Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

Supporting Information for

Palladium-Catalyzed Oxidative Annulation of *in-situ* Generated Enones to Pyrroles: A Concise Route to Functionalized Indoles

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Experimental procedures and analytical data

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1. General considerations

^{1}H and $^{13}C\{^{1}H\}$ NMR spectra were recorded on a Bruker DRX-400 spectrometer and all chemical shift values refer to δ_{TMS} = 0.00 ppm or $CDCl_3$ ($\delta(^{1}H)$, 7.26 ppm; $\delta(^{13}C)$, 77.16 ppm). The HRMS analysis was obtained on a Waters GC-TOF CA156 mass spectrometer. All the melting points were uncorrected. Analytical TLC plates, Sigma-Aldrich silica gel 60_{F200} were viewed by UV light (254 nm). Column chromatographic purifications were performed on SDZF silica gel 160. All the chemical reagents were purchased from commercial sources and used as received unless otherwise indicated. Compounds 1-methyl-2-phenylpyrrole (**1a**),[1] 1-ethyl-2-phenylpyrrole (**1b**),[2] 1-allyl-2-phenylpyrrole (**1c**),[3] 1-benzyl-2-phenyl- pyrrole (**1d**),[2] 1-methyl-2-(*p*-tolyl)pyrrole (**1e**),[1] 1-methyl-2-(*m*-tolyl)pyrrole (**1f**),[1] 1-methyl-2-(*o*-tolyl)pyrrole (**1g**),[4] 2-(4-methoxyphenyl)-1-methylpyrrole (**1h**),[1] 2-(4-fluorophenyl)-1-methylpyrrole (**1i**),[1] methyl 4-(1-methyl-pyrrol-2yl)benzoate (**1j**),[1] 4-(1-methyl-pyrrol-2-yl)benzonitrile (**1k**)[5] 2-(3-chlorophenyl)-1-methylpy- rrole (**1l**),[1] 1-methyl-2-(naphthalen-1-yl)pyrrole (**1m**),[1] 1-methyl-2-(thiophen-2-yl)- 1*H*-pyrrole (**1n**),[6] 1,2-dimethylpyrrole (**1o**),[7] 1,3-dimethyl-2-phenylpyrrole (**1p)**,[8] and 1-methyl-2-(4-nitrophenyl)pyrrole (**1r**).[4] 3-chloro-1-(*p*-tolyl)propan-1-one (**2b**), 3-chloro-1-(4-methoxyphenyl)propan-1-one (**2c**), 1-(4-(tert-butyl)phenyl)-3-chloropr- opan-1-one (**2d**), 1-([1,1'-biphenyl]4-yl)-3-chloropropan-1-one (**2e**), 3-chloro-1-(3,4 -dimethylphenyl)propan-1-one (**2h**), 3-chloro-1-(2,5-dimethylphenyl)propan-1-one (**2i**), 3-chloro-1-(2,4-dimethylphenyl)propan-1-one (**2j**), 3-chloro-1-(naphthalen-1-yl)-propan-1-one (**2k**), 3-chloro-1-(furan-2-yl)propan-1-one (**2l**), and 3-chloro-1-(thiophen-2-yl)propan-1-one (**2m**)[9] were known compounds and their spectroscopic features were in good agreement with those reported in the literatures.

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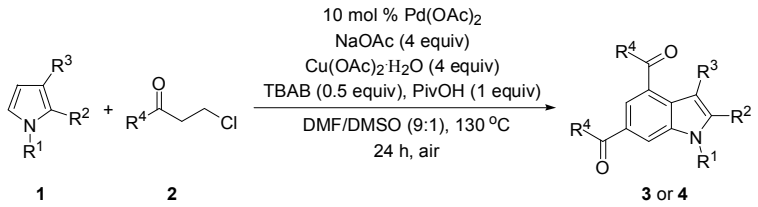
2. Experimental procedures

2.1 A typical procedure for the synthesis of 3 and 4 from the reactions of 1 with 2



Synthesis of 3a: A mixture of *N*-methyl-2-phenylpyrrole (**1a**) (32 mg, 0.2 mmol), 3-chloropropiophenone (**2a**) (133 mg, 0.8 mmol), $Pd(OAc)_2$ (4.5 mg, 0.02 mmol), $Cu(OAc)_2{\cdot}H_2O$ (160 mg, 0.8 mmol), TBAB (32 mg, 0.1 mmol), PivOH (20 mg, 0.2 mmol), and NaOAc (66 mg, 0.8 mmol) in 2.5 mL DMF/DMSO (v/v = 9:1) was stirred at 130 °C under an air atmosphere for 24 h. After cooled to ambient temperature, 10 mL CH_2Cl_2 was added and the resultant mixture was filtered through a short pad of celite, followed by rinsing with 10 mL CH_2Cl_2. The combined filtrate was washed with brine (10 mL) and separated. The organic phase was dried over anhydrous Na_2SO_4, filtered, concentrated under reduced pressure. The resulting residue was

purified by silica gel column chromatography (eluent: petroleum ether (60-90 ^{o}C)/EtOAc/CH_2Cl_2 (30:1:3, v/v/v)) to afford **3a** as a yellow liquid (56 mg, 68%).

2.2 Screening of reaction conditions

Table S1: Screening of conditions for the reaction of *N*-methyl-2-phenylpyrrole (**1a**) with 3-chloropropiophenone (**2a**)[a]

1a **2a** **3a**

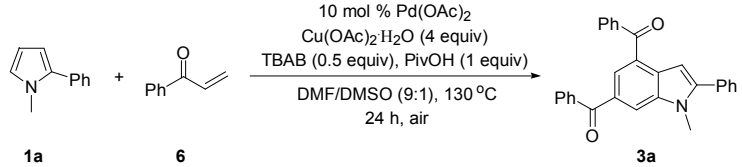
Entry	Catalyst	Base	Oxidant	Solvent	Additive	Yield[b] [%]
1	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	toluene		0
2	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	dioxane		7
3	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMSO		41
4	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF		42
5	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	toluene/DMSO (v/v = 10:1)		36
6	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO/HOAc (v/v/v = 20:2:1)		51
7	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	Dioxane/DMSO (v/v = 10:1)		48
8	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 2:1)		43
9	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)		64
10	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 20:1)		52
11	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2$	DMF/DMSO (v/v = 9:1)		61
12	$Pd(OAc)_2$	NaOAc	$CuCl_2$	DMF/DMSO (v/v = 9:1)		0
13	$Pd(OAc)_2$	NaOAc	CuOAc	DMF/DMSO (v/v = 9:1)		44
14	$Pd(OAc)_2$	NaOAc	AgOAc	DMF/DMSO (v/v = 9:1)		25
15	$Pd(OAc)_2$	NaOAc	Ag_2CO_3	DMF/DMSO (v/v = 9:1)		36
16	$Pd(OAc)_2$	NaOAc	Ag_2O	DMF/DMSO (v/v = 9:1)		12
17	$Pd(OAc)_2$	NaOAc	BQ	DMF/DMSO (v/v = 9:1)		0

18	$Pd(OAc)_2$	NaOAc	*t*BuOO*t*Bu	DMF/DMSO (v/v = 9:1)		17
19	$Pd(OAc)_2$	NaOAc	$K_2S_2O_8$	DMF/DMSO (v/v = 9:1)		trace
20	$Pd(OAc)_2$	NaOAc	DDQ	DMF/DMSO (v/v = 9:1)		0
21	$Pd(OAc)_2$	NaOAc	air	DMF/DMSO (v/v = 9:1)		13
22	$Pd(OAc)_2$	NaOAc	O_2	DMF/DMSO (v/v = 9:1)		14
23[c]	$Pd(OAc)_2$	NaOAc		DMF/DMSO (v/v = 9:1)		<1
24	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv)	65
25	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	PivOH (1.0 equiv)	66
26	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	70
27	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.2 equiv) + PivOH (0.5 equiv)	60
28	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	CuOAc (1.0 equiv)	59
29	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	4ÅMS	59
30	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	$FeCl_3$ (0.5 equiv)	51
31[c]	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv)+ PivOH (1.0 equiv)	55
32	$Pd(OAc)_2$	CsOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	48
33	$Pd(OAc)_2$	LiOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	59
34	$Pd(OAc)_2$	Na_2CO_3	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	39
35	$Pd(OAc)_2$	K_3PO_4	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	42
36		NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO	TBAB (0.5	trace

				(v/v = 9:1)	equiv) + PivOH (1.0 equiv)	
37	$PdCl_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	47
38	$Pd(PPh_3)_2Cl_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	57
39	$Pd(PPh_3)_4$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	30
40	$Pd_2(dba)_3$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	39
41[d]	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	50
42[e]	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	60
43[e,f]	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	48
44[e,g]	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	60
45[e,h]	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	68
46[e,i]	$Pd(OAc)_2$	NaOAc	$Cu(OAc)_2·H_2O$	DMF/DMSO (v/v = 9:1)	TBAB (0.5 equiv) + PivOH (1.0 equiv)	52

[a] Conditions: **1a** (0.2 mmol), **2a** (0.8 mmol), catalyst (0.02 mmol), base (0.8 mmol), oxidant (1.2 mmol), solvent (2.5 mL), air, 100 °C, 24 h. [b] Isolated yields based on **1a**. [c] Under 0.1 MPa N_2 atmosphere. [d] $Cu(OAc)_2·H_2O$ (0.6 mmol). [e] $Cu(OAc)_2·H_2O$ (0.8 mmol). [f] 80 °C. [g] 110 °C. [h] 130 °C. [i] 140 °C.

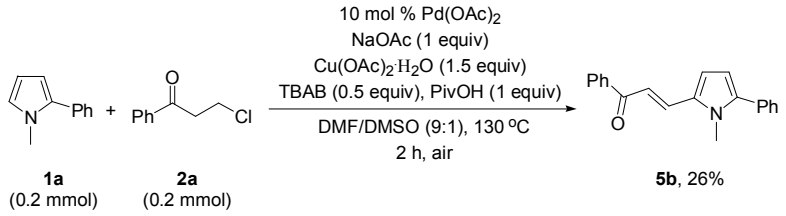
2.3 Reaction of pyrrole 1a with enone 6



1a **6** **3a**

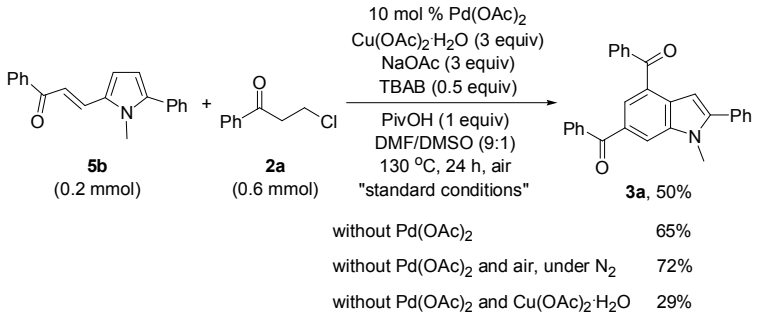
A mixture of *N*-methyl-2-phenylpyrrole (**1a**) (32 mg, 0.2 mmol), enone **6** (106 mg, 0.8 mmol), $Pd(OAc)_2$ (4.5 mg, 0.02 mmol), $Cu(OAc)_2{\cdot}H_2O$ (160 mg, 0.8 mmol), TBAB (32 mg, 0.1 mmol), and PivOH (20 mg, 0.2 mmol) in 2.5 mL DMF/DMSO (v/v = 9:1) was stirred at 130 °C under an air atmosphere for 24 h. After cooled to ambient temperature, 10 mL CH_2Cl_2 was added and the resultant mixture was filtered through a short pad of celite, followed by rinsing with 10 mL CH_2Cl_2. The combined filtrate was washed with brine (10 mL) and separated. The organic phase was dried over anhydrous Na_2SO_4, filtered, concentrated under reduced pressure. The resulting residue was purified by silica gel column chromatography (eluent: petroleum ether (60-90 °C)/EtOAc/CH_2Cl_2 (30:1:3, v/v/v)) to afford **3a** as a yellow liquid (49 mg, 59%).

2.4 Preparation of 5-alkenylated pyrrole intermediate 5b



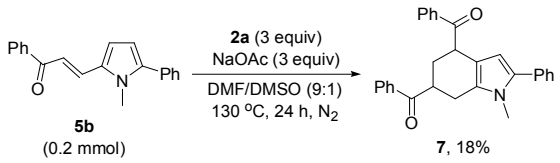
A mixture of *N*-methyl-2-phenylpyrrole (**1a**) (32 mg, 0.2 mmol), 3-chloropropiophenone (**2a**) (34 mg, 0.2 mmol), $Pd(OAc)_2$ (4.5 mg, 0.02 mmol), $Cu(OAc)_2{\cdot}H_2O$ (60 mg, 0.3 mmol), TBAB (32 mg, 0.1 mmol), PivOH (20 mg, 0.2 mmol), and NaOAc (17 mg, 0.2 mmol) in 2.5 mL DMF/DMSO (v/v = 9:1) was stirred at 130 °C under an air atmosphere for 2 h. After cooled to ambient temperature, 10 mL CH_2Cl_2 was added and the resultant mixture was filtered through a short pad of celite, followed by rinsing with 10 mL CH_2Cl_2. The combined filtrate was washed with brine (10 mL) and separated. The organic phase was dried over anhydrous Na_2SO_4, filtered, concentrated under reduced pressure. The resulting residue was purified by silica gel column chromatography (eluent: petroleum ether (60-90 °C)/EtOAc/CH_2Cl_2 (15:1:1, v/v/v)) to afford **5b** as a yellow solid (15 mg, 26%).

2.5 Reaction of 5b with 2a



A typical procedure for the reaction of 5b with 2a under the standard conditions: A mixture of **5b** (54 mg, 0.2 mmol), **2a** (101 mg, 0.6 mmol), $Pd(OAc)_2$ (4.5 mg, 0.02 mmol), $Cu(OAc)_2 \cdot H_2O$ (120 mg, 0.6 mmol), TBAB (32 mg, 0.1 mmol), PivOH (20 mg, 0.2 mmol), and NaOAc (49 mg, 0.6 mmol) in 2.5 mL DMF/DMSO (v/v = 9:1) was stirred at 130 °C under an air atmosphere for 24 h. After cooled to ambient temperature, 10 mL CH_2Cl_2 was added and the resultant mixture was filtered through a short pad of celite, followed by rinsing with 10 mL CH_2Cl_2. The combined filtrate was washed with brine (10 mL) and separated. The organic phase was dried over anhydrous Na_2SO_4, filtered, concentrated under reduced pressure. The resulting residue was purified by silica gel column chromatography (eluent: petroleum ether (60-90 °C)/EtOAc/CH_2Cl_2 (30:1:3, v/v/v)) to afford **3a** as a yellow liquid (42 mg, 50%).

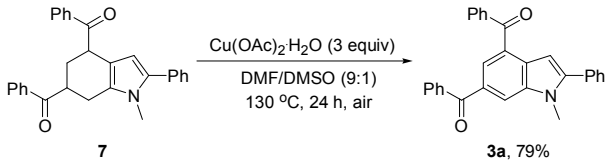
2.6 Preparation of intermediate 7



A mixture of **5b** (54 mg, 0.2 mmol), **2a** (101 mg, 0.6 mmol), and NaOAc (49 mg, 0.6 mmol) in 2.5 mL DMF/DMSO (v/v = 9:1) was stirred at 130 °C under a nitrogen atmosphere for 24 h. After cooled to ambient temperature, 10 mL CH_2Cl_2 was added and the resultant mixture was filtered through a short pad of celite, followed by rinsing with 10 mL CH_2Cl_2. The combined filtrate was washed with brine (10 mL) and separated. The organic phase was dried over anhydrous Na_2SO_4, filtered,

concentrated under reduced pressure. The resulting residue was purified by silica gel column chromatography (eluent: petroleum ether (60-90 °C)/EtOAc/CH_2Cl_2 (30:1:1, v/v/v)) to afford **7** as a white solid (15 mg, 18%).

2.7 Dehydrogenative aromatization of 7



7 **3a**, 79%

A mixture of **7** (42 mg, 0.1 mmol) and $Cu(OAc)_2 \cdot H_2O$ (60 mg, 0.3 mmol) in 1.5 mL DMF/DMSO (v/v = 9:1) was stirred at 130 °C under an air atmosphere for 24 h. After cooled to ambient temperature, 10 mL CH_2Cl_2 was added and the resultant mixture was filtered through a short pad of celite, followed by rinsing with 10 mL CH_2Cl_2. The combined filtrate was washed with brine (10 mL) and separated. The organic phase was dried over anhydrous Na_2SO_4, filtered, concentrated under reduced pressure. The resulting residue was purified by silica gel column chromatography (eluent: petroleum ether (60-90 °C)/EtOAc/CH_2Cl_2 (10:1:1, v/v/v)) to afford **3a** as a yellow liquid (32 mg, 79%).

3. X-Ray crystallographic studies

X-Ray diffraction studies for compound **4d** was carried out on a SMART APEX diffractometer with graphite-monochromated Mo Kα radiation (λ= 0.71073 Å). Cell parameters were obtained by global refinement of the positions of all collected reflections. Intensities were corrected for Lorentz and polarization effects and empirical absorption. The structures were solved by direct methods and refined by full-matrix least squares on F^2. All non-hydrogen atoms were refined anisotropically. All hydrogen atoms were placed in calculated positions. Structure solution and refinement were performed by using the SHELXL-97 package. The X-ray crystallographic files, in CIF format, are available from the Cambridge Crystallographic Data Centre on quoting the deposition numbers CCDC 1002185 for **4d**. Copies of this information may be obtained free of charge from The Director,

CCDC, 12 Union Road, Cambridge CB2 IEZ, UK (Fax: +44-1223-336033; e-mail: deposit@ccdc.cam.ac.uk or www: http://www. ccdc.cam.ac.uk).

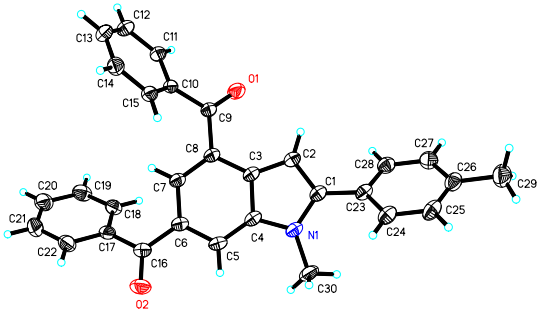
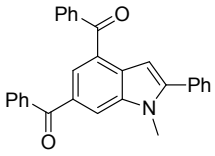
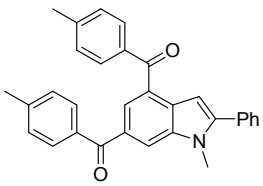


Figure 1. Molecular structure of **4d**.

4. Analytical data

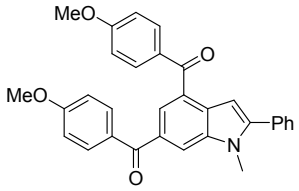


(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (3a): Yield 68%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.17 and 7.95 (s each, 1:1 H, aromatic CH), 7.86 (m, 4 H, aromatic CH), 7.56 (m, 4 H, aromatic CH), 7.48 (m, 7 H, aromatic CH), 7.08 (s, 1 H, 3-H of indolyl), 3.88 (s, 3 H, NCH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.8 and 196.4 (Cq, C=O), 147.7, 138.9, 138.7, 138.5, 131.7, 130.8 and 129.9 (Cq), 132.5, 132.2, 130.2, 130.1, 129.6, 129.0, 128.9, 128.4, 126.8, 116.2, and 103.6 (CH), 31.8 (CH_3). HRMS Calcd for $C_{29}H_{21}NO_2$ $[M+H]^+$: 416.1651; Found: 416.1659.



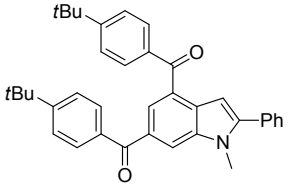
(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis(p-tolylmethanone) (3b): Yield 73%. Yellow solid. M.p.: 224-227 ^{o}C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.13 and 7.92 (s each, 1:1 H, aromatic CH), 7.78 (m, 4 H, aromatic CH), 7.55 (m, 2 H, aromatic CH), 7.50 (m, 2 H, aromatic CH), 7.45 (m, 1 H, aromatic CH), 7.29 (m, 3 H, aromatic CH),

7.26 (m, 1 H, aromatic CH), 7.02 (m, 1 H, 3-H of indolyl), 3.87 (s, 3 H, NCH_3), 2.44 and 2.43 (s each, 3:3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.6 and 196.3 (Cq, C=O), 147.3, 143.3, 142.93, 138.8, 135.9, 135.8, 131.8, 130.6 and 128.9 (Cq), 130.4, 130.4, 129.6, 129.1, 129.1, 128.9, 128.8, 126.2, 115.9 and 103.4 (CH), 31.7 (NCH_3), 21.79 and 21.76 (CH_3). HRMS Calcd for $C_{31}H_{25}NO_2$ $[M+H]^+$: 444.1964; Found: 444.1970.



(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis((4-methoxyphenyl)methanone)

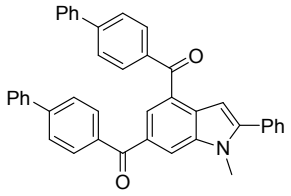
(3c): Yield 69%. Yellow solid. M.p.: 140-143 ^oC. ^1H NMR (400 MHz, $CDCl_3$) δ 8.09 (s, 1 H, aromatic CH), 7.87 (m, 5 H, aromatic CH), 7.54 (m, 2 H, aromatic CH), 7.52-7.41 (m, 3 H, aromatic CH), 6.96 (m, 5 H, aromatic CH and 3-H of indolyl), 3.87 (m, 9 H, NCH_3 and 2×OCH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 195.6 and 195.4 (Cq, C=O), 163.3, 163.1, 146.9, 138.8, 131.8, 131.2, 131.0, 130.7, 130.4 and 129.1 (Cq), 132.6, 132.6, 129.5, 128.9, 128.8, 125.5, 115.4, 113.7, 113.7 and 103.2 (CH), 55.6 (2×OCH_3), 31.7 (NCH_3). HRMS Calcd for $C_{31}H_{25}NO_4$ $[M+H]^+$: 476.1862; Found: 476.1862.



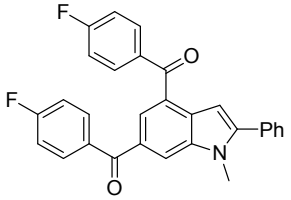
(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis((4-(tert-butyl)phenyl)methanone)

(3d): Yield 67%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.18 and 7.96 (s each, 1:1 H, aromatic CH), 7.83 (m, 4 H, aromatic CH), 7.57 (d, J = 7.1 Hz, 2 H, aromatic CH), 7.54-7.45 (m, 7 H, aromatic CH), 7.11 (s, 1 H, 3-H of indolyl), 3.89 (s, 3 H, NCH_3), 1.37 and 1.36 (s each, 9:9 H, 2×*t*Bu). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.5 and 196.1 (Cq, C=O), 156.1, 155.9, 147.5, 138.9, 135.9, 135.7, 131.8, 130.7 and 128.6 (Cq), 130.3, 130.2, 129.6, 128.9, 128.8, 126.7, 125.3, 115.9 and 103.5 (CH),

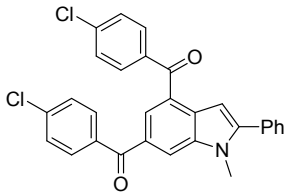
35.2 (NCH_3), 31.8 ($C(CH_3)_3$), 31.30 and 31.29 ($C(CH_3)_3$). HRMS Calcd for $C_{37}H_{37}NO_2$ $[M+H]^+$: 528.2903; Found: 528.2912.



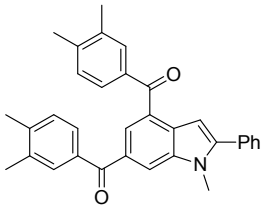
(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis([1,1'-biphenyl]-4-ylmethanone) (3e): Yield 71%. Yellow solid. M.p.: 102-105 ^oC. ^1H NMR (400 MHz, $CDCl_3$) δ 8.23 (s, 1 H, aromatic CH), 8.03 (d, J = 0.8 Hz, 1 H, aromatic CH), 7.96 (m, 4 H, aromatic CH), 7.72 (m, 4 H, aromatic CH), 7.63 (m, 4 H, aromatic CH), 7.58 (d, J = 6.9 Hz, 2 H, aromatic CH), 7.52 (m, 2 H, aromatic CH), 7.46 (m, 5 H, aromatic CH), 7.40 (m, 2 H, aromatic CH), 7.13 (s, 1 H, 3-H of indolyl), 3.90 (s, 3 H, NCH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.4 and 196.1(Cq, C=O), 147.7, 145.3, 145.0, 140.2, 140.1, 139.0, 137.4, 137.2, 131.7, 130.1 and 128.6 (Cq), 130.8, 130.7, 129.6, 129.09, 129.06, 129.02, 128.9, 128.3, 128.2, 127.44, 127.42, 127.12, 127.10, 126.8, 116.1, 103.6 (CH), 31.8 (NCH_3). HRMS Calcd for $C_{41}H_{29}NO_2$ $[M+H]^+$: 568.2277; Found: 568.2269.



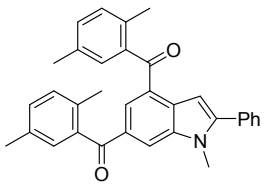
(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis((4-fluorophenyl)methanone) (3f): Yield 67%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.12 (s, 1 H, aromatic CH), 7.88 (m, 5 H, aromatic CH), 7.57-7.44 (m, 5 H, aromatic CH), 7.18 (m, 2 H, aromatic CH), 7.13 (m, 2 H, aromatic CH), 6.99 (s, 1 H, 3-H of indolyl), 3.88 (s, 3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 195.2 and 194.9 (Cq, C=O), 165.5 and 165.3 (Cq, d each, J = 252.0 Hz and 253.0 Hz, *i*-C of C_6H_4F), 147.8, 138.8, 134.8 and 134.6 (Cq, d each, J = 3.2 Hz and 3.0 Hz, *p*-C of C_6H_4F), 131.5, 130.7, 129.8 and 128.4 (Cq), 132.7 and 132.6 (CH, d each, J = 9.1 Hz and 9.0 Hz, *m*-C of C_6H_4F), 129.5, 129.1, 128.9, 126.0, 116.1, 115.58 and 115.57 (CH, d each, J = 21.8 Hz and 21.7 Hz, *o*-C of C_6H_4F), 103.4 (CH), 31.77 (CH_3). HRMS Calcd for $C_{29}H_{19}F_2NO_2$ $[M+H]^+$: 452.1462; Found: 452.1458.



(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis((4-chlorophenyl)methanone) (3g): Yield 70%. Yellow solid. M.p.: 208-211 ^{o}C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.11 (s, 1 H, aromatic CH), 7.87 (d, *J* = 1.3 Hz, 1 H, aromatic CH), 7.79 (m, 4 H, aromatic CH), 7.56-7.46 (m, 6 H, aromatic CH), 7.44 (m, 3 H, aromatic CH), 7.00 (s, 1 H, 3-H of indolyl), 3.87 (s, 3 H, NCH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 195.4 and 195.0 (Cq, C=O), 148.0, 139.0, 138.8, 138.7, 136.8, 136.7, 131.5, 130.8, 129.6 and 128.2 (Cq), 131.5, 131.4, 129.5, 129.1, 128.9, 128.8, 128.8, 126.1, 116.3 and 103.5 (CH), 31.8 (NCH_3). HRMS Calcd for $C_{29}H_{19}Cl_2NO_2$ $[M+H]^+$: 484.0871; Found: 484.0867.

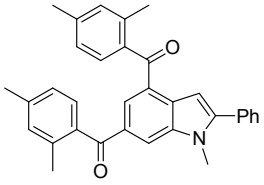


(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis((3,4-dimethylphenyl)methanone) (3h): Yield 65%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 and 7.91 (s each, 1:1 H, aromatic CH), 7.70 and 7.66 (s each, 1:1 H, aromatic CH), 7.58 (m, 4 H, aromatic CH), 7.47 (m, 3 H, aromatic CH), 7.22 (t, 2 H, aromatic CH), 7.04 (s, 1 H, 3-H of indolyl), 3.87 (s, 3 H, NCH_3), 2.34, 2.33 and 2.32 (s each, 3:6:3 H, 4×CH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.8 and 196.5 (Cq, C=O), 147.2, 142.0, 141.6, 138.9, 136.8, 136.8, 136.4, 136.2, 131.8, 130.6 and 130.5 (Cq), 131.3, 131.2, 129.5, 129.5, 128.9, 128.8, 128.2, 128.0, 126.1, 115.7 and 103.4 (CH), 31.7 (NCH_3), 20.11, 20.09, 19.91 and 19.89 (CH_3). HRMS Calcd for $C_{33}H_{29}NO_2$ $[M+H]^+$: 472.2277; Found: 472.2283.

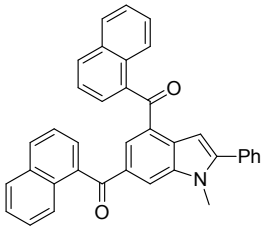


(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis((2,5-dimethylphenyl)methanone) (3i): Yield 55%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 (s, 1 H, aromatic

CH), 7.81 (d, *J* = 1.3 Hz, 1 H, aromatic CH), 7.56 (m, 2 H, aromatic CH), 7.54-7.45 (m, 3 H, aromatic CH), 7.25 (s, 1 H, 3-H of indolyl), 7.14 (m, 6 H, aromatic CH), 3.85 (s, 3 H, NCH_3), 2.31 2.30, 2.27 and 2.25 (s each, 3:3:3:3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 199.5 and 198.4 (Cq, C=O), 148.6, 139.7, 139.2, 139.2, 134.85, 134.80, 133.3, 133.1, 131.7 and 130.5 (Cq), 130.9, 130.8, 130.8, 129.6, 129.1, 128.93, 128.88, 128.6, 128.3, 116.5, 104.2 (CH), 31.8 (NCH_3), 21.0 and 19.6 (CH_3). HRMS Calcd for $C_{33}H_{29}NO_2$ $[M+H]^+$: 472.2277; Found: 472.2278.

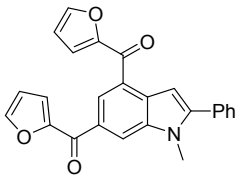


(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis((2,4-dimethylphenyl)methanone) (3j): Yield 50%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.08 (d, *J* = 1.0 Hz, 1 H, aromatic CH), 7.86 (d, *J* = 1.3 Hz, 1 H, aromatic CH), 7.56 (m, 2 H, aromatic CH), 7.50 (m, 2 H, aromatic CH), 7.48 (m, 1 H, aromatic CH), 7.26 (t, 2 H, aromatic CH), 7.18 (d, *J* = 0.6 Hz, 1 H, 3-H of indolyl), 7.08 (d, *J* = 3.7 Hz, 2 H, aromatic CH), 7.02 (t, *J* = 7.4 Hz, 2 H, aromatic CH), 3.83 (s, 3 H, NCH_3), 2.38, 2.37, 2.36 and 2.33 (s each, 3:3:3:3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 199.1 and 198.1 (Cq, C=O), 148.2, 140.5, 140.3, 139.0, 137.2, 137.0, 136.7, 136.2, 131.7, 130.80, 130.78 and 129.3 (Cq), 131.95, 131.90, 129.5, 129.4, 129.0, 128.9, 128.85, 127.84, 126.0, 125.9, 116.6 and 104.1 (CH), 31.7 (NCH_3), 21.5, 20.2 and 20.1 (CH_3). HRMS Calcd for $C_{33}H_{29}NO_2$ $[M+H]^+$: 472.2277; Found: 472.2283.

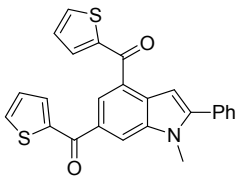


(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis(naphthalen-1-ylmethanone) (3k): Yield 52%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.29 and 7.91 (s each, 1:1 H, aromatic CH), 8.15 (d, *J* = 8.3 Hz, 1 H, aromatic CH), 8.02 (d, *J* = 8.3 Hz, 1 H, aromatic CH), 7.87 (m, 4 H, aromatic CH), 7.59 (m, 2 H, aromatic CH), 7.52 (m, 5 H,

aromatic CH), 7.48 (m, 2 H, aromatic CH), 7.44 (m, 2 H, aromatic CH), 7.40 (s, 1 H, 3-H of indolyl), 7.36 (t, 1 H, aromatic CH), 7.33 (d, *J* = 7.1 Hz, 1 H, aromatic CH), 3.86 (s, 3 H, NCH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 198.3 and 197.2 (Cq, C=O), 148.9, 139.2, 137.05, 136.8, 133.83, 133.75, 131.6, 131.2, 131.0, 130.8 and 129.1 (Cq), 131.1, 131.0, 129.6, 129.3, 129.1, 128.9, 128.5, 128.0, 127.4, 127.15, 127.09, 126.43, 126.37, 125.89, 125.86, 124.40, 124.37, 116.9 and 104.4 (CH), 31.8 (NCH_3). HRMS Calcd for $C_{37}H_{25}NO_2$ $[M+H]^+$: 516.1964; Found: 516.1959.

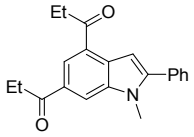


(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis(furan-2-ylmethanone) (3l): Yield 71%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.54 (d, *J* = 1.2 Hz, 1 H, aromatic CH), 8.34 (s, 1 H, aromatic CH), 7.73 (t, 2 H, aromatic CH), 7.56 (dd, *J* = 8.1, 1.4 Hz, 2 H, aromatic CH), 7.51 (m, 2 H, aromatic CH), 7.46 (m, 1 H, aromatic CH), 7.34 (d, *J* = 3.4 Hz, 1 H, furyl H), 7.30 (d, *J* = 3.4 Hz, 1 H, furyl H), 7.16 (s, 1 H, 3-H of indolyl), 6.62 (m, 2 H, furyl H), 3.89 (s, 3 H, NCH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 182.6 and 181.8 (Cq, C=O), 153.0, 152.9, 147.8, 138.9, 131.7, 130.7 and 128.1 (Cq), 147.1, 146.8, 129.6, 129.0, 128.9, 124.7, 120.5, 120.2, 115.8, 112.4, 112.3 and 103.4 (CH), 31.7 (NCH_3). HRMS Calcd for $C_{25}H_{17}NO_4$ $[M+H]^+$: 396.1236; Found: 396.1240.

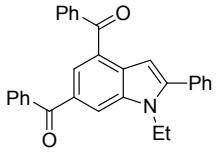


(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis(thiophen-2-ylmethanone) (3m): Yield 63%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.23 (d, *J* = 1.3 Hz, 1 H, aromatic CH), 8.20 (s, 1 H, aromatic CH), 7.75 (dd, *J* = 3.8 and 1.0 Hz, 1 H, thienyl H), 7.72 (m, 3 H, thienyl H), 7.56 (m, 2 H, aromatic CH), 7.54-7.43 (m, 3 H, aromatic CH), 7.19 (dd, *J* = 4.9 and 3.8 Hz, 1 H, thienyl H), 7.16 (dd, *J* = 4.7 and 4.0 Hz, 1 H, thienyl H), 7.08 (d, *J* = 0.5 Hz, 1 H, 3-H of indolyl), 3.89 (s, 3 H, NCH_3).

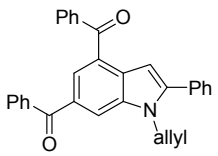
$^{13}C\{^{1}H\}$NMR (100 MHz, $CDCl_3$) δ 188.1 and 187.7 (Cq, C=O), 147.4, 144.5, 144.0, 138.8, 131.7, 130.6, 130.4 and 129.2 (Cq), 134.9, 134.5, 134.2, 133.8, 129.6, 129.0, 128.9, 128.2, 128.1, 124.1, 115.3 and 103.2 (CH), 31.8 (NCH_3). HRMS Calcd for $C_{25}H_{17}NO_2S_2$ $[M+H]^+$: 428.0779; Found: 428.0781.



1,1'-(1-Methyl-2-phenyl-1*H*-indole-4,6-diyl)bis(propan-1-one) (3n): Yield 35%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.43 and 8.23 (s each, 1:1 H, aromatic CH), 7.56 (d, J = 7.0 Hz, 2 H, aromatic CH), 7.49 (m, 3 H, aromatic CH), 7.40 (s, 1 H, 3-H of indolyl), 3.86 (s, 3 H, NCH_3), 3.18 (m, 4 H, 2×CH_3CH_2), 1.30 (m, 6 H, 2×CH_3CH_2). $^{13}C\{^{1}H\}$NMR (100 MHz, $CDCl_3$) δ 202.0 and 200.2 (Cq, C=O), 148.2, 139.3, 131.8, 129.9, 129.8 and 128.2 (Cq), 129.5, 129.0, 128.8, 122.6, 114.5 and 104.4 (CH), 32.9 and 32.0 (CH_3CH_2), 31.66 (NCH_3), 8.8 and 8.7 (CH_3CH_2). HRMS Calcd for $C_{21}H_{21}NO_2$ $[M+H]^+$: 320.1651; Found: 320.1651.

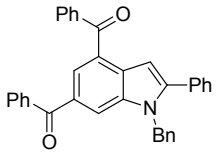


(1-Ethyl-2-phenyl-1H-indole-4,6-diyl)bis(phenylmethanone) (4a): Yield 62%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.20 and 7.94 (s each, 1:1 H, aromatic CH), 7.86 (m, 4 H, aromatic CH), 7.59-7.44 (m, 11 H, aromatic CH), 7.06 (s, 1 H, 3-H of indolyl), 4.33 (q, 2 H, CH_2CH_3), 1.37 (t, 3 H, CH_2CH_3). $^{13}C\{^{1}H\}$NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.4 (Cq, C=O), 147.3, 138.6, 138.5, 137.6, 132.0, 131.1, 129.8 and 128.5 (Cq), 132.5, 132.2, 130.2, 130.1, 129.4, 129.0, 128.9, 128.4, 126.8, 116.4 and 104.0 (CH), 39.3 (CH_2CH_3), 15.9 (CH_2CH_3). HRMS Calcd for $C_{30}H_{23}NO_2$ $[M+H]^+$: 430.1807; Found: 430.1806.

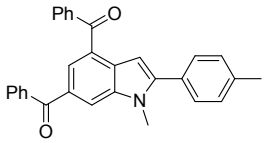


(1-Allyl-2-phenyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4b): Yield 61%.

Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.10 and 7.96 (s each, 1:1 H, aromatic CH), 7.88 (d, *J* = 7.5 Hz, 2 H, aromatic CH), 7.82 (d, *J* = 7.5 Hz, 2 H, aromatic CH), 7.61–7.43 (m, 11 H, aromatic CH), 7.12 (s, 1 H, 3-H of indolyl), 6.02 (m, 1 H, CH_2CH=CH_2), 5.28 and 5.00 (m each, 1:1 H, CH_2CH=CH_2), 4.85 (s, 2 H, CH_2CH=CH_2). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.3 (Cq, C=O), 147.6, 138.7, 138.4, 138.2, 131.7, 131.1, 130.0 and 128.6 (Cq), 133.4, 132.5, 132.2, 130.2, 130.1, 129.3, 129.1, 128.9, 128.4, 128.3, 126.9, 117.4, 117.1 and 103.9 (CH), 46.9 (CH_2CH=CH_2). HRMS Calcd for $C_{31}H_{23}NO_2$ $[M+H]^+$: 442.1807; Found: 442.1819.

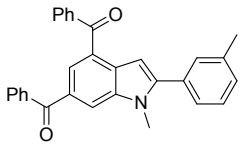


(1-Benzyl-2-phenyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4c): Yield 50%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98 (d, *J* = 1.1 Hz, 1 H, aromatic CH), 7.90 (m, 3 H, aromatic CH), 7.66 (m, 2 H, aromatic CH), 7.58 (m, 1 H, aromatic CH), 7.50 (m, 5 H, aromatic CH), 7.44 (m, 3 H, aromatic CH), 7.37 (t, 2 H, aromatic CH), 7.30 (m, 3 H, aromatic CH), 7.20 (s, 1 H, 3-H of indolyl), 7.00 (m, 2 H, aromatic CH), 5.49 (s, 2 H, CH_2Ph). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.9 and 195.9 (Cq, C=O), 147.8, 138.6, 138.1, 138.0, 137.3, 131.6, 131.1, 129.9 and 128.8 (Cq), 132.5, 132.1, 130.2, 130.1, 129.4, 129.1, 129.1, 128.9, 128.4, 128.3, 127.7, 126.9 and 126.1 (CH), 48.1 (CH_2Ph). HRMS Calcd for $C_{35}H_{25}NO_2$ $[M+H]^+$: 492.1964; Found: 492.1953.

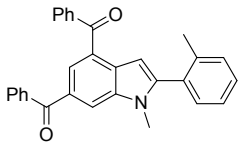


(1-Methyl-2-(*p*-tolyl)-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4d): Yield 68%. Yellow solid. M.p.: 138-140 ^{o}C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.16 and 7.94 (s each, 1:1 H, aromatic CH), 7.85 (m, 4 H, aromatic CH), 7.57 (m, 2 H, aromatic CH), 7.51-7.42 (m, 6 H, aromatic CH), 7.32 (d, *J* = 7.9 Hz, 2 H, aromatic CH), 7.05 (s, 1 H, 3-H of indolyl), 3.87 (s, 3 H, NCH_3), 2.44 (s, 3 H, CH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.4 (Cq, C=O), 147.9, 139.1, 138.8, 138.7, 138.5, 130.9, 129.7, 128.7 and 128.2 (Cq), 132.4, 132.1, 130.1, 130.0, 129.6, 129.4, 128.3, 126.9, 116.1

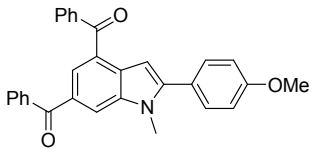
and 103.3 (CH), 31.7 (NCH_3), 21.5 (CH_3). HRMS Calcd for $C_{30}H_{23}NO_2$ $[M+H]^+$: 430.1807; Found: 430.1792.



(1-Methyl-2-(*m*-tolyl)-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4e): Yield 67%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.16 and 7.94 (s each, 1:1 H, aromatic CH), 7.89-7.80 (m, 4 H, aromatic CH), 7.57 (m, 2 H, aromatic CH), 7.48 (m, 4 H, aromatic CH), 7.40 (m, 2 H, aromatic CH), 7.35 (d, *J* = 7.5 Hz, 1 H, aromatic CH), 7.29-7.25 (m, 1 H, aromatic CH), 7.06 (d, *J* = 0.5 Hz, 1 H, 3-H of indolyl), 3.88 (s, 3 H, NCH_3), 2.45 (s, 3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.4 (Cq, C=O), 148.0, 138.9, 138.7, 138.7, 138.6, 131.6, 130.9 and 129.84 (Cq), 132.5, 132.2, 130.3, 130.17, 130.1, 129.8, 128.7, 128.4, 126.9, 126.6, 116.2 and 103.5 (CH), 31.8 (NCH_3), 21.6 (CH_3). HRMS Calcd for $C_{30}H_{23}NO_2$ $[M+H]^+$: 430.1807; Found: 430.1805.

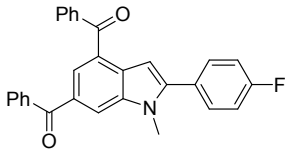


(1-Methyl-2-(*o*-tolyl)-1H-indole-4,6-diyl)bis(phenylmethanone) (4f): Yield 66%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.16 and 7.97 (s each, 1:1 H, aromatic CH), 7.85 (m, 4 H, aromatic CH), 7.57 (m, 2 H, aromatic CH), 7.48 (m, 4 H, aromatic CH), 7.40 (m, 1 H, aromatic CH), 7.37-7.28 (m, 3 H, aromatic CH), 6.93 (d, *J* = 0.4 Hz, 1 H, 3-H of indolyl), 3.65 (s, 3 H, NCH_3), 2.23 (s, 3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.5 (Cq, C=O), 147.0, 138.8, 138.5, 137.9, 137.87, 131.4 and 129.8 (Cq), 132.4, 132.2, 130.8, 130.5, 130.2, 130.1, 129.5, 128.4, 128.4, 126.8, 125.9, 116.0 and 103.7 (CH), 30.9 (NCH_3), 20.1 (CH_3). HRMS Calcd for $C_{30}H_{23}NO_2$ $[M+H]^+$: 430.1807; Found: 430.1803.

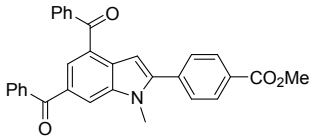


(2-(4-Methoxyphenyl)-1-methyl-1*H*-indole-4,6-diyl)bis(phenylmethanone)

(4g): Yield 51%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.15 and 7.94 (s each, 1:1 H, aromatic CH), 7.85 (m, 4 H, aromatic CH), 7.56 (m, 2 H, aromatic CH), 7.48 (m, 6 H, aromatic CH), 7.03 (m, 3 H, aromatic CH and 3-H of indolyl), 3.88 (s, 3 H, NCH_3), 3.86 (s, 3 H, OCH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.4 (Cq, C=O), 160.3, 147.8, 138.8, 138.7, 138.6, 131.0, 129.6, 128.1 and 124.0 (Cq), 132.4, 132.1, 130.9, 130.1, 130.1, 128.4, 127.0, 116.1, 115.1, 114.4 and 103.1 (CH), 55.5 (OCH_3), 31.7 (NCH_3). HRMS Calcd for $C_{30}H_{23}NO_3$ $[M+H]^+$: 446.1756; Found: 446.1764.

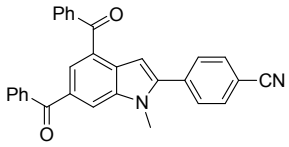


(2-(4-Fluorophenyl)-1-methyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4h): Yield 59%. Yellow solid. M.p.: 147-150 ^{o}C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.16 and 7.94 (s each, 1:1 H, aromatic CH), 7.85 (m, 4 H, aromatic CH), 7.60-7.51 (m, 4 H, aromatic CH), 7.48 (m, 4 H, aromatic CH), 7.20 (m, 2 H, aromatic C), 7.06 (s, 1 H, 3-H of indolyl), 3.85 (s, 3 H, CH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.8 and 196.4 (Cq, C=O), 163.2 (Cq, d, J = 248.1 Hz, *i*-C of C_6H_4F), 146.6, 138.8, 138.6, 138.4, 130.7 and 127.81 (Cq, d, J = 3.4 Hz, *p*-C of C_6H_4F) (Cq), 132.5, 132., 131.4 (CH, d, J = 8.3 Hz, *m*-C of C_6H_4F), 130.1, 130.1, 128.4, 126.9, 116.2, 116.0 (CH, d, J = 21.7 Hz, *o*-C of C_6H_4F), and 103.6 (CH), 31.7 (CH_3). HRMS Calcd for $C_{29}H_{20}FNO_2$ $[M+H]^+$: 434.1556; Found: 434.1551.

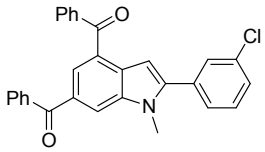


Methyl 4-(4,6-dibenzoyl-1-methyl-1*H*-indol-2-yl)benzoate (4i): Yield 62%. Yellow solid. M.p.: 206-209 ^{o}C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.17 (m, 3 H, aromatic CH), 7.94 (d, J = 1.3 Hz, 1 H, aromatic CH), 7.90-7.81 (m, 4 H, aromatic CH), 7.64 (d, J = 8.4 Hz, 2 H, aromatic CH), 7.57 (m, 2 H, aromatic CH), 7.47 (m, 4 H, aromatic CH), 7.15 (d, J = 0.4 Hz, 1 H, 3-H of indolyl), 3.96 (s, 3 H, NCH_3), 3.89 (s, 3 H, CO_2Me). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.7 and 196.3 (Cq, C=O), 166.6 (Cq, *C*O_2Me), 146.2, 139.2, 138.5, 138.3, 136.0, 130.6, 130.5, 130.4, and 128.7

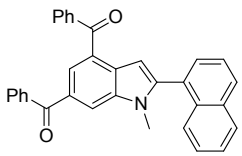
(Cq), 132.6, 132.3, 130.1, 130.1, 130.1, 129.4, 128.4, 126.8, 116.3 and 104.5 (CH), 52.4 (CO_2CH_3), 31.9 (CH_3). HRMS Calcd for $C_{31}H_{23}NO_4$ $[M+H]^+$: 474.1705; Found: 474.1697.



4-(4,6-Dibenzoyl-1-methyl-1*H*-indol-2-yl)benzonitrile (4j): Yield 52%. Yellow solid. M.p.: 214-217 ^oC. ^1H NMR (400 MHz, $CDCl_3$) δ 8.17 and 7.94 (s each, 1:1 H, aromatic CH), 7.88-7.77 (m, 6 H, aromatic CH), 7.68 (d, *J* = 8.4 Hz, 2 H, aromatic CH), 7.58 (m, 2 H, aromatic CH), 7.48 (m, 4 H, aromatic CH), 7.16 (s, 1 H, 3-H of indolyl), 3.88 (s, 3 H, NCH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.6 and 196.2 (Cq, C=O), 145.0, 139.3, 138.4, 138.2, 136.2, 130.9, 130.4, 128.9, 118.5 and 112.5 (Cq), 132.7, 132.6, 132.4, 130.1, 130.1, 129.9, 128.4, 126.9, 116.4 and 105.0 (CH), 32.0 (CH_3). HRMS Calcd for $C_{30}H_{20}N_2O_2$ $[M+H]^+$: 441.1603; Found: 441.1607.

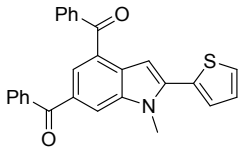


(2-(3-Chlorophenyl)-1-methyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4k): Yield 60%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.16 and 7.95 (s each, 1:1 H, aromatic CH), 7.84 (m, 4 H, aromatic CH), 7.57 (m, 3 H, aromatic CH), 7.52-7.42 (m, 7 H, aromatic CH), 7.09 (s, 1 H, 3-H of indolyl), 3.87 (s, 3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.7 and 196.3 (Cq, C=O), 145.9, 138.9, 138.5, 138.4, 134.8, 133.5, 130.6, 130.4 and 128.7 (Cq), 132.6, 132.3, 130.1, 130.1, 129.5, 129.1, 128.4, 127.6, 126.8, 116.3 and 104.1 (CH), 31.8 (CH_3). HRMS Calcd for $C_{29}H_{20}ClNO_2$ $[M+H]^+$: 450.1261; Found: 450.1253.

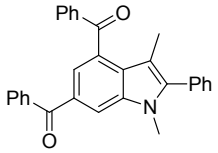


(1-Methyl-2-(naphthalen-1-yl)-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4l): Yield 61%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.21 (s, 1 H, aromatic CH), 8.00 (m, 2 H, aromatic CH), 7.95 (d, *J* = 7.8 Hz, 1 H, aromatic CH), 7.89 (m, 4 H,

aromatic CH), 7.66 (d, *J* = 8.3 Hz, 1 H, aromatic CH), 7.59 (m, 4 H, aromatic CH), 7.53 (m, 2 H, aromatic CH), 7.48 (m, 4 H, aromatic CH), 7.14 (s, 1 H, 3-H of indolyl), 3.62 (s, 3 H, NCH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.5 (Cq, C=O), 145.9, 138.7, 138.5, 138.4, 133.7, 132.5, 130.9, 130.0, 129.4 and 128.6 (Cq), 132.5, 132.2, 130.2, 130.1, 129.9, 129.1, 128.6, 128.4, 127.2, 126.8, 126.5, 125.8, 125.3, 116.1, 115.1 and 105.0 (CH), 31.4 (CH_3). HRMS Calcd for $C_{33}H_{23}NO_2$ $[M+H]^+$: 466.1807; Found: 466.1804.

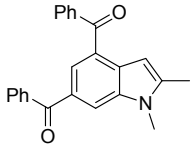


(1-Methyl-2-(thiophen-2-yl)-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4m): Yield 42%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.13 and 7.93 (s each, 1:1 H, aromatic CH), 7.84 (m, 4 H, aromatic CH), 7.56 (m, 2 H, aromatic CH), 7.47 (m, 5 H, aromatic CH and thienyl H), 7.33 (dd, *J* = 3.6 and 1.0 Hz, 1 H, thienyl H), 7.20 (d, *J* = 0.5 Hz, 1 H, 3-H of indolyl), 7.17 (dd, *J* = 5.0 and 3.7 Hz, 1 H, thienyl H), 3.97 (s, 3 H, NCH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 196.7 and 196.2 (Cq, C=O), 140.3, 138.9, 138.6, 138.4, 132.8, 130.54 and 128.3 (Cq), 132.5, 132.2, 130.1, 130.0, 128.4, 128.4, 128.03, 128.02, 127.7, 127.0, 116.0 and 104.2 (CH), 31.73 (CH_3). HRMS Calcd for $C_{27}H_{19}NO_2S$ $[M+H]^+$: 422.1215; Found: 422.1192.

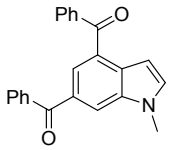


(1,3-Dimethyl-2-phenyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4n): Yield 63%. Yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 8.09 and 7.65 (s each, 1:1 H, aromatic CH), 7.94 (d, *J* = 7.5 Hz, 2 H, aromatic CH), 7.83 (d, *J* = 7.3 Hz, 2 H, aromatic CH), 7.60-7.51 (m, 3 H, aromatic CH), 7.48 (m, 6 H, aromatic CH), 7.40 (d, *J* = 6.8 Hz, 2 H, aromatic CH), 3.71 (s, 3 H, NCH_3), 1.99 (s, 3 H, CH_3). $^{13}C\{^1H\}$NMR (100 MHz, $CDCl_3$) δ 197.3 and 196.6 (Cq, C=O), 144.1, 138.6, 138.0, 137.1, 131.5, 131.1, 129.6 and 109.7 (Cq), 133.4, 132.1, 130.8, 130.7, 130.1, 128.8, 128.7, 128.6, 123.4 and 114.5 (CH), 31.5 (NCH_3), 11.9 (CH_3). HRMS Calcd for $C_{30}H_{23}NO_2$

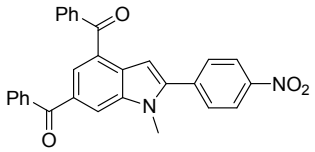
$[M+H]^+$: 430.1807; Found: 430.1797.



(1,2-Dimethyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4o): Yield 36%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.06 and 7.87 (s each, 1:1 H, aromatic CH), 7.81 (m, 4 H, aromatic CH), 7.55 (t, 2 H, aromatic CH), 7.46 (m, 4 H, aromatic CH), 6.76 (s, 1 H, 3-H of indolyl), 3.78 (s, 3 H, NCH_3), 2.50 (s, 3 H, CH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 197.0 and 196.5 (Cq, C=O), 144.2, 138.8, 138.6, 138.0, 131.1, 128.8 and 127.4 (Cq), 132.3, 132.0, 130.1, 130.0, 128.3, 128.3, 126.6, 115.2 and 102.2 (CH), 30.0 (NCH_3) , 13.3 (CH_3). HRMS Calcd for $C_{24}H_{19}NO_2$ $[M+H]^+$: 354.1494; Found: 354.1489.

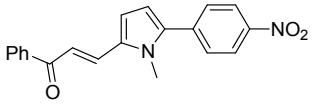


(1-Methyl-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4p): Yield 17%. Yellow liquid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12 (s, 1 H, aromatic CH), 7.91 (d, J = 1.0 Hz, 1 H, aromatic CH), 7.83 (m, 4 H, aromatic CH), 7.57 (m, 2 H, aromatic CH), 7.47 (m, 4 H, aromatic CH), 7.38 (d, J = 3.0 Hz, 1 H, 2-H of indolyl), 6.93 (d, J = 2.6 Hz, 1 H, 3-H of indolyl), 3.92 (s, 3 H, NCH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 196.9 and 196.5 (Cq, C=O), 138.6, 138.4, 137.2, 131.2, 130.1 and 129.0 (Cq), 134.8, 132.5, 132.3, 130.2, 130.1, 128.4, 126.1, 115.9 and 102.9 (CH), 33.4 (NCH_3). HRMS Calcd for $C_{23}H_{17}NO_2$ $[M+H]^+$: 340.1338; Found: 340.1339.

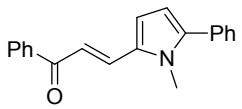


(1-Methyl-2-(4-nitrophenyl)-1*H*-indole-4,6-diyl)bis(phenylmethanone) (4q): Yield 38%. Yellow solid. M.p.: 206-209 ^{o}C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.36 (m, 2 H, aromatic CH), 8.17 and 7.95 (s each, 1:1 H, aromatic CH), 7.84 (m, 4 H, aromatic CH), 7.74 (m, 2 H, aromatic CH), 7.58 (m, 2 H, aromatic CH), 7.48 (m, 4 H, aromatic CH), 7.21 (s, 1 H, 3-H of indolyl), 3.91 (s, 3 H, NCH_3). ^{13}C{^{1}H}NMR (100 MHz,

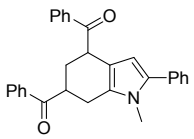
$CDCl_3$) δ 196.5 and 196.1 (Cq, C=O), 147.8, 144.5, 139.4, 138.3, 138.1, 138.0, 131.1, 130.3 and 129.0 (Cq), 132.7, 132.4, 130.1, 130.1, 130.05, 128.4, 126.9, 124.1, 116.4 and 105.3 (CH), 32.0 (CH_3). HRMS Calcd for $C_{29}H_{20}N_2O_4$ $[M+H]^+$: 461.1501; Found: 461.1499.



(*E*)-3-(1-Methyl-5-(4-nitrophenyl)-1*H*-pyrrol-2-yl)-1-phenylprop-2-en-1-one (5a): Yield 35%. Yellow solid. M.p.: 221-224 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.30 (m, 2 H, aromatic CH), 8.03 (m, 2 H, aromatic CH), 7.89 (d, *J* = 15.2 Hz, 1 H, C*H*=CHCOPh), 7.58 (m, 3 H, aromatic CH), 7.51 (m, 2 H, aromatic CH), 7.43 (d, *J* = 15.2 Hz, 1 H, CH=C*H*COPh), 6.95 and 6.48 (d each, *J* = 4.1 Hz and 4.1 Hz, 1:1 H, pyrrolyl CH), 3.80 (s, 3 H, NCH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 189.7 (Cq, C=O), 146.9, 138.7, 138.6, 136.9 and 134.0 (Cq), 132.8, 132.0, 129.2, 128.8, 128.4, 124.2, 118.6, 113.0 and 112.7 (CH), 32.8 (CH_3). HRMS Calcd for $C_{20}H_{16}N_2O_3$ $[M+H]^+$: 333.1239; Found: 333.1233.



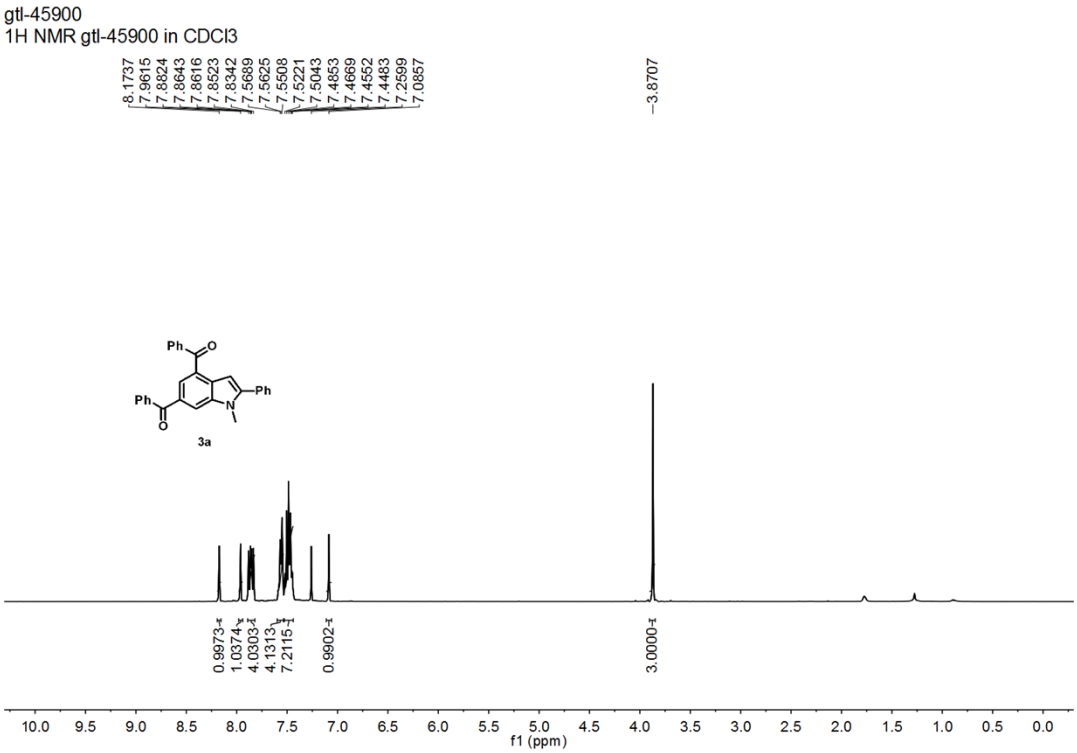
(*E*)-3-(1-Methyl-5-phenyl-1*H*-pyrrol-2-yl)-1-phenylprop-2-en-1-one (5b): Yield 26%. Yellow solid. M.p.: 96-99 °C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08-8.01 (m, 2 H, aromatic CH), 7.92 (d, *J* = 15.1 Hz, 1 H, C*H*=CHCOPh), 7.57 (t, 1 H, aromatic CH), 7.50 (m, 2 H, aromatic CH), 7.46 (m, 2 H, aromatic CH), 7.41 (m, 3 H, aromatic CH and CH=C*H*COPh), 7.37 (d, *J* = 6.8 Hz, 1 H, aromatic CH), 6.96 (d, *J* = 4.0 Hz, 1 H, pyrrolyl H), 6.35 (d, *J* = 3.9 Hz, 1 H, pyrrolyl H), 3.74 (s, 3 H, NCH_3). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 189.8 (Cq, C=O), 140.8, 139.0, 132.32 and 131.97 (Cq), 132.8, 132.4, 129.1, 128.7, 128.6, 128.3, 128.0, 116.6, 112.7 and 111.2 (CH), 32.4 (NCH_3). HRMS Calcd for $C_{20}H_{17}NO$ $[M+H]^+$: 288.1388; Found: 288.1387.



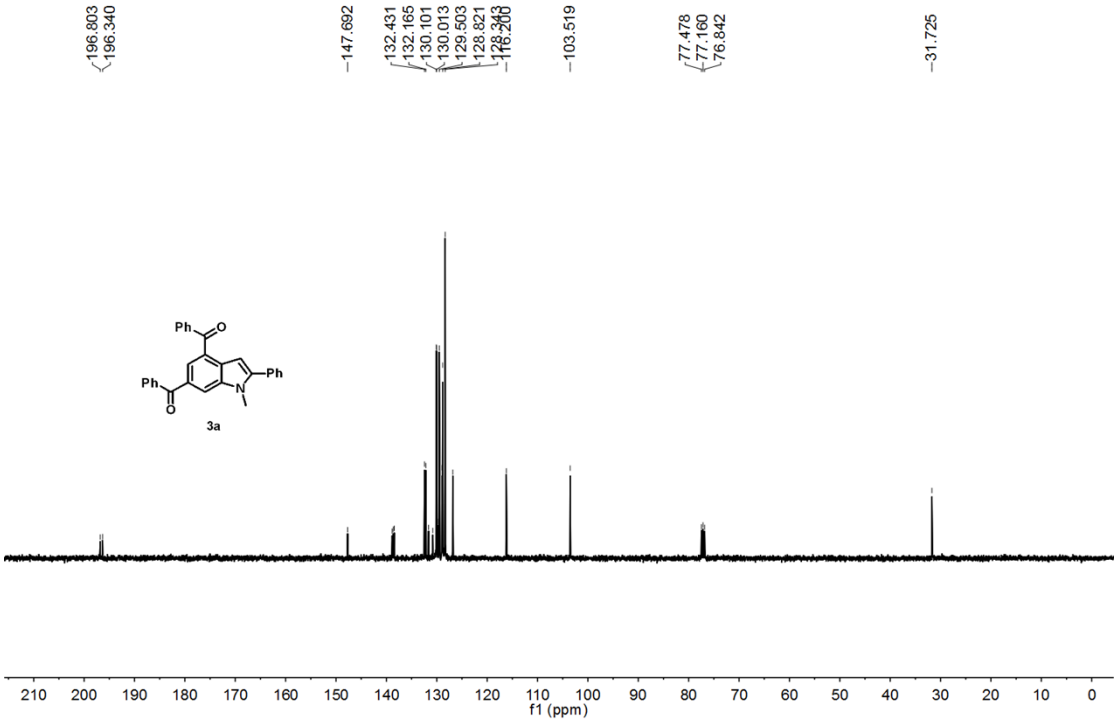
(1-Methyl-2-phenyl-4,5,6,7-tetrahydro-1*H*-indole-4,6-

diyl)bis(phenylmethanone) (7): Yield 18%. Yellow solid. M.p.: 155-158 ^{o}C. ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.30 (d, *J* = 7.2 Hz, 2 H, aromatic CH), 8.09 (d, *J* = 7.3 Hz, 2 H, aromatic CH), 7.59 (m, 2 H, aromatic CH), 7.53 (m, 4 H, aromatic CH), 7.34 (m, 2 H, aromatic CH), 7.27 (m, 3 H, aromatic CH), 5.83 (s, 1 H, 3-H of tetrahydroindole), 4.80 and 4.51 (m each, 1:1 H, 4-H and 6-H of tetrahydroindole), 3.50 (s, 3 H, NCH_3), 3.08 and 2.82 (dd each, *J* = 15.8, 10.8 Hz, 1:1 H, 7-CH_2 of tetrahydroindole), 2.60 and 2.01 (m each, 1:1 H, 5-CH_2 of tetrahydroindole). ^{13}C{^{1}H}NMR (100 MHz, $CDCl_3$) δ 203.1 and 202.1 (Cq, C=O), 137.4, 136.0, 134.2, 133.4, 130.1 and 112.90 (Cq), 133.3, 133.2, 129.03, 128.99, 128.9, 128.8, 128.8, 128.4, 126.8 and 107.1 (CH), 39.9 and 39.7 (4-CH and 6-CH of tetrahydroindole), 31.6 (NCH_3), 30.0 and 24.1 (5-CH_2 and 7-CH_2 of tetrahydroindole). HRMS Calcd for $C_{29}H_{25}NO_2$ $[M+H]^+$: 420.1964; Found: 420.1966.

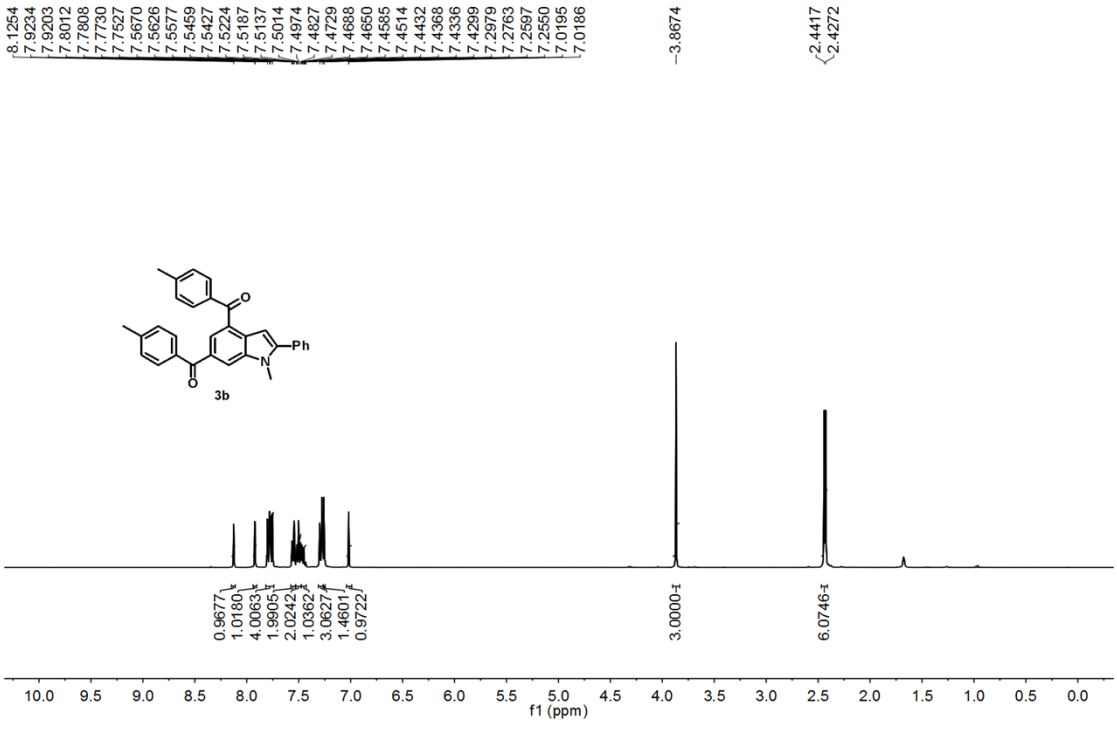
5. Copies of NMR spectra for new compounds



gtl-45900
13C NMR gtl-45900 CDCl3



gtl-42300
1H NMR gtl-42300 in CDCl3



gtl-42300
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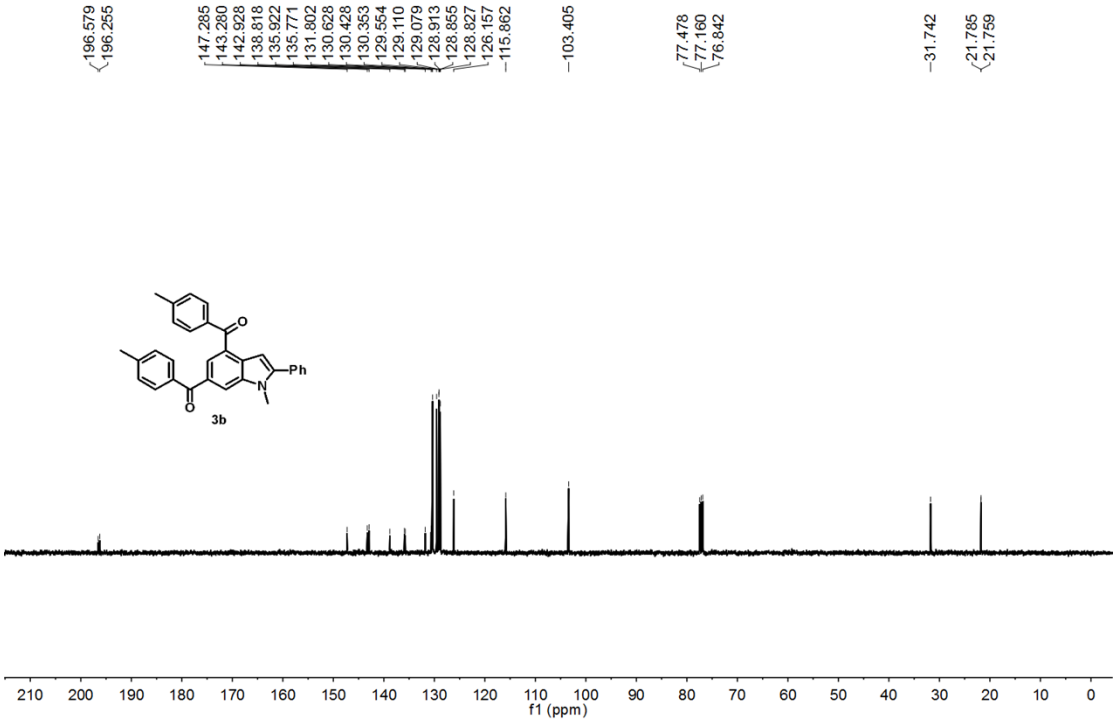
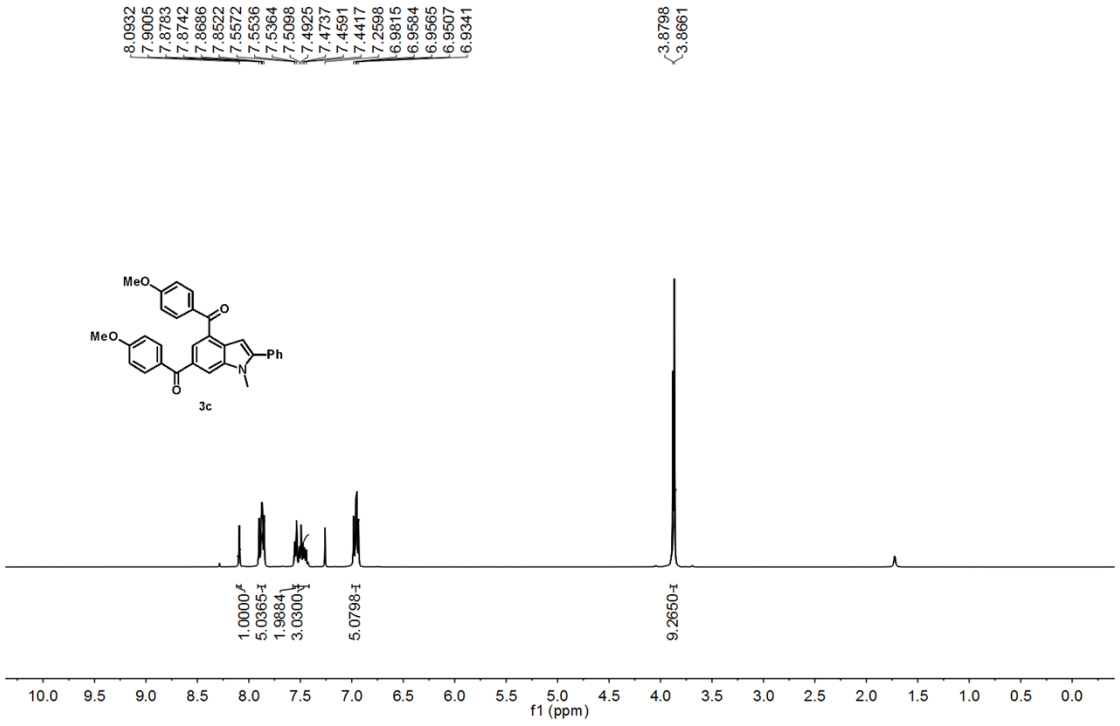
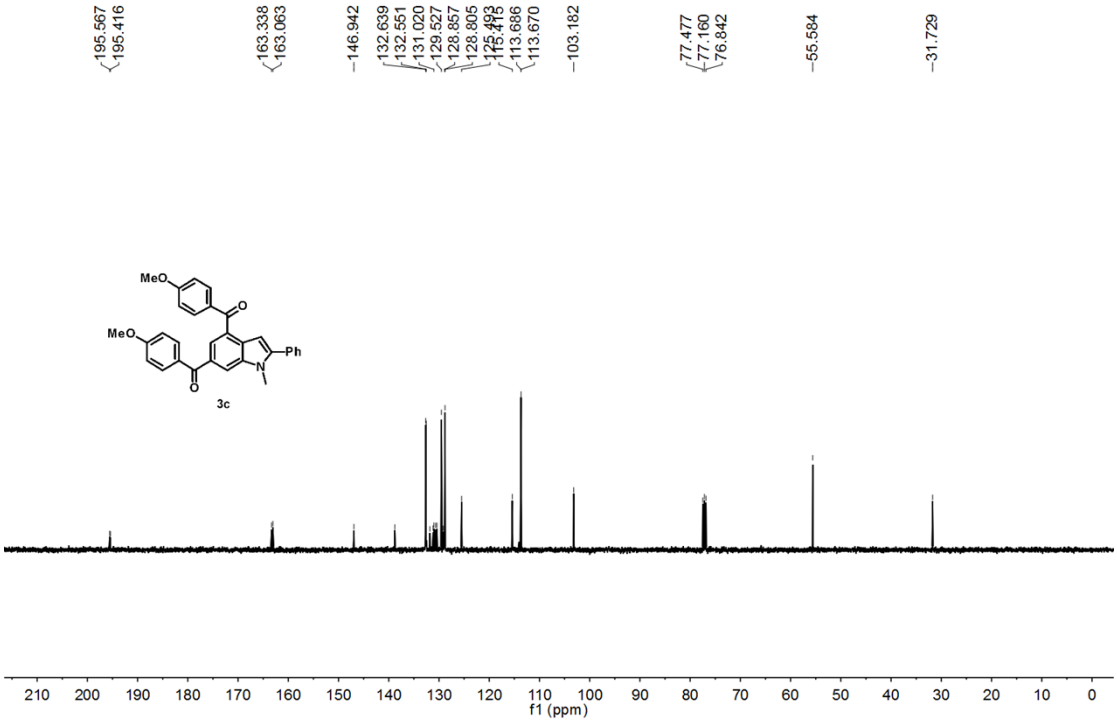


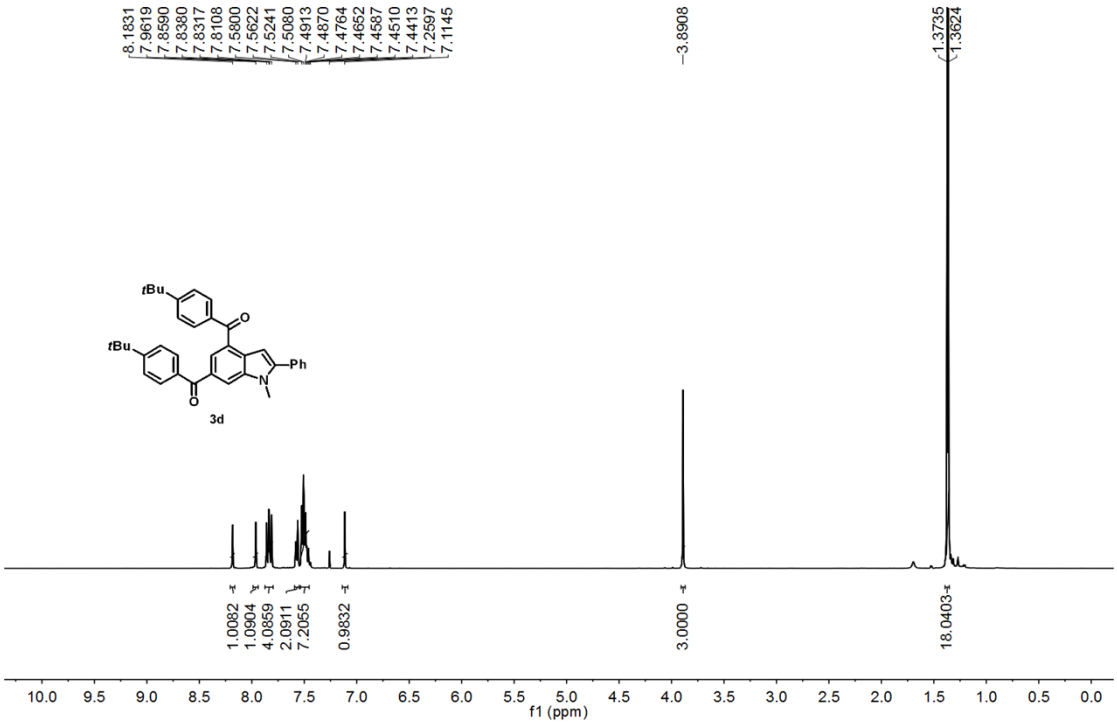
gtl-43102
1H NMR gtl-43102 in CDCl3



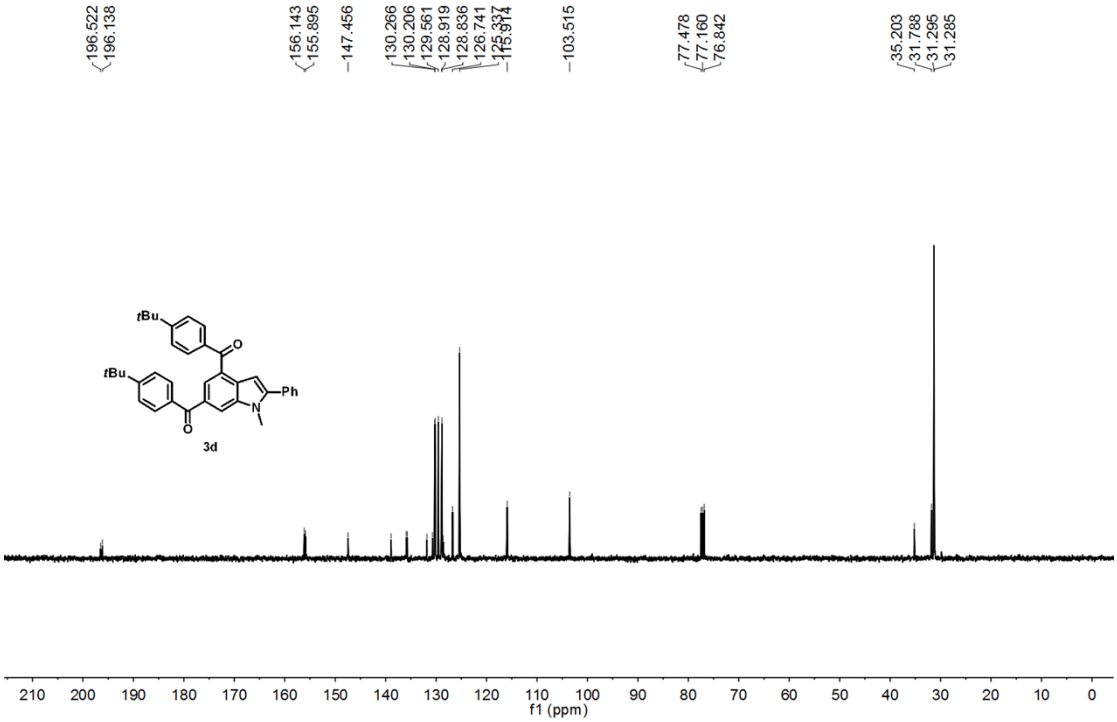
gtl-43102
13C NMR gtl-43102 CDCl3



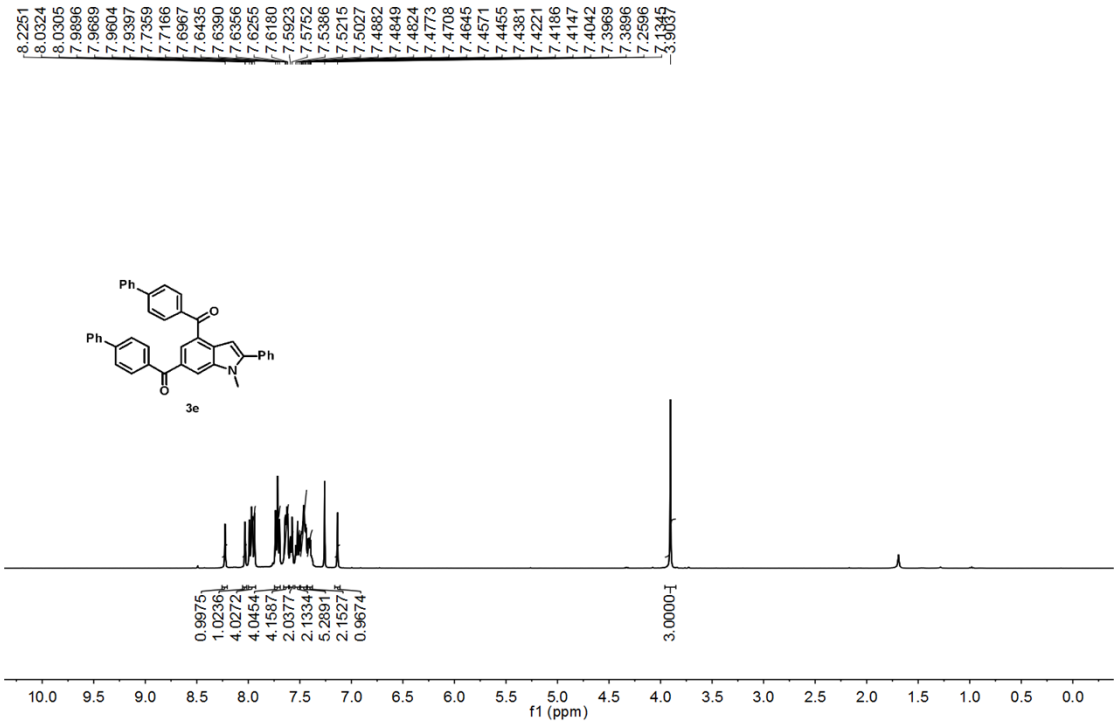
gtl-43200
1H NMR gtl-43200 in CDCl3



gtl-43200
13C NMR gtl-43200 CDCl3



gtl-43403
1H NMR gtl-43403 in CDCl3



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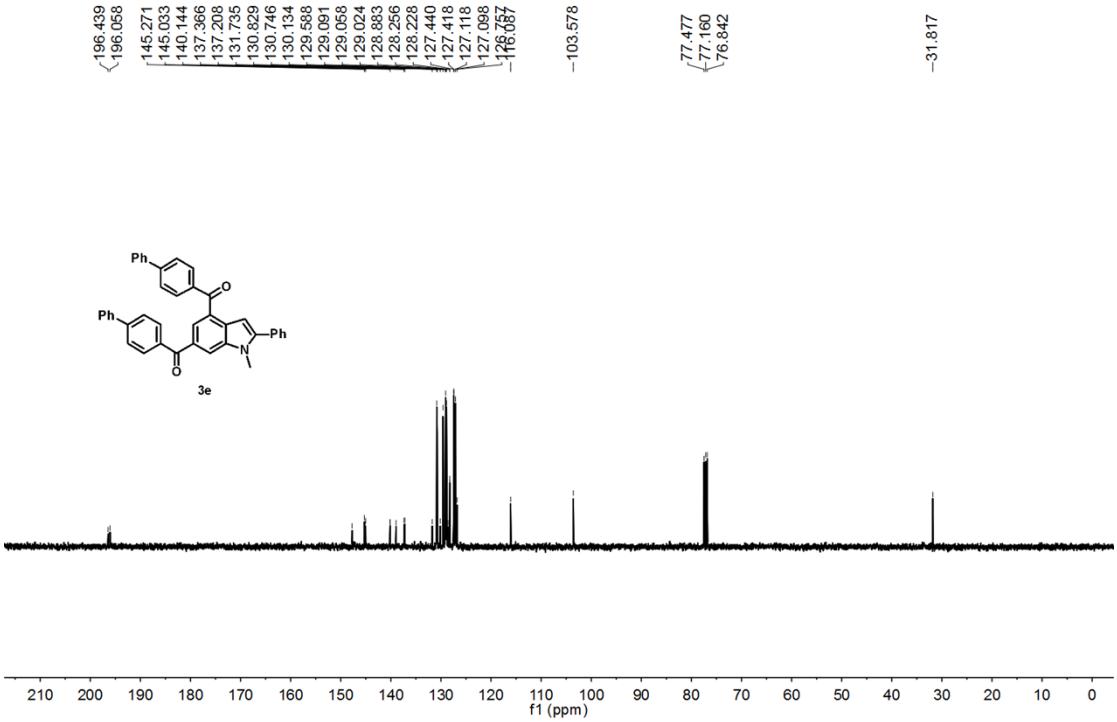
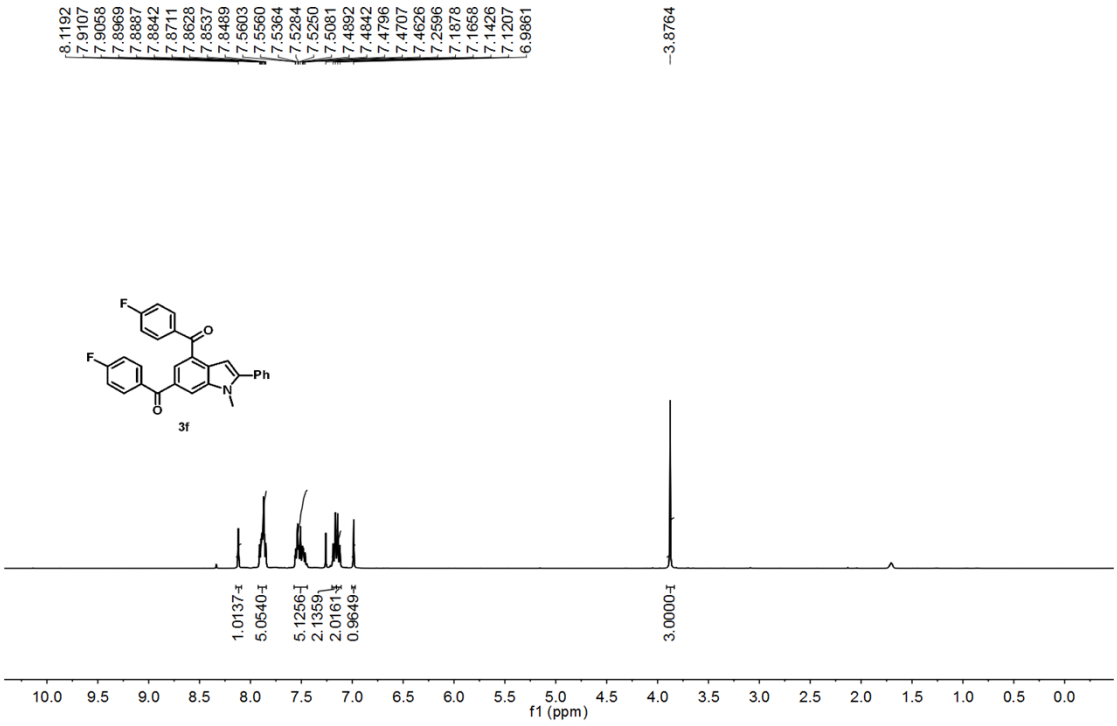
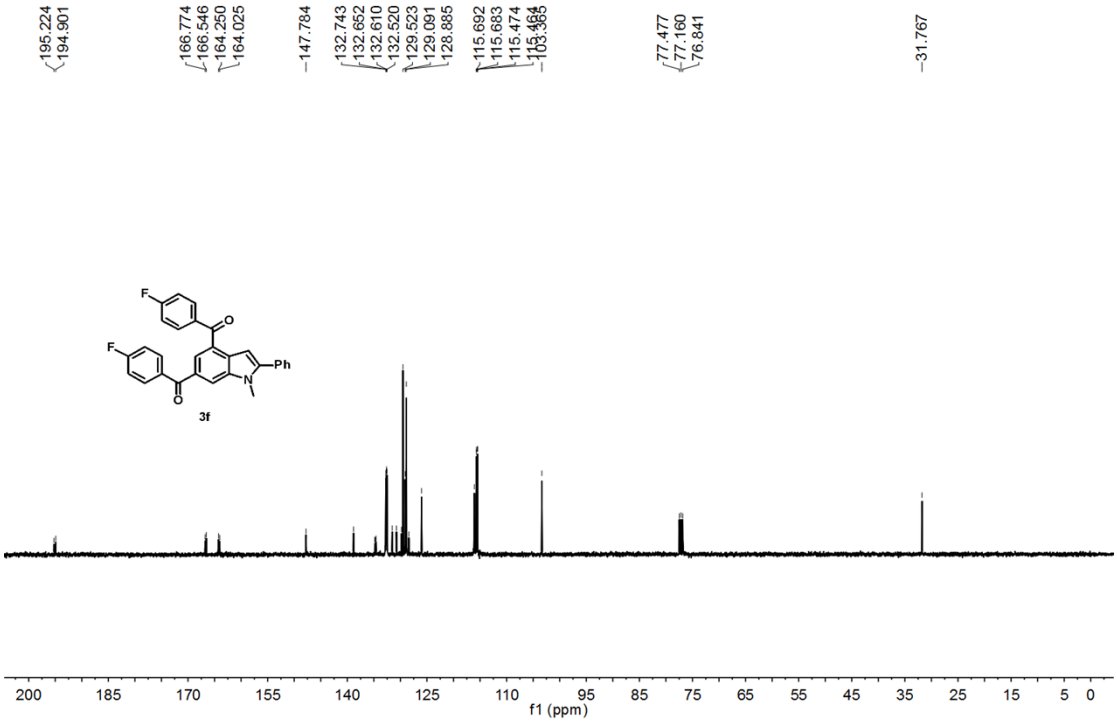


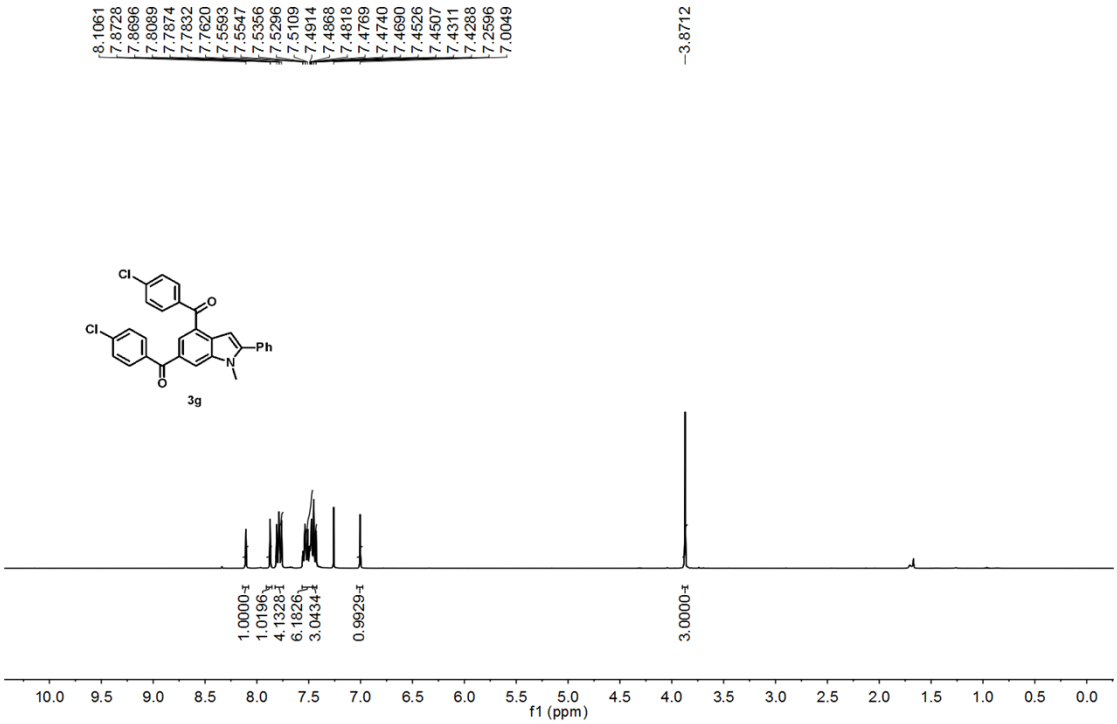
gtl-42900
1H NMR gtl-42900 in CDCl3



gtl-42900
13C NMR gtl-42900 CDCl3



gtl-43000
1H NMR gtl-43000 in CDCl3



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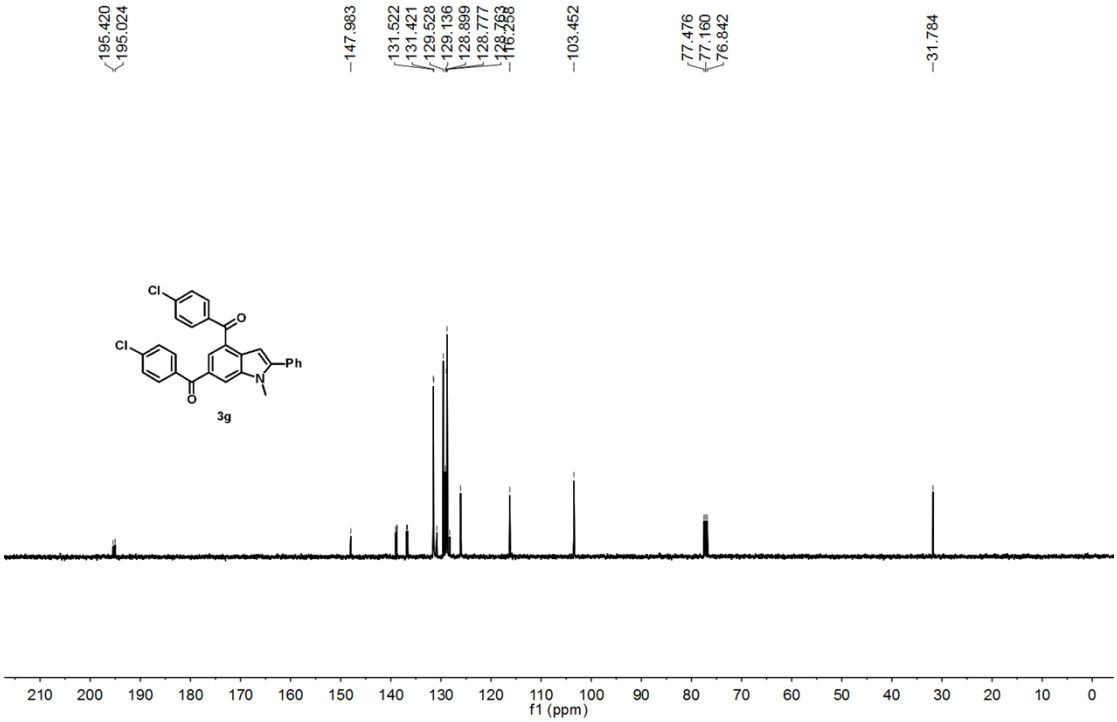
13C NMR gtl-43000 CDCl3



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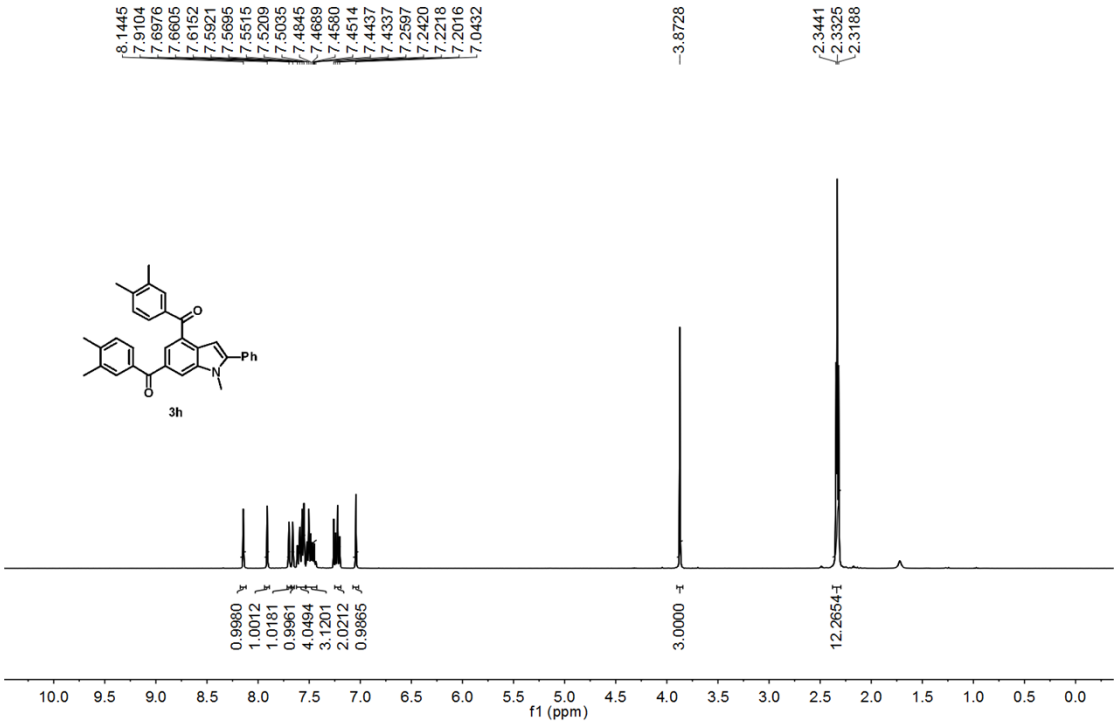
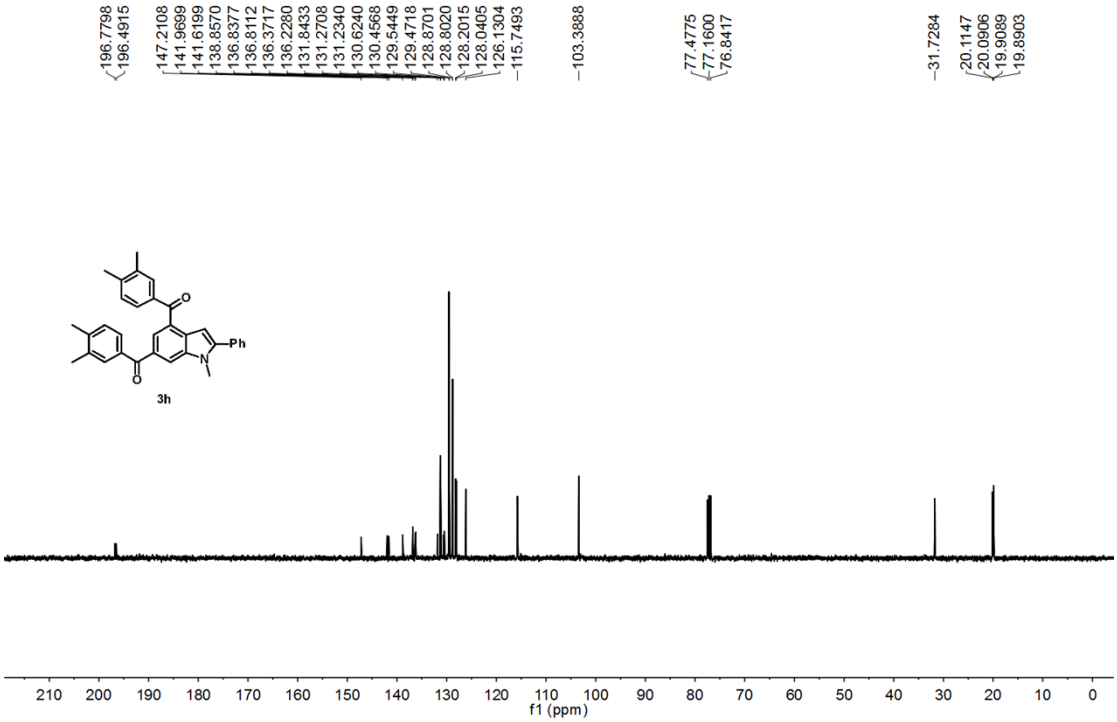
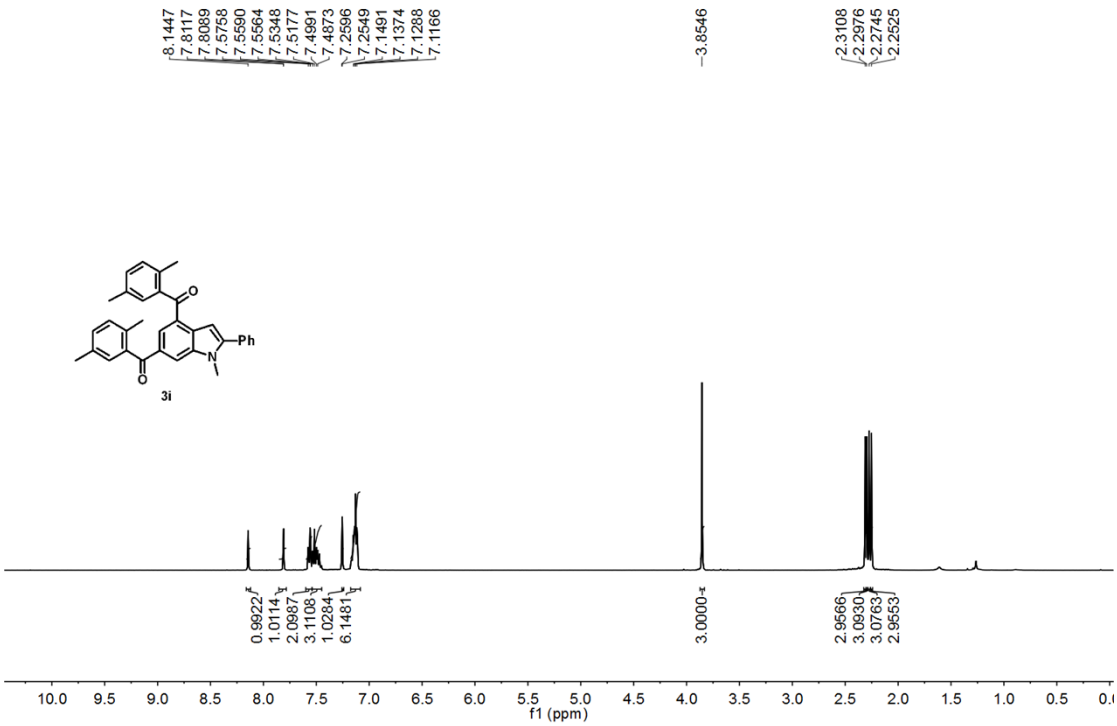


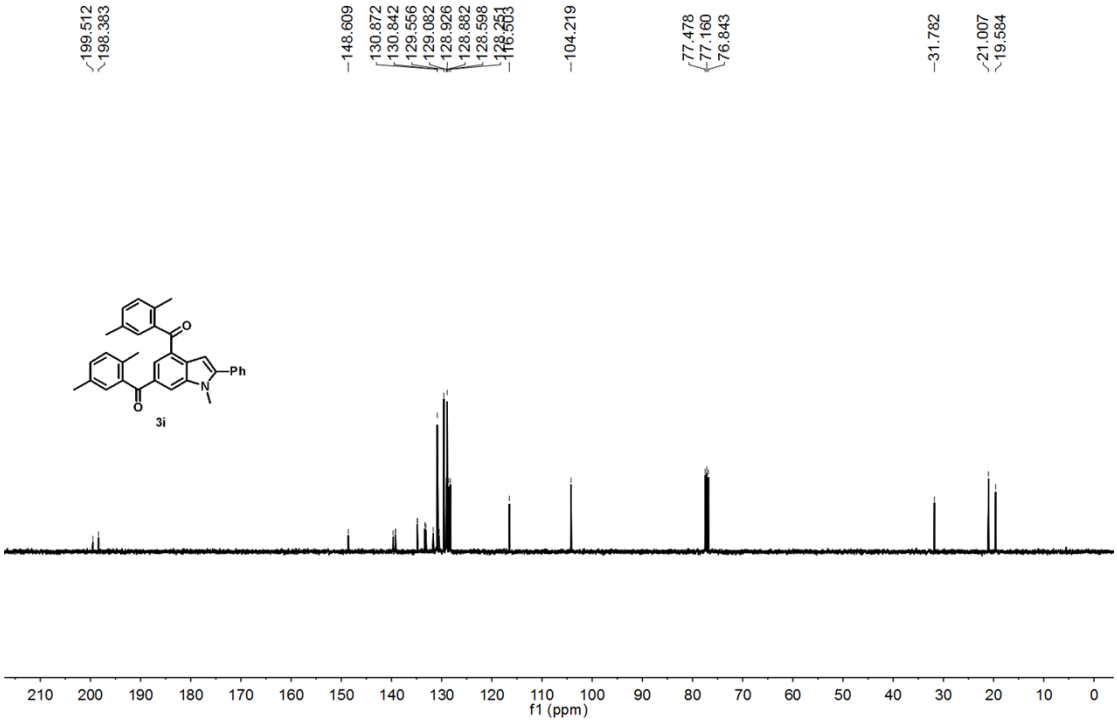
gtl-43300
13C NMR gtl-43300 CDCl3



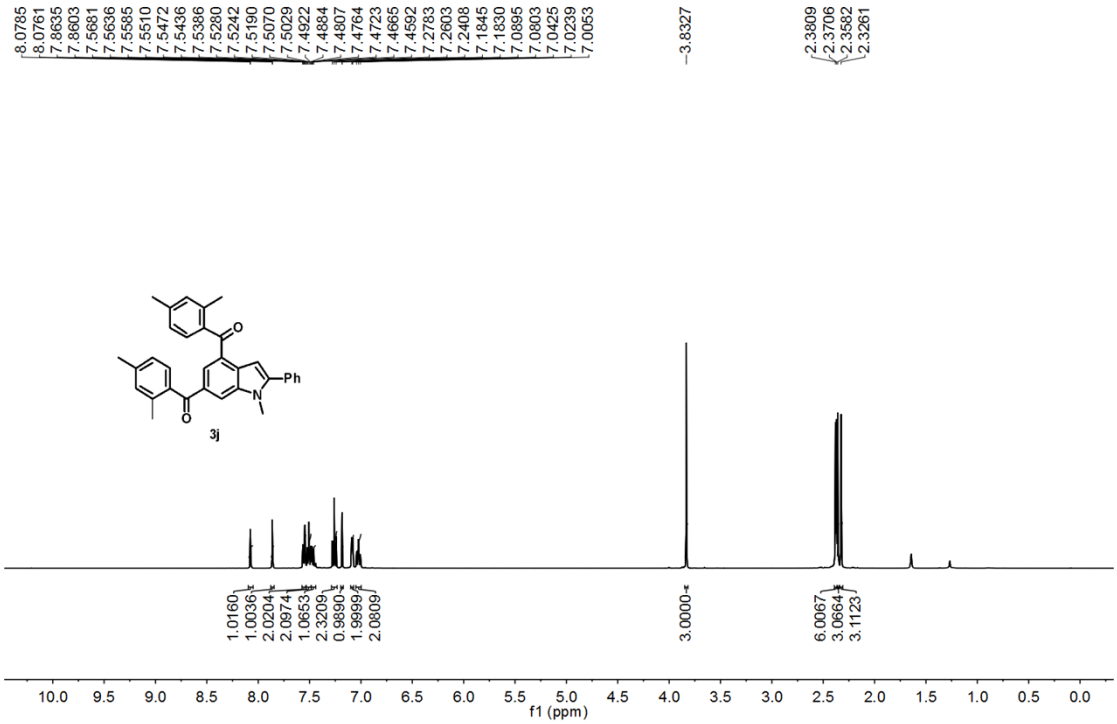
gtl-43600 in CDCl3



gtl-43600
13C NMR gtl-43600 CDCl3



gtl-43900
1H NMR gtl-43900 in CDCl3



gtl-43900
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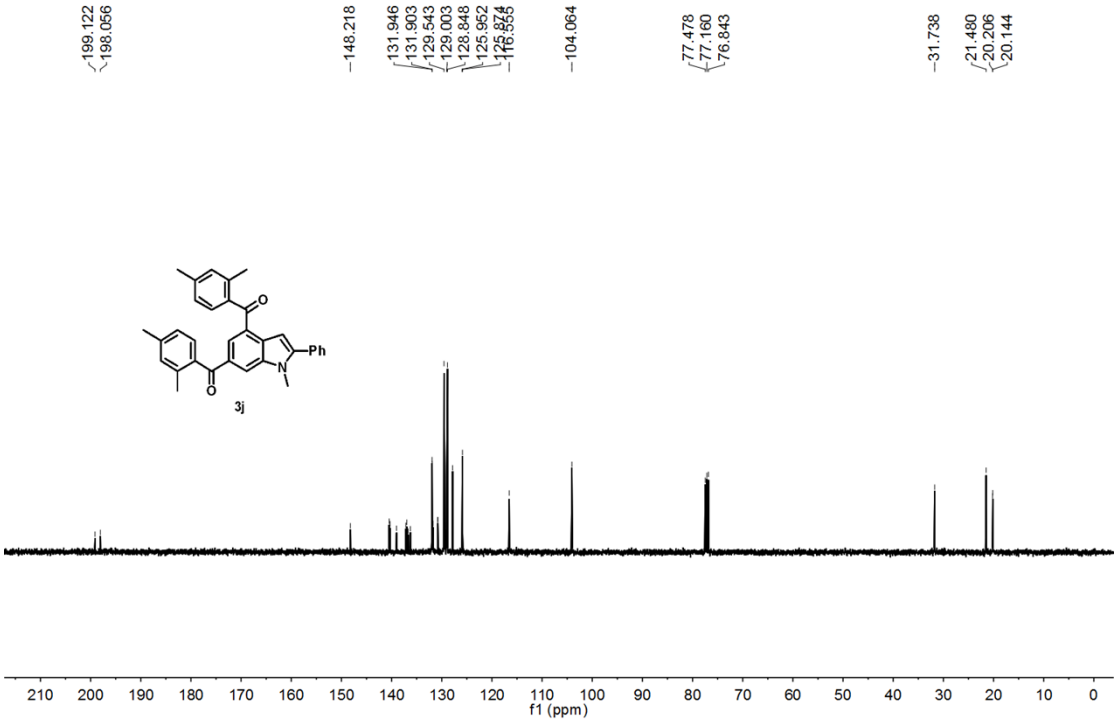

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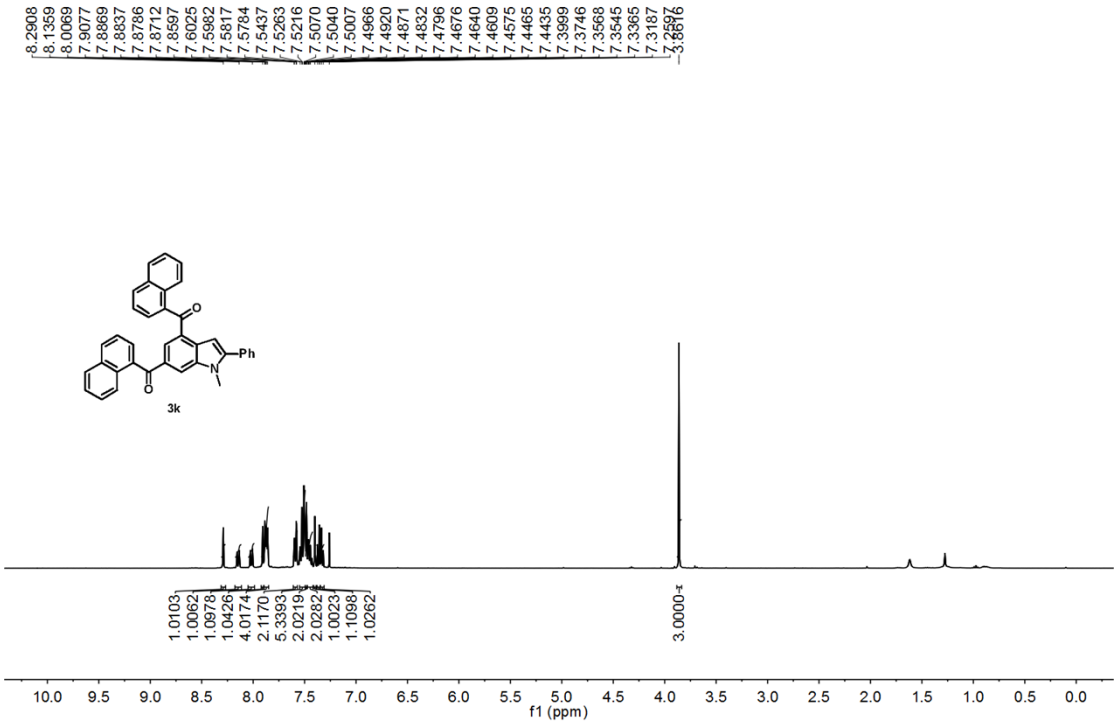

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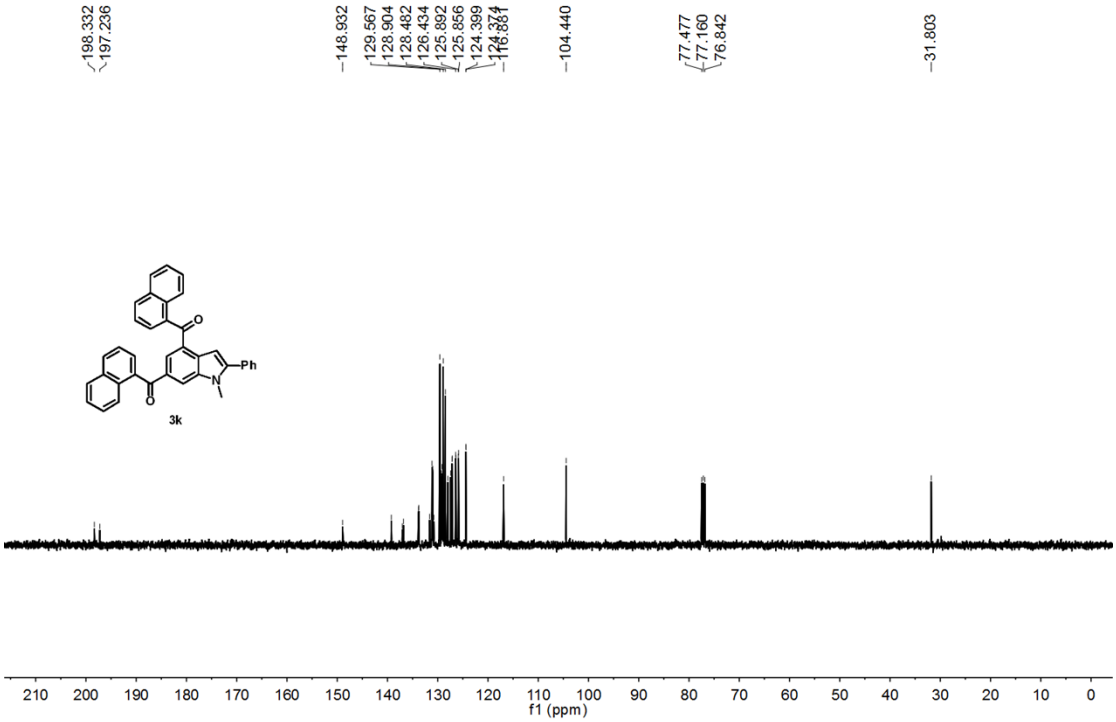
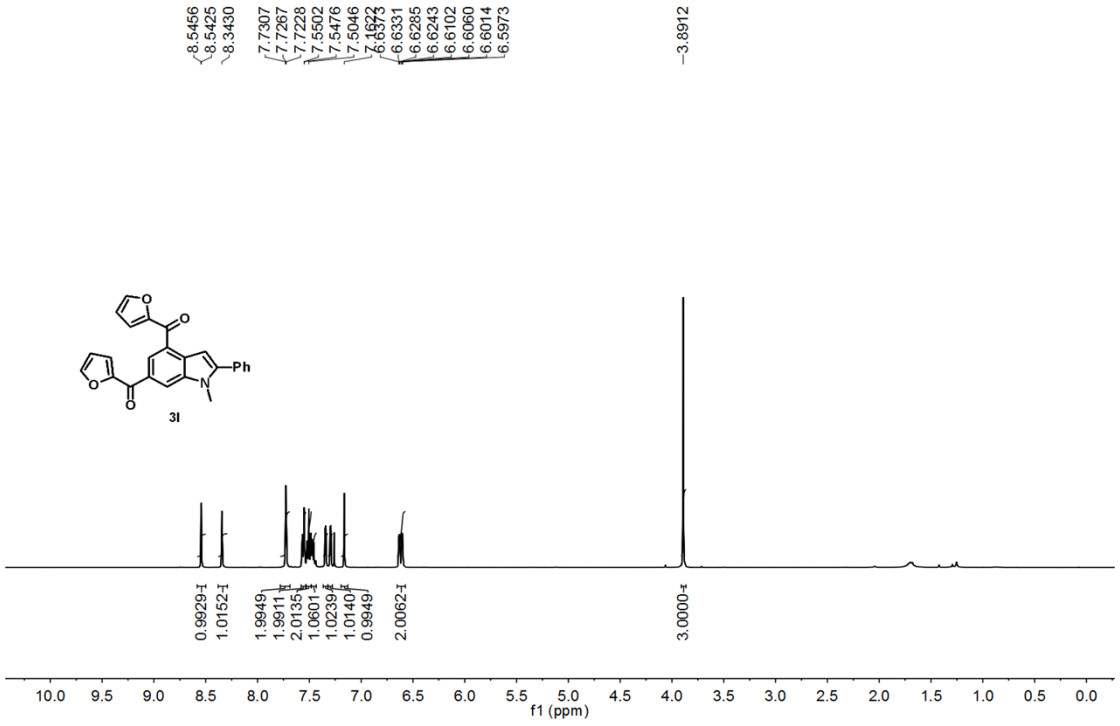


gtl-44100
1H NMR gtl-44100 in CDCl3



gtl-44100
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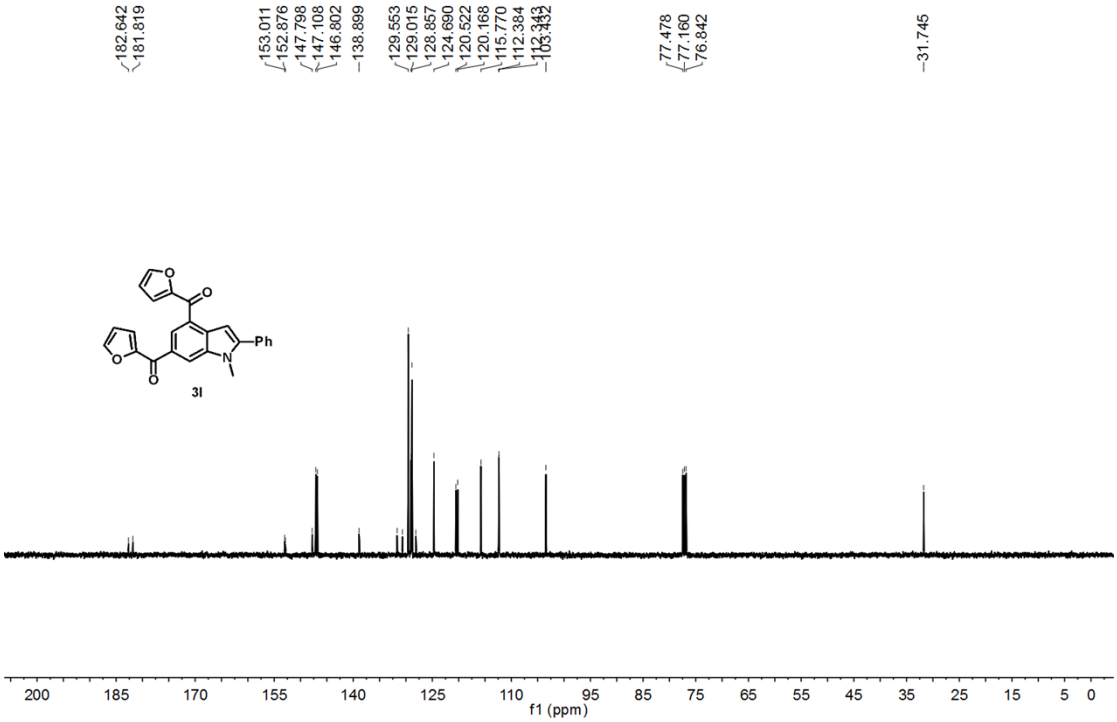

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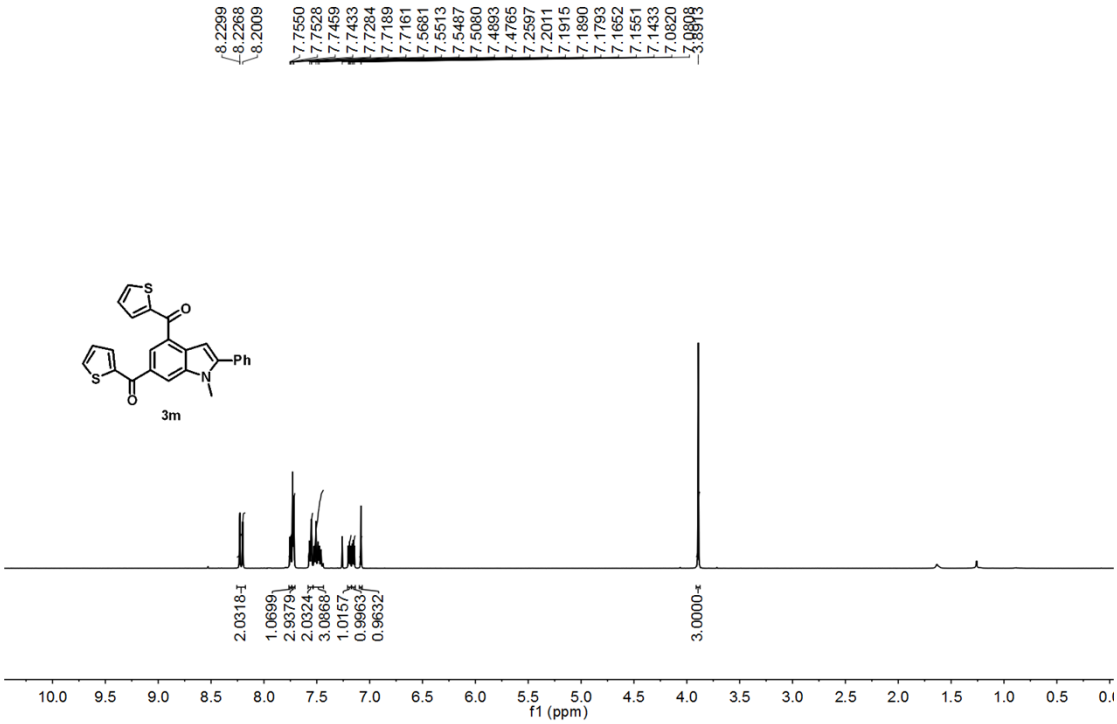

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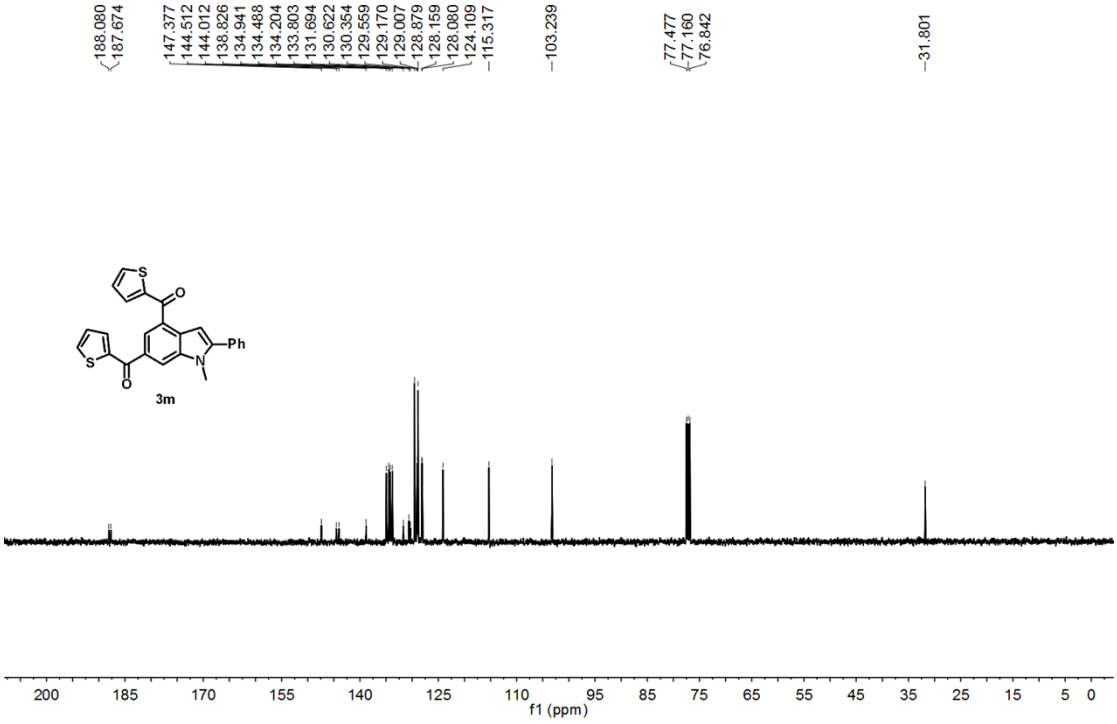

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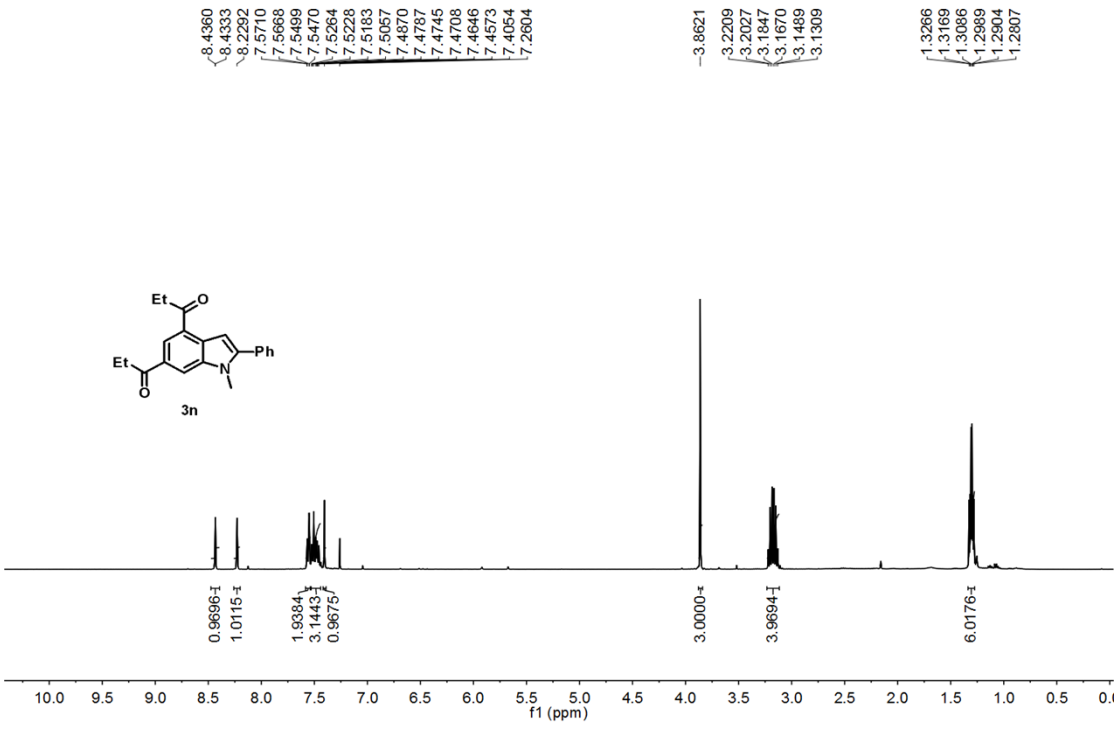
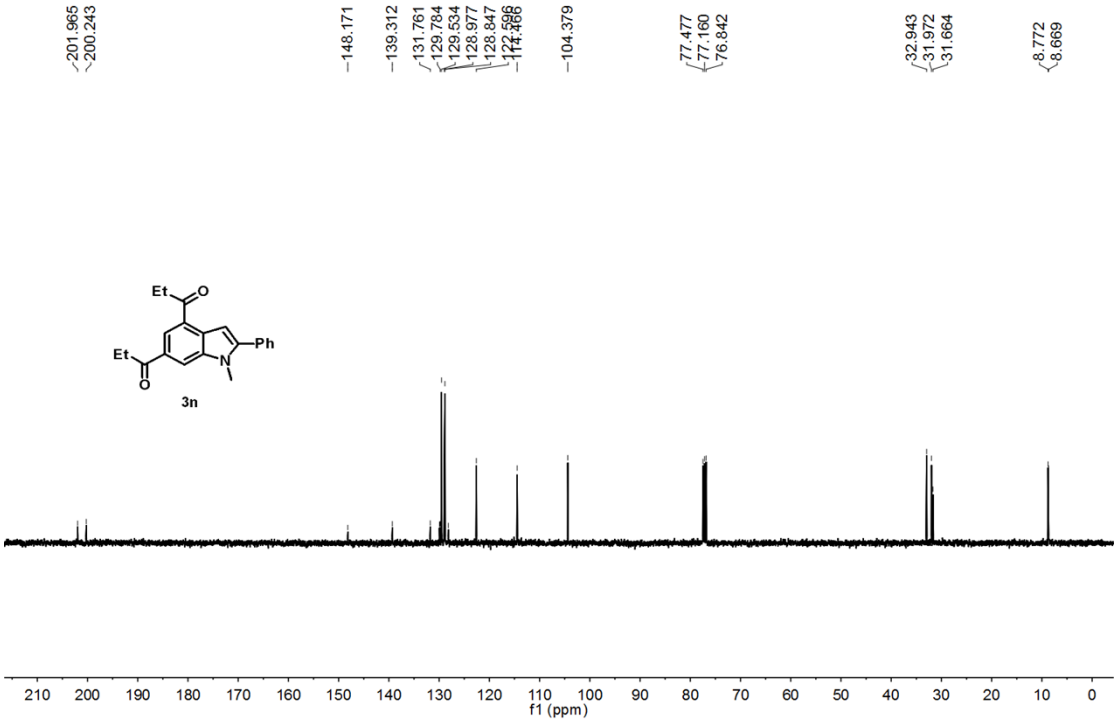
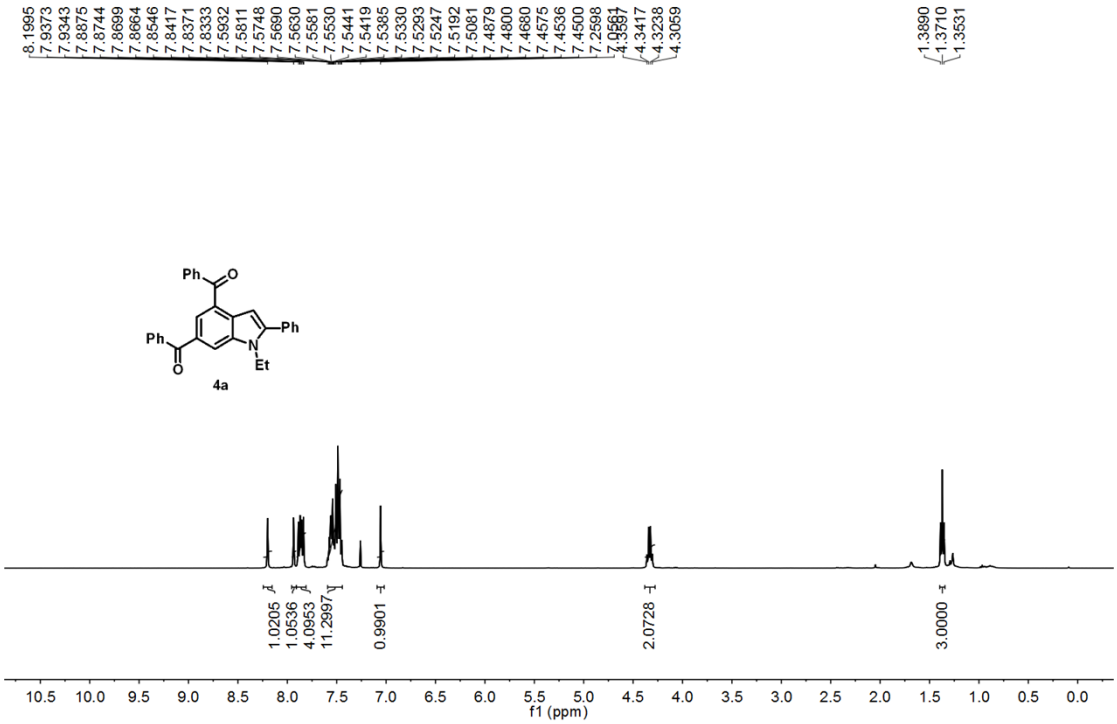


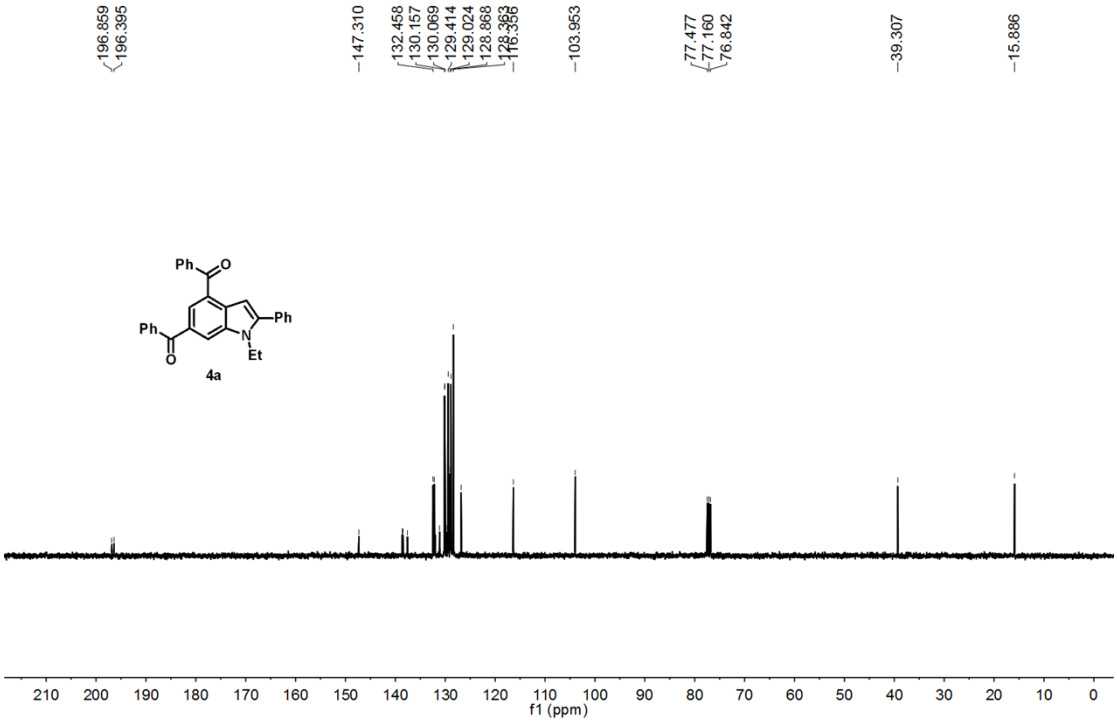
gtl-44604
13C NMR gtl-44604 CDCl3



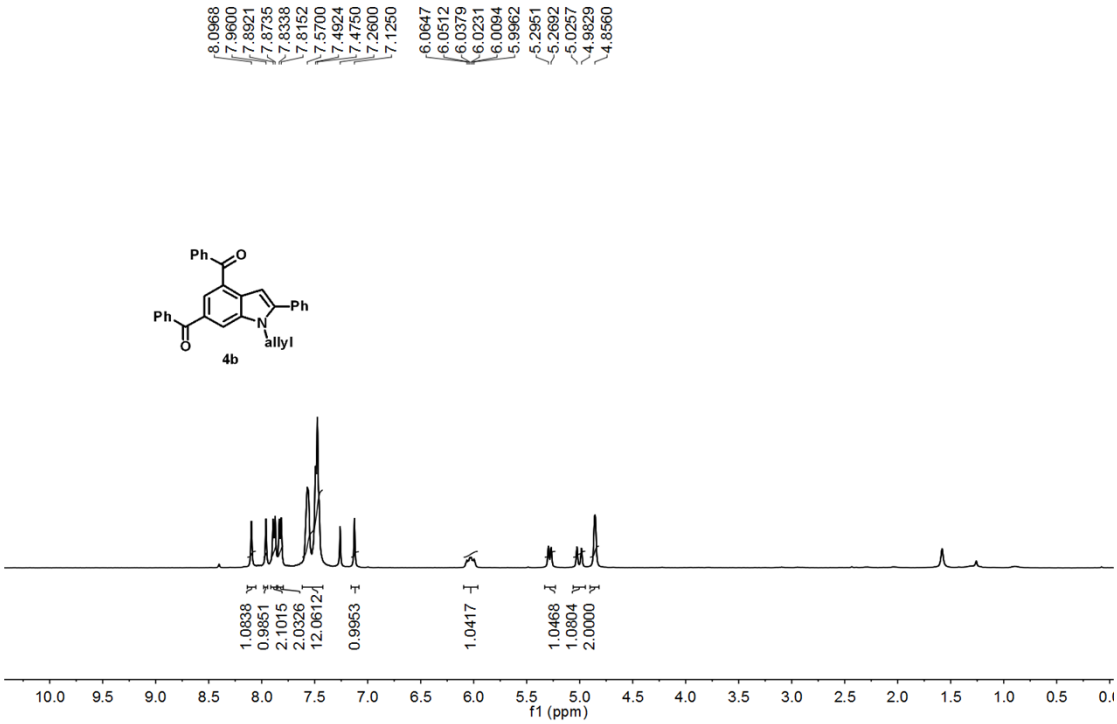
gtl-38900
1H NMR gtl-38900 in CDCl3



gtl-38900
13C NMR gtl-38900 CDCl3



gtl-47108 in CDCl3



gtl-47108
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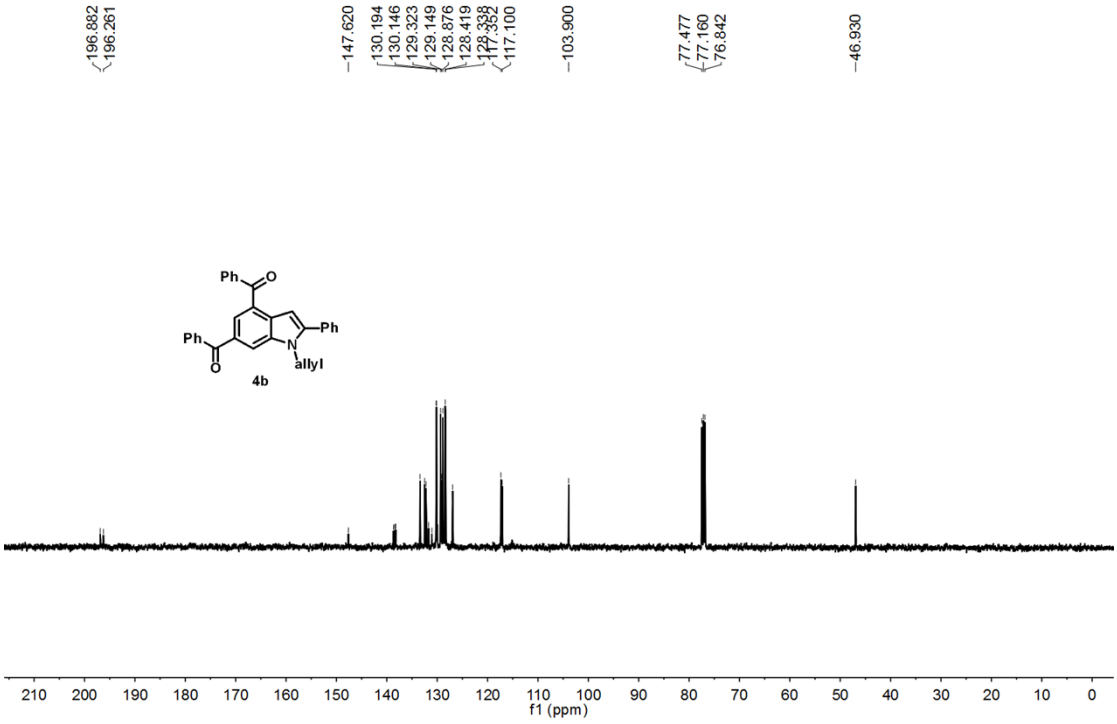

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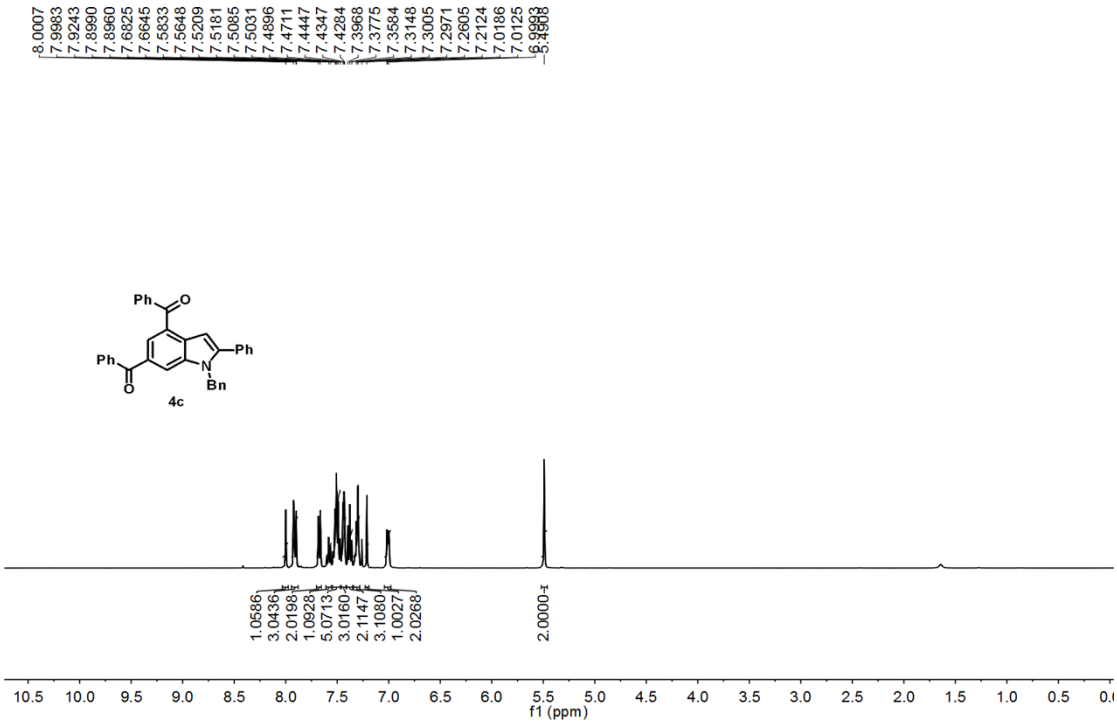

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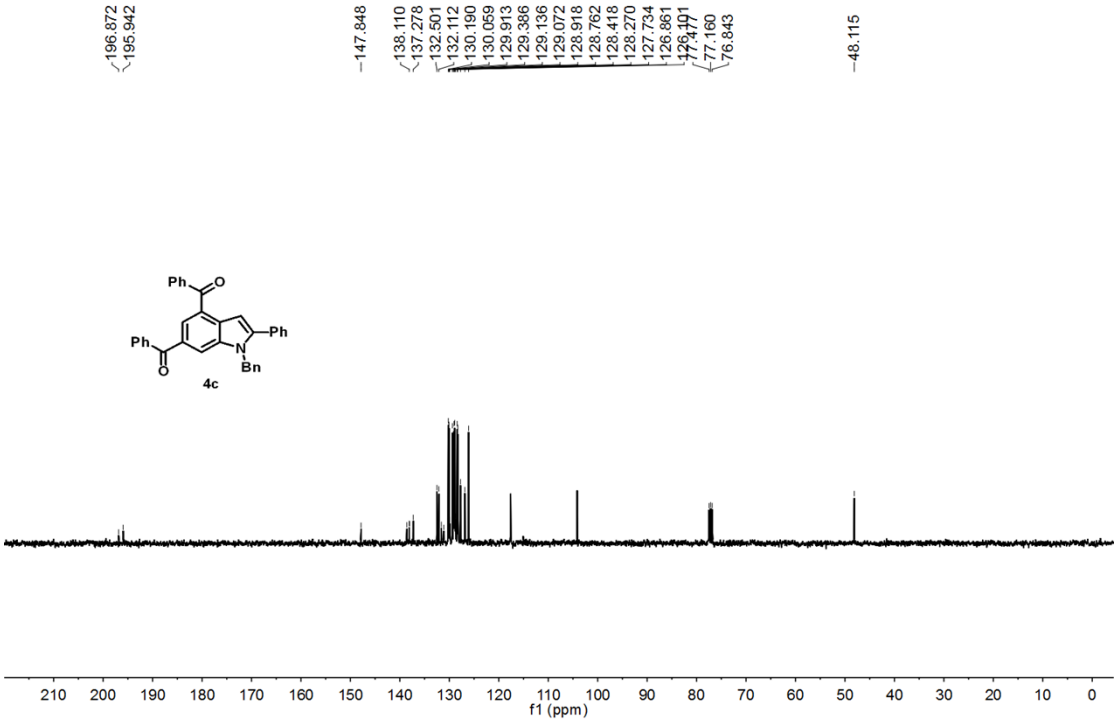
13C NMR gtl-39200 CDCl3



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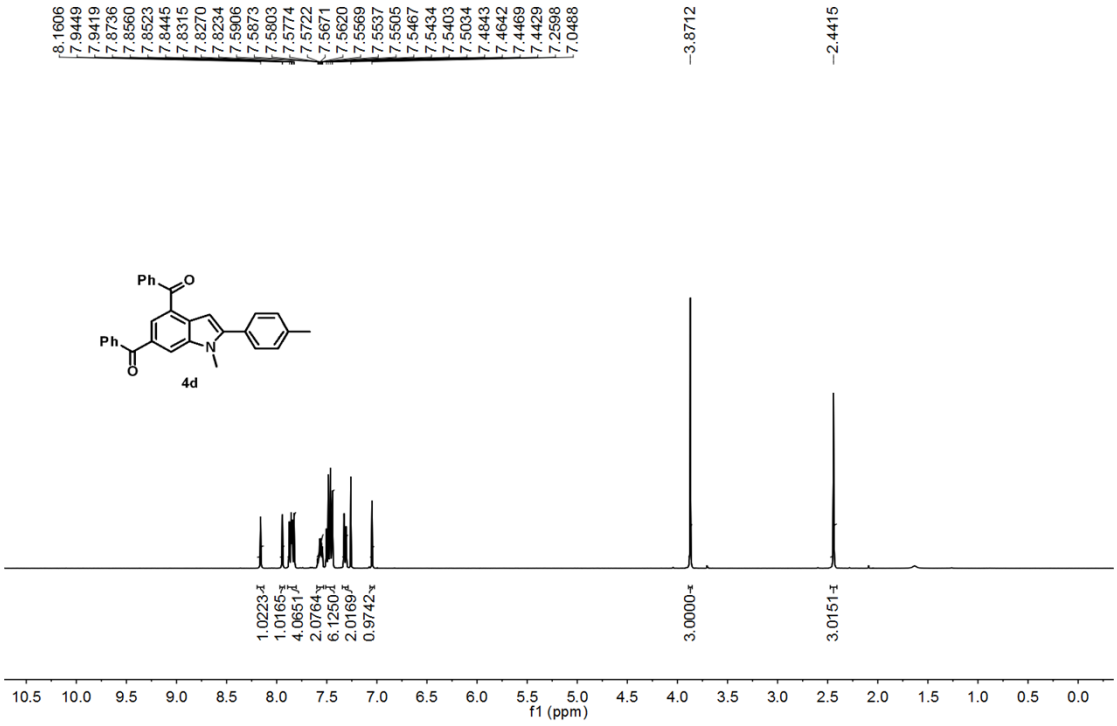

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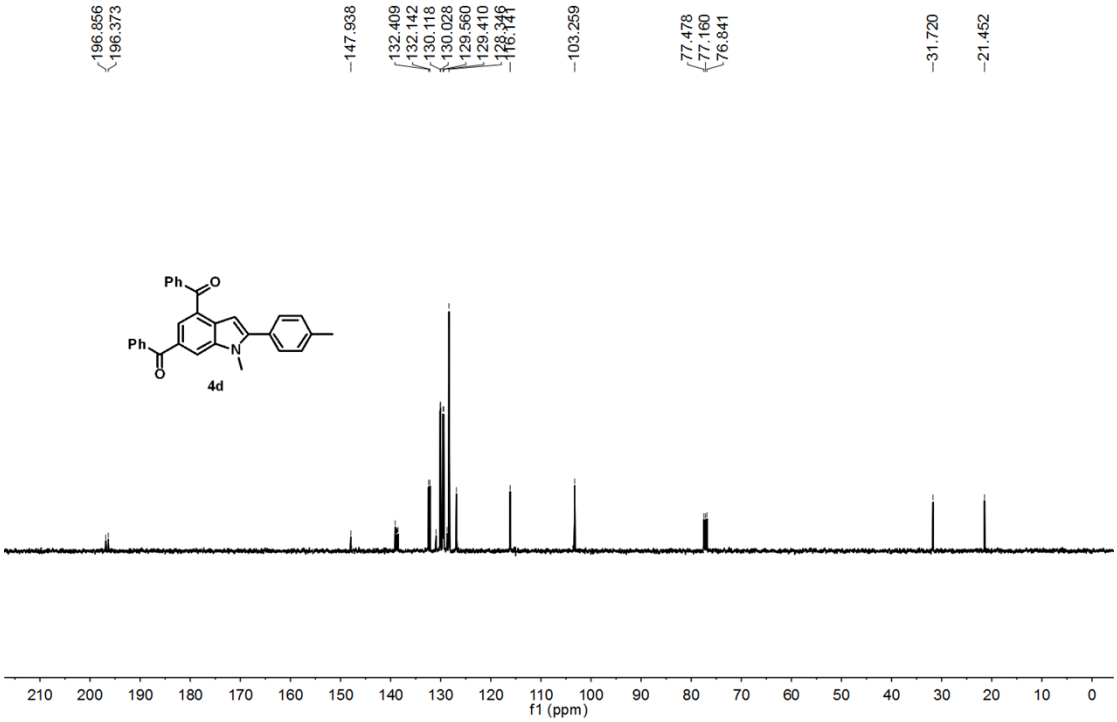

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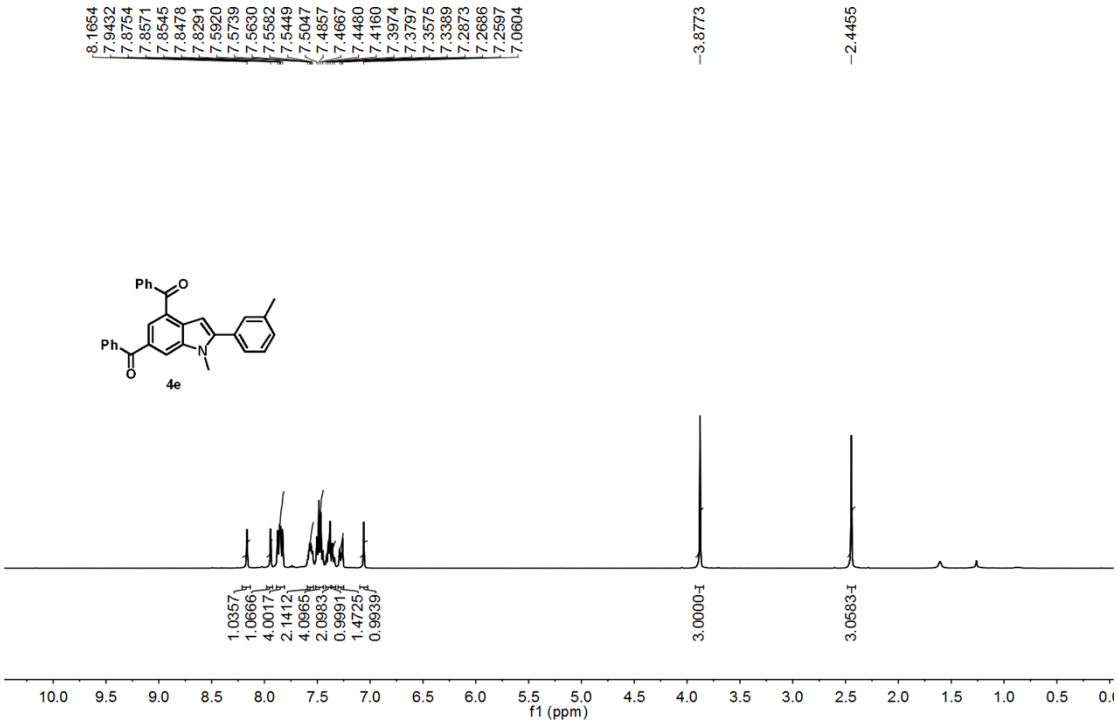

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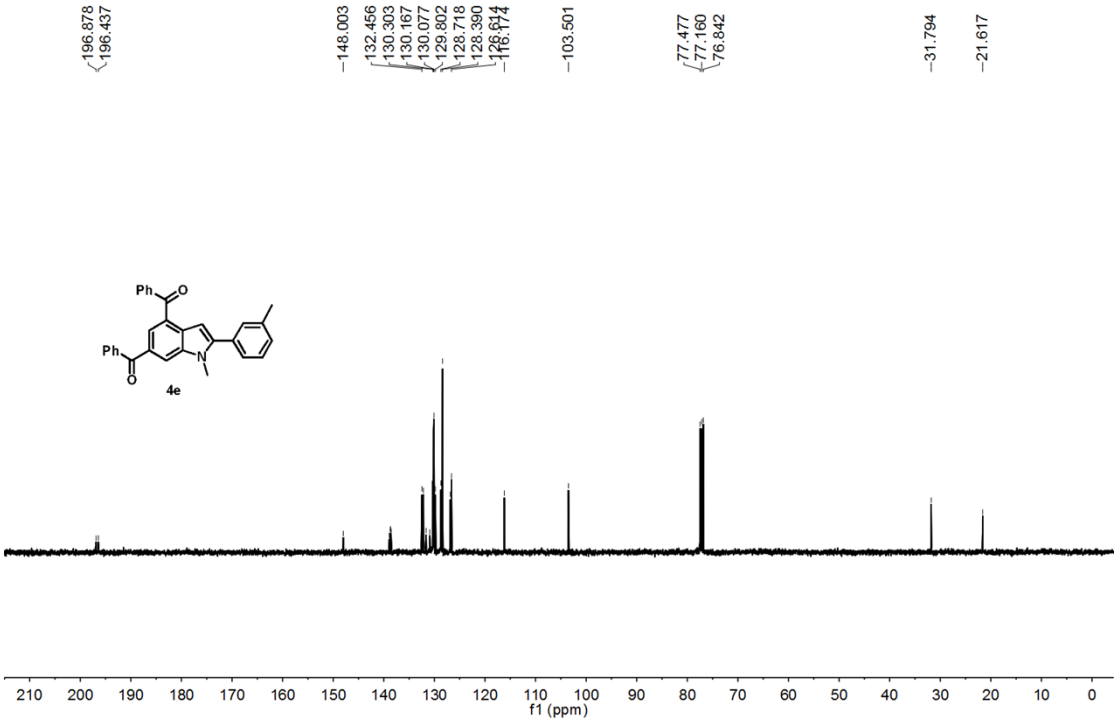

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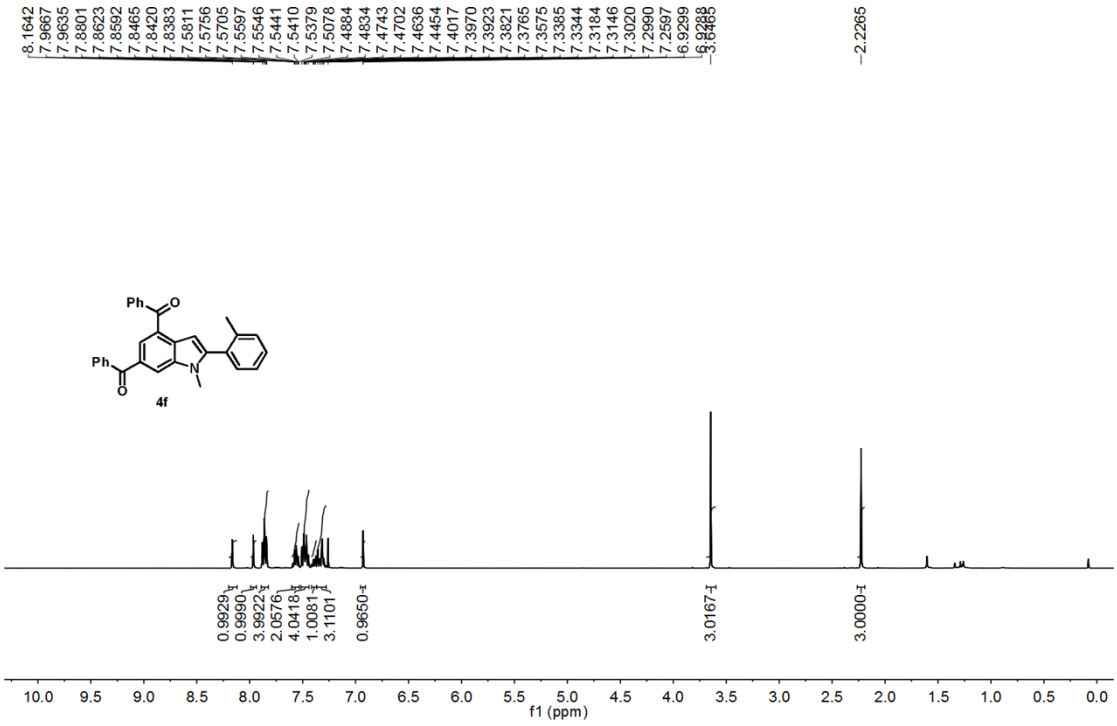

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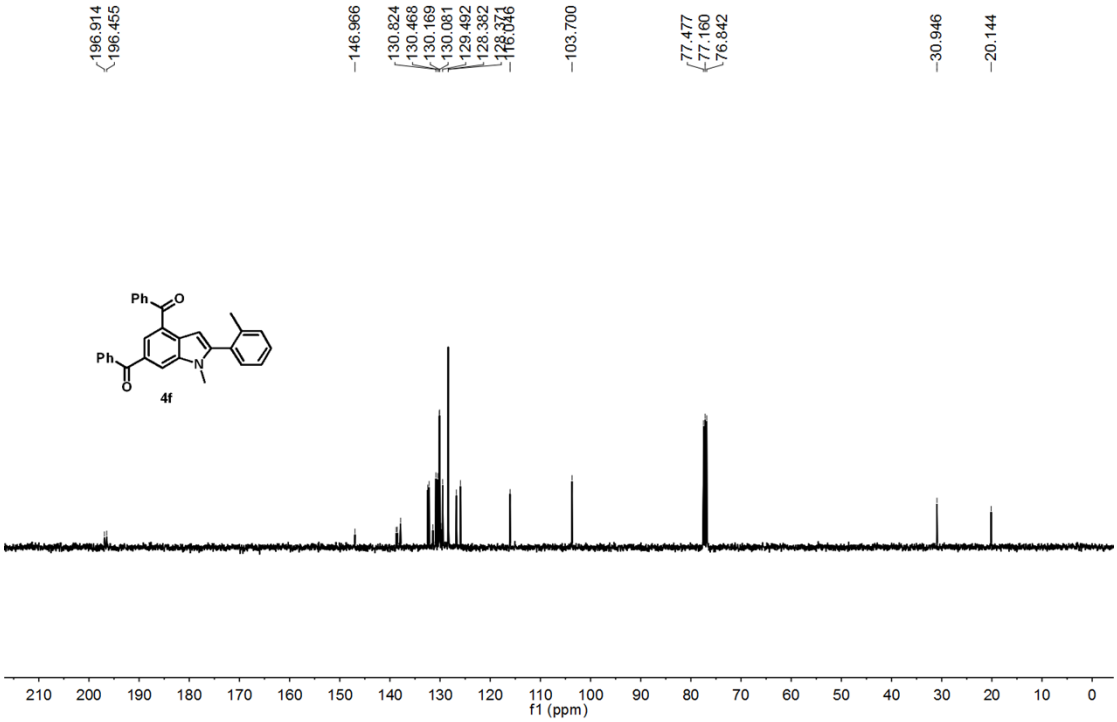
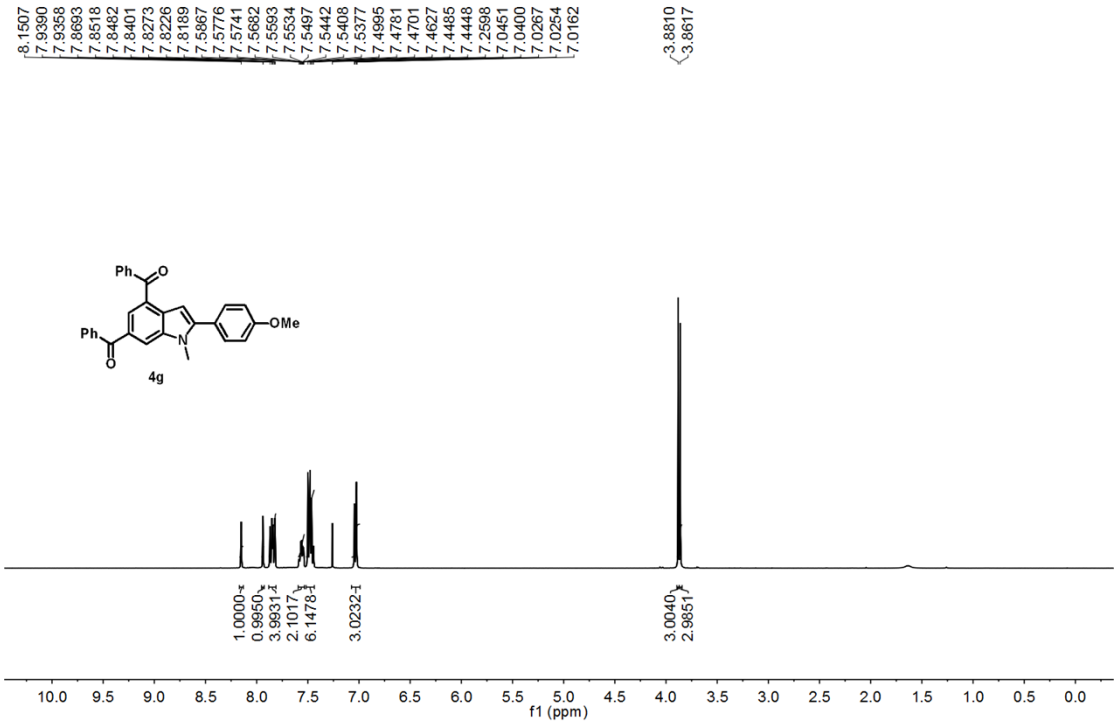


gtl-38300-1
1H NMR gtl-38300 in CDCl3



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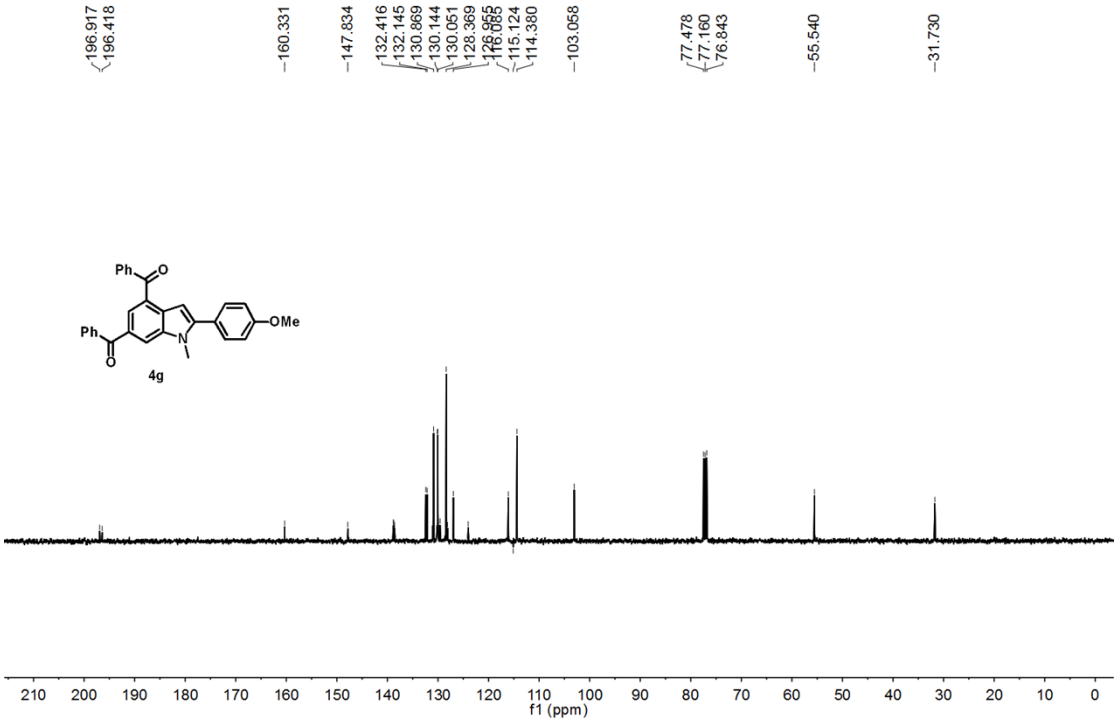
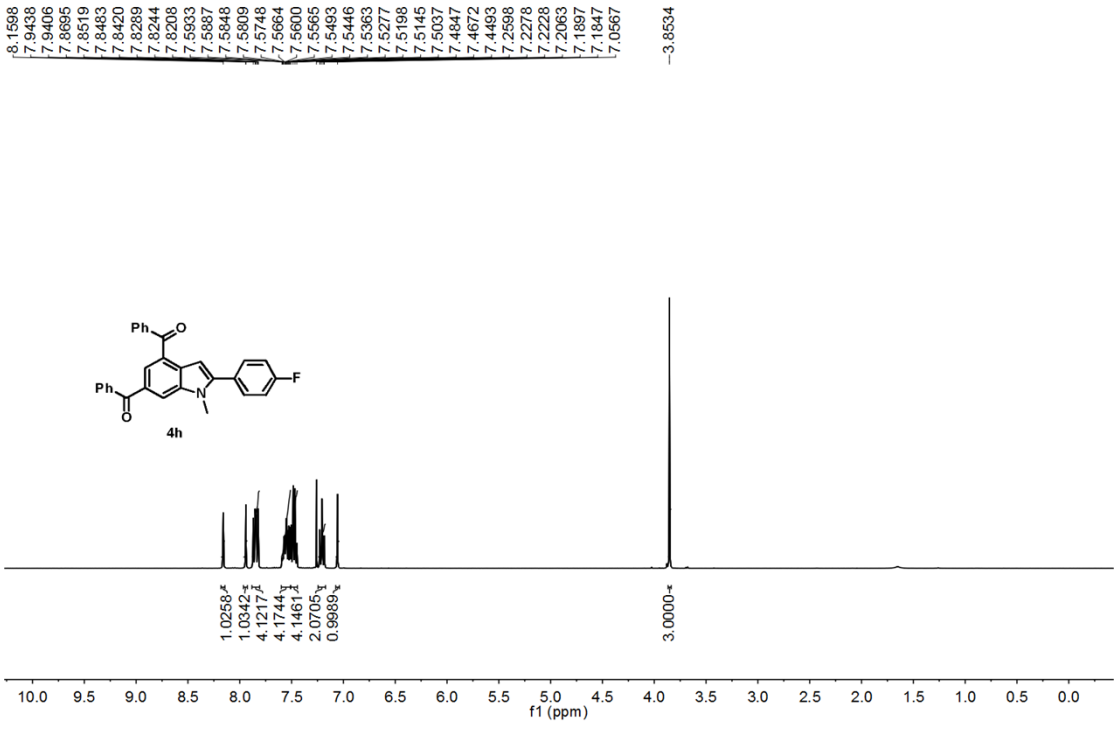
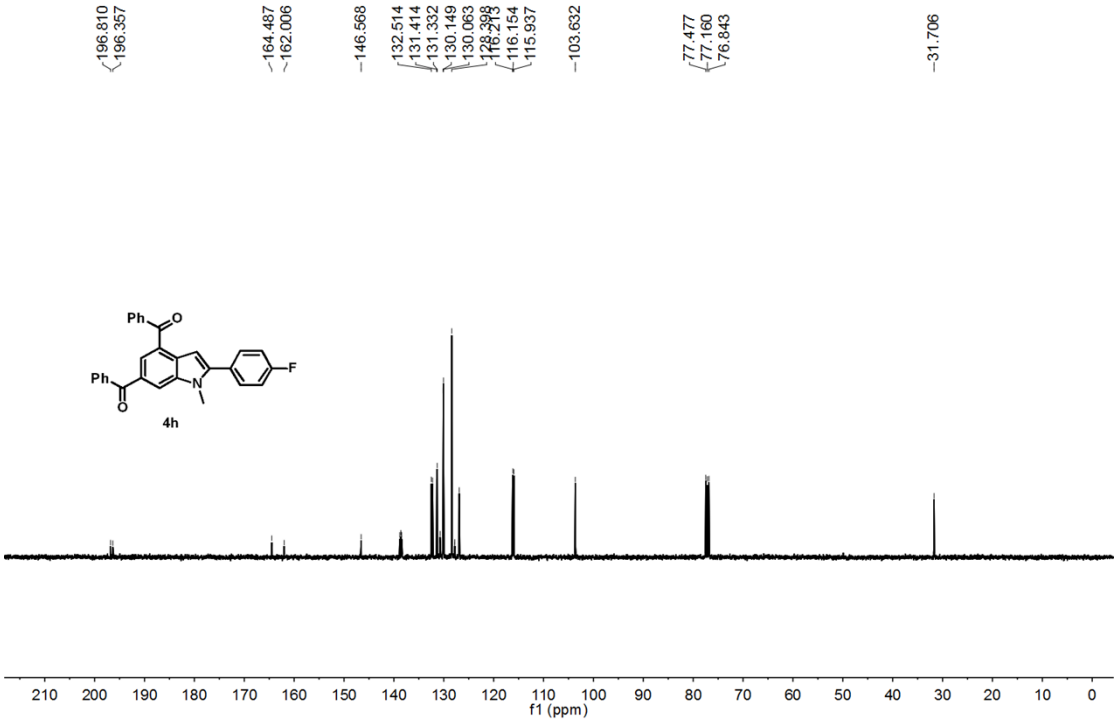


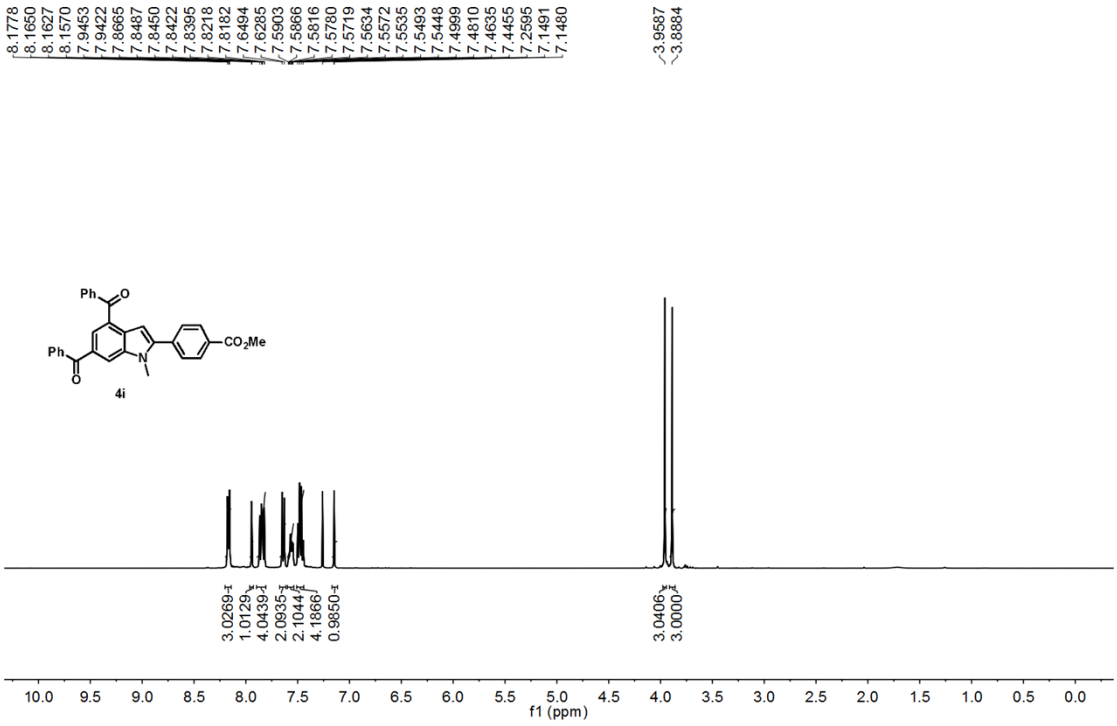
gtl-37400
1H NMR gtl-37400 in CDCl3



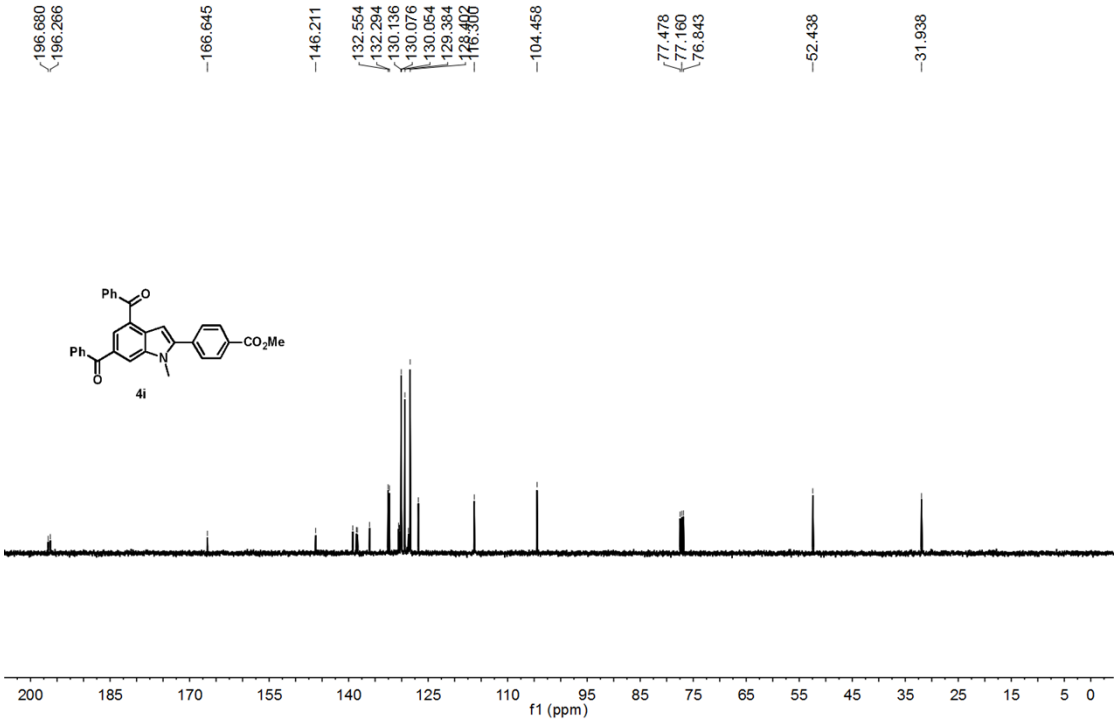
gtl-37400
13C NMR gtl-37400 CDCl3



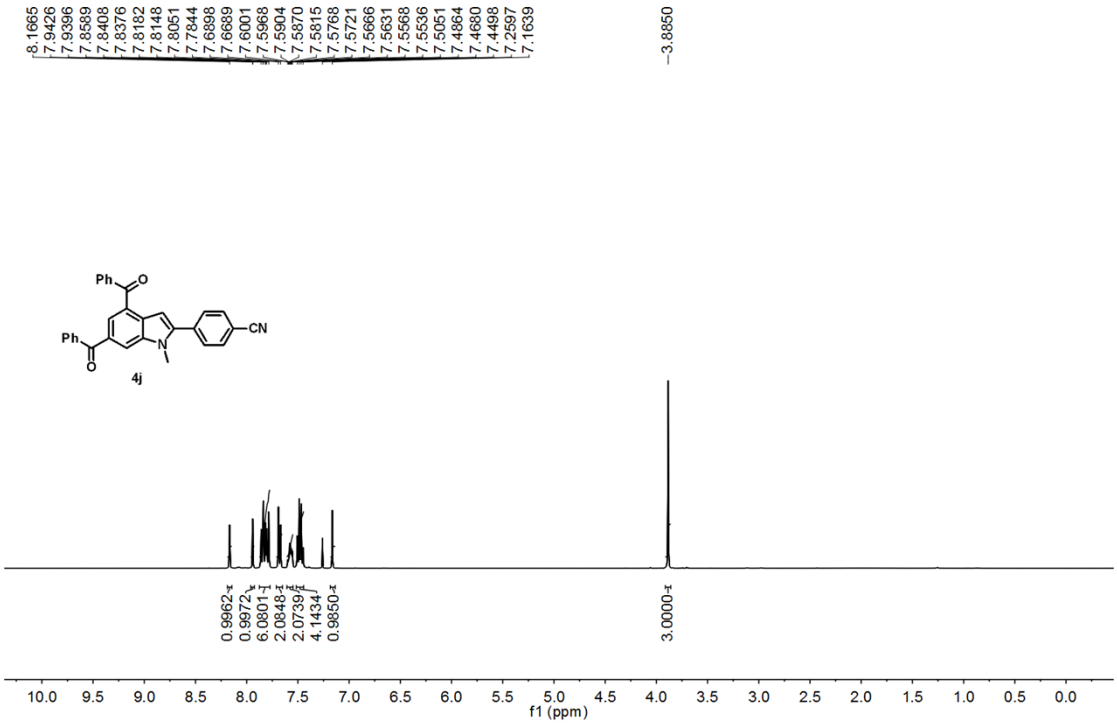
gtl-41200
1H NMR gtl-41500 in CDCl3



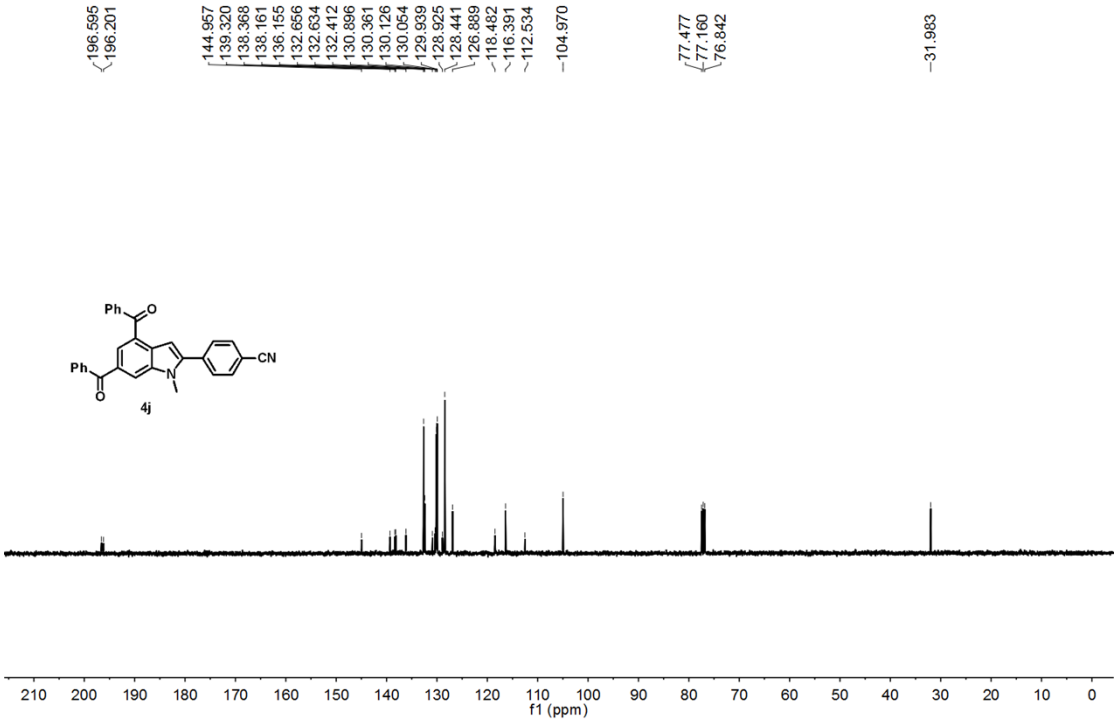
gtl-41200
13C NMR gtl-41500 CDCl3



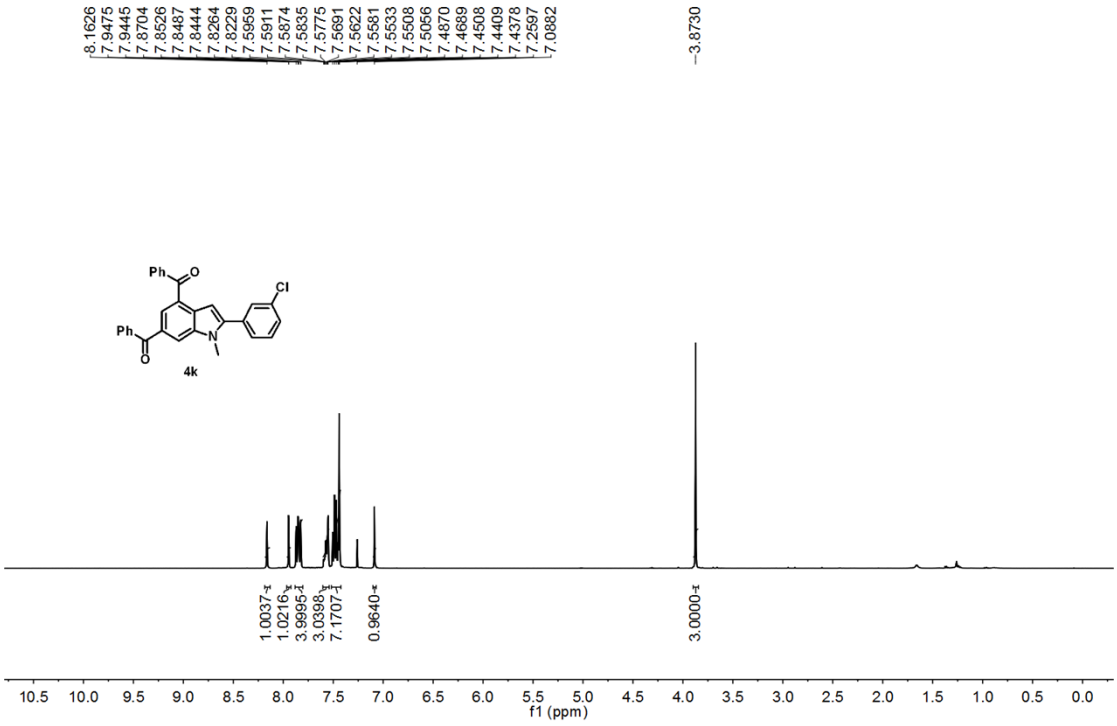
gtl-40900
1H NMR gtl-40900 in CDCl3



gtl-40900
13C NMR gtl-40900 CDCl3



gtl-37700
1H NMR gtl-37700 in CDCl3



gtl-37700
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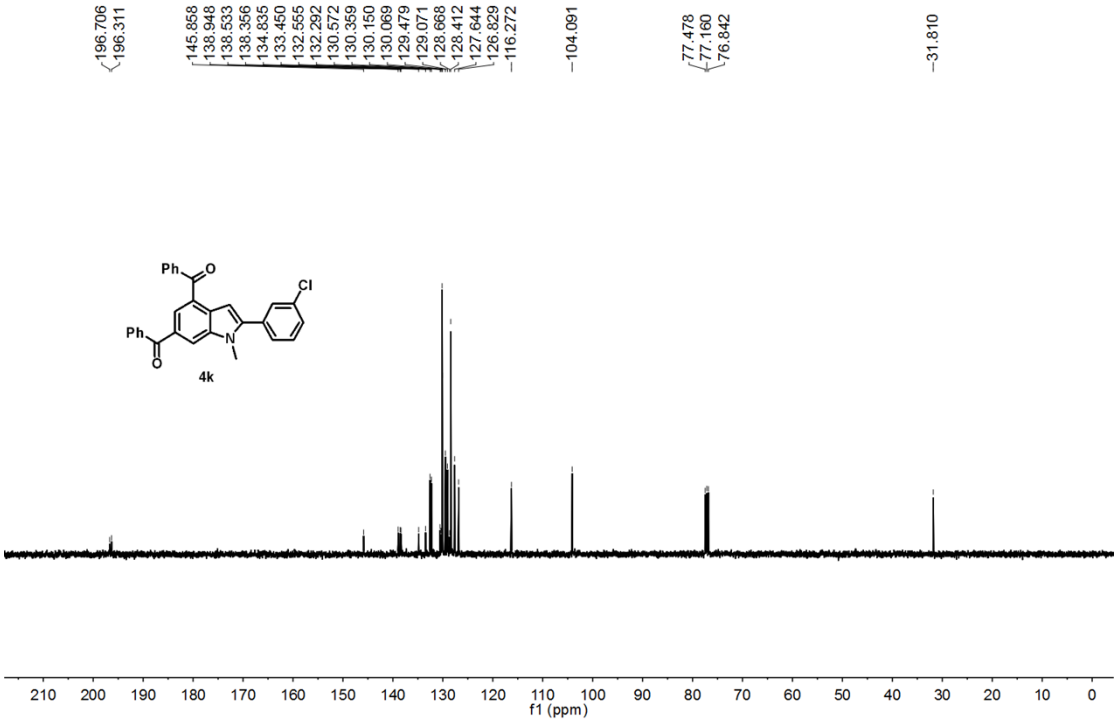
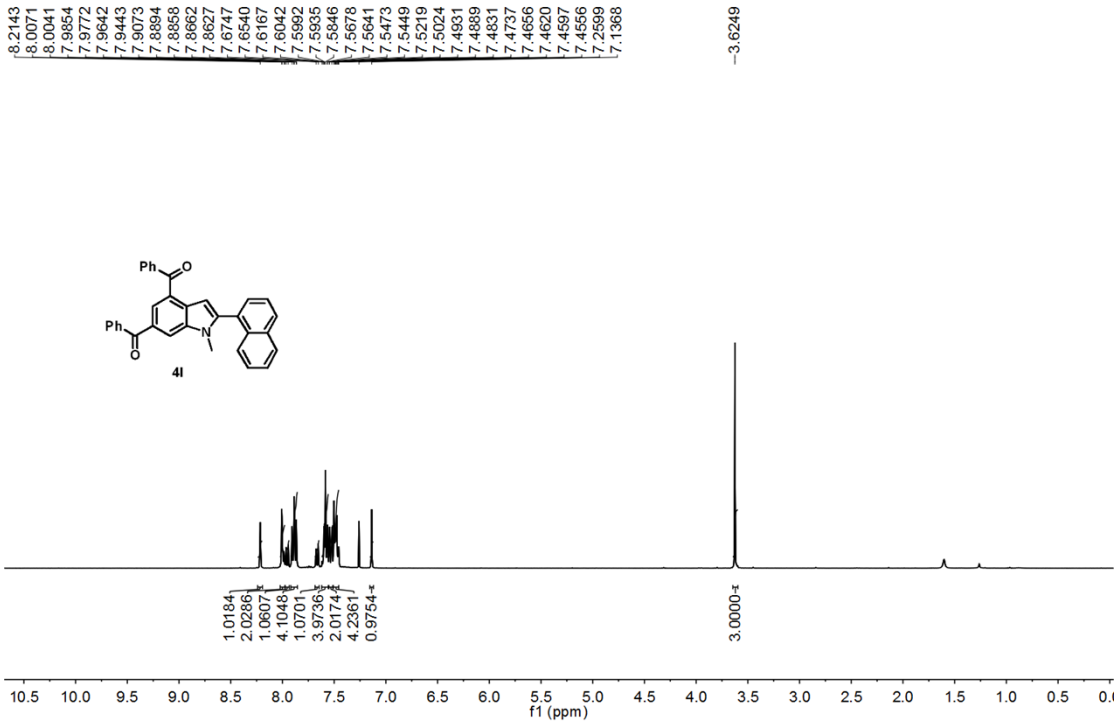
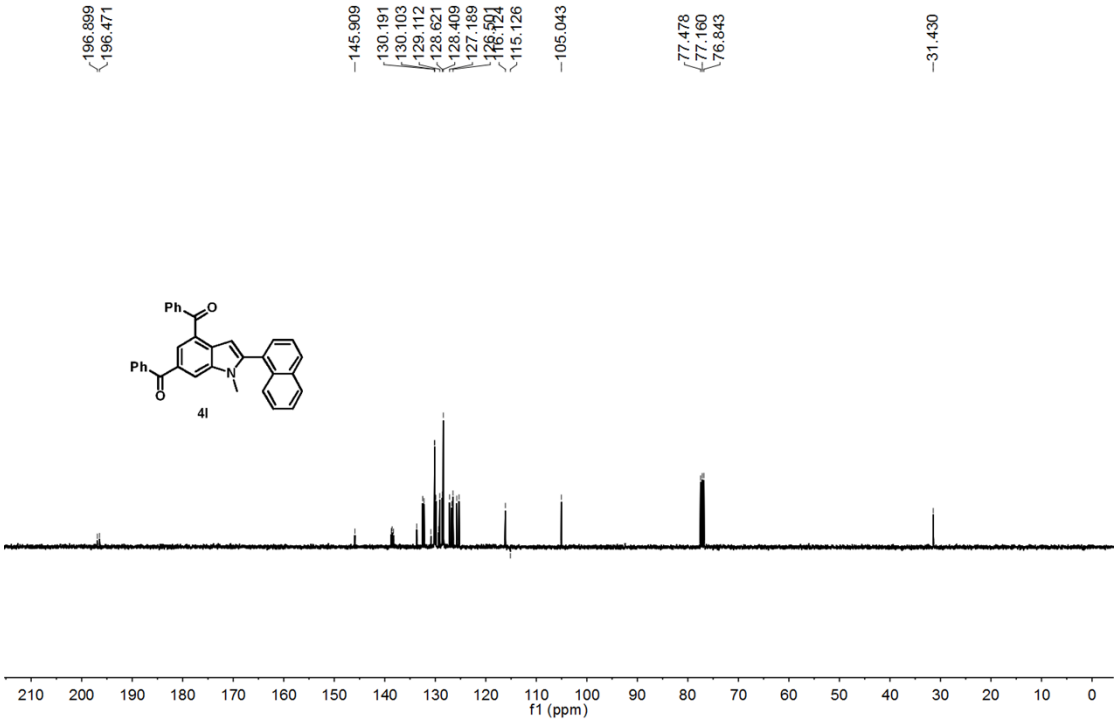


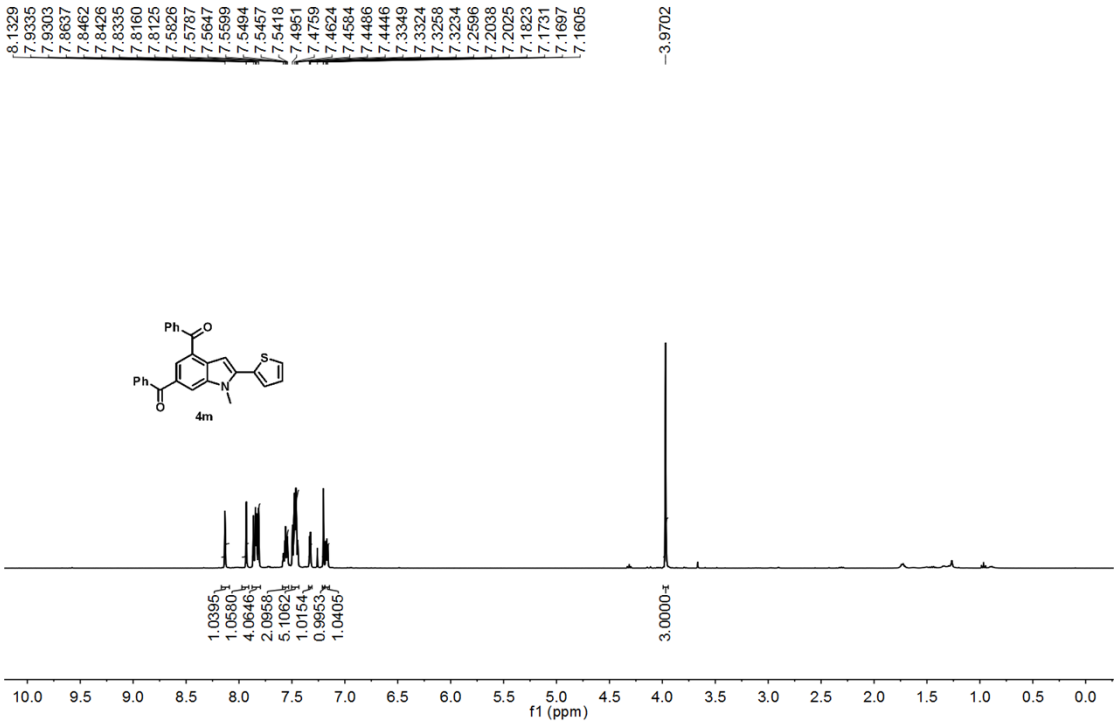
gtl-401-1
1H NMR gtl-401-1 in CDCl3



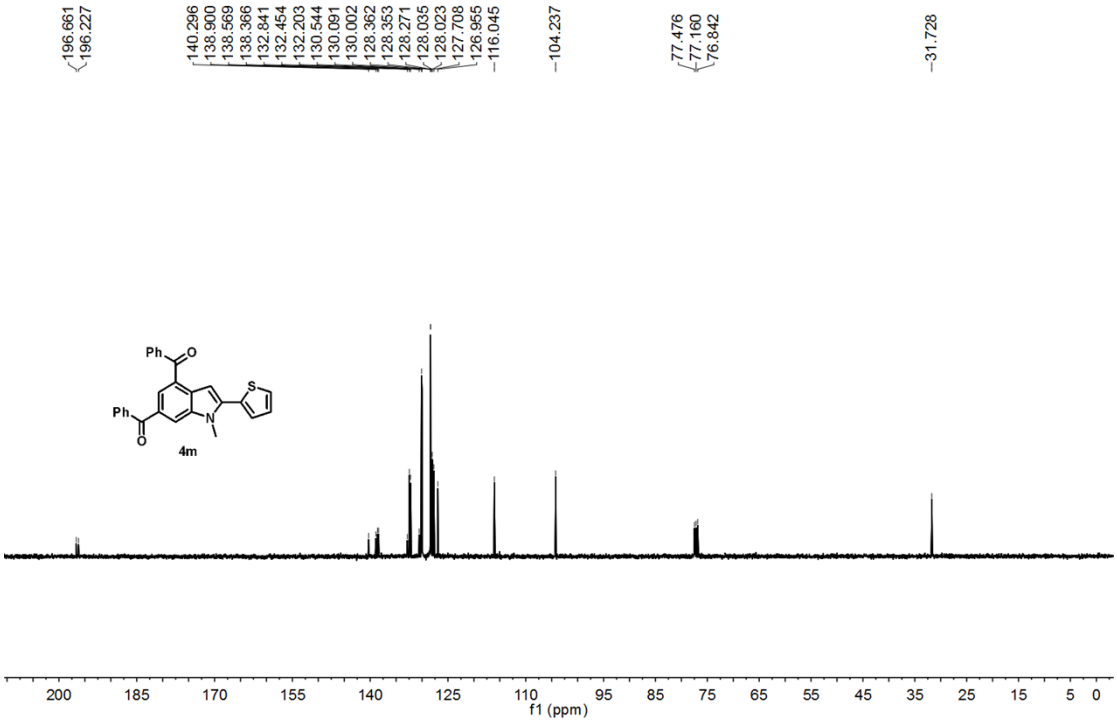
gtl-40100
13C NMR gtl-40100 CDCl3



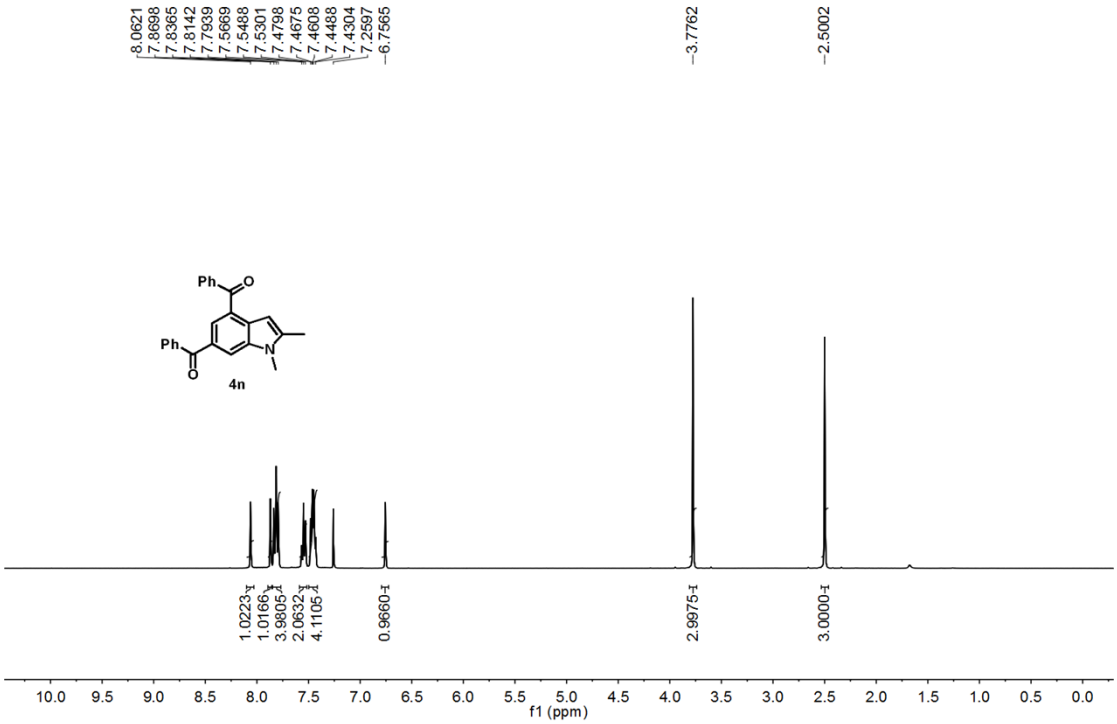
gtl-41600
1H NMR gtl-41600 in CDCl3



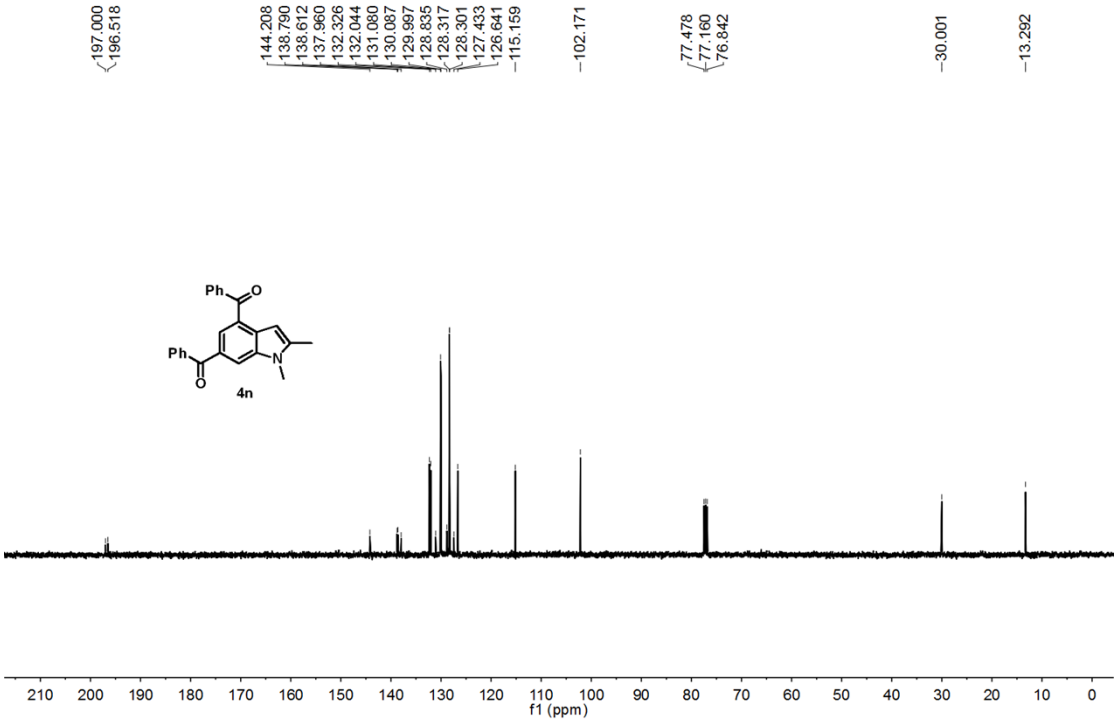
gtl-41600
13C NMR gtl-41600 CDCl3



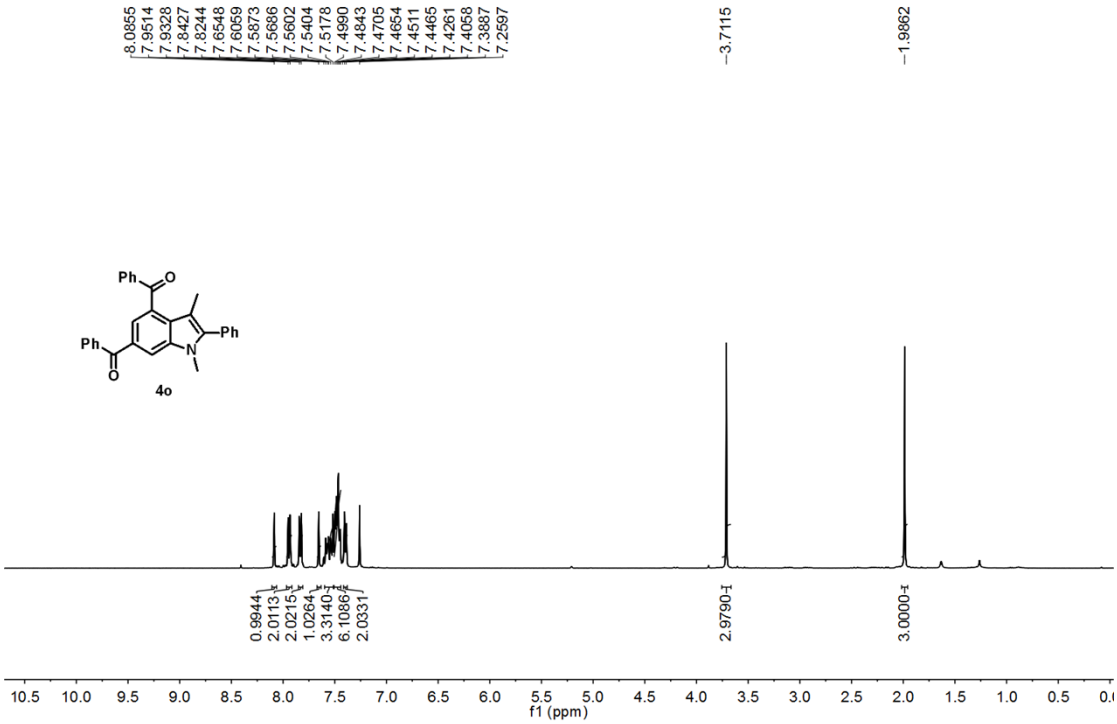
gtl-38100
1H NMR gtl-29000 in CDCl3



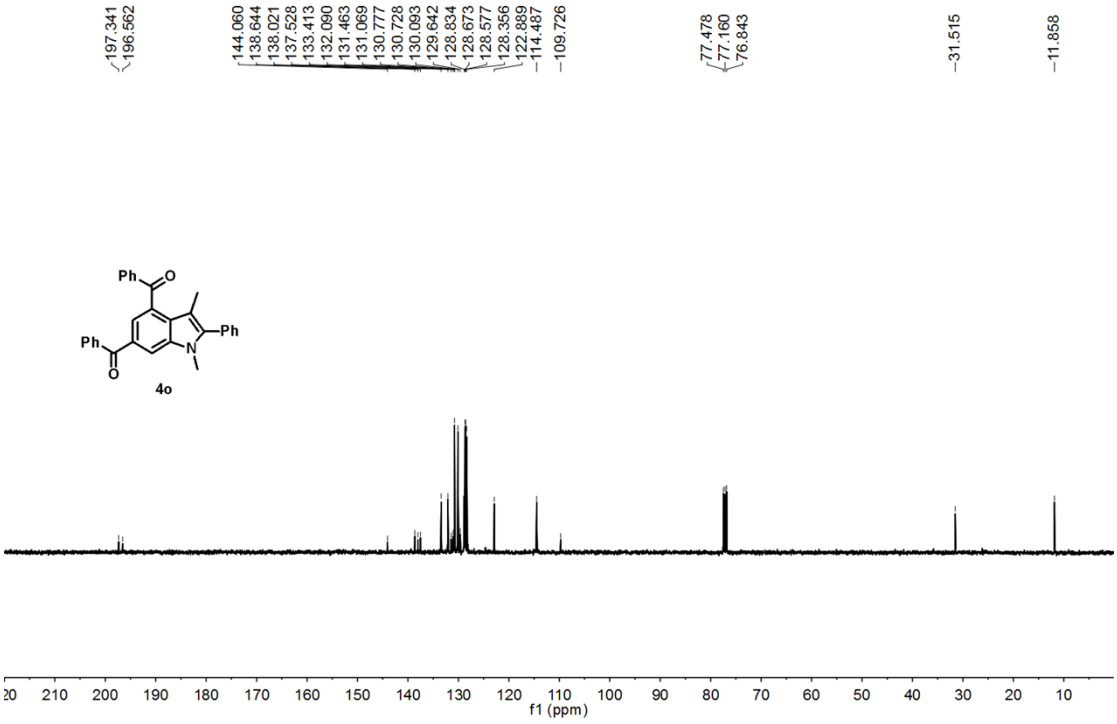
gtl-38100
13C NMR gtl-29000 CDCl3



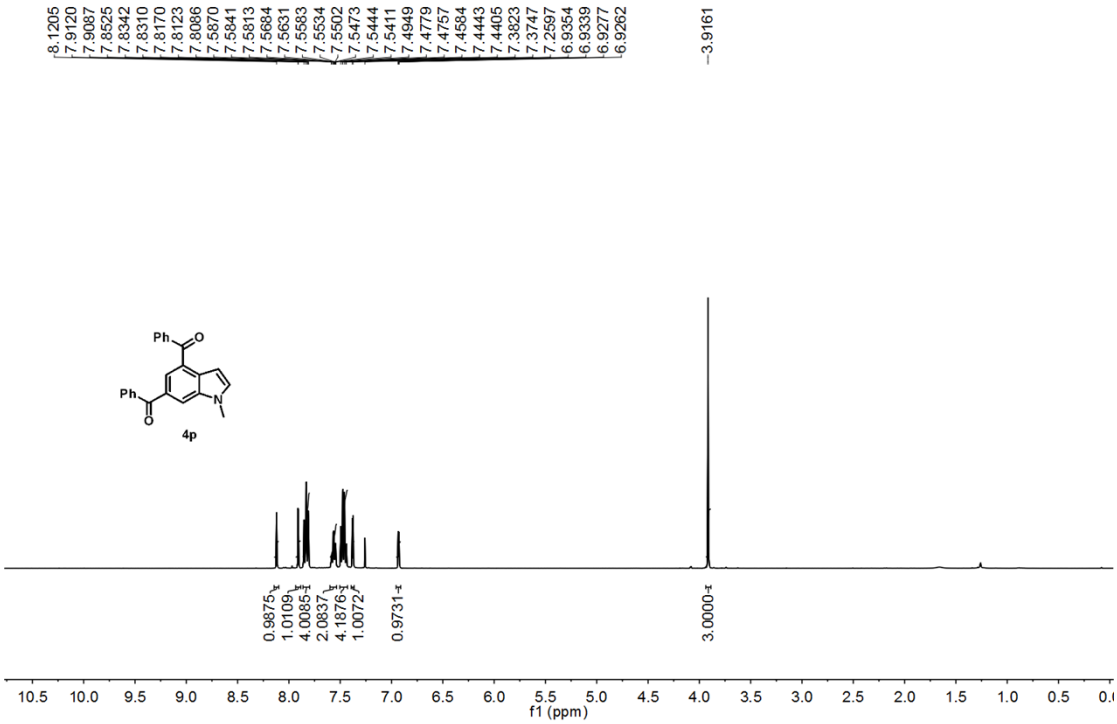
gtl-33200
1H NMR gtl-33200 in CDCl3



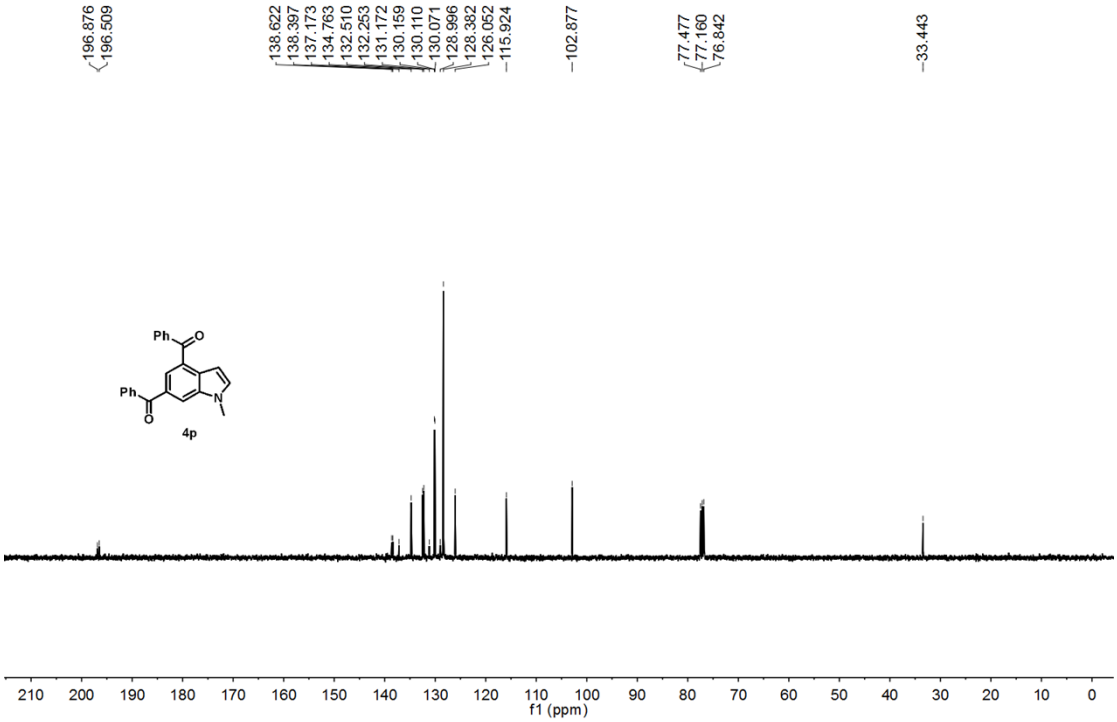
gtl-33200
13C NMR gtl-33200 CDCl3



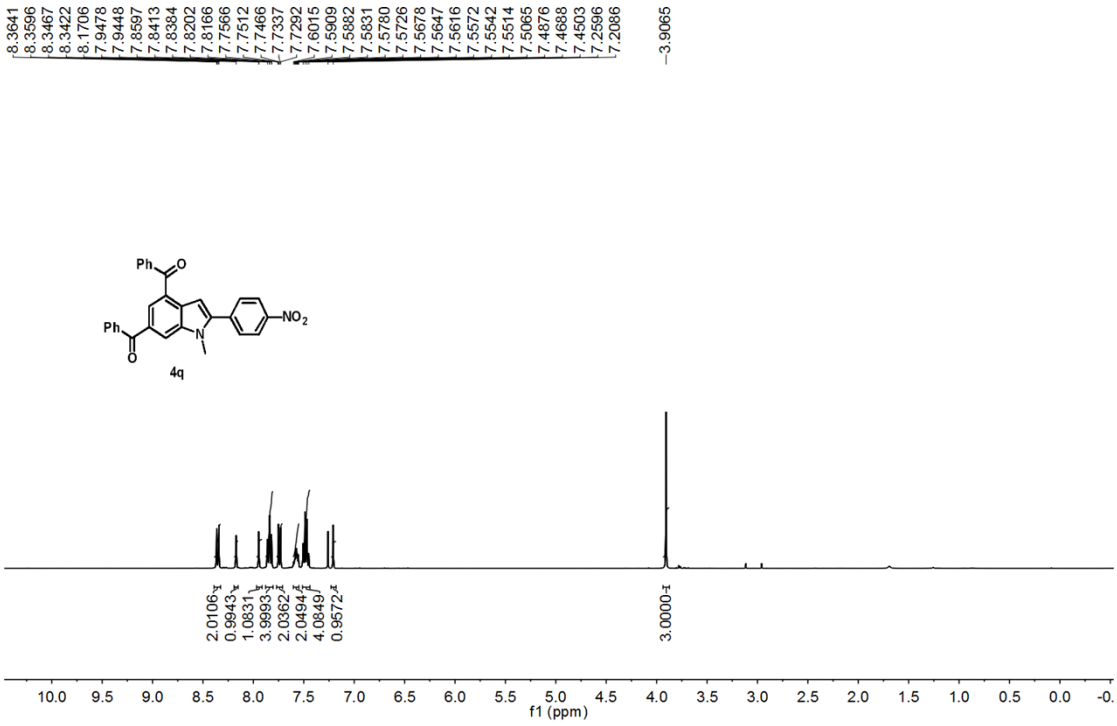
gtl-46100 in CDCl3



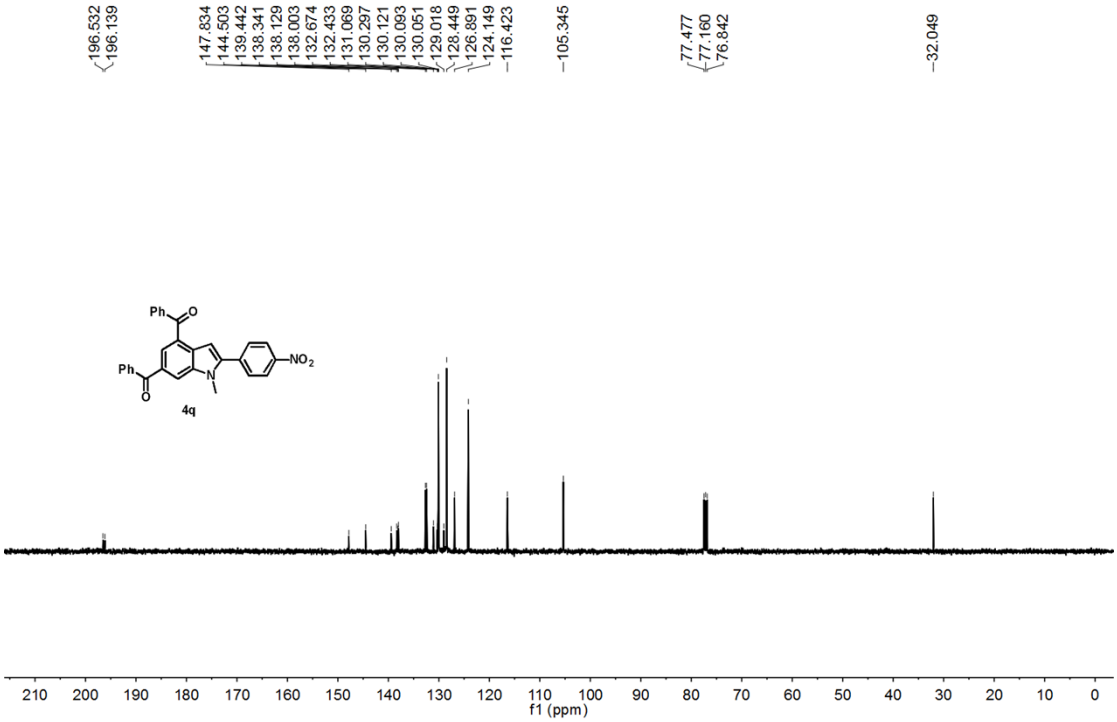
gtl-46100
13C NMR gtl-46100 CDCl3



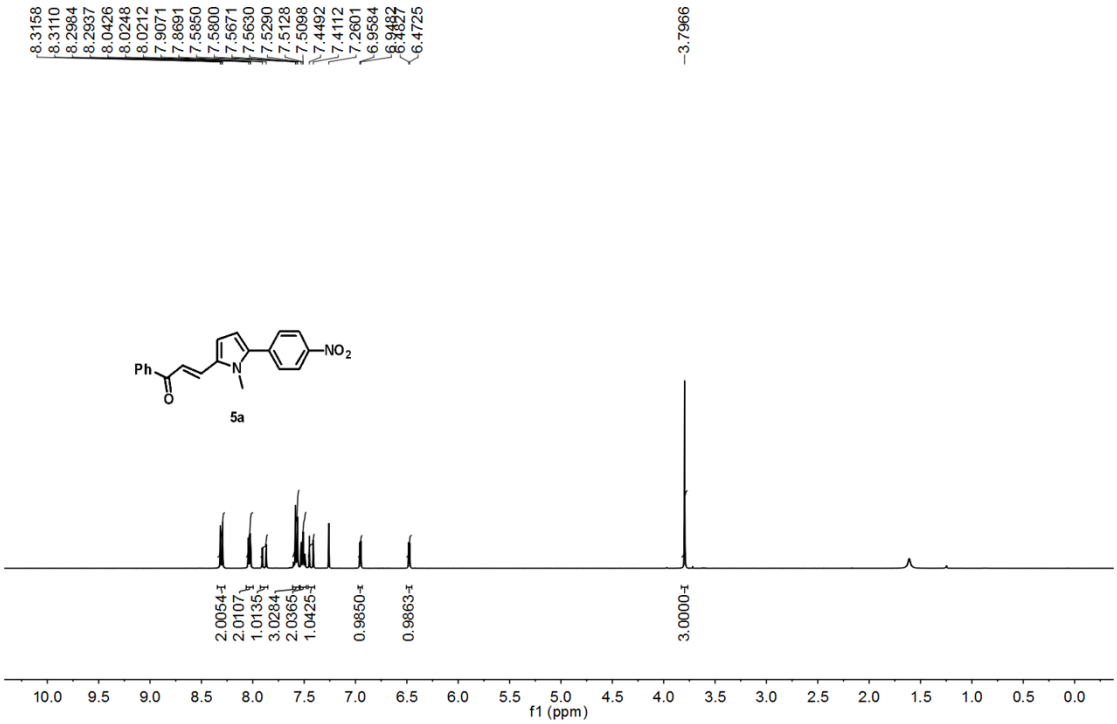
gtl-41000
1H NMR gtl-41000 in CDCl3



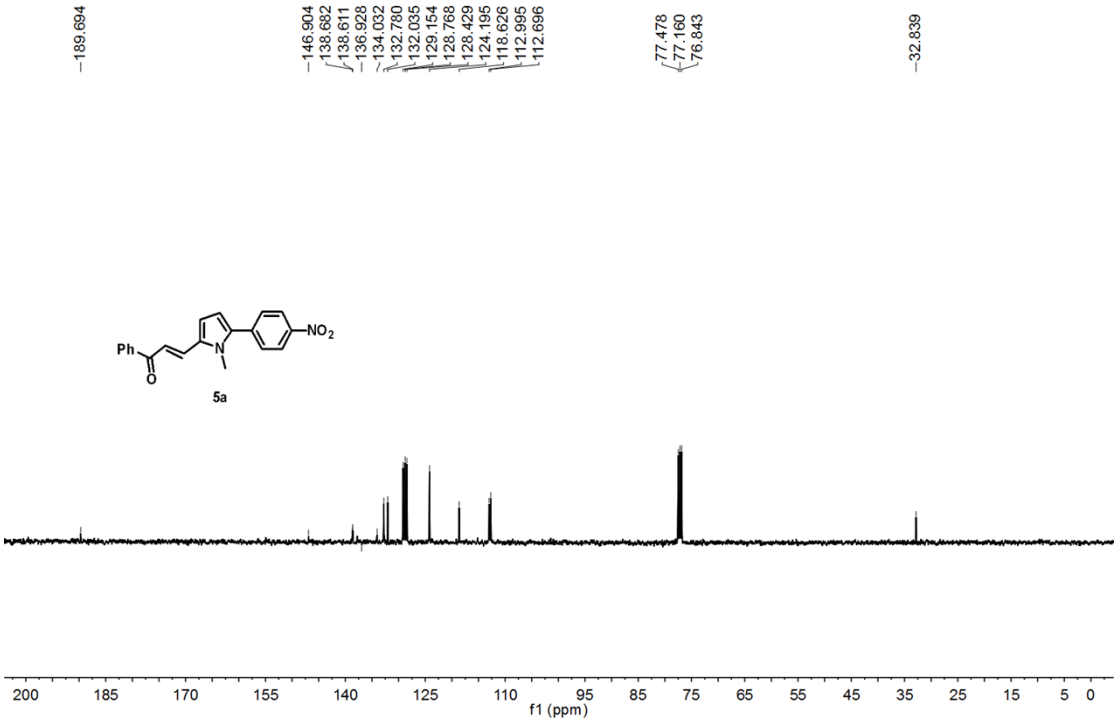
gtl-41000
13C NMR gtl-41000 CDCl3



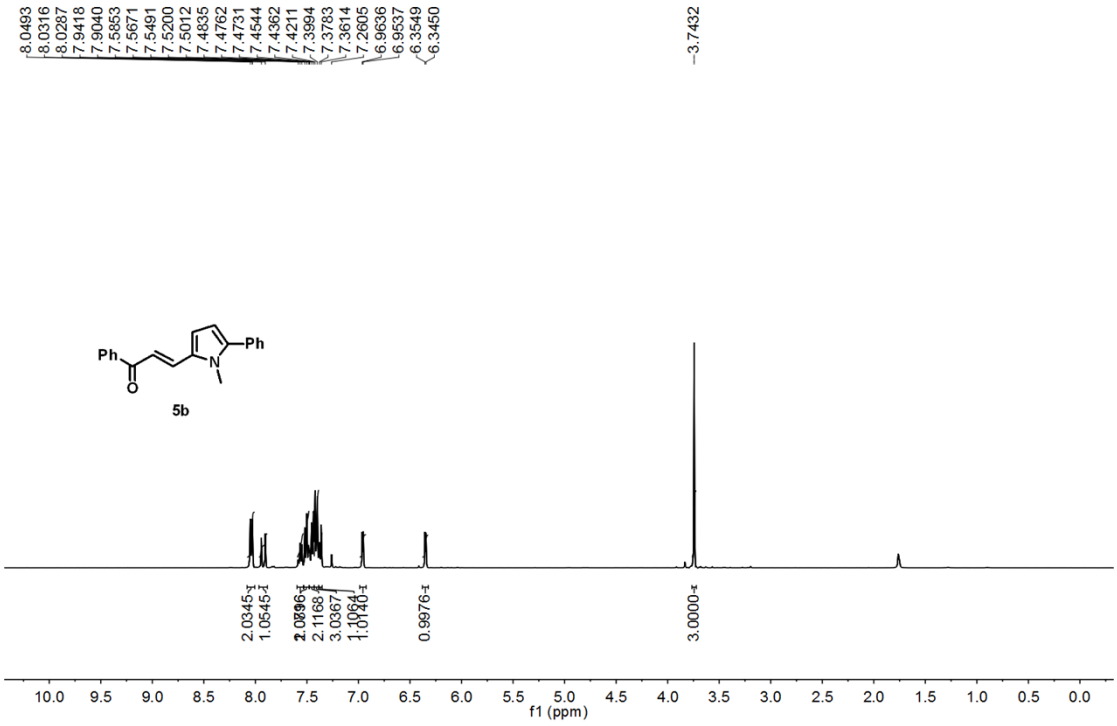
gtl-41001
1H NMR gtl-41001 in CDCl3



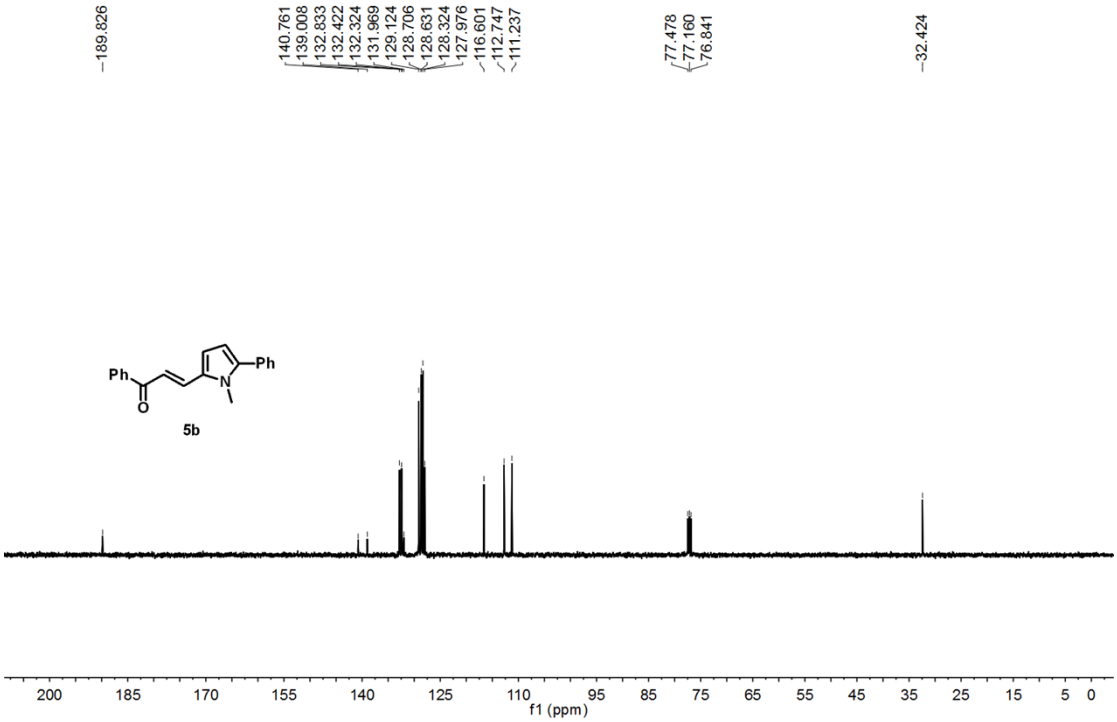
gtl-41001
13C NMR gtl-41001 CDCl3



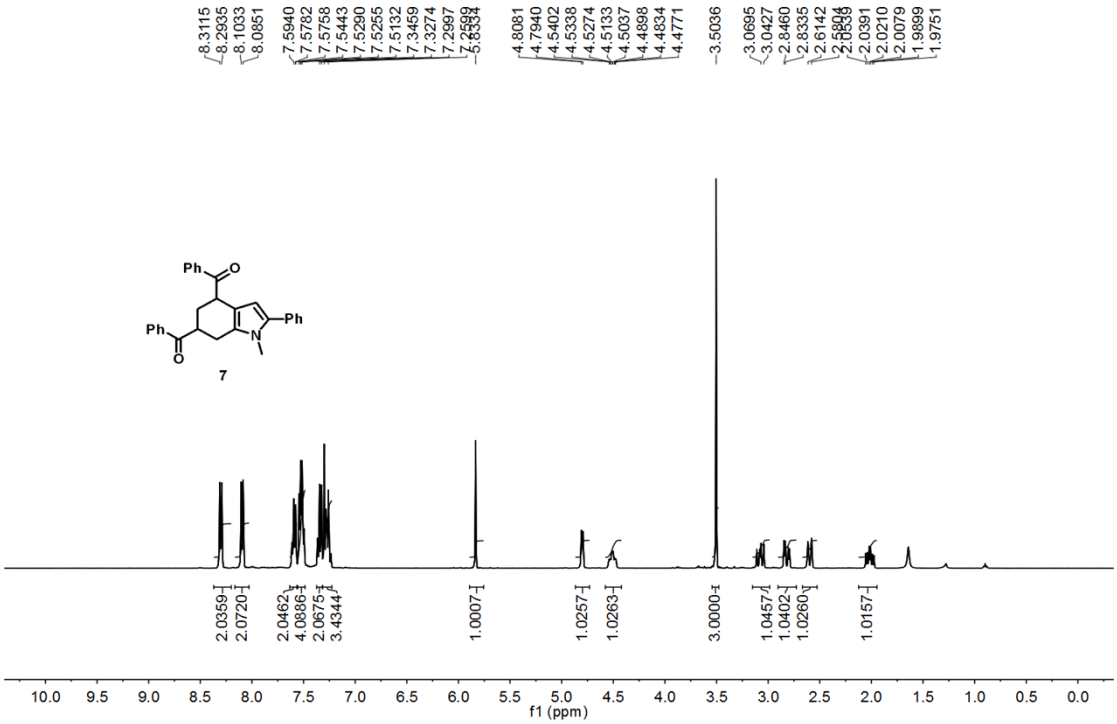
gtl-45404
1H NMR gtl-25404 in CDCl3



gtl-45404
13C NMR gtl-25404 CDCl3



gtl-46201
1H NMR gtl-46201 in CDCl3



gtl-46201
13C NMR gtl-46201 CDCl3