Electronic Supplementary Material (ESI) for RSC Advances.

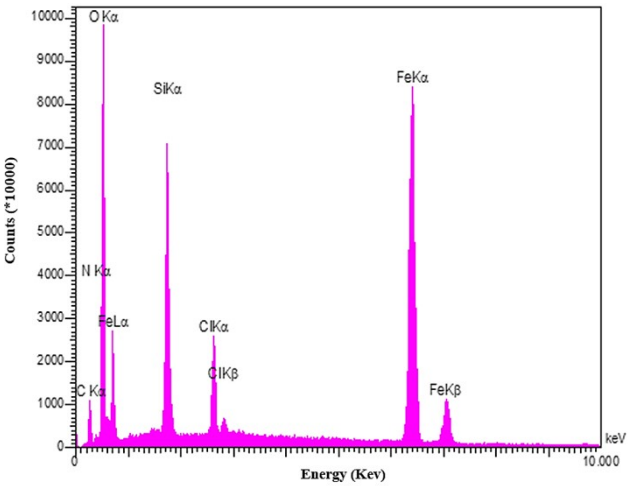


Fig. S1 Energy-dispersive X-ray spectroscopy of Fe_3O_4@SiO_2-APTES-Fe_2L^{DAR}.

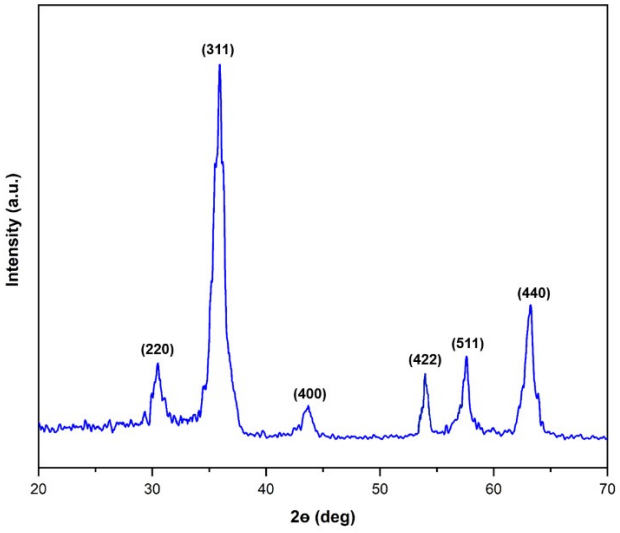


Fig. S2 XRD pattern of Fe_3O_4@SiO_2-APTES-Fe_2L^{DAR}.

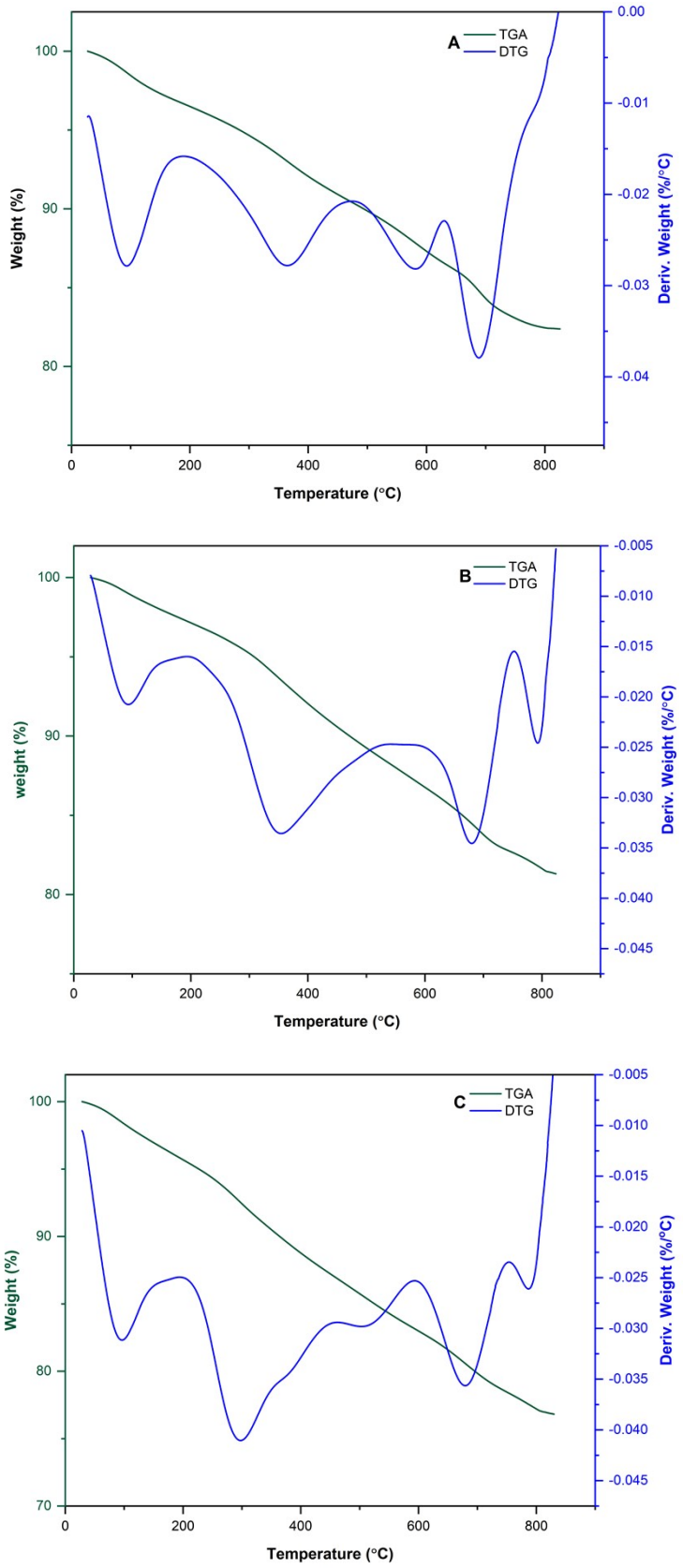


Fig. S3 (A) TGA and DTG analyses of Fe_3O_4@SiO_2-APTES, (B) Fe_3O_4@SiO_2-APTES-H_2L^{DAR}, (C) Fe_3O_4@SiO_2-APTES-Fe_2L^{DAR}.

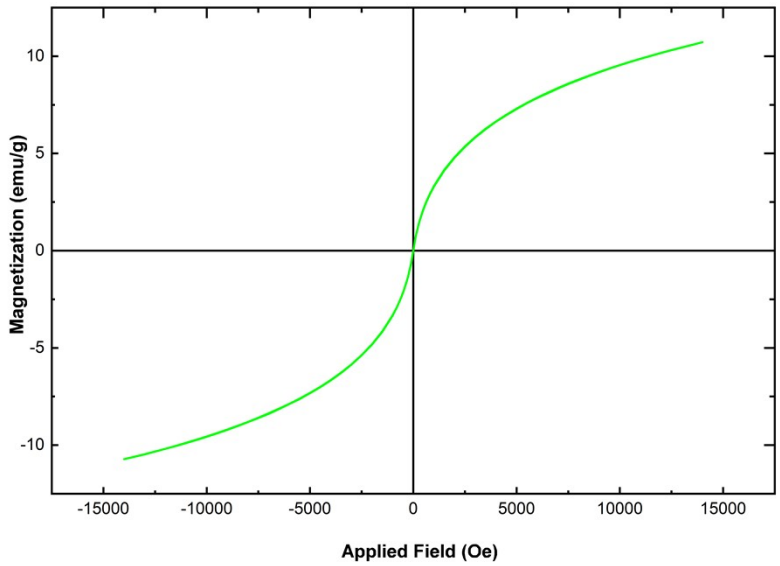


Fig. S4 Magnetization curve of Fe_3O_4@SiO_2-APTES-Fe_2L^{DAR}.

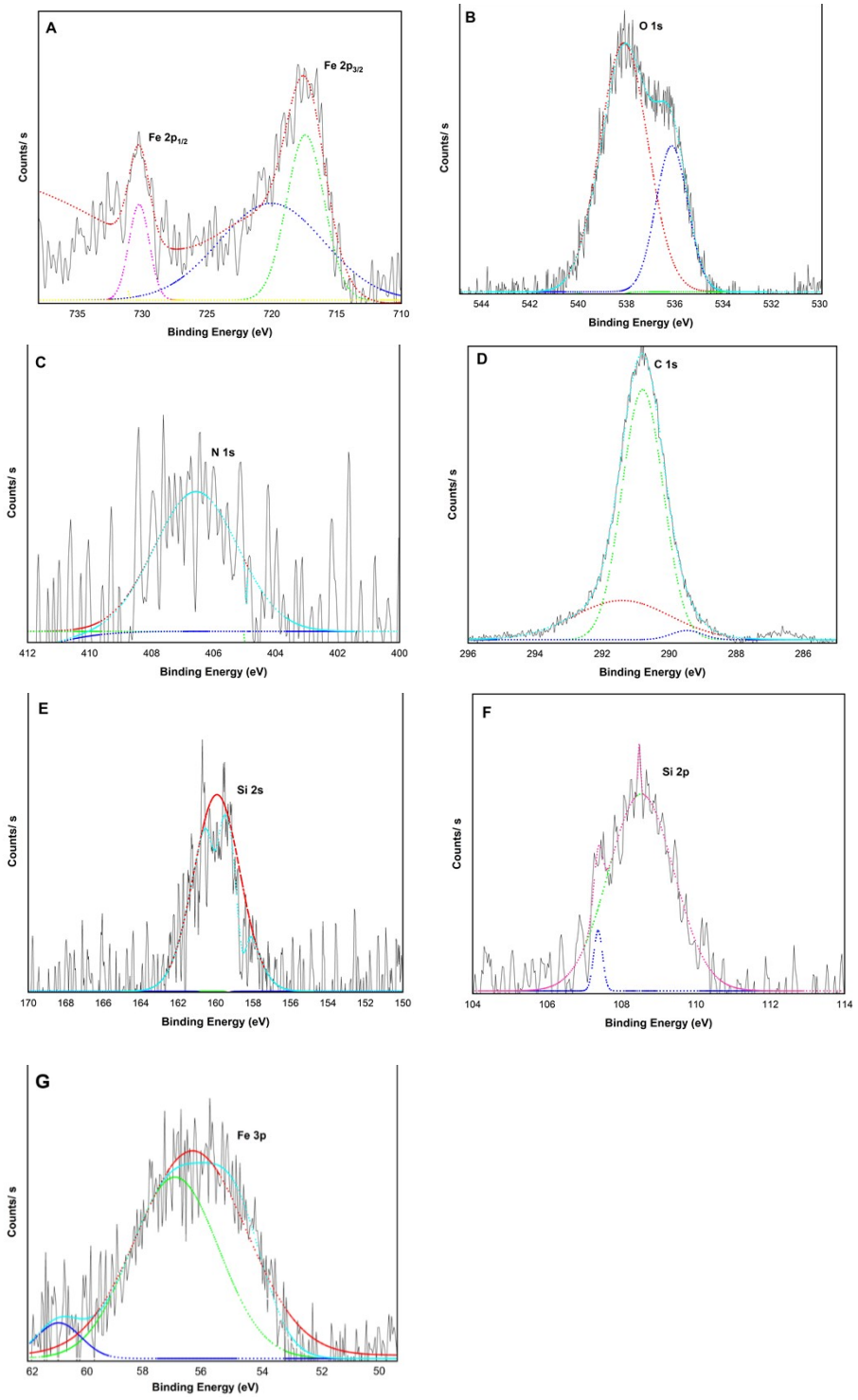
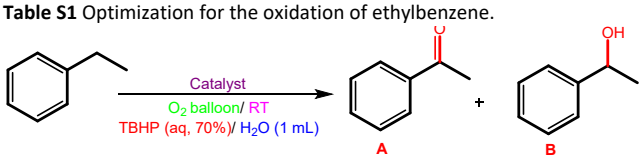


Fig. S5 The X-ray photoelectron spectroscopy (XPS) spectrum of catalyst (Fe_3O_4@SiO_2-APTES-Fe_2L^{DAR}).

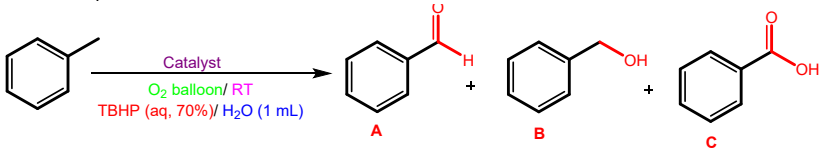
Table S1 Optimization for the oxidation of ethylbenzene.



Entry	Catalyst [mg]	Solvent	O_2/Ar	TBHP[a]	T [h]	Conv.[b] [%]	Sel.[c] [%] to A
1	30.0[d]	H_2O	O_2	2	8	83	100
2	30.0[d]	H_2O	O_2	-	2	6	100
3	30.0[d]	H_2O	O_2	-	5	21	100
4	30.0[d]	H_2O	O_2	-	10	52	100
5	30.0[d]	H_2O	O_2	-	15	100	100
6	40.0[e]	H_2O	O_2	-	15	100	100
7	40.0[e]	H_2O	O_2	-	12	87	100
8	20.0[f]	H_2O	O_2	-	15	94	100
9	10.0[g]	H_2O	O_2	-	15	71	100
10[h]	-	H_2O	O_2	-	15	trace	trace
11[i]	30.0[d]	H_2O	O_2	-	15	trace	trace

Reaction conditions: Catalyst, Substrate (1 mmol), O_2 balloon, H_2O (1 mL), T= room temperature.
[a] TBHP (70% in H_2O).
[b] Conversions were determined by GC using biphenyl as an internal standard (molar ratio of substrate to internal standard is 1:1).
[c] Selectivity % = [(Product %) / (Products %)] × 100.
[d] 30.0 mg ≈ 0.9 mol% [e] 40.0 mg ≈ 1.2 mol% [f] 20.0 mg ≈ 0.6 mol%. [g] 10.0 mg ≈ 0.3 mol%.
[h] In the absence of catalyst.
[i] Reference test in the presence of $FeCl_3$.

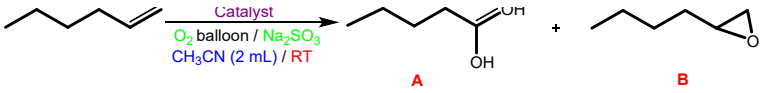
Table S2 Optimization for the oxidation of toluene.



Entry	Catalyst type and amount[mg]	Solvent	O_2/Ar	TBHP[a]	T [h]	Conv.[b] [%]	Sel.[c] [%] to C
1[d]	-	H_2O	O_2	2	20	-	-
2[e]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 30.0	H_2O	O_2	2	18	45	100
3[e]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 30.0	H_2O	O_2	3	18	72	100
4[f]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 40.0	H_2O	O_2	3	18	83	100
5[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	H_2O	O_2	3	18	87	100
6[f]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 40.0	H_2O	O_2	3	20	91	100
7[e]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 30.0	H_2O	O_2	3	20	82	100
8[h]	Fe_3O_4@SiO_2APTES 40.0	H_2O	O_2	3	20	94	1
9[i]	Fe_3O_4@SiO_2APTESH_2L^{DAR} 40.0	H_2O	O_2	3	20	95	3
10[l]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 40.0	H_2O	Ar	3	20	95	8

Reaction conditions: Catalyst, Substrate (1 mmol), O_2 balloon, H_2O (1 mL), T = room temperature.
[a] TBHP (70% in H_2O).
[b] Conversions were determined by GC using biphenyl as an internal standard (molar ratio of substrate to internal standard is 1:1).
[c] Selectivity % = [(Product %) / (Products %)] × 100.
[d] In the absence of catalyst .
[e] 30.0 mg ≈ 0.9 mol%. [f] 40.0 mg ≈ 1.2 mol%. [g] 50.0 mg ≈ 1.5 mol%.
[h] Reference test in the presence of Fe_3O_4@SiO_2APTES.
[i] Reference test in the presence of Fe_3O_4@SiO_2APTESH_2L^{DAR}.
[l] Reference test in the absence of O_2 balloon (under Ar atmosphere)

Table S3 Optimization for the oxidation of 1-hexene.

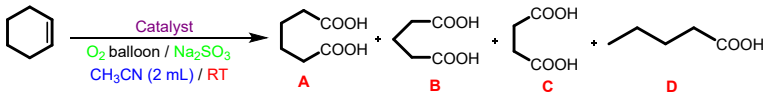


Entry	Catalyst type and amount[mg]	Solvent	O_2/Ar	Na_2SO_3[a] [mmol]	T [h]	Conv.[b] [%]	Sel.[c] [%] to A
1[d]	-	CH_3CN	O_2	2	15	-	-
2[e]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 30.0	CH_3CN	O_2	2	15	48	100
3[f]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 40.0	CH_3CN	O_2	2	15	72	100
4[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	CH_3CN	O_2	2	15	94	100
5[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	CH_3CN	O_2	2	10	57	100
6[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	CH_3CN	O_2	1	15	67	100
7[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	CH_3CN	O_2	3	15	90	100
8[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	CH_3CN	O_2	-	15	24	100
9[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	Solvent free	O_2	2	15	-	-
10[g]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	H_2O/acetone	O_2	2	15	84	100
11[h]	Fe_3O_4@SiO_2APTES 50.0	CH_3CN	O_2	2	15	-	-
12[i]	Fe_3O_4@SiO_2APTESH_2L^{DAR} 50.0	CH_3CN	O_2	2	15	-	-
13[l]	Fe_3O_4@SiO_2APTESFe_2L^{DAR} 50.0	CH_3CN	Ar	2	15	74	100
14[m]	$FeCl_3$ 50.0	CH_3CN	O_2	2	15	-	-

Reaction conditions: Catalyst, Substrate (2 mmol), O_2 balloon, CH_3CN (2 mL), T = room temperature.
[a] Na_2SO_3 (2 mmol, 0.24 g).
[b] Isolated yield.
[c] Selectivity % = [(Product %) / (Products %)] × 100.
[d] In the absence of catalyst.
[e] 30.0 mg ≈ 0.9 mol%. [f] 40.0 mg ≈ 1.2 mol%. [g] 50.0 mg ≈ 1.5 mol%.
[h] Reference test in the presence of Fe_3O_4@SiO_2APTES.
[i] Reference test in the presence of Fe_3O_4@SiO_2APTESH_2L^{DAR}.
[l] Reference test in the absence of O_2 balloon (under Ar atmosphere).

[m] Reference test in the presence of $FeCl_3$.

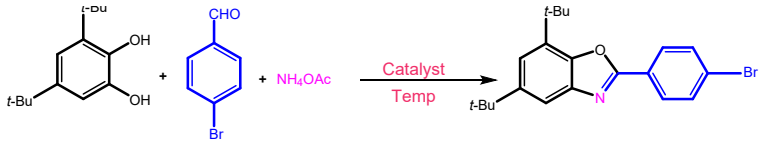
Table S4 Optimization for the oxidation of cyclohexene.



Entry	Catalyst [mg]	Solvent	O_2/Ar	Na_2SO_3[a]	T [h]	Yield [b] [%]
1[f]	-	CH_3CN	O_2	2	24	-
2	30.0[c]	CH_3CN	O_2	2	24	57
3	40.0[d]	CH_3CN	O_2	2	24	78
4	50.0[e]	CH_3CN	O_2	2	24	87
5	50.0[e]	CH_3CN	O_2	2	15	65
6	50.0[e]	CH_3CN	O_2	1	24	58
7	50.0[e]	CH_3CN	O_2	-	24	-
8	50.0[e]	H_2O/acetone	O_2	2	24	80
9	50.0[e]	Toluene	O_2	2	24	-
10	50.0[e]	$CHCl_3$	O_2	2	24	-
11	30.0[c]	Solvent free	O_2	2	24	-

Reaction conditions: Catalyst, Substrate (2 mmol), O_2 balloon, CH_3CN (2 mL), T= room temperature.
[a] Na_2SO_3 (2 mmol, 0.24 g).
[b] Isolated yield.
[c] 30.0 mg ≈ 0.9 mol% [d] 40.0 mg ≈ 1.2 mol% [e] 50.0 mg ≈ 1.5mol%.
[f] In the absence of catalyst.

Table S5 Optimization of the reaction conditions for synthesis of 7. 5,7-Di-tert-butyl-2-(4-bromophenyl)benzo[d]oxazole.



Entry	Catalyst/mol %	solvent	temp.	time (h)	Yield[a] (%)
1[c]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	Solvent free	25	24	-
2[c]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	Solvent free	70	24	18
3[c]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	H_2O	25	24	36
4[c]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	H_2O	70	24	97
5[c]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	H_2O	70	10	97
6[c]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	H_2O	70	7	75
7[b]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/0.9	H_2O	70	10	79
8[d]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.5	H_2O	70	10	95
9[d]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.5	H_2O	70	7	71
10[e]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	H_2O	70	10	84
11[f]	Fe_3O_4@SiO_2-APTESFe_2L^{DAR}/1.2	H_2O	70	10	77
12[g]	Fe_3O_4@SiO_2-APTESH_2L^{DAR}/1.2	H_2O	70	10	17
13[h]	Fe_3O_4@SiO_2-APTES/1.2	H_2O	70	10	8
14[i]	$FeCl_3$/1.2	H_2O	70	10	-
15	-	H_2O	70	10	-

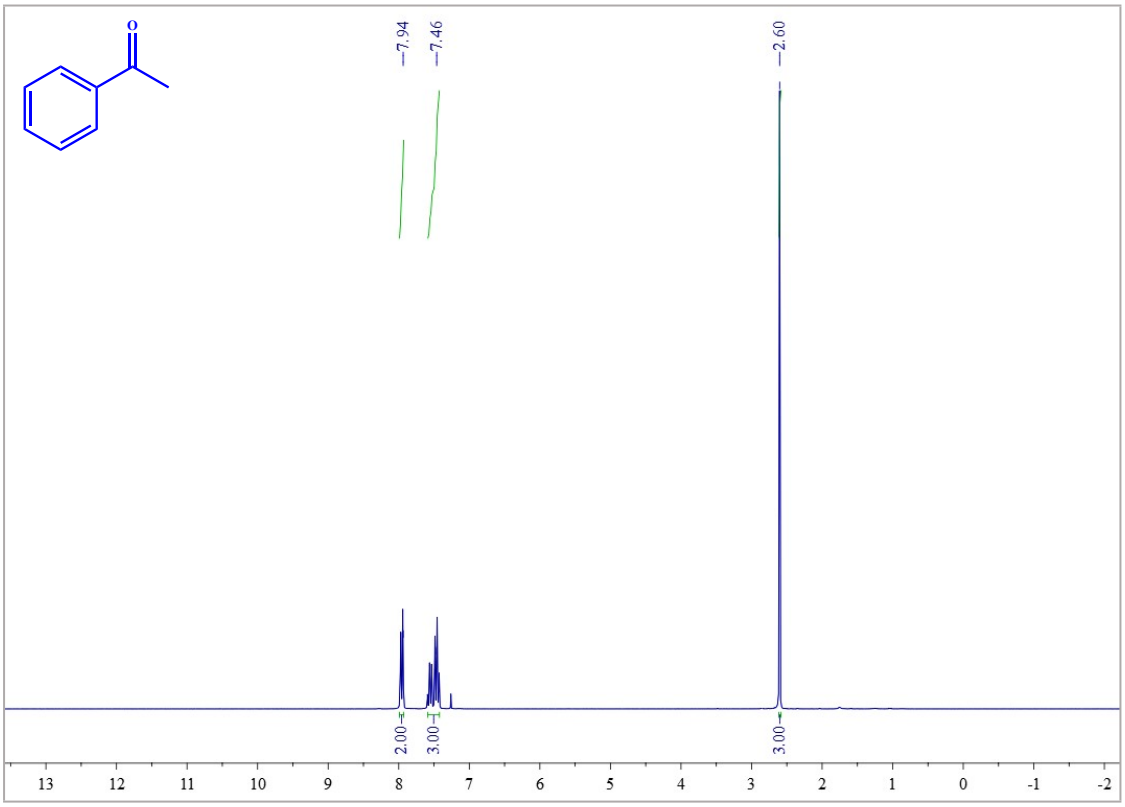
Reaction conditions: Catalyst, 3,5-di-tert-buthylbenzene-1,2-diol (0.5 mmol), NH_4OAc (0.5 mmol) and 4-bromobenzaldehyde (0.5 mmol), H_2O (7 mL).
[a] Isolated yield.
[b] 30.0 mg ≈ 0.9 mol% [c] 40.0 mg ≈ 1.2 mol% [d] 50.0 mg ≈ 1.5 mol%.
[e] NH_4OAc (1 mmol) [f] NH_4OAc (2 mmol).
[g] Reference test in the presence of Fe_3O_4@SiO_2-APTES-H_2L^{DAR}.
[h] Reference test in the presence of Fe_3O_4@SiO_2-APTES.
[i] Reference test in the presence of $FeCl_3$.

^{1}H NMR spectra for all synthesized compounds

C-H bond oxidation

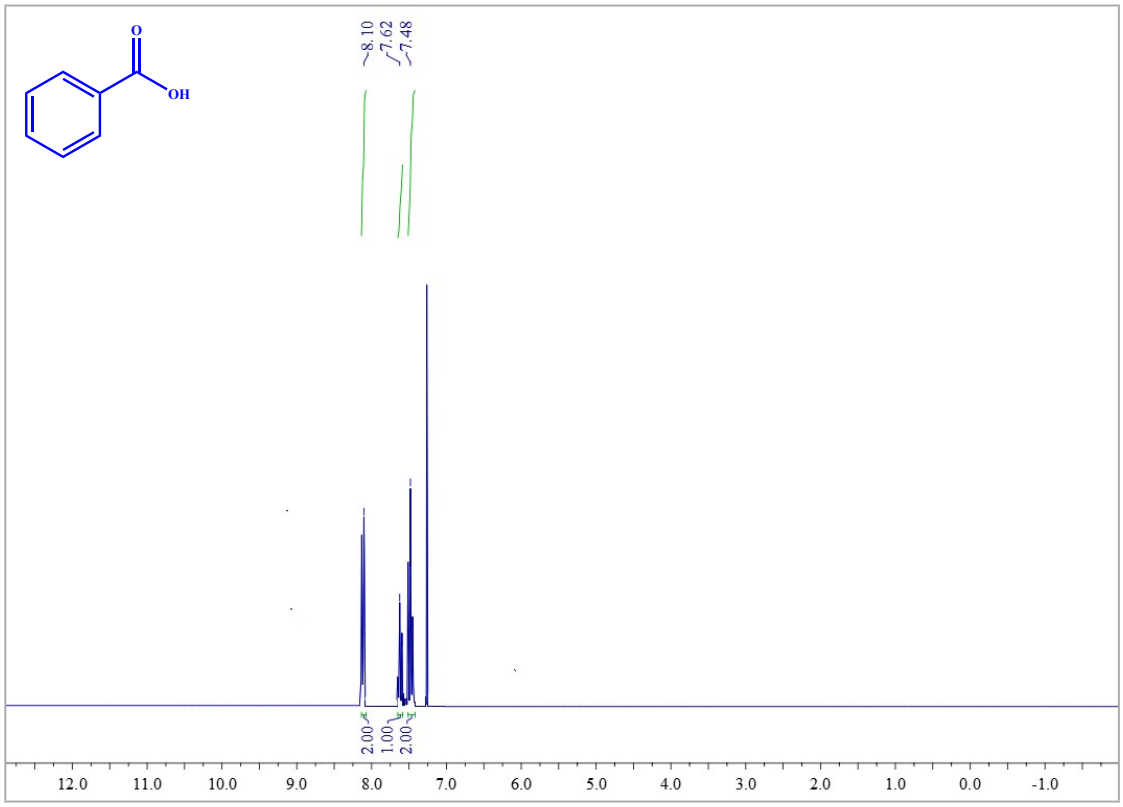
1. Acetophenone

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 7.96-7.94 (m, 2H), 7.56-7.43 (m, 3H), 2.5 (s, 3H).



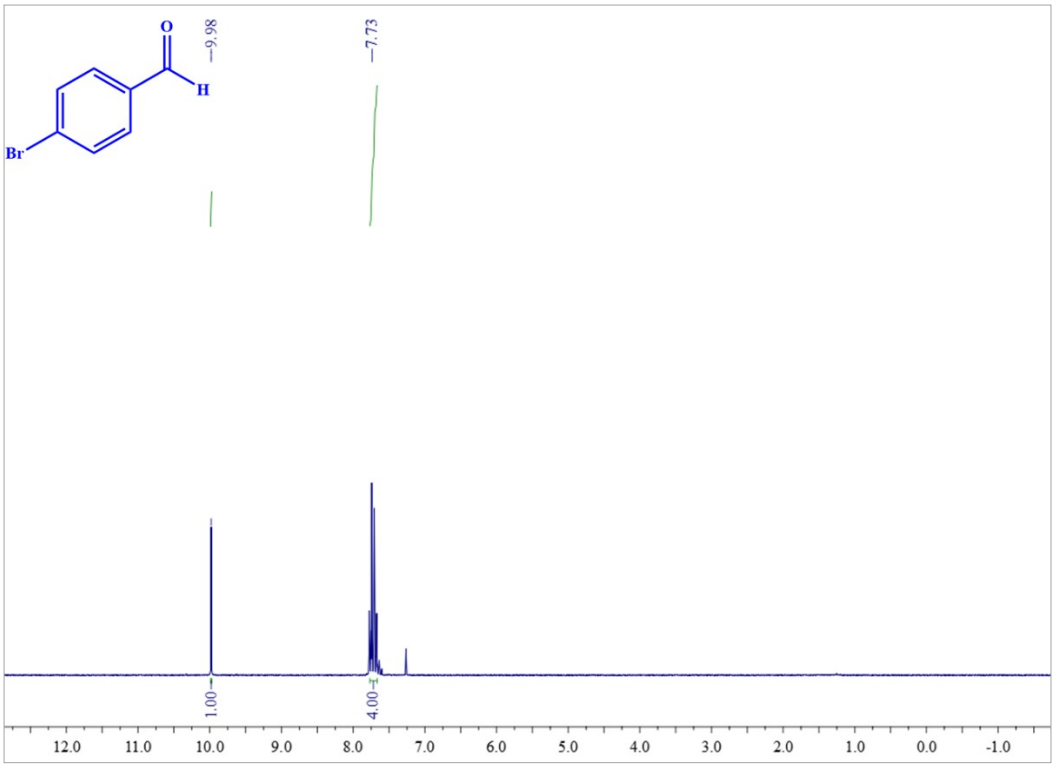
2. Benzoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 8.1 (d, *J*= 8 Hz, 2H), 7.62 (t, *J*= 8 Hz, 1H), 7.48 (t, *J*= 8 Hz, 2H).



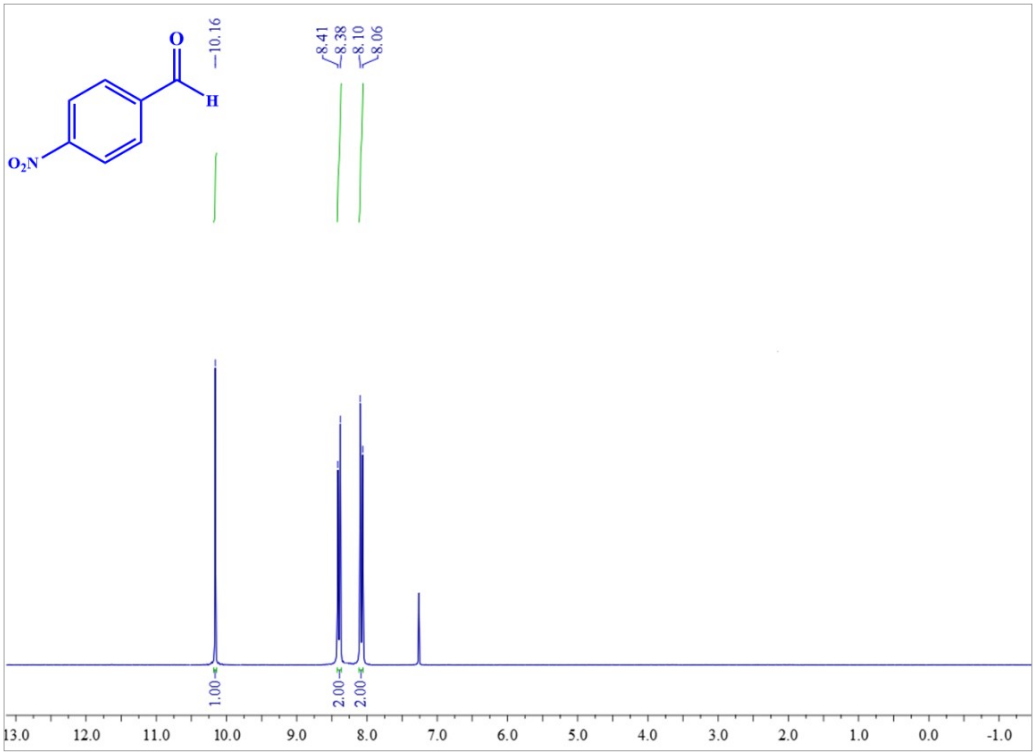
3. 4-Bromobenzaldehyde

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 9.98 (s, 1H), 7.73-7.64 (m, 4H).



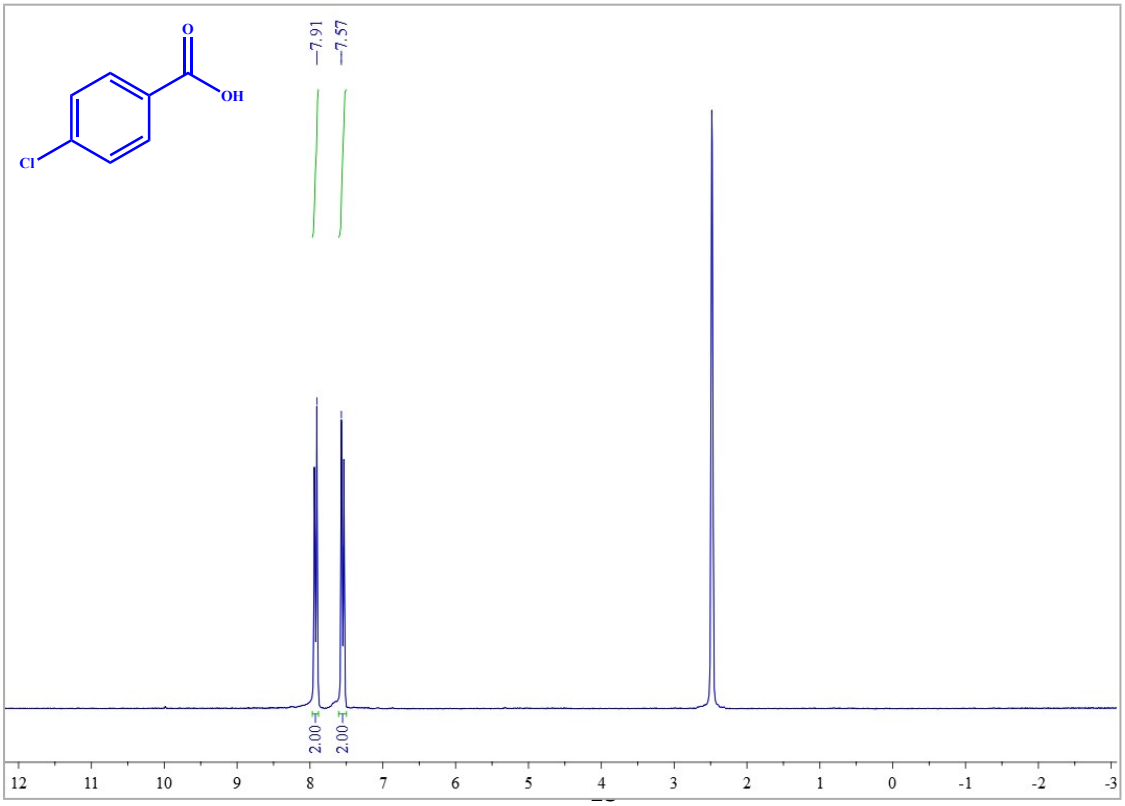
4. 4-Nitrobenzaldehyde

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 10.16 (s, 1H), 8.41-8.38 (d, d, *J*= 8 Hz, 2H), 8.10-8.06 (d, *J*= 8 Hz, 2H).



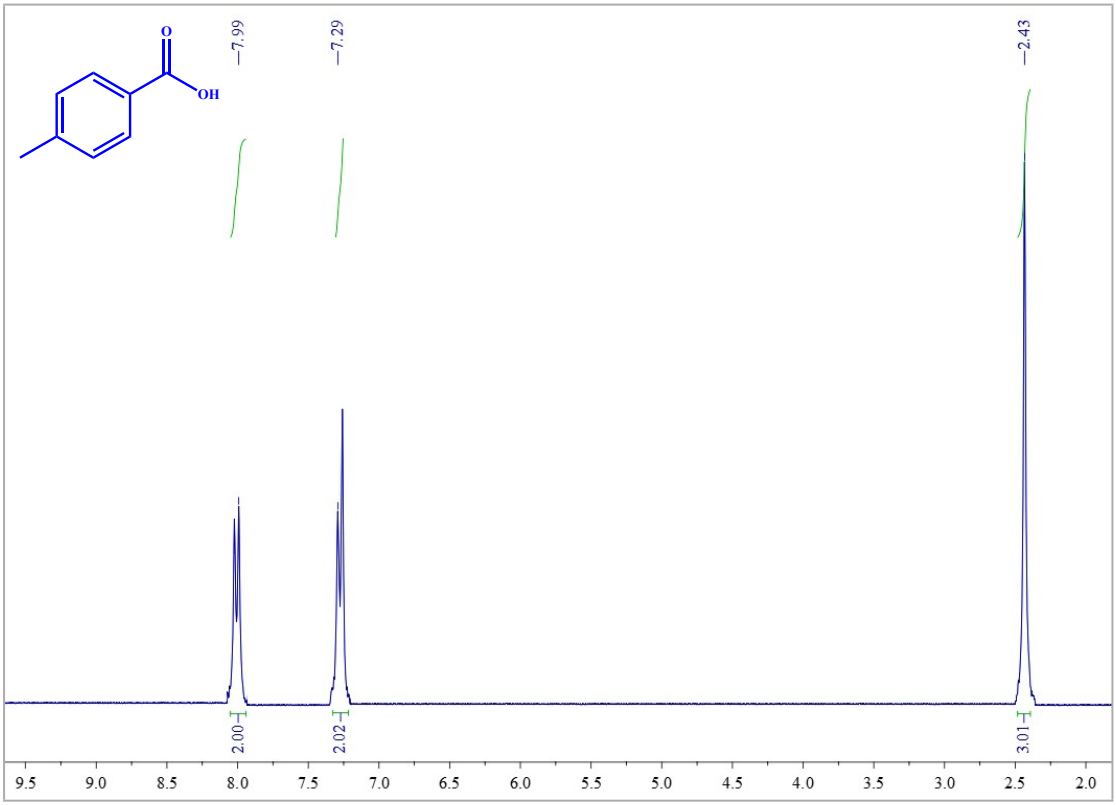
5. 4-Chlorobenzoic acid

^{1}H NMR (250 MHz, d_6-DMSO): δ (ppm) = 7.91 (d, *J*= 8 Hz, 2H), 7.57 (d, *J*= 8 Hz, 2H).



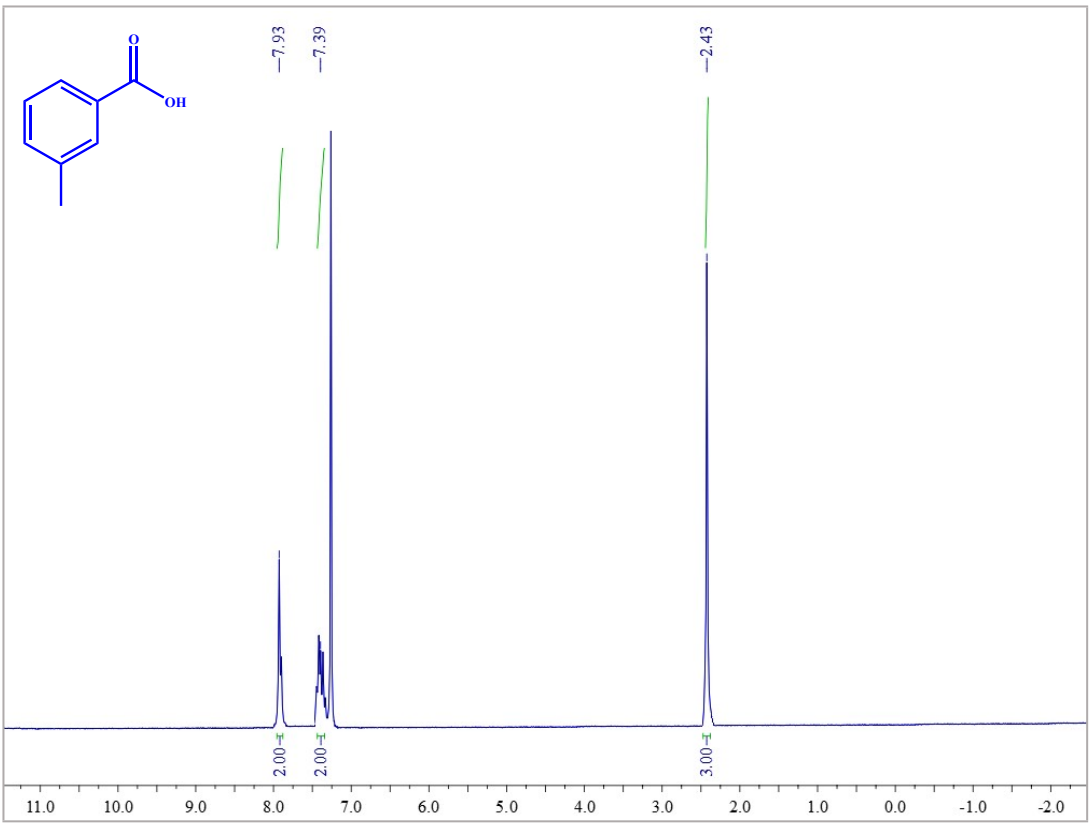
6. 4-Methylbenzoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 7.99 (d, *J*= 8 Hz, 2H), 7.629 (d, *J*= 8 Hz, 2H), 2.43 (s, 3H).



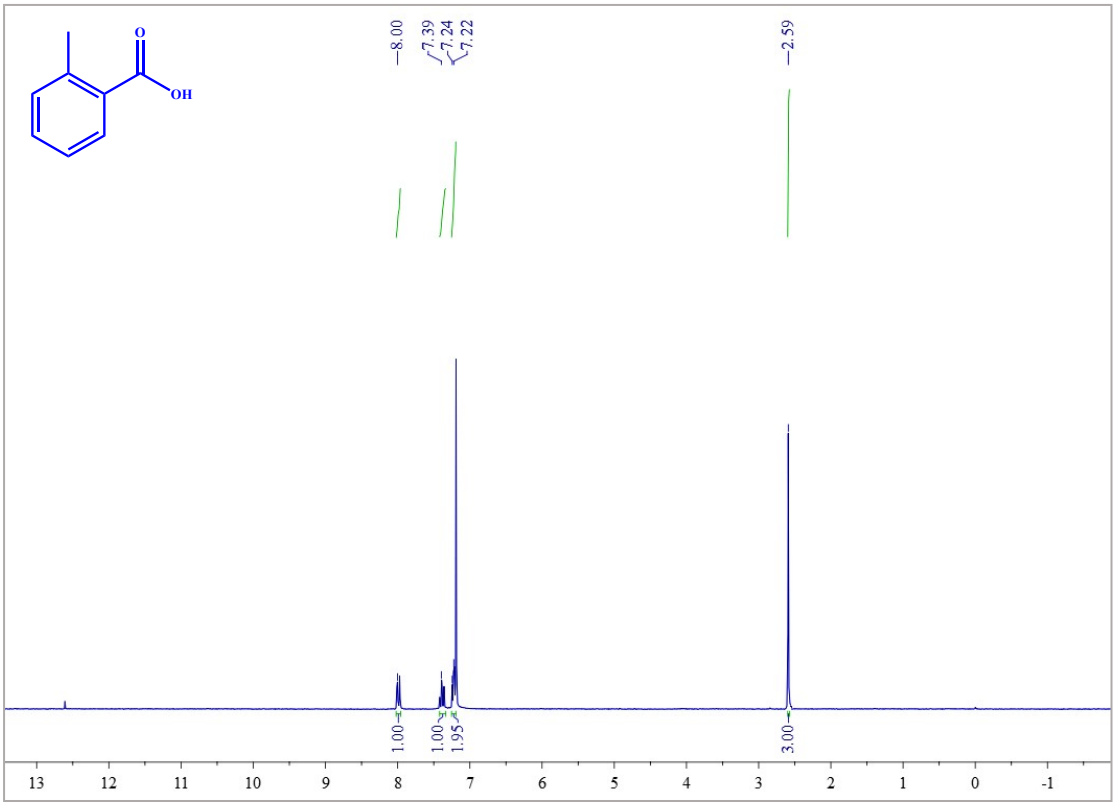
7. 3-Methylbenzoica acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 7.93 (d, *J*= 8 Hz, 2H), 7.39 (m, 2H), 2.43 (s, 3H).



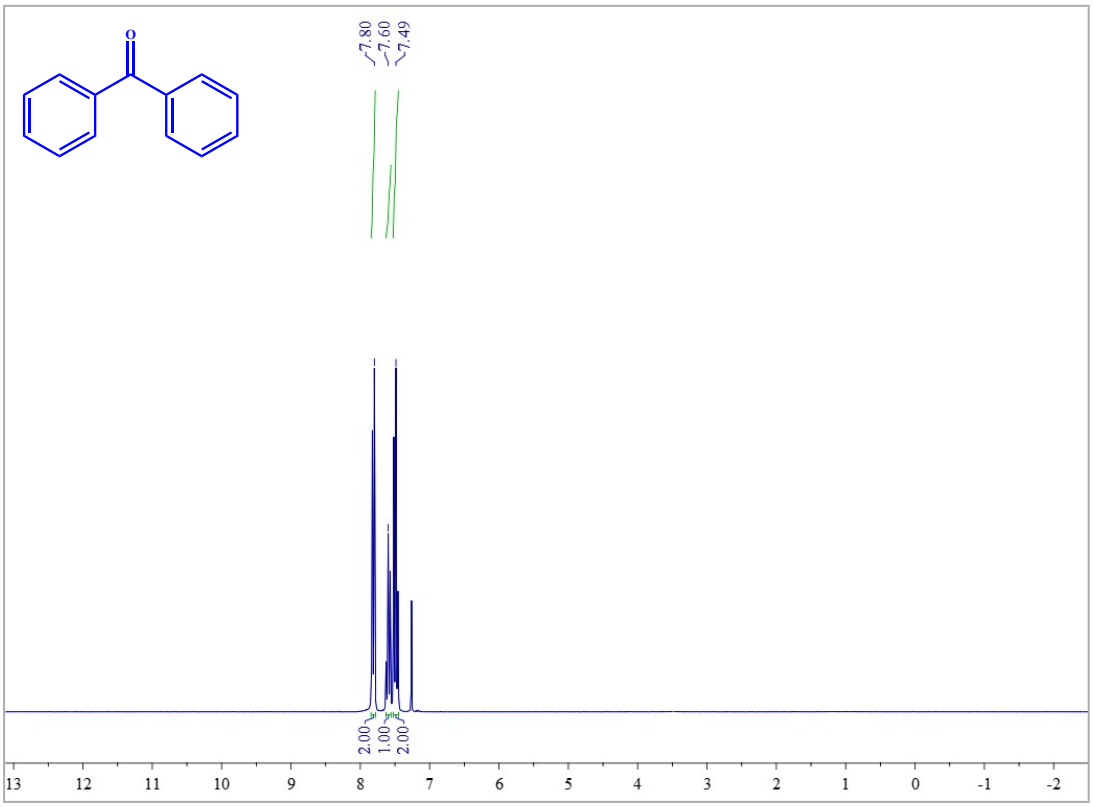
8. 2-Methylbenzoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 8 (d, *J*= 8 Hz, 1H), 7.39-7.22 (m, 3H), 2.59 (s, 3H).



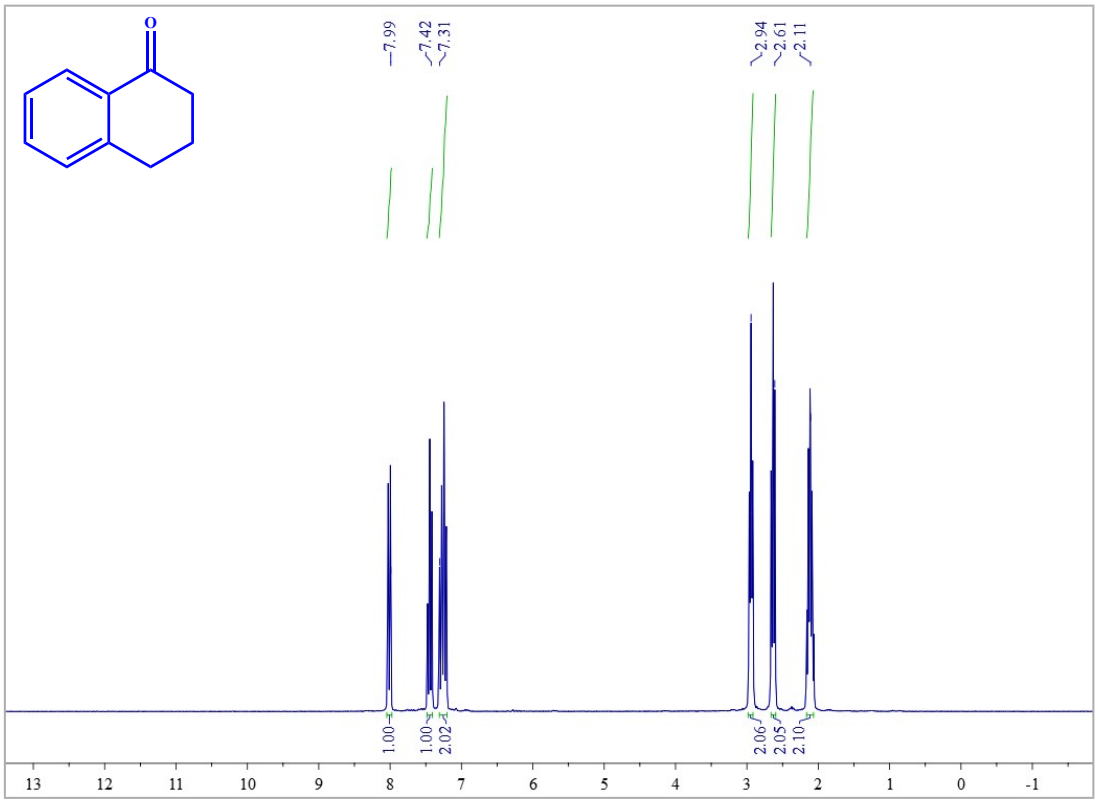
9. Benzophenone

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 7.80 (m, 2H), 7.60-7.49 (m, 3H).



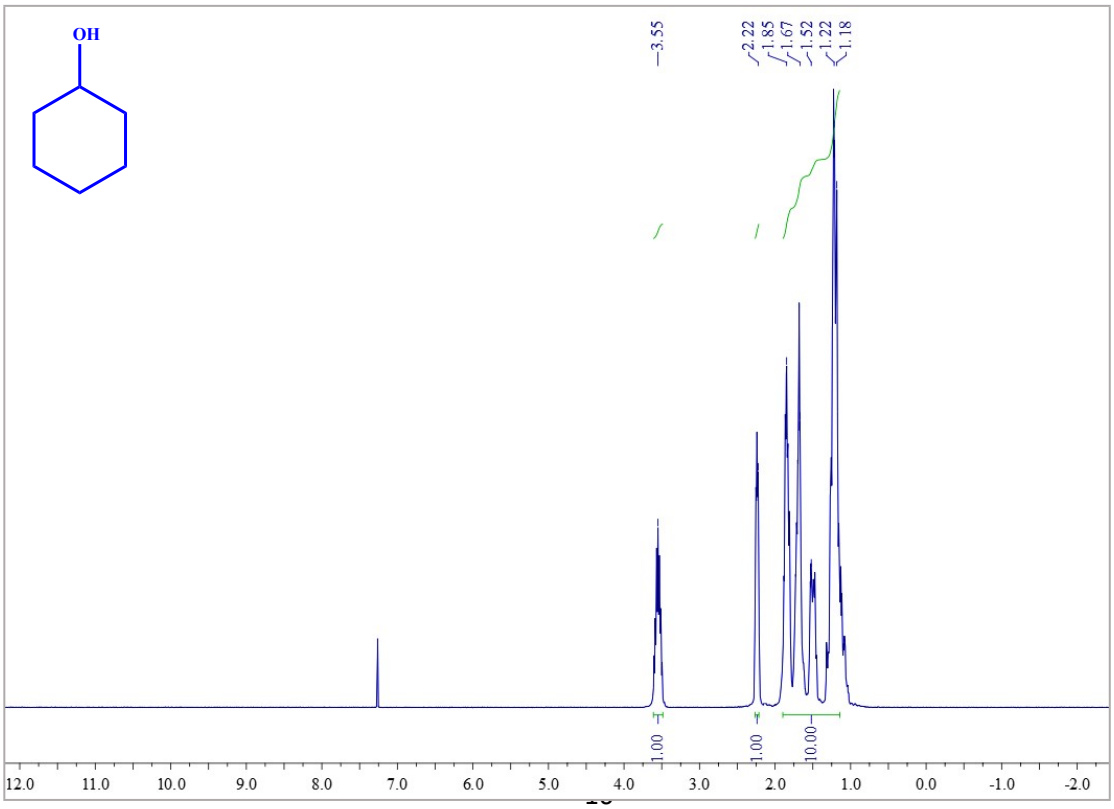
10. 1-Tetralone

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 7.99 (d, *J*= 8 Hz, 1H), 7.42 (m, 1H), 7.31 (m, 2H), 2.94 (m, 2H), 2.61 (m, 2H), 2.11 (m, 2H).



11. Cyclohexanol

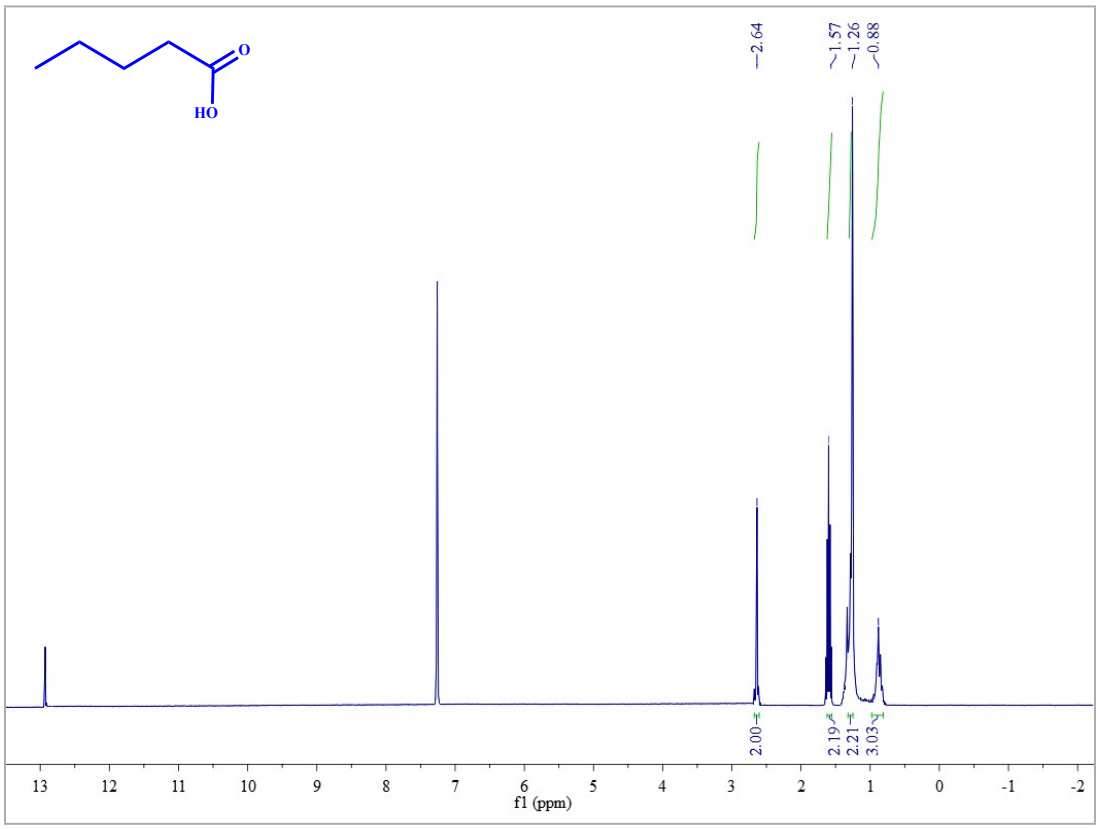
^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 3.55 (m, 1H), 2.22 (s, 1H), 1.18-1.55 (m, 10H).



C=C bond oxidation

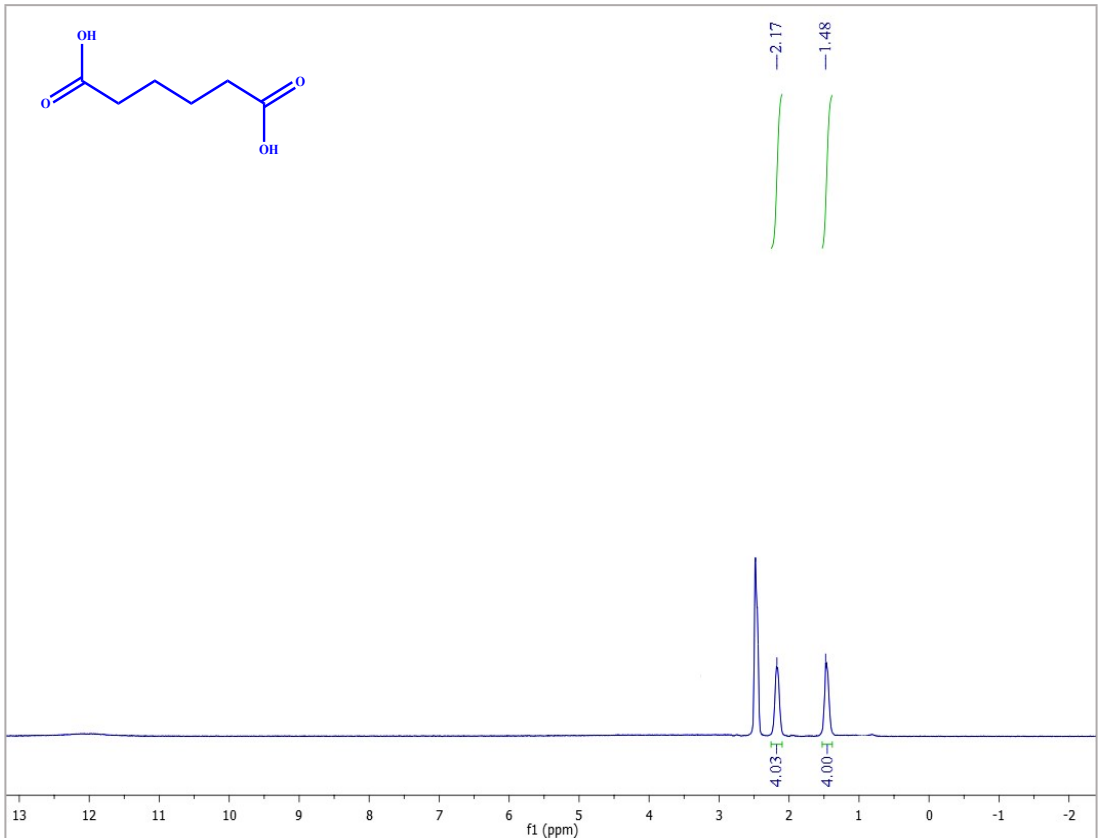
1. 1-Pentanoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 2.64 (m, 2H), 1.57 (m, 2H), 1.26 (m, 2H), 0.88 (m, 3H).



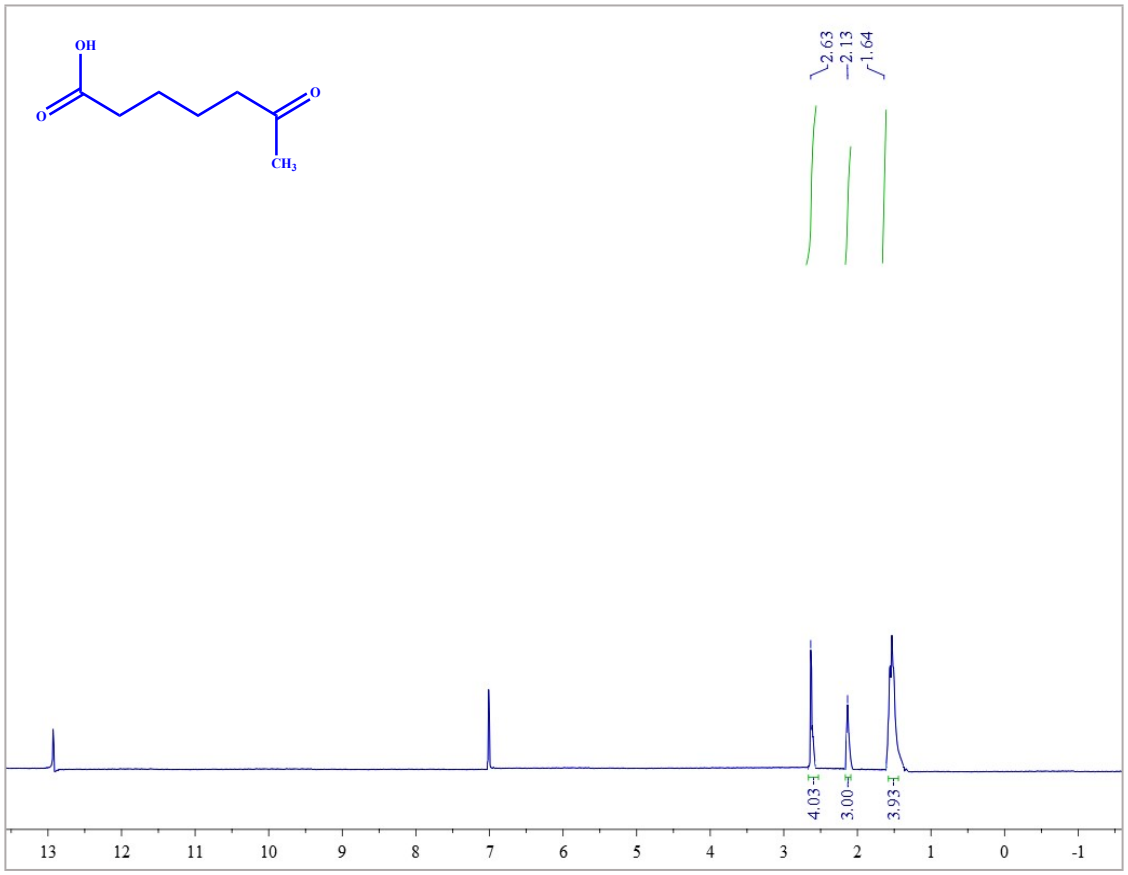
2. Adipic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 2.17 (m, 4H), 1.48 (m, 4H).



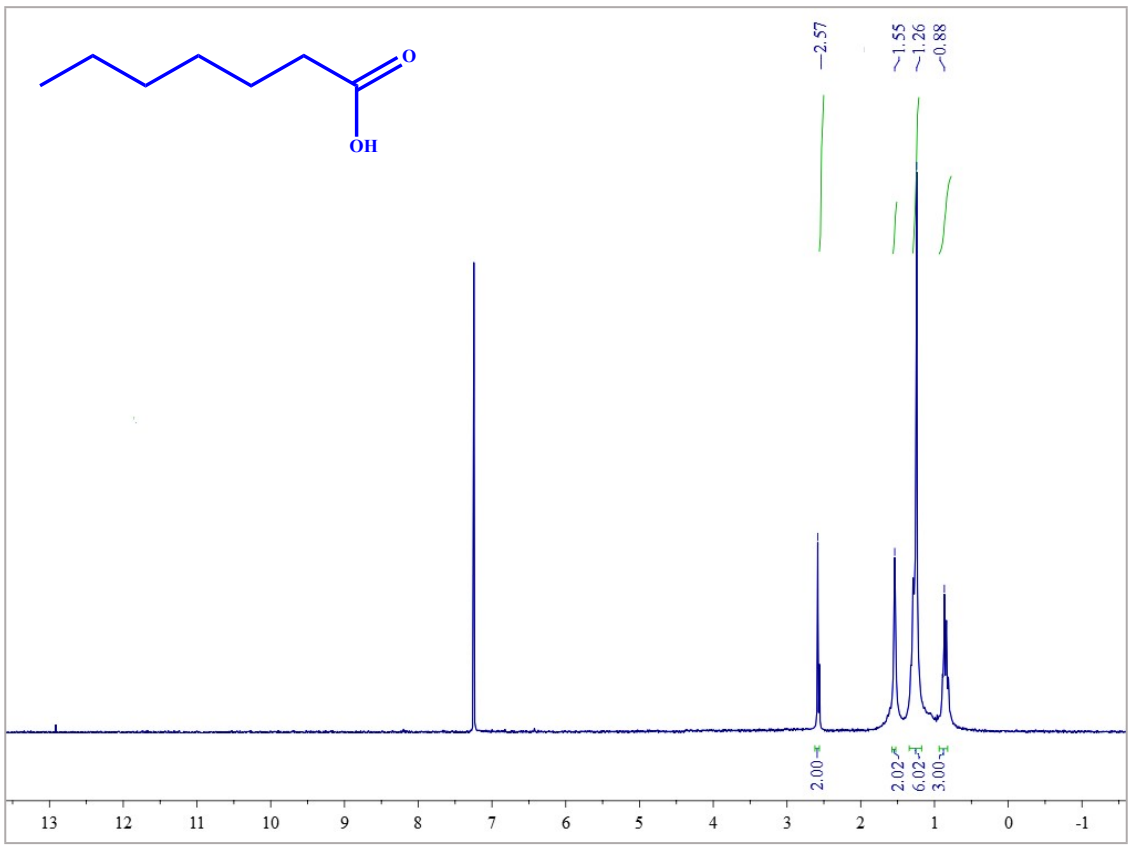
3. 6-Oxoheptanoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 2.63 (m, 4H), 2.13 (s, 3H), 1.64 (m, 4H).



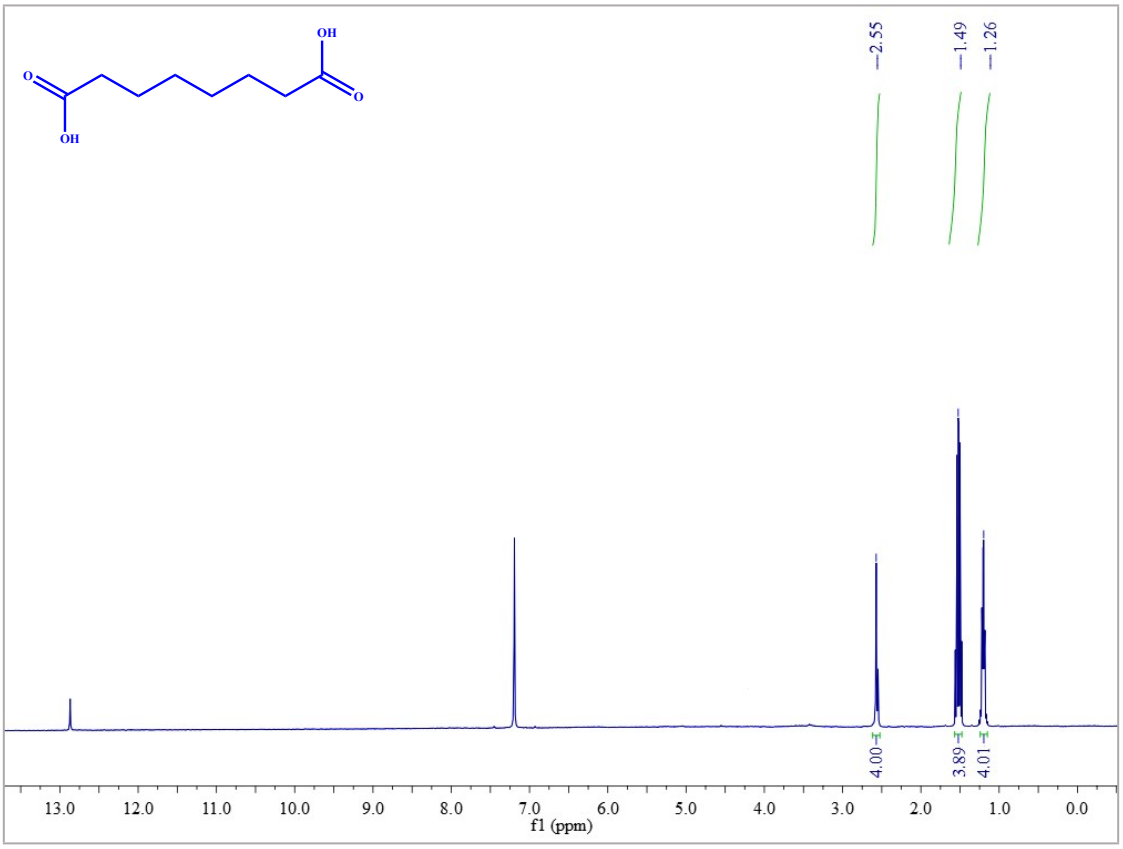
4. Heptanoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 2.57 (m, 2H), 1.55 (m, 2H), 1.26 (m, 6H), 0.88 (m, 3H).



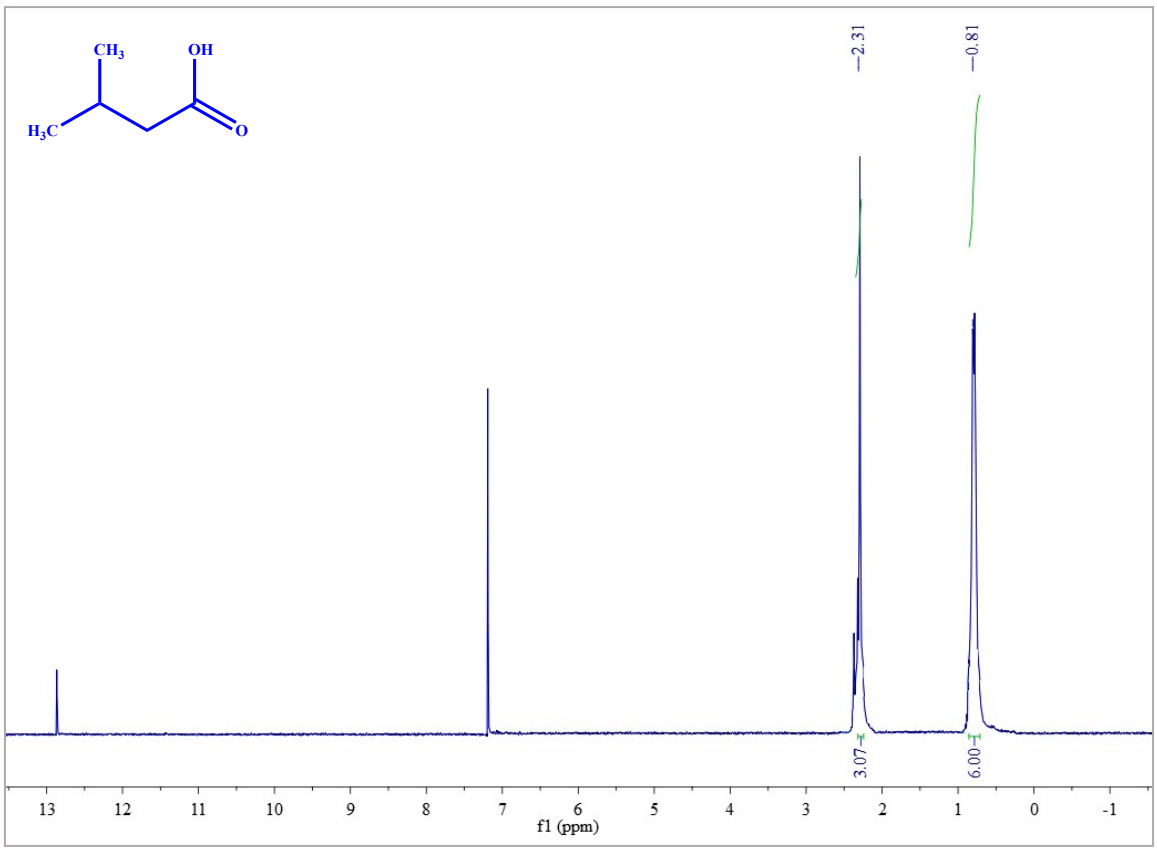
5. Octanedioic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 2.55 (m, 4H), 1.49 (m, 4H), 1.26 (m, 4H).



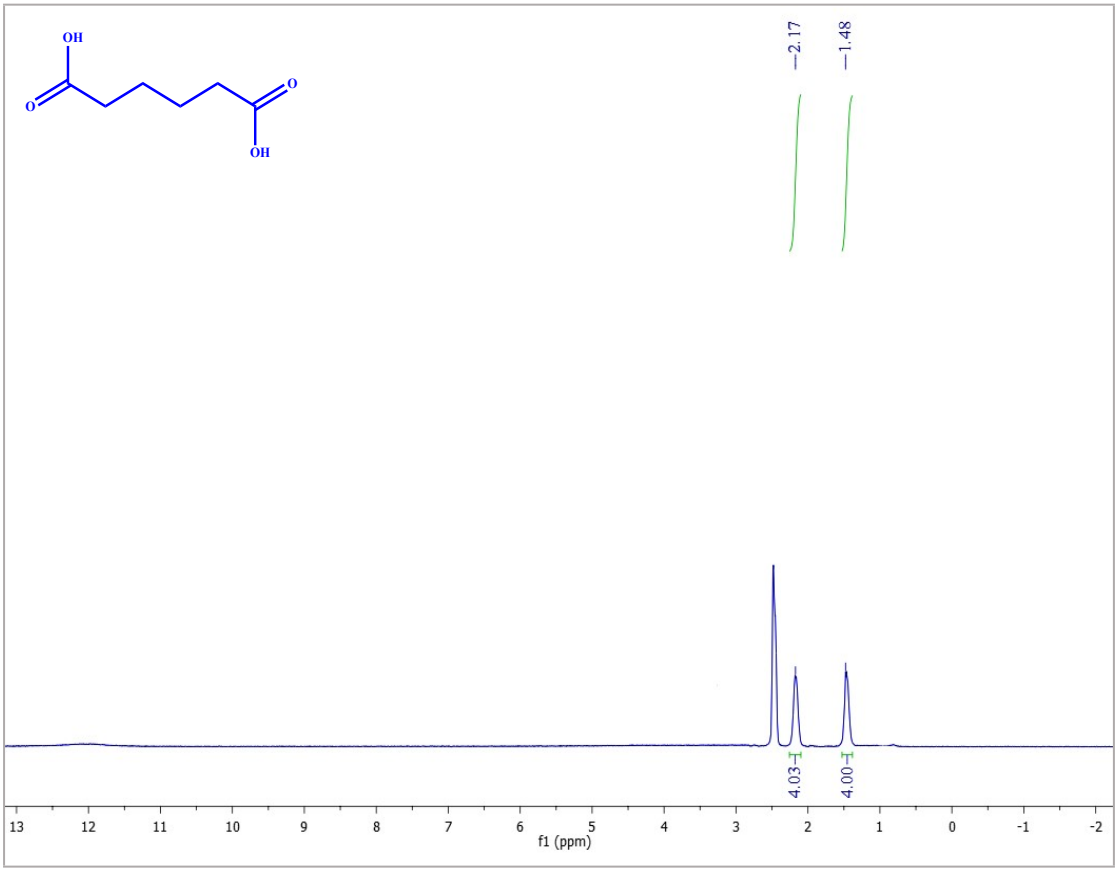
6. 3-Methylbutanoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 2.31 (m, 3H), 0.81 (m, 6H).



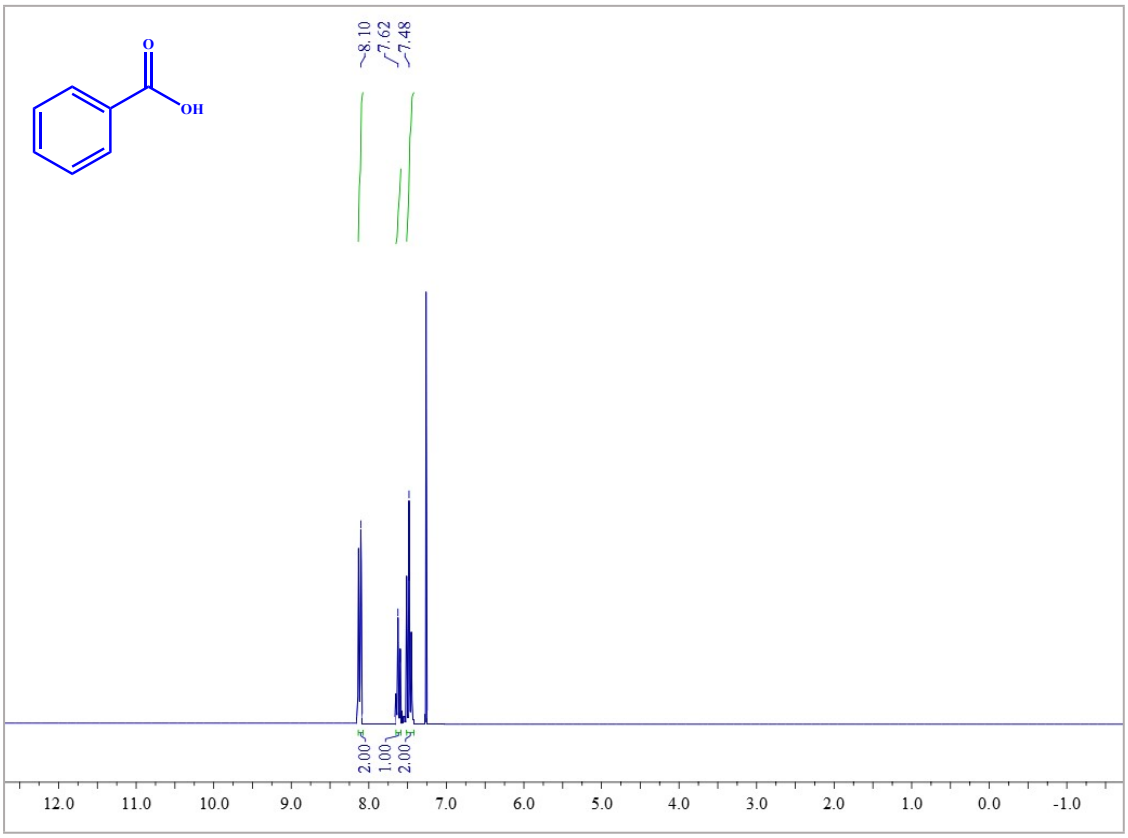
7. Adipic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 2.17 (m, 4H), 1.48 (m, 4H).



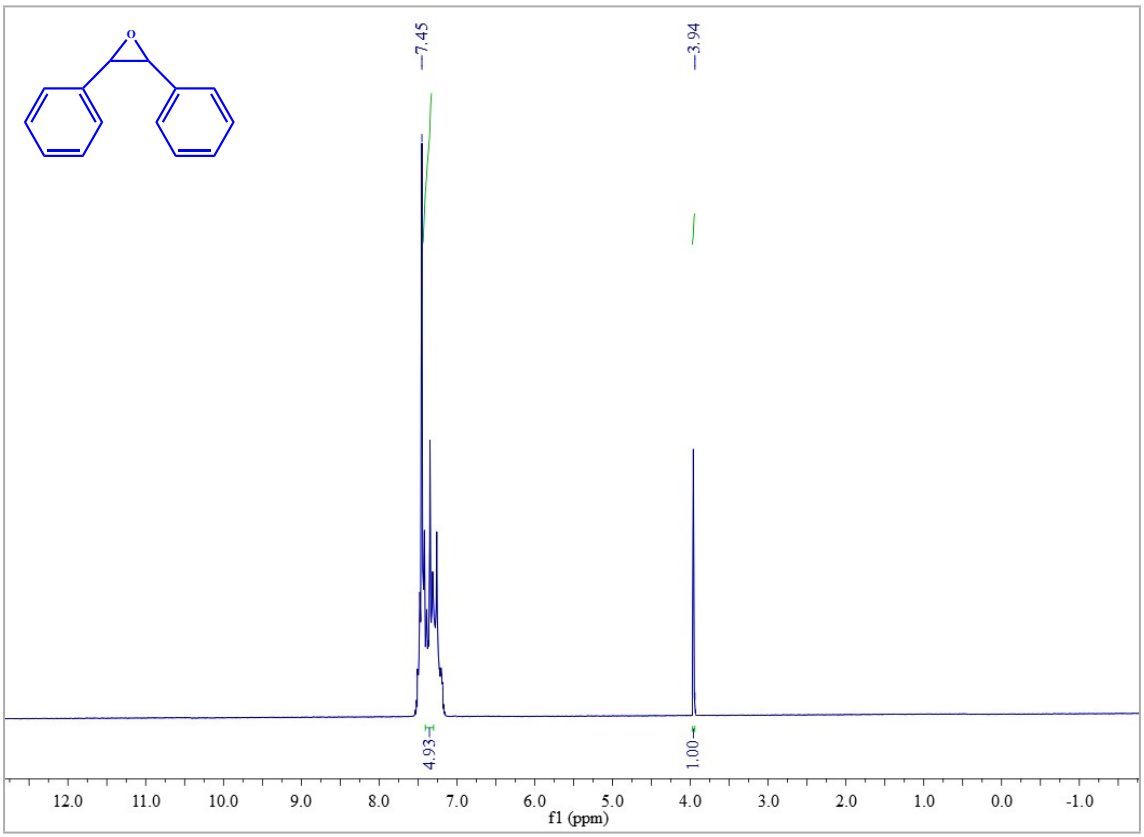
8. Benzoic acid

^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 8.1 (d, *J*= 8 Hz, 2H), 7.62 (t, *J*= 8 Hz, 1H), 7.48 (t, *J*= 8 Hz, 2H).



9. Cis-stilben oxide

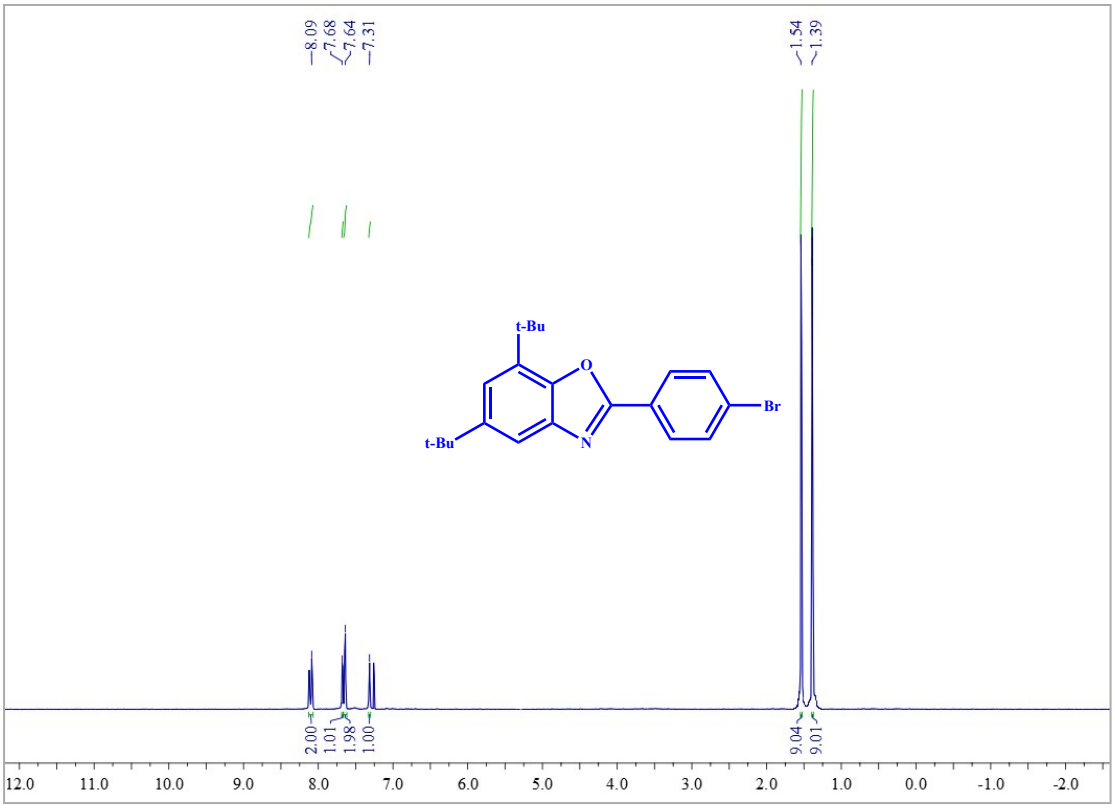
^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 7.45 (m, 10H), 3.94 (s, 2H).



Benzoxazoles NMR

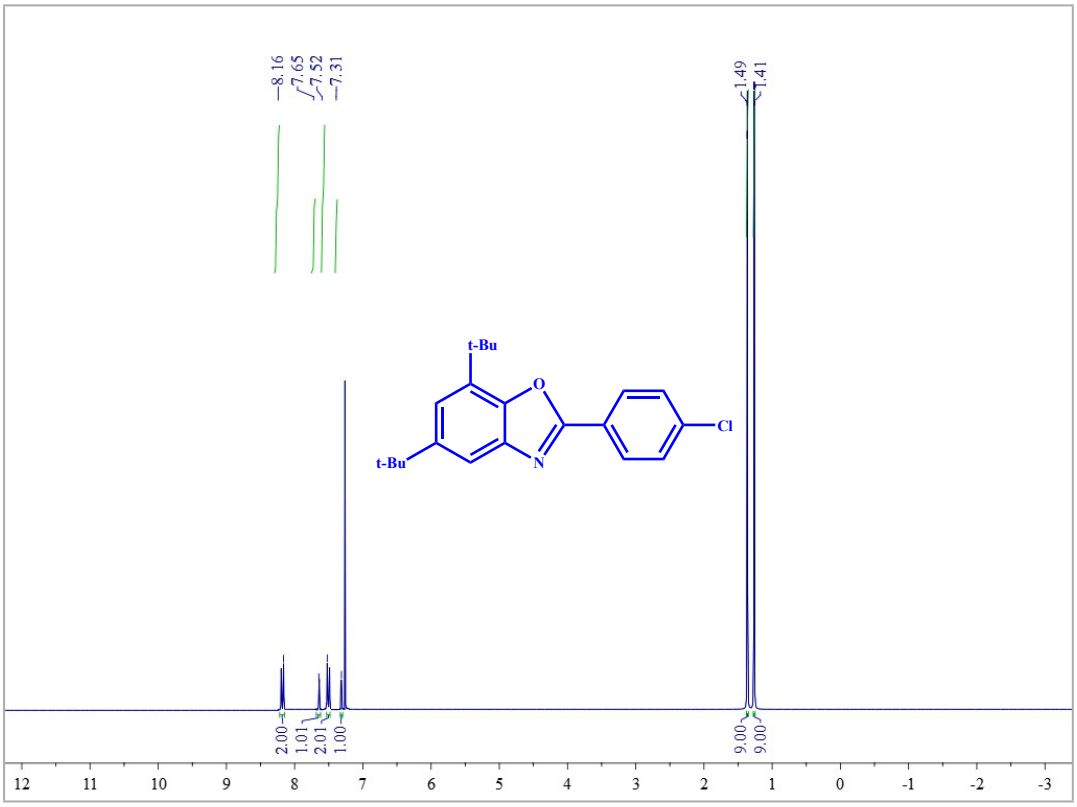
1. 5,7-Di-tert-butyl-2-(4-bromophenyl)benzo[d]oxazole

Refined by column chromatography on silica gel and laundered with ethyl acetate/n-hexane (1:100). Isolated yield: 327 mg, 97%. Pale yellow liquid; ^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 1.39 (s, 9H), 1.54 (s, 9H), 7.31 (d, *J*= 2.5 Hz, 1H), 7.64 (d, *J*= 7.5 Hz, 2H), 7.68 (d, *J*= 2.5 Hz, 1H), 8.09 (d, *J*= 7.5 Hz, 2H).



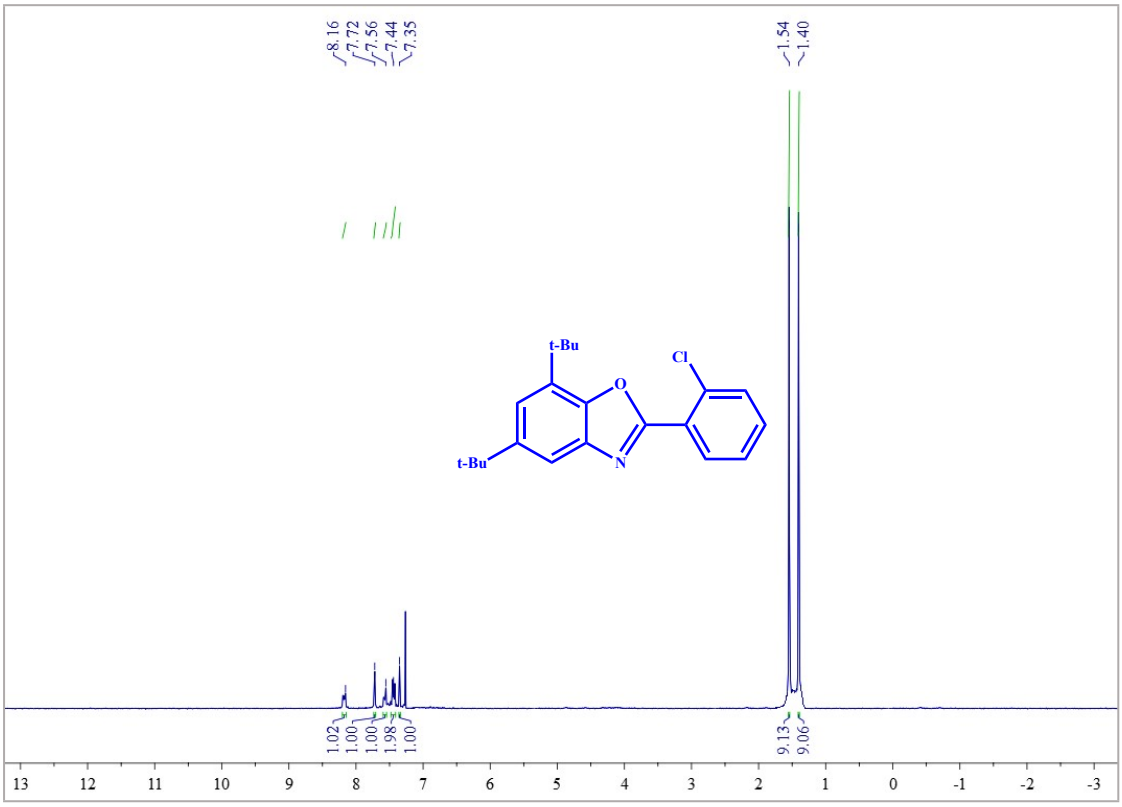
2. 5,7-Di-tert-butyl-2-(4-chlorophenyl)benzo[d]oxazole

Refined by column chromatography on silica gel and laundered with ethyl acetate/n-hexane (1:100). Isolated yield: 321 mg, 94%. Colorless liquid; ^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 1.41 (s, 9H), 1.49 (s, 9H), 7.31 (d, *J*= 2.5 Hz, 1H), 7.65 (d, *J*= 2.5 Hz, 1H), 7.52 (d, *J*= 7.5 Hz, 2H), 8.16 (d, *J*= 7.5 Hz, 2H).



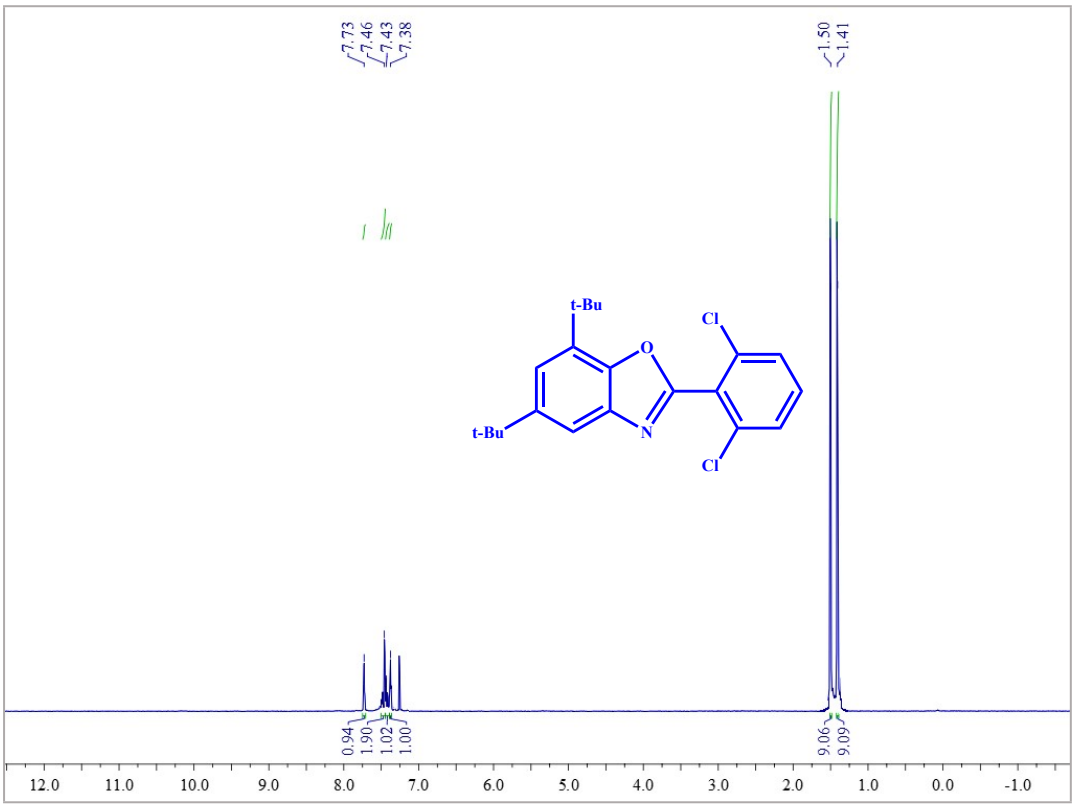
3. 5,7-Di-tert-butyl-2-(2-chlorophenyl)benzo[d]oxazole

Refined by column chromatography on silica gel and laundered with ethyl acetate/n-hexane (1:100). Isolated yield: 321 mg, 92%. Colorless liquid; ^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 1.40 (s, 9H), 1.50 (s, 9H), 7.35 (d, *J*= 2.5 Hz, 1H), 7.44 (m, 2H), 7.56 (d, *J*= 7.5 Hz, 1H), 7.72 (d, *J*= 2.5 Hz, 1H), 8.16 (d, *J*= 7.5 Hz, 1H).



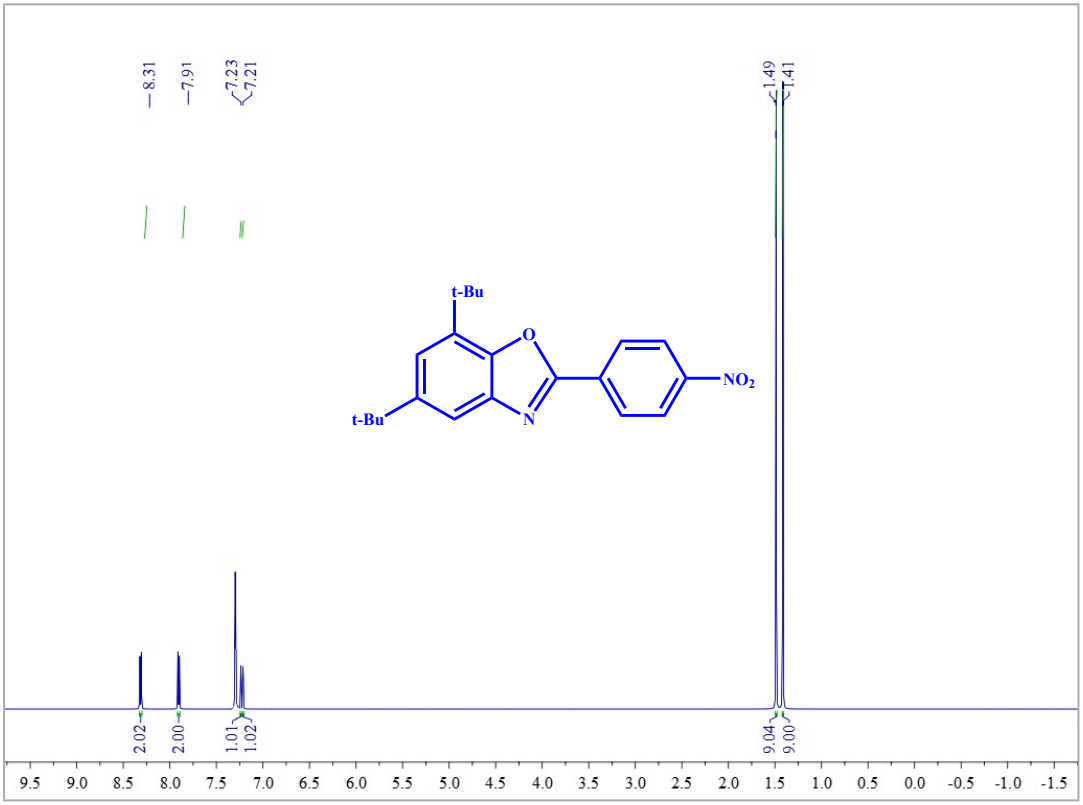
4. 5,7-Di-tert-butyl-2-(2,6-dichlorophenyl)benzo[d]oxazole

Refined by column chromatography on silica gel and laundered with ethyl acetate/n-hexane (1:100). Isolated yield: 330 mg, 90%. Colorless liquid; ^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 1.41 (s, 9H), 1.50 (s, 9H), 7.38-7.46 (m, 4H), 7.73 (d, *J*= 2.5 Hz, 1H).



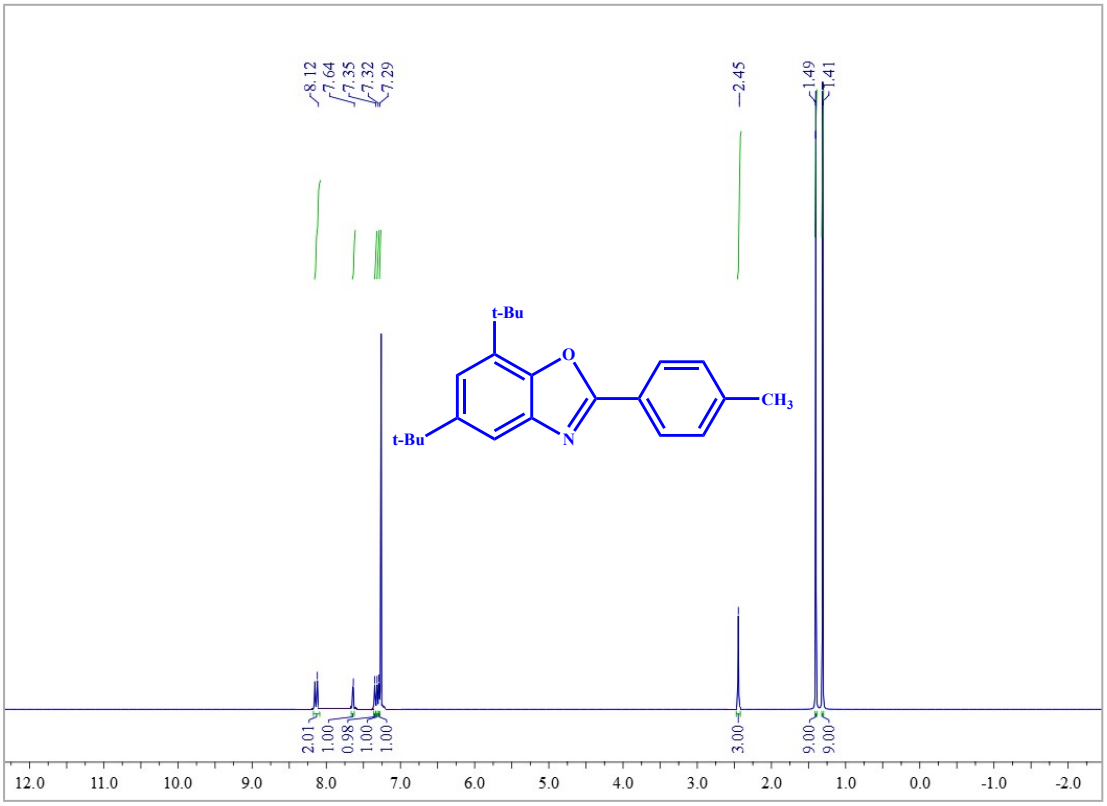
5. 5,7-Di-tert-butyl-2-(4-nitrophenyl)benzo[d]oxazole

Refined by column chromatography on silica gel and laundered with ethyl acetate/n-hexane (1:100). Isolated yield: 287 mg, 88%. Pale yellow liquid; ^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 1.41 (s, 9H), 1.49 (s, 9H), 7.21 (d, *J*= 2.5 Hz, 1H), 7.23 (d, *J*= 2.5 Hz, 1H), 7.91 (d, *J*= 7.5 Hz, 2H), 8.31 (d, *J*= 7.5 Hz, 2H).



6. 5,7-Di-tert-butyl-2-(p-tolyl)benzo[d]oxazole

Refined by column chromatography on silica gel and laundered with ethyl acetate/n-hexane (1:100). Isolated yield: 293 mg, 80%. Pale yellow liquid; ^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 1.41 (s, 9H), 1.49 (s, 9H), 2.45 (s, 3H), 7.29 (d, *J*= 4 Hz, 1H), 7.32-7.35 (d, *J*= 8 Hz, 2H), 7.64 (d, *J*= 4 Hz, 1H), 8.12 (d, *J*= 8 Hz, 2H).



7. 5,7-Di-tert-butyl-2-(4-methoxyphenyl)benzo[d]oxazole

Refined by column chromatography on silica gel and laundered with ethyl acetate/n-hexane (1:100). Isolated yield: 154 mg, 78%. Colorless liquid; ^{1}H NMR (250 MHz, $CDCl_3$): δ (ppm) = 1.41 (s, 9H), 1.49 (s, 9H), 3.81 (s, 3H), 7.05 (d, *J*= 9 Hz, 2H), 7.17 (s, 1H), 7.21 (s, 1H), 7.63 (d, *J*= 9Hz, 2H).