Supporting Information

Unravelling a Bench-Stable Zinc-amide Compound as a Highly Active Multitasking Catalyst for the Radical-Mediated Selective Alk(en)ylation of Unactivated Carbocycles Under Mild Conditions

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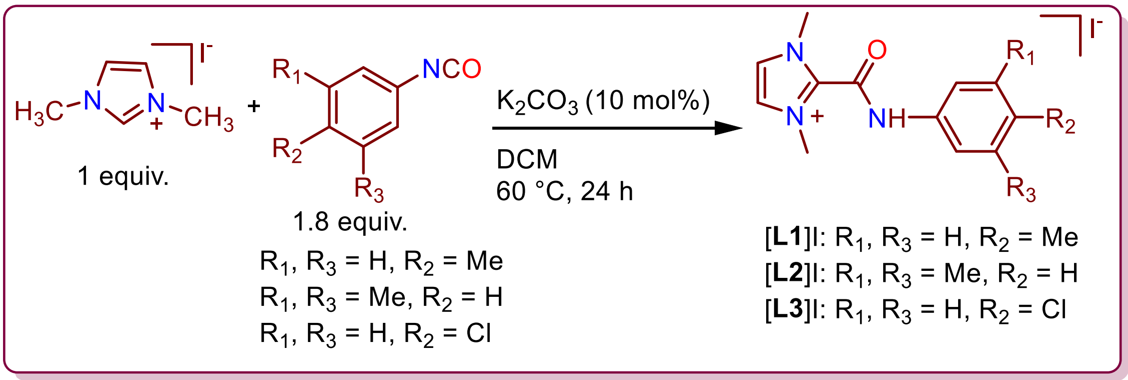
General experimental description:

All the reactions were performed under an argon atmosphere using a glove box and/or standard Schlenk techniques unless stated otherwise. All non-deuterated solvents used for the synthesis were distilled, degassed by standard methods and kept under an inert atmosphere over 4 Å molecular sieves, whereas the deuterated solvents were used as received from the commercial sources. NMR spectra were recorded using the Bruker 400 and 500 MHz FT-NMR spectrometers at ambient temperature and all the $^1H/^{13}C\{^1H\}$ NMR spectra were referenced internally to the residual solvent signals. ^{19}F NMR spectra were referenced externally to α,α,α -trifluorotoluene (0.05% $CDCl_3$, δ = -63.73 ppm). The ESI-MS spectra were measured with an Agilent 6545A Q-TOF Mass spectrometer. Infrared spectra were recorded on an ATR 4000 Series Spectrometer. UV-vis absorption spectra were recorded on JASCO V-650 spectrometer. Either Metrohm autolab potentiostat or galvanostat MAC90009 instrument was used for the electrochemical analysis. EPR spectra were measured using JES-FA200 ESR Spectrometer. Zinc salts (from TCI) and all other chemicals were purchased from commercial sources and used directly without further purification. Employed amidated azolium salts ([**L1-3**]I) were synthesized according to the literature procedures.[1]

1. General procedure for the synthesis of amidated imidazolium salts [L1-3]I:

The *N*,*N*-dimethylimidazolium salt and the amidated imidazolium salts were prepared using the reported procedure.[1] In a pressure tube (25 mL), *N*,*N*-dimethylimidazolium salt (0.373 mmol, 1 equiv.), isocyanate (0.671 mmol, 1.8 equiv.), and K_2CO_3 (0.1 equiv.) were stirred in DCM (2 mL) at 60 °C (oil bath temperature) for 24 h. After completion of the reaction, the reaction mixture was purified by column chromatography using methanol and DCM solvent mixture (1:10) as eluent to provide the desired C2-amidated imidazolium salts.[1]

Scheme S1: Synthesis and characterization of amidated imidazolium salts [L1-3]I



[**L1**]I: [**L1**]I was synthesized according to the general procedure[1] using 100 mg of imidazolium salt and 107.0 mg of *p*-tolyl-isocyanate (yield: 140 mg, 0.391 mmol, 87%). ^{1}H NMR (400 MHz, DMSO-d_6) δ 11.30 (s, 1H), 7.92 (s, 2H), 7.59 (d, J = 10.5 Hz, 2H), 7.25 (d, J = 8.4 Hz, 2H), 3.98 (s, 6H), 2.30 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 150.7, 138.0, 135.0, 134.3, 129.5, 123.9, 120.4, 36.4, 20.6 ppm. IR (KBr): 3428, 1683, and 1530 cm^{-1}.

[**L2**]I**:** [**L2**]I was synthesized according to the general procedure[1] using 100 mg of imidazolium salt and 118.3 mg of 3,5-dimethyl phenyl isocyanate (yield: 138 mg, 0.371 mmol, 83%). ^{1}H NMR (400 MHz, DMSO-d_6) δ 11.22 (s, 1H), 7.89 (s, 2H), 7.32 (s, 2H), 6.91 (s, 1H), 3.95 (s, 6H), 2.29 (s, 6H) ppm. ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 150.8, 138.4, 138.1, 136.7, 127.2, 123.9, 118.1, 36.3, 21.0 ppm. IR (KBr): 3451, 1685, and 1528 cm^{-1}.

[**L3**]I**:** [**L3**]I was synthesized according to the general procedure[1] using 100 mg of imidazolium salt and 102.7 mg of 4-chlorophenyl isocyanate (yield: 137 mg, 0.363 mmol, 81%). ^{1}H NMR (400 MHz, DMSO-d_6) δ 11.49 (s, 1H), 7.89 (s, 2H), 7.72 (d, J = 8.9 Hz, 2H), 7.52 (d, J = 8.9 Hz, 2H), 3.98 (s, 6H) ppm. ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 151.1, 137.7, 135.9, 129.4, 129.1, 124.1 122.2, 36.4 ppm. IR (KBr): 3427, 1615, and 1531 cm^{-1}.

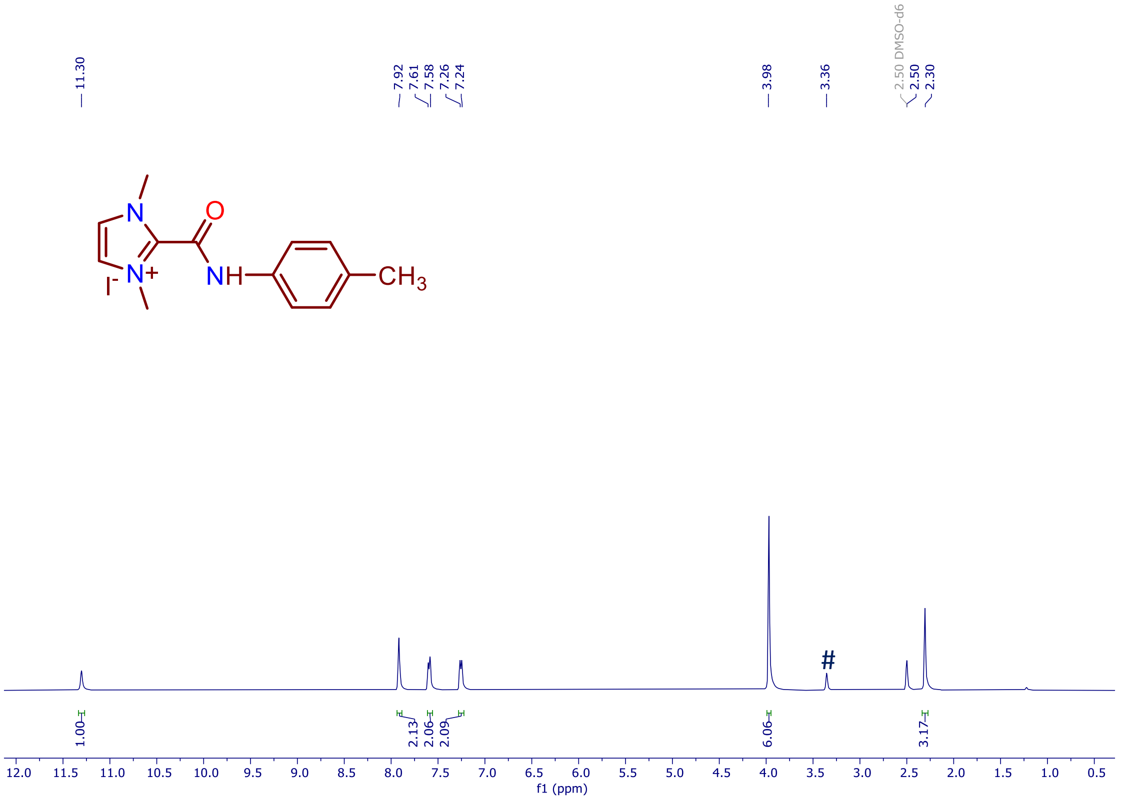


***Figure S1**. ^{1}H NMR of [**L1**]I in DMSO-d_6. # indicates the solvent impurity of H_2O in DMSO-d_6*

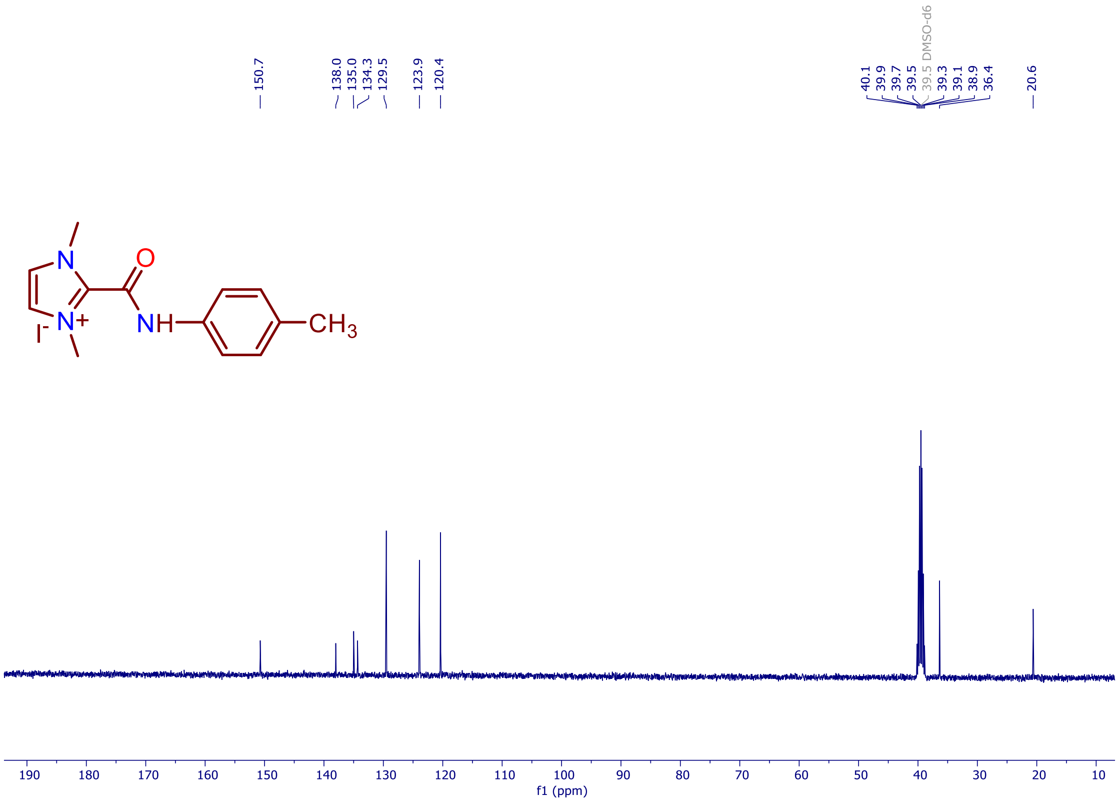


Figure S2. *$^{13}C\{^1H\}$ NMR of [**L1**]I in DMSO-d_6*

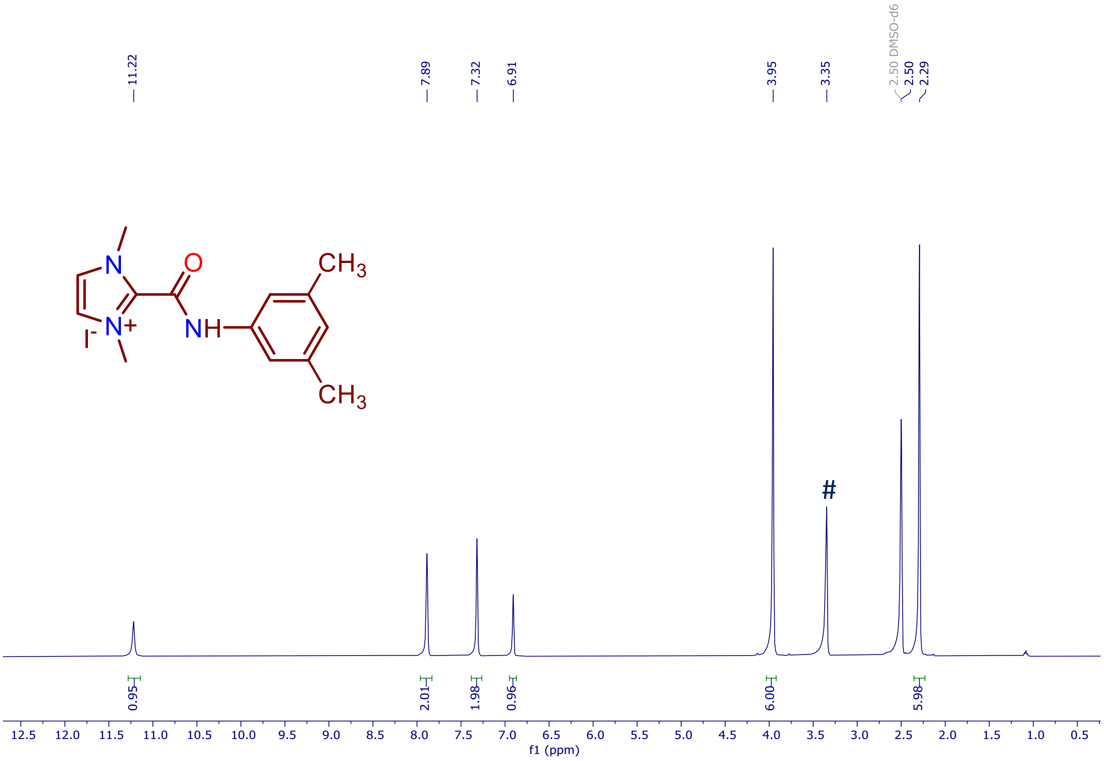


Figure S3. *^1H NMR of [**L2**]I in DMSO-d_6. # indicates the solvent impurity of H_2O in DMSO-d_6*

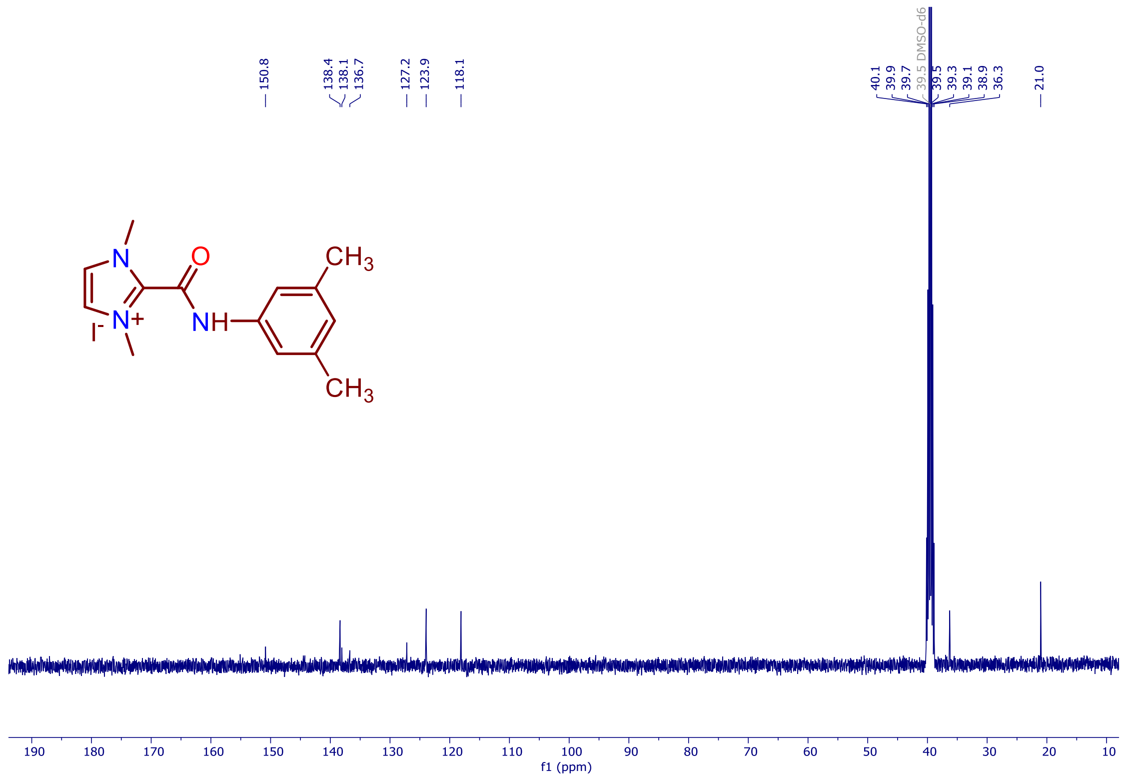


Figure S4. *$^{13}C\{^{1}H\}$ NMR of [**L2**]I in DMSO-d_6*

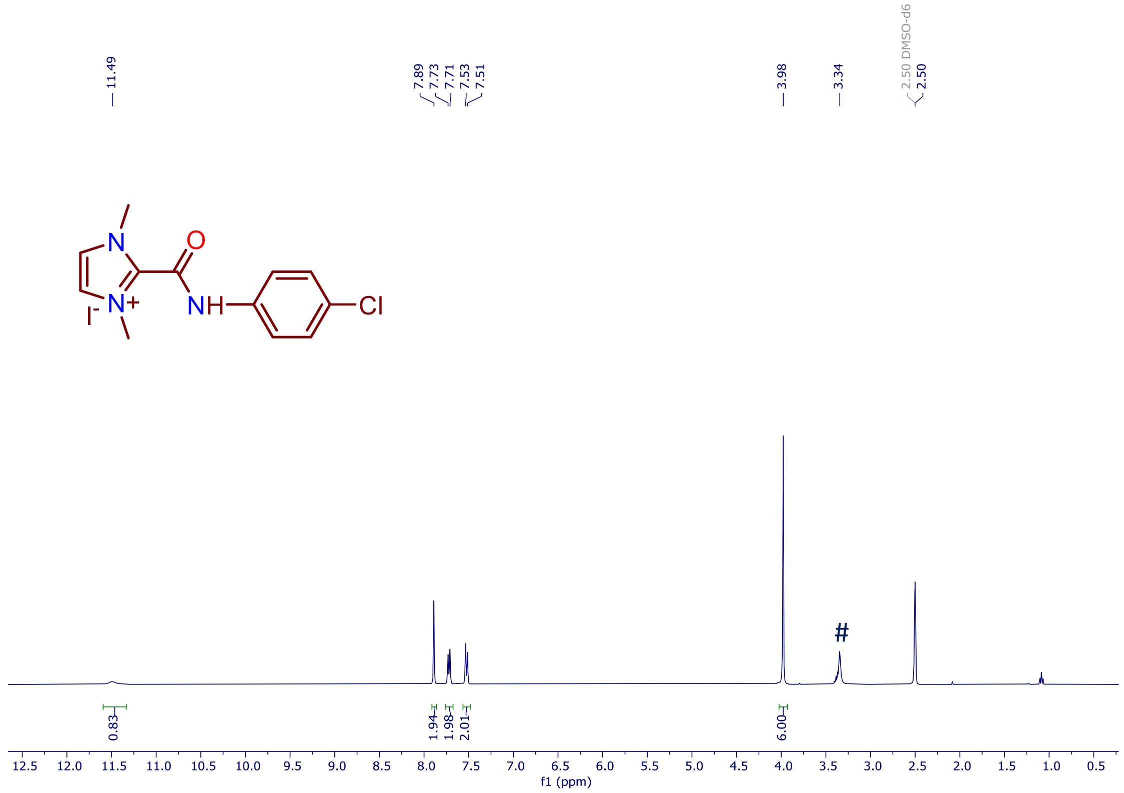


Figure S5. *^{1}H NMR of [**L3**]I in DMSO-d_6. # indicates the solvent impurity of H_2O in DMSO-d_6*

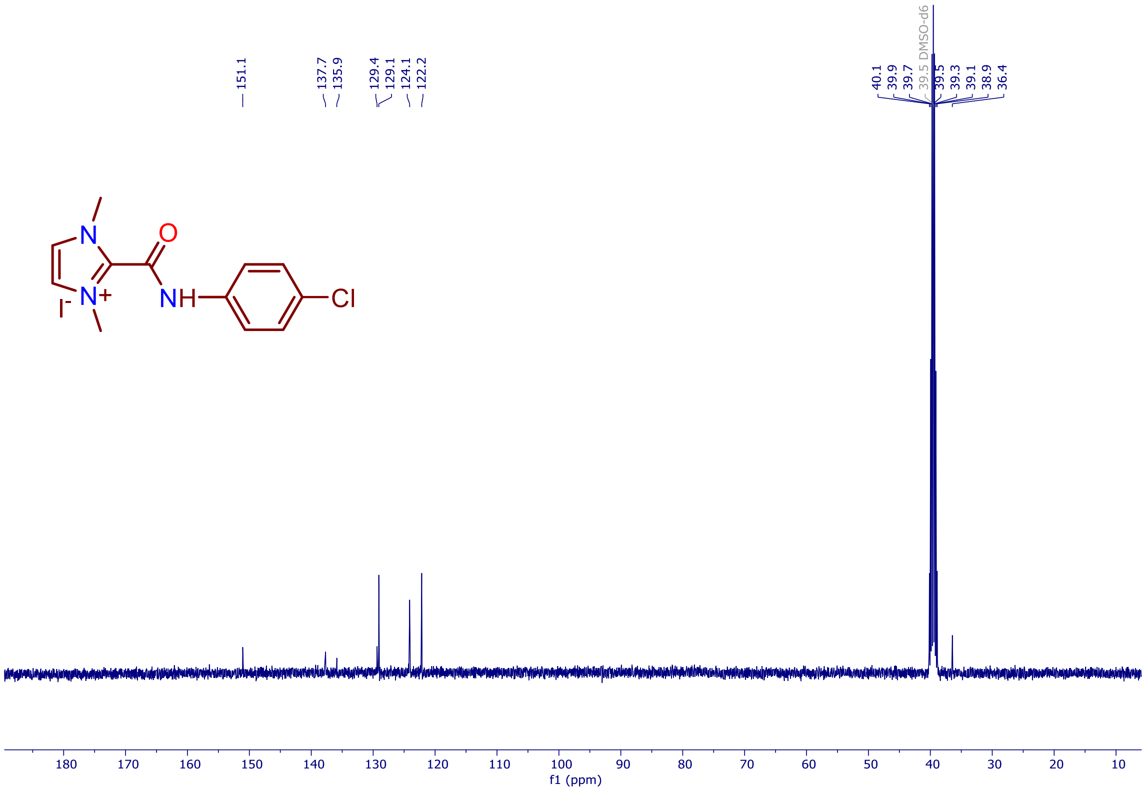


Figure S6. *^{13}C{^{1}H} NMR of [**L3**]I in DMSO-d_6*

2. General Procedure for the synthesis of compounds (Zn1-3):

The metal precursor $ZnCl_2$ (1 equiv.) and amidated imidazolium salts [**L1-3**]I (2 equiv.) were added to the Schlenk tube containing 2 mL of methanol with constant stirring at ambient temperature for 12 h. After that, all the volatiles were removed under high vacuum and the residue was then dissolved in dichloromethane. After filtration, the filtrate was concentrated and precipitated with diethyl ether. The precipitate was then collected and dried to yield a white solid.

*Compound **Zn1***. Compound **Zn1** was synthesized according to the general procedure using 100 mg of [**L1**]I (yield: 207.1 mg, 0.243 mmol, 87%). Suitable crystals of **Zn1** for single-crystal X-ray diffraction study was obtained *via* slow evaporation from methanolic solution of **Zn1**. ^{1}H NMR (400 MHz, DMSO-d_6) δ 11.96 (s, 2H), 7.87 (s, 4H), 7.64 (d, J = 10.5 Hz, 4H), 7.24 (d, J = 8.4 Hz, 4H), 3.97 (s, 12H), 2.30 (s, 6H) ppm. ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 150.9, 138.3, 137.5, 134.8, 129.4, 123.8, 120.5, 36.3, 20.6 ppm. IR (KBr): 3483, 1682, and 1529 cm^{-1}.

*Compound **Zn2***. Compound **Zn2** was synthesized according to the general procedure using 100 mg of [**L2**]I (yield: 175.0 mg, 0.199 mmol, 83%). ^{1}H NMR (400 MHz, DMSO-d_6) δ 11.24

(s, 2H), 7.93 (s, 4H), 7.34 (s, 4H), 6.90 (s, 2H), 3.97 (s, 12H), 2.29 (s, 12H) ppm. ^{13}C{^{1}H} NMR (101 MHz, DMSO-*d*$_6$) δ 150.8, 138.4, 138.1, 136.7, 127.2, 123.9, 118.1, 36.3, 21.0 ppm. IR (KBr): 3448, 1686, and 1530 cm^{-1}.

*Compound **Zn3***. Compound **Zn3** was synthesized according to the general procedure using 100 mg of [**L3**]I yield: 166.9 mg, 0.187 mmol, 79%). ^{1}H NMR (400 MHz, DMSO-*d*$_6$) δ 11.51 (s, 2H), 7.93 (s, 4H), 7.74 (d, *J* = 8.9 Hz, 4H), 7.53 (d, *J* = 8.9 Hz, 4H), 3.98 (s, 12H) ppm. ^{13}C{^{1}H} NMR (101 MHz, DMSO-*d*$_6$) δ 151.1, 137.7, 135.9, 129.4, 129.1, 124.1 122.2, 36.4 ppm. IR (KBr): 3435, 1690, and 1528 cm^{-1}.

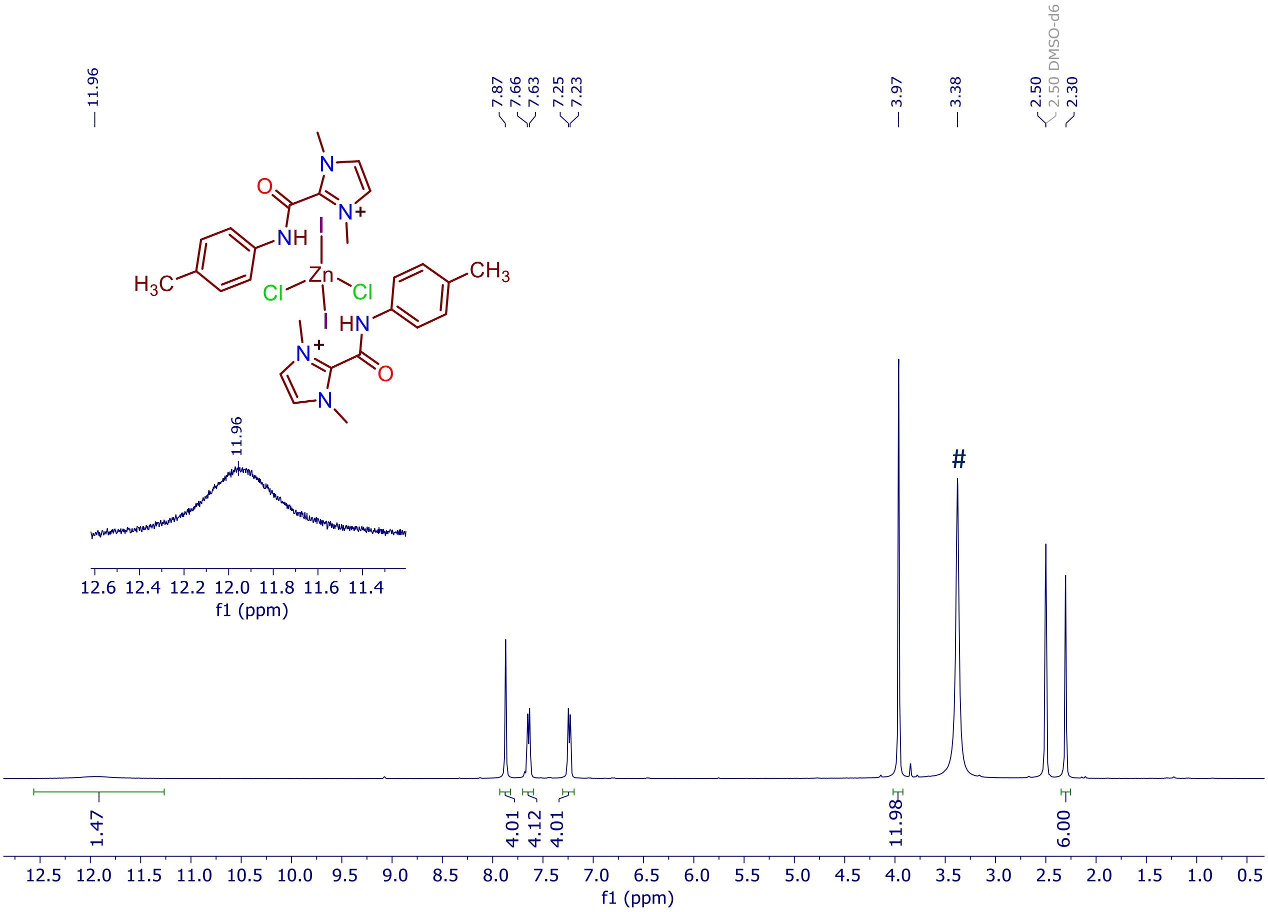


***Figure S7**. ^{1}H NMR of compound **Zn1** in DMSO-d$_6$. # indicates solvent impurity of H_2O in DMSO-d$_6$*

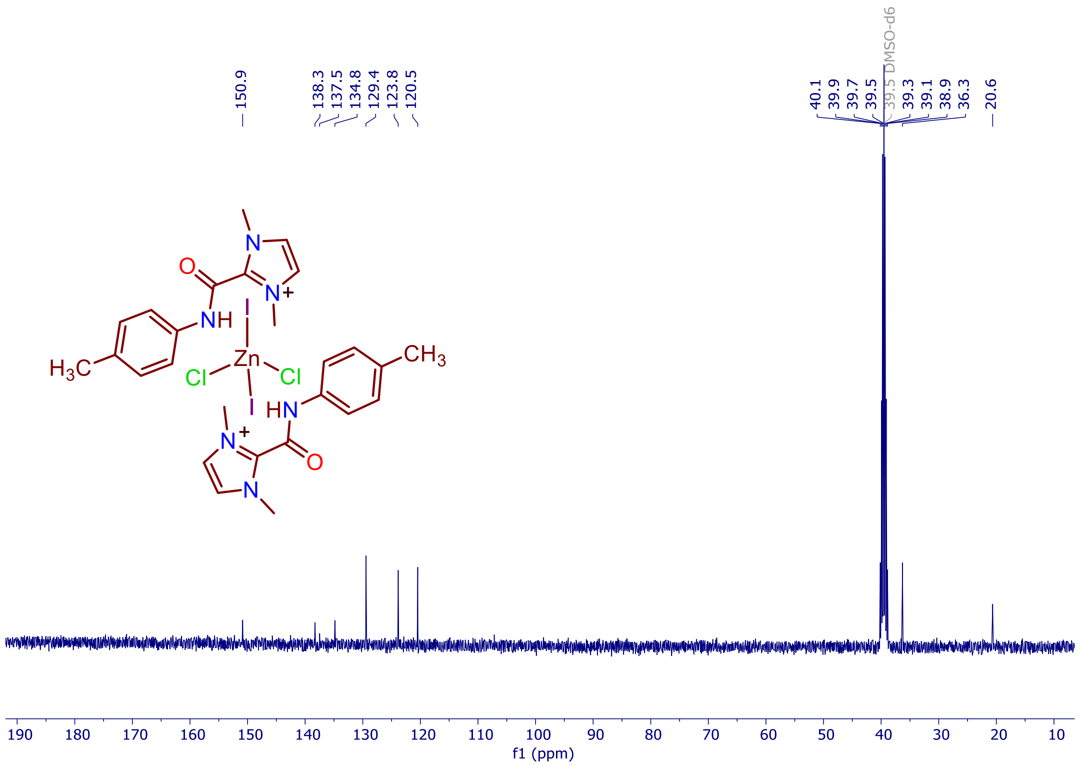


Figure S8. *$^{13}C\{^{1}H\}$ NMR of compound* ***Zn1*** *in DMSO-d_6*

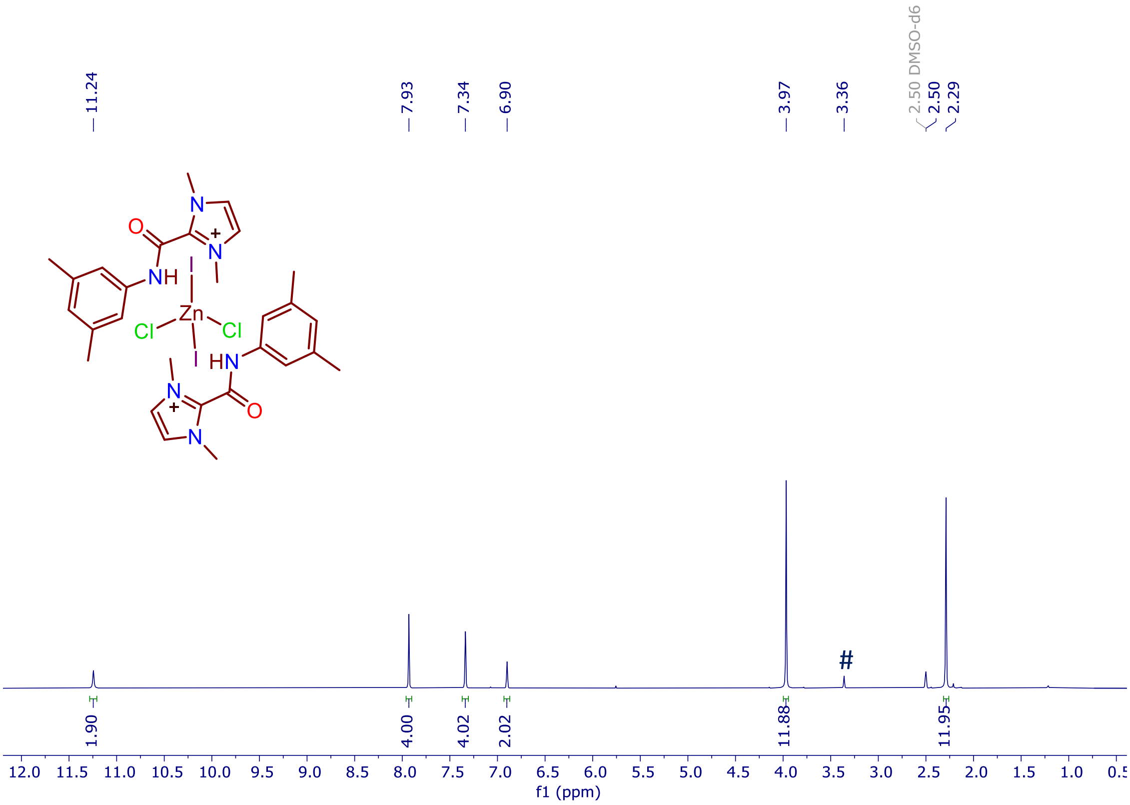


Figure S9. *^{1}H NMR of compound* ***Zn2*** *in DMSO-d_6. # indicates solvent impurity of H_2O in DMSO-d_6*

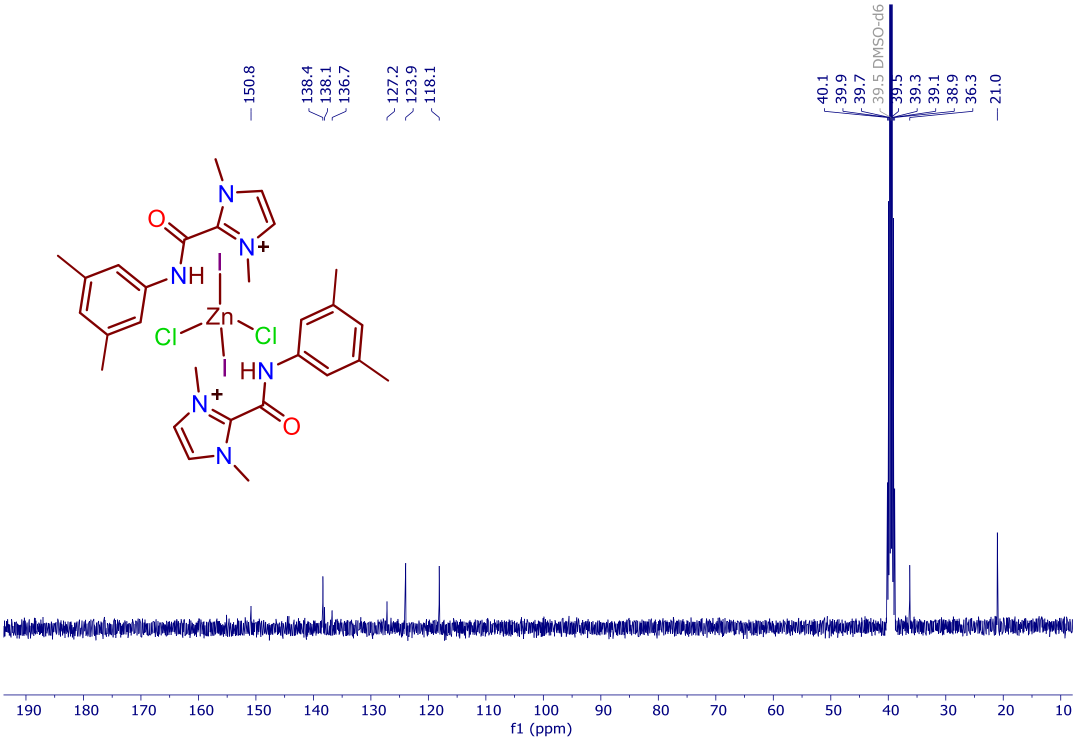


***Figure S10**. $^{13}C\{^1H\}$ NMR of compound **Zn2** in DMSO-d$_6$*

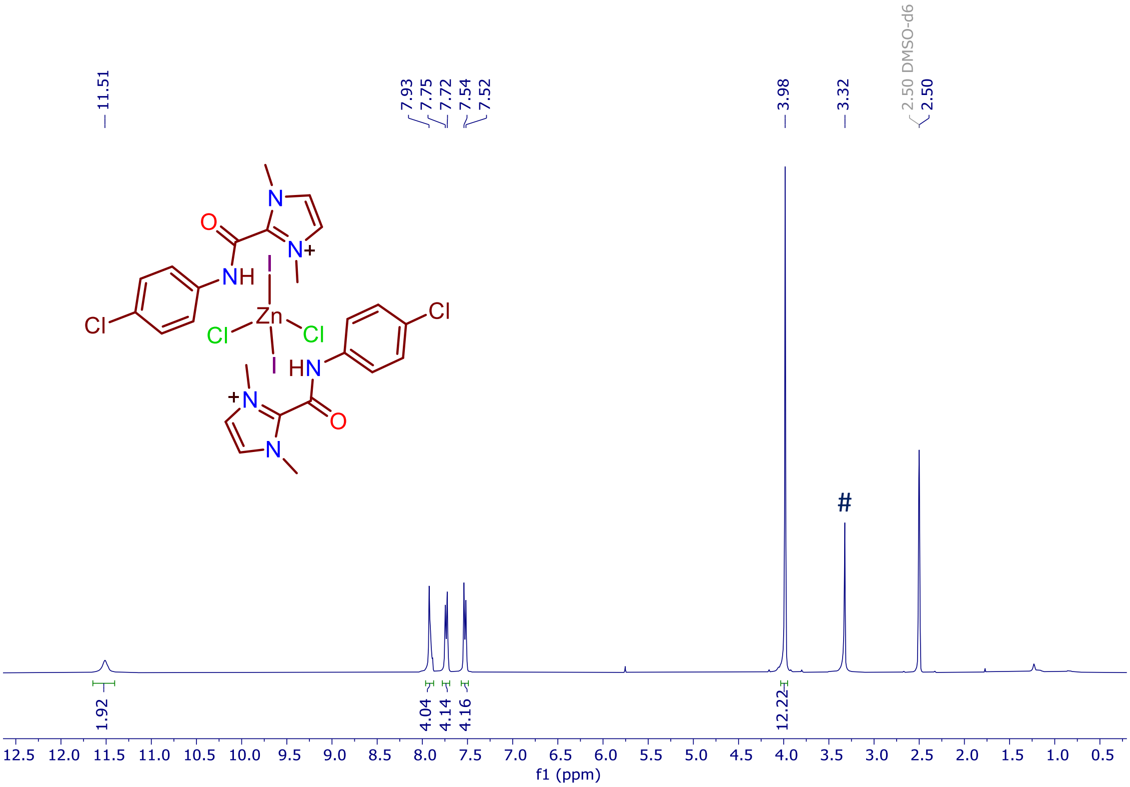


***Figure S11**. ^1H NMR of compound **Zn3** in DMSO-d$_6$. # indicates solvent impurity of H_2O in DMSO-d$_6$*

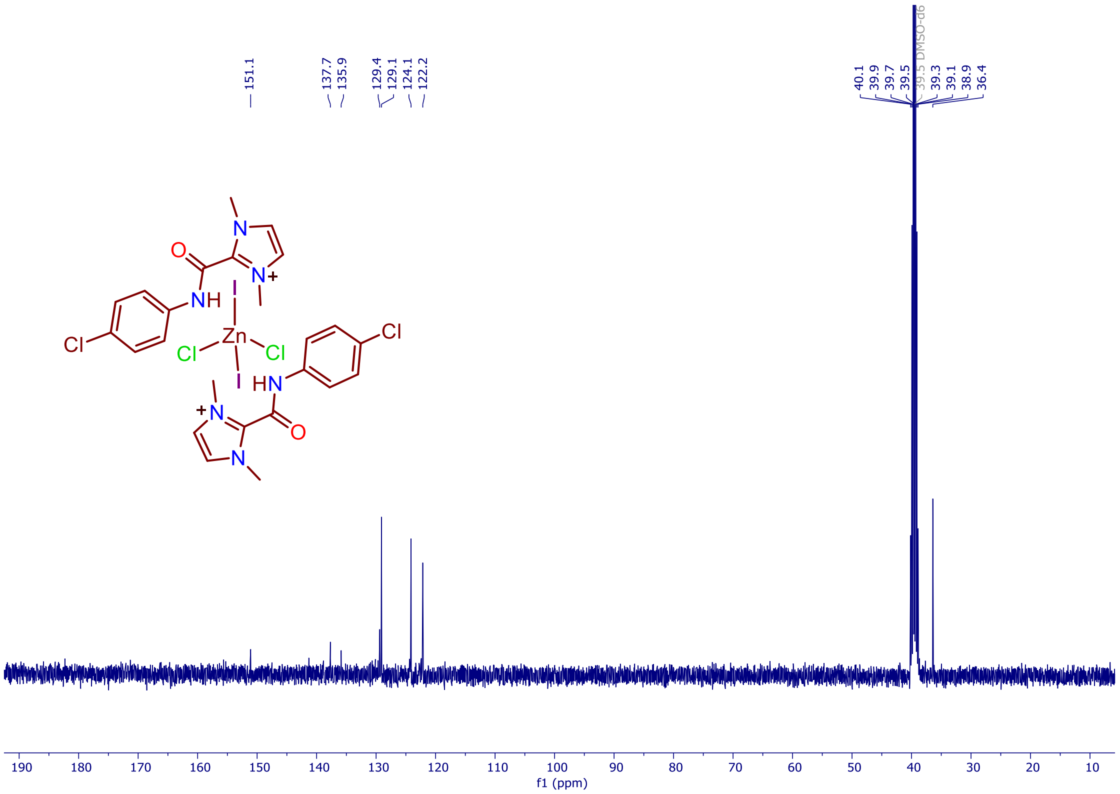


***Figure S12**. $^{13}C\{^{1}H\}$ NMR of compound **Zn3** in DMSO-d_6*

3. General procedure for alkenylation, double functionalization and alkylation reaction:

a) General procedure for the alkenylation of indene:

An oven-dried pressure tube (25 mL) was charged with indene (0.5 mmol), alcohol (0.55 mmol), LiO^{t}Bu (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%) followed by the addition of benzene (2 mL). Then, the tube was kept in an oil bath at 70 °C and heated for 24 h. After completion of the reaction, the desired products were isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

b) General procedure for the double functionalization of indene:

An oven-dried pressure tube (25 mL) was charged with indene (0.5 mmol), alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%) followed by the addition of benzene (2 mL). Then, the tube was kept in an oil bath at 70 °C and heated for 24 h. After completion of the reaction, the desired products were isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

c) General procedure for the alkenylation of fluorene:

An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), alcohol (0.55 mmol), LiO^{t}Bu (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%), followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. After completion of the reaction, the desired products were isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

d) General procedure for the alkylation of fluorene:

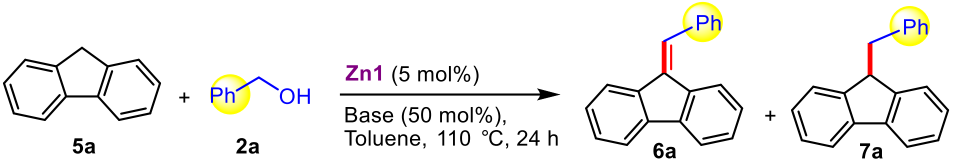
An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%), followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. After completion of the reaction, the desired products were isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

Table S1: Optimization of the reaction conditions for the selective C-alkenylation and double functionalization of indene[a]

Entry	**Catalyst**	**Temp (℃)**	**Base**	**Indene: alcohol**	**Solvent**	**Yield (%)**	
						3a	**4a**
1	[**L1**]I + $ZnBr_2$	70	LiO^{t}Bu	1: 1.1	Benzene	61	-
2	[**L1**]I + $ZnCl_2$	70	LiO^{t}Bu	1: 1.1	Benzene	68	-
3	[**L1**]I + $Zn(OAc)_2$	70	LiO^{t}Bu	1: 1.1	Benzene	29	-
4	[**L1**]I + $Zn(OTf)_2$	70	LiO^{t}Bu	1: 1.1	Benzene	35	-
5	**Zn1**	**70**	**LiO^{t}Bu**	**1: 1.1**	**Benzene**	**83**	-
6	**Zn2**	70	LiO^{t}Bu	1: 1.1	Benzene	59	-
7	**Zn3**	70	LiO^{t}Bu	1: 1.1	Benzene	67	-
8	**Zn1**	70	NaO^{t}Bu	1: 1.1	Benzene	37	15
9	**Zn1**	70	CsOH	1: 1:1	Benzene	12	43
10	**Zn1**	70	LiO^{t}Bu	1: 1.1	DCE	53	-
11	**Zn1**	70	LiO^{t}Bu	1: 1.1	Toluene	41	-
12	**Zn1**	**70**	**CsOH**	**1: 2.2**	**Benzene**	**10**	**71**
13	**Zn1**	70	LiO^{t}Bu	1: 2.2	Benzene	80	-
14	-	70	LiO^{t}Bu	1: 1.1	Benzene	<10	-
15	**Zn1**	70	-	1: 1.1	Benzene	trace	-
16	$ZnCl_2$	70	LiO^{t}Bu	1: 1.1	Benzene	trace	-
17	[**L1**]I	70	LiO^{t}Bu	1: 1.1	Benzene	30	-
18	**Zn1**	70	LiO^{t}Bu	1: 1.1	DMF	19	-
19	**Zn1**	70	LiO^{t}Bu	1: 1.1	DMSO	26	-
20	**Zn1**	70	LiO^{t}Bu	1: 1.1	Acetonitrile	32	-
21	**Zn1**	70	LiO^{t}Bu	1: 1.1	Dioxane	21	-
22	**Zn1**	60	LiO^{t}Bu	1: 1.1	Benzene	77	-
23	**Zn1**	80	LiO^{t}Bu	1: 1.1	Benzene	84	-
24	**Zn1**	90	LiO^{t}Bu	1: 1.1	Benzene	79	-
25	**Zn1**	70	NaOH	1: 1.1	Benzene	20	33
26	**Zn1**	70	Na_2CO_3	1: 1.1	Benzene	17	-
27	**Zn1**	70	K_2CO_3	1: 1.1	Benzene	14	-
28	**Zn1**	70	CsOH	1: 2.5	Benzene	12	73
29	**Zn1**	70	CsOH	1: 3	Benzene	18	70

[a]**Reaction conditions**: indene (0.5 mmol), benzyl alcohol (0.55-1.5 mmol), **Zn1** (5 mol%), and base (0.25 mmol) in solvent (2 mL) at 60-90 ℃ for 24 h.

Table S2. Optimization of the reaction conditions for selective C-alkenylation and C-alkylation of fluorene[a]



Entry	Catalyst	Base	Solvent	Temp (°C)	Fluorene : alcohol	Yield (%)	
						6a	7a
1	**Zn1**	LiO^{t}Bu	Benzene	70	1: 1.1	23	-
2	**Zn1**	**LiO^{t}Bu**	**Toluene**	**110**	**1: 1.1**	**78**	-
3	**Zn1**	NaO^{t}Bu	Toluene	110	1: 1.1	54	19
4	**Zn1**	CsOH	Toluene	110	1: 1.1	Not separable	
5	**Zn1**	**CsOH**	**Toluene**	**110**	**1: 2.2**	**7**	**83**
6	**Zn1**	NaOH	Toluene	110	1: 2.2	28	37
7	[**L1**]I + $ZnCl_2$	CsOH	Toluene	110	1: 2.2	-	67
8	-	LiO^{t}Bu	Toluene	110	1: 1.1	15	-
9	**Zn1**	LiO^{t}Bu	DMF	110	1: 1.1	31	-
10	**Zn1**	LiO^{t}Bu	DMSO	110	1: 1.1	27	-
11	**Zn1**	LiO^{t}Bu	Acetonitrile	110	1: 1.1	33	-
12	**Zn1**	LiO^{t}Bu	Toluene	90	1: 1.1	46	-
13	**Zn1**	LiO^{t}Bu	Toluene	100	1: 1.1	57	-
14	**Zn1**	LiO^{t}Bu	Toluene	120	1: 1.1	77	-
15	**Zn1**	LiO^{t}Bu	Toluene	130	1: 1.1	75	-
16	**Zn1**	Na_2CO_3	Toluene	110	1: 1.1	14	-
17	**Zn1**	K_2CO_3	Toluene	110	1: 1.1	19	-

[a] **Reaction conditions**: fluorene (0.5 mmol), benzyl alcohol (0.55-1.1 mmol), **Zn1** (5 mol%), and base (0.25 mmol) in solvent (2 mL) at 90-130 °C for 24 h.

4. General synthetic method for the alkylation of fluorene in gram scale:

An oven-dried pressure tube (25 mL) was charged with fluorene (1 gm, 6.01 mmol), benzyl alcohol (13.2 mmol), CsOH (3.00 mmol, 50 mol%), and **Zn1** (0.300 mmol, 5 mol%), followed by the addition of toluene (15 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. After completion of the reaction, the desired product (**7a**) was isolated by column chromatography over silica gel using hexane.

5. General procedure for competitive experiments:

*(a) Alkylation of fluorene (**5a**) with primary alcohol and secondary alcohol*:

An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), benzyl alcohol (1.1 mmol), 1-phenylethan-1-ol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%), followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. After completion of the reaction, the products **7a** (yield: 68%, 87.1 mg) and **7o** (yield: 23%, 31.1 mg) were isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

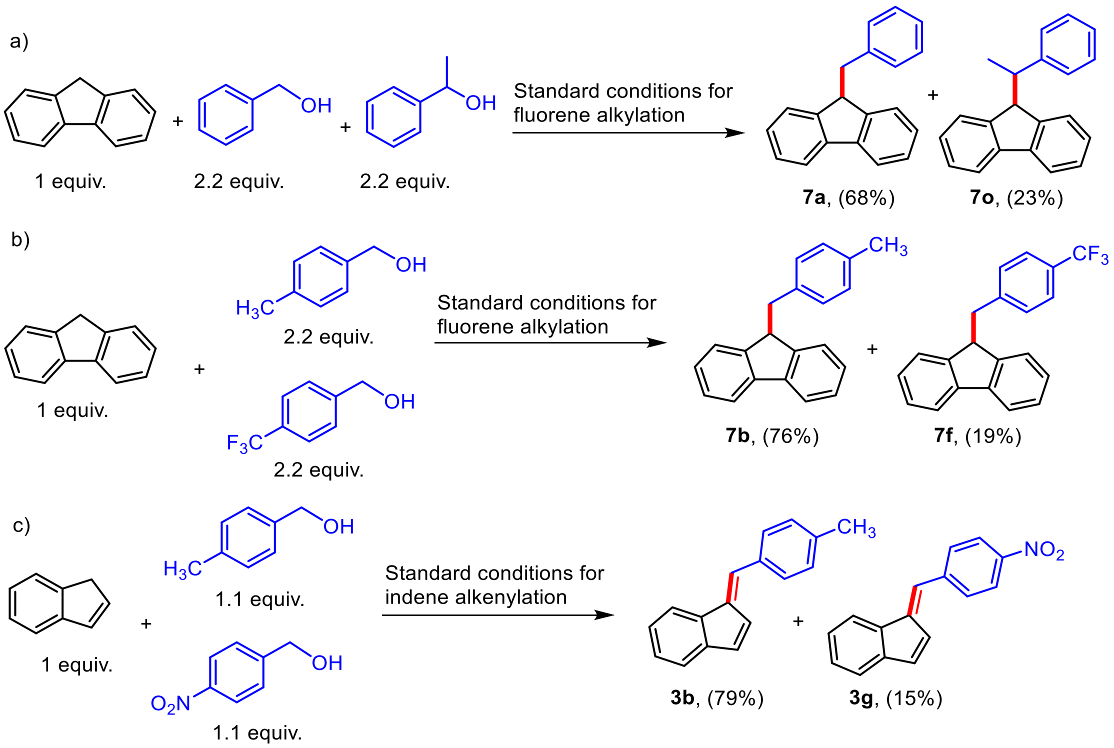
*(b) Alkylation of fluorene (**5a**) with electron-rich and electron-deficient benzyl alcohol*:

An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), 4-methyl benzyl alcohol (1.1 mmol), 4-(trifluoromethyl)benzyl alcohol (1.1 mmol), CsOH (0.250 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%), followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. After completion of the reaction, the products **7b** (yield: 76%, 102.7 mg) and **7f** (yield: 19%, 31 mg) were isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

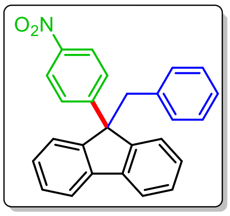
(c) *Alkenylation of indene (**1a**) with electron-rich and electron-deficient benzyl alcohol*:

An oven-dried pressure tube (25 mL) was charged with indene (0.5 mmol), 4-methyl benzyl alcohol (0.55 mmol), 4-nitrobenzyl alcohol (0.55 mmol), LiO^{t}Bu (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%), followed by the addition of benzene (2 mL). Then, the tube was kept in an oil bath at 70 °C and heated for 24 h. After completion of the reaction, the products **3b** (yield: 79%, 86.2 mg) and **3g** (yield: 15%, 18.7 mg) were isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

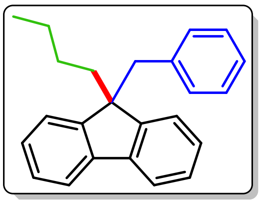
Scheme S2. Competitive experiments



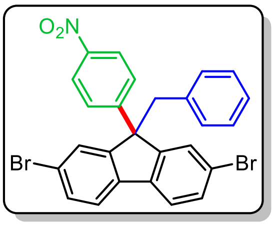
6. Late-stage modification of mono-alkylated fluorene:



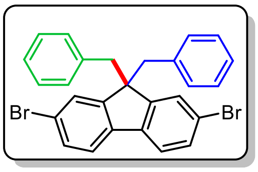
(a) Compound **8a** was synthesized following the reported procedure.[2] A mixture of 9-benzyl-9*H*-fluorene **7a** (0.4 mmol), 4-chloronitrobenzene (0.6 mmol), CsOH (0.6 mmol), and acetonitrile (1.0 mL) in a Schlenk tube (25 mL) was heated at 80 °C for 24 h. After completion of the reaction, the mixture was successively washed with brine solution, followed by extraction with ethyl acetate. The combined organic layer was concentrated under vacuum and then purified through column chromatography over silica gel using ethyl acetate/hexane as eluent, providing **8a** in 77% isolated yield.



(b) Compound **8b** was synthesized following the reported procedure.[2] A mixture of 9-benzyl-9*H*-fluorene **7a** (0.4 mmol), 1-bromobutane (1.6 mmol), TBAI (0.08 mmol), NaOH (0.2 g) and water (1 mL) in a Schlenk tube (25 mL) was heated at 80 °C for 24 h. After completion of the reaction, the mixture was successively washed with brine solution, followed by extraction with ethyl acetate. The combined organic layer was concentrated under vacuum, then purified through column chromatography over silica gel using ethyl acetate and hexane as eluent, giving **8b** in 73% isolated yield.



(c) Compound **8c** was synthesized following the reported procedure.[2] A mixture of 9-benzyl-2,7-dibromo-9*H*-fluorene **7w** (0.2 mmol), 4-chloronitrobenzene (0.3 mmol), $CsOH \cdot H_2O$ (0.3 mmol), and acetonitrile (1.0 mL) in a Schlenk tube (25 mL) was heated at 80 °C for 24 h and then the reaction progress was monitored by TLC. After completion of the reaction, the mixture was successively washed with brine solution, followed by extraction with ethyl acetate. The combined organic layer was concentrated under vacuum and then purified through column chromatography over silica gel using ethyl acetate and hexane as the eluent, giving **8c** in 69% isolated yield.



(d) Compound **8d** was synthesized following the reported procedure.[3] A mixture of 9-benzyl-2,7-dibromo-9*H*-fluorene **7w** (0.2 mmol), benzyl bromide (0.8 mmol), TBAI (0.08 mmol), NaOH (0.2 g) and water (1 mL) in a Schlenk tube (25 mL) was heated at 80 °C for 24 h. After completion of the reaction, the mixture was successively washed with brine solution, followed by extraction with ethyl acetate. The combined organic layer was concentrated under vacuum, then purified through column chromatography over silica gel using ethyl acetate and hexane as eluent, giving **8d** in 71% isolated yield.

7. EPR analysis:

(a) *Procedure for singly reduced product of* ***Zn1***:

In a Schlenk tube, **Zn1** (1 equiv.) and LiO^{t}Bu (1 equiv.) were added followed by the addition of methanol (2 mL). The reaction mixture was stirred for 1 h. During that time colour of the solution changed to light yellow. Then, EPR measurement of this solution was carried out at room temperature under inert condition.

EPR Detail:

The one-electron reduced product of **Zn1a** was analysed by X-band EPR (Bruker) at room temperature. The parameters during the data collection were following. Microwave frequency 9.43 GHz; Microwave Power 0.99 MW; Modulation frequency 100 kHz; Modulation amplitude 0.2 mT.

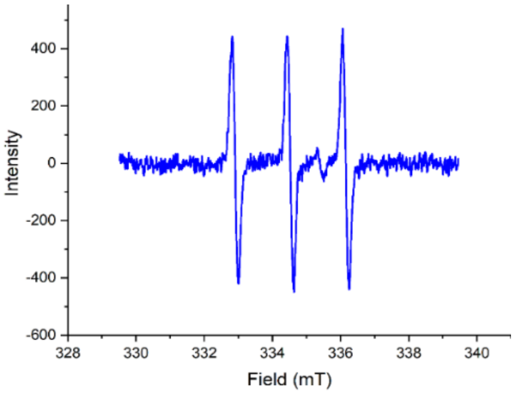


Figure S13. *EPR signal obtained from singly reduced product of* ***Zn1***

*(b) Procedure for singly reduced product of [****L1****]I*:

In a Schlenk tube, [**L1**]I (1 equiv.) and LiO^{t}Bu (1 equiv.) were added followed by methanol (2 mL). The reaction mixture was stirred for 1 h, no change in colour was observed during that time. Then the EPR measurement of this solution was carried out both at room temperature and liquid nitrogen temperature. No signal was observed.

EPR Detail:

The resultant solution was analysed by X-band EPR (Bruker) at room temperature. The parameters during the data collection were following. Microwave frequency 9.43 GHz; Microwave Power 2 MW; Modulation frequency 100 kHz; Modulation amplitude 0.35 mT.

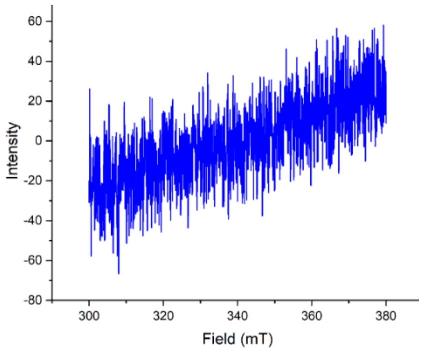
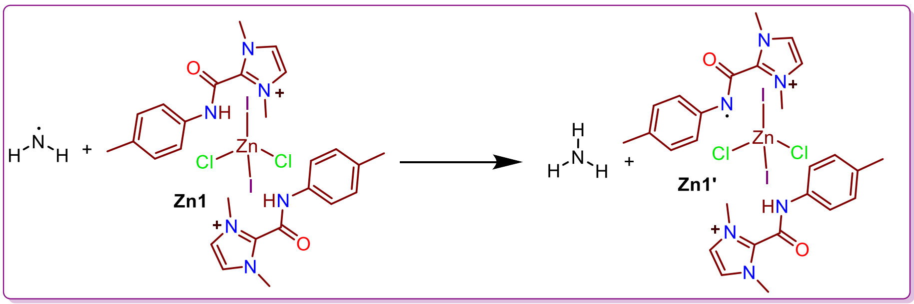


Figure S14. *EPR spectrum obtained from [**L1**]I and base*

8. Calculation of bond-dissociation enthalpy (BDE):

The BDE (N-H) values of amidated compound (**Zn1**) were estimated following the theoretical scheme reported by Zipse *et al*.[4] The stabilities of amidyl radicals (**Zn1**-radical) relative to the reference aminyl radical (·NH_2) were calculated as the reaction enthalpies at 298.15 K for the hydrogen atom transfer reaction shown below (ΔH_{rxn}). We could estimate the BDE of **Zn1** by adding the calculated reaction enthalpy to the experimentally determined BDE value of ammonia (450.1 kJ mol^{-1} = 107.6 kcal mol^{-1}).



Geometry optimizations of amidated compounds (**Zn1** and **Zn2**) and their corresponding radical forms (**Zn1'** radical and **Zn2'** radical) were performed at the B3LYP/6-31G(d). Thermochemical corrections to 298.15 K with a scaling factor of 0.9806 were used to obtain an enthalpy of the systems at the same level of theory. Components of the calculations are summarized in Table S3. The Gaussian 16, Revision B.01 program was used for all calculations.

Table S3. Enthalpies and BDE data

	H_{298}/Hf
NH_3	-56.519520
$NH_2\bullet$	-55.856231
Zn1	-2497.418608
Zn1’	-2496.773001

ΔH_{rxn} = -11.0943768 kcal mol^{-1}

BDE = 96.5 kcal mol^{-1}

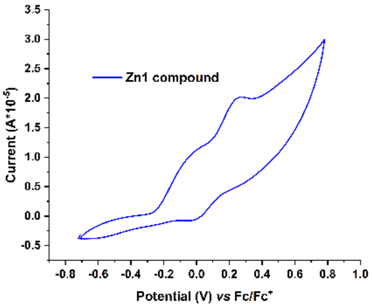
	H_{298}/Hf
NH_3	-56.519520
$NH_2\bullet$	-55.856231
Zn2	-2575.961881
Zn2’	-2575.310970

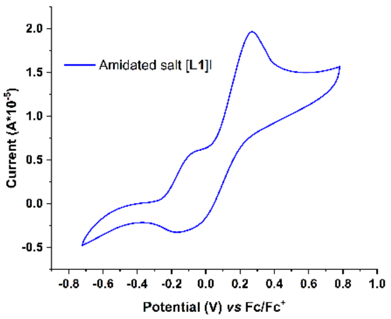
ΔH_{rxn} = -7.768573 kcal mol^{-1}

BDE = 99.8 kcal mol^{-1}

9. Electrochemical analysis of the [L1]I and compound Zn1:

The electrochemical measurements, cyclic voltammetry (CV), of the synthesized amidated imidazolium salt [**L1**]I and compound **Zn1** were carried out at ambient temperature with Metrohm auto lab potentiostat and galvanostat MAC90009 instrument, respectively. The measurements of [**L1**]I and **Zn1** were performed at a sweep rate of 100 mV/sec with three-electrode configuration such as auxiliary electrode: Pt wire; working electrode: Glassy carbon; reference electrode: Ag/Ag$^+$. All the measurements were calibrated externally using Ferrocene ($E_{1/2}$, Fc/Fc$^+$ = 0.22 volts *vs* Ag/Ag$^+$).





10. Control experiments for establishing a radical pathway:

(a) Radical scavenger experiments:

(i) An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), 4-methyl benzylalcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), **Zn1** (0.025 mmol, 5 mol%), and radical scavengers, BHT/ Galvinoxy/ $CuCl_2$ (0.5 mmol) followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. After completion of the reaction, the desired product (**7b**) was isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

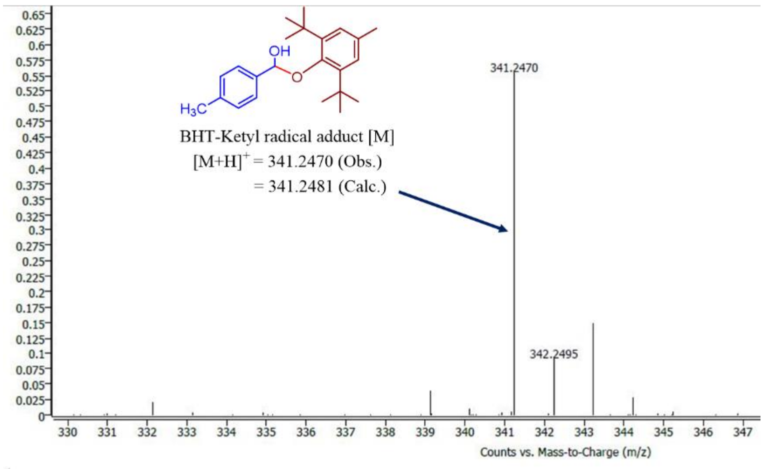


Figure S15. *Portion of ESI mass spectrum of the reaction mixture of standard fluorene alkylation in presence of BHT. M (molecular ion) corresponds to (p-tolyl)methanol radical trapped with BHT.*

(ii) An oven-dried pressure tube (25 mL) was charged with benzylalcohol (0.5 mmol), CsOH (0.25 mmol, 50 mol%), **Zn1** (0.025 mmol, 5 mol%), and BHT (0.5 mmol) followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. An aliquot from the reaction mixture was analysed by ESI-MS analysis.

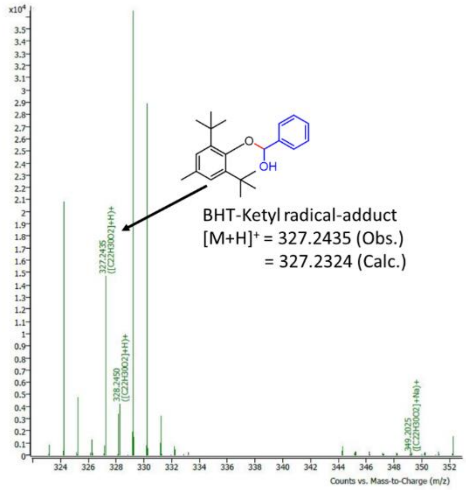


Figure S16. *Portion of ESI mass spectrum of the reaction mixture of a stoichiometric reaction between benzyl alcohol and BHT in presence of* ***Zn1****. M corresponds to benzyl alcohol radical trapped with BHT.*

(b) Hydrogen atom transfer experiment:

An oven-dried pressure tube (25 mL) was charged with cyclobutanol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%) followed by the addition of toluene (2 mL).

Then, the tube was kept in an oil bath at 110 °C and heated for 12 h. After completion of the reaction, the reaction was analysed by NMR spectroscopy.

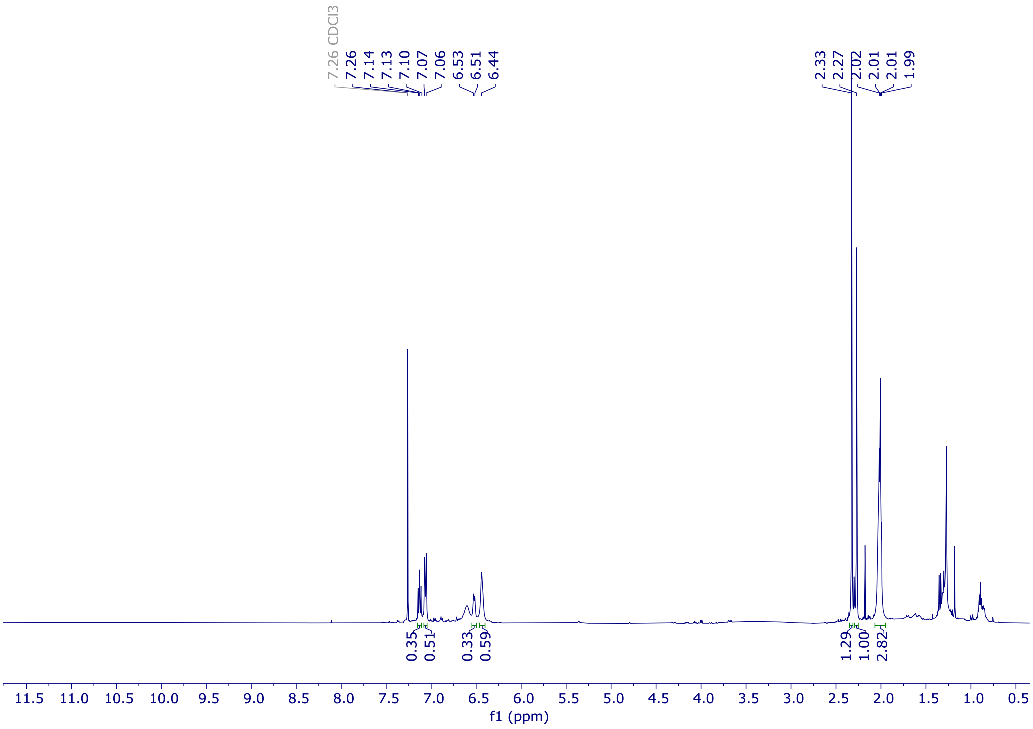


Figure S17. *^{1}H NMR spectrum of reaction mixture for the dehydrogenation of cyclobutanol*

11. Control experiments for detecting catalytic intermediates:

(a) Alcohol dehydrogenation:

An oven-dried pressure tube (25 mL) was charged with benzyl alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%) and **Zn1** (0.025 mmol, 5 mol%) followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 12 h. After completion of the reaction, the reaction mixture was analysed by GC-MS, using mesitylene as internal standard.

(b) Alkenylation of fluorene:

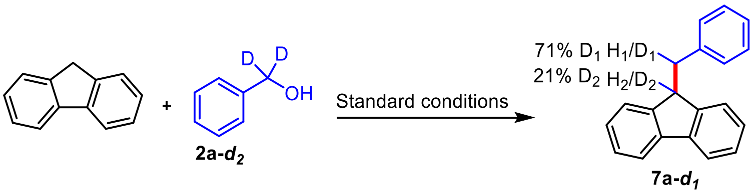
An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), benzaldehyde (0.5 mmol), LiO^{t}Bu (0.25 mmol, 50 mol%) followed by the addition of toluene (2 mL) with or without catalyst (**Zn1**). The reaction mixture was kept for heating at 110 °C for 24 h. After completion of the reaction, the desired product (**6a**) were isolated by column chromatography over silica gel using hexane as eluent.

*(c) Zinc catalyzed hydrogenation of intermediate (**6a**) by alcohol (**2a**)*:

An oven-dried pressure tube (25 mL) was charged with **Zn1** (0.025 mmol, 5 mol%), 9-benzylidene-9H-fluorene (**6a**), benzyl alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%) and toluene (2 mL). The reaction mixture was heated at 110 °C for 24 h. After completion of the reaction, the desired product (79%, **7a**) was isolated by column chromatography over silica gel using hexane as eluent.

(d) Deuterium labelling experiment:

An oven-dried pressure tube (25 mL) was charged with fluorene (0.25 mmol), deuterated benzyl alcohol (0.55 mmol), CsOH (0.12 mmol, 25 mol%) and **Zn1** (0.012 mmol, 5 mol%), followed by the addition of toluene (2 mL). Then, the tube was kept in oil bath at 110 °C and heated for the specified time. After completion of the reaction, the desired product was isolated by column chromatography over silica gel using hexane. The ^{1}H NMR analysis of the product **7a-*d₁*** revealed the 21-71% D incorporation in the product.



		H_1	H_2
Signal δ	7.24 [2H]	3.10 [2H]	4.23 [1H]
Integral Value	2.00	0.58	0.79
Calculated ratio	-	{(2-0.58)/2}*100 = 71%	{(1-0.79)/1}*100 = 21%

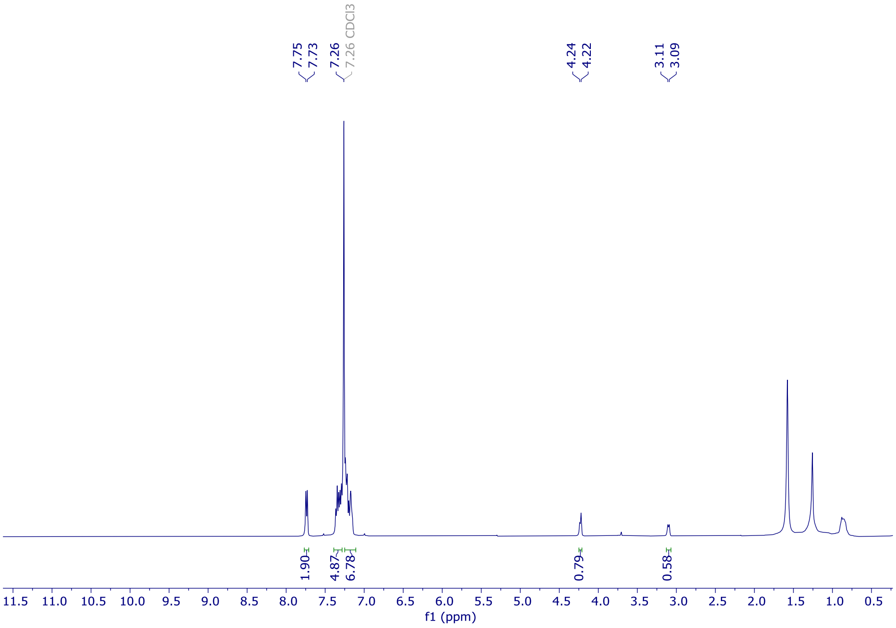


Figure S18. *^{1}H NMR of deuterium labelling experiment in $CDCl_3$*

(e) Procedure for mercury dropping test:

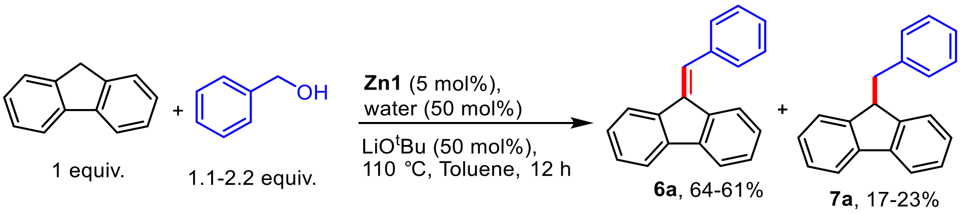
An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), 4-methyl benzylalcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), **Zn1** (0.025 mmol, 5 mol%), and mercury (40 mg, 2 mmol, 4 equiv. w.r.t. fluorene) followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and heated for 24 h. After completion of the reaction, the desired product (**7b**) was isolated by column chromatography over silica gel using hexane/ethyl acetate as eluent.

(f) Detection of intermediate:

An oven-dried pressure tube (25 mL) was charged with **Zn1** (0.025 mmol, 5 mol%), (E)-1-benzylidene-1H-indene (**3a**), benzyl alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%) and benzene (2 mL). The reaction mixture was heated at 70 °C for 24 h. After completion of the reaction, the desired product (66%, **4a**) was isolated by column chromatography over silica gel using hexane as eluent.

(g) Alk(en)ylation of fluorene in presence of water:

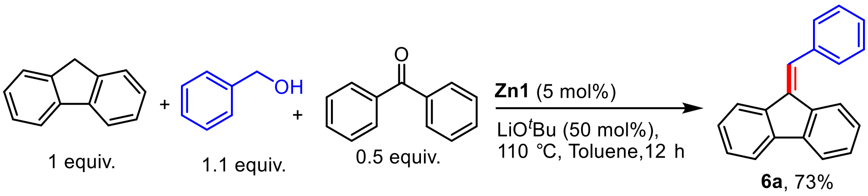
An oven-dried pressure tube (25 mL) was charged with **Zn1** (0.025 mmol, 5 mol%), fluorene (0.5 mmol), benzyl alcohol (0.55-1.1 mmol), LiO^{t}Bu (0.25 mmol, 50 mol%), water (0.25 mmol) and toluene (2 mL). The reaction mixture was heated at 110 °C for 24 h. After completion of the reaction, the reaction mixture was analysed by GC-MS using mesitylene as internal standard.



(h) Alk(en)ylation of fluorene in presence of benzophenone:

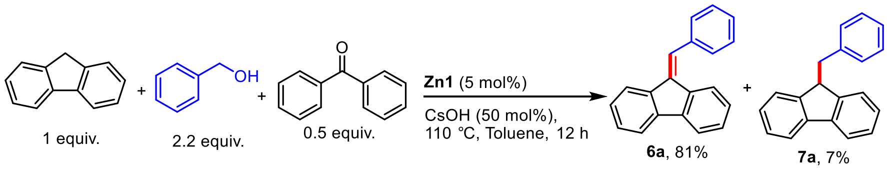
(i) An oven-dried pressure tube (25 mL) was charged with **Zn1** (0.025 mmol, 5 mol%), fluorene (0.5 mmol), benzyl alcohol (0.55 mmol), LiO^{t}Bu (0.25 mmol, 50 mol%), benzophenone (0.25 mmol) and toluene (2 mL). The reaction mixture was heated at 110 °C for 12 h. After completion of the reaction, the desired product (73%, **6a**) was isolated by column chromatography over silica gel using hexane as eluent.

Scheme S3. Alkenylation of fluorene in presence of benzophenone



(ii) An oven-dried pressure tube (25 mL) was charged with **Zn1** (0.025 mmol, 5 mol%), fluorene (0.5 mmol), benzyl alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), benzophenone (0.25 mmol) and toluene (2 mL). The reaction mixture was heated at 110 °C for 12 h. After completion of the reaction, **6a** was obtained in 81% along with **7a** in 7% after column chromatography over silica gel using hexane as eluent.

Scheme S4. Alkylation of fluorene in presence of benzophenone



12. *In situ* NMR monitoring of the functionalization of fluorene and indene:

(a) *In situ* NMR monitoring experiment for fluorene:

An oven-dried pressure tube (25 mL) was charged with fluorene (0.5 mmol), alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%), followed by the addition of toluene (2 mL). Then, the tube was kept in an oil bath at 110 °C and progress of the reaction was monitored over the interval of 6 h and 12 h, using ^{1}H NMR analysis.

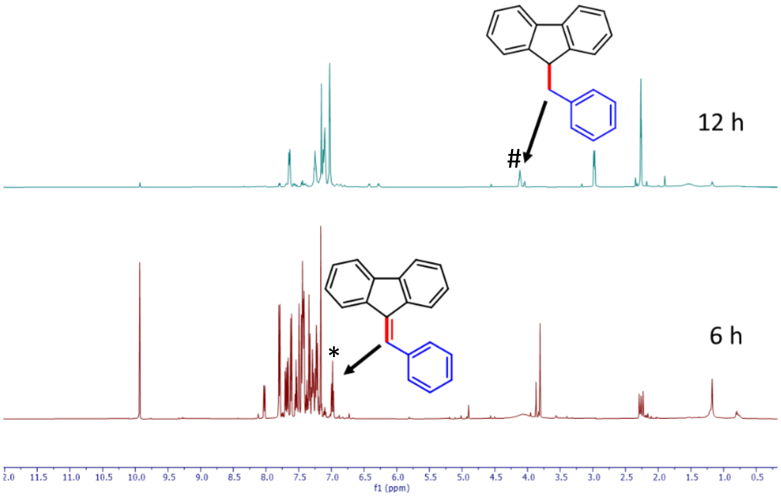


Figure S19. *Stacked ^{1}H NMR spectra of crude reaction for the alkylation of fluorene at different time interval. * alkenylated product, # alkylated product*

(b) *In situ* NMR monitoring experiment for indene:

An oven-dried pressure tube (25 mL) was charged with indene (0.5 mmol), alcohol (1.1 mmol), CsOH (0.25 mmol, 50 mol%), and **Zn1** (0.025 mmol, 5 mol%), followed by the addition of benzene (2 mL). Then, the tube was kept in oil bath at 70 °C and the progress of the reaction was monitored over the interval of 4-12 h, using ^{1}H NMR analysis.

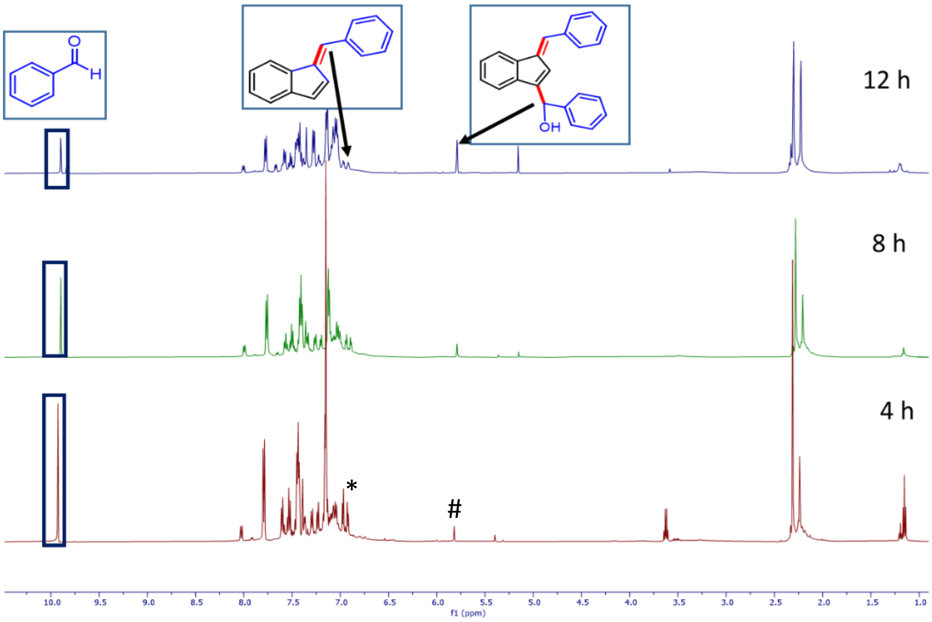


***Figure S20**. Stacked ^{1}H NMR spectra of crude reaction for the alkenylation of indene at different time interval. * alkenylated product, # double functionalized product*

13. UV-visible absorption studies for kinetics analysis:

*(a) Calculation of the reaction order with respect to catalyst loading [**Zn1**]*:

Different sets of reactions were conducted by varying the catalyst loading (3-6 mol%) keeping the other factors constant. An aliquot (5 µL) of the reaction mixture was then taken from the Schlenk tube after certain time interval and the spectral changes were monitored at 262 nm with appropriate dilution.

(b) Calculation of the reaction order with respect to 4-methylbenzyl alcohol

Different set of reactions were conducted by varying the alcohol amount (1.4-2.6 equiv.) keeping the other factors constant. An aliquot (5 µL) of the reaction mixture was then taken from the Schlenk tube after certain time interval and the spectral changes were monitored at 262 nm with appropriate dilution.

14. Molar magnetic susceptibility calculation for compound Zn1:

According to Evans method, $\chi_g = \frac{CL(R-R_0)}{10^9(m-m_0)}$ ……...Eq. (1)

where, χ_g = Gram magnetic susceptibility

C = Balance calibration constant = 1

L = Height of sample in tube in cm

R = Reading for tube + sample

R_0 = Reading for the empty tube

m = Weight of tube + sample in gram

m_0 = Weight of the empty tube in gram

Molar magnetic susceptibility, $\chi_m = \chi_g$ * Molecular weight of sample ……..Eq. (2)

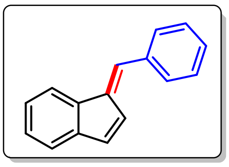
	Compound **Zn1**
Molecular weight	850.686
L	1.7 cm
R	-6
R_0	-6
m	1.7573 g
m_0	1.7367 g

For **Zn1**,

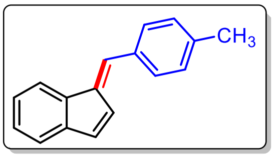
From Eq. (1), $\chi_g = 0$

Hence, from Eq. (2) $\chi_m = 0$

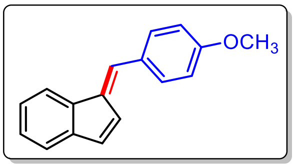
15. Analytical data of isolated compounds:



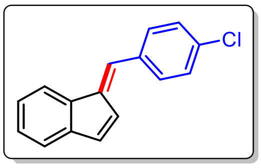
(E)-1-benzylidene-1H-indene (Compound-3a):[5] Following the general procedure 3(a), the titled compound was isolated as yellow solid (86.8 mg, 0.425 mmol, 85% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.73 (d, *J* = 8.2 Hz, 1H), 7.64 (d, *J* = 7.7 Hz, 2H), 7.53 (s, 1H), 7.46 (t, *J* = 7.66 Hz, 2H), 7.39-7.35 (m, 2H), 7.31-7.28 (m, 2H), 7.08-7.04 (m, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 142.2, 140.2, 137.6, 137.1, 134.7, 130.4, 128.9, 128.8, 128.5, 127.7, 126.2, 125.3, 121.1, 119.3 ppm.



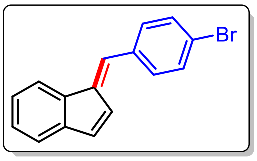
(E)-1-(4-methylbenzylidene)-1H -indene (Compound-3b):[5] Following the general procedure 3(a), the titled compound was isolated as yellow solid (90.5 mg, 0.415 mmol, 83% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.72 (d, *J* = 7.8 Hz, 1H), 7.55 (d, *J* = 7.7 Hz, 2H), 7.50 (s, 1H), 7.36-7.34 (m, 1H), 7.29-7.25 (m, 4H), 7.09-7.08 (m, 1H), 7.05-7.03 (m, 1H), 2.43 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 142.1, 139.4, 138.7, 137.7, 134.3, 130.4, 129.6, 129.0, 127.5, 126.3, 125.2, 121.1, 119.2, 21.5 ppm.



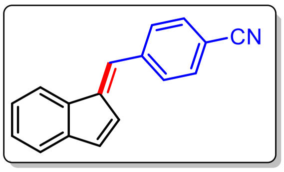
(E)-1-(4-methoxybenzylidene)-1H-indene (Compound-3c):[5] Following the general procedure 3(a), the titled compound was isolated as yellow solid (103 mg, 0.440 mmol, 88% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (d, J = 6.1 Hz, 1H), 7.60 (d, J = 8.8 Hz, 2H), 7.47 (s, 1H), 7.35 (d, J = 7.0 Hz, 1H), 7.22-7.25 (m, 2H), 7.08 (d, J = 5.5 Hz, 1H), 7.03 (d, J = 5.7 Hz, 1H), 6.98 (d, J = 8.8 Hz, 2H), 3.87 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 160.1, 141.9, 138.3, 137.8, 133.9, 131.9, 129.8, 128.8, 127.3, 126.0, 125.1, 121.0, 119.0, 114.4, 55.5 ppm.



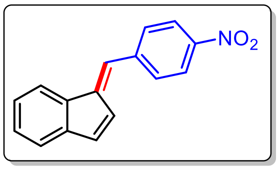
(E)-1-(4-chlorobenzylidene)-1H-indene (Compound-3d):[5] Following the general procedure 3(a), the titled compound was isolated as yellow solid (90.7 mg, 0.380 mmol, 76% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, J = 7.0 Hz, 1H), 7.53 (d, J = 8.6 Hz, 2H), 7.42-7.40 (m, 3H), 7.34-7.32 (m, 1H), 7.29-7.22 (m, 2H), 7.04 (d, J = 5.6 Hz, 1H), 6.97-6.96 (m, 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 142.2, 140.7, 137.4, 135.5, 135.2, 134.5, 131.5, 129.1, 127.9, 127.3, 125.8, 125.5, 121.2, 119.3 ppm.



(E)-1-(4-bromobenzylidene)-1H-indene (Compound-3e):[6] Following the general procedure 3(a), the titled compound was isolated as yellow solid (104.7 mg, 0.370 mmol, 74% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, J = 7.1 Hz, 1H), 7.56 (d, J = 8.6 Hz, 2H), 7.46 (d, J = 8.4 Hz, 2H), 7.40 (s, 1H), 7.33 (d, J = 6.2 Hz, 1H), 7.30-7.22 (m, 2H), 7.04 (d, J = 4.2 Hz, 1H), 6.96 (d, J = 5.6 Hz, 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 142.2, 140.8, 137.4, 136.0, 135.3, 132.0, 131.7, 127.9, 127.3, 125.8, 125.5, 122.8, 121.2, 119.4 ppm.

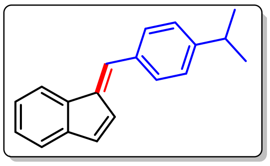


(E)-4-((1H-inden-1-ylidene)methyl)benzonitrile (Compound-3f):[6] Following the general procedure 3(a), the titled compound was isolated as yellow solid (76.8 mg, 0.335 mmol, 67% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71-7.65 (m, 5H), 7.42 (s, 1H), 7.32-7.22 (m, 3H), 7.06 (d, J = 5.6 Hz, 1H), 6.89 (d, J = 5.6 Hz, 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 143.0, 142.3, 141.6, 137.0, 136.7, 132.5, 130.7, 128.6, 126.0, 125.8, 125.4, 121.5, 119.7, 118.9, 111.5 ppm.

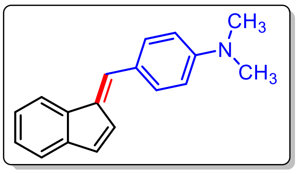


(E)-1-(4-nitrobenzylidene)-1H-indene (Compound-3g):[6] Following the general procedure 3(a), the titled compound was isolated as yellow solid (78.5 mg, 0.315 mmol, 63% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.28 (d, J = 8.9 Hz, 2H), 7.72-7.67 (m, 3H), 7.45 (s, 1H), 7.33-7.23 (m, 3H), 7.09 (d, J =

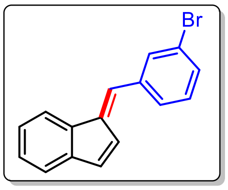
5.6 Hz, 1H), 6.90 (d, *J* = 5.6 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 147.2, 143.6, 143.5, 142.4, 137.1, 137.0, 130.8, 128.7, 125.9, 125.5, 125.4, 124.0, 121.5, 119.7 ppm.



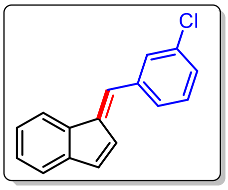
(E)-1-(4-isopropylbenzylidene)-1H-indene (Compound-3h): Following the general procedure 3(a), the titled compound was isolated as yellow solid (99.7 mg, 0.405 mmol, 81% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.71 (d, *J* = 8.25 Hz, 1H), 7.57 (d, *J* = 7.43 Hz, 2H), 7.49 (s, 1H), 7.33-7.30 (m, 3H), 7.24-7.21 (m, 2H), 7.08-7.07 (m, 1H), 7.03-7.01 (m, 1H), 3.00-2.93 (m, 1H), 1.30 (d, *J* = 7.0 Hz, 6H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 149.6, 142.1, 139.5, 137.7, 134.7, 134.2, 130.5, 129.0, 127.5, 127.0, 126.3, 125.2, 121.1, 119.2, 34.1, 24.0 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{19}H_{19}$ 247.1487; Found 247.1479.



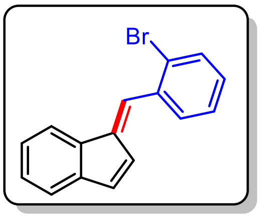
(E)-4-((1H-inden-1-ylidene)methyl)-N,N-dimethylaniline (Compound-3i):[5] Following the general procedure 3(a), the titled compound was isolated as yellow solid (101 mg, 0.410 mmol, 82% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.72-7.71 (m, 1H), 7.60 (d, *J* = 8.8 Hz, 2H), 7.45 (s, 1H), 7.35-7.34 (m, 1H), 7.23-7.21 (m, 2H), 7.17 (d, *J* = 5.5 Hz, 1H), 7.01 (d, *J* = 5.6 Hz, 1H), 6.76 (d, *J* = 7.1 Hz, 2H), 3.04 (s, 6H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 150.6, 141.5, 138.2, 135.8, 132.4, 132.1, 130.0, 126.5, 126.1, 125.0, 124.7, 120.9, 118.7, 112.2, 40.3 ppm.



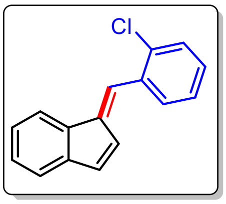
(E)-1-(3-bromobenzylidene)-1H-indene (Compound-3j): Following the general procedure 3(a), the titled compound was isolated as yellow solid (97.7 mg, 0.345 mmol, 69% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.75 (s, 1H), 7.67 (d, *J* = 6.8 Hz, 1H), 7.52-7.47 (m, 2H), 7.39 (s, 1H), 7.34-7.28 (m, 3H), 7.24-7.22 (m, 1H), 7.05 (d, *J* = 5.6 Hz, 1H), 6.96 (d, *J* = 5.6 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 142.3, 141.4, 139.1, 137.3, 135.6, 132.9, 131.2, 130.3, 128.8, 128.1, 126.8, 125.8, 125.5, 122.9, 121.2, 119.4 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{16}H_{12}Br$ 283.0122; Found 283.0112.



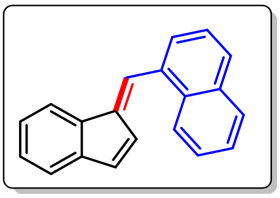
(E)-1-(3-chlorobenzylidene)-1H-indene (Compound-3k):[5] Following the general procedure 3(a), the titled compound was isolated as yellow solid (81.1 mg, 0.340 mmol, 68% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, *J* = 7.1 Hz, 1H), 7.59 (s, 1H), 7.47 (d, *J* = 7.2 Hz, 1H), 7.40-7.36 (m, 2H), 7.33-7.30 (m, 2H), 7.28-7.22 (m, 2H), 7.05 (d, *J* = 5.6 Hz, 1H), 6.97 (d, *J* = 5.6 Hz, 1H) ppm. $^{13}C\{^{1}H\}$ NMR (101 MHz, $CDCl_3$) δ 142.3, 141.4, 138.8, 137.3, 135.6, 134.7, 130.0, 130.0, 128.4, 128.3, 128.1, 126.9, 125.8, 125.5, 121.3, 119.4 ppm.



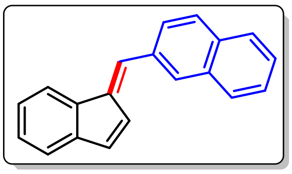
(E)-1-(2-bromobenzylidene)-1H-indene (Compound-3l): Following the general procedure 3(a), the titled compound was isolated as yellow solid (86.3 mg, 0.305 mmol, 61% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.38 (d, J = 6.8 Hz, 1H), 7.27 (s, 1H), 7.21 (d, J = 6.7 Hz, 1H), 6.98 (t, J = 6.9 Hz, 1H), 6.94-6.86 (m, 4H), 6.82 (t, J = 6.9 Hz, 1H), 6.64 (d, J = 5.6 Hz, 1H), 6.43 (d, J = 5.6 Hz, 1H) ppm. $^{13}C\{^{1}H\}$ NMR (101 MHz, $CDCl_3$) δ 142.3, 141.4, 139.1, 137.3, 135.6, 132.9, 131.2, 130.3, 128.8, 128.1, 126.8, 125.8, 125.5, 122.9, 121.2, 119.4 ppm. HRMS (ESI) m/z: $[M + NH_4]^+$: Calcd. for $C_{16}H_{11}BrNH_4$ 300.0388; Found 300.0472.



(E)-1-(2-chlorobenzylidene)-1H-indene (Compound-3m):[7] Following the general procedure 3(a), the titled compound was isolated as yellow solid (70.4 mg, 0.295 mmol, 59% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.78 (d, J = 6.5 Hz, 1H), 7.66 (d, J = 7.8 Hz, 2H), 7.61 (d, J = 7.7 Hz, 1H), 7.39 (t, J = 7.7 Hz, 1H), 7.35-7.20 (m, 4H), 7.03 (d, J = 5.6 Hz, 1H), 6.82 (d, J = 5.6 Hz, 1H) ppm. $^{13}C\{^{1}H\}$ NMR (101 MHz, $CDCl_3$) δ 142.6, 141.6, 137.0, 137.0, 135.2, 133.0, 132.7, 129.6, 128.1, 127.7, 127.5, 126.2, 125.5, 125.2, 121.2, 119.8 ppm.

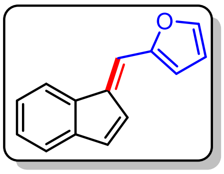


(E)-1-((1H-inden-1-ylidene)methyl)naphthalene (Compound-3n):[8] Following the general procedure 3(a), the titled compound was isolated as yellow solid (99.1 mg, 0.390 mmol, 78% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.18-8.16 (m, 2H), 7.94-7.85 (m, 3H), 7.66 (d, J = 7.1 Hz, 1H), 7.60-7.54 (m, 3H), 7.39-7.29 (m, 3H), 7.02 (d, J = 5.5 Hz, 1H), 6.86 (d, J = 5.7 Hz, 1H) ppm. $^{13}C\{^{1}H\}$ NMR (101 MHz, $CDCl_3$) δ 142.9, 142.2, 137.0, 134.3, 134.1, 133.7, 132.3, 129.5, 128.9, 128.7, 127.9, 127.2, 126.7, 126.3, 125.6, 125.3, 124.8, 121.2, 119.6 ppm.

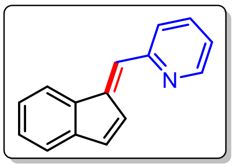


(E)-2-((1H-inden-1-ylidene)methyl)naphthalene (Compound-3o): Following the general procedure 3(a), the titled compound was isolated as yellow solid (101.7 mg, 0.400 mmol, 80% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.06 (s, 1H), 7.91-7.86 (m, 3H), 7.79-7.75 (m, 2H), 7.66 (s, 1H), 7.54-7.51 (m, 2H), 7.36 (d, J = 8.8 Hz, 1H), 7.29-7.28 (m, 2H), 7.16 (d, J = 5.7 Hz, 1H), 7.08 (d, J = 5.6 Hz, 1H) ppm. $^{13}C\{^{1}H\}$ NMR (101 MHz, $CDCl_3$) δ 142.2, 140.4, 137.7, 134.9, 134.6, 133.6, 133.2, 130.3, 128.9, 128.5, 128.4, 127.8, 127.7, 127.5, 126.8, 126.6, 126.3, 125.4, 121.1, 119.3 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{20}H_{15}$ 255.1174; Found 255.1169.

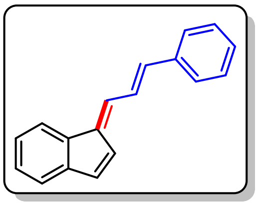
(E)-2-((1H-inden-1-ylidene)methyl)furan (Compound-3p):[7] Following the general procedure 3(a), the titled compound was isolated as yellow solid (62.1 mg, 0.319 mmol, 64% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.66-7.62 (m, 2H), 7.44-7.42 (m, 1H), 7.37-7.35 (m, 1H), 7.29-7.22 (m, 2H), 7.14 (s, 1H), 7.04-7.02 (m, 1H), 6.71 (s, 1H), 6.56 (s, 1H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 153.4, 144.9, 142.2, 137.5, 136.7, 133.7, 127.4, 125.0, 121.2, 119.1, 115.0, 114.3, 112.4 ppm.



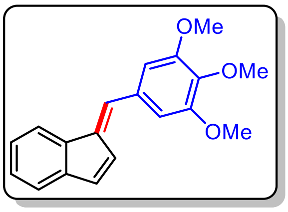
(E)-2-((1H-inden-1-ylidene)methyl)pyridine (Compound-3q):[8] Following the general procedure 3(a), the titled compound was isolated as yellow solid (67.7 mg, 0.330 mmol, 66% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.73 (d, J = 5.43 Hz, 1H), 7.72-7.65 (m, 3H), 7.51 (d, J = 7.9 Hz, 1H), 7.37 (s, 1H), 7.31- 7.17 (m, 4H), 7.03 (d, J = 5.7 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 155.8, 150.2, 142.9, 142.6, 137.8, 136.5, 135.9, 128.2, 127.6, 126.2, 126.0, 125.4, 122.5, 121.2, 119.6 ppm.



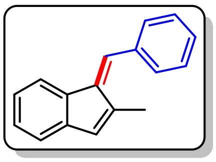
(E)-1-((E)-3-phenylallylidene)-1H-indene (Compound-3r):[7] Following the general procedure 3(a), the titled compound was isolated as yellow solid (88.0 mg, 0.385 mmol, 77% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.53 (d, J = 7.2 Hz, 1H), 7.43 (d, J = 7.8 Hz, 2H), 7.39-7.32 (m, 1H), 7.27 (t, J = 7.9 Hz, 2H), 7.22-7.20 (m, 2H), 7.14-7.09 (m, 3H), 6.87-6.82 (m, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 142.7, 140.9, 138.0, 136.9, 136.8, 132.5, 128.9, 128.7, 128.7, 127.6, 127.2, 125.7, 125.1, 124.7, 121.3, 119.5 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{18}H_{15}$ 231.1174; Found 231.1166.



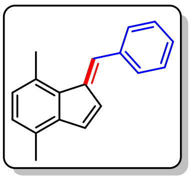
(E)-1-(3,4,5-trimethoxybenzylidene)-1H-indene (Compound-3s): Following the general procedure 3(a), the titled compound was isolated as yellow solid (92.7 mg, 0.314 mmol, 63% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, J = 7.1 Hz, 1H), 7.43 (s, 1H), 7.34-7.32 (m, 1H), 7.25-7.21 (m, 2H), 7.03 (s, 2H), 6.84 (s, 2H), 3.93 (s, 6H), 3.92 (s, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 153.5, 142.0, 139.7, 138.8, 137.6, 134.7, 132.6, 128.9, 127.6, 126.0, 125.3, 121.2, 119.2, 107.6, 61.1, 56.3 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{19}H_{19}O_3$ 295.1334; Found 295.1329.



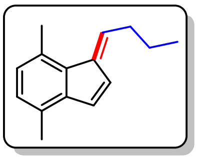
(E)-1-benzylidene-2-methyl-1H-indene (Compound-3t): Following the general procedure 3(a), the titled compound was isolated as yellow solid (45 mg, 0.205 mmol, 41% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 8.06 (d, J = 16.9 Hz, 1H), 7.85 (d, J = 19.0 Hz, 1H), 7.69-7.62 (m, 2H), 7.54-7.41 (m, 5H), 7.30-7.28 (d, J = 7.6 Hz, 2H), 2.46 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 142.1, 139.4, 138.7, 137.7, 134.3, 130.4, 129.6, 129.0, 127.5, 126.3, 125.2, 121.1, 119.2, 21.5 ppm. GC-MS: *m/z* 218 (M$^+$).



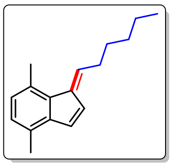
(E)-1-benzylidene-4,7-dimethyl-1H-indene (Compound-3u): Following the general procedure 3(a), the titled compound was isolated as yellow solid (88 mg, 0.379 mmol, 76% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.65 (s, 1H), 7.57-7.55 (m, 2H), 7.44 (t, J = 7.9 Hz, 2H), 7.37-7.34 (m, 1H), 7.08 (s, 1H), 7.01-6.93 (m, 3H), 2.66 (s, 3H), 2.43 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 143.3, 141.8, 138.0, 133.4, 133.2, 131.7, 130.7, 130.6, 129.1, 128.6, 128.4, 128.0, 126.5, 21.5, 18.1 ppm. GC-MS: *m/z* 232 (M$^+$).

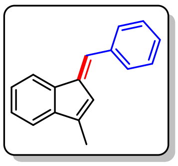


(E)-1-butylidene-4,7-dimethyl-1H-indene (Compound-3v): Following the general procedure 3(a), the titled compound was isolated as yellow liquid (68 mg, 0.343 mmol, 69% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 6.97-6.94 (m, 2H), 6.90-6.84 (m, 2H), 6.72 (s, 1H), 2.60-2.58 (m, 2H), 2.53 (s, 3H), 2.41 (s, 3H), 1.63-1.58 (m, 2H), 1.01 (t, J = 8.01 Hz, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 143.0, 142.1, 137.2, 132.9, 130.2, 129.3, 128.6, 128.1, 127.8, 125.2, 33.0, 23.5, 21.2, 18.1, 14.0 ppm. GC-MS: *m/z* 198 (M$^+$).

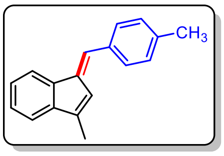


(E)-1-hexylidene-4,7-dimethyl-1H-indene (Compound-3w): Following the general procedure 3(a), the titled compound was isolated as yellow liquid (73.7 mg, 0.326 mmol, 65% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.01-6.96 (m, 2H), 6.93-6.87 (m, 2H), 6.73 (t, J = 8.2 Hz, 1H), 2.66-2.62 (m, 2H), 2.56 (s, 3H), 2.43 (s, 3H), 1.62-1.57 (m, 2H), 1.40-1.39 (m, 4H), 1.00-0.94 (m, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 142.8, 142.1, 137.5, 135.7, 132.9, 130.2, 129.3, 128.1, 127.7, 125.1, 31.7, 31.0, 29.9, 22.7, 21.2, 18.1, 14.2 ppm. GC-MS: *m/z* 226 (M$^+$).

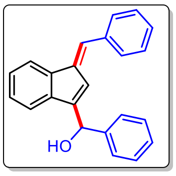




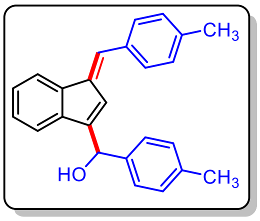
(E)-1-benzylidene-3-methyl-1H-indene (Compound-3x): Following the general procedure 3(a), the titled compound was isolated as yellow solid (77 mg, 0.352 mmol, 71% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.69 (d, J = 7.5 Hz, 1H), 7.61 (d, J = 8.3 Hz, 2H), 7.43 (t, J = 7.8 Hz, 2H), 7.36-7.29 (m, 5H), 6.81 (s, 1H), 2.28 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 144.8, 143.1, 139.6, 138.4, 137.4, 130.2, 128.8, 128.1, 127.5, 126.0, 125.4, 122.7, 118.9, 118.8, 13.6 ppm. GC-MS: *m/z* 218 (M^{+}).



(E)-3-methyl-1-(4-methylbenzylidene)-1H-indene (Compound-3y): Following the general procedure 3(a), the titled compound was isolated as yellow liquid (86 mg, 0.370 mmol, 74% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.61 (d, J = 7.5 Hz, 1H), 7.45 (d, J = 8.1 Hz, 2H), 7.26-7.15 (m, 6H), 6.76 (s, 1H), 2.33 (s, 3H), 2.21 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 144.2, 142.9, 138.7, 138.5, 138.2, 134.6, 130.1, 129.5, 127.3, 126.2, 125.3, 122.7, 118.8, 118.8, 21.5, 13.5 ppm. GC-MS: *m/z* 232 (M^{+}).

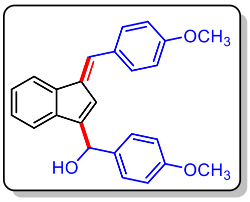


(E)-(1-benzylidene-1H-inden-3-yl)(phenyl)methanol (Compound-4a):[5] Following the general procedure 3(b), the titled compound was isolated as yellow solid (110 mg, 0.355 mmol, 71% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.60 (d, J = 7.5 Hz, 1H), 7.55 (d, J = 7.0 Hz, 2H), 7.39 (m, 3H), 7.35 (t, J = 8.1 Hz, 2H), 7.28-7.19 (m, 4H), 7.15-7.04 (m, 4H), 5.82 (s, 1H), 2.32 (br., 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 149.6, 141.7, 140.1, 138.8, 138.7, 137.0, 130.3, 128.9, 128.8, 128.6, 128.5, 128.3, 127.4, 127.2, 125.5, 122.2, 120.3, 119.2, 72.4 ppm.

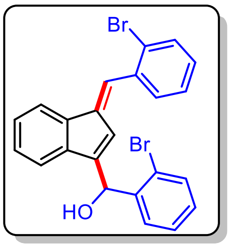


(E)-(1-(4-methylbenzylidene)-1H-inden-3-yl)(p-tolyl)methanol (Compound-4b):[7] Following the general procedure 3(b), the titled compound was isolated as yellow solid (113.3 mg, 0.335 mmol, 67% yield). ^{1}H NMR (500 MHz, CDCl$_3$) δ 7.79-7.76 (m, 1H), 7.67-7.64 (m, 2H), 7.55 (d, J = 5.3 Hz, 1H), 7.46 (t, J = 6.1 Hz, 2H), 7.35-7.34 (m, 2H), 7.30-7.22 (m, 6H), 5.94 (s, 1H), 2.51 (d, J = 6.1 Hz, 3H), 2.43 (d, J = 5.8 Hz, 3H) ppm. ^{13}C{^{1}H} NMR (126 MHz, CDCl$_3$) δ 149.3, 140.1, 138.8, 138.5, 138.0, 137.7, 134.2, 130.3, 129.5, 129.3, 128.3, 127.1, 127.1, 125.2, 121.9, 120.3, 119.0, 72.1, 21.4, 21.2 ppm.

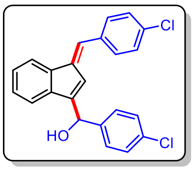
(E)-(1-(4-methoxybenzylidene)-1H-inden-3-yl)(4-methoxyphenyl)methanol (Compound-4c):[7] Following the general procedure 3(b), the titled compound was isolated as yellow solid (135.2 mg, 0.365 mmol, 73% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68 (d, J = 7.5 Hz, 1H), 7.62 (d, J = 8.8 Hz, 2H), 7.43-7.39 (m, 3H), 7.21-7.12 (m, 3H), 7.09-7.07 (m, 1H), 6.97 (d, J = 8.8 Hz, 2H), 6.87 (d, J = 8.7 Hz, 2H), 5.89 (s, 1H), 3.85 (s, 3H), 3.78 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 160.0, 159.5, 149.1, 139.9, 139.0, 136.9, 134.0, 131.9, 129.7, 128.6, 128.3, 126.9, 125.3, 121.7, 120.2, 119.0, 114.4, 114.1, 72.0, 55.5, 55.4 ppm.



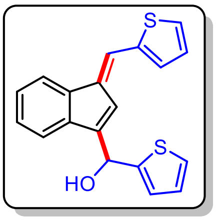
(E)-(1-(2-bromobenzylidene)-1H-inden-3-yl)(bromophenyl)methanol (Compound-4d): Following the general procedure 3(b), the titled compound was isolated as yellow solid (131.0 mg, 0.280 mmol, 56% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.62 (d, J = 7.7 Hz, 1H), 7.52-7.44 (m, 4H), 7.32 (d, J = 7.8 Hz, 1H), 7.21 (t, J = 7.5 Hz, 1H), 7.11-6.98 (m, 6H), 6.74 (s, 1H), 6.16 (s, 1H), 2.62 (br., 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 149.0, 140.5, 140.3, 140.2, 137.9, 136.9, 133.1, 133.0, 132.6, 129.9, 129.6, 129.2, 128.0, 127.9, 127.5, 127.5, 125.8, 125.2, 123.7, 122.6, 120.2, 71.0 ppm. HRMS (ESI) m/z: [M + Na]$^+$: Calcd. for $C_{23}H_{16}Br_2ONa$ 490.9447; Found 490.9439.



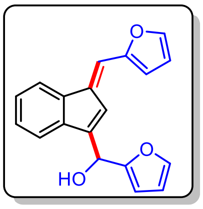
(E)-(1-(4-chlorobenzylidene)-1H-inden-3-yl)(4-chlorophenyl)methanol (Compound-4e):[5] Following the general procedure 3(b), the titled compound was isolated as yellow solid (125.1 mg, 0.330 mmol, 66% yield). ^{1}H NMR (400 MHz, $CDCl_3$) 7.66 (d, J = 7.2 Hz, 1H), 7.56 (d, J = 6.6 Hz, 2H), 7.47-7.44 (m, 4H), 7.37 (s, 1H), 7.34-7.32 (m, 2H), 7.23-7.16 (m, 2H), 7.10 (m, 1H), 6.98 (s, 1H), 5.83 (s, 1H), 2.51 (br., 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 149.6, 140.6, 139.8, 139.2, 138.5, 135.8, 132.0, 131.8, 131.6, 128.8, 127.7, 127.4, 125.9, 122.9, 122.2, 122.0, 120.5, 119.4, 71.6 ppm.



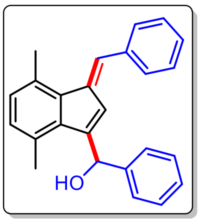
(E)-thiophen-2-yl(1-(thiophen-2-ylmethylene)-1H-inden-3-yl)methanol (Compound-4f): Following the general procedure 3(b), the titled compound was isolated as yellow solid (109.6 mg, 0.340 mmol, 68% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.61-7.59 (m, 1H), 7.56 (s, 1H), 7.50 (s, 1H), 7.42 (s, 1H), 7.23-7.18 (m, 3H), 7.08 (s, 1H), 6.66 (d, J = 3.4 Hz, 1H), 6.50-6.49 (m, 1H), 6.34-6.31 (m, 2H), 5.98 (s, 1H), 2.87 (br., 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 154.3,



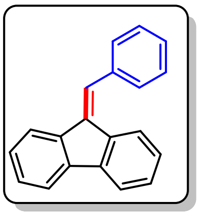
153.2, 145.6, 144.9, 142.6, 139.9, 138.3, 135.0, 127.1, 125.2, 123.9, 120.2, 119.0, 115.2, 114.4, 112.4, 110.5, 107.9, 65.8 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{19}H_{15}OS_2$ 323.0564; Found 323.0558.



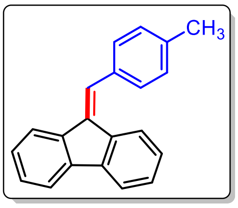
(E)-furan-2-yl(1-(furan-2-ylmethylene)-1H-inden-3-yl)methanol (Compound-4g):[7] Following the general procedure 3(b), the titled compound was isolated as yellow solid (101.6 mg, 0.350 mmol, 70% yield). ^{1}H NMR (500 MHz, CDCl$_3$) δ 7.62-7.60 (m, 1H), 7.57 (d, J = 2.1 Hz, 1H), 7.48 (s, 1H), 7.42 (d, J = 0.9 Hz, 1H), 7.23-7.17 (m, 3H), 7.10 (s, 1H), 6.67 (d, J = 3.5 Hz, 1H), 6.51-6.50 (m, 1H), 6.35-6.31 (m, 2H), 5.99 (s, 1H), 2.60 (s, 1H) ppm. ^{13}C{^{1}H} NMR (126 MHz, CDCl$_3$) δ 154.5, 153.3, 145.8, 144.9, 142.6, 140.0, 138.4, 135.2, 127.3, 125.2, 123.9, 120.3, 119.1, 115.2, 114.5, 112.4, 110.5, 107.9, 65.9 ppm.



(E)-(1-benzylidene-4,7-dimethyl-1H-inden-3-yl)(phenyl)methanol (Compound-4h): Following the general procedure 3(b), the titled compound was isolated as yellow solid (121.8 mg, 0.360 mmol, 72% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.49 (s, 1H), 7.39 (d, J = 8.4 Hz, 2H), 7.28-7.24 (m, 3H), 7.19-7.17 (m, 4H), 7.10 (s, 1H), 6.93 (s, 1H), 6.80-6.72 (m, 2H), 6.02 (s, 1H), 2.52 (s, 3H), 2.13 (s, 3H), 2.07 (s, 1H) ppm. ^{13}C{^{1}H} NMR (126 MHz, CDCl$_3$) δ 149.5, 142.1, 141.1, 139.6, 137.9, 135.3, 132.8, 131.0, 130.6, 130.5, 129.6, 129.1, 128.8, 128.7, 128.2, 128.0, 127.7, 125.3, 72.6, 22.0, 20.6 ppm.

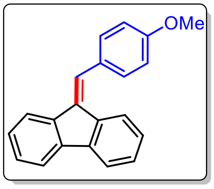


9-benzylidene-9H-fluorene (Compound-6a):[9] Following the general procedure 3(c), the titled compound was isolated as yellow solid (99.1 mg, 0.390 mmol, 78% yield). ^{1}H NMR (500 MHz, CDCl$_3$) δ 7.78 (d, J = 7.0 Hz, 1H), 7.74 (d, J = 7.4 Hz, 2H), 7.59-7.52 (m, 5H), 7.46-7.40 (m, 3H), 7.35 (t, J = 8.1 Hz, 2H), 7.11 (t, J = 7.1 Hz, 1H) ppm. ^{13}C{^{1}H} NMR (126 MHz, CDCl$_3$) δ 141.5, 139.4, 139.4, 137.2, 136.4, 135.4, 133.9, 130.7, 128.9, 128.5, 127.2, 126.9, 125.7, 124.4, 120.4, 119.9, 119.7 ppm.

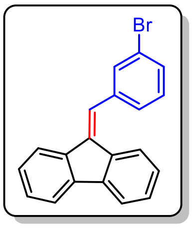


9-(4-methylbenzylidene)-9H-fluorene (Compound-6b):[10] Following the general procedure 3(c), the titled compound was isolated as yellow solid (101.9 mg, 0.380 mmol, 76% yield). ^{1}H NMR (500 MHz, CDCl$_3$) δ 7.81 (d, J = 7.8 Hz, 1H), 7.74 (d, J = 6.9 Hz, 2H), 7.69 (d, J = 7.6 Hz, 2H), 7.52 (d, J = 9.9 Hz, 2H), 7.41-7.32 (m, 3H), 7.29 (d, J = 7.6, 2H), 7.10 (t, J = 7.7 Hz, 1H), 2.47 (s, 3H) ppm. $^{13}C\{^1H\}$ NMR (126 MHz, $CDCl_3$) δ 141.3, 139.8, 139.2, 138.1, 136.8, 136.2, 134.0, 129.4, 129.4, 128.5, 128.2, 127.7, 127.1, 126.7, 124.5, 120.3, 119.8, 119.7, 21.6 ppm.

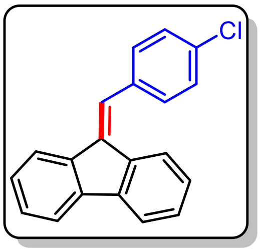
9-(4-methoxybenzylidene)-9H-fluorene (Compound-6c):[10] Following the general procedure 3(c), the titled compound was isolated as yellow solid (115.1 mg, 0.405 mmol, 81% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.71-7.63 (m, 4H), 7.54 (s, 1H), 7.45 (d, J = 8.3 Hz, 2H), 7.31-7.22 (m, 3H), 7.08-7.02 (m, 1H), 6.88 (d, J = 6.8 Hz, 2H), 3.74 (s, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 159.5, 141.1, 139.7, 138.9, 136.7, 135.4, 130.9, 129.0, 128.4, 127.9, 127.5, 126.9, 126.6, 124.2, 120.2, 119.8, 119.6, 113.9, 55.1 ppm.



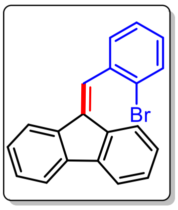
9-(3-bromobenzylidene)-9H-fluorene (Compound-6d):[11] Following the general procedure 3(c), the titled compound was isolated as yellow solid (109.9 mg, 0.329 mmol, 66% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.80-7.75 (m, 3H), 7.63-7.59 (m, 3H), 7.51-7.50 (m, 1H), 7.48-7.37 (m, 5H), 7.16 (t, J = 7.6 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 141.5, 139.4, 139.2, 138.8, 137.5, 136.2, 134.5, 129.8, 129.2, 128.9, 128.6, 128.1, 127.5, 127.2, 126.9, 125.3, 124.4, 120.4, 119.9, 119.7 ppm.



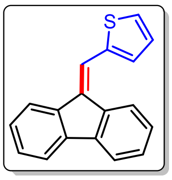
9-(4-chlorobenzylidene)-9H-fluorene (Compound-6e):[10] Following the general procedure 3(c), the titled compound was isolated as yellow solid (106.8 mg, 0.370 mmol, 74% yield). ^1H NMR (500 MHz, $CDCl_3$) δ 7.77 (d, J = 7.9 Hz, 1H), 7.73 (d, J = 7.9 Hz, 2H), 7.59-7.56 (m, 2H), 7.53 (d, J = 8.7 Hz, 2H), 7.46-7.40 (m, 3H), 7.35 (t, J = 8.1 Hz, 2H), 7.11 (t, J = 7.1 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (126 MHz, $CDCl_3$) δ 141.5, 139.4, 139.4, 137.2, 136.4, 135.4, 133.9, 130.7, 128.9, 128.5, 127.2, 126.9, 125.7, 124.4, 120.4, 119.9, 119.7 ppm.



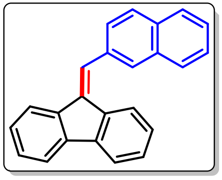
9-(2-bromobenzylidene)-9H-fluorene (Compound-6f): Following the general procedure 3(c), the titled compound was isolated as yellow solid (98.3 mg, 0.295 mmol, 59% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.93 (d, J = 7.6 Hz, 1H), 7.82-7.80 (m, 3H), 7.75 (d, J = 7.7 Hz, 1H), 7.67 (s, 1H), 7.51-7.39 (m, 5H), 7.33 (t, J = 7.9 Hz, 1H), 7.15 (t, J = 7.9 Hz, 1H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 141.4, 139.4, 139.1, 137.3, 137.2, 136.4, 132.9, 131.5, 129.7, 128.8, 128.6, 127.2, 127.2, 126.8, 126.0, 124.4, 124.1, 120.7, 119.9, 119.7 ppm. HRMS (ESI) m/z: $[M + H]^+$: Calcd. for $C_{20}H_{14}Br$ 335.0261; Found 335.0257.



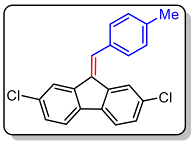
2-((9H-fluoren-9-ylidene)methyl)thiophene (Compound-6g):[10] Following the general procedure 3(c), the titled compound was isolated as yellow solid (92.4 mg, 0.355 mmol, 71% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.77 (d, J = 7.7 Hz, 1H), 7.74-7.67 (m, 4H), 7.40-7.22 (m, 5H), 6.75 (s, 1H), 6.56 (s, 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 152.3, 143.9, 141.1, 140.4, 139.0, 136.2, 132.8, 128.6, 128.0, 127.2, 126.9, 125.8, 119.9, 119.7, 119.7, 115.7, 112.8, 112.5 ppm.



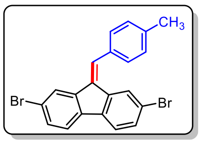
9-(naphthalen-2-ylmethylene)-9H-fluorene (Compound-6h):[12] Following the general procedure 3(c), the titled compound was isolated as yellow solid (105.0 mg, 0.345 mmol, 69% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07-8.06 (m, 2H), 7.83-7.75 (m, 3H), 7.55 (d, J = 7.1 Hz, 1H), 7.50-7.44 (m, 3H), 7.28-7.18 (m, 3H), 7.16 (s, 1H), 6.92 (d, J = 5.5 Hz, 1H), 6.75 (d, J = 5.7 Hz, 1H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 142.9, 142.2, 137.0, 134.3, 134.1, 133.7, 132.3, 129.5, 128.9, 128.7, 127.9, 127.2, 126.6, 126.3, 125.6, 125.3, 124.8, 121.2, 119.6 ppm.



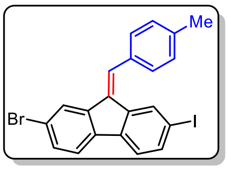
2,7-dichloro-9-(4-methylbenzylidene)-9H-fluorene (Compound-6i): Following the general procedure 3(c), the titled compound was isolated as yellow solid (123.0 mg, 0.365 mmol, 73% yield). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.54 (d, J = 2.0 Hz, 1H), 7.52 (d, J = 1.8 Hz, 1H), 7.48 (s, 1H), 7.39 (s, 1H), 7.37 (s, 1H), 7.31 (d, J = 7.8 Hz, 2H), 7.18-7.16 (m, 2H), 7.15 (s, 1H), 7.13-7.11 (m, 1H), 2.33 (s, 3H) ppm. ^{13}C{^{1}H} NMR (126 MHz, $CDCl_3$) δ 141.3, 139.1, 138.7, 138.0, 136.5, 134.1, 133.2, 132.8, 132.6, 130.2, 129.5, 129.4, 128.6, 128.2, 124.5, 120.7, 120.6, 120.6, 21.6 ppm. HRMS (ESI) m/z: $[M + H]^+$: Calcd. for $C_{21}H_{15}Cl_2$ 337.0551; Found 337.0547.



2,7-dibromo-9-(4-methylbenzylidene)-9H-fluorene (Compound-6j):[11] Following the general procedure 3(c), the titled compound was isolated as white solid (159.8 mg, 0.375 mmol, 75% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.64 (d, J = 6.0 Hz, 2H), 7.40 (s, 1H), 7.27-7.21 (m, 6H), 7.12-7.09 (d, J = 7.9 Hz, 2H), 2.29 (s, 3H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 141.3, 139.1, 139.0, 138.1, 136.8, 133.8, 132.7, 131.3, 130.9, 130.3, 129.5, 129.4, 127.3, 123.5, 121.2, 121.0, 120.9, 120.8, 21.6 ppm.

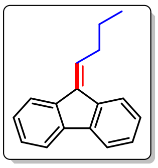


2-bromo-7-iodo-9-(4-methylbenzylidene)-9H-fluorene (Compound-6k): Following the general procedure 3(c), the titled compound was isolated as yellow solid (167.9 mg, 0.355 mmol, 71% yield). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.84 (s, 1H), 7.63 (s, 1H), 7.46- 7.39 (m, 2H), 7.22-7.20 (m, 3H), 7.14-7.09 (m,

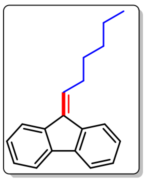


4H), 2.28 (s,3H) ppm. $^{13}C\{^1H\}$ NMR (126 MHz, $CDCl_3$) δ 141.3, 141.0, 139.4, 139.0, 138.2, 137.7, 137.3, 137.1, 136.7, 133.5, 133.0, 132.6, 131.3, 130.8, 130.2, 130.1, 129.4, 127.1, 123.4, 121.3, 121.1, 120.9, 120.8, 92.5, 92.2, 21.6 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{21}H_{15}BrI$ 474.9384; Found 474.9379.

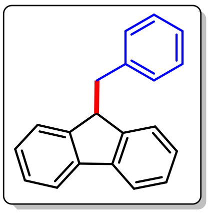
9-butylidene-9H-fluorene (Compound-6l): Following the general procedure 3(c), the titled compound was isolated as white solid (75 mg, 0.341 mmol, 68% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.93 (d, J = 7.9 Hz, 1H), 7.83-7.72 (m, 3H), 7.43-7.33 (m, 4H), 6.80 (t, J = 7.4 Hz, 1H), 2.87 (q, J = 8.6 Hz, 2H), 1.78 (q, J = 8.5 Hz, 2H), 1.15 (t, J = 7.6 Hz, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 140.9, 139.5, 138.7, 137.7, 135.6, 131.2, 127.7, 127.4, 127.0, 126.9, 125.1, 119.9, 119.8, 119.6, 31.5, 23.0, 14.2 ppm. GC-MS: *m/z* 220 (M^+).



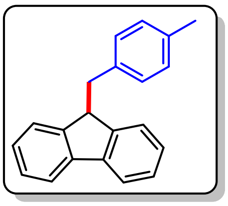
9-hexylidene-9H-fluorene (Compound-6m): Following the general procedure 3(c), the titled compound was isolated as white liquid (79 mg, 0.318 mmol, 64% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.97-7.75 (m, 4H), 7.44-7.36 (m, 4H), 6.83 (t, J = 7.5 Hz, 1H), 2.94-2.88 (m, 2H), 1.82-1.76 (m, 2H), 1.55-1.47 (m, 4H), 1.05 (t, J = 6.8 Hz, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 140.9, 139.5, 138.6, 137.7, 135.4, 131.4, 127.7, 127.4, 126.9, 126.9, 125.1, 119.9, 119.7, 119.5, 31.9, 29.5, 29.4, 22.7, 14.2 ppm. GC-MS: *m/z* 248 (M^+).



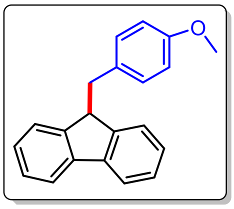
9-benzyl-9H-fluorene (Compound-7a):[12] Following the general procedure 3(d), the titled compound was isolated as white solid (106.3 mg, 0.415 mmol, 83% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.76 (d, J = 7.6 Hz, 2H), 7.39-7.29 (m, 5H), 7.28-7.18 (m, 7H), 4.25 (t, J = 7.6 Hz, 1H), 3.13 (d, J = 7.6 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 146.9, 140.9, 139.9, 129.7, 128.4, 127.2, 126.8, 126.5, 125.0, 120.0, 48.8, 40.2 ppm.



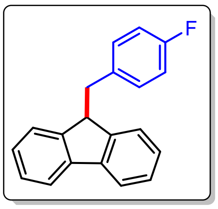
9-(4-methyllbenzyl)-9H-fluorene (Compound-7b):[13] Following the general procedure 3(d), the titled compound was isolated as white solid (109.5 mg, 0.405 mmol, 81% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.62 (d, J = 7.7 Hz, 2H), 7.23 (t, J = 7.5 Hz, 2H), 7.13-7.06 (m, 4H), 7.01 (s, 4H), 4.07 (t, J = 7.6 Hz, 1H), 2.96 (d, J = 7.6 Hz, 2H), 2.25 (s, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 147.0, 140.9, 136.8, 135.9, 129.5, 129.0, 127.1, 126.7, 125.0, 119.9, 48.9, 39.8, 21.2 ppm.



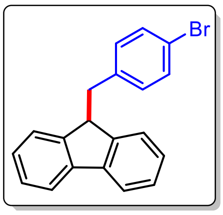
9-(4-methoxybenzyl)-9H-fluorene (Compound-7c):[13] Following the general procedure 3(d), the titled compound was isolated as white solid (120.2 mg, 0.420 mmol, 84% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.87 (d, J = 7.7 Hz, 2H), 7.49 (t, J = 7.4 Hz, 2H), 7.40-7.32 (m, 4H), 7.25 (d, J = 6.2 Hz, 2H), 6.98 (d, J = 8.4 Hz, 2H), 4.31 (t, J = 7.7 Hz, 1H), 3.91 (s, 3H), 3.19 (d, J = 7.7 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 158.1, 146.9, 140.9, 131.8, 130.5, 127.1, 126.6, 124.9, 119.8, 113.7, 55.2, 48.9, 39.1 ppm.



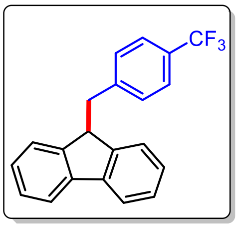
9-(4-fluorobenzyl)-9H-fluorene (Compound-7d):[13] Following the general procedure 3(d), the titled compound was isolated as white solid (94.6 mg, 0.345 mmol, 69% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.58 (d, J = 7.5 Hz, 2H), 7.20 (t, J = 7.5 Hz, 2H), 7.11-7.03 (m, 4H), 6.95 (t, J = 6.2 Hz, 2H), 6.81 (t, J = 8.09, 2H), 4.02 (t, J = 7.5 Hz, 1H), 2.94 (d, J = 7.5 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 162.9, 160.5, 146.6, 141.0, 135.3, 135.3, 131.0, 130.9, 127.3, 126.8, 124.9, 120.0, 115.2, 115.0, 48.8, 39.2 ppm. ^{19}F NMR (471 MHz, CDCl$_3$) δ -116.79 ppm.



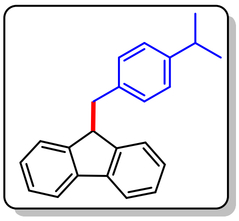
9-(4-bromobenzyl)-9H-fluorene (Compound-7e):[13] Following the general procedure 3(d), the titled compound was isolated as white solid (119.0 mg, 0.355 mmol, 71% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.73 (d, J = 10.8 Hz, 2H), 7.40-7.34 (m, 4H), 7.24-7.20 (m, 4H), 7.04 (d, J = 10.6 Hz, 2H), 4.20 (t, J = 7.8 Hz, 1H), 3.10 (d, J = 7.8 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 146.3, 140.9, 138.5, 131.3, 127.3, 126.7, 124.7, 120.2, 48.4, 39.3 ppm.



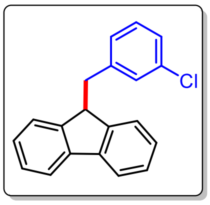
9-(4-(trifluoromethyl)benzyl)-9H-fluorene (Compound-7f):[13] Following the general procedure 3(d), the titled compound was isolated as white solid (108.6 mg, 0.355 mmol, 67% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.63 (d, J = 7.7 Hz, 2H), 7.42 (d, J = 7.9 Hz, 2H), 7.28-7.24 (m, 1H), 7.18-7.08 (m, 7H), 4.14 (t, J = 7.4 Hz, 1H), 3.09 (d, J = 7.6 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 146.3, 143.8, 141.0, 129.9, 127.5, 127.4, 126.9, 126.9, 125.3, 125.3, 125.2, 125.2, 124.9, 124.8, 120.1, 120.0, 48.5, 39.8 ppm. ^{19}F NMR (471 MHz, CDCl$_3$) δ -63.25 ppm.



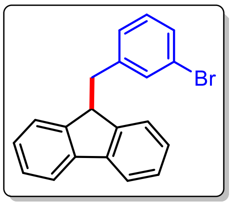
9-(4-isopropylbenzyl)-9H-fluorene (Compound-7g):[14] Following the general procedure 3(d), the titled compound was isolated as white solid (116.3 mg, 0.390 mmol, 78% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81 (d, J = 7.5 Hz, 2H), 7.42 (t, J = 7.4 Hz, 2H), 7.31-7.24 (m, 8H), 4.28 (t, J = 7.7 Hz, 1H), 3.14 (d, J = 7.7 Hz, 2H), 3.05-2.98 (m, 1H), 1.37 (d, J = 7.0 Hz, 6H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 147.1, 147.0, 140.9, 137.3, 129.5, 127.2, 126.7, 126.5, 125.0, 119.9, 48.9, 39.8, 33.9, 24.1 ppm.



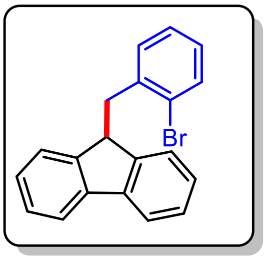
9-(3-chlorobenzyl)-9H-fluorene (Compound-7h):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (95.9 mg, 0.330 mmol, 66% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.75 (d, J = 7.9 Hz, 2H), 7.37 (t, J = 7.9 Hz, 2H), 7.26-7.19 (m, 7H), 7.09-7.07 (m, 1H), 4.22 (t, J = 7.7 Hz, 1H), 3.10 (d, J = 7.5 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 146.5, 141.9, 141.0, 134.1, 129.7, 129.6, 127.9, 127.4, 126.9, 126.7, 124.9, 120.0, 48.5, 39.8 ppm.

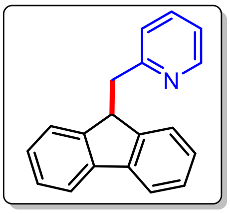


9-(3-bromobenzyl)-9H-fluorene (Compound-7i):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (113.9 mg, 0.340 mmol, 68% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.64 (d, J = 7.7 Hz, 2H), 7.29-7.24 (m, 3H), 7.16-7.12 (m, 3H), 7.09-6.99 (m, 4H), 4.10 (t, J = 7.6 Hz, 1H), 2.98 (d, J = 8.2 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 146.4, 142.2, 140.9, 132.6, 129.9, 129.6, 128.3, 127.4, 126.9, 124.9, 122.4, 120.1, 48.5, 39.8 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{21}H_{15}BrI$ 474.9384; Found 474.9370.

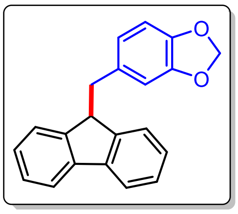


9-(2-bromoobenzyl)-9H-fluorene (Compound-7j): Following the general procedure 3(d), the titled compound was isolated as white solid (102.2 mg, 0.305 mmol, 61% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (d, J = 7.9 Hz, 2H), 7.38 (t, J = 7.9 Hz, 2H), 7.28-7.21 (m, 7H), 7.10 (s, 1H), 4.24 (t, J = 7.7 Hz, 1H), 3.12 (d, J = 7.5 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 146.5, 142.0, 141.0, 134.1, 129.7, 129.6, 127.9, 127.4, 127.0, 126.7, 125.0, 120.0, 48.5, 39.8 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{20}H_{16}Br$ 337.0417; Found 337.0401.

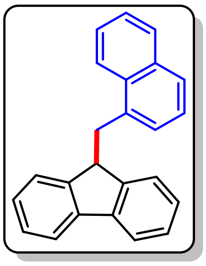




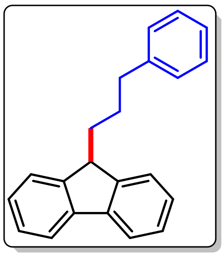
2-((9H-fluoren-9-yl)methyl)pyridine (Compound-7k):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (92.6 mg, 0.360 mmol, 72% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.55 (s, 1H), 7.60 (d, *J* = 7.8 Hz, 2H), 7.41 (t, *J* = 7.8 Hz, 1H), 7.20 (t, *J* = 7.7 Hz, 2H), 7.06 (t, *J* = 7.3 Hz, 3H), 6.94 (d, *J* = 7.7 Hz, 2H), 6.84 (d, *J* = 10.3 Hz, 1H), 4.51 (t, *J* = 7.8 Hz, 1H), 3.08 (d, *J* = 7.7 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 159.8, 149.5, 147.1, 140.7, 136.2, 127.1, 126.8, 124.7, 124.4, 121.7, 119.9, 47.2, 42.5 ppm.



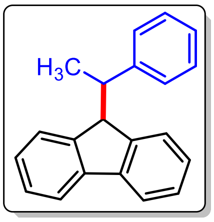
5-((9H-fluoren-9-yl)methyl)benzo[d][1,3]dioxole (Compound-7l):[8] Following the general procedure 3(d), the titled compound was isolated as white solid (114.1 mg, 0.380 mmol, 76% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.76 (d, *J* = 7.6 Hz, 2H), 7.39 (t, *J* = 7.2 Hz, 2H), 7.30-7.24 (m, 4H), 6.77 (d, *J* = 6.7 Hz, 2H), 6.65 (d, *J* = 7.9 Hz, 1H), 5.93 (s, 2H), 4.18 (t, *J* = 7.5 Hz, 1H), 3.05 (d, *J* = 7.6 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 147.5, 146.7, 146.0, 140.9, 133.5, 127.2, 126.7, 124.9, 122.6, 119.8, 109.7, 108.0, 100.9, 49.1, 39.6 ppm.



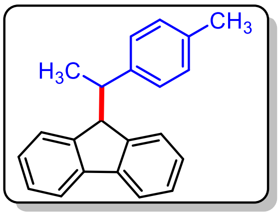
9-(naphthalen-1-ylmethyl)-9H-fluorene (Compound-7m):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (121.0 mg, 0.395 mmol, 79% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.37-24 (d, *J* = 8.3 Hz, 1H), 8.0 (d, *J* = 7.9 Hz, 1H), 7.90 (d, *J* = 8.3 Hz, 1H), 7.83 (d, *J* = 7.7 Hz, 2H), 7.65-7.57 (m, 2H), 7.50 (t, *J* = 7.6 Hz, 1H), 7.41 (t, *J* = 7.5 Hz, 2H), 7.32 (d, *J* = 7.0 Hz, 1H), 7.23 (d, *J* = 7.4 Hz, 2H), 7.14 (d, *J* = 7.6 Hz, 2H), 4.31 (t, *J* = 7.9 Hz, 1H), 3.37 (d, *J* = 7.9 Hz, 2H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 147.1, 140.8, 136.1, 134.1, 132.1, 129.2, 128.3, 127.6, 127.3, 126.7, 126.2, 125.8, 125.4, 125.2, 123.8, 119.9, 47.7, 38.1 ppm.



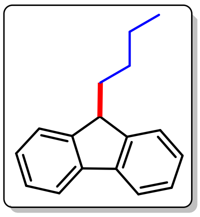
9-(3-phenylpropyl)-9H-fluorene (Compound-7n):[14] Following the general procedure 3(d), the titled compound was isolated as white solid (100.9 mg, 0. 355mmol, 71% yield). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.88 (d, *J* = 7.6 Hz, 2H), 7.61 (d, *J* = 7.5 Hz, 2H), 7.51-7.48 (m, 2H), 7.45-7.42 (m, 2H), 7.39-7.37 (m, 2H), 7.31-7.30 (m, 1H), 7.23 (d, *J* = 4.1 Hz, 2H), 4.13-4.10 (m, 1H), 2.70 (t, *J* = 7.7 Hz, 2H), 2.21-2.17 (m, 2H), 1.67-1.63 (m, 2H) ppm. $^{13}C\{^1H\}$ NMR (126 MHz, $CDCl_3$) δ 147.4, 142.3, 141.3, 128.5, 128.3, 127.0, 126.9, 125.8, 124.4, 119.9, 47.4, 36.2, 32.6, 27.3 ppm.



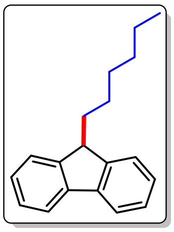
9-(1-phenylethyl)-9H-fluorene (Compound-7o):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (101.3 mg, 0.375 mmol, 75% yield).^1H NMR (500 MHz, $CDCl_3$) δ 7.72-7.68 (m, 7.4 Hz, 2H), 7.47 (d, J = 7.8 Hz, 1H), 7.37-7.24 (m, 8H), 7.10 (t, J = 7.5 Hz, 1H), 6.82 (d, J = 7.6 Hz, 1H), 4.29 (d, J = 4.7 Hz, 1H), 3.67- 3.63 (m, 1H), 0.93 (d, J = 7.2 Hz, 3H) ppm. $^{13}C\{^1H\}$ NMR (126 MHz, $CDCl_3$) δ 146.7, 144.8, 142.0, 141.5, 128.3, 128.2, 127.2, 127.2, 126.9, 126.4, 126.4, 125.8, 124.4, 119.8, 119.8, 54.4, 42.1, 14.2 ppm.



9-(1-(p-tolyl)ethyl)-9H-fluorene (Compound-7p):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (110.9 mg, 0.390 mmol, 78% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.61 (t, J = 8.2 Hz, 2H), 7.39 (d, J = 7.5 Hz, 1H), 7.26 (t, J = 7.2 Hz, 1H), 7.20 (t, J = 7.3 Hz, 2H), 7.11 (d, J = 7.5 Hz, 2H), 6.99-7.06 (m, 3H), 6.75 (d, J = 7.4 Hz, 1H), 4.18 (s, 1H), 3.58-3.51 (m, 1H), 2.27 (s, 3H), 0.78 (d, J = 11.5 Hz, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 146.8, 144.8, 141.9, 141.7, 141.5, 135.8, 129.0, 128.0, 127.1, 127.1, 126.9, 126.4, 125.8, 124.4, 119.8, 119.7, 54.4, 41.6, 21.2, 14.0 ppm.

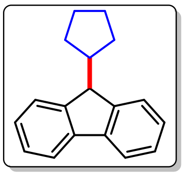


9-butyl-9H-fluorene (Compound-7q): Following the general procedure 3(d), the titled compound was isolated as white solid (81.1 mg, 0.365 mmol, 73% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.86 (d, J = 4.4 Hz, 2H), 7.62 (d, J = 5.5 Hz, 2H), 7.48-7.41 (m, 4H), 4.08 (s, 1H), 2.12 (s, 2H), 1.39 (s, 2H), 1.29 (s, 2H), 0.94 (s, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 147.7, 141.2, 126.9, 126.9, 124.4, 119.9, 47.5, 32.9, 27.9, 23.1, 14.0 ppm. HRMS (ESI) *m/z*: $[M + H]^+$: Calcd. for $C_{17}H_{19}$ 223.1487; Found 223.1482.

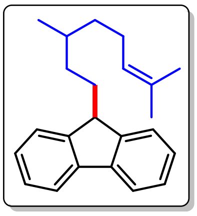


9-hexyl-9H-fluorene (Compound-7r):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (85.1 mg, 0.340 mmol, 68% yield). ^1H NMR (400 MHz, $CDCl_3$) δ 7.74 (d, J = 7.5 Hz, 2H), 7.50 (d, J = 7.2 Hz, 2H), 7.38-7.27 (m, 4H), 3.95 (t, J = 6.0 Hz, 1H), 2.01-1.95 (m, 2H), 1.26-1.14 (m, 8H), 0.82 (t, J = 6.1 Hz, 3H) ppm. $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 147.8, 141.2, 126.9, 126.9, 124.5, 119.9, 47.6, 33.2, 31.8, 29.8, 25.8, 22.8, 14.2 ppm.

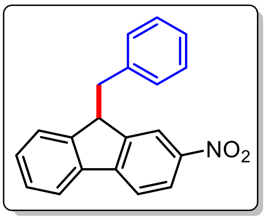
9-cyclopentyl-9H-fluorene (Compound-7s):[2] Following the general procedure 3(d), the titled compound was isolated as white solid (77.3 mg, 0.330 mmol, 66% yield). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.97-7.90 (m, 2H), 7.76-7.72 (m, 2H), 7.56-7.50 (m, 2H), 7.48-7.42 (m, 2H), 4.18 (t, J = 5.4 Hz, 1H), 2.55 (s, 1H), 1.95 (s, 2H), 1.76-1.56 (m, 6H) ppm. ^{13}C{^{1}H} NMR (126 MHz, $CDCl_3$) δ 147.2, 141.5, 126.9, 126.6, 125.2, 119.7, 51.3, 44.5, 30.1, 25.3 ppm.



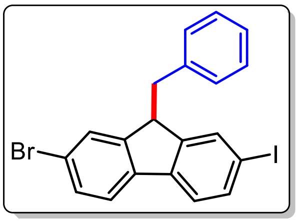
9-(3,7-dimethyloct-6-en-1-yl)-9H-fluorene (Compound-7t):[14] Following the general procedure 3d, the titled compound was isolated as white solid (94.3 mg, 0.310 mmol, 62% yield). ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.89-7.87 (m, 2H), 7.66-7.64 (m, 2H), 7.51-7.42 (m, 4H), 5.23-5.22 (m, 1H), 4.11-4.10 (m, 1H), 2.19-2.13 (m, 2H), 2.07-1.98 (m, 2H), 1.86-1.82 (m, 3H), 1.78 (d, J = 4.0 Hz, 1H), 1.72 (d, J = 4.1 Hz, 3H), 1.56-1.50 (m, 1H), 1.47-1.40 (m, 1H), 1.37-1.32 (m, 1H), 1.19-1.13 (m, 1H), 0.99-0.97 (m, 3H) ppm. ^{13}C{^{1}H} NMR (126 MHz, $CDCl_3$) δ 147.7, 147.7, 141.3, 131.0, 126.9, 126.9, 125.1, 124.4 124.4, 119.9, 47.7, 36.9, 32.7, 32.5, 30.4, 25.8, 25.6, 19.6, 17.7 ppm.



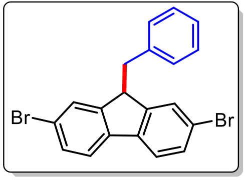
9-benzyl-2-nitro-9H-fluorene (Compound-7u): Following the general procedure 3d, the titled compound was synthesised (61% NMR conversion). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.49-7.42 (m, 2H), 7.25-7.13 (m, 8H, merged with starting material), 7.01 (m, 2H), 4.03 (t, J = 5.4 Hz, 1H), 3.06-2.88 (m, 2H) ppm. ^{13}C{^{1}H} NMR (126 MHz, $CDCl_3$) δ 147.6, 146.0, 141.5, 140.2, 139.5, 131.1, 129.7, 128.7, 128.4, 127.7, 127.4, 127.1, 126.4, 124.9, 124.6, 120.7, 118.5, 112.4, 109.3, 48.7, 48.5, 40.5 ppm.



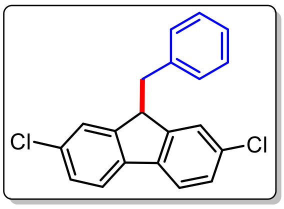
9-benzyl-2-bromo-7-iodo-9H-fluorene (Compound-7v): Following the general procedure 3(d), the titled compound was isolated as white solid (170.6 mg, 0.371 mmol, 74% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.46 (d, J = 7.9 Hz, 1H), 7.27 (s, 2H), 7.19-7.13 (m, 5H), 7.04 (s, 1H), 6.96 (d, J = 8.2 Hz, 2H), 3.83 (t, J = 7.6 Hz, 1H), 2.84-2.80 (m, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 148.6, 148.2, 139.3, 138.8, 138.6, 136.3, 134.1, 130.5, 129.5, 128.5, 128.2, 126.8, 121.5, 121.2, 121.1, 92.4, 48.6, 39.6 ppm. HRMS (ESI) m/z: [M + H]$^+$: Calcd. for $C_{20}H_{15}BrI$ 462.9383; Found 462.9378.



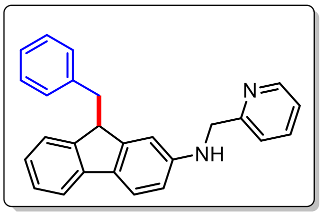
9-benzyl-2,7-dibromo-9H-fluorene (Compound-7w): Following the general procedure 3(d), the titled compound was isolated as white solid (151.1 mg, 0.365 mmol, 73% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.44-7.36 (m, 4H), 7.25-7.16 (m, 5H), 7.07 (d, J = 7.6 Hz, 2H), 4.07-4.01 (m, 1H), 2.96 (d, J = 7.8 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 148.5, 138.9, 138.7, 130.6, 129.5, 128.6, 128.4, 127.0, 121.3, 121.0, 48.8, 39.8 ppm. HRMS (ESI) *m/z*: [M + H]$^+$: Calcd. for $C_{20}H_{15}Br_2$ 414.9521; Found 414.9518.



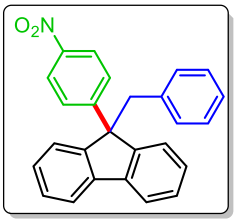
9-benzyl-2,7-dichloro-9H-fluorene (Compound-7x):[14] Following the general procedure 3(d), the titled compound was isolated as white solid (123.5 mg, 0.380 mmol, 76% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.44-7.36 (m, 4H), 7.25-7.19 (m, 3H), 7.16 (s, 2H), 7.07 (d, J = 7.2 Hz, 2H), 4.07-4.01 (m, 1H), 2.97 (d, J = 7.5 Hz, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 148.3, 138.7, 138.4, 132.8, 129.5, 128.5, 127.7, 126.9, 125.3, 120.8, 48.7, 39.7 ppm.



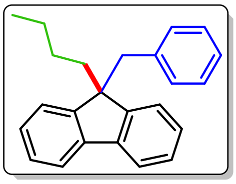
9-benzyl-N-(pyridin-2-ylmethyl)-9H-fluoren-2-amine (Compound-7y): Following the general procedure 3(d), the titled compound was isolated as yellow liquid (114.1 mg, 0.315 mmol, 63% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.44 (s, 1H), 7.42-7.28 (m, 3H), 7.07-7.03 (m, 7H), 6.97-6.90 (m, 3H), 6.45 (d, J = 7.2 Hz, 1H), 6.26 (s, 1H), 5.42 (br., 1H), 4.21 (s, 2H), 3.92 (t, J = 7.6 Hz, 1H), 2.88-2.86 (m, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 158.4, 148.9, 148.7, 147.2, 145.8, 141.4, 140.0, 136.8, 131.0, 129.9, 129.6, 128.3, 128.2, 127.0, 126.2, 124.7, 124.5, 122.2, 121.7, 120.6, 118.4, 112.4, 109.3, 49.1, 48.5, 40.2 ppm. HRMS (ESI) *m/z*: [M + H]$^+$: Calcd. for $C_{26}H_{23}N_2$ 363.1861; Found 363.2071.



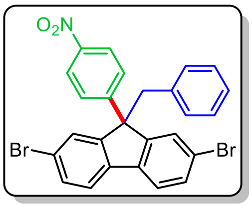
9-benzyl-9-(4-nitrophenyl)-9H-fluorene (Compound-8a): Following the general procedure, the titled compound was isolated as white solid (145.3 mg, 0.384 mmol, 77 % yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 (d, J = 8.9 Hz, 2H), 7.55-7.53 (m, 2H), 7.47 (d, J = 9.0 Hz, 2H), 7.32-7.31 (m, 6H), 6.93 (t, J = 7.4 Hz, 1H), 6.83 (t, J = 7.8 Hz, 2H), 6.47 (d, J = 7.1 Hz, 2H), 3.85 (s, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 152.8, 149.5, 146.7, 140.9, 135.5, 130.0, 128.0, 127.9, 127.6, 127.0, 126.1, 124.9, 123.7, 120.2, 60.1, 43.6 ppm. HRMS (ESI) *m/z*: [M + H]$^+$: Calcd. for $C_{26}H_{20}NO_2$ 378.1494; Found 378.1488.



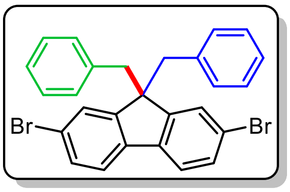
9-benzyl-9-butyl-9H-fluorene (Compound-8b):[2] Following the general procedure, the titled compound was isolated as white solid (114.0 mg, 0.365 mmol, 73% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.43 (s, 2H), 7.17 (d, J = 8.9 Hz, 6H), 6.87-6.81 (m, 3H), 6.55 (d, J = 7.5 Hz, 2H), 3.04 (s, 2H), 2.06-2.02 (m, 2H), 1.03-0.97 (m, 2H), 0.56 (t, J = 7.4 Hz, 3H), 0.49-0.52 (m, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 149.5, 141.0, 137.4, 130.4, 127.1, 127.0, 126.7, 125.9, 123.8, 119.8, 55.9, 47.0, 38.7, 26.1, 23.2, 13.8 ppm. HRMS (ESI) *m/z*: [M + H]$^+$: Calcd. for $C_{24}H_{25}$ 313.1956; Found 313.1947.



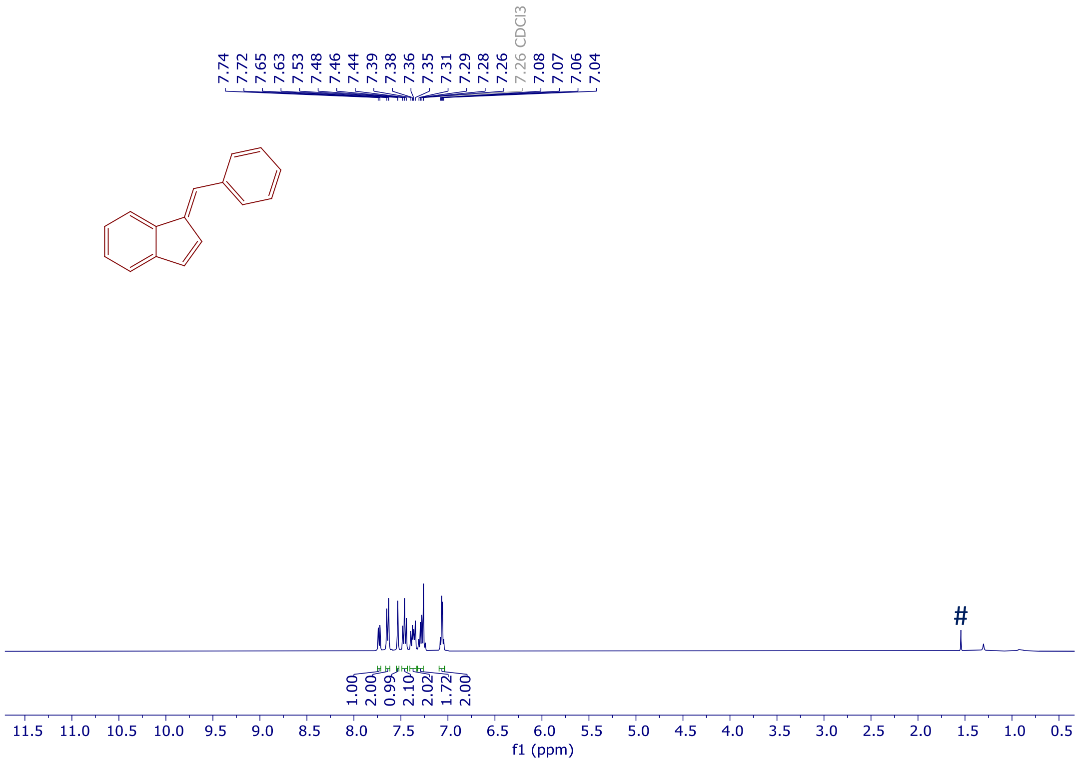
9-benzyl-2,7-dibromo-9-(4-nitrophenyl)-9H-fluorene (Compound-8c):[2] Following the general procedure, the titled compound was isolated as white solid (184.6 mg, 0.345 mmol, 69% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 8.16 (d, J = 8.7 Hz, 2H), 7.44-7.32 (m, 8H), 6.97-6.84 (m, 3H), 6.44 (d, J = 7.7 Hz, 2H), 3.80 (s, 2H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 151.4, 150.7, 147.1, 138.9, 134.7, 132.7, 131.4, 130.0, 128.2, 128.0, 127.4, 126.5, 125.1, 124.1, 121.7, 121.6, 60.2, 43.6 ppm. HRMS (ESI) *m/z*: [M + H]$^+$: Calcd. for $C_{26}H_{18}Br_2NO_2$ 535.9686; Found 535.9678.



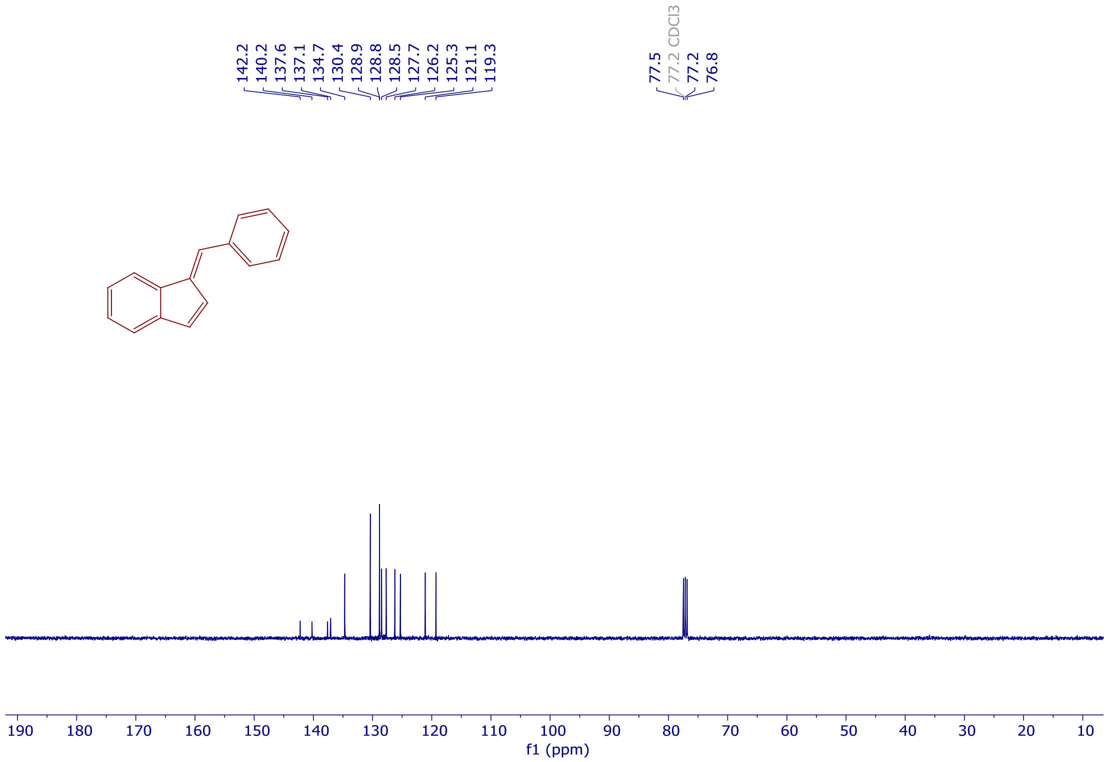
9,9-dibenzyl-2,7-dibromo-9H-fluorene (Compound-8d):[3] Following the general procedure, the titled compound was isolated as white solid (179.0 mg, 0.355 mmol, 71% yield). ^{1}H NMR (400 MHz, CDCl$_3$) δ 7.54 (s, 2H), 7.35-7.33 (d, J = 8.1 Hz, 2H), 7.17-7.15 (m, 2H), 7.02-7.00 (m, 6H), 6.71-6.69 (m, , 3.27 (s, 4H) ppm. ^{13}C{^{1}H} NMR (101 MHz, CDCl$_3$) δ 150.3, 138.9, 136.2, 130.6, 130.3, 128.0, 127.5, 126.4, 121.2, 120.7, 57.2, 45.2 ppm.



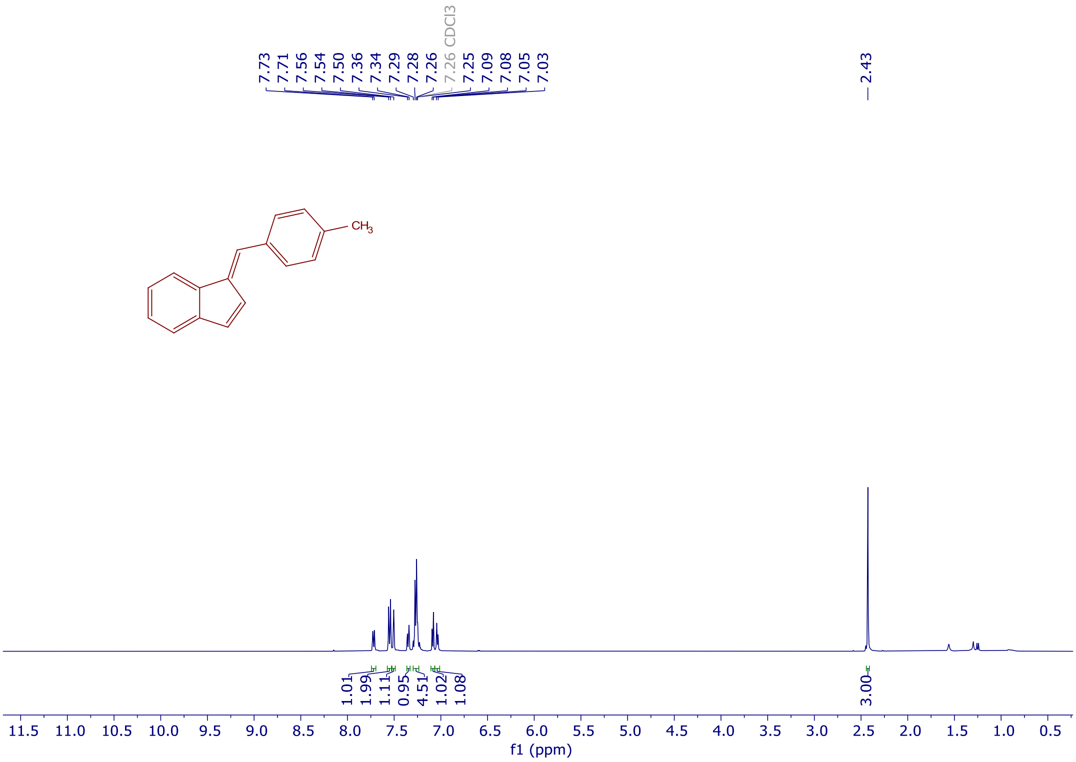
16. ^{1}H and ^{13}C{^{1}H} spectra of the isolated compounds:



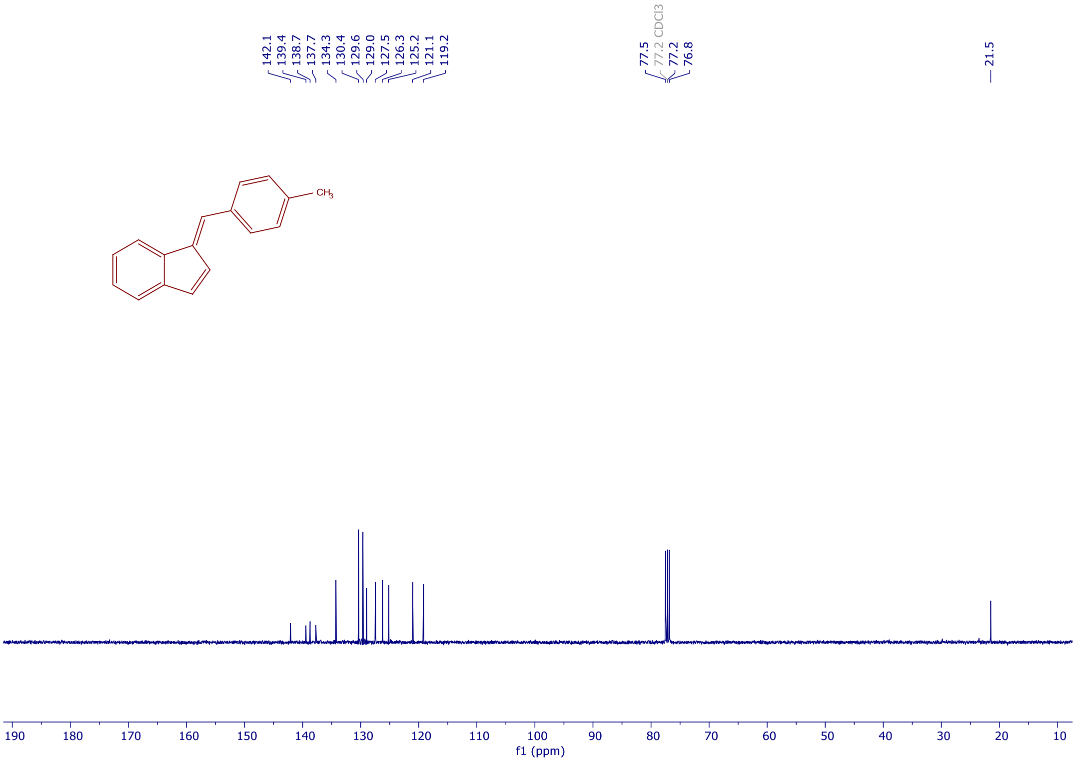
*^{1}H NMR of (E)-1-benzylidene-1H-indene (Compound-**3a**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



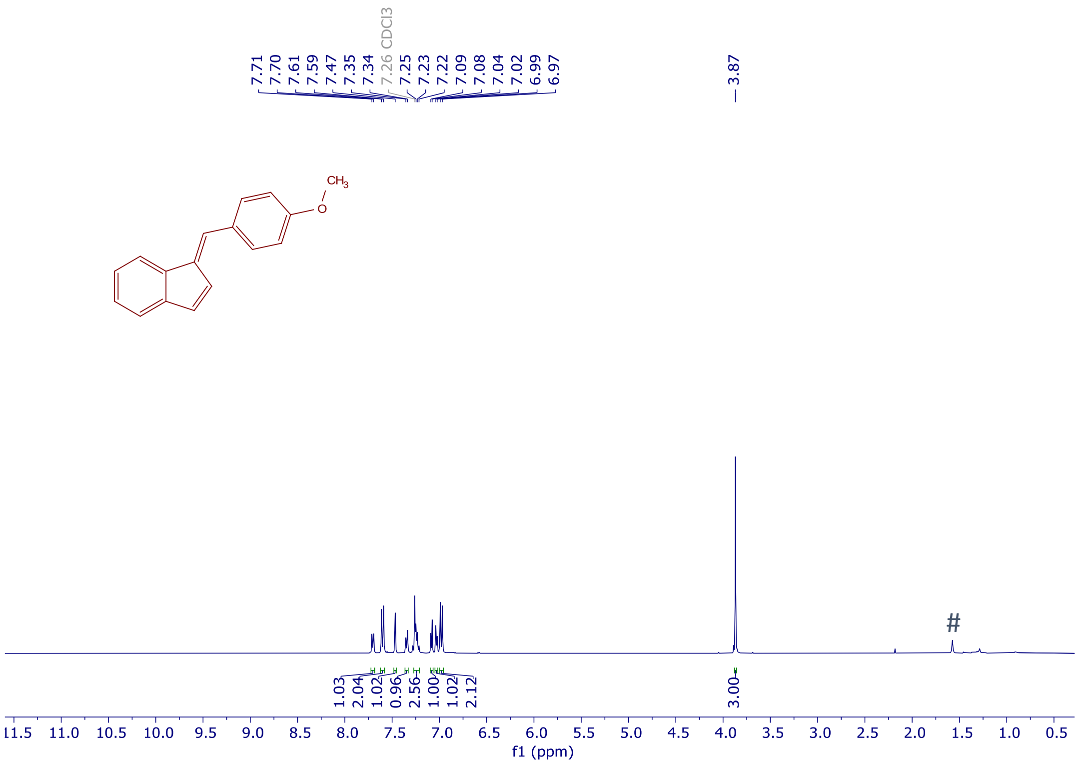
*^{13}C{^{1}H} NMR of (E)-1-benzylidene-1H-indene (Compound-**3a**) in $CDCl_3$*



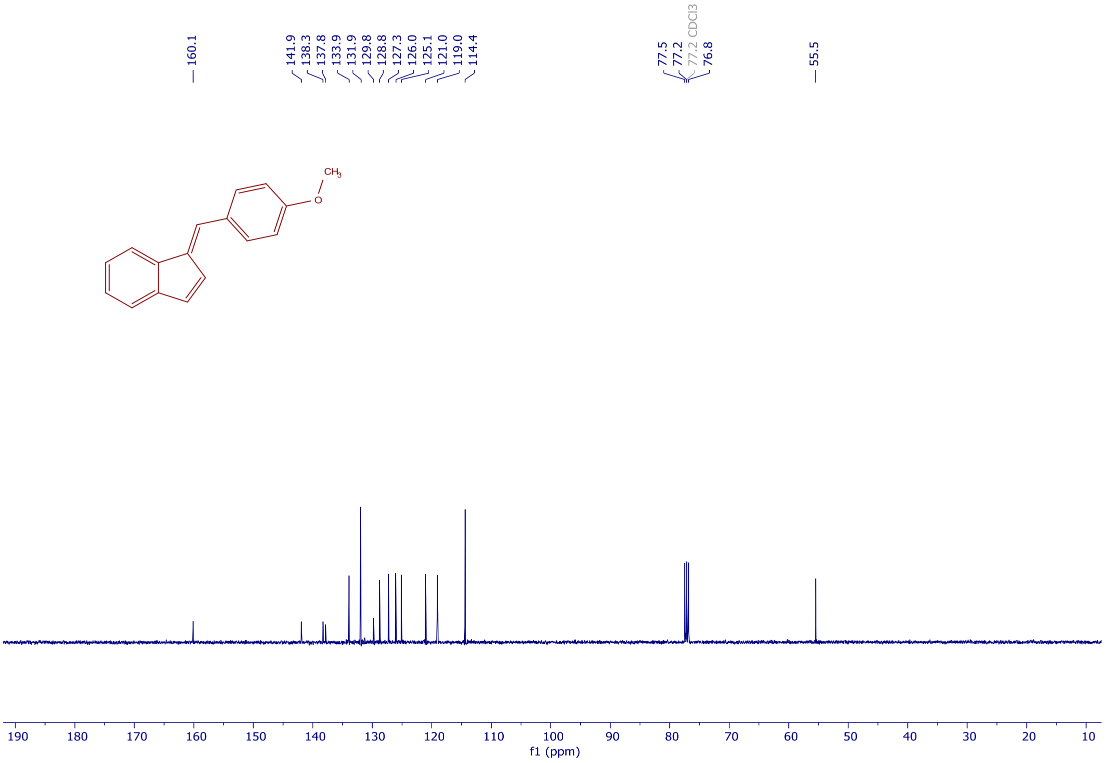
*^{1}H NMR of (E)-1-(4-methylbenzylidene)-1H-indene(Compound-**3b**) in $CDCl_3$*



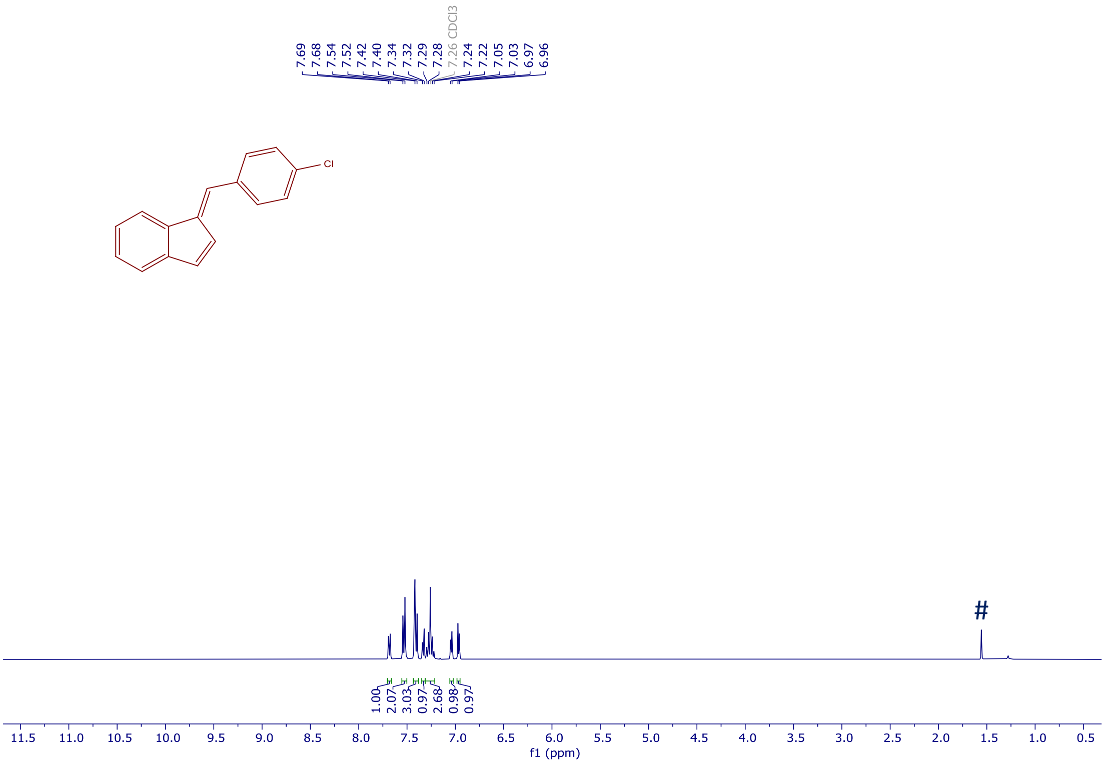
*$^{13}C\{^{1}H\}$ NMR of (E)-1-(4-methylbenzylidene)-1H-indene (Compound-**3b**) in $CDCl_3$*



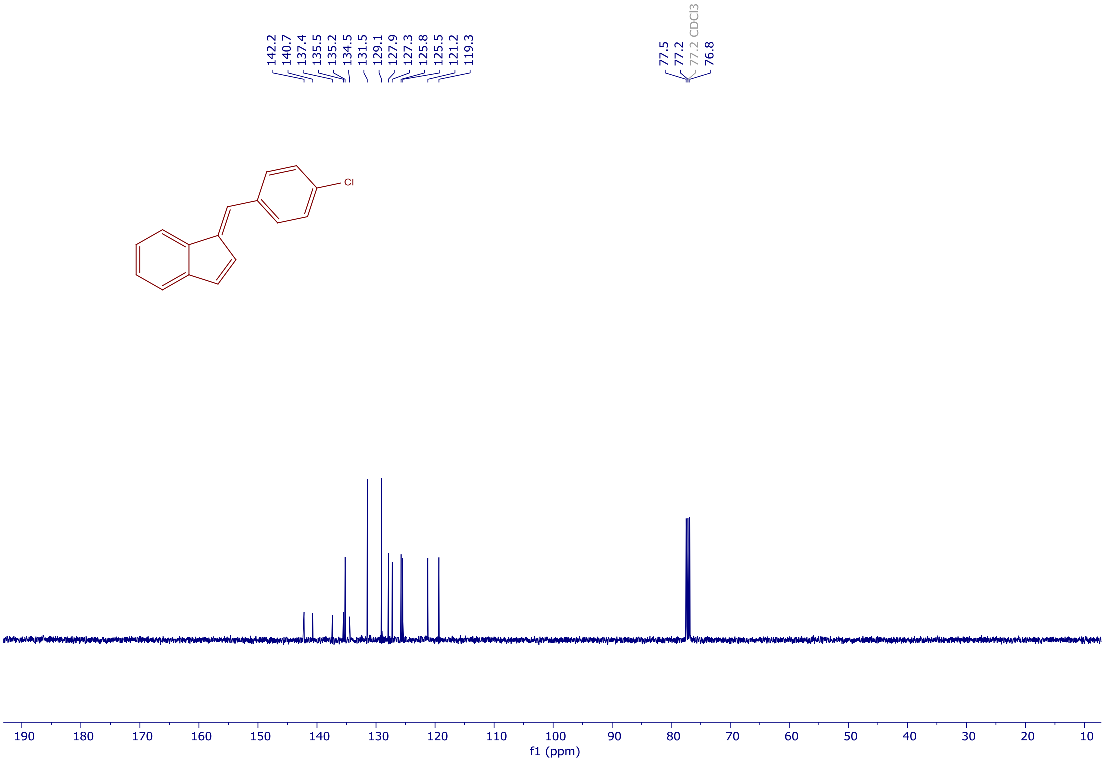
^{1}H NMR of (E)-1-(4-methoxybenzylidene)-1H-indene (Compound-**3c**) in CDCl$_3$. # indicates the solvent impurity of H$_2$O in CDCl$_3$



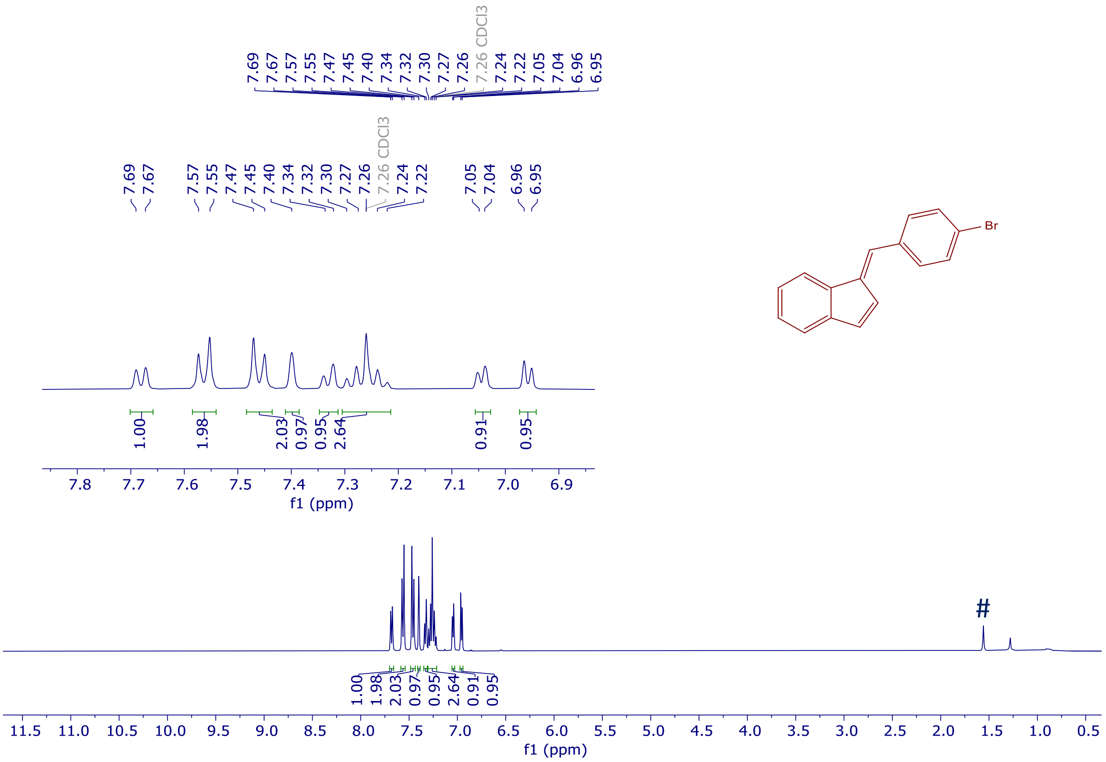
^{13}C{^{1}H} NMR of (E)-1-(4-methoxybenzylidene)-1H-indene (Compound-**3c**) in CDCl$_3$



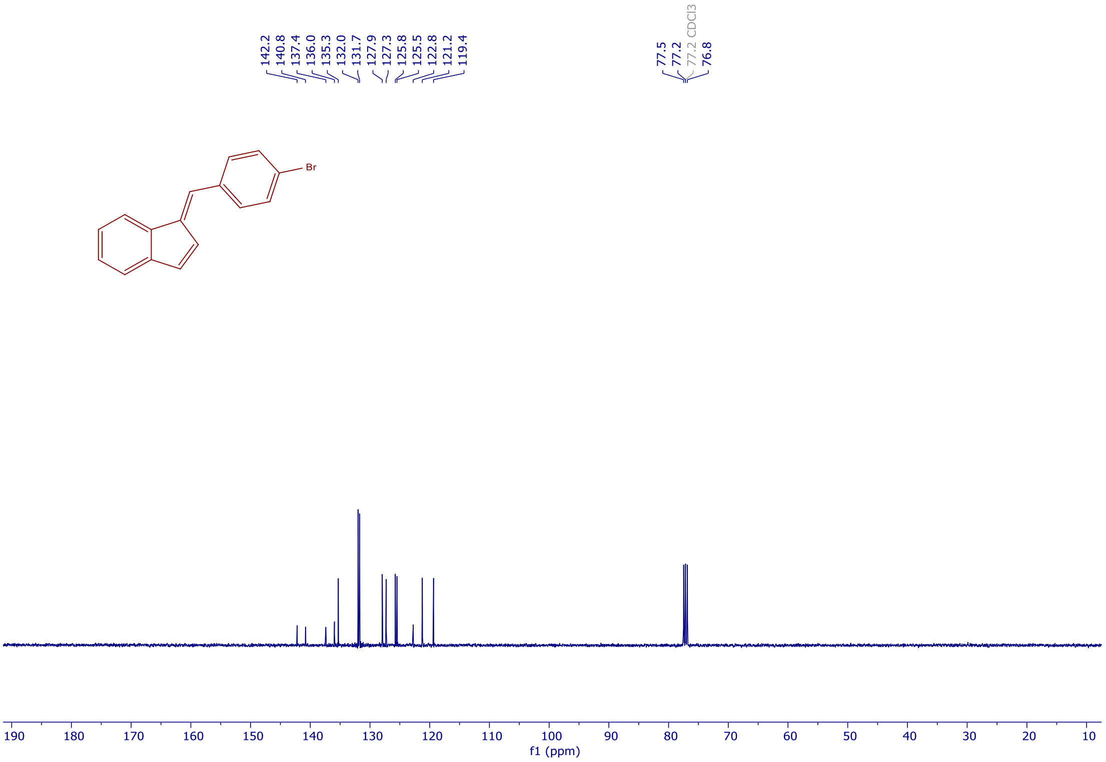
*^{1}H NMR of (E)-1-(4-chlorobenzylidene)-1H-indene (Compound-**3d**) in CDCl$_3$. # indicates the solvent impurity of H_2O in CDCl$_3$*



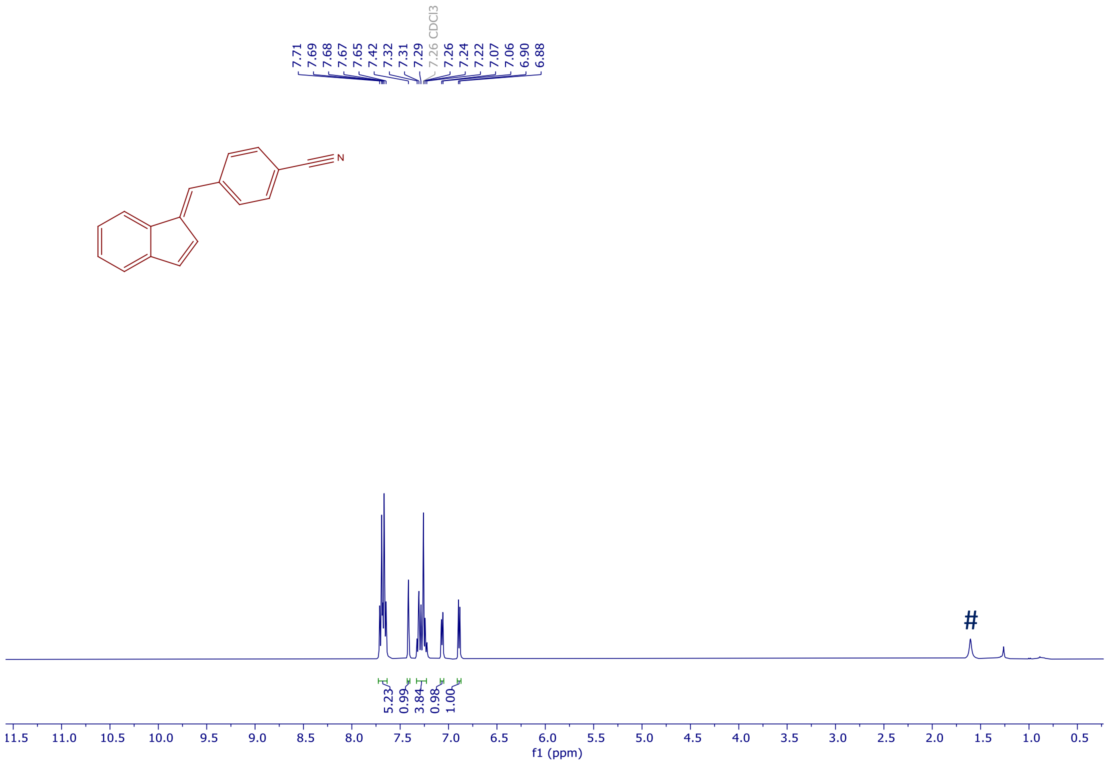
*^{13}C{^{1}H} NMR of (E)-1-(4-chlorobenzylidene)-1H-indene (Compound-**3d**) in CDCl$_3$*



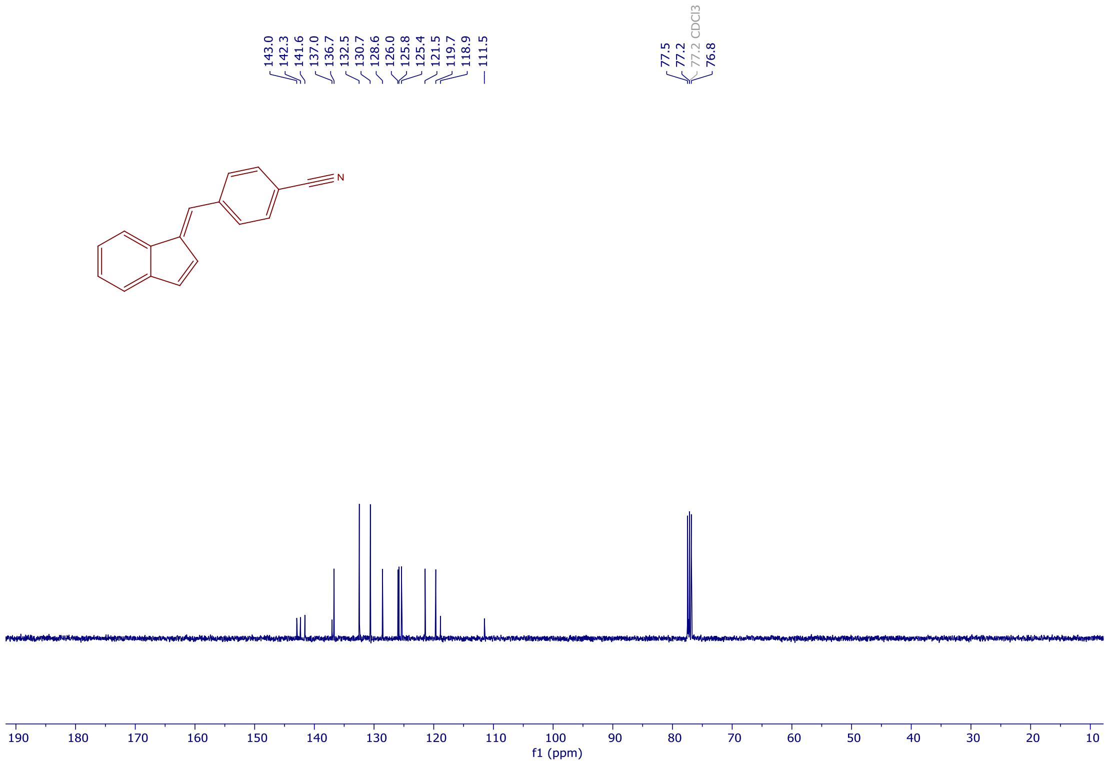
*^{1}H NMR of (E)-1-(4-bromobenzylidene)-1H-indene (Compound-**3e**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



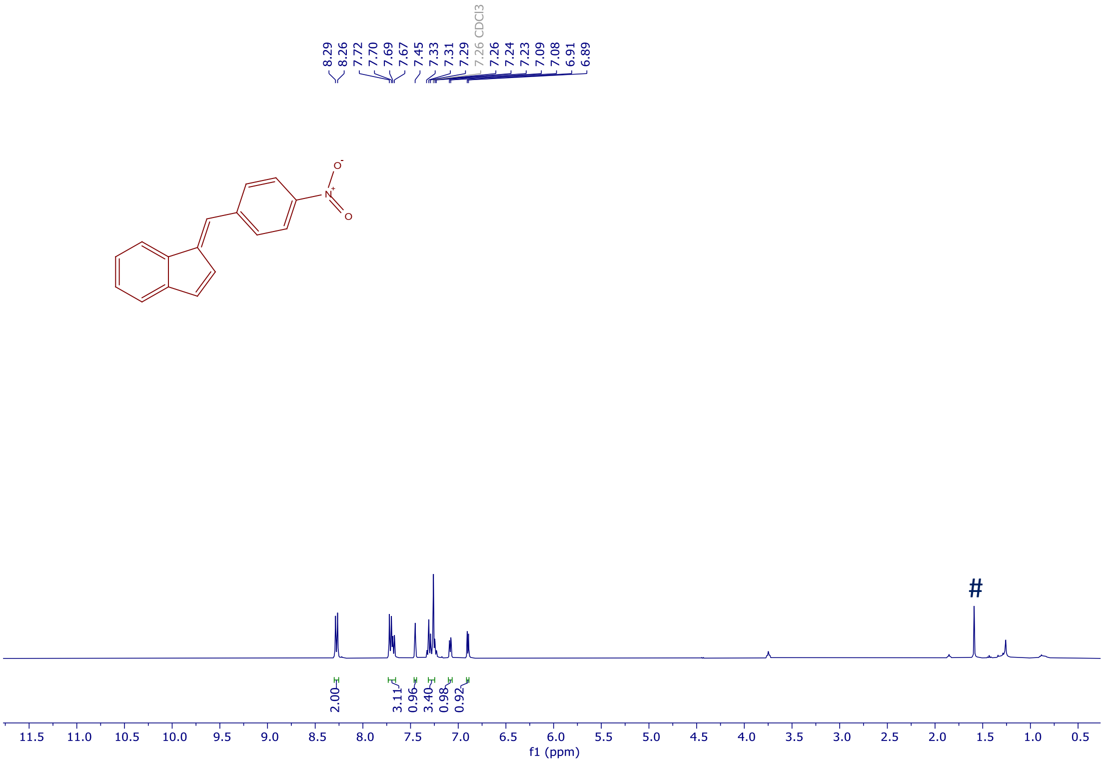
*$^{13}C\{^1H\}$ NMR of (E)-1-(4-bromobenzylidene)-1H-indene (Compound-**3e**) in $CDCl_3$*



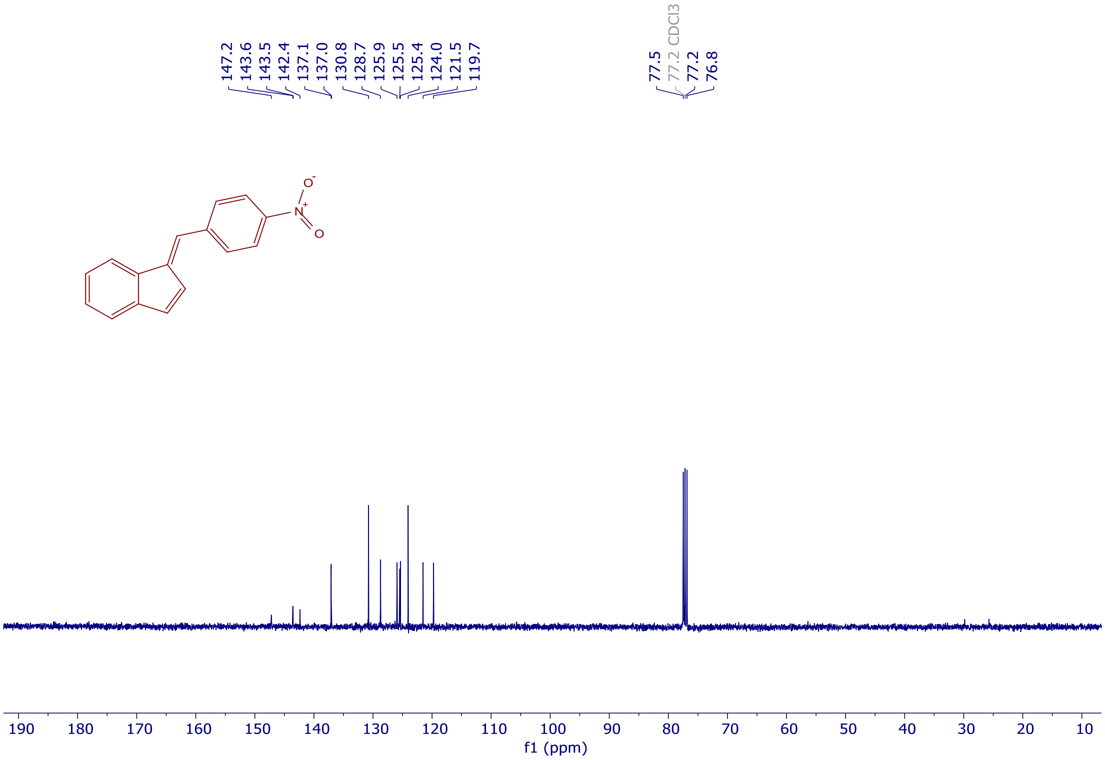
*^{1}H NMR of ((E)-4-((1H-inden-1-ylidene)methyl)benzonitrile (Compound-**3f**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



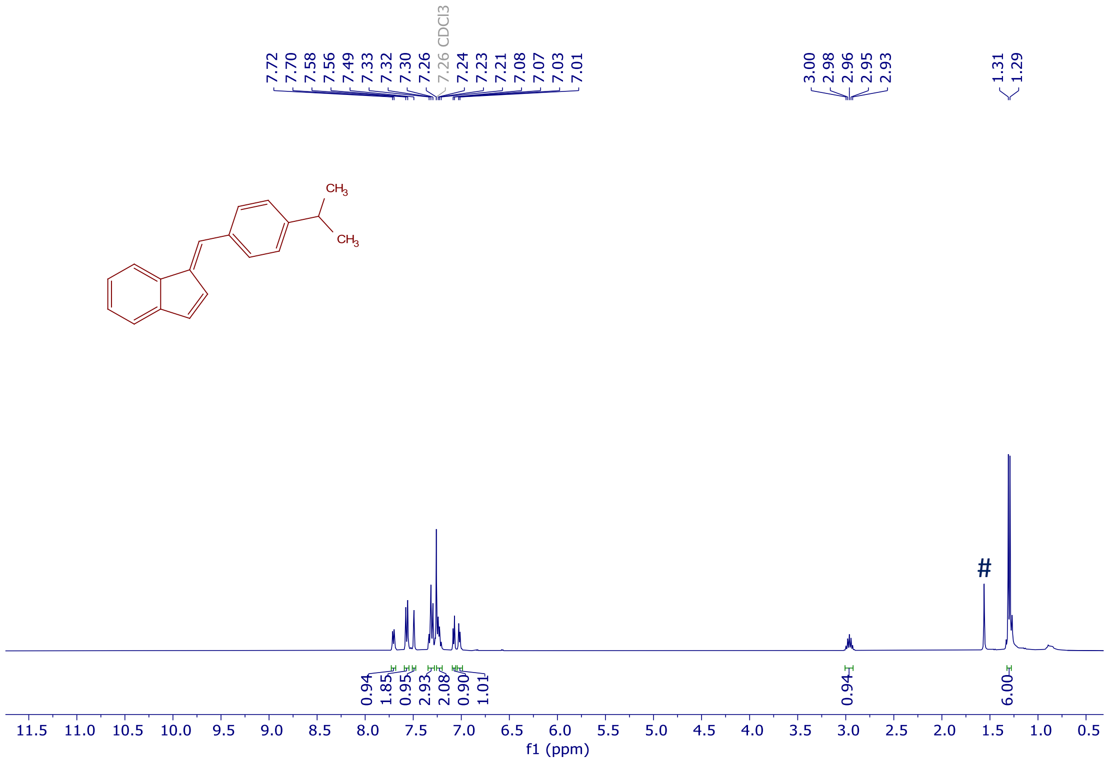
*$^{13}C\{^1H\}$ NMR of ((E)-4-((1H-inden-1-ylidene)methyl)benzonitrile (Compound-**3f**) in $CDCl_3$*



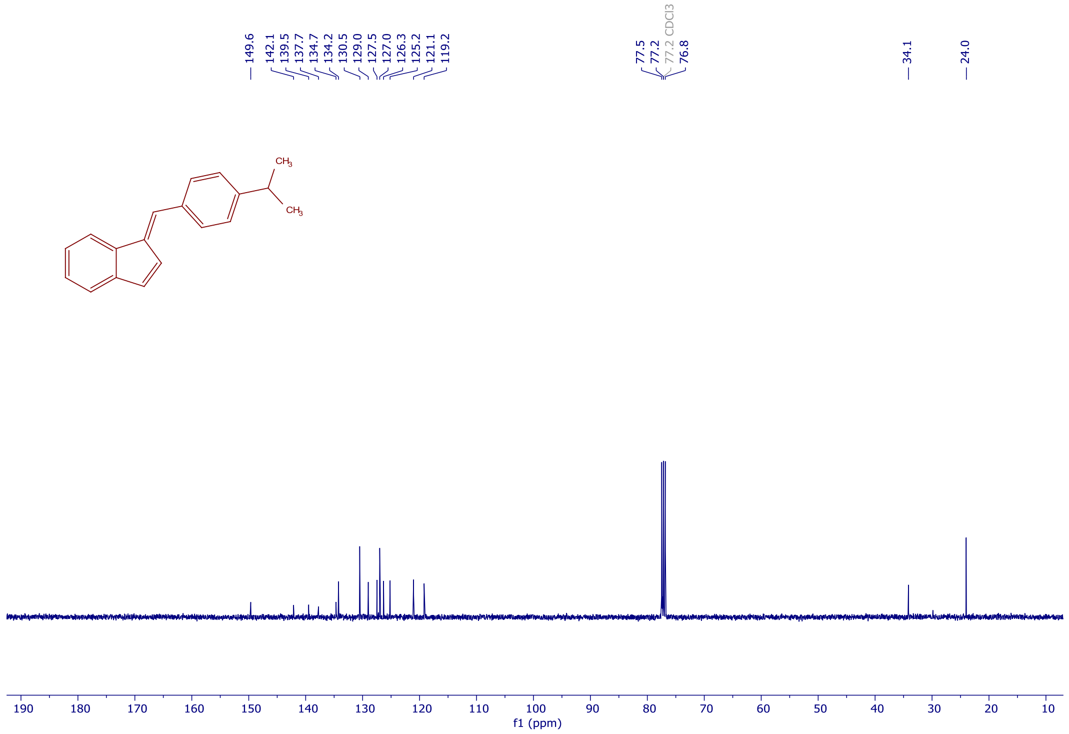
*^{1}H NMR of (E)-1-(4-nitrobenzylidene)-1H-indene (Compound-**3g**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



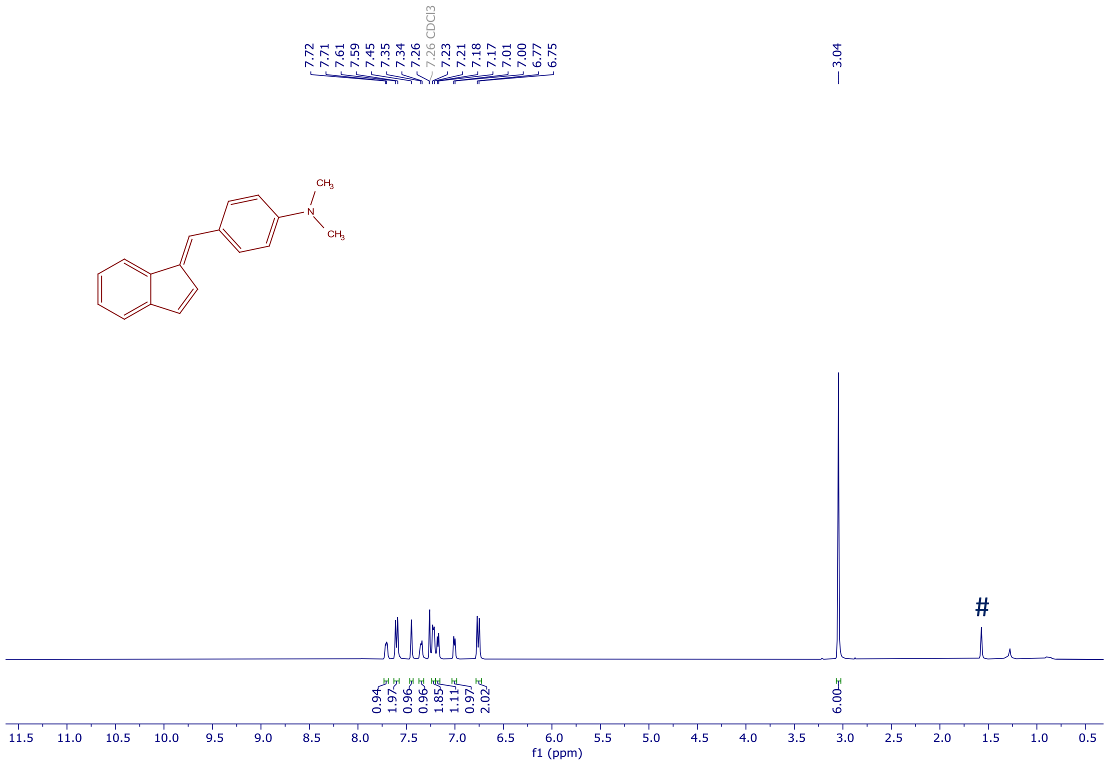
*$^{13}C\{^1H\}$ NMR of (E)-1-(4-nitrobenzylidene)-1H-indene (Compound-**3g**) in $CDCl_3$*



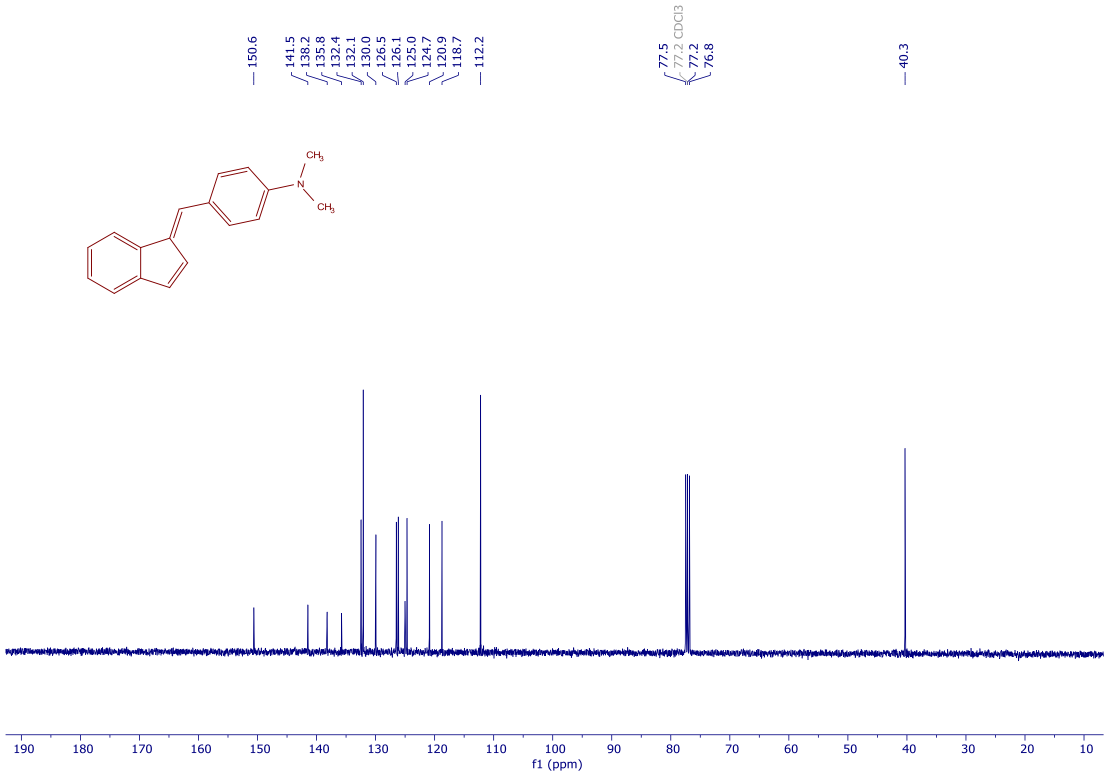
*^{1}H NMR of (E)-1-(4-isopropylbenzylidene)-1H-indene (Compound-**3h**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



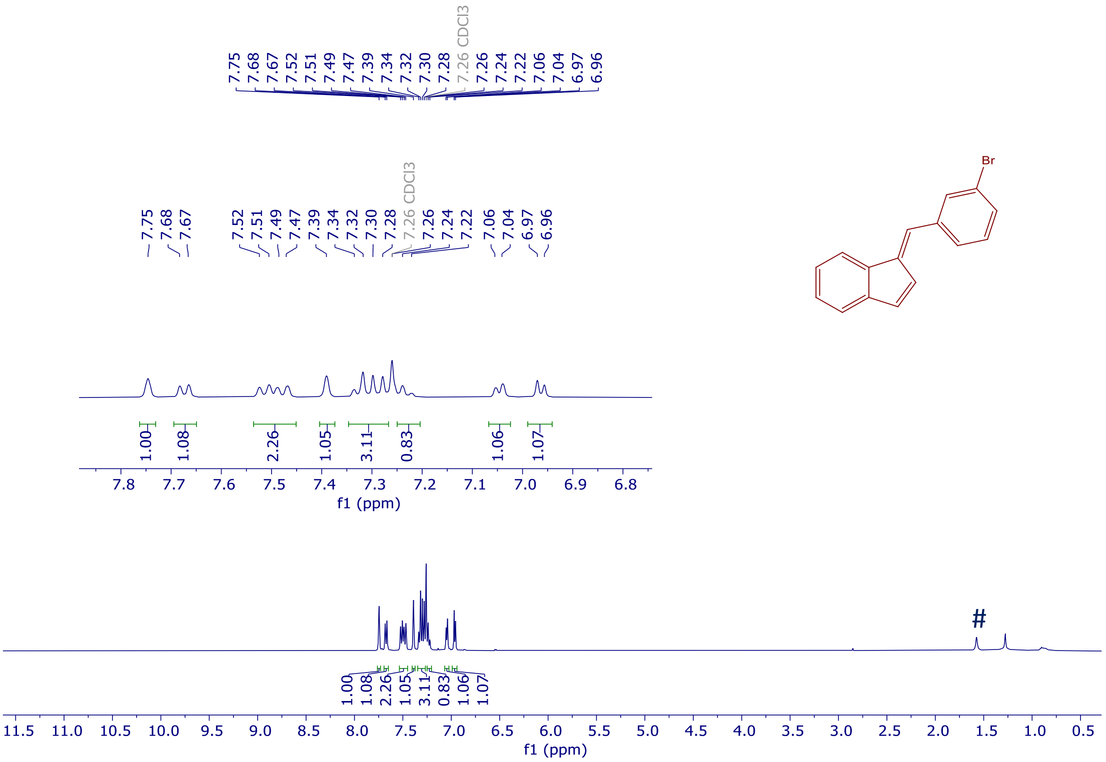
*$^{13}C\{^1H\}$ NMR of (E)-1-(4-isopropylbenzylidene)-1H-indene (Compound-**3h**) in $CDCl_3$*



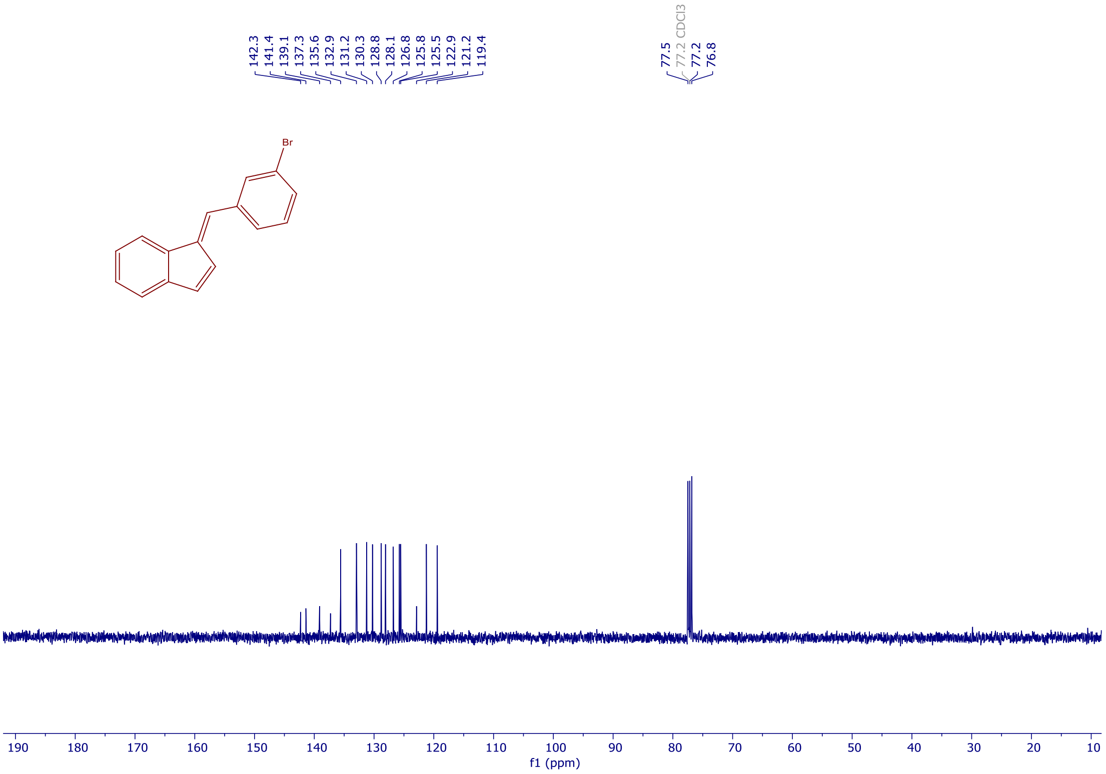
*^{1}H NMR of (E)-4-((1H-inden-1-ylidene)methyl)-N,N-dimethylaniline (Compound-**3i**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



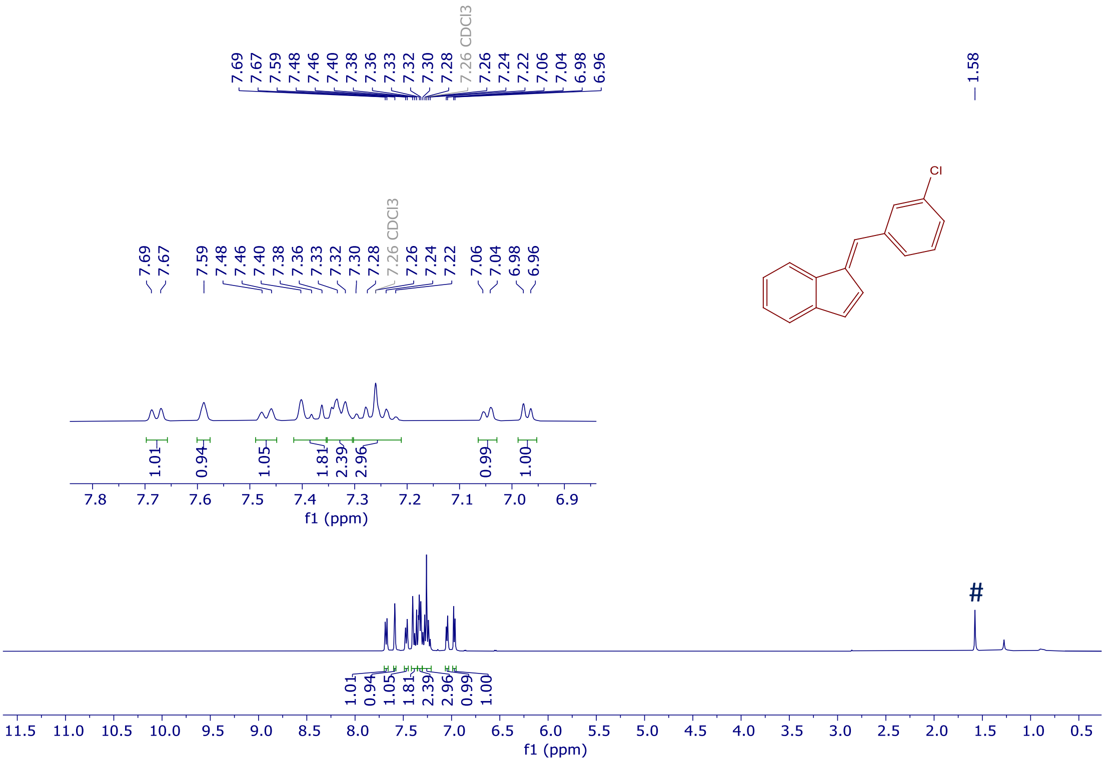
*^{13}C{^{1}H} NMR of (E)-4-((1H-inden-1-ylidene)methyl)-N,N-dimethylaniline (Compound-**3i**) in $CDCl_3$*



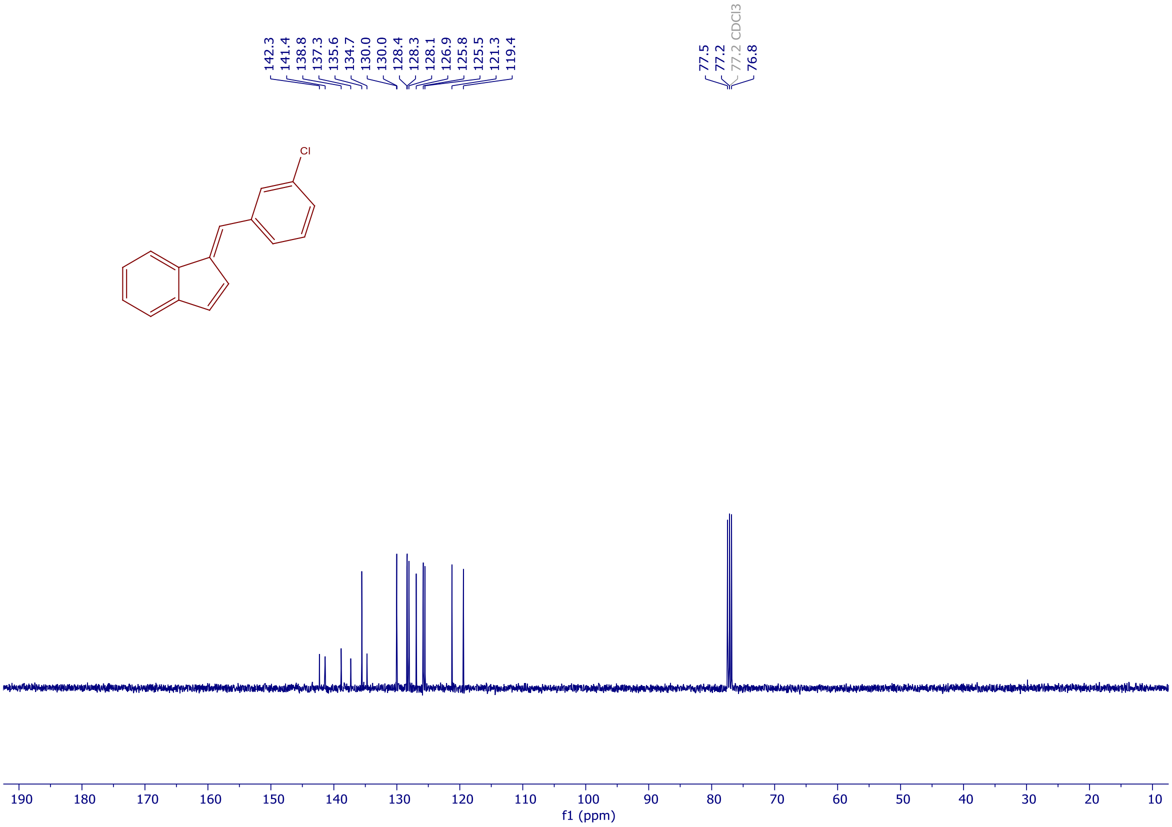
*^{1}H NMR of (E)-1-(3-bromobenzylidene)-1H-indene (Compound-**3j**) in CDCl$_3$. # indicates the solvent impurity of H$_2$O in CDCl$_3$*



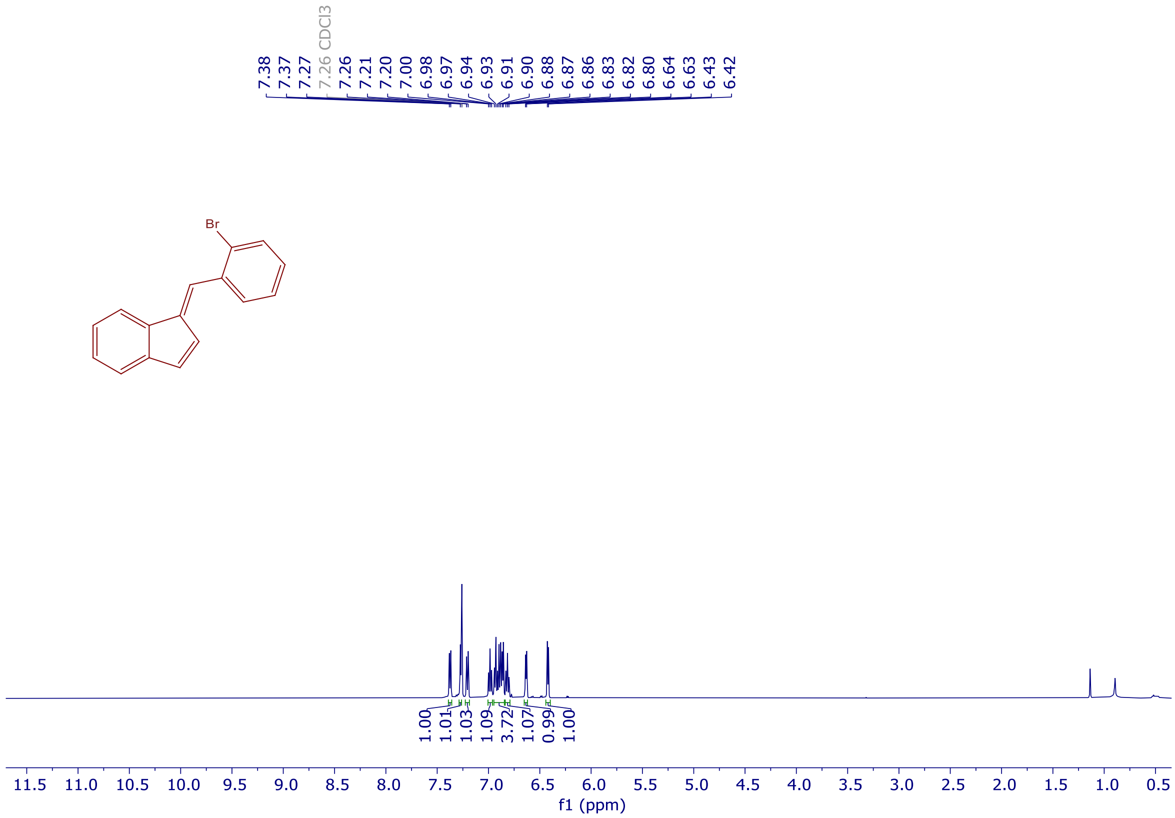
*^{13}C{^{1}H} NMR of (E)-1-(3-bromobenzylidene)-1H-indene (Compound-**3j**) in CDCl$_3$*



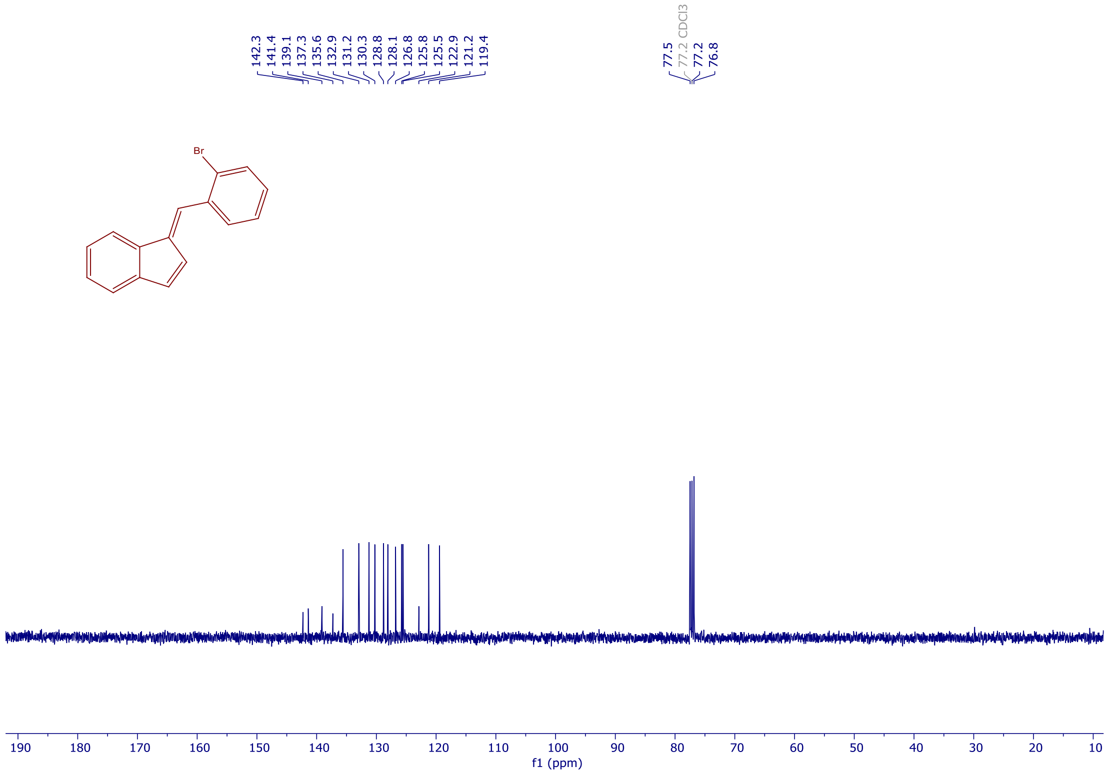
*^{1}H NMR of (E)-1-(3-chlorobenzylidene)-1H-indene (Compound-**3k**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



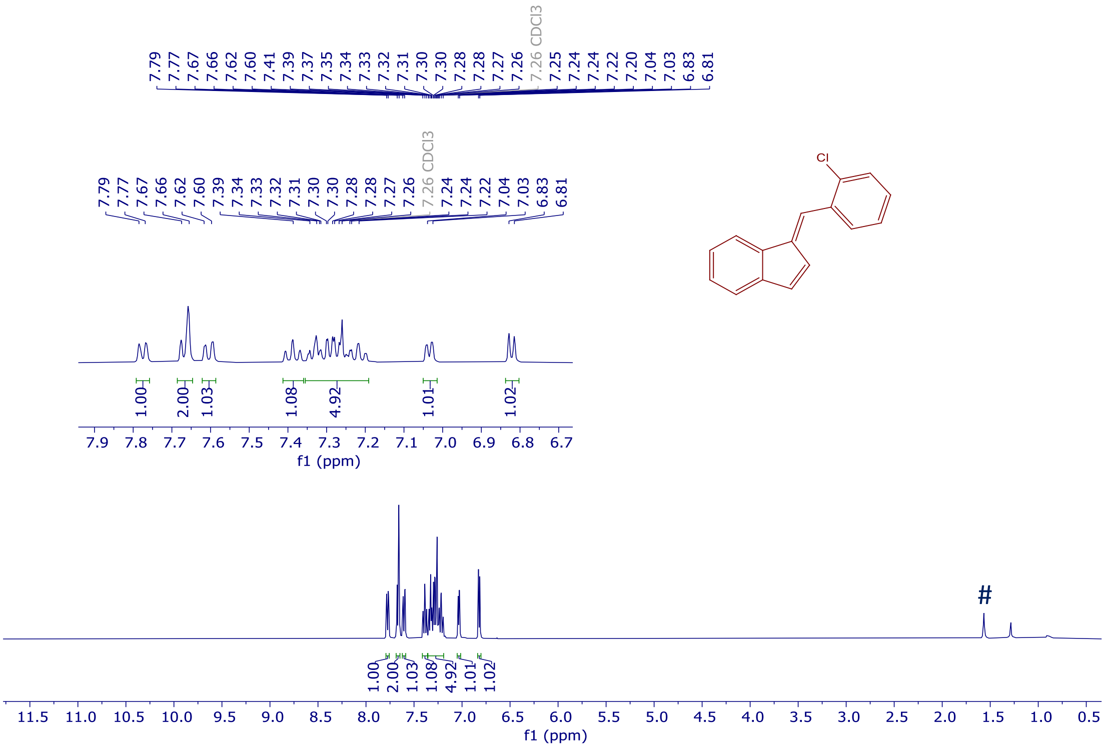
*^{13}C{^{1}H} NMR of (E)-1-(3-chlorobenzylidene)-1H-indene (Compound-**3k**) in $CDCl_3$*



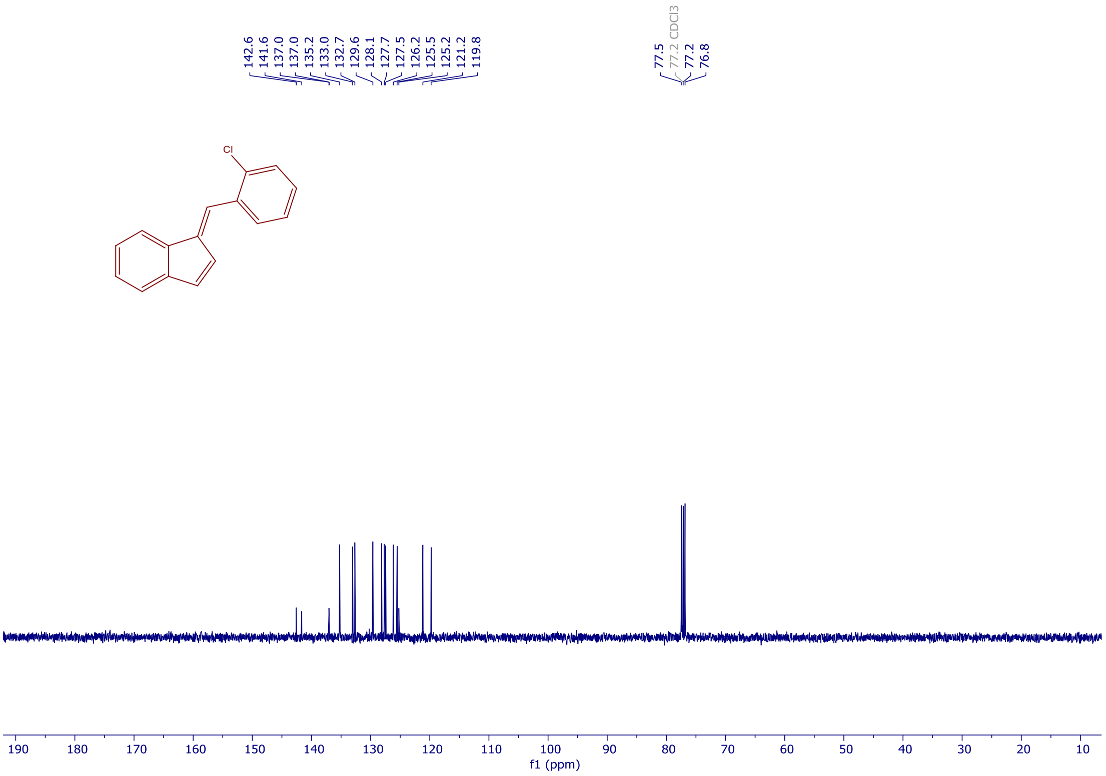
*^{1}H NMR of (E)-1-(2-bromobenzylidene)-1H-indene (Compound-**3l**) in CDCl$_{3}$.*



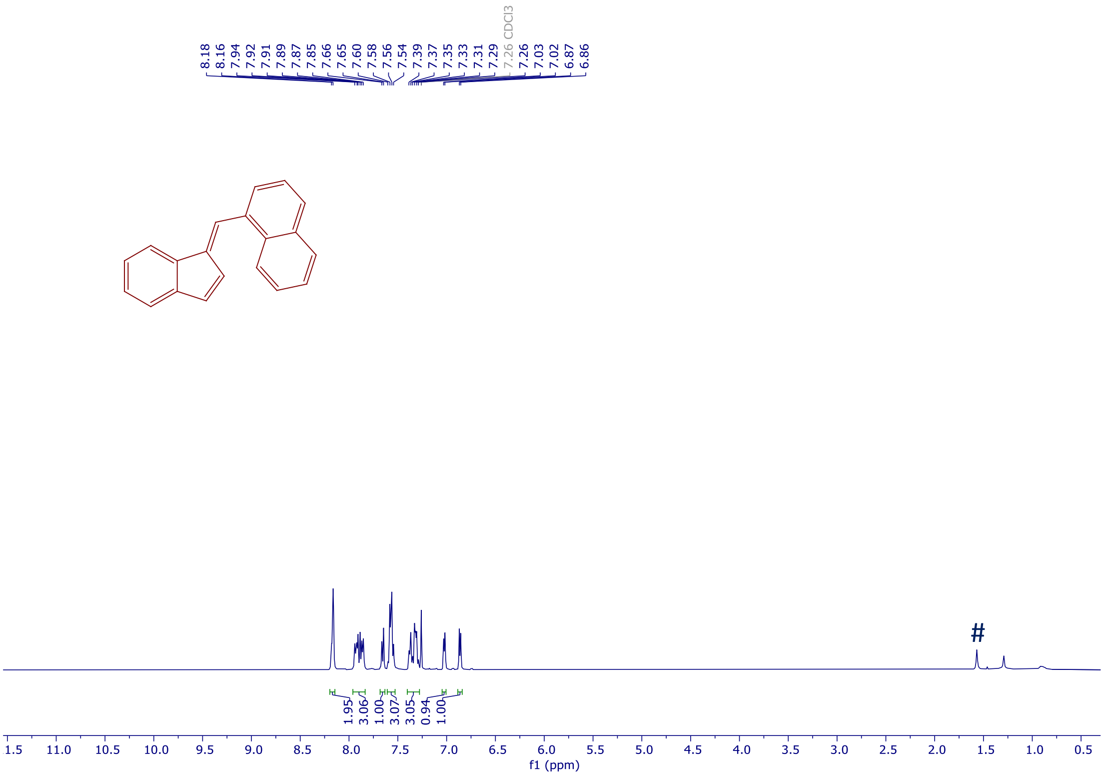
*^{13}C{^{1}H} NMR of (E)-1-(2-bromobenzylidene)-1H-indene (Compound-**3l**) in CDCl$_{3}$*



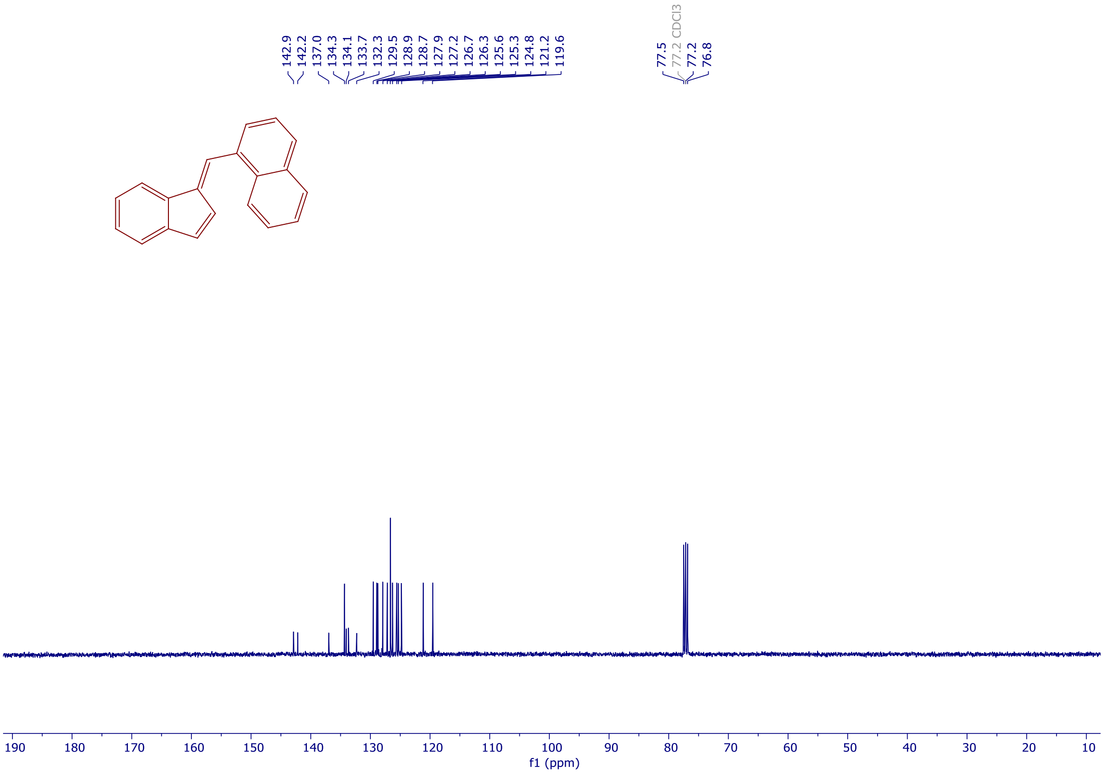
*^{1}H NMR of (E)-1-(2-chlorobenzylidene)-1H-indene (Compound-**3m**) in CDCl$_3$. # indicates the solvent impurity of H_2O in CDCl$_3$*



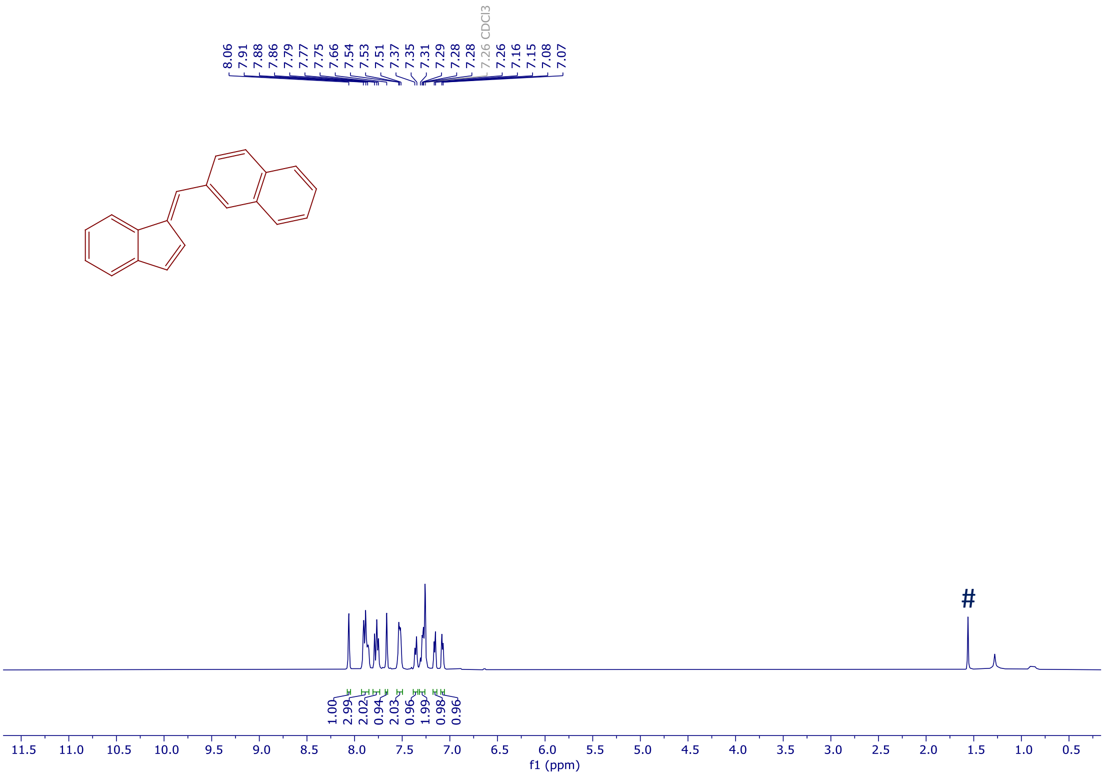
*^{13}C{^{1}H} NMR of (E)-1-(2-chlorobenzylidene)-1H-indene (Compound-**3m**) in CDCl$_3$*



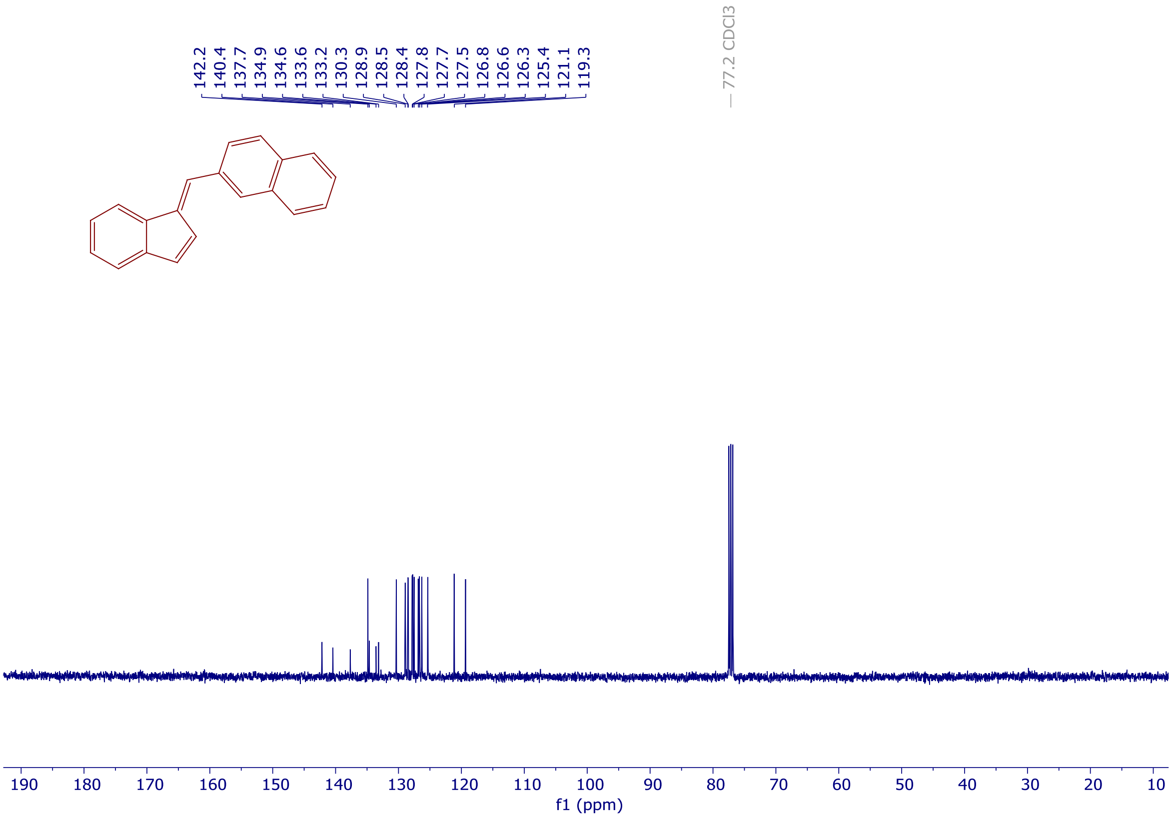
^{1}*H NMR of (E)-1-((1H-inden-1-ylidene)methyl)naphthalene(Compound-**3n**) in* $CDCl_3$*. # indicates the solvent impurity of* H_2O *in* $CDCl_3$



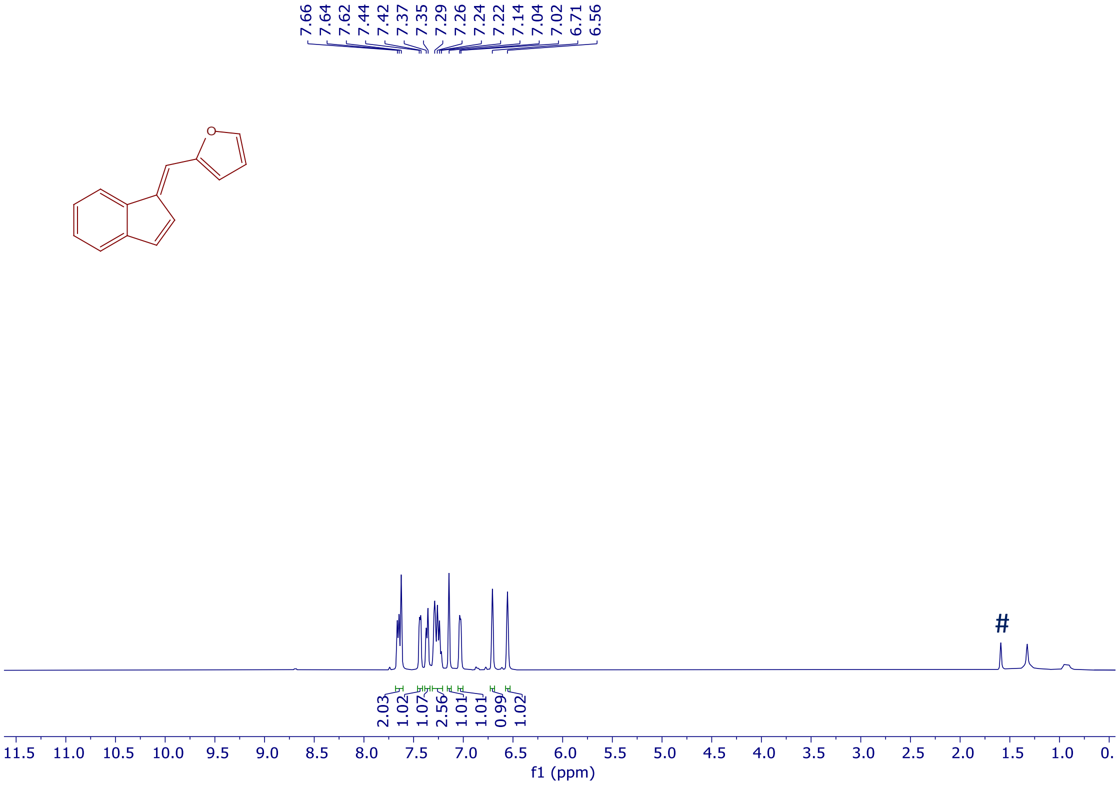
^{13}*C{*^{1}*H} NMR of (E)-1-((1H-inden-1-ylidene)methyl)naphthalene (Compound-**3n**) in* $CDCl_3$



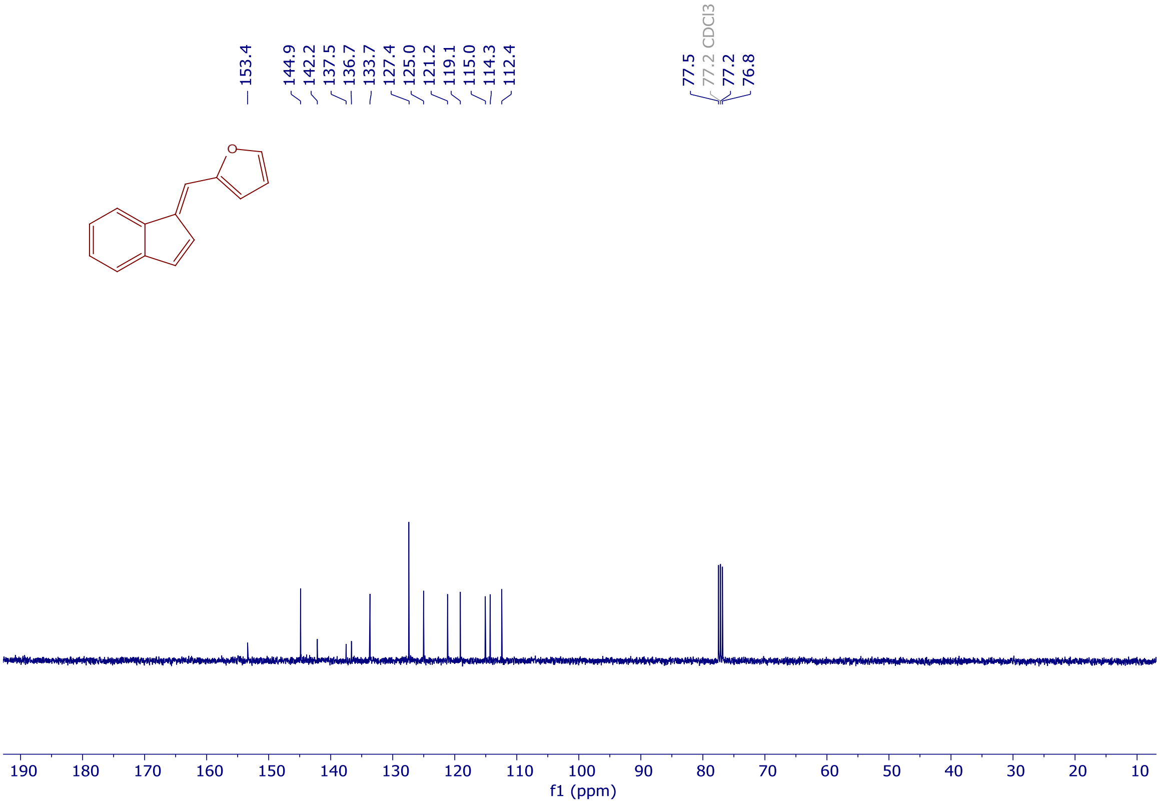
^{1}H NMR of *(E)-2-((1H-inden-1-ylidene)methyl)naphthalene (Compound-**3o**) in CDCl$_3$. # indicates the solvent impurity of H$_2$O in CDCl$_3$*



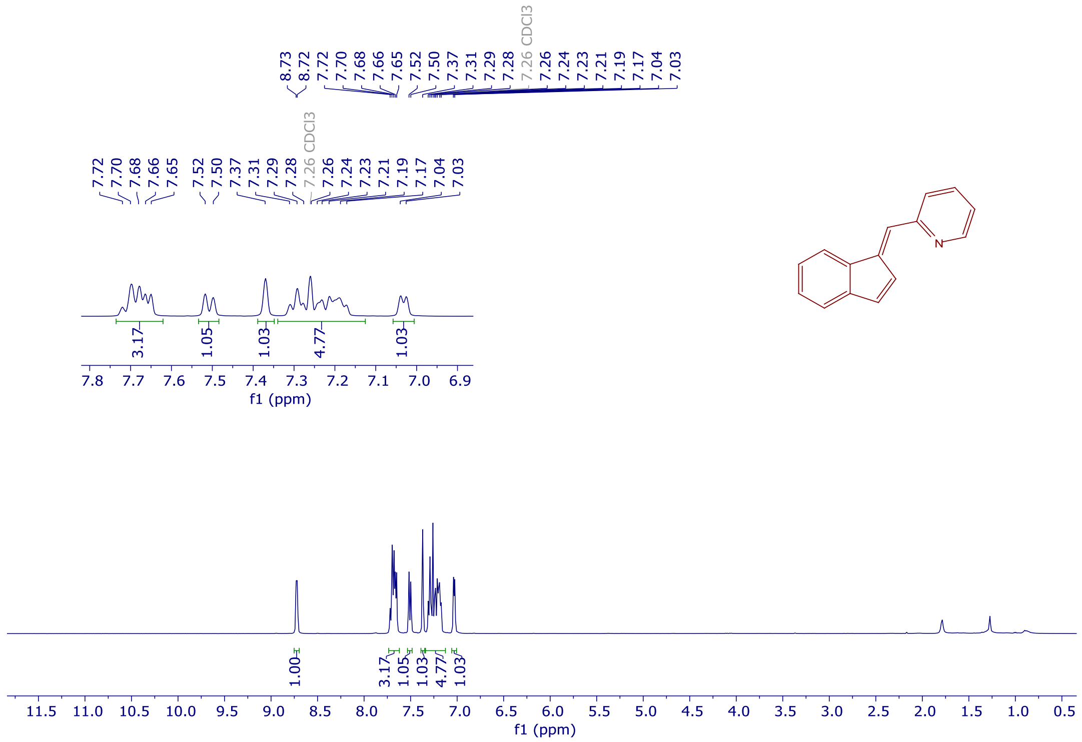
^{13}C{^{1}H} NMR of *(E)-2-((1H-inden-1-ylidene)methyl)naphthalene (Compound-**3o**) in CDCl$_3$*



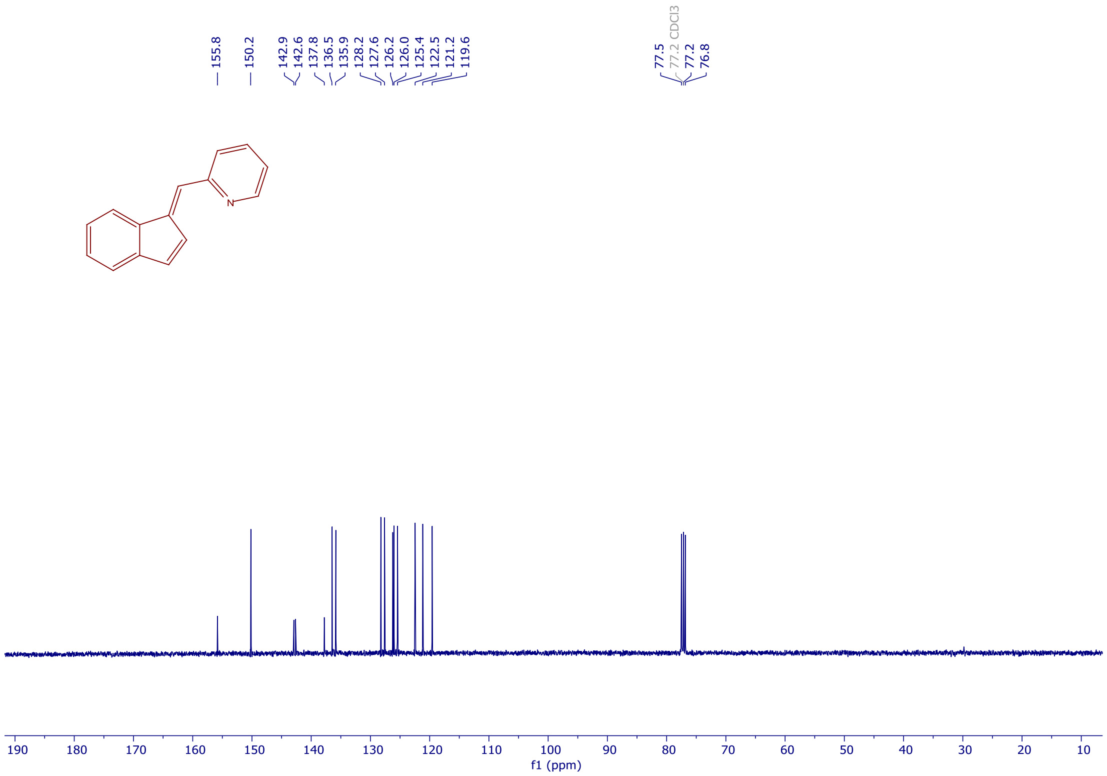
*^{1}H NMR of (E)-2-((1H-inden-1-ylidene)methyl)furan (Compound-**3p**) in $CDCl_3$ (*). # indicates the solvent impurity of H_2O in $CDCl_3$*



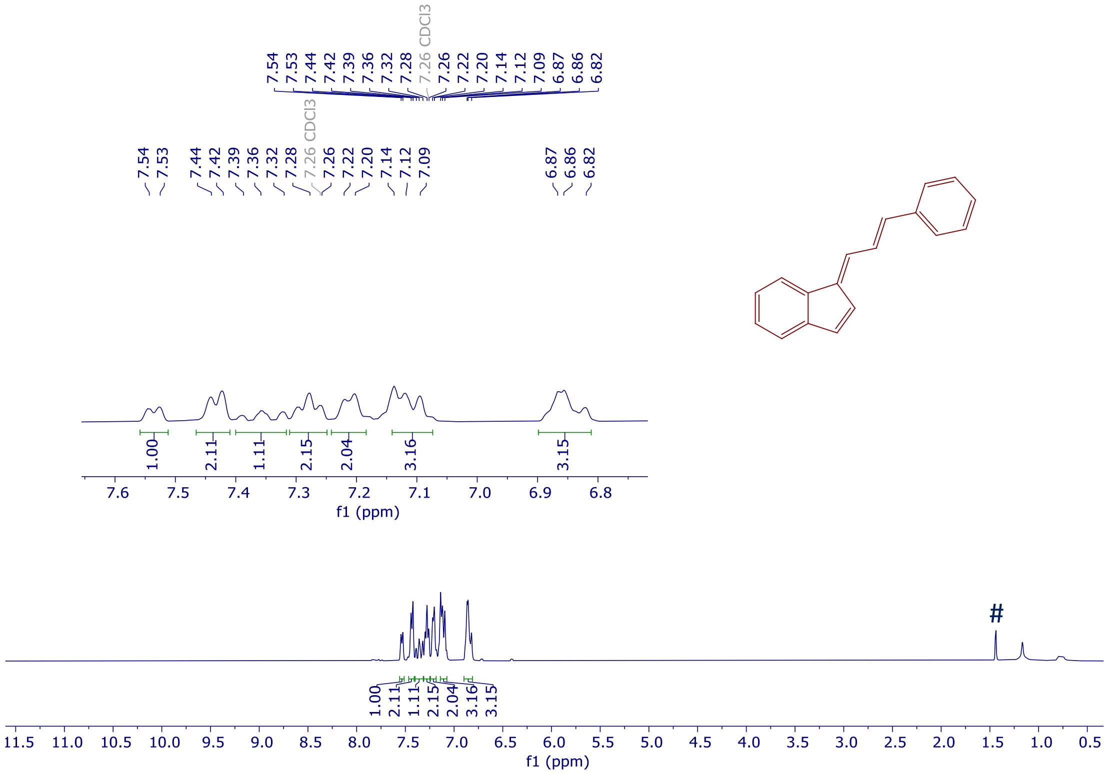
*$^{13}C\{^1H\}$ NMR of (E)-2-((1H-inden-1-ylidene)methyl)furan (Compound-**3p**) in $CDCl_3$*



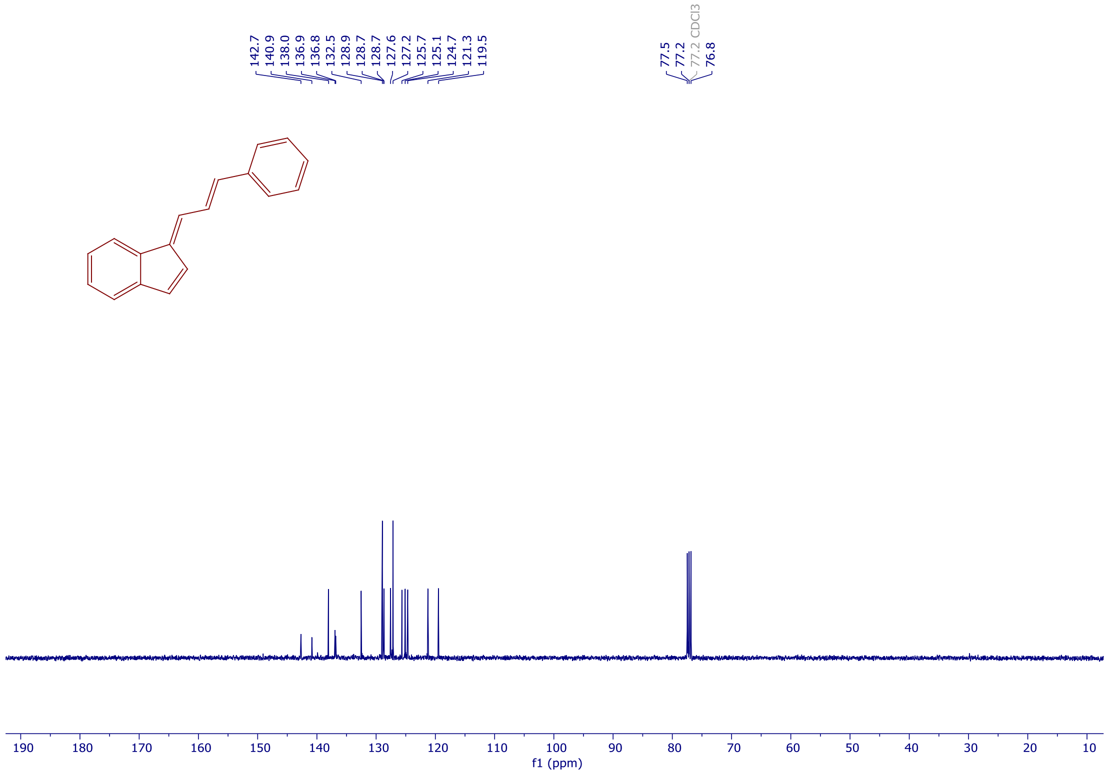
^{1}H NMR of (E)-2-((1H-inden-1-ylidene)methyl)pyridine (Compound-**3q**) in $CDCl_3$



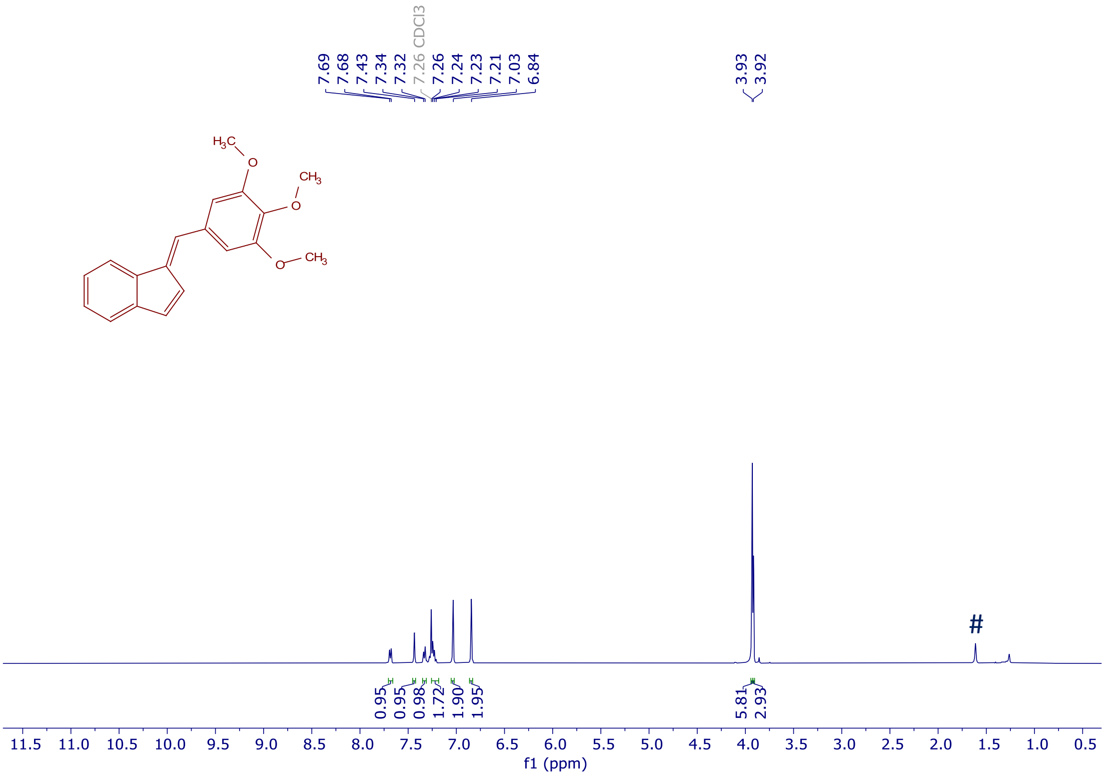
$^{13}C\{^1H\}$ NMR of (E)-2-((1H-inden-1-ylidene)methyl)pyridine (Compound-**3q**) in $CDCl_3$



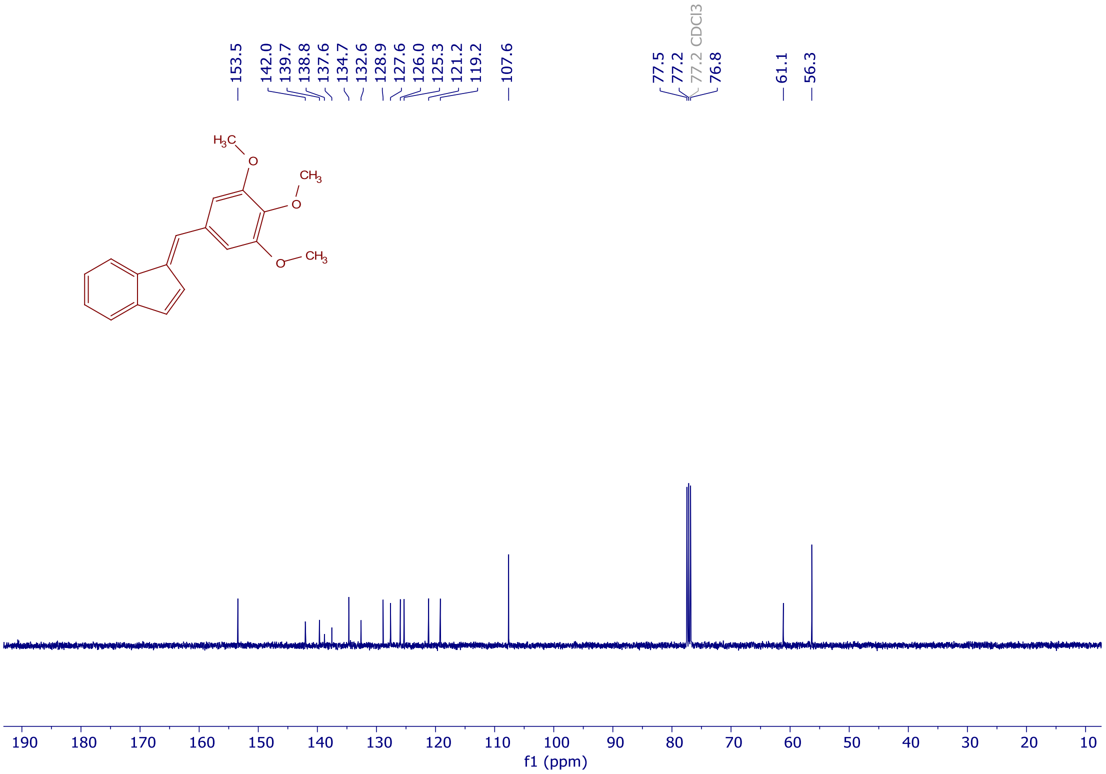
*^{1}H NMR of (E)-1-((E)-3-phenylallylidene)-1H-indene (Compound-**3r**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



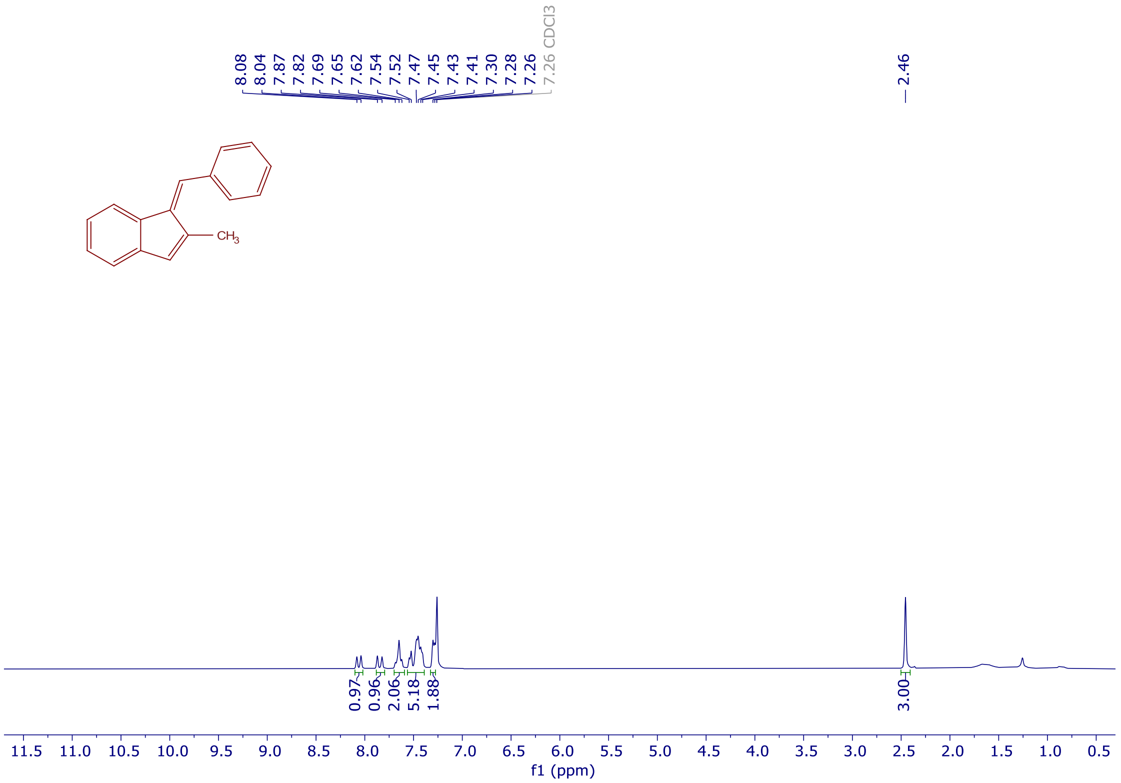
*$^{13}C\{^{1}H\}$ NMR of (E)-1-((E)-3-phenylallylidene)-1H-indene (Compound-**3r**) in $CDCl_3$*



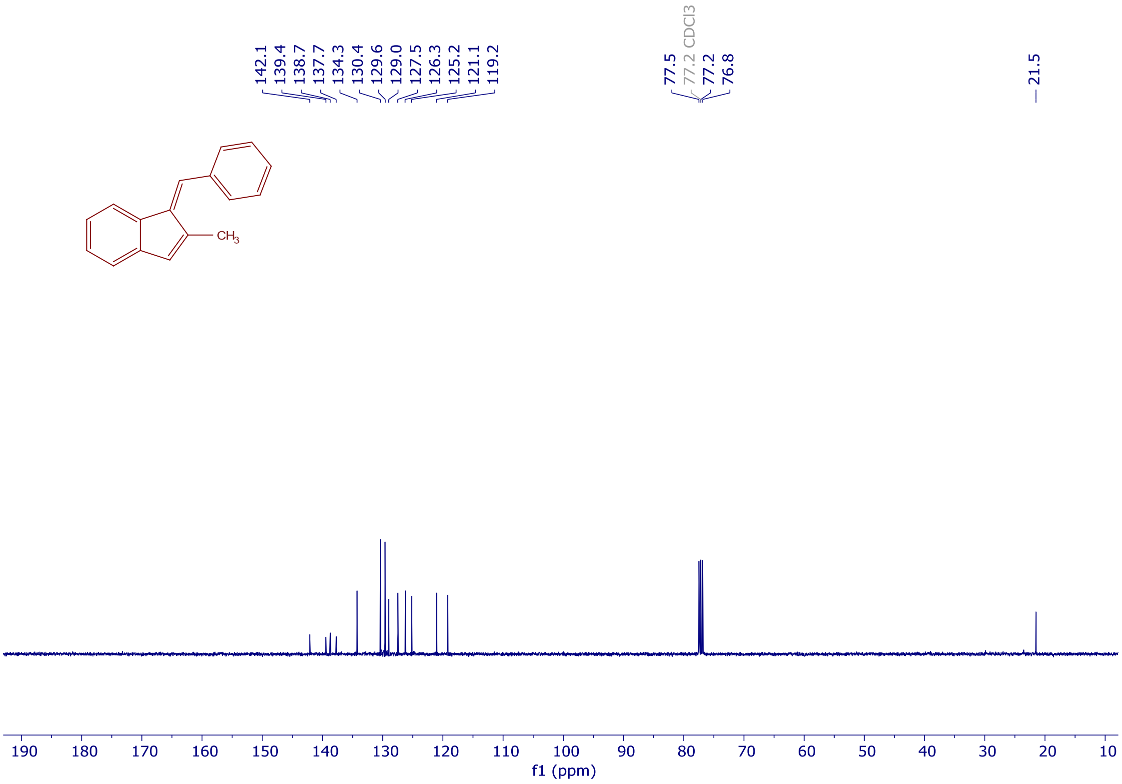
*^{1}H NMR of (E)-1-(3,4,5-trimethoxybenzylidene)-1H-indene (Compound-**3s**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



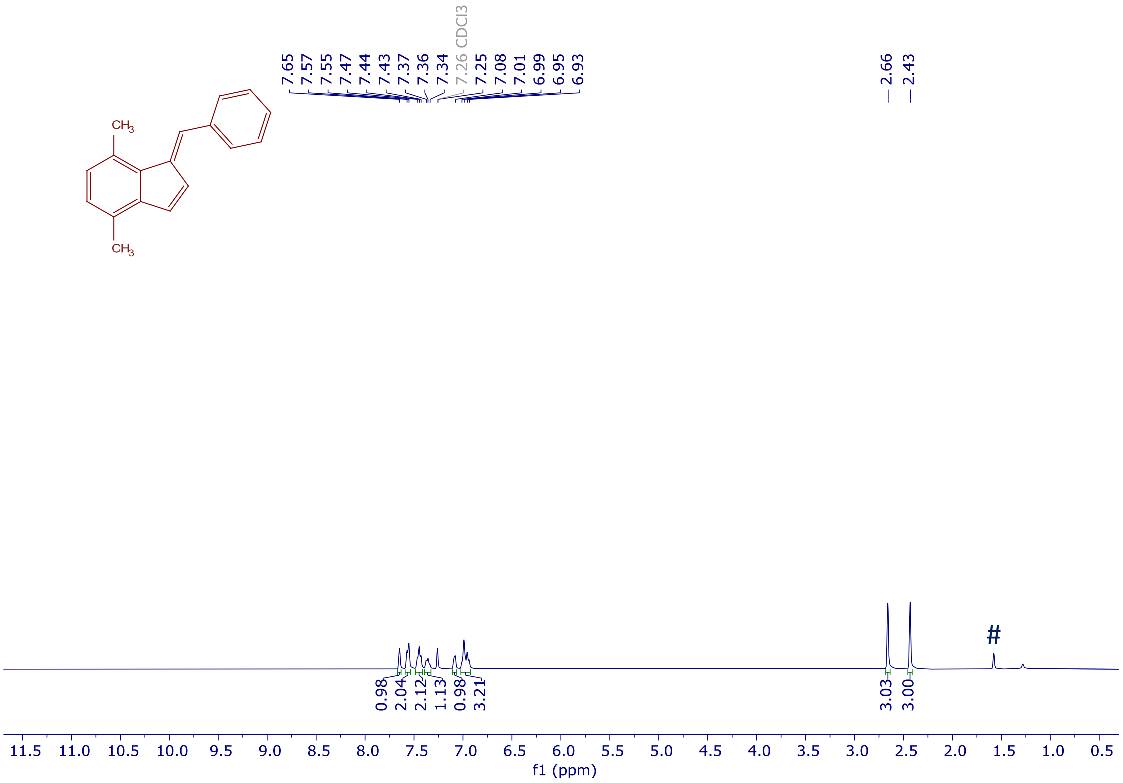
*$^{13}C\{^1H\}$ NMR of (E)-1-(3,4,5-trimethoxybenzylidene)-1H-indene (Compound-**3s**) in $CDCl_3$*



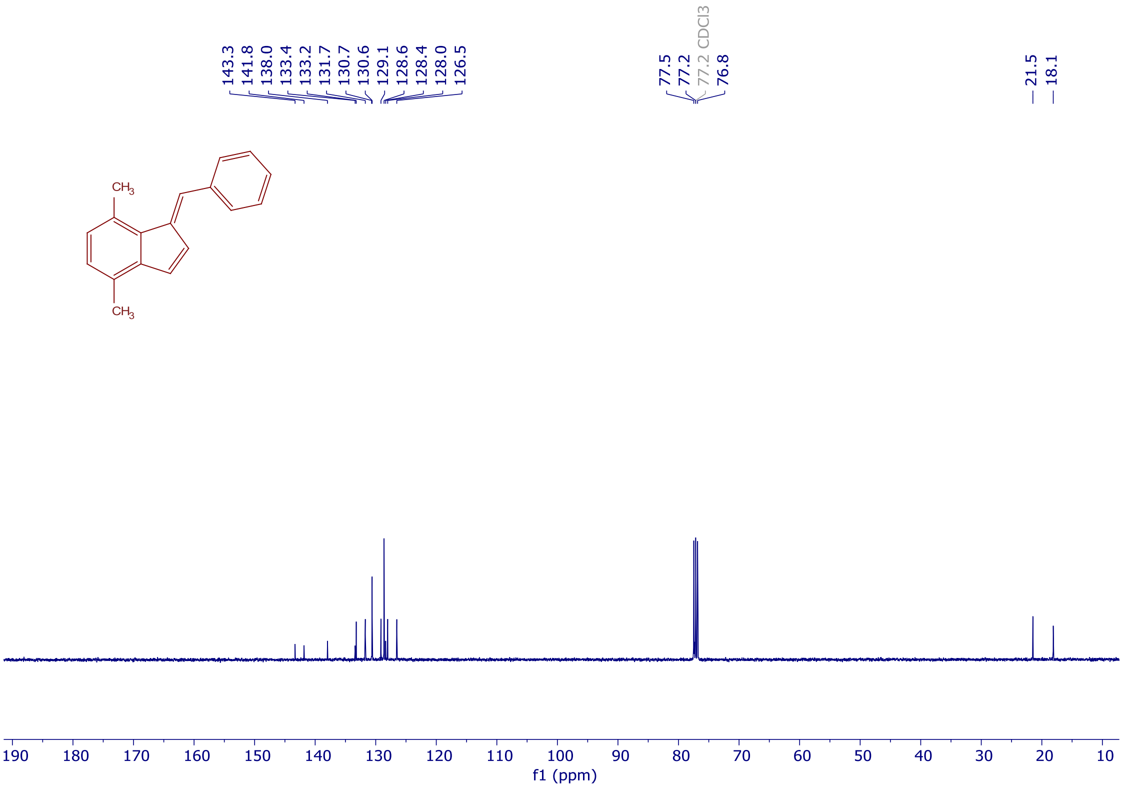
^{1}H NMR of (E)-1-benzylidene-2-methyl-1H-indene (Compound-**3t**) in CDCl$_3$



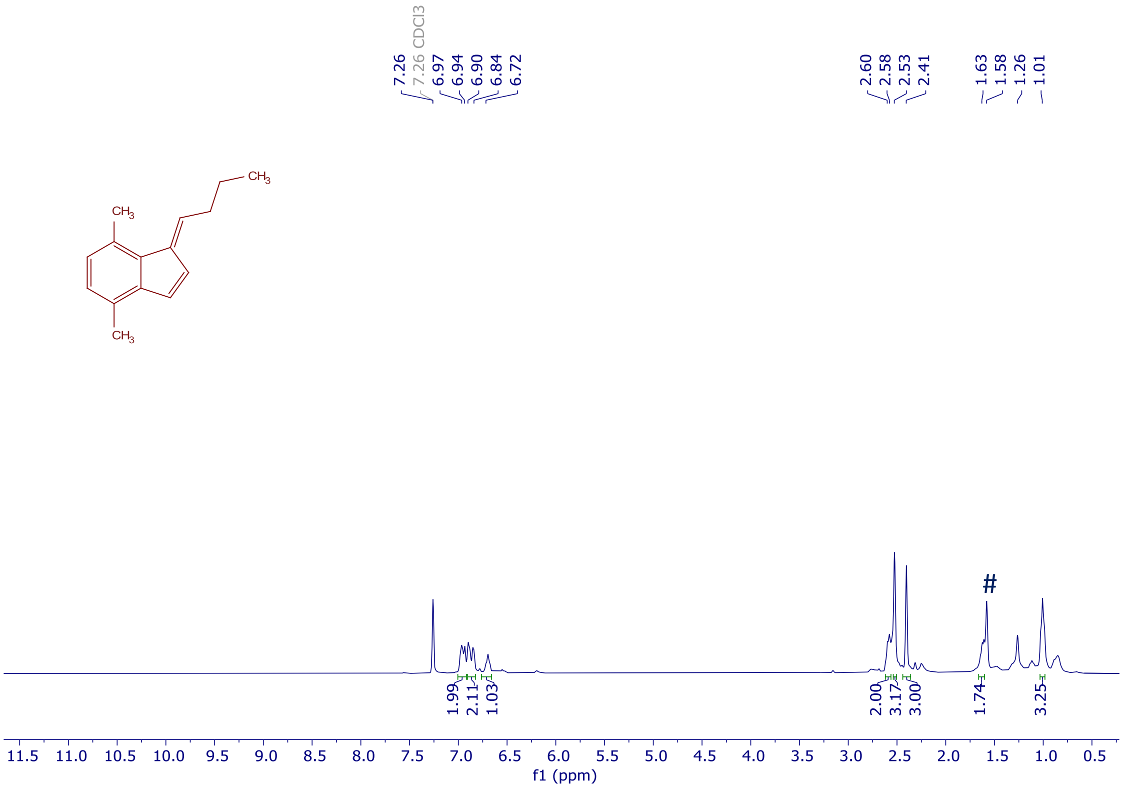
^{13}C{^{1}H} NMR of (E)-1-benzylidene-2-methyl-1H-indene (Compound-**3t**) in CDCl$_3$



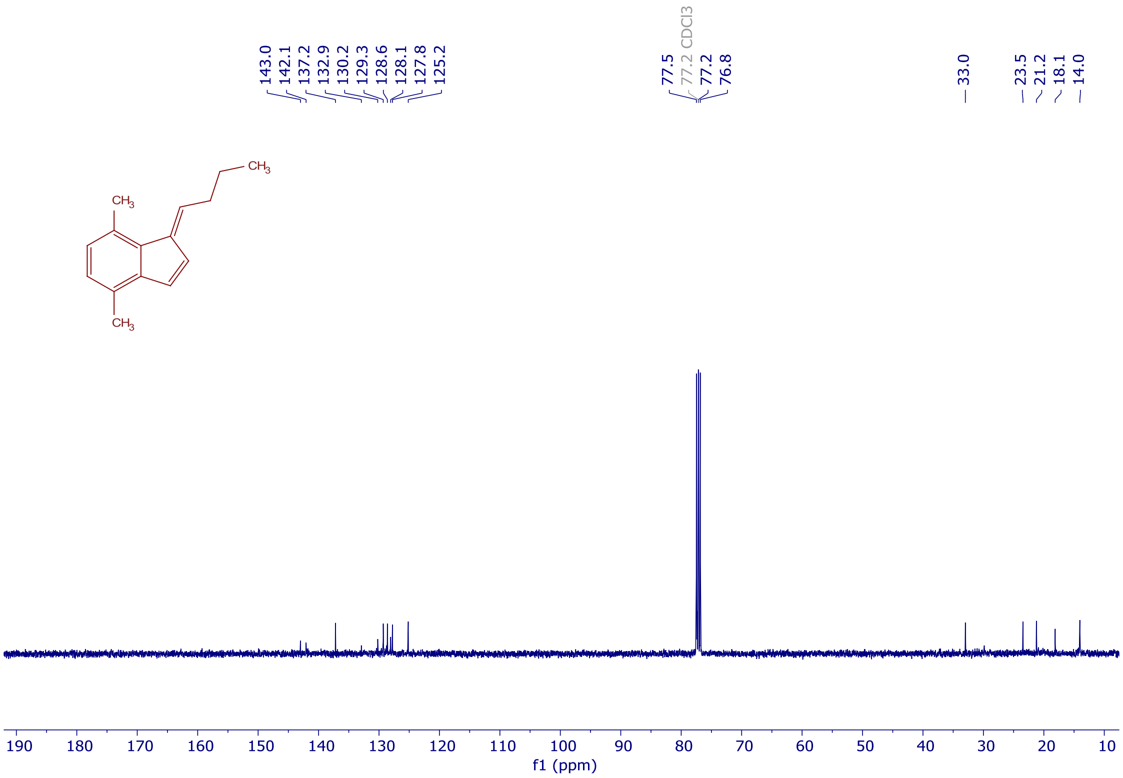
*^{1}H NMR of (E)-1-benzylidene-4,7-dimethyl-1H-indene (Compound-**3u**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



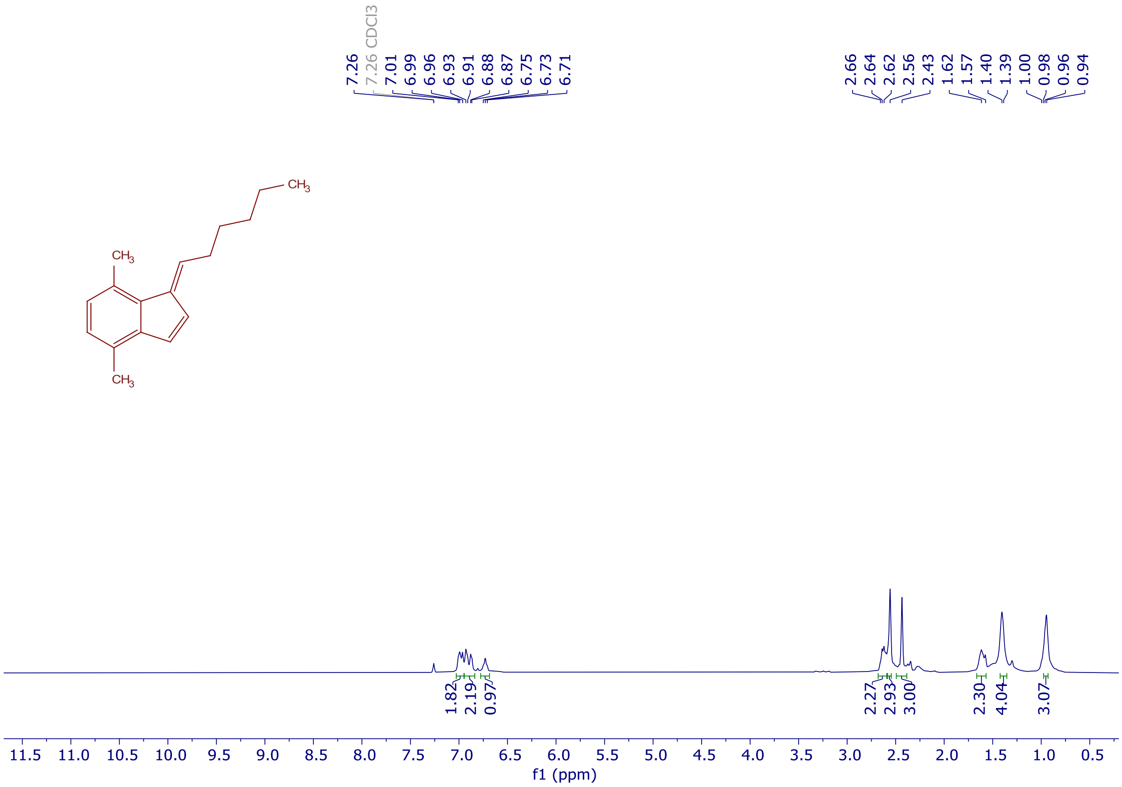
*$^{13}C\{^1H\}$ NMR of (E)-1-benzylidene-4,7-dimethyl-1H-indene (Compound-**3u**) in $CDCl_3$*



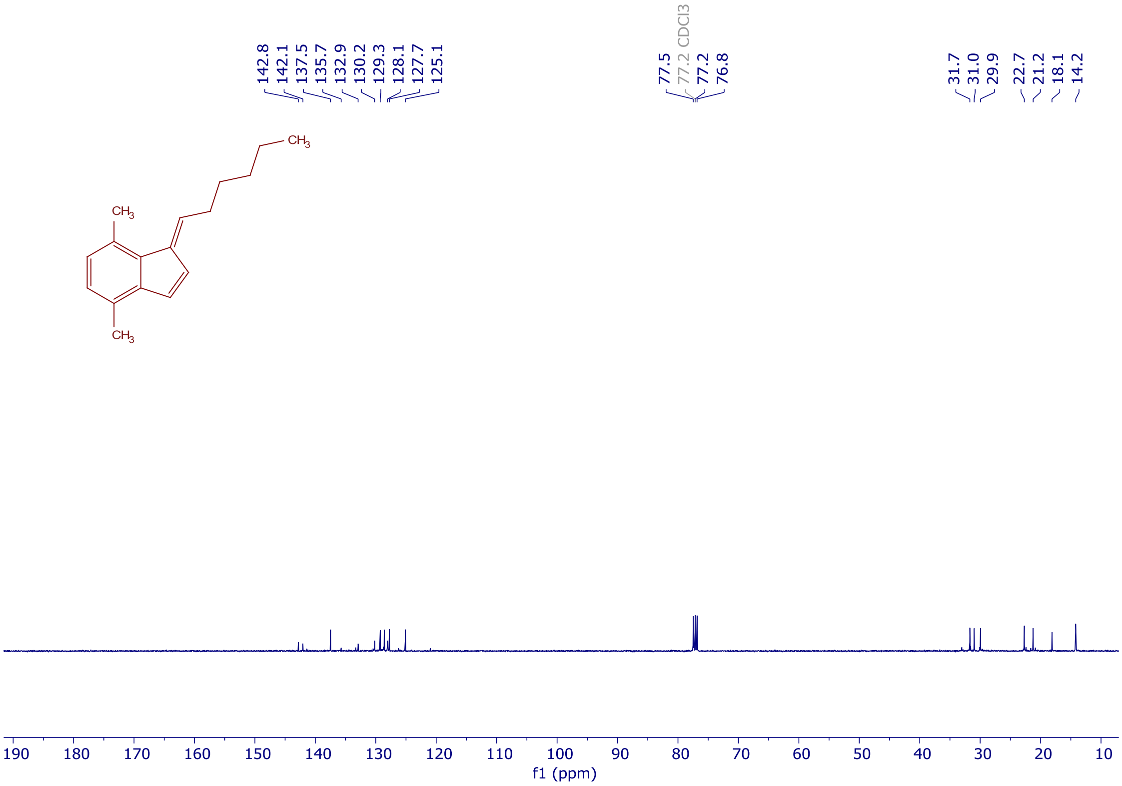
*^{1}H NMR of (E)- (1-butylidene-4,7-dimethyl-1H-indene (Compound-**3v**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



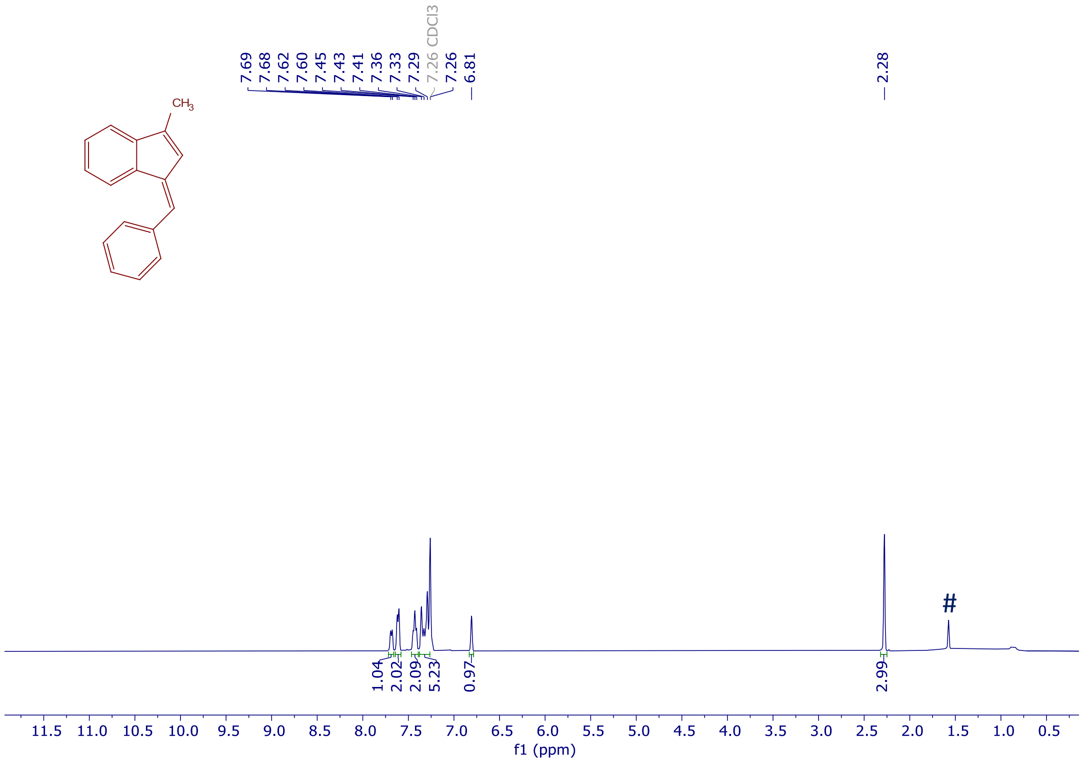
*$^{13}C\{^1H\}$ NMR of (E)- (1-butylidene-4,7-dimethyl-1H-indene (Compound-**3v**) in $CDCl_3$*



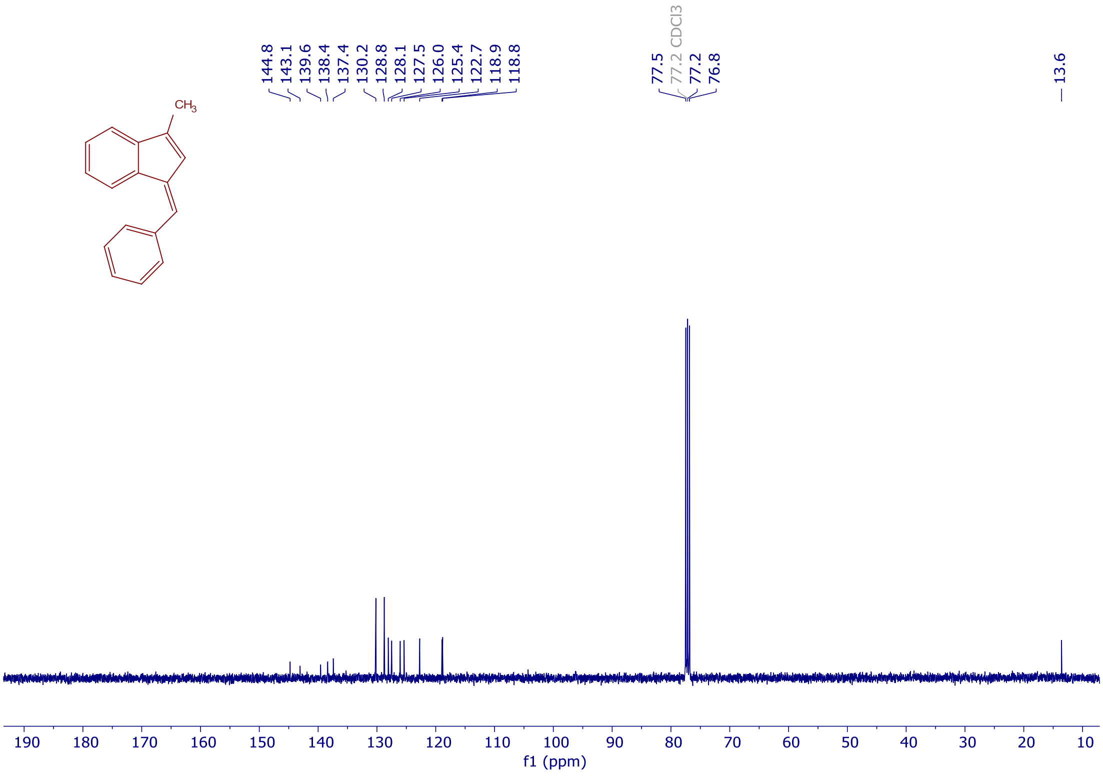
*^{1}H NMR of CDCl$_3$ (E)-1-hexylidene-4,7-dimethyl-1H-indene (Compound-**3w**) in CDCl$_3$*



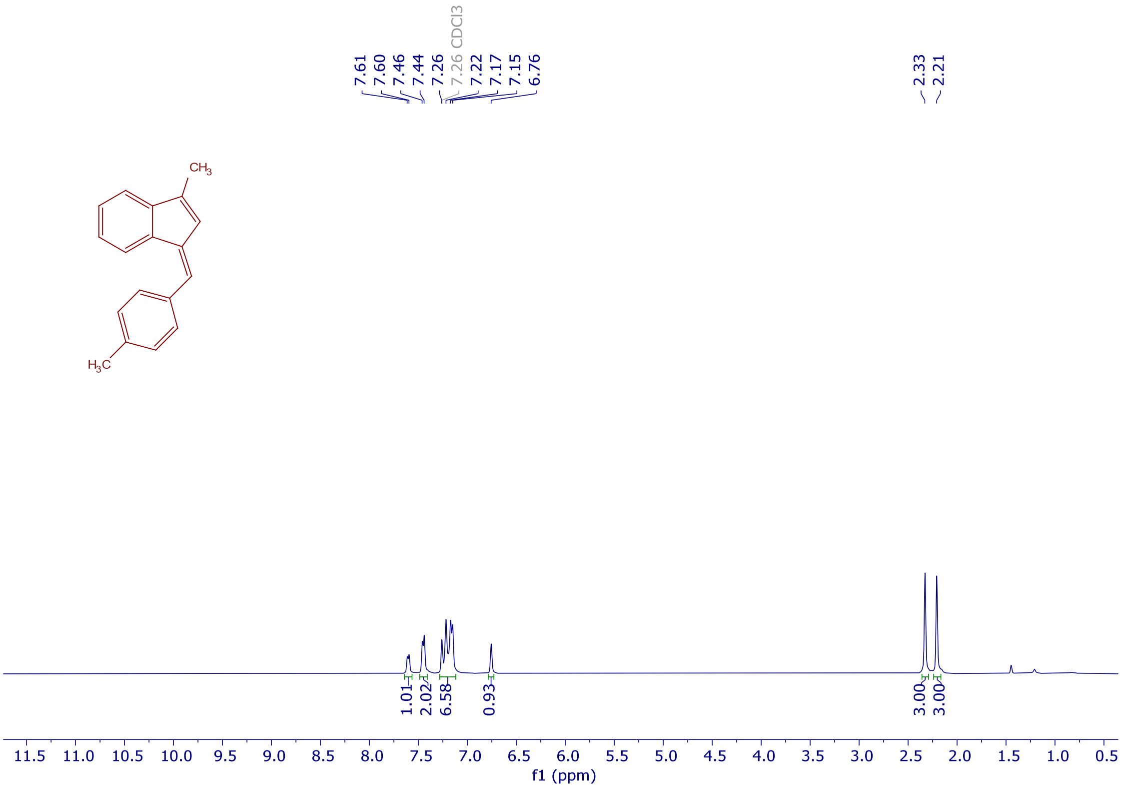
*^{13}C{^{1}H} NMR of (E)-1-hexylidene-4,7-dimethyl-1H-indene (Compound-**3w**) in CDCl$_3$*



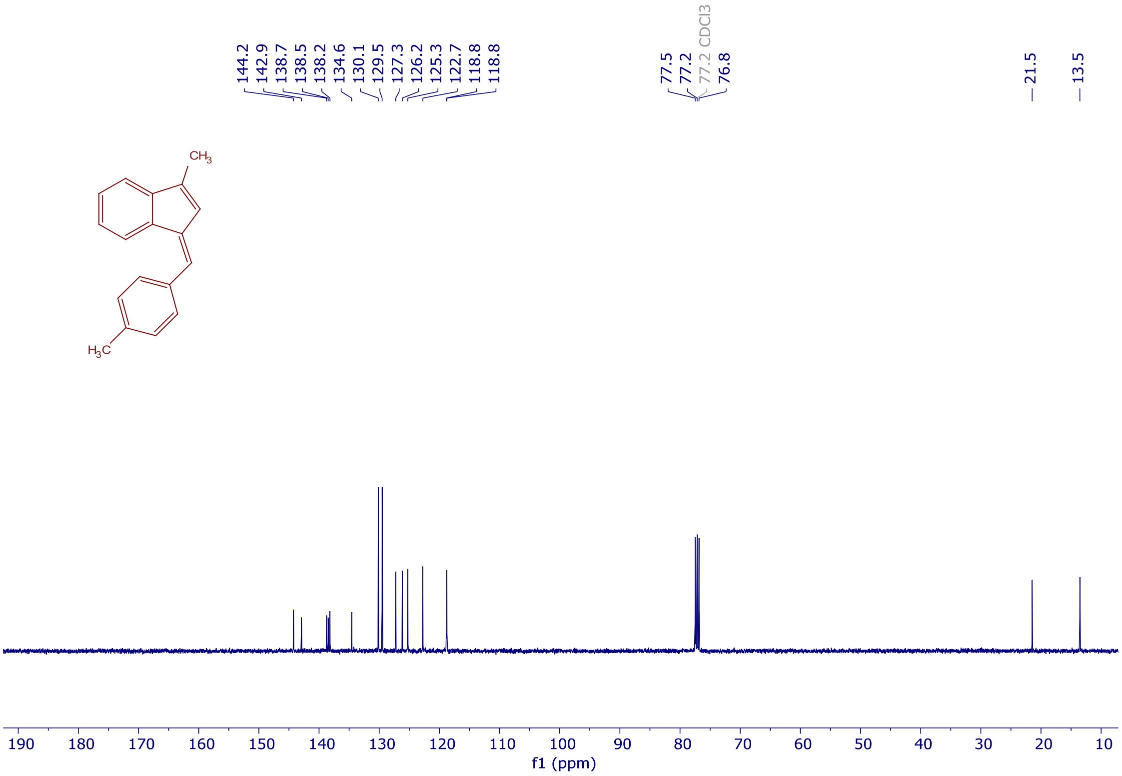
*^{1}H NMR of (E)-1-benzylidene-3-methyl-1H-indene (Compound-**3x**) in $CDCl_3$*



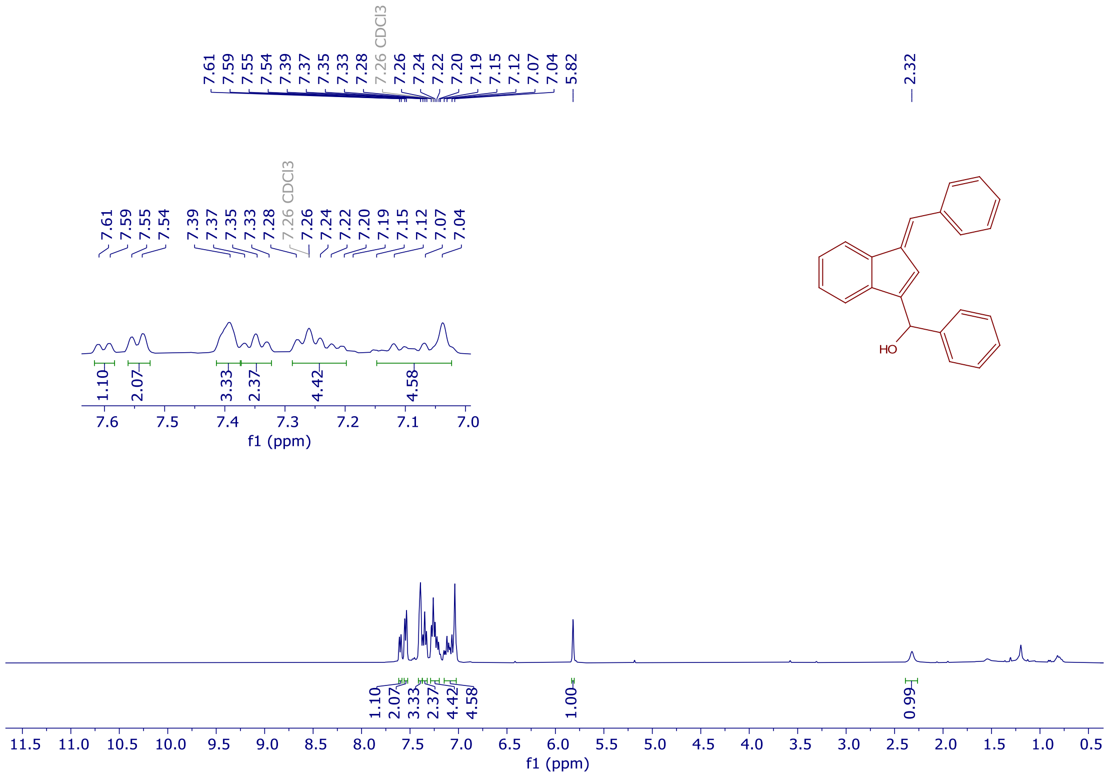
*^{13}C{^{1}H} NMR of (E)-1-benzylidene-3-methyl-1H-indene (Compound-**3x**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



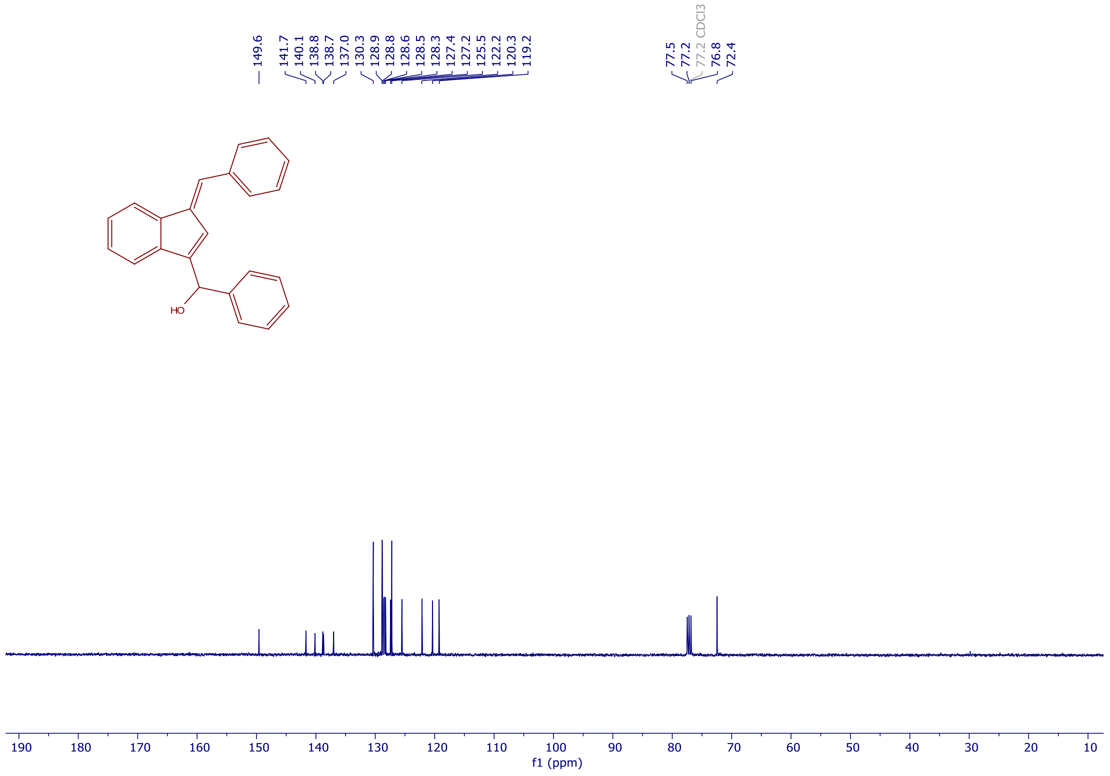
^{1}*H NMR of (E)-3-methyl-1-(4-methylbenzylidene)-1H-indene (Compound-**3y**) in* $CDCl_3$



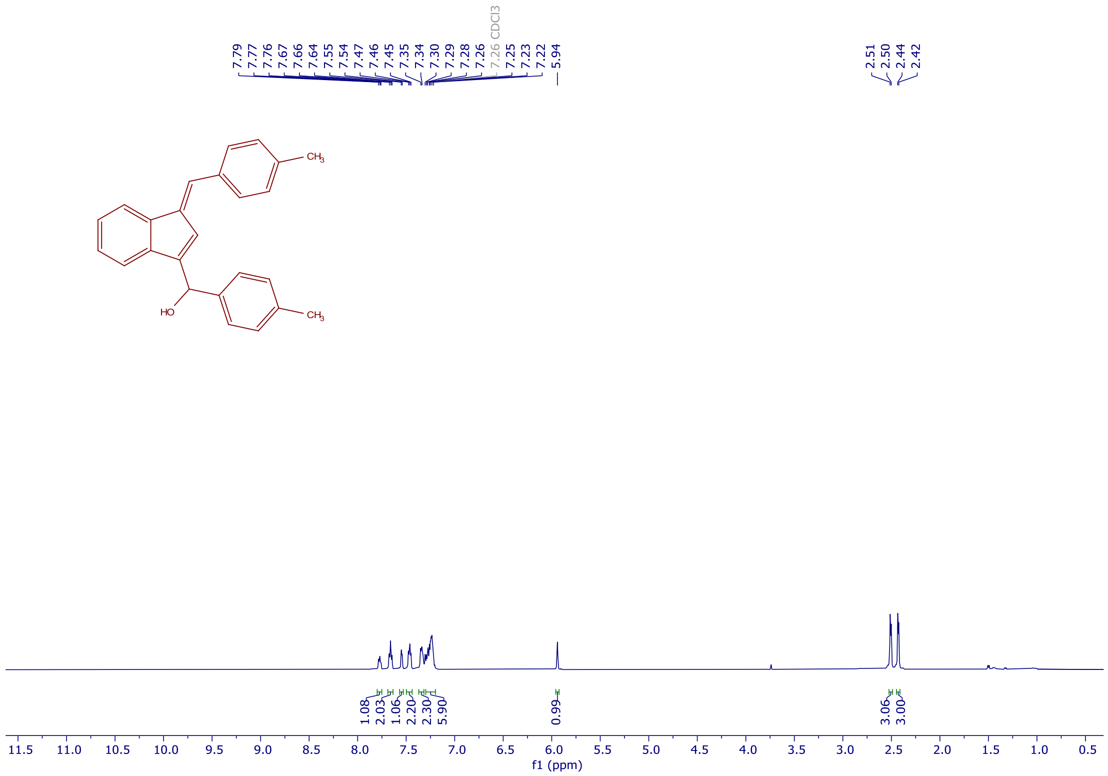
^{13}*C{*^{1}*H} NMR of (E)-3-methyl-1-(4-methylbenzylidene)-1H-indene (Compound-**3y**) in* $CDCl_3$



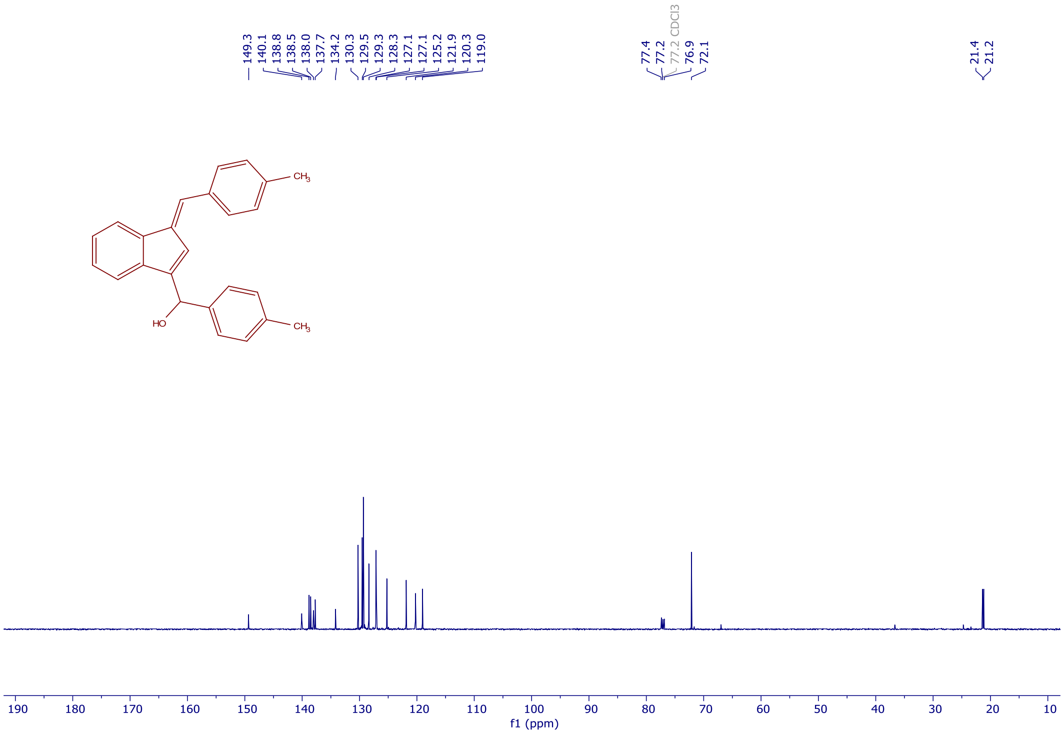
*^{1}H NMR of (E)-(1-benzylidene-1H-inden-3-yl)(phenyl)methanol (Compound-**4a**) in $CDCl_3$*



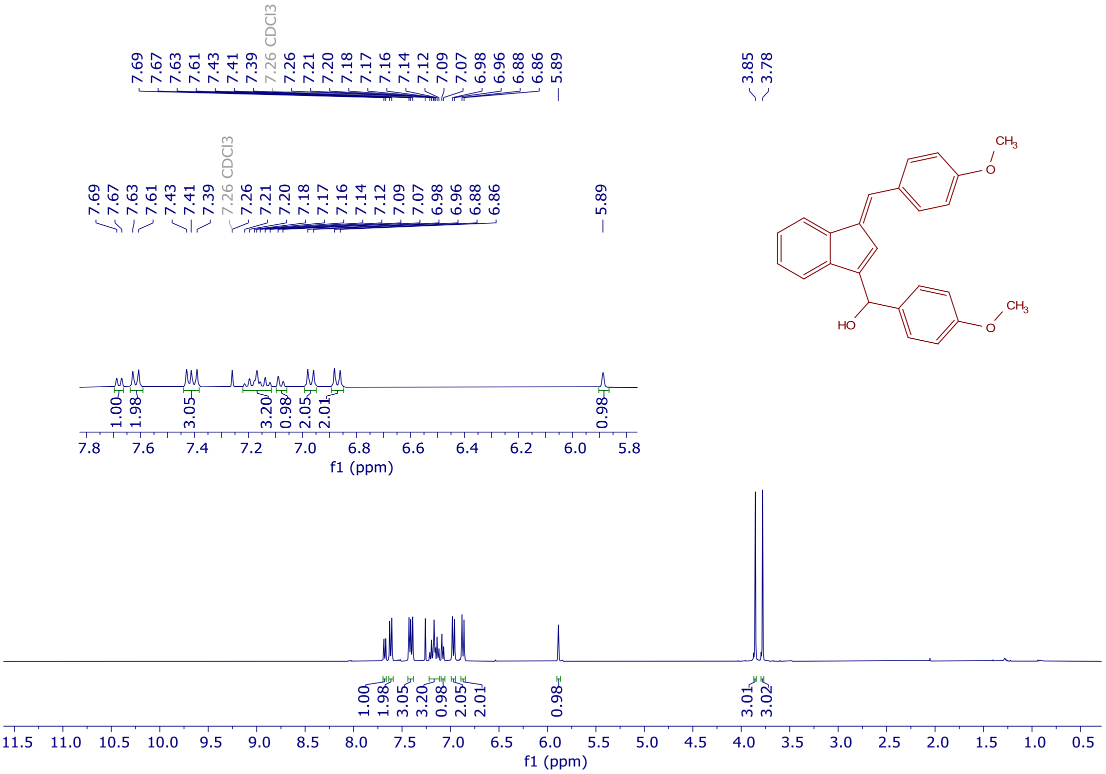
*$^{13}C\{^{1}H\}$ NMR of (E)-(1-benzylidene-1H-inden-3-yl)(phenyl)methanol (Compound-**4a**) in $CDCl_3$*



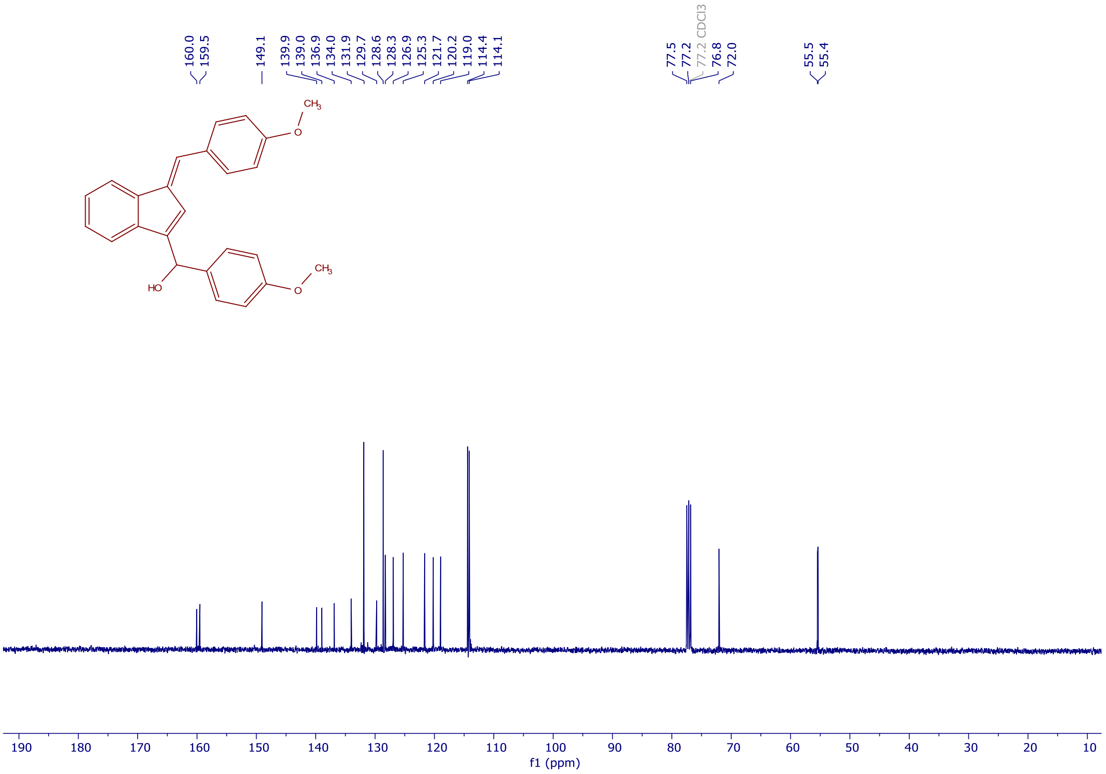
*^{1}H NMR of (E)-(1-(4-methylbenzylidene)-1H-inden-3-yl)(p-tolyl)methanol (Compound-**4b**) in CDCl$_3$*



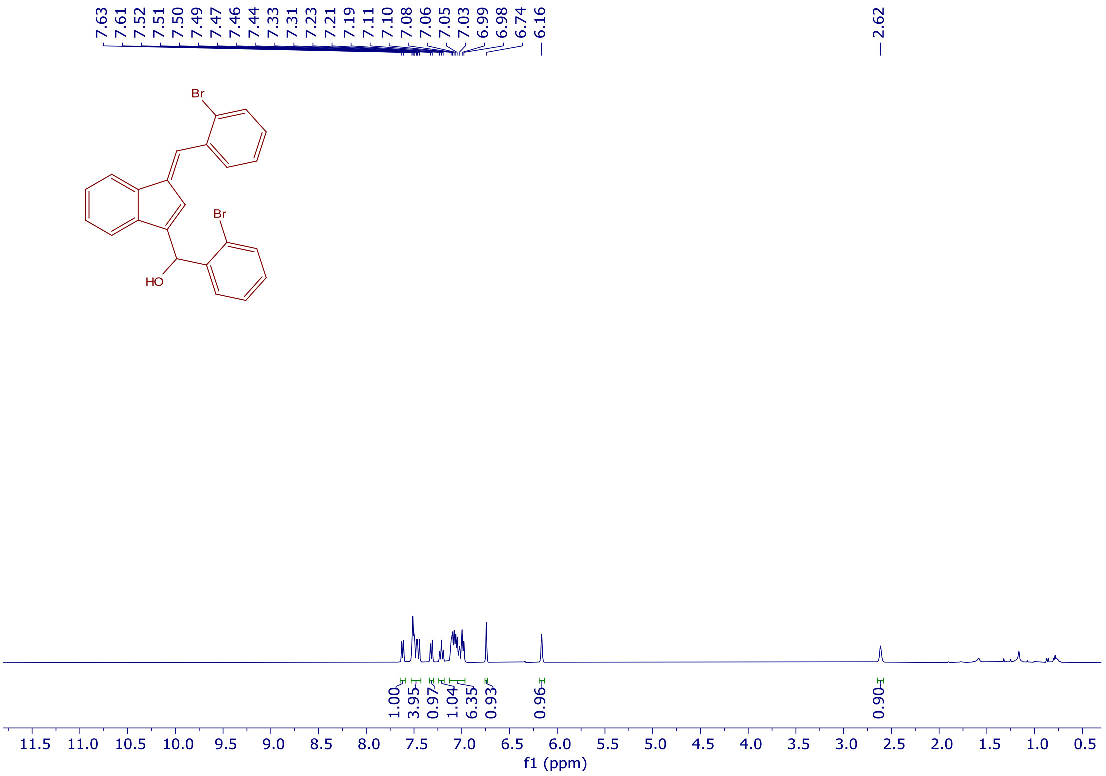
*^{13}C{^{1}H} NMR of (E)-(1-(4-methylbenzylidene)-1H-inden-3-yl)(p-tolyl)methanol (Compound-**4b**) in CDCl$_3$*



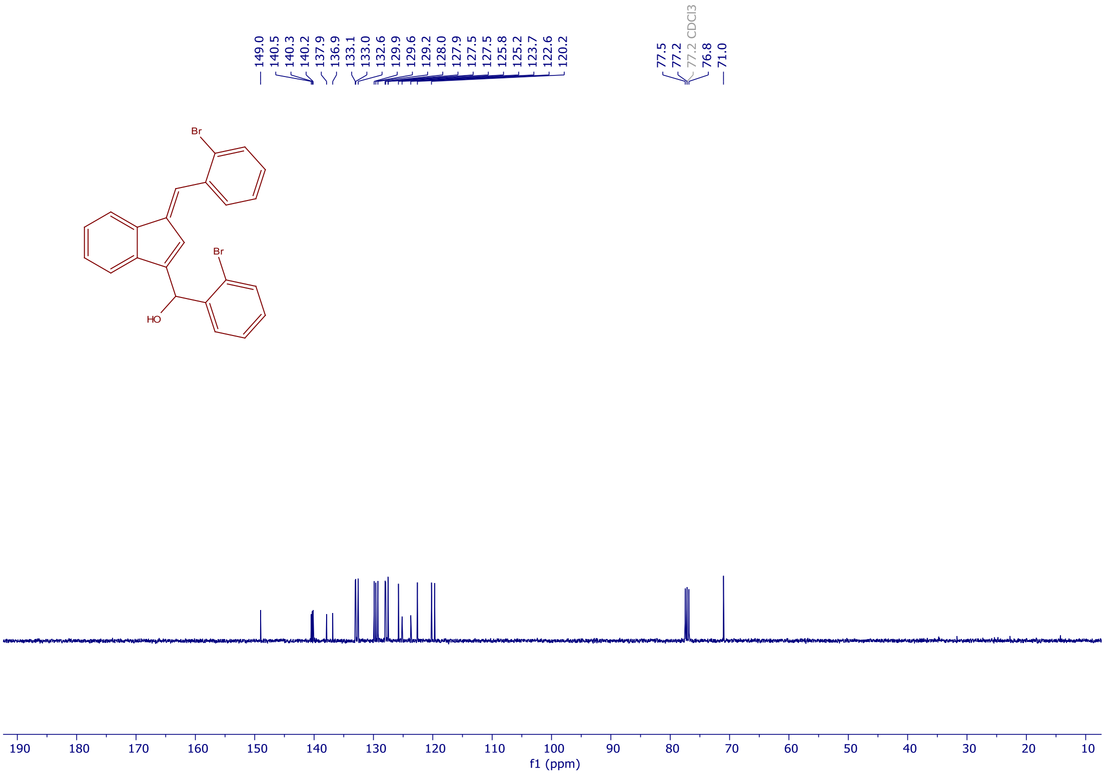
*^{1}H NMR of (E)-(1-(4-methoxybenzylidene)-1H-inden-3-yl)(4-methoxyphenyl)methanol (Compound-**4c**) in $CDCl_3$*



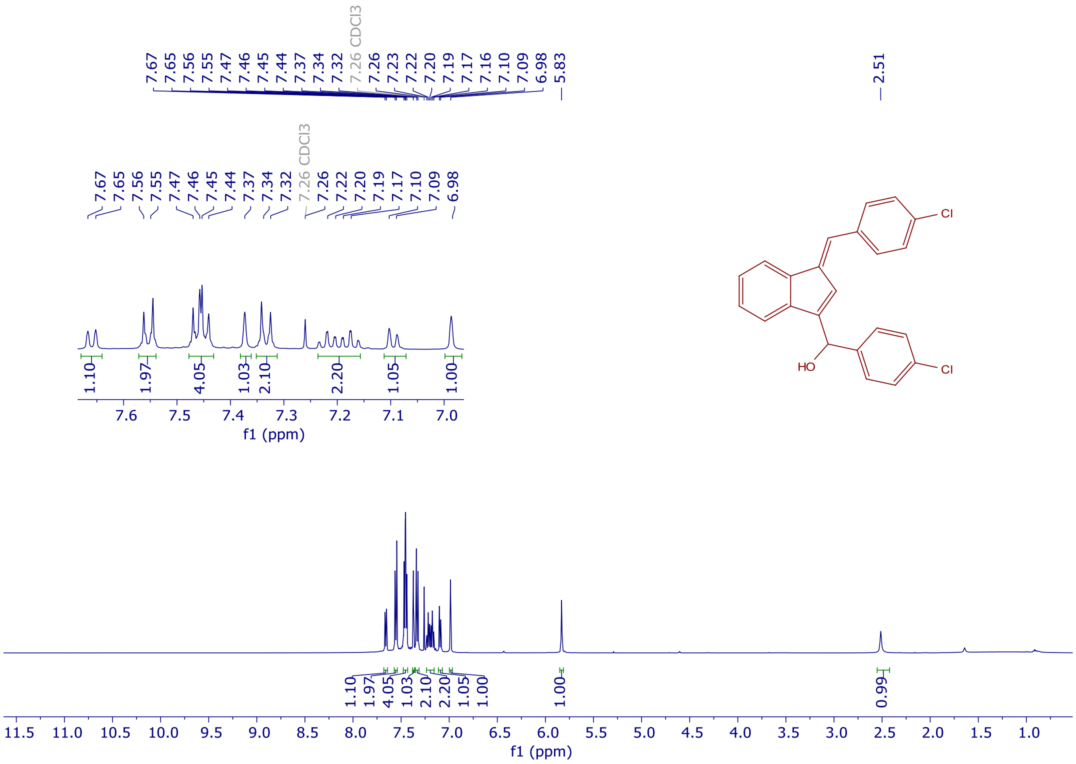
*^{13}C{^{1}H} NMR of (E)-(1-(4-methoxybenzylidene)-1H-inden-3-yl)(4-methoxyphenyl)methanol (Compound-**4c**) in $CDCl_3$*



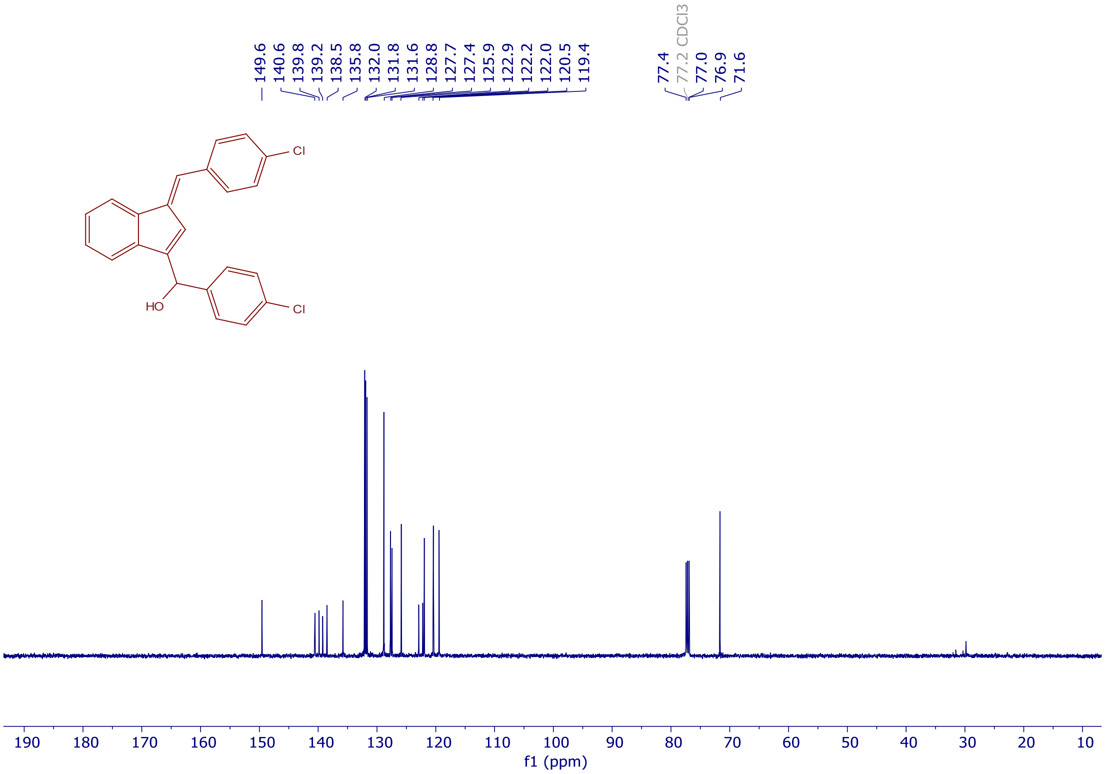
^{1}H NMR of (E)-(1-(2-bromobenzylidene)-1H-inden-3-yl)(2-bromophenyl)methanol (Compound-**4d**) in $CDCl_3$



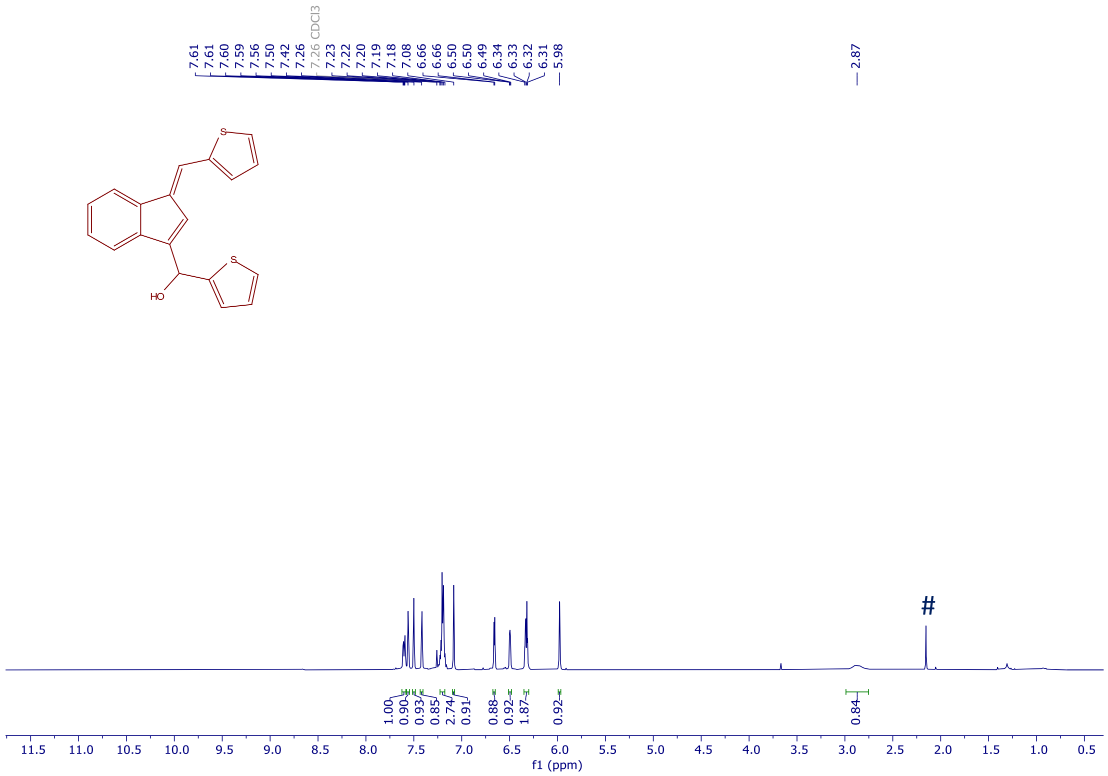
$^{13}C\{^{1}H\}$ NMR of (E)-(1-(2-bromobenzylidene)-1H-inden-3-yl)(2-bromophenyl)methanol (Compound-**4d**) in $CDCl_3$



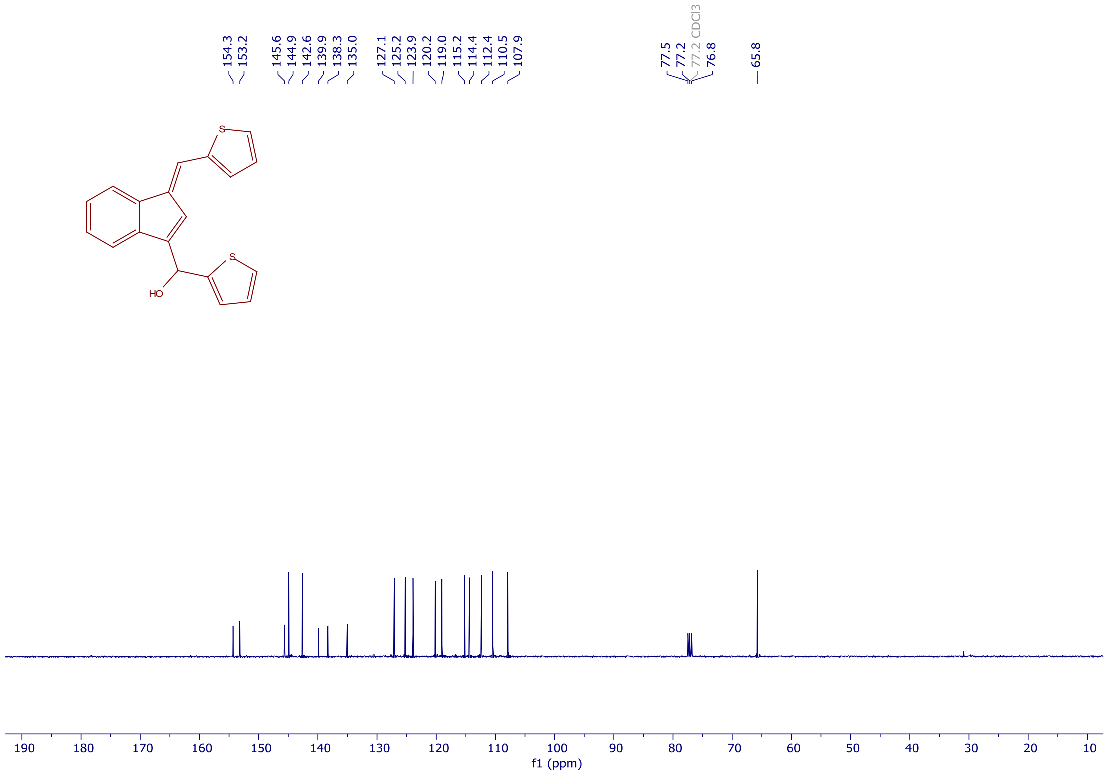
*^{1}H NMR of (E)-(1-(4-chlorobenzylidene)-1H-inden-3-yl)(4-chlorophenyl)methanol (Compound-**4e**) in $CDCl_3$*



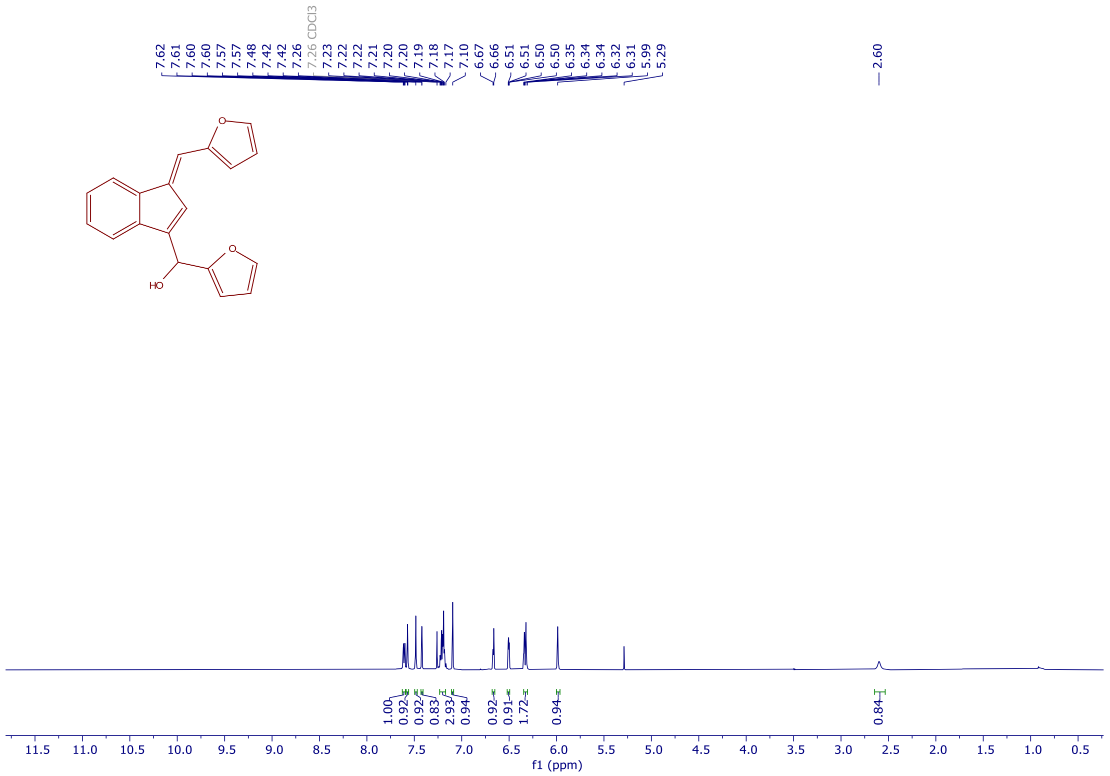
*^{13}C{^{1}H} NMR of (E)-(1-(4-chlorobenzylidene)-1H-inden-3-yl)(4-chlorophenyl)methanol (Compound-**4e**) in $CDCl_3$*



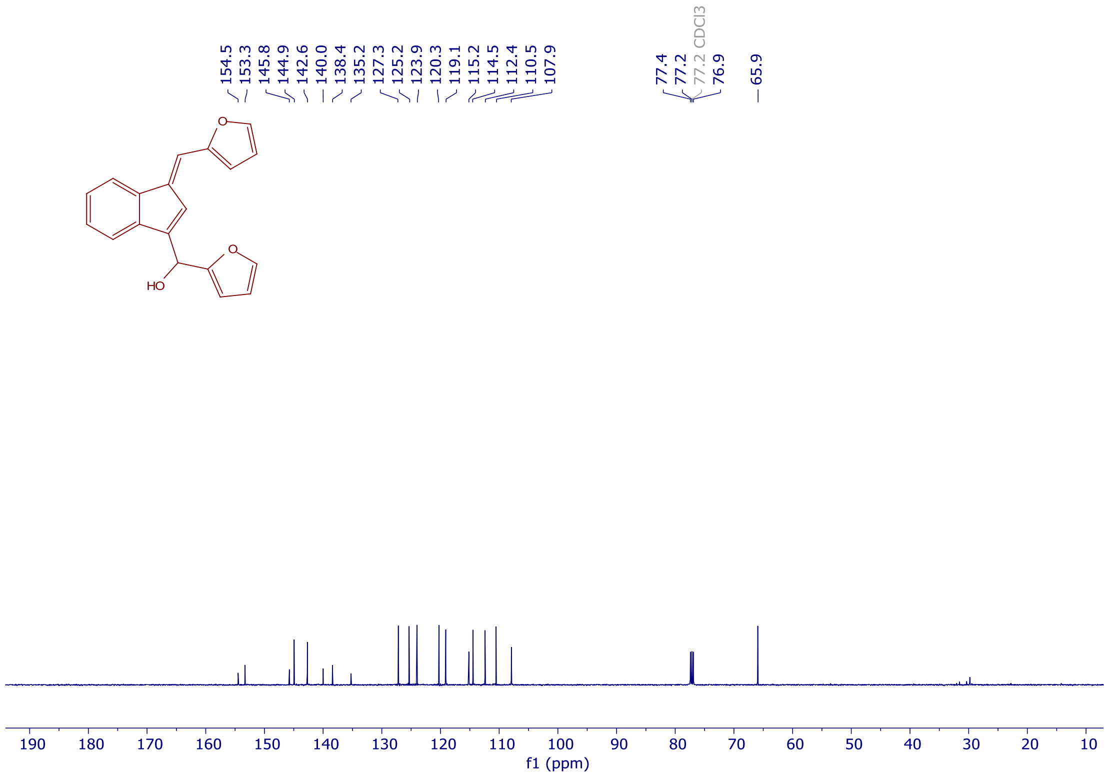
*^{1}H NMR of (E)-thiophen-2-yl(1-(thiophen-2-ylmethylene)-1H-inden-3-yl)methanol. (Compound-**4f**). # indicates the solvent impurity of acetone in CDCl$_3$*



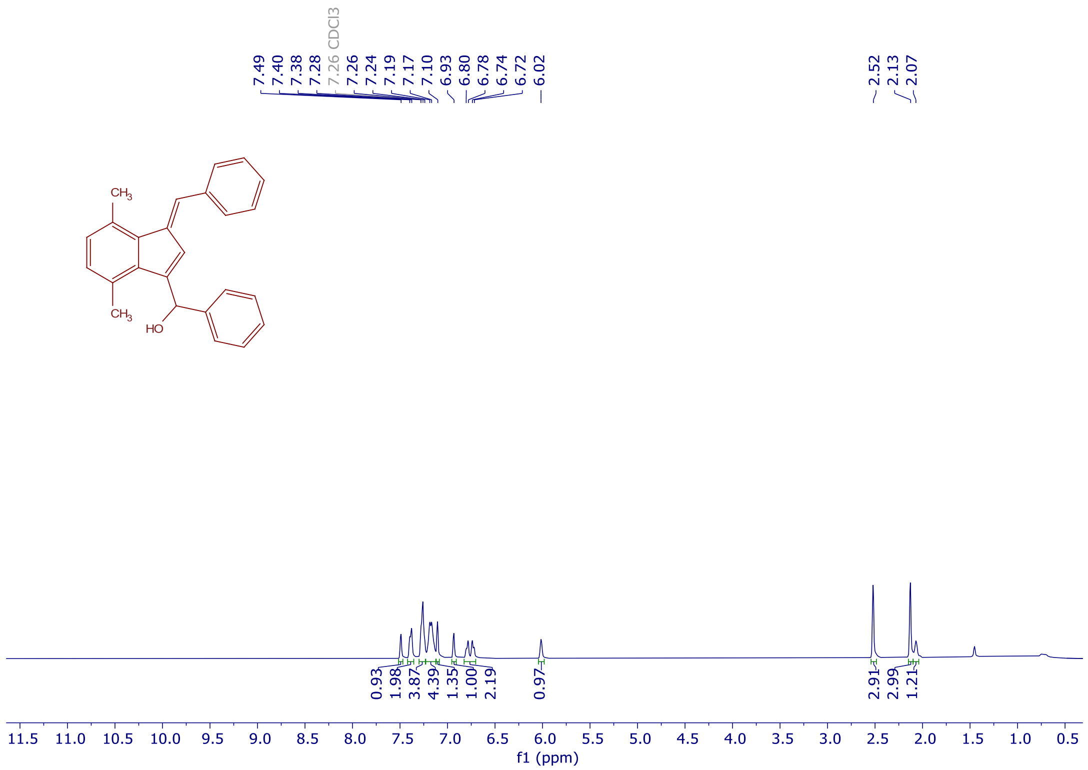
*^{13}C{^{1}H} NMR of (E)-furan-2-yl(1-(furan-2-ylmethylene)-1H-inden-3-yl)methanol (Compound-**4f**) in CDCl$_3$*



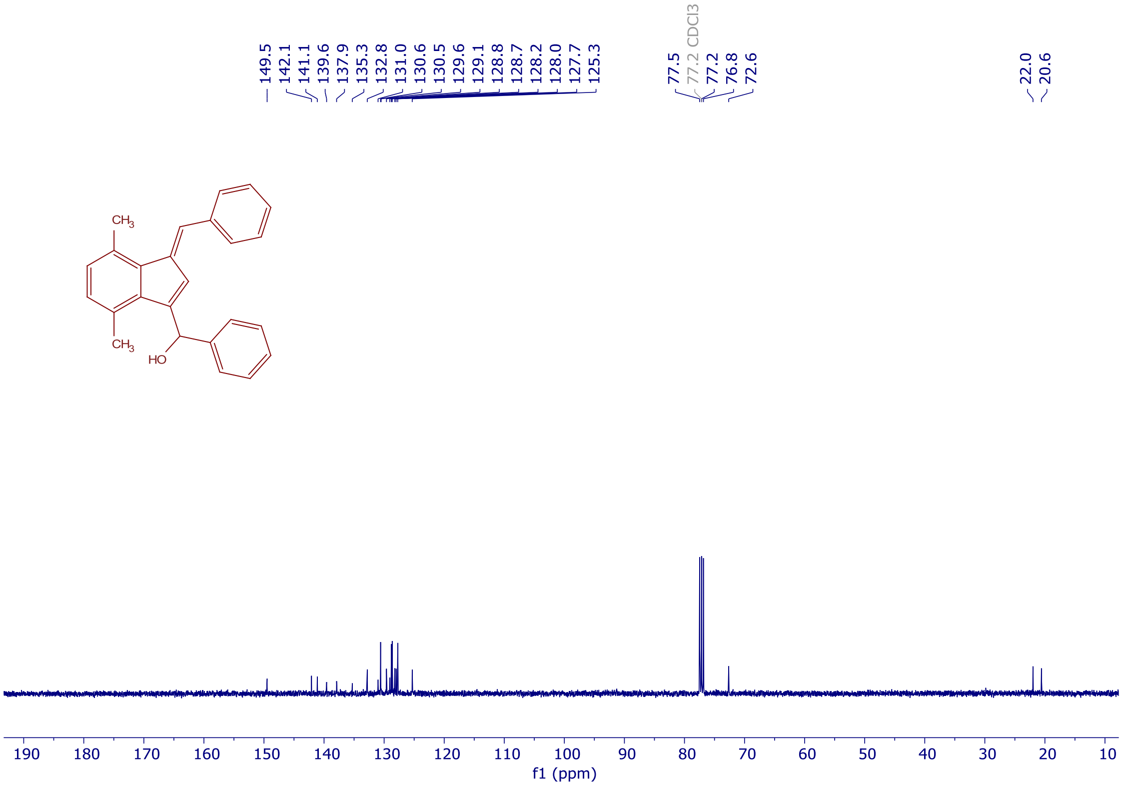
*^{1}H NMR of (E)-furan-2-yl(1-(furan-2-ylmethylene)-1H-inden-3-yl)methanol (Compound-**4g**) in CDCl$_3$*



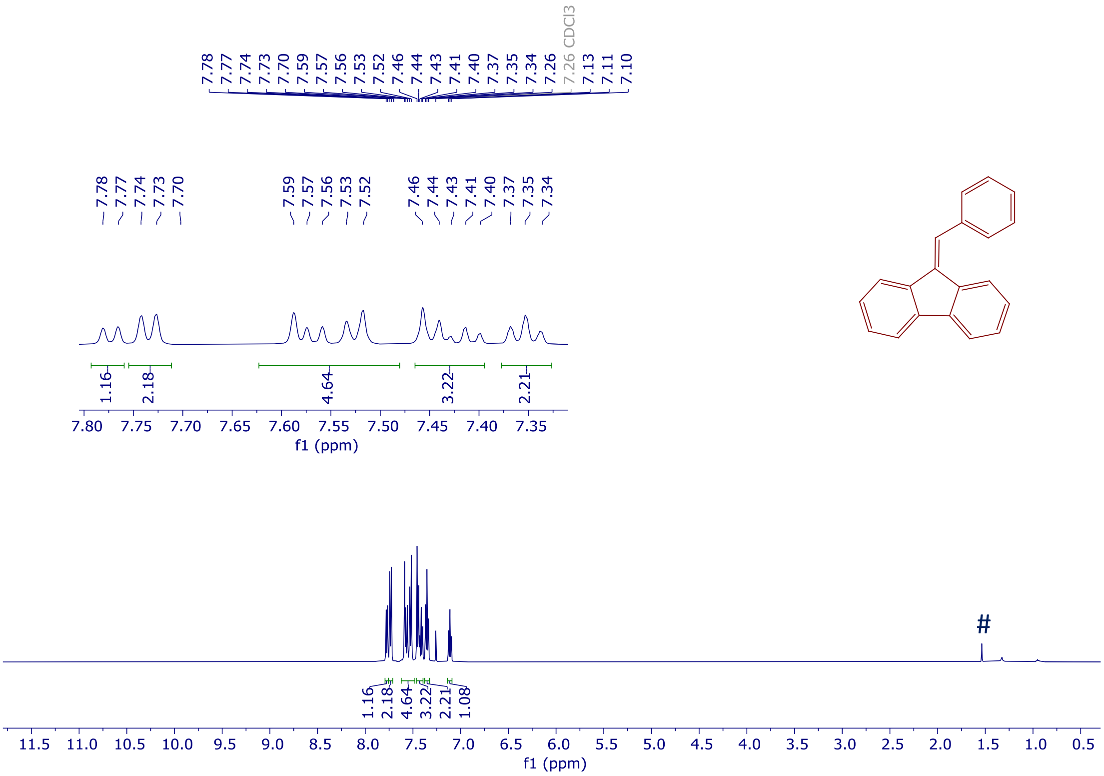
*^{13}C{^{1}H} NMR of (E)-furan-2-yl(1-(furan-2-ylmethylene)-1H-inden-3-yl)methanol (Compound-**4g**) in CDCl$_3$*



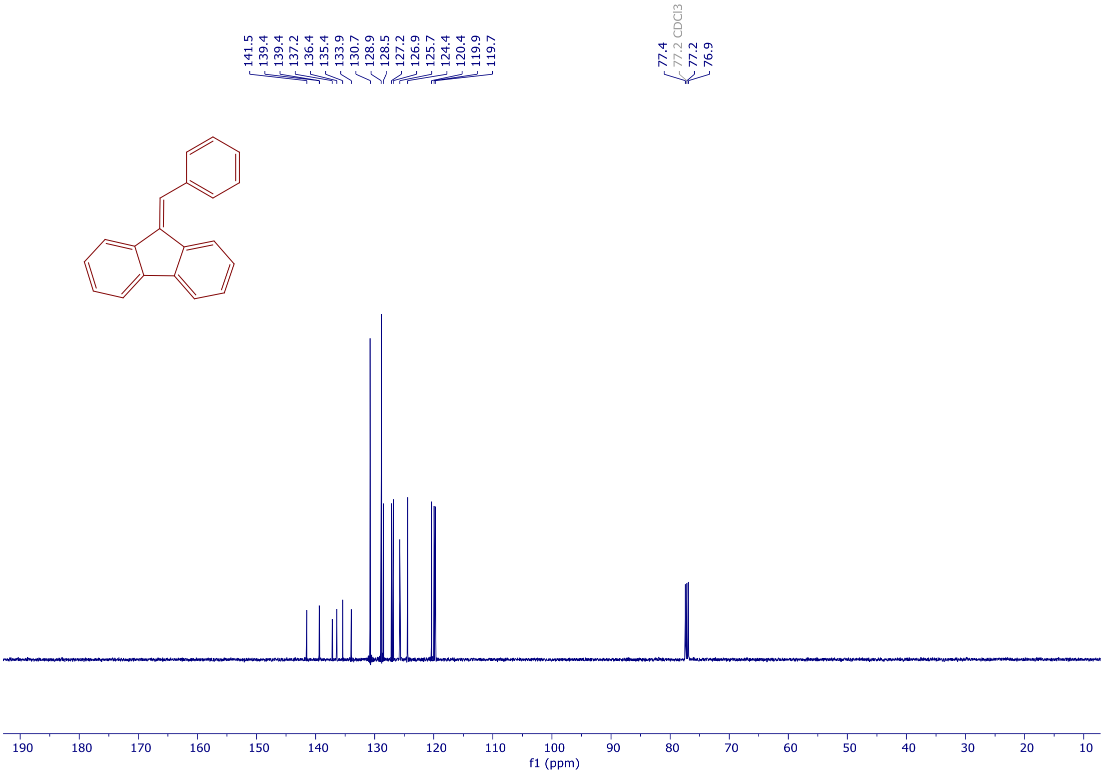
*^{1}H NMR of (E)- (1-benzylidene-4,7-dimethyl-1H-inden-3-yl)(phenyl)methanol (Compound-**4h**) in CDCl$_3$*



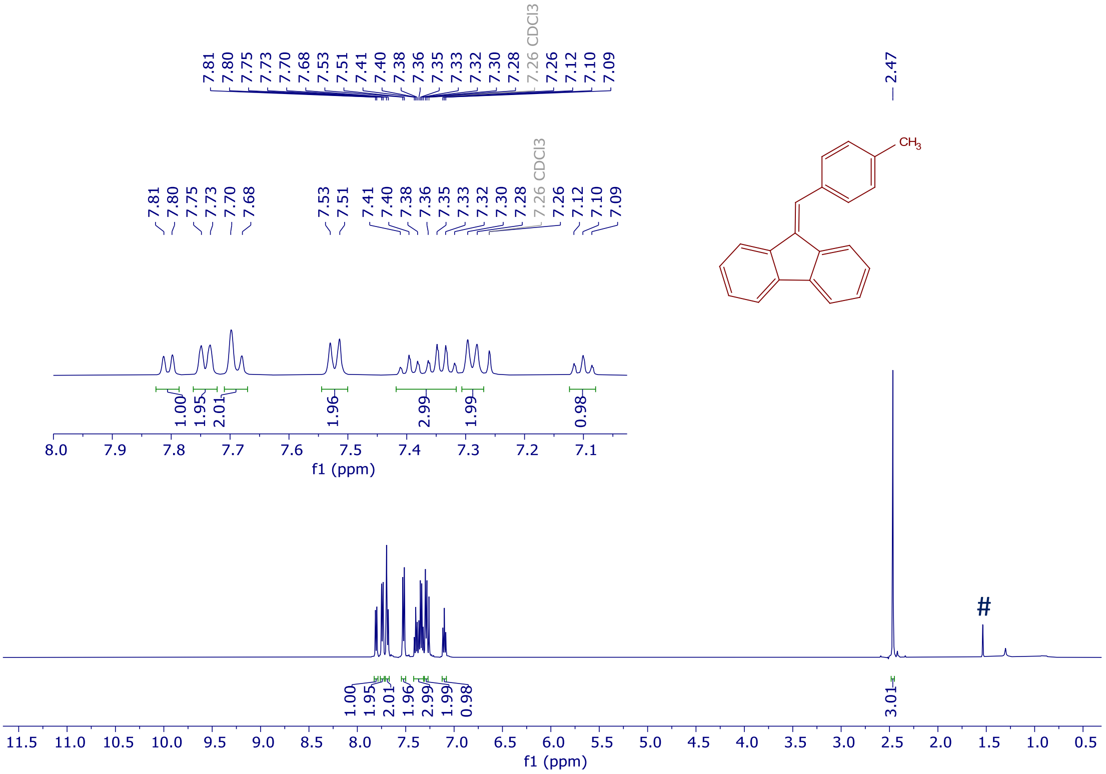
*^{13}C{^{1}H} NMR of (E)-(1-benzylidene-4,7-dimethyl-1H-inden-3-yl)(phenyl)methanol (Compound-**4h**) in CDCl$_3$*



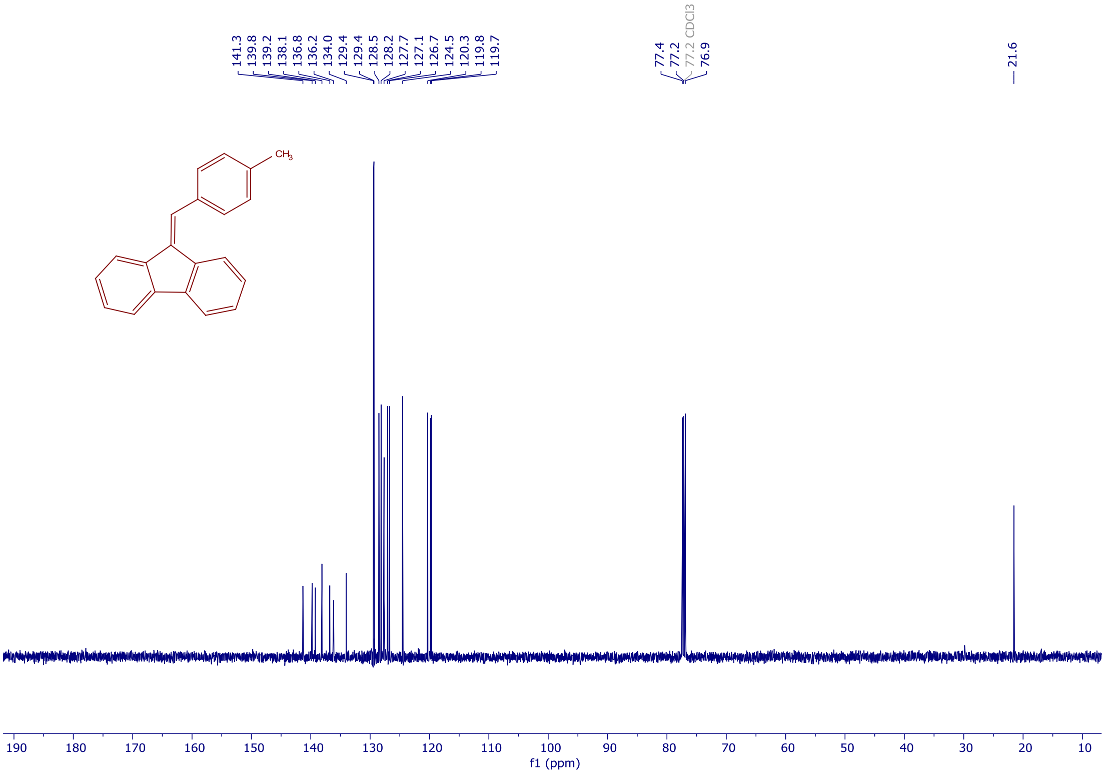
*^{1}H NMR of 9-benzylidene-9H-fluorene (Compound-**6a**) in CDCl$_3$. # indicates the solvent impurity of H_2O in CDCl$_3$*



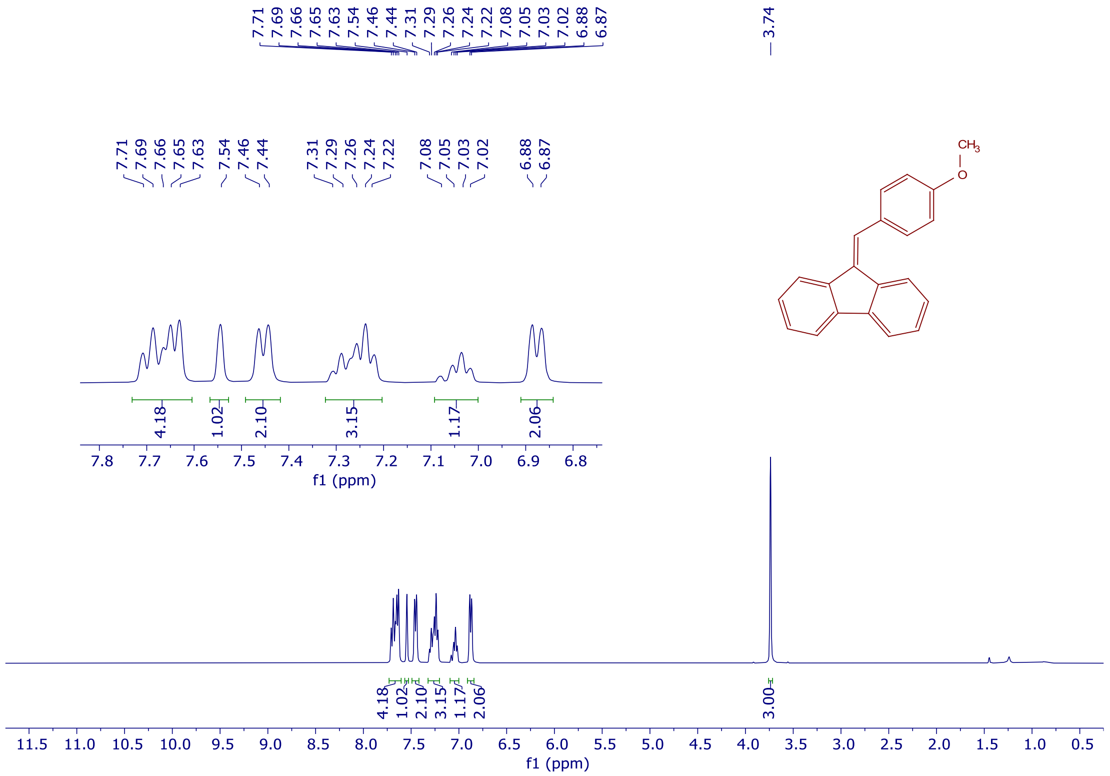
*^{13}C{^{1}H} NMR of 9-benzylidene-9H-fluorene(Compound-**6a**) in CDCl$_3$*



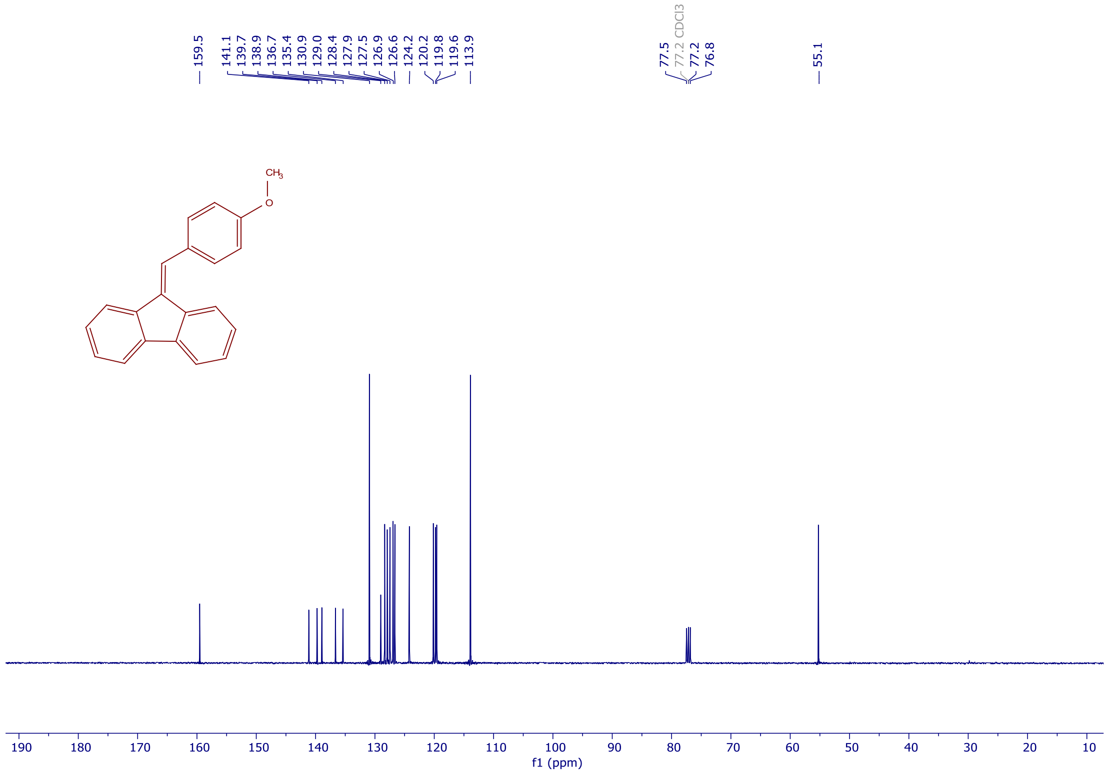
^{1}H NMR of 9-(4-methylbenzylidene)-9H-fluorene (Compound-**6b**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$



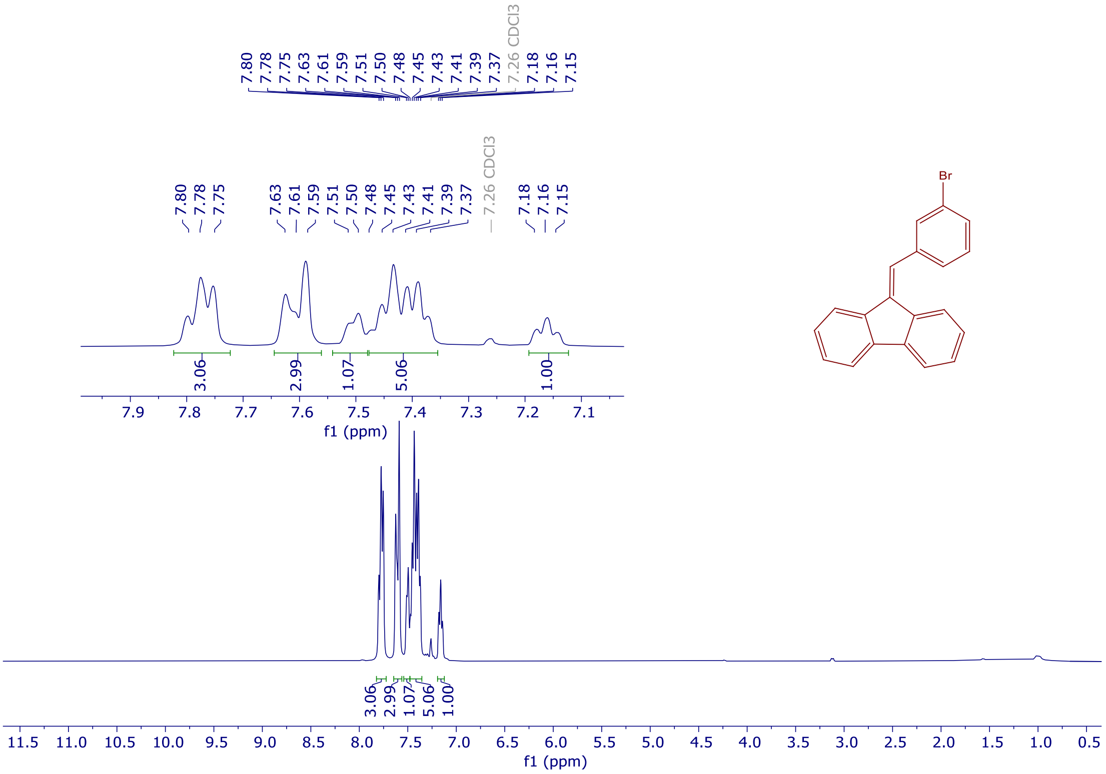
$^{13}C\{^1H\}$ NMR of 9-(4-methylbenzylidene)-9H-fluorene (Compound-**6b**) in $CDCl_3$



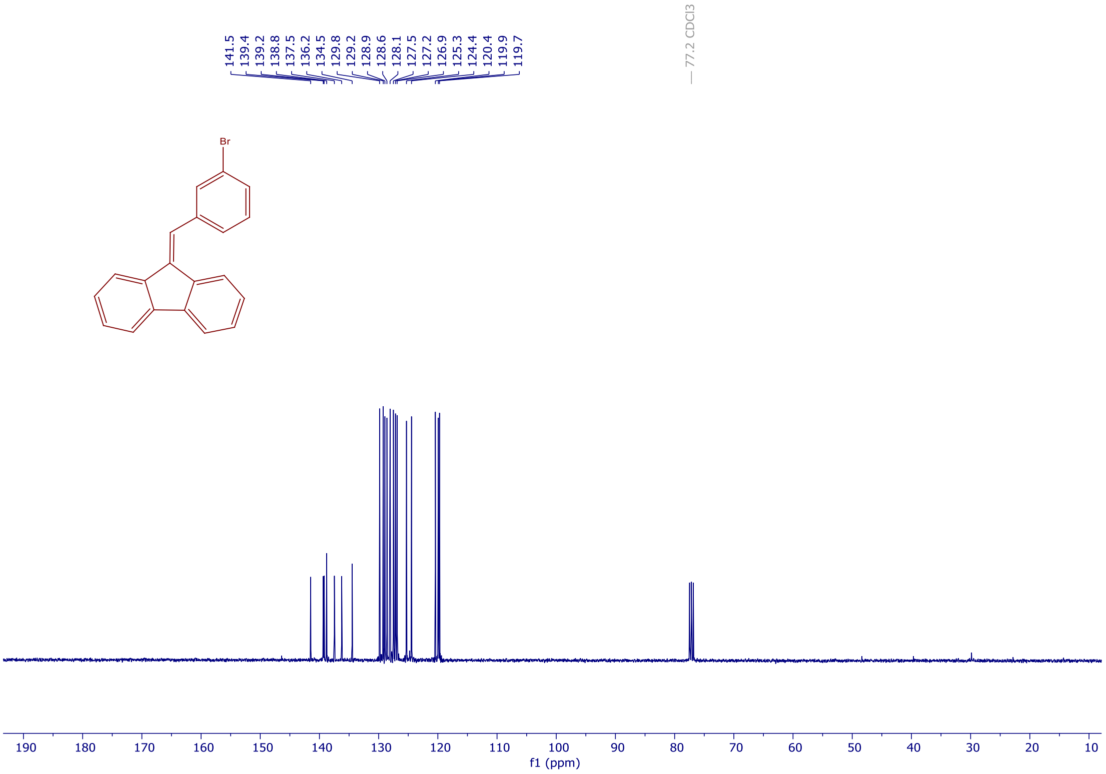
*^{1}H NMR of 9-(4-methoxybenzylidene)-9H-fluorene (Compound-**6c**) in CDCl$_3$*



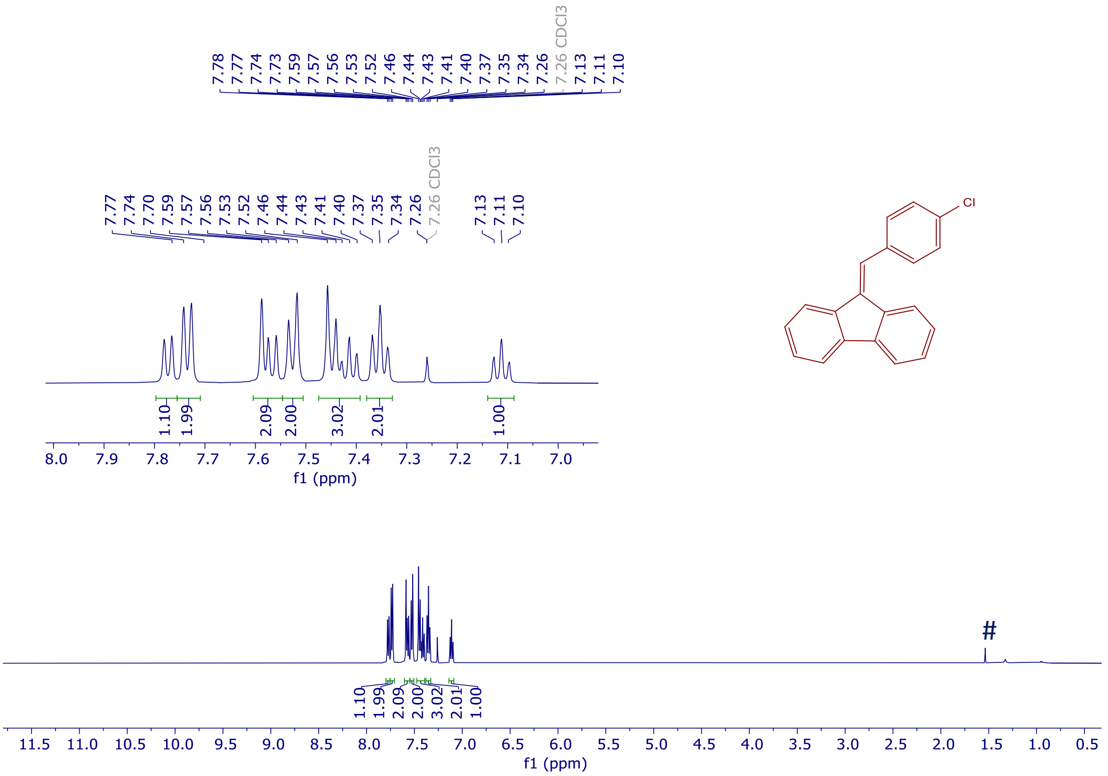
*^{13}C{^{1}H} NMR of 9-(4-methoxybenzylidene)-9H-fluorene (Compound-**6c**) in CDCl$_3$*



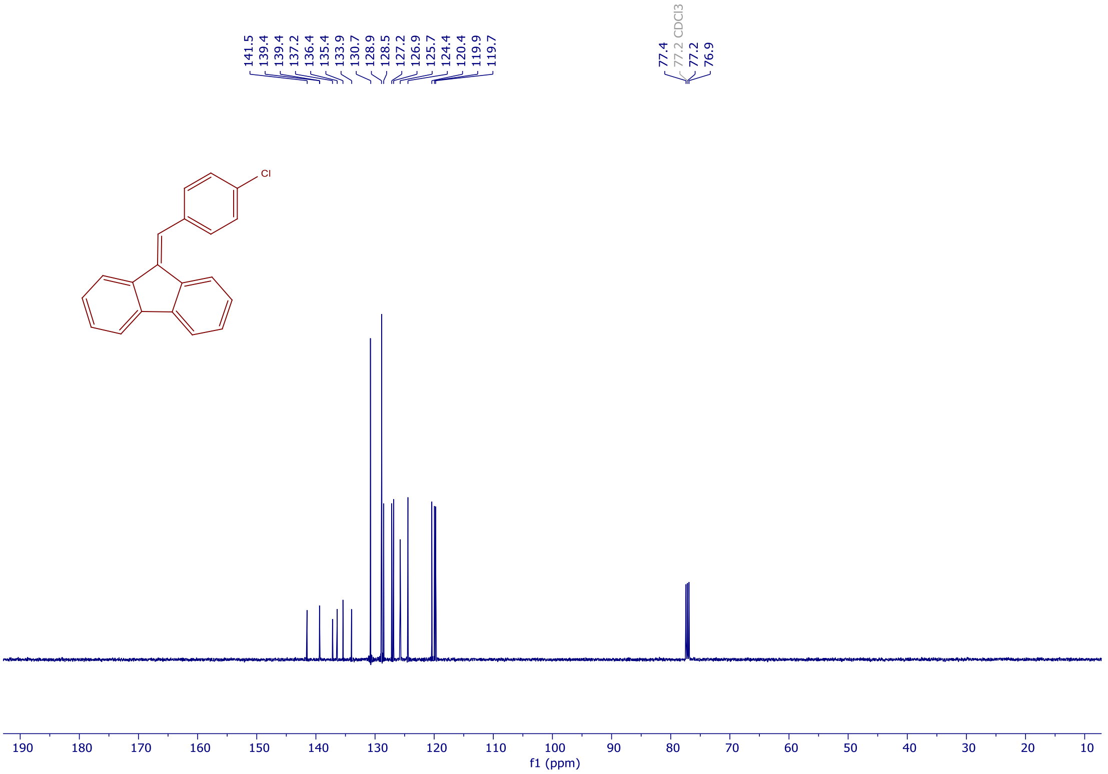
*^{1}H NMR of 9-(3-bromobenzylidene)-9H-fluorene (Compound-**6d**) in CDCl$_3$*



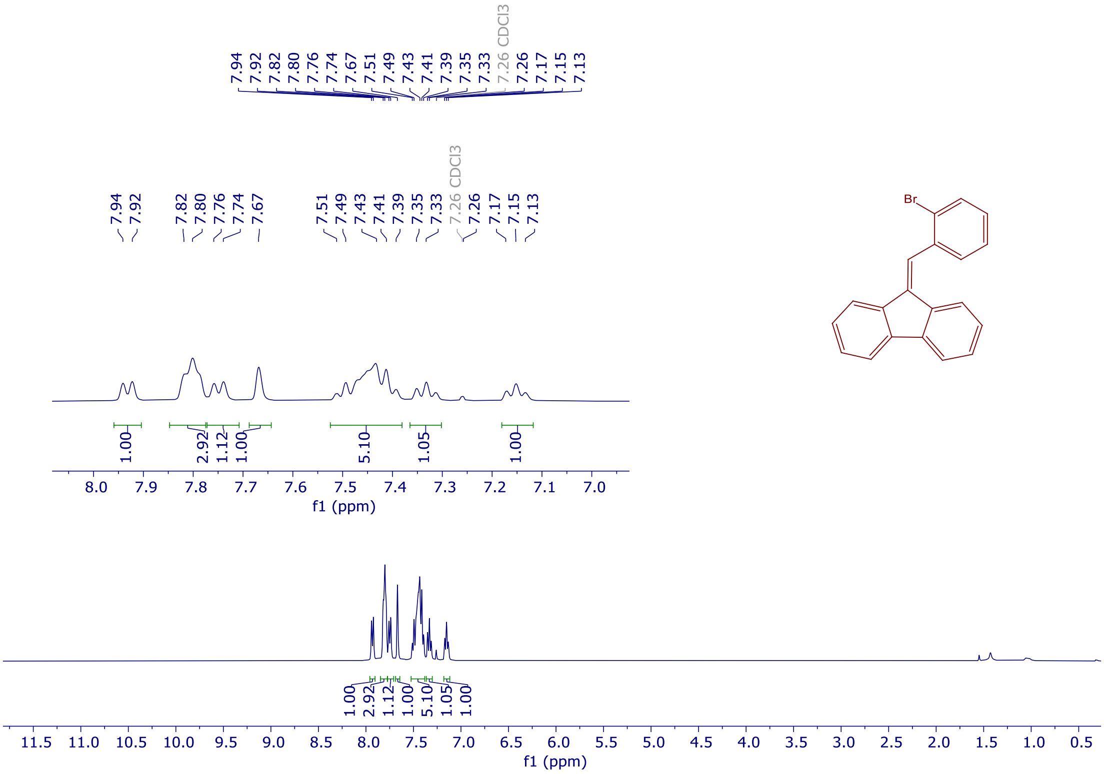
*^{13}C{^{1}H} NMR of 9-(3-bromobenzylidene)-9H-fluorene (Compound-**6d**) in CDCl$_3$*



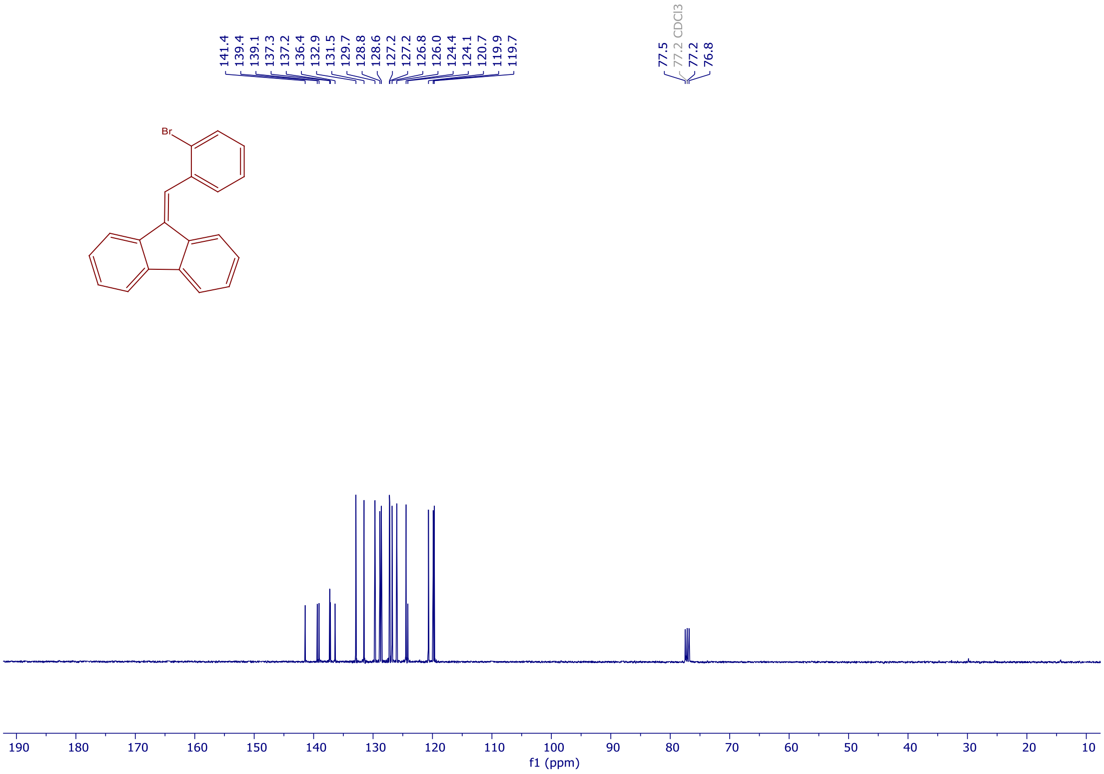
*^{1}H NMR of 9-(4-chlorobenzylidene)-9H-fluorene (Compound-**6e**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



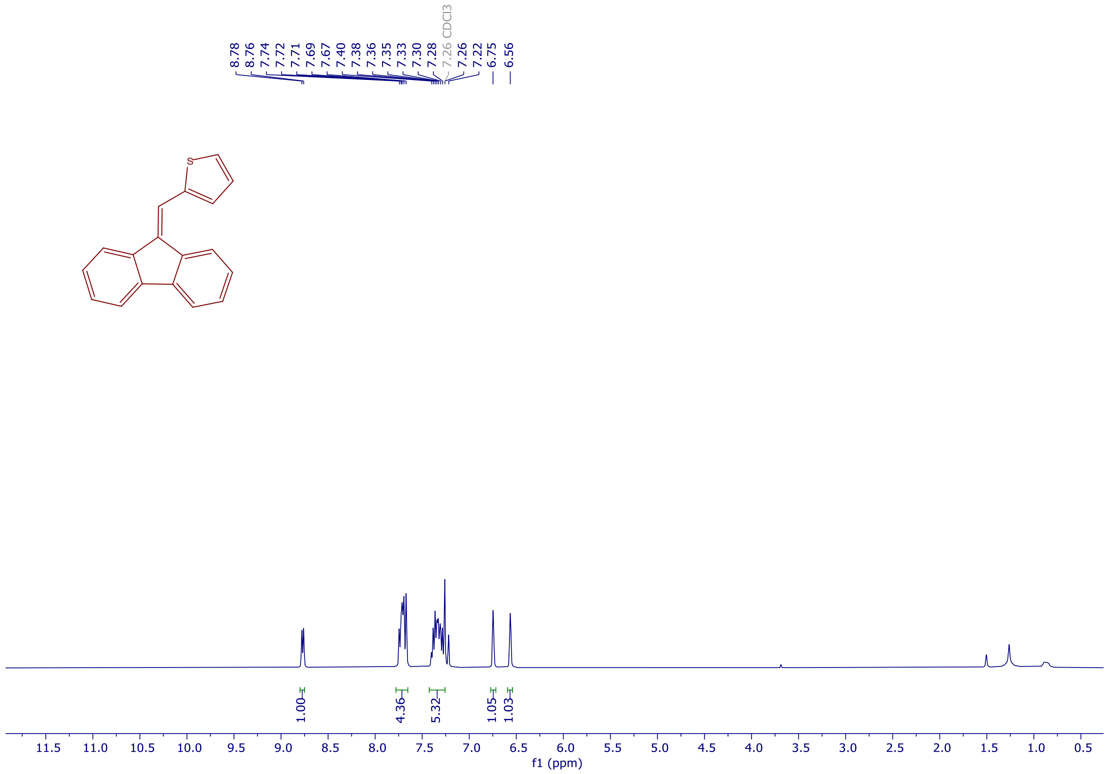
*$^{13}C\{^1H\}$ NMR of 9-(4-chlorobenzylidene)-9H-fluorene (Compound-**6e**) in $CDCl_3$*



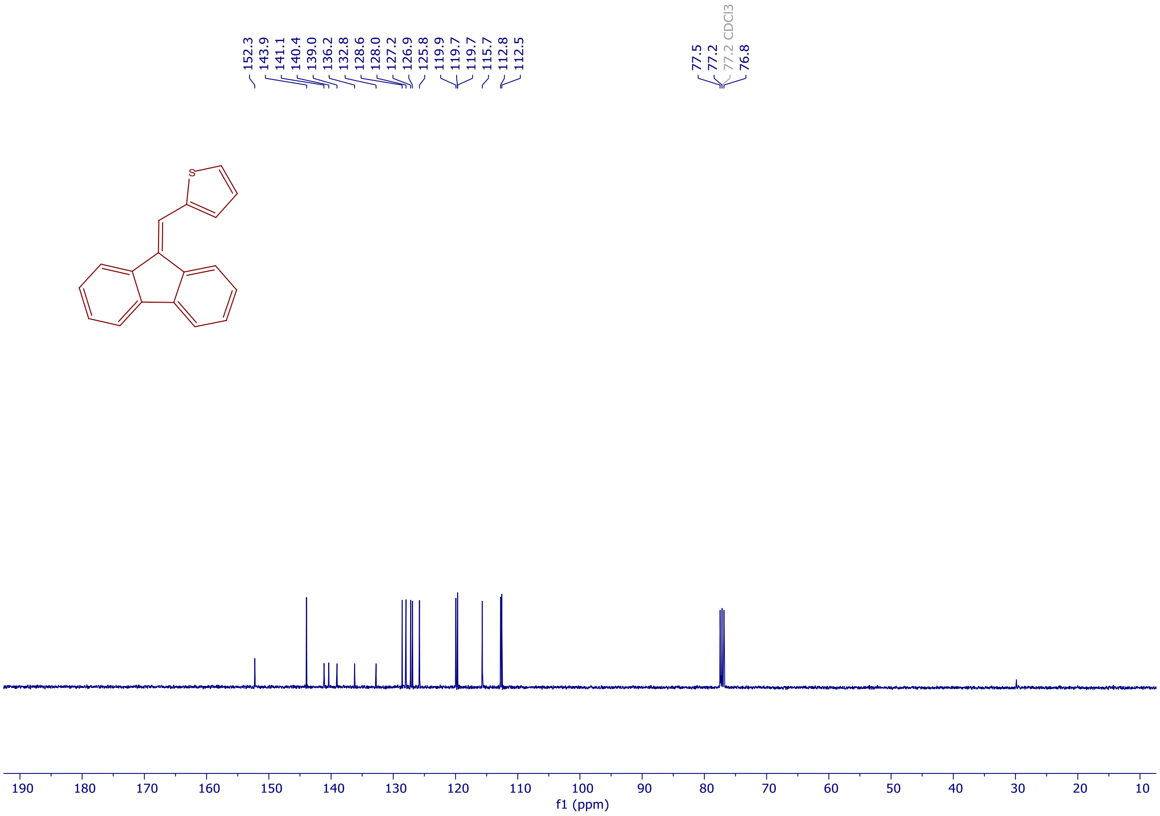
*^{1}H NMR of 9-(2-bromobenzylidene)-9H-fluorene (Compound-**6f**) in CDCl$_3$*



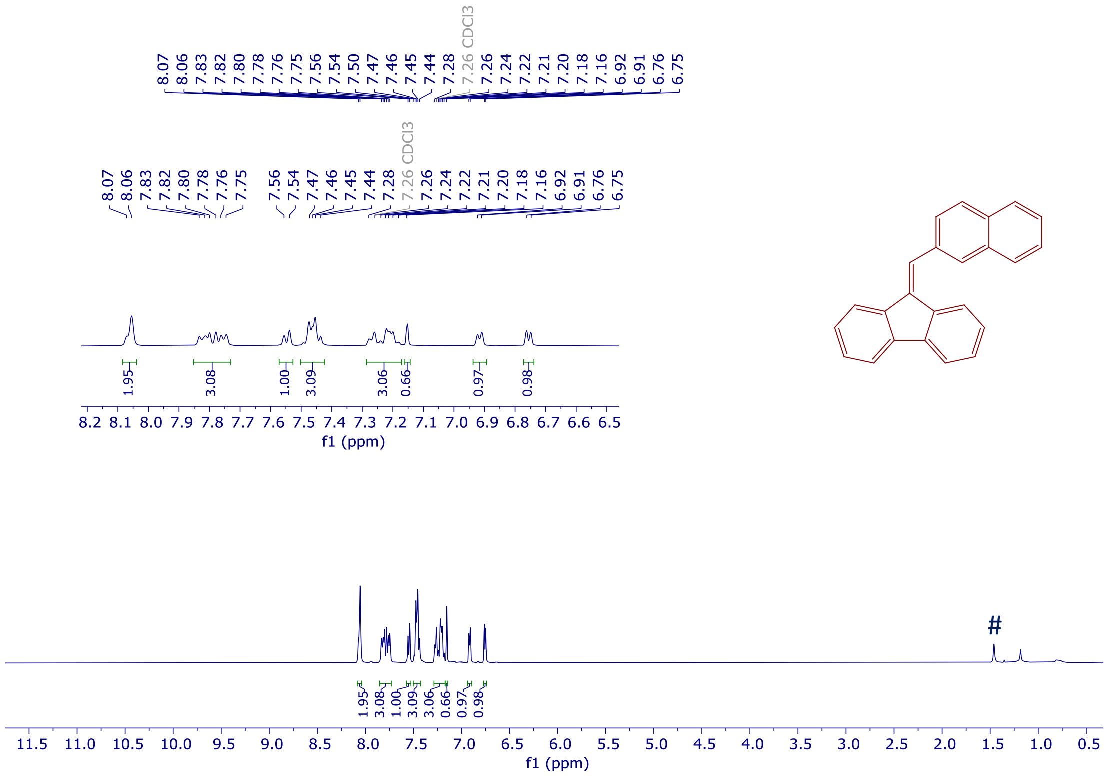
*^{13}C{^{1}H} NMR of 9-(2-bromobenzylidene)-9H-fluorene (Compound-**6f**) in CDCl$_3$*



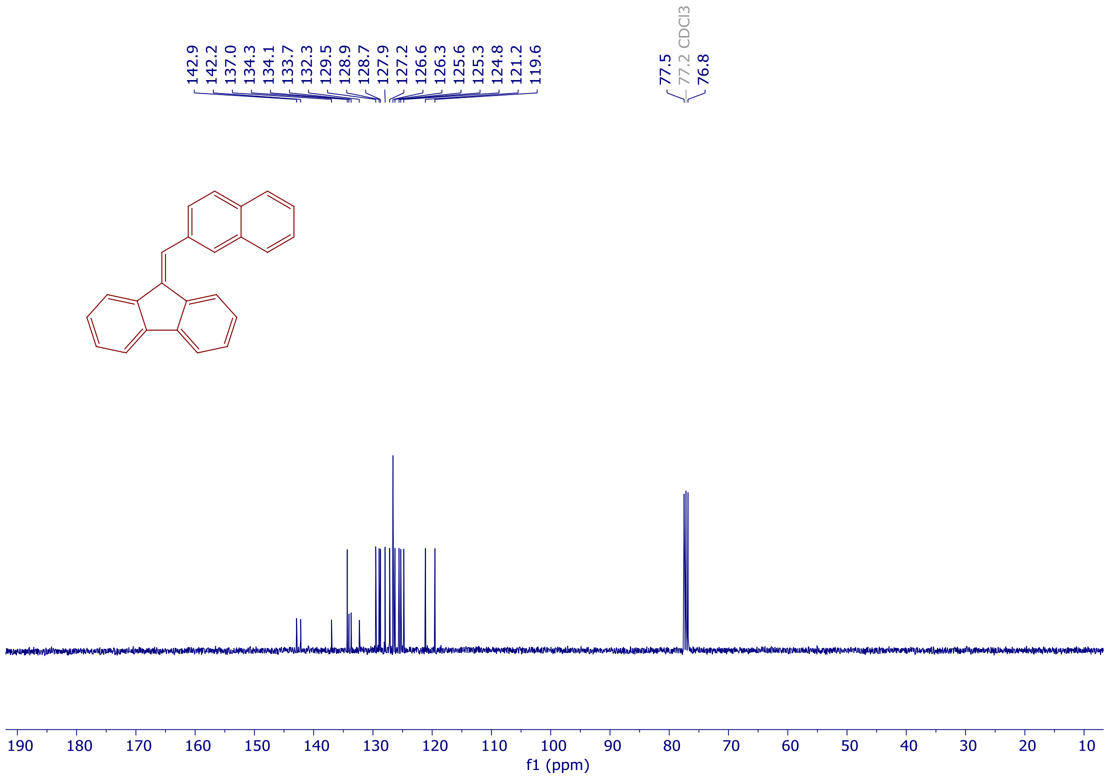
*^{1}H NMR of 2-((9H-fluoren-9-ylidene)methyl)thiophene (Compound-**6g**) in $CDCl_3$*



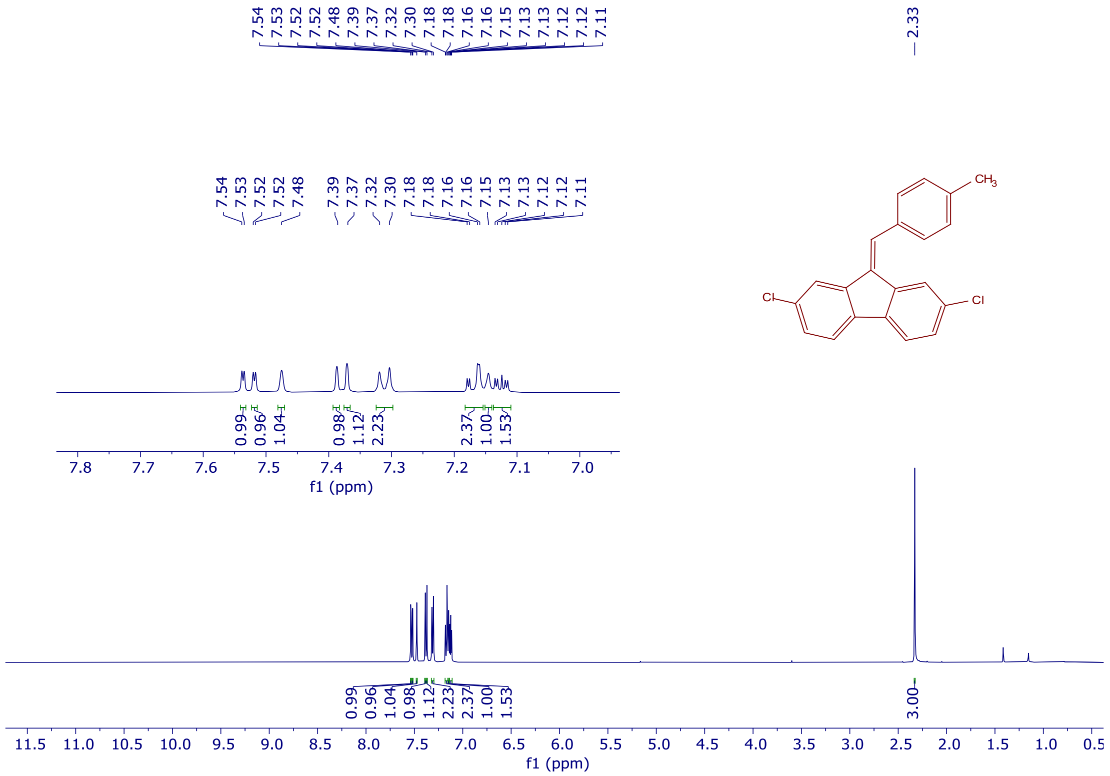
*$^{13}C\{^{1}H\}$ NMR of 2-((9H-fluoren-9-ylidene)methyl)thiophene (Compound-**6g**) in $CDCl_3$*



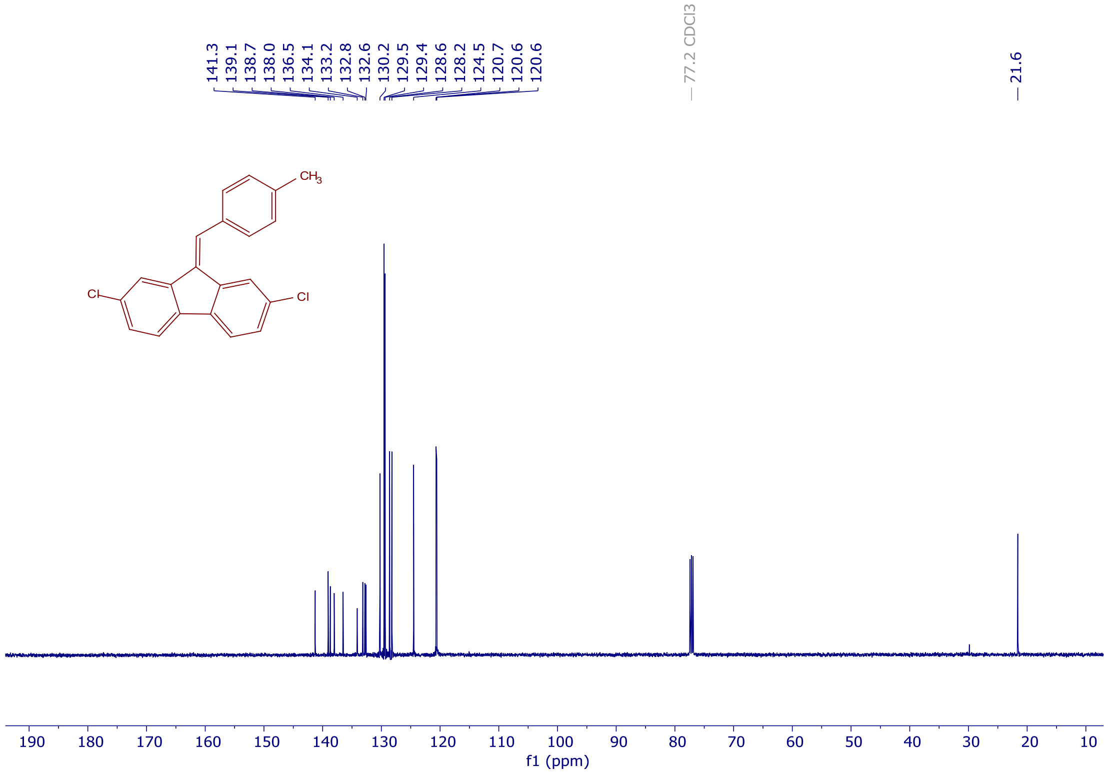
*^{1}H NMR of 9-(naphthalen-2-ylmethylene)-9H-fluorene (Compound-**6h**) in CDCl$_3$. # indicates the solvent impurity of H_2O in CDCl$_3$*



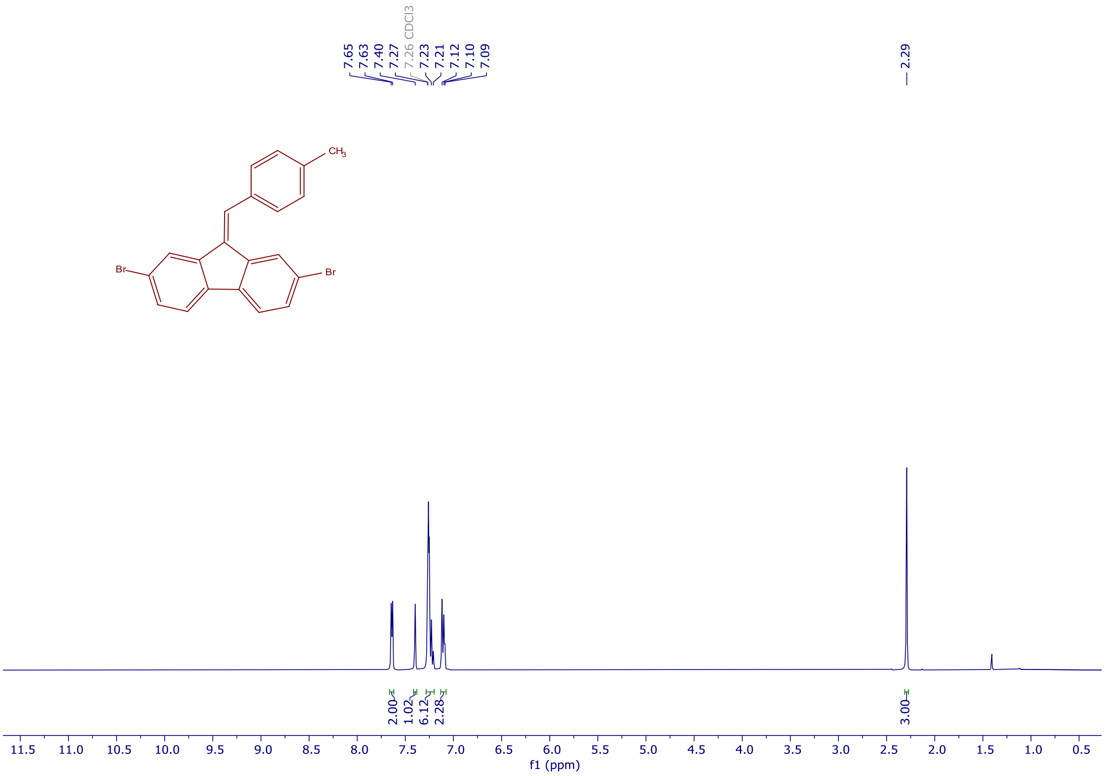
*^{13}C{^{1}H} NMR of 9-(naphthalen-2-ylmethylene)-9H-fluorene(Compound-**6h**) in CDCl$_3$*



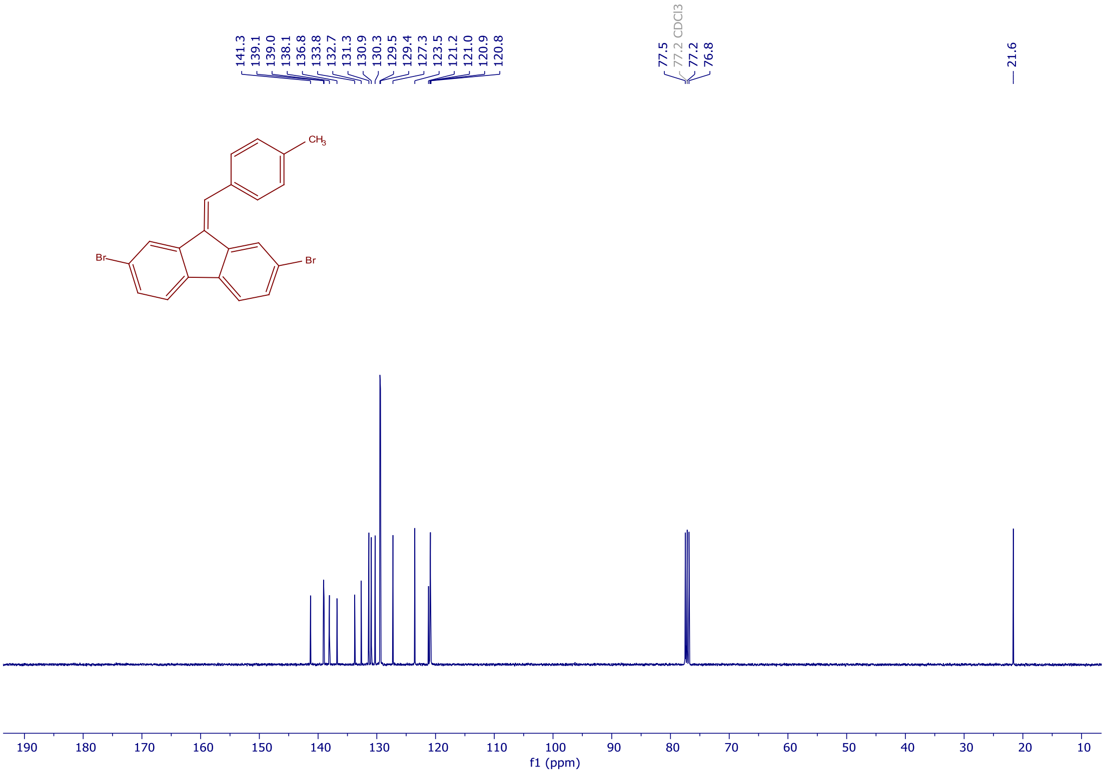
*^{1}H NMR of 2,7-dichloro-9-(4-methylbenzylidene)-9H-fluorene (Compound-**6i**) in CDCl$_3$*



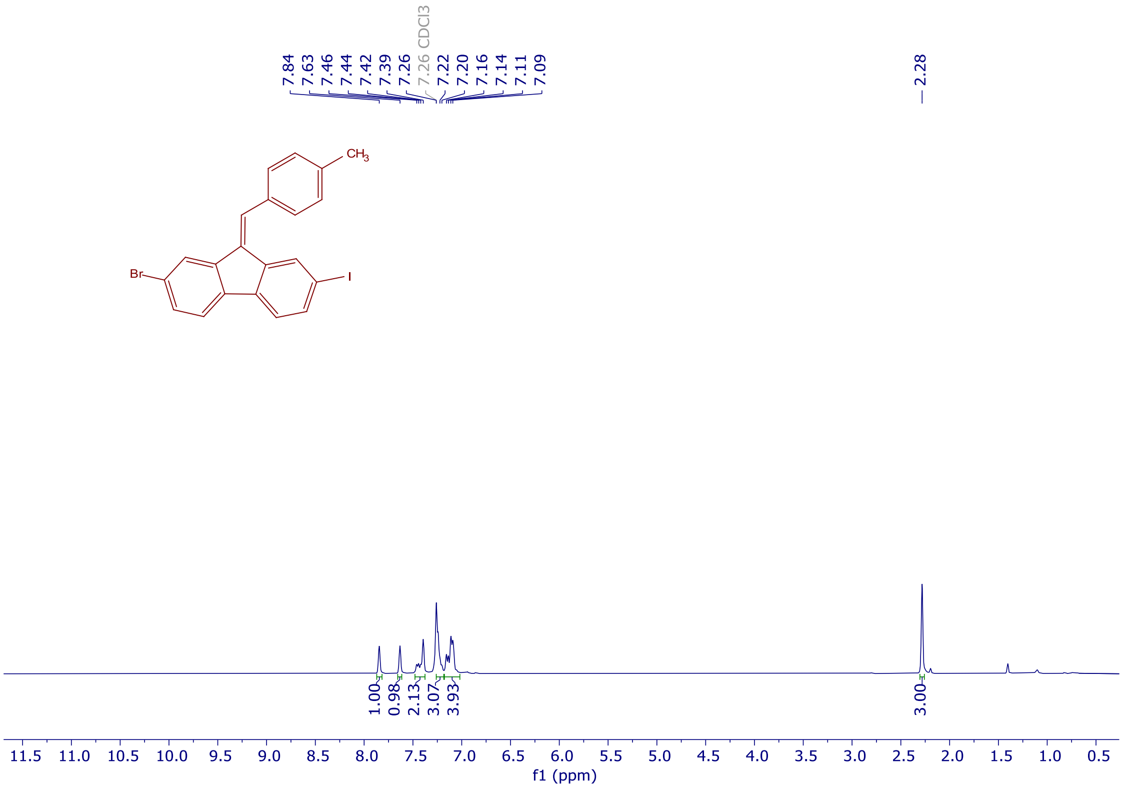
*^{13}C{^{1}H} NMR of 2,7-dichloro-9-(4-methylbenzylidene)-9H-fluorene (Compound-**6i**) in CDCl$_3$*



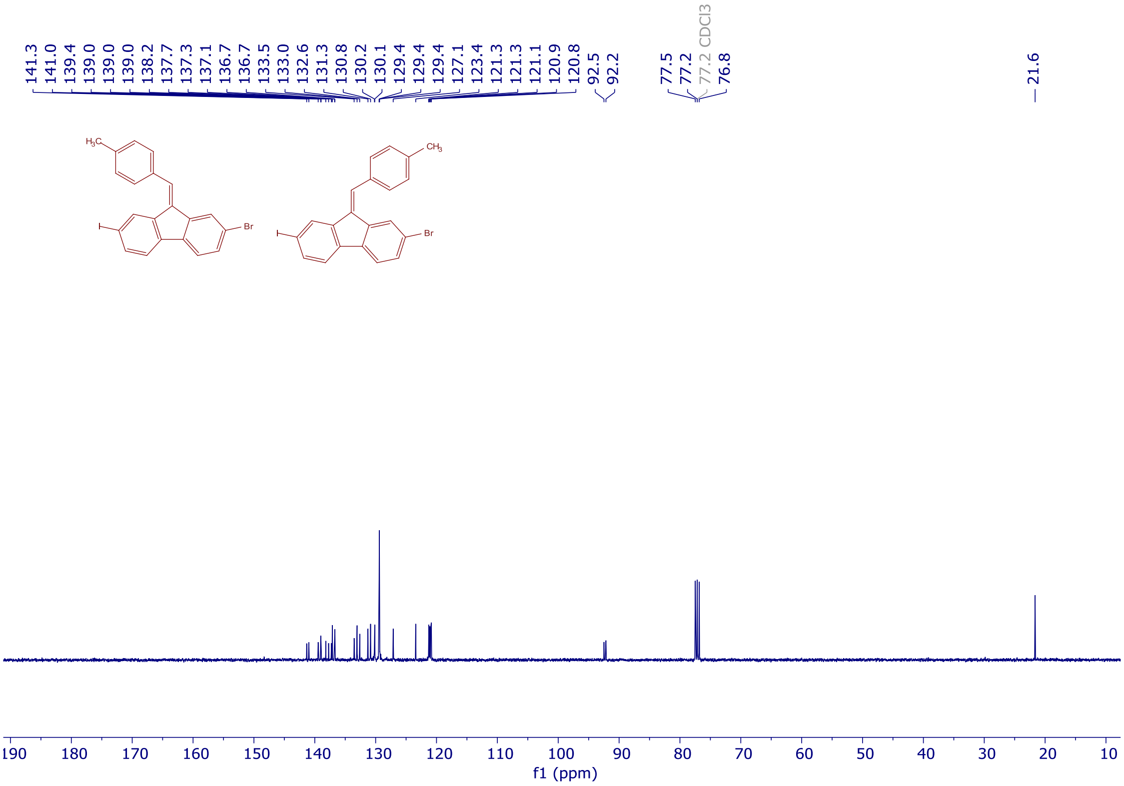
*^{1}H NMR of 2,7-dibromo-9-(4-methylbenzylidene)-9H-fluorene (Compound-**6j**) in CDCl$_3$*



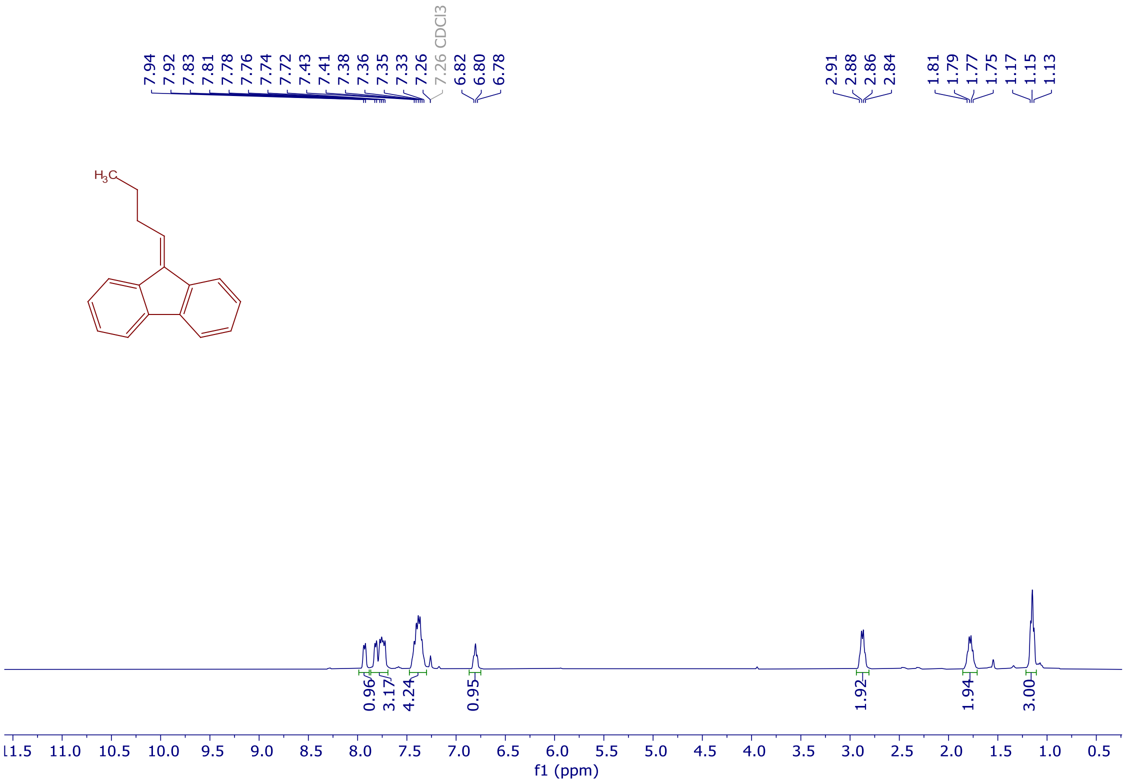
*^{13}C{^{1}H} NMR of 2,7-dibromo-9-(4-methylbenzylidene)-9H-fluorene (Compound-**6j**) in CDCl$_3$*



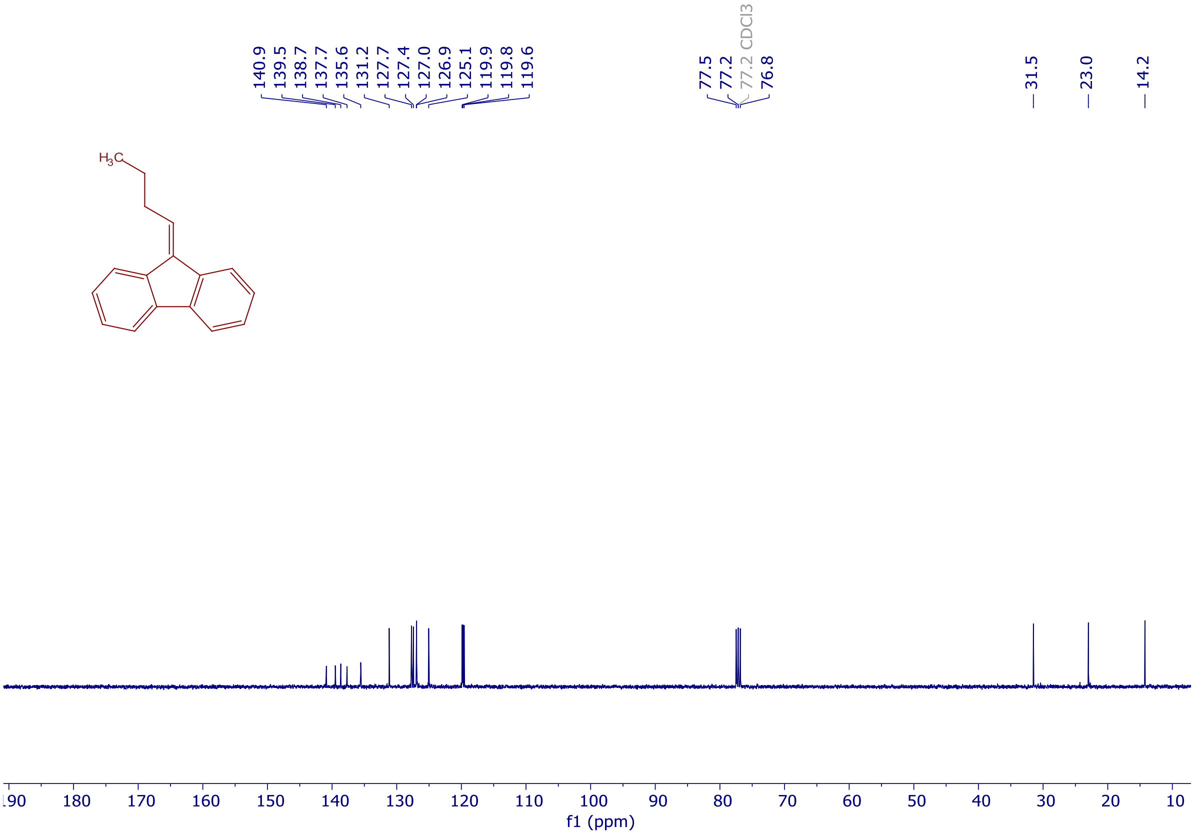
*^{1}H NMR of 2-bromo-7-iodo-9-(4-methylbenzylidene)-9H-fluorene (Compound-**6k**) in CDCl$_3$*



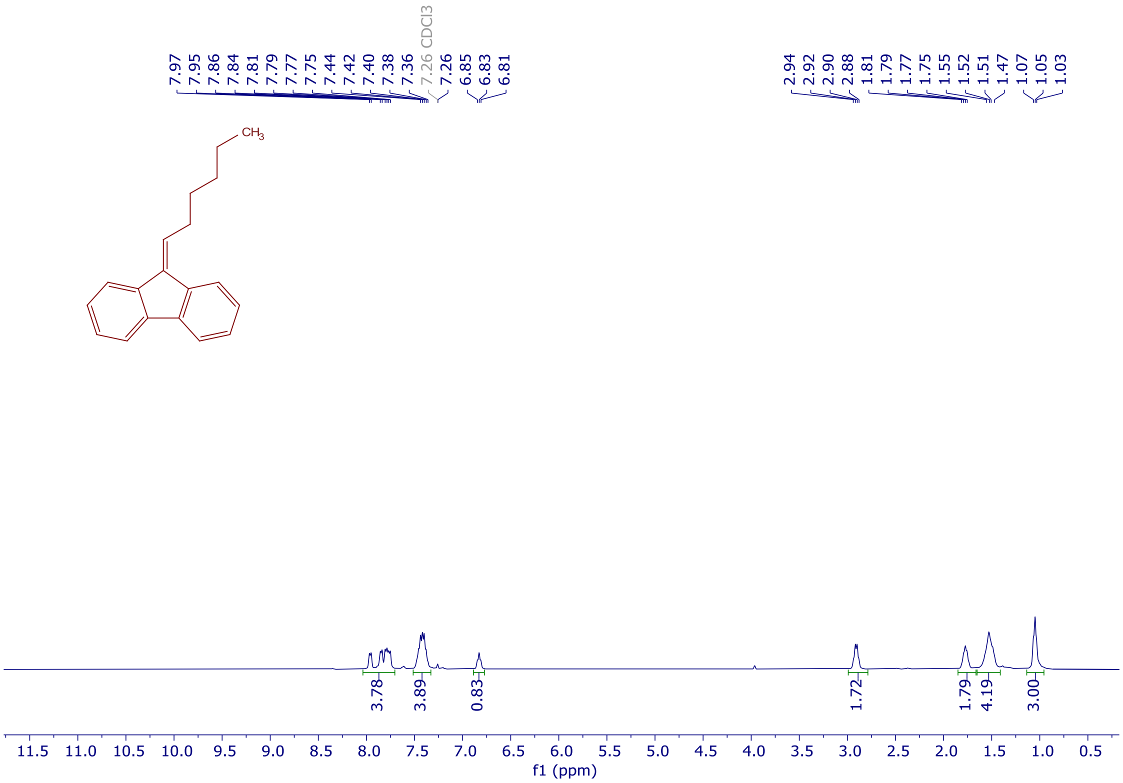
*^{13}C{^{1}H} NMR of 2-bromo-7-iodo-9-(4-methylbenzylidene)-9H-fluorene (Compound-**6k**) in CDCl$_3$*



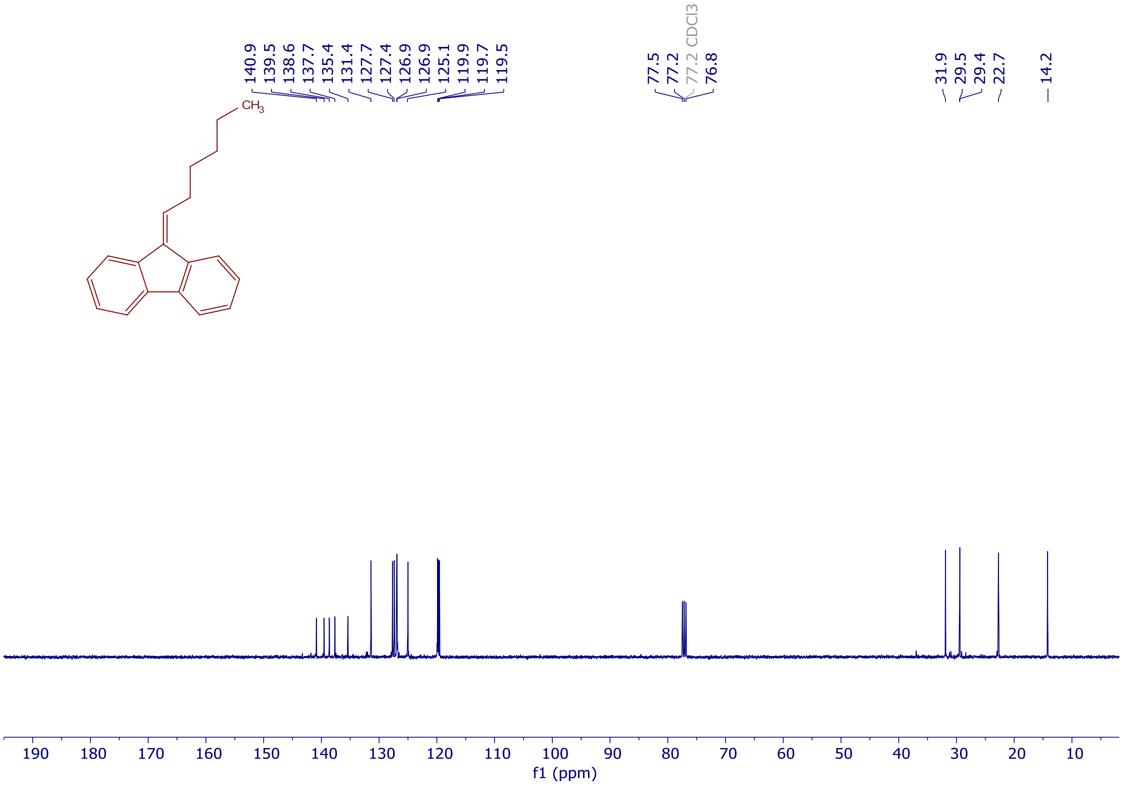
*^{1}H NMR of 9-butylidene-9H-fluorene (Compound-**6l**) in CDCl$_3$*



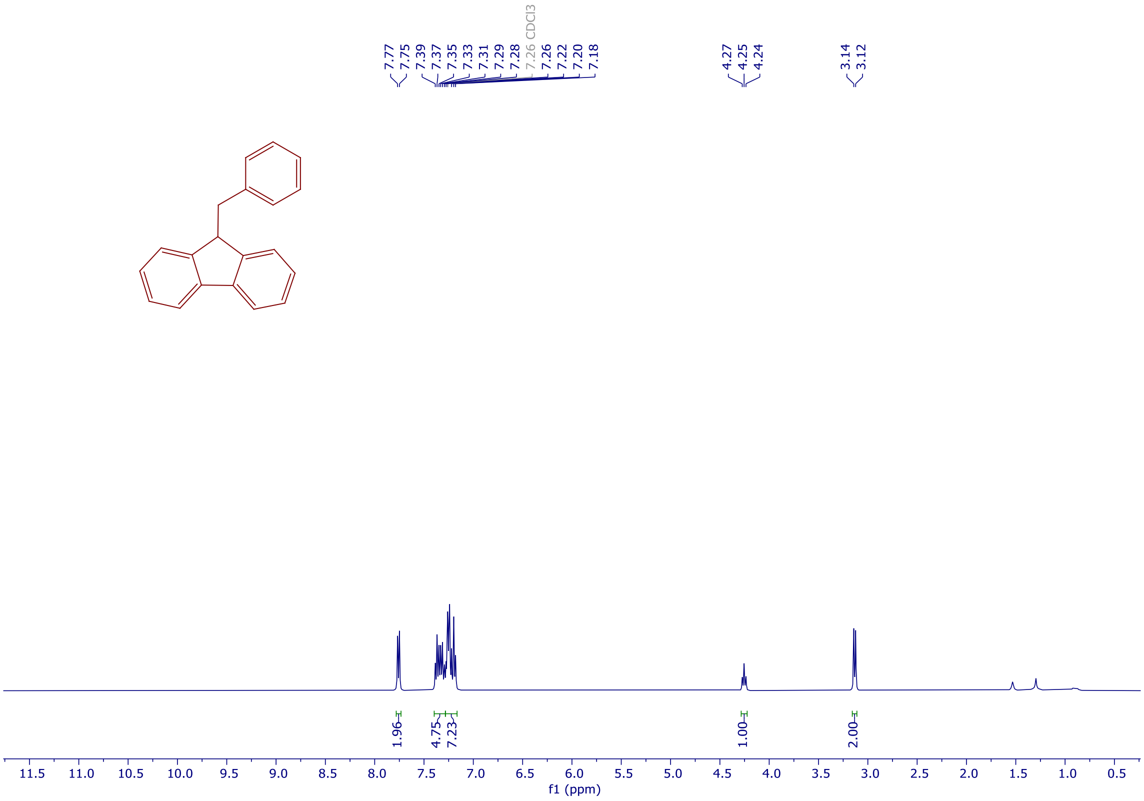
*^{13}C{^{1}H} NMR of 9-butylidene-9H-fluorene (Compound-**6l**) in CDCl$_3$*



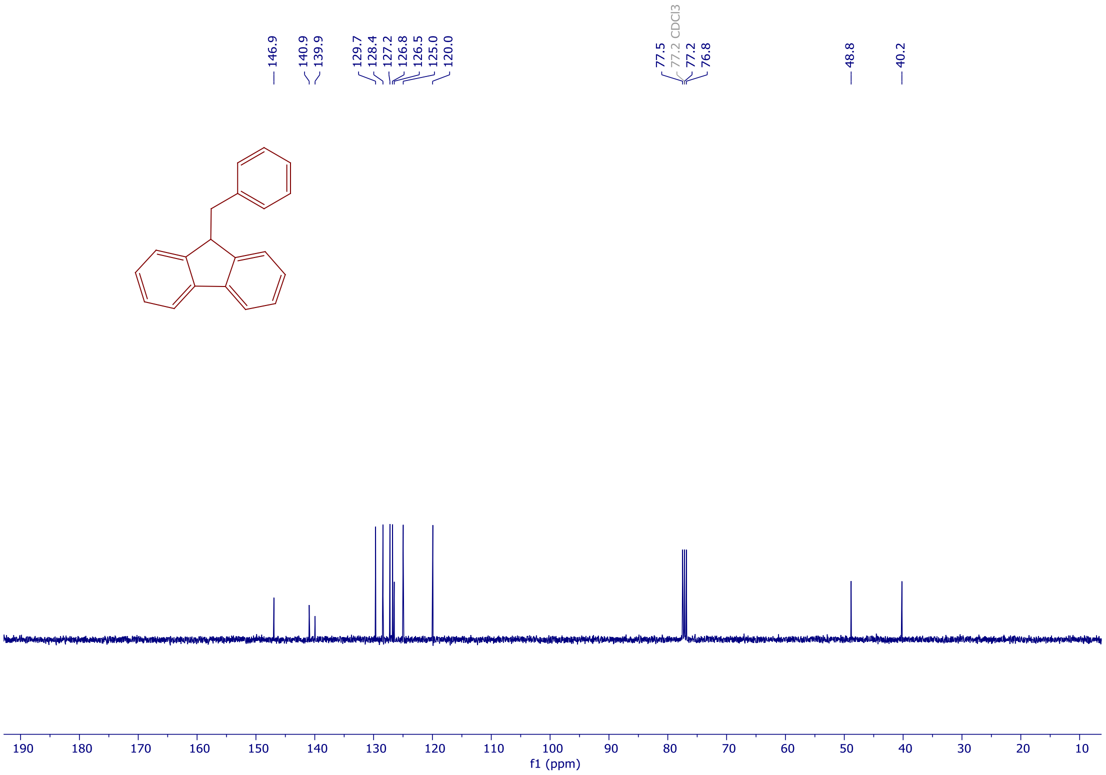
^{1}H NMR of hexylidene-9H-fluorene (Compound-**6m**) in $CDCl_3$



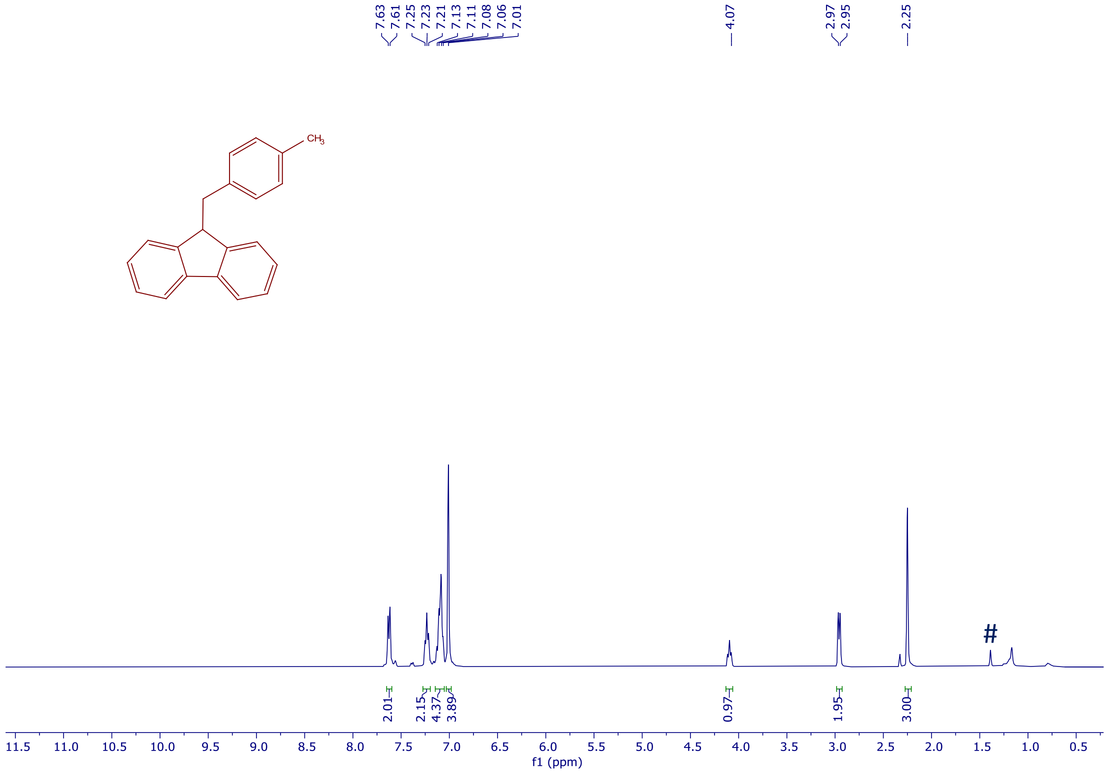
$^{13}C\{^1H\}$ NMR of hexylidene-9H-fluorene (Compound-**6m**) in $CDCl_3$



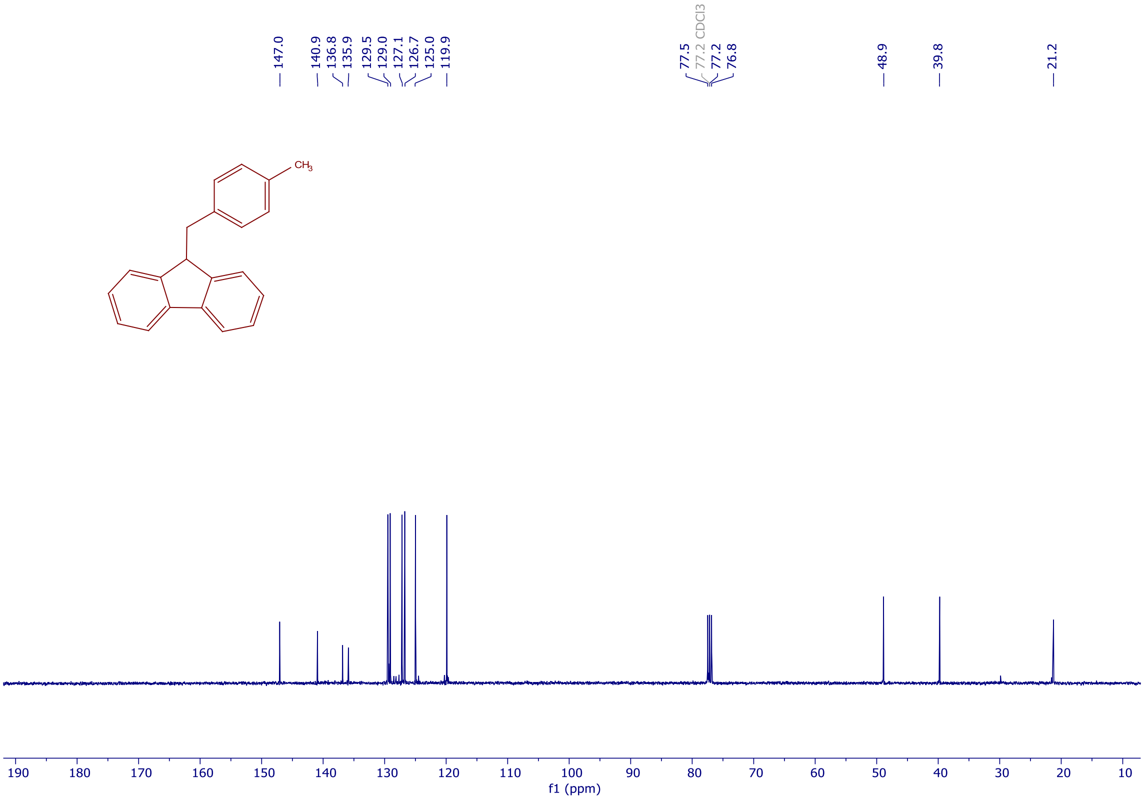
^{1}H NMR of 9-benzyl-9H-fluorene (Compound-**7a**) in CDCl$_3$



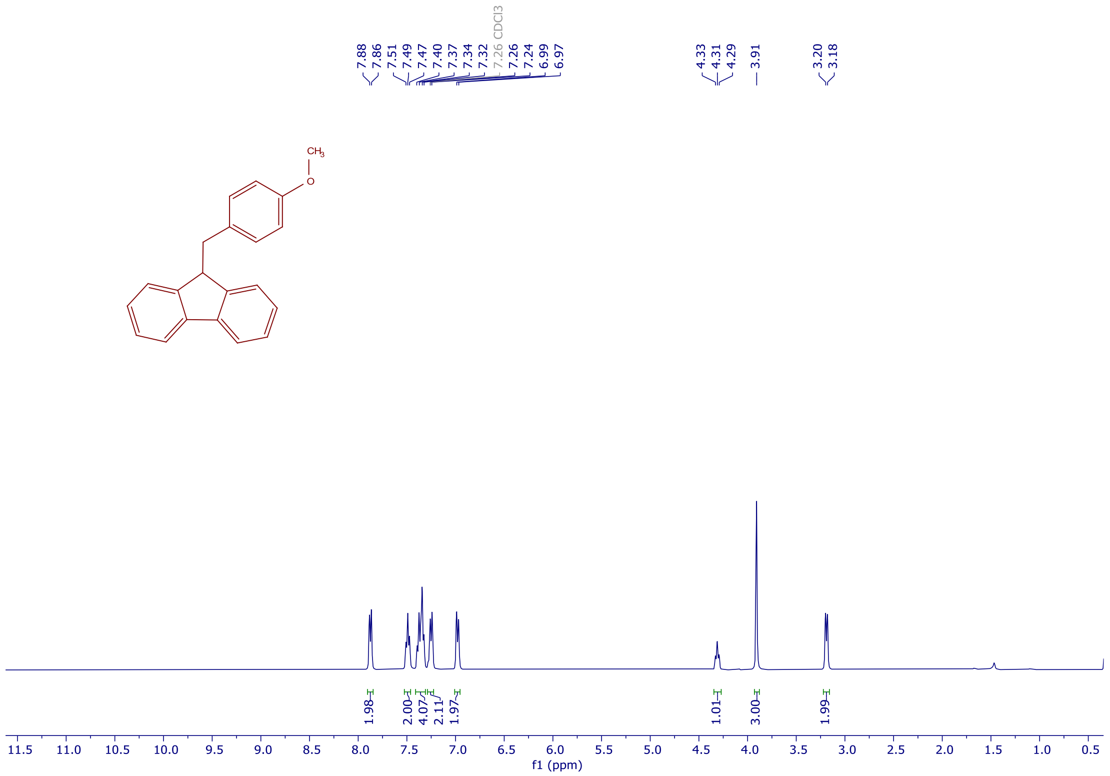
^{13}C{^{1}H} NMR of 9-benzyl-9H-fluorene (Compound-**7a**) in CDCl$_3$



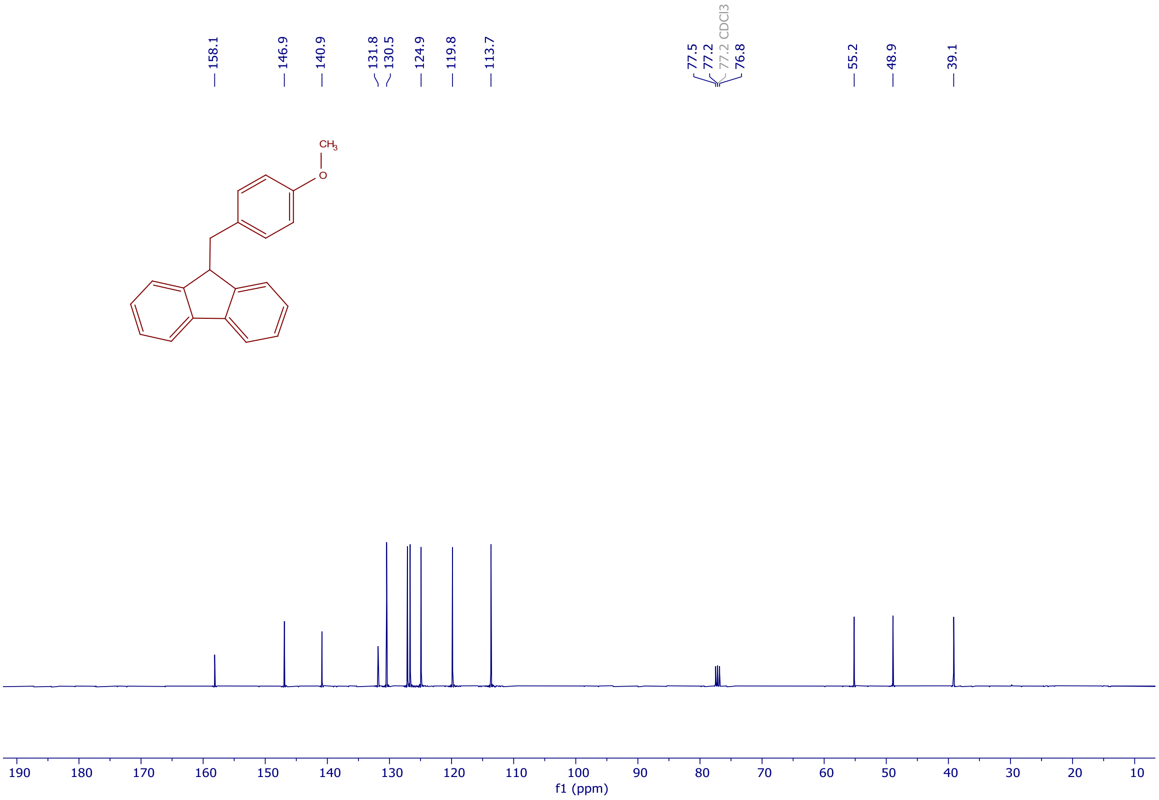
*^{1}H NMR of 9-(4-methylbenzyl)-9H-fluorene (Compound-**7b**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



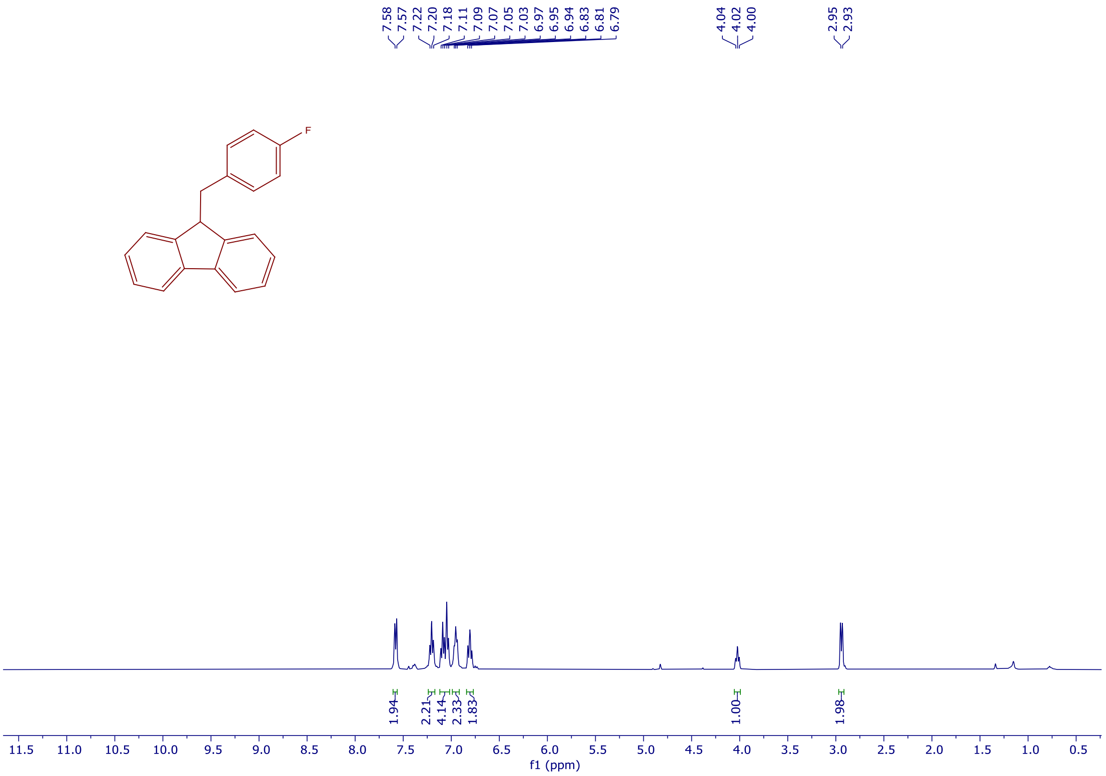
*$^{13}C\{^1H\}$ NMR of 9-(4-methylbenzyl)-9H-fluorene (Compound-**7b**) in $CDCl_3$*



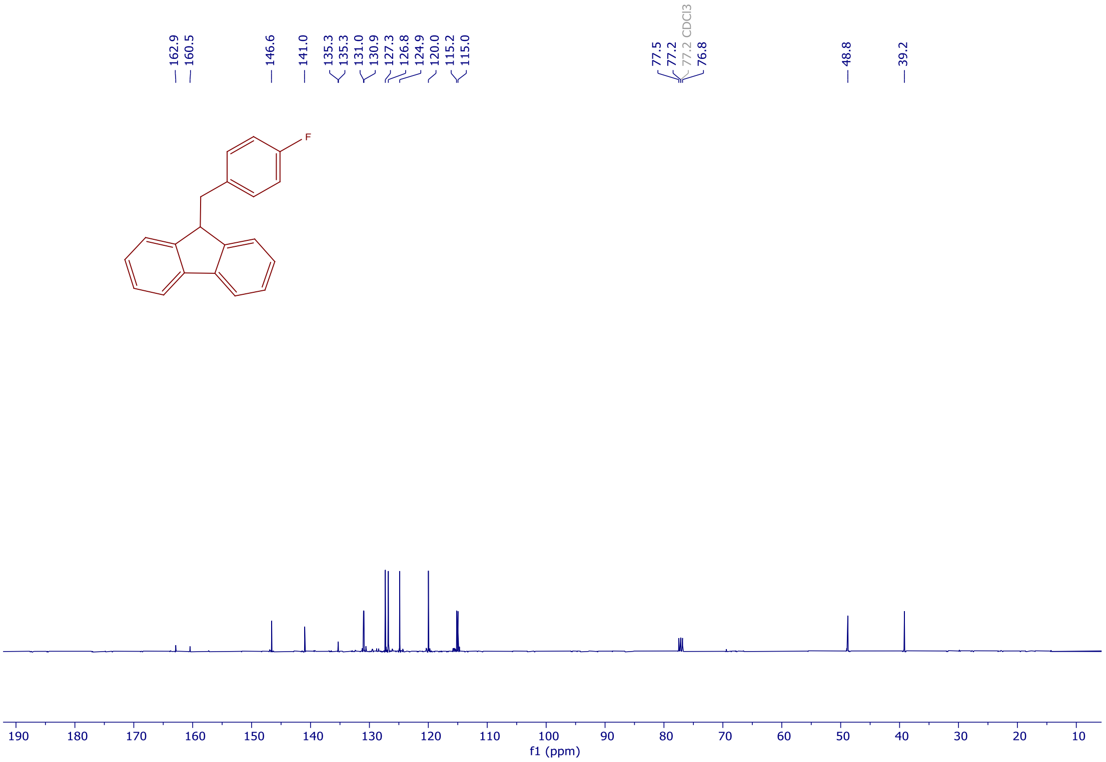
^{1}*H NMR of 9-(4-methoxybenzyl)-9H-fluorene (Compound-**7c**) in CDCl*$_{3}$



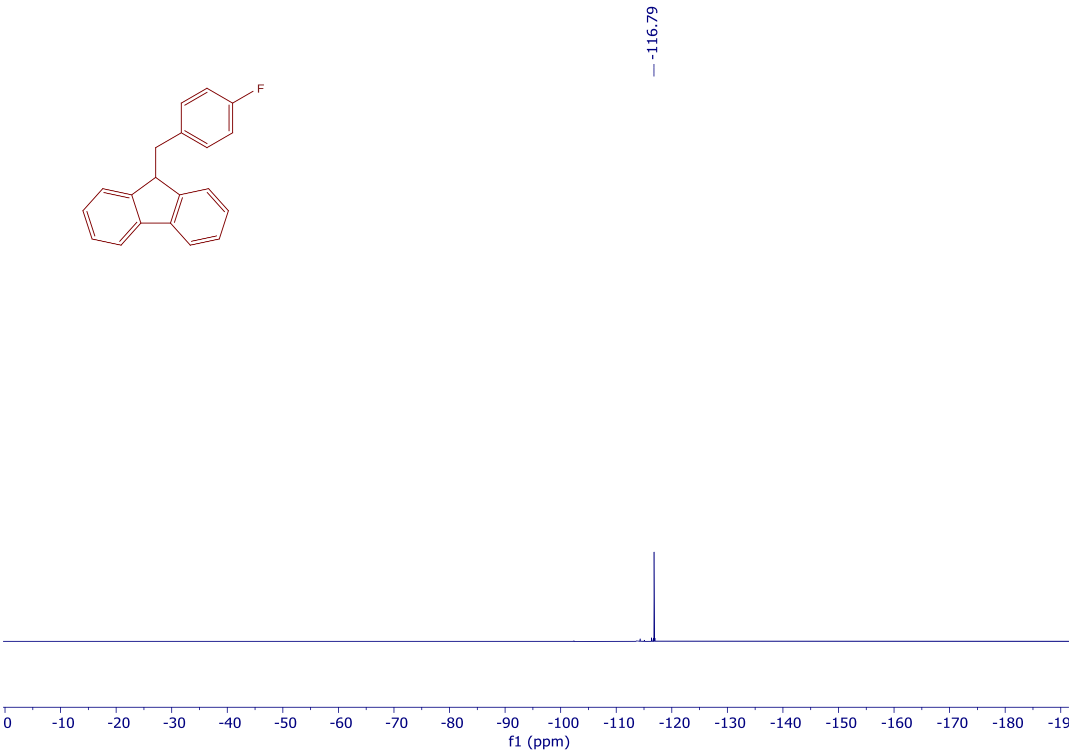
^{13}*C{*^{1}*H} NMR of 9-(4-methylbenzyl)-9H-fluorene(Compound-**7c**) in CDCl*$_{3}$



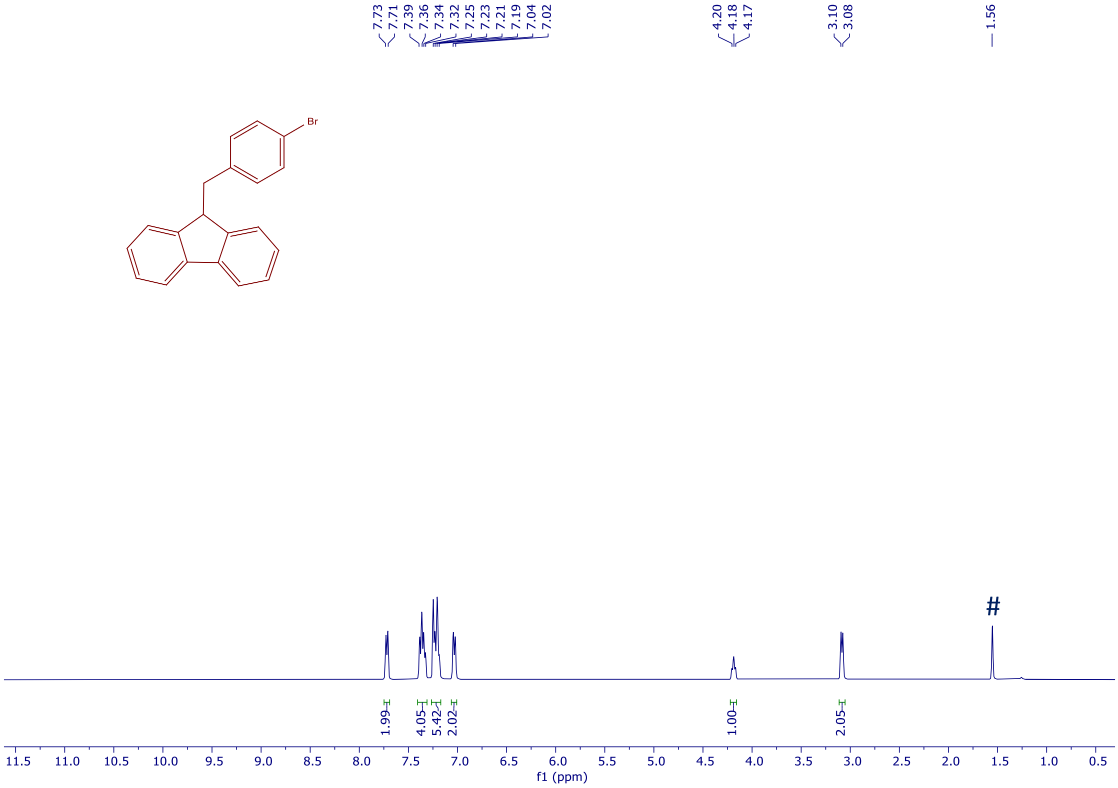
*^{1}H NMR of 9-(4-fluorobenzyl)-9H-fluorene (Compound-**7d**) in CDCl$_3$*



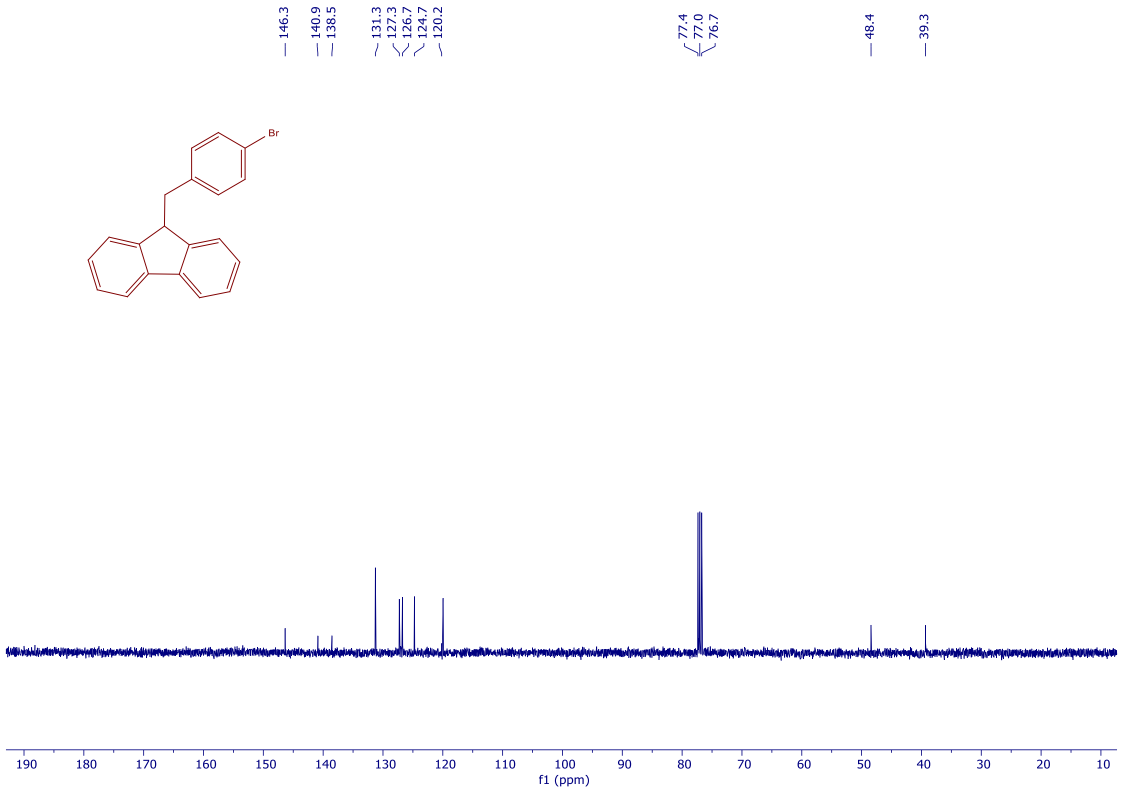
*^{13}C{^{1}H} NMR of 9-(4-fluorobenzyl)-9H-fluorene (Compound-**7d**) in CDCl$_3$*



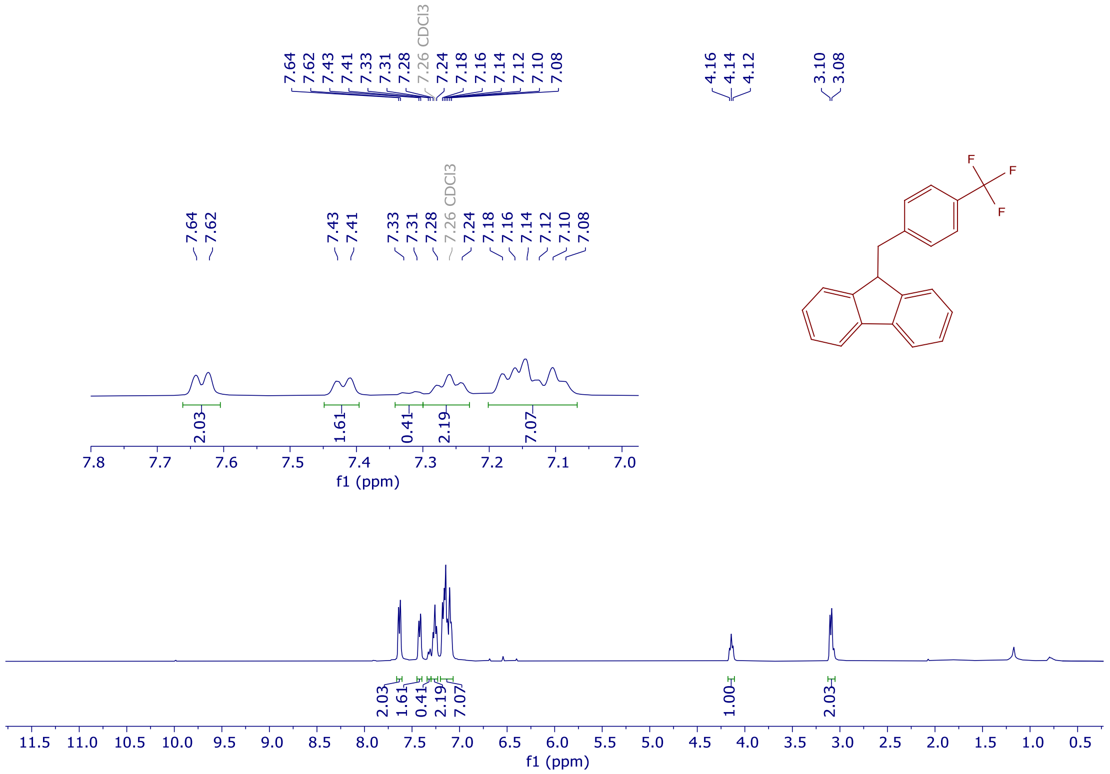
^{19}*F NMR of 9-(4-fluorobenzyl)-9H-fluorene (Compound-****7d****) in* $CDCl_3$



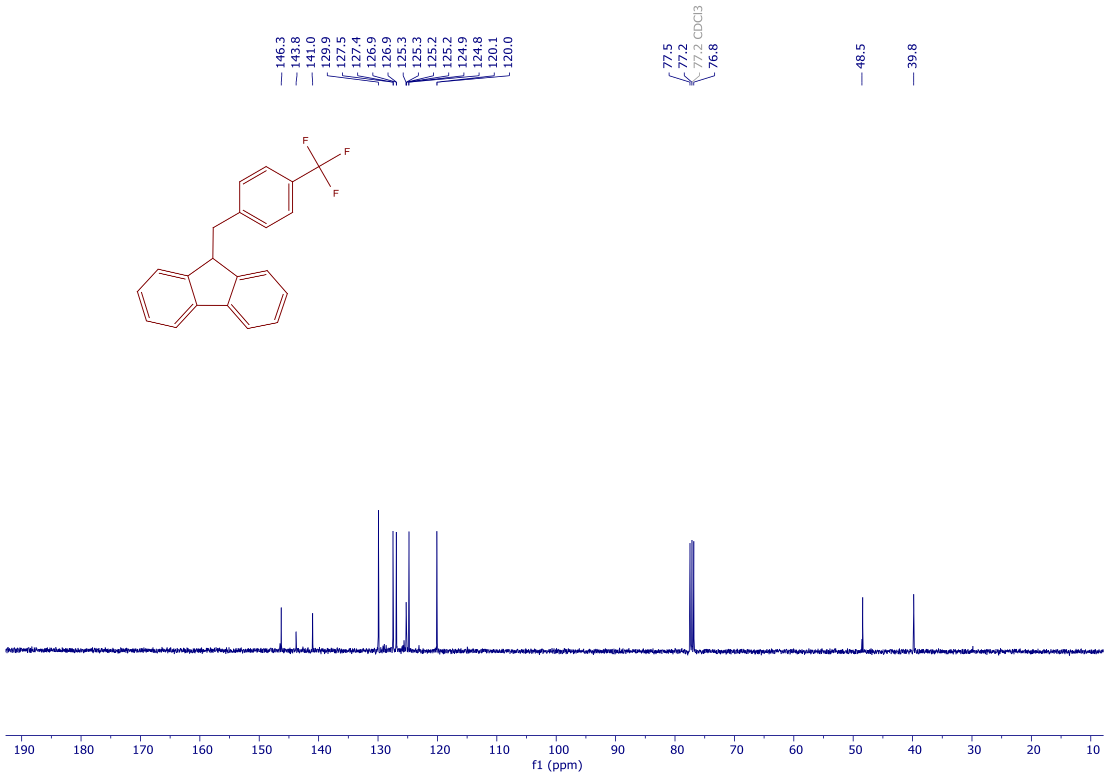
^{1}*H NMR of 9-(4-bromobenzyl)-9H-fluorene (Compound-****7e****) in* $CDCl_3$*. # indicates the solvent impurity of* H_2O *in* $CDCl_3$



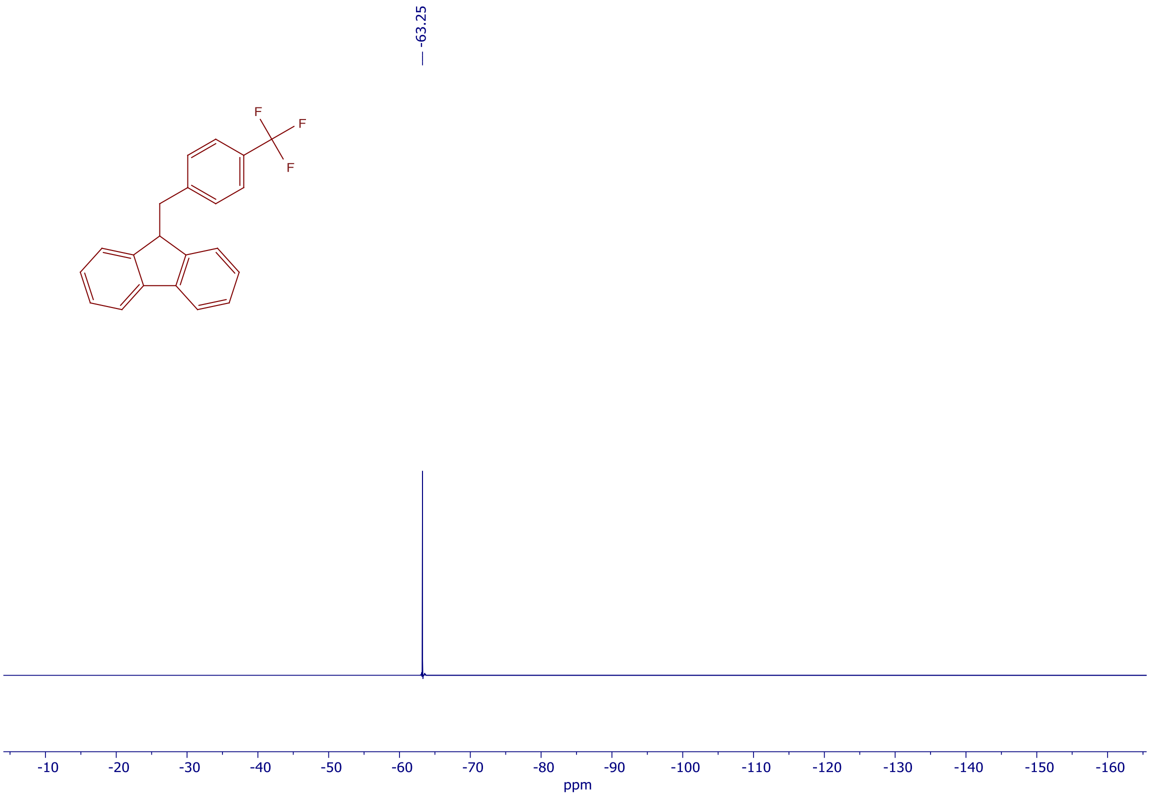
^{13}C{^{1}H} *NMR of 9-(4-bromobenzyl)-9H-fluorene (Compound-**7e**) in CDCl$_3$*



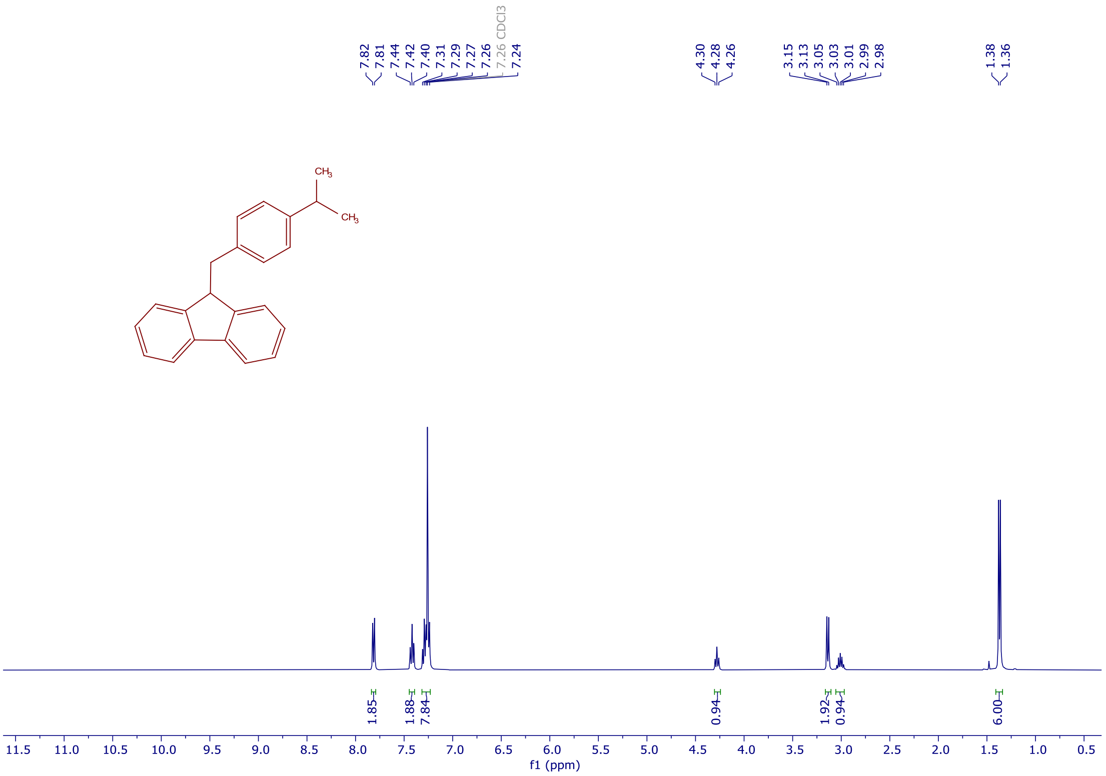
^{1}H *NMR of 9-(4-(trifluoromethyl)benzyl)-9H-fluorene (Compound-**7f**) in CDCl$_3$*



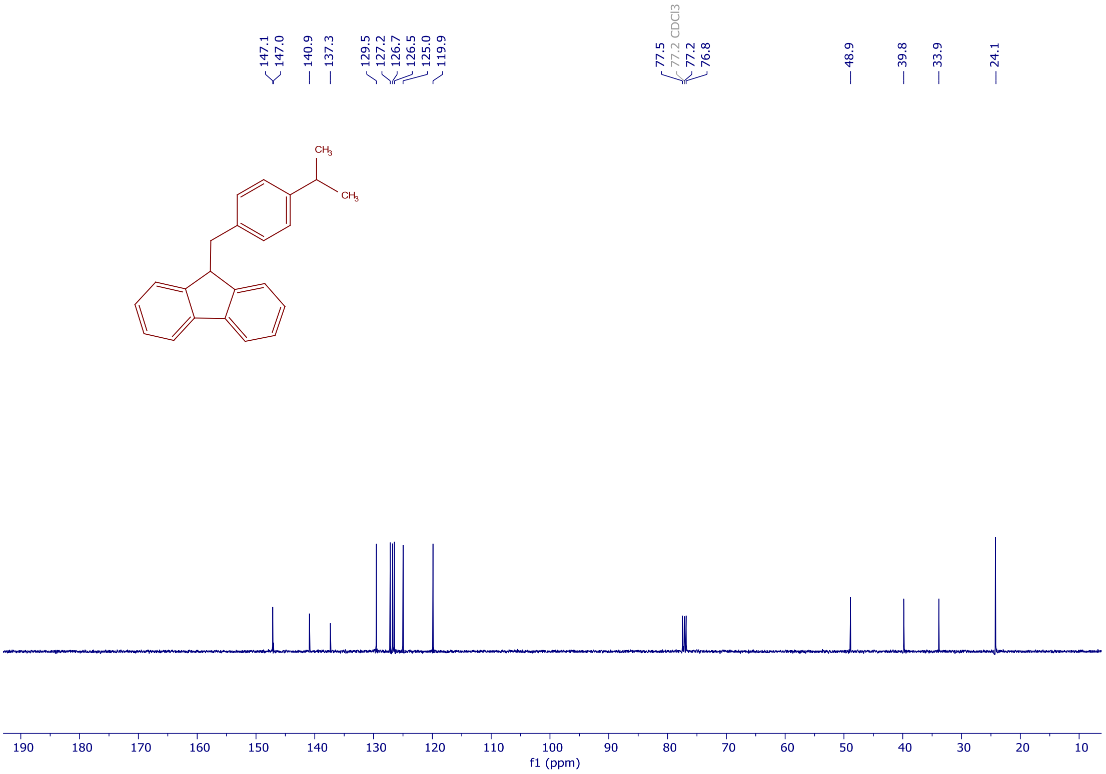
*$^{13}C\{^{1}H\}$ NMR of 9-(4-(trifluoromethyl)benzyl)-9H-fluorene (Compound-**7f**) in $CDCl_3$*



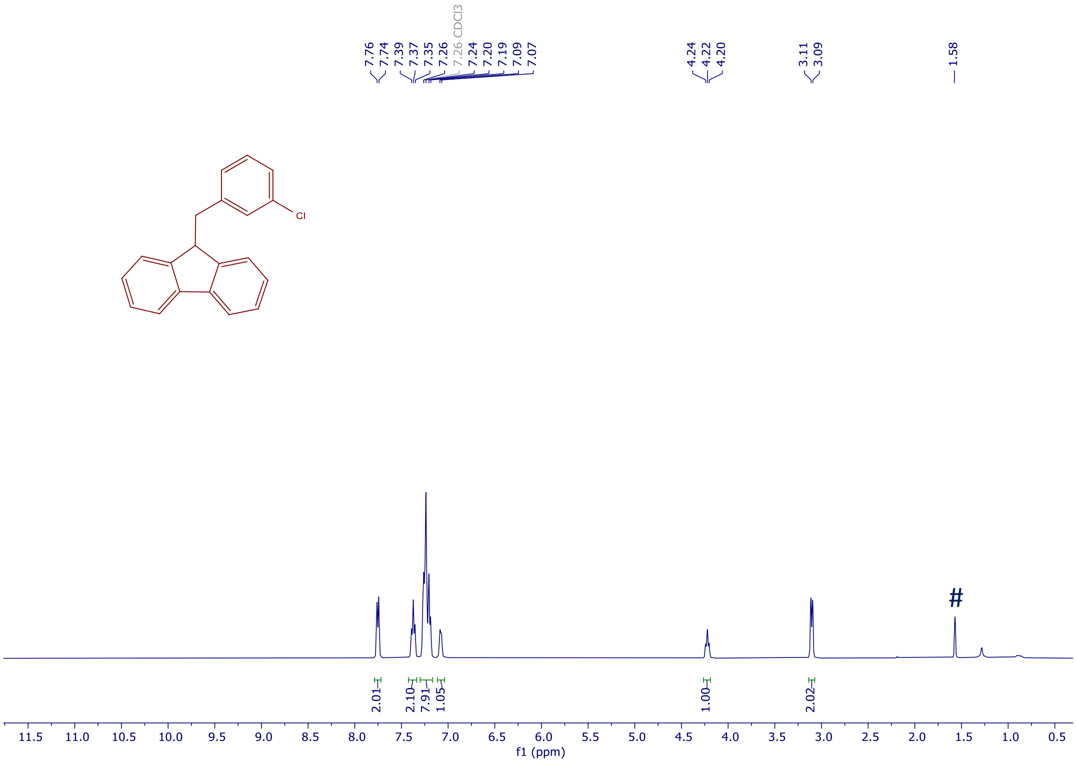
*^{19}F NMR of 9-(4-(trifluoromethyl)benzyl)-9H-fluorene (Compound-**7f**) in $CDCl_3$*



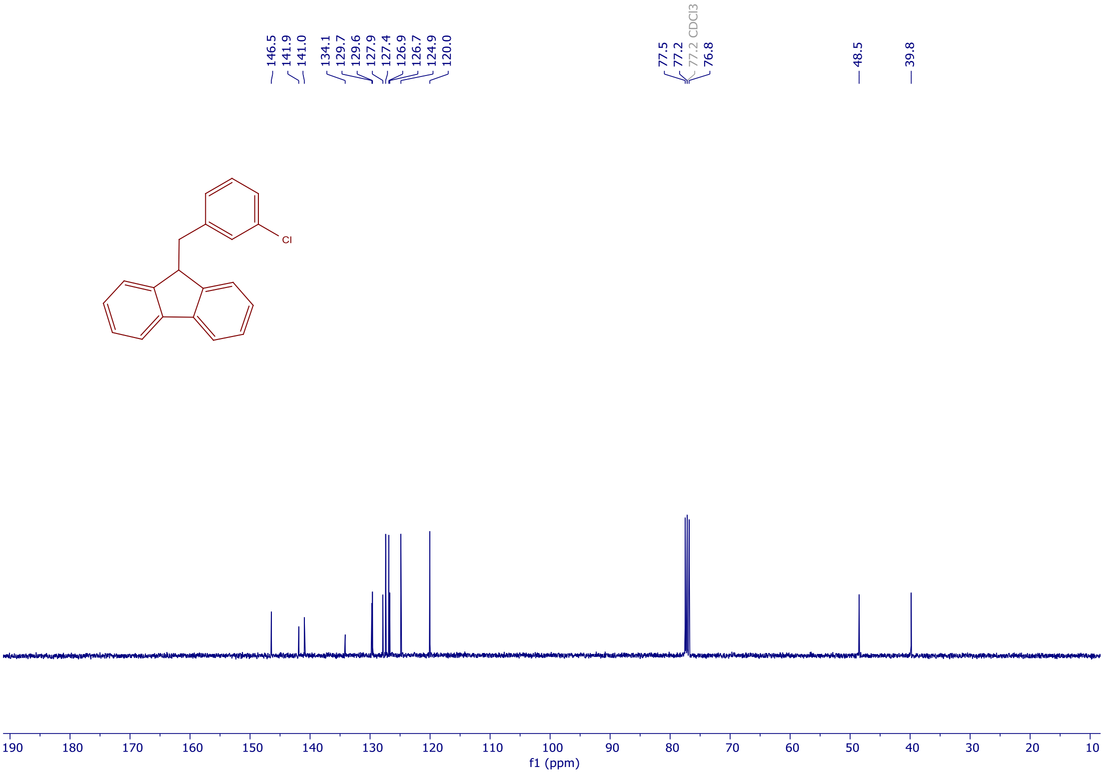
*^{1}H NMR of 9-(4-isopropylbenzyl)-9H-fluorene (Compound-**7g**) in CDCl$_3$*



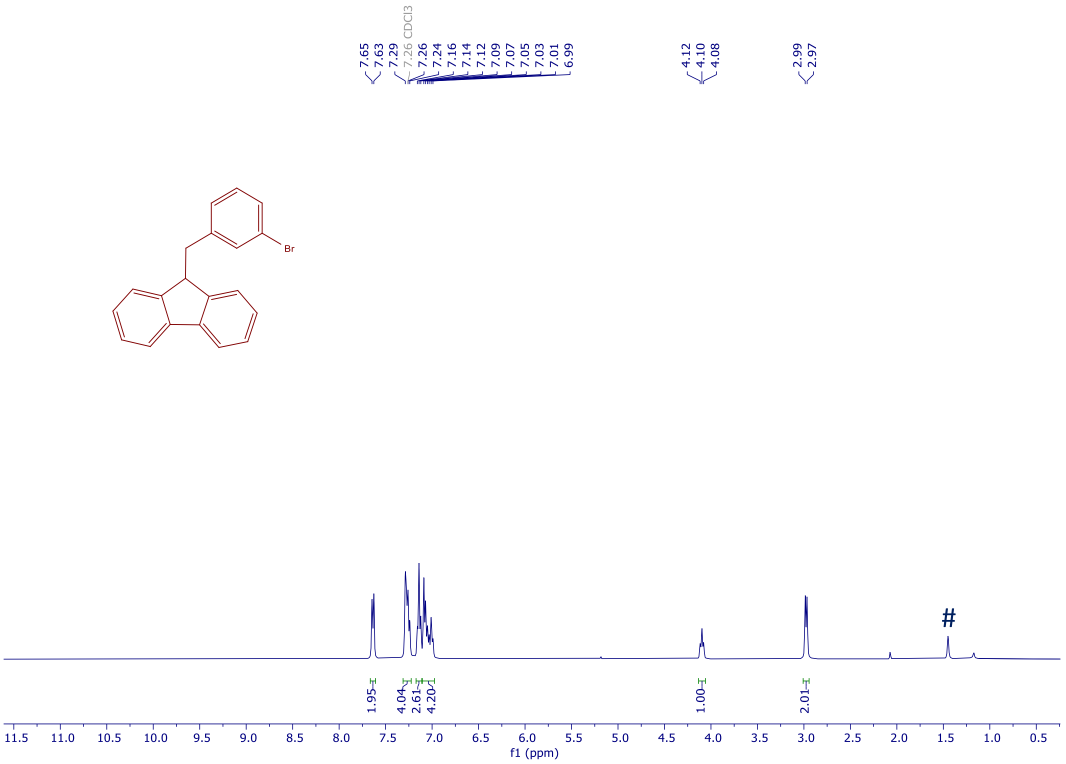
*^{13}C{^{1}H} NMR of 9-(4-isopropylbenzyl)-9H-fluorene (Compound-**7g**) in CDCl$_3$*



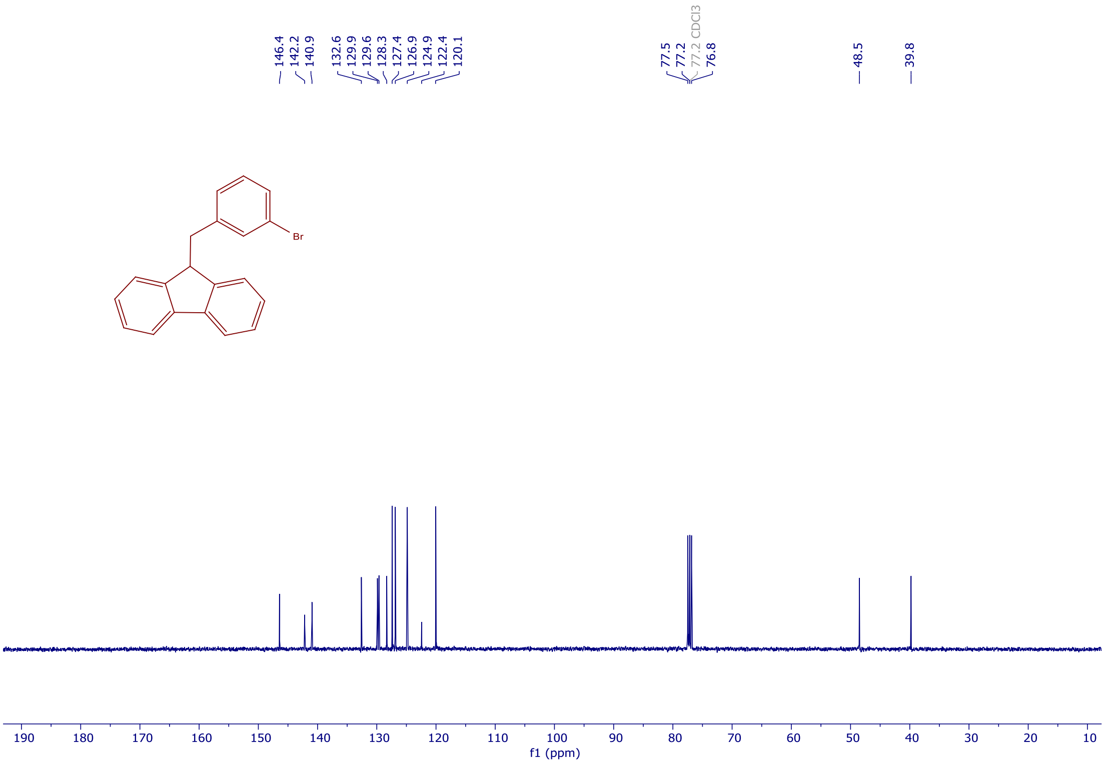
*^{1}H NMR of 9-(3-chlorobenzyl)-9H-fluorene(Compound-**7h**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



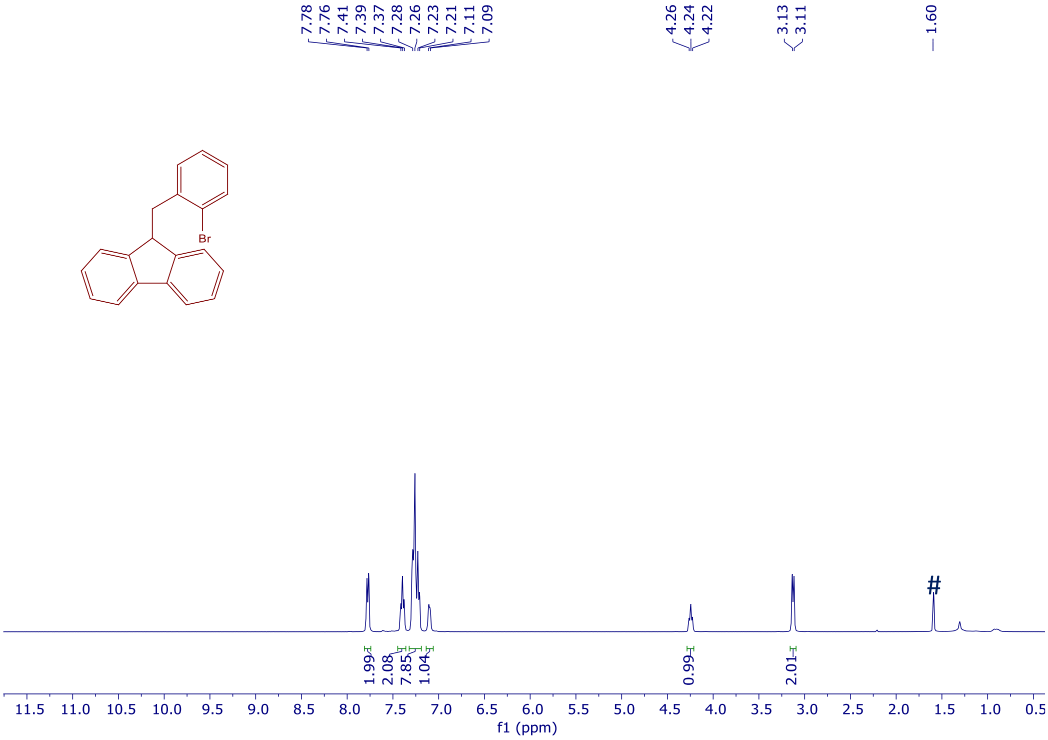
*$^{13}C\{^1H\}$ NMR of 9-(3-chlorobenzyl)-9H-fluorene(Compound-**7h**) in $CDCl_3$*



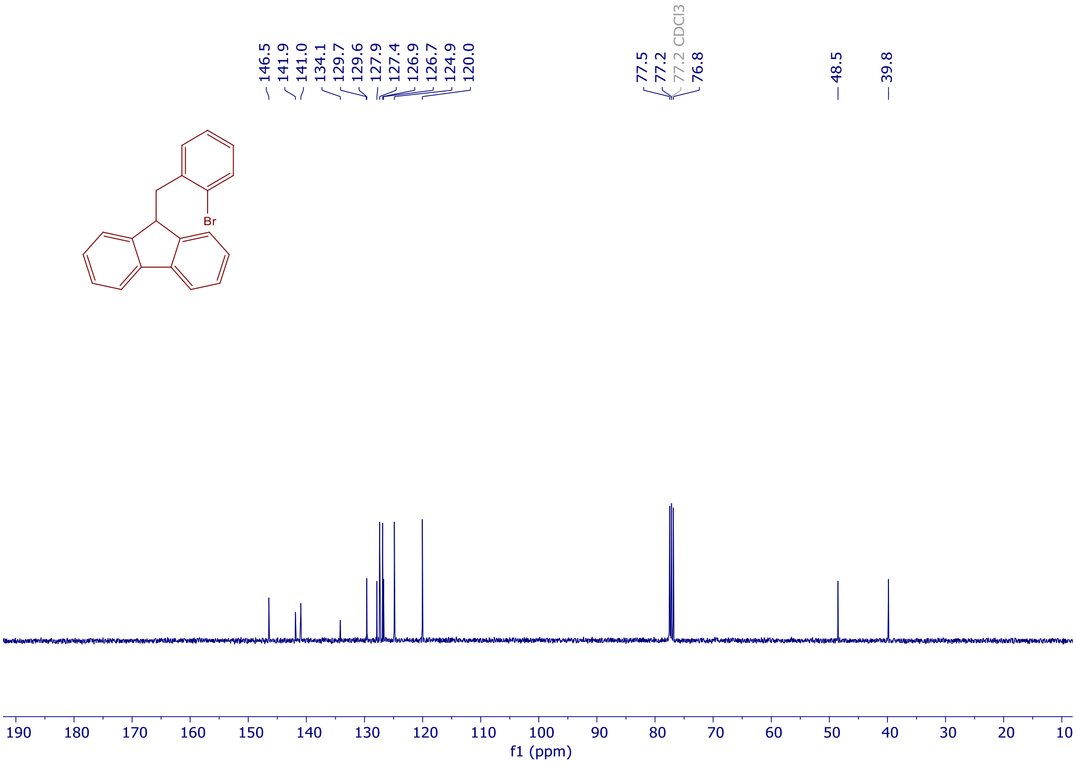
*^{1}H NMR of 9-(3-bromobenzyl)-9H-fluorene(Compound-**7i**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



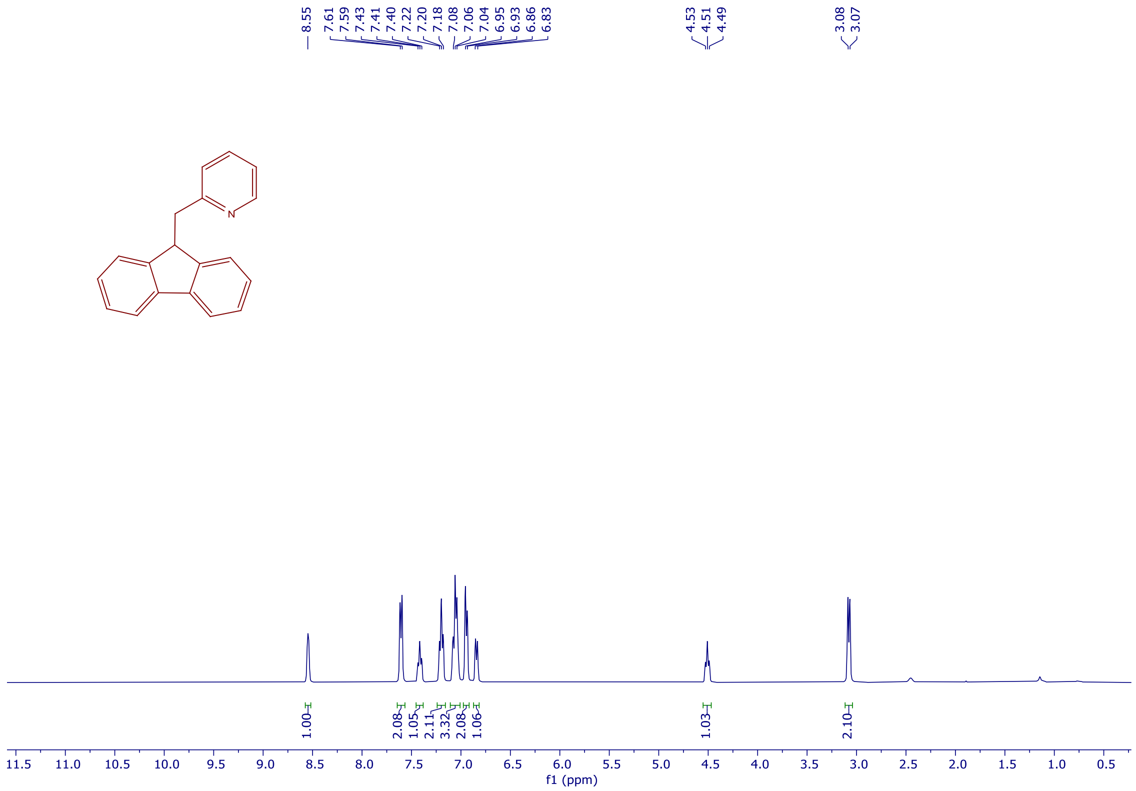
*$^{13}C\{^1H\}$ NMR of 9-(3-bromobenzyl)-9H-fluorene(Compound-**7i**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



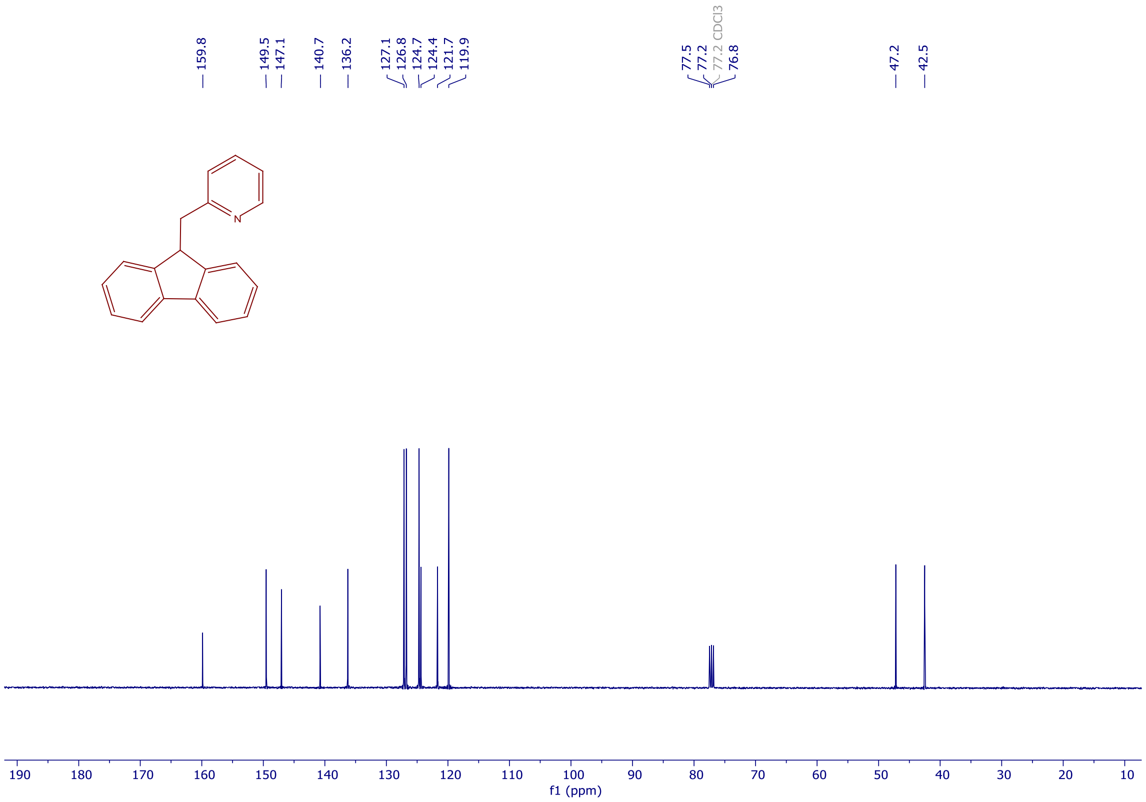
*^{1}H NMR of 9-(2-bromoobenzyl)-9H-fluorene (Compound-**7j**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



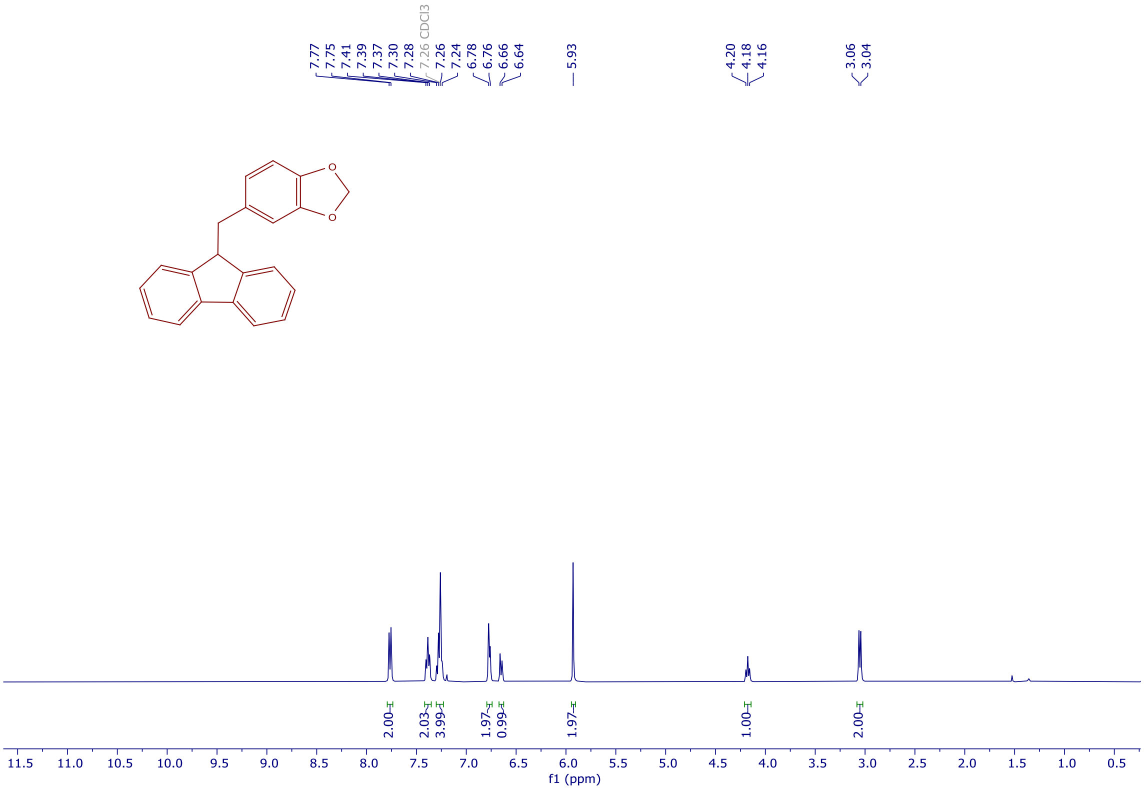
*$^{13}C\{^{1}H\}$ NMR of 9-(2-bromobenzyl)-9H-fluorene (Compound-**7j**) in $CDCl_3$*



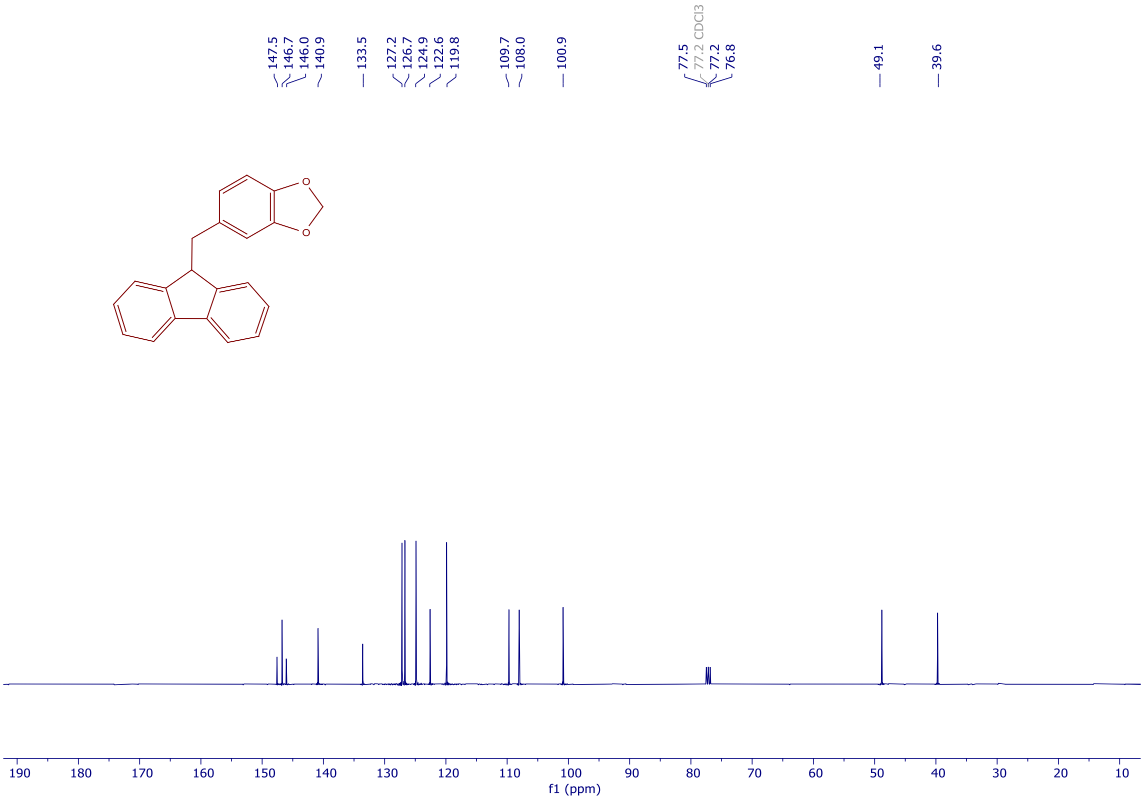
^{1}*H NMR of 2-((9H-fluoren-9-yl)methyl)pyridine (Compound-**7k**) in* $CDCl_3$



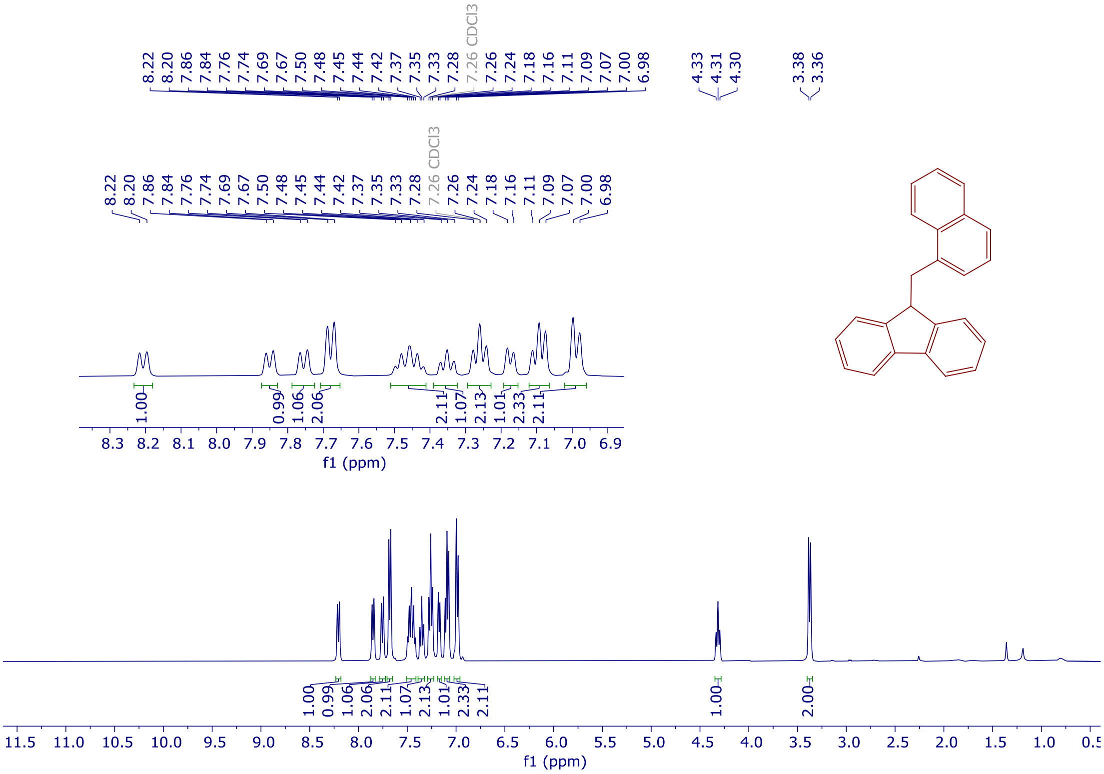
$^{13}C\{^{1}H\}$ *NMR of 2-((9H-fluoren-9-yl)methyl)pyridine (Compound-**7k**) in* $CDCl_3$



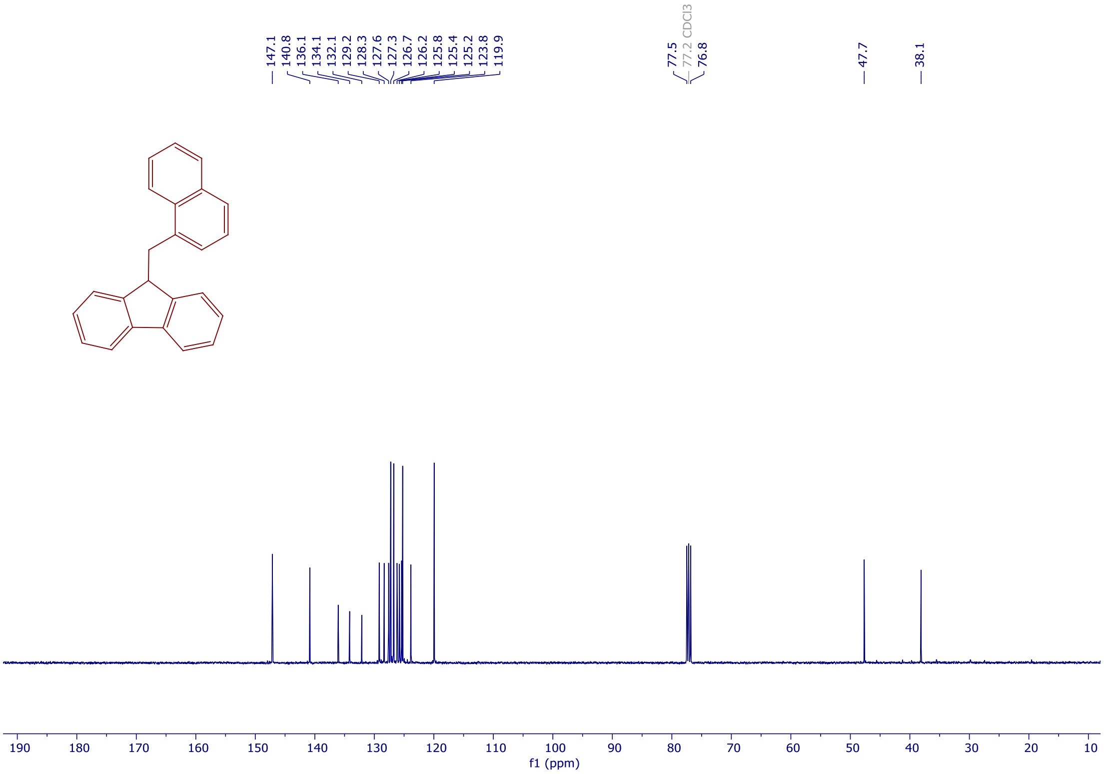
*^{1}H NMR of 5-((9H-fluoren-9-yl)methyl)benzo[d][1,3]dioxole (Compound-**7l**) in $CDCl_3$*



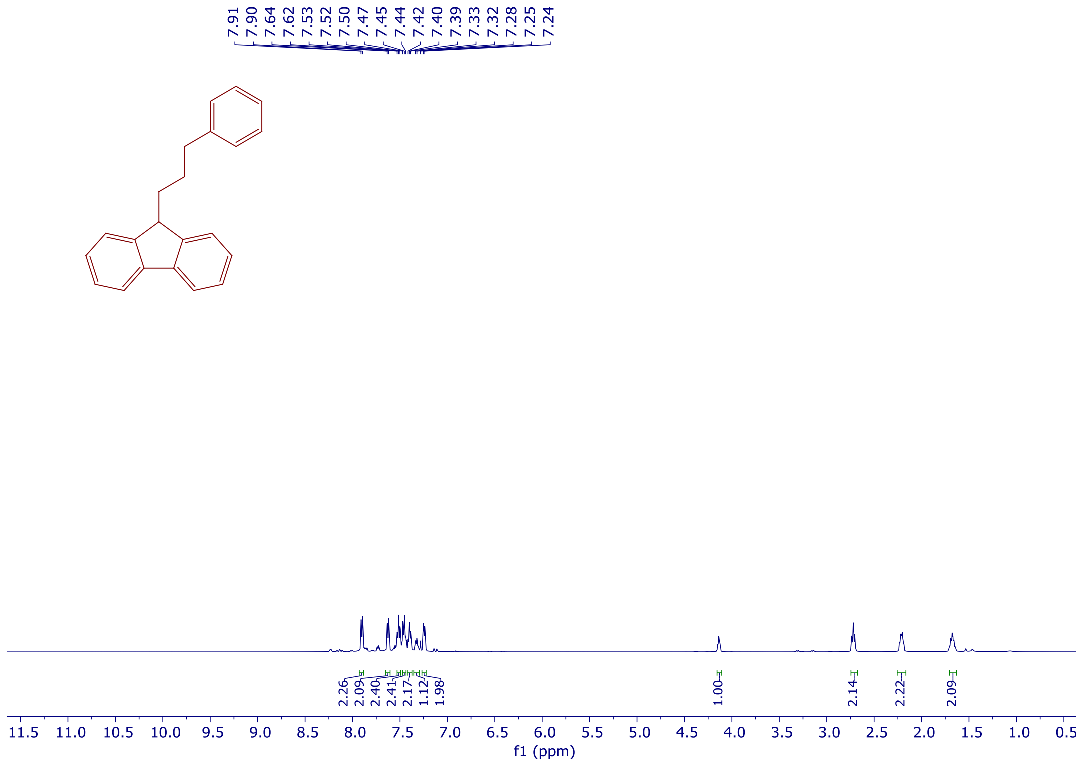
*^{13}C{^{1}H} NMR of 5-((9H-fluoren-9-yl)methyl)benzo[d][1,3]dioxole (Compound-**7l**) in $CDCl_3$*



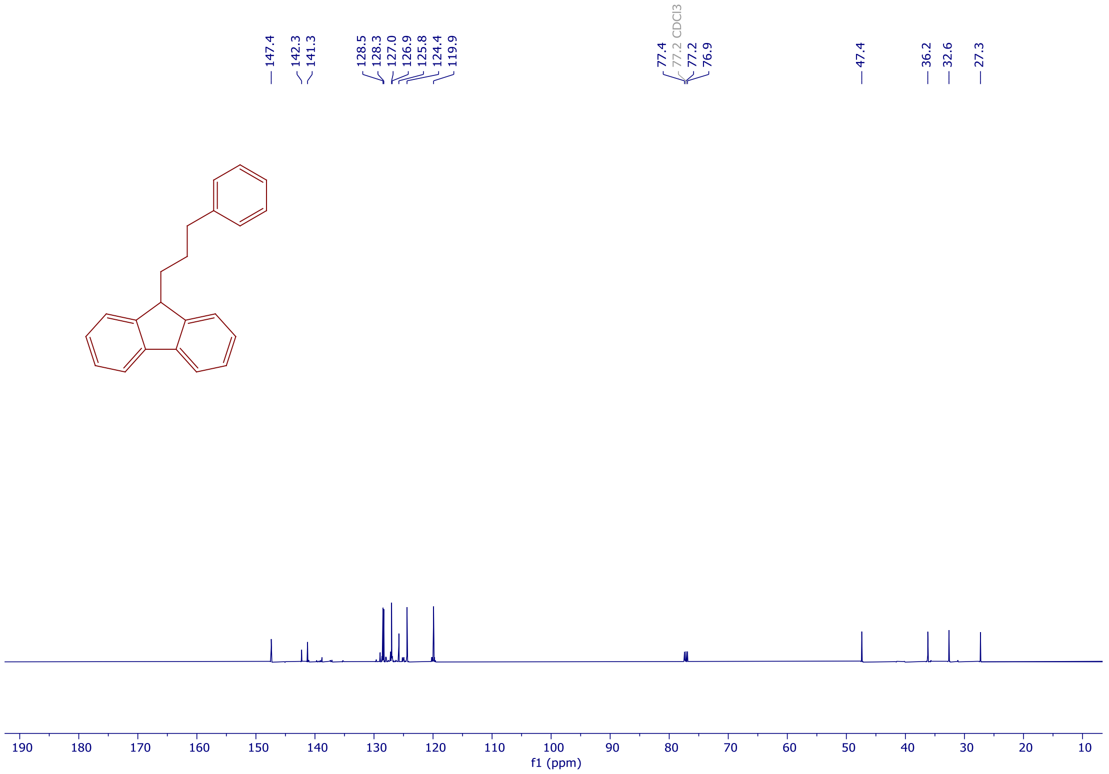
^{1}*H NMR of 9-(naphthalen-1-ylmethyl)-9H-fluorene (Compound-**7m**) in CDCl*$_3$



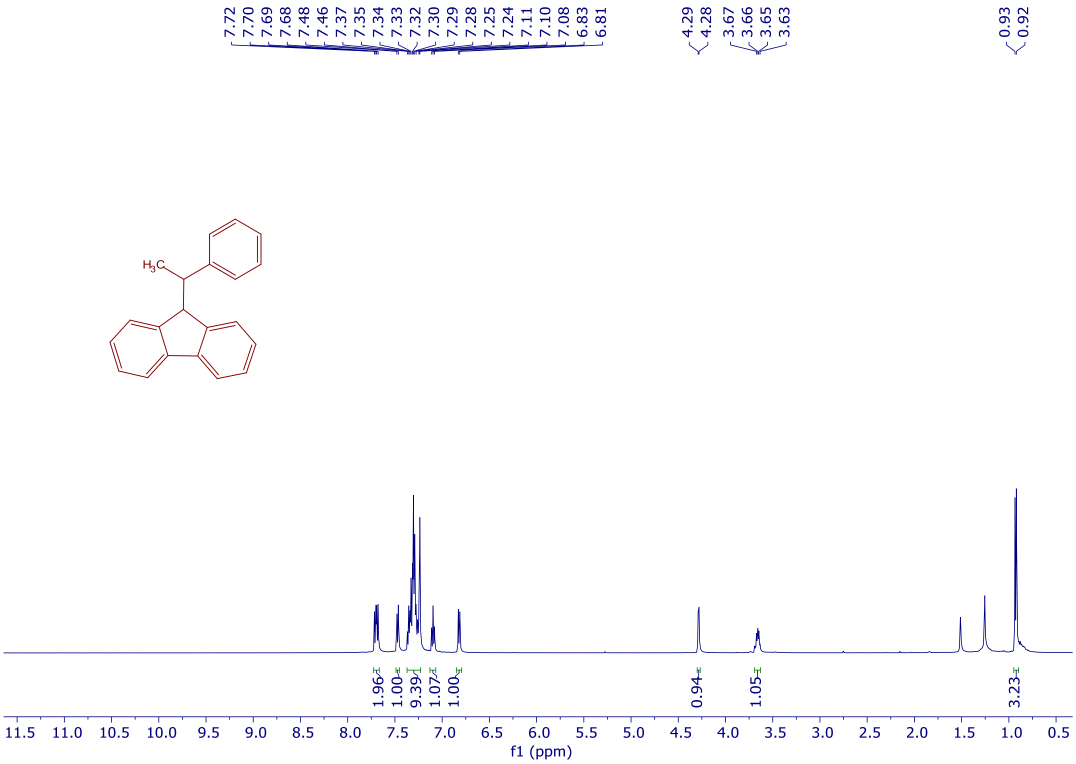
^{13}*C{*^{1}*H} NMR of 9-(naphthalen-1-ylmethyl)-9H-fluorene (Compound-**7m**) in CDCl*$_3$



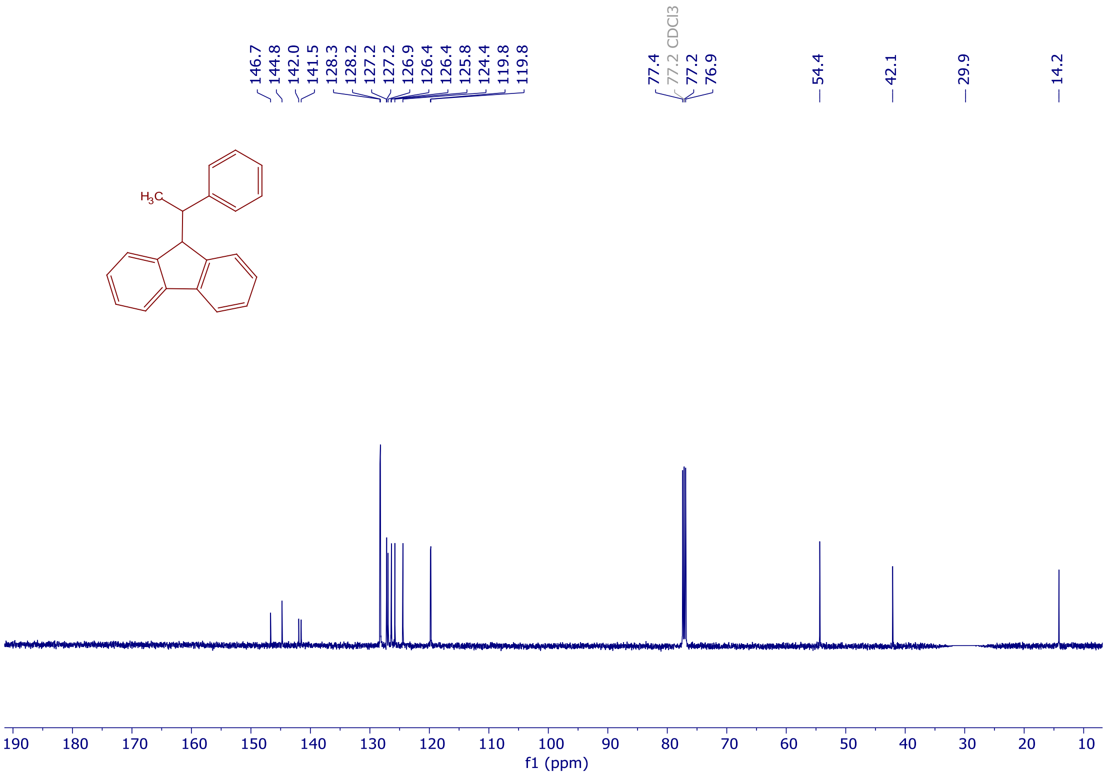
*^{1}H NMR of 9-(3-phenylpropyl)-9H-fluorene (Compound-**7n**) in $CDCl_3$*



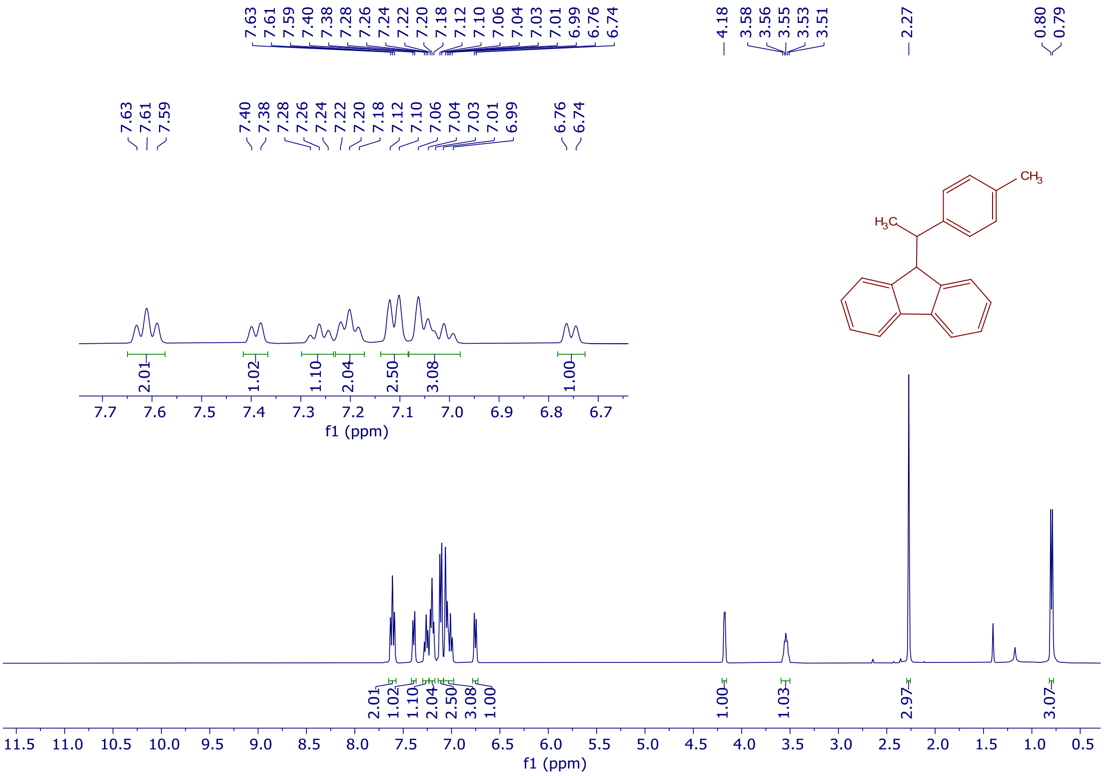
*^{13}C{^{1}H} NMR of 9-(3-phenylpropyl)-9H-fluorene (Compound-**7n**) in $CDCl_3$*



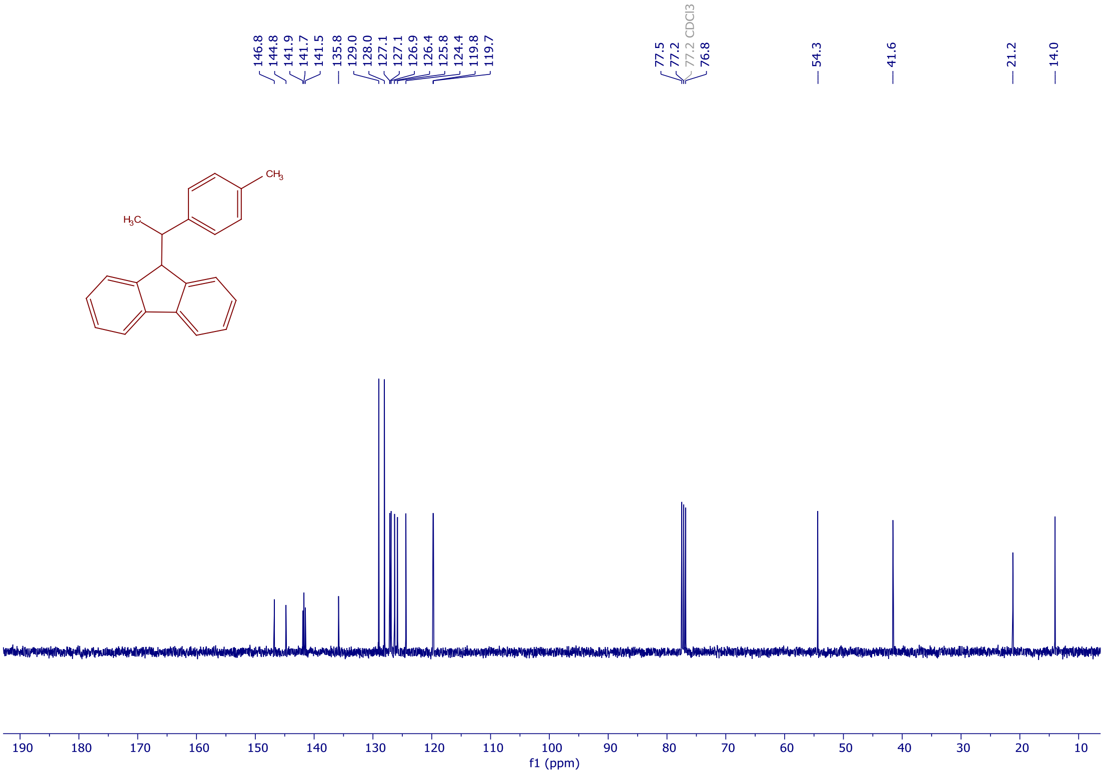
*^{1}H NMR of 9-(1-phenylethyl)-9H-fluorene (Compound-**7o**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



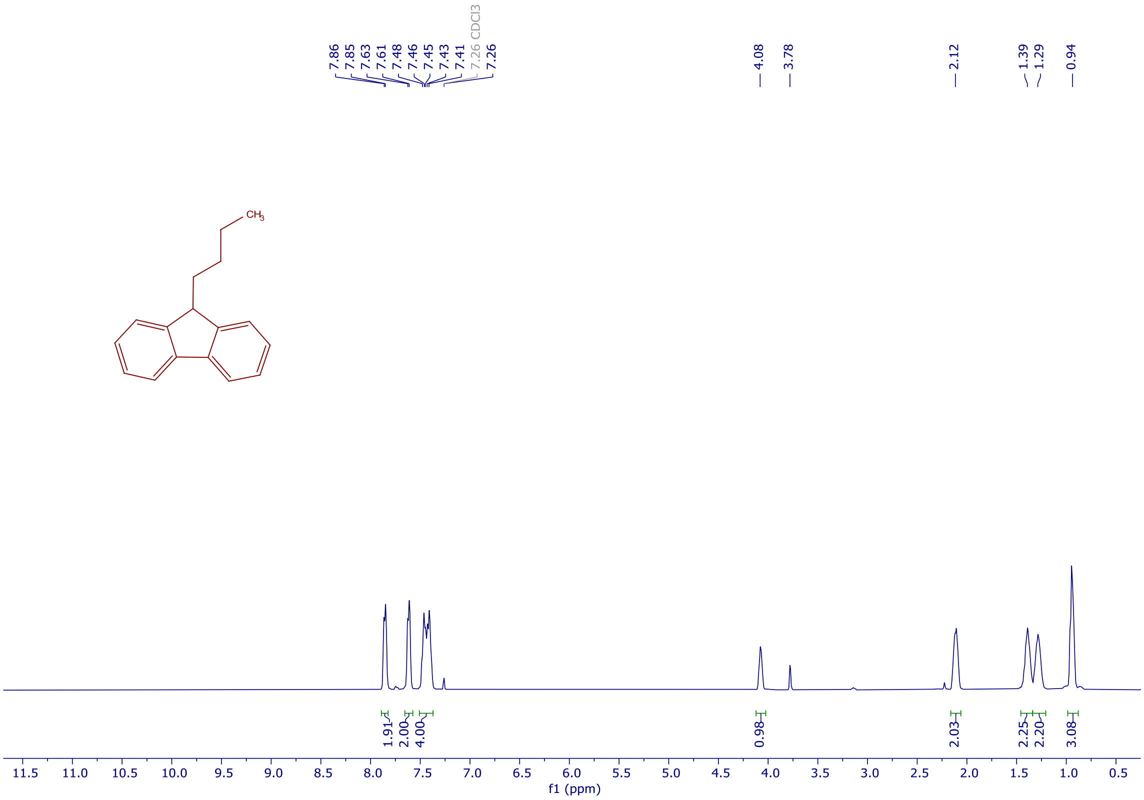
*$^{13}C\{^1H\}$ NMR of 9-(1-phenylethyl)-9H-fluorene (Compound-**7o**) in $CDCl_3$*



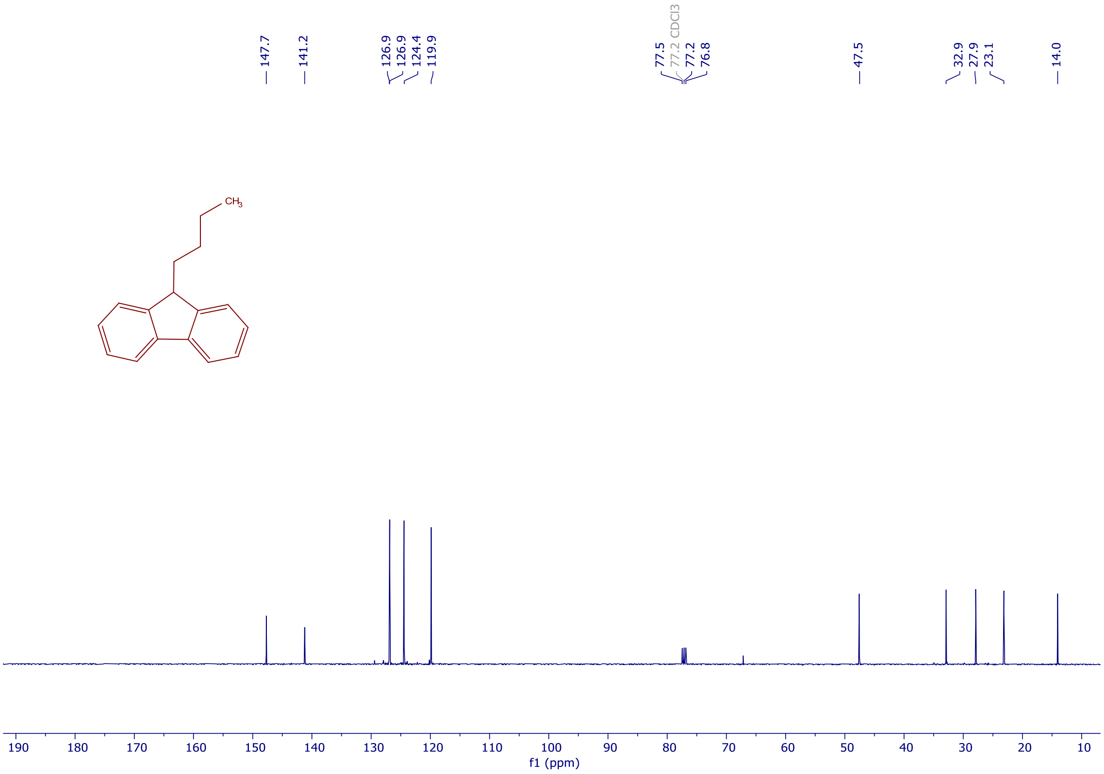
*^{1}H NMR of 9-(1-(p-tolyl)ethyl)-9H-fluorene (Compound-**7p**) in CDCl$_3$. # indicates the solvent impurity of H_2O in CDCl$_3$*



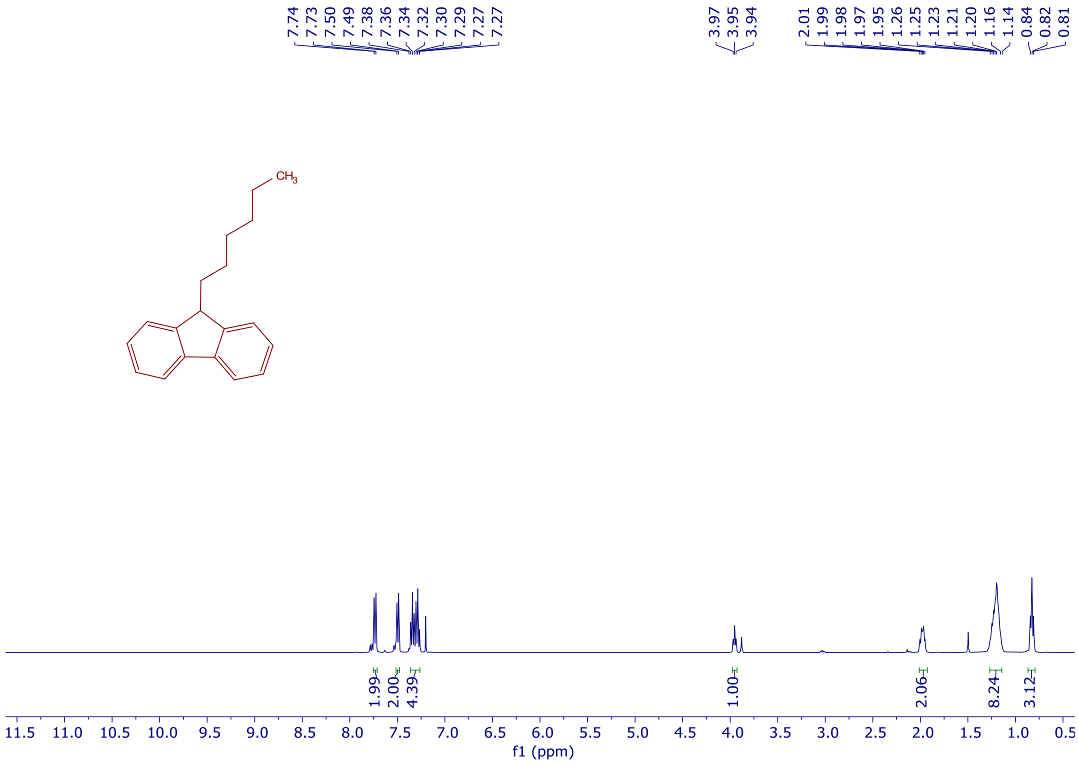
*^{13}C{^{1}H} NMR of 9-(1-(p-tolyl)ethyl)-9H-fluorene (Compound-**7p**) in CDCl$_3$*



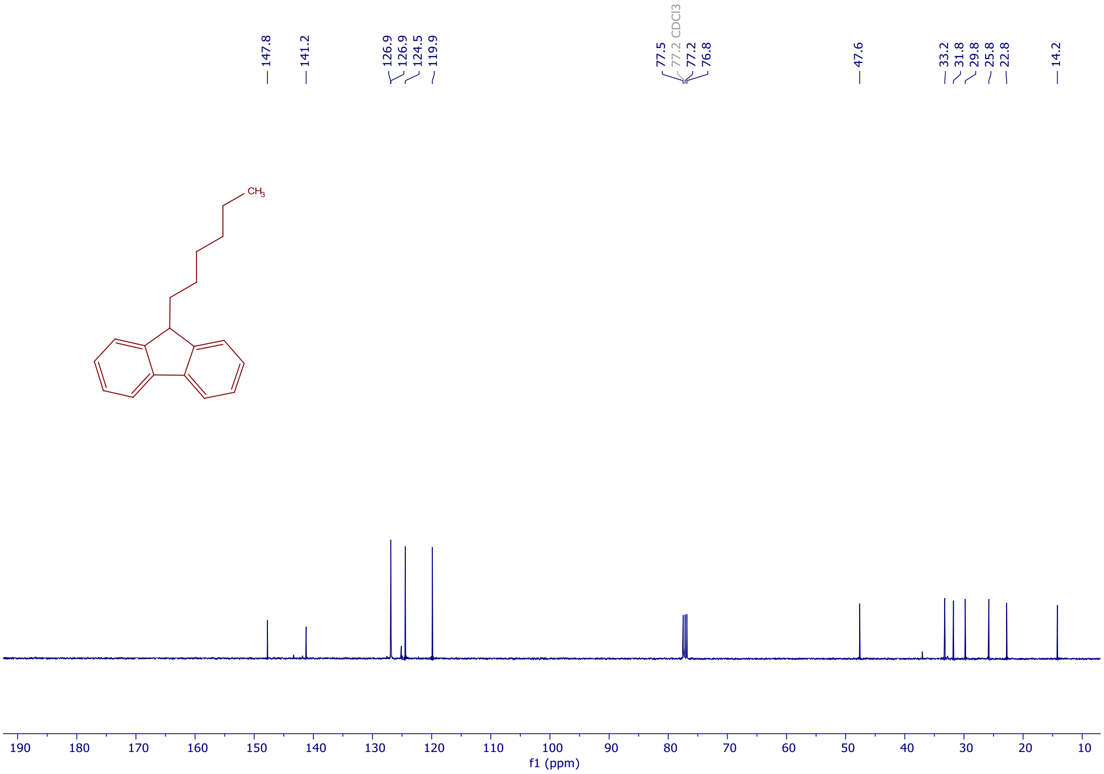
*^{1}H NMR of 9-butyl-9H-fluorene (Compound-**7q**) in $CDCl_3$*



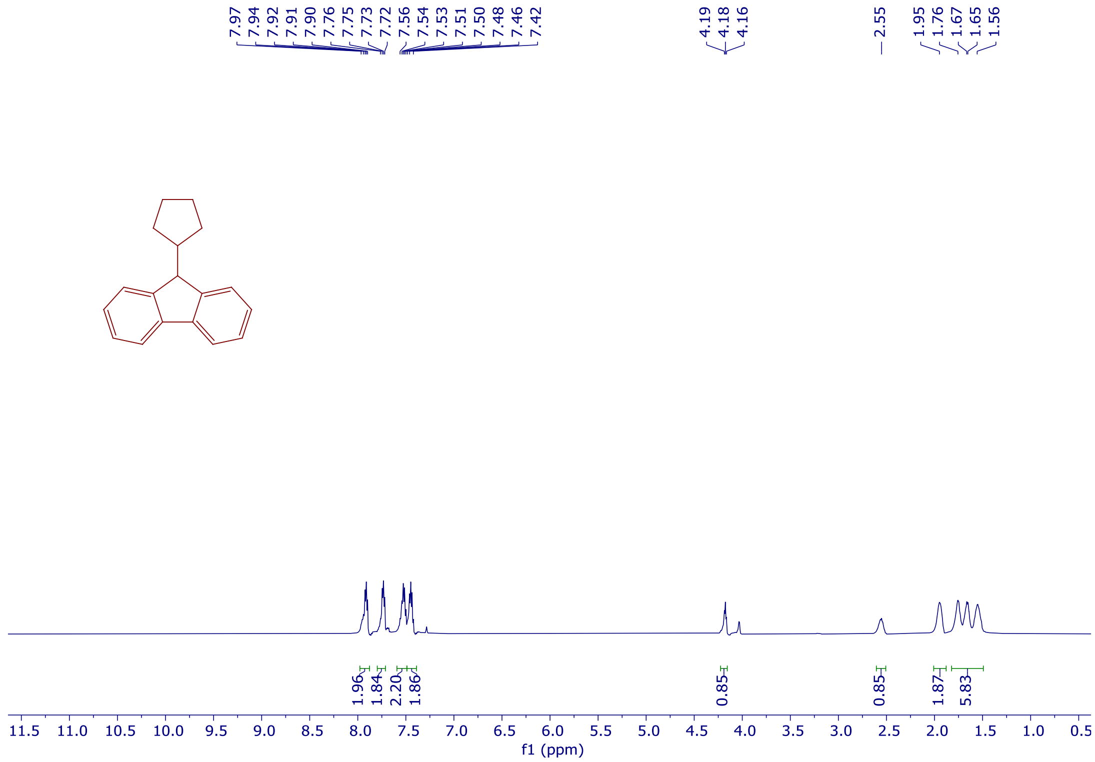
*$^{13}C\{^{1}H\}$ NMR of 9-butyl-9H-fluorene (Compound-**7q**) in $CDCl_3$*



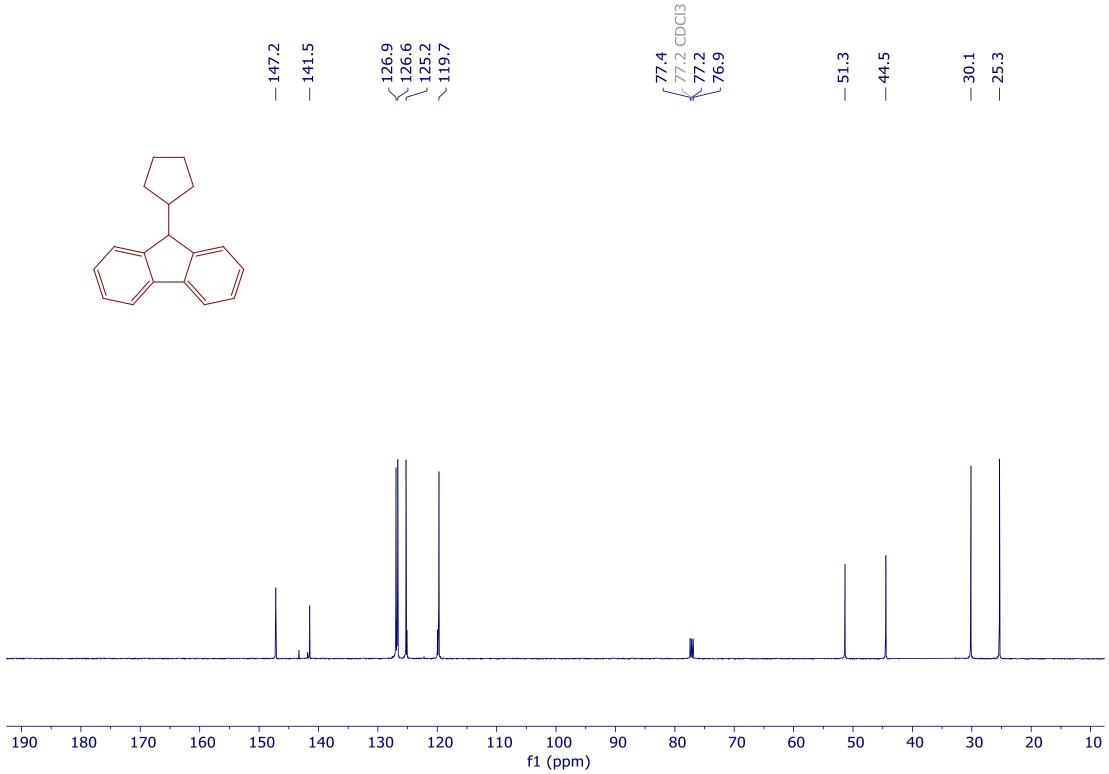
*^{1}H NMR of 9-hexyl-9H-fluorene (Compound-**7r**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



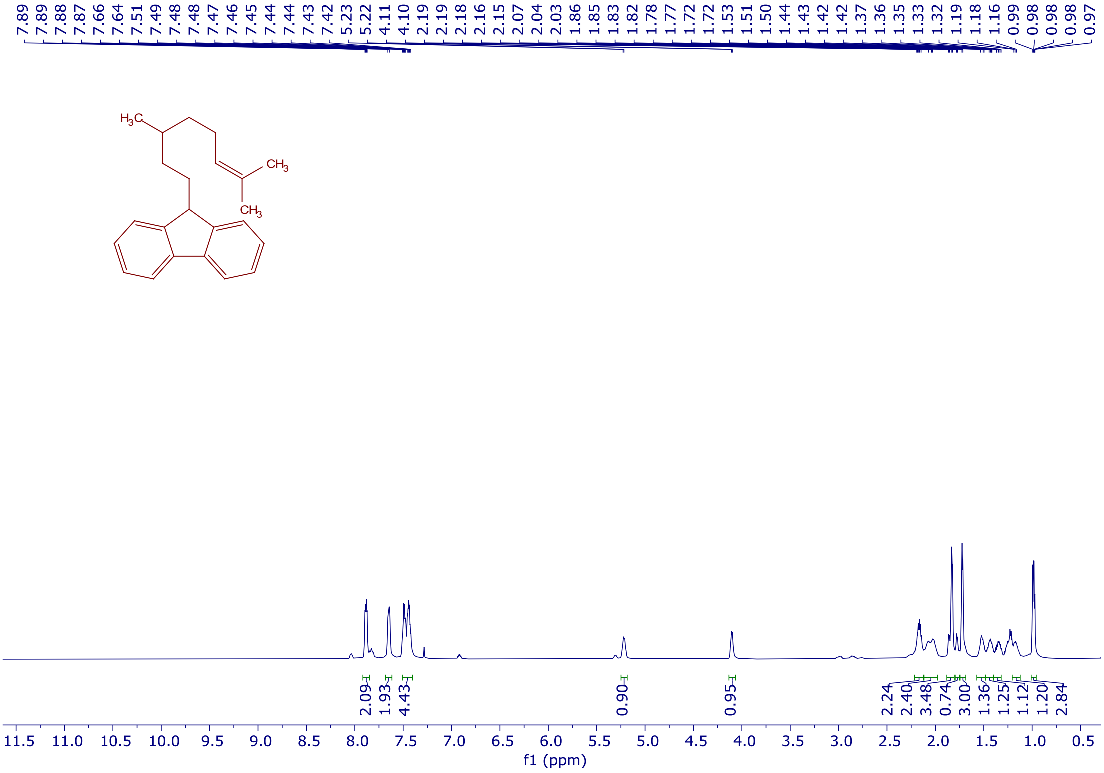
*$^{13}C\{^1H\}$ NMR of 9-hexyl-9H-fluorene (Compound-**7r**) in $CDCl_3$*



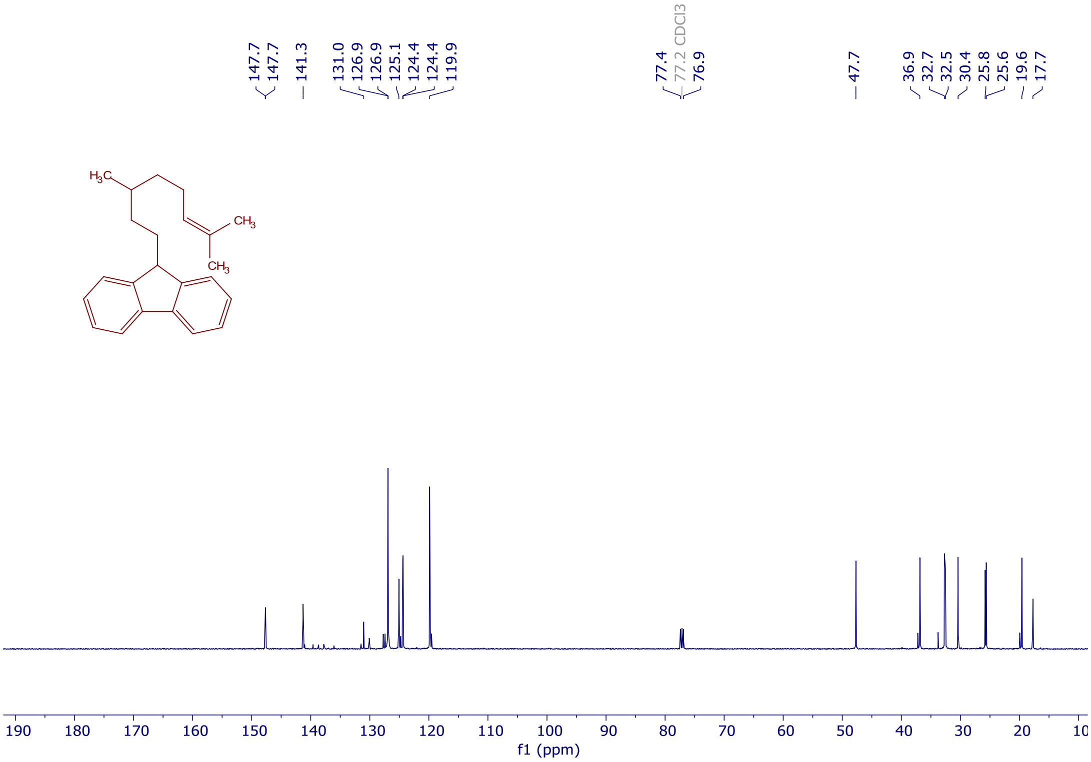
*^{1}H NMR of 9-cyclopentyl-9H-fluorene (Compound-**7s**) in CDCl$_3$*



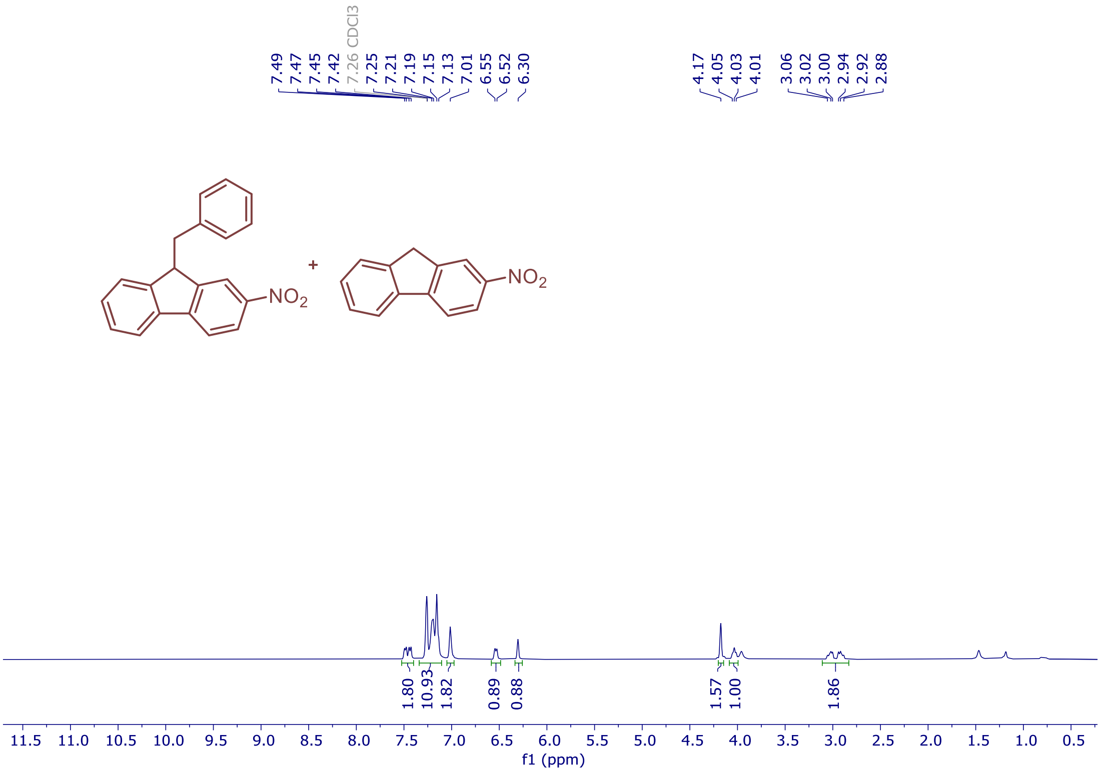
*^{13}C{^{1}H} NMR of 9-cyclopentyl-9H-fluorene (Compound-**7s**) in CDCl$_3$*



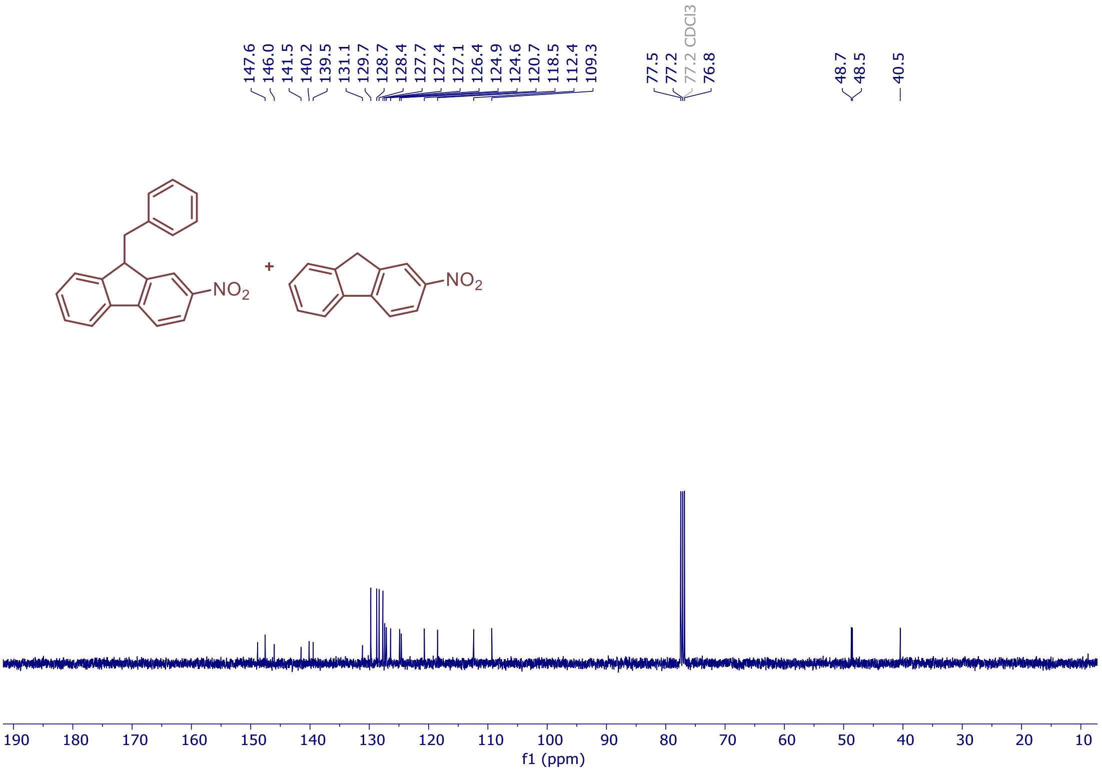
*^{1}H NMR of 9-(3,7-dimethyloct-6-en-1-yl)-9H-fluorene (Compound-**7t**) in CDCl$_3$*



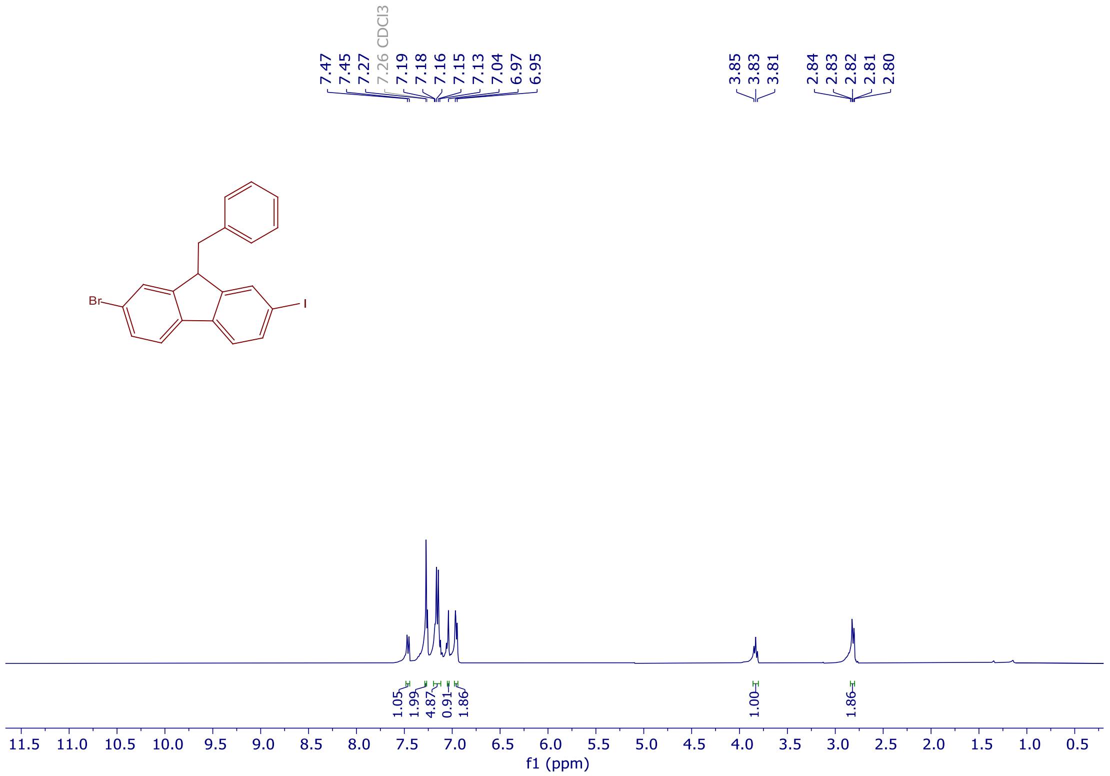
*^{13}C{^{1}H} NMR of 9-(3,7-dimethyloct-6-en-1-yl)-9H-fluorene (Compound-**7t**) in CDCl$_3$*



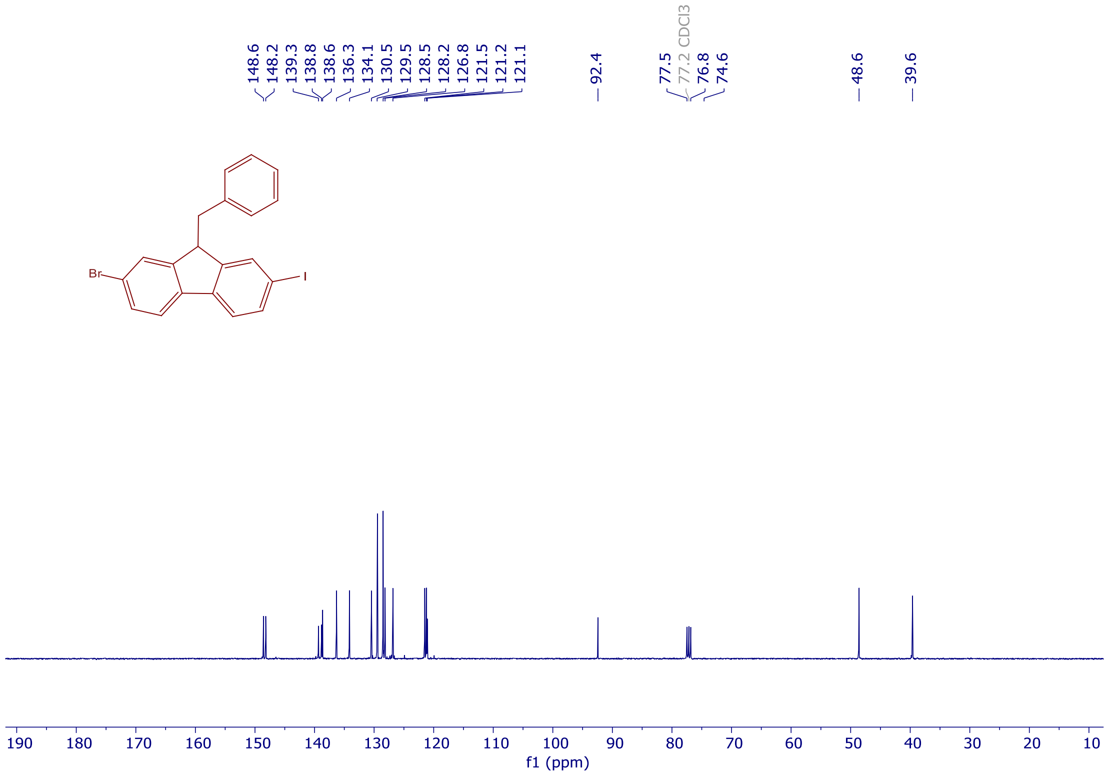
*^{1}H NMR of the reaction mixture of 9-benzyl-2-nitro-9H-fluorene (Compound-**7u**) and 2-nitro-9H-fluorene (Starting material) in CDCl$_3$*



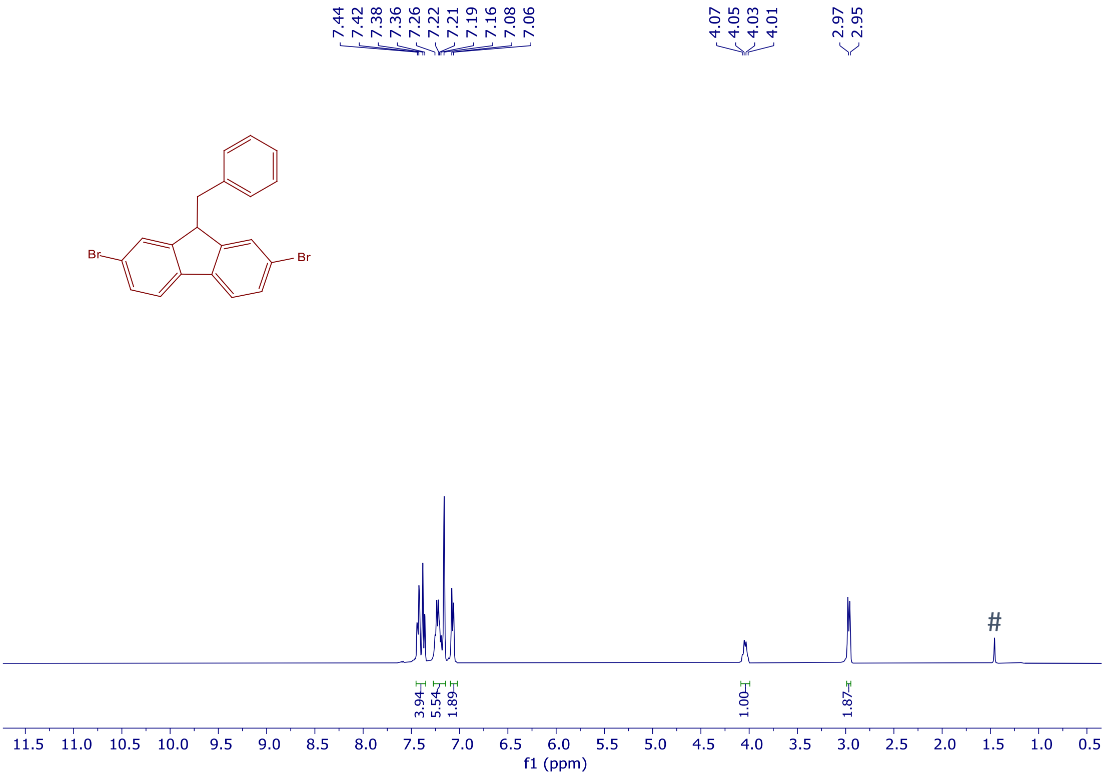
*^{13}C{^{1}H} NMR of the reaction mixture of 9-benzyl-2-nitro-9H-fluorene (Compound-**7u**) and 2-nitro-9H-fluorene in CDCl$_3$*



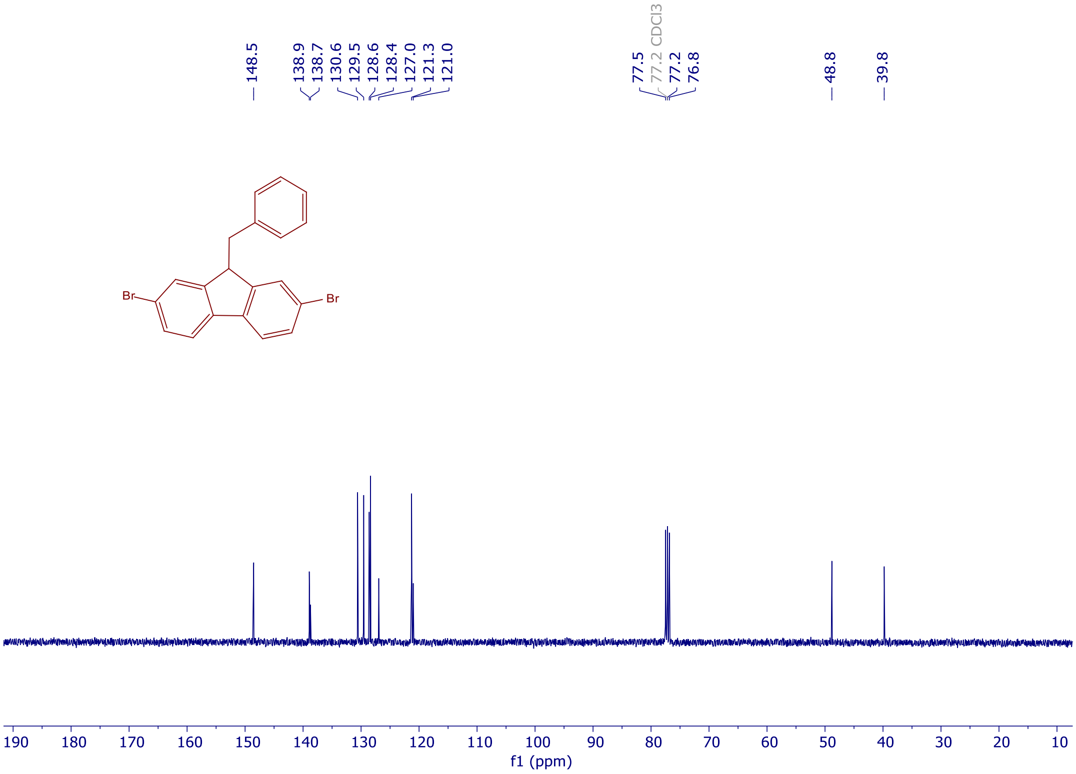
*^{1}H NMR of 9-benzyl-2-bromo-7-iodo-9H-fluorene (Compound-**7v**) in CDCl$_3$*



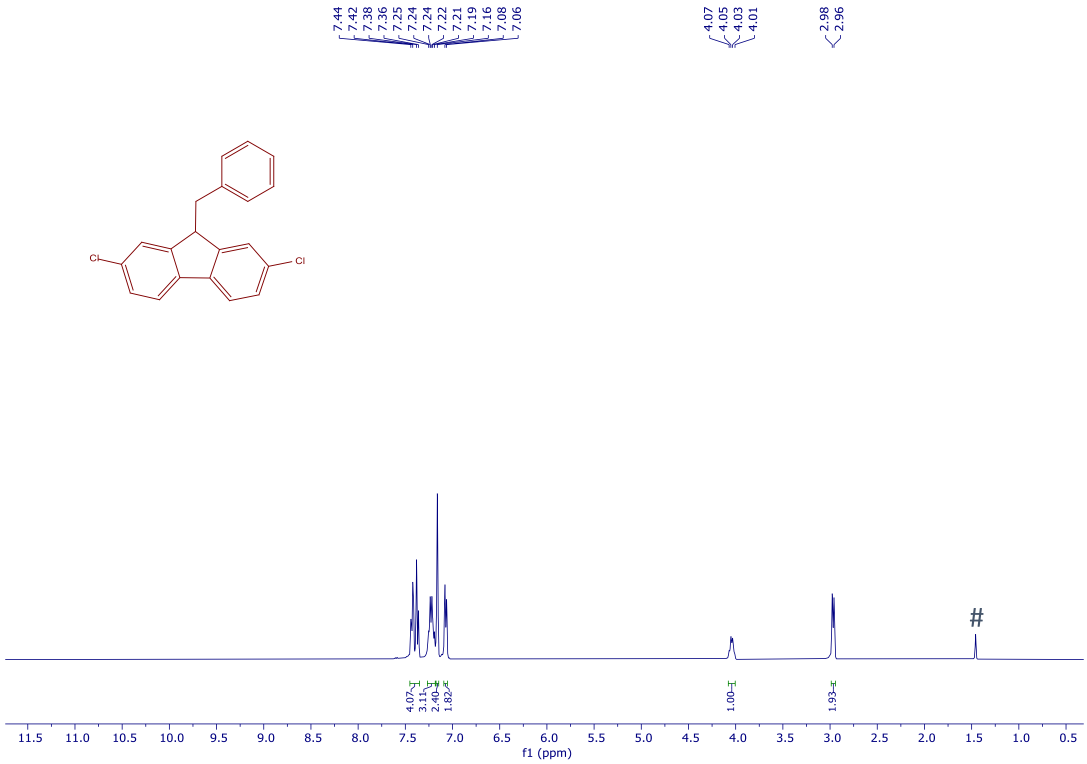
*^{13}C{^{1}H} NMR of 9-benzyl-2-bromo-7-iodo-9H-fluorene (Compound-**7v**) in CDCl$_3$*



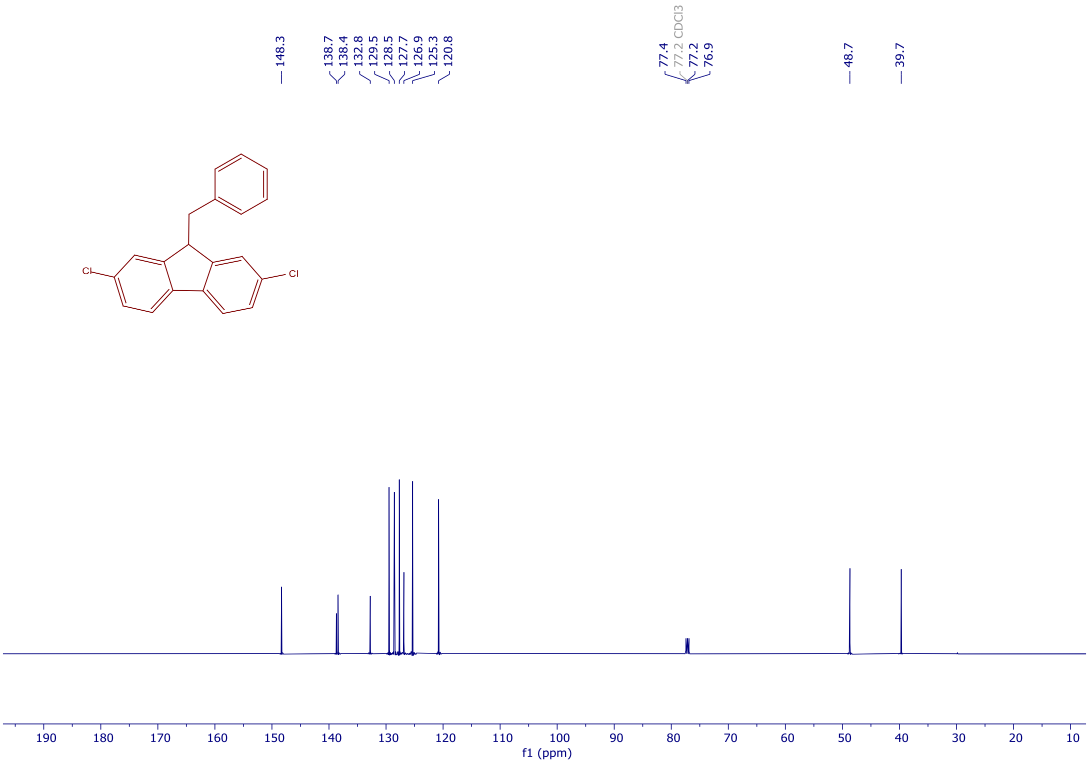
*^{1}H NMR of 9-benzyl-2,7-dibromo-9H-fluorene (Compound-**7w**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



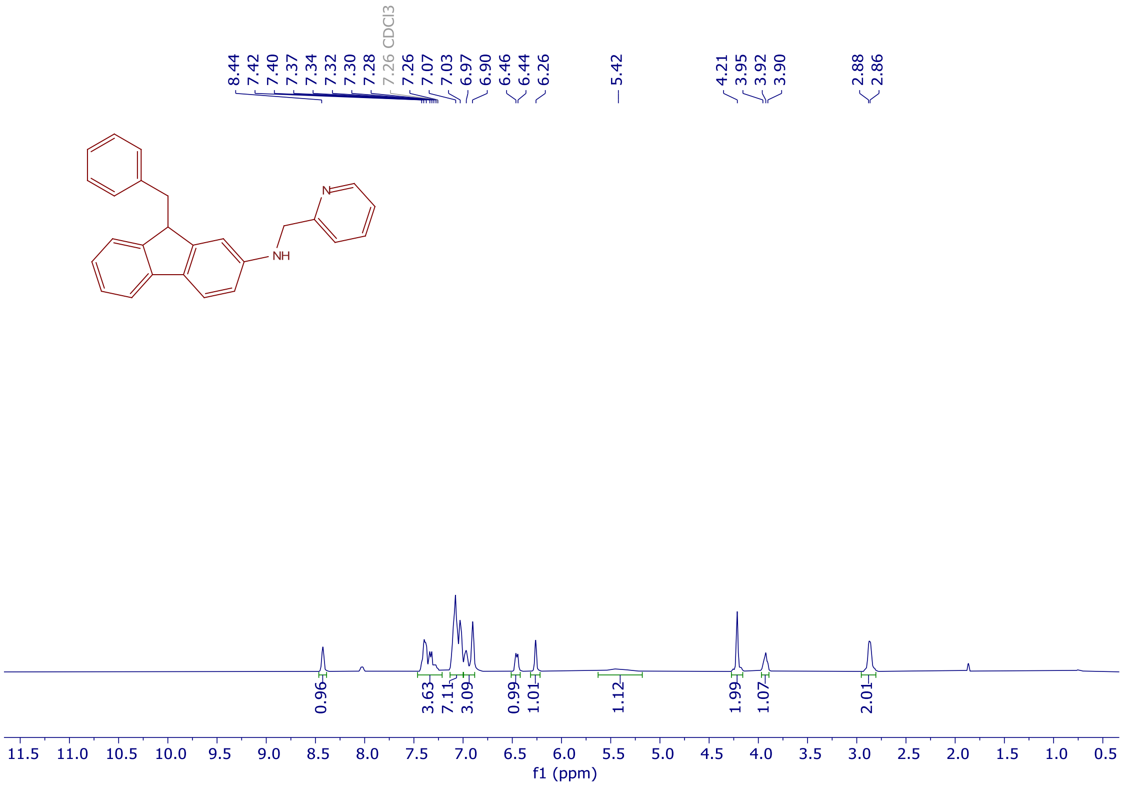
*^{13}C{^{1}H} NMR of 9-benzyl-2,7-dibromo-9H-fluorene (Compound-**7w**) in $CDCl_3$*



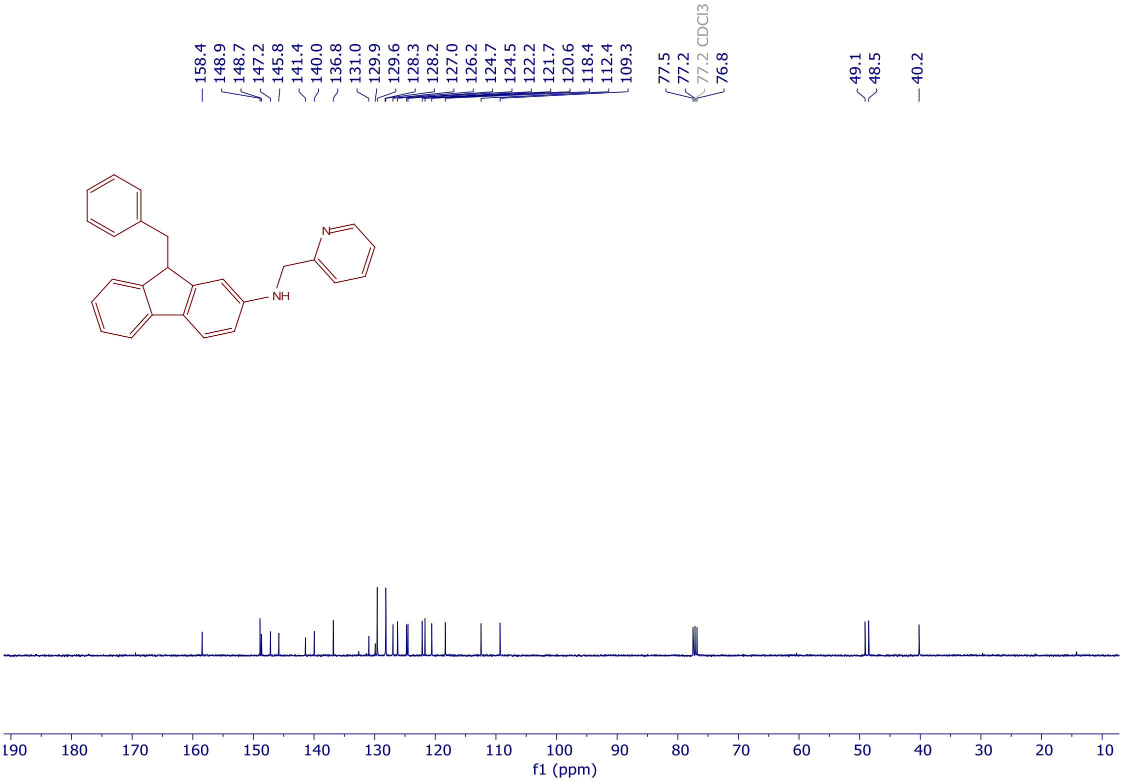
*^{1}H NMR of 9-benzyl-2,7-dichloro-9H-fluorene (Compound-**7x**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



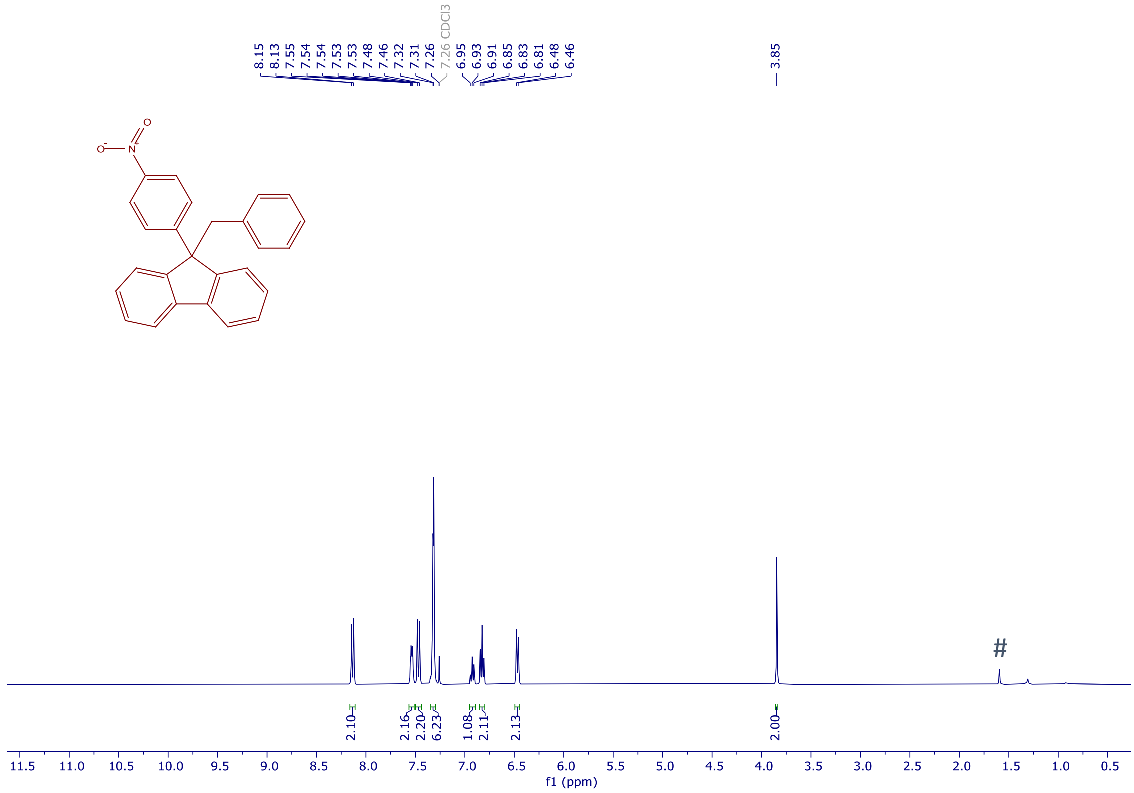
*^{13}C{^{1}H} NMR of 9-benzyl-2,7-dichloro-9H-fluorene (Compound-**7x**) in $CDCl_3$*



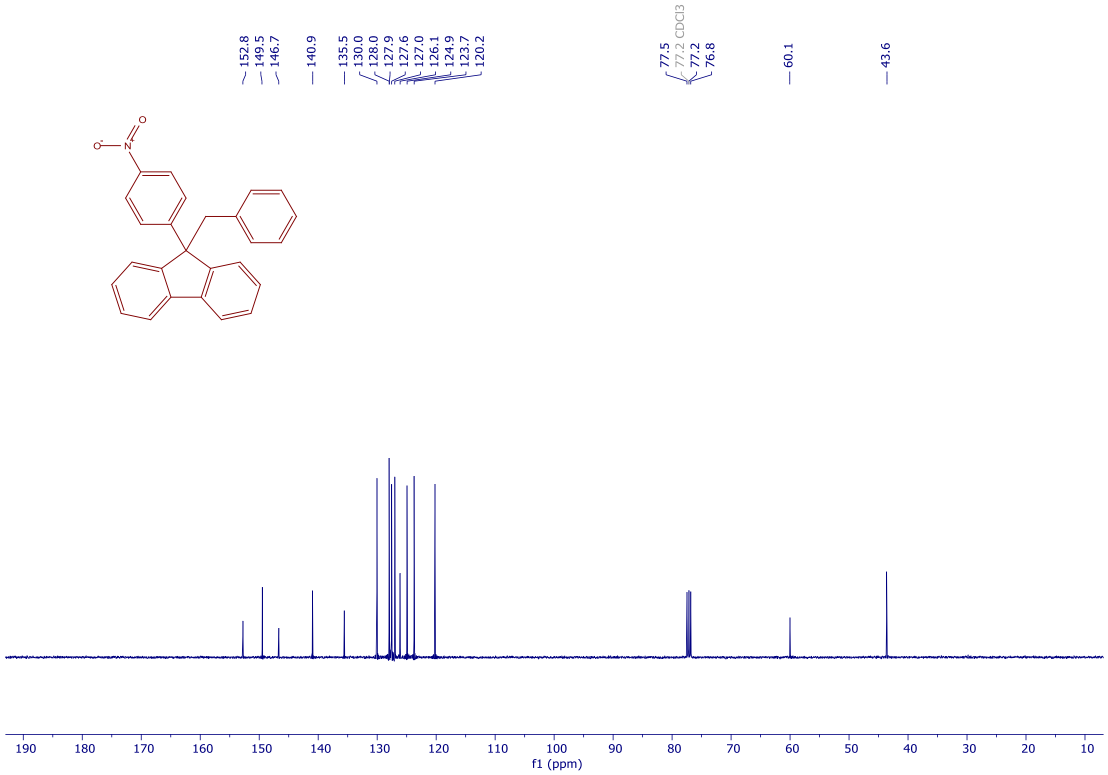
*^{1}H NMR of 9-benzyl-N-(pyridin-2-ylmethyl)-9H-fluoren-2-amine (Compound-**7y**) in CDCl$_3$*



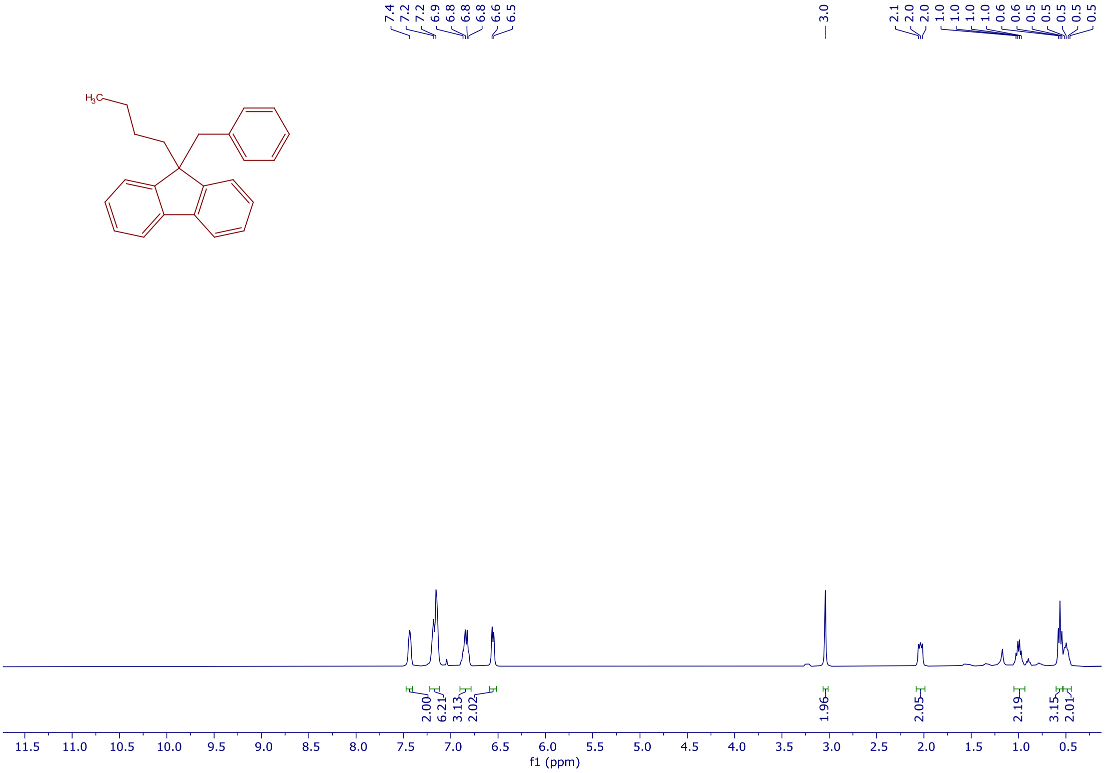
*^{13}C{^{1}H} NMR of 9-benzyl-N-(pyridin-2-ylmethyl)-9H-fluoren-2-amine (Compound-**7y**) in CDCl$_3$*



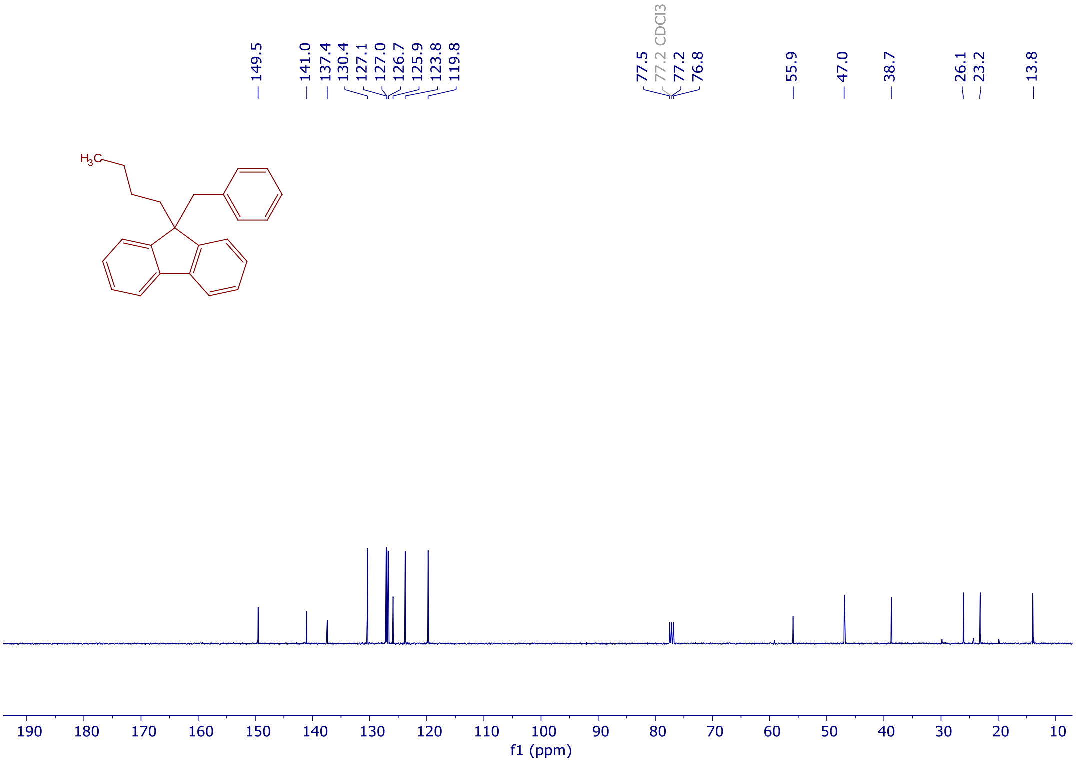
*^1H NMR of 9-benzyl-9-(4-nitrophenyl)-9H-fluorene (Compound-**8a**) in $CDCl_3$. # indicates the solvent impurity of H_2O in $CDCl_3$*



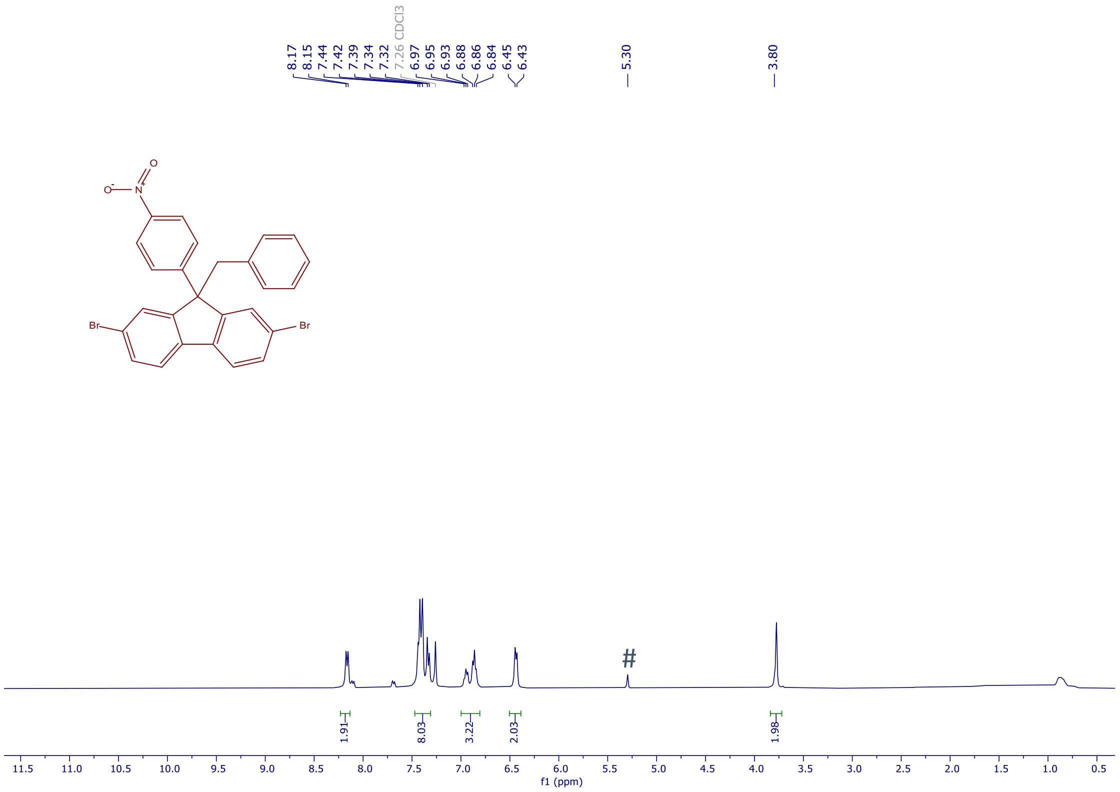
*$^{13}C\{^1H\}$ NMR of 9-benzyl-9-(4-nitrophenyl)-9H-fluorene (Compound-**8a**) in $CDCl_3$*



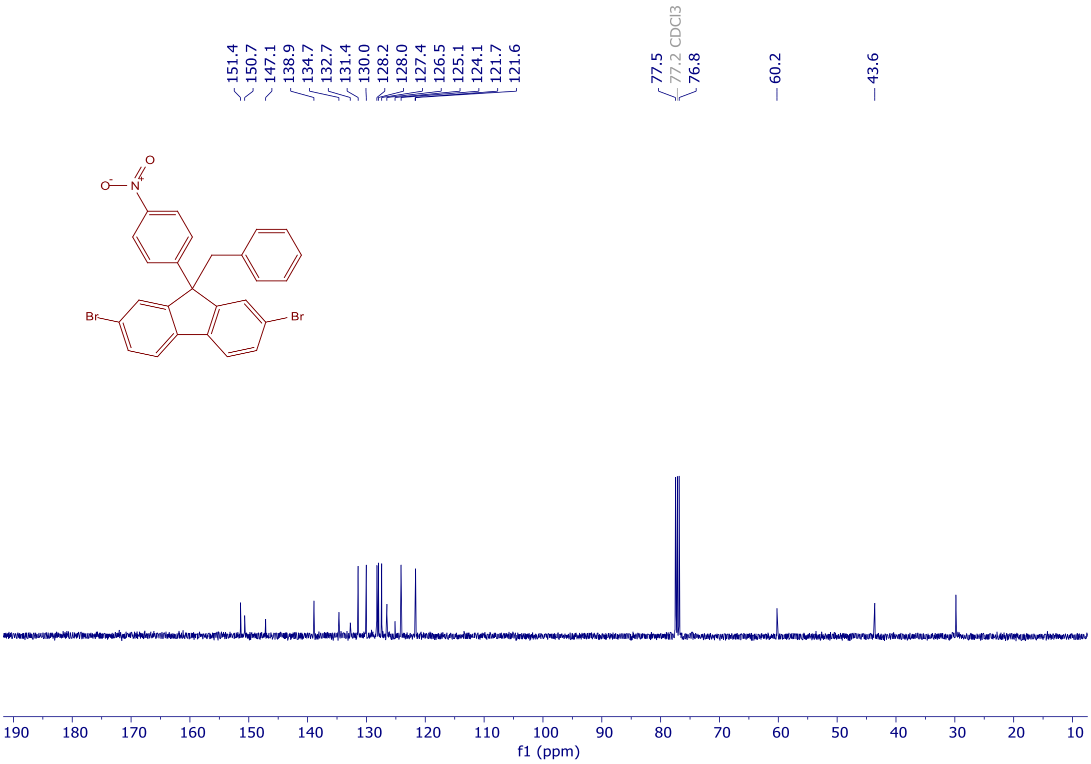
^{1}H NMR of 9-benzyl-9-(4-nitrophenyl)-9H-fluorene (Compound-**8b**) in CDCl$_3$



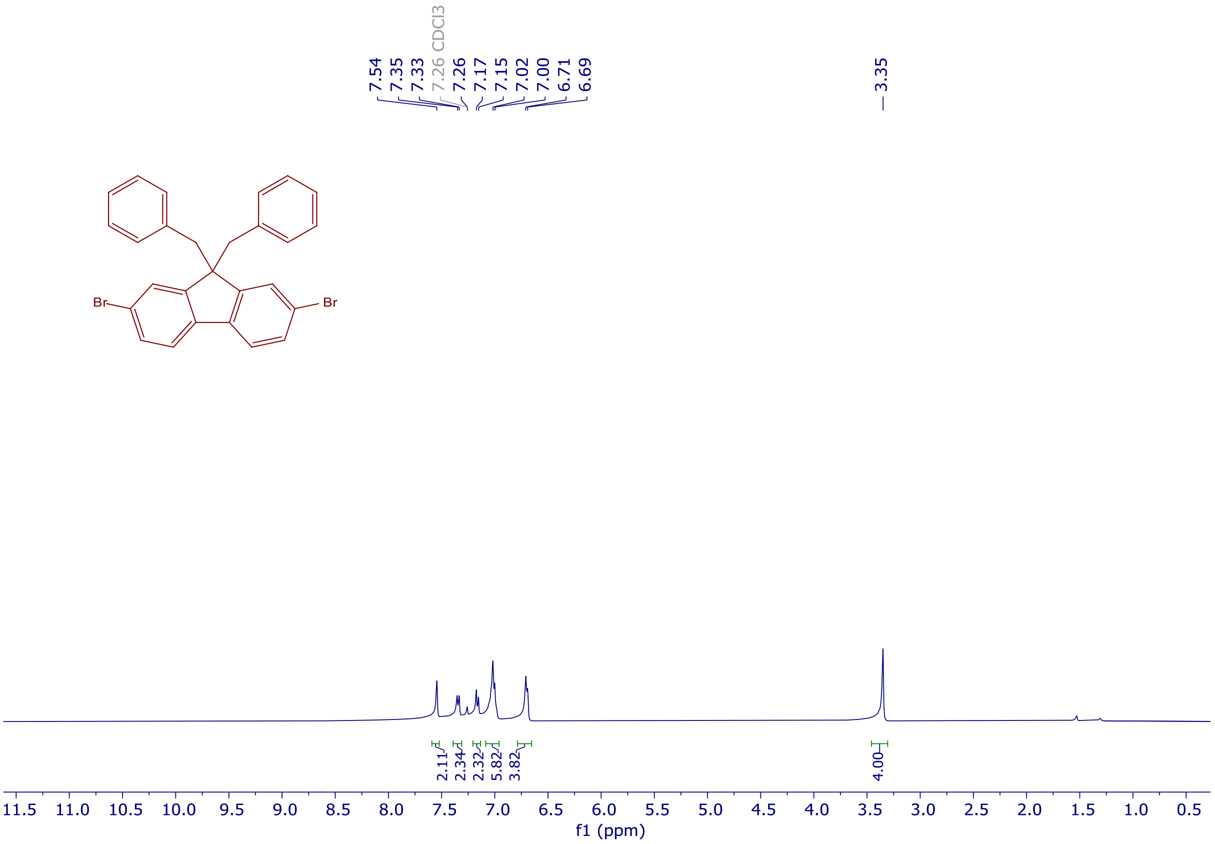
^{13}C{^{1}H} NMR of 9-benzyl-9-(4-nitrophenyl)-9H-fluorene (Compound-**8b**) in CDCl$_3$



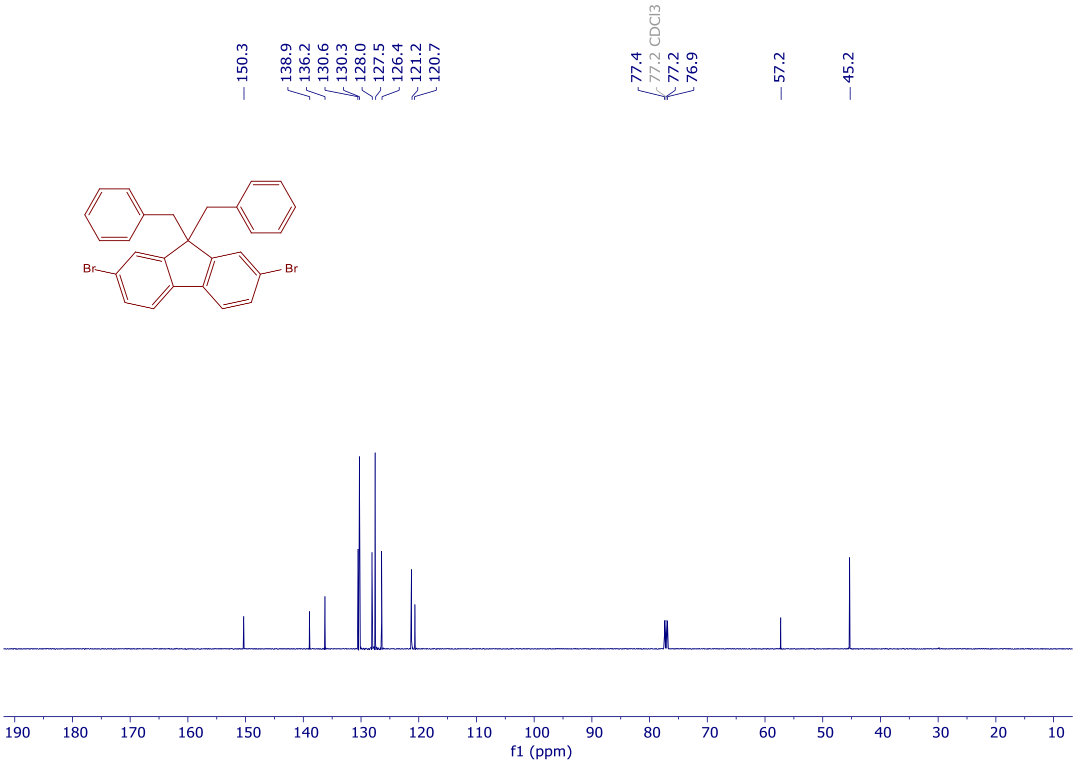
*^{1}H NMR of 9-benzyl-2,7-dibromo-9-(4-nitrophenyl)-9H-fluorene (Compound-**8c**) in $CDCl_3$. # indicates the solvent impurity of DCM in $CDCl_3$*



*$^{13}C\{^1H\}$ NMR of 9-benzyl-2,7-dibromo-9-(4-nitrophenyl)-9H-fluorene (Compound-**8c**) in $CDCl_3$*



*^{1}H NMR of 9,9-dibenzyl-2,7-dibromo-9H-fluorene (Compound-**8d**) in $CDCl_3$*



*^{13}C{^{1}H} NMR of 9,9-dibenzyl-2,7-dibromo-9H-fluorene (Compound-**8d**) in $CDCl_3$*

17. Mechanistic study:

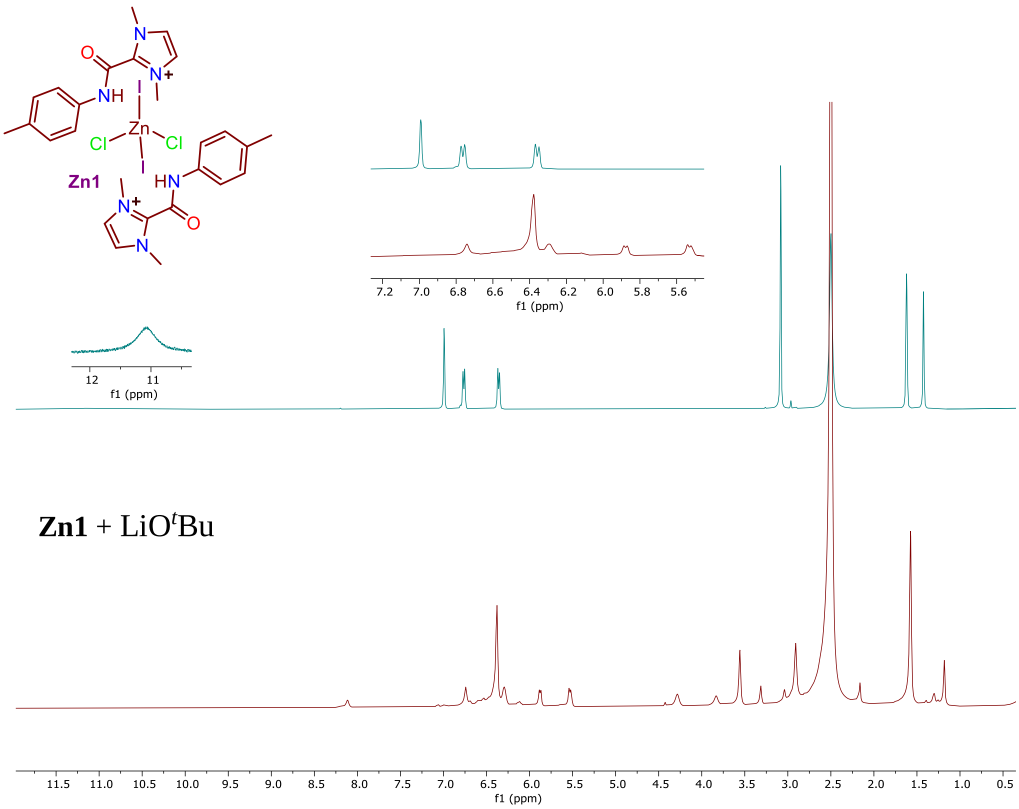


Figure S21. *Stacking of ^{1}H spectra between **Zn1** and Compound **Zn1a** (showing NMR broadening after 1 h) in DMSO-d_6*

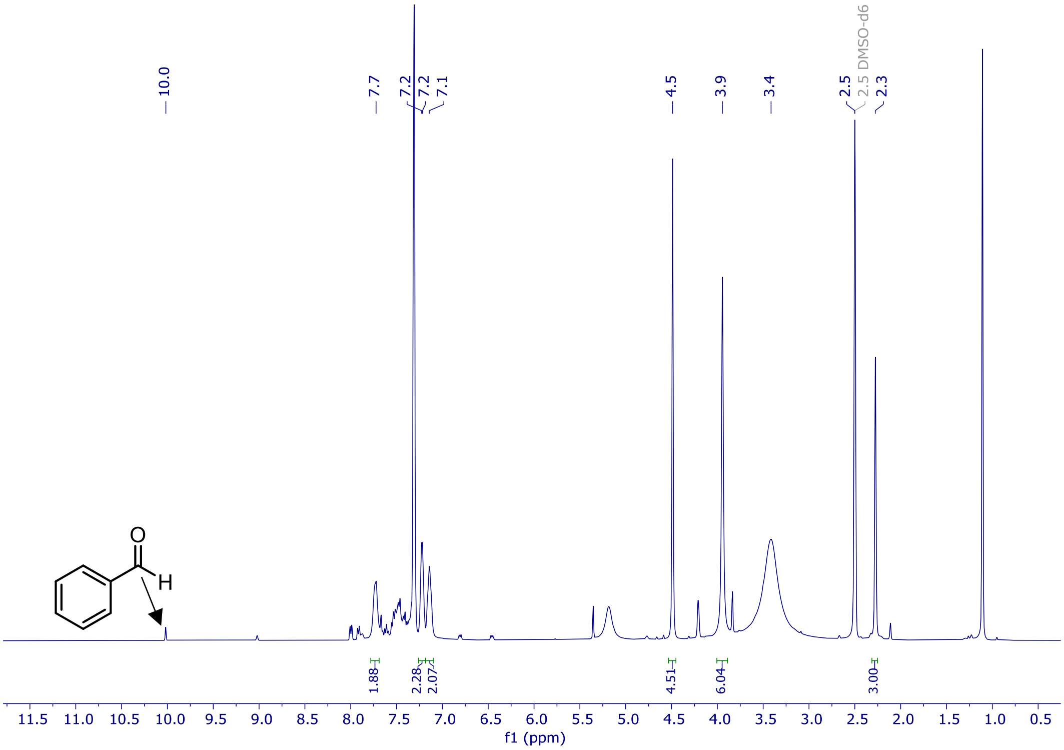


Figure S22. *^{1}H NMR spectrum of the reaction mixture in DMSO-d_6 after adding of 1 equiv. of benzyl alcohol to the reaction mixture (Figure S21) showing formation of the benzaldehyde*

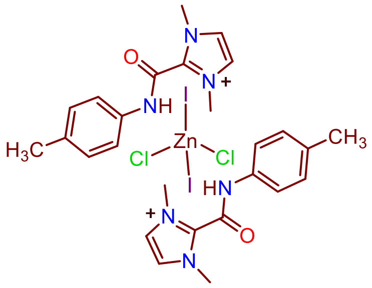
Table S4. Crystallographic data for the compound **Zn1**

Compound	**Zn1**
CCDC No	2284848
Empirical formula	$C_{26}H_{32}Cl_2I_2N_6O_2Zn$
Formula weight	850.67
Crystal system	Monoclinic
Space group	*C* 2/c
a (Å)	22.888(3)
b (Å)	9.043(11)
c (Å)	17.724(2)
α (°)	90
β (°)	115.556(4)
γ (°)	90
V (Å^3)	3309.3(7)
Z	4
D calc (Mg/m^3)	1.707
F (000)	1664
μ (mm^{-1})	2.802
θ Range (°)	1.937 to 24.999
Crystal size (mm)	0.150 x 0.120 x 0.100
No. of total reflns collected	10412
No. of unique reflns [I > 2σ(I)]	2866
Data/restraints/ parameters	2866/ 0 / 180
Goodness-of-fit on F^2	0.984
Final R indices [I > 2σ(I)]	0.0653, 0.1688
R indices (all data)	0.0932, 0.1905

18. Computational data

All the calculations were performed using the Gaussian 16, Revision B.01 program.[15] All structures were optimized with B3LYP[16] functional. Metals (Zn) and I were treated with LANL2DZ basis set with an effective core potential, while the other atoms were treated using using 6-31G**, a double-ζ Pople type basis set.

Cartesian Coordinates of all the optimized geometries:



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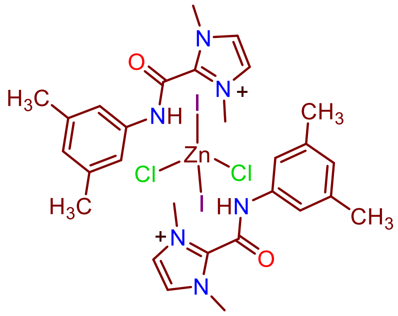

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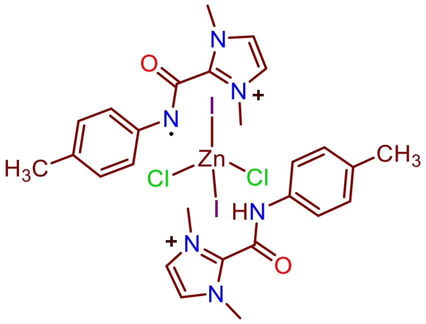

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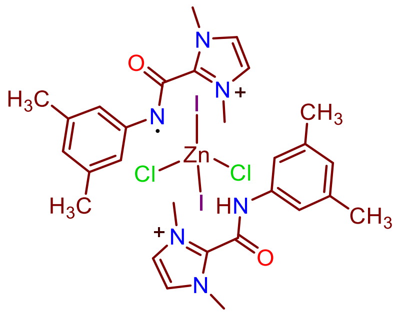

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