Supporting Information

AN IONIC LIQUID-BASED GREEN SYNTHESIS STRATEGY: SYNTHESIS OF DIHYDROPYRIMIDINONES BY THREE-COMPONENT BIGINELLI-TYPE REACTION OF ALIPHATIC ALDEHYDES, AROMATIC ALDEHYDES AND UREA

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1. The recycle of IL

In order to implement the concept of green chemistry and reduce energy consumption, the recycling of ionic liquids was also examined. The results are shown in Table S1. As the number of cycles increased from 1 to 4, the yield decreased only slightly, which shows that the ionic liquid still has a catalytic effect on the reaction after the cycle.

Table S1. The recycle of IL[a]

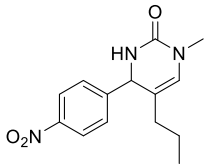
Entry	Cycle numbers	Yield[b](%)
1	1	85
2	2	84
3	3	83
4	4	79

[a]Reaction conditions: 4-nitrobenzaldehyde (1 mmol), *n*-hexanal (1.5 mmol), methylurea (1.5 mmol), 90℃, 6 h, 1 mL 30% IL aqueous solution. [b]Yields of pure products isolated by chromatography.

2. Characterization of compounds

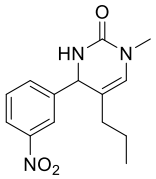
2.1 ^{1}H-NMR, ^{13}C-NMR data of DHPMs products

1-methyl-4-(4-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



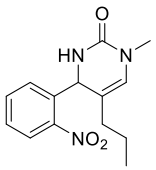
4a: 81% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.20 (d, *J* = 8.5 Hz, 2H), 7.46 (d, *J* = 8.5 Hz, 2H), 5.82 (s, 1H), 5.46 (s, 1H), 5.03 (s, 1H), 3.08 (s, 3H), 1.86 – 1.54 (m, 2H), 1.34 – 1.11 (m, 2H), 0.85 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.48, 149.99, 147.72, 129.37, 127.99, 125.21, 124.23, 59.06, 39.21, 31.63, 22.63, 14.15.

1-methyl-4-(3-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



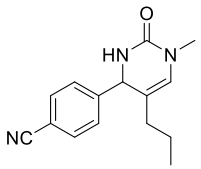
4b: 83% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.09 (dt, *J* = 3.5, 1.5 Hz, 2H), 7.59 (d, *J* = 7.8 Hz, 1H), 7.48 (t, *J* = 7.9 Hz, 1H), 5.79 (s, 1H), 5.42 (s, 1H), 4.99 (s, 1H), 3.03 (s, 3H), 1.65 (t, *J* = 8.8 Hz, 2H), 1.41 – 1.27 (m, 2H), 0.79 (t, *J* = 5.1 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.50, 150.01, 147.74, 129.39, 128.01, 124.25, 123.70, 113.02, 59.08, 34.48, 31.65, 22.65, 14.17.

1-methyl-4-(2-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



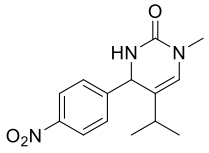
4c: 83% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.86 (d, *J* = 7.1 Hz, 1H), 7.59 (t, *J* = 7.6 Hz, 1H), 7.52 (d, *J* = 7.8 Hz, 1H), 7.40 (t, *J* = 7.7 Hz, 1H), 6.01 (s, 1H), 5.57 (s, 1H), 5.27 (s, 1H), 3.03 (s, 3H), 1.71 – 1.65 (m, 2H), 1.26 – 1.16 (m, 2H), 0.79 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.48, 149.99, 147.72, 129.37, 127.99, 125.21, 124.23, 113.00, 59.06, 34.46, 31.63, 22.63, 14.15.

4-(1-methyl-2-oxo-5-propyl-1, 2, 3, 4-tetrahydropyrimidin-4-yl)benzonitrile



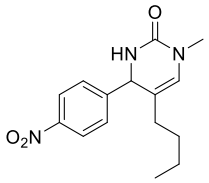
4d: 84% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.53 (d, *J* = 7.6 Hz, 2H), 7.30 (t, *J* = 17.5 Hz, 2H), 6.37 (s, 1H), 5.72 (s, 1H), 4.86 (s, 1H), 2.90 (s, 3H), 1.60 (t, *J* = 14.3 Hz, 2H), 1.25 (ddd, *J* = 20.6, 13.2, 6.7 Hz, 2H), 0.76 (t, *J* = 6.9 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.51, 147.75, 129.40, 128.02, 125.24, 124.26, 116.45, 113.03, 59.09, 34.49, 31.66, 22.66, 14.18.

5-isopropyl-1-methyl-4-(4-nitrophenyl)-3,4-*dihydropyrimidi*n-2(1*H*)-one



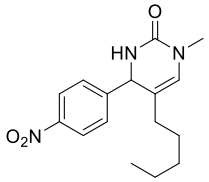
4e: 81% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.11 (d, *J* = 8.7 Hz, 2H), 7.40 (d, *J* = 8.7 Hz, 2H), 6.40 (s, 1H), 5.81 (s, 1H), 5.00 (s, 1H), 3.01 (s, 3H), 1.89 (dt, *J* = 13.5, 6.7 Hz, 1H), 0.93 (d, *J* = 6.8 Hz, 3H), 0.89 (d, *J* = 6.8 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) 153.49, 150.00, 147.73, 129.38, 128.00, 125.22, 124.24, 59.07, 34.47, 31.64, 22.64.

5-butyl-1-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one



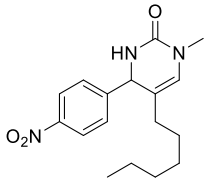
4f: 85% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.13 (d, *J* = 8.7 Hz, 2H), 7.40 (d, *J* = 8.7 Hz, 2H), 5.79 (s, 1H), 5.75 (s, 1H), 4.97 (s, 1H), 2.99 (s, 3H), 1.67 (t, *J* = 11.9 Hz, 2H), 1.23 – 1.11 (m, 4H), 0.78 (t, *J* = 6.2 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.48, 149.99, 147.72, 127.99, 125.21, 124.23, 123.68, 59.06, 34.46, 31.63, 30.34, 22.63, 14.15.

1-methyl-4-(4-nitrophenyl)-5-pentyl-3,4-dihydropyrimidin-2(1H)-one



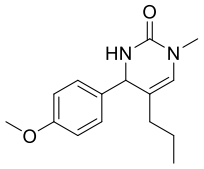
4g: 79% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.09 (d, *J* = 8.6 Hz, 2H), 7.39 (d, *J* = 8.5 Hz, 2H), 6.01 (s, 1H), 5.75 (s, 1H), 4.97 (s, 1H), 2.98 (s, 3H), 1.64 (t, *J* = 7.5 Hz, 2H), 1.09 (d, *J* = 10.0 Hz, 6H), 0.76 (t, *J* = 6.9 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.48, 149.99, 147.72, 127.99, 125.21, 124.23, 113.00, 59.06, 34.46, 31.63, 30.34, 26.99, 22.63, 14.15.

5-hexyl-1-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one



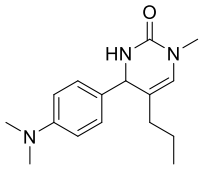
4h: 77% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.18 (d, *J* = 8.4 Hz, 2H), 7.44 (d, *J* = 8.4 Hz, 2H), 5.88 (s, 1H), 5.80 (s, 1H), 5.02 (s, 1H), 3.05 (s, 3H), 1.70 (t, *J* = 7.2 Hz, 2H), 1.18 (d, *J* = 7.3 Hz, 8H), 0.83 (t, *J* = 6.9 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.40, 149.91, 147.64, 127.91, 125.13, 124.15, 112.92, 58.98, 34.38, 31.56, 30.27, 28.78, 26.91, 22.55, 14.07.

4-(4-methoxyphenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



4i: 85% Yellow oily liquid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.12 (d, *J* = 8.6 Hz, 2H), 6.78 (d, *J* = 8.6 Hz, 2H), 5.68 (s, 1H), 5.11 (s, 1H), 4.80 (s, 1H), 3.72 (s, 3H), 2.99 (s, 3H), 1.69 – 1.56 (m, 2H), 1.31 (dd, *J* = 14.6, 7.2 Hz, 2H), 1.14 (t, *J* = 7.0 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.46, 149.97, 129.35, 127.97, 125.19, 124.21, 112.98, 59.04, 54.17, 34.44, 31.61, 22.61, 14.13.

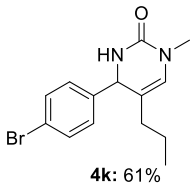
4-(4-(dimethylamino)phenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



4j: 86% Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.05 (d, *J* = 8.7 Hz, 2H), 6.60 (d, *J* = 8.7 Hz, 2H), 5.66 (s, 1H), 5.09 (s, 1H), 4.75 (s, 1H), 2.99 (s, 3H), 2.86 (s, 6H), 1.70 – 1.54 (m, 2H), 1.26 – 1.12 (m, 2H), 0.77 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.48, 152.38,

129.37, 127.99, 125.21, 124.23, 113.00, 59.06, 39.02, 34.46, 31.63, 22.63, 14.15.

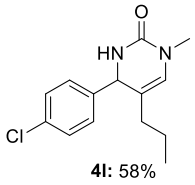
4-(4-bromophenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



4k: 61%

White solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.44 (d, *J* = 8.3 Hz, 2H), 7.14 (d, *J* = 8.3 Hz, 2H), 5.75 (s, 1H), 5.34 (s, 1H), 4.87 (s, 1H), 3.04 (s, 3H), 1.78 – 1.59 (m, 2H), 1.45 – 1.25 (m, 2H), 0.83 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.50, 147.74, 129.39, 128.01, 125.23, 124.25, 123.70, 59.08, 34.48, 31.65, 22.65, 14.17.

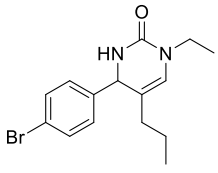
4-(4-chlorophenyl)-1-methyl-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



4l: 58%

White solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.30 (d, *J* = 8.4 Hz, 2H), 7.21 (d, *J* = 8.4 Hz, 2H), 5.76 (s, 1H), 5.23 (s, 1H), 4.89 (s, 1H), 3.06 (s, 3H), 1.69 (dd, *J* = 16.9, 8.7 Hz, 2H), 1.38 (dt, *J* = 14.8, 7.4 Hz, 2H), 0.83 (t, *J* = 7.3 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 153.48, 147.72, 129.37, 127.99, 125.21, 124.23, 123.68, 59.06, 34.46, 31.63, 22.63, 14.15.

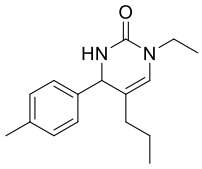
4-(4-bromophenyl)-1-ethyl-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one



4m: 63%

White solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.47 (d, *J* = 8.3 Hz, 2H), 7.16 (d, *J* = 8.5 Hz, 2H), 5.80 (s, 1H), 5.29 (s, 1H), 4.87 (s, 1H), 3.61 – 3.30 (m, 2H), 1.79 – 1.61 (m, 2H), 1.50 – 1.27 (m, 2H), 1.19 (t, *J* = 7.1 Hz, 3H), 0.86 (t, *J* = 7.4 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 152.74, 142.10, 131.90, 128.70, 123.16, 121.95, 113.61, 59.01, 41.62, 32.48, 20.23, 14.24, 13.67.

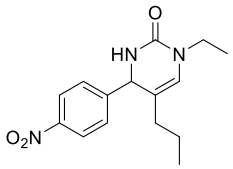
1-ethyl-5-propyl-4-(p-tolyl)-3,4-dihydropyrimidin-2(1*H*)-one



4n: 81%

Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.18 (d, *J* = 13.8 Hz, 4H), 5.79 (s, 1H), 4.87 (s, 1H), 3.94 (s, 1H), 3.62 – 3.40 (m, 2H), 2.35 (s, 3H), 1.85 – 1.63 (m, 2H), 1.48 – 1.30 (m, 2H), 1.22 (t, *J* = 7.1 Hz, 3H), 0.86 (t, *J* = 6.2 Hz, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 152.82, 140.13, 137.83, 129.32, 126.86, 122.59, 114.38, 59.35, 41.57, 32.55, 21.06, 20.23, 14.26, 13.69.

1-ethyl-4-(4-nitrophenyl)-5-propyl-3,4-dihydropyrimidin-2(1*H*)-one

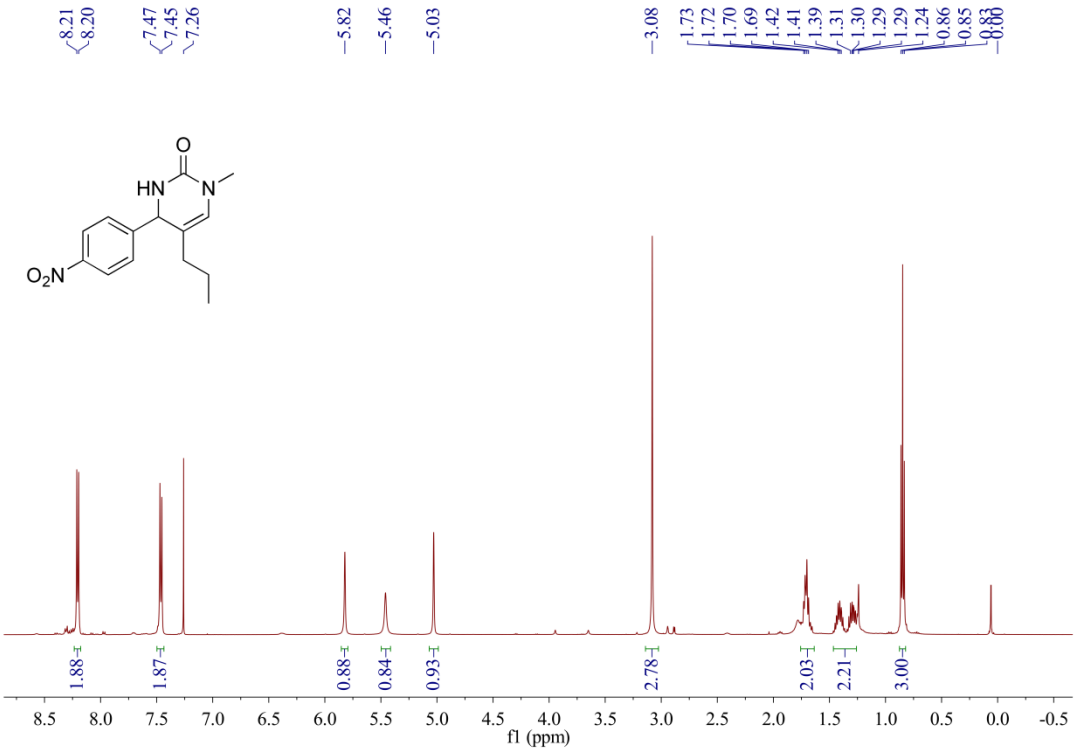


4o: 88%

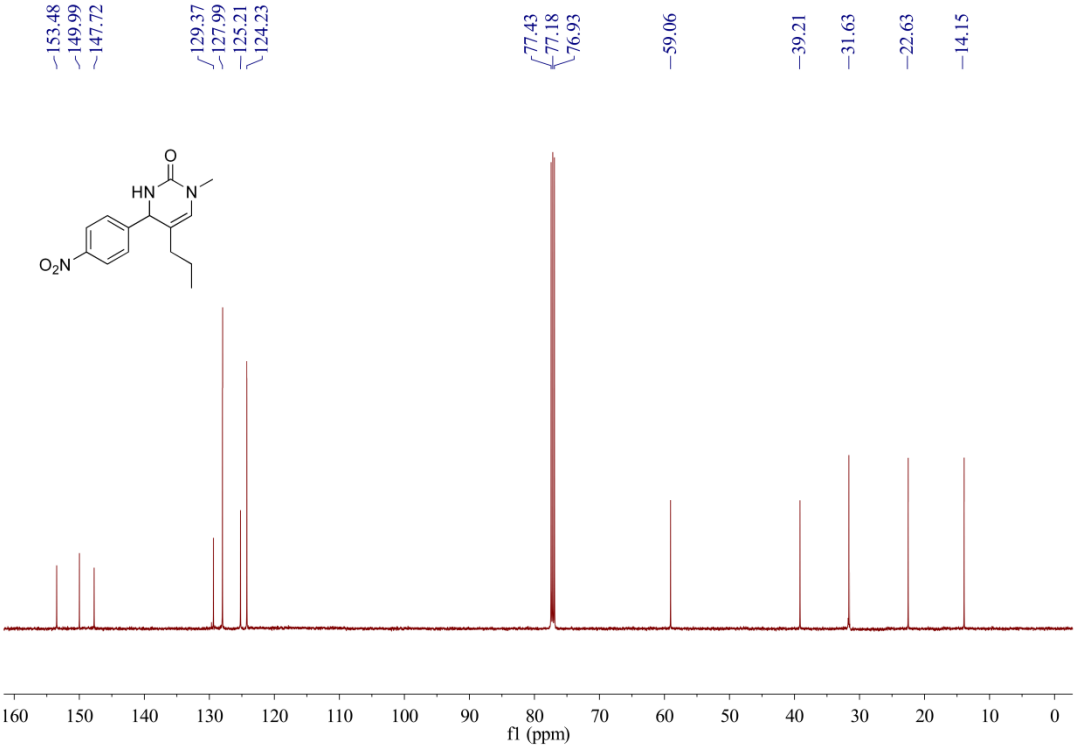
Yellow solid, ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.13 (d, *J* = 8.6 Hz, 2H), 7.38 (d, *J* = 8.6 Hz, 2H), 5.78 (s, 1H), 5.59 (s, 1H), 4.94 (s, 1H), 3.47 – 3.37 (m, 2H), 1.72 – 1.57 (m, 2H), 1.38 – 1.31 (m, 2H), 1.12 (t, *J* = 7.1 Hz, 3H), 0.79 (t, *J* = 7.3 Hz, 2H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 152.71, 150.04, 147.64, 127.86, 124.16, 123.80, 112.89, 58.87, 41.73, 32.45, 20.25, 14.23, 13.63.

^{1}H-NMR, ^{13}C-NMR spectra for DHPMs products

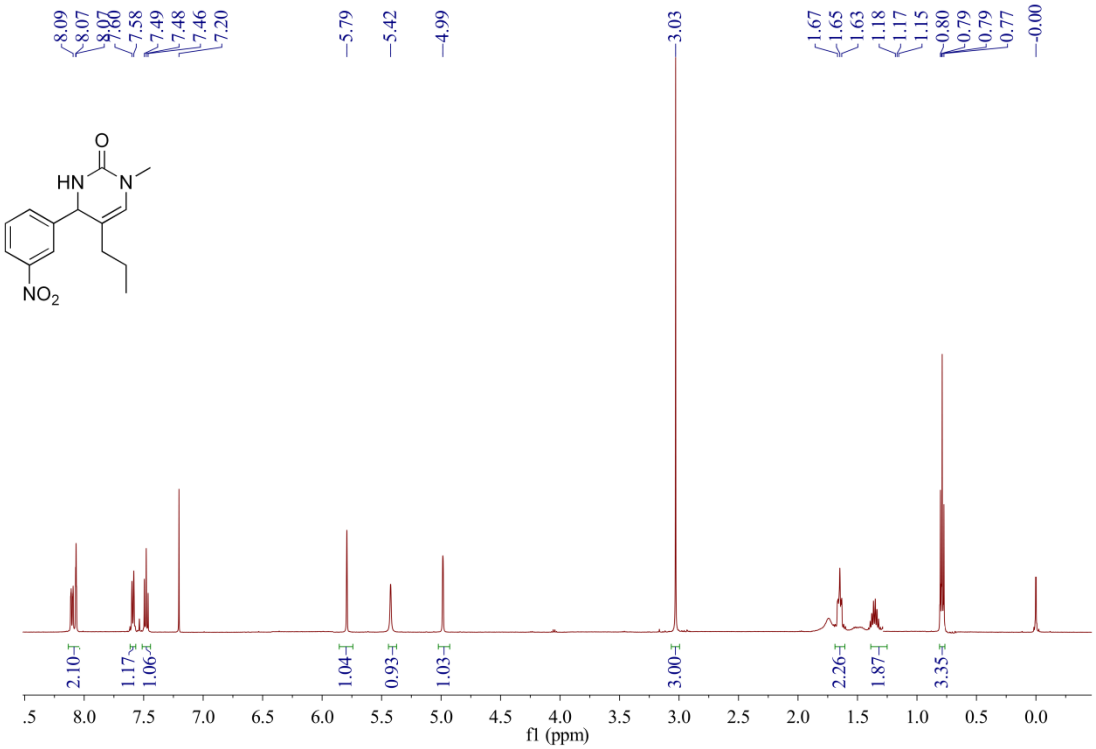
^{1}H NMR spectra of compound **4a**



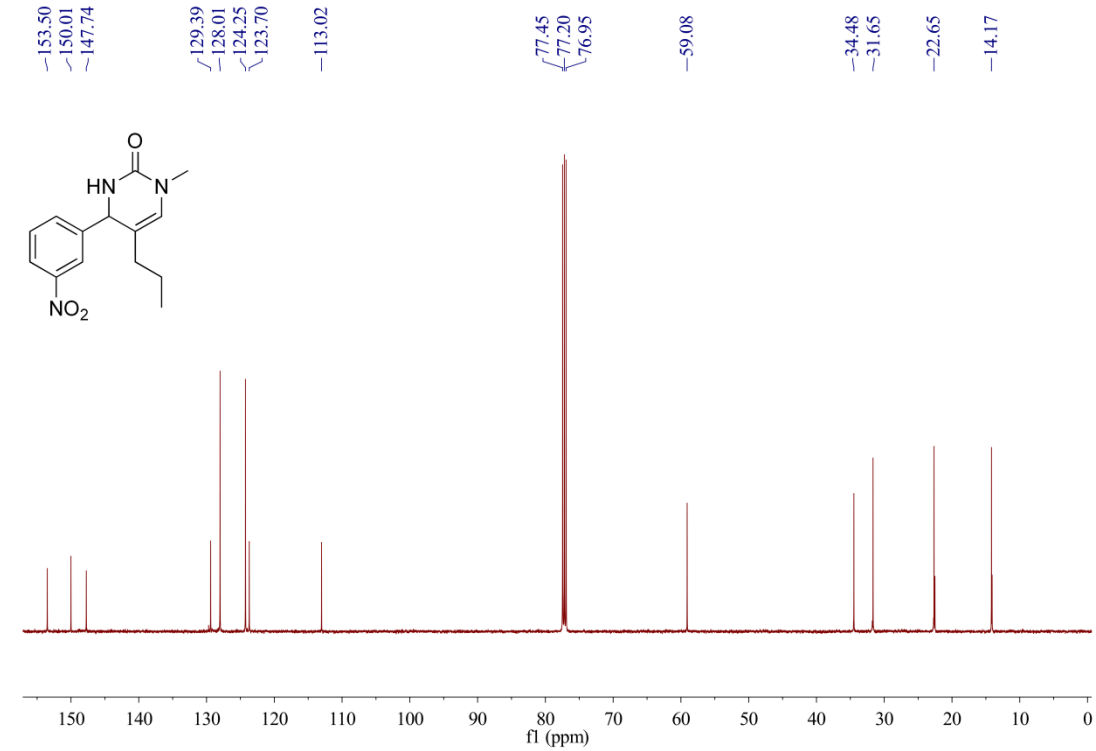
^{13}C NMR spectra of compound **4a**



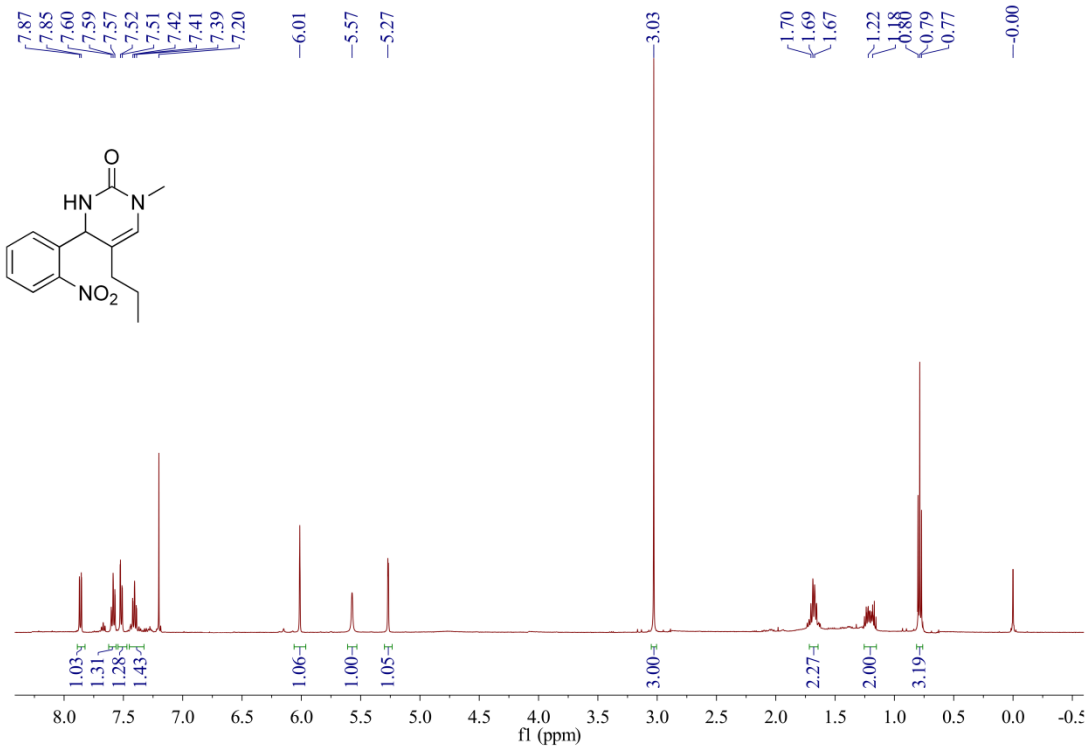
^{1}H NMR spectra of compound **4b**



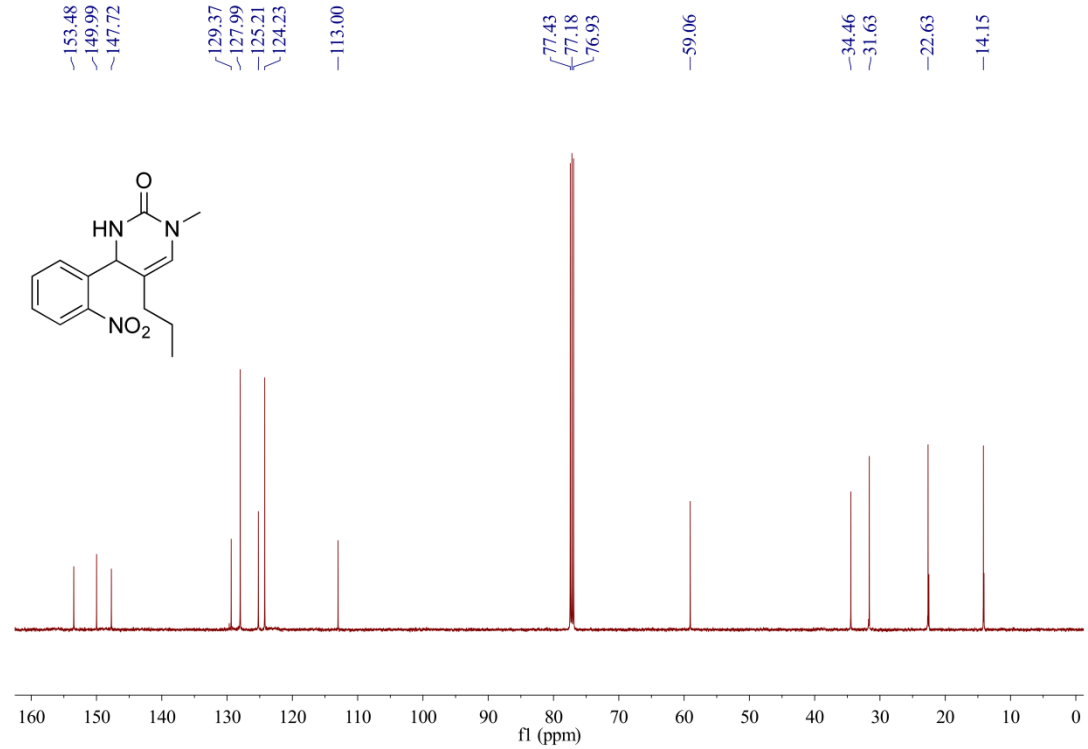
^{13}C NMR spectra of compound **4b**



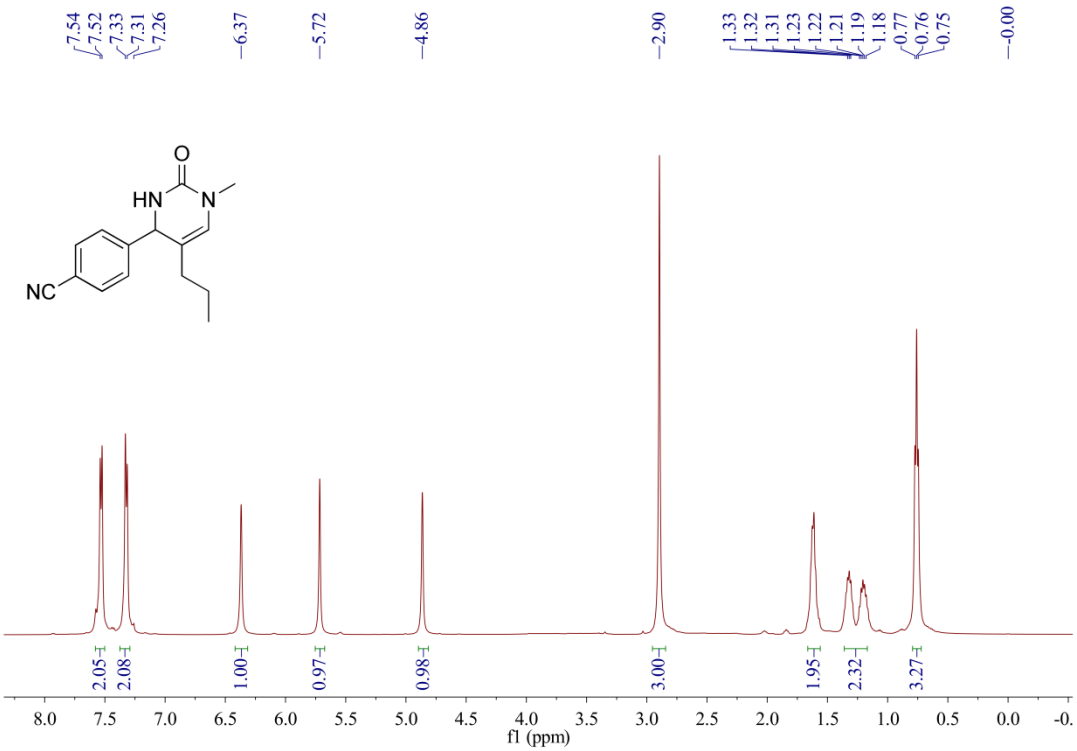
^{1}H NMR spectra of compound **4c**



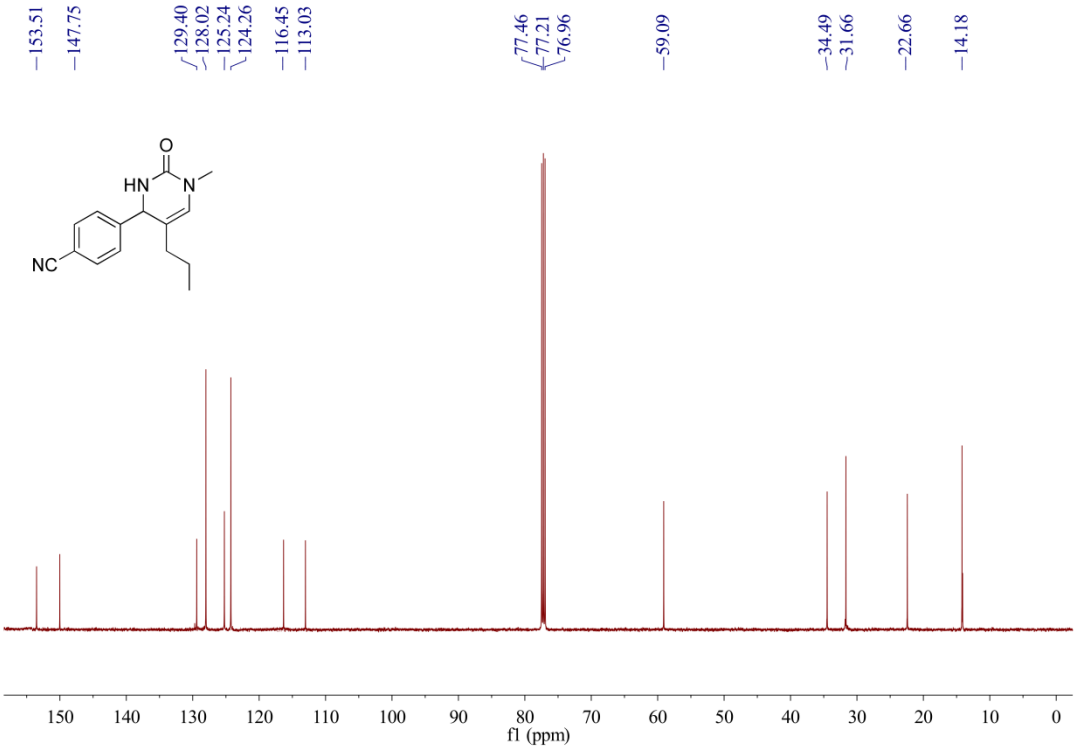
^{13}C NMR spectra of compound **4c**



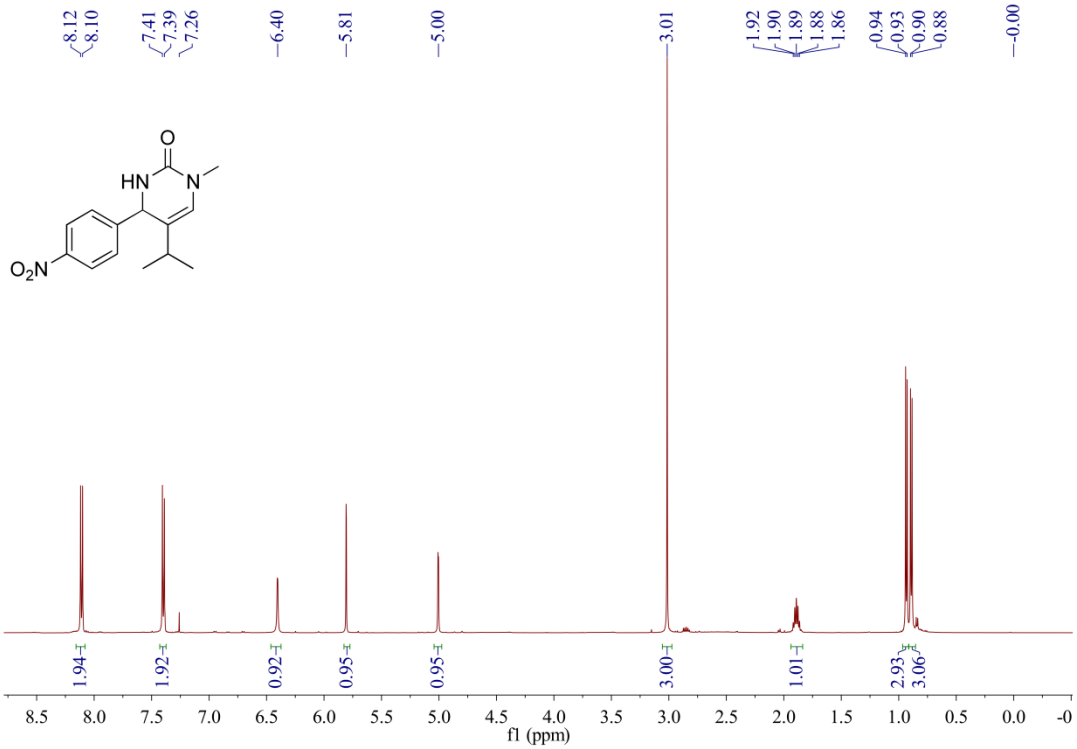
^{1}H NMR spectra of compound **4d**



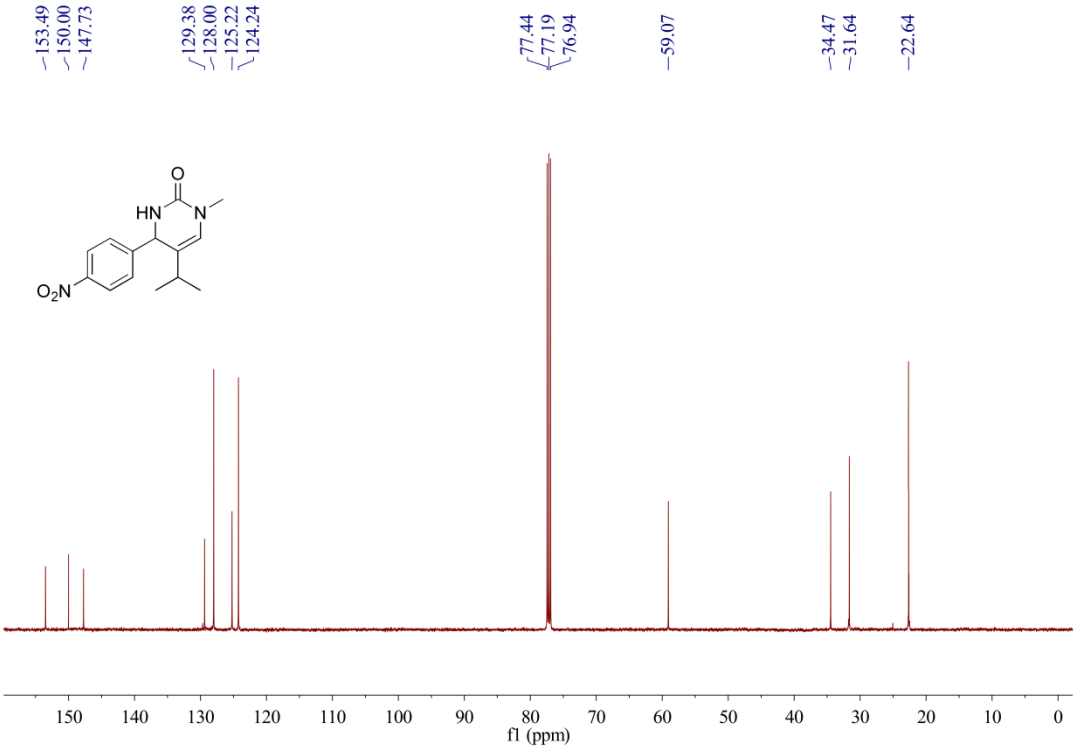
^{13}C NMR spectra of compound **4d**



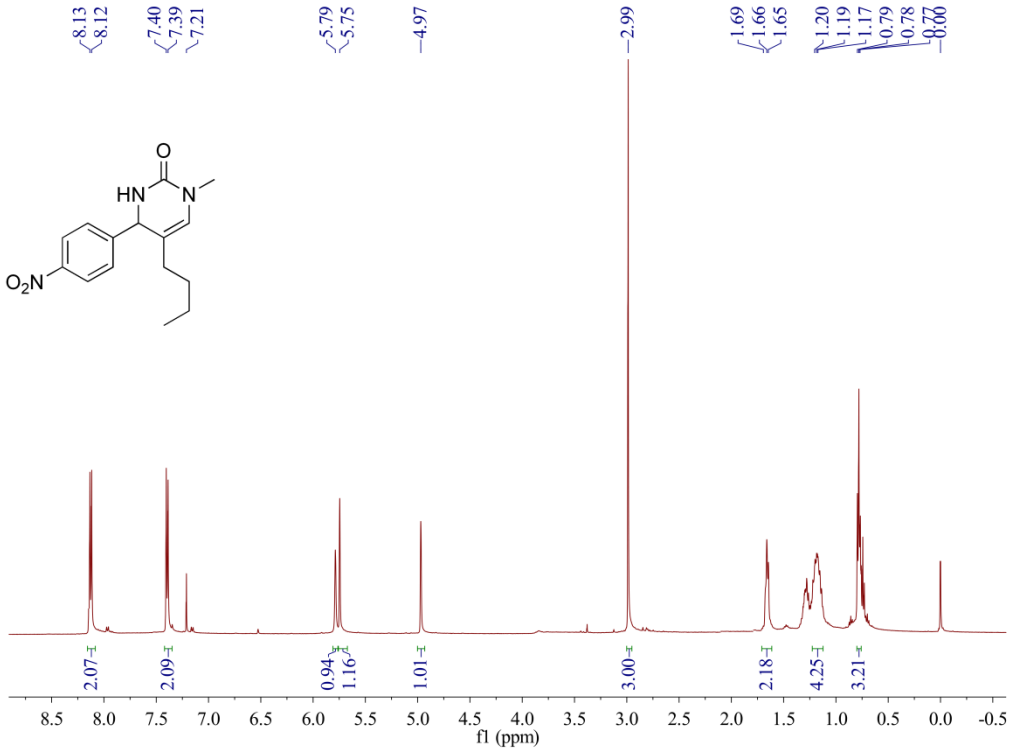
^{1}H NMR spectra of compound **4e**



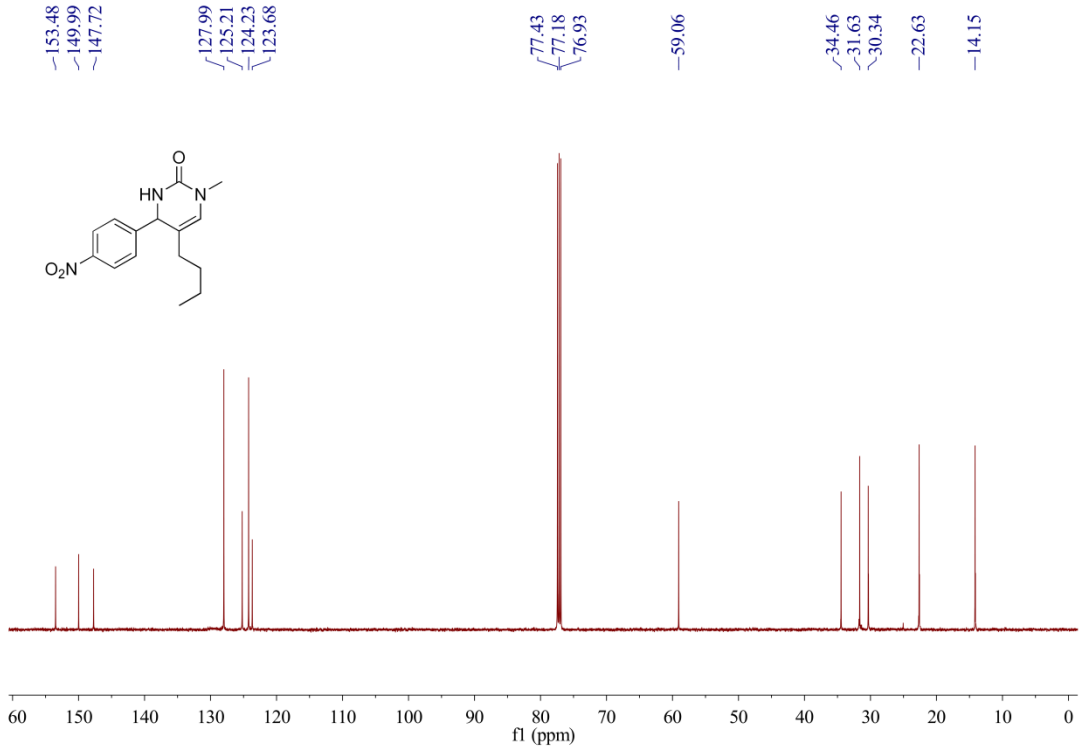
^{13}C NMR spectra of compound **4e**



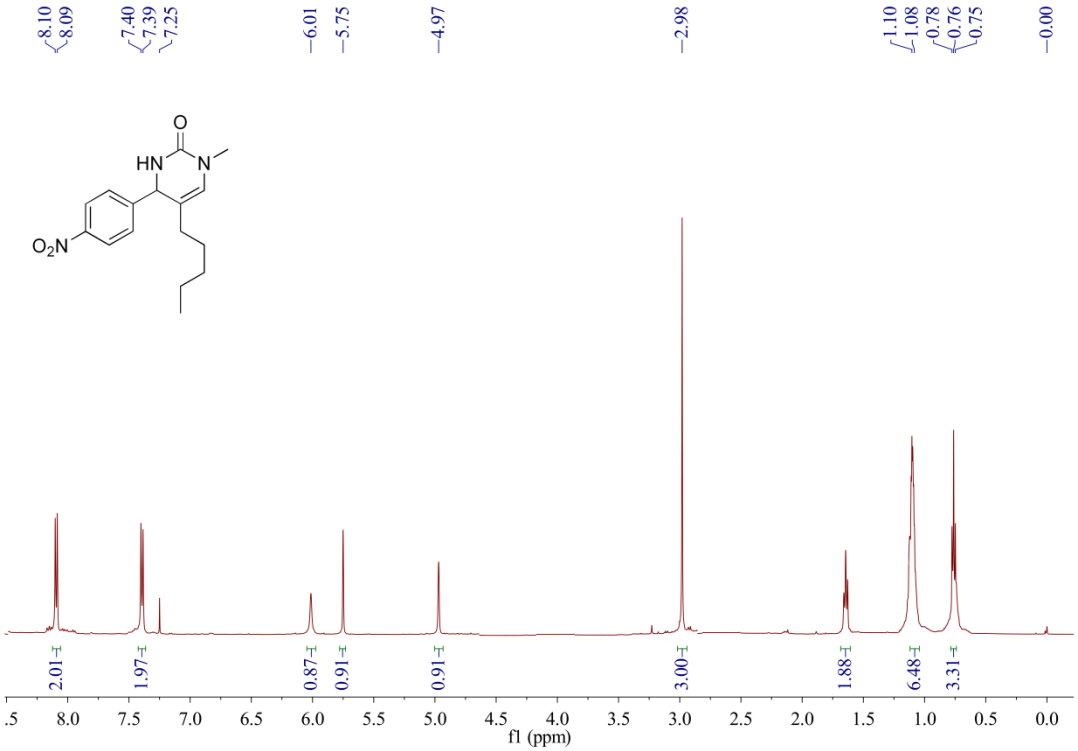
^{1}H NMR spectra of compound **4f**



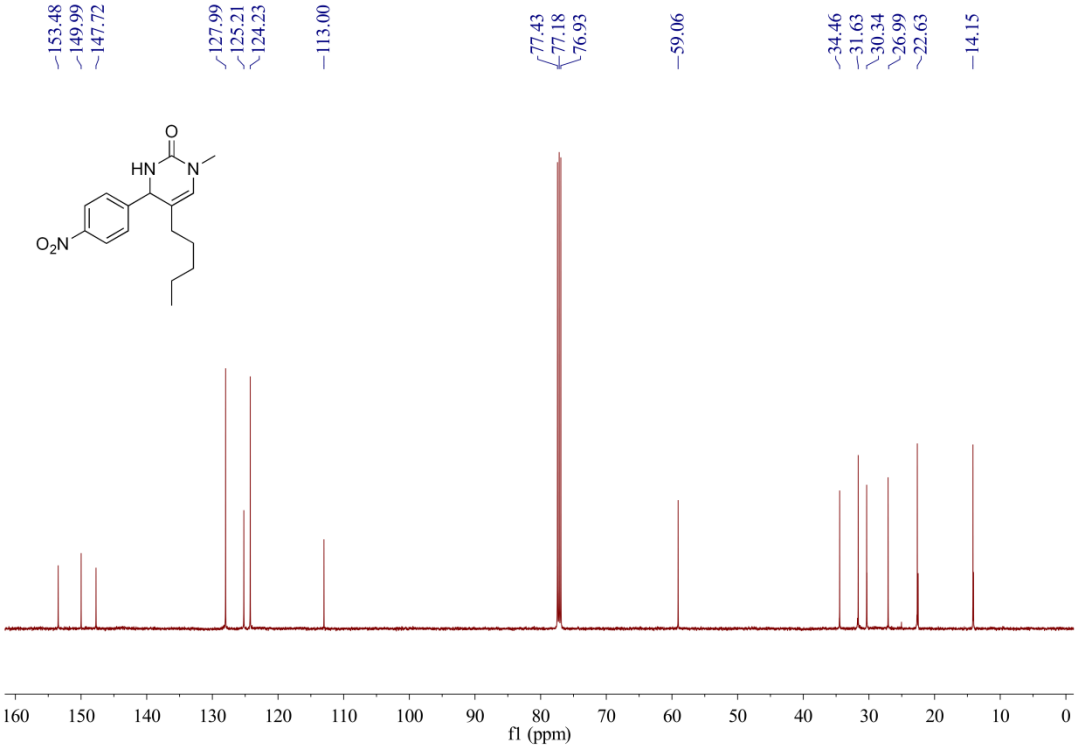
^{13}C NMR spectra of compound **4f**



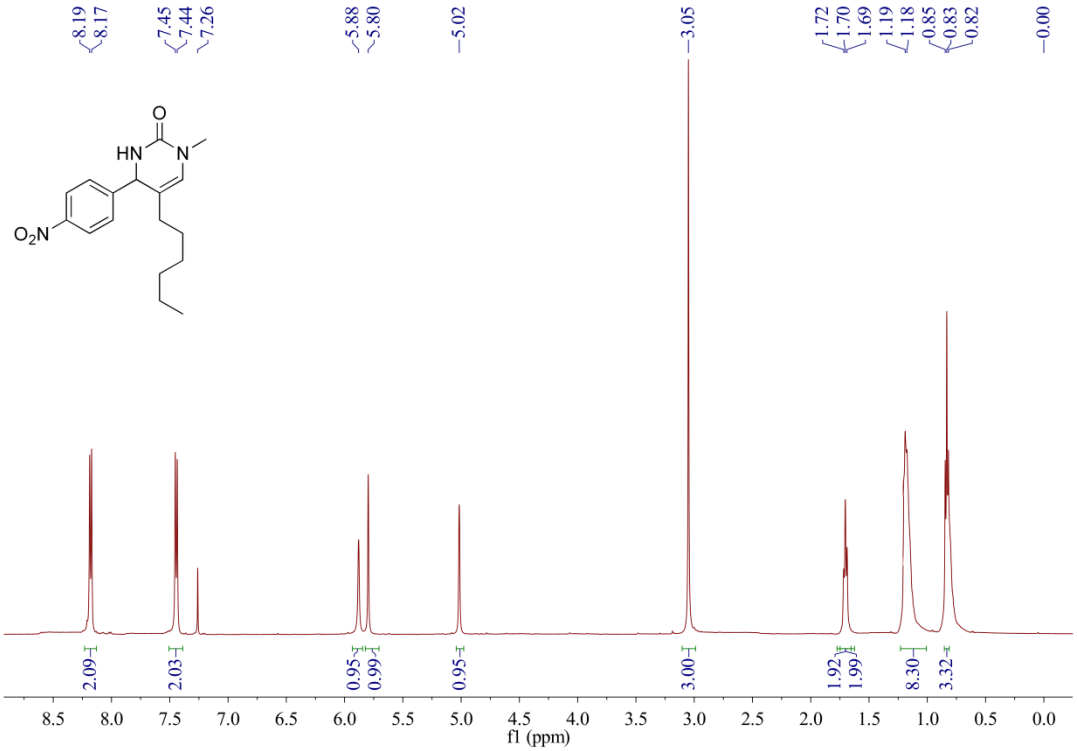
^{1}H NMR spectra of compound**4g**



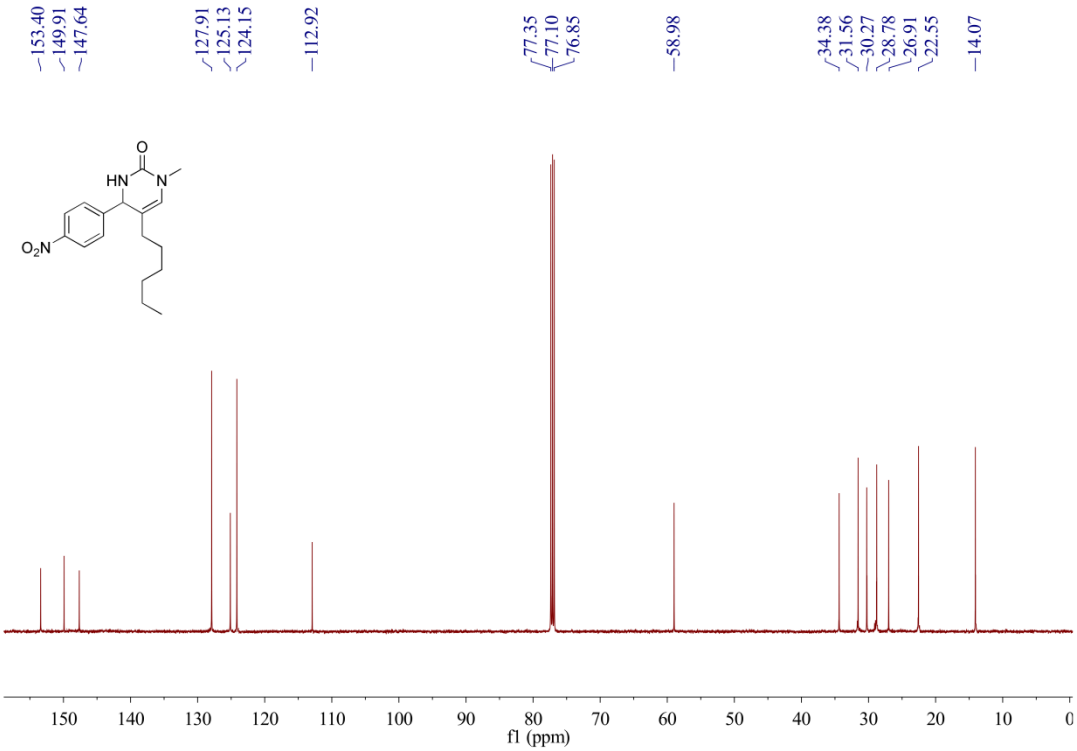
^{13}C NMR spectra of compound **4g**



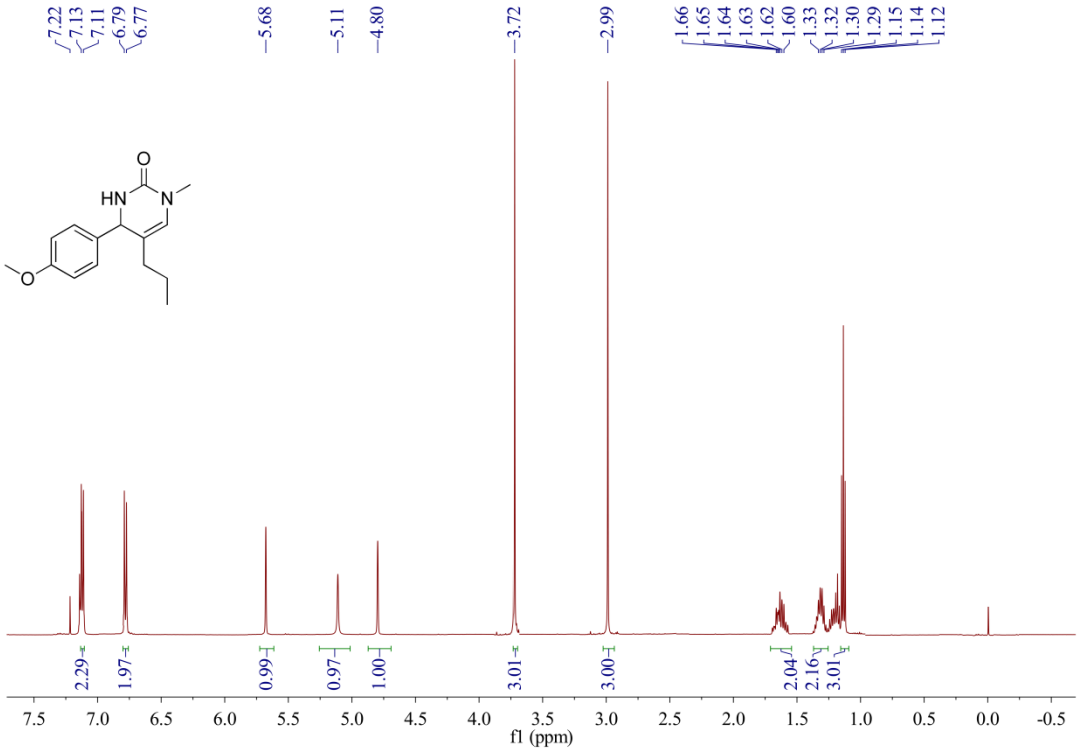
^{1}H NMR spectra of compound**4h**



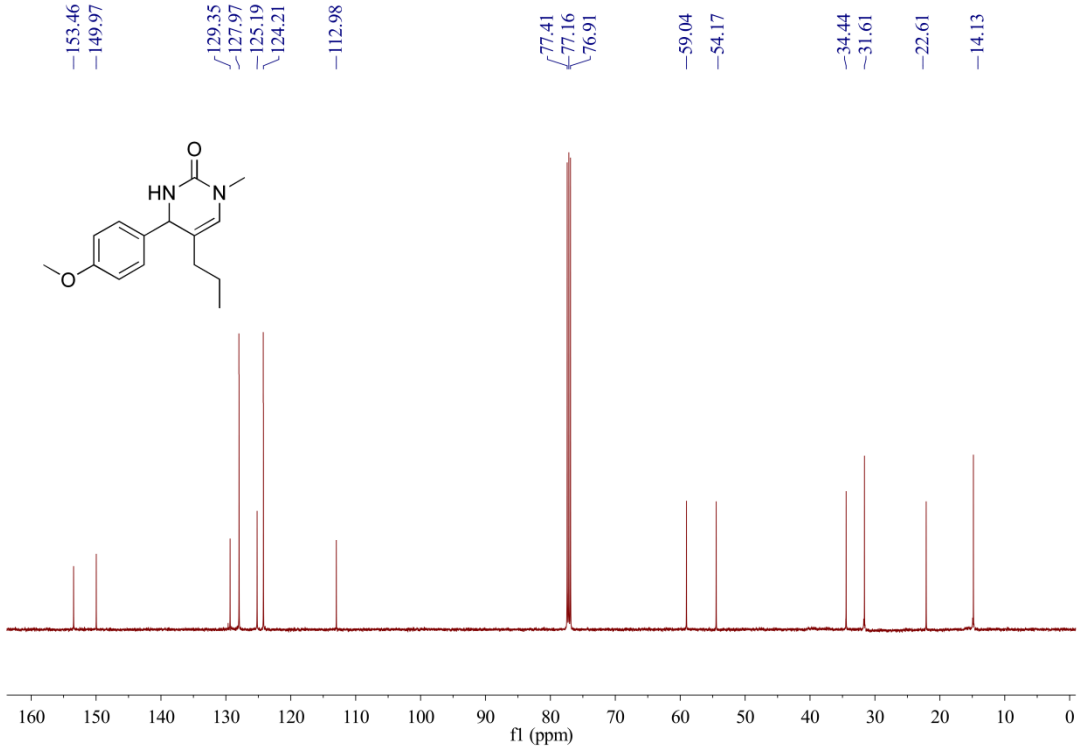
^{13}C NMR spectra of compound **4h**



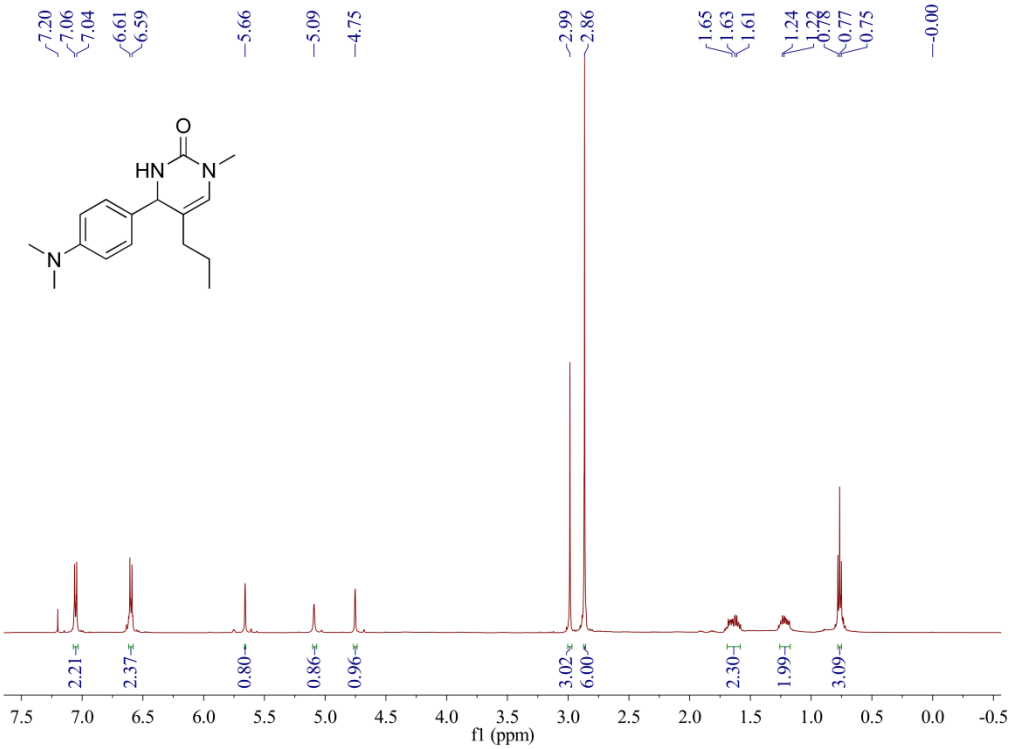
^{1}H NMR spectra of compound**4i**



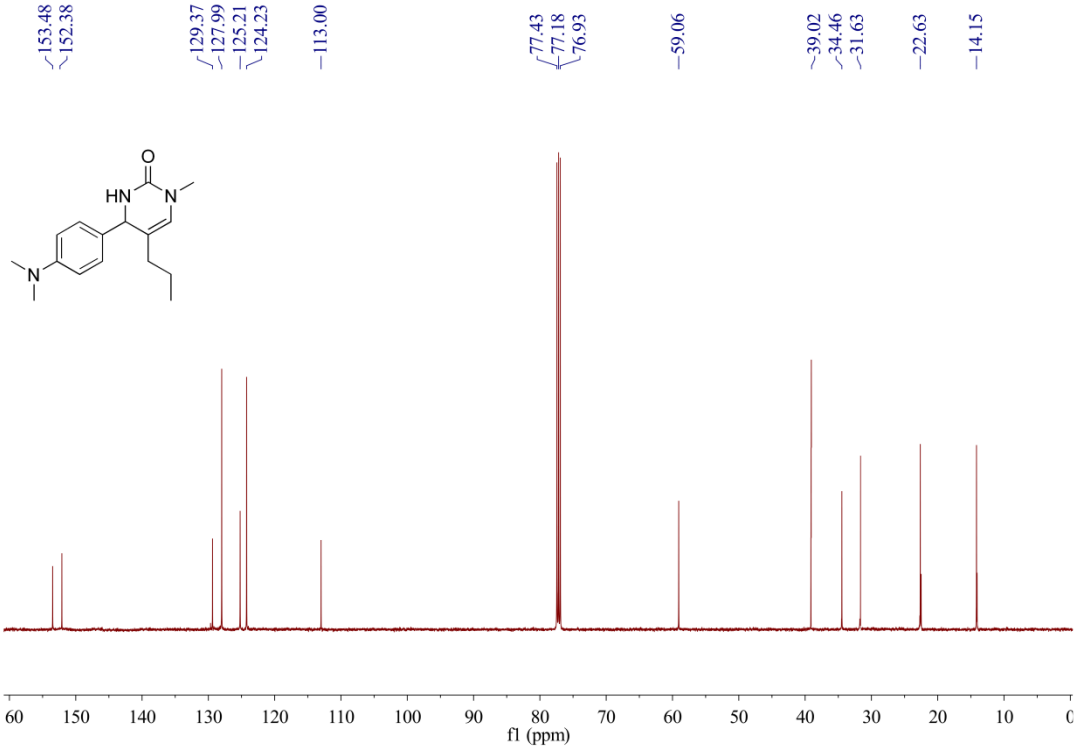
^{13}C NMR spectra of compound **4i**



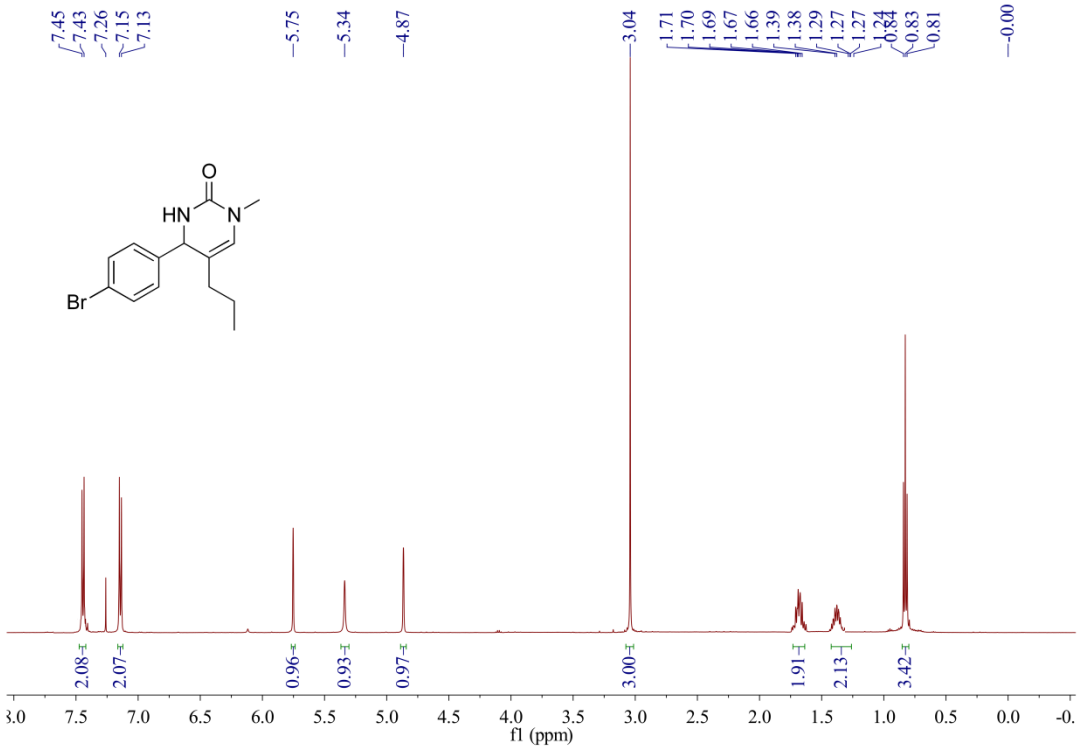
^{1}H NMR spectra of compound**4j**



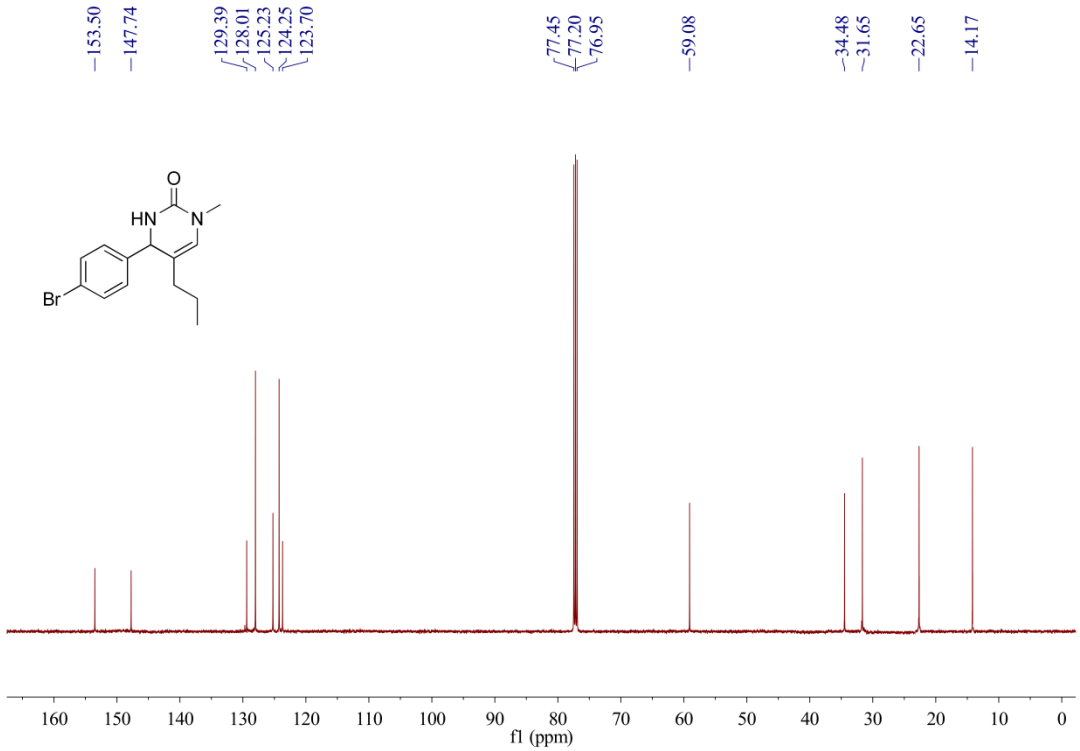
^{13}C NMR spectra of compound **4j**



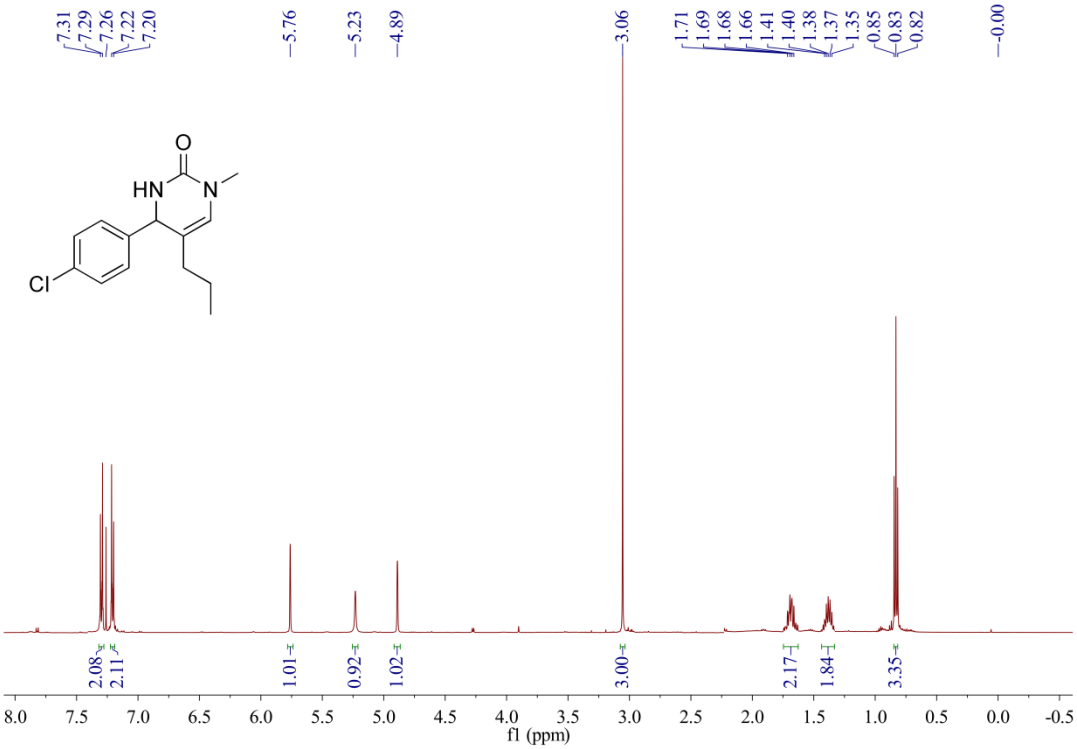
^{1}H NMR spectra of compound**4k**



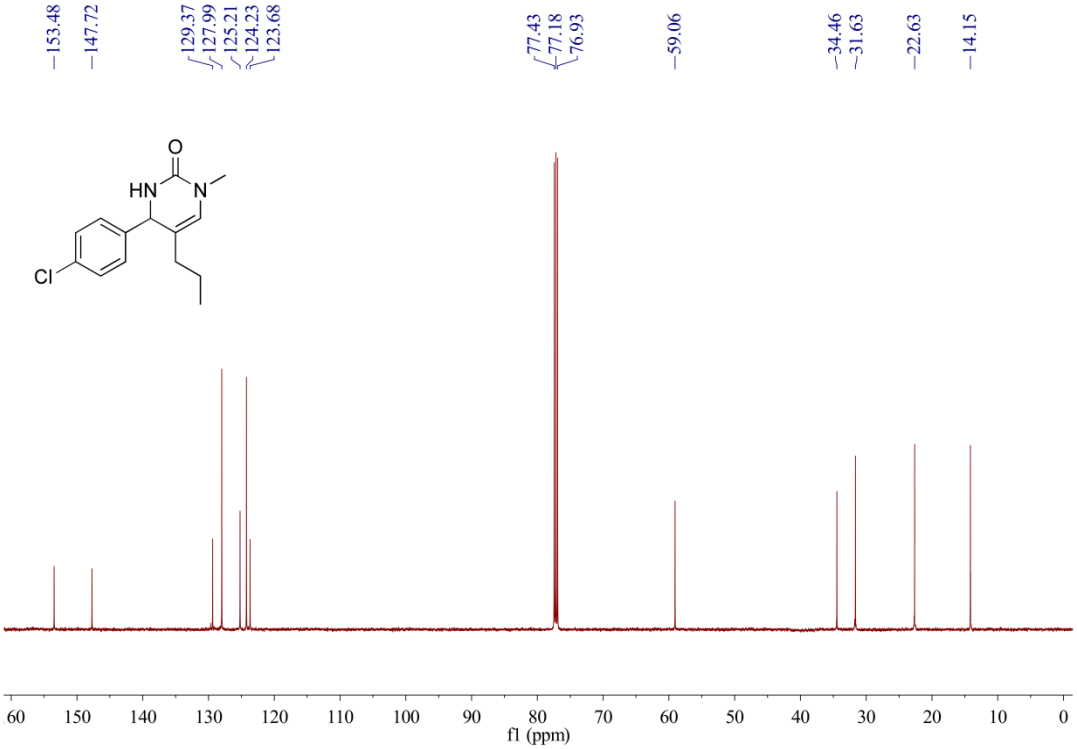
^{13}C NMR spectra of compound **4k**



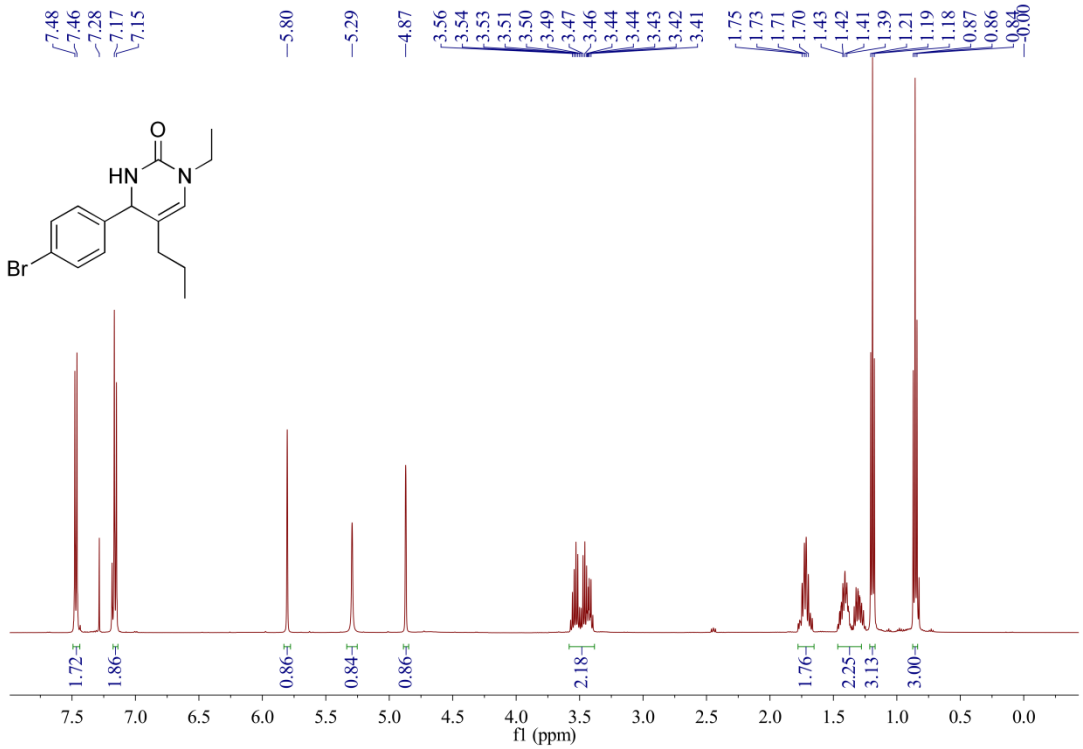
^{1}H NMR spectra of compound **4l**



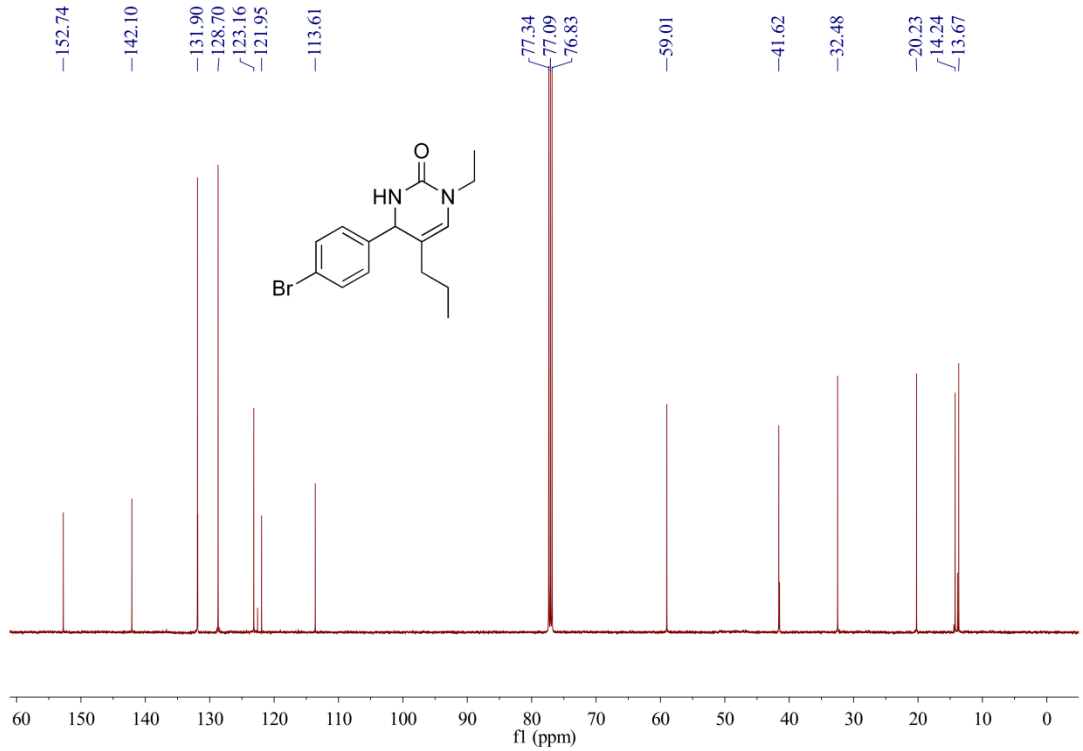
^{13}C NMR spectra of compound **4l**



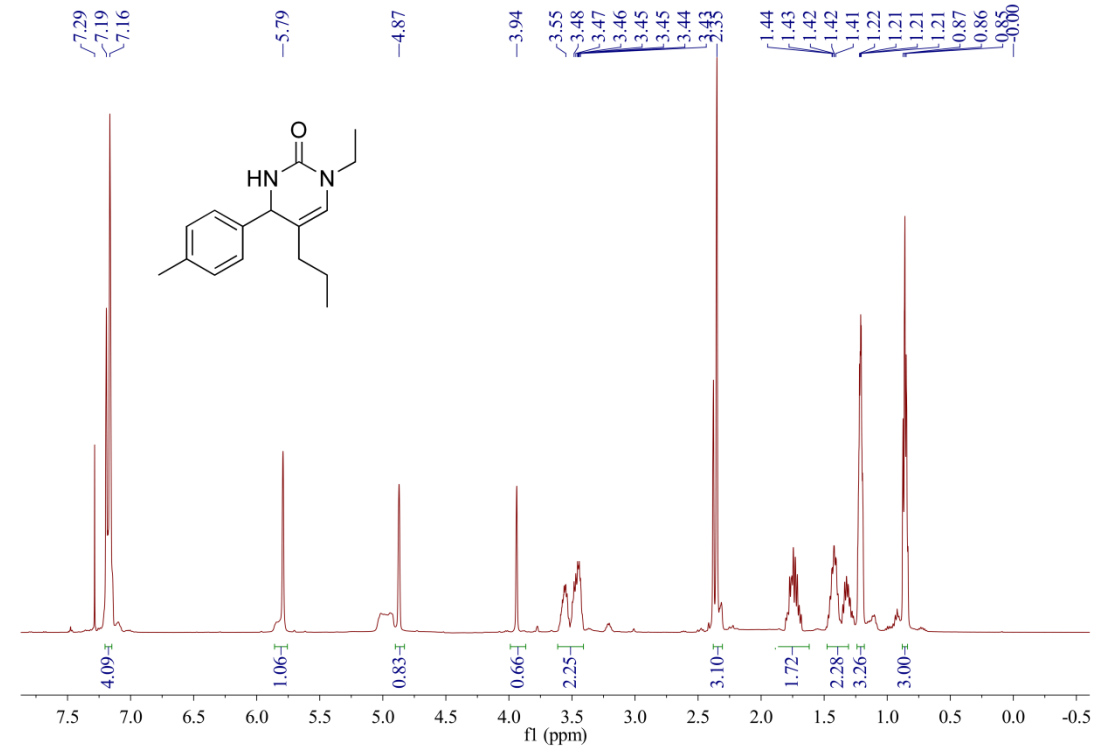
^{1}H NMR spectra of compound **4m**



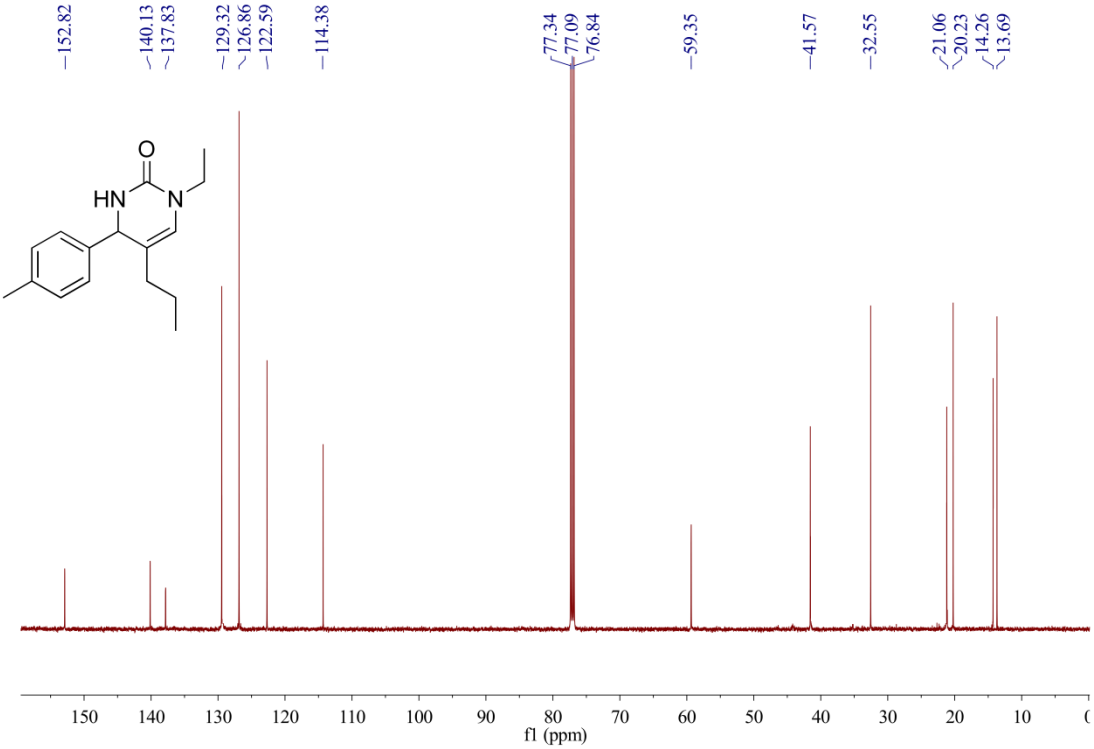
^{13}C NMR spectra of compound **4m**



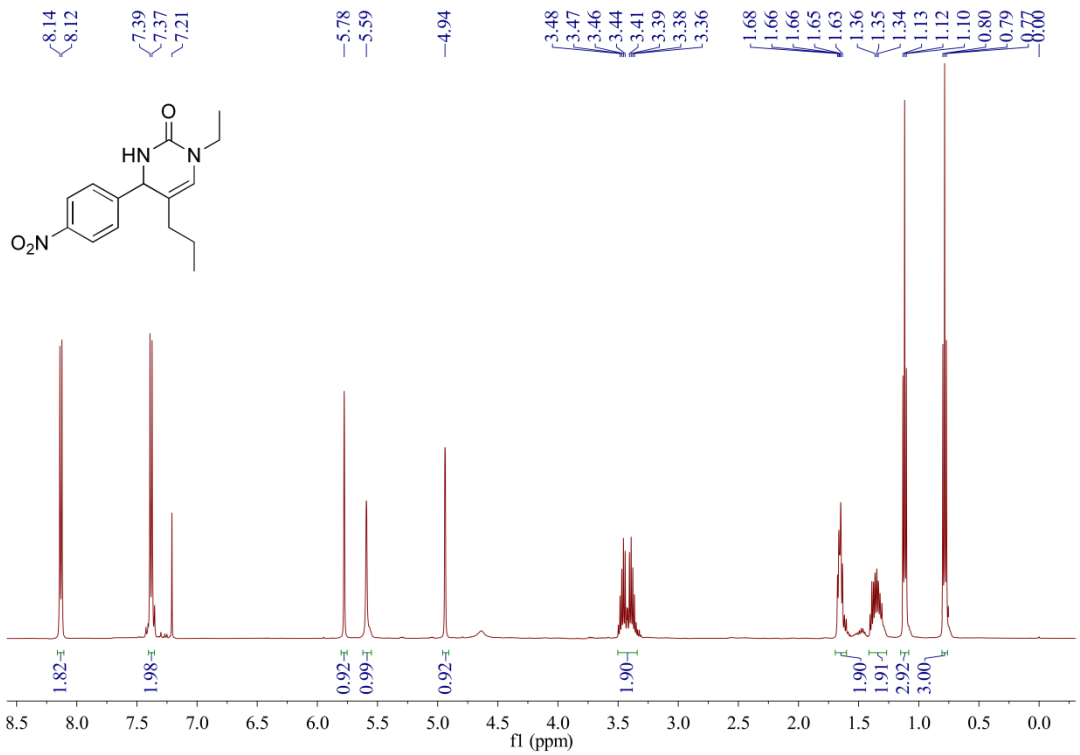
^{1}H NMR spectra of compound **4n**



^{13}C NMR spectra of compound **4n**



^{1}H NMR spectra of compound **4o**



^{13}C NMR spectra of compound **4o**