

Supporting Information

Palladium-Catalyzed Synthesis of 1*H*-Indenes and Phthalimides via Isocyanide Insertion

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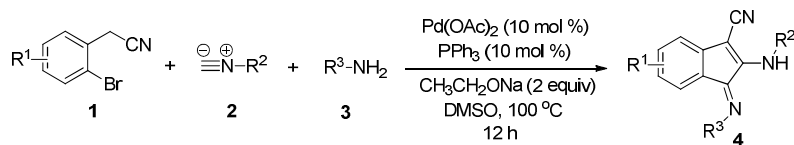
1. General Information

NMR spectra were obtained using a Bruker Avance 400 spectrometer (^1H at 400 MHz, and ^{13}C at 101 MHz). Chemical shifts for ^1H NMR spectra are reported in parts per million (ppm) from tetramethylsilane with the solvent resonance as the internal standard (CDCl_3 : δ 7.26 ppm or $\text{DMSO}-d_6$: δ 2.50 ppm). Chemical shifts for ^{13}C NMR spectra are reported in parts per million (ppm) from tetramethylsilane with the solvent as the internal standard (CDCl_3 : δ 77.0 ppm or $\text{DMSO}-d_6$: δ 39.5 ppm). IR spectra were obtained either as potassium bromide pellets or as liquid films between two potassium bromide pellets with a spectrometer. GC-MS was obtained using electron ionization. HRMS was obtained with a LCMS-IT-TOF mass spectrometer.

Materials. Unless stated otherwise, commercial reagents were used without further purification. All reagents were weighed and handled in air at room temperature.

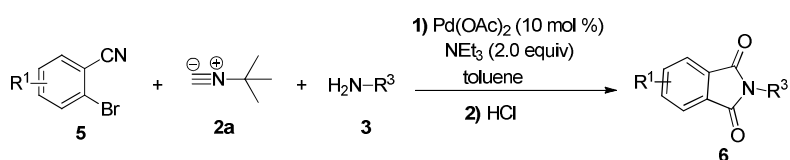
2. General Experimental Procedure: Synthesis of Compounds 4 and 6

General Procedure: Synthesis of 1*H*-indenes 4



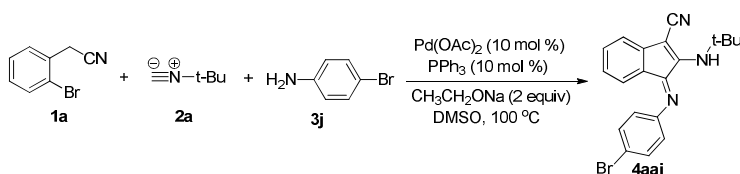
The reaction mixture of **1** (0.1 mmol), **2** (0.2 mmol), **3** (0.2 mmol), $\text{Pd}(\text{OAc})_2$ (10 mol %), PPh_3 (10 mol %), $\text{CH}_3\text{CH}_2\text{ONa}$ (2.0 equiv), DMSO (0.5 mL) at $100\text{ }^\circ\text{C}$ for 12 h, and monitored periodically by TLC. Upon completion, the reaction mixture was diluted with water (30 mL) and extracted with ethyl acetate ($3 \times 30\text{ mL}$). The combined organic layers were washed with water and brine, dried over Na_2SO_4 and filtered. The solvent was removed under vacuum. The residue was purified by flash column chromatography to afford 1*H*-indenes **4**.

General Procedure: Synthesis of phthalimides **6**



The reaction mixture of **5** (0.12 mmol), **2a** (0.15 mmol), **3** (0.1 mmol), Pd(OAc)₂ (10 mol %), NEt₃ (2.0 equiv), toluene (1 mL) at 100 °C for 12 h, followed by the addition of HCl (4 M, 0.5 mL) for 1 h, and monitored periodically by TLC. Upon completion, the reaction mixture was diluted with water (30 mL) and extracted with ethyl acetate (3 × 30 mL). The combined organic layers were washed with water and brine, dried over Na₂SO₄ and filtered. The solvent was removed under vacuum. The residue was purified by flash column chromatography to afford phthalimides **6**.

General Procedure: Procedure for Large Scale Reaction



The reaction mixture of 2-(2-bromophenyl)acetonitriles **1a** (2 mmol), *tert*-butyl isocyanide **2a** (4 mmol), 4-bromoaniline **3a** (4 mmol), Pd(OAc)₂ (10 mol %), PPh₃ (10 mol %), CH₃CH₂ONa (2.0 equiv), DMSO (5 mL) at 100 °C for 24 h, and monitored periodically by TLC. Upon completion, the reaction mixture was diluted with water (30 mL) and extracted with ethyl acetate (3 × 30 mL). The combined organic layers were washed with water and brine, dried over Na₂SO₄, and filtered. The solvent was removed under vacuum. The residue was purified by flash column chromatography to afford 1*H*-indene **4aaj**.

3. Further Modification of Product **4aaj**

(i) Hydrolysis of **4aaj**

The reaction mixture of **4aaj** (0.2 mmol), HCl (4 M, 0.5 mL), toluene (1 mL) at 100 °C for 1 h, and monitored periodically by TLC. Upon completion, the reaction mixture was diluted with water (30 mL) and extracted with ethyl acetate (3 × 30 mL). The combined organic layers were washed with water and brine, dried over Na₂SO₄, and filtered. The solvent was removed under vacuum. The residue was purified by flash column chromatography to afford *tert*-butylamino)-1-oxo-1*H*-indene-3-carbonitrile **8**.

(ii) Arylation of **4aaj**

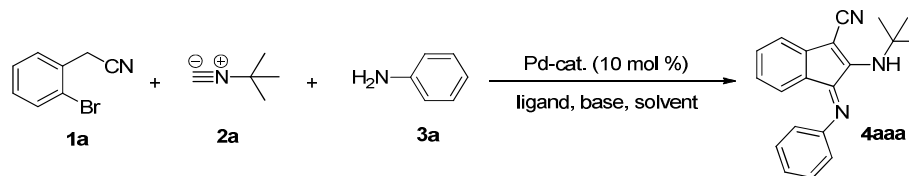
The reaction mixture of **4aaj** (0.2 mmol), phenylboronic acid (0.22 mmol), Pd(PPh₃)₄ (2.0 mol %), K₂CO₃ (1.0 mmol), DMF (1 mL), H₂O (1 mL) at 90 °C for 12 h under a N₂ atmosphere, and monitored periodically by TLC. Upon completion, the reaction mixture was diluted with water (30 mL) and extracted with ethyl acetate (3 × 30 mL). The combined organic layers were washed with water and brine, dried over Na₂SO₄, and filtered. The solvent was removed under vacuum. The residue was purified by flash column chromatography to afford product **10**.

(iii) Reduction reaction of **4aaj**

The reaction mixture of **4aaj** (0.2 mmol), NaBH₄ (2.0 equiv), THF (2 mL), RT, 2 h, and monitored periodically by TLC. Upon completion, the reaction mixture was diluted with water (30 mL) and extracted with ethyl acetate (3 × 30 mL). The combined organic layers were washed with water and brine, dried over Na₂SO₄, and filtered. The solvent was removed under vacuum. The residue was purified by flash column chromatography to afford product **11**.

4. Screening of the Reaction Conditions S1 and S2

Table S1. Optimization of reaction conditions for 1*H*-indene 4aaa^a



entry	catalyst	ligand	base	solvent	yield (%) ^b
1	Pd(OAc) ₂	PPh ₃	NEt ₃	DMSO	77
2	-	PPh ₃	NEt ₃	DMSO	ND
3	Pd(OAc) ₂	-	NEt ₃	DMSO	trace
4	Pd(OAc) ₂	PPh ₃	-	DMSO	ND
5	Pd(OAc) ₂	PPh ₃	NEt ₃	DMA	37
6	Pd(OAc) ₂	PPh ₃	NEt ₃	DMF	21
7	Pd(OAc) ₂	PPh ₃	NEt ₃	toluene	trace
8	Pd(OAc) ₂	PPh ₃	NEt ₃	dioxane	ND
9	PdCl ₂	PPh ₃	NEt ₃	DMSO	27
10	PdBr ₂	PPh ₃	NEt ₃	DMSO	17
11	PdCl ₂ (PPh ₃) ₂	PPh ₃	NEt ₃	DMSO	38
12	Pd(OAc) ₂	PPh ₃	Cs ₂ CO ₃	DMSO	ND
13	Pd(OAc) ₂	PPh ₃	CsF	DMSO	trace
14	Pd(OAc) ₂	PPh ₃	KPF ₆	DMSO	ND
15	Pd(OAc) ₂	PPh ₃	CH ₃ ONa	DMSO	82
16	Pd(OAc) ₂	PPh ₃	CH ₃ COONa	DMSO	17
17	Pd(OAc) ₂	PPh ₃	<i>t</i> -BuOK	DMSO	88
18	Pd(OAc) ₂	PPh ₃	CH ₃ CH ₂ ONa	DMSO	92 (90)

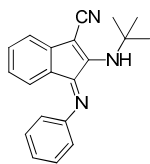
^aAll reactions were carried out under the following conditions: **1a** (0.1 mmol), **2a** (0.2 mmol), **3a** (0.2 mmol), Pd catalyst (10 mol %), ligand (10 mol %), base (2.0 equiv), solvents (0.5 mL) at 100 °C for 12 h. ^bYields determined by GC based on **1a** with n-dodecane as internal standard. Isolated yield is in the parentheses. ND = not detected.

Table S2. Optimization of reaction conditions for phthalimide 6aaa^a

entry	catalyst	ligand	base	solvent	yield (%) ^b
1	Pd(OAc) ₂	PPh ₃	NEt ₃	1,4-dioxane	21
2	-	PPh ₃	NEt ₃	1,4-dioxane	ND
3	Pd(OAc) ₂	-	NEt ₃	1,4-dioxane	19
4	Pd(OAc) ₂	PPh ₃	-	1,4-dioxane	ND
5	Pd(OAc) ₂	-	NEt ₃	DMSO	ND
6	Pd(OAc) ₂	-	NEt ₃	DMF	ND
7	Pd(OAc) ₂	-	NEt ₃	PhCl	trace
8	Pd(OAc) ₂	-	NEt ₃	toluene	52
9	Pd(OAc) ₂	-	<i>t</i> -BuOK	toluene	trace
10	Pd(OAc) ₂	-	K ₂ CO ₃	toluene	trace
11	Pd(OAc) ₂	-	CH ₃ ONa	toluene	32
12	Pd(OAc) ₂	-	Cs ₂ CO ₃	toluene	28
13	Pd(OAc) ₂	-	CsF	toluene	41
14	PdCl ₂	-	NEt ₃	toluene	29
15	Pd(dba) ₂	-	NEt ₃	toluene	trace
16	PdBr ₂	-	NEt ₃	toluene	25
17	PdCl ₂ (PPh ₃) ₂	-	NEt ₃	toluene	14
18 ^c	Pd(OAc) ₂	-	NEt ₃	toluene	61
19 ^d	Pd(OAc) ₂	-	NEt ₃	toluene	72 (69)

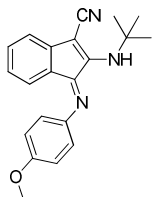
^aAll reactions were carried out under the following conditions: **5a** (0.12 mmol), **2a** (0.15 mmol), **3a** (0.1 mmol), Pd catalyst (10 mol %), ligand (20 mol %), base (2.0 equiv), solvents (1 mL) at 80 °C for 12 h, followed by the addition of HCl (4 M, 0.5 mL) for 1 h. ^bYields determined by GC based on **3a** with n-dodecane as internal standard. Isolated yield is in the parentheses. ND = not detected. ^c90 °C. ^d100 °C.

5. Analytical Data



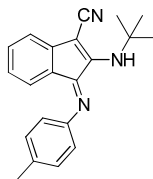
(E)-2-(tert-Butylamino)-1-(phenylimino)-1H-indene-3-carbonitrile (4aaa):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 90% (27.1 mg) as red liquid. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.44 (t, $J = 7.8$ Hz, 2H), 7.27 (dd, $J = 12.5$, 5.1 Hz, 1H), 7.20 (t, $J = 7.5$ Hz, 1H), 7.08 (d, $J = 7.5$ Hz, 1H), 6.96 (d, $J = 7.6$ Hz, 2H), 6.60 (t, $J = 7.6$ Hz, 1H), 6.52 (s, 1H), 6.39 (d, $J = 7.5$ Hz, 1H), 1.62 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 160.9, 149.3, 148.0, 147.3, 133.7, 129.5, 125.7, 125.2, 123.3, 122.2, 118.9, 118.2, 118.0, 77.1, 52.5, 30.1. **IR** (KBr, cm^{-1}): 3359, 2924, 2194, 1613, 1459, 1375, 1250, 1155, 1025, 754, 581, 497. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{19}\text{N}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 324.1471; found 324.1476.



(E)-2-(tert-Butylamino)-1-((4-methoxyphenyl)imino)-1H-indene-3-carbonitrile (4aab):

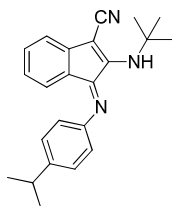
Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 82% (27.2 mg) as a red solid: 132-134 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.21-7.14 (m, 1H), 7.05 (d, $J = 7.5$ Hz, 1H), 6.99-6.87 (m, 4H), 6.65-6.55 (m, 2H), 6.51 (s, 1H), 3.86 (s, 3H), 1.58 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 160.6, 157.5, 148.2, 147.3, 142.0, 133.5, 125.3, 123.3, 122.2, 120.1, 119.0, 118.0, 114.7, 76.7, 55.5, 52.4, 30.1. **IR** (KBr, cm^{-1}): 3447, 2925, 2193, 1606, 1455, 1373, 1236, 1168, 1030, 829, 752, 570. **HRMS** (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{21}\text{N}_3\text{ONa}$ $[\text{M} + \text{Na}]^+$ 354.1577; found 354.1582.



(E)-2-(tert-Butylamino)-1-(p-tolylimino)-1H-indene-3-carbonitrile (4aac):

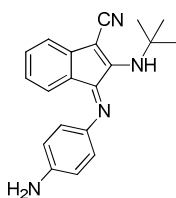
Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 89% (28.0 mg) as a red solid: 160-161 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.25-7.13 (m, 3H), 7.08-7.03 (m, 1H), 6.84 (d, $J = 8.2$ Hz, 2H), 6.59 (t, $J = 7.4$ Hz, 1H), 6.48 (t, $J = 5.8$ Hz, 2H), 2.40 (s, 3H), 1.59 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 160.7, 148.1, 147.3, 146.6, 134.9, 133.5, 130.0, 125.6, 123.2, 122.3,

119.0, 118.3, 118.0, 77.2, 52.4, 30.1, 21.0. **IR** (KBr, cm^{-1}): 3332, 2922, 2194, 1607, 1458, 1371, 1214, 1096, 750, 570, 498. **HRMS** (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{21}\text{N}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 338.1628; found 338.1629.



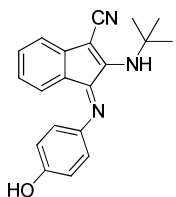
(E)-2-(tert-Butylamino)-1-(p-tolylimino)-1H-indene-3-carbonitrile (4aad):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 87% (30.1 mg) as red liquid. **^1H NMR** (400 MHz, CDCl_3 , δ ppm) 7.26 (d, $J = 7.8$ Hz, 2H), 7.17 (t, $J = 7.5$ Hz, 1H), 7.05 (d, $J = 7.5$ Hz, 1H), 6.87 (d, $J = 7.7$ Hz, 2H), 6.59 (t, $J = 7.5$ Hz, 1H), 6.50 (s, 1H), 6.48 (d, $J = 7.6$ Hz, 1H), 3.01 – 2.89 (m, 1H), 1.58 (s, 9H), 1.30 (s, 3H), 1.28 (s, 3H); **^{13}C NMR** (101 MHz, CDCl_3 , δ ppm) 160.5, 148.1, 147.3, 146.7, 146.1, 133.5, 127.4, 125.5, 123.3, 122.3, 119.0, 118.3, 118.0, 76.8, 52.4, 33.7, 30.1, 24.2. **IR** (KBr, cm^{-1}): 3317, 2963, 2193, 1606, 1456, 1370, 1292, 1210, 1096, 840, 752, 708, 575, 496. **HRMS** (ESI) m/z : calcd for $\text{C}_{23}\text{H}_{25}\text{N}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 366.1941; found 366.1944.



(E)-1-((4-Aminophenyl)imino)-2-(tert-butylamino)-1H-indene-3-carbonitrile

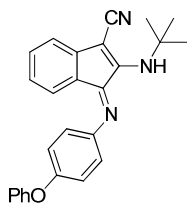
(4aae): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 83% (26.2 mg) as a red solid: 150-152 °C. **^1H NMR** (400 MHz, CDCl_3 , δ ppm) 7.17 (t, $J = 7.5$ Hz, 1H), 7.05 (d, $J = 7.7$ Hz, 1H), 6.83 (t, $J = 8.5$ Hz, 3H), 6.73 (d, $J = 8.3$ Hz, 2H), 6.62 (t, $J = 7.6$ Hz, 1H), 6.54 (s, 1H), 3.75 (s, 2H), 1.57 (s, 9H); **^{13}C NMR** (101 MHz, CDCl_3 , δ ppm) 159.6, 148.5, 147.2, 144.5, 140.0, 133.2, 125.1, 123.2, 122.3, 120.7, 119.3, 117.9, 115.7, 76.4, 52.3, 30.1. **IR** (KBr, cm^{-1}): 3362, 2923, 2192, 1607, 1457, 1371, 1292, 1211, 1096, 835, 750, 580, 496. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{21}\text{N}_4$ $[\text{M} + \text{H}]^+$ 317.1761; found 317.1759.



(E)-2-(tert-Butylamino)-1-((4-hydroxyphenyl)imino)-1H-indene-3-carbonitrile

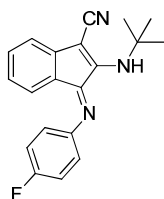
(4aaf): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding

86% (27.3 mg) as a red solid: 167-169 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.20 (t, $J = 7.4$ Hz, 1H), 7.07 (d, $J = 7.5$ Hz, 1H), 6.99-6.93 (m, 2H), 6.88 (d, $J = 8.3$ Hz, 2H), 6.71-6.56 (m, 3H), 6.35 (s, 1H), 1.60 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 160.5, 154.1, 148.6, 147.1, 141.7, 133.5, 125.4, 123.3, 122.2, 120.2, 119.3, 118.0, 116.3, 76.8, 52.5, 30.0. **IR** (KBr, cm^{-1}): 3303, 2928, 2194, 1605, 1513, 1456, 1368, 1218, 839, 752. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{19}\text{N}_3\text{ONa}$ [$\text{M} + \text{Na}$] $^+$ 340.1420; found 340.1422.



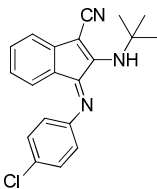
(E)-2-(tert-Butylamino)-1-((4-phenoxyphenyl)imino)-1H-indene-3-carbonitrile

(4aag): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 73% (28.7 mg) as a red solid: 180-182 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.38 (t, $J = 7.9$ Hz, 2H), 7.21 (t, $J = 7.3$ Hz, 1H), 7.14 (t, $J = 7.4$ Hz, 1H), 7.08 (t, $J = 7.5$ Hz, 5H), 6.96 (d, $J = 8.6$ Hz, 2H), 6.68-6.59 (m, 2H), 6.52 (s, 1H), 1.60 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 161.1, 157.3, 154.8, 148.1, 147.4, 144.4, 133.8, 129.9, 125.5, 123.4, 123.3, 122.2, 120.1, 120.0, 118.9, 118.7, 118.1, 76.9, 52.5, 30.1. **IR** (KBr, cm^{-1}): 3318, 2925, 2192, 1602, 1487, 1370, 1290, 1232, 1095, 1012, 856, 750, 697, 569, 493. **HRMS** (ESI) m/z : calcd for $\text{C}_{26}\text{H}_{24}\text{N}_3\text{O}$ [$\text{M} + \text{H}$] $^+$ 394.1914; found 394.1911.



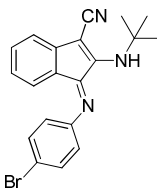
(E)-2-(tert-Butylamino)-1-((4-fluorophenyl)imino)-1H-indene-3-carbonitrile

(4aah): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 81% (25.8 mg) as a red solid: 127-129 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.19 (t, $J = 7.5$ Hz, 1H), 7.12 (t, $J = 8.5$ Hz, 2H), 7.05 (d, $J = 7.5$ Hz, 1H), 6.91 (dd, $J = 8.3, 4.9$ Hz, 2H), 6.61 (t, $J = 7.5$ Hz, 1H), 6.47 (s, 1H), 6.43 (d, $J = 7.5$ Hz, 1H), 1.59 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 161.6, 160.4 (d, $J = 242.8$ Hz), 159.2, 147.9, 147.4, 145.1 (d, $J = 3.0$ Hz), 133.9, 125.5, 123.4, 122.1, 119.9 (d, $J = 8.0$ Hz), 118.8, 118.2, 116.3 (d, $J = 2.3$ Hz), 77.1, 52.5, 30.1. **IR** (KBr, cm^{-1}): 3320, 2972, 2192, 1607, 1505, 1456, 1358, 1293, 1215, 1089, 1010, 840, 751, 571, 491. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{18}\text{N}_3\text{FNa}$ [$\text{M} + \text{Na}$] $^+$ 342.1377; found 342.1383.



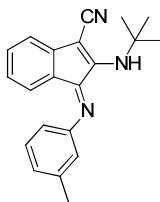
(E)-2-(tert-Butylamino)-1-((4-chlorophenyl)imino)-1H-indene-3-carbonitrile (4aai):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 78% (26.1 mg) as a red solid: 170-172 °C. ¹H NMR (400 MHz, CDCl₃, δ ppm) 7.42-7.35 (m, 2H), 7.21-7.17 (m, 1H), 7.05 (d, *J* = 7.5 Hz, 1H), 6.92-6.85 (m, 2H), 6.63-6.59 (m, 1H), 6.43-6.41 (m, 2H), 1.58 (s, 9H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 161.5, 147.7, 147.6, 147.4, 134.0, 130.6, 129.6, 125.6, 123.4, 122.0, 119.8, 118.7, 118.2, 77.5, 52.5, 30.1. IR (KBr, cm⁻¹): 3317, 2925, 2193, 1611, 1522, 1462, 1371, 1218, 1095, 1014, 836, 749, 569. HRMS (ESI) *m/z*: calcd for C₂₀H₁₈N₃CINa [M + Na]⁺ 358.1081; found 358.1084.



(E)-1-((4-Bromophenyl)imino)-2-(tert-butylamino)-1H-indene-3-carbonitrile (4aaj):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 74% (28.0 mg) as a red solid: 172-173 °C. ¹H NMR (400 MHz, CDCl₃, δ ppm) 7.53 (t, *J* = 5.6 Hz, 2H), 7.19 (t, *J* = 7.5 Hz, 1H), 7.04 (d, *J* = 7.5 Hz, 1H), 6.83 (t, *J* = 5.5 Hz, 2H), 6.61 (t, *J* = 7.5 Hz, 1H), 6.43 (s, 1H), 6.42 (d, *J* = 7.2 Hz, 1H), 1.58 (s, 9H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 161.5, 148.1, 147.8, 147.4, 134.0, 132.6, 125.7, 123.5, 122.0, 120.2, 118.7, 118.3, 118.2, 77.6, 52.5, 30.0. IR (KBr, cm⁻¹): 3322, 2974, 2192, 1609, 1523, 1460, 1375, 1212, 1079, 1010, 939, 834, 746, 571, 492. HRMS (ESI) *m/z*: calcd for C₂₀H₁₈N₃BrNa [M + Na]⁺ 402.0576, 404.0558; found 402.0579, 404.0564.

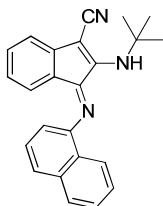


(E)-2-(tert-Butylamino)-1-(*m*-tolylimino)-1H-indene-3-carbonitrile (4aak):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 84% (26.5 mg) as red liquid. ¹H NMR (400 MHz, CDCl₃, δ ppm) 7.28 (dd, *J* = 14.6, 6.9 Hz, 1H), 7.17 (t, *J* = 7.5 Hz, 1H), 7.04 (dd, *J* = 6.9, 4.2 Hz, 2H), 6.79-6.70 (m, 2H), 6.58 (t, *J* = 7.5 Hz, 1H), 6.48 (s, 1H), 6.40 (d, *J* = 7.5 Hz, 1H), 2.37 (s, 3H), 1.58 (s, 9H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 160.3, 148.8, 147.6, 146.8, 139.1, 133.1, 128.9, 125.4, 125.3, 122.9, 121.8, 118.5, 118.3, 117.6, 114.6, 76.7, 52.0, 29.6, 21.0. IR

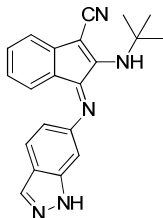
(KBr, cm^{-1}): 3320, 2970, 2192, 1607, 1505, 1456, 1369, 1292, 1215, 1088, 1012, 839, 751, 570, 490.

HRMS (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{21}\text{N}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 338.1628; found 338.1627.



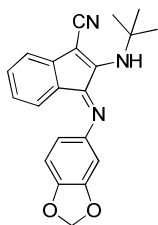
(E)-2-(tert-Butylamino)-1-(naphthalen-1-ylimino)-1H-indene-3-carbonitrile (4aal):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 84% (29.5 mg) as red liquid. **^1H NMR** (400 MHz, DMSO, δ ppm) 7.99 (d, $J = 8.1$ Hz, 1H), 7.83 (d, $J = 8.3$ Hz, 1H), 7.71 (d, $J = 8.3$ Hz, 1H), 7.60-7.45 (m, 3H), 7.19 (t, $J = 7.5$ Hz, 1H), 7.13-7.00 (m, 2H), 6.94 (d, $J = 7.4$ Hz, 1H), 6.51 (t, $J = 7.6$ Hz, 1H), 6.00 (d, $J = 7.5$ Hz, 1H), 1.58 (s, 9H); **^{13}C NMR** (101 MHz, DMSO, δ ppm) 161.7, 148.4, 147.2, 145.6, 134.5, 134.2, 128.6, 127.3, 126.8, 126.5, 125.6, 125.5, 124.5, 123.9, 123.4, 122.1, 119.0, 117.8, 113.3, 76.0, 52.9, 29.9. **IR** (KBr, cm^{-1}): 3364, 2924, 2191, 1608, 1516, 1457, 1372, 1292, 1212, 1085, 1015, 834, 750, 640, 489. **HRMS** (ESI) m/z : calcd for $\text{C}_{24}\text{H}_{21}\text{N}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 374.1628; found 374.1631.



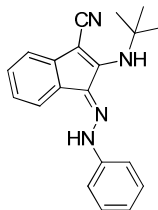
(E)-1-((1H-Indazol-6-yl)imino)-2-(tert-butylamino)-1H-indene-3-carbonitrile

(4aam): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 80% (27.3 mg) as a red solid: 192-193 $^{\circ}\text{C}$. **^1H NMR** (400 MHz, CDCl_3 , δ ppm) 11.60 (s, 1H), 8.13 (s, 1H), 7.79 (d, $J = 8.5$ Hz, 1H), 7.14 (t, $J = 7.5$ Hz, 1H), 7.08-7.00 (m, 2H), 6.79 (d, $J = 8.5$ Hz, 1H), 6.52-6.48 (m, 2H), 6.33 (d, $J = 7.6$ Hz, 1H), 1.59 (s, 9H); **^{13}C NMR** (101 MHz, CDCl_3 , δ ppm) 161.3, 148.3, 148.0, 147.3, 140.8, 134.7, 133.9, 125.8, 123.4, 122.3, 122.0, 121.0, 119.0, 118.1, 113.8, 98.1, 76.9, 52.6, 30.1. **IR** (KBr, cm^{-1}): 3320, 2925, 2193, 1612, 1523, 1459, 1367, 1209, 1072, 1016, 946, 857, 749, 639, 569, 504. **HRMS** (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{19}\text{N}_5\text{Na}$ $[\text{M} + \text{Na}]^+$ 364.1533; found 364.1537.



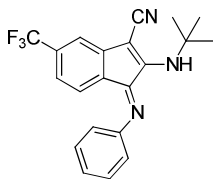
(E)-1-(Benzo[d][1,3]dioxol-5-ylimino)-2-(tert-butylamino)-1H-indene-3-carbonitrile (4aan):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 92% (31.7 mg) as a red solid: 130-132 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.18 (t, $J = 7.3$ Hz, 1H), 7.04 (d, $J = 7.5$ Hz, 1H), 6.83 (d, $J = 8.1$ Hz, 1H), 6.65 (dt, $J = 14.8, 7.4$ Hz, 2H), 6.51 (s, 1H), 6.47 (s, 1H), 6.41 (d, $J = 8.1$ Hz, 1H), 6.02 (s, 2H), 1.58 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 161.1, 148.5, 148.1, 147.3, 145.3, 143.3, 133.6, 125.5, 123.3, 122.0, 118.9, 118.0, 111.1, 108.7, 101.5, 100.8, 76.8, 52.4, 30.1. **IR** (KBr, cm^{-1}): 3316, 2971, 2190, 1601, 1471, 1369, 1096, 1033, 930, 860, 812, 745, 563, 492. **HRMS** (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{19}\text{N}_5\text{O}_2\text{Na}$ $[\text{M} + \text{Na}]^+$ 368.1369; found 368.1371.



(E)-2-(tert-Butylamino)-1-(2-phenylhydrazono)-1H-indene-3-carbonitrile (4aa0):

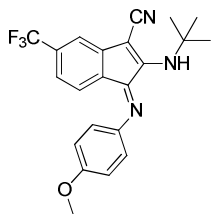
Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 64% (20.2 mg) as a red solid: 132-133 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 8.83 (s, 1H), 7.44 (d, $J = 7.5$ Hz, 1H), 7.37 (t, $J = 7.8$ Hz, 2H), 7.25 – 7.17 (m, 4H), 7.06 (t, $J = 7.4$ Hz, 1H), 6.99 (t, $J = 7.4$ Hz, 1H), 6.19 (s, 1H), 1.58 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 150.2, 144.1, 142.9, 136.4, 130.6, 129.6, 123.0, 122.9, 122.1, 121.7, 119.8, 118.2, 114.0, 74.5, 52.3, 30.2. **IR** (KBr, cm^{-1}): 3364, 2971, 2187, 1594, 1517, 1456, 1362, 1307, 1215, 1009, 921, 749, 691, 488. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{21}\text{N}_4$ $[\text{M} + \text{H}]^+$ 317.1761; found 317.1760.



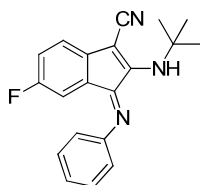
(E)-2-(tert-Butylamino)-1-(phenylimino)-5-(trifluoromethyl)-1H-indene-3-carbonitrile (4baa):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 74% (27.3 mg) as a red solid: 126-127 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.41 (t, $J = 7.7$ Hz, 2H), 7.23 (s, 2H), 6.90 (d, $J = 7.9$ Hz, 2H), 6.81 (d, $J = 7.9$ Hz, 1H), 6.54 (s, 1H), 6.42 (d, $J = 7.8$ Hz, 1H), 1.58 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 159.7, 148.7, 148.3, 148.2, 135.9 (q, $J =$

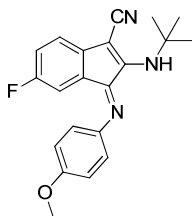
32.2 Hz), 129.6, 126.2 (d, $J = 271.6$ Hz), 125.7, 125.5, 124.8, 120.4 (q, $J = 4.1$ Hz), 118.2, 118.0, 114.4 (q, $J = 3.7$ Hz), 76.8, 52.8, 30.0. **IR** (KBr, cm^{-1}): 3321, 2976, 2195, 1605, 1525, 1475, 1430, 1357, 1318, 1264, 1214, 1167, 1126, 1075, 951, 883, 834, 767, 710, 569, 509. **HRMS** (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{18}\text{N}_3\text{F}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 392.1345; found 392.1338.



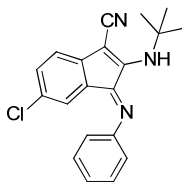
(E)-2-(tert-Butylamino)-1-((4-methoxyphenyl)imino)-5-(trifluoromethyl)-1H-indene-3-carbonitrile (4bab): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 85% (33.9 mg) as a red solid: 157-159 °C. **^1H NMR** (400 MHz, CDCl_3 , δ ppm) 7.26 (s, 1H), 6.95 (q, $J = 8.9$ Hz, 4H), 6.88 (d, $J = 7.9$ Hz, 1H), 6.73 (d, $J = 7.9$ Hz, 1H), 6.60 (s, 1H), 3.86 (s, 3H), 1.59 (s, 9H); **^{13}C NMR** (101 MHz, CDCl_3 , δ ppm) 159.1, 158.0, 148.6, 148.2, 141.4, 134.9 (q, $J = 32.2$ Hz), 125.0, 124.9, 124.8, 126.4 (q, $J = 271.2$ Hz), 120.3 (q, $J = 4.1$ Hz), 118.4, 114.7, 114.3 (q, $J = 3.6$ Hz), 76.5, 55.5, 52.7, 30.0. **IR** (KBr, cm^{-1}): 3310, 2974, 2193, 1603, 1506, 1431, 1356, 1317, 1249, 1168, 1124, 1034, 951, 883, 834, 725, 516. **HRMS** (ESI) m/z : calcd for $\text{C}_{22}\text{H}_{20}\text{N}_3\text{OF}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 422.1451; found 422.1453.



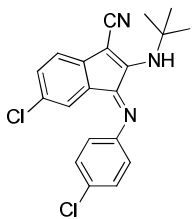
(E)-2-(tert-Butylamino)-6-fluoro-1-(phenylimino)-1H-indene-3-carbonitrile (4caa): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 81% (25.8 mg) as red liquid. **^1H NMR** (400 MHz, CDCl_3 , δ ppm) 7.43 (t, $J = 7.7$ Hz, 2H), 7.25 (d, $J = 8.0$ Hz, 1H), 6.98-6.86 (m, 4H), 6.42 (s, 1H), 6.07 (dd, $J = 8.6, 2.0$ Hz, 1H), 1.58 (s, 9H); **^{13}C NMR** (101 MHz, CDCl_3 , δ ppm) 160.1 (d, $J = 2.7$ Hz), 159.5 (d, $J = 240.3$ Hz), 148.7, 147.9 (d, $J = 2.3$ Hz), 142.8 (d, $J = 2.4$ Hz), 129.7, 125.6, 123.1 (d, $J = 8.2$ Hz), 119.5 (d, $J = 22.7$ Hz), 118.8, 118.4 (d, $J = 7.8$ Hz), 118.0, 113.6 (d, $J = 25.9$ Hz), 77.6, 52.5, 30.0. **IR** (KBr, cm^{-1}): 3325, 2972, 2193, 1612, 1523, 1468, 1372, 1265, 1204, 1066, 832, 768, 701, 573, 512. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{18}\text{N}_3\text{FNa}$ $[\text{M} + \text{Na}]^+$ 342.1377; found 342.1382.



(E)-2-(tert-Butylamino)-6-fluoro-1-((4-methoxyphenyl)imino)-1H-indene-3-carbonitrile (4cab): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 84% (29.3 mg) as a red solid: 146-147 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 6.91 (ddd, $J = 16.7, 11.4, 5.5$ Hz, 6H), 6.45 (s, 1H), 6.35 (dd, $J = 8.7, 2.1$ Hz, 1H), 3.84 (s, 3H), 1.56 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 159.5 (d, $J = 2.8$ Hz), 159.4 (d, $J = 239.3$ Hz), 157.9, 148.2 (d, $J = 2.3$ Hz), 142.8 (d, $J = 2.4$ Hz), 141.3, 123.1 (d, $J = 8.2$ Hz), 120.1, 119.3 (d, $J = 2.3$ Hz), 118.9, 118.3 (d, $J = 7.8$ Hz), 114.8, 113.2 (d, $J = 2.5$ Hz), 76.3, 55.5, 52.4, 30.0. **IR** (KBr, cm^{-1}): 3320, 2972, 2192, 1609, 1510, 1465, 1371, 1246, 1106, 1031, 947, 828, 730, 574, 511. **HRMS** (ESI) m/z : calcd for $\text{C}_{22}\text{H}_{20}\text{N}_3\text{OF}_3\text{Na}$ $[\text{M} + \text{Na}]^+$ 372.1483; found 372.1482.

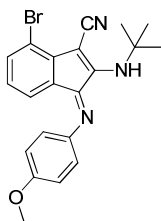


(E)-2-(tert-Butylamino)-6-chloro-1-(phenylimino)-1H-indene-3-carbonitrile (4daa): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 88% (29.5 mg) as a red solid: 178-180 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.43 (t, $J = 7.6$ Hz, 2H), 7.26 (t, $J = 7.4$ Hz, 1H), 7.13 (d, $J = 8.0$ Hz, 1H), 6.93 (dd, $J = 14.2, 8.0$ Hz, 3H), 6.49 (s, 1H), 6.30 (s, 1H), 1.58 (s, 9H); $^{13}\text{C NMR}$ (101 MHz, CDCl_3 , δ ppm) 159.8, 148.6, 147.9, 145.7, 133.1, 129.6, 128.6, 125.7, 123.3, 118.8, 118.5, 118.1, 76.8, 52.7, 30.0. **IR** (KBr, cm^{-1}): 3319, 2977, 2194, 1605, 1521, 1455, 1318, 1208, 1118, 1067, 893, 815, 700, 574, 505. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{18}\text{N}_3\text{ClNa}$ $[\text{M} + \text{Na}]^+$ 358.1081; found 358.1084.

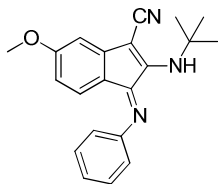


(E)-2-(tert-Butylamino)-6-chloro-1-((4-chlorophenyl)imino)-1H-indene-3-carbonitrile (4dai): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 73% (26.9 mg) as a red solid: 176-178 °C. $^1\text{H NMR}$ (400 MHz, CDCl_3 , δ ppm) 7.40 (d, $J = 8.0$ Hz, 2H), 7.15 (d, $J = 8.0$ Hz, 1H), 6.95 (d, $J = 8.0$ Hz, 1H), 6.88 (d, $J = 8.0$ Hz, 2H), 6.43 (s, 1H),

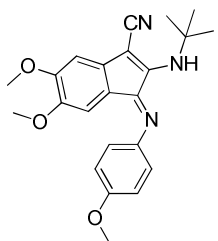
6.41 (s, 1H), 1.57 (s, 9H); ^{13}C NMR (101 MHz, CDCl_3 , δ ppm) 160.2, 147.7, 146.8, 145.9, 133.4, 131.2, 129.8, 128.7, 125.6, 123.1, 119.8, 119.0, 118.4, 76.8, 52.7, 30.0. **IR** (KBr, cm^{-1}): 3320, 2977, 2194, 1609, 1522, 1458, 1318, 1210, 1092, 1009, 833, 664, 571, 503. **HRMS** (ESI) m/z : calcd for $\text{C}_{20}\text{H}_{17}\text{N}_3\text{Cl}_2\text{Na}$ $[\text{M} + \text{Na}]^+$ 392.0692; found 392.0688.



(E)-4-Bromo-2-(tert-butylamino)-1-((4-methoxyphenyl)imino)-1H-indene-3-carbonitrile (4eab): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 79% (32.3 mg) as red liquid. ^1H NMR (400 MHz, CDCl_3 , δ ppm) 7.23 (d, $J = 8.1$ Hz, 1H), 6.94 (d, $J = 8.8$ Hz, 2H), 6.87 (d, $J = 8.7$ Hz, 2H), 6.70 (s, 1H), 6.59 (d, $J = 7.5$ Hz, 1H), 6.43 (t, $J = 7.8$ Hz, 1H), 3.84 (s, 3H), 1.60 (s, 9H); ^{13}C NMR (101 MHz, CDCl_3 , δ ppm) 159.0, 157.7, 151.1, 144.2, 141.5, 138.3, 124.6, 124.4, 124.3, 120.1, 119.9, 114.8, 113.5, 77.1, 55.6, 53.0, 30.7. **IR** (KBr, cm^{-1}): 3308, 2970, 2195, 1598, 1507, 1457, 1415, 1368, 1245, 1124, 1030, 839, 739, 567, 507. **HRMS** (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{20}\text{N}_3\text{OBrNa}$ $[\text{M} + \text{Na}]^+$ 432.0682, 434.0664; found 432.0687, 434.0666.

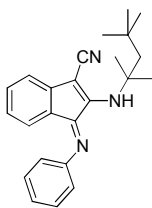


(E)-2-(tert-Butylamino)-5-methoxy-1-(phenylimino)-1H-indene-3-carbonitrile (4faa): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 52% (17.2 mg) as a red solid: 122-124 $^\circ\text{C}$. ^1H NMR (400 MHz, CDCl_3 , δ ppm) 7.39 (t, $J = 7.6$ Hz, 2H), 7.20 (t, $J = 7.4$ Hz, 1H), 6.92 (d, $J = 7.8$ Hz, 2H), 6.61-6-60 (m, 2H), 6.28 (d, $J = 8.4$ Hz, 1H), 6.04 (d, $J = 8.4$ Hz, 1H), 3.77 (s, 3H), 1.59 (s, 9H); ^{13}C NMR (101 MHz, CDCl_3 , δ ppm) 164.5, 159.8, 150.2, 149.5, 149.4, 129.5, 127.4, 124.9, 119.1, 118.5, 114.9, 108.0, 104.4, 75.7, 55.6, 52.5, 30.0. **IR** (KBr, cm^{-1}): 3314, 2968, 2192, 1601, 1523, 1473, 1365, 1214, 1110, 1023, 846, 768, 700, 560. **HRMS** (ESI) m/z : calcd for $\text{C}_{21}\text{H}_{21}\text{N}_3\text{ONa}$ $[\text{M} + \text{Na}]^+$ 354.1577; found 354.1578.



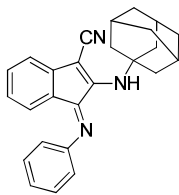
(E)-2-(tert-Butylamino)-5,6-dimethoxy-1-((4-methoxyphenyl)imino)-1H-indene-3-carbonitrile

e-3-carbonitrile (4gab): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 69% (27.0 mg) as a red solid: 174-175 °C. ¹H NMR (400 MHz, CDCl₃, δ ppm) 6.96-6.90 (m, 4H), 6.59 (s, 1H), 6.53 (s, 1H), 6.15 (s, 1H), 3.89 (s, 3H), 3.82 (s, 3H), 3.41 (s, 3H), 1.56 (s, 9H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 160.4, 157.4, 153.8, 148.9, 144.6, 142.9, 142.2, 120.4, 119.5, 114.6, 113.4, 110.4, 102.0, 74.7, 56.2, 56.1, 55.6, 52.4, 30.0. IR (KBr, cm⁻¹): 3311, 2943, 2188, 1604, 1496, 1296, 1241, 1074, 1026, 834, 547. HRMS (ESI) m/z: calcd for C₂₃H₂₅N₃O₃Na [M + Na]⁺ 414.1788; found 414.1789.



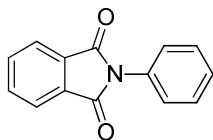
(E)-1-(Phenylimino)-2-((2,4,4-trimethylpentan-2-yl)amino)-1H-indene-3-carbonitrile

le (4aba): Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 61% (21.8 mg) as a red solid: 145-147 °C. ¹H NMR (400 MHz, CDCl₃, δ ppm) 7.44 (t, *J* = 7.0 Hz, 2H), 7.28-7.19 (m, 2H), 7.09 (d, *J* = 7.4 Hz, 1H), 6.95 (d, *J* = 8.0 Hz, 2H), 6.63 (s, 1H), 6.60 (t, *J* = 8.2 Hz, 1H), 6.40 (d, *J* = 7.5 Hz, 1H), 1.96 (s, 2H), 1.66 (s, 6H), 1.11 (s, 9H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 160.9, 149.2, 147.6, 133.7, 129.5, 125.7, 125.1, 123.2, 122.1, 119.0, 118.2, 118.0, 76.7, 56.4, 53.2, 31.8, 31.6, 30.2. IR (KBr, cm⁻¹): 3264, 2971, 2192, 1622, 1518, 1456, 1369, 1216, 1013, 757, 612, 497. HRMS (ESI) m/z: calcd for C₂₄H₂₇N₃Na [M + Na]⁺ 380.2097; found 380.2100.

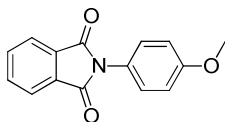


(E)-2-(Adamantan-1-ylamino)-1-(phenylimino)-1H-indene-3-carbonitrile (4aca):

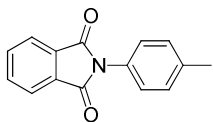
Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 57% (21.6 mg) as a red solid: 140-142 °C. ¹H NMR (400 MHz, CDCl₃, δ ppm) 7.41 (t, *J* = 7.8 Hz, 2H), 7.23 (t, *J* = 7.5 Hz, 1H), 7.17 (t, *J* = 7.3 Hz, 1H), 7.04 (d, *J* = 7.5 Hz, 1H), 6.93 (d, *J* = 7.4 Hz, 2H), 6.56 (t, *J* = 7.4 Hz, 1H), 6.43 (s, 1H), 6.35 (d, *J* = 7.5 Hz, 1H), 2.24 (s, 3H), 2.18 (s, 6H), 1.77 (dd, *J* = 32.0, 12.2 Hz, 6H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 161.0, 149.3, 147.6, 147.4, 133.6, 129.5, 125.6, 125.1, 123.2, 122.2, 119.5, 118.2, 118.0, 77.0, 52.9, 42.2, 35.9, 29.6. IR (KBr, cm⁻¹): 3310, 2912, 2191, 1602, 1520, 1453, 1356, 1300, 1235, 1178, 1103, 710, 577. HRMS (ESI) m/z: calcd for C₂₆H₂₅N₃Na [M + Na]⁺ 402.1941; found 402.1936.



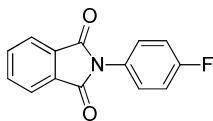
2-Phenylisoindoline-1,3-dione (6aaa)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 69% (15.4 mg) as a white solid: 200-202 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 8.00-7.93 (m, 2H), 7.83-7.77 (m, 2H), 7.54-7.49 (m, 2H), 7.47- 7.38 (m, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.3, 134.4, 131.8, 131.7, 129.1, 128.1, 126.6, 123.8. **IR** (KBr, cm⁻¹): 2916, 1704, 1608, 1515, 1461, 1388, 1298, 1174, 1097, 1026, 879, 823, 715, 527. **HRMS** (ESI) m/z: calcd for C₁₄H₉NO₂Na [M + Na]⁺ 246.0525; found 246.0521.



2-(4-Methoxyphenyl)isoindoline-1,3-dione (6aab)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 79% (19.9 mg) as a white solid: 158-160 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.97-7.90 (m, 2H), 7.80-7.74 (m, 2H), 7.37-7.30 (m, 2H), 7.05-6.98 (m, 2H), 3.85 (s, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.6, 159.3, 134.3, 131.9, 128.0, 124.3, 123.7, 114.5, 55.5. **IR** (KBr, cm⁻¹): 2915, 1704, 1608, 1515, 1461, 1388, 1298, 1250, 1174, 1097, 1026, 879, 824, 716, 528. **HRMS** (ESI) m/z: calcd for C₁₅H₁₁NO₃Na [M + Na]⁺ 276.0631; found 276.0633.

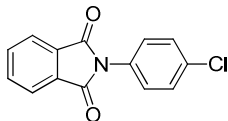


2-(p-Tolyl)isoindoline-1,3-dione (6aac)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 73% (17.3 mg) as a white solid: 204-205 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.98-7.91 (m, 2H), 7.81-7.75 (m, 2H), 7.31 (s, 4H), 2.41 (s, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.4, 138.2, 134.3, 131.9, 129.8, 129.0, 126.5, 123.7, 21.2. **IR** (KBr, cm⁻¹): 2922, 1712, 1598, 1484, 1370, 1238, 1176, 1081, 874, 779, 709, 626, 526, 444. **HRMS** (ESI) m/z: calcd for C₁₅H₁₁NO₂Na [M + Na]⁺ 260.0682; found 260.0678.

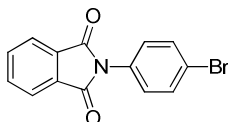


2-(4-Fluorophenyl)isoindoline-1,3-dione (6aah)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 62% (14.9 mg) as a white solid: 180-191 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.98-7.91 (m, 2H), 7.82-7.75 (m, 2H), 7.46-7.38 (m, 2H), 7.23-7.15 (m, 2H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.2, 162.0 (d, *J* = 246.5 Hz), 134.5,

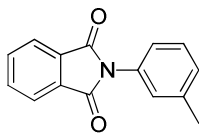
131.7, 128.4 (d, $J = 8.7$ Hz), 127.6 (d, $J = 3.2$ Hz), 123.8, 116.1(d, $J = 2.2$ Hz). **IR** (KBr, cm^{-1}): 3064, 1711, 1603, 1513, 1464, 1283, 1173, 1113, 1077, 880, 827, 791, 711, 523. **HRMS** (ESI) m/z : calcd for $\text{C}_{14}\text{H}_9\text{NO}_2\text{F}$ [$\text{M} + \text{H}$] $^+$ 242.0612; found 242.0608.



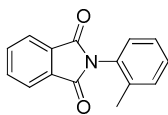
2-(4-Chlorophenyl)isoindoline-1,3-dione (6aai)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 64% (16.4 mg) as a white solid: 194-195 °C. **¹H NMR** (400 MHz, CDCl_3 , δ ppm) 7.99-7.92 (m, 2H), 7.83-7.77 (m, 2H), 7.49-7.45 (m, 2H), 7.44-7.37 (m, 2H); **¹³C NMR** (101 MHz, CDCl_3 , δ ppm) 167.0, 134.6, 133.8, 131.7, 130.3, 129.3, 127.7, 123.9. **IR** (KBr, cm^{-1}): 2918, 1711, 1498, 1396, 1173, 1120, 1085, 884, 819, 711, 517. **HRMS** (ESI) m/z : calcd for $\text{C}_{14}\text{H}_8\text{NO}_2\text{ClNa}$ [$\text{M} + \text{Na}$] $^+$ 280.0136; found 280.0131.



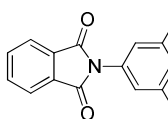
2-(4-Bromophenyl)isoindoline-1,3-dione (6aaj)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 67% (20.1 mg) as a white solid: 188-190 °C. **¹H NMR** (400 MHz, CDCl_3 , δ ppm) 7.97-7.94 (m, 2H), 7.81-7.79 (m, 2H), 7.63 (d, $J = 8.7$ Hz, 2H), 7.36 (d, $J = 8.7$ Hz, 2H); **¹³C NMR** (101 MHz, CDCl_3 , δ ppm) 166.9 134.6, 132.3, 131.6, 130.8, 128.0, 123.9, 121.8. **IR** (KBr, cm^{-1}): 2917, 1708, 1492, 1395, 1172, 1120, 884, 817, 711, 507. **HRMS** (ESI) m/z : calcd for $\text{C}_{14}\text{H}_8\text{NO}_2\text{BrNa}$ [$\text{M} + \text{Na}$] $^+$ 323.9631, 325.9611; found 323.9624, 325.9610.



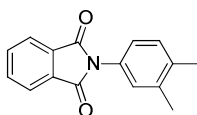
2-(*m*-Tolyl)isoindoline-1,3-dione (6aak)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 70% (16.6 mg) as a white solid: 176-178 °C. **¹H NMR** (400 MHz, CDCl_3 , δ ppm) 7.99-7.92 (m, 2H), 7.81-7.75 (m, 2H), 7.39 (t, $J = 7.7$ Hz, 1H), 7.23 (d, $J = 7.9$ Hz, 3H), 2.42 (s, 3H); **¹³C NMR** (101 MHz, CDCl_3 , δ ppm) 167.4, 139.2, 134.4, 131.8, 131.5, 129.1, 129.0, 127.3, 123.8, 123.7, 21.4. **IR** (KBr, cm^{-1}): 2922, 1714, 1599, 1489, 1372, 1243, 1178, 1080, 875, 779, 713, 627, 527, 443. **HRMS** (ESI) m/z : calcd for $\text{C}_{15}\text{H}_{11}\text{NO}_2\text{Na}$ [$\text{M} + \text{Na}$] $^+$ 260.0682; found 260.0680.



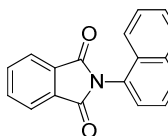
2-(o-Tolyl)isoindoline-1,3-dione (6aap)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 71% (16.8 mg) as a white solid: 178-180 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.96-7.93 (m, 2H), 7.80-7.77 (m, 2H), 7.41-7.29 (m, 3H), 7.20 (d, *J* = 7.6 Hz, 1H), 2.21 (s, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.4, 136.6, 134.4, 132.1, 131.2, 130.6, 129.5, 128.8, 126.9, 123.8, 18.1. **IR** (KBr, cm⁻¹): 2922, 1713, 1598, 1492, 1458, 1371, 1283, 1238, 1181, 1076, 873, 779, 710, 625, 525, 441. **HRMS** (ESI) *m/z*: calcd for C₁₅H₁₁NO₂Na [M + Na]⁺ 260.0682; found 260.0680.



2-(3,5-Dimethylphenyl)isoindoline-1,3-dione (6aaq)²: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 72% (18.1 mg) as a white solid: 133-134 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.97-7.91 (m, 2H), 7.80-7.74 (m, 2H), 7.05 (s, 1H), 7.02 (s, 2H), 2.38 (s, 6H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.5, 138.9, 134.3, 131.9, 131.4, 130.1, 124.5, 123.7, 21.3. **IR** (KBr, cm⁻¹): 2919, 1769, 1710, 1644, 1504, 1458, 1376, 1239, 1091, 1015, 948, 876, 798, 705, 523, 447. **HRMS** (ESI) *m/z*: calcd for C₁₆H₁₃NO₂Na [M + Na]⁺ 274.0838; found 274.0834.

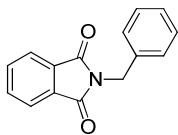


2-(3,4-Dimethylphenyl)isoindoline-1,3-dione (6aar)³: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 75% (18.8 mg) as a white solid: 188-189 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.97-7.91 (m, 2H), 7.80-7.75 (m, 2H), 7.26 (d, *J* = 2.1 Hz, 1H), 7.18 (s, 1H), 7.15-7.12 (m, 1H), 2.32 (s, 3H), 2.31 (s, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.6, 137.7, 137.1, 134.3, 131.9, 130.3, 129.2, 127.8, 124.2, 123.7, 19.9, 19.5. **IR** (KBr, cm⁻¹): 2919, 1769, 1712, 1644, 1501, 1458, 1415, 1376, 1241, 1173, 1097, 1016, 949, 878, 799, 750, 706, 606, 565, 524, 446. **HRMS** (ESI) *m/z*: calcd for C₁₆H₁₃NO₂Na [M + Na]⁺ 274.0838; found 274.0834.

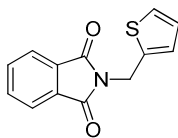


2-(Naphthalen-1-yl)isoindoline-1,3-dione (6aal)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 70% (19.1 mg) as a white solid: 180-181 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 8.07-7.92 (m, 4H), 7.87-7.79 (m, 2H), 7.64-7.58 (m,

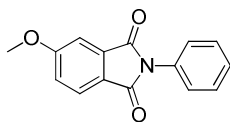
2H), 7.56-7.46 (m, 3H); ^{13}C NMR (101 MHz, CDCl_3 , δ ppm) 167.8, 134.5, 132.1, 130.3, 123.0, 128.9, 128.6, 128.3, 127.1, 127.0, 126.6, 125.4, 124.0, 122.5. IR (KBr, cm^{-1}): 2926, 1708, 1625, 1512, 1376, 1280, 1085, 1085, 956, 822, 706, 526, 468. HRMS (ESI) m/z : calcd for $\text{C}_{18}\text{H}_{11}\text{NO}_2\text{Na}$ [$\text{M} + \text{Na}$] $^+$ 296.0682; found 296.0680.



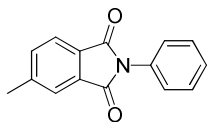
2-Benzylisoindoline-1,3-dione (6aas)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 52% (12.3 mg) as a white solid: 112-113 °C. ^1H NMR (400 MHz, CDCl_3 , δ ppm) 7.89-7.82 (m, 2H), 7.74-7.69 (m, 2H), 7.45 (d, $J = 7.1$ Hz, 2H), 7.37-7.27 (m, 3H), 4.87 (s, 2H); ^{13}C NMR (101 MHz, CDCl_3 , δ ppm) 168.1, 136.4, 134.0, 132.2, 128.6, 128.5, 127.8, 123.4, 41.6. IR (KBr, cm^{-1}): 2921, 1765, 1709, 1581, 1432, 1386, 1328, 1182, 1059, 935, 757, 709, 620, 518. HRMS (ESI) m/z : calcd for $\text{C}_{15}\text{H}_{11}\text{NO}_2\text{Na}$ [$\text{M} + \text{Na}$] $^+$ 260.0682; found 260.0677.



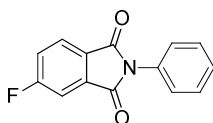
2-(Thiophen-2-ylmethyl)isoindoline-1,3-dione (6aat)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 49% (11.9 mg) as a white solid: 123-124 °C. ^1H NMR (400 MHz, CDCl_3 , δ ppm) 7.89-7.81 (m, 2H), 7.75- 7.67 (m, 2H), 7.21 (dd, $J = 5.1, 1.2$ Hz, 1H), 7.14 (dd, $J = 3.4, 0.5$ Hz, 1H), 6.93 (dd, $J = 5.1, 3.5$ Hz, 1H), 5.02 (s, 2H); ^{13}C NMR (101 MHz, CDCl_3 , δ ppm) 167.6, 138.1, 134.1, 132.1, 127.7, 126.9, 125.9, 123.4, 35.7. IR (KBr, cm^{-1}): 2924, 1768, 1716, 1464, 1428, 1388, 1322, 1236, 1176, 1084, 1033, 933, 842, 721, 637, 527. HRMS (ESI) m/z : calcd for $\text{C}_{13}\text{H}_9\text{NO}_2\text{SNa}$ [$\text{M} + \text{Na}$] $^+$ 266.0246; found 266.0242.



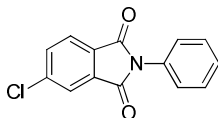
5-Methoxy-2-phenylisoindoline-1,3-dione (6baa)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 57% (14.4 mg) as a white solid: 178-179 °C. ^1H NMR (400 MHz, CDCl_3 , δ ppm) 7.85 (d, $J = 8.3$ Hz, 1H), 7.53-7.47 (m, 2H), 7.46-7.36 (m, 4H), 7.23 (dd, $J = 8.3, 2.3$ Hz, 1H), 3.95 (s, 3H); ^{13}C NMR (101 MHz, CDCl_3 , δ ppm) 167.1, 167.0, 165.0, 134.4 131.9, 129.1, 128.0, 126.6, 125.5, 123.7, 120.5, 108.2, 56.2. IR (KBr, cm^{-1}): 2921, 1713, 1640, 1545, 1461, 1406, 1321, 1115, 832, 706, 640, 521. HRMS (ESI) m/z : calcd for $\text{C}_{15}\text{H}_{11}\text{NO}_3\text{Na}$ [$\text{M} + \text{Na}$] $^+$ 276.0631; found 276.0627.



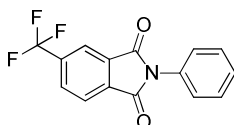
5-Methyl-2-phenylisoindoline-1,3-dione (6caa)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 54% (12.8 mg) as a white solid: 197-199 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.83 (d, *J* = 7.6 Hz, 1H), 7.75 (s, 1H), 7.58 (dd, *J* = 7.6, 0.6 Hz, 1H), 7.53-7.47 (m, 2H), 7.45-7.33 (m, 3H), 2.55 (s, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 167.5, 167.4, 145.8, 135.0, 132.2, 131.8, 129.2, 129.0, 128.0, 126.6, 124.3, 123.7, 22.1. **IR** (KBr, cm⁻¹): 2915, 1700, 1455, 1091, 930, 840, 739, 690, 566, 501. **HRMS** (ESI) *m/z*: calcd for C₁₅H₁₁NO₂Na [M + Na]⁺ 260.0682; found 260.0678.



5-Fluoro-2-phenylisoindoline-1,3-dione (6daa)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 47% (11.3 mg) as a white solid: 148-150 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.96 (dd, *J* = 8.1, 4.5 Hz, 1H), 7.63 (d, *J* = 6.8 Hz, 1H), 7.53-7.39 (m, 6H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 166.6 (d, *J* = 256.1 Hz), 166.2, 165.9 (d, *J* = 2.7 Hz), 134.6 (d, *J* = 9.5 Hz), 131.5, 129.2, 128.3, 127.5 (d, *J* = 2.7 Hz), 126.5, 126.2 (d, *J* = 9.4 Hz), 121.5 (d, *J* = 23.5 Hz), 111.5 (d, *J* = 24.8 Hz). **IR** (KBr, cm⁻¹): 2919, 1704, 1605, 1481, 1271, 1104, 896, 864, 802, 747, 697, 570, 503. **HRMS** (ESI) *m/z*: calcd for C₁₄H₉NO₂F [M + H]⁺ 242.0612; found 242.0610.

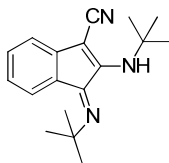


5-Chloro-2-phenylisoindoline-1,3-dione (6eaa)¹: Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 51% (13.1 mg) as a white solid: 189-190 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.93 (d, *J* = 1.5 Hz, 1H), 7.89 (d, *J* = 8.0 Hz, 1H), 7.76 (dd, *J* = 8.0, 1.7 Hz, 1H), 7.53-7.50 (m, 2H), 7.45-7.38 (m, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 166.3, 166.0, 141.2, 134.5, 133.4, 131.4, 129.8, 129.2, 128.3, 126.5, 125.0, 124.2. **IR** (KBr, cm⁻¹): 2919, 1710, 1598, 1385, 1166, 1107, 910, 845, 733, 691, 564, 502. **HRMS** (ESI) *m/z*: calcd for C₁₄H₈NCIO₂Na [M + Na]⁺ 280.0136; found 280.0132.



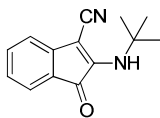
2-Phenyl-5-(trifluoromethyl)isoindoline-1,3-dione (6faa): Purified *via* flash

column chromatography with 10% ethyl acetate/petroleum ether, yielding 42% (12.2 mg) as a white solid: 88-90 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 8.23 (s, 1H), 8.09 (q, *J* = 8.0 Hz, 2H), 7.56-7.50 (m, 2H), 7.48-7.35 (m, 3H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 165.9, 165.8, 136.5 (d, *J* = 33.2 Hz), 134.6, 132.5, 131.5 (d, *J* = 3.6 Hz), 131.2, 129.3, 128.5, 126.5, 124.4, 121.7, 121.04 (d, *J* = 3.8 Hz), 121.3 (d, *J* = 271.8 Hz). **IR** (KBr, cm⁻¹): 2957, 1713, 1445, 1396, 1326, 1261, 1122, 1021, 864, 801, 748, 691, 601, 505. **HRMS** (ESI) *m/z*: calcd for C₁₅H₉NO₂F₃ [M + H]⁺ 292.0580; found 292.0577.



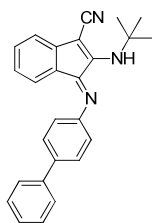
(E)-2-(tert-Butylamino)-1-(tert-butylimino)-1H-indene-3-carbonitrile (7aa):

Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 98% (27.5 mg) as a red solid: 152-153 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.53 (d, *J* = 7.7 Hz, 1H), 7.24 (t, *J* = 7.9 Hz, 1H), 7.10 (d, *J* = 7.5 Hz, 1H), 6.90 (t, *J* = 7.6 Hz, 1H), 6.72 (s, 1H), 1.52 (s, 9H), 1.51 (s, 9H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 156.1, 149.2, 148.9, 132.4, 128.2, 122.7, 121.7, 119.4, 118.0, 74.7, 55.7, 51.8, 30.0, 29.4. **IR** (KBr, cm⁻¹): 3263, 2970, 2191, 1623, 1518, 1457, 1369, 1293, 1216, 1072, 1013, 760, 711, 613, 498. **HRMS** (ESI) *m/z*: calcd for C₁₈H₂₃N₃Na [M + Na]⁺ 304.1784; found 304.1783.



2-(tert-butylamino)-1-oxo-1H-indene-3-carbonitrile (8):

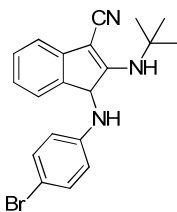
Purified *via* flash column chromatography with 10% ethyl acetate/petroleum ether, yielding 94% (42.5 mg) as a yellow solid: 138-139 °C. **¹H NMR** (400 MHz, CDCl₃, δ ppm) 7.35 (t, *J* = 8.1 Hz, 2H), 7.01 (d, *J* = 7.4 Hz, 1H), 6.92 (t, *J* = 7.4 Hz, 1H), 5.80 (s, 1H), 1.52 (s, 9H); **¹³C NMR** (101 MHz, CDCl₃, δ ppm) 190.7, 149.1, 143.3, 137.4, 124.8, 124.7, 124.4, 118.8, 117.9, 78.5, 52., 29.9. **IR** (KBr, cm⁻¹): 3340, 2927, 2182, 1728, 1625, 1517, 1458, 1365, 1285, 1202, 1046, 753, 540, 498. **HRMS** (ESI) *m/z*: calcd for C₁₄H₁₄N₂ONa [M + Na]⁺ 249.0998; found 249.1002.



(E)-1-([1,1'-biphenyl]-4-ylimino)-2-(tert-butylamino)-1H-indene-3-carbonitrile (10):

Purified *via* flash column chromatography with 40% ethyl acetate/petroleum ether, yielding 77% (58.0

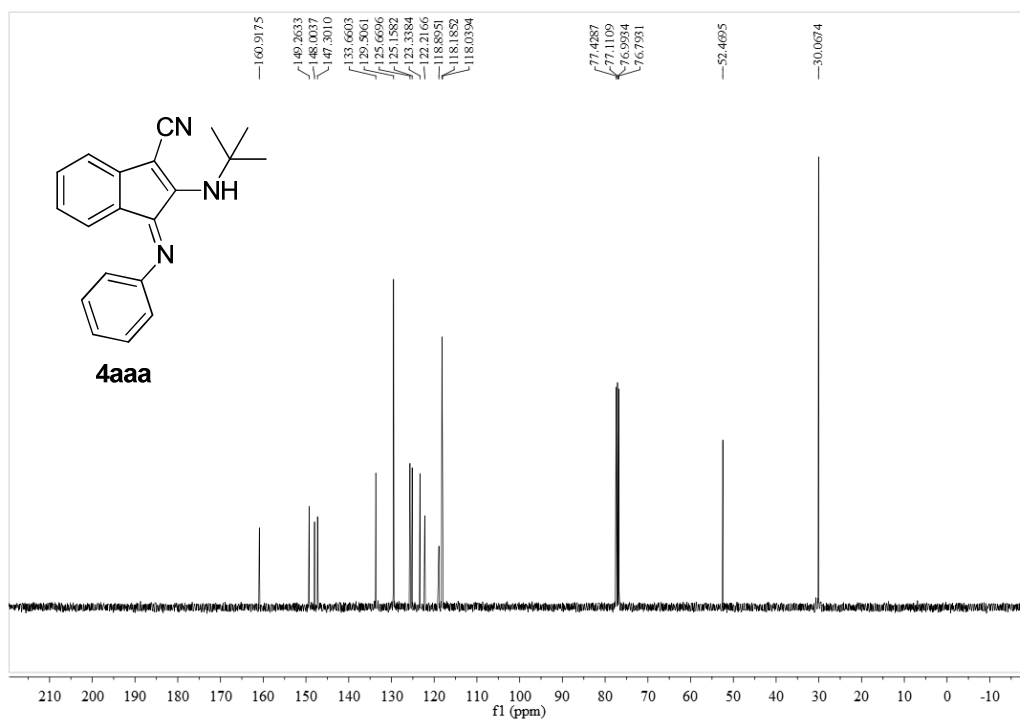
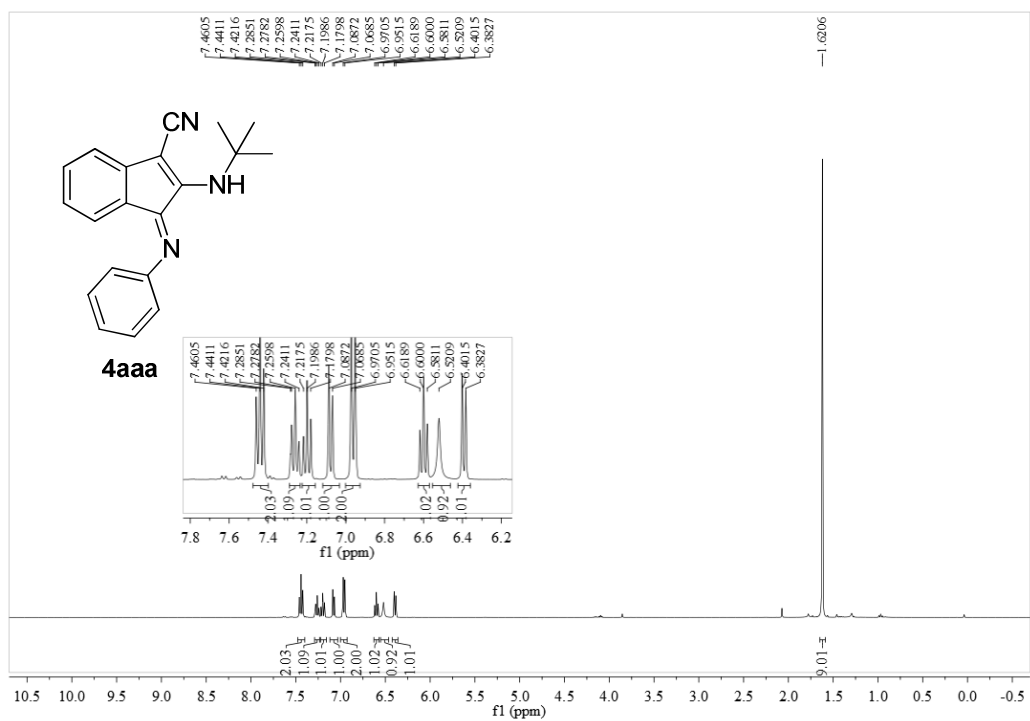
mg) as a red solid: 132-133 °C. ¹H NMR (400 MHz, CDCl₃, δ ppm) 7.67 (dd, *J* = 7.5, 5.7 Hz, 4H), 7.47 (t, *J* = 7.6 Hz, 2H), 7.37 (t, *J* = 7.3 Hz, 1H), 7.19 (t, *J* = 7.4 Hz, 1H), 7.05 (dd, *J* = 15.8, 7.9 Hz, 3H), 6.67-6.44 (m, 3H), 1.61 (s, 9H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 161.0, 148.4, 148.0, 147.4, 140.3, 138.1, 133.8, 128.9, 128.1, 127.4, 126.8, 125.7, 123.4, 122.3, 118.9, 118.1, 52.5, 30.1. IR (KBr, cm⁻¹): 3319, 2925, 2192, 1730, 1605, 1520, 1455, 1369, 1253, 1209, 1093, 747, 572, 495. HRMS (ESI) *m/z*: calcd for C₂₆H₂₄N₃ [M + H]⁺ 378.1965; found 378.1964.

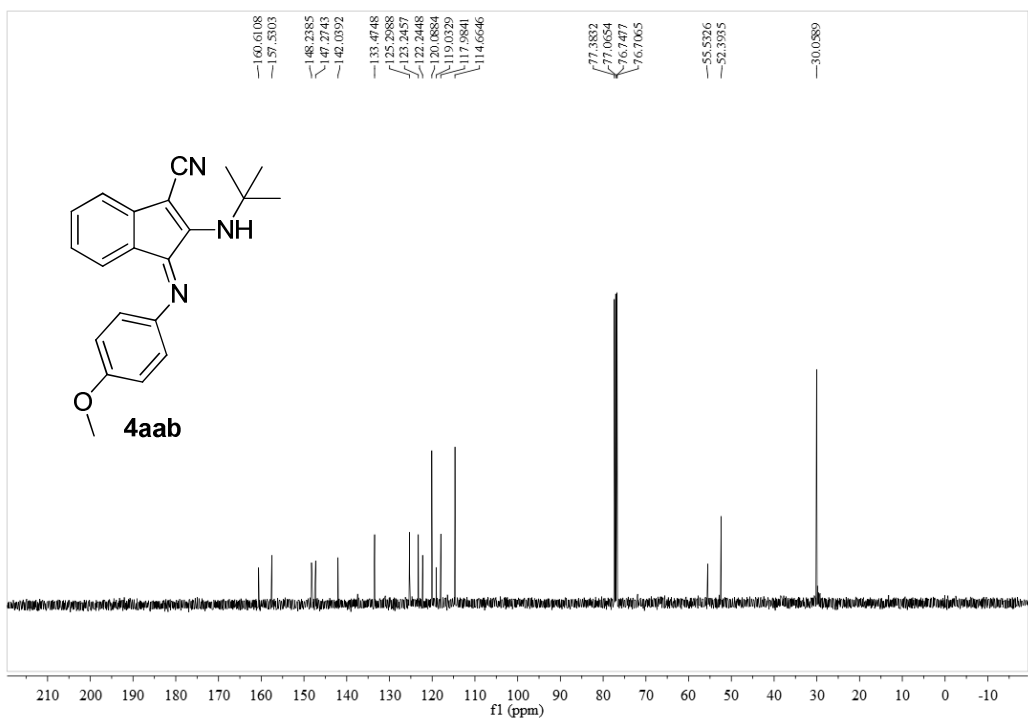
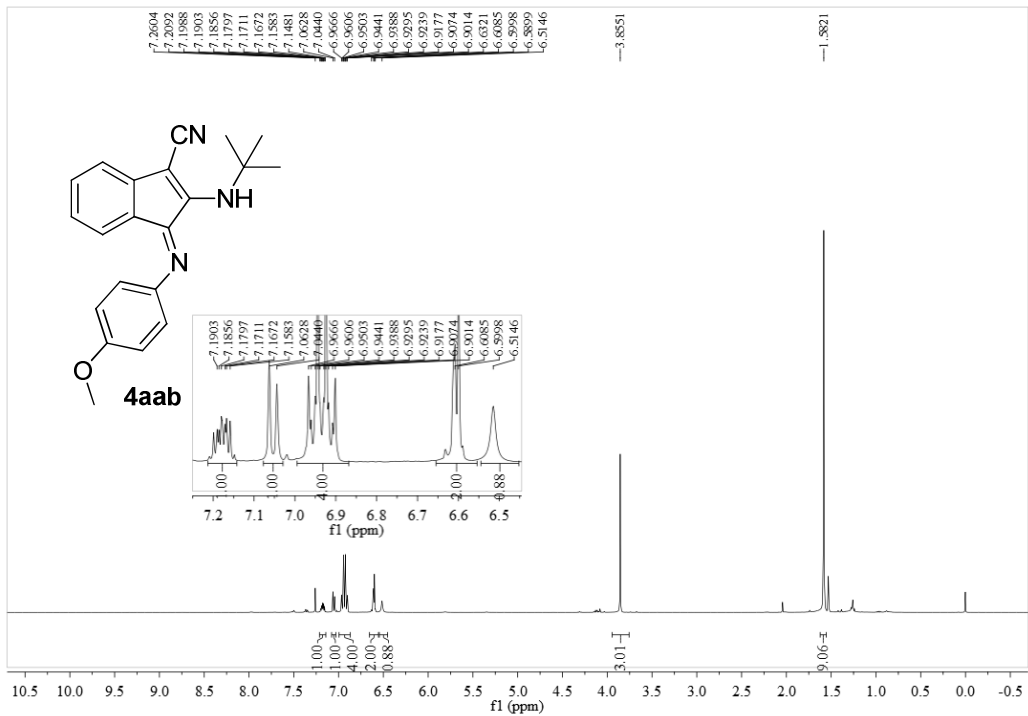


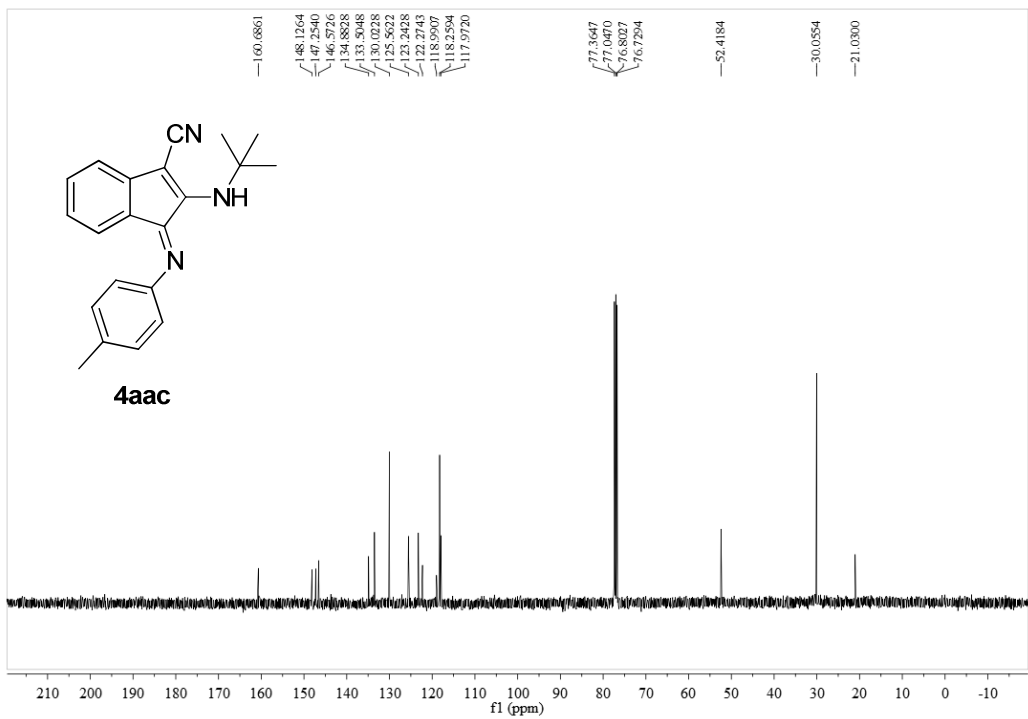
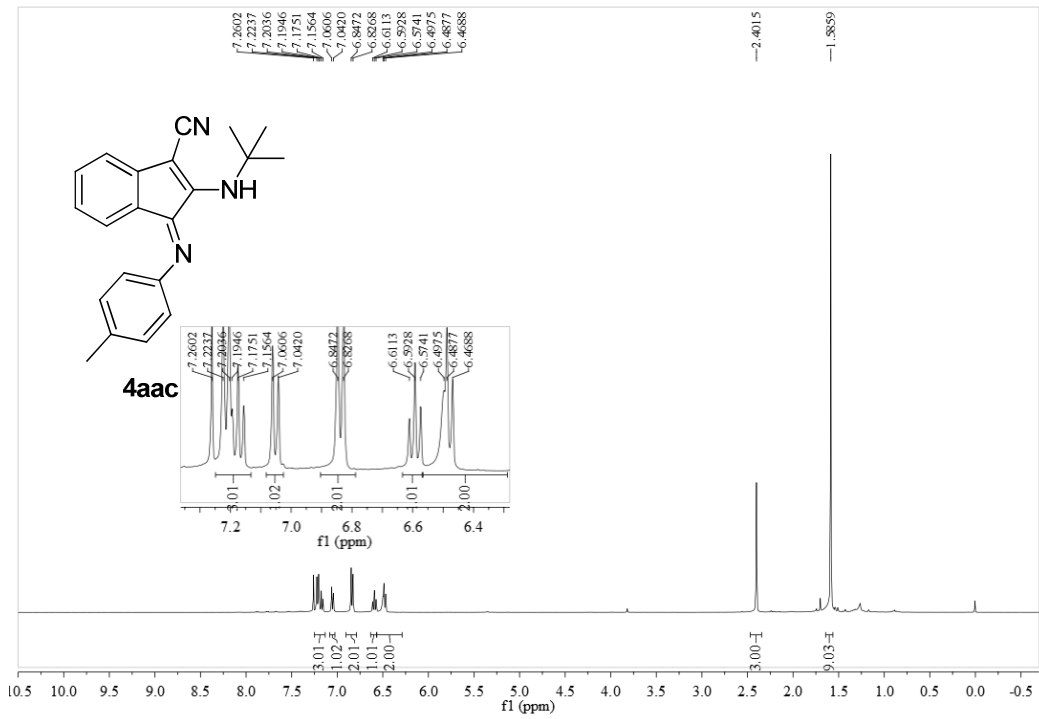
1-((4-bromophenyl)amino)-2-(tert-butylamino)-1H-indene-3-carbonitrile (11):

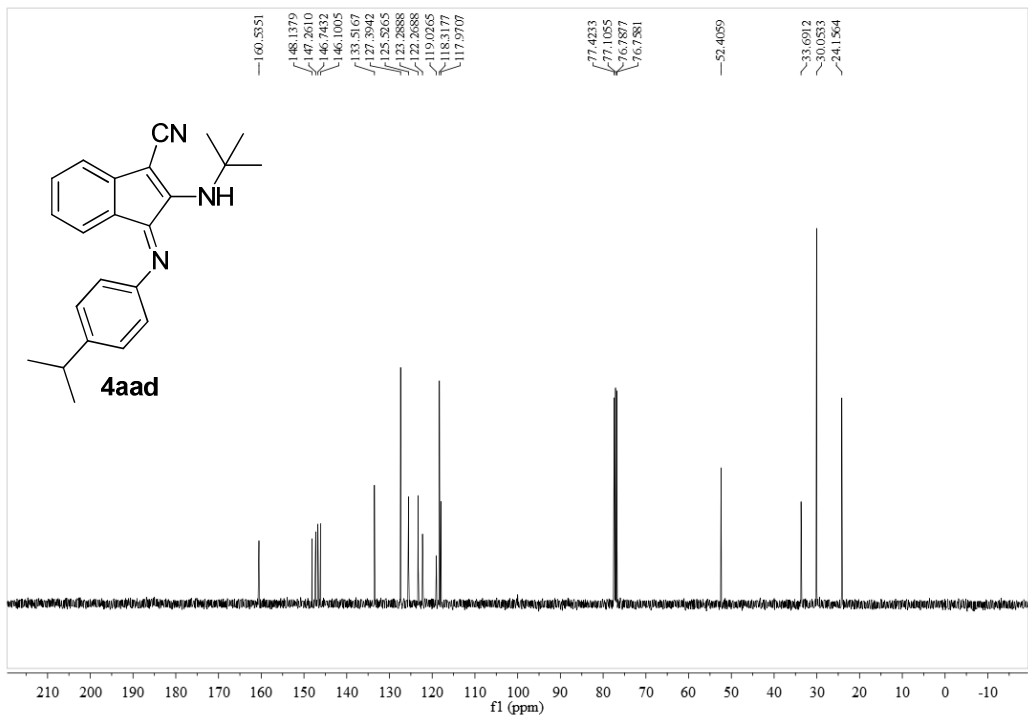
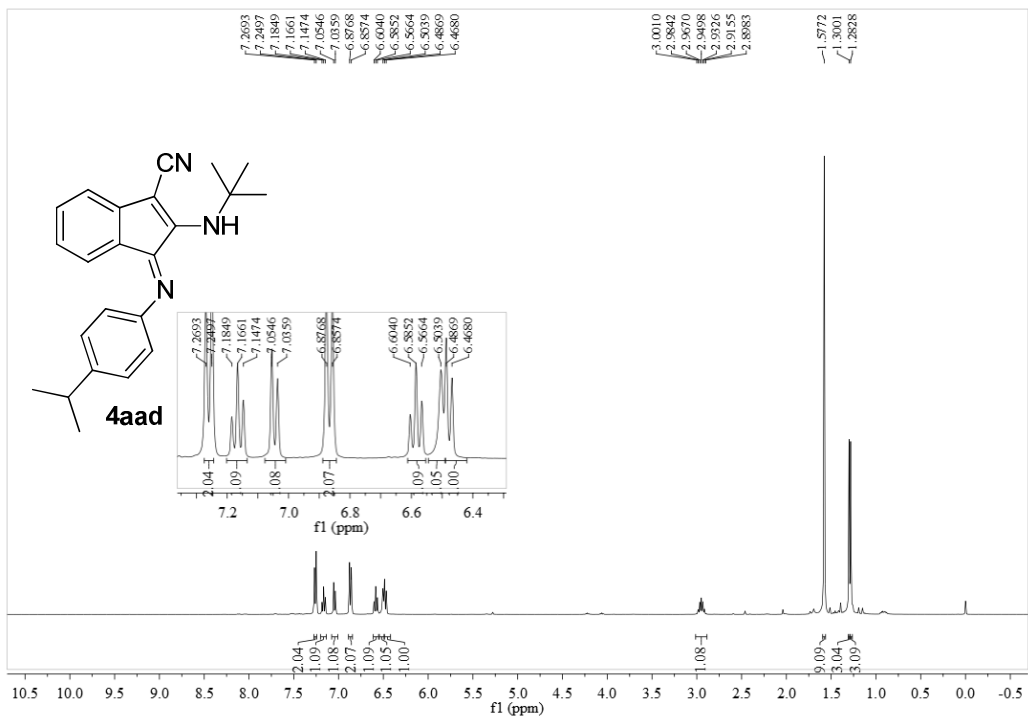
Purified *via* flash column chromatography with 20% ethyl acetate/petroleum ether, yielding 91% (69.3 mg) as a yellow liquid. ¹H NMR (400 MHz, CDCl₃, δ ppm) 7.31-7.14 (m, 5H), 7.04 (d, *J* = 7.3 Hz, 1H), 6.90 (t, *J* = 7.1 Hz, 1H), 6.39 (d, *J* = 8.6 Hz, 2H), 5.58 (s, 1H), 4.92 (s, 1H), 1.48 (s, 9H); ¹³C NMR (101 MHz, CDCl₃, δ ppm) 160.0, 144.3, 142.0, 132.9, 132.2, 129.3, 123.2, 122.3, 119.3, 117.5, 116.1, 111.6, 75.2, 63.2, 53.0, 29.9. IR (KBr, cm⁻¹): 3321, 2927, 2192, 1731, 1607, 1522, 1458, 1369, 1290, 1209, 1073, 749, 557, 495. HRMS (ESI) *m/z*: calcd for C₂₀H₂₀N₃BrNa [M + Na]⁺ 404.0733, 404.0714; found 404.0740, 404.0724.

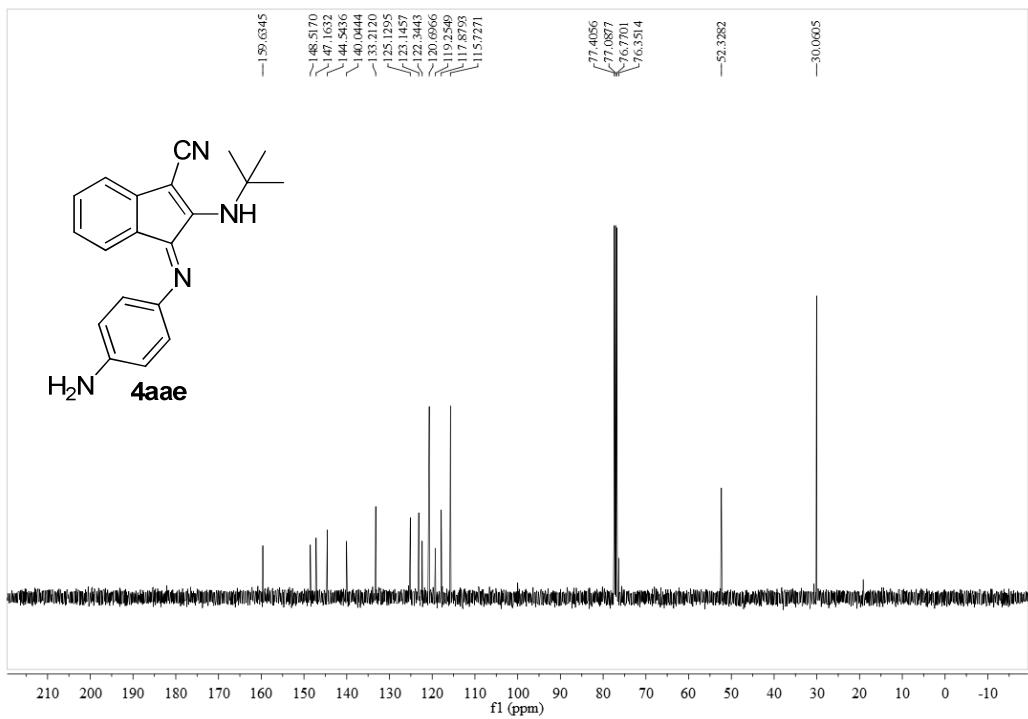
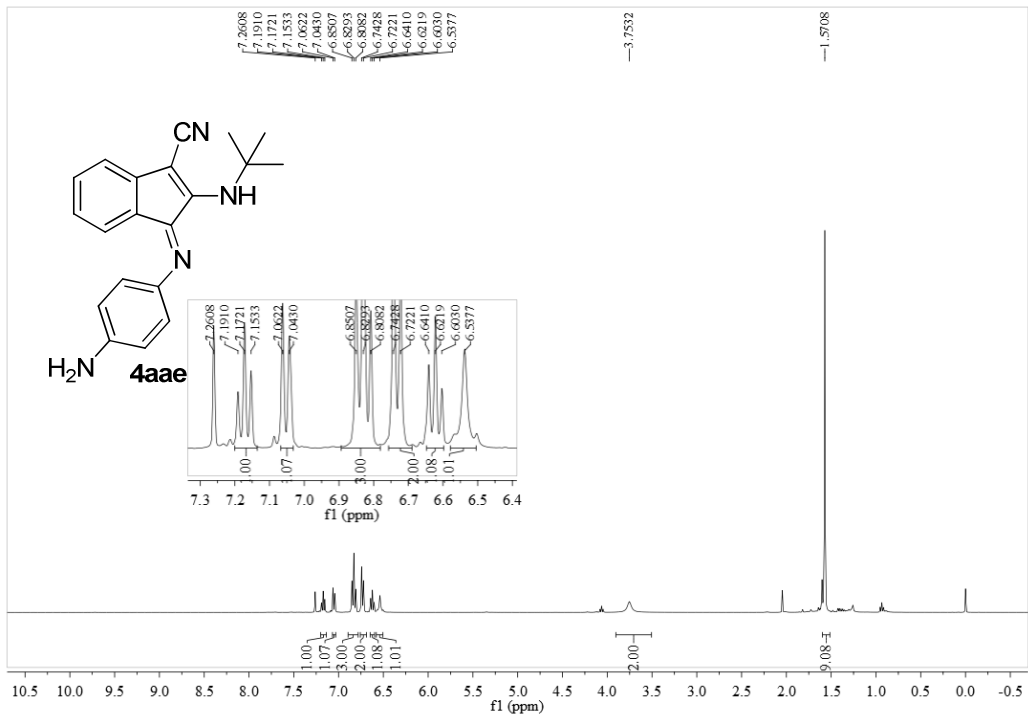
6. Copies of ^1H NMR and ^{13}C NMR Spectra of Products

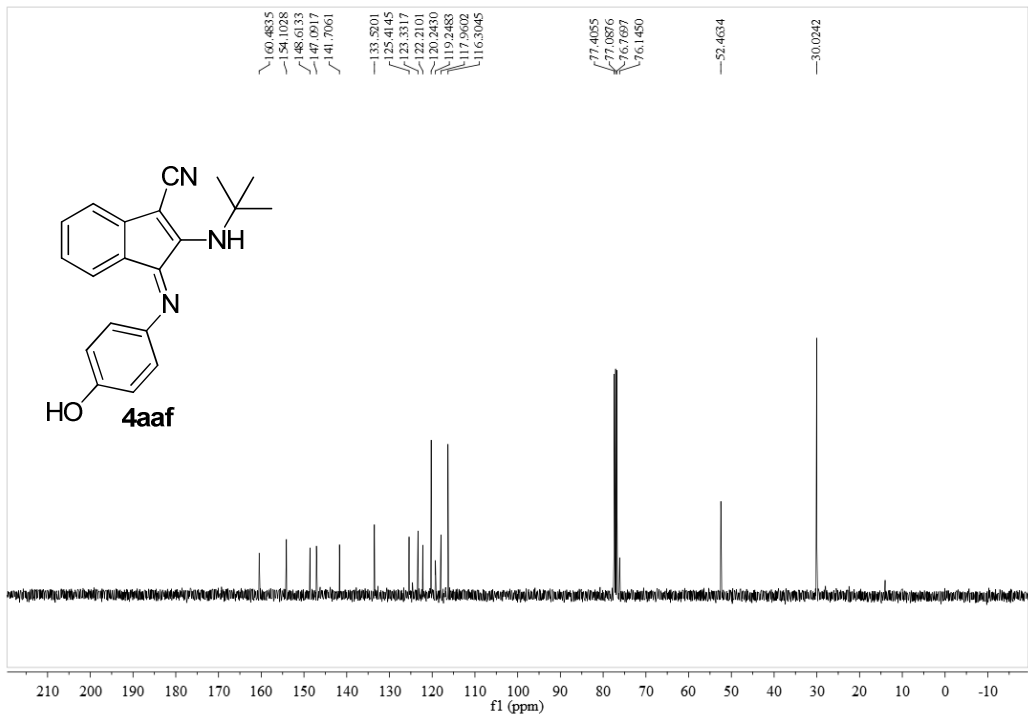
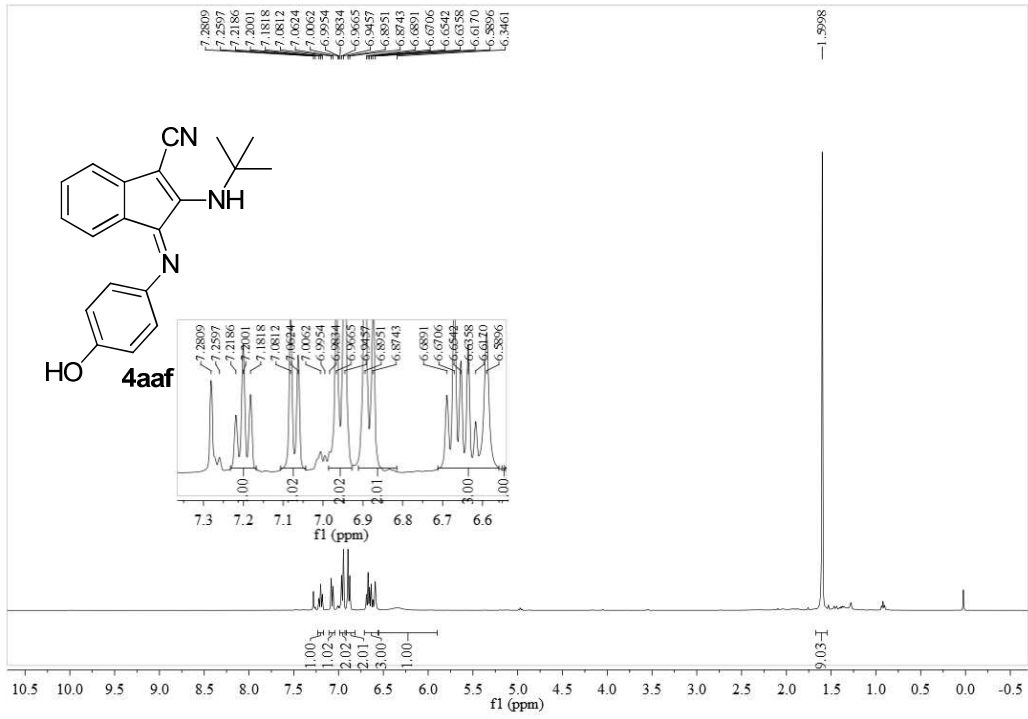


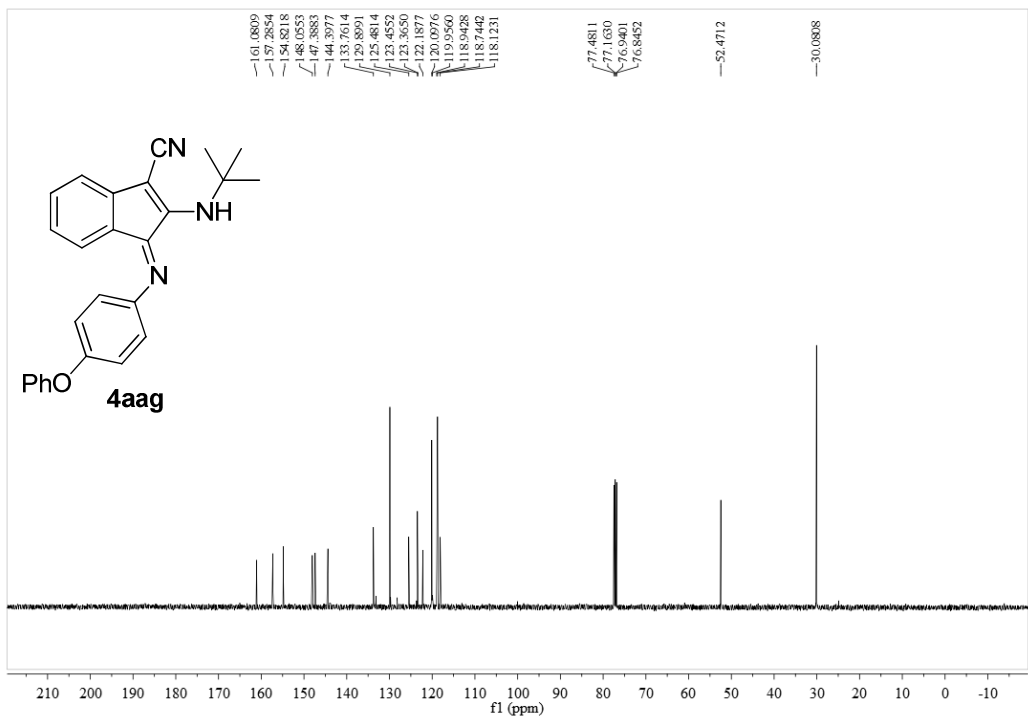
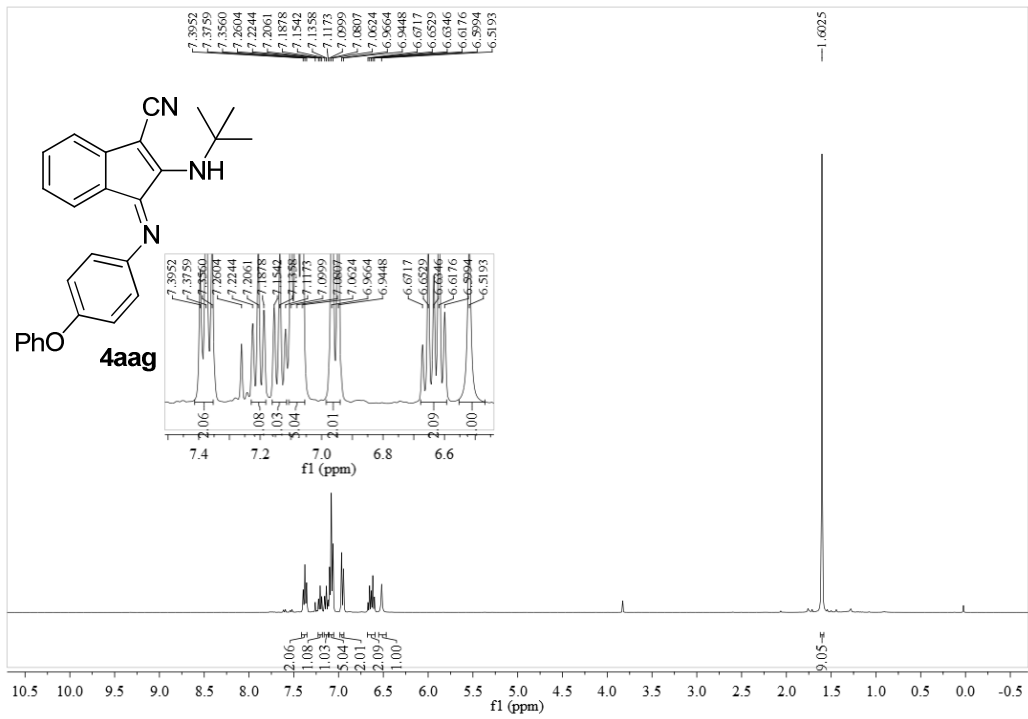


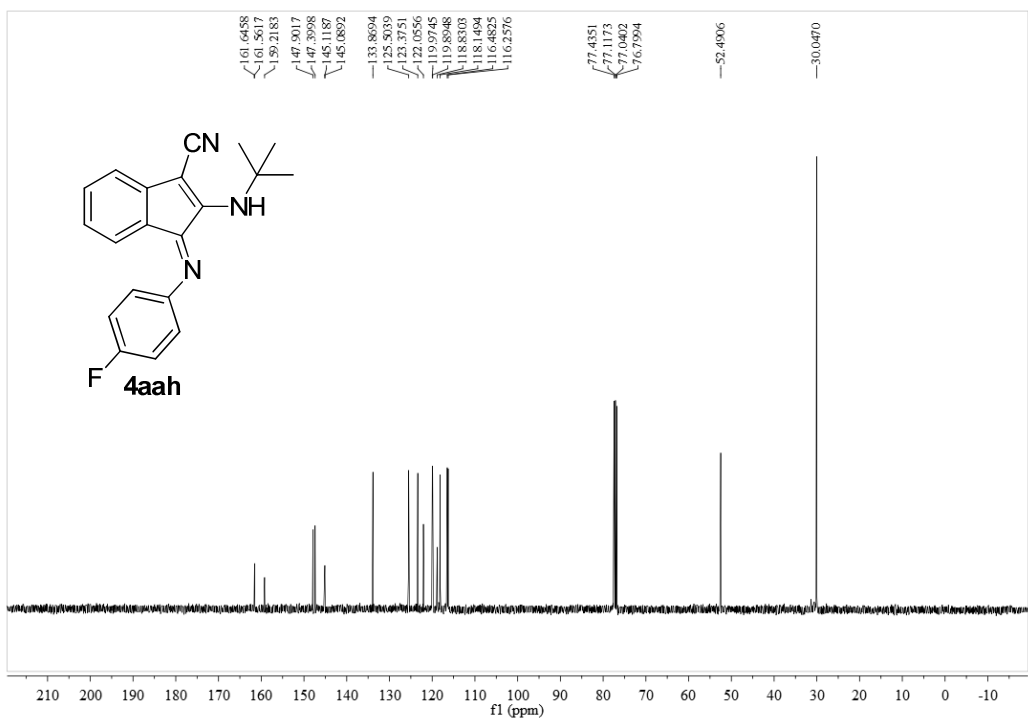
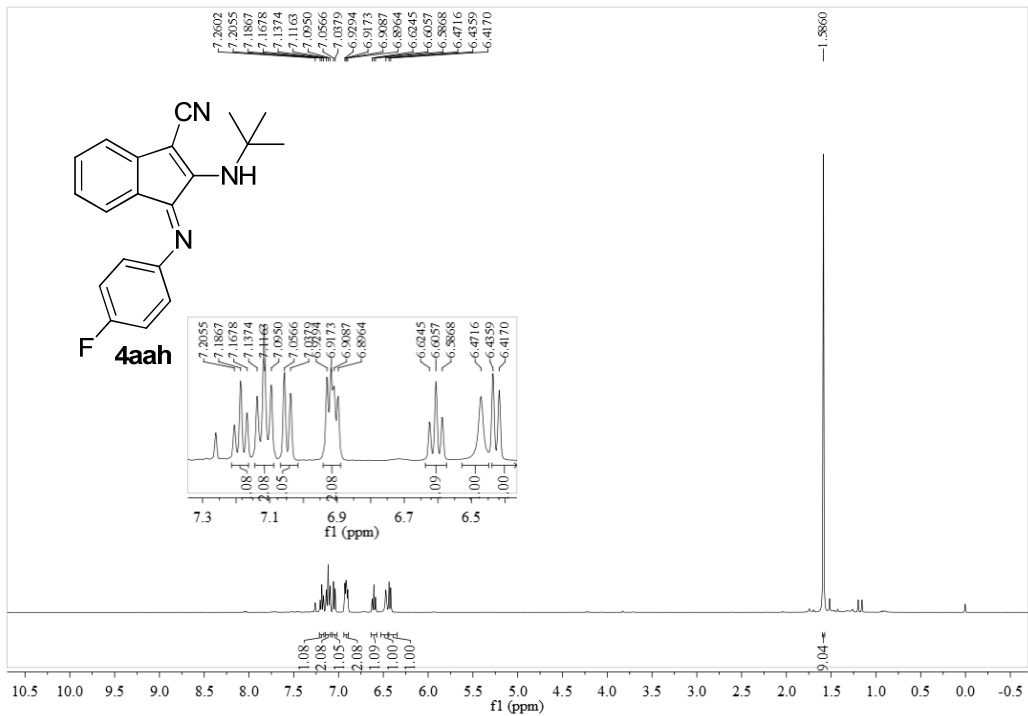


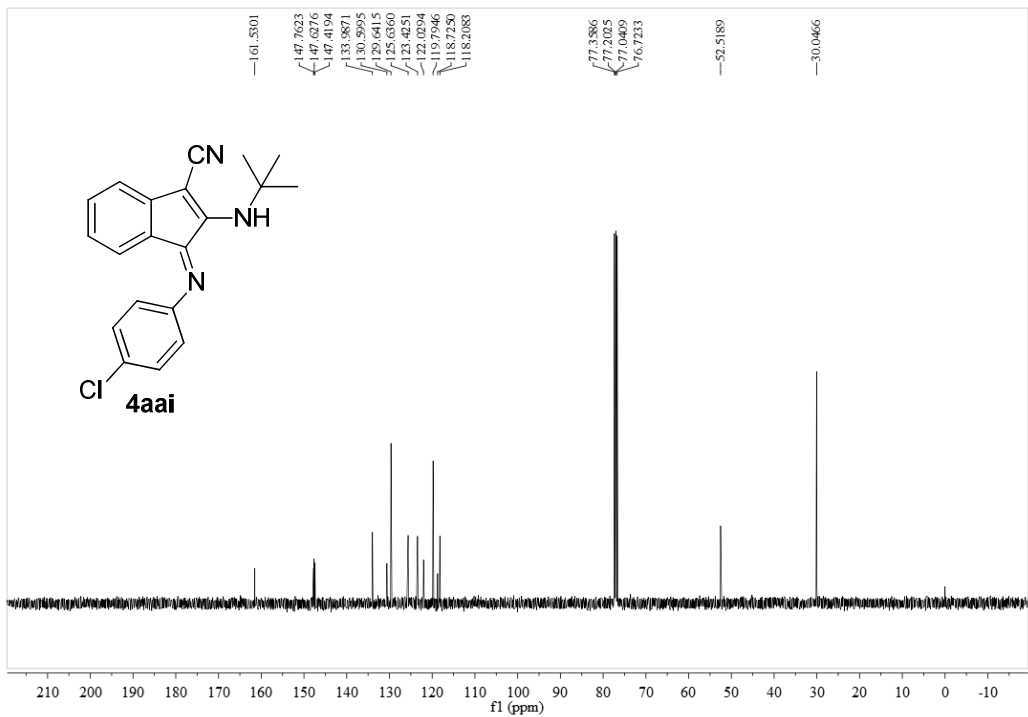
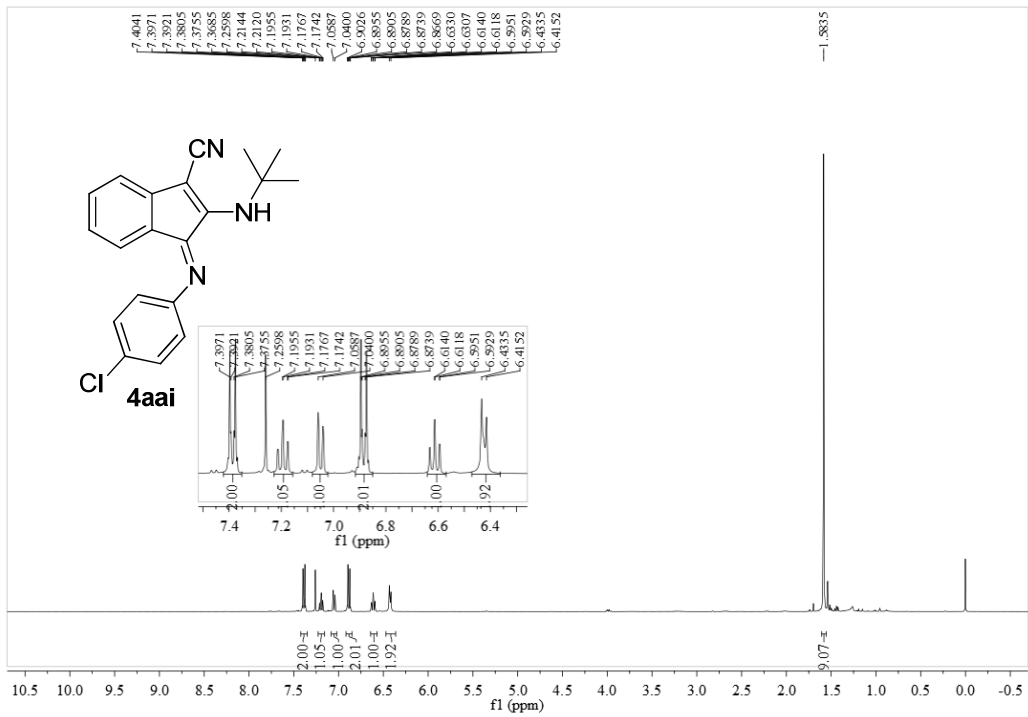


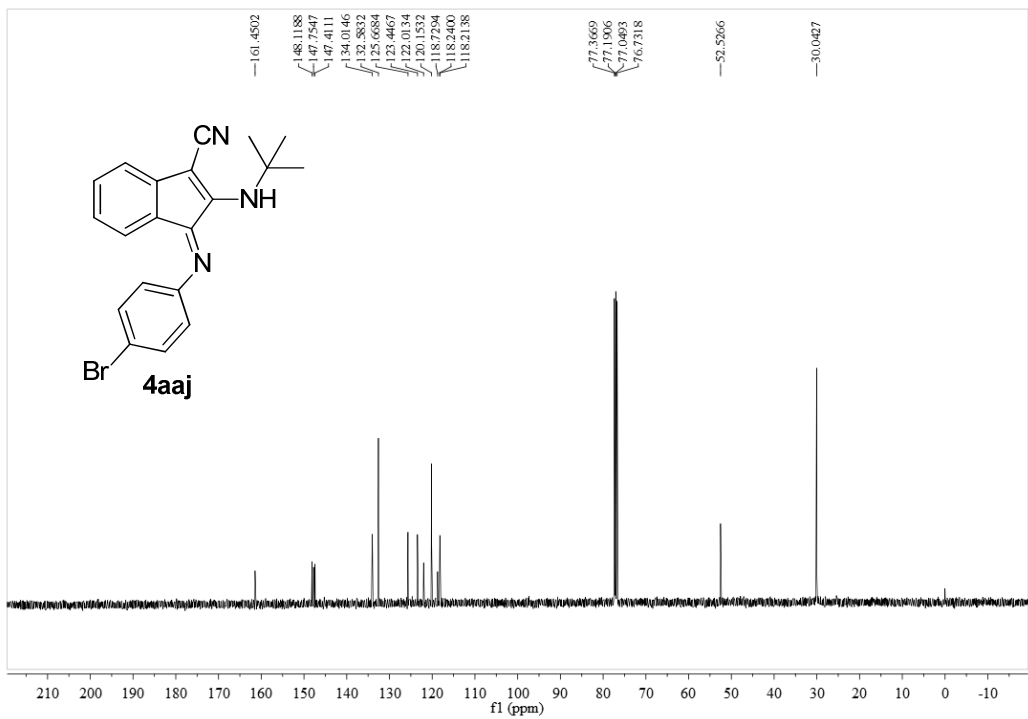
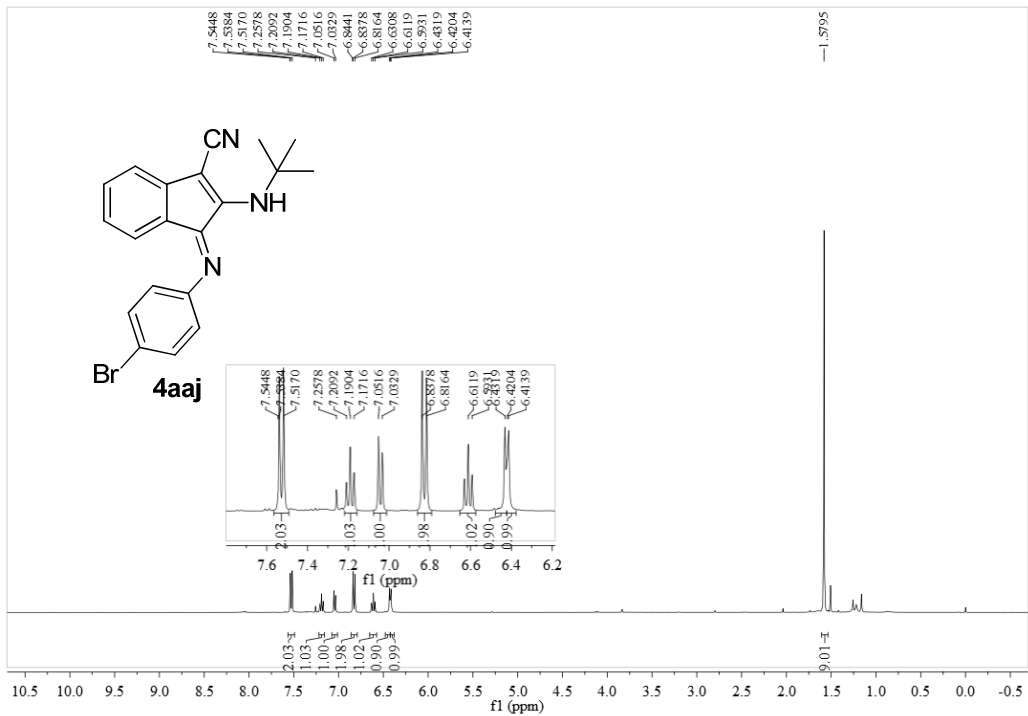


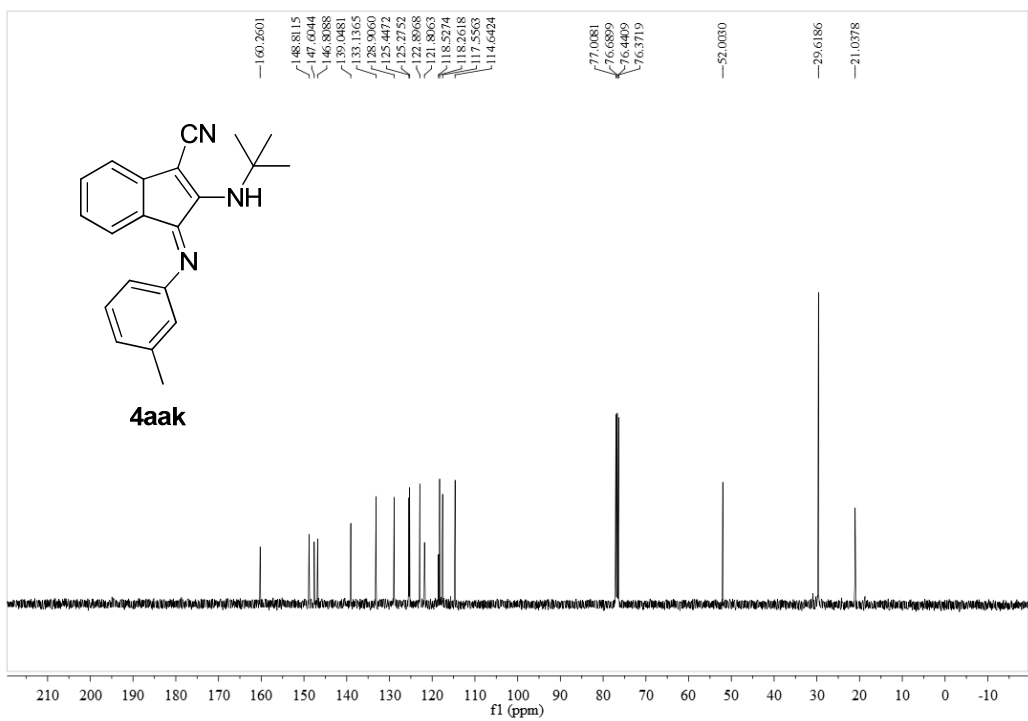
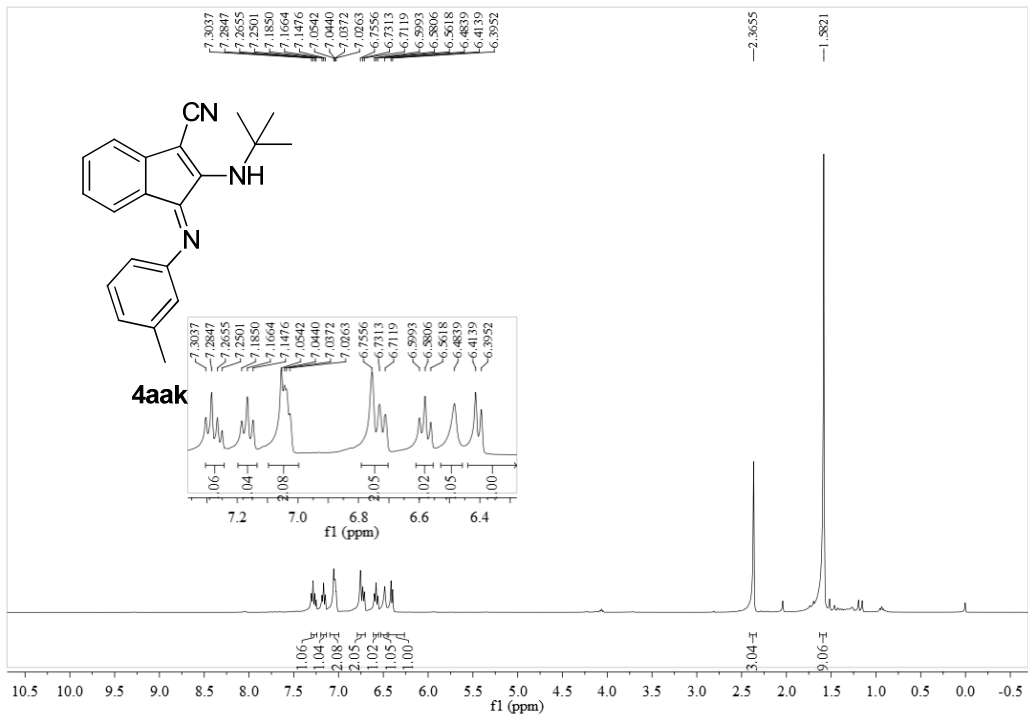


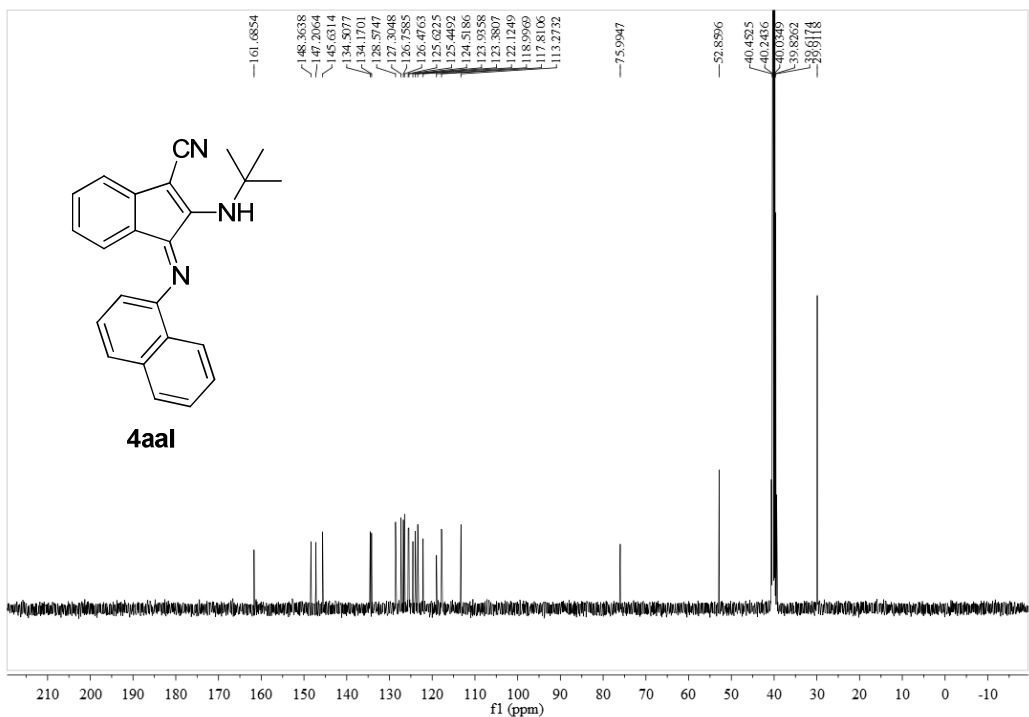
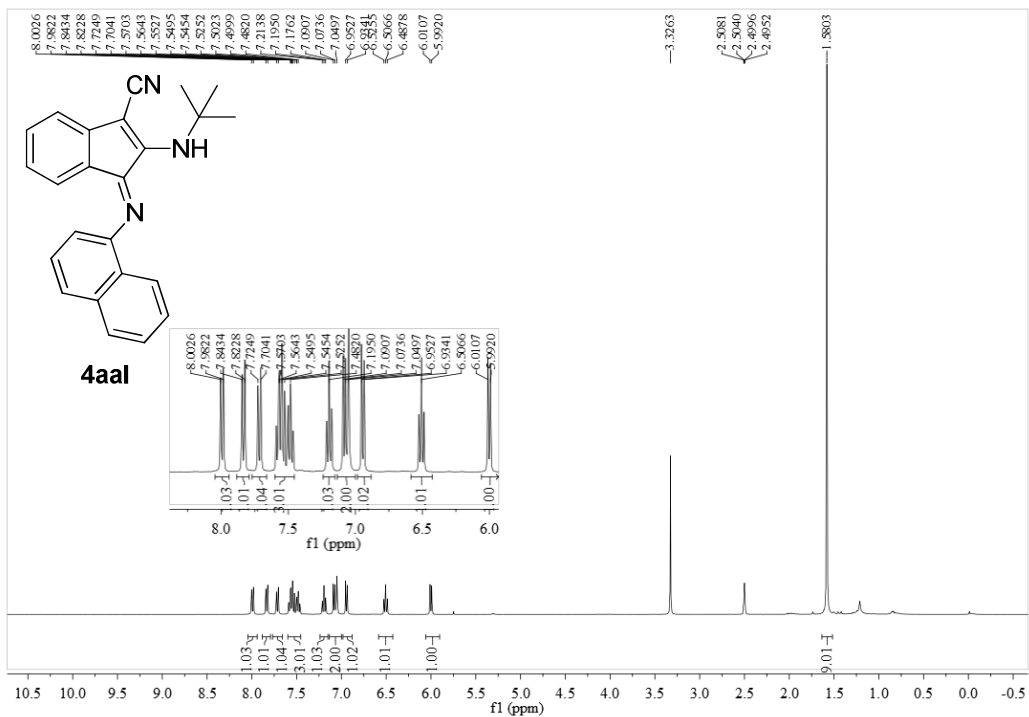


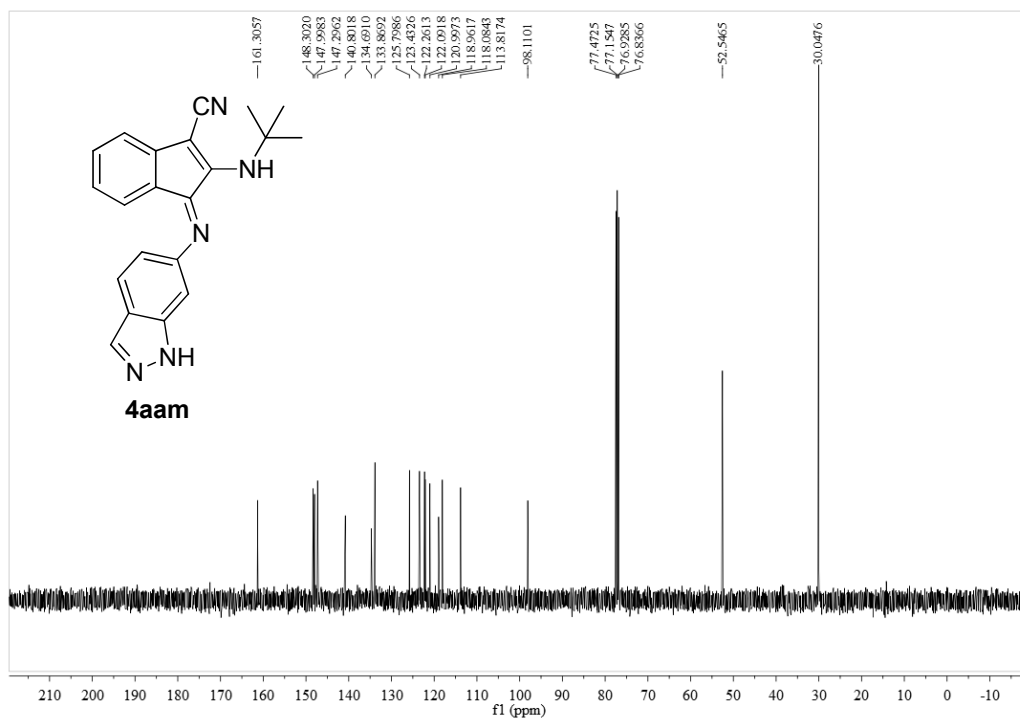
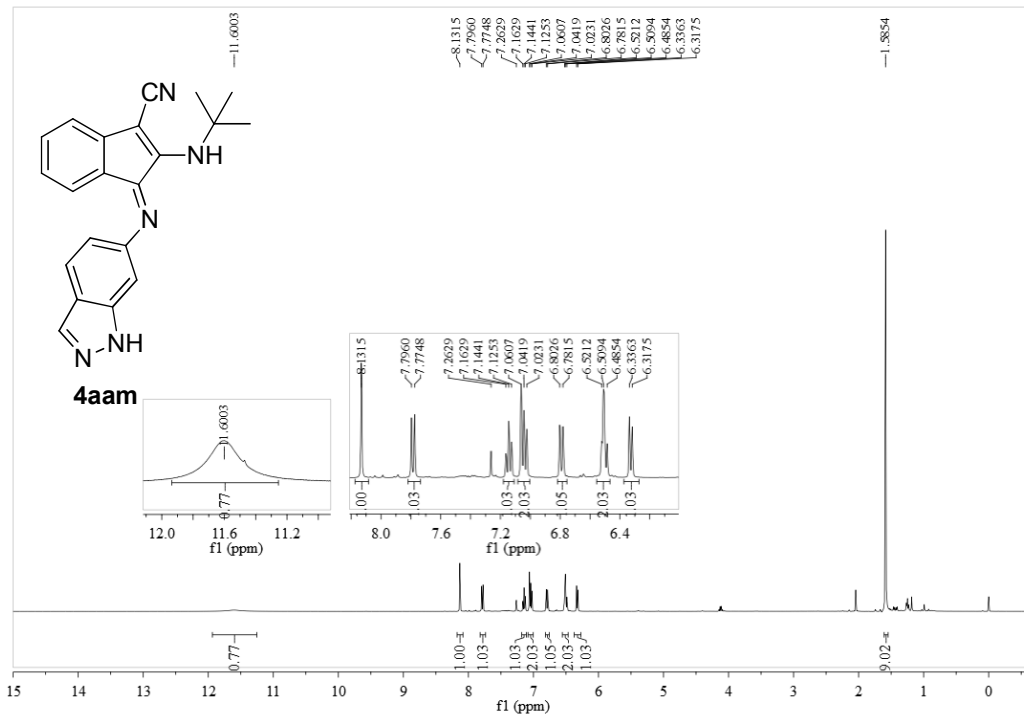


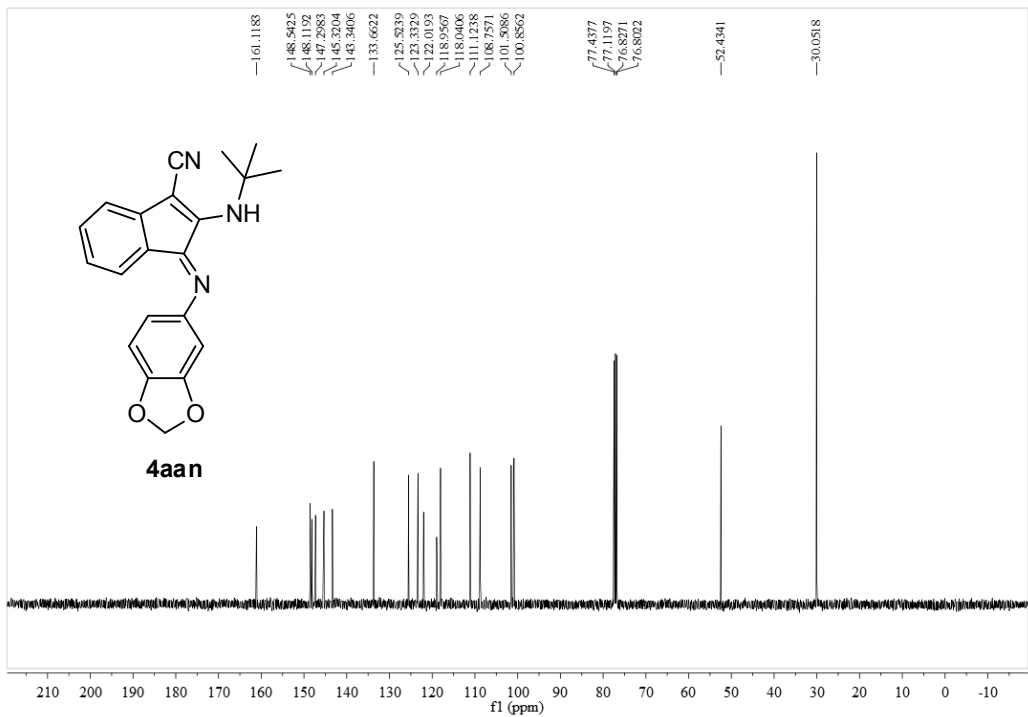
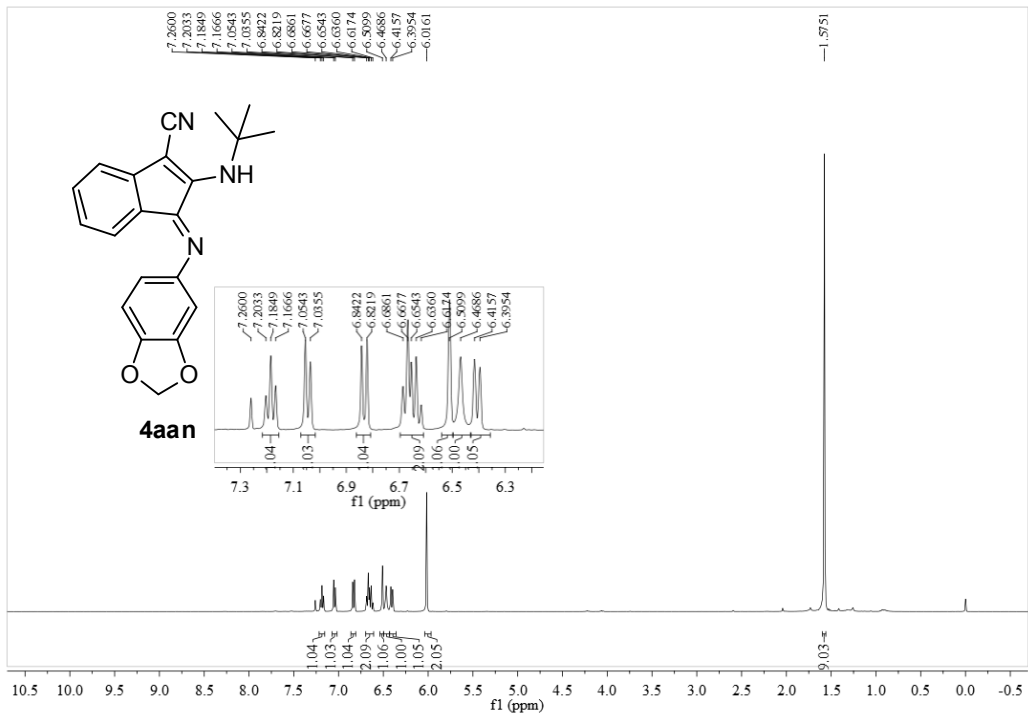


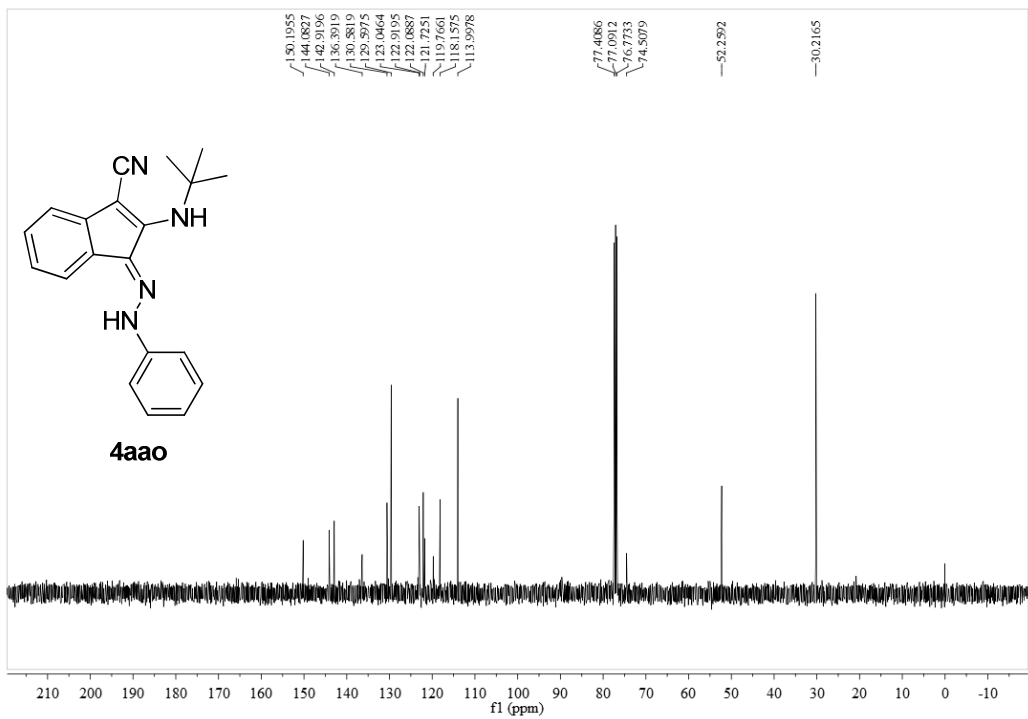
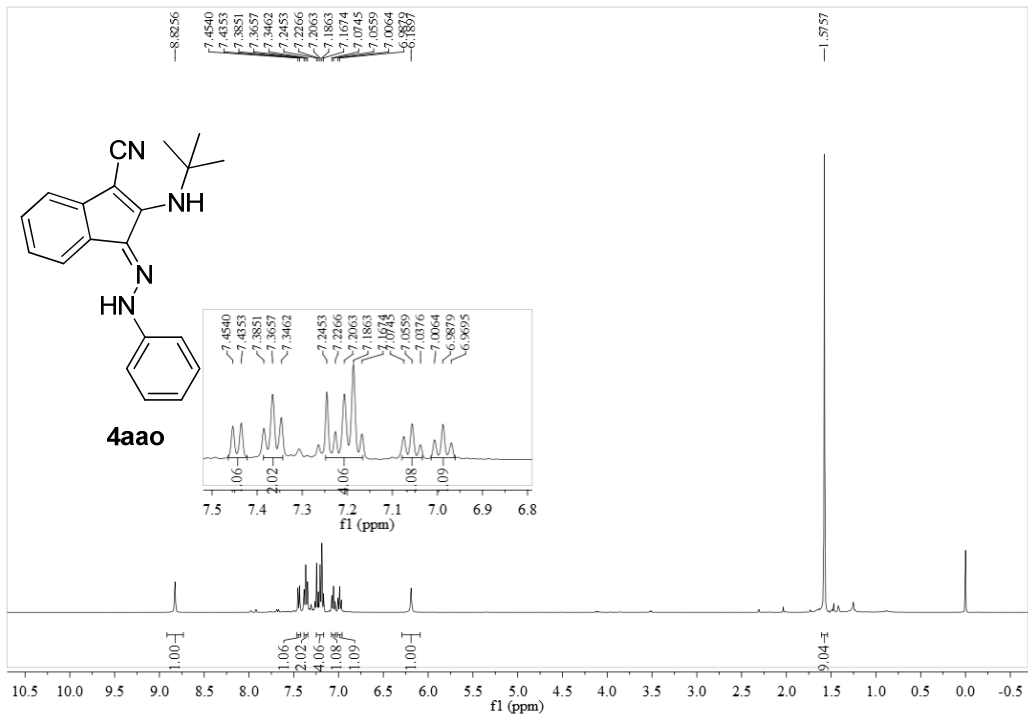


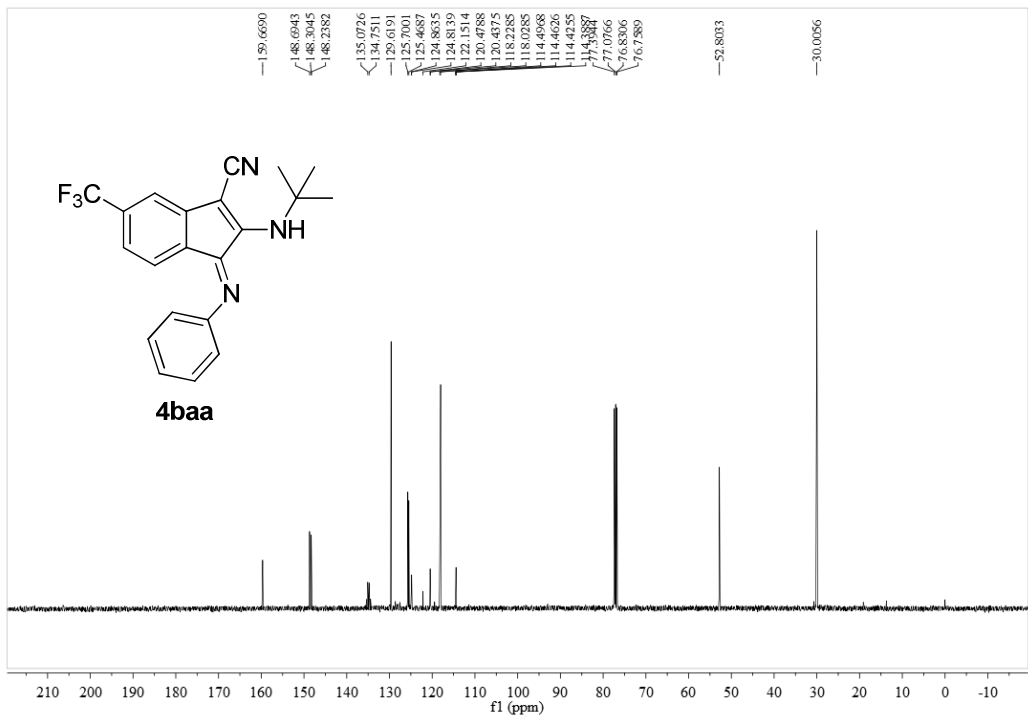
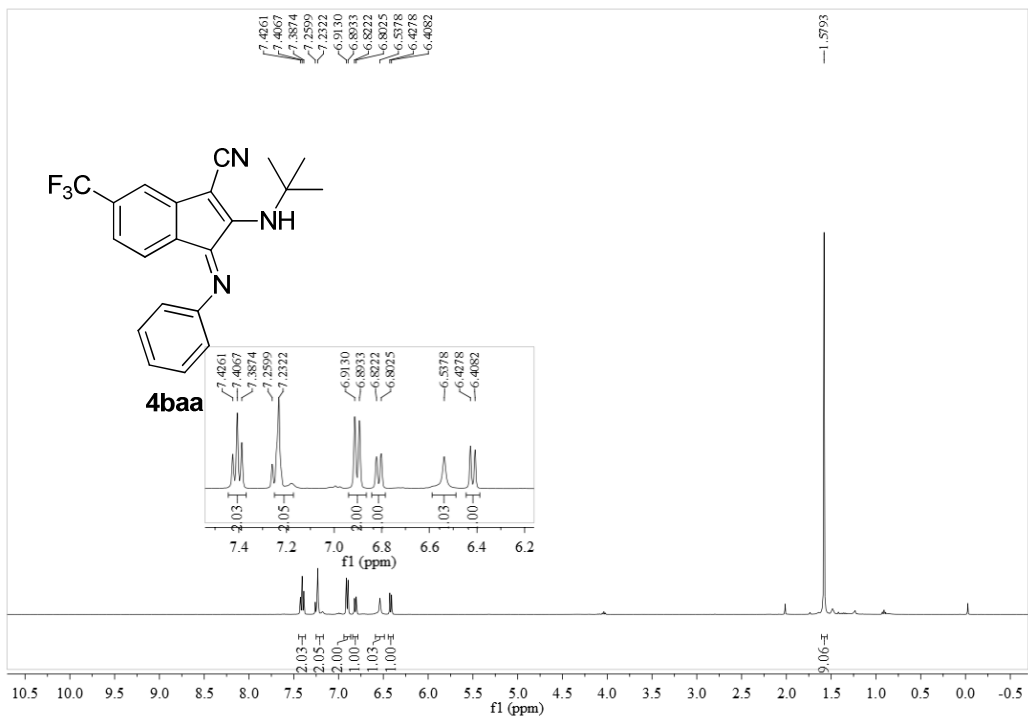


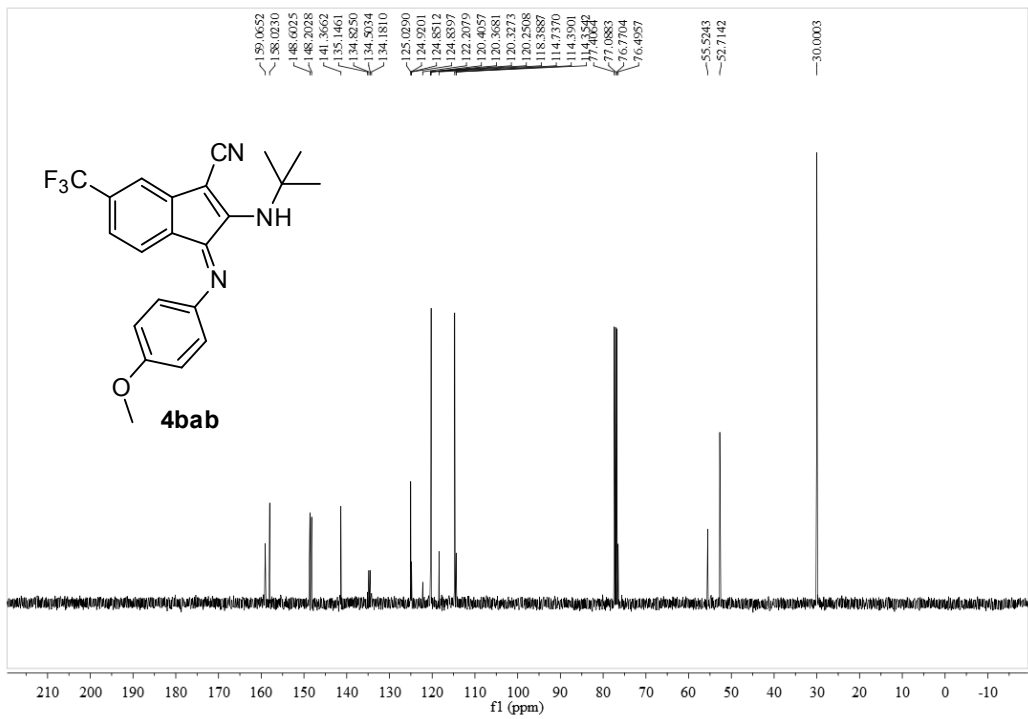
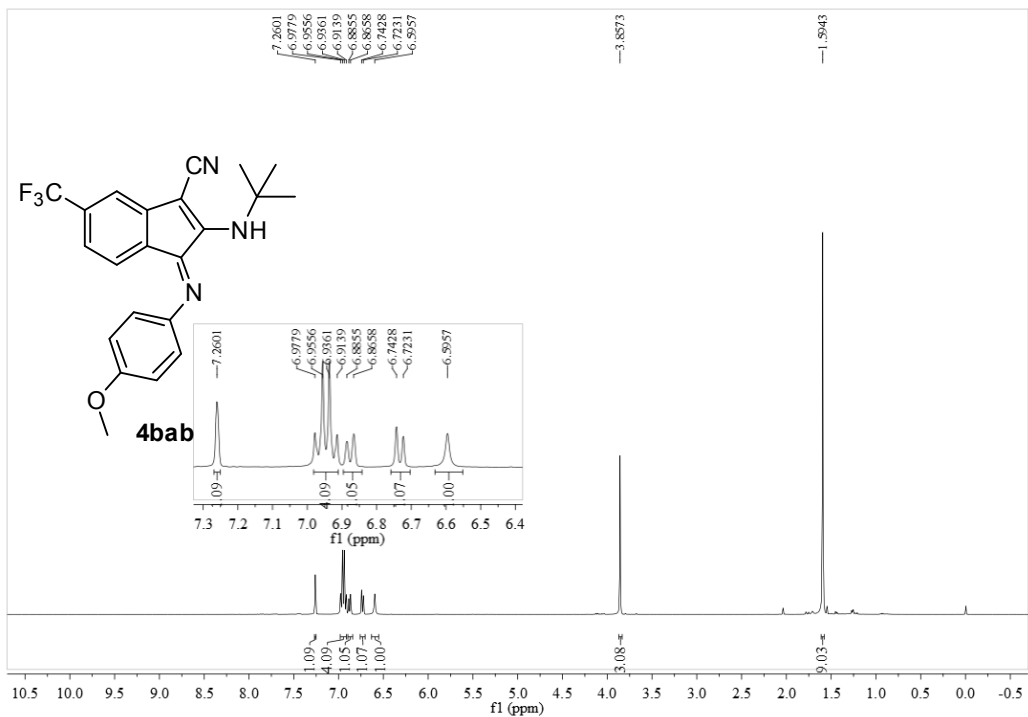


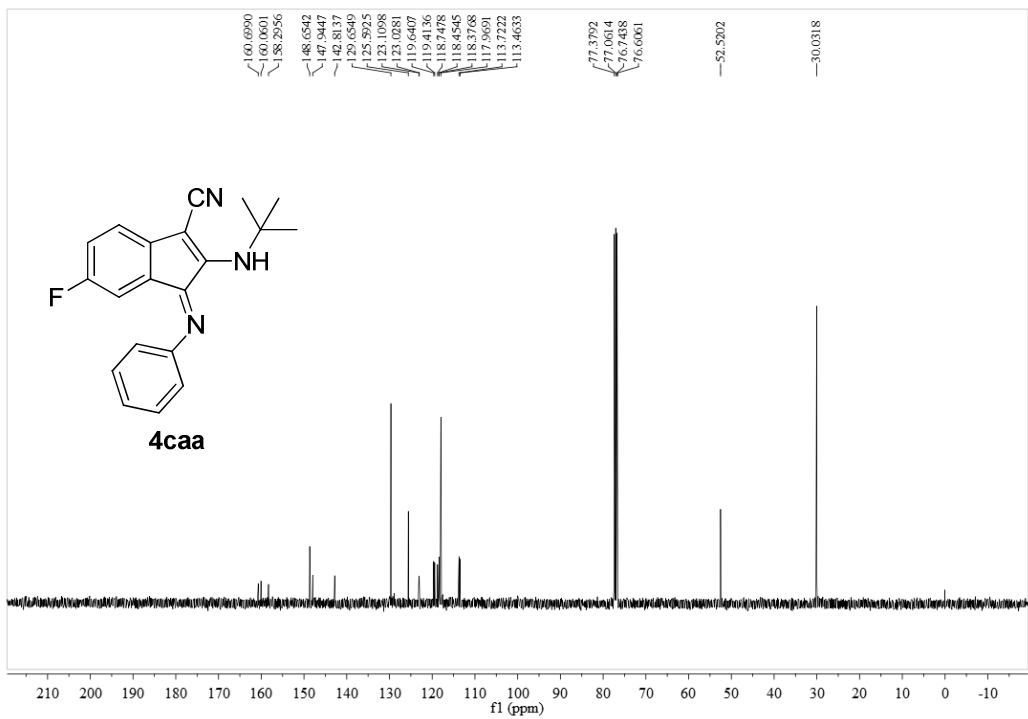
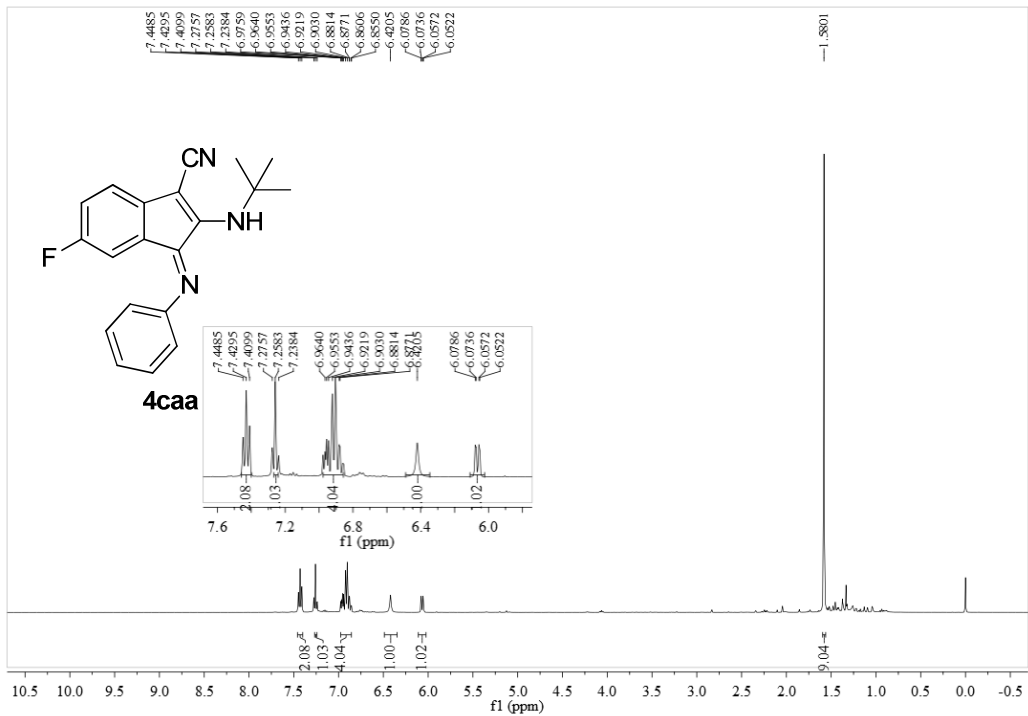


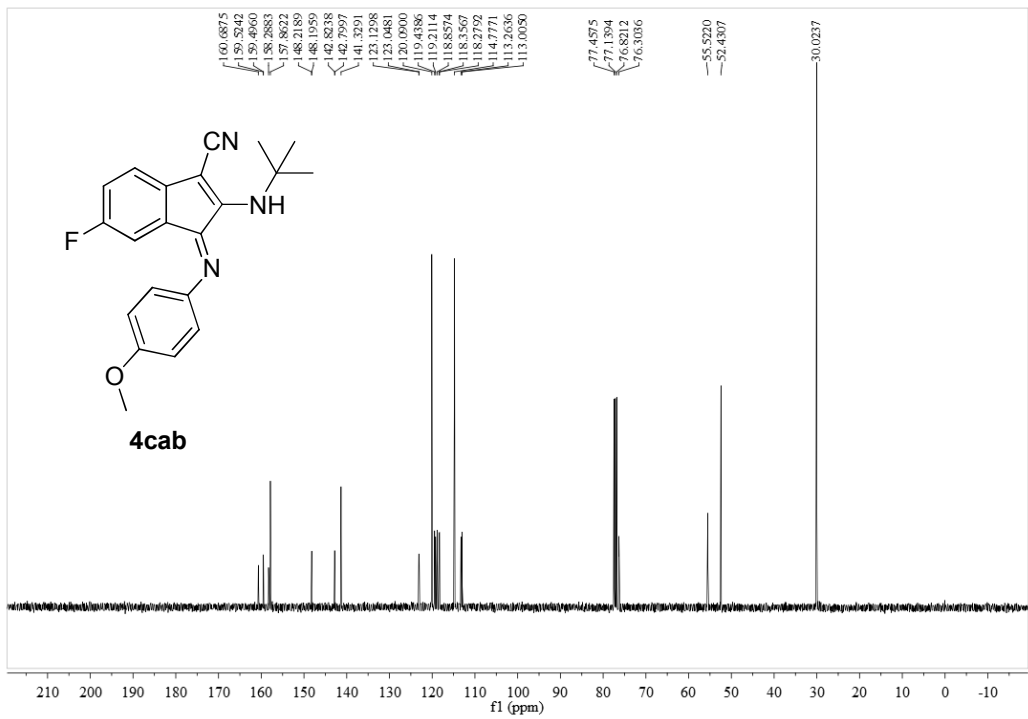
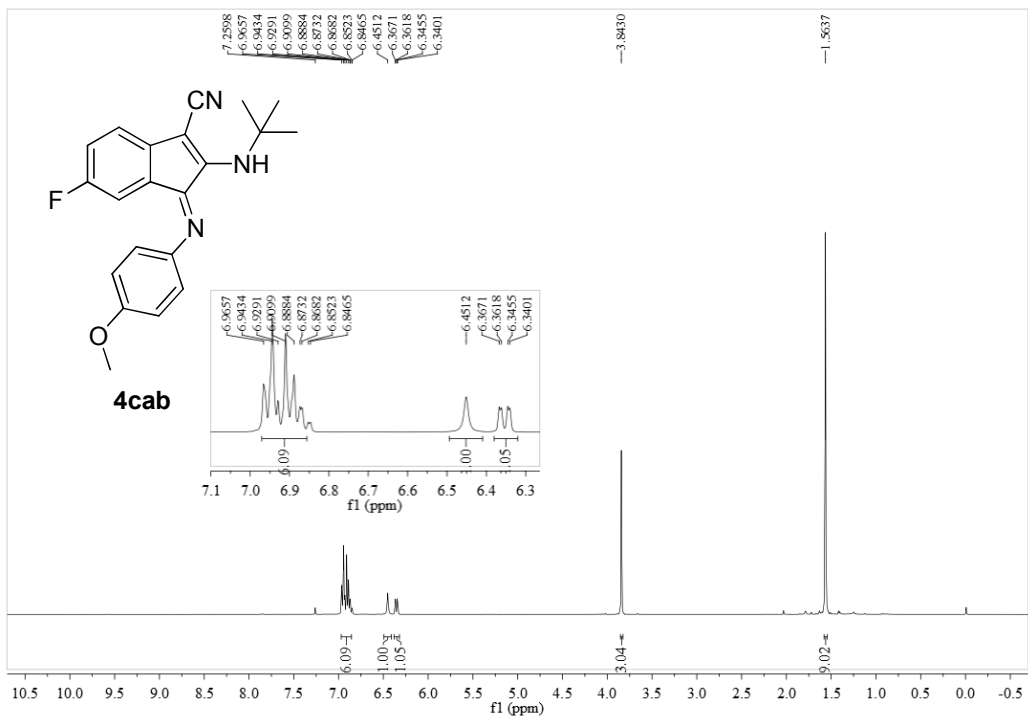


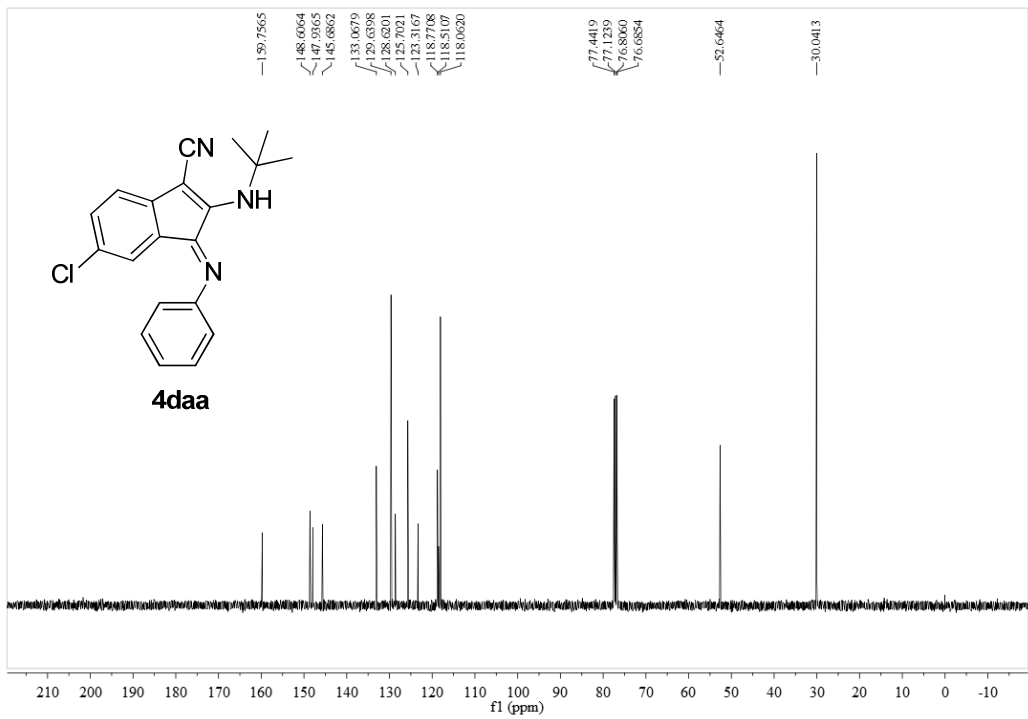
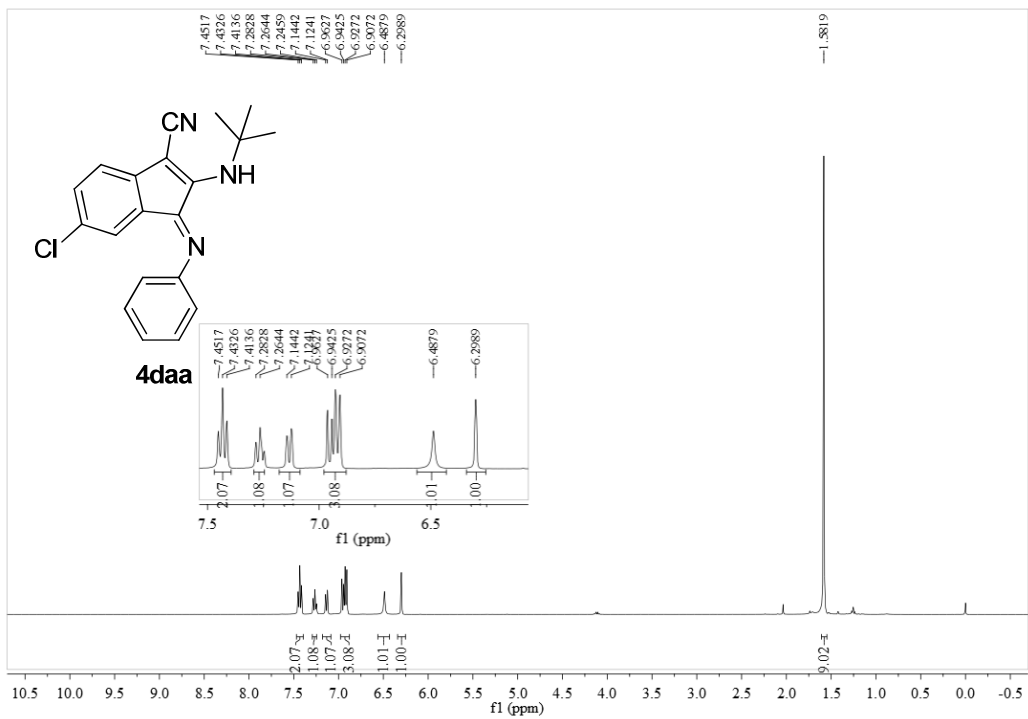


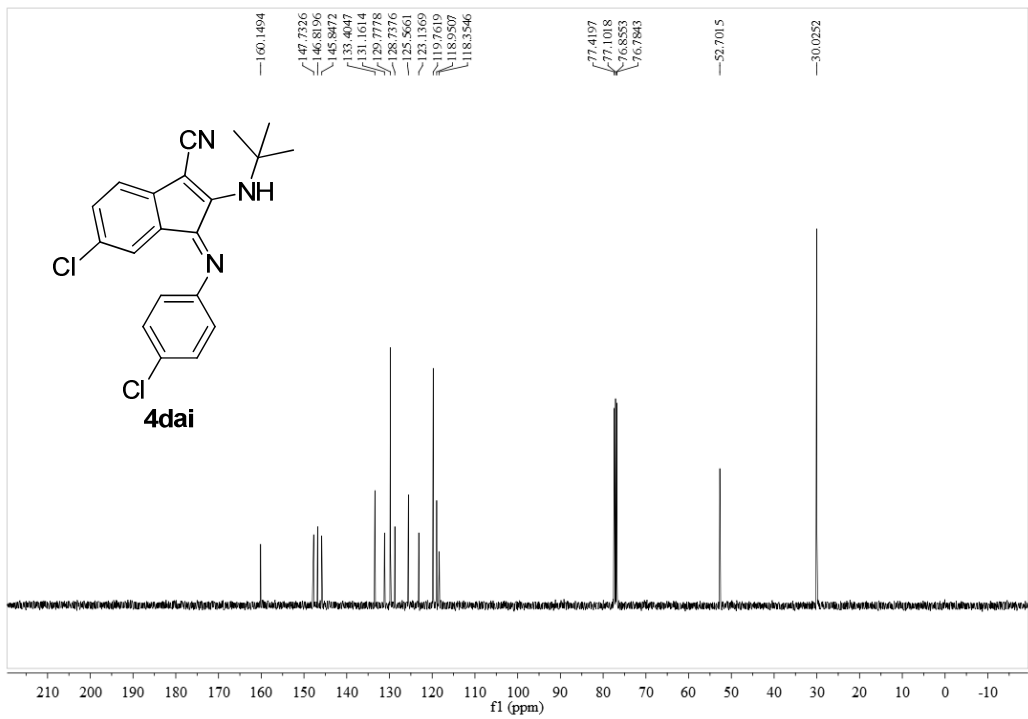
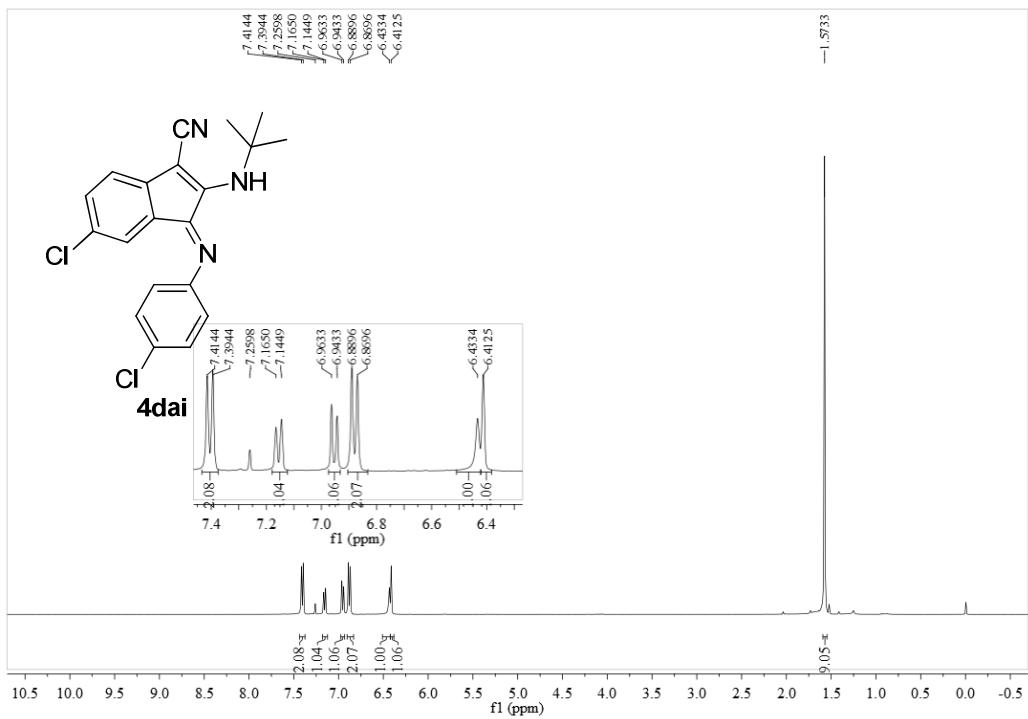


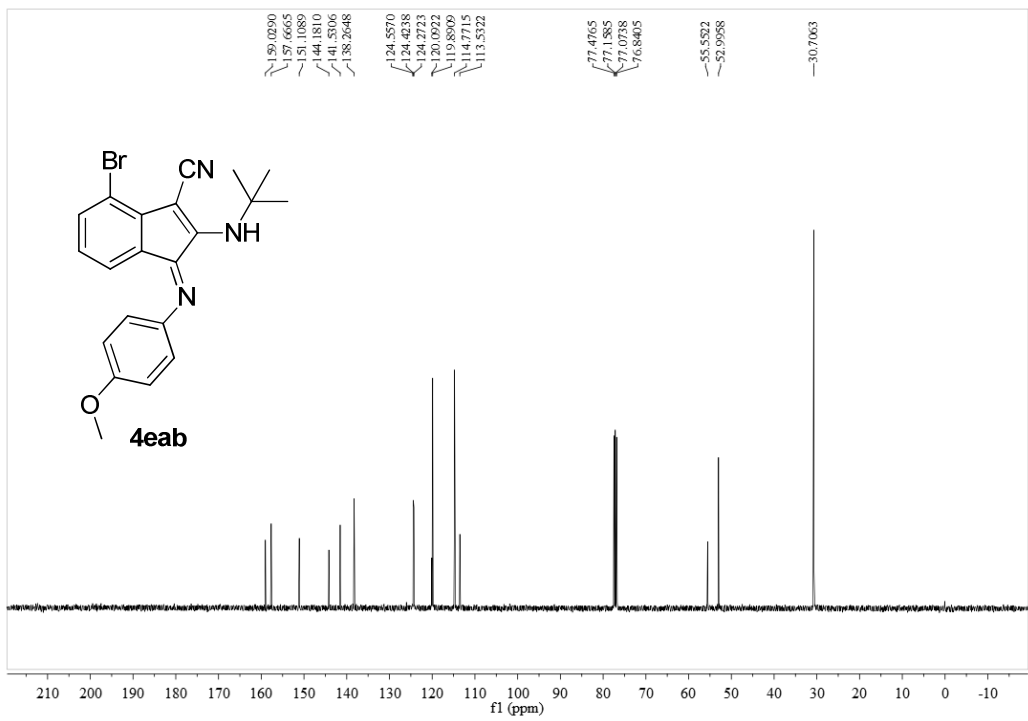
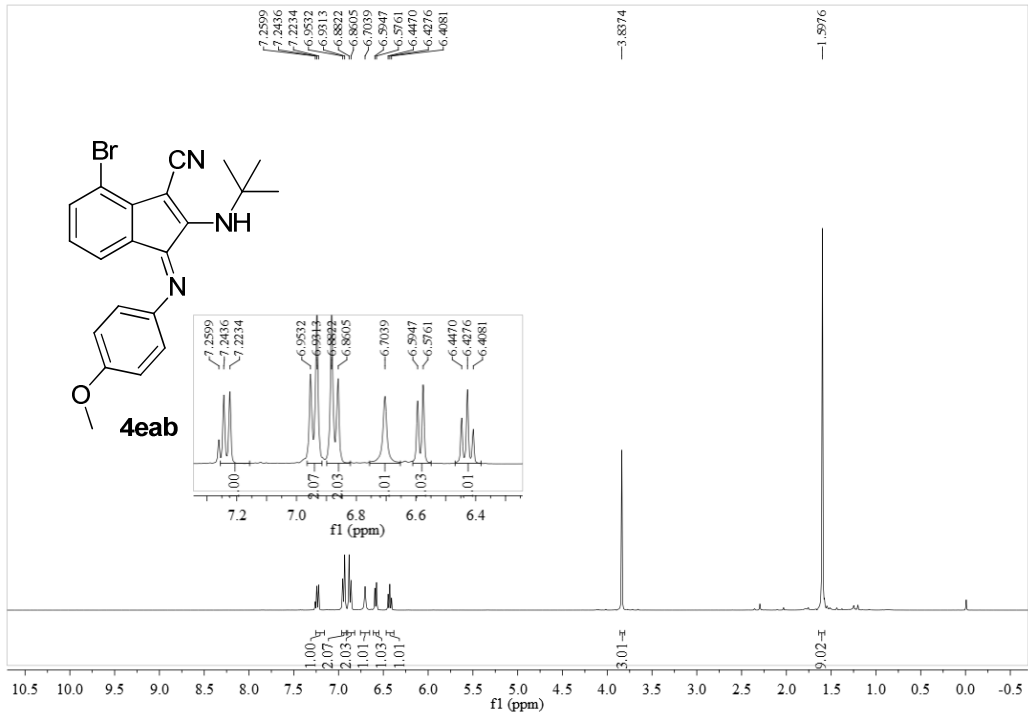


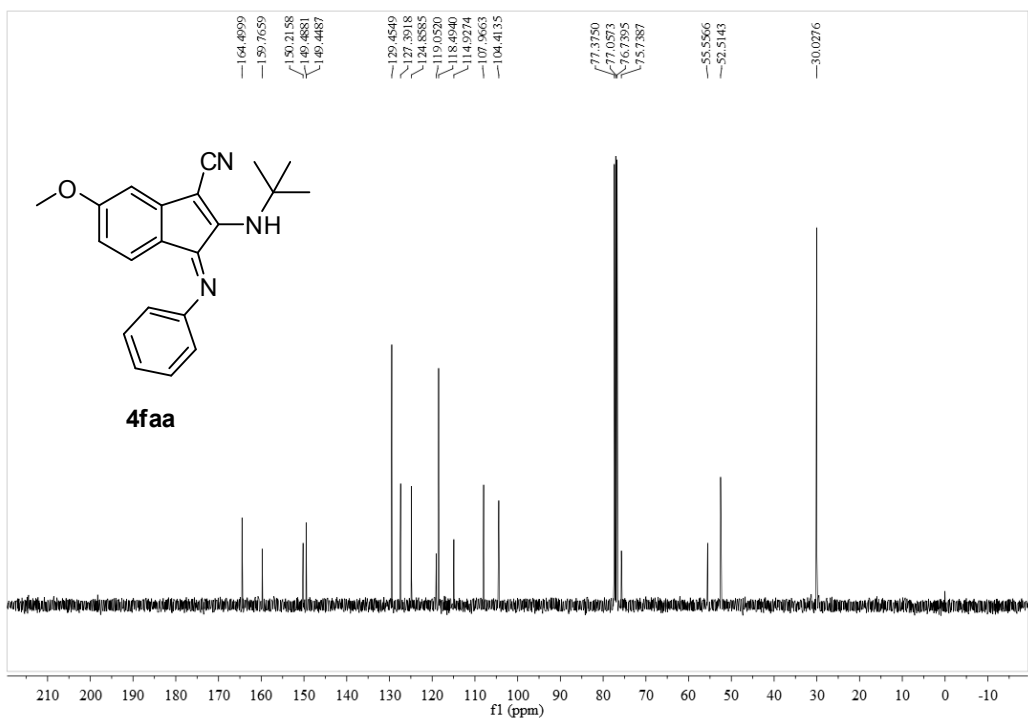
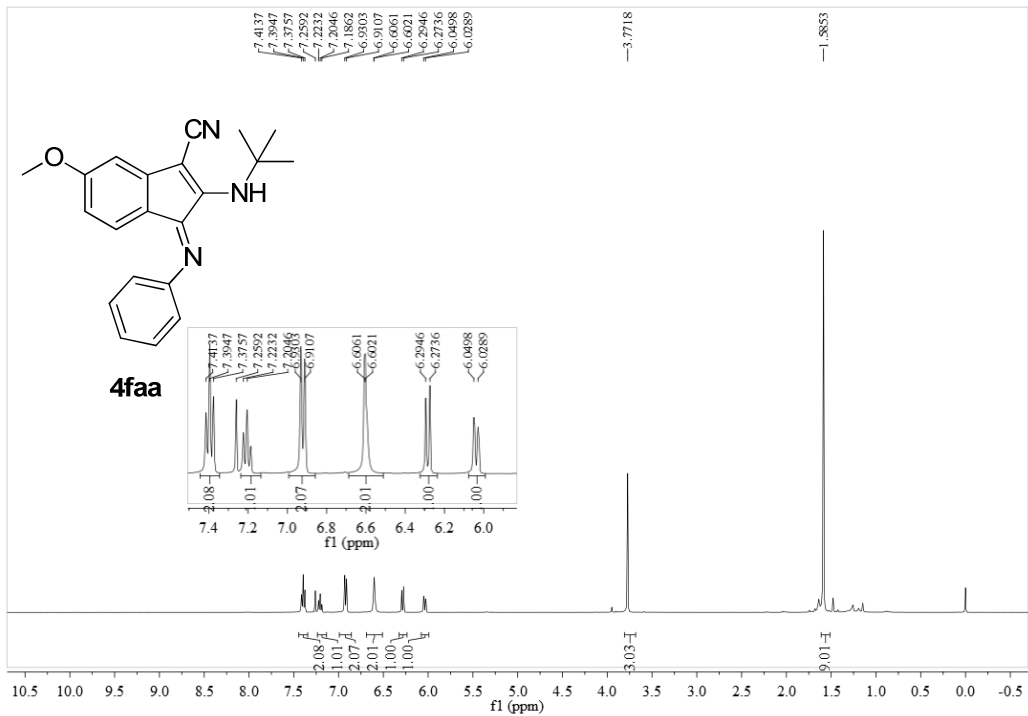


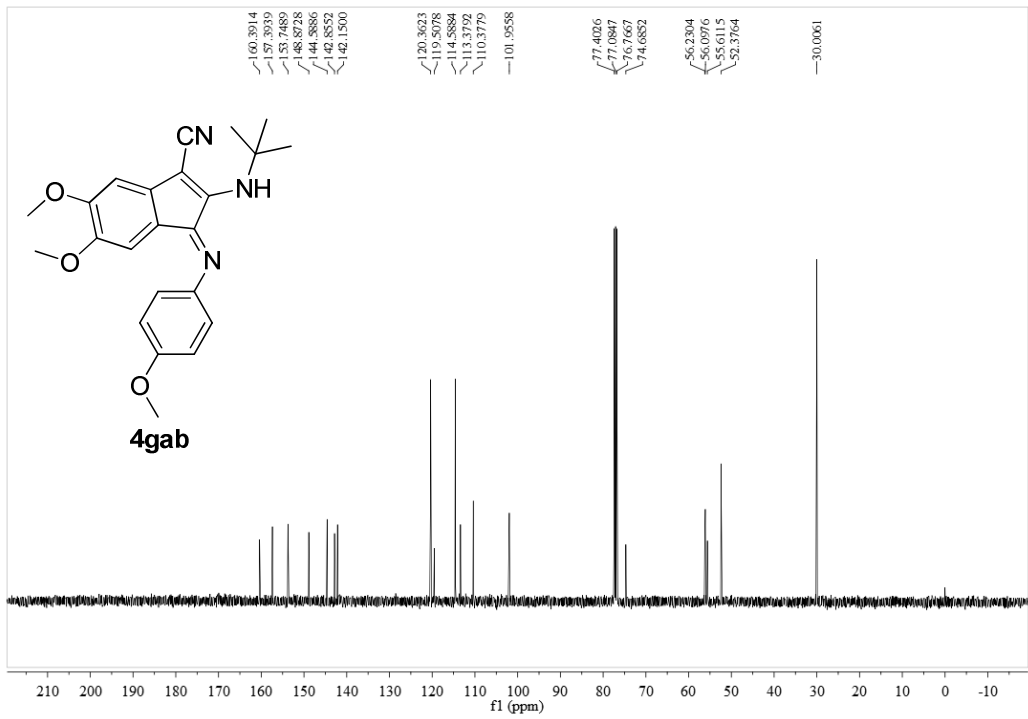
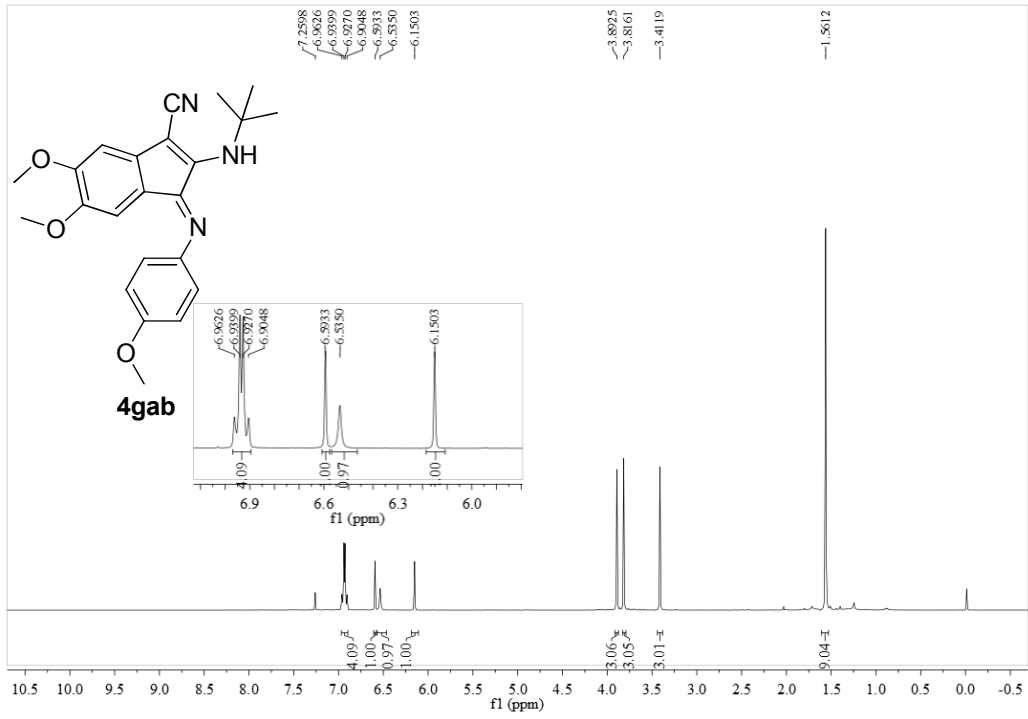


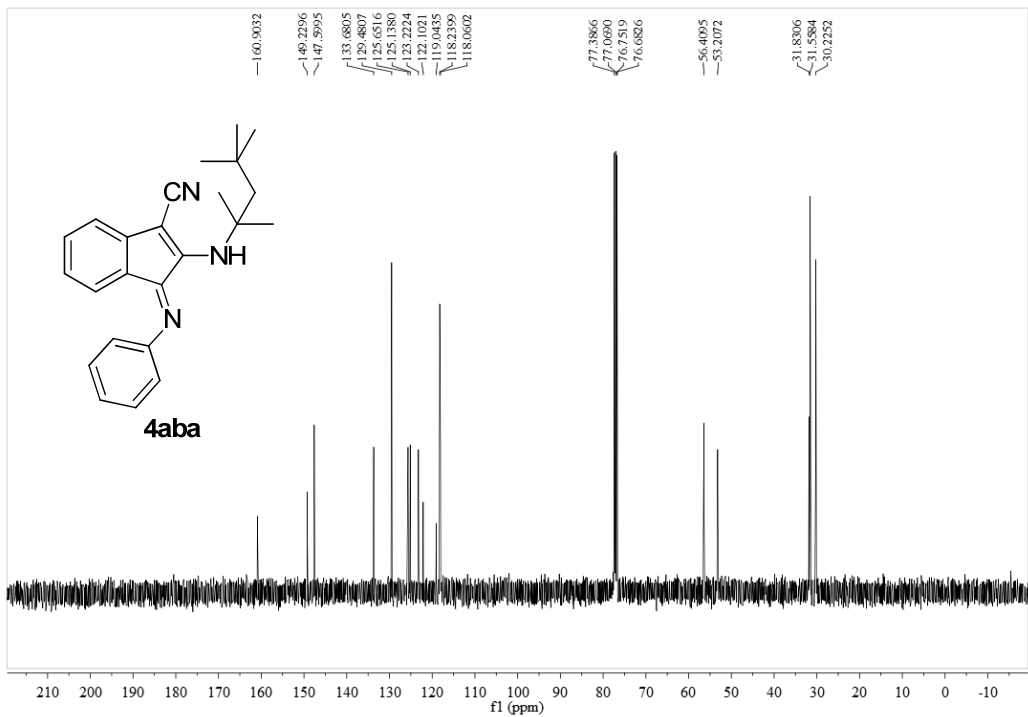
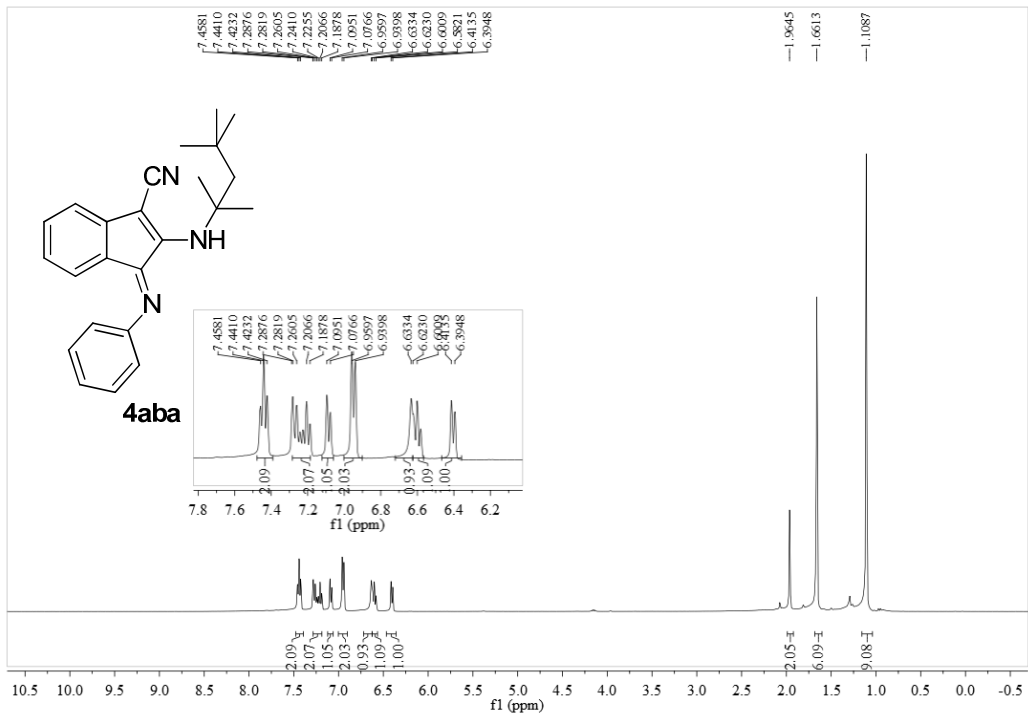


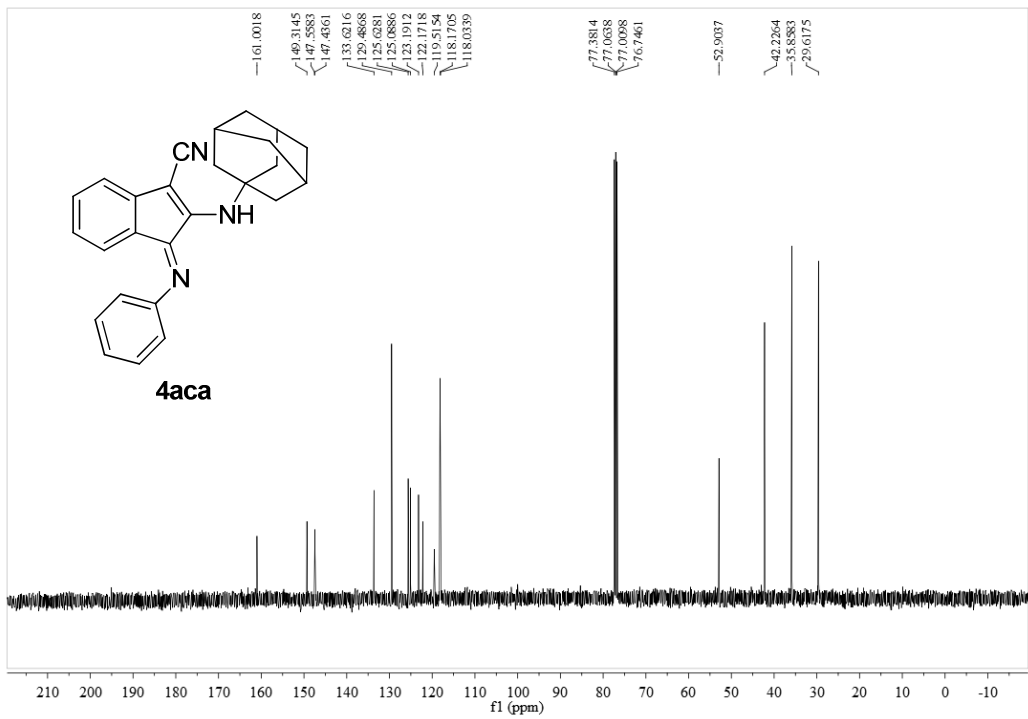
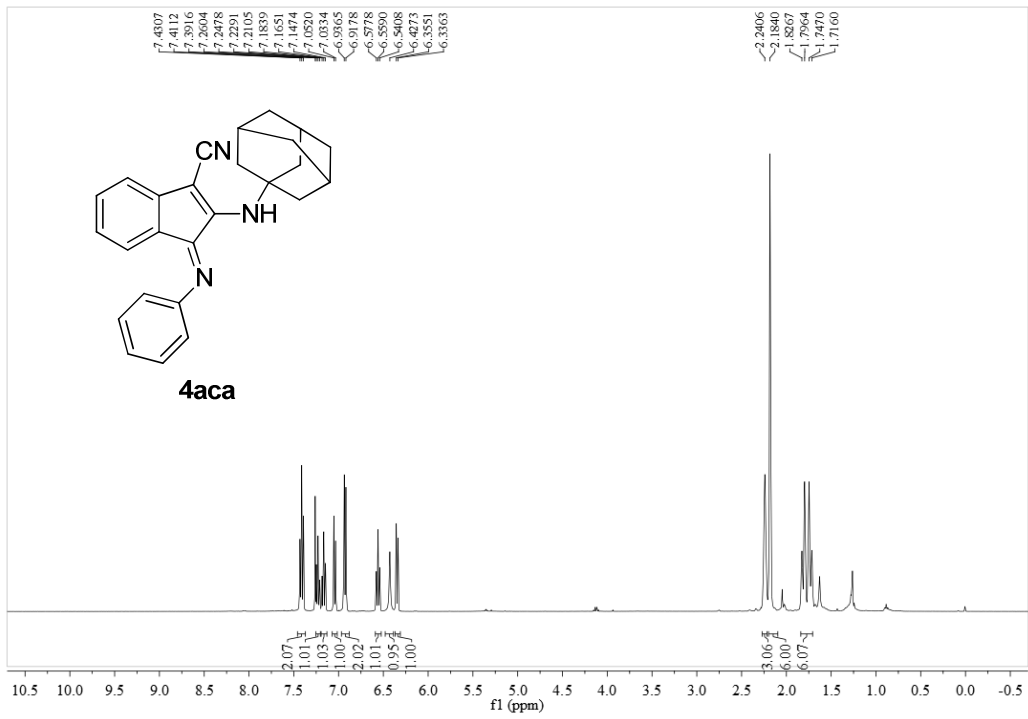


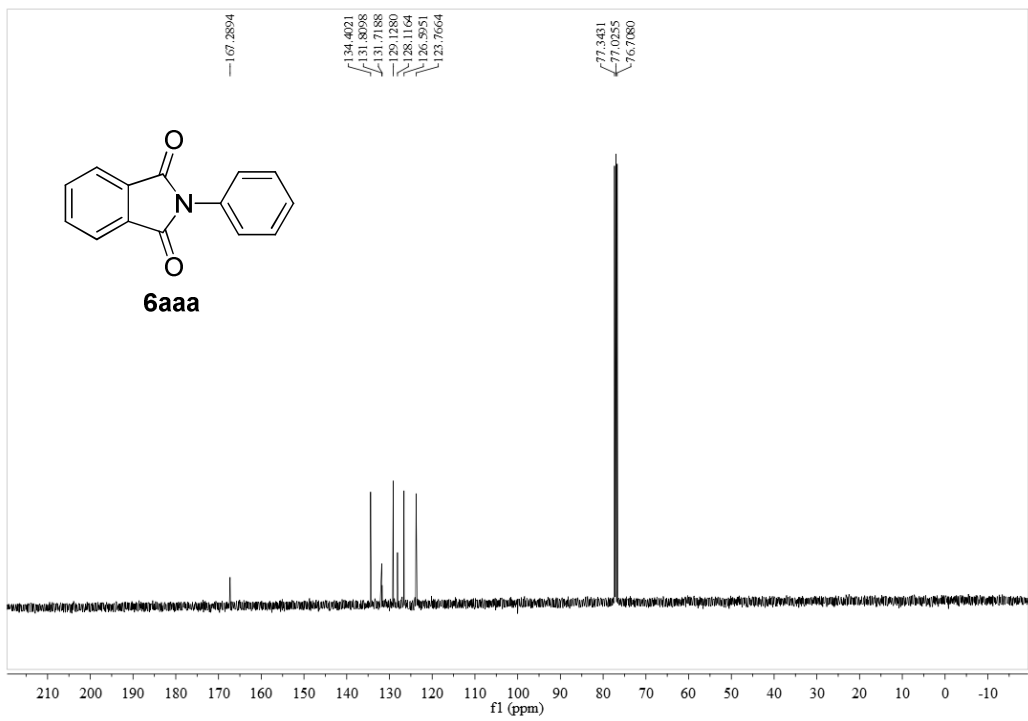
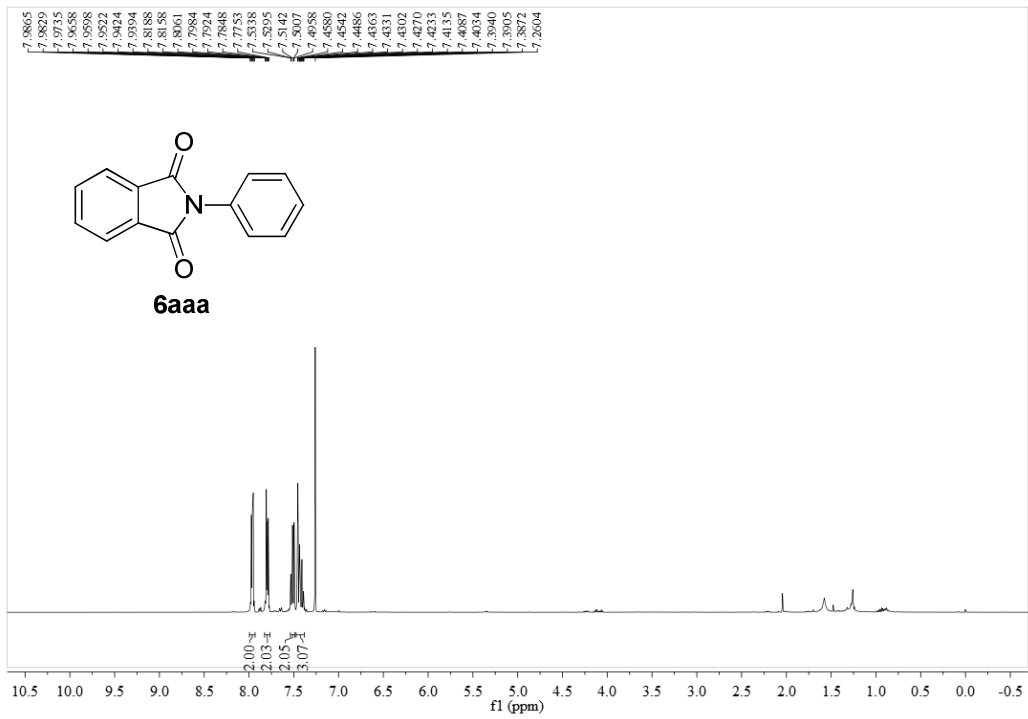


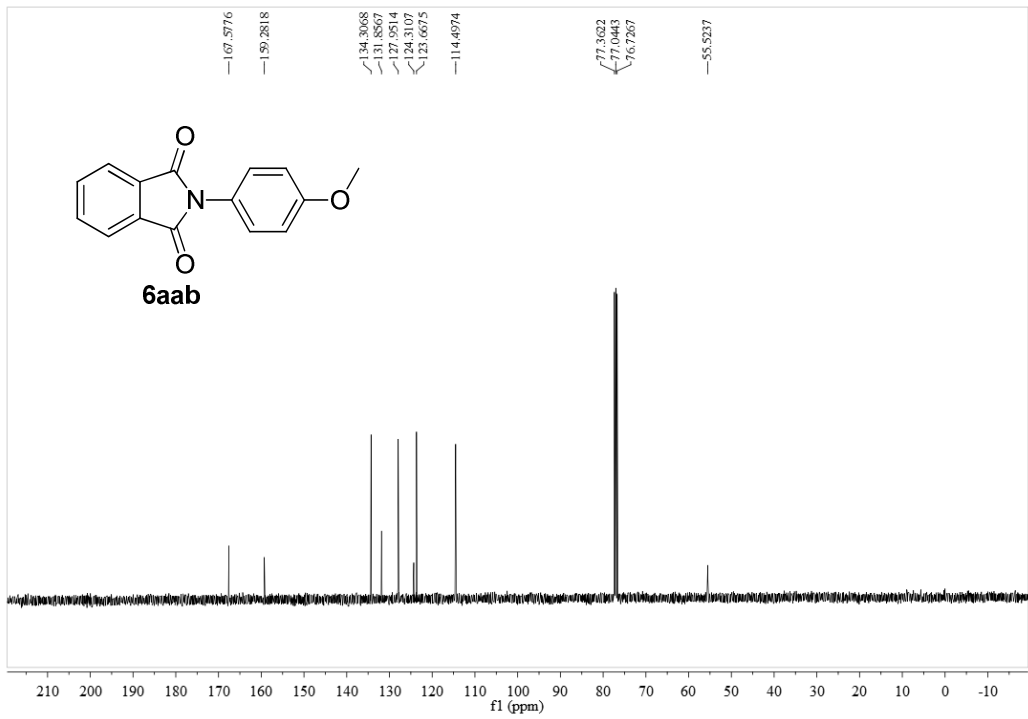
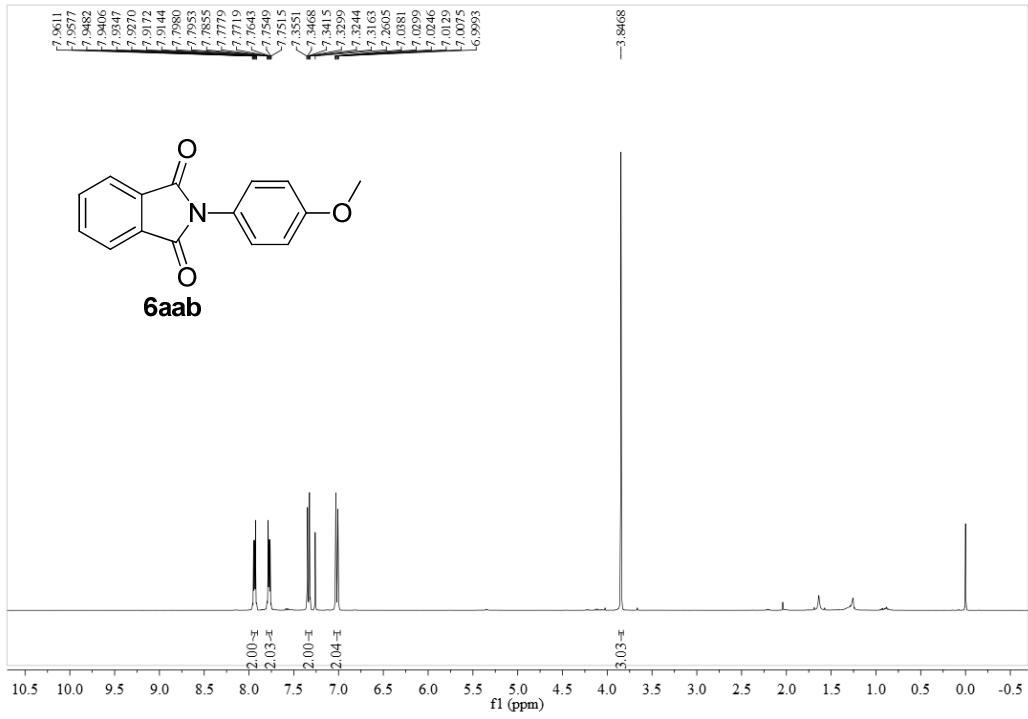


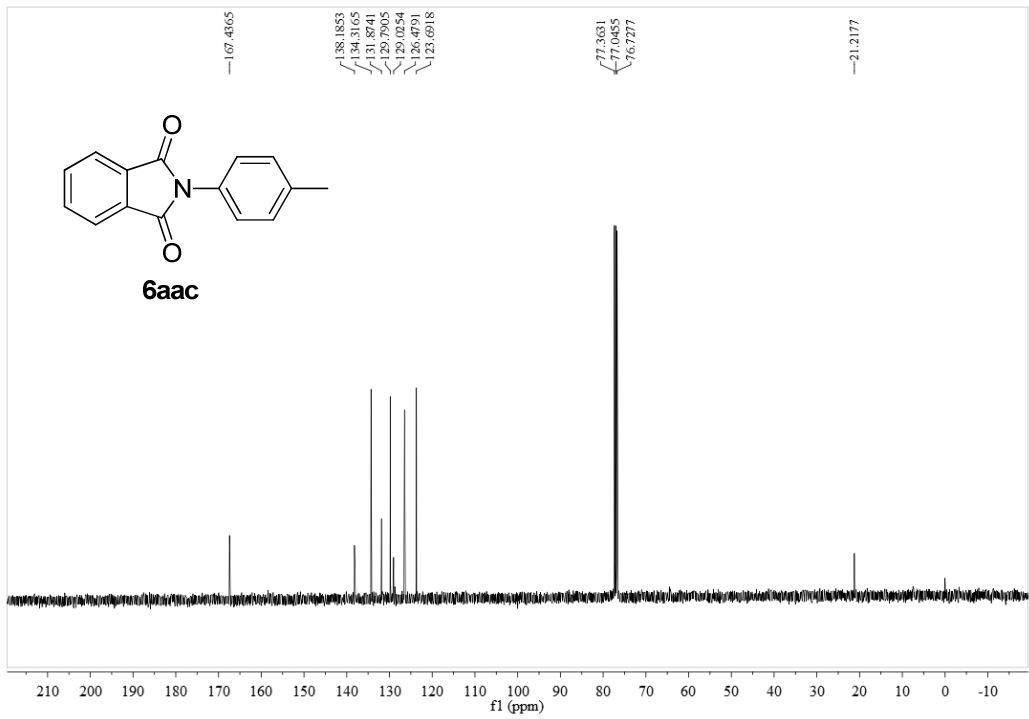
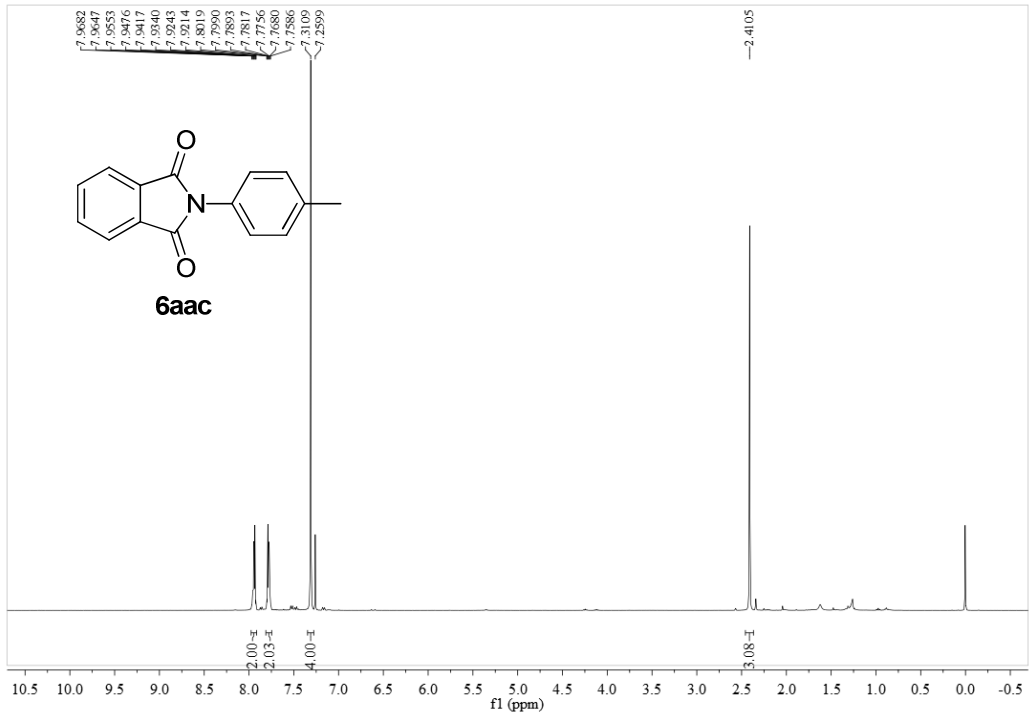


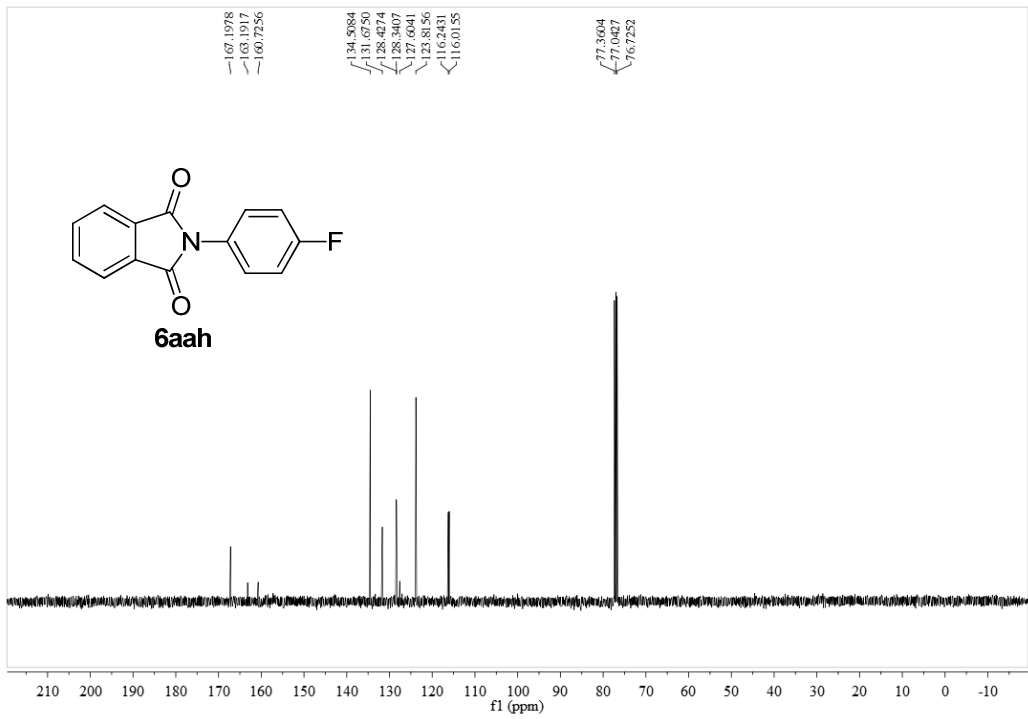
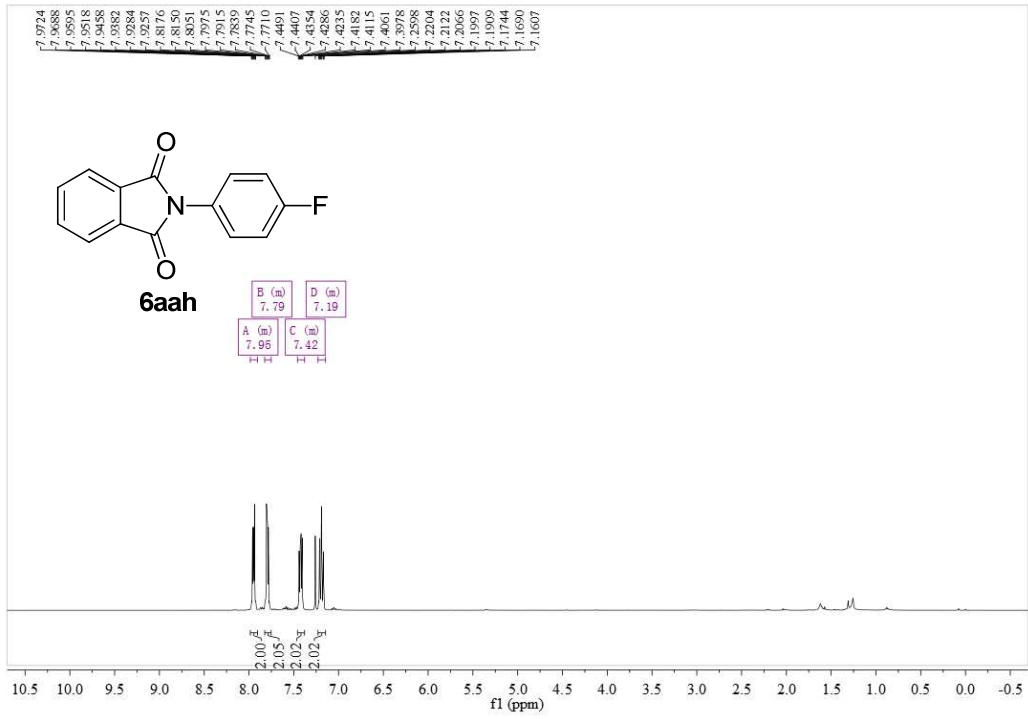


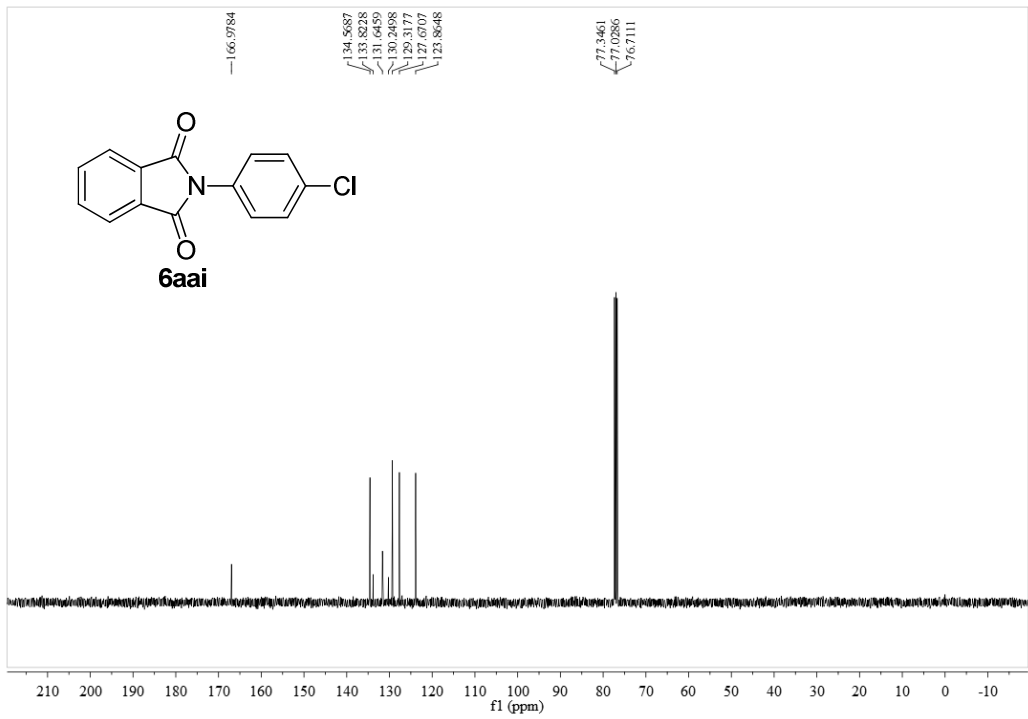
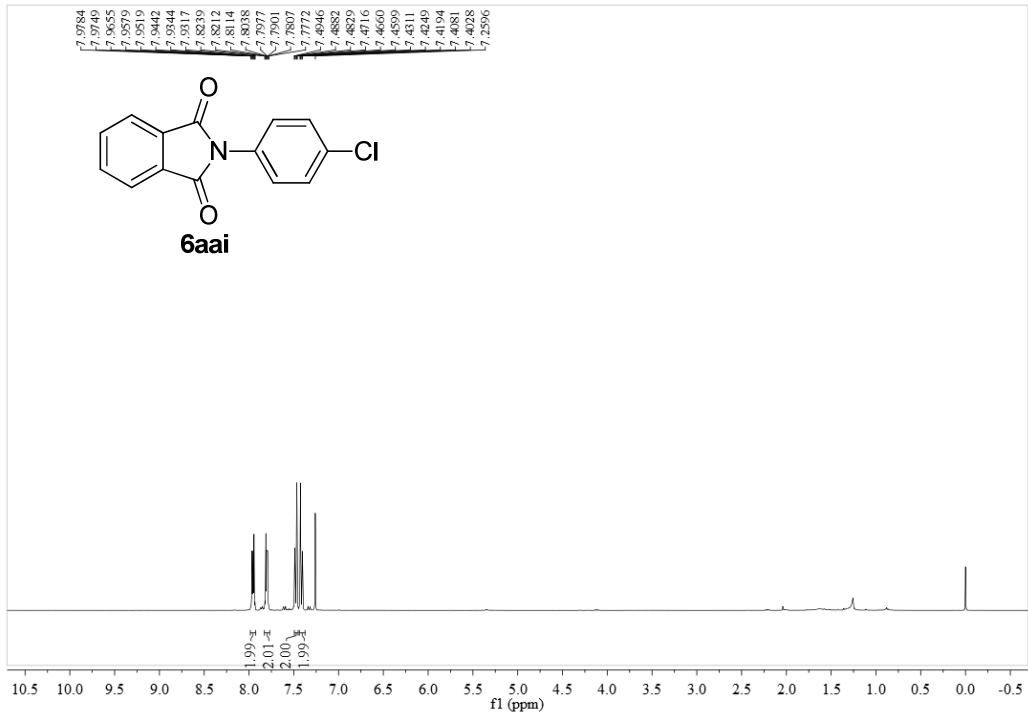


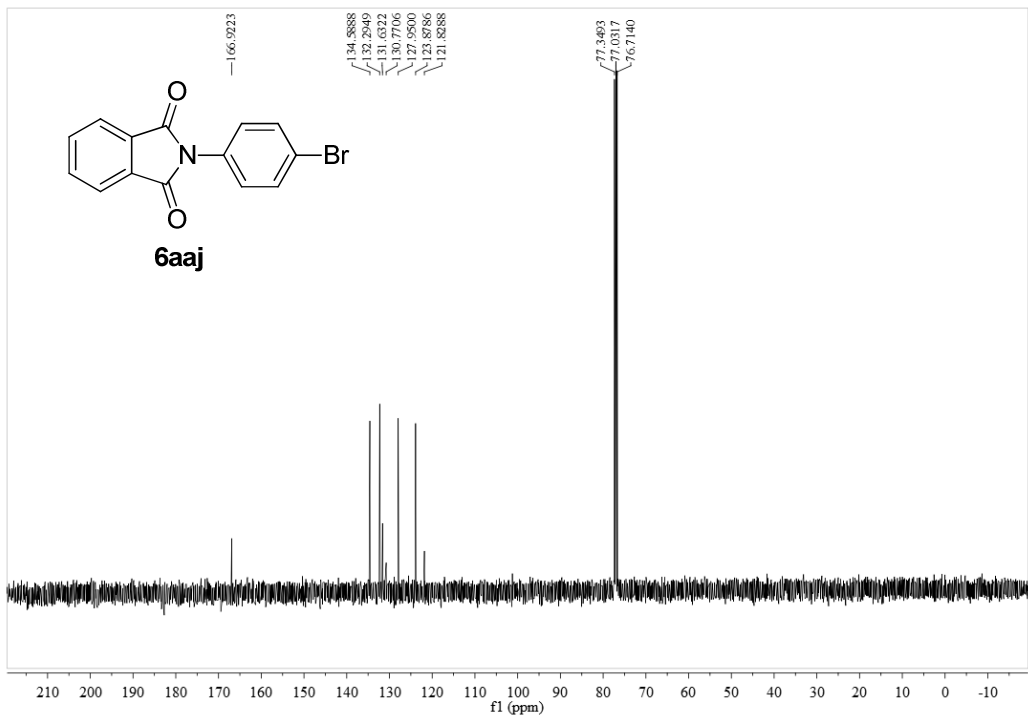
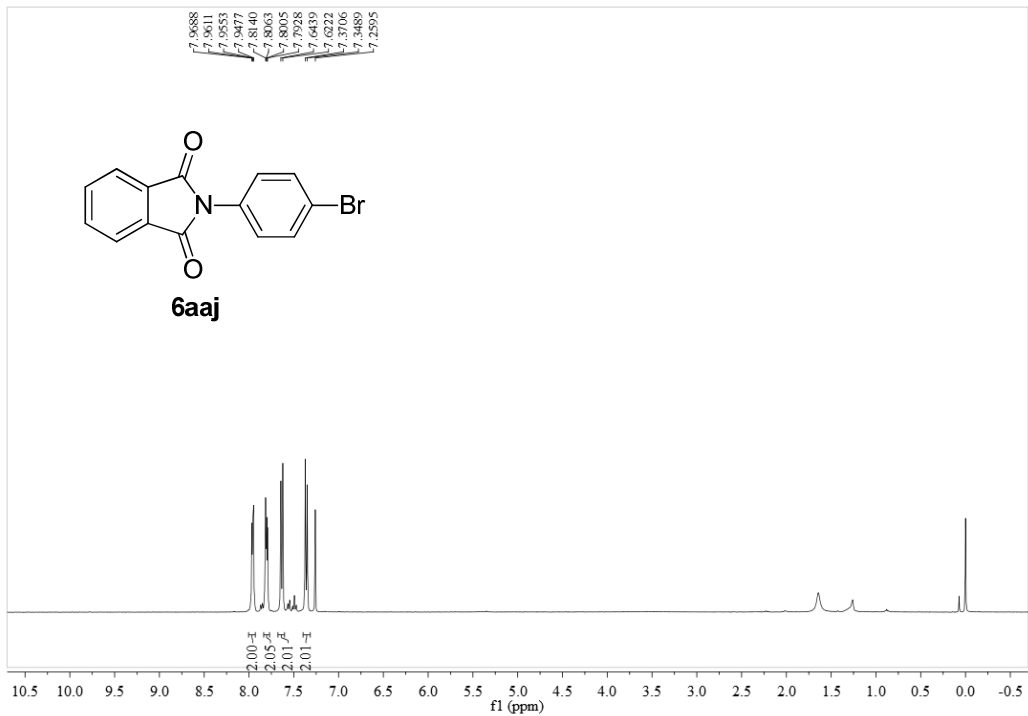


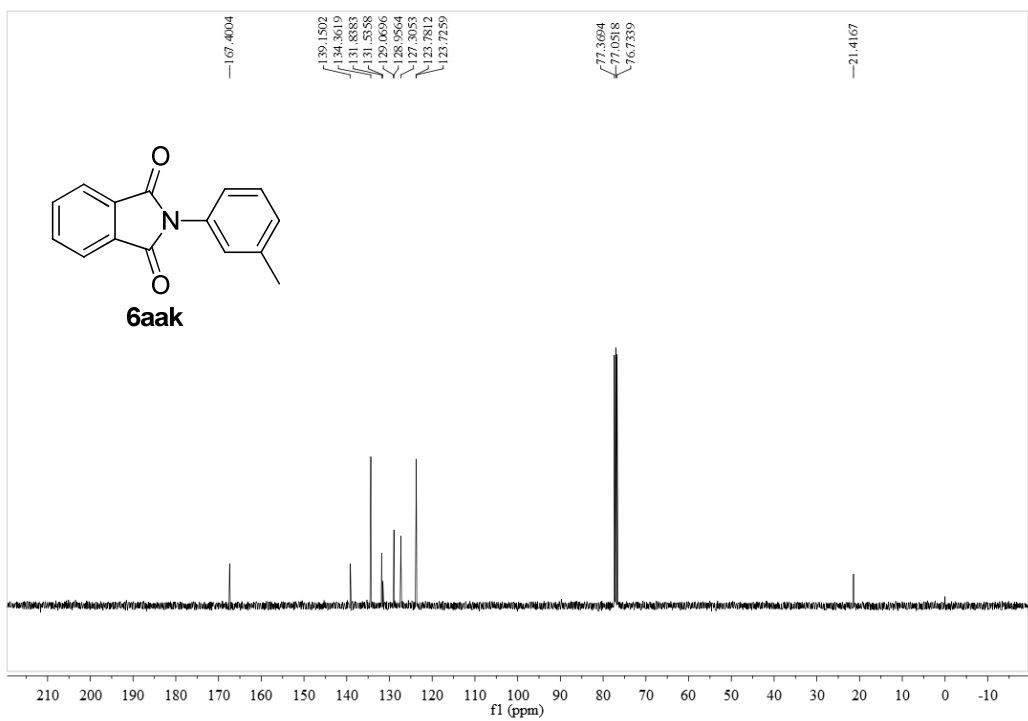
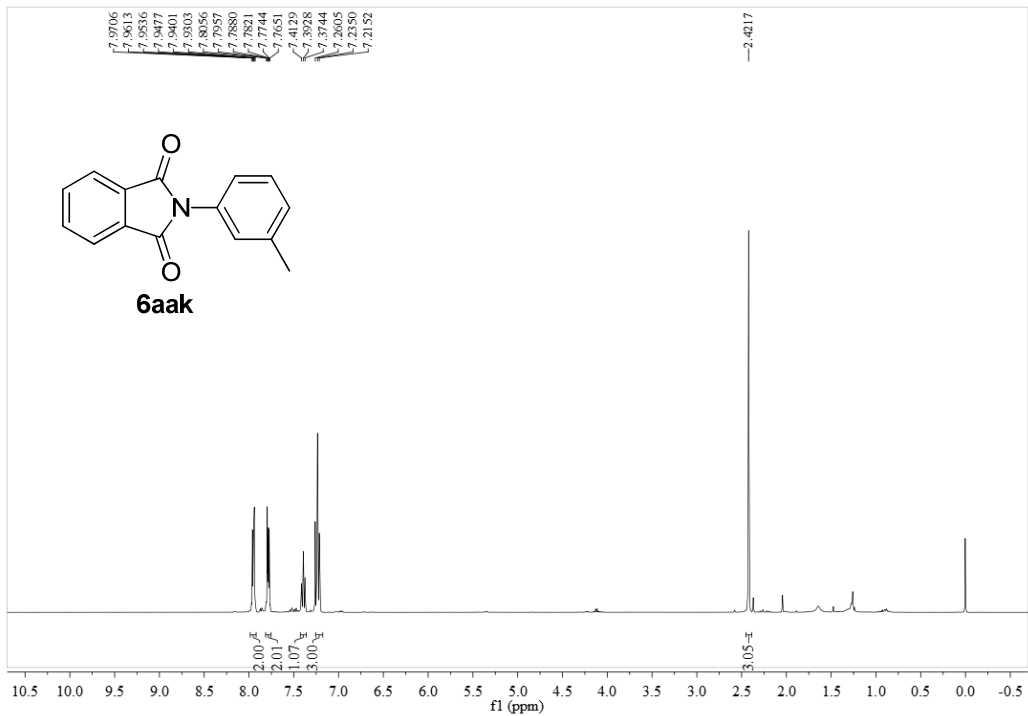


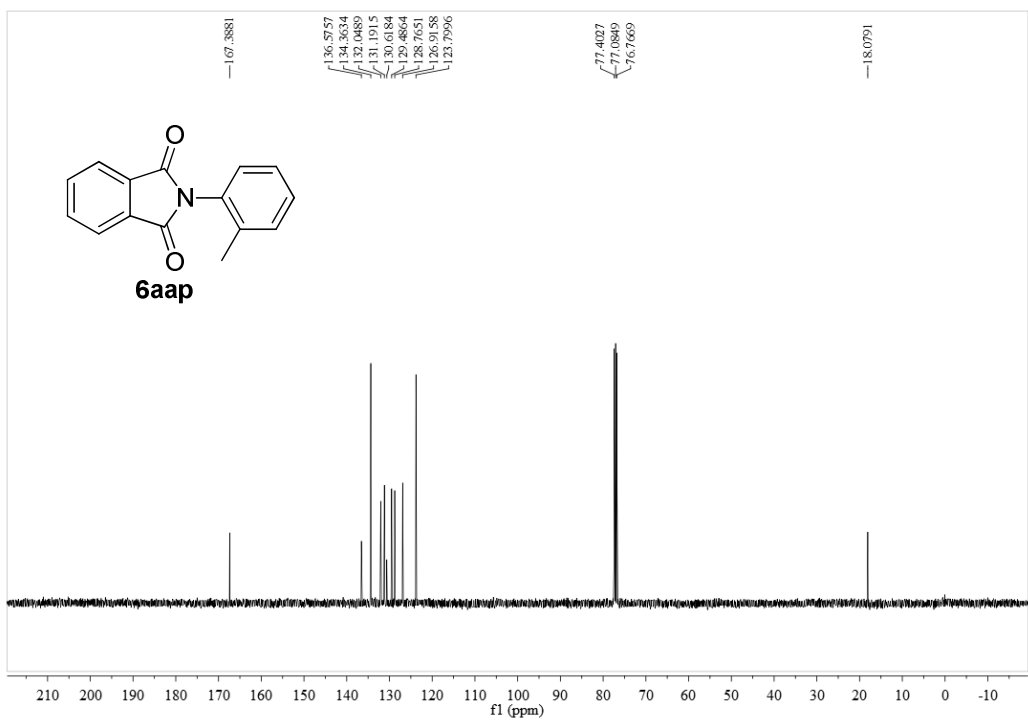
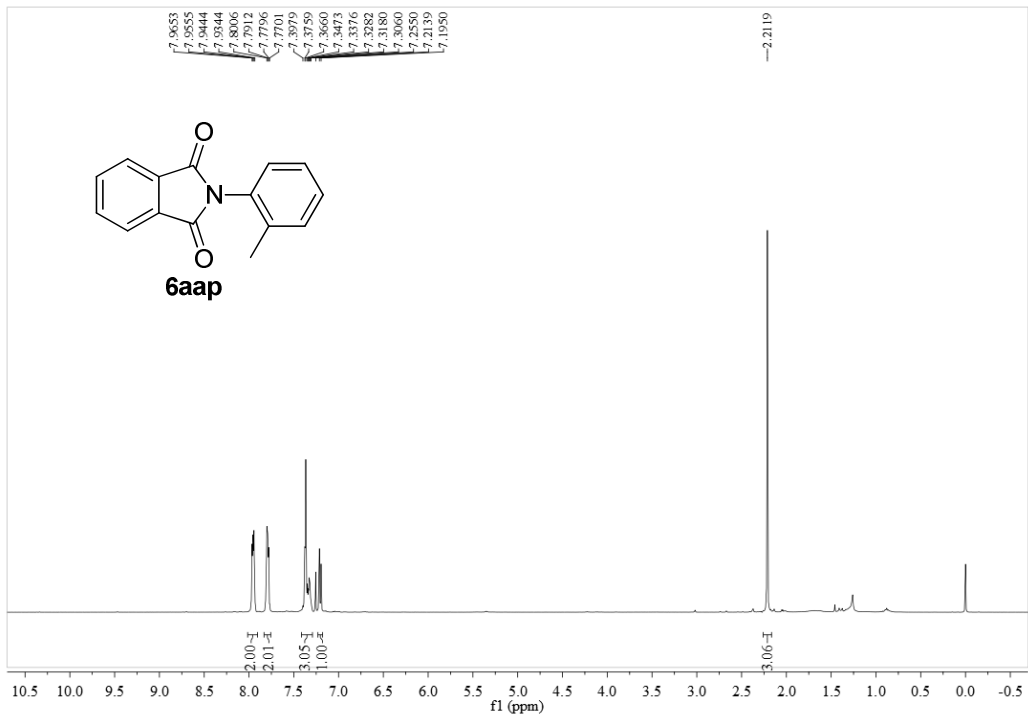


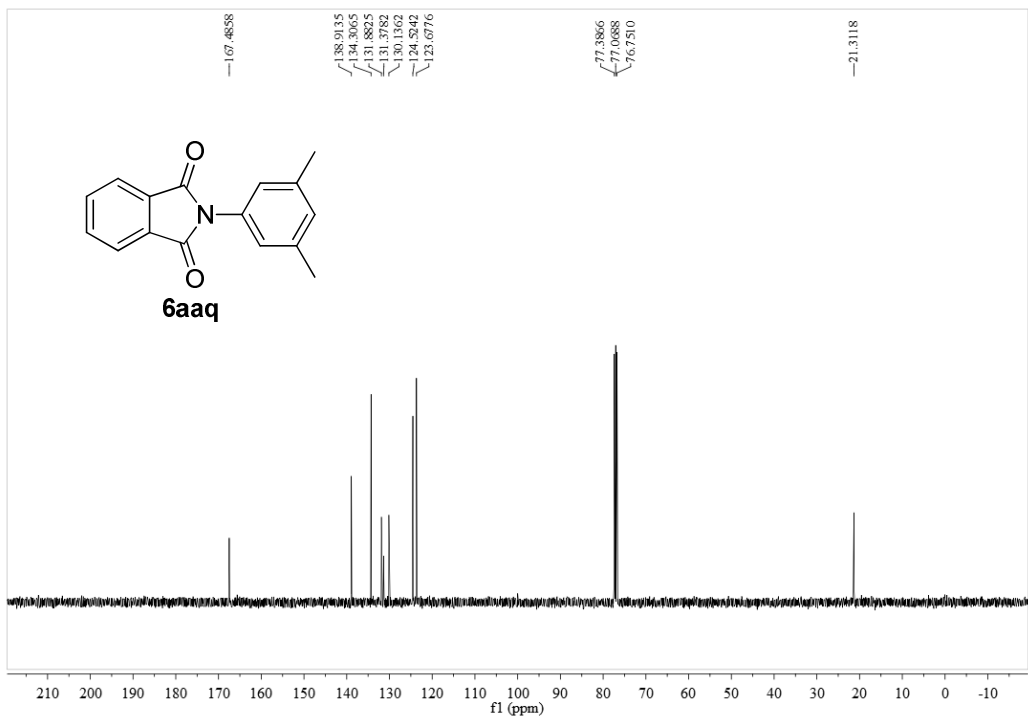
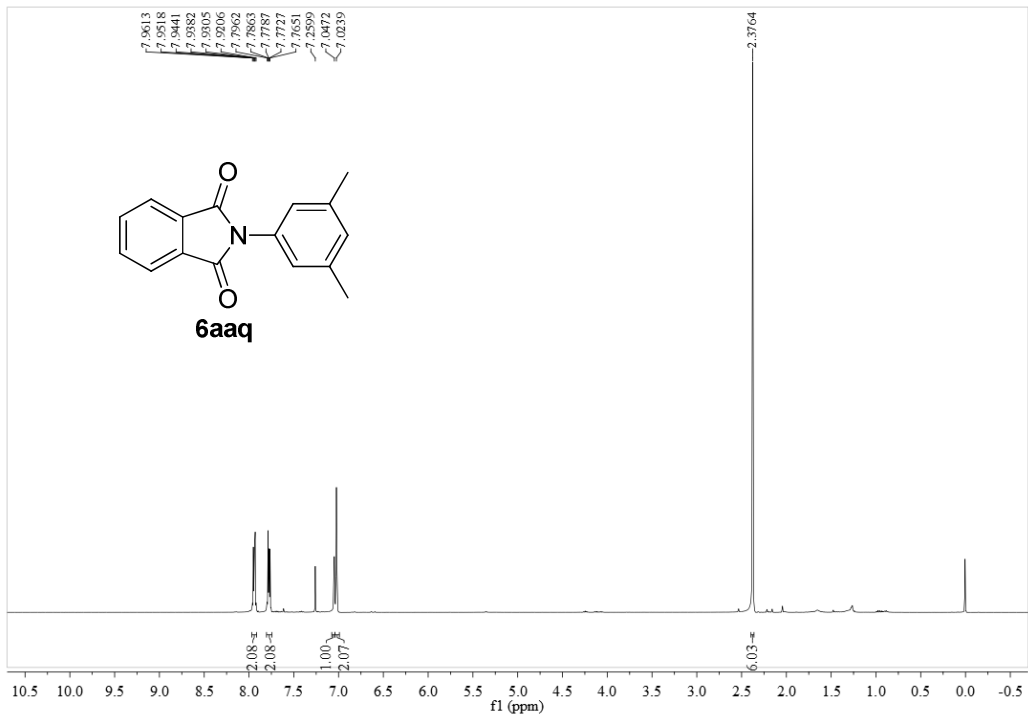


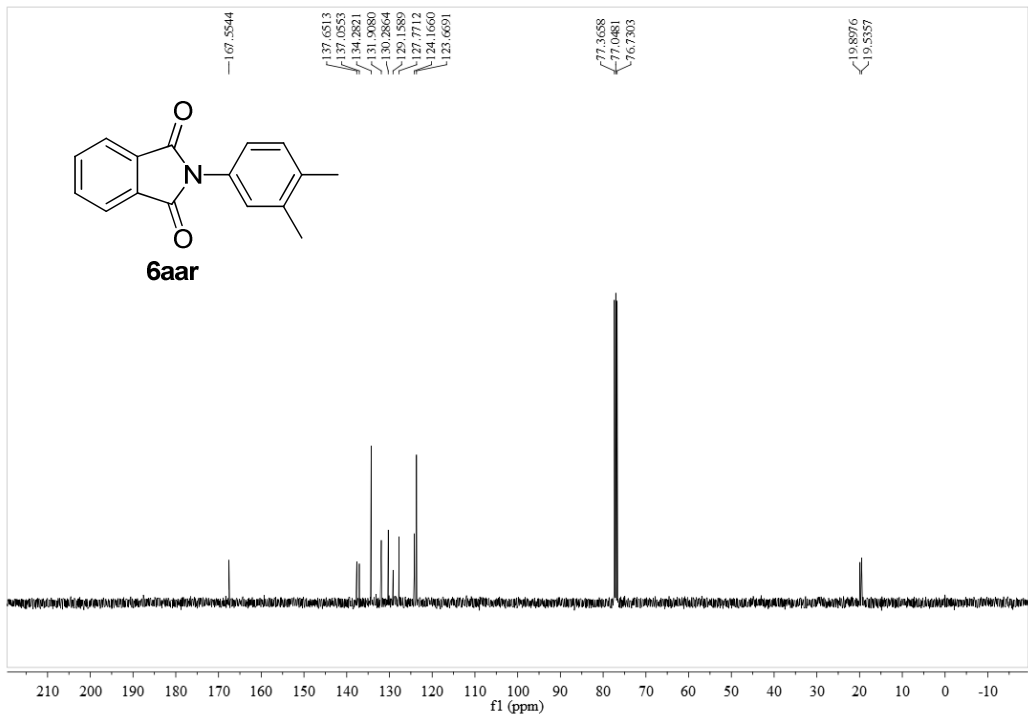
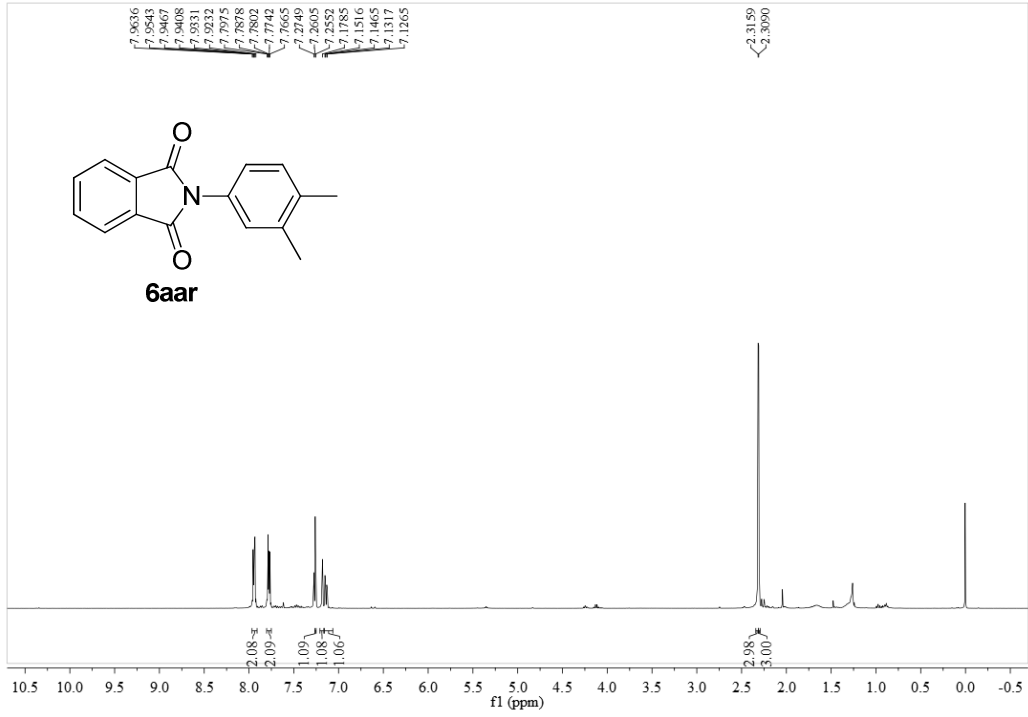


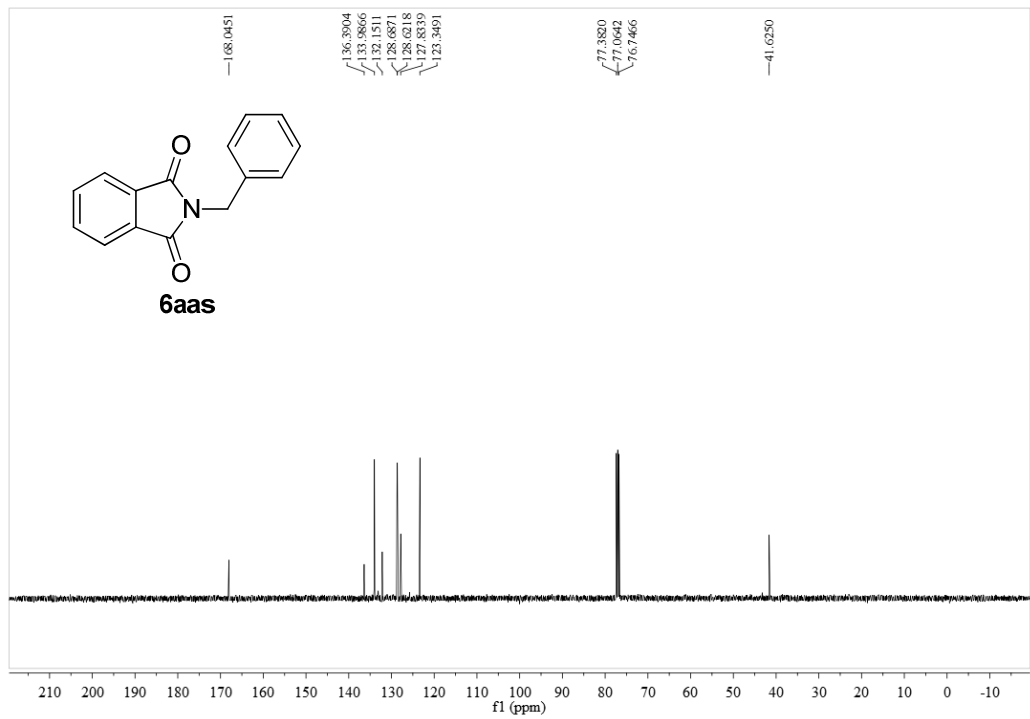
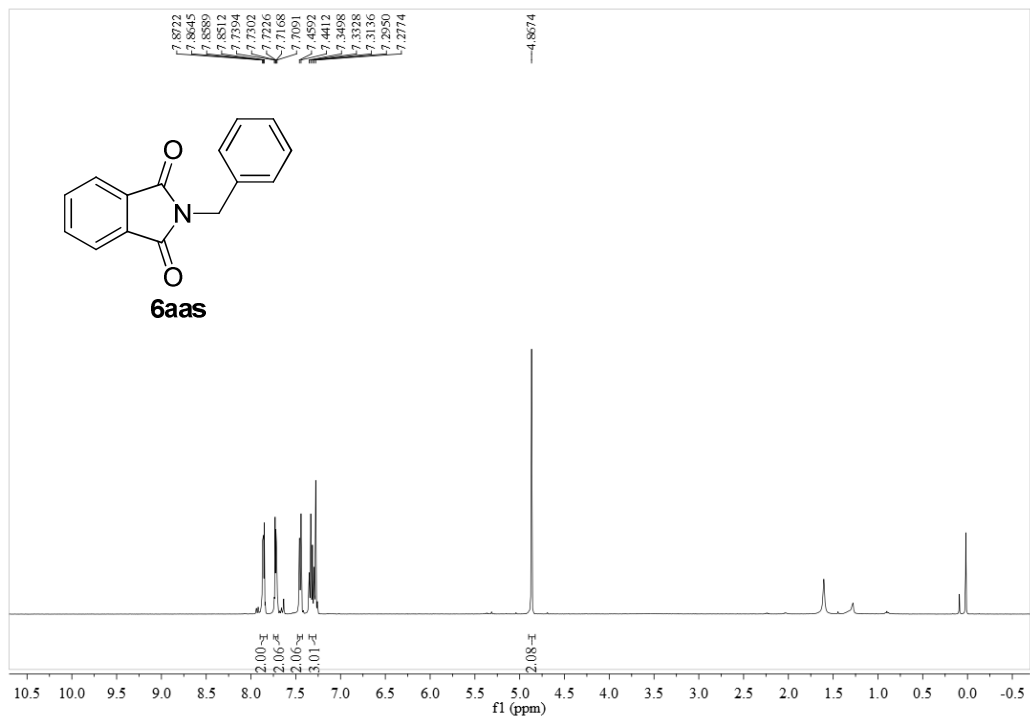


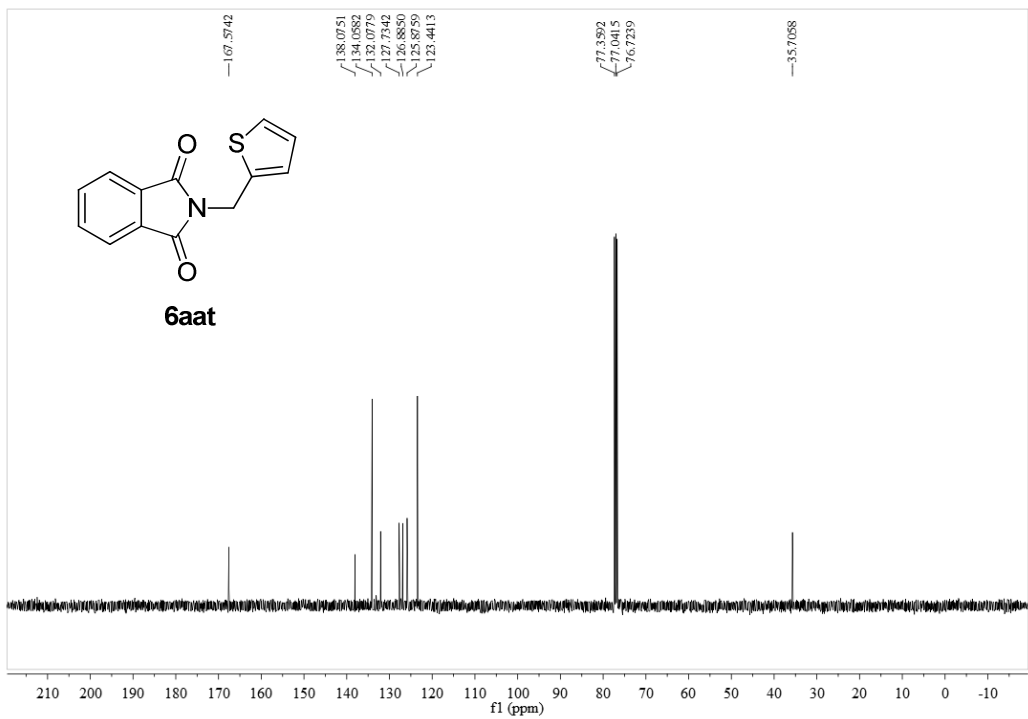
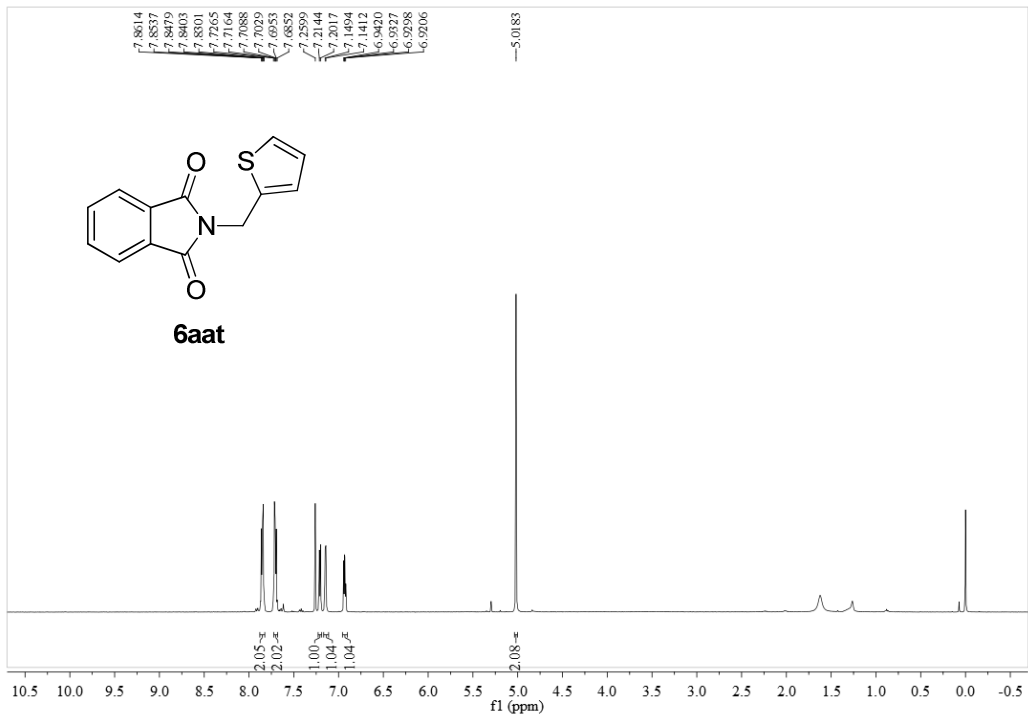


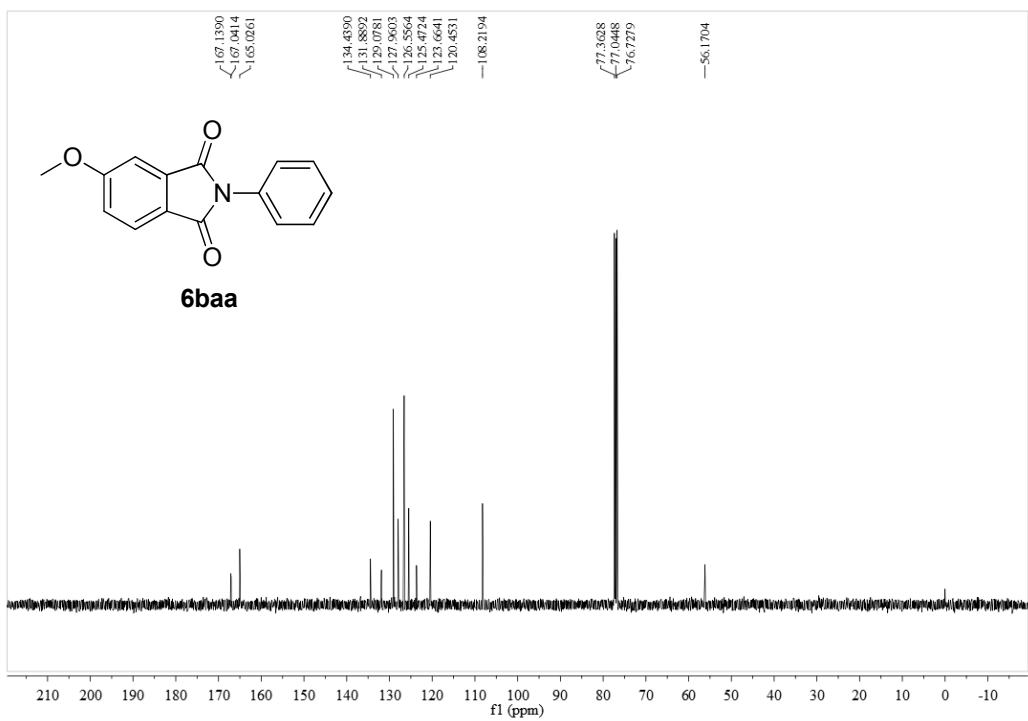
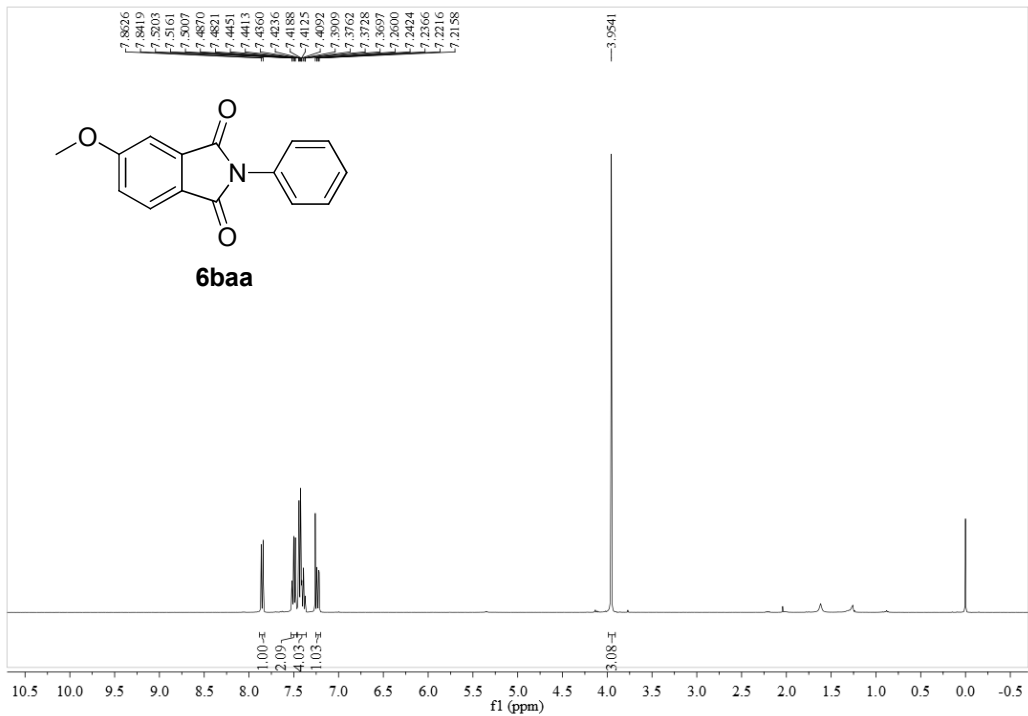


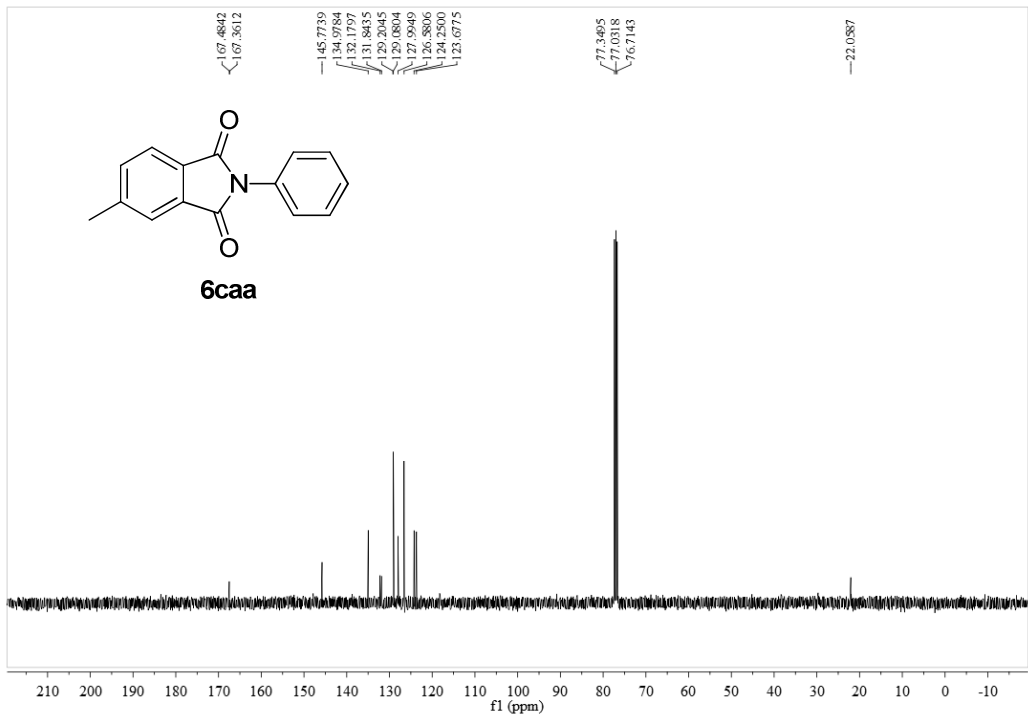
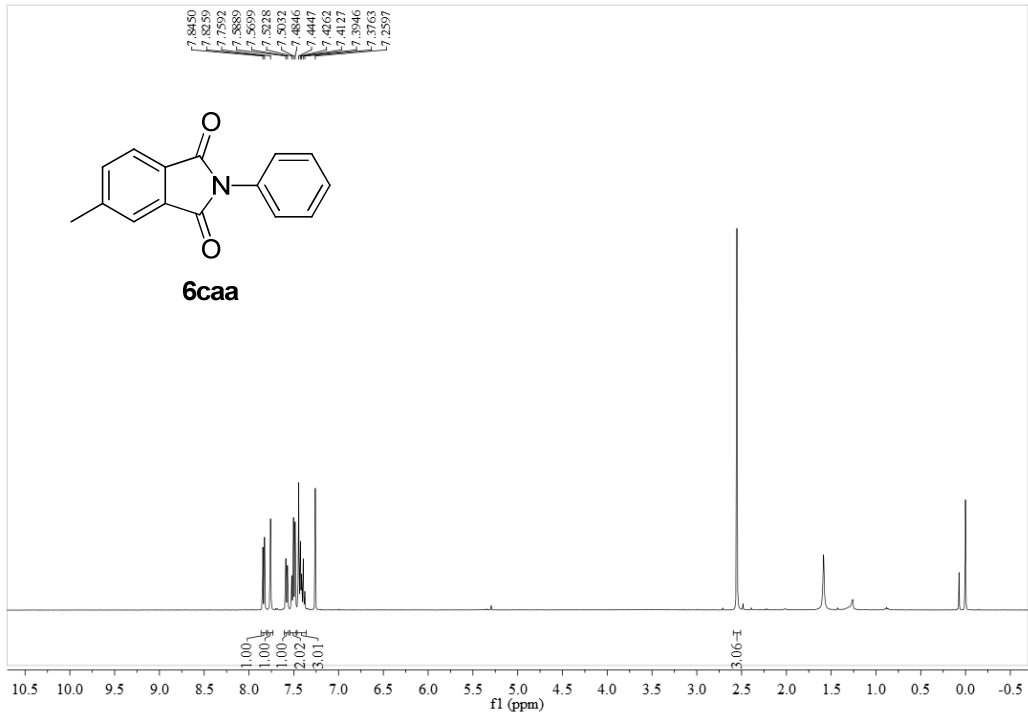


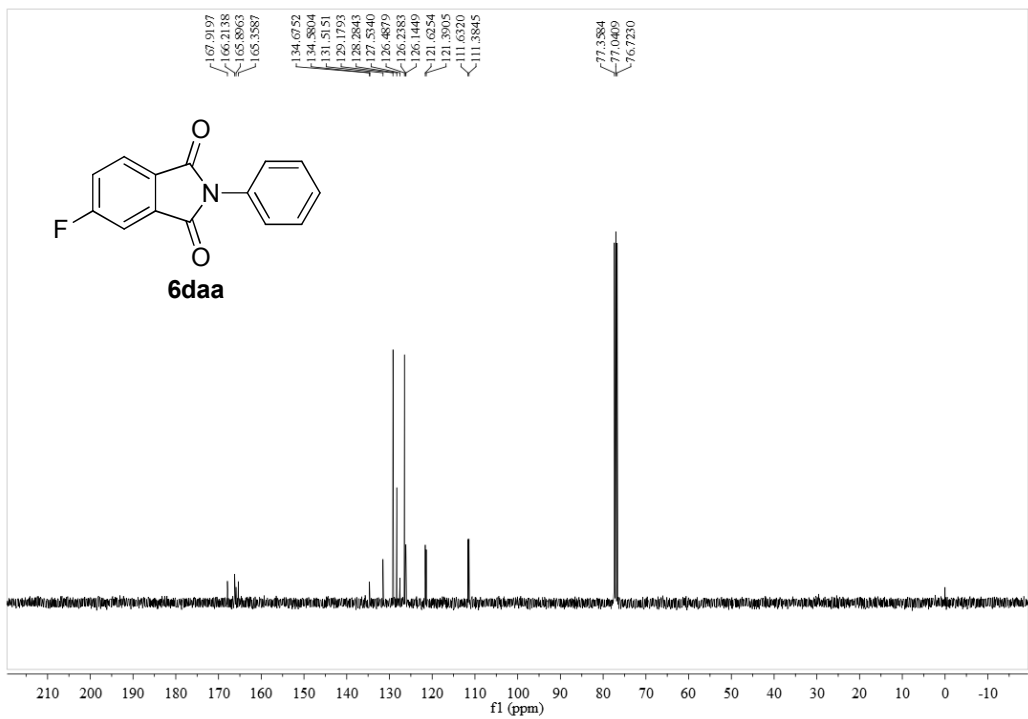
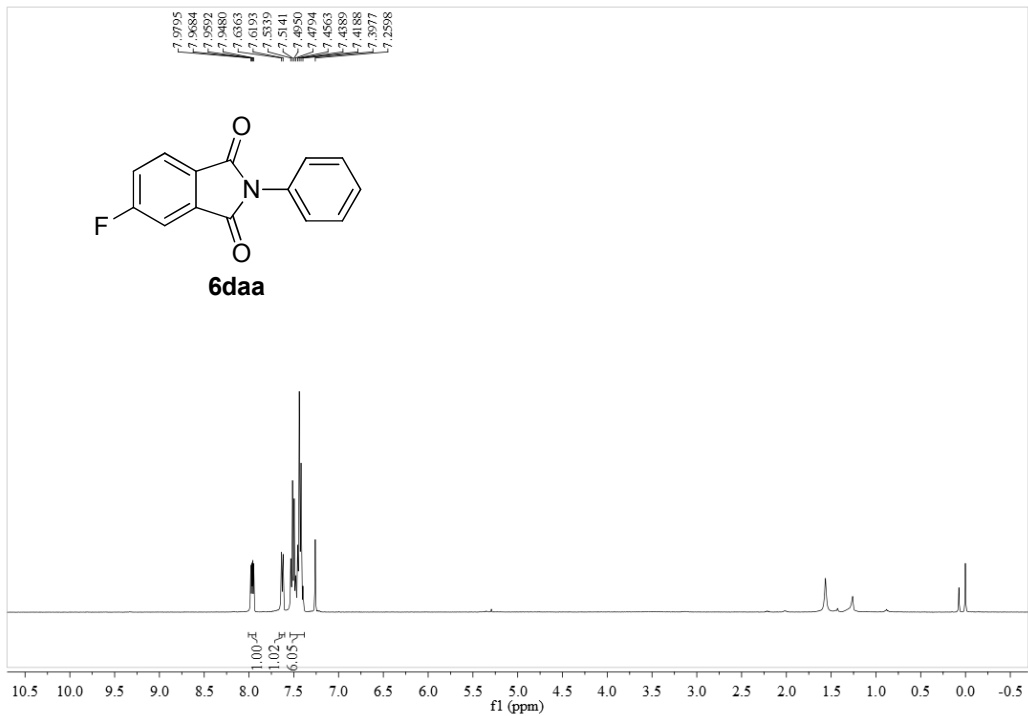


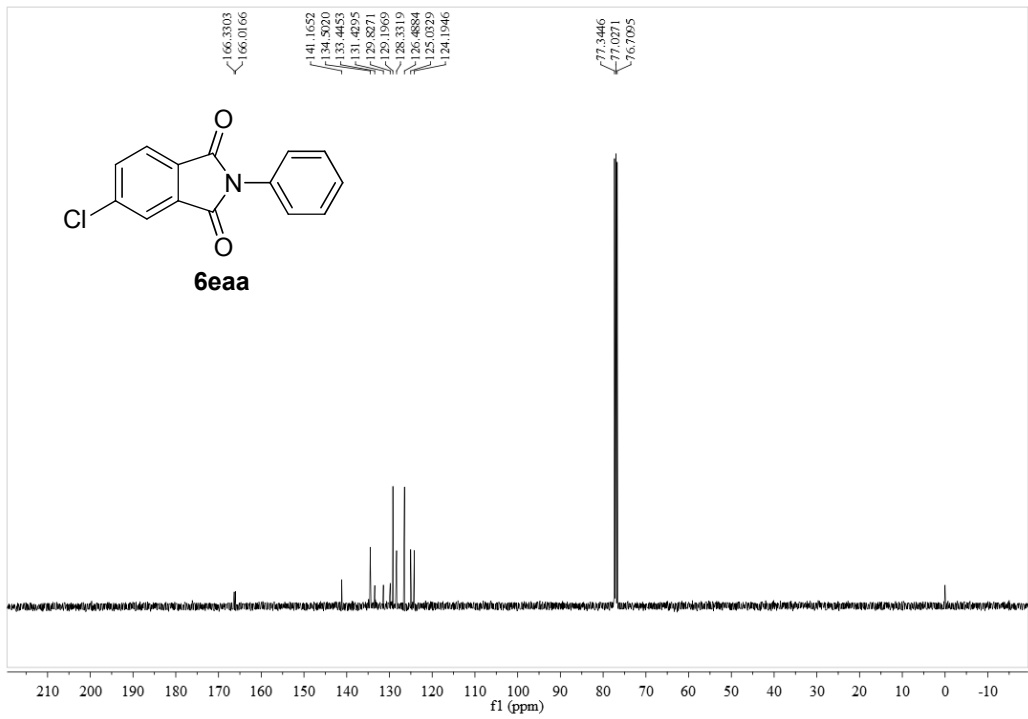
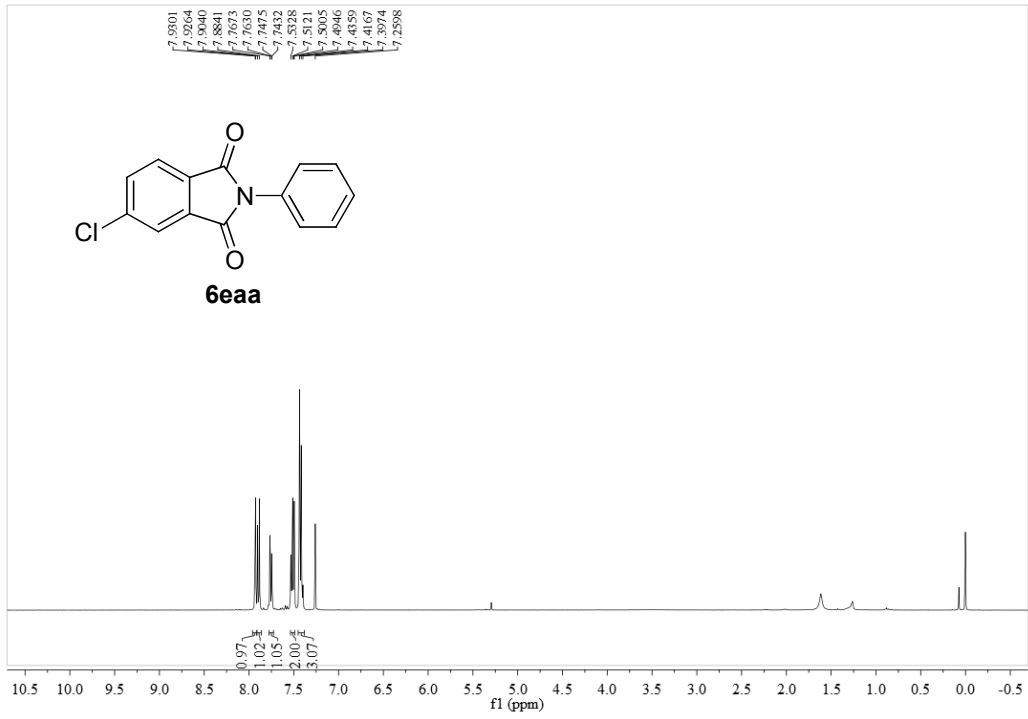


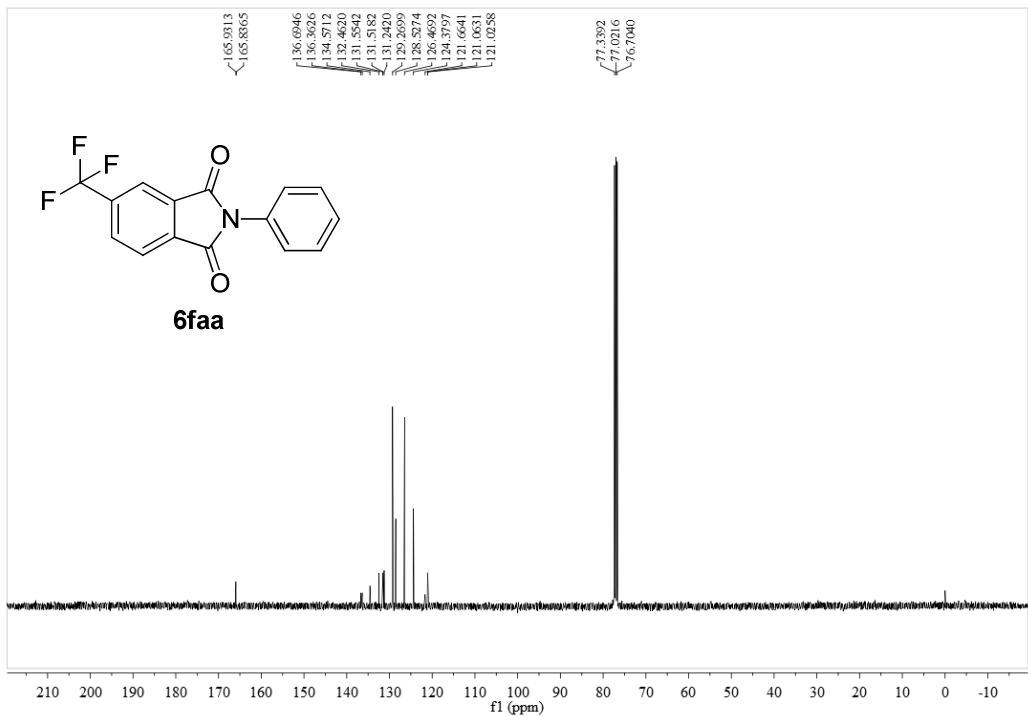
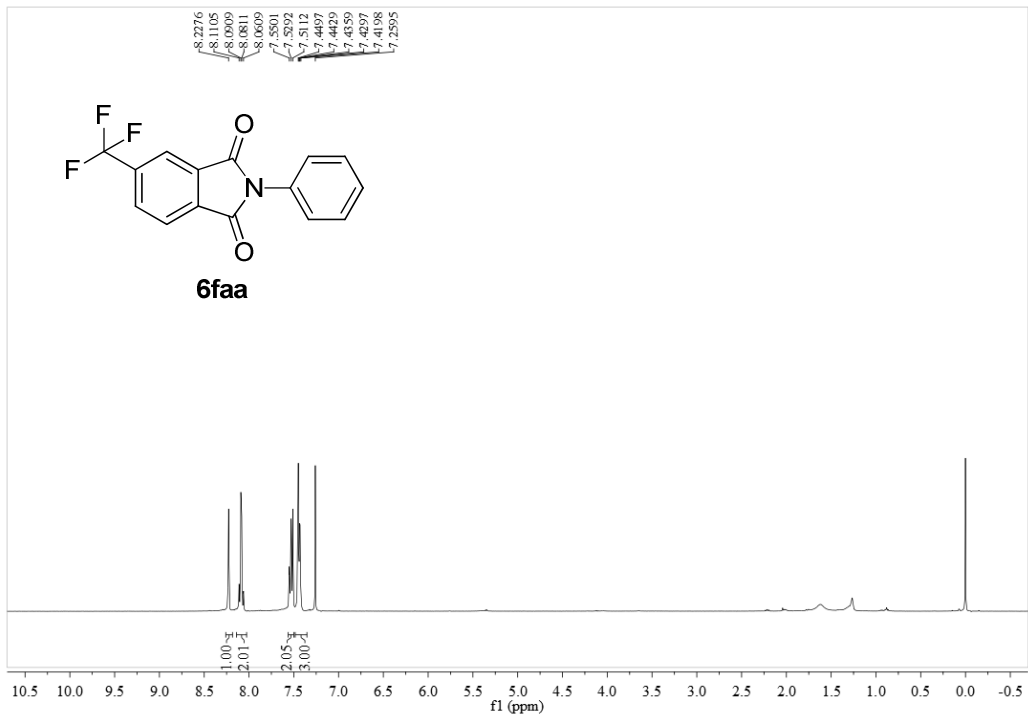


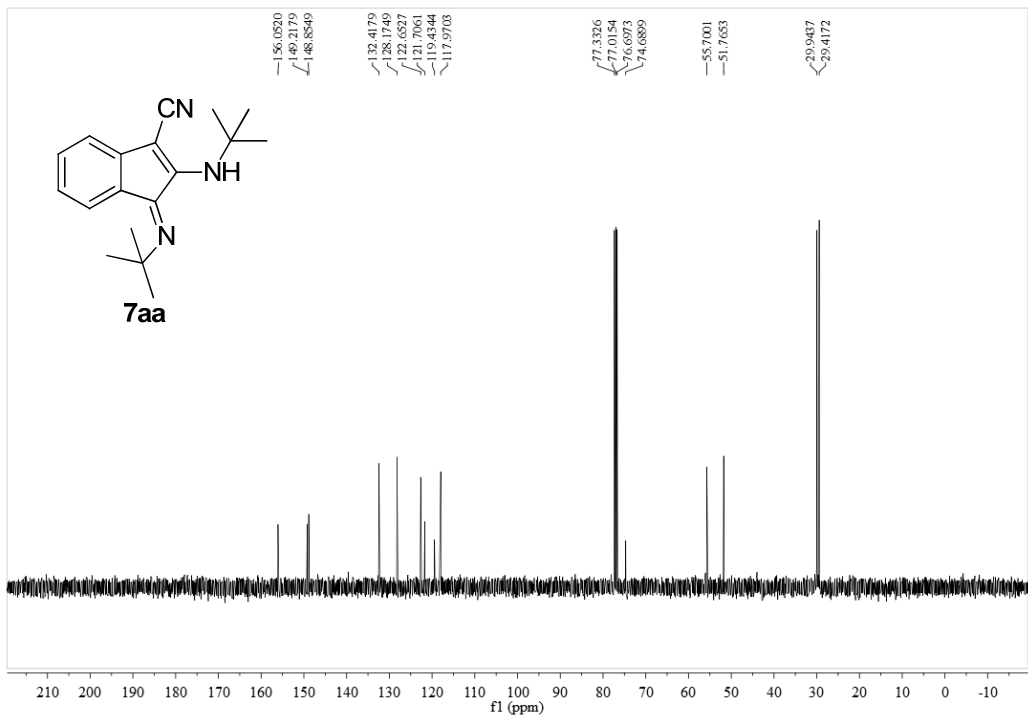
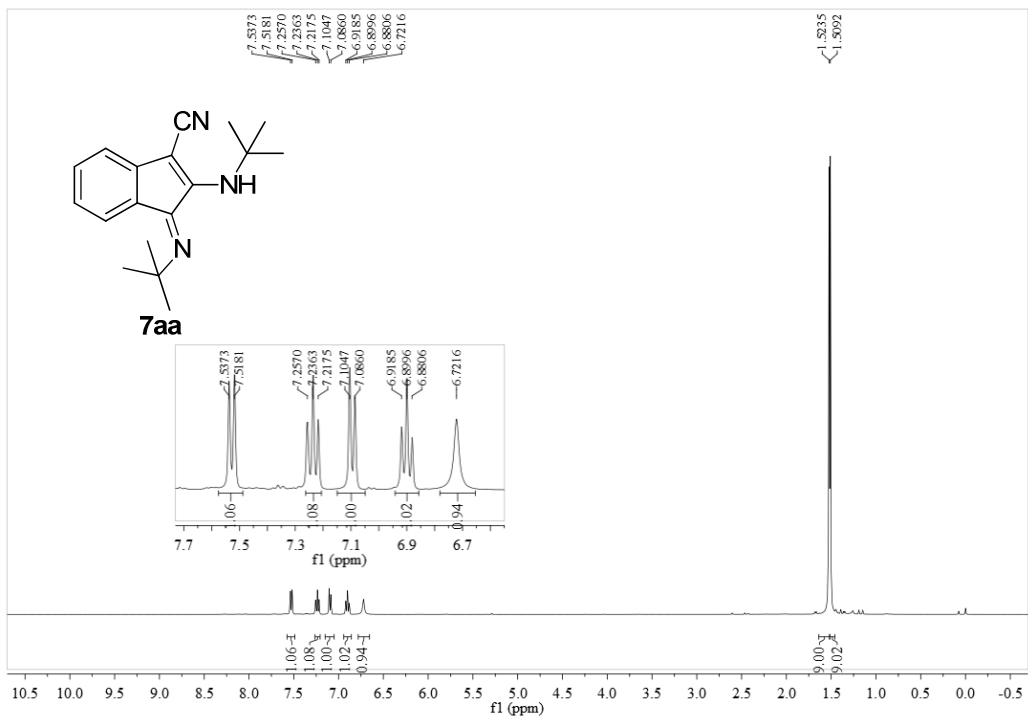


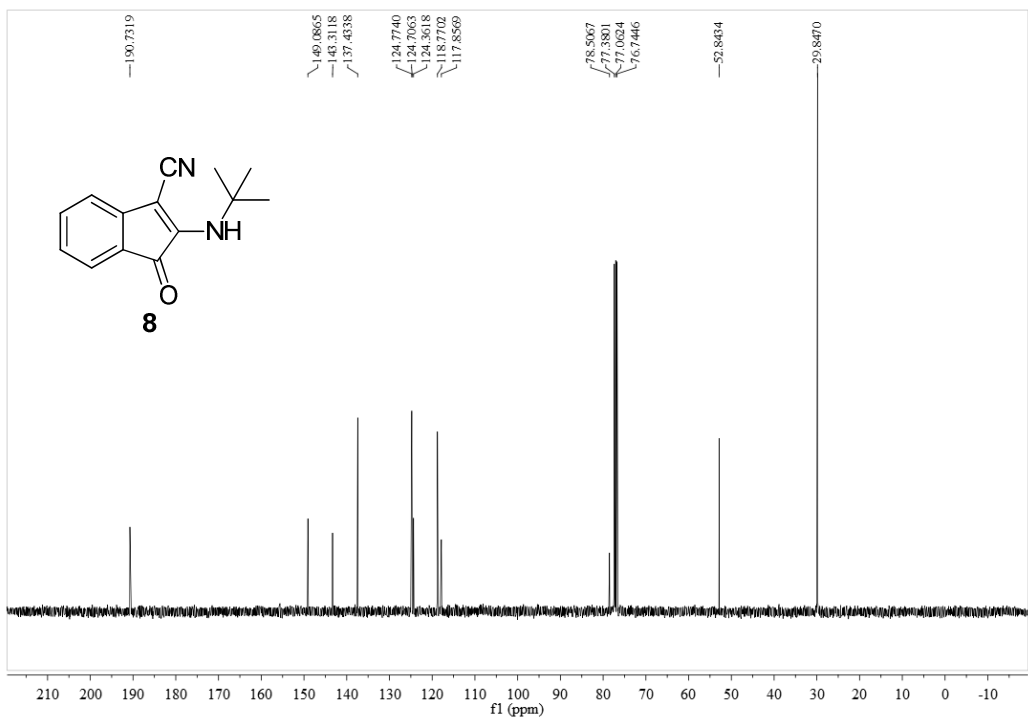
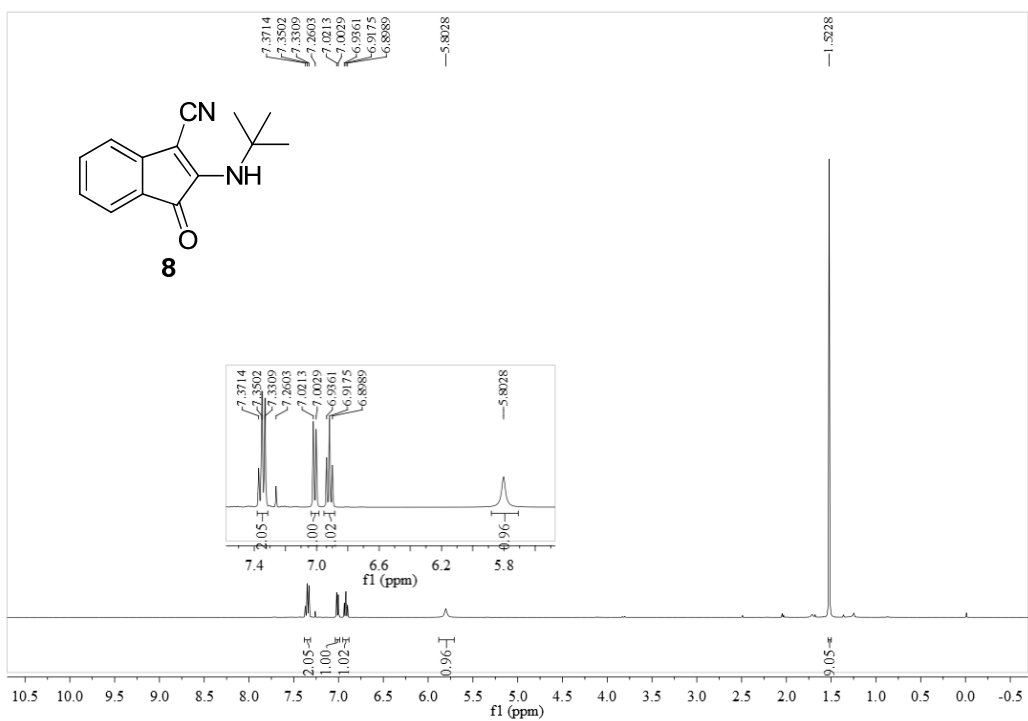


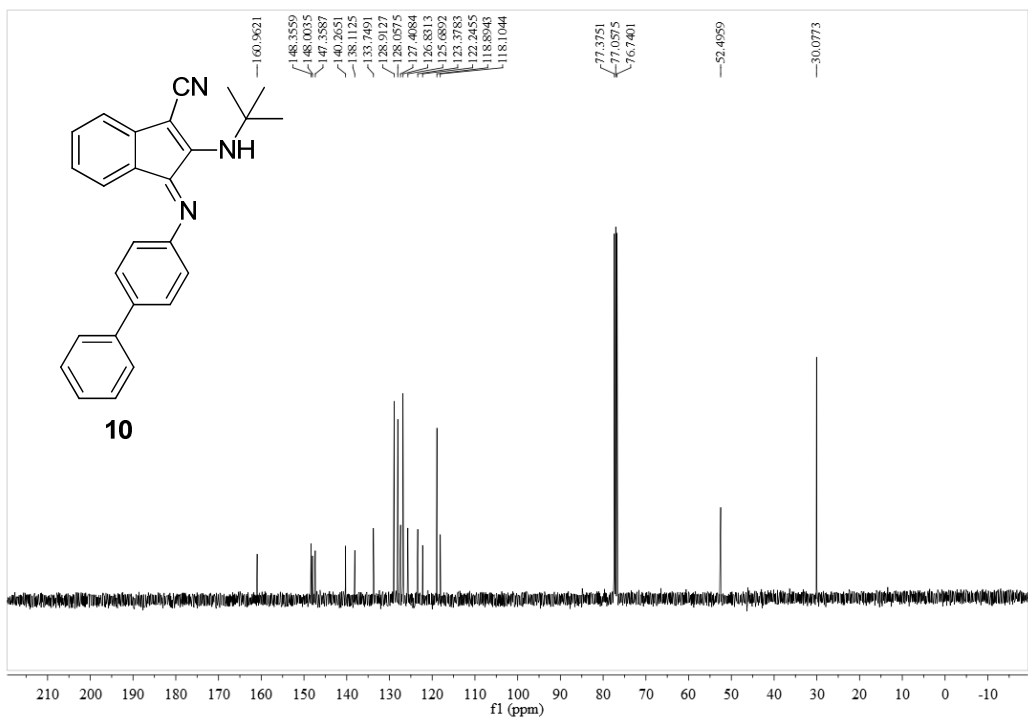
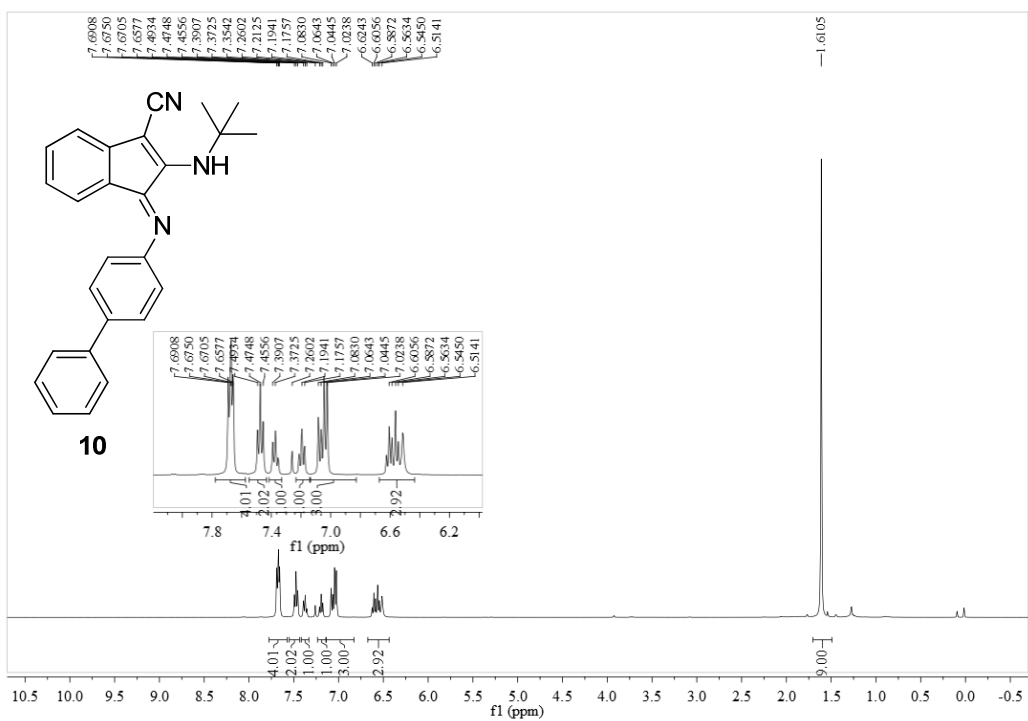


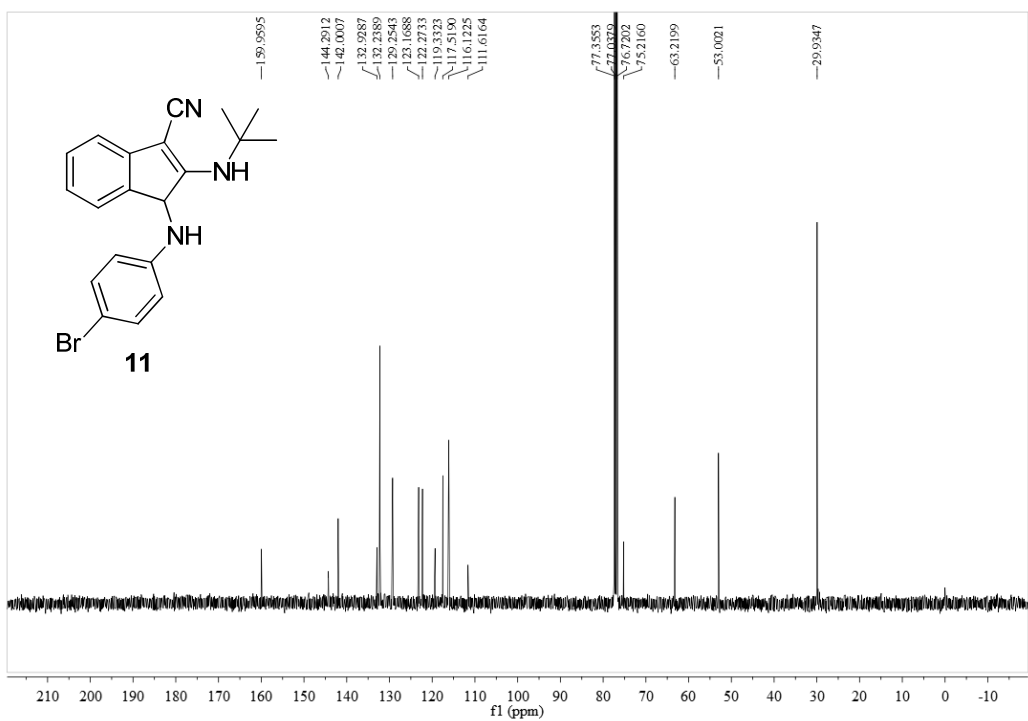
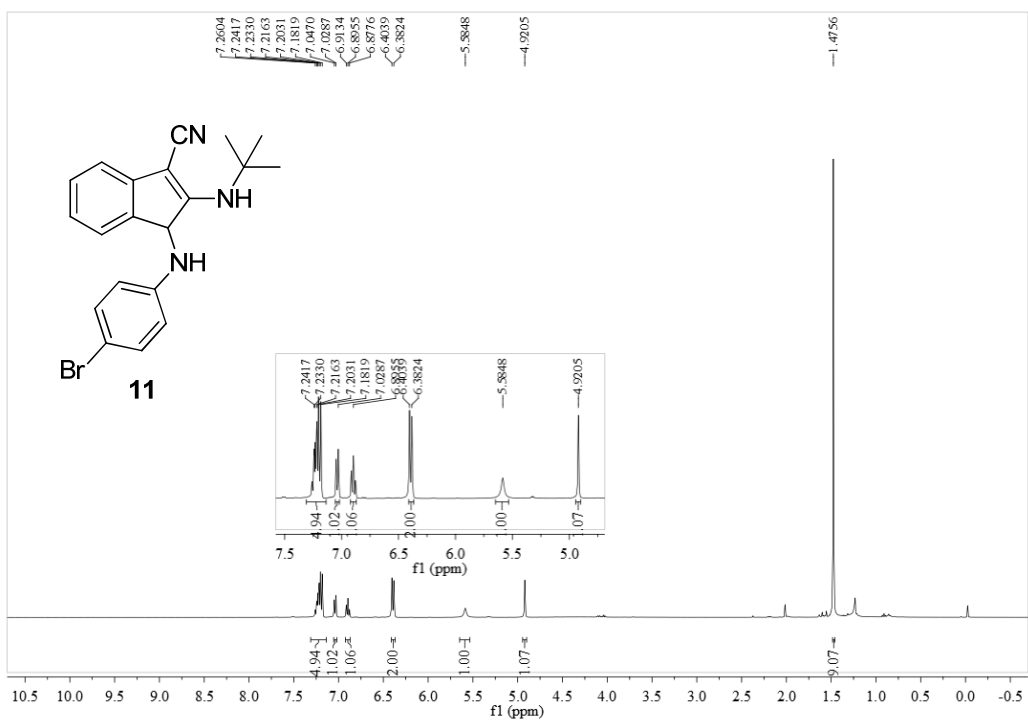




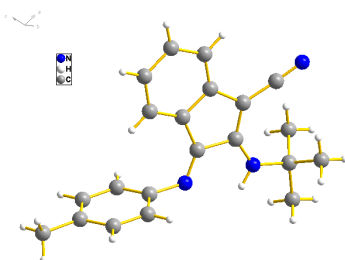








7. X-Ray Structure for Compound 4aac



Formula	C ₂₁ H ₂₁ N ₃
M _r	315.41
Crystal size (mm)	0.3×0.22×0.22
Crystal system	Monoclinic
Space group	P2 ₁
<i>a</i> (Å)	9.6502(2)
<i>b</i> (Å)	10.11860(10)
<i>c</i> (Å)	9.8060(2)
<i>α</i> (°)	90
<i>β</i> (°)	112.655(2)
<i>γ</i> (°)	90
<i>V</i> (Å ³)	883.64(3)
<i>F</i> (000)	336
<i>Z</i>	2
<i>D_c</i> (g cm ⁻³)	1.185
<i>μ</i> (mm ⁻¹)	0.55
<i>θ</i> range (°)	4.9, -73.4
Ref.meas./indep.	3109, 2307
Obs.ref.[<i>I</i> >2σ(<i>I</i>)]	2254
<i>R</i> _{int}	0.0177
<i>R</i> ₁ [<i>I</i> ≥ 2σ(<i>I</i>)] ^a	0.0325
<i>ωR</i> ₂ (all data) ^b	0.0833
Goof	1.08
Δρ(max,min)(eÅ ⁻³)	0.13, -0.19

^a $R_1 = \frac{\sum ||F_o| - |F_c||}{\sum |F_o|}$. ^b $wR_2 = \frac{[\sum w(|F_o|^2 - |F_c|^2)|^2]}{\sum w(|F_o|^2)^2}]^{1/2}$

8. References

- (1) Wang, M.; Lu, J.; Ma, J.; Zhang, Z.; Wang, F. *Angew. Chem. Int. Ed.* **2015**, *54*, 14061-14065.
- (2) Worlikar, S. A.; Larock, R. C. *J. Org. Chem.* **2008**, *73*, 7175-7180.
- (3) Marchetti, L.; Katak, A.; Davis, R.; DeBoef, B. *Org. Lett.* **2015**, *17*, 358-361.