



Supporting Information

for

Synthesis of (Z)-3-[amino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-ones using an Eschenmoser coupling reaction

Lukáš Marek, Lukáš Kolman, Jiří Váňa, Jan Svoboda and Jiří Hanusek

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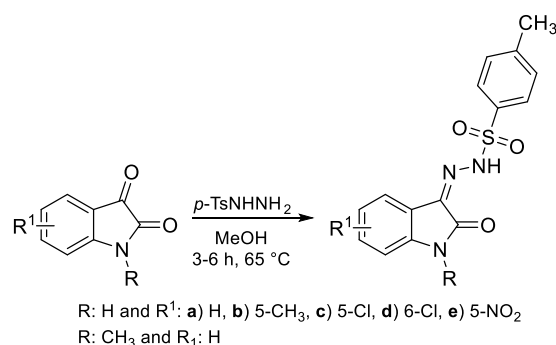
Synthetic procedures, characterization data and copies of spectra

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Synthesis of starting 3-bromooxindoles 1a–e

1. step:



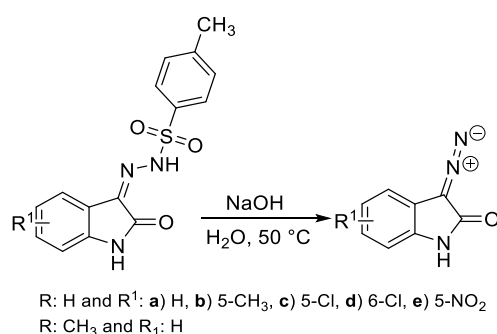
The corresponding isatin (*n*; mmol) was suspended in CH₃OH (*V*_{CH₃OH}; mL) at 50 °C. *p*-Toluenesulfonylhydrazine (1.1 equiv) was added in one portion and the reaction mixture was refluxed (*t*_{reflux}) and the reaction progress was monitored using TLC. The reaction mixture was concentrated to half of its volume and cooled to 0–5 °C. The precipitated mixture of (*Z*)/(*E*)-isomers of isatin *p*-tosylhydrazone was filtered off and washed with cold CH₃OH.

Table S1 – Molar and weight amounts of reactants and product yields and their melting points.

R	R ¹	<i>n</i> _{isatin} [mmol]	<i>m</i> _{isatin} [g]	<i>V</i> _{CH₃OH} [mL]	<i>t</i> _{reflux} [h]	Yield [g]	Yield [%]	m.p. [°C]
H	H	24	3.53	100	3	6.2	82	199-202
H	5-CH ₃	12.5	2.01	40	6	3.3	79	194-196
H	5-Cl	41	7.44	135	3	12.8	89	232-233
H	6-Cl	14	2.54	65	3	3.7	75	210-211
H	5-NO ₂	20	3.84	90*	4	5.4	75	209-210
CH ₃	H	30	5.00	170*	1	8.4	83	194-196

*In THF

2. step:



The corresponding isatin *p*-tosylhydrazone (*n*; mmol) was suspended in water (10 mL per each 1 mmol of *p*-tosylhydrazone) and 10% aqueous NaOH (4.5 mmol per each 1 mmol of tosylhydrazone; 4.5 equiv) was added in one portion. The suspension was stirred at 50 °C until dissolution of all solid material accompanied by a change of the solution color to orange (*t*_{heating}). Then, the reaction mixture was cooled and extracted with EtOAc (4 × 75 mL). The combined organic layers were washed with water (50 mL), dried with anhydrous Na₂SO₄, and evaporated. Yields and ¹H NMR data are given below.

Table S2 – Molar and weight amounts of reactants and product yields and their melting points.

R	R¹	<i>n</i>_{hydrazone} [mmol]	<i>m</i>_{hydrazone} [g]	<i>m</i>_{NaOH} [g]	<i>t</i>_{heating} [h]	Yield [g]	Yield [%]	m.p. [°C]
H	H	15.8	5.00	2.84	3	2.4	96	165-167
H	5-Me	9.1	3.00	1.63	3	1.5	96	190-194
H	5-Cl	20.0	7.00	3.60	2	3.5	89	219-224
H	6-Cl	8.6	3.00	1.55	2	1.5	92	192 (dec.)
H	5-NO ₂	13.9	5.00	2.50	5	2.3	81	255 (dec.)
CH ₃	H	6.0	2.00	1.10	3	0.9	86	83-85

3-Diazooxindole:

¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.66 (bs, 1H, NH), 7.39 (d, *J* 7.6 Hz, 1H, ArH), 7.09 (t, *J* 7.6 Hz, 1H, ArH), 6.99 (t, *J* 7.6 Hz, 1H, ArH), 6.91 (d, *J* 7.8 Hz, 1H, ArH). ¹³C NMR (100 MHz, DMSO-*d*₆) δ: ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 167.9 (C=O), 132.7, 125.2, 121.3, 119.3, 117.1, 110.0, 60.3 (>C=N⁺=N⁻) correspond to Ref. [1].

5-Methyl-3-diazooxindole:

¹H NMR (500 MHz, DMSO-*d*₆) δ: 10.53 (bs, 1H, NH), 7.18 (s, 1H, ArH), 6.88 (d, *J* 8.0, 1H, ArH), 6.79 (d, *J* 7.9 Hz, 1H, ArH). ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 168.1 (C=O), 130.5, 130.4, 125.9, 119.7, 117.2, 109.8, 60.1 (>C=N⁺=N⁻), 20.9 (CH₃).

5-Chloro-3-diazooxindole:

¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.77 (bs, 1H, NH), 7.53 (d, *J* 2.1 Hz, 1H, ArH), 7.09 (dd, *J* 8.3 and 2.2 Hz, 1H, ArH), 6.87 (d, *J* 8.3 Hz, 1H, ArH). ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 167.4 (C=O), 131.4 (C_{quart}), 125.3 (C_{quart}), 124.7 (CH), 119.1 (C_{quart}), 119.0 (CH), 111.1 (CH), 60.8 (>C=N⁺=N⁻) correspond to Ref. [2]

6-Chloro-3-diazooxindole:

¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.79 (bs, 1H, NH), 7.38 (d, *J* 8.2 Hz, 1H, ArH), 7.00 (dd, *J* 8.2 and 1.6 Hz, 1H, ArH), 6.89 (d, *J* 1.5 Hz, 1H, ArH) corresponds to Ref [3]. ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 167.7 (C=O), 133.7, 129.4, 121.0, 120.4, 116.1, 110.0, 60.6 (>C=N⁺=N⁻).

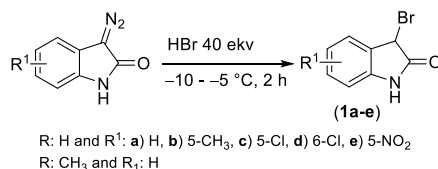
5-Nitro-3-diazooxindole:

¹H NMR (400 MHz, DMSO-*d*₆) δ: 8.44 (d, *J* 2.3 Hz, 1H, ArH), 8.01 (dd, *J* 8.7 and 2.4 Hz, 1H, ArH), 7.05 (d, *J* 8.7 Hz, 1H, ArH), 3.62 (vbs, 1H, NH + H₂O) corresponds to Ref [2]. ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 167.8 (C=O), 141.8, 138.5, 121.6, 118.5, 114.9, 109.7, 61.7 (>C=N⁺=N⁻).

3-Diazo-*N*-methyloxindole:

¹H NMR (400 MHz, CDCl₃) δ: 7.16-7.24 (m, 2H, ArH), 7.10 (t, *J* 7.5 Hz, 1H, ArH), 6.93 (d, *J* 8.0 Hz, 1H, ArH), 3.34 (s, 3H, CH₃) corresponds to Ref. [4].

3. step:



The corresponding well-grinded diazooxindole (*n*; mmol) was added in small portions to a stirred cold (−10 °C) aqueous 46% HBr (40 equiv.; *V*_{HBr}) solution until the nitrogen evolution ceased. Then, the reaction mixture was stirred for the next 2 hours at room temperature to complete the reaction (monitoring by TLC; SiO₂ plates, CHCl₃/CH₃OH 9:1). The final suspension was filtered and washed with water until neutral pH of the filtrate and the solid residue was dried.

Table S3 – Molar and weight amounts of reactants and product yields and their melting points.

	R	R ¹	<i>n</i> [mmol]	<i>m</i> [g]	<i>V</i> _{HBr} [ml]	Yield [g]	Yield [%]	M. p. [°C]
1a	H	H	15.1	2.4	72	2.6	82	162-163
1b	H	5-Me	15.0	2.6	68	2.4	71	173-174
1c	H	5-Cl	13.0	2.5	61	3.1	96	179 (dec.)
1d	H	6-Cl	10.5	2.0	50	2.4	92	162-163
1e	H	5-NO ₂	10.8	2.2	51	3.1	91	205 (dec.)
1a'	CH ₃	H	3.6	0.6	30	0.6	76	123-124

3-Bromooxindole (1a)

¹H NMR (500 MHz, DMSO-*d*₆) δ: 10.79 (bs, 1H, NH), 7.33 (d, *J* = 7.59 Hz, 1H, ArH), 7.29-7.25 (m, 1H, ArH), 7.02 (td, *J* = 1.02 and 7.59 Hz, 1H, ArH), 6.85 (d, *J* = 7.59 Hz, 1H, ArH), 5.72 (s, 1H, CH). ¹³C NMR-APT (125 MHz, CDCl₃) δ (ppm) 174.7 (C=O), 140.9 (C_{quart}), 130.4 (CH), 126.6 (C_{quart}), 126.3 (CH), 123.4 (CH), 110.6 (CH), 38.9 (CH) correspond to Ref. [1]

3-Bromo-5-methyloxindole (1b)

¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.67 (bs, 1H, NH), 7.14 (s, 1H, Ar-H₄), 7.06 (d, *J* 7.9 Hz 1H, Ar-H), 6.74 (d, *J* 7.9 Hz, 1H, Ar-H), 5.66 (s, 1H, CH), 2.25 (s, 3H, CH₃). ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 173.7 (C=O), 140.0 (C_{quart}), 131.4 (C_{quart}), 130.6 (CH), 127.2 (C_{quart}), 126.6 (CH), 110.0 (CH), 40.8 (CH), 20.6 (CH₃).

3-Bromo-5-chlorooxindole (1c)

¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.90 (bs, 1H, NH), 7.38 (s, 1H, Ar-H₄), 7.31 (dd, *J* 8.3 and 2.0 Hz, 1H, Ar-H₆), 6.87 (d, *J* 8.3 Hz, 1H, Ar-H₇), 5.69 (s, 1H, CH). ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 173.4 (C=O), 141.3 (C_{quart}), 130.1 (CH), 129.2 (C_{quart}), 126.2 (C_{quart}), 126.0 (CH), 111.8 (CH), 39.6 (CH).

3-Bromo-6-chlorooxindole (1d)

¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.92 (bs, 1H, NH), 7.35 (d, *J* = 8.0 Hz, 1H, Ar-H₄), 7.07 (dd, *J* = 8.0 and 1.7 Hz, 1H, Ar-H₅), 6.88 (d, *J* = 1.4 Hz, 1H, Ar-H₇), 5.71 (s, 1H, CH). ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 173.7 (C=O), 143.9 (C_{quart}), 134.4 (C_{quart}), 127.5 (CH), 126.1 (C_{quart}), 122.1 (CH), 110.3 (CH), 39.6 (CH).

3-Bromo-5-nitrooxindole (1e)

¹H NMR (500 MHz, DMSO-*d*₆) δ: 11.49 (bs, 1H, NH), 8.22 (dd, *J* = 8.7 and 1.8 Hz, 1H, ArH), 8.18 (s, 1H, ArH), 7.06 (d, *J* = 8.7 Hz, 1H, ArH), 5.82 (s, 1H, CH). ¹³C NMR (125 MHz, DMSO-*d*₆) δ: 174.1 (C=O), 148.8 (C_{quart}), 142.6 (C_{quart}), 128.4 (C_{quart}), 127.3 (CH), 121.6 (CH), 110.6 (CH), 38.7 (CH).

3-Bromo-*N*-methyloxindole (1a')

¹H NMR (400 MHz, CDCl₃) δ: 7.41 (d, *J* = 7.5 Hz, 1H, ArH), 7.35 (t, *J* = 7.8 Hz, 1H, ArH), 7.11 (t, *J* = 7.3 Hz, 1H, ArH), 6.83 (d, *J* = 7.8 Hz, 1H, ArH), 5.27 (s, 1H, CH), 3.23 (s, 3H, CH₃) corresponds to Ref. [1].

3-Hydroxy-*N*-methyloxindole

N-Methylisatin (2.5 g, 15.5 mmol) was suspended in CH₃OH (45 mL) and NaBH₄ (0.75 g, 19.8 mmol) was added in one portion at 0 °C under vigorous stirring. After 5 min, the homogeneous decolorized solution was evaporated at 30 °C under vacuum and the residue was extracted with EtOAc (100 mL). The organic phase was washed with water (2 × 20 mL) and brine (40 mL), dried with anhydrous Na₂SO₄, and evaporated. The crude product was crystallized from CH₃OH (5 mL) by cooling to -20 °C. White crystals (1.9 g, 75%); m.p. 151-152 °C; ¹H NMR (400 MHz, CDCl₃) δ 7.47 (d, *J* 7.3 Hz, 1H, Ar-H), 7.33 (t, *J* 7.7 Hz, 1H, Ar-H), 7.11 (t, *J* 7.5 Hz, 1H, Ar-H), 6.82 (d, *J* 7.8 Hz, 1H, Ar-H), 5.12 (s, 1H, CH), 4.71 (bs, 1H, OH), 3.18 (s, 3H, CH₃); ¹³C NMR (100 MHz, CDCl₃) δ (ppm): 177.4, 143.7, 129.7, 127.1, 125.1, 123.2, 108.4, 69.8, 26.2 corresponds to Ref. [5].

3-Hydroxyoxindole

Prepared from isatin (4.56 g, 31 mmol) and NaBH₄ (1.50 g, 19.8 mol) in CH₃OH (60 mL) at 0 °C using the same procedure as for 3-hydroxy-*N*-methyloxindole. The solid residue was triturated with Et₂O (15 mL) at 0 °C. White crystals (2.3 g, 50%); m. p. 165-167 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.23 (bs, 1H, NH), 7.28 (d, *J* = 7.3 Hz, 1H, ArH), 7.20 (t, *J* = 7.6 Hz, 1H, ArH), 6.95 (t, *J* = 7.4 Hz, 1H, ArH), 6.78 (d, *J* = 7.6 Hz, 1H, ArH), 6.18 (d, *J* = 7.6 Hz, 1H, CH), 4.82 (d, *J* = 7.6 Hz, 1H, OH). ¹³C NMR (100 MHz, DMSO-*d*₆) δ: 178.1 (C=O), 142.3, 129.4, 129.1, 124.9, 121.7, 109.6, 69.3 correspond to Ref. [6]

3-Chlorooxindole (1f)

trans-β-Nitrostyrene (3.2 g, 14 mmol) was dissolved in DCM (50 mL) and acetyl chloride (2.1 mL, 30 mmol) was added in one portion. Anhydrous FeCl₃ (4.6 g, 29 mmol) was added in three

portions at 25 °C during 15 min and the resulting mixture was stirred for 5 hours. Then, the reaction mixture was diluted with 3% aq. HCl (200 mL) and extracted with DCM (3 × 50 mL). The organic layer was washed with water (2 × 50 mL), brine (50 mL), dried with anhydrous Na₂SO₄, and evaporated. The crude product was dissolved in DCM (70 mL), evaporated with SiO₂ (15 g) and purified using preparative flash liquid chromatography (silica gel cartridge 40 g, mobile phase CHCl₃/CH₃OH, gradient 0–5% CH₃OH during 15 min). The pure product was obtained by crystallization from hot toluene. White crystals (0.8 g, 35%); m.p. 161-162 °C; ¹H NMR (400 MHz, CDCl₃) δ 10.77 (bs, 1H, NH), 7.36 (d, *J* 7.4 Hz, 1H, Ar-H), 7.29 (t, *J* 7.6 Hz, 1H, Ar-H), 7.03 (td, *J* 7.6 and 0.8 Hz, 1H, Ar-H), 6.87 (d, *J* 7.8 Hz, 1H, Ar-H), 5.57 (s, 1H, CH). ¹³C NMR (100 MHz, CDCl₃) δ (ppm): 173.3, 142.5, 130.4, 126.6, 125.7, 122.4, 110.2, 52.3 corresponds to Ref. [7].

Diethyl (2-oxoindolin-3-yl)phosphate (1g)

Isatin (4.4 g, 30 mmol) was suspended in dry acetonitrile (50 mL) and diethyl phosphite (4.15 g; 30 mmol) and anhydrous well-grinded K₂CO₃ (0.44 g, 3 mmol) were added under argon atmosphere. The reaction mixture was stirred at 60 °C for 1.5 h and then filtered. Residual salts were washed with acetonitrile (2 × 10 mL) and the combined filtrates were evaporated. The crude oily product (8.5 g, 99%) was dissolved in EtOAc (40 mL), evaporated with neutral alumina (30 g) and submitted to preparative flash chromatography (silica gel cartridge 80 g, mobile phase *n*-hexane/EtOAc, gradient 0–100% EtOAc during 20 min). The resulting colorless oil (7.1 g, 84%) slowly solidified to white solid with m.p. 86 – 88 °C. ¹H NMR (500 MHz, CDCl₃) δ 9.59 (bs, 1H, NH), 7.50 (d, *J* 7.5 Hz, 1H, Ar-H), 7.25 (t, *J* 7.8 Hz, 1H, Ar-H), 7.02 (t, *J* 7.5 Hz, 1H, Ar-H), 6.91 (d, *J* 7.8 Hz, 1H, Ar-H), 5.58 (d, *J* 12.8 Hz, 1H, C-H), 4.16-4.34 (m, 4H, 2× OCH₂), 1.30-1.40 (m, 6H, 2× CH₃) correspond to [8].

(Z)-3-[Amino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (5aa)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–20% EtOAc during 20 min). Yellow crystals (623 mg, 88%); mp 239 – 240 °C (EtOH) (Ref. [1] gives 238-239 °C); ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.36 (bs, 1H, NH), 9.45 (bd, *J* = 3.2 Hz, 1H, ½NH₂), 8.03 (bd, *J* = 2.8 Hz, 1H, ½NH₂), 7.55-7.64 (m, 3H, Ar-H), 7.48-7.55 (m, 2H, Ar-H), 6.76-6.88 (m, 2H, Ar-H), 6.52 (t, 1H, *J* = 7.6 Hz, Ar-H₆), 6.01 (d, *J* = 7.7 Hz, 1H, Ar-H₄) correspond to Ref. [1].

(Z)-3-[Amino(4-methoxyphenyl)methylidene]-1,3-dihydro-2H-indol-2-one (5ab)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–30% EtOAc during 20 min). Yellow crystals (655 mg, 82%); mp 229.5 – 231 °C (toluene/CH₃OH) (mp 178-180 °C given in Ref. [32] is probably interchanged with thiazole derivative with mp 230-233 °C also cited therein); ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.34 (bs, 1H, NH), 9.47 (bd, *J* = 3.3 Hz, 1H, ½NH₂), 7.91 (bd, *J* = 3.0 Hz, 1H, ½NH₂), 7.48 (AA'XX', *J* = 8.7 Hz, 2H, Ar-H), 7.12 (AA'XX', *J* = 8.7 Hz, 2H, Ar-H), 6.72-6.88 (m, 2H, Ar-H), 6.56 (t, *J* = 7.5 Hz, 1H, Ar-H), 6.24 (d, *J* = 7.7 Hz, 1H, Ar-H₄), 3.85 (s, 3H, OCH₃) correspond to Ref. [9].

(Z)-3-[Amino(4-chlorophenyl)methylidene]-1,3-dihydro-2H-indol-2-one (5ac)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–40% EtOAc during 20 min). Yellow crystals (754 mg, 93%); mp 237.5 – 238.5 °C (toluene/CH₃OH) (Ref. [1] gives 234-236 °C dec.); ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.39 (bs, 1H, NH), 9.41 (bd, *J* = 3.2 Hz, 1H, ½NH₂), 8.03 (bs, 1H, ½NH₂), 7.66 (AA'XX', *J* = 8.4 Hz, 2H, Ar-H), 7.55 (AA'XX', *J* = 8.4 Hz, 2H, Ar-H), 6.78-6.88 (m, 2H, Ar-H), 6.57 (t, *J* = 7.5 Hz, 1H, Ar-H), 6.06 (d, *J* = 7.8 Hz, 1H, Ar-H₄), corresponds to Ref. [1].

(Z)-3-[Amino(4-iodophenyl)methylidene]-1,3-dihydro-2H-indol-2-one (5ac')

The crude product was purified using column chromatography (silica gel, EtOAc). Yellow solid 500 mg, 93%); mp 242 – 245 °C; ¹H NMR (500 MHz, CDCl₃) δ 9.47 (bs, 1H, ½NH₂), 7.90 (d, *J* = 8.3 Hz, 1H, Ar-H), 7.86 (bs, 1H, NH), 7.69 (AA'XX', *J* = 8.4 Hz, 2H, Ar-H), 7.46 (AA'XX', *J* = 8.4 Hz, 2H, Ar-H), 7.32 (d, *J* = 8.3 Hz, 1H, Ar-H), 6.99 (t, *J* = 7.6 Hz, 1H, Ar-H), 6.89 (d, *J* = 7.7 Hz, 1H, Ar-H), 6.72 (t, *J* = 7.6 Hz, 1H, Ar-H), 6.36 (t, *J* = 7.6 Hz, 1H, Ar-H₄), 5.13 (bs, 1H, ½NH₂); ¹³C NMR (125 MHz, CDCl₃) δ (ppm): 170.7, 158.0, 157.9, 138.5, 135.6, 134.7, 132.5, 129.6, 124.9, 123.8, 120.8, 118.5, 109.1, 96.9; HRMS: calcd. for C₁₅H₁₂IN₂O [M+H⁺]: calcd. 362.9989, found 362.9989; Elemental analysis: calcd. C: 49.75, H: 3.06, N: 7.73, I: 35.00; found C: 49.64, H: 3.01, N: 7.68, I: 34.85 (for sample cryst. from CH₃OH/toluene). IR (cm⁻¹): 3349 (s), 3261 (s), 1615 (s).

(Z)-3-[Amino(4-trifluoromethylphenyl)methylidene]-1,3-dihydro-2H-indol-2-one (5ad)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–80% EtOAc during 20 min). Yellow crystals

(884 mg, 97%); mp 244 – 245 °C (toluene) (Ref. [1] gives 233-237 °C dec.); ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.43 (bs, 1H, NH), 9.43 (bs, 1H, ½NH₂), 8.09 (bs, 1H, ½NH₂), 7.97 (AA'XX', *J* = 8.0 Hz, 2H, Ar-H), 7.77 (AA'XX', *J* = 7.9 Hz, 2H, Ar-H), 6.78-6.89 (m, 2H, Ar-H), 6.55 (t, 1H, *J* = 7.3 Hz, Ar-H₆), 5.95 (d, *J* = 7.7 Hz, 1H, Ar-H₄) correspond to Ref. [1].

(Z)-3-[Amino(phenyl)methylidene]-5-methyl-1,3-dihydro-2H-indol-2-one (5ba)

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane 90:10). Beige solid (526 mg, 84%); mp 206 – 208 °C; ¹H NMR (500 MHz, CDCl₃) δ: 9.51 (bs, 1H, ½NH₂), 8.37 (bs, 1H, NH), 7.47 – 7.68 (m, 5H, Ar-H), 6.80 (d, *J* = 8.1 Hz, 1H, Ar-H), 6.10 (s, 1H, Ar-H₄), 5.23 (bs, 1H, ½NH₂), 2.08 (s, 3H, CH₃). ¹³C NMR (125 MHz, CDCl₃) δ (ppm): 171.2, 159.3, 136.0, 133.5, 130.4, 129.6, 129.1, 127.8, 124.7, 124.0, 119.2, 108.8, 95.9, 21.4; HRMS: calcd. for C₁₆H₁₅N₂O [M+H⁺]: calcd. 251.1179, found 251.1175; Elemental analysis: calcd. C: 76.78, H: 5.64, N: 11.19; found C: 76.70, H: 5.65, N: 11.16 (for sample cryst. from MeCN/*n*-hexane). IR (cm⁻¹): 3354 (s), 3265 (s), 1643 (s).

(Z)-3-[Amino(4-methoxyphenyl)methylidene]-5-methyl-1,3-dihydro-2H-indol-2-one (5bb)

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane/MeCN 75:20:5). Yellow solid (482 mg, 69%); mp 213 – 215 °C; ¹H NMR (500 MHz, CDCl₃) δ: 9.52 (bs, 1H, ½NH₂), 8.27 (bs, 1H, NH), 7.53 (AA'XX', *J* = 8.6 Hz, 2H, Ar-H), 7.04 (AA'XX', *J* = 8.7 Hz, 2H, Ar-H), 6.79 (s, 2H, Ar-H_{6,7}), 6.32 (s, 1H, Ar-H₄), 5.19 (bs, 1H, ½NH₂), 3.91 (s, 3H, CH₃); ¹³C NMR (125 MHz, CDCl₃) δ (ppm): 171.2, 161.3, 159.4, 133.4, 129.6, 129.5, 128.3, 124.9, 123.9, 119.1, 114.3, 108.7, 95.7, 55.5, 21.4; HRMS: calcd. for C₁₆H₁₇N₂O₂ [M+H⁺]: calcd. 281.1285, found 281.1279; Elemental analysis: calcd. C: 72.84, H: 5.75, N: 9.99; found C: 72.44, H: 5.60, N: 9.58 (for sample cryst. from MeCN/EtOAc). IR (cm⁻¹): 3374 (s), 3282 (s), 1609 (s).

(Z)-3-[Amino(4-chlorophenyl)methylidene]-5-methyl-1,3-dihydro-2H-indol-2-one (5bc)

The crude product was purified using column chromatography (silica gel, EtOAc/CH₃OH 99:1). Yellow solid (520 mg, 73%); mp 246 – 247.5 °C; ¹H NMR (500 MHz, CDCl₃) δ: 9.49 (bs, 1H, ½NH₂), 8.27 (bs, 1H, NH), 7.53 (s, 4H, Ar-H), 6.80 (s, 2H, Ar-H), 6.18 (s, 1H, Ar-H₄), 5.14 (bs, 1H, ½NH₂), 2.12 (s, 3H, CH₃); ¹³C NMR (125 MHz, CDCl₃) δ (ppm): 171.1, 157.7, 136.6, 134.3, 133.6, 129.84, 129.45, 129.37, 124.38, 124.29, 119.1, 108.9, 96.2, 21.4; HRMS: calcd. for C₁₆H₁₄ClN₂O [M+H⁺]: calcd. 285.0789, found 285.0784; Elemental analysis: calcd. C: 67.49, H: 4.60, N: 9.84, Cl: 12.45; found C: 67.70, H: 4.57, N: 9.58, Cl: 12.29 (for sample cryst. from MeCN/EtOAc). IR (cm⁻¹): 3362 (s), 3269 (s), 1640 (s).

(Z)-3-[Amino(4-trifluoromethylphenyl)methylidene]-5-methyl-1,3-dihydro-2H-indol-2-one (5bd)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–80% EtOAc during 15 min). Yellow solid (891 mg, 93%); mp 234 – 236 °C; ¹H NMR (500 MHz, CDCl₃) δ: 10.31 (bs, 1H, NH), 9.38

(bd, $J = 4.0$ Hz, 1H, $\frac{1}{2}\text{NH}_2$), 8.06 (bd, $J = 4.0$ Hz, 1H, $\frac{1}{2}\text{NH}_2$), 7.97 (AA'XX', $J = 8.1$ Hz, 2H, Ar-H), 7.75 (AA'XX', $J = 8.0$ Hz, 2H, Ar-H), 6.65-6.61 (m, 2H, Ar-H_{6,7}), 5.68 (s, 1H, Ar-H₄), 1.93 (s, 3H, CH₃); ^{13}C NMR (125 MHz, CDCl₃) δ (ppm): 170.3, 158.2, 139.7, 134.3, 130.4 (q, $^2J_{\text{C-F}} = 32.0$ Hz), 129.1, 127.9, 126.0 (q, $^3J_{\text{C-F}} = 3.7$ Hz), 124.4, 124.1 (q, $^1J_{\text{C-F}} = 272$ Hz), 123.4, 117.9, 108.9, 94.5, 21.2.; HRMS: calcd. for C₁₇H₁₄F₃N₂O [M+H⁺]: calcd. 319.1058, found 319.1059; Elemental analysis: not measured due to fluorine content damaging column. IR (cm⁻¹): 3497 (w), 3441 (s), 1652 (s), 1605 (s).

(Z)-3-[Amino(phenyl)methylidene]-5-chloro-1,3-dihydro-2H-indol-2-one (5ca)

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane/THF 70:25:5). Yellow solid (373 mg, 97%); mp 192.5 – 193 °C; ^1H NMR (500 MHz, CDCl₃) δ 9.60 (bs, 1H, $\frac{1}{2}$ NH₂), 9.07 (bs, 1H, CONH), 7.45 – 7.68 (m, 5H, Ar-H), 6.92 (dd, $J = 8.2, 1.9$ Hz, 1H, Ar-H₆), 6.83 (d, $J = 8.2$ Hz, 1H, Ar-H₇), 6.24 (d, $J = 1.7$ Hz, 1H, Ar-H₄), 5.42 (s, 1H, $\frac{1}{2}$ NH₂); ^{13}C NMR (125 MHz, CDCl₃) δ 171.2, 160.6, 135.3, 134.1, 130.9, 129.3, 127.6, 126.1, 125.8, 123.0, 118.4, 109.9, 95.3; HRMS: calcd. for C₁₅H₁₂ClN₂O [M+H⁺]: calcd. 271.0633, found 271.0652; Elemental analysis: calcd. C: 66.55, H: 4.10, N: 10.35, Cl: 13.10; found C: 66.30, H: 3.99, N: 10.18, Cl: 13.05 (for sample cryst. from *n*-hexane/EtOAc). IR (cm⁻¹): 3356 (m), 3271 (s), 1636 (s), 1612 (s).

(Z)-3-[Amino(4-methoxyphenyl)methylidene]-5-chloro-1,3-dihydro-2H-indol-2-one (5cb)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–50% EtOAc during 20 min). Yellow solid (658 mg, 73%); mp 235 – 237 °C (toluene/CH₃OH); ^1H NMR (400 MHz, DMSO-*d*₆) δ : 10.48 (bs, 1H, NH), 9.54 (bd, $J = 3.2$ Hz, 1H, $\frac{1}{2}\text{NH}_2$), 8.21 (bs, 1H, $\frac{1}{2}\text{NH}_2$), 7.49 (AA'XX', $J = 8.6$ Hz, 2H, Ar-H), 7.16 (AA'XX', $J = 8.6$ Hz, 2H, Ar-H), 6.85 (dd, $J = 8.2, 1.8$ Hz, 2H, Ar-H₆), 6.78 (d, $J = 8.2$ Hz, 1H, Ar-H₇), 6.13 (d, $J = 1.4$ Hz, 1H, Ar-H₄), 3.87 (s, 3H, OCH₃); ^{13}C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 170.1, 161.6, 161.0, 134.5, 129.6, 127.3, 126.8, 123.7, 121.5, 116.6, 114.4, 109.8, 93.0, 55.5; HRMS: calcd. for C₁₆H₁₄ClN₂O₂ [M+H⁺]: calcd. 301.0744, found 301.0744; Elemental analysis: calcd. C: 63.90, H: 4.36, N: 9.31, Cl: 11.79; found C: 63.70, H: 4.37, N: 9.31, Cl: 11.48 (cryst. from toluene/CH₃OH). IR (cm⁻¹): 3348 (s), 1605 (s).

(Z)-3-[Amino(4-chlorophenyl)methylidene]-5-chloro-1,3-dihydro-2H-indol-2-one (5cc)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–40% EtOAc during 20 min). Yellow solid (824 mg, 90%); mp 281.5 – 283 °C (EtOH/H₂O); ^1H NMR (500 MHz, DMSO-*d*₆) δ : 10.55 (bs, 1H, NH), 9.51 (bd, $J = 3.9$ Hz, 1H, $\frac{1}{2}\text{NH}_2$), 8.35 (bd, $J = 3.8$ Hz, 1H, $\frac{1}{2}\text{NH}_2$), 7.70 (AA'XX', $J = 8.5$ Hz, 2H, Ar-H), 7.57 (AA'XX', $J = 8.5$ Hz, 2H, Ar-H), 6.87 (dd, $J = 8.2, 2.1$ Hz, 2H, Ar-H₆), 6.80 (d, $J = 8.2$ Hz, 1H, Ar-H₇), 5.93 (d, $J = 2.0$ Hz, 1H, Ar-H₄); ^{13}C NMR (125 MHz, DMSO-*d*₆) δ (ppm): 170.2, 160.4, 135.4, 135.0, 134.1, 130.1, 129.5, 126.6, 124.0, 122.1, 116.7, 110.2, 93.5; HRMS: calcd. for C₁₅H₁₁Cl₂N₂O [M+H⁺]: calcd. 305.0249, found 305.0251; Elemental analysis: calcd. C: 59.04, H: 3.30, N: 9.18, Cl: 23.24; found C: 59.41, H: 3.17, N: 8.95, Cl: 22.92 (for sample cryst. from EtOH/H₂O). IR (cm⁻¹): 3460 (s), 1609 (s).

(Z)-3-[Amino(4-iodophenyl)methylidene]-5-chloro-1,3-dihydro-2H-indol-2-one (5cc')

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane 90:10). Yellow solid (520 mg, 87%); mp 257 – 258 °C; ¹H NMR (500 MHz, CDCl₃) δ (ppm): 9.59 (bs, 1H, ½NH₂), 7.99 (bs, 1H, NH), 7.93 (d, *J* = 8.2 Hz, 2H, AA'XX', Ar-H), 7.31 (d, *J* = 8.2 Hz, 2H, AA'XX', Ar-H), 6.95 (dd, *J* = 8.3 and 2.0 Hz, 1H, Ar-H), 6.81 (d, *J* = 8.3 Hz, 1H, Ar-H), 6.34 (d, *J* = 1.8 Hz, 1H, Ar-H₄); 5.25 (bs, 1H, ½NH₂); ¹³C NMR (125 MHz, CDCl₃) δ (ppm): 170.6, 159.0, 138.6, 134.6, 133.9, 129.3, 126.1, 125.8, 123.4, 118.4, 109.9, 97.4; HRMS: calcd. for C₁₅H₁₁ClIN₂O [M+H⁺]: calcd. 396.9599, found 396.9600; Elemental analysis: calcd. C: 45.43, H: 2.54, N: 7.06, Cl: 8.94, I: 32.00; found C: 45.12, H: 2.37; N: 6.85 (for sample cryst. from CH₃OH/*i*-PrOH). IR (cm⁻¹): 3339 (s), 1646 (s), 1603 (s).

(Z)-3-[Amino(4-trifluoromethylphenyl)methylidene]-5-chloro-1,3-dihydro-2H-indol-2-one (5cd)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–80% EtOAc during 15 min). Yellow solid (770 mg, 76%); mp 235 – 238 °C (toluene/CH₃OH); ¹H NMR (400 MHz, DMSO-*d*₆) δ: 10.58 (bs, 1H, NH), 9.52 (bs, 1H, ½NH₂), 8.41 (bs, 1H, ½NH₂), 8.00 (AA'XX', *J* = 8.0 Hz, 2H, Ar-H), 7.78 (AA'XX', *J* = 7.9 Hz, 2H, Ar-H), 6.87 (dd, *J* = 8.2, 1.7 Hz, 2H, Ar-H₆), 6.87 (d, *J* = 8.2 Hz, 1H, Ar-H₇), 5.76 (s, 1H, Ar-H₄); ¹³C NMR (125 MHz, DMSO-*d*₆) δ (ppm): 170.0, 159.6, 139.1, 135.0, 130.7 (q, ²*J*_{C-F} = 32.1 Hz), 129.0, 126.1 (q, ³*J*_{C-F} = 5.0 Hz), 124.0 (q, ¹*J*_{C-F} = 270 Hz), 123.8, 122.1, 116.5, 110.0, 93.5; HRMS: calcd. for C₁₆H₁₁ClF₃N₂O [M+H⁺]: calcd. 339.0512, found 339.0516; Elemental analysis: not measured due to fluorine content damaging column. IR (cm⁻¹): 3350 (s), 1651 (s), 1606 (s).

(Z)-3-[Amino(phenyl)methylidene]-6-chloro-1,3-dihydro-2H-indol-2-one (5da)

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane 80:20). Yellow solid (323 mg, 84%); mp 212 – 214 °C; ¹H NMR (500 MHz, CDCl₃) δ 9.51 (bs, 1H, ½ NH₂), 9.35 (bs, 1H, NH), 7.44 – 7.65 (m, 5H, Ar-H), 6.94 (d, *J* = 1.9 Hz, 1H, Ar-H₇), 6.63 (dd, *J* = 8.3, 1.9 Hz, 1H, Ar-H₅), 6.18 (d, *J* = 8.3 Hz, 1H, Ar-H₄), 5.39 (bs, 1H, ½ NH₂); ¹³C NMR (125 MHz, CDCl₃) δ 171.4, 159.9, 136.7, 135.6, 130.7, 129.3, 128.7, 127.7, 123.1, 120.4, 119.0, 109.6, 95.3; HRMS: calcd. for C₁₅H₁₂ClN₂O [M+H⁺]: calcd. 271.0633, found 271.0633; Elemental analysis: calcd. C: 66.55, H: 4.10, N: 10.35, Cl: 13.10, found C: 66.20, H: 3.95, N: 10.19, Cl: 12.89 (for sample cryst. from CH₃OH/toluene). IR (cm⁻¹): 3406 (m), 1609 (s).

(Z)-3-[Amino(4-methoxyphenyl)methylidene]-6-chloro-1,3-dihydro-2H-indol-2-one (5db)

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane 80:20). Yellow solid (523 mg, 87%); mp 229.5 – 231 °C; ¹H NMR (500 MHz, CDCl₃) δ: 9.50 (bs, 1H, ½NH₂), 8.38 (bs, 1H, NH), 7.50 (AA'XX', *J* = 8.7 Hz, 2H, Ar-H), 7.04 (AA'XX', *J* = 8.7 Hz, 2H, Ar-H), 6.90 (d, *J* = 1.9 Hz, 1H, Ar-H₇), 6.67 (dd, *J* = 8.3 and 1.9 Hz, 1H, Ar-H₅), 6.37 (d, *J* = 8.3 Hz, 1H, Ar-H₄), 5.28 (bs, 1H, ½NH₂), 3.91 (s, 3H, CH₃); ¹³C NMR (125 MHz, CDCl₃) δ (ppm): 170.9, 161.5, 160.0, 136.2, 129.4, 128.5, 127.8, 123.4, 120.5, 119.1, 114.5,

109.4, 95.0, 55.5. HRMS: calcd. for $C_{16}H_{14}ClN_2O_2$ [$M+H^+$]: calcd. 301.0738, found 301.0744; Elemental analysis: calcd. C: 63.90, H: 4.36, N: 9.31, Cl: 11.79; found C: 63.87, H: 4.25, N: 9.09, Cl: 11.68 (for sample cryst. from CH_3OH /toluene). IR (cm^{-1}): 3406 (m), 1610 (s).

(Z)-3-[Amino(4-chlorophenyl)methylidene]-6-chloro-1,3-dihydro-2H-indol-2-one (5dc)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–60% EtOAc during 20 min). Yellow solid (861 mg, 94%); mp 228 – 229.5 °C (CH_3OH/H_2O); 1H NMR (400 MHz, $DMSO-d_6$) δ (ppm): 10.54 (bs, 1H, NH), 9.43 (bd, $J = 2.6$ Hz, 1H, $\frac{1}{2}NH_2$), 8.23 (bs, 1H, $\frac{1}{2}NH_2$), 7.65 (AA'XX', $J = 8.4$ Hz, 2H, Ar-H), 7.55 (AA'XX', $J = 8.4$ Hz, 2H, Ar-H), 6.81 (d, $J = 1.7$ Hz, 1H, Ar-H₇); 6.62 (dd, $J = 8.3$ and 1.9 Hz, 1H, Ar-H₅), 6.02 (d, $J = 8.3$ Hz, 1H, Ar-H₄); ^{13}C NMR (100 MHz, $DMSO-d_6$) δ (ppm): 170.1, 159.6, 137.2, 135.1, 134.1, 129.9, 129.2, 126.7, 123.4, 119.4, 118.0, 108.7, 93.2; HRMS: calcd. for $C_{15}H_{11}Cl_2N_2O$ [$M+H^+$]: calcd. 305.0249, found 305.0247; Elemental analysis: calcd. C: 59.04, H: 3.30, N: 9.18, Cl: 23.23; found C: 59.41, H: 3.19, N: 8.85, Cl: 22.97. IR (cm^{-1}): 3457 (m), 1651 (s), 1596 (s).

(Z)-3-[Amino(4-iodophenyl)methylidene]-6-chloro-1,3-dihydro-2H-indol-2-one (5dc')

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane/THF 70:25.5). Orange solid (510 mg, 86%); mp 256 – 260 °C; 1H NMR (500 MHz, $CDCl_3$) δ (ppm): 9.46 (bs, 1H, $\frac{1}{2}NH_2$), 8.09 (bs, 1H, NH), 7.90 (d, $J = 8.2$ Hz, 2H, AA'XX', Ar-H), 7.30 (d, $J = 8.2$ Hz, 2H, AA'XX', Ar-H), 6.90 (d, $J = 1.8$ Hz, 1H, Ar-H₇); 6.69 (dd, $J = 8.3$ and 1.9 Hz, 1H, Ar-H₅), 6.25 (d, $J = 8.3$ Hz, 1H, Ar-H₄), 5.20 (bs, 1H, $\frac{1}{2}NH_2$); ^{13}C NMR (125 MHz, $CDCl_3$) δ (ppm): 170.7, 158.3, 138.6, 136.4, 134.9, 129.4, 129.1, 122.8, 120.7, 119.1, 109.5, 97.1, 95.6; HRMS: calcd. for $C_{15}H_{11}ClIN_2O$ [$M+H^+$]: calcd. 396.9599, found 396.9605; Elemental analysis: calcd. C: 45.43, H: 2.54, N: 7.06, Cl: 8.94, I: 32.00; found C: 45.33, H: 2.39, N: 6.90 (for sample cryst. from CH_3OH /toluene and triturated with *n*-hexane). IR (cm^{-1}): 3452 (m), 1662 (s), 1605 (s).

(Z)-3-[Amino(4-trifluoromethylphenyl)methylidene]-6-chloro-1,3-dihydro-2H-indol-2-one (5dd)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/EtOAc + 3% TEA, gradient 0–60% EtOAc during 20 min). Yellow solid (796 mg, 78%); mp 292 – 295 °C (toluene/ CH_3OH); 1H NMR (400 MHz, $DMSO-d_6$) δ (ppm): 10.59 (bs, 1H, NH), 9.46 (bs, 1H, $\frac{1}{2}NH_2$), 8.29 (bs, 1H, $\frac{1}{2}NH_2$), 7.96 (AA'XX', $J = 8.0$ Hz, 2H, Ar-H), 7.76 (AA'XX', $J = 8.0$ Hz, 2H, Ar-H), 6.82 (s, 1H, Ar-H₇); 6.60 (d, $J = 8.3$ Hz, 1H, Ar-H₅), 5.91 (d, $J = 8.4$ Hz, 1H, Ar-H₄); ^{13}C NMR (100 MHz, $DMSO-d_6$) δ (ppm): 170.1, 159.1, 139.3, 137.4, 130.6 (q, $^2J_{C-F} = 32.1$ Hz), 129.0, 126.9, 126.1 (q, $^3J_{C-F} = 3.7$ Hz), 124.1 ($^1J_{C-F} = 271$ Hz), 123.2, 119.4, 117.9, 108.8, 93.4; HRMS: calcd. for $C_{16}H_{11}ClF_3N_2O$ [$M+H^+$]: calcd. 339.0512, found 339.0518; Elemental analysis: not measured due to fluorine content damaging column. IR (cm^{-1}): 3497 (m), 1663 (s), 1605 (s).

(Z)-3-[Amino(phenyl)methylidene]-5-nitro-1,3-dihydro-2H-indol-2-one (5ea)

Yellow solid (445 mg, 79%); mp 295 - 298 °C; ¹H NMR (500 MHz, DMSO-*d*₆) δ: 11.12 (bs, 1H, NH), 9.56 (bd, *J* = 3.7 Hz, 1H, ½NH₂), 8.69 (bd, *J* = 3.4 Hz, 1H, ½NH₂), 7.81 (dd, *J* = 8.6 and 2.4 Hz, 1H, Ar-H₆), 7.70 (m, 1H, Ar-H_p), 7.64 (m, 2H, Ar-H_m), 7.57 (m, 2H, Ar-H_o), 6.95 (d, *J* = 8.6 Hz, 1H, Ar-H₇), 6.84 (d, *J* = 2.3 Hz, 1H, Ar-H₄); ¹³C NMR (125 MHz, DMSO-*d*₆) δ (ppm): 170.5, 162.9, 141.4, 140.7, 134.6, 130.8, 129.3, 127.8, 125.3, 119.0, 112.0, 108.4, 92.5; HRMS: calcd. for C₁₅H₁₁N₃O₃Na [M+Na⁺]: calcd. 304.0698, found 304.0696; Elemental analysis: calcd. C: 64.05, H: 3.94, N: 14.94; found C: 64.20, H: 3.90, N: 14.55. IR (cm⁻¹): 3430 (s), 1661 (s), 1616 (s).

(Z)-3-[Amino(4-methoxyphenyl)methylidene]-5-nitro-1,3-dihydro-2H-indol-2-one (5eb)

The crude product was purified using flash chromatography (silica gel, CHCl₃/CH₃OH; gradient changing from 99:1 to 90:10 during 25 min). Yellow solid (455 mg, 73%); mp 328 - 331 °C; ¹H NMR (500 MHz, CDCl₃) δ: 11.11 (bs, 1H, NH), 9.56 (bs, 1H, ½NH₂), 8.58 (bs, 1H, ½NH₂), 7.82 (dd, *J* = 8.6 and 2.3 Hz, 1H, Ar-H₆), 7.53 (AA'XX', *J* = 8.6 Hz, 2H, Ar-H_{3',5'}), 7.18 (AA'XX', *J* = 8.7 Hz, 2H, Ar-H_{2',6'}), 7.09 (d, *J* = 2.0 Hz, 1H, Ar-H₄), 6.95 (d, *J* = 8.6 Hz, 1H, Ar-H₇), 3.88 (s, 3H, OCH₃); ¹³C NMR (125 MHz, DMSO-*d*₆) δ (ppm): 170.9, 163.3, 161.6, 141.5, 141.0, 130.0, 127.0, 125.8, 119.2, 114.9, 112.4, 108.7, 92.7, 55.9; HRMS: calcd. for C₁₆H₁₄N₃O₄ [M+H⁺]: calcd. 312.0979, found 312.0981; Elemental analysis: calcd. C: 61.73, H: 4.21, N: 13.50; found C: 61.68, H: 4.17, N: 13.44 (for sample cryst. from EtOH/toluene). IR (cm⁻¹): 3464 (m), 1659 (s), 1605 (s).

(Z)-3-[Amino(4-chlorophenyl)methylidene]-5-nitro-1,3-dihydro-2H-indol-2-one (5ec)

Yellow solid (489 mg, 78%); mp >372 °C (dec.); ¹H NMR (500 MHz, CDCl₃) δ: 11.14 (bs, 1H, NH), 9.52 (bd, *J* = 3.6 Hz, 1H, ½NH₂), 8.69 (bd, *J* = 3.3 Hz, 1H, ½NH₂), 7.82 (dd, *J* = 8.6 and 2.3 Hz, 1H, Ar-H), 7.53 (AA'XX', *J* = 8.6 Hz, 2H, Ar-H), 7.82 (dd, *J* = 8.6 and 2.3 Hz, 1H, Ar-H₆), 7.72 (AA'XX', *J* = 8.5 Hz, 2H, Ar-H), 7.61 (AA'XX', *J* = 8.5 Hz, 2H, Ar-H), 6.95 (d, *J* = 8.7 Hz, 1H, Ar-H₇), 6.87 (d, *J* = 2.3 Hz, 1H, Ar-H₄); ¹³C NMR (125 MHz, DMSO-*d*₆) δ (ppm): 170.6, 161.6, 141.7, 140.9, 135.7, 133.6, 130.1, 129.6, 125.3, 119.4, 112.1, 108.7, 92.8; HRMS: calcd. for C₁₅H₁₁ClN₃O₃ [M+H⁺]: calcd. 316.0489, found 316.0490; Elemental analysis: calcd. C: 57.07, H: 3.19, N: 13.31; found C: 57.13, H: 3.10, N: 13.10. IR (cm⁻¹): 3454 (s), 1652 (s), 1609 (s).

(Z)-3-[Amino(4-trifluoromethylphenyl)methylidene]-5-nitro-1,3-dihydro-2H-indol-2-one (5ed)

The crude product was purified using flash chromatography (silica gel, *n*-hexane/EtOAc; gradient changing from 85:15 to 20:80 during 20 min). Yellow solid (674 mg, 97%); mp 345 °C (decomp.); ¹H NMR (500 MHz, CDCl₃) δ: 11.18 (bs, 1H, NH), 9.50 (bs, 1H, NH), 8.78 (s, 1H, NH), 8.03 (AA'XX', *J* = 8.1 Hz, 2H, Ar-H), 7.78 – 7.85 (m, 4H, Ar-H, AA'XX' + Ar-H), 6.96 (d, *J* = 8.6 Hz, 1H, Ar-H), 6.63 (d, *J* = 2.1 Hz, 1H, Ar-H₄); ¹H NMR (125 MHz, DMSO-*d*₆) δ (ppm): 170.3, 160.9, 141.6, 140.7, 138.6, 130.9 (q, ²*J*_{C-F} = 32.1 Hz), 128.9, 126.3, 124.9, 124.0 (q, ¹*J*_{C-F} = 272.5 Hz), 119.3, 111.8, 108.5, 92.8; HRMS: calcd. for C₁₆H₁₁F₃N₃O₃ [M+H⁺]:

calcd. 350.0747, found 350.0748; Elemental analysis: not measured due to fluorine content damaging analyzer. IR (cm⁻¹): 3478 (m), 3403 (m), 1667 (s), 1609 (m).

(Z)-3-[Phenyl(phenylamino)methylidene]-1,3-dihydro-2H-indol-2-one (6aa)

The crude product was purified using column chromatography (neutral alumina, *n*-hexane/EtOAc 3:2). Yellow solid (721 mg, 77%); mp 322.5 – 325 °C; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.01 (bs, 1H, NH), 10.73 (bs, 1H, NH), 7.52 – 7.61 (m, 3H, Ar-H), 7.44 – 7.51 (m, 2H, Ar-H), 7.14 (t, *J* = 7.7 Hz, 2H, Ar-H), 6.98 (t, *J* = 7.3 Hz, 1H, Ar-H), 6.90 (t, *J* = 7.5 Hz, 1H, Ar-H₆), 6.85 (d, *J* = 7.5 Hz, 1H, Ar-H₇), 6.77 – 6.82 (m, 2H, Ar-H), 6.53 (t, *J* = 7.5 Hz, 1H, Ar-H₅), 5.77 (d, *J* = 7.8 Hz, 1H, Ar-H₄); ¹³C NMR (125 MHz, DMSO-*d*₆) δ (ppm): 170.3, 155.5, 138.3, 136.9, 132.5, 131.8, 130.3, 129.6, 128.7, 124.7, 124.0, 123.8, 120.2, 118.3, 116.4, 109.3, 98.7; HRMS: calcd. for C₂₁H₁₇N₂O [M+H⁺]: calcd. 313.1341, found 313.1343; Elemental analysis: calcd. C: 80.75, H: 5.16, N: 8.97; found C: 80.80, H: 5.09, N: 8.74 (for sample cryst. from EtOAc/EtOH). IR (cm⁻¹): 3083 (m), 1641 (s), 1607 (s).

(Z)-3-[4-Methoxyphenylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6ab)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 20:1:0.1). Yellow solid (827 mg, 88%); mp 271 – 273 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 11.89 (bs, 1H, NH), 10.66 (bs, 1H, NH), 7.48 – 7.56 (m, 3H, Ar-H), 7.39 – 7.47 (m, 2H, Ar-H), 6.79 – 6.91 (m, 4H, Ar-H), 6.72 (AA'XX', *J* = 9.0 Hz, 2H, Ar-H), 6.52 (dt, *J* = 7.3 and 1.2 Hz, 1H, Ar-H), 5.74 (d, *J* = 7.8 Hz, 1H, Ar-H₄), 3.65 (s, 3H, OCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 170.2, 157.2, 156.4, 136.4, 132.8, 131.4, 129.9, 129.3, 128.6, 125.2, 124.2, 123.2, 119.9, 117.8, 114.1, 109.1, 96.9, 55.2; HRMS: calcd. for C₂₂H₁₉N₂O₂ [M+H⁺]: calcd. 343.1441, found 343.1448; Elemental analysis: calcd. C: 77.17, H: 5.30, N: 8.18; found C: 77.56, H: 5.25, N: 8.20. IR (cm⁻¹): 3063 (m), 1641 (s), 1607 (s).

(Z)-3-[4-Chlorophenylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6ac)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 10:1:0.1). Yellow solid (503 mg, 48%); mp 286.5 – 288 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 11.95 (bs, 1H, NH), 10.75 (bs, 1H, NH), 7.51 – 7.62 (m, 3H, Ar-H), 7.41 – 7.51 (m, 2H, Ar-H), 7.18 (AA'X'X', *J* = 8.8 Hz, 2H, Ar-H), 6.91 (t, *J* = 7.5 Hz, 1H, Ar-H₆), 6.85 (d, *J* = 7.5 Hz, 1H, Ar-H₇), 6.81 (AA'X'X', *J* = 8.8 Hz, 2H, Ar-H), 6.54 (t, *J* = 7.5 Hz, 1H, Ar-H₅), 5.81 (d, *J* = 7.7 Hz, 1H, Ar-H₄); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 170.3, 155.7, 137.8, 136.9, 132.5, 130.3, 129.6, 128.9, 128.7, 128.3, 124.4, 123.96, 123.87, 120.2, 118.3, 109.4, 98.6; HRMS: calcd. for C₂₁H₁₆ClN₂O [M+H⁺]: calcd. 347.0946, found 347.0953; Elemental analysis: calcd. C: 72.73, H: 4.36, N: 8.08, Cl: 10.22; found C: 72.76, H: 4.33, N: 7.99, Cl: 9.93. IR (cm⁻¹): 3087 (m), 1642 (s), 1613 (s).

(Z)-3-[Phenyl(4-trifluoromethylphenylamino)methylidene]-1,3-dihydro-2H-indol-2-one (6ad)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 20:1:0.1). Yellow solid (488 mg, 43%); mp 282 – 284 °C; ¹H NMR (400

MHz, DMSO-*d*₆) δ 12.11 (bs, 1H, NH), 10.83 (bs, 1H, NH), 7.56 – 7.67 (m, 3H, Ar-H), 7.50 – 7.55 (m, 2H, Ar-H), 7.47 (AA'XX', *J* = 8.6 Hz, 2H, Ar-H), 6.94 (t, *J* = 7.5 Hz, 1H, Ar-H₆), 6.84 – 6.91 (m, 3H, Ar-H), 6.56 (t, *J* = 7.6 Hz, 1H, Ar-H₅), 5.85 (d, *J* = 7.9 Hz, 1H, Ar-H₄); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 170.3, 154.5, 142.7, 137.3, 132.5, 130.6, 129.8, 128.6, 126.1 (q, *J* = 3.6 Hz), 124.3 (q, *J* = 272 Hz, CF₃), 124.5, 123.6, 123.6 (q, *J* = 32.1 Hz), 121.7, 120.3, 118.7, 109.5, 100.1; HRMS: calcd. for C₂₂H₁₆F₃N₂O [M+H⁺]: calcd. 381.1209, found 381.1216; Elemental analysis: not measured due to fluorine content damaging column. IR (cm⁻¹): 3149 (m), 1650 (m), 1611 (s).

(Z)-3-[4-Nitrophenylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6ae)

The crude product was purified using column chromatography (silica gel, DCM/CH₃OH/TEA 20:1:0.05). Orange solid (339 mg, 32%); mp 281 – 283.5 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 12.19 (bs, 1H, NH), 10.89 (bs, 1H, NH), 7.97 (AA'XX', *J* = 9.1 Hz, 2H, Ar-H), 7.60 – 7.70 (m, 3H, Ar-H), 7.53 – 7.59 (m, 2H, Ar-H), 6.97 (t, *J* = 7.6 Hz, 1H, Ar-H₆), 6.87 (d, *J* = 7.7 Hz, 1H, Ar-H₇), 6.82 (AA'XX', *J* = 9.1 Hz, 2H, Ar-H), 6.58 (t, *J* = 7.6 Hz, 1H, Ar-H₅), 5.88 (d, *J* = 7.7 Hz, 1H, Ar-H₄); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 170.2, 153.0, 145.6, 142.0, 137.8, 132.4, 130.9, 130.0, 128.6, 125.2, 125.0, 123.3, 120.52, 120.48, 119.1, 109.7, 102.0; HRMS: calcd. for C₂₁H₁₆N₃O₃ [M+H⁺]: calcd. 358.1186, found 358.1195; Elemental analysis: calcd. for hemihydrate C: 68.84, H: 4.40, N: 11.47; found C: 68.82, H: 4.37, N: 11.25. IR (cm⁻¹): 3100 (m), 1645 (s), 1583 (s).

(Z)-3-[Methylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6af)

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane 4:1). Yellow solid (475 mg, 76%); mp 256 – 258 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.40 (bs, 1H, NH), 10.09 (d, *J* = 5.0 Hz, 1H, NH), 7.59 – 7.65 (m, 3H, Ar-H), 7.34 – 7.47 (m, 2H, Ar-H), 6.73 – 6.83 (m, 2H, Ar-H), 6.40 – 6.50 (m, 1H, Ar-H), 5.52 (d, *J* = 7.7 Hz, 1H, Ar-H), 2.74 (d, *J* = 4.8 Hz, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ 170.0, 161.9, 135.7, 132.9, 129.9, 129.6, 127.5, 125.0, 122.0, 119.6, 116.7, 108.8, 94.1, 30.5; HRMS: calcd. for C₁₆H₁₅N₂O [M+H⁺]: calcd. 251.1184, found 251.1191; Elemental analysis: calcd. C: 76.78, H: 5.64, N: 11.19; found C: 76.46, H: 5.54, N: 11.11 (for sample cryst. from toluene/CH₃OH). IR (cm⁻¹): 3139 (m), 1634 (s), 1610 (s).

(Z)-3-[*n*-Pentylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6ag)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 20:1:0.1). Yellow solid (652 mg, 71%); mp 214 – 216 °C; ¹H NMR (400 MHz, CDCl₃) δ 10.21 (t, *J* = 5.6 Hz, 1H, NH), 9.29 (bs, 1H, CONH), 7.52 – 7.63 (m, 3H, Ar-H), 7.34 – 7.45 (m, 2H, Ar-H), 6.84 – 6.98 (m, 2H, Ar-H), 6.54 – 6.61 (m, 1H, Ar-H), 5.68 (d, *J* = 7.8 Hz, 1H, Ar-H₄), 3.11 (dd, *J* = 13.5, 6.8 Hz, 2H, NCH₂), 1.51 – 1.63 (m, 2H, CH₂), 1.19 – 1.38 (m, 4H, 2×CH₂), 0.87 (t, *J* = 7.0 Hz, 3H, CH₃); ¹³C NMR-APT (100 MHz, CDCl₃) δ (ppm): 170.8 (C=O), 161.9 (C_q), 135.0 (C_q), 133.2 (C_q), 129.6 (C-H), 129.3 (C-H), 127.6 (C-H), 125.2 (C_q), 122.1 (C-H), 120.2 (C-H), 117.4 (C-H), 109.0 (C-H), 94.6 (C_q), 43.9 (CH₂), 30.4 (CH₂), 28.7 (CH₂), 22.2 (CH₂), 13.9 (CH₃); HRMS: calcd. for C₂₀H₂₁N₂O [M+H⁺]: calcd.

307.1805, found 307.1809; Elemental analysis: calcd. for hemihydrate C: 78.40, H: 7.24, N: 9.14; found C: 78.40, H: 7.27, N: 8.98. IR (cm⁻¹): 3137 (m), 1643 (s), 1589 (s).

(Z)-3-[Benzylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6ah)

The crude product was purified using column chromatography (neutral alumina, DCM/i-PrOH/conc. NH₃ 30:1:0.1). Light yellow solid (696 mg, 71%); mp 231 – 234 °C; ¹H NMR (400 MHz, CDCl₃) δ 10.52 (bt, *J* = 5.8 Hz, 1H, NH), 8.85 and 8.91 (2×bs, 1H, NH), 7.40 – 7.59 (m, 3H, Ar-H), 7.28 – 7.37 (m, 4H, Ar-H), 7.23 – 7.28 (m, 1H, Ar-H), 7.17 – 7.23 (m, 2H, Ar-H), 6.85 – 6.93 (m, 2H, Ar-H), 6.58 (dt, *J* = 7.0 and 1.9 Hz, 1H, Ar-H), 5.70 (d, *J* = 7.8 Hz, 1H, Ar-H₄), 4.34 (d, *J* = 6.5 Hz, 2H, CH₂); ¹³C NMR-APT (100 MHz, CDCl₃) δ (ppm): 170.8, 161.6, 138.3, 135.1, 132.7, 129.8, 129.4, 128.7, 127.7, 127.4, 126.8, 125.1, 122.6, 120.4, 117.7, 109.1, 95.6, 47.6; HRMS: calcd. for C₂₂H₁₉N₂O [M+H⁺]: calcd. 327.1492, found 327.1498; Elemental analysis: calcd. C: 80.96, H: 5.56, N: 8.58; found C: 81.30, H: 5.41, N: 8.35 (for sample cryst. from CH₃OH/H₂O). IR (cm⁻¹): 3131 (m), 1640 (s), 1606 (s).

(Z)-3-[Cyclohexylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6ai)

The crude product was purified using column chromatography (neutral alumina, DCM/i-PrOH/conc. NH₃ 20:1:0.1). Yellowish solid (613 mg, 64%); mp 343 – 346 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.47 (bs, 1H, NH), 10.41 (d, *J* = 9.6 Hz, 1H, NH), 7.66 – 7.71 (m, 3H, Ar-H), 7.47 – 7.51 (m, 2H, Ar-H), 6.80 – 6.85 (m, 2H, Ar-H), 6.44 – 6.53 (m, 1H, Ar-H), 5.47 (d, *J* = 7.7 Hz, 1H, Ar-H₄), 3.05 - 3.16 (m, 1H, NCH), 1.76 - 1.84 (m, 2H, 2×½CH₂), 1.63 – 1.72 (m, 2H, 2×½CH₂), 1.07 – 1.52 (m, 6H, 3 × CH₂); ¹³C NMR-APT (100 MHz, DMSO-*d*₆) δ (ppm): 169.9, 160.1, 135.6, 133.0, 129.9, 129.6, 127.4, 124.8, 122.0, 119.5, 116.6, 108.7, 94.0, 51.4, 33.7, 24.8, 23.9; HRMS: calcd. for C₂₁H₂₃N₂O [M+H⁺]: calcd. 319.1805, found 319.1810; Elemental analysis: calcd. C: 79.21, H: 6.96, N: 8.80; found C: 79.31, H: 6.99, N: 8.69. IR (cm⁻¹): 3137 (m), 1638 (s), 1605 (s), 1582 (s).

(Z)-5-Methyl-3-[phenyl(phenylamino)methylidene]-1,3-dihydro-2H-indol-2-one (6ba)

The crude product was purified using column chromatography (silica gel, DCM/CH₃OH/TEA 20:1:0.05). Yellow solid (674 mg, 69%); mp 258 – 261 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 12.03 (bs, 1H, CONH), 10.62 (bs, 1H, NH), 7.52 – 7.59 (m, 3H, Ar-H), 7.46 (dd, *J* = 7.5 and 1.3 Hz, 2H, Ar-H), 7.13 (t, *J* = 7.8 Hz, 2H, Ar-H), 6.97 (t, *J* = 7.4 Hz, 1H, Ar-H), 6.80 (d, *J* = 7.8 Hz, 2H, Ar-H), 6.73 (q, *J* = 7.9 Hz, 2H, Ar-H), 5.60 (s, 1H, Ar-H₄), 1.92 (s, 3H, CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 170.4, 155.9, 138.8, 134.6, 132.9, 130.1, 129.4, 129.0, 128.66, 128.29, 124.22, 124.16, 124.13, 122.6, 119.04, 119.01, 108.9, 98.2, 21.2; HRMS: calcd. for C₂₂H₁₉N₂O [M+H⁺]: calcd. 327.1492, found 327.1501; Elemental analysis: calcd. C: 80.96, H: 5.56, N: 8.58; found C: 81.10, H: 5.52, N: 8.58. IR (cm⁻¹): 3222 (m), 1641 (s), 1626 (s).

(Z)-5-Chloro-3-[phenyl(phenylamino)methylidene]-1,3-dihydro-2H-indol-2-one (6ca)

The crude product was purified using column chromatography (silica gel, DCM/CH₃OH 99:1). Yellow solid (380 mg, 89%); mp 242 - 243 °C; ¹H NMR (500 MHz, CDCl₃) δ 11.98 (bs, 1H, NH), 8.62 (bs, 1H, NH), 7.50 – 7.59 (m, 3H, Ar-H), 7.39 – 7.43 (m, 2H, Ar-H), 7.13 (t, *J* = 7.9

Hz, 2H, Ar-H), 7.01 (t, $J = 7.3$ Hz, 1H, Ar-H), 6.92 (dd, $J = 8.2$ and 2.0 Hz, 1H, Ar-H₆), 6.84 (d, $J = 8.3$ Hz, 1H, Ar-H₇), 6.78 – 6.82 (m, 2H, Ar-H), 5.88 (d, $J = 1.9$ Hz, 1H, Ar-H₄); ^{13}C NMR (125 MHz, CDCl_3) δ (ppm): 170.7, 158.2, 138.4, 133.8, 132.4, 130.3, 129.5, 128.8, 128.5, 126.14, 126.09, 124.8, 123.3, 123.1, 118.8, 109.9, 97.1; HRMS: calcd. for $\text{C}_{21}\text{H}_{16}\text{ClN}_2\text{O}$ $[\text{M}+\text{H}^+]$: calcd. 347.0946, found 347.0958; Elemental analysis: calcd. C: 72.73, H: 4.36, N: 8.08, Cl: 10.22; found C: 72.28, H: 4.19, N: 7.97, Cl: 10.52 (for sample cryst. from EtOAc/*n*-hexane). IR (cm^{-1}): 3148 (m), 1645 (s), 1608 (s).

(Z)-6-Chloro-3-[phenyl(phenylamino)methylidene]-1,3-dihydro-2H-indol-2-one (6da)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH_3 20:1:0.1). Yellow solid (749 mg, 72%); mp 319 – 322 °C; ^1H NMR (400 MHz, $\text{DMSO}-d_6$) δ 11.98 (bs, 1H, NH), 10.87 (bs, 1H, NH), 7.51 – 7.59 (m, 3H, Ar-H), 7.46 (m, 2H, Ar-H), 7.15 (t, $J = 7.8$ Hz, 2H, Ar-H), 7.00 (t, $J = 7.3$ Hz, 1H, Ar-H), 6.86 (d, $J = 1.8$ Hz, 1H, Ar-H₇), 6.82 (d, $J = 7.9$ Hz, 2H, Ar-H), 6.60 (dd, $J = 8.3$ and 1.8 Hz, 1H, Ar-H₅), 5.71 (d, $J = 8.3$ Hz, 1H, Ar-H₄); ^{13}C NMR (100 MHz, $\text{DMSO}-d_3$) δ (ppm): 170.2, 156.9, 138.4, 137.7, 132.5, 130.3, 129.5, 129.0, 128.5, 127.6, 124.6, 123.0, 123.0, 119.7, 118.9, 109.1, 97.1; HRMS: calcd. for $\text{C}_{21}\text{H}_{16}\text{ClN}_2\text{O}$ $[\text{M}+\text{H}^+]$: calcd. 347.0946, found 347.0954; Elemental analysis: calcd. C: 72.73, H: 4.36, N: 8.08, Cl: 10.22; found C: 72.82, H: 4.36, N: 8.09, Cl: 10.01 (for sample cryst. from EtOAc/*n*-hexane). IR (cm^{-1}): 3064 (m), 1646 (s), 1605 (s).

(Z)-5-Nitro-3-[phenyl(phenylamino)methylidene]-1,3-dihydro-2H-indol-2-one (6ea)

The crude product was purified using flash chromatography (silica gel, DCM/EtOAc + 3% TEA; gradient changing from 0–50% EtOAc during 20 min). Orange solid (529 mg, 74%); mp 267 – 269 °C; ^1H NMR: 11.98 (vbs, 2H, $2\times$ NH), 7.85 (dd, $J = 8.6$ and 2.2 Hz, 1H, Ar-H₆), 7.54 – 7.67 (m, 3H, Ar-H), 7.50 (d, $J = 7.0$ Hz, 2H, Ar-H_o), 7.18 (d, $J = 7.7$ Hz, 2H, Ar-H_m), 7.05 (t, $J = 7.6$ Hz, 1H, Ar-H_p), 7.00 (d, $J = 8.7$ Hz, 1H, Ar-H₇), 6.91 (d, $J = 7.7$ Hz, 2H, Ar-H_o), 6.60 (s, 1H, Ar-H₄). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$, -60 °C) δ (ppm): 170.5, 158.6, 141.8, 141.0, 138.0, 131.8, 130.2, 129.3, 128.7, 128.2, 124.9, 124.6, 123.3, 119.5, 112.8, 108.6, 96.4 HRMS: calcd. for $\text{C}_{21}\text{H}_{15}\text{NaN}_3\text{O}_3$ $[\text{M}+\text{Na}^+]$: calcd. 380.1011, found 380.1016; Elemental analysis: calcd. C: 70.58, H: 4.23, N: 11.76; found C: 70.79, H: 4.22, N: 11.61. IR (cm^{-1}): 3351 (m), 1654 (s), 1622 (m). IR (cm^{-1}): 3351 (m), 1654 (s), 1622 (m).

(Z)-5-Methyl-3-[methylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6bf)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH_3 25:1:0.1). Beige solid (521 mg, 66%); mp 255 – 257.5 °C; ^1H NMR (400 MHz, $\text{DMSO}-d_6$) δ 10.27 (bs, 1H, NH), 10.06 (bq, $J = 5.1$ Hz, 1H, NH), 7.58 – 7.67 (m, 3H, Ar-H), 7.37 – 7.44 (m, 2H, Ar-H), 6.66 (d, $J = 7.8$ Hz, 1H, Ar-H₇), 6.59 (d, $J = 7.8$ Hz, 1H, Ar-H₆), 5.31 (s, 1H, Ar-H₄), 2.74 (d, $J = 5.1$ Hz, 3H, NCH_3), 1.87 (s, 3H, CH_3); ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$) δ (ppm): 170.1, 161.6, 133.6, 133.0, 129.7, 129.6, 127.6, 127.5, 125.1, 122.5, 117.5, 108.4, 94.2, 30.4, 21.3; HRMS: calcd. for $\text{C}_{17}\text{H}_{17}\text{N}_2\text{O}$ $[\text{M}+\text{H}^+]$: calcd. 265.1335, found 265.1339; Elemental analysis: calcd. C: 77.25, H: 6.10, N: 10.60; found C: 77.36, H: 6.08, N: 10.43 (for sample cryst. from $\text{CH}_3\text{OH}/\text{CHCl}_3$). IR (cm^{-1}): 3137 (m), 1628 (s), 1607 (s).

(Z)-5-Chloro-3-[methylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6cf)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 30:1:0.1). Yellow solid (605 mg, 71%); mp 256 – 258 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.54 (bs, NH, 1H), 10.17 (bq, *J* = 5.1 Hz, 1H, NH), 7.61 – 7.71 (m, 3H, Ar-H), 7.39 – 7.46 (m, 2H, Ar-H), 6.72 – 6.81 (m, 2H, Ar-H), 5.36 (d, *J* = 1.3 Hz, 1H, Ar-H₄), 2.77 (d, *J* = 5.1 Hz, 3H, NCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 169.8, 162.8, 134.2, 132.5, 130.1, 129.8, 127.3, 126.8, 123.8, 121.2, 116.1, 109.8, 93.5, 30.6; HRMS: calcd. for C₁₆H₁₄ClN₂O [M+H⁺]: calcd. 285.0789, found 285.0793; Elemental analysis: calcd. C: 67.49, H: 4.60, N: 9.84, Cl: 12.45; found C: 67.30, H: 4.57, N: 10.08, Cl: 12.38 (for sample cryst. from CH₃OH/H₂O).

(Z)-6-Chloro-3-[methylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (6df)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 15:1:0.1). Yellow solid (719 mg, 84%); mp 274 – 276.5 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.54 (bs, 1H, NH), 10.10 (bq, *J* = 5.1 Hz, 1H, NH), 7.57 – 7.71 (m, 3H, Ar-H), 7.38 – 7.45 (m, 2H, Ar-H), 6.77 (d, *J* = 1.9 Hz, 1H, Ar-H₇), 6.49 (dd, *J* = 8.3 and 2.0 Hz, 1H, Ar-H₆), 5.44 (d, *J* = 8.3 Hz, 1H, Ar-H₄), 2.75 (d, *J* = 5.1 Hz, 3H, NCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 169.8, 162.4, 136.6, 132.6, 130.1, 129.7, 127.4, 126.0, 124.0, 119.2, 117.3, 108.6, 93.6, 30.6; HRMS: calcd. for C₁₆H₁₄ClN₂O [M+H⁺]: calcd. 285.0789, found 285.0795; Elemental analysis: calcd. C: 67.49, H: 4.60, N: 9.84, Cl: 12.45; found C: 67.76, H: 4.56, N: 10.01, Cl: 12.27. IR (cm⁻¹): 3143 (m), 1641 (s), 1608 (s), 1591 (s).

(Z)-3-[Methylamino(phenyl)methylidene]-5-nitro-1,3-dihydro-2H-indol-2-one (6ef)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 15:1:0.1). Orange solid (656 mg, 74%); mp 332.5 – 335.5 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 11.12 (bs, 1H, CONH), 10.21 (bq, *J* = 5.1 Hz, 1H, NH), 7.76 (dd, *J* = 8.6 and 2.3 Hz, 1H, Ar-H₆), 7.65 – 7.73 (m, 3H, Ar-H), 7.42 – 7.52 (m, 2H, Ar-H), 6.92 (d, *J* = 8.6 Hz, 1H, Ar-H₇), 6.33 (d, *J* = 2.2 Hz, 1H, Ar-H₄), 2.82 (d, *J* = 5.1 Hz, 3H, NCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ (ppm): 170.2, 163.9, 140.78, 140.76, 132.0, 130.4, 129.9, 127.2, 125.5, 118.5, 111.5, 108.3, 92.9, 30.9; HRMS: calcd. for C₁₆H₁₄N₃O₃ [M+H⁺]: calcd. 296.1030, found 296.1023; Elemental analysis: calcd. C: 65.08, H: 4.44, N: 14.24; found C: 65.40, H: 4.27, N: 14.09. IR (cm⁻¹): 3107 (m), 1644 (s), 1585 (s).

(Z)-3-[Dimethylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (7aa)

The crude product was purified using column chromatography (silica gel, EtOAc/*n*-hexane 2:1). Yellow solid (220 mg, 83%); mp 239 – 240 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ: 9.99 (bs, 1H, NH), 7.58 – 7.64 (m, 1H, Ar-H), 7.54 (t, *J* = 7.4 Hz, 2H, Ar-H_m), 7.44 (d, *J* = 7.6 Hz, 2H, Ar-H_o), 6.66 – 6.72 (m, 2H, Ar-H), 6.29 – 6.37 (m, 1H, Ar-H), 5.35 (d, *J* = 7.8 Hz, 1H, Ar-H₄), 3.11 (bs, 6H, 2×CH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, 60 °C) δ: 165.3, 160.9, 136.9,

135.5, 130.4, 129.9, 128.9, 127.3, 121.3, 118.5, 117.8, 107.7, 97.2, 43.5; HRMS: calcd. for $C_{17}H_{17}N_2O$ $[M+H]^+$: calcd. 265.1341, found 265.1341; Elemental analysis: calcd. C: 77.25, H: 6.10, N: 10.60; found C: 76.65, H: 6.07, N: 10.43. IR (cm^{-1}): 3123 (m), 1644 (s), 1540 (s).

(Z)-3-[Dimethylamino(4-methoxyphenyl)methylidene]-1,3-dihydro-2H-indol-2-one (7ab)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/CH₃OH, gradient 0–5% CH₃OH during 20 min). Yellow solid (140 mg, 48%); mp 272 – 273 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ : 9.76 (bs, 1H, NH), 7.37 (AA'XX', J = 8.6 Hz, 2H, Ar-H), 7.07 (AA'XX', J = 8.5 Hz, 2H, Ar-H), 6.66 – 6.74 (m, 2H, Ar-H), 6.73 – 6.83 (m, 2H, Ar-H), 6.39 (m, 1H, Ar-H), 5.56 (d, J = 7.0 Hz, 1H, Ar-H), 3.86 (s, 3H, OCH₃), 3.11 (bs, 6H, 2×NCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆) δ 165.4, 161.1, 136.7, 131.9, 127.7, 127.4, 121.0, 118.5, 117.7, 114.4, 107.6, 96.8, 55.2, 43.4 (bs); HRMS: calcd. for $C_{18}H_{19}N_2O_2$ $[M+H]^+$: calcd. 295.1447, found 295.1448; Elemental analysis: calcd. C: 73.45, H: 6.16, N: 9.52; found C: 73.36, H: 6.22, N: 9.46. IR (cm^{-1}): 3007 (m), 1643 (s), 1606 (s), 1538 (s).

(Z)-3-[4-Chlorophenyl(dimethylamino)methylidene]-1,3-dihydro-2H-indol-2-one (7ac)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/CH₃OH, gradient 0–5% CH₃OH during 20 min). Yellow solid (179 mg, 60%); mp 239.5 – 241 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ : 9.84 (bs, 1H, NH), 7.59 (AA'XX', J = 7.9 Hz, 2H, Ar-H), 7.47 (AA'XX', J = 8.1 Hz, 2H, Ar-H), 6.68 – 6.75 (m, 2H, Ar-H), 6.41 (m, 1H, Ar-H), 5.52 (d, J = 7.6 Hz, 1H, Ar-H), 3.11 (bs, 6H, 2×NCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, 60 °C) δ 165.3, 159.4, 137.0, 135.3, 134.1, 132.0, 129.1, 127.0, 121.6, 118.7, 117.8, 107.8, 97.3, 43.6 (bs); HRMS: calcd. for $C_{17}H_{16}ClN_2O$ $[M+H]^+$: calcd. 299.0951, found 299.0954; Elemental analysis: calcd. C: 68.34, H: 5.06, N: 9.38, Cl: 11.87; found C: 68.67, H: 5.09, N: 9.30, Cl: 11.69. IR (cm^{-1}): 3085 (m), 1651 (s), 1544 (s).

(Z)-3-[Dimethylamino(phenyl)methylidene]-5-methyl-1,3-dihydro-2H-indol-2-one (7ba)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 10:1:0.1). Yellow solid (740 mg, 53%); mp 214 – 215 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ : 9.69 (bs, 1H, NH), 7.40 – 7.62 (m, 5H, Ar-H), 6.50 – 6.60 (m, 2H, Ar-H), 5.20 (s, 1H, Ar-H₄), 3.11 (s, 6H, 2×NCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, 60 °C) δ : 165.6, 160.7, 135.6, 134.8, 130.3, 130.0, 128.9, 127.5, 126.6, 121.9, 118.8, 107.3, 97.4, 43.5, 20.8; HRMS: calcd. for $C_{18}H_{19}N_2O$ $[M+H]^+$: calcd. 279.1492, found 279.1497; Elemental analysis: calcd. C: 77.67, H: 6.52, N: 10.06; found C: 77.79, H: 6.56, N: 9.99. IR (cm^{-1}): 3388 (m), 1639 (s)..

(Z)-6-Chloro-3-[dimethylamino(phenyl)methylidene]-1,3-dihydro-2H-indol-2-one (7ca)

The crude product was purified using column chromatography (neutral alumina, DCM/*i*-PrOH/conc. NH₃ 10:1:0.1). Yellow solid (280 mg, 19%); mp 269 – 270.5 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ : 9.95 (bs, 1H, NH), 7.61 (t, J = 7.2 Hz, 1H, Ar-H_p), 7.54 (t, J = 7.3 Hz, 2H, Ar-H_m), 7.44 (d, J = 7.3 Hz, 2H, Ar-H_o), 6.71 (d, J = 1.5 Hz, 1H, Ar-H₇), 6.36 (d, J = 7.0 Hz, 1H, Ar-H₅), 5.33 (d, J = 6.4 Hz, 1H, Ar-H₄), 3.13 (s, 6H, 2×NCH₃); ¹³C NMR (100 MHz, DMSO-*d*₆, 60 °C) δ : 165.1, 161.7, 137.8, 135.1, 130.6, 129.9, 129.0, 126.4, 125.4, 118.5, 118.1,

107.5, 96.1, 43.7; HRMS: calcd. for $C_{17}H_{16}ClN_2O$ $[M+H]^+$: calcd. 299.0951, found 299.0952; Elemental analysis: calcd. C: 68.34, H: 5.06, N: 9.38, Cl: 11.87; found C: 68.63, H: 5.17, N: 9.20, Cl: 11.71. IR (cm^{-1}): 3068 (m), 1652 (s), 1606 (m), 1542 (s).

(Z)-3-[1-Aminoethylidene]-1,3-dihydro-2H-indol-2-one (10a)

The crude product was purified using column chromatography (neutral alumina, EtOAc/CH₃OH/TEA 6:1:0.05). White solid (310 mg, 89%); mp 261 – 262.5 °C (Ref. [10] gives 225 °C); ¹H NMR (500 MHz, DMSO-*d*₆) δ 10.18 (bs, 1H, NH), 9.31 (vbs, 1H, $\frac{1}{2}$ NH₂), 7.95 (vbs, 1H, $\frac{1}{2}$ NH₂), 7.22 (d, *J* = 6.7 Hz, 1H, Ar-H), 6.90 (t, *J* = 7.3 Hz, 1H, Ar-H), 6.86 (d, *J* = 7.3 Hz, 1H, Ar-H), 6.82 (t, *J* = 7.5 Hz, 1H, Ar-H), 2.37 (s, 3H, CH₃) corresponds to Ref. [11]. HRMS: calcd. for $C_{10}H_{11}N_2O$ $[M+H]^+$: calcd. 175.0866, found 175.0865; Elemental analysis: calcd. C: 68.95, H: 5.79, N: 16.08; found C: 68.86, H: 5.76, N: 16.14. IR (cm^{-1}): 3359 (m), 1633 (s), 1538 (s).

(Z)-3-[(1-Methylamino)ethylidene]-1,3-dihydro-2H-indol-2-one (10b)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/CH₃OH + 5% TEA, gradient 0–5% CH₃OH during 15 min). White solid (320 mg, 85%); mp 281 - 282 °C (Ref. [12] gives 280 °C); ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.33 (bs, 1H, NH), 10.05 (bs, 1H, NH), 7.31 (d, *J* = 6.8 Hz, 1H, Ar-H), 6.81 - 6.93 (m, 3H, Ar-H), 3.06 (d, *J* = 5.1 Hz, 3H, NCH₃), 2.42 (s, 3H, CH₃) corresponds to Ref. [12]; ¹³C NMR (100 MHz, DMSO-*d*₆, 60 °C) δ: 169.4, 161.8, 135.0, 125.2, 121.0, 119.5, 117.4, 108.3, 93.2, 28.9, 15.1. HRMS: calcd. for $C_{11}H_{13}N_2O$ $[M+H]^+$: calcd. 189.1022, found 189.1020; Elemental analysis: calcd. C: 70.19, H: 6.43, N: 14.88; found C: 70.07, H: 6.42, N: 15.00. IR (cm^{-1}): 3131 (m), 1630 (s), 1590 (s).

(Z)-3-[(1-Phenylamino)ethylidene]-1,3-dihydro-2H-indol-2-one (10c)

The crude product was purified using preparative flash liquid chromatography (silica gel, mobile phase DCM/CH₃OH + 5% TEA, gradient 0–5% CH₃OH during 20 min). Yellow solid (312 mg, 62%); mp 222 - 223 °C (Ref. [13] gives 215-216 °C); ¹H NMR (500 MHz, CDCl₃) δ 12.02 (bs, 1H, NH), 8.74 (bs, 1H, NH), 7.41 (t, *J* = 7.8 Hz, 2H, Ar-H_m), 7.37 (d, 1H, Ar-H), 7.26 (t, *J* = 7.5 Hz, 1H, Ar-H_p), 7.20 (d, *J* = 7.6 Hz, 2H, Ar-H_o), 6.97 – 7.10 (m, 3H, Ar-H), 2.51 (s, 3H, CH₃) corresponds to Ref. [13]; ¹³C NMR (100 MHz, DMSO-*d*₆, 60 °C) δ: 169.6, 156.9, 137.7, 136.0, 129.1, 125.3, 124.5, 124.2, 122.5, 120.1, 118.7, 108.8, 96.8, 16.9. HRMS: calcd. for $C_{16}H_{15}N_2O$ $[M+H]^+$: calcd. 251.1179, found 251.1177; Elemental analysis: calcd. C: 76.78, H: 5.64, N: 11.19; found C: 76.56, H: 5.59, N: 11.35. IR (cm^{-1}): 3155 (m), 1651 (s), 1613 (s).

Eschenmoser coupling of diethyl (2-oxoindolin-3-yl)phosphate (**1g**) with thiobenzamide (**2a**)

A closeable vial was charged with thiobenzamide (**2a**, 343 mg, 0.25 mmol) and DMF (2.5 mL). Diethyl (2-oxoindolin-3-yl)phosphate (**1g**, 0.25–1 mmol, 1–4 equiv) and in one case also tetrabutylammonium bromide (161 mg, 20 mol %) were added. The closed vial was stirred at the given temperature and time below. Samples of the reaction mixture were analyzed by HPLC (Phenomenex Synergi Polar-RP 250 × 4.6 mm column; isocratic mode, mobile phase acetonitrile/water 1:1, flow rate 1 mL·min⁻¹) equipped with a UV–vis detector (245 nm). Calibration series for starting compounds and products were determined. Results are summarized in the following Table 4:

Table S4 – HPLC monitoring of reaction **1g** with **2a**.

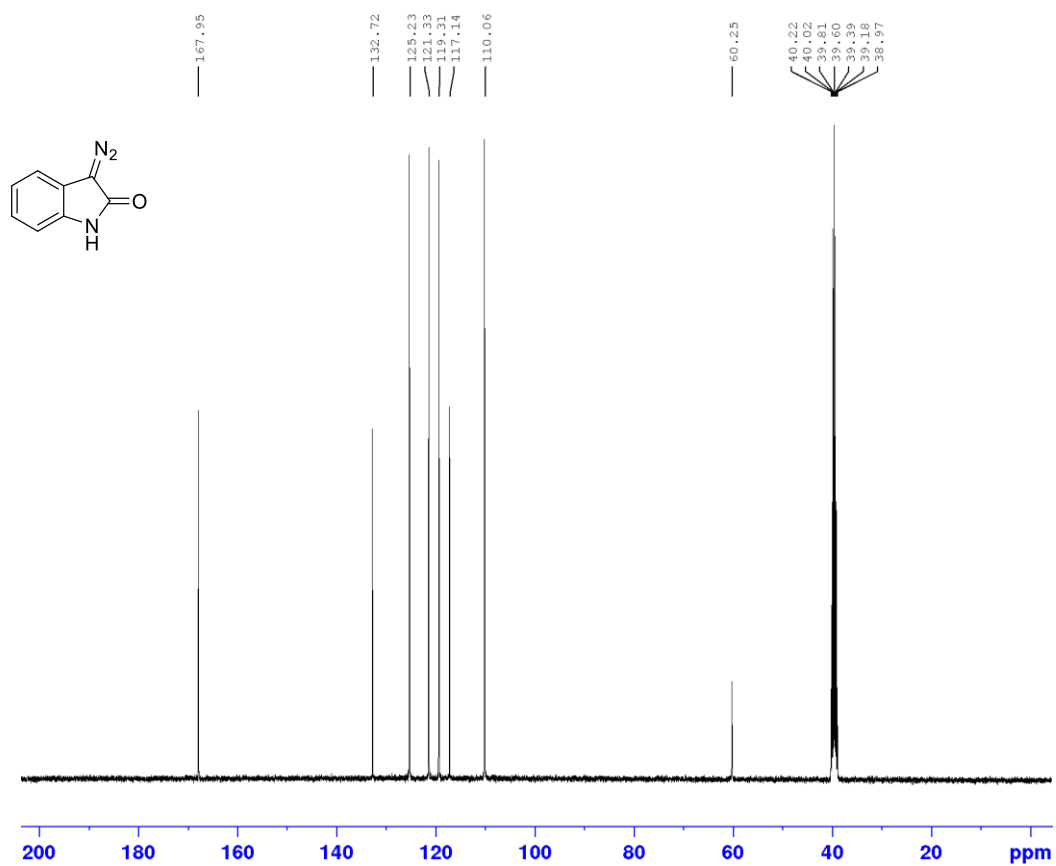
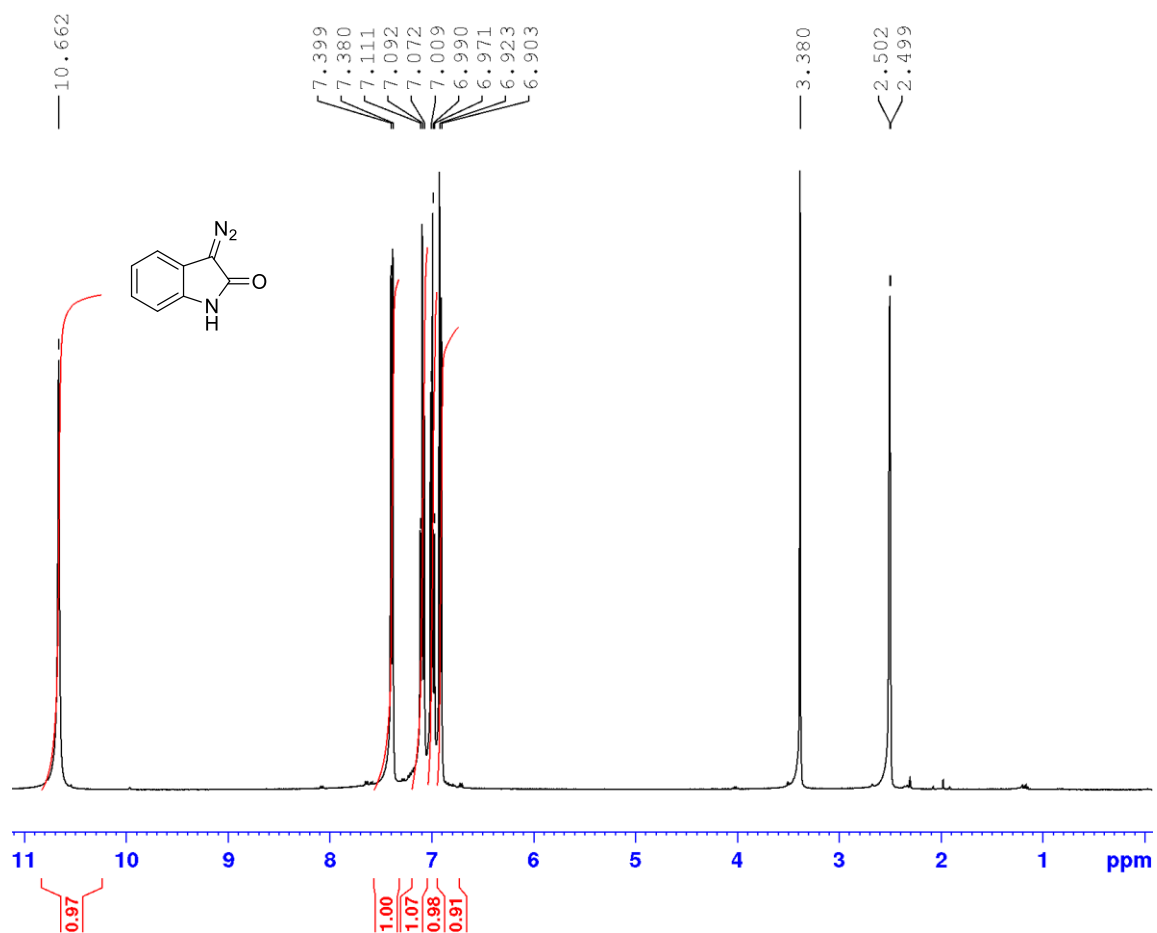
Run	Temperature [°C]	Phosphate [equiv.]	Time [h]	Conversion (%)		5aa (%)
				Thiobenzamide	Phosphate	
1	25	1	150	5	4	traces
2	80	1	6	25	17	2
			32	91	>99	21
3	80	2	6	29	22	5
			32	98	>99	38
4	80	3	6	36	18	9
			32	>99	>99	37
5	80	4	6	41	10	15
			32	>99	>99	31
6	80	1*	19	27	94	17

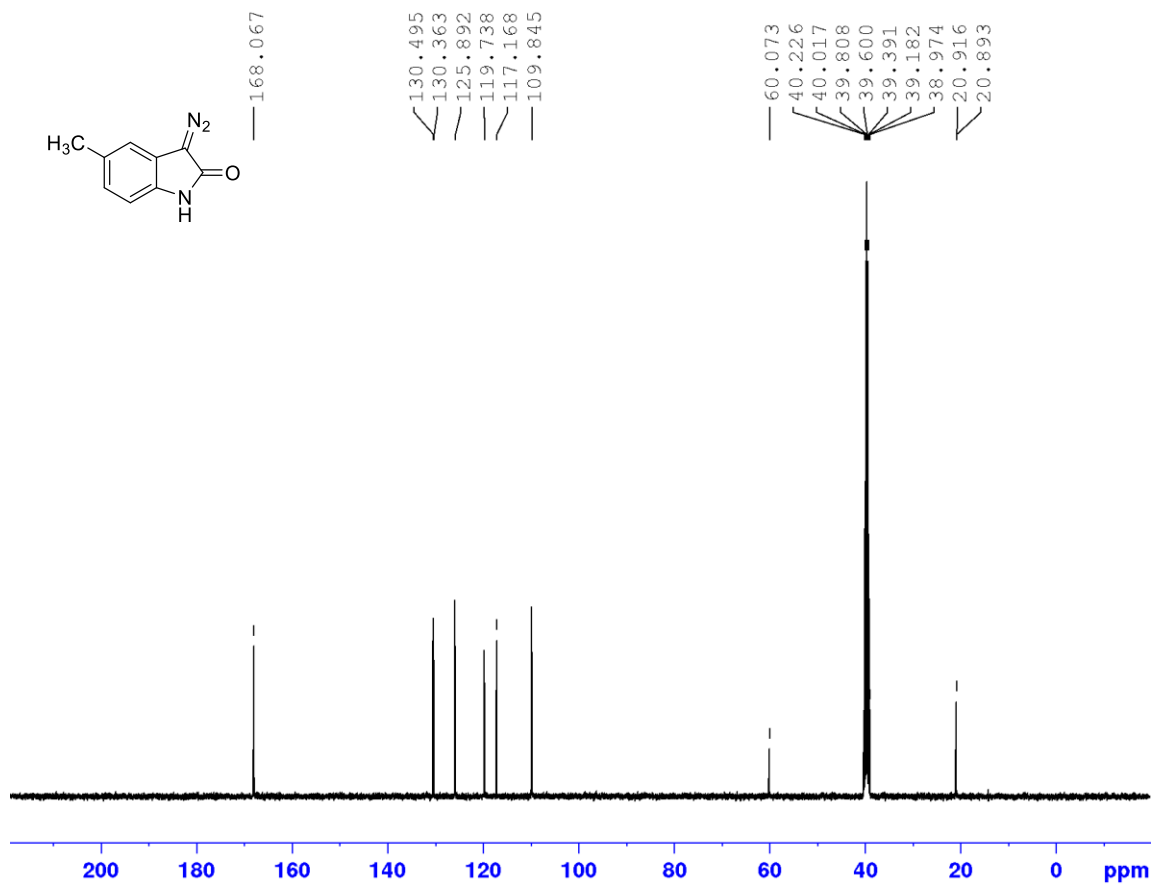
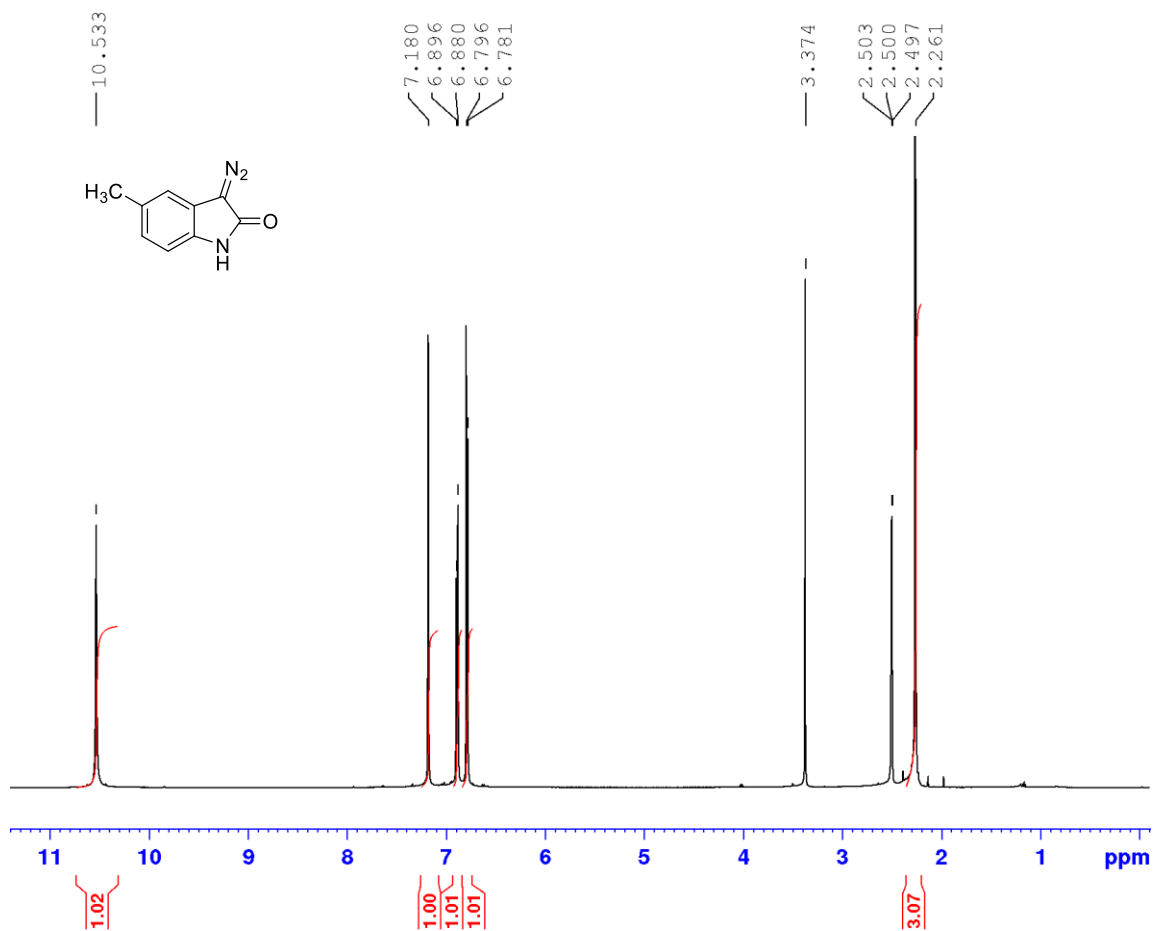
*addition of 20 mol % TBAB

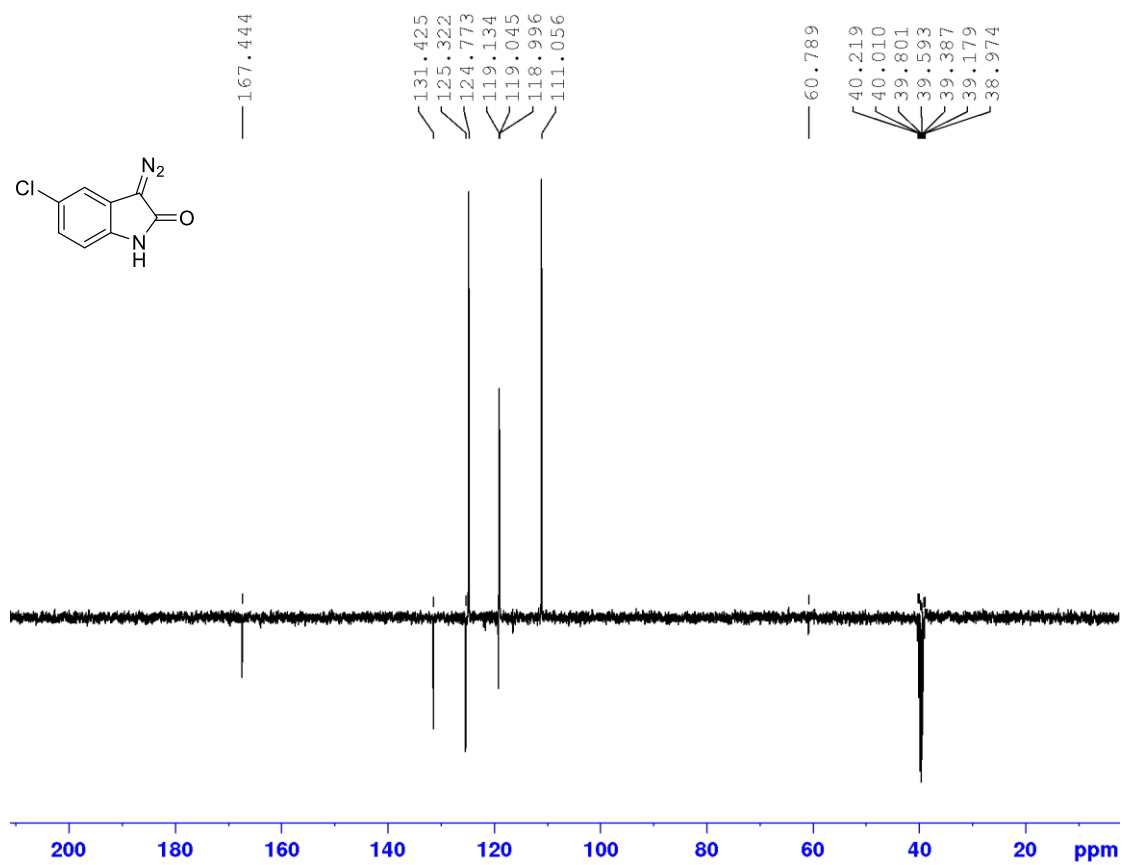
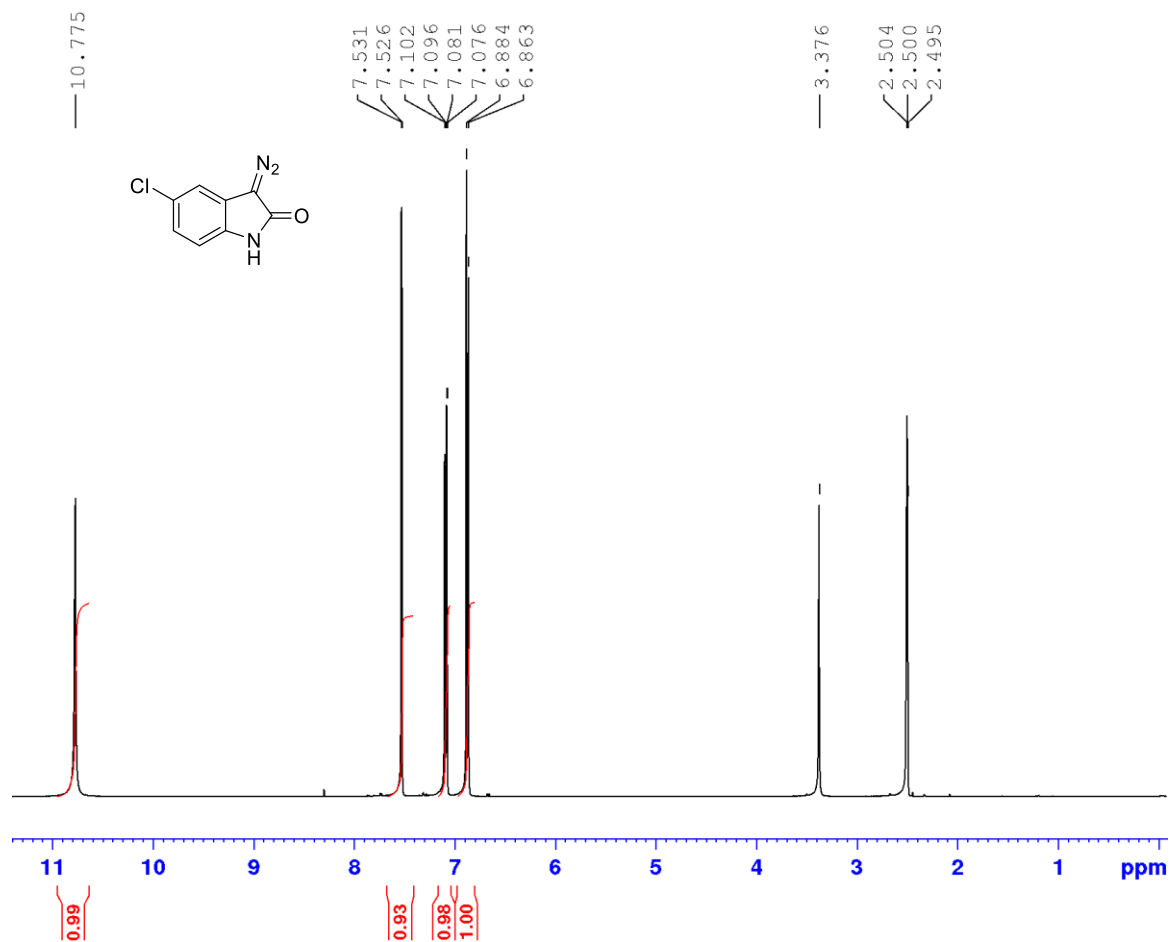
Eschenmoser coupling of 3-hydroxy-*N*-methyloxindole with thiobenzamide (**2a**) giving (*Z*)-3-[amino(phenyl)methylidene]-1-methyl-1,3-dihydro-2*H*-indol-2-one (**11**)

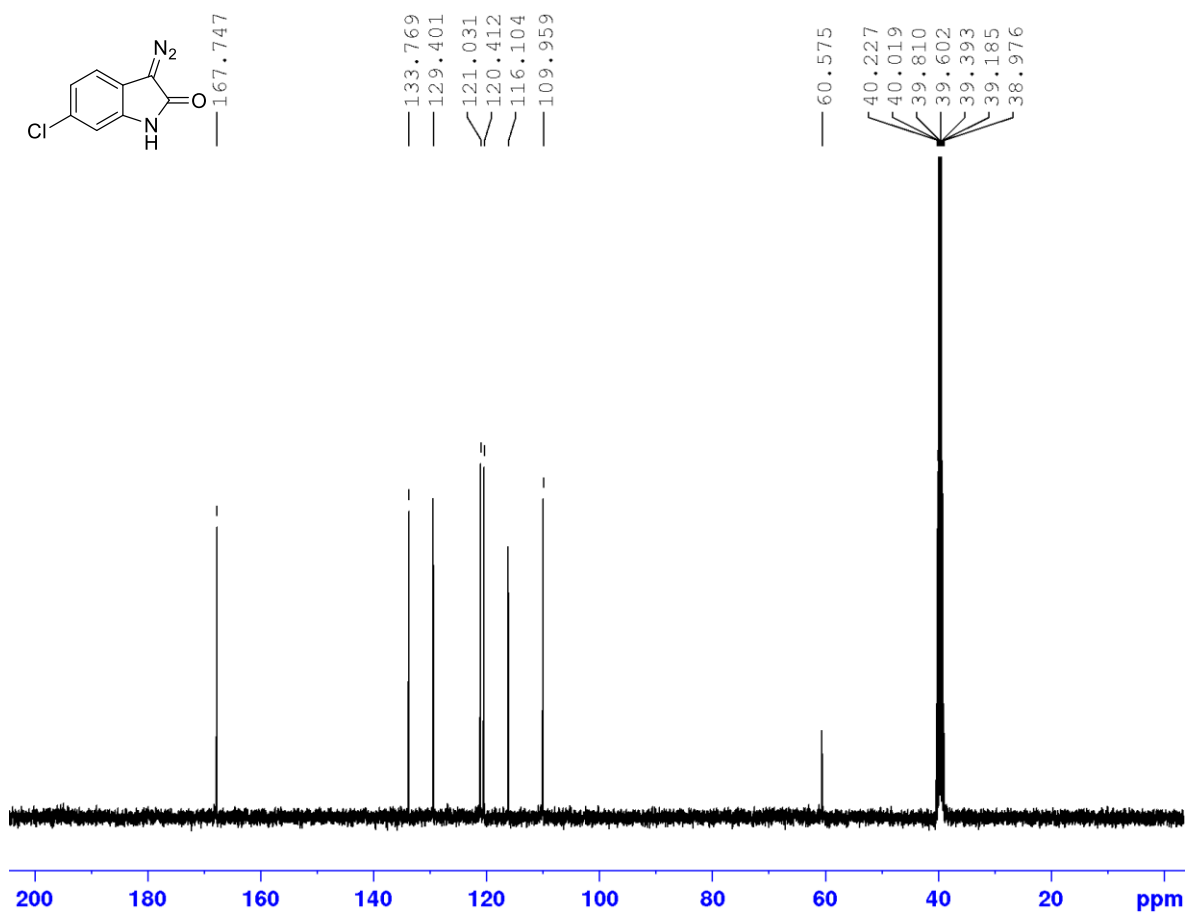
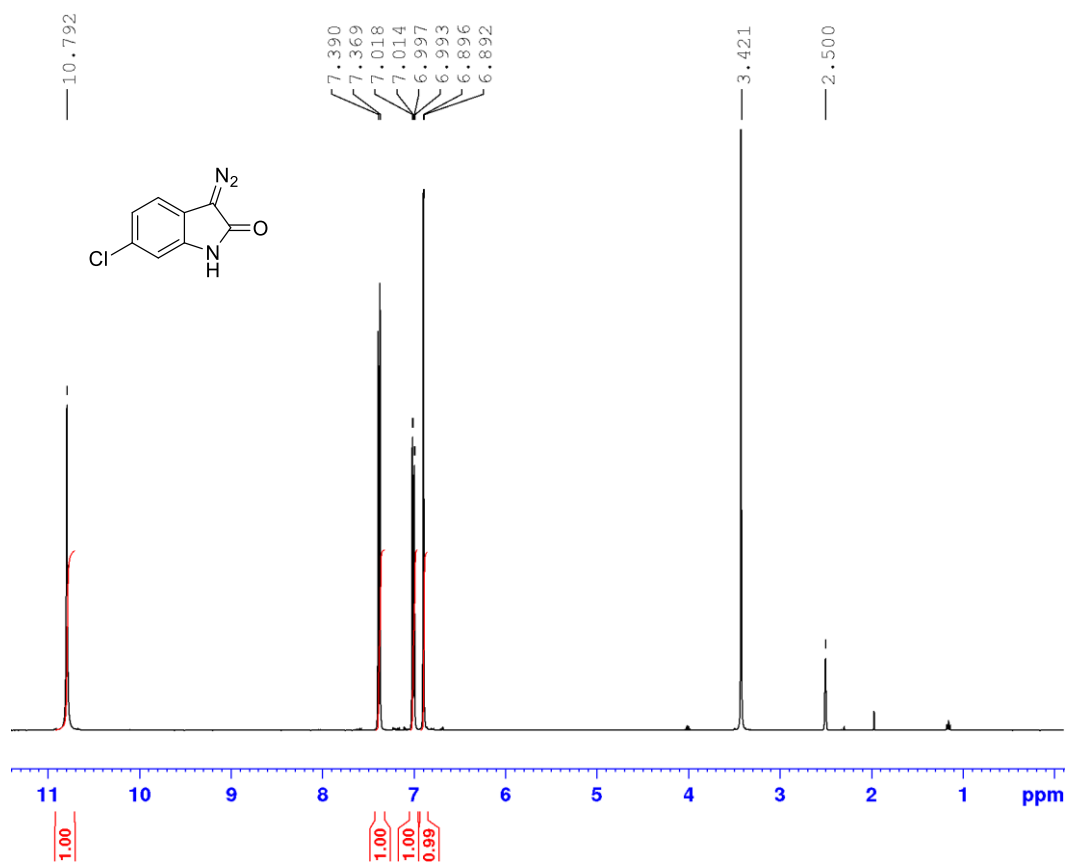
3-Hydroxy-*N*-methyloxindole (163 mg, 1 mmol) was dissolved in dry DMF (10 mL) under argon atmosphere. The solution was cooled to –20 °C and triflic anhydride (160 µL, 1 mmol) was added dropwise through a gas-tight syringe during 10 min. The reaction mixture was stirred for 40 min, during which time the temperature reached 0 °C. Then, thiobenzamide (**2a**, 137 mg, 1 mmol) was added and reaction mixture was further stirred overnight at room temperature. The reaction mixture was evaporated and the residue was dissolved in DCM (20 mL) and evaporated with silica gel (5 g). Preparative flash chromatography (silica gel cartridge; mobile phase DCM/EtOAc + 3 % TEA, gradient of EtOAc 0–20% during 20 min) gave 220 mg (87%) of yellow crystals of **11** with m.p. 174 – 176.5 °C. ¹H NMR (400 MHz, DMSO-*d*₆) δ 9.47 (bd, *J* 3.7 Hz, 1H, ½NH₂), 8.16 (bd, *J* 3.4 Hz, 1H, ½NH₂), 7.43-7.67 (m, 5H, Ar-H), 6.82-7.02 (m, 2H, Ar-H), 6.57-6.65 (m, 1H, Ar-H), 6.10 (d, *J* 7.8 Hz, 1H, Ar-H), 3.27 (s, 3H, CH₃) corresponds to Ref. [1].

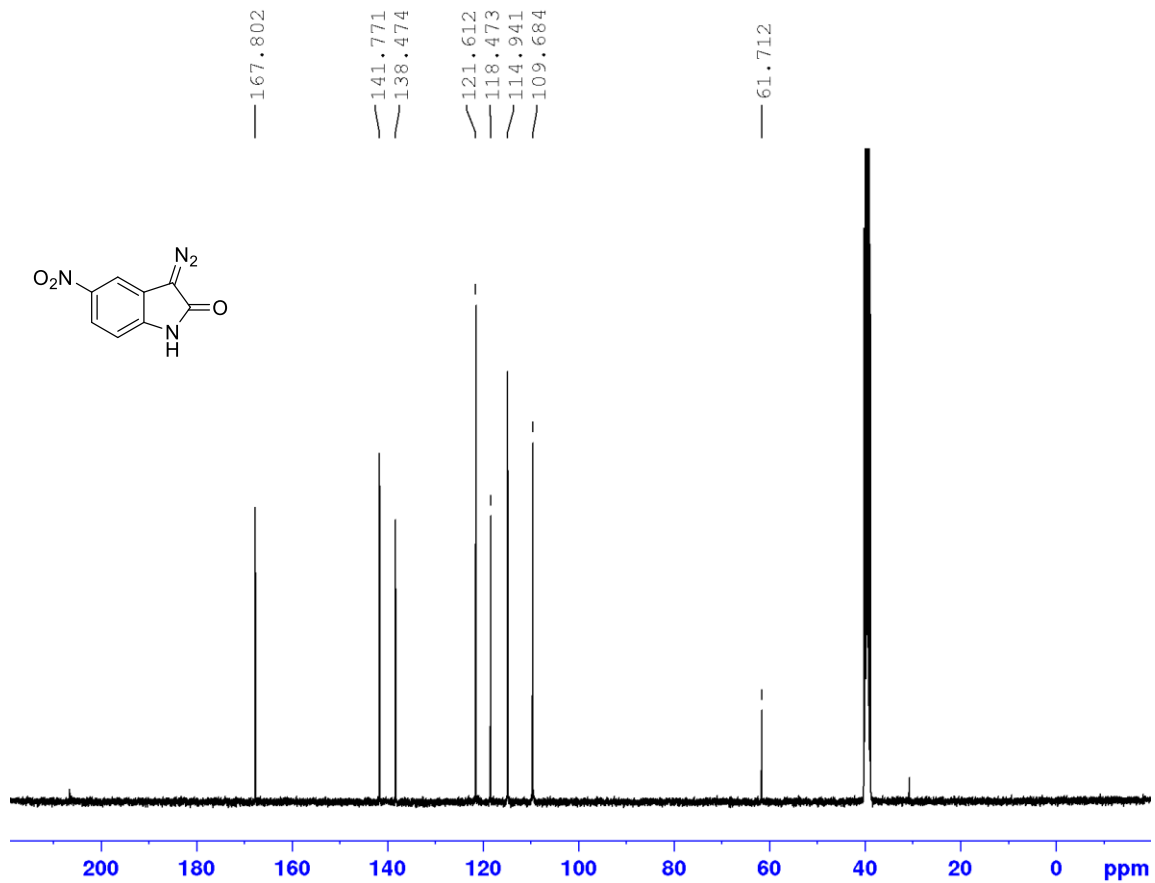
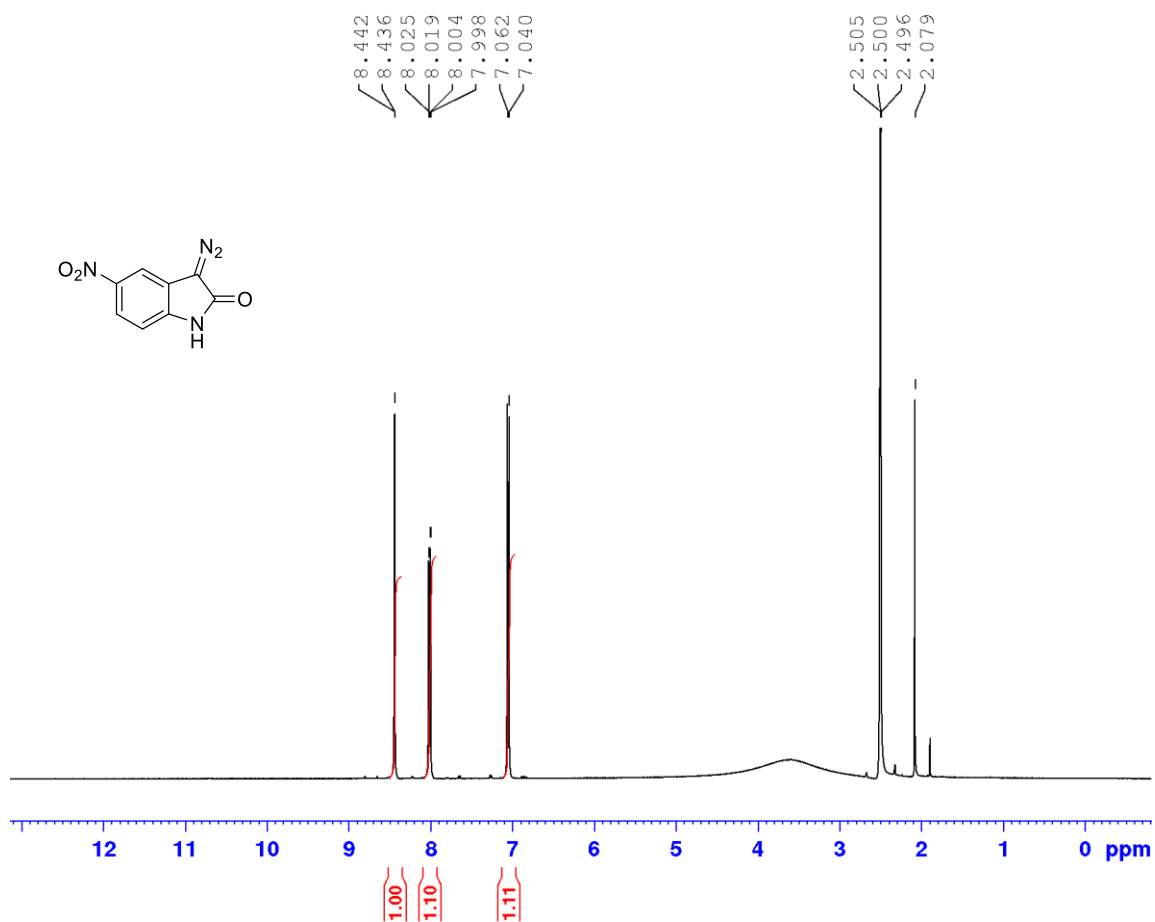
The Rhe reaction of 3-hydroxyoxindole with thiobenzamide (**2a**) giving (*Z*)-3-[amino(phenyl)methylidene]-1,3-dihydro-2*H*-indol-2-one (**5aa**) was carried out in the same manner.

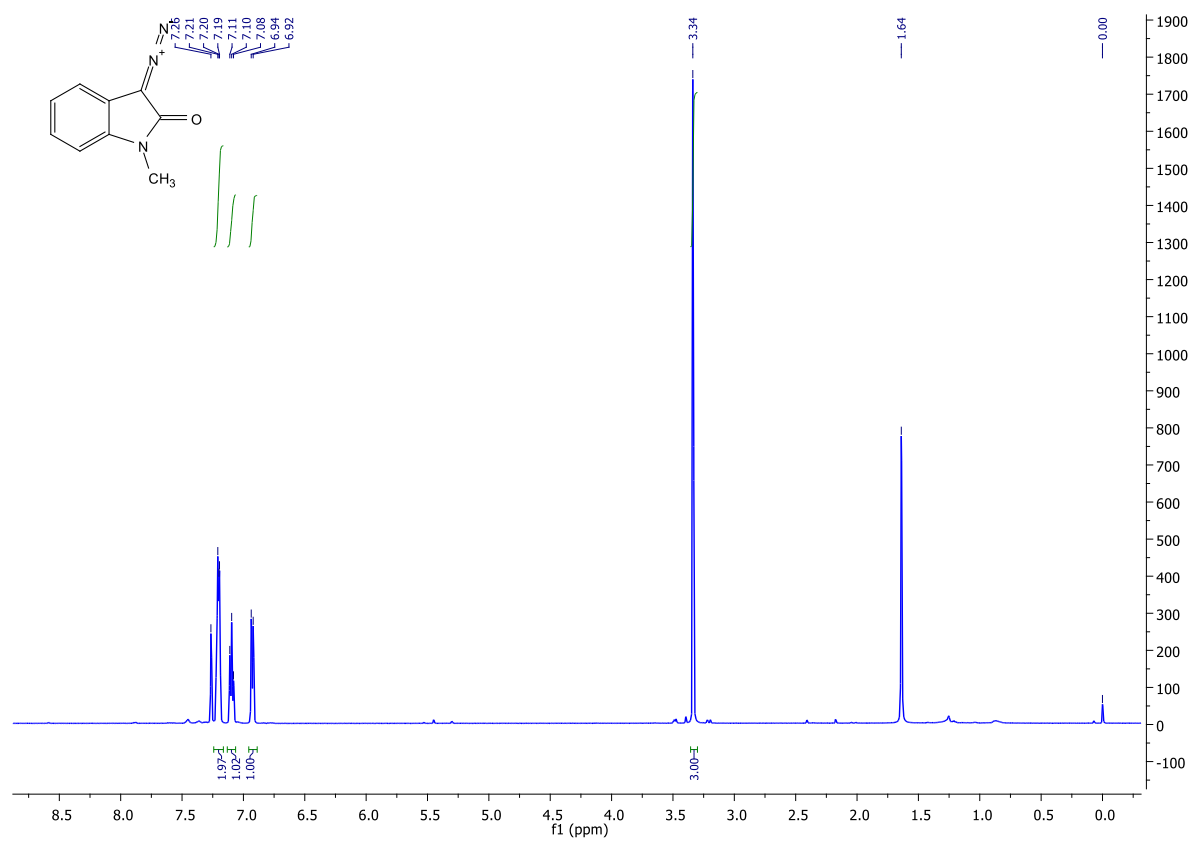


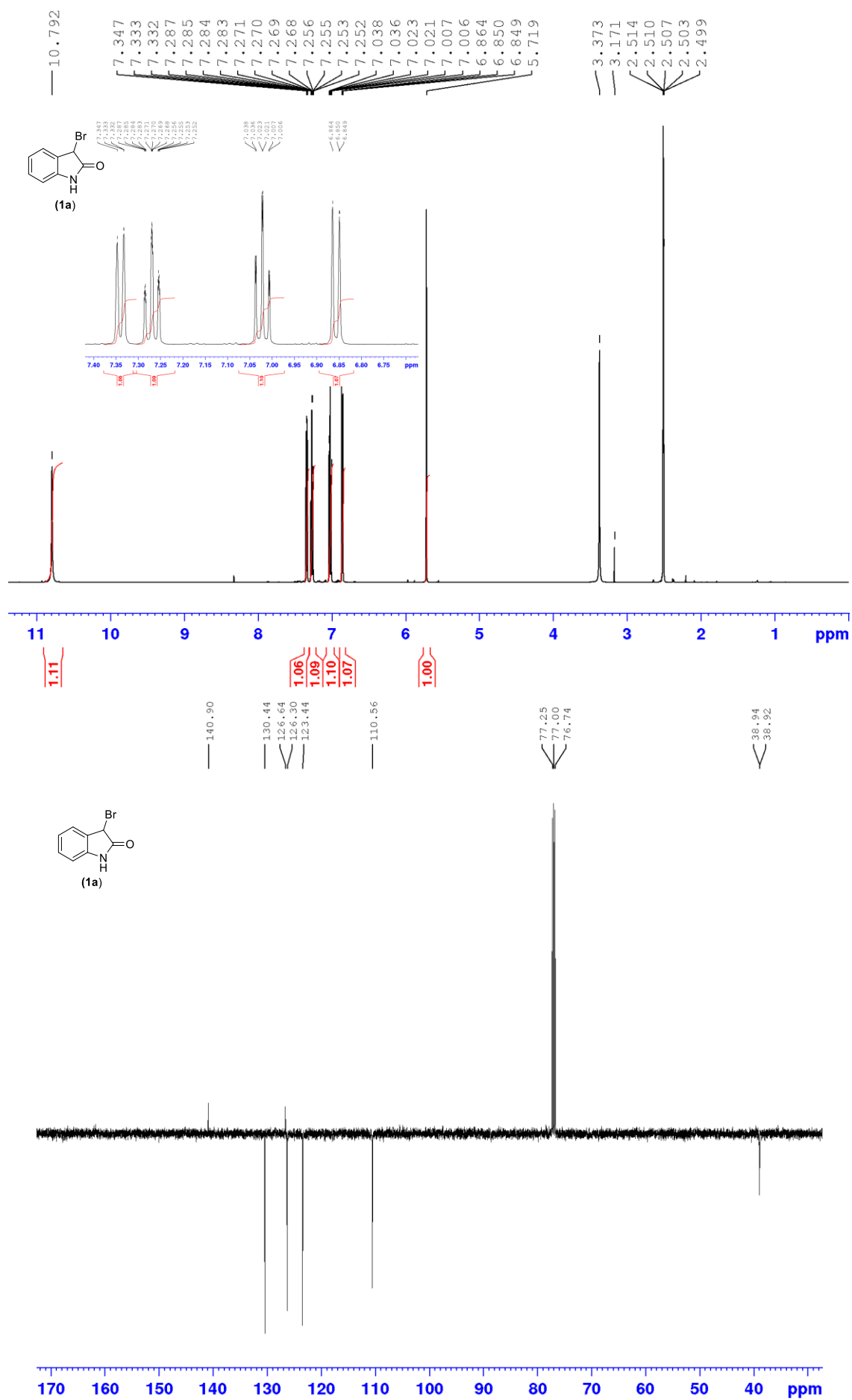


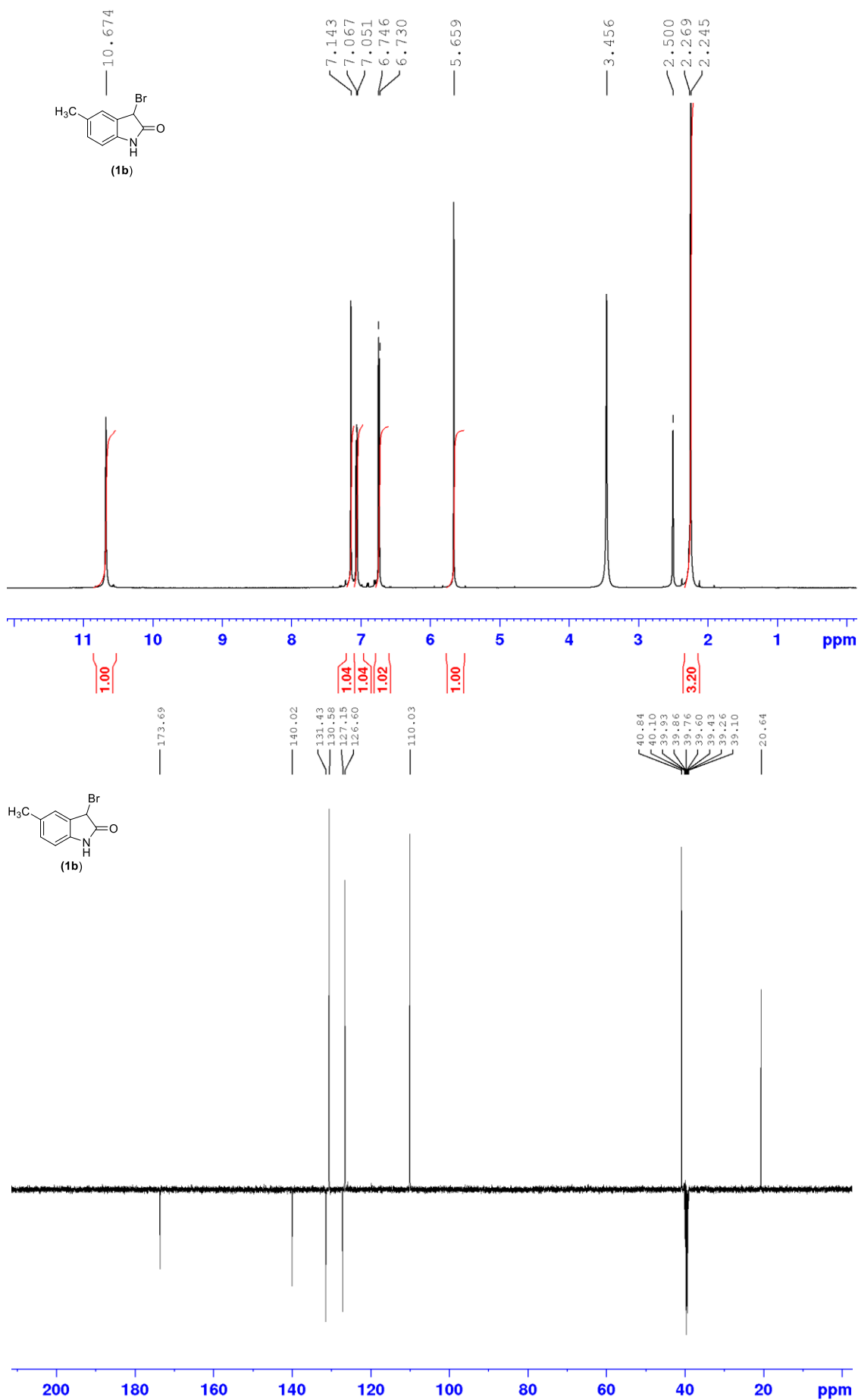


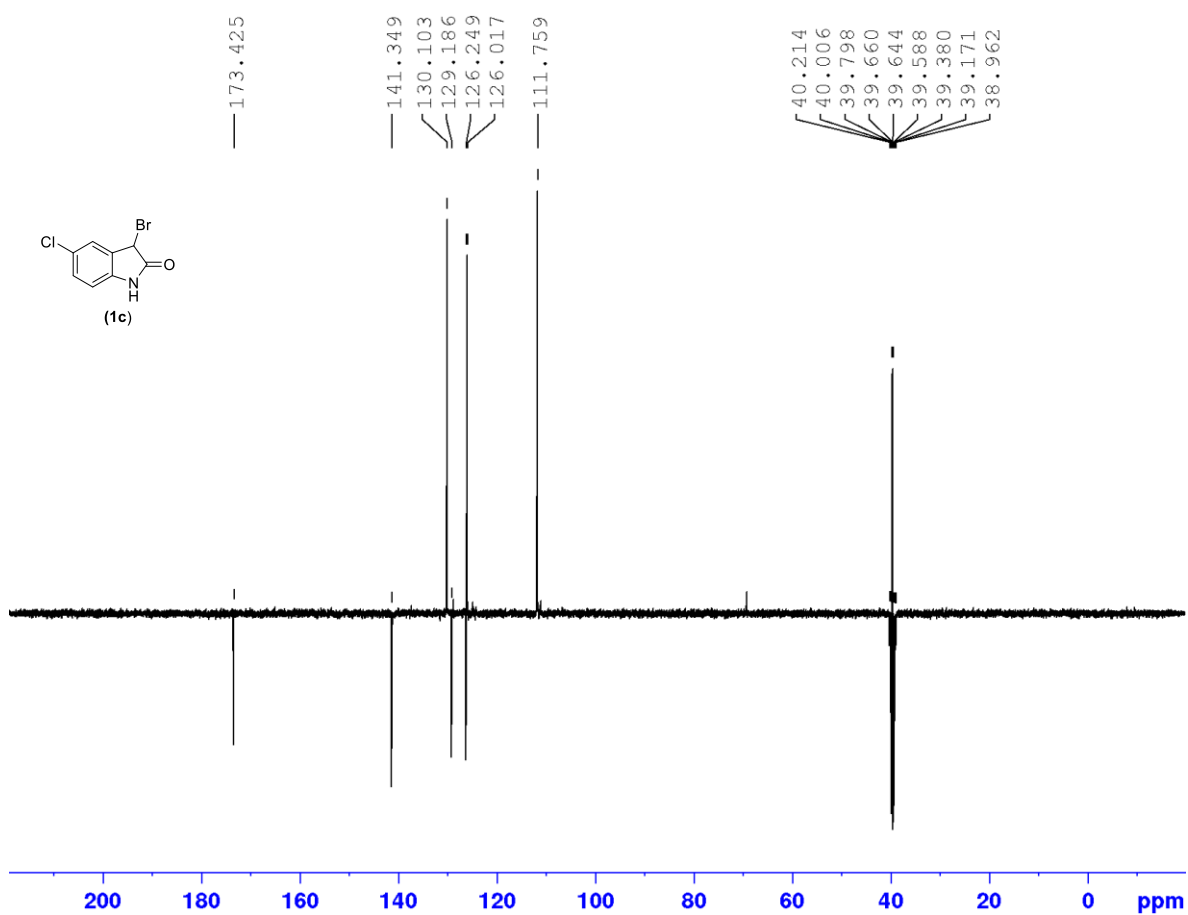
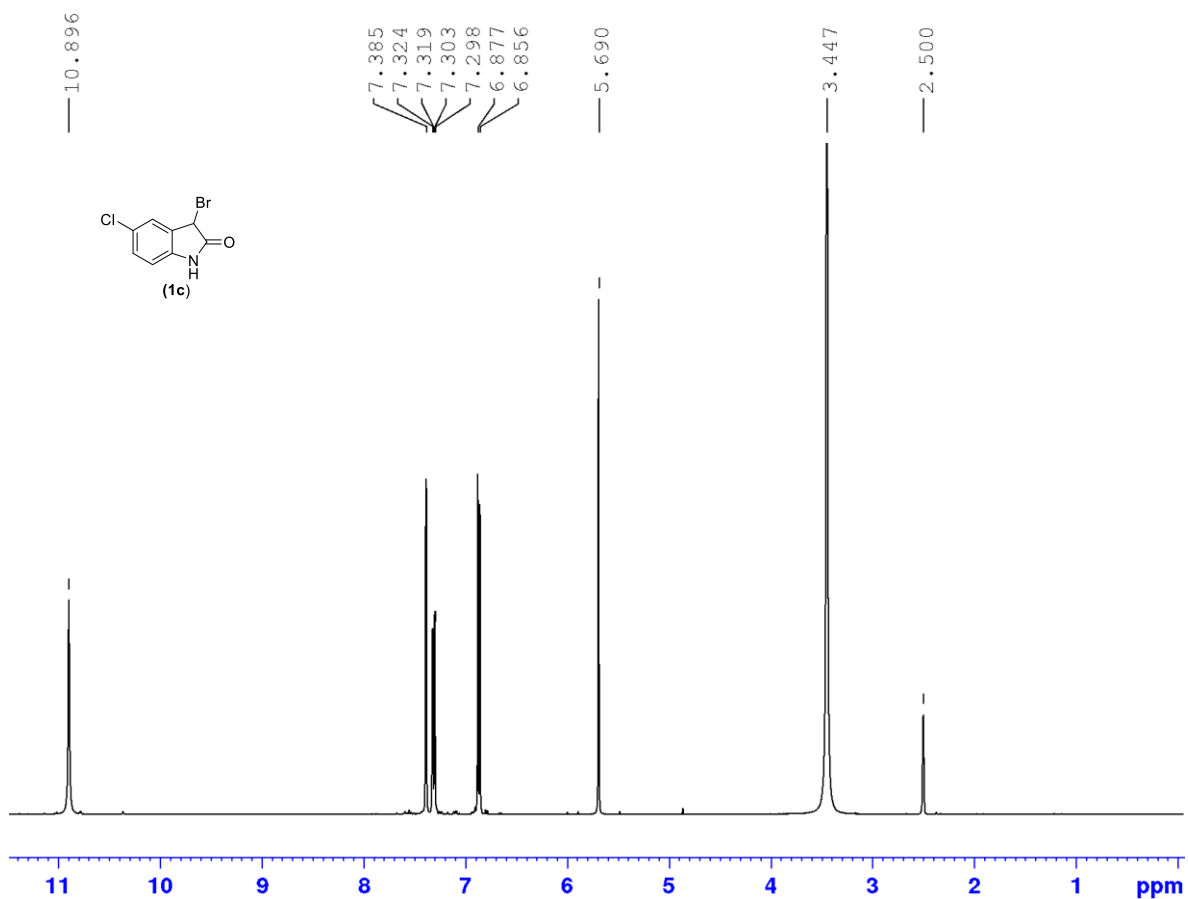


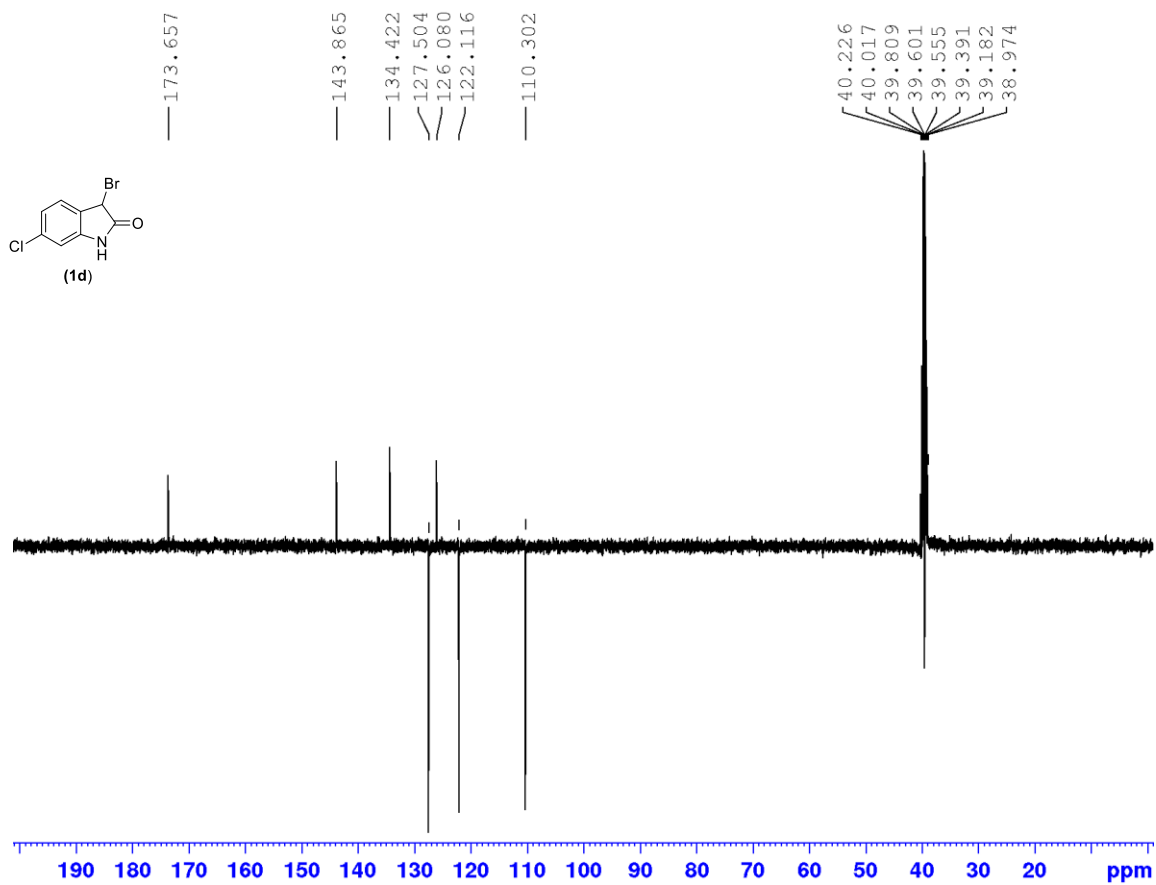
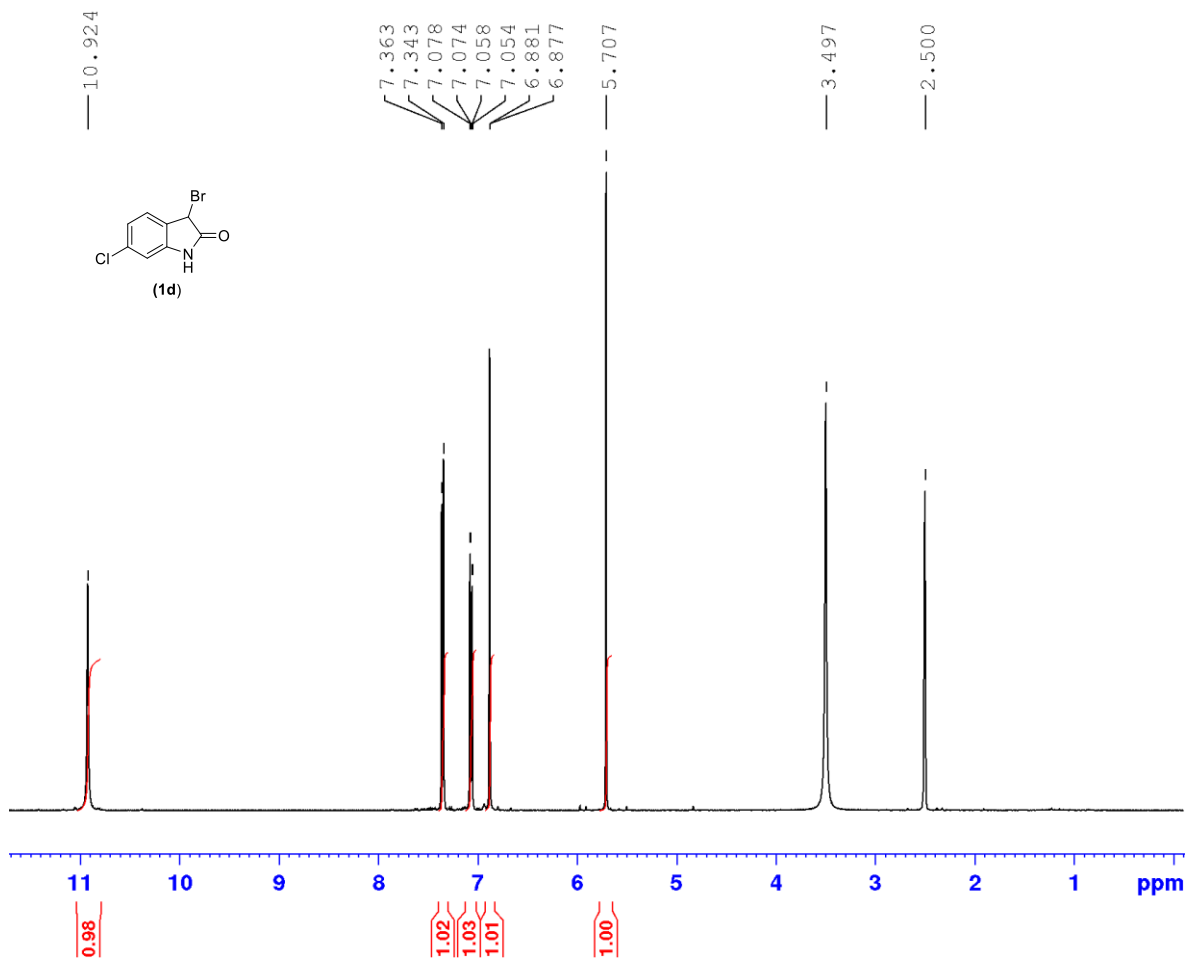


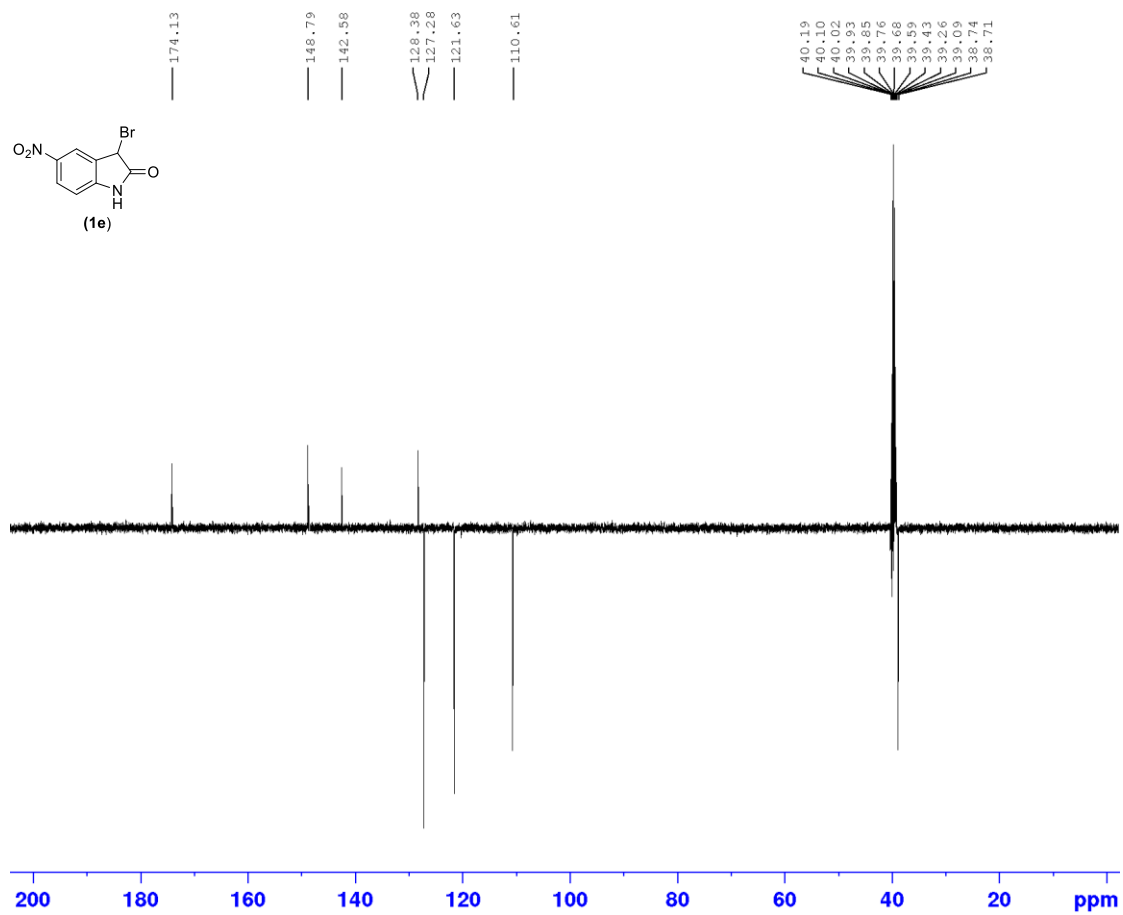
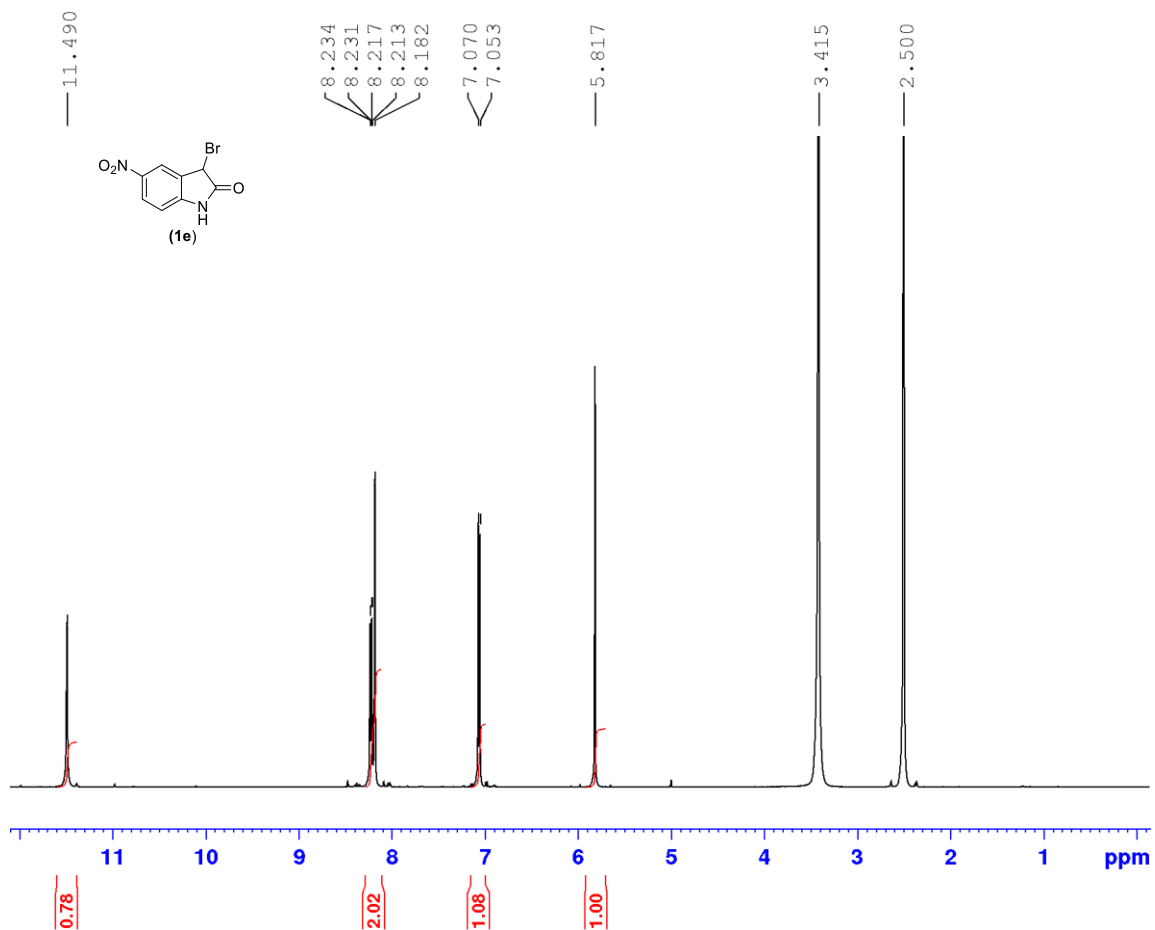


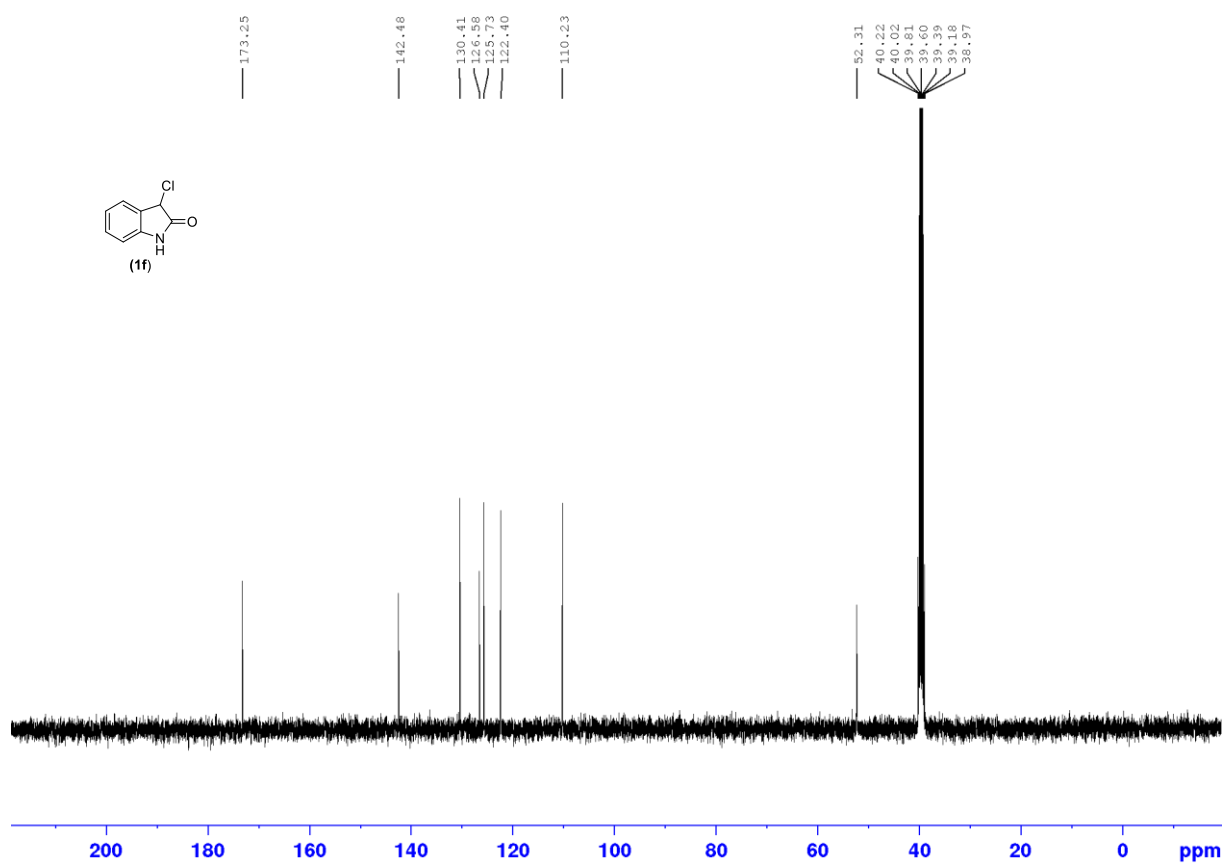
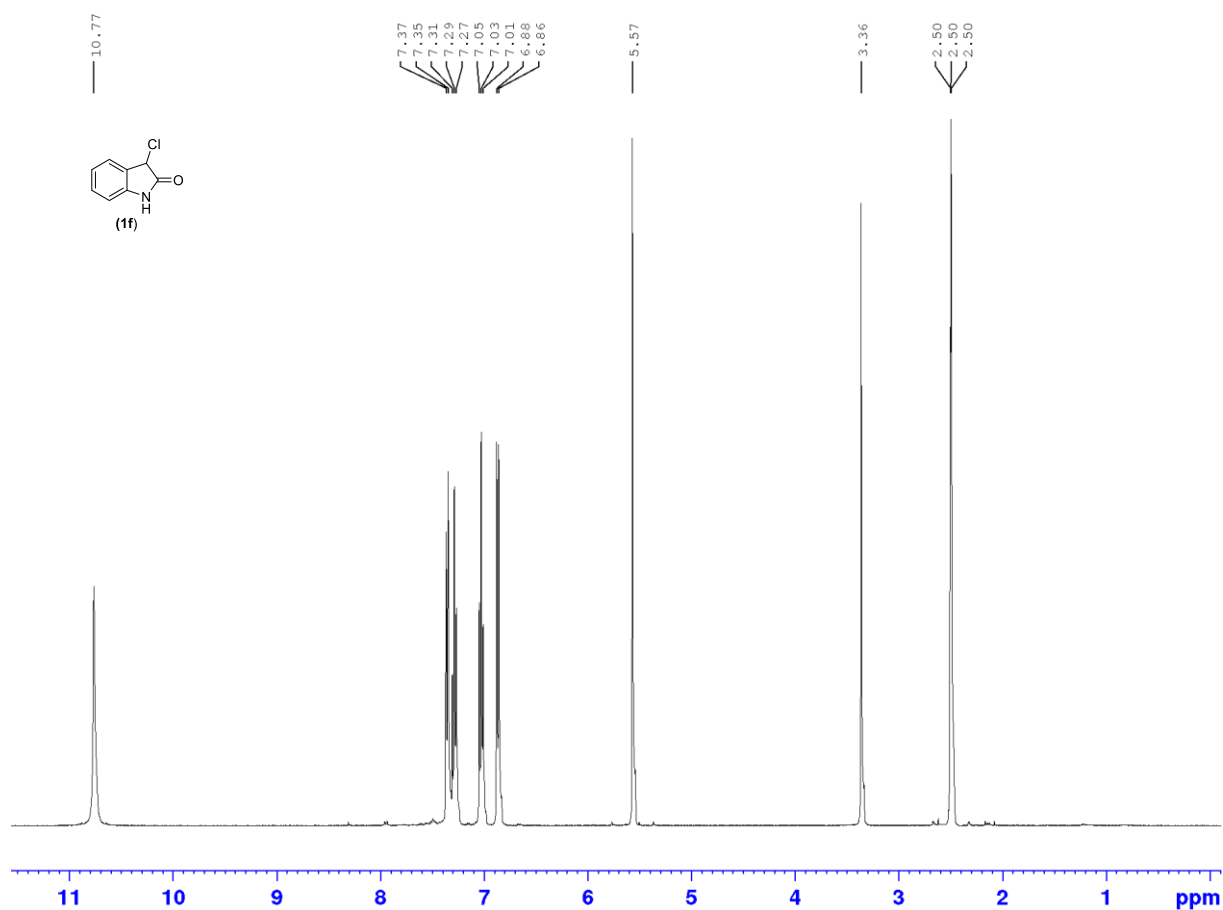


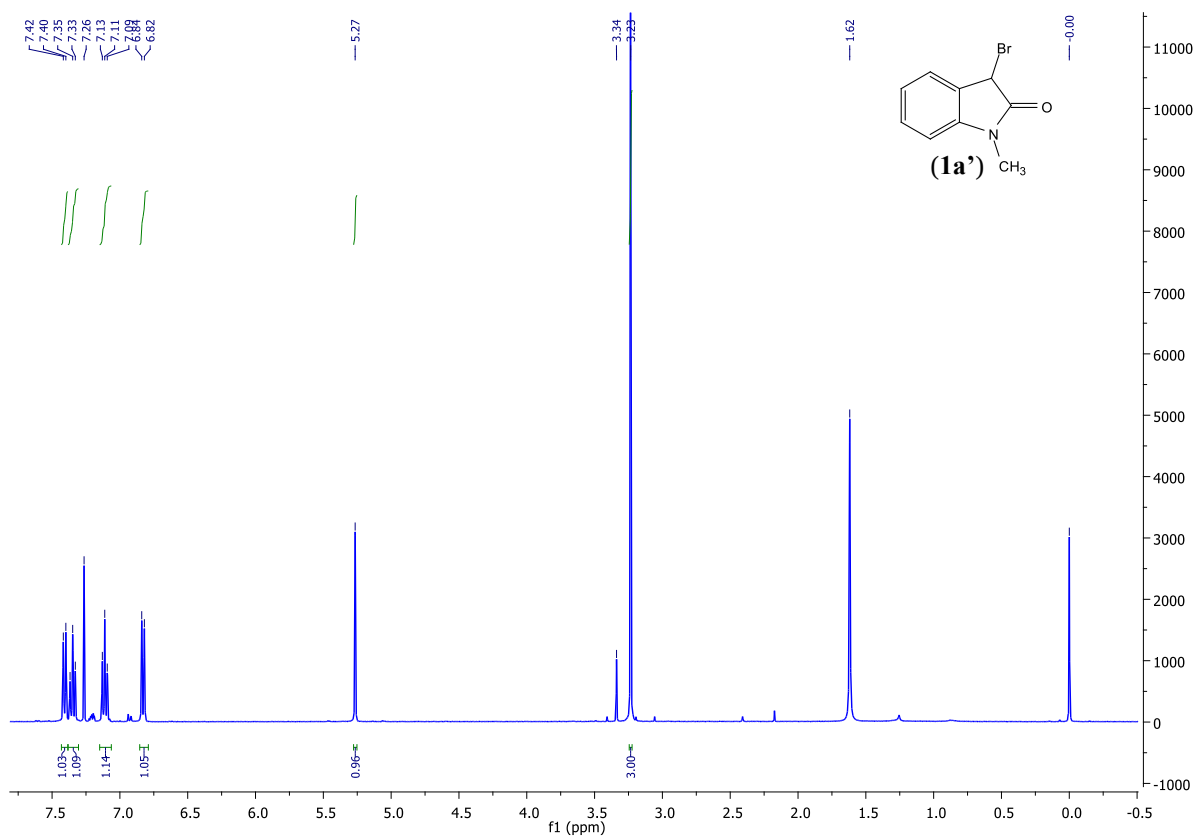
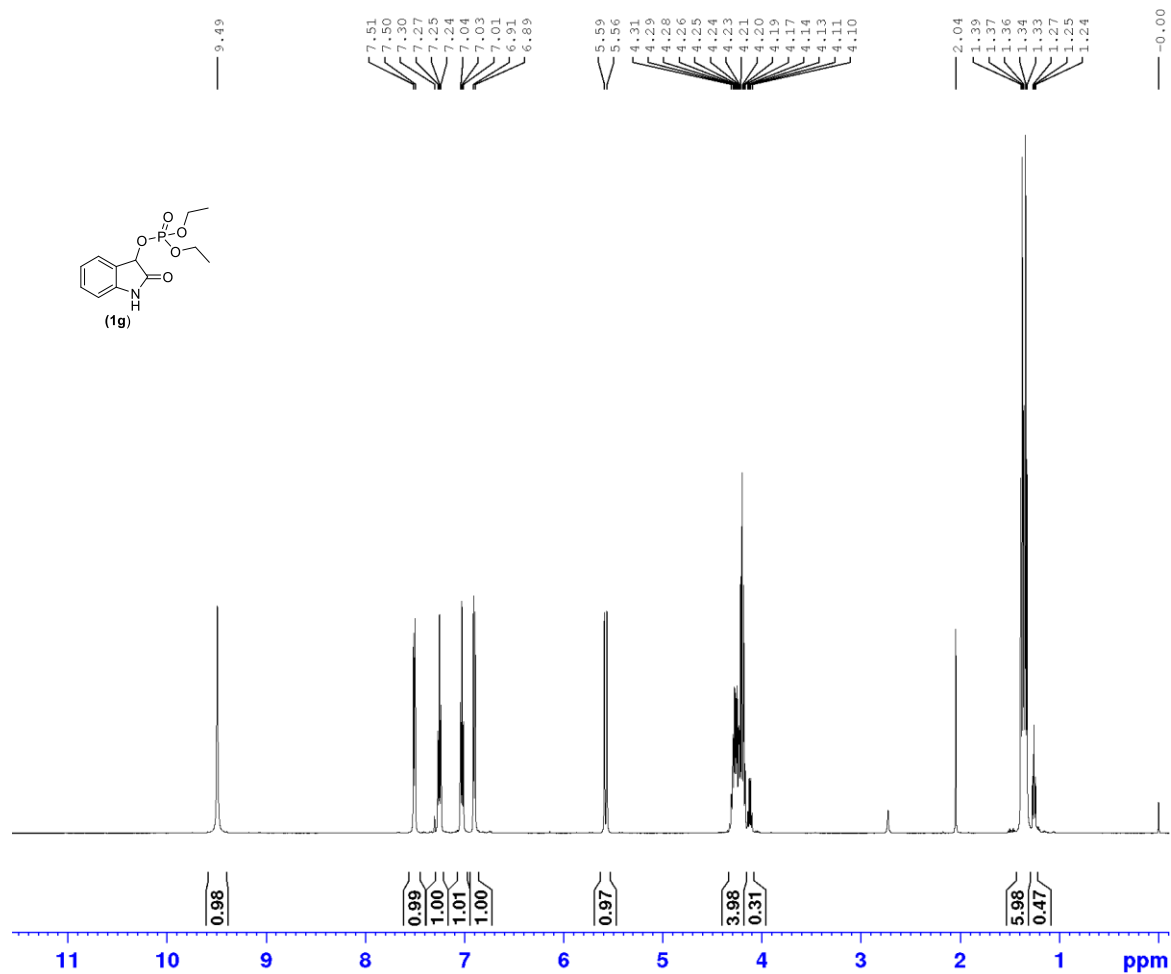


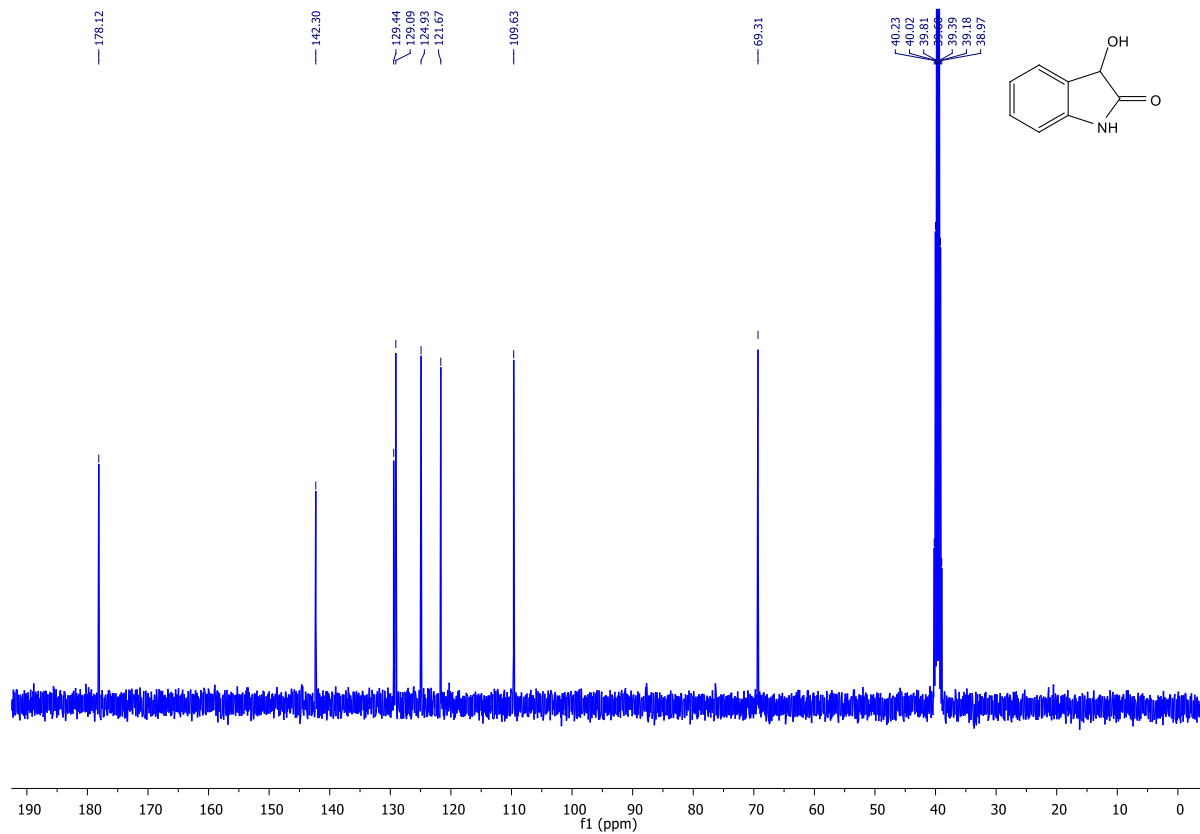
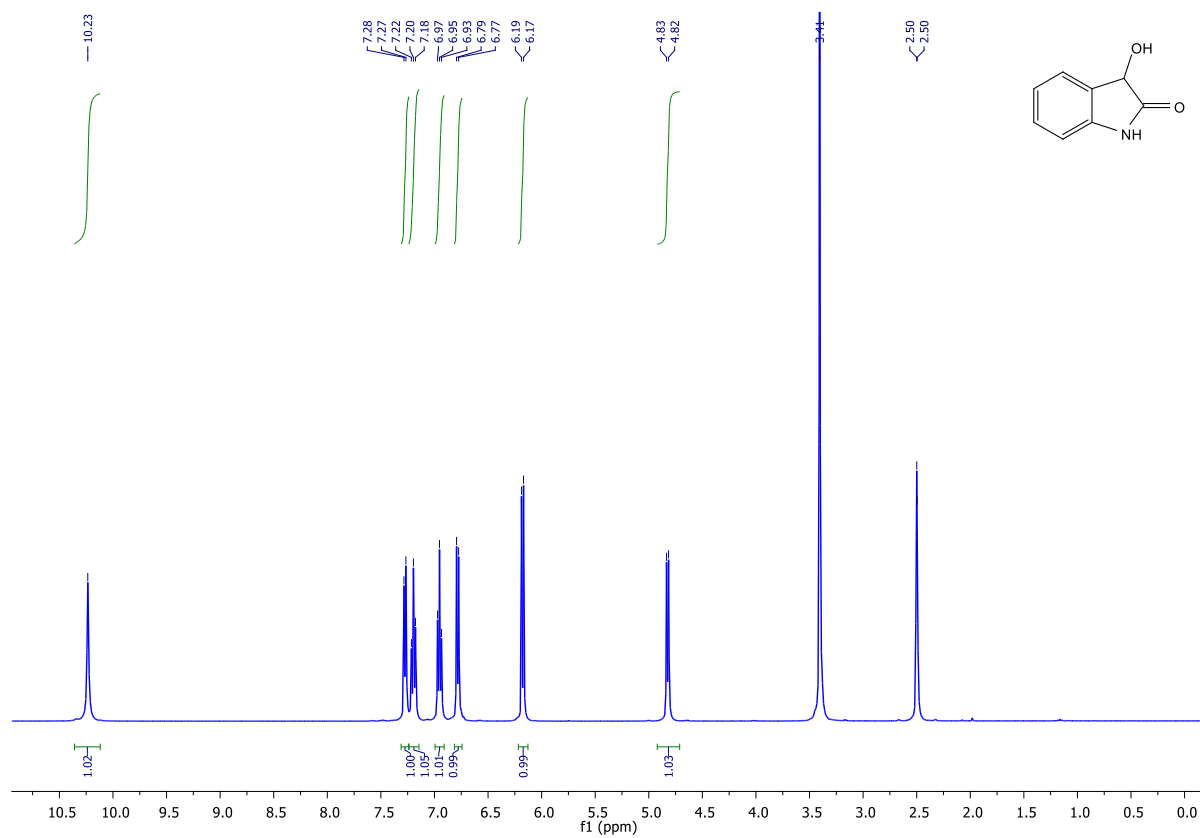


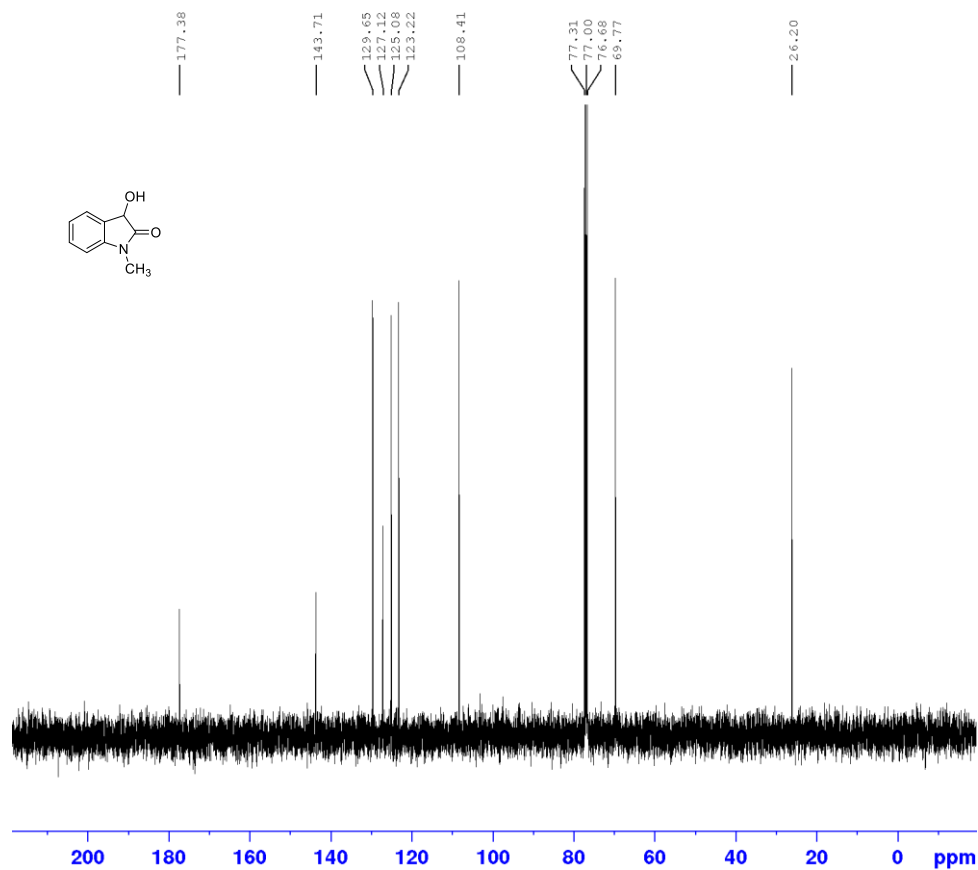
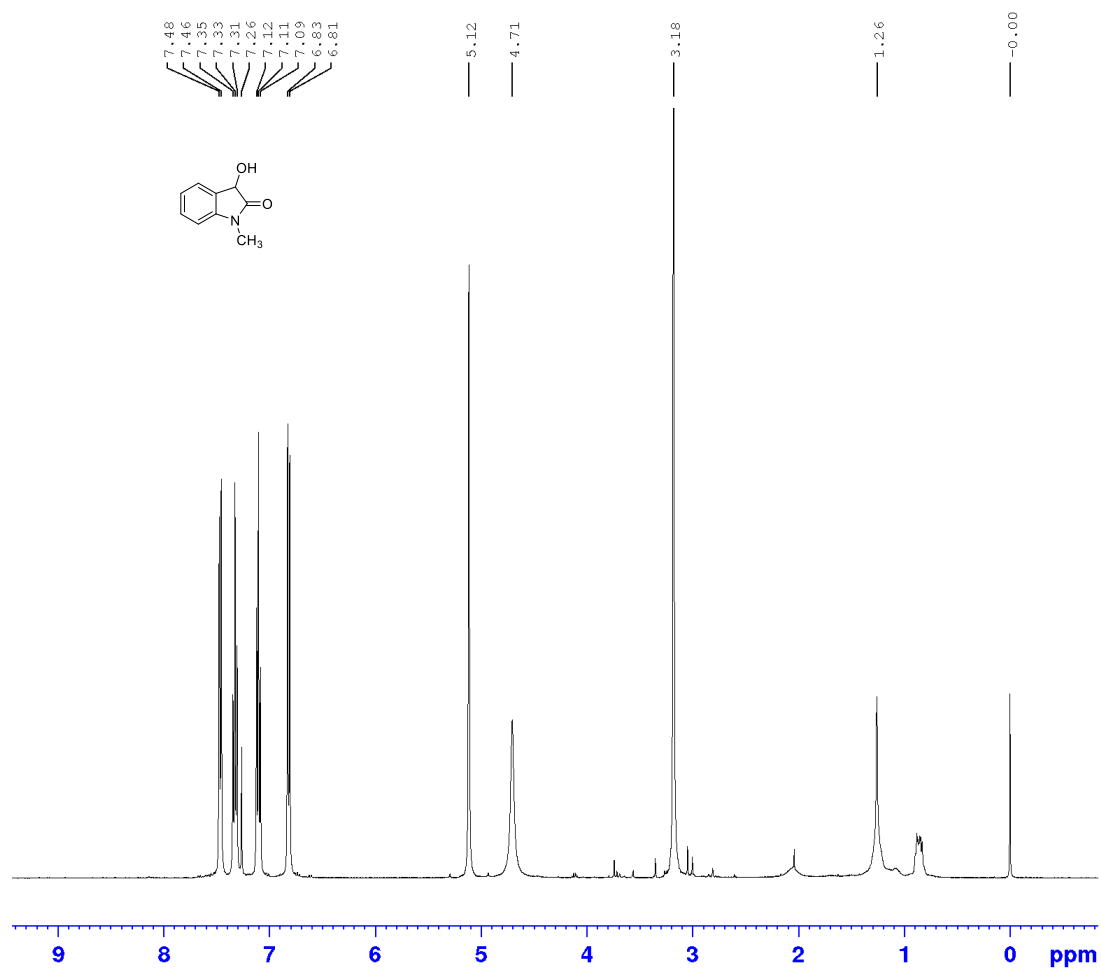


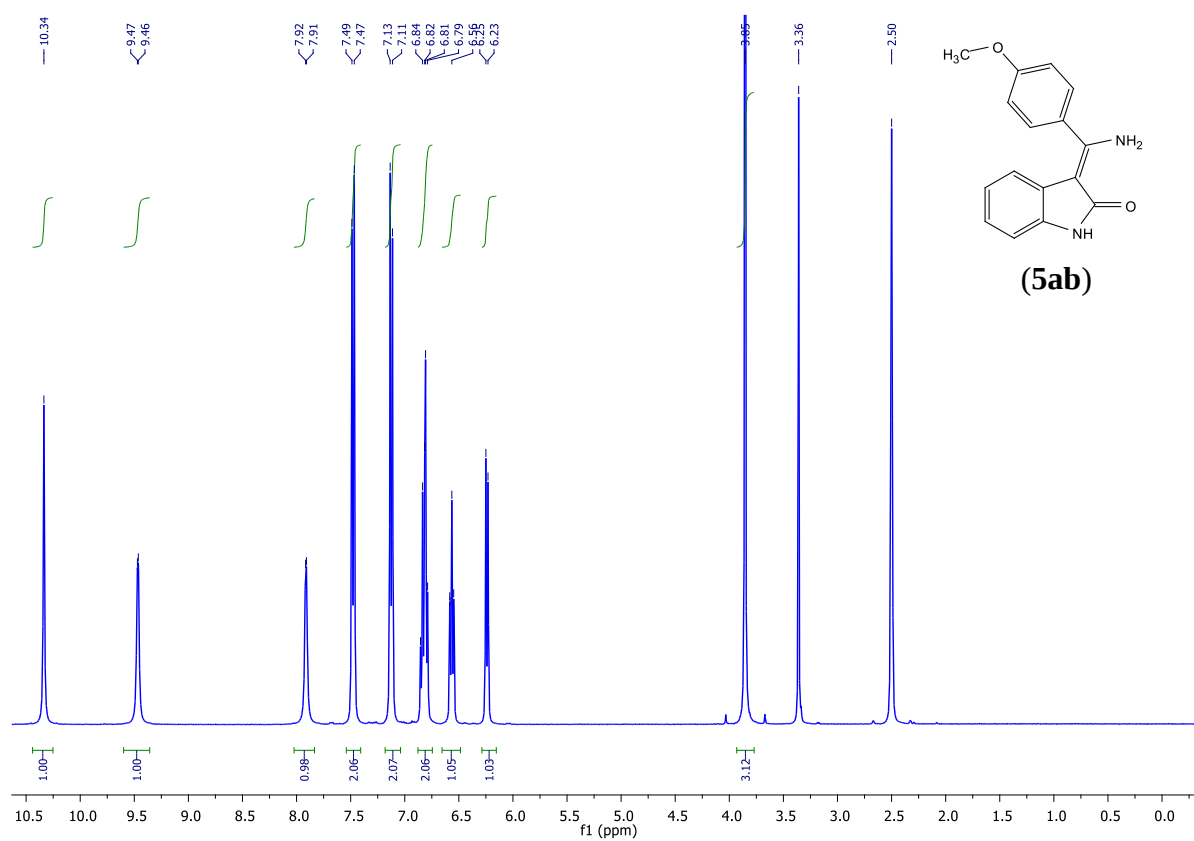
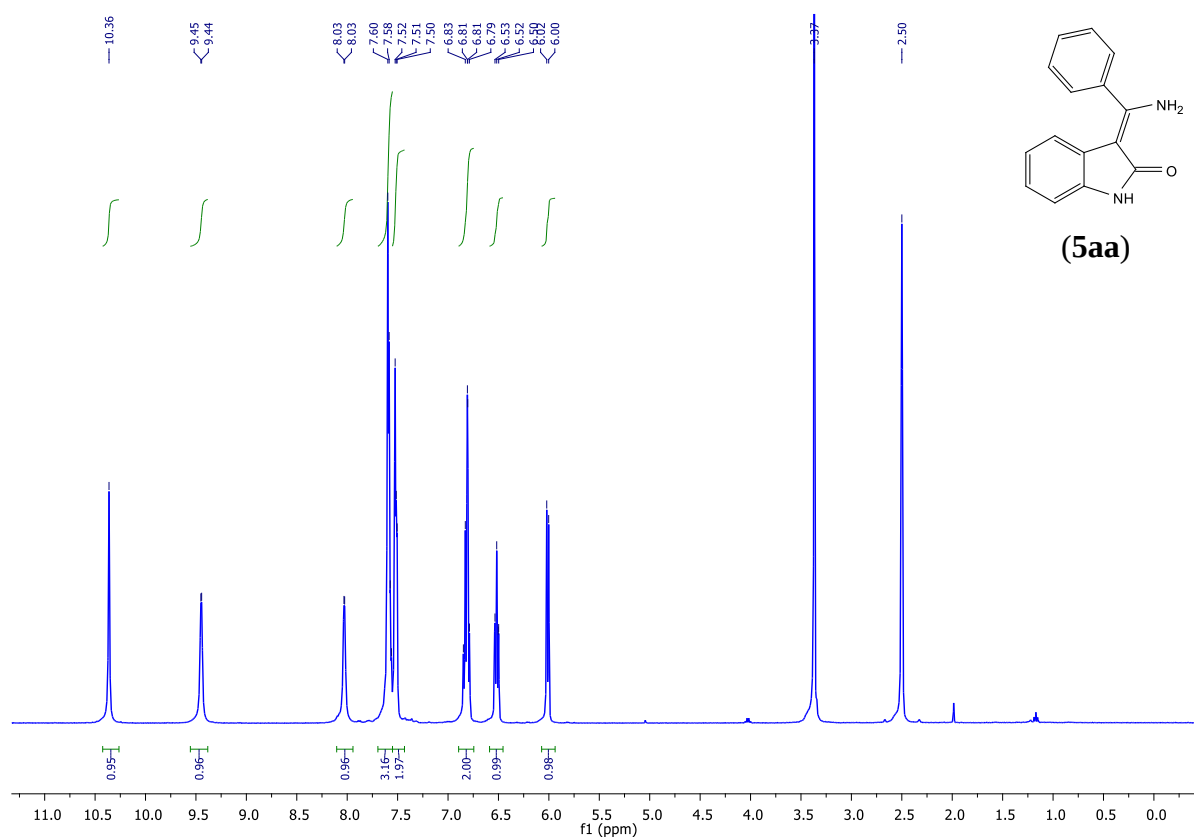


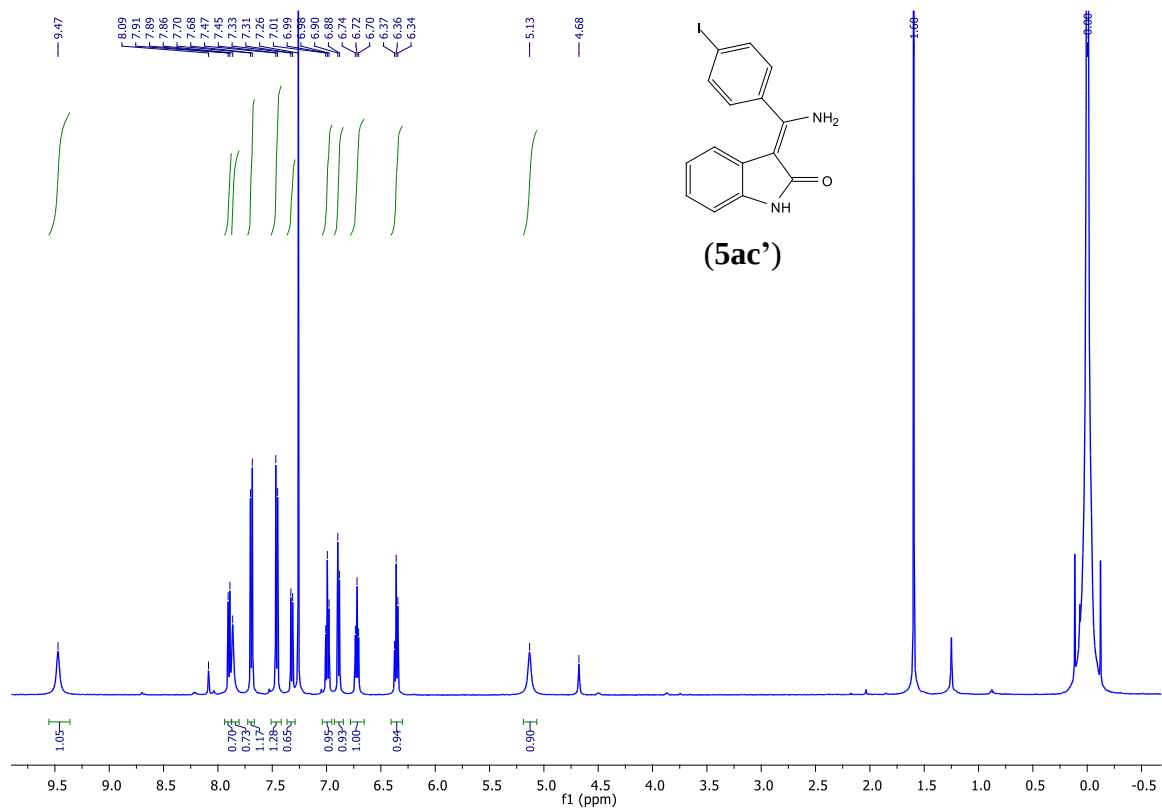
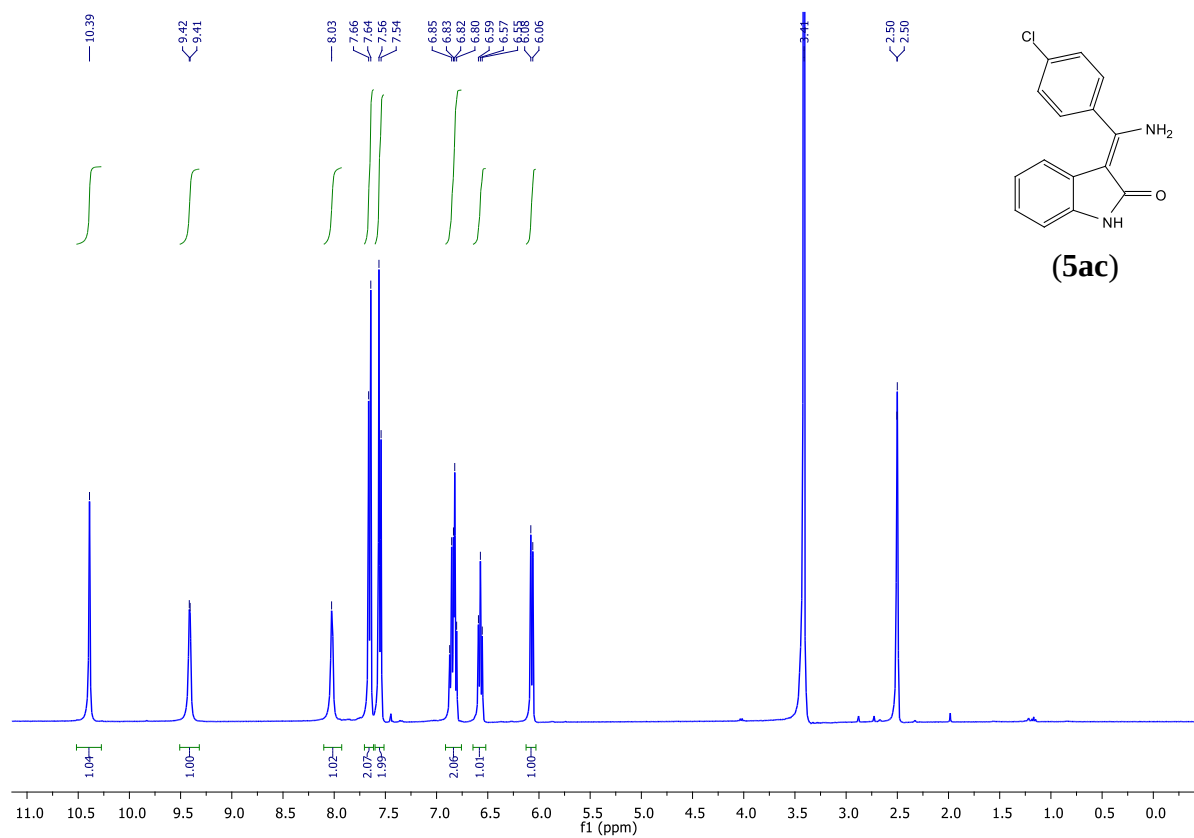


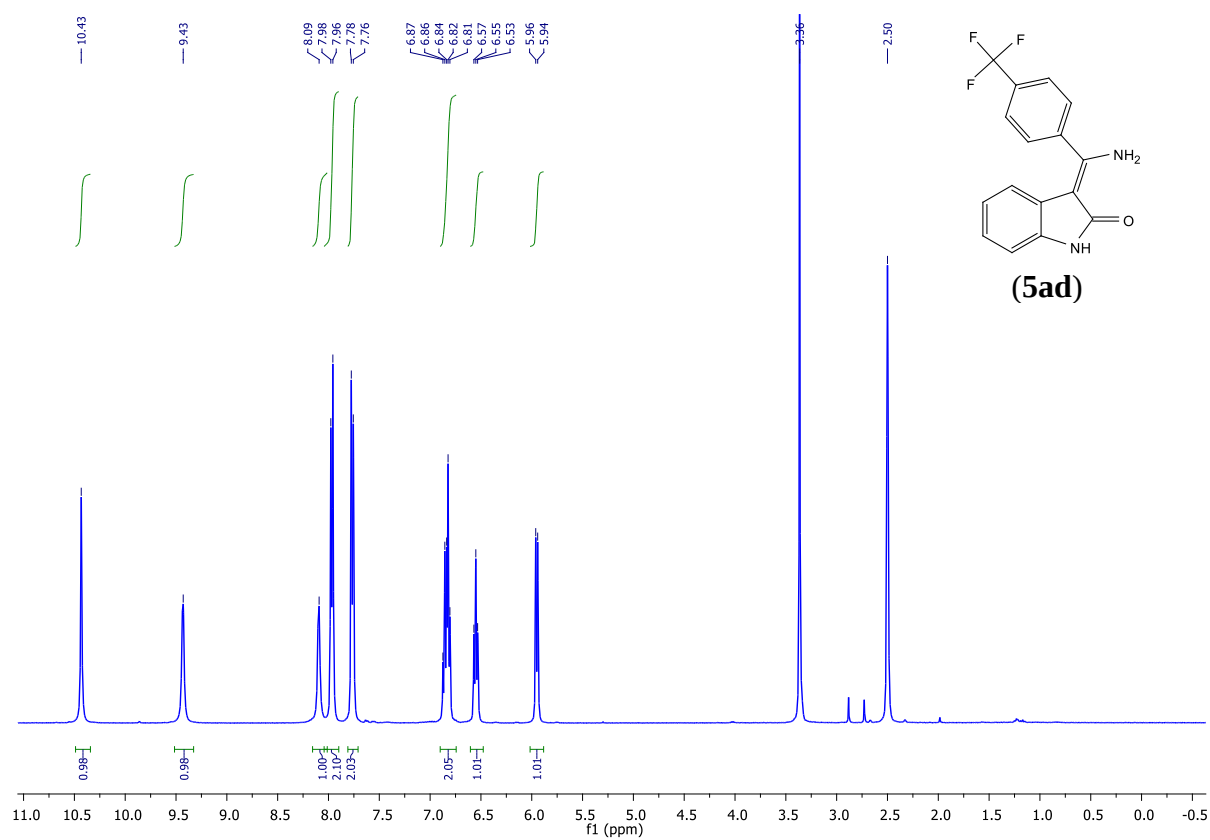
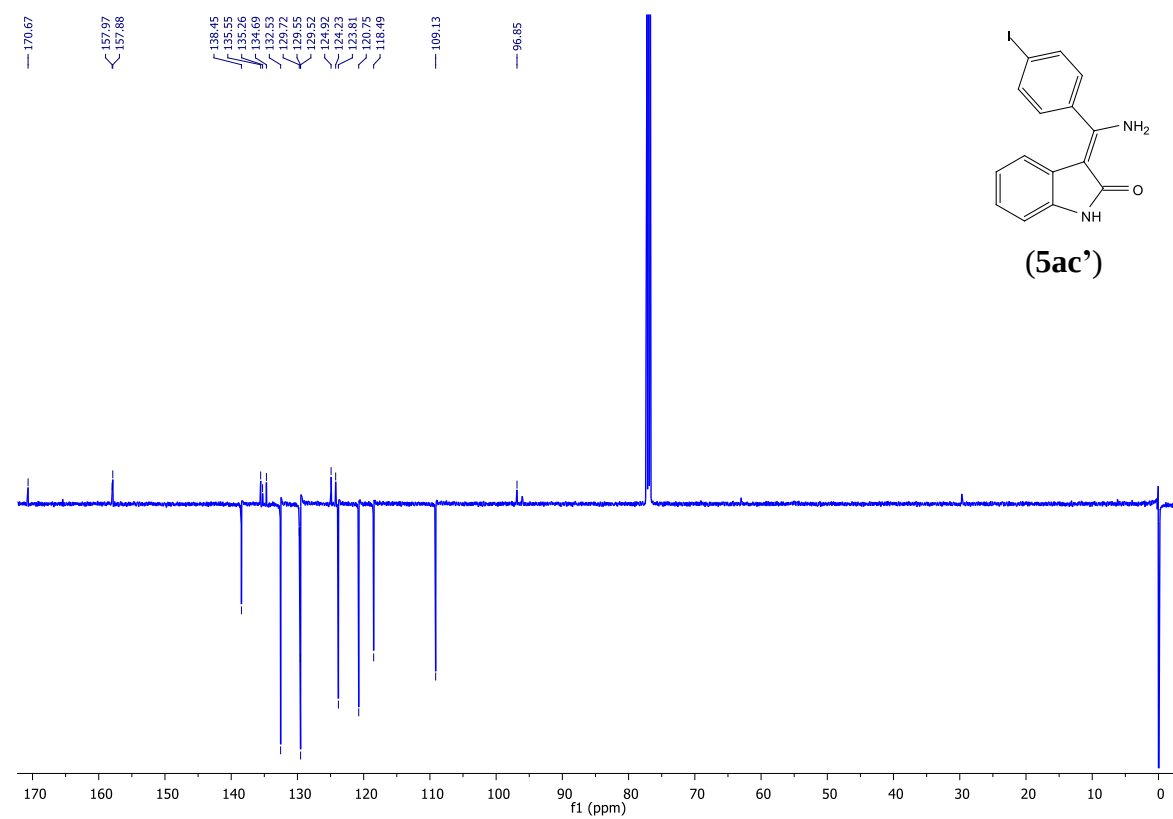


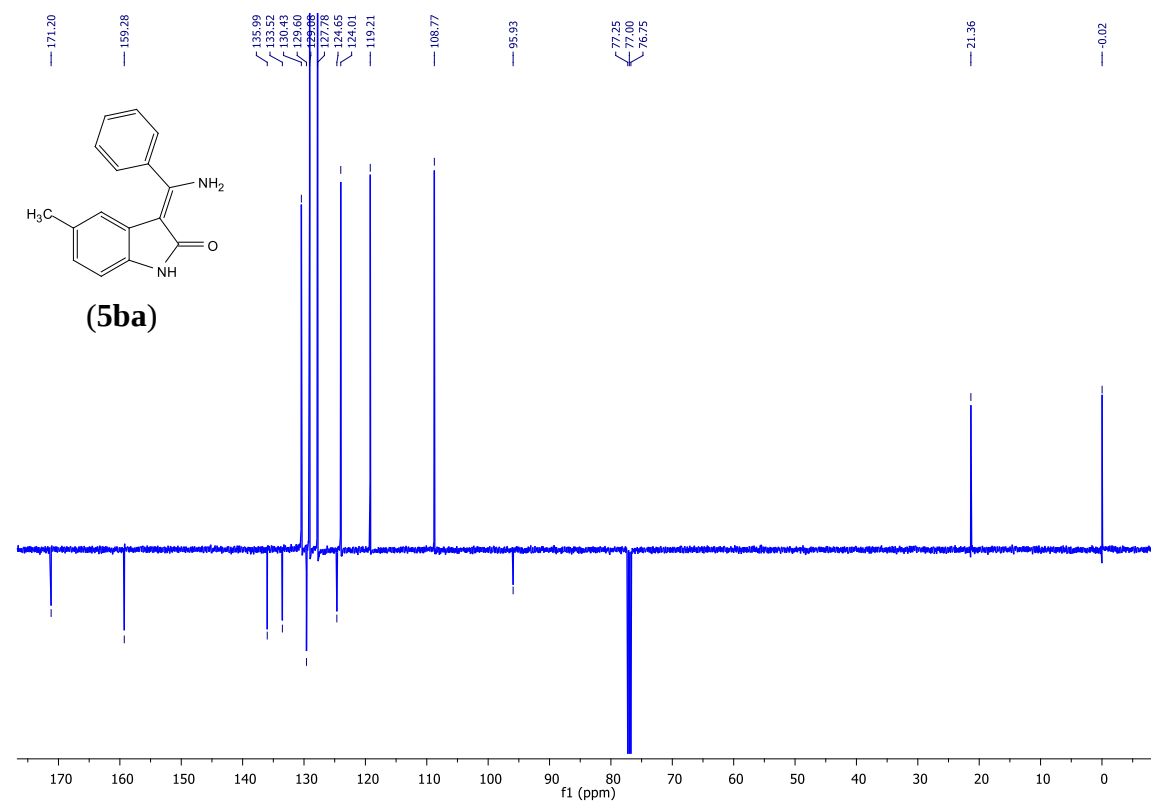
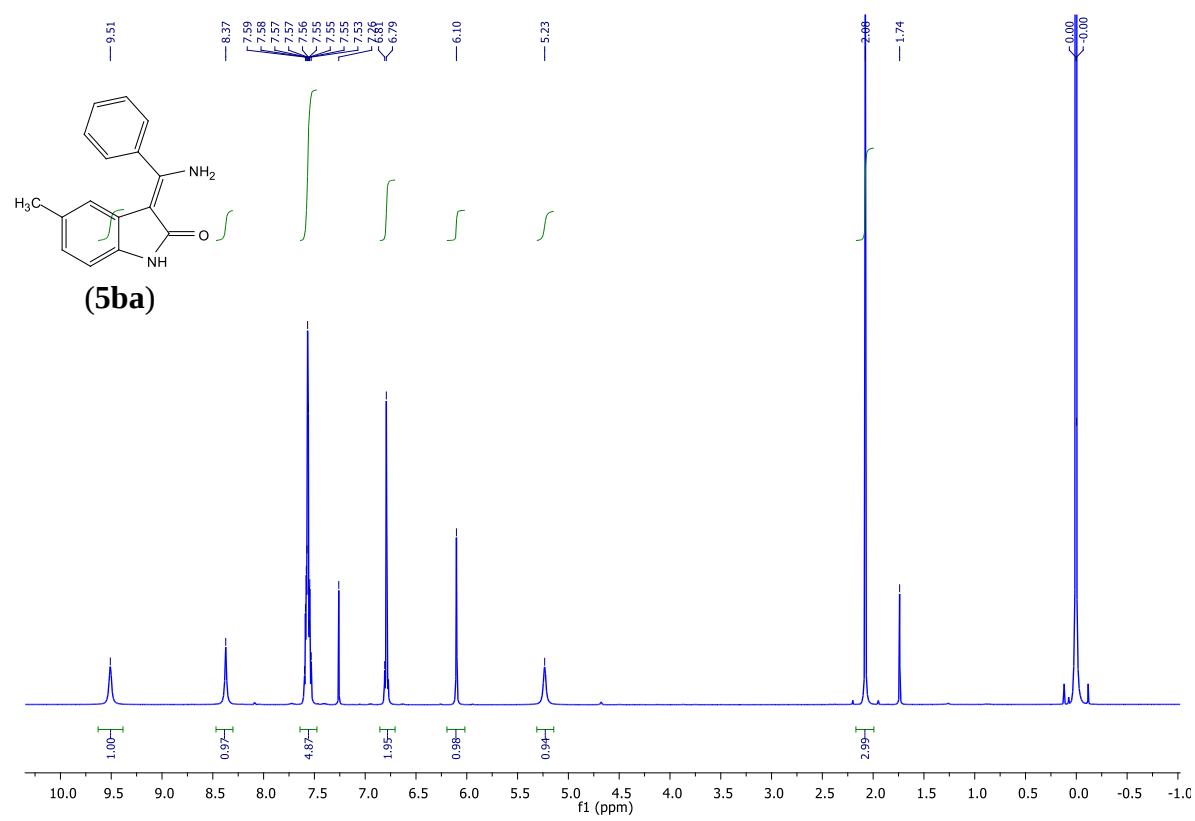


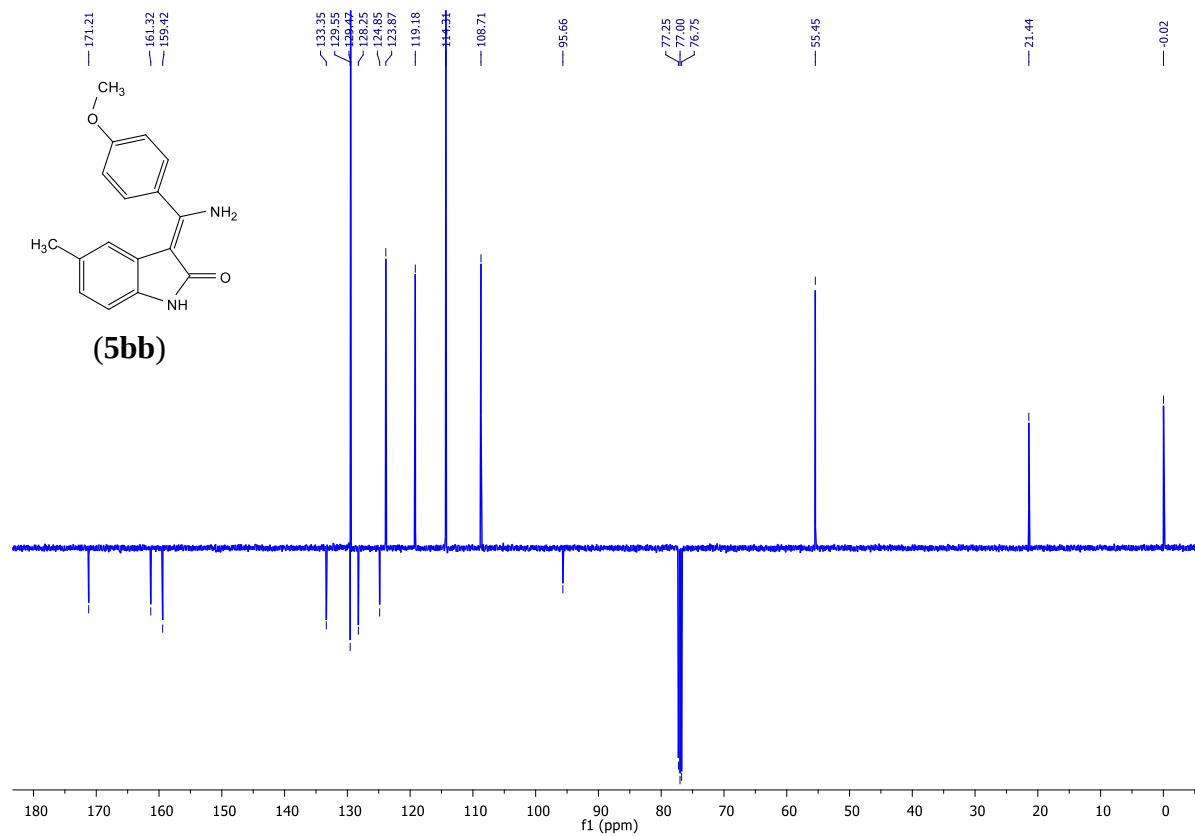
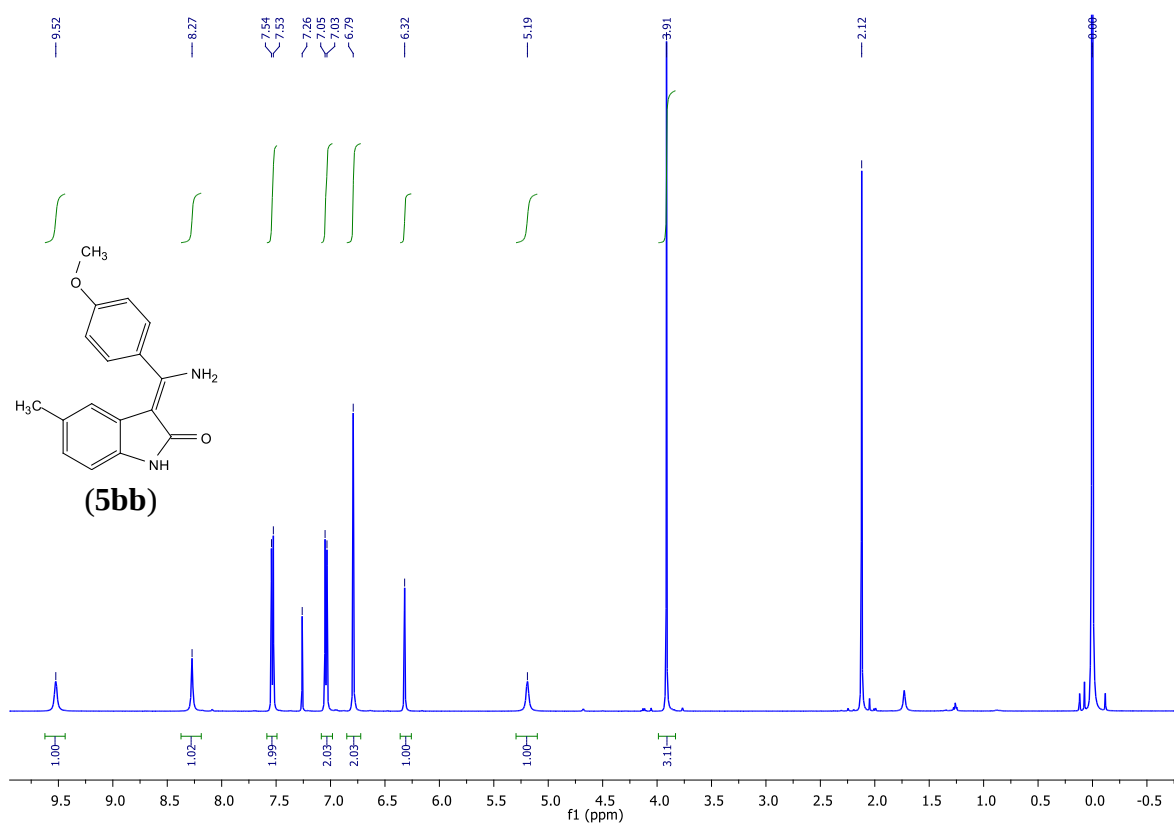


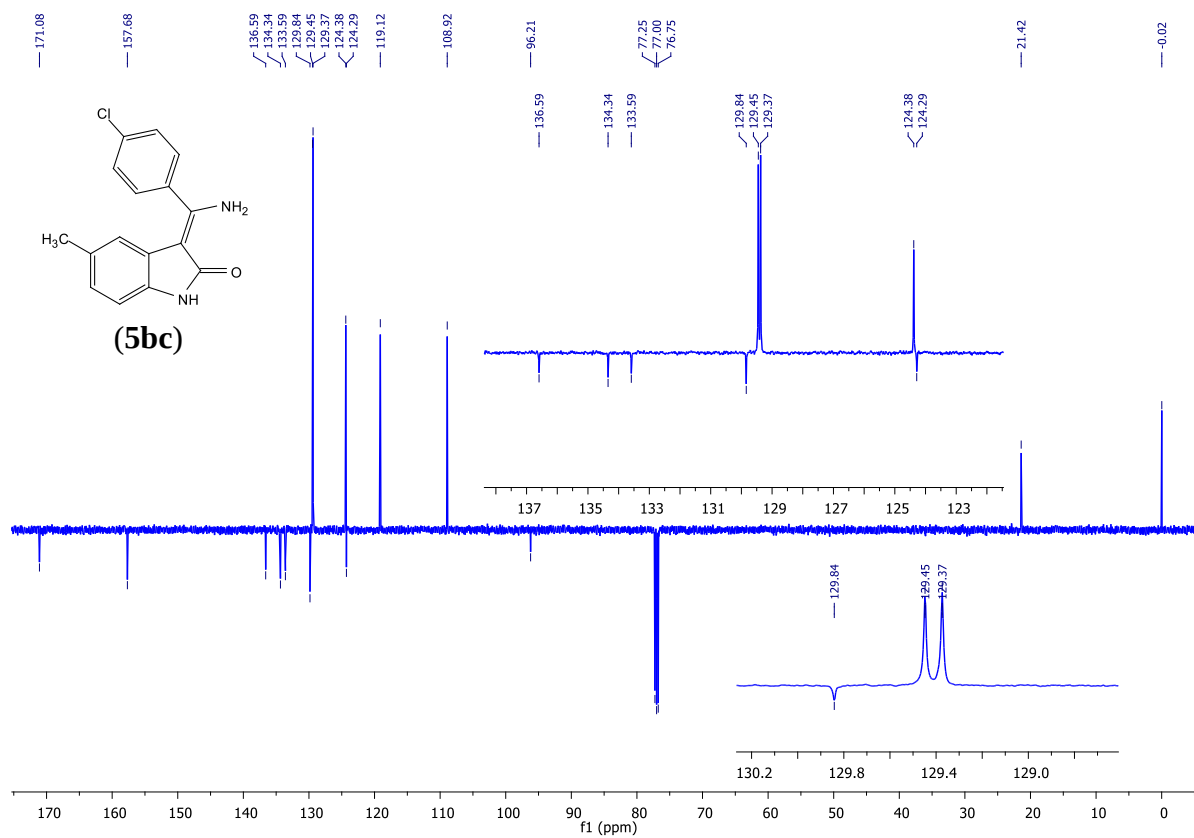
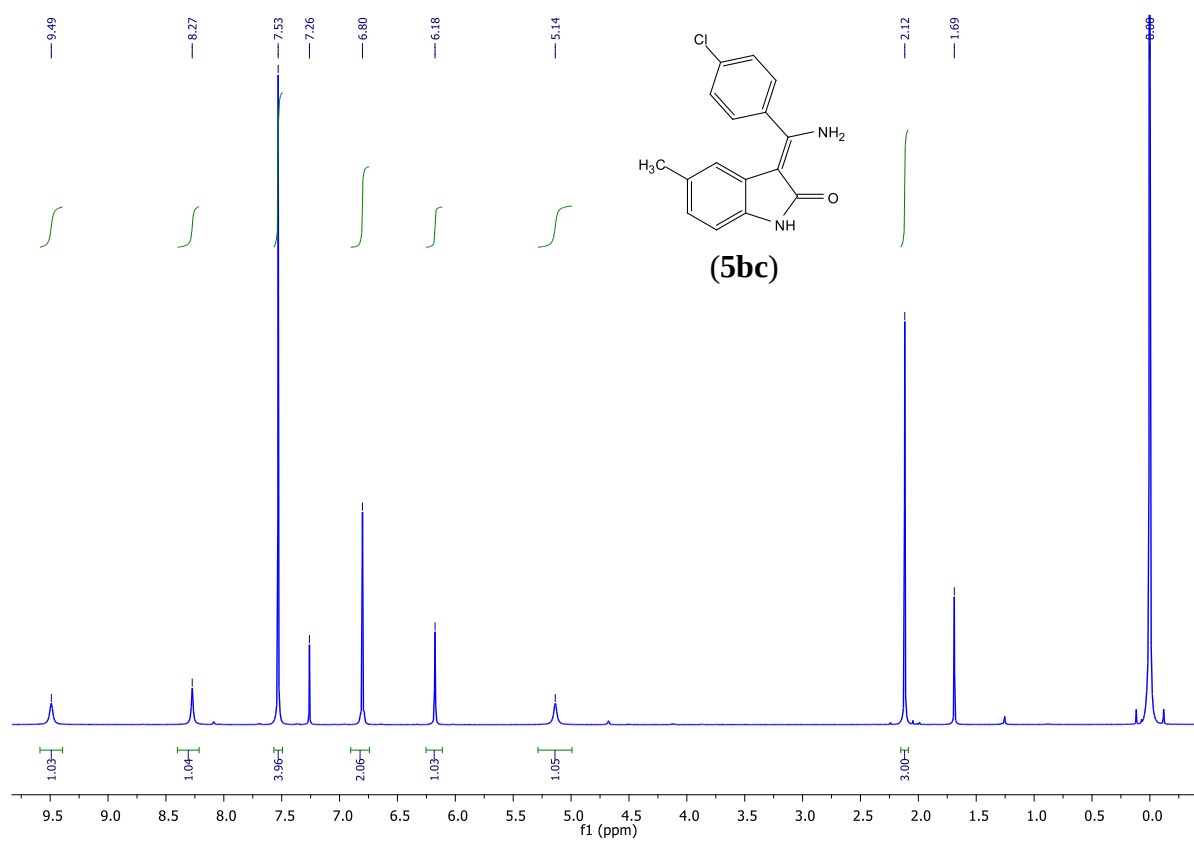


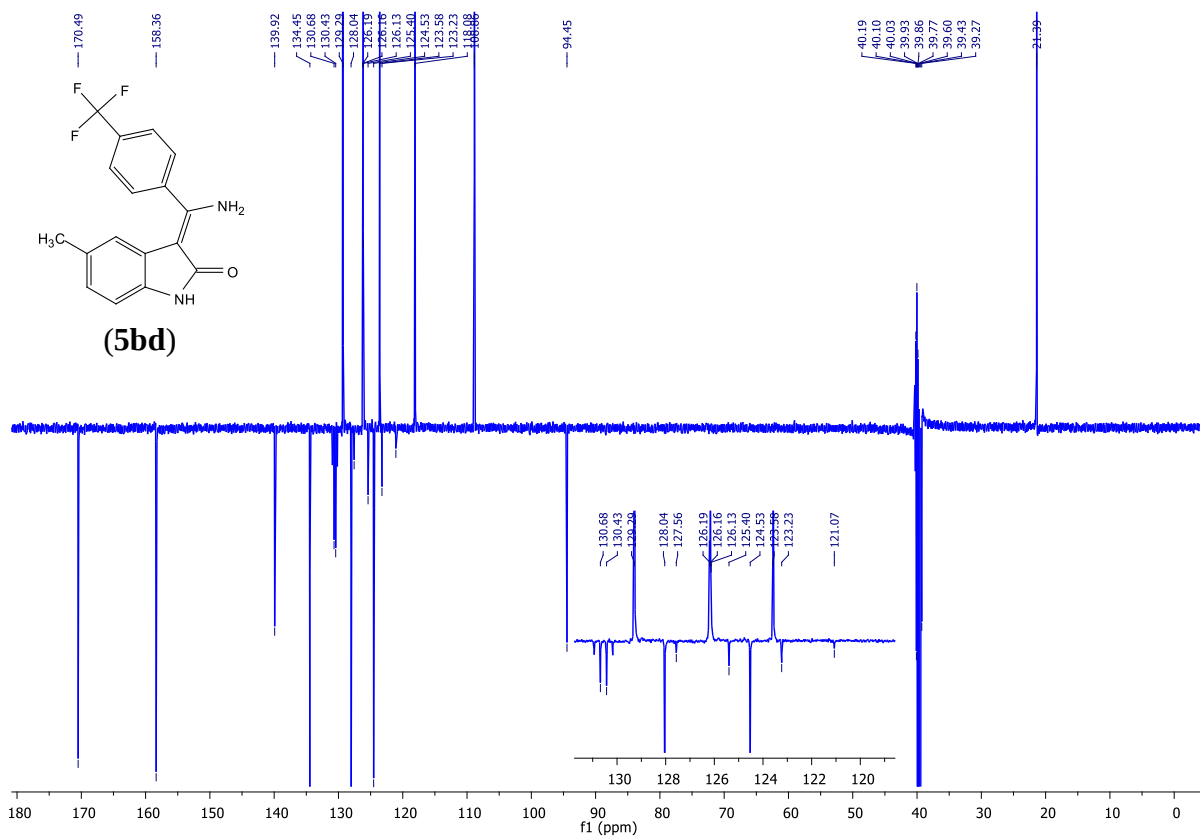
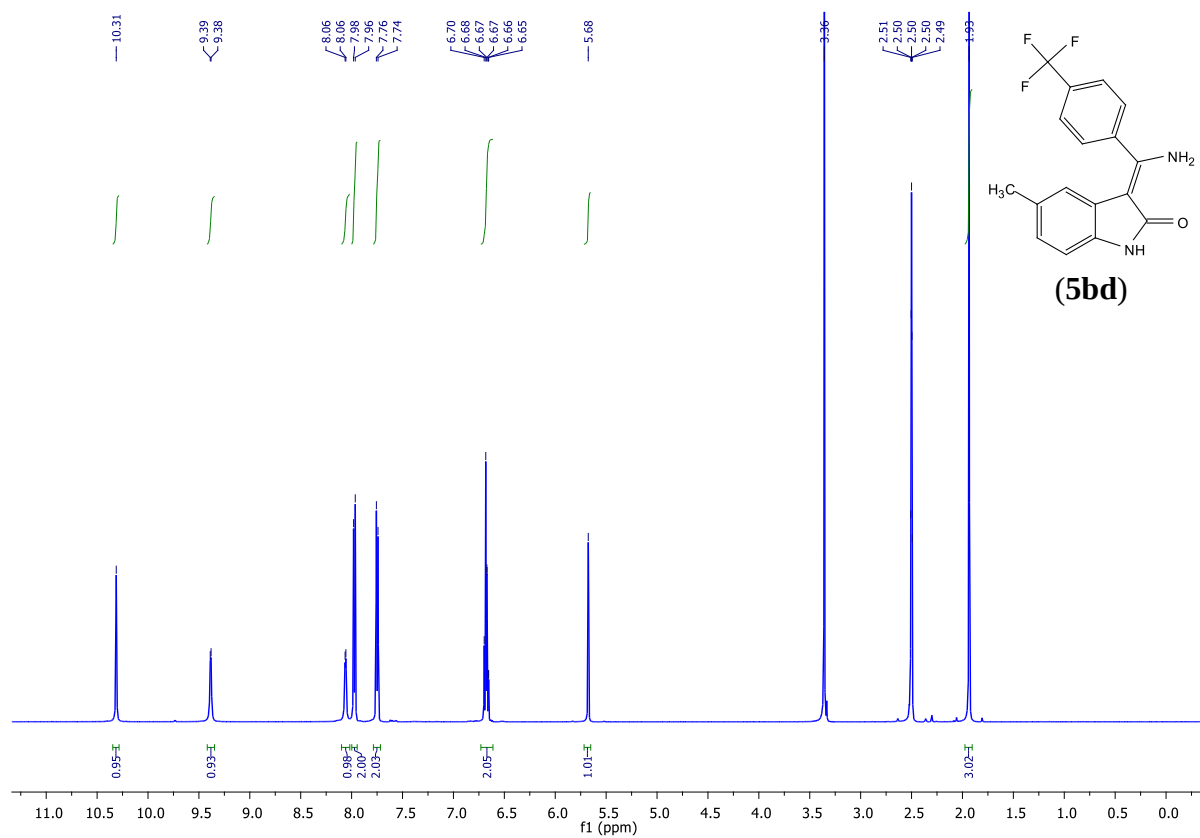


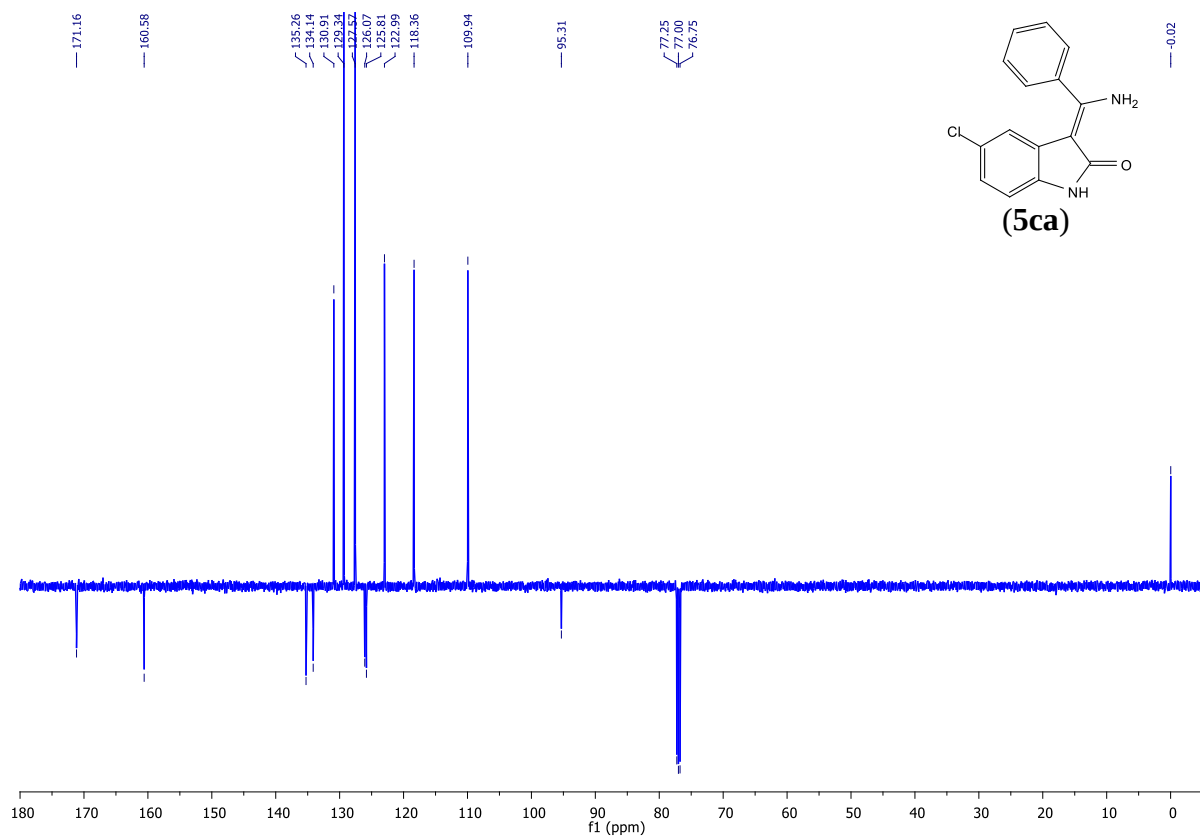
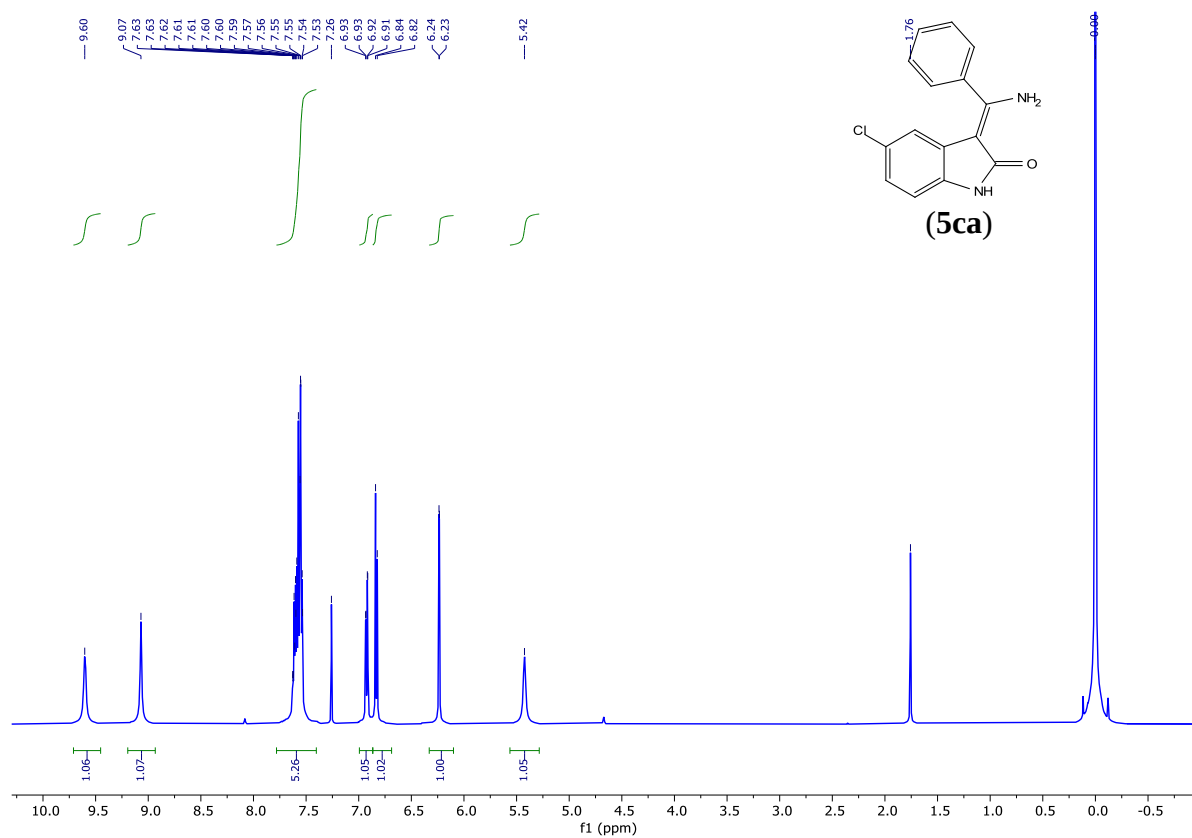


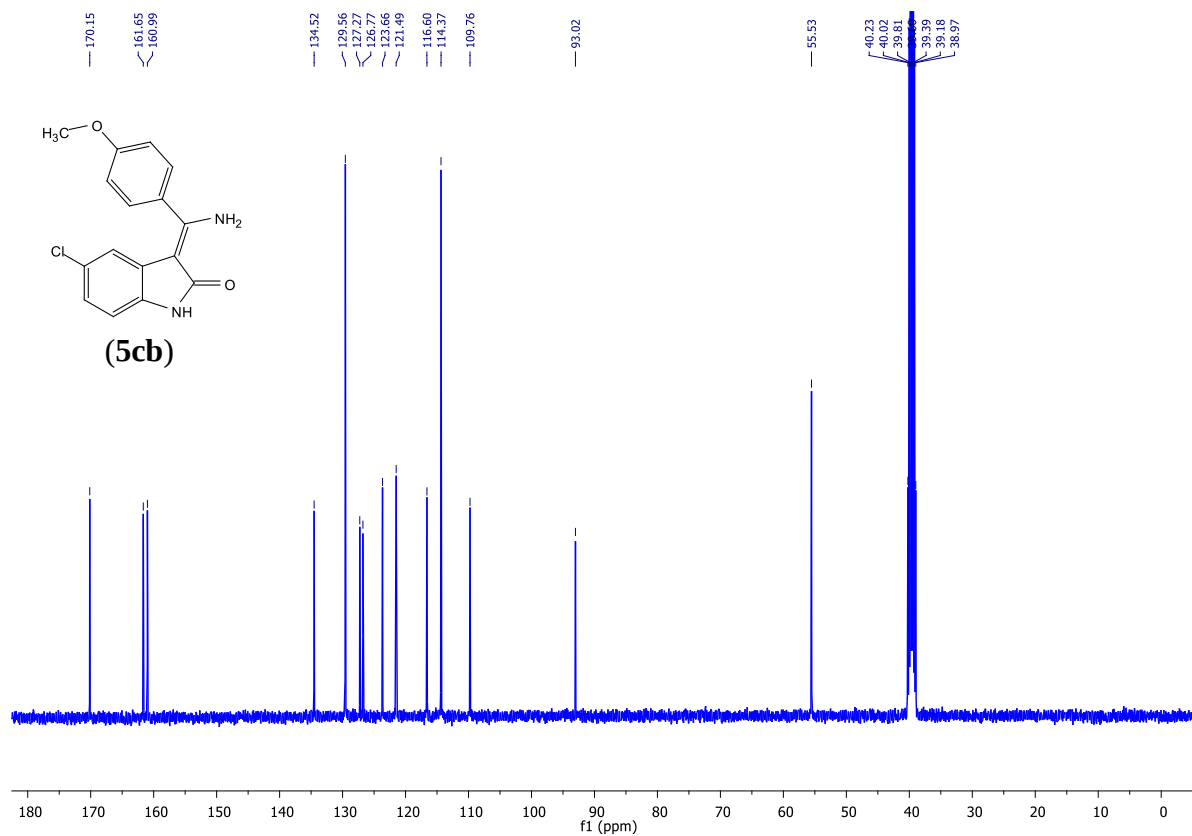
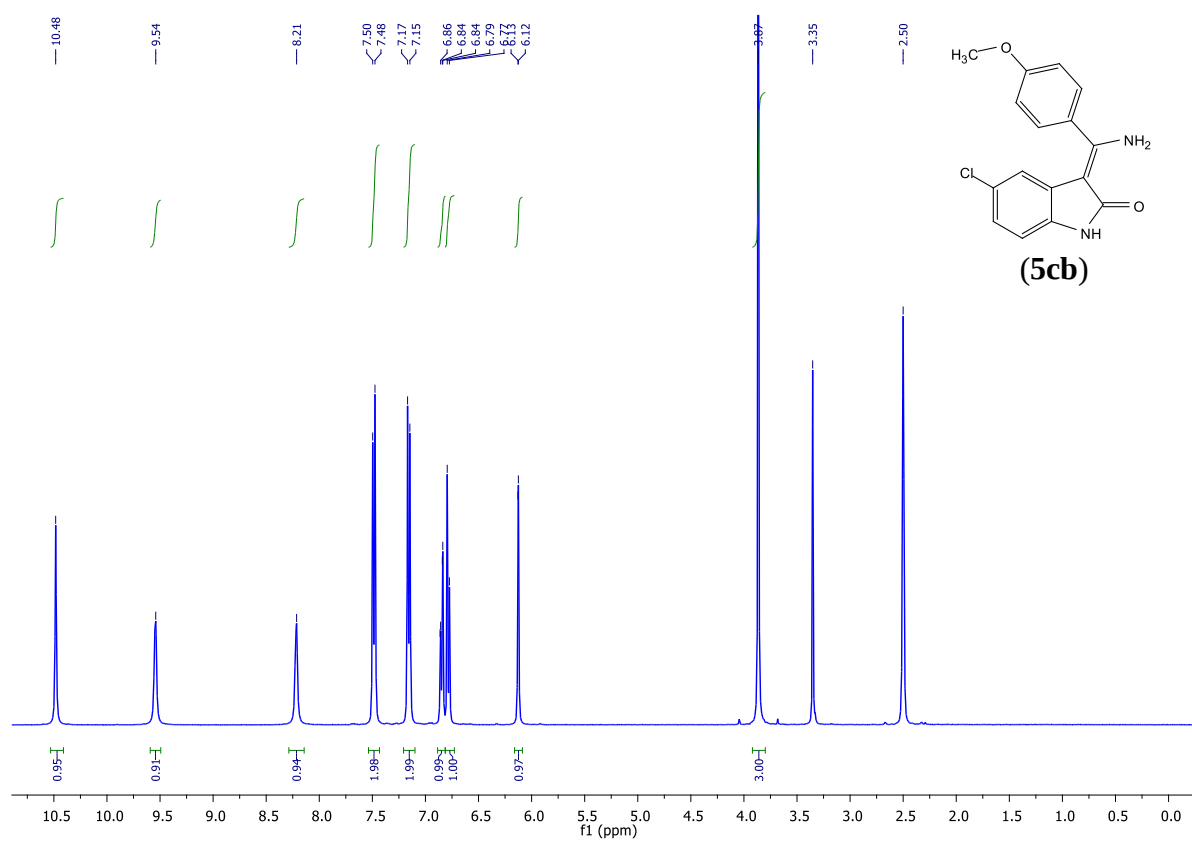


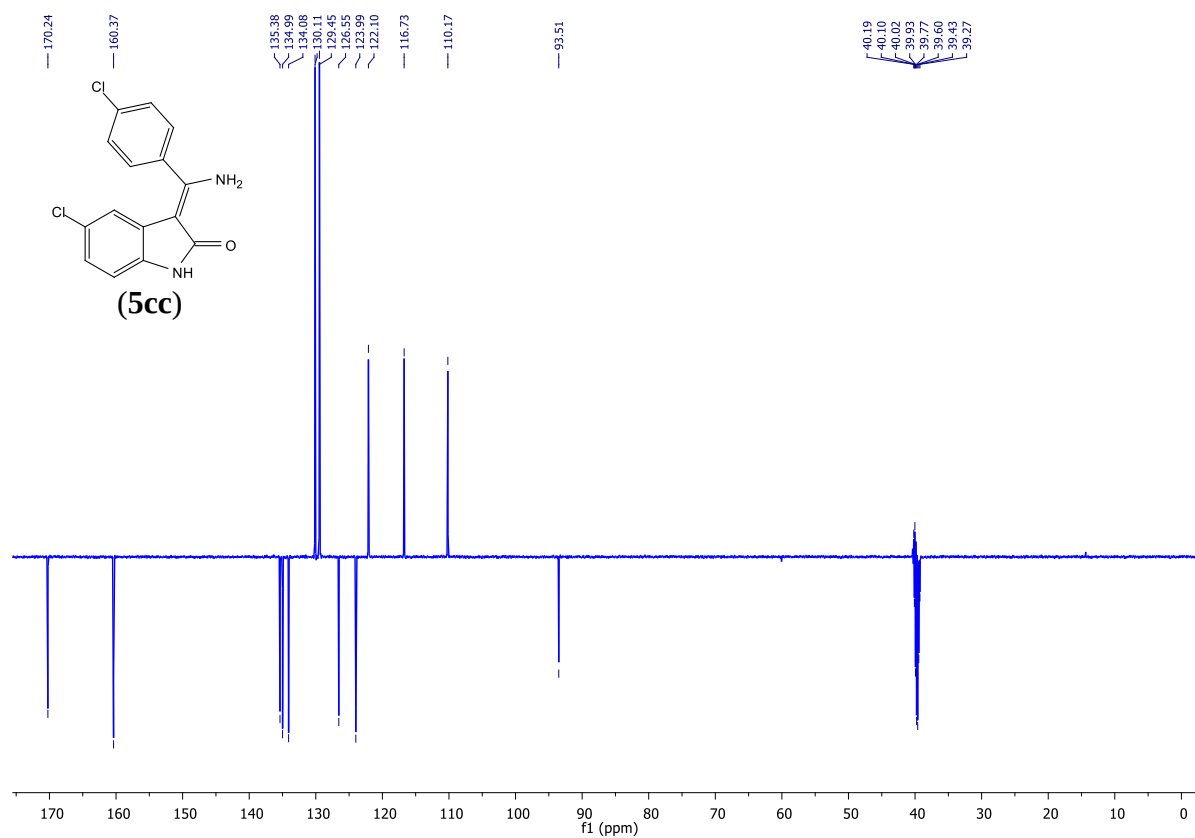
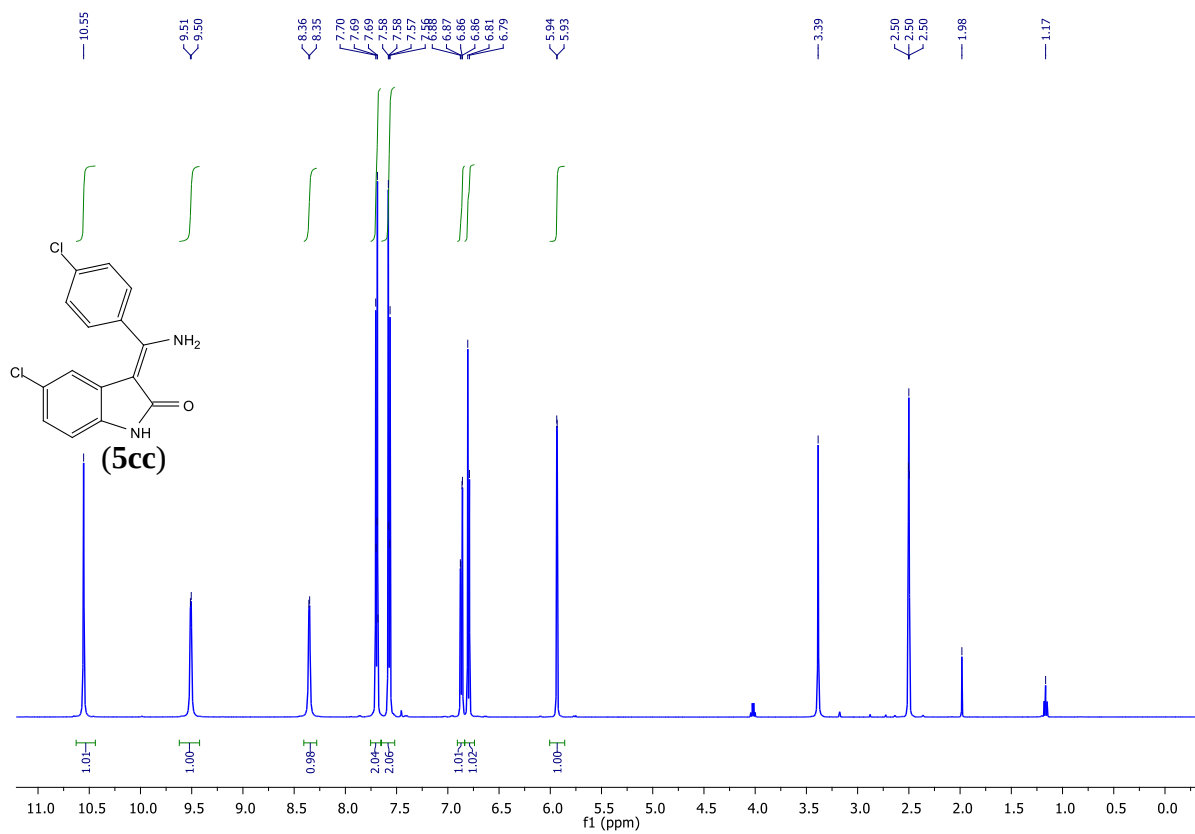


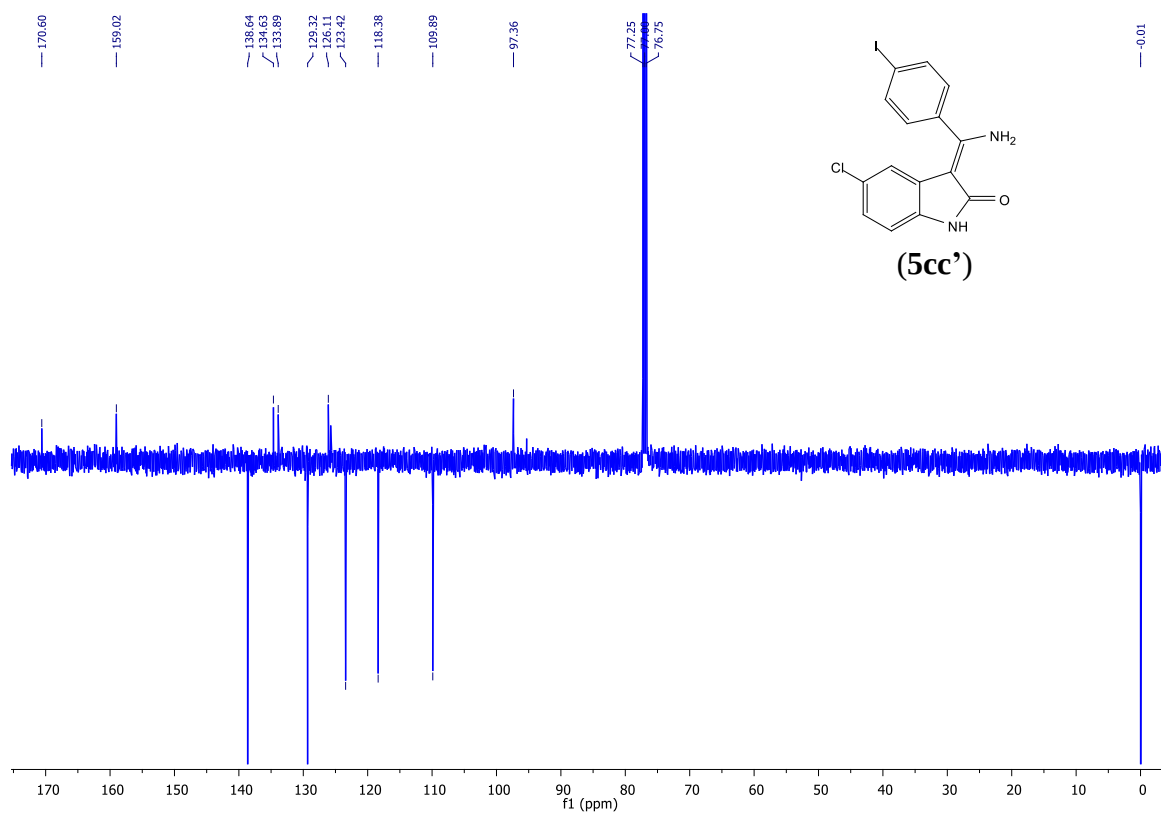
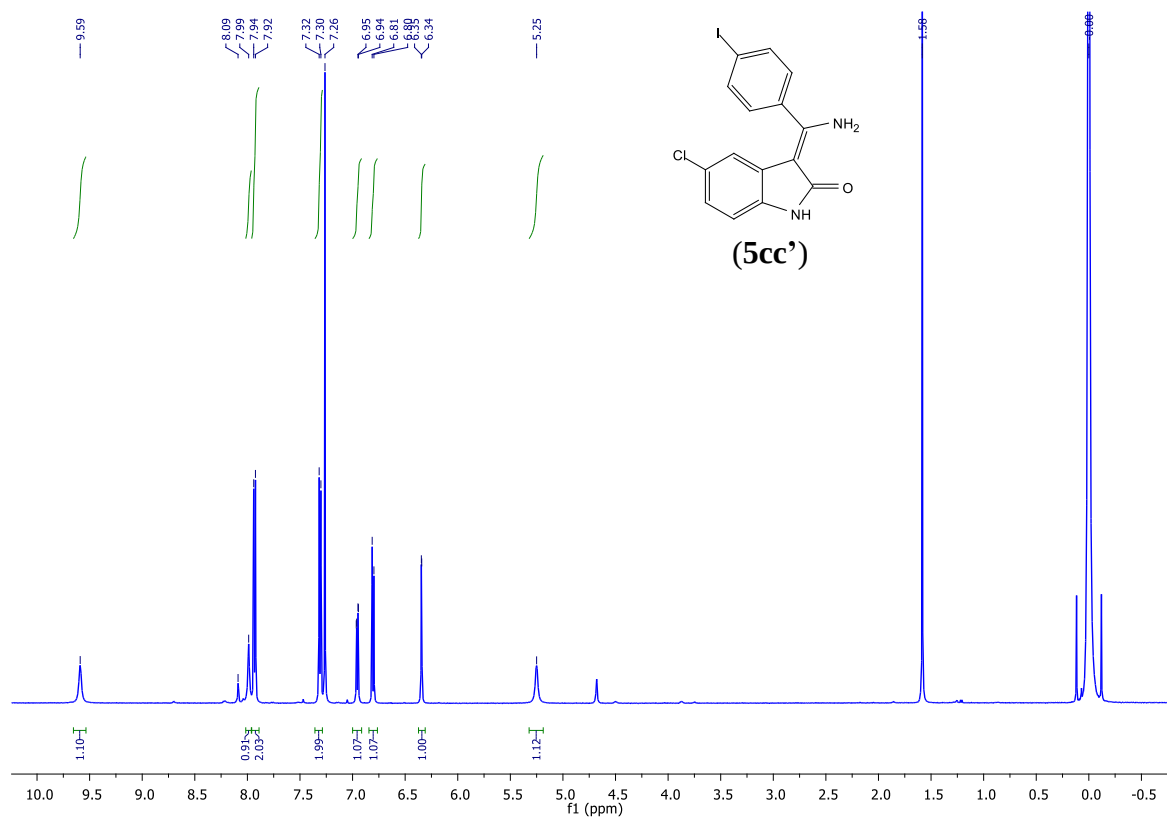


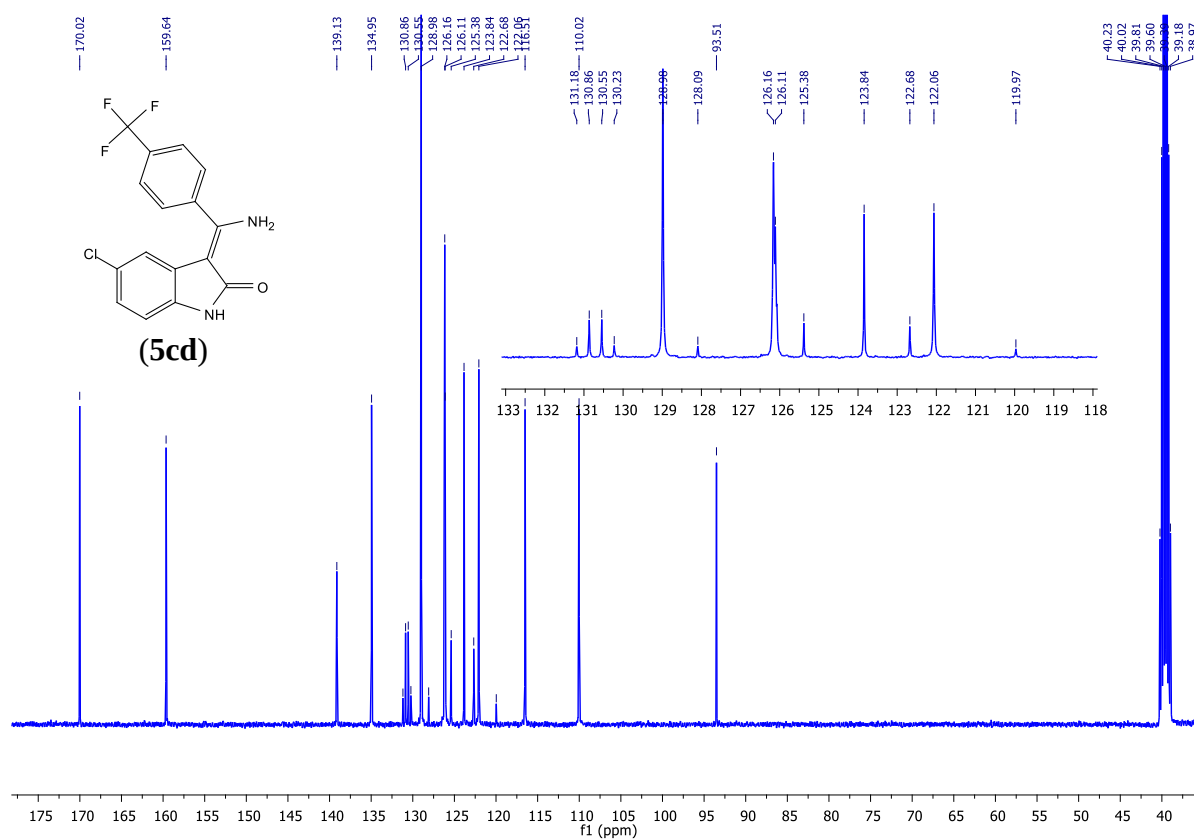
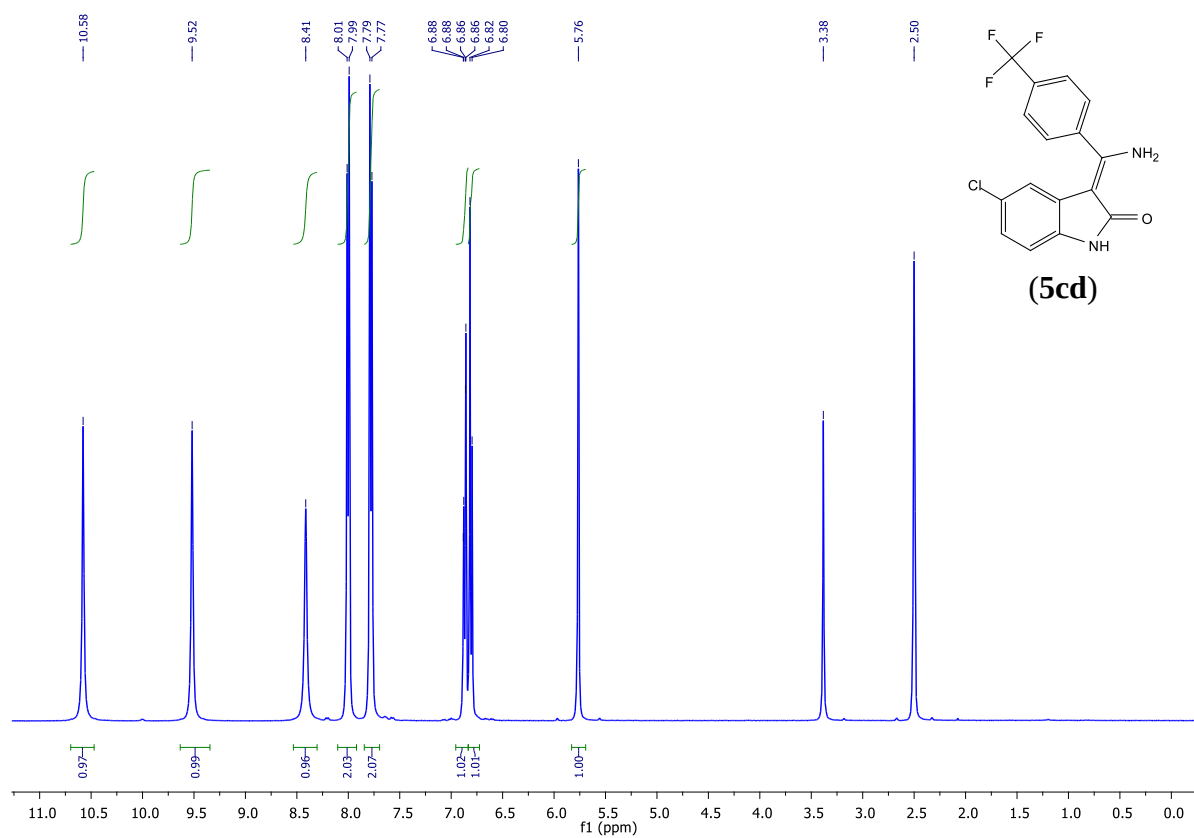


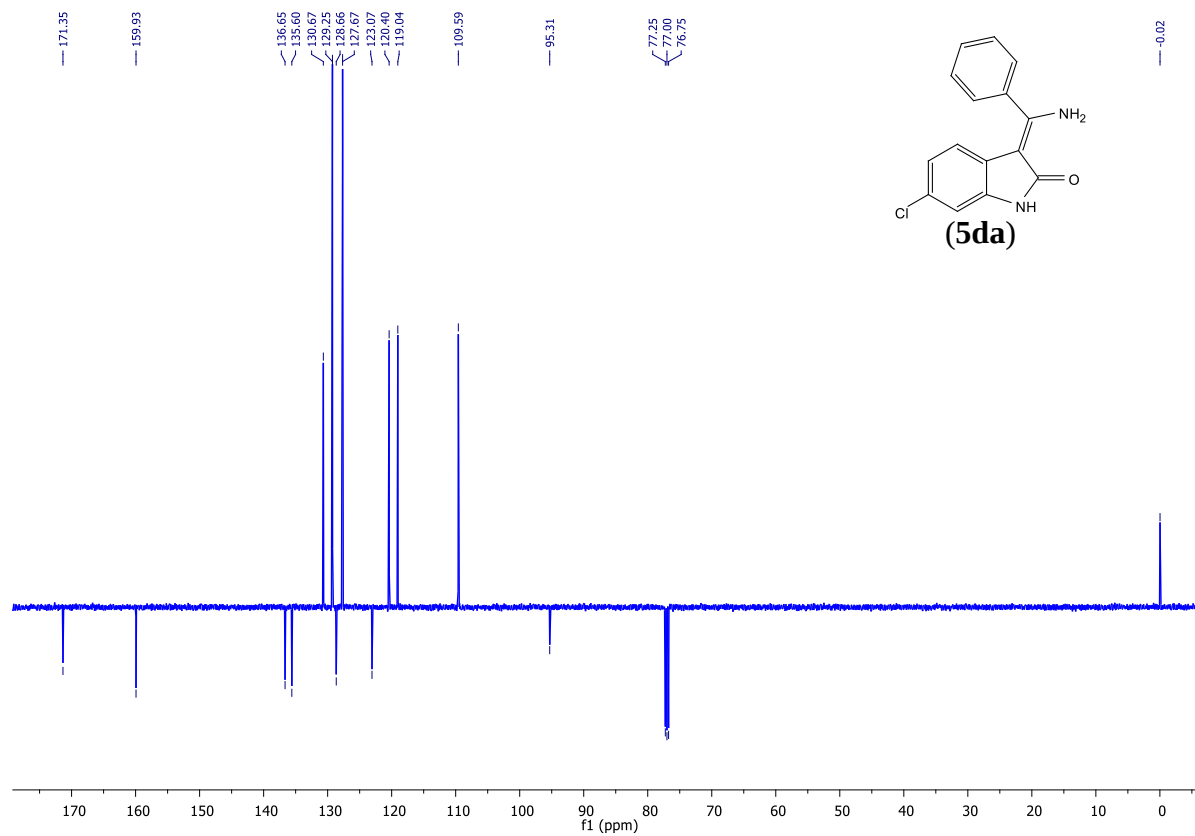
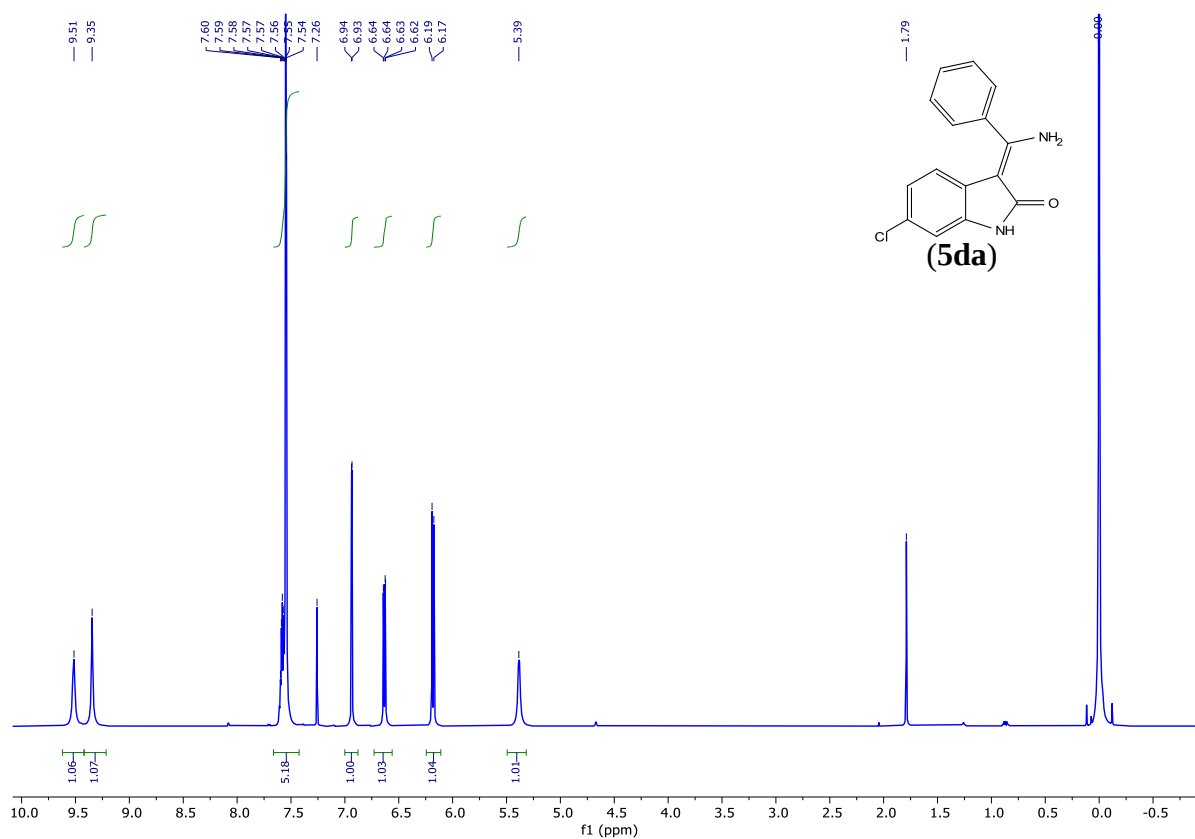


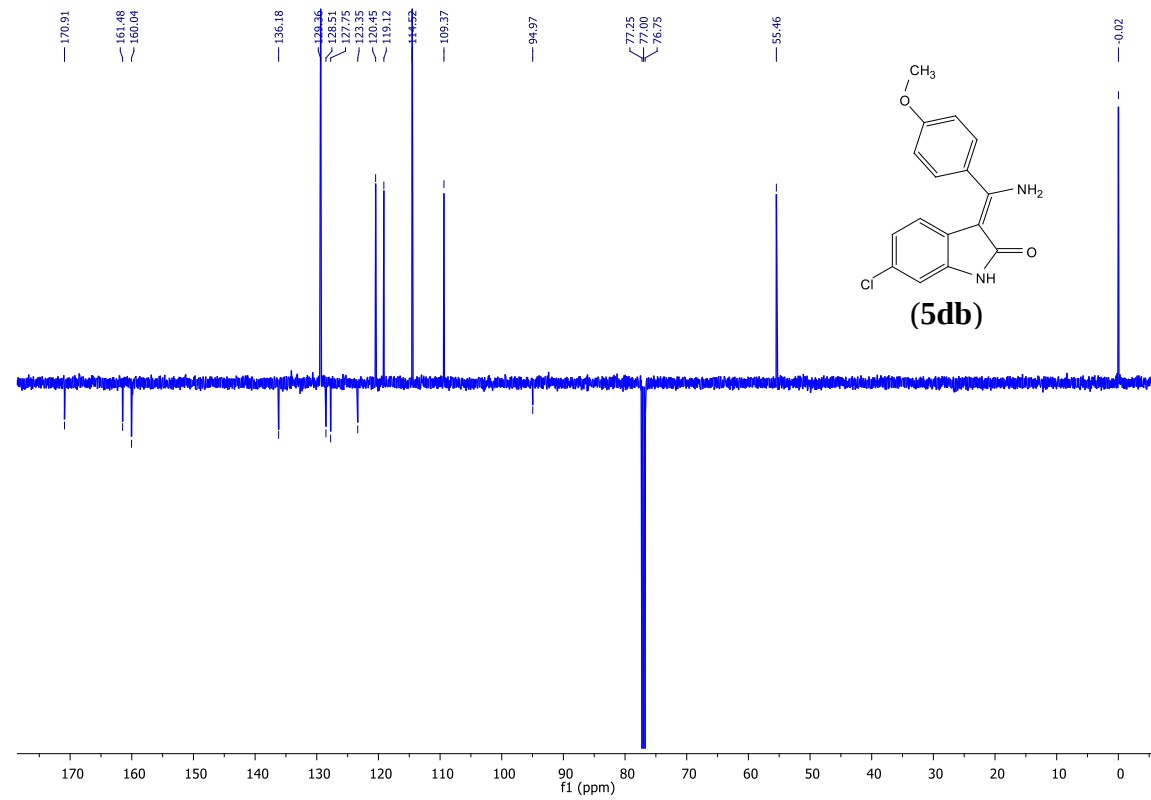
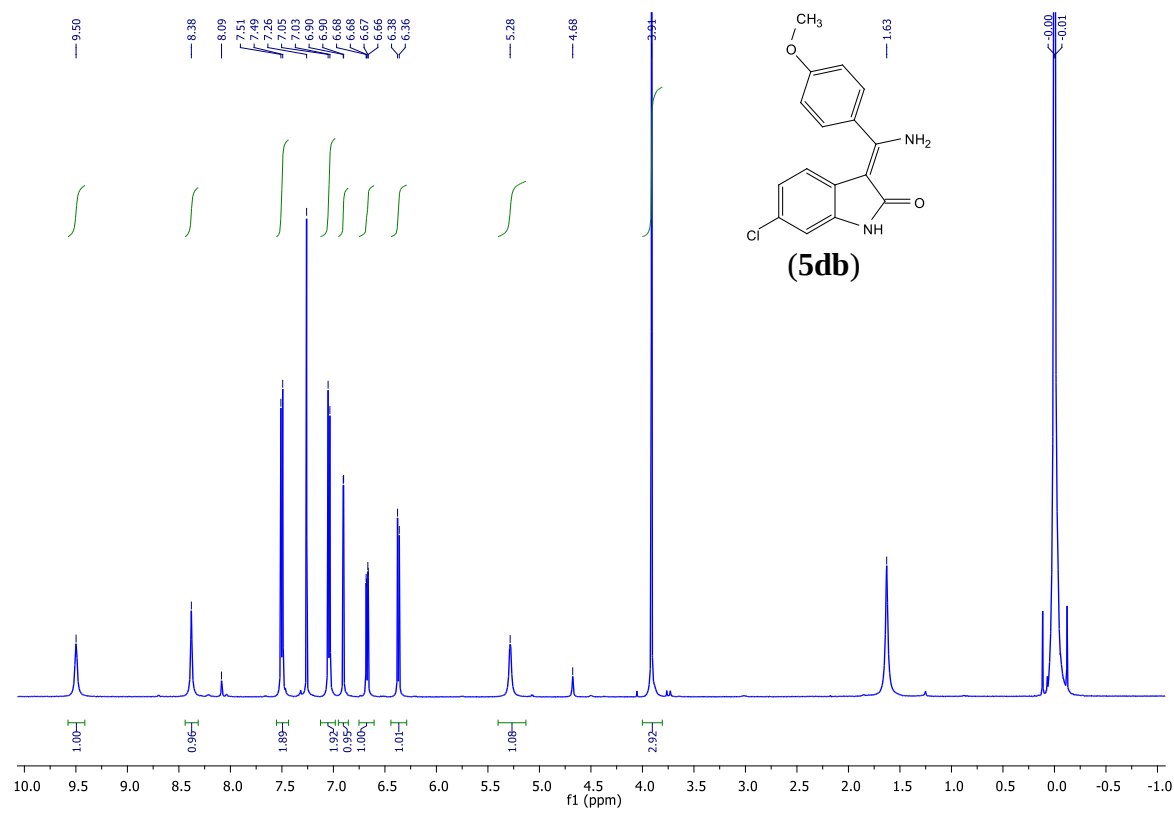


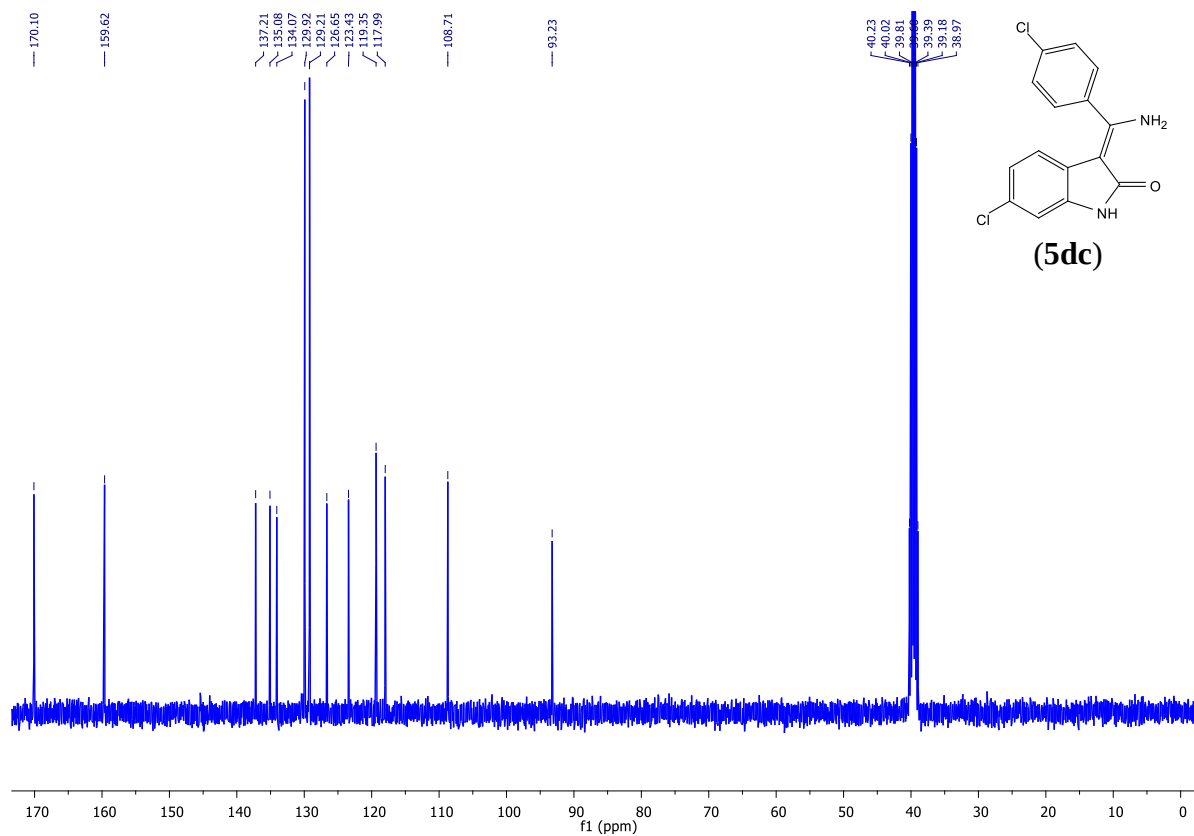
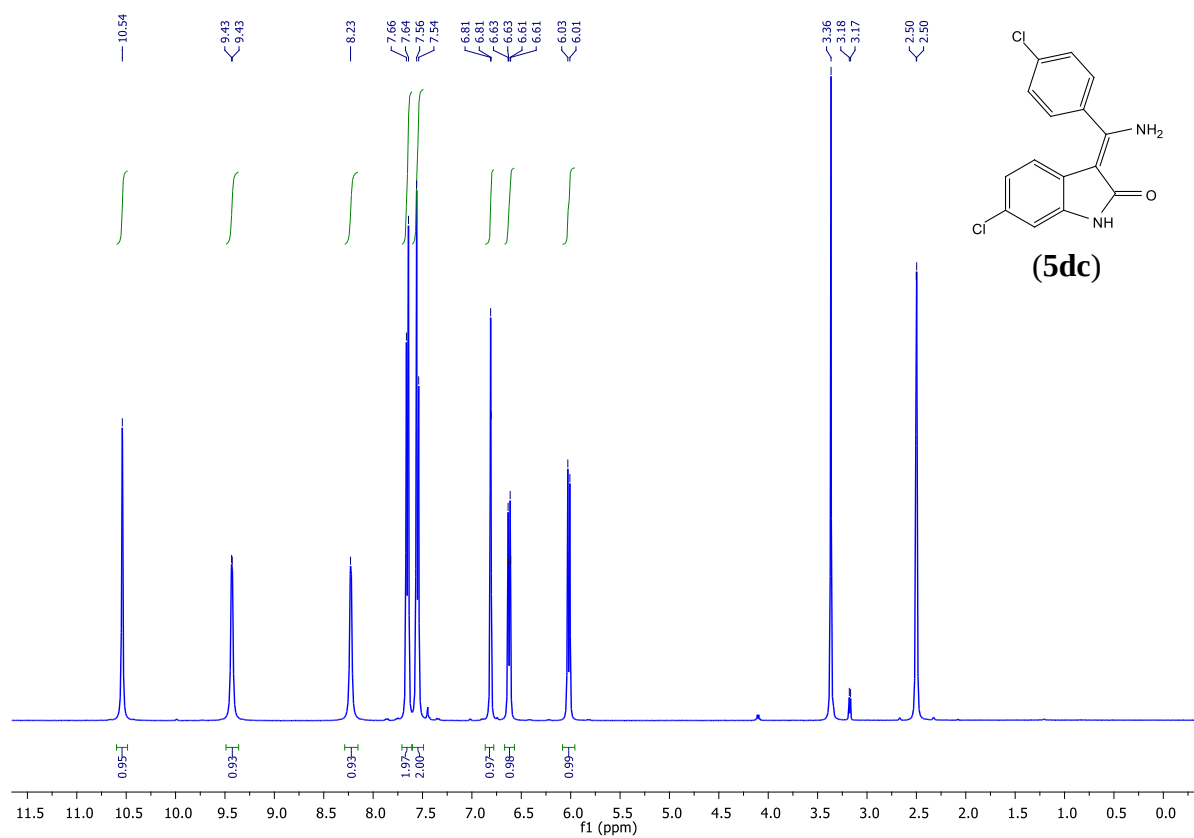


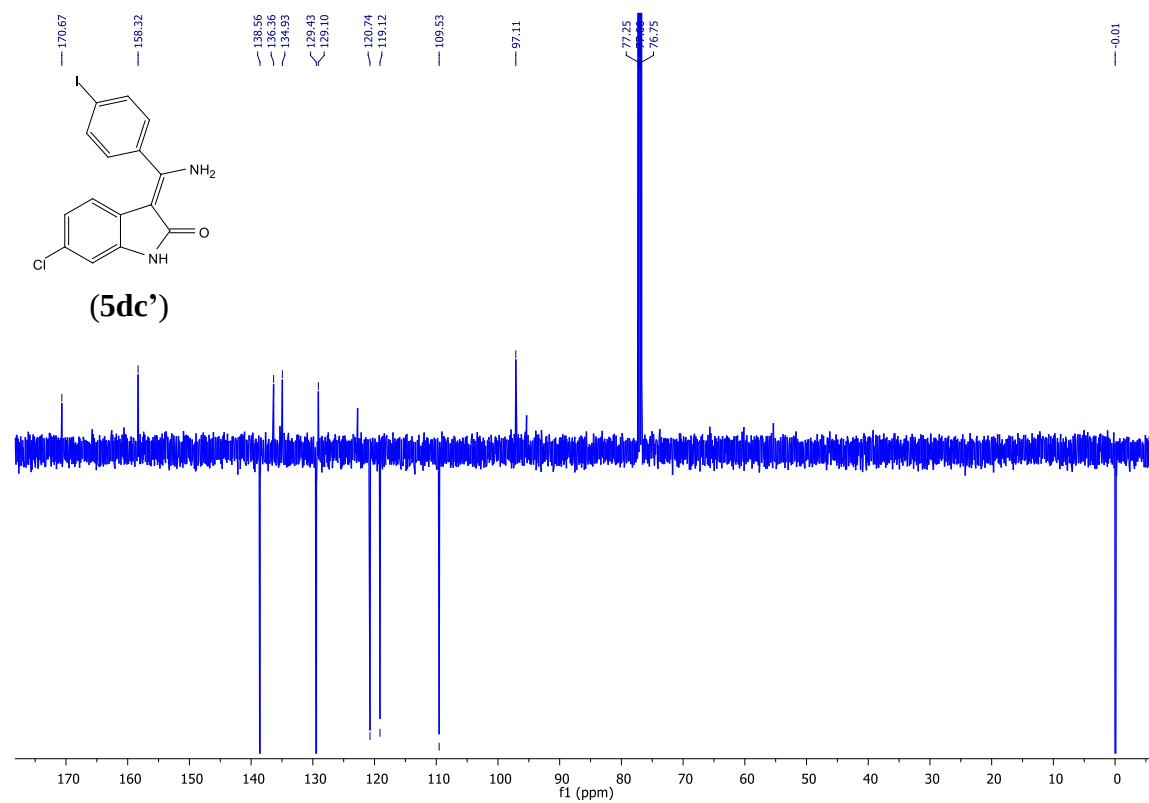
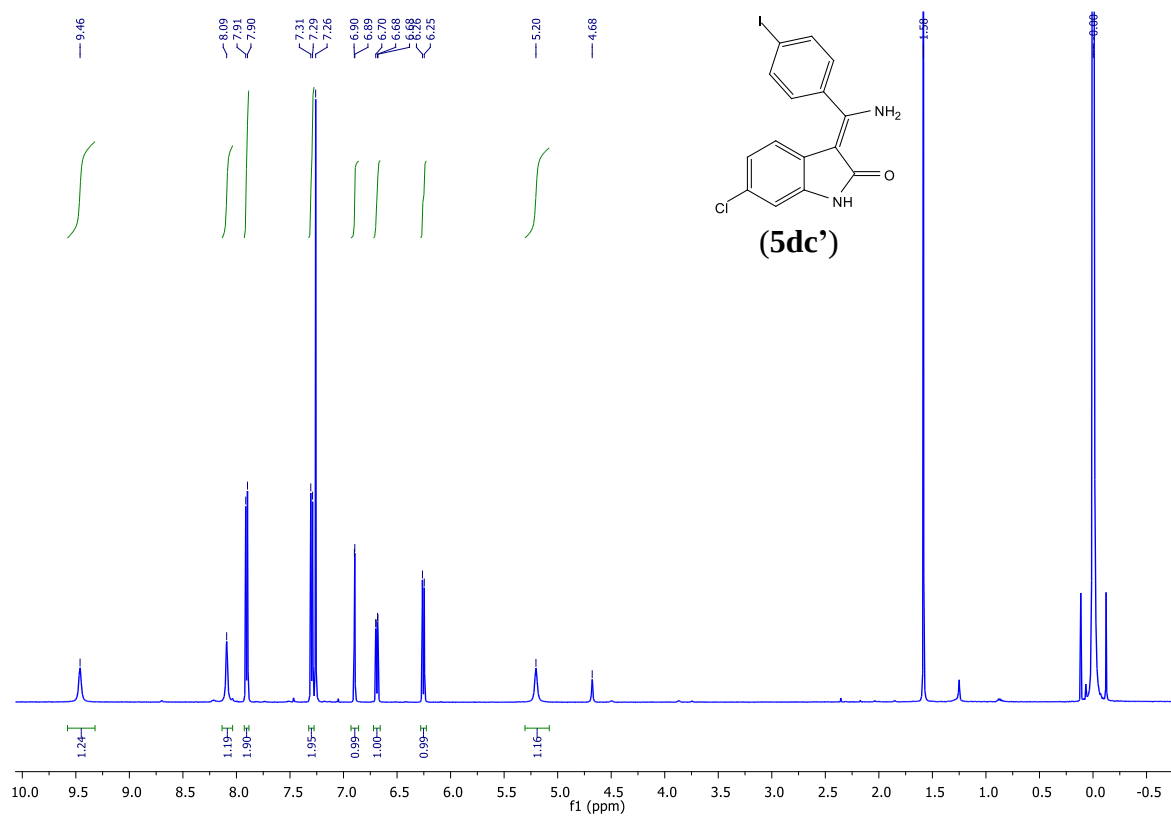


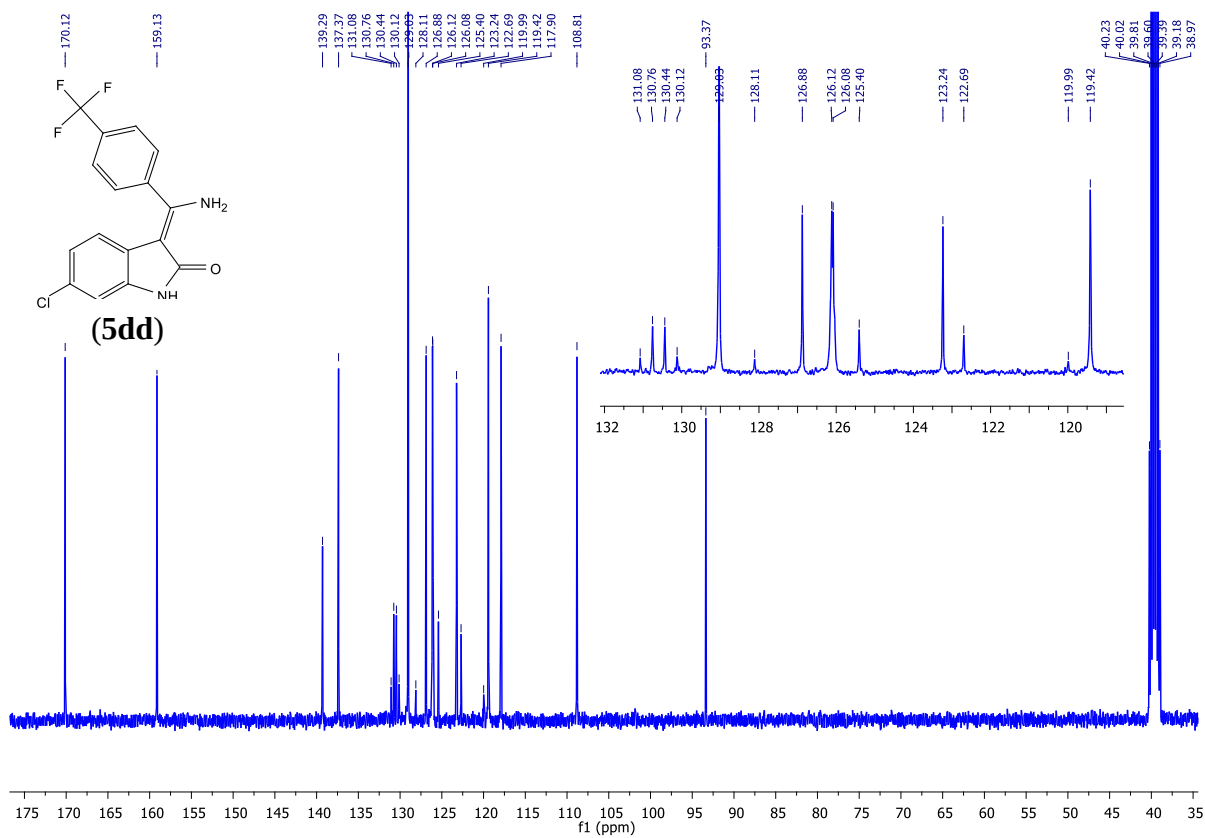
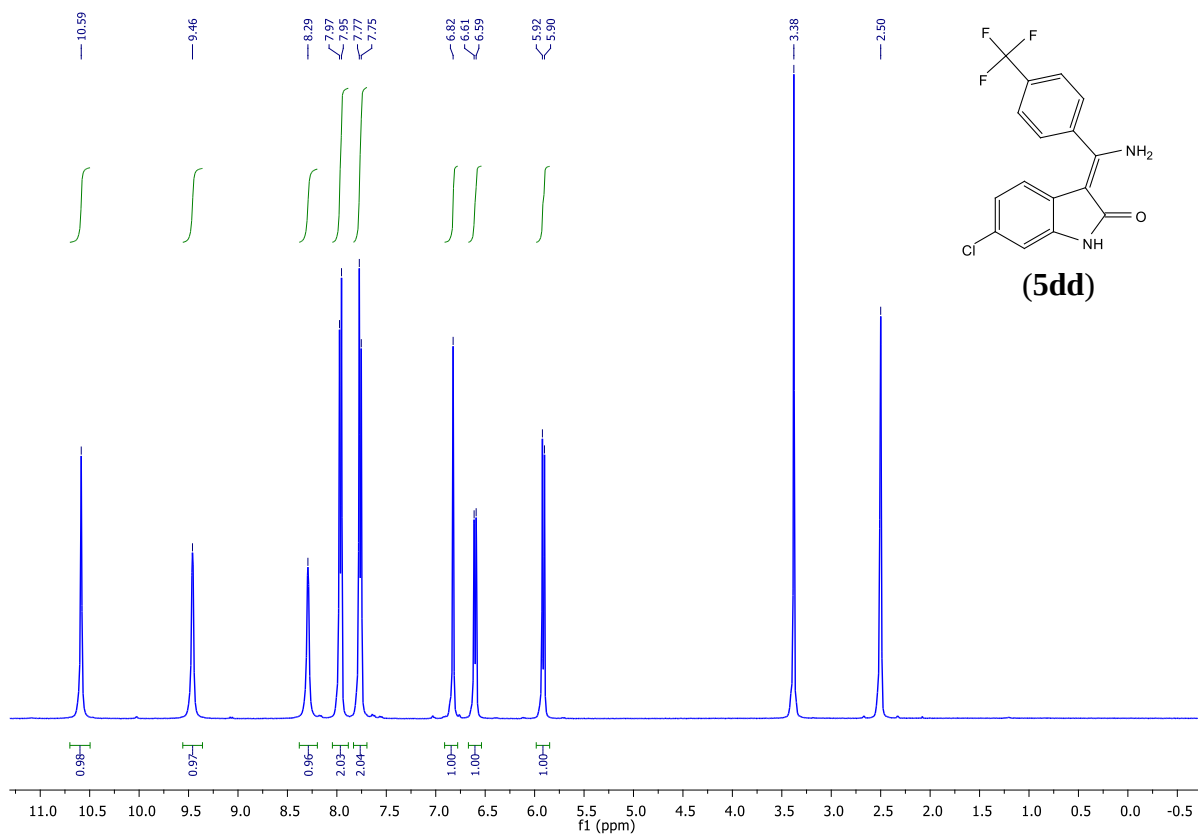


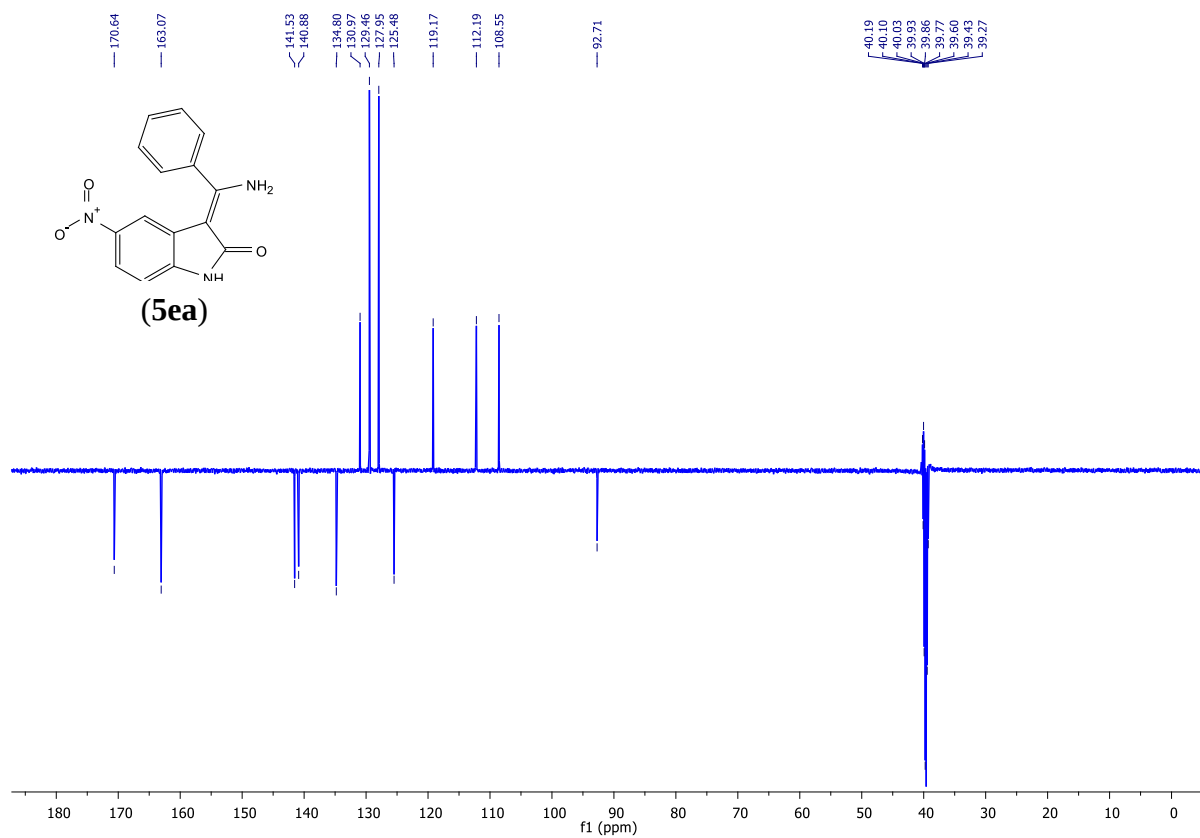
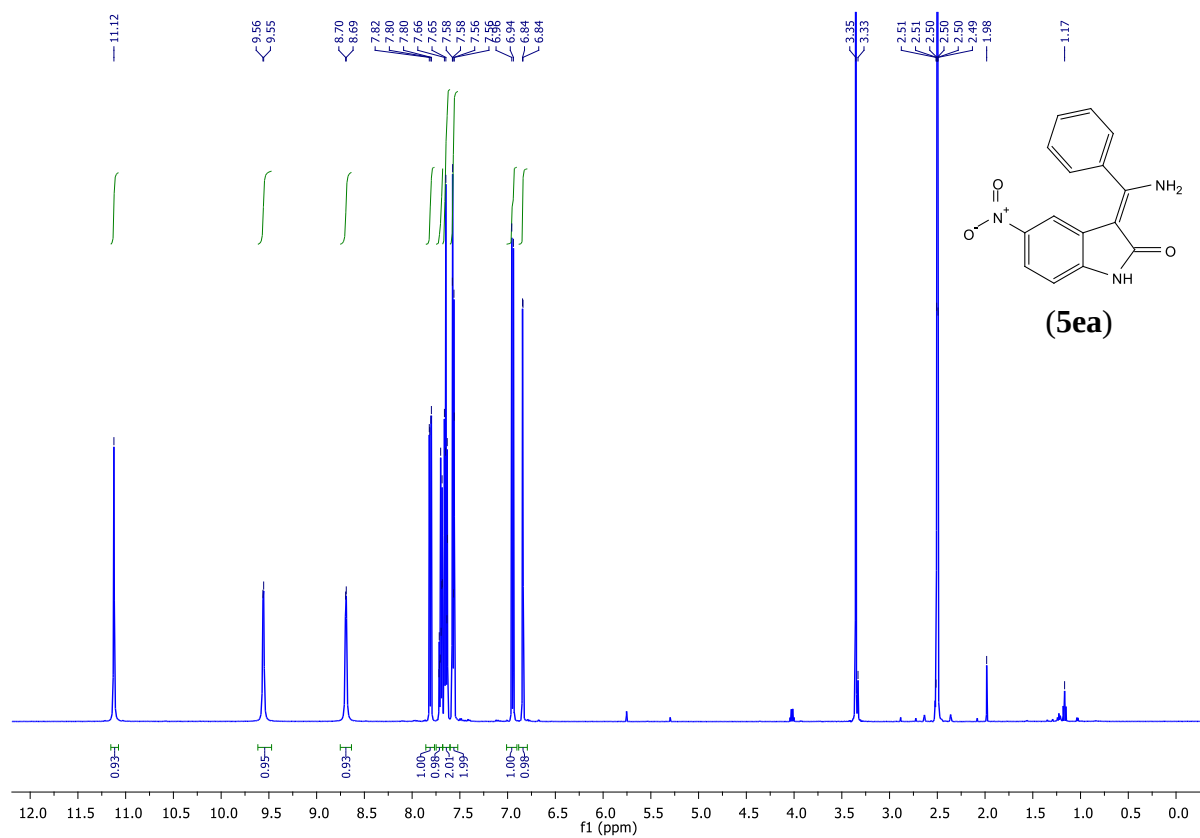


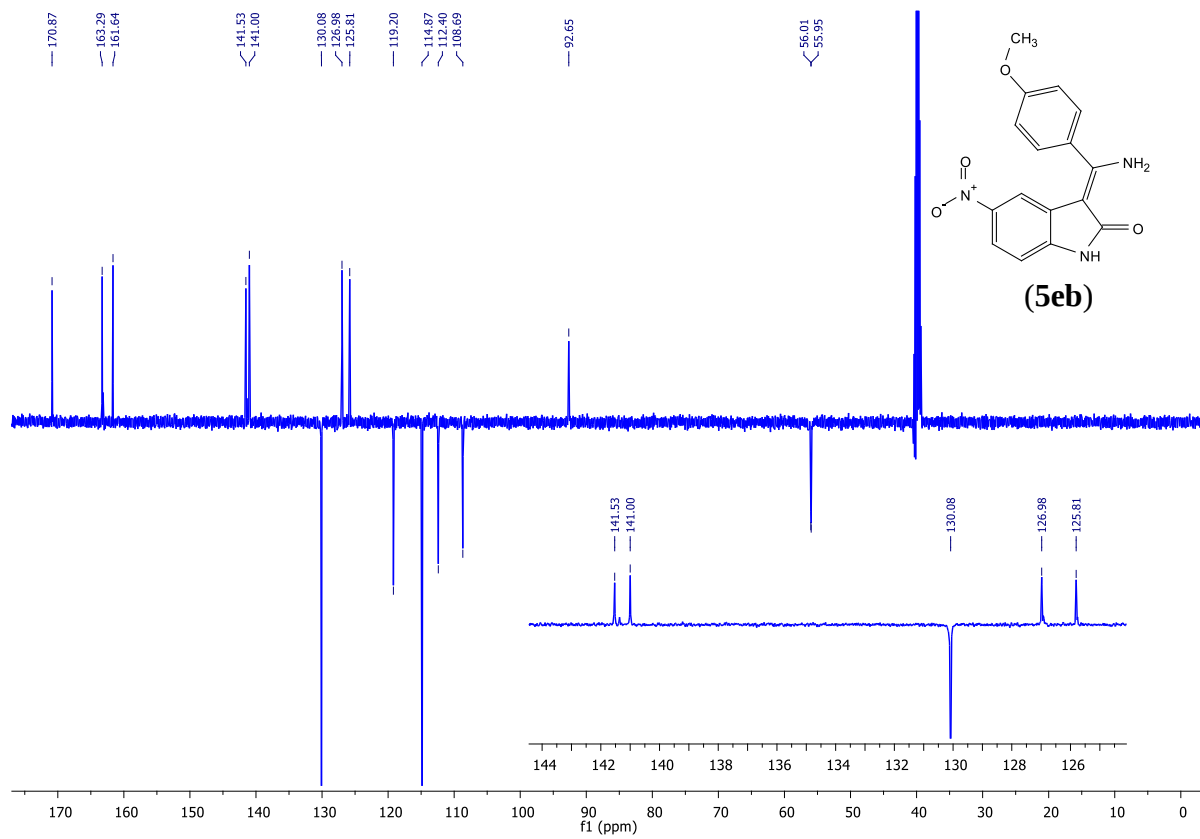
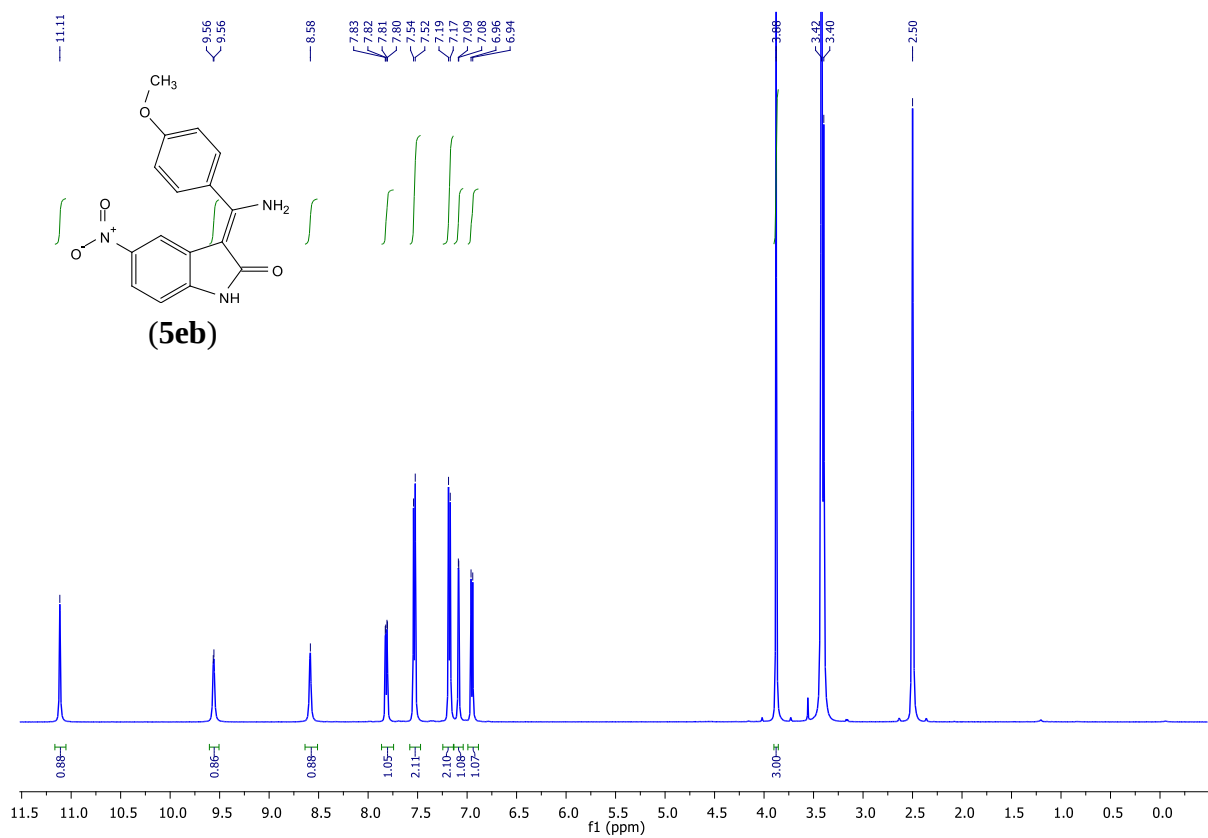


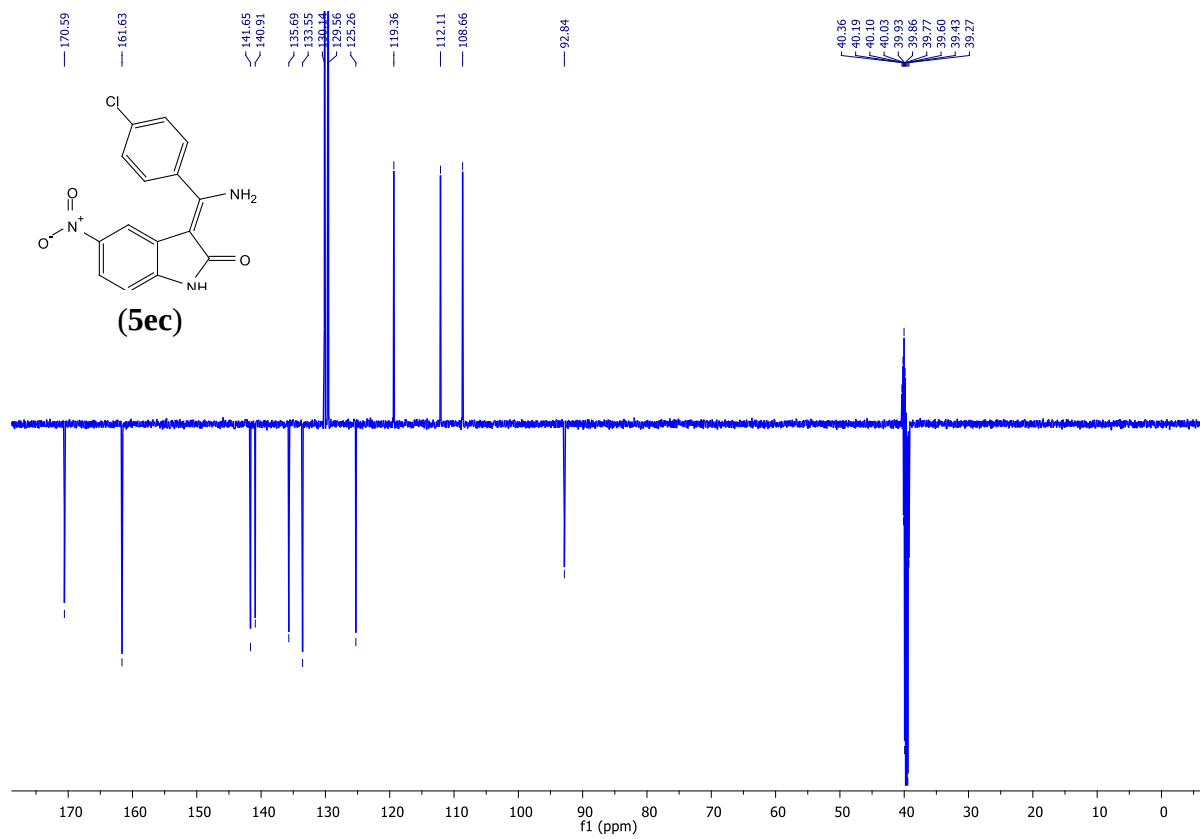
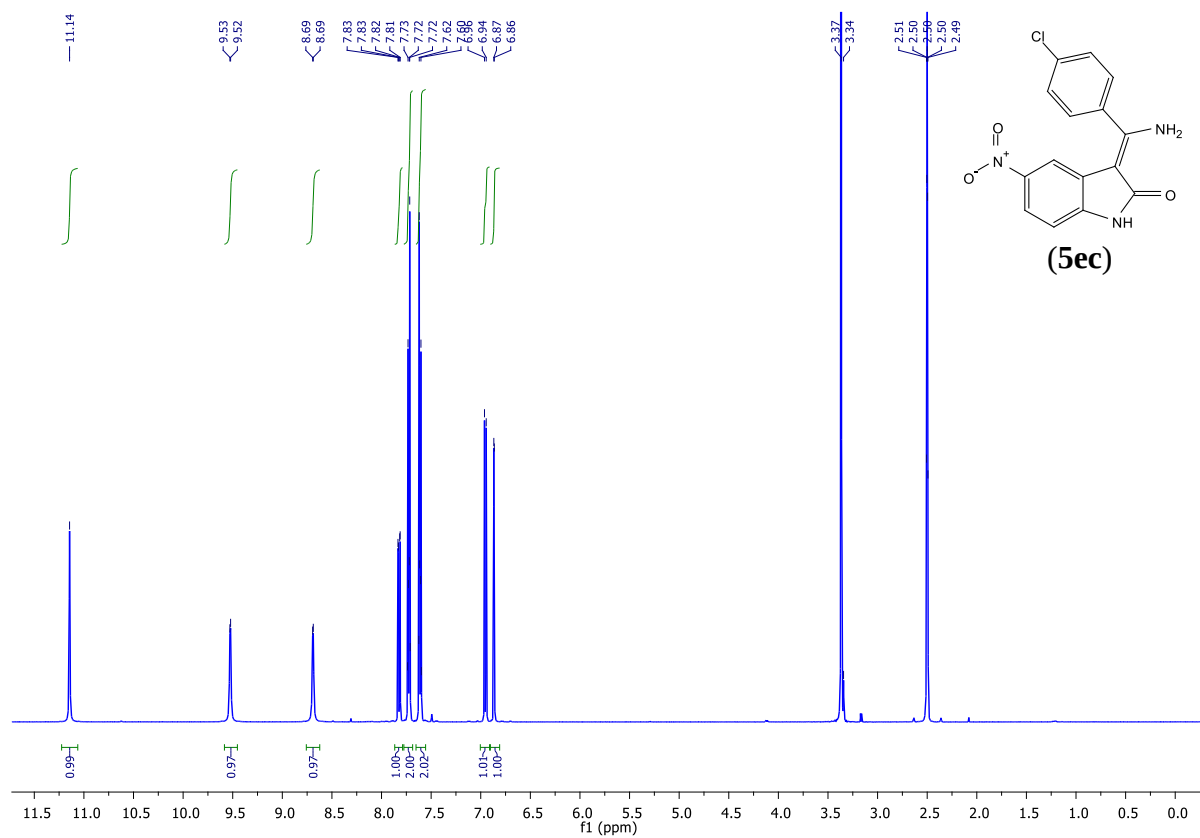


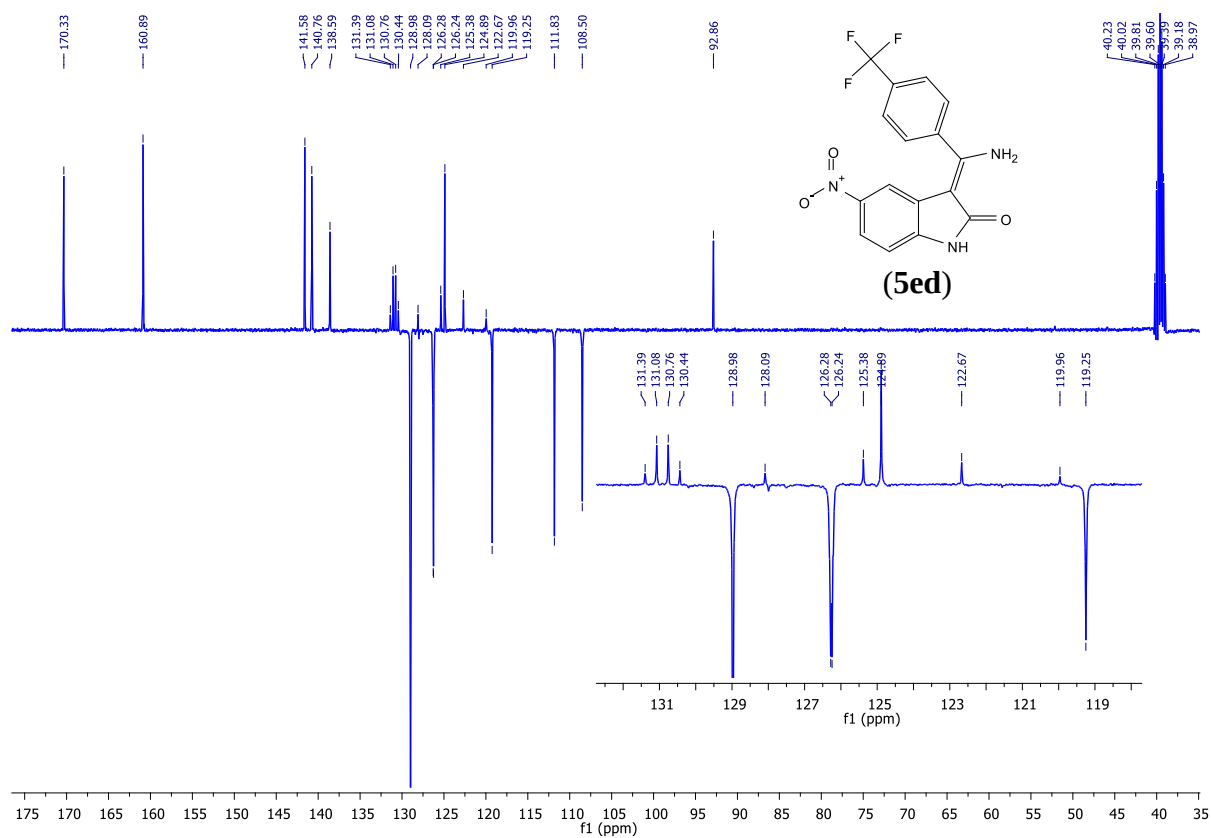
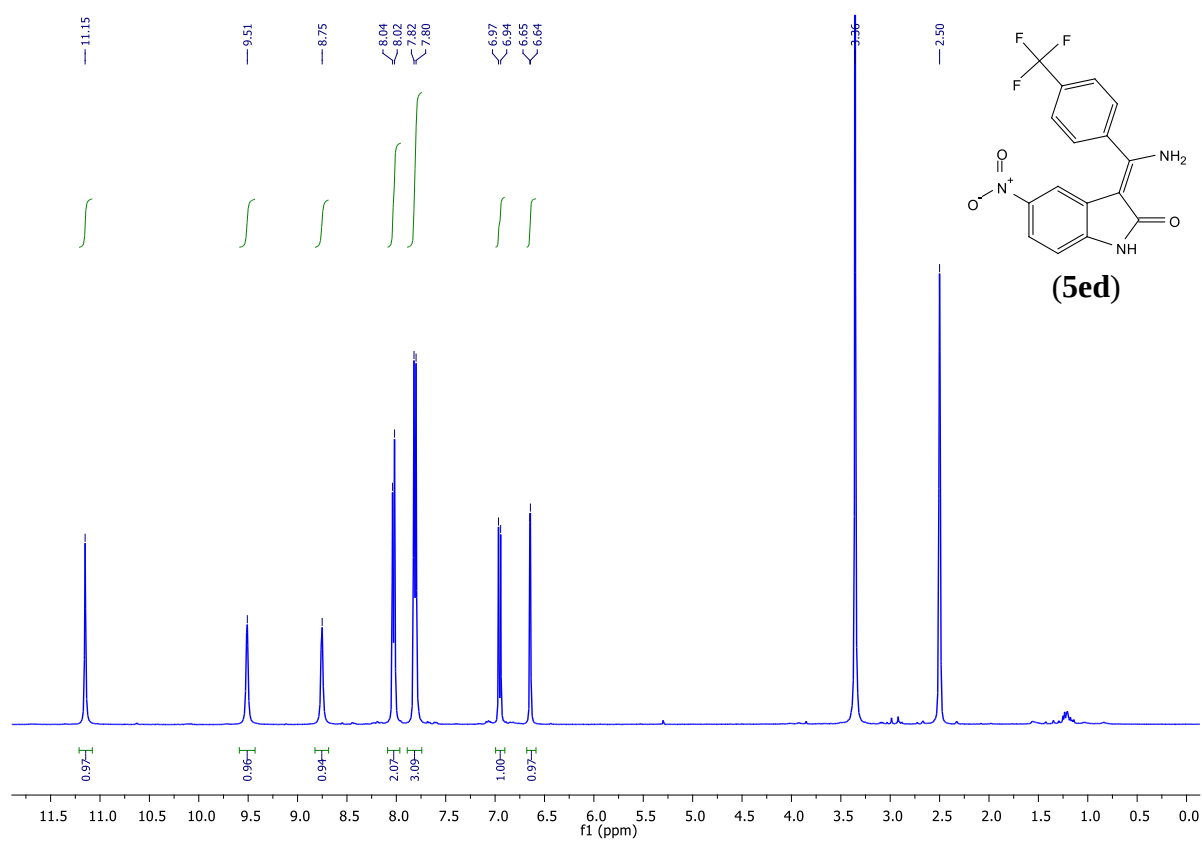


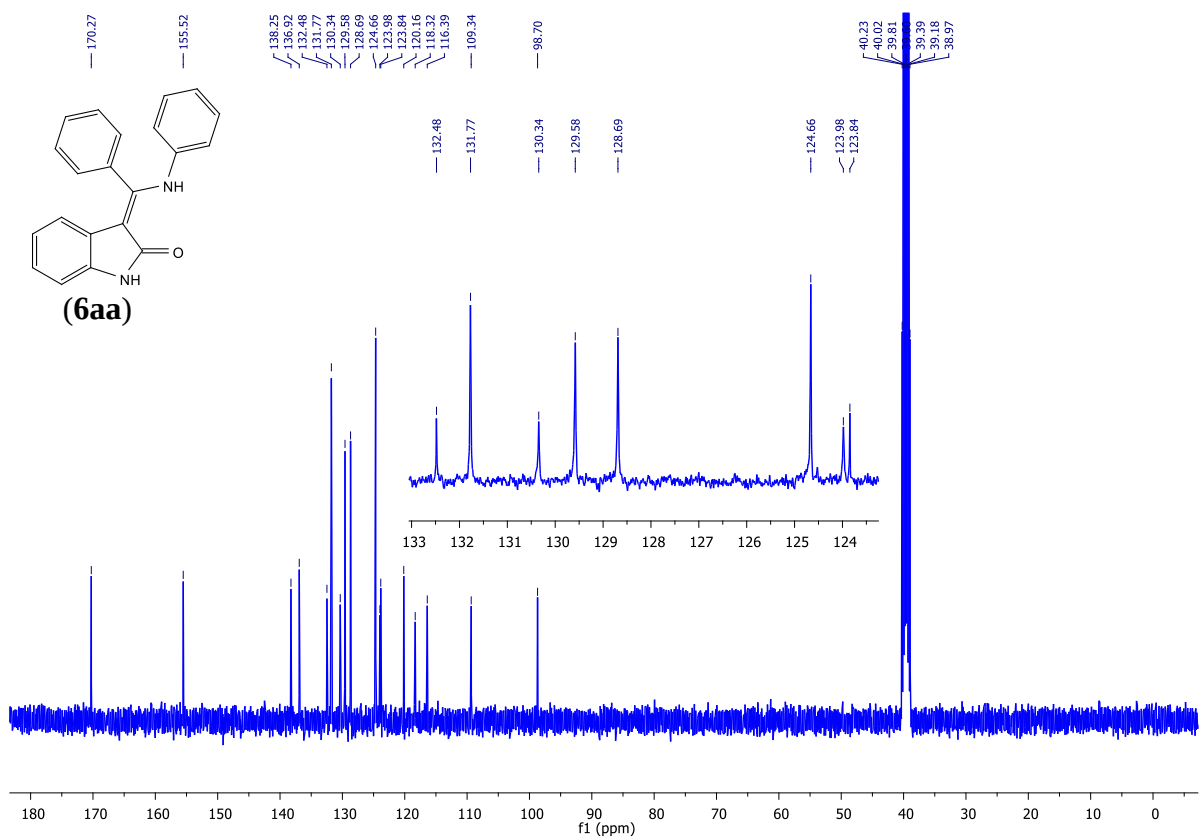
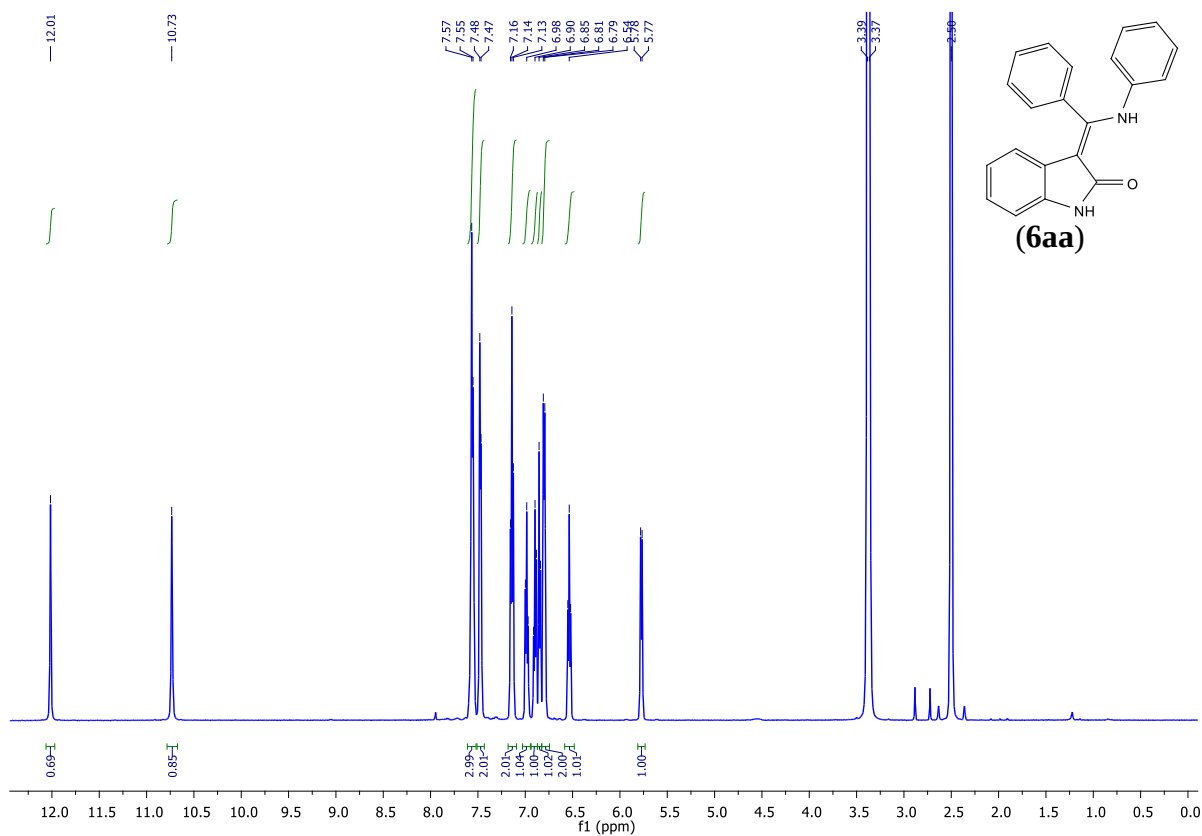


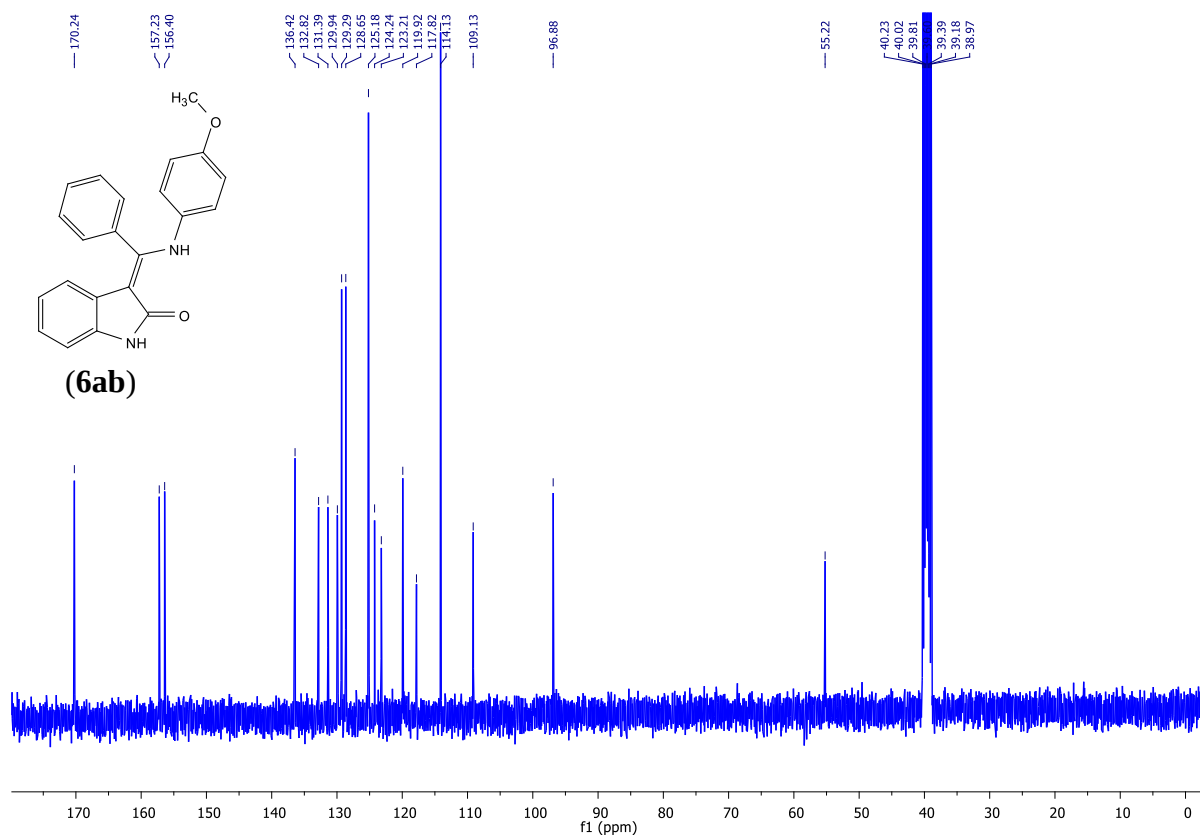
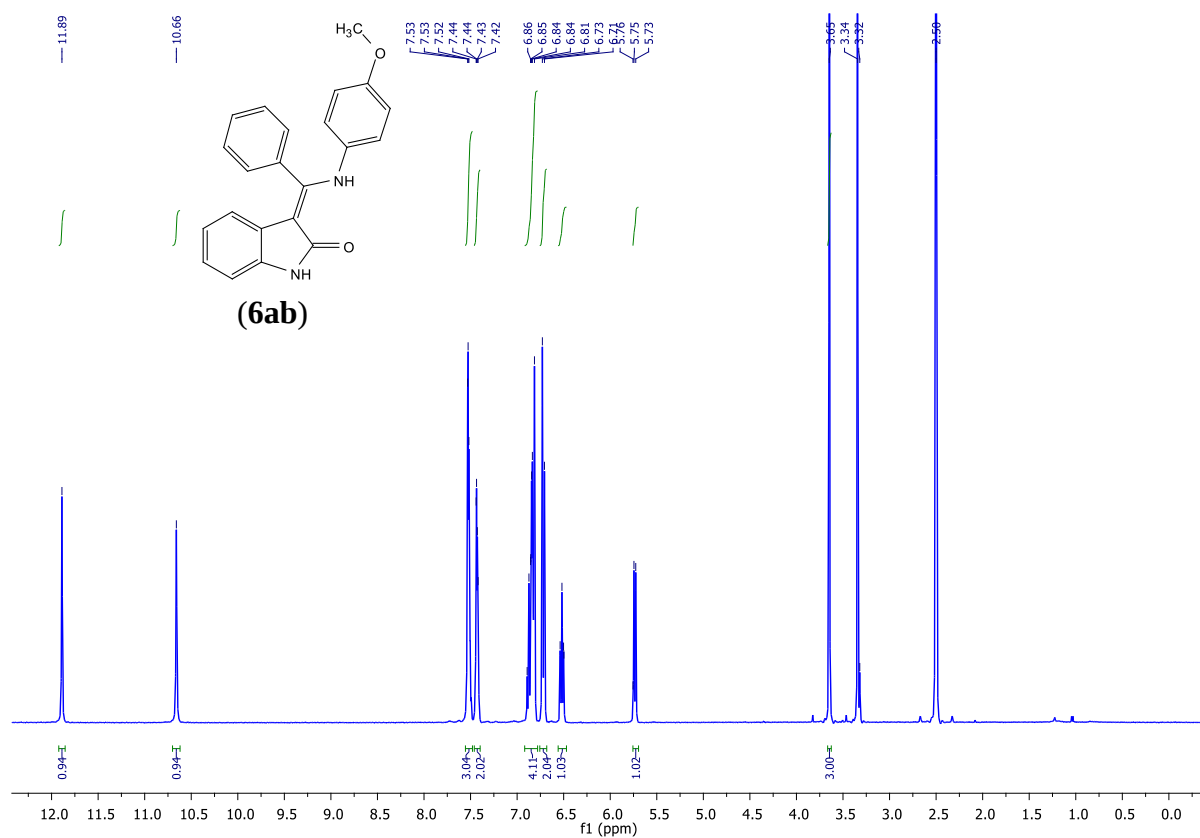


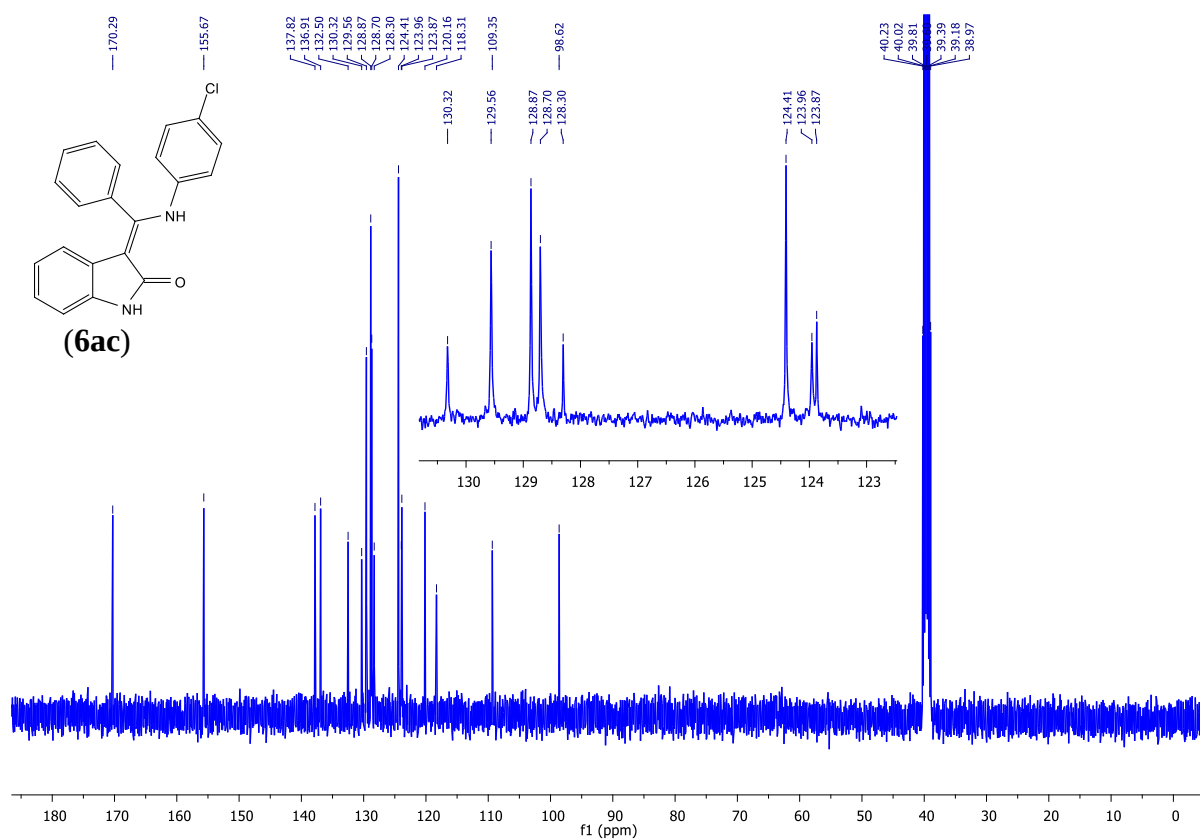
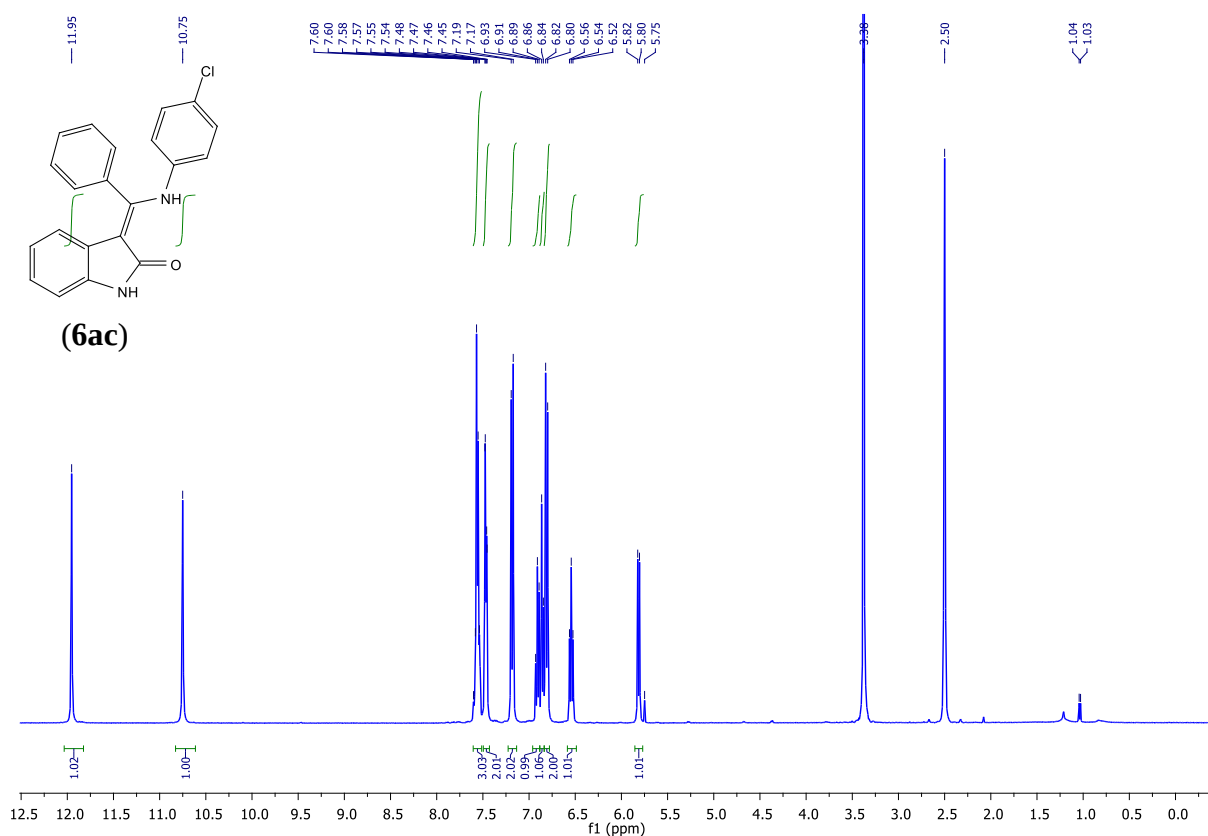


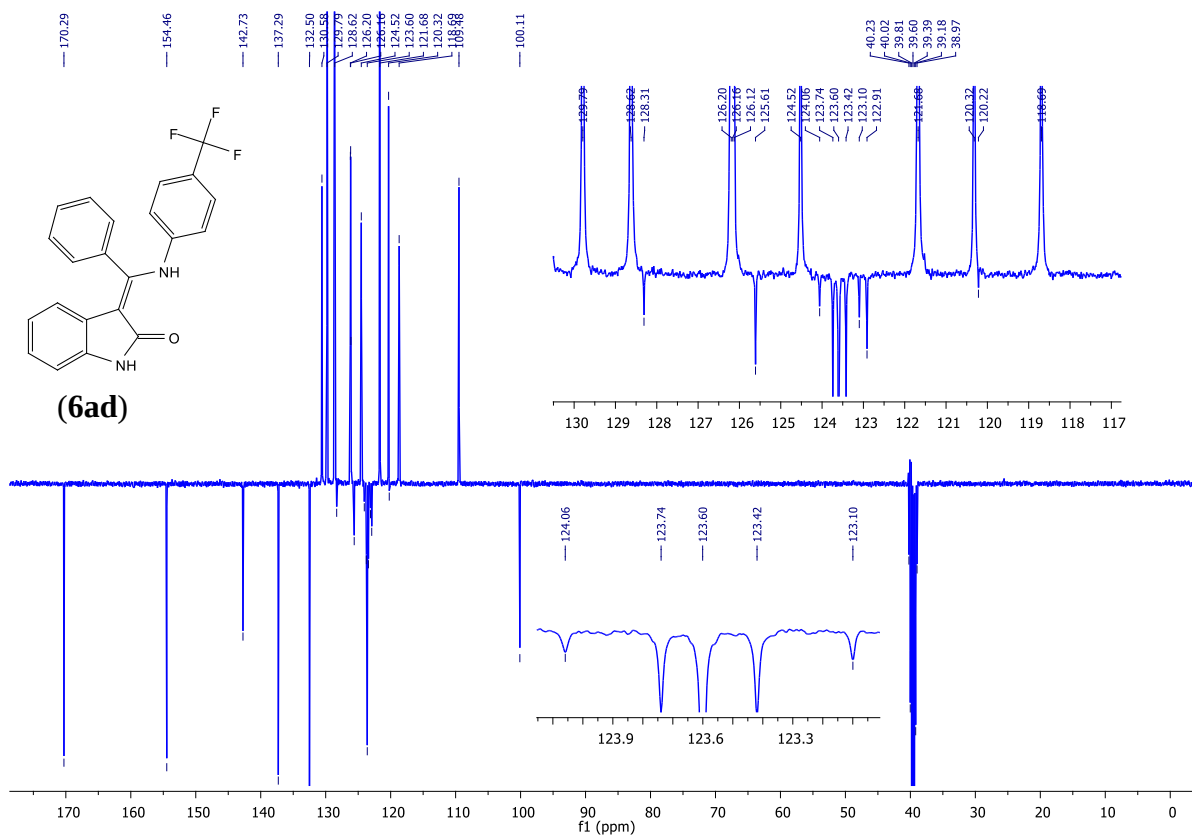
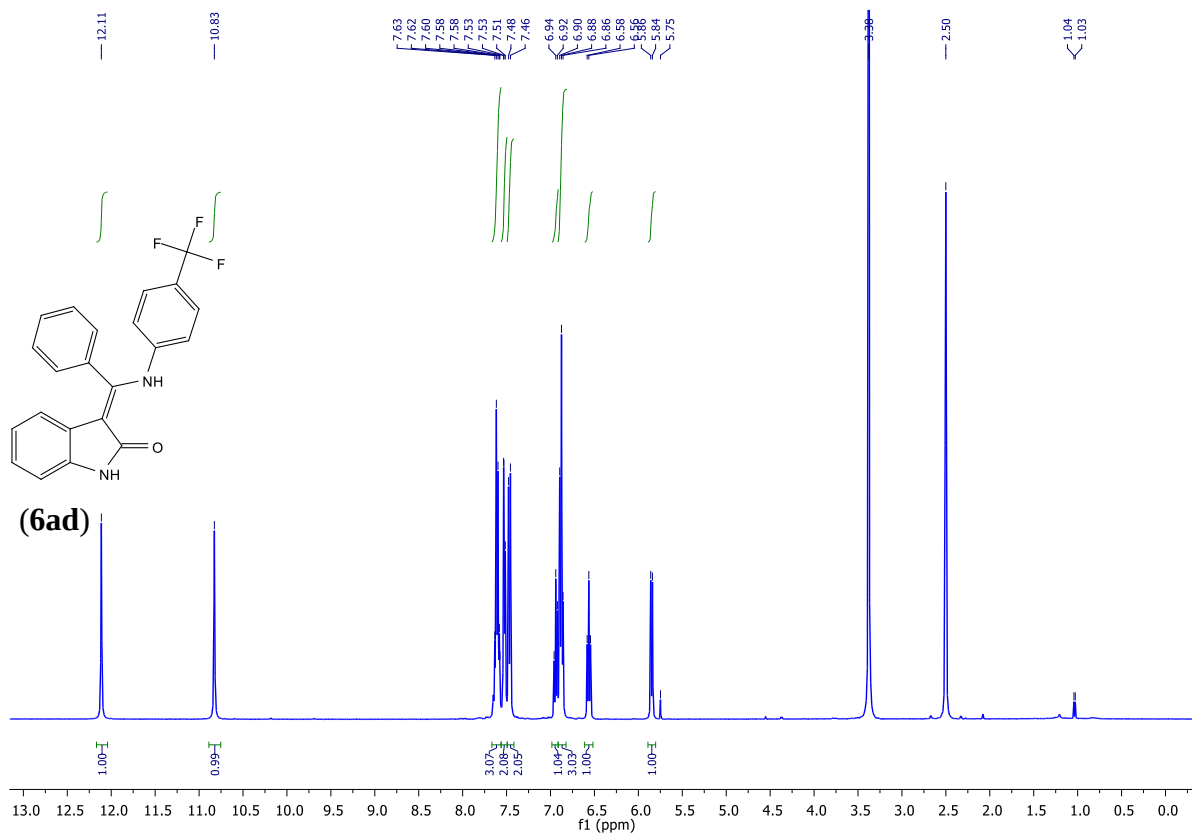


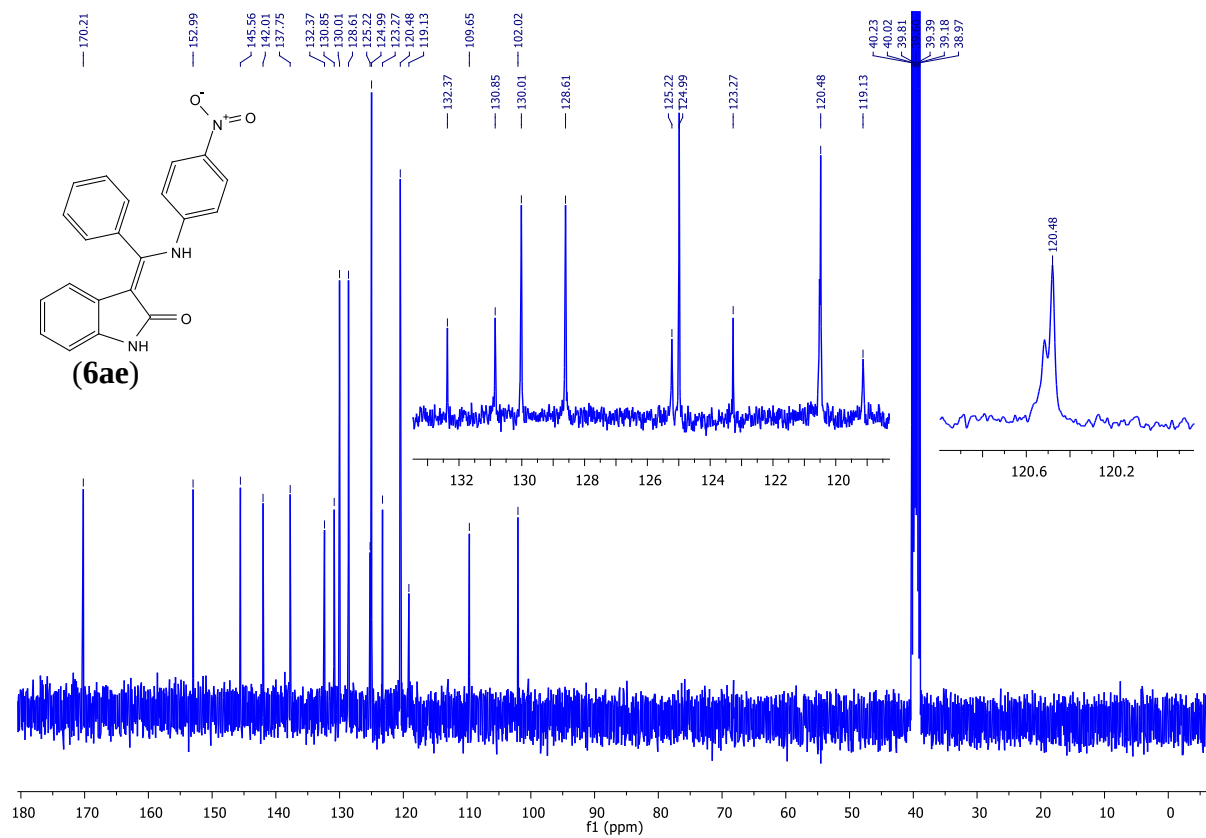
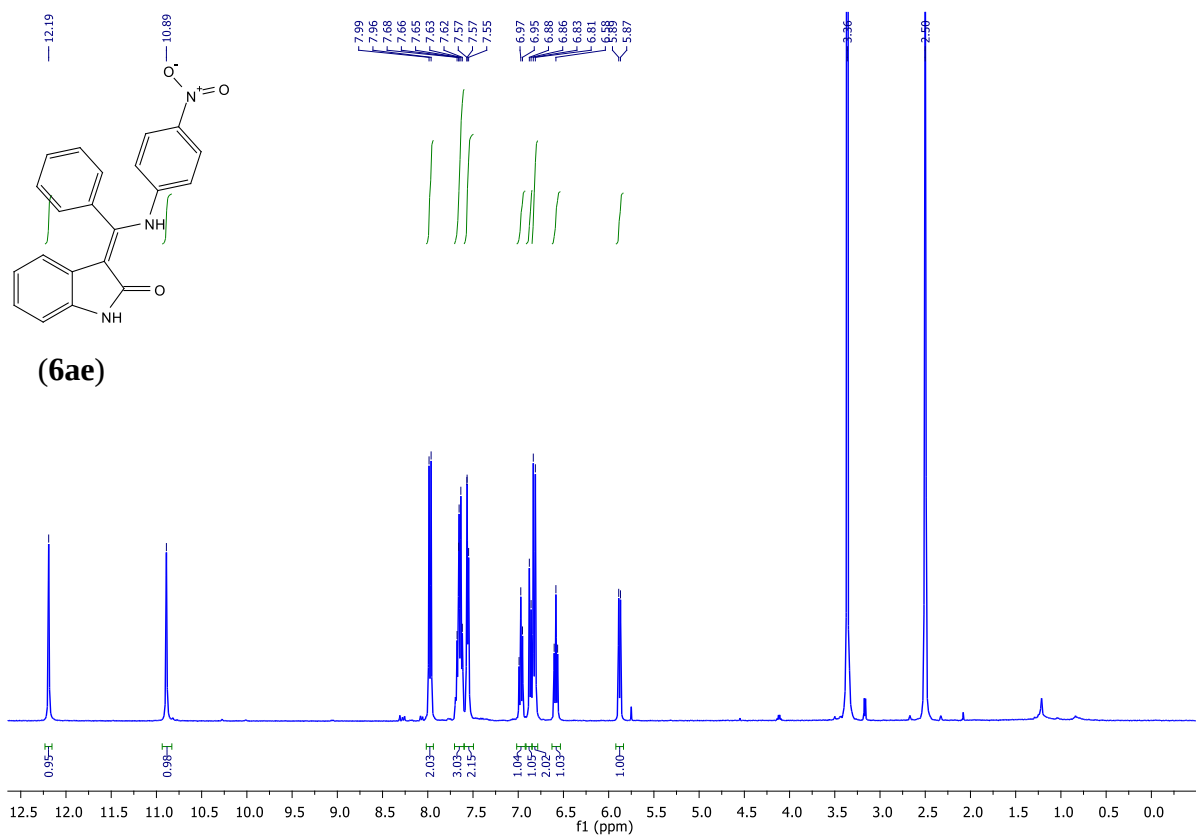


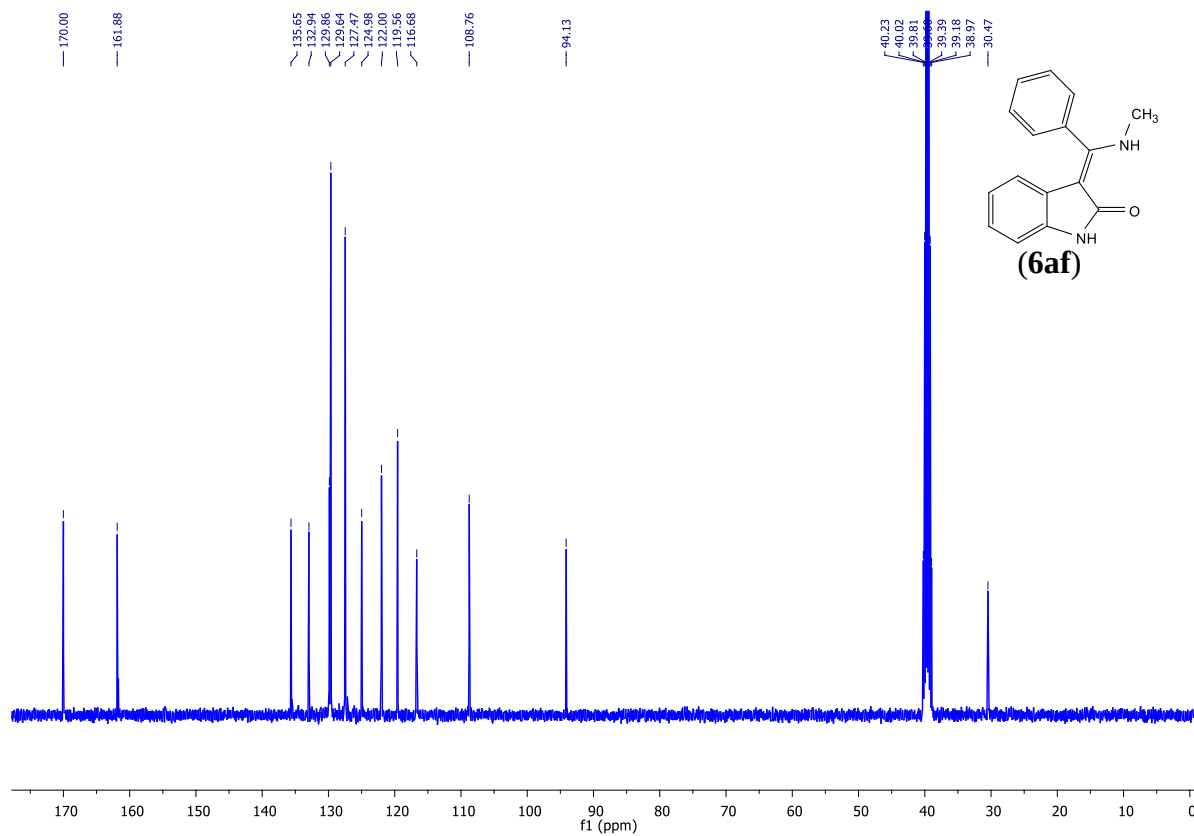
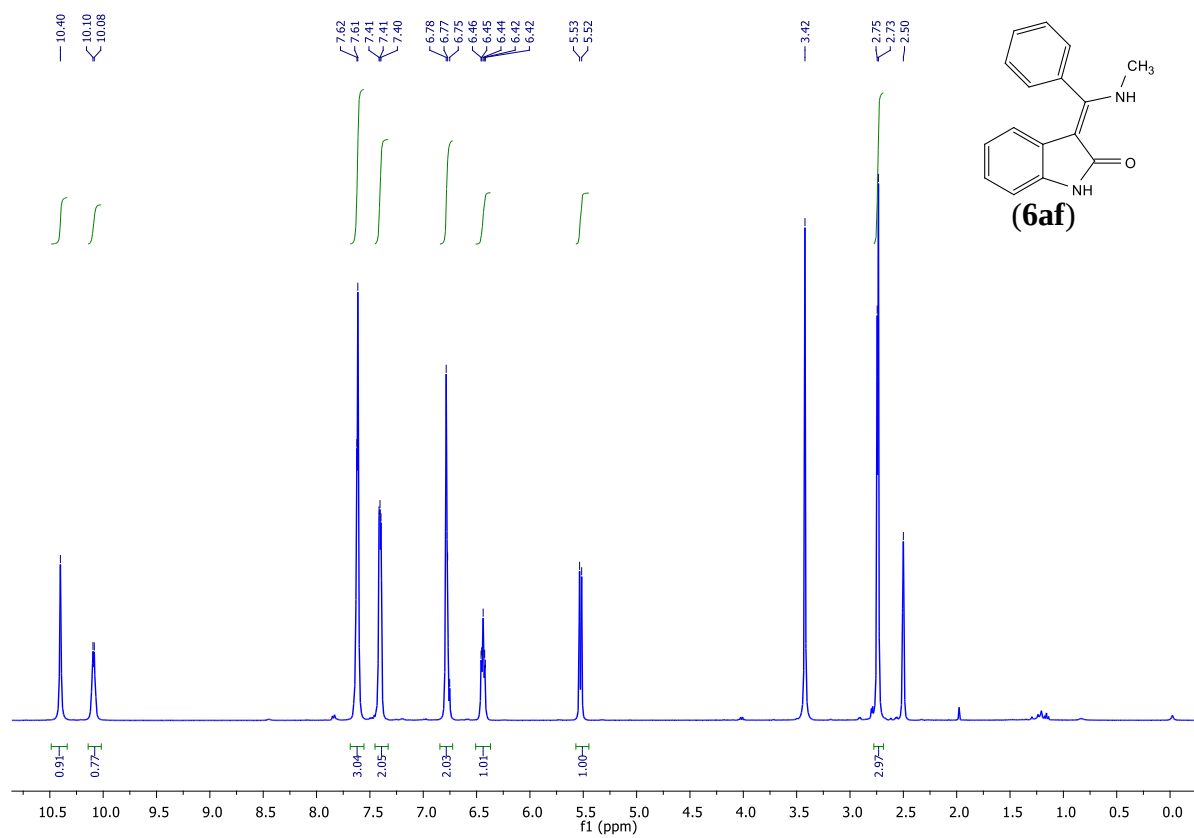


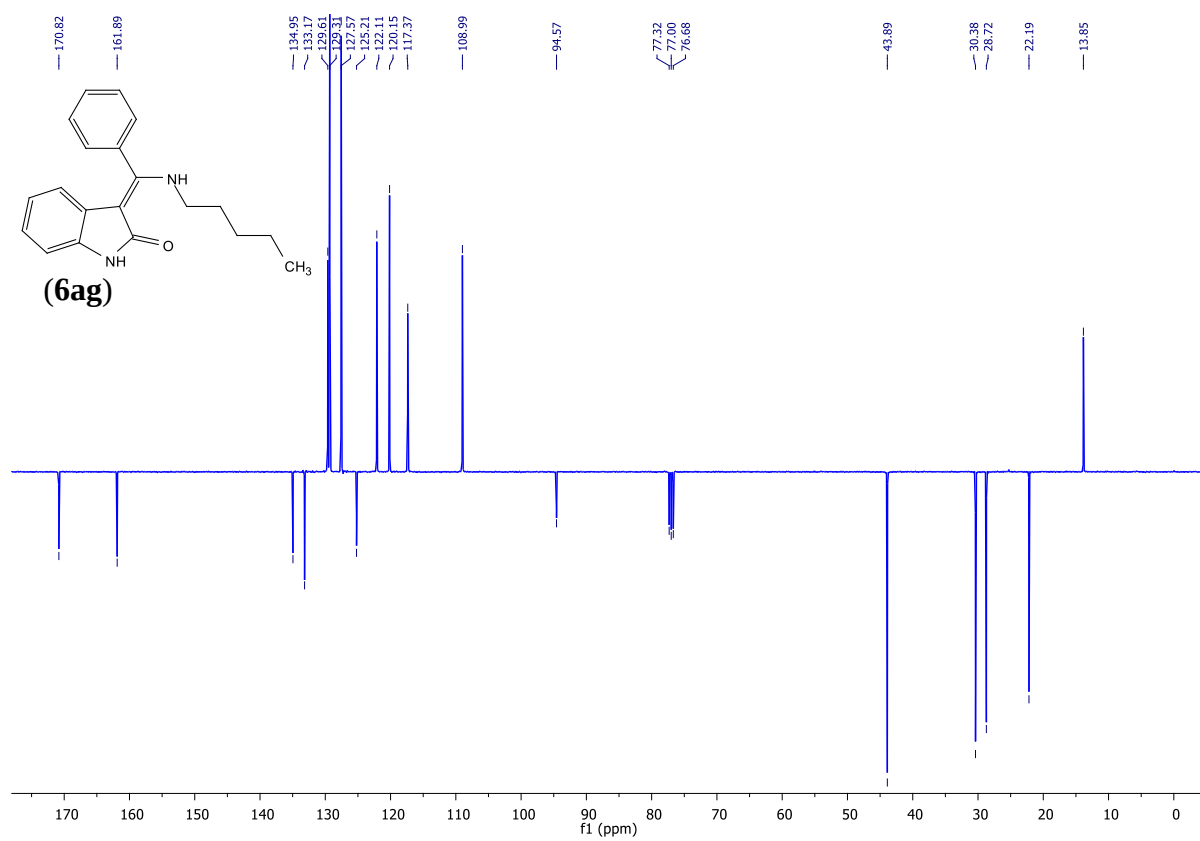
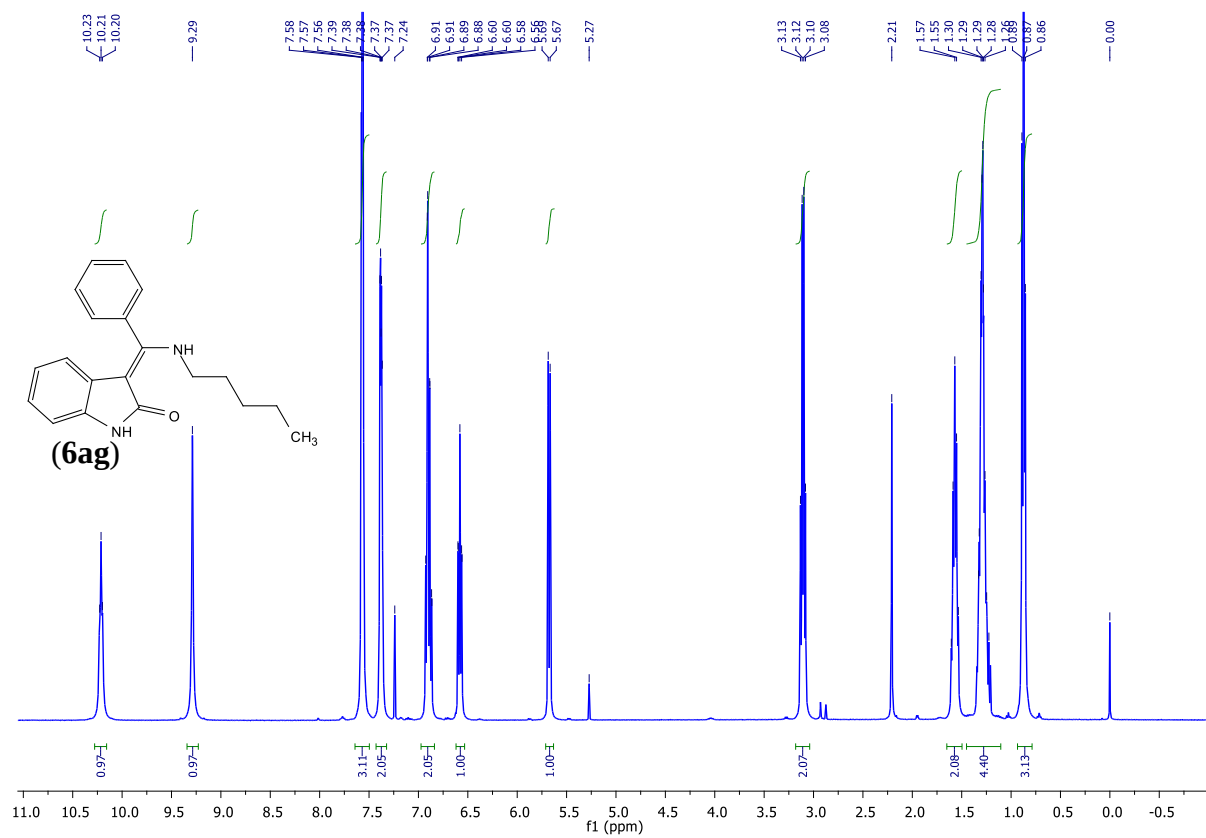


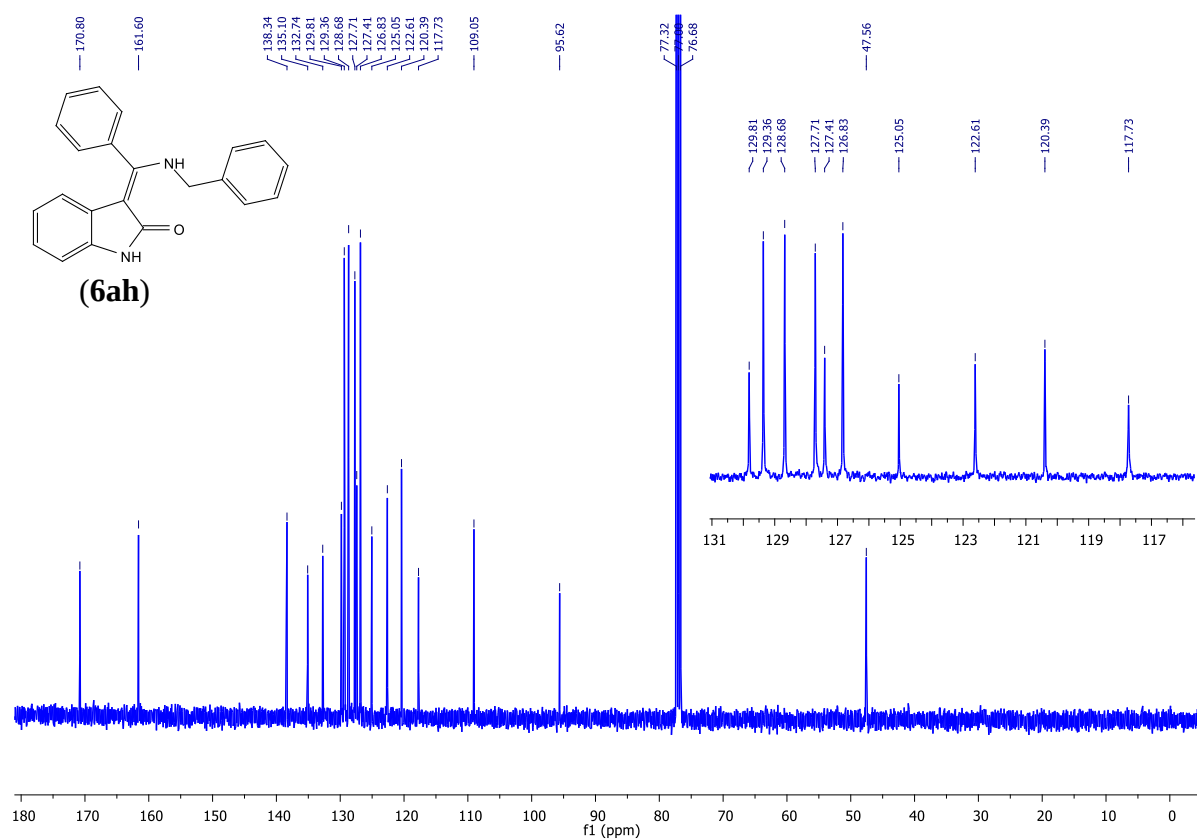
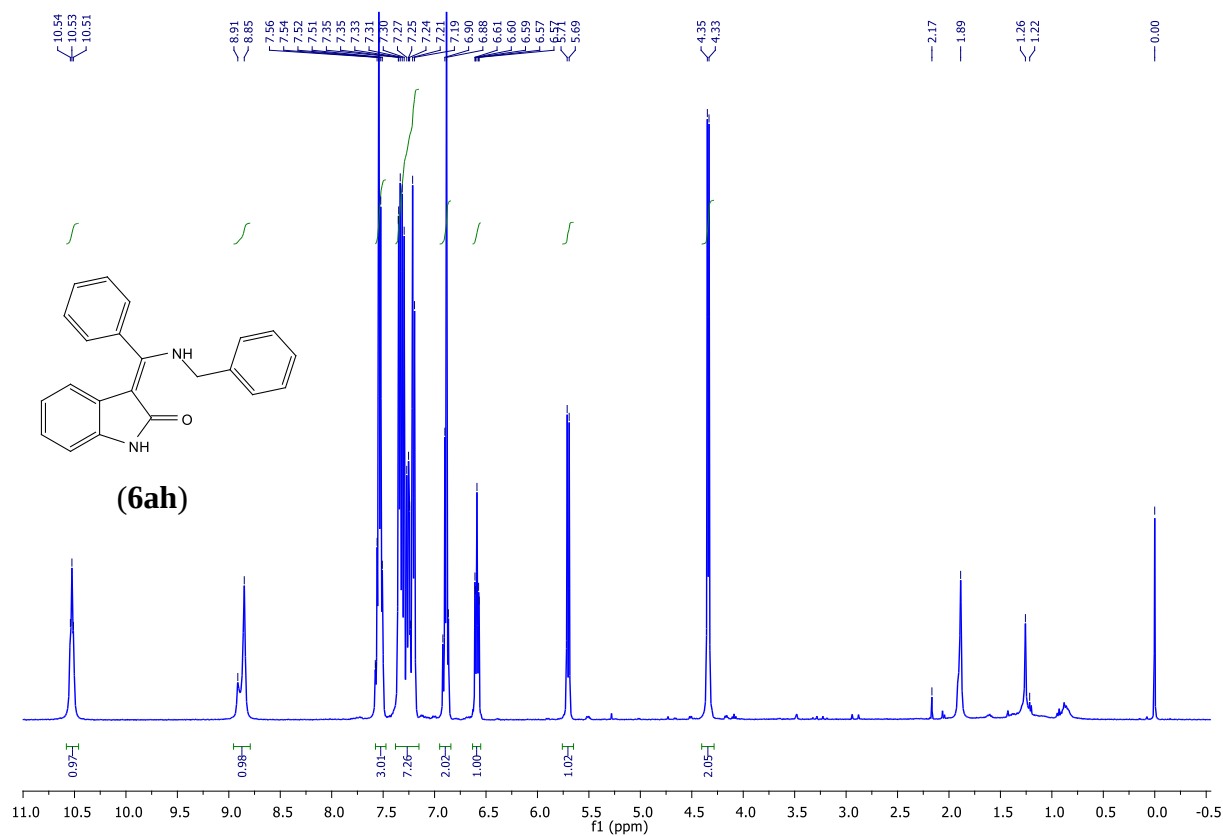


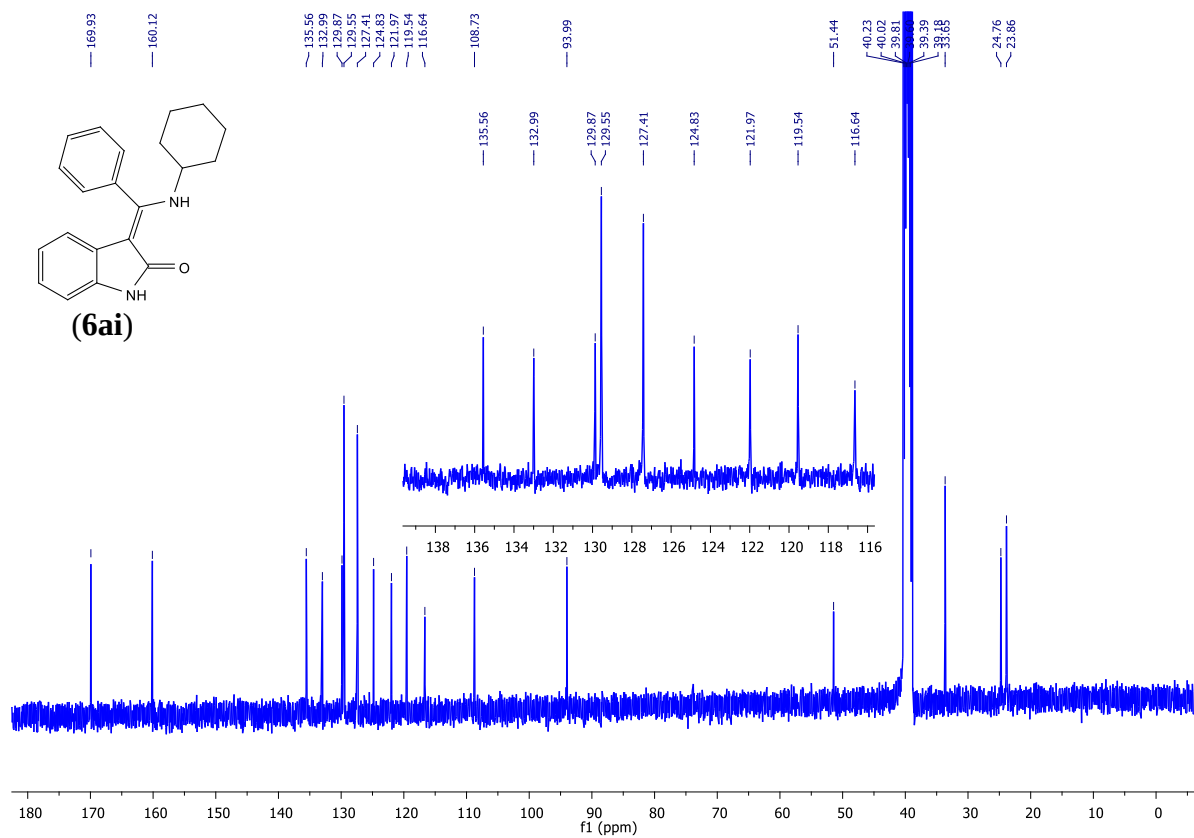
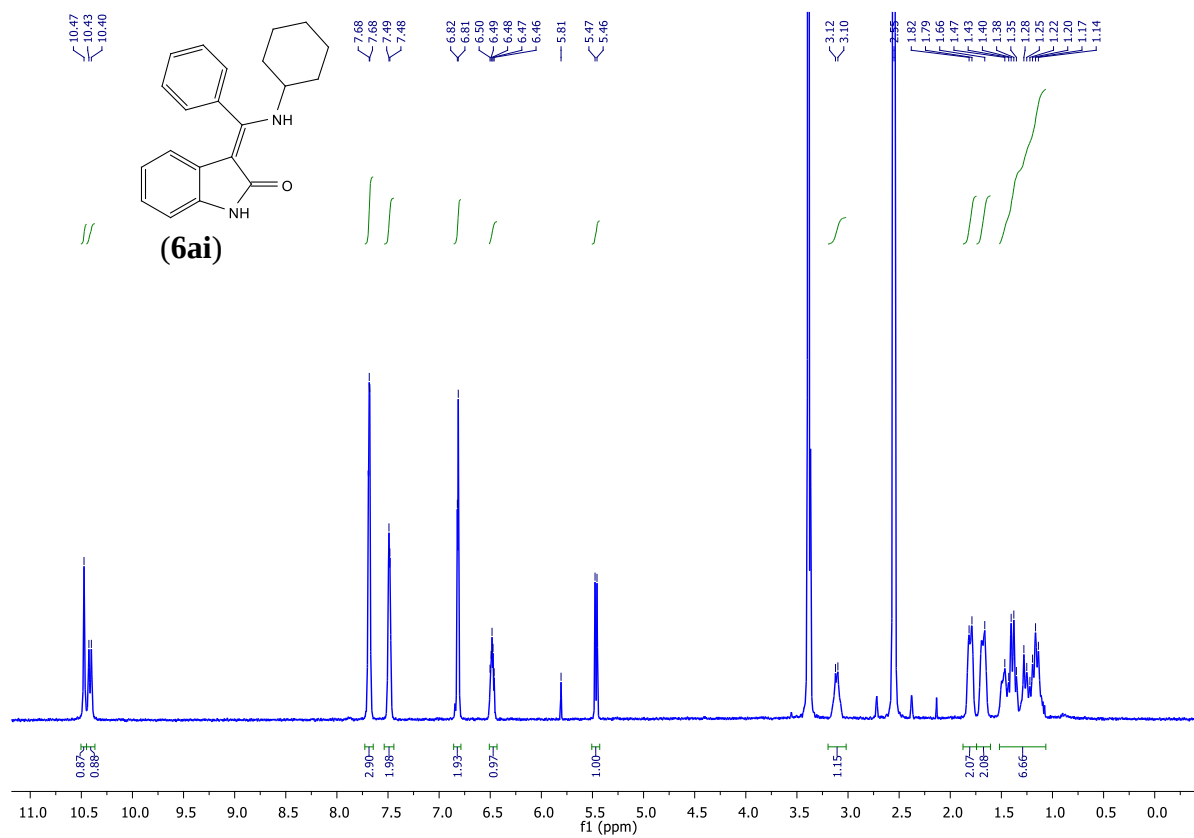


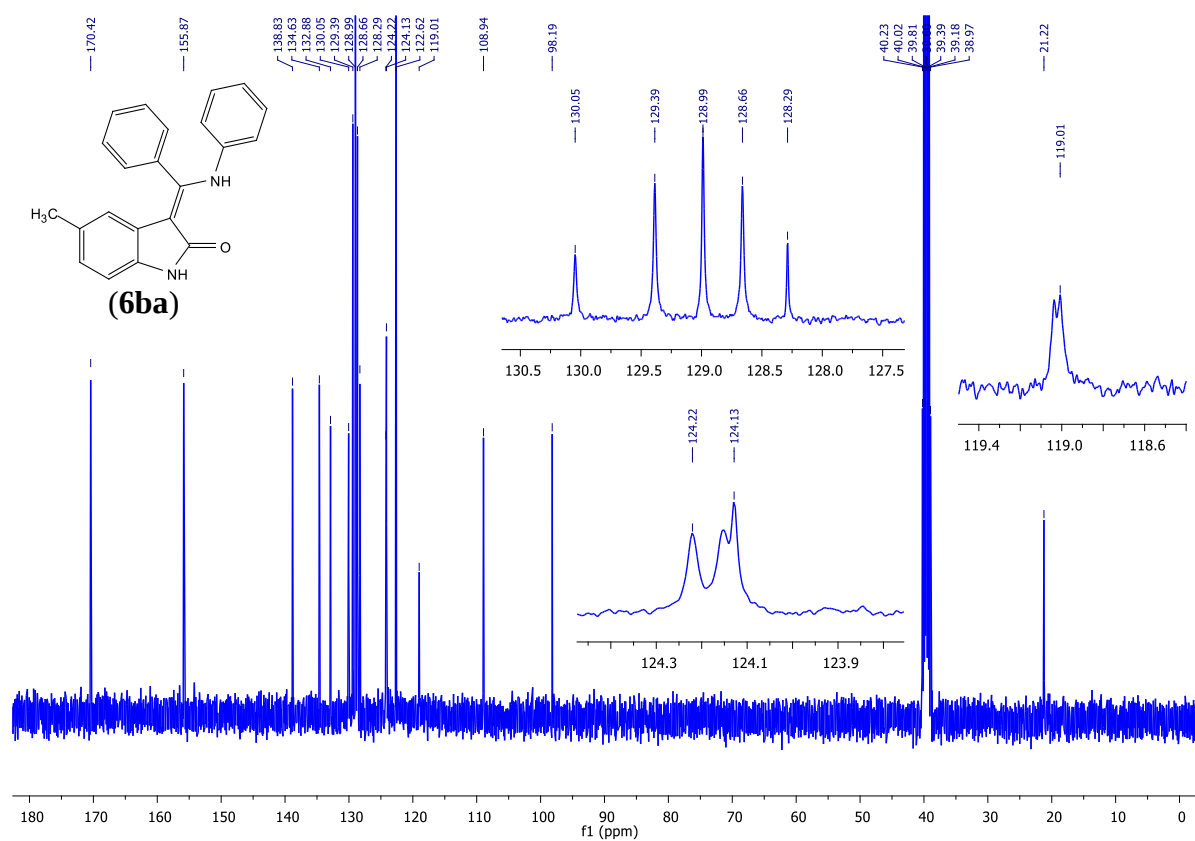
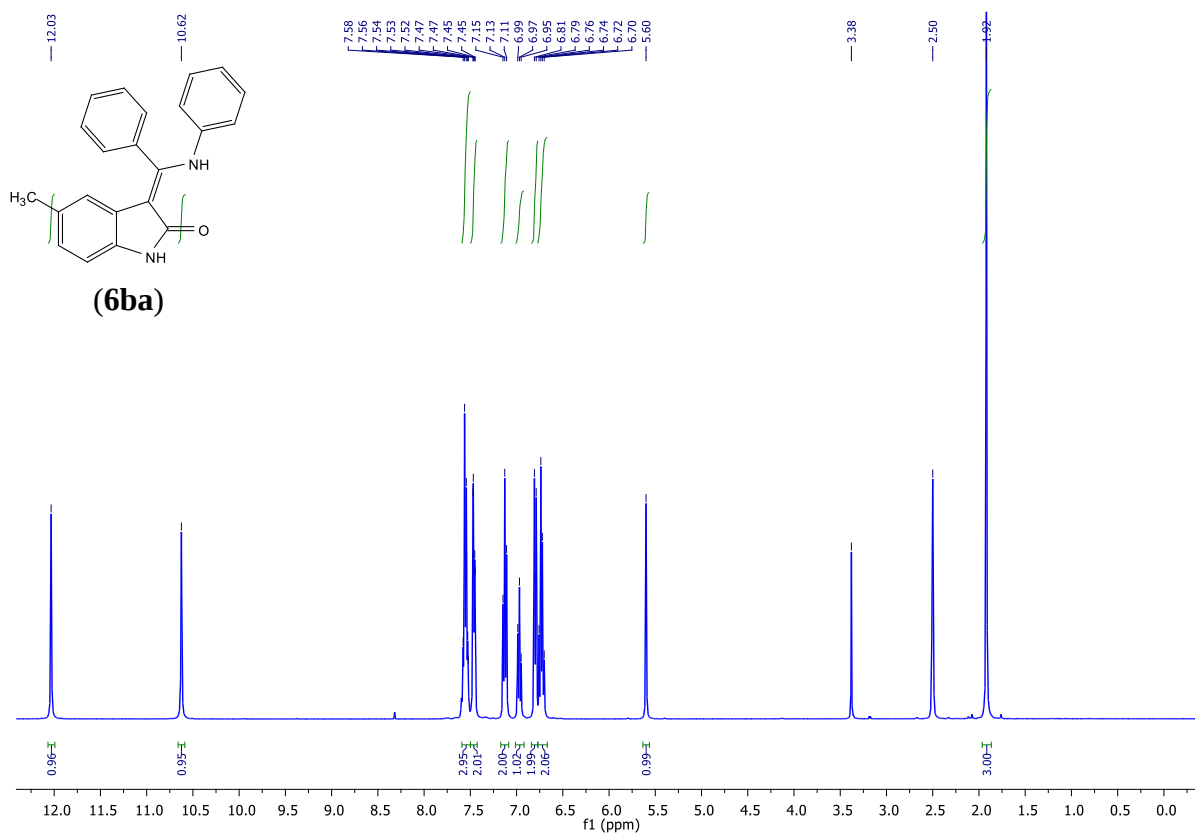


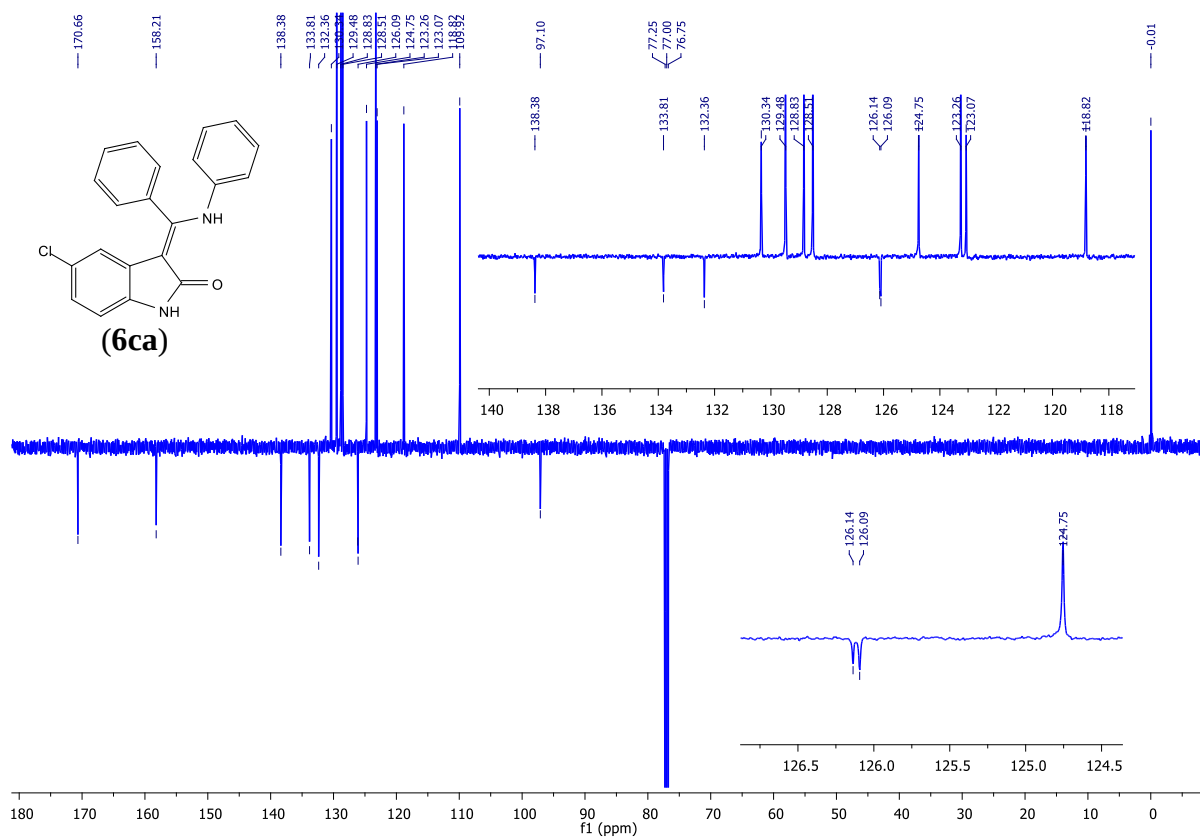
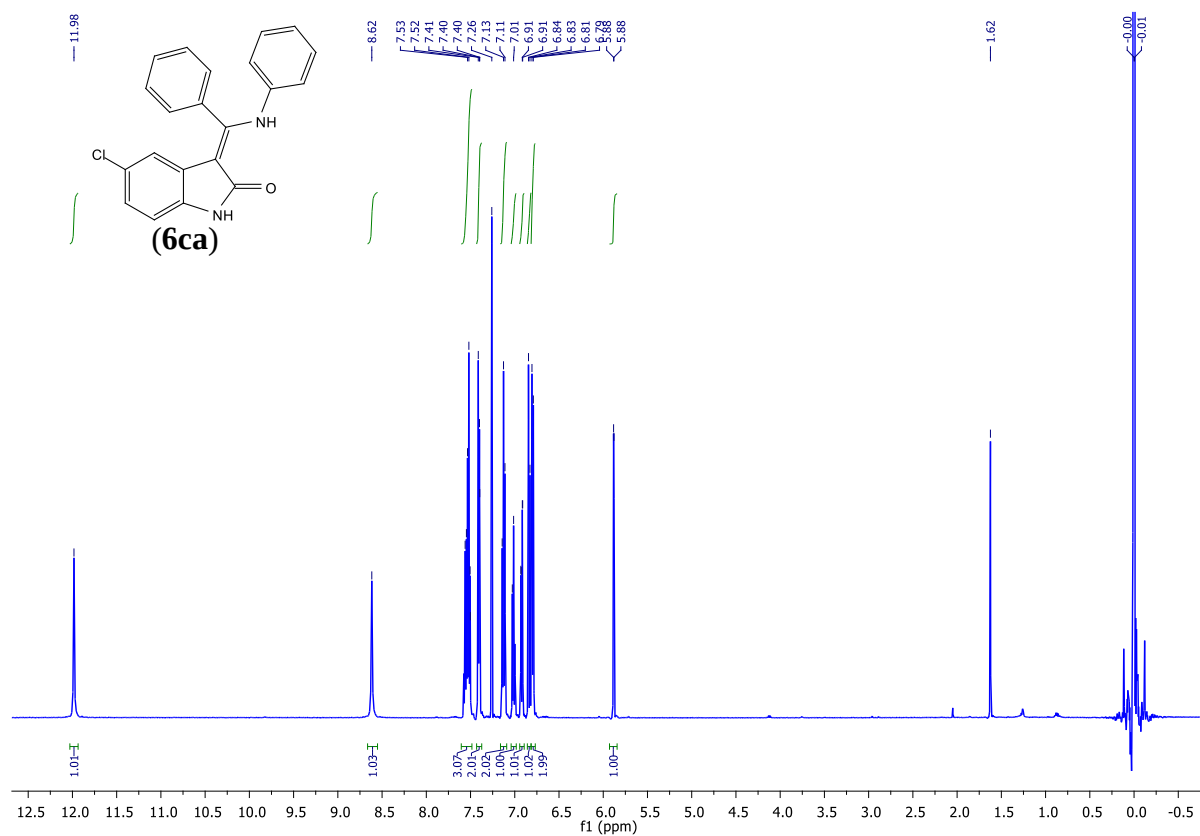


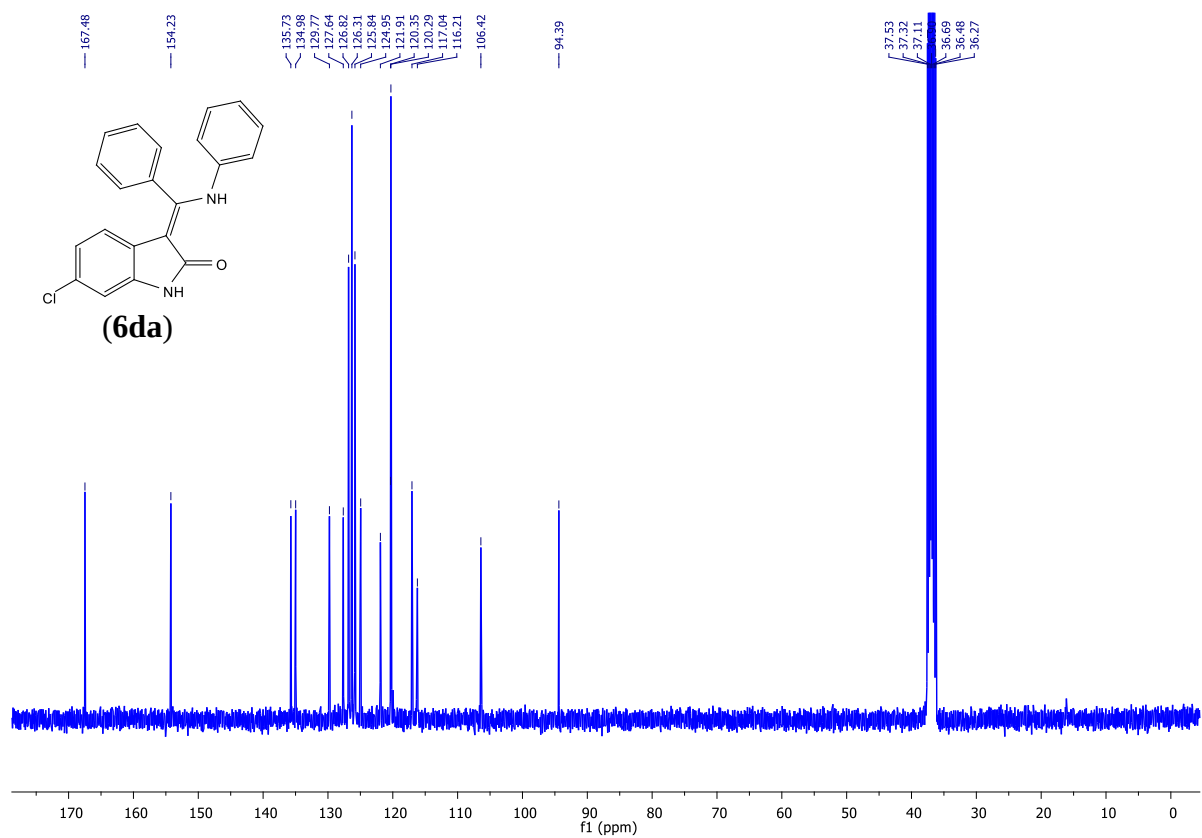
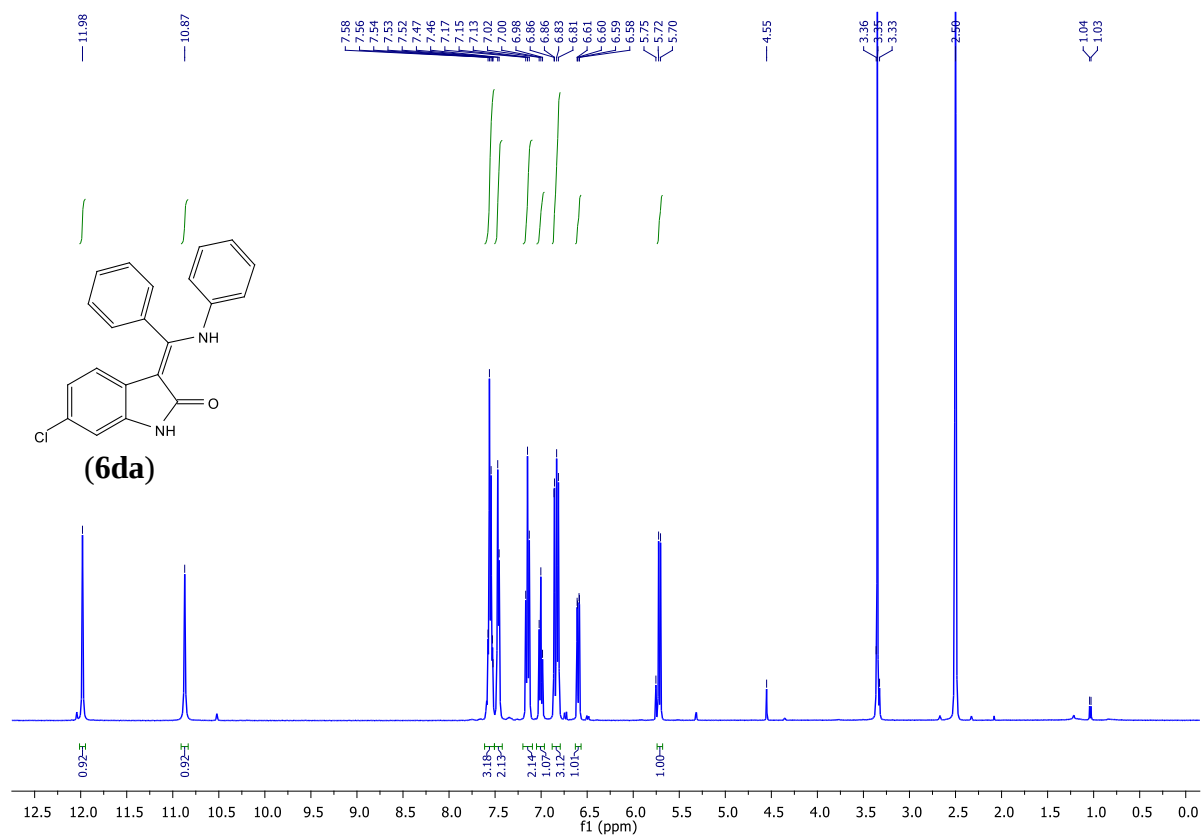


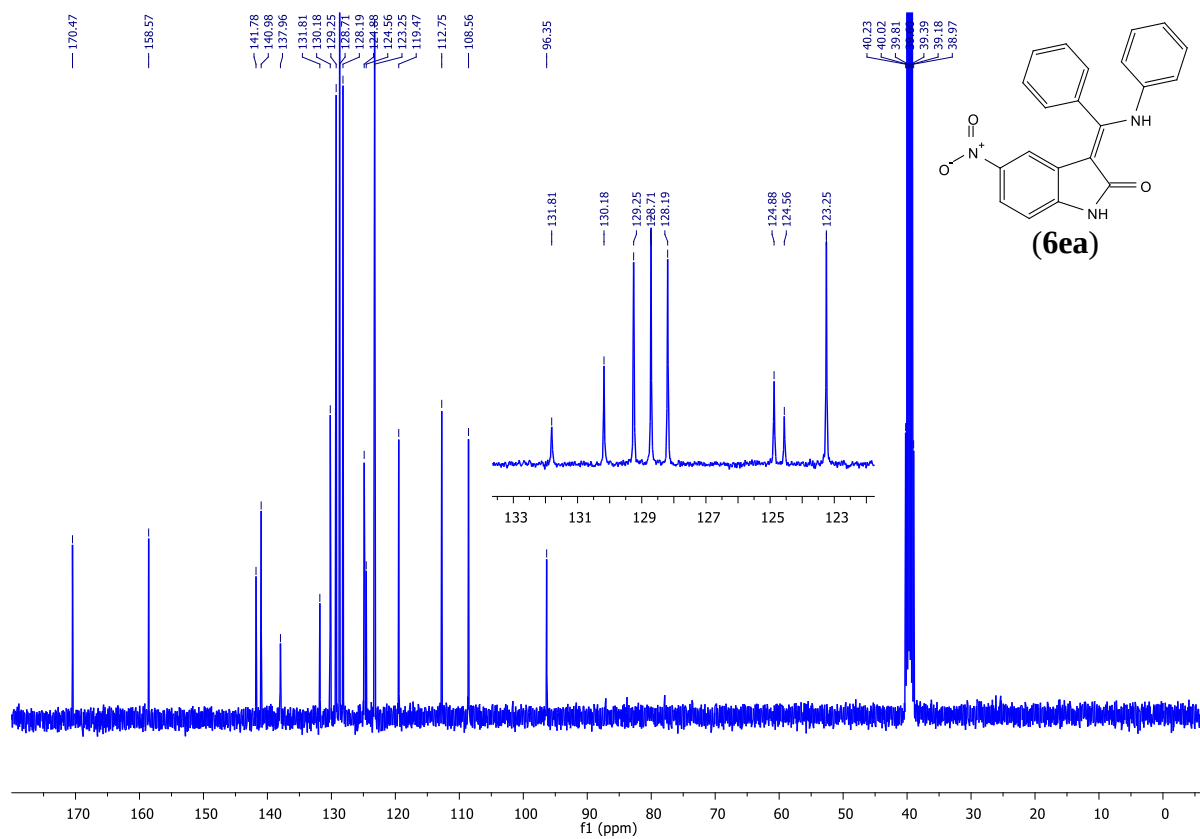
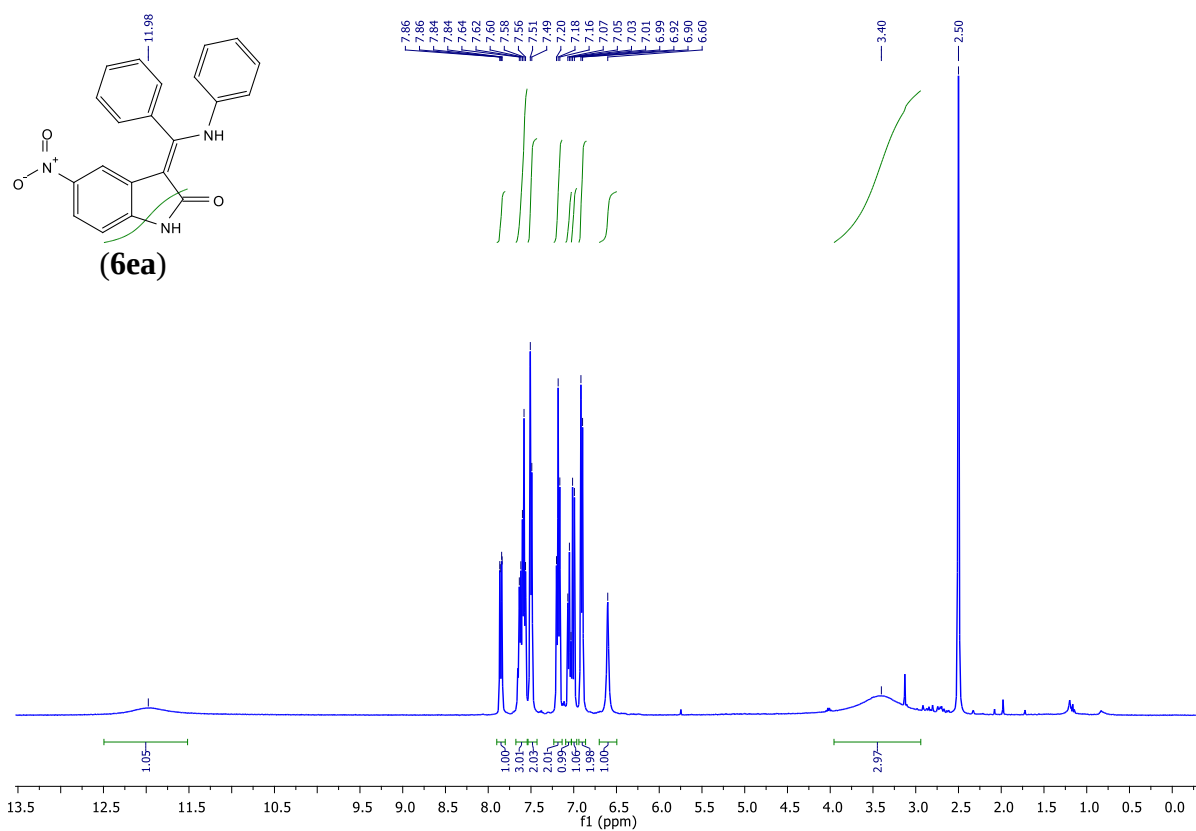


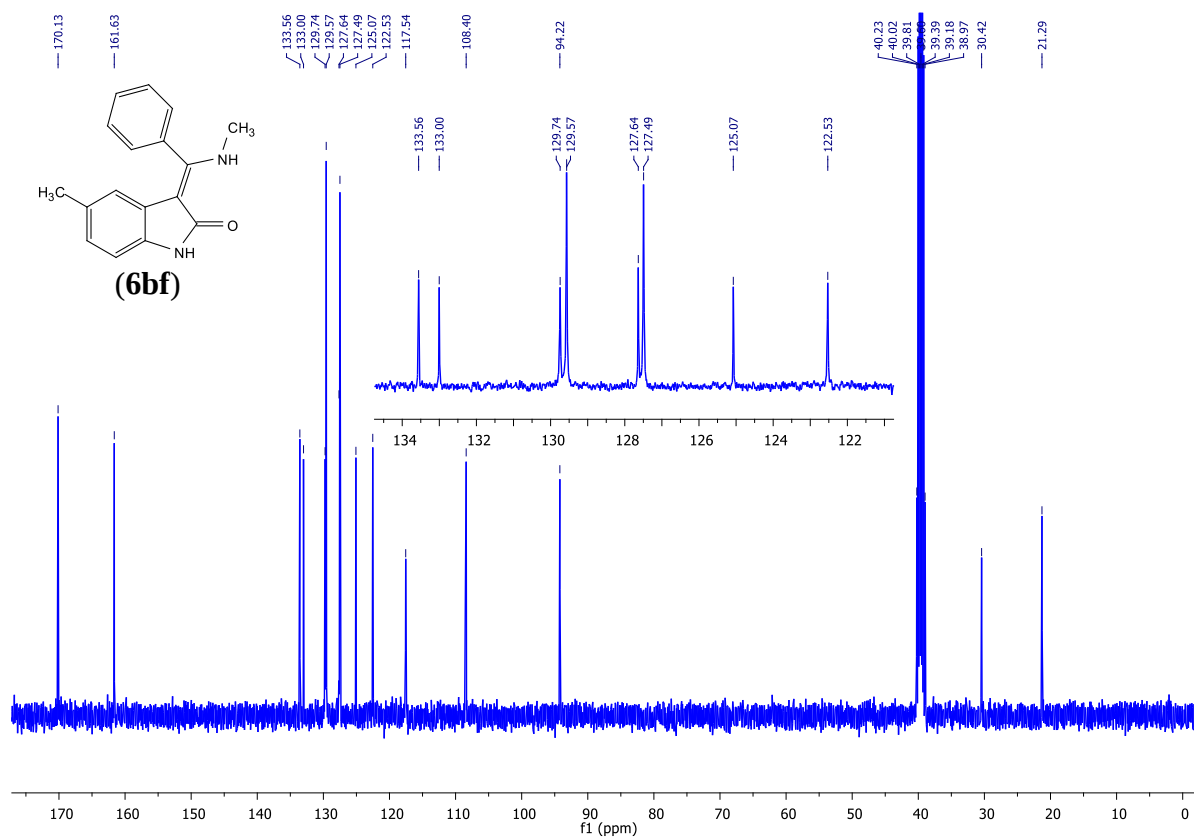
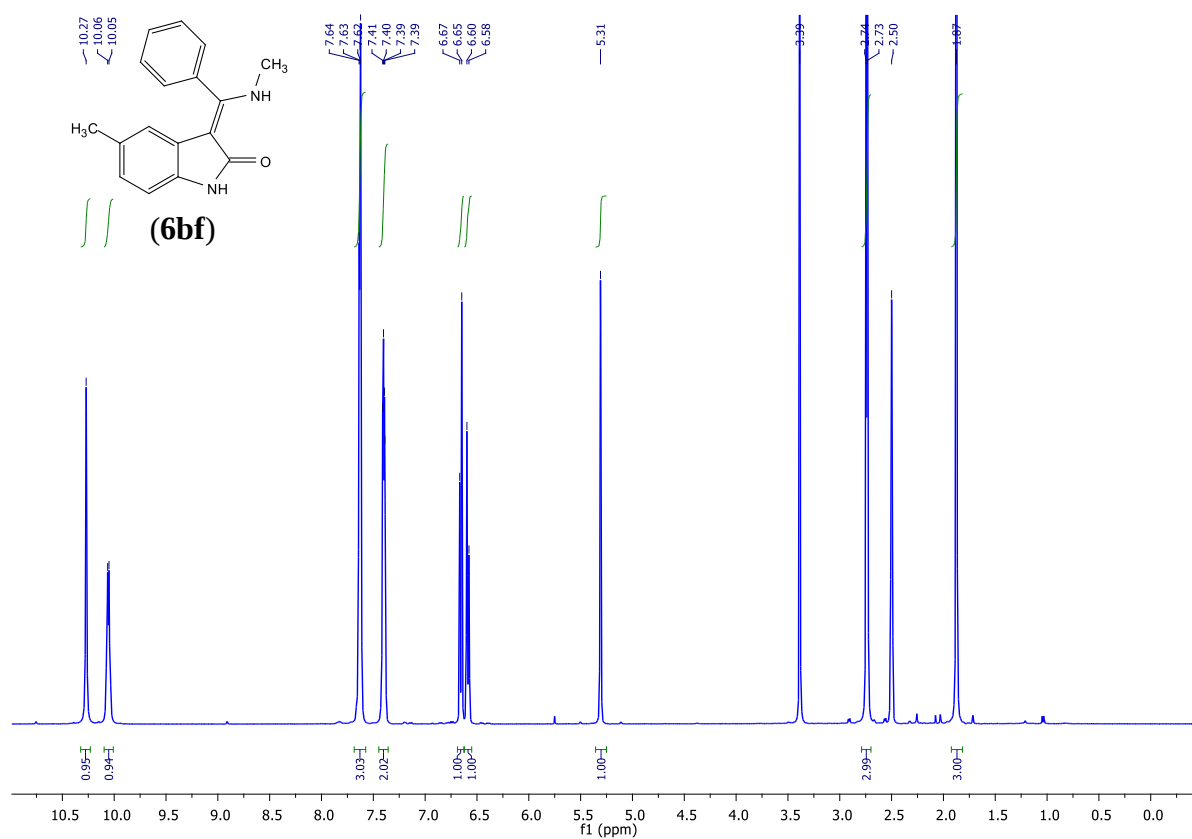


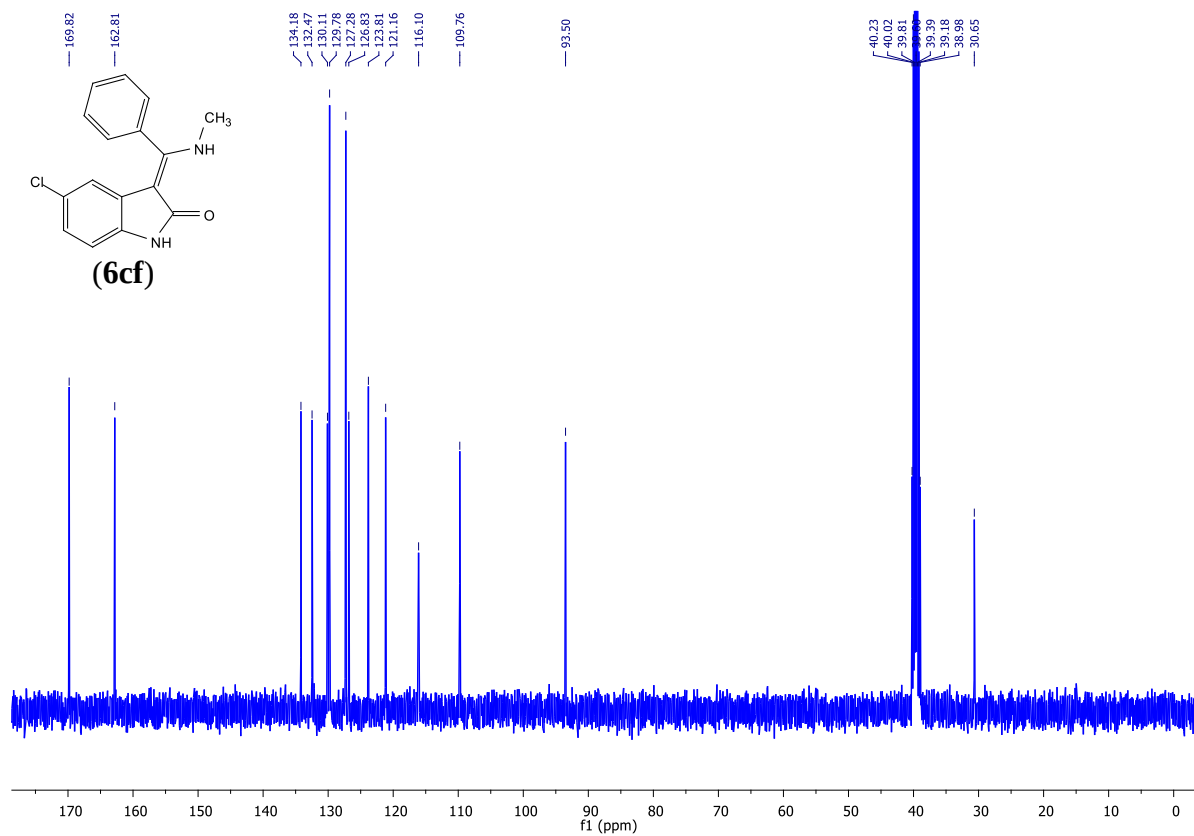
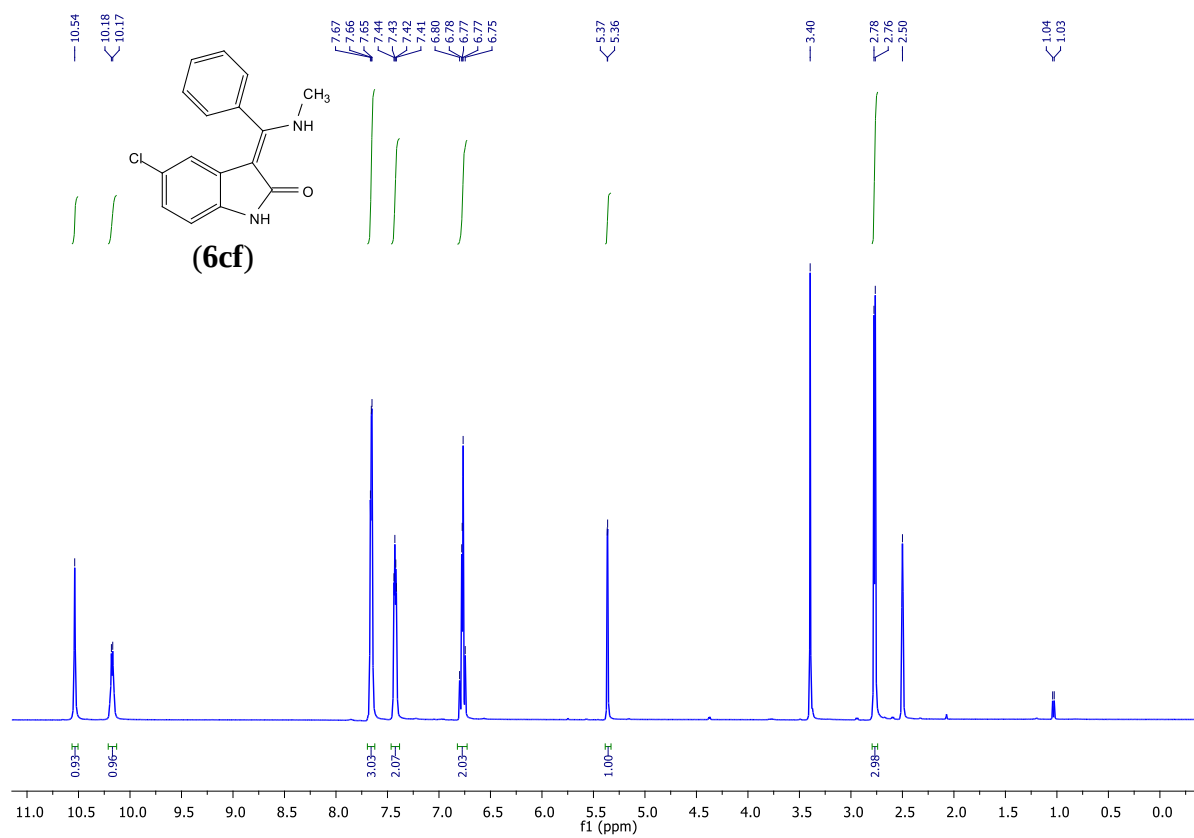


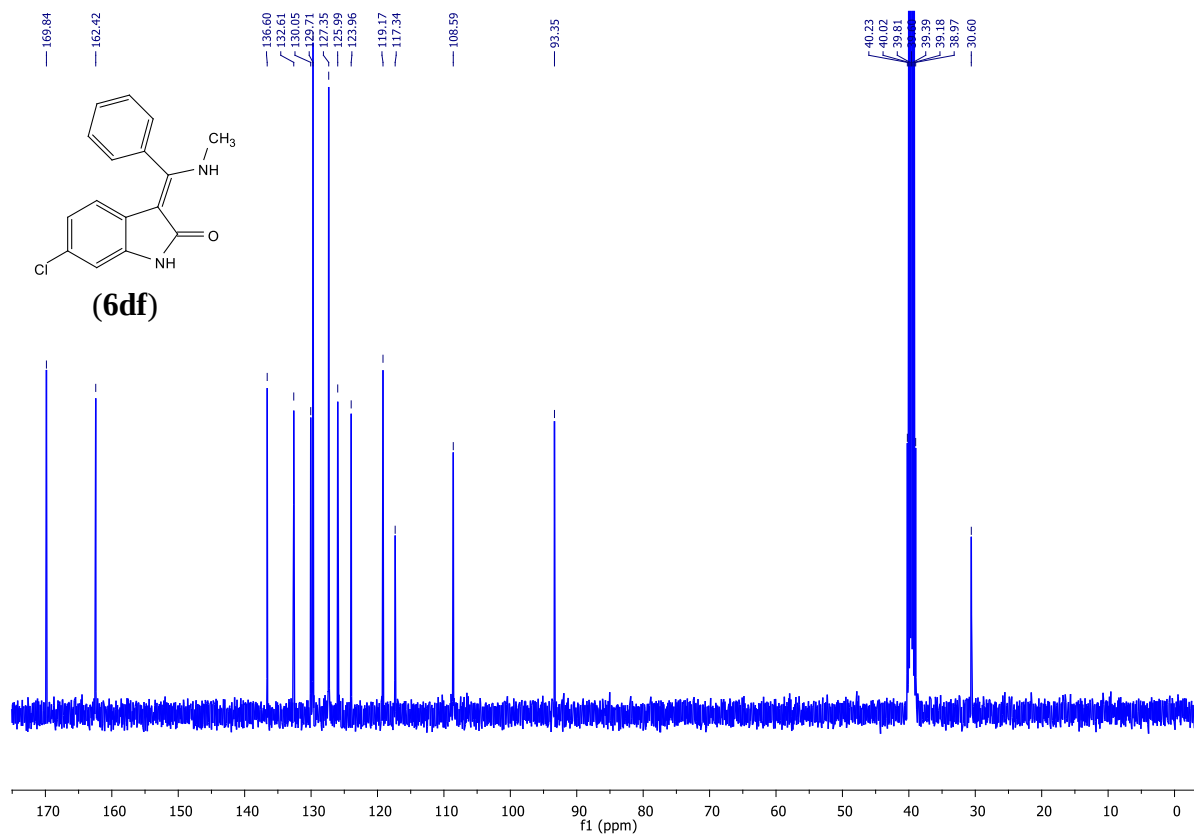
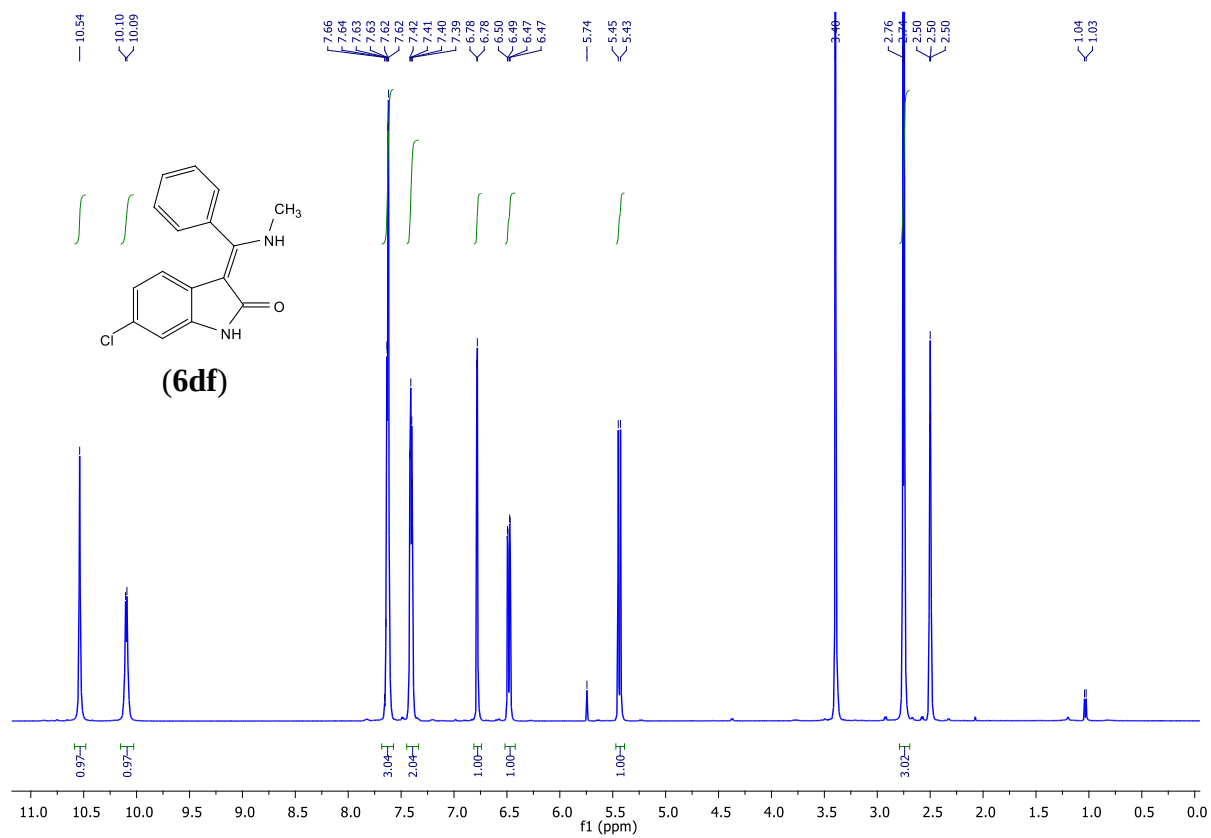


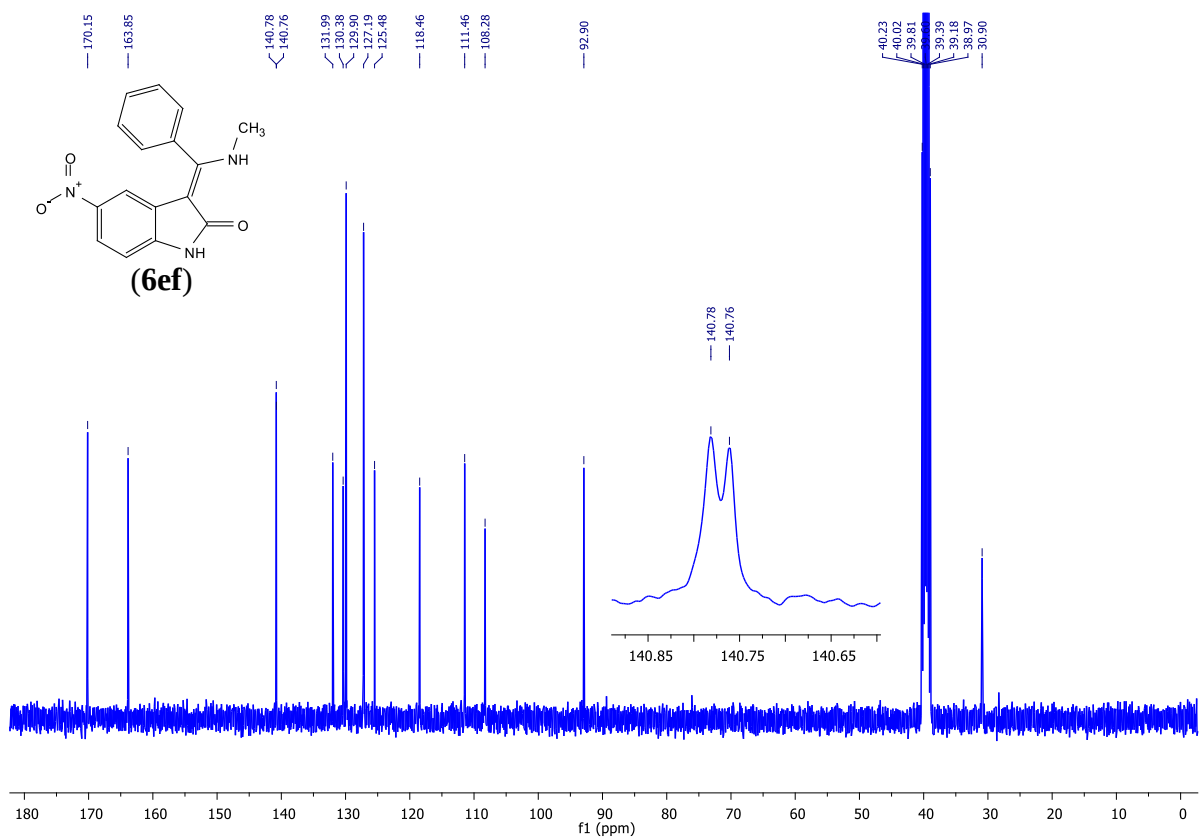
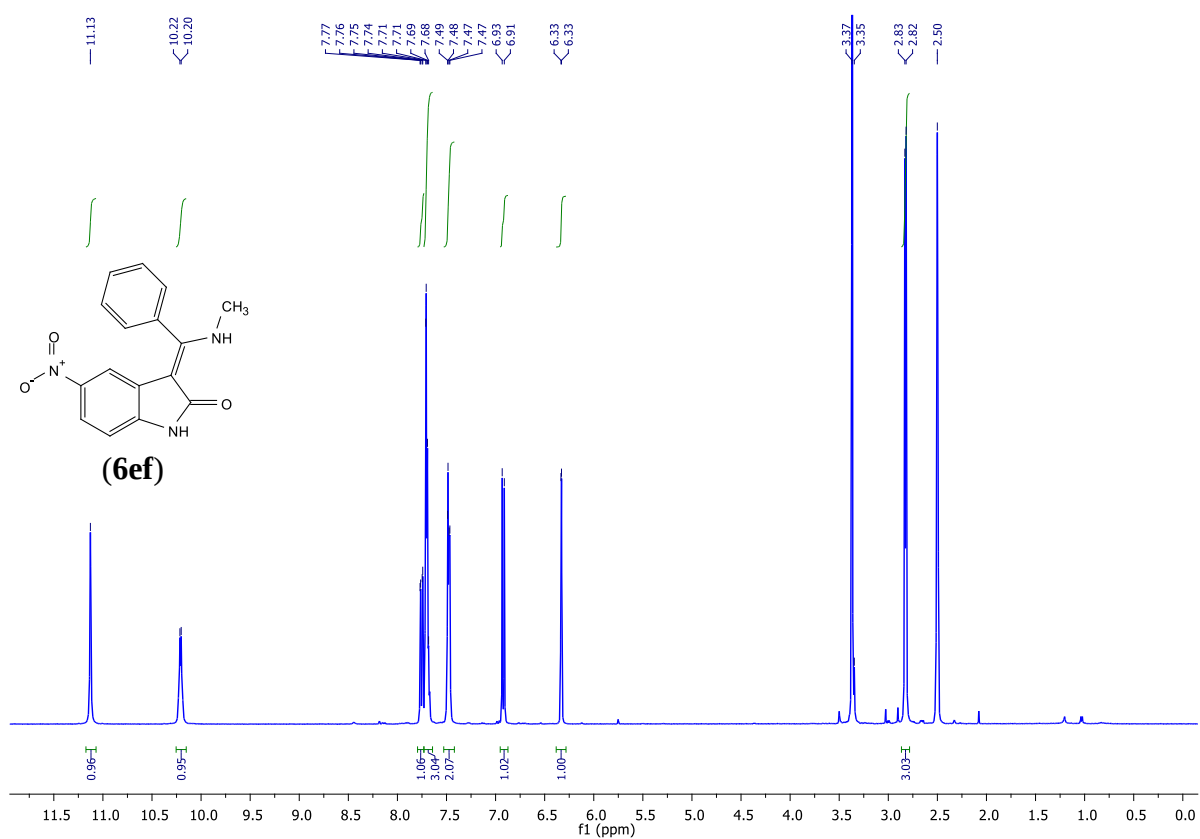


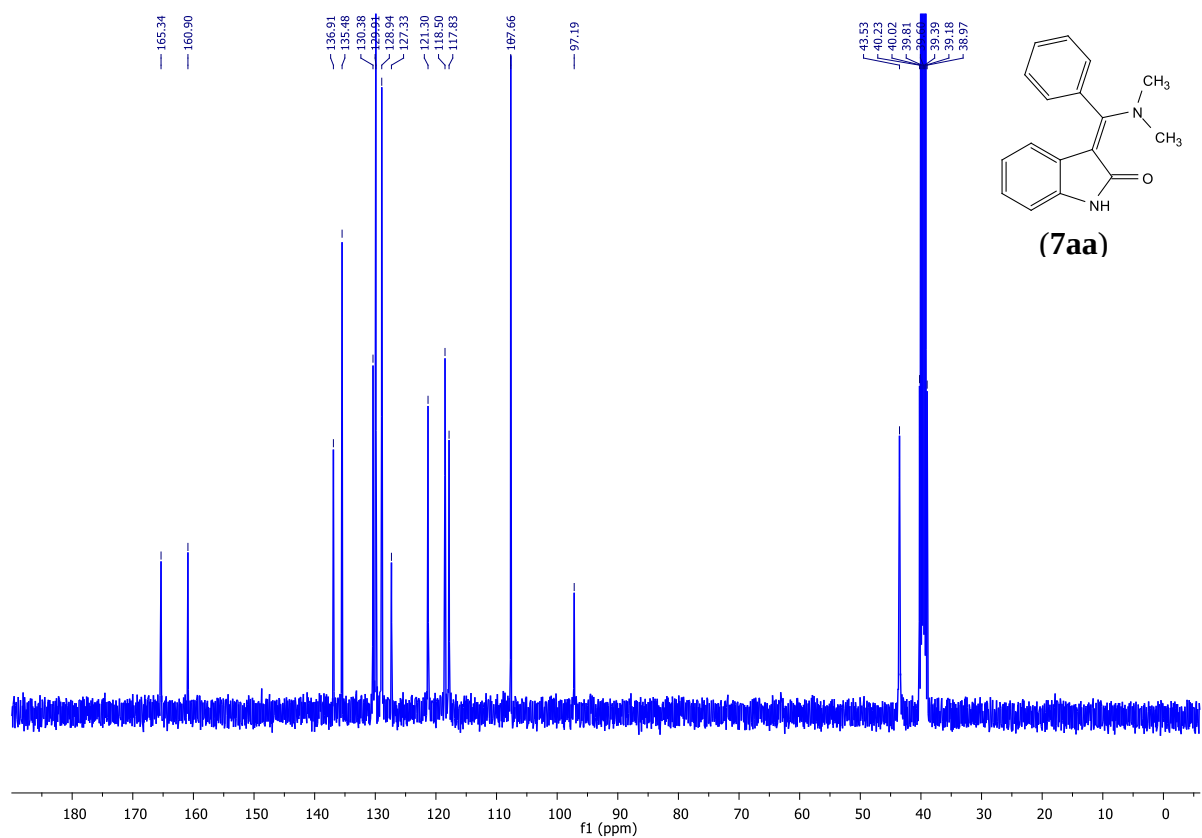
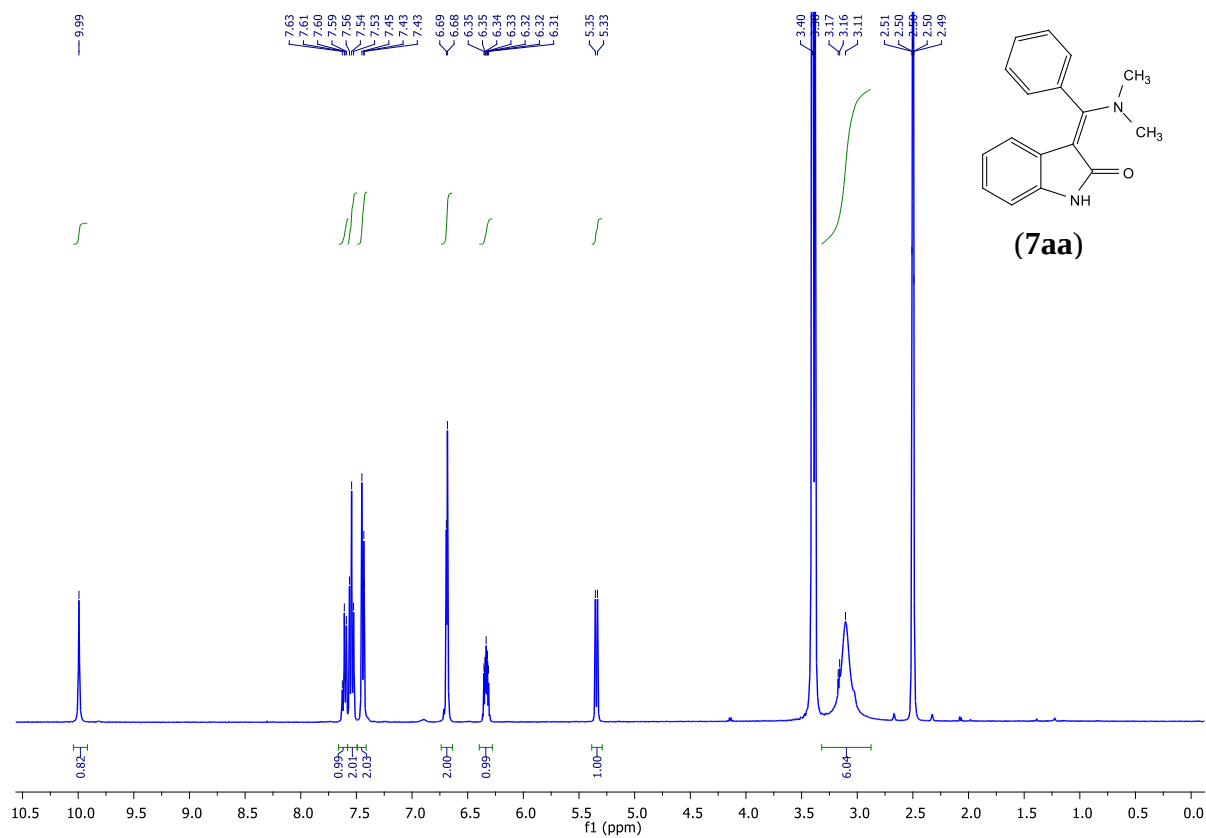


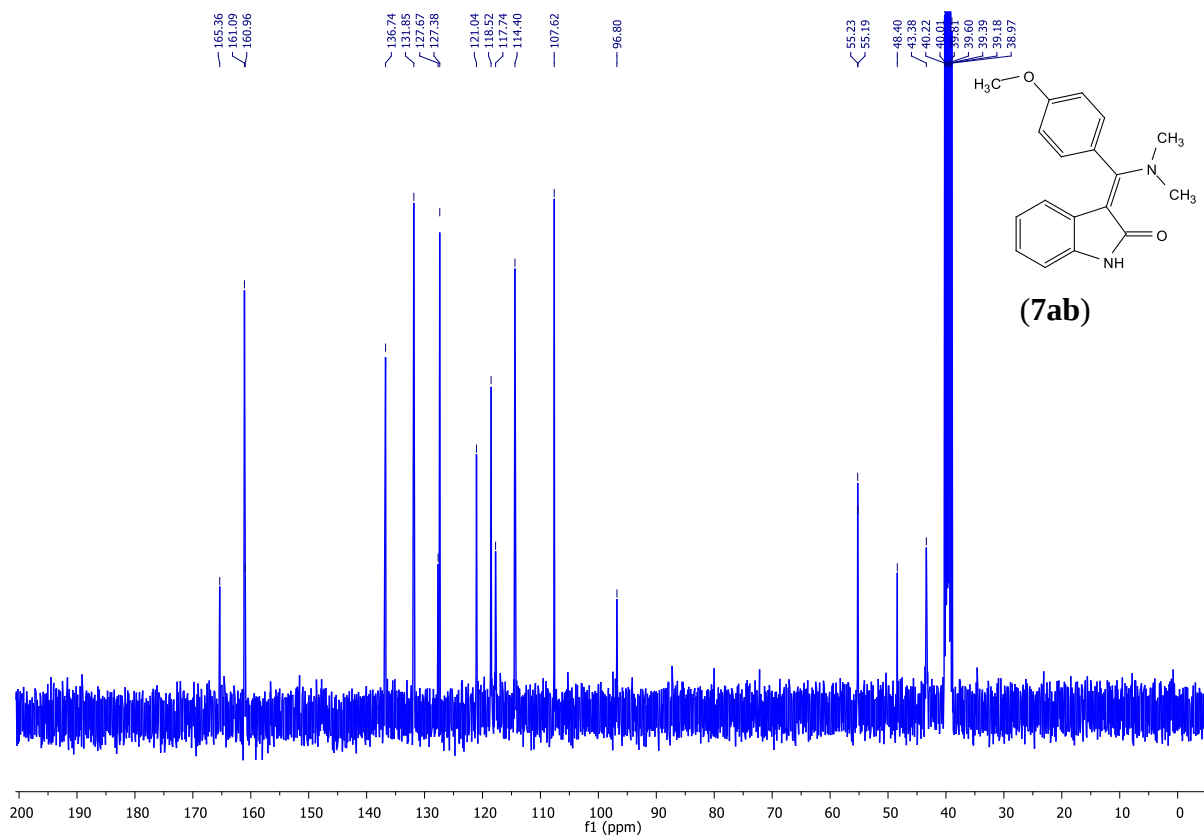
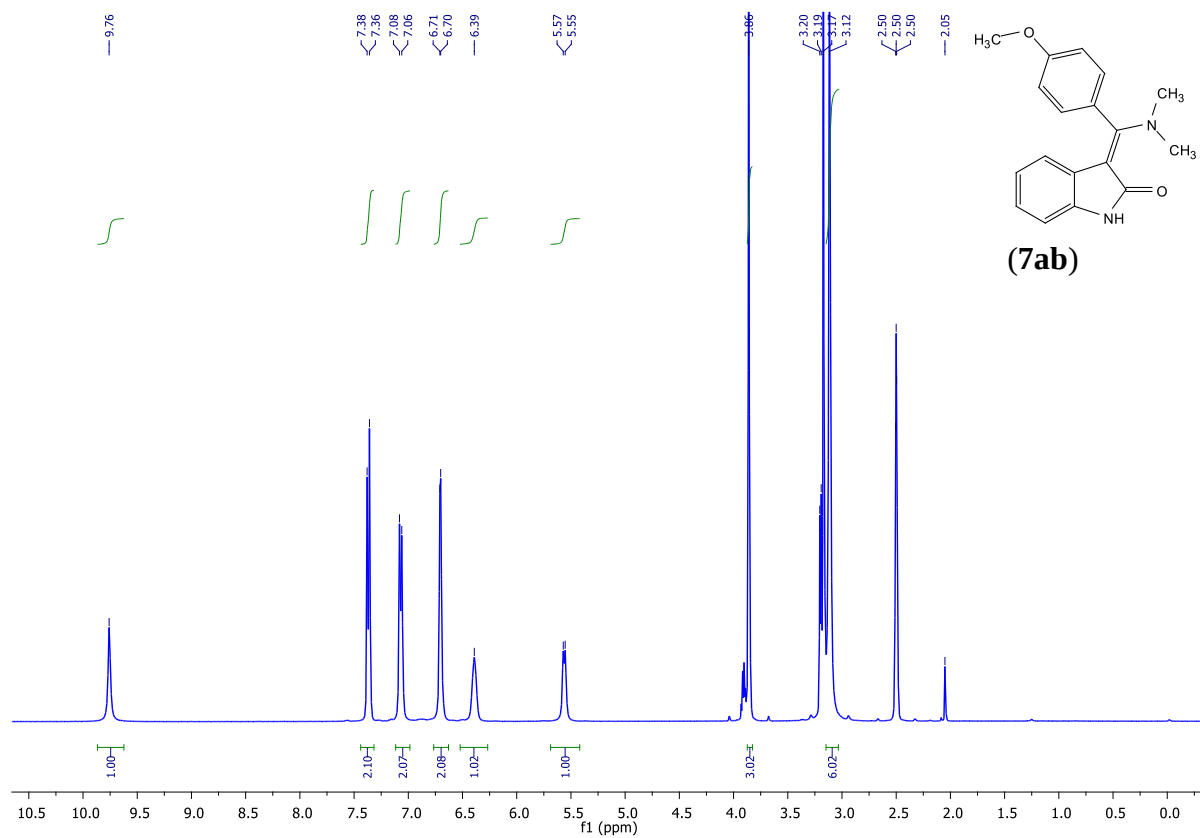


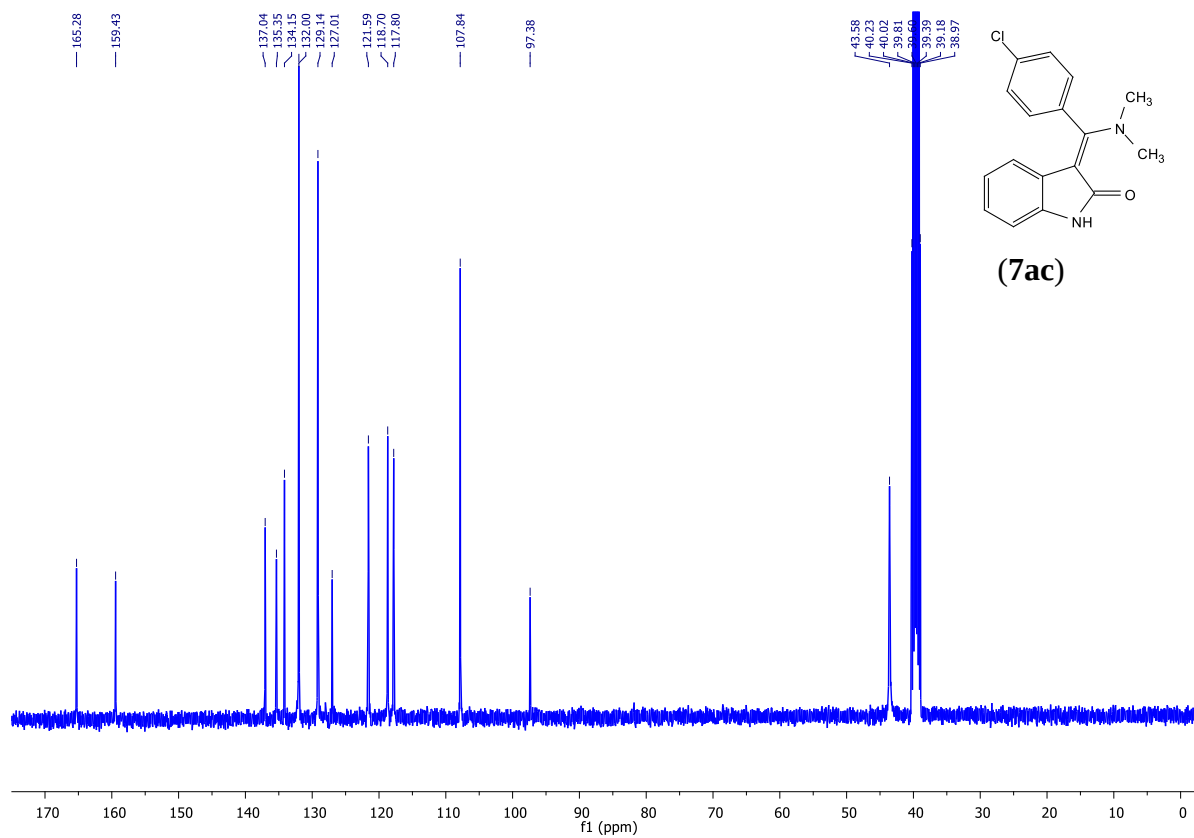
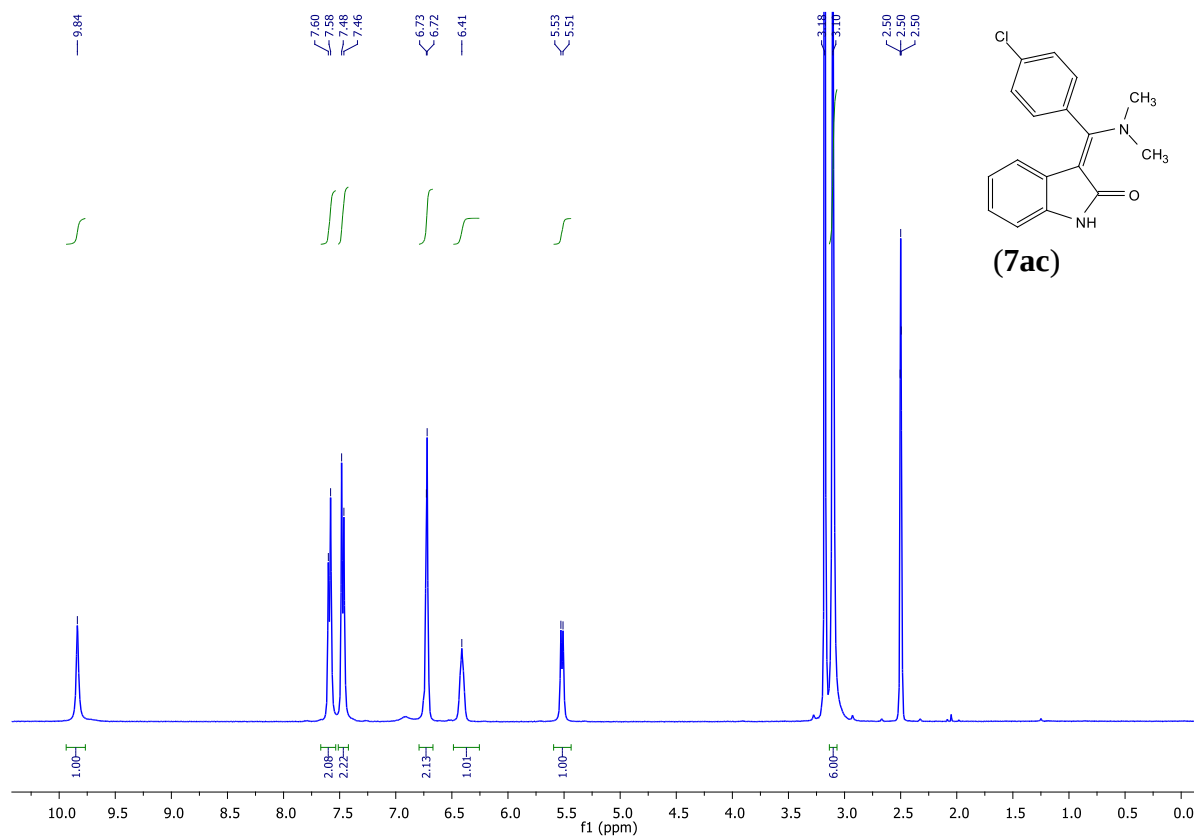




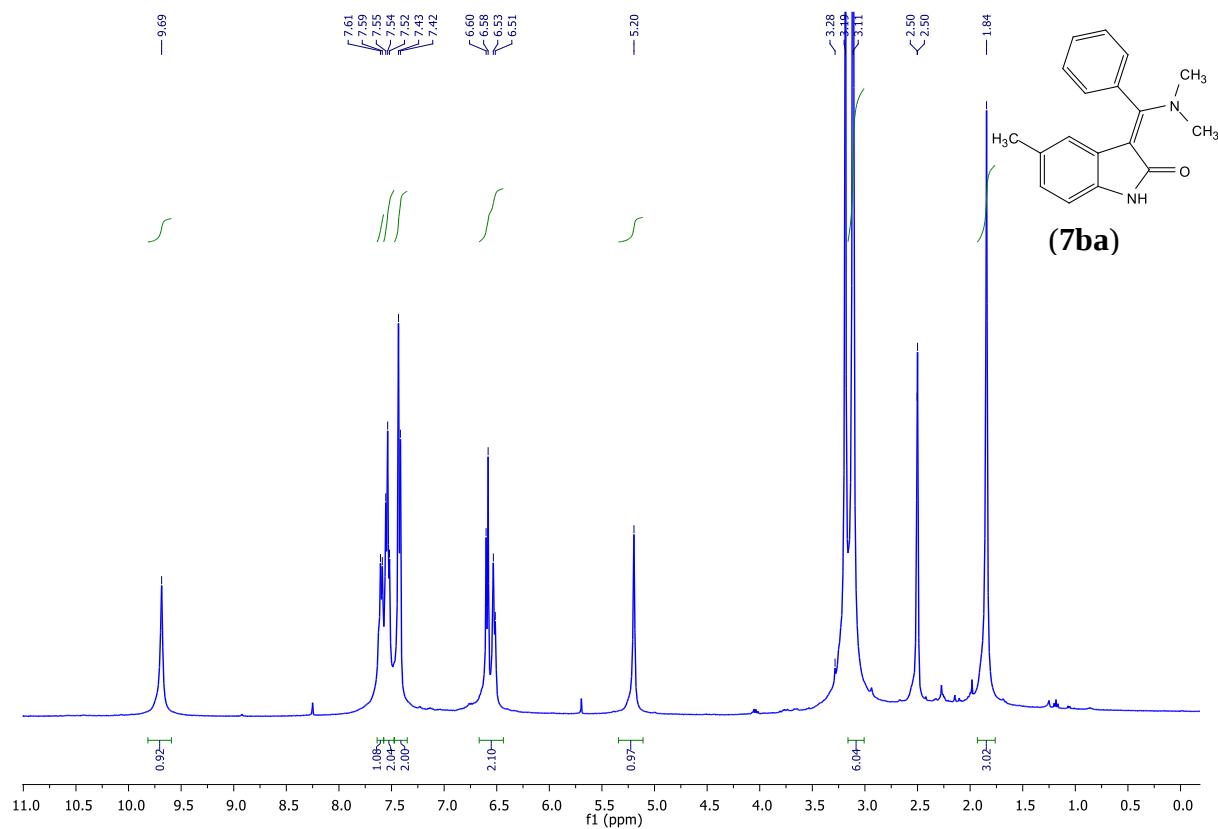




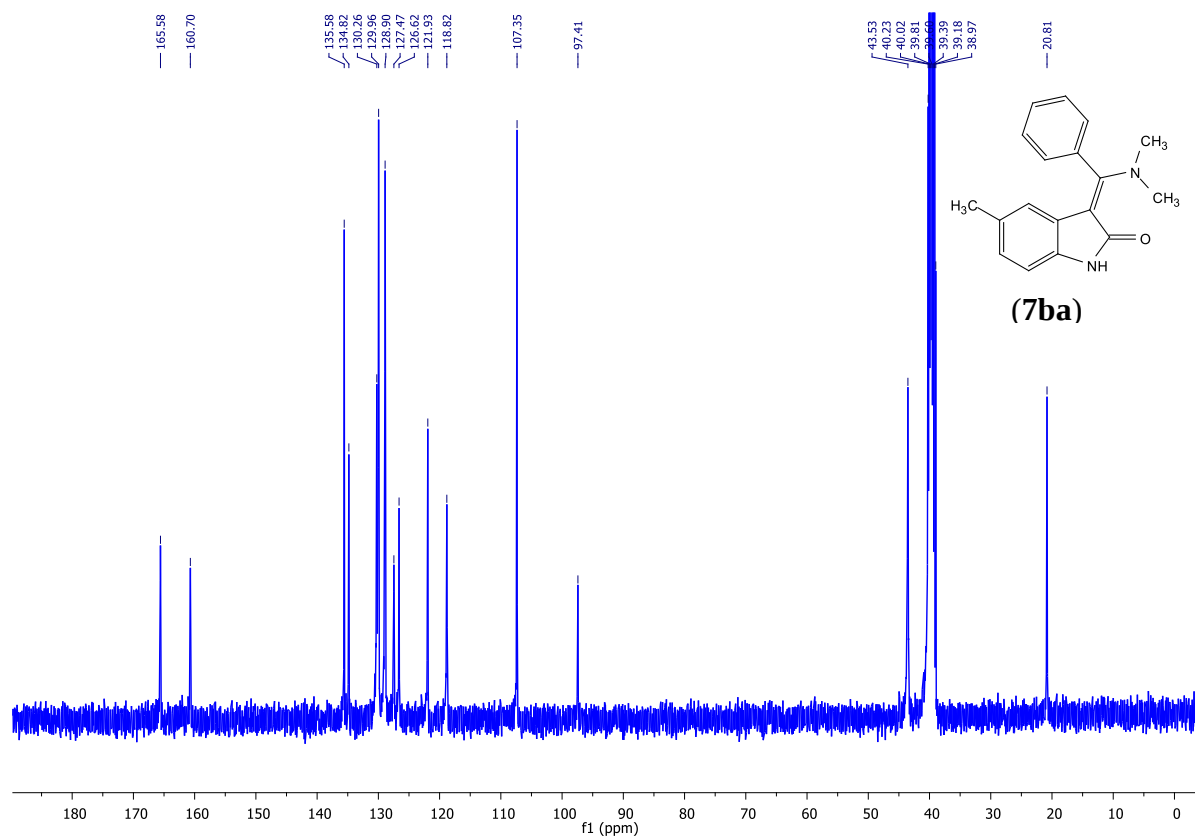




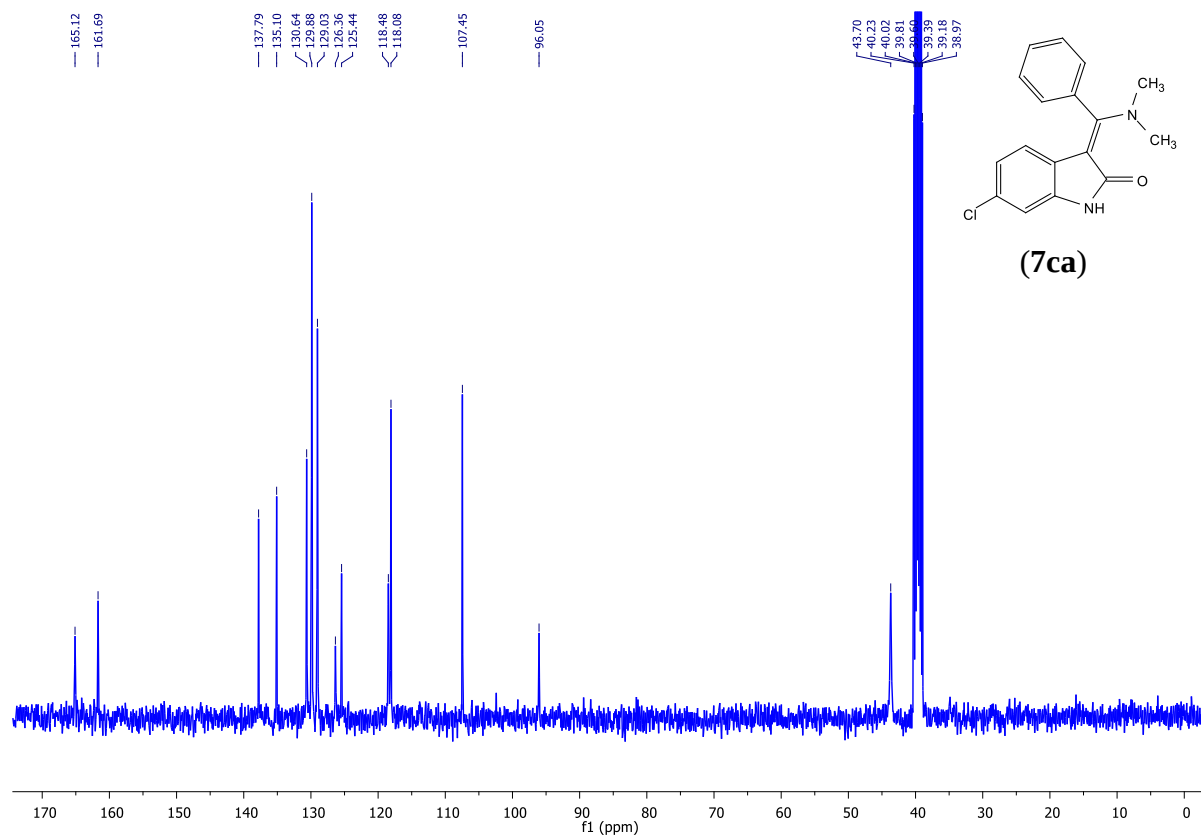
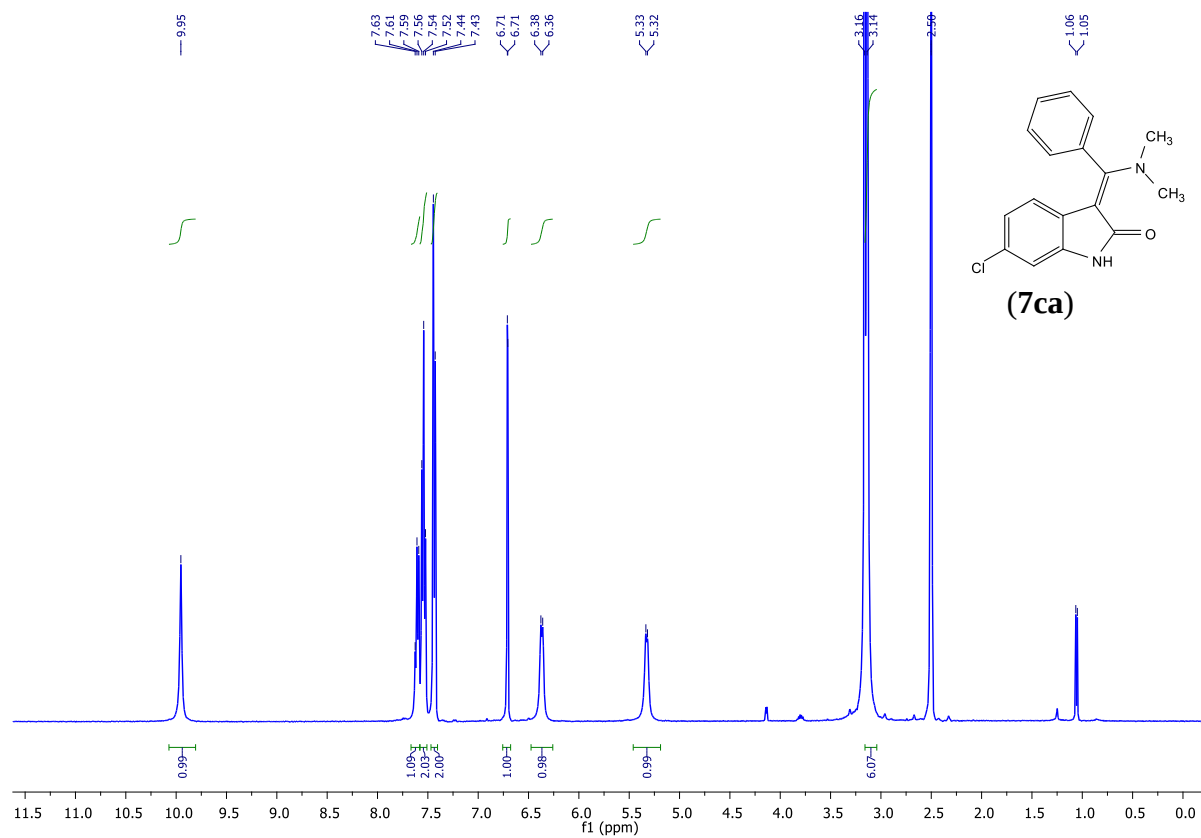
At 60 °C.

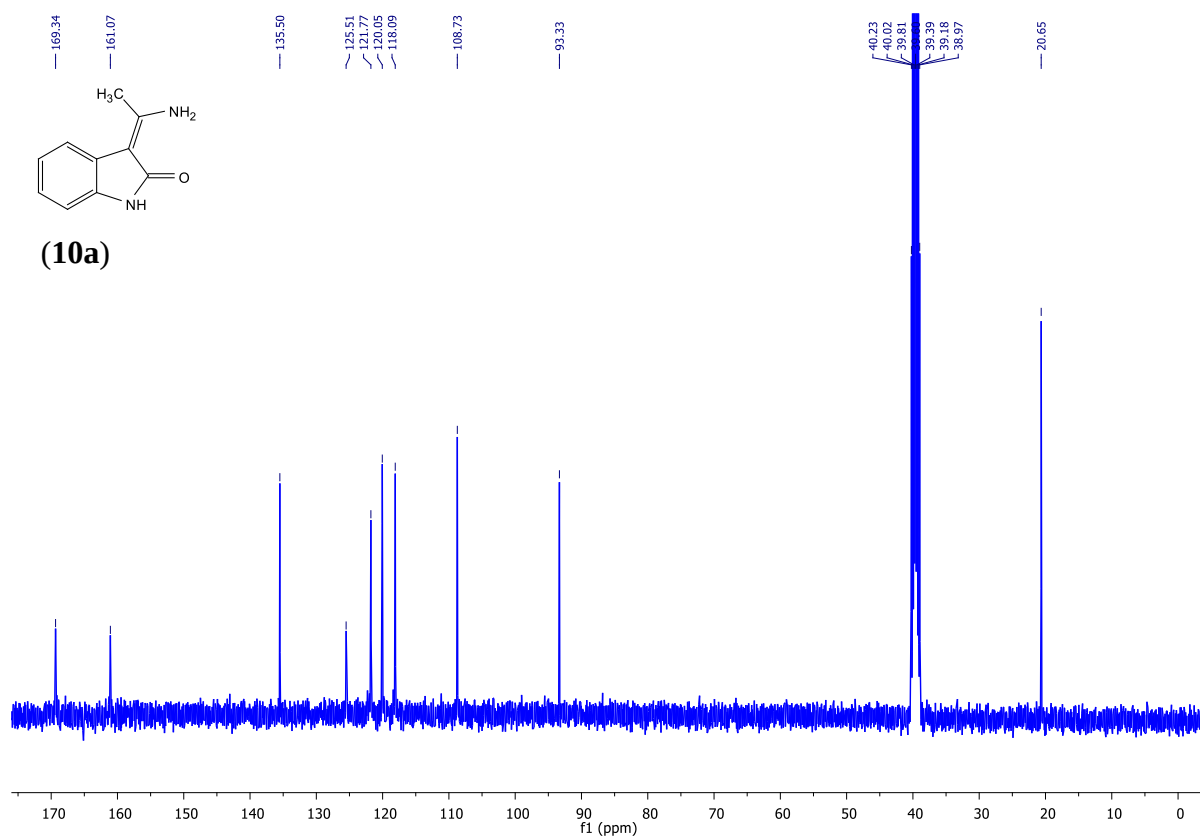
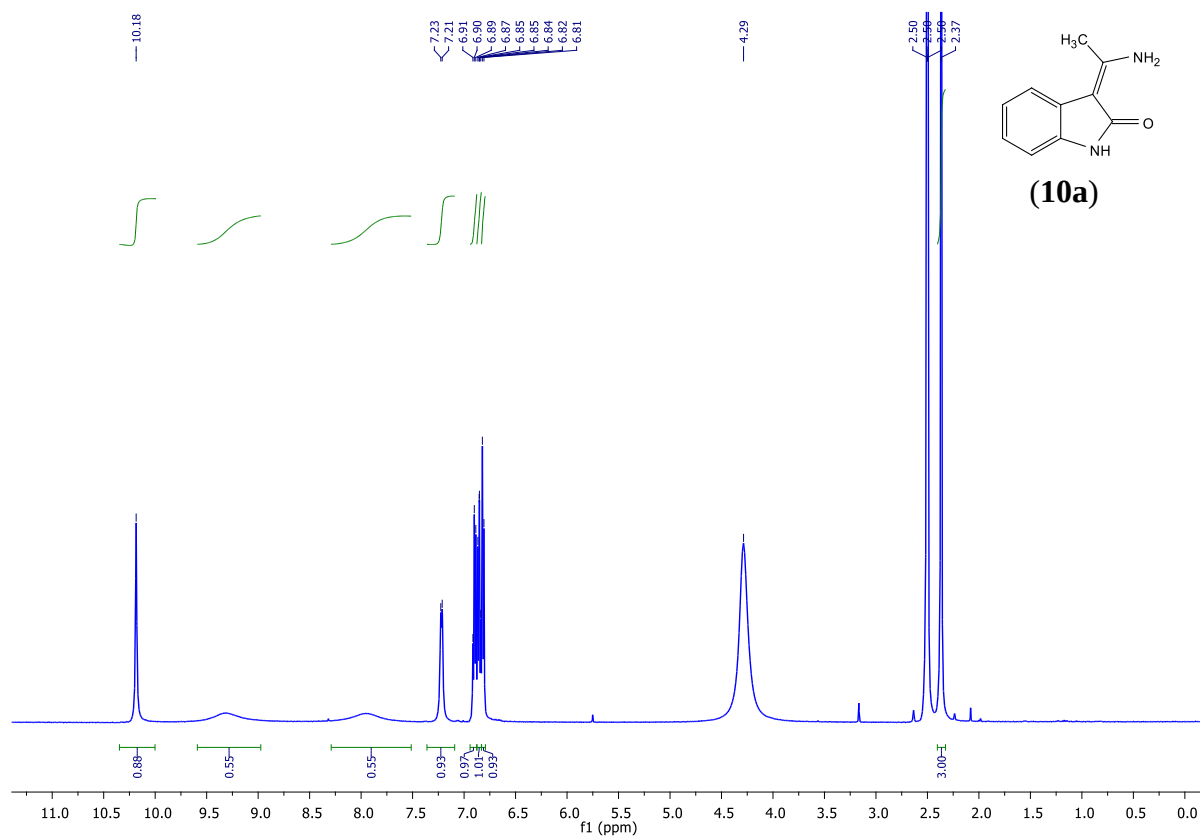


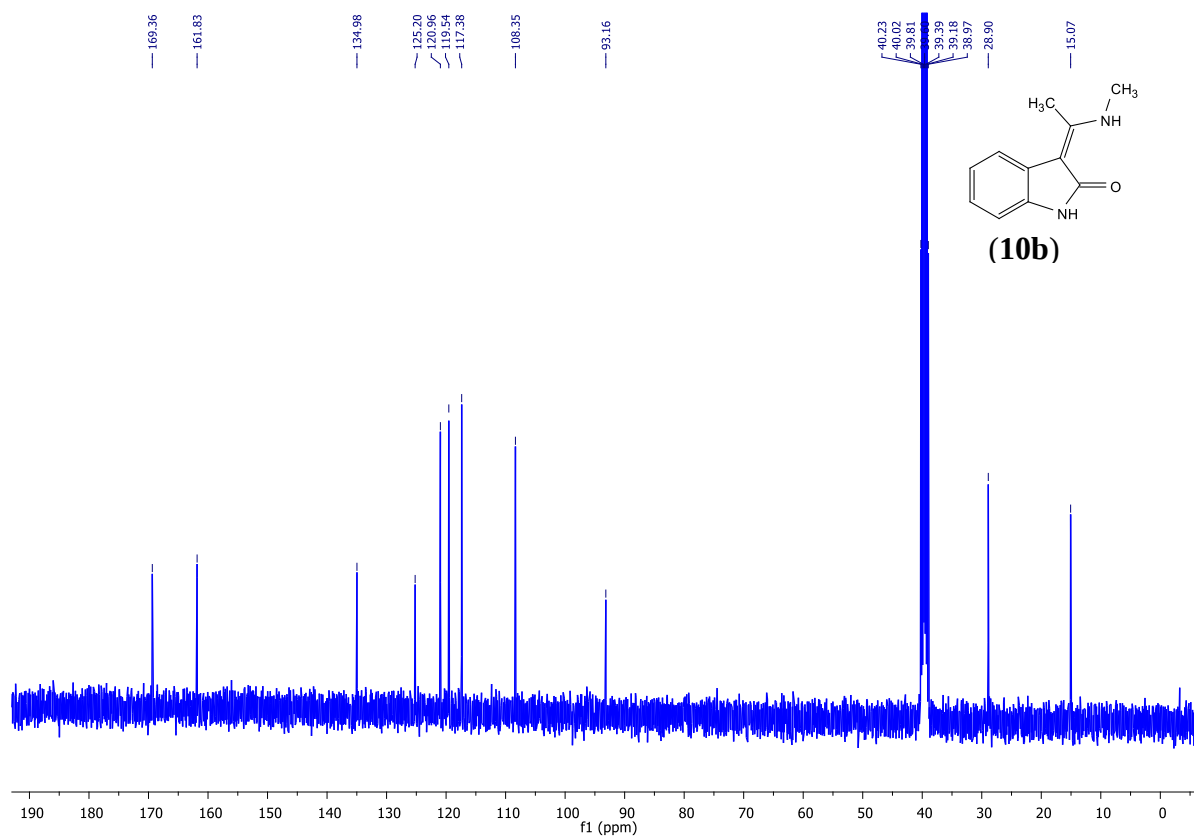
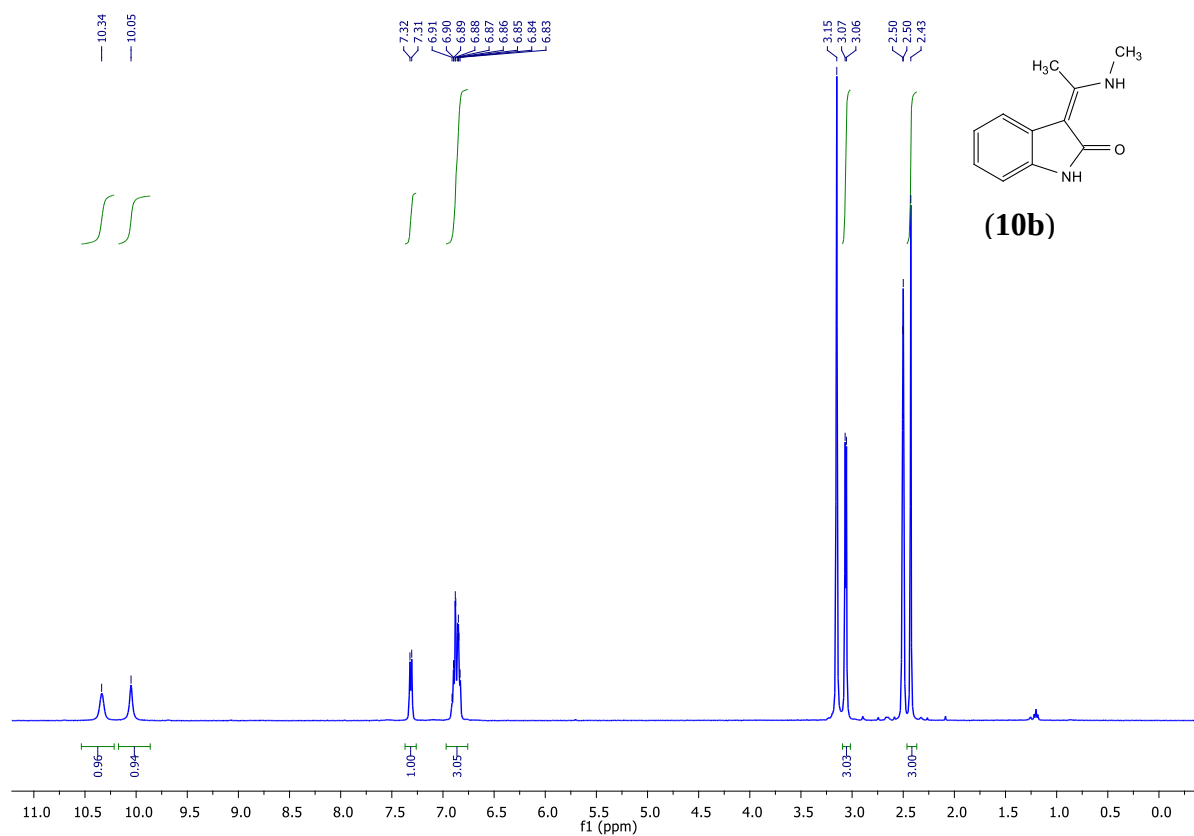
At 60 °C.

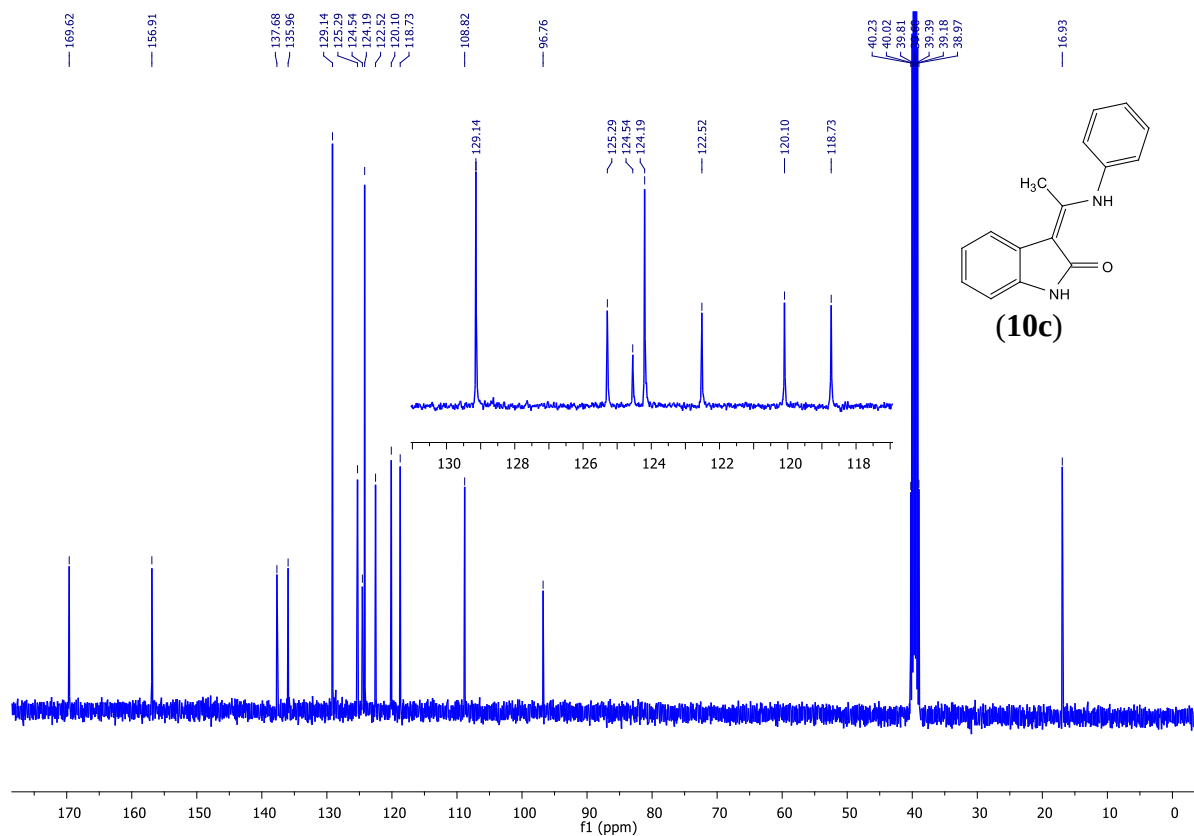
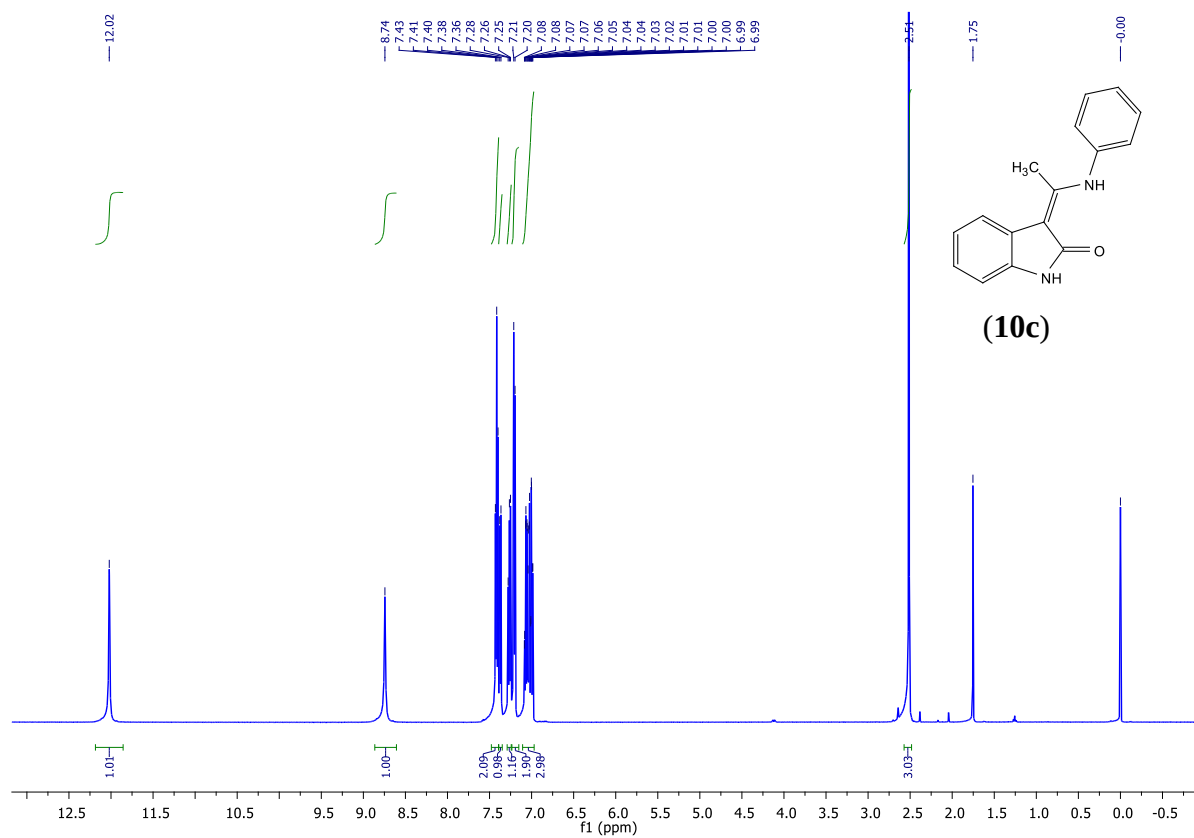


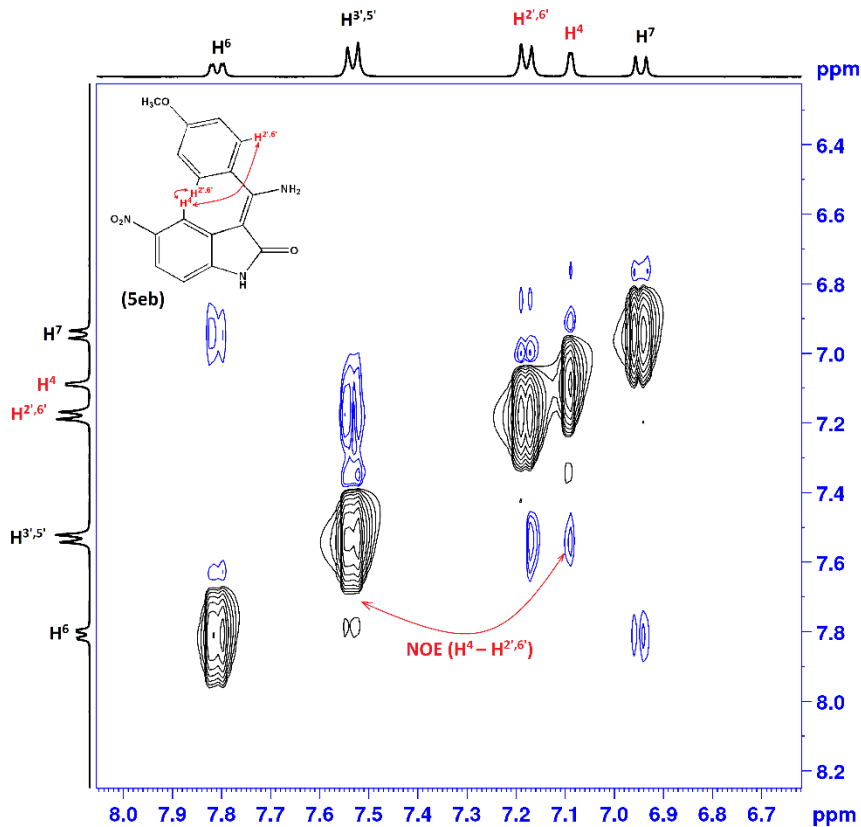
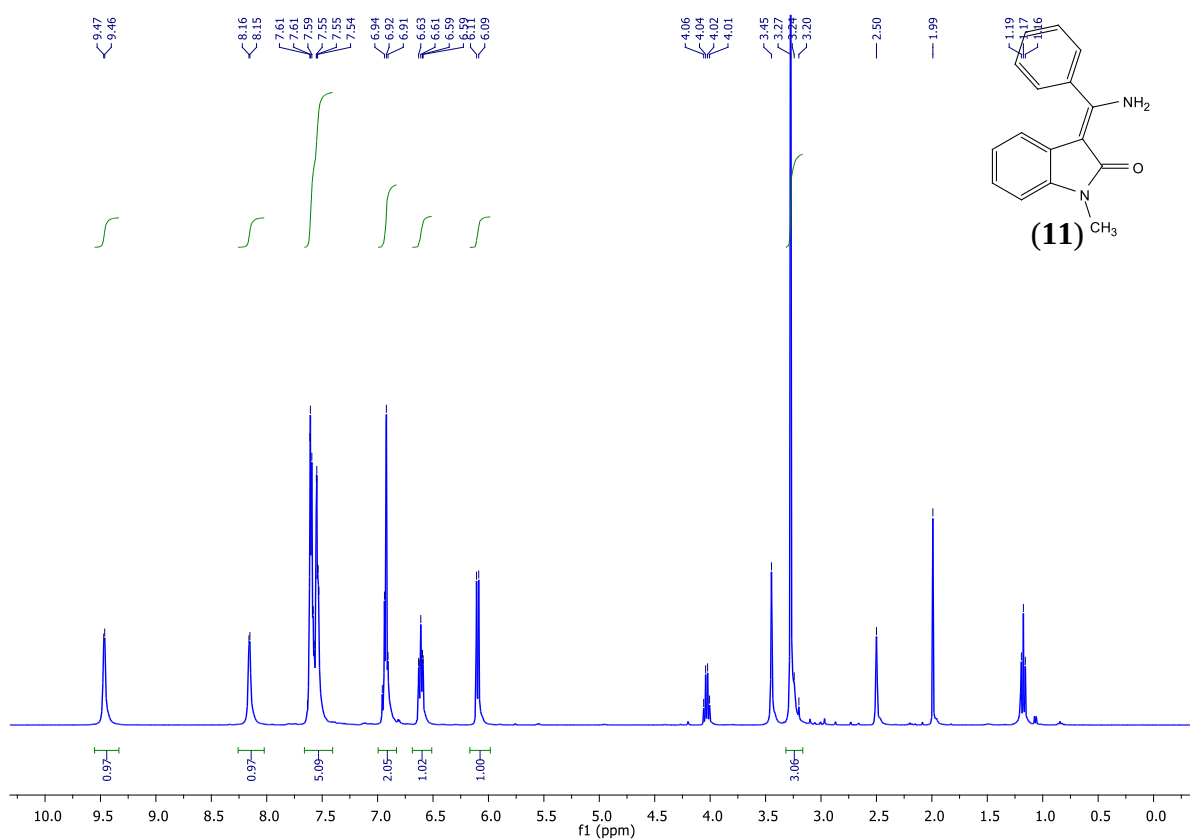
At 60 °C.











Current Data Parameters
NAME servis
EXPNO 6453
PROCNO 1

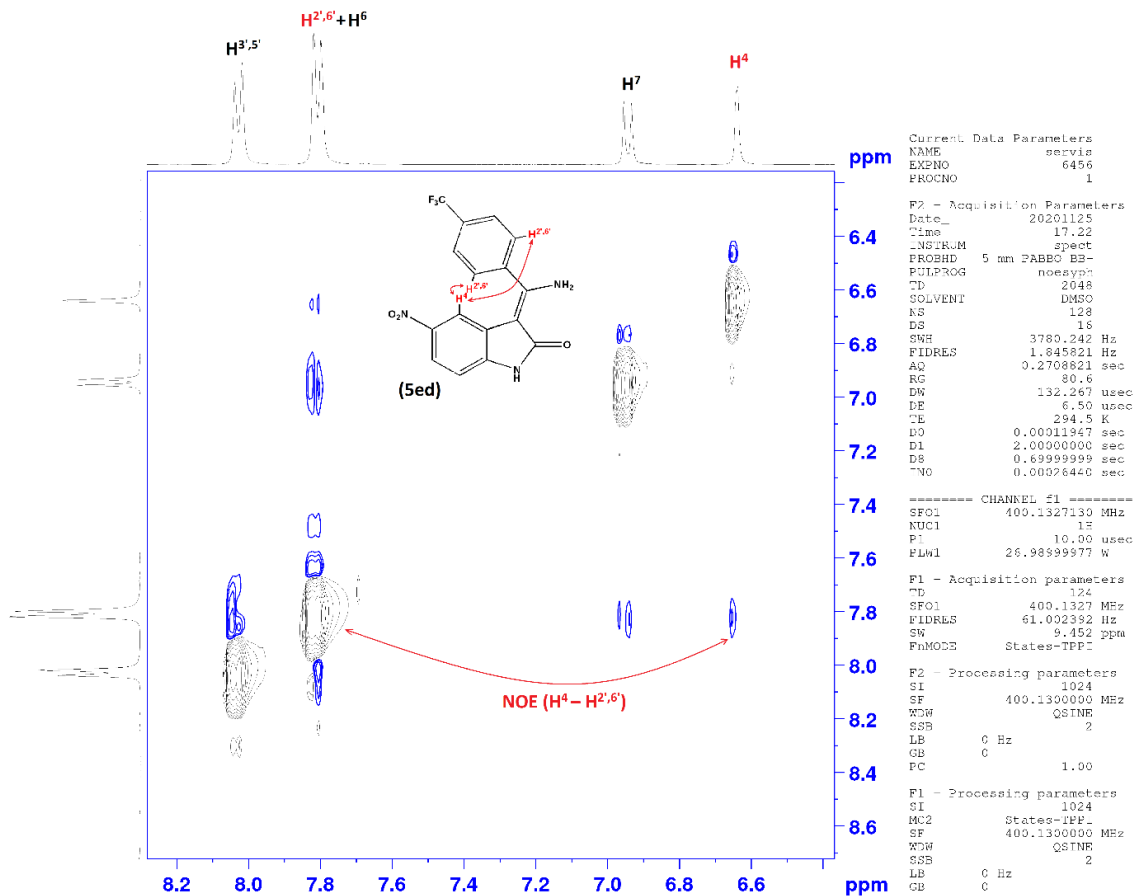
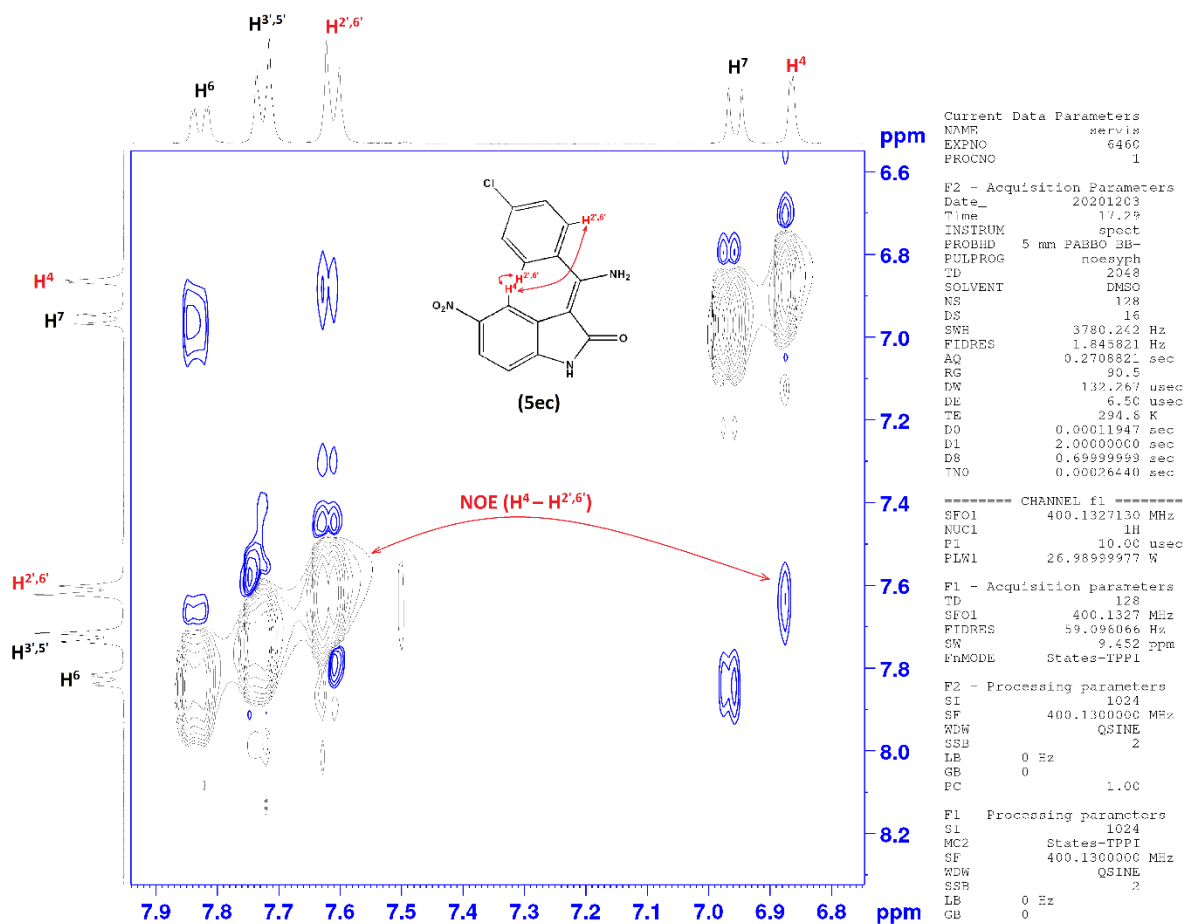
F2 - Acquisition Parameters
Date_ 20200924
Time 16.37
INSTRUM spect
PROBHD 5 mm PABBO BB-
PULPROG noesyph
TD 2048
SOLVENT DMSO
NS 128
DS 16
SWH 3780.242 Hz
FIDRES 1.845821 Hz
AQ 0.2708821 sec
RG 64
DE 132.267 usec
TE 297.9 K
DO 0.00011947 sec
D1 2.0000000 sec
D8 0.69999999 sec
INO 0.00026440 sec

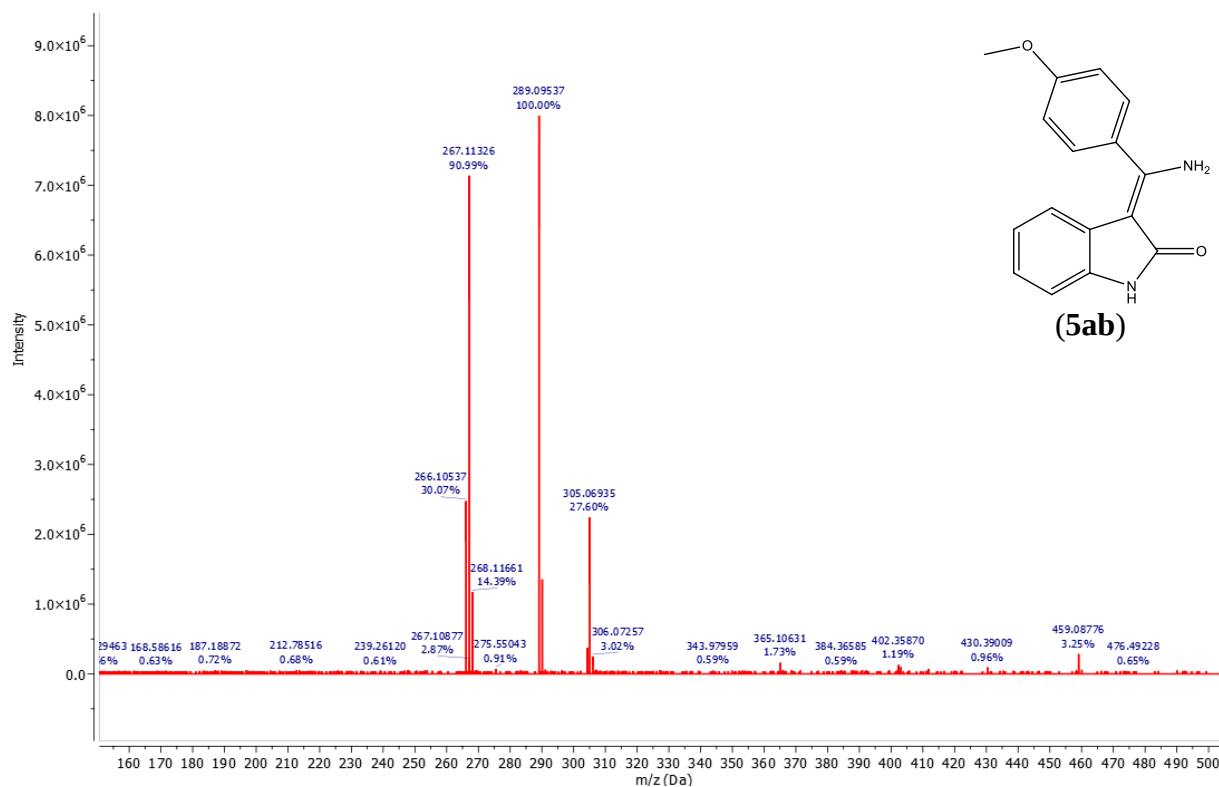
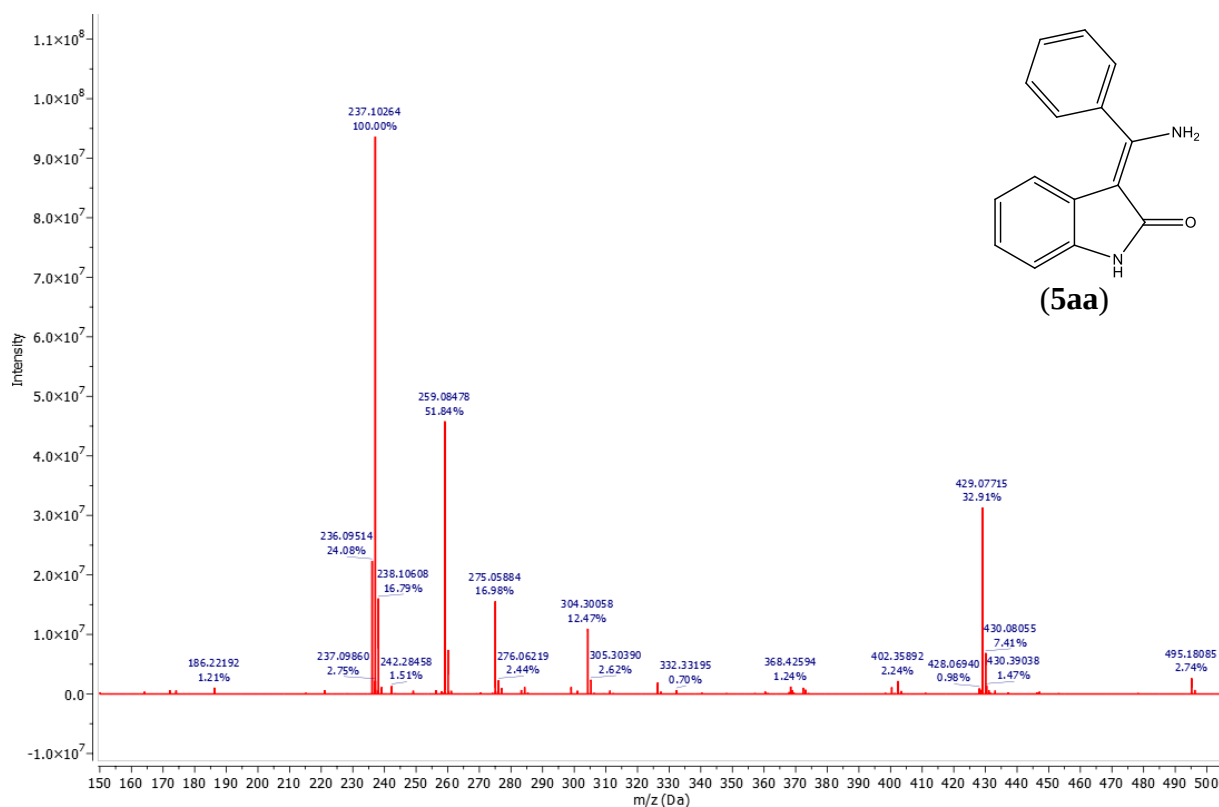
===== CHANNEL f1 =====
SF01 400.1327130 MHz
NUC1 1H
P1 10.00 usec
PL1 26.98999977 W

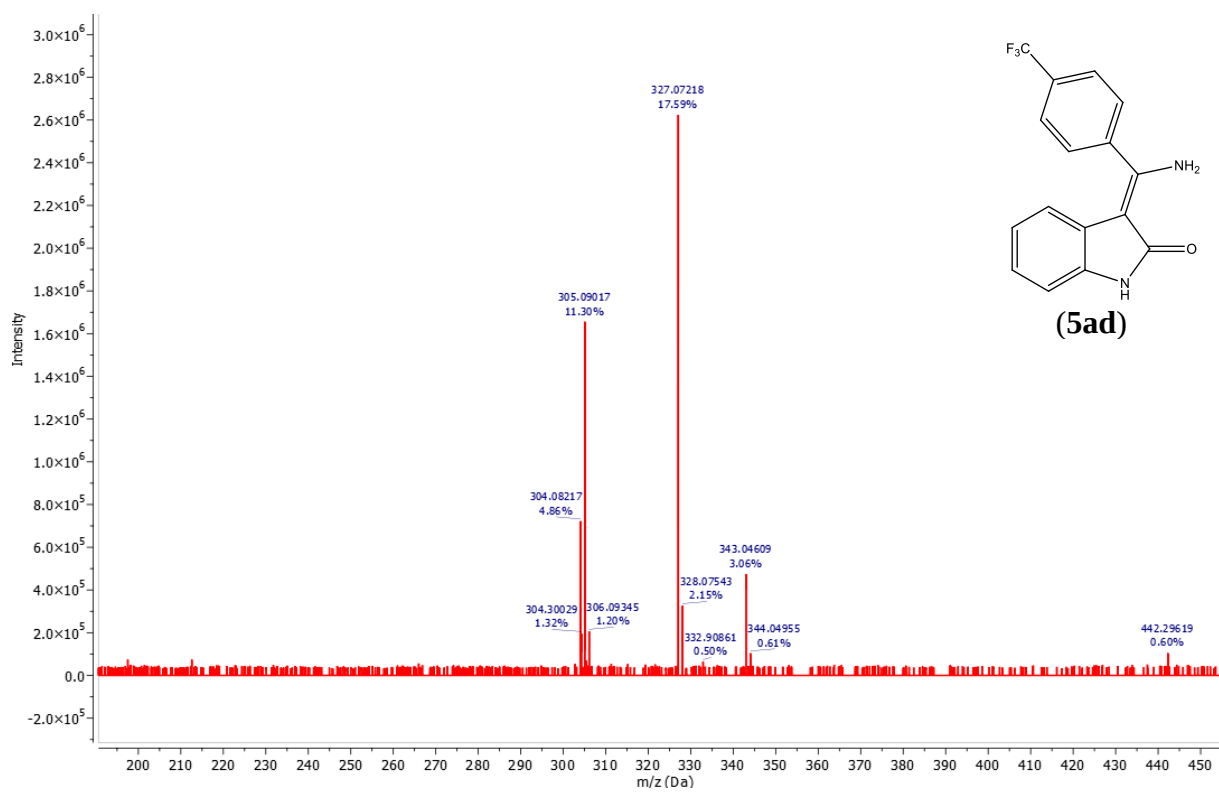
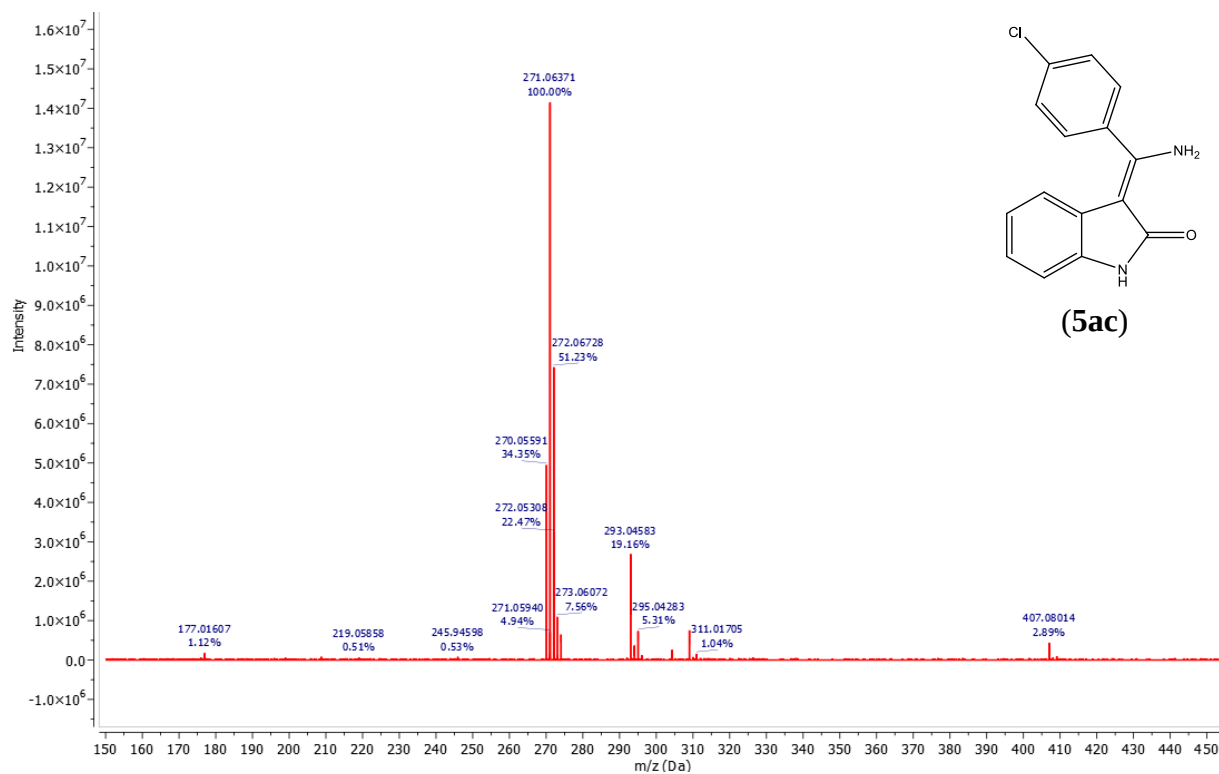
F1 - Acquisition parameters
TD 128
SF01 400.1327 MHz
FIDRES 59.096066 Hz
SW 9.452 ppm
FnMODE States-TPP1

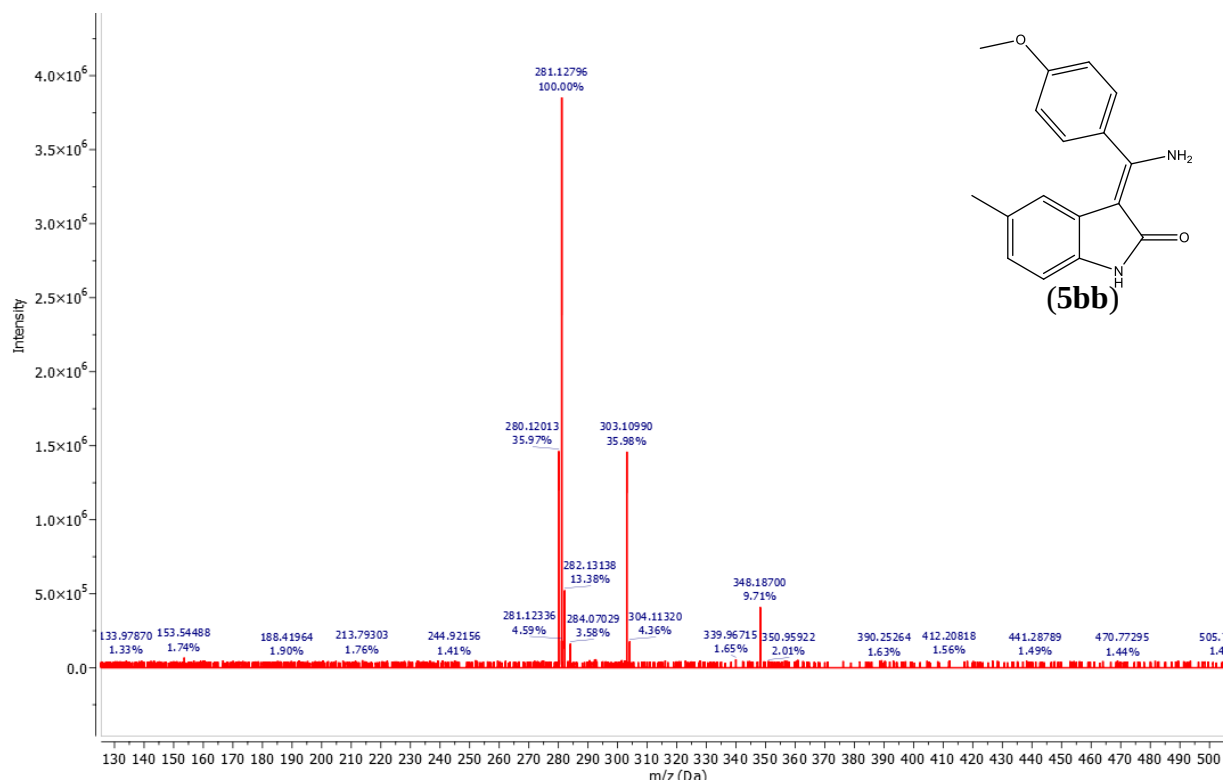
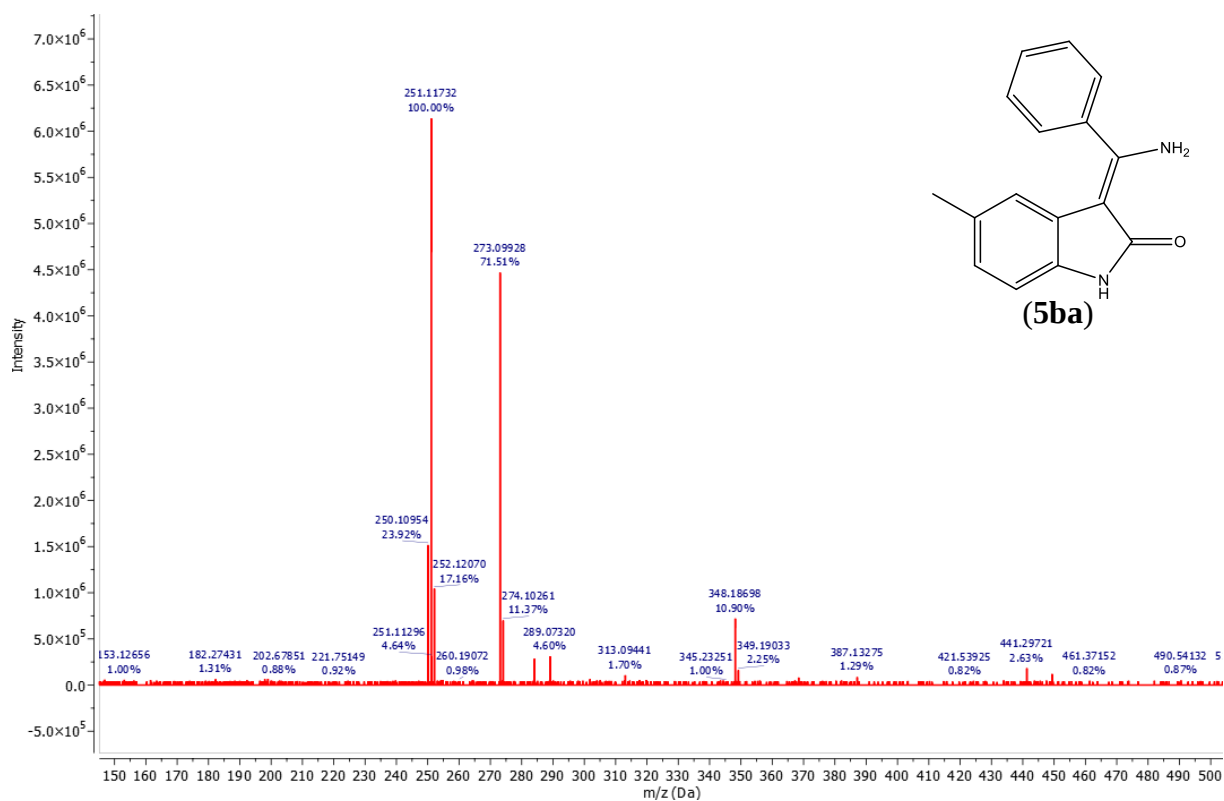
F2 - Processing parameters
SI 1024
SF 400.130033 MHz
WDW QSINE
SSB 2
LB 0 Hz
GB 0
PC 1.00

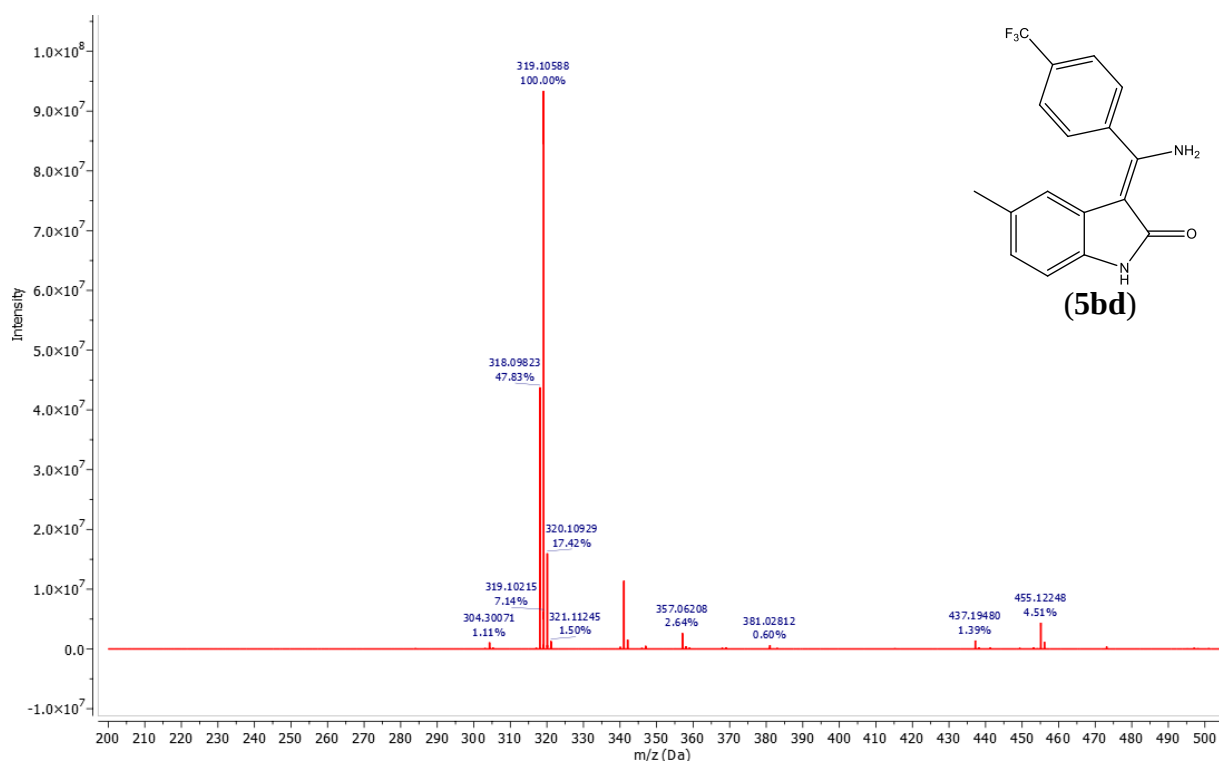
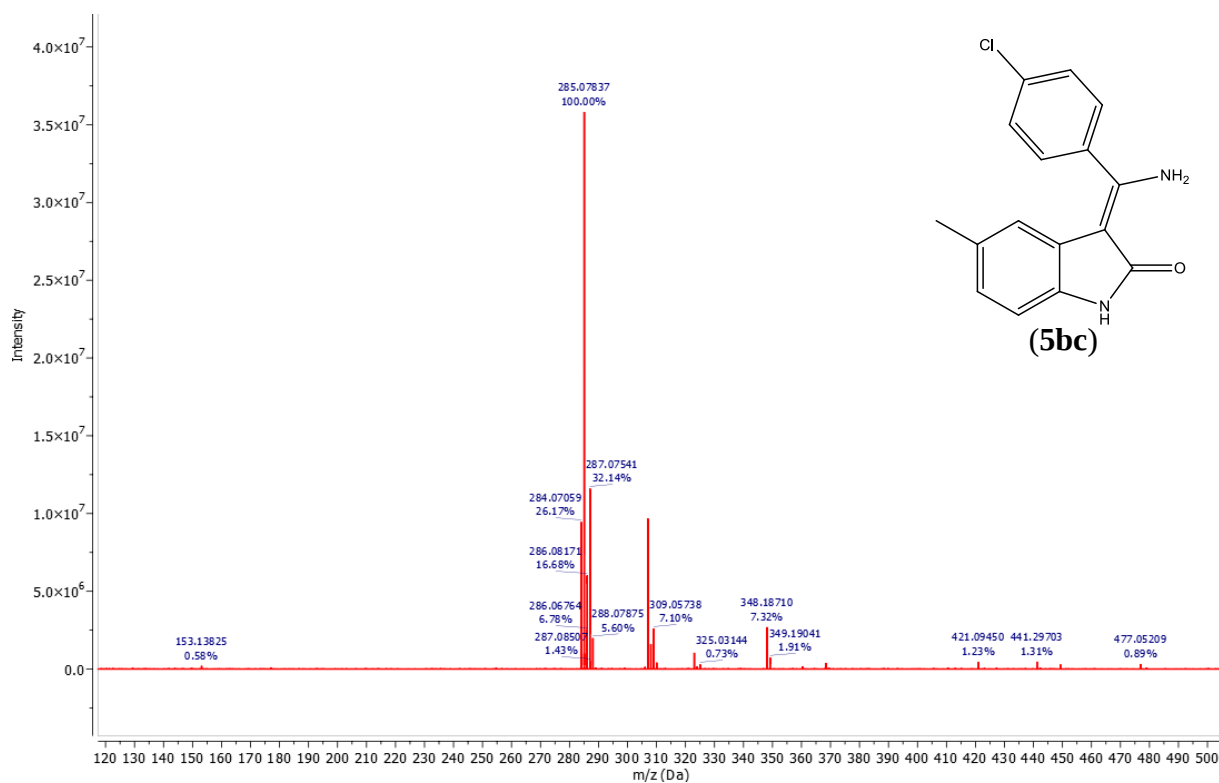
F1 - Processing parameters
SI 1024
MC2 States-TPPI
SF 400.130033 MHz
WDW QSINE
SSB 2
LB 0 Hz
GB 0

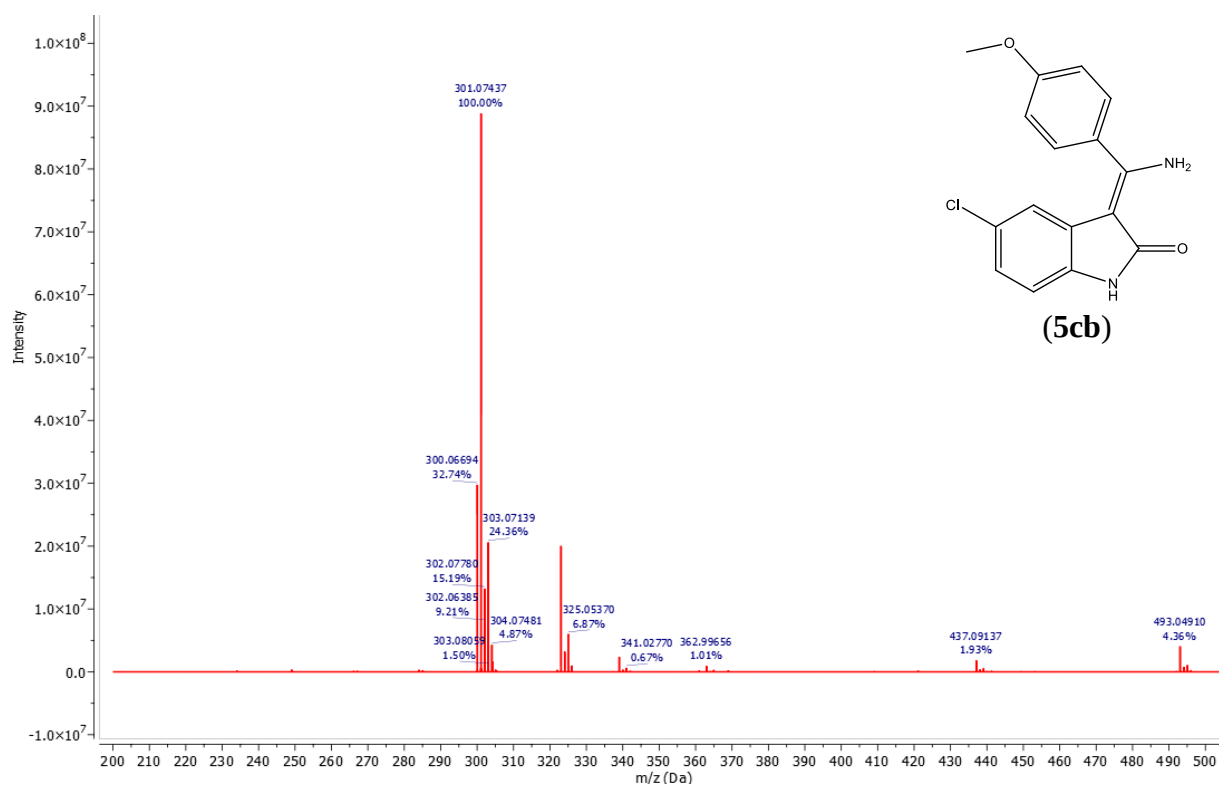
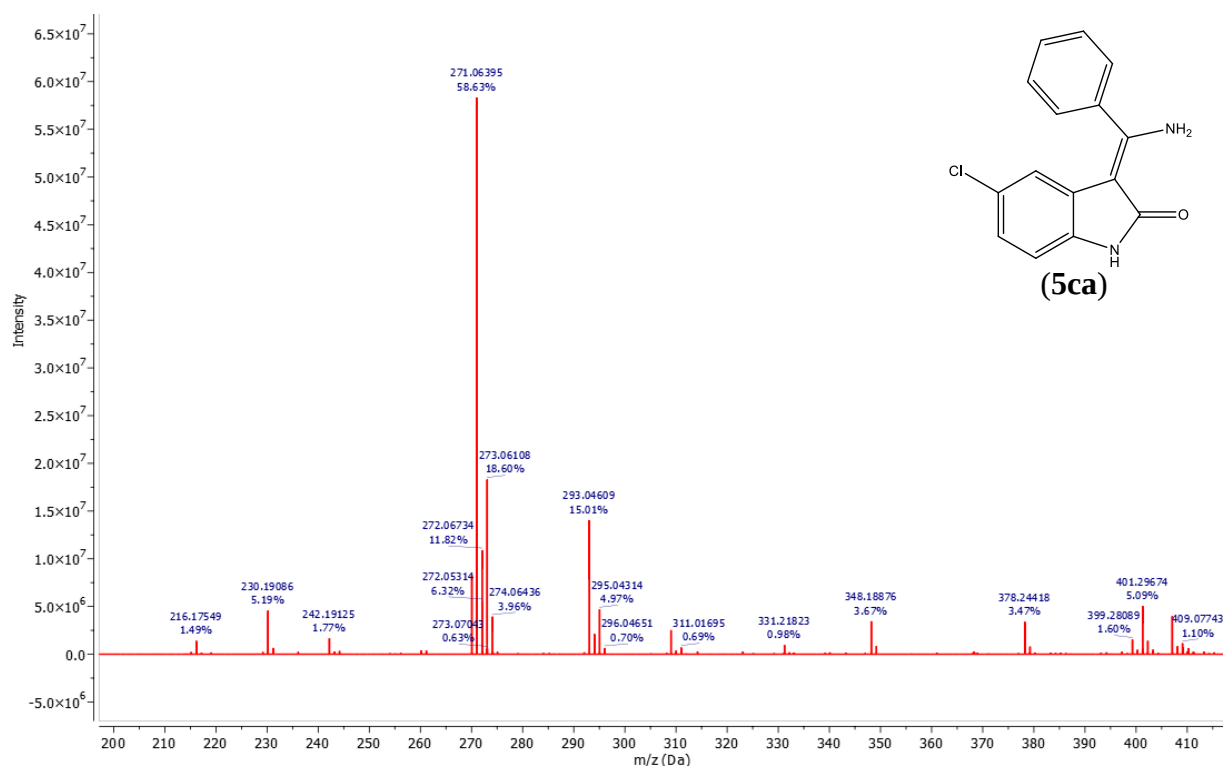


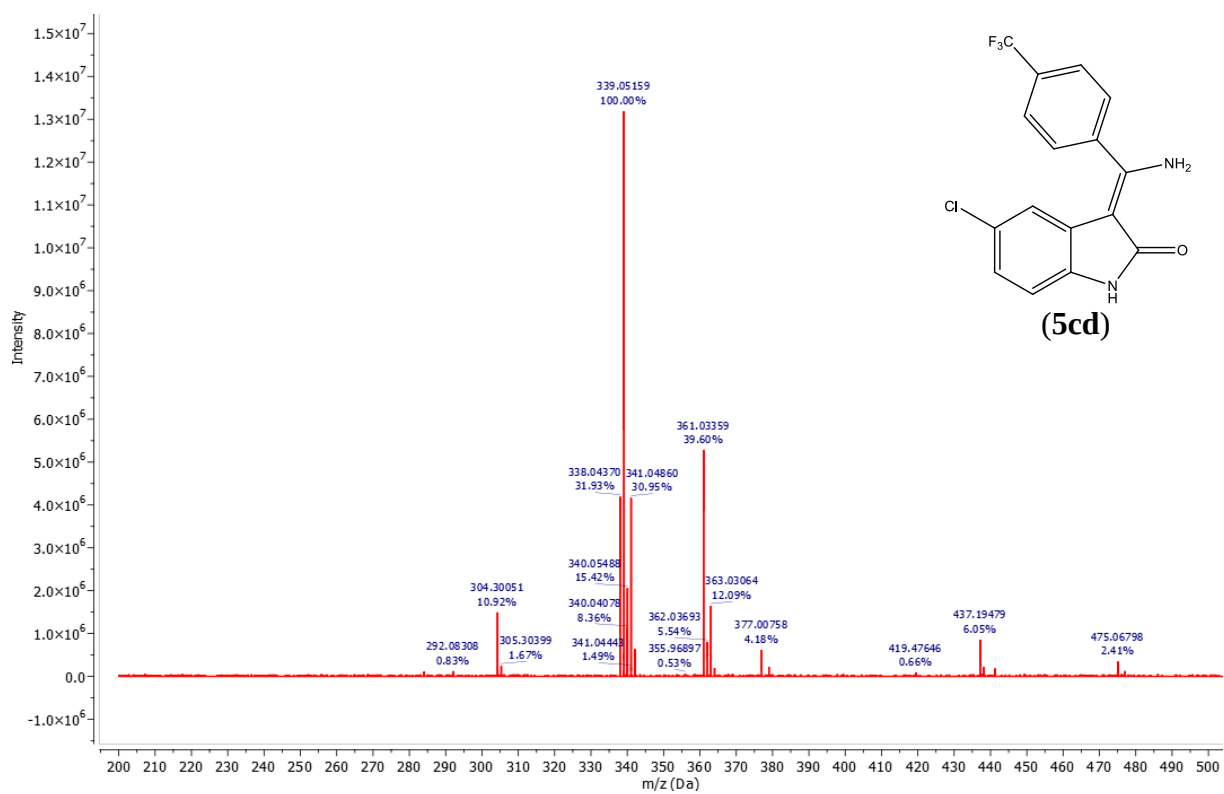
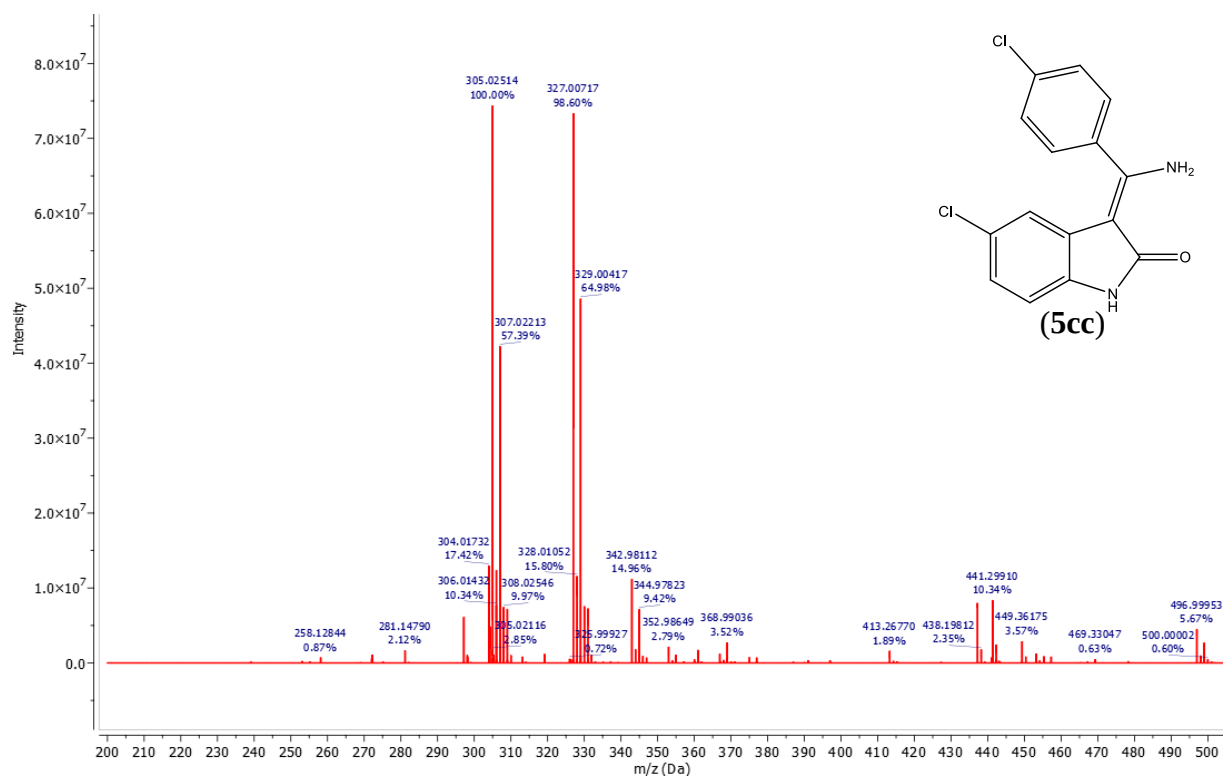


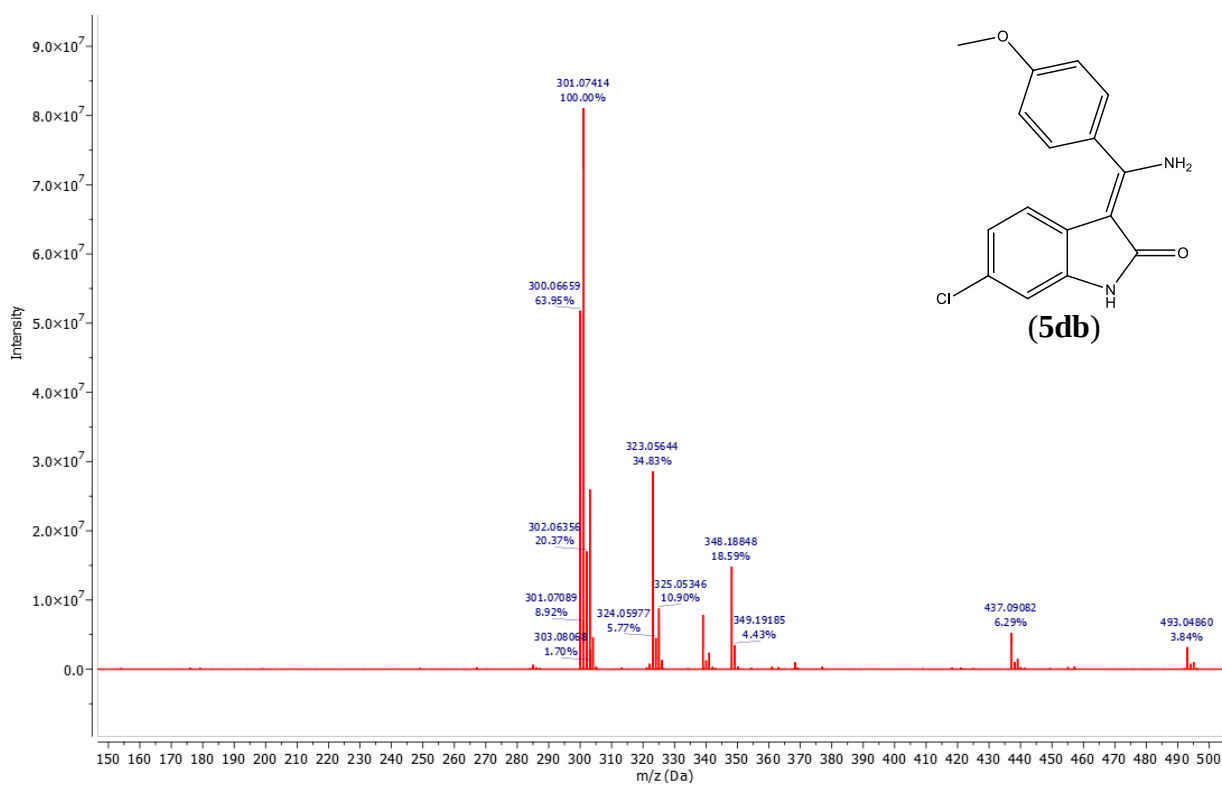
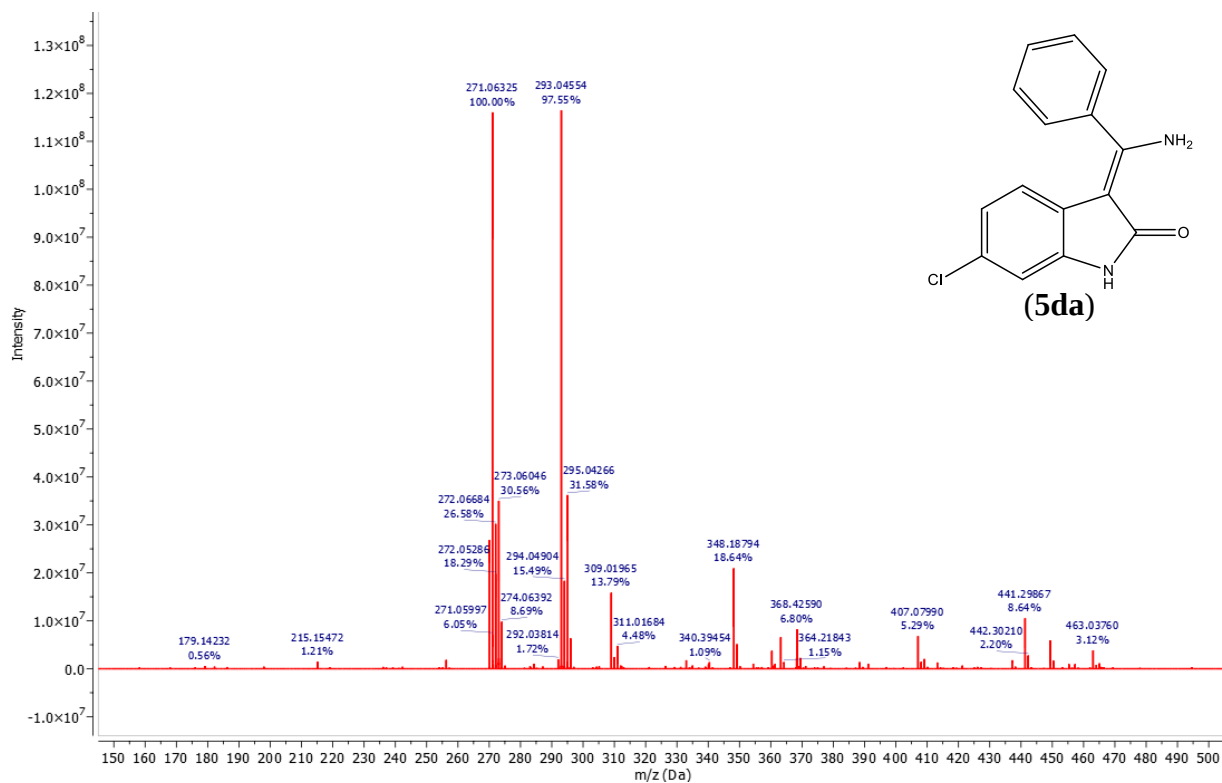


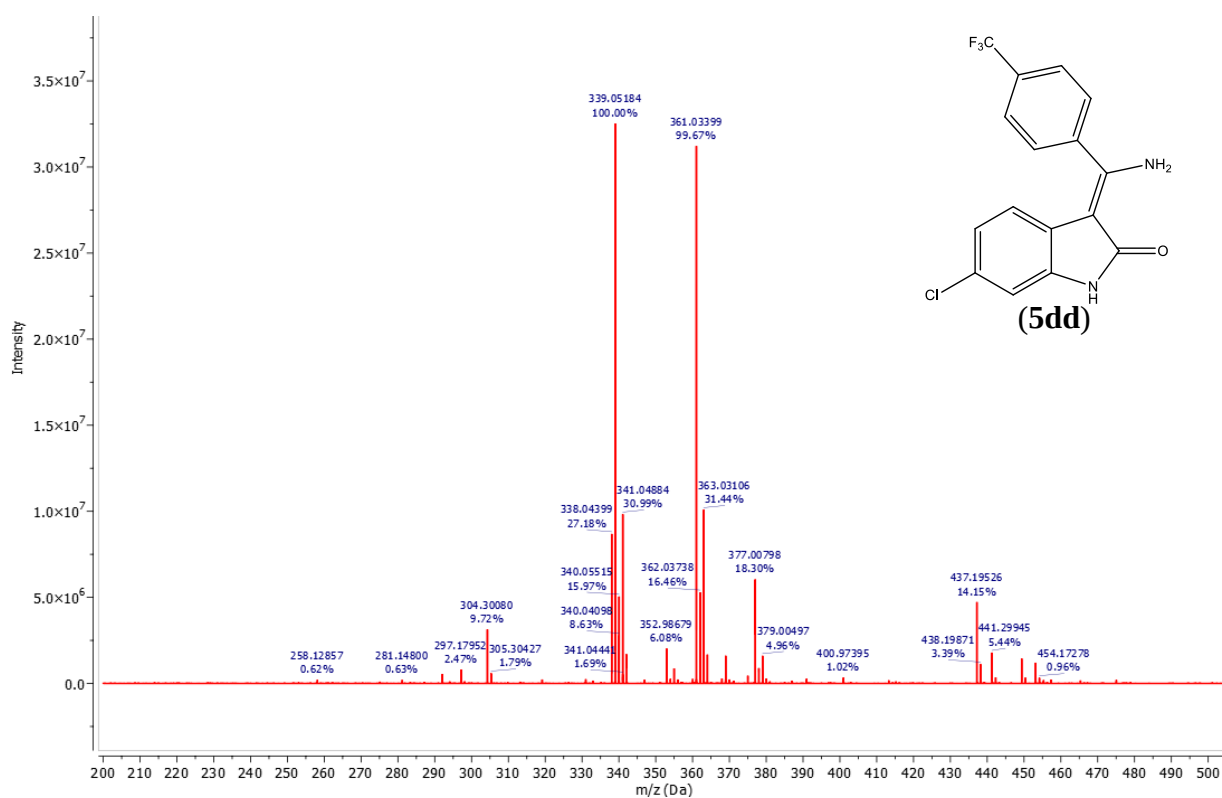
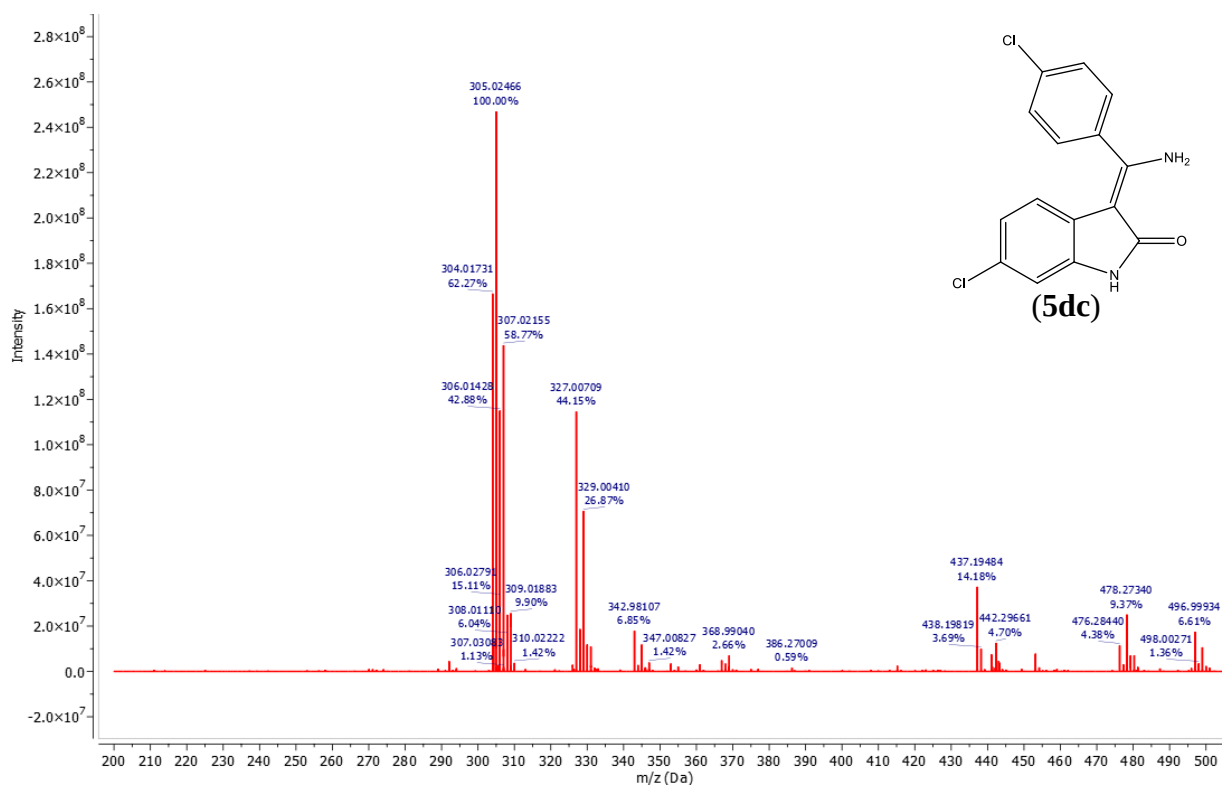


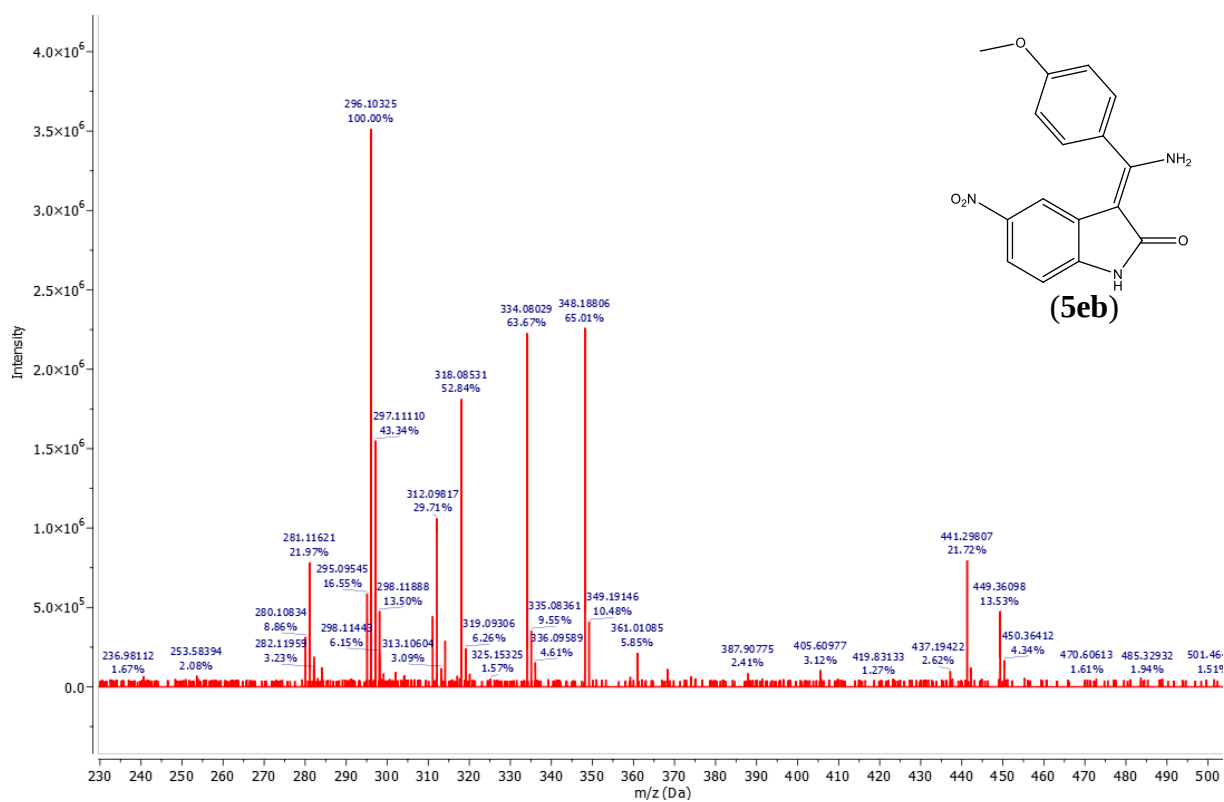
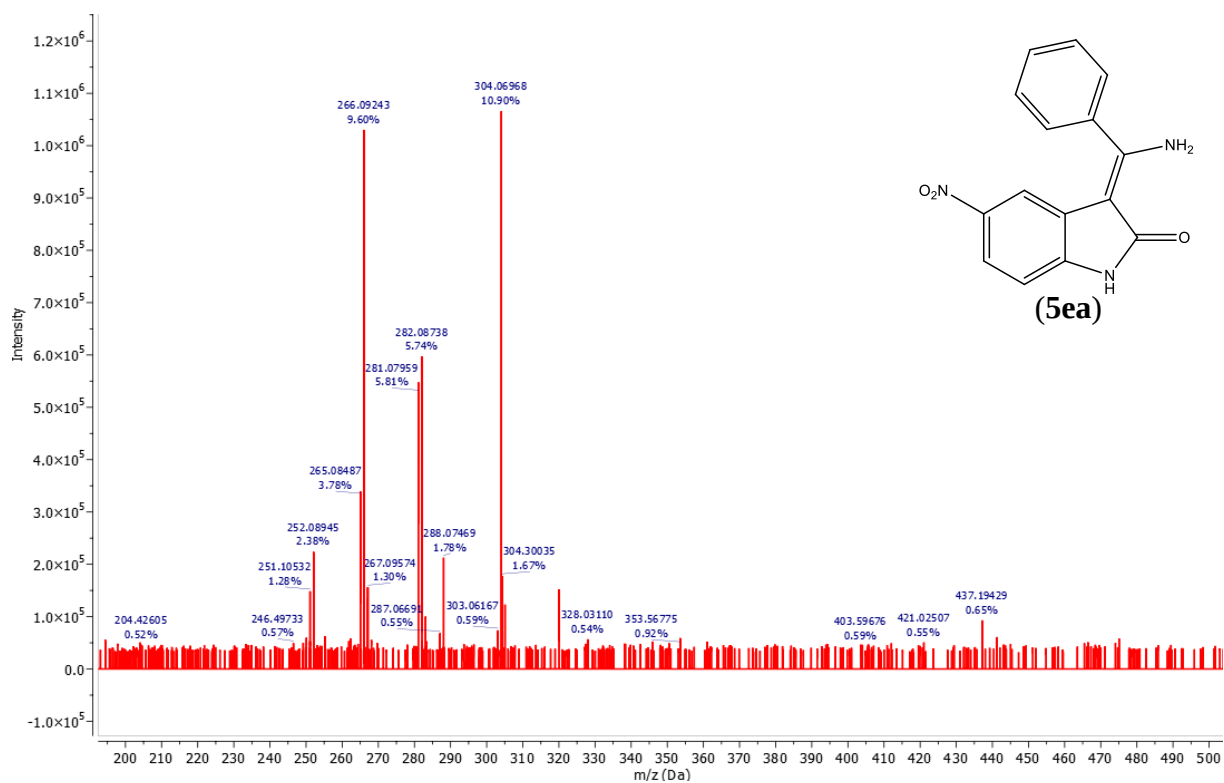


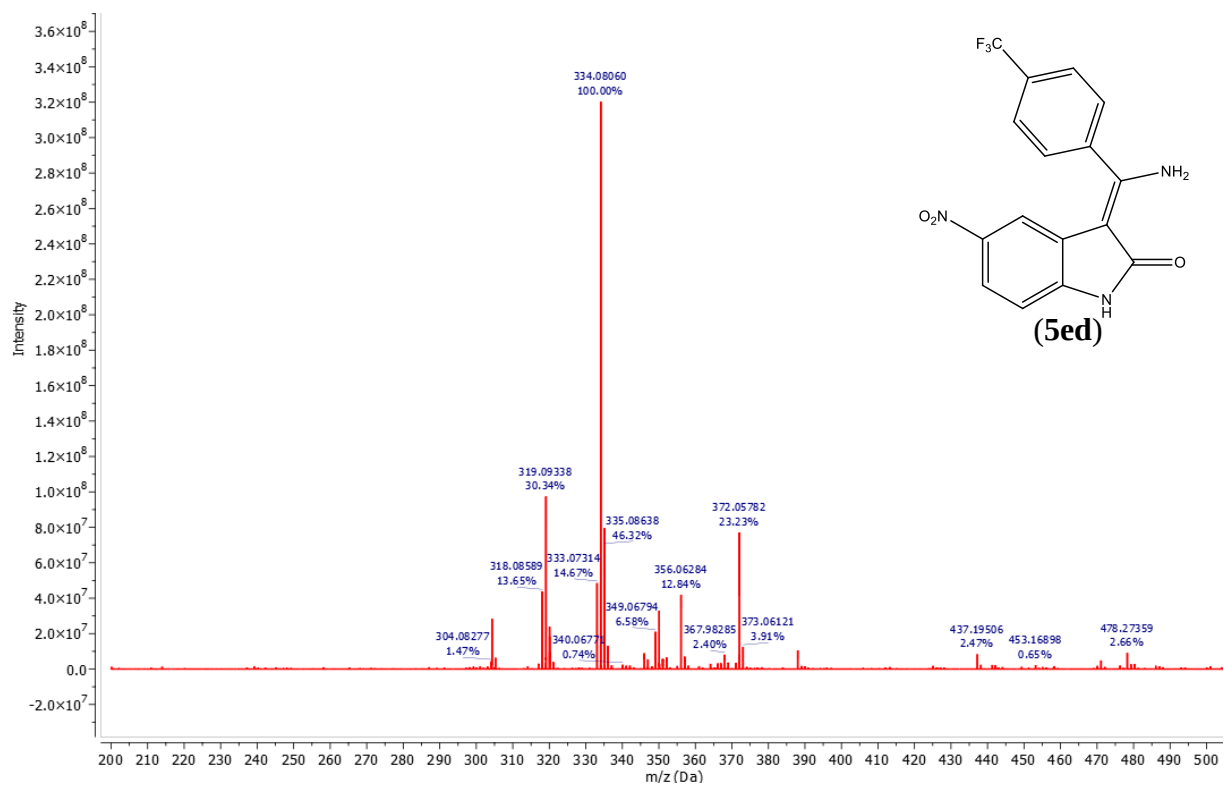
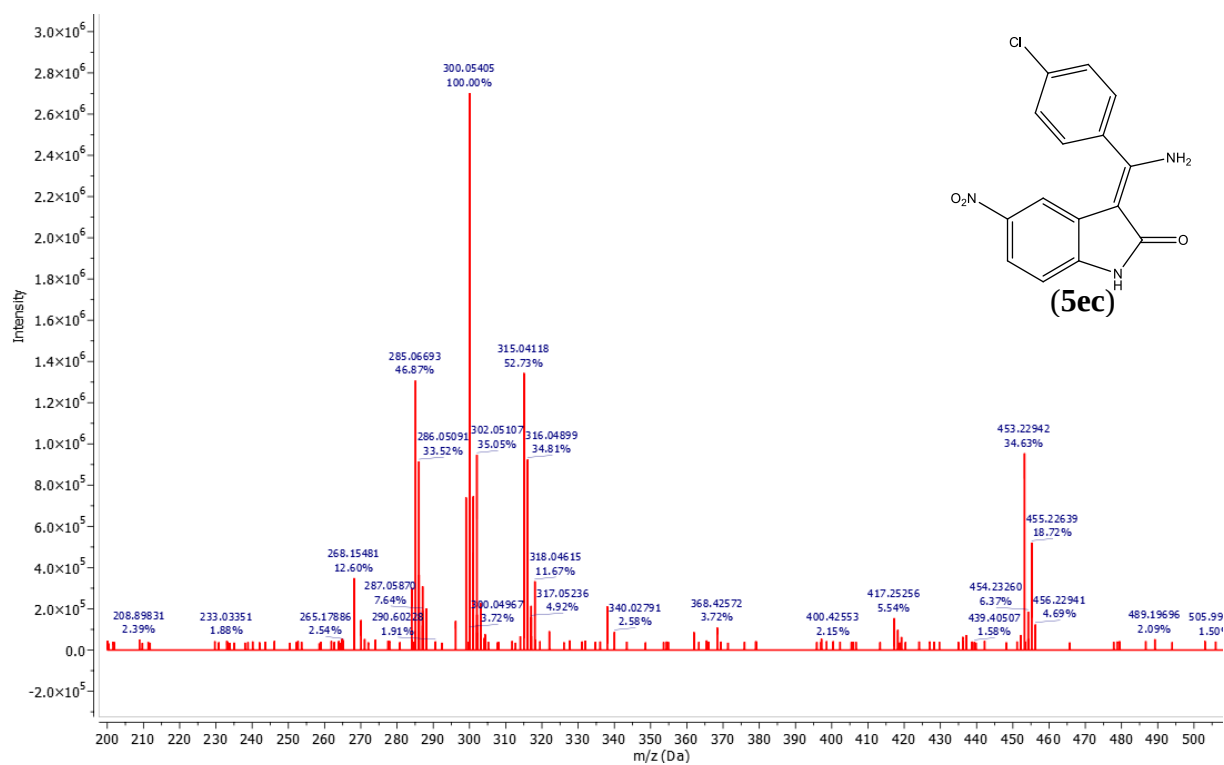


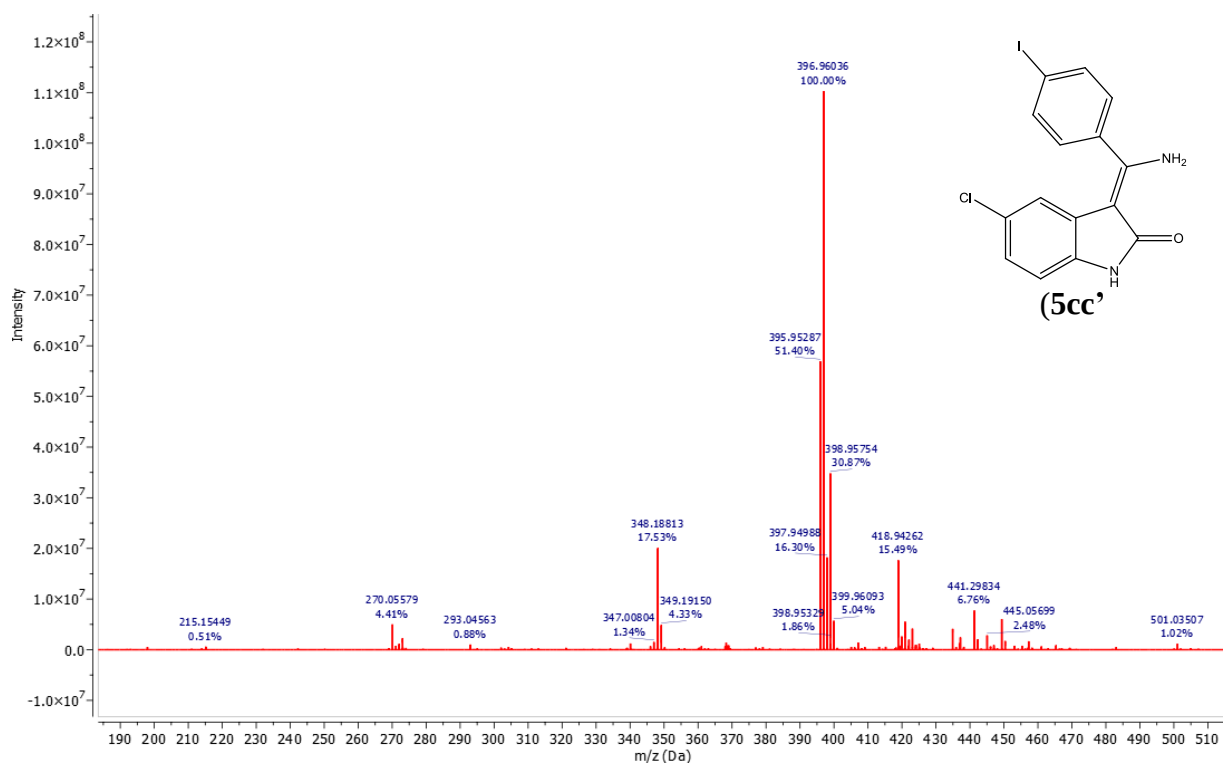
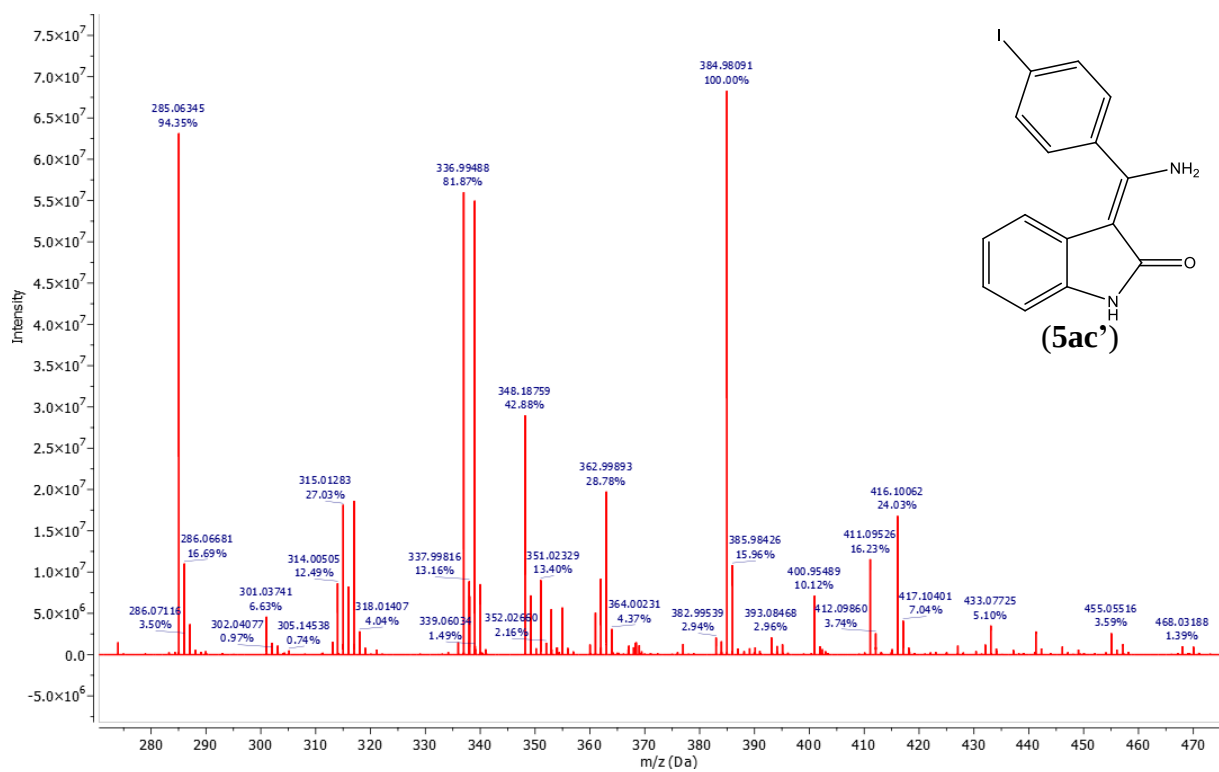


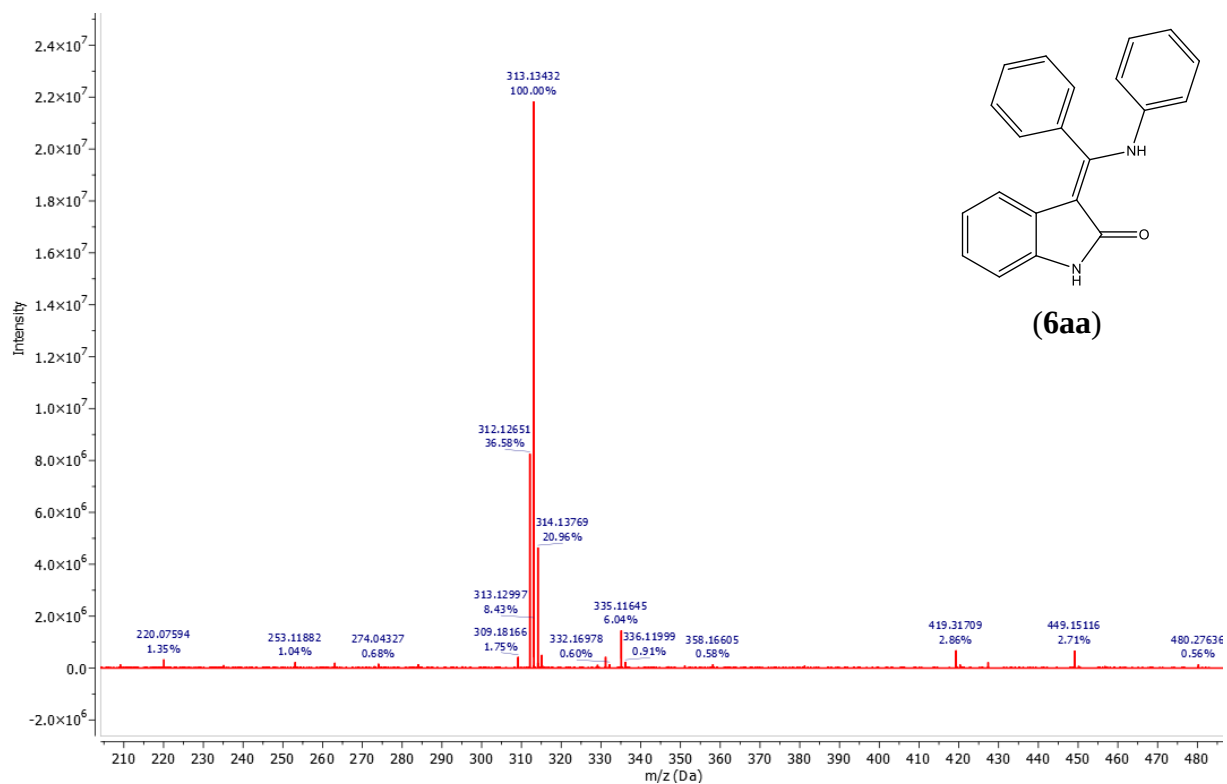
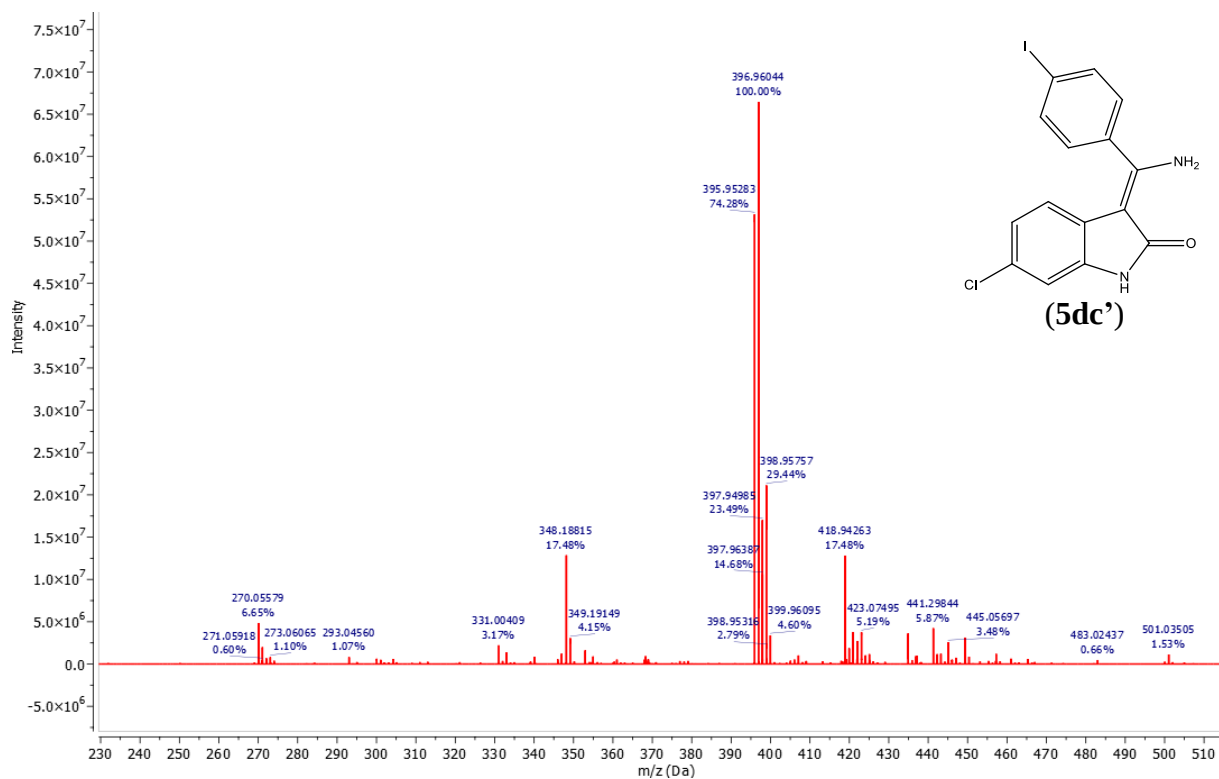


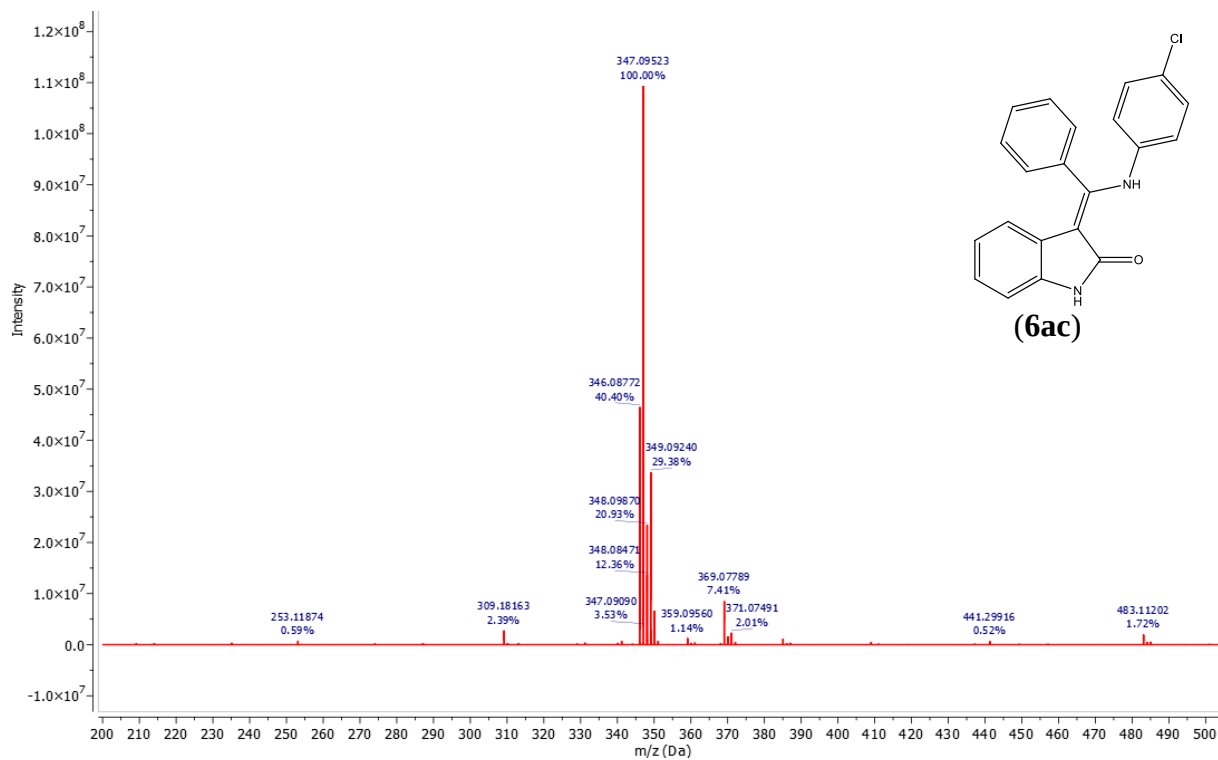
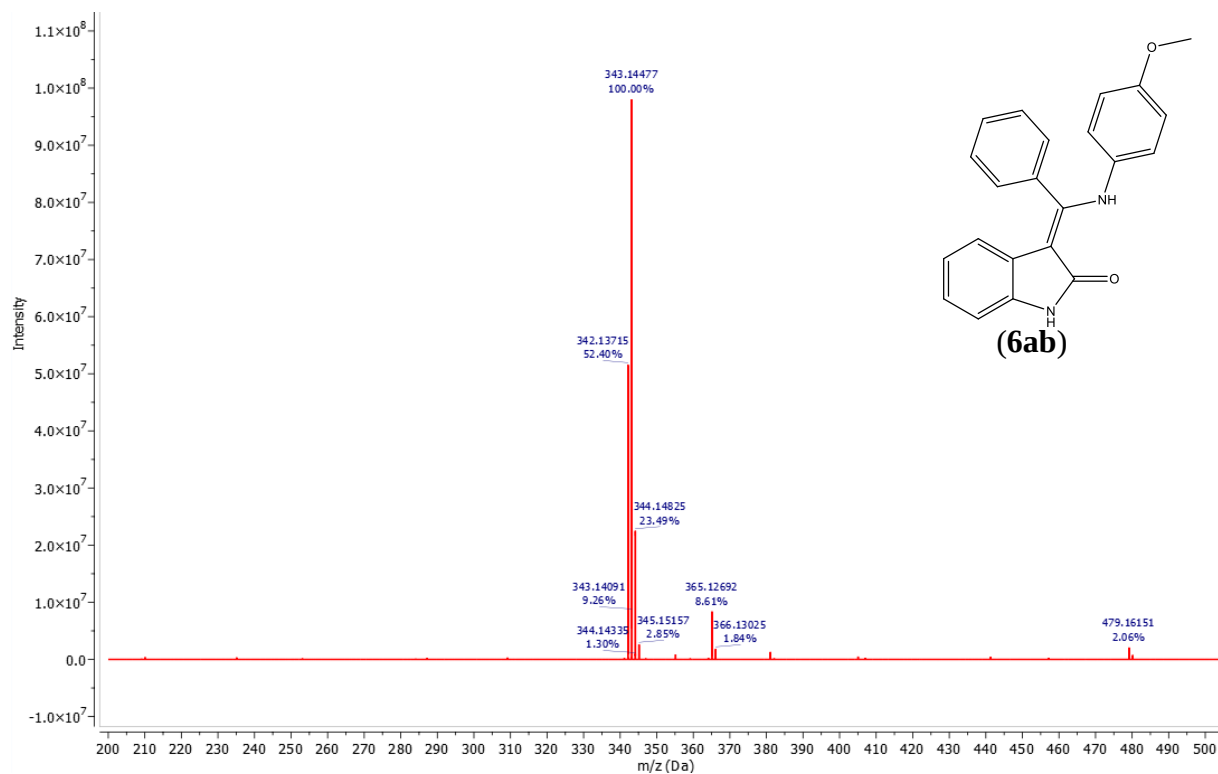


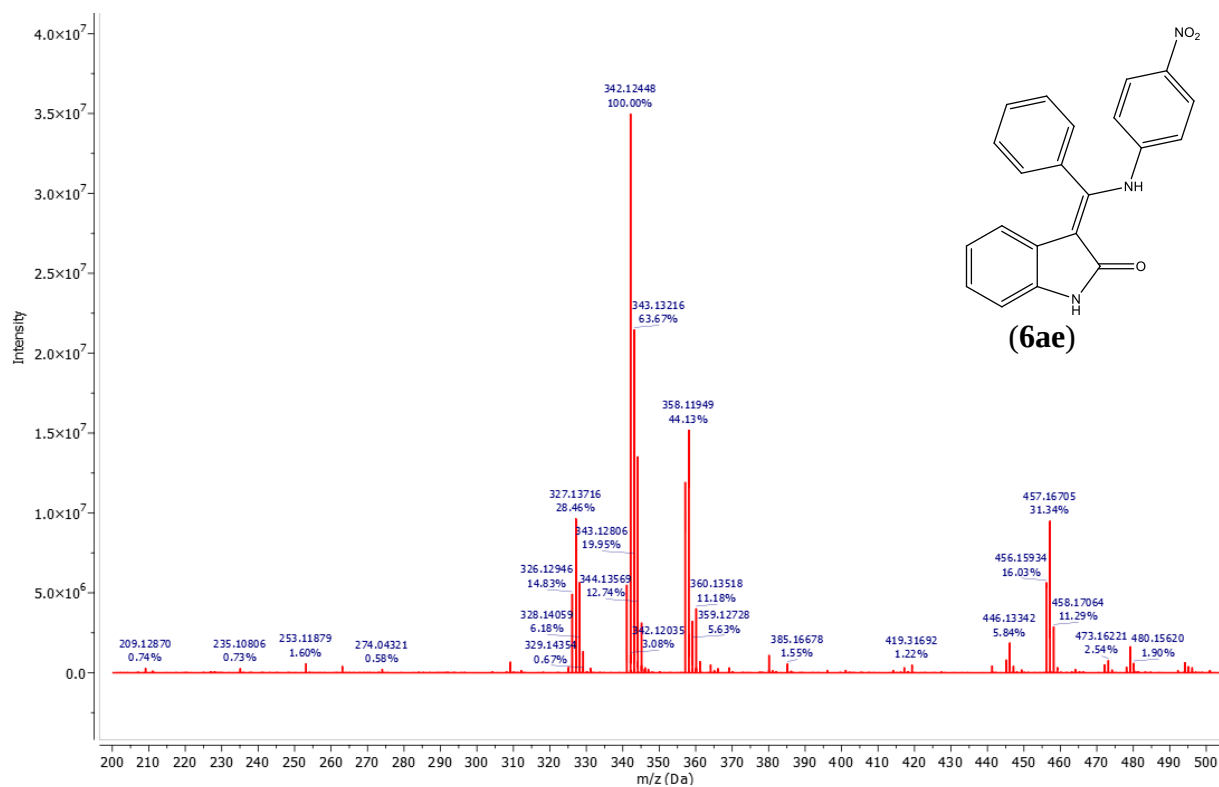
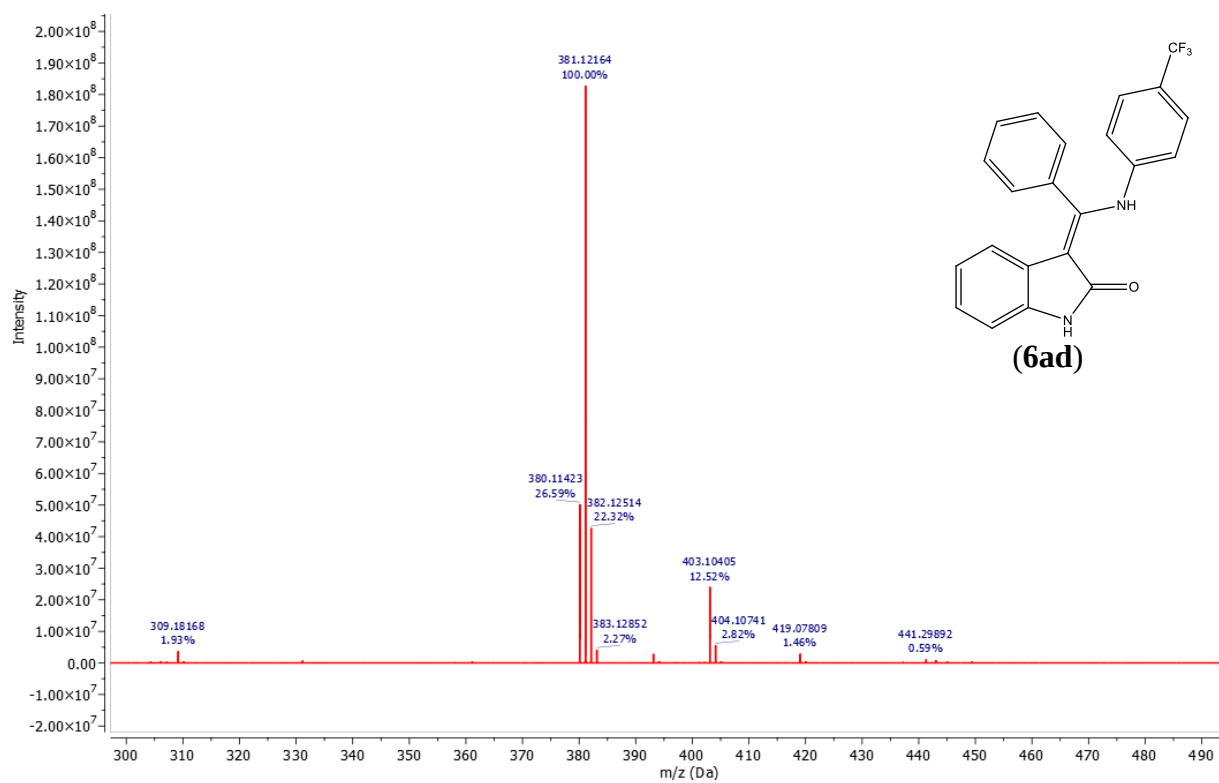


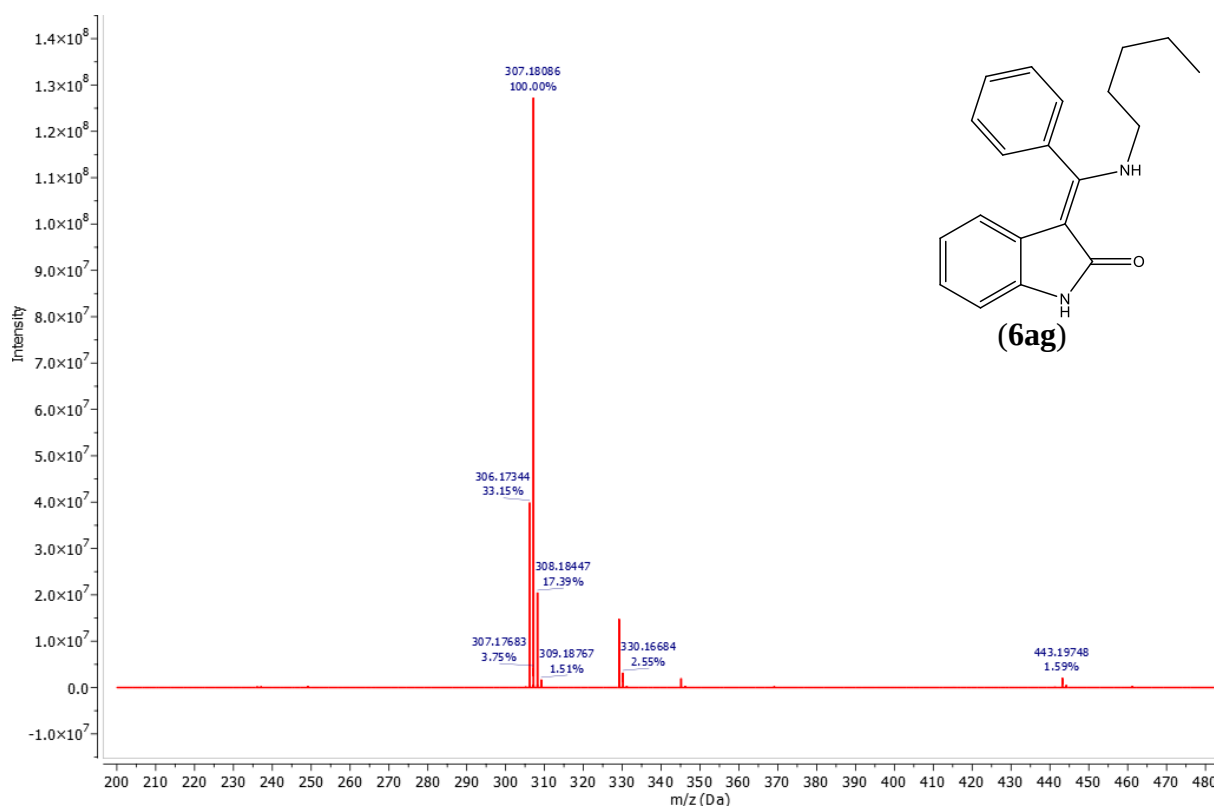
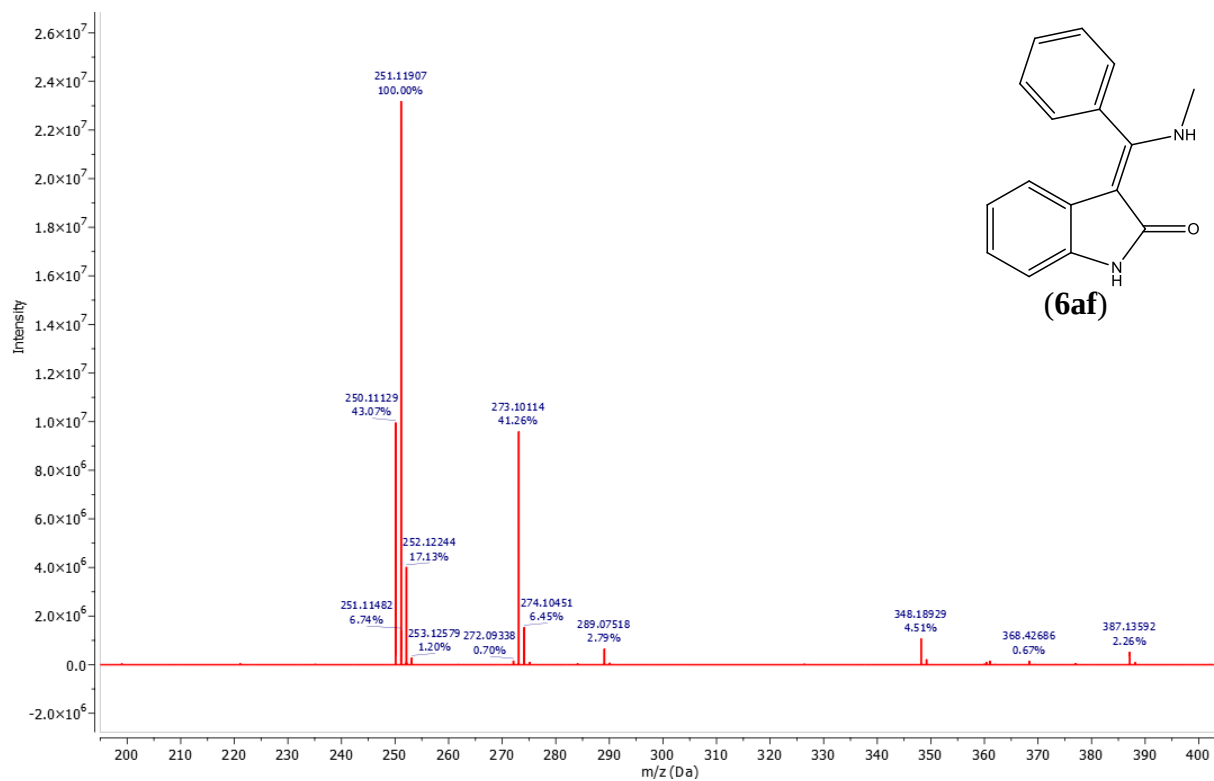


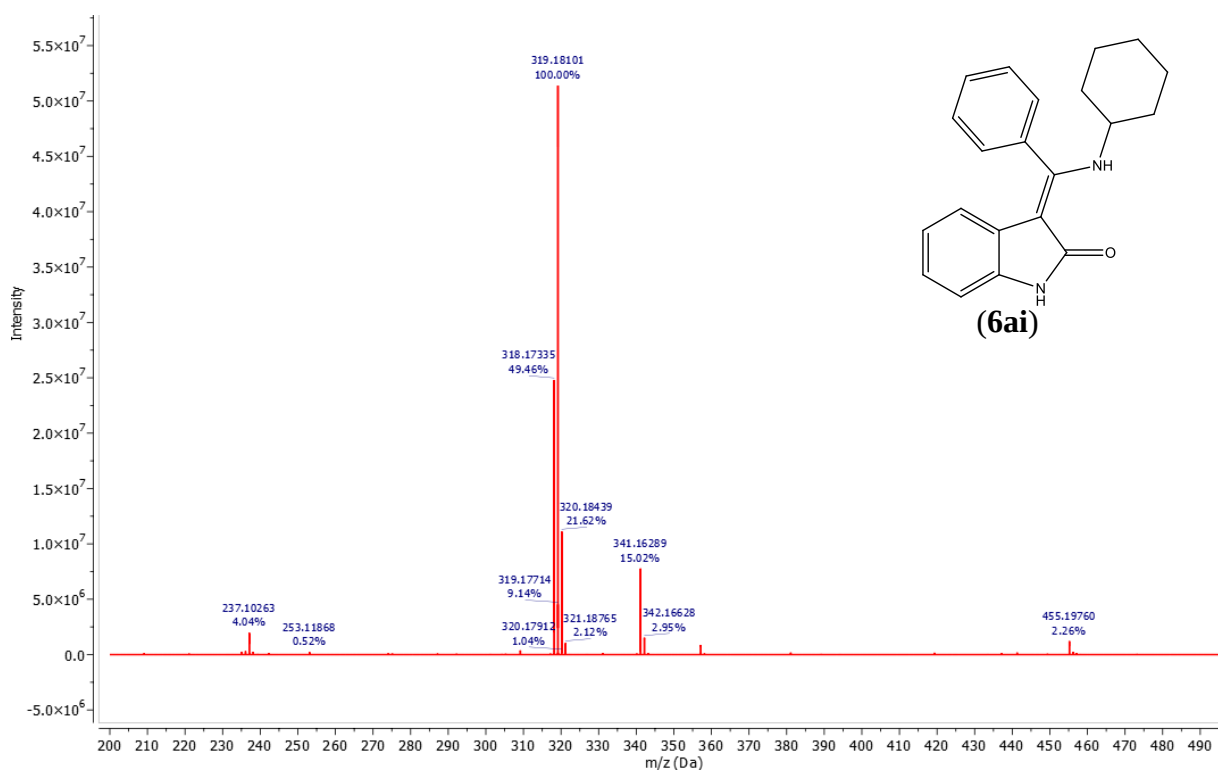
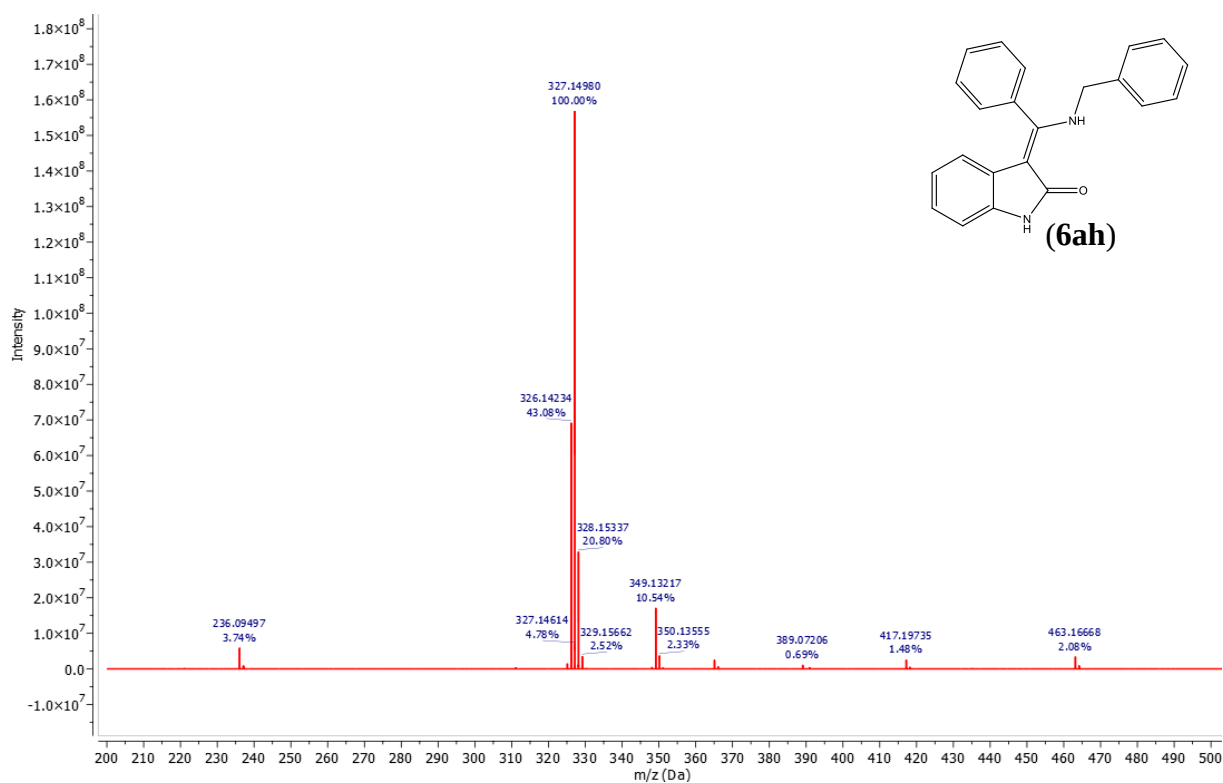


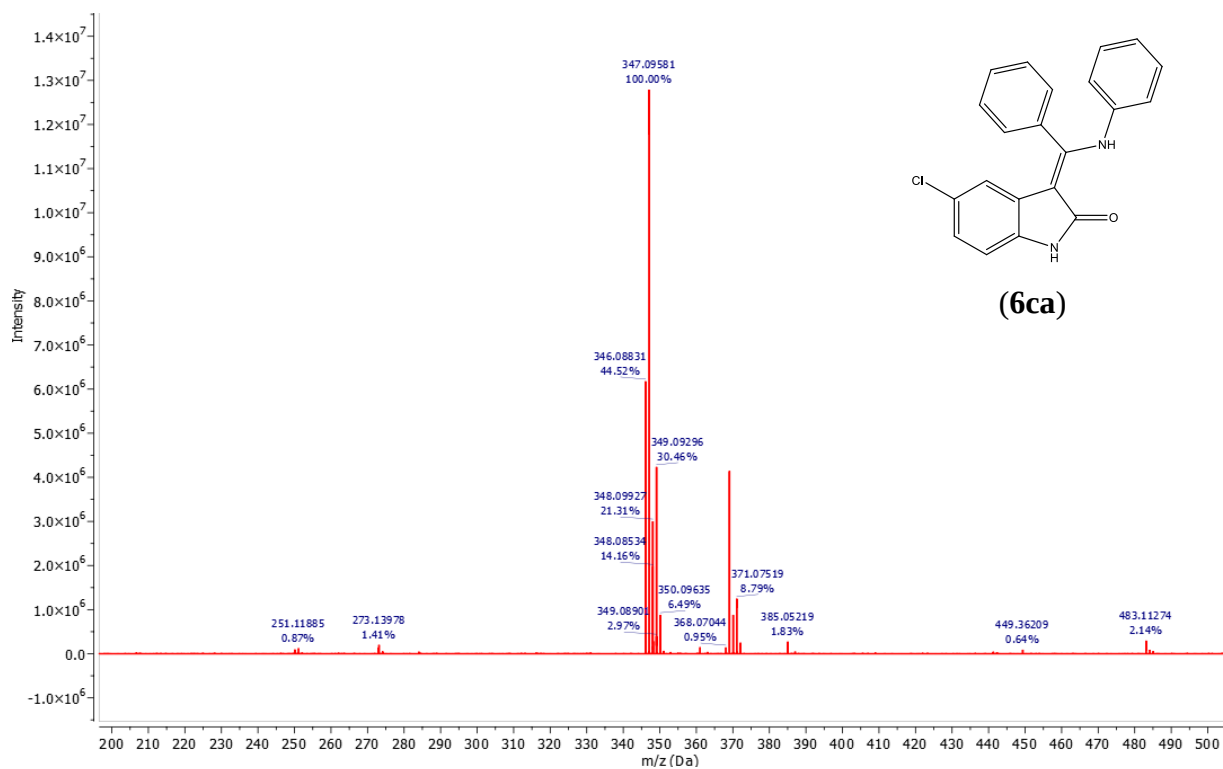
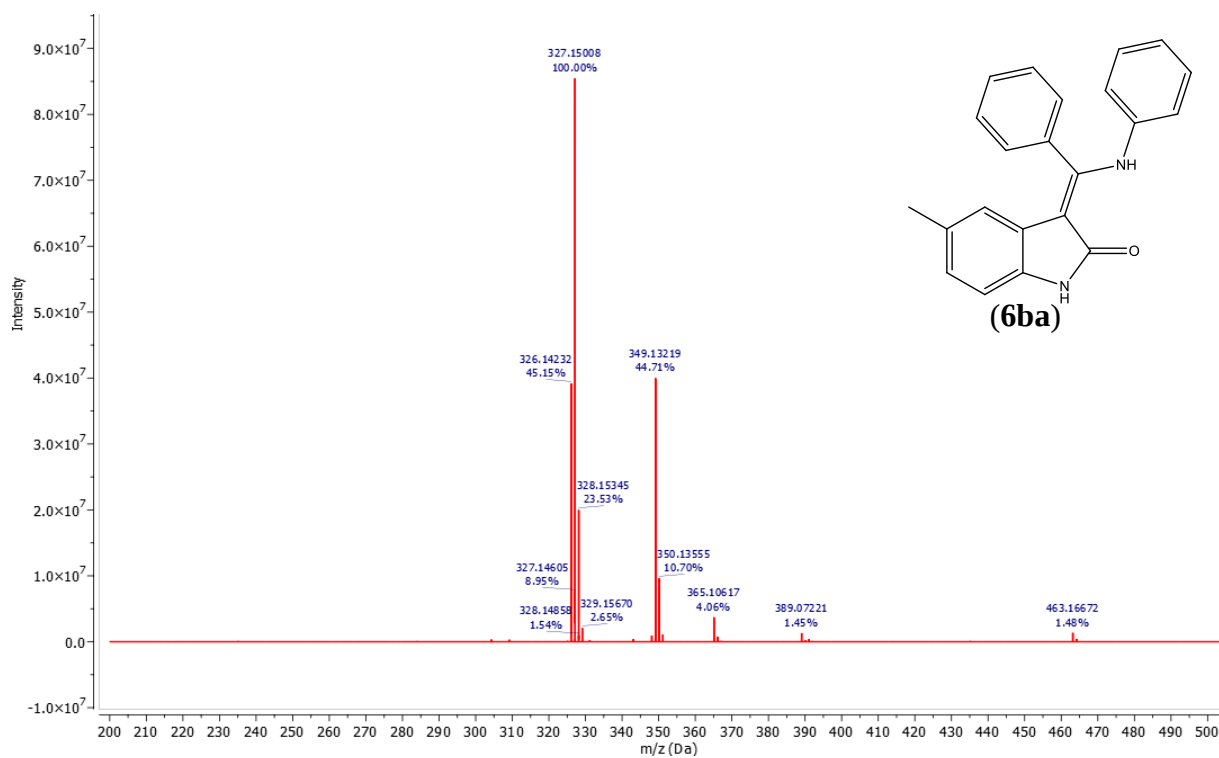


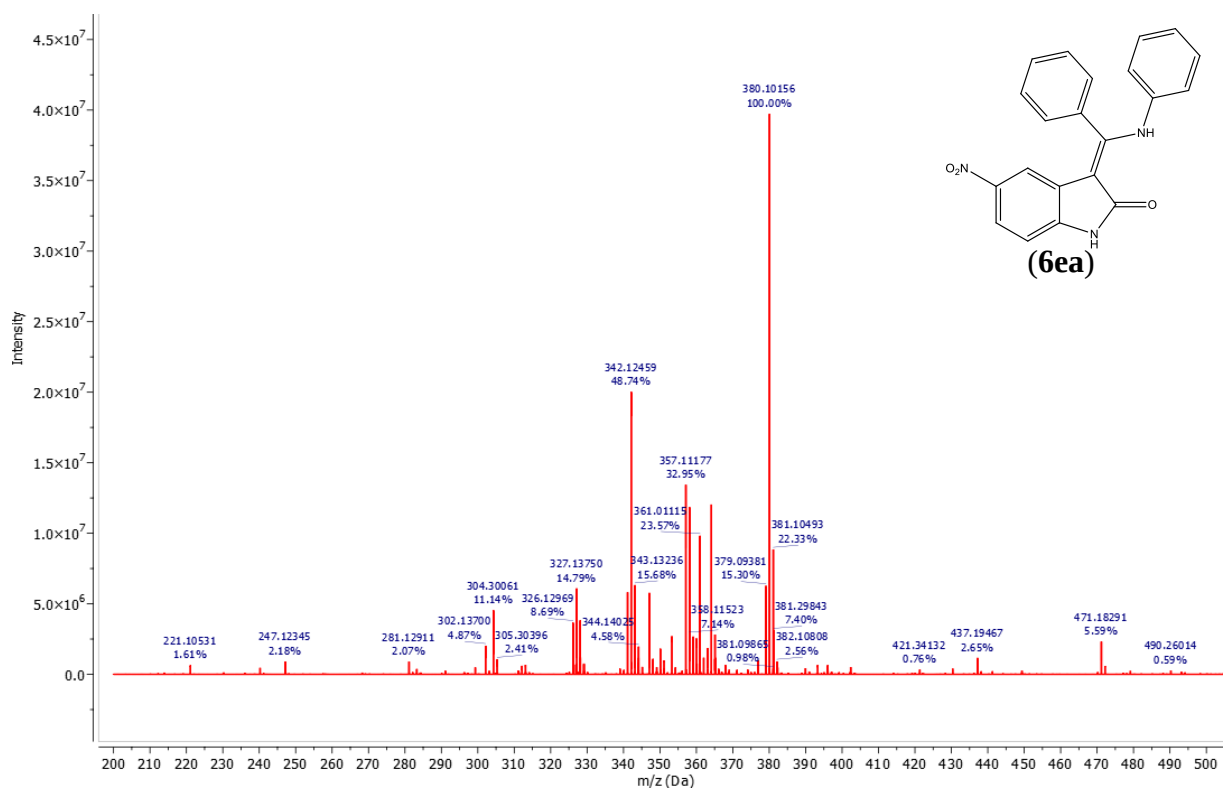
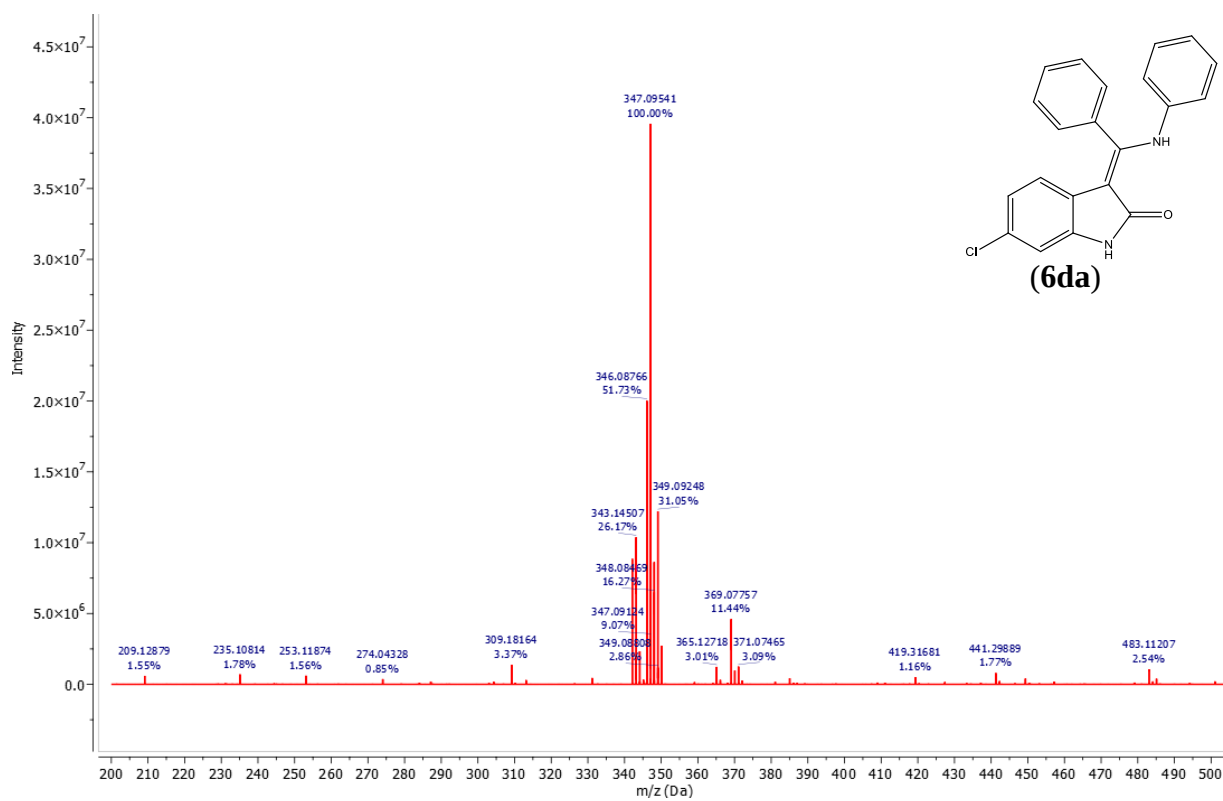


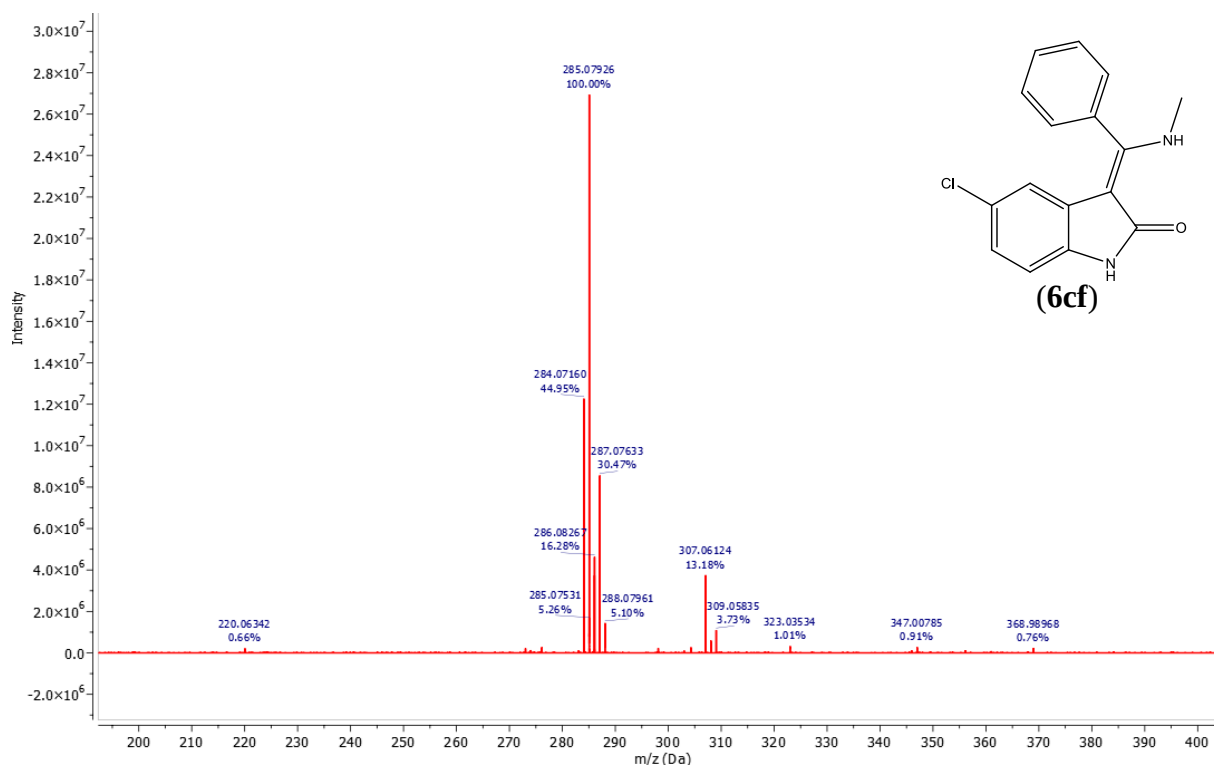
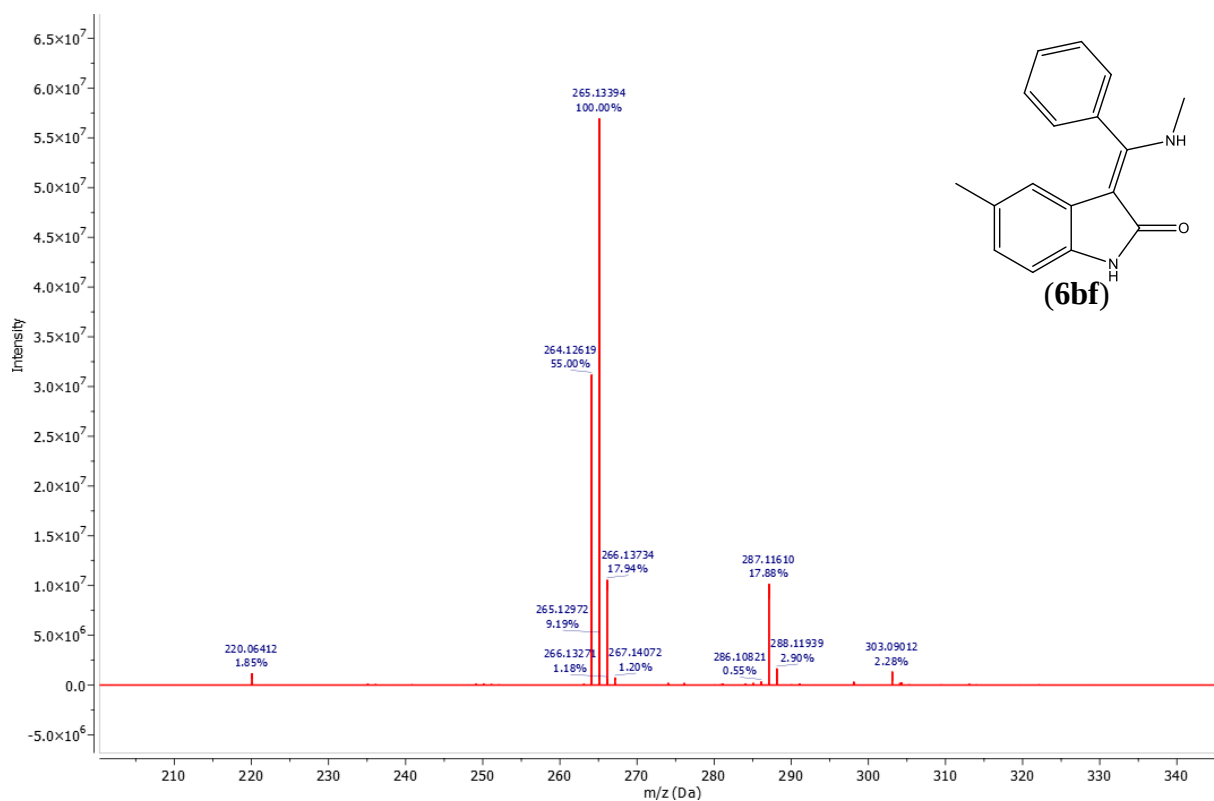


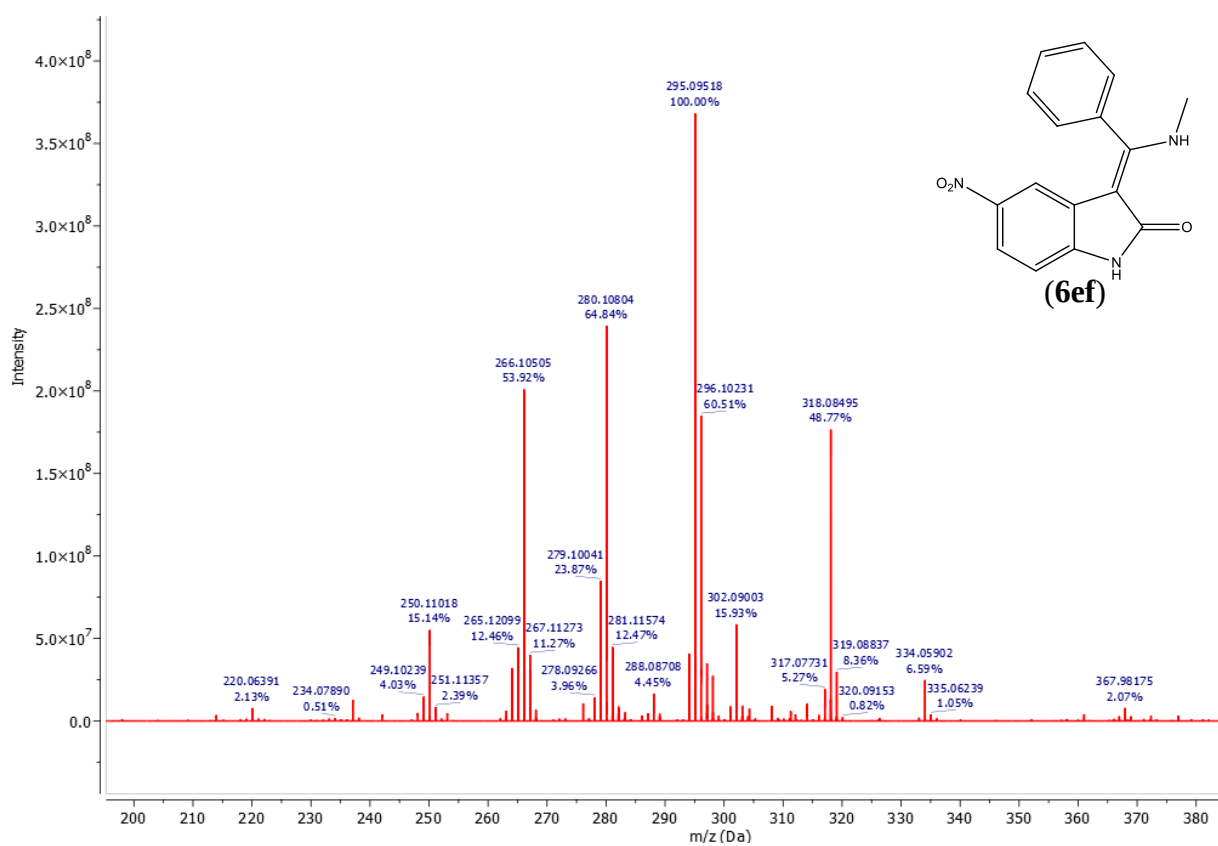
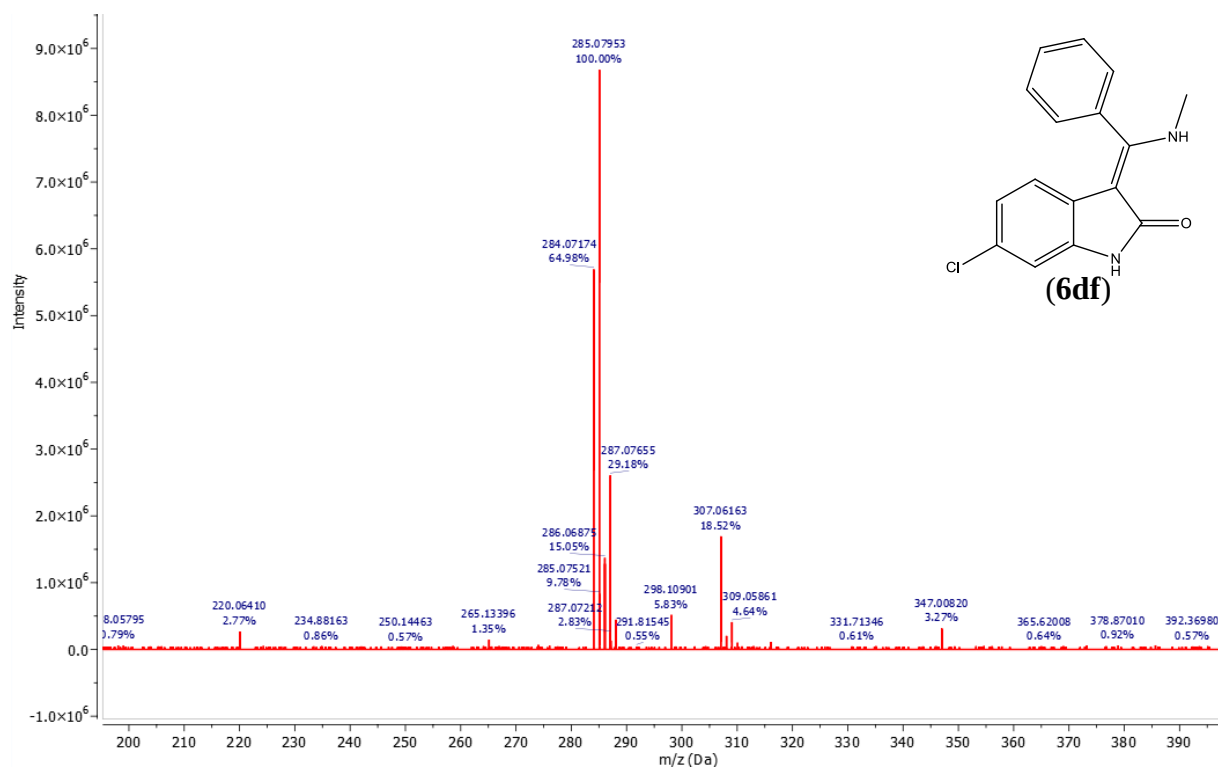


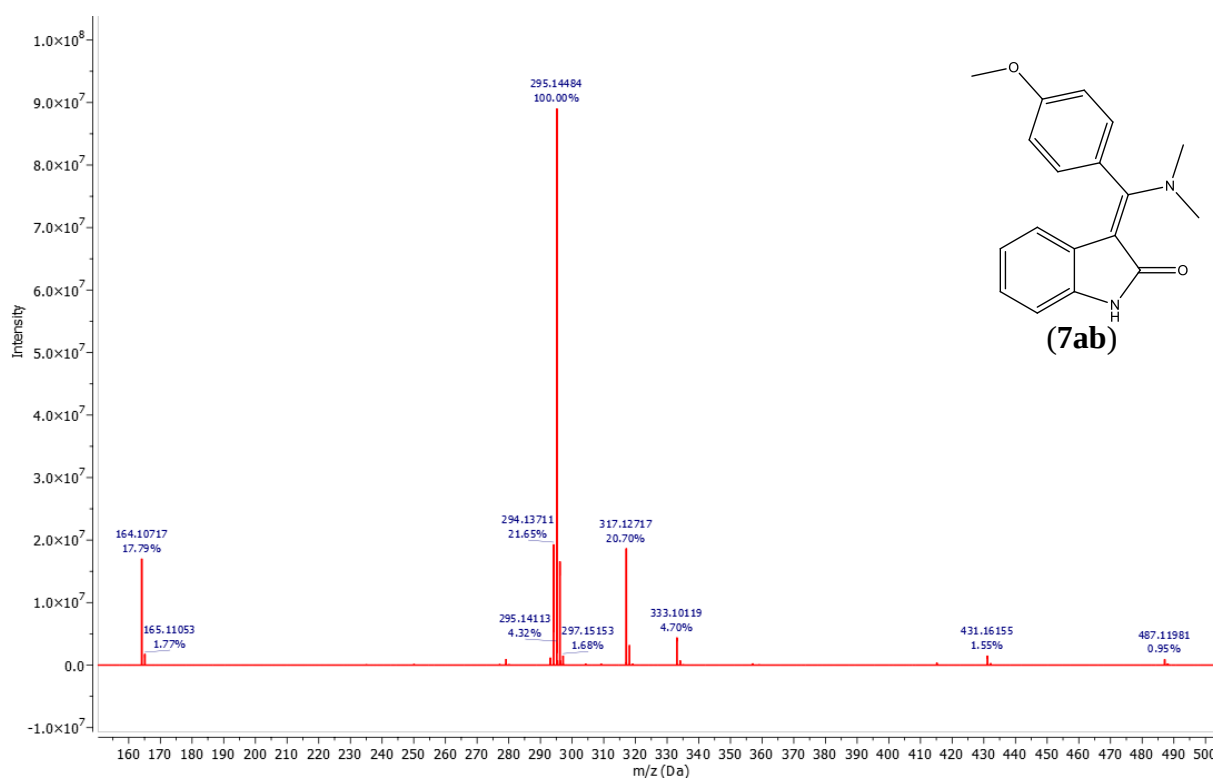
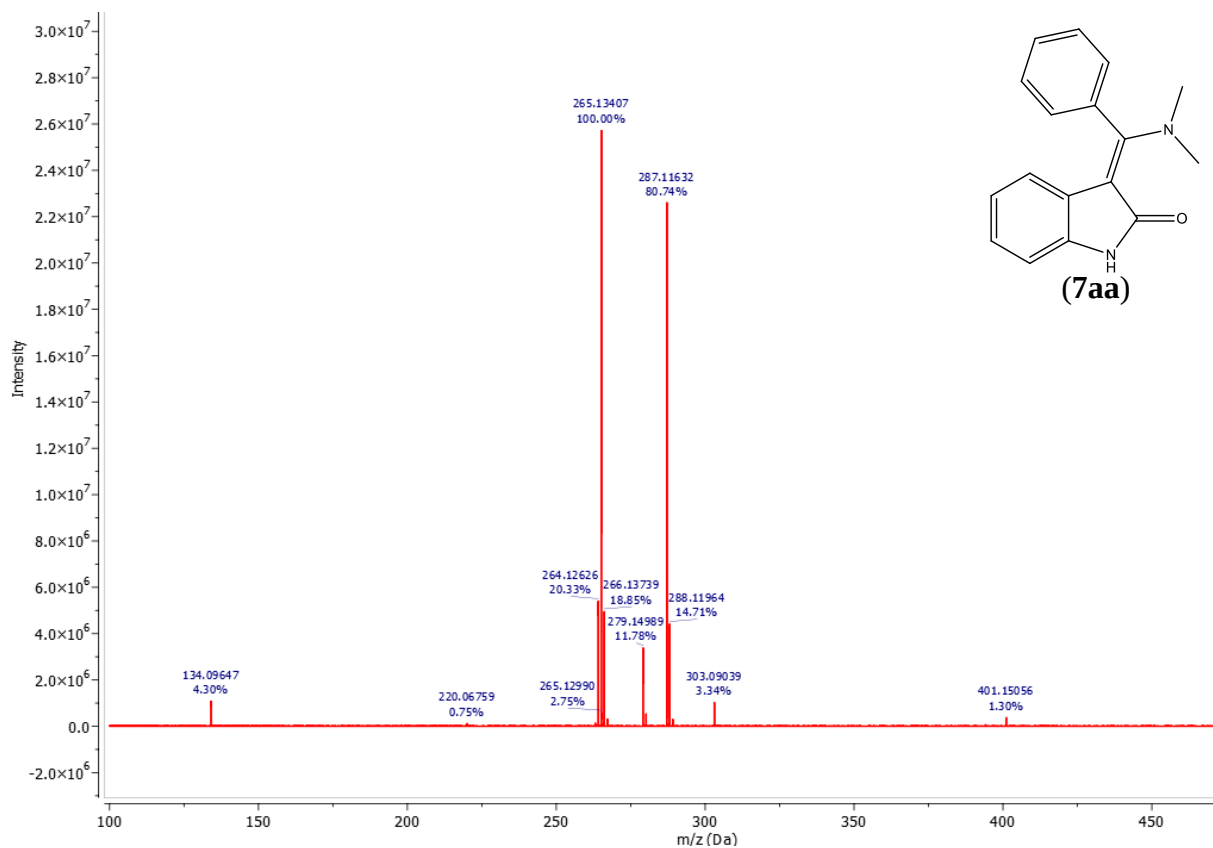


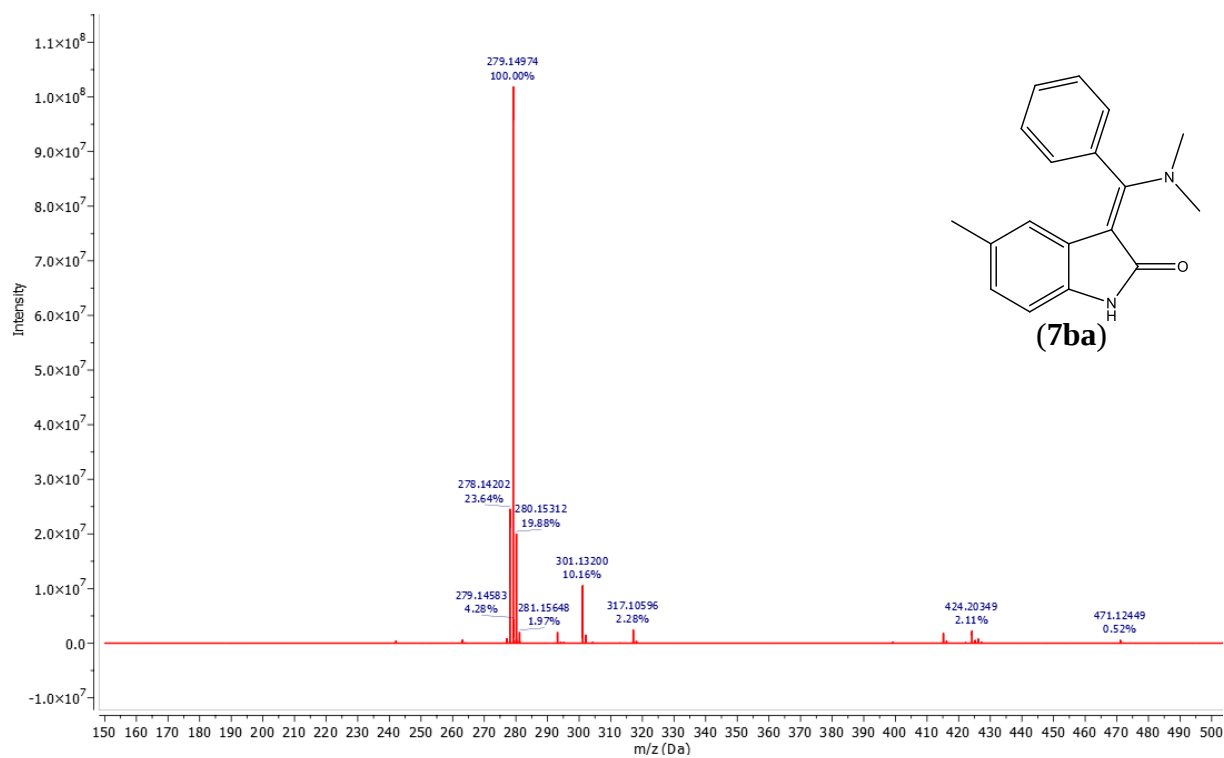
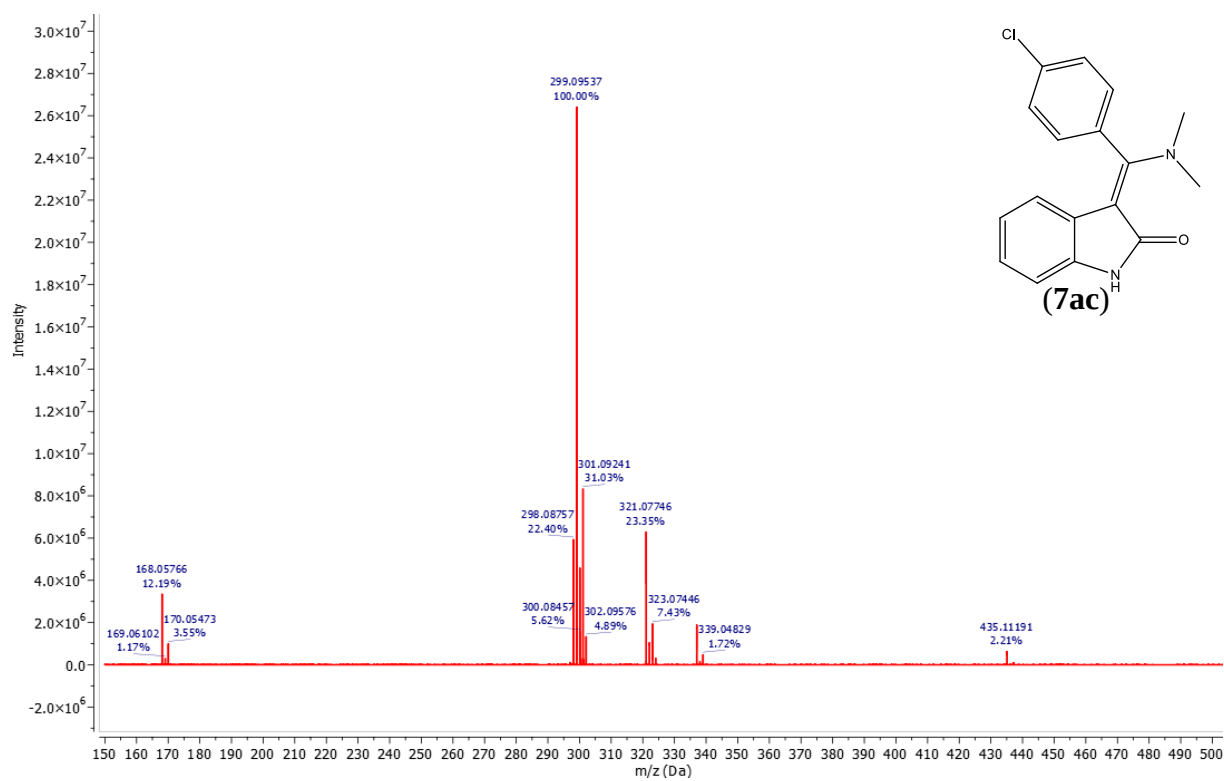


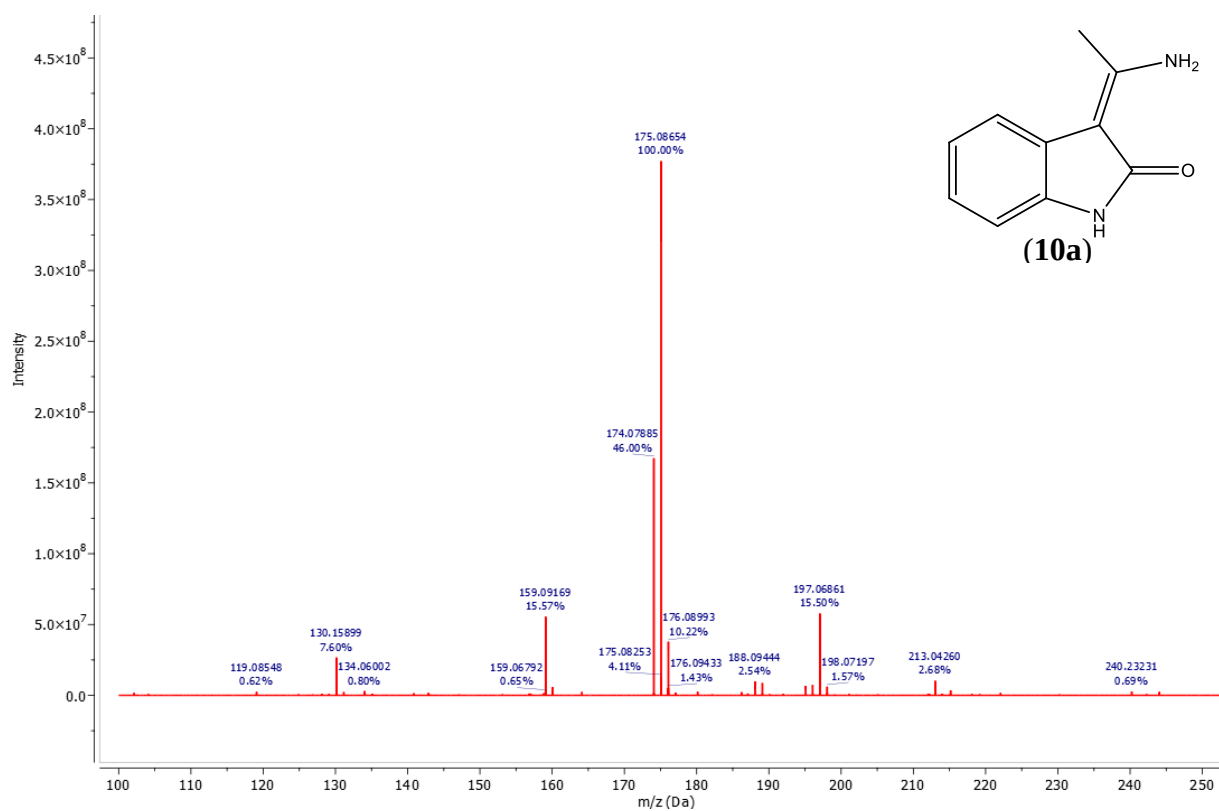
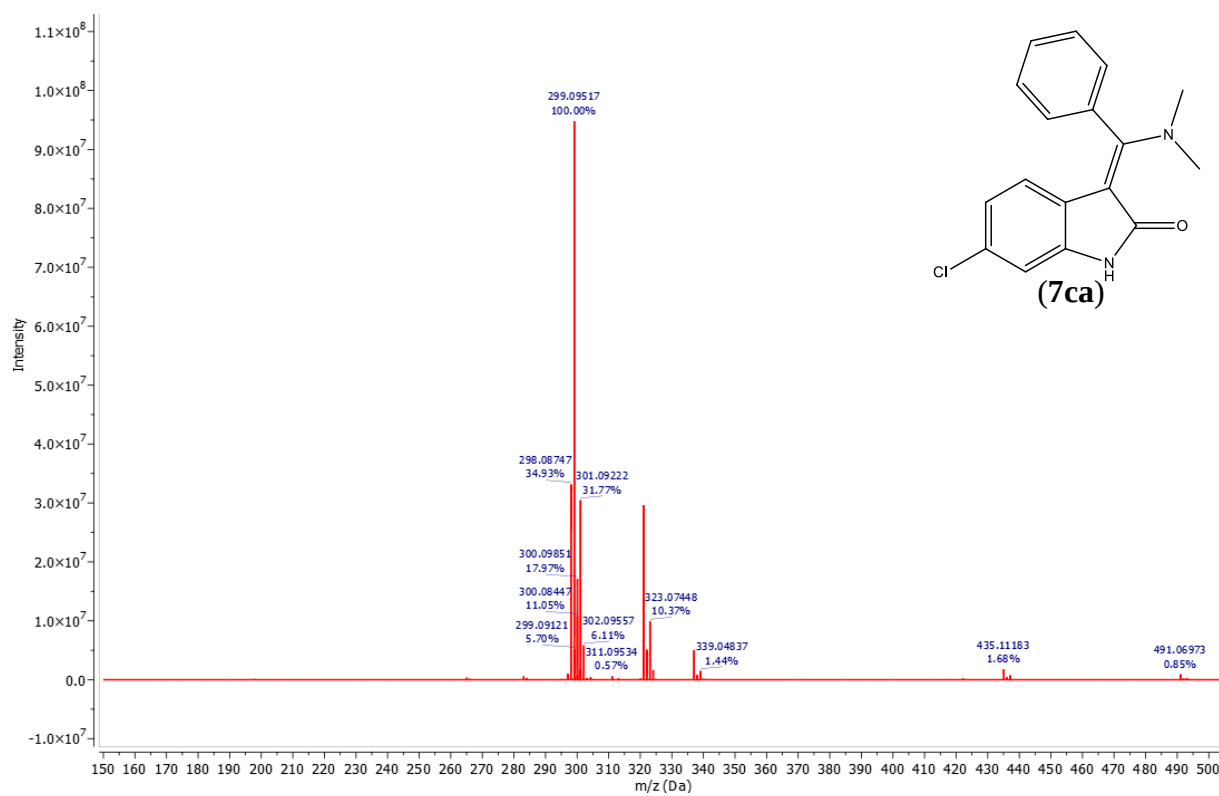


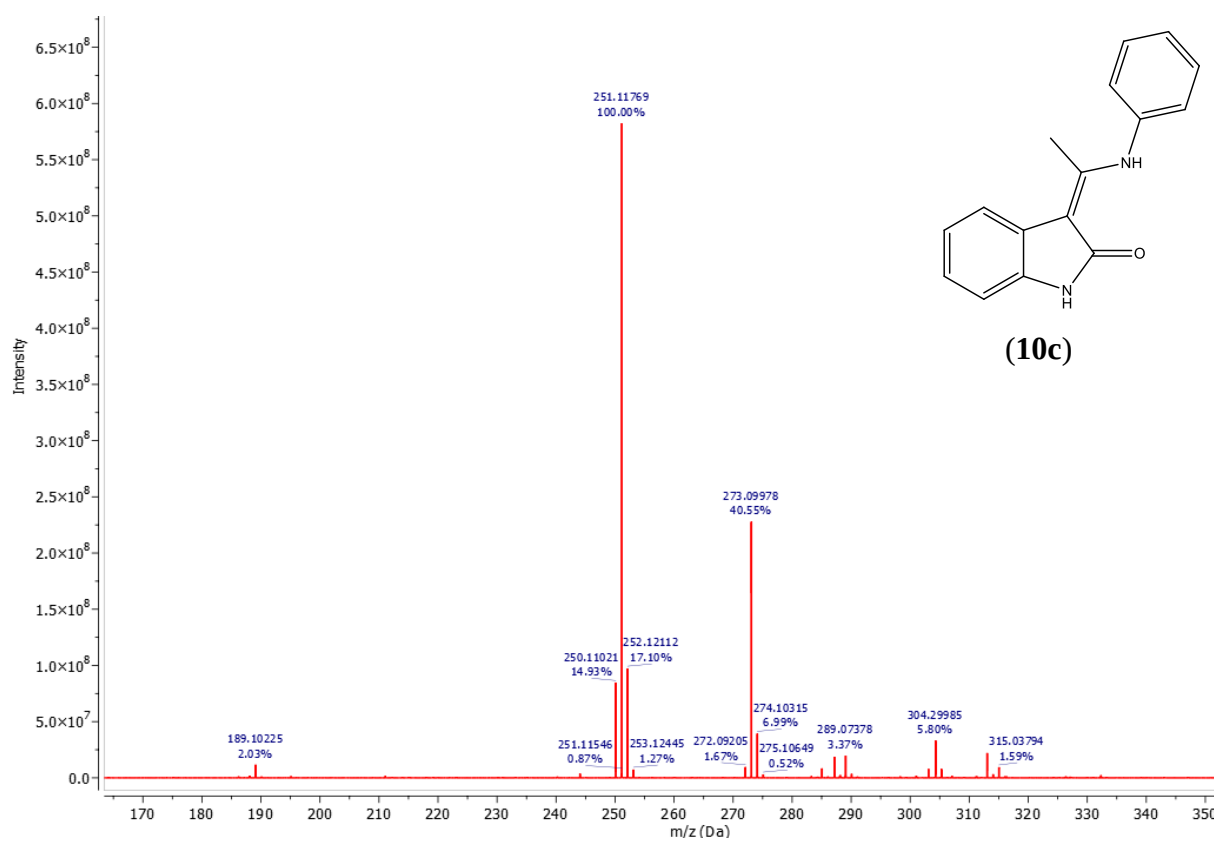
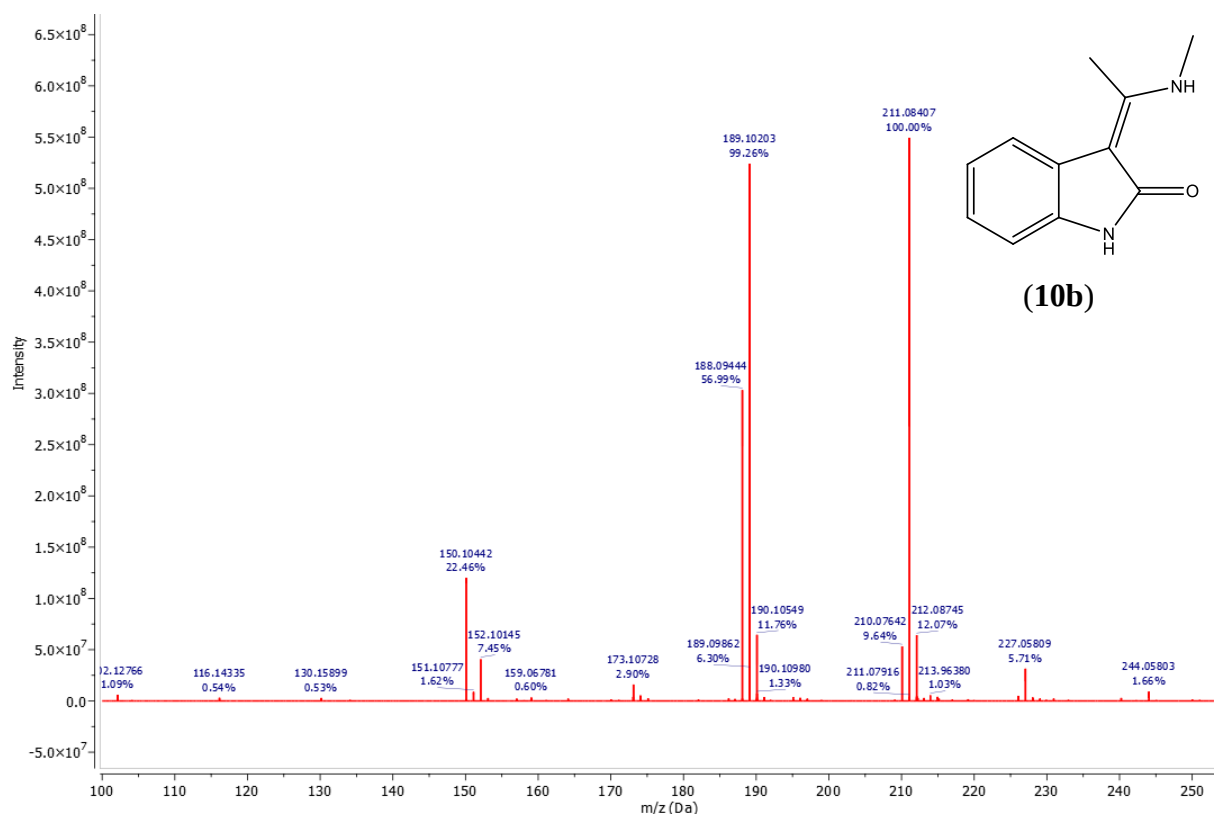


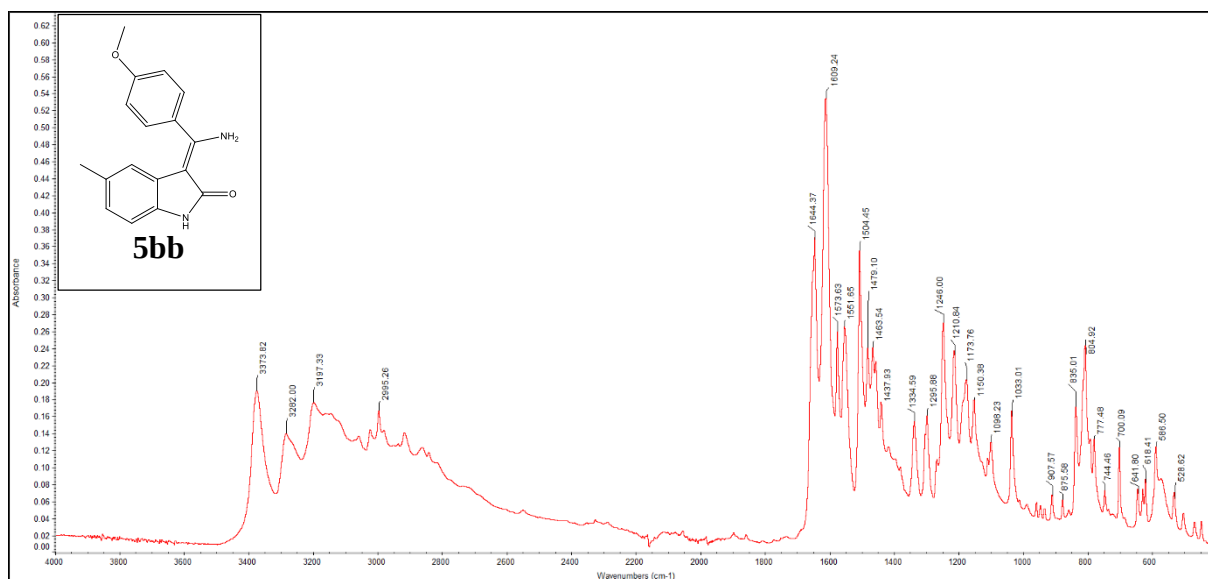
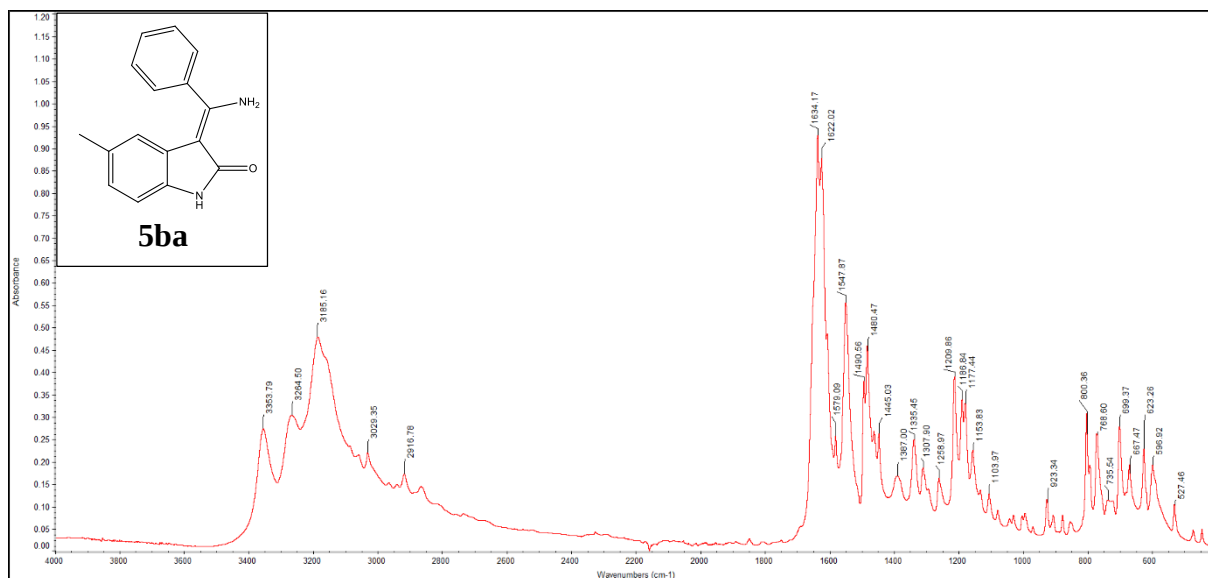
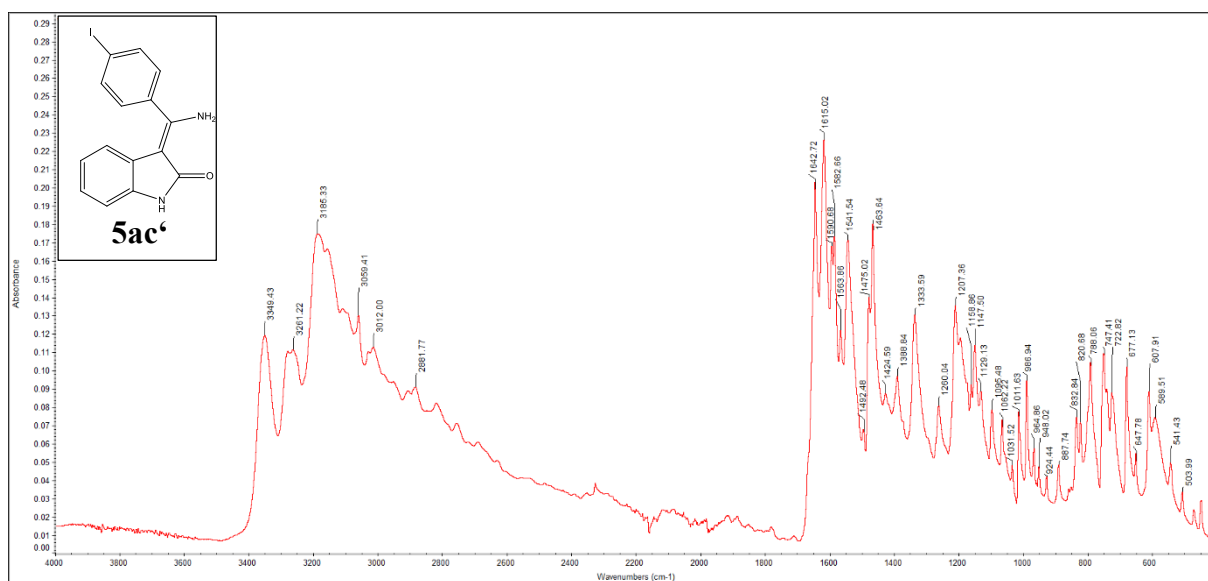


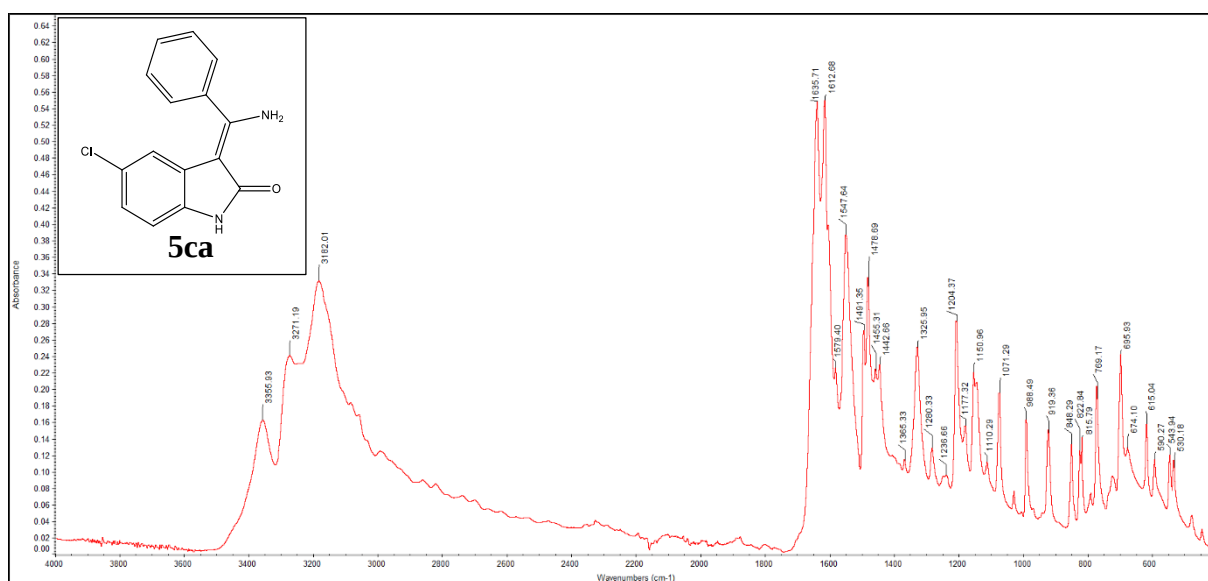
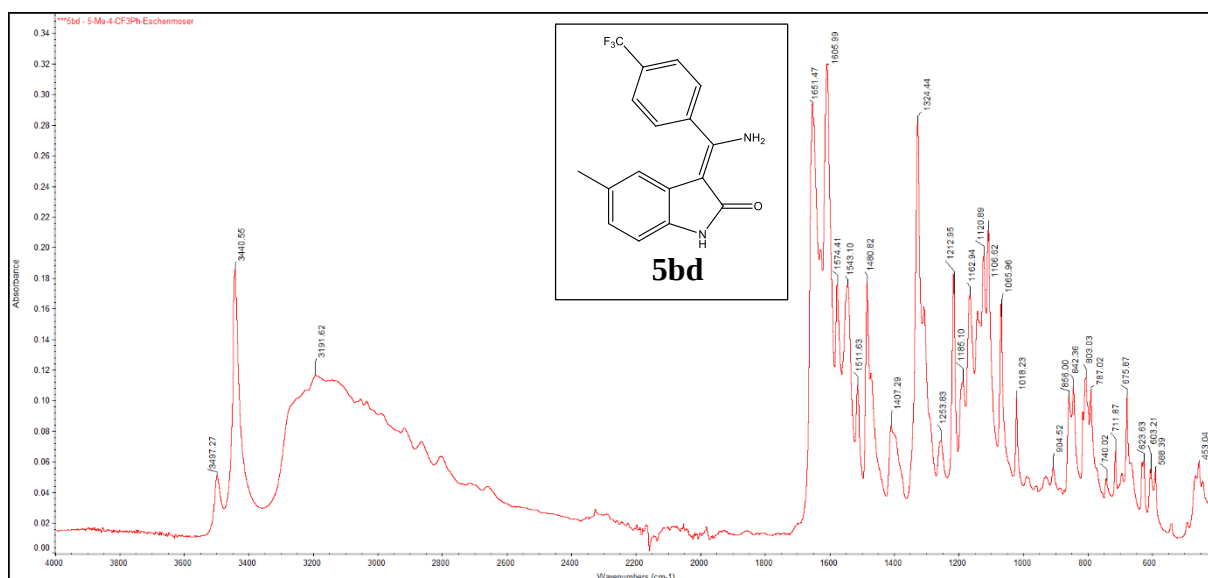
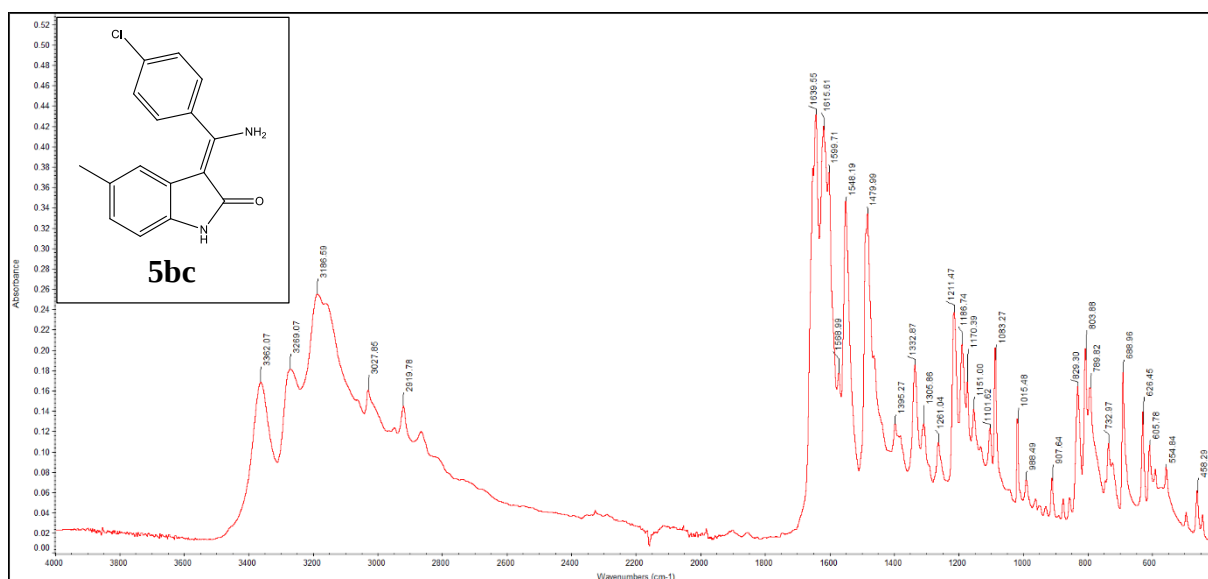


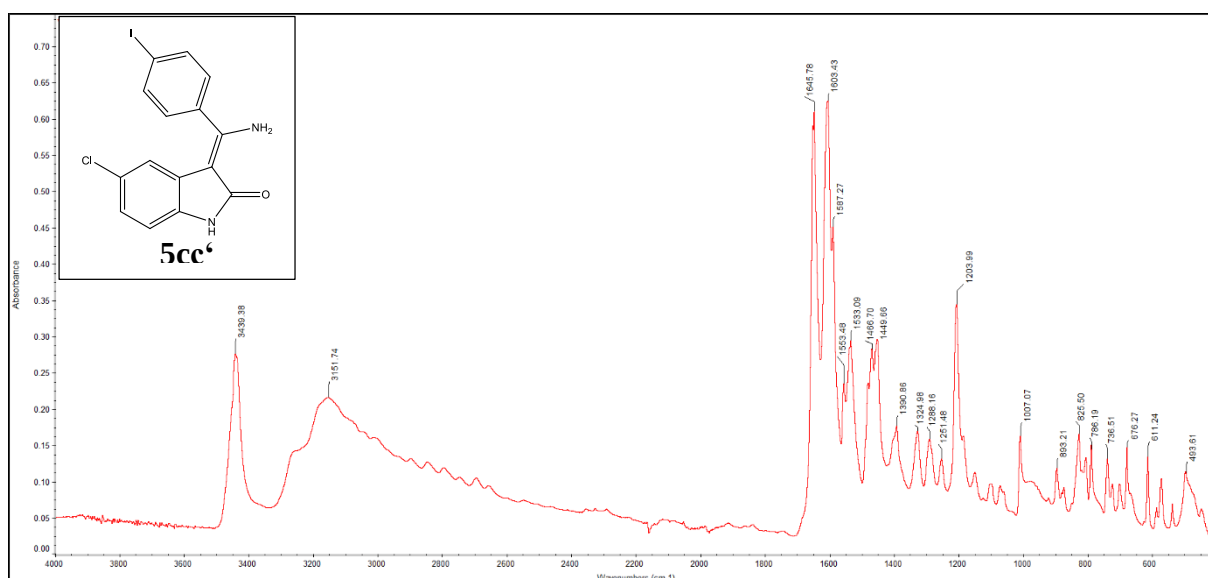
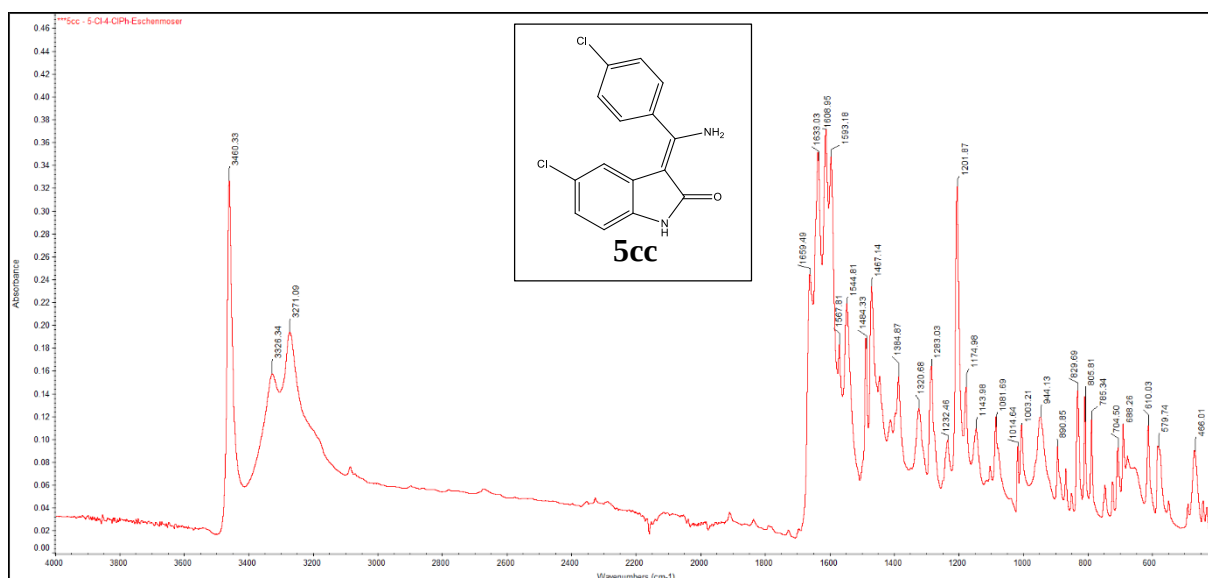
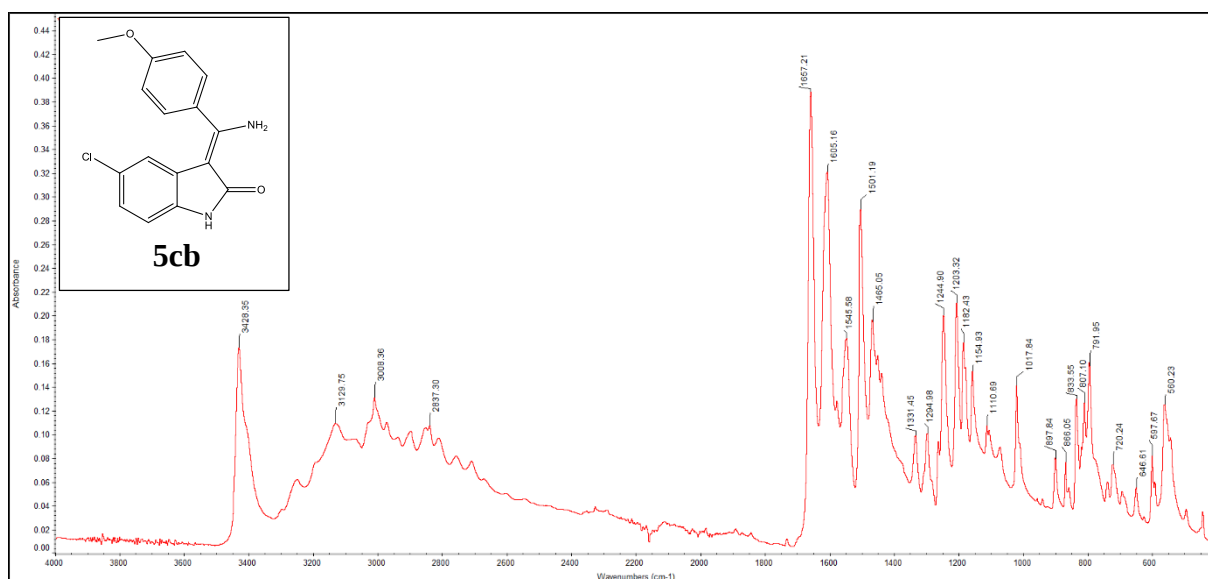


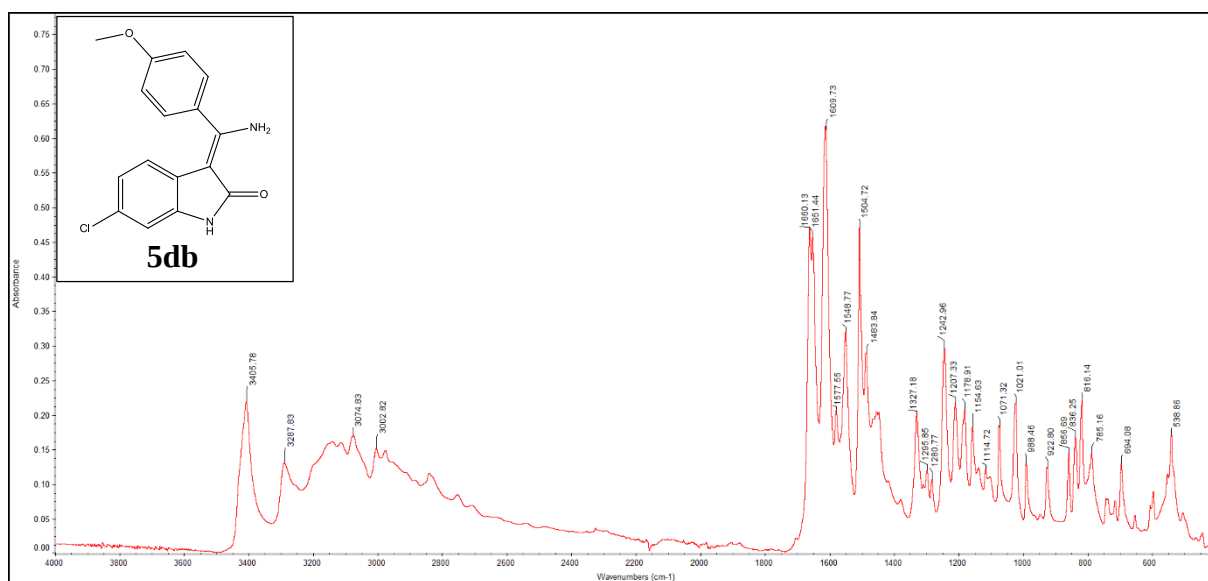
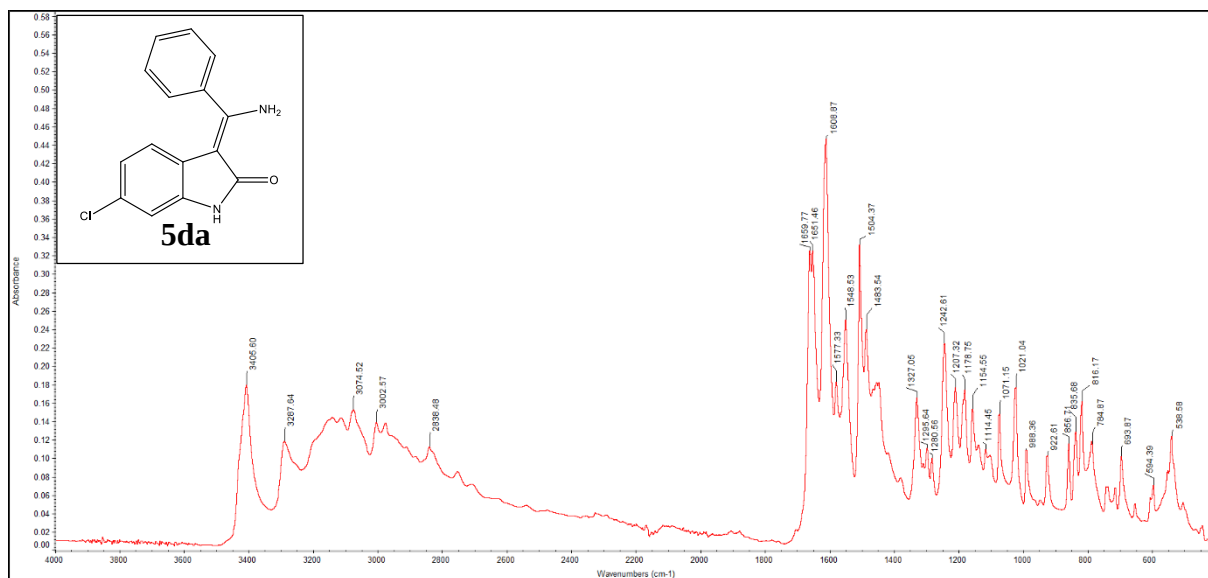
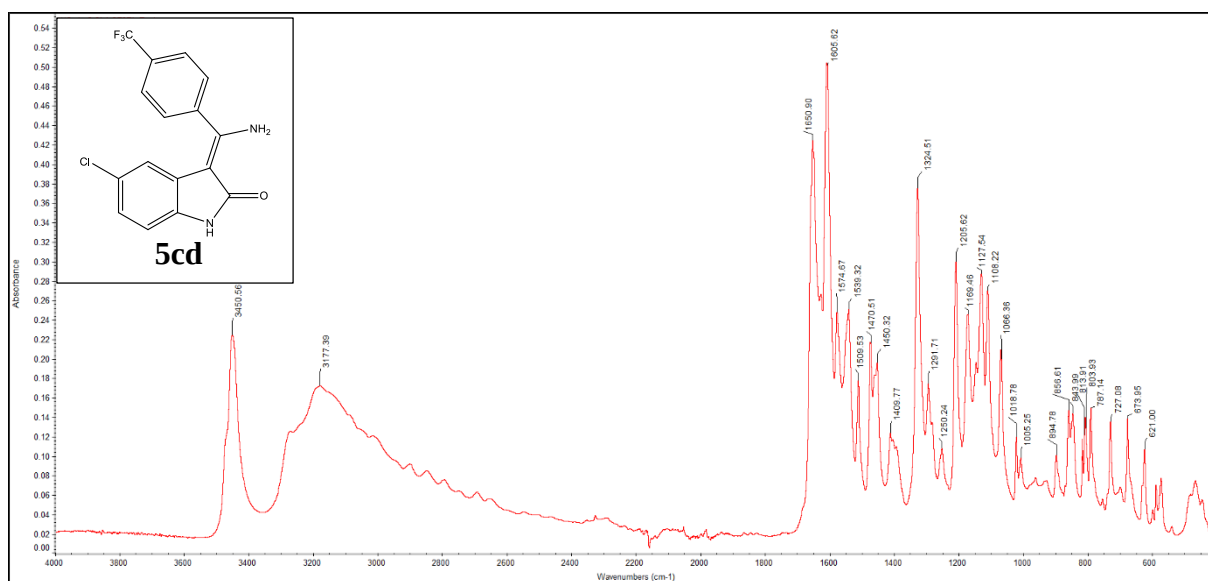


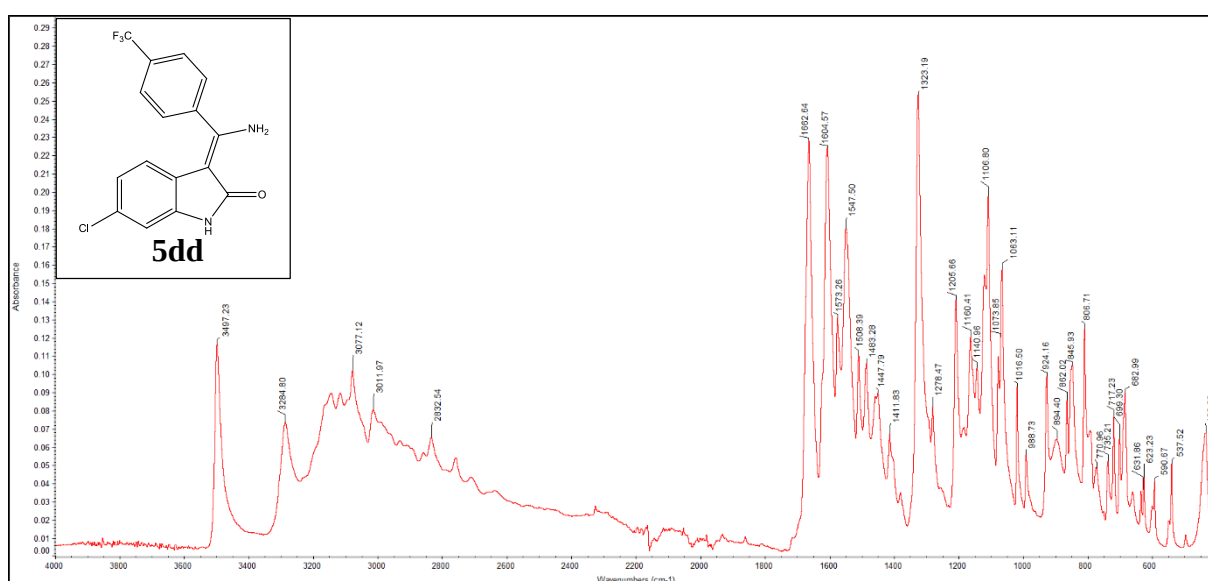
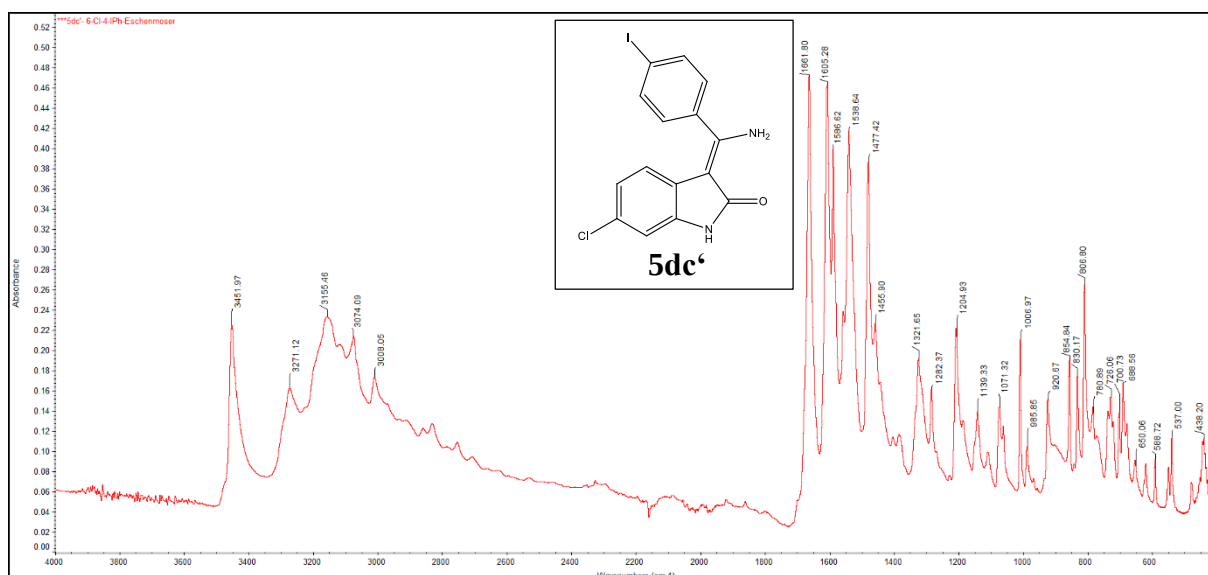
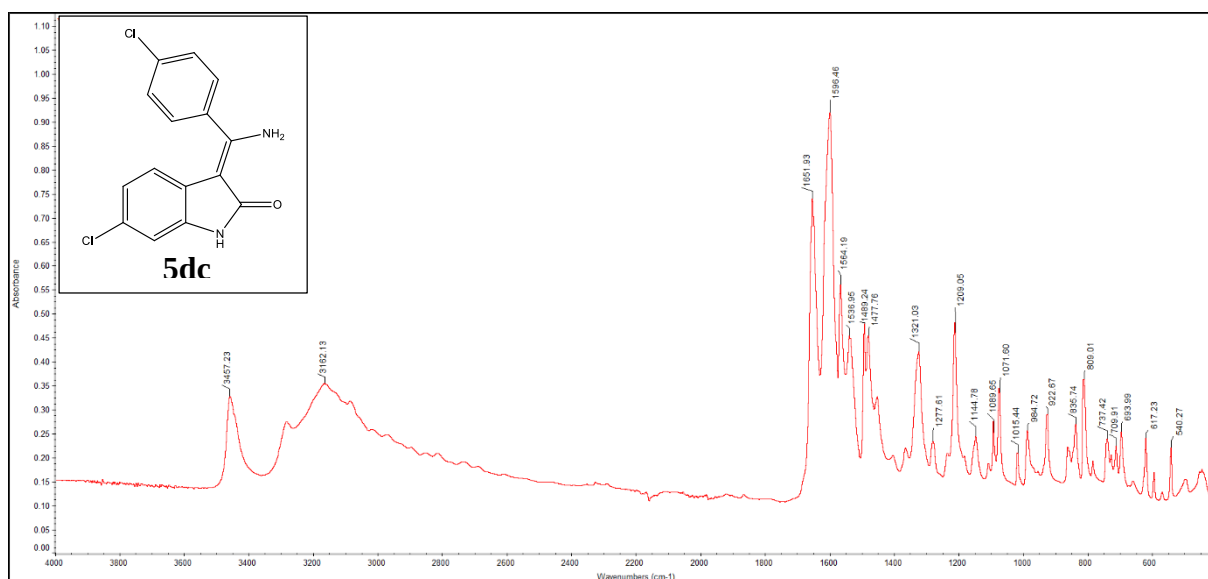


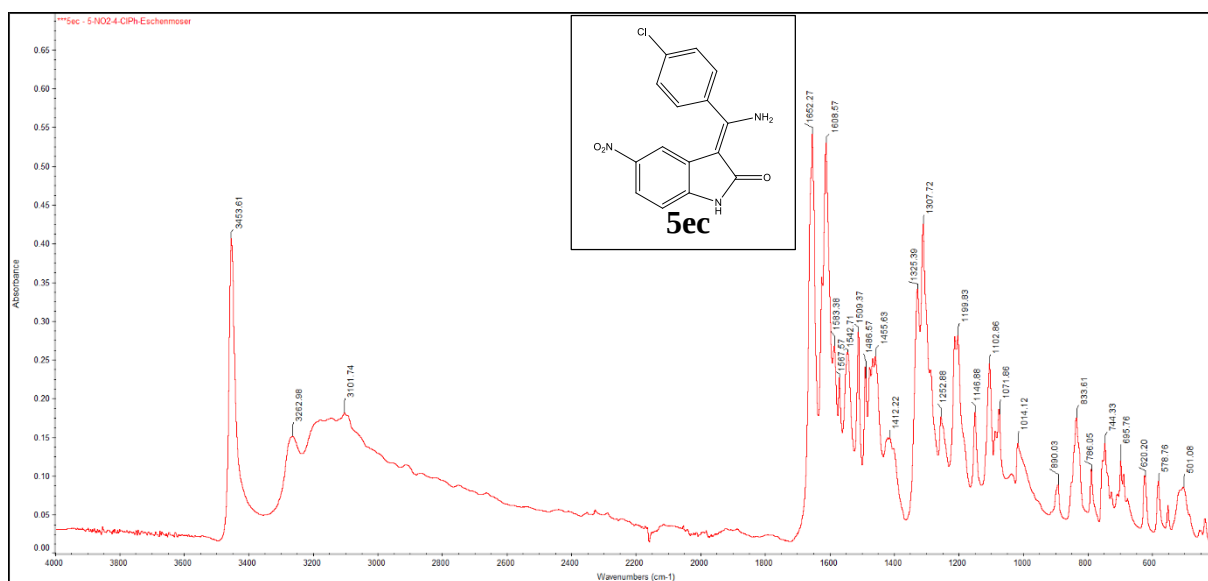
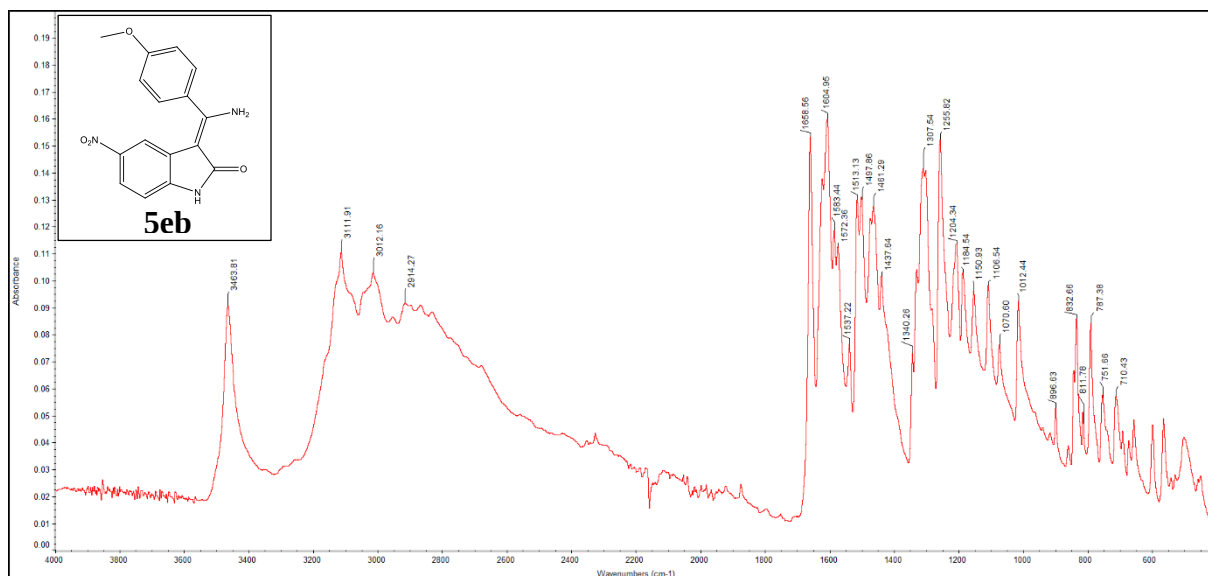
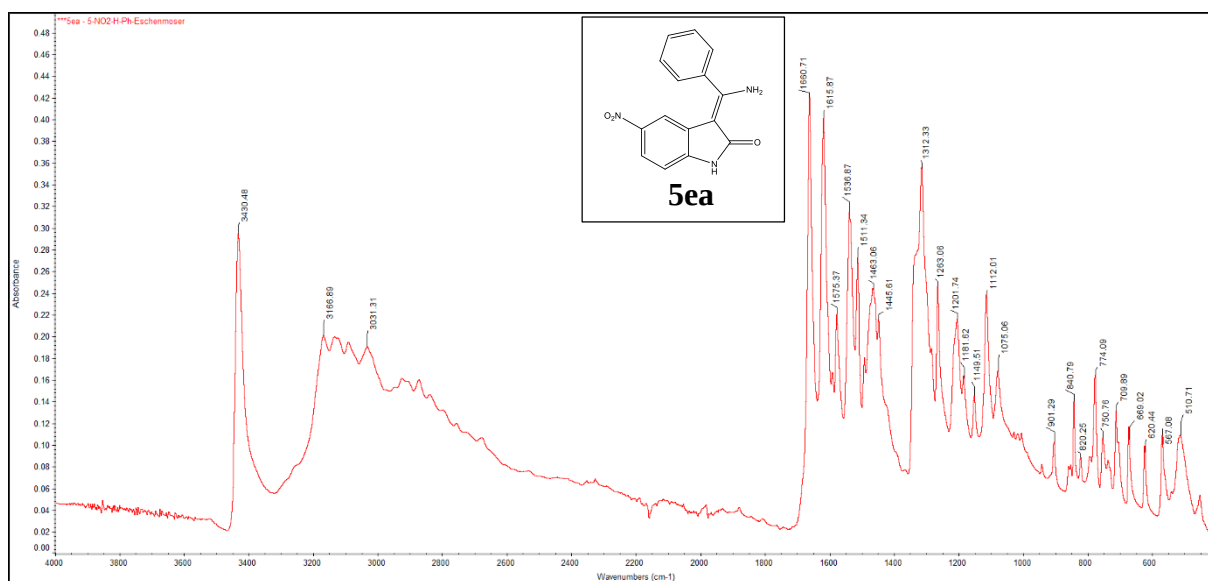


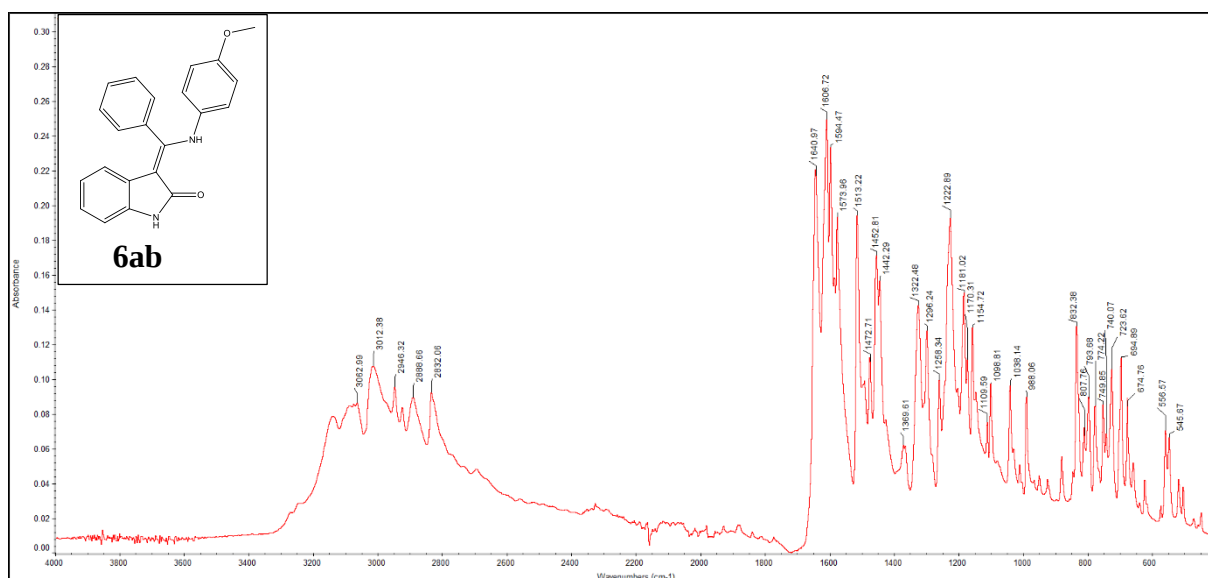
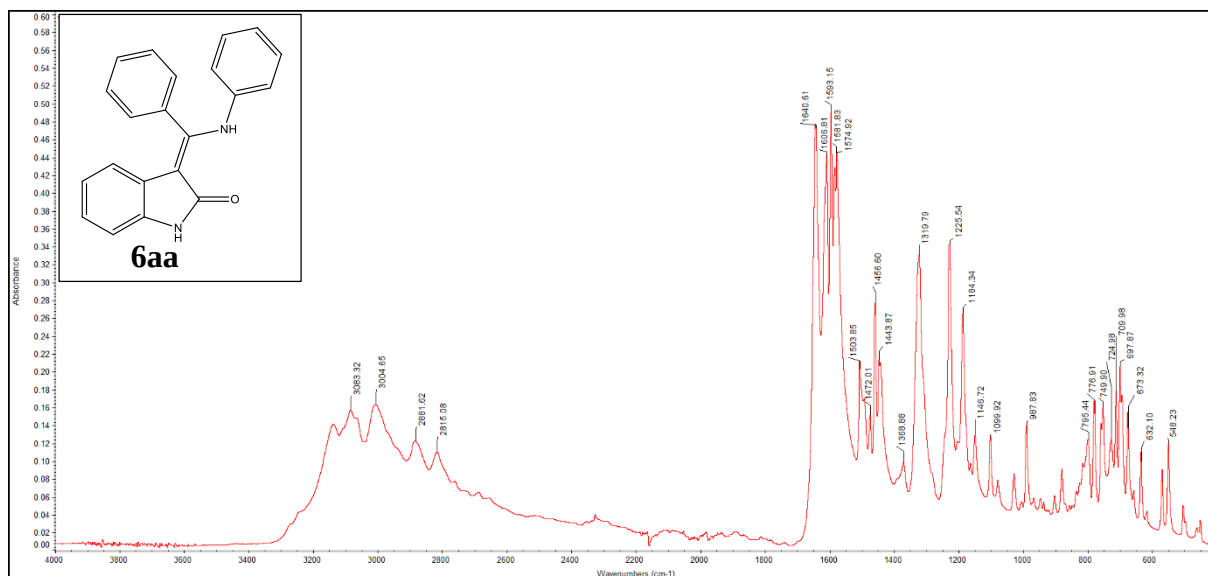
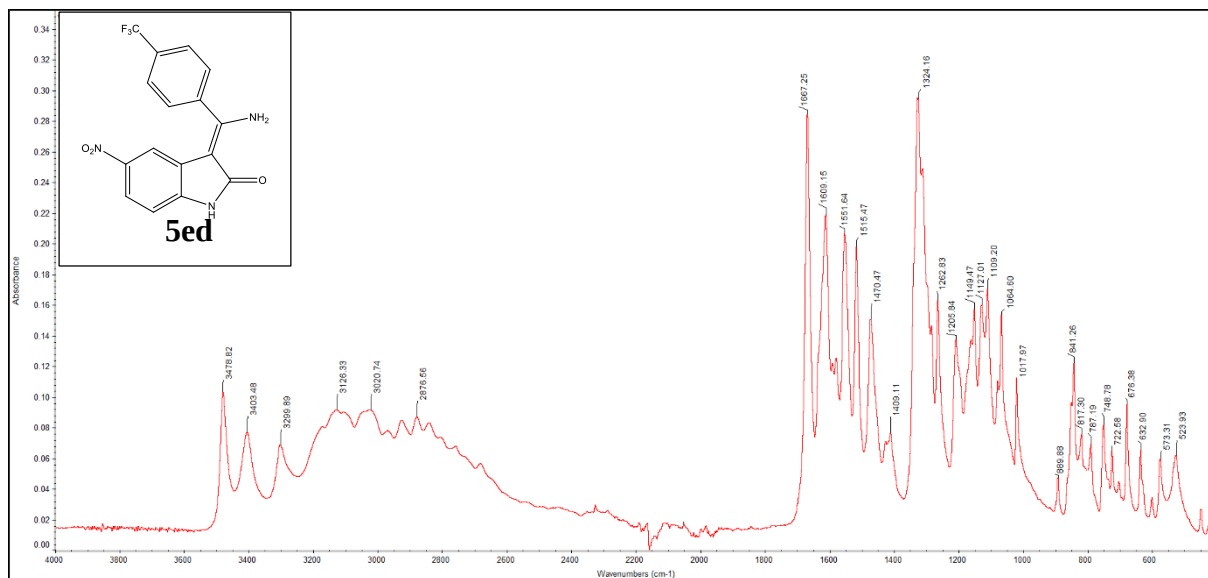


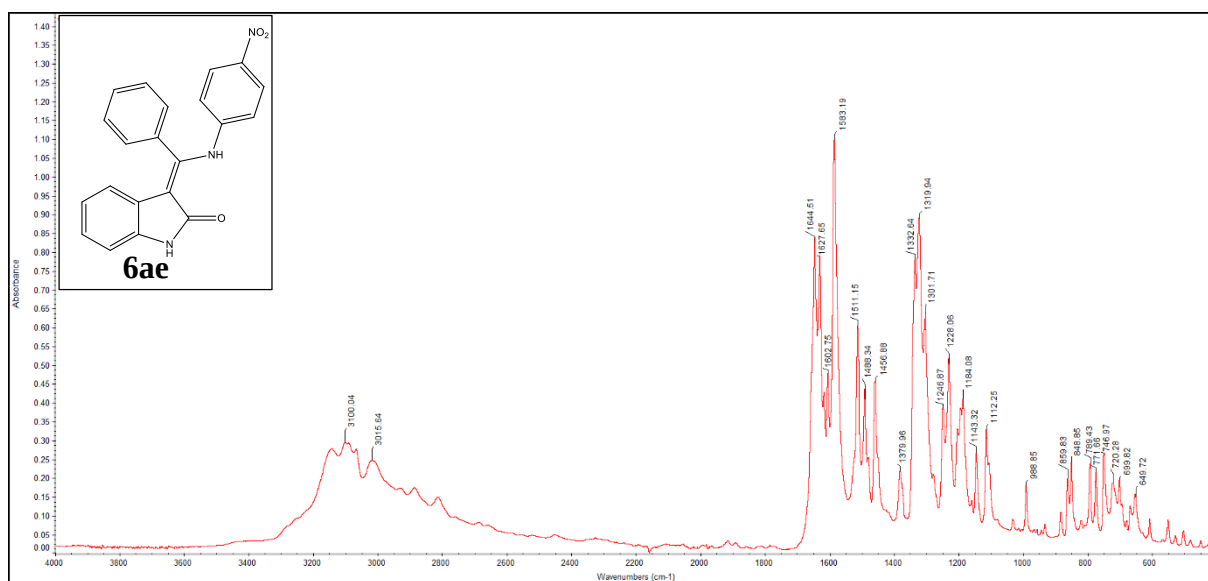
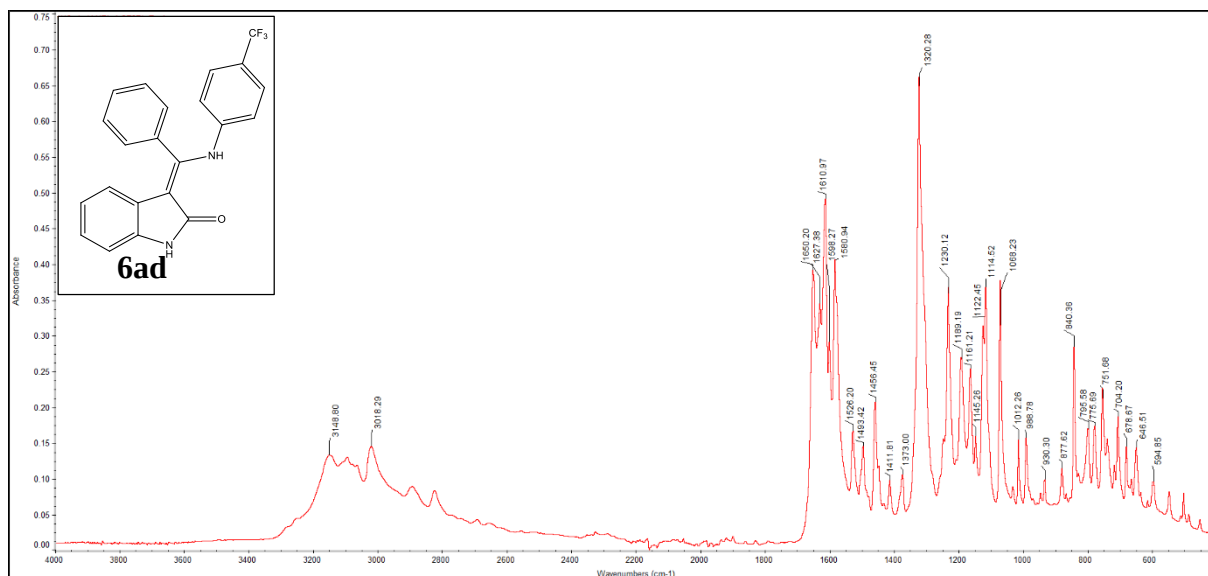
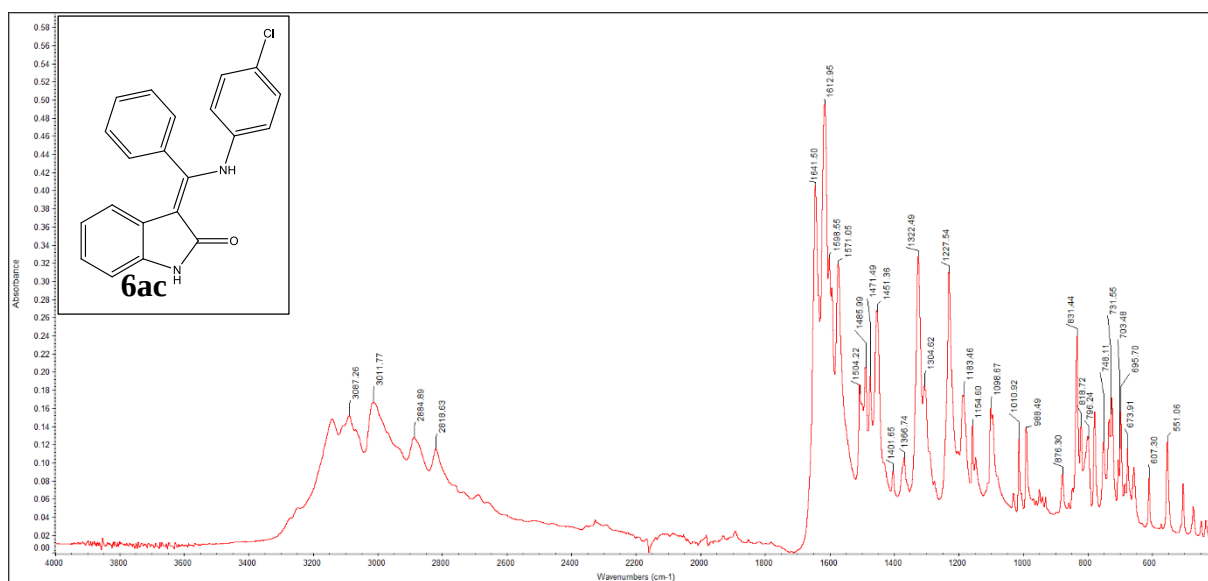


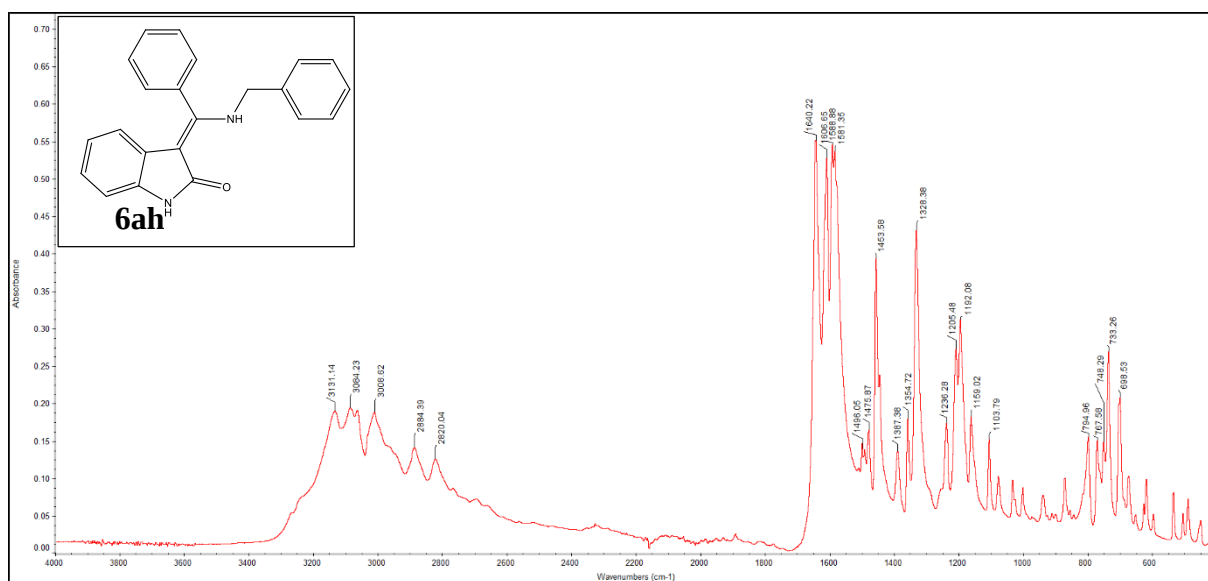
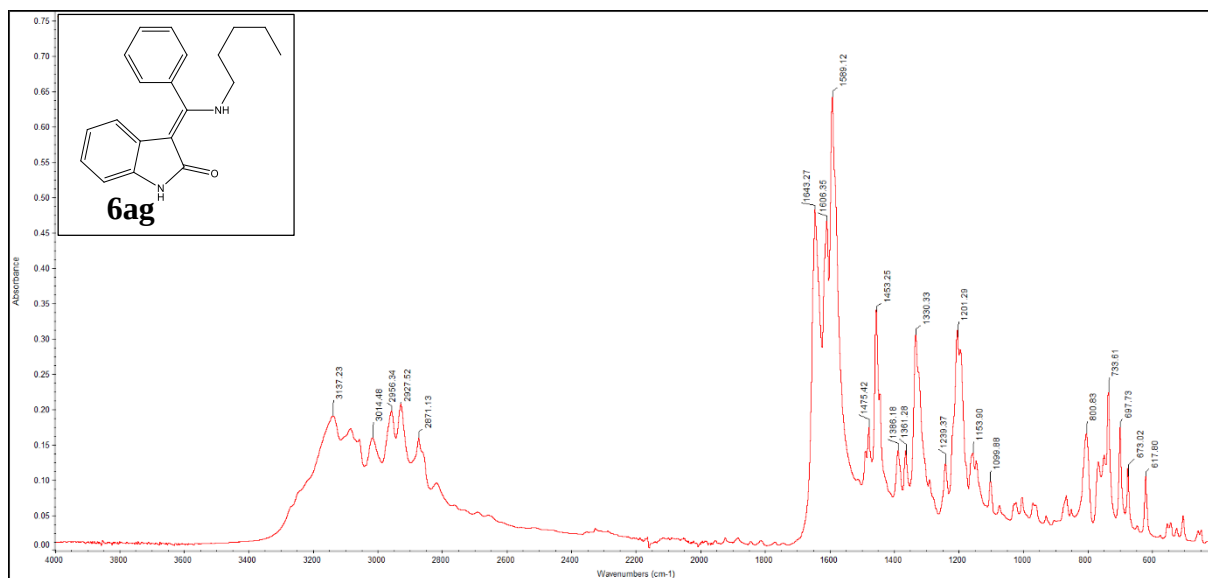
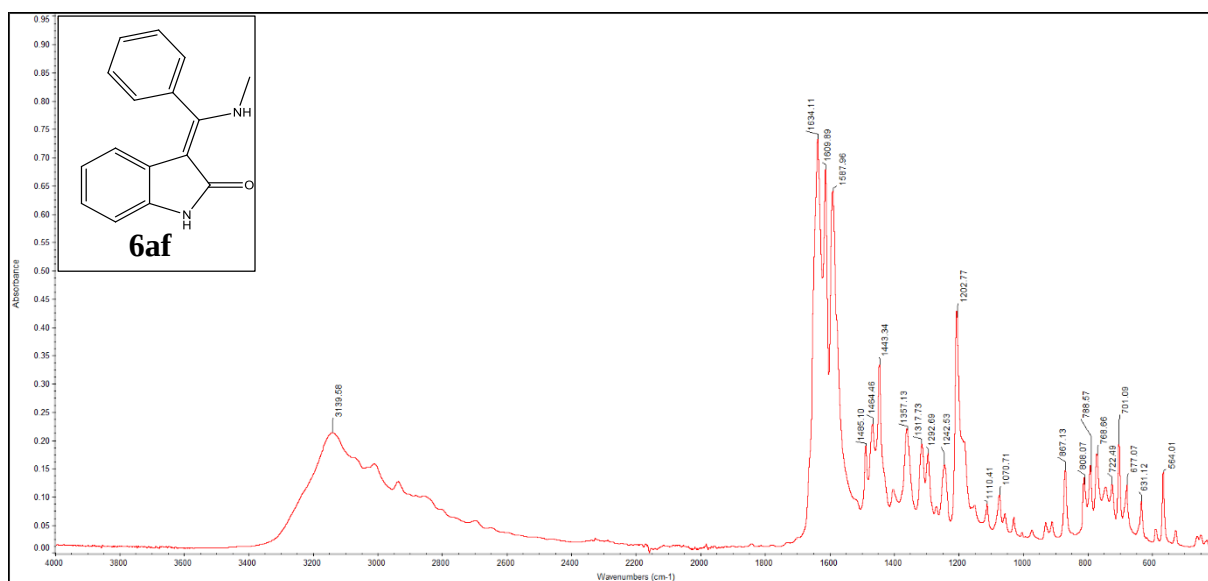


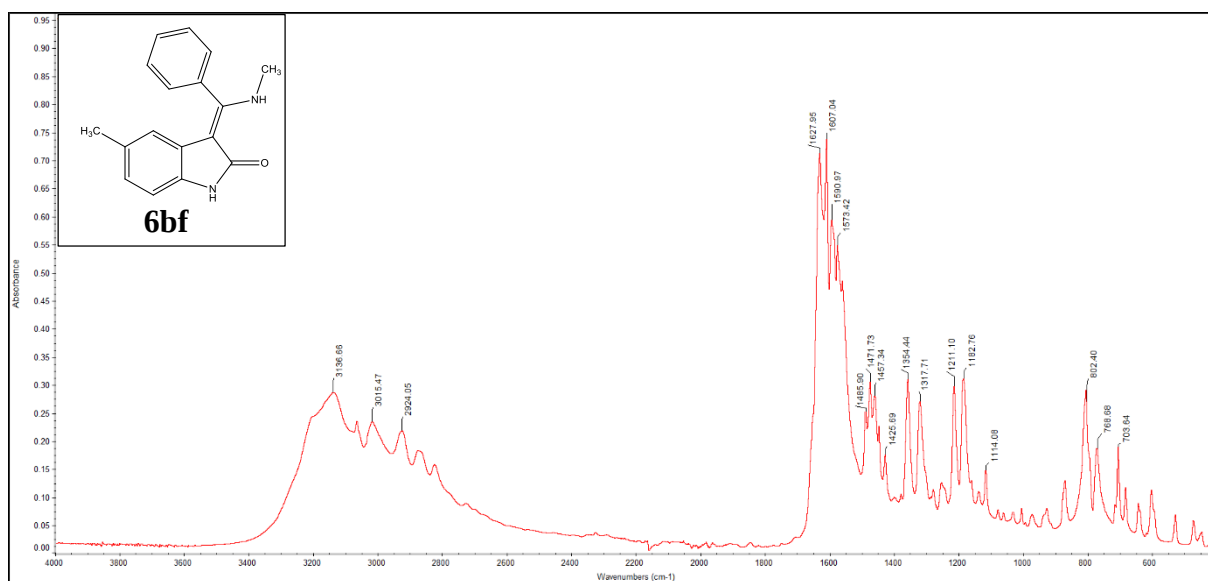
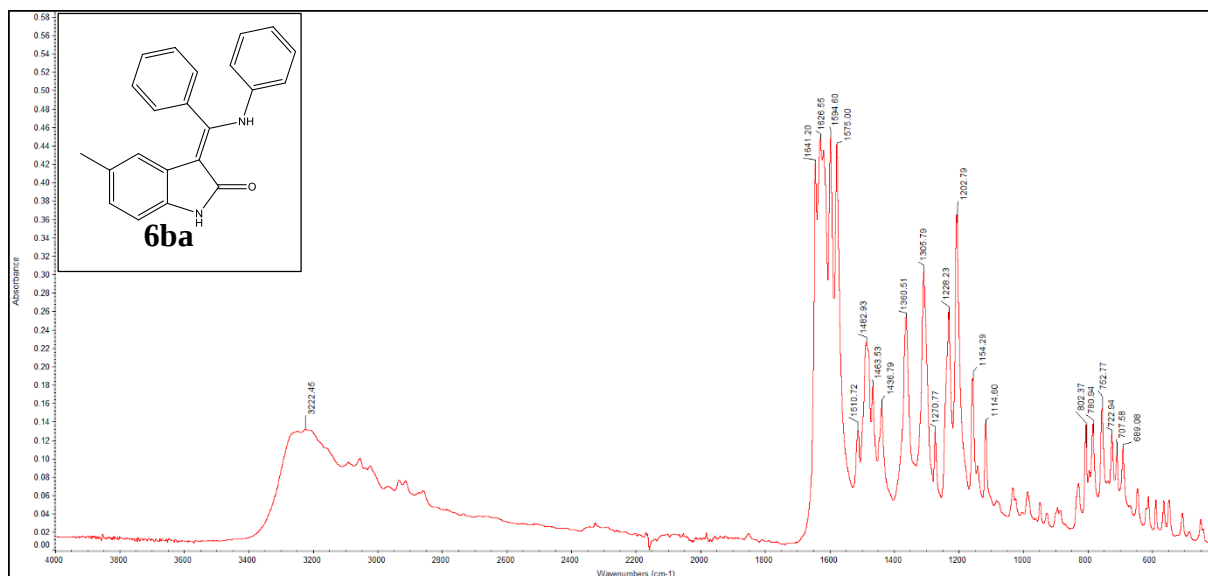
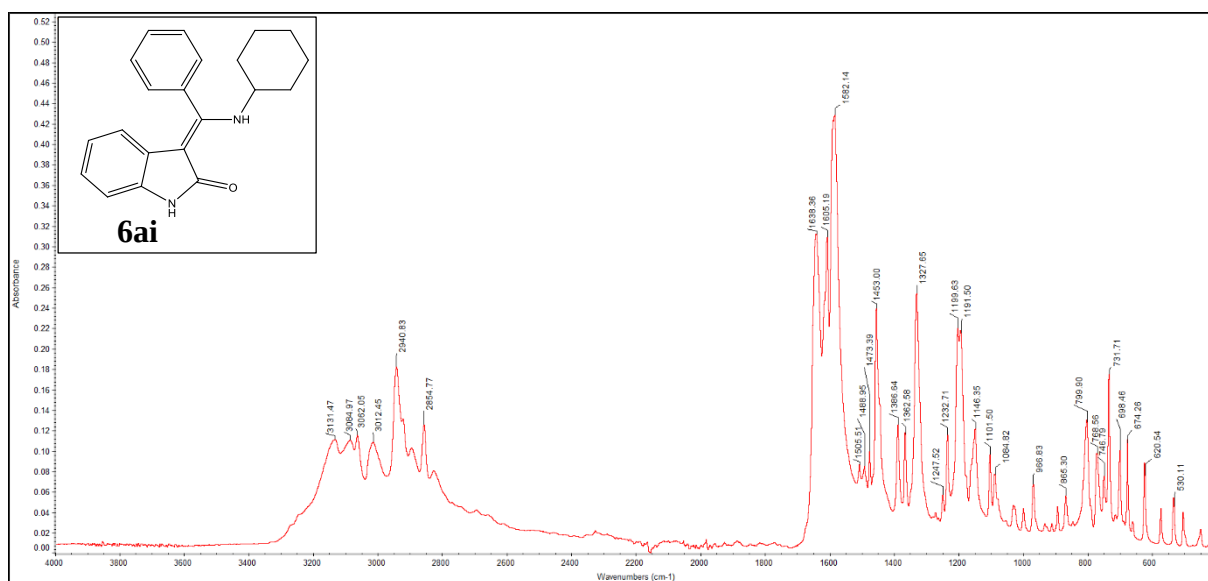


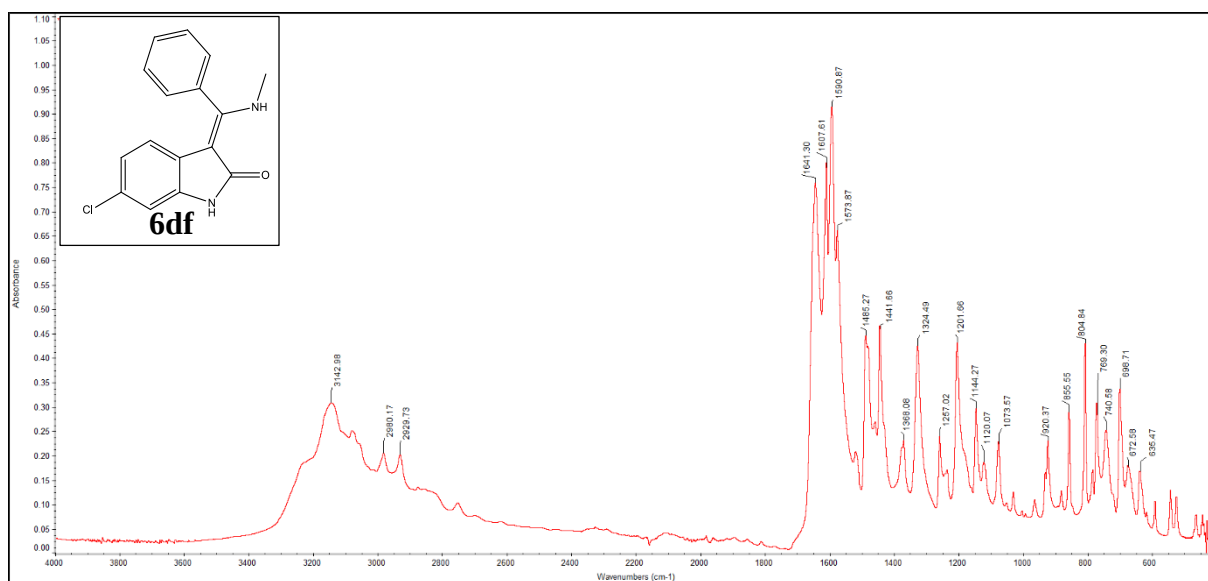
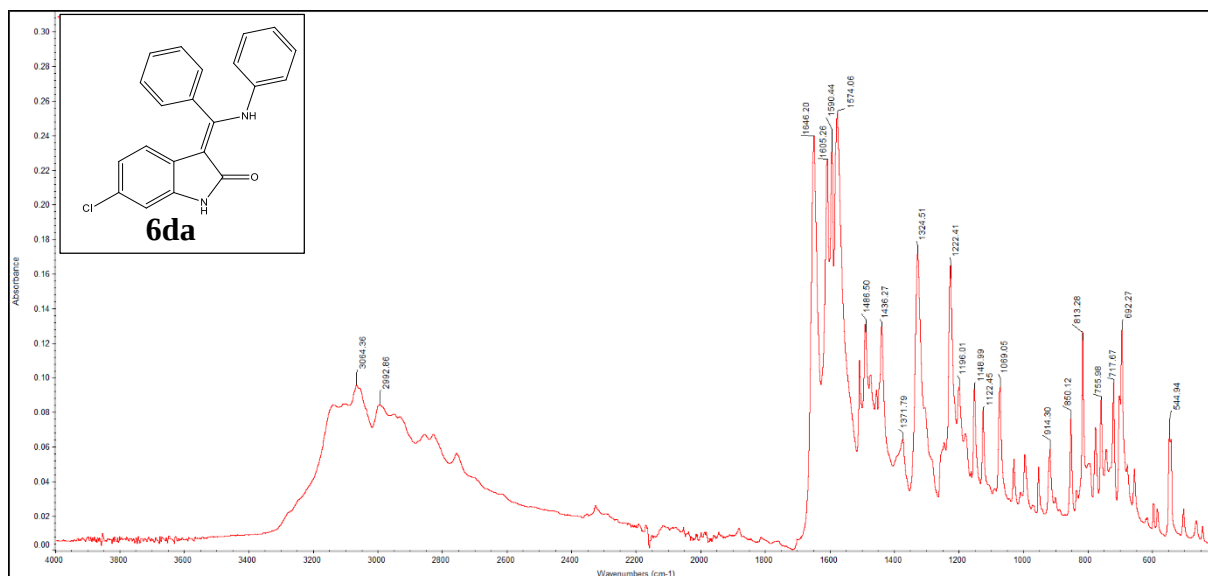
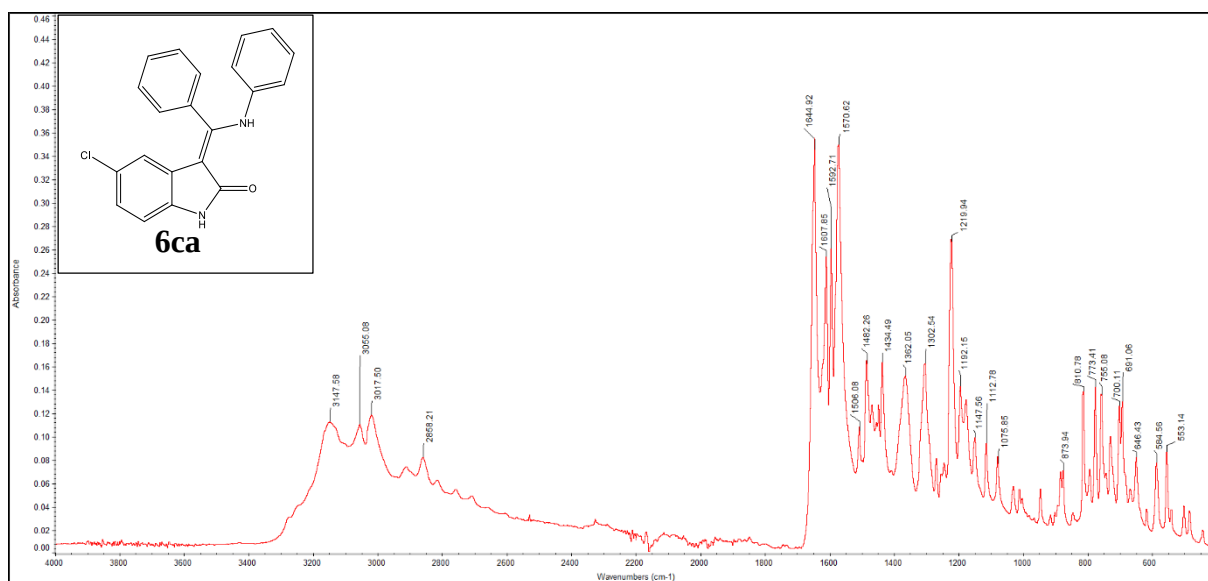


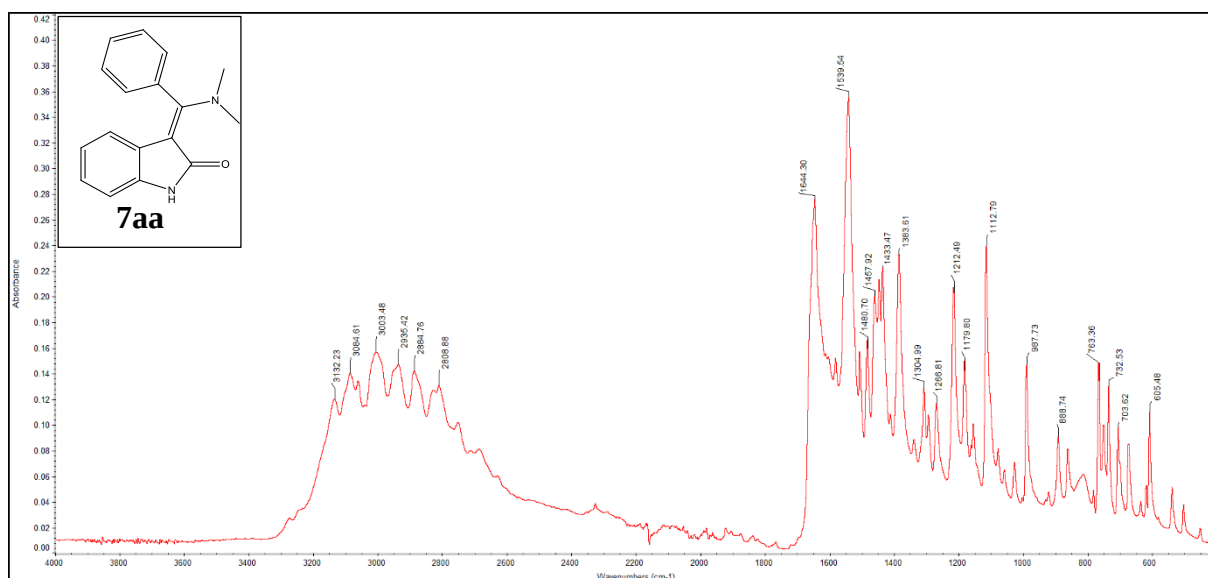
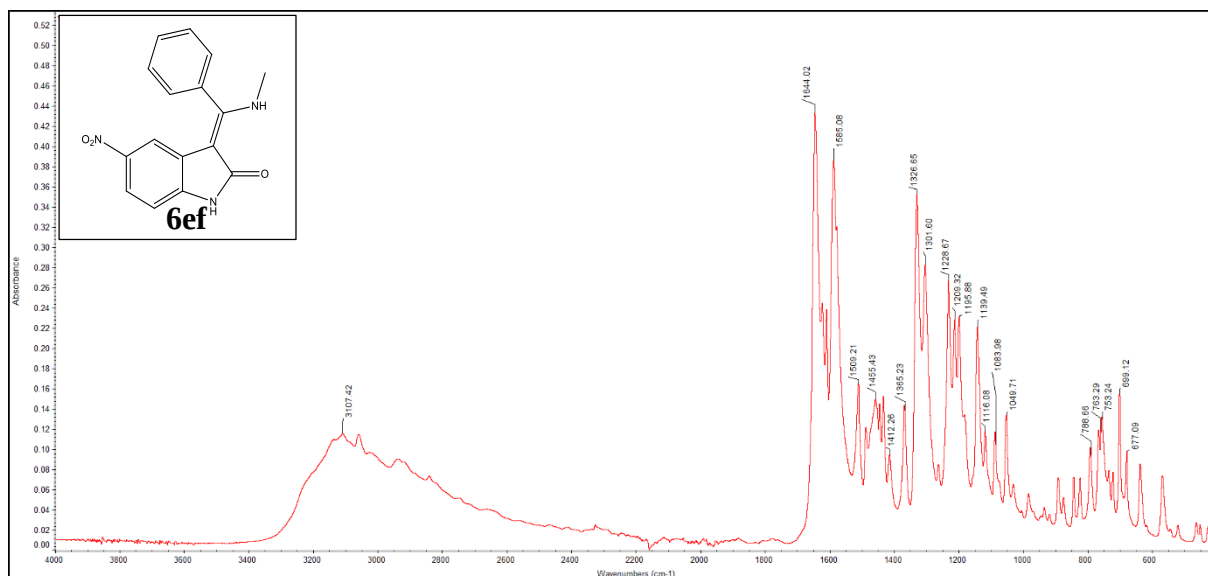
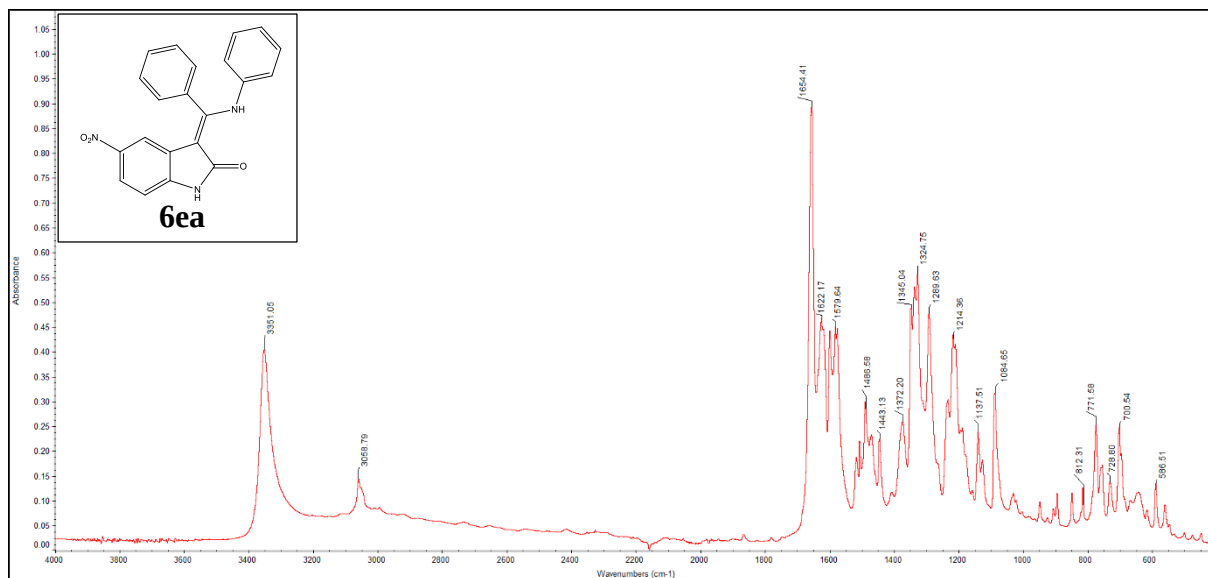


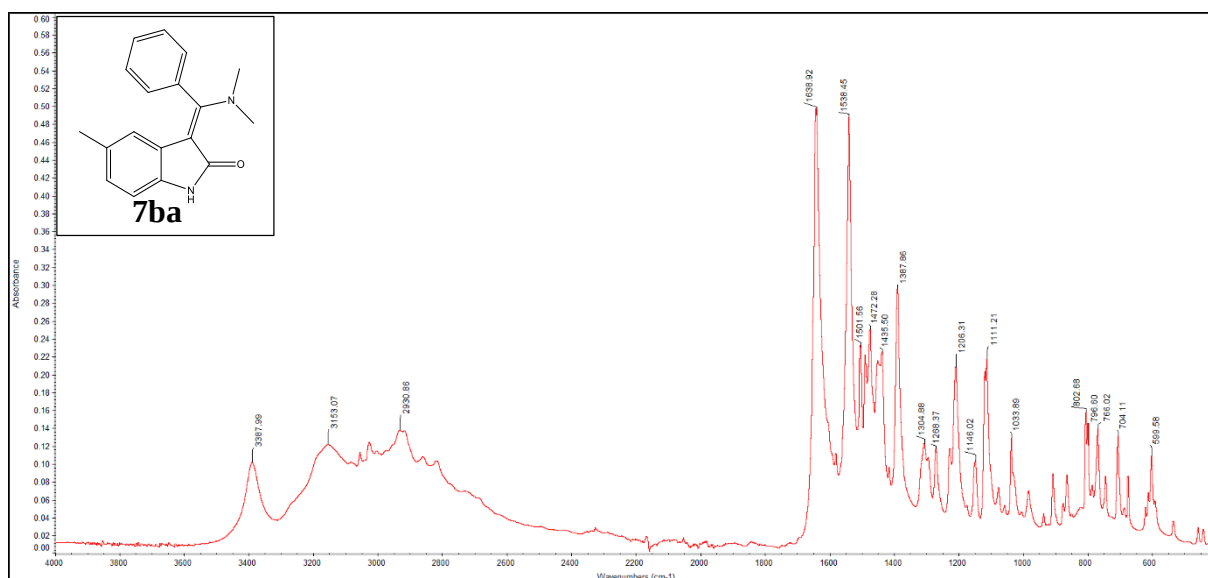
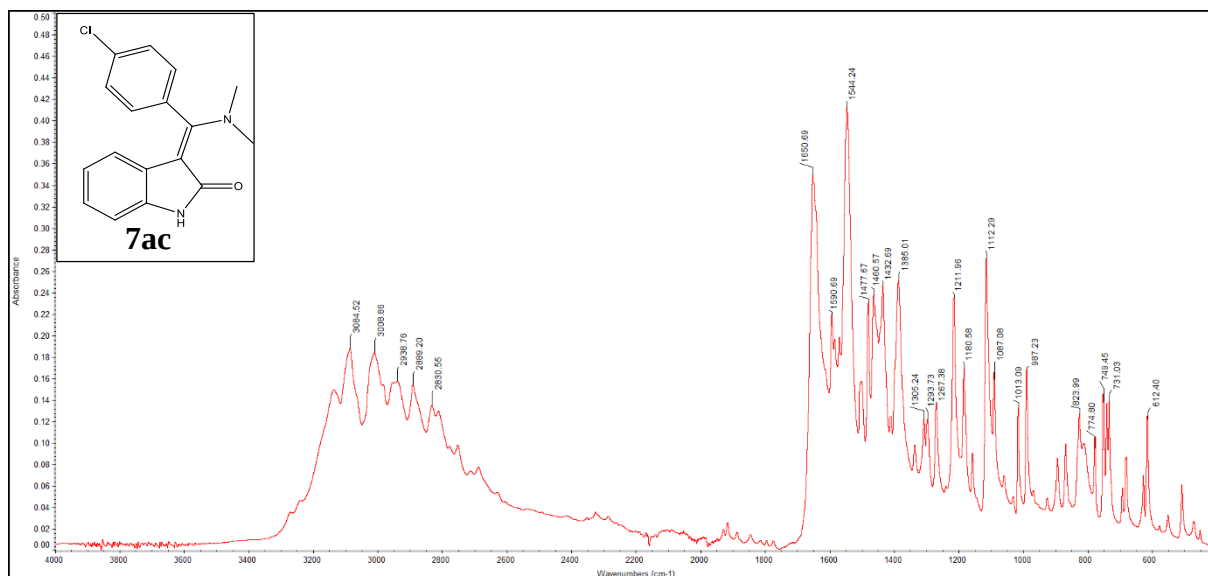
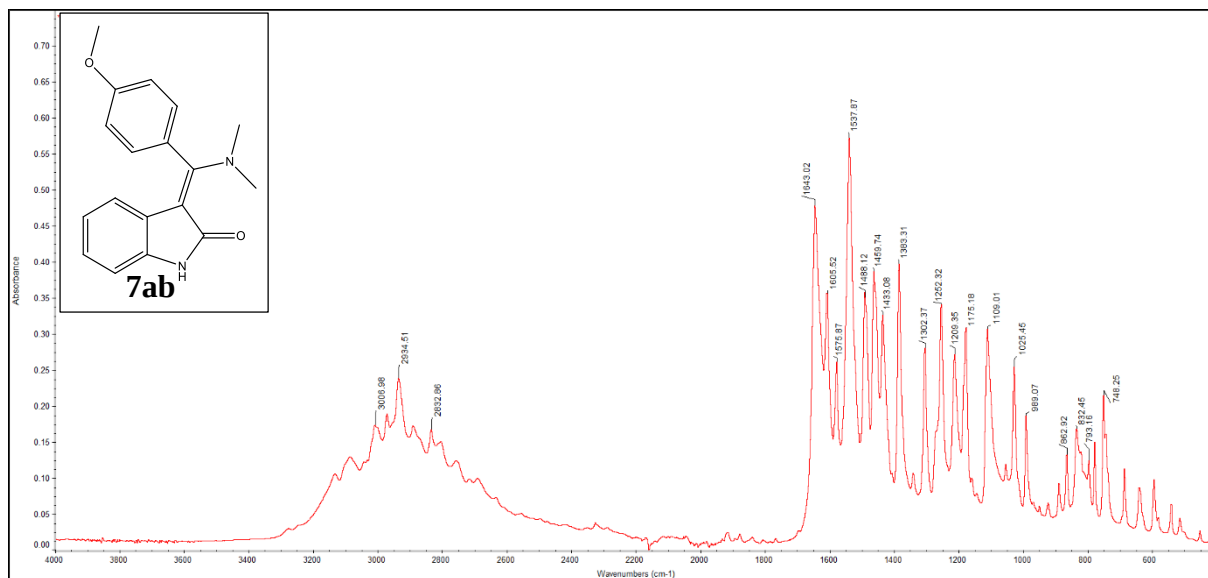


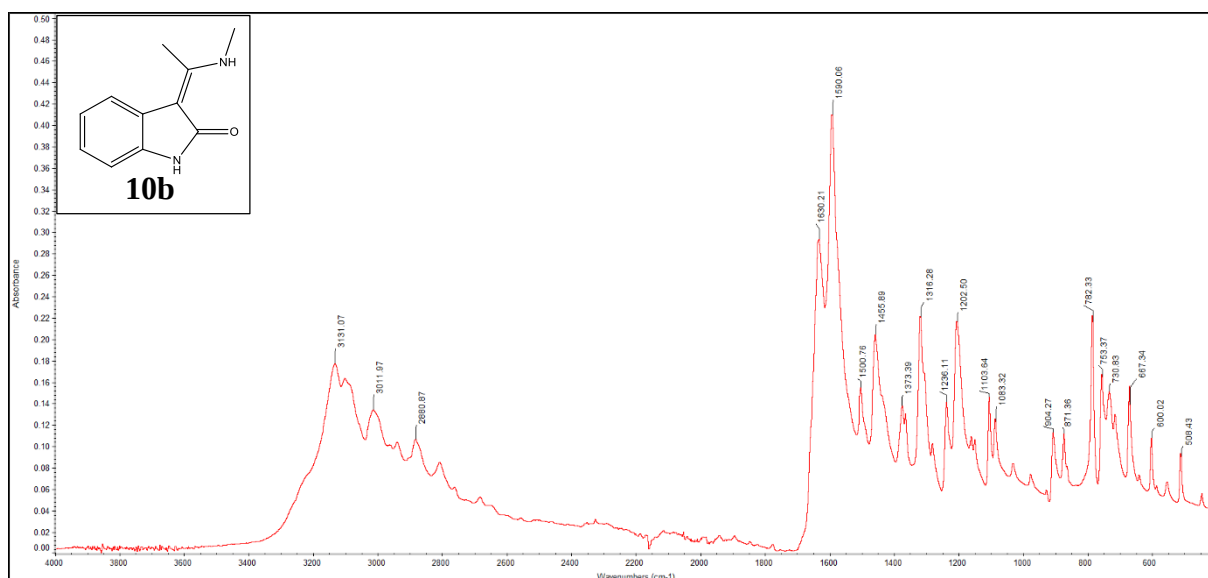
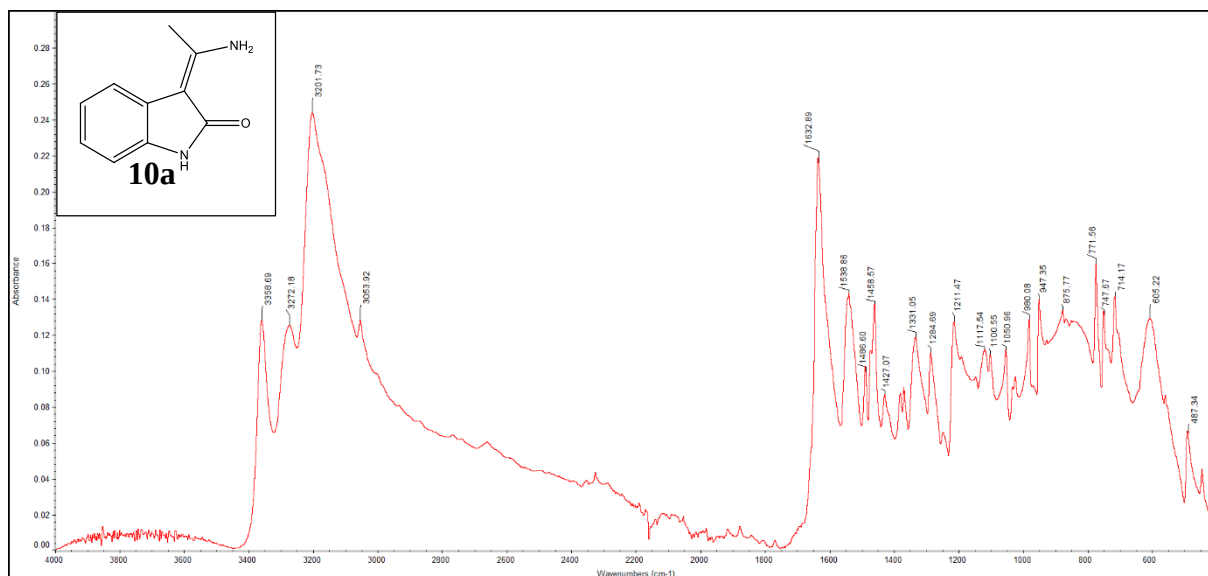
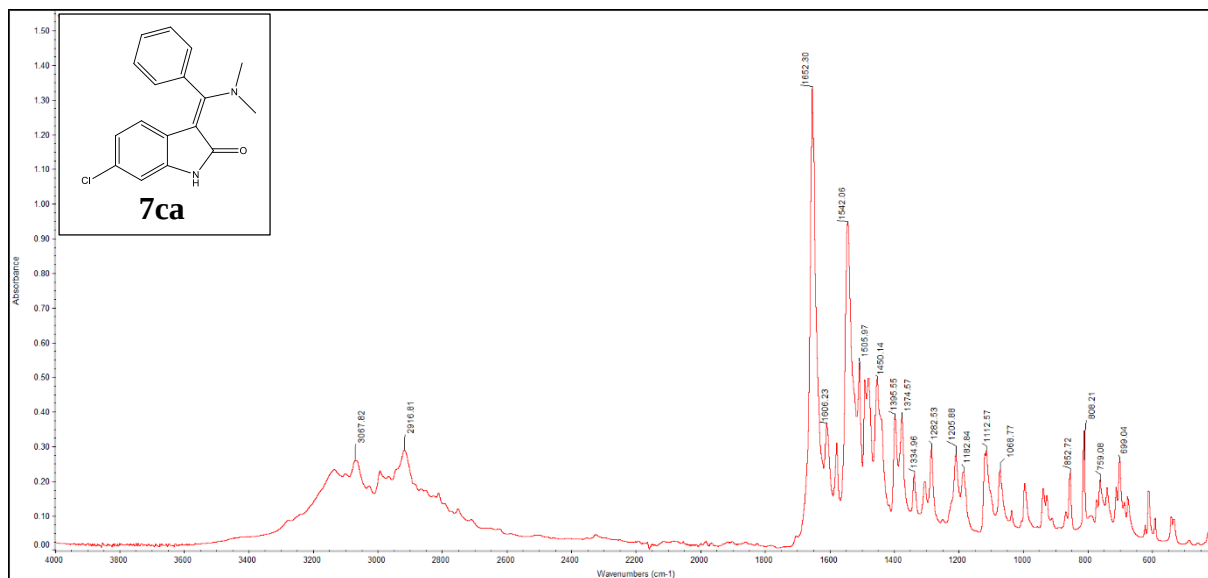


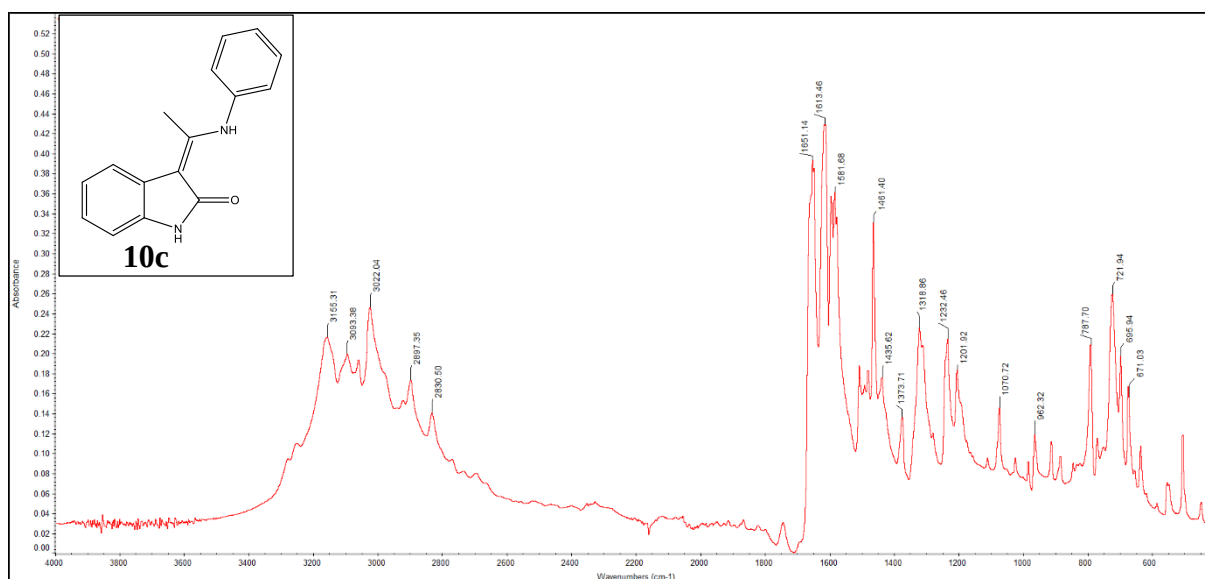












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