Supporting Information

Pyrazole-pyridine-pyrazole (NNN) ruthenium (II) complex catalyzed acceptorless-dehydrogenation of alcohols to aldehydes

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1. General remarks

All commercial reagents were ordered from Innochem, Sigma-Aldrich, Aladdin, TCI, Titan, Energy Chemical, or Bidepharm. Nitrogen (99.9%) were supplied by Southwest Institute of Chengdu. Unless otherwise stated, commercial reagents were used without purification. All manipulations of air- and moisture-sensitive reagents were performed under an inert atmosphere of nitrogen in either a vacuum atmosphere or using standard Schlenk techniques. All solvents were reagent grade or higher, unless otherwise noted, all operations were performed under the nitrogen balloon and all solvents were performed degassing and dehydrating treatment. ^{1}H and ^{13}C and ^{31}P nuclear magnetic resonance (NMR) spectra were recorded on Bruker AVANCE IIIHD 400 NMR spectrometers. Multiplets were assigned as s (singlet), d (doublet), t (triplet), dd (doublet of doublet), and m (multiplet). Single crystal X-ray structure analyses data were collected on a Bruker D8 VENTURE. High resolution mass spectra (HRMS) were obtained with a Shimadzu (LCMS-IT-TOF). The data are given as mass units per charge (m/z) and intensities of signals are given in brackets. Gas chromatography and mass spectrometry (GC-MS) analysis was conducted on a Shimadzu GCMS-QP2020 (SH-Rtx-35MS, 30 m × 0.25 mm × 0.25 μm)

2. Typical Procedure for the AD of Alcohols

After **1** (0.005 mmol, 4.7 mg), K_3PO_4 (0.8 mmol, 169.8 mg) were added into a dried Schlenk tube equipped with a N_2 balloon, and it was subject to three cycles of vacuum and refilled with nitrogen. Benzyl alcohol (1.0 mmol, 104 μL) and anhydrous toluene (2 mL) were sequentially injected into the tube. The solution was stirred at a certain temperature for 2 days. At the end of the reaction, the mixture was cooled to room temperature and diluted with dichloromethane and filtered with Celite. The yield was calculated by GC-MS and isolating.

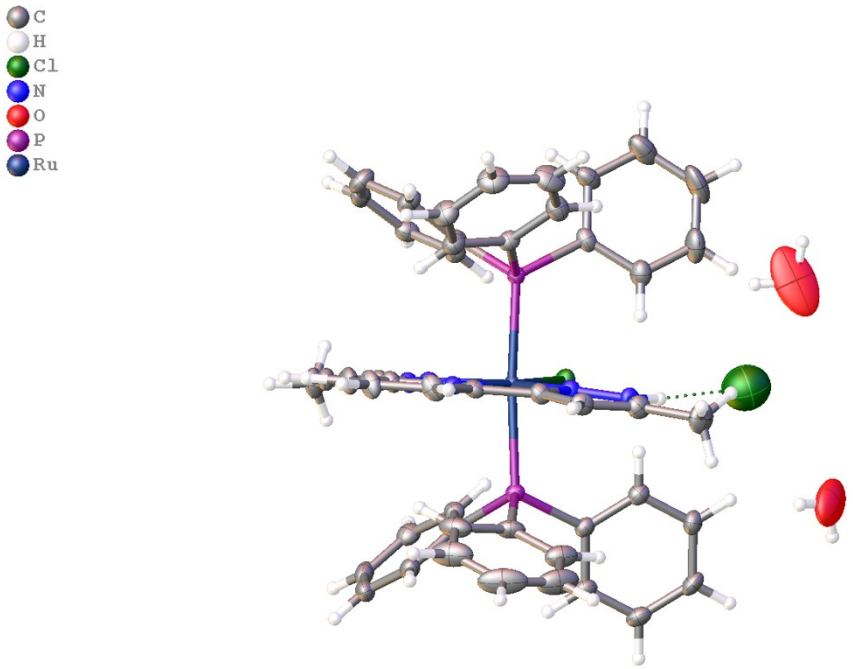
Isolation method: After the reaction was cooled to room temperature, the solid was removed by filtration and the filtrate was concentrated to 1 mL, subsequently it was purified by column chromatography on silica gel with ethyl acetate/petroleum ether (1/30, v/v) as eluent to obtain the aldehyde product.

3. Typical Procedure for the sequential AD/condensation reaction between the primary and secondary alcohols

After **1** (0.005 mmol, 4.7 mg), K_3PO_4 (0.8 mmol, 169.8 mg) were added into a dried Schlenk tube equipped with a N_2 balloon, and it was subject to three cycles of vacuum and refilled with nitrogen. Benzyl alcohol (0.5 mmol, 52 μL), 1-phenylethanoland (1.0 mmol, 122 μL) and anhydrous toluene (2 mL) were sequentially injected into the tube. The solution was stirred at a certain temperature for 3 days. At the end of the reaction, the mixture was slowly cooled to room temperature and diluted with 3 mL of CH_2Cl_2 and filtered with diatomite. The filtrate was evaporated to afford a brown residue, and the residue was purified by flash column chromatography on silica gel to afford the product.

4. X-ray single crystal data of 1, 2, 3

4.1 X-ray single crystal data of **1**
CCDC No. of **1** is **2278192**.

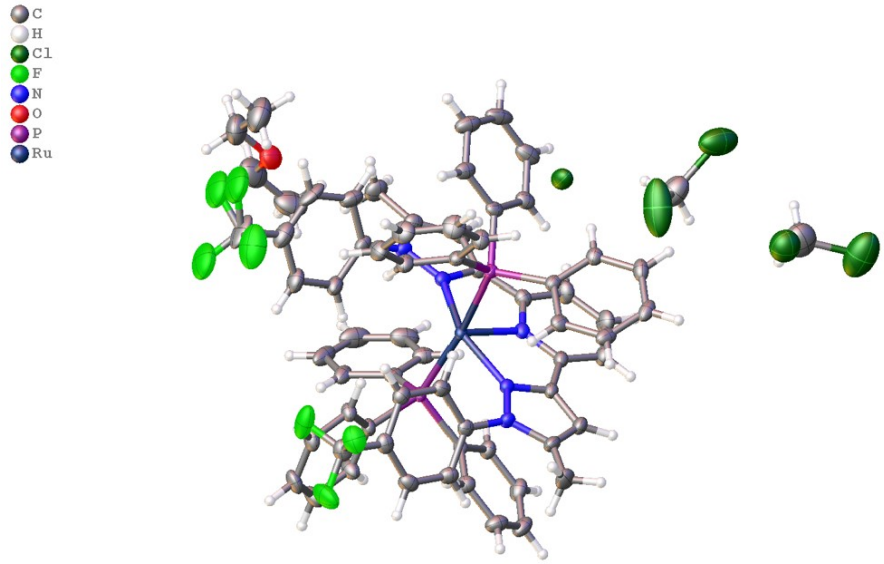


Empirical formula	$C_{49}H_{47}Cl_2N_5O_2P_2Ru$
Formula weight	970.82
Temperature/K	138.0
Crystal system	monoclinic
Space group	$P2_1/c$
a/Å	11.8398(8)

b/Å	21.1962(19)
c/Å	19.7092(17)
α/°	90
β/°	94.851(3)
γ/°	90
Volume/Å^3	4928.5(7)
Z	4
ρ_{calc}g/cm^3	1.308
μ/mm^{-1}	0.533
F(000)	1996.0
Crystal size/mm^3	0.19 × 0.16 × 0.14
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	3.844 to 50
Index ranges	-14 ≤ h ≤ 11, -25 ≤ k ≤ 21, -23 ≤ l ≤ 23
Reflections collected	39187
Independent reflections	8644 [R_{int} = 0.0575, R_{sigma} = 0.0498]
Data/restraints/parameters	8644/6/562
Goodness-of-fit on F^2	1.045
Final R indexes [I>=2σ (I)]	R_1 = 0.0610, wR_2 = 0.1930
Final R indexes [all data]	R_1 = 0.0682, wR_2 = 0.2014
Largest diff. peak/hole / e Å^{-3}	1.63/-1.22

4.2 X-ray single crystal data of **2**

CCDC No. of **2** is **2278195**.

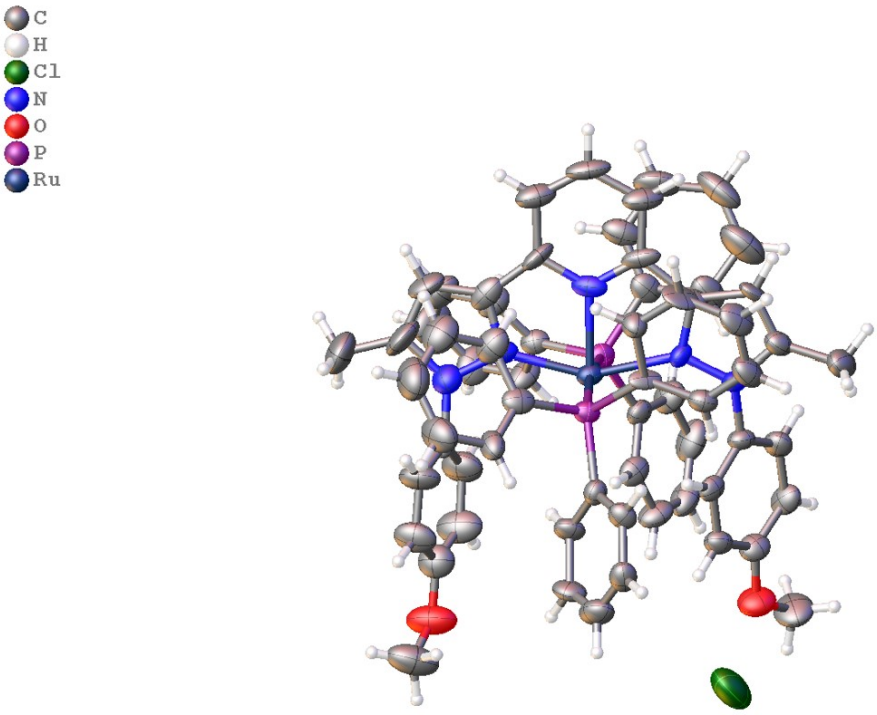


Empirical formula	$C_{69}H_{63}Cl_5F_6N_5OP_2Ru$
Formula weight	1432.50
Temperature/K	120.0
Crystal system	triclinic
Space group	P-1
a/Å	11.5849(7)

b/Å	13.4625(10)
c/Å	22.7608(16)
α/°	103.721(3)
β/°	95.440(2)
γ/°	101.016(2)
Volume/Å^3	3348.2(4)
Z	2
ρ_{calc}g/cm^3	1.421
μ/mm^{-1}	0.545
F(000)	1466.0
Crystal size/mm^3	0.5 × 0.39 × 0.25
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	4.096 to 50
Index ranges	-13 ≤ h ≤ 13, -16 ≤ k ≤ 16, -27 ≤ l ≤ 27
Reflections collected	65880
Independent reflections	11738 [R_{int} = 0.0669, R_{sigma} = 0.0397]
Data/restraints/parameters	11738/264/804
Goodness-of-fit on F^2	1.042
Final R indexes [I>=2σ (I)]	R_1 = 0.0469, wR_2 = 0.1206
Final R indexes [all data]	R_1 = 0.0544, wR_2 = 0.1263
Largest diff. peak/hole / e Å^{-3}	1.28/-1.27

4.3 X-ray single crystal data of **3**

CCDC No. of **3** is **2256798**.



Empirical formula	$C_{63}H_{55}ClN_5O_2P_2Ru$

Formula weight	1112.58
Temperature/K	293.15
Crystal system	monoclinic
Space group	$P2_1/n$
a/Å	11.2274(5)
b/Å	24.983(2)
c/Å	21.3970(14)
α/°	90
β/°	94.992(6)
γ/°	90
Volume/Å^3	5979.1(8)
Z	4
ρ_{calc}g/cm^3	1.236
μ/mm^{-1}	0.405
F(000)	2300.0
Crystal size/mm^3	0.4 × 0.3 × 0.3
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	5.962 to 50
Index ranges	-13 ≤ h ≤ 13, -28 ≤ k ≤ 29, -25 ≤ l ≤ 25
Reflections collected	24187
Independent reflections	10456 [R_{int} = 0.0886, R_{sigma} = 0.1185]
Data/restraints/parameters	10456/0/671
Goodness-of-fit on F^2	1.094
Final R indexes [I>=2σ (I)]	R_1 = 0.1322, wR_2 = 0.2438
Final R indexes [all data]	R_1 = 0.1558, wR_2 = 0.2571
Largest diff. peak/hole / e Å^{-3}	1.05/-2.00

5. Synthesis of complexes

5.1 Synthesis of **L1-L3**

Pyridine 2,6-dicarboxylate (15 g) and anhydrous ethanol (120 mL) were added to a 250 mL round bottom flask equipped with reflux condensation device and acetyl chloride (25 mL) was added dropwise and the reaction was carried out at room temperature for 24 h. After the reaction, the solid was removed by filtration and the filtrate was neutralized with sodium carbonate solution, and then extracted with ethyl acetate to obtain a white solid diethyl pyridine 2,6-dicarboxylate. Diethyl pyridine 2,6-dicarboxylateand sodium ethanol (8.87 g) were added to toluene (60 mL) solution under ice bath, and toluene solution dissolved with 0.12 mol acetone was added drop by drop to react for 6h, gradually producing a yellow turbidity. Raise to room temperature and continue the reaction for 10h, the toluene was removed by filtration, and the solid was neutralized

with dilute hydrochloric acid to pH=5, then extracted with ethyl acetate to obtain a yellow solid 1,1'-(pyridine-2,6-diyl) bis(butane-1,3-dione). Added 1,1'-(pyridine-2,6-diyl) bis(butane-1,3-dione) (2mmol), glacial acetic acid (4mL), and ethanol (30mL) to a 100mL round bottom flask, stirred at room temperature for 30 minutes, and then added an ethanol solution containing 4mmol of hydrazine hydrate through a constant pressure drop funnel. Then, raised the temperature to 65℃ and continued the reaction for 24h. After the reaction, concentrated the solution to about 10mL, and then neutralized with sodium carbonate solution to pH=8, subsequently, extracted with ethyl acetate to obtain yellow solid **L1**. **L2-L3** were readily prepared by adding different hydrazine reagents (**L2**: 4-(Trifluoromethyl) phenylhydrazine; **L3**: (4-methoxyphenyl) hydrazine).

5.2 Synthesis of **1**

The mixture of ligand **L1** (1 mmol) and $RuCl_3 \bullet 3H_2O$ (1 mmol) was heated in refluxed ethanol (60 mL) for 5h. The color of the solution changed from black-brown to red-brown slowly and further generated red-brown precipitates. After being cooled to room temperature, the precipitates were filtered, washed with EtOH and Et_2O, and dried under vacuum. Then, Methanol (30 mL) and TPP (2 mmol) were added into the filtered precipitate and the mixture was heated at reflux for 6 h. At the end of the reaction, the solvent was removed under vacuum to give a solid substance. The resultant solid was dissolved in dichloromethane followed by removal of the insoluble impurities by filtration to give a red solution. After the diethyl ether was added to the solution, the orange solid complex **1** was recrystallized from the solution, in yields of 50%. ^{1}H NMR (400 MHz, DMSO-d_6) δ 13.50 (s, 2H), 7.43 – 7.38 (m, 1H), 7.29 (t, J = 7.2 Hz, 6H), 7.16 (qt, J = 12.4, 7.3 Hz, 24H), 7.00 (d, J = 7.8 Hz, 2H), 6.44 (s, 2H), 2.14 (s, 6H). ^{31}P{^{1}H} NMR (162 MHz, methanol-d_4) δ 24.36. HRMS (ESI-TOF) m/z: $[M-Cl]^+$ calcd for $C_{49}H_{43}ClN_5P_2Ru$, 900.1726; found, 900.1726.

5.3 Synthesis of **2**

The mixture of ligand **L2** (1 mmol) and $RuCl_3 \bullet 3H_2O$ (1 mmol) was heated in refluxed ethanol (60 mL) for 5h. The color of the solution changed from black-brown to red-brown slowly and further generated red-brown precipitates. After being cooled to room temperature, the precipitates were filtered, washed with EtOH and Et_2O, and dried under

vacuum. Then, Methanol (30 mL), TPP (2 mmol) and NaH (1mmol) were added to the filtered precipitate and refluxed for 6 h. After the reaction completed, the solvent was removed under vacuum to give a solid substance. The resultant solid was dissolved in dichloromethane followed by removal of the insoluble impurities by filtration to give a bright red solution. After the diethyl ether was added to the solution, the red solid complex **2** was recrystallized from the solution, in yields of 40%. ^{1}H NMR (400 MHz, Methylene Chloride-d_2) δ 7.95 (t, J = 7.9 Hz, 1H), 7.70 (d, J = 7.9 Hz, 2H), 7.22 (t, J = 7.4 Hz, 6H), 7.13 (d, J = 8.3 Hz, 4H), 7.00 (t, J = 7.4 Hz, 12H), 6.65 (dt, J = 8.6, 4.8 Hz, 12H), 6.58 (s, 2H), 6.28 (d, J = 8.2 Hz, 4H), 1.88 (s, 6H), -6.33 (t, J = 23.3 Hz, 1H). ^{31}P{^{1}H} NMR (162 MHz, DMSO-d6) δ 42.12. HRMS (ESI-TOF) m/z: $[M-Cl]^+$ calcd for $C_{63}H_{50}F_6N_5P_2Ru$, 1154.2484; found, 1154.2485.

5.4 Synthesis of **3**

The mixture of ligand **L3** (1 mmol) and $RuCl_3 \bullet 3H_2O$ (1 mmol) was heated in refluxed ethanol (60 mL) for 5h. The color of the solution changed from black-brown to red-brown slowly and further generated red-brown precipitates. After being cooled to room temperature, the precipitates were filtered, washed with EtOH and Et_2O, and dried under vacuum. Methanol (30 mL), TPP (2 mmol) and NaH (1mmol) were added to the filtered precipitate and refluxed for 6 h. At the end of the reaction, the solvent was removed under vacuum to give a solid substance. The resultant solid was dissolved in dichloromethane followed by removal of the insoluble impurities by filtration to give a red solution. After the diethyl ether was added to the solution, the orange red solid complex **3** was recrystallized from the solution, in yields of 45%. ^{1}H NMR (400 MHz, Methylene Chloride-d_2) δ 7.84 (t, J = 7.9 Hz, 1H), 7.56 (d, J = 7.8 Hz, 2H), 7.19 (t, J = 7.4 Hz, 6H), 6.97 (t, J = 7.5 Hz, 12H), 6.65 (dt, J = 8.6, 4.9 Hz, 12H), 6.39 (d, J = 0.9 Hz, 2H), 6.32 (d, J = 8.9 Hz, 4H), 6.00 (d, J = 8.9 Hz, 4H), 3.77 (s, 6H), 1.83 (s, 6H), -6.28 (t, J = 23.6 Hz, 1H). ^{31}P{^{1}H} NMR (162 MHz, Methylene Chloride-d_2) δ 42.03. HRMS (ESI-TOF) m/z: $[M-Cl]^+$ calcd for $C_{63}H_{56}N_5O_2P_2Ru$, 1078.2947; found, 1078.2947.

6. NMR spectra and HRMS data of complex 1, 2, and 3

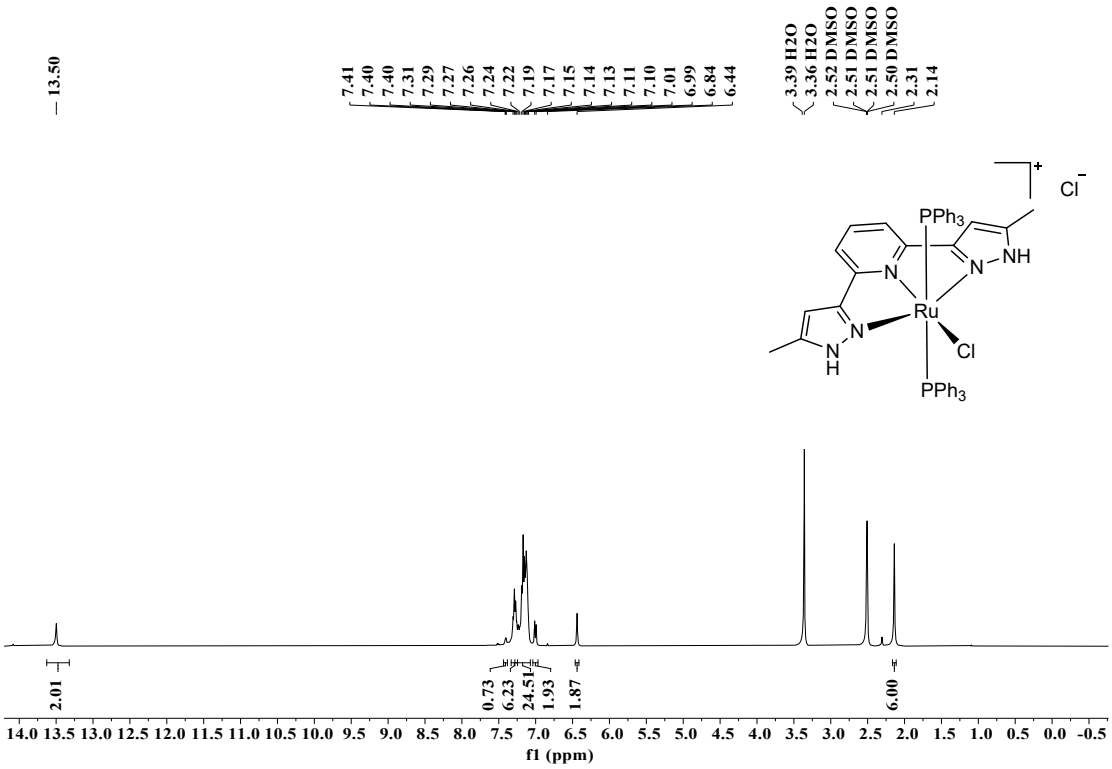


Figure S1: ^{1}H NMR spectrum of 1 (400.1 MHz, DMSO-d_6).

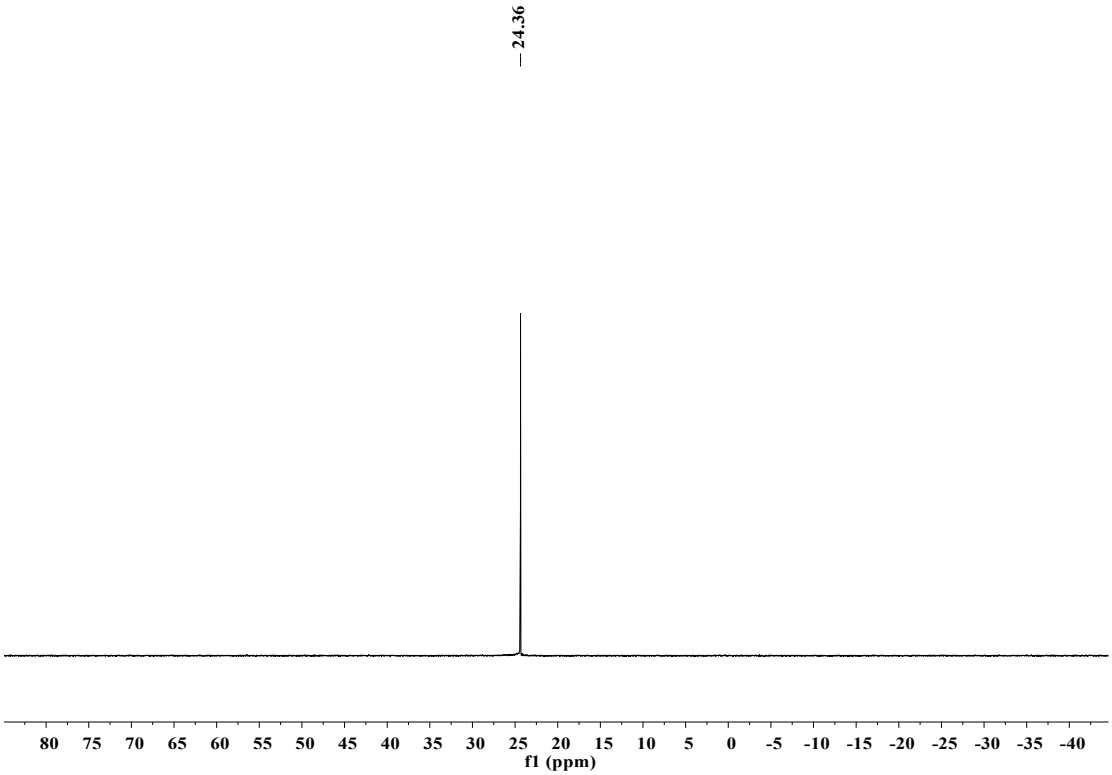


Figure S2: $^{31}P\{^{1}H\}$ NMR spectrum of 1 (162.0 MHz, DMSO-d_6).

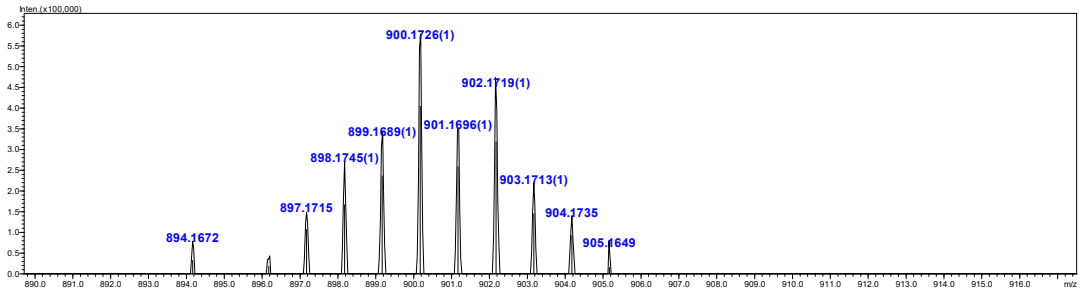


Figure S3: HRMS results of complex 1.

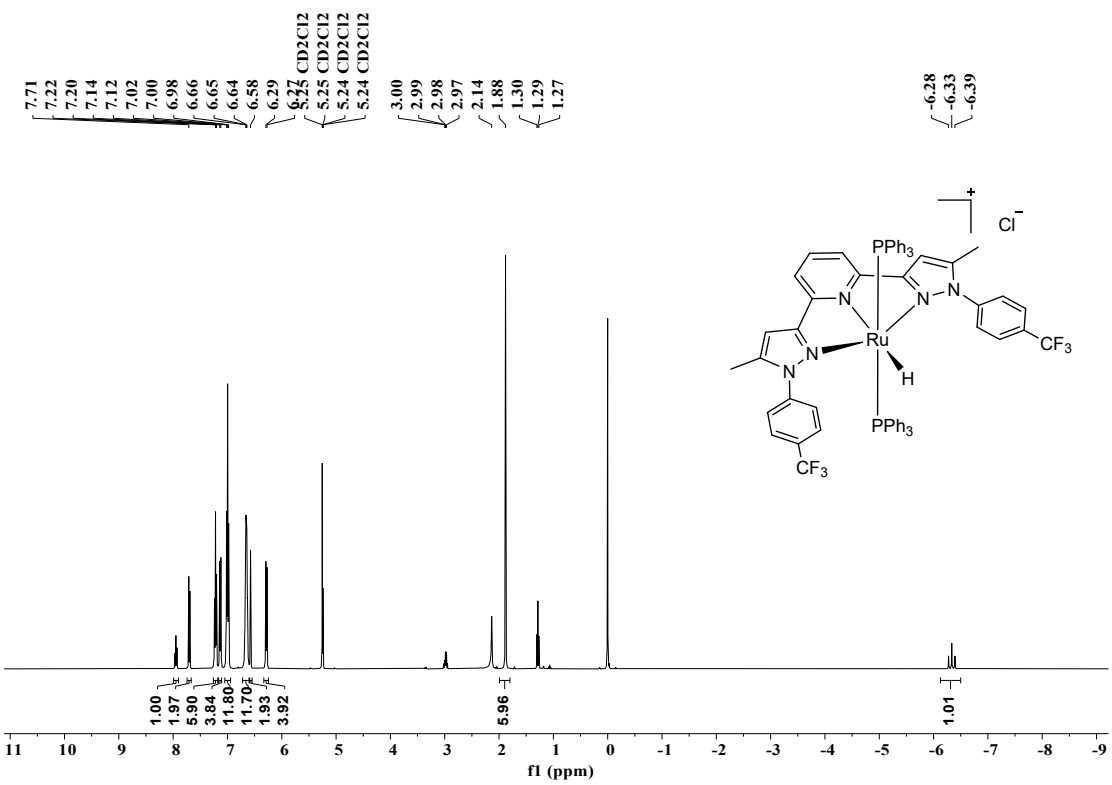


Figure S4: ^{1}H NMR spectrum of 2 (400 MHz, CD_2Cl_2).

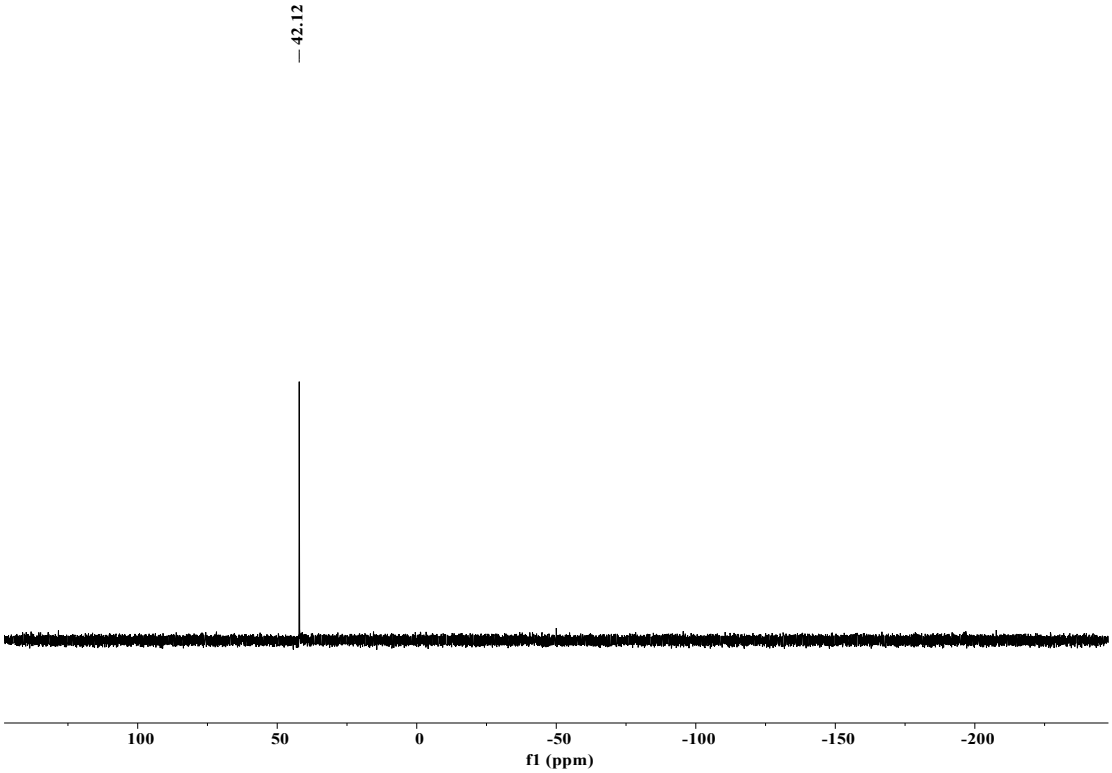


Figure S5: $^{31}P\{^{1}H\}$ NMR spectrum of 2 (162 MHz, DMSO).

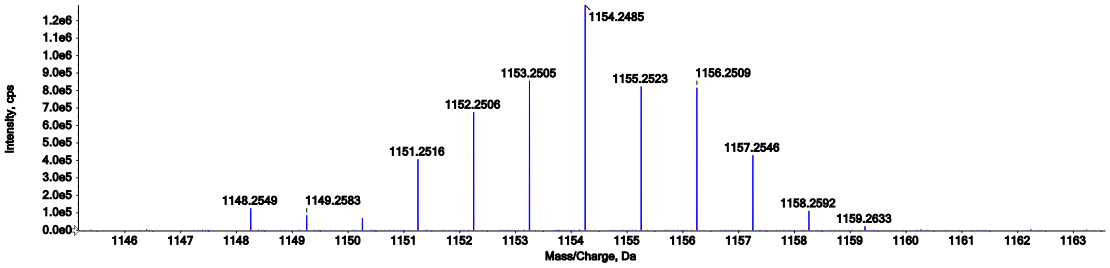


Figure S6: HRMS results of 2.

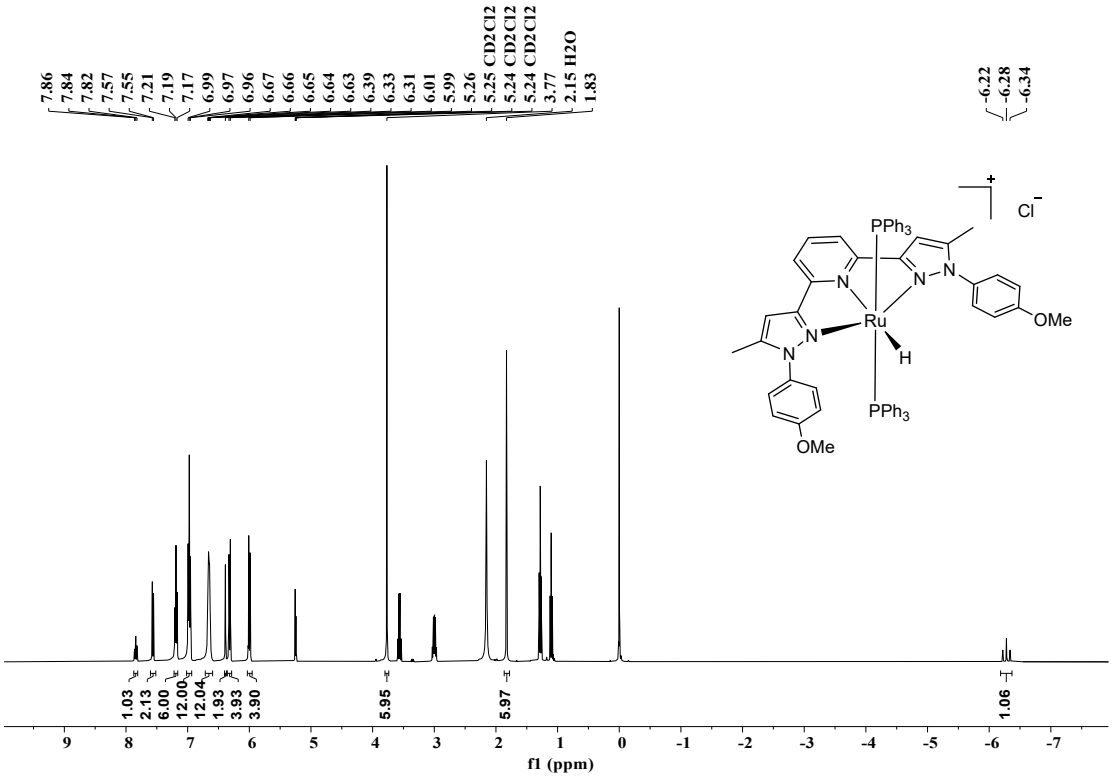


Figure S7: ^{1}H NMR spectrum of 3 (400 MHz, CD_2Cl_2).

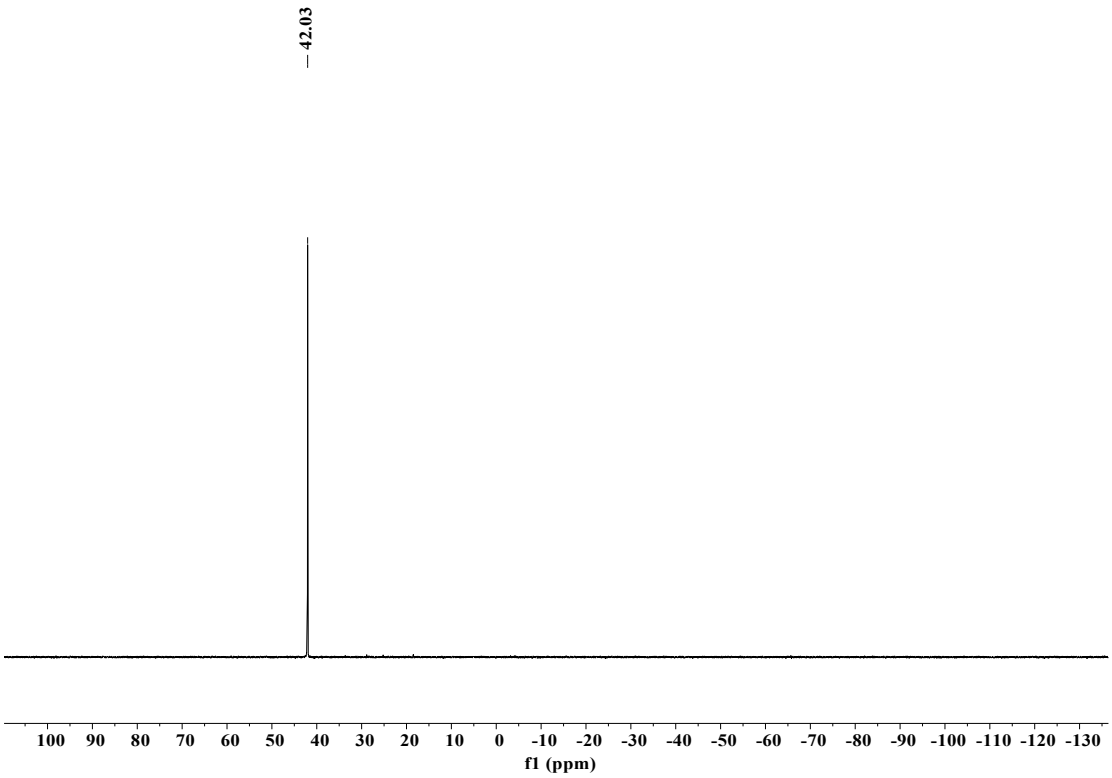


Figure S8: $^{31}P\{^{1}H\}$ NMR spectrum of 3 (162.0 MHz, CD_2Cl_2).

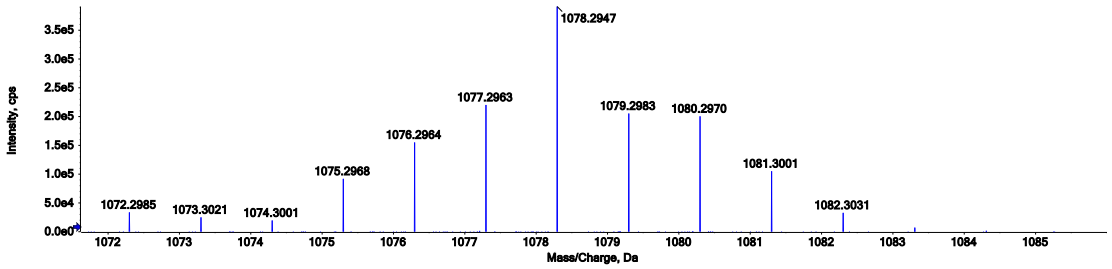
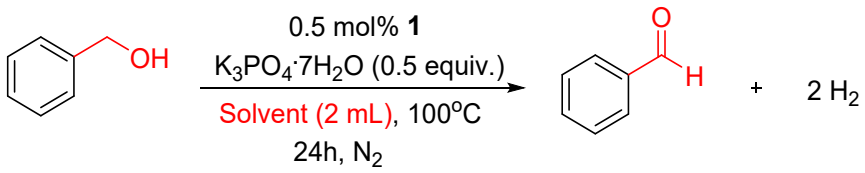


Figure S9: HRMS results of 3.

7. Optimization of reaction conditions for the AD of alcohols to aldehydes

Reaction procedure: Unless other conditions are specified, Ru catalyst, base were added into a dried Schlenk tube equipped with a balloon, and it was subject to three cycles of vacuum and refilled with nitrogen. Then solvent and alcohols were sequentially injected into it. The mixture was stirred at corresponding temperature. At the end of the reaction, the mixture was slowly cooled to room temperature and diluted with 3 mL of CH_2Cl_2.

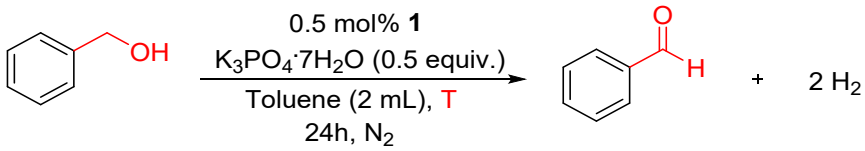
Table S1: Effect of reaction solvent on acceptorless dehydrogenative of primary alcohols [a].



entry	solvent	conv (%)	yield of aldehyde (%)
1	1,4-dioxane	3	3
2[b]	THF	9	9
3[b]	1,2-dimethoxyethane	5	5
4	DMF	19	19
5	DMA	37	35
6	Toluene	49	38

[a]Reaction Condition: 0.5 mol% **1**, benzyl alcohol (1.0 mmol, 104 µL), and $K_3PO_4·7H_2O$ (0.5 mmol) in solvent (2 ml) at 100°C for 24 h under the nitrogen atmosphere. [b]The reaction temperature is the boiling point of the solvent. All conversions and yields were determined by GC-MS.

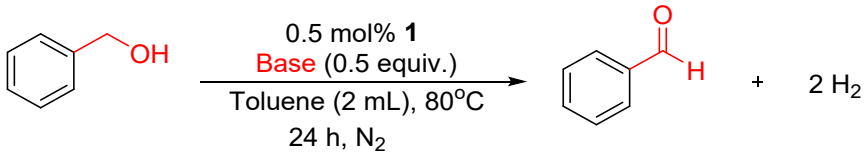
Table S2: Effect of reaction temperature on acceptorless dehydrogenative of primary alcohols [a].



entry	T (°C)	conv (%)	yield of aldehyde (%)
1	50	16	16
2	60	26	26
3	70	29	29
4	80	46	46
5	90	51	49
6	100	60	56
7	110	56	39
8	120	44	26

[a]Reaction Condition: 0.5 mol% **1**, benzyl alcohol (1.0 mmol, 104 µL), and $K_3PO_4·7H_2O$ (0.5 mmol) in toluene (2 ml) for 24 h under the nitrogen atmosphere. All conversions and yields were determined by GC-MS.

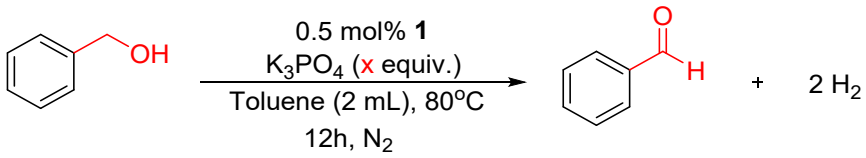
Table S3: Effect of bases on acceptorless dehydrogenative of primary alcohols [a].



entry	base	conv (%)	yield of aldehyde (%)	Selectivity(%)
1	KOH	50	9	18
2	K_2CO_3	27	27	100
3	KAc	22	22	100
4	$K_2HPO_4 \cdot 3H_2O$	16	16	100
5	$K_3PO_4 \cdot 7H_2O$	46	46	100
6	K_3PO_4	54	54	100
7	$K_4P_2O_7$	47	47	100
8	$(CH_3CH_2)_3N$（0.5）	3	3	100
9	Na_3PO_4	38	35	92
10	Li_3PO_4	7	7	100
11	Cs_2CO_3	53	8	15

[a]Reaction Condition: 0.5 mol% **1**, benzyl alcohol (1.0 mmol, 104 μL), and base (0.5 mmol) in toluene (2 ml) at 80°C for 24h under the nitrogen atmosphere. All conversions and yields were determined by GC-MS.

Table S4: Effect of base loadings on acceptorless dehydrogenative of primary alcohols [a].

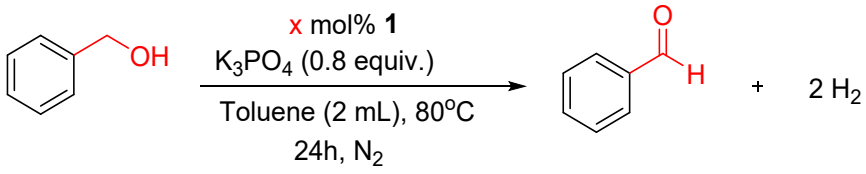


entry	base	quantity of base	conv (%)	yield of aldehyde (%)
1	K_3PO_4	None	2	2
2	K_3PO_4	0.5 equiv.	43	43
3	K_3PO_4	0.6 equiv.	35	35
4	K_3PO_4	0.7 equiv.	49	49
5	K_3PO_4	0.8 equiv.	56	56

6	K_3PO_4	1.0 equiv.	41	41

[a]Reaction Condition: 0.5 mol% **1**, benzyl alcohol (1.0 mmol, 104 µL), and K_3PO_4 (x equiv.) in toluene (2 ml) at 80°C for 20 h under the nitrogen atmosphere. All conversions and yields were determined by GC-MS.

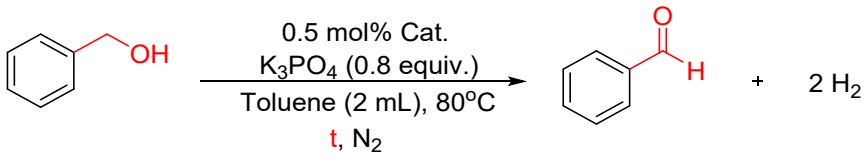
Table S5: Effect of the quantity of catalyst on acceptorless dehydrogenative of primary alcohols [a].



entry	quantity of catalyst (mol %)	conv (%)	yield of aldehyde (%)
1	None	trace	trace
2	0.3	43	43
3	0.5	62	62
4	1	76	51

[a]Reaction Condition: x mol% **1**, benzyl alcohol (1.0 mmol, 104 µL), and K_3PO_4 (0.8 mmol) in toluene (2 ml) at 80°C for 24 h under the nitrogen atmosphere. All conversions and yields were determined by GC-MS.

Table S6: Effect of Reaction time on acceptorless dehydrogenative of primary alcohols [a].

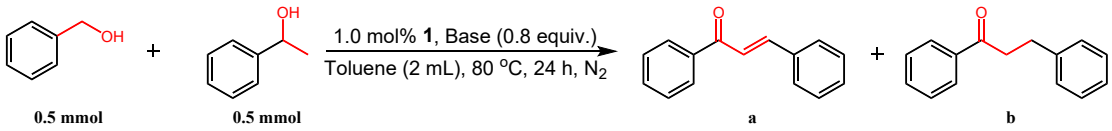


entry	Reaction time (h)	conv (%)	yield of aldehyde (%)
1	24	62	62
2	48	>99	>98
3	72	>99	>99

[a]Reaction Condition: 0.5 mol% **1**, benzyl alcohol (1.0 mmol, 104 µL), and K_3PO_4 (0.8 equiv.) in toluene (2 ml) at 80°C under the nitrogen atmosphere. All conversions and yields were determined by GC-MS.

8. Optimization of the conditions for sequential AD/condensation reactions between primary and secondary alcohols

Table S7: Effect of bases on the sequential AD/condensation reactions between primary and secondary alcohols.

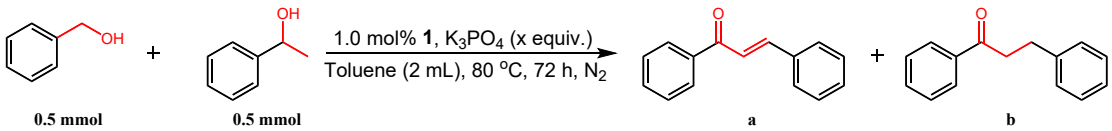


0.5 mmol 0.5 mmol a b

entry	base	conv (%)	yield of a (%)	yield of b (%)
1	KTB	90	8	83
2	K_3PO_4	34	29	5
3	KOH	87	5	82
4	NaOH	75	5	70
5	Cs_2CO_3	24	18	6

Reaction conditions: primary alcohol (0.5 mmol), secondary alcohols (0.5 mmol), Base (0.8 mmol), **1** (1.0 mol %), toluene (2 mL), reaction time (24 h), N_2 balloon, and isolated yield in the table. [a]The experimental data represent the yield of *α, β*-unsaturated ketones, [b]The experimental data represent the yield of saturated ketones.

Table S8: Effect of base loadings on the sequential AD/condensation reactions between primary and secondary alcohols.

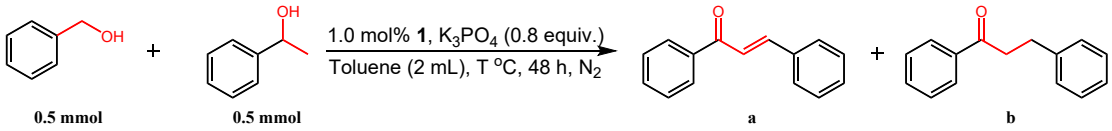


0.5 mmol 0.5 mmol a b

entry	quantity of base	conv (%)	yield of a (%)	yield of b (%)
1	0.2 equiv.	9	9	trace
2	0.4 equiv.	28	28	trace
3	0.6 equiv.	47	43	4
4	0.8 equiv.	75	70	5
5	1.0equiv.	53	46	7

Reaction conditions: primary alcohol (0.5 mmol), secondary alcohols (0.5 mmol), K_3PO_4 (x mmol), **1** (1.0 mol %), toluene (2 mL), reaction time (72 h), N_2 balloon, and isolated yield in the table. [a]The experimental data represent the yield of *α, β*-unsaturated ketones, [b]The experimental data represent the yield of saturated ketones.

Table S9: Effect of reaction temperature on the sequential AD/condensation reactions between primary and secondary alcohols.

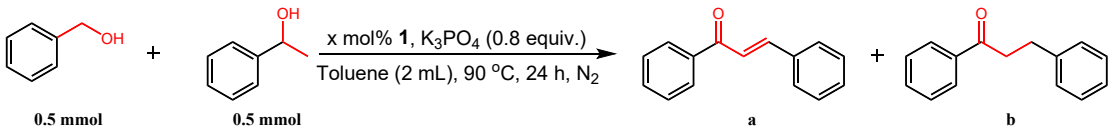


0.5 mmol 0.5 mmol a b

entry	T (°C)	conv (%)	yield of a (%)	yield of b (%)
1	70	36	31	5
2	80	61	56	5
3	90	72	64	8
4	100	77	60	17
5	120	>99	trace	>99

Reaction conditions: primary alcohol (0.5 mmol), secondary alcohols (0.5 mmol), K_3PO_4 (0.8 mmol), **1** (1.0 mol %), toluene (2 mL), reaction time (48 h), N_2 balloon, and isolated yield in the table. [a]The experimental data represent the yield of *α, β*-unsaturated ketones, [b]The experimental data represent the yield of saturated ketones.

Table S10: Effect of the quantity of catalyst on the sequential AD/condensation reactions between primary and secondary alcohols.



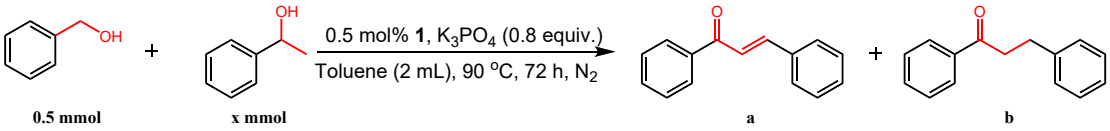
0.5 mmol 0.5 mmol a b

entry	quantity of catalyst (mol %)	conv (%)	yield of a (%)	yield of b (%)
1	0.2	21	19	2
2	0.5	47	45	2
3	1.0	41	36	5
4	1.5	38	19	9
5	2.0	39	34	5

Reaction conditions: primary alcohol (0.5 mmol), secondary alcohols (0.5 mmol), K_3PO_4 (0.8 mmol), **1** (x mol %), toluene (2 mL), reaction time (24 h), N_2 balloon, and isolated yield in the table. [a]The experimental data represent the yield of *α, β*-unsaturated ketones, [b]The experimental data represent the yield of saturated ketones.

Table S11: Effect of ratio of primary alcohol to secondary alcohol on the sequential

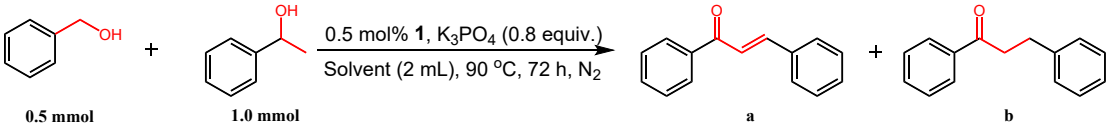
AD/condensation reactions between primary and secondary alcohols.



entry	ratio of primary alcohol to secondary alcohol	conv (%)	yield of a (%)	yield of b (%)
1	0.5:1	82	78	4
2	0.5:0.75	51	46	5
3	0.5:0.5	49	43	6

Reaction conditions: primary alcohol (0.5 mmol), secondary alcohols (x mmol), K_3PO_4 (0.8 mmol), **1** (0.5 mol %), toluene (2 mL), reaction time (72 h), N_2 balloon, and isolated yield in the table. [a]The experimental data represent the yield of *α, β*-unsaturated ketones, [b]The experimental data represent the yield of saturated ketones.

Table S12: Effect of reaction solvent on the sequential AD/condensation reactions between primary and secondary alcohols.



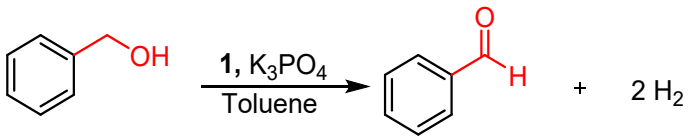
entry	Solvent (2 mL)	conv (%)	yield of a (%)	yield of b (%)
1	toluene	86	84	2
2	DMA	15	15	trace
3	DMF	16	16	trace
4	chlorobenzene	80	73	7
5	THF	52	32	20

Reaction conditions: primary alcohol (0.5 mmol), secondary alcohols (1.0 mmol), K_3PO_4 (0.8 mmol), **1** (0.5 mol %), solvent (2 mL), reaction time (72 h), N_2 balloon, and isolated yield in the table. [a]The experimental data represent the yield of *α, β*-unsaturated ketones, [b]The experimental data represent the yield of saturated ketones.

9. Mechanistic study

9.1 Control experiments[a]

Table S13: Data for control experiments[a]



entry	**1**	K_3PO_4	Toluene	conv. (%) [b]	Yield (%) [b]
1	√	√	√	>99%	98
2	-	-	√	NR[c]	NR[c]
3	√	-	√	2	2
4	-	√	√	4	4

[a]The reaction was performed in the presence of benzyl alcohol (1.0 mmol), **1** (0.5 mmol%), and K_3PO_4 (0.8 equiv.) in toluene (2 mL) at 80 °C for 48 h under N_2. [b]Conversions and yields were determined by GC-MS. [c]NR, no reaction.

9.2 NMR analysis

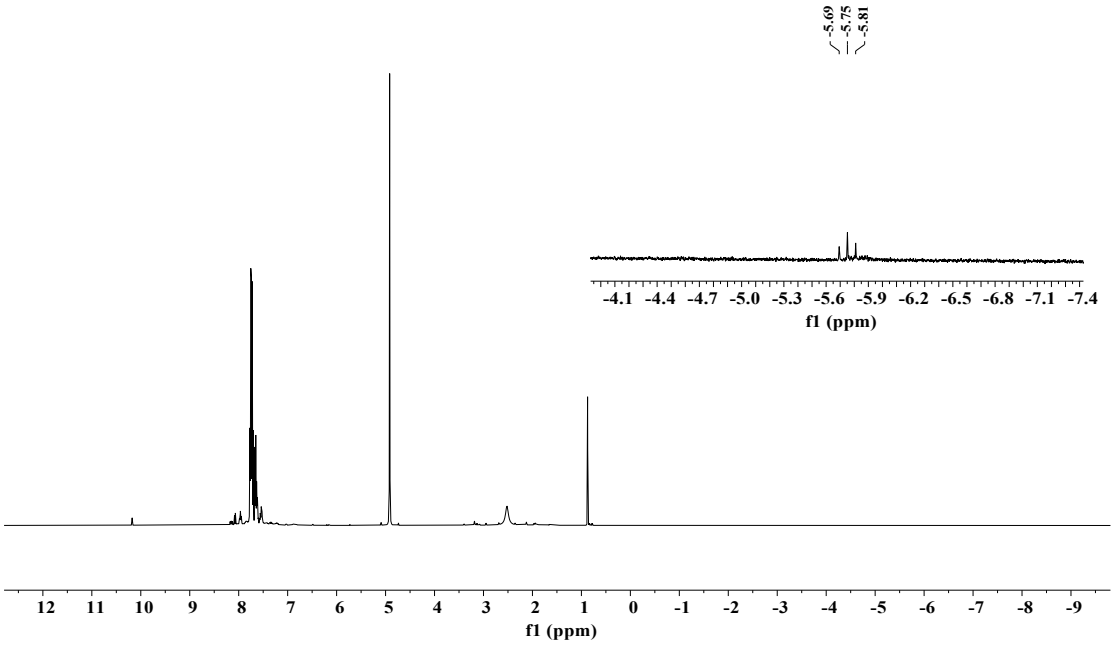


Figure S10: ^{1}H NMR evidence for the formation of Ru-H species in Complex 1 System

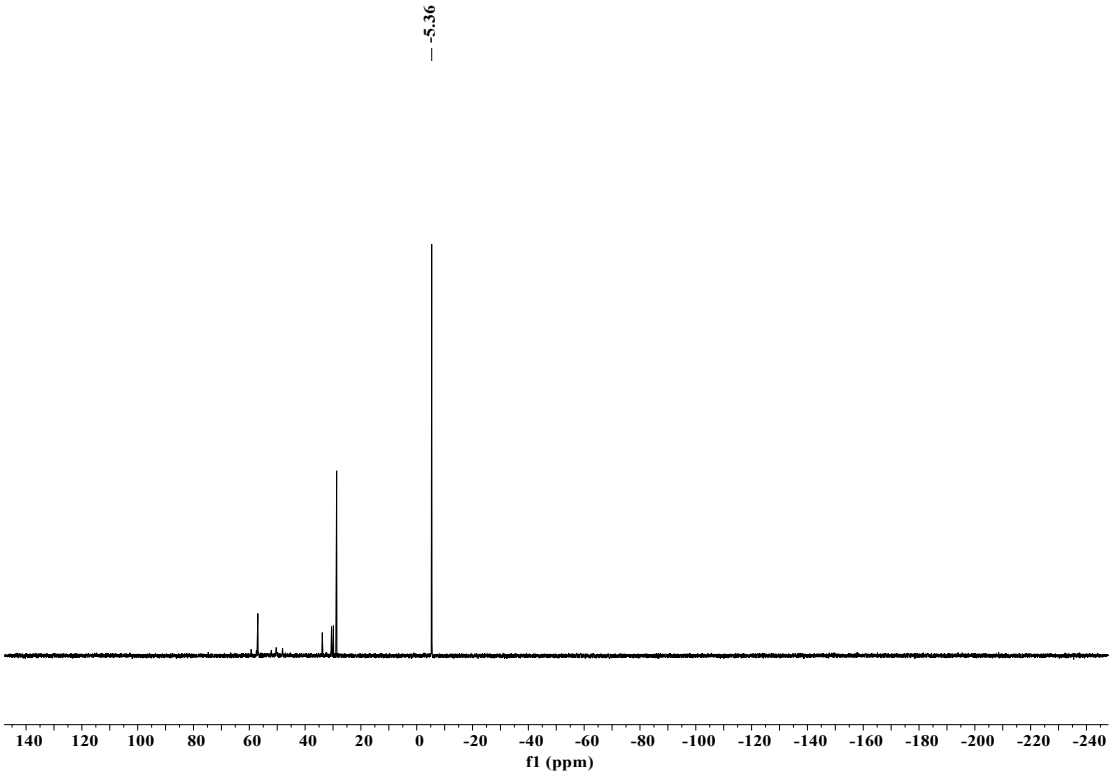


Figure S11: the observed dissociated PPh_3 ligand in $^{31}P\{^1H\}$ NMR spectra in Complex 1 System.

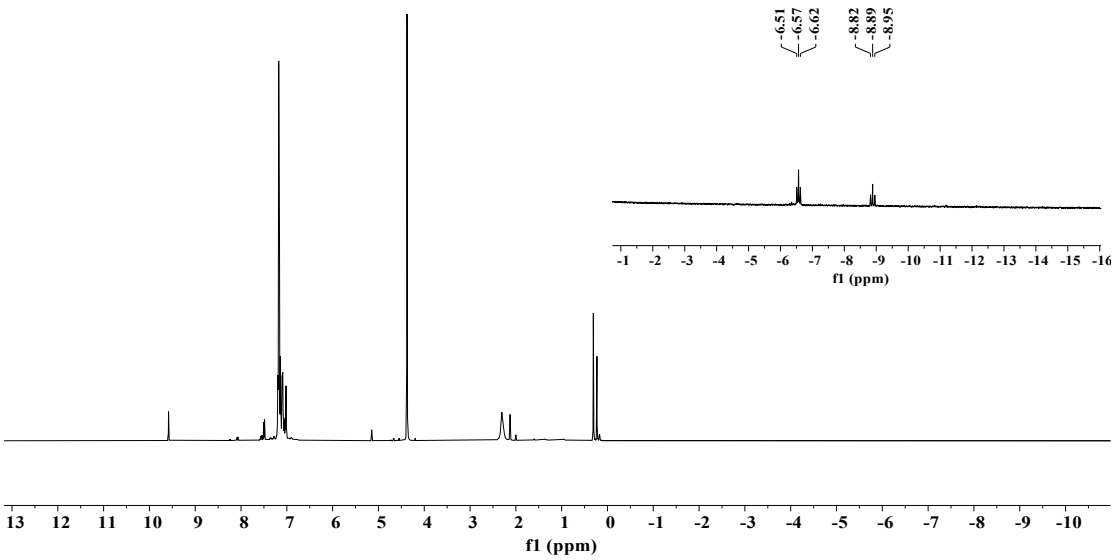


Figure S12: ^1H NMR evidence for the formation of Ru-H species in Complex 2 System

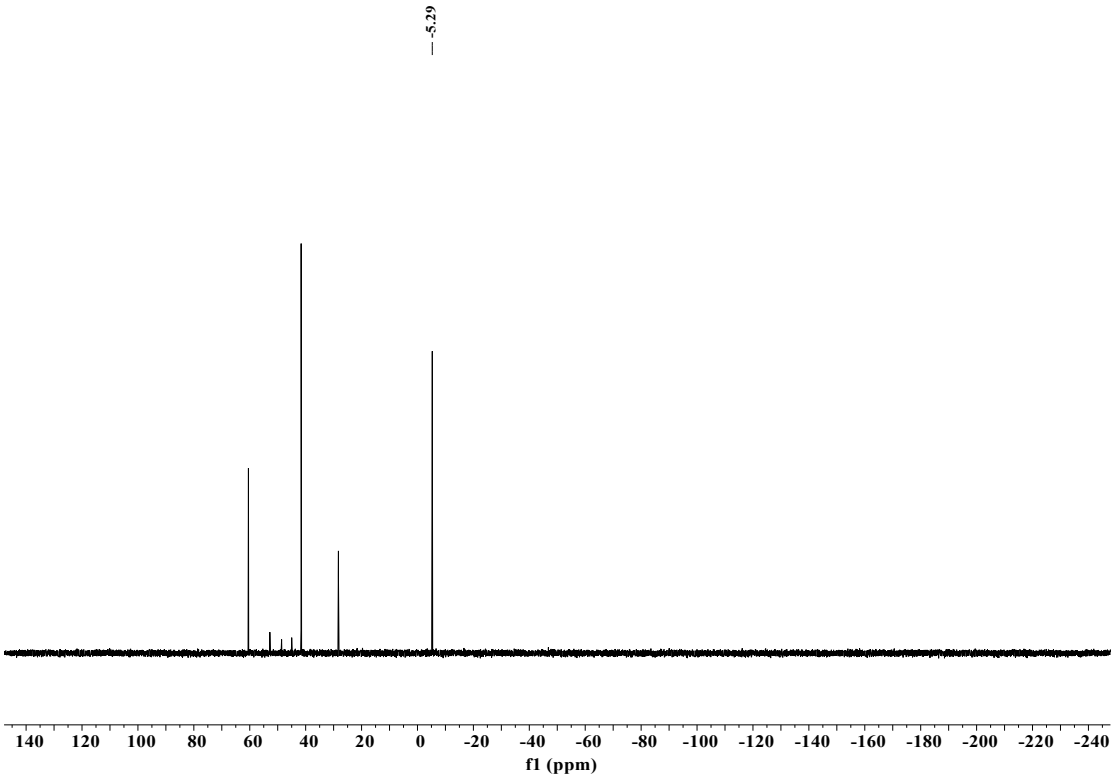


Figure S13: the observed dissociated PPh_3 ligand in $^{31}P\{^1H\}$ NMR spectra in Complex 2 System.

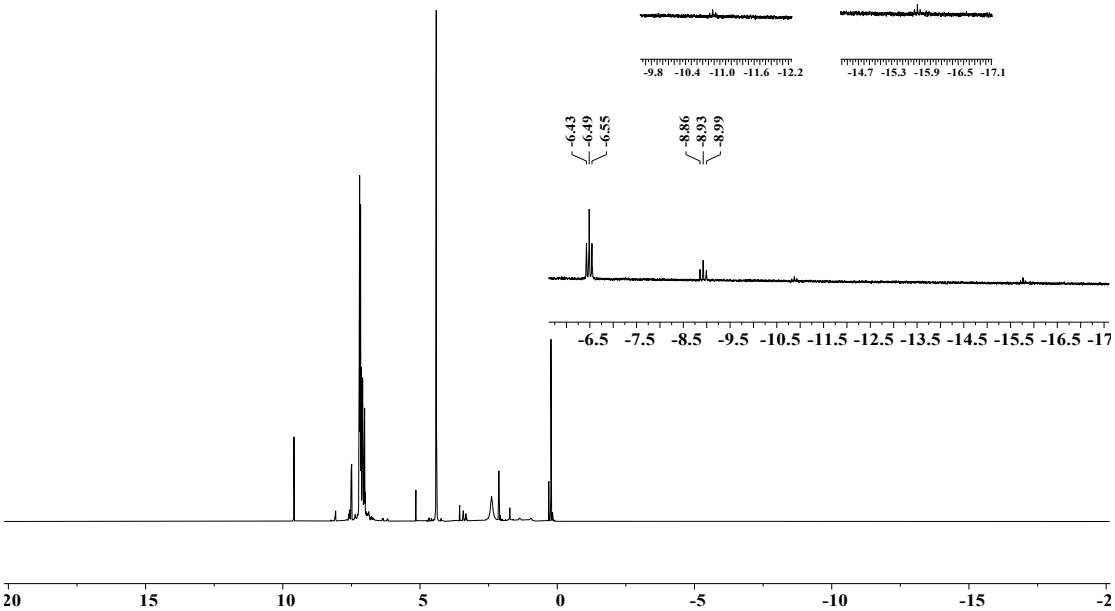


Figure S14: ^1H NMR evidence for the formation of Ru-H species in Complex 3 System

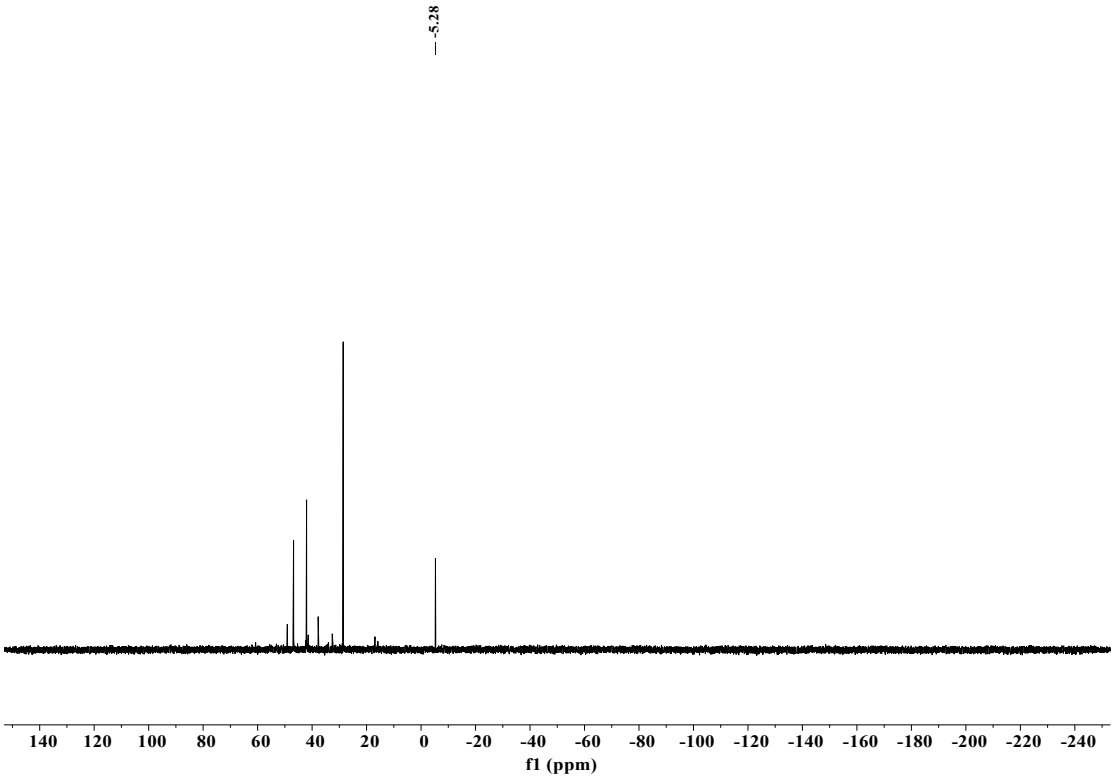
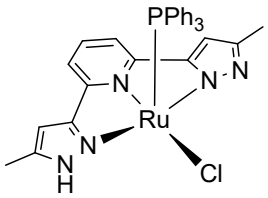
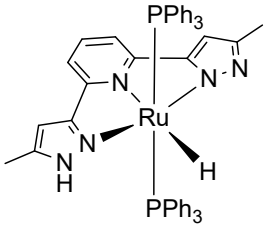


Figure S15: the observed dissociated PPh_3 ligand in $^{31}P\{^1H\}$ NMR spectra in Complex 3 System.

9.3 The key active species was detected by the HRMS

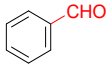


Exact Mass: $[M+H]^+$ = 638.0809
found: $[M+H]^+$ = 638.0818

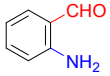


Exact Mass: $[M+H]^+$ = 866.2110
found: $[M+H]^+$ = 866.2086

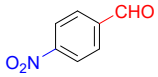
10. Characterization data of substrates



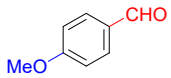
Benzaldehyde (**4a**).^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.05 (s, 1H), 7.95 – 7.86 (m, 2H), 7.71 – 7.63 (m, 1H), 7.56 (t, *J* = 7.5 Hz, 2H). $^{13}C\{^1H\}$ NMR (101 MHz, Chloroform-*d*) δ 192.38, 136.38, 134.45, 129.70, 128.98.



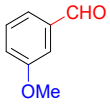
Anthranilaldehyde (**4b**)**.** ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.88 (s, 1H), 7.49 (dd, *J* = 7.8, 1.6 Hz, 1H), 7.32 (ddd, *J* = 8.5, 7.1, 1.7 Hz, 1H), 6.82 – 6.61 (m, 2H), 6.16 (s, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 194.12, 149.96, 135.75, 135.23, 118.83, 116.36, 116.05.



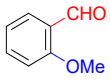
4-nitrobenzaldehyde (**4c**)**.** ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.17 (s, 1H), 8.39 (d, *J* = 8.4 Hz, 2H), 8.09 (d, *J* = 8.4 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.38, 151.09, 140.06, 130.49, 124.29.



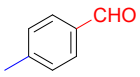
4-methoxybenzaldehyde (**4d**)**.** ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.85 (s, 1H), 7.80 (d, *J* = 8.8 Hz, 2H), 6.97 (d, *J* = 8.8 Hz, 2H), 3.85 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.75, 164.58, 131.93, 129.91, 114.29, 55.54.



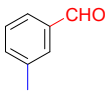
3-methoxybenzaldehyde (**4e**)**.** ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.95 (s, 1H), 7.50 – 7.33 (m, 3H), 7.15 (d, *J* = 6.7 Hz, 1H), 3.83 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 192.08, 160.12, 137.79, 130.01, 123.44, 121.42, 112.10, 55.41.



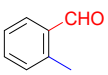
2-methoxybenzaldehyde (**4f**)**.** ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.47 (d, *J* = 0.8 Hz, 1H), 7.82 (dd, *J* = 7.7, 1.8 Hz, 1H), 7.55 (ddd, *J* = 8.3, 7.3, 1.8 Hz, 1H), 7.07 – 6.95 (m, 2H), 3.92 (d, *J* = 1.2 Hz, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 189.78, 161.83, 135.96, 128.47, 124.81, 120.63, 111.64, 55.62.



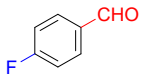
4-methylbenzaldehyde (**4g**)**.** ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.97 (s, 1H), 7.78 (d, *J* = 8.1 Hz, 2H), 7.33 (d, *J* = 7.9 Hz, 2H), 2.44 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 191.96, 145.53, 134.21, 129.83, 129.71, 21.87.



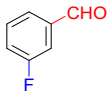
3-methylbenzaldehyde (**4h**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.96 (s, 1H), 7.78 – 7.56 (m, 2H), 7.40 (d, *J* = 7.3 Hz, 2H), 2.40 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 192.53, 138.87, 136.47, 135.26, 129.99, 128.86, 127.17, 21.13.



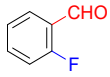
2-methylbenzaldehyde (**4i**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.26 (s, 1H), 7.79 (dd, *J* = 7.7, 1.5 Hz, 1H), 7.47 (td, *J* = 7.5, 1.6 Hz, 1H), 7.35 (td, *J* = 7.5, 1.2 Hz, 1H), 7.25 (d, *J* = 7.6 Hz, 1H), 2.66 (s, 3H).^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 192.81, 140.59, 134.14, 133.65, 132.05, 131.77, 126.32, 19.57.



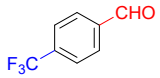
4-fluorobenzaldehyde (**4j**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.96 (s, 1H), 7.90 (dd, *J* = 8.8, 5.4 Hz, 2H), 7.20 (t, *J* = 8.6 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.45, 166.48 (d, *J* = 256.5 Hz), 132.97 (d, *J* = 2.7 Hz), 132.20 (d, *J* = 9.6 Hz), 116.31 (d, *J* = 22.4 Hz).



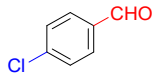
3-fluorobenzaldehyde (**4k**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.00 (d, *J* = 1.9 Hz, 1H), 7.68 (dt, *J* = 7.5, 1.2 Hz, 1H), 7.61 – 7.49 (m, 2H), 7.34 (tdd, *J* = 8.3, 2.7, 1.1 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.88 (d, *J* = 2.4 Hz), 163.08 (d, *J* = 249.4 Hz), 138.40 (d, *J* = 6.2 Hz), 130.79 (d, *J* = 7.7 Hz), 126.05 (d, *J* = 3.0 Hz), 121.56 (d, *J* = 21.7 Hz), 115.31 (d, *J* = 21.7 Hz).



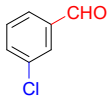
2-fluorobenzaldehyde (**4l**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.35 (d, *J* = 0.7 Hz, 1H), 7.86 (ddd, *J* = 7.6, 7.0, 1.9 Hz, 1H), 7.60 (dddd, *J* = 8.4, 7.3, 5.4, 1.9 Hz, 1H), 7.26 (tt, *J* = 7.5, 1.0 Hz, 1H), 7.16 (ddd, *J* = 10.5, 8.4, 1.0 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 187.15 (d, *J* = 6.6 Hz), 164.67 (d, *J* = 258.6 Hz), 136.34 (d, *J* = 9.1 Hz), 128.68 (d, *J* = 1.8 Hz), 124.63 (d, *J* = 3.7 Hz), 124.16 (d, *J* = 8.0 Hz), 116.49 (d, *J* = 20.5 Hz).



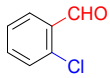
4-(trifluoromethyl)-benzaldehyde (**4m**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.13 (s, 1H), 8.07 – 8.01 (m, 2H), 7.84 (d, *J* = 8.0 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 191.07, 138.65, 135.54 (q, *J* = 32.7 Hz), 129.87, 126.06 (q, *J* = 3.8 Hz), 123.42 (q, *J* = 272.8 Hz).



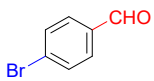
4-chlorobenzaldehyde (**4n**). ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.00 (s, 1H), 7.94 – 7.89 (m, 2H), 7.64 (dd, *J* = 8.5, 1.8 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 192.45, 139.84, 135.26, 131.57, 129.76.



3-chlorobenzaldehyde (**4o**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.96 (s, 1H), 7.82 (t, *J* = 1.8 Hz, 1H), 7.75 (dt, *J* = 7.6, 1.3 Hz, 1H), 7.57 (ddd, *J* = 8.0, 2.2, 1.2 Hz, 1H), 7.47 (t, *J* = 7.8 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.82, 137.78, 135.39, 134.35, 130.37, 129.20, 127.98.

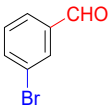


2-chlorobenzaldehyde (**4p**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.50 – 10.47 (m, 1H), 7.92 (dt, *J* = 7.8, 1.6 Hz, 1H), 7.56 – 7.50 (m, 1H), 7.45 (dt, *J* = 8.1, 1.4 Hz, 1H), 7.39 (td, *J* = 7.4, 1.2 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 189.79, 137.92, 135.13, 132.45, 130.60, 129.36, 127.29.

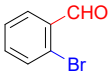


4-bromobenzaldehyde (**4q**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.99 (s, 1H), 7.76 (d, *J* = 8.5 Hz, 2H), 7.69 (d, *J* = 8.5 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 191.03, 135.07, 132.43,

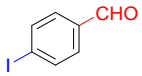
130.96, 129.75.



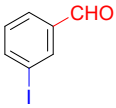
3-bromobenzaldehyde (**4r**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.94 (s, 1H), 7.97 (t, *J* = 1.8 Hz, 1H), 7.84 – 7.63 (m, 2H), 7.40 (t, *J* = 7.8 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.70, 137.96, 137.25, 132.25, 130.63, 128.39, 123.33.



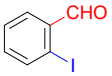
2-bromobenzaldehyde (**4s**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.39 (s, 1H), 7.96 – 7.90 (m, 1H), 7.70 – 7.64 (m, 1H), 7.51 – 7.41 (m, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 191.73 (d, *J* = 1.8 Hz), 135.32, 133.87, 133.46, 129.82, 127.90, 127.07.



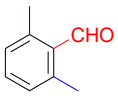
4-iodobenzaldehyde (**4t**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.97 (s, 1H), 7.92 (d, *J* = 8.4 Hz, 2H), 7.60 (d, *J* = 8.4 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 191.41, 138.41, 135.56, 130.81, 102.85.



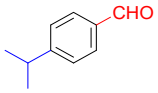
3-iodobenzaldehyde (**4u**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.93 (s, 1H), 8.22 (s, 1H), 7.96 (dt, *J* = 7.8, 1.4 Hz, 1H), 7.85 (dt, *J* = 7.6, 1.3 Hz, 1H), 7.30 (t, *J* = 7.7 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.70, 143.17, 138.41, 137.97, 130.73, 128.91, 94.68.



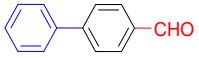
2-iodobenzaldehyde (**4v**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.07 (d, *J* = 0.8 Hz, 1H), 7.96 (dd, *J* = 7.9, 1.1 Hz, 1H), 7.88 (dd, *J* = 7.7, 1.8 Hz, 1H), 7.47 (tt, *J* = 7.6, 1.0 Hz, 1H), 7.29 (ddd, *J* = 7.9, 7.3, 1.8 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 195.74, 140.65, 135.48, 135.11, 130.26, 128.73, 100.73.



2,6-Dimethylbenzaldehyde (**4w**).^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.64 (s, 1H), 7.34 (t, *J* = 7.6 Hz, 1H), 7.11 (d, *J* = 7.6 Hz, 2H), 2.63 (s, 6H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 193.55, 141.14, 132.98, 132.44, 129.71, 20.50.

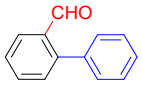


4-isopropylbenzaldehyde (**4x**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.98 (s, 0H), 7.86 – 7.79 (m, 0H), 7.42 – 7.37 (m, 0H), 3.00 (p, *J* = 6.9 Hz, 0H), 1.29 (d, *J* = 6.9 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 192.00, 156.22, 134.55, 130.00, 127.14, 34.47, 23.63.

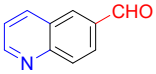


Biphenyl-4-carboxaldehyde (**4y**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.08 (s, 1H), 8.00 – 7.94

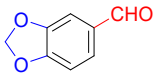
(m, 2H), 7.80 – 7.75 (m, 2H), 7.69 – 7.63 (m, 2H), 7.54 – 7.42 (m, 3H). $^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 191.93, 147.17, 139.71, 135.22, 130.29, 129.05, 128.51, 127.69, 127.39.



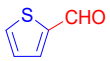
Biphenyl-2-carboxaldehyde (**4z**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.02 (d, *J* = 0.8 Hz, 1H), 8.07 (dd, *J* = 7.8, 1.4 Hz, 1H), 7.66 (td, *J* = 7.5, 1.5 Hz, 1H), 7.58 – 7.36 (m, 8H). $^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 192.42, 145.98, 137.77, 133.74, 133.58, 130.80, 130.12, 128.45, 128.14, 127.80, 127.58.



Quinoline-6-formaldehyde (**4aa**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.17 (s, 1H), 9.02 (dd, *J* = 4.2, 1.7 Hz, 1H), 8.35 – 8.26 (m, 2H), 8.23 – 8.12 (m, 2H), 7.50 (dd, *J* = 8.3, 4.3 Hz, 1H). $^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 191.47, 153.06, 150.79, 137.49, 134.29, 133.64, 130.72, 127.68, 126.69, 122.21.

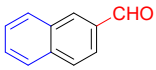


Piperonal (**4ab**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.78 (s, 1H), 7.38 (dd, *J* = 7.9, 1.6 Hz, 1H), 7.29 (d, *J* = 1.7 Hz, 1H), 6.90 (d, *J* = 7.9 Hz, 1H), 6.05 (s, 2H). $^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 190.25, 153.08, 148.68, 131.83, 128.64, 108.31, 106.80, 102.12.

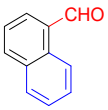


Thiophene-2-Carboxaldehyde (**4ac**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.95 (d, *J* = 1.3 Hz, 1H), 7.79 (ddd, *J* = 8.0, 4.3, 1.2 Hz, 2H), 7.23 (ddd, *J* = 4.8, 3.7, 1.1 Hz, 1H). $^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 183.05, 143.99, 136.44, 135.17, 128.38.

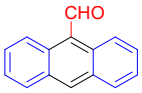
Thiophene-3-Carboxaldehyde (**4ad**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.92 (d, *J* = 0.8 Hz, 1H), 8.12 (dd, *J* = 2.9, 1.2 Hz, 1H), 7.54 (dd, *J* = 5.1, 1.1 Hz, 1H), 7.37 (ddd, *J* = 5.1, 2.9, 0.8 Hz, 1H). $^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 184.98, 143.02, 136.79, 127.43, 125.32.



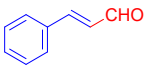
2-Naphthaldehyde (**4ae**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.17 (d, *J* = 0.6 Hz, 1H), 8.34 (d, *J* = 1.5 Hz, 1H), 8.12 – 7.85 (m, 4H), 7.63 (dddd, *J* = 22.8, 8.2, 6.9, 1.3 Hz, 2H).$^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 192.25, 136.45, 134.56, 134.12, 132.65, 129.54, 129.12 (d, *J* = 2.8 Hz), 128.09, 127.10, 122.76.



1-Naphthaldehyde (**4af**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 10.39 (s, 1H), 9.28 (dd, *J* = 8.6, 1.1 Hz, 1H), 8.08 (dt, *J* = 8.4, 1.0 Hz, 1H), 7.98 – 7.89 (m, 2H), 7.70 (ddd, *J* = 8.4, 6.8, 1.4 Hz, 1H), 7.60 (tdd, *J* = 6.8, 3.1, 1.0 Hz, 2H).$^{13}C\{^{1}H\}$ NMR (101 MHz, Chloroform-*d*) δ 193.55, 136.69, 135.30, 133.73, 131.41, 130.54, 129.08, 128.50, 126.98, 124.89.

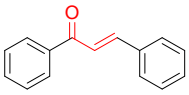


9-Anthracenecarboxaldehyde (**4ag**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 11.50 (s, 1H), 8.97 (dd, *J* = 9.0, 1.0 Hz, 2H), 8.64 (s, 1H), 8.04 (dd, *J* = 8.5, 1.3 Hz, 2H), 7.68 (ddd, *J* = 9.0, 6.6, 1.4 Hz, 2H), 7.55 (ddd, *J* = 7.9, 6.6, 1.0 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 192.96, 135.20, 132.09, 131.03, 129.27, 129.10, 125.66, 124.65, 123.52.

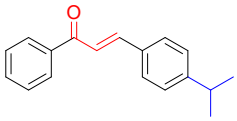


Cinnamaldehyde (**4ah**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 9.66 (d, *J* = 7.7 Hz, 1H), 7.55 – 7.48 (m, 2H), 7.46 – 7.34 (m, 4H), 6.67 (dd, *J* = 16.0, 7.7 Hz, 1H).^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 193.69, 152.79, 134.01, 131.29, 129.12, 128.54 (d, *J* = 6.0 Hz).

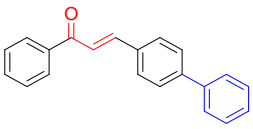
Cyclopropanecarboxaldehyde (**4ai**). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.88 (d, *J* = 5.8 Hz, 1H), 1.81 (d, *J* = 5.8 Hz, 0H), 1.10 – 1.03 (m, 4H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 201.52, 22.65, 7.25.



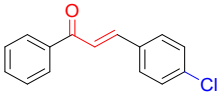
(E)-chalcone (**5c**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.09 – 8.02 (m, 2H), 7.85 (d, *J* = 15.7 Hz, 1H), 7.70 – 7.41 (m, 9H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.58, 144.87, 138.25, 134.92, 132.81, 130.57, 128.99, 128.66, 128.54, 128.48, 122.14.



(E)-3-(4-isopropylphenyl)-1-phenylprop-2-en-1-one (**5d**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.05 (dt, *J* = 7.8, 1.2 Hz, 2H), 7.85 (d, *J* = 15.6 Hz, 1H), 7.66 – 7.45 (m, 6H), 7.31 (d, *J* = 8.2 Hz, 2H), 2.97 (p, *J* = 6.9 Hz, 1H), 1.30 (d, *J* = 6.9 Hz, 6H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.68, 152.00, 144.99, 138.40, 132.69, 132.56, 128.65, 128.62, 128.50, 127.12, 121.22, 34.17, 23.80.

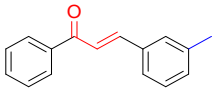


(E)-3-([1,1'-biphenyl]-4-yl)-1-phenylprop-2-en-1-one (**5e**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.13 – 8.03 (m, 2H), 7.89 (d, *J* = 15.7 Hz, 1H), 7.79 – 7.38 (m, 13H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.51, 144.42, 143.33, 140.14, 138.29, 133.87, 132.81, 129.01, 128.95, 128.66, 128.54, 127.93, 127.62, 127.08, 121.92.

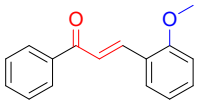


(E)-3-(4-chlorophenyl)-1-phenylprop-2-en-1-one (**5f**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.06 – 8.01 (m, 2H), 7.78 (d, *J* = 15.7 Hz, 1H), 7.67 – 7.47 (m, 6H), 7.41 (d, *J* = 8.5 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.23, 143.31, 138.04, 136.44, 133.39, 132.94, 129.60, 129.26,

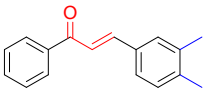
128.69, 128.51, 122.48.



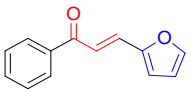
(E)-1-phenyl-3-(m-tolyl) prop-2-en-1-one (**5g**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.09 – 8.00 (m, 2H), 7.82 (d, *J* = 15.7 Hz, 1H), 7.65 – 7.44 (m, 6H), 7.30 (dt, *J* = 28.7, 7.7 Hz, 2H), 2.43 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.60, 145.08, 138.64, 138.28, 134.85, 132.76, 131.44, 129.08, 128.87, 128.63, 128.52, 125.74, 121.90, 21.38.



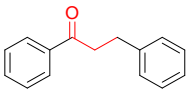
(E)-3-(2-methoxyphenyl)-1-phenylprop-2-en-1-one (**5h**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.09 – 7.99 (m, 2H), 7.80 (d, *J* = 15.7 Hz, 1H), 7.63 – 7.57 (m, 1H), 7.56 – 7.48 (m, 3H), 7.35 (t, *J* = 7.9 Hz, 1H), 7.26 (dt, *J* = 7.6, 1.2 Hz, 1H), 7.18 (t, *J* = 2.1 Hz, 1H), 6.98 (ddd, *J* = 8.2, 2.7, 1.0 Hz, 1H), 3.86 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.51, 159.97, 144.76, 138.18, 136.27, 132.83, 129.97, 128.65, 128.53, 122.38, 121.12, 116.33, 113.48, 55.36.



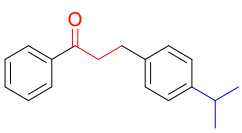
(E)-3-(3,4-dimethylphenyl)-1-phenylprop-2-en-1-one (**5i**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.05 (dd, *J* = 7.1, 1.8 Hz, 2H), 7.81 (d, *J* = 15.7 Hz, 1H), 7.65 – 7.37 (m, 6H), 7.21 (d, *J* = 7.7 Hz, 1H), 2.33 (d, *J* = 2.6 Hz, 6H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 190.68, 145.20, 139.91, 138.43, 137.24, 132.64, 132.57, 130.28, 129.67, 128.59, 128.49, 126.20, 120.94, 19.91, 19.79.



(E)-3-(furan-2-yl)-1-phenylprop-2-en-1-one (**5j**[a]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.11 – 8.00 (m, 2H), 7.71 – 7.44 (m, 6H), 6.74 (d, *J* = 3.5 Hz, 1H), 6.54 (dd, *J* = 3.5, 1.8 Hz, 1H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 189.83, 151.68, 144.93, 138.16, 132.77, 130.68, 128.62, 128.44, 119.33, 116.24, 112.69.

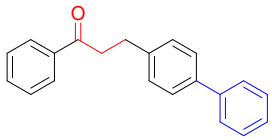


1,3-diphenylpropan-1-one (**5c**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.05 – 7.99 (m, 2H), 7.66 – 7.55 (m, 1H), 7.50 (dd, *J* = 8.4, 7.0 Hz, 2H), 7.42 – 7.22 (m, 5H), 3.35 (dd, *J* = 8.5, 6.9 Hz, 2H), 3.13 (dd, *J* = 8.5, 6.9 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 199.25, 141.36, 136.89, 133.14, 128.67, 128.60, 128.51, 128.11, 126.21, 40.50, 30.17.

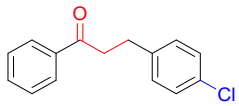


3-(4-isopropylphenyl)-1-phenylpropan-1-one (**5d**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.00 (dd, *J* = 8.4, 1.4 Hz, 2H), 7.70 – 7.40 (m, 3H), 7.22 (d, *J* = 2.4 Hz, 4H), 3.33 (dd, *J* = 8.6, 6.9 Hz, 2H), 3.08 (dd, *J* = 8.5, 6.9 Hz, 2H), 2.99 – 2.79 (m, 1H), 1.28 (d, *J* = 6.9 Hz, 6H). ^{13}C{^{1}H} NMR (101

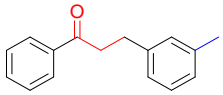
MHz, Chloroform-*d*) δ 199.40, 146.73, 138.60, 136.92, 133.05, 128.62, 128.37, 128.08, 126.59, 40.57, 33.74, 29.74, 24.08.



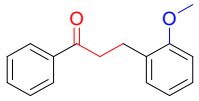
3-([1,1'-biphenyl]-4-yl)-1-phenylpropan-1-one (**5e**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.10 – 8.03 (m, 2H), 7.70 – 7.58 (m, 5H), 7.52 (td, *J* = 7.9, 5.2 Hz, 4H), 7.45 – 7.39 (m, 3H), 3.41 (dd, *J* = 8.4, 6.8 Hz, 2H), 3.21 (dd, *J* = 8.4, 6.9 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 199.19, 141.06, 140.53, 139.19, 136.95, 133.18, 129.00, 128.87, 128.72, 128.16, 127.35, 127.23, 127.10, 40.42, 29.82.



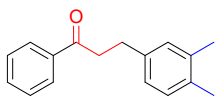
3-(4-chlorophenyl)-1-phenylpropan-1-one (**5f**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 7.98 (dd, *J* = 8.4, 1.4 Hz, 2H), 7.63 – 7.53 (m, 1H), 7.51 – 7.43 (m, 2H), 7.28 (d, *J* = 8.5 Hz, 2H), 7.21 (d, *J* = 8.5 Hz, 2H), 3.30 (dd, *J* = 8.1, 6.9 Hz, 2H), 3.07 (t, *J* = 7.5 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 198.83, 139.82, 136.75, 133.22, 131.86, 129.91, 128.69, 128.62, 128.05, 40.14, 29.38.



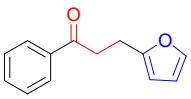
1-phenyl-3-(m-tolyl) propan-1-one (**5g**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.10 – 7.94 (m, 2H), 7.65 – 7.57 (m, 1H), 7.55 – 7.47 (m, 2H), 7.26 (t, *J* = 7.5 Hz, 1H), 7.17 – 7.05 (m, 3H), 3.35 (dd, *J* = 8.7, 6.8 Hz, 2H), 3.10 (dd, *J* = 8.6, 6.9 Hz, 2H), 2.41 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 199.32, 141.31, 138.16, 136.91, 133.12, 129.32, 128.67, 128.52, 128.11, 126.96, 125.48, 40.59, 30.11, 21.49.



3-(2-methoxyphenyl)-1-phenylpropan-1-one (**5h**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.08 – 7.98 (m, 2H), 7.62 – 7.55 (m, 1H), 7.53 – 7.44 (m, 2H), 7.26 (ddd, *J* = 8.9, 4.3, 1.9 Hz, 2H), 7.00 – 6.87 (m, 2H), 3.88 (s, 3H), 3.37 – 3.30 (m, 2H), 3.12 (dd, *J* = 8.7, 6.7 Hz, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 200.00, 157.57, 137.00, 132.97, 130.22, 129.55, 128.59, 128.17, 127.59, 120.59, 110.29, 55.23, 38.98, 25.78.



3-(3,4-dimethylphenyl)-1-phenylpropan-1-one (**5i**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.06 – 8.00 (m, 2H), 7.65 – 7.57 (m, 1H), 7.51 (dd, *J* = 8.4, 6.9 Hz, 2H), 7.17 – 7.01 (m, 3H), 3.34 (dd, *J* = 8.7, 6.9 Hz, 2H), 3.07 (dd, *J* = 8.6, 6.9 Hz, 2H), 2.31 (d, *J* = 6.2 Hz, 6H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 199.42, 138.76, 136.93, 136.70, 134.33, 133.08, 129.85, 128.65, 128.11, 125.79, 40.77, 29.74, 19.84, 19.40.



3-(furan-2-yl)-1-phenylpropan-1-one (**5j**[b]). ^{1}H NMR (400 MHz, Chloroform-*d*) δ 8.00 (dd, *J* = 8.3, 1.4 Hz, 2H), 7.63 – 7.54 (m, 1H), 7.52 – 7.45 (m, 2H), 7.34 (dd, *J* = 1.9, 0.9 Hz, 1H), 6.31 (dd, *J* = 3.2, 1.9 Hz, 1H), 6.08 (dd, *J* = 3.2, 0.9 Hz, 1H), 3.36 (dd, *J* = 8.3, 6.7 Hz, 2H), 3.17 – 3.08 (m, 2H). ^{13}C{^{1}H} NMR (101 MHz, Chloroform-*d*) δ 198.65, 154.79, 141.12, 136.76, 133.16, 128.64, 128.05, 110.28, 105.34, 36.94, 22.52.

11. NMR spectra of substrates

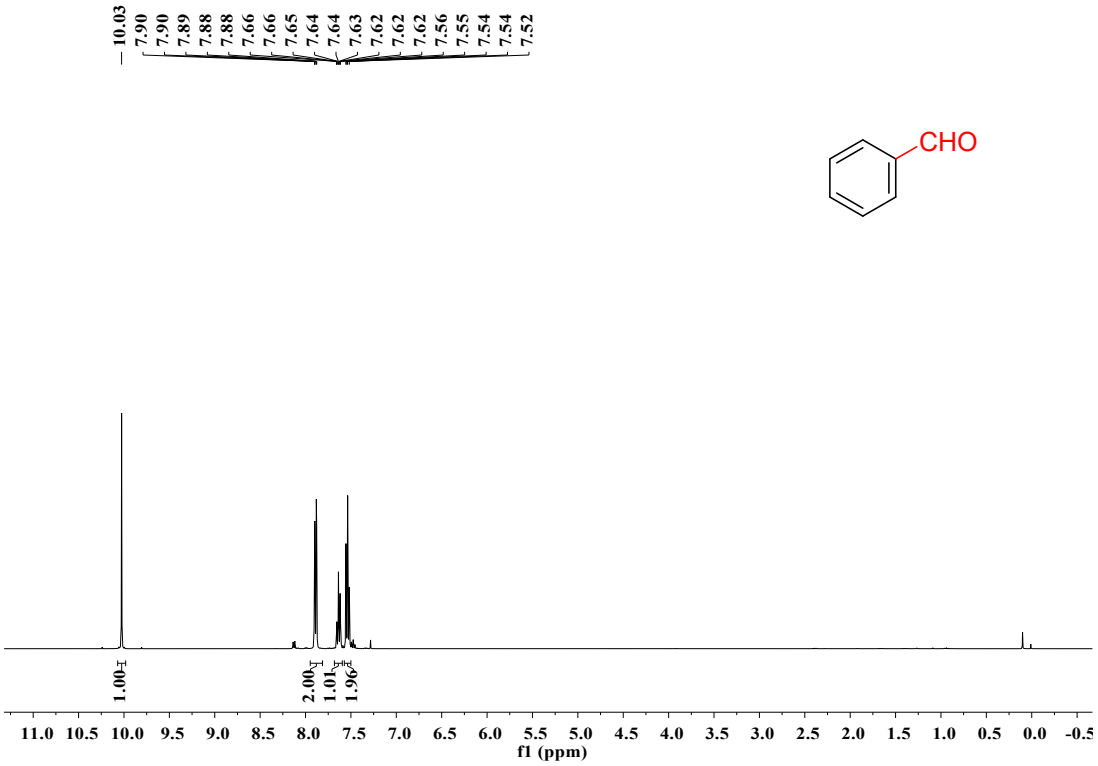


Figure S16: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of benzaldehyde (4a).

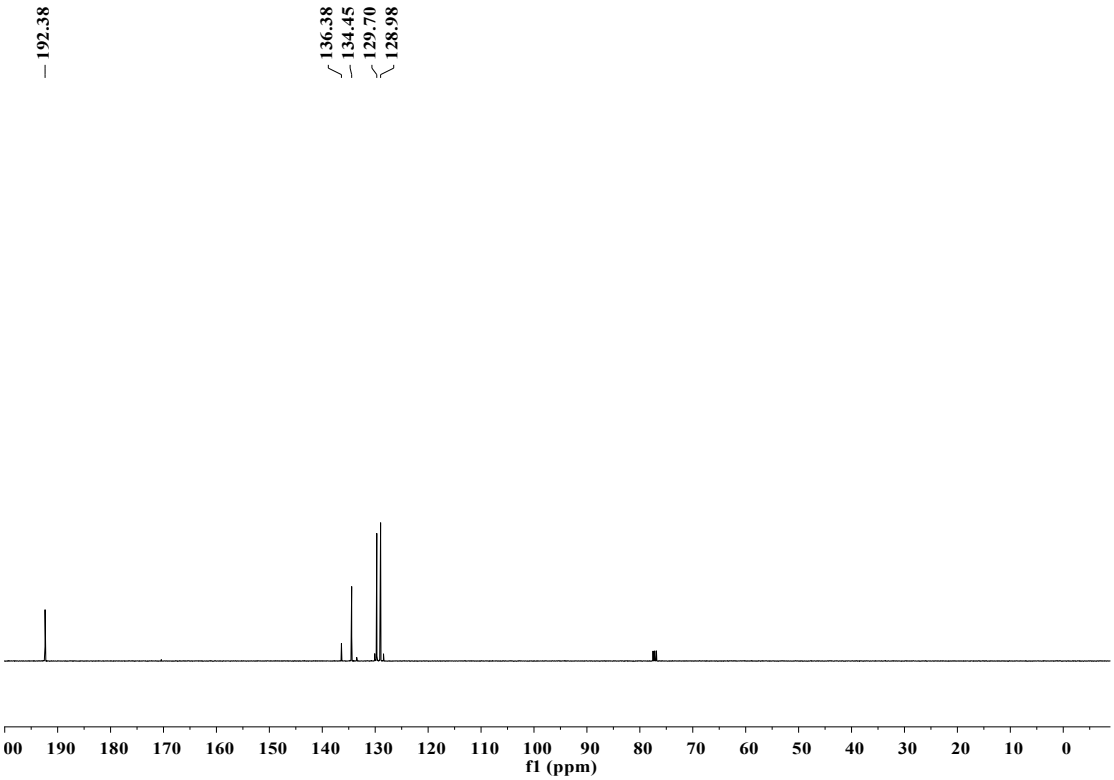


Figure S17: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of benzaldehyde (4a).

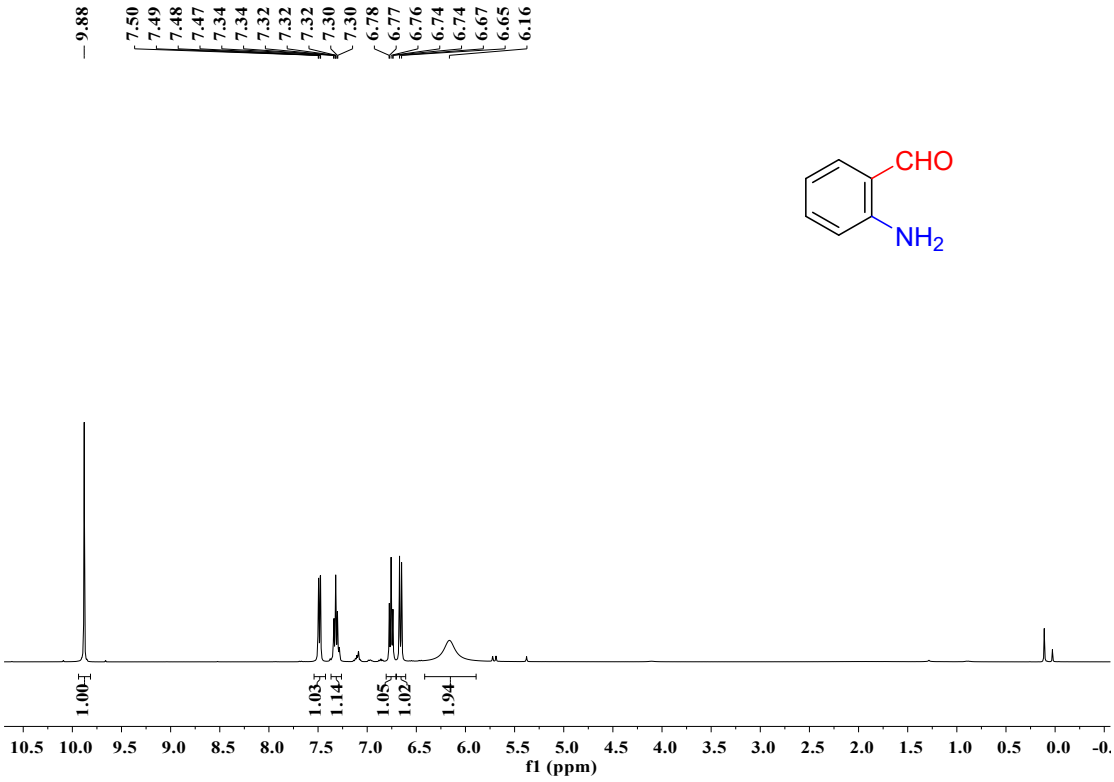


Figure S18: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of anthranilaldehyde (4b).

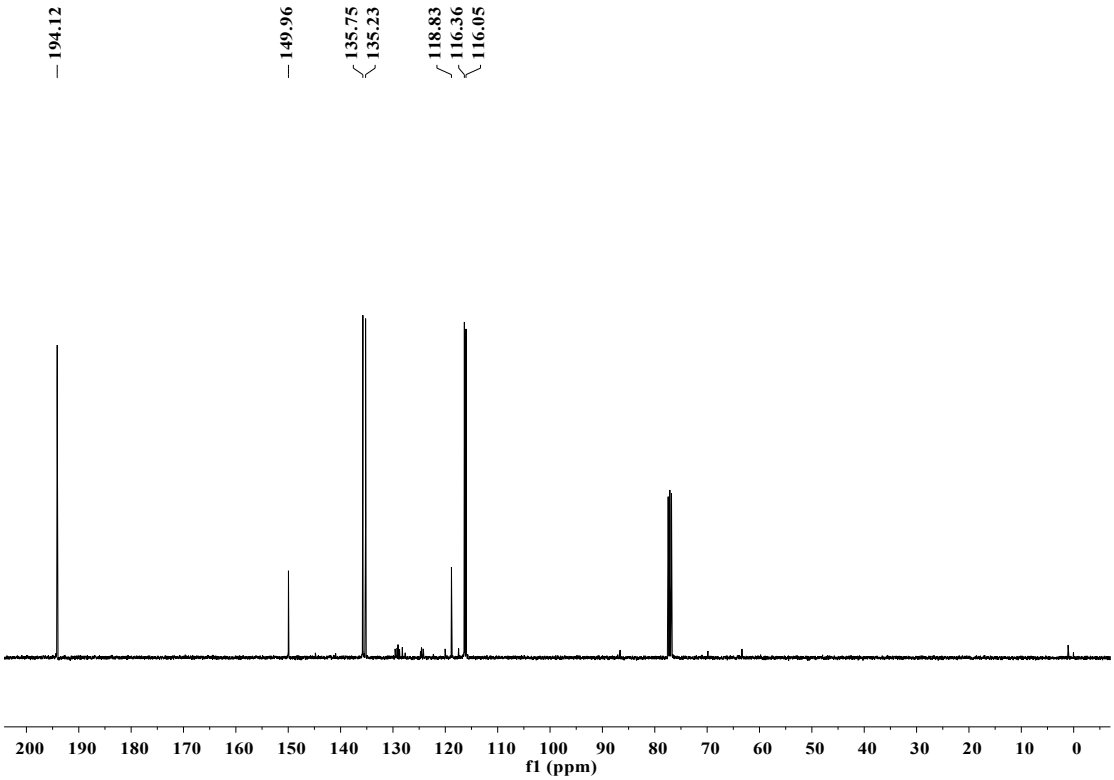


Figure S19: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of anthranilaldehyde (4b).

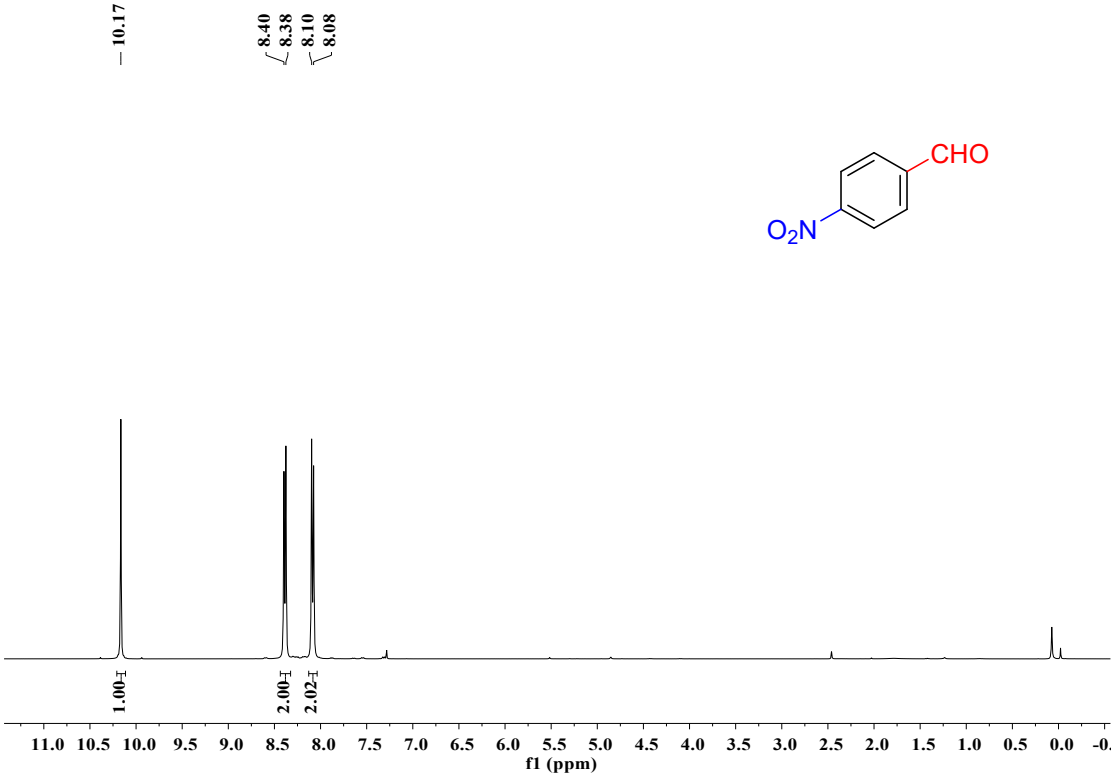


Figure S20: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-nitrobenzaldehyde (4c).

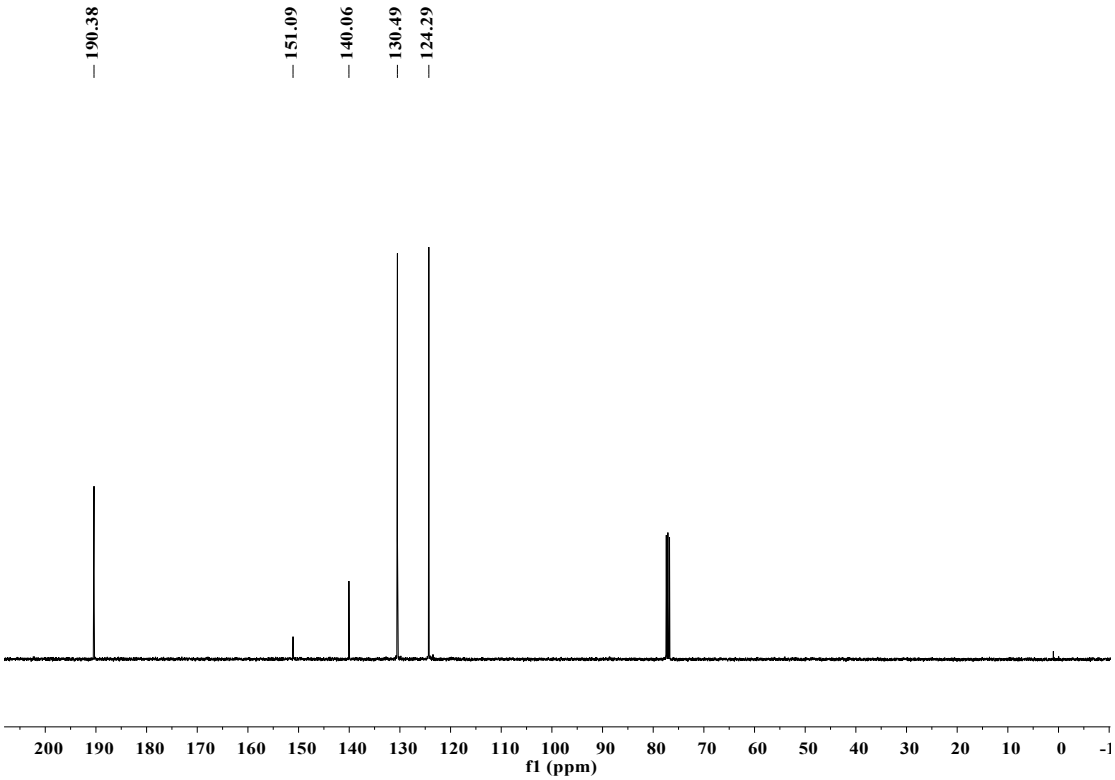


Figure S21: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-nitrobenzaldehyde (4c).

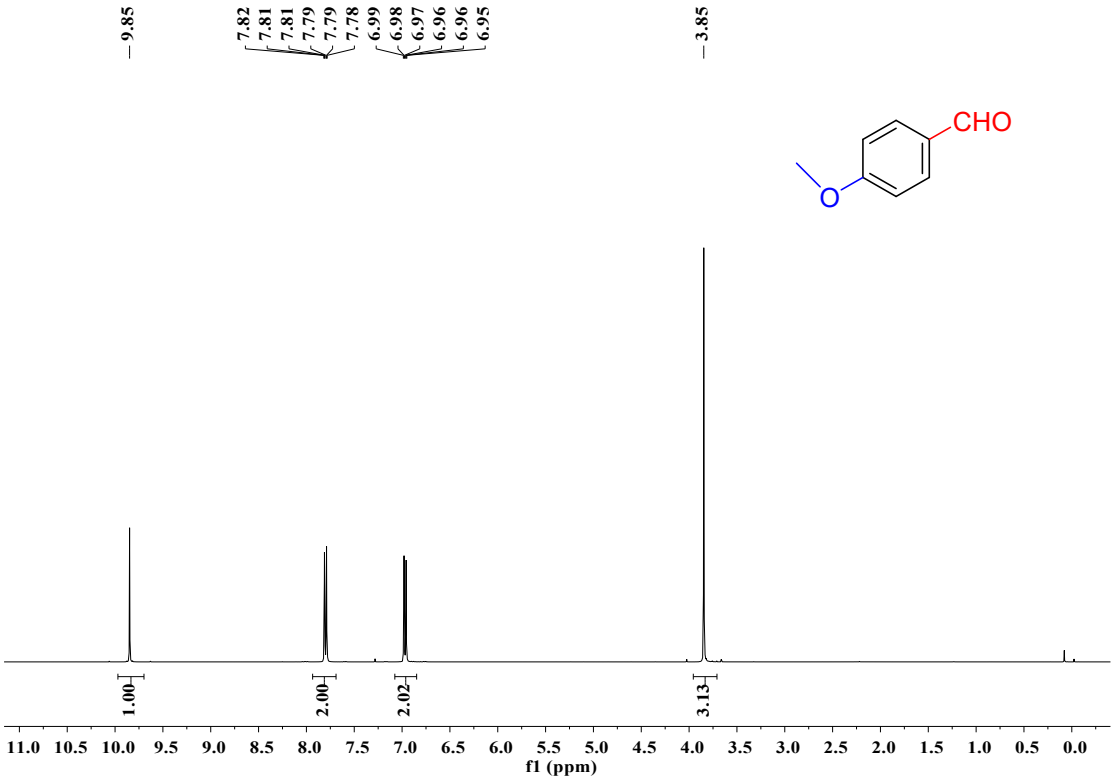


Figure S22: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-methoxybenzaldehyde (4d).

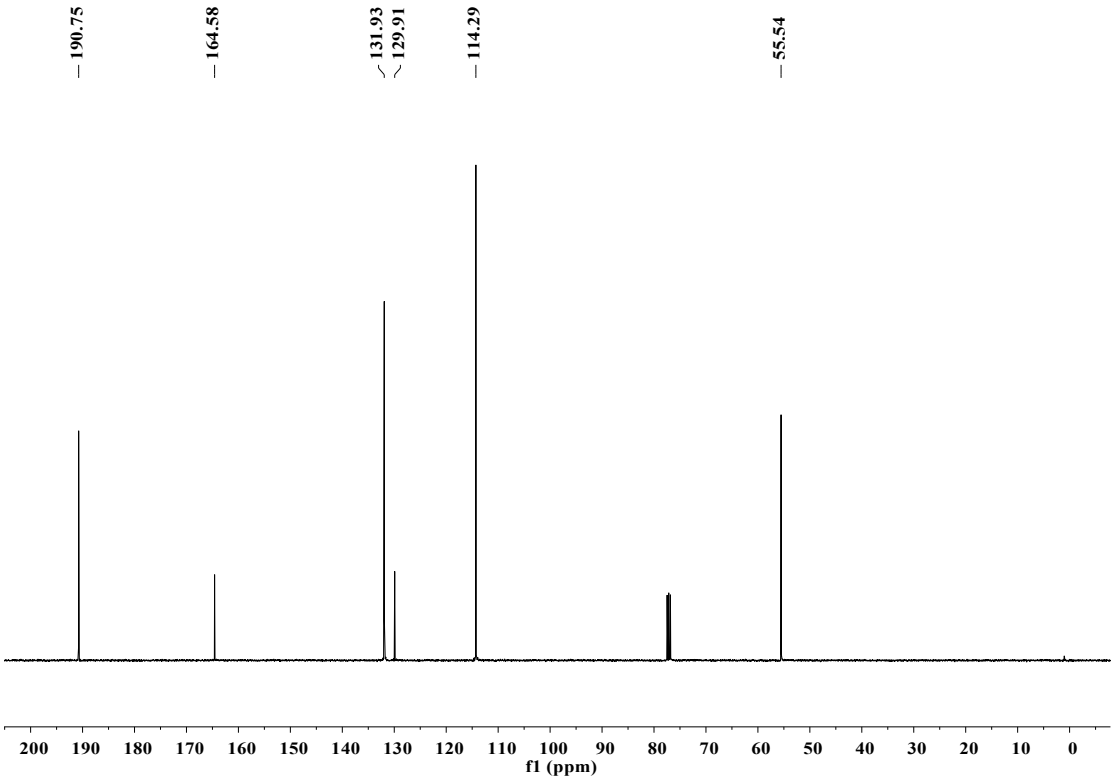


Figure S23: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-methoxybenzaldehyde (4d).

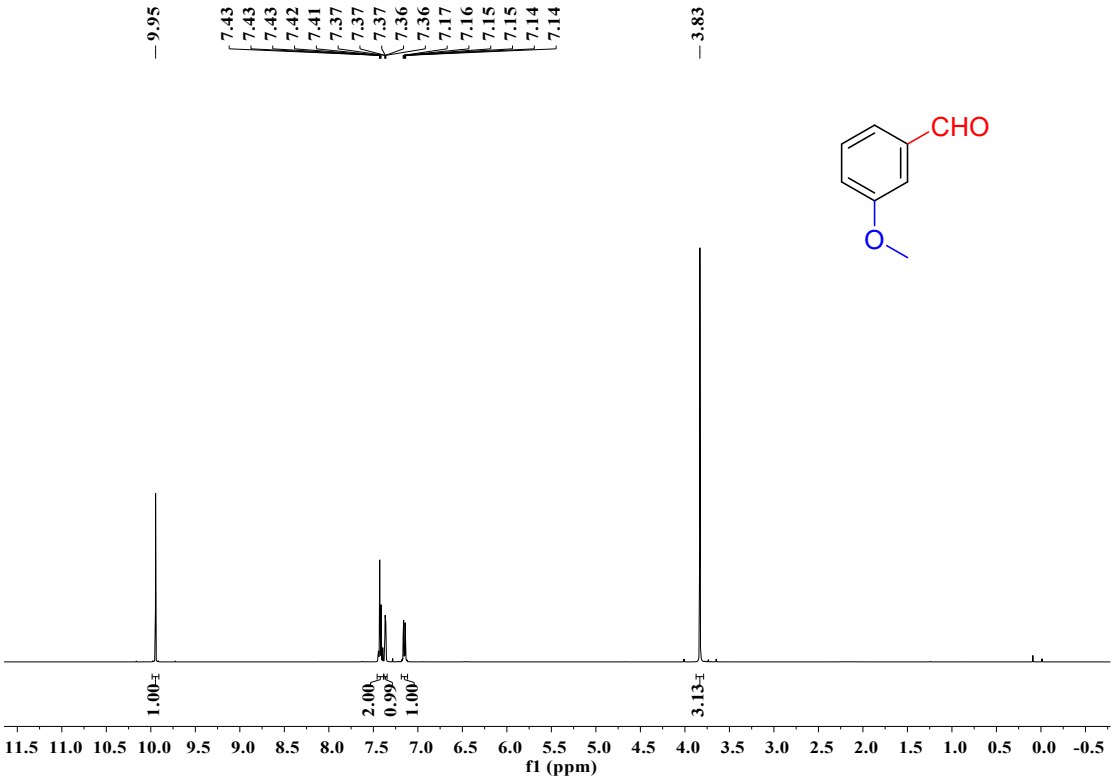


Figure S24: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-methoxybenzaldehyde (4e).

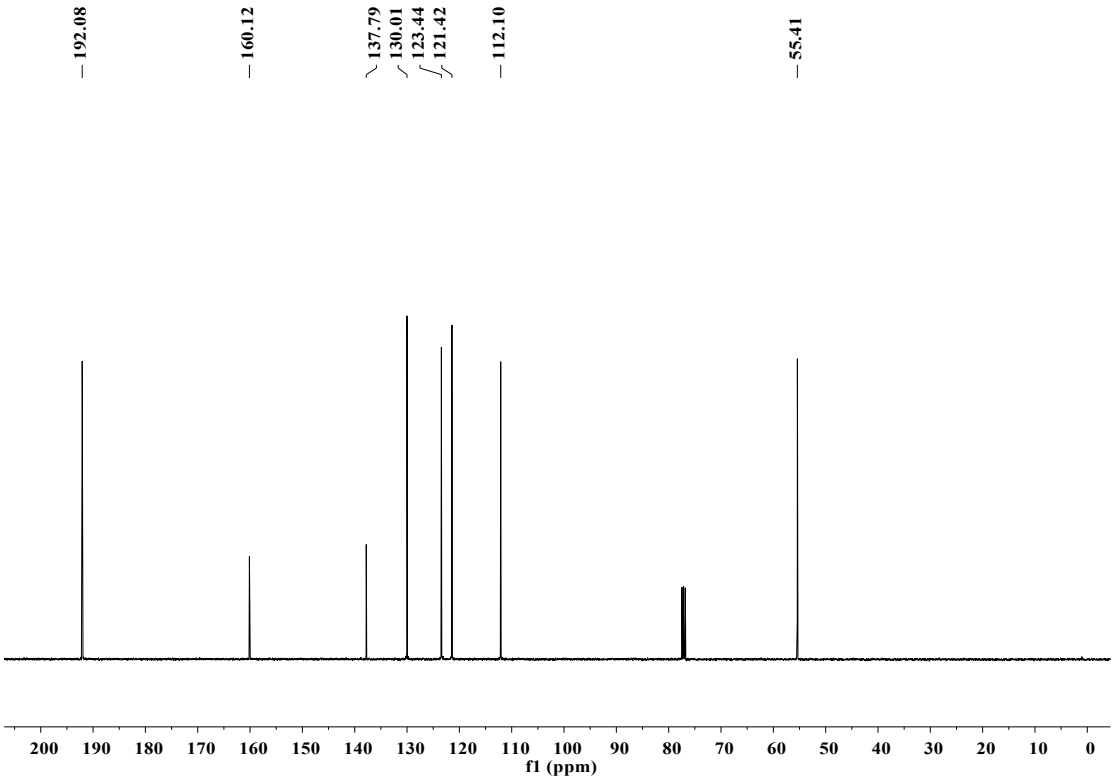


Figure S25: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 3-methoxybenzaldehyde (4e).

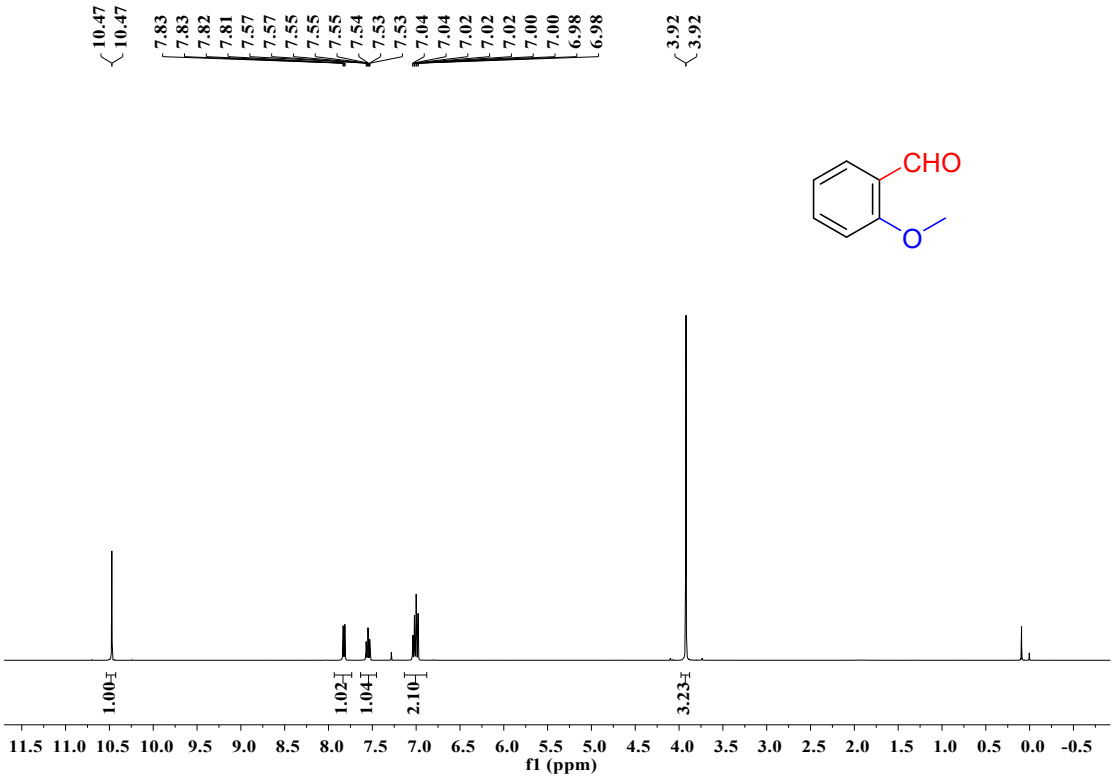


Figure S26: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2-methoxybenzaldehyde (4f).

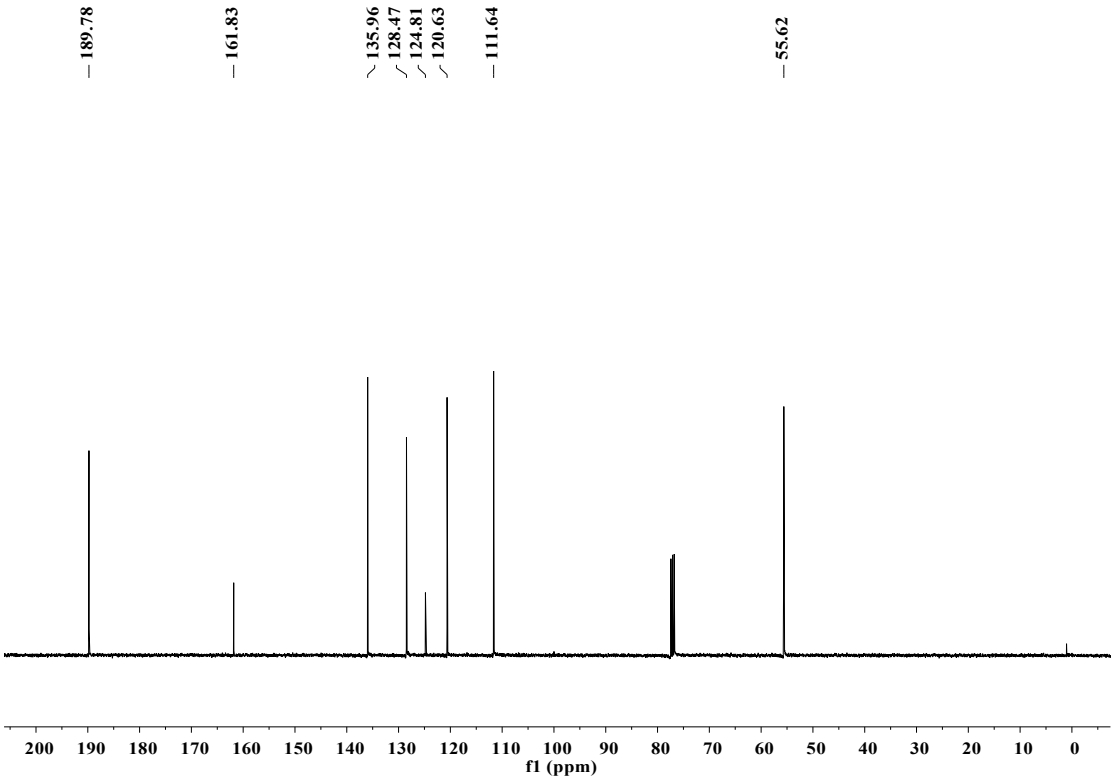


Figure S27: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2-methoxybenzaldehyde (4f).

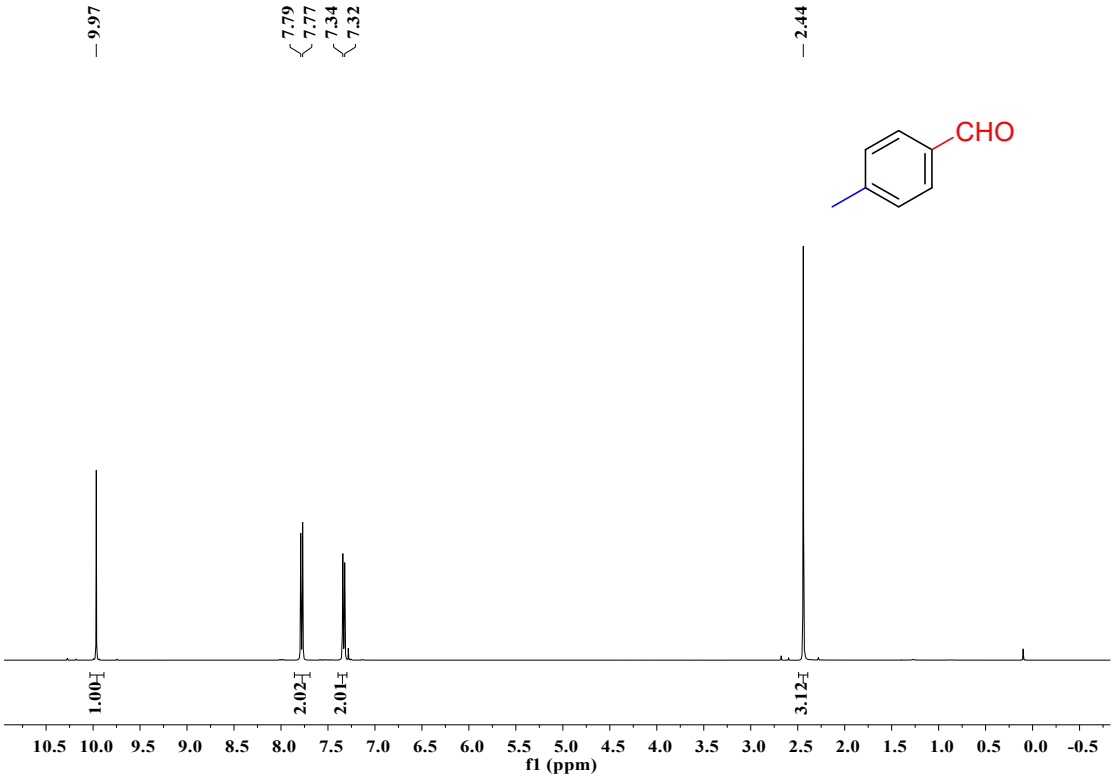


Figure S28: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-methylbenzaldehyde (4g).

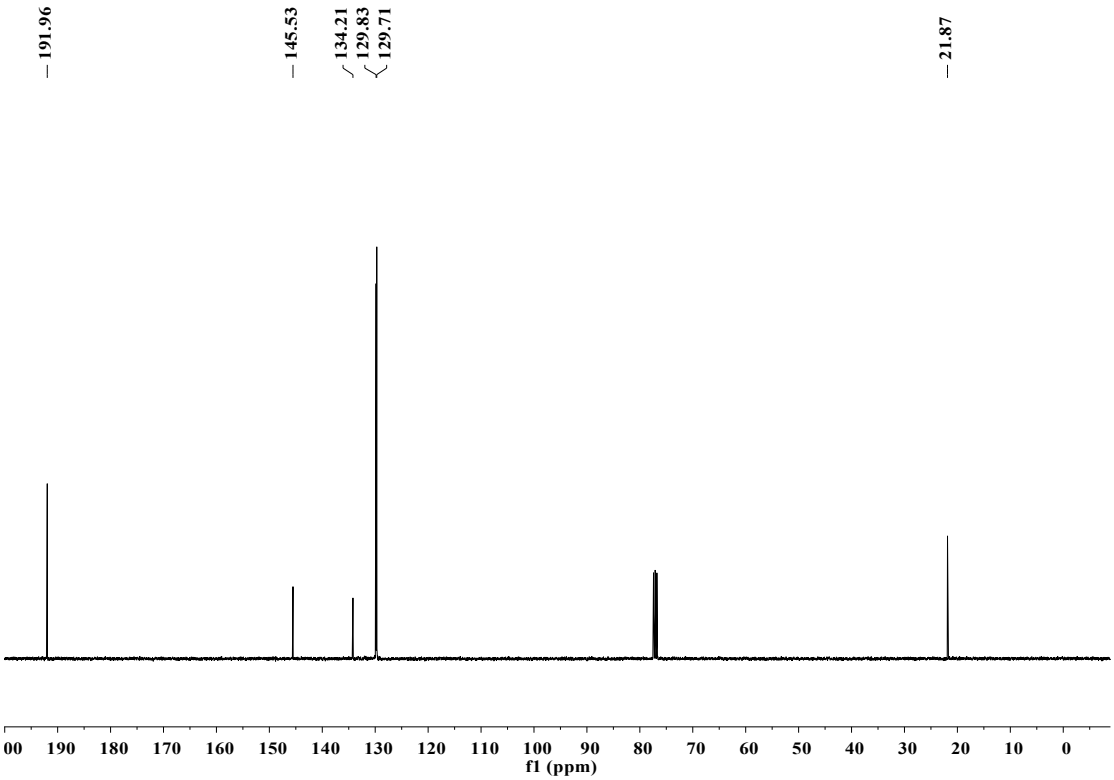


Figure S29: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-methylbenzaldehyde (4g).

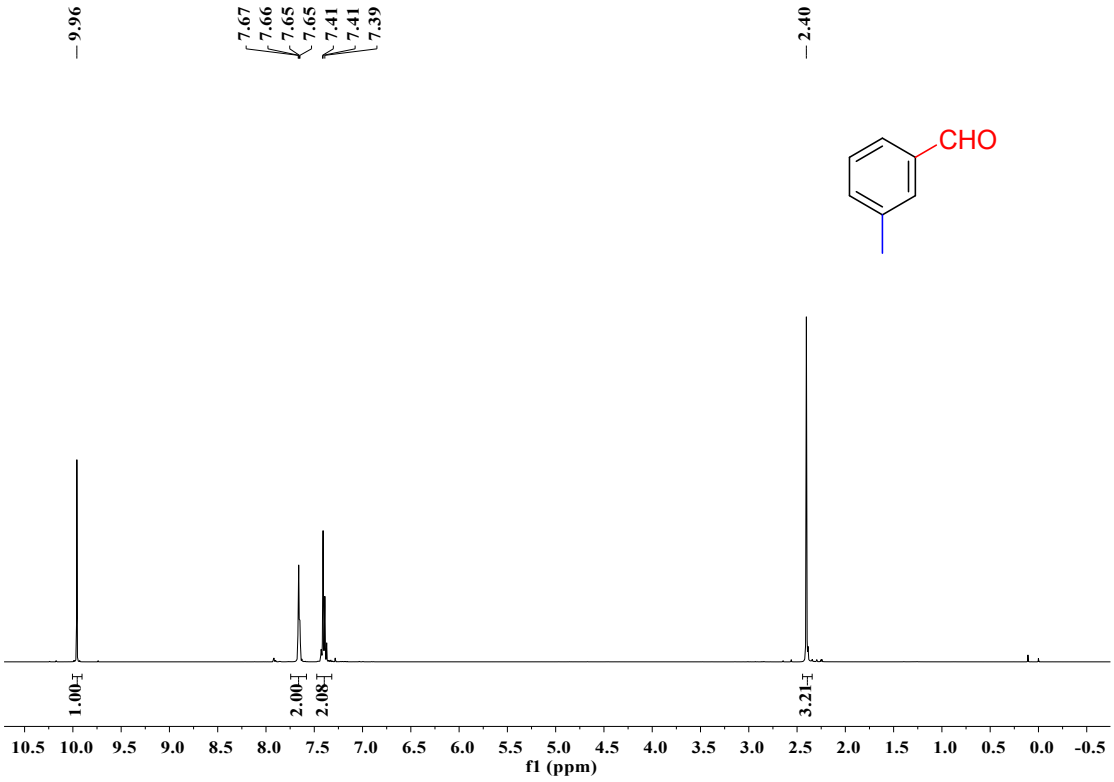


Figure S30: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-methylbenzaldehyde (4h).

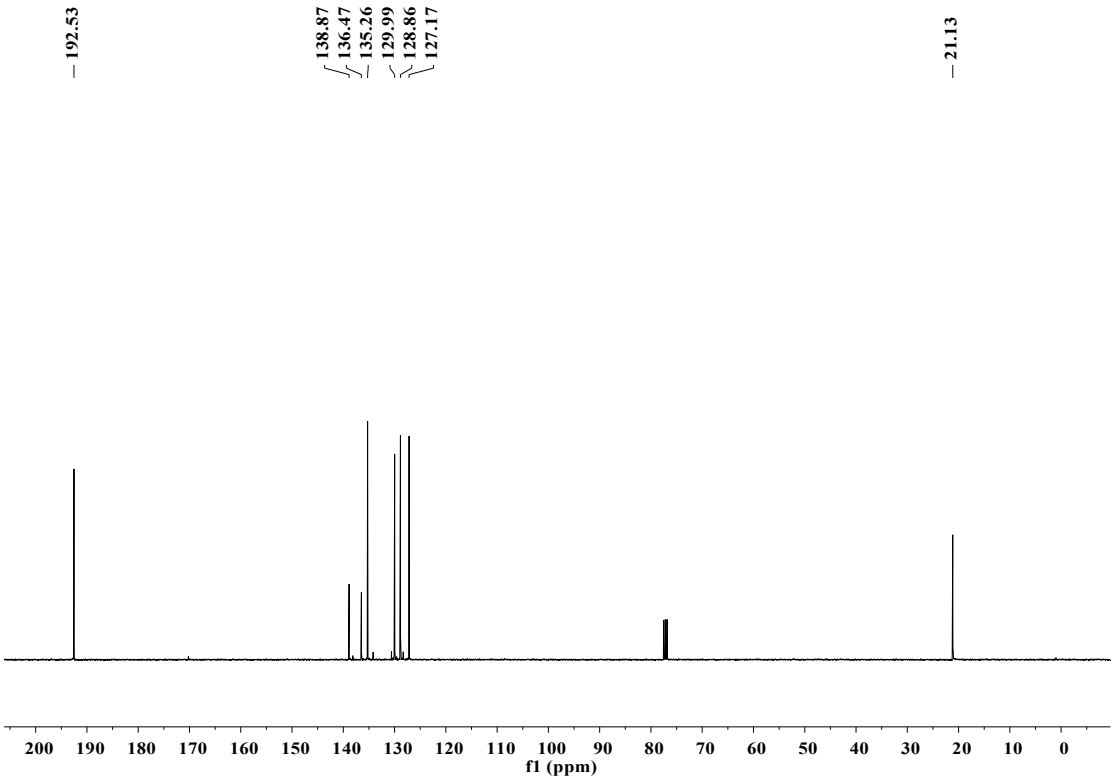


Figure S31: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 3-methylbenzaldehyde (4h).

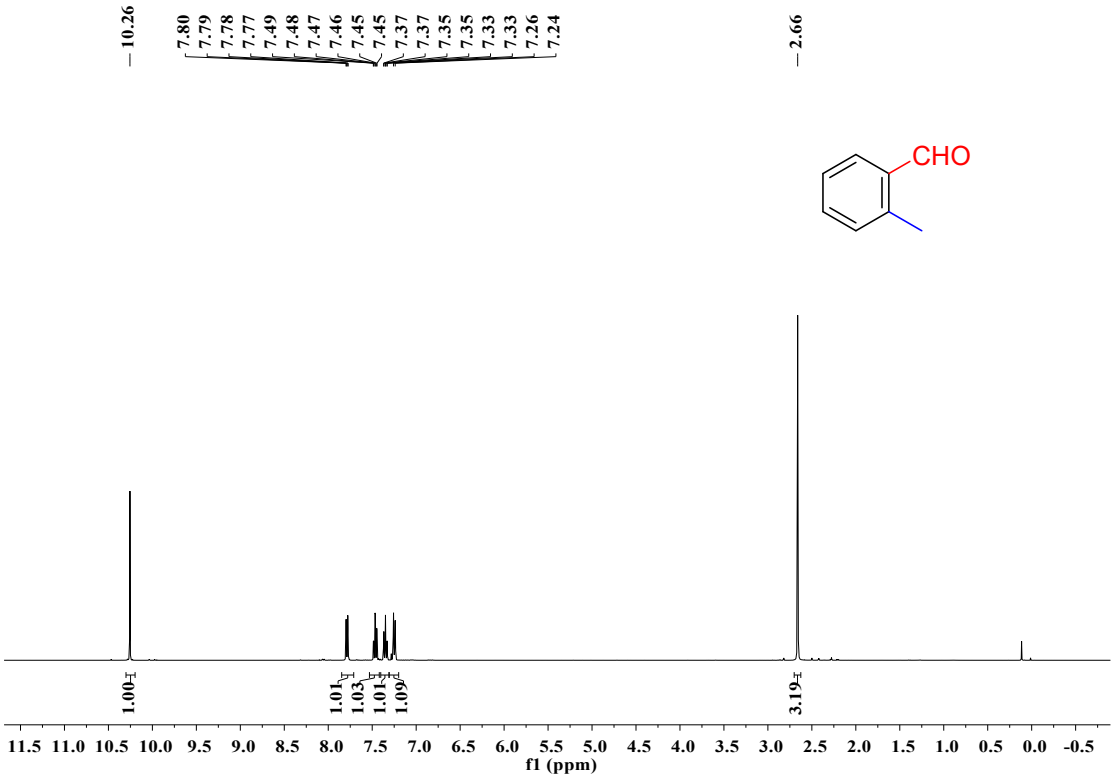


Figure S32: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2-methylbenzaldehyde (4i).

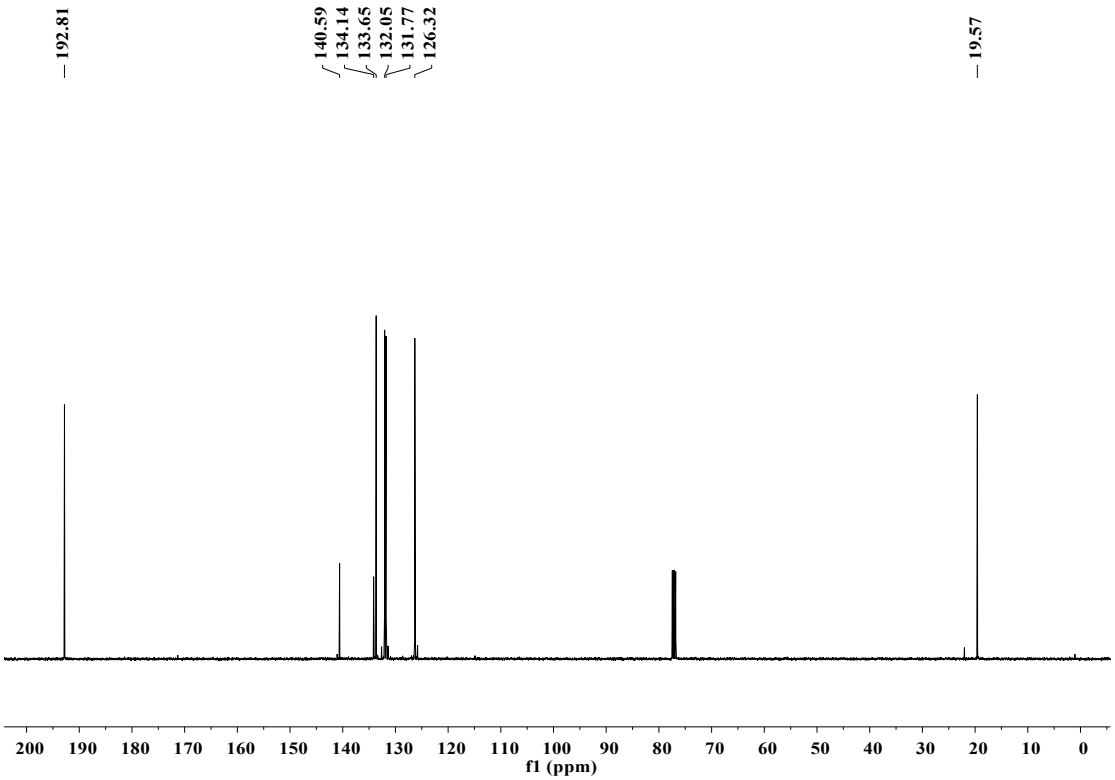


Figure S33: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2-methylbenzaldehyde (4i).

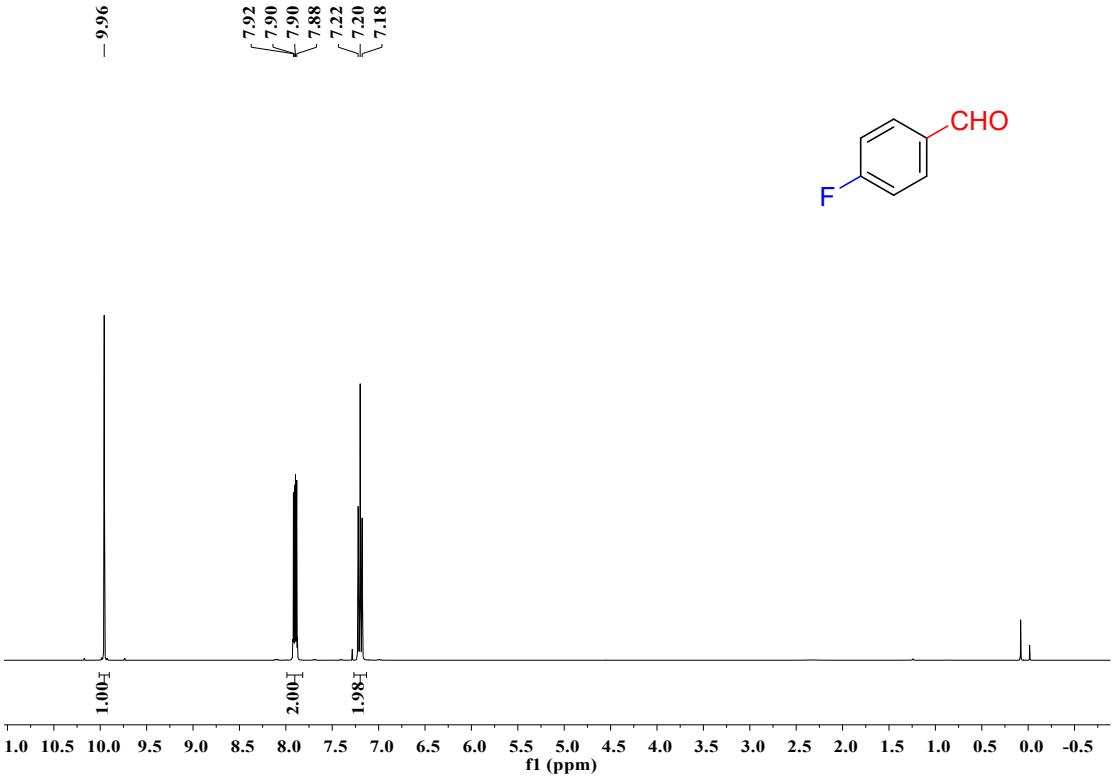


Figure S34: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-fluorobenzaldehyde (4j).

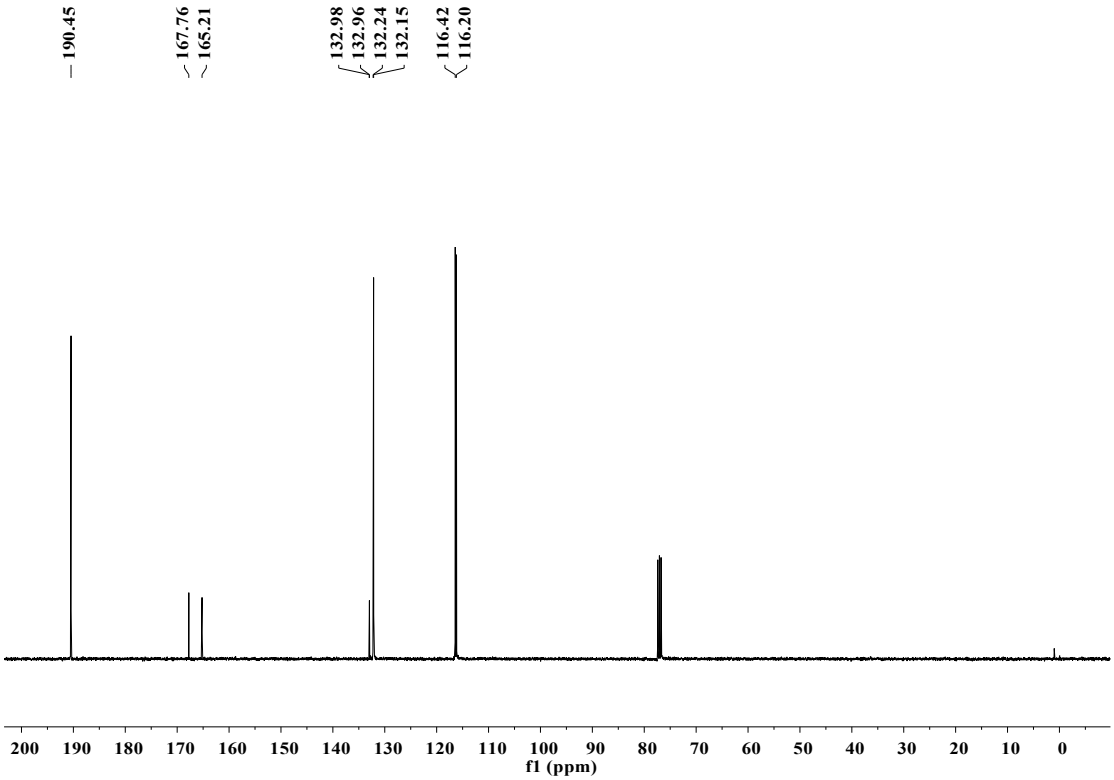


Figure S35: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-fluorobenzaldehyde (4j).

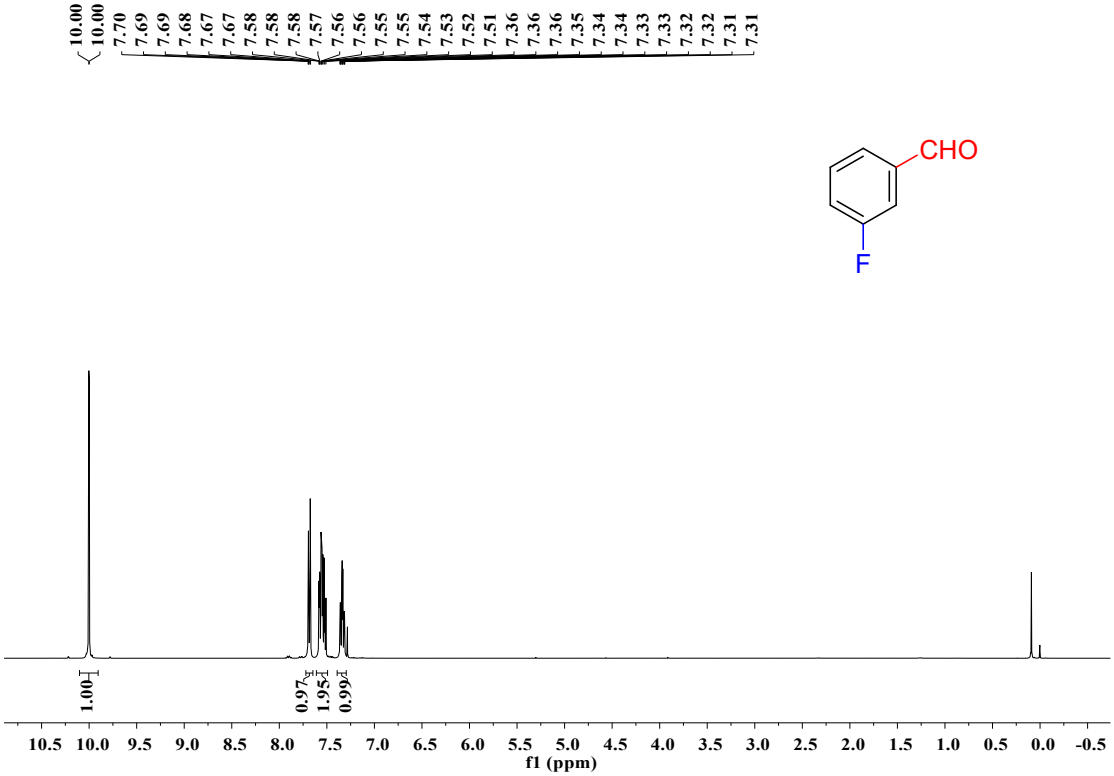


Figure S36: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-fluorobenzaldehyde (4k).

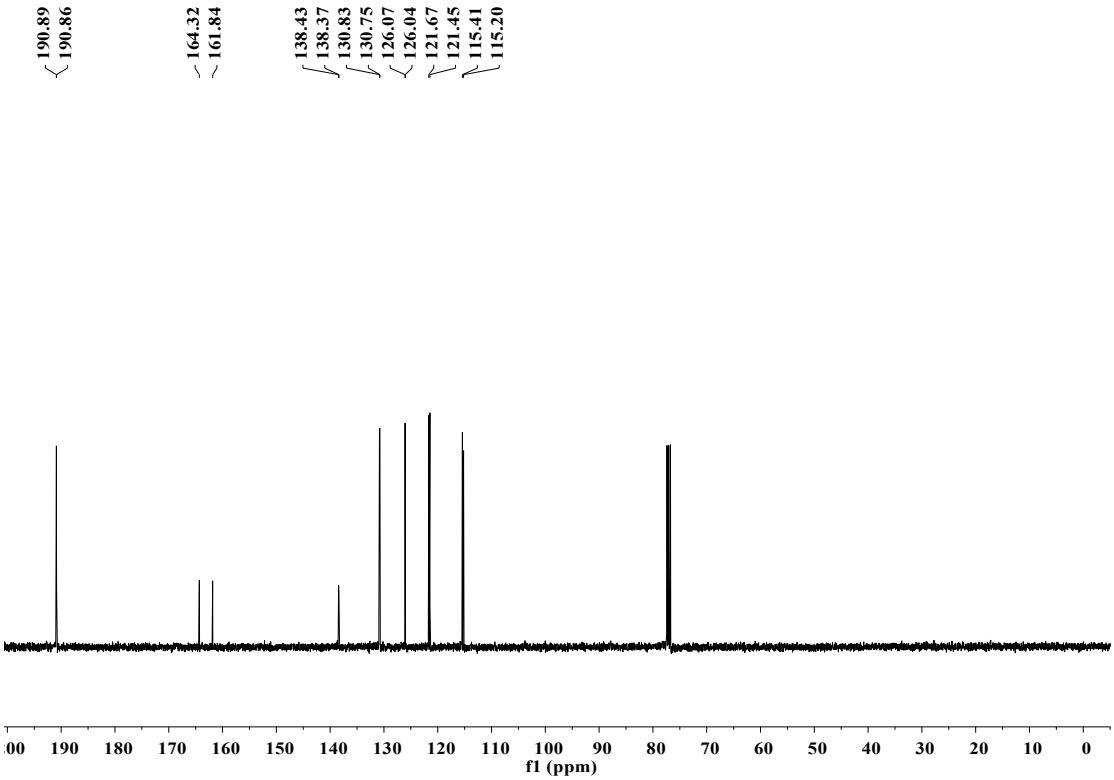


Figure S37: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 3-fluorobenzaldehyde (4k).

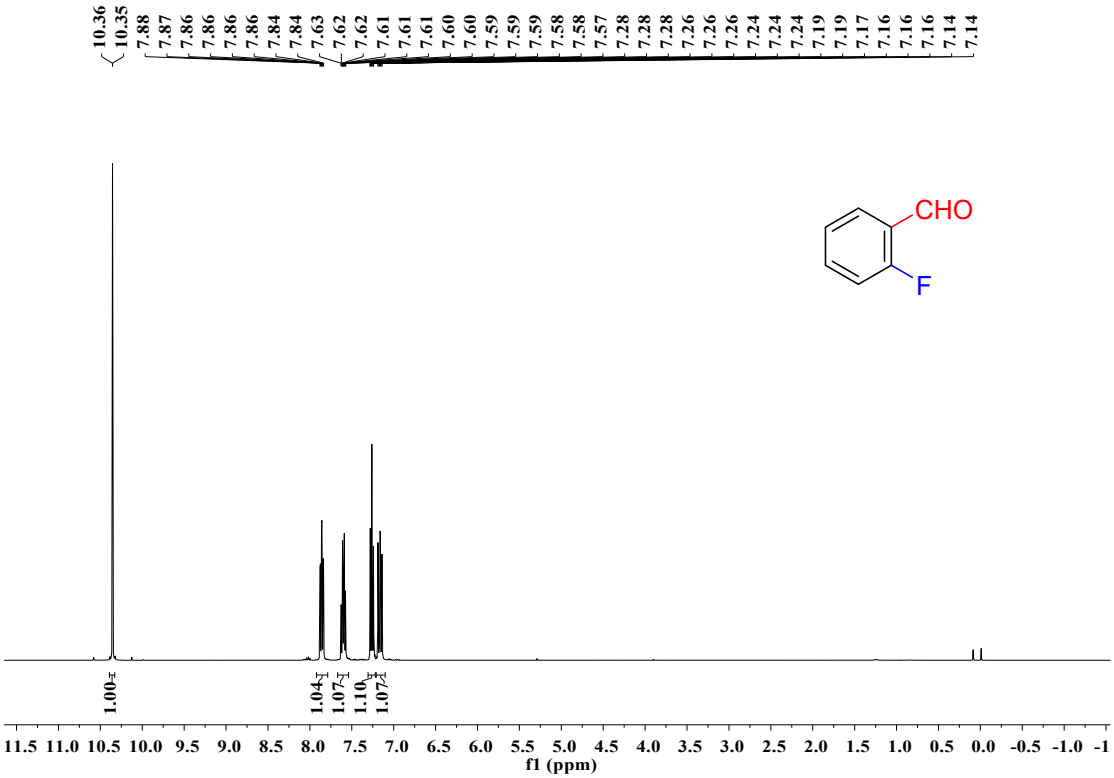


Figure S38: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2-fluorobenzaldehyde (4l).

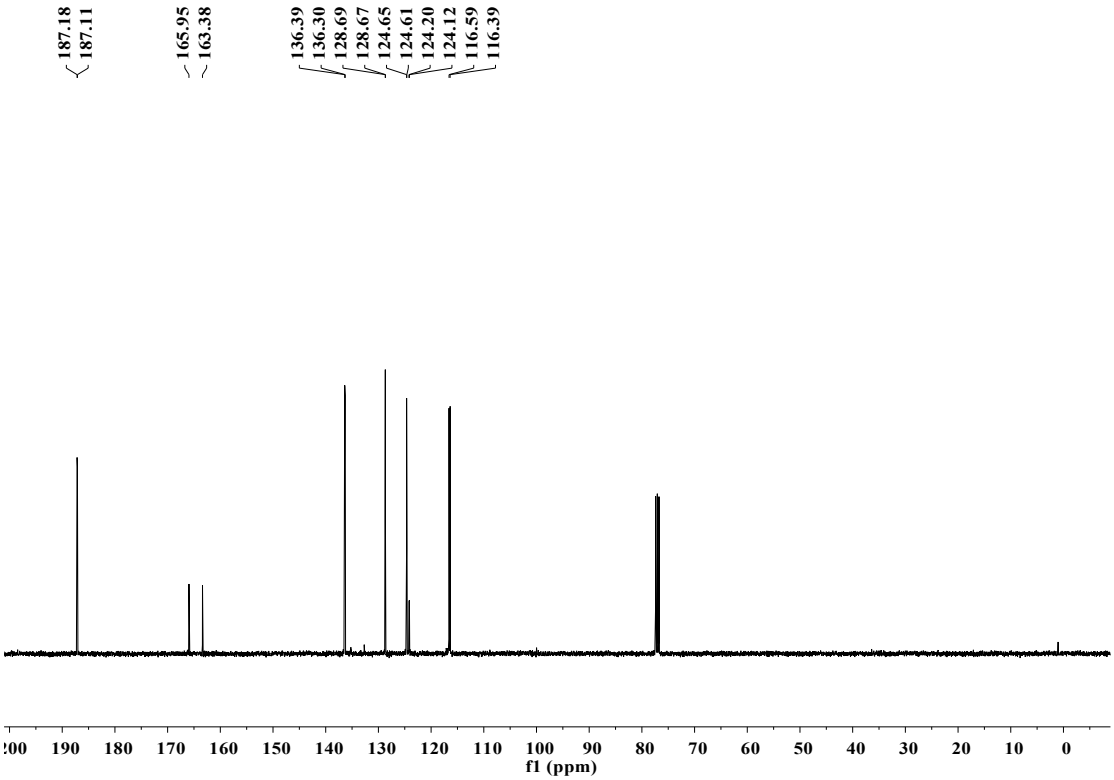


Figure S39: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2-fluorobenzaldehyde (4l).

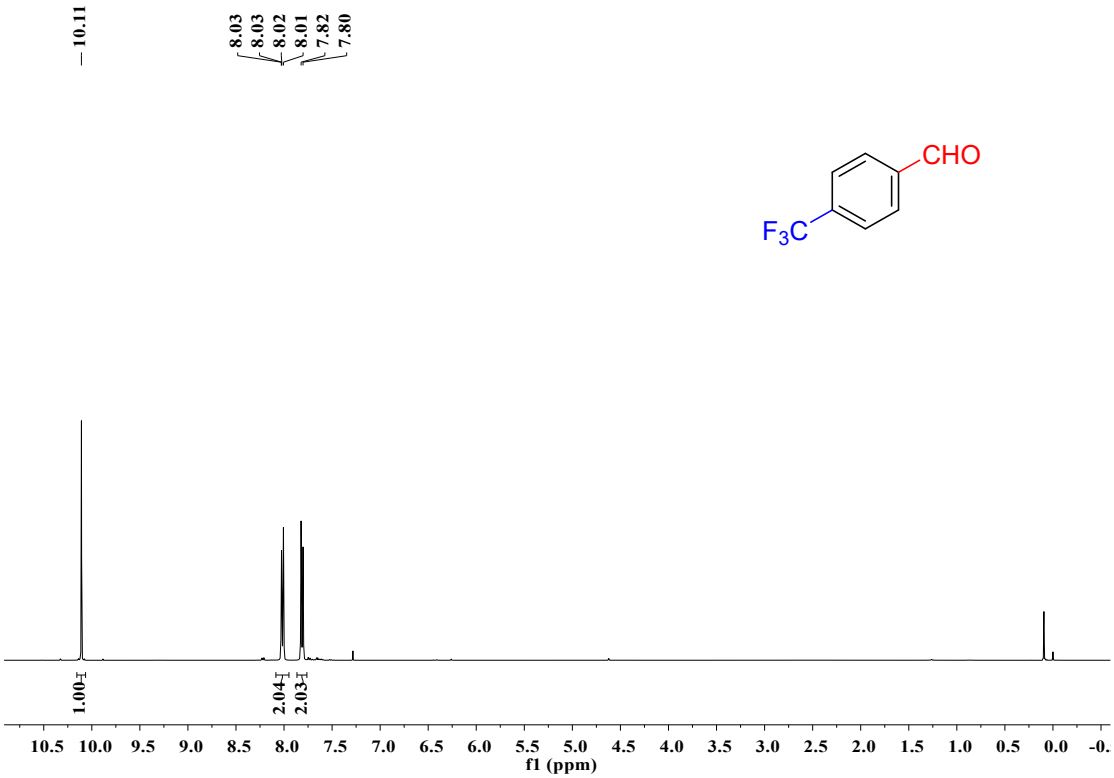


Figure S40: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-(trifluoromethyl)-benzaldehyde (4m).

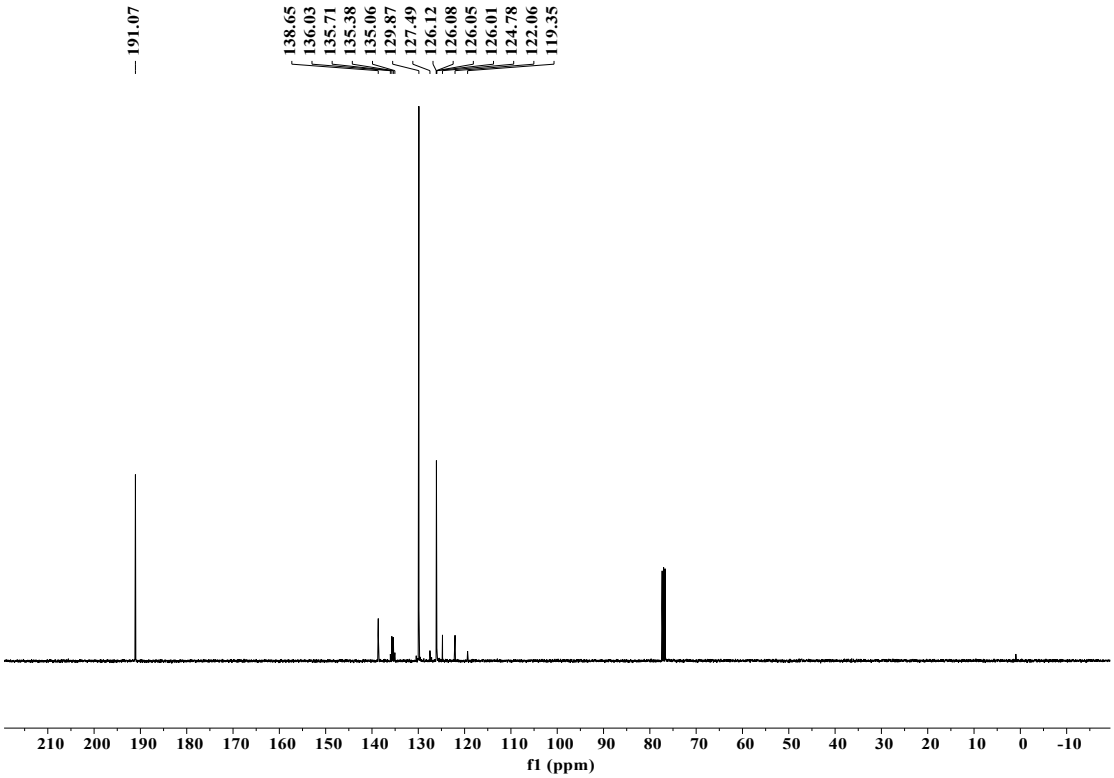


Figure S41: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-(trifluoromethyl)-benzaldehyde (4m).

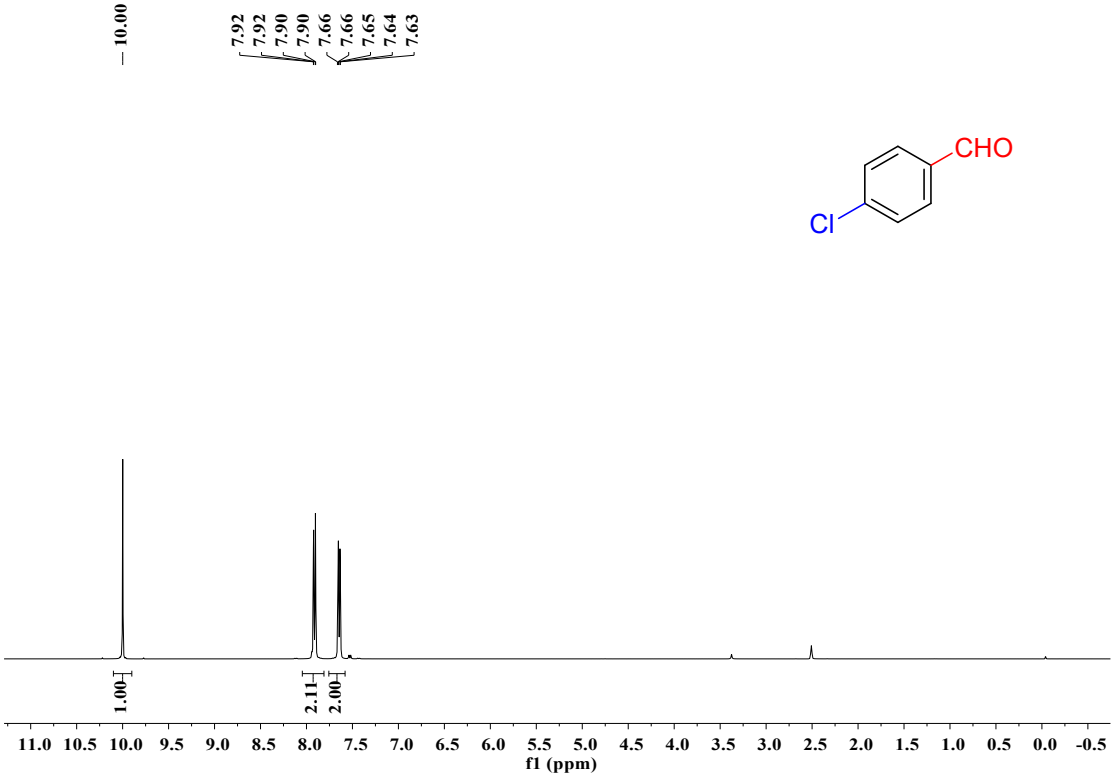


Figure S42: ^{1}H NMR spectrum of (400 MHz, DMSO) of 4-chlorobenzaldehyde (4n).

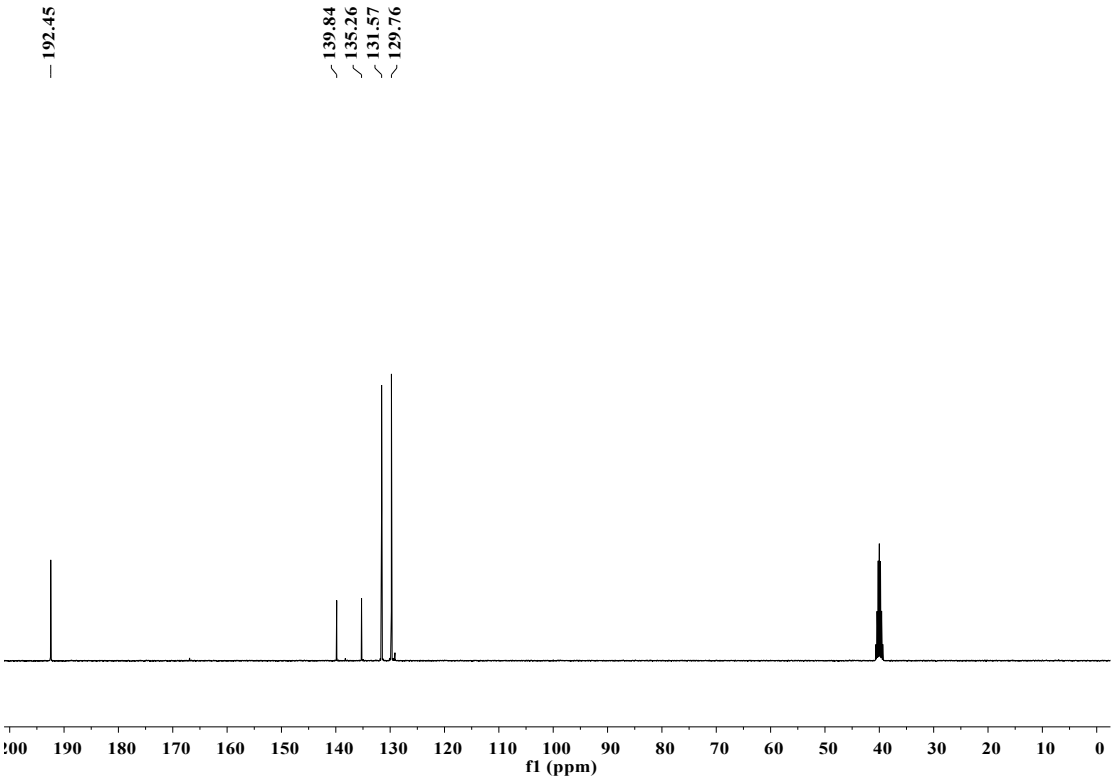


Figure S43: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, DMSO) of 4-chlorobenzaldehyde (4n).

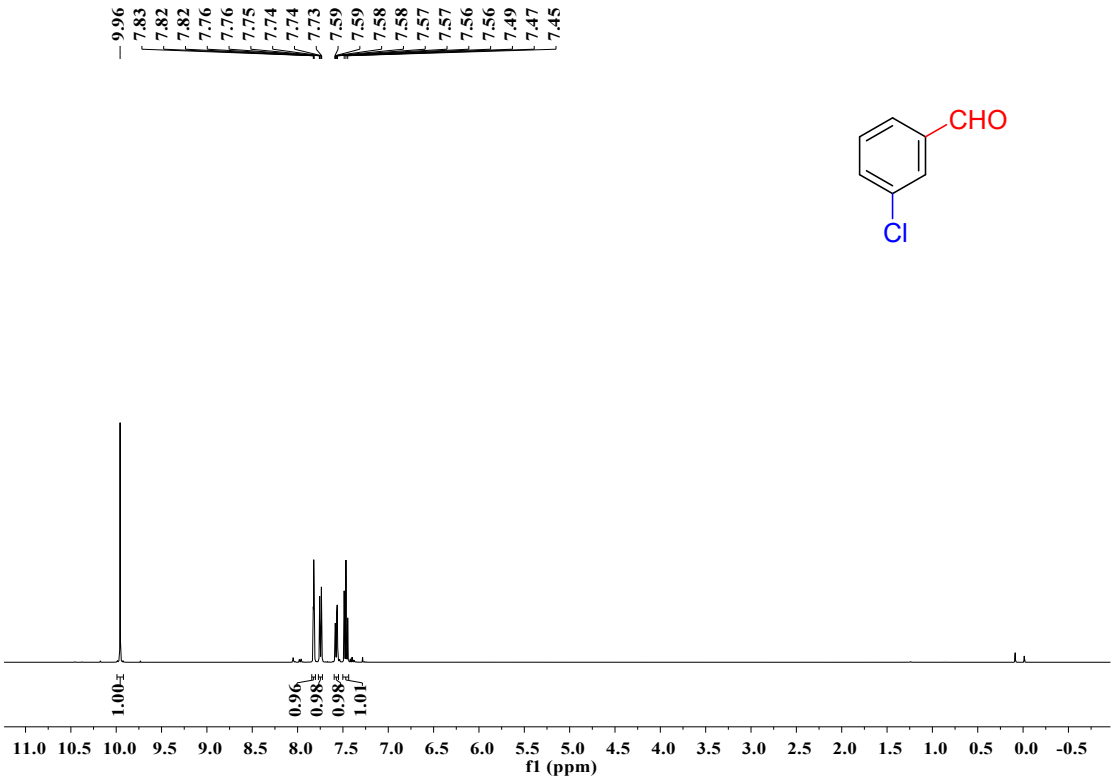


Figure S44: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-chlorobenzaldehyde (4o).

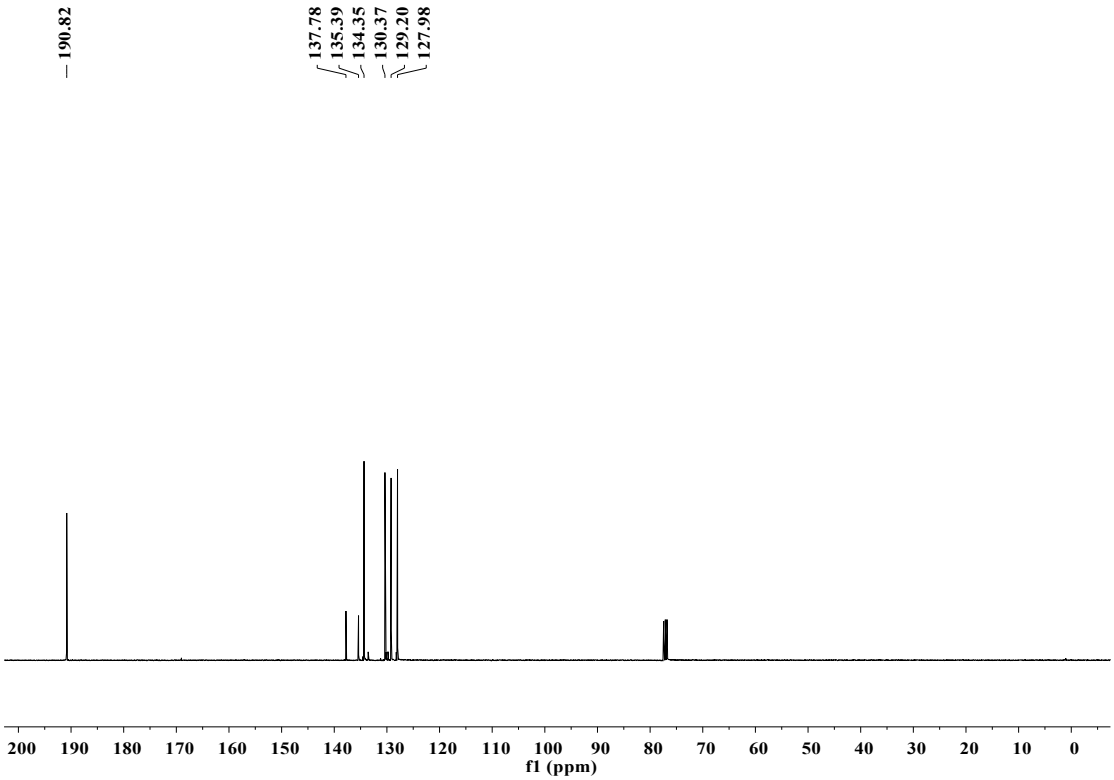


Figure S45: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 3-chlorobenzaldehyde (4o).

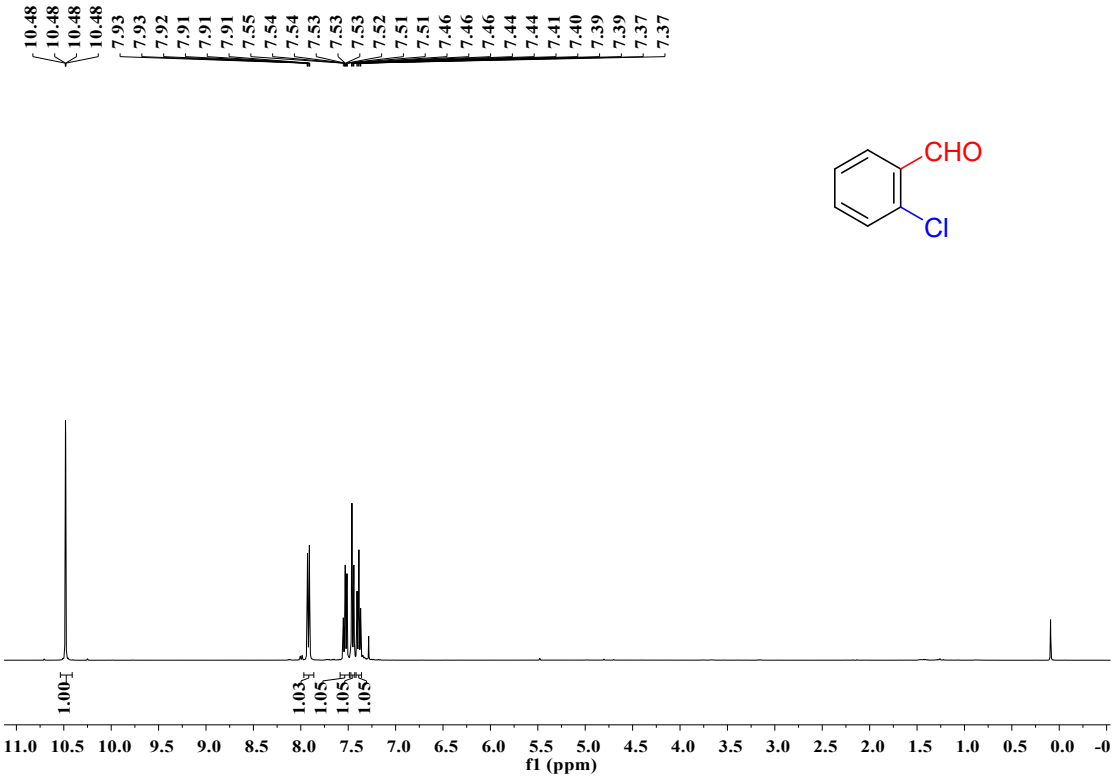


Figure S46: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2-chlorobenzaldehyde (4p).

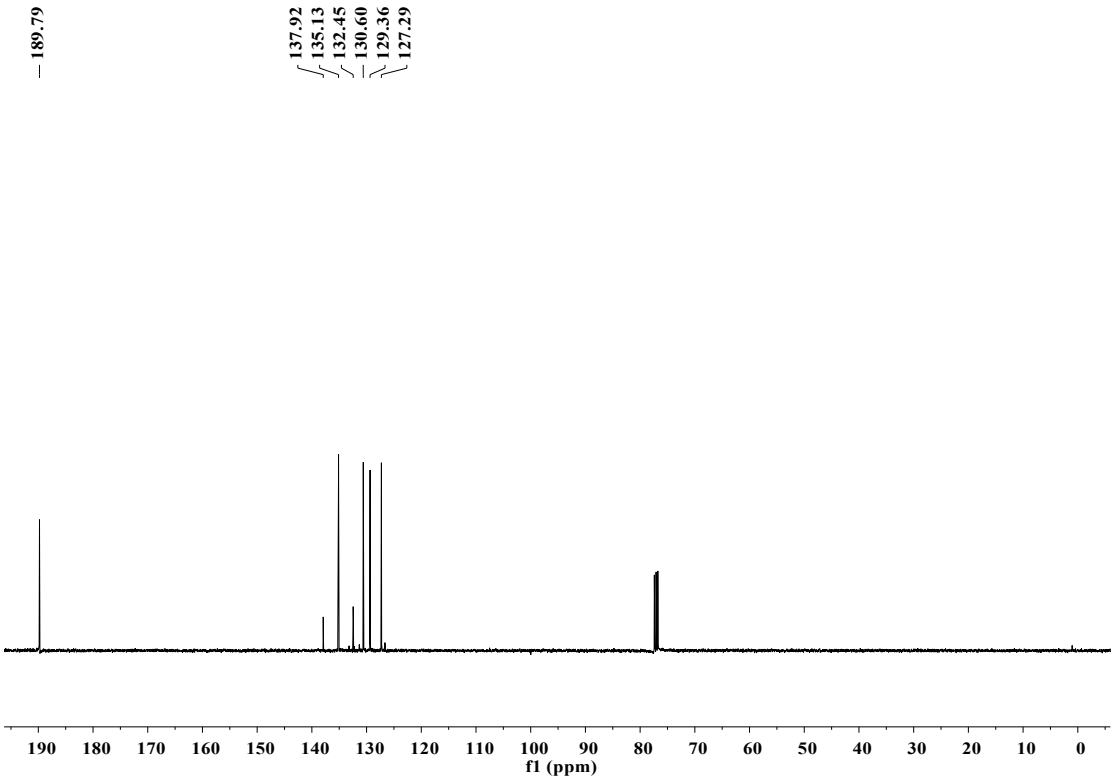


Figure S47: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2-chlorobenzaldehyde (4p).

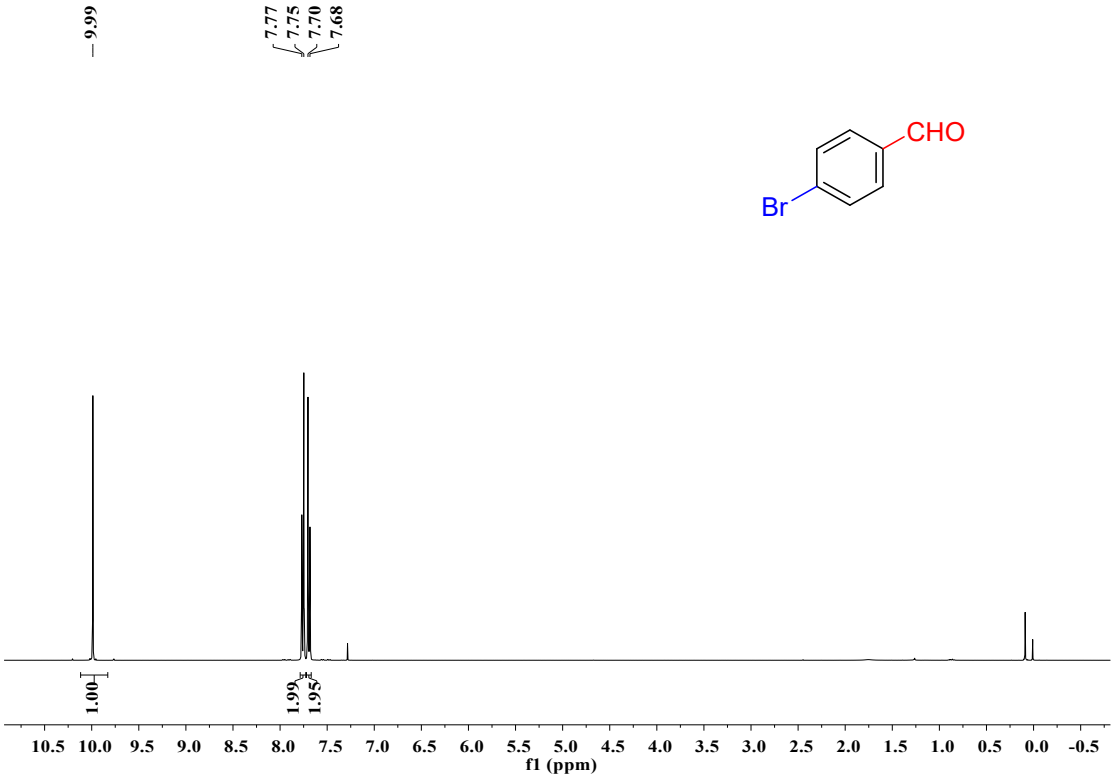


Figure S48: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-bromobenzaldehyde (4q).

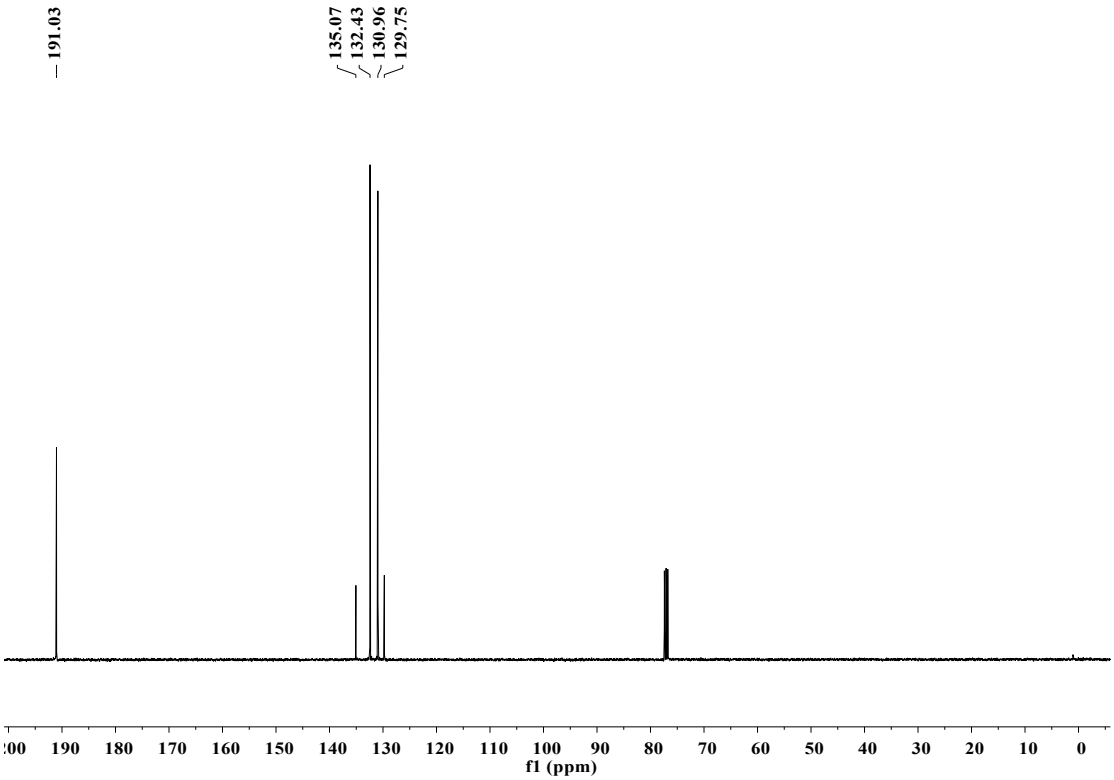


Figure S49: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-bromobenzaldehyde (4q).

Figure S50

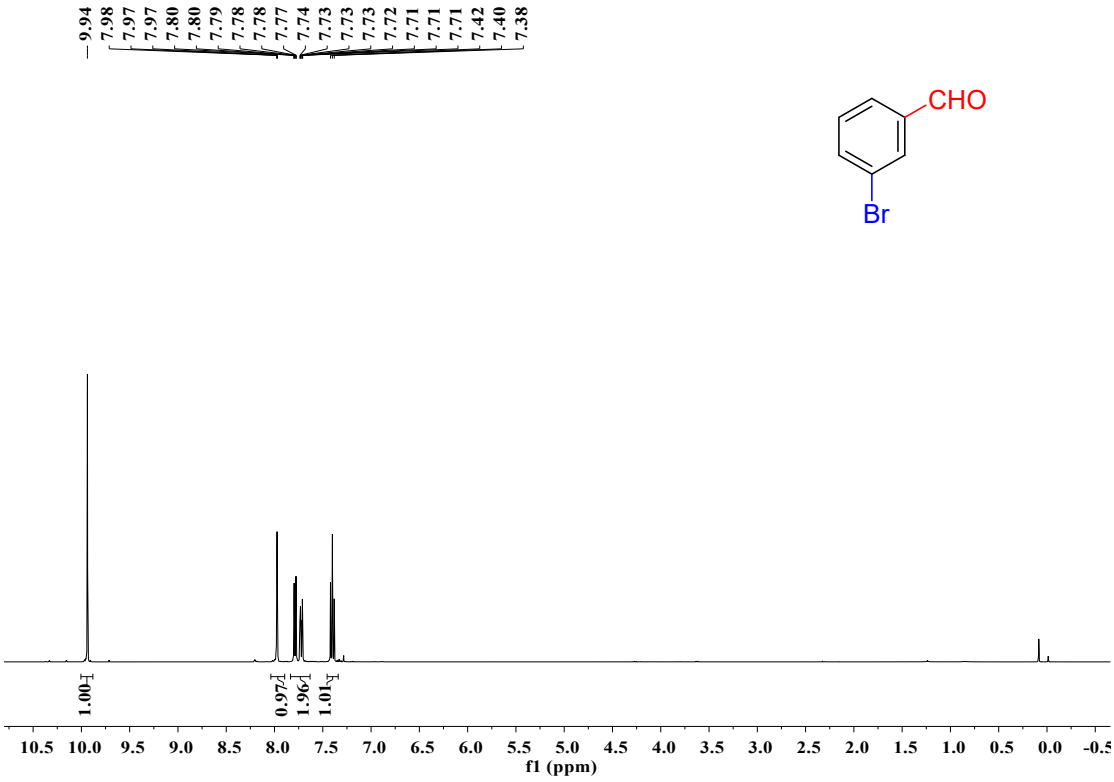


Figure S51: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-bromobenzaldehyde (4r).

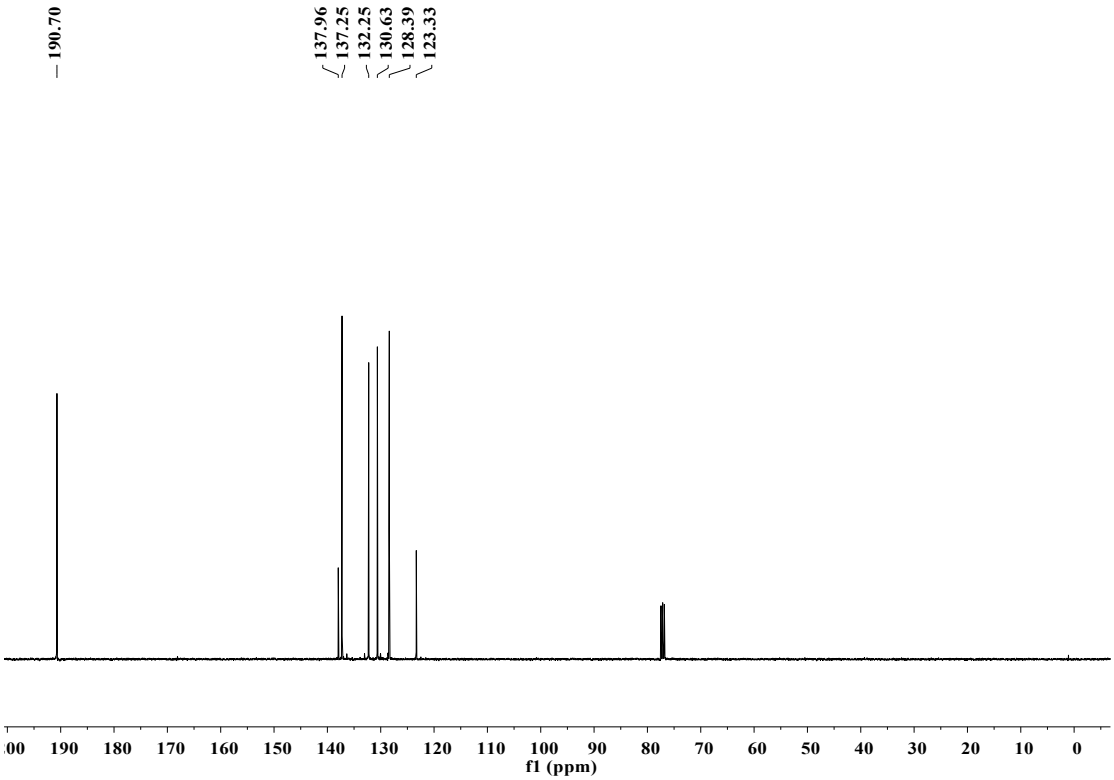


Figure S52: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 3-bromobenzaldehyde (4r).

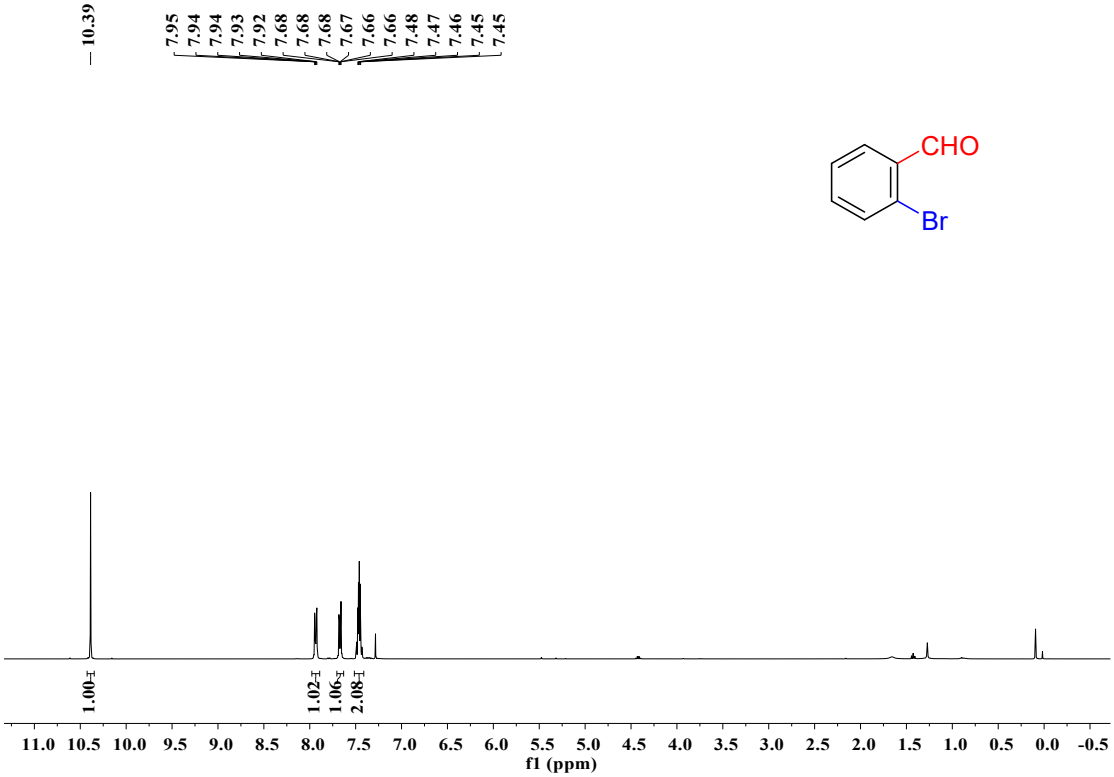


Figure S53: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2-bromobenzaldehyde (4s).

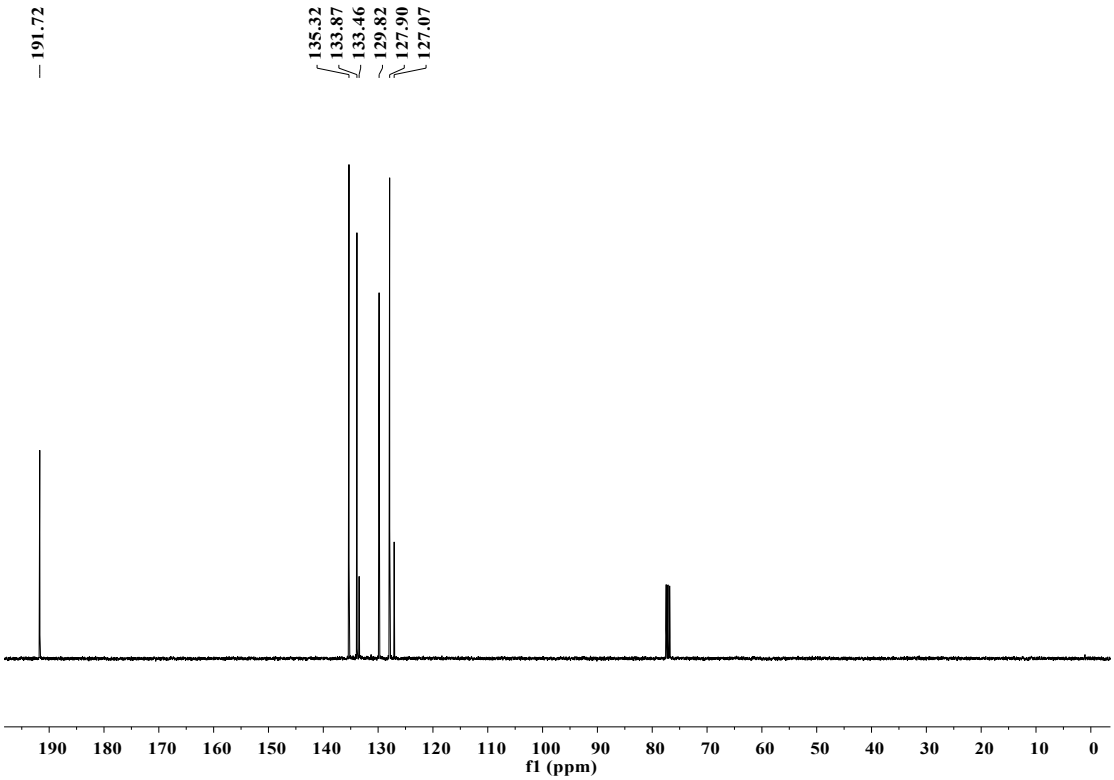


Figure S54: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2-bromobenzaldehyde (4s).

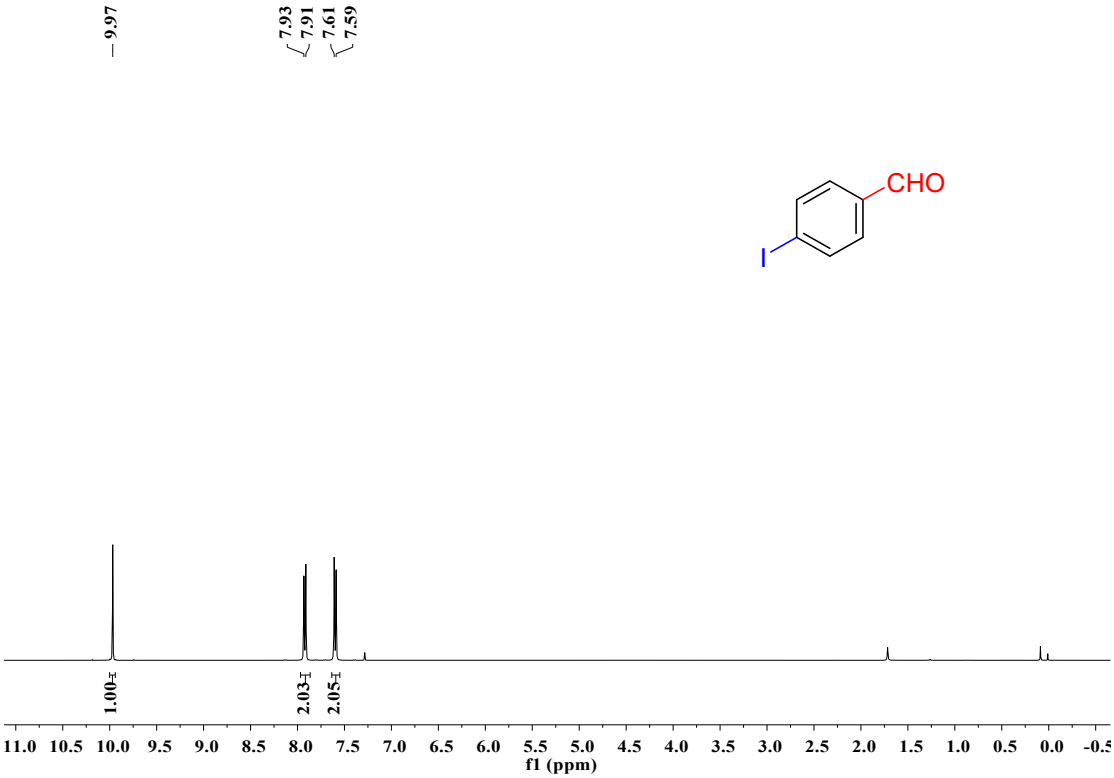


Figure S55: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-iodobenzaldehyde (4t).

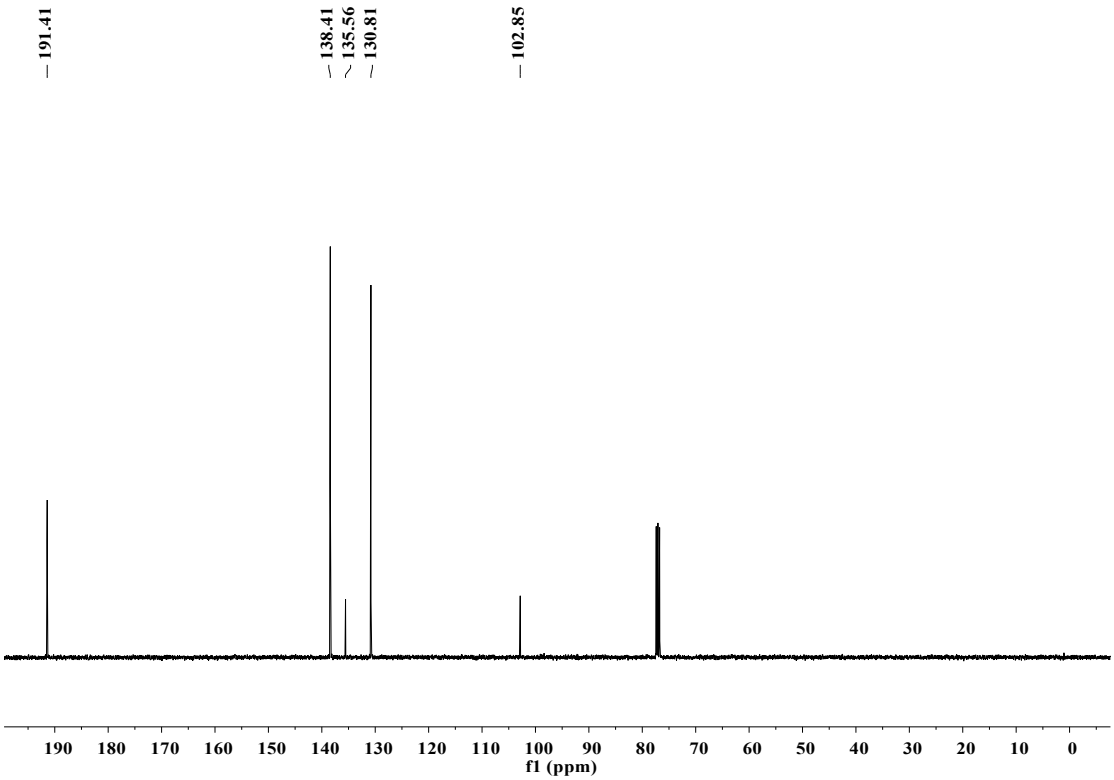


Figure S56: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-iodobenzaldehyde (4t).

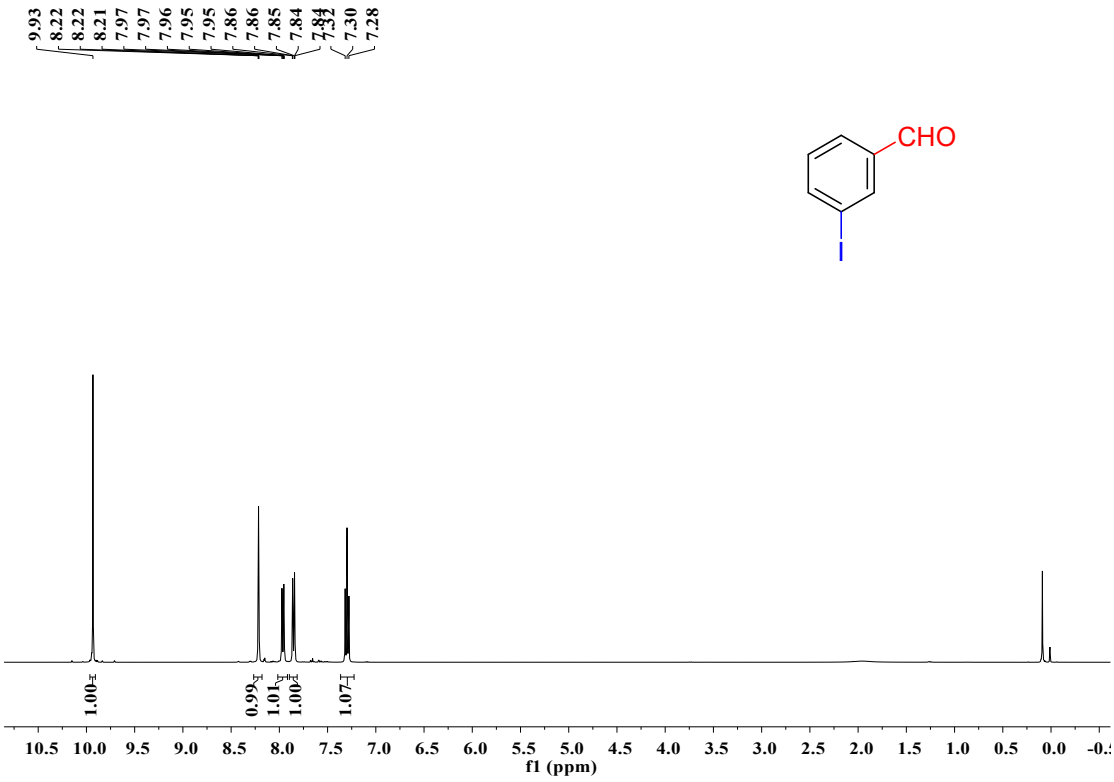


Figure S57: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-iodobenzaldehyde (4u).

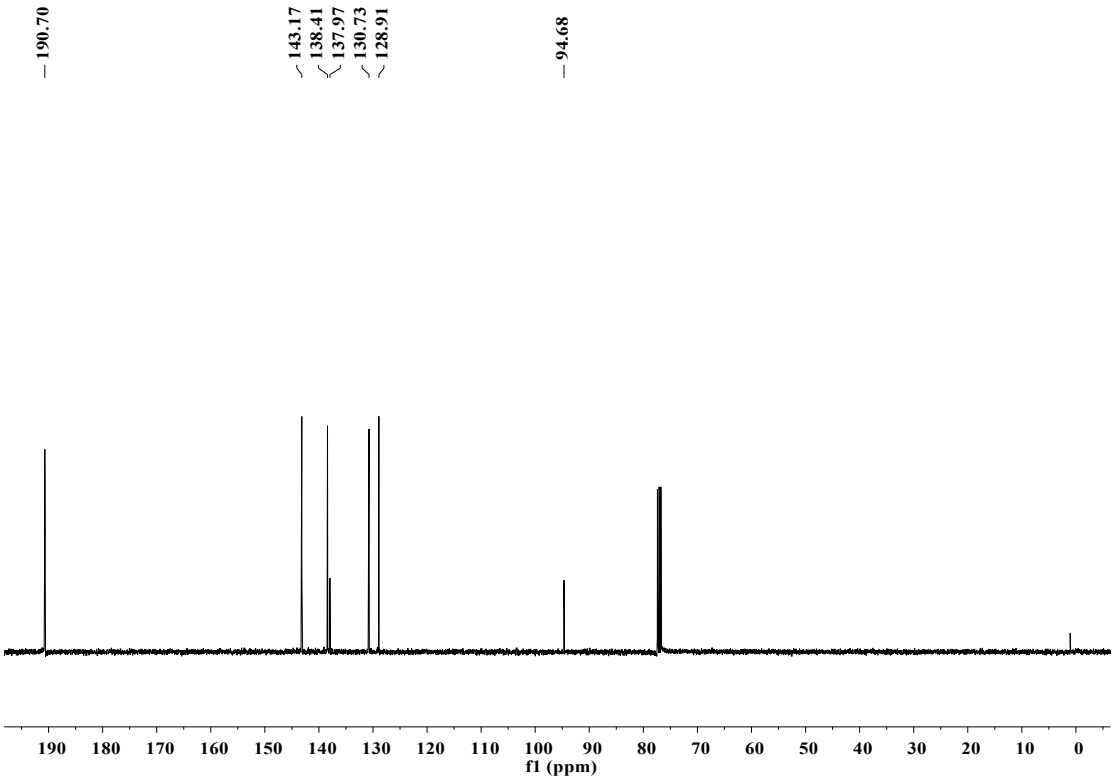


Figure S58: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 3-iodobenzaldehyde (4u).

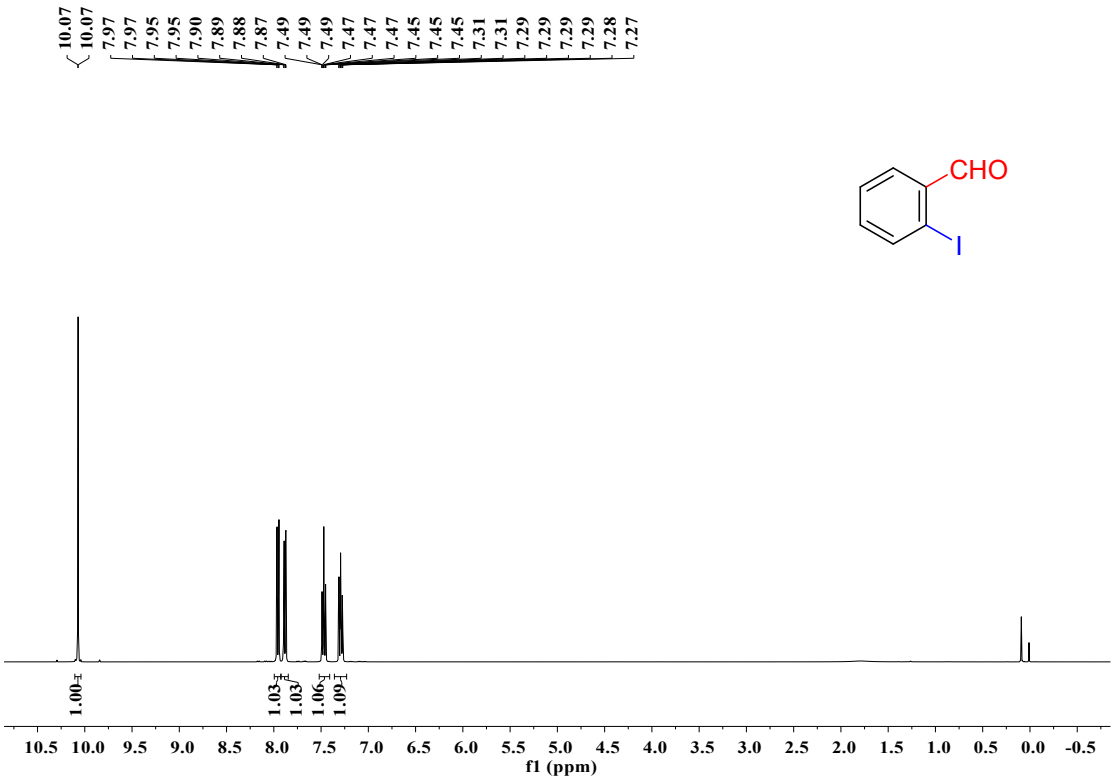


Figure S59: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2-iodobenzaldehyde (4v).

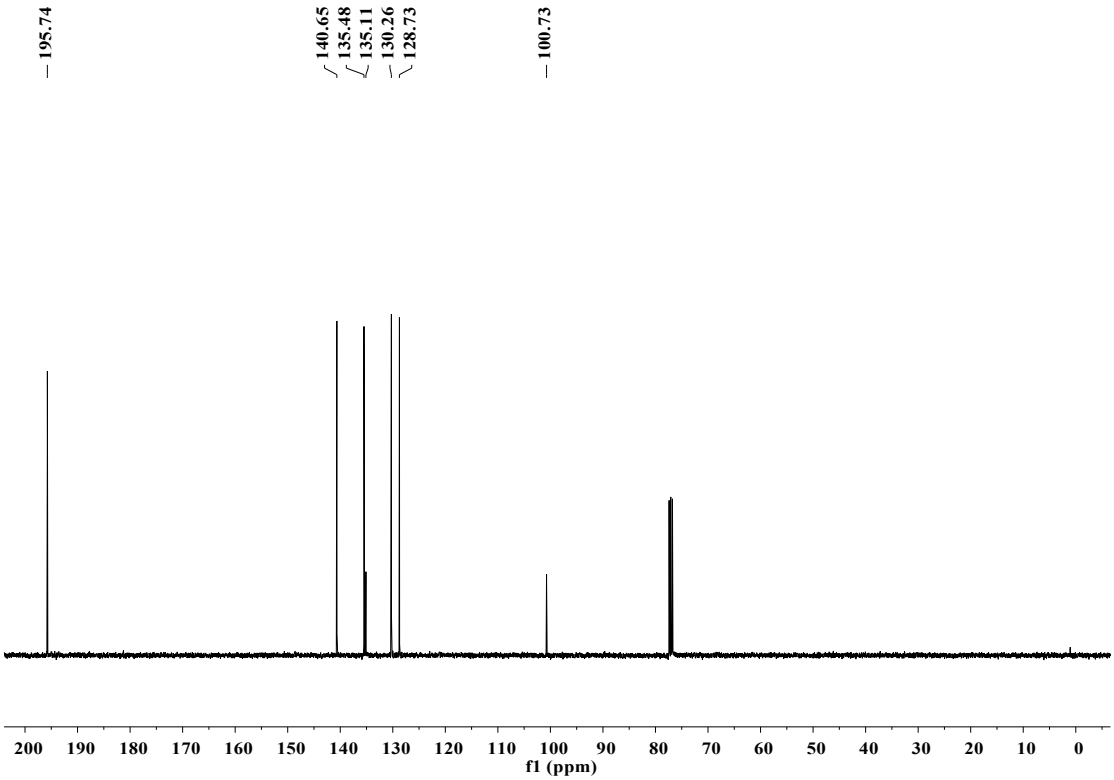


Figure S60: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2-iodobenzaldehyde (4v).

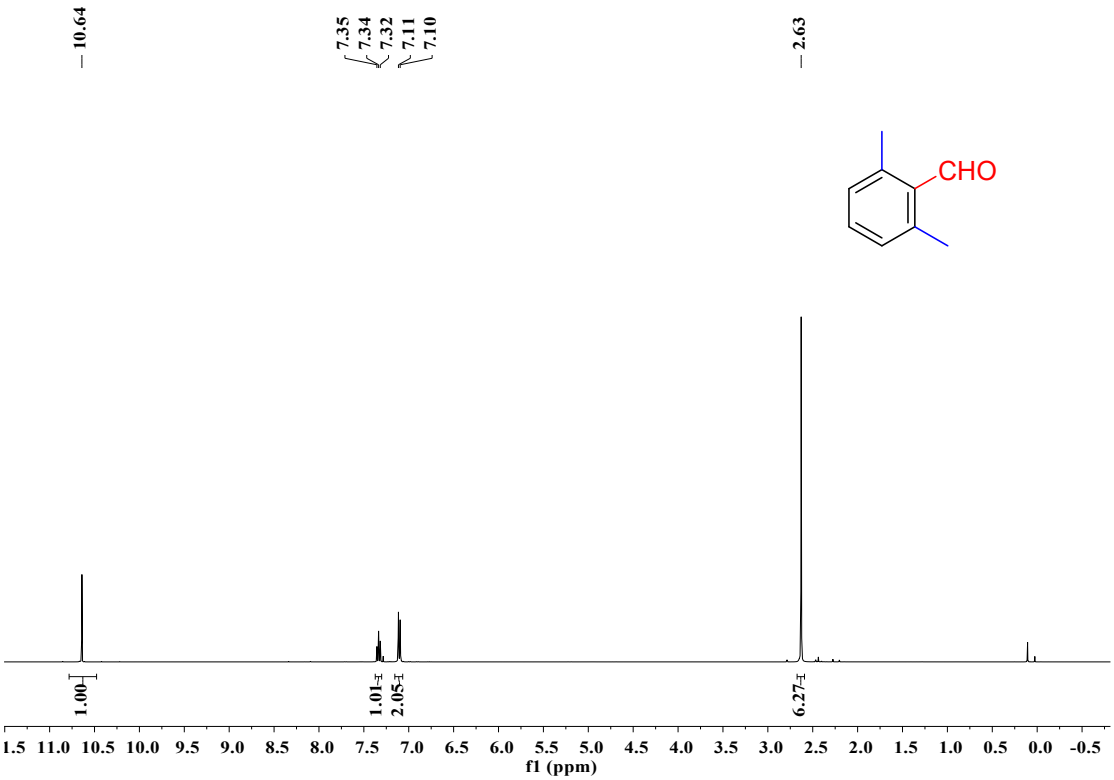


Figure S61: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2,6-Dimethylbenzaldehyde (4w).

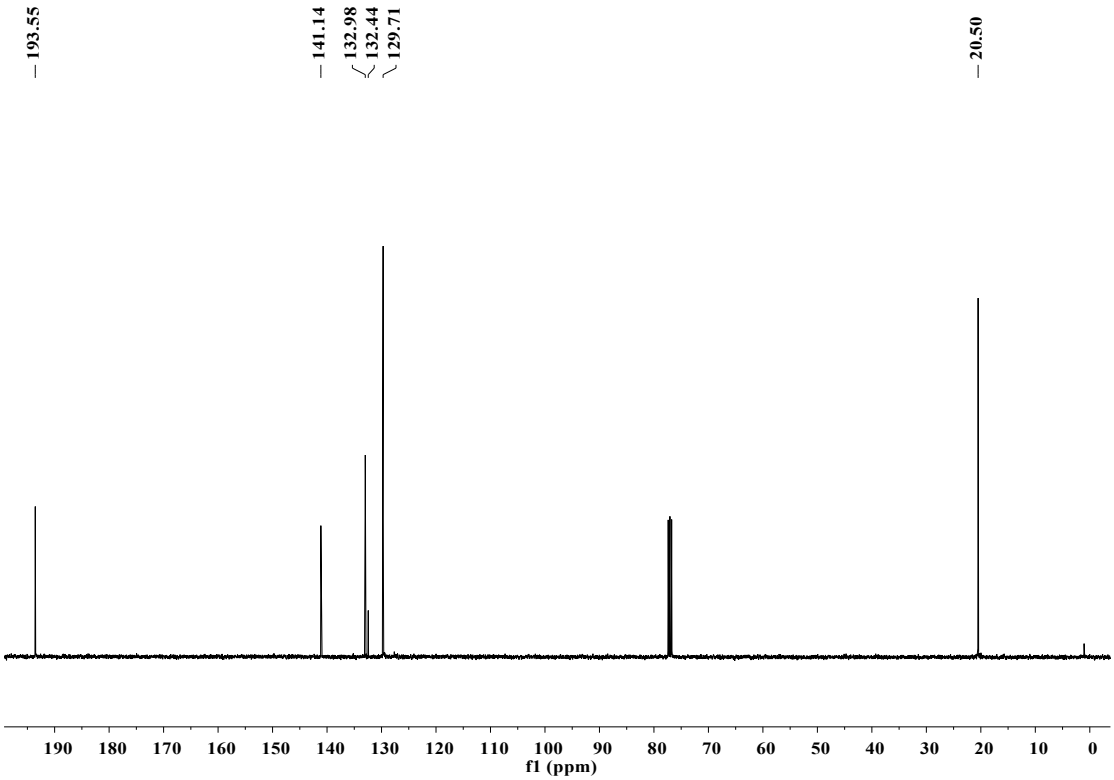


Figure S62: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2,6-Dimethylbenzaldehyde (4w).

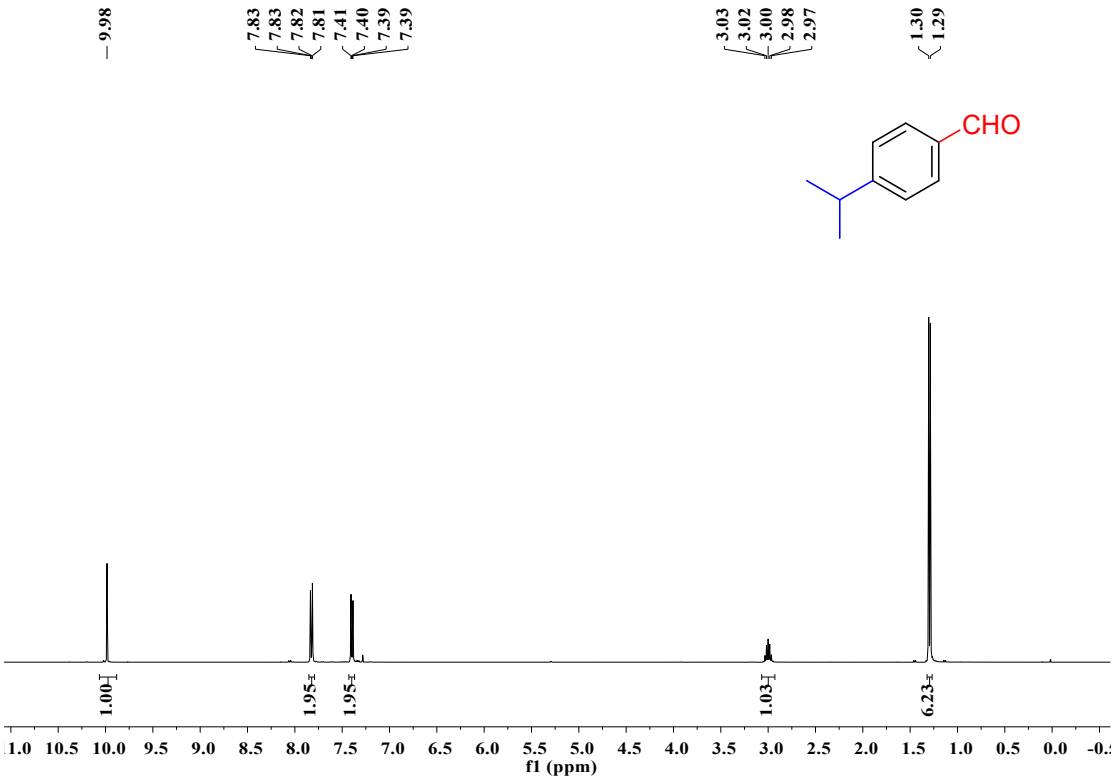


Figure S63: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 4-isopropylbenzaldehyde (4x).

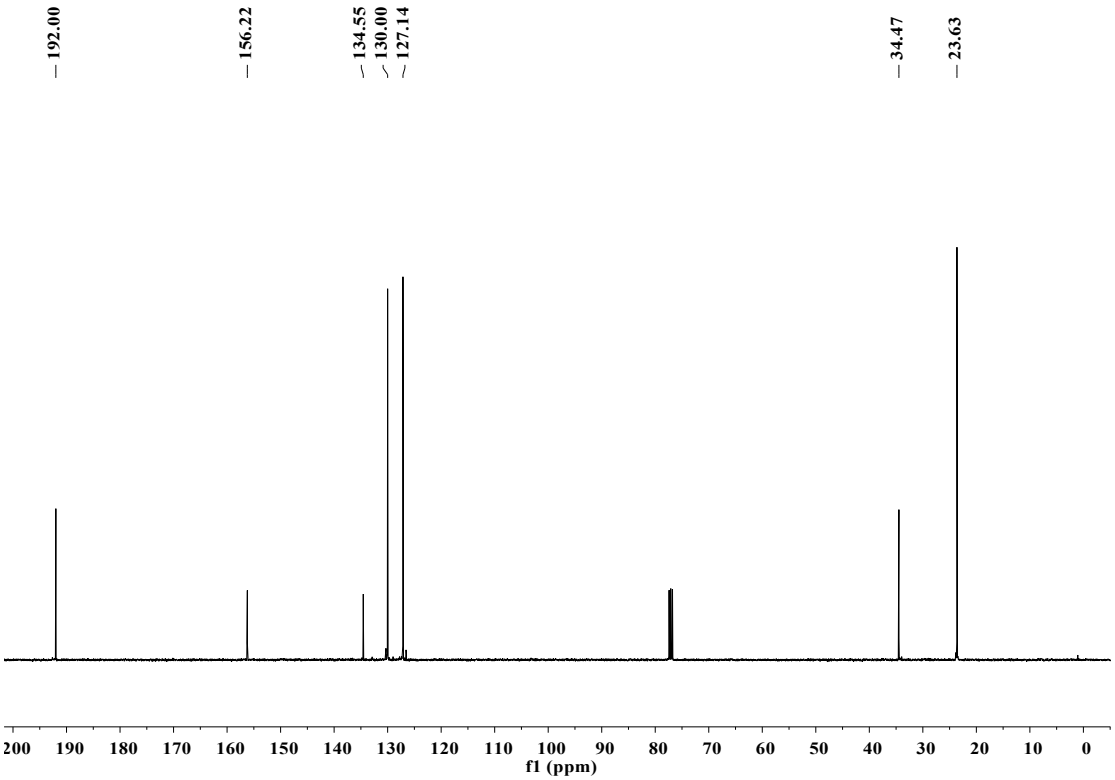


Figure S64: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 4-isopropylbenzaldehyde (4x).

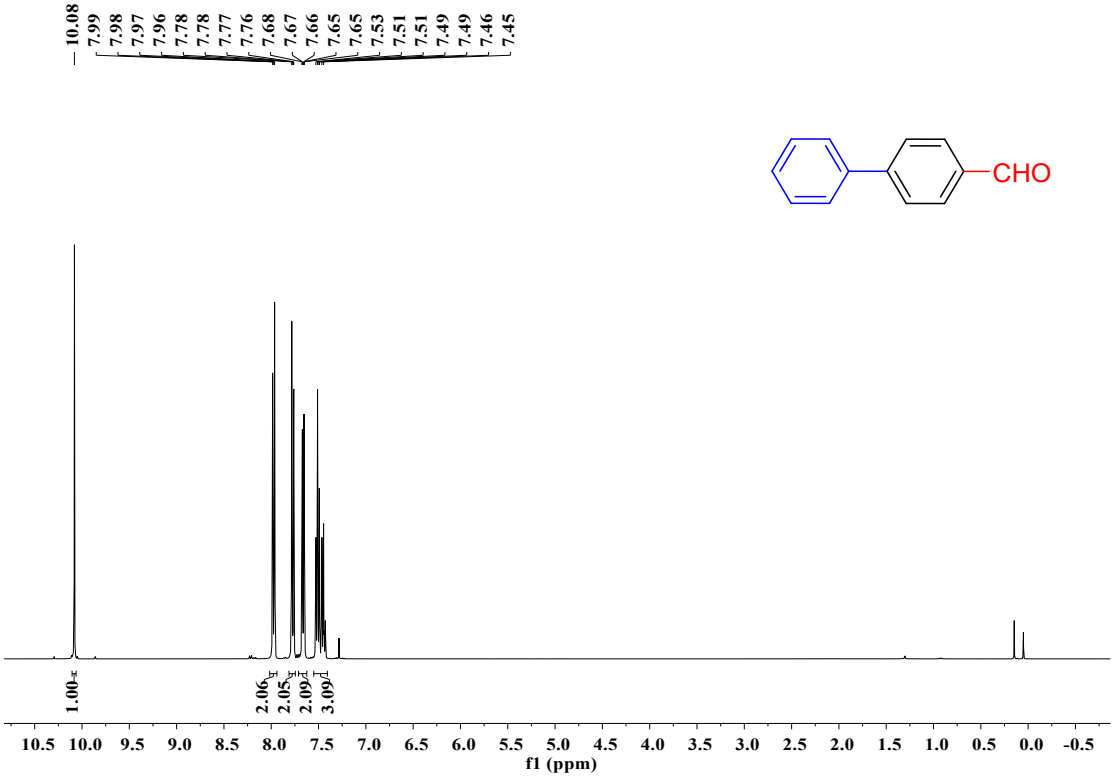


Figure S65: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of biphenyl-4-carboxaldehyde (4y).

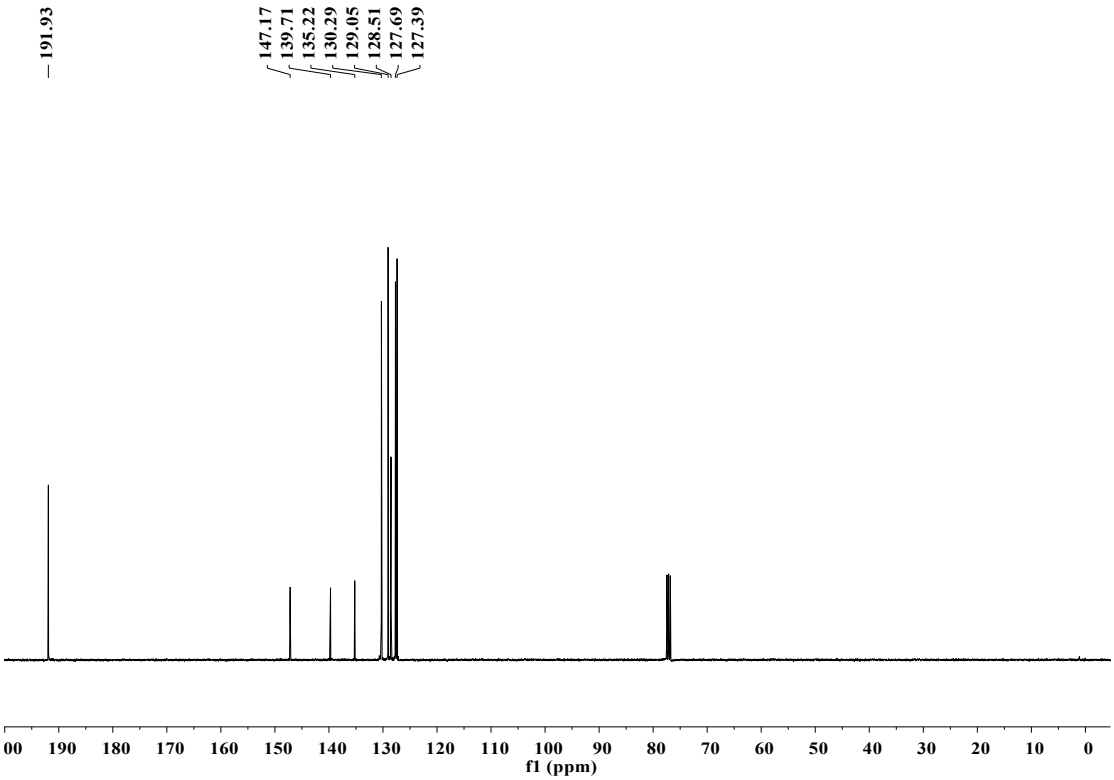


Figure S66: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of biphenyl-4-carboxaldehyde (4y).

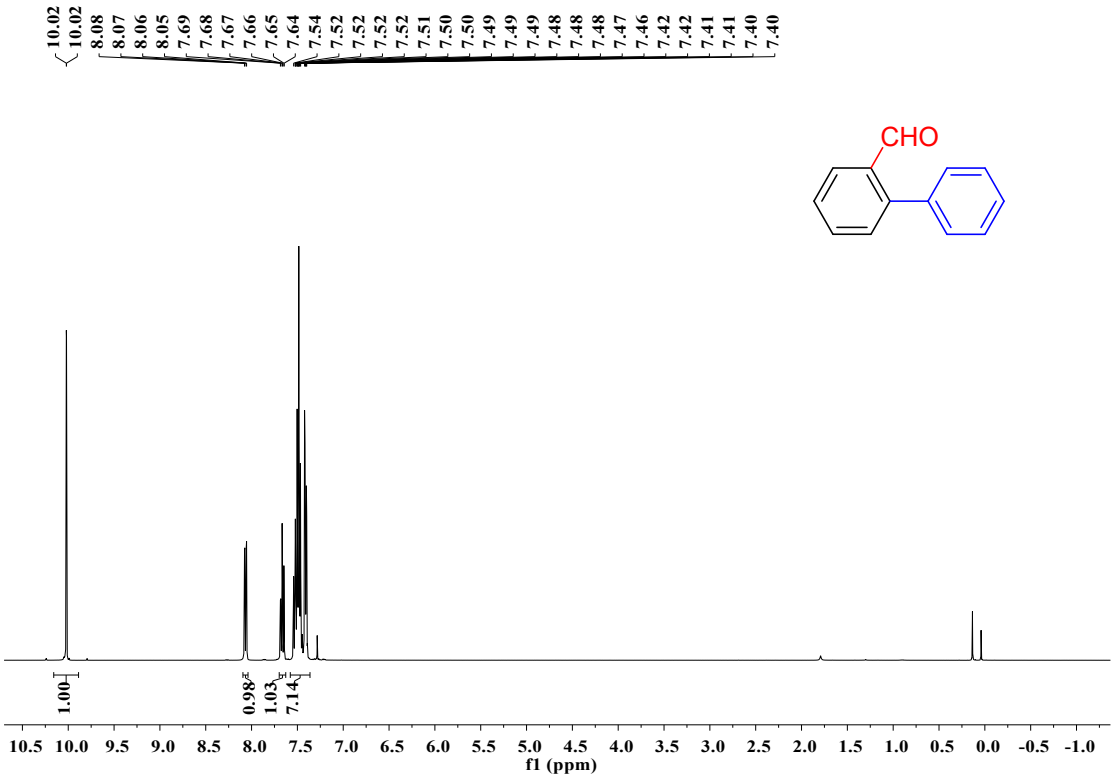


Figure S67: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of biphenyl-2-carboxaldehyde (4z).

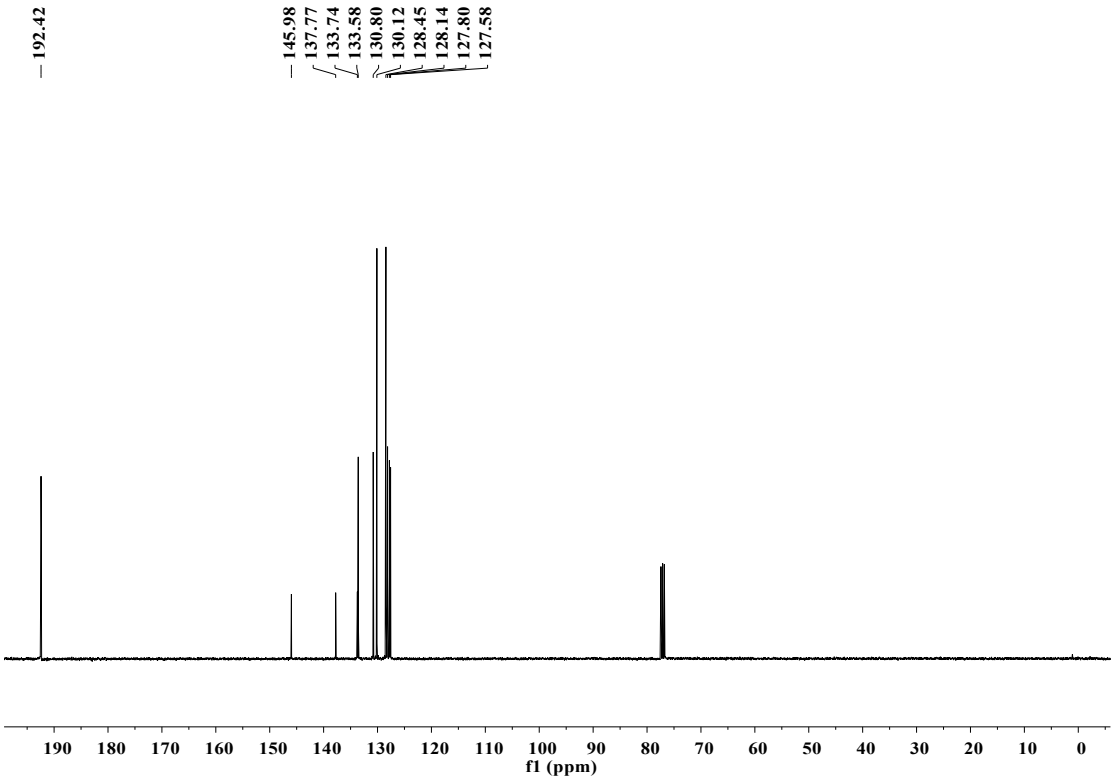


Figure S68: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of biphenyl-2-carboxaldehyde (4z).

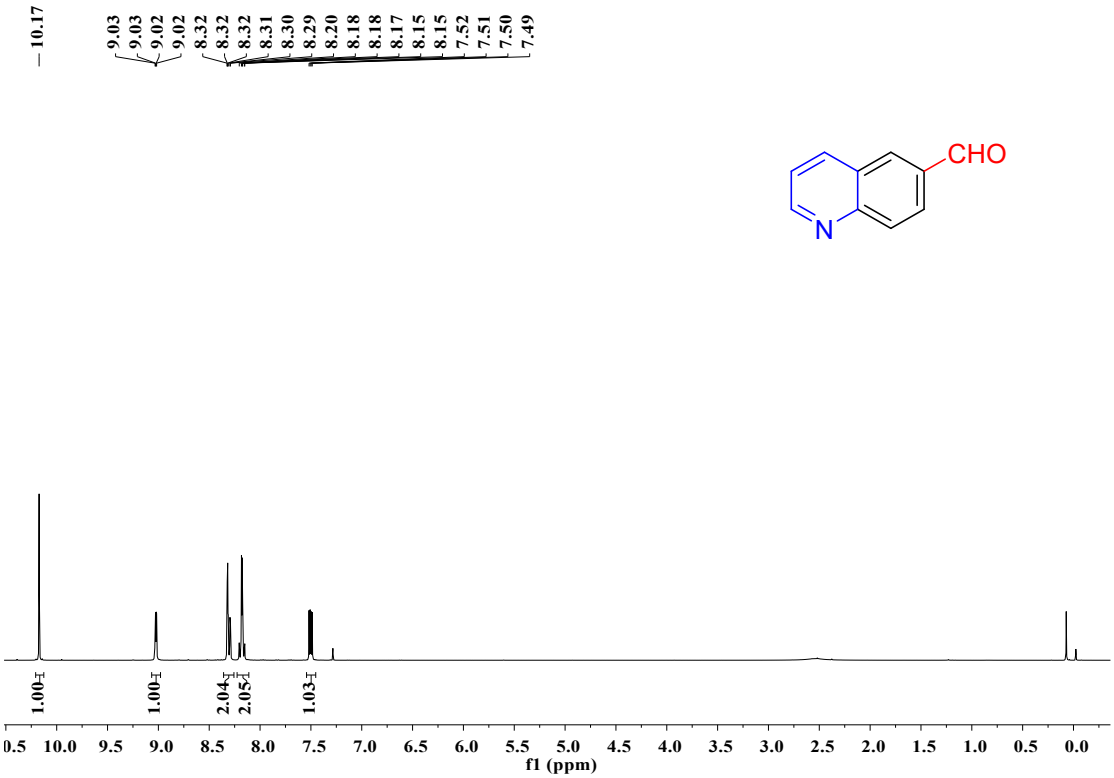


Figure S69: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of quinoline-6-formaldehyde (4aa).

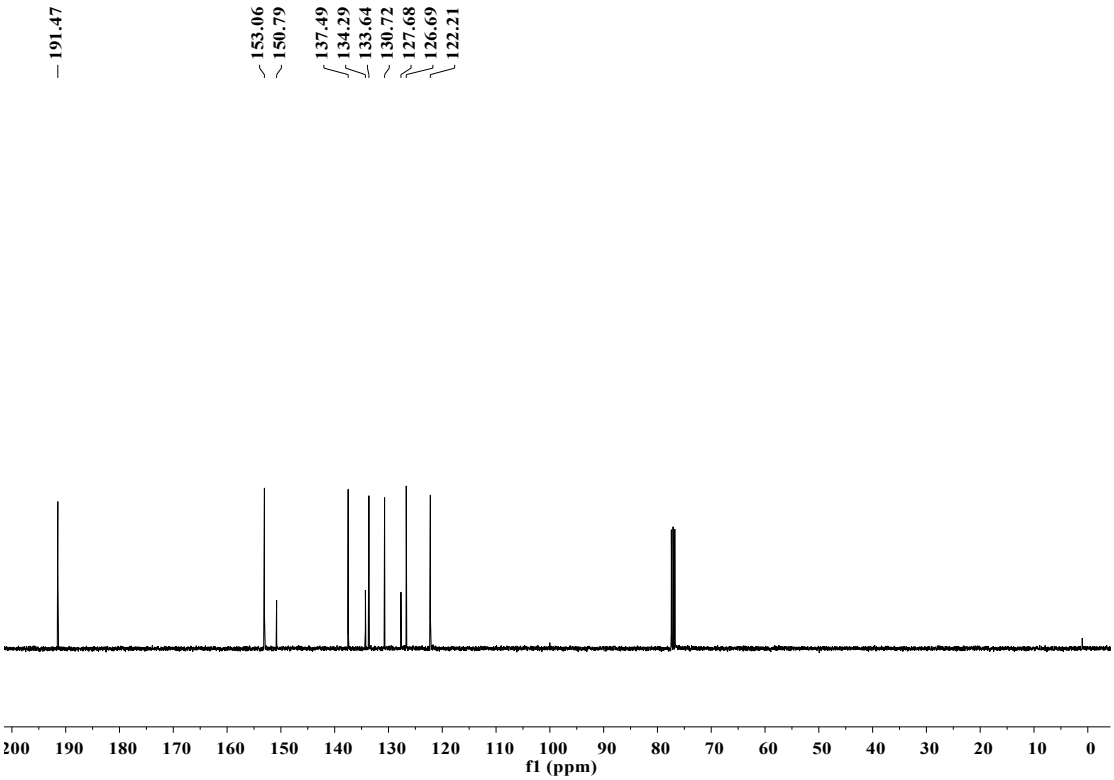


Figure S70: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of quinoline-6-formaldehyde (4aa).

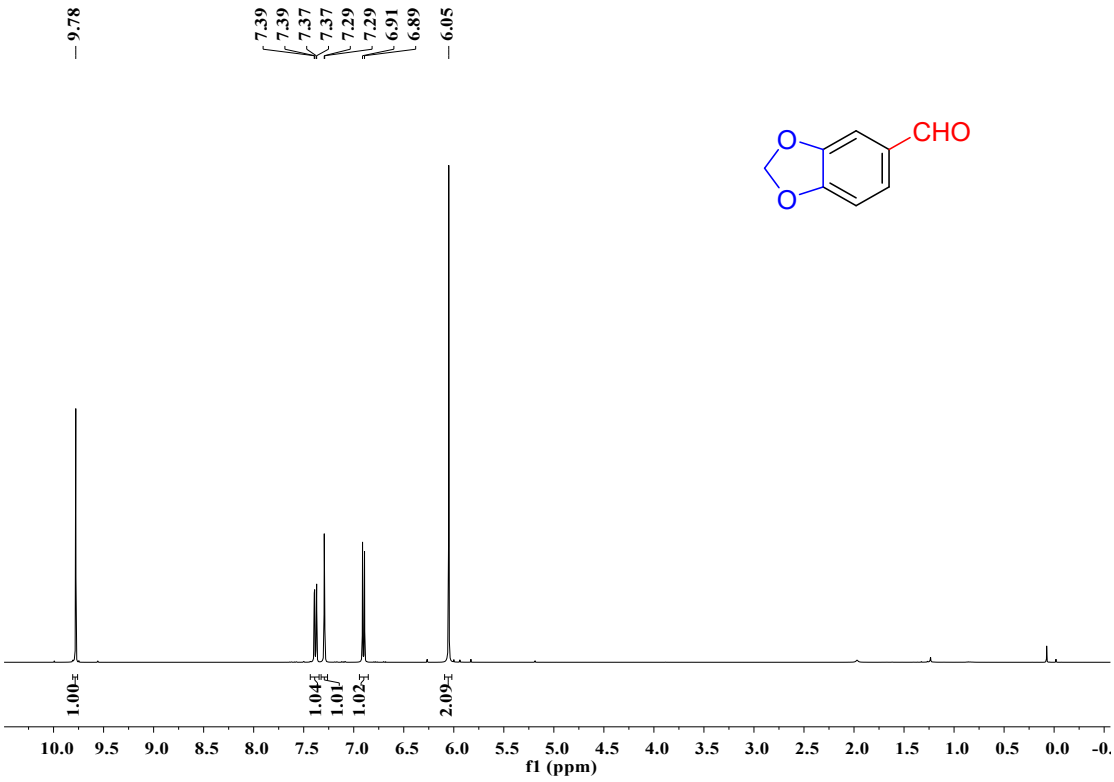


Figure S71: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of piperonal (4ab).

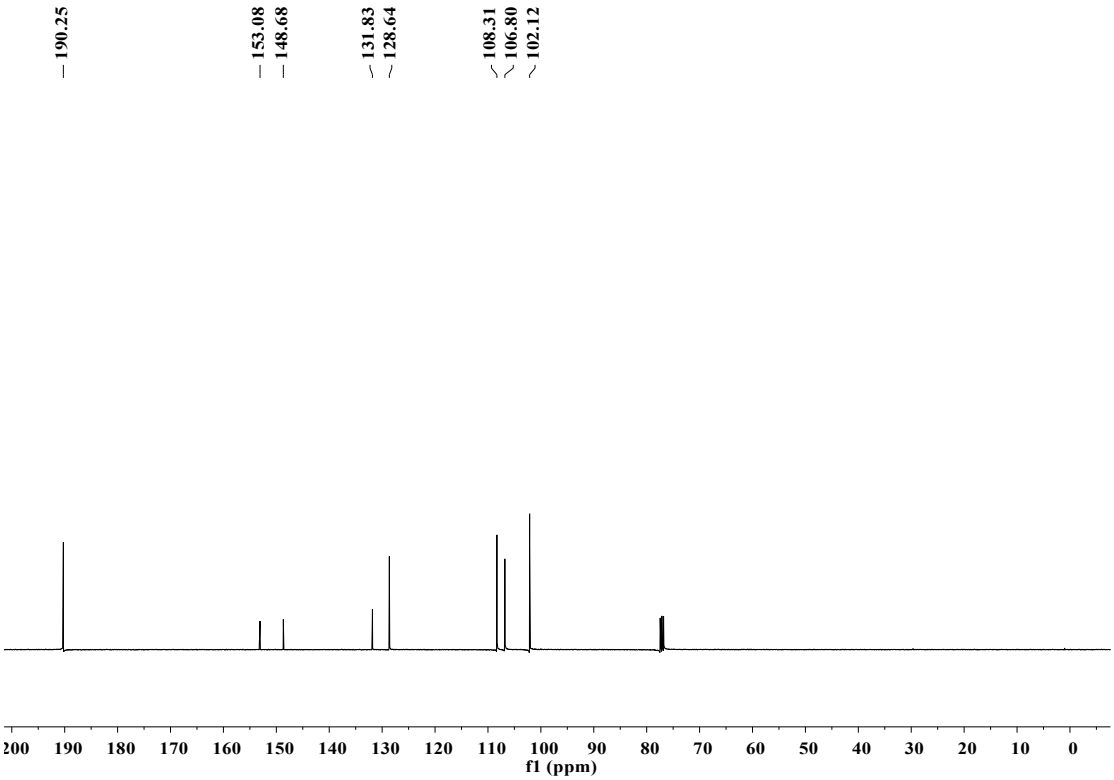


Figure S72: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of piperonal (4ab).

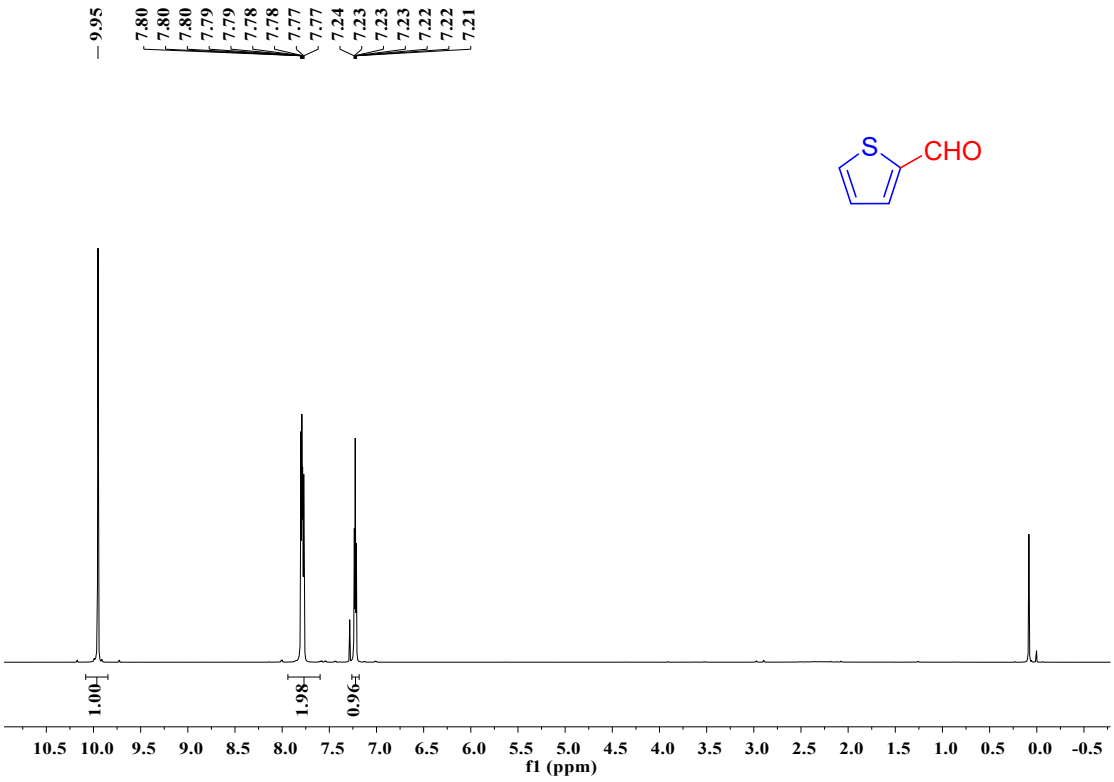


Figure S73: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of thiophene-2-Carboxaldehyde (4ac).

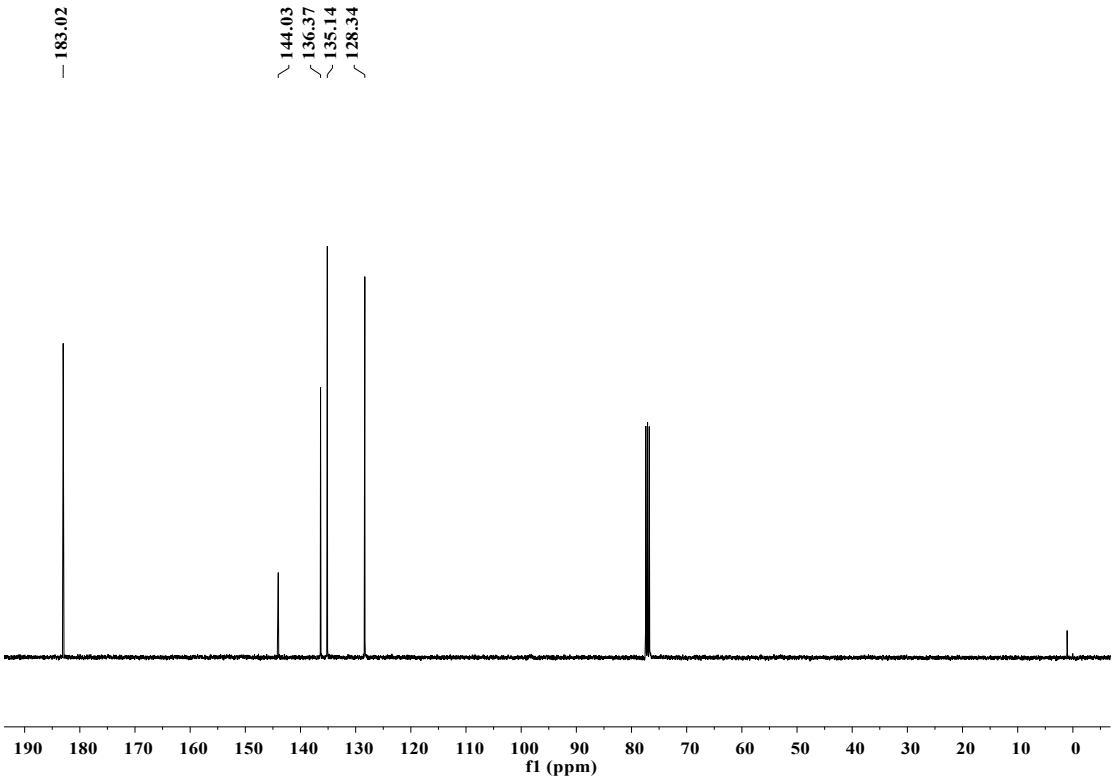


Figure S74: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of thiophene-2-Carboxaldehyde (4ac).

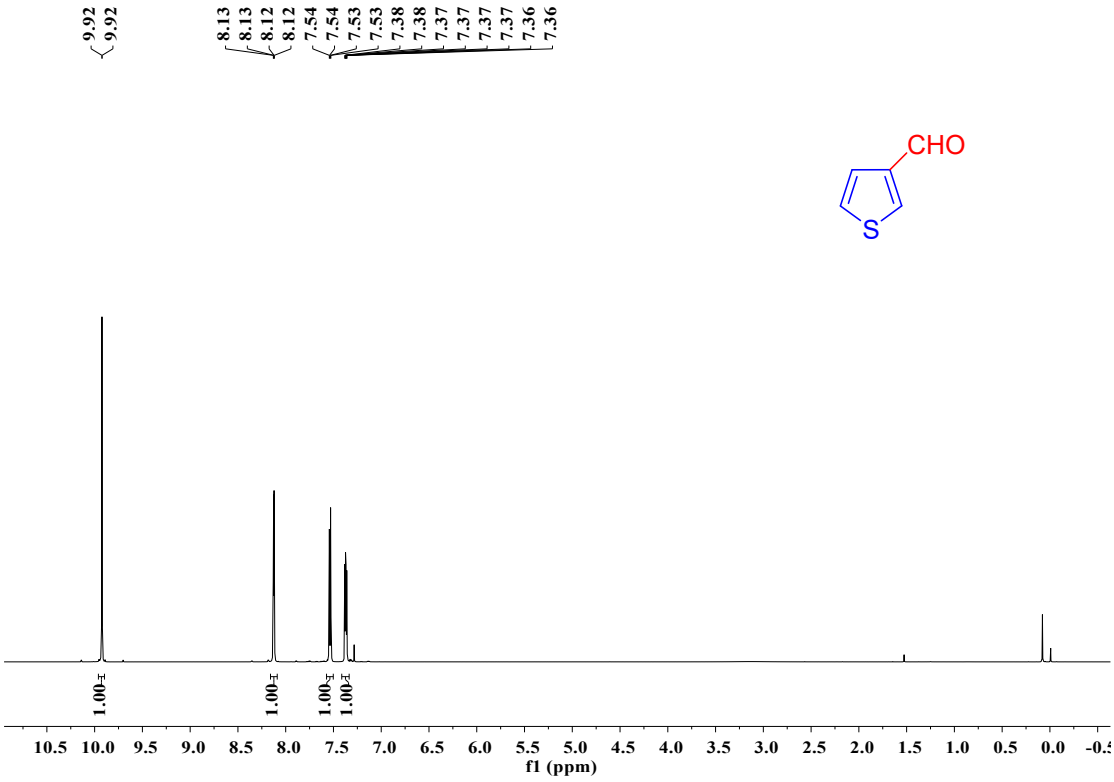


Figure S75: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of thiophene-3-Carboxaldehyde (4ad).

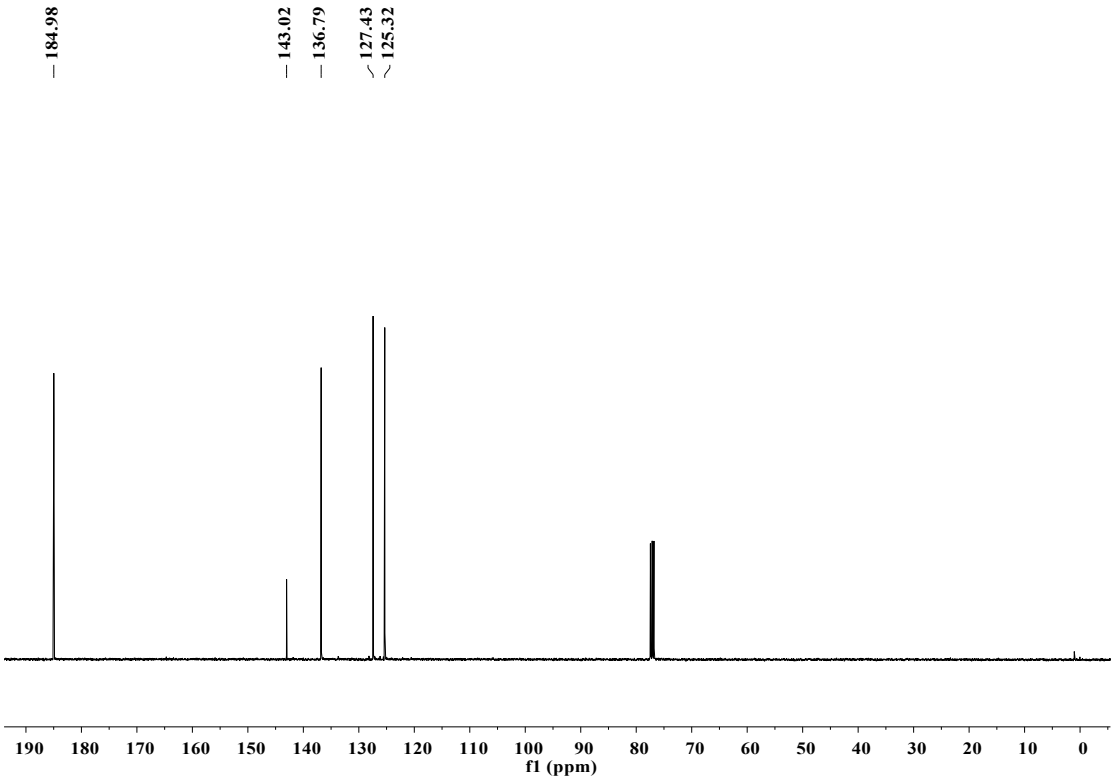


Figure S76: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of thiophene-3-Carboxaldehyde (4ad).

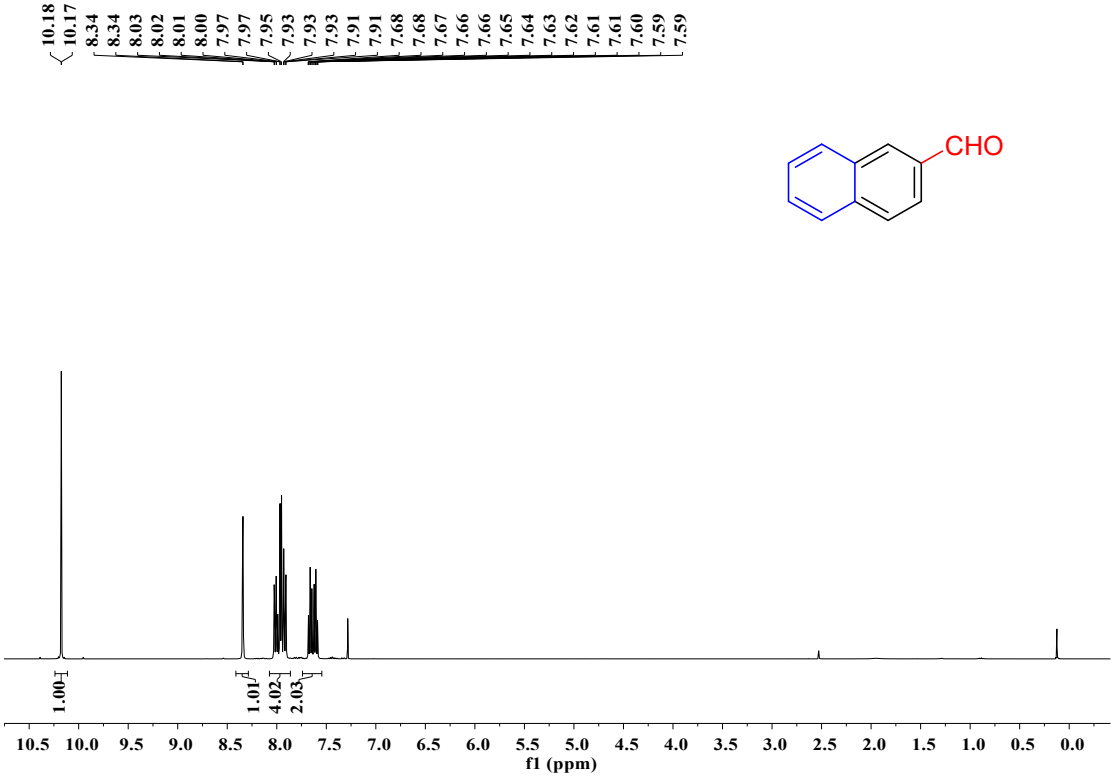


Figure S77: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 2-naphthaldehyde (4ae).

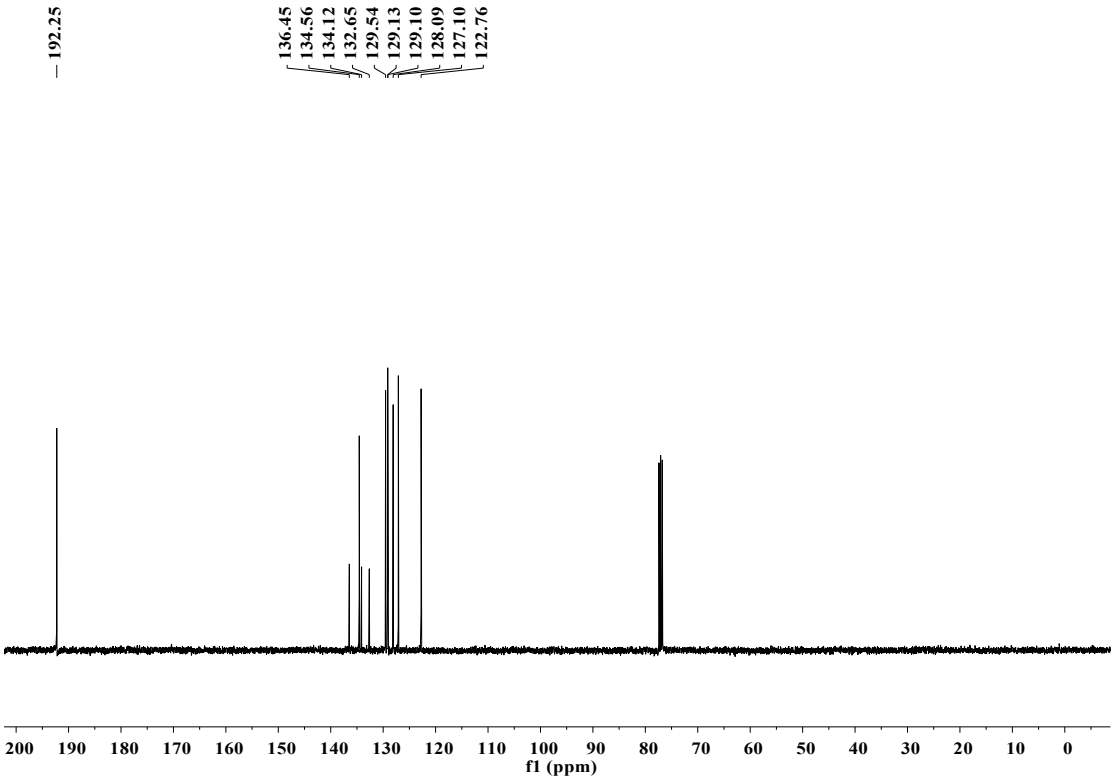


Figure S78: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 2-naphthaldehyde (4ae).

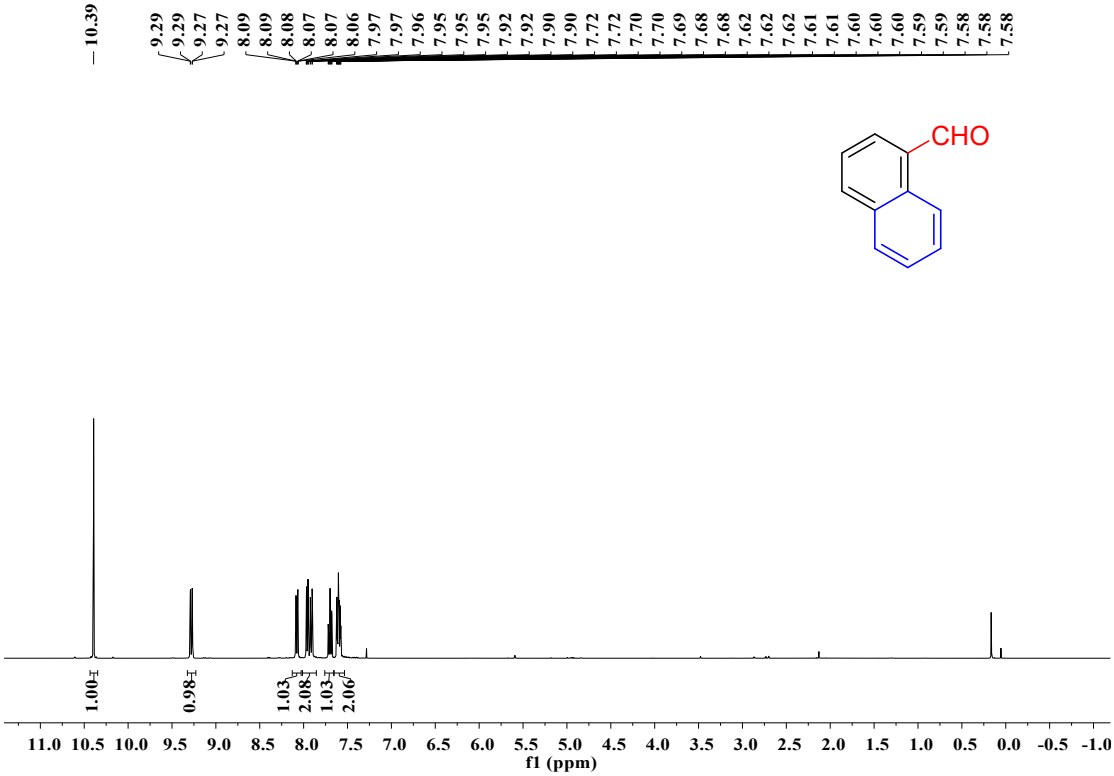


Figure S79: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 1-naphthaldehyde (4af).

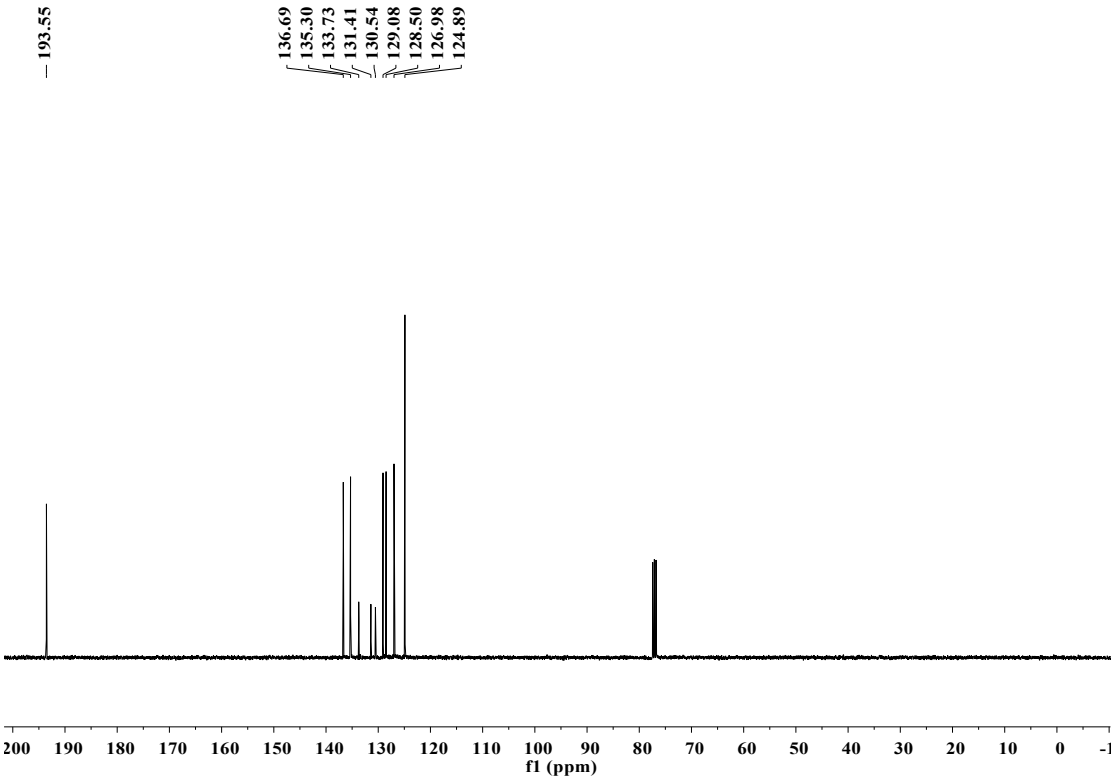


Figure S80: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 1-naphthaldehyde (4af).

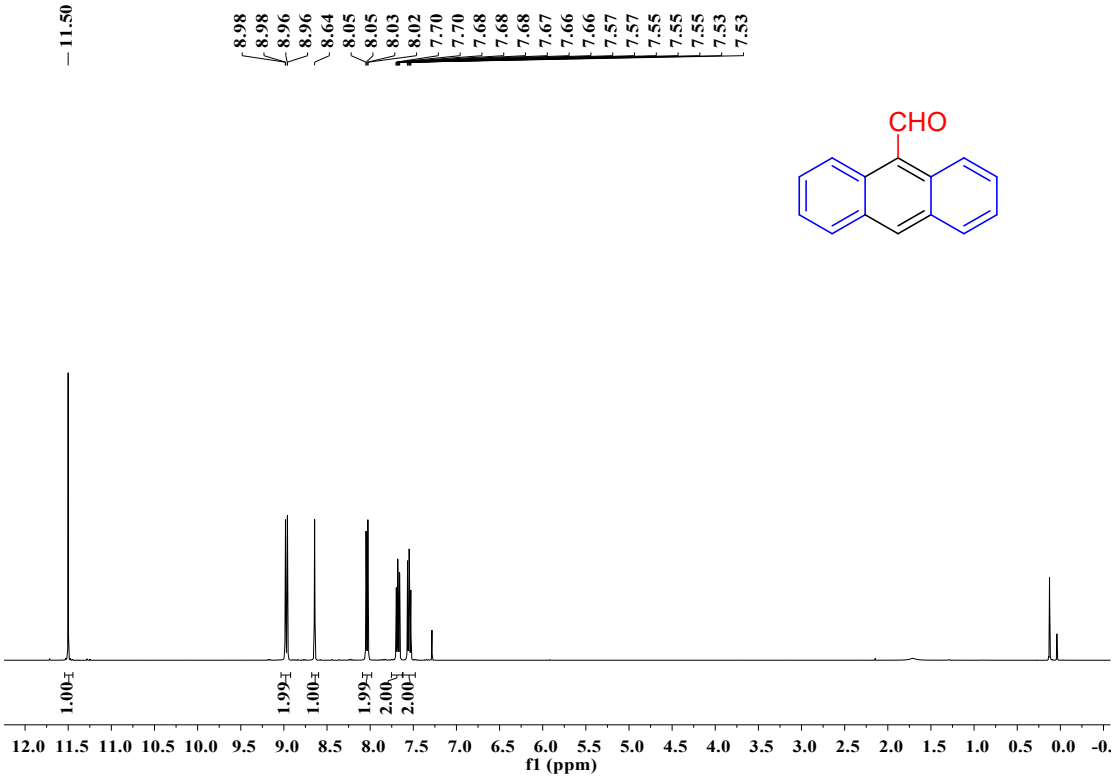


Figure S81: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 9-anthracenecarboxaldehyde (4ag).

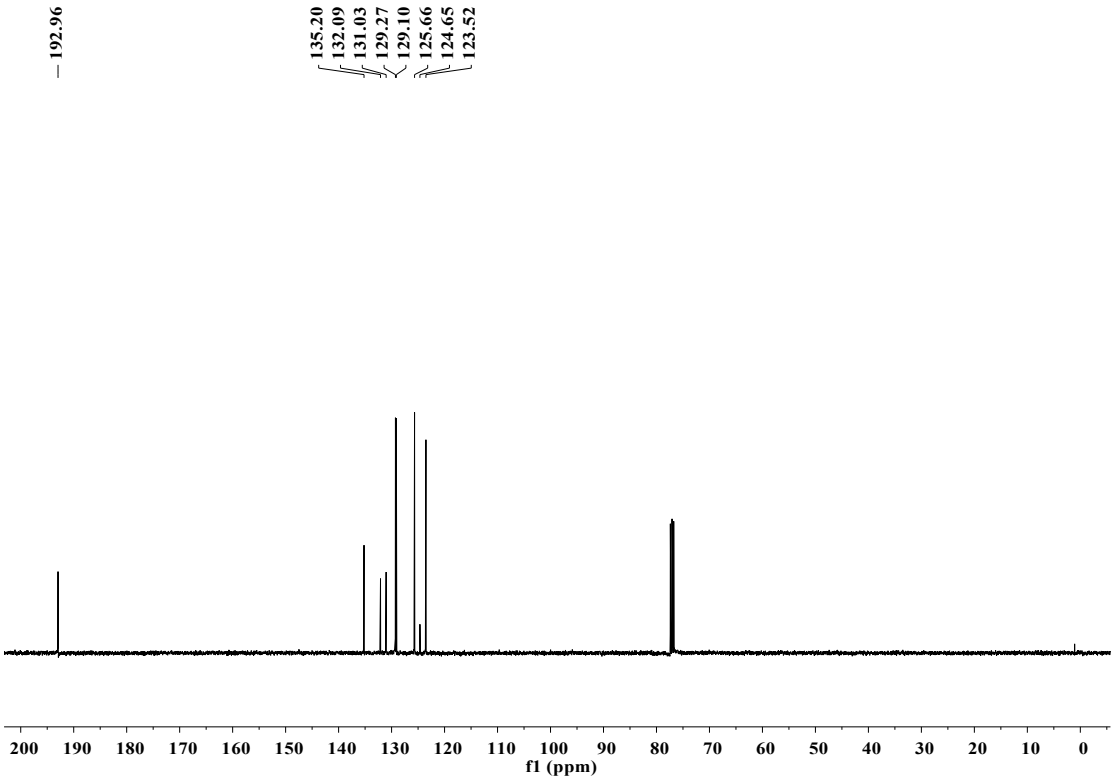


Figure S82: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of 9-anthracenecarboxaldehyde (4ag).

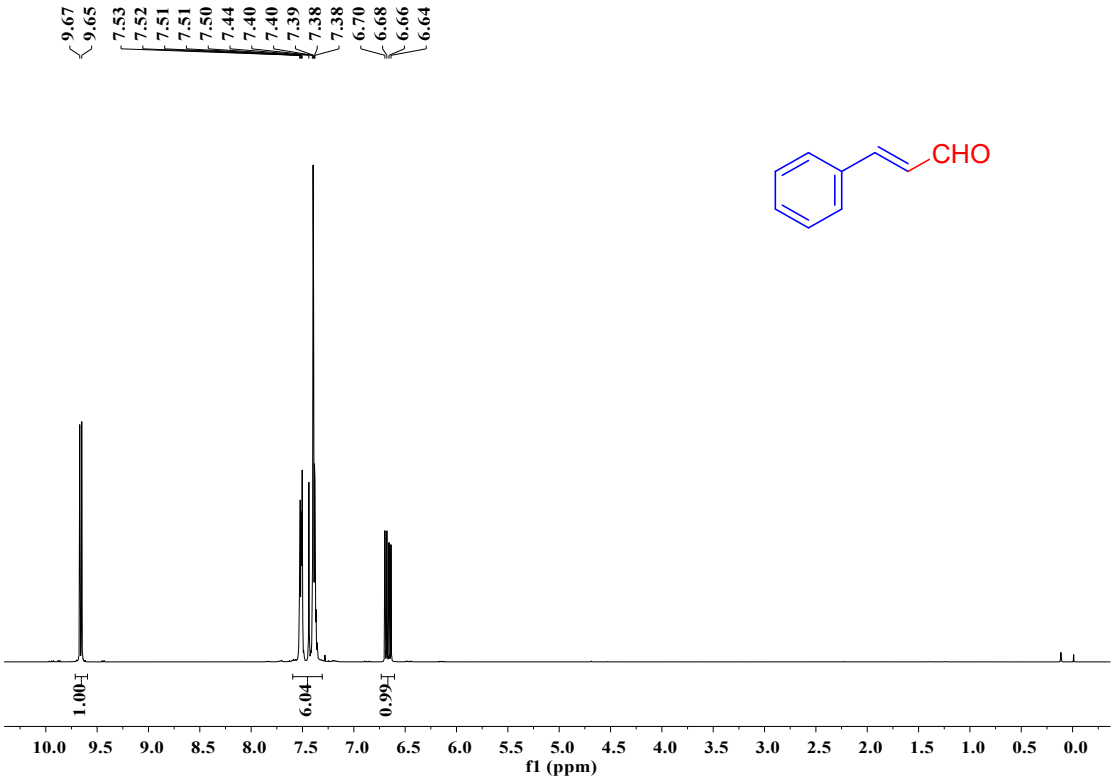


Figure S83: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of cinnamaldehyde (4ah).

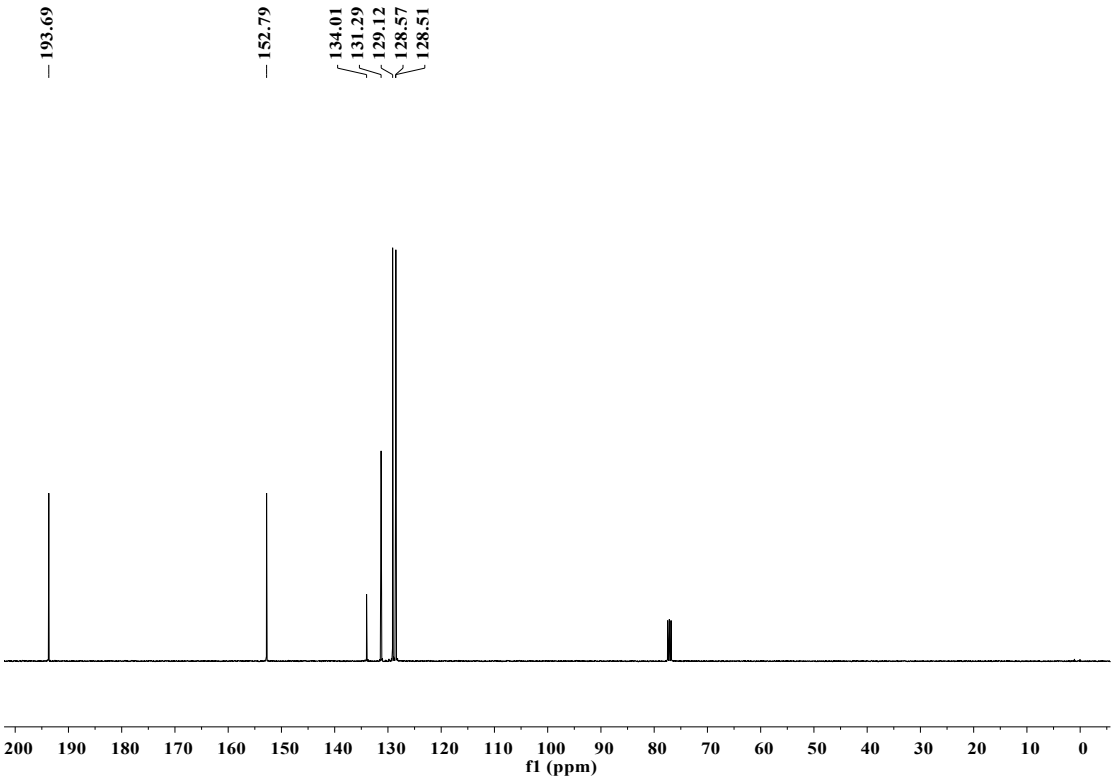


Figure S84: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of cinnamaldehyde (4ah).

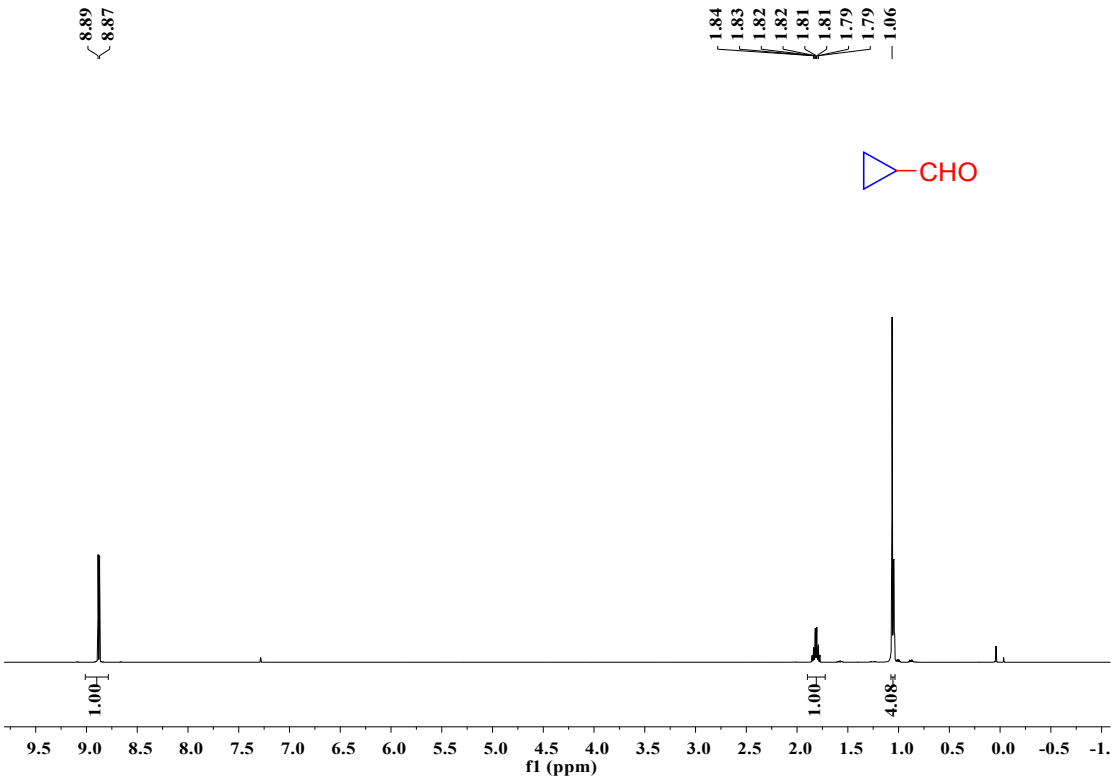


Figure S85: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of cyclopropanecarboxaldehyde (4ai).

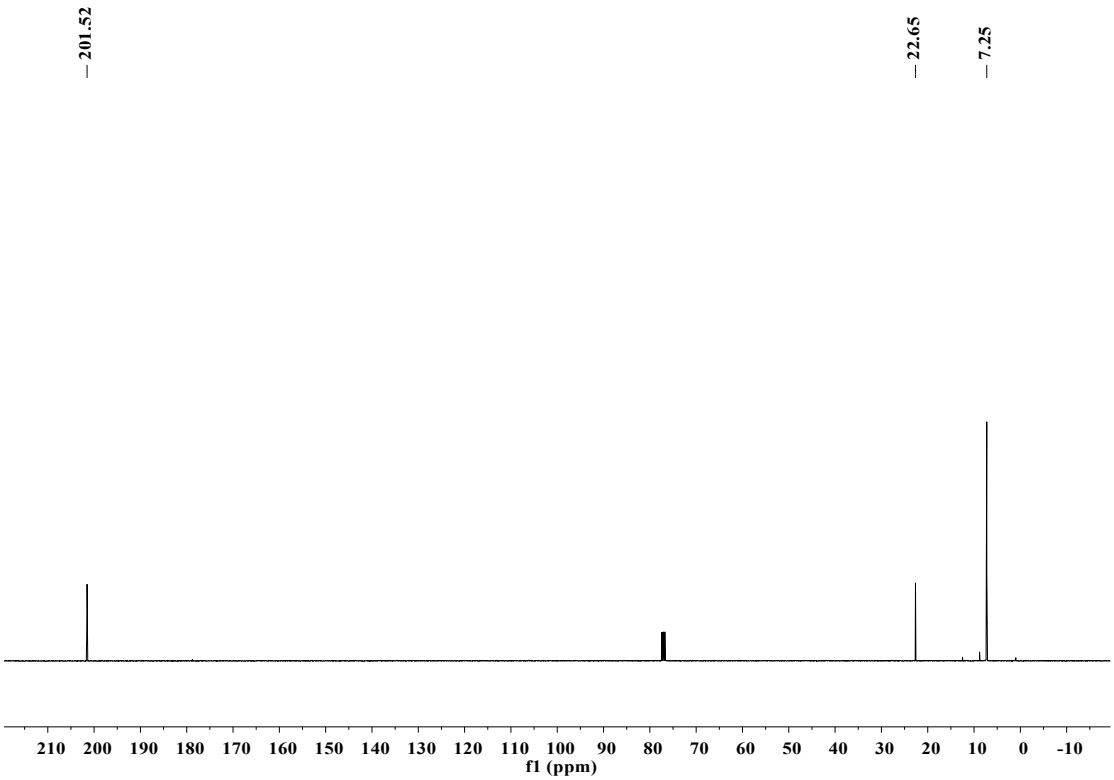


Figure S86: $^{13}C\{^{1}H\}$ NMR spectrum of (101 MHz, $CDCl_3$) of cyclopropanecarboxaldehyde (4ai).

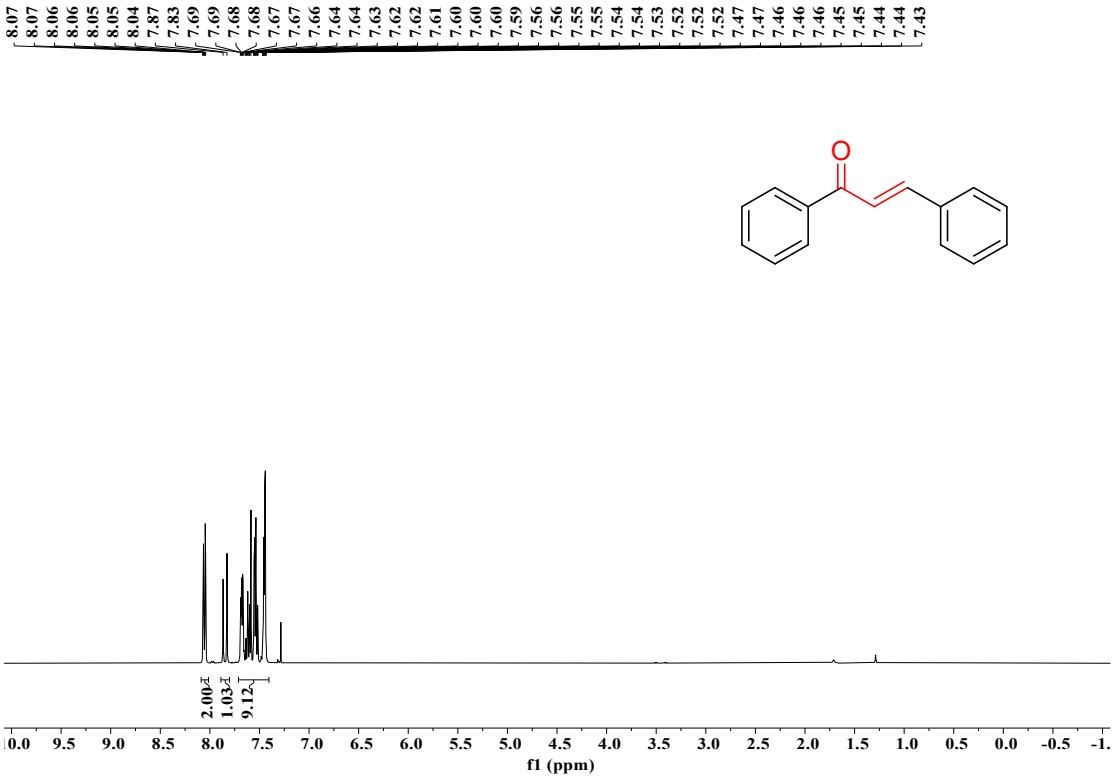


Figure S87: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-chalcone (5c[a]).

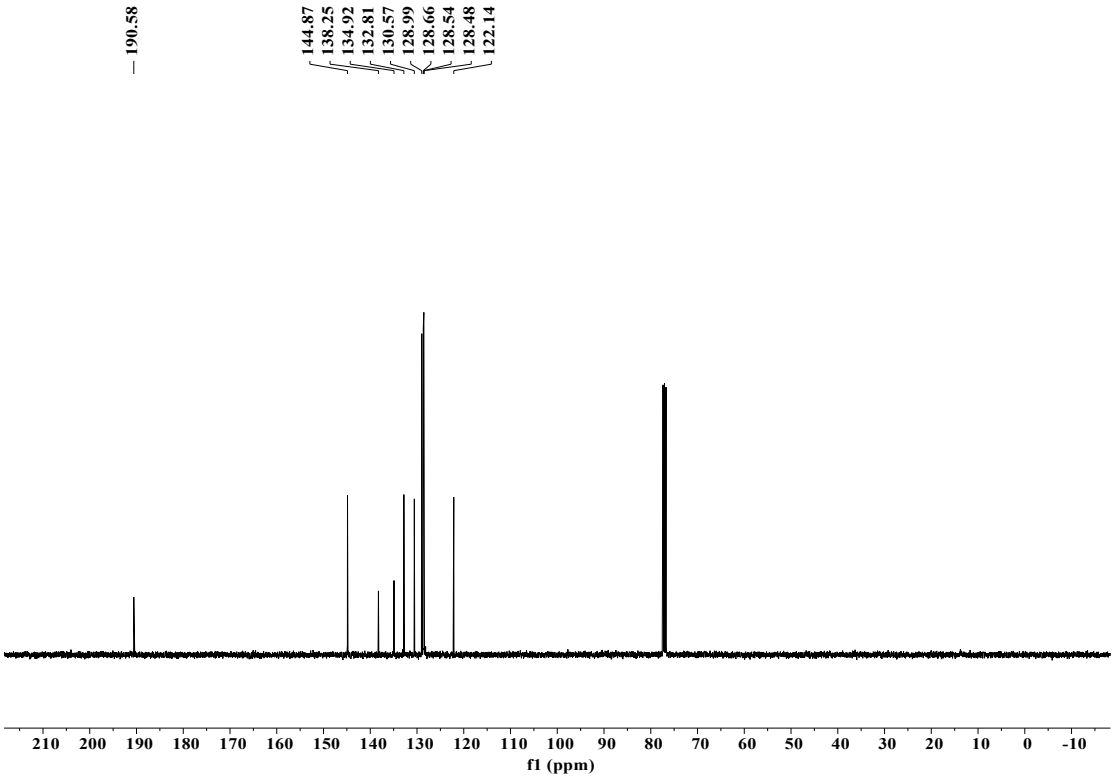


Figure S88: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-chalcone (5c[a]).

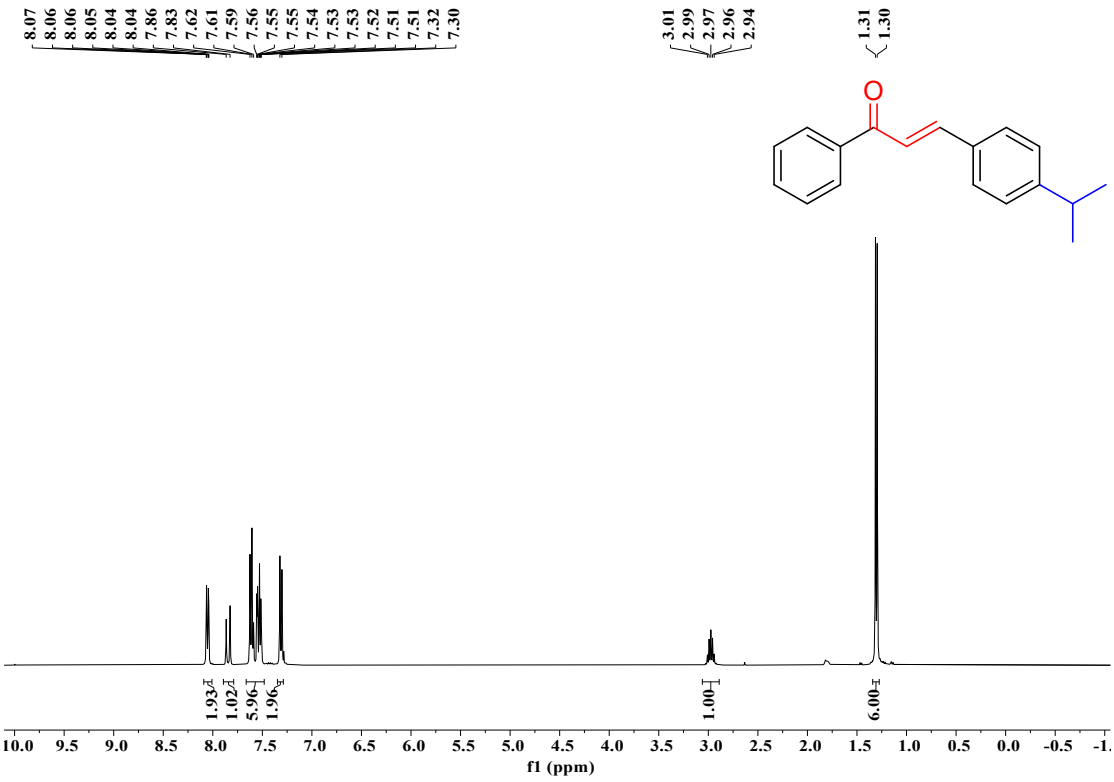


Figure S89: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(4-isopropylphenyl)-1-phenylprop-2-en-1-one (5d[a]).

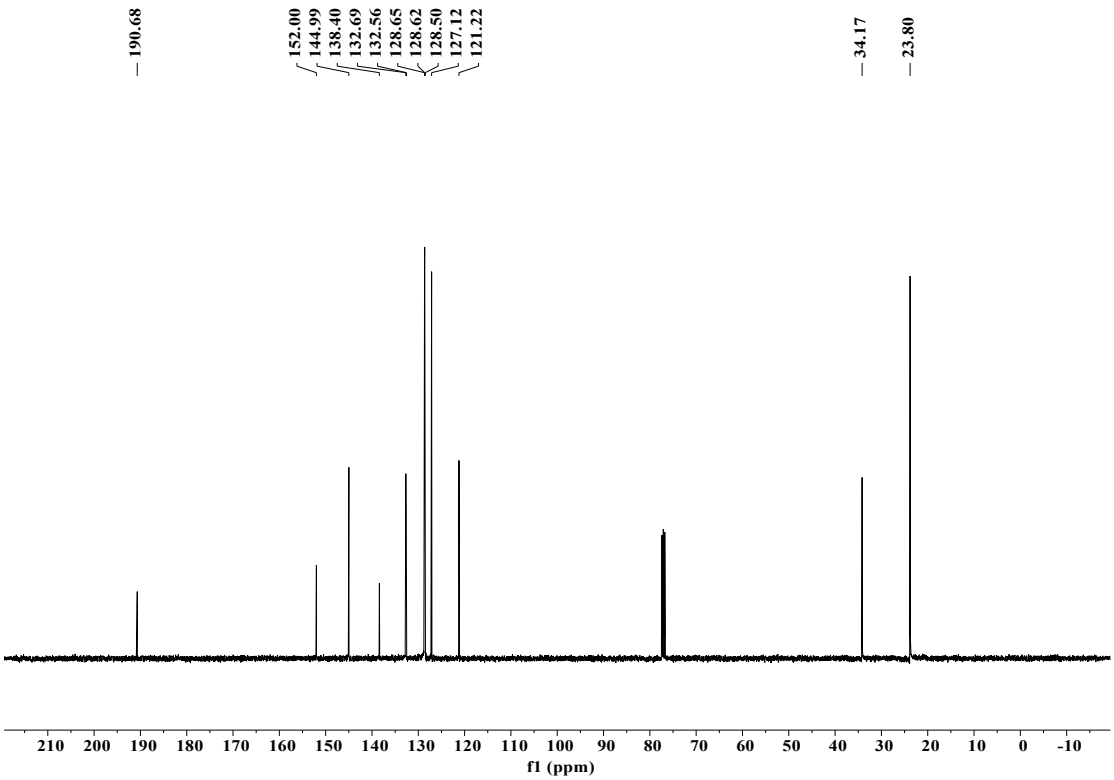


Figure S90: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(4-isopropylphenyl)-1-phenylprop-2-en-1-one (5d[a]).

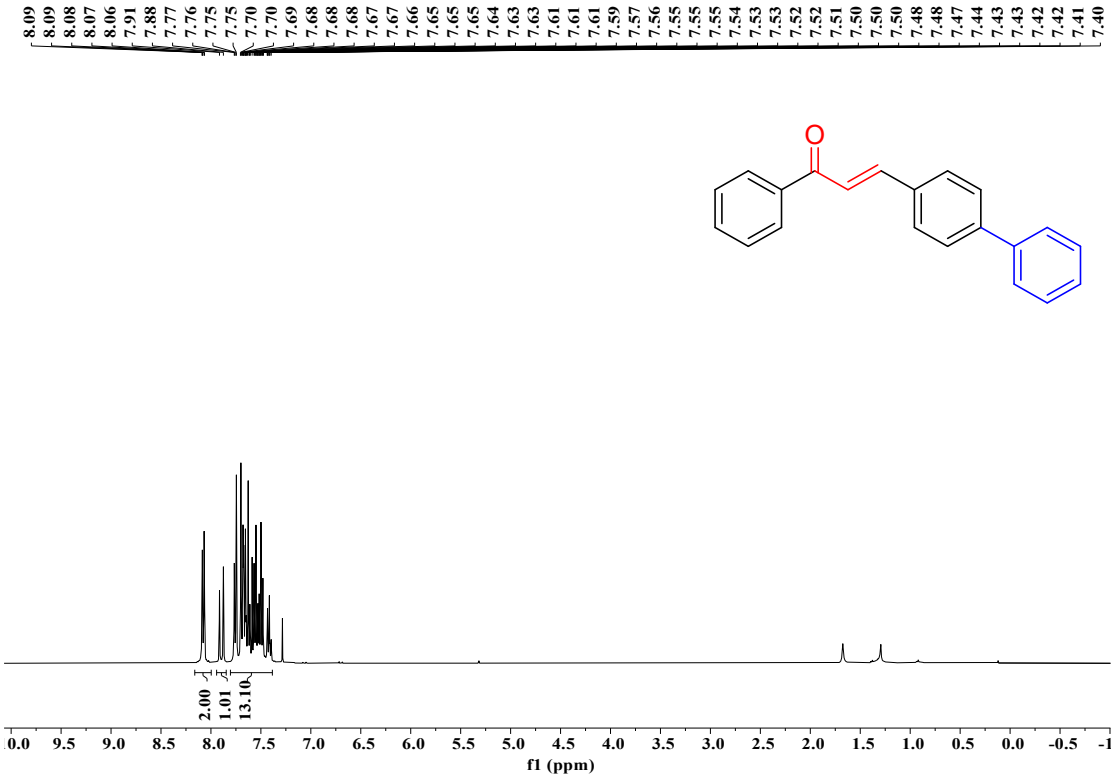


Figure S91: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-([1,1'-biphenyl]-4-yl)-1-phenylprop-2-en-1-one (5e[a]).

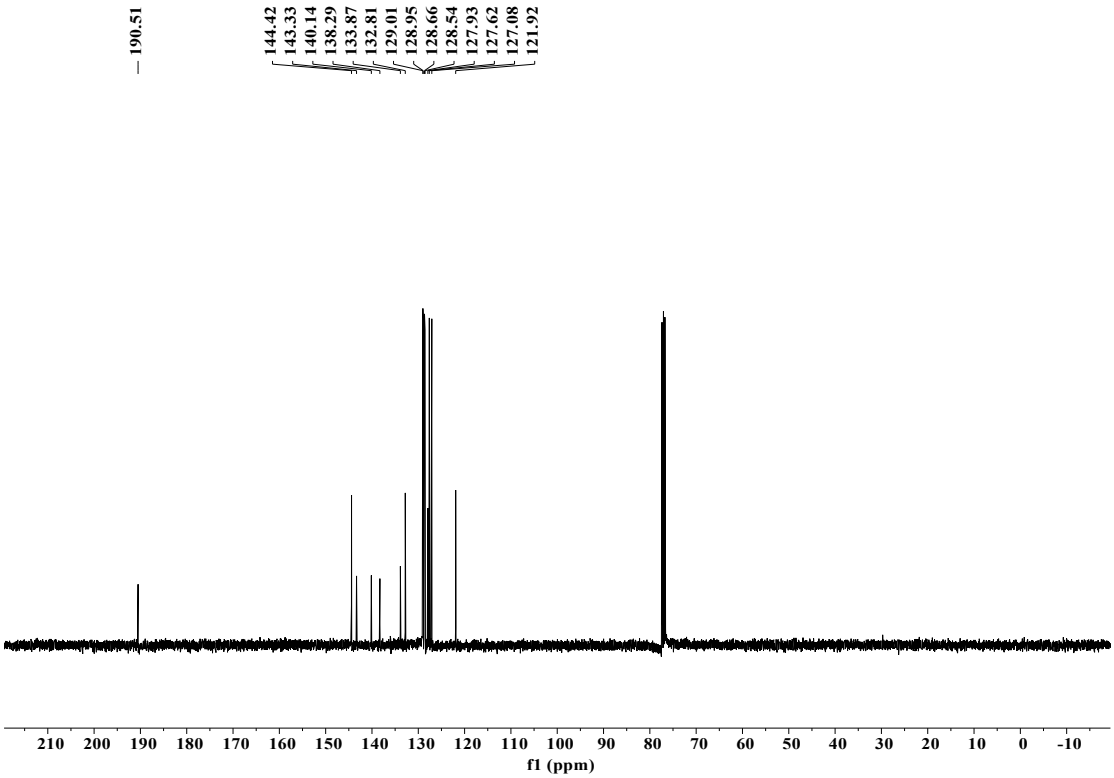


Figure S92: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-([1,1'-biphenyl]-4-yl)-1-phenylprop-2-en-1-one (5e[a]).

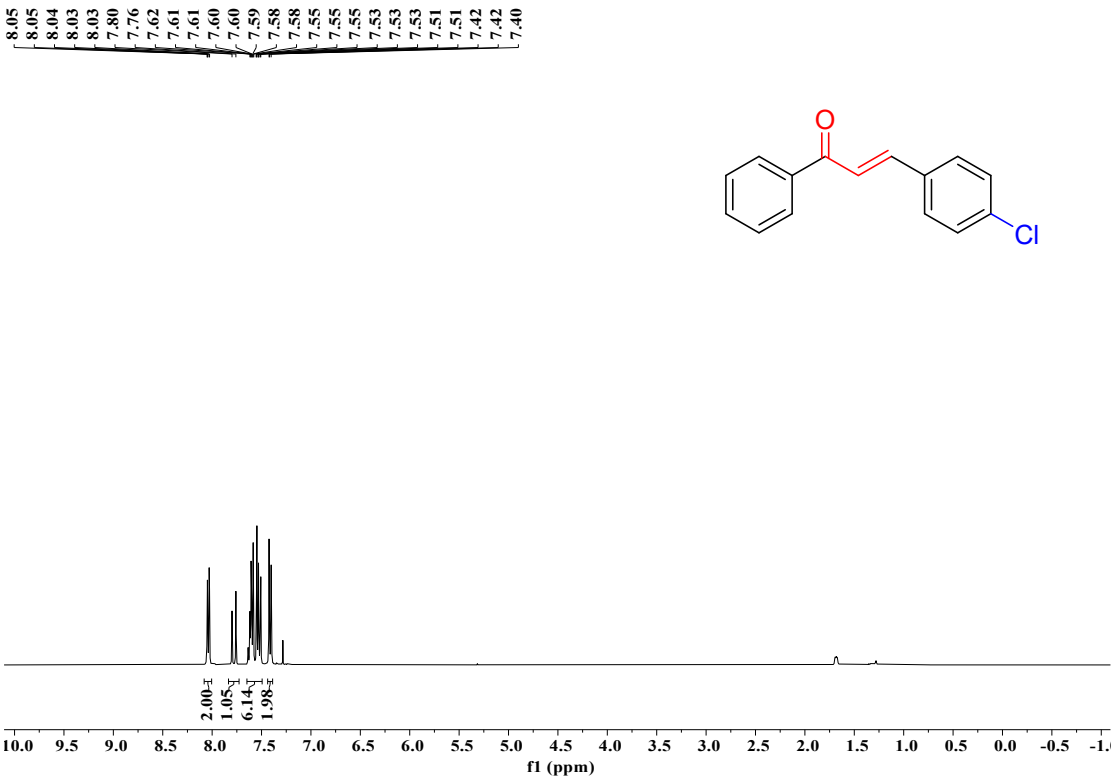


Figure S93: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(4-chlorophenyl)-1-phenylprop-2-en-1-one (5f[a]).

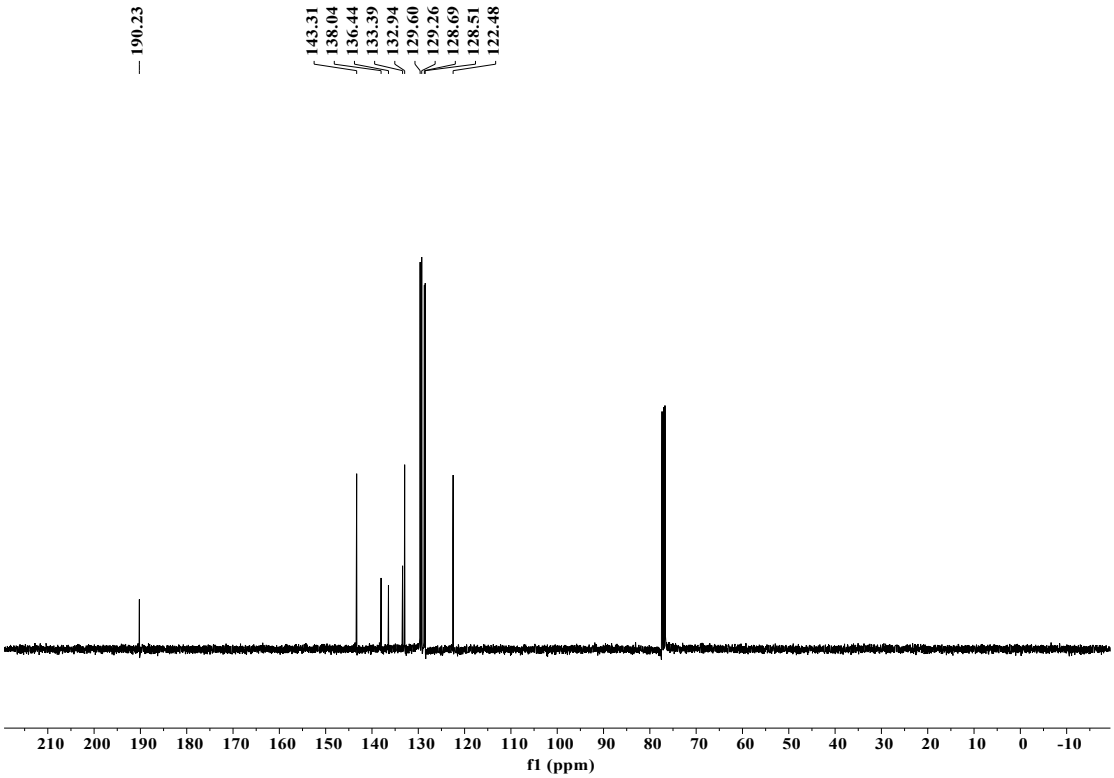


Figure S94: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(4-chlorophenyl)-1-phenylprop-2-en-1-one (5f[a]).

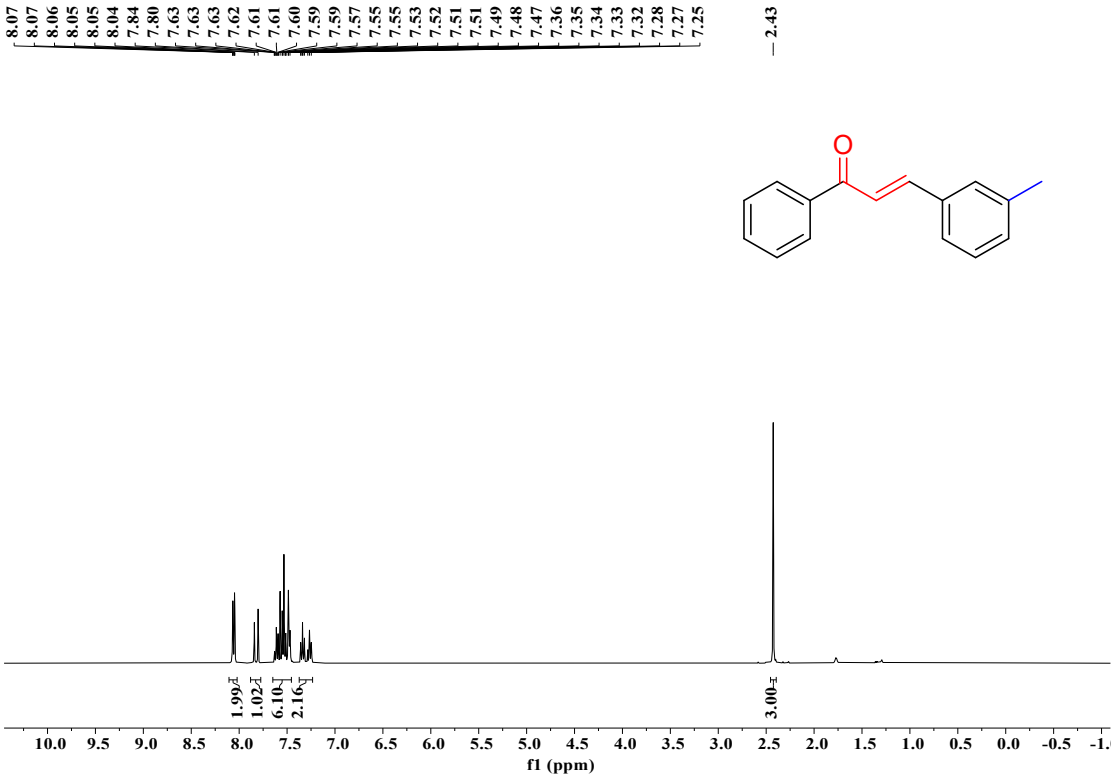


Figure S95: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-1-phenyl-3-(m-tolyl) prop-2-en-1-one (5g[a]).

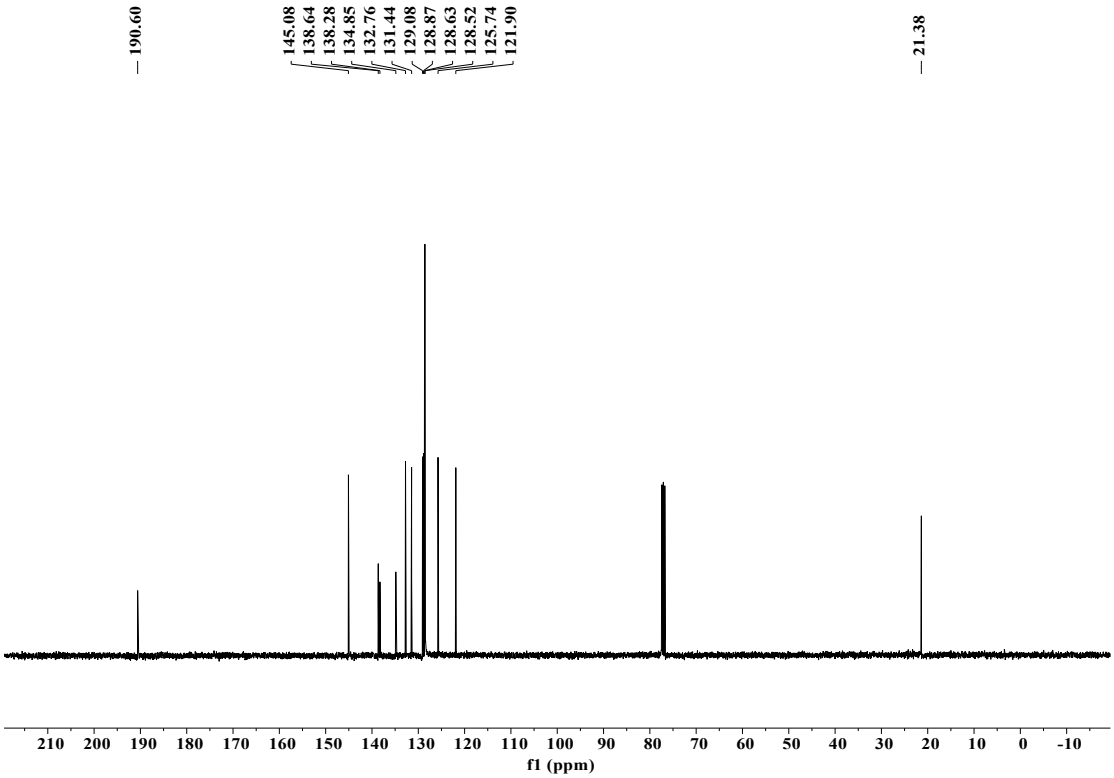


Figure S96: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-1-phenyl-3-(m-tolyl) prop-2-en-1-one (5g[a]).

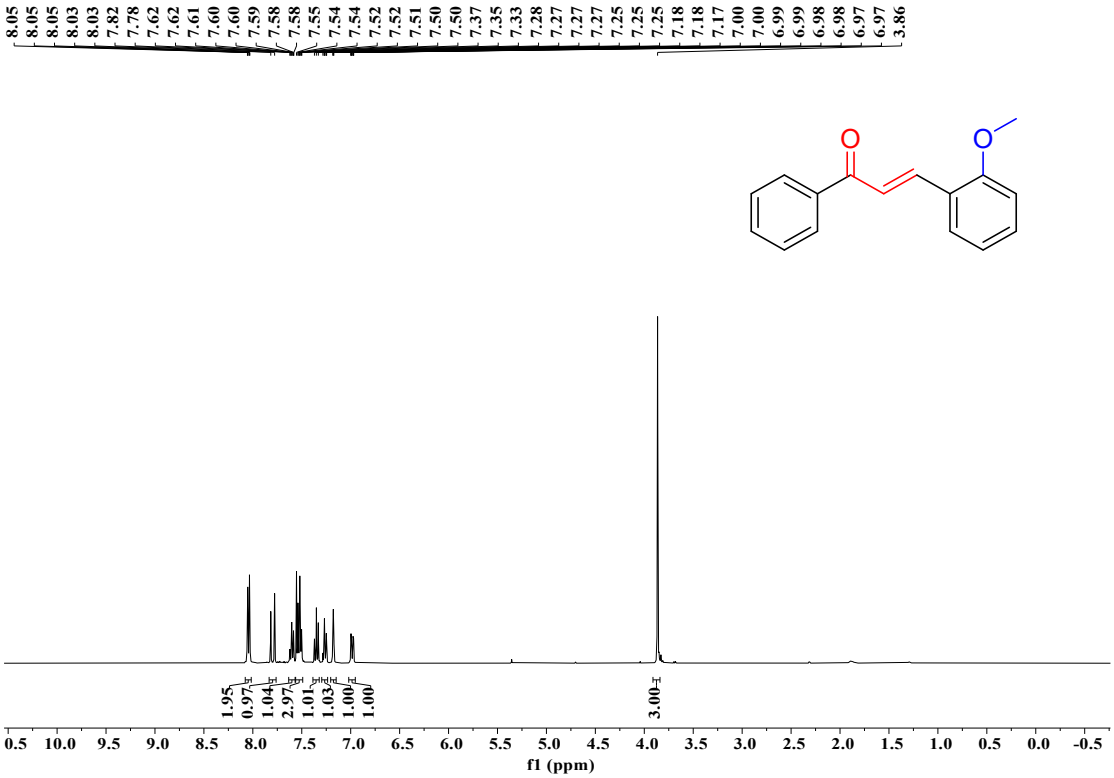


Figure S97: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(2-methoxyphenyl)-1-phenylprop-2-en-1-one (5h[a]).

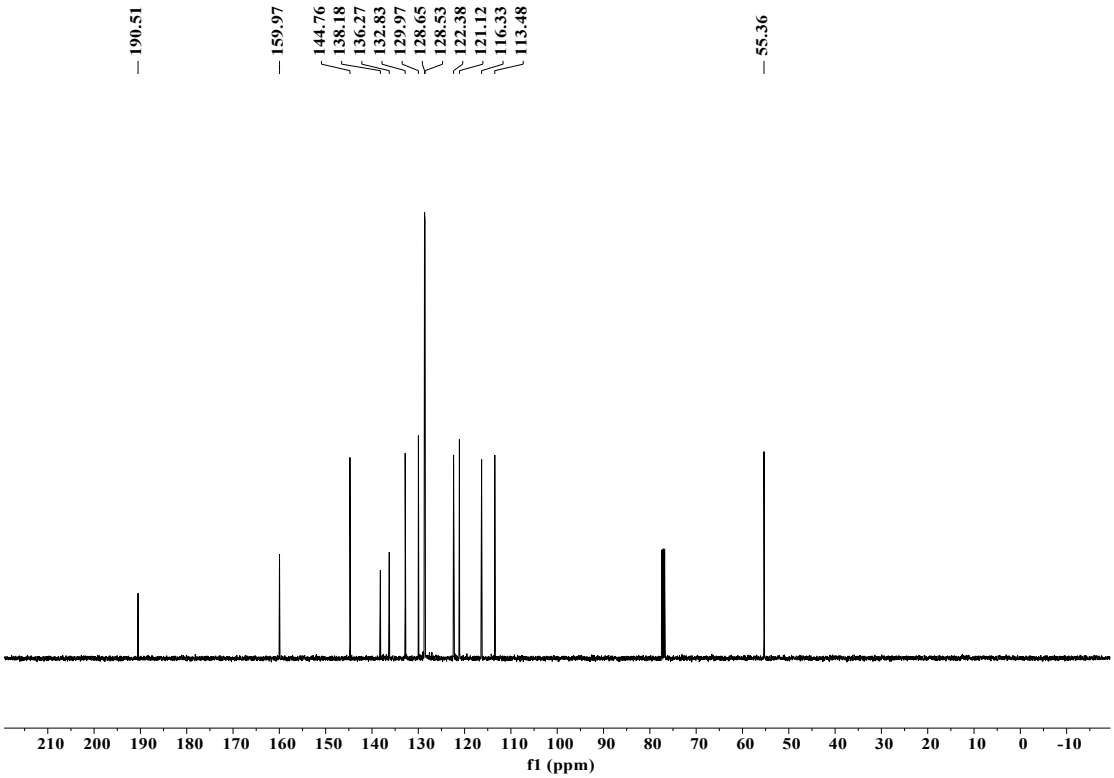


Figure S98: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(2-methoxyphenyl)-1-phenylprop-2-en-1-one (5h[a]).

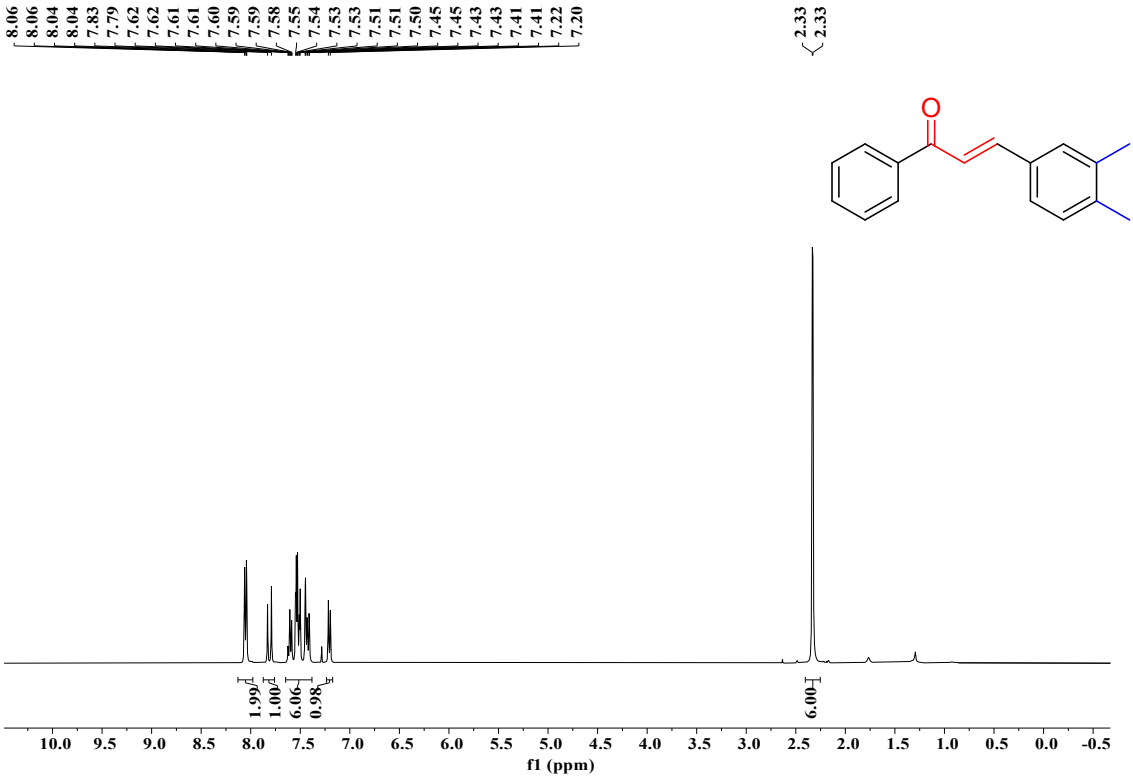


Figure S99: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(3,4-dimethylphenyl)-1-phenylprop-2-en-1-one (5i[a]).

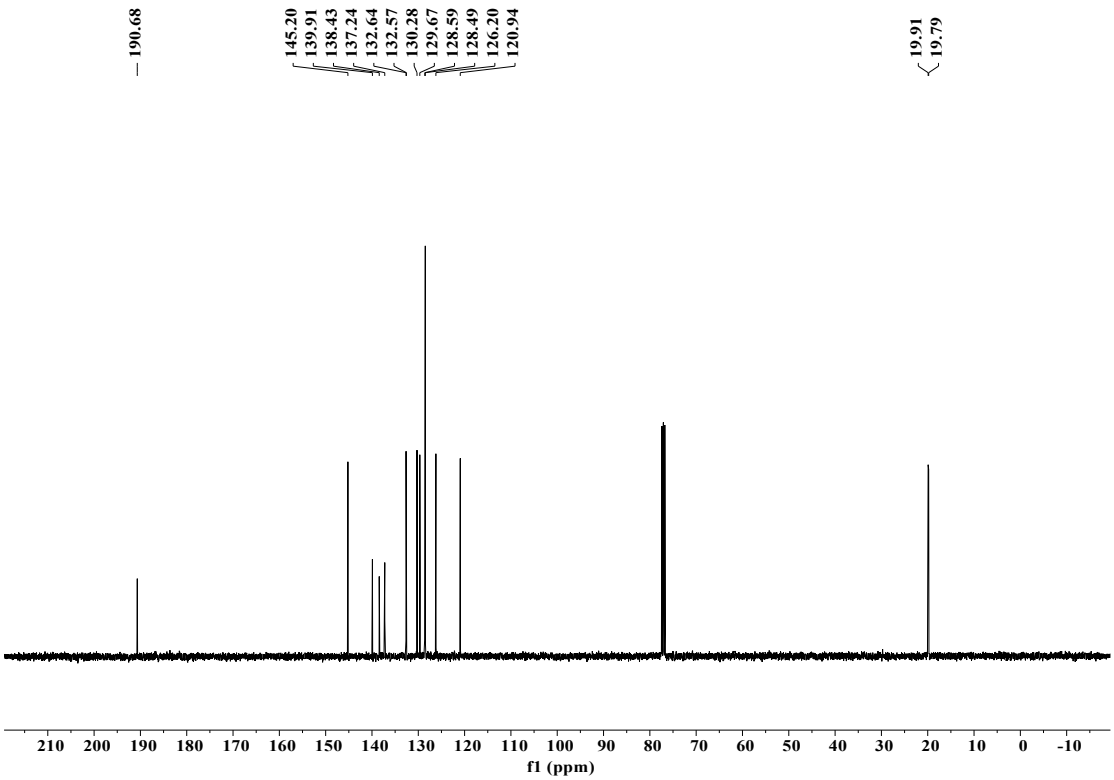


Figure S100: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(3,4-dimethylphenyl)-1-phenylprop-2-en-1-one (5i[a]).

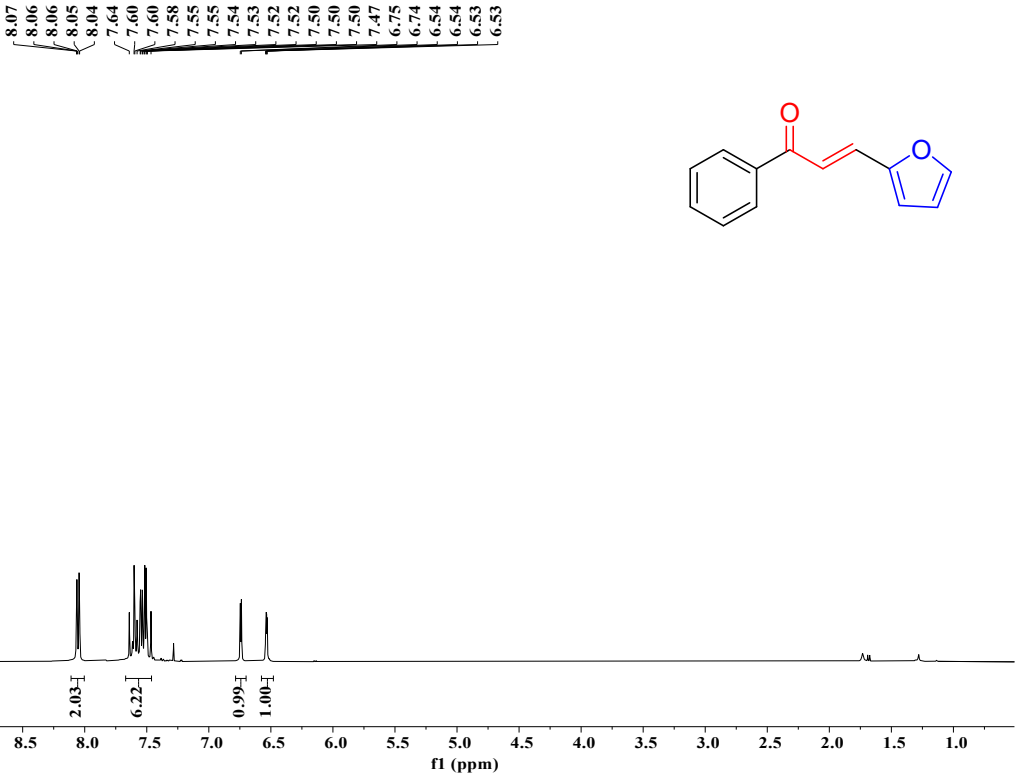


Figure S101: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(furan-2-yl)-1-phenylprop-2-en-1-one (5j[a]).

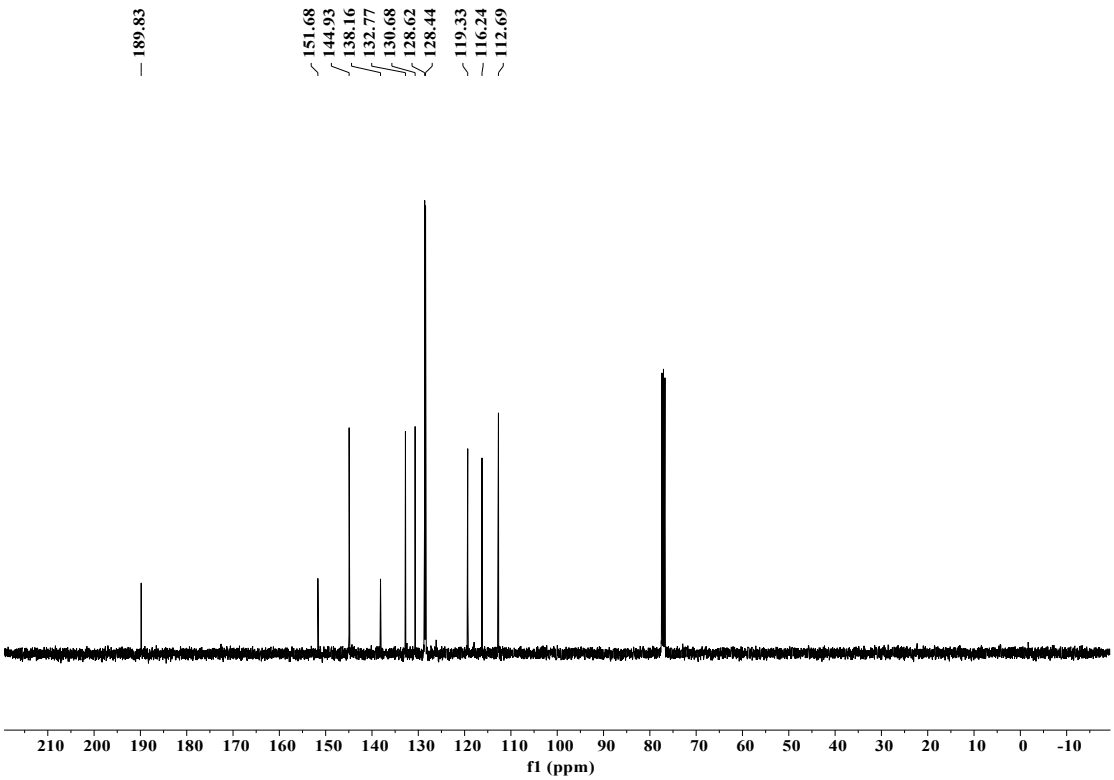


Figure S102: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of (E)-3-(furan-2-yl)-1-phenylprop-2-en-1-one (5j[a]).

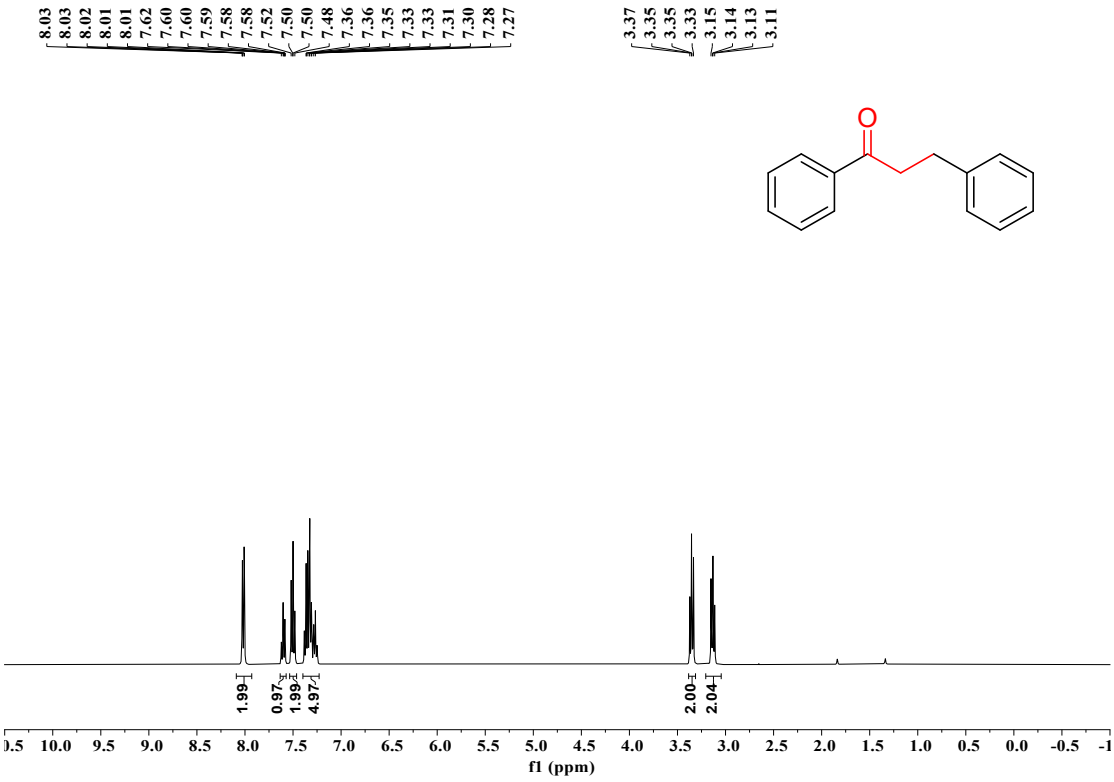


Figure S103: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 1,3-diphenylpropan-1-one (5c[b]).

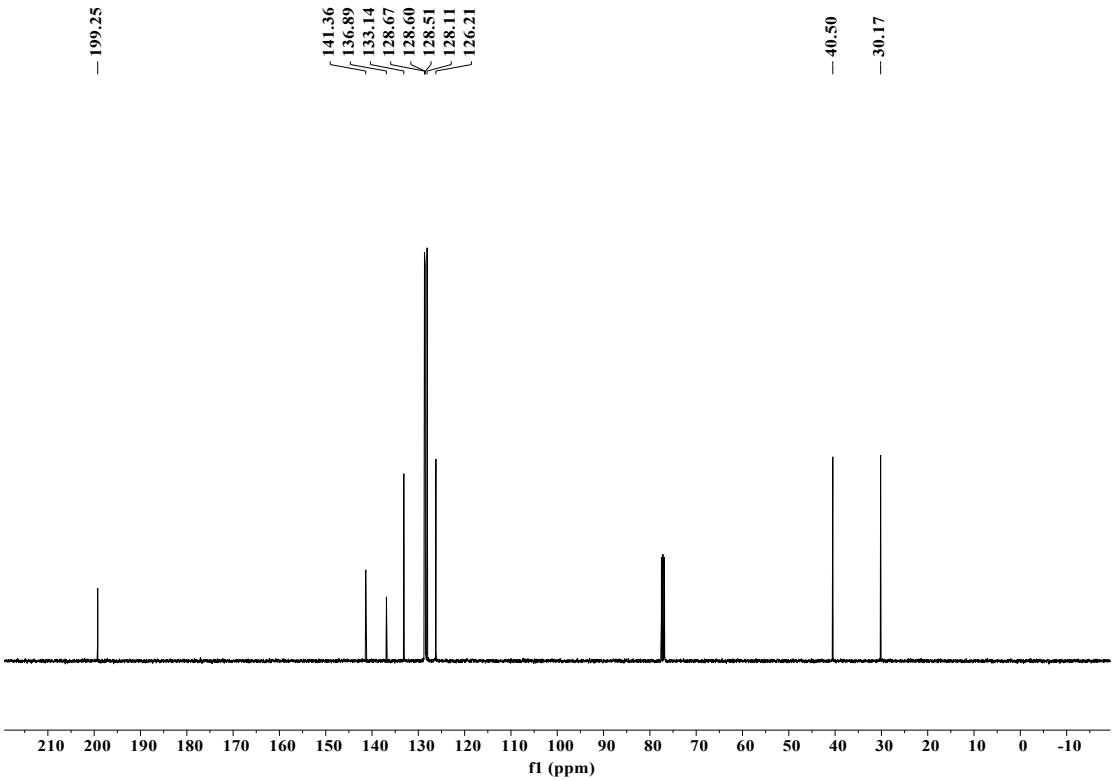


Figure S104: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 1,3-diphenylpropan-1-one (5c[b]).

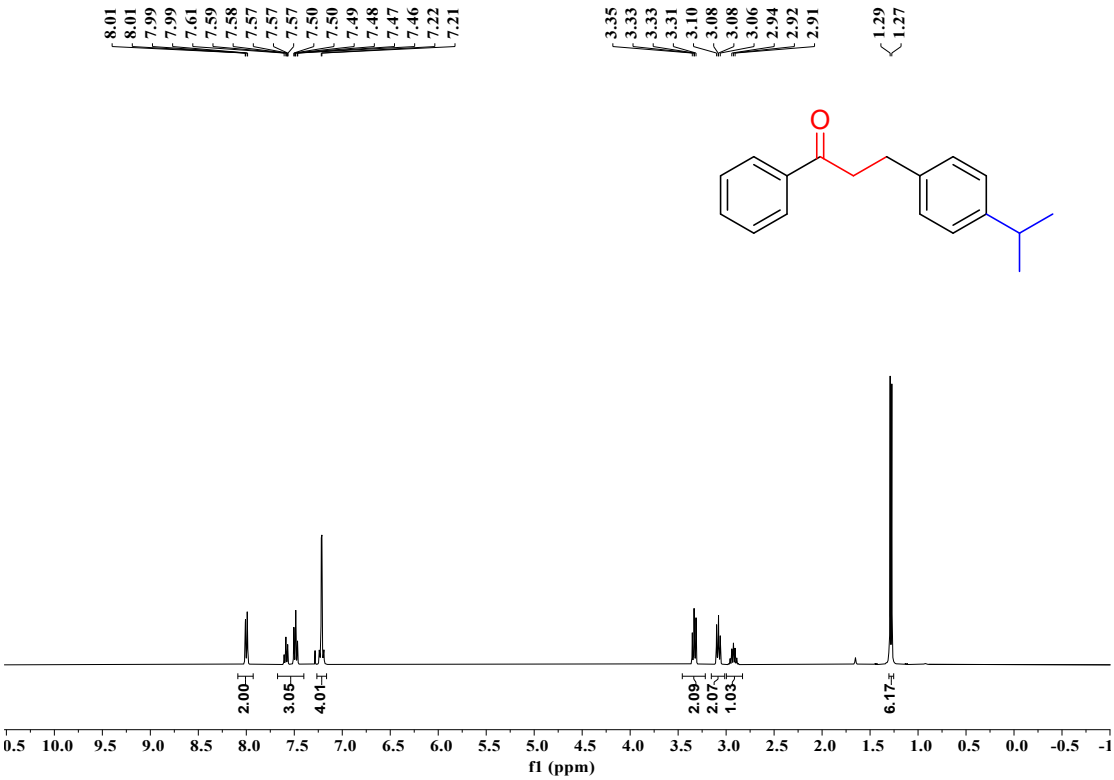


Figure S105: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-(4-isopropylphenyl)-1-phenylpropan-1-one (5d[b]).

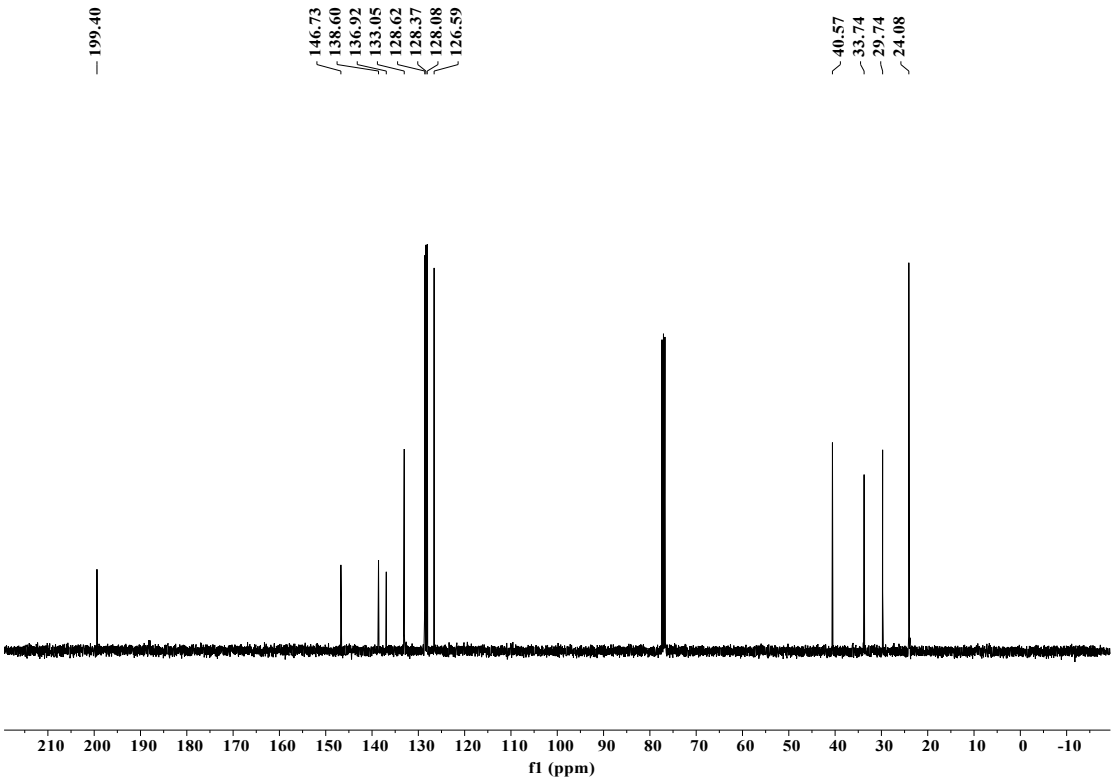


Figure S106: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 3-(4-isopropylphenyl)-1-phenylpropan-1-one (5d[b]).

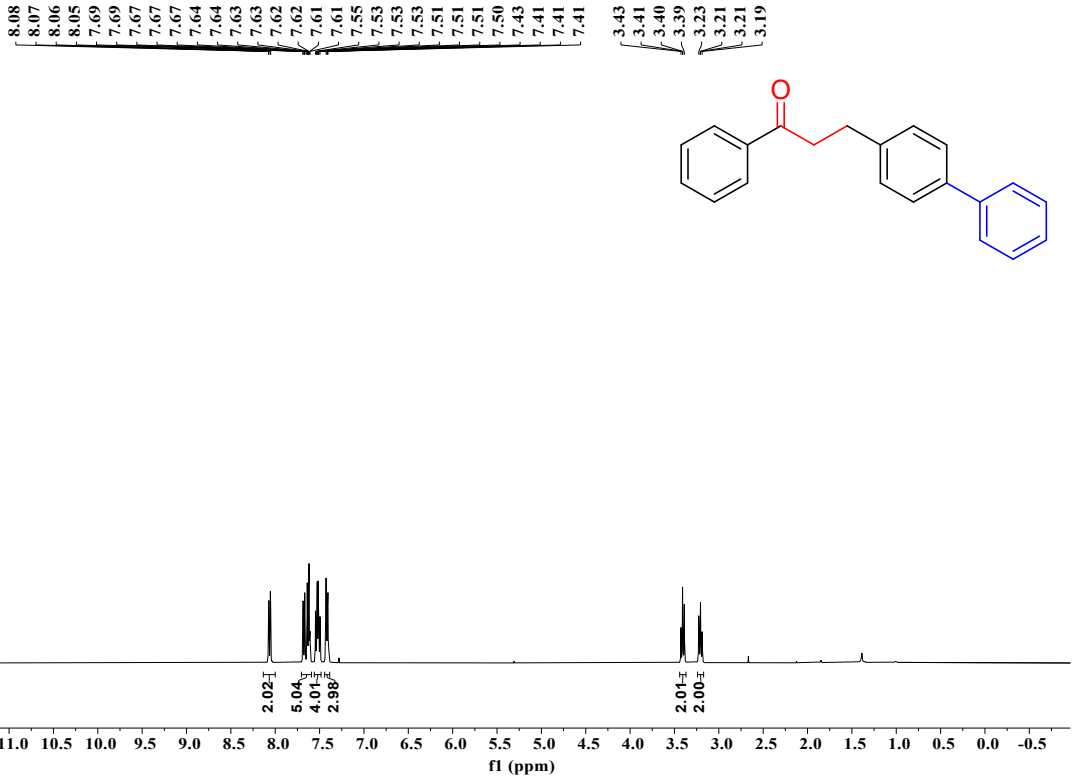


Figure S107: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-([1,1'-biphenyl]-4-yl)-1-phenylpropan-1-one (5e[b]).

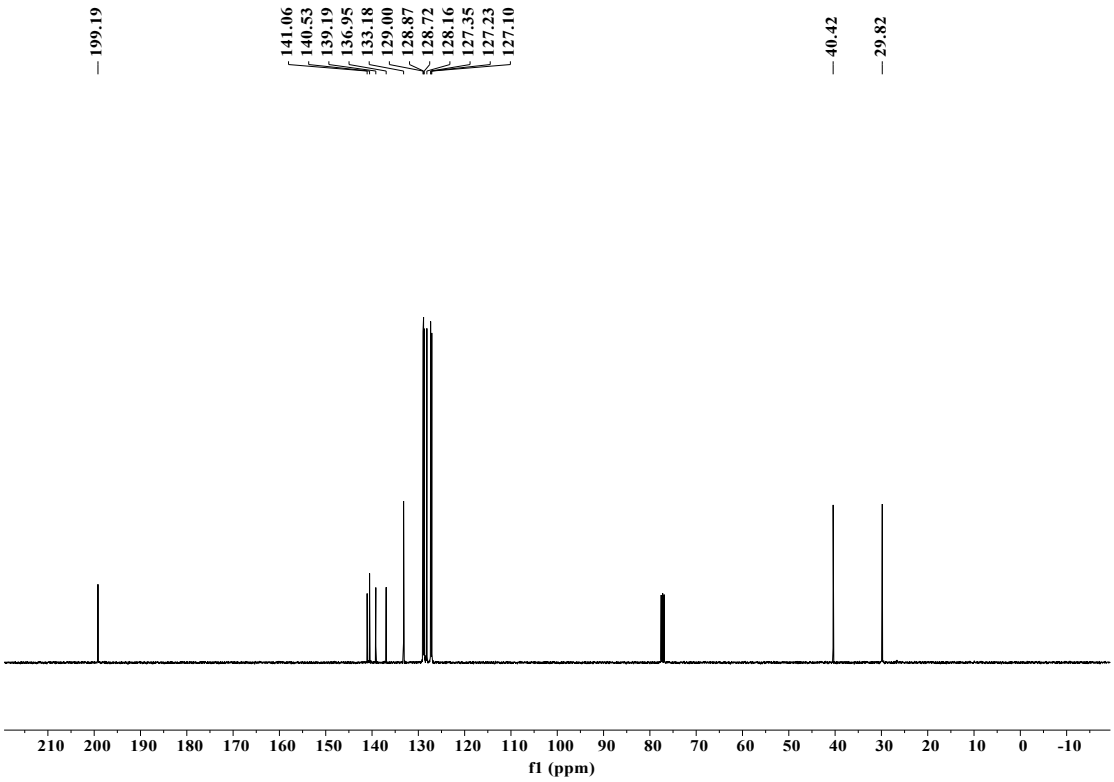


Figure S108: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 3-([1,1'-biphenyl]-4-yl)-1-phenylpropan-1-one (5e[b]).

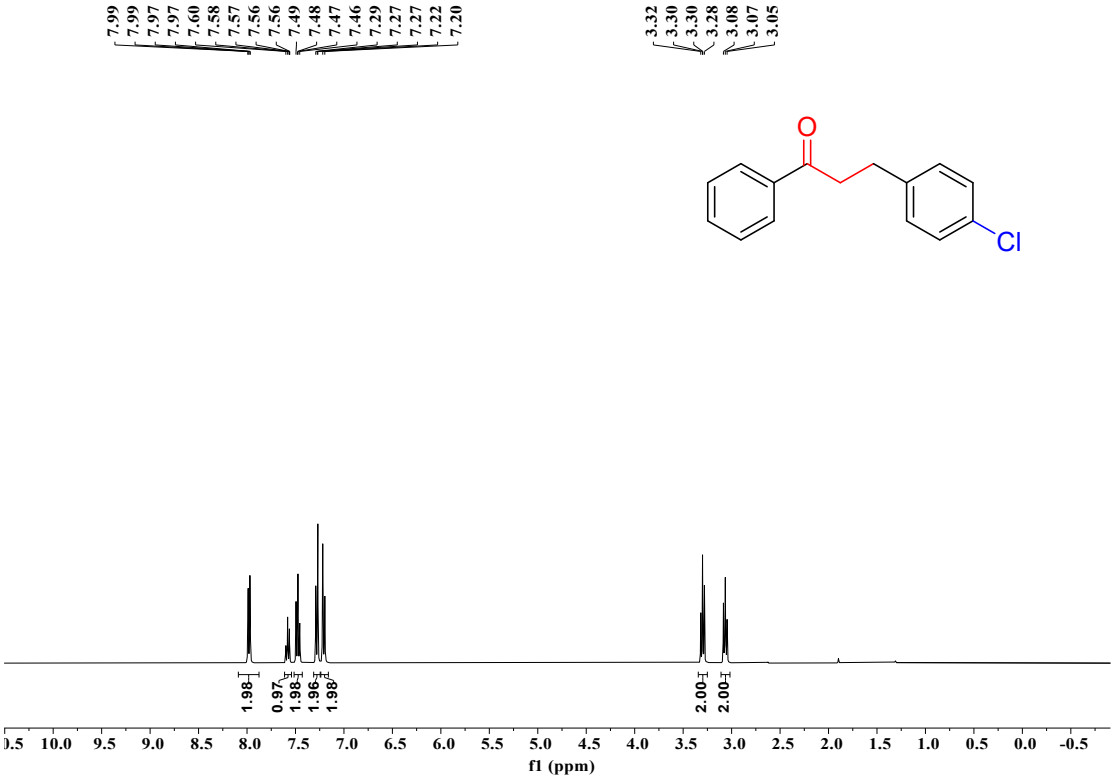


Figure S109: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-(4-chlorophenyl)-1-phenylpropan-1-one (5f[b]).

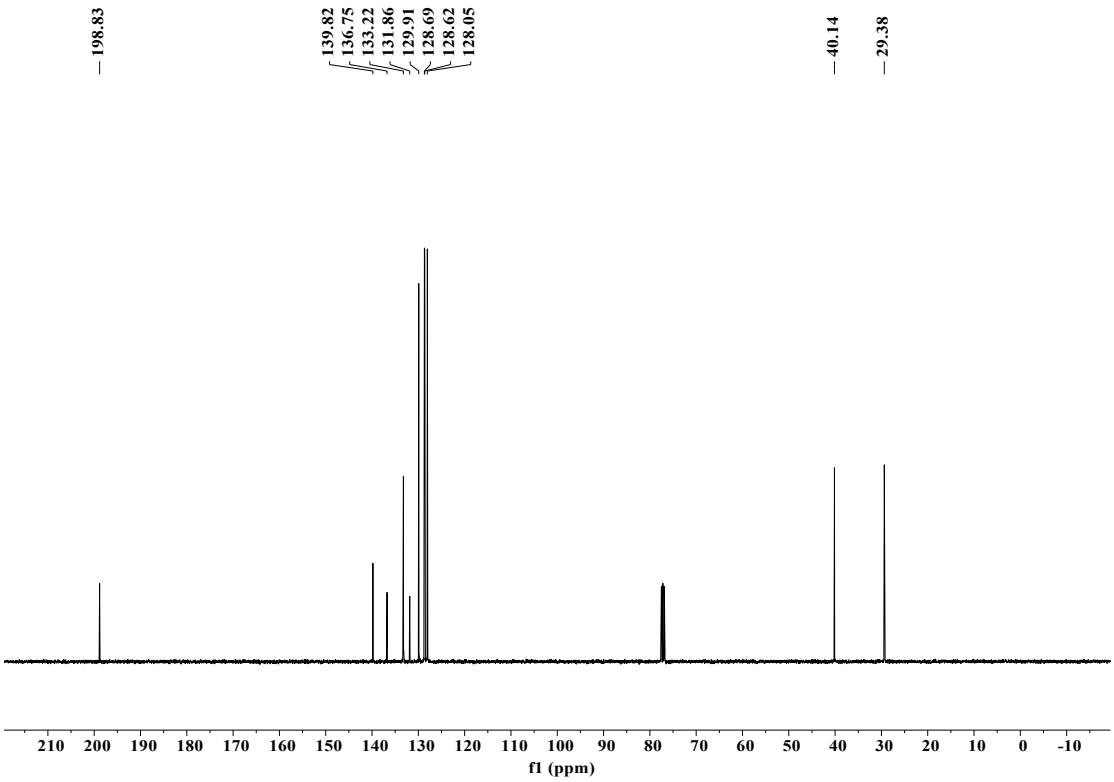


Figure S110: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 3-(4-chlorophenyl)-1-phenylpropan-1-one ($5f^b$).

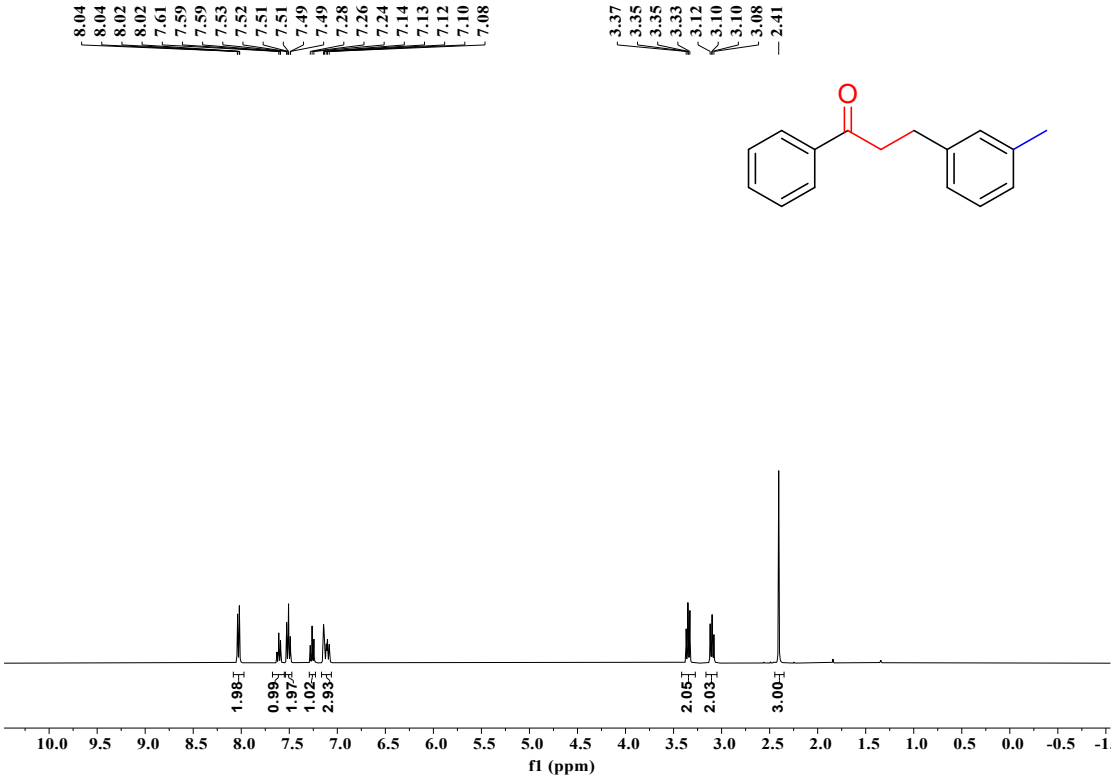


Figure S111: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 1-phenyl-3-(m-tolyl) propan-1-one ($5g^b$).

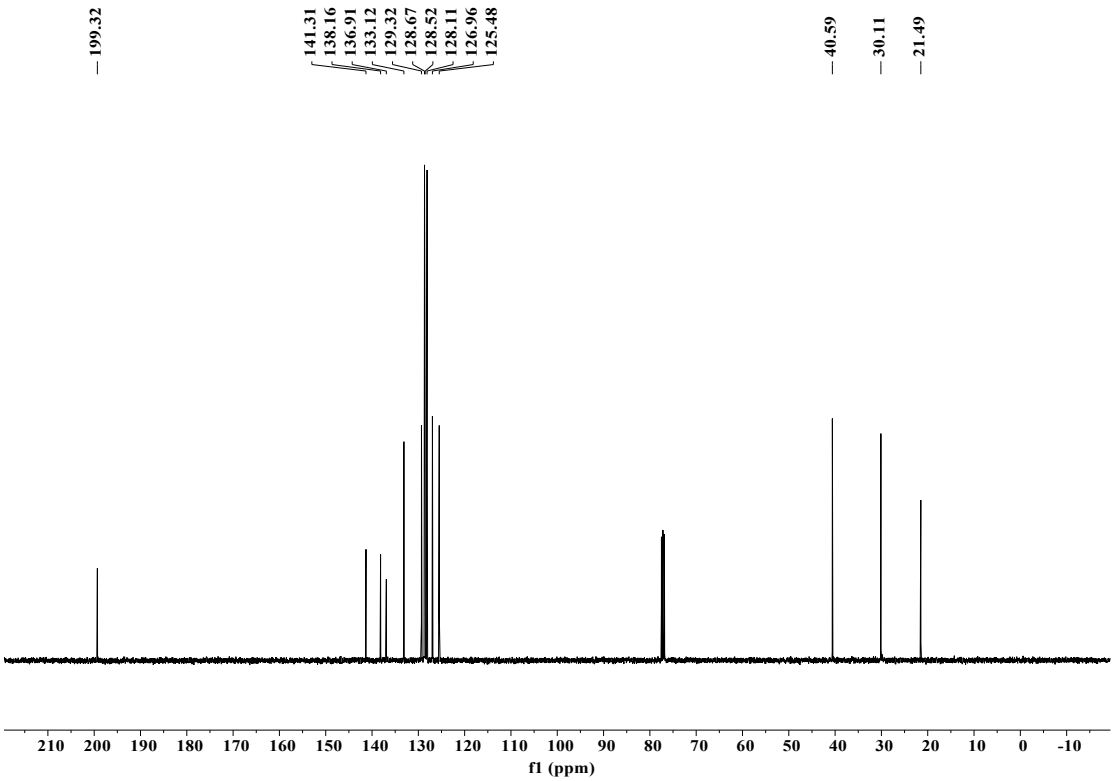


Figure S112: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 1-phenyl-3-(m-tolyl) propan-1-one (5g[b]).

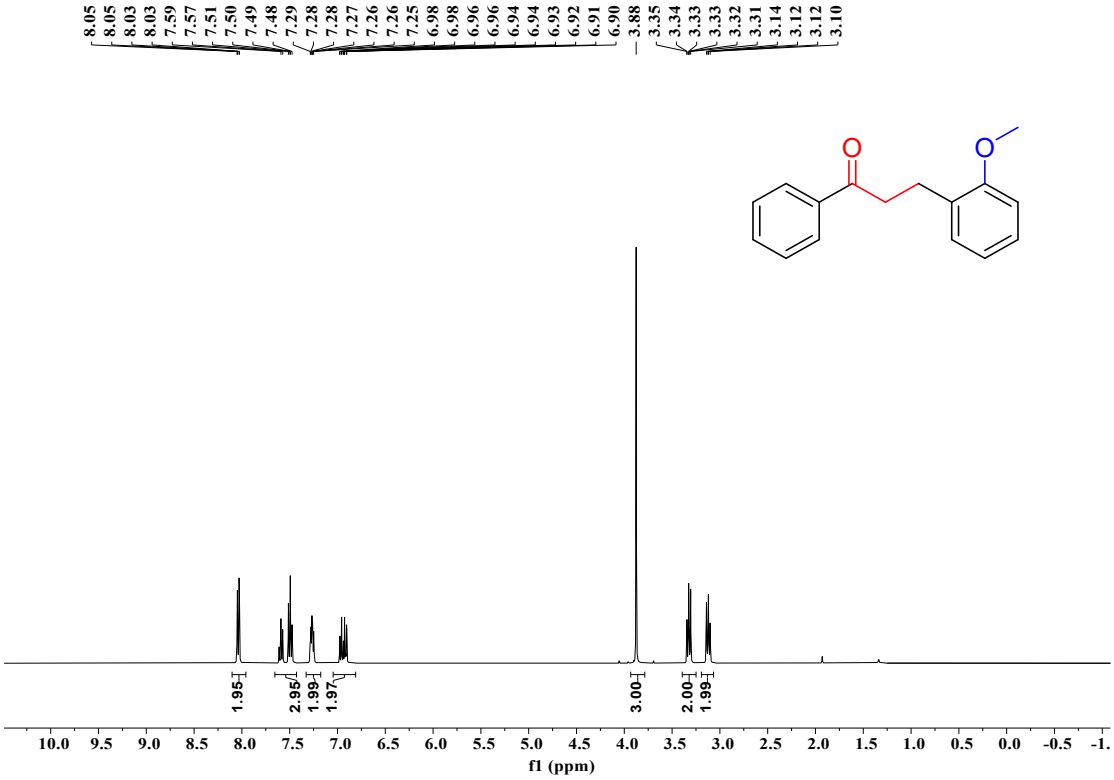


Figure S113: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-(2-methoxyphenyl)-1-phenylpropan-1-one (5h[b]).

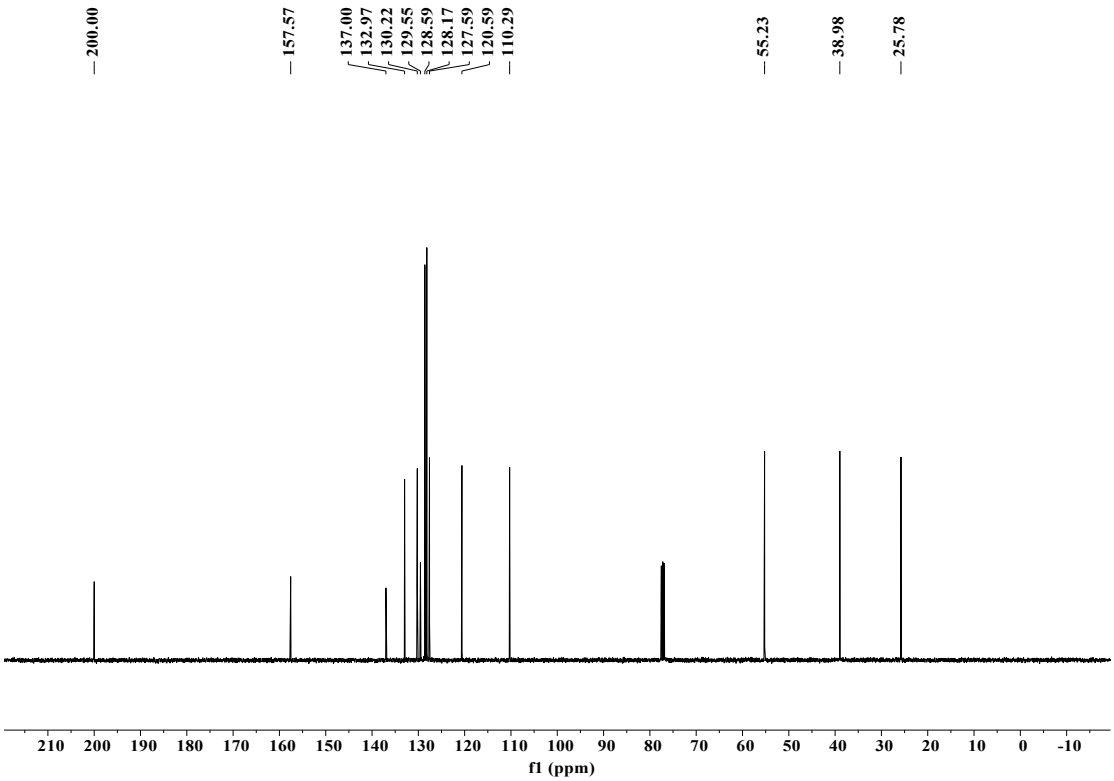


Figure S114: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 3-(2-methoxyphenyl)-1-phenylpropan-1-one (5h[b]).

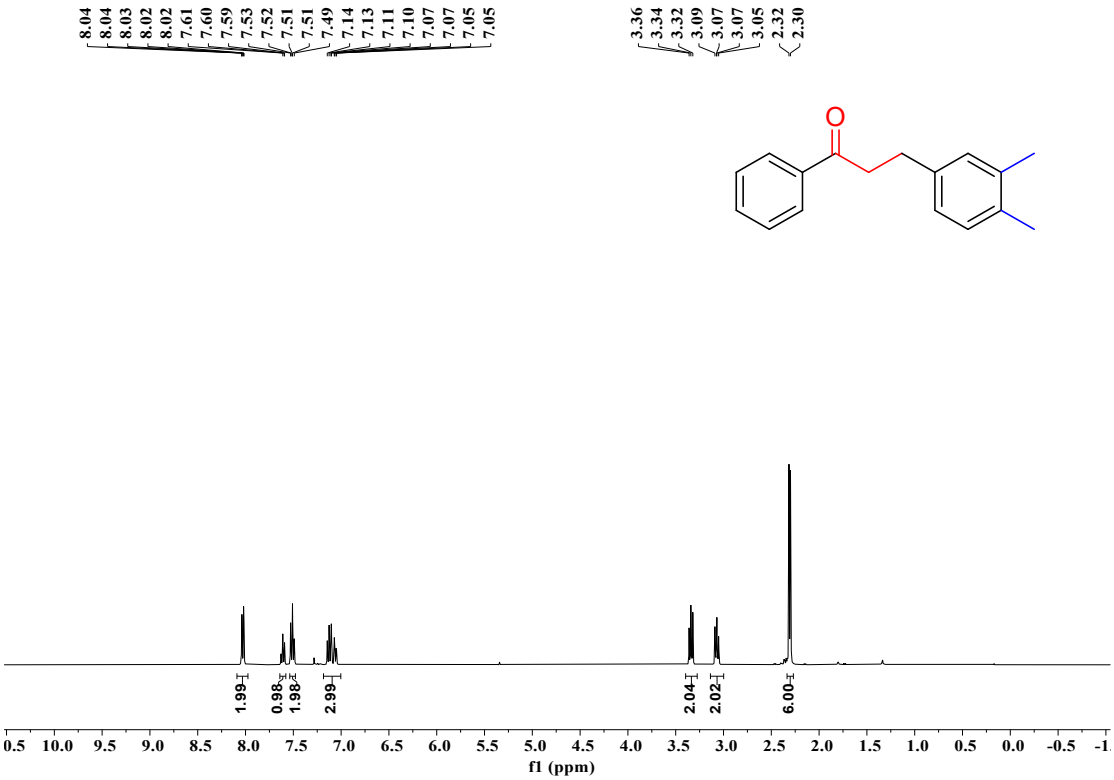


Figure S115: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-(3,4-dimethylphenyl)-1-phenylpropan-1-one (5i[b]).

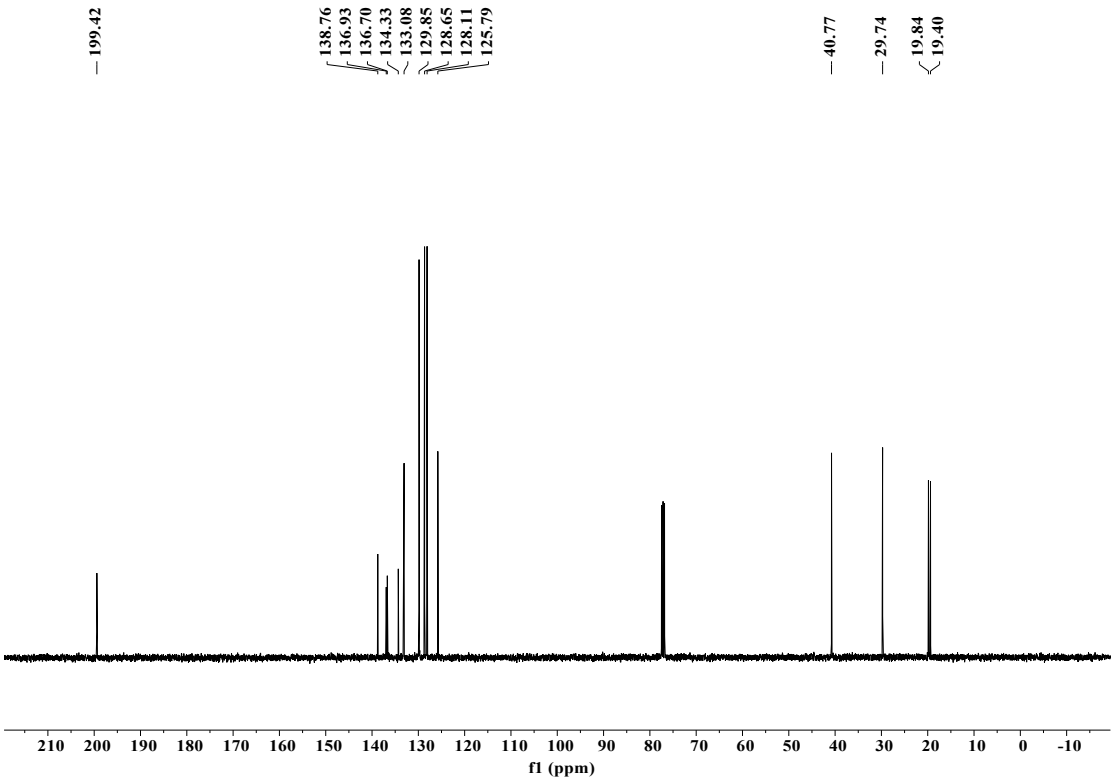


Figure S116: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 3-(3,4-dimethylphenyl)-1-phenylpropan-1-one (5i[b]).

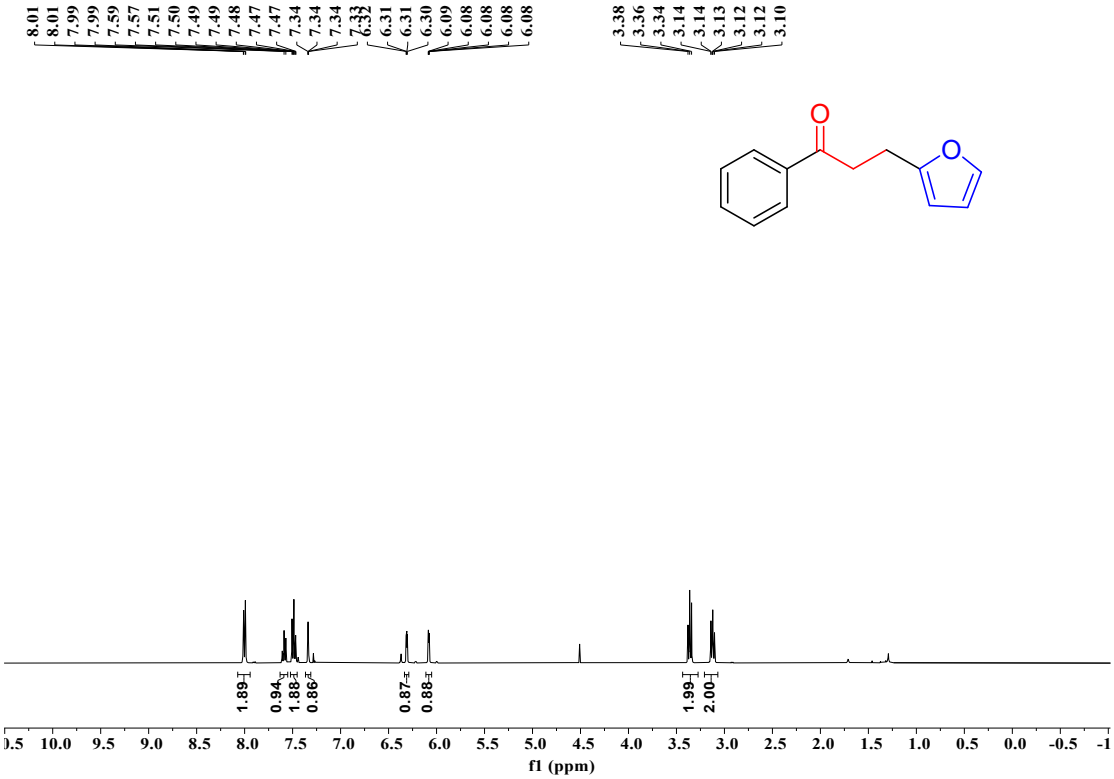


Figure S117: ^{1}H NMR spectrum of (400 MHz, $CDCl_3$) of 3-(furan-2-yl)-1-phenylpropan-1-one (5j[b]).

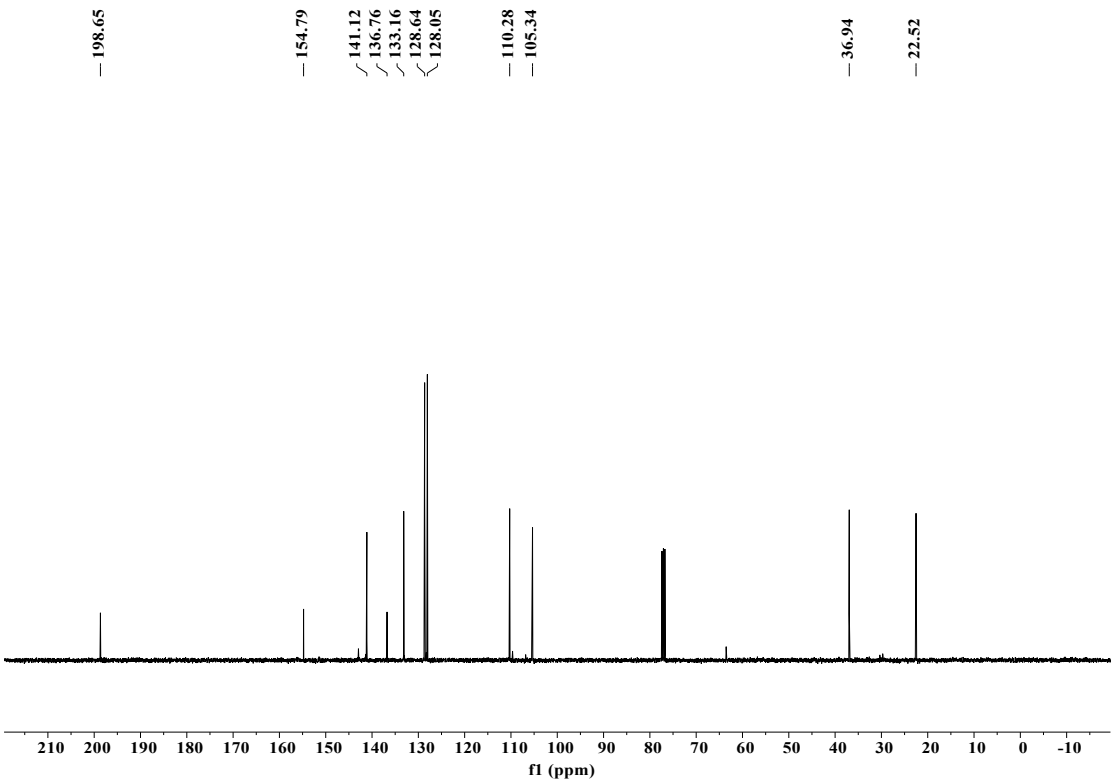


Figure S118: $^{13}C\{^{1}H\}$ NMR spectrum of (400 MHz, $CDCl_3$) of 3-(furan-2-yl)-1-phenylpropan-1-one (5j[b]).