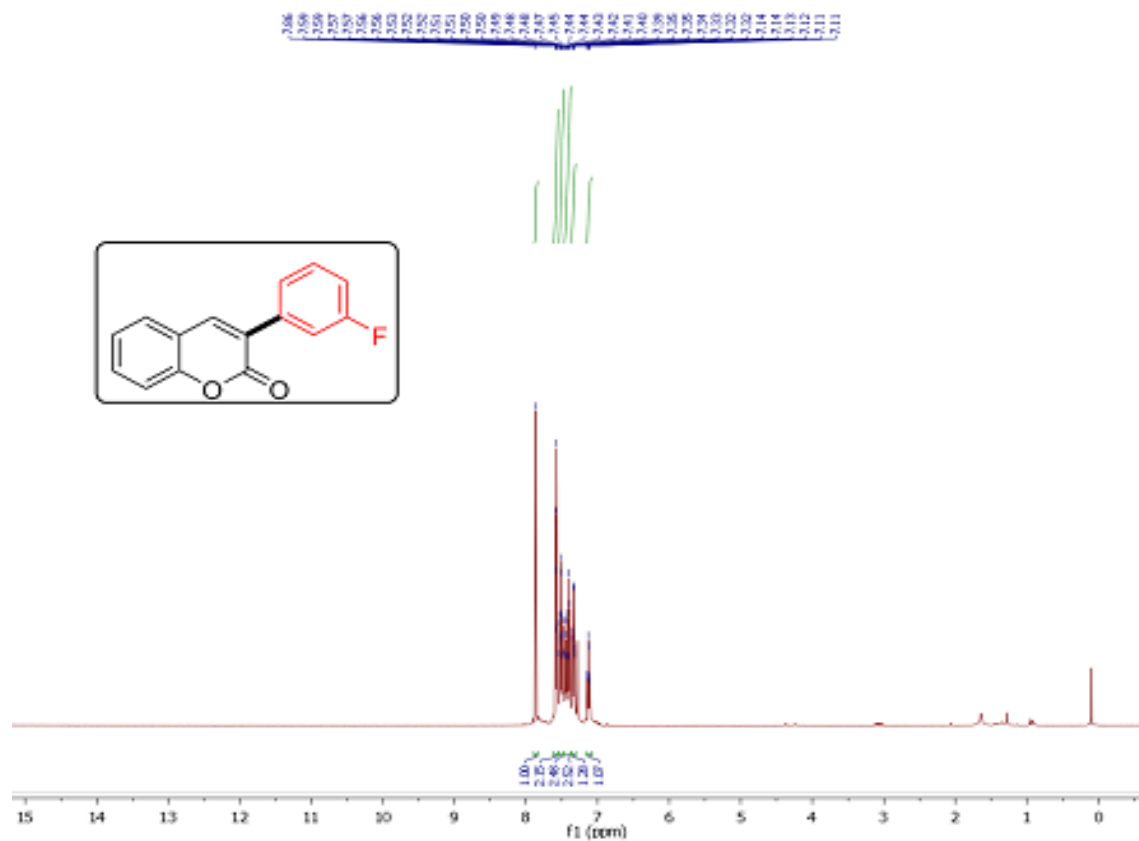
Compound **3g**



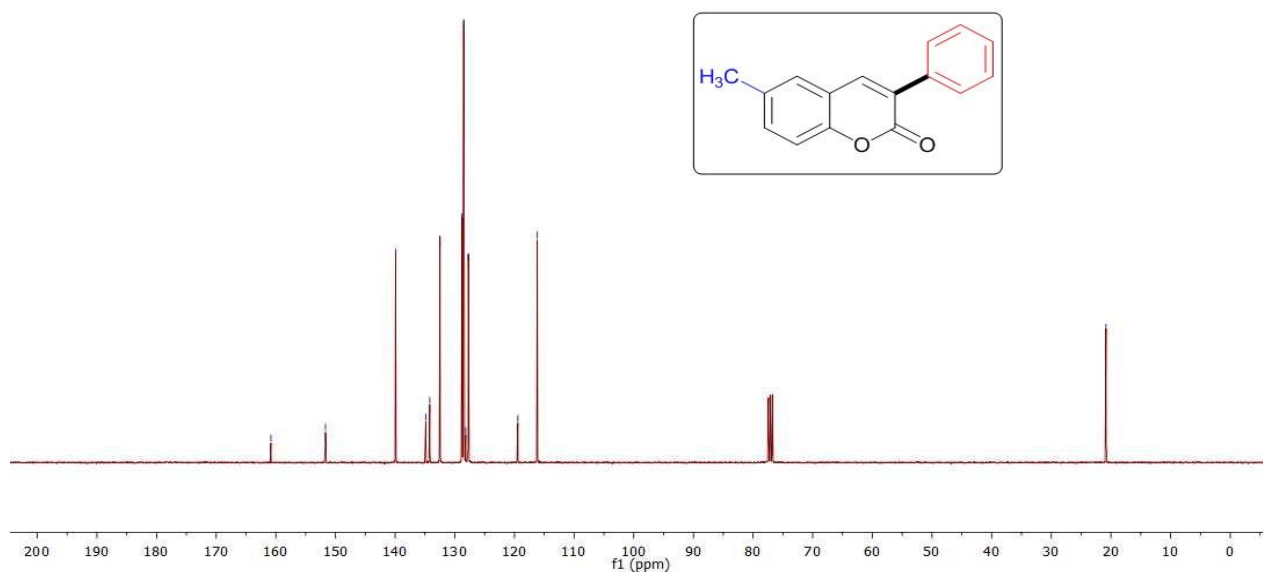
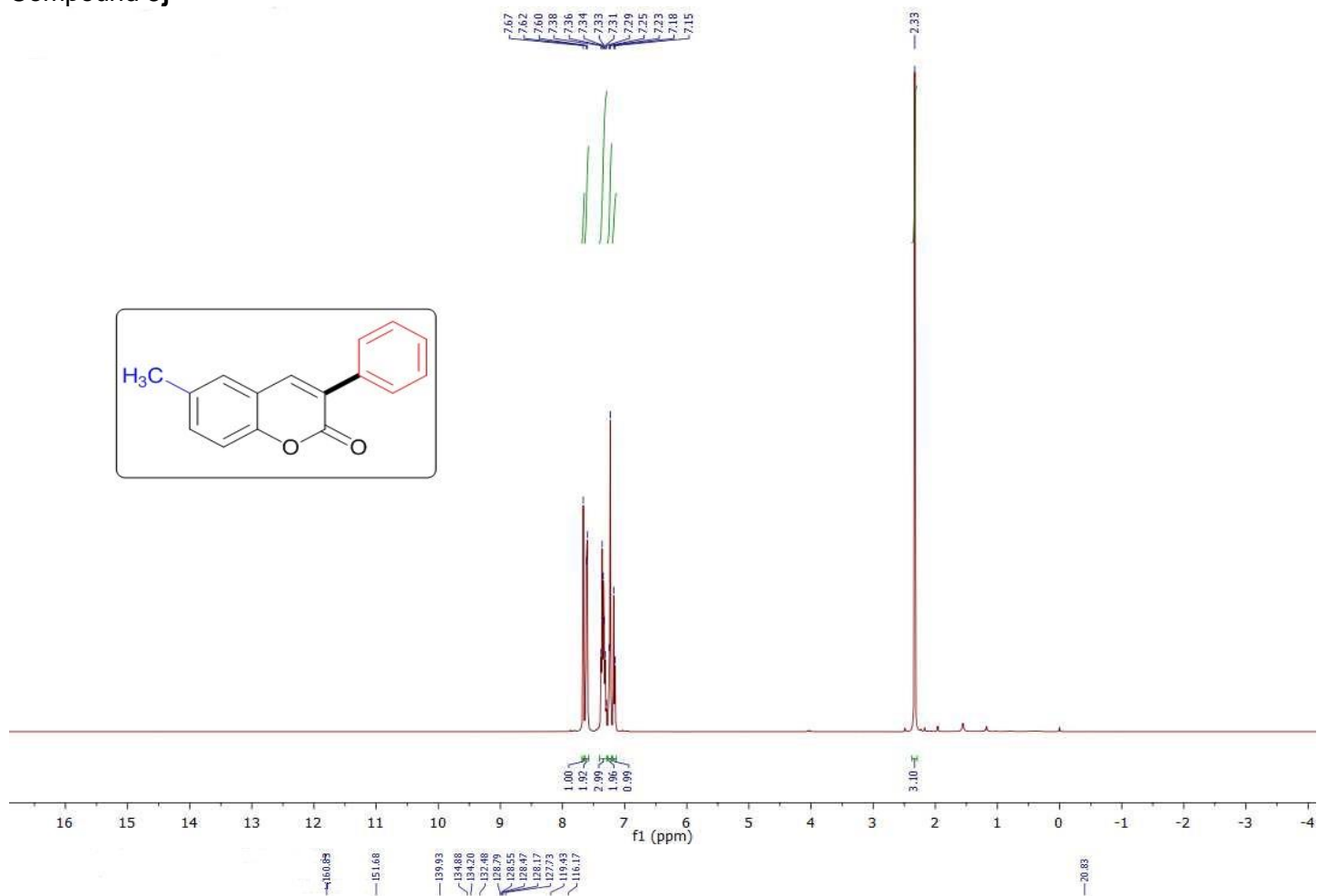
Compound 3h



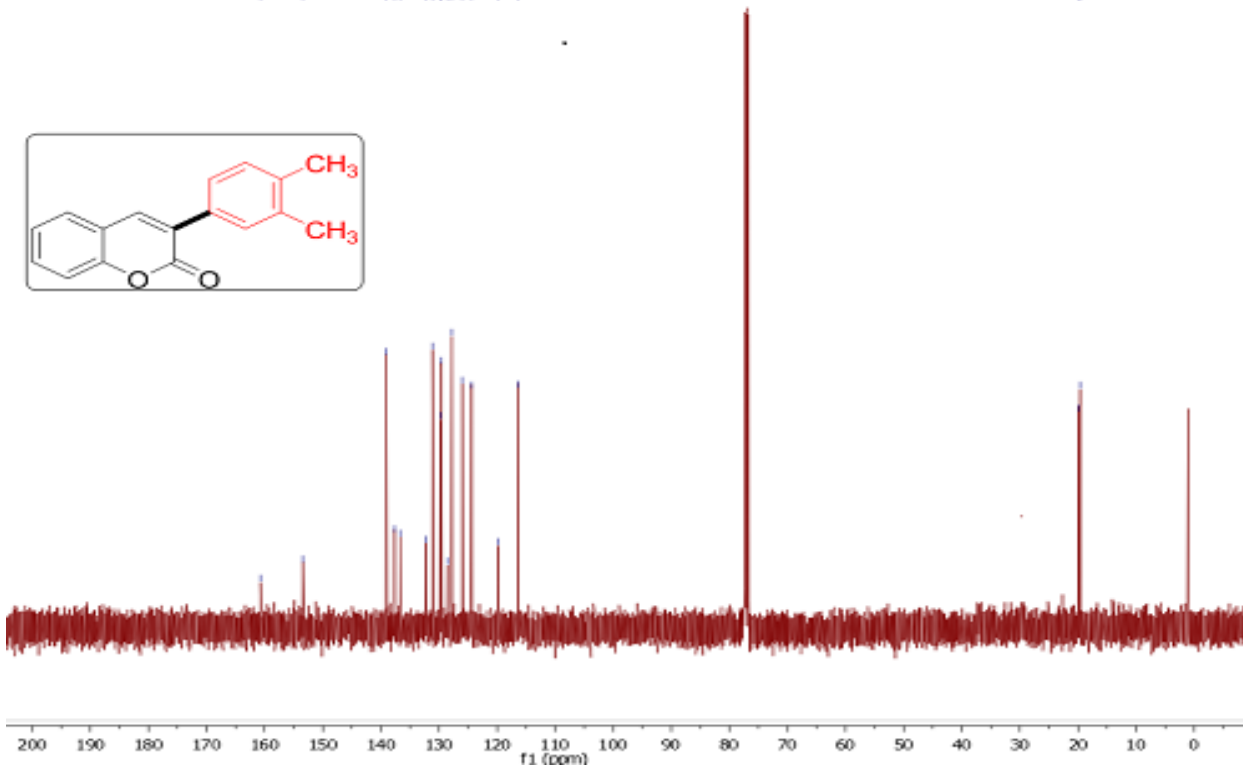
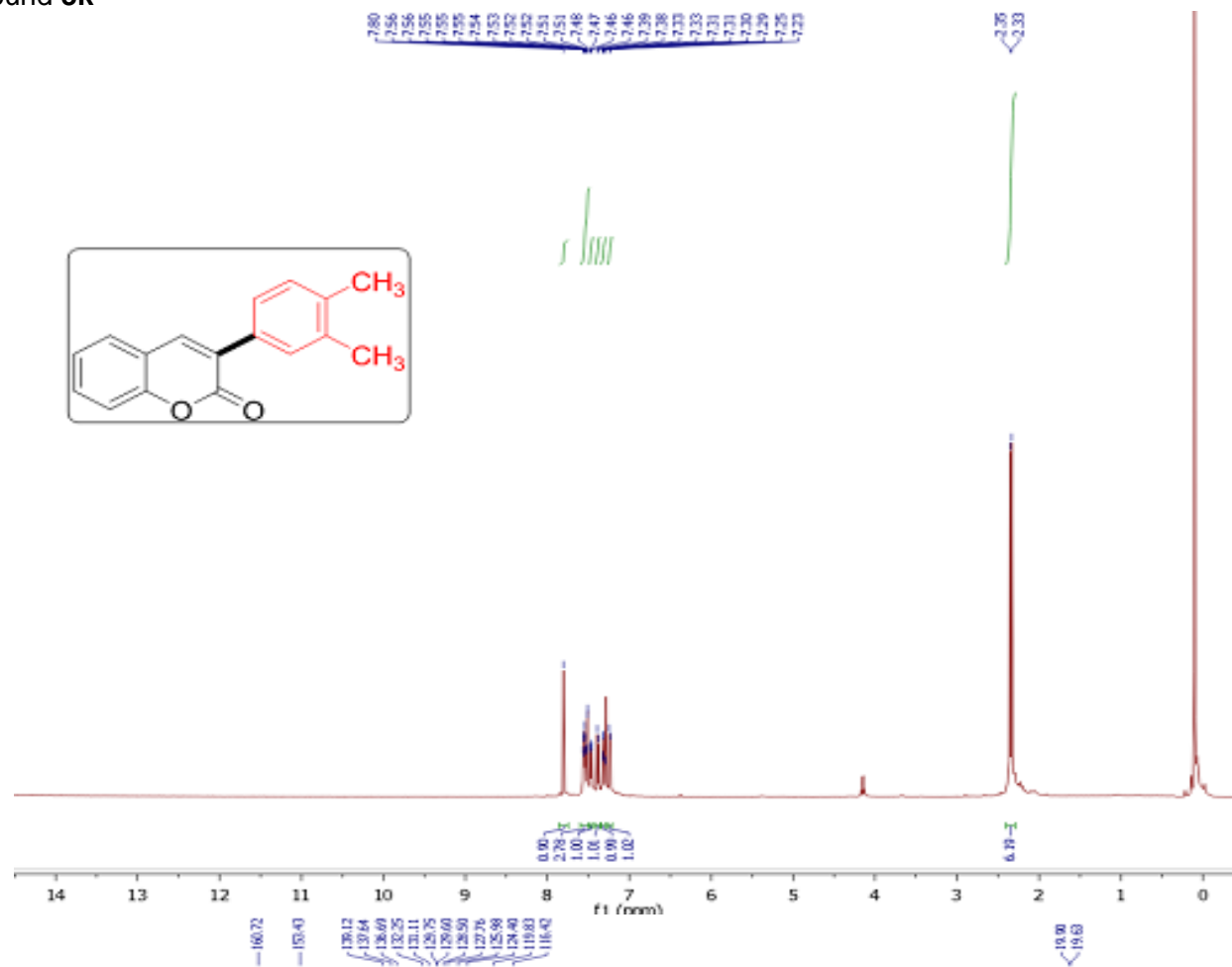
Compound **3i**



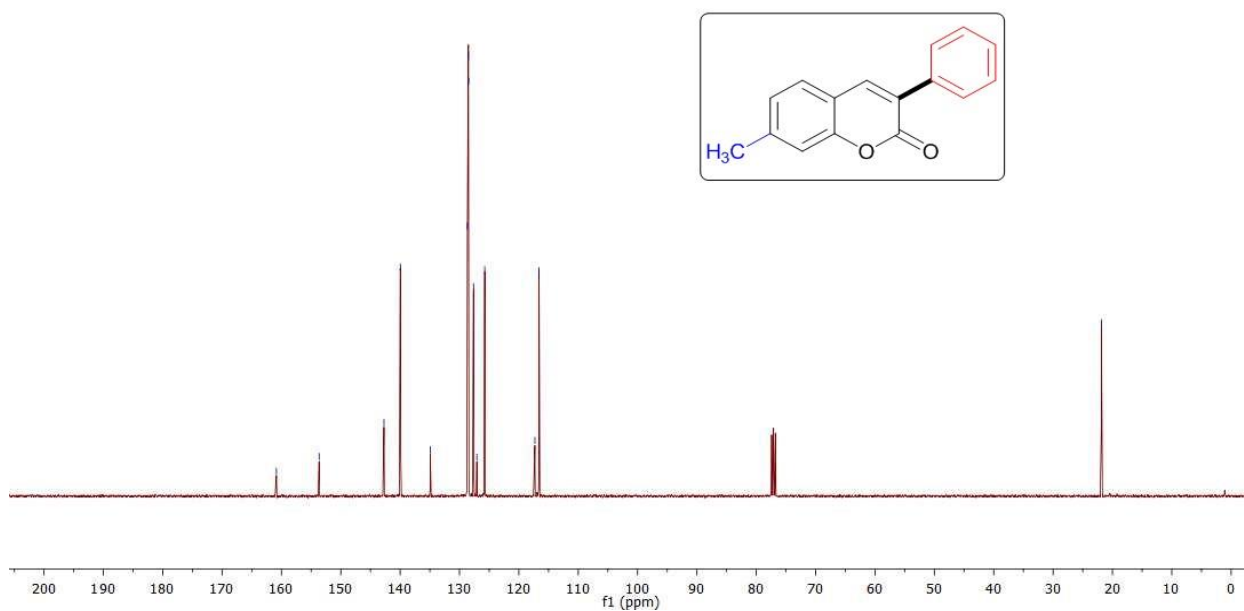
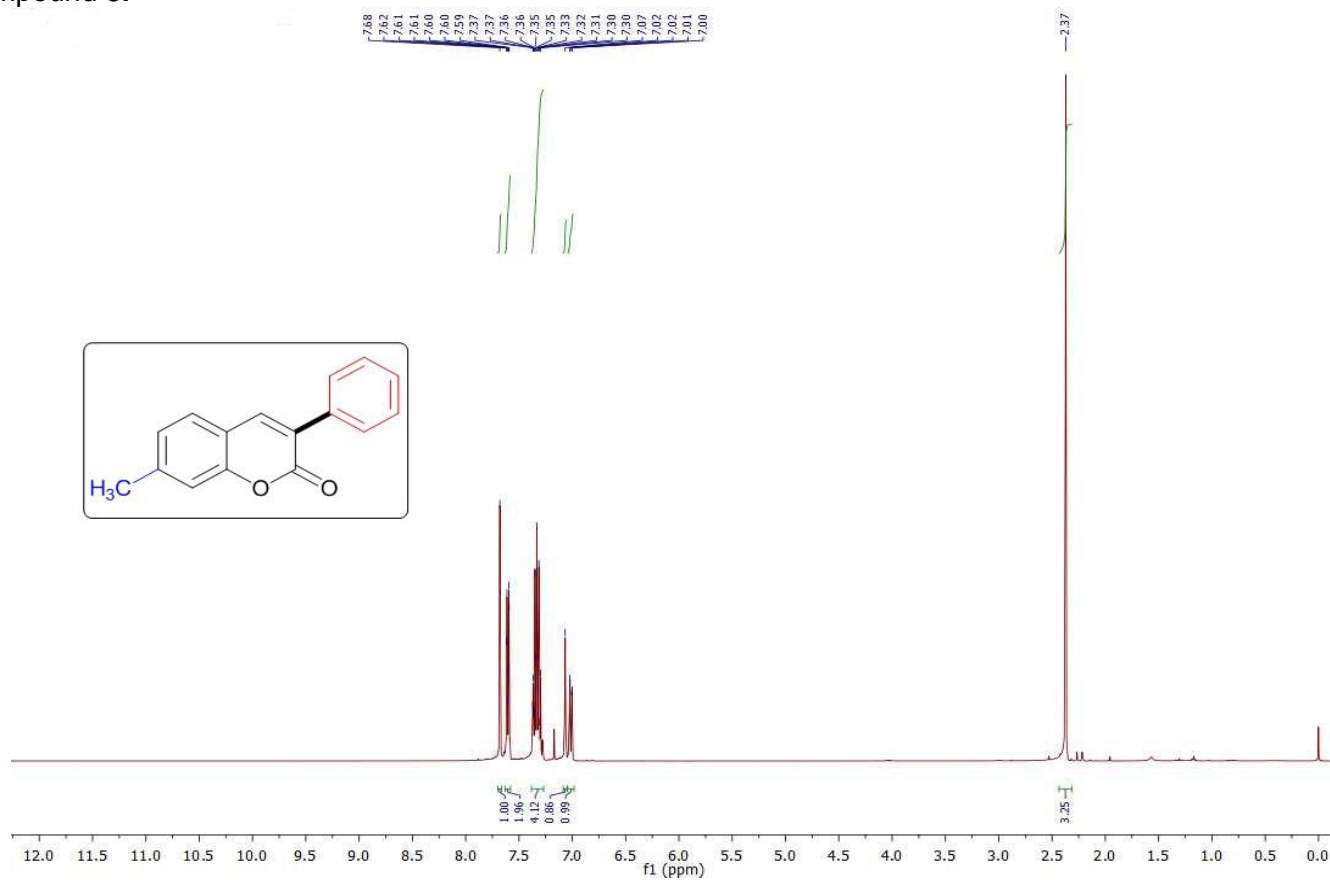
Compound 3j



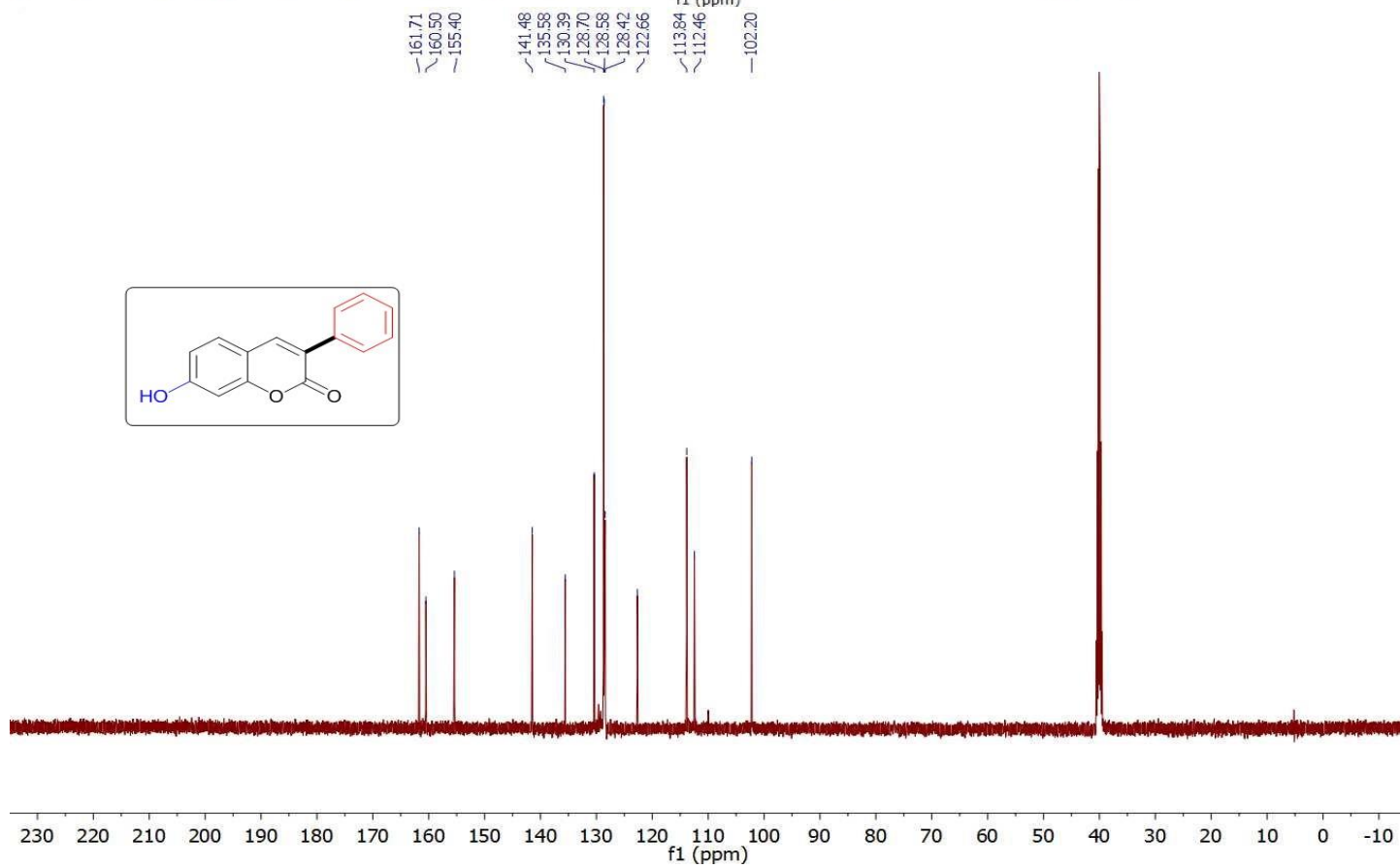
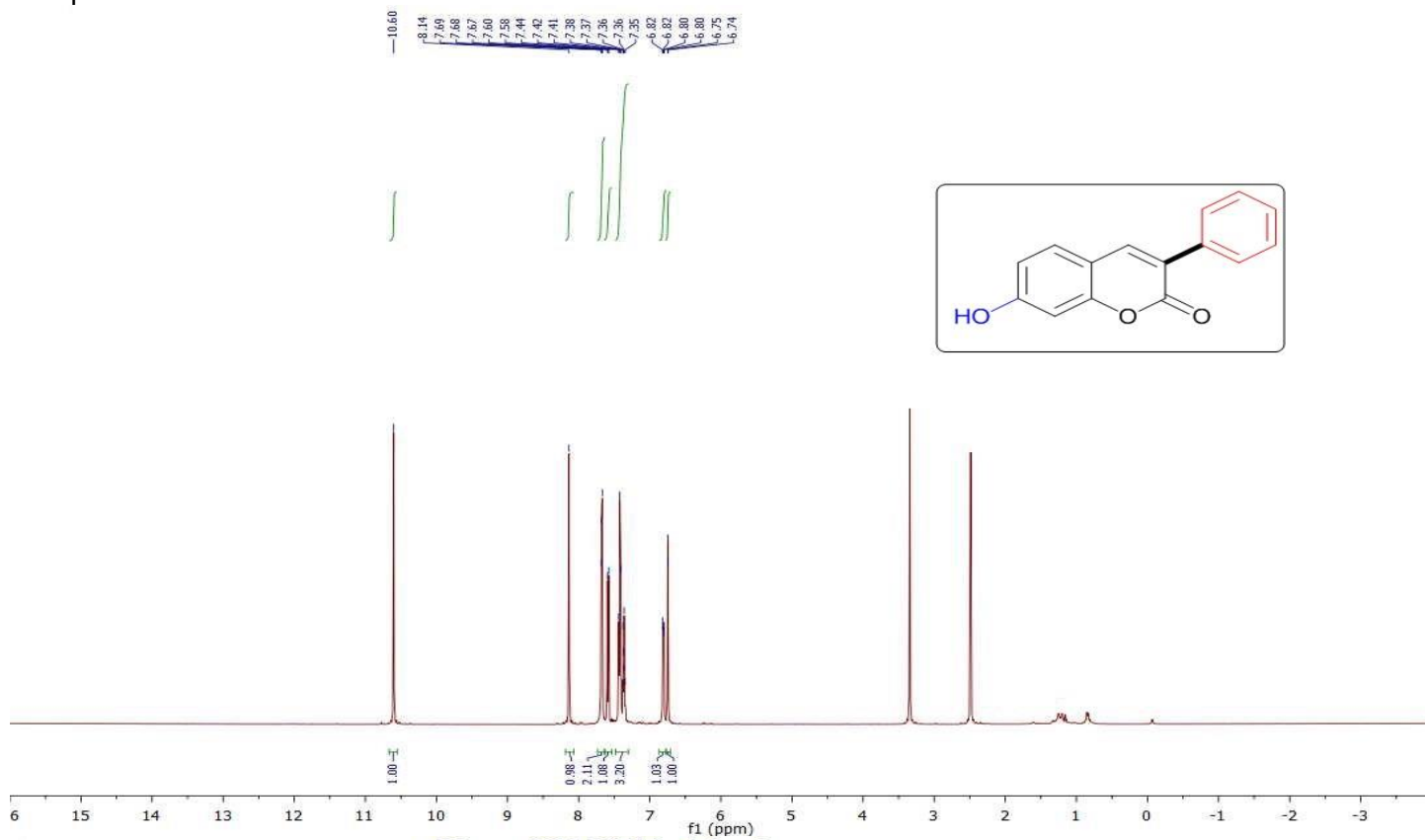
Compound **3k**



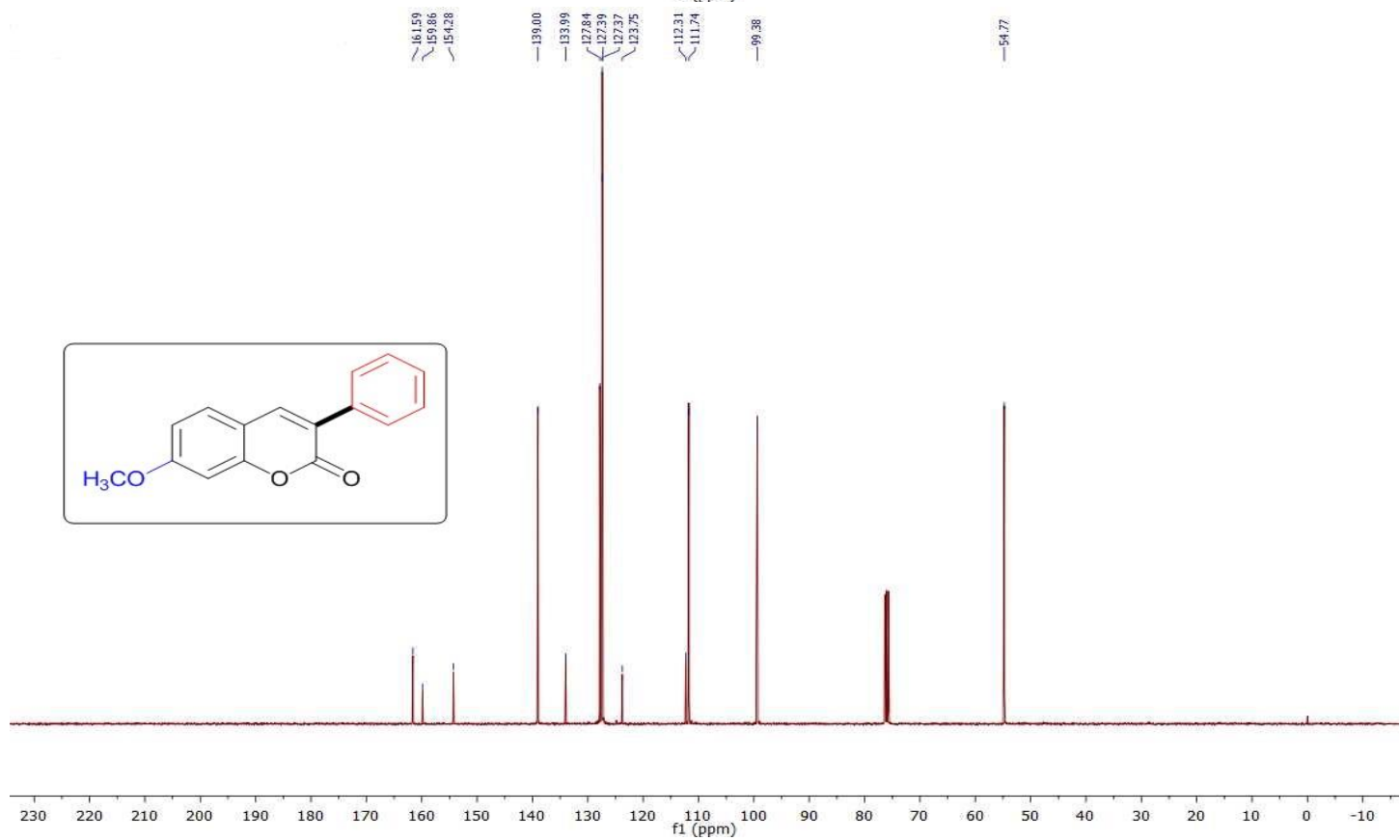
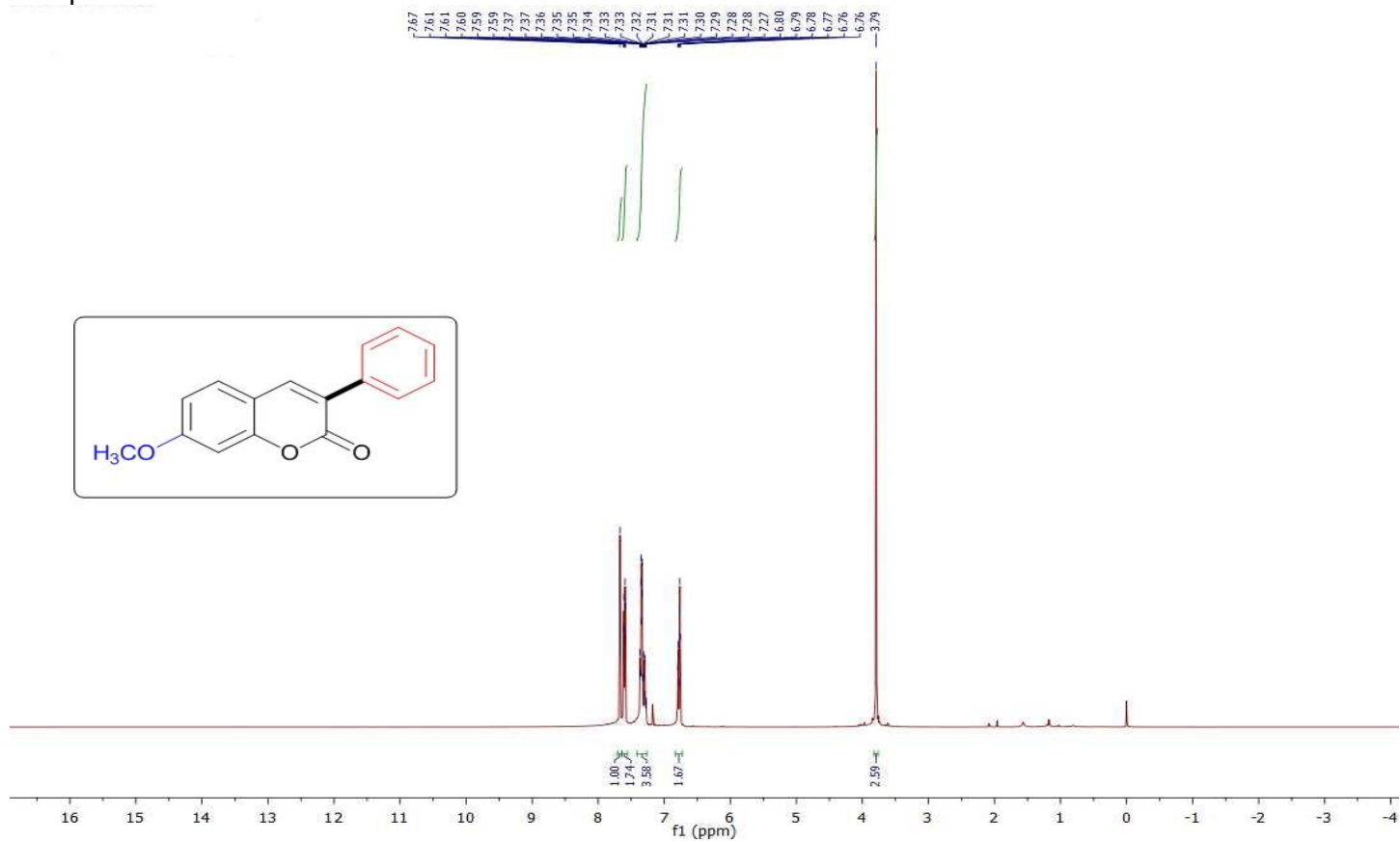
Compound **31**



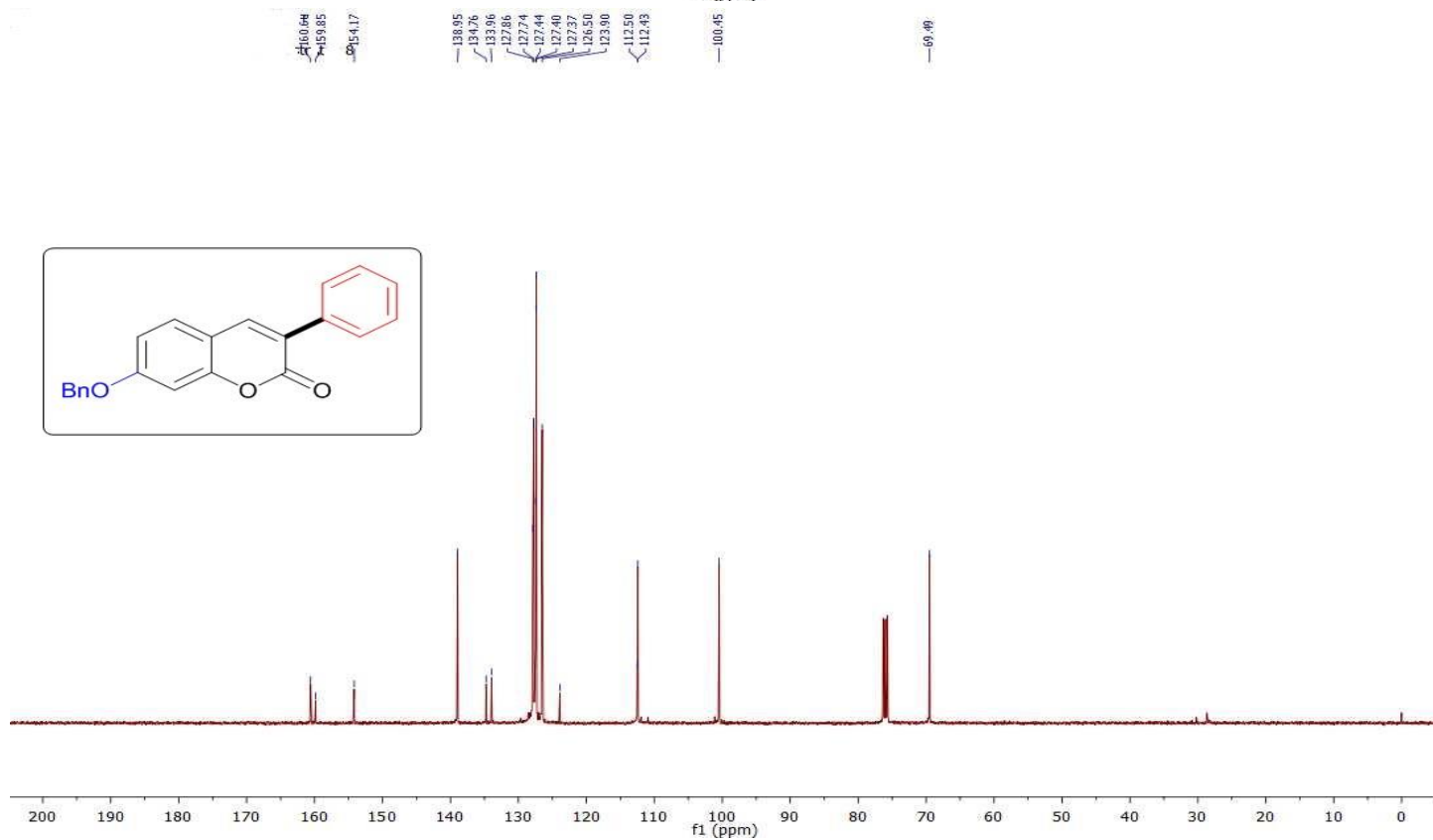
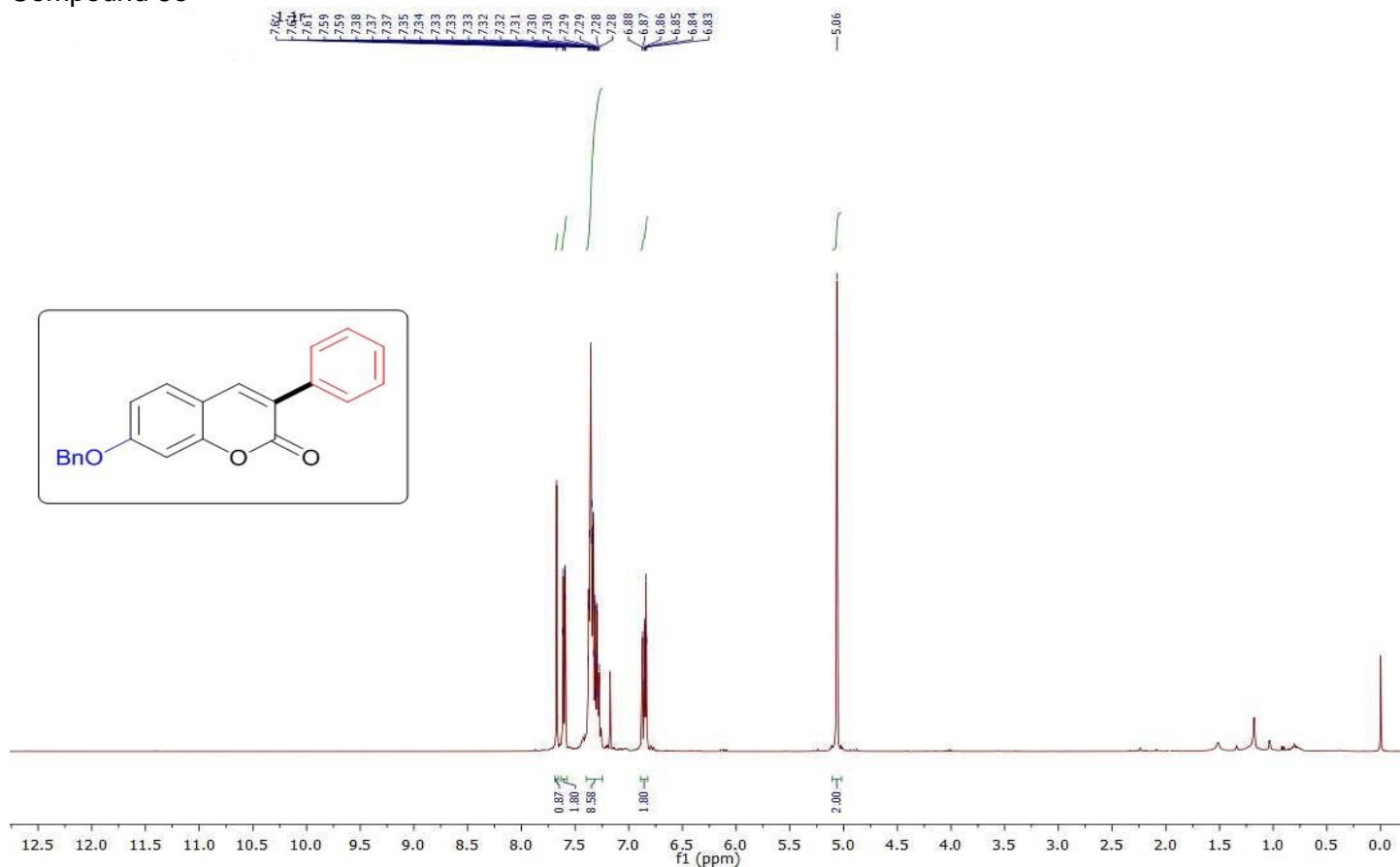
Compound 3m



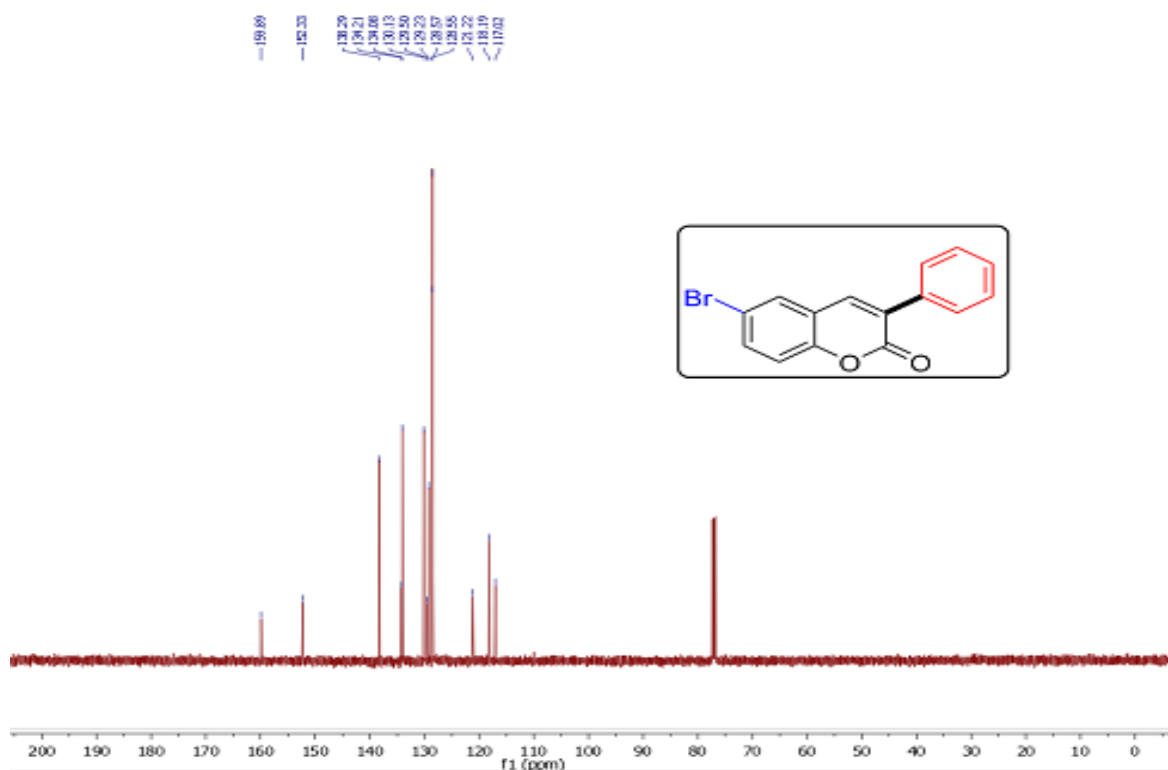
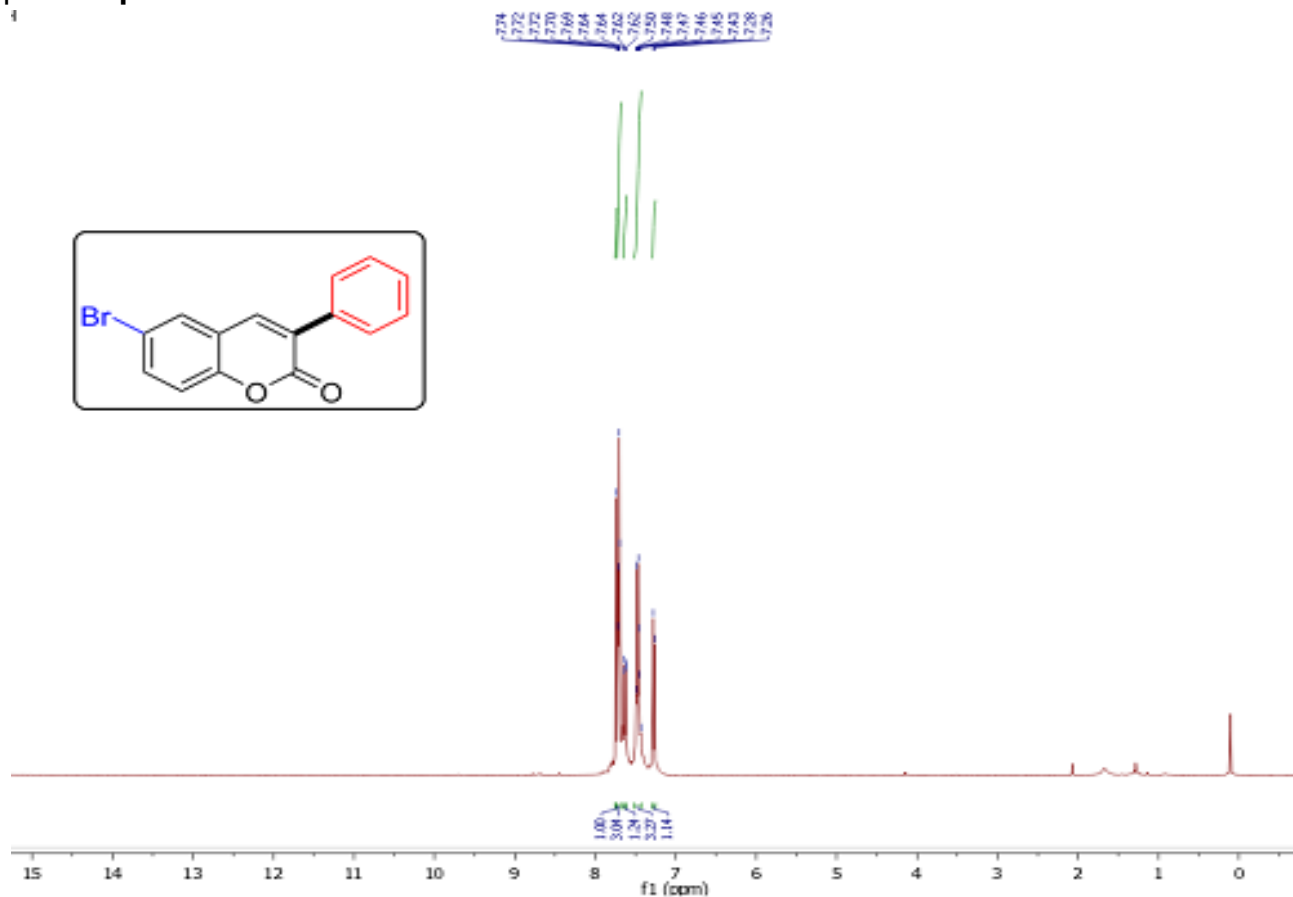
Compound 3n



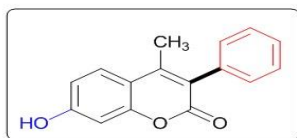
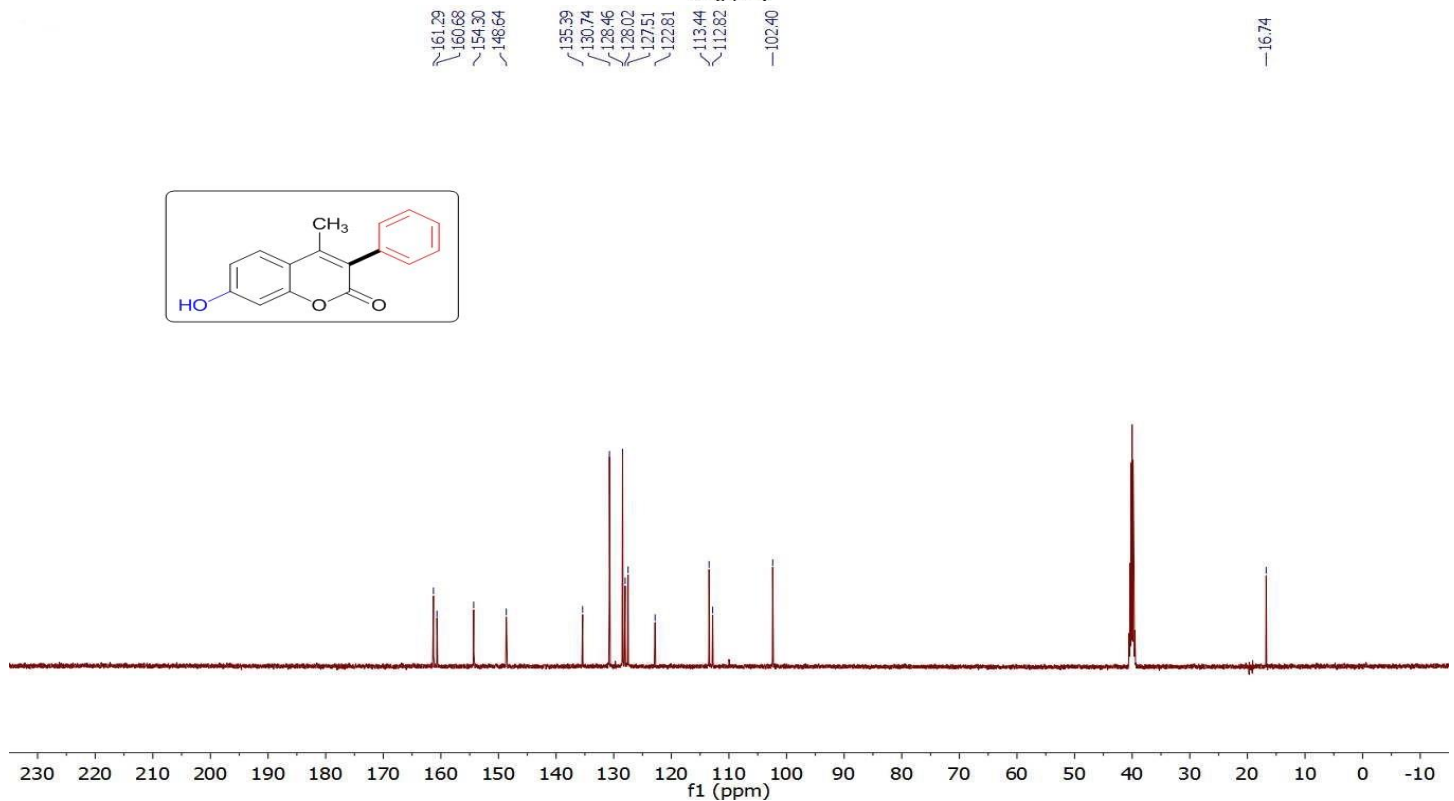
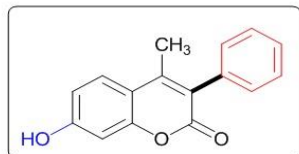
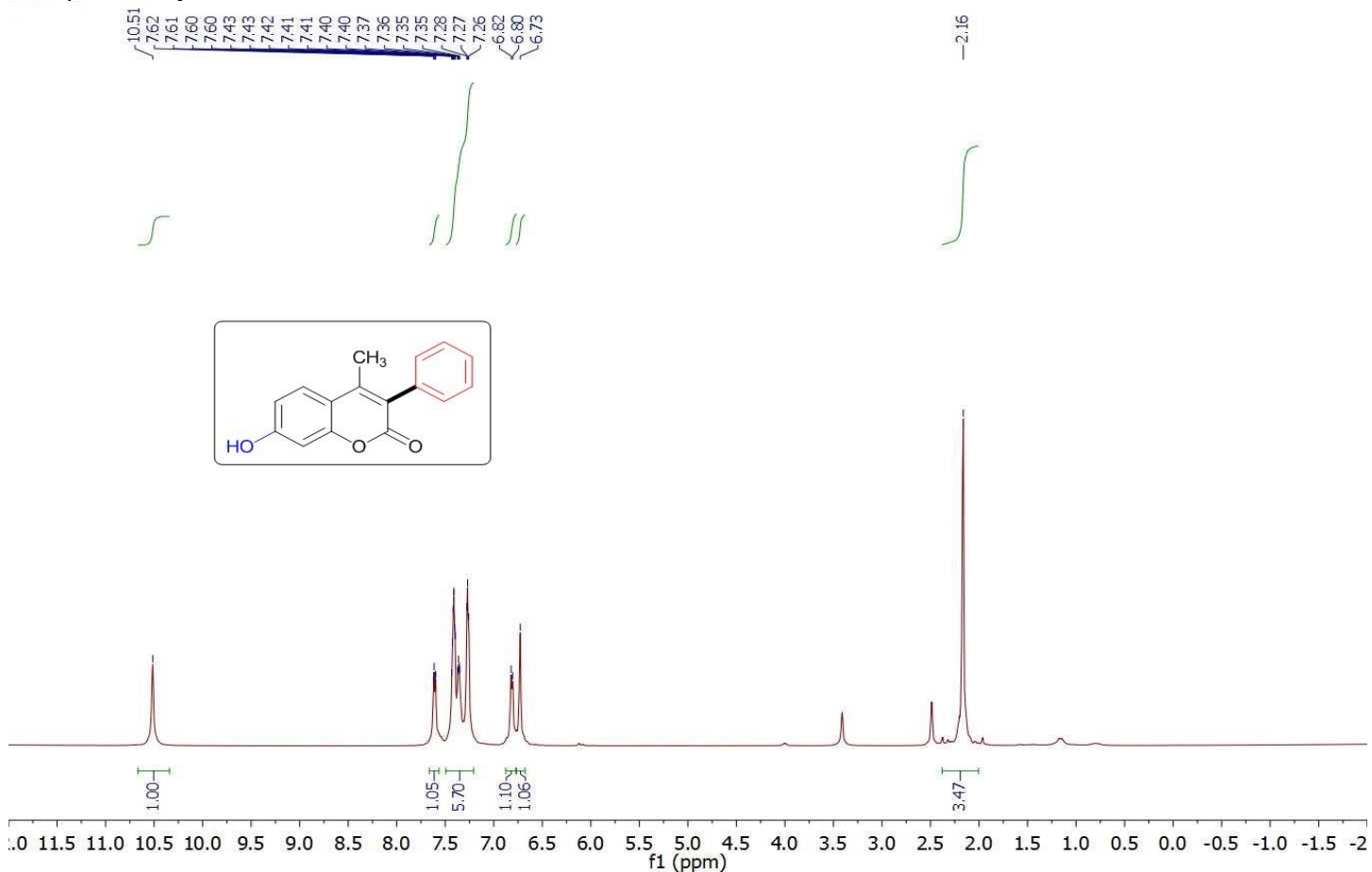
Compound 3o



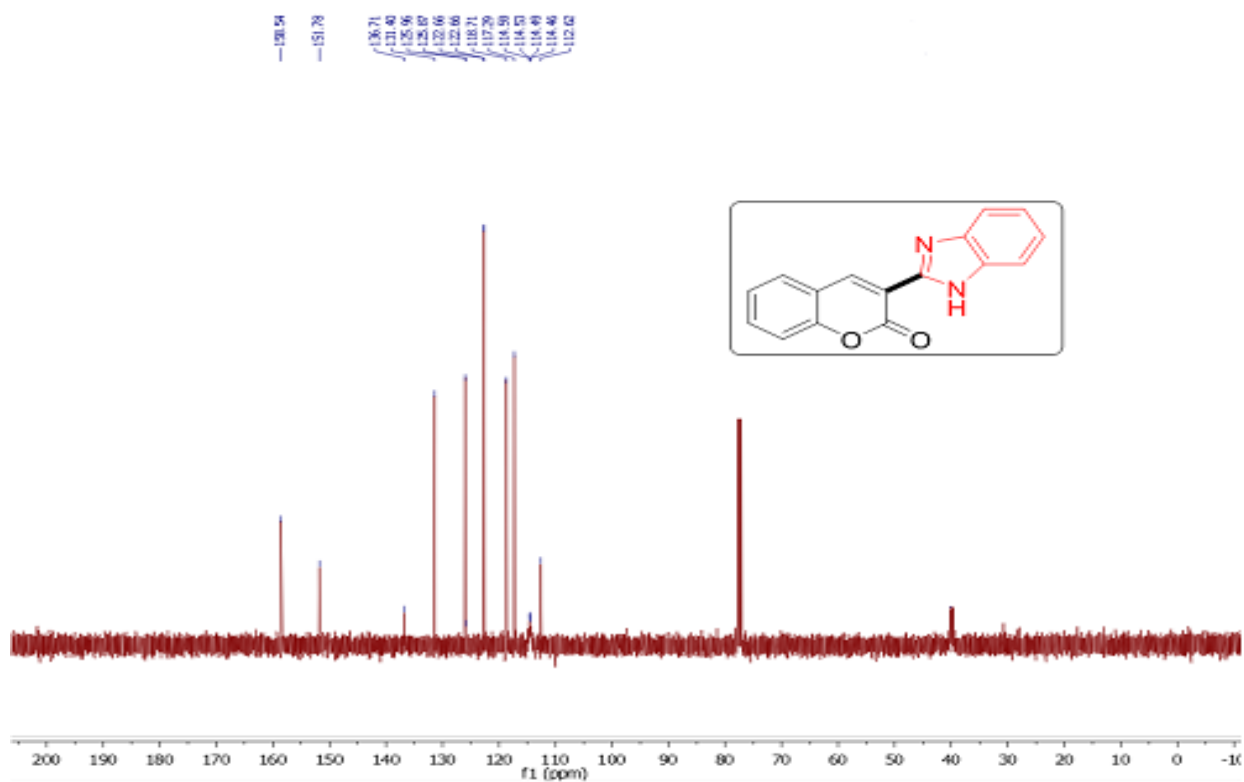
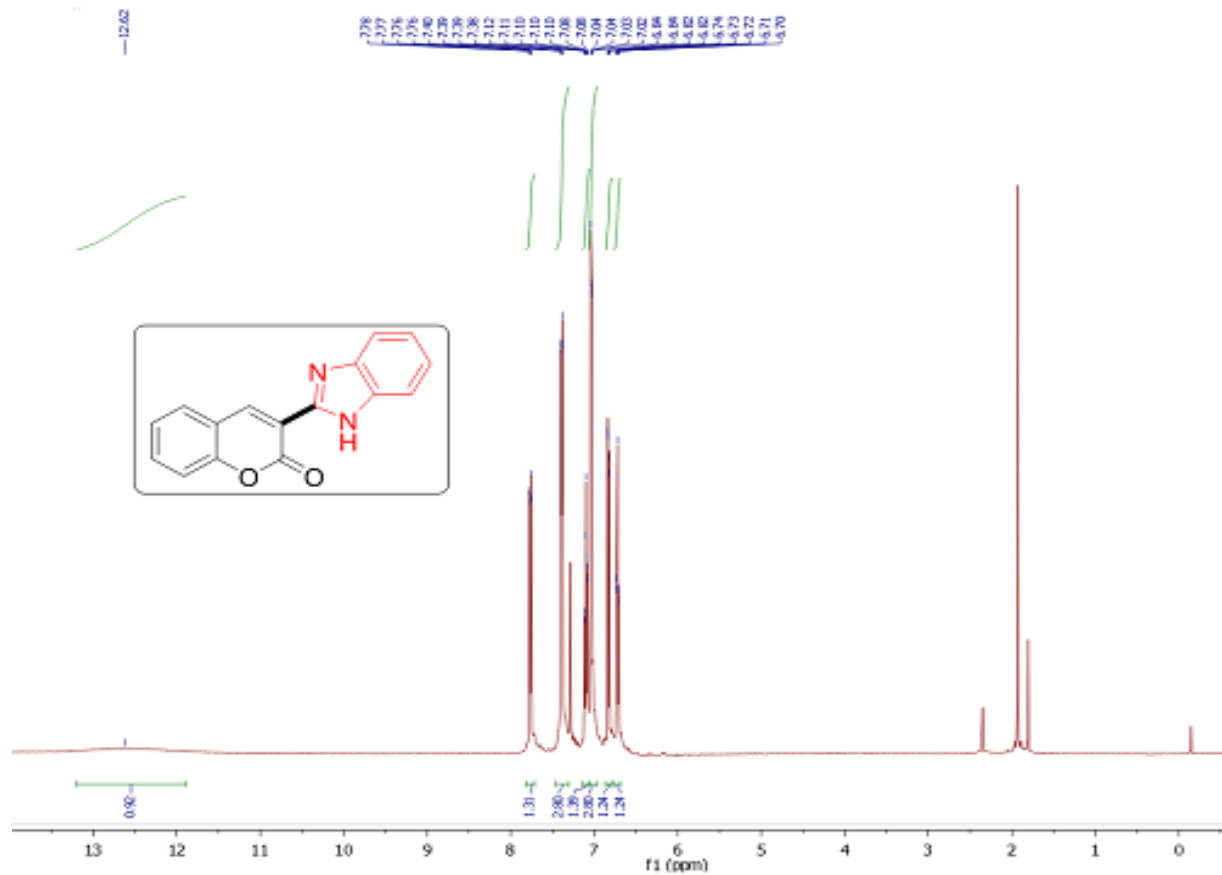
Compound 3p



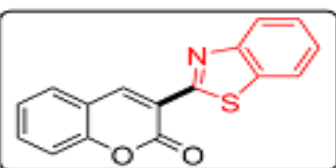
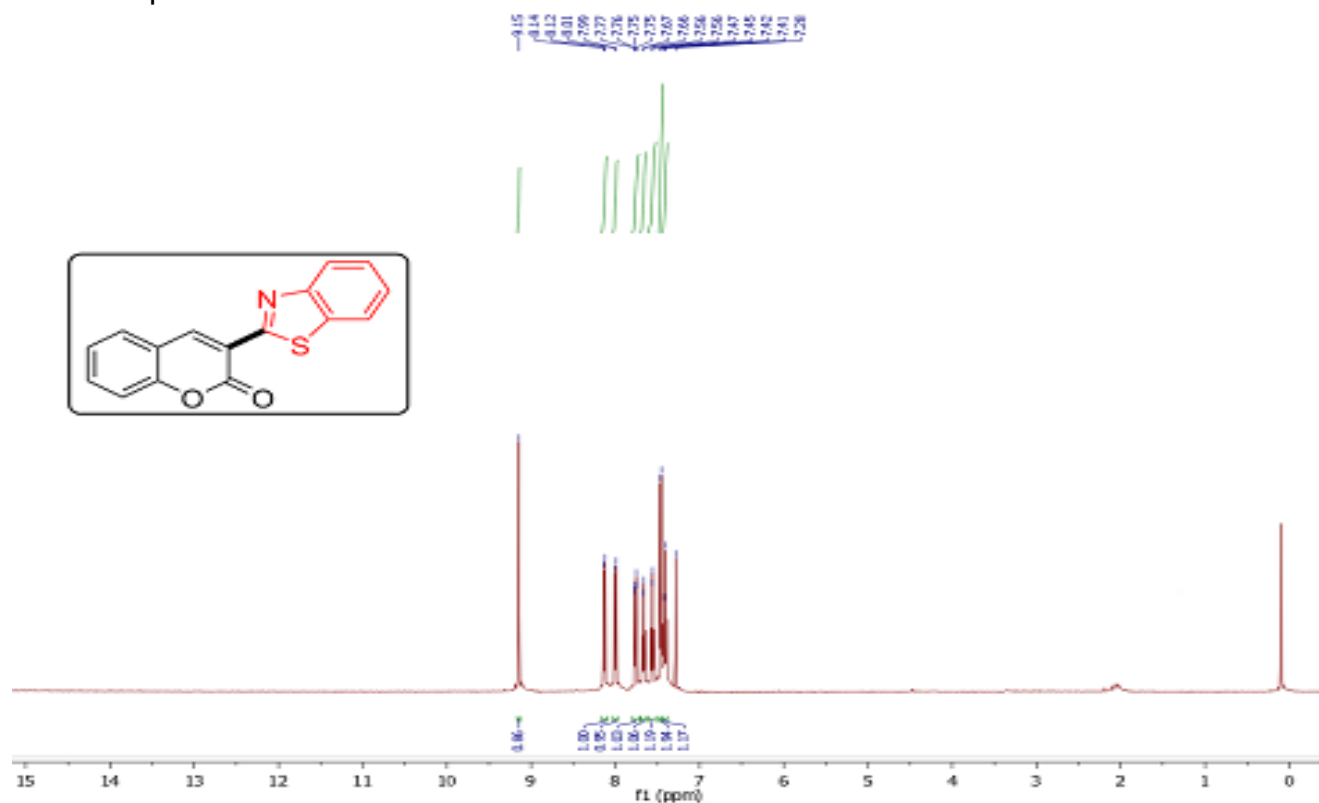
Compound **3q**



Compound **3s**



Compound 3t



156.04
155.00
150.87
150.03
141.78
136.65
133.33
129.47
128.62
128.51
128.29
122.78
121.78
120.08
118.08
116.78

